



Study of Condensable Ion Production by Resonant Laser Ionization

Jose Luis Henares Gonzalez

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THESE

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Spécialité Physique

Préparée au sein de l'Université de Caen Normandie

Study of Condensable Ion Production by Resonant Laser Ionization

**Présentée et soutenue par
Jose Luis HENARES GONZÁLEZ**

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ED SIMEM



*“Every act of perception is, to some degree, an act of creation
and every act of memory is, to some degree, an act of imagination.”*
Gerald M. Edelman (Physiology Nobel Prize)

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Preface

Context

On-line isotope separators are of great interest in the generation of high intensity pure short-lived radioactive ion beams (RIBs). For that reason, different ion source techniques are under development worldwide to provide pure RIBs. During the decade of 1980 a technique, called Resonant Ionization Laser Ion Source (RILIS), based on the step-wise resonant photo-excitation and ionization of atoms of interest, was developed for frontier research in several domains. The RILIS is a selective ion source which provides high ion beam intensities, a low ratio of contaminants and it gives access to exotic and rare nuclei at on-line nuclear facilities like ISOLDE at CERN (Geneva, Switzerland) [1] or ISAC at TRIUMF (Vancouver, Canada) [2].

Nowadays, the Resonant Ionization Laser Ion Source (RILIS) technology has emerged as an important tool for several applications including Radioactive Ion Beam generation (RIB) [3, 4, 5], atomic spectroscopy [6, 7, 8] or ultra-trace analysis [9, 10]. RILIS is extensively used worldwide and the number of accessible elements increases each year. The main advantages are: a reduced production of isobaric impurities, an inherent Z-selectivity, a relatively high efficiency and a pulsed structure of the ion beam. All of these permit to use the RILIS as a universal and reliable ion source. The RILIS technique requires detailed laser spectroscopy information on atomic spectra and excitation schemes of all elements of interest. This allowed the use of RILIS as a tool for spectroscopy development, at the same time this information provided the feedback needed to the technique breakthrough. Up to now more than 51 elements are available with the RILIS [11, 12]. Nevertheless the probability of non-selective surface ionization of unwanted isobar contaminants must be minimized, in the case of the RILIS using a hot-cavity. In conclusion, the recent development of the RILIS concerns ionization efficiency, emittance and, isotopic and isomeric selectivity.

General objective of the thesis

The objective of the present work is the study and implementation of a Resonant Ionization Laser Ion Source in an off-line test bench at GANIL (Caen, France). The test bench was first successfully adapted and optimized for the RILIS from an ECR (Electron Cyclotron Resonance) test bench (chapters 3 and 4). The optimization included simulations and experiences focused on the search of the optimum extraction and transport parameters. In addition, the RILIS test bench was used to develop a low energy beam profiler that will be installed for SPIRAL2. This device was extensively used to characterize the ion beam.

Tin and zinc ion beams were produced by a RILIS and an experimental research was performed for both elements to find alternative and possibly more efficient resonant atomic transition schemes to the already known ones. The study included spectroscopy measurements and bibliographical research (section 3.4).

The influence of the RILIS ion source geometry to the ion beam characteristics was not studied in full depth yet. In this work a study was performed with different ion source shapes, in order to improve the RILIS performance. Efficiency, emittance and the arrival time of the generated ions were measured and compared for different ion source settings (chapter 5). In addition, the electric field generated by the resistive heating can have impact on the ion extraction by a small potential drop and corresponding acceleration in the ion source tubes, thus it may modify the ionization efficiency for laser ionized atoms.

The RILIS technique is known for its selectivity and ability to produce pure ion beams. In the case of using a hot-cavity to perform the laser-atom interaction, the production of alkali contamination due to surface ionization at high temperatures is still an unsolved problem. For this reason, two contamination reduction methods were studied. One concerning the control of the electric field generated by resistive heating, and the second by the study of low work function materials in order to reduce the surface ionization of alkali contaminants (section 5.5).

Finally, a new technique to measure the energy distribution of the ion beam was developed, in order to understand the underlying physics concerning the ionization process inside the ion source. This technique may resolve some of the open and unexplained ion source behaviours. First measurements and interpretation are provided (section 5.4).

Préface

Contexte

Le principal intérêt des séparateurs d'isotopes en-ligne est la génération de faisceaux d'ions radioactifs de forte intensité et de courte durée de vie (RIB en acronyme anglais). Pour cette raison, différentes techniques de production d'ions sont en cours de développement dans le monde entier pour fournir des faisceaux d'ions radioactifs de grande pureté. Au cours de la décennie de 1980, une technique appelée source d'ions par ionisation laser résonante (RILIS en acronyme anglais) a été développée pour la recherche d'avant-garde dans plusieurs domaines. RILIS est basée sur la photo-excitation résonante par étapes successives des atomes d'intérêt. Il est une source d'ions sélective qui fournit des intensités élevées de faisceau avec un faible taux de contaminants, ce que permet d'accéder à de noyaux exotiques auprès d'installations nucléaires en-ligne comme ISOLDE au CERN (Genève, Suisse) [1] ou ISAC à TRIUMF (Vancouver, Canada) [2].

De nos jours, la technique de source d'ions par ionisation laser résonante (RILIS) a émergé comme un outil très important pour différentes applications: la production de faisceaux radioactifs [3, 4, 5], la spectroscopie atomique [6, 7, 8] ou l'analyse de ultra-traces [9, 10]. Cette technique est largement utilisée dans le monde entier et le nombre d'éléments accessibles augmente chaque année. Les principaux avantages sont: une réduction de la production d'impuretés isobariques, une sélectivité intrinsèque en Z , une efficacité élevée et une structure pulsée du faisceau d'ions. Tous ces éléments permettent de l'utiliser en tant que source d'ions universelle et fiable. En outre, la technique nécessite des informations détaillées sur les spectres atomiques et les schémas d'excitation des éléments d'intérêt. Cela a permis l'utilisation comme un outil pour le développement de la spectroscopie, dont les informations contribuent à améliorer la technique. Jusqu'à présent, plus de 51 éléments peuvent être produits par la méthode RILIS [11, 12]. Néanmoins, l'ionisation de surface non sélective des contaminants isobariques doit être réduite au minimum dans le cas d'une RILIS avec cavité chaude. En conclusion, le développement de RILIS concerne l'efficacité d'ionisation, l'emittance et la sélectivité, isotopique et isomérique.

Objectif général de la thèse

L'objectif de ce travail est l'étude et l'implantation d'une source d'ions par ionisation laser résonante (RILIS) sur un banc de test hors-ligne au GANIL (Caen, France). Le banc de test a été adapté et optimisé avec succès pour RILIS à partir d'un banc de test pour source d'ions ECR (Electron Cyclotron Resonance) (chapitres 3 et 4). L'optimisation a consisté en des simulations et des expériences pour trouver les paramètres optimaux d'extraction et transport. De plus, le banc de test RILIS a été utilisé pour développer un profileur de faible intensité qui sera installé dans SPIRAL2. Ce dispositif a été largement utilisé pour caractériser le faisceau d'ions.

Grâce à RILIS, des faisceaux d'ions d'étain et de zinc ont été produits et une recherche spectroscopique a été réalisée sur ces deux éléments pour trouver des schémas d'ionisation alternatifs à ceux déjà connues. L'étude comprenait des mesures de spectroscopie et des recherches bibliographiques (section 3.4).

L'influence de la géométrie de la source d'ions RILIS sur la génération des faisceaux d'ions n'avait pas encore été étudiée en profondeur. Dans ce travail, afin d'améliorer le comportement RILIS, une étude a été réalisée avec différentes géométries de la source d'ions. L'efficacité, l'emittance et temps de vol des ions produits ont été mesurés et comparés pour les différents paramètres de la source (chapitre 5). En outre, L'impact du champ électrique, généré par le chauffage résistif, sur l'extraction des ions dans les tubes de la source a été étudié. Ainsi, le champ électrique peut modifier l'efficacité d'ionisation pour les atomes ionisés par laser.

La technique RILIS est connue pour sa sélectivité et sa capacité à produire des faisceaux d'ions purs. Cependant, dans le cas d'une source d'ions utilisant une cavité chaude, la production de polluants due à l'ionisation de surface à haute température est encore un problème non résolu. C'est pourquoi, deux méthodes ont été étudiées pour réduire la génération de polluants. La première concerne le contrôle du champ électrique généré par chauffage résistif, et la seconde l'étude des matériaux avec un faible travail de sortie afin de réduire l'ionisation de surface des contaminants alcalins (section 5.5).

Finalement, une nouvelle technique pour mesurer la distribution en énergie du faisceau d'ions a été développée, afin de comprendre les processus physiques mis en jeu lors de l'ionisation à l'intérieur de la source d'ions. Cette technique peut aider à comprendre certains comportements encore inexplicables. Des mesures préliminaires et leur interprétation sont présentées (section 5.4).

Prólogo

Contexto

El principal interés de los separadores de isótopos en línea es la generación de haces de iones radioactivos de vida corta y alta intensidad (RIB en su acrónimo inglés). Por esa razón, diferentes técnicas de producción de iones están siendo desarrolladas en todo el mundo para proporcionar haces de iones radioactivos de alta pureza. Durante la década de 1980, una técnica llamada fuente de iones por ionización resonante láser (RILIS en su acrónimo inglés) fue desarrollada para la investigación de vanguardia. RILIS está basado en un proceso de foto-excitación resonante por pasos e ionización posterior de los átomos de interés. RILIS es una fuente de iones selectiva que proporciona haces de iones de alta intensidad con baja proporción de contaminantes, lo que permite el acceso a núcleos exóticos en instalaciones nucleares en línea como ISOLDE en el CERN (Ginebra, Suiza) [1] o ISAC en TRIUMF (Vancouver, Canadá) [2].

Hoy en día, la tecnología de la fuente de iones por ionización resonante láser se ha convertido en una herramienta fundamental en varias aplicaciones, tal como la generación de haces de iones radioactivos [3, 4, 5], la espectroscopía atómica [6, 7, 8] o el análisis de ultra-trazas [9, 10]. RILIS es utilizada en todo el mundo y el número de elementos accesibles aumenta cada año. Las principales ventajas son: la reducción de la producción de impurezas isobáricas, una inherente selectividad en Z , una eficacia relativamente alta y una estructura pulsada del haz de iones. Todas estas ventajas permiten utilizar RILIS como fuente de iones universal y fiable. La técnica RILIS requiere información detallada sobre los espectros atómicos y los esquemas de excitación de los elementos de interés. Por ello, RILIS sirve como una herramienta para el desarrollo de la espectroscopía, al mismo tiempo que esta información proporciona la retroalimentación necesaria para el avance de la técnica. Actualmente, Más de 51 elementos están disponibles mediante esta técnica [11, 12]. Sin embargo, la ionización térmica no selectiva de los contaminantes isobáricos debe minimizarse en el caso del uso de cavidades calientes. En conclusión, el desarrollo de RILIS afecta a la eficiencia de ionización, la emitancia y la selectividad, isotópica e isomérica.

Objetivo general de la tesis

El objetivo del presente trabajo es el estudio y la implementación de una fuente de iones por ionización resonante láser (RILIS) en un banco de pruebas en GANIL (Caen, Francia). El banco de pruebas ha sido adaptado y optimizado para RILIS a partir de un banco de pruebas ECR (Electron Cyclotron Resonance) (capítulos 3 y 4). La optimización incluye simulaciones y experimentos para encontrar los parámetros óptimos de extracción y transporte. Por otra parte, el banco de pruebas RILIS se ha utilizado para desarrollar un detector de perfil de baja intensidad que será instalado en SPIRAL2. Además este detector se ha utilizado para caracterizar el haz de iones.

Mediante RILIS, se han producido haces de iones de estaño y zinc y se ha realizado una amplia investigación experimental en ambos elementos para encontrar esquemas de ionización alternativos (y posiblemente más eficientes) a los ya conocidos. El estudio incluye medidas de espectroscopía y búsqueda bibliográfica (sección 3.4).

La influencia de la geometría de la fuente de iones en las características del haz de iones no ha sido aún estudiada en profundidad. Por ello, con el fin de mejorar el rendimiento de RILIS, en este trabajo se ha realizado un estudio con diferentes geometrías de las fuentes de iones. Se han analizado y comparado la eficiencia, la emitancia y el tiempo de vuelo de los iones generados para los diferentes parámetros de la fuente (capítulo 5). Por otro lado, se ha estudiado cómo el campo eléctrico, generado por el calentamiento resistivo, puede tener un impacto en la extracción de iones dentro de la cavidad de la fuente, lo que permite mejorar la eficacia de ionización de los átomos.

La técnica RILIS es conocida por su selectividad y su capacidad de producir haces de iones puros. Sin embargo, cuando se usa una cavidad caliente para realizar la interacción láser-átomo, la producción de contaminantes alcalinos, debido a la ionización térmica a altas temperaturas, es un problema todavía sin resolver. Por esta razón, y con el fin de reducir la ionización térmica de contaminantes alcalinos, se han estudiado dos métodos de reducción. El primero relacionado con el control del campo eléctrico generado por calentamiento resistivo, y el segundo empleando materiales que presentan un bajo valor en la función de trabajo (sección 5.5).

Por último, se ha desarrollado una nueva técnica para medir la distribución de energía del haz de iones con el fin de comprender la física subyacente en relación con el proceso de ionización dentro de la fuente de iones. Esta técnica puede ayudar a resolver ciertos comportamientos de la fuente de iones todavía no explicados. Se proporcionan las medidas preliminares y la interpretación de los resultados (sección 5.4).

Chapter 1

Radioactive-ion beams

1.1 Introduction

The study of the structure of matter and its properties dates back to the ancient history, when civilizations (like Greeks or Indians) tried to find out the smallest and most simple form of matter: the atom. But the definitive inception was not until 1886, when H. Becquerel, P. Curie and M. Curie, following the studies performed by Röntgen, discovered radioactivity. From that moment the experimental research on atoms started. In 1934, Fermi published the first article on the production of radioactive nuclei, leading to the discovery of nuclear fission and the production of elements lying beyond what was until then the Periodic Table.

Nuclear and atomic physicists try to understand the atomic properties and interactions (mass, radius, half-life, decay modes, reaction modes, cross sections, spin, magnetic and electric dipole moments, excited states, etc.). Today there are 118 elements in the periodic system (IUPAC [13, 14]) and more than 3100 nuclides were produced and partly studied experimentally (fig. 1.1). Radioactive nuclei exhibit an unique opportunity to study the structure of elements. A huge number of nuclear studies (e.g. nuclear structure, nuclear masses, astrophysics and nucleosynthesis processes, etc.) depend on the availability of radioactive-ion beams (RIBs), which are produced worldwide using particle accelerator and on-line mass separator facilities.

In the recent decades many new properties for nuclei far from stability were discovered. For example: New nuclear structures have been observed (e.g. neutron halo observed on ^{11}Li [16], super neutron-rich nuclei [17, 18]) and new elements are accessible (e.g. the discovery of a new element $Z=117$ [19]). ^{40}Mg and ^{42}Al were experimentally measured, which indicates that the neutron drip line is placed farther from stable isotopes than expected [20], and the two-protons radioactivity was finally observed experimentally at GANIL, GSI and MSU [21, 22, 23]. These experiments rely on the production of intense radioactive-ion beams.

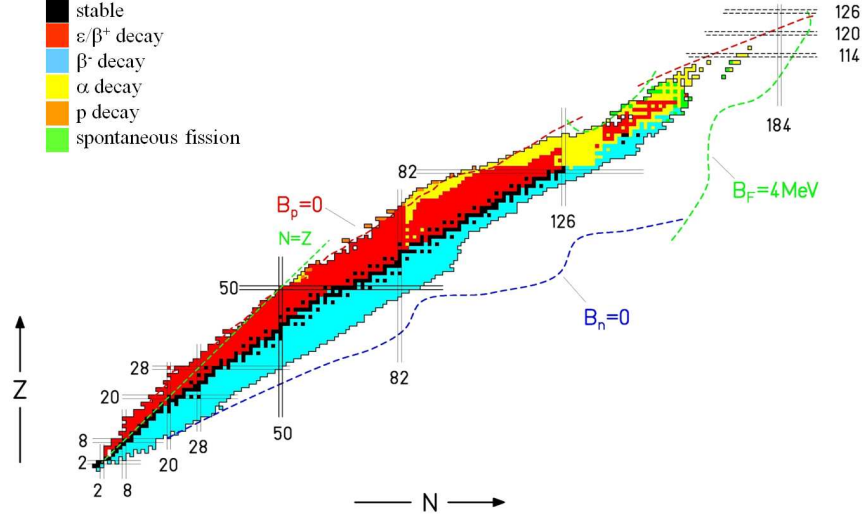


Figure 1.1– Representation of the atomic nuclei chart. The regions are presented as stable isotopes (black), or by its disintegration modes (coloured). In addition, the expected boundary limits are marked [15].

Nowadays, the forthcoming research objectives in particle accelerator and mass separator facilities need the production of “exotic” isotopes (i.e. short lived low-yield radioisotopes far-off β -stability) [24]. Moreover, one major prerequisite for extending those experiments to short lived low yield radioisotopes is the availability of pure isobaric ion beams [25]. For example, experiments performed at GANIL required highly pure ^{18}F beam for the study of the astrophysically important $^{18}\text{F}(p,\alpha)^{15}\text{O}$ reaction, relevant for γ -ray production in novae [26], or the selective production of polonium isotopes at RILIS-ISOLDE for the study of its ground state and isomeric properties [6, 27].

1.2 Production of radioactive-ion beams

The Radioactive-Ion Beam (RIB) production is possible using either particle accelerator facilities or nuclear reactors. A particle accelerator is a device that uses electro-magnetic fields to drive charged particles to high speeds, and to keep them in well-defined beams for further use in engineering or scientific research. The source generates the particle beam at the beginning of the facility. The production of exotic radioactive-ion beams is complex and entail the problem of low production cross sections and yields several orders of magnitude lower than for stable isotopes. In addition the detection of short-lived isotopes is critical due to its short life-times [28]. Finally the non-selective production and the consequent contamination of the radioactive-ion beam decreases the reliability of the experiments.

Two methods are mainly used for the production of radioactive-ion beams allowing the exploration of nuclear properties: the in-flight method (section 1.2.1) and the ISOL (Isotope Separator On-Line) technique (section 1.2.2).

1.2.1 In-Flight method

The in-flight method consist of the bombardment of a thin target with a high-intensity heavy ion beam. While the interaction occurs, either fission, fusion or fragmentation reactions will happen depending on the nature of the projectile-target couple and the energy involved in the reaction. The produced radioactive nuclei are highly ionized with an energy determined by the incoming projectile energy, the reaction Q value and the energy loss in the target. The generated radioactive-ion beams are selected with a fragment separator and can be used to initiate new nuclear reactions or to produce an ion beam (fig. 1.2).

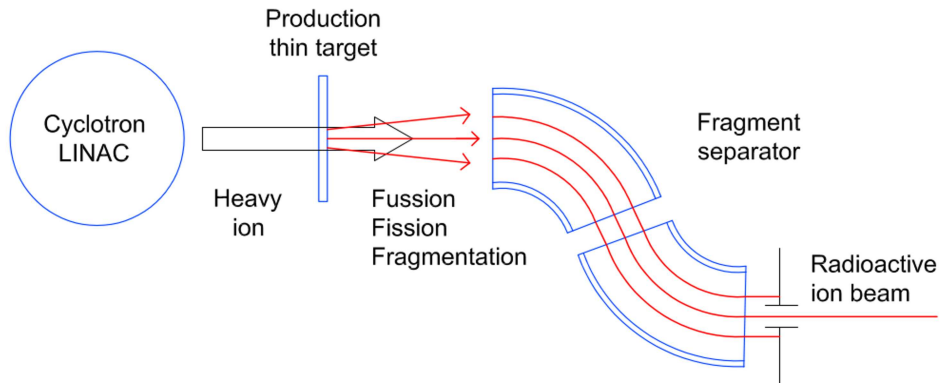


Figure 1.2– Scheme of radioactive-ion beam production by in-flight method (also called high energy fragmentation method). A heavy ion beam from a cyclotron or a Linear accelerator (LINAC) interacts with a thin target, producing radioactive-ion beams by nuclear reactions. The produced radioactive-ion beam is selected with a mass spectrometer.

The in-flight method can provide radioactive-ion beams with high energies and a large variety of isotopes. This method, due to the high velocity of fragments and prevention of any wall collision, gives access to very short-lived isotopes. However, the method presents several drawbacks: The process is non-selective, in some cases low production yields can be obtained, and the particle beam quality is bad, being deteriorated due to the kinematics of nuclear reactions. This production method is used in several laboratories such at S800 at MSU (USA) [29], LISE at GANIL (France) [30], GSI (Germany) [31] and RIBF at RIKEN (Japan) [32], and will be used at FRIB at MSU (USA) [33] and FAIR at GSI (Germany) [34].

1.2.2 ISOL method

Radioactive nuclei can be produced by the ISOL (Isotope Separator On-Line) method through different nuclear reactions (i.e. fission, spallation, fragmentation, etc.), when an incoming primary beam (e.g. 500 MeV proton beam delivered at ISAC-I at TRIUMF or 40 MeV deuteron beam of SPIRAL2 at GANIL) interacts with target atoms [35, 36]. Several consecutive steps are involved in the ISOL process (fig. 1.3).

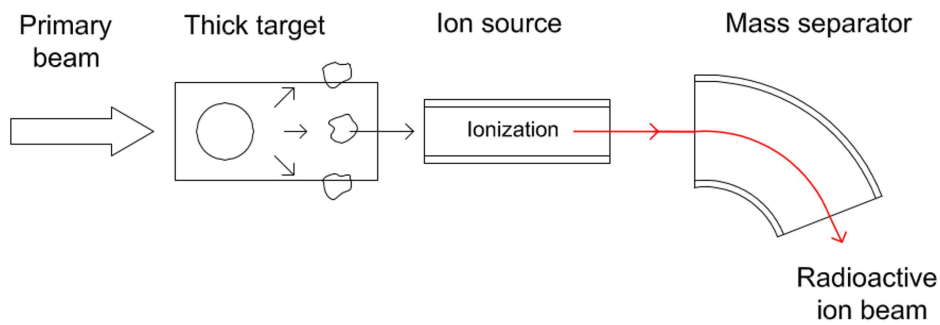


Figure 1.3– Scheme of radioactive-ion beam production by the ISOL method. The interaction of a primary beam with a thick target induces nuclear reactions and produces radioactive atoms, which are transferred to an ionization cavity where they are ionized. In the last step, the radioactive-ion beams are extracted and mass selected with a mass separator.

The production step implies the interaction of a primary stable ion projectile beam with a thick production target. The projectile beam has enough energy to induce nuclear reactions that produce radioactive atoms. The target is thick enough to stop the reaction products. According to the primary beam and the selected target, different reaction mechanisms can produce a wide range of radioactive isotopes. The produced radioactive atoms can diffuse out of the target due to the thermal energy. To enhance this process the target is heated to high temperature (>2000 °C). Afterwards, radioactive atoms effuse from the target cavity towards the ion source. The geometry of the transfer cavity has to be conceived in a way to facilitate the effusion to the ion source. The radioactive isotopes are ionized and extracted to form a radioactive-ion beam which is mass separated and can be directly used in low energy experiments or accelerated and sent to high energy experiments.

The ISOL method can provide very often high production yields (related to the thickness of the target) and it can produce a good optical-quality ion beam. However the diffusion of the elements from the target is a slow process and short-lived isotopes might not be released. In addition, the method cannot generate refractory elements or chemically active elements. The efficiency of the ISOL technique is related to the half-life of the extracted isotopes. The selectivity of the technique depends also on the chosen pair projectile-target.

The ISOL method is used at ISOLDE-CERN (Switzerland) [37], ISAC at TRIUMF (Canada) [38], SPIRAL1 at GANIL (France) [30], ALTO at IPN (France) [39], and it is also planned to be used in the future project SPES under construction at LNL (Italy) [40], RAON (Korea) [41], Beijing RIB facility [42], SPIRAL2 under consideration at GANIL (France) [43], and the European installation for radioactive ion production (EURISOL) in the far future [44].

1.3 Ionization methods

An ion source is a device that creates atomic or molecular ion beams. Conventional ion sources at on-line facilities are surface ionization sources, Forced Electron Beam Induced Arc Discharge (FEBIAD) ion sources, Electron Cyclotron Resonance (ECR) ion sources and Resonant Ionization Laser Ion Source (RILIS) [45]. The ion sources dedicated to the production of Radioactive-Ion Beams (RIB) have to be highly efficient, selective (to reduce the isobar contamination) and fast (to limit the decay losses of short-lived isotopes). In addition, in the case of radioactive beams produced by the ISOL method, the ion sources must operate steadily for extended periods of time (weeks to months) in a high radiation area and operating at high temperature ($>2000^{\circ}\text{C}$) to prevent the sticking of radioactive atoms or ions to cold surfaces. The choice of the most appropriate ion source is crucial to maximize the intensity, the beam quality and the number of different radioactive-ion beams that can be provided. A short introduction of the most common types of techniques for ion beam production will be given including the Resonant Ionization Laser Ion Source in this thesis.

1.3.1 Surface Ionization

The surface ionization (also named thermal ionization) is a physical process where the atoms are desorbed from a hot metallic surface due to temperature, and consequently ionized [46]. To maximize the efficiency, a high work function material (φ) is used in a way that atoms with low Ionization Potential (IP) may lose one electron when desorbing from the hot surface (fig. 1.4). The surface ionization was the first type of ionization method applied at ISOL facilities [47].

The ionization efficiency depends on the ionization potential, the surface material work function and also the plasma potential inside the hot cavity [48]. The surface ionization of positive ions is very efficient for elements with a small ionization potential like alkalis, which show high efficiencies. Other elements like rare-earth, heavier alkaline-earth and the boron group elements are surface ionized with relative low efficiencies. A dedicated surface ion source has been studied and developed for SPIRAL2 [49].

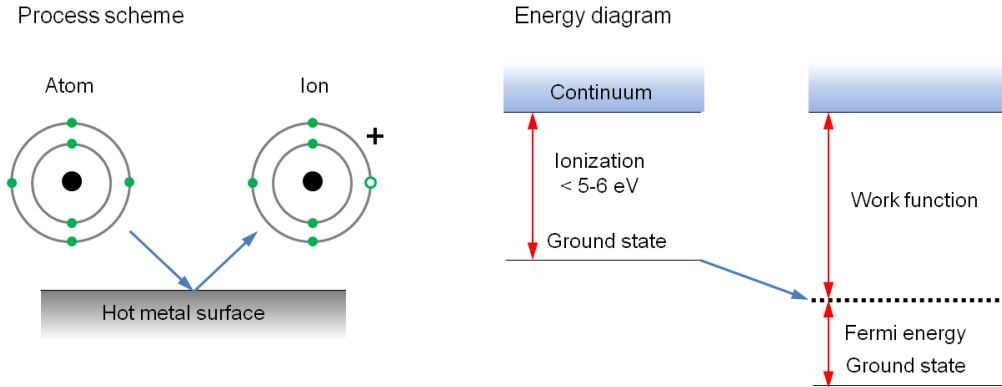


Figure 1.4– Positive surface ionization process. When an atom interacts with a hot metal surface, the energy of thermal electron emitted from the surface can be transferred to the atom resulting in ionization, if the energy exceed the element ionization potential.

Surface ionization is a competitive process and source of impurities and contamination in the Resonant Ionization Laser Ion Source (RILIS) (section 2.4).

1.3.2 Forced Electron Beam Induced Arc Discharge (FEBIAD)

The Forced Electron Beam Induced Arc Discharge (FEBIAD) is an ionization technique based on electron impact and atom-ion collisions [50, 51, 52] (fig. 1.5). In this technique an electrode grid (typically graphite) is used as anode for the continuous extraction of fast electrons from a heated cathode surface. The plasma chamber is surrounded by magnet coils for better confinement of the plasma.

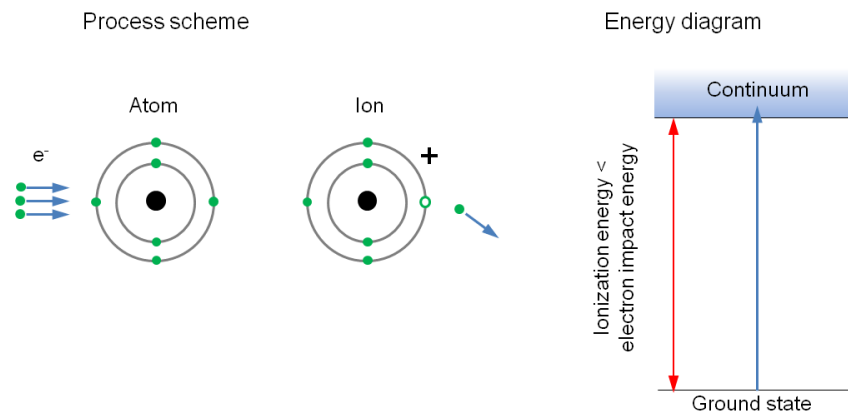


Figure 1.5– Ionization by electron impact (FEBIAD ionization). A fast electron beam interacts with an atom and the collisions provide enough energy to remove one electron.

One of the main disadvantages is the low selectivity of the process. Practically every atom entering the plasma volume can get ionized regardless its chemical nature. This ion source is used in the cases when other techniques are not able to produce required elements.

For the GANIL SPIRAL1 upgrade a FEBIAD ion source, developed by ISOLDE (VADIS VD5 [53, 54]), was coupled to the SPIRAL1 carbon target to produce new radioactive-ion beams. This source can reach 1900°C to decrease adsorption losses from the cavity wall. An upgrade of SPIRAL1 facility at GANIL is being studied with a new FEBIAD ion source in order to extend its production capabilities to condensable elements [55, 56].

1.3.3 Electron Cyclotron Resonance (ECR)

In the Electron Cyclotron Resonance (ECR) ion source, the ions are generated by means of ionization in a plasma generated by the process of electron cyclotron resonance. It does not have hot surfaces inside the plasma chamber, for that reason it is also called *cold* ion source.

The process starts with the injection of a high-frequency electromagnetic field (microwaves) into the cavity at the frequency corresponding to the electron cyclotron resonance, defined by the magnetic field applied to a region inside the volume (fig. 1.6). The magnets around the plasma chamber must provide a minimum magnetic field to generate a well for the electrons that will turn around the magnetic field lines and stay confined in the ion source where the electron cyclotron resonance condition is fulfilled:

$$\omega_c = \frac{q \cdot B}{m} \quad (1.1)$$

where ω_c is the angular frequency of the cyclotron motion, q is the elementary charge, B the magnetic field strength and m is the mass of the electron. The alternating electric field of the microwaves is set to be synchronous with the gyration period of the free electrons of the gas, and increases their perpendicular kinetic energy. Subsequently, when the energized free electrons collide with the gas in the volume, they can cause ionization if their kinetic energy is larger than the ionization energy of the atoms or molecules [57].

Depending on the frequency, ECR ion sources can produce single-charged or/and multi-charged states ions. ECR ion sources are especially well adapted to the ionization of noble gases and gaseous compounds, due to the good magnetic confinement and the high energy electrons that are usually provided by these sources.

In the SPIRAL1 facility the multi-charged ECR ion source (called *Nanogan III*) was connected to a thick carbon target allowing to produce multi-charged Radioactive-ion beams from 2001 [58]. *Nanogan III* was developed to produce fundamentally gaseous elements. In the per-

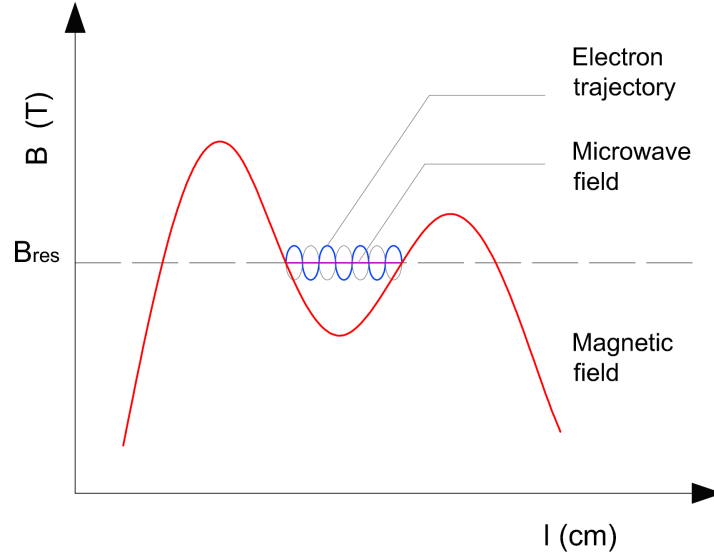


Figure 1.6– Diagram of the Electron Cyclotron Resonance (ECR) ionization technique. A plasma is produced due to the superposition of a magnetic field and a high-frequency electro-magnetic field (microwave) at the electron cyclotron resonance frequency (B_{res}).

spective of radioactive-ion beam production with SPIRAL2, the ECR Ion Source *MONOBOB II* (a fully radiation hard ECR ion source) was implemented [59]. It uses symmetric magnetic confinement created by four coils and it is optimized for the injection of a 2.45 GHz microwave.

1.3.4 Laser Resonance Ionization

Despite the fact that the ion beam generation methods can produce a wide range of isotopes with high performance, they often lack of selectivity due to the production of isobars and molecular side-bands. In many cases, the resolving power of mass separators is not high enough to resolve isobars and these contaminants are transmitted along with the isotope of interest [60]. In many cases, the production rate of the latter is several orders of magnitude lower than that of the isobaric contaminants [25].

The selectivity can be increased by the use of Resonant Ionization Laser Ion Source (RILIS), an elementally selective ionization process based on step-wise resonant photo-excitation (fig. 1.7). By this method the highest ion beam purity can be obtained compared to the other ionization techniques [61]. Due to this advantage, Resonant Ionization Laser Ion Sources are widely used at radioactive-ion beam facilities today. The RILIS process will be explained in detail in chapter 2.

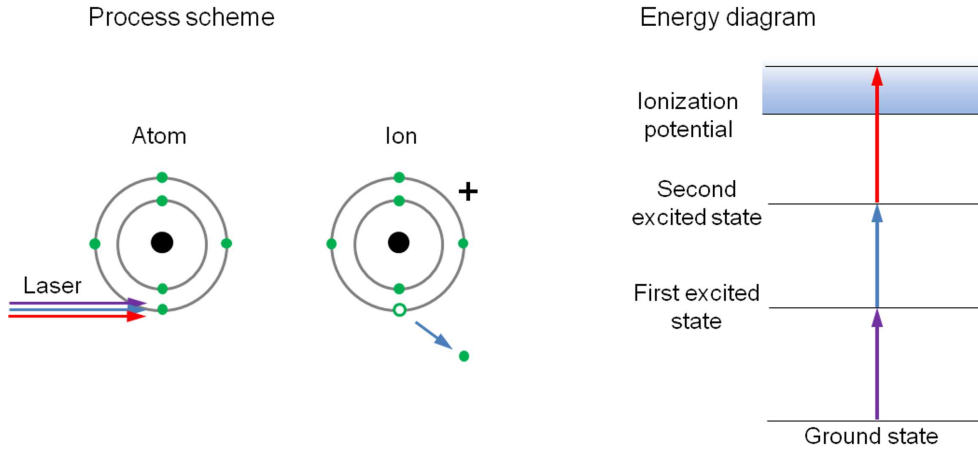


Figure 1.7– Diagram of the multi-step resonant photo-excitation process to selectively ionize a requested element. The resonance occurs when the laser frequency matches the atomic transition frequency, with the consequent excitation of the valence electron and ionization.

1.4 GANIL-SPIRAL2 radioactive ion beam facility

1.4.1 GANIL

GANIL (*Grand Accélérateur National d'Ions Lourds*) is an international facility located in Caen (France) for research with stable and radioactive-ion beams. Its experimental program ranges from nuclear and atomic physics to radiotherapy, and from condensed matter to astrophysics [30].

The GANIL particle accelerator complex is constituted by 5 isosynchronous cyclotrons and can produce a wide variety of accelerated ion beams, from carbon to uranium, allowing the generation of radioactive-ion beams by ISOL and in-flight methods. Its characteristics have been extended in 2001 by the SPIRAL1 facility (*Système de Production d'Ions Radioactifs Accélérés en Ligne*) in order to produce ISOL accelerated Radioactive-Ion Beams using projectile fragmentation [62, 63, 64].

1.4.2 SPIRAL2

In the near future GANIL will host the SPIRAL2 facility, for fundamental nuclear physics and interdisciplinary research, which is based on a high intensity linear particle accelerator (fig. 1.8) [65, 66, 67]. The SPIRAL2 project is divided into two phases:

1.4.2.1 SPIRAL2 Phase-1

The SPIRAL2 Phase-1 corresponds to the building of the superconductive LINAC accelerator and 3 experimental areas: NFS, S^3 and DESIR. The superconducting linear accelerator (LINAC) will produce intense beams of protons (33 MeV), deuterons (40 MeV), ^3He and heavy ions (up to 14.5 MeV/u). The first 5 mA proton beam were produced with the RFQ (Radio Frequency Quadrupole) in December 2015.

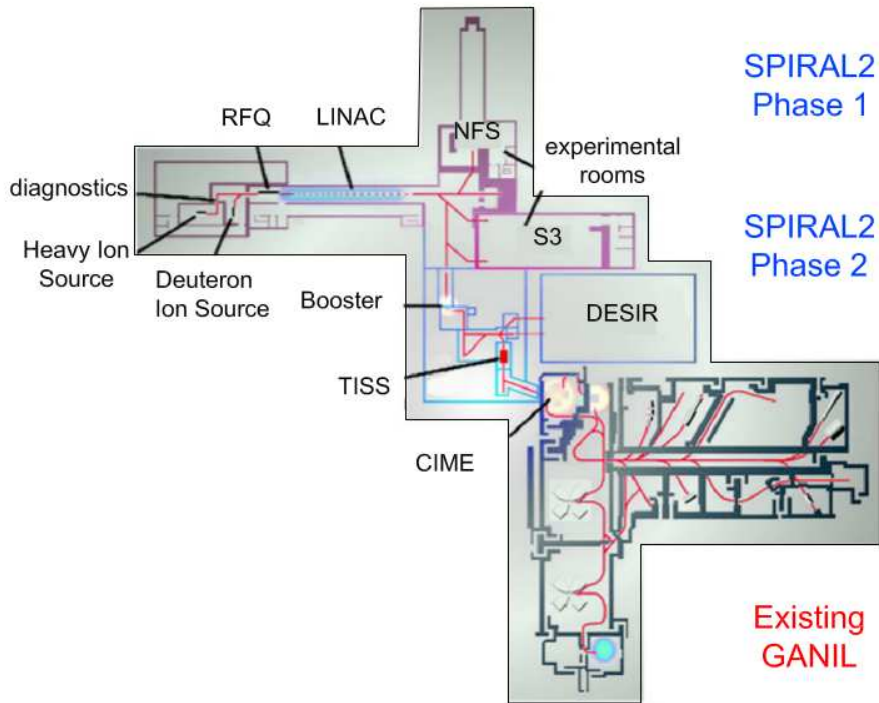


Figure 1.8– Floor plan of the existing accelerator facility at GANIL and the planned SPIRAL2 which consist of two independent phases: The LINAC main accelerator, NFS and S^3 (Phase-1) and the RIB production building (Phase-2).

The experimental areas are: NFS (Neutrons For Science) for the study and use neutron beams [68], the Super Separator Spectrometer (S^3) to produce and study radioactive-ion beams by fusion evaporation [69] and DESIR (*Désintégration, excitation et stockage d'ions radioactifs*) facility for the study of very low energy exotic nuclei [70]. A low energy branch will be connected to the S^3 spectrometer for the production and study of low energy beams of good optical quality, based on the in-gas laser ionization and spectroscopy technique [71].

1.4.2.2 SPIRAL2 Phase-2

The Phase-2 of SPIRAL2, still under discussion, is aimed to produce ion beams of short-lived radioactive nuclei by the ISOL method for research in fundamental physics, nuclear physics or studies of reactions in stellar processes (fig. 1.9).

The production of high intensity RIBs of neutron-rich nuclei will be based on the fission of uranium target induced by neutrons, which are obtained from a 200 kW deuteron beam impinging on graphite converter or by a direct irradiation with a deuteron, ^3He , or ^4He beam. After production, the radioactive-ion beam is injected in a charge booster to increase the charge state and is then injected into the existing *Cyclotron pour Ion de Moyenne Energie* (CIME), which is well adapted for separation and acceleration of ions in the energy range from about 3 to 10 MeV/u for masses $A \sim 100$ -150.

SPIRAL2 beams can be used in the present experimental area of GANIL. The produced radioactive atoms will be ionized by different ion source techniques, according to the nature of the element and the requested characteristics of the ion beam. The ion source system can be exchanged due to ion beam requirements [56].

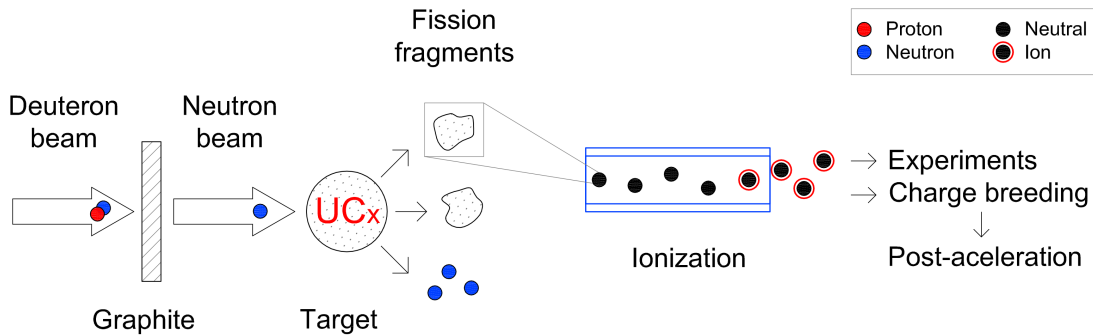


Figure 1.9– Scheme of radioactive-ion beam production by ISOL method for SPIRAL2-GANIL. A deuteron beam interacts with a graphite converter that produces a neutron beam. The consequent impact of the neutron beam in a UCx target induces fission and produces radioactive atoms, which are transferred to an ionization cavity where they are ionized. Then, the radioactive-ion beams are extracted, mass selected, accelerated or sent to a low energy radioactive-ion beam facility (DESIR).

The Target Ion Source System (TISS) of SPIRAL2 Phase-2 will eventually consists of a 2.7 kg UCx target heated at 2000 K by an ohmic furnace, and the ion source connected in the opposite side of the incoming primary beam (fig. 1.10). The future target and ion source system requires a 90 degree turn between the production area and the extraction area.

In the case of RILIS, this connection is performed via a double tube in elbow-shape, where the tube close to the target acts as transfer tube and the second tube is the ionization cavity. The radioactive atoms, after effusing out of the target, will be transferred to the ionization tube, where the laser beams and the radioactive atoms interact. Then, the radioactive-ion beams are extracted by a high voltage potential.

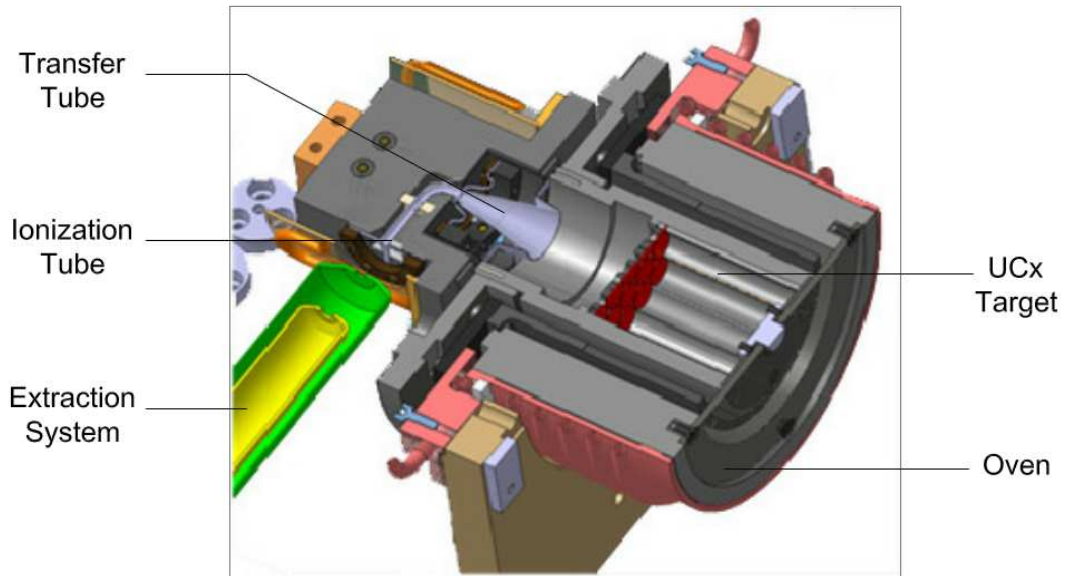


Figure 1.10– Target module front-end picture of the Uranium Carbide target furnace and ion source for SPIRAL2 Phase-2 in the RILIS setup. The ionizer/transfer tube assembly is marked.

The objective of this work is the study of a Resonant Ionization Laser Ion Source (RILIS), for a future implementation as part of the R&D project in the framework of SPIRAL2 Phase-2. The ionizer/transfer system behavior needed for the RILIS has been particularly studied. The developments presented here will be also useful for the laser ion source conceived for the S^3 low energy branch [71] and the future implementation of the RILIS in the SPIRAL1 facility.

Chapter 2

Resonant Ionization Laser Ion Source

2.1 Resonant photo-excitation process

The Resonant Ionization Laser Ion Source (RILIS) is based on the resonant photo-excitation process. The atoms are ionized via stepwise atomic resonant excitation, which is followed by ionization in the last transition. Optical excitation occurs when the laser radiation frequency matches the atomic transition frequency [72]. Since the atoms of each element have their own specific energy level structure, the ionization process has a higher selectivity compared to other non-specific ion source generation techniques.

By this method a variety of elements are accessible to be selectively ionized, if the excitation schemes are known. 80% of chemical elements have an ionization potential between 5 and 9 eV (fig. 2.1). The approximate 2 eV needed for each atomic transition fall in the visible spectrum range. Some elements are not accessible due to their high ionization potential (noble gases or some halogens) or because the first accessible excitation step requires a light wavelength below 200 nm (i.e. this radiation will not be transmitted in air due to UV absorption) or other transitions outside of the laser tuning range. Some elements have a low ionization potential and therefore can be easily ionized. This is the case of groups I and II in the periodic table (alkali metals and alkaline earth metals). In hot-cavity RILIS, they will be present as surface ionization contaminants, due to the resistive heating of the ionization cavity to high temperatures (typically 2000 K).

The resonant ionization process starts with the atom in its ground state or a low-lying thermally populated state. Valence electrons are stepwise excited by resonant absorption of laser light. They are advanced to a first excited state (FES) using a strong atomic transition. For most elements, suitable transitions for this excitation step are in the blue (energy of photons 2.5-3.5 eV) or ultraviolet (energy of photons >3.5 eV) wavelength regions.

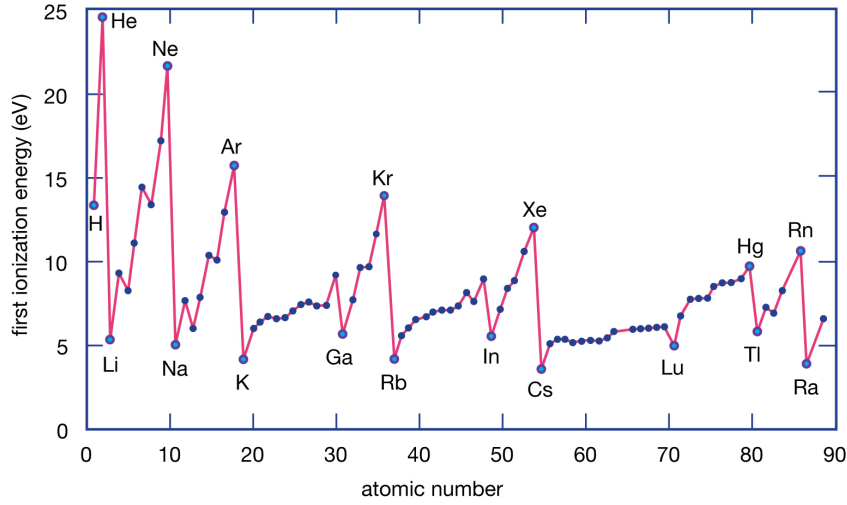


Figure 2.1– Ionization potential for several chemical elements. It is possible to observe that the average ionization potential does not exceed 9 eV [73].

The saturation of each transition level is desired for an efficient ion production. This situation is experimentally verified by measurements of saturation curves and adequate setting of injected laser power (section 3.4.2).

The first transition generally has a cross section in the order of 10^{-10} cm² and it is easily saturated with a microjoule laser pulse. However, for the remaining atomic transitions, higher power is needed for saturation due to lower cross sections. Following the first step excitation, the atom is driven to a higher excited state by another strong atomic transition connected to the first excited state. In some cases, this second excited state (SES) can already generate ionization, if located close to or even above the ionization potential. If not, a third excitation step will then ionize the atom. With the combination of two or three laser beams it is thus possible to ionize the predominant part of the elements of the periodic table.

The final atomic transition is crucial for the efficiency of the method. Fortunately there are different ways to realize this final step (fig. 2.2). The ionizing step could be reached via non-resonant transition into the continuum, above the first ionization potential of the element, or resonantly via an auto-ionizing state (i.e. an eigenstate located above the first ionization potential) or Rydberg states (a highly excited level with high principal quantum number n , located just below the ionization potential). The disadvantage of the non-resonant process is its relatively low cross section ($\sim 10^{-17}$ cm²), which is several orders of magnitude smaller than for transitions using auto-ionizing states or Rydberg states. Both, ionization via Rydberg or auto-ionizing states offers higher cross section because they are resonant processes ($\sim 10^{-12}$ - 10^{-13} cm²).

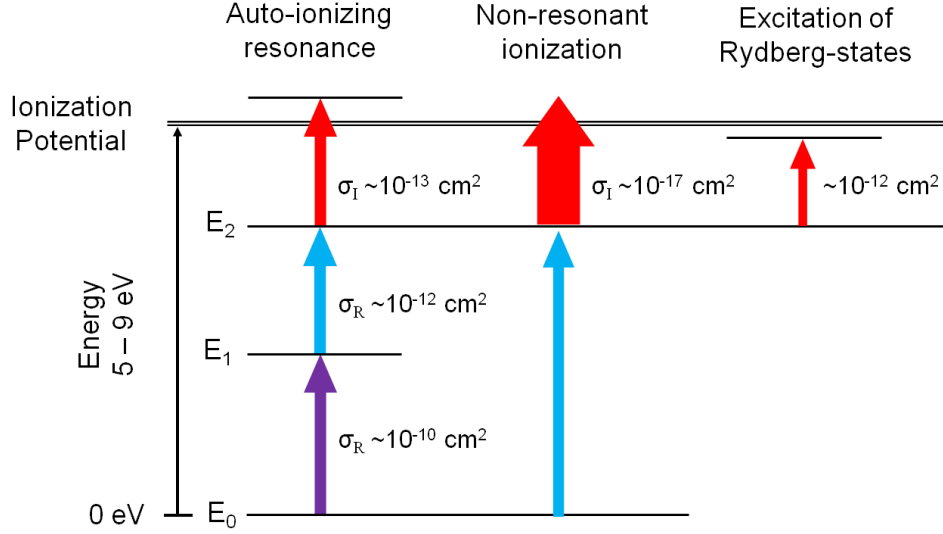


Figure 2.2– Alternative ways to reach the ionization potential for the final step: Auto-ionizing resonances, non-resonant ionization to the continuum and excitation of Rydberg-states. The cross section of each atomic transition is also indicated.

The atoms excited to high-lying Rydberg states are ionized in combination with a non-resonant mechanism of ionization: Thermal radiation, collisions, electric fields, IR lasers, etc. The ionization of laser-excited Rydberg states in a hot cavity is predominantly carried out by the blackbody radiation field. For an efficient and selective ionization, an individual excitation scheme and strong atomic transitions are needed for each element of interest. The first two excitation steps can be typically found in literature (NIST database [74] or Kurucz tables [75]) while extensive spectroscopic investigations are usually needed to find suitable high-lying Rydberg states or auto-ionizing states. In addition, some of the transitions are not accessible within the available wavelength tuning range of the laser system used. For this reason, high-power lasers are often applied in the last step for a non-resonant transition to the continuum.

2.2 RILIS requirements

A Resonant Ionization Laser Ion Source is composed of two parts: The interaction device (ion source) and the laser system (fig. 2.3). The ion source is connected to the hot furnace target area. It is installed on a high voltage (HV) platform at the isotope production front end of the on-line mass separator. This is often based technically on a conventional surface ionization source and ideally provides confinement of the radioactive atomic vapor until it is ionized. The laser system allows the resonant ionization of radioactive atoms. The laser laboratory is separated from the target ion source area in a temperature controlled room.

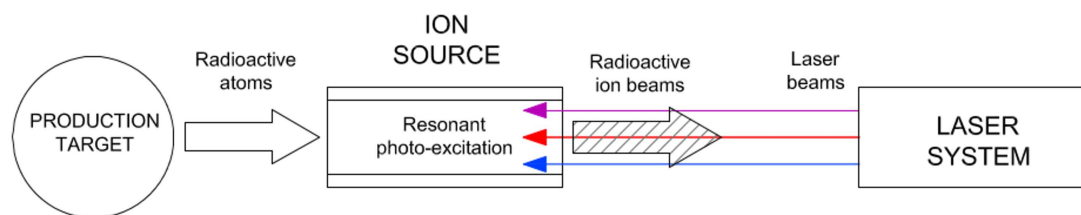


Figure 2.3– Resonant Ionization Laser Ion Source diagram. The radioactive atoms coming from the target are introduced into the ion source. Here the laser beams produce the resonant photo-excitation of the radioactive atoms and a radioactive-ion beam is generated.

The laser system consists of a set of two or three tunable lasers, which allow to select the proper wavelength for resonant photo-excitation with a high energy per pulse. The laser beams, which are overlapped in space and time, are transported from the laser room to the ion source through the mass separator and the extraction system, and then the ionization takes place inside the ionization tube. Finally, the generated radioactive-ion beams are extracted and mass selected. An introduction to the laser technology and a description of its origin can be found in appendix A.

The main requirements of a laser ion source for on-line applications are universality, selectivity, efficiency, reliability and rapidity [48]. One of the fundamental characteristics that motivated the development of RILIS is the **universality** of the method. As already mentioned, 80% of the chemical elements of the periodic table can be ionized by the RILIS technique. The **selectivity** (ionization of one element at a time) is the strong point of the RILIS technique and its main advantage in comparison with other ion generation techniques [76]. The combination of laser element selective ionization and mass separation allows to select precisely a single isotope (fig. 2.4). The selectivity can be improved by modifying the conventional RILIS configuration (i.e. the laser-atom interaction) or by use of purification equipments to the RILIS line (section 2.4.1). These methods allow the production of highly pure ion beams.

The **efficiency** of the technique depends on the atomic species and the ionization schemes chosen. For some elements, the efficiency can reach values up to 40% [77]. In general for most of the elements, ionization efficiencies vary between 5 and 30% [1]. The **reliability** of RILIS is determined by the long-term stability of the laser system. The capability to adapt the method to any possible setting without a big effort is called the versatility of the system. The laser system is outside the radiation area and the only thing that remains in the radiation area is the ionization tube, which is simple and robust. Finally, the **rapid nature** of the process allows to reduce decay losses in the ion source. The ionization time is negligible compared to the release time from the production target, and the effusion time in the target and ion source system.

Z	53	¹²⁵ I 59.4 D	¹²⁶ I 13.11 D	¹²⁷ I 100	¹²⁸ I 24.99 M	¹²⁹ I 15700000 Y	¹³⁰ I 12.36 H	¹³¹ I 8.0207 D	Resonant laser ionization
	52	¹²⁴ Te 4.74	¹²⁵ Te 7.07	¹²⁶ Te 18.84	¹²⁷ Te 9.35 H	¹²⁸ Te 31.74	¹²⁹ Te 69.6 M	¹³⁰ Te 34.08	
	51	¹²³ Sb 42.79	¹²⁴ Sb 60.20 D	¹²⁵ Sb 2.758 Y	¹²⁶ Sb 12.46 D	¹²⁷ Sb 3.85 D	¹²⁸ Sb 9.01 H	¹²⁹ Sb 4.40 H	
	50	¹²² Sn 4.63	¹²³ Sn 129.2 D	¹²⁴ Sn 5.79	¹²⁵ Sn 9.64 D	¹²⁶ Sn -	¹²⁷ Sn 2.10 H	¹²⁸ Sn 59.07 M	
	49	¹²¹ In 23.1 S	¹²² In 1.5 S	¹²³ In 5.98 S	¹²⁴ In 3.11 S	¹²⁵ In 2.36 S	¹²⁶ In 1.60 S	¹²⁷ In 1.09 S	
	48	¹²⁰ Cd 50.8 S	¹²¹ Cd 13.5 S	¹²² Cd 5.24 S	¹²³ Cd 2.1 S	¹²⁴ Cd 1.25 S	¹²⁵ Cd 0.65 S	¹²⁶ Cd 0.506 S	
47	¹¹⁹ Ag 2.1 S	¹²⁰ Ag 1.23 S	¹²¹ Ag 0.78 S	¹²² Ag 0.52 S	¹²³ Ag 0.293 S	¹²⁴ Ag 0.172 S	¹²⁵ Ag 166 M S	Magnetic deflection	
		72	73	74	75	76	77	78	
		n							

Figure 2.4– Schematic explanation of the RILIS Z-selectivity. In this case, ^{125}Sn is selected. All different isotopes of the same element are ionized (blue). Then the selectivity takes place when the mass of the isotope is selected by the mass spectrometer (yellow). The combination of element selective resonant laser ionization and mass selectivity by means of a mass separator ensures the nuclei selectivity (green).

2.2.1 Laser specifications for the RILIS technique

There is a wide variety of lasers, but the RILIS technique demands some specific laser requirements in order to access many elements. For that reason, the possible laser types that finally can be used are limited. A suitable laser for the RILIS has to fulfill the following requirements: High output power, high repetition rate, appropriate wavelength selection, possibility for easy generation of higher harmonics, GHz spectral linewidth and suitable synchronization between pulses from several laser resonators.

2.2.1.1 Pulsed laser

The output power has to be sufficient to transfer the energy to the atomic transition for efficient resonant photo-excitation. Non-resonant transitions (specially for the ionization step) have a cross-section below 10^{-17} cm². In this case, a CW (continuous wave) laser beam from a frequency doubled Nd:YAG laser would require several kW for a 3 mm spot size to saturate a non-resonant transition [78]. A pulsed laser is a way to reach a peak power close to this value. The Q-switch technique can be used to deliver pulsed laser beams with a large amount of light radiation in a short period of time, where the peak power depends on the pulse length.

This technique is based on the accumulation of the energy provided by the pump laser and a sudden release afterwards. This is reached by increasing the resonator losses controlled by the Q-factor (Q stands for quality). During the low Q-factor, while the gain medium is being pumped, the high cavity losses inhibits feed-back in the system and prevents the laser emission. The energy is accumulated in the form of population inversion between the fundamental and the excited state, if the accumulation time is shorter compared to the decay period.

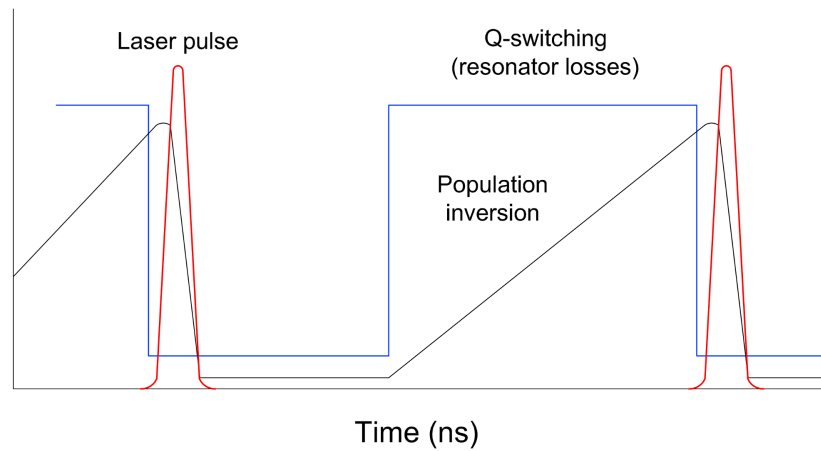


Figure 2.5– Diagram of Q-switch mechanism. The reduction of the cavity losses in population inversion causes the emission of an intense laser pulse.

Then, the system is suddenly changed to a high Q-factor (or low loss mode) if the system is in a high grade of population inversion. Then induced emission occurs and will be strongly amplified, increasing the number of photons in the cavity and causing a sudden emission of energy in a form of an intense laser pulse (fig. 2.5). The laser pulse is generated in a short pulse due to the immediate exhaustion of the accumulated energy and decrease of population inversion, until the gain medium only provides rather low gain and the laser emission stops. If the Q-factor is switched again to a low value (high loss mode) the system restarts a cycle and the gain medium starts again to accumulate energy [79].

2.2.1.2 Repetition rate

For efficient ion-source operation the temporal and spatial overlap of laser light and atoms has to be optimized. In a hot-cavity typically there is a continuous flow of atoms through the ionizer, that when using pulsed lasers is only temporally filled with the laser light. The inverse of the atom residence time within the laser interaction region gives the required laser

frequency. For a hot-cavity RILIS, it was calculated that a ~ 10 kHz laser repetition rate provides the necessary interaction for the required duty cycle [80]. One can calculate the most probable velocity of atoms (v_p) given by the Maxwell-Boltzmann distribution:

$$v_p = \sqrt{\frac{2k_B T}{m}} \quad (2.1)$$

where k_B is the Boltzmann's constant, T is the temperature [K] and m is the mass of the particle [kg]. The most probable velocity of Sn atoms at 2000 K is $v_p \simeq 530$ m/s, and their free-flight path between two laser pulses at 10 kHz repetition rate is of the order of few centimeters, which is the approximate length of the ionizer. However collision effects and sticking times on surfaces are not taken into account, which on average increase the residence time of the atoms in the ionizer and therefore increase the probability of laser-atom interaction.

2.2.1.3 Wavelength selection

The laser system has to be tunable over a wide range of wavelengths to allow the selection of the required electronic transitions in a specific laser ionization scheme. The length of the resonator sets the free spectral range $\Delta\nu$, where the frequency spacing in the cavity fulfills the relation:

$$\Delta\nu = \frac{c}{2L} \quad (2.2)$$

where c is the speed of light [m/s] and L is the length of the cavity [m]. The frequencies that are constructively superimposed are called longitudinal modes of the resonator. The spectral width of the resonator modes $\delta\nu$ is inversely proportional to the finesse (F):

$$\delta\nu = \frac{\Delta\nu}{F} \quad (2.3)$$

The finesse of a resonator is determined by the reflectivity of the cavity mirrors (R_1, R_2):

$$F = \frac{\pi(R_1 \cdot R_2)^{1/4}}{1 - (R_1 \cdot R_2)^{1/2}} \quad (2.4)$$

Moreover, the cavity mirrors are coated to have high reflectivity in a certain wavelength range (e.g. 840-940 nm) [81]. Several techniques can provide fine laser wavelength control through the selection of a particular longitudinal cavity mode, such as birefringent-etalon or grating prisms configurations.

Birefringence is the optic property of some materials, with non-isotropic refractive index, to split a light beam into two linear polarized beam lights, whose polarization planes are perpendicular to each other, the ordinary and extraordinary rays. Due to the birefringence, the ordinary and extraordinary components experience different refractive index (n_o and n_e respectively). As a consequence, propagation changes with the direction of the light and causes a phase shift. If a p-polarized laser beam is traveling through a birefringent medium, it will be transformed into elliptical polarization, with the subsequent reflections and losses for the s-polarized fraction when passing through with cavity elements at Brewster's angle.

A Lyot filter is a special type of birefringent optical element which produces a narrow passband of transmitted wavelengths using several birefringent plates oriented at Brewster's angle. The free spectral range depends on the thinnest plate (L_{thin}) and the spectral width of the resonator modes depends on the thickest plate (L_{thick}).

$$\Delta\nu_{Lyot} = \frac{c}{\Delta n \cdot L_{thin}} \quad (2.5)$$

$$\delta\nu_{Lyot} = \frac{c}{2^{N-1}} \Delta n \cdot L_{thick} \quad (2.6)$$

The angle between the optical axis of the crystal and the propagation direction of the laser beam can be changed by rotating the Lyot filter, that will modify Δn and therefore the selected wavelength. In addition, the etalon filter (also known as Fabry-Pérot interferometer) consist of one or several parallel reflecting plates, where the transmission spectrum depends on the resonances of the interferometer. In this way some wavelengths will be selected depending on the interferometer parameters (fig. 2.6).

The **diffraction grating** is an alternative way of wavelength selection. It consists of a surface-treated mirror that disperses the incident light at an angle that depends on the wavelength and the grating parameters.

$$\theta_m = \arcsin\left(\frac{p\lambda}{s} - \sin\theta_i\right) \quad (2.7)$$

where θ_m is the angle between the diffracted ray and the grating's normal vector, θ_i is the incidence angle, s is the distance pattern, λ is the wavelength and p is an integer representing the propagation mode.

A diffraction grating can be used to select a narrowband spectrum from a broadband laser range by means of the propagation mode. The diffraction grating allows scanning the tunable laser continuously over a wide range of wavelengths [82].

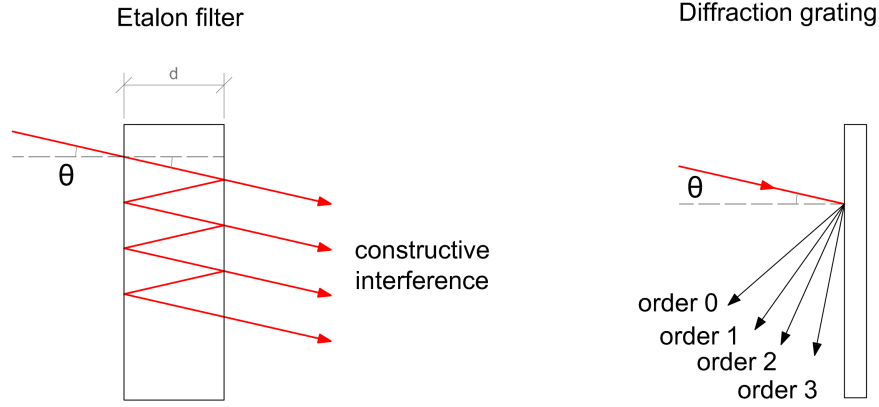


Figure 2.6— Two ways of wavelength selection: (Left) The etalon is a Fabry-Pérot interferometer where the additive interference of the successive reflections can reduce the linewidth. (Right) The diffraction grating is a surface-treated material in a periodic structure that produces the diffraction of light into several beams that travel in different directions, as the diffraction angle depends on the wavelength.

