Electron Emission from Insulators Irradiated by Slow Highly Charged Ions

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Total electron emission yields have been measured for the first time resulting from impact of slow highly charged Ar\textsuperscript{q+} (q ≤ 17), Xe\textsuperscript{q+} (q ≤ 50) and Hg\textsuperscript{q+} (q ≤ 68) ions on clean insulating LiF(001) and CaF\textsubscript{2}(111) surfaces at various impact angles. The surprisingly large yields show that even for the highest projectile charge states, a local charge-up of the surface poses no barrier for electron emission. We demonstrate that this is due to a strong sub-surface contribution in the potential electron emission process which is considerably more efficient in insulators because of the increased inelastic mean free path and the production of secondary electrons.

I. INTRODUCTION

The interaction of slow highly charged ions (HCI) with solid surfaces results in the formation of “hollow atoms” [1–6], an exotic atomic state far from equilibrium. Whereas electronic transitions which eventually restore the equilibrium could largely be identified for metal surfaces in experimental and theoretical studies during the last decade [3, 5, 6], similar processes at insulator surfaces are much less understood [7, 8]. The main differences as compared to HCI-metal interactions are (i) the different dielectric response of insulating surfaces (modified image charge acceleration, reduced hole mobility, etc.), (ii) local charging-up of the surface (leading to a partial repulsion of the projectile by positive hole charges on the surface and a possible reduction in the number of emitted electrons), and (iii) the wide band gap (altering transfer, excitation and emission of electrons). In principle, electron emission is caused by deposition of both kinetic and potential (= internal) energy of the projectile ions. For slow HCI, the potential electron emission (PE) component [5, 7, 9–11] usually dominates over kinetic electron emission (KE) [11–14].

In order to study the response of an insulator surface to the strong perturbation introduced by a slowly approaching highly charged ion, electron emission experiments have been carried out at the Max Planck Institute of Nuclear Physics in Heidelberg/Germany for highly charged ions using a specially designed setup built by TU Wien/Austria. In this contribution we report on recent results for the impact velocity and impact angle dependent electron emission from different insulator target surfaces (LiF and CaF\textsubscript{2}) during bombardment with slow HCI. Accompanying simulations for electron emission and transport aid the analysis and interpretation of these experimental results.

II. EXPERIMENTAL METHOD

The highly charged ions (HCI) used for the present study were extracted from the Heidelberg electron beam ion trap (EBIT). Details of this source can be found elsewhere [15]. Electron beam intensities of the EBIT were kept between 300 and 350 mA and the electron gun was biased negatively between 8 and 12 kV in order to allow production of He-like Xe\textsuperscript{52+} even at a drift tube bias of only 3 kV. The electron beam is compressed by an 8 T magnetic field to about 50 µm diameter. The corresponding space charge potential was calculated and included in the calculation of the ion beam velocity. Isotope-pure \textsuperscript{40}Ar and \textsuperscript{129}Xe gases were injected to produce the highly charged ions, while mercury was injected exploiting its finite gas pressure at room temperature. Since the mercury was not isotope-pure, there is an intrinsic uncertainty of its charge state of ±1. The EBIT was operated in leaky mode (also known as dc mode), where the produced ions are stored in the trap by a confining axial potential applied to a set of drift tubes. The drift tube facing the beamline was kept at a lower potential than the one facing the electron gun, so that HCI, after reaching the appropriate temperature via collisions with the electron beam, could escape the trap. Since they pass the confining drift tube with, ideally, zero kinetic energy, this potential defines their final kinetic energy when they hit the grounded target. After escaping the trap, the ions passed an electrostatic lens and a set of deflection plates, which were used to focus the beam on the entrance slits of the 90° analyzing mag-

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After the magnet the beam was again focused by a quadrupole triplet and entered a switching magnet, which steered the beam into one of the beamlines. The beam was subsequently focused and steered by a very compact segmented Einzel lens, recently termed “Sikler lens” [16], so that it finally entered the experimental chamber via a circular beam shape defining aperture with 1 mm diameter, followed by a somewhat larger negatively biased electron repeller aperture (Fig. 1(a)). Our experimental chamber included a target holder which allowed X, Y, Z, and θ manipulation of the target, where θ is the ion angle of incidence with respect to the target surface normal and could be adjusted with an accuracy of 1°. The insulating CaF$_2$(111) and LiF(001) single crystal targets were freshly cleaved in air and mounted on the target holder with elastic steel clips to ensure good thermal conductivity. The targets were heated inside the vacuum chamber via a coaxial heating element, where 10 W of heating power was sufficient to reach target temperatures of more than 400°C. Elevated temperatures were repeatedly applied to clean the target surface between measurements.

