Fragmentation of multicharged C$_{70}^{q+}$ prepared in collisions with F$^+$ ions at 3 keV

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Abstract. Ionization and successive evaporation of C$_{70}^{q+}$ is studied as a function of the internal energy using collision induced dissociation under energy control. Multicharged ions C$_{70}^{q+}$ are prepared in F$^+$ (3 keV) + C$_{70}$ → F$^-$ + C$_{70}^{q+}$ + ne collisions. Up to seven successive evaporation of C$_2$ are observed in an excitation energy range from 40 to 100 eV. The dissociation energies of C$_{70-2m}^{2+}$ (m=1-7) are determined using a statistical cascade model to reproduce the excitation energy distribution of C$_{70}$ parent ions for each dissociation channel. Results are in good agreement with previous theoretical calculations.

1. Introduction

The fragmentation of mono and multicharged C$_{60}$ has drawn great attention from broad scientific communities during the past decades. The specific scientific interest of the fullerene molecule, considered now as model system, is related to its exceptional symmetric and stable structure[1][2][3][4][5]. It offers the opportunity to easily prepare a high-density mass selected jet in the laboratory. Fragmentations of C$_{60}$ or C$_{70}$ depend sensitively on its charge and on its internal excitation energy [6][7][8]. In collisions between multi-charged ions and C$_{70}$, multi-charged C$_{70}$ with relatively low internal energy can be prepared via electron capture process.

In this work, we present a study on the fragmentation patterns of C$_{70}^{q+}$ with well-controlled internal excitation energy in F$^+$ + C$_{70}$ → F$^-$ + C$_{70}^{q+}$ + ne collisions using the CIDEC method (collision induced dissociation under energy control) [7]. Very briefly, the mass spectrum of the recoil ions is recorded in coincidence with the kinetic energy loss of the scattered negative ions F$^-$ leading to the measurement of the internal energy distribution of the charged C$_{70}$ parent ions. Using the Arrhenius law we have estimated the dissociation rates for each step in the fragmentation cascade. The internal energy distributions have been reproduced with a cascade statistical model without any assumption on the excitation energy.

2. Experimental set-up

The experimental arrangement has been described in detail in ref. [9] [10]. A relatively pure C$_{70}$ powder around 98% (C$_{60}$ is around 2%) is evaporated from an oven heated at 650°C corresponding to
the initial internal energy estimated theoretically to 6 eV. The C\textsubscript{70} jet crosses the fluorine ion beam delivered by an ECR ion source at 3 keV energy. Downstream from the interaction region, the scattered projectile ions are analyzed by a cylindrical electrostatic analyzer (R=200 mm) and detected with a channeltron. The charged C\textsubscript{70} or fragments are extracted from the collision region and then analyzed with a Time of Flight (TOF) spectrometer equipped with multichannel plates associated with a multi-anode detector. The electrons ejected from the projectile are accelerated to 20 keV and detected by a silicon surface barrier detector PIPS (Passive Implanted Planar Silicon) in coincidence with the other products in an event-by-event mode. Recoil ion spectra are recorded in coincidence with the detection of F\textsuperscript{-} outgoing projectile and the number of ejected electrons. The energy loss calibration is done using an Argon gas target. To record a spectrum, the scattered F\textsuperscript{-} ions are selected according to their kinetic energy loss by scanning the voltage of the electrostatic analyzer. For each voltage, the TOF of recoil ions and the ejected electron number are measured in multi-coincidence and in event-by-event data acquisition mode.

3. Results and discussions

A typical 2D spectrum named in the following EX-RI (Excitation-Recoil Ion) (figure 1(a)) is built by plotting the Recoil Ion TOF along the horizontal axis and the voltage of the projectile analyser along the vertical axis. The vertical scale was converted to the excitation energy of the target in the F\textsuperscript{+}+C\textsubscript{70} -> F\textsuperscript{-}+C\textsubscript{70}\textsuperscript{2+} reaction based on the CIDEC method[7]. Partial projections on horizontal axis for low and high excitation energies parts of the figure 1(a) are shown in figures 1(b) and 1(c) respectively. For the low energy part, C\textsubscript{70}\textsuperscript{2+}, C\textsubscript{60}\textsuperscript{2+} and C\textsubscript{70}\textsuperscript{3+} peaks are observed. C\textsubscript{60}\textsuperscript{2+} and C\textsubscript{70}\textsuperscript{2+} peaks correspond to the collision for which two electrons capture process occurs leaving the molecule intact with an excitation lower than 40 eV. C\textsubscript{70}\textsuperscript{3+} peak corresponds to the double capture process followed by ionization. As seen on figure 1a, the ionization process starts at relatively low energy, lower than 20 eV. Comparable to the third ionization potential of C\textsubscript{70} estimated to 13.6 eV, this value shows that the

