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Abstract

The present paper reports on a recent study of the production of multiply charged fullerene beams based on an ECR ion source. As collision studies in fundamental physics are demanding intense beams of multiply charged ions of small molecules, clusters and particularly of fullerenes, we have further developed the ion source ECRIS MONO1000[1], originally devoted to produce singly charged ions, towards the production of multiply charged fullerene beams. In this paper, the test measurements performed at the ELISA facility will be described. Typical mass spectra (from pure C_{60} and C_{70} powder) will be shown and the influence of several source parameters (HF-power, support gas, gas pressure, …) will be discussed specifying the conditions necessary for an optimum ion source operation.

I. Introduction

The present development has been initiated mainly by the demand of the collision physics community. Indeed, there is an increasing interest in using beams of multiply charged C_n^{q+} ions in relation with studies of fundamental properties of carbon clusters. As an example, the dissociation energy of C_{60}^{q+} molecules with respect to C_2 emission, which has been discussed very controversially during many years, has been determined to be 9.8 eV only very recently[2]. Another example, the study of the kinetic-energy release, in reactions $C_{60}^{q+} \rightarrow C_{60-2m}^{(q-1)+} + C_{2m}^{+}$, has shown a charge dependence[3]. In order to test the stability criteria for finite-size, multiply charged systems, further experiments with different multiply charged carbon clusters are necessary. The present collaboration between the University of Århus, the CIRIL and the GANIL aims to test the production of multiply charged fullerene and carbon cluster ions with the ECRIS MONO1001 (derived from the ECRIS MONO1000[1]). The experiments have been performed at the ELISA facility[4] in Århus where the analyzing dipole magnet allows a high mass separation up to a maximum mass over charge ratio of ~1500 at an extraction voltage of 22 kV. Previous measurements have already shown that, under specific conditions, intense beams of multiply charged fullerene ions (> enA) can be produced with an ECR type ion source[5].

II. Description of the ion source and of measurements performed at GANIL

The MONO1001 ECRIS is operating at 2,45 GHz , corresponding to a closed resonance surface of 875 Gauss. Its longitudinal and radial magnetic confinement structure is created by two magnetic rings of permanent magnets. Those rings are separated by 80 mm and their internal diameter is about 50 mm producing an axial-symmetric magnetic confinement with a

last closed magnetic surface at 2000 Gauss[6]. A spectroscopic study[7] has established that the MONO1001 ECR plasma, and consequently the distribution of high energy electrons, does not have a flat profile but looks rather like a plasma torus corresponding to the ECR resonance surface. This means that there exists a zone of high energy electron density where the probability for ionization is, for that reason, higher. Preliminary experiments have been performed at GANIL by measuring the ionization efficiency of solid elements and fullerenes. The results are summarized in the Table 1. Typical ionization efficiencies are in the range of 0.1 to 0.2%. These results were expected as the electronic density is rather low ($\approx 7.10^{10}$ e$^-$/cm^2 for a resonance surface of 875 Gauss). For solid elements and powders like fullerenes the standard GANIL oven, which is characterized in more detail elsewhere[8], has been used. Mass spectra of the extracted ions, which could be measured only at low extraction voltages (700 V) due to the limitation by the analyzing magnet, showed the production of fullerene ions in charge states up to q = 4.

III Fullerene beam tests performed at the ELISA facility

III.a Typical mass spectra

Figures 1 and 2 show typical mass spectra measured with pure C_{60} and C_{70} powders using the support gas H_2, respectively. The analyzing device allowed us to extract ions at a voltage of 22 kV and to separate ion masses up to m/q = 900. The spectra can be divided into three zones: the right hand part shows the singly charged C_{60}^+ (C_{70}^+) ions and the corresponding evaporation products C_{60-2m}^+ (C_{70-2m}^+), the intensity of which decreases with increasing m, i.e. with increasing loss of the number of C_2-units. In the C_{70}-case, the intensity of the C_{60}^+-fragment is strongly enhanced due to the high stability of C_{60}. A second part shows multiply charged $C_{60/70}^{q+}$ ions and the evaporation products partly mixed with smaller mostly singly charged carbon clusters (for example m/q = 15 : C_{60}^{4+} - C_{15}^+). Finally, at small

