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Branching ratios of successive emission (up to three) of C_{2n}^+ (n = 1–5) fragments in asymmetrical fission processes of C_{60}^{r+} ions (r = 4–6)

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Abstract. Highly charged C_{60} molecules are produced in collisions between neutral C_{60} and multiply charged ions within a large range of temperatures. Successive emission of one, two or three light monocharged fragments referred as one-, two- and three-step processes have been observed. The experimental mass branching ratios for the emission of one C_{2n}^+ fragment from C_{60}^{6+}, C_{60}^{5+} and C_{60}^{4+} ions are compared with the theoretical values using a statistical model. From hotter C_{60}^{6+} ions, branching ratios for three-step processes have been measured and the data are in good agreement with an estimation using the branching ratios in one-step process.

PACS. 36.40.Ei Phase transitions in clusters

1 Introduction

Fragmentation of finite size systems is one of the important deexcitation pathways of hot atomic clusters. Recently, special attention has been devoted to the study of fragmentation of highly charged clusters like C_{60} or sodium [1,2] and more precisely to the decay via fission reactions by the ejection of charged fragments that has no equivalent process in solid state physics. In this paper, we present a systematic measurement of fission processes of multicharged C_{60}^{4-6+} ions at various excitation energies leading to the successive emission of one, two or three light monocharged fragments with even number of carbon.

2 Experimental set-up

Details of the experimental apparatus are given in reference [3]. In brief, neutral C_{60} molecules are evaporated in an oven at a temperature of ≃500 °C. The neutral C_{60} jet crosses perpendicularly a beam of multiply charged ions Ar^{8+} delivered by the new nanogan III E.C.R. Electron Cyclotron Resonance source. The charged reaction products (electrons and recoil ions) are extracted by a transverse an electric field. The ionised C_{60}^{r+} molecules and fragments are analyzed according to their size to charge ratio m/q by a time of flight mass spectrometer. The ions are detected by two M.C.P. Multi Channel Plate and a multianode of 61 pixels linked to 61 individual discriminators. This detector is useful for studying fragmentation processes with a large multiplicity implying the detection of several identical particles (for example: two or three carbon dimmers arriving at the same time). The number "n" of ejected electrons for each event is measured using a semi conductor device placed at the opposite side of the T.O.F. Time Of Flight tube. The outgoing projectile Ar^{6+} is selected and its signal is used as the common stop trigger in the event-by-event acquisition mode. The r initial charge state of the parent ion of C_{60}^{r+} can be estimated using the electron number conservation law ($r = n + 2$). The detection efficiencies of the recoil ions have been estimated to 70% for monocharged light fragments (C_2^+, C_4^+, $C_6^+ \ldots$), 50% for C_{60}^+ and 65% for multicharged C_{60}^{r+} ions. The detection efficiency of the ejected electrons is estimated to be close to 100% in this experiment. It allows to deduce that the energy of ejected electrons from the autoionisation processes is lower than 100 eV. Multicoincidence measurements are performed between the outgoing projectile, the number of ejected electrons and the charged fragments of recoil ions. For each hit on the M.C.P. detector, the number of pixels activated by the electron beam outgoing from the second M.C.P. is also recorded. This additional information is used to well separate the multifragmentation process from the asymmetrical fission process. Indeed, the first process leads to the detection of multiple singly

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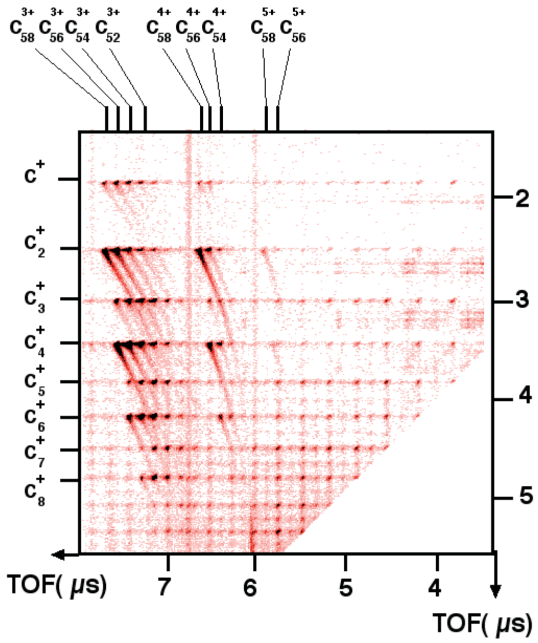

Fig. 1. Correlated two fragments spectrum associated with the detection of two electrons. The C_{60}^{4+} parent ions are selected.