![Figure 1](https://example.com/f1.png)
ionization process is probably a direct process analogue to what is observed in interaction with femtosecond laser[11]. The figure 1c shows the high-energy part from 40 to 100eV of figure 1(a). Peaks of C_{70-2m}^{2+} (m=1-9) and C_{70-2m}^{3+} (m=1-7) are attributed to the successive evaporation processes of C_2. Between 40 to 60 eV, the ionization process of C_{70}^{2+} competes with the C_2 evaporation channel. As expected, the dissociation of C_{70}^{2+} and C_{70}^{3+} presents a statistical behavior. Indeed, for the first C_2 emission the average excitation energy of the C_{70}^{2+} parent ion is about 53 eV, which is much larger than the dissociation energy around 9.5 eV. For the second and third evaporation, an extra amount of energy of about 8 and 16 eV is required, respectively. In the time range of the experiment (extraction time is about of 1.7 µs) up to nine C_2 evaporation have been observed leading to C_{52}^{2+} daughter ion. The corresponding internal energy of the C_{70}^{2+} parent ion is about 100 eV. From the projection onto the vertical axis of each spot of the EX-RI spectrum (figure 1a), we obtained the excitation energy distribution of the parent C_{70}^{2+} ions for the evaporative sequence. Each distribution is deconvoluted from the experimental resolution (5.5eV) and fitted with Gaussian distributions N(C_{70-2m}^{2+}, E) (figure 2).

It is noticeable that the distribution corresponding to the C_{60}^{2+} fragment corresponds to a higher stability of fullerene and that the distribution associated to C_{58}^{2+} and C_{56}^{2+} are broadened. The experimental evaporative sequence of C_{70}^{2+} ions leading to the production of C_{70-2m}^{2+} has been simulated using a cascade model (see the paper [6] for details) up to m=7. We have used the Arrhenius law with the heat capacity of C = 0.0138 eV/K. On figure 3, the modeled excitation energy distributions of C_{70-2m}^{2+} are compared to the experimental distributions N (C_{70-2m}^{2+}, E).

It is difficult to adjust at the same time the centers, intensities and widths. While centers of each distribution are well reproduced, slight disagreements are observed for both intensities and widths. In order to obtain better agreement, additional competitive fragmentation channels should probably be introduced in the model[12]. Indeed the first emitted fragment could be a C_4 or C_6 followed by successive emission C_2 especially for high excitation energy. Although these fragmentation channels are probably very weak, they could explain the broadening of the experimental distributions especially for C_{58}^{2+} and C_{56}^{2+}. The obtained dissociation energies of C_{70-2m}^{2+} (m=1-7) (table 1), treated as free parameters in the model, are found in good agreement with recent calculations[13]. The small and high values for C_{62}^{2+} and C_{60}^{2+} respectively clearly indicate the well-known high stability structure of C_{60} molecule. Comparison with previous measurements on dissociation energies for C_{60-2m}^{2+} (m=1-5) [6] using a C_{60} powder shows also a good agreement.

Figure 2. Internal energy distribution of C_{70}^{2+} parent ions for successive C_2 evaporations. The amplitude of the C_{70}^{2+} peak is divided by a factor 20.

Figure 3. Comparison between experimental and adjusted distributions from C_{68}^{2+} to C_{56}^{2+}.
Table 1. Adjusted pre-exponential factors and comparison between theoretical and experimental dissociation rates obtained with the cascade model from parent ions $C_{70}^{2+}$.

|          | $A$ (s$^{-1}$) | $D_{\text{exp}}$(eV) | $D_{\text{theo}}$(eV) [11] |
|----------|---------------|----------------------|-----------------------------|
| $C_{70}^{2+}$ | 4 $10^{22}$   | 9.4                  | 9.44                        |
| $C_{68}^{2+}$ | 1 $10^{19}$   | 8.5                  | 8.6                         |
| $C_{66}^{2+}$ | 5 $10^{19}$   | 9.0                  | 8.26                        |
| $C_{64}^{2+}$ | 1 $10^{20}$   | 8.5                  | 8.53                        |
| $C_{62}^{2+}$ | 5 $10^{17}$   | 6.8                  | 6.77                        |
| $C_{60}^{2+}$ | 1 $10^{23}$   | 10.1                 | 10.18                       |
| $C_{58}^{2+}$ | 1 $10^{23}$   | 8.3                  | 8.45                        |
| $C_{56}^{2+}$ | 1 $10^{19}$   | 8.2                  | 8.54                        |

4. Conclusion
As a conclusion, we have applied the CIDEC method to measure the internal excitation energy of multicharged C702+ prepared in the primary two-electron capture process, $F^+ + C70 \rightarrow F^- + C70^2+$. The internal energy distribution of C702+ for the evaporation channels, $C70^2+ \rightarrow C70-2m^2+ + mC2$ has been simulated using a cascade statistical evaporation model. The dissociation energies $Dm^2+$ of doubly charged fullerenes have been derived and compared with success to theoretical values.

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