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Above-surface potential sputtering of protons by highly charged ions

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We present a description for potential-energy-induced sputtering of protons by slow, highly charged ions in front of insulator surfaces, based on the classical over-barrier model. Proton emission results from the localized Coulomb explosion after breaking the covalent C-H bond in hydrocarbons at the surface. The model describes the very steep dependence on the projectile charge ($\sim Q^4 - Q^6$) for moderately high charge states ($Q \lesssim 10$), in agreement with the experiment. It furthermore predicts the saturation of the Q dependence for even higher charge states. [S1050-2947(96)06510-9]

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I. INTRODUCTION

The impact of a charged particle with a surface leads to the emission of secondary ions and neutral particles from the surface. Two basic mechanisms for sputtering of target atoms can be distinguished: ‘‘kinetic’’ sputtering, which results from the transfer of kinetic energy from the projectile to target atoms in the surface in a binary collision sequence, and ‘‘potential’’ sputtering, which results from the dissipation of the potential energy carried into the collision by the projectile and the subsequent conversion into the kinetic energy of target atoms. For the surface interaction involving very slow highly charged ions, available from electron cyclotron resonance and electron beam ion sources, with surfaces, the second mechanism is generally believed to be dominant.

Pioneering experimental studies of the charge (Q) dependence of the secondary-ion yield were performed by Arifov *et al.* [1] and Bitenskii, Murakhmetov, and Parilis [2]. They found the yield to display a significant charge (Q) dependence for nonmetals in contrast to metals. An intriguing Coulomb explosion model to explain these findings was put forth by Parilis [3]. According to this model, multiple charge transfer to the projectile leads to a strong local charging of the impact region. As ‘‘rushing in’’ of the screening charge and reneutralization is suppressed in insulators, ionized target atoms are ejected due to the mutual Coulomb repulsion. This model has been used to fit several more recent experimental data [4–8], most of them taken in an energy regime where kinetic sputtering significantly contributes.

Very recent measurements at low energies performed on LiF as a prototype insulator have sharpened and, to some extent, modified this picture [9,10]: It was found that the overwhelming fraction of sputtered particles is neutral rather than charged, thereby ruling out the Coulomb explosion as the dominant mechanism. The dominance of neutral particles has been previously observed also for semiconductors [4]. Furthermore, the yield of neutral particles was found to be proportional to the potential energy of the incident ion (or approximately proportional to Q^2). These observations were interpreted in terms of multiple electronic defect production (‘‘color centers’’) induced by sequential resonant charge transfer to the projectile in close analogy to electron and photon stimulated desorption (ESD and PSD) [11,12]. At the same time, a very small component of ionized sputtered particles was identified, the yield of which displayed a stronger Q dependence. The latter could possibly be interpreted in terms of a Coulomb explosion, however, as a subdominant process.

Studies of sputtering of protons originating from impurities or the deposition of hydrocarbons at the surface display a remarkably strong Q dependence, ranging from $\sim Q^3$ in the kinetic sputtering regime [5,8] to $\sim Q^5 - Q^6$ in the pure potential-energy regime [13,14]. Equally remarkable is the high absolute yield of the order of one proton per incident highly charged ion with $Q \simeq 20$, which surpasses corresponding yields of other ionized fragments (e.g., F^+ or Li^+ for LiF) by orders of magnitude. The extreme sensitivity of the proton sputtering to the charge state of the projectile could be of importance for surface diagnostics of ‘‘real’’ (i.e., dirty) surfaces for technological applications [13,14] as well as for organic and biological materials since this sputtering mechanism appears to be rather insensitive to specific properties of the insulator surface.

In the following, we present a simple model analyzing sputtering of protons from an insulator surface consisting of a deposited layer of hydrocarbons of sufficient thickness such that the properties of the substrate are not important. The model employs the classical over-barrier model for resonant neutralization of highly charged ions [15,16]. It can be viewed as a variant of the Coulomb explosion model, which focuses, however, on the above-surface Coulomb interaction of the highly charged ion rather than its energy deposition in the surface. As there are no detailed data available on the properties of these surfaces, we aim here at a semiquantitative description and focus on the exploration of the surprisingly strong and heretofore unexplained Q dependence in the potential sputtering regime.

