Effect of Collective Response on Electron Capture and Excitation in Collisions of Highly Charged Ions with Fullerenes

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Projectile deexcitation Lyman x-ray emission following electron capture and K excitation has been studied in collisions of bare and Li-like sulphur ions (of energy 110 MeV) with fullerenes (C_{60}/C_{70}) and different gaseous targets. The intensity ratios of different Lyman x-ray lines in collisions with fullerenes are found to be substantially lower than those for the gas targets, both for capture and excitation. This has been explained in terms of a model based on "solidlike" effect, namely, wakefield induced stark mixing of the excited states populated via electron capture or K excitation: a collective phenomenon of plasmon excitation in the fullerenes under the influence of heavy, highly charged ions.

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The discovery of stable C_{60} and other fullerenes has led to a series of intensive studies on the structural and collisional properties of these symmetric quasispherical carbon clusters. The studies of the fragmentation and ionization of C_{60} in collisions with electrons or photons have received a great deal of attention due to their fundamental and practical importance in many areas of physics, chemistry, astrophysics, and material science. The collective excitation phenomenon, namely, the excitation of a giant plasmon dipole resonance, with an energy of $\sim$20–30 eV has been predicted [1] and observed experimentally [2,3]. The collective excitation as well as other properties of fullerenes or other types of clusters are of current interest in the physics of small particles since these studies are aimed to bridge the gap between solids and atoms.

The heavy ion atomic collision technique has been proven to be a powerful tool to study the structural and collisional properties of atoms, molecules, solids, and surfaces (see [4]). There have been many measurements on the ionization and fragmentations of fullerenes using low energy highly charged ions (HCI) or protons [5–8] as projectiles. The formation of hollow ions in collisions of such low energy HCIs with C_{60} and surfaces [4,9] is also known. The dynamics of different atomic processes in low energy collisions differ widely from those at high energies [10–12]. The potential importance of giant plasmon excitation in high energy heavy ion collisions was also noted [11]. The giant dipole resonance is a well-known phenomenon in nuclear reactions. In ion-solid collisions, the collective electronic response is known to give rise to a dynamic screening and a wake of electron density fluctuations trailing the ions [13,14]. Such a solid-state effect on REC (radiative electron capture) [15,16], resonant coherent excitation [17,18], and convoy electron production (see [13]) has been identified. The Stark splitting due to the wakefield has also been observed in the Lyα x-ray emission yields in e-capture by bare ions in thin solid targets [19,20]. A large enhancement in the multiple e-capture has been noticed in thin carbon foils in high energy collisions [21]. The last two measurements [19,21] are carried out at sufficiently high energies, i.e., 36 and 46 MeV/u, and therefore satisfy a single collision condition for such thin ($\sim$10 μg/cm^2) targets. However, the single collision does not necessarily hold for a lower energy heavy ion ($\sim$5 MeV/u) colliding with thin foils since the mean-free paths for electron capture become comparable with the foil thickness (see, e.g., Refs. [19,22]). At such collision energies, free C_{60}/C_{70} molecules become an excellent tool to investigate the "solidlike" effect. The fullerenes are "small" particles with diameter $\sim$10 Å over which a large electron density [23], similar to that of a solid foil, is sampled, while single collision conditions are satisfied. It has also been shown recently [10] that the charge state distribution of low energy Li ions colliding with fullerenes is almost similar to that of thin carbon foils. The present investigation is aimed to explore the effect of collective response in heavy ions colliding with fullerenes. We have measured the Ly x-ray intensities arising due to e-capture in the excited states of bare S ions in collisions with fullerenes and other gaseous targets. To study the K-shell excitation, Li-like ions were used (due to the existence of long lived metastable states He-like ions were not used).

