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# Kinetic energy release distributions for $C_2^+$ emission from multiply charged $C_{60}$ and $C_{70}$ fullerenes

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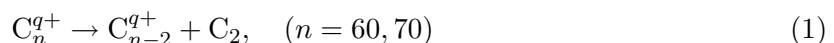
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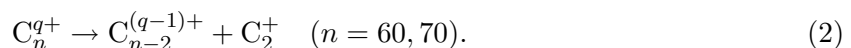
**Abstract.** We present a systematic study of experimental kinetic energy release distributions for the asymmetric fission processes  $C_{60}^{q+} \rightarrow C_{58}^{(q-1)+} + C_2^+$  and  $C_{70}^{q+} \rightarrow C_{68}^{(q-1)+} + C_2^+$  for mother ions in charge states  $q = 4-8$  produced in collisions with slow highly charged ions. Somewhat to our surprise, we find that the KERD for asymmetric fission from  $C_{60}^{q+}$  are considerably wider and have larger most likely values than the  $C_{70}^{q+}$  distributions in the corresponding charge states when  $q > 4$ .

## 1. Introduction

Collision experiments on the extraordinary stable and symmetric fullerenes are particularly useful for the identification of the main mechanisms behind electron transfer processes, energy deposition and fragmentation. Indeed a very large number of such studies has been carried out since the 1990s (see e.g. [1] and references therein). When fullerenes collide with slow highly charged atomic ions several electrons may be transferred and multiply ionized intact fullerenes may be produced. Depending on their internal excitation energy, fullerene ions may also fragment within the experimental time frame of a few microseconds. The kinetic energy release distributions (KERD) in these unimolecular fragmentation processes provide valuable information on the energetics and dynamics of the individual reactions, with positions and shapes governed by the potential energy surfaces describing the interactions of the separating fragments in the exit channel. Unlike neutral  $C_2$  evaporation,



for which kinetic energy release distributions have been measured earlier [2-4], much less is known about the  $C_2^+$  emission process (asymmetric fission),



In this work, we present the first systematic study of experimental KERD for asymmetric fission (2) of  $C_{60}^{q+}$  and  $C_{70}^{q+}$  with  $q = 4-8$ . A single experimental KERD has been reported by Senn *et al.* [5] in 1998, but the more recent results on kinetic energy releases in asymmetric fission of multiply charged fullerenes [6, 7] are given as single (typical) values only. There are also no theoretical studies that could suggest a functional form for KERD of (2).

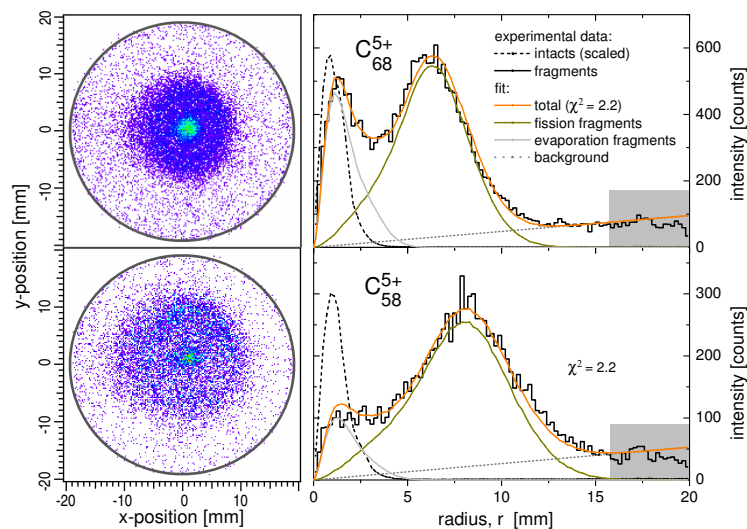
## 2. Experiment

For the present study, we have designed a new linear recoil-ion-momentum spectrometer which has been optimized for measurements of KERD for fragmenting complex molecules. The spectrometer, which is described in more detail in [8], consists of an acceleration region with 19 ring electrodes in a grounded housing with a small aperture for the *collinear* target jet, a field-free drift region, and a position sensitive detector with two microchannelplates (MCP, 40 mm in diameter) and a resistive anode. The dimensions of the spectrometer are chosen such that first order time focusing for different trajectory starting points is achieved. Collimated fullerene target jets effusing from a sublimation oven are crossed with a pulsed (2 kHz, 5  $\mu$ s pulse length) beam of 57 keV Xe<sup>19+</sup> ions ( $v = 0.4$  a.u.) from a 14.5 GHz Electron Cyclotron Resonance (ECR) ion source.

