Image Charge Acceleration of Multicharged Ions in Front of the Surface of an Insulator

C. Auth, T. Hecht, T. Igel, and H. Winter
Institut für Physik, Humboldt-Universität zu Berlin, Invalidenstrasse 110, D-10115 Berlin, Germany
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Multicharged xenon ions are scattered with keV energies from a LiF(100) surface under a glancing incidence. From the angular distributions of scattered projectiles, we deduce the existence of an attractive force acting on the projectiles on the incident path. We interpret this as the dielectric response of the insulator due to the presence of an ion. The interaction energies gained on the incident trajectory are reproduced by an "overbarrier" model of stepwise capture of electrons from the fluorine $2p$ band of LiF into Rydberg levels of the projectile.

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In recent years the interactions of multiply and/or highly charged ions with surfaces have been the subject of active research, which has profited from developments in ion-source technology for slow multiply and highly ionized atoms. The considerable amount of potential energy stored in those ions is liberated in collisions with a solid in complex multielectron processes [1]. These processes, in addition to the fundamental interest concerning the interaction mechanisms, are relevant with respect to practical aspects (e.g., plasma-wall interactions), and might have the potential for future technological applications (e.g., ion-beam modification and analysis of surfaces).

Most experimental work in this field has focused on measurements of x-ray [2] and electron emission [3–5] and has led in recent years to significant progress in the understanding of the interaction mechanisms [6,7]. The interaction between ion and metal *in front* of the surface is described by a classical "overbarrier" model [8], where the neutralization of the ions proceeds at rather large distances from the surface by the resonant transfer of conduction band electrons into (multiply excited) Rydberg levels of the projectiles, forming so called "hollow atoms." Since most of the inner shell vacancies of the projectiles survive this first step, the high initial potential energy is still available. Then at and *below* the surface plane the inner shells will be rapidly filled in close encounters with target atoms and conduction electrons of high densities to complete the relaxation sequence.

Recently, we have shown that the trajectory of multicharged ions is affected by a pronounced dynamic image force produced by the charged projectile in front of the surface [9]. In scattering ions under glancing angles from clean and flat metal surfaces we have deduced from angular distributions for scattered projectiles information on the image charge acceleration of ions on the incoming trajectory. Since the image interaction depends strongly on the charge of the projectile, one obtains from image interaction energies information on neutralization in front of the surface. The data for metal surfaces give clear evidence for the formation of hollow atoms and are astonishingly well reproduced by an overbarrier model [6–11].

In contrast to the interaction of multicharged ions with metal targets, studies with insulators are rare. Aside from some early work, detailed investigations focused on sputtering [12] and on electron emission phenomena [13,14] have reported the first results. In the analysis of the data obtained in those studies, information is needed on the effect of the dielectric response of the target on the projectile itself and, in particular, on the neutralization sequence in front of the surface. Before this work was performed, it was an open issue how the trajectory of a (multi)charged atomic particle is affected by the presence of an insulating dielectric medium and how charge transfer between insulator and ion proceeds.

The method applied in our studies is based on the analysis of angular distributions for scattered projectiles [9]. In brief, scattering of ions from a surface under a glancing angle Φ proceeds under "surface channeling" conditions, where the energies for the motion normal as well as for the motion parallel with respect to the surface are conserved. These energies amount to $E_z = E_0 \sin^2\Phi \approx E_0\Phi^2$ and $E_\parallel = E_0 \cos^2\Phi \approx E_0$ (E_0 = projectile energy). At $\Phi \approx 1°$ the parallel motion proceeds with E_0, whereas normal energies are about 4 orders of magnitude smaller. For keV ion beams the normal motion proceeds with eV energies, and the projectiles do not overcome the repulsive potential of the surface and are reflected in front of the topmost layer of surface atoms. For conservation of energy the projectiles are reflected specularly from the target surface: $\Phi_{\text{in}} = (E_y/E_\parallel)^{1/2} = \Phi_{\text{out}}$.

