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Correlation between multiple ionization and fragmentation of C60 in 2-MeV Si2+ collisions: Evidence for fragmentation induced by internal excitation

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Fragment ions from C60 induced by 2 MeV (ν=1.7 a.u.) Si2+ impacts are measured in coincidence with the number distributions of secondary electrons under conditions of single-electron loss and single-electron capture collisions. Multifragmentation, leading to disintegration of cage structure, is found to occur at surprisingly low charge states of r=3. Also, we find that mass distributions of fragment ions are nearly the same for low and capture collisions provided that the number of electrons ejected, due to electronic energy deposition from an incident ion, are the same. Present results indicate evidently that the internal excitation, rather than the charge state r of transiently formed prefragmented parent ions C60r++, plays the essential role in C60 fragmentation in fast heavy ion collisions.

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Collision-induced ionization and fragmentation of C60 has been the subject of intensive theoretical and experimental studies recently (see Ref. [1] and references therein). Leading motivation of study is that microscopic understanding of these fundamental collision processes is important in basic collision physics and in various applied fields involving polyatomic particles, like fullerenes, lying between atoms and solids. Particular effort has been devoted to elucidation of the role of excitation manners (electronic or vibrational) in C60-fragmentation process using various incident ions and charge states. Evidence with the number distributions of outgoing projectile particles using 2.0 MeV-energy heavy ions where the electronic energy loss is dominant, greater importance of the internal excitation is demonstrated in the figure. The partial -loss (qf=3) collisions. This triple coincidence measurement provides detailed fragmentation profiles as a function of the charge states of prefragmented C60r++ parent ions, enabling us to examine the role of initial charge state r in fragmentation processes. It is noted that the present technique is similar to that used in SHCI experiments by Martin et al. [12].

The experiment was carried out using a 1.7 MV tandem Cockcroft-Walton accelerator of Kyoto University. Besides our newly developed detector of secondary electrons and an electrostatic charge selector for outgoing projectiles, the rest of experimental apparatus such as the collision chamber and the time-of-flight (TOF) spectrometer are similar to those described previously [13], and only an essential outline is given below. A well collimated beam of 2.0 MeV Si2+ was incident on a target of effusive molecular C60 beam produced by sublimation of high-purity (99.98%) powder at 550°C. A base pressure of the target chamber was kept below 1×10−6 Pa. Outgoing projectile ions from the collision chamber were charge selected by electrostatically and detected by a movable semiconductor detector. Product ions from C60 were extracted by an electric field of 615 V/cm perpendicular to the incident beam axis and detected by a two-stage multichannel-plate with a front voltage of −4.6 kV. Secondary electrons were extracted to the opposite direction of the product ions and detected by a semiconductor detector (active area=150 mm2) biased at +30 kV. A pulse height spectrum from the detector provides a nc distribution, since simultaneous detection of n electrons results in a total energy deposition of 30nc keV to the detector [14,15].

A typical example of TOF-nc coincidence spectra is shown in Fig. 1 obtained for 1e-loss collisions (Si2+→Si3+). Vertical and horizontal axes are the time-of-flight of product ions and the pulse height of electrons, respectively. Projection of this two-dimensional spectrum onto these axes provides, respectively, a total TOF and a total nc spectrum, as demonstrated in the figure. The partial nc spectrum for any
desired product ion can be obtained from the corresponding TOF region.

As the simplest cases, $n_e$ spectra for parent ions $C_{60}^{r+}$ ($r=1\sim3$) are depicted in the inset of Fig. 1. The principal peak in each $C_{60}^{r+}$-spectrum appears at the expected position of $n_e=r+1$, implying that the electron lost from a $Si^{2+}$ ion is safely detected as well as the $r$ electrons from a $C_{60}^{r+}$ ion. Spectator peaks observed in lower side of the principal peaks are known to originate from electron backscattering at the detector surface, depositing a part of the impacting energy (30 keV) into the detector [14,15]. Taking account of this backscattering effect, observed spectra were reproduced almost perfectly as shown by solid lines calculated using constant fitting parameters for the backscattering probability (0.17) and the backscattering $K$ factor (0.6). Note that these fitting parameters are nearly equivalent to published data [14]. Also we note that the electron-collection efficiency of the present experiment was estimated to be about 0.94.

Using these fitting parameters, $n_e$ distributions for individual fragment ions were deduced accurately. Results for $C_n^+$ ($n=1,3,$ and 11) are presented in Fig. 2 together with the total distributions obtained by the sum of all product ions. Data are plotted as a function of $r$ instead of $n_e$, using the relationship $r=n_e-1$ for loss and $r=n_e+1$ for capture collisions. The partial distribution shows clearly that the average charge state of prefragmented parent ions increases significantly with decreasing cluster size $n$. The total distributions for loss and capture collisions are considerably different from each other. This is due to different contributions from intact parent ions from which only a few electrons are ejected. We find, therefore, that the $1e$-capture process ac-

![FIG. 1. TOF-$n_e$ coincidence spectrum obtained for single-electron loss collisions of 2 MeV $Si^{2+}$ projectiles. Partial distributions for $C_{60}^{r+}$ ($r=1,2,3$) are shown in the inset.](image)

![FIG. 2. Electron number ($n_e$) distributions for $C_{60}^{r+}$ ($n=1,3,11$) as a function of $r$. Total distributions (closed squares) over all products ions are normalized to unity.](image)
only by 27% for 1e-loss collisions (61% for capture ones). The different results obtained in SHCI and our fast ion collisions may be attributed solely to a difference of internal excitation energy deposited into a C\textsubscript{60} molecule. This is because, in our lowly charged fast ions, C\textsubscript{60}\textsuperscript{r+} ions are likely produced in closer collisions receiving a larger amount of excitation energy compared to SHCI collisions.