With the target at room temperature, significant charging of the surface was observed even for the very small ion fluxes obtained from the EBIT (typically 10$^5$–10$^4$ ions/sec). Figure 2(a) shows the reduction in electron yield over time due to a charging-up of the surface even at a primary ion current of less than 1 pA. To counter this effect, both targets were heated during experiments. As seen in Fig. 2(b), a surface temperature of 180°C in the case of LiF and 60°C in the case of CaF$_2$ is sufficient to eliminate any macroscopic charging-up by the onset of ionic conductance leading to the restoration of the “original” electron yield. Between any two ion impact events (typically every ms) the surface can relax to a discharged state while it still behaves like an insulator during the impact of one individual ion (timescale typically 10-100 fs).

Base pressure of our chamber and the EBIT beamline was below 3 × 10$^{-10}$ mbar.

To detect the number of electrons from each ion impact, we used a slightly modified version of the electron statistics setup described by Lemell et al. [17]. The same setup was recently used to determine electron emission yields from a conducting Au(111) surface [18]. Electrons emitted from the surface were collected by a highly transparent grid and afterwards accelerated onto the active surface of a surface barrier detector biased at 27 kV (see Figs. 1(a) and 1(b)). Electron emission induced by single projectiles terminates within a time interval of well below 1 ps after the collision, much shorter than the time resolution of the applied detector electronics. Thus, n electrons emitted during a single ion impact and accelerated towards the surface barrier detector are registered like one electron of n × 27 keV rather than n individual 27 keV electrons. The number of electrons emitted for a particular ion impact event can therefore be evaluated from the detector pulse height distributions. More details on the electron statistics detection method and the appropriate correction of measured pulse height spectra can be found in [17, 19] and references therein.

The detector was mounted horizontally at 90° with respect to the ion beam direction, so that for impact angles below 45° electrons were emitted towards the grid and the collection efficiency was close to 100%. For impact angles below 45° the negative bias of the electron repeller electrode assisted the deflection of electrons (see Fig. 1(b)), but still the collection efficiency was reduced. This effect had to be accounted for, using calculated collection efficiencies for different impact angles obtained with the SIMION package (V. 8.0) which were verified experimentally using the Au target [18]. We have double-checked the angular dependence of the obtained yields by alternatively mounting the detector vertically below the target holder, along the axis of rotation of the target. The collection efficiency of this setup was always lower than in the original geometry, but not dependent on the ion impact angle.
anymore. The angular dependence of electron yields obtained in this way was found to be qualitatively the same as that of the collection-efficiency corrected yields using the original setup described above.

III. RESULTS AND DISCUSSIONS

Figure 3 shows total electron yields from clean single crystal LiF(001) and CaF$_2$(111) targets for normal incidence of highly charged Ar$^{q+}$ ($q \leq 17$), Xe$^{q+}$ ($q \leq 50$) and Hg$^{q+}$ ($q \leq 68$) ions at a constant impact velocity of $7 \times 10^5$ ms$^{-1}$, compared to yields obtained for a clean conducting Au(111) surface. We observe a clear dependence of the yield on the charge state of the incident ion, as would be expected for the dominant potential emission process. Surprisingly, electron yields from the CaF$_2$ surface are comparable to that from the conducting gold surface, which has a considerably lower work function, while the total electron yields from the LiF surface are even higher than that. Even more surprising, the yields show no saturation at higher charge states, which could be expected if one assumes that the impact zone is charged up microscopically. The extraction of electrons by the approaching HCl would then make the emission of additional electrons on a ps timescale more difficult.