This condition is violated for charged projectiles by the dynamical image interaction, and multicharged ions gain considerable normal energies E_{im} on the incident trajectory. Under the experimental conditions of our studies projectiles leave the surface neutralized (ion fractions here $<1\%$) and an image interaction on the outgoing path can be neglected. The normal energy on the incident path is enhanced from E_y to $E_y' = E_y + E_{\text{im}}$, resulting in an increased effective angle of incidence $\Phi_{\text{in}}' = (E_y'/E_\parallel)^{1/2} = \Phi_{\text{out}}'$. The image interaction energies E_{im} can be deduced from angular distributions for scattered projectiles, where image charge effects manifest

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themselves by scattering angles $\Phi_s' = \Phi_{in} + \Phi_{out}'$ larger than the scattering angle $\Phi_s = \Phi_{in} + \Phi_{out} = 2\Phi_{in}$ for specular reflection. E_{im} is obtained from the simple relation $E_{im} = E_0[(\Phi_s' - \Phi_{in})^2 - \Phi_{in}^2]$.

Essential components of our setup are sketched in Fig. 1. A well collimated beam of 50 keV multicharged xenon ions from a NANOGAN–10 GHz–ECR ion source [15] is directed onto the (100) face of a polished LiF monocrystalline target along a high-index crystallographic direction in the surface plane ("random"). Angular distributions of the (neutralized) projectiles are recorded in the plane of scattering by means of a channeltron detector mounted on a precision manipulator. At a base pressure of about 10^{-10} mbar the preparation of the surface is performed by cycles of grazing sputtering of the target ($\Phi_{in} \approx 2°$) at about 330 °C with 25 keV Ar^+ ions and subsequent annealing to temperatures of 400 °C.

Of the various procedures to avoid charging up of the target [16] we find keeping the target at temperatures of about 300 °C to be the most appropriate one, where LiF is a sufficiently good ionic conductor. For our method, well defined angular distributions of the scattered projectiles are an essential prerequisite, which means the time-consuming preparation of extremely clean and flat surfaces. In Fig. 2 we present as a representative example of our studies angular distributions obtained with 50 keV Xe^0 (full circles) and Xe^{12+} projectiles (open circles). A pronounced angular shift towards larger angles of scattering is found for the ions, as expected for an attractive force acting on the incident trajectory. To our knowledge these data represent the first demonstration of image charge effects on the trajectory of ions in front of the surface of an insulator.

The analysis of the data is straightforward. The intense narrow peak on the left side stems from a residual fraction of the direct beam and serves as a reference for the direction of the projectile beam (see Fig. 1). For the neutral beam we assume specular reflection conditions (no image force on incident and outgoing trajectory) and obtain from the angle of scattering $\Phi_s = 1.16°$ an angle of incidence $\Phi_{in} = \Phi_s/2 = 0.58°$ so that $E_y = E_0 \sin^2 \Phi_{in} = 5.1$ eV. For the Xe^{12+} ions we find $\Phi_s' = 2.27°$ and $E_y' = E_0 \sin^2(\Phi_s' - \Phi_{in}) = 43.5$ eV. The ions have gained in front of the surface an image interaction energy of $E_{im} = 38.4$ eV. In Fig. 3 we have summarized

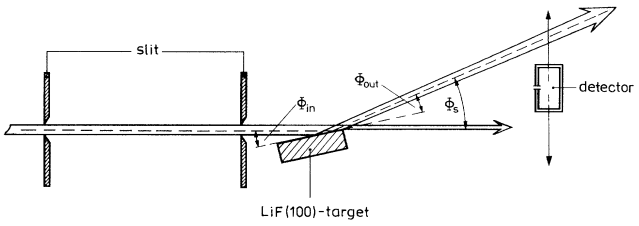

FIG. 1. Sketch of the experimental setup.

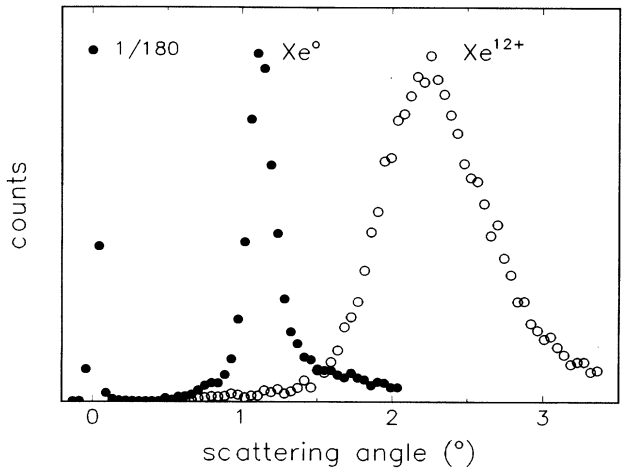

FIG. 2. Polar angular distributions for 50 keV Xe^0 (full circles) and Xe^{12+} (open circles) projectiles scattered from a clean and flat LiF(100) surface.

the energies observed for 50 keV Xe^{q+} ions for charges ranging from $q = 2$ to 15.

