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# Neutralization of noble-gas ions at very low energies

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Neutralization of noble-gas ions ($He^+$, $Ne^+$, $Ar^+$) on Pt(100) has been studied. It is common to the three kinds of projectiles that the ion yield shows a V-shaped incident-energy dependence at very low energies. The minimum position of the yield and the steepness of the V shape depend on the projectile ion species. The ion-yield change is governed by the change of the ion survival probability against Auger neutralization. Angular distributions show that the ion yield becomes higher in small-angle scattering with decreasing incident energy. A new empirical model is presented for the ion survival probability depending on penetration depth and residence time in the ion-surface interaction region.

Ion neutralization of a low-energy ion beam on metal surfaces has long been an important subject in surface science. Among several aspects of neutralization, the validity of Hagstrum's formula for the ion survival probability $P$,[1]

$$P = \exp\{-v_c[(1/v_{\text{in}\perp}) + (1/v_{\text{out}\perp})]\} , \quad (1)$$

is still in dispute, where $v_c$ is the characteristic velocity and $v_{\text{in}\perp}$ and $v_{\text{out}\perp}$ are the normal components of the velocity of the incoming and the outgoing ion, respectively. In most of the experimental and theoretical studies so far, it was shown that Eq. (1) holds approximately above the ordinary energy range of low-energy ion scattering (LEIS). Deviations from Eq. (1) were discussed and more sophisticated models have been presented.[2-8] Recently, the authors have reported the incident energy dependence of the scattered ion yield of $Ar^+$, $N^+$, and $N_2{}^+$ from a Pt(100) surface down to 10 eV and have shown that the ion survival probability does not fall to zero when energy goes to zero.[9] In these ion-surface interaction systems, the energy levels of the ground state of the projectiles are located at almost the same position just below the bottom of the Pt valence band, and the effect of resonance neutralization and collisional neutralization were discussed.

In this Communication, we report subsequent experimental results for neutralization of $He^+$ and $Ne^+$ on a Pt(100) surface. Since the energy levels of the lowest excited state of these species are located above the Fermi level of a Pt(100) surface, only Auger neutralization is allowed for transfer into neutral ground states. Therefore, if we measure the scattered ion yield down to an energy range as low as 10 eV, we can check whether the ion survival probability shows a V shape similar to the case of $Ar^+$, $N^+$, and $N_2{}^+$ or not. We can reach a better understanding of the neutralization of noble-gas ions at very low energies ($E_i \leq 100$ eV) through the experimental results.

The experimental setup is the same as the previous one.[9,10] Briefly, a Menzinger-type ion source (Colutron source) with a mass-selecting magnet installed in a UHV chamber and an efficient deceleration-lens system enabled us to obtain a (10–60)-nA ion current on a Pt(100) target surface. The scattered ions were detected with a rotatable quadrupole mass filter without energy analysis. The trajectories of transmitted ions were bent by 90° using a deflector before detection with an electron multiplier, in order to remove neutral species and low-energy secondary ions. However, after proper cleaning procedures, very little secondary-ion emission except for the $H^+$ ion was observed. The scattered ion yield normalized with the incident ion current was obtained by integrating the mass peak and the correction for the mass peak width $\Delta M$ was made.

The scattered $He^+$ and $Ne^+$ ion yields in the specular direction along the [001] azimuth as a function of the incident energy are shown in Figs. 1 and 2. The $Ar^+$ result has already been reported in Ref. 9. In contrast with reactive ion species such as $N^+$ and $N_2{}^+$, the ion yield of noble-gas ions is high in the whole energy range (10–400 eV). The scattered ion yield is proportional to the product of the scattering cross section and the ion survival probability $P$. The cross section is generally a smoothly de-

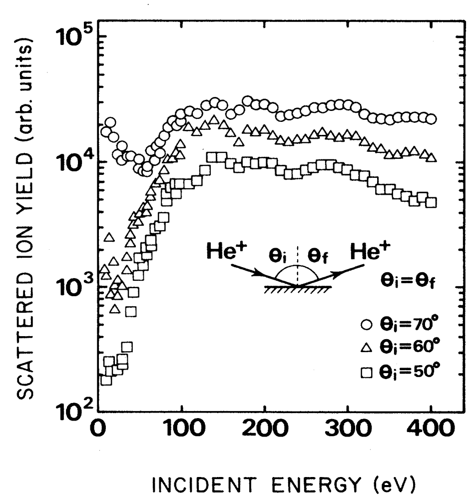


FIG. 1. Incident-energy dependence of the scattered $He^+$ ion yield for three specular scattering geometries along the [001] azimuth on Pt(100). The arbitrary units used are common to Fig. 1, Fig. 2, and Ref. 9 ($Ar^+$ incidence).