Bare and Li-like S ions of energy 110 MeV were obtained from a 14 MV BARC-TIFR Pelletron accelerator. A tightly collimated ion beam of diameter $\sim$2 mm interacts with the fullerene vapor at a distance of 6 mm from the nozzle of length 8 mm and opening 1 mm. The vapor target was formed from a mixture of C_{60}/C_{70} (76% C_{60}, 22% C_{70}, 2% higher fullerenes) [24], heated to about 550–600 °C in an oven. The typical effective target thickness was calculated to be 2×10^{10} per cm^2. For collisions with gas targets, the chamber was flooded with He, N_2, Ne, Ar, Kr, and Xe gases, and single collision conditions were ensured. A Si(Li) detector (resolution 160 eV at 5.9 keV) with 25 μm Be window and 6 μm mylar foil,

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mounted inside the vacuum chamber, at 90° with respect to the ion-beam direction was used to detect the x-rays emitted. Two other Si(Li) detectors were also mounted at 45° and 135°. The experiment was repeated for two other beam energies, i.e., 120 and 138 MeV.

A typical x-ray spectrum (background subtracted) is shown in Fig. 1 for bare S ions bombarding on fullerenes. The background spectrum was collected without any target, i.e., at $\sim 6 \times 10^{-7}$ mbar pressure and was found to be very small [see inset (a)]. A similar spectrum for N_2 is shown in inset (b). The different components of the Ly x-ray lines are clearly visible in all the spectra. However, the x-ray spectra are dominated by the hypersatellite line arising due to single capture. The normal satellite which may arise due to double or multiple capture is clearly negligible in both cases, i.e., for fullerene and N_2. This is, however, expected since double electron transfer in the present energy range is very small.

In the case of S^{13+} ions, only the normal component is visible in the x-ray spectrum (not shown). The total x-ray production cross section (per target atom) increases smoothly with Z_T (Fig. 2) and falls on a smooth curve (dashed line). For absolute normalization of the gas target data, Ar K x-ray yields were measured in collisions with 45 and 63 MeV $F^{5+,6+,8+}$ ions and known x-ray cross sections were used [25]. The absolute cross sections for fullerenes, using a normalization procedure derived from first principles [10], falls on a smooth curve and is in agreement with the Z_T dependence observed for the gas targets. The relatively larger error (~ 35–40%) in the case of fullerene target arises due to the large uncertainty in the vapor pressure data [26]. It may be noted that the oven had undergone several heating cycles and therefore an average value of the vapor pressure was used. The continuum distorted wave (CDW) calculations [27] reproduce closely the data for a low Z_T target such as helium and deviate largely for higher $Z_T \geq 7$ for which the collision systems are highly nonperturbative. The same calculations are shown to reproduce the cross sections much better at much higher energy [20], the perturbation strength being small. The cascade contributions [28], following capture into high Rydberg states, were also included along with the CDW predicted capture cross sections to derive the Ly x-ray cross sections. The contribution in the Lyα x-ray due to the M1 transition is negligible and was estimated to be about 0.6% for the present collision geometry considering the lifetime ($\tau_{M1} = 706$ ns) of the $2s - 1s$ M1 decay for S ions [29].

The ratios of intensities of different Ly x-ray lines are free from the normalization error and therefore can provide a more stringent test of models. The "Ly ratio" (R_1) of intensity of Lyβ + Lyγ to that of Lyα [i.e., I($\beta\gamma$)/I(α)], for an emission angle of 90° is plotted as a function of Z_T in Fig. 3(a). The intensity ratios are found to decrease smoothly (see dotted line) for all the gas targets. However, the ratio for the C_{60} is found to be substantially lower compared to the trend followed by the gas targets. The ratio R_1 for C_{60} was always (i.e., at all three energies) found to be lower (by about 15%) than the corresponding gas targets of similar atomic number, such as N_2. [The ratio I($\beta\gamma$)/I(α) also shows similar behavior, not shown.] The CDW calculation was found to reproduce the observed decreasing trend of the ratios except for very low Z_T target atoms (Fig. 3). This observation is found to be independent of the x-ray emission angles and is in agreement with the results at very high energy collisions ($v_p = 36$ a.u.) with thin solid foils [19]. In the case of S^{13+} ions, the K x-rays predominantly arise due to K-shell excitation to higher shells (see [30] for other second order processes) and in such cases the Ly ratios are shown in Fig. 3(b). It may be clearly seen that the results are similar to that for capture [i.e., in Fig. 3(a)].

