

Plasmon excitation in the interaction of protons and electrons with clean and potassium-covered Al surfaces

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We present a study of the low-energy electron emission resulting from the interaction of 5–100 keV protons colliding with clean and K-covered Al(111) surfaces at grazing incidence. In the whole energy range the spectra show structures that are associated with decay of both bulk and surface plasmons. We discuss the dependence of these structures with K coverage and compare them with those seen in the loss spectrum induced by 500 eV electron bombardment. We observe that the surface mode presents a stronger dependence with K coverage. Finally, we discuss the role of the ion velocity on the thresholds for surface and bulk plasmon production by direct excitation, by secondary electrons, and by electron capture processes.

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I. INTRODUCTION

The study of the collective excitation of electrons at a metal surface (surface and volume plasmons) is of fundamental interest for the understanding of surface properties and has been the subject of many experimental and theoretical works.^{1–23} In particular, for the case of an interaction of ions with surfaces,^{6–20} the volume plasmon excitation has been extensively discussed, while the surface plasmon excitation has been less frequently studied. The excitation processes involved in ion-surface collisions are either kinetic or potential processes depending on the potential energy of the incident ion and projectile energy.^{16,17} In the case of plasmons excited by highly charged ions or low-energy ions, potential effects such as plasmon-assisted electron capture and loss must be invoked.^{8,10,13,20} For low-energy protons Ritzau *et al.*¹⁰ and later Sánchez *et al.*¹² proposed a kinetic process where the excited secondary electrons of the metal are responsible for the excitation of the volume plasmon. Niehaus *et al.*¹⁸ and Winter *et al.*¹⁷ observed more prominent structures in a direction close to the sample normal which were ascribed to interference effects of Bloch waves. The different processes have been studied theoretically by several authors.^{6,7,9,10,14,17,18,20}

In the present paper we discuss the effect of an electropositive adsorbate on the excitation of the surface and bulk plasmons by ion and electron bombardment. Baragiola *et al.*¹¹ studied the dependence of the low-energy emission resulting from collisions of 100 eV He⁺ ions with Al surfaces covered with Cs. In that work the intensity of the structure observed at 11 eV decreased rapidly with Cs coverage. Because of this effect, the structure was attributed to a surface plasmon of short wavelength. More recently, Stolterfoht *et al.*¹⁹ have reported intense spectral structures near 6.5 and 11 eV produced by bombardment of Al with Ne⁴⁺, which were ascribed to surface and bulk plasmon decay. For the case studied here, 5–100 keV H⁺ hitting K-covered Al(111) surfaces, two similar structures are observed and followed as a function of the coverage. This method provides a clear identification of both surface and volume plasmons. Indeed,

as we could expect, the propagation of the electron oscillation parallel to the surface is more affected than the oscillation in the volume by the presence of potassium on the substrate. This is observed for incident ions as well as for incident electrons. An interesting feature is that *both* surface and bulk plasmon structures are present in the whole energy range, i.e., starting at 5 keV. In relation with this point, we discuss the effect of the velocity of the low-energy protons on the onset for plasmon excitation by direct processes, by secondary electrons, and by electron capture processes. Different approaches to these aspects have been discussed previously (see, for example, Refs. 9, 10, 14, 16, and 20). Finally, we show the existence of another electron structure at an energy that is in between of the ones for surface and bulk plasmon decay, and that might be associated with the excitation of a multipole plasmon of Al(111). The existence of such excitation and decay has been reported previously for incident photons^{21,22} and for incident electrons.^{16,23}

II. EXPERIMENTS

The experiment was performed in the Bariloche setup;²⁴ it consists of energy distribution measurements of electrons emitted during electron and grazing ion bombardment. The H⁺ ions were directed onto the Al surface with an incident energy ranging from 5 to 100 keV, at incident angles α in the range of 1°–10° with respect to the surface plane [inset of Fig. 1(b)] and at random azimuthal orientation. The electron spectra were measured with a custom made cylindrical mirror analyzer with 1% energy resolution, and the energy scale calibrated with the Al L_{VV} Auger peak (68 eV) induced by electron bombardment. The direction of observation was set at $\theta = 35^\circ$ (from the surface plane) and $\phi = 41^\circ$ [with respect to the incident beam direction, inset of Fig. 1(b)]. The angular acceptance was $\pm 2^\circ$. Observation at other angles (far from the sample normal) produced changes in the low-energy secondary maximum (in both position and intensity), but the structures assigned to plasmon decay remained at the same energy position. In order to measure the low-energy electrons the sample was biased to -5 V. The electron spectra are energy shifted by 5 eV and are not corrected for the

transmission function of the analyzer.