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II. THEORY

The theoretical description of proton sputtering is based on the following experimental observations [14,17]:

(a) The sputtering yield and its Q dependence is largely independent of the substrate as long as the surface is covered by a few monolayers of hydrocarbons. The approximate stoichiometric composition for deposited hydrocarbons is typically C_nH_{2n} for large molecules.

(b) The energy distribution of sputtered protons is highly nonthermal with a peak at $E_k \approx 10$ eV after correction of the projected distribution for the angular distribution relative to surface normal. This energy is consistent with the Coulomb explosion energy after breaking of the covalent C—H bond between the C and H atoms,

$$E_k \simeq 1/d_0, \tag{1}$$

where $d_0 \simeq 2$ a.u. is the bond length prior to breakup. Because of the asymmetric mass ratio $\mu_H \ll \mu_C$, almost all of the Coulomb explosion energy is carried away by the proton. Consequently, proton sputtering after covalent bond breaking is a comparatively fast, energetic process compared to the emission of heavier ions from a charged-up lattice, which may explain in part its high efficiency.

(c) The observed Q dependence is largely uncorrelated with the potential energy. For example, the gap in potential energy between $Q=8$ and 9 of Ar^{q+} projectiles, where the first L-shell hole appears (~400 eV), is not at all mirrored in a jump in the proton yield. This finding strongly argues for a mechanism that involves directly the effect of the strong Coulomb field of the projectile far away from equilibration as opposed to an energy dissipation and equilibration process.

The currently accepted scenario for neutralization of highly charged ions involves two stages [16,18]. The first stage consists of the formation of hollow atoms above the surface by multiple resonant over-barrier charge transfer. The buildup of this diffuse charge cloud provides transient screening and determines the energy gain due to image acceleration. The above surface interaction time is insufficient for a significant relaxation of potential energy. As the projectile reaches the surface this diffuse charge cloud is replaced (''peeled off'') by a more compact screening cloud having the size of the bulk screening length [19] (second-generation hollow atom), which is formed by quasiresonant and Auger capture into the L and/or M shell [20,21]. Only in this second stage does energy dissipation mostly by Auger decay lead to the relaxation to the ground state. The validity of this scenario for insulators is currently under intense investigation [22–24]. Initial theoretical simulations for multiply charged ions interaction with a LiF [25] surface indicate that the hollow-atom formation above the surface takes place, however, with a significantly reduced charge transfer rate and a corresponding slower buildup of the diffuse charge cloud.

It is intuitively clear that a strong Q dependence of the proton sputtering yield originates most likely from the Coulomb interaction between hydrocarbons and the highly charged ions in the above-surface phase prior to the formation of the compact screening cloud. Moreover, because of the light mass of the proton, the dissociation energy [Eq. (1)] is sufficient for the proton to escape from the surface, while the projectile is still above or near the surface. It is understood that additional sputtering events may result from the second stage of the neutralization and relaxation process. The latter is expected, however, to result in a weaker Q dependence and smaller absolute yields and will not be considered in the following.

Within this framework, proton sputtering requires the breaking of the covalent bond between two neighboring C and H atoms due to resonant charge transfer of two electrons to the projectile leaving behind a C^+ and a H^+ ion. In addition, the ionic charge state of the sputter fragments must be maintained for a characteristic correlation time τ such that dissociation occurs. The latter is determined by the requirement that the gain in kinetic energy is sufficient to overcome the surface binding energy. Furthermore, τ must be sufficiently long such that the proton escapes to distances from the surface where reneutralization becomes unlikely. Accordingly, we determine the sputtering rate per unit time as

