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Abstract

For nuclear reactions in which super-heavy nuclei can be formed, the essential difference between the fusion process followed by fission and non-equilibrium processes leading to fission-like fragments is the reaction time. Quite probable non-equilibrium processes, characterized by very short reaction times, are highlighted thanks to mass-angle correlations. However, long lifetime components associated with fission following fusion have been observed with two independent experimental techniques, providing evidence for the formation of compound nuclei with Z = 120 and 124, followed by mass asymmetric fission.

1 Introduction

In reactions between two very heavy nuclei, the tiny cross-sections associated with evaporation residue detection makes it very difficult to demonstrate the formation by fusion of super-heavy nuclei (atomic numbers Z > 110). Even if compound nuclei are formed, they decay dominantly by fission, symmetric or possibly asymmetric in mass. Therefore, experimental fusion cross-sections can only be reached through fission fragment detection. However, the distinction between fusion followed by fission (fusion-fission) and faster non-equilibrium processes (often called quasi-fission) is very tricky because the fission fragments and fission-like fragments from quasi-fission can be quite similar in mass, atomic number and energy [1, 2, 3].

In most of the experimental work, the discrimination between fusion-fission and quasi-fission reactions is somewhat arbitrary, based on considerations of the mass symmetry in the exit channel (ignoring thus any possible asymmetric fission) or the width of the mass and energy distributions. In fact, the objective difference between quasi-fission and fusion-fission is the reaction time [3, 4, 5]. After the fusion step, the nucleons are trapped within a potential pocket, and the composite system needs time to find its way to scission. By contrast, in quasi-fission reactions the nucleons of the system are not trapped and a very fast separation into two fission-like fragments takes place. Therefore, the most reliable experimental criterion that can be used to discriminate between fusion-fission and quasi-fission reactions is the reaction time. In the following, we shall first present recent reaction time measurements through mass-angular correlations, highlighting fast processes leading to fission-like fragments in reactions between very heavy nuclei. We shall then present results of experiments [5, 6] in which long lifetime components ($\tau > 10^{-18}$s) characteristic of fusion reactions were observed, associated with part of the reactions leading to fission-like fragment production.

2 Reaction time from mass-angle distributions

The correlation between the mass of the reaction products and their emission angle can be used to highlight binary reactions in which the sticking time is shorter or of the same order as the rotational period of the composite system [3, 4]. For such fast reactions, the angular distributions measured as a function of the mass asymmetry in the exit channel present maxima that can be linked to lifetimes of the composite system, assuming for each mass asymmetry a single fast component in the reaction time distribution. By contrast, for fusion-fission reactions associated with very long lifetimes, the composite system lives much more than one rotational period and flat angular distributions are expected.

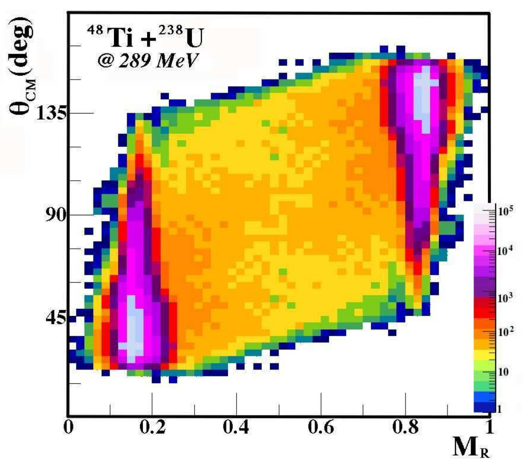

Figure 1: Correlation between the center-of-mass deflection angle (θ_{CM}) and the mass ratio (M_R).

Mass-angle distributions for heavy systems have been recently studied at the Heavy Ion Accelerator Facility at the Australian National University. In these experiments [4, 7], the two coincident fragments from binary reactions were measured by large area position sensitive multiwire proportional counters allowing the determination of the detection angle and of the mass asymmetry in the exit channel. Figure 1 presents, for the heaviest system presently studied at an energy above the Coulomb barrier, ^{48}Ti + ^{238}U at 289 MeV ($E/B \sim 1.1$), the cross-section $\frac{d^2\sigma}{d\theta_{CM} \times dM_R}$ as a function of the center-of-mass scattering angle θ_{CM} and of M_R, the ratio of fragment mass to compound nucleus mass. The minimum cross-sections observed for symmetric splittings at any angle confirm for this system the vanishing of symmetric fission already inferred from studies on lighter systems [3, 4, 7]. For mass asymmetric splittings, the angular distributions clearly present maxima between 30 and 90° for $M_R < 0.5$ (90 and 150° for $M_R > 0.5$), indicating reactions lasting less than the rotational period. Assuming reaction time distributions with a single fast component, most probable reaction times $\widetilde{t}_{react} \lesssim 10^{-20}$s can be inferred, even for the longest quasi-fission reactions associated with the largest deflection angles with respect to grazing trajectories. These quasi-fission times are in good agreement with fully microscopic quantum calculations [8]. Nevertheless, it must be stressed that, for mass asymmetries $0.3 < M_R < 0.4$ ($0.6 < M_R < 0.7$), a region possibly populated by mass asymmetric fission, the angular distributions are quite broad. It seems therefore difficult to exclude, in addition to the fast component associated with quasi-fission reactions, long lifetime components characteristic of fusion followed by asymmetric fission.