The sample was cleaned by cycles of 20 keV Ar^+ bombardment at 1° – 3° incident angle under continuous rotation of the azimuthal angle and sample annealing at 450°C . The cleanliness was verified by Auger electron spectroscopy (AES) before and after performing the measurements. The K deposition was obtained by evaporation from a heated alkali dispenser source (SAES getter, Italy). Auger measurements were performed to follow the increase of the K coverages step by step until saturation was reached. At room temperature the maximum coverage corresponds to one monolayer. After exposure the pressure in the chamber remained in the range of 10^{-10} T.

III. RESULTS AND DISCUSSION

A. Dependence with K coverage

Figure 1 shows electron energy spectra obtained (a) in the low-energy region (<25 eV, derivative mode) and (b) around the elastic peak (in the normal mode) for 508 eV electrons incident on an Al(111) surface covered with increasing amounts of K. The azimuthal direction of the incident electrons was set random; a typical spectrum in the low-energy region is shown in the inset of Fig. 1(a). On the high-energy edge of the broad structure seen in the inset we observe two shoulders at ~ 6.5 and ~ 11 eV which are attributed to the decay of surface and bulk plasmons, respectively. The two collective modes are better discernable in the first derivative dN/dE of the electron spectra.^{11–13,16} As it could be expected,²⁵ the surface plasmon contribution presents a stronger attenuation with K coverage, i.e., at 25% monolayer (ML) it becomes barely observable while the bulk plasmon intensity has decreased slightly. Measurements carried on at different incident directions presented a similar behavior. Figure 1(b) shows the electron energy loss spectrum (EELS) in the region corresponding to the surface and bulk plasmon losses. In correspondence with the above discussion for the low-energy part of the electron spectra, a preferential attenuation of the surface plasmon can be observed, although in this case the attenuation is not complete even near full coverage. This last phenomenon cannot be related to the inelastic mean free path λ , since λ for 500 eV electrons and for low-energy electrons (5–10 eV) are similar. The fact that a collective mode excited by 500 eV electrons [Fig. 1(b)] at the interface can be sustained even at coverages near one monolayer has been observed before by photon excitation.²⁵ In that work, performed at low temperatures, several alkali layers were needed to quench completely the substrate plasmon structure.

Figures 2(a)–2(d) show the electron energy spectra obtained for 5 and 40 keV H^+ ions incident on the Al(111) surface along a random azimuthal direction. The spectra were measured successively with increasing K deposition (from top to bottom). They are vertically shifted for comparison. The spectra shown in the right panels [(b) and (d)] are the derivative of those presented in the left ones [(a) and (c), respectively]. The general shape of these spectra corresponds to the typical energy electron distribution found in the literature.^{16,26} The dissymmetric broad structure is attributed

(a) 500 eV electrons on K/Al(111)

