Effects of collective excitation on ionization and fragmentation of C_{60} by high energy H^+ impacts

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The ionization and fragmentation cross sections of C_{60} for 0.2–2.0 MeV H^+ impacts have been measured. The results obtained show that the peak position of the ionization cross sections shifts to lower impact energies with increasing the charge state of C_{60}^{q+}. The theoretical calculations based on single and double plasmon excitation model were carried out, supporting the present experimental results. In addition, it is found that the cross sections for parent ions C_{60}^{q+} and fragment ions $C_{60-2m}^{(q-1)+}$ (q = 2–4, m = 1, 2) have the same energy dependence, while the energy dependence of cross sections for these ions at $m \geq 3$ differ with increasing the number of emitted C_2. The production of C_{58}^+ is interpreted fairly well within the double plasmon excitation.

Introduction

Studies of ionization and fragmentation dynamics of C_{60} have been conducted using various means such as charged particle (ions, electrons and C_{60} ions) impacts or photoabsorption.[1-5] Clarification of their detailed collision mechanisms are essential for understanding the electronic properties and stability of highly excited states of C_{60}. Recently, there has been considerable interest in a giant plasmon resonance of C_{60}.[2] This resonance occurs at around 20 eV with a width of about 10 eV, and may play an important role in the ionization and fragmentation processes of C_{60}.

Pioneering experimental studies of ionization and multifragmentation have been performed by LeBrun et al.[3] using 420–625 MeV $Xe^{18+,35+}$. Single ionization cross sections have been reproduced fairly well by the theoretical calculations using the first Born approximation on the basis of a single plasmon excitation model. In multiple photoabsorption, on the other hand, remarkable experimental results have been reported by Hunsche et al.,[4] who conclude that double ionization or singly charged C_{60-2m}^+ (m = 1–9) require a double plasmon excitation at 40 eV.

The purpose of this work is to provide cross section data relevant to ionization and fragmentation mechanisms by observation of cross sections, and to verify the applicability of the plasmon excitation model for multiple ionization cross sections in fast H^+-C_{60} collisions. The correlation between ionization and fragmentation processes is discussed.

Experimental method

We have measured ionization and fragmentation cross sections of C_{60} for 0.2–2.0 MeV H^+ impacts under the single collision condition. The H^+ beam was obtained from a 1.7 MV tandem Cockcroft-Walton accelerator at Kyoto University, and collimated by two adjustable slits to about 2×2 mm^2 before colliding with a target. The incident DC beam was converted into a pulsed beam using an electrostatic beam chopping system. The C_{60} vapor target was produced by heating high-purity (99.98%) C_{60} powder to 450 °C in a quartz oven. Fragment ion distribution was measured using a spatial focusing time-of-flight spectrometer. The target pressure was estimated to be about 8×10^9 /cm^3 in the collision region. To obtain MCP detection efficiency, we measured the relative detection efficiency (RDE) of C_{60}^{q+} ions. At the MCP front bias of 4.9 kV, RDEs of C_{60}^{q+} ions were about 45% (q = 1), 89% (q = 2), and unity (q = 3, 4), respectively.

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Results and discussions

Our spectra show that the peaks corresponding to parent ions C_{60}^{q+} and fragment ions C_{60-2m}^{q+} originate from even numbered neutral cluster emission. The charge state of these ions is clearly resolved up to q = 4. The results are similar to those obtained in electron impact experiments.[5] However, the present results are completely different from those obtained in heavy ion impact[1,3] or multiple photoabsorption experiments.[4] That is, no small fragments (C_n^+) were obtained in the present spectra, while they distinctly appear in the latter experiments.

Figure 1 shows the experimental results for the ionization cross sections for 0.2–2.0 MeV H^+ impacts on C_{60}, and for electron impacts by Dünser et al.[5] The solid lines in Fig. 1 are the results of theoretical calculations based on single and double-plasmon excitation models. Details regarding these models are given in Ref. 3. It should be noted that the peak position of the cross sections clearly shifts to lower impact energies with an increase in the charge state of C_{60}^{q+}. Obviously the present results are different from the general behavior observed in ion-atom collisions, namely, the peak position of the cross section shifts to higher impact energies with an increase in the ionization potential.

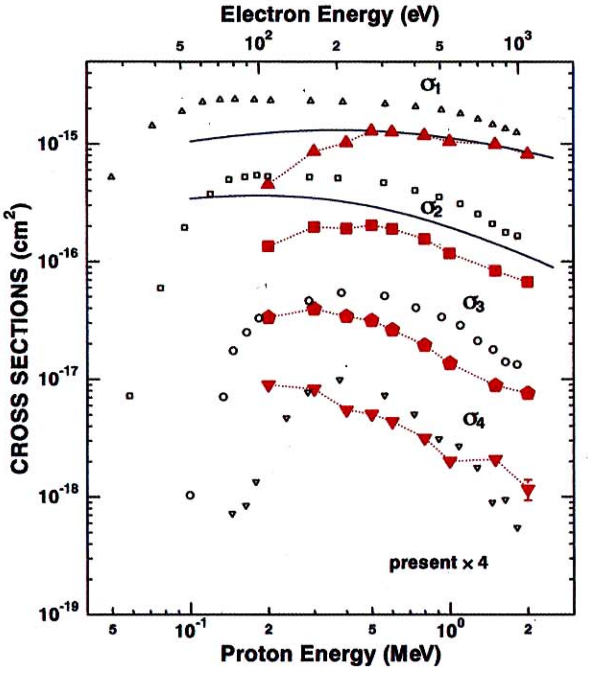

Fig. 1. Single to quadruple ionization cross sections of C_{60} for H^+ impacts (solid symbols). The open symbols are measurements for electron impacts (Ref. 5). The lines represent the values calculated using the plasmon excitation model.