3 Long lifetime components

The reaction time $\widetilde{t}_{react} \lesssim 10^{-20}$s inferred from the measured correlations between mass and angle for capture reactions (either complete fusion followed by fission or quasi-fission reactions) in the super-heavy nucleus domain clearly demonstrates the presence of non equilibrium processes. However, it does not rule out longer reaction times for a part of the events. Two different experimental techniques, which both present the advantage of being independent of any nuclear model, have been applied to detect long lifetime components ($\tau > 10^{-18}$s), characteristic of fusion reactions: the blocking technique in single crystals (section 3.1) and the X-ray fluorescence technique (section 3.2).

3.1 Long lifetime components from the blocking technique in single crystals

Three different systems have been studied in reverse kinematics with the blocking technique [5]: ^{208}Pb + Ge at 6.2 MeV/u, ^{238}U + Ni at 6.6 MeV/u and ^{238}U + Ge at 6.1 MeV/u, possibly leading to compound nuclei with $Z_{CN} = 114$, 120 and 124, respectively. All the coincident charged products were detected and identified by INDRA [9], a highly efficient detector array covering a solid angle close to 4π sr. In addition, the blocking patterns were measured for all the fragments detected around 20°. The reaction mechanism analysis performed with INDRA shows that, for the two heaviest systems, the reactions are always binary when one of the fragments is detected with $70 \leqslant Z \leqslant 85$ (only 2 fragments with $Z > 6$ in the exit channel). Furthermore, the sum of the atomic numbers of these two heavy fragments is precisely equal to the total number of protons in the system, as shown for example by figure 2 corresponding to the detection of a fission fragment with $70 \leqslant Z \leqslant 80$ in the ^{238}U + Ni system. In addition, these reactions are associated with negligible multiplicities of lighter charged products ($Z < 6$). The detection of a fragment with $70 \leqslant Z \leqslant 85$ provides thus us with an efficient selection of capture reactions.

Thermal vibrations of the atoms of the single crystals used as targets imply (see for example [10] and references therein) that all reactions lasting less than about 10^{-18}s lead to the same value of χ_{min}, the relative yield of fragments detected in the precise direction of the crystal axes. An increase of χ_{min} for capture reactions is thus straightforward evidence for fusion-fission. For the two heaviest systems studied, a significant χ_{min} increase (with respect to the one measured for either deep-inelastic or quasi-elastic reactions that gives us a reference for fast processes) was observed for fission fragments with $70 \leqslant Z \leqslant 85$, indicating the formation of compound nuclei with Z = 120 and 124. A minimum proportion of 10% of fusion-fission reactions was directly inferred from the χ_{min} increase for the detected fragments. By contrast, no long lifetime components could be evidenced for $Z_{CN} = 114$, possibly due to the compound nucleus neutron number being much lower than the one usually predicted for the nearest shell closure (N = 184).

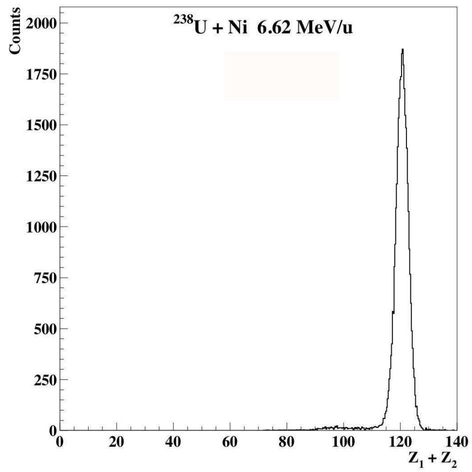

Figure 2: Sum Z_1+Z_2 of the atomic numbers of the two coincident fission-like fragments for $70 \leqslant Z_1 \leqslant 80$.