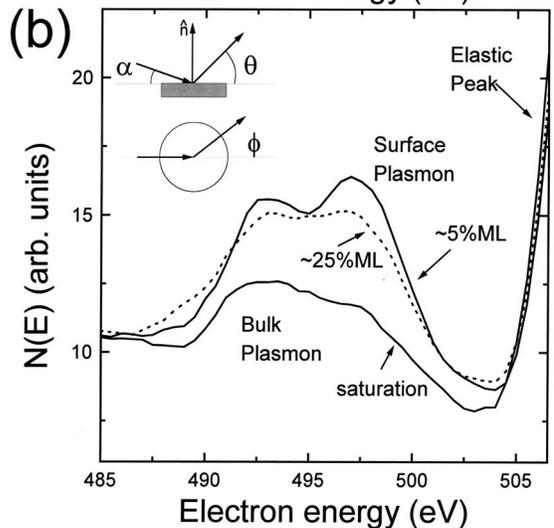
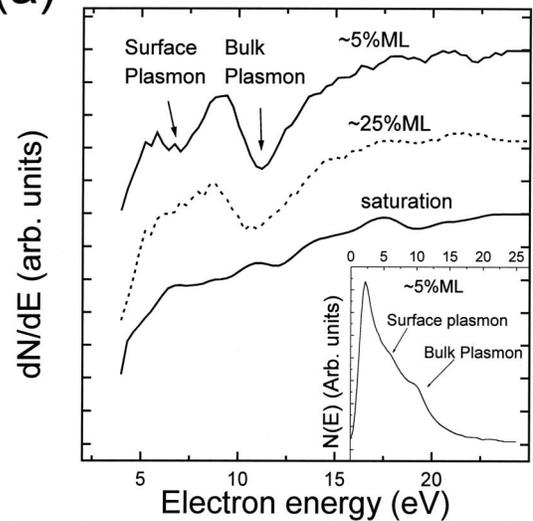


FIG. 1. (a) Derivative of the electron spectra measured during the scattering of 508 eV electrons at clean and K-covered Al surface. The inset shows the normal spectrum for 5% ML coverage. (b) Energy loss spectra for 508 eV electrons in the region of the first plasmon losses. The incident angle α is 35° and the observation angles are $\theta=35^\circ$ and $\phi=41^\circ$. The inset shows the angle definition.

to kinetic and potential electron emission from the surface. The low-energy edge of the spectra corresponding to near-zero-energy electrons shifts with increasing K coverage, reaching a value of about 2 eV for high coverage, which corresponds to the work function decrease of the sample. A somewhat surprising feature is that after the first adsorption (low coverage) the intensity of the low-energy secondary electron maximum decreases instead of increasing, as could be expected for a decrease in the work function. In order to check if this effect came from the sample polarization, we changed the sample bias (from 3 to 7 eV) and observed the same dependence. This effect could be due to the randomly adsorbed K atoms that can shadow the Al substrate and can produce scattering of the grazing ions at large angles, reduc-

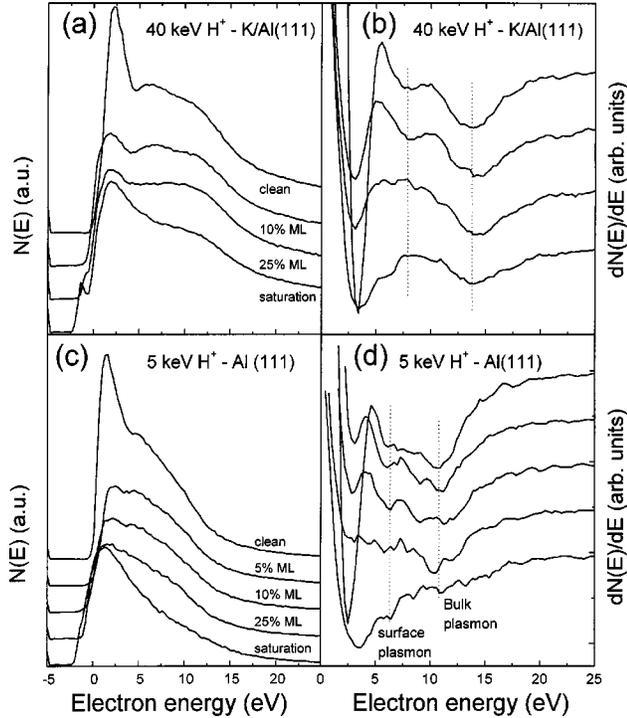


FIG. 2. Electron energy spectra $[N(E)]$ and its derivative $[dN(E)/dE]$ for H^+ at 40 keV [(a), (b)] and 5 keV [(c), (d)] incident on Al(111) for different K exposures. The vertical scale for each spectrum has been shifted for clarity. The incident angle α is 5° and the observation angles are $\theta=38^\circ$ and $\phi=44^\circ$.

ing the length of the ion trajectory at the surface and consequently reducing the number of ejected electrons. Note that a strong decrease in the specularly scattered projectiles is usually observed during the first stages of adsorption on flat surfaces.²⁷ At higher coverages the intensity of the secondary maximum increases again and becomes higher than for the clean surface.

