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The fragment ion distribution of C_{60} in close collision with fast carbon ions

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Abstract. We have measured the mass distribution of fragment ions of C_{60} produced by collisions with 15.6 MeV carbon ions in different charge states. Close collisions were selectively measured using the coincidence method with the change of the projectile charge state. For the electron capture and loss by $C^{5+,6+}$ projectile ions, which are the K-electron processes, the multifragmentation was observed evidently. In L-electron loss channels of the C^{2+} projectile, the peaks of the multiply ionized C_{60} ions and the multiply ionized fullerene-like fragment ions are more intense or as intense as the small fragment ions. We suppose that we can roughly classify the intermediate states of C_{60} through the measurement of fragmentation in coincidence with the change of charge state for various projectiles.

1. Introduction

There is increasing recent interest in the fragmentation of C_{60} induced by collisions with atoms, molecules, ions, electrons, photons and C_{60} [1–22]. In most of these studies, parent ions, C_{60}^{q+}, and fullerene-like fragment ions corresponding to the loss of even-numbered clusters were shown. The fragmentation processes leading to such even-numbered cluster loss were fully discussed there. The processes proposed there can be roughly classified into two categories: a successive C_2 evaporation [2] and an initial cleavage process involving the ejection of $C_n (n > 2)$ rings and chains [7]. However, not enough detailed studies of prompt multiple fragmentation have been done. LeBrun *et al* [1] measured the fragment-ion distribution for collisions of fast Xe ions with gaseous C_{60}. They postulated that small-size fragments come from multifragmentation of C_{60} and they reproduced the small fragment-ion distribution by calculation using a bond-percolation model with a bond-breaking probability as an adjustable free parameter. Walch *et al* [3] performed the coincidence measurement between fragment ions and projectile final charge state using slow highly charged ions (HCI); only peaks corresponding to small-size fragment ions were shown in their time-of-flight (TOF) spectra for many electron transfer processes. These small fragment ions seem to be produced by Coulomb explosion. Jin *et al* [18] pointed out the usefulness of the TOF correlation between the first and second fragment ions to investigate details of the fragmentation process in the collisions with slow HCI. Campbell and co-workers simulated the multifragmentation of the neutral and singly charged C_{60} system with high internal energy using statistical calculation and molecular dynamics calculation [15, 20, 22].

The fragmentation pattern is thought to relate to the intermediate states before fragmentation. The multifragmentation by fast-ion impact is especially thought to be

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caused by large energy transfer from the projectile ions. It should be important to find a tool of classification of the intermediate states for detailed studies of the fragmentation process. In slow HCI–C_{60} collisions, the number of transferred electrons seems to be useful to classify the Coulomb energy of intermediate states [3]. In slow collisions between C_{60}^{+} and gaseous atoms, the collision energies directly connect to the internal energies of intermediate states. In the fast-ion impact experiment, the measurement of projectile energy loss gives important information for internal energies of intermediate states. However, measurements of projectile energy loss lower than a few keV are not realistic in the MeV u^{-1} energy region. In this energy region, a large energy transfer to C_{60} mainly results from a strong Coulomb interaction between the projectile ion and electrons of C_{60} at small impact parameters. Therefore, the collisions accompanying large energy transfer can be selected using coincidence measurements with the changes of the projectile charge states because the charge-state change occurs predominantly when the projectile trajectories overlap with C_{60}.

We have studied the fragmentation process of C_{60} by the impact of a fast carbon ion through measurements of fragment-ion distributions. The fragment-ion distributions in close collisions were measured in coincidence with the outgoing projectile charge state as well as total fragment-ion distributions.