3.2 Long lifetime components from the X-ray fluorescence technique

During a collision between two heavy ions, vacancies are created in the inner electronic shells of the unified atom [11, 12]. The vacancies are thereafter filled by transitions of electrons from outer electronic shells, giving rise to X-ray fluorescence (in the case of very heavy atoms, the fluorescence quantum yield can be accurately estimated as 1.0). Considering the independent lifetimes of the electron vacancies and of the compound nucleus, sizable probabilities of X-ray fluorescence from the unified atom can only be observed when the compound nucleus lifetime is at least of the same order of magnitude as the vacancy lifetime. Nuclear lifetimes of excited uranium nuclei have been estimated with the fluorescence technique in good agreement [13, 14] with those inferred from the blocking technique in single crystals [15]. Since the lifetime of a vacancy in the K shell of a super-heavy atom is of the order of 10^{-18}s [16], the multiplicity of X_K rays with an energy characteristic of the unified atom provides us with a sensitive probe for long lifetime components.

Unlike the blocking technique that requires good quality single crystals as targets, the X-ray fluorescence technique can be in principle applied to any combination of projectile and target nuclei, giving thus access to investigations of a broader Z range. Furthermore, it makes possible the use of isotopically enriched targets in order to study the long lifetime component production as a function of the compound nucleus isospin. However, this technique had never been previously used in the super-heavy atom domain. Therefore, the system ^{238}U + ^{64}Ni at 6.6 MeV/A, leading to $^{302}_{120}$X compound nuclei, has been chosen for a first experiment [6], providing us both with a test of the fluorescence technique and a cross-check with the crystal blocking results.

The transitions from outer electronic shells to the K shell of $^{302}_{120}$X atoms have been calculated [17, 18] with a multi-configuration-Dirac-Fock (MCDF) approach [19, 20]. Only 3 main X_K transitions are predicted, shown in figure 3, yellow lines, for an ion with a charge 1+ for different nuclear lifetimes (the lines are broadened by the Weisskopf effect resulting from the finite nuclear lifetime [21] and by the Doppler effect due to the experimental set-up). For nuclear lifetimes associated with quasi-fission reactions ($\tau_{nucl} \sim 10^{-20}$s) [3], the 3 lines are so broad that the very weak fluorescence yield only gives rise to a continuous background. For longer nuclear lifetimes, 3 lines can still be observed, but the fluorescence probability is reduced by roughly a factor 30 between 10^{-17}s and

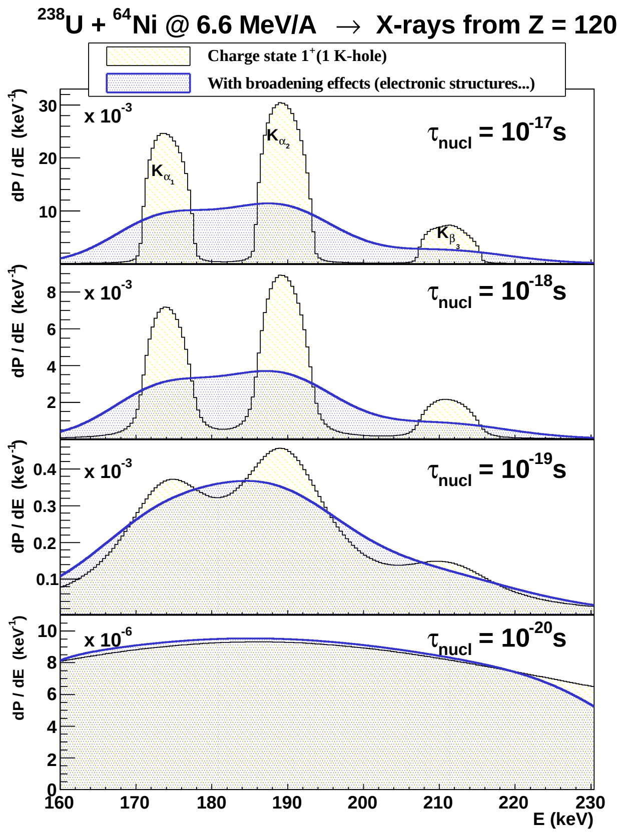

Figure 3: Dominant X_K lines for different nuclear lifetimes for Z= 120 atoms in a charge state 1+ (yellow curves) and for a more realistic electronic structure distribution (blue curves, see text). The Doppler effect associated with the experimental set-up described in section 3.2 is taken into account for the yellow and blue curves.