m/q-values small singly and doubly charged carbon clusters can be identified. In figure 3 we show the normalized intensity distribution of singly charged fullerene ions along the evaporation series. For C_{60} and C_{70} powder we observe the same intensity maxima, which are due to the higher stability and higher binding energies of the fullerenes like C_{60}^+, C_{56}^+, C_{50}^+, C_{44}^+ and C_{38}^+. This is in agreement with earlier findings[9]. In figure 4 the normalized intensity distributions of C_{60}^{q+} and C_{70}^{q+} ions are compared. The probability for the production of multiply charged fullerene ions drops faster for C_{70} than for C_{60}. For example, the ratio between the C_{60}^{3+} and the C_{70}^{3+} is roughly 5 and becomes even larger for higher charge states. Therefore, the use of C_{60} powder is more appropriate for getting high intensity beams of multiply charged fullerenes.

III.b Influence of the source parameters

In fact, the number of parameters for tuning the source is quite low: extraction voltage and gap between the plasma and the puller electrode; RF generator power; type and pressure of the support gas and the oven temperature. Due to the storage ring operating mode, the extraction voltage has been fixed at 22 kV. The most favorable gap distance was found to be ~55 mm which has been kept fixed in all test measurements. Three types of support gases have been tested. Figure 5 shows a comparison of the production of C_{60}^{q+} ions with O_2, He and H_2. It should be noted that the increased intensity of C_{60}^{4+} is due to the 'contamination' by C_{15}^+ fragments which seem to be less important in the case of O_2. The behavior of the source is similar for He and H_2 whereas for O_2 the intensity of C_{60}^{q+} (q>2) is decreasing more strongly with increasing charge. Furthermore, a strong "getter chemical effect" appeared with O_2 which obliged us to increase the gas pressure in order to maintain a steady state operation mode for the source. Therefore, most test measurements have been performed with H_2 as support gas. The RF power had to be decreased from 20-30 W with O_2

down to ≈ 1 W with H_2, which made the tuning of the source more difficult as it is the lower limit of the power generator. One of the paramount parameters for obtaining a stable beam is the total ion intensity delivered by the source: it should be of the order of 20 eμA which represents a constraint for the support gas pressure. Finally, the last parameter is the oven temperature. The $C_{60/70}^{+}$ beams appear at an oven power of 1.3 W which corresponds to a temperature of about 350 °C. With increasing oven power the beam intensity increases nearly linearly (see Table 2). The RF generator power has almost no influence on the oven heating especially at the low power level usually used. The operating time of the ion source is only limited by the load of the oven container which is 50 mg in our case. Typical operation times are of the order of 10 hours.

III.c Transport efficiency

Further changes of the source bench are necessary in order to improve the transport efficiency ($\approx 2\%$). The support of the ion source which was made out of iron and which has modified the magnetic field at the extraction region and therefore the extracted beam shape will be replaced by an non-magnetic frame. In order to adapt the acceptance of the analyzing magnet to the emittance of the source, an additional ion optics between source and analyzing magnet will be installed. This should focus the beam and enhance the transmission efficiency.

IV Conclusion

We have studied the conditions under which the ECR ion source MONO1001 is able to produce stable intense beams of fullerene ions : up to 80 enA of C_{60}^{+} with a transport efficiency of 2% corresponding to 4 eμA at the source. The influence of the gas pressure, the

oven temperature and the type of support gas has been evaluated. The successful test measurements allowed us to perform collision experiments with multiply charged fullerene beams showing a high stability for a time range of around 10 hours. The beam intensities were high enough to store ions such as C_{60}^{5+} in the storage ring ELISA. In the future, we aim to further improve the performances of this source for the production of other multiply charged molecular ions and to adapt it for the production of bio-molecular beams.

