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Effect of giant plasmon excitations in single and double ionization of C_{60} in fast heavy-ion collisions

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Single and multiple ionization of C_{60} in collisions with highly charged fast oxygen ions have been studied using the recoil-ion time-of-flight technique. The dependence of multiple-ionization cross sections on projectile charge state (q_p) was found to be drastically different from those for an atomic target, such as Ne. A model based on the giant dipole plasmon resonance explains quite well the observed q_p dependence for the single-and-double-ionization cross sections. But the same model deviates for triple and quadruple ionizations.

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Ion-atom collisions involving single-particle interactions with heavy ions have been a subject of study for a long time. But for multielectron interactions the studies performed up to now offer little insight. In some cases, like nanoclusters, the actual behavior of the system may be quite simple, even when the underlying equations are not [1]. A similar situation arises when a sufficiently large number of electrons collectively and coherently respond to photons, electrons, or fast-moving heavy ions [1]. Collective excitation plays a very important role in the electronic energy-loss mechanism when a charged particle interacts with a solid or cluster target [1,2]. In spite of this, very few studies have been made to understand this aspect of excitation in heavy-ion collisions. So far most of the studies on collective excitation of the C_{60} fullerenes or metal clusters involve the photoabsorption technique. The collective response of the target electrons has been observed in ion-solid [3] and ion-cluster [4] collisions. Studies on the ionization and fragmentation of fullerenes induced by highly charged ions or protons have been carried out by several groups [5–12]. The electron capture process has been investigated for low-energy ions colliding with C_{60} [13] and the classical over-the-barrier model has been used to explain the data. However, the dynamics of atomic processes in low-energy collisions differ widely from those at high energies [4,6,14] and out of these references only a few [4,6] deal with the collective excitation phenomenon.

C_{60} offers a very simple target with a solidlike electron density distribution coming from the delocalized outer-shell electrons. It is known to have a giant dipole plasmon resonance (GDPR) at $\sim$20 eV [15] having a width of about 10 eV and integrated oscillator strength 71 [1,15] (also see [16]). It was shown in an intense-laser-based experiment that the ionization of C_{60} is dominated by the GDPR excitation [17]. Cheng *et al.* [6] proposed a simplistic model for the fast-ion-induced GDPR followed by ionization. Though it provided a reasonable guideline to the experimental data, a rigorous test is awaited. The model was further tested to some extent by Tsuchida *et al.* [8] for proton beams with different velocities. The model predicts a weak velocity dependence but very strong projectile charge state (q_p) dependence, which would be a more stringent test of the model. A collective excitation causing a wakefield is shown [4] to influence the x-ray emission following electron capture in fullerenes. The influence of this process on single and multiple ionization is not very well understood. We investigate the q_p dependence of multiple ionization of C_{60} and a Ne gas target. The relative ionization cross sections for C_{60} in collisions with 50 MeV oxygen ions with different charge states are measured. The projectile velocity (v_p) was high enough (10.8 a.u.) so that the total electron capture probability can be considered small.

A Wiley-McLaren-type time-of-flight (TOF) recoil-ion mass spectrometer with the mass resolution $M/\Delta M=130$ has been used. The electric fields in the extraction and drift regions were 330 and 650 V/cm, respectively. A slit placed in front of the electron channel electron multiplier (CEM) was used to reduce the electron count rate. The base pressure in the chamber was better than 1×10^{-7} mbar. A well-collimated (1×1 mm^2) beam of 50 MeV O ions with charge states q_p=4+ to 8+ was provided by the BARC-TIFR Pelletron facility at TIFR. The beam was made to interact with the C_{60} vapor from the oven. In the case of the atomic target experiment, an effusive jet of Ne gas was used. The electron signal from the electron CEM was used as the start trigger and the recoil-ion signal from the microchannel plate (MCP) detector was used as the stop.