On the high-energy edge of the secondary electron distribution we observe two structures which have been ascribed to plasmon decay.^{10,12,13,16} The two structures are better discernable in the derivative spectra [Figs. 2(b) and 2(d)]. At 5 keV incidence the two dips are around 6.3 and 11 eV. These correspond to the structures observed with 508 eV incident electrons (Fig. 1). These two structures correspond in energy to the decay of the zero-momentum surface and volume plasmon of Al(111), respectively. At 40 keV incidence the same two structures are better observed, but shifted to higher energies (8 and 13.5 eV). These can be attributed to the decay of non-zero-momentum surface and volume plasmons of Al. The energy of the plasmon depends on the excitation process. At 40 keV incident energy the excitation of both plasmons may also take place by a direct kinetic effect,¹⁶ which is not allowed at 5 keV (this will be discussed at the end).

Similarly to the behavior observed in Fig. 1, a decrease in the intensity is observed for both plasmons with increasing exposure. Here again the surface and volume plasmon structures vary differently. At low exposure the 6.3–8 eV structure decreases first. With increasing exposures this one disappears when the 11–13.5 eV structure starts to decrease.

The trends are similar for 5 and 40 keV H^+ incident energies (Fig. 2) and for 508 eV electrons (Fig. 1). Note that the observation angles used for incident ions and electrons are different, which suggests that the effects described above are not a specific feature of the direction of observation. Measurements carried on at other observation angles showed the same dependence with coverage for the plasmon structures. These measurements support the identification of the surface plasmon. Indeed, the presence of adatoms changes rapidly the properties of the topmost layer of Al and the self-sustaining oscillation of the electron gas at the surface is more sensitive than the collective oscillation of the electron gas propagating in the volume. Note that contrary to the behavior of the low-energy edge, the position of surface and volume plasmons does not shift with the changes of the sample work function due to K adsorption. This is because the plasmon energies are fixed with respect to the sample Fermi level, and in the experiments the Fermi levels of analyzer and sample are in contact (or biased by -5 V) and the electron energy is measured with respect to the vacuum level of the analyzer, which does not change with K adsorption.

At 5 keV H^+ a structure appears in between of the surface and bulk plasmon dip (at about 9 eV) that is better seen for low K coverage. Since its energy corresponds to $0.8\hbar\omega_p$, this could be ascribed to the decay of the surface multipole plasmon.^{3,16,21,23} The multipole plasmon of Al(111) has been seen by photon excitation, where it appears at the same energy,^{21,25} and by electron bombardment.^{16,23} The excitation of the multipole plasmon by photons and electrons *in the alkali film* has been studied by Barman *et al.*²⁵ and Tsuei *et al.*²⁸ for the case of several layers of alkali films grown on Al at low temperature. In our measurements we cannot observe the structure that comes from K since at room temperature the coverage is at most one monolayer and the excitation of the K plasmon is weak and appears at very low energies (1–2 eV).^{25,28,29}

B. Discussion of threshold values

The fact that surface and bulk plasmon structures are seen in the whole energy range, starting at 5 keV, cannot be explained by a direct kinetic mechanism, and as discussed previously,^{10,12,14,16,17} other processes must be invoked.