$$\gamma_H = \frac{K_1}{\tau} N_H S_2(C^+, H^+, t, \tau), \tag{2}$$

where N_H is the number of H atoms in the surface area with radius ρ situated ''below'' the HCI from which resonant over-barrier charge transfer to the projectile is possible. K_1 is a geometric constant that determines the fraction of dissociating protons emitted into vacuum. Only if the direction of the C-H bond is such that the H^+ ion escapes towards the vacuum does proton sputtering take place. $S_2(C^+,H^+,t,\tau)$ denotes the two-particle autocorrelation function for the formation of ion pairs of C^+ and H^+. This correlation function describes the probability for finding at time t ion pairs of C^+ and H^+ belonging to the same bond and whose charge states remain frozen at time $t+\tau$ (i.e., correlated to their initial values at a time t) during the correlation time τ. The sputtering process is governed by two disparate time scales $\tau \ll t_I$, where t_I is the effective interaction time between the HCI and the surface layer

$$t_I = R_c / v_\perp \simeq \frac{2\sqrt{Q}G}{W v_\perp}, \tag{3}$$

with R_c the critical distance from the surface where over-barrier charge-transfer processes set in. A detailed determination of the correction factor G that accounts for the surface dielectric constants near insulator surfaces is given in Ref. [25]. For the following discussion it is sufficient to note that $G \simeq 1$, which we will use for surfaces with hydrocarbon deposits. In the potential-energy sputtering regime $v_\perp$ is typically of the order of $v_\perp \gtrsim 5 \times 10^{-3}$ delimited by both the initial impact velocity and the additional image acceleration for very slow, highly charged ions. Note that this acceleration can be partially compensated by the deceleration of the ion due to the repulsive interaction with the charged-up surface [25]. We therefore treat in the following $v_\perp$ to be a charge-independent constant. The interaction time is of the order of $t_i \simeq \sqrt{Q} \times 10^3$ for a typical ionization potential (work function) $W \simeq 0.4$ a.u., while τ can be estimated to be of the order $\lesssim 300$ a.u., as discussed below.

We approximate now the two-particle correlation function by the product of single-particle correlation functions

$$S_2(\mathrm{C}^+,\mathrm{H}^+,t,\tau)\simeq K_2 S(\mathrm{C}^+,t,\tau)S(\mathrm{H}^+,t,\tau), \tag{4}$$

where $S(x^+,t,\tau)$ ($x=C$ or H) denotes the single-particle autocorrelation function for the x ion describing the probability for remaining ionized during the time interval τ, thereby neglecting pair-correlation beyond the geometric correlation. K_2 gives the geometric probability that the C^+ and H^+ ions belong to the same pair and is therefore given by

$$K_2\simeq d_0^2/\rho^2. \tag{5}$$

In our numerical results, only the product of the constants $K=K_1K_2$ will enter. The autocorrelation function $S(x^+,t,\tau)$ can now be determined from the coupled rate equations for the time-dependent probabilities $p(x^+,t)$ for each constituent (x=C,H),

$$\begin{aligned}\frac{d}{dt}\,p(x^+,t)&=w_c(x,t)p(x,t)-r(x,t)p(x^+,t),\\ \frac{d}{dt}\,p(x,t)&=r(x,t)p(x^+,t)-w_c(x,t)p(x,t),\end{aligned} \tag{6}$$

with $0\leqslant t\leqslant t_I$ and initial conditions $p(x,0)=1$ and $p(x^+,0)=0$. In Eq. (6), $w_c(x,t)$ determines the rate for electron capture by the highly charged ion, which can be estimated from the classical over-barrier model, while r denotes the reneutralization rate due to charge transfer from neighboring target atoms. In Eq. (6) we have omitted for simplicity terms describing the net loss due to sputtering since in the present case the total sputtering probability per surface atom remains small.

Our estimate for w_c relies on the observation that for metals the staircase model of stepwise neutralization describes the charge state evolution reasonably well [26,27]. Its validity for insulators is still under investigation [22–25]. We use

$$w_c(t)\simeq c_I\left[2v_\perp\,\frac{Q}{R_c}\left(\frac{R(t)}{R_c}\right)\right], \tag{7}$$

where w_c is the total capture rate and

$$w_c(x,t)=f_x w_c(t), \tag{8}$$

where f_x is the stoichiometric fraction of the species x. We note that the differences in the ionization potentials can also be included in f_x since w_c is proportional to W^2. The prefactor c_I takes into account the fact that the experimental [20,22] and theoretical evidence from simulations [25] suggests that the capture rates from insulator surfaces are reduced compared to a metal. Numerical values will be discussed below. The estimate for the reneutralization rate in an insulator is more difficult. Clearly, r is expected to be considerably smaller than the characteristic screening rate in a metal (of the order of the plasmon frequency $\simeq$0.5). We will use the neutralization rate in the absence of the perturbation by the highly charged ion r_0 as a free parameter, however, with its order of magnitude delimited by the nearest-neighbor hopping matrix element (or, equivalently, the width of a narrow band) $r_0\lesssim 10^{-1}$.