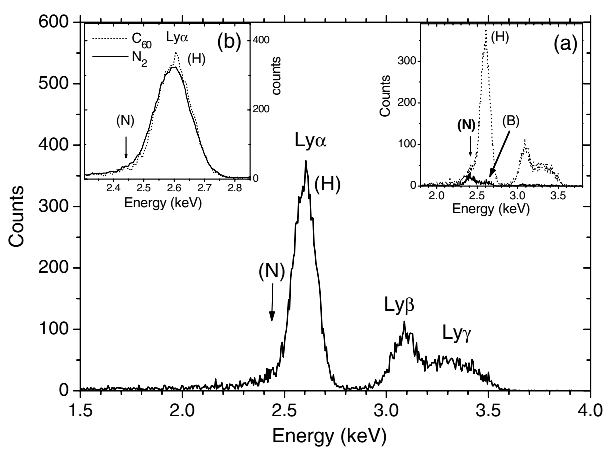

FIG. 1. Lyman x-ray spectrum (background subtracted) following electron capture in collisions of 110 MeV S^{16+} with fullerenes. The inset (a) shows the background spectrum (B) and that from fullerenes (dotted). Inset (b): Similar spectrum for N_2 (solid line) and the fullerenes (dotted). The normal- and hypersatellite components are denoted by (N) and (H), respectively.

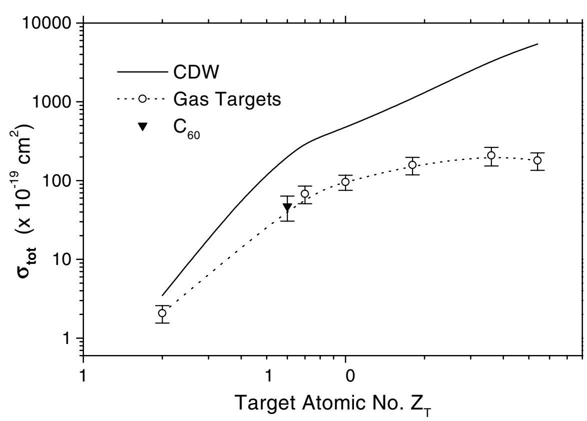

FIG. 2. Total Ly x-ray cross sections (per atom) for gas targets and fullerenes. The dotted line is to guide the eyes.

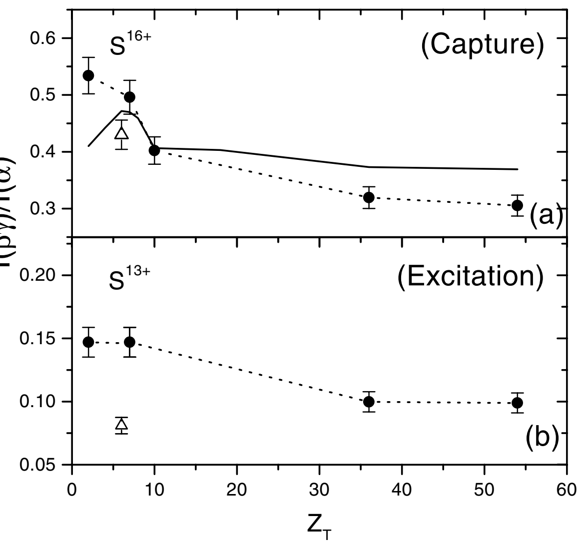

FIG. 3. The intensity ratio (R_1) versus Z_T for (a) S^{16+} and (b) S^{13+}. The solid line in (a) is CDW calculations and the dashed lines are to guide eyes.

