Separation of Potential and Kinetic Electron Emission for Grazing Impact of Multiply Charged Ar Ions on a LiF(001) Surface

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Projectile time-of-flight spectra and the number of emitted electrons have been determined in coincidence for grazing scattering of slow (0.45 keV/u) multiply charged Ar ions from an atomically clean and flat LiF(001) surface. By relating projectile energy loss to kinetic electron emission we were able to determine contributions from potential electron emission even in the presence of a considerable number of kinetically excited electrons. Our results suggest a practically complete use of the available potential energy for electron emission during grazing scattering in sharp contrast to findings for the normal incidence case.

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The impact of slow ions (impact velocity < 1 a.u. $=$ 25 keV/u) on solid surfaces is of genuine interest in plasma and surface physics, and related applications. The nature and intensity of the resulting inelastic processes depend on both the kinetic and the potential (i.e., internal) ion energy carried toward the surface [1–6]. For slow multiply charged ions (MCI) this potential energy can become comparable to or even considerably exceed the ion kinetic energy, resulting in additional electron emission or sputtering [potential electron emission (PE) [4,5,7,8], potential sputtering [9–11]], processes which are usually dominated by kinetic effects [kinetic electron emission (KE) [1–4], kinetic sputtering [12,13]]. The relative importance of ion induced PE and KE from solid surfaces is not easy to determine. Measurements performed under grazing angles of incidence are here of particular interest, since then the projectiles' interaction with the surface proceeds along a well-defined trajectory (surface channeling [6]). More detailed information can be obtained if electron emission is observed in coincidence with the angular distribution of scattered projectiles. Recently, such measurements for multiply charged Ar ion impact on Au(111) have permitted a clear distinction of the contributions from PE and KE [14].

In this Letter we describe an extension of such investigations for an insulator target, where electronic properties (dielectric response, band gap, limited charge carrier mobility) may strongly affect the interaction scenario and add complexity to its theoretical description [15,16]. We have used a LiF(001) crystal as a prototype wide-band gap insulator. An earlier attempt to separate PE and KE via coincidence measurements of electron emission and the angular distribution of scattered projectiles as utilized by Lemell *et al.* [14] for a Au(111) surface turned out to be unfeasible [17]. This was mainly caused by the fact that KE from LiF starts at a much smaller impact energy (i.e., a lower KE threshold than for Au), increases more rapidly with projectile velocity, and decreases with incident ion charge state q (i.e., less emission for higher q) [18,19]. We have now developed a new technique, which makes use of the close relationship between kinetic electron emission and inelastic energy loss of the projectile ions, in order to separate the KE from the PE contribution.

A key feature of our setup is the coincident measurement of time of flight (TOF) for grazing projectile scattering from the LiF(001) surface with the number of emitted electrons for each scattering event by means of an electron statistics detector [14]. A chopped beam of multiply charged Ar^{q+} ions (charge state $q = 2$–8) produced by an electron cyclotron resonance ion source hits the LiF(001) surface under high index (''random'') azimuthal orientation with an angle of incidence below 5°. Ar projectiles scattered close to the specular direction (i.e., planar surface-channeled projectiles) are recorded by means of a position-sensitive multichannel plate detector (MCP) equipped with a two-dimensional wedge and strip anode situated further down stream. Electrons emitted from the LiF surface are collected by a weak electric field, imposed by a highly transparent grid about 1 cm in front of the target. This grid shields the adjacent high electric field from a surface barrier detector (SBD) at $+25$ kV which accelerates the extracted electrons, resulting in detector pulse height distributions from which the number of electrons ejected per projectile impact on the surface is derived. The detection efficiency is close to 100% for all electrons emitted with a kinetic energy of <50 eV into the half solid angle [14]. These ''low-energy'' electrons make up practically 100% of the total electron yield for impact of Ar^{q+} up to $q = 8$ and more than 98% for impact of Ar^{9+}. The cleaved LiF(001) single crystal surface is kept at a base pressure in the mid 10^{-10} mbar range and prepared in situ by annealing at about 400 °C for typically 90 min. During the measurements the target is kept at a temperature of about 200 °C, where the ionic conductance of LiF is