The modification of the reneutralization rate due to the presence of a highly charged ion can be easily estimated from the over-barrier model. Because the ionized surface atom and the highly charged ion form a single, quasimolecular potential well, the electron transferred from nearby atoms is shared by the phase space of the surface atom and the projectile. Following similar considerations for ion-atom collisions [28], we use

$$r(x,t)=\frac{r_0(x)}{1+n_p^2(t)}, \tag{9}$$

where n_p denotes the principal action (quantum number) of the projectile level, in resonance with the target, which is given by $n_p(t)\simeq q(t)/\sqrt{2W}$ in terms of the time-dependent charge $q(t)$ with $q(t=0)=Q$. Since $n_p(t)$ is approximately proportional to Q for large Q, reneutralization of ionized surface atoms to be sputtered is strongly suppressed as the charge flow is diverted to the projectile.

The numerical solution of Eq. (6) gives the single-particle probability for ionization $p(x,t)$. Using the fact that $\tau\ll t_I$, the single-particle autocorrelation function for ionization can be approximated in terms of a Poisson process, i.e.,

$$S(x^+,t,\tau)=p(x^+,t)\exp[-\tau r(x,t)]. \tag{10}$$

In Eq. (10) the probability for finding species x ionized at time t is given by $p(x^+,t)$. The probability that it remains ionized during the interval $(t,t+\tau)$ is accounted for by the exponential factor $\sim\exp[-\tau r(x,t)]$, thereby assuming that the rates ω_c and r show little variation during the correlation time τ. Finally, the sputter yield induced by a highly charged ion above the surface follows from Eqs. (2), (3), and (7) as

$$\begin{aligned}Y(\mathrm{H}^+)&=\frac{N_\mathrm{H}K}{\tau}\int_0^{t_I}dt\;S(\mathrm{H}^+,t,\tau)S(\mathrm{C}^+,t,\tau)\\ &=\frac{N_\mathrm{H}K}{\tau}\int_0^{t_I}dt\;p(\mathrm{H}^+,t)p(\mathrm{C}^+,t)\\ &\quad\times\exp[-\tau(r(\mathrm{H},t)+r(\mathrm{C},t))].\end{aligned} \tag{11}$$

III. NUMERICAL RESULTS AND DISCUSSION

Obviously, the theoretical description presented in the preceding section depends on a number of poorly known parameters whose judicious choice is a prerequisite for the model to have predictive power, even on a semiquantitative level. The ‘‘hot’’ surface area from which charge electron capture to the projectile takes place is given within the over-barrier model by a disk with radius of $\rho\simeq 5$ a.u. It should be noted that within the classical over-barrier model this size is only weakly dependent on Q, i.e., the strong Q dependence is *not* due to an effective increase of the number of available hydrogen atoms. Assuming a deposition density of hydrocarbons of $\simeq$0.1 C–H pairs per unit area (in a.u.), $N_\mathrm{H}\sim 16$. Simulations for proton emission as a function of the launching angle θ relative to the surface normal, as determined by the initial C-H bond orientation, indicate that the protons can