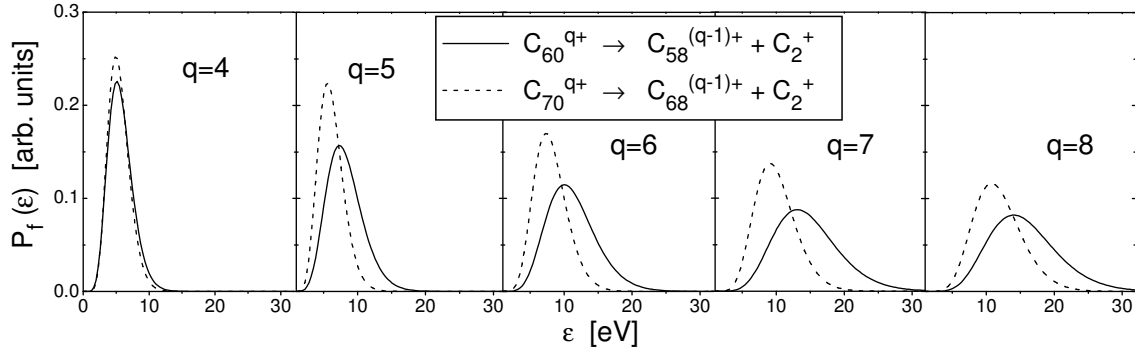
Intact ionized fullerenes and charged fragments are extracted towards the detector directly after the passage of the ion beam by homogeneous acceleration fields of 6.0 or 9.0 V/mm. The ion flight times, as deduced from the time differences between the extraction pulses and signals from the MCP, and the corresponding four anode corner signals, yielding the position on the detector, are stored on an event-by-event basis.

## 3. Data analysis

Two-dimensional detector images for multiply charged C<sub>58</sub> and C<sub>68</sub> fragments from fission (2) and evaporation (1) (see figure 1, left, for an example) are converted to radial intensity distributions (cf. figure 1, right), which are then used to extract distributions of kinetic energy releases,  $P(\epsilon)$ , by means of a simulation and fitting procedure. SIMION 7.0 simulations of the radial distributions are performed taking into account the actual initial conditions and assumed



**Figure 1.** (Color online) Left: Two-dimensional detector images for C<sub>68</sub><sup>5+</sup> (upper panel) and C<sub>58</sub><sup>5+</sup> (lower panel) due to fragmentation of C<sub>70</sub> and C<sub>60</sub> mother ions, respectively. Right: Corresponding experimental radial distributions with fitted/simulated distributions. Contributions from fission and evaporation are indicated. The background is assumed to be uniform (intensity linear in  $r$ ) up to  $r = 16$  mm. Events with  $r > 16$  mm are disregarded in the fit. For comparison, the radial distributions of intact C<sub>70</sub><sup>5+</sup> and C<sub>60</sub><sup>5+</sup> ions are indicated by dashed lines (not to scale).



**Figure 2.** Kinetic energy release distributions in asymmetric fission reactions of multiply charged  $C_{60}$  (—) and  $C_{70}$  (- - -), as functions of charge state,  $q$ . The distributions shown are optimized to reproduce the experimental data in the described simulation and fitting procedure.

probability distributions of the kinetic energy releases for evaporation,  $P_e(\epsilon)$ , and fission,  $P_f(\epsilon)$ . For different assumed analytical forms of the KERD for fission, the simulated total radial distributions (including contributions from evaporation, fission and background) are fitted to those obtained from the experiment by non-linear regression. The  $(P_e(\epsilon), P_f(\epsilon))$ -combinations which give the best fits are taken as the present results.