Second, there exists remarkable resemblance between s\textsubscript{r}-spectra in 1e-loss and s\textsubscript{r+1}-spectra in 1e-capture collisions; compare the spectra of, e.g., r=4 in loss with r=5 in capture collisions. On the contrary, distribution profiles at the same r are significantly different from each other. It is noted that one of r+1 electrons in 1e-capture collisions is captured into a projectile ion and the rest r electrons are purely ejected via ionization processes taking place simultaneously. Note that the simultaneous events among inelastic processes such as charge-changing, excitation, and ionization are typically observed in collisions of fast heavy ions with many-electron atomic targets [21]. Denoting the number of this pure ejection by n\textsubscript{r}, the present result indicates that overall multifragmentation profiles are governed by the amount of n\textsubscript{r}, rather than the apparent charge state r itself. This one-electron-shift of fragmentation profile is more explicitly depicted in Fig. 4 as a function of n\textsubscript{r} where n\textsubscript{r}=r in 1e-loss and n\textsubscript{r}=r−1 in 1e-capture collisions. Here, we plot relative intensities of intact parent ions C\textsubscript{60}\textsuperscript{r+}, medium sized fragments $\sum_{n=4}^{12}$ C\textsubscript{n} and the first three smallest ions $\sum_{n=1}^{3}$ C\textsubscript{n+} with respect to total intensities of all product ions. Plotted data for loss and capture collisions are found to coincide fairly well with each other.

It is known that the electronic energy deposition $E_e$ is shared between excitation and ionization of target electrons with a certain partition rate [22,23]. Thus, it may be possible to estimate the amount of internal excitation energy $E_{int}$ from the values of n\textsubscript{r}. For this estimation, however, exact values of partition rates are required. So far, no experimental and theoretical work has been reported referring to such partition rates for fullerene particles. For other collision systems, two theoretical papers are available for 0.2–10\textsuperscript{4} keV H\textsuperscript{+}+H\textsubscript{2}O [23] and 1.4 MeV/amu U\textsuperscript{32+}+Ne [22]. According to these studies, the partition rate to the excitation branch is about 0.2 at the present incident velocity [23]. As for the ionization branch with a rate of 0.8, it is further divided into two branches of kinetic energies of ejected electrons and their ionization potentials. The partition rate spent for ionization potentials $E_p$ is about 0.25 [22]. If we simply use these partition rates, we obtain $E_{p}(=E_e \times 0.8 \times 0.25) = E_{int}(=E_e \times 0.2)$. The present value of n\textsubscript{r} is related to $E_p$ by

$$E_p \geq \sum_{r=1}^{n} I_r,$$

where the rth ionization potential $I_r$ of C\textsubscript{60} may be calculated by $I_r$(eV) = 3.77 + 3.82r [24]. The internal energy $E_{int}$
that of C\textsubscript{60} occurs strongly (Fig. 3). According to the maximum entropy calculations made by Campbell \textit{et al.} [7], the multifragmentation starts at $E_{\text{int}} \approx 85$ eV and complete disintegration into smaller fragment ions occurs beyond 200 eV. Present rough estimations compare fairly well with these theoretical predictions, although present values seem to be not high enough to induce such strong multifragmentation as observed experimentally. In order to obtain more reliable values of $E_{\text{int}}$ from $n_i$, accurate partition rates in the present collision system are needed. From the close connection between $E_{\text{int}}$ and $n_i$, it follows convincingly that the internal energy of C\textsubscript{60}\textsuperscript{+} in 1e loss may be essentially equivalent to that of C\textsubscript{60}\textsuperscript{(r+1)+} in le-capture collisions, because of the same values of $n_i$ for both cases. Together with the experimental findings of resemblance of fragmentation profiles at the same $n_i$, it leads us to a conclusion that the fragmentation profile in fast collisions is essentially governed by the internal excitation energy rather than the apparent charge state $r$. For higher charge states ($n_i \sim r > 10$), however, the Coulomb explosion may also become responsible for the fragmentation.

In summary, we performed triple coincidence measurements for TOF, $n_i$ and $q_f$ to study correlated ionization and fragmentation processes of C\textsubscript{60}. A careful comparison of the fragmentation profiles between 1e-loss and 1e-capture collisions shows that the C\textsubscript{60} fragmentation is governed by the number of purely ejected target electrons, implying that the internal excitation energy is more important than the apparent charge state $r$ itself in 2-MeV Si\textsuperscript{2+} collisions. It should be addressed, however, that accurate values of energy-partition rates are urgently required to know the role of internal excitation effect more quantitatively.

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