To explain these observations, we use a model which has been used [19,20,31] in the case of ion-solid collisions in which the ratio R_1 was substantially ($\sim$ 20%–25%) lower than that for gas targets. The wakefield, due to the collective response of the condensed medium, depends on the plasma frequency [$\omega_p = (4\pi n_e e^2/m_e)^{1/2}$] which is about 21 eV in the present case (which is also nearly the same as the observed giant plasmon excitation energy in C_{60}). This is derived using an average electron density (n_e) $\sim 3.3 \times 10^{23}$ cm^{-3} [Eq. (7) in [13]]. The wakefield has a wavelength $\lambda_w = 2\pi v/\omega_p \sim$ 10 Å which is the same as the diameter of C_{60} (including the surrounding electron gas [23]) and is strong enough ($\sim$ 10 V/Å [19]) to cause a Stark splitting ($\omega_s \sim$ 3.8 eV [13]) of the excited states of $n = 2$. Neglecting the fine structure and QED effects, the wakefield leads to a Stark mixing of the $2s_o$ and $2p_o$ states of the projectile causing a change in the Lyα ($2p$-$1s$) cross section (σ_α). In the case of gaseous targets for which wakefield is absent, this can be written as $\sigma_\alpha^g = \sigma(2p_o) + \sigma(2p \pm 1)$ and CDW was used. For a cluster or condensed medium, this gets modified (see [20,31] for details), and the σ_α^c now depends on the $s - p$ coherence or mixing angle (ϕ) of the $2s_o$ and $2p_o$ states [31], ω_s, the lifetime of the $2p$ state ($\tau_p = 2.3 \times 10^{-14}$ s) [32], and the dwell time τ_d (5×10^{-17} sec).

As the projectile enters the condensed medium, due to the wakefield, the $2s_o$ and $2p_o$ states mix and, depending upon the values of ϕ, ω_s, and τ_d, the population in $2p_o$ will increase compared to a gas target. (The metastable $2s_o$ population now mixes with $2p_o$ which then rapidly decays to $1s$ enhancing Ly-α emission.) The model predicts an enhancement of 10% in the Lyα cross section for the present collision, therefore causing the intensity ratio, R_1, to decrease. The present study deals with very small target thickness and therefore smallest dwell time compared to earlier studies, thus, probing the wakefield induced Stark-mixing effect on a very small time scale ($\sim 10^{-17}$ s).

The CDW model (see Fig. 3) predicts the Z_T dependence of R_1 quite well for $Z_T \geq 6$. Therefore, one may safely expect the ratio R_1 to be nearly the same for $Z_T =$ 6 and 7. In order to explore the gas-cluster difference, we plot (in Fig. 4) the ratio (R_{cg}) of R_1 in the case of C_{60} to that for N_2, for all three beam energies. The ratio ($\sim$0.85) is much lower than the prediction (i.e., $\sim$1.0) of the CDW model, based on ion-atom collisions. The observed lower (by 15%) ratio for C_{60}, according to the model, reflects the existence of the solidlike wakefield effect. Including this effect, as discussed above, the calculated R_{cg} will be about 0.90 (see dotted line in Fig. 4) which is closer to the experimental results within the errors.

In the case of excitation, this ratio is found to be close to 0.5, i.e., even smaller compared to that for capture. In the Li-like ions an electron, already present in the $2s$ state, can mix with the $2p$ to give rise additional Ly-α photons following K excitation. Therefore, wakefield influences the capture as well as the excitation process, qualitatively, in the same way.