Kinetic processes. Using Lindhard's dielectric function formulation, the excitations produced in the inelastic scattering of a particle of charge Ze , mass m_0 , and velocity \mathbf{v} interacting with a free electron gas (FEG) may be described by the probability function^{30,31}

$$P_{\text{bulk}}(q, \omega) = \frac{(Ze)^2}{\pi^2 q^2} \text{Im} \left[\frac{-1}{\epsilon(q, \omega)} \right] \delta \left(\omega - \mathbf{q} \cdot \mathbf{v} + \frac{q^2}{2m_0} \right), \quad (1)$$

where \mathbf{q} and ω are the momentum and energy transfers to the FEG in units of \hbar . To describe plasmon excitations with this formalism we first look for the collective resonances in the dielectric function, given by $\epsilon(q, \omega) = 0$. This equation determines the plasmon dispersion curve $\omega_{\text{pi}}(q)$ and the range of undamped collective oscillations, $0 \leq q \leq q_c$ where q_c is the maximum momentum of the plasmon.

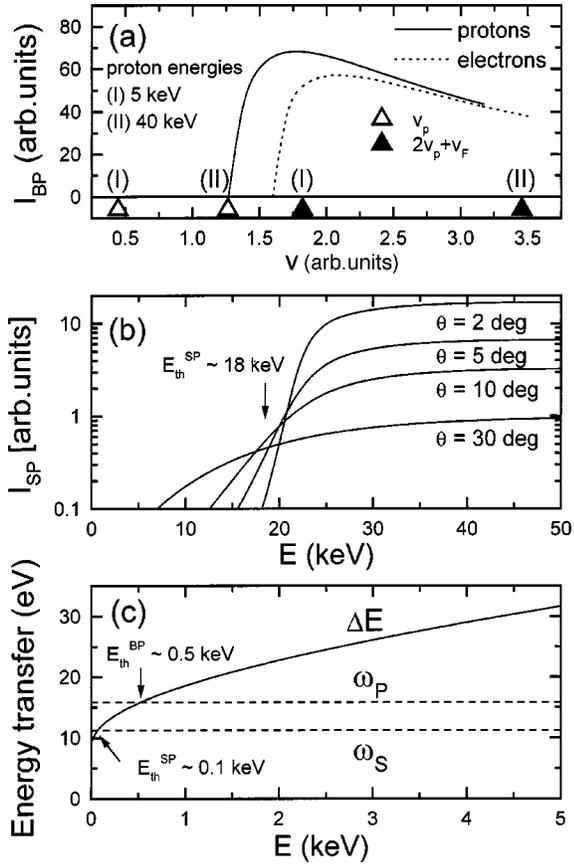


FIG. 3. (a) Probability for direct excitation of Al bulk plasmons by proton (solid line) and electron (dashed line) bombardment. The open triangles indicate the projectile velocity v_p for 5 and 40 keV incident protons. The solid triangles indicate the maximum velocity transferred to a secondary electron, $2v_p + v_F$. (b) Probability for direct excitation of Al surface plasmons by proton bombardment at different incidence angles. (c) Energy transfer to the free electron gas in Auger neutralization of protons vs the projectile energy.

Using Lindhard's dielectric function, with an r_s parameter appropriate for aluminum, $r_s = 2.07$ (corresponding to a plasma frequency $\omega_p = 0.582$), we have numerically determined the resonance curve, which may be approximated (within 0.5% precision) by the fitting function $\omega_{pl}(q) = \omega_p + 0.40q^2 + 0.405q^4$. The maximum wave vector q_c and the corresponding plasmon frequency $\omega_c \equiv \omega_{pl}(q_c)$ were numerically determined obtaining $q_c = 0.685$ and $\omega_c = 0.869$.

Using this formalism, we have integrated the probability of bulk plasmon excitation by either a proton or an electron (direct excitation process), obtaining the results shown in Fig. 3(a) where the solid line corresponds to protons and the dashed line to electrons. We obtained different *velocity thresholds* for each of these cases, at velocities $v_{th}^{(P)} = 1.27$ a.u. for incident protons and $v_{th}^{(e)} = 1.61$ a.u. for electrons; this corresponds to threshold energies $E_{th}^{(P)} = 40$ keV and $E_{th}^{(e)} = 35$ eV, respectively.

