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# X-ray studies of the interaction of N, O, and Ne hydrogenlike ions below surfaces

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We present in this paper some experiments on the interaction of slow $N^{6+}$, $O^{7+}$, and $Ne^{9+}$ ions below C or Si surfaces carried out by looking at the projectile and target x rays. The study of the x rays emitted by the ions, in contrast with studies of Auger electrons, allows the observation of a much larger part of the decay, not yet explored, below the surface. Moreover, the x rays emitted by the target atoms may identify the shell from which the electrons are captured. It is shown that the electron promotion mechanism, previously observed, which transfers, e.g., $K$ electrons of C targets into the $L$ shell of these ions, represents only a very small part of the interactions occurring at the first atomic layer and that the neutralization takes place, below the surface, mainly via Auger neutralization. [S1050-2947(97)01705-8]

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All the experiments to date on the interaction of relatively low charged ions (e.g., $C^{5+}$, $N^{6+}$, $O^{7+}$, $Ne^{9+}$, or $Ar^{q+}$, $8 \leqslant q \leqslant 16$) on surfaces have been dealing with the study of the Auger electrons emitted in flight by these ions [1–3]. The main objective of these studies was to observe the decay of these ions above or at the surface (below the surface, i.e., on the first few atomic layers, the Auger electrons are slowed down and cannot provide precise information). We present in this paper several experiments on the interaction of $N^{6+}$, $O^{7+}$, and $Ne^{9+}$ ions on various targets at large incidence angle ($\theta = 45°$) and at an energy of 15 keV/$q$ such that most of the observed decay takes place below the surface. In these experiments, instead of looking at the Auger electrons, we studied, with a low-energy SiLi detector, the x rays emitted in flight by the ions, i.e., some ''below the surface'' signals unperturbed by the presence of the ions inside the bulk (no energy loss in matter). The main interest in these ions of the second row of the Periodic Table is that the observed emission of these originally hydrogenic ions comes from the $K$ hole, after the collision ($K$ x ray or $KLL$ Auger), instead of the $L$ shell, which is the case for most ions of the third row of the Periodic Table (e.g., $Ar^{q+}$). In this case the lifetime for the filling of the $K$ hole is about ten times longer than that of one $L$ hole. At the most commonly used velocities (those corresponding to the extraction voltage of an electron cyclotron resonance (ECR) source, i.e., $\sim 10^6$ $\mathrm{ms}^{-1}$), the decay length is rather long, about 50 Å, and emission occurs well below the surface (the filling of one $L$ hole occurs along a range of about 5 Å [4]).

The x rays were detected by means of a windowless SiLi detector of high resolution and very low background, capable of detecting x rays down to the C $K$ line (277 eV). In most experiments this detector was protected against particles by a very thin Formvar window. At low energies calibrating the detector is a delicate operation (there is no radioactive sample delivering very-low-energy photons and the C, N, O, and K lines must be excited by electron bombardment or fluorescence). In our experiment we calibrated the detector by looking at the x rays emitted by the ions during their interaction with the residual gas of the beam line ($p \sim 10^{-8}$ mb). In these conditions the ions capture mainly one electron in their excited states and emit the He-like Lyman $\alpha$ lines ($\omega = 1$), which have a well-known energy [5] (Fig. 1). (Double-capture processes, less than 10%, lead to the formation of doubly excited Li-like ions decaying mainly via Au-

