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Charge-State Dependence of C_{60} – Multifragmentation in Collisions of 30 MeV Ne^{2-9+} Ions

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Abstract

An experimental study has been made on multifragmentation of free C_{60} molecules using highly charged neon ions at an incident energy of 30 MeV. Production cross sections for fragment ions were obtained systematically at projectile charge states $q = 2 \sim 9$ using time-of-flight coincidence method. Total cross sections for all product ions are found to increase approximately as $q^{1.2}$. On the other hand, cross sections for individual ions exhibit significantly different q-dependence for different ions. As an overall feature, most product ions begin to increase slowly at low q values and then sharply as q^2 for $q \geq 7$. Production of C_1^+ ions is found to be enhanced exhibiting a $q^{2.7}$ dependence for $q \geq 7$.

1. Introduction

Ionization of atoms and molecules by fast charged particles is one of the most fundamental inelastic atomic collisions. It is well known that multiply charged recoil ions are often produced predominantly when a highly charged fast ion collides with a high Z (atomic number) atomic target [1–4]. For polyatomic targets, molecular dissociation is also induced by impact of energetic ions [5,6]. A typical feature of this molecular fragmentation can be seen in collisions involving free C_{60} molecules, in which small-size carbon cluster ions are produced predominantly as a result of multifragmentation [7–12]. From a phenomenological viewpoint, multifragmentation of a C_{60} molecule seems to be analogous to multiple ionization of a heavy atom.

As reported previously [12], production cross sections of fragment ions are significantly smaller than the geometrical size of a C_{60} molecule. Together with other experimental results obtained using slow HCI's [7–10], it is suggested that the C_{60}-multifragmentation is induced only in close collisions in which a large amount of excitation energy may be transferred to the target. Since the total amount of energy deposition depends on the impact parameter between collision partners, it is expected that the charge state of incident particles plays an important role in the multifragmentation process.

2. Experiment

Highly charged Ne^{q+} ions of 1.52 MeV/u ($v = 7.7$a.u.) were produced from a heavy-ion-linear-accelerator at RIKEN (RILAC) with incident charge states $q = 2$, 4, 7, 8 and 9. A well collimated beam was incident on a gas phase C_{60} target in a crossed beam collision chamber. The C_{60} target was formed by heating a high purity powder at about 500°C using a temperature controlled oven. Time-of-flight coincidence measurement of fragment ions was carried out by chopping the incident neon beam to 145 ns width and 50 μs period [11]. Detection of fragment ions was made with a chevron type multichannel plate (MCP) at −5kV front voltage. Care was taken for determination of MCP detection efficiency for secondary ions with different mass. The incident beam current was measured with a biased Faraday cup located downstream of the collision chamber. The base pressure was kept below 2×10^{-8} Torr through the whole experiment. In order to minimize experimental errors arising from uncertainties of the beam current, the total yield of secondary ions was measured simultaneously. As the C_{60} target pressure was kept constant during measurements, the spectral intensity of a given fragment ion divided by the number of incident particles gives a relative production cross section of the fragment ion. The charge state dependence of production cross sections deduced in this way was determined with an accuracy better than 10%.

3. Results and discussion

Relative cross sections for the production of individual fragment ions C_n^+ ($n = 1 \sim 12$) and intact ionized parent ions C_{60}^+ ($r = 1 \sim 4$) are presented in Fig. 1, where correction due to the MCP detection efficiency is made for parent ions. Even-odd oscillations are observed for all the incident charge states q. With increasing q, however, the oscillation behavior becomes weaker and approaches power-law intensity distributions of the form of $n^{-\alpha}$, indicating that the multifragmentation is induced more preferentially as the projectile charge state increases. It should be noted that the experiment was made by the beam-chopping coincidence method, so that the present data include contributions from all impact parameter collisions. This is essentially different compared to our charge-changing coincidence experiments using 2 MeV $Li^{0\sim3+}$ ions, where the power-law behavior becomes remarkable as the initial-final charge difference increases. That is, charge-changing collisions are limited to small impact parameters.