In former papers we have pointed out that image interaction energies comprise important information on the ion-surface interaction. Since these energies are lower limits for the approach towards the surface, experiments with multicharged ions at low energies have to take this feature into account. A number of data for metal surfaces show clear indications for this effect [3–7], and our work shows clearly that this effect also has to be taken into account for a LiF surface. In this respect we give support for corresponding corrections performed in recent studies of sputtering [12].

The second aspect of image interaction energies is related to the neutralization of the ions in front of the surface, since the image charge interaction depends strongly on the effective charge of the particle. This feature has been studied in some detail for the interaction of multicharged ions with metal surfaces, and the available data can be reproduced fairly well by a classical overbarrier model of stepwise neutralization of the incident ion. In particular, a $q^{3/2}$ dependence predicted by this simple model is in agreement with the experimental observations reported so far [6–11].

An important quantity in the overbarrier model is the distance R_q for the onset of electron transfer to an ion of charge q. This distance is estimated from an overbarrier criterion: Transfer of valence electrons to the ion is classically allowed, if the effective potential barrier for an "active" electron is lowered to energies of occupied electronic states. The effective one-electron potential in front of the surface is given by the Coulomb potential of the ion core and its image charge and by the electronic self-image interaction. In front of a metal surface one

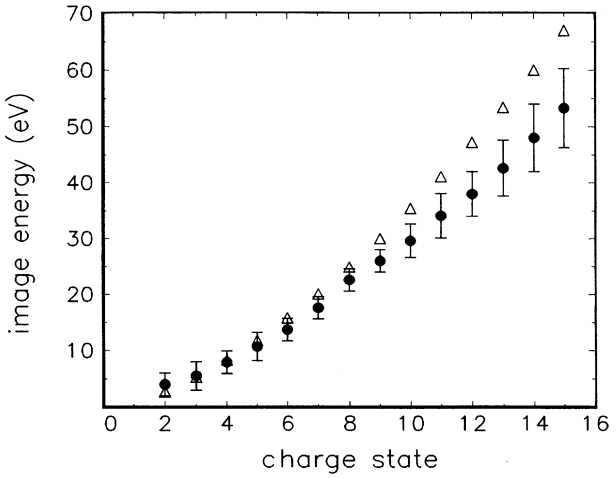

FIG. 3. Image interaction energies for the scattering of Xe^{q+} ions with a LiF(100) surface (full circles). The open triangles represent results obtained with a classical overbarrier model of stepwise neutralization in front of the surface.

finds, in good approximation, the simple relation $R_q = (2q)^{1/2}/W$, where W is the work function of the metal. Support for this classical estimate is given by Burgdörfer [17] from a discussion based on quantum-mechanical transition rates.

The energies displayed in Fig. 3 are similar to those found for metal surfaces. However, the electronic structure and the dielectric response of insulators are clearly different from metals. We adopt the overbarrier stair-case model [10] to the interaction of multicharged ions in front of a LiF surface. In contrast to a metal we have a broad energy gap, and the binding energies for the band of occupied electronic states (F $2p$) are clearly higher. A "work function" for F $2p$ electrons of about 12 eV can be found in the literature [16,18] where shifts of the band energies at the surface are a matter of speculation. Furthermore, the dielectric response of LiF clearly differs from that of a perfect conductor. The dynamic image potentials of the electron and ion core along the surface normal z are calculated for a projectile with velocity $\vec{v}_\parallel$ in local response from [19]

$$V_{\rm im}(z) = \frac{q}{2\pi}\int \frac{d\vec{Q}}{Q}\, e^{-Q(R+z)}\frac{1-\varepsilon(\omega)}{1+\varepsilon(\omega)}, \qquad (1)$$

where R is the position of the charge and $\varepsilon(\omega)$ is deduced from optical constants [20]; $\omega = \vec{Q}\cdot\vec{v}_\parallel$ and $\vec{Q}$ denotes the momentum $\vec{k}$ parallel to the surface $\vec{k} = (\vec{Q}, \vec{k}_z)$. In the limit $v_\parallel \to 0$ Eq. (1) gives

$$V_{\rm im}(z) = \frac{1-\varepsilon(0)}{1+\varepsilon(0)}\,\frac{q}{R+z}, \qquad (2)$$