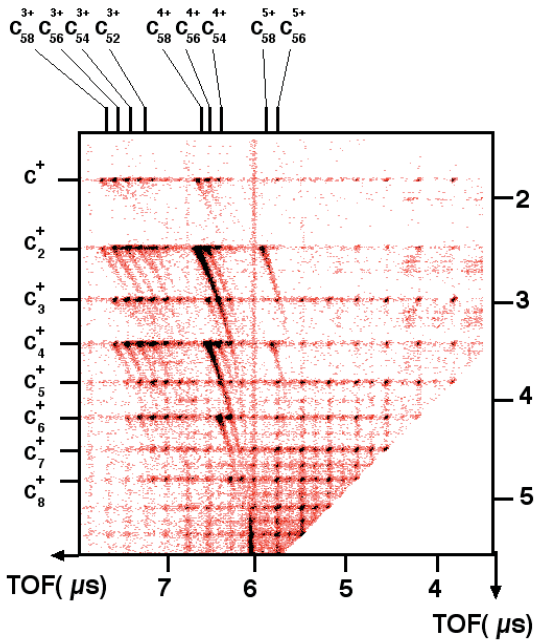

Fig. 2. Same as Figure 1 but with the detection of three electrons. The C_{60}^{5+} parent ions are selected.

charged C_n^+ fragments and each fragment activates mainly only one pixel, while the second process is characterized by the detection of a multicharged fullerene C_{60-2n}^{r+} that in the average activates at least three pixels. By selecting the number of pixels per hit, it is possible to separate the two processes and to extract special information concerning the minor process. As in the fragmentation of high charge states of parent C_{60}, the dominant process is the multifragmentation.

3 Experimental results

Figures 1–3, represent the correlation spectra for selected events with a number of ejected electrons equal to two, three and four respectively and with only two detected fragments. The additional criteria on the number of activated pixel ($\geq$3) ensured that one of the two fragments is a multicharged fullerene ion. The TOF of the heavy fragment is plotted along the X-axis and that of the light fragment is along the Y-axis.

The fragmentation pattern of C_{60}^{4+} parent ions (Fig. 1) shows mainly the light even monocharged fragments (C_2^+, C_4^+, C_6^+, C_8^+, C_{10}^+) in coincidence with C_{58}^{3+}, C_{56}^{3+}, C_{54}^{3+}, C_{52}^{3+}, C_{50}^{3+} ions. The long tail attached to each spot is due to the delayed fragmentation process inside the extraction zone. The $C_{60-2n}^{4+}-C_{2n}^+$ spots observed in Figure 1 are explained by the backscattering effect of electrons on the semi-conductor detector so that a small contribution of the fragmentation pattern of C_{60}^{5+} associated to the peak of 3 electrons is found in the spectrum of C_{60}^{4+} characterized by the peak of 2 electrons (see paper [4] for details). The recoil ion coincidence spectrum