As for the intact parent ions C_{60}^{r+}, the ionization cross sections exhibit also a power-law form $r^{-\alpha}$ as a function of the degree of ionization r. Estimated values of powers are 2.1 ($q = 2$), 2.7 ($q = 4$) and 2.9 ($q = 7 \sim 9$). It is interesting to point out that these values are nearly equivalent to those obtained in 1.05 MeV/u Ar^{q+} + Ar collisions

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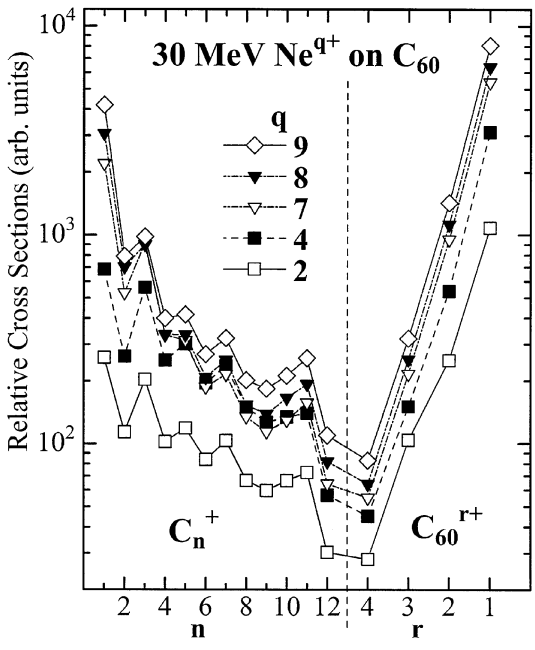

Fig. 1. Cross sections for the production of fragment ions C_n^+ and intact ions C_{60}^{r+} as a function of n or r.

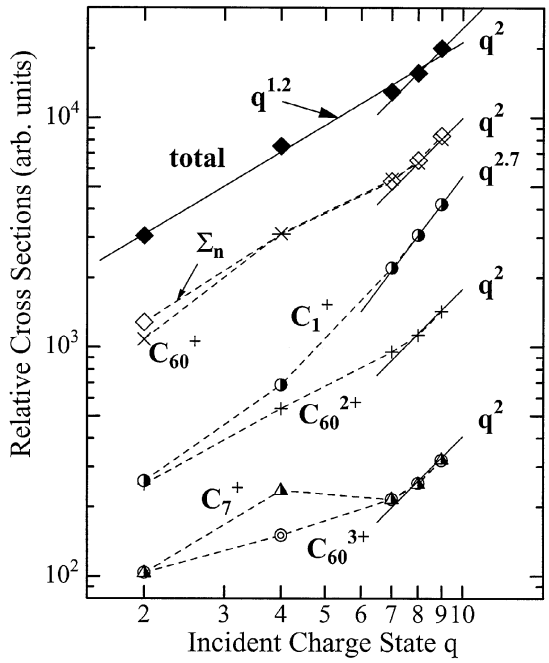

Fig. 2. Cross sections for various product ions as a function of the projectile charge state q. The symbol Σ_n represents the sum of cross sections over small fragment ions C_n^+ of $n = 1 \sim 12$, and "total" is the total cross sections of all product ions.

[3], indicating a similarity of C_{60} to a multielectron single atom.

The incident charge state dependence of the production cross sections are shown in Fig. 2 for some typical product ions. Here, the values denoted by Σ_n are the sums of cross sections over small fragment ions C_n^+ from $n = 1$ to 12, and "total" represents total production cross sections of all the produced ions including parent and their daughter ions. The "total" cross sections increase as $q^{1.2}$ as shown in the figure. An interesting result is that the single ionization production cross sections (C_{60}^+) are almost equivalent to those of total small fragment ions Σ_n at all the incident charges.

One can see clearly that, for most fragment ions, the cross section variation as a function of q cannot be expressed in a simple power-law form of q^β over the whole range of q. Instead, the cross sections increase slowly in the low q range and then increase steeper with β of about 2 in the higher q range. A careful analysis shows that the values of β in the low q range ($q \leq 7$) are different for different ions. As for intact parent ions C_{60}^{r+}, it was found that the values of β for $q \leq 7$ become smaller with increasing r. This result apparently contradicts the tendency observed in usual ion-atom collisions where multiple ionization has steeper q-dependence compared to single ionization [1,2]. It seems that the multiple ionization of C_{60} can not be described on the basis of the independent electron model [1]. Furthermore, it also indicates that multiply ionized parent ions may decay into small fragment ions with high probabilities as the value of r increases, leading to weaker q-dependence of the final production yields of these parent ions. It should be noted that the lightest ion C_1^+ increase as $q^{1.4}$ for $q = 2 \sim 4$ and $q^{2.7}$ for $q \geq 7$. These large values seem to support the assumption described above.