2.2.1.4 High harmonic generation

None fundamental radiation laser system can provide a full range of wavelengths. This limitation can be overcome by using non-linear effects of certain birefringent materials, which can be used for higher harmonic generation (i.e. frequency doubling, tripling or quadrupling). An input laser wave generates another wave, which is the sum of several incident photons (i.e. twice the optical frequency in the case of second harmonic generation). The birefringent materials generate a non-linear polarization of the incoming laser radiation. This polarization oscillates with twice fundamental frequency, and radiates an electromagnetic field with doubled frequency.

By this method a fundamental 1064 nm Nd:YAG laser beam provides a 532 nm output (by using an appropriate non-linear medium and the phase matching angle θ). Some of the most common crystalline materials are β -barium borate (BBO), potassium titanyl phosphate (KTP), and lithium triborate (LBO). The combined use of tunable lasers and high harmonic generation technique can extend the available wavelength range.

2.2.1.5 Spectral linewidth

The laser system might have a short spectral bandwidth to selectively choose the correct transition. However, the random thermal vibrations of the emitting atoms, while the energy of the atomic transition remains constant, produce a broadening of the emitted frequency.

A single Gaussian mode TEM₀₀ (transverse electromagnetic modes) is fully advised, which comes from the cavity propagation modes and the use of the previously described optical elements to select the wavelength (i.e. birefringent plate, etalon filter and grating mirror). For a single mode pulsed laser, the linewidth is Fourier limited (~ 30 MHz for a 30 ns pulse). For high ionization efficiency it is important that the laser linewidth is comparable to the Doppler broadened atomic linewidth due to the ion source conditions. For an atom in motion with velocity v , and a transition frequency at rest ω_0 , the Doppler shift of the angular frequency is given by:

$$\omega = \omega_0 \left(1 + \frac{v}{c} \right) \quad (2.8)$$

The number of atoms with velocity v_p (from the Boltzmann distribution, equation 2.1) and atomic mass m is given by:

$$n(v_p)\delta v = N \sqrt{\frac{m}{2\pi k_B T}} e^{\frac{-mv_p^2}{2k_B T}} dv \quad (2.9)$$

The distribution of the frequencies is then:

$$P(\omega) = P(\omega_0) \exp \left[\frac{-mc^2(\omega - \omega_0)^2}{2k_B T \omega^2} \right] \quad (2.10)$$

This is a Gaussian function centered at ω_0 with a width $\Delta\omega_D$ [83]:

$$\Delta\omega_D = \frac{2\omega_0}{c} \sqrt{\frac{2K_B T}{m} \ln 2} \quad (2.11)$$

2.2.1.6 System synchronization

Two or three laser beams are used to assure selective ionization for a given element. A temporal overlap and low pulse timing jitter are required, and the differences between pulse lengths should not exceed the atomic state lifetime [11]. The temporal lasing depends on several resonator parameters (e.g. pump power, gain medium position, resonator mirrors, etc.) and it can be actively modified by a Pockels cell installed inside the laser cavity. A Pockels cell is an electro-optical device that produces birefringence in a material induced by an electric field. With a Pockels cell, it is possible to actively control e.g. to Q-switch of the cavity and modulate the timing of each laser by a common master clock with direct access to the Q-switch drivers.

2.2.2 Available laser systems for the RILIS technique

The need of a reliable and robust tunable narrow-bandwidth laser system motivates an effort in searching the best laser system for the RILIS method. However, the special requirements limit the quantity of lasers available for this technique. Dye laser systems pumped by copper vapor lasers and frequency double/tripled Nd:YAG lasers have been extensively used historically in many facilities due to their unique range of wavelengths and output power. Nowadays, the use of solid-state titanium:sapphire (ti:sa) laser systems, which can be easily manipulated and maintained, has become an alternative. These two laser systems, which are characterized by a different gain medium, will be presented in the following.

2.2.2.1 Dye laser

The gain medium of a dye laser is a liquid dissolution of organic dye compounds in a liquid solvent, such as methyl-alcohol. Its main advantage is the availability of a wide variety of different dyes that allow (combined) the tunability in a wide range of frequencies. One of the main dyes used due to its superior stability and tunability is rhodamine 6G, with chemical formula $C_{26}H_{27}N_2O_3Cl$ and structure of several benzene rings. This molecule in solution has a complicated electronic structure, because each electronic level is subdivided into a vast number of vibrational and rotational levels (fig. 2.7).

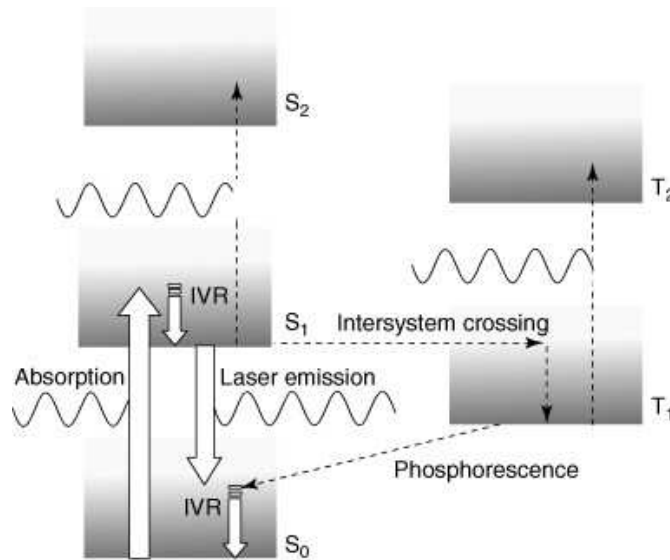


Figure 2.7– Jablonski diagram for a dye laser molecule. Each gradient region consist of several vibronic levels of the molecule. The vertical arrows show absorption and emission transitions. The radiative transitions follow the Franck-Condon principle [84].

The stimulated emission occurs through these vibrational and rotational levels. This makes possible to access a broad absorption and emission bandwidths, and it allows the selection of the required frequency. However, the emission has to be pulsed due to some processes that limit the population inversion for long periods. It is possible anyway to emit at continuous wave (cw) creating a high speed flux of the dye. The wavelength range is selected by the dye nature, approximately 10 dyes are needed to cover a range from 540 to 850 nm.

In the very first RILIS implementations, the dye lasers were pumped by copper vapor lasers (CVL). Nowadays, reliable 10 kHz Q-switched Nd:YAG pump laser can be used to efficiently pump the dye solution with the required 10 ns pulse duration [85].

The main advantages of the dye laser system are the relative high power (< 12 W, 1.2 mJ/pulse with 50 W laser pump), the short pulse duration (~ 8 ns) which increases the peak power, and the wide range of accessible wavelengths (540 nm to 850 nm). In addition, dye lasers can be pumped by 355 nm tripled Nd:YAG laser (20 W pump power) with a power output of ~ 3 W, and wavelength range from 375 to 550 nm. However the use of dyes and solvents needs maintenance (dye renovation), as well as surveillance and security systems [86].

2.2.2.2 Titanium:Sapphire Laser

Titanium:sapphire (ti:sa) lasers have recently emerged as an alternative to dye lasers for RILIS applications. A ti:sa is a vibronic laser, where the active medium is a sapphire crystal (matrix) with titanium ion impurities, which emit the laser radiation. The sapphire matrix is transparent and it has suitable thermal properties (i.e. it can be easily cooled). The emission light is due to electronic levels and vibrations of the crystal structure (phonons). This effect amplifies the possible emission and absorption lines, and it allows the selection of a specific wavelength (fig. 2.8).

In addition the absorption and emission curves are displaced such a way that an efficient pumping process is possible (fig. 2.9). The ti:sa lasers have the broadest spectral tuning range of all the currently known solid-state lasers. Therefore, they are perfect candidates for the RILIS. The ti:sa lasers are often optically pumped in the absorption range of 500 nm by a frequency doubled Nd:YAG laser. Nd:YAG is another solid-state laser of a YAG (Yttrium Aluminium Garnet) matrix doped by Neodymium ions. The characteristics of the laser system used during this work will be explained in section 3.2.2.

The ti:sa laser system is reliable, easy to handle, and (almost) maintenance free. Another advantage is a broad tuning range in the infra-red region from ~ 700 nm to 1000 nm. On the other hand, the weaknesses are the lower laser power compared to dye laser (< 5 W, 0.5 mJ/pulse with 20 W laser pump), and a longer pulse duration (~ 50 ns) that affects the laser-atom interaction.

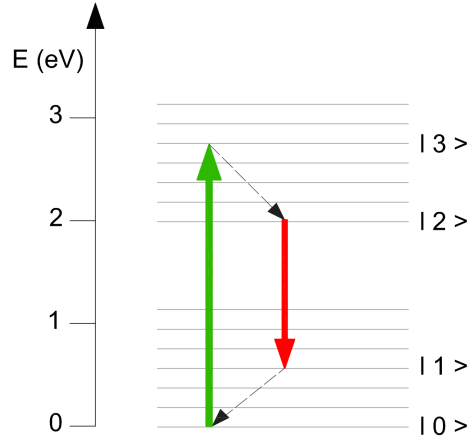


Figure 2.8– Jablonski diagram for a titanium:sapphire crystal. The lasing path in a four level scheme is marked by the arrows. The green arrow indicates the absorption of a photon (pump) and the red arrow shows a possible emission transition. Grey arrows indicate fast decays to vibronic ground states.

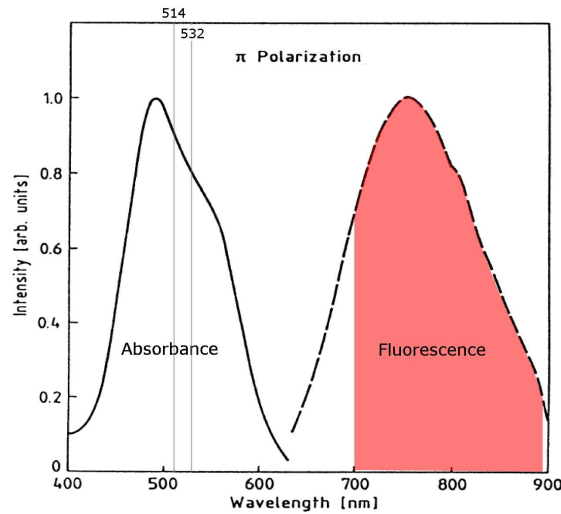


Figure 2.9– Absorption and fluorescence emission spectrum of a ti:sa laser. Doubled Nd:YAG (532 nm) and Ar laser (541 nm) can be used to pump the ti:sa crystal. The emission range of the ti:sa system used is indicated by a red area [87].

2.2.2.3 Comparison between dye and ti:sa lasers

As already mentioned, the two main tunable laser options for RILIS are dye and ti:sa. In table 2.1, the described characteristics are summarized. Other laser systems, such as Cr:Forsterite laser, are today under development but they did not pass the laboratory phase.

Table 2.1 – Comparative table of titanium:sapphire and dye laser options.

	Ti:sa	Dye
Active medium	1 ti:sa crystal	> 10 different dyes
Condition of aggregation	solid-state	liquid (solutions of organic molecules)
Tuning range	680 - 980 nm	540 - 850 nm
Pulse duration	~ 50 ns	~ 8 ns
Synchronization	Q-switch, pump power	optical delay lines
No. of developed schemes	43	47
Maintenance	none	renew dye solutions
Wavelength set	change optics set	change optics set + change dye

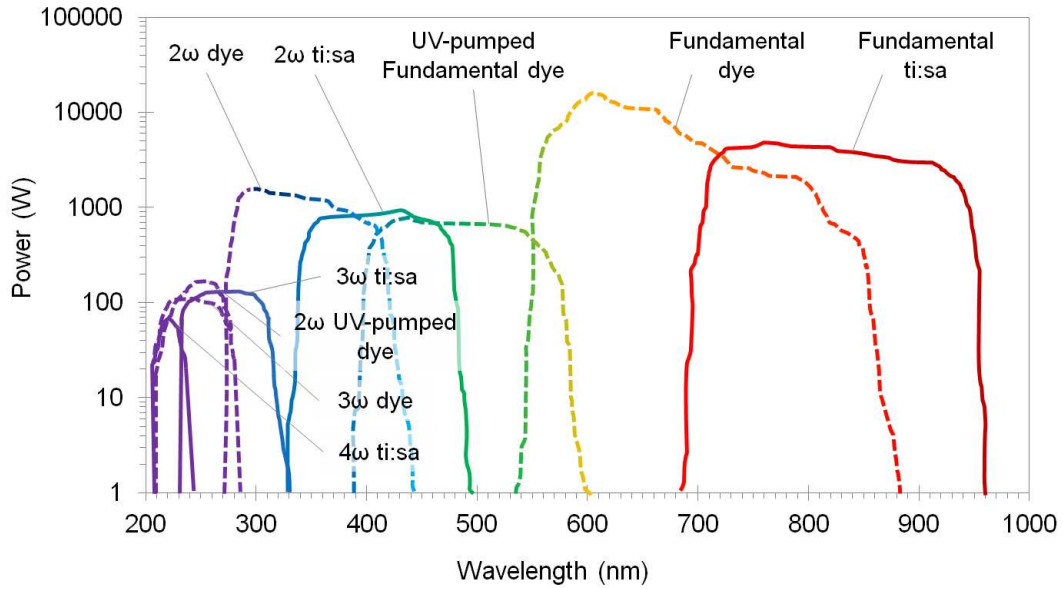


Figure 2.10– Accessible wavelengths for ti:sa (solid line) and dye laser (dashed line) systems as a function of the output power. The fundamental radiation, the second harmonic (2ω), third harmonic (3ω) and fourth harmonic (4ω) are shown.

In fig. 2.10, both systems are compared as a function of the output power. In general, a broad access to the visible wavelengths is observed. Green and UV pumped pulsed dye lasers have a fundamental tuning range that covers most of the visible spectrum, but the dye lasers used below 500 nm and across part of the near infrared range are either inefficient or unstable. The ti:sa lasers manage to reach wavelengths deeper in the IR region, but they lack of shorter wavelengths between 500 nm to 700 nm. For this reason, it is preferable to have both laser types available at the same RILIS installation [88].

The dye system can be pumped harder than the ti:sa crystal and higher pulse energies can be reached. However, the efficiency of the lasing process strongly depends on the dye nature used, thus the output power decreases for near infrared wavelengths. On the contrary, the ti:sa lasers provide an average output power for all wavelengths. A full ti:sa laser system was used during this thesis to provide the required wavelengths.

2.3 Laser-atom interaction devices

The interaction device of the ion source is the cavity where the ionization takes place. There are several techniques to confine the radioactive atoms in order to maximize the effect of the laser-atoms interaction. Historically, the first idea was to ionize the elements in an atomic beam, which was intersected by the laser light in order to produce the ionization [89]. Afterwards, other particular geometries were tested to increase the efficiency, such as cylindrical cavities [90, 91, 92, 93, 94] or capillary geometries [95, 96]. In the latter case, the idea was to confine the radioactive vapor to allow a selective ionization of the isotopes of interest, while maintaining a low ratio of contaminants from the container. Nowadays, the two main techniques are the hot cavity and the gas cell.

The geometries of the interaction device depend specifically on the nature of the radioactive atom, the technical requirements of the RILIS and the laser-atom interaction behavior. For these reasons, the hot cavity furnace is normally designed as two cylinders: a transfer tube that connects with the production area, followed by an ionization tube where the laser and the atoms interact. The gas cell is a cavity filled by gas where the radioactive vapor is stopped and confined until it is irradiated by the laser light. For off-line tests and laser spectroscopy measurements, the elements of study are placed directly inside the interaction device.

2.3.1 Hot cavity

The hot cavity RILIS interaction device is widely used for the production of ion beams from ISOL facilities. The objective of the cavity geometry is to keep the atoms confined in a volume to be irradiated by the laser light and to drive the ions during their drift towards the extraction region [97] (fig. 2.11). The mechanisms of confinement inside a hot cavity ion source have been studied for many years (section 2.5). To improve the ionization ratio of the short-lived radioactive species, the ion source must be operated at high temperatures to allow fast transport of the exotic nuclei to the ion extraction system. This temperature should be kept high enough to avoid that the atoms stick to the walls.

The ion confinement is a natural consequence of the electron emission by high temperature metallic surfaces, which builds a few eV negative potential well [47]. The potential prevents the photo-ions to hit the wall and recombine to neutral wall atoms. In addition, the ions are pushed towards the exit due to the internal electric field, created by the potential charge of the resistively heated tubular cavity, if the resistivity is high enough [97].

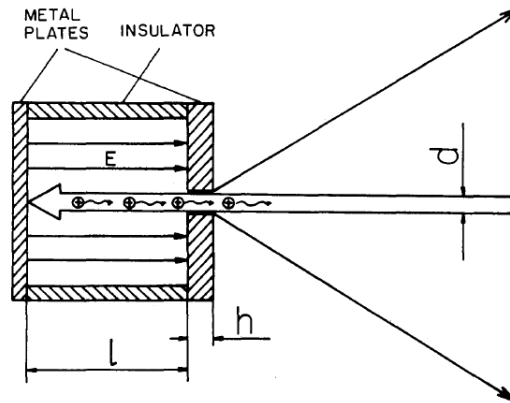


Figure 2.11– Scheme of one of the first designs of the hot cavity concept described by Andreev, Mishin and Letokhov in 1988, with the laser interaction indicated by an arrow [92].

The influence of the electric field on the ion extraction efficiency will be a fundamental part of this thesis (section 4.1.2). The hot cavity furnace is resistively heated by the Joule effect to temperatures around 1400 K to 2000 K for a fast release and a short wall sticking time. Consequently, thermal ionization of atoms may take place on the hot surface, particularly if the ionization potential of the atom is low (section 2.4). In addition, the ion source is set in a high-voltage platform to extract and accelerate the ion beam. The hot cavity is normally made of refractory metals (Ta, W, Nb) or refractory carbides (C, TaC) to ensure reliability at very high temperatures [95, 96, 98].

The ionization efficiency in the cavity (ε_{cavity}) of the ion source shown in figure 2.11, can be calculated from equation 2.12 [95], by comparing the rate of photo-ionization with the rate of diffusion as neutral atom out of the hot cavity [94]:

$$\varepsilon_{cavity} = \frac{f \cdot \varepsilon_{ion}}{f \cdot \varepsilon_{ion} + \frac{v}{4L}} \quad (2.12)$$

where ε_{ion} is the probability for resonant photo-ionization, f the repetition rate of the lasers [Hz], v is the thermal velocity of the atoms [m/s] and L is the cavity length [m]. As it can be seen from equation 2.12, a high efficiency requires a high pulse repetition rate of the lasers and a long tube length. It is important to note that this equation does not show any dependency

of the diameter of the tube on the efficiency. The saturation in all transitions is a prerequisite to maximize the ionization efficiency ε_{ion} . If the transitions are not saturated, the ionization can be increased by focusing the laser beams, which however reduces the effective ionization volume due to a lower quantity of atoms effectively interacting with the laser radiation, with a possible consequence of reduction of efficiency. Another formula, based on the kinetic theory of gas diffusion [97], calculates the ionization efficiency in a capillary $\varepsilon_{capillary}$, where $d \ll L$:

$$\varepsilon_{capillary} = \frac{f \cdot \varepsilon_{ion}}{f \cdot \varepsilon_{ion} + \frac{2}{3} \cdot \frac{d \cdot v}{L^2}} \quad (2.13)$$

Therefore the ionization efficiency increases when the ratio d/L^2 decreases. This formula established the conventional geometry of the RILIS hot-cavity ionizer with 3 mm diameter and 35 mm length. This hypothesis is discussed in this thesis by the modification of diameter and length ion source parameters. The different RILIS hot cavities approaches will be described in section 4.1 and the results in section 5.1.

2.3.2 Gas cell

A different approach for the interaction device is the gas cell, where the target is implemented in a chamber filled by Ar or He at a working pressure of 500 mbar [99]. This technique is under development since 1992 at Leuven Isotope Separator On-Line facility [100], and it has been implemented in other facilities such as IGISOL, at University of Jyväskylä [101].

In this technique the cyclotron beam enters into a gas cell and interacts with one or more uranium targets. Then the fission products are thermalized by the gas [102]. The fission products, which are in an ionized state, are neutralized through recombination processes with the electrons, which are mainly created by the primary beam passing through the gas cell [103]. The reaction products are transported together with the gas to the laser ionization area where they are resonantly photo-ionized (fig. 2.12). The laser beams are sent into the gas cell via the central axis or perpendicular to the axis, close to the exit hole. After ionization, the ions are sent through the exit hole and enter the a multipole ion guide system (SPIG) to improve the transport efficiency [104].

The gas cell is usually made of aluminium. There have been a wide variety of geometries from the first designs [25] to the actual Leuven geometry [105]. Other improvements were done, such as supersonic in-gas-jet used for laser spectroscopy [106]. For spectroscopy measurements, the limitation is the Doppler broadening caused by the atom velocity distribution, the pressure broadening (and additionally pressure shift) induced by collisions with the surrounding gas [107, 108], the laser-power broadening, and the intrinsic bandwidth of the laser [106].

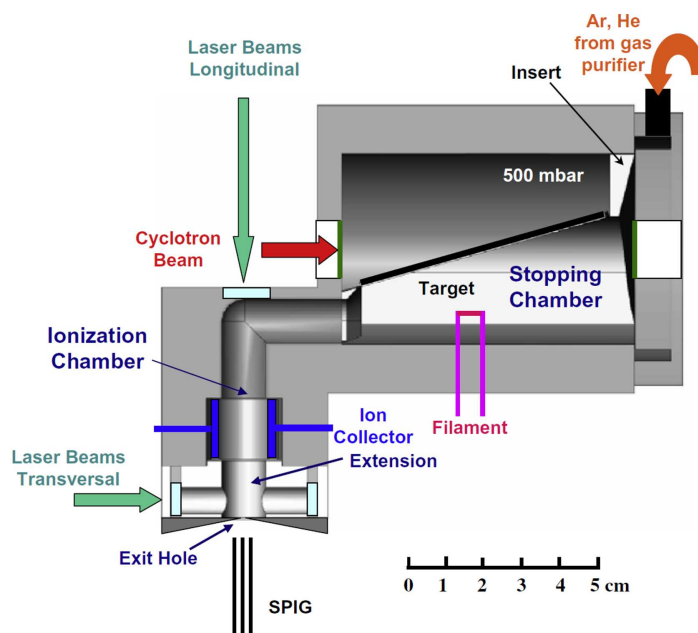


Figure 2.12– Scheme of the dual chamber laser ion source gas cell [105].

This technique will be used in the future S^3 at SPIRAL2 (GANIL), based on fusion-evaporation reactions, for the production of nuclei in the $N=Z$ region and heavy and super heavy elements [71].

2.3.3 Comparison of interaction devices

The interaction device for the RILIS has to be chosen carefully depending on the required element, and both systems have their advantages and disadvantages (table 2.2). In the hot-cavity technique, higher isotopic productions can be obtained due to the thick production target. Nevertheless, the gas cell approach is better adapted to the production of refractory elements or for some short-living isotopes [71].

Table 2.2 – Comparative table between hot cavity and gas cell interaction devices.

	Hot-cavity	Gas cell
Isotopic production in the target	High in target yield	Low in target yield
Target	Thick	Thin
Refractory elements	Stick to target material	Transported by gas to ionization region
Delay time (Short-lived isotope production)	Depends on chemical properties and diffusion behavior	Depends on the gas cell geometry and gas velocity (<100 ms)

2.4 Contaminant reduction and ion beam purification

In the last two decades laser photo-excitation for radioactive-ion beam generation has been implemented due to its extremely high **selectivity** compared to other radioactive-ion beam generation methods. One of the drawbacks of the hot-cavity method is the non selective surface ionization of unwanted isobaric contaminants due to the high temperature needed (section 1.3). The process has to take place into a hot furnace in order to maximize the ionization efficiency [93]. Due to this, the materials used for the ion source should operate satisfactory at high temperatures, and only few metals and ceramics are available. The high temperature causes surface ionization of contaminants and the production of molecular sidebands, which decrease drastically the selectivity in the hot cavity [109]. Surface ionization occurs during thermal desorption, when the atoms desorb from the walls of the hot cavity. The development of RILIS for radioactive-ion beam production is often focused on the research of techniques that can reduce the generation of contaminants by several ways: Materials that diminish the generation of surface ions [110, 111], systems to purify the ion beam after ionization [112], purification by quartz transfer line [113] or by applying specially oriented electric fields [114, 115].

2.4.1 Selectivity

The selectivity of a hot-cavity ion source can be calculated as the ratio of the total ionization efficiency (laser photo-ionization efficiency $\beta_{photo-ion}$ + surface ionization efficiency β_s) relative to the surface ionization efficiency (β_s) [47]. If $I_{laser-on}$ is the current obtained with resonant laser ionization and $I_{laser-off}$ is the current without resonant laser ionization, the selectivity S is equal to:

$$S = \frac{I_{laser-on}}{I_{laser-off}} = \frac{\beta_{photo-ion} + \beta_s}{\beta_s} \quad (2.14)$$

The production cross-section for isotopes far from stability is orders of magnitude lower than that of isobars closer to stability (fig. 2.13). For that reason, the selectivity is highly important for production of ion beams of exotic isotopes. However, the high temperature target and ion sources suffer from unwanted surface ionization of low ionization potential elements (i.e. alkalis) [114]. Therefore, the selectivity depends on the ionization potential of isobar atoms. In conclusion, the production of contaminants has a high dependence on the interaction device material, the geometry of the interaction device and the requested element [95]. The selectivity can be measured off-line if the element is produced by surface ionization and laser ionization. Other possibility is to measure relative contamination reduction of an alkali marker to analyze the contamination suppression. This technique will be described in chapter 5.5.

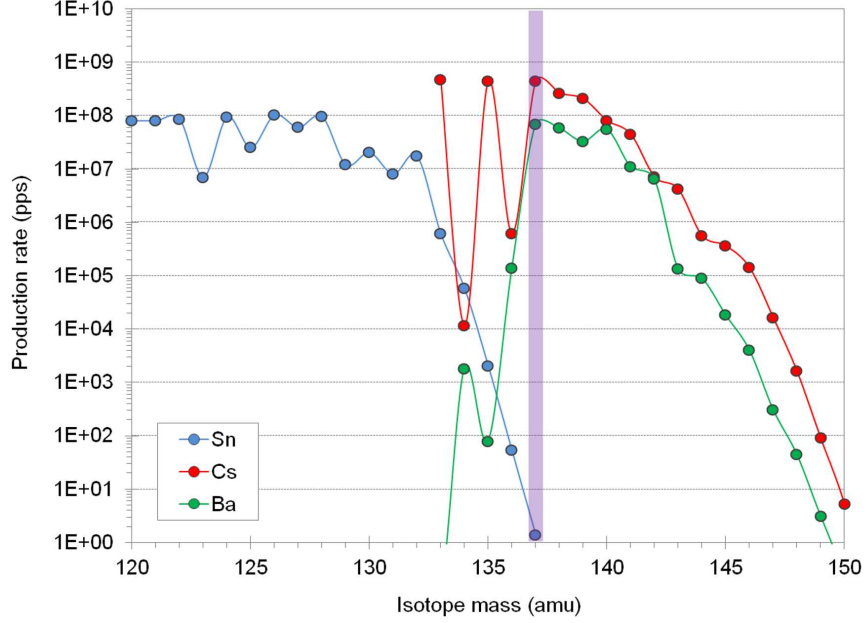


Figure 2.13– Example of the dependence of the selectivity on the mass of produced isotope. Production calculation of Sn, Cs and Ba isotopes for the SPIRAL2 Phase-2 UCx target (280g) after post-acceleration by CIME. If the production of ^{137}Sn is required (purple), 8 orders of magnitude more of ^{137}Cs and ^{137}Ba will also be generated contaminating the ^{137}Sn ion beam.

As already mentioned, surface ionization is a competing process to resonance ionization and a source of unwanted isobaric contaminations [116]. Therefore, one of the main developments for the RILIS is to limit the surface ionization inside the ion source. The thermal ionization process on a surface of a hot cavity is described by the Saha-Langmuir equation [117]:

$$\alpha_s = \frac{n_i}{n_0} = \frac{\sigma_i}{\sigma_0} \cdot \exp\left(\frac{\varphi - W_{IP}}{k_B T}\right) \quad (2.15)$$

where α_s is the degree of ionization, n_i and n_0 are the ion and neutral concentrations, σ_i and σ_0 are the statistical weights of ionic and atomic ground states respectively, φ is the work function of the material [eV], W_{IP} is the ionization potential of the element of interest [eV], k_B is the Boltzmann constant and T is the temperature [K]. According to equation 2.15, when $\varphi > W_{IP}$, the exponential is positive and the surface ionization degree increases. On the contrary, when $\varphi < W_{IP}$ the exponential is negative and the surface ionization degree decreases rapidly. The surface ionization efficiency β_s is defined as:

$$\beta_s = \frac{n_i}{n_i + n_0} = \frac{\alpha_s}{1 + \alpha_s} \quad (2.16)$$

According to equations 2.15 and 2.16, it is possible to conclude that the degree of surface ionization depends on the temperature, the ionization potential of the required element, and the work function of the hot-cavity. Rubidium will be used as a contaminant marker (section 5.5). For a rubidium ion beam ($W_{IP} = 4.177$ eV) with a tantalum ionizer ($\varphi = 4.28$ eV) the surface ionization efficiency at 2000 K was calculated to be 90.89%. On the contrary, if a low work function material is used, for example ZrC with $\varphi = 3.65$ eV, the surface ionization efficiency decreases to 2.35%. A decrease of the work function property will decrease the surface ionization with the consequent amelioration of the selectivity.

However, in the case of hot-cavity, it has been observed that the ionization efficiency can be significantly higher than the one given by the Saha-Langmuir equation [118]. The following hypothesis has been proposed to explain the experimental results: The existence of a negative thermal plasma in the cavity with a plasma sheath potential (Φ_p) [V], which is induced by the intense electron emission from the heated walls. This plasma can generate a confinement of ions inside the cavity [47, 118] and an enhancement of the degree of ionization. The electron density n_e is formulated by the Richardson-Duchmann expression:

$$n_e = 2 \cdot \left(\frac{2\pi m e k_B T}{h^2} \right)^{3/2} \cdot \exp\left(\frac{-\varphi \cdot \Phi_p}{k_B T}\right) \quad (2.17)$$

where n_e is the electron density near the surface, m is the electron mass [kg], e is the elementary charge [C] and h is the Planck's constant. As it can be seen in equation 2.17, the plasma sheath can modify the ion beam confinement.

The plasma sheath potential is defined as:

$$\Phi_p = \frac{k_B T}{2e} \cdot \ln\left(\frac{n_e}{n_0}\right) \quad (2.18)$$

Assuming a thermal equilibrium in the volume of the cavity, the degree of ionization α_h is given by:

$$\alpha_h = \alpha_s \exp\left(-\frac{\Phi_p}{k_B T}\right) \quad (2.19)$$

With equations 2.19 and 2.18 the ionization efficiency in a hot cavity β_h can be calculated as a function of the degree of ionization:

$$\beta_h = \frac{n_{ih}}{n_{ih} + n_0} = \frac{\alpha_h}{1 + \alpha_h} \quad (2.20)$$

To conclude, it is possible to consider that materials with a low work function will improve the selectivity of the RILIS technique by the reduction of unwanted surface ionization elements (equation 2.15). On the other hand, the plasma potential can enhance the efficiency due to a better ion confinement. However, further studies will be needed to analyze the implication of different materials over the plasma sheath and the ion confinement.

2.4.2 Low work function materials

The work function is the minimum amount of energy that an electron needs to leave a solid. As it was discussed, a hot-cavity made of a low work function (φ) material should cause an increase in the selectivity due to a diminution of the surface ionized ions [111]. The chosen material has to be reliable at the high temperatures ($\sim 2000^\circ\text{C}$) required for the hot-cavity furnace and must resist a high radiation environment. For these reasons, only a few materials are available. The work function intrinsically depends on the material structure and crystallographic configurations. Possible materials that can be used for contaminant suppression were investigated. The candidate materials were divided into four categories:

- Refractory metals: Tungsten (W), Tantalum (Ta), Niobium (Nb) and Rhenium (Re).
- Carbon based: Graphite (C), Titanium carbide (TiC), Zirconium carbide (ZrC), Tantalum carbide (TaC) and Hafnium carbide (HfC).
- Boride based: Hexa-gadolinium boride (GdB_6), Tetra-gadolinium boride (GdB_4) and Hexa-lanthanum boride (LaB_6).
- Oxides: Calcium oxide (CaO), Strontium oxide (SrO), Barium oxide (BaO) and quartz (SiO_2).

Only limited information can be found about the different materials. Most of the work function information is measured at temperatures below the typical ion source operating temperature, and in most of the cases the extrapolation to high temperatures is not possible because the temperature dependence of each material is not known [119]. In addition, the work function property changes with crystallographic variations, for this reason a chemically identical material can have different work function with stoichiometric structural variations.

Some work function and material properties, available to be used as interaction device, will be presented and discussed on the following sections. The values for the material properties are mainly obtained from material handbooks such as CRC Handbook of Chemistry and Physics [120], CRC Reference Handbook of chemical and physical data [121], Materials Handbook [122] and Smithells Metals Reference Book [123] and specialized material webpages [124, 125]. In addition, selectivity studies of different ion source materials will be presented.

2.4.2.1 Refractory metals

The most employed materials for the interaction device are the refractory metals: Tungsten (W), tantalum (Ta), niobium (Nb), rhenium (Re), etc. They have been chosen for their good performance at high temperatures as well as the possibility to create all geometries, well known behavior, strength, easy machining and lower prize (compared with other materials). This makes for the moment the most reliable solution (most facilities use pure tantalum for all parts). However, as it is observed in table 2.3, pure metals have a high work function too. In addition, Some of these materials suffer from carburization in presence of carbon at high temperature and this is a disadvantage when used with UCx targets for the production of radioactive nuclei. For that reason other elements are under study, such as Rhenium [49] or monolithic carbide, which do not carburate [126].

Table 2.3 – Work function and properties of pure refractory metals.

	W	Ta	Nb	Re
φ (eV)	4.54 [119] 4.77 [120]	4.28 [119] 4.4 [120]	4.02 [119] 4.41 [120]	4.8 [124] 5.1 [127]
Resistivity ($\Omega \cdot m$) at 300 K	$5.65 \cdot 10^{-8}$	$1.35 \cdot 10^{-7}$	$1.52 \cdot 10^{-7}$	$1.93 \cdot 10^{-7}$
Tensile Strength, Ultimate (MPa)	980	900	240	1070
Elastic Modulus (GPa)	411	182	105	463
Thermal Conductivity (W/m·K)	163.3	59.4	52.3	48.0
Density (g/cm ³)	19.25	16.69	8.57	21.02
Melting point (K)	3695	3290	2750	3459
Welding	Possible	Possible	Not possible	Possible

Several studies of selectivity have been carried out. Ta and Nb cavities were tested for laser ionization of Tm ($W_{IP} = 6.18$ eV) and a selectivity of 10^4 was observed at temperatures of 1600 K. However increasing the temperature quickly decreased the selectivity to 30 at 2300 K [95]. In addition, on-line Tl beams were produced using hot W and Nb cavities, which provided selectivities between 10 and 20 at ISOLDE (CERN) [128, 61]. Moreover, cavities made of W and tungsten impregnated with BaO and SrO (W(BaOSrO)) were studied with a Ga ion beam at ISOLDE (CERN). The tube coated with BaOSrO showed an outstanding selectivity close to 10^6 at low temperatures ($<1200^\circ\text{C}$), and the selectivity in the case of W was <10 at high temperature. The contamination reduction for W(BaOSrO) was limited due to the destruction of the coating at >1400 K, which limited the efficiency of the process [111]. Finally, a test of Ga ionization ($W_{IP} = 5.9993$ eV) was performed in the ISOLDE ion source, which provided selectivities bigger than 1000 for W(BaOSrO) and between 10 and 20 for W [109].

2.4.2.2 Carbides

Carbide materials are an alternative solution as interaction device. Some examples are: Graphite (C), tantalum carbide (TaC), zirconium carbide (ZrC), titanium carbide (TiC) and hafnium carbide (HfC). For some authors, the carbon based materials show (approximately) the same values of work function as the refractory metals, but for other authors they have lower values (table 2.4). These materials have higher resistivity due to its isolating nature (which makes possible a higher resistive heating with less electrical current). As ceramics, they have a fragile break, but their high elasticity modulus prevents from deformations in stress conditions compared to metals. They also display chemical inertness [129]. In addition, some authors claimed that the work functions of refractory carbides can be reduced by heating in vacuum due to a loss of carbon from the surface during the activation process at high temperatures [130]. They also have high melting points and therefore they can be heated to high temperatures (>2000 K)

Table 2.4 – Work function and properties for carbide-based ceramics.

	C	TaC	ZrC	TiC	HfC
φ (eV)	4.81	4.21 [130] 4.36 [123] 4.38 [132]	3.38 [131] 3.5 [129] 4.09 [133]	3.32 [131] 4.1 [129] 4.94 [123]	3.4 [131] 4.45 [123]
Resistivity ($\Omega \cdot m$) at 300 K	$6 \cdot 10^{-5}$	$4.1 \cdot 10^{-7}$	$4.3 \cdot 10^{-7}$	$2.2 \cdot 10^{-6}$	$1.1 \cdot 10^{-6}$
Tensile Strength, Ultimate (MPa)	-	96 - 291	90	258	-
Elastic Modulus (GPa)	4.8	285	406	449.5	352
Thermal Conductivity (W/m·K)	24	22	20.5	23	30
Density (g/cm ³)	2.27	14.5	6.73	4.93	12.67
Melting point (K)	3925	4150	3813	3430	4170

Carbide materials are nowadays a reliable alternative to the refractory metals but they need to be further studied. TaC cavities were tested, where the selectivity showed values of 10^4 at low temperatures (1600 K) and quickly decreased at high temperatures (selectivity of 30 at 2300 K) [95]. Recently, promising results with crystalline graphite were obtained where the selectivity could be increased by a factor of 30 - 50 for isotopes of mass 100 [126]. Graphite is often used in several facilities such as ISOLDE (CERN) [11]. The main problem is that the different allotropic species of C have different work functions [134, 135]. This limits the comparison between materials because a perfect characterization has to be performed before use.

As it can be observed in table 2.4, ZrC carbide has an average work function value of 3.65 eV and a high melting point of 3813 K. For that reason it can be a good candidate to decrease the production of contaminants at high temperatures.

2.4.2.3 Borides

Boride materials have a lower work function compared to the carbide-based or refractory metals (table 2.5). The work function seems to be independent of the temperature [136]. However boride-based materials have a difficult processing and they present worse properties in extreme conditions. Also some of them (like LaB_6) are chemically active at high temperatures and can be easily damaged by flux gases. Therefore, the highest working temperature is limited [131]. Also these ceramics become highly fragile when they are in contact with hot metal surfaces (e.g. for a GdB_6 cavity, the maximum working temperature is 1673 K) [111].

Table 2.5 – Work function and properties of boride-based ceramics.

	LaB_6	GdB_6	CaB_6	CeB_6	YB_4
φ (eV)	2.66 [136] 2.7 [123] 2.85 [116]	1.8 [116]	2.86 [136]	2.59 [136]	2.61 [131]
Elastic Modulus (GPa)	-	-	451	379	-
Thermal Conductivity (W/m·K)	-	-	39.3	33.9	-
Density (g/cm ³)	4.72	5.27	2.45	4.81	3.76
Melting point (K)	2480	2373	2508	2805	2570

Low work function materials were used at ISOLDE on-line runs to reduce non-specifically ionized isobaric contaminants. Tantalum foils combined with GdB_6 cavities were used to deliver 10^7 $^{140}\text{Nd}/\mu\text{C}$ and $5 \cdot 10^7$ $^{142}\text{Sn}/\mu\text{C}$. However, none information was given about the contamination reduction factor [127]. LaB_6 coating was tested and the selectivity ranged between 10^2 and 10^3 at 1473 K. In addition, the Ga ionization selectivity in the ISOLDE ion source with GdB_6 was also measured obtaining a result above 100 [109].

2.4.2.4 Oxides

Oxides materials have the lowest work function of the studied materials (table 2.6). The drawbacks are the complicated machining and the brittle transition at high temperatures. In addition, oxides generally have high chemical reactivity at high temperatures. ThO_2 has relatively low chemical reactivity and a high melting point of 3400 K, however the vapor pressure is very high and it is fragile. Finally they are generally insulators and the only solution is to fit them inside the ionizer cavity. However, the oxygen could migrate towards the hot-cavity material and degrade it during long duration functioning [137]. Some selectivity was reported for a quartz transfer line at 873 K [128], with no specification about the reduction. A test was performed with a quartz transfer line at 581 K providing 5 orders of magnitude reduction of ^{80}Rb . Moreover, ^{142}Cs contaminants were suppressed by 3 orders of magnitude [113].

Table 2.6 – Work function and properties for oxide-based ceramics.

	CaO	SrO	BaO	SiO ₂
φ (eV)	1.78 [131]	1.43 [131]	1.25 [131]	2.5 [138]
Density (g/cm ³)	3.34	4.7	5.72	2.6
Melting point (K)	2886	2804	2196	1986

2.4.2.5 Conclusion

The experiments performed by several laboratories are gathered in table 2.7. A selectivity enhancement was observed for refractory metal materials at temperatures lower than 1600 K (e.g. Ta, Nb). However, an increase of the temperature lead to a decrease of the selectivity.

Table 2.7 – Selectivity tests with several ion source materials.

Material	Average work function (eV)	Element	IP (eV)	Selectivity	Working temperature (K)	Reference
Ta	4.34	Tm	6.18	10 ⁴	<1600	[95]
		Tm	6.18	30	2300	[95]
Nb	4.22	Tm	6.18	10 ⁴	<1600	[95]
		Tm	6.18	30	2300	[95]
		Tl	6.11	10 - 20	2273	[128]
W	4.65	Tl	6.11	10-20	2273	[128]
		Ga	5.9993	<10	2273	[111]
				~10 - 20	2273	[109]
TaC	4.32	Tm	6.18	10 ⁴	<1600	[95]
		Tm	6.18	30	2300	[95]
<i>LaB</i> ₆	2.73	Ga	5.9993	10 ² - 10 ³	~1473	[111]
W(BaOSrO)		Ga	5.9993	10 ⁶	<1473	[111]
				> 1000	1373	[109]
<i>GdB</i> ₆	1.8			>10 ²	1673	[109]
Quartz	2.5	Li, K, Rb, Cs	5.39 - 3.89	~10 ³ - 10 ⁵	581	[113]
Crystalline C				30 - 50	-	[126]

TaC showed the same behavior as the refractory metals. Tungsten exhibited a low selectivity also at high temperatures but tungsten infiltrated W(BaOSrO) presented the highest selectivity (10⁶) at temperatures below 1200°C. However, this success was limited due to the material physical limitations at temperatures higher than 1473 K. Low work function materials displayed high selectivity (*LaB*₆, *GdB*₆, quartz) at low temperatures, and only *GdB*₆ presented high selectivity at 1400°C. As it can be observed, a decrease in the working temperature usually enhances the selectivity, but it decreases the ion production efficiency due to the decrease of the effusion efficiency and the increase of the sticking time.

More studies are needed to find a reliable material that can work at temperatures higher than 2000 K and simultaneously decrease the surface ion production. Due to its average work function value of 3.65 eV, zirconium carbide (ZrC) was chosen to be tested to reduce the surface ionization at high temperatures (section 5.5). In addition, it has a melting point at 3813 K, so it is a good candidate to work at 2000 K.

2.4.3 Oriented electric fields

Several attempts have been performed to minimize the contamination from the target by applying oriented electric fields [139]. The electric current used to heat the ion source by Joule effect, generates also an electric field from the voltage drop along the ionizer and atomizer.

This electric field can modify the direction of the surface ion motion and that of the laser ionized atoms [95, 140]. Investigations were performed at CERN with Ta hot-cavity with the reversal of the dc target heating current, where a suppression factor of 5 was measured for potassium, rubidium and cesium [114] (fig. 2.14). A study was performed in this work to determine the reduction of surface ions by oriented electric fields (section 5.5).

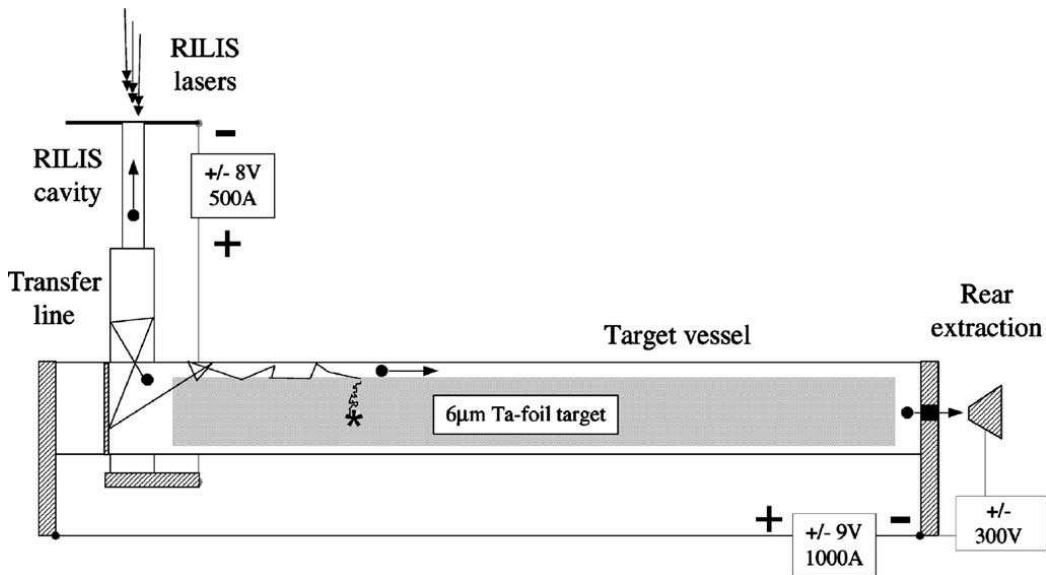


Figure 2.14– Scheme of the ISOLDE target Ta-174. It is equipped with a laser ion-source high temperature cavity, a target vessel that allows the reversal of the dc target heating current and a “rear” electrostatic extraction [114].

2.4.4 Laser Ion Source Trap

The main idea of the Laser Ion Source Trap (LIST) is to ionize the elements of interest in a RFQ trap, which is independent of the hot cavity where the surface ion contaminants are generated [141, 142]. The contaminant suppression is possible by installing a positively charged 10 V repeller electrode immediately in front of the atomizer cavity. Immediately behind, a radio-frequency quadrupole ion guide was set to trap the ions. Here is where the free-background neutrals are resonantly laser ionized. The RFQ field radially confines the ions generated by resonant laser photo-ionization [143]. The ions are then extracted by the ground extraction system (fig. 2.15).

This device was developed in a collaboration between CERN and Johannes-Gutenberg Universität Mainz [80]. TRIUMF TRILIS also developed a laser ion source trap [144], called Ion Guide Laser Ion Source (IG-LIS). By this method it is possible to reduce the amount of surface ionized isobaric contaminants by four orders of magnitude [145]. The first on-line test were performed at ISOLDE (CERN) [146] and at TRIUMF [4]. The LIST performance can suppress the contamination by $\sim 10^3 - 10^4$ orders of magnitude, but at the same time the efficiency decreases a factor $\sim 20 - 50$. This loss factor is due to the expansion of the atomic beam beyond the ionizer cavity [11].

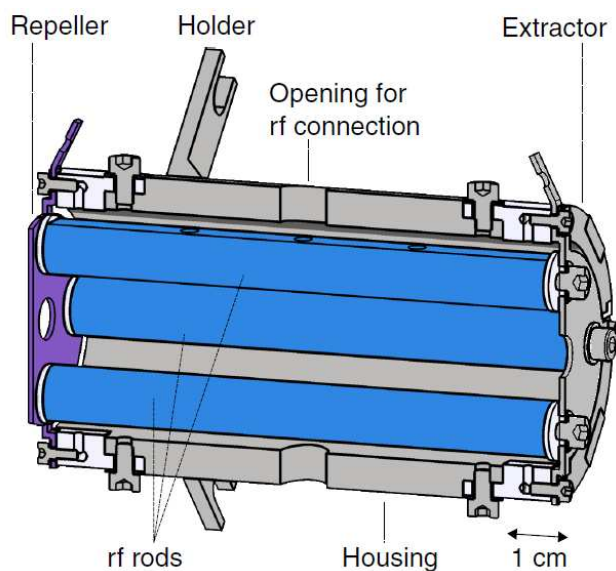


Figure 2.15– 3D-CAD sketch of the LIST device. A repeller electrode is mounted in the left side. The RFQ ion guide rods are shown in blue, and the housing is shown in grey [146].

The repeller electrode can operate on two different configurations. If a positive voltage is applied, the charged ions coming from the hot cavity are repelled and the contaminants cannot enter the RFQ ion guide (LIST mode). Alternatively, if a negative voltage is applied,

the positive ions can go through the electrode (ion guide mode). The efficiency in the ion guide mode is comparable to a normal RILIS operation without a LIST device. This enhanced laser ion source opens the opportunity of an improved resonance spectroscopy [146]. The main drawback is the diminution of the ionization efficiency. Anyway, the photo-ions are produced relatively free of any background and radially confined, which provides a low emittance ion beam [147].

2.5 Origins and evolution of the RILIS technique

In this section, an overview of the historic developments of the RILIS is given. These advancements led to the actual implementation of the RILIS technology. The rapid development of the laser technology and the early stage of the spectroscopy research produced a wide range of techniques for resonant ionization spectroscopy (RIS). Among them, multi-step resonant photo-ionization was first studied by Ambartzumian and Letokhov in 1971 [89] and then described in a textbook by Letokhov [148]. The resonant optical excitation and ionization was included as one of the techniques for spectroscopy resolution of atomic transitions. All this work was reviewed in detail by Samuel Hurst [149].

The resonant ionization spectroscopy appeared to be a versatile and powerful technique for the study of nuclear ground-state and isomeric properties. This technique also makes possible isomer selective ionization by precise laser wavelength tuning to a particular hyperfine structure component of the atomic transition line. These publications opened, at the same time, the possibility of using the resonant absorption to generate ion beams based on the element line information.

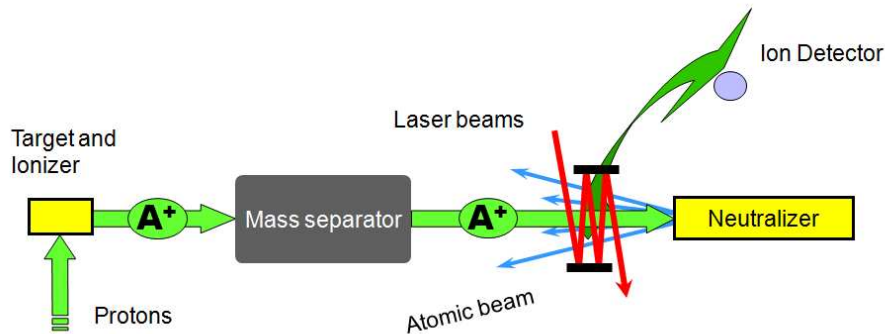


Figure 2.16– Picture that shows the setup for the spectroscopy experiment carried out by ISAN and LNPI in 1983 [150].

The very first experiments of laser resonant ionization spectroscopy of Radioactive Isotopes in Atomic Beams carried out at the Leningrad Nuclear Physics Institute (LNPI) in cooperation with the Institute of Spectroscopy of USSR Academy of Sciences (ISAN) in 1982 [151], at the ISOL facility at Gatchina (USSR). The main idea was to perform spectroscopy measurements in an atomic beam generated by evaporation of samples of radioactive isotopes from a tantalum crucible. The lasers were used to reach the ionization, and then the generated ion beam was extracted, detected and analyzed. One year later, the first on-line laser photo-ionization experiment was performed at the same facility by directing mass-separated ion beams of short-lived europium isotopes to the crucible followed by their neutralization, evaporation in the form of collimated atomic beam and laser resonance photo-ionization (fig. 2.16).

Throughout those experiments appeared the idea to use the photo-ionization method to generate radioactive-ion beams. The need of a confinement for a perfect overlapping between the laser light and the atoms was resolved confining the atom vapour in a pipe-shape cavity and facing the laser to the atom beam. The explanation of the availability of photo-ionization, and a compilation of the first ideas were published in 1985 by S.V. Andreev, V.I. Mishin, S.K. Sekatsky [152]. It was shown that a hot cavity resonant laser photo-ionization can provide ionization efficiencies above 50%, which compares very favorably with the typical 10^{-4} - 10^{-3} efficiencies of resonance ionization spectroscopy set-ups using unconfined atomic beams.

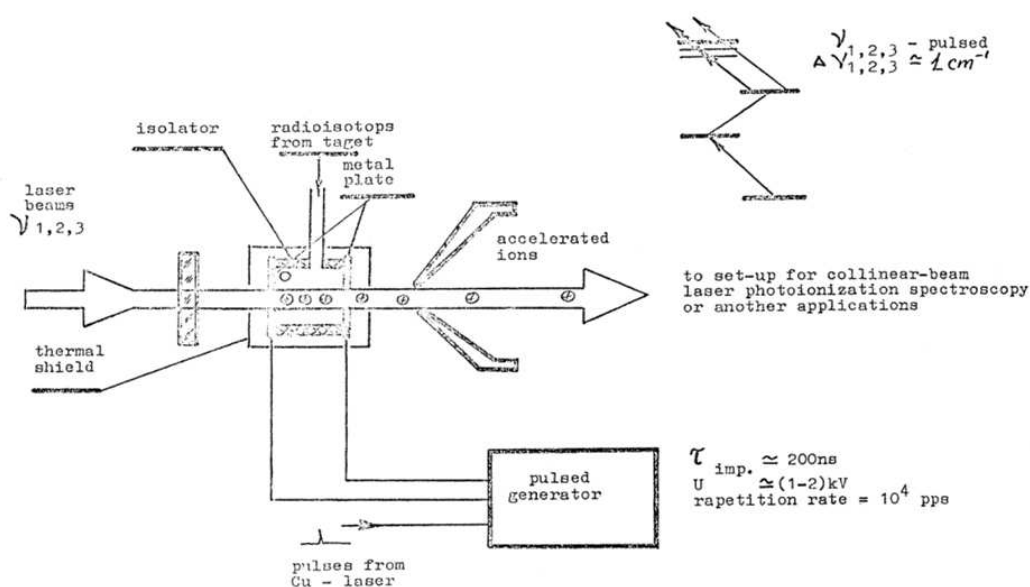


Figure 2.17– Explanation of the proposal of the experiment of laser photo-ionization pulsed source of radioactive atoms [126].

In 1984 there was a proposal of the Institute of Spectroscopy (Academy of Sciences of USSR) and ISOLDE (CERN, Switzerland) to start a campaign of experiments of laser photo-ionization pulsed ion source of radioactive atoms (fig. 2.17). The aim of the proposal was to create an isobar-selective ion source based on laser resonant atomic photo-ionization. In this case the laser system and the ion source were designed at the same time. Dye laser beams in the visible range were used to ionize different lanthanides.

The first concept of the hot-cavity for resonant ionization laser ion source was suggested by H.-J. Kluge, F. Ames, W. Ruster and K. Wallmeroth in 1985 [153]. The present hot cavity ion sources are coming from the tubular sources demonstrated in Gatchina [154] and CERN [95]. Since its first proposal by Kluge and collaborators, the resonant ionization laser ion source at ISOLDE (CERN, Switzerland) were further developed due to its unique selectivity and efficiency to obtain a high performance laser ion source [11]. In 1989 G.D. Alkhazov, V.S. Letokhov, V.I. Mishin, V.N. Panteleyev, V.I. Romanov, S.K. Sekatsky, V.N. Fedoseyev, demonstrated the high selective laser ion source method: High efficient Z-selective photo-ionization of atoms in a hot metal cavity followed by electrostatic confinement of the ions [155].

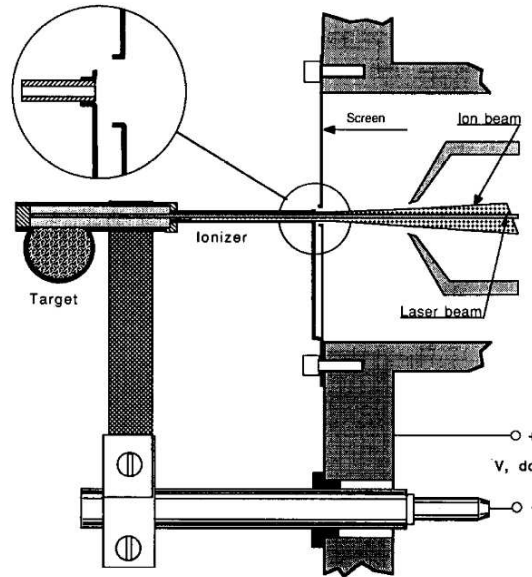


Figure 2.18– Schematic drawing of the target and hot cavity ionizer setup at the ISOLDE-3 mass separator [95].

During the year 1990 a laser ion source was developed for efficient production of isobarically pure ion beams at the on-line mass separator ISOLDE at CERN (Switzerland) [156] (fig. 2.18). Shortly before that, a similar facility development took place at the IRIS facility at the Petersburg Nuclear Physics Institute (PNPI) at Gatchina (Russia) [155, 154]. In 1992, a study of short-lived tin isotopes with resonant laser photo-ionization at the Heavy Ion Accelerator

UNILAC/GSI (Germany) was performed [157]. After all of these advances, the implementation of an on-line laser ion source at the new ISOLDE-PS Booster facility (CERN, Switzerland) started, due to its outstanding advantages in comparison to conventional ion sources. The first physics runs with the RILIS were carried out in 1994 [158]. Since 1993, ISOLDE is the main facility to generate radioactive beams produced by the RILIS technique at CERN.

The gas cell development started in 1992 at the Leuven Isotope Separator On-Line facility [100, 104, 25]. In 1998, Lettry et al. [140] measured the time profile distribution of the laser-ionized ion bunches and the efficiencies of several elements were presented. From 1999 to 2000, high intense radioactive-ion beams of nine different elements were produced at ISOLDE (CERN) [97] (fig. 2.19). In addition, a study of the duration of the ion packets at the output of the mass separator in relation to the voltage drop across the RILIS ionizer was presented in order to understand the physics involving the ion source [114, 159]. Furthermore, a systematic development of both off-line and on-line studies was achieved by the LISOL group of the University of Leuven [104, 160], who implemented a RILIS gas cell technology in an ion-guide type ion source.

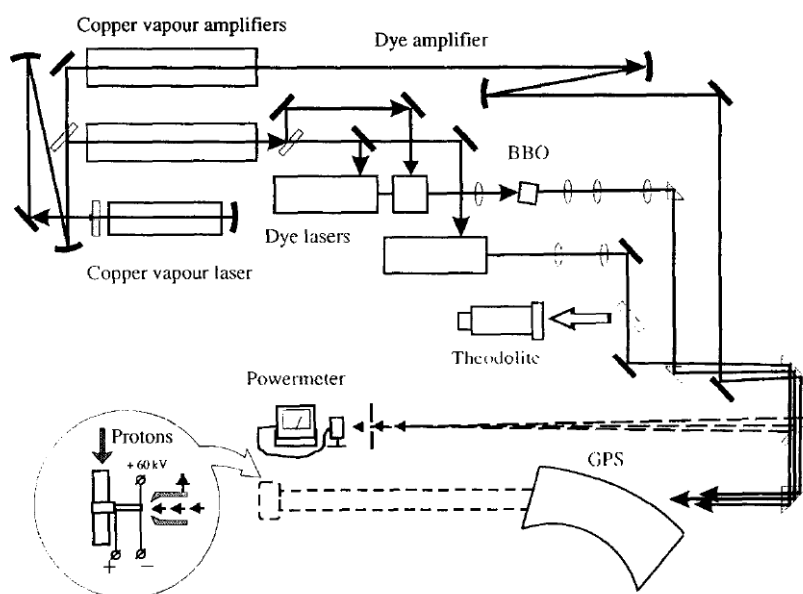


Figure 2.19– The former scheme of the laser ion source system at ISOLDE, which included a dye laser system (above) and an ionizer and mass selector (below) [161].

In 2000, the LARISSA group at the Johannes-Gutenberg Universität Mainz (Germany) started the development of a tunable all-solid-state laser system which competed with the dye laser technology used for RILIS in those days [162]. The LIST RFQ ion trap was first proposed in 2003 [80] as a combination of the advantages of a laser ion source with those of an ion trap. This device lead to perform a significant increase of the isobaric selectivity.

2.5. Origins and evolution of the RILIS technique

Based on the ti:sa laser designs of the LARISSA group of Mainz University a solid-state titanium:sapphire laser ion source was developed at the ISAC facility (TRIUMF, Canada) in 2002. During 2003, the first off-line demonstrations of ion beams were performed [163] and finally the first on-line radioactive-ion beam of Ga by a solid-state laser ion source was produced at TRIUMF (Vancouver, Canada) [2, 164, 165]. Throughout 2004, 24 elements have been generated at ISOLDE (CERN) with ionization efficiencies in the range from 0.5 to 15% [5]. The application of the resonant laser ionization technology was started in 2005 for the IGISOL mass separator facility in Jyväskylä (Finland) [166].

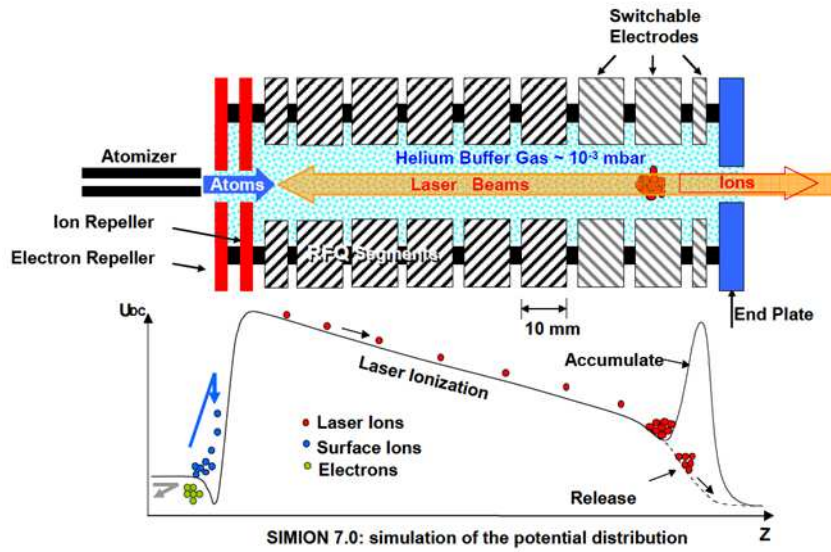


Figure 2.20– Schematic drawing of one of first LIST prototypes with a repeller electrode, a segmented linear RFQ ion trap, a gas inlet, a pulsed end-plate and an exit lens. The potential gradient along the symmetry axis is shown for both, the accumulation and release modes [80].

A fruitful campaign of spectroscopy research provided atomic element line information for many elements at Johannes-Gutenberg Universität Mainz (Germany) [167]. In the same year, the FURIOS laser ion source was completed at IGISOL (Jyväskylä, Sweden) and the first off-line laser ionisation tests were performed on Bi [101] and ^{99}Tc [168]. A hot-cavity LIS system with three ti:sa lasers was set up and operated at the off-line ion source test facility (ISTF-2) of the Holifield Radioactive-Ion Beam Facility (HRIBF) at Oak Ridge (ORNL, USA) [169]. In the same year, a design for a gas-cell prototype was being studied at ALTO facility at IPN-Orsay (France) [170]. On the other hand, an ion source combining a segmented linear radiofrequency quadrupole (RFQ) to a laser ion source was being built at TRIUMF [144].

Since 2007, the RILIS performance and capabilities have been gradually improved through a multi-stage upgrade of the laser system. Most of the facilities using dye lasers replaced the copper vapor pump lasers by Nd:YAG pump lasers and often with a complementary ti:sa laser

system [171, 1]. Other developments for the ion-guide technique appeared [172]. In addition a new LAser Resonance Ionization Spectroscopy (LARIS) laboratory was constructed at ISOLDE (CERN) for off-line studies. ^{26}Al beam was produced by a solid-state laser ion source at TRIUMF [173]. In 2009 a laser ion source development was started at GANIL (France) based on the ti:sa laser technology of University of Mainz and TRIUMF [174]. In addition, the design of a grating-tuned ti:sa laser for spectroscopy applications was presented at TRIUMF [82] and a novel development for time structure measurements of ion beams was performed at Oak Ridge (ORNL) [77].

Since 2010 the number of the accessible elements increased year by year. Spectroscopy measurements were performed [175, 8] and ionization schemes were found for many elements [176]. New applications were found, such as ultra-trace determination [9], and several technological advancements were carried out. The latter include the improvement of the reference cell [177], the narrow linewidth operation of the RILIS titanium:sapphire [178] and the evolution of the ion-gas-jet technique in the gas cell [179, 180] and first off-line tests of the KISS gas cell at RIKEN [181, 182]. The first applications of LIST experiments were carried out [146] (fig. 2.20). In addition, the first results of the off-line RILIS test bench at GANIL were presented [183]. The production of astatine ions and the measurement of the first ionization potential of astatine were finally achieved [178, 184, 185].

Today more than 600 isotopes are accessible by the RILIS technique. Figure 2.21 shows the current implementation of the hot-cavity technique around the world.

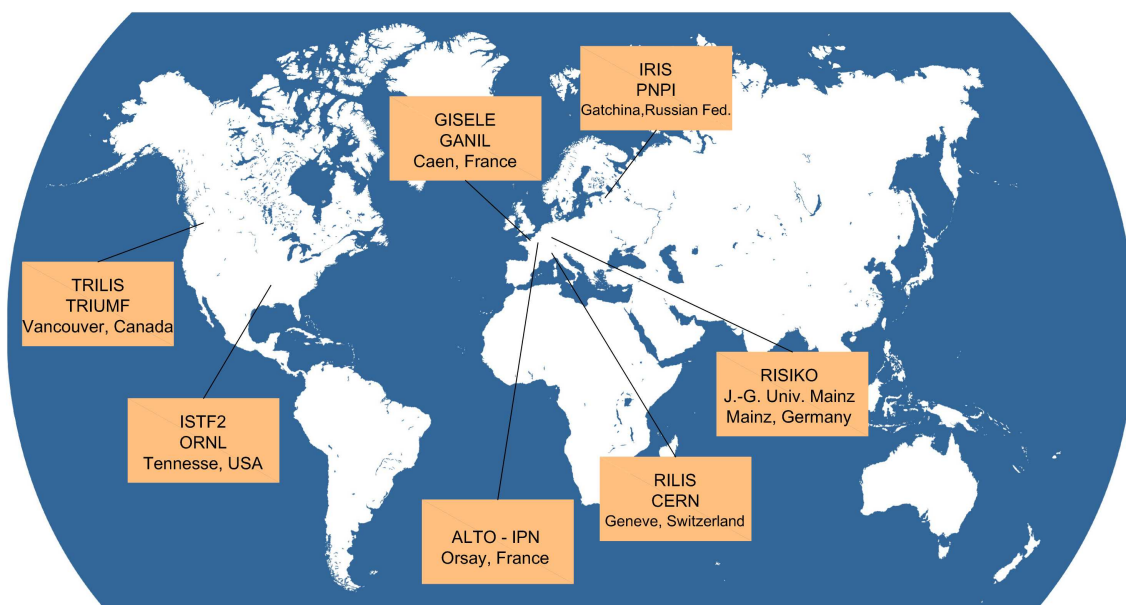


Figure 2.21– Existing resonant laser ion sources using hot-cavity devices at radioactive-ion beam facilities.