A post-collisional long range interaction causing the Stark mixing of the projectile degenerate states ($n = 2$) was invoked [33,34], in order to explain the electron capture process by bare ions. In the present case, the interaction between the polarized C_{60} molecule with giant dipole moment and the captured (or excited) electron could, in principle, produce an additional Stark mixing. However, this field is estimated to be very small [35] compared to the wakefield, and accordingly its effect on the Ly ratio is also negligibly small. The radiative relaxation cascade of hollow ions [9] may also suggest a reduction of the Ly ratio below that for the atomic targets. However, the hollow ion formation is important for

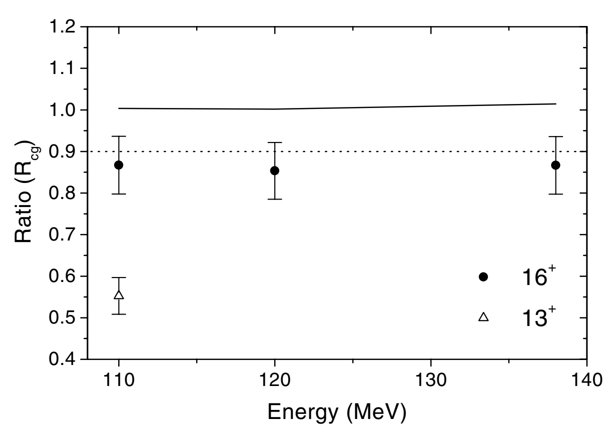

FIG. 4. The ratio (R_{cg}) of R_1 for fullerenes to that for N_2. Solid line: CDW (for 16+) based on ion-atom model. Dotted line: CDW including the Stark-mixing model (see text).

much lower energy collisions. The classical transport calculation [36] considering multiple scattering and high l-state diffusion (and predicting the increase in Ly-α) may not be applicable here, since the target thickness is extremely small.

In the case of ion-solid collisions, with $\kappa = 1$ (Sommerfeld parameter $\kappa = Z_P/v_P$) the Lyman ratio was observed to be 20%–25% less than that for gas targets [19]. In the present study, with similar κ (~ 1.3) but much lower t_d (than that in [19]), we observe this difference to be substantial ($\sim 15\%$) and slightly larger than the prediction (i.e., $\sim 10\%$) of the present model. This provides a scope for further theoretical works by including explicitly the giant dipole polarization mechanism of C_{60} in the model, since the collective response of the cluster giving rise the wakefield could be related to the Giant plasmon excitation under heavy ion impact.

However, the present observations are in qualitative agreement with the enhancement of the REC x-ray yields and the REC energy shifts due to solid-state effects in heavy ion channeling, with similar κ [15,16] and will also be useful to calculate the projectile energy loss in fullerenes since it depends on the plasma oscillation.

In conclusion, we have studied the electron capture and excitation processes via the measurements of projectile Ly x-rays in collisions of highly charged S ions with fullerene molecules and also for gaseous targets. The Lyman ratio, i.e., $I(\beta\gamma)/I(\alpha)$ in the case of fullerene target, falls well below those for the gaseous targets. These observations, most likely signify the existence of a solidlike effect, i.e., wakefield induced Stark mixing of the L sublevels, in a very small time scale: a collective response of the large electron density in the cluster (which could be related to the giant plasmon excitation). The K-excitation process is also found to be influenced by the solidlike effect qualitatively in the same way as in the case of capture.