Different from light ions like $C_1^+ \sim C_3^+$, production yields of intermediate fragment ions were found to be nearly constant in the range $q = 4 \sim 7$, as demonstrated for C_7^+ in Fig. 2. This fact indicates clearly the existence of a competing process that reduces the production of these ions. In order to see more clearly this somewhat surprising q-dependence, intensity ratios between C_n^+ and Σ_n are plotted in Fig. 3. One can see obviously that the larger fragments ($n > 5$) tend to decrease at $q = 4$ and stay nearly constant for $q \geq 7$. On the other hand, C_1^+ begins to increase sharply at $q = 4$, and an intermediate behavior is found for ions of $n = 2$ and 3. These results correspond to the power-law distri-

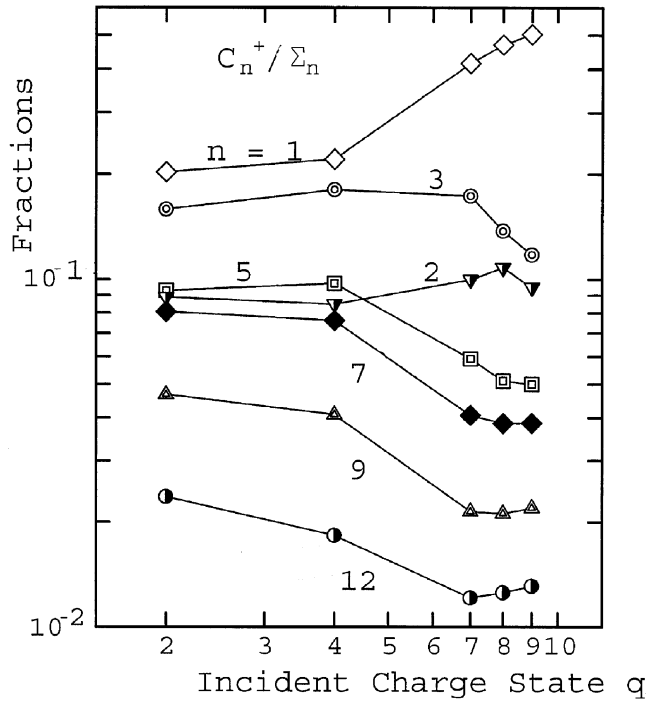

Fig. 3. Intensity ratios between C_n^+ and Σ_n as a function of q.

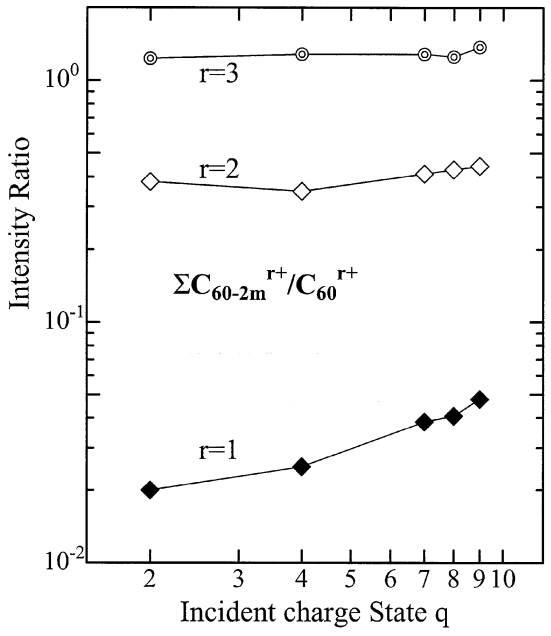

Fig. 4. Intensity ratios between the sum of daughter ions ΣC_{60-2m}^{r+} and intact ions C_{60}^{r+} at a given recoil charge state r.

bution $n^{-\alpha}$ discussed above (Fig. 1), implying the preferential enhancement of multifragmentation at large q values. This leads, consequently, to less production yields of intermediate fragment ions in the final distribution spectra.

Contrary to small fragment ions described above, large daughter ions C_{60-2m}^{r+} originate from relatively soft relaxation processes of initial excited C_{60} molecules [9]. Thus, the production cross sections of large daughter ions give a good indication of the degree of internal excitation. Figure 4 shows intensity ratios between total intensity of daughter ions and intact parent ions with the same charge state r. It is noted that a part of the daughter ions with charge r ($r > 2$) are originating from parent ions with higher charge in a decay scheme such as $C_{60}^{4+} \rightarrow C_{60-2m}^{3+} + C_{2m}^{+}$ [10]. One can see clearly that the degree of internal excitation of intact ions increases with increasing r. Another interesting characteristic is that the ratios for $r = 1$ increase with increasing projectile charge, while for $r = 2$ and 3 the values are nearly constant irrespective of q, implying apparently that the daughter ions have the same q-dependence as their parent ions. Together with the previous results described in Fig. 2, these results also suggest that the multiply charged parent ions ($r \geq 2$) are apt to decay into small fragments as the incident charge increases.

In conclusion, a strong q-dependence was observed for the production of C_1^+ ions in comparison with other fragment ions. We conclude that this result is another direct proof of the preferential multifragmentation taking place at higher incident charge states.

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