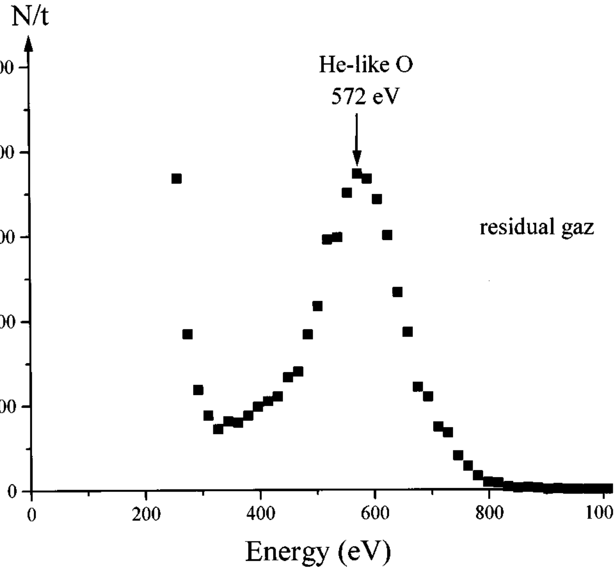

FIG. 1. $K$ x-ray spectrum from $O^{7+}$ collisions with the residual gas in the target chamber. Predominant single-electron capture to highly excited levels of $O^{6+}$ leads to a pure He-like $K$ x-ray spectrum useful for the calibration of the SiLi detector.

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ger transitions and do not contribute appreciably to the observed x rays.) It was then possible to measure the energy of the x rays emitted during the interaction of these ions with solid targets within 5-eV precision. We used two different targets: SiH (silicon covered by a single monolayer of hydrogen prepared following some well-established chemical procedures and having a well-defined atomic nature just below the first layer: no $SiO_2$) and C foils. These experiments were performed at the advanced electron cyclotron resonance (AECR) ion source of the 88-Inch cyclotron of the Lawrence Berkeley National Laboratory on the joint Berkeley-Livermore beam line facility at an energy of 15 keV/$q$. We present in Figs. 2 and 3 the $K$ x-ray spectra observed when N, O, and Ne H-like ions collide with SiH and C targets and in Fig. 4 the energy of the lines that are observed.

The energy of the x rays emitted during the interaction of $N^{6+}$ and $O^{7+}$ with Si and C targets is found to be equal to that of neutral atoms. This means that for N and O the $L$ shell of the ions is completely filled before the emission of a $K$ x ray (filling of the $K$ hole) and also that no signature of the formation of hollow atoms below the surface is observed. The broadening of the $KLL$ Auger lines that is observed below surfaces is thus most probably due to the straggling of the Auger electrons and not to an incomplete filling of the $L$ shell. As demonstrated with low-energy collisions by Folkerts and Morgenstern [6], $N^{6+}$ ions capture electrons in the $L$ and $M$ shells; the $L$ shell is partly filled by Auger transitions from the $M$ shell and partly through some direct collisional filling (the so-called side feeding). With $Ne^{9+}$ on Si and C targets the $K$ line lies at the mean energy of the $KL^4$ satellite. These results confirm those presented by Hustedt *et al.* [7], who found for certain targets, at comparable energies and incidence angles, a mean number of $4-5\,L$ electrons at the time of the $K$ hole filling. In the absence of any information on the number of $M$ electrons attached to the ion when the $K$ hole is filled, the incomplete filling of the $L$ shell could not signal the formation of hollow atoms. These authors, however, observed clear $KLM$ Auger transitions, which means that the $M$ shell has also been populated and that hollow atoms were probably formed.

In some collisions, e.g., $Ne^{9+}$ or $O^{7+}$ on C targets, it was possible to observe simultaneously the $K$ x rays of the projectile and eventually those of the target: the projectile being singly ionized in the $K$ shell, the fluorescence yields of both the projectile and the target are comparable (the x-ray signals are, in any case, much weaker than the Auger emissions). As shown in Fig. 3, no, or very few, C $K$ x rays are observed in these collisions.

These findings clearly establish that the captured electrons do not come from the $K$ shell of the target and that (i) there is no, or very few, electron promotion processes, e.g., transfer of projectile vacancy into the $K$ shell of the atoms below the surface, (ii) nor any Auger neutralization mechanism involving inner shells of the target as we previously suggested [8] (e.g., transfer of one $K$ electron of C into the $K$ shell of the ion and emission of a second one into the continuum: $KKK$ Auger). Such a transition, which leaves the C $K$ shell doubly ionized, would have led to the observation of C $K$ hypersatellite and satellite x-ray emission [9].

