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Interaction of slow $Ar^{(17,18)+}$ ions with C_{60}: An insight into ion-surface interactions

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The interaction of $Ar^{(17,18)+}$ ions with C_{60} has been studied by observing coincidences between Ar *K* x rays and the fullerene ions and fragments. At large distances the capture of electrons from C_{60} into excited states of the ion has been observed and compared to the interaction of the same ions with surfaces. Most of the observed events correspond to the capture of many electrons by the ion and the loss of all but one. These results show clearly the characteristic behavior of ions flying over a surface without any contact. [S1050-2947(96)50605-0]

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Highly charged ions approaching, at low velocity, a metal surface are known to capture, at large distances, conduction electrons into large *n* states of the projectile [1–4]. These highly excited ions then decay to the ground state through a cascade of Auger transitions ending, e.g., by the emission of a *K* x ray [5,6]. This cascade usually takes a time period longer than that needed for the ion to touch the surface and one cannot expect that many x rays are emitted by the ion prior to the impact. Thus the highly excited ions partly explode on the surface (the most weakly bound electrons are peeled off) [7] and a new capture process takes place below the surface [6], ending with the emission of characteristic x rays inside the bulk. Because of the acceleration of the ion by its own image on a metal surface [8], it is not possible to reduce its normal velocity and thus increase its transit time outside the surface to allow the observation of pure ''outside'' x rays. Thus one observes only a very weak x-ray spectrum emitted outside the surface [4,9–11] superimposed on a very intense spectrum characteristic of the ''inside'' interaction. We present in this Rapid Communication an experiment aimed at the observation of the x-ray spectrum that is mainly due to an above surface interaction. The idea is to look at the x rays emitted in flight by Ar^{17+} and Ar^{18+} ions interacting with a C_{60} vapor beam. Electron capture occurs at a distance (~20 Å) much larger than the radius (3.5 Å) of the C_{60} cage; therefore one may expect the geometrical cross section for the ''outside'' interaction to be much larger (~92%) than that for any head-on collision (penetration). Thus most of the observed spectrum would be characteristic of the x rays emitted outside the target, and mimic, more or less, what happens above a surface.

The Ar^{17+} and Ar^{18+} ions were produced by the Advanced Electron Cyclotron Resonance ion source of the 88-inch cyclotron of the Lawrence Berkeley National Laboratory at an energy of 10 keV/*q*. They were mass and charge analyzed on the joint Lawrence Berkeley National Laboratory–Lawrence Livermore National Laboratory (LBNL-LLNL) ion beam facility by two dipoles and sent into a vapor beam produced by heating C_{60} powder to 430 °C in an oven. The ion x rays were analyzed with a Si(Li) detector with a resolution of 147 eV (full width at half maximum) at 6 keV, and the charged fullerene ions, or fragments, by a time-of-flight apparatus (Fig. 1).

In Fig. 2 we present a scatter plot and projections of the recoil-ion time of flight versus the Ar *K* x-ray energy. The $K\alpha$ line [Fig. 2(c)] has an energy equal to the mean value of the ${}^1P_1$ and ${}^3P$ $(1s)(2p)\rightarrow(1s)^2$ transition (3139.6 and 3123.6 eV, respectively) [12] of the singly excited He-like Ar ion (Table I) and a width equal to the detector resolution (i.e., excluding any ''contamination'' by the satellite lines corresponding to an $L\rightarrow K$ transition in the presence of a

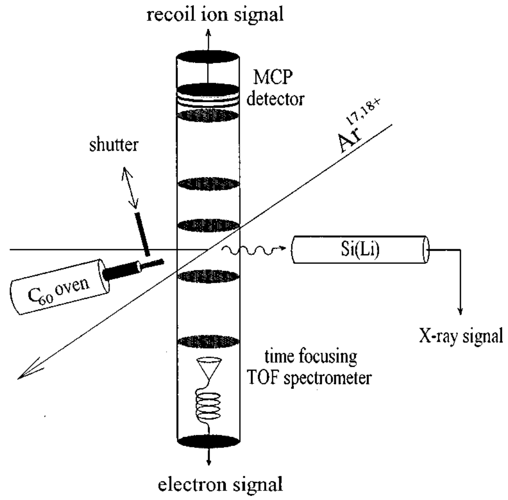

FIG. 1. Experimental setup. $Ar^{17,18+}$ ions intercept a fullerene beam produced by a low-temperature oven. The recoil ions are extracted at 90° by a vertical electric field and detected by a double-sided, time focusing time-of-flight (TOF) spectrometer in coincidence with the Ar x rays. The electron side of the TOF spectrometer is used for diagnostics.