high enough to prevent macroscopic charging up of the sample, while the microscopic situation during single impact events (e.g., local charging via electron extraction from the surface) is not yet affected. In our TOF setup, the MCP signal serves as "stop" pulse for a time-to-amplitude converter, while the "start" is obtained from the beam chopper. The beam chopper was operated at a variable frequency between 100 and 250 kHz to adjust the count rate at the MCP detector to typically 1000 projectiles per second. Therefore on the average less than 0.01 projectiles pass through the aperture in a single chopper cycle. A multiparameter analog-to-digital converter system digitizes the signals from the TOF electronics, the SBD as well as the position information from the wedge and strip anode and stores them in list mode for data evaluation. In a nonlinear transformation the TOF information is converted into an energy loss scale with the recorded impact positions on the MCP serving for correction of different flight paths.

In this way we measure for individual trajectories the projectile energy loss during grazing scattering from the surface in coincidence with the number of emitted electrons. A typical coincidence spectrum for 18 keV Ar^{3+} ion impact on clean LiF(001) under 3.8° angle of incidence is shown in Fig. 1(a). As a general trend we note that the mean number of emitted electrons increases with increasing energy loss. For further analysis mean values of electron number and projectile energy loss have been evaluated for certain cuts through such coincidence spectra. Cuts at constant energy loss (as indicated in Fig. 1(a) for the cases of 1, 2, or 3 keV energy loss) yield the respective mean number of emitted electrons [Fig. 1(b)]. Extrapolation of this curve in Fig. 1(b) to the hypothetical case of projectiles

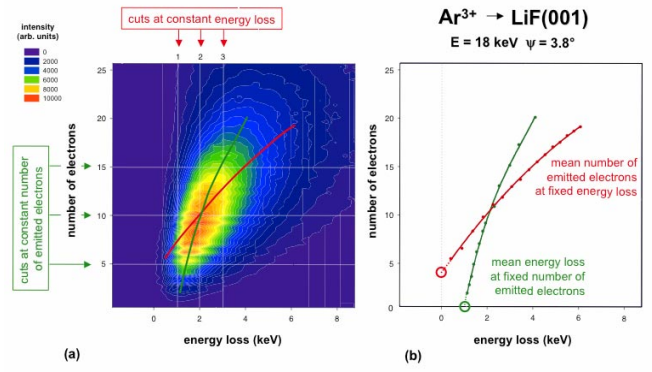

FIG. 1 (color online). (a) Coincidence spectrum for the number of emitted electrons vs projectile energy loss for 18 keV Ar^{3+} ions impinging on a clean LiF(001) surface under a grazing angle of incidence of 3.8°. (b) Cuts through the coincidence spectrum at constant energy loss provide the mean number of emitted electrons for a particular energy loss of the projectiles (red/black curve), while cuts for a given number of emitted electrons lead to the related mean energy losses (green/gray curve). The curves can be extrapolated to zero energy loss (red/black circle) and zero number of emitted electrons (green/gray circle), respectively (for further details cf. text).

with zero energy loss (actually not observable in our experiment) gives the electron emission contribution, which is not associated with kinetic energy loss of the projectile (indicated by a red/black circle in Fig. 1(b)]. Since these electrons cannot have been emitted at the expense of the projectile's kinetic energy, they must result from the projectile's potential energy, i.e., constitute the "pure" potential electron emission yield $\gamma_{\mathrm{PE}}(\Delta E \rightarrow 0)$.