Chapter 3

GISELE laser system

3.1 Off-line test bench

GISELE (GANIL Ion Source using Electron Laser Excitation) is an off-line test bench to develop, study and eventually implement RILIS for the production of on-line accelerated RIBs in the future ISOL facility at GANIL [174]. The experimental setup (fig. 3.1) consists of a laser laboratory (section 3) and an ion source (section 4).

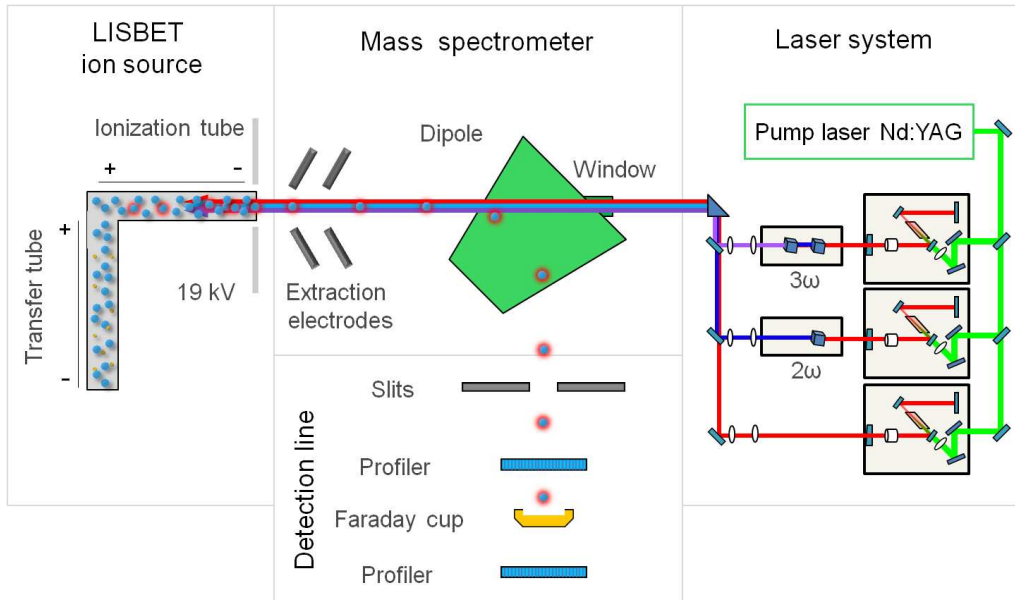


Figure 3.1– GISELE laser system and the off-line test bench for the implementation and development of RILIS at GANIL. The laser system, the ion source, mass spectrometer and detection line sections are indicated.

On the one hand, the laser system includes the laser cavity resonators, the laser transport optics and the laser beam diagnosis. On the other hand, the ion source consists of the interaction device, ion extraction optics, mass selection and ion detection beam. The data acquisition is controlled via a LabView program in the laser room and another at the test bench. It is possible to record the ion current, mass scan, beam profile shape and emittance. In the laser room, the laser light radiation is monitored by wave-meters, a master clock for laser synchronization and powermeters to measure the laser power output. In addition, live ion current measurement is integrated for optimization.

3.2 Laser system setup

The laser system consists of three pulsed tunable solid-state titanium:sapphire (ti:sa) laser resonators, pumped by a frequency doubled Nd:YAG laser (fig. 3.2). This complementary configuration allows two or three resonant photo-ionization process in a hot cavity by tuning each laser to the correct energy transition [186]. This system is actually used worldwide at on-line isotope separator facilities for the production of pure ion beams of shorted lived isotopes.

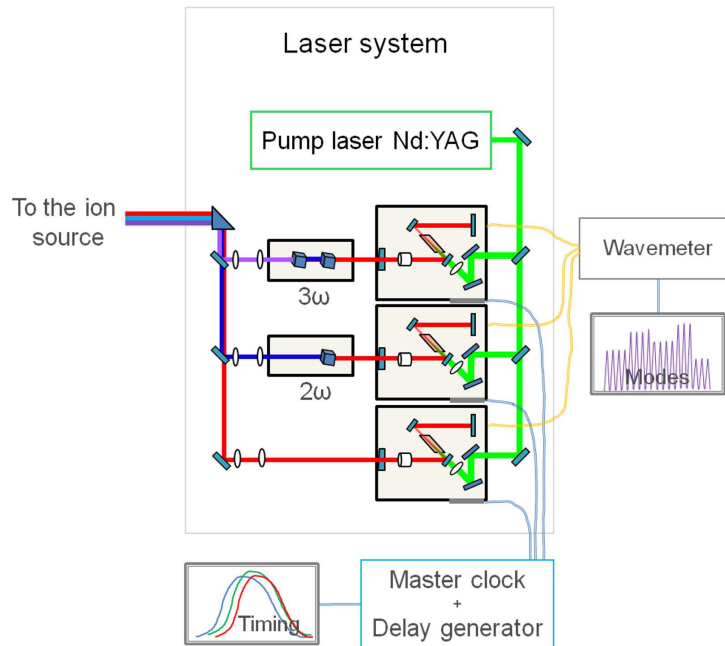


Figure 3.2– GISELE laser system setup. Three tunable titanium:sapphire lasers pumped by a frequency doubled Nd:YAG laser produce fundamental IR laser light. The fundamental laser radiation can be frequency mixed to access higher energy wavelengths. The laser cavities are monitored by a wave-meter and the delay is controlled by a master clock driving intracavity Pockels cells.

3.2.1 Nd:YAG pump laser

A Photonix Q-switched Nd:YAG laser is used as pump laser. The fundamental laser radiation (1064 nm) is intra-cavity doubled to generate the 532 nm needed for the ti:sa crystal absorption. The pump laser can provide an output power up to 70 W (7 mJ per pulse) at 10 kHz repetition rate with 120 ns of pulse width and $M^2=32.9$. This energy is enough to pump several laser cavities at the same time. A 10 kHz repetition rate laser system was used to minimize any possible duty cycle losses as explained in section 2.2.1. The laser pump power is measured before and after every experiment by a Coherent FieldMax II power meter (100 W maximum power) to control the long term power stability.

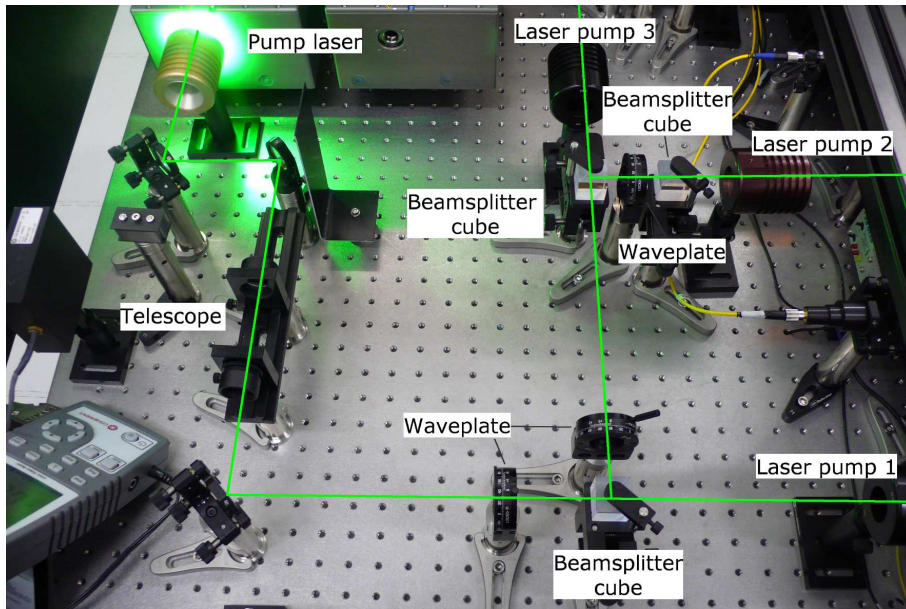


Figure 3.3– Pump laser transport towards the laser cavities. The Galileo telescope, half-wave plates and beam-splitter cubes are indicated.

The pump laser is expanded by a Galileo telescope composed by a f-75 (divergent lens with focal 75 mm) and a f150 (convergent lens with 150 mm). The telescope allows to control the size of the pump laser beam. Then, the pump laser is divided into several secondary laser beams, to independently control the power that goes to each laser cavity without modifying the general pump power (fig. 3.3). This is performed by half-wave plate ($\lambda/2$) in combination with beam-splitter cubes. The functioning of this system works as follows: the half-wave plate sets the incoming laser beam to a certain polarization, then if the laser beam is parallel polarized (p-polarized) the beam-splitter cube let the laser light go through it straight forward, and if the laser beam is perpendicular polarized (s-polarized), it will be reflected 45° as if a mirror were used. So, in an intermediate polarization between s-polarization and p-polarization, the

pump lasers is divided in two beams in a certain ratio that depends on the half-wave plate angle. This way it is possible to control the amount of pump laser power that goes to each cavity laser from a single pump laser.

3.2.2 Ti:sapphire lasers

The laser system was conceived at the Johannes Gutenberg-Universität Mainz (Germany) [162] the particular lay-out was developed and built at TRIUMF (Vancouver, Canada) [2]. The resonator consist of a z-cavity with a solid state gain material (ti:sa), two 0.5 inches diameter curved mirrors, a front-end mirror (high reflective) and a output coupler (fig. 3.4). The gain material is a 20 mm long ti:sa crystal rod with 6.35 mm diameter. It is cut at Brewsters angle to reduce the reflection losses in the surface. Nevertheless, the angle of incidence produces astigmatism and the resulting beam has an elliptical profile. To compensate this effect the 0.5 inch curved mirrors are set at angle of 18.1° referenced to the laser beam axis.

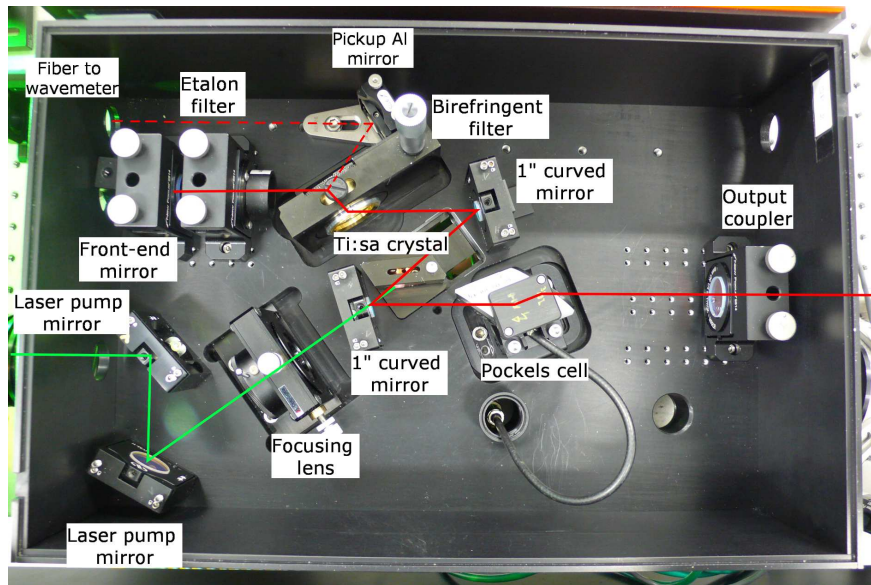


Figure 3.4– Ti:sapphire resonator with all of the opto-mechanics parts developed by TRILIS-TRIUMF [2]. The pump power enters to the cavity and it is focused into the ti:sa crystal. The amplification is provided by the resonator optics.

The pump laser is focused to the ti:sa crystal (generally in the back face to allow a higher laser pump without damaging the crystal) with a f88.9 convergent lens. The ti:sa lasers can develop a fundamental output power of about 3 W at 10 kHz repetition rate and 40 ns pulse width with an input pump power between 8 to 15 W. The output power was measured with a Coherent LabMax laser power meter (10 W maximum power). A detailed description of the laser system can be found in [167].

The laser wavelength selection is carried out by the characteristics of the resonator optics and the tuning elements inside the resonator. First, the mirror-set used in the resonator mirrors are coated to have high reflectivity ($\sim 99.9\%$) in different wavelength ranges and they can modulate the wavelengths that can be accessed. There are 4 mirror-sets for the curved mirrors and front-end mirror. In the case of the output coupler the 4 mentioned mirror-sets are available, but there is another mirror-set which can be used with the grating laser to access a broadband series of wavelengths. The mirror-set provided by Layertec are coated in the following ranges: SP-1 (655-755 nm), SP-2 (750-850 nm), SP-3 (840-940 nm), SP-4 (900-1050 nm) and an special broadband output coupler mirror SP-0 (650-1050 nm). The SP-0 mirror-set (also named as set-V) can be used with the other configurations to modify the wavelengths that can be produced.

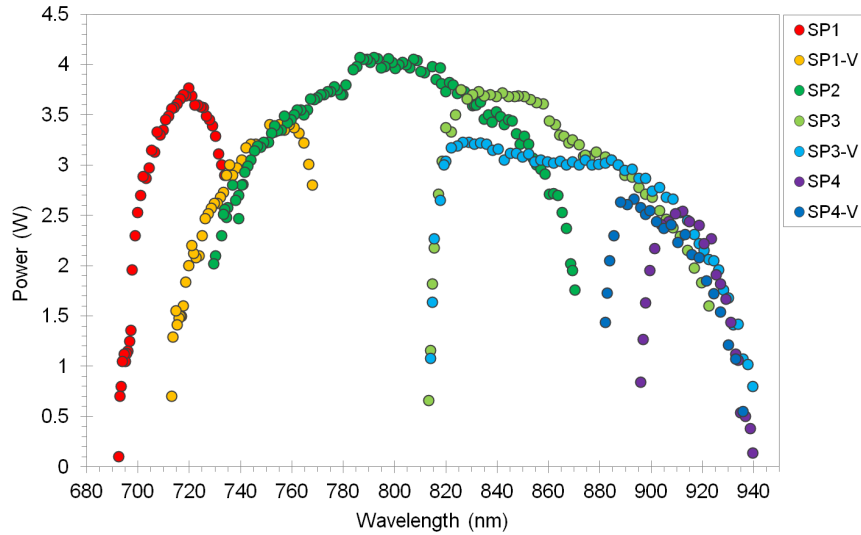


Figure 3.5– Laser wavelength selection for different mirror-set. SP-1: 655-755 nm, SP-2: 750-850 nm, SP-3: 840-940 nm, SP-4: 900-1050 nm and wavelengths produced by the use of the SP-0 output coupler (set-V): 650-1050 nm.

In addition, a Lyot birefringent filter (BRF) in combination with a thin etalon (0.2 or 0.3 mm thickness) can generate a spectral linewidth of about 5 GHz [52] as explained in section 2.2.1. The spectral bandwidth ensures the matching to the Doppler broadening of the optical transitions inside the hot atomizer cavity, maximizing the efficiency of the in-source ionization. In the case of the grating tuned laser, a gold coated blazed grating placed instead of the end mirror in Littrow configuration, replaces the etalon and the birefringent filter for wavelength selection. The grating tuned laser can reach 7 GHz of laser linewidth [82]. The output power has a high dependence in the chosen wavelength and the cavity mirror set. The titanium:sapphire laser can provide a wavelength emission range from 700 nm to 950 nm. The [fig. 3.5](#) shows the output power as a function of the wavelengths provided by each mirror-set, including those using the broadband SP-0 (set-V) mirror-set.

The laser wavelengths are monitored by a precision commercial laser wave-meter High Finesse Ångström WS6L. A reflexion from the etalon is sent to the fiber by a 0.5" Al mirror and the fiber is connected to the wave-meter. The wave-meter provides information on the spectral bandwidth and the wavelength. The laser mode affects the frequency mixing efficiency. To ensure a proper Gaussian mode at the exit of the cavity, a visual inspection with a divergent lens was performed, optimizing the resonator with the output coupler mirror.

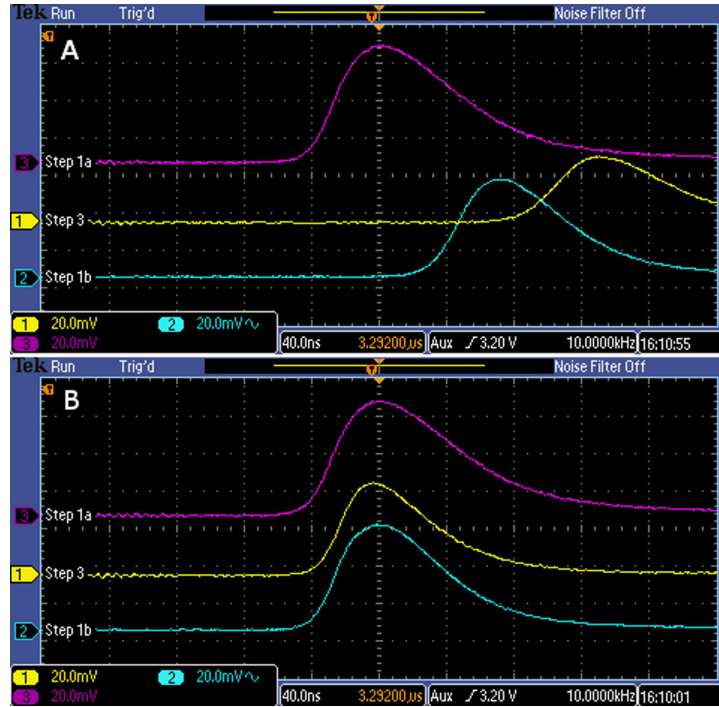


Figure 3.6– Pulse synchronization by Pockels cell. (A) De-synchronized lasers (Pockels cell off) and (B) synchronized lasers (Pockels cell on).

For an efficient resonant excitation and ionization laser pulses have to arrive synchronized to the ion source, however the laser cavities fire at different times producing laser desynchronization. The timing of a ti:sa can be modified by the laser pump power, where increasing the pump power increases the gain on the ti:sa crystal consequently firing earlier, and vice versa. If necessary, each ti:sa laser can be actively Q-switched with an intra-cavity Pockels cell and HV Behlke switch for temporal fine synchronization, permitting an temporal overlap better than 5 ns. In the fig. 3.6, the difference between Pockels cell synchronization and without synchronization can be observed. The timing of the laser pulse is characterized by a reflection from the etalon sent to a photodiode via a optic fiber. The delay time is driven by a generator master clock. and it is monitored on an oscilloscope.

3.2.3 High harmonic frequency generation

The limitation of the fundamental titanium:sapphire frequency radiation for resonant photo-excitation is the inaccessibility of high energetic transitions. Almost all elements have strong atomic transitions of 2.5 - 6 eV that need blue or UV laser radiation for excitation. To extend the fundamental wavelength range, high harmonic generation (HHG) can be produced using non-linear laser light interactions with optical crystal (e.g. β -barium borate). By this method the frequency of the ti:sa laser light can be doubled (second harmonic), tripled (third harmonic), and quadrupled (fourth harmonic) to expand the accessible wavelength range.

Higher harmonics are generated in two frequency conversion units (fig. 3.7) designed and manufactured at the Johannes Gutenberg-Universität Mainz (Germany) [88]. The wavelengths accessible with these ti:sa laser are approximately 700 to 950 nm for the fundamental radiation (ω), 350 to 475 nm for Second Harmonic Generation (2ω), 233 to 317 nm for Third Harmonic Generation (3ω), and 210 to 237 nm for Fourth Harmonic Generation (4ω). Wavelengths lower than 210 nm suffer of ultraviolet (UV) absorption on air propagation.

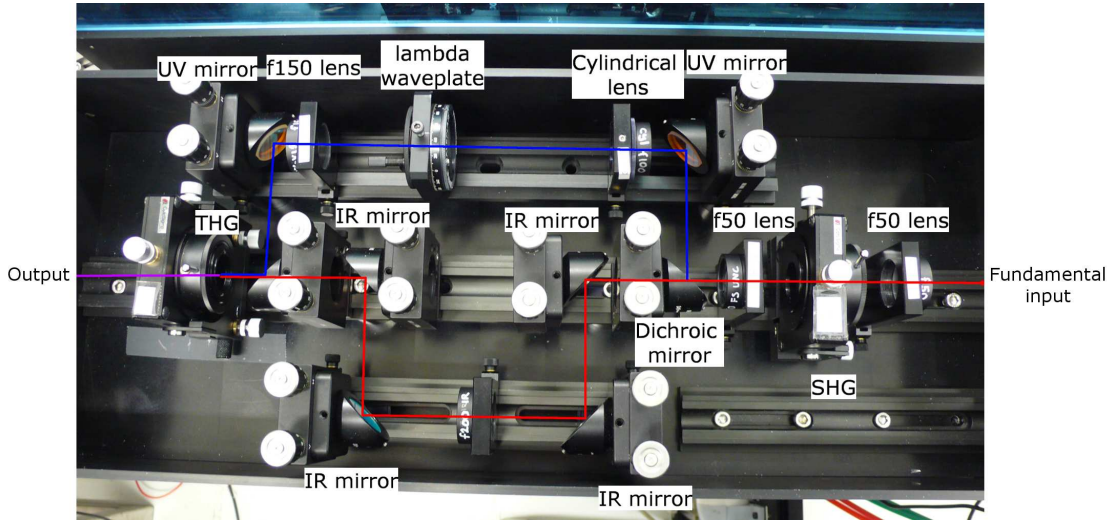


Figure 3.7– Frequency triple conversion unit developed at Johannes Gutenberg-Universität Mainz (Germany) and CERN [88]. The fundamental laser radiation arrives from the right part, then it is frequency doubled with a BBO crystal. Afterwards the doubled frequency laser beam and the remaining IR laser are split in two paths by a dichroic mirror and further recombined to generate tripled frequency light in another BBO crystal.

The second harmonic is generated by the use of a BBO non-linear crystal. The laser light is focused into the crystal, then parameters as crystal angles, crystal position and laser focus position are optimized, until higher harmonics are produced. The second harmonic is generated with a typical efficiency value of 25% input laser power. It is possible to perform intra-cavity doubling placing the non-linear crystal directly inside the resonator.

In the case of third harmonic generation, the IR laser light is doubled as explained before, and then the second harmonic and the remaining IR laser light are recombined again in another non-linear crystal to produce tripled frequency light. Third harmonic provides an efficiency of 10% of the input power. The quadrupling is just arranged by adding a second doubling stage for the second harmonics beam. Fourth harmonic generation has an efficiency of about 2% of the fundamental IR power.

The wavelength produced by high harmonic generation is not directly measured, it is calculated from the fundamental laser measured by the wave-meter and dividing afterwards the wavelength by 2, 3 or 4 in the case of frequency doubling, tripling, or quadrupling respectively. The typical output powers are about 1 W for second harmonic, 250 mW for third harmonic and ~ 50 mW for fourth harmonic. The output powers were measured after two dichroic mirrors.

3.2.4 Laser beam transport to the ion source

In order to maximize the efficiency of the ionization process, all laser beams must be overlapped in space and time inside the ion source. The laser beams have to be transported through a total distance of 20 meters to reach the ionization chamber (fig. 3.8). Each wavelength region (i.e. IR, second harmonic, third harmonic and fourth harmonic) has an independent mirror path with narrow-band optics adapted to the required wavelength. For a clear line-of-sight to the ion source all apertures and mirrors can accommodate multiple laser beams.

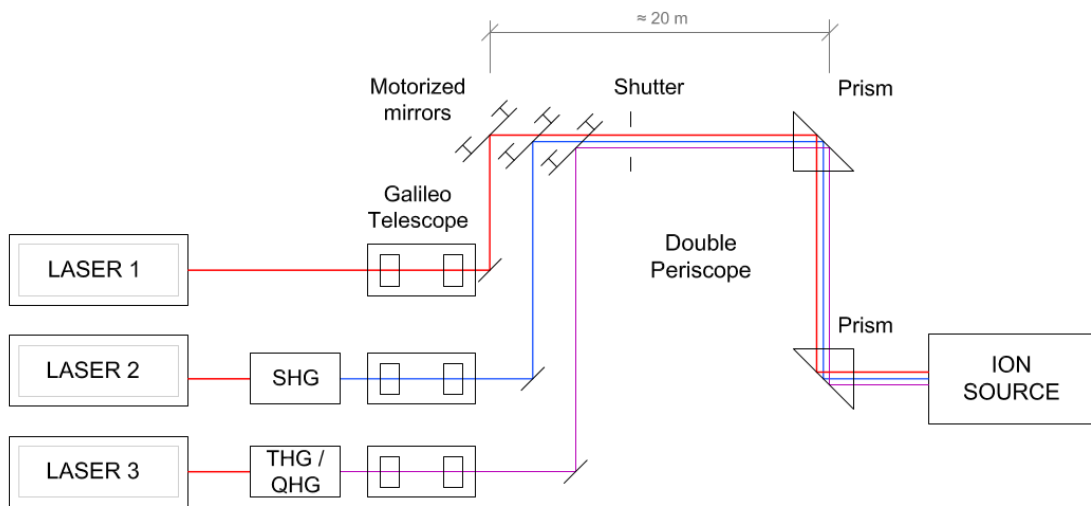


Figure 3.8– Laser transport from the laser cavity towards the ion source: Galileo telescope, double periscope, motorized mirrors and prisms.

3.2. Laser system setup

The transport consisted of a Galileo telescope to control the laser beam focalization in the interaction point (fig. 3.9). The laser beams were expanded by a factor of 4 by a divergent lens (focal -50 or -75 mm) and then focused by a convergent lens (focal 200 mm). This system allows to control the focus of the laser beams (modifying the distance between the lenses) in a distance of approximately 20 m to obtain the maximum ion signal. Due to the long path towards the ion source, a larger beam diameter will reduce diffraction limited spot size in the source.

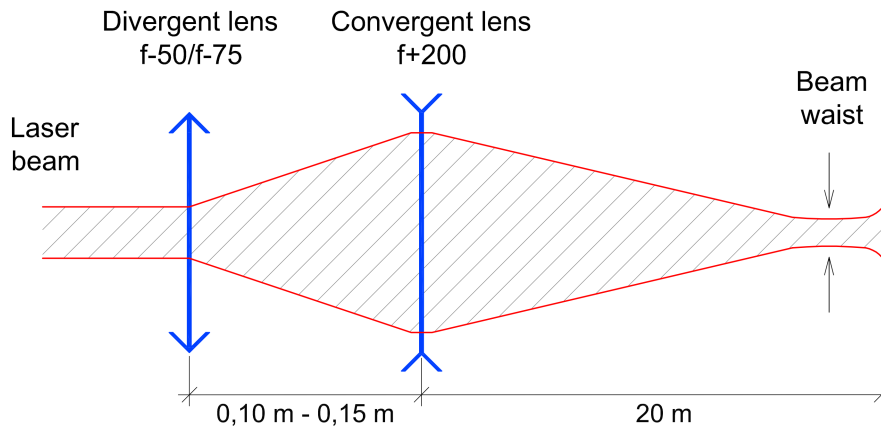


Figure 3.9– Diagram of Galileo telescope. The laser beams were expanded by a divergent lens and then focused by a convergent lens in order to control the laser spot size in the ion source at ~ 20 m.

Afterwards a pair of periscopes were used to transport the laser beams at 2.3 m height towards the experimental area. This system ensures the best laser safety conditions because none laser beam can be crossed or directly observed, and at the same time allows the transport of all laser beams in a long distance. First, each laser beam was sent up by a periscope (created by a pair of mirrors) to a 2.3 m height. Then laser beams are transported through an enclosure tube to the test bench area, and they are sent down by another periscope (in this case, a pair of prisms). Finally, the laser beams are coupled into the ion source chamber through a window in the back of the dipole and through the extraction electrodes. In addition, a mechanical shutter was placed at the exit of the laser room (inside the enclosure tube) to secure the delivery of the laser beams to the experimental area.

The laser beams are overlapped by cube beam-splitters (when the wavelengths are close enough) or by dichroic mirrors at the top of the periscope. Some of the mirrors are motorized, due to impractical access and security conditions, to control the spatial overlap between the laser pointing and the ion source, allowing a real-live optimization on ion beam signal intensity. Narrower-band optics (e.g. dielectric mirrors) were used for beam transport to avoid Fresnel reflection losses. However the transport losses, measured with the ion source chamber open,

were $\sim 50\%$ for IR laser beam and between 60% to 70% for doubled and tripled frequency light. Most of the losses come from the non-coated two prisms and the quartz window in the dipole mass spectrometer.

3.2.5 Laser beam diagnosis

A reference point was used in the laser room in order to control the correct overlap and spot size of the laser beams inside the ion source (fig. 3.10). A small fraction of the laser beams is picked before the shutter, and an alternative laser path was mounted inside the laser room to simulate the total length the lasers have to travel. At the end of this laser path the laser beams have the approximate same size as in the ion source. A screen was placed to visualize each laser spot and optimize the overlap of each laser position.

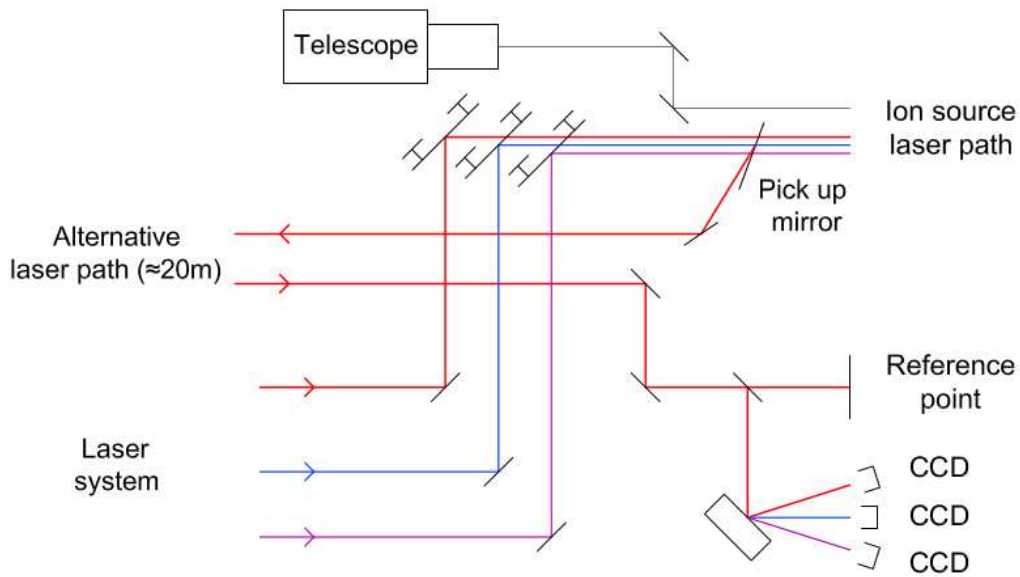


Figure 3.10– The reference point is installed in the laser room. A fraction of each laser beam is picked and sent through an alternative path to visualize the size and relative position of the laser beams, or alternatively send them to a diffraction grating and 3 CCD cameras (placed in different diffraction orders).

Furthermore, a diffraction grating was added to split the different wavelength components (i.e. each laser beam) in the different reflection orders which were recorded by three CCD cameras (one per laser beam reflection order). By this way it is possible to control and optimize the size of each laser beam. In the case of two close wavelengths (selected in the same reflection order) just blocking one of the laser beams will be enough to optimize the other.

In addition, an Optical Vision Ltd Sky-watcher telescope was included, facing the first periscope prism. With this device it is possible to look into the ion source (through the dipole window and the extraction optics) and to center the IR laser light (fig. 3.11). A CCD camera was set in the telescope to record the signal in a computer. The combination of these three systems (visual reference point, CCD cameras and telescope) assure the correct alignment and overlap of the laser beams into the ion source. The fine optimization of the relative positions and size is performed analyzing the ion beam signal enhancement.

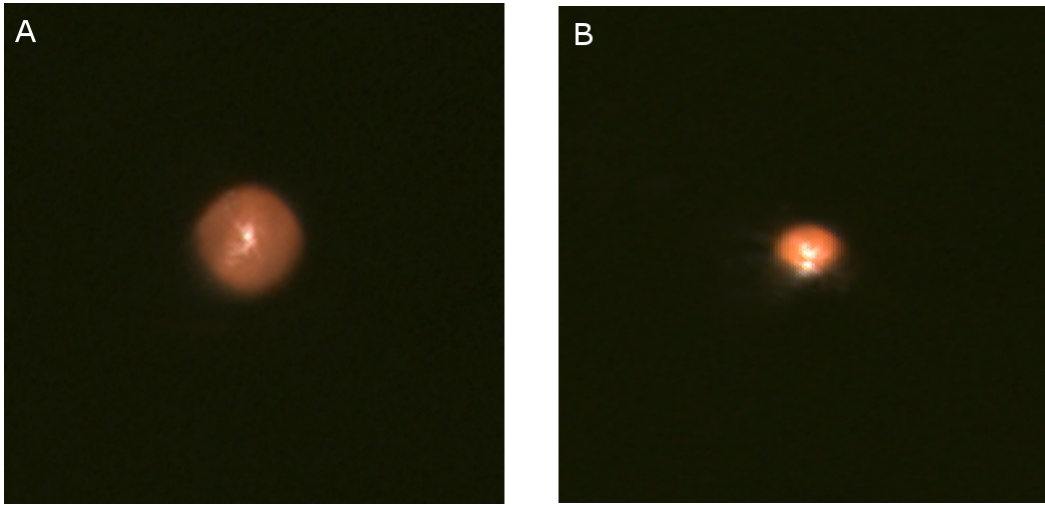


Figure 3.11– Images recorded by the CCD camera connected to the telescope for two ionization tube configurations: (A) 7 mm diameter and (B) 3 mm diameter. The red circle is the IR radiation generated by the ionization tube at medium temperature (~ 1100 K), and the white spot correspond to the IR laser reflection.

3.2.6 Laser monitoring and control system

A LabView program was integrated in the laser room to control the parameters of the laser system (fig. 3.12). The wavelengths are read by the wave-meter together with the ion beam signal (either from the ion source or from the reference cell). This allows to perform scans over different wavelengths to study atomic transitions. In addition, it is possible to optimize the laser beam parameters (wavelength, timing and laser spot overlap) by the live variation of the ion signal intensity. Finally, the ion source parameters (high voltage, current applied to the interaction device, etc.) are shown, but they can only be modified by a similar LabView program in the test bench command control adapted for ion source acquisition.



Figure 3.12– Command control front end with direct access to information concerning the laser system in the laser laboratory.

3.3 Reference cell

A reference cell provided by the LARISSA group from the Johannes Gutenberg-Universität Mainz (Germany) was installed in the laser room for spectroscopy measurements 3.13. The sample of the element of study is placed in an oven which is indirectly heated to provide an atomic flux. Then laser beams and atoms interact and the ions are drifted by several electrodes to a Secondary Electron Multiplier (SEM). The lasers ionize the atoms entering through a lateral window and crossing the atom flux.

The reference cell was optimized to record even small ion beams. By this compact system, it is possible to perform spectroscopy research through the scan of atomic transitions (by tuning the laser wavelengths) to search for non-known atomic resonances. The maximum working temperature is limited to 1770 K and the pressure for measurements is 10^{-7} mbar. The repeller and lenses can generate 4 to 30 V (positive), the SEM electrode 100 to 500 V (negative) and the SEM detector 1300 to 1650 V.

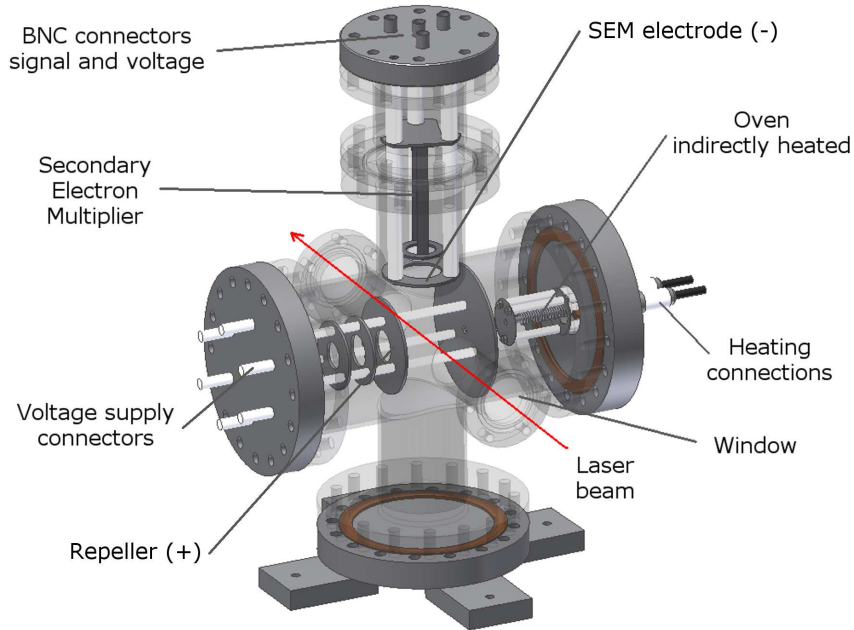


Figure 3.13– Reference cell diagram from the Johannes Gutenberg-Universität Mainz. A sample of interest is placed in the oven. Then it is heated by resistive heating to produce an atomic flux. The laser beams interact with the atomic flux producing an ion beam. The ion beam can be detected by a Secondary Electron Multiplier (SEM).

3.4 Resonant photo-excitation and ionization schemes

Since each element has its own unique energy level structure, specific atomic transitions have to be found for each atomic element [187]. Due to this, detailed laser spectroscopy information on atomic spectra and excitation schemes is required to access the elements of interest and drive the ionization process efficiently [5].

3.4.1 Ionization schemes research

Finding a new scheme includes database research, bibliographical research and experimental tests of a broad spectra of possibilities. An efficient ionization scheme should fulfill the next requirements: The atomic transitions have to be strong enough to provide highest population of excited levels, the atomic transition scheme has to follow the quantum mechanics selection rules, and the frequency of the transitions have to be reached by the ti:sa laser radiation (including high harmonics).

The first point is to identify the transition lines for the element. Kurucz database [75] provides extensive information of transitions but not all transitions are included. On the other hand, NIST database [74] has all lines but less specific information.

Using these databases, an energy level diagram can be drawn to have a first idea of the global scenario. The Ionization Potential (IP) represents the total energy needed to reach ionization. The IP can be analyzed to conclude if it can be reached by photons generated by a combination of three ti:sa lasers. Then, it is important to identify the transitions that can be reached by the ti:sa wavelength range (harmonics included) to a First Excited State (FES) and to calculate the thermal populations of low-lying atomic levels given by the Boltzmann distribution of each transition. Boltzmann distribution describes the population of levels at thermal distribution for given T [188]:

$$\frac{N_j}{N_i} = \frac{g_j}{g_i} \exp\left(\frac{-(E_j - E_i)}{k_B T}\right) \quad (3.1)$$

where N_j and N_i refer to the population of upper and lower levels respectively, E_j and E_i the energies of upper and lower levels respectively, k_B is the Boltzmann's constant, T the temperature [K] and g_j and g_i are the statistical weights of the levels calculated by the total angular momentum quantum number of the term (J):

$$g_j = (2J_j + 1) \quad (3.2)$$

For example, at 2000 K the population ratios of germanium (Ge) ground states: $4s^2 4p^2 {}^3P_0$ ($g_1 = 1$, $E_1 = 0 \text{ cm}^{-1}$), $4s^2 4p^2 {}^3P_1$ ($g_2 = 3$, $E_2 = 557.1341 \text{ cm}^{-1}$) and $4s^2 4p^2 {}^3P_2$ ($g_3 = 5$, $E_3 = 1409.9609 \text{ cm}^{-1}$) are respectively 21%, 42% and 38%. In thermal equilibrium, the lower energy level is more populated than the other two energy levels (this is the normal state of the system), but at 2000 K the $4s^2 4p^2 {}^3P_1$ level is more populated than the others. As a consequence, $4s^2 4p^2 {}^3P_1$ level will be a better starting state for an ionization scheme using a hot-cavity laser ion source.

In addition, the A-value is the transition probability (Einstein's A coefficient). It gives an idea of the efficiency of the transition, where 10^8 s^{-1} is a strong transition, 10^6 s^{-1} is an average transition and 10^4 s^{-1} is a weak transition. However the A-values for most of the transition lines are not known. Finally, only transitions with wavelengths corresponding to the ti:sa range can be reached. As a conclusion, to guarantee an efficient transition to the First Excited State the population of starting level has to be considered and the A-value as well as the optical transition, rejecting transitions with wavelengths outside of the ti:sa range.

For Second Excited States (SES) literature information on A-values or relative transition intensities has to be analyzed, in order to find an efficient transition. If not all levels are known, an experimental research for new transitions has to be carried out. In addition, it is important to know that the transition has to fulfill the following selection rules:

- The parity rule has to be kept to allow the transition: from «even» level to «odd» level or viceversa.
- J changes its value $-1/0/+1$ except for $0 \rightarrow 0$.

For the ionization step, first option is to compare the energy difference between the SES and the Ionization Potential. If it is <1.77 eV (photon energy for 700 nm light wavelength) a non-resonant ionization may be possible with infrared (IR) photons, efficiently generated by the Ti:Sa lasers. However, resonant transitions (i.e. Auto-Ionization States or Rydberg states) will provide the best ionization efficiency. Auto-Ionization States (AIS) above the ionization potential can be reached only if there is a wavelength in the Ti:Sa range and if the transition fulfill the selection rules.

3.4.2 Resonance transition saturation

Usually, three laser beams are used to ionize the atom of interest by means of its atomic transition resonant excitation. In order to measure the efficiency of the transitions, a resonance saturation measurement of each step is performed. It consists of the measurement of the ion intensity as a function of the laser power, in order to judge if the atomic transition is saturated enough. The laser power of one laser cavity is decreased while the others are maintained at full power. If transitions are saturated, it means the transition maximum population has been reached and the ionization process is carried at its maximum efficiency for the selected ionization scheme (fig. 3.14).

To analyze the saturation curve, the laser output power is measured on the laser table before the telescope expansion. The laser output power is decreased by the use of a half-wave plate ($\lambda/2$) and an optic polarizer. Half-wave plates turn the polarization direction of the laser beam, and then the polarized laser beams are sent to the optic polarizer. An optic polarizer only allows the transmission of the polarized light in one direction (maximum polarization) while proportionally absorbs/reflects the power for other polarization directions, reaching a minimum transmission at the perpendicular position of the maximum polarization. By this method, it is possible to scan different laser power intensities to measure the resonance transition saturation. The laser powers are measured directly after the resonator cavity for fundamental wavelength and after two dichroic mirrors for high harmonic wavelength. However the real laser power in the ion source is decreased due to the losses in the transport optics (section 3.2.4). For this

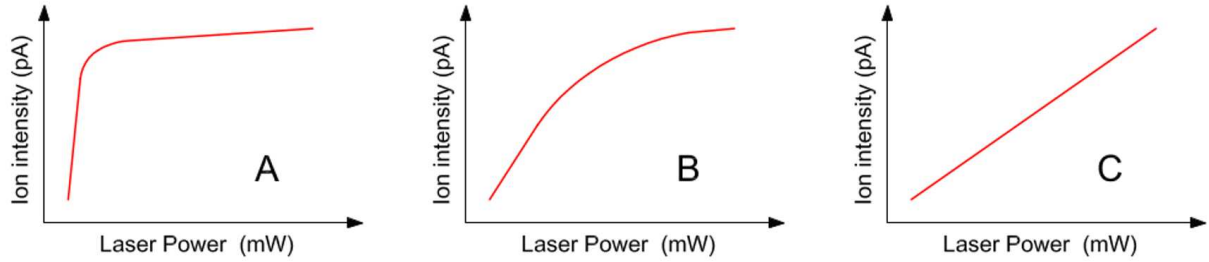


Figure 3.14– Saturation curves examples. Where (A and (B) are examples of a completely saturated and average saturated transition respectively, and (C) represents a non-saturated transition.

reason the laser power is also measured in the ion source with open chamber at maximum laser power, and the decreased laser powers are weighted (a linear fitting is applied). The parameters that affect a resonance saturation do not only depend on the laser power. An optimized focus size and a correct overlap in time and space for all lasers are expected to maximize the efficiency of the process. Nevertheless some atomic transitions are much easier saturated than others. As already commented, the Einstein A-value gives an idea of the transition probability, thus transitions with higher values ($>10^8 \text{ S}^{-1}$) will be easily saturated, and lower A-values ($<10^4 \text{ S}^{-1}$) will have very low probability of resonant ionization. Usually for the last step, auto-ionizing resonant transitions have high A values. Otherwise, a huge amount of laser power will be needed to saturate a non-resonant transition.

3.4.3 Requested elements

Several elements are requested for SPIRAL2 to be produced by RILIS from several letters of intent (section 1.4). These elements are: zinc (Zn), tin (Sn), yttrium (Y) and gallium (Ga). Sn and Zn ion beams were produced by resonant laser ionization in the GISELE test bench and, therefore, research on the best ionization scheme for these elements has been carried out.

3.5 Zinc ionization schemes

3.5.1 Physico-chemical properties of zinc

Zinc is a chemical element with atomic number 30. It is a transition metal and the first element of group 12 of the periodic table. It has an electron configuration: $[\text{Ar}] 3d^{10} 4s^2$ and its first ionization energy is 9.39 eV [74]. The melting point at atmospheric pressure is 692.68 K.

Zn has five stable isotopes, the most abundant isotope is ^{64}Zn with 48.63% natural abundance. The other isotopes found in nature are ^{66}Zn (28%), ^{67}Zn (4%) and ^{68}Zn (19%).

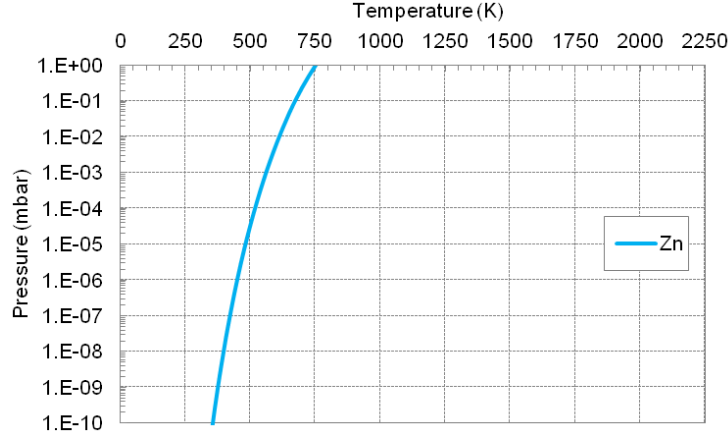


Figure 3.15– Zinc vapor pressure calculation. If an experimental pressure of $1 \cdot 10^{-6}$ mbar is considered, the evaporation temperature would be about 450 K [189].

Zinc presents high vapor pressure compared to other metals, for that reason it will be delivered at lower temperatures in the ion source (fig. 3.15). The pressure set in the ion source cavity is $\sim 1 \cdot 10^{-6}$ mbar, that corresponds to a release temperature from 450 K for zinc. Several ionization schemes were found and tested to produce a Zn ion beam by resonant laser ionization (section 3.5).

3.5.2 Atomic transitions research

A research on the available databases was performed together with experimental measurements in order to find a reliable ionization scheme for zinc. Zn has an ionization potential of 9.3941 eV (wavenumber: $75759.328 \text{ cm}^{-1}$) [74]. Many possible schemes are available to ionize Zn but only few of them have strong enough transitions and high cross sections. Historically, a three-step scheme was first used by A. Jokinen and collaborators [190, 191]. In this scheme the atoms were first resonantly excited from the ground state $3d^{10}4s^2 \ ^1S_0$ to the intermediate atomic state $3d^{10}4s4p \ ^1P_0^\circ$ using 213.9 nm UV radiation. Then they were excited to the $3d^{10}4s4d \ ^1D_2$ second excited state by using 636.2 nm radiation. Finally non-resonant ionization was reached using a copper vapor laser laser at 510.6 nm (fig. 3.16). This ionization scheme was used for several β -decay experiments at ISOLDE (CERN) [192, 193]. This ionization scheme requires a dye laser, because light with a wavelength of 636.2 nm is not reachable with a ti:sa laser. A research was performed to find a reliable ionization scheme for Zn using a titanium:sapphire laser system. Zn does not offer many intermediate states for a three-steps scheme. Due to this fact a two-step scheme was considered too.

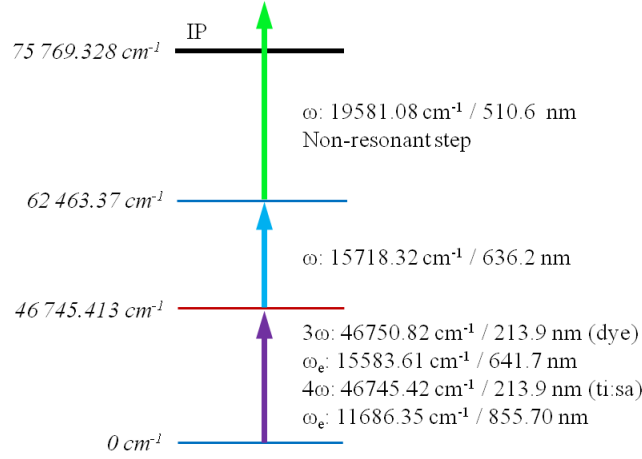


Figure 3.16– Three-steps ionization scheme used for Zn at ISOLDE. This process is only feasible with a dye laser system. For each transition, the following information is given: On the left side the wavenumber of the level and on the right side the radiation light needed for the transition. The wavenumber and wavelength of each transition are also indicated, where ω stands for fundamental radiation and $x\omega$ for frequency mixed radiation. For the latter, the equivalent fundamental radiation ω_e is provided.

3.5.2.1 First transitions

The ground state $3d^{10}4s^2S_0$ is «even» and it has a population ratio of 100%. To find a First Excited State (FES), a strong transition to «odd» state (with big A-value) is required to fulfill the selection rules (section 3.4.1). For the First Excited State (FES) there are only two possibilities (table 3.1): First, a strong UV excitation step (FES1), and second, a UV excitation step (FES2) with an A-value 4 orders of magnitude weaker compared to the first one.

The strong FES1 transition was chosen because, the A-value performance allows to excite electrons to the level of interest more efficiently than the FES2 transition for which the A-value is smaller.

Table 3.1 – Transitions to the First Excited States of Zn. Transition wavelengths are given for vacuum.

Name of transition	Initial state	Parity	Final state	Parity	A-value	Required wavelength	Required ti:sa wavelength
FES1	$3d^{10}4s^2\ ^1S_0$ 0 cm^{-1}	«even»	$3d^{10}4s4p\ ^1P_1^\circ$ $46\ 745.413\text{ cm}^{-1}$	«odd»	$7.04 \cdot 10^8\text{ s}^{-1}$	213.9247 nm 4ω	855.6989 nm
FES2	$3d^{10}4s^2\ ^1S_0$ 0 cm^{-1}	«even»	$3d^{10}4s4p\ ^3P_1^\circ$ $32\ 501.421\text{ cm}^{-1}$	«odd»	$2.96 \cdot 10^4\text{ s}^{-1}$	307.6789 nm 3ω	923.0366 nm

3.5.2.2 Intermediate states

Thirteen possible intermediate transitions (Second Excited States) were found by searching in Kurucz [75] and NIST [74] databases (table 3.2), which have reasonable A-value from FES1. None of them can be accessed with the fundamental laser radiation, but with frequency mixed radiation. All of these levels are $\ll\text{even}\gg$ so the transition from the already described FES is allowed.

Table 3.2 – Transitions from FES1 to Second Excited States of Zn. Transition wavelengths are given for vacuum.

Name of transition	Initial state	Parity	Final state	Parity	Wavelength/ Wavenumber	Required transition wavelength
SES1	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s5d\ ^1D_2$ 68338.51 cm ⁻¹	$\ll\text{even}\gg$	463.1110 nm 21593.097 cm ⁻¹	926.2219 nm 2ω
SES2	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s7s\ ^1S_0$ 70003.73 cm ⁻¹	$\ll\text{even}\gg$	429.9537 nm 23258.317 cm ⁻¹	859.9074 nm 2ω
SES3	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s6d\ ^1D_2$ 71050.47 cm ⁻¹	$\ll\text{even}\gg$	411.4370 nm 24305.057 cm ⁻¹	822.8740 nm 2ω
SES4	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s8s\ ^1S_0$ 71956.21 cm ⁻¹	$\ll\text{even}\gg$	396.6554 nm 25210.797 cm ⁻¹	793.3109 nm 2ω
SES5	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s7d\ ^1D_2$ 72516.98 cm ⁻¹	$\ll\text{even}\gg$	388.0245 nm 25771.567 cm ⁻¹	776.0490 nm 2ω
SES6	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s9s\ ^1S_0$ 73060.65 cm ⁻¹	$\ll\text{even}\gg$	380.0080 nm 26315.237 cm ⁻¹	760.0160 nm 2ω
SES7	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s8d\ ^1D_2$ 73395.42 cm ⁻¹	$\ll\text{even}\gg$	375.2344 nm 26650.007 cm ⁻¹	750.4688 nm 2ω
SES8	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s9d\ ^1D_2$ 73961.89 cm ⁻¹	$\ll\text{even}\gg$	367.4245 nm 27216.477 cm ⁻¹	734.8489 nm 2ω
SES9	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s10d\ ^1D_2$ 74348.18 cm ⁻¹	$\ll\text{even}\gg$	362.2825 nm 27602.767 cm ⁻¹	724.5650 nm 2ω
SES10	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s11d\ ^1D_2$ 74622.77 cm ⁻¹	$\ll\text{even}\gg$	358.7141 nm 27877.357 cm ⁻¹	717.4281 nm 2ω
SES11	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s12d\ ^1D_2$ 74825.05 cm ⁻¹	$\ll\text{even}\gg$	356.1300 nm 28079.637 cm ⁻¹	712.2599 nm 2ω
SES12	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s13d\ ^1D_2$ 74978.2 cm ⁻¹	$\ll\text{even}\gg$	354.1981 nm 28232.787 cm ⁻¹	708.3962 nm 2ω
SES13	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$\ll\text{odd}\gg$	$3d^{10}\ 4s14d\ ^1D_2$ 75097.09 cm ⁻¹	$\ll\text{even}\gg$	352.7128 nm 28351.677 cm ⁻¹	705.4256 nm 2ω

The A-values for transitions to $5d$, $7s$, $8s$ and $9s$ states (named respectively SES1, SES2, SES4 and SES6 in table 3.2) are about $1 \cdot 10^6 \text{ s}^{-1}$. The A-values for the rest of the lines are unknown, however 1D_2 states are singlets, thus reasonable transition strengths can be expected. The three last transitions of table 3.2 are near the limit of the ti:sa wavelength range. Therefore a lower lasing efficiency and a lower output power are expected. The second transitions were tested with a final ionizing non-resonant step.

3.5.2.3 Ionization states

For the final ionization state, it is usually more efficient to use an Auto-Ionizing State (AIS) instead of a non-resonant transition. However, most of the Zn Auto-Ionizing States are so high in energy that they are not reachable with laser light radiation, which are characterized by wavelengths higher than 100000 cm^{-1} (below 100 nm). Because of this, they are not useful for an ionization scheme. Nevertheless there are still some possible solutions at lower energies. Three «even» AIS and five «odd» AIS (table 3.3) can be tested for two-step and three-step schemes respectively. The details are indicated in table 3.3. The A-values for all transitions are unknown. The different options combining SES with ionization states will be discussed in the next section.

Table 3.3 – Possible transitions to Auto-Ionization States of Zn.

Name of transition	Initial state	Final state
AIS1	«Odd» state (table 3.1)	$3d^{10} 4p^2 \ ^3P_0$ 80175.04 cm^{-1}
AIS2	«Odd» state (table 3.1)	$3d^{10} 4p^2 \ ^3P_1$ 80394.20 cm^{-1}
AIS3	«Odd» state (table 3.1)	$3d^{10} 4p^2 \ ^3P_2$ 80792.10 cm^{-1}
AIS4	«Even» state (table 3.2)	$3d^9 4s^2 4p \ ^3P_1^\circ$ 90227 cm^{-1}
AIS5	«Even» state (table 3.2)	$3d^9 4s^2 4p \ ^3D_1^\circ$ 95209 cm^{-1}
AIS6	«Even» state (table 3.2)	$3d^9 4s^2 4p \ ^1P_1^\circ$ 95792 cm^{-1}
AIS7	«Even» state (table 3.2)	$3d^{10} 4p(^2P^\circ_{1/2})5s \ ^2[1/2]_1^\circ$ 101945 cm^{-1}
AIS8	«Even» state (table 3.2)	$3d^{10} 4p(^2P^\circ_{3/2})5s \ ^2[3/2]_1^\circ$ 103001 cm^{-1}

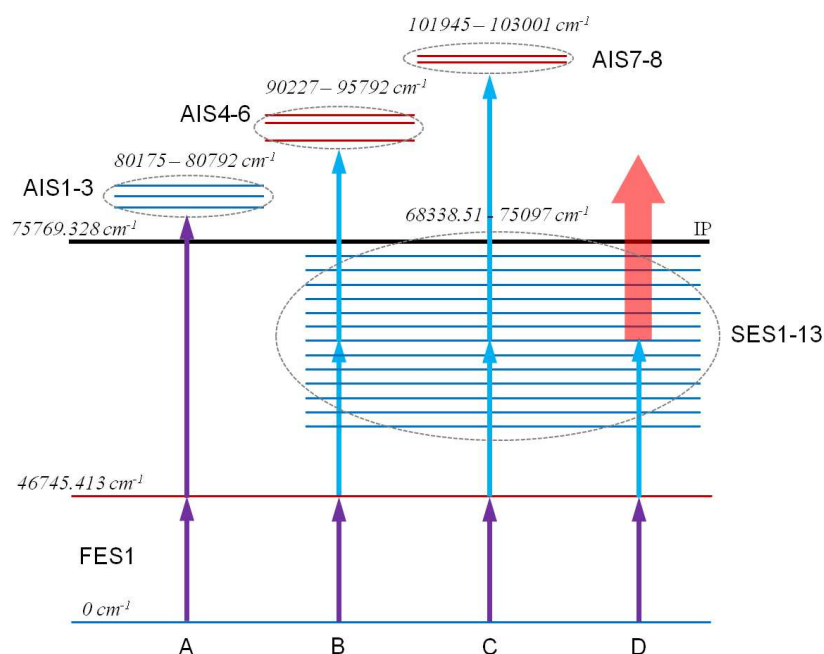


Table 3.4 – Transitions from FES1 to the possible AIS for the Zn two-step ionization schemes.

Name of transition	Second state	Final state	Wavelength/ Wavenumber	Required time/ wavelength
AIS1	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$3d^{10}\ 4p^2\ ^3P_0$ 80175.04 cm ⁻¹	299.1358 nm 33429.627	897.4076 nm 3ω
AIS2	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$3d^{10}\ 4p^2\ ^3P_1$ 80394.20 cm ⁻¹	297.1875 nm 33648.787	891.5626 nm 3ω
AIS3	$3d^{10}4s4p\ ^1P_1^\circ$ 46745.413 cm ⁻¹	$3d^{10}\ 4p^2\ ^3P_2$ 80792.10 cm ⁻¹	293.7143 nm 34046.687	881.1430 nm 3ω

Two of the three afore mentioned schemes were tested, namely AIS2: $3d^{10}\ 4p^2\ ^3P_1$ and AIS3: $3d^{10}\ 4p^2\ ^3P_2$. No auto-ionization behavior was observed in these cases. Figure 3.18 shows the results of the transition measurements (i.e. the intensity of ion signal as a function of the ionization step wavelength). Only a weak, non-resonant enhancement was observed for both AIS2 and AIS3 transitions. These two-step ionization schemes were found to be inefficient. Therefore, they were not be used for the system analyzed in this thesis.

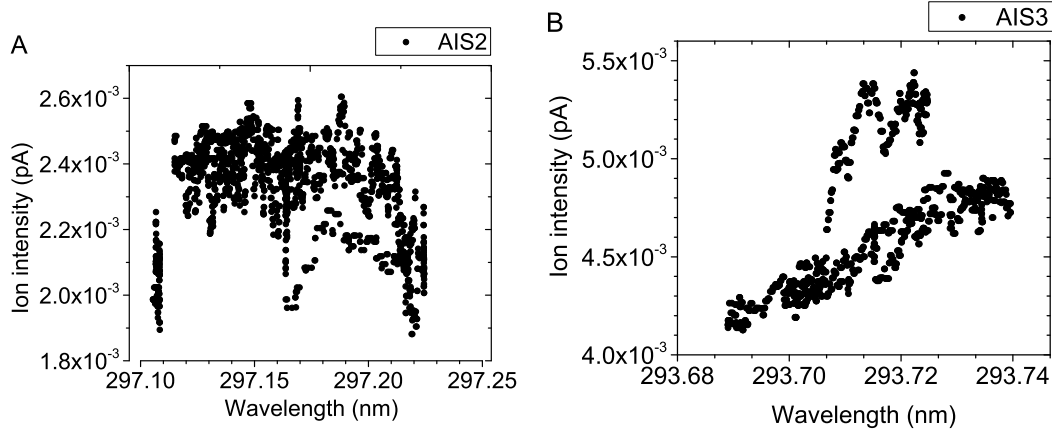


Figure 3.18– Zn Auto-Ionizing States (AIS) transitions scanned in a two-step ionization scheme: (A) AIS2 $3d^{10}\ 4p^2\ ^3P_1$ and (B) AIS3 $3d^{10}\ 4p^2\ ^3P_2$.

3.5.3.2 Three-step ionization (SES scan): Option D

Second Excited States were scanned to find the most efficient transition. As the transitions cannot reach the ionization potential, all SES were scanned with a final non-resonant ionization step of 3 W output power at ~ 910 nm. Figure 3.19 gathers all the measured SES transitions, and it shows the ion intensity signal as a function of the wavelength. For same reference cell parameters, some of the transitions were perfectly defined (e.g. SES1 $5d$, SES2 $7s$, SES3 $6d$, etc.) while others could not be determined (e.g. SES4 $8s$ and SES5 $7d$). The best results

3.5. Zinc ionization schemes

were found for SES1 (5d) and SES3 (6d) transitions (table 3.2). The comparison between both transitions showed that the signal measured in the SES1 case is 150 times higher than SES3 measured in the reference cell with same laser power.

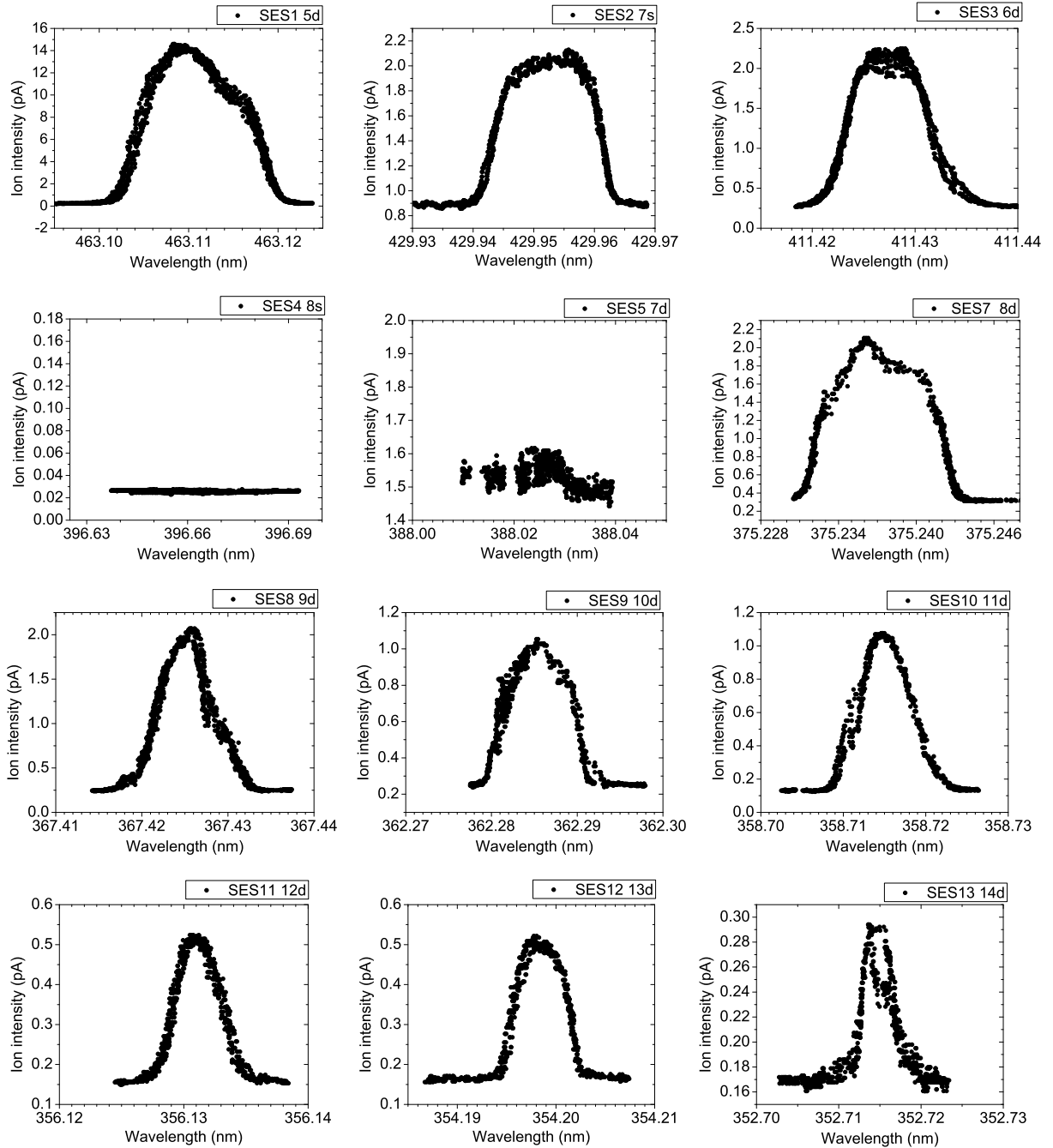


Figure 3.19– Second Excited State scans in a three-step ionization scheme.

3.5.3.3 Three-step ionization (AIS scan): Option B & C

In the case of three-step ionization schemes with a final Auto-Ionization State, most of the transitions are not accessible because they are out of the ti:sa wavelength range, and only some transitions can reach the AIS. As for the two-step scheme, the main issue is to determine whether these transitions are strong enough to efficiently ionize the atoms. Table 3.5 shows the possible combinations of SES (from table 3.2) and AIS (from table 3.3).

Table 3.5 – Zn three-step scheme combinations between Second Excited States (SES) and Auto-ionization States (AIS). Wavelengths in green were tested and wavelengths in red are outside the ti:sa wavelength range.