Figure 4 shows total electron yields from a freshly cleaved LiF surface for impact of highly charged Ar$^{q+}$ ($q \leq 17$) and Xe$^{q+}$ ($q \leq 50$) ions. While the impact velocity was kept constant for all charge states, impact angles were varied between 10° and 75° with respect to the surface normal. Kinetic electron emission (KE) yields for the present velocity regime of $7 \times 10^5$ ms$^{-1}$ have been measured for normal impact of Ar$^{q+}$ on LiF [20] as 25 electrons per ion, and should be somewhat higher for the heavier Xe ions. Still, this is up to an order of magnitude smaller than the total yields we have obtained. Kinetic emission yields are long known to vary with the incident ion’s impact angle according to an inverse cosine law [21]. Indeed, the total electron yields obtained for the insulating targets can be well fitted by using an inverse cosine law and a constant offset, as seen in Fig. 4.

$$\gamma(\theta) = \gamma_0 + \frac{c}{\cos \theta}.$$  \hspace{1cm} (1)

It is, furthermore, observed that the angular dependent part of the yield $c$ is almost independent of the primary ionic charge state $q$, while the constant part $\gamma_0$ shows a very strong charge state dependence. On the other hand, the angular dependent part varies strongly with the projectile velocity. This lead us to conclude that the angular dependence of the electron yields obtained for LiF is primarily due to KE. This conclusion is also supported by Fig. 5, which shows electron yields for Xe$^{44+}$ impact at several impact velocities and under various impact angles as a function of the normal component of the impact velocity. We have previously observed that for a gold target and pure PE yields, all data points can be fitted with a single curve in that way [18]. Yet for the insulating LiF target, this procedure does not work at all. If, however, we assume that the angular dependence is mostly due to KE and therefore dependent on the impact velocity $v$, we can fit the parameter $c$ in formula (1) by a linear regression vs. $v$. We can subtract this KE contribution from each of the total electron yields obtained according to an inverse cosine law. The remaining PE data points can then be fitted by a single curve (Fig. 5) and are almost independent of the impact velocity. This is in strong contrast to the case of the conducting Au surface where a decrease of the PE yield with increasing velocity was found [18, 22].

$$\gamma_{\text{PE}}(v, \theta) = \gamma_\infty + \frac{c}{\sqrt{v \cos \theta}}.$$  \hspace{1cm} (2)

A more direct way to eliminate any contribution of KE in our total yields is looking at the difference between yields obtained for two charge states at the same impact velocity. Figure 6a shows results obtained for Xe$^{51+}$ and Xe$^{13+}$ at the same impact velocity of $4.5 \times 10^5$ ms$^{-1}$, and the difference between them. The subtraction of Xe$^{13+}$ yields from the Xe$^{51+}$ results undesirably eliminates a part of the PE yield (although the potential energy of
FIG. 5: All measured electron yields for Xe$^{4+}$ impact on LiF for various impact angles (10°-70°) and impact velocities (from 2.2 keV/amu to 4.2 keV/amu) as a function of the velocity component normal to the surface. Opposite to the case of a conducting Au surface (c.f. text), the data points for LiF (●) are only aligned when measured at the same impact angle (connected by linear fits). After subtraction of the relevant kinetic electron emission contributions (c.f. text), all data points ( ●) end up on a single curve, which shows a very weak dependence on the impact velocity. Due to the uncertainties in the subtraction procedure, error bars for the corrected yields are larger.

Xe$^{13+}$ is only 1.6 keV, compared to 111 keV for Xe$^{31+}$), so the remaining yield is just the “excess” PE yield of Xe$^{51+}$ as compared to Xe$^{13+}$ on CaF$_2$. The contribution from KE, however, is fully eliminated as it only depends on the primary impact velocity of the ion. Image charge acceleration is negligible compared to the primary kinetic energy of more than 100 keV as it would amount to less than 500 eV even for Xe$^{50+}$ impact on a conducting surface [23]. The remaining yield thereby qualitatively represents pure PE. As can be seen in Fig. 6b, these “excess” PE yields do not (or only very weakly) depend on the impact angle of the primary ion, both for LiF and for CaF$_2$ targets and for different projectile charge states. These results support the conclusions drawn from Xe$^{4+}$ results (see Fig. 5) that the PE yields for these insulating targets do not strongly depend on the impact angle or on the component of the impact velocity normal to the surface, as in the case of a metal surface. For the CaF$_2$ surface, and also for the LiF surface at higher impact velocities, a slight decrease of the electron yield is observed with increasing impact angle.