The displacement of the threshold velocity for electrons with respect to that for protons can be explained by the recoil term $q^2/2m_0$ in Eq. (1). In fact, for protons this term may be neglected and the threshold velocity is simply determined by

$v_{th}^{(P)} = \omega_c/q_c$, whereas for electrons ($m_0 = m_e = 1$ a.u.) one must keep this term and so the threshold is displaced to $v_{th}^{(e)} = \omega_c/q_c + q_c/2$ (in a.u.).

The relevant velocities for the cases of incident protons with energies of (I) $E_p = 5$ keV and (II) $E_p = 40$ keV are indicated in Fig. 3(a) by the triangles inserted on the x axis. The open triangles indicate the incident proton velocity for each case ($v_p = 0.447$ and 1.265 a.u.); clearly, these velocities are (i) well below and (ii) just at the proton threshold [cf. solid line in Fig. 3(a)], so that no plasmon excitation would be expected from the direct mechanism. However, this is valid for well defined plasmons of $q < q_c$. Calculations³² and electron energy loss measurements^{33,34} show that energy losses with intensities even larger than that for low- q plasmon take place at $q \geq q_c$. These direct excitations would also generate electrons at an energy near that corresponding to a $q = q_c$ plasmon, i.e., with a few eV above the $q = 0$ plasmon, as is observed in our experiments for 40 keV.

Similarly, we may also analyze the alternative process of plasmon excitation by "secondary" electrons, which may occur after these electrons are excited by a close interaction with the incident proton.^{10,14,20} The conditions of maximum energy transfer for the collision of protons with electrons yields a maximum possible electron velocity $v_{max} = 2v_p + v_F$ (where v_p is the proton velocity and v_F the electron Fermi velocity). The solid triangles in Fig. 3(a) give the values of $v_{max} = 2v_p + v_F$, which correspond to the indirect process mediated by secondary electrons. By comparing the v_{max} values with the dashed curve in Fig. 3(a)—which shows the probability of plasmon excitation by electrons—it can be concluded that the indirect (secondary electron) mechanism provides a possible explanation to the observed excitation of plasmons by low-energy incident protons: in particular, the threshold would be around $v_p = 1.61$ a.u., i.e., 2.9 keV. A similar value has been previously reported in Ref. 15.

We turn now to the process of surface plasmon excitation. We give here only a simplified consideration of the threshold behavior. The probability of surface plasmon excitation by a charged particle interacting with a metallic surface following a reflecting trajectory may be calculated using the expression³⁵

$$P_{\text{surf}}(\mathbf{Q}, \omega) = \frac{(Ze)^2}{\pi^2 Q} \left[\frac{2Qv_{\perp}}{Q^2v_{\perp}^2 + (\omega - \mathbf{Q} \cdot \mathbf{v}_{\parallel})^2} \right]^2 \times \text{Im} \left[\frac{-1}{1 + \epsilon_s(\mathbf{Q}, \omega)} \right], \quad (2)$$

where now \mathbf{Q} denotes the momentum transfer parallel to the surface, \mathbf{v}_{\perp} and \mathbf{v}_{\parallel} are the components of the particle velocity perpendicular and parallel to the surface, and $\epsilon_s(\mathbf{Q}, \omega)$ is the "surface dielectric constant." This expression has a resonance at $1 + \epsilon_s(\mathbf{Q}, \omega) = 0$ corresponding to the conditions for surface plasmon excitation. We approximate $\epsilon_s(\mathbf{Q}, \omega)$ around the resonance by the plasmon-pole approximation,³⁶ which yields

$$\text{Im} \left[\frac{-1}{1 + \epsilon_s} \right] \cong \left(\frac{\pi}{4} \right) \omega_s [\delta(\omega - \omega_s) - \delta(\omega + \omega_s)]. \quad (3)$$

The probability for the process is obtained by integrating $P_{\text{surf}}(Q, \omega)$ over all the possible values of momentum and energy transfers (Q and ω).³⁵ In Fig. 3(b) we show the result for the case of an incident proton on aluminum (with $\omega_s = 0.41$ a.u.) for various angles of incidence between 2° and 30° . The intensity I_{sp} shown in the figure is proportional to the average number of surface plasmons excited by a particle reflected at the surface. We observe a threshold around 18–20 keV for small angles of incidence and a lower threshold for larger angles. In any case, the probability of direct plasmon excitation at the experimental energy of 5 keV is negligibly small.