	AIS4 $3d^9 4s^2 4p \ ^3P_1^\circ$ 90227. cm^{-1}	AIS5 $3d^9 4s^2 4p \ ^3D_1^\circ$ 95209 cm^{-1}	AIS6 $3d^9 4s^2 4p \ ^1P_1^\circ$ 95792 cm^{-1}	AIS7 $3d^{10} 4p(^2P^\circ_{1/2})5s \ ^2[1/2]_1^\circ$ 101945 cm^{-1}	AIS8 $3d^{10} 4p(^2P^\circ_{3/2})5s \ ^2[3/2]_1^\circ$ 103001 cm^{-1}
SES1: $3d^{10} 4s5d \ ^1D_2$ 68338.51 cm^{-1}	456.9 nm 2ω	372.2 nm 2ω	364.3 nm 2ω	297.6 nm 3ω	288.5 nm 3ω
SES2: $3d^{10} 4s7s \ ^1S_0$ 70003.73 cm^{-1}	494.5 nm	396.7 nm 2ω	387.8 nm 2ω	313.1 nm 3ω	303.1 nm 3ω
SES3: $3d^{10} 4s6d \ ^1D_2$ 71050.47 cm^{-1}	521.5 nm	413.9 nm 2ω	404.2 nm 2ω	323.7 nm	313.0 nm 3ω
SES4: $3d^{10} 4s8s \ ^1S_0$ 71956.21 cm^{-1}	547.3 nm	430.1 nm 2ω	419.5 nm 2ω	333.5 nm	322.1 nm
SES5: $3d^{10} 4s7d \ ^1D_2$ 72516.98 cm^{-1}	564.7 nm	440.7 nm 2ω	429.6 nm 2ω	339.8 nm	328.0 nm
SES6: $3d^{10} 4s9s \ ^1S_0$ 73060.65 cm^{-1}	582.5 nm	451.5 nm 2ω	439.9 nm 2ω	346.2 nm	334.0 nm
SES7: $3d^{10} 4s8d \ ^1D_2$ 73395.42 cm^{-1}	594.1 nm	458.4 nm 2ω	446.5 nm 2ω	350.3 nm 2ω	337.8 nm
SES8: $3d^{10} 4s9d \ ^1D_2$ 73961.89 cm^{-1}	614.8 nm	470.7 nm 2ω	458.1 nm 2ω	357.4 nm 2ω	344.4 nm
SES9: $3d^{10} 4s10d \ ^1D_2$ 74348.18 cm^{-1}	629.8 nm	479.4 nm	466.3 nm 2ω	362.4 nm 2ω	349.0 nm
SES10: $3d^{10} 4s11d \ ^1D_2$ 74622.77 cm^{-1}	640.9 nm	485.8 nm	472.4 nm	366.0 nm 2ω	352.4 nm 2ω
SES11: $3d^{10} 4s12d \ ^1D_2$ 74825.05 cm^{-1}	649.3 nm	490.6 nm	476.9 nm	368.7 nm 2ω	354.9 nm 2ω
SES12: $3d^{10} 4s13d \ ^1D_2$ 74978.2 cm^{-1}	655.8 nm	494.3 nm	480.5 nm	370.8 nm 2ω	356.9 nm 2ω
SES13: $3d^{10} 4s14d \ ^1D_2$ 75097.09 cm^{-1}	660.9 nm	497.2 nm	483.2 nm	372.5 nm 2ω	358.4 nm 2ω

The transitions out of the ti:sa range are marked in red, and the measurements carried out with our system are marked in green. These transitions were chosen due to the easy characterization with a same cavity mirror set.

A weak, non-resonant enhancement was observed for all of the tested Auto-Ionization States and no measurement was recorded. In addition, no wavelength dependence was observed for SES9, SES10, SES11, SES12 and SES13 transitions. None of the tested SES-AIS transition combinations were strong enough to provide efficient resonant ionization.

3.5.3.4 Zn saturation measurement

Saturation measurements were performed in the ion source test bench (described in next chapter) for the transitions that showed the best performance in the reference cell: FES1 ($3d^{10}4s4p^1P_1^\circ$), SES1 ($3d^{10}4s5d^1D_2$) and SES3 ($3d^{10}4s6d^1D_2$) (fig. 3.20). The non-resonant ionization transition was tested and, since no saturated behavior was observed, none point was recorded.

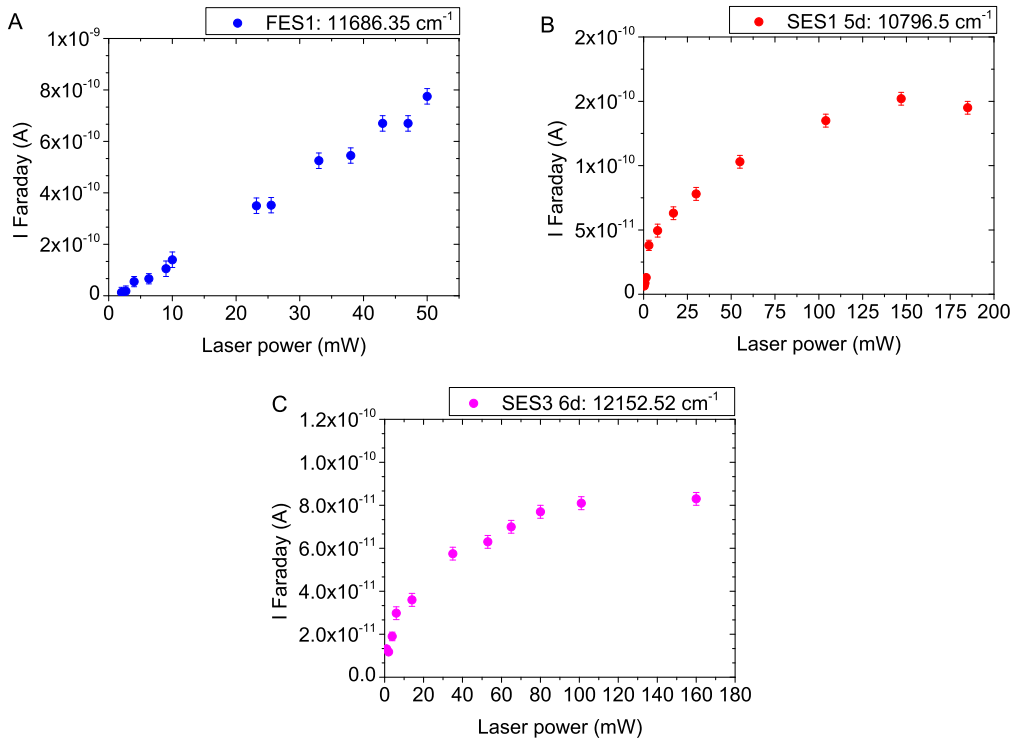


Figure 3.20– Saturation measurement for Zn three-step schemes (A) FES1: $3d^{10}4s4p^1P_1^\circ$, (B) SES1: $3d^{10}4s5d^1D_2$ and (C) SES3: $3d^{10}4s6d^1D_2$. The laser power was measured after the resonator in the case of fundamental radiation and after two dichroic mirrors in the case of frequency mixed radiation.

For the first excitation step (FES1), no saturation was observed with a 50 mW laser output power (~ 15 mW into the ion source). For second excitation steps (SES1 and SES3), saturation was reached, and the saturation laser output powers were determined to be 200 mW (~ 60 mW into the ion source) for both transitions. Finally, no saturation behavior was observed with a laser output power of 3 W (1.5 W into the ion source) for the non-resonant ionization step.

3.5.4 Conclusion

Twelve ionization schemes were tested for Zn, where most of them were found to be inadequate for an efficient resonant ionization. In particular, two-step and three-step schemes were tested. In the two-step cases, the low efficiency of the tripled ti:sa implied a low transition efficiency. For that reason these schemes were rejected. In the case of three-step schemes with final AIS no strong resonance was found. Therefore, they were also rejected for Zn ion production. Finally, a three-step scheme with a final non-resonant transition was chosen for the production of Zn ions in the GISELE test bench.

In this case, FES1 transition was chosen due to its high A-value. However no saturation was observed with ~ 15 mW laser power injected in the ion source (fig. 3.20). Moreover, the SES1 transition was used, due to the enhancement observed in the reference cell compared to SES3 transition. In addition, the SES1 transition was saturated, which ensures a maximum excitation efficiency. In the case of the ionization step, none resonant AIS was found to be used in combination with the measured transitions (table 3.5). For this reason a non-resonant step was chosen at 913 nm. The transition scans for the first and second steps can be found in figure 3.21.

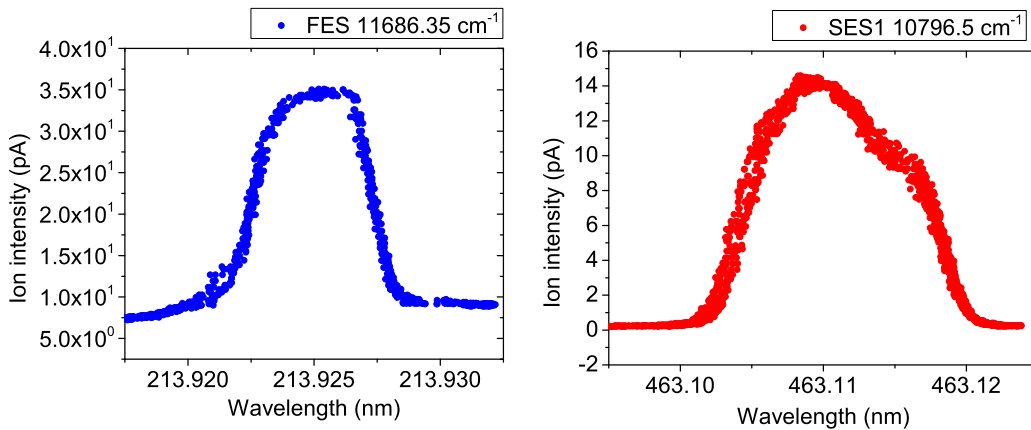


Figure 3.21– First Excitation Step (FES1) and Second Excitation Step (SES1) transitions scans for Zn. The wavenumber measured by the wavemeter is indicated in the upper part of the figures.

In conclusion, tisa lasers were set to photo-ionize Zn using the three-step scheme shown in figure 3.22. The Zn atoms were ionized with two resonant transitions. Firstly, a transition from the $4s^2 \ ^1S_0$ atomic ground state to the $4s4p \ ^1P_1^\circ$ state at $46745.413 \text{ cm}^{-1}$ by using quadrupled frequency photons of 213.9247 nm (46745.42 cm^{-1}). A second transition was induced from the previous level to the $3d^{10} 4s5d \ ^1D_2$ state at 68338.51 cm^{-1} by using doubled frequency photons of 463 nm (21598.27 cm^{-1}). Finally a non-resonant excitation was set across the ionization potential at 913 nm.

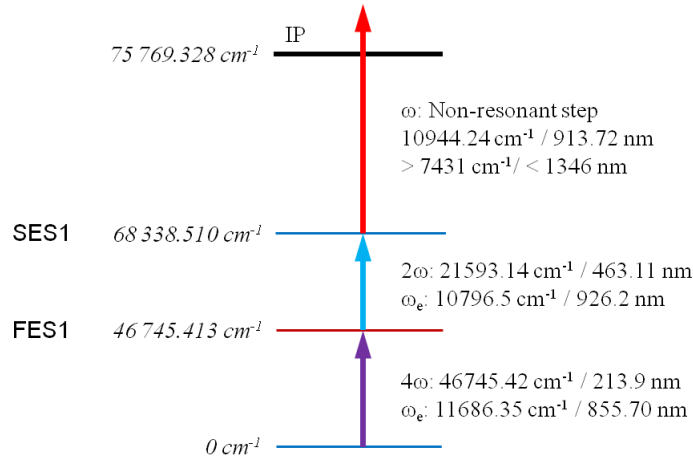


Figure 3.22– Ionization scheme chosen for resonant laser ionization of Zn at GANIL with a final non-resonant ionization transition. For each transition, the following information is given: On the left side the wavenumber of the level and on the right side the vacuum wavelength for the transition. The wavenumber and wavelength of each transition are given, where ω stands for fundamental radiation and $x\omega$ stands for frequency mixed radiation. For the latter, the equivalent fundamental radiation ω_e is also provided.

3.6 Tin ionization schemes

3.6.1 Physico-chemical properties of tin

Tin is a chemical element with atomic number 50. It is a post-transition metal of the group 14 of the periodic table. It presents an electron configuration of: $[\text{Kr}] 4d^{10} 5s^2 5p^2$ with a first ionization energy of 7.34 eV [74] and a melting point at atmospheric pressure of 505.08 K. With 10 stable isotopes, Sn has the largest number of stable isotopes in the periodic table. Their atomic mas numbers go from 112 to 124.

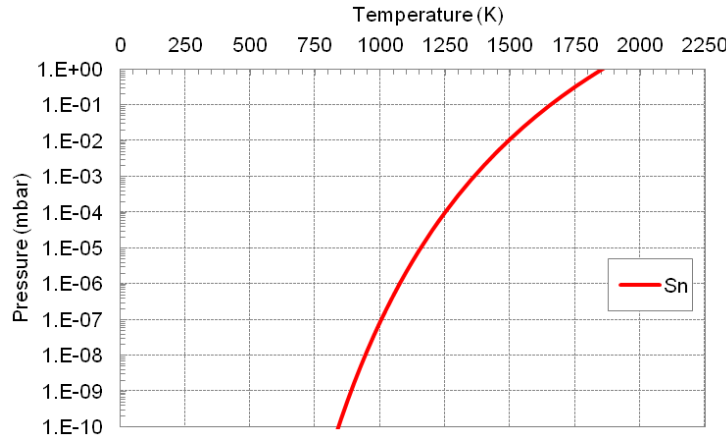


Figure 3.23– Tin vapor pressure calculation. If an experimental pressure of $1 \cdot 10^{-6}$ mbar, the evaporation temperature would be 1100 K [189].

The most abundant Sn isotopes are ^{120}Sn (32.58% of natural abundance), ^{118}Sn (24.22%), and ^{116}Sn (14.54%). Sn has a medium-high vapor pressure. If a pressure of about $1 \cdot 10^{-6}$ mbar is considered, Sn will be delivered at 1100 K (fig. 3.23). Sn has an ionization potential of 7.3434 eV [74]. The surface ionization efficiency of Sn is considerably lower ($\epsilon_{surf} = 0.006\%$) than the one of rare-earth elements [95]. The ionization schemes available in the literature were compared in order to efficiently ionize Sn by multi-step resonant excitation.

3.6.2 Possible ionization schemes

A study was carried out in order to compare the already known Sn ionization schemes and find the most reliable solution for the laser system. Tin has a relative medium ionization potential for RILIS of 7.3434 eV (59232.69 cm^{-1} in wavenumber) [74], which eases the resonant ionization process and prevents surface ionization. Several possibilities were found available for ti:sa laser systems in different laboratories.

The first use of stepwise resonant photo-excitation process for Sn was described by F. Scheerer [194]. It consisted of a three-step scheme, as described in table 3.6. A variation of this scheme was presented by V. Fedoseyev [97], which used the transition from the initial state $5s^25p^2 \ ^3P_1$ at 1691.806 cm^{-1} for the first step of excitation. This ionization scheme has been extensively used in ISOLDE (CERN) runs as the conventional scheme for Sn. Several spectroscopy [195], decay experiments [196] and ISOL technology studies [197] have been performed using this scheme. In addition, another variation was described by Y. Liu [169] at Oak Ridge. In this case, the first transition occurs from the ground state $5s^25p^2 \ ^3P_0$. Figure 3.24 (A-B-C schemes) shows an overview of these three ionization scheme options.

3.6. Tin ionization schemes

KU Leuven also reported two-step schemes [198], as indicated in table 3.7. These schemes are identified as D and E schemes in (fig. 3.24). The ionization scheme D is suitable for a ti:sa system. However, the scheme E is not convenient because the AIS-E transition (627.2180 nm) cannot be produced by a ti:sa system. The only possibilities are to use the FES-E transition and then scan for an unknown AIS or Rydberg state, or to use the fundamental ti:sa radiation for non-resonant ionization.

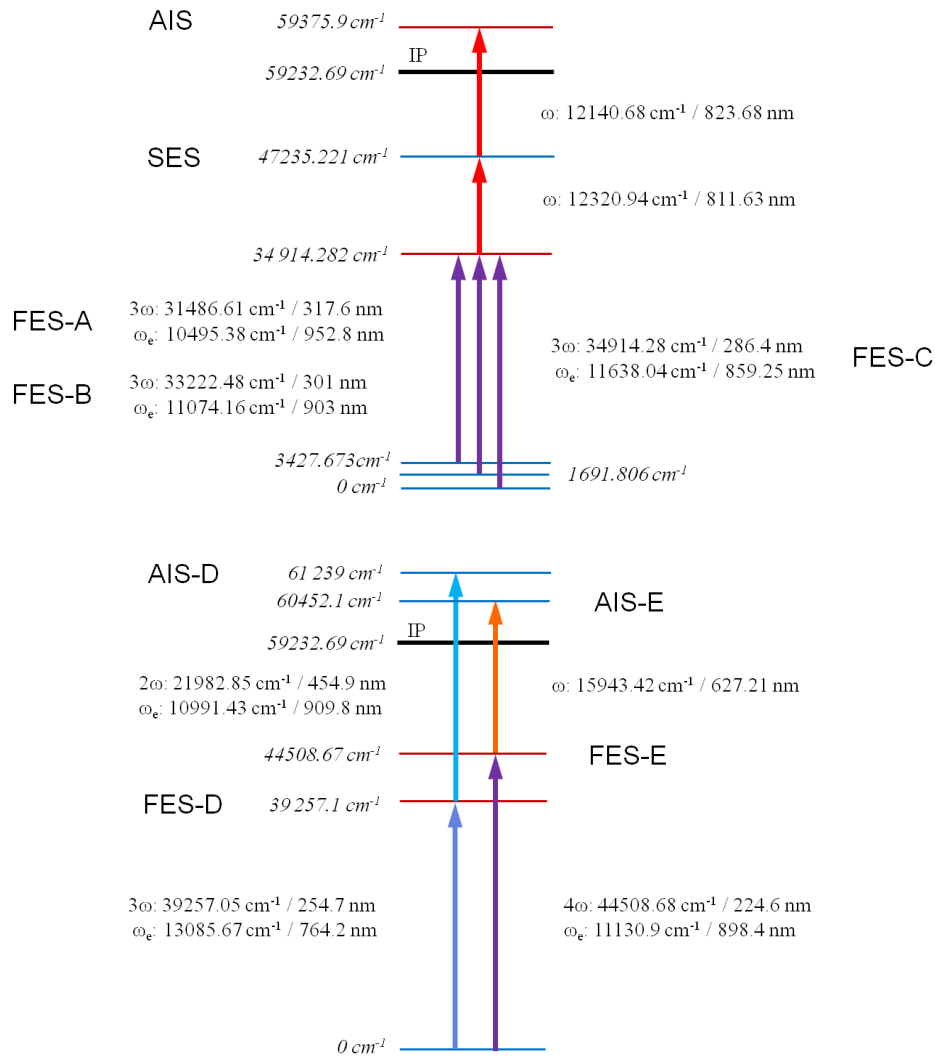


Figure 3.24— Ionization schemes for Sn: (A) Scheerer three-step scheme, (B) ISOLDE three-step scheme, (C) Oak Ridge three-step scheme and (D,E) Leuven two-step ionization scheme. The wavenumber and wavelength of each transition is given, where ω stands for fundamental radiation and $x\omega$ for frequency mixed radiation. For the latter, the equivalent fundamental radiation ω_e is also provided.

Table 3.6 – Tin three-step resonant photo-excitation schemes.

Name of transition	Initial state	Final state	Wavelength/ Wavenumber	Required ti:sa wavelength
FES-A	$5s^25p^2\ ^3P_2$ 3427.673 cm^{-1}	$5s^25p6s\ ^3P_1^\circ$ 34914.282 cm^{-1}	317.5953 nm 31486.609 cm^{-1}	952.7860 nm 3ω
FES-B	$5s^25p^2\ ^3P_1$ 1691.806 cm^{-1}	$5s^25p6s\ ^3P_1^\circ$ 34914.282 cm^{-1}	301.0010 nm 33222.476 cm^{-1}	903.0031 nm 3ω
FES-C	$5s^25p^2\ ^3P_0$ 0 cm^{-1}	$5s^25p6s\ ^3P_1^\circ$ 34914.282 cm^{-1}	286.4157 nm 34914.282 cm^{-1}	859.2472 nm 3ω
SES	$5s^25p6s\ ^3P_1^\circ$ 34914.282 cm^{-1}	$5s^25p6p\ ^3P_2$ 47235.221 cm^{-1}	811.6265 nm 12320.939 cm^{-1}	811.6265 nm ω
AIS	$5s^25p6p\ ^3P_2$ 47235.221 cm^{-1}	$5s^25p9s\ ^3P_2^\circ$ 59374.2 cm^{-1}	823.793 nm 12138.972 cm^{-1}	823.793 nm ω

Table 3.7 – Leuven two-step scheme for Sn. E scheme is not accessible for the ti:sa wavelength range.

Name of transition	Initial state	Final state	Wavelength/ Wavenumber	Required ti:sa wavelength
FES-D	$5s^25p^2\ ^3P_0$ 0 cm^{-1}	$5s^25p6s\ ^1P_1^\circ$ 39257.053 cm^{-1}	254.7313 nm 39257.053 cm^{-1}	764.194 nm 3ω
AIS-D	$5s^25p5d\ ^3D^\circ$ 39257.053 cm^{-1}	- 61240 cm^{-1}	454.77 nm 21982.853 cm^{-1}	909.8 nm 2ω
FES-E	$5s^25p^2\ ^3P_0$ 0 cm^{-1}	$5s^25p5d\ ^3D_1^\circ$ 44508.68 cm^{-1}	224.606 nm 44508.68 cm^{-1}	898.424 nm 4ω
AIS-E	$5s^25p5d\ ^3D_1^\circ$ 44508.68 cm^{-1}	- 60452.1 cm^{-1}	627.2180 nm 15943.423 cm^{-1}	x

3.6.3 Comparison and discussion

In order to compare the different ionization schemes, the reference cell unit (section 3.3) was used with a natural sample of Sn. The variations of the generated ion current were compared. The three-step schemes can be easily compared by the use of four ti:sa cavities, because they share the same SES and AIS. Two of the three-step schemes were compared switching from one scheme to another for the same reference cell parameters (i.e. resistive heating of the reference cell oven). FES-B transition ($5s^25p^2\ ^3P_1$), FES-C transition ($5s^25p^2\ ^3P_0$) and B+C ionization schemes (together) were compared by this method.

The comparison showed an enhancement of the ion current by a factor of 2 for the C ionization scheme, with respect to the B scheme. In addition, an improvement by a factor of 3 was obtained by combining FES-B and FES-C (fig. 3.25).

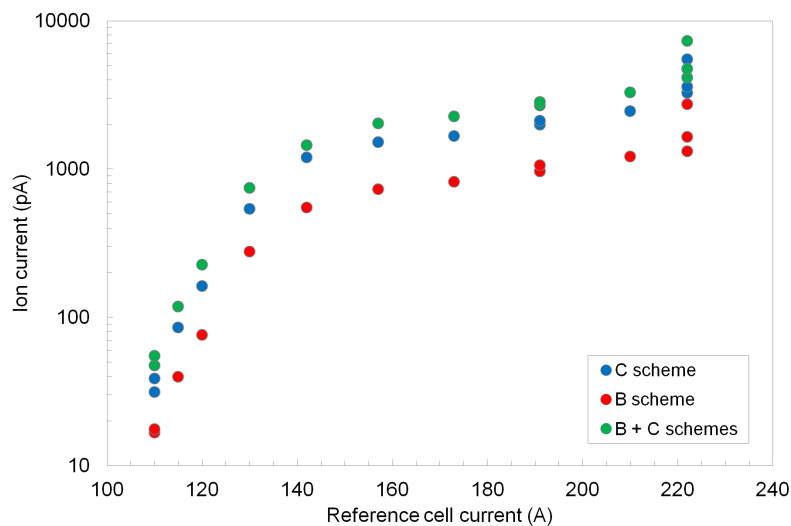


Figure 3.25– First Excitation State comparison for Sn three-step ionization schemes. B scheme, C scheme and B+C are compared. The ion current in the reference cell is represented as a function of the oven current.

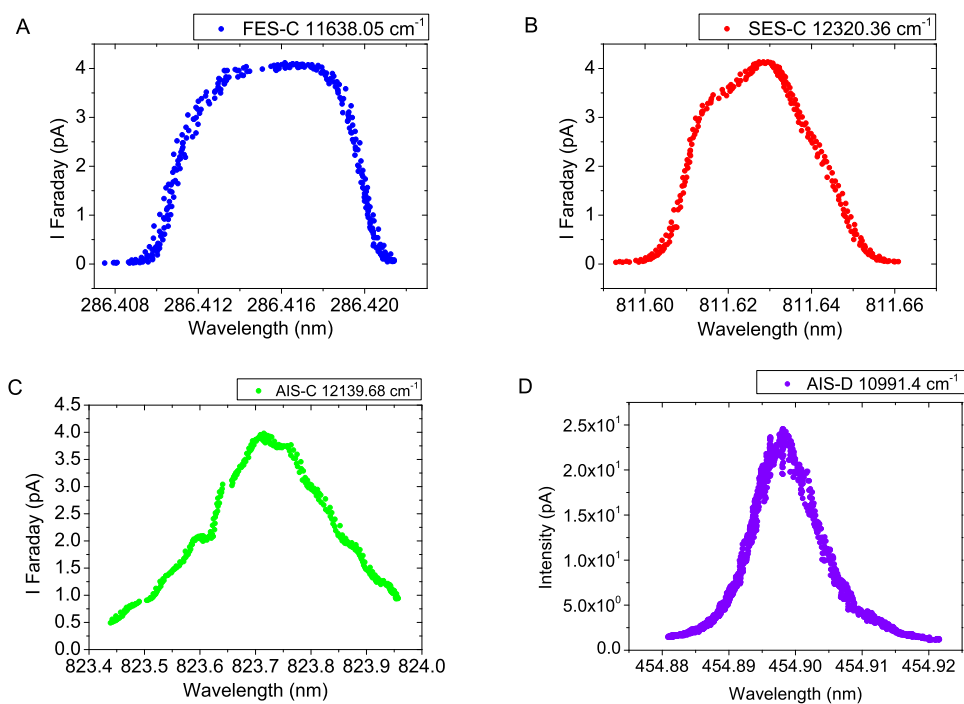


Figure 3.26– (A) First Excited State (FES), (B) Second Excited State (SES) and (C) Auto-Ionization State (AIS) transition scans for C ionization scheme and (D) Auto-Ionization State for D ionization scheme (KU Leuven scheme). The wavenumber measured by the wavemeter is indicated on the upper part of the figures.

One of the advantages of the $5s^25p^23P_0$ transition (C scheme) is that it has a population ratio of 43% at 2000 K, compared to the 38% and 18% for $5s^25p^23P_1$ (B scheme) and $5s^25p^23P_2$ (A scheme) respectively. In addition, the C scheme was compared with the Leuven D scheme in the reference cell. An increase by a factor of 13 was observed for the C scheme. A transition scan was performed for the transitions of the C scheme and for the AIS-D transition (fig. 3.26).

Finally, the First Excited State of the E scheme (FES-E) was used in order to search for an Auto-ionization State or Rydberg state. A laser cavity equipped with a grating mirror (instead of the birefringent-etalon system) was used to scan over a wide range of wavelengths. However, no AIS resonance was observed.

3.6.4 Conclusion

Five ionization schemes were tested in order to find the optimal alternative for a full time-shared RILIS laser system. The best option was based on the use of both B+C schemes to obtain the highest ionization efficiency. B+C schemes can provide a higher efficiency of ion production, however this solution needs 4 laser cavities. In order to perform the routine work with 3 laser cavities, the C scheme (fig. 3.27) was chosen for the production of resonant laser photo-excitation, due to its high performance compared to other ionization schemes.

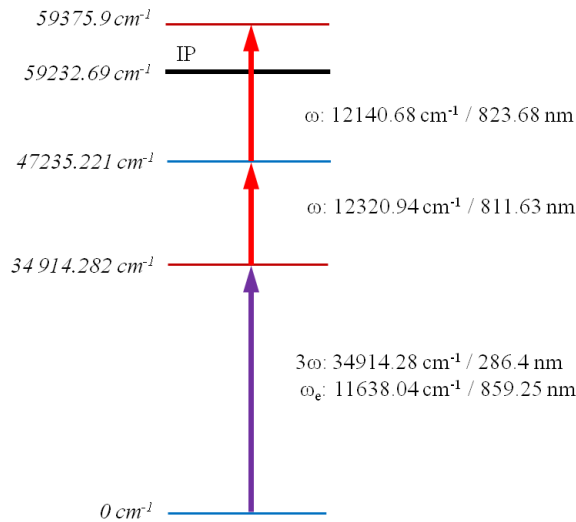


Figure 3.27– Ionization scheme chosen for resonant laser ionization of Sn at GANIL. For each transition, the following information is given: On the left side the wavenumber of the level and on the right side the radiation light needed for the transition. The wavenumber and wavelength of each transition are given, where ω stands for fundamental radiation and $x\omega$ stands for frequency mixed radiation. For the latter, the equivalent fundamental radiation ω_e is also provided.

3.6. Tin ionization schemes

Saturation measurements were performed for the C scheme in the off-line test bench for the interaction device geometry with 7 mm diameter and 35 mm length ionizer (LISBET-D7L35), which will be described in section 4.1. In this scheme, saturation was observed for the first excitation step at 11638.05 cm^{-1} (3^{rd} harmonic: 286.41 nm) with 80 mW of laser power injected into the ion source, and for the second excitation step at 12320.36 cm^{-1} (811.63 nm) with 1.8 W of laser power in the ion source. For the ionization step, beginning of saturation behaviour was observed at 12140.68 cm^{-1} (823.79 nm) with an output power of 1.65 W in the ion source (fig. 3.28).

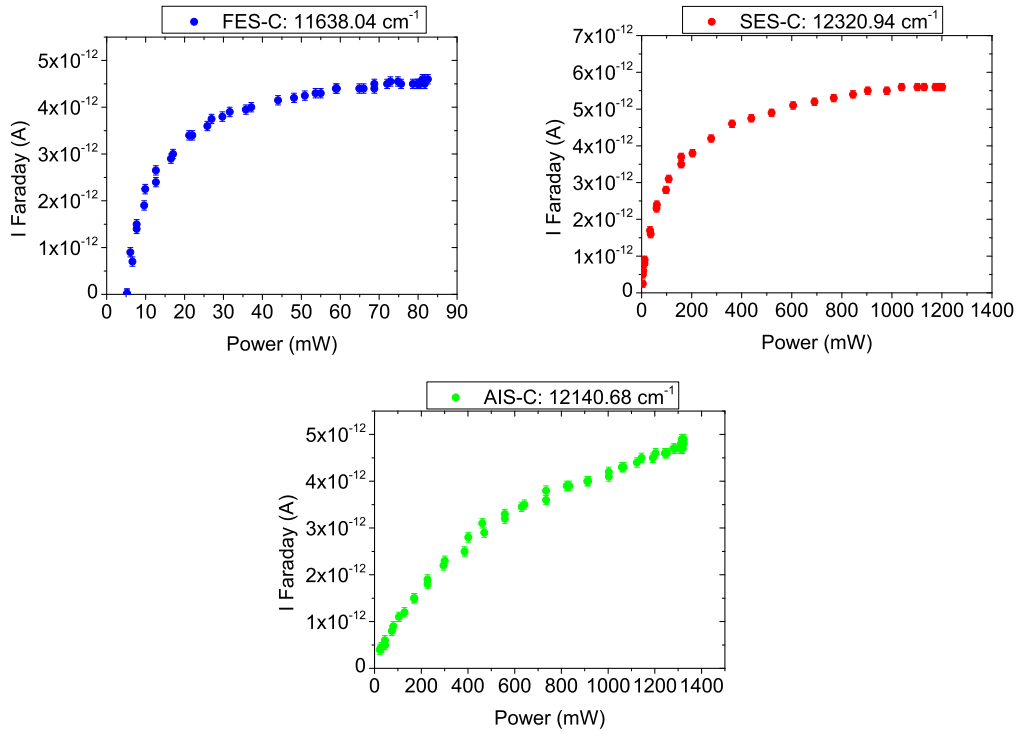


Figure 3.28– Saturation measurement using the Sn three-step scheme (C ionization scheme) for LISBET-D7L35.

Chapter 4

Ion source test bench

The ion source test bench (fig. 4.1) consists of the ion source body (which includes the interaction device), the extraction optics for an optimum ion beam leading, a magnetic sector field mass spectrometer to actively select the required isotope, and finally the detection line to measure the ion beam properties (emittance, mass scans, etc.).

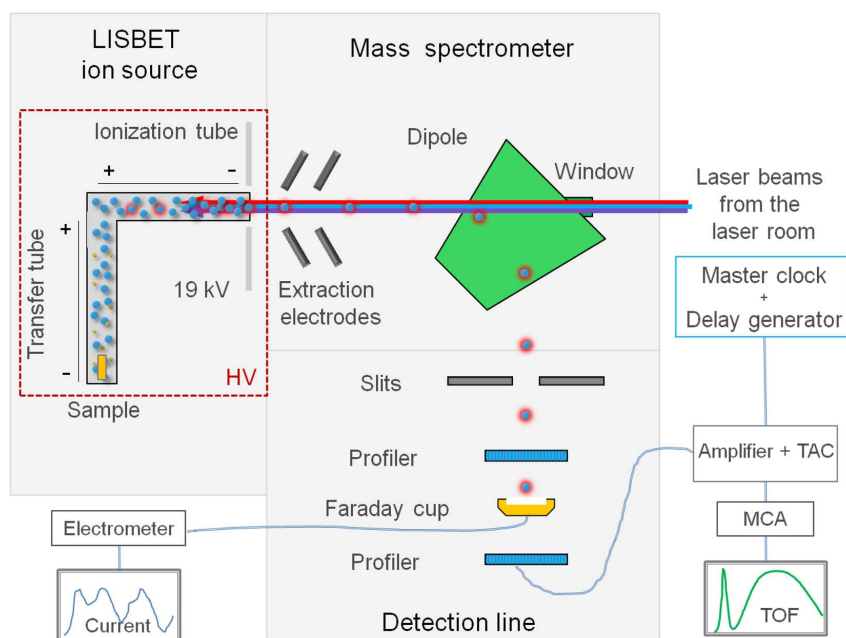


Figure 4.1– Ion source test bench. After ionization the laser ions are extracted, mass selected by a dipole mass spectrometer and finally detected with one of the two low intensity ion beam profilers or with a Faraday cup.

The test bench was previously used to study and develop ECR ion sources for SPIRAL1 and SPIRAL2 Phase-2 [59]. The former test bench was designed for ion currents of μA up to mA, and emittance of the order of $80 \pi \cdot \text{mm} \cdot \text{mrad}$. A RILIS usually produces ion beams with currents of pA up to nA and emittance of about $10 \pi \cdot \text{mm} \cdot \text{mrad}$. For a proper use, an adaptation of the test bench has been carried out. The ion source is mounted in a high voltage (HV) platform to drift the ions towards the extraction system, where the high voltage can be tuned up to 19 kV and the single extraction electrode has been optimized to a double extraction electrode set (section 4.6). A shielding was implemented in the ion source to avoid the radiative heating of the extraction elements. The Faraday cup has been replaced by the new Faraday cup type for SPIRAL2, to extend the measure to low ion beam intensities from 0.1 pA to 10 μA . Finally, the test bench was also used to study and develop a low intensity ion beam profiler prototype for SPIRAL2 [199] (section 4.4.2). Two profiler prototypes have been installed before and after the Faraday cup to guarantee a centered alignment of the ion beam.

4.1 Ion source

The ion source body is the structure where the laser light radiation interacts with the atomic vapor. The ion source consists of a transfer tube and an ionization tube with a 90 degree turn. This geometry is the consequence to the spatial configuration of the target and ion source assembly of the production module in SPIRAL2 Phase-2, as described in section 1.4. The ionization tube and the transport tube have independent voltage connectors and they can be resistively heated by an electrical current to temperatures up to 2000 K. For off-line measurements in the test bench, the target is not present, the transport tube is used as atomizer (fig. 4.1). Samples containing the element of study are placed in the transport tube/atomizer and they are resistively heated until total evaporation by an electrical current. The neutral species effuse through the transport tube into the ionization tube where they are ionized. Different geometry configurations have been studied and compared to analyze the dependence of the geometry over the ionization efficiency.

4.1.1 Interaction device development

LISBET (Laser Ion Source Body using Efficient Techniques) is an ion source prototype designed to investigate the geometries of the on-line ion source layout by varying the diameter and length of the ionization tube (fig. 4.2). It consists of two tantalum tubes, an atomizer tube with 7 mm diameter and 80 mm length, and an ionizer tube with 3 or 7 mm diameter and 35 or 60 mm length (table 4.1). All tubes have 0.2 mm thickness to provide a high electrical resistivity that will lead to significant electric potential at high temperatures ($>1400 \text{ K}$).

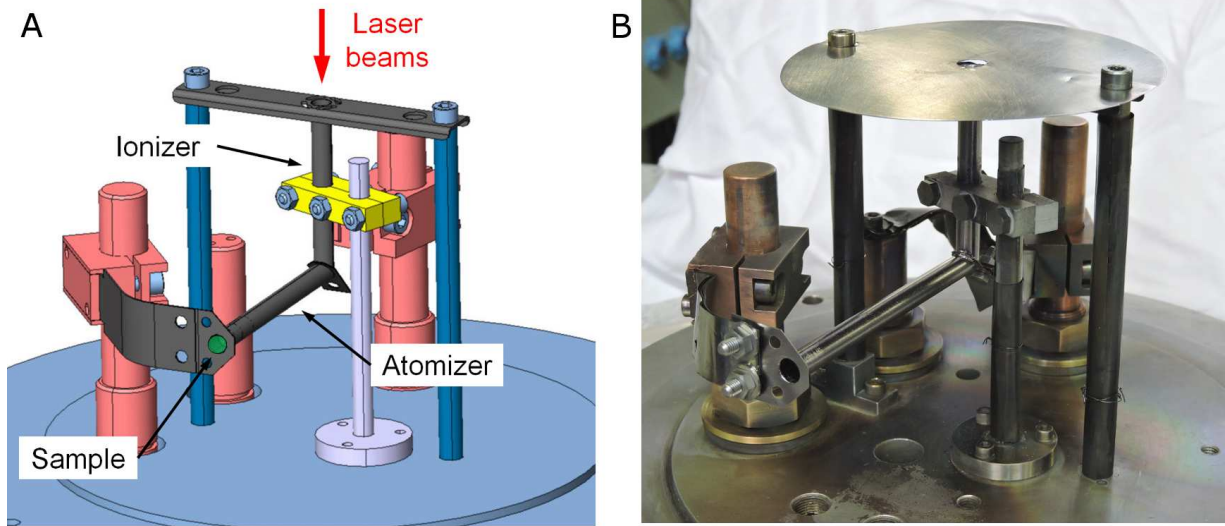


Figure 4.2– LISBET ion source: (A) CAD design and (B) ion source test prototype.

Table 4.1 – Comparative table of each laser ion source body used at GANIL.

Name	Ionizer Diameter	Ionizer length	Atomizer Diameter	Atomizer length
LISBET-D7L60	7 mm	60 mm	7 mm	80 mm
LISBET-D3L60	3 mm	60 mm	7 mm	80 mm
LISBET-D7L35	7 mm	35 mm	7 mm	80 mm
LISBET-D3L35	3 mm	35 mm	7 mm	80 mm

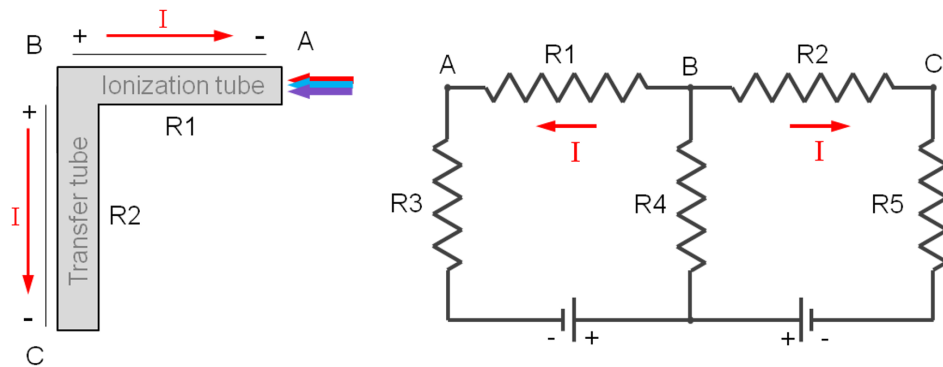


Figure 4.3– Electric scheme of LISBET. (Right) LISBET diagram with the ionizer connectors (A-B) and the ionizer/transfer connectors (B-C). (Left) the corresponding electric diagram, where the ionizer is the resistance R1, the atomizer/transfer the resistance R2 and R3, R4 and R5 are the resistances of the cables.

4.1.2 Electric field configurations

The resistive heating current generates an electric field potential of several volts (depending on the applied current) inside the ionizer tube and transfer/atomizer tube (fig. 4.3). Due to this, the voltage drop due to resistive heating can modify its trajectories. That means by the control of the direction of the electric field, the ions will be drifted to the extraction system or otherwise they will be rejected back to the production target area. The effect of the electric potential was tested to be used as contaminant reduction technique (chapter 5.5). The electric potential was measured directly on the ion source connectors as shown in section 4.1.5. The four configurations tested for the direction of the electric field are shown in figure 4.4A-D .

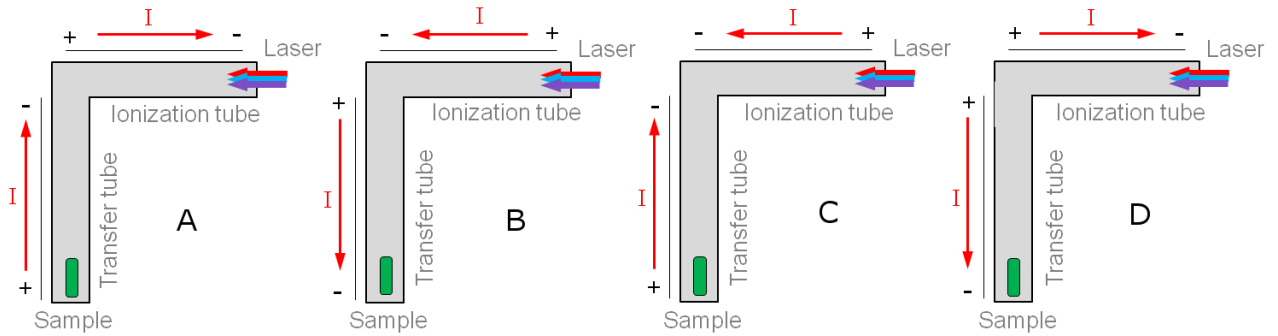


Figure 4.4– The possible configurations for the electric current in the ion source: (A) electric field towards the extraction region. (B) electric field repelling all ions backwards to the “target place”; (C) electric field forward in the transfer tube and backwards for the ionization tube; (D) electric field repelling the ions generated in the transfer tube and pushing forwards the ions generated in the ionization tube.

4.1.3 Low electron emission work function materials

For some experiments, a low work function material (ZrC) was fitted into the 7 mm diameter ion source configuration, in order to study the reduction of surface ion production inside the ion source (as described in section 2.4). ZrC was chosen for its reliability at high temperature (melting point at 3430 K) and low work function (3.65 eV). The material was developed by the SPCTS laboratory *Science des Procédés Céramiques et de Traitements de Surface* from the Université de Limoges [200]. The material consists of 7 parts of 5 mm long, with 3 mm inner diameter and 7 mm outer diameter. The parts were inserted into the ionizer tube as seen in figure 4.5. The electrical resistivity of ZrC carbides depends on its atomic crystalline configuration.

The electrical resistivity of $6 \cdot 10^{-4} \pm 2 \cdot 10^{-4} \Omega \cdot \text{m}$ was measured. This indicates that ZrC is an isolator when compared to the tantalum ion source electric resistivity of $1.35 \cdot 10^{-7} \Omega \cdot \text{m}$. The studies carried out with ZrC to decrease the generation of surface ions are described in chapter 5.5.

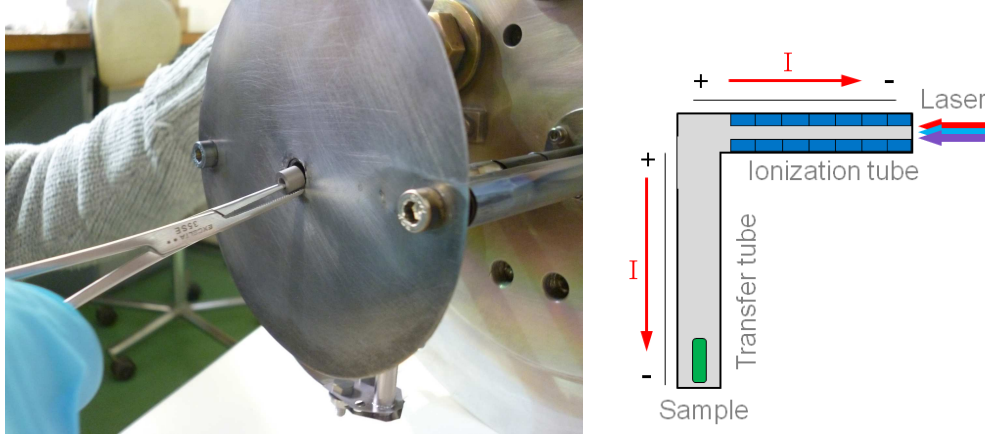


Figure 4.5– Insertion of ZrC tubes into the 7 mm diameter ionizer tube and schematic of the final assembly.

4.1.4 Ion source temperature calibration

A proper temperature calibration is fundamental to control the atomic release and to characterize the ion source. The calibration was performed by a home-made thermocouple consisting of a pair of W-Re wires (W5%Re/W26%Re) inserted inside the ionizer tube and the transfer/atomizer tube. The thermocouple gives the temperature T [°C] as a function of the measured electric potential U [V] following the formula:

$$T = 0.024786 \cdot U^3 - 0.97545 \cdot U^2 + 64.669 \cdot U + 19.27. \quad (4.1)$$

This type of thermocouple allows to measure temperatures from 673 to ~ 2100 K. Both thermocouple ends were placed approximately in the middle of each tube. A ceramic cap was also placed at the sharp end of the wire to prevent direct contact with the tube walls. The power supply used to heat the tubes is current regulated. The measurements were repeated more than 3 times to ensure a proper calibration. At temperatures higher than 2200 K the thermocouples started to fail and none point could be recorded.

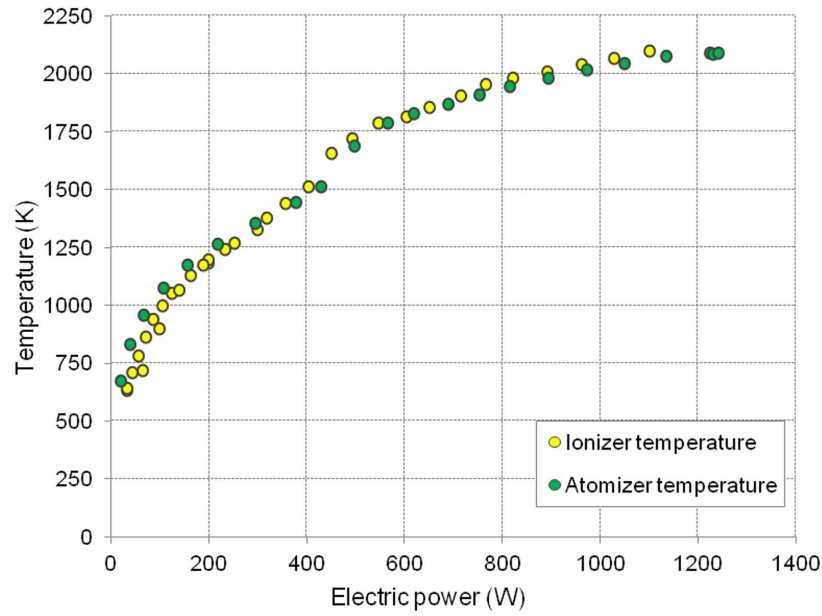


Figure 4.6– Calibration measurement for LISBET ion source with 7 mm ionizer diameter.

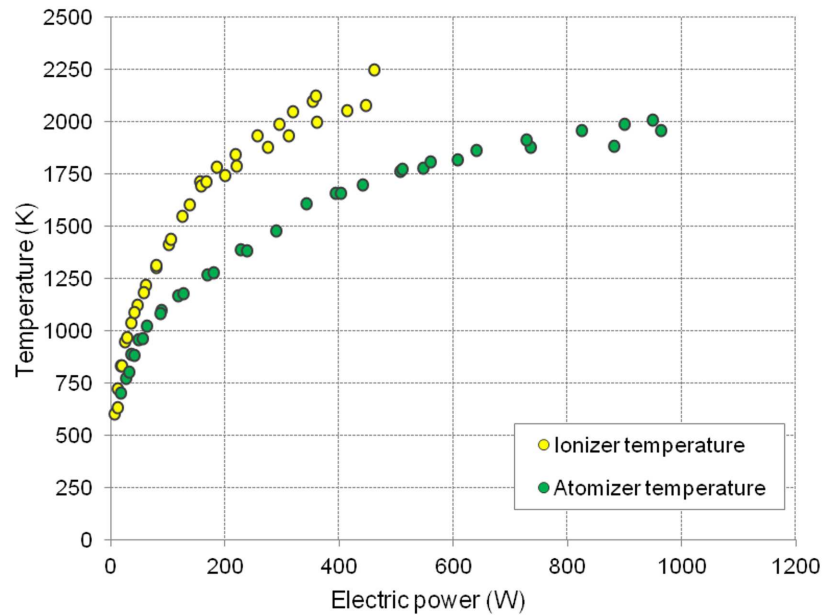


Figure 4.7– Calibration measurement for LISBET ion source with 3 mm diameter.

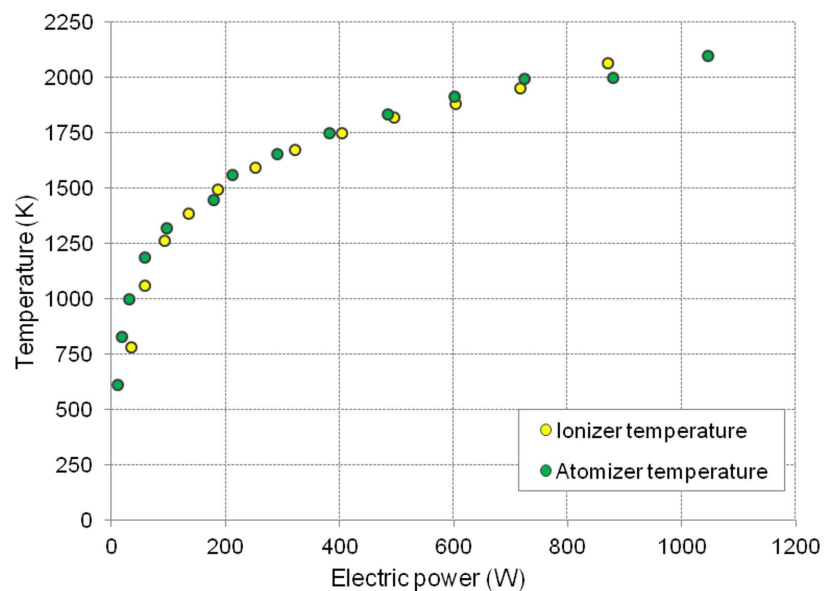


Figure 4.8– Calibration measurement with the ZrC inserted into LISBET ion source with 7 mm diameter.

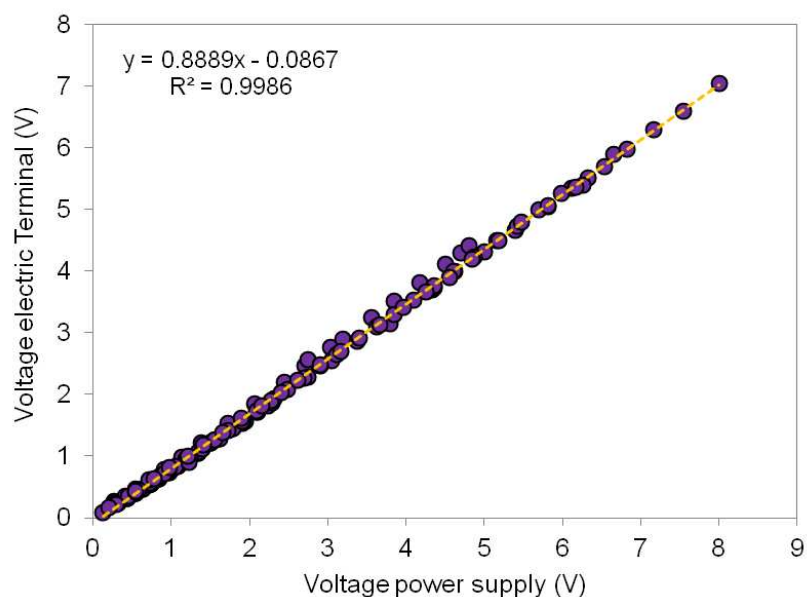


Figure 4.9– Comparison of the voltage drop measured in the ion source terminal connection (ionizer) as a function of the voltage delivered by the power supply.

In [figure 4.6](#), the measured temperatures as a function of the electric power supplied to the LISBET ion sources with ionizer diameter of 7 mm is shown. Both tube lengths (35 mm and 60 mm length) were measured and the same curve was obtained. A similar electric power is needed to heat both ionizer and atomizer tubes.

In the case of LISBET ion sources with ionizer of 3 mm diameter ([fig. 4.7](#)), the ionization tube needs less electric power to reach high temperatures. The atomizer calibration is similar to LISBET ion source with 7 mm ionizer diameter.

In addition, a temperature calibration was performed with the ZrC material inserted into the ionizer tube and the calibration curve was measured ([fig. 4.8](#)). This calibration curve is similar to the result with LISBET ion source with 7 mm ionizer diameter, for that reason it is possible to conclude that the effect of ZrC on the resistivity of the system is negligible.

As described in [section 4.1.2](#), the electric potential drop along the tube will be a fundamental part of the study. For that reason, the voltages generated due to the resistive heating were recorded. A difference between the voltage measured in the power supply and the voltage measured directly from the ion source connection was observed. This difference is constant for all ion source configurations and comes from the ohmic resistivity of the cables. In [figure 4.9](#) a compilation of data from all ion sources is presented.

For future studies, only electric potentials measured in the ion source connection were considered. A comparison of the voltages between different ion source solutions can be observed in [table 4.2](#), at temperatures of about 2000 K for the ionizer tube. However, this is not the real voltage conditions because the voltage inside the ion source (in the ionizer/transfer tube tab) could not be measured, and a diminution of these values may be expected. The resistance of the cables, from the power supply to the front-end ion source, was calculated fitting the results from [figure 4.9](#) obtaining a resistance of 0.0045 ohms for the ionizer cable and 0.006 ohms for the atomizer cable. As seen in [table 4.2](#), ionizers with 7 mm diameter have a larger electric potential drop compared to the 3 mm diameter at same temperatures. The smallest electrical potential has been registered for LISBET-D3L35.

Table 4.2 – Currents and voltages measured for different ion sources at 2000 K. The voltages were measured directly in the terminal connector.

	Current	Ionizer tube	Temperature	Current	Transfer tube	Temperature
Version	(A)	Voltage (V)	(K)	(A)	Voltage (V)	(K)
LISBET-D7L60	162	5.46	2031	140	5.27	1984
LISBET-D7L35	166.4	5.4	2061	146.1	6.6	2017
LISBET-D3L60	95	4.3	2075	147	5.34	1987
LISBET-D3L35	90.6	3.6	2098	140.9	5.53	1882

4.1.5 Interaction device thermo-electric study

A thermal study has been performed by the design department with ANSYS to characterize the temperature behavior in LISBET ion source. The results show that the geometry with an ionizer Ta tube of 3 mm diameter is more favorable to produce an homogeneous temperature. This is coherent because the conductive section is smaller, thus the effect of thermal conduction and heating loss in the extremities is lower. In addition, the radiation loss is smaller because the radiative surfaces are smaller.

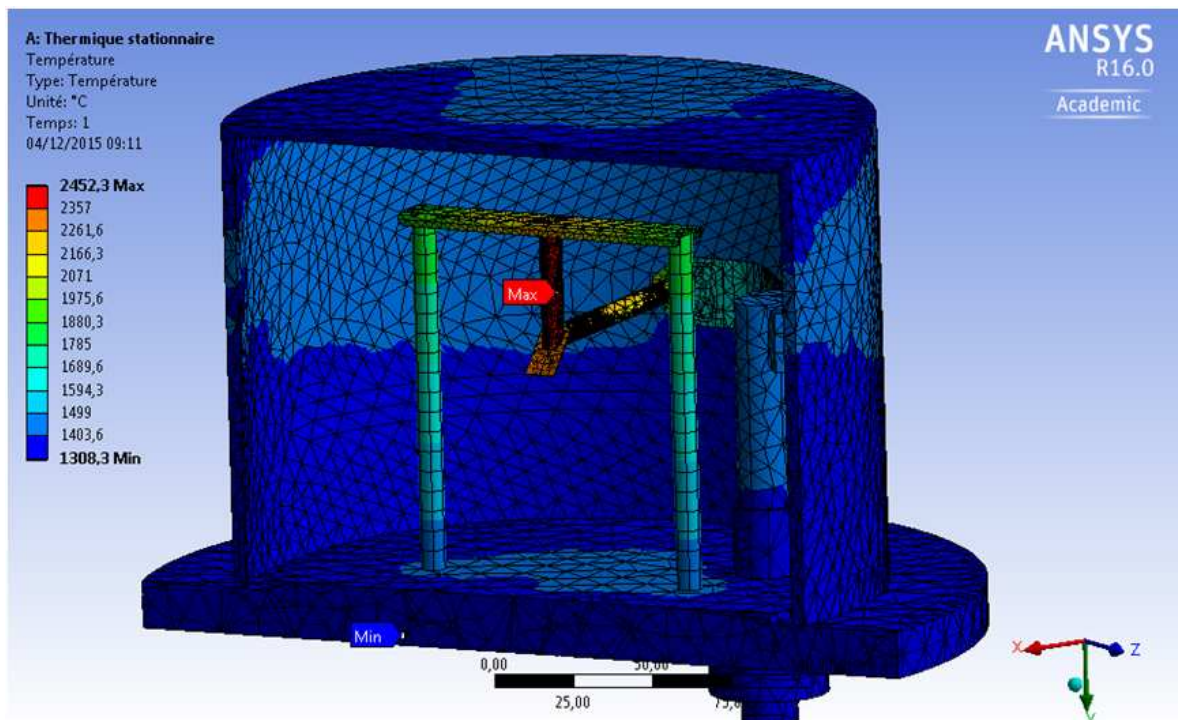


Figure 4.10– Thermal simulation of LISBET-D7L35 with mutual radiation configuration between the interaction device and the chamber.

A comparison has been performed to observe the mutual influence of the radiation between the ion source and the chamber. Figure 4.10 shows that even at 2400°C the radiation between the interaction device and the ion source chamber is lower than 5%, for this reason it was considered negligible. In addition, a thermo-electric calculation has been performed for the electric current directions. The electric current flowing from the transfer tube gives higher and more homogeneous temperatures than the other configurations (Fig. 4.11). No cold or hot spot was observed. The temperatures were similar to the measured temperatures if a 5% is added due to mutual radiation. As a conclusion, the temperature calibration was correctly performed. Best behavior was found for LISBET ionizers with 3 mm diameter.

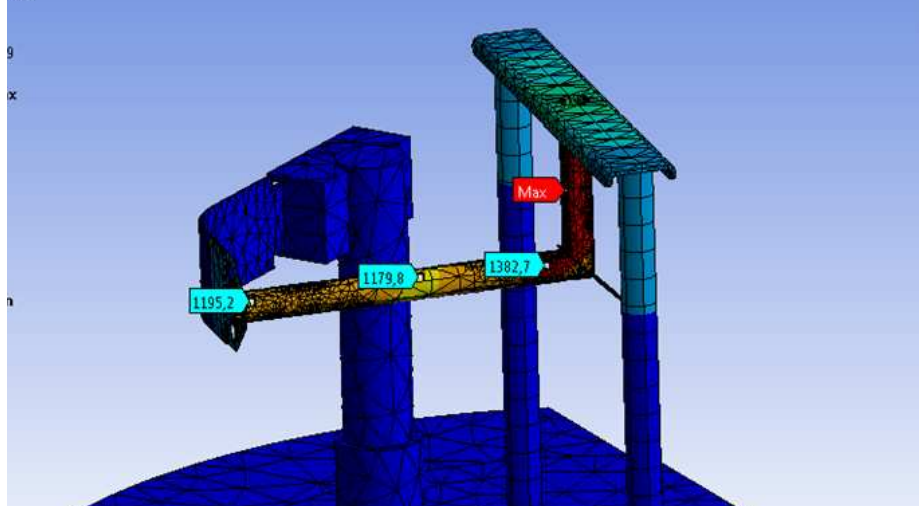


Figure 4.11– Thermo-electric calculation of LISBET-D7L35 with the electric current arriving from the elbow tab.

4.2 Ion extraction

The extraction system set at ground potential directs the ion beams from the high voltage platform ion source to the dipole magnet mass spectrometer. In the first tests, it consisted of a single extraction electrode. The extraction system is motorized and the distances can be changed on-line during ion production for optimization. The distances and voltages of the extraction electrodes were optimized by SIMION simulations [201], resulting in an addition of a second extraction electrode to improve the ion beam quality. The studies carried out to find the best parameters of distances, geometries and voltages for the extraction electrodes can be found in section 4.6.



Figure 4.12– Extraction system motorized elements and once mounted in the off-line test bench.

4.3 Magnetic sector field mass spectrometer

A magnetic sector field mass spectrometer is a device that deflects the incoming ion beam by applying a magnetic field in the sagittal plane. The particles are deflected due to the Lorentz force $\vec{F}$:

$$\vec{F} = q(\vec{E} + v \times \vec{B}) \quad (4.2)$$

where q is the particle charge, $\vec{E}$ the electric field (in this case $= 0$), v the particle velocity and $\vec{B}$ the magnetic field. The isotopes of interest can be selected through a variation of the magnetic field. The magnetic field is controlled by the current flowing through the dipole coils. A range of currents can be applied up to 265 A.

The dipole magnet also permits mass scan measurements by the recording of the signal intensity in the Faraday cup while the current of the dipole is modified. The dipole magnet was built forming an angle of 102 degree in a central trajectory with radius 405.59 mm and it has a faced magnetic shunt entry mounted at 31.62 degree to focus the ion beams. The focal distance is approximately 655 mm. The acceptance of the magnetic mass separator is $150 \pi \cdot \text{mm} \cdot \text{mrad}$ and the $B\rho$ is 0.2209048 T·m. The maximum mass that can be selected is ~ 290 with a current of 265 A at 19 kV extraction potential.

4.4 Detection system

The detection system gives information about the ion beam characteristics and allows recording for further analysis. It consist of a pair of motorized metal plate slits, a Faraday cup to detect the ion beam intensity and two low intensity ion beam profilers to measure the profile and emittance of the ion beam (section 5.2). The metal plate slits are placed in the image point of the dipole mass spectrometer to select the ion beam of interest. As described in section 4, the detection system has been adapted to measure ion beams generated by RILIS.

4.4.1 Faraday cup

The ion current was recorded by a Faraday cup (FC) system developed for SPIRAL2. The Faraday cup has been designed for intensities from pA up to μA , with a sensor diameter of 30 mm. The sensor measures the logarithm of the current over the voltage. It also incorporates electronics to increase the signal to noise ratio.

Table 4.3 – Faraday cup error measured for a chosen caliber set.

Caliber	Range	Error
1 pA/V	0 - 10 pA	± 0.008 pA
10 pA/V	0 - 100 pA	± 0.05 pA
100 pA/V	0 - 1 mA	± 0.4 pA
1 nA/V	0 - 10 mA	± 3 pA

The Faraday cup caliber can be changed to measure ion currents of very different amplitude. However, the caliber induces an intrinsic error on the measured current intensity generated by its electronic. The error depends on the chosen caliber and it was measured with the average background noise without ion beam (table 4.3). This error will be considered for efficiency analysis.

4.4.2 Low intensity ion beam profiler

The test bench had installed a wire profiler, which could only detect ion beam intensities higher than μA . Therefore, it was replaced by a low intensity ion beam profiler prototype developed by the ion beam detection group at GANIL [199]. Two low-intensity profilers of this type were placed before and after the Faraday cup. This configuration guaranteed that the ion beam was perfectly centered in the Faraday cup and the ion beam size was smaller than the detector size. The profiler device consists of an aluminum foil placed at 45° on the way of the ion beam, that emits secondary electrons during the ion beam interaction. The secondary electrons are driven by both magnetic and electric fields to the detector (fig. 4.13).

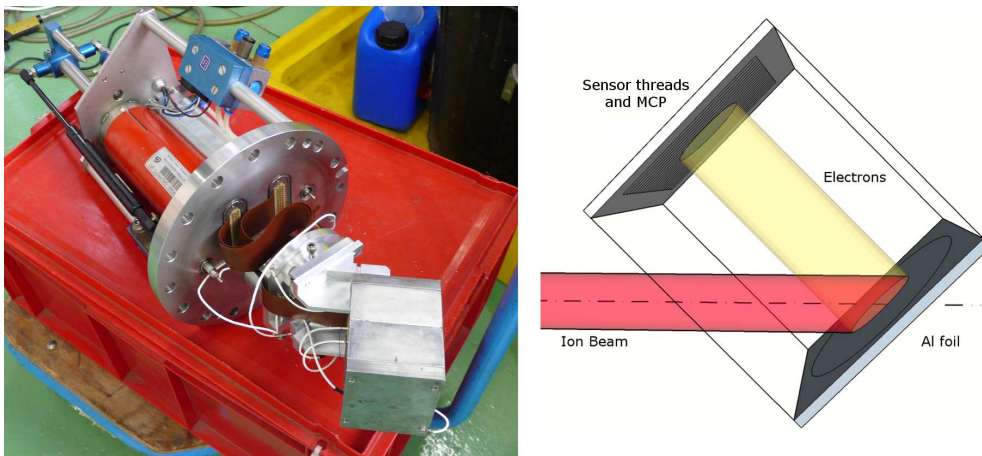


Figure 4.13– Low intensity ion beam profiler and a sketch of the detection head showing the aluminum foil that generates secondary electrons and the MCP.

The secondary electrons are detected by a micro channel plate (MCP), which increases the electron signal. The detector (MCP) consist of a regular array of tiny threads (micro-channels) densely distributed over the whole surface, horizontally and vertically to provide spatial resolution on X and Y axis. The size of the MCP is approximately 30 mm. The electrons are drifted by both electric and magnetic fields to improve the resolution. The magnetic field can be modified by changing the magnets placed on the sides of the detection cavity. The resolution of the detector is 1 mm in the horizontal axis and 0.7 mm in the vertical axis.

The aluminium foil was set to a typical electric potential of -2.2 kV and the electric potential set to the MCP can be used as gain with values between 0.7 and 1.65 kV. The working electric potential is 100 mV/thread with a breaking limit of 500 mV/thread. The integration time can be adjusted to provide enough signal/background ratio and to avoid saturation of the detector. Emittance and profile measurements can be recorded by this apparatus with ion beam currents below 1 pA (fig. 4.14). The next considerations have to be taken into account: The current in the MCP introduces a noise when a voltage higher than 1.7 kV is applied. The thread can be damaged above 2.5 kV or at vacuum pressures higher than 10^{-5} mbar. Finally, the low intensity profiler has been modified to detect and characterize the arrival time of the ion bunches. This upgrade is explained in chapter 5.3.