Our results therefore not only demonstrate the surprisingly large values of total and also purely potential electron emission yields for insulating targets, but also show drastic differences in the dependence of the PE yields on impact angle when compared to a conducting surface. To explore the reason behind these differences, we have performed Monte Carlo simulations based on the classical-over-the-barrier (COB) model [2] modified for insulators [24] to study potential electron emission during the approach of the ion to the surface. For insulating targets, the electron capture probabilities depend on the history of the impact process (local charges) and on the position of the HCI with respect to the lattice, so these simulations proved to be much more demanding. We have hence only performed these simulations for the comparably simple case of Ar$^{11+}$ and Ar$^{17+}$ colliding with a LiF and a Au surface. The calculations clearly show that the above surface part of the electron emission process, which is responsible for a large fraction of the total electron yield for conducting Au surfaces [18, 22], plays a much less important role for insulators. While the measurements yield a (potential) electron emission of 38 e$^-$/ion for Ar$^{11+}$ and 66 e$^-$/ion for Ar$^{17+}$ impact on LiF, the simulation of the above-surface part results only in 3.5 and 9.0 e$^-$/ion, respectively. Due to the increased binding energy of valence electrons in LiF (12 eV) compared to those of the conduction band in gold (5 eV), the hollow atom (HA) above a LiF surface is formed at a smaller distance and much less time is available for any deexcitation processes above the surface (see Fig. 7). Potential electron emission is therefore dominated by sub-surface emission processes, at least for the present velocity regime.

Every (Auger) electron produced inside the target undergoes elastic and inelastic scattering events, thereby ini-

FIG. 6: (a) Subtraction of Xe$^{13+}$ yields from Xe$^{31+}$ yields measured at the same impact velocity cancels out any contribution of kinetic electron emission, showing the qualitative impact-angle dependence of pure potential emission from insulators. (b) The remaining PE yields show a very weak dependence on the projectile angle of impact for both LiF and CaF$_2$ targets and various charge states.

http://www.sssj.org/ejssnt (J-Stage: http://www.jstage.jst.go.jp/browse/ejssnt/)
FIG. 8: Calculated secondary electron yields for primary electrons generated with different energies at a certain depth $z$ beneath the surface of a conducting Au(111) and an insulating LiF(001) surface. Due to the increased inelastic mean free path especially for slow electrons in the insulator, secondary electrons have a much better chance to escape into vacuum.

The surprisingly high electron yields from the insulating LiF surface therefore are caused by (c.f. Fig. 7): (i) The delayed electron capture and deexcitation leaving more potential energy stored in the HA at the time of impact and (ii) the sub-surface decay via Auger emission leading to secondary electrons which have a higher chance of escaping the surface due to the larger mean free path (much in the same way as kinetic energy deposited in insulators leads to higher KE yields as compared to the conducting targets). Auger decay of the HA formed inside the solid after surface penetration will be completed within a few nm [26, 27], which in the case of LiF and CaF$_2$ is comparable to the mean escape depth $\lambda$ of the produced electrons. This explains why the sub-surface PE yield for LiF does only slightly change with impact angle or impact velocity. For increasing impact angles (at constant impact velocity) the time spent by the projectile between the first electron capture and its impact increases and thus also the above surface part of the potential electron emission gets larger. This, however, leads to the emission of fewer (but more energetic) electrons with a reduced probability for electron multiplication below the surface (in about 50% of the cases their trajectories are completely outside of the solid). Together with an increased probability for projectile reflection, a reduction of the PE electron yield at grazing impact angles is therefore at least qualitatively plausible.

IV. CONCLUSIONS

We have obtained total electron yields for HCl impact on clean LiF(001) and CaF$_2$(111) surfaces using an electron statistics detector. With primary ion charge states of up to 68$^+$, we have found surprisingly large yields from the insulating targets. By subtracting kinetic electron emission we have shown that the potential electron emission yields from these surfaces do not strongly depend on impact angle or impact velocity in the present regime of 4 up to $9 \times 10^5$ ms$^{-1}$. These findings are explained by a strong contribution of sub-surface electron emission, where each of the Auger electrons emitted during the deexcitation of the hollow atom can create a cascade of secondary electrons, which have a high probability to escape into vacuum due to the increased mean free path in LiF and CaF$_2$.