References

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Singly charged ions	Faraday cup current (eμA)	Ionization efficiency (%) for the singly charged and (for all the charges)	Material
H	400	30 (60)	H_2
^3He	1700	20	^3He
^4He	1500	20	^4He
F	100	15	SF_6
Ne	700	90	Ne
S	120	40 (90)	SO_2 or SF_6
Ar	500	90	Ar
Ca	6	1	Ca oven
Cr	2	0.16	Cr oven
Fe	25	....	ferrocene
Kr	120	90	Kr
Xe	....	60 (90)	Xe
Er	0.013 (0.180)	0.2 (0.4)	Er oven
Pb	1.2 (6)	0.1 (0.15)	Pb oven
C_{60}	0.023 (0.5)	0.2	C_{60} oven

Table 1

Oven power (W)	Temperature (°C)	I_{FC} C_{60}^{1+} (enA)	Consumption of fullerene powder (mg/hour)
2 W	410	<2	≈1
2.2 W	430	≈10	≈7
2.5 W	450	≈20	≈15

Table 2
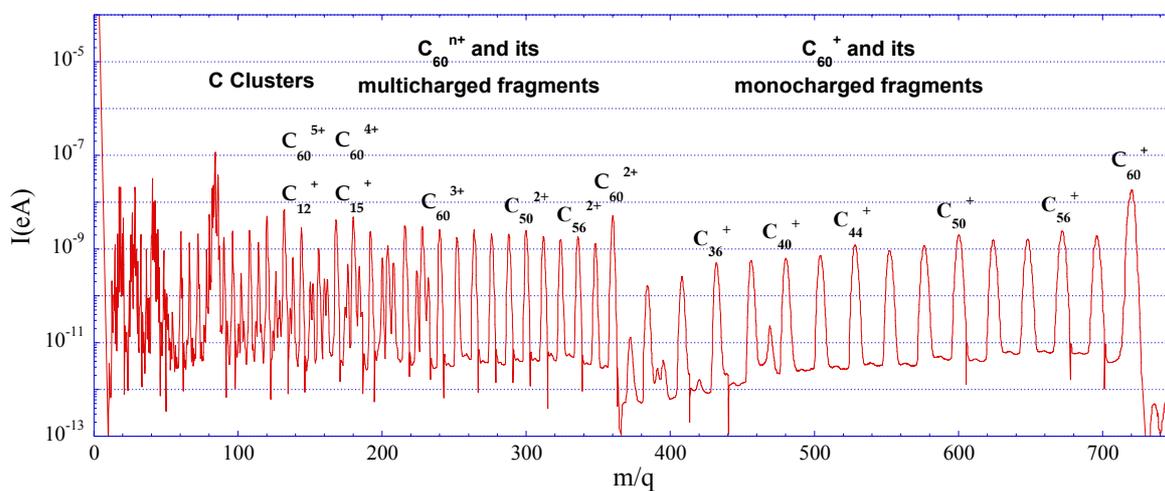
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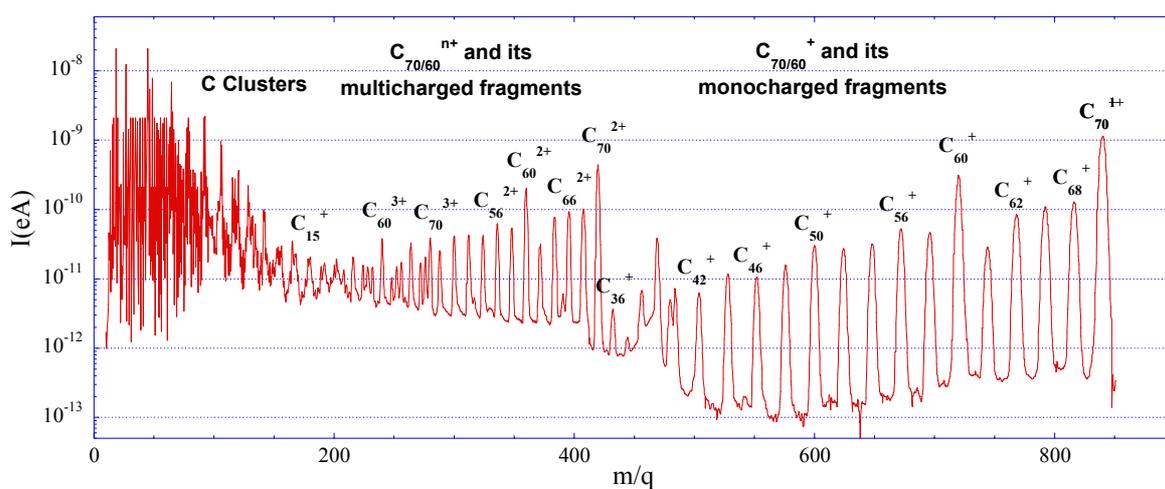
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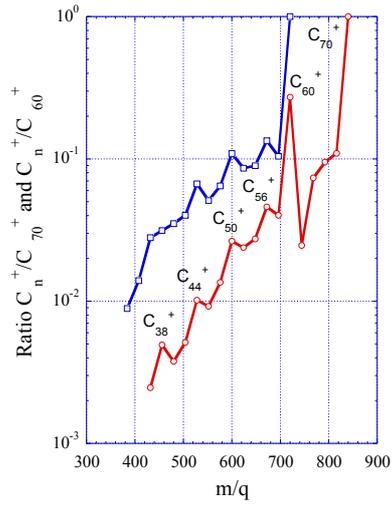