We have not performed additional calculations for electrons, but we expect a qualitative behavior similar to that in Fig. 3(a). Hence the excitation of surface plasmons at proton energies around 5 keV may be expected to occur not as a direct excitation process by the protons, but as a result of the excited “secondary” electrons.

Electron capture process. Finally, we have considered also the threshold conditions for the alternative process of plasmon excitation in the electron capture^{10,14,16,20} during the neutralization of H^+ into H^0 . The energy-momentum balance for this process may be expressed in terms of the energy transfer to the FEG, ΔE , as follows³¹ (in atomic units):

$$\begin{aligned} \Delta E(q) &= \frac{1}{2} (\mathbf{v}_e - \mathbf{v}_p)^2 + \mathbf{q} \cdot \mathbf{v}_p - \omega_0 \\ &= \frac{1}{2} (\mathbf{v}_e - \mathbf{v}_p)^2 + \mathbf{q} \cdot \mathbf{v}_p + I - \phi - E_F, \end{aligned} \quad (4)$$

where $\frac{1}{2}v_e^2$ (with $v_e \leq v_F$) is the kinetic energy of a FEG electron before being captured by the proton, \mathbf{q} is the momentum transfer from the proton to the FEG, either in the excitation of an Auger electron or in the excitation of a plasmon with $q \leq q_c$ (note that the total momentum transfer from the ion in this process is actually $\mathbf{q} + \mathbf{v}_e - \mathbf{v}_p$, which includes the momentum transfer to the captured electron, $\mathbf{v}_p - \mathbf{v}_e$), $I = -E_{\text{bound}}$ is the ionization energy of the atomic state ($I = 13.6$ eV for H^0 outside the solid or 11.6 eV near the surface), ϕ is the work function of the metal, E_F is the Fermi energy ($\phi = 4.3$ eV and $E_F = 11.7$ eV for Al), and $w_0 = E_{\text{bound}} - U_0 = -I + \phi + E_F$ is the energy of the atomic level with respect to the bottom of the FEG band U_0 (with $U_0 = -\phi - E_F = -16$ eV).

In the capture process mediated by plasmon excitation the energy ΔE is absorbed by the plasmon. The condition for this process is therefore $\Delta E(q) = \omega_{\text{pl}}(q)$. To find the threshold condition we consider the most favorable situation, which is obtained when the electron to be captured is initially with the maximum possible energy, $\frac{1}{2}v_e^2 = E_F$, and we also assume the maximum value of $\mathbf{q} \cdot \mathbf{v}_p|_{\text{max}} = q_c v_p$. In this case,

$$\Delta E(q)_{\text{max}} = \frac{1}{2} v_F^2 + v_F(v_F + q_c) + I - \phi. \quad (5)$$

The function $\Delta E(q)_{\text{max}}$, for $I = 13.6$ eV, is plotted in Fig. 3(c) as a function of the proton energy. As may be observed, the thresholds for bulk and surface plasmon excitation are located at quite low energies (~ 0.5 and ~ 0.1 keV, respectively) so that both processes are allowed at the energies of the current experiment.

IV. CONCLUSIONS

In conclusion, we discuss in this paper the energy distributions of the electrons attributed to plasmon decay induced by grazing collisions of 5–40 keV H^+ with an Al surface and the effect of adding a K overlayer on this surface. This method allows a clear identification of the surface and bulk plasmon decay structures. At low ion energies another structure at electron energies between the surface and bulk plasmon is seen for low K coverage. A discussion of the threshold energies corresponding to the different plasmon excitation mechanisms shows that the plasmon-assisted capture from Al valence band electrons to H^+ bound states should be allowed down to less than 1 keV projectile energy. We finally call the attention to the fact that part of the electron emission attributed to plasmon decay could result from excitation of damped plasmons^{33,34} with $q \geq q_c$ together with single-particle excitations in the same region,³² an effect that should be important near 40 keV projectile energies.

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