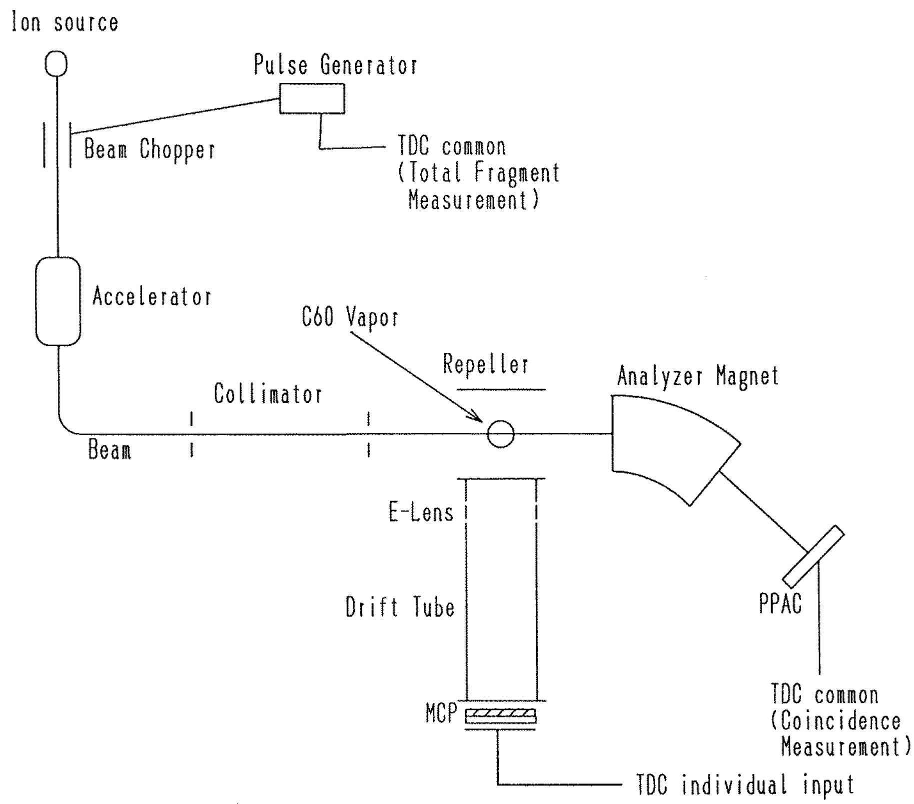


Figure 1. A simplified overview of the experimental set-up. A beam chopper was used to generate a pulsed projectile beam for the total fragment measurement. The drawing is not to scale.

2. Experimental

We measured the mass-to-charge ratio (m/q) of the fragment ions using a TOF method. In figure 1, a schematic diagram of the experimental set-up is shown. A 15.6 MeV C^{q+} beam ($q = 2$, 4, 5, 6) from the heavy-ion linac at RIKEN (RILAC) was collimated with a double-slit system upstream of a collision chamber and passed through a C_{60} vapour target produced by an oven heated to 693 K (C^{5+} impact) and 723 K ($C^{2+,4+,6+}$ impact). We used a beam chopped by pulses of 145 ns width and 50 μs period to measure the total fragment-ion distributions. The beam modulation pulse was used as the common timing pulse for total TOF measurements. In coincidence measurements with charge change of the projectiles, we used a continuous beam without beam modulation. The TOF spectrometer was located at 90° with respect to the beam axis. The C_{60} ions and the fragment ions produced in the collision region were extracted by an electric field, focused by an electrostatic lens and passed through an 18 cm long flight tube. A rather weak extraction electric field of 150 V cm^{-1} was used in order that the TOF peaks were well separated from each other. A couple of multichannel plates (MCP) were used as the fragment ion detector. The total acceleration voltage of product ions was 4.0 kV and the detection efficiencies may not be so high for singly charged fullerene-like fragment ions and C_{60}^{+} [3, 15, 21]. The final charge state of the projectile ion was analysed by a dipole magnet located downstream of the collision chamber. The projectile ions with a selected charge state hit a two-dimensional position-sensitive parallel-plate avalanche counter (PPAC). The common timing signals for coincidence measurements were produced by this PPAC. The common timing signal either from the beam modulation pulse or from the PPAC pulse started the time window of 40 μs (the TOF of C_{60}^{+} is about 30 μs). A timing pulse from the TOF spectrometer within this time window was used as the individual input of a multi-hit (eight-hit) time-to-digital converter (multi-hit TDC). The trailing edge of the time window was used as the common stop signal of the multi-hit TDC. The double-pulse resolution of the fragment ion detector was set at 210 ns by data analysis software to avoid the distortion of TOF spectra caused by ringing pulses from the MCP. If plural fragment ions with the same mass-to-charge ratio are produced, our system cannot detect all the fragment ions consequently.