 

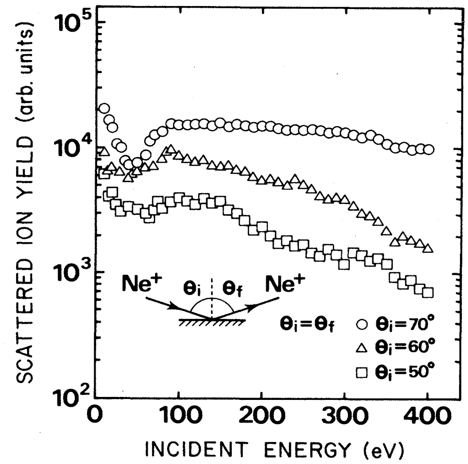


FIG. 2. Incident energy dependence of the scattered $Ne^+$ ion yield for three specular scattering geometries.

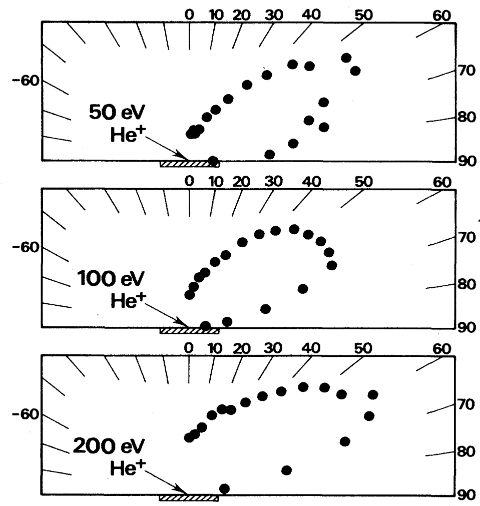


FIG. 3. Angular distributions of scattered $He^+$ ions at $\theta_i = 60°$ with $E_i = 50$, 100, and 200 eV. An arbitrary scale is chosen.

creasing function of energy and $P$ is an increasing function in the ordinary energy range of LEIS. Then the ion yield has a maximum at some incident energy.[11] In the present system, the maximum point is seen at about 100–150 eV with $He^+$ and $Ne^+$ and above 400 eV with $Ar^+$. Besides the usual energy dependence, the ion yield shows commonly a V shape in the very low energy regime for the three kinds of projectile ions. The minimum turning point is 20 eV with $He^+$, 50–60 eV with $Ne^+$, and 100 eV with $Ar^+$ for the incidence angles $\theta_i = 60°$ and 50°. At $\theta_i = 70°$, the turning point is shifted slightly to higher energies. The energy dependence of the ion yield is strong for $He^+$ and $Ar^+$, and weak for $Ne^+$.

The projectile dependence of the turning-point energy shows that the V shape of the ion yield in the energy dependence reflects predominantly the change of the ion survival probability rather than the change of the scattering cross section. The differential scattering cross section calculated with a binary single-collision approximation shows a much more moderate incident energy dependence as compared with the steep V shape. In the same way, it is easy to see that the yield increase above the turning point is not due to enhanced reionization.[12–14] The reionization process becomes more significant as the ground-state energy level of the projectiles is located closer to the valence-band bottom of the target. Then, if reionization comes into play, the reionization probability will be larger with $Ar^+$ than with $He^+$ and the threshold energy of reionization will be lower with $Ar^+$ than with $He^+$. However, the experimental result shows the opposite: the turning point energy of $He^+$ is lower than that of $Ar^+$. Moreover, it is unlikely that the reionization takes place at such a very low energy on a Pt surface with an almost filled valence band. Therefore, we can conclude that the V-shaped energy dependence of the ion yield is dominated by the change of the ion survival probability against Auger neutralization.