so that for metals we have $[|\varepsilon(0)| \gg 0]$ $q/(R+z)$ and $-1/4R$ for the induced potentials due to the ion core and the electronic self-interaction. For LiF $\varepsilon(0) = 8.65$ is reported [20] so that the corresponding potentials are reduced by a factor of 0.79. A treatment of the overbarrier model for finite ε has been presented by Barany and Setterlind [21]. However, since $\varepsilon(0)$ for LiF is related to a low frequency resonance (38 meV), Eq. (2) is already a poor approximation for relatively low velocities. Towards optical frequencies $\varepsilon(\omega)$ drops to 1.92 so that, in general, the potential barrier shows a dependence on $v_\parallel$.

For illustration we show in Fig. 4 the effective potential for an active electron in the plane containing the surface normal and ion core ($q = 6$) at a distance of $R = 10.8$ a.u. from the electronic reference plane (positioned a few a.u. in front of the topmost layer of surface atoms) for $v_\parallel = 0.1$ a.u. The solid line represents the potential in front of LiF, the dashed line is for a perfect conductor, and the dotted line is the Coulomb potential of the ion core. From the figure it is evident that the effective barrier is lowered for a reduced image interaction so that R_q for a given electronic energy is larger for LiF than for a metal. R_q will be within the limits $\sqrt{2q}/E_B < R_q < q/E_B$, where the upper bound holds for vanishing response [22] and E_B denotes the minimal binding energy for valence band electrons. For LiF we assume $E_B = 12$ eV $= 0.44$ a.u. [18] so that $7.9 < R_q < 13.6$ a.u.; from calculations using Eq. (1) we obtain $R_q = 10.6$ a.u. (compare Fig. 4).

The finding that R_q is about one-third larger for LiF than for a metal is a general trend found in our calculations. Since, furthermore, the image potentials in the range of R_q for the velocity in these studies ($v_\parallel = 0.122$ a.u.) calculated from Eq. (1) are reduced to about 60% of the potential in front of metal surfaces, we expect for a given E_B image interaction energies $E_{\rm im}$ less than half of the size for a perfect conductor. The combined effects of larger R_q and reduced $V_{\rm im}$ on image interaction energies are

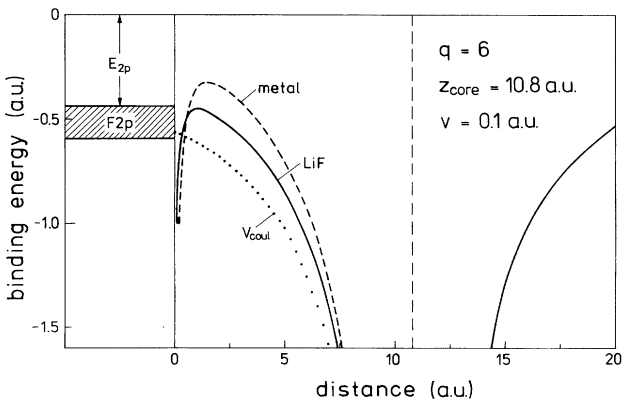

FIG. 4. Electronic potential for the interaction of an ion with core charge $q = 6$ at a distance of 10.8 a.u. in front of a metal (dashed line) and a LiF (solid line) surface. The dotted line represents the Coulomb potential $V_{\rm Coul}$ of the ion core. The potential for the LiF surface is obtained with help of Eq. (1) for $v_\parallel = 0.1$ a.u.

comparable to the ratio of E_B to work functions for metals and lead, despite the very different features of insulator and metal, to comparable E_{im}.

The overall agreement of the simple model (open triangles in Fig. 3) with the data is astonishingly good. However, we have to point out that some approximations (in particular, the electronic self-image interaction close to the image plane and the resulting potential barrier) are crude and need future more realistic treatments. In this respect it is also important to stress that in grazing collisions the projectiles interact with the target over rather large lateral extensions, so that in contrast to normal incidence eventual local processes could be shared over different sites in the surface plane.

In conclusion, we have reported the first observations of an image charge acceleration of multicharged ions in front of the surface of an insulator. The interpretation of the data implies the formation of hollow atoms on the incident path. We expect that the broad energy gap for the insulator (i.e., missing unoccupied electronic states above the valence band) will suppress the reionization of captured electrons. Then in contrast to metals electrons are expected to survive the original capture, until these loosely bound electrons forming the hollow atom are peeled off in close encounters with the surface.

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