These findings, however, seem to be in contradiction with the accurate results obtained by Meyer *et al.* [1] for $N^{7+}\rightarrow C$ or Hustedt *et al.* [7] for $Ne^{9+}\rightarrow C$, who clearly observed the $KVV$ ($V$ denotes valence) Auger lines of carbon. These results were first explained by Meyer *et al.* [1] by a Fano-Lichten–type electron promotion mechanism transferring one $L$ vacancy of the projectile (heavier partner of the collision) into the $K$ shell of the target (the lighter partner).

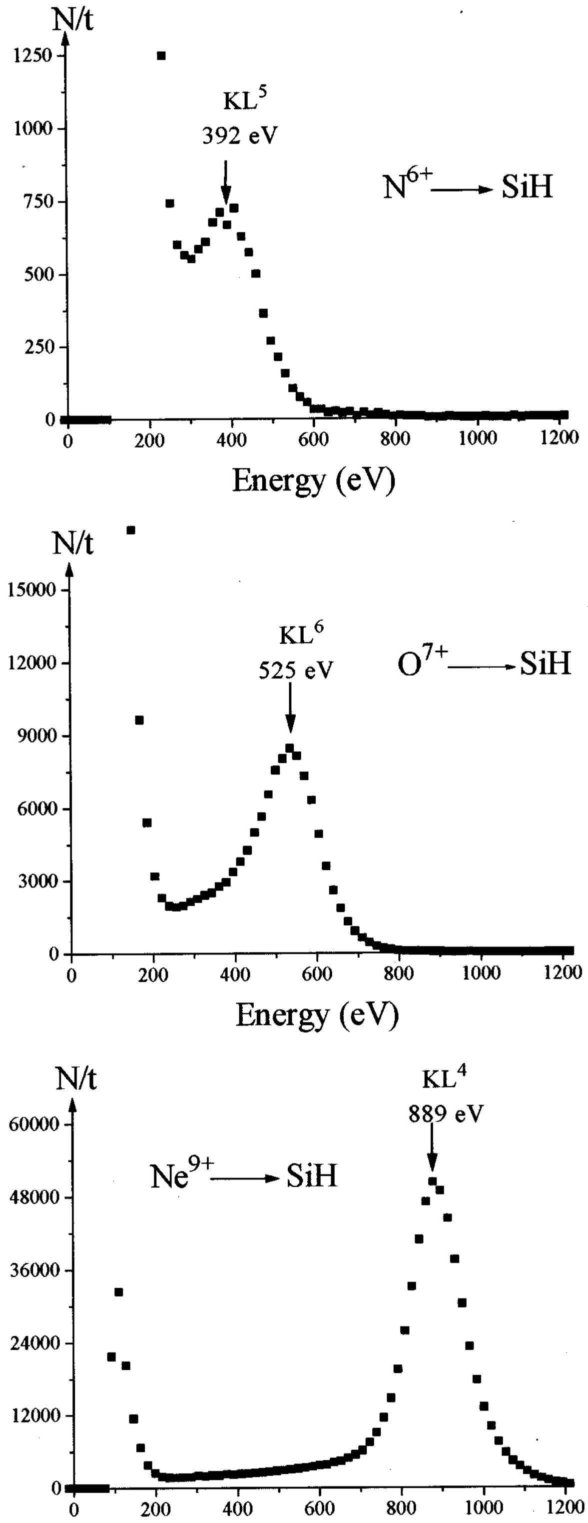

FIG. 2. $K$ x-ray spectra from $N^{6+}$, $O^{7+}$, and $Ne^{9+}$ impact on SiH surfaces. The arrows indicate the location of the $KL^n$ satellite line matching the peak in each spectrum.