The intrinsic efficiency of the MCP was obtained using an empirical formula [14]. The collection efficiency for the C_{60} ions is unity. The temperature dependence was measured by taking the data at different oven temperatures in some cases. All the data sets were collected at approximately constant oven temperature of 400 °C. The areas under the C_{60} recoil-ion peaks, corrected for the detector efficiency and temperature variation and normalized with the integrated beam current, give the relative cross sections for single and multiple ionization. The cross sections had standard deviation $\sim$20% arising mainly due to the uncertainty in C_{60} vapor density [5,9].

To study the effect of the GDPR in ionization of C_{60}, LeBrun *et al.* [6,7] used a model based on linear response theory and the dipole approximation which was originally developed for nuclear GDR studies [18]. We use the same model here. The electrons are treated as a free-electron gas and the GDPR is described as an excitation of a harmonic oscillator with characteristic frequency ω ($E=\hbar\omega=20$ eV

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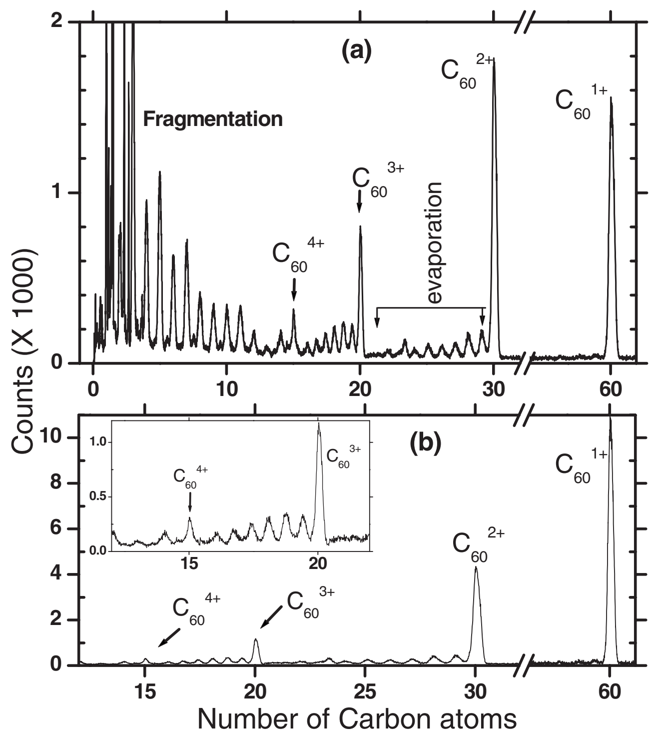

FIG. 1. (a) Typical TOF spectrum of C_{60} in collision with 50 MeV O^{8+} ions. (b) Same spectrum with efficiency correction. Inset shows $C_{60}{}^{3+}$ and $C_{60}{}^{4+}$.

here). The required energy transfer $\Delta\epsilon$ to excite the GDPR, is then calculated [6,8]. The GDPR is characterized by an oscillator strength distribution $f(E)$ which is approximated to be a Gaussian. The effective number of excitations of the oscillator is given as

$$N(b) = \int_0^\infty dE \frac{f(E)}{E} \Delta\epsilon, \qquad (1)$$

and the probability for excitation to the nth state is given by

$$P_n(b) = \frac{[N(b)]^n}{n!} \exp[-N(b)]. \qquad (2)$$

Finally the cross section of excitation is given by $\sigma_n(pl) = 2\pi \int b\, db\, P_n(b)$.

A typical TOF spectrum is shown in Fig. 1 in which C_{60} ions with recoil-ion charge states q_R from 1+ to 4+ are clearly visible. The ionized products are accompanied by C_2 evaporation products which were negligible for single ionization. The fraction of all the C_2 evaporation products together was found to be significant compared to $C_{60}{}^{2+}$ yields ($\sim 25\%$) or $C_{60}{}^{3+}$ yields ($\sim 50\%$). The similarity of the spectrum with that for low-energy collisions [14] is noticeable.