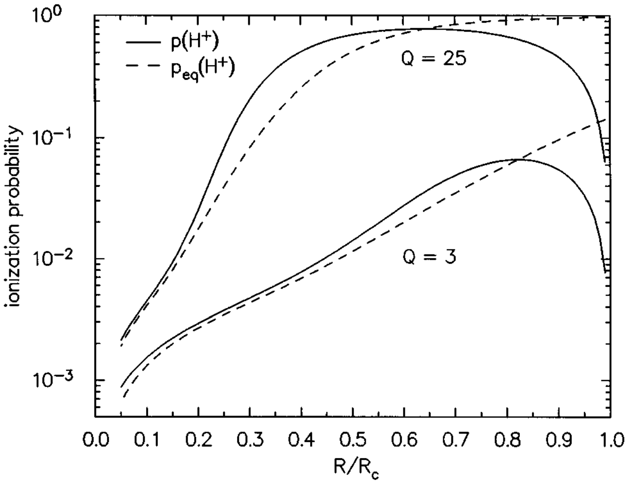

FIG. 1. Ionization probability of hydrogen $p(\mathrm{H}^+, R/R_c)$ as a function of the scaled distance R/R_c from the surface of the highly charged ion with the initial charge states $Q=25$ and 3 ($c_I=0.2$, $r_0=0.05$). Solid line, numerical solution of Eq. (6); dashed line, equilibrium solution of Eq. (6) assuming instantaneous equilibrium at a given distance R.

escape only for intermediate launching angles ($35° \lesssim \theta \lesssim 55°$). For smaller angles, scattering at the projectile reflects the proton back to the surface. At large angles, the combined fields of the projectile image and of the local charge-up of the surface lead to trajectories with long dwell times near the surface, which favors reneutralization. The constant K_1 can therefore be estimated as $K_1 \simeq 0.125$. Accordingly, $K=K_1K_2 \simeq 2\times10^{-2}$. Both K_1 and K_2 may contain weak dependences on Q, which we will neglect in the following. All multiplicative constants entering Eq. (11) do not carry any Q dependence.

Due to the exponential dependence of Eq. (11) on the correlation time τ, the absolute yield depends strongly on τ. We have determined τ by solving the equation of motion for the C^+-H^+ Coulomb explosion, requiring that the kinetic energy exceed typical molecular binding energies ($E_B \sim 0.1$ a.u.) and that the distance exceeds the critical distance for over-barrier recapture (often referred to as freezing distance) $\simeq 4$ a.u. and find $\tau \simeq 200$ a.u. We allow the ionization potentials for the first and second electron of the C-H bond to be different and choose $W_1=0.37$ a.u. and $W_2=0.55$ a.u. Because of the symmetry of the expression Eq. (11) in H^+ and C^+ we can associate W_1 with H^+ and W_2 with C^+ (or vice versa) without loss of generality.

The resulting Q dependence is primarily influenced by the choice of r_0 and c_I. Values of $c_I \ll 1$ have been found [25] in over-barrier simulations for insulators (LiF). We have varied r_0 between 0.008 ($\simeq 3.3\times10^{14}$ sec^{-1}) and 0.05 ($\simeq 2\times10^{15}$ sec^{-1}), which are comparable to rates used in the Coulomb explosion model [7] and are also of the order of typical Auger neutralization rates. We note that, according to Eq. (6), $p(x,t)$ displays, over a limited range, an approximate scaling in r_0/c_I so that the extrapolation of the results to other parameters is possible.

Figure 1 shows the approach of the ionization probability $p(\mathrm{H}^+)$ toward ionization equilibrium as a function of the scaled distance R/R_c from the solid for $Q=3$ and 25 and $r_0=0.05$, $c_I=0.2$. The equilibrium ionization probability $p_{\mathrm{eq}}=\omega_c/(\omega_c+r)$ is defined through $dp/dt=0$ in Eq. (6). As the projectile reaches the critical distance R_c for over-barrier charge transfer ($R/R_c=1$), $p(\mathrm{H}^+)$ rises from zero toward the equilibrium ionization. We observe characteristic changes as a function of Q. For small Q the equilibrium is rapidly reached as the ion approaches the surface ($R<R_c$), but the equilibrium lies at low levels of ionization, i.e., the overwhelming fraction of surface atoms in the hot surface are neutral. By contrast, for highly charged ions the ionization equilibrium is dramatically shifted toward ionization of surface atoms. However, this equilibrium is only slowly reached at smaller R. At small distances the charge state of the projectile is reduced by hollow-atom formation, thereby reducing the equilibrium ionization probability. The Q dependence of the ionization dynamics considerably influences the Q dependence of $Y(\mathrm{H}^+)$.