3. Result and discussion

The TOF spectra of total fragment-ion distribution are shown in figure 2(*a*). These spectra are the sum of all eight channels of our multi-hit TDC. Peaks observed in these TOF spectra are categorized as follows.

(1) Peaks due to singly, doubly, and triply (probably quadruply) charged C_{60} ions.

(2) Peaks corresponding to losses of even-numbered small carbon clusters $(C_2, C_4, \ldots)$ or to sequential losses of carbon dimers from C_{60}^{q+} [2]. The relative intensities of these peaks to their parent peaks C_{60}^{q+} become higher with the charge q.

(3) Peaks corresponding to singly charged small fragment ions C_n^{+} ($n = 1$–15). The peak intensities of the odd-numbered fragments are higher than the neighbouring even-numbered fragments up to $n = 7$.

These three features of the total fragment-ion distributions are qualitatively similar to other studies, especially the study using fast, highly charged, Xe impact by LeBrun *et al* [1].

The intensity of C_{56}^{q+} is almost the same as C_{58}^{q+} or a little larger than C_{58}^{q+} for all q (figure 2(*b*)). This fact might indicate that the branching ratio of C_4 emission is not very small compared with C_2 emission as reported in other collision systems. Radi *et al* [12]

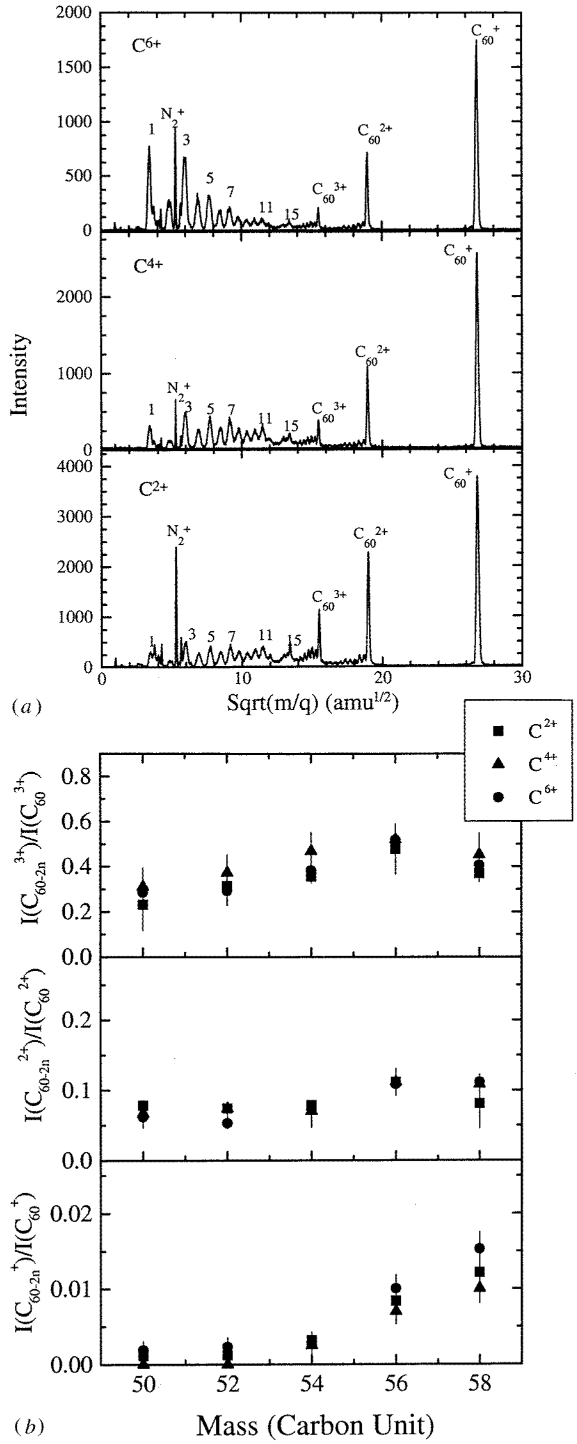


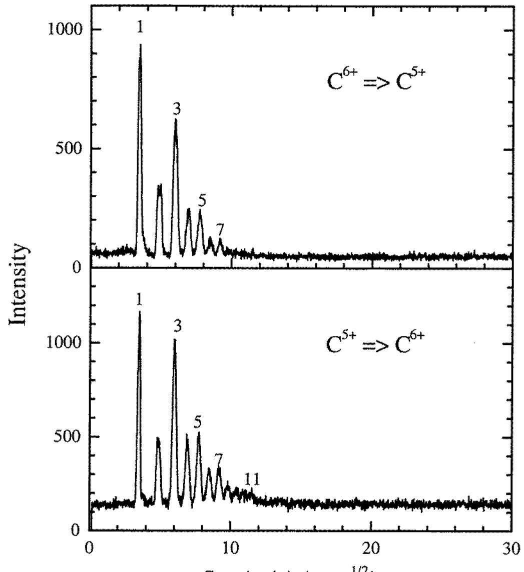
Figure 2. (*a*) TOF spectra for total fragment ion distribution in the collisions with $C^{2,4,6+}$ at 15.6 MeV. The number above each peak shows the number of atoms in the fragment ion. The peak of $n = 15$ includes the peak of C_{60}^{4+}. We can also see the small peak for