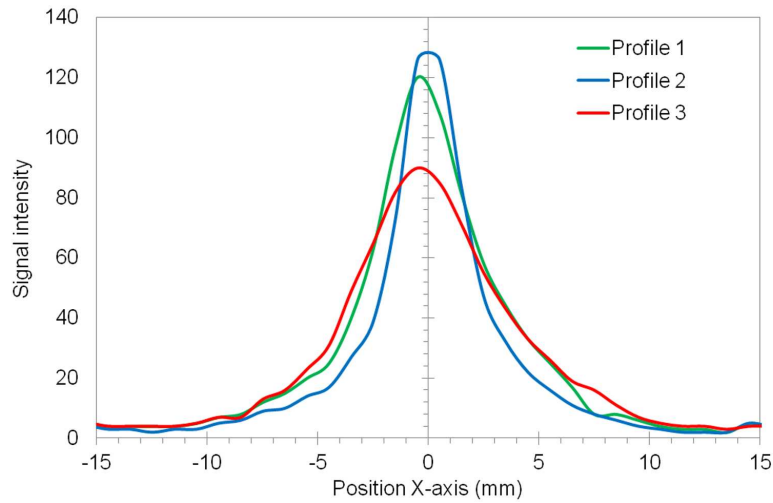


Figure 4.14– Example of X-axis profiler measurements for different ion beam extraction parameters with ion current intensities about 10 pA.

4.5 Test bench monitoring and control system

In parallel to the program described in section 3.2.6, a LabView program was integrated in the test bench to control the parameters of the ion source (fig. 4.15). By this program it

was possible to control the current applied to the ionization tube and atomizer tube and set heating ramps, the position of the extraction electrodes and of the slit plates. It also allows to control the current applied to the dipole and the high voltage of the ion source platform and the electrodes voltage. In addition it is possible to read and record the ion current signal from the Faraday cup and remotely change the caliber.

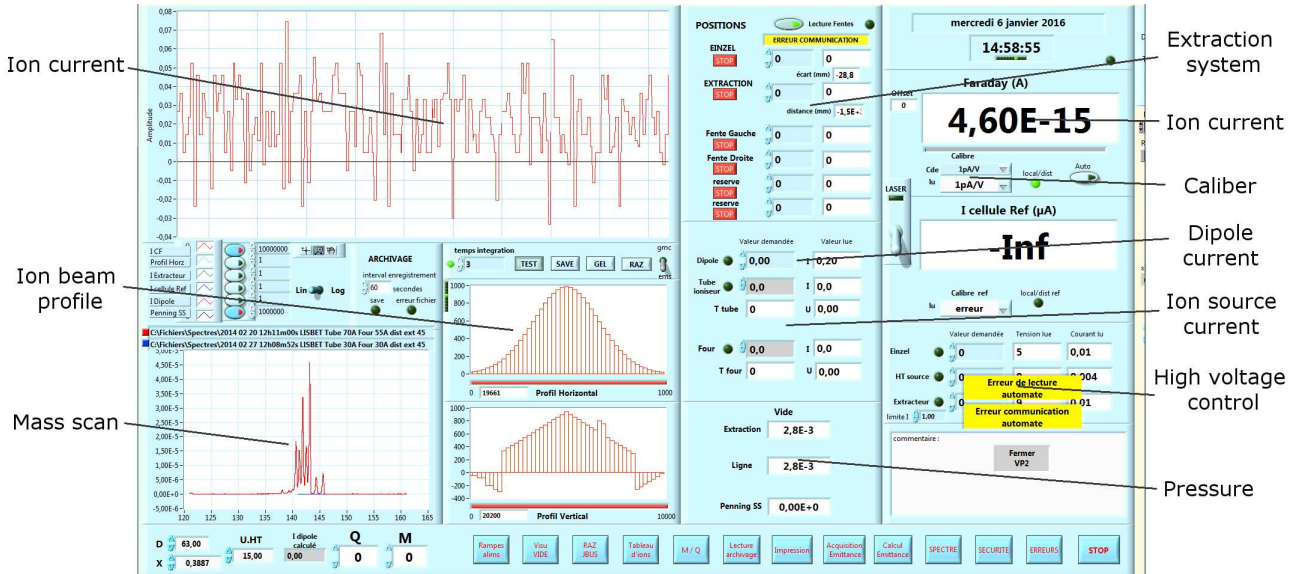


Figure 4.15– Command control front end with direct access to information concerning the ion source parameters in the test bench.

Integrated programs have been added to perform mass measurements (varying the dipole current and reading the ion current intensity) and emittance measurements (controlling the slit position and profiler parameters). Three pressure sensors are placed in different places of the test bench to monitor the pressure during the experiments, one next to the ion source, another in the extraction system and a final one in the detection line.

4.6 Extraction system optimization

The off-line test bench was simulated using SIMION [201] to optimize the extraction efficiency, and understand the underlying physics in the electric field interaction inside the ion source. By this simulation the quality of the beam was improved and information concerning the extraction and ions time of flight was obtained. The results presented here correspond to the optimization of the extraction system.

4.6.1 Simulation description

The ion source is mounted in a high voltage (HV) platform to generate a drift towards the extraction system, set at ground, where the electric field gradient pushes and confines the ions. In addition, as already explained, the DC electrical heating current flowing through the ion source creates a weak electric field that drives the ions axial movement. Thus, the electric field experienced by an ion is a superposition of the field produced by the DC electric heating current and the externally applied acceleration of the extraction system (fig. 4.16). Both modify the propagation behavior of the particles.

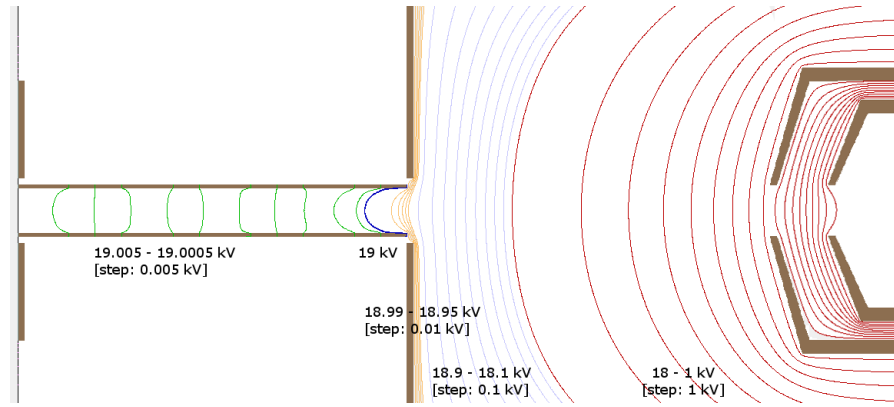


Figure 4.16– Picture of the equi-potential lines of the electric voltage generated by the resistive heating (5 V) and the high voltage (19 kV).

4.6.1.1 Simulation parts

A three-dimension ion-transport SIMION simulation has been developed (fig. 4.17). The simulation consist of an ionizer tube (figure A) and the extraction system (figure B) with one or two extraction electrodes. The ion source housing and the extraction system chamber were also simulated to avoid boundary effects (figure C). The ion source housing was set to high voltage and the extraction chamber to ground. The atomizer was not included because none laser ionized atom is expected there.

The ionizer tube was divided into 15 parts and each part was set to a voltage following a progressive potential all along the ionizer tube. The total voltage drop considered was 5.4 V for ionizers with 7 mm diameter and 4 V for 3 mm diameter ionizers. The voltage drop is added to the 19 kV extraction voltage (fig. 4.18). In addition, an extraction electrode was placed at the exit of the ion source to provide an electric field gradient that guide the ions towards the dipole mass spectrometer. It has an extraction hole of 8 mm diameter and it can be placed in a range of distances from 10 to 60 mm referenced to the ionizer exit.

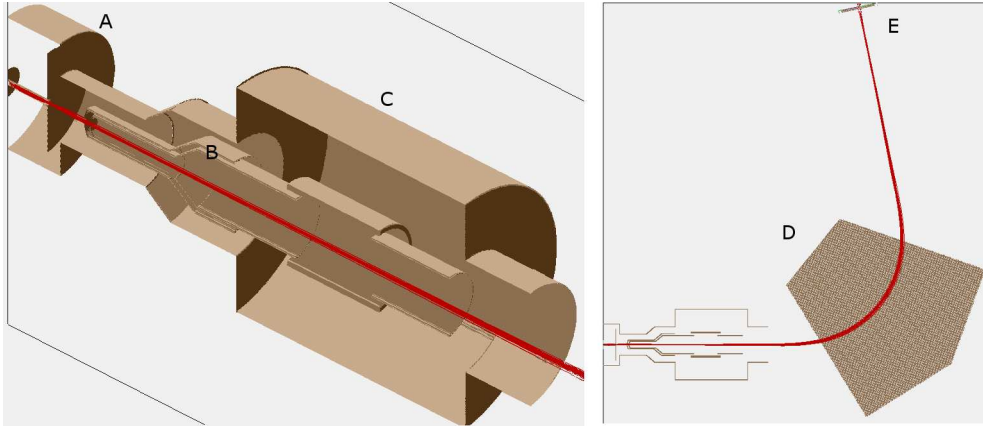


Figure 4.17– SIMION simulation geometries used to optimize the ion beam generation. The geometry consist of: (A) The ion source body, (B) the extraction system, (C) the ion source and extraction system housing, (D) the dipole magnetic mass spectrometer and (E) the detection point.

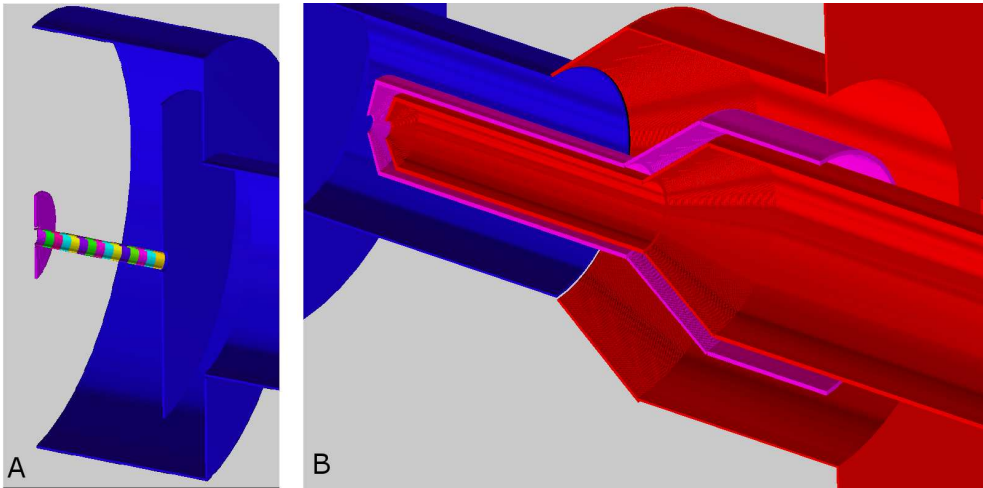


Figure 4.18– (A) The ionizer was divided into several parts to simulate the voltage drop. Each color corresponds to one of the 15 divisions. (B) Extraction system simulation with first extraction electrode (pink) and ground extraction electrode (red).

The dipole magnetic mass spectrometer was included in the simulation to analyze the energy dispersion (figure D), described in section 5.4. The dipole was generated using a simplified version of the CAD drawings. However, it was observed that the magnet focuses correctly in horizontal direction, but the vertical focusing was weaker than it should. For that reason, in some cases the simulation was performed without dipole, and only the horizontal emittance was considered. Finally the detection point was set either in the Faraday cup or in the low intensity profiler position (figure E).

SIMION divides the simulation space into potential arrays (PA). The resolution of the PA was set to 0.1 mm for the ion source and extraction system, 5 mm for the dipole and 5 mm for the detection. These parameters will not modify the precision of the results and at the same time the simulation will allow for more particles in less processing time to run.

4.6.1.2 Simulation parameters

The chosen isotope for the simulation was ^{124}Sn (mass 124) with +1 charge state. The generation of the ions inside the ion source was defined as a homogeneous cylindrical distribution (radial direction is considered) with diameter and length equal to the ion source, simulating the presence of atoms filling the tube when a laser pulse arrives.

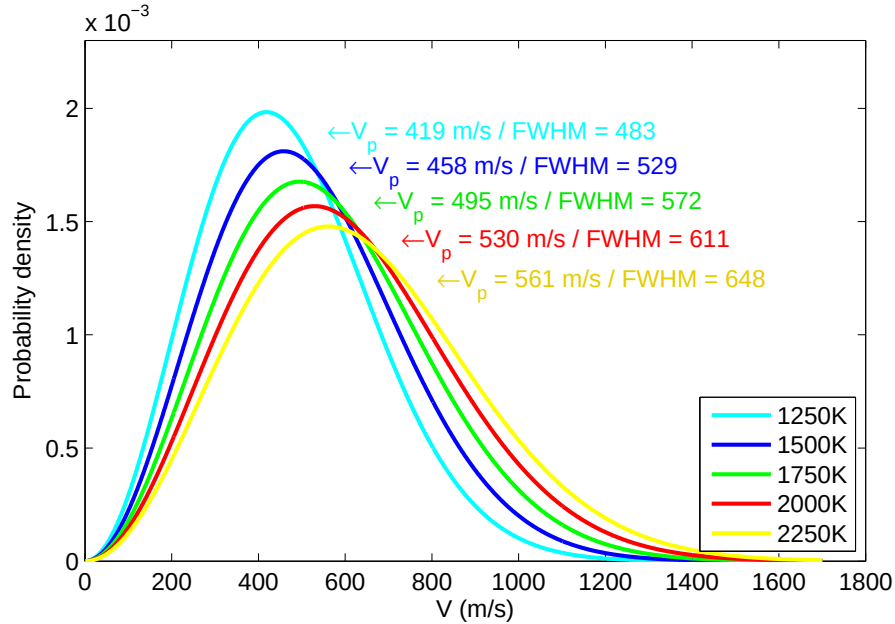


Figure 4.19– Maxwell-Boltzmann velocity distribution of Sn ions at different temperatures. The most probable speed for each distribution is indicated.

A Maxwell-Boltzmann velocity distribution for the ion initial speed was assumed. [Figure 4.19](#) shows the distributions for several temperatures and the most probable speed is indicated. Two configurations were used for the ion initial velocity direction, one with an isotropic distribution in 2π and another considering a 30 degree cone distribution. The latter was used to optimize the extraction system, due to the increase in the number of ions getting out of the ion source. The plasma potential which may prevent collisions with the walls was not included in the simulation model.

4.6.1.3 Output data

The simulation output is a file that includes the propagation information of each particle in the detection point (Faraday cup or profiler). Runs of 10000 to 50000 particles were performed to ameliorate the statistics. A typical information output is: ion number, Time of Flight (TOF) in μs , mass (amu), charge, position (x , y , z), azimuthal and elevation angles, velocity (v_x , v_y , v_z) and kinetic energy (eV) of the particle. With this information it is possible to know the number of ions that arrive to the detector and reconstruct the ion beam size and profile at the detection point, the time structure, calculate the emittance and estimate the energy dispersion. A MATLAB program was developed to analyze the simulation output.

4.6.2 First electrode distance optimization

Simulations with one extraction electrode have been performed to maximize the transport efficiency. In this case, LISBET ion source with geometry 7 mm diameter and 60 mm length ionizer was studied. The extraction electrode distances 15, 30, 45 and 60 mm were analyzed (the extraction electrode range covers from 10 to 60 mm). The ion source was set to 19 kV and the extraction electrode was set to ground (0 V). The initial velocity distribution of atoms was considered in a 30 degree cone distribution.

The results are shown in [figure 4.20](#) and [table 4.4](#). The distribution spread (A), the beam profile (B) and the angular distribution (C) in the detection point were analyzed. The circle in the distribution spread (A) marks the size of the Faraday cup sensor (30 mm). The angular distribution (C) can provide the emittance if the position is multiplied by the angle (in mm·mrad). The dipole mass spectrometer deflection was not considered in this simulation, but the total distance to the detector was taken into account.

Table 4.4 – Summary of the simulations performed with one extraction electrode at 19 kV.

Distance	Ion distribution	Profile maximum intensity	Angular distribution	Transport efficiency
15 mm	Max	100	112 mm·mrad	91.09%
30 mm		100	88 mm·mrad	94.05%
45 mm		150	50 mm·mrad	95.43%
60 mm	Min	250	15 mm·mrad	95.8%

Best results were obtained with 60 mm distance (IV), which showed highest peak intensity (250 counts) and smallest angular distribution (15 mm·mrad) (figures IV-B and IV-C). The angular distribution was symmetric for all simulations.

The transport efficiency was also recorded obtaining 91.1% for 15 mm, 94.1% for 30 mm, 95.4% for 45 mm and 95.8% for 60 mm. The ions are lost due to collisions with the ion source cavity walls or with the extraction electrodes. An increasing of lost ions was observed for small distances due to collisions with the extraction electrode.

4.6.3 Two step extraction optimization

Preliminary studies showed that an intermediate extraction electrode can improve the ion beam distribution. A double extraction electrode allows also to vary the potential of the intermediate electrode, instead of varying the single electrode position, for ion beam transport optimization. This is fundamental in the case of high radiation conditions where no motorization of extraction system is possible. The intermediate extraction electrode (also named first electrode) can be set to an electric potential between the ion source potential (19 kV) and the second extraction electrode potential (0 V).

Table 4.5 – Optimum electric potential values found for distances: 15, 30, 45 and 60 mm for the two electrodes configuration at 19 kV.

Distance	Electric potential (V)
15 mm	6500
30 mm	7500
45 mm	85000
60 mm	10000

The electric potentials of the first extraction electrode were optimized in the simulations for each distance, in order to obtain a small and homogeneous ion beam distribution (table 4.5). Only the optimized solutions are discussed here. The distances were measured between the exit of the ionizer and the first extraction electrode. The second extraction electrode was placed at a distance of 12 mm from the first extraction electrode. This distance was chosen due to preliminar ion beam measurements in the test bench.

Figure 4.21 and table 4.6 show the results of ion propagation with two extraction electrodes. A similar behavior as with single extraction electrode can be observed with an amelioration of the beam characteristics when the distance was increased. A decreasing of the distribution spread and angular distribution was observed for distances 45 mm (VII) and 60 mm (VIII). The angular distribution was symmetric for all simulations. 45 mm beam profile (VII-B) shows a maximum peak.

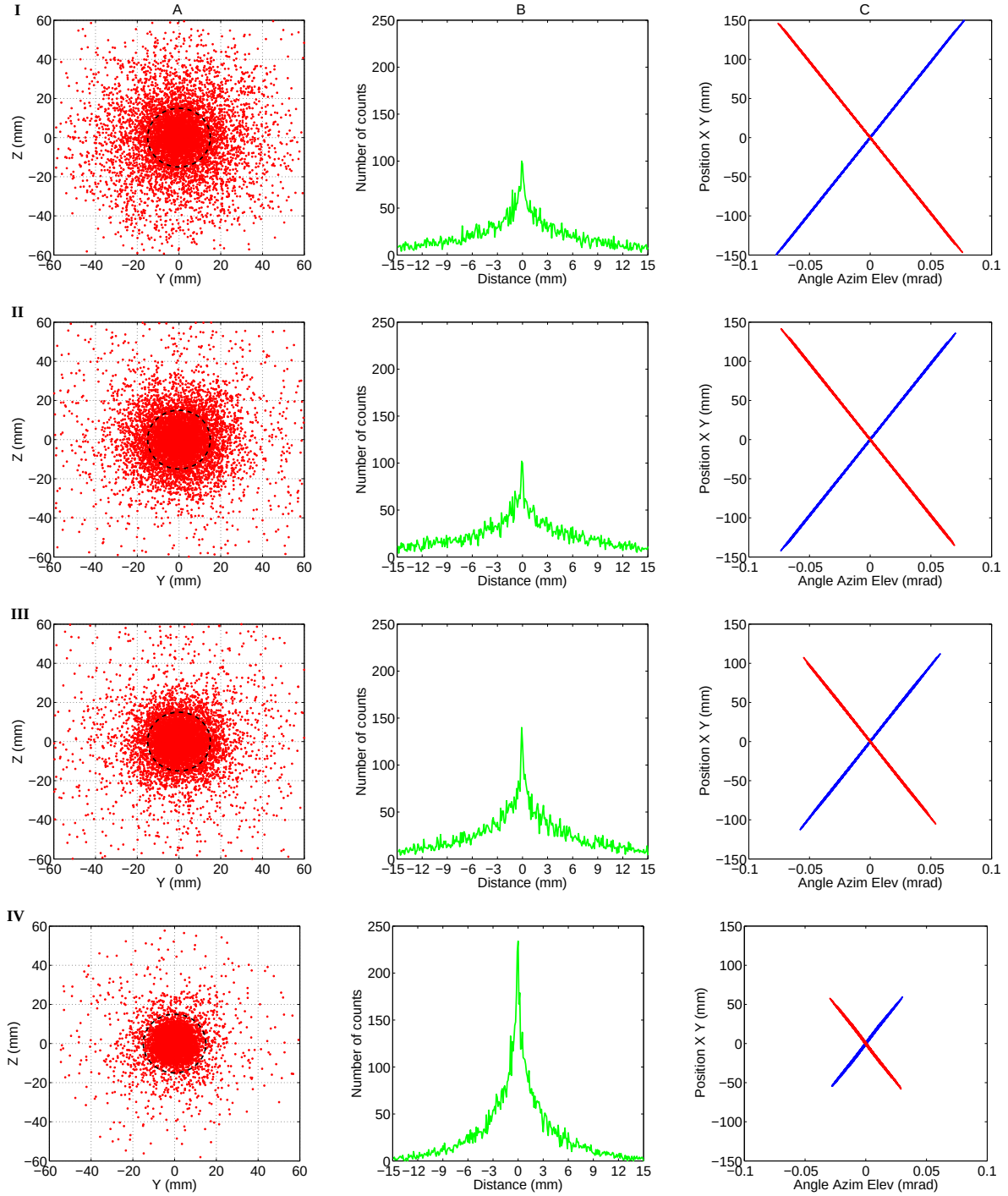


Figure 4.20– Simulation comparison for (I) 15 mm, (II) 30 mm, (III) 45 mm, and (IV) 60 mm distances with one extraction electrode. (A) The distribution spread, (B) the horizontal ion beam profile and (C) the angular distribution are shown. The dotted circle in the distribution spread corresponds to the size of the Faraday cup (30 mm diameter). The angular distributions are: x-azimuthal (red) and y-elevation (blue).

4.6. Extraction system optimization

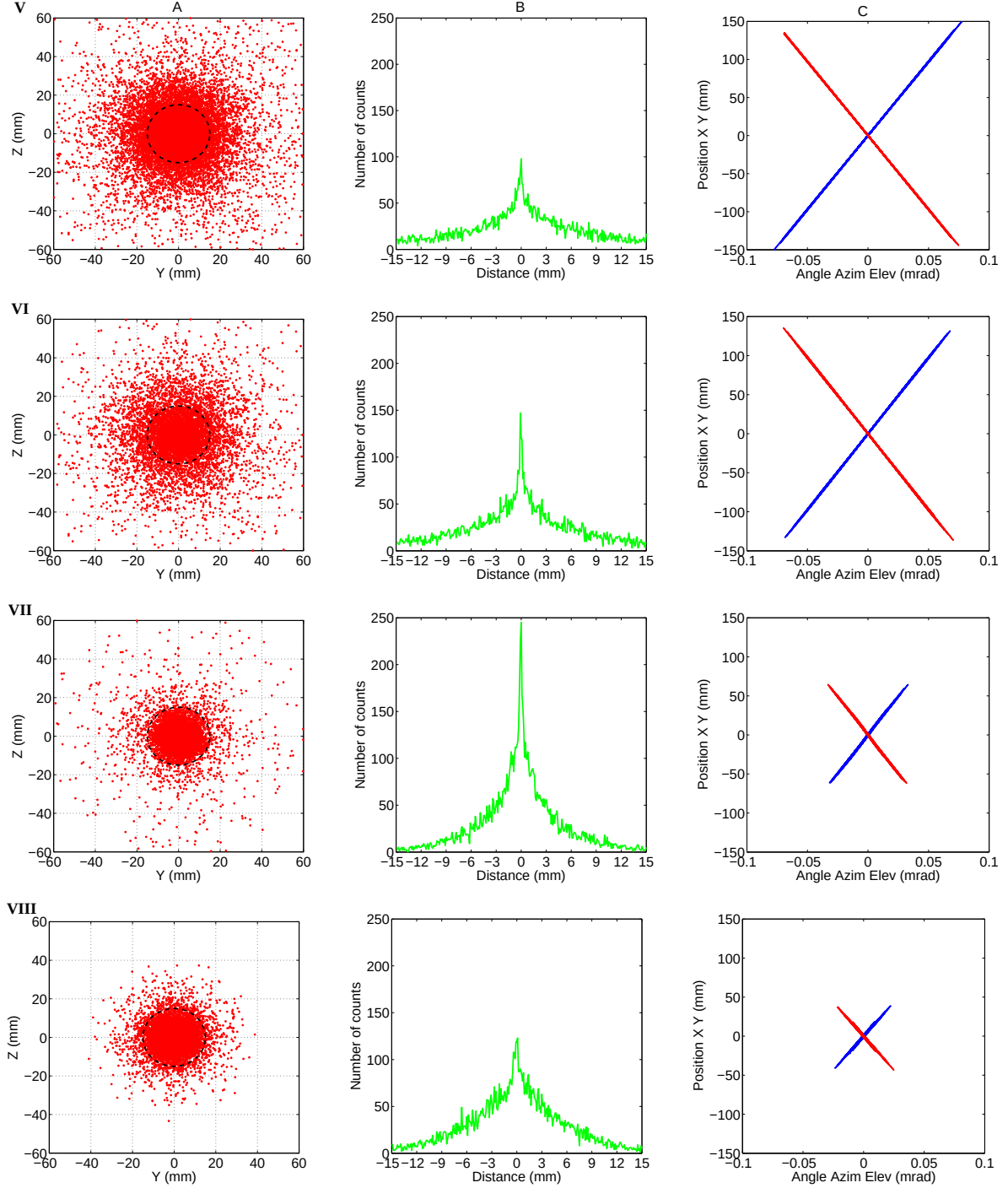


Figure 4.21– Simulation comparison for the distances (V) 15 mm, (VI) 30 mm, (VII) 45 mm, and (VIII) 60 mm with two extraction electrodes. The distribution spread (A), the beam profile (B) and the angular distribution (C) are shown. The Faraday cup size corresponds to the dotted circle in the distribution spread (30 mm diameter). The angular distributions are: x-azimuthal (red) and y-elevation (blue).

Table 4.6 – Summary of the simulations performed with two extraction electrodes.

Distance	Ion distribution	Profile maximum intensity	Angular distribution	Transport efficiency
15 mm	Max	100	105 mm·mrad	91.77%
30 mm		150	91 mm·mrad	96.18%
45 mm	Min	250	33 mm·mrad	95.94%
60 mm	Min	125	10 mm·mrad	96.24%

The simulations recorded $\sim 95\%$ of generated ions arriving to the detection with 30, 45 and 60 mm, with the exception of 15 mm distance option that recorded 91.7%. Similar to one extraction electrode, an increasing of lost ions was found for small distances due to collisions with the first electrode.

The best spatial distribution was established for a distance of 60 mm and 10 kV voltage, however the maximum intensity distribution close to the center was measured at 45 mm with 8.5 kV. On the other hand, the minimum angular distribution was found for 60 mm and 10 kV.

The conclusion is that the optimum parameters are in the range between 45 and 60 mm distances and between 8.5 and 10 kV voltages on the first extraction electrode (table 4.6).

4.6.4 Comparison with measurements

The simulations were compared to the experiment in the test bench with the same parameters. The Faraday cup was used to record the intensity of the ion signal and compare the different options. When the first extraction voltage is set to 0 V, it corresponds to the case using a single extraction electrode, because the second electrode does not affect the extraction field. The second extraction electrode was placed at 12 mm from the first electrode.

Figure 4.22 shows the measurements for a constant ion parameter (i.e. the intensity of the ion beam coming from the source was constant). Each measurement was repeated several times to avoid errors due to the ion source release. An optimum ion beam quality was found with the first extraction electrode placed at 55 mm from the ion source and a voltage of 9.5 kV (fig. 4.23 shows the equivalent simulation).

The optimum distance for the second extraction electrode was optimized directly in the test bench with a Sn ion beam obtaining an optimum signal at 9 mm between the first and the second electrodes.

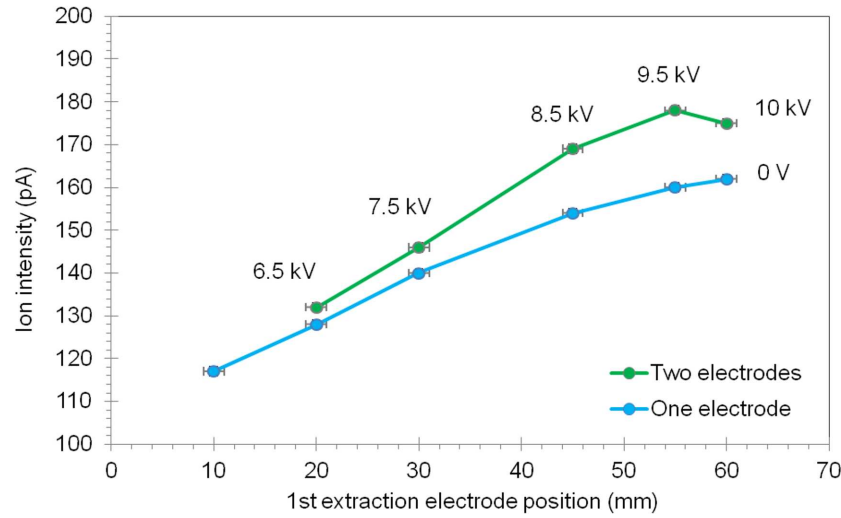


Figure 4.22– Sn ion current intensities generated at several distances of the first extraction electrode. One electrode configuration: First and second electrodes set to 0 V. Two electrodes configuration: First electrode at different voltages and second electrode set to 0 V.

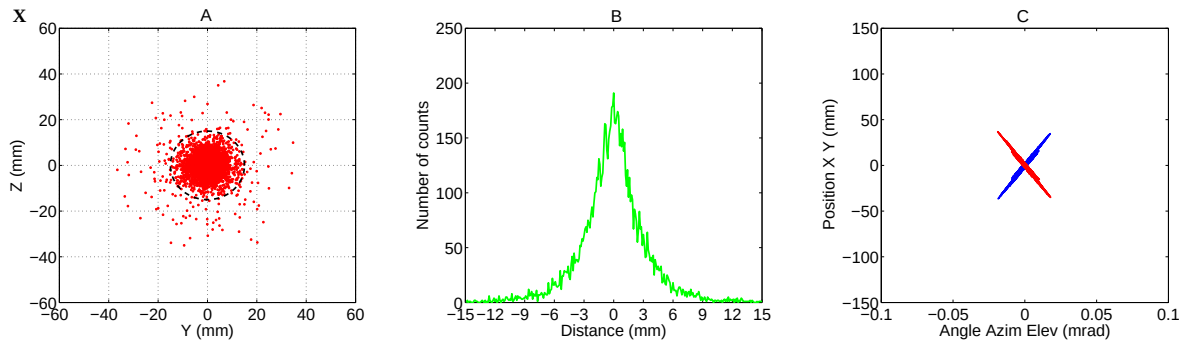


Figure 4.23– Optimized simulation parameters: 55 mm between the ion source and the first extraction electrode at 9.5 kV and 9 mm between the electrodes. (A) The distribution spread, (B) the beam profile and (C) the angular distribution are shown.

As conclusion, the extraction system was optimized with transport efficiencies close to 100% for the ions with initial velocity in a 30 degree cone distribution. The figure 4.24) shows a comparison between the ion beam profile obtained in the simulations and the one measured in the test bench with the optimized parameters. Most of the ions will arrive to the detector minimizing the transport error for the efficiency measurements.

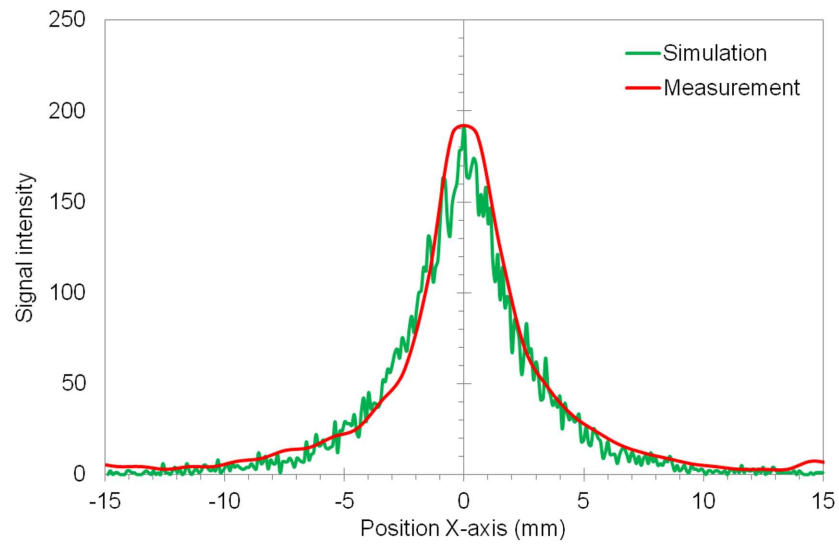


Figure 4.24– Comparison between the simulated ion beam profile and the profile measured with the low intensity profiler.

Chapter 5

Ion source geometry study

5.1 Geometry comparison as a function of the efficiency

The study of the ion source geometries is a fundamental part of this thesis. The objective is to find the solution that can provide the highest efficiency while keeping the ion beam quality. The effect of the ion source geometry has not been studied yet, and most of the laboratories use a conventional ion source configuration: An ionizer tantalum tube with 3 mm diameter and 35 mm length. In section 4.1 four different options (two diameters, two lengths) were described in order to be studied. The code of each ion source described there will be used (table 4.1), where for example LISBET-D7L60 stands for an ion source with 7 mm diameter and 60 mm length ionizer. In this section, LISBET-D7L35, LISBET-D7L60, LISBET-D3L35, LISBET-D3L60 and LISBET-D7L35 with the ZrC insert (3 mm of inner diameter) were compared by means of efficiency measurements.

5.1.1 Efficiency measurement

The ion production efficiency can be calculated as the total integrated ion current compared to the amount of atoms in the sample. The ratio between the total number of ions measured after the magnet mass spectrometer (K_{Total}) and the known quantity of atoms of interest in the atomizer (N_{iso}) provides the efficiency (ε_{ff}):

$$\varepsilon_{ff} = \frac{K_{Total}}{N_{iso}} \quad (5.1)$$

This efficiency includes the effusion efficiency of the atoms before ionization ($\varepsilon_{effusion}$), the efficiency of the photo-ionization process ($\varepsilon_{photoion}$) and the transport efficiency of ions through the extraction system and the magnet mass spectrometer ($\varepsilon_{transport}$):

$$\varepsilon_{ff} = \varepsilon_{effusion} \cdot \varepsilon_{photoion} \cdot \varepsilon_{transport} \quad (5.2)$$

$\varepsilon_{transport}$ can be considered close to 100% due to the optimization of the test bench described before (section 4.6). Therefore the measured total efficiency represents the product of $\varepsilon_{effusion}$ and $\varepsilon_{photoion}$.

The ion source was gradually heated until the sample was completely released, and further heating did not increase the ion current. The current was continuously recorded until it decreased below 10 pA. Figure 5.1 shows an example of the total current for an efficiency measurement, where the peaks correspond to rises of temperature. The method used to quantify the atoms in the sample and the detected ions will be described in the following sections.

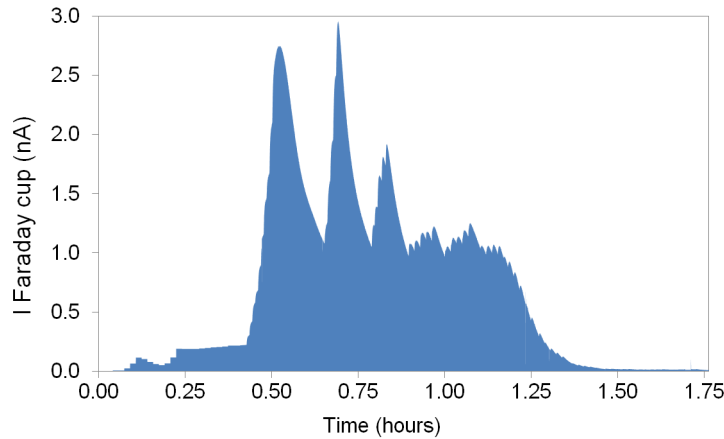


Figure 5.1– Efficiency measurement example. The ion current is accumulated during the experiment and the integral of the signal gives the total number of generated ions.

5.1.1.1 Sample quantification

The quantity of atoms in a sample can be calculated from the concentration and the known volume of the element. For the sample preparation two options were used, which consisted of a diluted sample and a sample without dilution. These methods to calculate the number of atoms in these two cases will be discussed in the following lines.

The following notation will be used:

- Q the concentration of the element of interest in the nitric acid solution in grams per liter.
- V_E the volume of solute in liters.
- V_S the volume of the solvent (in our case distilled water) in liters.
- V_T the volume of the dissolution ($V_E + V_S$) in liters.
- D the dissolution factor.
- m the mass in grams.
- M_A the atomic mass of the element.
- N_A the Avogadro's number.
- n the natural abundance of the chosen isotope.
- N_{atom} the total number of atoms in the sample.
- N_{iso} the proportional number of isotopes.

For a **non diluted** sample the total number of atoms included in the sample corresponds to:

$$N_{atom} = \frac{Q \cdot V_E \cdot N_A}{M_A} \quad (5.3)$$

And the error associated to this quantity is:

$$\Delta N_{atom} = N_{atom} \cdot \sqrt{\left(\frac{\Delta Q}{Q}\right)^2 + \left(\frac{\Delta V_E}{V_E}\right)^2 + \left(\frac{\Delta M_A}{M_A}\right)^2} \quad (5.4)$$

For a **diluted sample** the dissolution is defined as:

$$D = \frac{V_E}{V_E + V_S} \quad (5.5)$$

where the error of the dissolution is:

$$\Delta D = D \cdot \sqrt{\left(\left(\frac{V_S}{V_E + V_S}\right) \Delta V_E\right)^2 + \left(\frac{\Delta V_S}{V_E + V_S}\right)^2} \quad (5.6)$$

Then the total number of atoms in the diluted sample can be calculated as:

$$N_{atom} = \frac{Q \cdot V_E \cdot D}{M_A} \cdot N_A = \frac{Q \cdot V_E}{M_A} \cdot \left(\frac{V_E}{V_E + V_S}\right) \cdot N_A \quad (5.7)$$

where the error of the total number of atoms in the sample can be calculated as:

$$\Delta N_{atom} = N_{atom} \cdot \sqrt{\left(\frac{\Delta Q}{Q}\right)^2 + \left(\frac{\Delta V_E}{V_E}\right)^2 + \left(\frac{\Delta M_A}{M_A}\right)^2 + \left(\frac{\Delta D}{D}\right)^2} \quad (5.8)$$

In this expression, $\Delta M_A/M_A$ can be considered close to 0. Finally the **total number of isotopes** that can be ionized for a chosen element is:

$$N_{iso} = N_{atom} \cdot n \quad (5.9)$$

with an error:

$$\Delta N_{iso} = N_{iso} \cdot \sqrt{\left(\frac{\Delta N_{atom}}{N_{atom}}\right)^2 + \left(\frac{\Delta n}{n}\right)^2} \quad (5.10)$$

As an example, a sample of 0.5 μl of standard Sn solution (118.71 atomic mass) with a concentration of 1 g/l contains $2.54 \cdot 10^{15} \pm 4.57 \cdot 10^{14}$ atoms of Sn. Therefore, the quantity of ^{124}Sn can be obtained from the total quantity by considering the relative natural abundance (5.79%). In this case, the sample contains $1.47 \cdot 10^{14} \pm 2.64 \cdot 10^{13}$ ^{124}Sn atoms.

5.1.1.2 Detected ions

The acquisition system records the ion current (I) in amperes, which is measured in a Faraday cup. The ion current can be divided by the electric charge to obtain the quantity of ions detected per second (K_{ion}) as seen in equation 5.11, where q is the electric charge of the particles ($1.602176 \cdot 10^{-19}$ C) for a monocharged ion:

$$K_{ion} = \frac{I}{q} \quad (5.11)$$

The integrated quantity of atoms during the experiment gives the total quantity of ions detected in a range of time (K_{Total}). This number represents the ions successfully ionized, extracted and transported to the detector (equation 5.12), where t represents the duration of the experiment in seconds.

$$K_{Total} = \sum_{i=0}^t K_{ion} \quad (5.12)$$

The error in the amount of ions measured by the Faraday cup can be calculated as:

$$\Delta K_{ion} = K_{ion} \sqrt{\left(\frac{\Delta I}{I}\right)^2 + \left(\frac{\Delta q}{q}\right)^2} \quad (5.13)$$

$$\Delta K_{Total} = \sum_{i=0}^t K_{Total} \sqrt{\left(\frac{\Delta K_{ion}}{K_{ion}}\right)^2} \quad (5.14)$$

In equation 5.13, the quantity $\Delta q/q$ can be supposed close to zero.

5.1.1.3 Total efficiency

The total efficiency ε_{ff} can be calculated as the amount of ions measured by the Faraday cup (K_{Total}) divided by the total number of atoms in the atomizer for a chosen isotope (N_{iso}).

$$\varepsilon_{ff} = \frac{K_{Total}}{N_{iso}} \cdot 100 \quad (5.15)$$

The error in the efficiency is:

$$\Delta \varepsilon_{ff} = \varepsilon_{ff} \sqrt{\left(\frac{\Delta N_{iso}}{N_{iso}}\right)^2 + \left(\frac{\Delta K_{Total}}{K_{Total}}\right)^2} \quad (5.16)$$

5.1.2 Sample preparation

A stable atomic sample is needed in order to perform off-line studies. The sample has to have a minimum amount of atoms to provide enough atomic flux through the interaction device, and to be able to accomplish reliable efficiency measurements. At the same time, it should not be too big in order to avoid a huge atomic release that can pollute the experimental chamber. An ideal sample should last for a one day experiment, because this allows the repetition of experiments and minimizes the impact of failed measurements. The knowledge of the optimum sample size will be fundamental to perform off-line measurements with radioactive long lived isotopes due to the quantity regulations of these isotopes.

The sample is generated by the deposition in a metallic foil of a drop of liquid solution (nitric acid) containing a calibrated quantity of the atoms of interest. The liquid is then evaporated (different evaporation techniques were tested, which are explained below). The calibrated quantity of the atoms of interest stays as dry products on the metallic foil, which can be folded and placed in the atomizer.

Two micro-pipettes were used in order to deposit the required volume of liquid solution in the metallic foil: a 1-100 μl micro-pipette for diluted samples (to reduce the quantity of atoms), and a 0.1-2 μl micro-pipette for non-diluted samples. Non-diluted samples were preferred to reduce the errors due to dilution. Tests were first performed with a titanium (Ti) foil. However, the composition of commercial titanium has a certain percentage of impurities, such as 200 ppm of tin, which may diffuse out at high temperatures and increase the quantity of atoms of interest. A tantalum (Ta) foil, containing only 2 ppm of tin, was used later on instead of the titanium foil. Due to the chemical characteristics of tantalum, the release temperature of the atoms (i.e. the temperature at which the first signal is observed) was decreased and the temperature needed for a complete evaporation of the sample was also reduced. In table 5.1, the impurities present in commercial tantalum and titanium are compared.

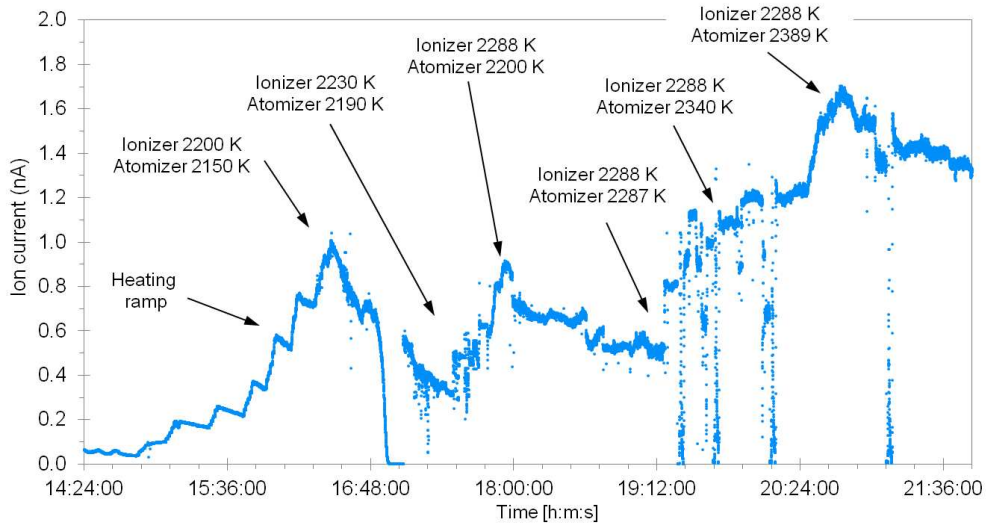


Figure 5.2– Example of Sn background signal at temperatures higher than 2000 K after an efficiency measurement. The sample was almost finished at 14:30 (2000 K) and a second heating ramp was performed, which produced an increase of the ion current. The temperature of each step is indicated.

In the case of Sn ion production, it was observed that tin diffuses out of the metallic surfaces (LISBET and the foil) at temperatures above 2000 K. An example of a background signal can be found in figure 5.2. This signal comes from the inherent Sn impurities of the structural ion source materials, since the diffusion process is activated at high temperatures. In order to avoid contamination from the ion source a limit of 2000 K was set for the experiments. The typical contamination elements composing an ion source made of tantalum and titanium can be found in table 5.1.

For the liquid drop evaporation, the sample was heated in vacuum at $\sim 100^\circ\text{C}$ in an oven. It was observed that the liquid solution created bubbles due to the vapor pressure and, when the

5.1. Geometry comparison as a function of the efficiency

Table 5.1 – Typical contamination elements that can be found in a commercial titanium (Ti) and tantalum (Ta) [202].

Element	Z	Impurities	
		Titanium	Tantalum
Na	11		
Al	13	300 ppm	5 ppm
K	19		
Ca	20	20 ppm	2 ppm
Ti	22		20 ppm
Mn	25	100 ppm	2 ppm
Fe	26	1500 ppm	30 ppm
Co	27		1 ppm
Cu	29	5 ppm	2 ppm
Nb	41		<500 ppm
Sn	50	200 ppm	2 ppm

bubbles exploded, a partial quantity of the sample was lost. This effect decreased the reliability of the samples. For this reason, the samples were dried in air about 2 hours until the complete evaporation of solvent was achieved. Once evaporated, the sample was folded in a cylinder shape.

Finally, the sample was placed inside the LISBET transfer tube in order to use it as atomizer. The extreme of the atomizer was closed with a tantalum cap to prevent the sample to fall down. The cap was tight to limit the escape of atoms. An atom flux was achieved by resistively heating the atomizer from 1400 K to 2000 K. Samples containing between 10^{13} to 10^{15} atoms of Zn, Rb and Sn were prepared by this method. The calculation of the quantity of atoms included in one sample is fundamental for a proper calculation of the efficiency equation (section 5.1.1). The description of the calibrated solutions can be found in table 5.2.

Table 5.2 – Samples used to produce an atomic vapor inside the hot-cavity. Zn and Sn were laser ionized and Rb was surface ionized.

Isotope of interest	Quantity μl	Concentration g/l	Dissolution	Total atoms of the isotope of interest	Error
^{64}Zn	8	1	1:10	$3.58 \cdot 10^{15}$	$7 \cdot 10^{13}$
^{124}Sn	8	1	1:10	$2.4 \cdot 10^{14}$	$3 \cdot 10^{13}$
^{124}Sn	10	1	1:10	$2.9 \cdot 10^{14}$	$4 \cdot 10^{13}$
^{124}Sn	10	1	1:20	$1.5 \cdot 10^{14}$	$2 \cdot 10^{13}$
^{124}Sn	0.5	1	-	$1.5 \cdot 10^{14}$	$3 \cdot 10^{13}$
^{85}Rb	10	0.05	1:50	$5.1 \cdot 10^{13}$	$4 \cdot 10^{12}$
^{85}Rb	1	1	1:20	$2.5 \cdot 10^{14}$	$2 \cdot 10^{13}$

5.1.3 Background reduction for efficiency measurements

In addition to the background signal from the ion source impurities described in the previous section, the residual Sn from the sample can also generate a background signal that could affect the efficiency measurement. A study was performed to characterize the influence of this Sn background signal over the total measured ion current. After an efficiency measurement, it was observed that the background signal without sample could increase up to hundreds of pA at temperatures close to 2000 K.

The Sn background signal was followed closely and it slowly decreased after several days of heating. The amount of remaining signal diminished up to stabilize to a minimum quantity at a certain temperature (fig. 5.3). It was observed that the background followed an exponential distribution when none sample was inserted into the ion source. A study was performed to characterize the evolution of the background signal by using a 7 mm ionizer tube diameter, in order to remove it from the efficiency calculation for all ion source geometries.

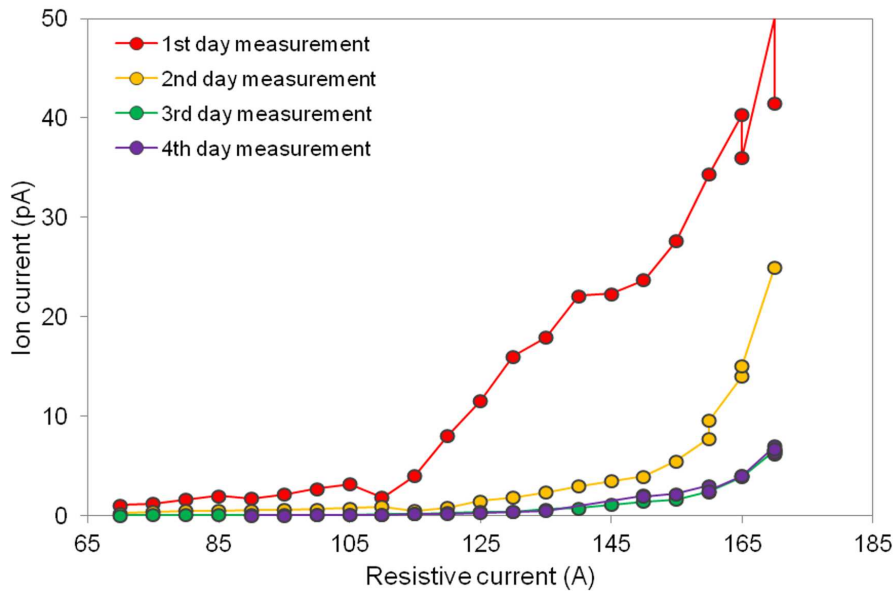


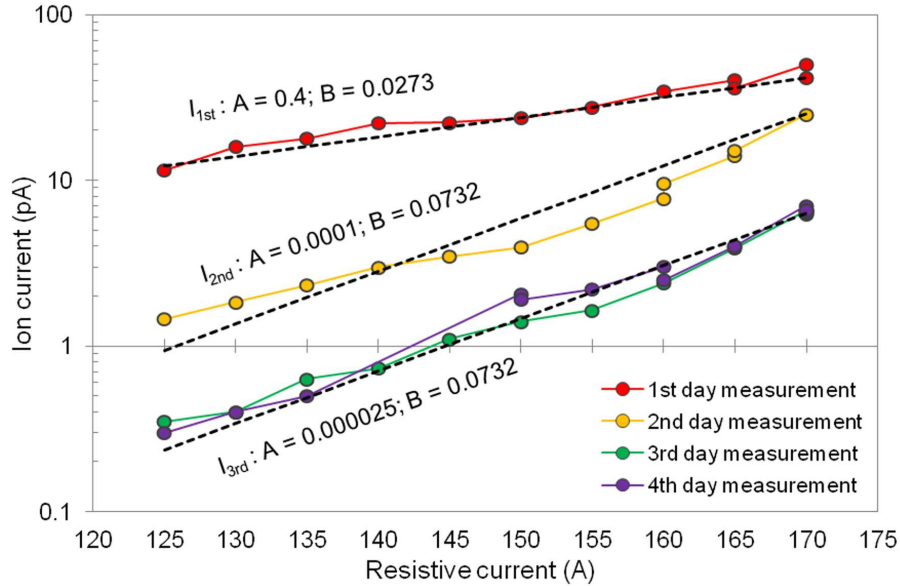
Figure 5.3– Ion current measured without sample in several consecutive experiments. A decreasing signal was observed for the measurements performed the first (red dots) and second (yellow dots) days. A stabilization of the signal can be observed in the third (green dots) and fourth (purple dots) measurements. In the cases where two different values of the ion current are provided for the same resistive current, they correspond to measurements performed at different times.

5.1. Geometry comparison as a function of the efficiency

An exponential distribution was used in order to estimate the remaining quantity of Sn background before an efficiency measurement with a new sample. This distribution followed the equation:

$$I = A \cdot \exp(B \cdot C) \quad (5.17)$$

where I is the ion current detected in the Faraday cup, A and B are fitting parameters and C is the resistive heating current set to the ion source in amperes. In [figure 5.4](#) the fitting curves used to fit the experimental data are presented. The parameter B converged after several days of heating to a value close to 0.0732.



[Figure 5.4](#)– Fitting of the ion current signal background for different experiments. A convergence in the B parameter can be observed after the second day of measurements.

A cleaning routine was applied before an efficiency measurement to reduce and control the background signal: The ion source was heated without sample, and the ion current signal was recorded. The remaining sample inside the ion source runs out and the signal decreases to a certain value. An estimation of the remaining background is performed by means of the last recorded value. Finally, a background correction is applied to the efficiency measurement, by removing the estimated background at each temperature from the measured ion current.

[Figure 5.5](#) shows a test of the background correction. The cleaning routine is represented as red dots. The presence of several ion signals for the same current is due to the decreasing of the signals with time.

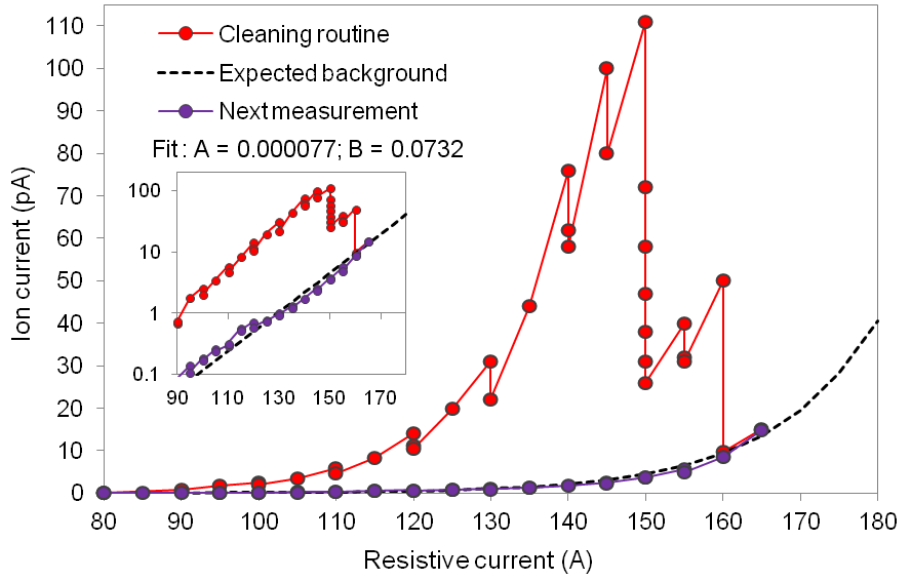


Figure 5.5– Example of background calculation for efficiency experiments. The last point of the cleaning routine (red dots) is fitted with the calculated parameters $A = 0.000077$ and $B = 0.0732$ (discontinuous line) and the purple dots show the next measurement. A logarithmic scale is shown in the small figure.

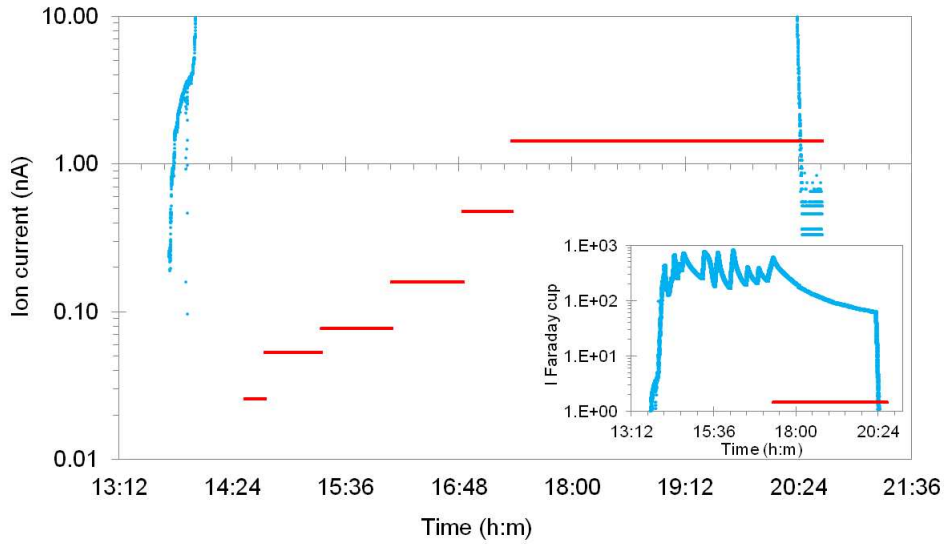


Figure 5.6– Example of background correction for an efficiency measurement. The ion current (blue) and the background correction (red) are shown as a function of time. The last value from the cleaning routine gave an exponential background fitting with parameters $A = 2.45 \cdot 10^{10}$ and $B = 0.0732$. The values given by the exponential are removed for each current heating value (red). The efficiency in this case decreased from 24.44% to 24.33%.

These points correspond to measurements performed at different times. Finally, the last point of the measurement was fitted by equation 5.17, as described before (dashed line). In order to analyze the accuracy of the fit, the measurement was repeated in the same conditions (purple points). It is possible to observe that the fit reproduced perfectly the expected background.

The background behavior as a function of heating current was estimated from the last point recorded during the cleaning routine. The estimated background was then removed from the following measurements with a new sample. In some cases, the background correction was applied retroactively for the efficiency measurements without cleaning procedure using the last point recorded in the previous measurement.

Figure 5.6 shows an example of the background correction for an efficiency measurement. In most of the cases the difference was included in the efficiency error, because the samples were finished before 2000 K, thus the correction was small. Nevertheless, the background correction was performed for all Sn efficiency measurements.

5.1.4 Experiment requirements

In order to study the efficiency of the different geometry options, a sample of natural Sn atoms (table 5.2) was placed inside the atomizer. The efficiency measurements were repeated at least three times and several sample sizes were also tested for reproducibility. The errors include sample preparation error, detection error and deviation error due to the different measurements. Before an efficiency measurement, the alignment and the output power of each laser was verified. Figure 5.7 shows a comparison of the saturation measurements performed with two different diameter ionizers, 7 and 3 mm diameter at 2000 K. The diameter of the laser beam corresponds to the diameter of the ionizer. Both diameters show the same saturation behavior.

5.1.5 Efficiency results for different LISBET geometries

Tables 5.3 and 5.4 gather the efficiency measurements for the different LISBET configurations and figure 5.8 shows examples of the total ion currents measured for each LISBET geometry.

An overall efficiency of $24 \pm 3\%$ was obtained for the **LISBET-D7L35** (ion source with 7 mm diameter ionizer and 35 mm length) from three samples containing $2.9 \cdot 10^{14}$ and $1.5 \cdot 10^{14}$ ^{124}Sn atoms. Diluted and non-diluted samples were also tested. The background correction was directly removed from the raw signal. Two measurements (#7 and #9) failed due to errors in the cap fitting.

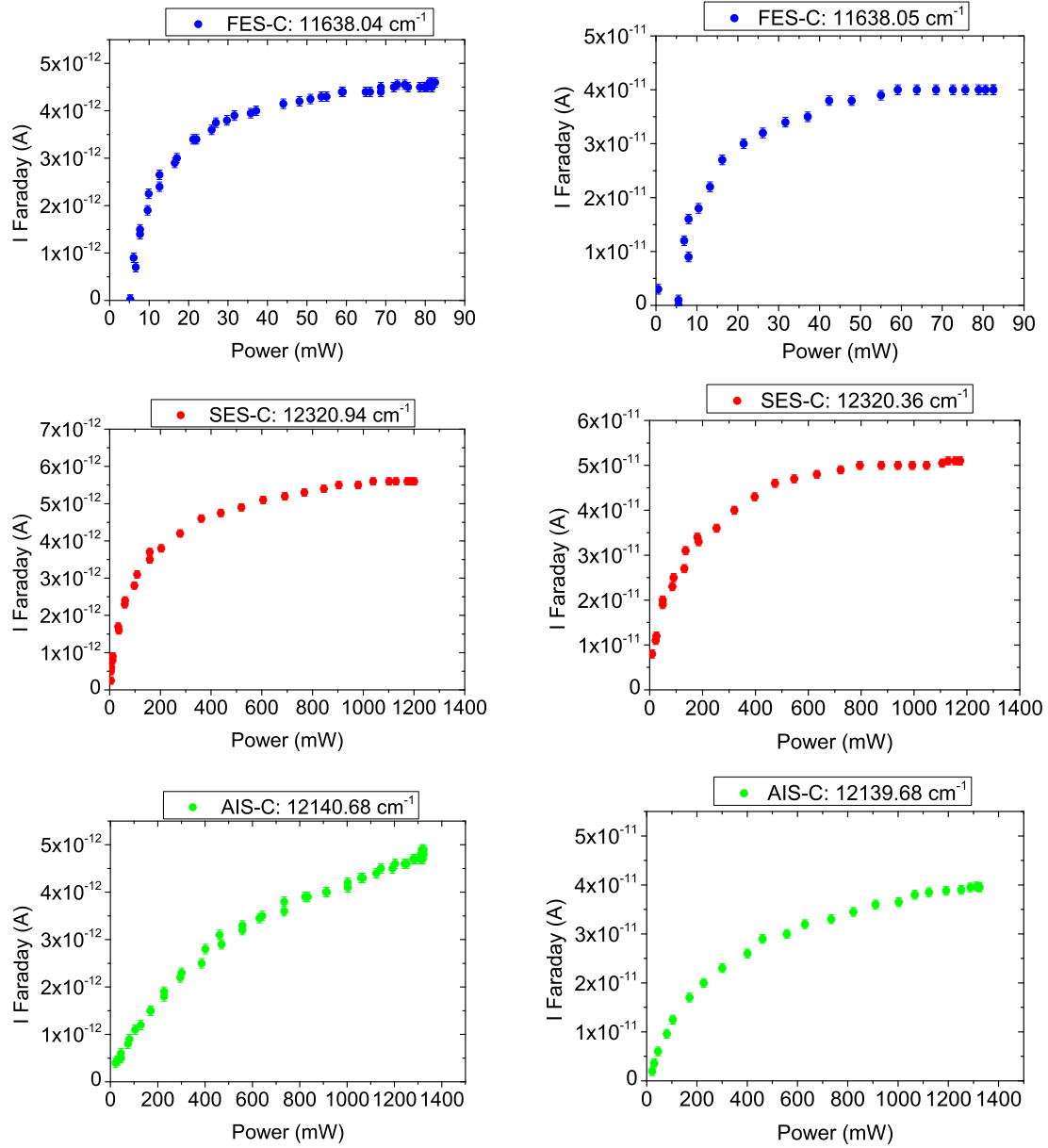


Figure 5.7– Saturation measurements performed with the Sn three-step scheme for LISBET-D7L35 (left column) and LISBET-D3L35 (right column). Both show a similar behavior where the first and second transitions are completely saturated and the third transition is not completely saturated.

5.1. Geometry comparison as a function of the efficiency

Table 5.3 – Efficiency results for the studied LISBET configurations. The red arrows indicate the heating current direction. The size of the laser beam corresponds to the size of the ionizer diameter with the exception of measurement #12. ε_{raw} is the efficiency directly recorded. ε_{bkgd} is the efficiency after background correction. An asterisk (*) indicates that the background correction was performed retroactively without cleaning. ε_{mean} is the mean efficiency of the measurements.

LISBET-D7L35

	Code	Sample (μ l)	Number of atoms	ε_{raw}	ε_{bkgd}	Error	Comments	ε_{mean}	Mean error
	#7	10 (1:10)	$2.9 \cdot 10^{14}$	14.3%	-	-	Sample failed	24%	$\pm 3\%$
	#8	10 (1:10)	$2.9 \cdot 10^{14}$	25.1%	24.8%*	$\pm 3\%$			
	#9	10 (1:10)	$2.9 \cdot 10^{14}$	16.4%	-	-	Sample failed		
	#10	10 (1:20)	$1.5 \cdot 10^{14}$	24.4%	24.3%	$\pm 3\%$			
	#11	0.5	$1.5 \cdot 10^{14}$	24.0%	23.9%	$\pm 4\%$			
	#12	0.5	$1.5 \cdot 10^{14}$	9.5%	9.5%	$\pm 2\%$	3 mm laser spot	10%	$\pm 2\%$

LISBET-D7L60

	Code	Sample (μ l)	Number of atoms	ε_{raw}	ε_{bkgd}	Error	Comments	ε_{mean}	Mean error
	#1	0.5	$1.5 \cdot 10^{14}$	10.7%	-	-	Misalignment	19%	$\pm 4\%$
	#2	0.5	$1.5 \cdot 10^{14}$	18.4%	18.2%*	$\pm 4\%$			
	#3	0.5	$1.5 \cdot 10^{14}$	18.6%	18.4%*	$\pm 4\%$			
	#4	0.5	$1.5 \cdot 10^{14}$	17.9%	17.8%	$\pm 4\%$			
	#5	0.5	$1.5 \cdot 10^{14}$	12.1%	-	-	Misalignment		
	#6	0.5	$1.5 \cdot 10^{14}$	22.4%	22.3%	$\pm 4\%$			

LISBET-D3L35

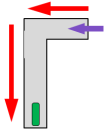
	Code	Sample (μ l)	Number of atoms	ε_{raw}	ε_{bkgd}	Error	Comments	ε_{mean}	Mean error
	#17	0.5	$1.47 \cdot 10^{14}$	6.2%	6.2%	$\pm 2\%$		7%	$\pm 2\%$
	#18	0.5	$1.47 \cdot 10^{14}$	7.0%	6.9%	$\pm 2\%$			
	#19	0.5	$1.47 \cdot 10^{14}$	4.4%	-	-	Sample failed		
	#20	0.5	$1.47 \cdot 10^{14}$	6.5%	6.4%	$\pm 1\%$			

LISBET-D3L60

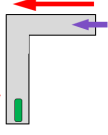
	Code	Sample (μ l)	Number of atoms	ε_{raw}	ε_{bkgd}	Error	Comments	ε_{mean}	Mean error
	#13	0.5	$1.5 \cdot 10^{14}$	7.8%	7.6	$\pm 2\%$		8%	$\pm 2\%$
	#14	0.5	$1.5 \cdot 10^{14}$	7.2%	7.1%	$\pm 2\%$			
	#15	0.5	$1.5 \cdot 10^{14}$	10.7%	9.7%	$\pm 2\%$			
	#16	0.5	$1.5 \cdot 10^{14}$	6.9%	6.8%	$\pm 2\%$			

Table 5.4 – Efficiency results for the studied LISBET configurations. (continuation).