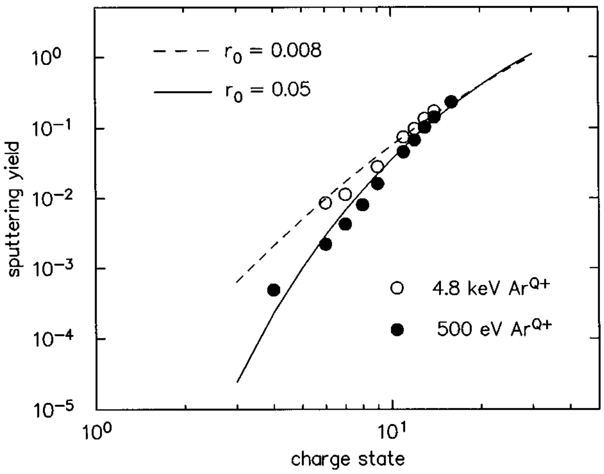

FIG. 2. Typical charge state Q dependence of the proton sputtering yield $Y(\mathrm{H}^+)$ from a hydrocarbon surface for different parameters ($c_I=0.2$, $r_0=0.05$ and $c_I=0.1$, $r_0=0.008$) (see the text). Experimental data: ●, Ar^{q+} at 500 eV; ○, Ar^{q+} 4.8 keV [17].

The sputtering yield Y as a function of the charge state Q of a structureless projectile for two different sets of parameters ($c_0=0.2$, $r_0=0.05$ and $c_I=0.1$, $r_0=0.008$) is displayed in Fig. 2. For a wide range of values of the parameters r_0 and c_I consistent with the limits discussed above, we find a remarkably steep increase with Q. For small $Q \lesssim 10$, the increase is consistent with a power law Q^β ($4 \lesssim \beta \lesssim 6$), in agreement with experimental data [13,14]. Moreover, the order of magnitude of the absolute sputtering yield is reasonably well reproduced, given the uncertainties in the parameter values. The present model not only reproduces the major trends in available data but also predicts weakening of the Q dependence of the sputtering yield with a further increase of Q. This onset of saturation is not only due to the exhaustion of the supply of hydrogen in the hot area. In fact, the number of sputtered protons remains relatively small compared to the number of available H atoms. The predicted weak Q dependence as $Q \rightarrow \infty$ is primarily due to the delayed approach of ionization equilibrium as a function of R. Equilibration becomes increasingly difficult to establish within the limited above-surface interaction time as the latter is intrinsically limited by the image acceleration of the projectile [26,29].

Our present results may also shed some light on related experiments of sputtering of ions from LiF surfaces in the potential-energy regime [9,10]. Yields of F^+ and Li^+ also display a strong Q dependence ($\sim Q^{\beta}$, $\beta \approx 3$), however, with absolute yields orders of magnitude below the ones for $Y(H^+)$ in the potential sputtering regime while exhibiting different impact velocity dependences in the kinetic regime. In view of Eq. (11), these findings can be at least qualitatively explained. The Coulomb repulsion resulting from the ionic cage of an alkali-halide crystal is weaker than the one resulting from a broken covalent bond because the larger atomic separation and because the forces exerted by the ionic cage are largely directed in the surface plane rather than along the surface normal. Since, furthermore, the mass of the sputtered particle is much larger, this translates into a considerably larger dissociation (or correlation) time τ resulting in an exponential suppression of the ionic sputtering yield. In addition, recent experimental evidence [24] suggests a strong influence of occupied electronic surface states on the neutralization of projectile ions in front of alkali-halide surfaces. The presence of weakly bound surface electrons would increase the reneutralization rates r_0 compared to the estimate for insulators and further reduce the sputter yield of ionized fragments.

IV. CONCLUSION

In conclusion, we have presented a simple theoretical framework for the description of proton sputtering induced by highly charged ions above the surface. We find a steep rise of the yield as a function of the charge state, in agreement with experiments, which is predicted to saturate for large Q. We stress that the total sputtering yield will also contain a contribution from at or below the surface. The latter is, however, expected to have a much weaker Q dependence. A comparison with experiments with better characterized surfaces for which a microscopic determination of relevant parameters is feasible would be very desirable.

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