C_{60-2n}^{4+}. (*b*) The intensity ratio of fullerene-like fragment ions $C_{50,52,54,56,58}^{q+}$ to their parent ion C_{60}^{q+} for $q = 1, 2, 3$. Full square, C^{2+} impact; full triangle, C^{4+} impact; full circle, C^{6+} impact. The errors reflect statistical fitting errors.

also reported that a branching ratio of C_4 emission to C_2 emission is not negligible for the decay of C_{60}^{+}. Hohmann *et al* [7, 20] also reported that the even-numbered cluster emission was an important initial fragmentation process for the photofragmentation of C_{60} by intense pulse laser. Ehlich *et al* [15] reported that the production of fullerene-like fragments was probably due to a combination of cleavage process and successive metastable C_2 loss in the case of very slow collisions between C_{60}^{+} and various gaseous targets.

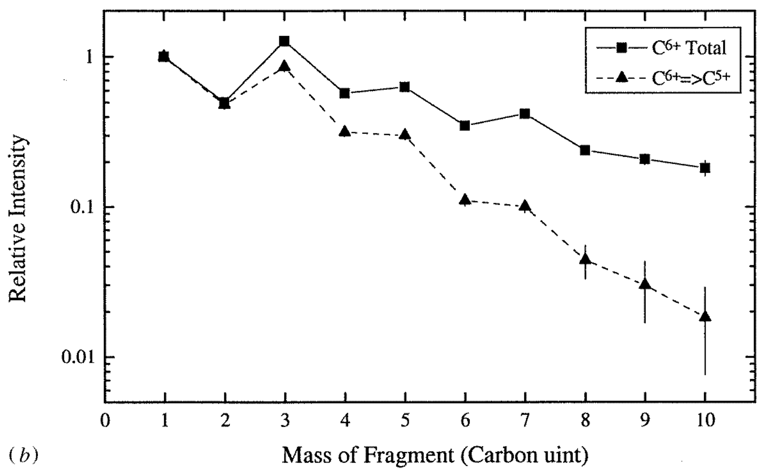
As shown in figure 2(*b*), the intensity ratios between C_{60-2n}^{q+} and C_{60}^{q+}, $I(C_{60-2n}^{q+})/I(C_{60}^{q+})$, seem to be independent of projectile charge states. A similar feature was reported in [7] for the photofragmentation by a nanosecond laser although the laser pulse width was much larger than our collision time scale. They showed that the fullerene-like fragment distribution did not change very much with increasing laser intensity. Because total fragment ion distribution in an ion–molecule collision is obtained by integration over the impact parameter and because fullerene-like fragment ions are produced by rather lower energy transfer as caused by any of our projectiles, there might be little projectile dependence of $I(C_{60-2n}^{q+})/I(C_{60}^{q+})$.