The calculated cross sections for single and double plasmon excitation are compared. The calculated values show a good agreement with the experimental results to within a factor of about 4. In Fig. 1, the calculated cross sections were normalized to the measured single ionization cross sections at 0.6 MeV. This discrepancy may be attributed to the following: the first is the uncertainty of target pressure measurements, and second is due to the ambiguity of detection efficiency measurements for a detector. Finally, the calculation is based on the assumption that plasmon excitation relaxes only when decay occurs via emission of electrons (ionization). The peak energies of σ_1 and σ_2 are at about 0.45 and 0.25 MeV, and the results supported qualitatively, the present experimental results. Thus, it can be concluded that single and double ionization of C_{60} is induced predominantly via plasmon excitation. In addition, it should be noted that the simple model can be applied to explain the ionization processes of C_{60} by fast particles such as H^+ and Xe^{35+}.

Regarding proton and electron impacts, the ionization cross sections for proton impacts are about a factor of 6 smaller than those for electron impacts. However, both cross sections appear to be identical in the high energy region. On the other hand, the energy dependence of σ_1 and σ_2 are very different, for e.g., σ_1 and σ_2 for electron impacts have a broad cross section maximum showing a flat plateau, but not so for proton impacts. This behavior is also observed in electron-C_{60} ion collisions. The plateau in electron impact experiments can be explained by the effects of factors such as distortion, electron exchange and resonance channel between electron and C_{60}. On the other hand, σ_3 and σ_4 for both proton and electron impacts have a similar energy dependence. Hence, it is suggested that the triple and quadruple ionization are occur via the same specific excitation channels.

Figure 2 shows the ratio of cross sections for parent ions C_{60}^{q+} and fragment ions $C_{60-2m}^{(q-1)+}$ ($q = 2$–4). The two remarkable features are (a) the results indicate that the cross sections for $C_{58,56}^{(q-1)+}$ and C_{60}^{q+} ($q = 2$–4) have a similar energy dependence, and (b) the ratio shows a minimum at about 0.6 MeV with an increase in the number of emitted C_2. These results indicate that C_{60}^{q+} and $C_{58,56}^{(q-1)+}$ ($q = 2$–4) ions were produced via the same intermediate state, while it must be emphasized that the production mechanisms of C_{60-2m}^{q+} ($q \geq 3$)differ depending on the number of C_2 emitted. In (a), it is suggested that C_{60}^{2+} and C_{58}^{+} ions are produced by double plasmon excitation, via two pathways, double ionization and emission of both one electron and C_2. In multiphoton absorption experiments, Hunsche et al.[4] have reported that the production of both C_{60}^{2+} and C_{60-2m}^{+} ($m = 1$–9) ions requires a double plasmon excitation at 40 eV. In the present results, the energy dependence of the cross sections for C_{56}^{+} are clearly different from that for C_{60}^{2+}. Hence, further studies to elucidate the production mechanism for C_{56}^{+} are required.

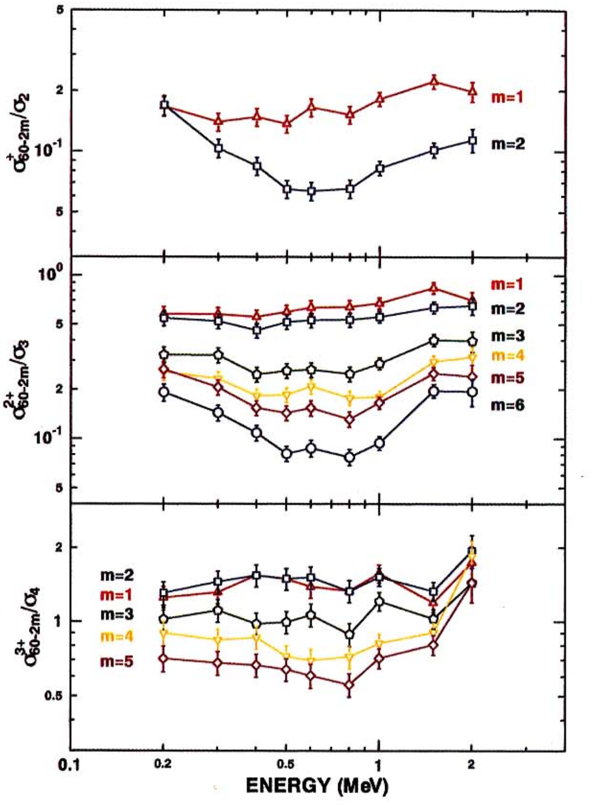

Fig. 2. The ratio of cross sections for parent ions C_{60}^{q+} and fragment ions $C_{60-2m}^{(q-1)+}$ as a function of incident energies.

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