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# Subthreshold plasmon excitation in proton Al(111) collisions

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Electron energy distributions arising from 2–6-keV protons grazingly incident on an Al(111) surface are reported for two different azimuthal angles of incidence and for a range of observation angles. Although the protons have energies that are an order of magnitude below the generally accepted threshold for plasmon excitation, the spectra strongly suggest that excitation and subsequent decay of bulk plasmons takes place. To explain this remarkable observation we propose that a subthreshold proton may excite a bulk plasmon in a process in which not only energy and momentum are transferred to the plasmon but also, mainly momentum, to the target.

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## I. INTRODUCTION

Bombardment of a clean metal surface with ions in the keV energy range leads to the emission of electrons. The emission process is called potential electron emission (PEE) when the electrons are emitted at the expense of the potential energy of the projectile [1–4], and kinetic electron emission (KEE) when the electrons are emitted at the expense of the kinetic energy of the projectile [5–8]. One possible channel for both PEE and KEE is the excitation and decay of collective excitations in the electron gas of the metal. These excitations behave in a particlelike manner, and the associated particles are called ''plasmons.'' The particlelike behavior manifests itself in that a plasmon has both energy *and* momentum. The relation between the energy $\hbar\omega$ and momentum $\hbar k$ of the plasmon is described by the plasmon dispersion relation. In a nearly free electron metal like Al, the plasmon dispersion can be described as [9]

$$\omega(k)\approx\left(\omega_p^2+\frac{3}{5}v_F^2k^2\right)^{1/2}, \tag{1}$$

$$\omega_p=\left(\frac{ne^2}{\epsilon_0 m}\right)^{1/2}, \tag{2}$$

with $\omega(k)$ the frequency and $k$ the wave vector of the plasmon, $\omega_p$ the classical plasma frequency, $v_F$ the Fermi velocity, $n$ the electron number density, $\epsilon_0$ the dielectric constant of vacuum, and $e$ and $m$ the charge and mass of an electron. For Al, $\hbar\omega_p$ has a value of 15.7 eV. Equation (1) is a good approximation for the dispersion relation up to the critical wave vector $k_c$, i.e., the wave vector where the dispersion enters the single-particle continuum [9]. For Al $k_c$ has a value of approximately $0.634k_F$ (with $k_F$ the Fermi wave vector). From electron-impact experiments it is known that besides these (bulk) plasmons, collective excitations localized at the surface may also be excited. These excitations are called surface plasmons, and typically have an energy of $\hbar\omega_p/\sqrt{2}$ [10]. Excitations of bulk and surface plasmons by impact of electrons show up as characteristic energy losses in the energy distributions of backscattered electrons, and have been extensively studied in the past [5,9,10]. Theoretical descriptions are based on the screened Coulomb interaction between the incoming electron and the target electrons [5].

The contribution of the excitation and decay of plasmons to PEE has been investigated quite recently, both theoretically [11,12] and experimentally [13,14]. The results were obtained for incident noble-gas ions with their relatively large potential energy. In the experiments described in this paper, however, protons have been used for which the available potential energy is not sufficient to excite a plasmon. Kinetic excitation of plasmons by protons, on the other hand, is possible, has been studied for some time now, and has a firm theoretical basis [5,15–17]. Since the excitation process is also based on the screened Coulomb interaction between the incoming proton and the target electrons, the theoretical descriptions are similar as in case of incoming electrons [5]. When the kinetic excitation of a plasmon is considered as a process in which only the incoming ion and the plasmon are involved, simply applying energy and momentum conservation shows that this may only occur at proton energies higher than approximately 40 keV [5,18,19]. Subsequent decay of the excited plasmons predominantly takes place via the excitation of electron-hole pairs, whereas radiative decay only constitutes a minor channel [5]. In this paper we will present experimental spectra that strongly suggest that excitation and decay of plasmons in electron-hole pairs also occurs at proton energies far below the threshold of 40 keV. To solve the problem of the conservation of energy and momentum, we propose an explanation in which the excess momentum is transferred to the Al target.