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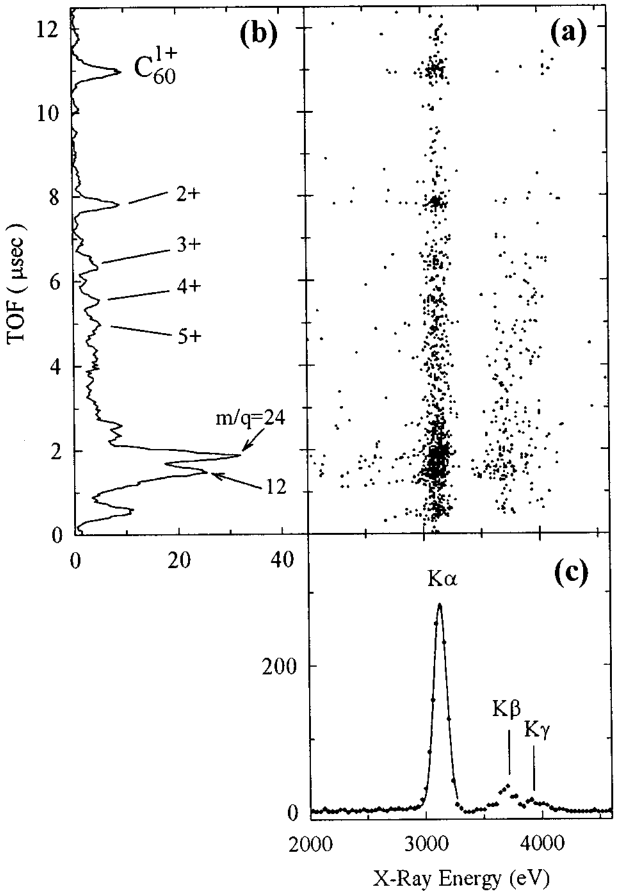

FIG. 2. Ar K x-ray–recoil ion coincidence data from Ar^{17+} (170 keV) collisions with C_{60}. (a) C_{60} product ion time-of-flight vs x-ray energy. (b) Projection of (a) on the time-of-flight axis. (c) Projection of (a) on the x-ray energy axis.

certain number of L spectator electrons, as is observed below a surface). This fact is somewhat surprising because the interaction with C_{60} is expected to be intermediate between that with a gas target (mainly capture of one electron) and that with a solid, where, according to the resonant neutralization model [1], many electrons of the conduction band are captured predominantly into excited states of the ion having roughly the same binding energy. Here, the capture of the first electron from a C_{60} will populate a state with principal quantum number near $n=13$. Furthermore, owing to the increase of the binding energy of the electrons due to the positive charge buildup on the C_{60}, the subsequent capture occurs into slightly lower n states of the ion. We are thus dealing with capture processes filling various excited states of relatively large principal quantum number. By observing the x rays, we detect the last step of the decay cascade from highly excited ions with a relatively large number of electrons. Unfortunately the observation of the KL^1 line does not allow us to determine the number of outermost (M, N, etc.) shell electrons that were present at the time of the x-ray emission (the energy shift due to the presence of outermost shell spectator electrons is not large enough to be detected). In order to determine the number of electrons still attached to the ion at the time of the x-ray emission, we reproduced the same experiment with Ar^{18+} ions where the two K vacancies are filled sequentially [13,14]. The corresponding spectrum (Fig. 3) shows the H-like line and a small bump at the energy of the He-like line which comes mainly from the charge-exchange processes along the beam line, transforming Ar^{18+} ions into Ar^{17+} (as experimentally proven by the study of the beam interaction with the residual gas). However, a very small contribution of the He-like $K\alpha$ line cannot be completely excluded. This result shows that after the filling of the first K hole the ion has no more electrons to fill the second one, and ends its life mainly with just one electron.

TABLE I. Energy of the measured $2p \rightarrow 1s$ lines presented in Figs. 2(c) and 3. $\overline{E}$=energy (theory and experiments of H-like and He-like Ly-α lines, assuming a statistical population). When one more electron is present in the ion this energy is reduced by ~26 eV.

	E $2p \rightarrow 1s$ line	$\overline{E}$ (Ref. [12])
Ar^{17+}/C_{60}	3131±10 eV	3129 eV (He-like)
Ar^{18+}/C_{60}	3330±10 eV	3319.8 eV (H-like)

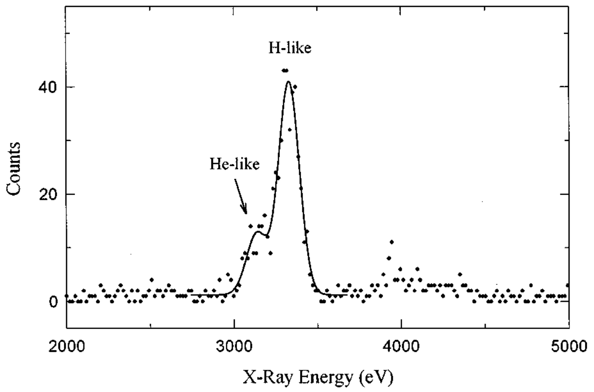

FIG. 3. X-ray spectrum observed from 180-keV Ar^{18+} collisions with C_{60}.