Since the single- and double-plasmon-excitation energies are about 20 and 40 eV, respectively, it is energetically not possible to have evaporation due to plasmon excitation. The evaporation of C_2 fragment from $C_{60}{}^{2+}$ ions needs at least 53.9 eV of (appearance) energy deposition [20,21] although the C_2 binding energy is ~ 10 eV. Therefore, even for double plasmon excitation C_2 evaporation is very unlikely. Evaporation from $C_{60}{}^{3+}$ requires an appearance energy of ~ 70 eV. The internal energy required for evaporation and fragmentation is relatively large and can be supplied in a very close collision. Figures 2(a)–2(d) show the q_R distributions of the relative ionization cross sections (σ_I, not including evaporation products) for different q_p=4+, 5+, 6+, and 8+. The model prediction [normalized to the experimental single ionization cross section (σ_{SI})] agrees well with the double-ionization cross sections (σ_{DI}). A similar trend is also observed for q_p=5+, 6+, and 8+. Therefore, the ratio of single- to double-ionization cross sections is approximately equivalent to the ratio of single- to double-plasmon resonance cross sections.

This is in partial agreement with the observation using an intense laser [17] which concludes that single and double plasmon excitations lead to single and double ionization, respectively. Accordingly, we compare the calculated plasmon-

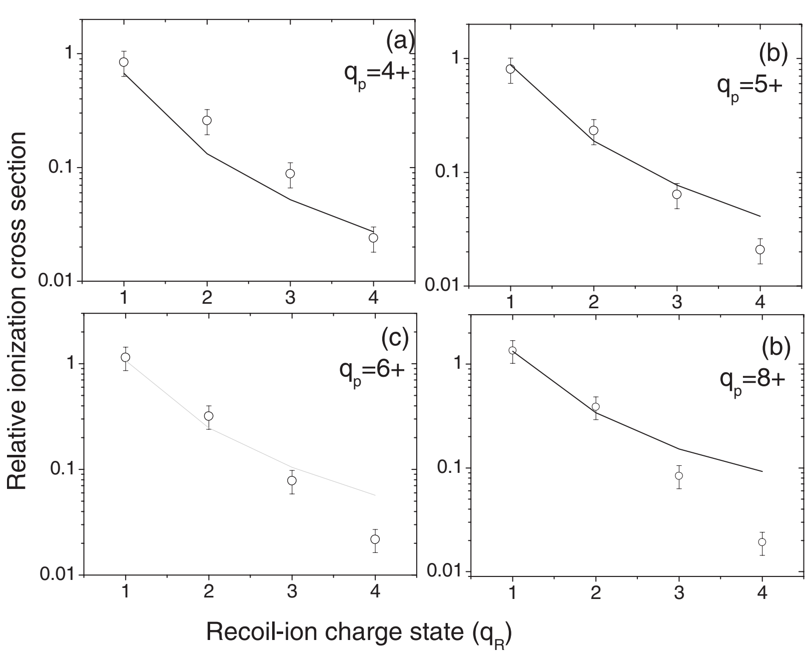

FIG. 2. Relative ionization cross sections of C_{60} versus q_R for q_p=4+ (a), 5+ (b), 6+ (c), and 8+ (d). The lines are the normalized GDPR model predictions.