Plotting the extrapolated $\gamma_{\mathrm{PE}}(\Delta E \rightarrow 0)$ values for different Ar^{q+} projectiles as a function of the potential energy of the ions (i.e., the sum of all ionization potentials for producing an ion with a given charge q) supports this interpretation. In Fig. 2 we find a linear relationship between the pure PE yields and the potential energy brought towards the surface by the different Ar^{q+} projectile ions, while no dependence on impact energy (varied between 18 and 54 keV) could be observed within our experimental errors. Moreover, our data points lie right at the border allowed by energy conservation (shaded triangular region in Fig. 2). As shown by Hagstrum [7,8] PE is due to Auger processes such as, e.g., Auger neutralization, resonant capture followed by Auger deexcitation, or multiple resonant capture followed by autoionization [5,20]. All these processes require a minimum potential energy of at least twice the binding energy W_ϕ of the highest occupied state of the solid (which in the case of metal targets corresponds to the work function). The maximum possible number of electrons n_{max} emitted via PE is therefore given by

$$n_{\mathrm{max}} = E_{\mathrm{pot}}/(2W_\phi). \quad (1)$$

Surprisingly, this maximum possible number of PE electrons is actually obtained from our experimental data, taking into account a binding energy of about 12 eV [21] for the highest occupied states in the $F^-(2p)$ valence band of LiF (solid line in Fig. 2).

This remarkable finding suggests that up to the highest ion charge state/potential energy which we have applied

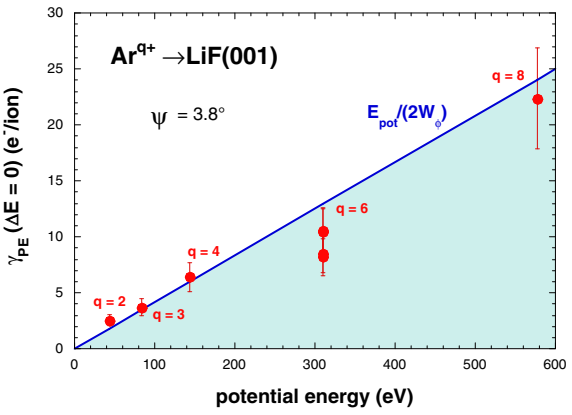

FIG. 2 (color online). Pure potential electron emission contribution (cf. text) as a function of available potential energy (full circles) as compared to model predictions (solid line).

(Ar^{8+}, $E_{pot} \approx 580$ eV), the electronic properties of the alkali halide (limited hole mobility, possible reduction of the electron capture rate due to hole formation, and eventually necessary capture of more tightly bound electrons, see, e.g., [16]) impose no limitations on its ability to provide sufficient electrons for complete neutralization and deexcitation within the (limited) surface interaction time. We note that these PE yields for grazing Ar^{q+} impact are by more than a factor of 2 larger than PE yields by Vana *et al.* [19] for normal incident Ar^{q+} on LiF. It is, however, important to stress that in grazing collisions the projectiles interact with many different (F^-) sites over a rather large lateral extension. In contrast to that Wirtz *et al.* [16] have shown that for normal impact on a LiF surface the rates for capture from neighboring sites are considerably (typically 1 order of magnitude) smaller than for capture from the closest fluorine atom directly ''underneath'' the projectile. Further neutralization from the same site is also less probable because it means capture from a more tightly bound electron. Our findings are also consistent with observations on the image charge acceleration of multiply charged ions in grazing scattering on a LiF surface [22]. There the interaction energies gained by the projectiles also point to a complete projectile neutralization along the grazing scattering flight path.

The much smaller PE efficiency for normal incident projectiles suggests a noncomplete deexcitation and maybe even an incomplete neutralization of the multiply charged Ar projectiles during their above-surface interaction phase. This again raises the question about the existence of a so-called "trampoline" effect [16,23], in which (non-neutralized) projectiles under certain circumstances might be repelled by the positive hole charges on the surface. If this trampoline effect would exist a pure electronic interaction of the projectile ion with the target without any significant momentum transfer to individual target nuclei would be possible. This offers important implications for using slow highly charged ions as a tool for gentle nanostructuring of surfaces [5,11].