Figure 3(*a*) shows the TOF spectra in coincidence with electron capture by bare projectiles (C^{6+}–C^{5+}) and the projectile K-shell electron loss (C^{5+}–C^{6+}). Fullerene-like fragment ions are completely absent here. A similar TOF spectrum was observed for the electron capture by C^{5+} projectiles. As for the electron loss of C^{4+} projectiles, the peaks of doubly and triply charged fullerene-like fragment ions were observed but their intensities were much lower than those of the small fragment ions. We note that the absence of the fullerene-like fragment ions is not from instrumental origins. The events in which all the channels of multi-hit TDC were hit account for less than 3% of the events of each measurement. In most of the events, not all the channels of our TDC were hit before the fullerene-like fragment ion reached the MCP. As for the detection efficiency of singly charged heavy cluster ions, Walch *et al* [3] reported that the detection efficiency was saturated above 10 kV and that the (relative) detection efficiency for C_{60}^{+} was 36% of the saturated efficiency for the acceleration voltage of 4.2 kV. According to [21], the detection efficiency for singly charged heavy cluster ions increases exponentially with ion velocity at the entrance of MCP in the case of low acceleration voltage. Assuming that the detection efficiency is 36% at 4.2 kV, we could estimate the (relative) detection efficiency at about 30% for the acceleration voltage of 4.0 kV using the exponential formula. We measured yields and pulse-height distributions of the MCP output signals for $C_{60}^{q+}(q = 1, 2, 3)$ over the acceleration voltage from 2.8 to 4.8 kV. Assuming that the efficiency for C_{60}^{q+} depends only on the ion energy at the entrance of the MCP†, we can say that the efficiency of 30% is reasonable and that the detection efficiency appears to be almost saturated above the energy 9 keV. Judging from figure 3(*a*), the TOF peak height for singly charged fullerene ions is thought to be not higher than the fluctuation of the background level. Therefore, after correction for the efficiency, the intensity of each singly charged fullerene ion is estimated to be at most about 5% of the intensities of C^{+}. According to our measurements, the (relative) detection efficiencies for doubly charged fullerene ions is also estimated to be about 90%. In [3], the (relative) detection efficiency for C_{60}^{2+} was estimated to be 81%. Those for triply and more highly charged fullerene ions appear to be almost unity (saturated). Consequently, the intensities of multiply charged fullerene ions are negligible compared with those of small fragment ions. Moreover, we may also rule out the argument that the residual C_{60} systems always dissociate to a neutral heavy fragment and small fragment ions in the overlap collision because the heavy fragment, which has a lower ionization potential than the small fragments, tends to carry more positive charge than the small fragments and because large clusters are generally more stable for Coulomb dissociation than small clusters. Therefore,

† As for secondary electron yield by fullerene impact on surface, Vana *et al* [28] reported no influence of the fullerene charge state on the total electron emission yield by slow fullerene ion impact on gold surface.



(*a*)



(*b*)

Figure 3. (*a*) TOF spectra in coincidence with the electron capture by bare projectiles (C^{6+}–C^{5+}) and the projectile K-shell electron loss (C^{5+}–C^{6+}). The meaning of the number above each peak is the same as figure 2(*a*). (*b*) Relative intensities of small fragment ions up to $n = 10$ are shown for total fragment measurement by C^{6+} impact (full square) and for the electron capture channel of C^{6+} (full triangle). The errors reflect statistical fitting errors.

we can conclude that multifragmentation is the dominant process in the cases of projectile K-shell electron loss and electron capture by projectiles with 1s-vacancies.

In figure 3(*b*), relative intensities of small fragment ions up to $n = 10$ are shown for total fragment measurement by C^{6+} impact and for the electron-capture channel of C^{6+}. The decrease of intensity towards a heavier fragment ion is more rapid for the electron-capture channel than for the total fragment measurement. Similarly, in the case of C^{5+} projectiles, the decrease is more rapid for the channels of electron capture and K-electron loss by the projectile than for the total fragment measurement. It indicates that the intermediate states produced in the charge-changing channels of $C^{5+,6+}$ projectiles are higher excited and/or ionized among the intermediate states followed by multifragmentation in collisions with $C^{5+,6+}$ projectiles.