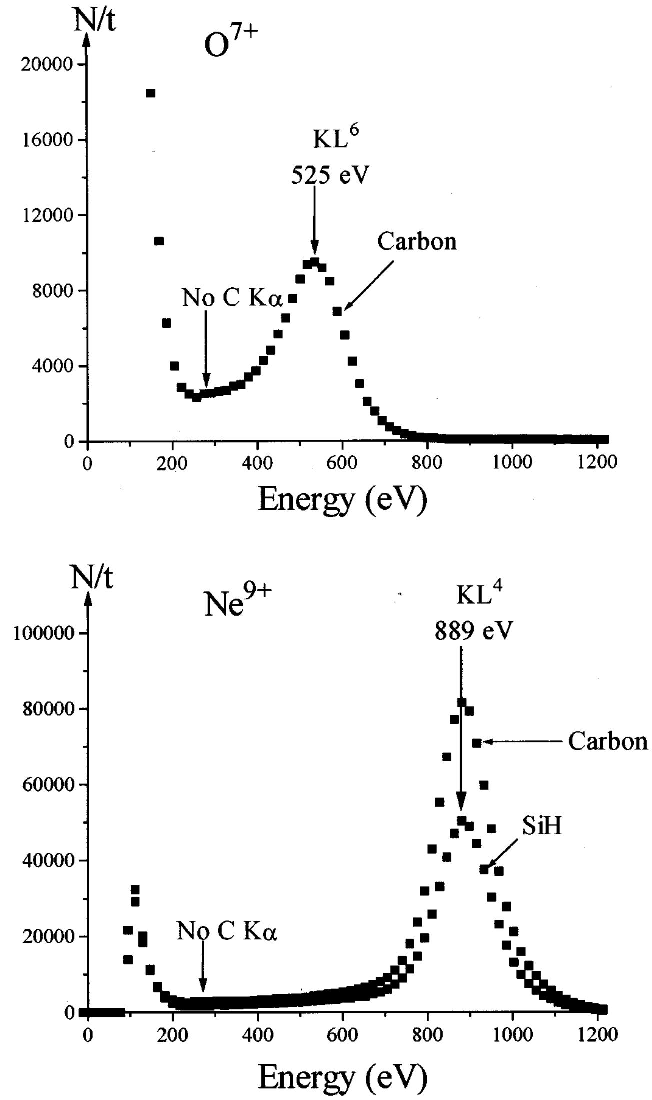

FIG. 3. *K* x-ray spectra from $O^{7+}$ impact on C and $Ne^{9+}$ impact on C and SiH surfaces. No C $K\alpha$ emission is present above background for the spectra from the C targets.

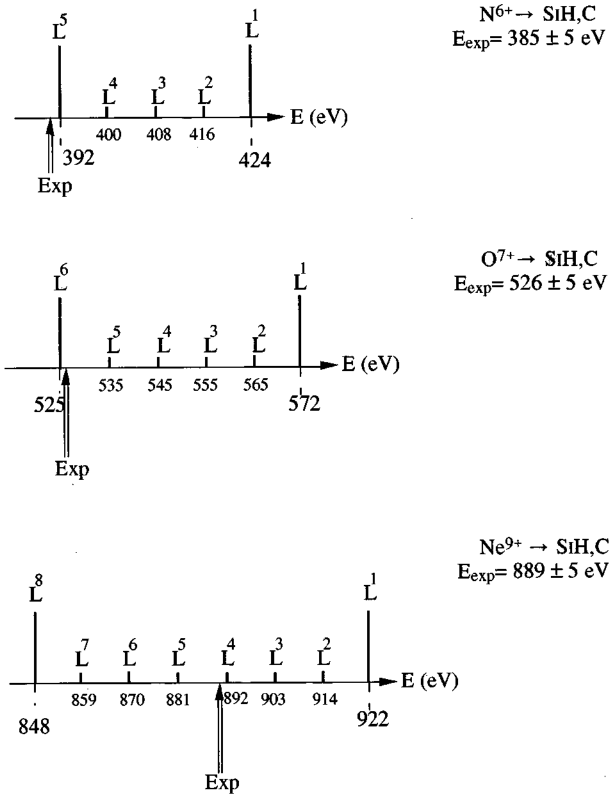

FIG. 4. Comparison of experimental $K\alpha$ energies from $N^{6+}$, $O^{7+}$, and $Ne^{9+}$ collisions with SiH and C targets with theoretical predictions for the $KL^x$ satellites ($x$ is the number of $L$ electrons present at the time of the $K\alpha$ emission).