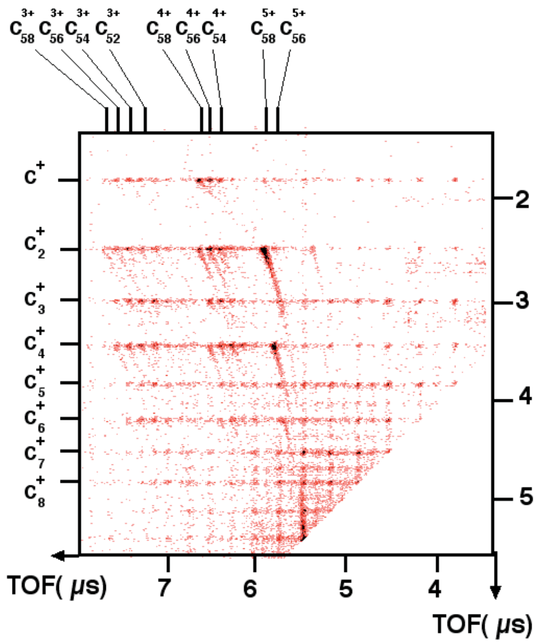

Fig. 3. Same as Figure 1 but with the detection of four electrons. The C_{60}^{6+} parent ions are selected.

of C_{60}^{5+} parent ions is shown in Figure 2. The strong $C_{58}^{4+}-C_2^+$ and $C_{56}^{4+}-C_4^+$ spots indicate the dominant fission channels. The smaller spots, observed in coincidence with C_{60-2n}^{3+} ions, are explained coming from two contributions. The first small contribution is attributed to the emission of a doubly charged light fragment and is not discussed in this paper [5]. The main contribution

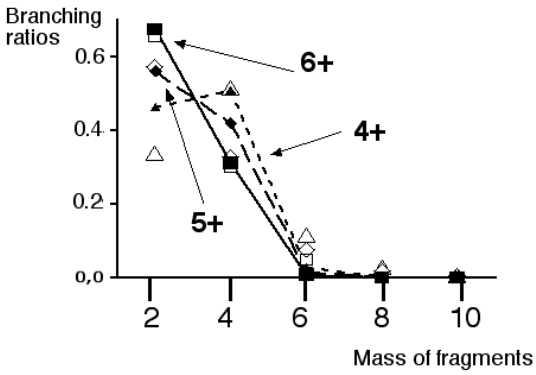

Fig. 4. Branching ratios for the one-step emission $C_{60}^{r+} \rightarrow C_{60-2m}^{(r-1)+} + C_{2m}^+$ as a function of $2m$; ✧, △) (□, ◊, △) experimental values for C_{60}^{6+}, C_{60}^{5+} and C_{60}^{4+} ions respectively. (■, ◆, ▲) theoretical values for C_{60}^{6+}, C_{60}^{5+} and C_{60}^{4+} ions. Lines are to guide the eye.

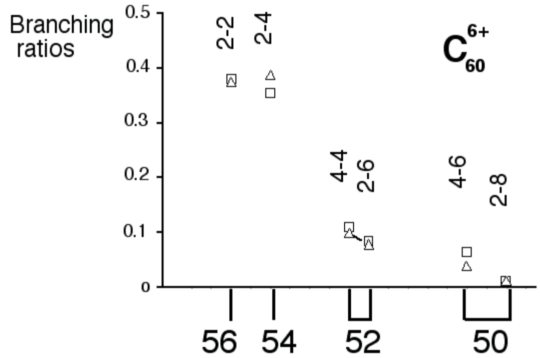

Fig. 5. Branching ratios for the two-step asymmetrical fission processes; (□) experimental values, (△) theoretical values $C_{60}^{r+} \rightarrow C_{60-2m}^{(r-1)+} + C_{2m}^+ \rightarrow C_{60-2m-2n}^{(r-2)+} + C_{2m}^+ + C_{2n}^+$.

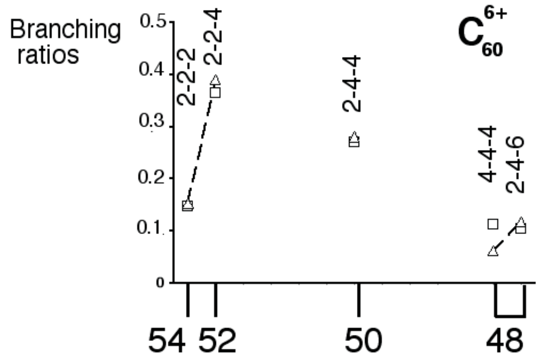

Fig. 6. Branching ratios for the three-step successive emissions; (□) experimental values, (△) theoretical values; $C_{60}^{r+} \rightarrow C_{60-2m}^{(r-1)+} + C_{2m}^+ \rightarrow C_{60-2m-2n}^{(r-2)+} + C_{2m}^+ + C_{2n}^+ \rightarrow C_{60-2m-2n-2p}^{(r-3)+} + C_{2m}^+ + C_{2n}^+ + C_{2p}^+$.