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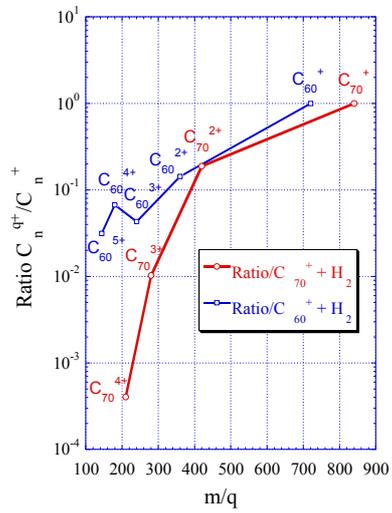
Figure 4
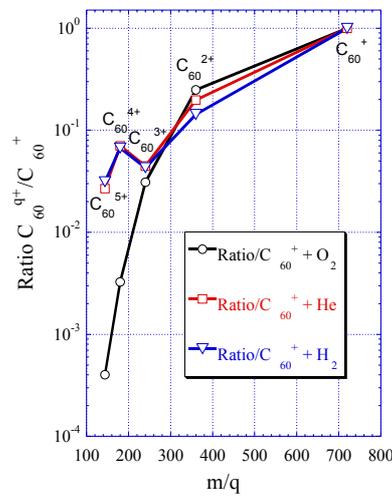
Figure 5

Table 1: Ion intensities produced with the ECRIS MONO1001

Table 2 : Intensities of the C_{60}^{1+} beam and consumption of the fullerene powder as a function of the oven power (transport efficiency : 2%)

Figure 1: Mass spectrum at an extraction voltage of 22 kV using pure C_{60} powder (support gas: H_2)

Figure 2: Mass spectrum at an extraction voltage of 22 kV using pure C_{70} powder (support gas: H_2)

Figure 3: Normalized distribution of singly charged fullerenes produced from C_{60} and C_{70} (support gas : H_2)

Figure 4: Comparison of C_{60}^{q+} and C_{70}^{q+} distributions (support gas : H_2)

Figure 5: Evolution of the C_{60}^{q+} distribution as a function of the support gas