As can be seen in the scatter plot [Fig. 2(a)], the K x rays are correlated with the highly charged fullerene ions $C_{60}{}^{q+}$ or fragments, whose time-of-flight spectra have been previously observed [15,16]. When corrected for the less than unity efficiency [15] for detection C_{60} ions with $q=1,2$, the data of Fig. 2 show that ≈45% of the x-ray intensity is associated with C_{60} ions, and 55% with the light fragment ions.

These results must be compared with those of Walch *et al.* [15] on Ar^{8+} colliding with C_{60}. These authors studied the coincidence between the scattered projectiles and the recoil ions; they found correlation between either nearly neutralized Ar ions (many electrons captured and retained) and charged fragments (destruction of the fullerene), or highly charged projectiles (mostly Ar^{7+}) and fullerene $C_{60}{}^{q+}$ ($1<q<6$). They observed that ≈64% of the total collision cross section was associated with C_{60}-ion production vs 36% of the light fragment ions. At large distances, the transfer of C_{60} electrons populates high-n states of the ion via resonant neutralization and leads to a cascade of Auger transitions (such a capture into high-n states cannot occur inside a solid or at its surface because of the short interaction distances). At much closer distances, i.e., one C_{60} cage radius plus one or two angstroms, Ar ions capture carbon electrons into the *M* and *N* shells. In the case of Ar^{8+} (neonlike ions), this close capture process populates mainly the *M* shell, i.e., the ground configuration. The product ions are thus stable and retain most of the captured electrons, as observed by Walch *et al.* Of course it is not known how many electrons may have been freed in those collisions producing fragment ions. The ratio of fragment to $\mathrm{C}_{60}{}^{q+}$ production observed by Walch *et al.* is consistent with the ratio of cross sections for capture of seven or more electrons to that for up to six electrons, as predicted by the classical barrier model.

Assuming that fragmentation occurs by overcharging the fullerene (to perhaps near 11 units [16]), our observation of the relative intensity of x rays associated with fragments vs those with $\mathrm{C}_{60}{}^{q+}$ ions suggest that Ar^{17+} collisions reaching separations inside those for capture of the first few electrons, result in removal of more electrons from C_{60} than is predicted by the classical barrier model. A rapid Auger emission process during the collision and/or penetration (tunneling) of the classical barrier may explain this observation.

In order to explain the complete removal of all captured electrons (minus one), we have to estimate the mean number of Auger steps needed for the electrons to reach the *L* shell. The Auger decay of ions having several electrons in highly excited states proceeds through some very specific cascade paths, as described in Ref. [11].

(i) The Auger rates are known to be maximum for the lowest electron energies; thus the cascade proceeds through the closest, energetically allowed levels. Hence a large number of steps has to take place before the excited electrons reach the innermost shell (e.g., $n=10 \rightarrow n=8 \rightarrow n=6. \ldots$).

(ii) Although the Auger rates for two electrons in Rydberg states are quite low, the total rate for levels having many electrons scales like $m^{1.5}$ (or more, *m* being the number of electrons). The decay of a given *n* state then starts to be substantially fast when many electrons are present in the shell.

These two rules of thumb lead to a very large number of steps for the decay of these very excited levels. (The $n=8$ level, for instance, in a cascade starting at $n=10$, ‘‘waits’’ to decay until it gets enough electrons.) We calculated previously [11] the lifetimes of the Auger transitions for some typical ions having 17 electrons in $n=10$. These numbers show that a cascade of more than 15 Auger transitions is needed before the first electron reaches the *L* shell of the ion. The ion is fully (minus one electron) reionized, long before the last remaining electron has reached the *L* shell, and then decays through the emission of a pure He-like (or H-like) characteristic x ray, in agreement with the experimental results. These results also prove that the capture of conduction (or valence) electrons of the target populates, outside the surface, the high-*n* states of the ion ($n>10$ for Ar^{17+}), in agreement with the resonant neutralization model when an ion is flying parallel to the ‘‘surface.’’

In about 35% of the events one also observes some coincidences between the capture of one or two electrons ($\mathrm{C}_{60}{}^{1+}$ or $\mathrm{C}_{60}{}^{2+}$) and the emission of He-like lines [Fig. 2(a)]. Obviously these processes mainly come from capture events at very large distances (~20 Å), i.e., when the ion stays just a short time at the barrier radius and captures one electron (or two). However, we notice in this spectrum the absence of any $K\beta$ and γ lines and an extremely weak contribution at the energy of the series limit that shows that large-*n* states are populated and decay through an Yrast cascade, i.e., that capture takes place into large-*l* states. This is not surprising for collisions involving very large impact parameters.

In conclusion we have observed some events that show the very specific behavior of an ion interacting with a surface without contact. We have shown that capture occurs into large-*n* states of the ion in an original situation where there is no dynamical screening of the ion (large ‘‘parallel’’ velocity, the ion flies over a quite large fraction of the surface). We have then observed what happens when an ion captures many electrons and cannot be easily refed (it escapes from the capture area much faster than it loses electrons through Auger decays). This specific behavior where an ion captures and loses many electrons in a very short time will certainly be of interest for studying the interaction of highly charged ions with insulators where one can expect to observe some ion backscattering.

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