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[1] G. F. Bertsch, A. Bulgac, D. Tománek, and Y. Wang, Phys. Rev. Lett. **67**, 2690 (1991).
[2] I.V. Hertel *et al.*, Phys. Rev. Lett. **68**, 784 (1992).
[3] D. L. Litchenberger, Chem. Phys. Lett. **176**, 203 (1991).
[4] R. Morgenstern, T. Schlatholter, and R. Hoekstra, *The Physics of Electronic and Atomic Collisions*, edited by Itikawa *et al.*, AIP Conf. Proc. No. 500 (AIP, New York, 2000), p. 65.
[5] L. Chen *et al.*, Phys. Scr. **T80**, 52 (1999).
[6] A. Reinkoster, U. Werner, N. M. Kabachnik, and H. O. Lutz, Phys. Rev. A **64**, 023201 (2001).
[7] J. Opitz *et al.*, Phys. Rev. A **62**, 022705 (2000).
[8] T. Schlatholter, O. Hadjar, R. Hoekstra, and R. Morgenstern, Phys. Rev. Lett. **82**, 73 (1999).
[9] U. Thumm, Phys. Rev. A **55**, 479 (1997); Comments At. Mol. Phys. **34**, 119 (1999).
[10] A. Itoh, H. Tsuchida, T. Majima, and N. Imanishi, Phys. Rev. A **59**, 4428 (1999).
[11] T. LeBrun *et al.*, Phys. Rev. Lett. **72**, 3965 (1994).
[12] B. Walch, C. L. Cocke, R. Voelpel, and E. Salzborn, Phys. Rev. Lett. **72**, 1439 (1994).
[13] J. Burgdorfer, Nucl. Instrum. Methods Phys. Res., Sect. B **67**, 1 (1992).
[14] P. M. Echenique, R. H. Ritchie, and W. Brandt, Phys. Rev. B **20**, 2567 (1979).
[15] L. C. Tribedi *et al.*, Phys. Rev. A **51**, 1312 (1995).
[16] J. M. Pitarke, R. H. Ritchie, and P. M. Echenique, Phys. Rev. B **43**, 62 (1991).
[17] S. Datz *et al.*, Phys. Rev. Lett. **40**, 843 (1978).
[18] T. Azuma *et al.*, Phys. Rev. Lett. **83**, 528 (1999).
[19] J. P. Rozet *et al.*, Phys. Rev. Lett. **58**, 337 (1987).
[20] J. P. Rozet *et al.*, *Proceedings of the 3rd International Workshop on High Energy Ion-Atom Collisions*, edited by D. Berenyi and G. Hock, Lecture Notes in Physics (Springer-Verlag, Berlin, 1988), Vol. 294, p. 307.
[21] H. Brauning *et al.*, Phys. Rev. Lett. **86**, 991 (2001).
[22] H. D. Betz, D. Roschenthaler, and J. Rothermel, Phys. Rev. Lett. **50**, 34 (1983).
[23] O. Hadjar, R. Hoekstra, R. Morgenstern, and T. Schlatholter, Phys. Rev. A **63**, 033201 (2001).
[24] Obtained from MER corporation, Tucson, AZ.
[25] B. B. Dhal *et al.*, Phys. Rev. A **62**, 022714 (2000).
[26] J. Abrefah, D. R. Olander, M. Balooch, and W. J. Siekhaus, Appl. Phys. Lett. **60**, 1313 (1992).
[27] Dź. Belkic, J. Phys. B **11**, 3529 (1978).
[28] K. Omidvar, At. Data Nucl. Data Tables **28**, 1 (1983).
[29] R. Marrus and P. J. Mohr, Adv. At. Mol. Phys. **14**, 181 (1978).
[30] U. Tiwari *et al.*, Phys. Rev. A **58**, 4494 (1998); M. Chabot *et al.*, J. Phys. B **27**, 111 (1994).
[31] I. A. Sellin, L. Liljeby, S. Mannervik, and S. Hultberg, Phys. Rev. Lett. **42**, 570 (1979).
[32] H. Panke, F. Bell, H. D. Betz, W. Stehling, E. Spindler, and R. Laubert, Phys. Lett. **53A**, 457 (1975).
[33] J. Burgdorfer, Phys. Rev. A **24**, 1756 (1981).
[34] J. Burgdorfer and L. J. Dubé, Phys. Rev. Lett. **52**, 2225 (1984).
[35] L. G. Gerchikov, A.V. Solovyov, J. P. Connerade, and W. Greiner, J. Phys. B **30**, 4133 (1997).
[36] J. Kemmler, J. Burgdorfer, and C. O. Reinhold, Nucl. Instrum. Methods Phys. Res., Sect. B **67**, 168 (1992).