LISBET-D7L35 (inversed polarity)

	Code	Sample (μl)	Number of atoms	ε_{raw}	$\varepsilon_{\text{bkgd}}$	Error	Comments	$\varepsilon_{\text{mean}}$	Mean error
	#32	10 (1:10)	$1.5 \cdot 10^{14}$	4.4%	4.0%	$\pm 1\%$		5%	$\pm 1\%$
	#33	10 (1:10)	$1.5 \cdot 10^{14}$	5.1%	5.0%	$\pm 1\%$			

LISBET-D7L60 (inversed polarity)

	Code	Sample (μl)	Number of atoms	ε_{raw}	$\varepsilon_{\text{bkgd}}$	Error	Comments	$\varepsilon_{\text{mean}}$	Mean error
	#24	8 (1:10)	$3.58 \cdot 10^{15}$	3.9%*	3.6%	$\pm 1\%$		4%	$\pm 1\%$
	#25	10 (1:10)	$2.9 \cdot 10^{14}$	2.2%*	-	-	Sample failed		
	#26	10 (1:10)	$2.9 \cdot 10^{14}$	2.0%*	-	-	Sample failed		
	#27	10 (1:10)	$2.9 \cdot 10^{14}$	5.3%*	5.2%	$\pm 1\%$			
	#28	10 (1:10)	$2.9 \cdot 10^{14}$	2.4%*	-	-	Sample failed		
	#29	10 (1:10)	$2.9 \cdot 10^{14}$	3.6%*	3.4%	$\pm 1\%$			
	#30	10 (1:10)	$2.9 \cdot 10^{14}$	4.5%*	4.2%	$\pm 1\%$			
	#31	10 (1:10)	$2.9 \cdot 10^{14}$	3.9%*	3.7%	$\pm 1\%$			

5.1. Geometry comparison as a function of the efficiency

The efficiency of the **LISBET-D7L60** configuration (7 mm diameter and 60 mm length ionizer) resulted to be $19 \pm 4\%$ from four samples containing $1.5 \cdot 10^{14}$ atoms. The background correction and the data dispersion increased the total error. It was also observed that laser misalignments could reduce the efficiency to 12%.

A total efficiency of $7 \pm 2\%$ was measured for **LISBET-D3L35** (3 mm diameter and 35 mm length ionizer). The background correction was small and thus the efficiency error decreased. The measurement #19 failed due to a failure of the recording system.

Finally, an efficiency value of $8 \pm 2\%$ was obtained for **LISBET-D3L60** (3 mm diameter and 60 mm length ionizer).

In addition, two complementary measurements were performed. In the first one, laser beams were focused to a three millimeters spot size with **LISBET-D7L35**. An efficiency of $10 \pm 2\%$ was obtained (measurement #12). In the second, a test to observe the effect of the electric field drift (which will be used in section 5.5) was performed for **LISBET-D7L35** and **LISBET-D7L60** where the polarity was inverse for repelling all ions backwards. A decrease of the efficiency to $5 \pm 1\%$ was obtained (measurements #24 - #33).

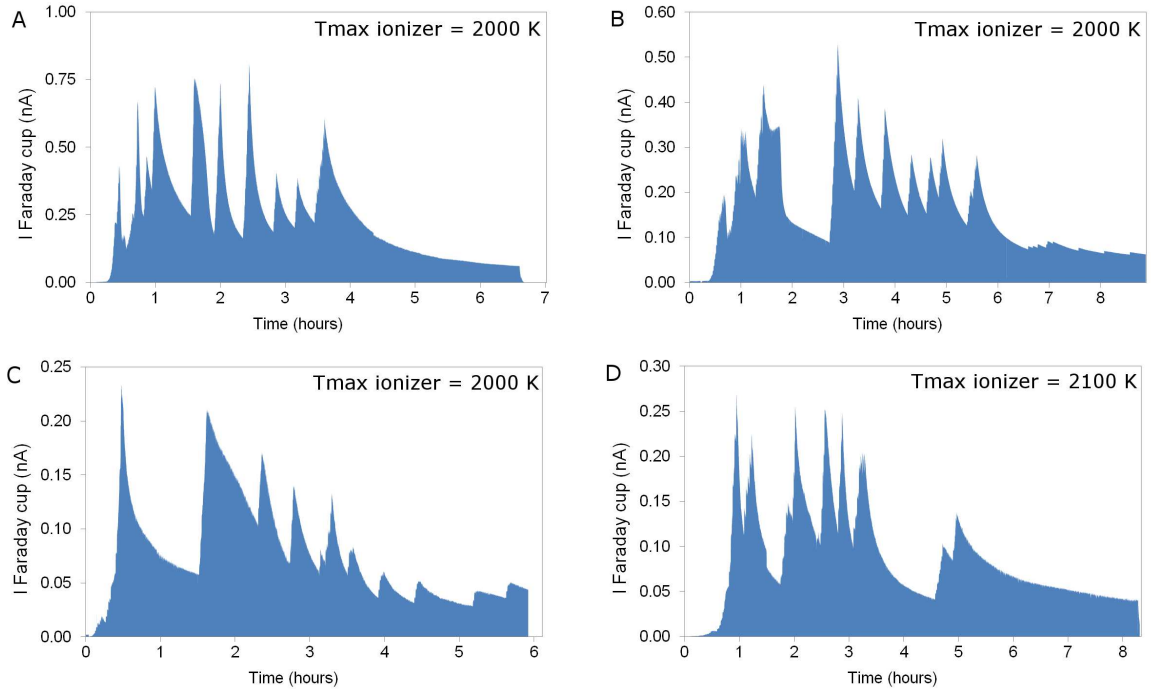


Figure 5.8– Ion current accumulated with (A) LISBET-D7L35, (B) LISBET-D7L60, (C) LISBET-D3L35 and (D) LISBET-D3L60 ion sources for ^{124}Sn efficiency measurements.

5.1.5.1 Simulations

Four different geometries have been studied with the SIMION simulation described in chapter 4.6. The different ionizer geometries have been tested to compare the propagation of ions from the ionizer to the detection point. The dipole was included and the optimized parameters were used. The electric potential of each LISBET geometry corresponds to the one measured in the terminal connectors (section 4.1.5). An isotropic distribution was considered for the initial velocity direction.

Table 5.5 – Propagation efficiencies simulated with SIMION. The efficiency was calculated as the comparison between the number of particles generated (10000) and the number of particles that arrived to the detector. The losses due to impacts inside the ionizer cavity or in the extraction parts are indicated.

LISBET geometry	Simulated efficiency	Ionizer losses	Extraction losses
LISBET-D7L35	48.34%	50.79%	0.80%
LISBET-D7L60	33.04%	66.40%	0.56%
LISBET-D3L35	15.28%	84.65%	0.07%
LISBET-D3L60	11.04%	88.93%	0.03%

The simulations gave more than 30% of generated ions arriving to the detector for LISBET geometries with 7 mm diameter and between 10% and 15% with 3 mm diameter.

As SIMION cannot reproduce a recycling effect (neutralization followed by re-ionization), every ion that touched the cavity walls was lost. Several behaviors were observed: First, most of the losses in the case of 3 mm diameter ionizers came from neutralization within the ionization tube, while 7 mm diameter ionizers provided a lower number of ions neutralized in the ionization tube. Second, 3 mm diameter ionizers provided smaller losses in the extraction, as the extracted ion beam is smaller, while this value increased in the case of 7 mm ionizers (fig. 5.9).

Finally, an increase of length for both ionizers increased the neutralization within the cavity. It is possible to consider that inside ionizers with 60 mm length the probability to touch the walls is higher compared to the ionizers with 35 mm length. Ionizers with 7 mm diameter and 35 mm length are more favorable to extract the ions once they are produced. Thus they might be more suitable for isotopes with short half-lives.

The simulations did not reproduce the experimental data (25% efficiency for 7 mm diameter ionizers and 7% efficiency for 3 mm diameter ionizers) but they give the same trend for the diameter change: The efficiency increases for 7 mm diameter compared to 3 mm diameter ionizer.

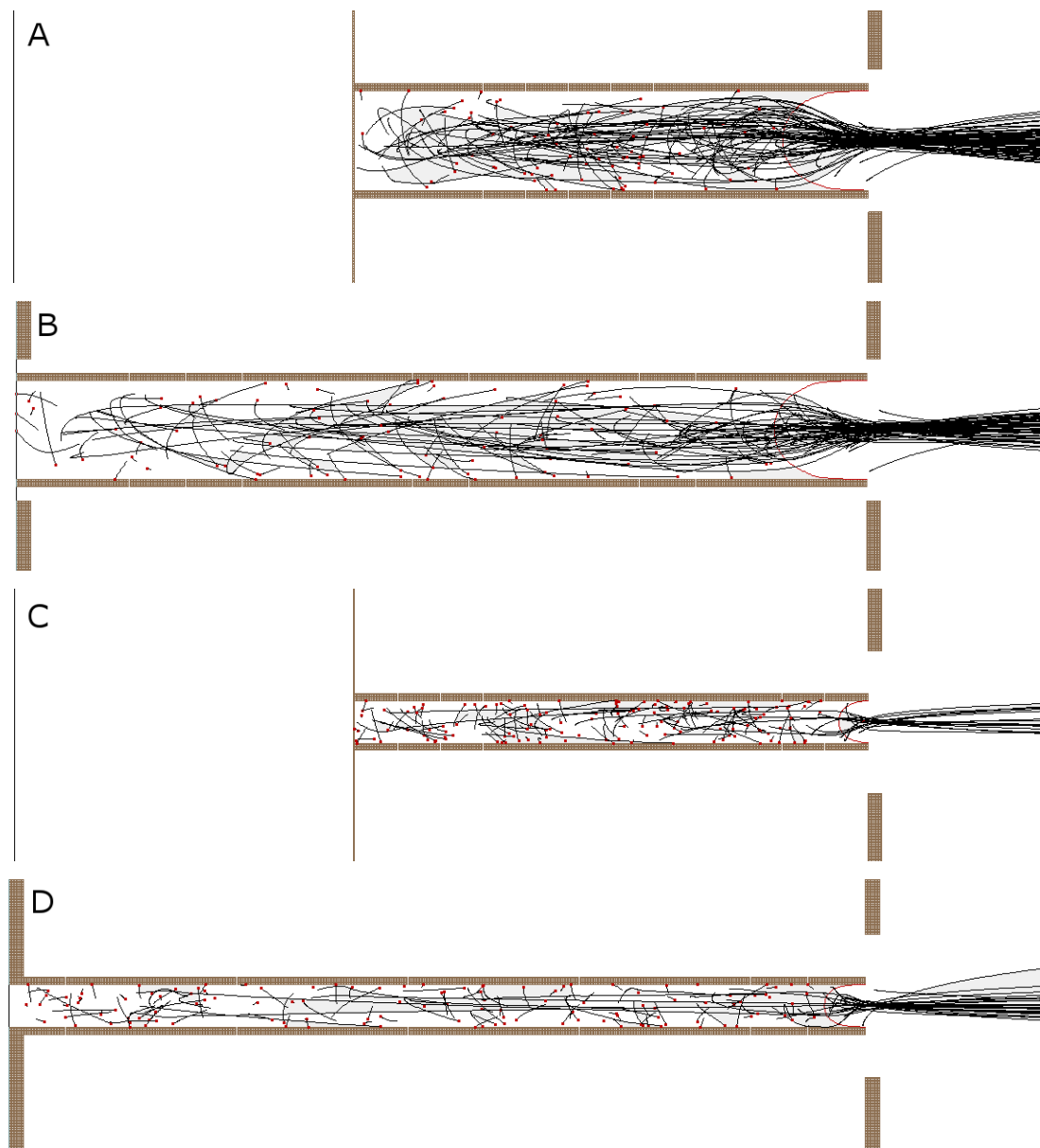


Figure 5.9– Simulation examples of the ionizer and the extraction electrodes for: (A) LISBET-D7L35, (B) LISBET-D7L60, (C) LISBET-D3L35 and (D) LISBET-D3L60. The ion path is represented by black lines, the equi-potential extraction line of 19 kV is shown in red. The ion impact on a surface is marked by a red square. 150 particles are represented in each figure.

The extraction potential (19 kV) enters several millimeters into the ionizer, and it also can generate a focalization effect (red line in figure 5.10). It was also observed in the simulations that the extraction potential depth (into the ionizer tube) changed with the geometry (table 5.6). The particles generated inside the ionizer only feel the electric drift field generated by the heating current flowing through the tube. Due to this fact, the ions at the beginning of the tube have to travel all along the hot-cavity until they meet the 19 kV extraction potential and they are accelerated (fig. 5.10).

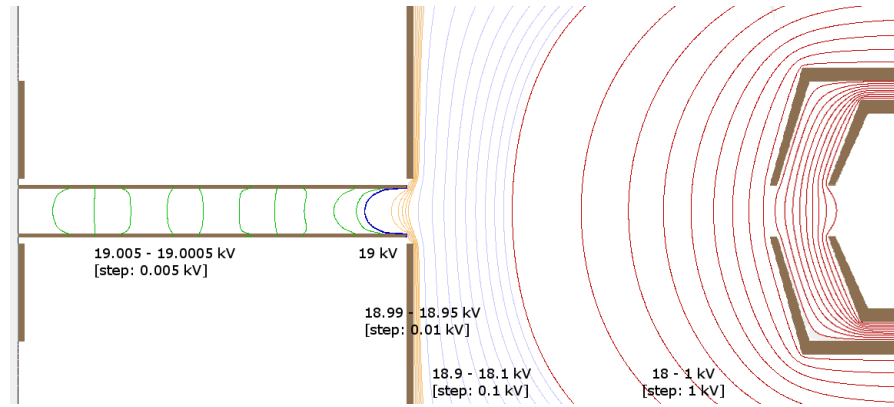


Figure 5.10– Electric field distribution of the ion source and the extraction electrode system for LISBET-D7L60 geometry. The electric field equi-potentials are represented by different colors. The blue line represents the 19 kV extraction potential, the green lines show the potential inside the ion source. Yellow, grey and red represent the potentials till the first extraction electrode.

Table 5.6 – Extraction potential depth calculated from SIMION simulations. The electric potential and the electric field for each geometry due to resistive heating is also provided.

Version	19 kV extraction depth	Electric potential at 2000 K	Electric field at 2000 K
LISBET-D7L35	6 mm	5.4 V	1.54 V/cm
LISBET-D7L60	6.5 mm	5.46 V	0.9 V/cm
LISBET-D3L35	2 mm	3.6 V	1.03 V/cm
LISBET-D3L60	3 mm	4.3 V	0.72 V/cm

5.1.6 Efficiency discussion

Table 5.7 and figure 5.11 gather the Sn efficiency measurements performed for all LISBET options. All measurements have been performed with the optimized double extraction electrode system and the preparation of the Sn sample using a Ta foil (to allow a fast release of the sample). All measured efficiencies (table 5.3) are within the range of the average error.

5.1. Geometry comparison as a function of the efficiency

Table 5.7 – Overall efficiencies for different ion source geometries (from tables 5.3 and 5.4). When not indicated, laser beams have the same diameter as the ionizer diameter.

Name	Ionizer diameter (mm)	Ionizer length (mm)	Efficiency	Error
LISBET-D7L35	7	35	24%	$\pm 3\%$
LISBET-D7L60	7	60	19%	$\pm 4\%$
LISBET-D3L35	3	35	7%	$\pm 2\%$
LISBET-D3L60	3	60	8%	$\pm 2\%$
LISBET-D7L35 (3 mm laser spot)	7	35	10%	$\pm 2\%$
LISBET-D7L35 (Rev. polarity)	7	35	5%	$\pm 1\%$
LISBET-D7L60 (Rev. polarity)	7	60	4%	$\pm 1\%$

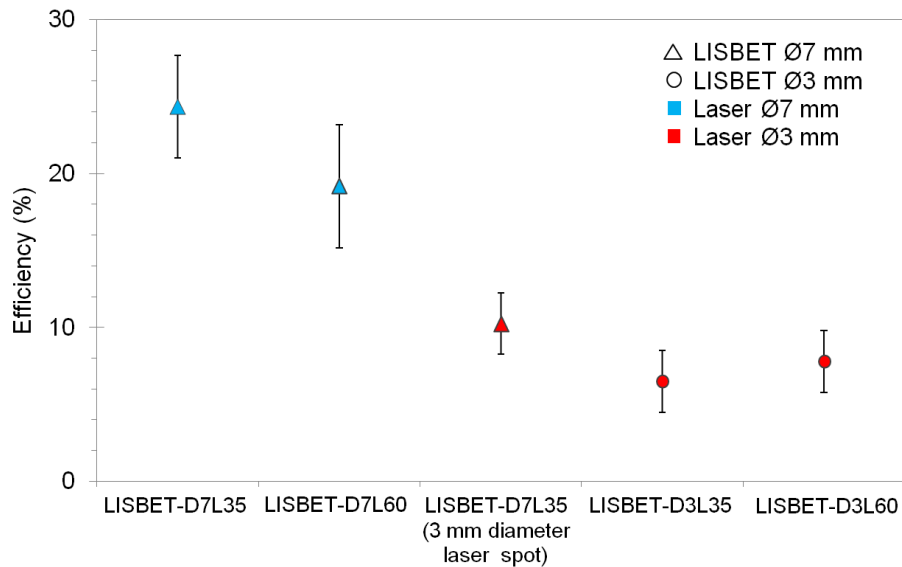


Figure 5.11– Efficiencies of different ion source geometries. The error of the measurement is indicated (from table 5.3).

In comparison, the ISOLDE facility at CERN (Geneva, Switzerland) measured an efficiency for Sn of 9% with 3 mm diameter and 30 mm length Ta ionizer and similar ionization scheme (different first excitation step) [97]. Furthermore, an efficiency of 22% was measured at ISTF2 Oak Ridge (ORNL, USA) with a Ta ionizer of 30 mm length with an inner diameter of 3 mm, preceded by a 7 mm diameter and 90 mm length atomizer in the same axis [169].

5.1.6.1 Length parameter discussion

An increase of efficiency was expected for an increase of length, due to the increase of laser-atom interaction probability (i.e. each atom can see two laser pulses instead of one). The efficiency increase observed at Oak Ridge, compared to ISOLDE efficiency, has been explained due to an ionization process in the whole length system (ionizer + transfer tube) (fig. 5.12). In this case, the **LISBET-D7L60** efficiency ($19\% \pm 4\%$) is comparable to the Oak Ridge (ISTF2). However, **LISBET-D7L35** provided an efficiency of $24\% \pm 3\%$, which does not show a dependence to the length parameter.

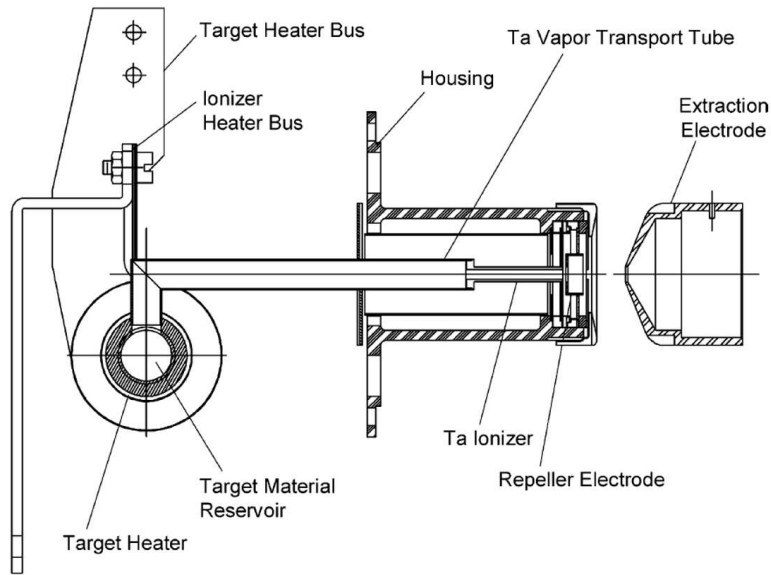


Figure 5.12– Schematic drawing of the hot-cavity ion source at Oak Ridge with 3 mm diameter and 35 mm length ionizer, and 7 mm diameter and 90 mm length atomizer [169].

In section 2.3, a theoretical photo-ionization efficiency approximation was described for hot-cavities with diameter $\ll$ length: $\epsilon_{capillary} = f \cdot \epsilon_{ion} / (f \cdot \epsilon_{ion} + 2 \cdot d \cdot v / 3 \cdot L^2)$.

In order to compare the geometries, saturation in all transitions was considered ($\epsilon_{ion}=0.43$). The laser repetition rate is 10 kHz and the most probable ion velocity is 530 m/s for Sn at 2000 K (section 2.1). The theoretical photo-ionization efficiencies of LISBET ion sources obtained by this formula are 39.58% (LISBET-D3L35), 41.77% (LISBET-D3L60), 35.78% (LISBET-D7L35) and 40.24% (LISBET-D7L60). The results are gathered in table 5.8.

According to the equation 2.13, the efficiency is higher for a smaller ionizer diameter and is also higher for longer ionizer length due to the increase of laser-atom interaction. An ion source with small diameter and long length should confine the atoms and increase their residence time inside the ion source, which should enhance the efficiency.

5.1. Geometry comparison as a function of the efficiency

Table 5.8 – Comparative table of the theoretical ionization efficiency in a capillary hot-cavity given by 2.13 equation.

	3 mm ionizer diameter	7 mm ionizer diameter
Ionizer length 35 mm	39.58%	35.78%
Ionizer length 60 mm	41.77%	40.24%

The formula takes into account the effusion of atoms with a linear gradient of the vapor density from max in the beginning of tube to zero at the exit (approximation). Ion extraction is assumed to be 100% due to the plasma potential. A change in the ionizer length did not modified the efficiency out of the error bar. **LISBET-D3L60** ($8\pm2\%$) compared to **LISBET-D3L35** ($7\pm2\%$), and in the case of **LISBET-D7L35** ($24\pm3\%$) compared to **LISBET-D7L60** ($19\pm4\%$) (fig. 5.11).

Table 5.6 shows the electric field of each ion source calculated by the electric potential in the terminal connectors for each ionizer length. One observes that **LISBET-D7L35** provides an electric field of 1.54 V/cm while other solutions have lower values. The increase of the electric field could provide an extra longitudinal acceleration that diminishes the neutralization of the ions inside the ionizer.

The SIMION simulations showed that an extension of the length has the same consequence for both diameters (table 5.7). Ionizers with 60 mm length showed an increase of neutralization within the cavity, while this value decreased for ionizers with 35 mm length.

5.1.6.2 Diameter parameter discussion

The **LISBET-D3L35** ionizer is similar to the ISOLDE configuration. With an efficiency of $7\%\pm2\%$ and 9% respectively, comparable results were measured. A decrease of efficiency was expected with the enlargement of diameter due to the lower probability of interaction. Equation 2.13 shows that an enlargement of diameter decreases the theoretical ion source efficiency. However, an increase of efficiency was measured for bigger ionizer diameters: **LISBET-D7L35** ($24\pm3\%$) and **LISBET-D7L60** ($19\pm4\%$).

The hypothesis to explain the efficiency are based on the increase of interaction volume between atoms and photons, and the decrease of contact surface for 7 mm diameter ionizers compared to 3 mm diameter ionizers. The contact surface ratio can be characterized by the surface divided by the volume of the ionizer, obtaining a ratio of $0.\bar{6}$ for 3 mm diameter ionizer and $0.\bar{3}$ for 7 mm diameter ionizer. Lowering the contact surface ratio for 7 mm diameter ionizers also decreases the probability of ion collision with the ionizer cavity walls, which neutralizes the ions. Moreover, a bigger diameter also ease the laser alignment and ion signal optimization.

An enlargement of diameter also increases the electric field potential (table 5.6). This potential can drift the ions towards the extraction system, increasing the probability of leaving the ionizer without collisions. Moreover, ion sources with 3 mm diameter have several complications which do not ensure a perfect laser alignment. Small angles between the laser beams and the ion source (or even between each laser beam) can lead to a decrease of the ion signal, while this effect was attenuated for bigger diameters.

In addition, the laser focalization can modify the efficiency. As already described, an efficiency measurement was performed reducing the laser focus to 3 mm for an ionizer with 7 mm diameter (**LISBET-D7L35** measurement #12). The obtained efficiency of $10 \pm 2\%$ is comparable to the one measured with **LISBET-D3L35** ($7 \pm 2\%$). The decrease compared to **LISBET-D7L35** can be explained with the lower interaction of the lasers with the atoms (due to un-illuminated regions), and the small increase compared to **LISBET-D3L35** due to a lower neutralization in the cavity.

5.1.7 Optimum diameter simulation

A simulation has been performed to investigate the diameter effect into the efficiency. The geometry used as reference was **LISBET-D7L60** with 19 kV extraction voltage and an isotropic distribution of the initial velocity. The voltage potential was weighted for each diameter from the known values: 5 V for **LISBET-D7L60** and ~ 4 V for **LISBET-D3L60**.

Table 5.9 – Simulations performed with different ionizer diameters. The efficiency was calculated as the ratio between the number of detected particles to the number of particles generated in the cavity. The losses due to the impact inside the ionizer cavity or in the extraction parts are indicated. The extraction electrode diameter was 8 mm for all simulations.

Ionizer diameter	Simulated efficiency	Ionizer losses	Extraction losses
3 mm	11.04%	88.93%	0.03%
4 mm	15.79%	84.09%	0.12%
5 mm	20.18%	79.42%	0.40%
6 mm	25.36%	73.92%	0.72%
7 mm	33.04%	65.84%	1.12%
8 mm	32.44%	65.81%	1.75%

Ionizer diameters from 3 to 8 mm were compared (table 5.9) and the highest efficiency was found for the ionizer with diameter 7 mm. The losses due to neutralization in the cavity increased for ionizer diameters lower than 6 mm diameter, and the extraction losses increased for the ionizer diameters higher than 7 mm diameter. The optimum ionizer diameter corresponds to the one that minimizes both, the neutralization inside and outside the ionizer cavity.

5.2 Geometry comparison as a function of the emittance

The quality of an ion beam is fundamental for its proper transport and further use in any mass spectrometer or accelerator facility. It similarly determines the mass resolution and transport efficiency at on-line mass separator facilities [203]. Therefore a precise knowledge of the emittance is necessary for a full characterization of the generated ion beams. The slit-grid method was used in this thesis to measure the rms (root mean square) emittance with a standard deviation of 2σ error. In the next section a theoretical approximation of the emittance and the treatment calculation will be described.

5.2.1 General definition of the emittance

Considering a particle beam propagating through an axis. Each particle is characterized by a set of variables of position (x, y, z) and velocity (v_x, v_y, v_z) (fig. 5.13). An ion beam can be defined by its emittance figure. The emittance figure delimits a region of phase space where the values of the described variables are included (x, y, z, v_x, v_y, v_z) . If a transverse phase-space of horizontal motion is considered, two transversal emittances can be defined: horizontal and vertical emittance. The emittance, considered at a fixed z distance (z the propagation axis), is a 2D figure $(x, v_x$ or $y, v_y)$. A tangent to angle approximation transforms the expression to (x, x') or (y, y') parameters, where x' and y' represent the divergence angles given in mrad.

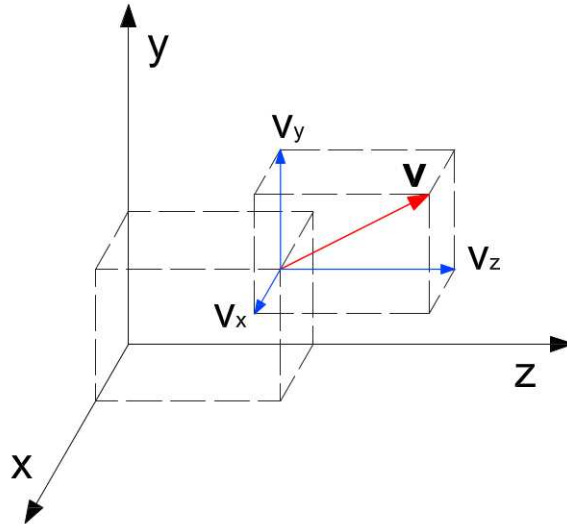


Figure 5.13– Definition of position and velocity for a particle propagating through an axis.

The geometric emittance corresponds to the area filled by the particle distribution (x, x') . The emittance figures are theoretically described by ellipses. This area is usually divided by π ,

giving the units for the emittance as $\pi \cdot \text{mm} \cdot \text{mrad}$. The geometric emittance in the case of an ellipse is equal to the semi-axes of the equivalent ellipse. The rms (root mean square) emittance is a statistical emittance which gives consideration to the weights of the density distribution [204]. The emittance can be expressed in $\text{mm} \cdot \text{mrad}$ or $\pi \cdot \text{mm} \cdot \text{mrad}$ being the latter equal to the area of the correlated ellipse. The procedure to calculate the rms emittance will be described in the next point.

5.2.1.1 Emittance diagnosis

The slit-grid method was used to measure the rms (root mean square) emittance. In this method, the ion beam is scanned by a pair of metal plates with a defined slit distance (fig. 5.14). The position and divergence of each slice of the ion beam is recorded by a low-intensity profiler detector (section 4.4.2). This method allows to measure the ion beam intensity as a function of the particle position and its angle ($I_{xx'}$) and the density profile $\rho(x, x')$. A MATLAB code was integrated to analyze the data from the ion beam profiler and calculate the rms emittance.

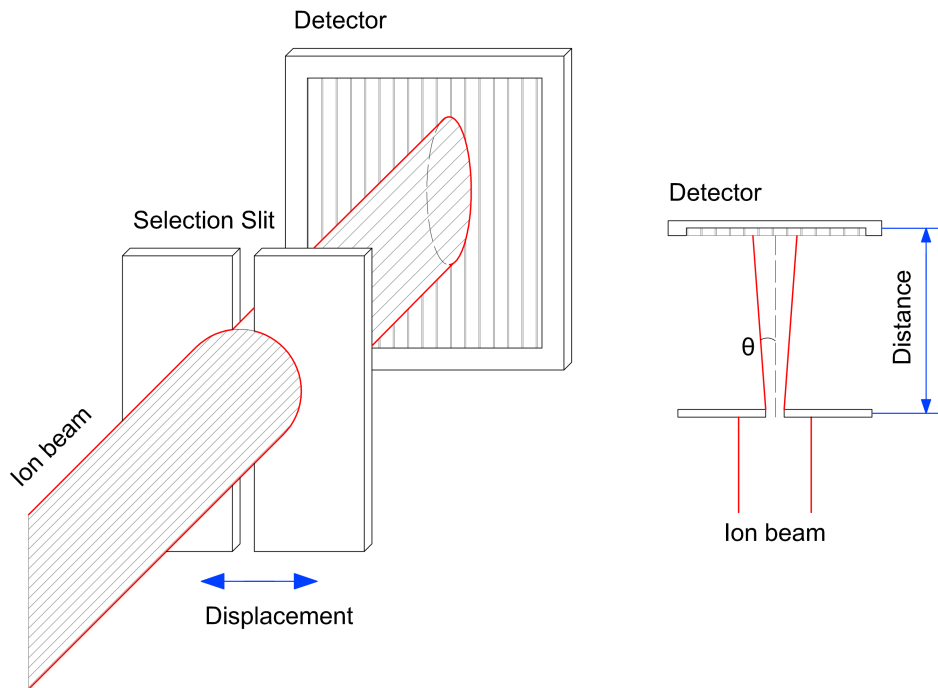


Figure 5.14– Emittance characterization by the slit-grid method. The ion beam is scanned by a pair of semi-closed metal plates. The truncated ion beam going through the slit between the plates is recorded by a low-intensity ion beam profiler.

5.2.1.2 Emittance treatment

The emittance data are stored as an array of beam intensities (I) at each slit positions (x) and divergence angles (x') [204]. The moments of the beam distribution can be calculated as follows:

The total intensity measured I_{total} :

$$I_{total} = \sum_{i=1}^N \sum_{j=1}^M I(x_i, x'_j) \quad (5.18)$$

The central value of displacement x_c :

$$x_c = \frac{\sum_{i=1}^N \sum_{j=1}^M x_i I(x_i, x'_j)}{I_{total}} \quad (5.19)$$

The central value of angular divergence x'_c :

$$x'_c = \frac{\sum_{i=1}^N \sum_{j=1}^M x'_j I(x_i, x'_j)}{I_{total}} \quad (5.20)$$

And the mean values $\langle x^2 \rangle$, $\langle x'^2 \rangle$ and $\langle xx' \rangle$:

$$\langle x^2 \rangle = \frac{\sum_{i=1}^N \sum_{j=1}^M (x_i - x_c)^2 I(x_i, x'_j)}{I_{total}} \quad (5.21)$$

$$\langle x'^2 \rangle = \frac{\sum_{i=1}^N \sum_{j=1}^M (x'_j - x'_c)^2 I(x_i, x'_j)}{I_{total}} \quad (5.22)$$

$$\langle xx' \rangle = \frac{\sum_{i=1}^N \sum_{j=1}^M (x_i - x_c)(x'_j - x'_c) I(x_i, x'_j)}{I_{total}} \quad (5.23)$$

where $\chi_{rms} = \sqrt{\langle x^2 \rangle}$ and $\chi'_{rms} = \sqrt{\langle x'^2 \rangle}$ are the beam widths, and $\sqrt{\langle xx' \rangle}$ is the correlation. The rms emittance is then defined as:

$$\epsilon_{rms} = \sqrt{\langle x^2 \rangle \langle x'^2 \rangle - \langle xx' \rangle^2} \quad (5.24)$$

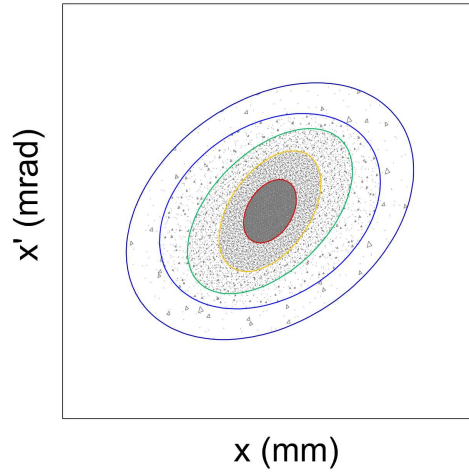


Figure 5.15– Emittance figure example. Each pixel represents the position by means of its divergence where the iso-intensity are represented.

The related emittance figure consist of iso-intensities lines, with the position of the beam (x) as a function of the divergence (x') as it can be seen in [figure 5.15](#).

5.2.1.3 Emittance incertitude

In order to avoid over estimated emittances due to the background noise, the ion intensity is considered only above a threshold equivalent to 5% of the ion maximum intensity. This value corresponds to a 2σ error. The emittance error was calculated by the method described in [\[205\]](#).

In addition, it is possible to calculate the angular resolution $\delta_{x'}$ of the emittance-meter given in mrad with the slits aperture δ_x (spatial resolution on x), the distance between two profiler threads δ_e and the distance slit-profiler L [\[206\]](#).

$$\delta_{x'} = \frac{\delta_x + \delta_e}{L} \quad (5.25)$$

The minimum emittance $\delta\epsilon_{min}$ that a system can record is:

$$\delta\epsilon_{min} = \frac{\delta_x \cdot \delta_{x'}}{\pi} \quad (5.26)$$

Implementing the parameters of the off-line test bench: $L = 500$ mm, $\delta_x = 0.6$ mm and $\delta_e = 1$ mm. The resulting angular resolution $\delta_{x'}$ is equal to 3.2 mrad. The minimum emittance that the system can measure is $\delta\epsilon_{min} = 0.61 \pi \cdot \text{mm} \cdot \text{mrad}$. This limitation of the system is enough for the measurements that were carried out.

5.2.2 Emittance results for different LISBET geometries

The rms (root mean square) emittances 2σ of the four configurations of the ion source have been measured and compared with the method previously described (figure 5.16).

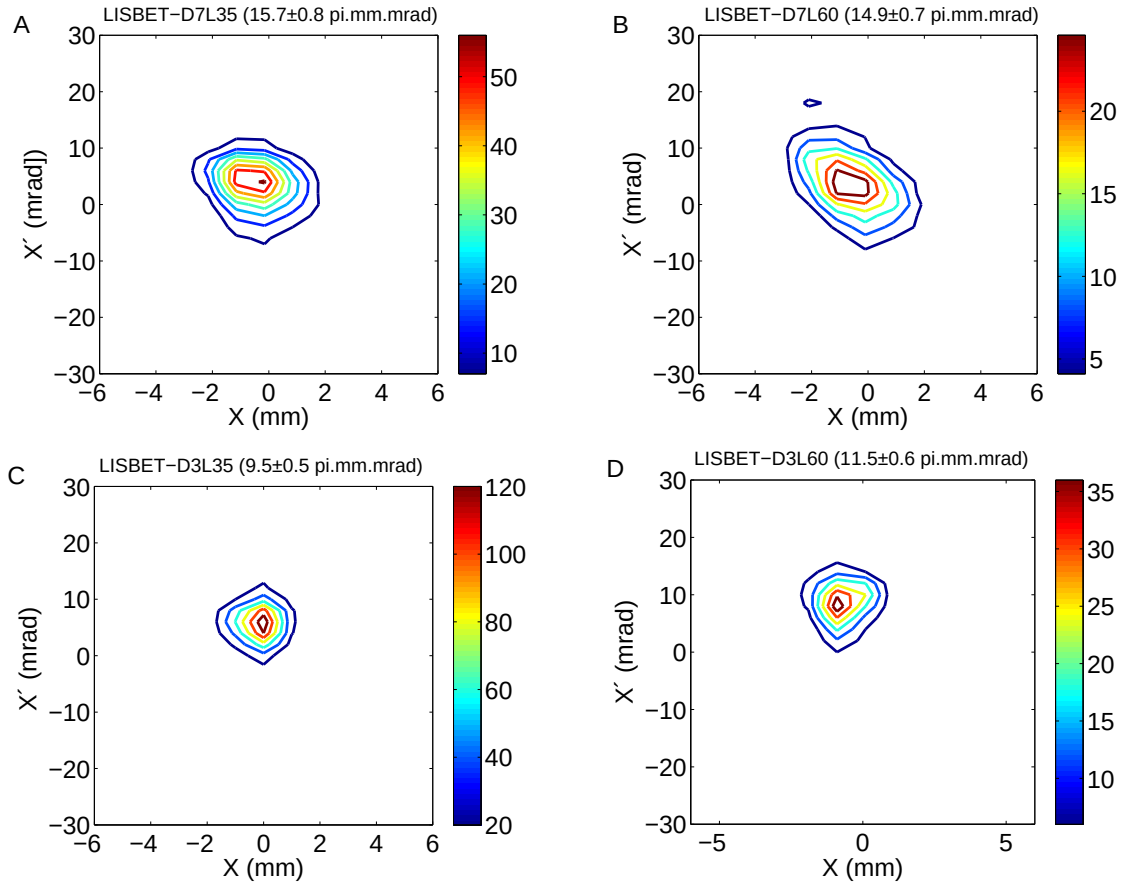


Figure 5.16– Rms emittances 2σ measured with (A) LISBET-D7L35 ($15.7 \pm 0.8 \pi \cdot \text{mm} \cdot \text{mrad}$), (B) LISBET-D7L60 ($14.9 \pm 0.7 \pi \cdot \text{mm} \cdot \text{mrad}$), (C) LISBET-D3L35 ($9.5 \pm 0.5 \pi \cdot \text{mm} \cdot \text{mrad}$) and (D) LISBET-D3L60 ($11.5 \pm 0.6 \pi \cdot \text{mm} \cdot \text{mrad}$).

Each emittance was measured with a new sample. The low intensity profiler placed at the end of the detection line (after the Faraday cup) was used to measure the emittances. The large distance between the slit plates to the profiler (500 mm) increases the resolution of the emittances. The ion current intensity during the acquisitions was in the range from 1 pA up to 1 nA. The lasers were optimized like for the efficiency measurements and the extraction system parameters were the same (19 kV extraction voltage and optimized extraction electrodes). The emittances were measured at about 1800 K.

The next rms emittances 2σ were obtained: $15.7 \pm 0.8 \pi \cdot \text{mm} \cdot \text{mrad}$ for **LISBET-D7L35**, $14.9 \pm 0.7 \pi \cdot \text{mm} \cdot \text{mrad}$ for **LISBET-D7L60**, $9.5 \pm 0.5 \pi \cdot \text{mm} \cdot \text{mrad}$ for **LISBET-D3L35** and $11.5 \pm 0.6 \pi \cdot \text{mm} \cdot \text{mrad}$ for **LISBET-D3L60**. The emittance was smaller for smaller ionizer diameter, due to the decrease of the ion spread in space at the exit of the ionizer. The figures 5.16 show the analyzed emittance iso-intensity contours of the four LISBET geometries.

5.2.2.1 Simulations

The emittance has also been studied by the simulation described in section 4.6 for the different ion source configurations. The extraction voltage was set to 19 kV and the already explained optimized extraction parameters were used. The detection point was set in the position of the last low intensity ion beam profiler (like for the emittance experiments). The emittances are evaluated directly by a Lua program [207] included in the SIMION simulation. No threshold was applied to calculate the simulated emittances. For that reason, all points arriving to the detector were included in the calculation and consequently a bigger values of emittance were obtained.

The distribution of the simulated points collected in the detector can be observed in figure 5.17. The average value of ten series of 1000 ions each were simulated for each geometry. The average emittance for **LISBET-D7L35** was $43 \pi \cdot \text{mm} \cdot \text{mrad}$. For **LISBET-D7L60** $43 \pi \cdot \text{mm} \cdot \text{mrad}$. In the case of **LISBET-D3L35** $23 \pi \cdot \text{mm} \cdot \text{mrad}$ and **LISBET-D3L60** $26 \pi \cdot \text{mm} \cdot \text{mrad}$ were recorded.

Some ions are drifted to the left part of figures 5.17-A, 5.17-B and 5.17-D . This dispersion was not observed in the measurements, but the threshold removed these values from the emittance figure. The simulations provided similar behaviors compared to LISBET configurations (higher emittances for bigger diameter ionizers). However, the emittance values were increased in the simulations due to the inexistent threshold to the data (in the case of emittance experiments a threshold at 5% of maximum intensity was applied). In addition, an increase of emittance can be also explained due to a weaker focusing of the simulated magnet mass spectrometer (this was observed in preliminary simulations).

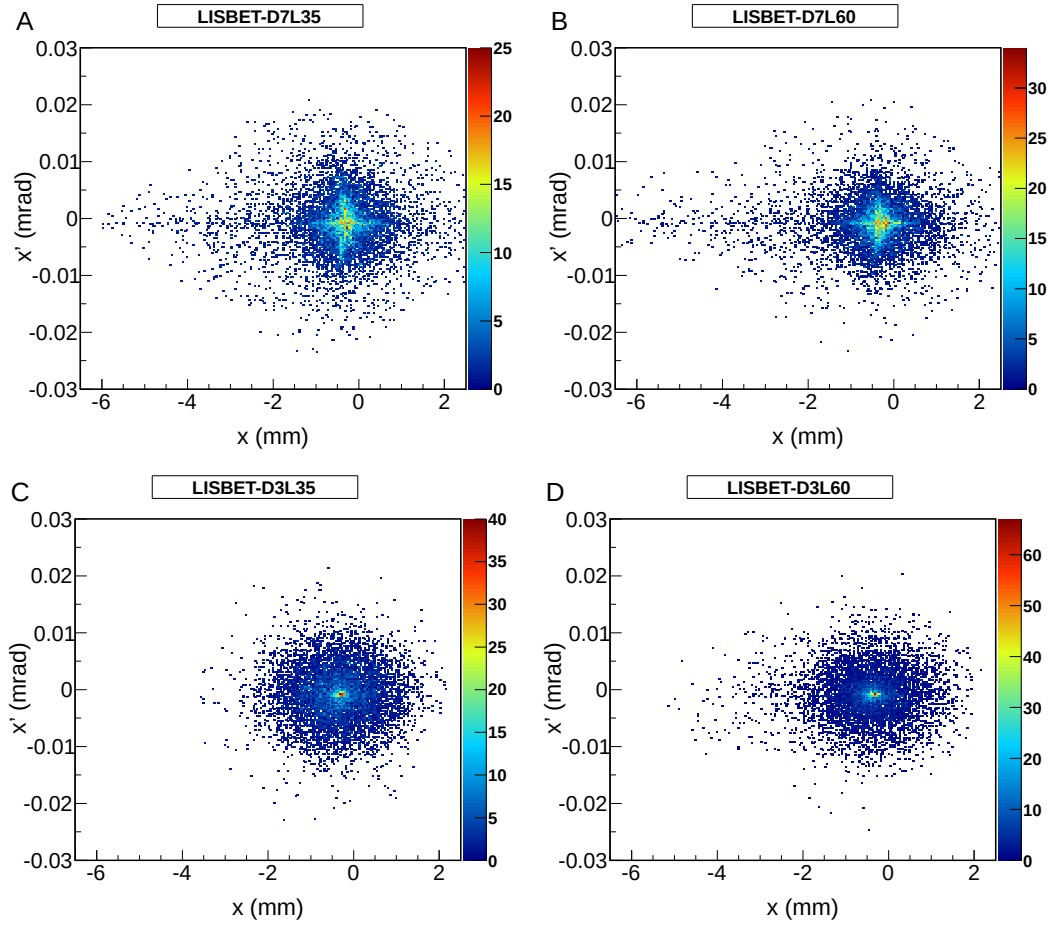


Figure 5.17– Emittance simulations for different LISBET configurations: (A) LISBET-D7L35, (B) LISBET-D7L60, (C) LISBET-D3L35 and (D) LISBET-D3L60. An spatial drift of ions towards the left part of the figure can be observed for LISBET-D7L35, LISBET-D7L60 and LISBET-D3L60.

5.2.2.2 Temperature change

A variation of the emittance value depending on the temperature was noticed. For this reason, the emittances as a function of the ion source working temperature (from 1400 K to 2100 K) were measured (fig. 5.18). The emittance stays constant at low temperatures, but an increase was observed from 1400 K to 2100 K for all geometries, and a high dispersion of results was observed in the case of **LISBET-D7L60**.

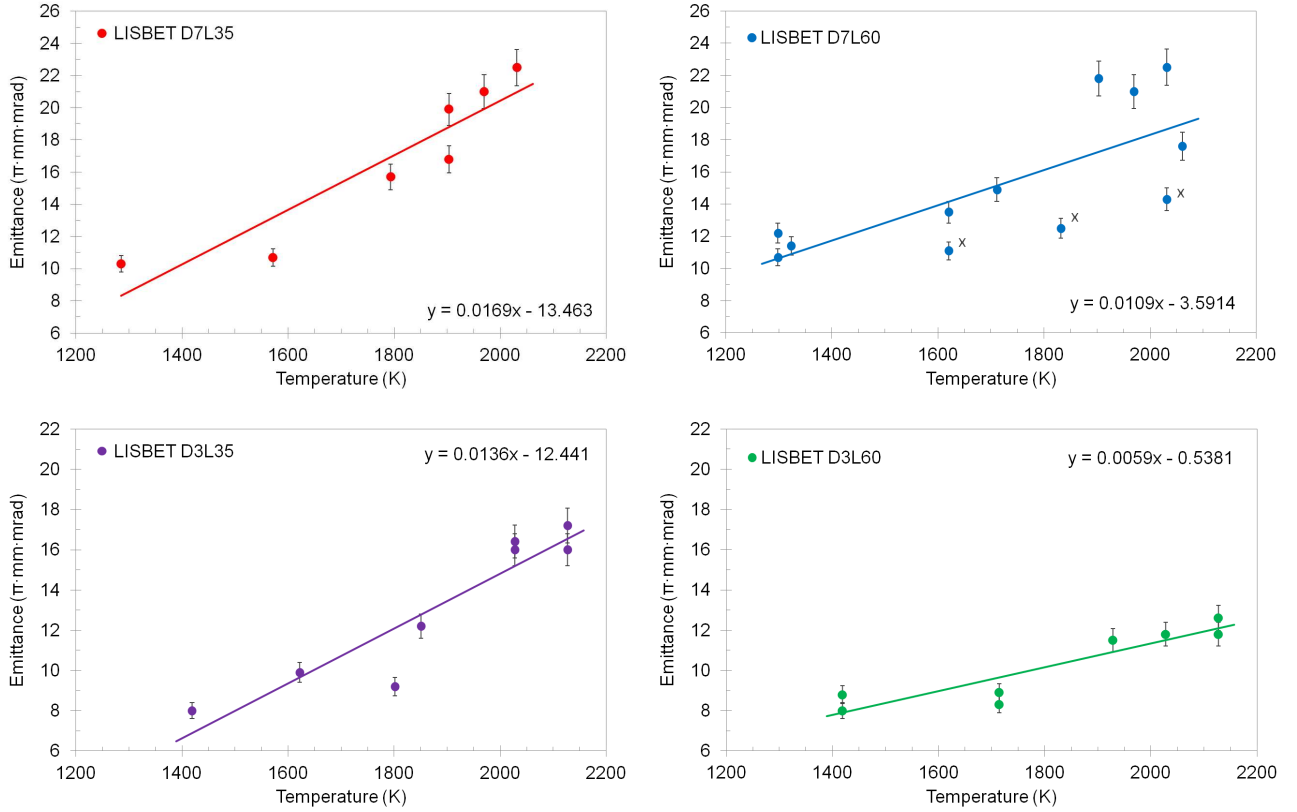


Figure 5.18– Emittance measurement evolution as a function of the working temperature for (A) LISBET-D7L35, (B) LISBET-D7L60, (C) LISBET-D3L35 and (D) LISBET-D3L60.

5.2.3 Emittance discussion

The emittance results for different LISBET configurations are gathered in the table 5.10. A bigger ionizer diameter increases the emittance, while no effect was observed for a variation of length. The increase of emittance for 7 mm ionizer diameters was explained due to a bigger volume in the ionizer exit.

Table 5.10 – Overall emittances for different ion source geometries measured at the low intensity ion beam profiler. The extraction potential was set to 19 kV and the optimized extraction electrode parameters were used. The emittances were measured at about 1800 K.

Name	Ionizer diameter (mm)	Ionizer length (mm)	Rms emittance 2σ	Error
LISBET-D7L35	7	35	$15.7 \pi \cdot \text{mm} \cdot \text{mrad}$	± 0.8
LISBET-D7L60	7	60	$14.9 \pi \cdot \text{mm} \cdot \text{mrad}$	± 0.7
LISBET-D3L35	3	35	$9.5 \pi \cdot \text{mm} \cdot \text{mrad}$	± 0.5
LISBET-D3L60	3	60	$11.5 \pi \cdot \text{mm} \cdot \text{mrad}$	± 0.6

It was observed that the extraction potential generates a lens effect when entering into the ionizer (fig. 5.9). This effect was pronounced for 7 mm diameter ionizers, where the extraction potential enters up to 6 millimeters. The lens effect can also increase the emittance due to the tangential electric potential that the ion experience in this region.

The increase of the emittance due to the temperature was observed, where each LISBET geometry showed a different behavior. The increase might be explained by the experimental conditions, since an increase of temperature also increases the dispersion of the atoms and ions in the produced beam.

Another hypothesis is that all measurements were performed with new samples placed after opening the ion source chamber. This could increase the presence of alkali atoms in the ion source, with the consequent enhanced production of alkali ions at high temperatures.

These alkalis can reach ion currents of $1\ \mu\text{A}$. Thus the increase of emittance may be related to space-charge effect due to the enhanced alkali production at high temperatures. The emittance values marked with an X in figure 5.18-B were performed after several heatings with the same sample. The decrease of alkali production and the consequent reduction of the space-charge effect also reduced the emittance value. Further studies are needed to understand how this can affect the ion source requirements.

As a comparison, the charge booster $1+$ to $n+$ that increases only the charge before the acceleration in the CIME cyclotron has an acceptance of about $40\ \pi\cdot\text{mm}\cdot\text{mrad}$, DESIR-HRS has an RFQ cooler (SHIRaC) with acceptance of $80\ \pi\cdot\text{mm}\cdot\text{mrad}$ and the transport lines at GANIL have an acceptance of $80\ \pi\cdot\text{mm}\cdot\text{mrad}$. All ion beams produced by the different LISBET configurations are smaller than these values and can be transported without losses.

5.3 Ion time structure study

5.3.1 Acquisition system

Due to the nature of the RILIS ionization process (i.e the ionization is carried out by pulsed lasers) the ions are generated in pulsed bunches. The temporal shape of the ion bunches can provide valuable information of the ion production and extraction (e.g. the individual velocity of the ions) [3]. A setup was implemented to detect and to quantify the time-of-flight of the ions after mass separation, where the objective is to study the temporal structure of the ions generated inside the hot-cavity. The concept of suitable time structure acquisition systems were described by J. Lettry et al. [114] at CERN and later by Y. Liu et al. [208] at Oak Ridge. They studied and explained the processes which occur inside the hot cavity for RILIS ionization of different elements. They were able to simulate the observed ion time profiles based on ion motions driven by the electric field resulting from resistive heating and assuming a thermal plasma formed in the hot cavity that confines the ions.

5.3.1.1 Faraday Cup solution

The time structure detection system based on a Faraday cup (FC) was developed first. The main idea is to use a fast response of the Faraday cup system (section 4.4) combined with a digital acquisition system to reconstruct the shape of the ion time structure, through the measurement of the current intensity.

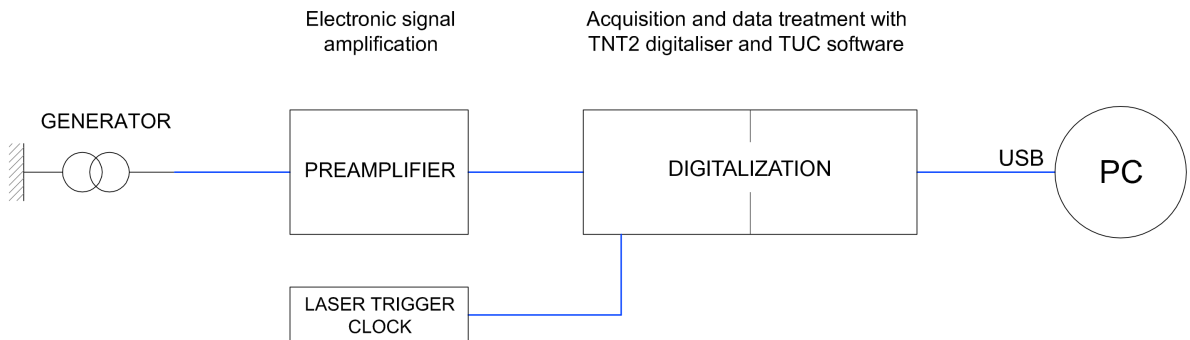


Figure 5.19– Diagram of the electronic chain and data acquisition system used to study the ion time structure with a Faraday cup or a pulse generator. A TNT2 CAEN digitalizer card together with a TUC (TNT USB Control) software was used to record the time structure signal.

A pre-test was performed with a pulse generator (Tektronix AFG3102) in order to validate this setup (fig. 5.19). The pulse generator was used to simulate the ion signal coming from the Faraday cup during an experiment. The output signal of the generator was processed by a preamplifier which integrates the current and provides an output voltage proportional to the charge collected and is then sent to a Digital Pulse Processor (TNT2 CAEN) monitored by a PC. In order to correctly trigger the signal from the Faraday cup, the acquisition system was correlated with the pump laser pulse master-clock. However, the noise level coming from the test bench environment degraded the signal-to-noise ratio too much which forced us to find another solution. Nevertheless, this solution could be interesting to be tested at the SPIRAL2 facility, where the background conditions may be better.

5.3.1.2 Low intensity ion beam profiler solution

The use of a low intensity ion beam profiler [199] connected to a standard acquisition system was the second option tested to measure the time structure of the ions. The idea was to measure the time-of-flight between the ions detected by the profiler and the pump laser pulse master-clock. The profiler is based on a micro-channel plate (MCP) detector which collects electrons produced by the interaction of ions with an emissive foil located along the beam axis. It allows the reconstruction of the beam spot profile and provides a fast time signal (positive signal of about 2.5 ns rise time). The signal was first inversed with an inversion module and amplified using a fast amplification module (AR4) and sent afterwards to a constant fraction discriminator (ENERTEC CFD).

The advantage of the AR4 module is that it amplifies the signal, without losing its time resolution too much. The threshold of the CFD was adjusted to be above the noise level (-248 mV), such that no counts are recorded when the beam is off. No inconvenient background noise has been seen in the inspection module. The NIM output signal from the CFD was sent to the **stop** input of the Time-to-Amplitude Converter (ORTEC TAC). In addition, a rate-meter device was connected to control the amount of signal being processed by the electronics to avoid electronic saturation and/or an irreversible deterioration of the detector. The measurements were performed with a beam intensity of 5000 counts/s (fig. 5.20).

The external trigger pulse from the laser master-clock was converted to the NIM standard and sent to a dual gate generator module (Le Croy DGG 993). A delay of 20 μ s, which corresponds to the approximate travel time of Sn ions at 19 kV from the ionizer to the detector, was set to have a more convenient acquisition window. This signal corresponds to the **start** input of the TAC. The range of the TAC module was adapted to record a full time spectrum. A Time-To-Amplitude Converter module transforms the small time interval between a start signal and a stop one to an analog output pulse proportional to the measured time [209]. The latter was recorded with an AMPTEK multi-channel analyzer (MCA-8000D).

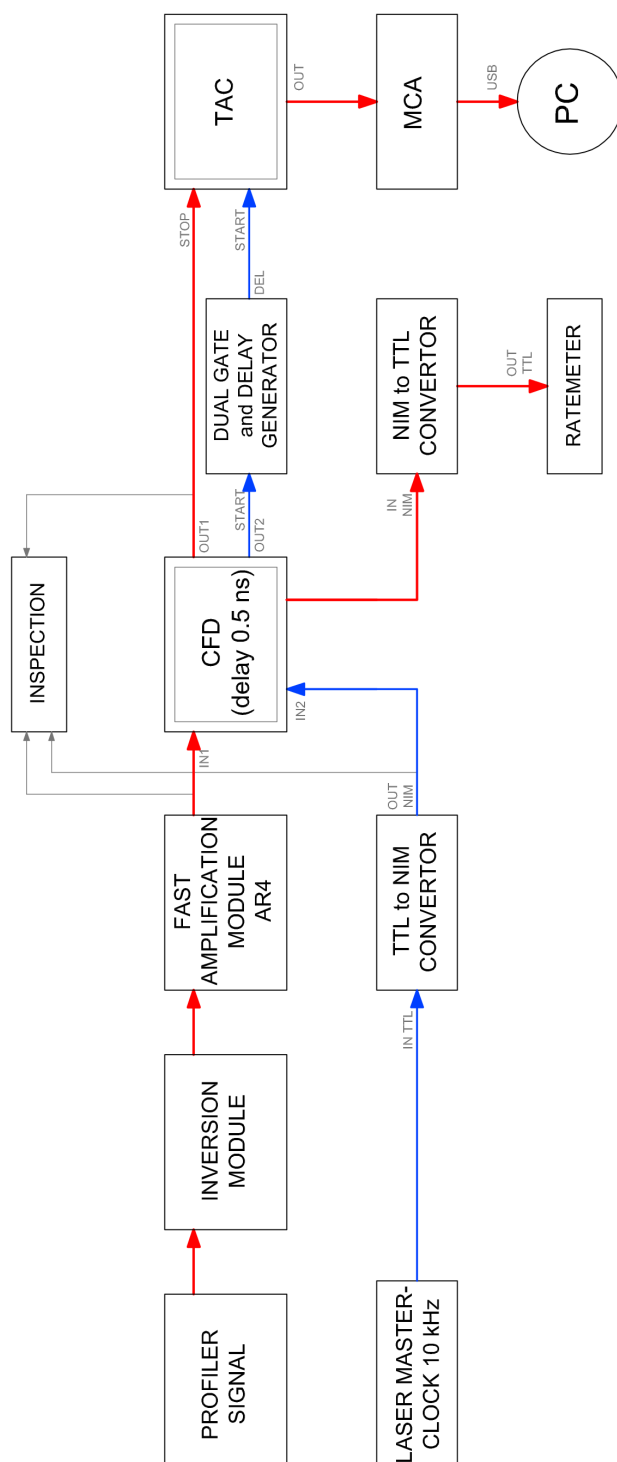


Figure 5.20– Diagram of the electronics chain and data acquisition used to record the ion time structure using a low intensity ion beam profiler.

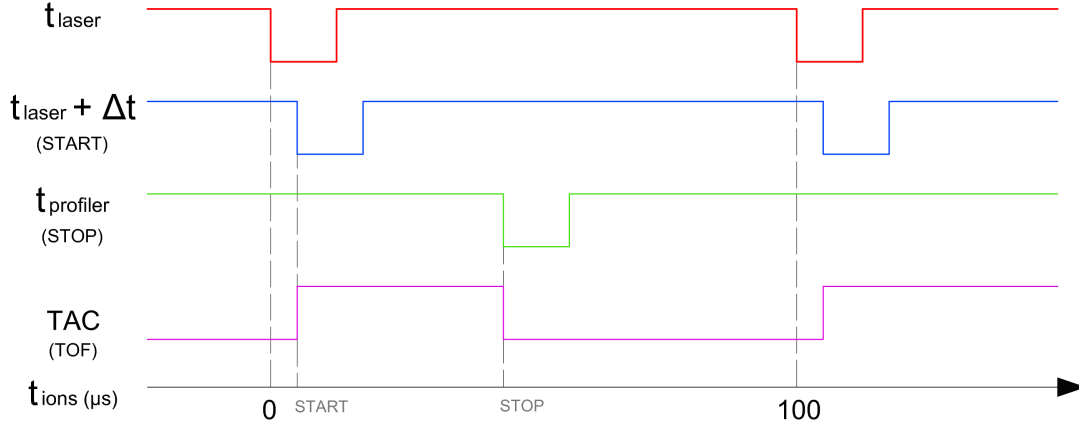


Figure 5.21– Diagram of signal time line to determine the time-of-flight of ions. t_{laser} is the master-clock signal, Δt the delay, $t_{profiler}$ the profiler signal and TAC the time-of-flight signal.

A diagram of the signal steps can be found in appendix 5.21. A calibration of the TAC was performed with a pulse generator in order to check the linearity of the amplification and to calibrate the obtained spectrum (in μs). The calibration curve can be found in figure 5.22. The resulting calibration equation of the MCA acquisition system for 4096 channels is:

$$\tau(s) = N_{channel} \cdot 0.023461 - 0.005967 \quad (5.27)$$

where $N_{channel}$ is the channel number and $\tau(s)$ is the corresponding time in seconds.

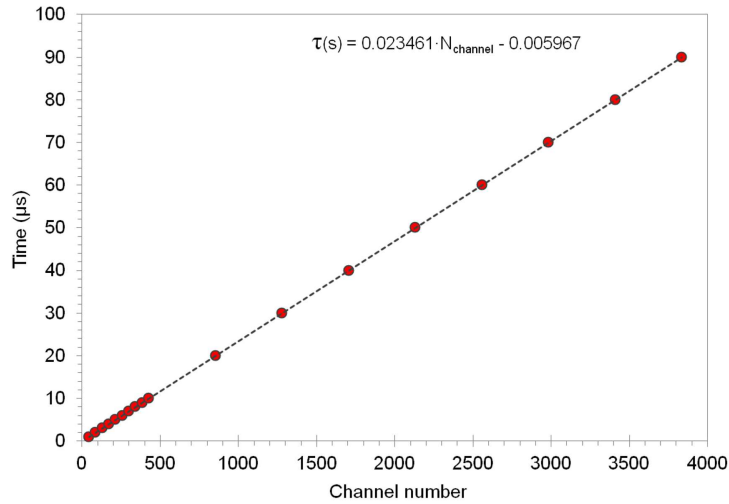


Figure 5.22– Calibration of the acquisition system, where the time is presented as a function of the channel number. The reference signal was set to different times: From 1 to 10 μs (with 1 μs step) and then from 10 to 90 μs (with 10 μs step)

5.3.2 RILIS time profile structures

The time structure of each individual ion bunch, produced by the RILIS technique in a hot-cavity, consists of one narrow peak (typically few microseconds wide) followed by a broad structure (also named main peak) (fig. 5.23). It is widely considered that the first narrow peak is constituted by the atoms ionized close to the ionization tube exit where the extraction field is present, and the second broad peak is composed of the atoms ionized deep inside the ionization tube [114, 140, 208].

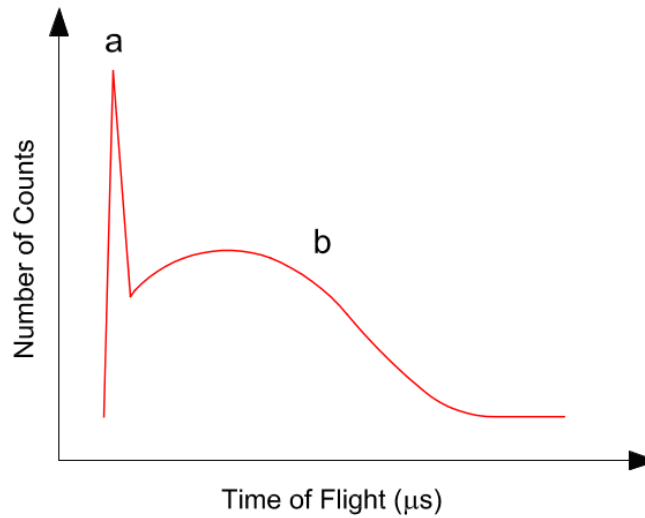


Figure 5.23– Time profile structure measurement diagram example. The number of counts is represented as a function of the time-of-flight of the ions for a range of time. The time structure of a hot-cavity RILIS consists of a narrow peak (a) followed by a broad peak (b) (see text for explanations).

5.3.2.1 Ion time-of-flight simulations

A simulation based analysis for the LISBET-D7L60 geometry (with electric potential of 5.4 V) was performed in order to investigate the effect of the starting position of the ions compared to their time-of-flight. Therefore, the ionizer tube length was divided into 60 parts of 1 mm each, and the time-of-flight of which ions are generated in each slice of the ionizer were recorded. By this, it was possible to analyze the approximate walk of each ion for a set of parameters. Figure 5.24 shows the resulting data.

The simulation confirms that the ions are generated close to the extraction region at the exit of the ionizer (from 0 to 6 mm in figure 5.24) and they are extracted in a compact bunch (the narrow peak) which have a time-of-flight between 2.5 and 5 μ s.