At our impact energy, the ion velocity, 7.2 au, is so high compared with the velocities of valence electrons of C_{60} that a 1s-electron of C_{60} is predominantly captured by the projectile. In the process of projectile K-electron loss, a 1s-vacancy may be produced in C_{60} at the same time because the probability of projectile K-electron loss is large when projectile ions pass close to a carbon nucleus of C_{60}. Therefore, there is a large possibility of producing a 1s-vacancy inside C_{60} in these processes. Does the 1s-vacancy play an important role in the multifragmentation? Aksela *et al* [8] reported on the product-ion distribution in the photoexcitation and photoionization of the 1s-electron. After the 1s-vacancy is produced by photoexcitation (ionization), it decays through the Auger process with the emissions of one or a few electrons. Singly, doubly or triply charged C_{60} ions are produced and the fullerene-like fragment ions are also produced through rearrangement of the C_{60} ions with valence holes. However, they reported that the intensities of small fragment ions were very low and that the dominant fragment process was a neutral C_2 or C_4 emission following the photoexcitation and photoionization of the 1s-electron. We suppose that the 1s-vacancy production is not directly connected to the multifragmentation but that the 1s-vacancy plays only a role of increasing the charge state of the residual C_{60} system through the Auger decay.

In carbon ion–carbon atom collisions, the probability of projectile K-electron loss is thought to be maximum when the impact parameter is around the K-shell radius of C^{5+} (0.17 au). As for the electron capture from K-shell in C_{60}, the probability becomes maximum at a little larger impact parameter. In such close collisions, the recoil energy of the target carbon nucleus close to the projectile trajectory should be considered. According to a perturbation calculation [23], the probability of C^{5+} electron loss (weighted by $2\pi b$, where b is the impact parameter) has a maximum impact parameter of about $b = 0.2$ au and is larger than half of the maximum value in the region from $b = 0.05$ to 0.4 au. The recoil energy ranges from 0.4 to 25 eV in this impact parameter region and is about 2 eV at $b = 0.2$ au. As estimated in [22], the internal energy of more than 210 eV is necessary for multifragmentation of a neutral C_{60} system. Another experimental study suggests that an internal energy of more than 100 eV is necessary to produce small fragment ions [15]. Therefore, we think that the recoil energy alone can rarely cause multifragmentation even if all of the recoil energy is used for fragmentation. If recoil energy is large enough compared with the binding energy of a carbon atom, a carbon atom (or atomic ion) could be knocked off directly. However, because the binding energy per bond is about 4.8 eV [24, 25], the probability of direct knock-off is thought to be small.

Figure 4 shows the TOF spectra for the L-electron loss cases of the C^{2+} projectile. The peaks corresponding to multiply ionized C_{60} ions and multiply ionized fullerene-like fragment ions become more intense here than in the total fragment distribution by C^{2+} impact, although the corrected intensity of C_{60}^{+} is still larger than the intensities of multiply charged C_{60}^{q+}.

A smaller m/q region becomes more intense in double L-electron loss than single L-electron loss. The intensity ratio of the fullerene-like fragment ions as C_{60-2n}^{q+} to the parent ion C_{60}^{q+}, $I(C_{60-2n}^{q+})/I(C_{60}^{q+})$, is also larger in double L-electron loss than in single L-electron loss, that is, the probability of the fragmentation is larger in double L-electron loss than in single L-electron loss. It indicates that higher excited intermediate states are generated in double L-electron loss than in single L-electron loss. Using the L-electron loss of C^{2+}, we