This apparent discrepancy may be explained, as discussed below, by the fact that Auger spectroscopy displays only the small part of the decay of the projectile occurring just at the surface where the C atoms lie.

The N and O projectiles have been found to have all their electrons, except in the *K* shell, at the time they emit x rays. The main decay path of an ion having a lifetime of the order of $5\times10^{-14}$ s (e.g., N) at a velocity of $\sim10^6$ $ms^{-1}$, is therefore ~50 Å, i.e., well below the surface; x-ray emission thus occurs very likely long after the ion is quasineutralized. So we observe in x-ray spectroscopy the whole decay of the last inner hole of the ion, whereas in Auger spectroscopy this decay is observed only in the first (one or two) monolayers. At a grazing incidence, however, the ion may be specularly reflected or incoherently scattered and a larger part of the interaction may be observed by Auger spectroscopy.

In all the experiments performed with Auger spectroscopy discussed above, the surfaces were carefully analyzed and the C deposits never exceeded one monolayer. In our experiment the C foils were contaminated on the first monolayer by O compounds such as $H_2O$ and CO, as proven by Auger analysis (20% contamination of O in a six-monolayer C foil). Most of the x rays are then emitted by the ions, below the first O compound monolayer of the surface, inside the carbon substrate. In Auger spectroscopy one observes the whole Auger emission of C from the first monolayer, but only a small part of the signature of the projectile *K* hole filling. The observation of the disappearance of the N projectile Auger lines with increasing incidence angles [1], while the intensity of the target Auger lines remains constant, confirms this view. Consequently, in Auger spectroscopy, the relative intensity of the target emission is greatly enhanced with respect to that of the projectile. In x-ray spectroscopy the observed relative intensity of the photons emitted by the target and the projectile must be much closer to the actual emission rates. The transfer of a C *K* electron into the *L* shell of the projectile is then a much smaller fraction of the whole interaction than appears in Auger spectroscopy (less than 5–10 % of the overall events and hence difficult to identify in our experiments) and holds only in the first atomic monolayer of C available on the surface. So, as could be expected, it is a binary collision between the projectile and a single atom of the surface. Below this first layer the ion must complete its neutralization by capturing other electrons of the substrate into its *L* shell (and *M* shell for Ne). In the present experiments there were very few C atoms on the top layer of the surface and the main part of the interaction took place below this layer inside the C substrate from where we do not observe any C *K* x-ray emission. Subsequently, the ion captures *L* (conduction or valence) electrons of C instead of *K* electrons through another mechanism more characteristic of a bulk interaction: the Auger neutralization process. This mechanism, first considered by Hagstrum [10] as responsible for the ion neutralization at close distances for ion velocities lower than or comparable to the Fermi velocity, directly transfers the conduction electrons of the solid into the ion vacancies while another electron is ejected into the continuum.

From these experiments one concludes that the electron promotion mechanisms, which have been unambiguously observed in $N^{6+}$, $O^{7+}$, and $Ne^{9+}$ collisions on C surfaces, take place just at the surface (in the first monolayer) and thus represent only a part of the first step of the neutralization process. This finding is consistent with the Fano-Lichten promotion mechanism, which is a binary collision process. Below the surface the ion interacts with several atoms at the same time. The main part of the neutralization, i.e., the capture of many valence or conduction electrons, occurs in the bulk through the Auger neutralization process.

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