#### 4. Results and discussion

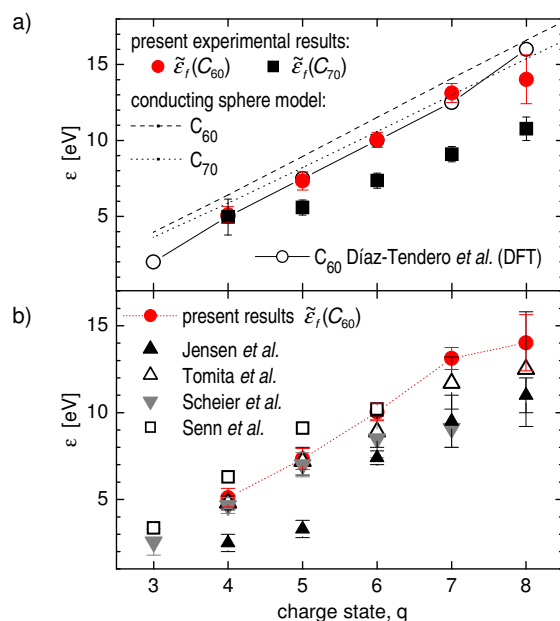
The fission KERD,  $P_f(\epsilon)$ , yielding the best fit results are presented in figure 2. For both  $C_{60}^{q+}$  and  $C_{70}^{q+}$  the width of the distributions increases with  $q$  (probably linked to the increase of the reverse activation barriers). We find that the KERD for  $C_{60}^{4+} \rightarrow C_{58}^{3+} + C_2^+$  and  $C_{70}^{4+} \rightarrow C_{68}^{3+} + C_2^+$  are very similar (cf. left panel of figure 2). For higher charge states  $q$  of the mother ions, however, the distributions for  $C_{70}$  are narrower and shifted towards lower energies.

These KERD have been obtained using the 'model-free' approach by Klots [9],

$$P(\epsilon) = a\epsilon^l \exp\left(-l\frac{\epsilon}{\tilde{\epsilon}}\right), \quad (3)$$

where  $a$  is a normalization factor,  $\tilde{\epsilon}$  is the position of the maximum of the distribution, i.e. the most probable KER value, and  $l$  is a parameter related to the interaction potential between the separating fragments ( $0 \leq l \leq 1$ ). Following Gluch *et al.* [3] and Climen *et al.* [4], we take  $l = 0.5$  for the evaporation KERD, corresponding to a Langevin-like long range interaction between  $C_2$  and the heavier, charged fragment. The overall best fits to our experimental data are obtained by tentatively using equation 3 also for fission and taking  $l = 8.5$  ( $C_{60}$ ) and  $l = 10.0$  ( $C_{70}$ ). For the moment we refrain from an attempt to interpret these values and only regard the resulting functions  $P_f(\epsilon)$  as efficient ways to parameterize the experimental data.

In figure 3, the most likely kinetic energy release values for *fission* of  $C_{60}^{q+}$  and  $C_{70}^{q+}$ ,  $\tilde{\epsilon}_f$ , are shown as functions of  $q$ . In the case of  $C_{60}$ , there is almost perfect agreement with theoretical values for the reverse activation barriers obtained by high level Density Functional Theory (DFT) transition state calculations [10] (cf. figure 3a). A comparison with earlier experimental kinetic energy release measurements is shown in part b of figure 3. The wide experimental distributions which we obtain show that kinetic energy releases may also be several eV smaller or larger than the reverse activation barrier. The lower energies may be explained as due to couplings of the reaction coordinate with other internal degrees of freedom leading to a situation in which the reverse barrier may partially be transformed to internal energy. The larger kinetic energy release



**Figure 3.** Comparison of the present most probable KER values for asymmetric fission of  $C_{60}^{q+}$  and  $C_{70}^{q+}$  with, (a), results from DFT calculations [10] (with lines to guide the eye) and an electrostatic model [6, 11], (b), other experiments on  $C_{60}$  yielding single typical values by Scheier *et al.* [12], Tomita *et al.* [7], Senn *et al.* [5] and Jensen *et al.* [6].

values may possibly relate to remaining electronic excitations. At the moment, the mechanism behind this excitations is not completely clear and needs to be studied in further detail.

We believe that the observed differences between  $C_{60}$  and  $C_{70}$  are, at least partly, related to a larger polarizability due to the larger overall size of  $C_{70}$ . This effect, however, which is taken into account in the electrostatic model calculations [6, 11] plotted in figure 3a, cannot fully explain the observed differences, which are unexpectedly large. Additionally, the probably lower  $C_{70}$  fission barriers [8], the larger number of vibrational degrees of freedom for  $C_{70}$  and  $C_{68}$  in comparison to  $C_{60}$  and  $C_{58}$ , the inhomogeneous charge distribution on  $C_{70}$  ions [13] and the non-spherical shape may play a role. Also the fact that there is a larger number of  $C_{68}$  isomers and that several transformations between them are needed to reach the energetically most stable one, may lead to a situation where less excess energy is released as relative translational energy of the fragments.

### Acknowledgments

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