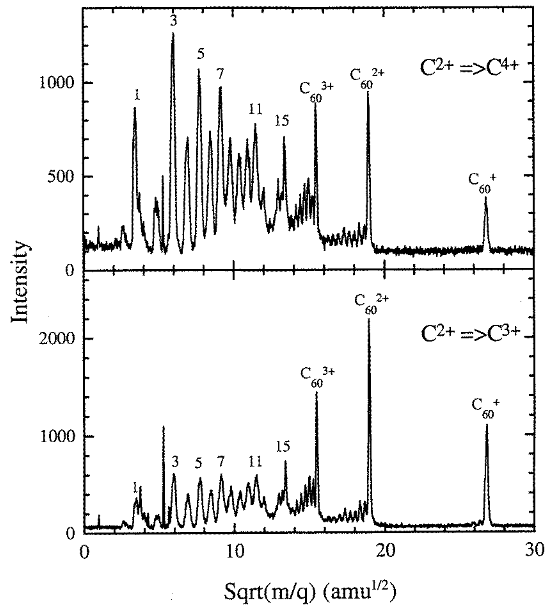


Figure 4. TOF spectra for the L-electron loss cases of the C^{2+} projectile. The meaning of the number above each peak is the same as figure 2(*a*). The peak of $n = 15$ includes the peak of C_{60}^{4+} and we can see the small peak for C_{60-2n}^{4+} as in figure 2(*a*).

can observe the fragmentation via the intermediate states whose internal energy is lower when in close collision with $C^{5+,6+}$ but higher when in the distant collision with C^{2+}.

Moreover, enhancement of small fragment ions for higher-Z (atomic number) projectiles was also observed in our recent experiment where the fragment distributions were measured for various projectiles with low charge states in the MeV region [26]. When in close collision with projectiles carrying many electrons, the screening effect of the projectile nuclear charge seems to decrease and the electrons of C_{60} close to the projectile trajectory feel a stronger force from the projectile ion than the force expected by the projectile charge state. There also seems to be excitation and ionization by projectile electrons, which is known as the antiscreening effect in ion–atom collisions [27]. Therefore, the intermediate states with higher excited energy appear to be generated in close collision with high-Z projectiles than with low-Z projectiles with the same charge state.

From our present measurement, we postulate the fragmentation process in fast collisions as follows. First, direct excitation (including ionization) of valence electrons of C_{60} is caused by projectile ions during collisions. 1s-electrons of C_{60} may be excited or ionized during close collisions. Next, the excited system produced by collisions electronically relaxes. The degree of ionization of the residual C_{60} system becomes higher if further autoionization is possible during the relaxation. Dissociation and/or rearrangement also occurs as a competitive process of the electronic relaxation. Finally, the fragmentation takes place. In charge transfer processes of highly charged fast-projectile ions, where the trajectories of the projectiles overlap with C_{60}, the strong force from projectile ions especially causes high and multiple excitation of valence electrons. If 1s-vacancies are produced in C_{60}, the Auger relaxation of the 1s-vacancy results in an additional ionization of the valence electrons. Moreover, recoil energy of a carbon nucleus of C_{60} near the projectile trajectory increases the internal energy of the intermediate states. In such manner, a multiply ionized and highly excited residual system is created and it decays to multiple small fragments. Further investigations are necessary for the fragmentation, for example, mass correlation, initial kinetic-energy distribution and correlation of fragment ions, and so on.

4. Summary

In summary, we performed selective measurements for close collisions using the coincidence method with the change of projectile-charge states, as well as total fragment-ion distribution measurements. Our total fragment-ion distributions show the peaks of C_{60}^{q+}, C_{60-2n}^{q+} ($q = 1, 2, 3$, and probably 4) and the small cluster C_n^+ up to $n = 15$. For the charge-change channel of $C^{5+,6+}$ projectile ions, the multifragmentation was observed evidently. In these close collisions, the multiple ionization and excitation of valence electrons are important. The influences of 1s-vacancy relaxation and recoil energy of a carbon nucleus in C_{60} are thought to be superimposed upon the excitation of valence electrons. In the L-electron loss of the C^{2+} projectile, the peaks of the multiply ionized C_{60} ions and the multiply ionized fullerene-like fragment ions are more intense or as intense as the small fragment ions. In collisions with various fast-projectile ions, using the coincidence method with the charge-transfer channel, it will be possible to make a rough classification of the intermediate excited states of C_{60} systems and to perform a selective study of relaxation processes from such various intermediate states.

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