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[1] J. P. Briand, L. de Billy, P. Charles, S. Essabaa, P. Briand, R. Geller, J. P. Desclaux, S. Bliman, and C. Ristori, Phys. Rev. Lett. 65, 159 (1990).
[2] J. Burgdörfer, P. Lerner, and F. W. Meyer, Phys. Rev. A 44, 5674 (1991).
[3] J. Burgdörfer, in Fundamental Processes and Applications of Atoms and Ions, edited by C. D. Lin (World Scientific, Singapore, 1993).
[4] R. Morgenstern, and J. Das, Europhysics News 25, 3 (1994).
[5] A. Arnau, F. Aumayr, P. M. Echenique, M. Grether et al., Surf. Sci. Reports 27, 113 (1997).
[6] HP. Winter and F. Aumayr, J. Phys. B: At. Mol. Opt. Phys. 32, R39 (1999).
[7] F. Aumayr and HP. Winter, Springer Tracts in Modern Physics 225, 79 (2007).
[8] J. Burgdörfer and C. Lemell, Springer Tracts in Modern Physics 225, 1 (2007).
[9] H. D. Hagstrum, Phys. Rev. 96, 325 (1954).
[10] H. D. Hagstrum, Phys. Rev. 96, 336 (1954).
[11] R. Baragiola, in Chap. IV in Low energy Ion-Surface Interactions, edited by J. W. Rabalais (Wiley, 1993).
[12] D. Hasselkamp, in Particle Induced Electron Emission II, edited by G. Höhler (Springer, Heidelberg, 1992), pp. 1.
[13] J. Schou, Scanning Microsc. 2, 607 (1988).
[14] M. Rössler and W. Brauer, in Particle Induced Electron Emission I, edited by G. Höhler (Springer, Berlin, 1991).
[15] J. R. Crespo López-Urrutia, B. Bapat, B. Feuerstein, D. Fischer, H. Löch, R. Moshammer, and J. Ullrich, Hyperfine Interactions 146/147, 109 (2003).
[16] A. Lapierre, private communication (2007).
[17] C. Lemell, J. Stockl, H. P. Winter, and F. Aumayr, Rev. Sci. Instrum. 70, 1653 (1999).
[18] W. Meissl, M. C. Simon, J. R. Crespo López-Urrutia, H. Tawara, J. Ullrich, HP. Winter, and F. Aumayr, Nucl. Instrum. Meth. Phys. Res. B 256, 520 (2007).
[19] F. Aumayr, G. Lakits, and HP. Winter, Appl. Surf. Sci. 47, 139 (1991).
[20] M. Vana, F. Aumayr, P. Varga, and HP. Winter, Nucl. Instrum. Meth. Phys. Res. B 100, 284 (1995).
[21] L. A. Dietz and J. C. Sheffield, Rev. Sci. Instrum. 44, 183 (1973).
[22] H. Kurz, F. Aumayr, D. Schneider, M. A. Briere, J. W. McDonald, and HP. Winter, Phys. Rev. A 49, 4693 (1994).
[23] F. Aumayr, H. Kurz, D. Schneider, M. A. Briere, J. W. McDonald, C. E. Cunningham, and HP. Winter, Phys. Rev. Lett. 71, 1943 (1993).
[24] L. Wirtz, C. O. Reinhold, C. Lemell, and J. Burgdörfer, Phys. Rev. A 67, 12903 (2003).
[25] K. Tökösi, D. Varga, L. Kövér, and Mukoyama, Spectr. Rel. Phenom. 76 (1995).
[26] M. Hattass, T. Schenkel, A. V. Hamza, A. V. Barnes, M. W. Newman, J. W. McDonald, T. R. Niedermayr, G. A. Machicoane, and D. H. Schneider, Phys. Rev. Lett. 82, 4795 (1999).
[27] C. Lemell, A. S. El-Said, W. Meissl, I. C. Gebeshuber, C. Trautmann, M. Toulouse, J. Burgdörfer, and F. Aumayr, Solid State Electronics 51, 1398 (2007).