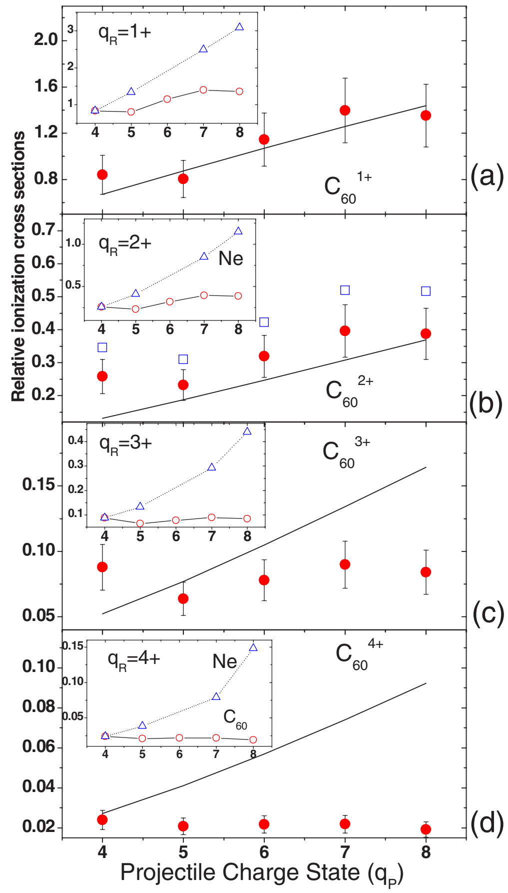

FIG. 3. (Color online) Relative cross sections for C_{60}: (a) single ionization, (b) double ionization, (c) q_R=3+, and (d) q_R=4+. Insets show the cross sections for Ne (△) and C_{60} (○). Ne data are normalized to C_{60} data at q_p=4+. The squares in (b) represent the yields of double ionization + associated evaporations.

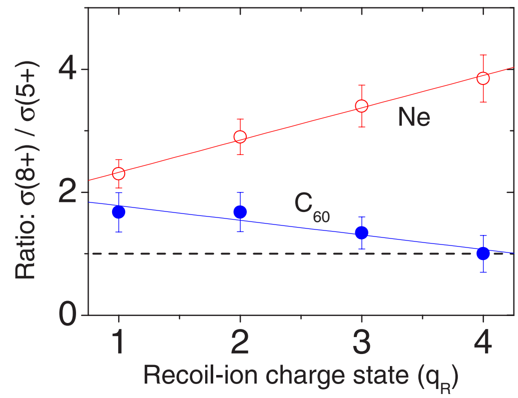

FIG. 4. (Color online) Ratio of ionization cross section for 8+ to that for 5+ for C_{60} as well as Ne.

excitation cross sections to those for the ionization. However, this assumption is not an absolute requirement here since we compare only the relative cross sections with the model, although the ratios are comparable as shown above.

A more stringent test of the model would be to compare the q_p dependence of σ_I. The single-, double-, triple- (σ_{TI}) and quadruple- (σ_{QI}) ionization cross sections (i.e., not including evaporation products) versus q_p are shown in Figs. 3(a)–3(d) along with the respective GDPR predictions (normalized to one point). The single plasmon excitation cross sections agree very well with the σ_{SI} data [Fig. 3(a)]. For double ionization the theory still agrees with the trend shown by the experiments [σ_{DI}, Fig. 3(b)]. For the sake of completeness, the squares [shown in Fig. 3(b)] represent the evaporation contribution added to the double ionization and it is obvious that the slope of the variation is similar to that for double ionization. For the triple and quadruple ionizations the model predicts steep q_p dependence, whereas the data show nearly q_p independent, behavior. This indicates that single and double ionization are influenced largely through the excitation of the giant plasmon mode. One expects σ_{SI} to vary as q_p^2 according to the Bohr-Lindhard [Eq. (8) in Ref. [23]] or quantum-mechanical first-order models. However, we compare these observations with the experimental results for a pure ion-atom collision system, such as that for Ne atom, and plot the ionization cross sections versus q_p in the insets. It can be seen from Fig. 3 that σ_I increase either almost linearly (for single and double ionization) or faster (for higher ionizations). In single and double ionization of C_{60}, σ_{SI} and σ_{DI}, respectively, increase linearly but with much smaller slopes than those for the Ne target. For example, in the case of single ionization of C_{60} the observed slope (∼0.15) is only about one-fourth of that (∼0.60) for Ne. The result for double ionization is similar. This indicates that in such ionization the molecule is acting like a single system although the slopes are much lower than those expected for single ion-atom collisions. On the other hand, the slopes are almost entirely governed by the GDPR prediction for C_{60} (q_R=1,2) as shown in Figs. 3(a) and 3(b).

For H-like and bare ions there is a substantial increase in the Ne recoil-ion cross sections for q_R=3+ and 4+ which is partly due to *K* (shell)–*K* (shell) electron transfer. A similar *K*-*K* transfer contribution is also expected for the O-C collision system. However, for C_{60} the cross sections do not show any such enhancement. Such close collisions most likely would also lead to fragmentation (through nuclear stopping) in the fullerene target and therefore the *K*-*K* transfer is not observed.