The distinctive energy dependence of the ion survival probability is associated with the angular distribution of scattered ions shown in Figs. 3, 4, and 5. With $He^+$ and $Ne^+$, the lobe position is nearly the specular direction above 100 eV. Below 50 eV, the lobe of $Ne^+$ shifts 10° from the specular direction towards the surface and the contribution of small-angle scattering to the ion yields increases. These features are most clearly seen in the $Ar^+$ case. At 50 eV, the lobe position of $Ar^+$ scattering is 20° towards the surface from the specular direction. Even at 100 eV, the ions are ejected in the parallel direction to the surface.

The experimental result that the angular distributions shift towards the surface as the incident energy decreases represents two important features which characterize the ion scattering on the surface at large incidence angle with

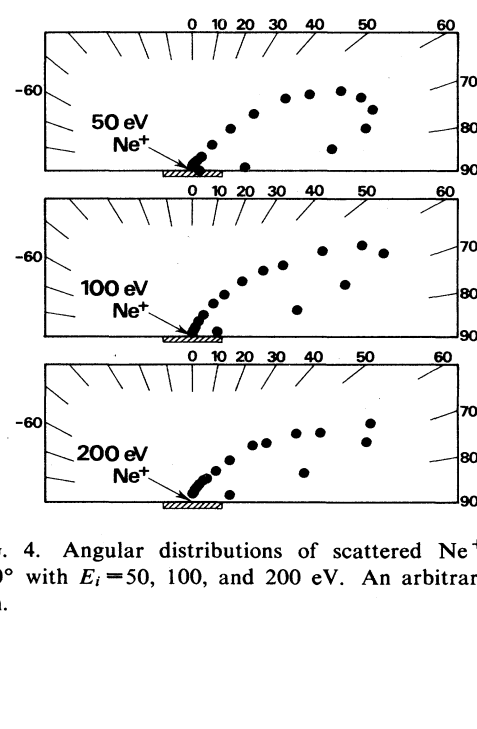


FIG. 4. Angular distributions of scattered $Ne^+$ ions at $\theta_i = 60°$ with $E_i = 50$, 100, and 200 eV. An arbitrary scale is chosen.

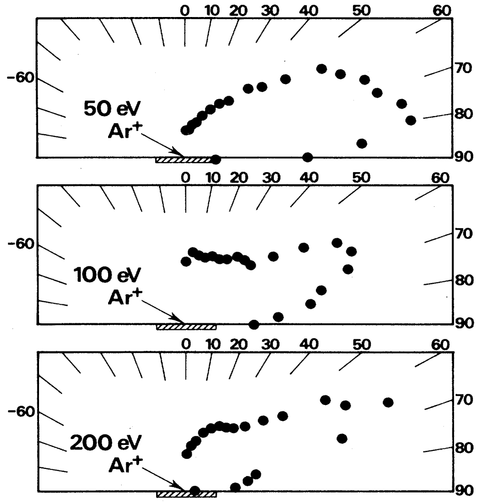


FIG. 5. Angular distributions of scattered $Ar^+$ ions at $\theta_i=60°$ with $E_i=50$, 100, and 200 eV. An arbitrary scale is chosen.

very low energies. First, the blocking effect is (or appears to become) unimportant. With decreasing energy, the ions are scattered from shallower regions of the surface potential. An ion sees a small-corrugation surface potential generated by the overlapping of surface atomic wave functions. Then, an ion reflected at a small angle travels along the nearly flat ion-surface interaction potential and is scattered softly. Second, the ion survival probability is large for the ions that collide with a large impact parameter and do not penetrate deep regions in the surface. Such ions are scattered at small scattering angles. These overviews lead to the following conclusion. At very low energies the ion survival probability is determined by the penetration depth of ions into the ion-surface interaction region rather than the residence time in the vicinity of the surface.