is due to the two-step successive fission process where only one monocharged light fragment is detected. In Figure 3, the spectrum of C_{60}^{6+} parent ions shows two intense spots attributed to the one step emission of C_2^+or C_4^+. The spots in coincidence with C_{60-2n}^{4+} ions are attributed mainly to the two-step successive emission of two light fragments from C_{60}^{6+} ions. The spots in coincidence with the C_{60-2n}^{3+} ions are attributed to the contributions of three-step emission. To measure precisely the contributions of such two-step and three-step emission processes, we extracted from the event files the number of events for each channel characterized by the coincidence detection of three ions $C_{60-2n-2m}^{4+}$–C_{2n}^+–C_{2m}^+ and four ions $C_{60-2n-2m-2p}^{4+}$–C_{2n}^+–C_{2m}^+–C_{2p}^+ (not show here). A special care has been taken to detection efficiency corrections. Details of the analysis will be given in a forthcoming paper. From these measurements, we determined the ratios of one-, two- and three-step successive emission events with respect to the total asymmetrical fission events involving the emission of even fragments to be equal to 58%, 35% and 7% respectively for C_{60}^{6+} ions. The branching ratios for the one step emission are presented in Figure 4 for C_{60}^{4+}, C_{60}^{5+} and C_{60}^{6+} ions.

It is noteworthy that the population distributions shift towards lighter fragment C_2^+ as the charge state of the parent C_{60}^{r+} increases. Similar population distributions have been already reported in paper [6] for C_{60}^{9+}–C_{60}^{4+} ions prepared in Xe^{25+}–C_{60} collisions.

Following the paper [6] we have calculated the relative population distribution as a function of the number of carbon atoms of the light fragment for each C_{60}^{r+} ion (r = 4–6) using a statistical model. The reaction rate constant $A(E^*)$ is thus given by

$$A(E^*) = n_s \nu_\circ \left(1 - \frac{B}{E^*}\right)^{3m-7}$$

where n_s, the degeneracy factor, is the number of atoms on the surface of the C_m fullerene, $n_s = m$, and $\nu_\circ = 2.7 \times 10^{-13}$ s^{-1} is the Debye frequency of C_{60}.

The fission barrier B for each reaction channel $C_{60}^{r+} \rightarrow C_{60-2m}^{(r-1)+} + C_{2m}^+$ has been estimated using the atomization energy [7] and the ionisation potential of C_p. For one step emission processes, the temperature of C_{60}^{r+} is estimated to about 2600 K. A slight variation of temperature around this value does not change the branching ratios for different charge of C_{60}^{r+} (r = 4–6). The theoretical values are compared to the experimental results in Figure 4. The general tendencies are well reproduced showing that the statistical model gives a good description for the asymmetrical fission of C_{60}^{r+} ions.

For the emission of two or three fragments, the temperatures have been estimated to 3500 K and 4300 K respectively for the C_{60}^{6+} ions. In Figures 5 and 6, the branching ratios for two- and three-step emission for C_{60}^{6+} ions are compared with the estimations using the branching ratios for one-step fission of C_{60}^{6+}, C_{60}^{5+} and C_{60}^{4+} ions (Fig. 4). We have taken the assumption that the branching ratio for the emission of a given fragment does not depend too much on the size of the fullerene. A rather good agreement between the experimental data and the model is obtained.

4 Conclusion

We have measured the successive emission of light fragments from highly charged C_{60}. The branching ratios have been relatively well reproduced using a simple assumption. In future studies, the weaker channels involving the emission of odd fragments should be taken into account.

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