The increase in σ_I with q_p can also be quantified by plotting the ratio of σ_I for q_p=8+ to that for 5+ (i.e., the lowest charge states obtained using a post stripper). This ratio (Fig. 4) gradually increases for Ne with q_R whereas for C_{60} it decreases, approaching 1.0. These behaviors are opposite to each other and cannot be understood based on a pure ion-atom collision model. For triple ionization the cross sections show [Fig. 3(c)] a very weak q_p dependence and the slope of the dependence is much less than that for an atomic target or for the GDPR prediction. For quadruple ionization of C_{60} [Fig. 3(d)] the cross section is independent of q_p. For Ne, however, a much steeper increase is seen for both the triple and quadruple ionizations. The slope of the ionization cross sections increases with q_R for Ne whereas for C_{60} this behavior is opposite, i.e., the q_p dependence is gradually reduced with higher ionization. The exact reason for this behavior is not understood.

The higher degrees of ionization, such as q_R=3+, and 4 +, are mainly produced in close collisions for which evaporation and fragmentation and solidlike dynamical screening [24] are also possible processes, if the collision takes place inside the cage. For single ionization the evaporation product is negligible. In the case of double ionization the evaporation yield is about 25% and is almost independent of q_p. In the present case, the TOF ($\leqslant 10\ \mu$s) is less than that in Refs. [19,20]. Since evaporation is a statistical process, the decay of the metastable parent ions depends on the internal energy and the flight time. On a longer time scale, a small fraction of the main ionization peak can still be contributed by the ions with sufficient internal energy (for evaporation) which did not decay by evaporation during the flight. This contribution can be shown to be small using the Maxwell-Boltzmann distribution. This fraction, being independent of q_p, will not change the slope of the yields and thereby the agreement with the model remains valid. For the triple and quadruple ionizations the evaporation yield is higher (50% or more) and again a part of the ionization peak can be contributed by this process. For triple and quadruple ionizations the GDPR model does not explain the observation anyway. For these ionizations the excitation energy is very large and the C_{60} is fragmented apart from ionization+evaporation. It is shown that for $q_R \geqslant 3$ superasymmetric fission competes with evaporation [21,22] and therefore one has to consider all these channels.

One may gain some insight by invoking the observations in ion-solid collision processes. In ion-solid collisions an equilibrium charge state is reached within a thickness of about a few tens of μg/cm^2, which is measured in terms of postcollisional q_p. The dynamical screening [24] of the swift ions and wakefield-induced effects are also known in ion-solid interactions [3] as well as in ion-fullerene collisions [4]. In close collisions with the molecular shells (inside the cage) the projectile may be subjected to dynamical screening [4] (with wavelengths ~ tens of angstroms) in a highly polarizable molecule like C_{60}. However, one cannot expect a equilibration to take place within the C_{60} molecule. Since in ion-sold collision experiments much thicker targets are used, the q_p distributions are the results of such microscopic distributions arising from many successive layers. In this sense, collision studies with fullerene may provide microscopic input to develop a more detailed model for collisions in solids. However, a self-consistent model, inclusive of evaporation, fragmentation, ionization, and dynamical screening, needs to be developed. Analysis of the evaporation and fragmentation yields is in progress and details will be presented elsewhere. The present study is limited to a narrow range of q_p which will be extended to heavier projectiles with higher q_p in order to have a deeper insight into the problem.

In conclusion, heavy-ion impact ionization of C_{60} molecules is studied in fast collisions. A model based on the GDPR has been used to explain the observed q_p dependence of the ionization cross section of C_{60}. The single and double ionization are largely dominated by the excitation of single or double plasmons, respectively. However, for triple and quadruple ionization the model fails to reproduce the observed q_p independence. Single and multiple ionizations of C_{60} show considerable difference from the results with an atomic target, such as Ne. A theoretical model possibly including ionization, evaporation, fragmentation, and solidlike dynamical screening mechanism may be required to explain the multiple ionization of C_{60}.

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