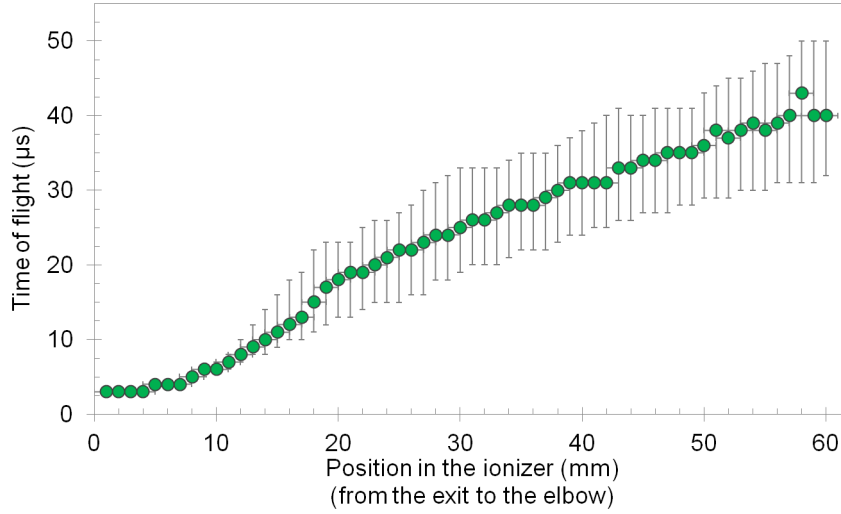


Figure 5.24– Ion beam simulation where the starting position of the ions in the ionizer was compared to their time of flight. The zero position was set at the exit of the ionizer tube. The vertical error bars come from the temporal spread of the ion bunch arriving to the detection.

The ions with the longest time-of-flight are produced in farther position and have to travel all the way along the tube to the extraction field at the tube exit. In addition, the time spread of the ions in the narrow peak is at a minimum (the error bar in [figure 5.24](#) shows the temporal spread), while the temporal spread increases for the ions within the ionizer due to the low extraction field with respect to the thermal velocity distribution of ions.

It is possible to conclude that the time structure of an ion bunch can give information about the ionization location and propagation behavior of atoms. This information can be used to analyze the ionization efficiency and optimize each ion source configuration.

5.3.2.2 Time structures of LISBET ion sources

Time structures were measured for all LISBET geometries with the modified low intensity ion beam profiler. Each measurement corresponds to the integration of $6 \cdot 10^6$ ion pulses. The time structures were measured for 7 mm diameter ionizers and 3 mm diameter ionizers ([fig. 5.25](#)). All measurements were shifted $5 \mu s$ in the figures for a better visualization of the narrow peak. A variation of the time structure shape was observed as a function of the temperature.

In the case of 7 mm diameter ionizers, the time structure at low temperatures (1400 K) showed a prominent first narrow peak (Np), a second peak (Sp) and a small main peak (Mp). Increasing the temperature reduced the narrow peak and the second peak, and increased the main peak.

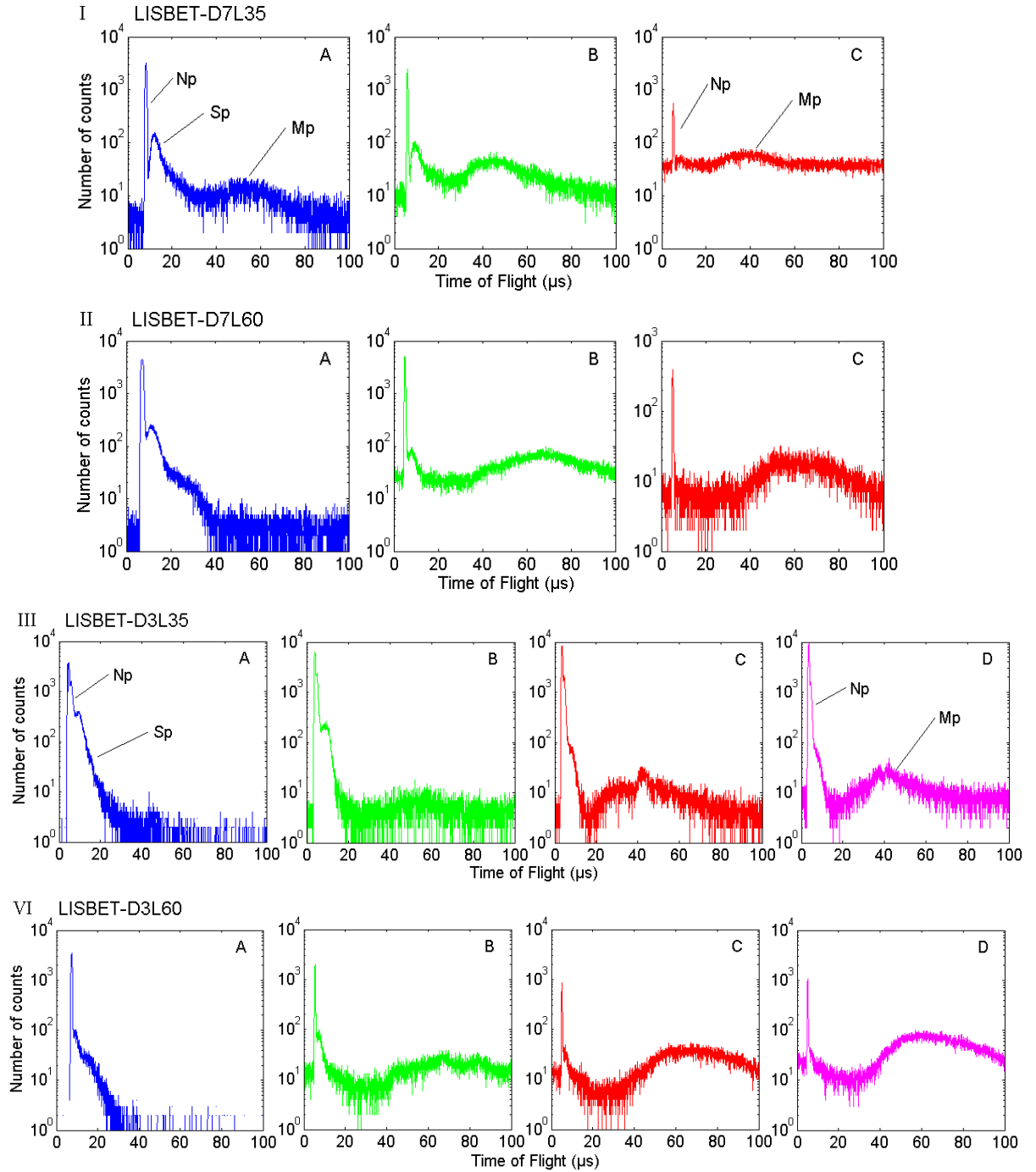


Figure 5.25— Time structure measurements for LISBET-D7L35 (I), LISBET-D7L60 (II), LISBET-D3L35 (III) and LISBET-D3L60 (IV) at temperatures: (A) 1400 K, (B) 1700 K, (C) 1900 K and (D) 2100 K. *Np* stands for narrow peak, *Sp* for second peak and *Mp* for main peak.

No main peak was observed for **LISBET-D7L60** at low temperatures (1400 K) (fig. 5.25/II). The main peak appeared at medium temperatures (1700 K). At high temperature (1900 K) only the narrow peak and the main peak were observed for both LISBET configurations. In all cases, the size of the narrow peak was proportionally smaller at high temperature, compared to the one measured at low temperature.

On the other hand, 3 mm diameter ionizers showed a similar behavior. The narrow peak (Np) decreased with increasing temperature and the main peak (Mp) appeared at 1700 K (fig. 5.25). The second peak (Sp) was placed closer to the narrow peak (Np) and it did not disappear completely within the heating process.

It is also possible to observe that the main broad peak position changes depending on the ionizer length (fig. 5.25). The average position and width of main peak for different LISBET configurations can be found in table 5.11. In the case of **LISBET-D3L60**, the main peak was cut due to the recording window limit of 100 μs .

Table 5.11 – Summary of the measured main peak positions at 1900 K for different LISBET configurations. The main peaks are referenced to the first narrow peak.

	Position (μs)	Width (μs)
LISBET-D7L35	35	~ 30
LISBET-D7L60	55	~ 90
LISBET-D3L35	35	~ 35
LISBET-D3L60	55	~ 70

To analyze the evolution of the time structures as a function of the temperature, the integrated number of ions in the main peak ($N_{MainPeak}$) was divided by the total number of ions recorded (N_{Total}) (fig. 5.26). For low temperatures, most of the ions were generated in the narrow peak therefore the ratio is low.

However, this behavior changes for high temperatures where the quantity of ions included in the main peak increases. **LISBET-D7L35** and **LISBET-D7L60** reached 80% of ions included in the main peak at 1900 K. On the other hand, **LISBET-D3L60** reached this value at 2000 K and **LISBET-D3L35** at 2200 K.

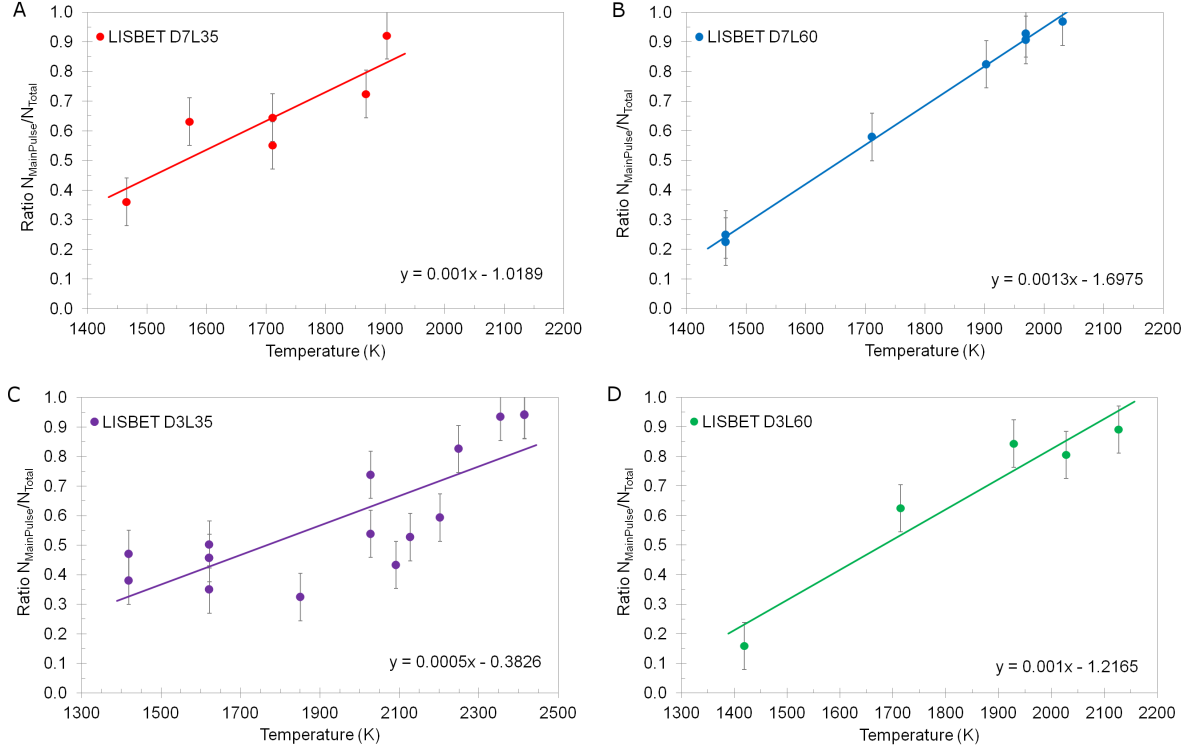


Figure 5.26– Ratios between the ions in the main broad peak ($N_{MainPeak}$) divided by the total number of ions (N_{Total}) as a function of the temperature for (A) LISBET-D7L35, (B) LISBET-D7L60, (C) LISBET-D3L35 and (D) LISBET-D3L60.

5.3.3 Simulations

The simulation described above was also used to calculate the time structures of the different LISBET configurations (fig. 5.27). The ions were generated with initial velocity direction of 30 degree cone distribution to increase the number of particles getting out of the ion source, due to the high neutralization inside the ionizer. This artifact gives a similar efficiency trend compared to the isotropic distribution but higher statistics were recorded.

The narrow peak and the main peak are also present in the simulations. Longer ionizers result in a broader time-of-flight distribution of the ions. No difference was observed with a variation of the ionizer diameter. The simulated ions reached the detector in less time compared to experiments. However, some parameters as the electric field potential (probably lower than estimated in the measurements due to losses in the connections) can increase the time-of-flight of ions. A satisfying explanation could not be found to explain the presence of the measured second peak with the simulation.

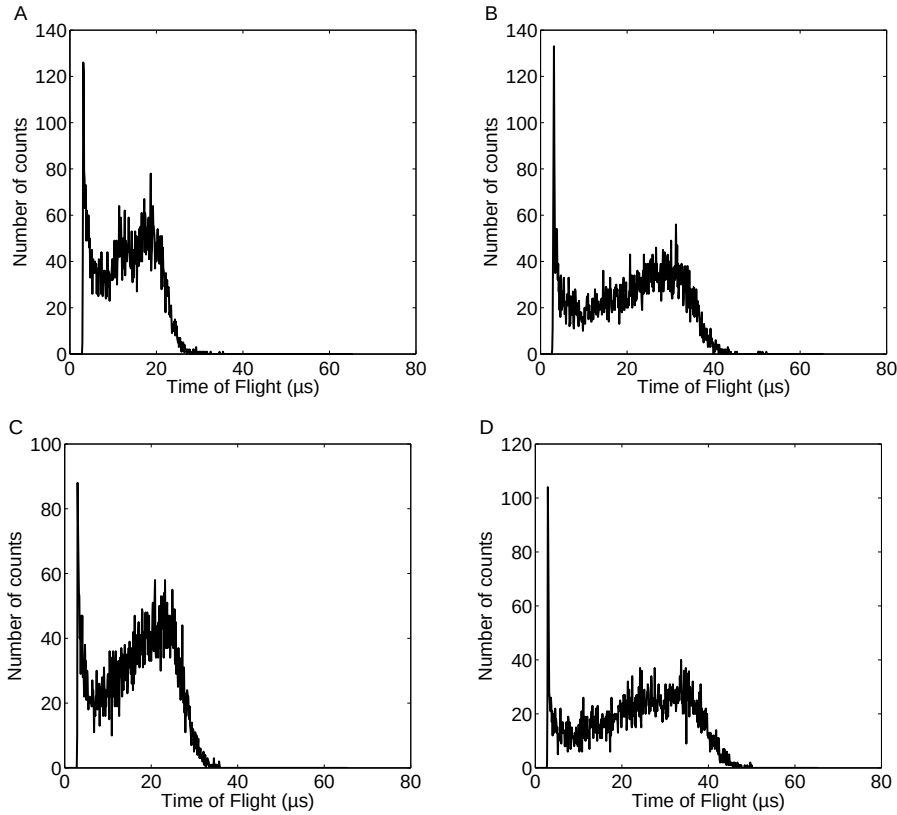


Figure 5.27– Time structure simulation for the different LISBET configurations at 2000 K. (A) LISBET-D7L35, (B) LISBET-D7L60, (C) LISBET-D3L35 and (D) LISBET-D3L60.

Figure 5.28 shows the initial position of the ions and the resulting detected time structure. The ions at the exit of the ionizer are fast extracted due to the extraction potential gradient, while the ions generated inside the tube define the main peak. It was observed that the ions generated at the entrance of the ionizer (close to the elbow) have a higher speed due to the higher electric potential in this region. These ions gather the ions generated in the middle of the tube (with relative less speed) defining the shape of the main peak.

5.3.4 Time structure discussion

The time structures can change when the ion source geometry changes. As already shown, the diameter of the ionizer does not affect the main peak, while the ionizer length modifies the position and width of the main peak. A change of the ionizer length changes the distance the ions have to travel to exit the cavity, as explained in the time-of-flight analysis ([fig. 5.24](#)). For that reason the main peak shows a temporal spread in the case of 60 mm length ionizers.

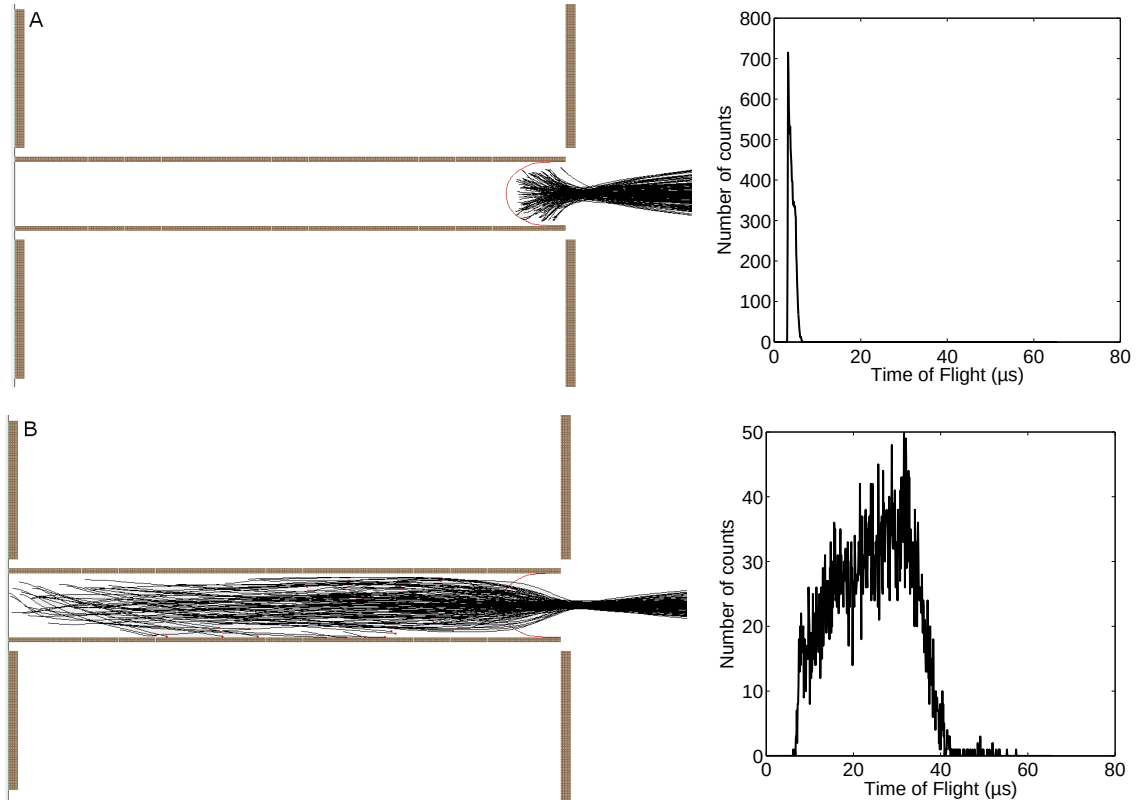


Figure 5.28– Simulation comparing the initial position of ions: Generated at the exit of the ionizer (A) or inside the ionizer tube (B). In the first case the ions are generated in a narrow peak and in the second case the are spread in time.

The narrow peak was noticed for all geometries and it was also observed in the simulations. The narrow peak height changes as a function of the temperature, decreasing for more than 1400 K. A relation between the ions in the main peak and the ions in the narrow peak was calculated, where most of the ions were included in the main peak at 1900 K (for 7 mm diameter ionizers) and 2100 K (for 3 mm diameter ionizers).

A second peak structure was also observed which disappeared for 7 mm diameter ionizer at 1900 K, but it did not completely disappear for 3 mm diameter ionizer at 2100 K. A hypothesis is that the narrow peak consists of the ions generated in the 19 kV extraction region that are fully accelerated when ionized, and the second peak are the ions generated close to the 19 kV extraction region. Since the electric potential is higher in the 7 mm diameter ionizers, the ions in the second peak are merged together in the narrow peak.

The main peak appeared for all LISBET configurations at 1700 K. It consists of the ions coming from the inside of the ionizer. It is broader in the case of longer ionizers, with similar width for both diameters. The main peak shape shows that the ions are merged due to different acceleration speeds. This also generates a depth between the narrow peak and the main peak observed in the simulation.

The main peak tail was cut in **LISBET-D7L60** and **LISBET-D3L60** due to electronic restrictions. The TAC module triggered by the master-clock laser signal only allows an acquisition window of 100 μ s. A possible development to increase the total time structure window could be to increase the time between 2 pulses, with the consequent extension of the acquisition window. This could permit to distinguish between different ion beam time structures.

The time structures were also analyzed in the simulations where the narrow peak and the main peak could be reproduced. The structures measured in the simulations are faster than the experiments. The possible reasons are that the extraction potential used in the simulations was the one measured in the terminal connectors. As already commented, the real electric potential has to be lower due to the electric connectors between the electric terminals and the ion source. A lower electric potential will affect the propagation speed of the ions. In additions, the change of atomic vapor density along the ion source cavity was not taken into account. Nevertheless the peak was placed in a similar environment and the form of the different structures are similar in shape.

5.4 Energy distribution scans

The energetic distribution of an ion beam is a fundamental information for its proper selection, transport and acceleration. In order to measure the energy distribution, the time structure measurement method was combined with the slit-grid method, used in the emittance measurement (section 5.2). The result of the measurement is the ion beam spatial distribution of each ion compared to the time of flight of individual ions.

5.4.1 Method description

The idea is to perform time structure measurements with the slits closed to its maximum (0.3 mm). In this configuration the ion beam is scanned by moving the slits in the whole dipole selection range. Each slice of the ion beam represents a range of energies selected by the magnetic mass separator (fig. 5.29). As the magnetic mass spectrometer is an energy selector, it is possible to transform each position of the beam to a defined energy. Valuable information of the ion beam energy drift can be obtained from the energetic distribution of the ion bunch from the inside of the ion source. However, the energy selected by the magnet mass spectrometer is the convolution between the energy distribution and the spatial distribution of ions. The measurements can only provide a qualitative measurement of the energy dispersion.

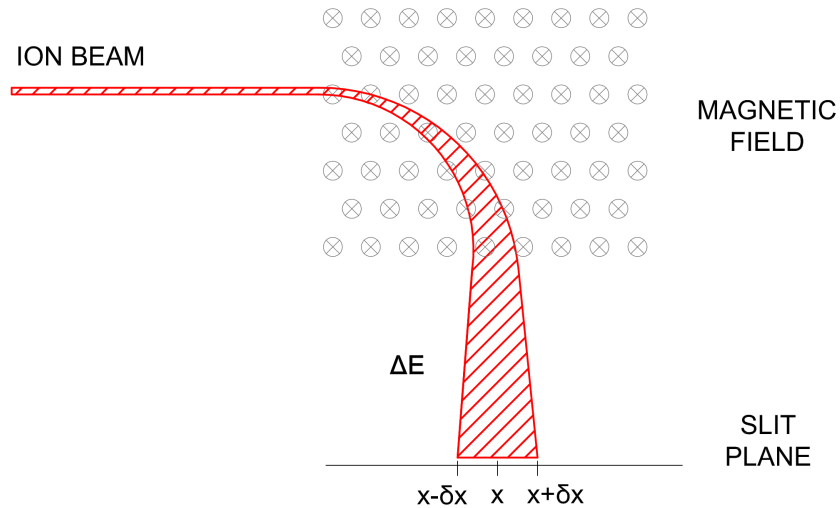
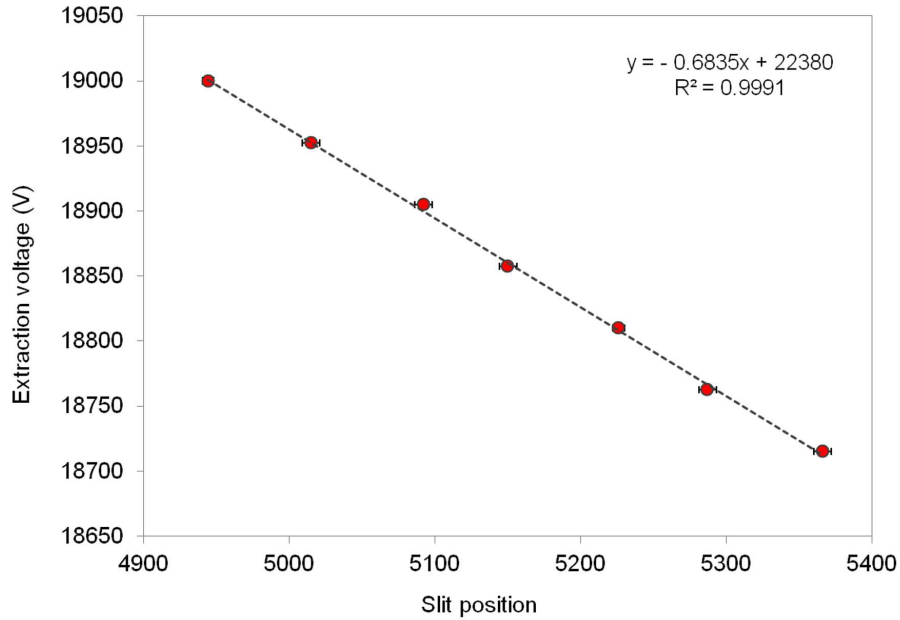


Figure 5.29– Diagram of the ion beam energy scan. Each position of the slits (x) corresponds to an energy selected by the magnet mass spectrometer.

5.4.1.1 Energy window

The main limitation of this technique is the resolution of the magnet mass spectrometer and the energetic dispersion ΔE in the acquisition point. Considering that the magnetic mass selector deviates the ions by its kinetic energy, it is possible to approximate the energetic dispersion at the exit of the dipole by measuring the deviation of the ion beam when the kinetic energy of the particles is increased or decreased (i.e. the extraction electric potential set in the ion source platform was decreased or increased). The variations of energy up to 1.5% are shown in [figure 5.30](#) and the consequent deviation can be observed in the slit position.



[Figure 5.30](#)– Calibration of the kinetic energy centroid as a function of the position after the dipole. The applied energy is represented as a function of the slit position (errors are included). One slit step corresponds to 0.03 mm

5.4.2 Energy distribution for the LISBET-D7L35 geometry

The energy distribution of each ion compared to its time of flight was recorded. The measurement was performed using the slit grid method, where the ion beam was scanned by the use of a pair of slits closed to its maximum. Each slice of the ion beam (slit position) represents a range of energies selected by the magnetic mass separator. The result of the energy scan for **LISBET-D7L35** can be observed in [figure 5.31](#) compared to a conventional time structure measurement. It has the same components as the time profile (first narrow peak and broad main peak) but the energy distribution can be observed too.

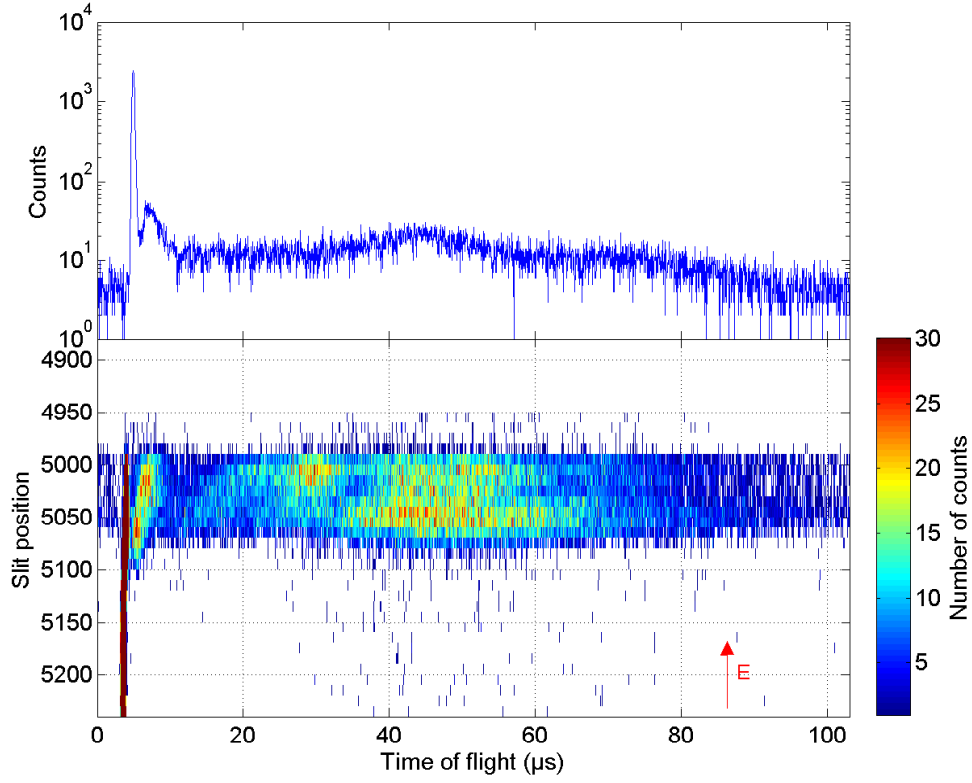


Figure 5.31– Time profile measurement of ^{124}Sn (above) and the energy distribution measured by the magnetic mass spectrometer (below) for LISBET-D7L35 with same conditions. The increment of the extraction energy is marked by an arrow.

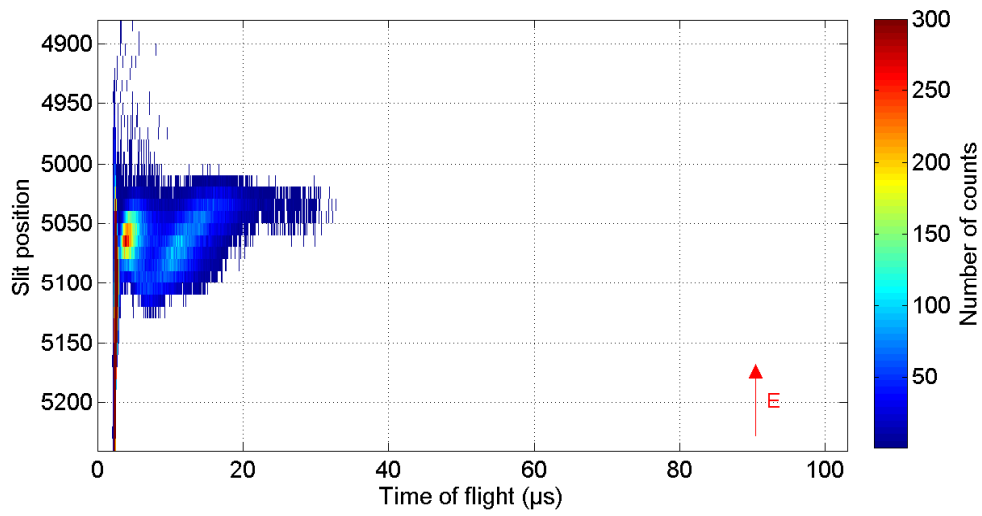


Figure 5.32– Energy distribution measured in the same conditions as figure 5.31 reversing the electric field potential in the ionizer.

The time profile of the energy scan showed a narrow peak containing ions with a broad distribution of energies, from the extraction energy (19 keV) to lower energies, which ended in a long and smooth tail. The main peak (the ions coming from the inside of the ionizer) was perfectly confined in a range corresponding to the extraction energy and the electric potential along the tube.

In addition, a scan of the energy distribution was performed by reversing the electric field in the atomizer (fig. 5.32), where only the ions produced near the exit were extracted. From the comparison of figures 5.31 and 5.32 it is possible to observe that, in the latter, the ions generated inside the ionizer tube cannot escape due to the electric field potential. They are neutralized when touching the cavity walls or they are confined backwards to the transfer/atomizer tube. The electric potential measured in the terminal connectors was approximately 5 V for both.

5.4.2.1 Simulations

The energy scans were simulated with SIMION for all the LISBET. The results of the simulations are shown in figure 5.33. The ions were only generated inside the ionizer.

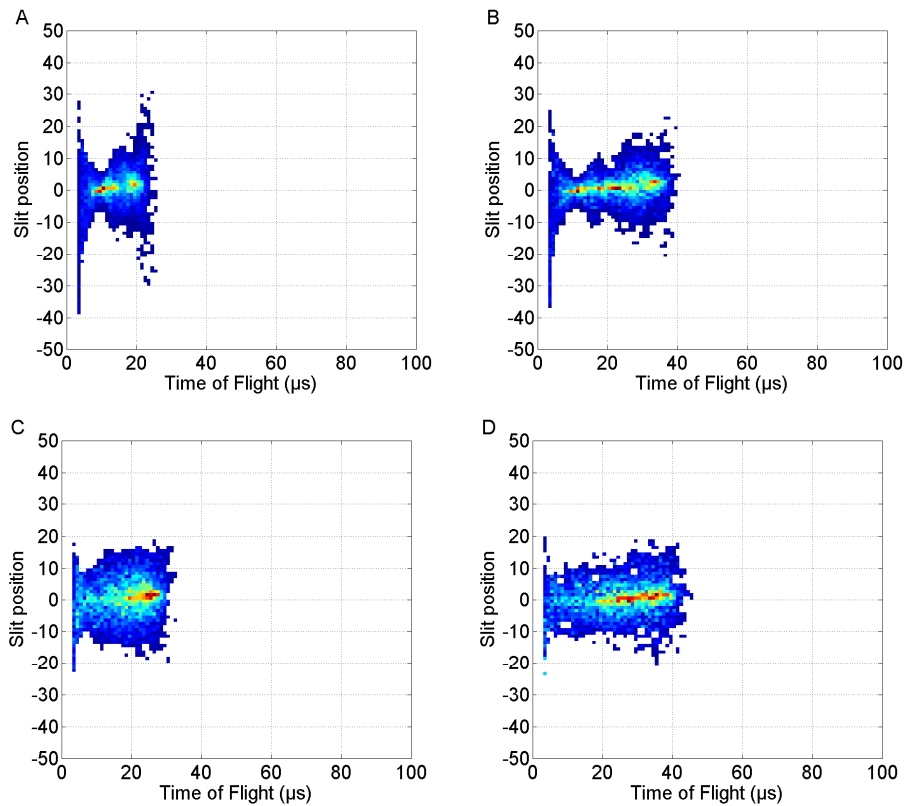


Figure 5.33– Simulations of energy scans for the different LISBET configurations: (A) LISBET-D7L35, (B) LISBET-D7L60, (C) LISBET-D3L35 and (D) LISBET-D3L60.

The simulations reproduced the structures observed in the experimental energy scans: A broad energy distribution in the case of the narrow peak and a confined structure, for the ions coming from the inner part of the ionizer tube (yellow and red areas in the figures).

However, a different behavior was observed for each ion source geometry. An increasing in the ionizer length provided ions with higher time-of-flight (as already discussed in the time structure section). In the case of 7 mm diameter ionizer, the experiments showed a much broader energy dispersion of the narrow peak than in the simulations.

The energy distribution of the ions from the inside of the ionizer depends on the electric potential applied to the ionizer. Figure 5.34 shows the kinetic energy in the detection point as a function of the starting position of ions for **LISBET-D7L60**. The ions generated at the beginning of the ionizer have an energy slightly higher than the extraction potential (19 kV) due to the electric potential in the tube. Moreover, the ions generated at the exit of the ionizer have an energy that depends on the extraction potential in the region where they were ionized. A particle generated 2 mm from the exit of the ionizer has an energy of 18.9 keV instead of 19 keV.

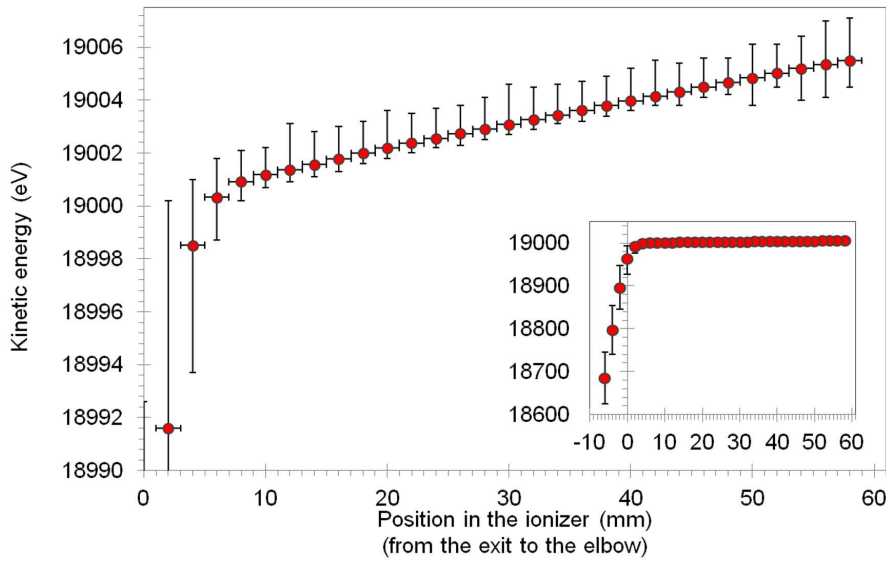


Figure 5.34— Ion beam simulation where the starting position of the ions in the ionizer was compared to their time of flight. The zero position was set at the exit of the ionizer tube. The vertical error bars come from the temporal spread of the ion bunch arriving to the detection point.

Figure 5.35 presents the simulated energy spread of each LISBET configuration. The prominent peak between 19000 and 19006 eV corresponds to the ions from the inside of the ionizer, at approximately 19 kV (the extraction potential).

The long tail before this peak corresponds to the ions generated in the extraction potential region, with an energy that depends on the equi-potential extraction line where they were ionized. The confined peak showed a distribution of energies $\Delta E \sim 5$ eV for all the LISBET configurations.

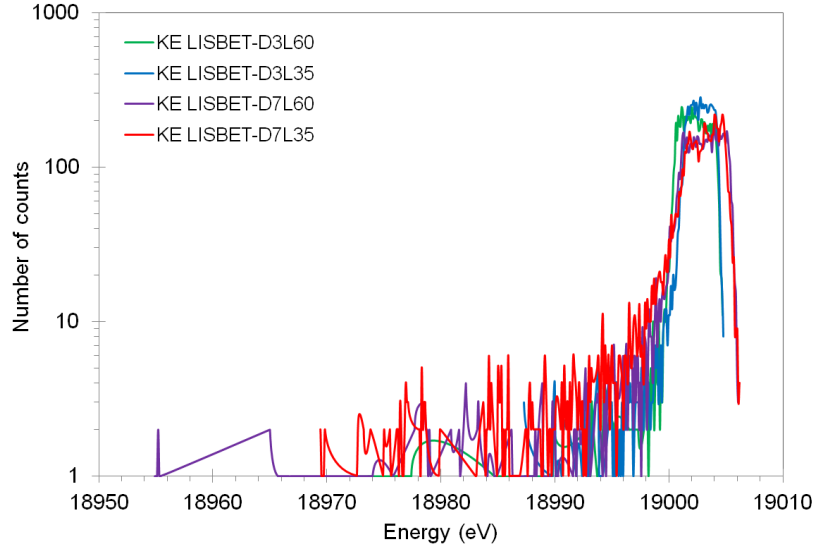


Figure 5.35– Simulated energy spread for each LISBET configuration. The number of counts is presented as a function of the energy.

5.4.3 Energy scan discussion

It was observed that the narrow peak on the time profile of the energy scans consisted of a broad energy distribution, while the main peak had a well defined energy distribution. This time structure could be reproduced for the different LISBET geometries in the simulations shown in figure 5.33. The energy dispersion of the narrow peak was also observed in the emittance simulations (fig. 5.17) in section 5.2.2.1, where some ions were drifted to the left part of the figure.

An hypothesis to explain the broad energy distribution of the narrow peak is the following: The ions generated at the exit of the ionizer may have less energy if they are ionized in a region where the extraction potential decreases. It was already discussed that the extraction field potential penetrates several millimeters inside the ionizer (fig. 5.10), thus it creates a extraction potential gradient with the consequent selection by the mass spectrometer at lower energies.

In addition, the penetration of the extraction field potential creates a focalization effect (explained in section 5.10), where the focal point is placed inside the ionizer at a distance of few mm from its end. This increases the ion spatial dispersion. This hypothesis could also explain why the narrow peak showed distributions (at slit position) in a higher energies compared to the extraction energy, since the spatial distribution has also an effect in the measurement.

The relative quantities of ions in the narrow peak and in the main pulse have been measured. The results show that 7 mm diameter ionizers ensure more than 90% of ionized atoms confined in the proper energy at 1900 K, while ion source geometries with 3 mm diameter have to reach 2200 K to ensure the same percentage.

With the energy scan method, it is not possible to know the precise energy of the ion bunch, just the convolution between the energy and the spatial spread. For this reason the energy distribution of the confined ions was considered the one analyzed by the simulations, which has an energy dispersion of ~ 5 eV for the ions confined in the ionizer. Considering this value for the main peak, one can deduce that the energy distribution of the ions in the narrow peak could be larger than 100 eV when they are ionized at few millimeters from the ionizer exit.

The narrow peak poses the problem of selection in the mass separator system, since the ions cannot be properly selected due to the broad energetic distribution, while the main pulse is perfectly selectable. The ions produced with such energy distribution will pass through the mass separator to the focal plane positions corresponding to different masses, thus produce contamination for lower masses.

The broad energy distribution of the narrow peak may cross other isotopes with the consequent isotopic contamination. For example, the ^{117}Sn isotope may contain contributions from the narrow peaks of ^{118}Sn , ^{119}Sn , ^{120}Sn , etc. isotopes, which are selected by the magnetic mass spectrometer (fig. 5.36). This isotopic contamination may destroy the selectivity and it can affect the ultra-trace determination [210] and the measurements of isomers [211]. This hypothesis has been considered in some studies [212], but further analysis and development will be needed in order to reduce the afore mentioned effects.

In the efficiency measurements, ions are coming from the narrow and main peaks. However, below temperatures of 1400 K no ions from the main peak were recorded and most of the ions came from the narrow peak. Some authors developed the hypothesis that a plasma potential could be generated at certain temperatures that might confine ions with thermal energies [117, 208]. This plasma potential would reduce the neutralization of the ions as it prevents the ions from hitting the cavity walls. The appearance of plasma potential is temperature dependent (e.g for Ta cavity $T > 2200\text{K}$ is needed). In our case, the electric potential was the only effect considered due to the lower temperature used, compared to high temperature surface ion sources.

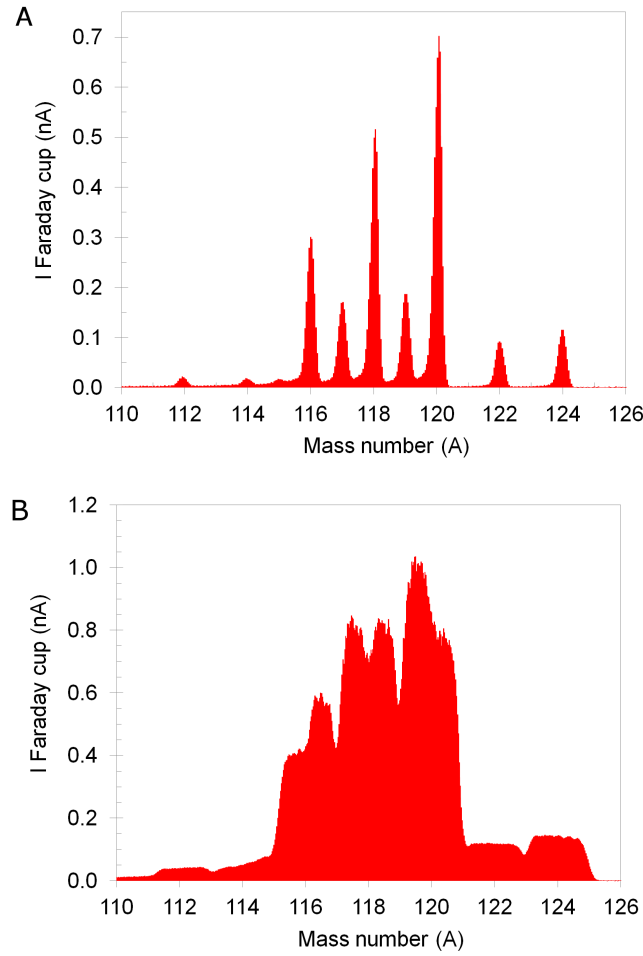


Figure 5.36– Mass spectrum of Sn with the generated isotopes. (A) Scan with the metal plate slits in normal position and (B) metal plate slits wide open. The spectrum is broadened due to the energy displacement of one of the isotopes with respect to the other masses.

In conclusion, the temperature can increase the quantity of usable ions since they are generated in the energy-confined main pulse. On the contrary, the ions cannot be properly selected and used in an accelerator if the narrow peak is dominant. Further studies will be needed to understand and reduce the effect of the narrow peak for the ion beam selection.

5.5 Research for the improvement of the selectivity

The RILIS technique is known for its atomic selectivity. However, as discussed in section 2.4, the hot-cavity behavior suffers from a decrease of the selectivity due to the generation of unwanted contaminants and molecular sidebands, which contribute to the ion beam. The surface ionization occurs during thermal desorption of atoms from the hot-cavity walls at high temperature. The reduction of alkali contaminants in the hot-cavity is one of the main research fields for the RILIS technique. In this chapter, the studies carried out to diminish the alkali production will be described. The relative contamination reduction of an alkali marker was measured in order to analyze the contamination suppression.

5.5.1 Element of study

A representative surface ionization element was chosen, which cannot be affected by external contamination. The possible contaminant marker options were: Na, Al, K, Rb, Cs, Ca and Ba.

Table 5.12 – Representative surface ionization elements, which could be used as contaminant marker. IP is the ionization potential and I_{dipole} the dipole current where are found during the experiments.

Element	IP (eV)	Reference isotope	Natural abundance (%)	I_{dipole} (A) [HV = 19 kV]
Na	5.1391	^{23}Na	100	74.04
Al	5.9858	^{27}Al	100	80.39
K	4.3407	^{39}K	93.26	97.28
Rb	4.1771	^{85}Rb	72.15	147.95
Cs	3.8939	^{133}Cs	100	176.12
Ca	6.1132	^{40}Ca	96.97	
Ba	5.2117	^{138}Ba	71.66	192.47

Among the elements included in table 5.12, Na, K, Ca and Ba were discarded because they are present in the background of the hot-cavity ion source signal when heating. This presence will mask the sample release at high temperatures. Al, Rb and Cs are good candidates because they were not found in the background. Al has a relative high vapor pressure (melting point at 1200 K for a pressure $\sim 10^{-6}$ mbar), thus high temperature will be needed to perform reliable measurements with the consequent consumption of time.

Cs release is too fast due to its high vapor pressure, thus high temperature tests cannot be performed with small size samples. On the other hand, while increasing the sample size risks of ion source pollution.

Finally, the chosen candidate was Rb due to its intermediate alkali ionization potential, its high vapor pressure and its boiling point at 961 K. These intermediate properties and the fact that Rb is not present as impurity in the ion source background, make Rb a suitable and reliable contaminant marker.

5.5.2 Electric field potential comparison

The contamination reduction was tested by the control of the electric field potential generated due to resistive heating. As presented in section 4.1.2, the DC electrical heating current flowing through the ionization and transfer/atomizer tube is expected to create a weak electric field that would guide the ions axial movement [114]. This electric field can modify the direction of the surface ion motion and that of the laser ionized atoms. The effect of the electric field was observed for Sn ions in the previous sections. In the LISBET ion source, the direction of the electrical current can be chosen separately for the ionizer and the atomizer (section 4.1). This direction was chosen in such a way that the resulting electric field drifted the ions toward the extraction potential or rejected them back.

Figure 5.37 shows the four solutions proposed to control the electric field direction. Rubidium was chosen to measure the total ion current by placing a sample with a known amount of atoms into the atomizer (described in section 5.1.2). For these experiments, the **LISBET-D7L35** configuration was used (7 mm diameter and 35 mm length ionizer tube). The isotope ^{85}Rb , with a natural abundance of 72.15%, was considered for the efficiency studies. The samples were prepared with a total amount of $5.1 \cdot 10^{13} \pm 4 \cdot 10^{12}$ and $2.5 \cdot 10^{14} \pm 2 \cdot 10^{13}$ ^{85}Rb atoms. The relative total ion current signals for each solution was used to analyze the relative contamination reduction. One measurement was performed for each electric field configuration.

The atomizer has a cap that closes the tube to prevent the sample from falling down. This cap has also the effect of recycling the surface ions inside the atomizer. In the on-line target ion source system the cap is replaced by the target container, which acts as a cavity. The ions created in the transfer tube can penetrate the target container and be neutralized, delayed due to effusion and lost due to radioactive decay. Therefore, measurements without cap were performed to simulate the presence of the target container.

In the case without the cap, it was considered that the sample releases 50% of Rb atoms to the atomizer and the other 50% to the target cavity. A different reduction factor is expected with and without the atomizer cap. For this reason, the measurements were performed with and without the cap.

The relative efficiency in the case of the ion source with the cap can be considered as a lower limit of the reduction factor due to recycling effects in the atomizer (Δ_{low}). On the other hand, the relative efficiencies without the cap can be considered as a higher limit of the

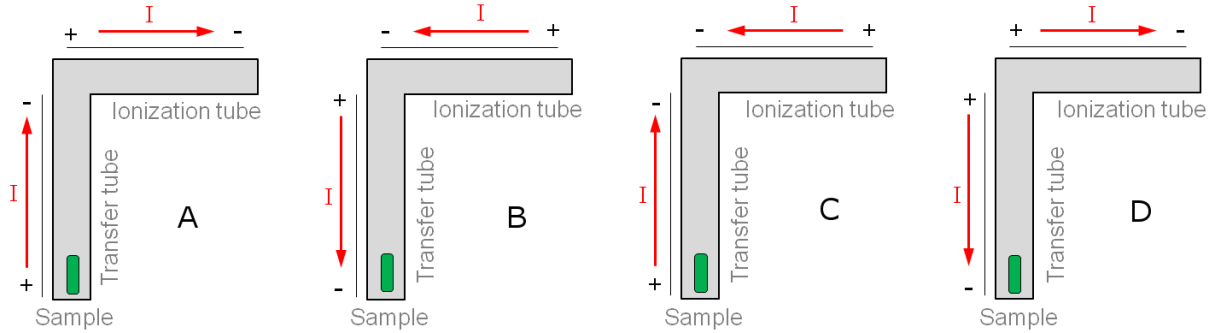


Figure 5.37– The possible configurations for the electric current in the ion source: (A) electric field towards the extraction region. (B) electric field repelling all ions backwards to the “target place”; (C) electric field forward in the transfer tube and backwards for the ionization tube; (D) electric field repelling the ions generated in the transfer tube and pushing forward the ions generated in the ionization tube.

reduction factor (Δ_{high}). In this case, the re-absorption of the impurities by the target cavity was considered. Also radioactive isotopes could decay in the target cavity. The real values for the contamination reduction should be between both values.

The efficiencies of each option (electric field and low function material) were compared to these two reference values (Δ_{low} and Δ_{high}) to obtain the contamination reduction factor. Samples with $5.1 \cdot 10^{13} \pm 4 \cdot 10^{12}$ ^{85}Rb atoms were used to measure Δ_{low} and samples with $2.5 \cdot 10^{14} \pm 2 \cdot 10^{13}$ ^{85}Rb atoms were used to measure Δ_{high} . The sample with $5.1 \cdot 10^{13}$ atoms comes from an old standard solution and the calibration could not be assured (this also explains the higher error provided by this sample). However, the unknown calibration does not affect the relative reduction factor.

5.5.2.1 Contamination reduction results

The electric field potential was set in order to produce an electric field potential that pushes the ions towards the extraction system (option A of [figure 5.38](#)). An efficiency of $75 \pm 12\%$ was obtained with the tube cap, while $29 \pm 3\%$ was obtained without cap. These efficiencies were considered as the references Δ_{low} and Δ_{high} . In these cases the surface ions drift forwards in both the atomizer and the ionizer, giving the highest ion extraction efficiency. In [figure 5.38](#) the total ion current during the measurement with the atomizer cap can be observed.

The second option was to set the electric field in order to produce an electric field potential that repels the ions to the target cavity (option B). The ions will be stored in the transfer tube. They can be re-injected in the case of having the cap or can be thrown out without the cap.

5.5. Research for the improvement of the selectivity

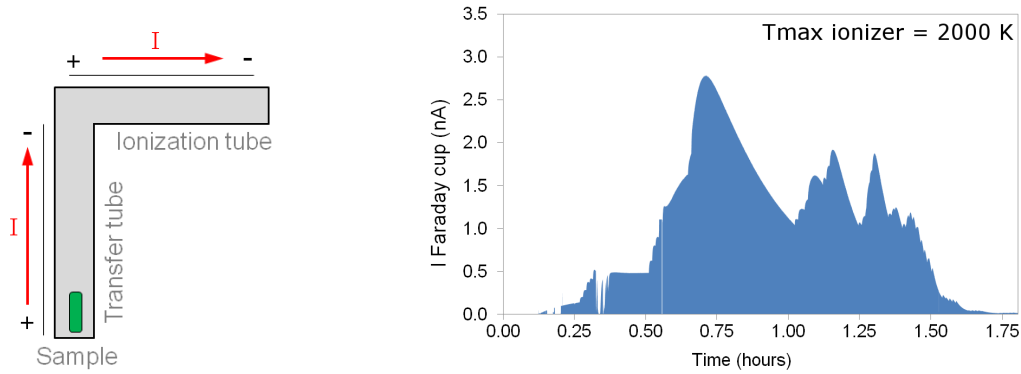


Figure 5.38– (Left) Configuration of the electric current in the ion source used for the ion current measurement. Option A: electric field towards the extraction system (forwards). (Right) Ion current measured as a function of time.

In the option B, an efficiency of $37 \pm 8\%$ was obtained with the cap (reduction factor of 2 ± 0.5 compared to Δ_{low}), while $14 \pm 2\%$ was obtained without the cap (reduction factor of 2.0 ± 0.4 compared to Δ_{high}). The [figure 5.39](#) presents the total ion current recorded during the experiment. As the surface ions are captured by the electric field, most of them cannot reach the extraction electrode potential.

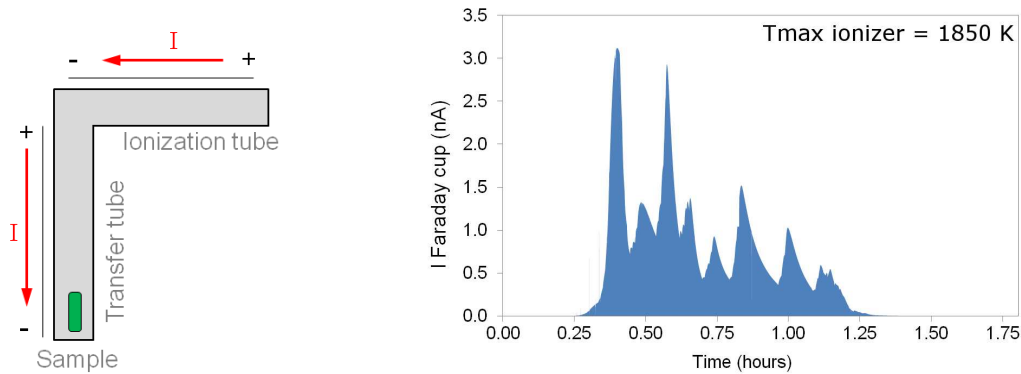


Figure 5.39– (Left) Configuration of the electric current in the ion source used for the ion current measurement. Option B: Electric field potential towards the target (backwards). (Right) Ion current measured as a function of time.

In the third case, the electric potential was set in order to generate a dip going to the elbow of the ion source body (option C). The surface ions generated in the transfer tube will be sent forwards and the ions generated in the ionization tube will be pushed back.

Option C is not a convenient option because the surface ions will be accelerated to the ionizer and the laser ionized elements will be rejected back. However this option was measured in order to complete the study.

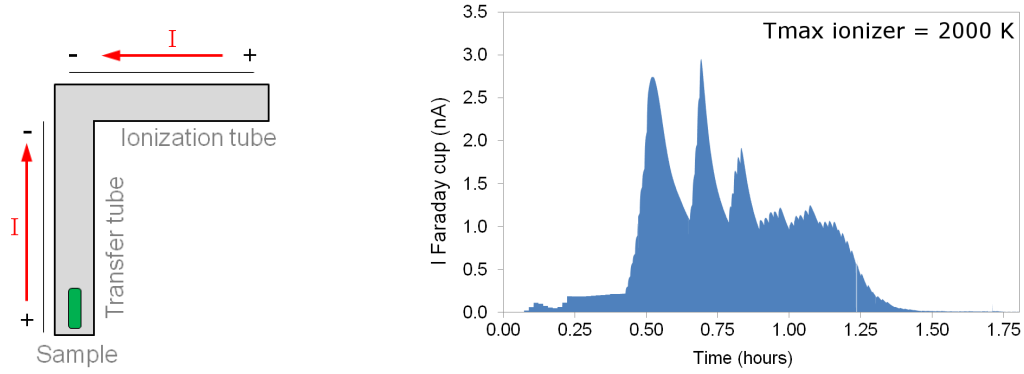


Figure 5.40– (Left) Configuration of the electric current in the ion source used for the ion current measurement. Option C: Electric field potential backwards for the ionization tube and forwards for the transfer tube. (Right) Ion current measured as a function of time.

The measured efficiencies were $54 \pm 9\%$ with cap (reduction factor of 1.4 ± 0.3 times Δ_{low}) and $17 \pm 2\%$ without cap (reduction factor of 1.7 ± 0.3 times Δ_{high}) respectively (fig. 5.40).

Finally, the option D was investigated. In this case, the direction of the electrical current of the ionization tube was chosen in order to create an electric field that pushes the ions generated in the ionization tube towards the extraction region, whereas the direction of the electrical current for the transfer tube was set to repel back the surface ions generated in the transfer tube (figure 5.41).

The D solution combines the contaminant reduction properties without compromising the ion production rate. For that reason, it was used for most of the results presented in this thesis. This option provided an efficiency of $45 \pm 8\%$ with cap (reduction factor of 1.7 ± 0.4 times Δ_{low}) and $12 \pm 1\%$ without cap (reduction factor of 2.4 ± 0.4 times Δ_{high}).

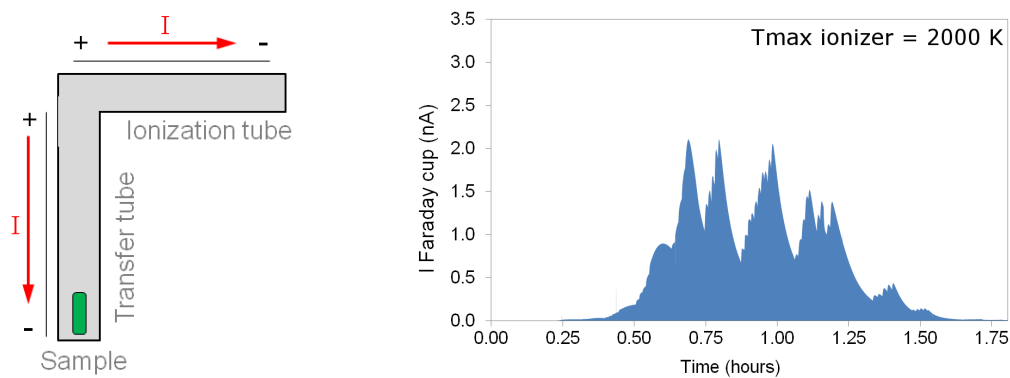


Figure 5.41– (Left) Configuration of the electric current in the ion source used for the ion current measurement. Option D: Electric field potential forwards for the ionization tube and backwards for the transfer tube. (Right) Ion current measured as a function of time.

5.5.3 Effect of ZrC as contamination reduction agent

The possibility of using a material with a low work function material was studied in order to reduce the ionization and recycling of alkali contaminants (section 2.4.2). For this purpose, a rod of **ZrC**, with an inner diameter of 3 mm and an outer diameter of 7 mm, was inserted into the **LISBET-D7L35** configuration.

A test was performed with the configuration D+ZrC of the electric field (repelling backwards the ions generated in the transfer tube and pushing forwards the ions generated in the ionization tube) to determine the expected reduction in surface ionization figure 5.42.

The efficiency with the low work function material inside LISBET was $15 \pm 1\%$ with cap (reduction factor of 5 ± 0.9 times Δ_{low}) and $2.7 \pm 0.3\%$ without cap (reduction factor of 11 ± 2 times Δ_{high}).

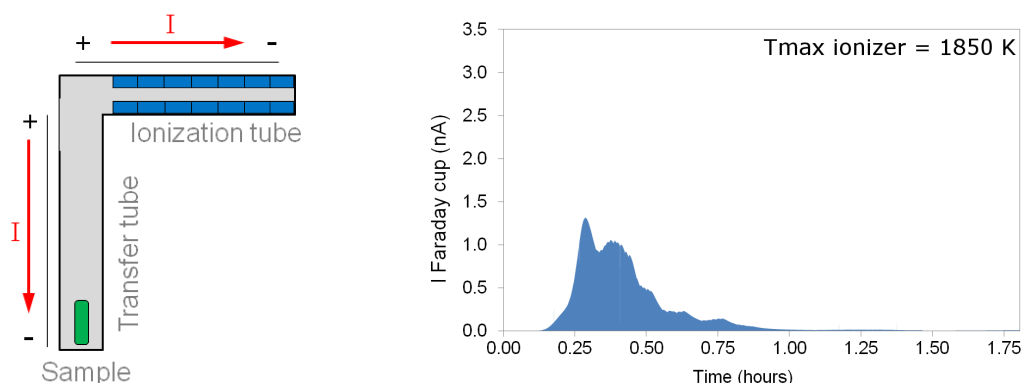


Figure 5.42– (Left) Configuration of the electric current in the ion source used for the ion current measurement. Option D+ZrC: Electric field potential set in the same conditions as D, but the ZrC ceramics were inserted into the ionizer. (Right) Ion current measured as a function of time. The vertical axis is constant for all figures to observe the ion current reduction.

5.5.4 Study of the contamination reduction due to the ionizer diameter

A measurement using **LISBET-D3L35** was carried out in order to distinguish the contamination suppression due to the use of **ZrC** and due to the geometry. The insertion of **ZrC** modifies the inner diameter of the ionizer, which could reduce the conductance of ions from the atomizer/transfer tube to the ionizer tube. This decrease could modify the ionization behavior and mask the real effect of the ZrC material.

The transversal areas of ionizers with 7 and 3 mm diameters are 38.48 mm² and 7.07 mm², respectively. This means that decreasing the ionizer diameter from 7 mm to 3 mm decreases the conductance area by more than 5 times. It is also important to indicate that the transfer tube has a diameter of 7 mm. In this case the ions might find an easier path, while in the case of a 3 mm diameter the change between both cavities is abrupt and a lot of ions could be lost in the transition.

A sample containing $2.5 \cdot 10^{14} \pm 2 \cdot 10^{13}$ atoms was used instead the one of $5.1 \cdot 10^{13} \pm 4 \cdot 10^{12}$ atoms considered in the previous sections. In this way, the error associated with the number of atoms in the sample was minimized. However, a change of the sample size also required to repeat the reference measurement. For this reason, the Rb measurements with a 3 mm diameter ionizer with cap were compared to a new reduction factor Δ_{low2} .

The **LISBET-D3L35** device (3 mm diameter ionizer) was first set in the option A to measure the efficiency with the cap. An efficiency of $80 \pm 6\%$ was obtained, which was used as Δ_{low2} . Option D provided an efficiency of $25 \pm 2\%$ (reduction factor of 3.3 ± 0.3 compared to Δ_{low2}) and $2.8 \pm 0.2\%$ (reduction factor of 10 ± 1 compared to Δ_{high}).

5.5.5 Contamination reduction discussion

Table 5.13 gathers the results of the contamination reduction measurements. It was demonstrated that a reduction of the unwanted surface ions can be achieved by the control of the electric field direction in a resistive heating configuration. An approximate reduction factor of 2 was measured when reversing the electric field potential (option B). The configuration where the laser ionized atoms were accelerated in the ionizer and the alkalis were repelled in the atomizer (option D) also provided a contamination reduction factor of approximately 2.

In addition, the influence of **ZrC** as low work function material was studied. A reduction of the surface contaminants by a factor between 5 and 11 was obtained. However, the reduction can be due to the reduction of the volume in the LISBET device. As it was measured, a reduction of the ionizer diameter also reduced the generation of contaminant surface ions by a factor between 3 and 10.

Nevertheless, a small improvement can be observed if the lower factor of LISBET-D3L60 (factor 3) and lower factor of ZrC (factor 5) are compared. This could mean that the work function value is still too high and the ZrC structure has to be optimized. Values between 3.38 and 4.09 eV were found in the literature (section 2.4.2). A change in the manufacture parameters (by the University of Limoges) could provide a diminution of the work function value.

5.5. Research for the improvement of the selectivity

Table 5.13 – Reduction factors measured for the different electric potential directions. The symbol + indicates an electric field direction towards the extraction system and – corresponds to a backwards direction.

LISBET-D7L35 (Ionizer/atomizer)	Sample (μl)	Efficiency with cap	Reduction factor with cap Lower limit Δ_{low}	Sample (μl)	Efficiency without cap	Reduction factor without cap Higher limit Δ_{high}
Option A (+/+)	$5.1 \cdot 10^{13}$	$75 \pm 12\%$	1	$2.5 \cdot 10^{14}$	$29 \pm 3\%$	1
Option B (-/-)	$5.1 \cdot 10^{13}$	$37 \pm 8\%$	2 ± 0.5	$2.5 \cdot 10^{14}$	$14 \pm 2\%$	2.0 ± 0.4
Option C (-/+)	$5.1 \cdot 10^{13}$	$54 \pm 9\%$	1.4 ± 0.3	$2.5 \cdot 10^{14}$	$17 \pm 2\%$	1.7 ± 0.3
Option D (+/-)	$5.1 \cdot 10^{13}$	$45 \pm 8\%$	1.7 ± 0.4	$2.5 \cdot 10^{14}$	$12 \pm 1\%$	2.4 ± 0.4
ZrC ceramic (+/-)	$5.1 \cdot 10^{13}$	$15 \pm 1\%$	5.0 ± 0.9	$2.5 \cdot 10^{14}$	$2.7 \pm 0.3\%$	11 ± 2

LISBET-D3L35 (Ionizer/atomizer)	Sample (μl)	Efficiency with cap	Reduction factor with cap Lower limit Δ_{low2}	Sample (μl)	Efficiency without cap	Reduction factor without cap Higher limit Δ_{high}
Option A (+/+)	$2.5 \cdot 10^{14}$	$81 \pm 6\%$	1	$2.5 \cdot 10^{14}$		
Option D (+/-)	$2.5 \cdot 10^{14}$	$25 \pm 2\%$	3.3 ± 0.3	$2.5 \cdot 10^{14}$	$2.8 \pm 0.2\%$	10 ± 1

5.5.6 RILIS ion production in the LISBET device with ZrC

5.5.6.1 Efficiency of LISBET-D7L35 + ZrC for Sn

Zirconium carbide was studied as a low work function material for contaminant reduction. However, it is also important to know the production efficiency of the isotope of interest for further applications. The efficiency for Sn with **LISBET-D7L35 + ZrC** was determined (fig. 5.43) by obtaining a total efficiency of $10 \pm 2\%$ (table 5.14).

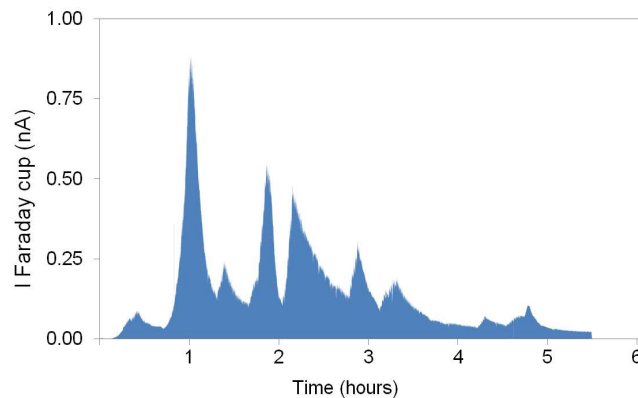
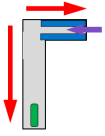


Figure 5.43– Ion current accumulated with LISBET-D3L35 + ZrC for ^{124}Sn efficiency measurement.