10^{-19}s. It must be stressed however that, in fusion reactions, the atoms are actually formed with broad distributions of charge states and electronic configurations giving rise to slight shifts in the transition energies. Therefore, the 3 lines possibly detected in coincidence with fission fragments merge, even for the longest nuclear lifetimes (figure 3, top), in a single peak at an energy around 190 keV, with a width of about 50 keV (blue lines in figure 3).

During the experiment, the fission fragments were detected between 16° and 70° by telescopes (ionization chambers followed by double-sided silicon strip detectors). Coincident photons were detected by 3 planar germanium detectors, operated under vacuum and covering a solid angle $\Omega \approx 0.8$ sr . The 3 detectors were located at the same polar angle ($\theta = 127°$), but at 3 different azimuthal angles ($\phi = 30, 150$ and 270° with respect to a vertical plane perpendicular to the beam direction). The photon energy spectrum measured by the germanium detector located at $\phi = 270°$ in coincidence with fission-like fragments (fragments detected with $35 \leqslant Z \leqslant 90$) is presented in the top left panel of figure 4 and in its bottom left panel after background subtraction. Two peaks are clearly seen around 150 and 190 keV. The peak at 150 keV is precisely located at the energy observed in singles measurements (as well as at the energy observed in the random coincidence spectra) for the 158.8 keV γ transition from the decay of the first rotational band of uranium nuclei (the energy of the γ-ray is shifted by the Doppler effect towards lower energy due to the backward angle of the germanium detectors). After correction for random coincidences (right panels in figure 4), the 150

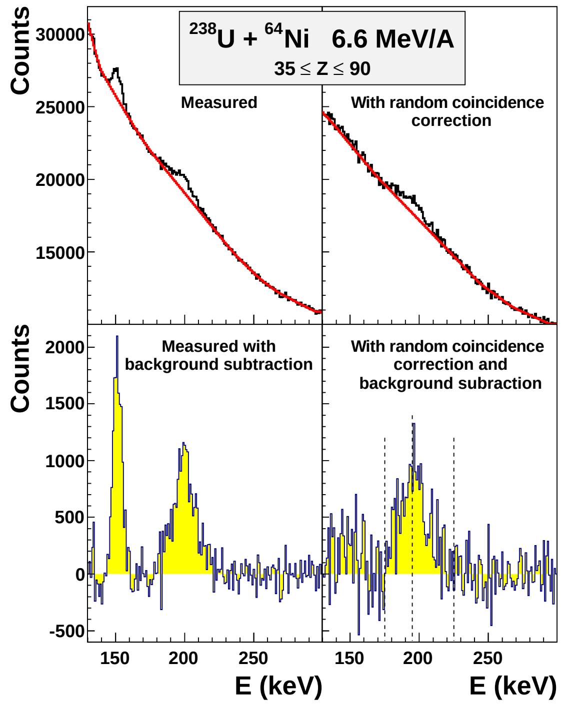

Figure 4: Energy spectrum of photons in coincidence with fission fragments (upper left panel); Spectrum with background subtraction (lower left panel) ; Spectrum with random coincidence correction(upper right panel); spectrum with random coincidences correction and background subtraction (lower right panel)

keV peak is strongly suppressed, confirming the random aspect of these coincidences with fission-like fragments. A peak at 200 keV, arising from the 211 keV γ-transition of the same cascade decay as the 158.8 keV one, is also observed in the singles and random spectra. The broad peak observed in the left panel of fig 3 around 190 keV contains therefore random coincidences with 200 keV γ-rays. However, the peak measured at 190 keV in coincidence with fission fragments is much broader than the random ones at 150 and 200 keV ($\Gamma \sim 50$ keV for the 190 keV peak and $\Gamma \sim 8$ keV for the 200 keV one). Furthermore, since the 211 keV transition feeds the 158.8 keV one during the decay cascade, the peak observed in single and random spectra at 200 keV is much smaller than the one at 150 keV. Therefore, the probability measured for random coincidences with 200 keV γ-rays is much smaller than the one with the 150keV γ-rays. Consequently the peak at 190 keV is not eliminated by the random correction, as shown by the right panels of figure 4, and the good suppression of the more probable 150 keV peak ensures that the 190 keV peak after random correction corresponds to true coincidences with fission-like fragments. Such fission-like fragments can arise either from quasi-fission reactions or from uranium fission or from compound nucleus fission.