Since surface-channeled projectiles interact with the surface along well-defined and calculable trajectories [6], the technique presented here in principle also allows investigation of PE yields as a function of the closest distance of projectile approach toward the surface. For not too high ion charge states this could be an alternate way to determine distance dependent Auger rates [24–26].

Returning again to Fig. 1, we now analyze cuts for a constant number of emitted electrons. The mean energy loss associated with the emission of a particular number of emitted electrons (green/gray curve) can be extrapolated to the hypothetical case of zero electron emission (indicated as a green/gray circle in Fig. 1(b)]. This pure energy loss ΔE, which has not led to emission of electrons, is found to rise linearly with impact energy (see Fig. 3)

$$\Delta E \sim E_{\mathrm{kin}} \tag{2}$$

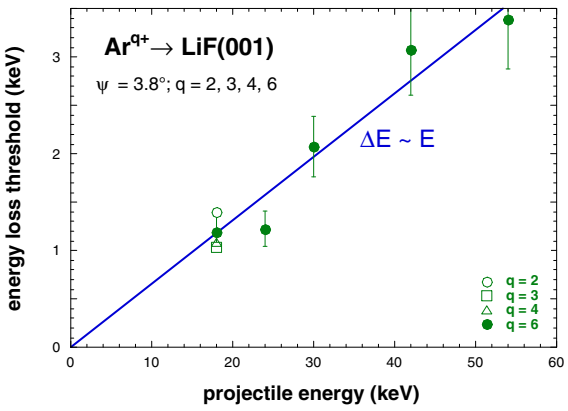

FIG. 3 (color online). Pure energy loss (cf. text) as a function of projectile impact energy.

while its dependence on the ion charge state is quite weak. For grazing scattering of protons from LiF(001), Auth *et al.* [27] have already found such a linear dependence of the projectile energy loss on incident ion energy. They assumed a stopping power $S(z)$ which exponentially decreases with increasing ion-surface distance z

$$S(z) = -dE/dx(z) = S_0(E_{kin}) \exp(-z/z_0) \tag{3}$$

and integrated the energy loss along the ion trajectory to obtain

$$\Delta E \sim S_0(E_{\mathrm{kin}}) {E_{\mathrm{kin}}}^{1/2}. \tag{4}$$

The linear dependence of pure energy loss on kinetic projectile energy shown in Fig. 3 is therefore consistent with a velocity-proportional stopping power

$$S_0(E_{\mathrm{kin}}) \sim v \sim {E_{\mathrm{kin}}}^{1/2}. \tag{5}$$

Such a linear v dependence is well established for metallic targets at low projectile velocity ($v \leq 1$ a.u.) [28], but surprisingly was also observed for an insulating LiF target in both transmission [29] and scattering experiments [27] with protons as projectiles. Coincidence measurements for grazing scattering of keV protons [30] and for hydrogen and helium atoms on LiF(001) have recently revealed that the principal energy loss mechanism is caused by excitation of surface excitons, as shown by well separated equidistant peaks in the energy loss spectra in cases where no electron is emitted [30,31].

In conclusion, we have applied a coincidence technique to study electron emission from the interaction of slow multiply charged Ar ions with an atomically clean and flat LiF(001) surface under grazing scattering conditions. Since kinetic electron emission is always associated with kinetic energy loss, we can determine contributions from potential electron emission even in cases where the majority of ejected electrons results from kinetic emission. Surprisingly, the such obtained PE yields agree with an almost complete conversion of the potential energy into

electron emission via Auger type processes, indicating complete neutralization and deexcitation of the projectiles within the available above-surface interaction time. Considerably smaller PE yields for perpendicular MCI impact on LiF suggest a reinvestigation of the so-called trampoline effect. Moreover, our technique presented here permits the determination of PE yields in dependence of the closest distance of projectile approach toward the surface as long as not too high ion charge states are involved.

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