Table 5.14 – Efficiency results for LISBET-D7L35 + ZrC. The red arrows indicate the heating current direction. The size of the laser beam corresponds to the size of the ZrC inner diameter. ε_{raw} is the efficiency directly recorded. ε_{bkgd} is the efficiency after background correction. ε_{mean} is the mean efficiency of the measurements.

LISBET-D7L35 + ZrC

	Code	Sample (μ l)	Number of atoms	ε_{raw}	ε_{bkgd}	Error	Comments	ε_{mean}	Mean error
	#21	0.5	$1.5 \cdot 10^{14}$	11.8%*	11.7%	$\pm 2\%$			
	#22	0.5	$1.5 \cdot 10^{14}$	1.7%*	-	-	ZrC moved	10%	$\pm 2\%$
	#23	0.5	$1.5 \cdot 10^{14}$	10.3%*	9.8%	$\pm 2\%$			

The efficiency of **LISBET-D7L35 + ZrC** (3 mm inner diameter) was similar to **LISBET-D3L35**. The small difference between both measurements was explained due to the imperfections of the ZrC material, which did not fit perfectly with the Ta tube. The ZrC rods could be displaced into the ionizer tube with the consequent ionization in the extremity of the 7 mm diameter ionizer (fig. 5.44).

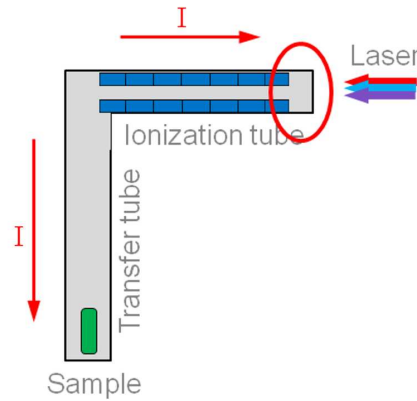


Figure 5.44– Example of an imperfect fit of the ZrC rods into the 7 mm diameter ionizer. It can produce an increase of the efficiency and the emittance due to the enhancement of the ionizer diameter.

5.5.6.2 Emittance results of LISBET-D7L35 + ZrC for Sn

The emittance of **LISBET-D7L35 + ZrC** was recorded to provide the properties of the ion beam. The laser beam sizes were set to a 3 mm spot, because the ZrC rod has an inner diameter of 3 mm. An rms emittance of $12.5 \pm 0.7 \pi \cdot \text{mm} \cdot \text{mrad}$ was obtained for a Sn ion beam.

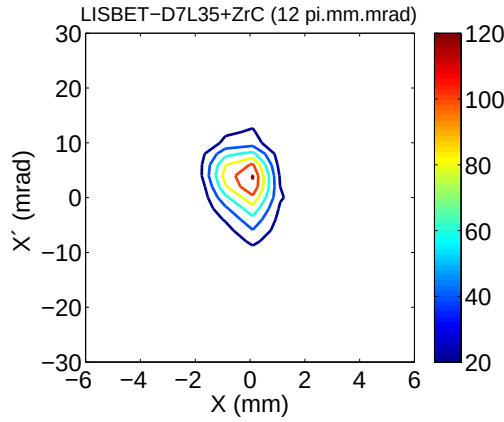


Figure 5.45– Rms emittance 2σ of LISBET-D7L35 + ZrC ($12.5 \pm 0.7 \pi \cdot \text{mm} \cdot \text{mrad}$).

Figure 5.45 shows the emittance iso-intensity contours for **LISBET-D7L35 + ZrC**. The insertion of the **ZrC** rod results in an effective diameter that is equivalent to the one of the **LISBET-D3L35** configuration. In the case of the **LISBET-D7L35 + ZrC** (3 mm inner diameter) an emittance close to the one of **LISBET-D3L35** was measured (12.5 and 9.5 $\pi \cdot \text{mm} \cdot \text{mrad}$ respectively). The increasing compared to the 3 mm diameter configurations, can be due to an imperfect fit of the ZrC rods inside the ionizer, as explained before.

5.5.7 Production discussion

A comparison was performed between 3 mm diameter ionizer, 7 mm diameter ionizer and the use of ZrC for the production of laser ions (Sn) and contaminants (Rb). In table 5.15 it is possible to observe that the 7 mm diameter ionizer is 3.4 times more efficient for laser ionization of Sn than the 3 mm diameter ionizer.

Table 5.15 – Comparison between the ion productions obtained with 3 mm diameter and 7 mm diameter ionizers. The ratios *high* and *low* compare the production of impurities with and without atomizer cap.

Ionizer	Sn production	Rb production (low)	Rb production (high)
7 mm diameter	$24\% \pm 3\%$	$45\% \pm 8\%$	$12\% \pm 1\%$
3 mm diameter	$7\% \pm 2\%$	$25\% \pm 2\%$	$2.8\% \pm 0.2\%$
Ratio	3.4	1.8	4.3

	Sn production	Rb production (low)	Rb production (high)
ZrC (3 mm diameter)	$10\% \pm 2\%$	$15\% \pm 1\%$	$2.7\% \pm 0.3\%$
3 mm diameter	$7\% \pm 2\%$	$25\% \pm 2\%$	$2.8\% \pm 0.2\%$
Ratio	1.4	0.6	1

However, the 7 mm diameter ionizer generates approximately the same increase of contamination (between 1.8 and 4.3). Then, as already explained, an increase of the diameter reduces the neutralization of ions in the cavity but it has also the consequence of an increase of alkali contamination.

The comparison between **LISBET-D3L35** and **LISBET-D7L35 + ZrC** (same inner diameter) showed an increase of the Sn production (due to the imperfect fit of the ZrC into the ionizer tube). However, a higher reduction of impurities was observed in the lower ratio (0.6) while the higher ratio resulted in the same production of Rb (1). Perhaps there is some reduction of impurities due to the presence of ZrC but the main effect is due to the ionizer diameter. Further studies will be needed to characterize the effect of the ionizer diameter over the contamination production, and to prove the impact of ZrC over the contamination reduction. An improvement of this value is expected with a new structurally modified ZrC.

Chapter 6

Conclusion and outlook

Conclusions

A Resonant Ionization Laser Ion Source (RILIS) has been developed for the production of radioactive-ion beams in SPIRAL2 Phase-2, a new ISOL facility, which is expected to be constructed at GANIL (Caen, France). Particle accelerator and mass separator facilities have an enormous interest in the RILIS technique due to its higher selectivity compared to other ionization methods to produce short lived isotopes.

The **RILIS** technique is based on the resonant photo-excitation process, where the atoms are ionized via stepwise atomic resonant excitation until ionization occurs in the last transition. The ionization process is selective due to the unique energy level structure of each element. More than 80% of the chemical elements can be accessed with the RILIS technique.

As reviewed in bibliography, most of the facilities using the RILIS hot-cavity have a “standard” ion source, which comes from surface ion source designs. As of today, few studies have been performed to optimize the geometries involved. For that reason, the study of the ion source geometry and its impact on the ion beam properties is the main objective of this work.

Another fundamental point is that surface ionized elements, which will be generated at high temperatures (~ 2000 K), will cause the contamination of the produced ion beam. It is known that low work function materials can reduce the surface ionization inside the ion source. An investigation of low work function materials working at high temperatures was performed in this work, in order to minimize the contributions from contaminant elements. To complete the study, the use of electric field potentials to reduce the contaminants has been proven.

In the first place, an **off-line test bench** has been installed at GANIL for the development of the RILIS technique. It consists of a solid-state titanium:sapphire (ti:sa) laser system, a hot cavity ion source, an extraction system, a magnet mass spectrometer, and a detection system.

The test bench has been adapted by including a new Faraday cup and two low intensity beam profilers. The extraction system has been fully optimized by means of experiments and simulations in order to provide the highest transport efficiency of the produced ion beams. A double extraction electrode has been installed after optimization.

In the view of the ion beam species requested in the SPIRAL2 letters of intent, a new three-step **Zn** ionization scheme was developed:

- First step: Transition from the $4s^2\ ^1S_0$ atomic ground state to the $4s4p\ ^1P_1^\circ$ state, which was induced by quadrupled frequency photons with a wavelength of 213.9 nm (46745.42 cm^{-1}).
- Second step: Transition from the previous state to the $3d^{10}\ 4s5d\ ^1D_2$ state with doubled frequency photons with a wavelength of 463 nm (21598.27 cm^{-1}).
- Third step: Non-resonant excitation across the ionization potential induced by photons with a wavelength of 913 nm.

Furthermore, the already known ionization schemes for **Sn** have been carefully compared in order to identify the best ionization scheme for this element. The best configuration was a three-step ionization scheme with:

- First step: Transition from the $5s^25p^2\ ^3P_0$ atomic ground state to the $5s^25p6s\ ^3P_1^\circ$ state, which was induced by tripled frequency photons of 286.4 nm.
- Second step: Transition from the previous state to the $5s^25p6p\ ^3P_2$ state, induced by photons of 811.6 nm.
- Third step: Resonant ionization to the auto-ionization $5s^25p9s\ ^3P_2^\circ$ state was induced by 823.8 nm fundamental radiation

The **ion source** geometry for radioactive-ion beam production at SPIRAL2 has been tested through the modification of different geometry parameters. The ion source consists of a transfer/atomizer tube and an ionizer tube designed in an elbow shape, which is adapted to the Target Ion Source System (TISS) configuration for SPIRAL2 Phase-2. This ion source permits to independently control the direction and magnitude of the current that is responsible for the resistive heating of the tubes up to high temperatures.

The efficiency, the emittance and the time structure were analyzed with a Sn ion beam as a function of the diameter and length of the ionizer tube. An increase of the ionizer diameter substantially ameliorated the ion beam production. The electric potential gradient along the tube, generated by the heating current, together with the reduction of neutralization due to the decrease of the contact surface, produced an amelioration of the ion extraction.

The **efficiencies** of the different ion source configurations have been compared. An increase in the ionizer length was expected to ameliorate the efficiency, however, no improvement was observed. In the case of the ionizer diameter, an efficiency of approximately 24% was obtained for 7 mm and an efficiency of 8% was obtained for 3 mm diameter ionizers. The simulations showed that an ionizer with a diameter of 7 mm should provide the best transport efficiency.

The **rms emittance** 2σ has been measured for the different LISBET ion source configurations. No significant difference has been observed in the emittance of ionizers with different lengths. In the case of 7 mm ionizer diameters, an emittance of approximately $15.5 \pi \cdot \text{mm} \cdot \text{mrad}$ has been obtained, while $10.5 \pi \cdot \text{mm} \cdot \text{mrad}$ has been obtained for 3 mm ionizer diameters. The emittance values obtained with all the LISBET geometries were small enough to ensure the transport and acceleration of ions in accelerator and mass separator facilities at GANIL without losses.

In conclusion, an enlargement of the ionizer diameter produced an increase of the isotopic production efficiency, while the emittance did not increase significantly. An ion source with 7 mm diameter and 35 mm length should provide the best behaviour for the production of short-lived isotopes due to the faster extraction from the ion source (due to the length) and the diminution of the neutralization within the hot-cavity (due to the diameter).

The **time structures** and energy scans have been measured for all the LISBET configurations and they show that the time structure consists of a narrow peak and a broad main peak that appears at certain temperature. The narrow peak is produced by the ions generated in the 19 kV extraction potential depth region inside the ionizer and the ions generated in the last millimeters at the exit of the ion source. The main peak is caused by the ions coming from the inside of the ionizer.

A proportional weight ratio was observed between both structures at different temperatures, for this reason a comparison was performed and it was observed that at high temperatures the main peak increases while the narrow peak decreases. The increasing of ions in the main peak at high temperature was explained as the result of the diminution of neutralization due to the electric potential generated by the heating current. At 1700 K, the use of 7 mm diameter ionizers provided a higher number of ions in the main peak compared to 3 mm diameter ionizers.

The fact that the main peak appears at high temperatures, raises a question about the reliability of the efficiency measurements because only the ions from the narrow peak are recorded at low temperatures. For that reason, the efficiency measurement should be considered as average result. A more exact determination of the efficiency would be the one performed directly at 2000 K. However, this sudden increase of temperature will also increase the pressure, with the consequent risk of oxidation of tantalum parts.

To fully characterize the ion beam, a technique to measure the convolution of the **energy and the spatial drift** of the ions generated by the RILIS technique has been developed. The technique provided information on the energy distribution inside and outside the ionizer. It was concluded that the narrow peak (measured in the time structure) consists of a broad distribution of energies, which can complicate the isotope selection. The ions coming from inside the ionizer have an estimated energy dispersion of about 5 eV.

Additionally, in order to study and optimize the selectivity of the process, two methods have been studied in order to decrease the production of contaminants (due to the surface ionization) with an alkali marker (Rb). The first technique was based on the control of the **electric field potential** direction. By this method, a decrease of a factor of 2 was reached by reversing the electric field potential direction in the atomizer. The second technique implied the insertion of **ZrC** ceramics in the ionizer tube. Further reduction of the contamination at high temperatures was expected in this case. As described in bibliography, ZrC materials could have a low work function property that diminishes the alkali contamination. However, no significant improvement was found with the material configuration studied. Preliminary work on the microstructure of ZrC has to be carried out in order to reduce the work function before new investigations.

From the point of view of the contamination generation, it has been observed using the LISBET geometries that an enlargement of the ionizer diameter, improved the production of laser ionizer elements, at the same time the production of contaminants was also increased due to the surface ionization. A comparative study has to be performed connecting the RILIS ion production and contaminants as a function of the geometry and the temperature.

As conclusion, the test bench has been optimized with the optimal extraction and transport parameters and the main objectives of this thesis were accomplished. At the same time, the ionization schemes of zinc and tin have been chosen that provide the best operation for a ti:sa laser system. Different geometries of the ion source have been tested, where an enlargement of the ionizer diameter increased the isotope production efficiency, with small variations of emittance. The geometry with 7 mm diameter and 35 mm length ionizer (LISBET-D7L35) has proven to offer the best performance. Finally a reduction of the contamination production was observed using the electric field potential.

Outlook

The GISELE off-line test bench has been successfully commissioned and optimized for the RILIS technique. The achievement of the objectives open new lines of work of great interest in order to develop the technique.

GISELE is now fully operational for spectroscopic and efficiency measurements, for this reason a study has to be carried out to find efficient ionization schemes using an all solid-state ti:sa laser for zirconium (Zr), erbium (Er) or chromium (Cr) which will be used in SPIRAL2- S^3 . In addition, the research of ionization schemes with a possible final AIS for the rest of the elements could provide an enhancement of the production efficiency.

The efficiency improvement obtained due to the increase of the ionizer diameter has to be confirmed with other RILIS ionized elements (e.g. gallium or zinc), considering also the dependence in the efficiency of the resonance and/or ionization transitions saturation. Another possible optimization is the decrease of the thickness of the ionizer to provide a higher electric field potential (limited by the material strength). An increase of the electric field in the transfer/atomizer tube will enhance the alkali suppression without affecting the laser generated ions. It should also be taken into consideration that an increase of the electric potential in the ionizer will increase the energy spread of the ion beam. This energy dispersion will not affect the low energy experiments due to the use of a cooler system before the setup [55], but it could affect the charge booster in an energy window between 3 to 7 eV [213].

The narrow peak in the time structure of the ion beam is generated due to the extraction potential depth into the ionizer. A possibility to reduce this effect is to place a metallic grid to homogenize the electric potential distribution at the exit of the ionizer. However, this also can reduce the amount of laser light that enters the cavity with the consequent diminution of the laser-atom interaction, in addition this technique would not improve the energy dispersion of the ions generated outside the ion source in the extraction gap. A study of the optimum grid ensuring homogeneity of the extraction potential has to be carried out in order to increase the number of ions with the correct energy.

Addressing the issue of the characterization of the low work function materials can be performed by measuring the atom flux with a quartz microbalance and the ion current signal with a Faraday cup, as explained in appendix B. Rubidium can be the perfect candidate to measure the work function of the selected material. It is important to note that a modification of the ion source chamber has to be carried out to allow the measurement of both parameters at the same time.

An additional development of the RILIS technique would consist in the improvement of the laser alignment control. By the methods explained in this thesis, the alignment at the entrance of the ionizer was assured. However, small angles can produce a variation in the illumination of the deeper part of the ionizer tube, which would reduce the laser-atom interaction. Once the ion source is removed for maintenance, it will be possible to place a double iris to correct the laser alignment.

An important percentage of the ion beams requested in the SPIRAL2 letters of intent could be produced through the RILIS technique. The works presented in this thesis provided explanations to some physical behaviours related to the RILIS technique and they open new perspectives of research and development. It is fundamental to continue these works since RILIS is going to be used in the SPIRAL2-S³ installations (gas cell) and SPIRAL2-Phase2 (hot cavity). The technique could also be considered in the framework of SPIRAL1 (hot cavity) in the near future.

Conclusion et perspectives

Conclusions

Une source d'ions par ionisation résonante laser (RILIS en acronyme anglais) a été développée pour la production de faisceaux d'ions radioactifs dans le cadre de SPIRAL2 Phase-2, une nouvelle installation ISOL, qui pourrait être construite au GANIL (Caen, France). Les accélérateurs de particules et les séparateurs de masse montrent un énorme intérêt à la technique RILIS en raison de sa grande sélectivité en comparaison à d'autres méthodes d'ionisation pour la production d'isotopes de courte durée de vie.

La technique **RILIS** est basée sur le processus de photo-excitation résonante, où les atomes sont ionisés par étapes successives d'excitation résonante atomique jusqu'à ce que l'ionisation se produise dans la dernière transition. En raison de la structure unique de niveaux d'énergie de chaque élément, le procédé d'ionisation est sélectif. Plus de 80% des éléments chimiques peuvent être produits par la technique RILIS.

Une recherche bibliographique montre que la plupart des installations utilisent une RILIS à cavité chaude basée sur une source d'ions "standard" à thermo-ionisation (ionisation par contact avec les surfaces chaudes). A ce jour, peu d'études ont été réalisées pour optimiser cette géométrie. L'objectif principal de ce travail est donc l'étude de la géométrie de la source d'ions et son impact sur les propriétés du faisceau d'ions.

En outre, les éléments à faible potentiel d'ionisation, facilement ionisés par simple contact avec les surfaces chaudes de la cavité (~ 2000 K), provoquent la contamination du faisceau d'ions. Les matériaux à faible travail de sortie peuvent réduire la thermo-ionisation à l'intérieur de la source d'ions. Afin de minimiser les contributions des éléments contaminants, une recherche de matériaux à faible travail de sortie qui peuvent être portés à haute température et l'utilisation des potentiels électriques ont été étudiés dans ce travail.

Un **banc de test** a été installé au GANIL pour le développement de la technique RILIS. Il se compose d'un système laser titane:saphir (ti:sa), d'une source d'ions à cavité chaude, d'un système d'extraction, d'un spectromètre magnétique de masse, et d'un système de détection.

Le banc de test a été modifié en incluant une nouvelle coupelle de Faraday et deux profileurs de basse intensité. En outre, le système d'extraction a été entièrement optimisé grâce à des expériences et des simulations, afin de trouver la meilleure efficacité de transport des faisceaux d'ions produits. Une double électrode d'extraction a été installée après optimisation.

Pour accéder aux faisceaux d'ions demandées dans les lettres d'intention de SPIRAL2, un nouveau schéma d'ionisation à trois étapes pour le **Zn** a été développé:

- Première étape: Transition de l'état fondamental atomique $4s^2\ ^1S_0$ à l'état $4s4p\ ^1P_1^\circ$, induite par des photons de fréquence quadruplé avec une longueur d'onde de 213,9 nm ($46745,42\text{ cm}^{-1}$).
- Deuxième étape: La transition de l'état précédent à l'état $3d^{10}\ 4s5d\ ^1D_2$ avec des photons de fréquence doublée avec une longueur d'onde de 463 nm ($21.598,27\text{ cm}^{-1}$).
- Troisième étape: Excitation non résonante au delà du potentiel d'ionisation, induite par des photons d'une longueur d'onde de 913 nm.

De plus, les systèmes d'ionisation déjà connus pour le **Sn** ont été soigneusement comparés afin d'identifier le meilleur schéma d'ionisation pour cet élément. La meilleure configuration trouvée est un schéma d'ionisation en trois étapes avec:

- Première étape: Transition de l'état atomique fondamental $5s^25p^2\ ^3P_0$ à l'état $5s^25p6s\ ^3P_1^\circ$, induite par des photons de fréquence triplée de 286,4 nm.
- Deuxième étape: La transition de l'état précédent à l'état $5s^25p6p\ ^3P_2$, induite par des photons de 811,6 nm.
- Troisième étape: Transition de l'état précédent à l'état auto-ionisant $5s^25p9s\ ^3P_2^\circ$ induite par le rayonnement fondamental 823,8 nm, produisant l'ionisation résonante.

La géométrie de la **source d'ions** pour la production de faisceaux d'ions radioactifs à SPIRAL2 a été testée en modifiant les différents paramètres géométriques. La source d'ions est constituée d'un tube transfert/atomiseur et d'un tube d'ionisation conçu en forme de coude, qui est adapté à la configuration de l'ensemble cible-source (TISS en acronyme anglais) pour SPIRAL2 Phase-2. Cette source d'ions permet de contrôler de façon indépendante la direction et la valeur du courant qui est induit le chauffage des tubes par effet Joule jusqu'à des températures élevées.

L'efficacité, l'emittance et la structure en temps des faisceaux d'ions de Sn ont été analysées en fonction du diamètre et de la longueur du tube d'ionisation. Une augmentation du diamètre de l'ioniseur améliore sensiblement la production d'ions. Le gradient de potentiel électrique dans le tube généré par le courant de chauffage, combiné avec la réduction de la neutralisation due à la diminution de la surface de contact, a produit une amélioration de l'extraction des ions.

Les **efficacités** des différentes configurations de sources d'ions ont été comparées. Une augmentation de la longueur était prévu pour améliorer l'efficacité. Cependant, aucune amélioration n'a pas été observée en modifiant la longueur. Une efficacité d'environ 24% a été obtenue avec les ioniseurs de 7 mm de diamètre, et une efficacité de 8% a été obtenue avec les ioniseurs de 3 mm de diamètre. Les simulations ont montré qu'un ioniseur de 7 mm de diamètre, doit assurer la meilleure efficacité de transport.

L'**emittance rms** 2σ a été mesurée pour les différentes configurations de source d'ions. Aucune différence significative n'a pas été observée dans l'emittance des ioniseurs avec des longueurs différentes. Dans les cas des ioniseurs avec 7 mm de diamètre, une emittance d'environ $15,5 \pi \cdot \text{mm} \cdot \text{mrad}$ a été obtenue, tandis que pour ceux de 3 mm de diamètre, une emittance de $10,5 \pi \cdot \text{mm} \cdot \text{mrad}$ a été obtenu. Les valeurs d'emittance obtenues avec toutes les géométries étaient suffisamment petites pour assurer le transport et l'accélération des ions dans les accélérateurs et les installations de séparation de masse du GANIL sans pertes.

En conclusion, un agrandissement du diamètre de l'ioniseur a produit une augmentation de l'efficacité de la production isotopique, tandis que l'emittance n'a pas augmenté de manière significative. Une source d'ions avec 7 mm de diamètre et 35 mm de longueur devrait fournir le meilleur comportement pour la production d'isotopes à courte durée vie en raison de l'extraction plus rapide de la source d'ions (dû à la longueur) et la diminution de la neutralisation à l'intérieur de la cavité chaude (dû à le diamètre).

Les **structures temporelles** et les distributions en énergie ont été mesurées pour toutes les configurations. La structure temporelle consiste en un pic étroit et un pic large principal qui apparaît a une certaine température. Le pic étroit est produit par les ions générés dans la région du potentiel d'extraction (19 kV) à l'intérieur de l'ioniseur et dans les derniers millimètres à la sortie de la source d'ions. Le pic principal est dû aux ions provenant de l'intérieur de l'ioniseur.

Le rapport entre les deux structures varie avec la température. Pour cette raison, les structures temporelles ont été comparées, et il a été possible d'observer que à haute température le pic principal augmente et le pic étroit diminue. Cette augmentation a été expliqué comme le résultat de la diminution de la neutralisation dû à le potentiel électrique généré par le courant de chauffage à haute température. À 1700 K, l'utilisation d'ioniseurs de 7 mm de diamètre a fourni un nombre plus élevé d'ions dans le pic principal à 1700 K par rapport aux ioniseurs de 3 mm de diamètre.

Le fait que le pic principal apparaisse à 1700 K, soulève une question au sujet de la fiabilité des mesures d'efficacité car seuls les ions du pic étroit sont enregistrés à basse température. Pour cette raison, les mesures d'efficacité doivent être considérées comme des résultats moyens. Une détermination plus exacte de l'efficacité serait celle effectuée uniquement à 2000 K. Cependant, cette augmentation soudaine de la température pourrait augmenter également la pression, avec le risque d'oxydation des pièces en tantale.

Pour finaliser la caractérisation du faisceau d'ions, une technique pour mesurer la convolution de l'**énergie et de la déviation spatiale** des ions générés par la technique RILIS a été développée. La technique a fourni des informations sur la distribution de l'énergie des ions à l'intérieur et à l'extérieur de l'ioniseur. Il a été conclu que le pic étroit (mesuré dans la structure temporelle) est constituée d'une large distribution en énergies, ce qui peut compliquer la sélection des isotopes. Les ions provenant de l'intérieur de l'ioniseur ont une dispersion en énergie estimée à environ 5 eV.

Enfin, deux méthodes ont été étudiées afin de diminuer la production des polluants avec un marqueur alcalin (Rb). La première est basée sur le contrôle de la direction du **potentiel électrique**. Par ce procédé, une diminution d'un facteur 2 a été atteinte en inversant le sens du potentiel électrique dans l'atomiseur. La seconde technique implique l'insertion de **ZrC** dans le tube ioniseur. Une réduction supplémentaire de la contamination à des températures élevées était prévue dans ce cas. Comme décrit dans la littérature, les matériaux ZrC devraient avoir un travail de sortie plus bas ce qui pourrait diminuer les polluants alcalins. Cependant, aucune amélioration significative n'a pas été observée avec la configuration étudiée. Des travaux sur la microstructure de ZrC doivent être effectuées afin de réduire le travail de sortie de ce matériau avant de nouvelles recherches.

Il a été observé que l'augmentation du diamètre de l'ioniseur améliore la production d'éléments ionisés par laser en même temps qu'il augmente la production des contaminants provenant de la thermo-ionisation par contact. À partir de cette conclusion, une étude comparative doit être réalisée mettant en relation la production d'ions RILIS et les polluants en fonction de la géométrie et de la température.

En conclusion, le banc de test a été optimisé avec les paramètres optimaux d'extraction et de transport et les principaux objectifs de cette thèse ont été atteints. Par ailleurs, les schémas d'ionisation du zinc et de l'étain ont été sélectionnés pour fournir le meilleur fonctionnement du système laser titane:saphire. Différentes géométries de la source d'ions ont été testées, et un diamètre plus grand augmente l'efficacité de production des isotopes, sans augmenter l'émittance. La géométrie avec l'ioniseur de 7 mm de diamètre et 35 mm de longueur (LISBET-D7L35) a fourni le meilleur comportement. Finalement, la réduction des contaminants par la méthode du potentiel électrique a été observée.

Perspectives

Le banc de test GISELE a été mis en service et a été optimisé avec succès pour la technique RILIS. La réalisation des objectifs ouvre de nouvelles voies de travail d'un grand intérêt pour le développement de la technique.

Actuellement GISELE est pleinement opérationnelle pour des mesures spectroscopiques et d'efficacité, pour ce raison une étude doit être réalisée afin de trouver les schémas d'ionisation plus efficaces en utilisant un système laser Ti:S pour l'zirconium (Zr), erbium (Er) ou chromium (Cr) qui seront utilisés dans SPIRAL2- S^3 . De plus, une étude des possibles états auto-ionisants doit être réalisée sur le reste d'éléments pour augmenter les efficacités de production.

Ensuite, l'amélioration de l'efficacité obtenue grâce à l'augmentation du diamètre de l'ioniseur doit être confirmée pour d'autres éléments (par exemple Ga ou Zn), en considérant également la dépendance de l'efficacité avec la saturation de la résonance et/ou des transitions d'ionisation. Une autre possibilité d'optimisation est la diminution de l'épaisseur de l'ioniseur pour fournir un plus grand potentiel électrique. Ceci est limité par la tenue mécanique du matériau. Une augmentation du potentiel électrique dans le tube transfert/atomiseur permettra d'améliorer la suppression des alcalins sans affecter les ions générés par laser. Il faut tenir compte qu'une augmentation du potentiel électrique dans l'ioniseur augmentera également la dispersion en énergie du faisceau d'ions. Cette dispersion en énergie n'affectera pas les expériences de basse énergie en raison de l'utilisation d'un système cooler avant l'expérience [55], mais cela pourrait affecter le booster de charge dans une fenêtre d'énergie entre 3 à 7 eV [213].

Le pic étroit dans la structure temporelle du faisceau d'ions est généré en raison du potentiel d'extraction dans l'ioniseur. Une possibilité pour réduire cet effet consiste à placer une grille métallique pour homogénéiser la distribution du potentiel électrique à la sortie de l'ioniseur. Cependant, cela peut également réduire la quantité de lumière laser qui pénètre dans la cavité et aussi la diminution de l'interaction laser-atome. De plus, cette technique ne permet pas d'éviter la dispersion en énergie des ions générés en dehors de la source d'ions dans l'extraction. Une étude de la grille en assurant une homogénéité optimale du potentiel d'extraction doit être effectuée afin d'augmenter le nombre d'ions avec l'énergie correcte.

Si on aborde la caractérisation des matériaux de faible travail de sortie, elle peut être effectuée en mesurant le flux d'atomes avec une microbalance à quartz et le signal de courant ionique avec une coupelle de Faraday, comme expliqué dans l'annexe B. Le rubidium peut être un candidat idéal pour mesurer le travail de sortie de la matière choisie. Toutefois, une modification de la chambre de la source d'ions doit être effectuée pour permettre la mesure de ces deux paramètres à la fois.

Un développement supplémentaire de la technique RILIS consisterait à améliorer le contrôle de l'alignement du laser. L'alignement à l'entrée de l'ioniseur était assuré par les méthodes expliquées dans cette thèse. Toutefois, les petits angles peuvent produire une variation de l'éclairage de la partie plus profonde du tube ioniseur, ce qui réduirait l'interaction laser-atome. Une fois que la source d'ions est retirée pour maintenance, il est possible de placer un double diaphragme pour corriger l'alignement du laser.

Un pourcentage important des faisceaux d'ions demandés dans les lettres d'intention de SPIRAL2 pourrait être produit par la technique RILIS. Les travaux présentés dans cette thèse ont fourni des explications à certains aspects physiques liés à la technique RILIS et ouvrent de nouvelles perspectives en terme de recherche et développement. Il est fondamental de continuer ces travaux puisque RILIS va être utilisé au sein des installations SPIRAL2-S³ (cellule gazeuse) et SPIRAL2-Phase2 (cavité chaude). Il pourrait aussi être considéré dans le cadre de SPIRAL1 (cavité chaude) dans le futur proche.

Conclusión y perspectivas

Conclusiones

Se ha desarrollado una fuente de iones por ionización resonante láser (RILIS en el acrónimo inglés) para la producción de haces de iones radioactivos en SPIRAL2 Phase-2, una nueva instalación ISOL que será construida en GANIL (Caen, Francia). Esta técnica tiene un enorme interés dentro de los aceleradores de partículas y los separadores de masas, debido a su mayor selectividad en comparación con otros métodos de ionización para la producción de isótopos de corta duración.

La técnica **RILIS** se basa en el proceso de foto-excitación resonante, donde los átomos son ionizados a través de sucesivas etapas de excitación resonante atómica hasta la ionización producida en la última transición. El proceso de ionización es selectivo debido a la estructura de niveles energéticos única para cada elemento. Un factor clave es que más del 80% de los elementos químicos podrían ser producidos con esta técnica.

Como se puede observar en la bibliografía, la mayor parte de las instalaciones que utilizan una cavidad caliente RILIS, se sirven de una fuente de iones “estándar” procedente de los diseños de las fuentes por ionización térmica. Hasta hoy se han realizado pocos estudios destinados a optimizar las geometrías implicadas. Por ello, el principal objetivo de este trabajo es, precisamente, el estudio de la geometría de la fuente de iones y su impacto en las propiedades de los haces de iones generados.

Otro punto importante es que los elementos ionizados térmicamente a altas temperaturas (~ 2000 K) contaminan el haz de iones producido. Es sabido que los materiales que presentan una baja función de trabajo pueden reducir la ionización térmica en el interior de la fuente de iones. Se ha realizado una investigación empleando materiales de este tipo con el fin de minimizar las contribuciones de los elementos contaminantes que permitan, además, el funcionamiento a altas temperaturas. Para terminar, se ha probado el uso del potencial del campo eléctrico con la misma finalidad de reducir los contaminantes.

El primer paso ha sido la instalación de un **banco de pruebas** en GANIL para el desarrollo de la técnica RILIS, compuesto por: Un sistema láser titanio:zafiro (ti:sa), una fuente de iones con cavidad caliente, un sistema de extracción, un espectrómetro de masas magnético, y un sistema de detección.

El banco de pruebas se ha adaptado mediante la inserción de un nuevo detector tipo copa de Faraday y dos perfiladores de haz de baja intensidad. El sistema de extracción se ha optimizado por medio de experimentos y simulaciones que proporcionan la eficiencia más alta en el transporte de los haces de iones producidos. Tras dicha optimización se ha instalado un doble electrodo de extracción.

Se ha desarrollado un nuevo esquema de ionización para el **Zn** debido a los haces de iones solicitados para SPIRAL2:

- Primer paso: La transición del estado atómico fundamental $4s^2\ ^1S_0$ al estado $4s4p\ ^1P_1^\circ$, inducida mediante fotones de frecuencia cuádruple con una longitud de onda de 213,9 nm ($46.745,42\text{ cm}^{-1}$).
- Segundo paso: La transición desde el estado anterior al estado $3d^{10}\ 4s5d\ ^1D_2$ con fotones de doble frecuencia con una longitud de onda de 463 nm ($21598,27\text{ cm}^{-1}$).
- Tercer paso: Excitación no resonante, a través del potencial de ionización, inducida mediante fotones con una longitud de onda de 913 nm.

Por otra parte, se han estudiado cuidadosamente los esquemas de ionización conocidos del **Sn**. Se ha comprobado que la mejor configuración es el siguiente esquema de ionización:

- Primer paso: Transición del estado fundamental atómico $5s^25p^2\ ^3P_0$ al estado $5s^25p6s\ ^3P_1^\circ$, inducida mediante fotones de frecuencia triplicada de 286,4 nm.
- Segundo paso: La transición desde el estado anterior al estado $5s^25p6p\ ^3P_2$, inducida mediante fotones de 811,6 nm.
- Tercer paso: Ionización de resonancia al estado auto-ionizante $5s^25p9s\ ^3P_2^\circ$, inducido mediante radiación fundamental 823,8 nm

Se ha ensayado la geometría de la **fuentes de iones** para la producción de haces de iones radioactivos en SPIRAL2, a través de la modificación de los diferentes parámetros geométricos. La fuente de iones, adaptada a la configuración del conjunto blanco-fuente (TISS en el acrónimo inglés) para SPIRAL2 Phase-2, se compone de un tubo de transferencia/atomizador y un tubo ionizador diseñado en forma de codo. Esta fuente de iones permite controlar de forma independiente la dirección y la magnitud de la corriente eléctrica, que es responsable del calentamiento resistivo de los tubos.

La eficiencia, la emitancia y la estructura temporal de un haz de iones de Sn se han estudiado en función del diámetro y la longitud del tubo ionizador. Un aumento del diámetro del ionizador mejora sustancialmente la producción de haces de iones. El gradiente de potencial eléctrico a lo largo del tubo, generado por el calentamiento resistivo, junto con la reducción de la neutralización debido a la disminución de la superficie de contacto, produce una mejora de la extracción de iones.

Al analizar la **eficiencia** de las diferentes configuraciones de la fuente de iones, se ha observado que un aumento de la longitud no proporciona una mejora sustancial de los resultados, al contrario de lo que se esperaba. En cuanto al diámetro, se han obtenido eficiencias aproximadas del 24% para el caso de 7 mm, mientras que los ionizadores con diámetro 3 mm no superan el 8%. Las simulaciones confirman estos resultados ya que muestran que un ionizador con un diámetro de 7 mm proporciona la mejor eficiencia del transporte.

Al medir la **emitancia rms** 2σ para las diferentes configuraciones de la fuente de iones LISBET, ninguna diferencia significativa se ha observado al variar la longitud. En el caso de diámetros de 7 mm se han obtenido emitancias de aproximadamente $15,5 \pi \cdot \text{mm} \cdot \text{mrad}$, mientras que diámetros de 3 mm proporcionaron $10,5 \pi \cdot \text{mm} \cdot \text{mrad}$. Estos valores de emitancia obtenidos en las diferentes geometrías LISBET son lo suficientemente pequeños como para asegurar el transporte y la aceleración de los iones en las instalaciones de aceleración y separación de masa de GANIL sin pérdidas.

Visto que ni la eficiencia, ni la emitancia varían significativamente con la longitud del ionizador, una fuente de iones con 7 mm de diámetro y 35 mm de longitud debería proporcionar el mejor comportamiento para la producción de isótopos de corta duración debido a la extracción más rápida de la fuente de iones (debido a la longitud) y una disminución de la neutralización (debido al diámetro).

Los resultados obtenidos en las **estructuras temporales** y los escáneres de energía muestran que en todas las configuraciones la estructura temporal está formada por un pico estrecho y un amplio pico principal que aparece a partir de una cierta temperatura. El pico estrecho se produce por los iones generados en la región del potencial de extracción (19 kV) en el interior del ionizador y por los iones generados en los últimos milímetros de la salida de la fuente de iones. El pico principal se forma por los iones procedentes de la parte interior del ionizador.

La relación proporcional entre ambos picos varía con la temperatura, por ello se ha realizado una comparativa de estructuras temporales en función de la misma, donde se observa que a mayores temperaturas el pico principal aumenta mientras que el pico estrecho disminuye. Esto se explica como resultado de la disminución de la neutralización debido al potencial eléctrico generado por la corriente de calentamiento resistivo. Estableciendo una temperatura de 1700 K, se ha observado que el uso de ionizadores de 7 mm de diámetro ha proporcionado un mayor número de iones en el pico principal, comparado con los ionizadores de 3 mm de diámetro.

La aparición del pico principal a altas temperaturas plantea la pregunta sobre la fiabilidad de las medidas de eficiencia, ya que a bajas temperaturas sólo se registran los iones procedentes del pico estrecho. Por esta razón, las medidas de eficiencia deben considerarse como un resultado promedio. Una determinación más exacta de la eficiencia debería ser llevada a cabo a 2000 K, sin olvidar que este aumento repentino de la temperatura aumenta la presión, con el consiguiente riesgo de oxidación de las partes de tantalio.

Para finalizar la caracterización, se ha desarrollado una técnica para medir la convolución de la distribución **energética y la divergencia espacial** de los iones generados por la técnica de RILIS, que ha proporcionado información sobre la distribución de energía dentro y fuera del ionizador. El resultado muestra que el pico estrecho (de la medida en la estructura temporal) se compone de una amplia distribución de energías, que puede complicar la selección de isótopos. Los iones procedentes del interior del ionizador poseen una dispersión de energía estimada en, aproximadamente, 5 eV.

Complementariamente, con el fin de estudiar y mejorar la selectividad del proceso, se han ensayado dos métodos para disminuir la producción de contaminantes (causados por la ionización térmica) con un marcador alcalino (Rb). El primero se basa en el control de la dirección del **potencial de campo eléctrico**. Mediante este método, una disminución de un factor 2 ha sido alcanzado gracias a la inversión de la dirección del campo eléctrico en el atomizador. El segundo consiste en la inserción de **ZrC** en el tubo ionizador. Como se ve en la bibliografía, materiales como el ZrC tienen un valor bajo de la función de trabajo lo que podría reducir la contaminación alcalina. Sin embargo, ninguna mejora significativa se ha encontrado. Un estudio de la microestructura del ZrC debería realizarse con el fin de analizar y reducir la función de trabajo antes de realizar nuevas investigaciones.

Desde la perspectiva de la generación de contaminantes, se ha observado que el aumento del diámetro del ionizador, bien que mejora la producción de elementos ionizados mediante láser, también incrementa la producción de contaminantes debidos a la ionización térmica. A partir de esta conclusión, se debería realizar un estudio comparativo donde se relacionen la producción de iones RILIS y contaminantes en función de la geometría y la temperatura.

Para concluir, el banco de pruebas ha sido optimizado con los parámetros óptimos de extracción y transporte, cumpliendo el objetivo principal establecido para esta tesis. A su vez, se han seleccionado los esquemas de ionización de zinc y estaño que proporcionan el mejor funcionamiento para el sistema láser titanio:zafiro. Se han ensayado diferentes geometrías de la fuente de iones, observando que un mayor diámetro del ionizador proporciona un aumento de la eficiencia de producción de los isótopos, sin apenas aumentar la emitancia. La geometría de 7 mm de diámetro y 35 mm de longitud (LISBET-D7L35) ofrece el mejor comportamiento. Finalmente, se ha logrado reducir la generación de contaminantes alcalinos con el método del potencial del campo eléctrico.

Perspectivas

El banco de pruebas GISELE ha sido optimizado y puesto a punto para el desarrollo de la técnica RILIS. La ejecución de los objetivos marcados abre nuevas líneas de trabajo de gran interés de cara al desarrollo de la técnica.

En la actualidad GISELE está en pleno funcionamiento para realizar medidas de espectroscopía y de eficiencia, por ello debería llevarse a cabo un estudio con el fin de encontrar esquemas de ionización más eficaces utilizando el sistema láser de titanio:zafiro para zirconio (Zr), erbio (Er) o cromo (Cr) que serán utilizados en SPIRAL2- S^3 . Además un estudio de posibles estados auto-ionizantes en el resto de elementos debería realizarse con el fin de aumentar las eficiencias de producción.

En segundo lugar, la mejora de la eficiencia obtenida por el aumento del diámetro del ionizador tiene que confirmarse para otros elementos generados mediante RILIS (por ejemplo galio o zinc), considerando también que la eficiencia depende de la saturación de las resonancias y/o transiciones de ionización. Otro posible aspecto de optimización es la disminución del espesor del ionizador, proporcionando un mayor potencial de campo eléctrico (limitado por la resistencia del material). Dicho aumento del campo eléctrico en el tubo de transferencia/atomizador mejoraría la supresión alcalina sin afectar a los iones generados mediante láser. Hay que tener en cuenta que un aumento del potencial eléctrico en el ionizador aumentaría la divergencia energética del haz de iones. Esta dispersión de energía no afecta a los experimentos de baja energía debido a la utilización de un sistema de enfriamiento antes del experimento [55], pero podría afectar al booster de carga en una ventana de energía entre 3-7 eV [213].

El pico estrecho en la estructura temporal del haz de iones se genera debido al potencial de extracción dentro del ionizador. Este efecto podría reducirse mediante una rejilla metálica para homogeneizar la distribución de potencial eléctrico en la salida del ionizador. Sin embargo, esta rejilla también puede reducir la cantidad de luz láser que entra en la cavidad con la consiguiente disminución de la interacción láser-átomo, además esta técnica no mejoraría la dispersión de energía de los iones generados fuera de la fuente de iones en la zona de extracción. Un estudio de la red que asegure la homogeneidad del potencial de extracción tiene que llevarse a cabo con el fin de aumentar el número de iones con la energía correcta.

Si se aborda la caracterización de los materiales de baja función de trabajo, es recomendable realizarla mediante la medición del flujo atómico con una microbalanza de cuarzo y la señal de corriente de iones con una copa de Faraday, como se explica en el apéndice B. Rubidio podría ser el candidato perfecto para medir la función de trabajo del material seleccionado. Hay que tener en cuenta que la cámara de experimentación de la fuente de iones tiene que ser modificada para permitir la medición de los dos parámetros al mismo tiempo.

Un desarrollo adicional de la técnica RILIS consistiría en la mejora del control de alineamiento láser. Por los métodos explicados en esta tesis, se ha asegurado el alineamiento en la entrada del ionizador. Sin embargo, pequeños ángulos pueden producir una variación en la iluminación de la parte más profunda del tubo ionizador, lo que reduciría la interacción láser-átomo. Una vez retirada la fuente de iones para el mantenimiento, sería posible colocar un doble diafragma para corregir la alineación láser.

Un porcentaje importante de los haces de iones solicitados para SPIRAL2 podrían ser producidos a través de la técnica de RILIS. Los trabajos presentados en ésta tesis proporcionan explicaciones a ciertos aspectos físicos relacionados con la técnica RILIS y abren nuevas perspectivas de investigación y desarrollo. Es fundamental continuar con éstos trabajos ya que la técnica RILIS va a ser utilizada en las instalaciones SPIRAL2-S³ (célula gaseosa) y SPIRAL2-Phase2 (cavidad caliente). Ésta técnica también podría ser considerada en el marco de SPIRAL1 (cavidad caliente) en un futuro próximo.

Appendix A

Laser fundamentals

A laser is a device that emits light through a process of optical amplification based on the stimulated emission of electromagnetic radiation [79]. The word laser is an acronym for *Light Amplification by stimulated Emission of Radiation*. The key of the lasing concept is the stimulated emission process (fig. A.1) where a photon interacting with an initially excited atom can cause this atom to go to the fundamental state emitting a new photon added to the one that incited the transition.

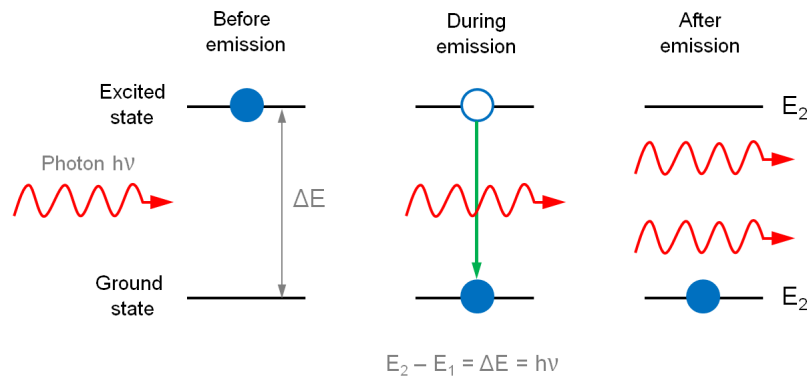


Figure A.1– Diagram of the stimulated emission process. Before the emission the atom is in an excited state. When a photon arrives, the decay of the atom occurs emitting a photon with identical properties as the one who produced the emission.

Originally the idea comes from a work of Albert Einstein in 1917 about quantum theory of radiation [214]. In this article, he suggested a system in thermal equilibrium consisting of a cavity filled by light and a number equal of atoms in their fundamental state and excited state, which can emit and absorb radiation. The equilibrium is reached when the absorption of a photon is as fast as the emission. The absorption rate is proportional to the number of photons. This proportional constant was called coefficient B. In equilibrium, the emission rate

has to be the same as the absorption rate. For that, the emission has to be described by the same coefficient B and it has to be proportional to the number of photons too. By this method, Einstein proposed a new type of emission that can only exist in the presence of an electric field. This means the higher the quantity of present photons, the greater the efficiency of the process will be. The stimulated emission generates the decay of an atom adding a new photon to the radiation field. This may create an avalanche process; the increment of photons stimulates the emission of more photons in the medium. This is the principle of the light amplification by stimulated emission [215].

To generate the stimulated emission a gain medium is needed. As we have seen, the atoms of the material have to be excited. Atoms in the fundamental state absorb the radiation while the excited atoms can amplify the light by the stimulated emission. Both processes are linked and the global balance absorption/amplification depends on the relative fraction between the quantities of atoms in the fundamental state and excited state. To provide a full amplification, the latter has to lead to the first one. This is called population inversion. Most known materials can absorb light but none of them amplifies the light in standard conditions.

The mechanism to induce the population inversion is not essentially complicated. It is enough to provide energy to the active medium. However the population inversion is not viable in time. When the atoms emit radiation, the fraction of excited atoms decreases and the population inversion disappears. Then an initial amplifier material will turn into absorbent material. To maintain the amplification process, an external agent that excites the atoms and preserves the population inversion is needed. The mechanism responsible for maintaining the population inversion is called *pump*. There are a wide range of pump techniques which depend on the gain medium to generate amplification. The common request is to be efficient enough to compensate the atomic decay and maintain the population inversion [216].

These two elements (the active medium and the population inversion mechanism) are not sufficient to produce laser light. In order to increase the photon density passing through the active medium, it is confined in a cavity formed by mirrors (fig. A.2). One of the mirrors has high reflectivity and the other is partially reflecting to let the laser light escape from the cavity. By this method the brightness will be increased, the control of the wavelength will be possible, and the spectral bandwidth generated by the gain curve of the material will be reduced. The frequencies f of the resonant radiation in the cavity have to fulfill the relation $f = N \cdot c / (2L)$, where N is an integer number, c is the speed of light [m/s] and L is the length of the cavity [m]. These frequencies are called longitudinal modes of the cavity. In addition, multiple sweeps have the benefit of an increment of the amplification efficiency back feeding multiple times. On the other hand, just the photons which are aligned with the cavity axis will be amplified.

The beam of light generated by a laser can have many properties that are unique. In a general description of the emitted beam light, it has the properties to be monochromatic,

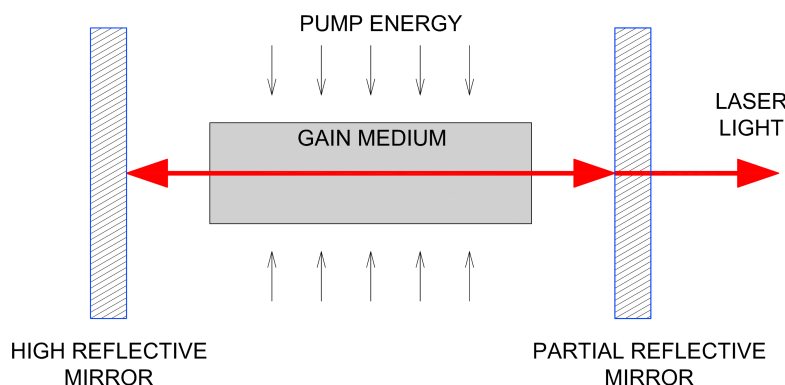


Figure A.2– Schematic explaining the operation of a laser system. The gain material confined by two mirrors (one of them is partially reflecting). A pump element is needed to sustain the emission process.

coherent, and have directional propagation. The **monochromatic light** contains just one wavelength. Of course, it is not possible to reach a perfect monochromatic field. However the laser emits in a narrow frequency interval that can be considered monochromatic. An electromagnetic wave is **coherent** when its phase does not change randomly in space or time when propagating. Coherence is a consequence of the stimulated emission where each photon is emitted identical to the previous. The **directional propagation** is a direct consequence of the cavity configuration, because only the photons that travel in the perpendicular direction of the mirrors will contribute to the amplification.

The discovery of the laser is a combination of the effort and contributions of numerous scientists during the whole XXth century. The idea of the laser appears in the combination of basic concepts, such as the stimulated emission and techniques, as the optic pump or the maser (a microwave laser) [217]. T. H. Maiman et al. succeeded to amplify the visible light by stimulated emission for the first time in the year 1960 [218]. But due to the number of previous patents and authorships (including Nobel prizes to Charles H. Townes, Nikolay Basov and Alexander Prokhorov, for their research to lead to the maser), it is impossible to describe the discovery of the laser focused on only one persons research. Just a year later of its demonstration, the first research applications were published. During the 60's there was a vertiginous development and lasers were increasingly used for scientific research and industrial technology. Nowadays the two main scientific domains of frontier research are: the shortest laser pulse and highest peak-power laser pulse [219]. The impact of this new apparatus was enormous, a few years after its first development, it was implemented in many fields and presented a whole range of possibilities for the industry and research.

Appendix B

Work function measurement

Introduction

The objective is to measure the work function of a material (φ) by means of the surface ionization process. For a correct measurement, it is necessary to consider that the surface ionization is the main ionization process. In this case, a hot-cavity ion source fulfill this requirement. A microbalance set at the exit of the ion source and a Faraday cup placed after the magnet mass separator will be needed in order to perform the experiment. An easily ionized atom has to be chosen to perform the experiments. Rb is a perfect candidate due to its low ionization potential (Rb 4.177 eV).

Ionization efficiency measurement

The microbalance can measure the atomic flux (Φ_{total}) that exit the ion source at a certain temperature (T). Then the microbalance is removed and the ion current signal is measured in the Faraday cup. This measure gives the ionic flux (Φ_{ion}). The ionization efficiency is:

$$\epsilon_{ion} = \frac{\Phi_{ion}}{\Phi_{total}} \quad (\text{B.1})$$

This efficiency has an error ($\Delta\epsilon_{ion}$) equal to:

$$\Delta\epsilon_{ion} = \epsilon_{ion} \cdot \sqrt{\left(\frac{\Delta\Phi_{ion}}{\Phi_{ion}}\right)^2 + \left(\frac{\Delta\Phi_{total}}{\Phi_{total}}\right)^2} \quad (\text{B.2})$$

From the ionization efficiency it is possible to deduce the ionization degree value (α_{ion}) and its error ($\Delta\alpha_{ion}$):

$$\alpha_{ion} = \frac{\epsilon_{ion}}{1 - \epsilon_{ion}} \quad (B.3)$$

$$\Delta\alpha_{ion} = \frac{\Delta\epsilon_{ion}}{(\epsilon_{ion})^2 \cdot (\epsilon_{ion}^{-1} - 1)^2} \quad (B.4)$$

The Saha-Langmuir equation defines the ionization degree as:

$$\alpha_{ion} = \frac{n_i}{n_0} = \frac{\sigma_i}{\sigma_0} \cdot \exp\left(\frac{\varphi - W_{IP}}{k_B T}\right) \quad (B.5)$$

where n_i and n_0 are the ion and neutral concentrations, σ_i and σ_0 are the statistical weights of ionic and atomic ground states respectively, φ is the work function of the material [eV], W_{IP} is the ionization potential of the element of interest [eV], k_B is the Boltzmann constant and T is the temperature [K].

Work function value

Gathering B.4 with B.5 the equation results (logarithm form):

$$\ln(\alpha_{ion}) = \ln\left(\frac{\sigma_i}{\sigma_0}\right) + \frac{\varphi - W_{IP}}{k_B T} \quad (B.6)$$

The equation B.6 has the form of:

$$y = ax + b \quad (B.7)$$

where: $y = \ln(\alpha_{ion})$, $a = \ln(\frac{\sigma_i}{\sigma_0})$, $b = \frac{\varphi - W_{IP}}{k_B}$ and $x = 1/T$.

The slope given by the equation B.7 permits to obtain the work function with the following equation:

$$\varphi = k_B \cdot b + W_{IP} \quad (B.8)$$

Example

An estimation of the ionization degree for Rb is gather in table B.1. The values of $1/T$ and $\text{Ln}(\alpha_{ion})$ can be plotted as seen in figure B.1. The slope of the graph (4837.8) corresponds to a work function of 3.76 eV for the hot-cavity material. By this method, it will be possible to compare different materials in order to reduce the contamination production inside the hot-cavity.

Table B.1 – Estimated values of the ionization efficiency to calculate the work function value of a material.

T (K)	Ionization efficiency	Ionization degree	$\text{Ln}(\alpha_{ion})$	$1/T$
500	0.004%	$4.00 \cdot 10^{-5}$	-10.13	$2.00 \cdot 10^{-3}$
750	0.150%	$1.50 \cdot 10^{-3}$	-6.50	$1.33 \cdot 10^{-3}$
1000	0.614%	$6.18 \cdot 10^{-3}$	-5.09	$1.00 \cdot 10^{-3}$
1250	1.573%	$1.60 \cdot 10^{-2}$	-4.14	$8.00 \cdot 10^{-4}$
1500	2.917%	$3.00 \cdot 10^{-2}$	-3.51	$6.67 \cdot 10^{-4}$
1750	4.337%	$4.53 \cdot 10^{-2}$	-3.09	$5.71 \cdot 10^{-4}$
2000	5.837%	$6.20 \cdot 10^{-2}$	-2.78	$5.00 \cdot 10^{-4}$
2250	7.515%	$8.13 \cdot 10^{-2}$	-2.51	$4.44 \cdot 10^{-4}$

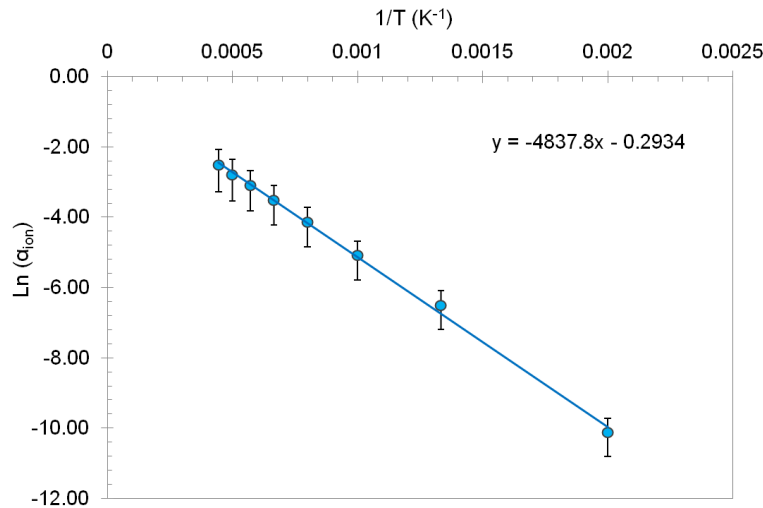


Figure B.1– Work function calculation example by the measurement of the surface ionization degree of Rb. The logarithm of the ionization degree as a function of the inverse of the temperature.

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- J.L. Henares, L. Hijazi, N. Lecesne, B. Bastin, T. Kron, J. Lassen, F. Le Blanc, R. Leroy, B. Osmond, S. Raeder, F. Schneider and K. Wendt. Hot-cavity studies for Resonance Ionization Laser Ion Source. *Nuclear Instruments and Methods for Physics Research A* (2015). doi: 10.1016/j.nima.2015.10.061.
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Abstract

This doctoral thesis describes the implementation, optimization and development of a Resonant Ionization Laser Ion Source (RILIS) at the GANIL facility (Caen, France). The RILIS is a selective ion source technique which is based on a step-wise resonant excitation process where the elements of interest are ionized via atomic resonant excitation by laser radiation. The off-line RILIS test bench at GANIL consists of three tunable titanium:sapphire lasers and a hot-cavity ion source. In this thesis, a new three-step Zn ionization scheme was developed and the already known ionization schemes for Sn have been compared in order to identify the best ionization scheme for a titanium:sapphire laser system. Furthermore, several configurations of the ion source geometry were tested (two diameters and two lengths) in order to provide an optimal ionization efficiency and ion beam emittance. An increasing of the ionizer diameter was found to enhance the production of laser ionized elements, while no enhancement was observed by modifying the ionizer length. The time-of-flight of the generated ions was also studied for the different ion source configurations at several temperatures. The time-of-flight can provide crucial information about the ion formation inside the ion source cavity. In addition, a technique to measure the convolution of the spatial and energetic spread of the ion bunches at the detection point was developed. Finally, two contamination reduction techniques were studied: Firstly, the ions trajectories through the ion source cavity can be modified by the electric field generated due to resistive heating. Therefore, a study of the appropriate configuration of the electric field direction was performed to reduce the alkali contamination of the ion beam. Secondly, a low work function material was tested in order to reduce the production of alkali contaminants and molecular sidebands generated inside the ion source.

Résumé

Cette thèse doctorale décrit l'implantation, l'optimisation et le développement d'une source d'ions par ionisation laser résonante (RILIS en acronyme anglais) à GANIL (Caen, France). RILIS est une technique sélective de production d'ions qui est basé sur un processus d'excitation résonante par étapes successives dans lequel les éléments d'intérêt sont ionisés via l'excitation de résonance atomique par le rayonnement laser. Le banc de test RILIS à GANIL consiste en trois lasers titane:saphir et une source d'ions à cavité chaude. Dans cette thèse, un nouveau schème d'ionisation a été développé pour Zn et les schèmes d'ionisation déjà connues pour Sn ont été comparés afin de déterminer le meilleur schéma d'ionisation pour un système laser titane:saphir. Par ailleurs, plusieurs configurations de la géométrie de la source d'ions ont été testées (deux diamètres et deux longueurs) afin de trouver l'efficacité d'ionisation et l'emittance optimales. L'augmentation du diamètre a produit une amélioration de la production d'ions par laser, par contre aucun changement n'a pas été observé en modifiant la longueur. Le temps de vol des faisceaux d'ions ont aussi été étudiés pour les différentes configurations à plusieurs températures. Le temps de vol peut fournir des informations sur la formation des ions à l'intérieur de la cavité de la source d'ions. En outre, une technique pour mesurer la convolution de la distribution spatiale et énergétique du faisceau d'ions a été développée. Finalement, deux techniques ont été testées pour réduire les polluants: En première lieu, le mouvement des ions à travers la source d'ions peut être modifié par le champ électrique, provenant de l'effet Joule. Par conséquent, une étude de la configuration la plus appropriée a été effectuée pour réduire la pollution des faisceaux d'ions par des alcalins. En deuxième lieu, un matériau avec faible travail de sortie a été testé pour réduire la production des polluants alcalins dans la source d'ions.

Extracto

La presente tesis doctoral describe la instalación, la optimización y el desarrollo de una fuente de iones mediante ionización resonante láser (RILIS) en el laboratorio GANIL (Caen, Francia). RILIS es una técnica selectiva de producción de iones basada en un proceso de excitación resonante multi-paso, donde los elementos de interés son ionizados mediante sucesivas excitaciones resonantes atómicas por radiación láser. El banco de pruebas en GANIL consiste en tres láseres titanio:zafiro sintonizables y una fuente de iones (cavidad caliente). En esta tesis un nuevo esquema de ionización se ha desarrollado para Zn y los diferentes esquemas de ionización para Sn han sido comparados con el objetivo de identificar la mejor solución para un sistema láser titanio:zafiro. Por otro lado, varias configuraciones de la geometría de la fuente de iones han sido analizadas (dos diámetros y dos longitudes) con el fin de estudiar las eficiencias de ionización y emitancia óptimas. Un incremento del diámetro del ionizador ha producido un aumento de la producción de elementos ionizados por láser, mientras que ningún cambio se ha observado modificando la longitud del ionizador. El tiempo de vuelo de los iones a diferentes temperaturas también se ha estudiado, pues éste proporciona información crucial sobre la formación de los iones dentro de la cavidad. Además, una técnica se ha desarrollado para medir la convolución de la distribución espacial y energética de los haces de iones. Finalmente, dos técnicas para reducir la producción de contaminantes han sido estudiadas: En la primera, las trayectorias de los iones a través de la fuente de iones pudieron modificarse mediante el campo eléctrico generado por el efecto Joule. Por ello, se realizó un estudio de la configuración del campo eléctrico más apropiada para reducir la contaminación del haz de iones. En segundo lugar, un material con bajo valor de la función de trabajo ha sido examinado para reducir la producción de contaminantes alcalinos dentro de la fuente de iones.

Abstract

This doctoral thesis describes the implementation, optimization and development of a Resonant Ionization Laser Ion Source (RILIS) at the GANIL facility (Caen, France). The RILIS is a selective ion source technique which is based on a step-wise resonant excitation process where the elements of interest are ionized via atomic resonant excitation by laser radiation. The off-line RILIS test bench at GANIL consists of three tunable titanium:sapphire lasers and a hot-cavity ion source. In this thesis, a new three-step Zn ionization scheme was developed and the already known ionization schemes for Sn have been compared in order to identify the best ionization scheme for a titanium:sapphire laser system. Furthermore, several configurations of the ion source geometry were tested (two diameters and two lengths) in order to provide an optimal ionization efficiency and ion beam emittance. An increasing of the ionizer diameter was found to enhance the production of laser ionized elements, while no enhancement was observed by modifying the ionizer length. The time-of-flight of the generated ions was also studied for the different ion source configurations at several temperatures. The time-of-flight can provide crucial information about the ion formation inside the ion source cavity. In addition, a technique to measure the convolution of the spatial and energetic spread of the ion bunches at the detection point was developed. Finally, two contamination reduction techniques were studied: Firstly, the ions trajectories through the ion source cavity can be modified by the electric field generated due to resistive heating. Therefore, a study of the appropriate configuration of the electric field direction was performed to reduce the alkali contamination of the ion beam. Secondly, a low work function material was tested in order to reduce the production of alkali contaminants and molecular sidebands generated inside the ion source.

Key words: *Radioactive nuclear beams, Resonance ionization spectroscopy, Photoionization, Ion sources, Solid-state lasers, Electronic excitation, Ionization – Efficiency, Thermionic emission.*

Résumé

Cette thèse doctorale décrit l'implantation, l'optimisation et le développement d'une source d'ions par ionisation laser résonante (RILIS en acronyme anglais) à GANIL (Caen, France). RILIS est une technique sélective de production d'ions qui est basé sur un processus d'excitation résonante par étapes successives dans lequel les éléments d'intérêt sont ionisés via l'excitation de résonance atomique par le rayonnement laser. Le banc de test RILIS à GANIL consiste en trois lasers titane:saphir et une source d'ions à cavité chaude. Dans cette thèse, un nouveau schéma d'ionisation a été développé pour Zn et les schémas d'ionisation déjà connues pour Sn ont été comparés afin de déterminer le meilleur schéma d'ionisation pour un système laser titane:saphir. Par ailleurs, plusieurs configurations de la géométrie de la source d'ions ont été testées (deux diamètres et deux longueurs) afin de trouver l'efficacité d'ionisation et l'emittance optimales. L'augmentation du diamètre a produit une amélioration de la production d'ions par laser, par contre aucun changement n'a pas été observé en modifiant la longueur. Le temps de vol des faisceaux d'ions ont aussi été étudiés pour les différentes configurations à plusieurs températures. Le temps de vol peut fournir des informations sur la formation des ions à l'intérieur de la cavité de la source d'ions. En outre, une technique pour mesurer la convolution de la distribution spatiale et énergétique du faisceau d'ions a été développée. Finalement, deux techniques ont été testées pour réduire les polluants: En première lieu, le mouvement des ions à travers la source d'ions peut être modifié par le champ électrique, provenant de l'effet Joule. Par conséquent, une étude de la configuration la plus appropriée a été effectuée pour réduire la pollution des faisceaux d'ions par des alcalins. En deuxième lieu, un matériau avec faible travail de sortie a été testé pour réduire la production des polluants alcalins dans la source d'ions.

Mots clés: *Faisceaux d'ions radioactifs, Spectrométrie d'ionisation résonante, Photoionisation, Sources d'ions, Lasers à solide, Excitation électronique, Ionisation – Efficacité, Effet thermoionique.*

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