The multiplicities of the 190 keV photons for different bins in detected atomic numbers Z are presented in table 1. The maximum multiplicity is reached for $70 \leqslant Z \leqslant 79$. Since this Z selection is only associated with capture reactions (see discussion in section 3.1 and figure 2), the coincident photons are emitted by the composite system or by its fission(-like) fragments. Emission from a fission(-like) fragment should be associated with significantly different Doppler shifted energies measured at $\phi = 30$ and $270°$ ($\delta E \sim 20$ keV), whereas emission from the composite system should lead to identical energies, due to the symmetry of the detection set-up with respect to the beam axis. The energy spectra measured for $70 \leqslant Z \leqslant 85$ at $\phi = 30$ and $270°$ are presented in the upper part of figure 5. The difference of the two spectra (normalized to the surface of the peak between 175 and 225 keV) is shown in the lower part of the figure. No statistically significant difference can be seen between the spectra which is a clear evidence for emission from the composite system. Therefore, considering its energy and its width predicted by MCDF calculations, the peak at 190 keV must be associated to X_K fluorescence from Z=120 atoms.

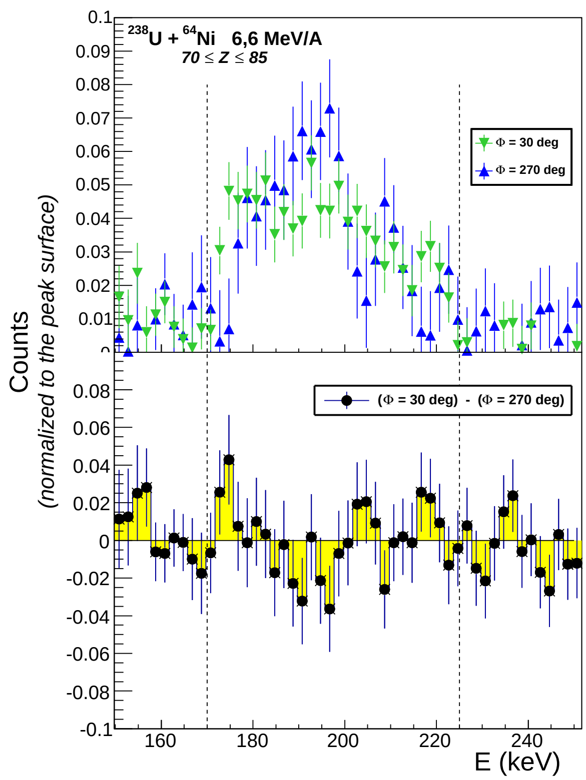

Figure 5: Upper panel: Energy spectra, normalized to the area of the peak between 175 and 225 keV, of photons detected at $\phi = 30°$ (green) and $270°$ (blue). Lower panel: Difference of the two spectra.

The X_K multiplicity $M_{X_K} \sim 0.1$ measured for $70 \leqslant Z \leqslant 79$ is very high. It is indeed of the same order than the K-vacancy creation probability that can be inferred [6] from the one measured in coincidences with elastically scattered projectiles, $P_K^{elast} \sim 0.27$ (elastic scattering and fusion reactions correspond to similar atomic impact parameters). Therefore, M_{X_K} can only be taken into account considering in the reaction time distribution sizable proportions of long lifetime components with $\tau \gtrsim 10^{-18}$s. Assuming an exponential reaction time distribution, at least 50% of the capture reactions associated with this Z bin would correspond to fusion-fission reactions. Considering the isotopically enriched target used for the X-ray fluorescence experiment, these conclusions are in good agreement with the ones inferred from the blocking technique in single crystals.

Table 1: Multiplicity of photons with energy between 175 and 225 keV in coincidence with a fission fragment with atomic number between Z_{min} and Z_{max}

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Z_{min}	Z_{max}	Multiplicity
35	49	0.064 ± 0.002
50	65	0.052 ± 0.002
70	79	0.102 ± 0.004
80	90	0.067 ± 0.004

4 Conclusions

Reaction time measurements give access to unique pieces of information on the reaction mechanisms involved between two very heavy ions at energies slightly above the fusion barrier. Mass-angle correlations highlight fast non-equilibrium reactions and suggest a vanishing of the mass-symmetric fission cross-section for the heaviest systems. In a complementary approach, long lifetime components observed for two very heavy systems testify to mass asymmetric fission following fusion. However, fusion has been evidenced for these systems in experiments in which the fission fragments were detected backward of the grazing angle, whereas most of the cross-section associated with capture reactions was located inside the grazing angle. Therefore, experiments allowing the measurement of long lifetime component probabilities over broad angular and mass ranges are now highly desirable in order to determine cross-sections for fusion as well as for non-equilibrium processes.

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