We can construct a simple analytical model to reproduce the V-shape ion yield by applying a Lee and George model[8] for derivation of the energy dependence of $v_c$ at very low energies and combining with the usual treatment at high energies as shown schematically in Fig. 6. We assume the ion-surface interaction potential is expressed by the repulsive potential of an exponential form, $V(s)=B\exp(-bs)$, which is a good approximation at very low energies, and the exponential transition rate of Auger neutralization, $R_t(s)-A\exp(-as)$, where $s$ is the distance from the surface. We further assume that the parallel component of velocity is nearly conserved in specular scattering. The closest-approach distance $s_0$ is given by

$$s_0=(-1/b)\ln(E_\perp/B)\,, \tag{2}$$

where $E_\perp$ is a perpendicular energy. Insertion of this relation into the characteric velocity defined as $v_c=(A/a)\exp(-as_0)$ yields

$$v_c=(A/a)(E_\perp/B)^{a/b}\,. \tag{3}$$

Since the repulsive interaction and Auger transition are

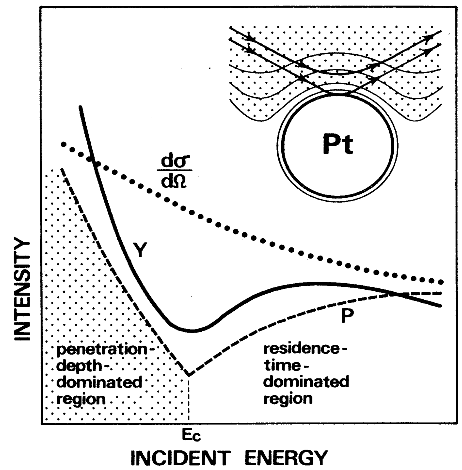


FIG. 6. Explanation of the empirical model for the incident energy dependence of the differential scattering cross section $d\sigma/d\Omega$ (dotted line), ion survival probability $P$ (dashed line), and ion yield $Y$ (solid line). Below the critical energy $E_c$, $P$ is dominated by the penetration depth into the ion-surface interaction potential, and above $E_c$, $P$ is dominated by the residence time in the vicinity of the surface. Inset: In the screened area Auger neutralization takes place.

based on the overlapping of the same wave functions,[15] it is reasonable to take $a=b$ and Eq. (3) reduces to

$$v_c=AE_\perp/aB\,, \tag{4}$$

which is proportional to $v_i^2$. This relation has been reported in an $Al^+$ sputtering experiment from an Al surface,[16] although an explanation in line with the Lee and George model was criticized.[17] There exists a critical closest-approach distance $s_c$, where the gradient of the ion survival probability changes from negative to positive. The distance $s_c$ corresponds to a critical incident energy $E_c$ (normal component $E_{c\perp}$) through Eq. (2). The V shape of the ion survival probability $P$ is reproduced, if we take

$$v_c=AE_\perp/aB, \quad \text{for } s_0 \geqq s_c\,, \tag{5}$$

$$v_c=AE_{c\perp}/aB, \quad \text{for } s_0 < s_c\,. \tag{6}$$

The physical image of this simplified model is that Auger neutralization takes place outside the sphere designated by $s_c$. From Eqs. (1) and (5) it is seen that at $E \leqq E_c$ the linear relation in $v_c$ to $E_\perp$ is superior to the inverse square root relation of the residence time. That is to say, $P$ is determined by the penetration depth of ions into the surface interaction region. At $E>E_c$, $P$ is given by Hagstrum's formula with a constant $v_c$. $P$ is controlled by the residence time in the vicinity of the surface. The parameter $B$ becomes larger with increasing atomic number. The value of $E_c$ appears to be correlated with the atomic number by Eq. (2) using a similar value of $s_c$ for the three kinds of noble-gas ions. However, since $v_c$ is expected to reflect a complex shape of the interaction potential depending on the incidence angle and projectile species, further quantiative discussion seems to be not fruitful at this stage.

In summary, we have measured the energy dependence of the scattered ion yield for three noble-gas ion projectiles ($He^+$, $Ne^+$, and $Ar^+$) from a Pt(100) surface in the 10–400 eV energy range. Observed V-shape energy dependence of the ion survival probability is due to the change of the Auger neutralization transition rate as a function of the penetration depth and the residence time of the projectile ion. This change is associated with the change of the angular distributions. The present experimental result indicates a possibility to apply the noble-gas ion beam to the rainbow scattering experiment in the very-low-energy regime. If the angular distribution of scattered ions is measured two dimensionally at normal incidence, a map of the electron density profile as viewed from the ion neutralization will be obtained, which is complementary with the experiment using alkali metal ion (neutral) projectiles.[18]

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