## II. KINETIC THRESHOLD FOR PLASMON EXCITATION

We will briefly discuss the threshold behavior of the kinetic excitation of plasmons by protons. The dispersion relation according to Eq. (1) for a bulk plasmon in Al is shown in Fig. 1. The width of the curve, as indicated by the shaded area, corresponds to the finite lifetime of the plasmon, and is mainly caused by interband transitions [9]. The energy loss $\Delta E_{\mathrm{ion}}$ of an ion that induces a certain excitation can simply be written in terms of its change in absolute momentum $\Delta(\hbar k_{\mathrm{ion}})$

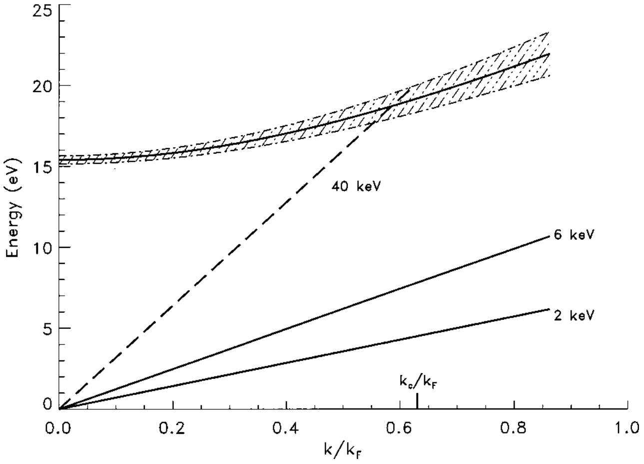

FIG. 1. Dispersion relation for a bulk plasmon in Al according to Eq. (1), with the width of the curve indicated by the shaded area. Also shown are energy-loss functions of protons that induce an excitation in the solid, for proton energies of 2, 6, and 40 keV. $k_c$ and $k_F$ denote critical and Fermi wave vectors for Al, respectively. Conservation of energy and momentum implies that only protons with energies higher than 40 keV may excite bulk plasmons.

$$\Delta E_{\mathrm{ion}}=\frac{\hbar k_{\mathrm{ion}}}{m_{\mathrm{ion}}}\Delta(\hbar k_{\mathrm{ion}})=v_{\mathrm{ion}}\Delta(\hbar k_{\mathrm{ion}}) \quad (\Delta k_{\mathrm{ion}}\ll k_{\mathrm{ion}}), \tag{3}$$

with $v_{\mathrm{ion}}$ the velocity of the incoming ion. The absolute momentum transfer is equal to the change in absolute momentum of the ion only if the ion is not deflected during the excitation process. Energy-loss functions according to Eq. (3) are shown in Fig. 1 for protons with an energy of 40 keV, and for protons with energies corresponding to the minimum and maximum energies used in our experiments. If in the excitation process only the proton and the plasmon are involved, conservation of energy and momentum implies that the energy-loss function of the ion has to intersect with the plasmon dispersion curve. For the proton energies used in our experiments this is clearly not the case; only for proton energies higher than approximately 40 keV does an intersection occur (see also Ref. [5]). Rösler and Brauer also calculated the energy distributions of electrons emitted due to the interaction of 60-keV (and higher energy) protons with an Al crystal and found rather smooth curves with high intensities at low electron energies and decreasing tails toward higher energies [5]. This electron emission is mainly caused by the excitation of single electrons via the interaction with screened protons. On top of these tails, at energies of approximately 11–12 eV, a small shoulder is visible, which is ascribed to the excitation and decay of bulk plasmons. The energy of the corresponding electrons is roughly given by $\hbar\omega_p-\phi$, i.e., $\hbar$ multiplied by the classical plasma frequency minus the work function of the crystal (for Al, $\phi\simeq 4-4.5$ eV depending on which surface has been used [20]). A study of the derivatives of the spectra also reveals small contributions from the excitation and decay of surface plasmons [5]. These theoretical findings have also been observed experimentally [18,19].

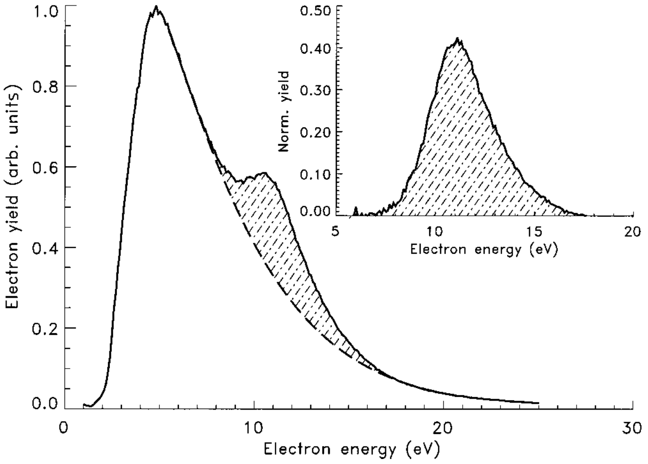

FIG. 2. Electron energy distribution obtained with 4-keV protons incident at 5° on Al(111) in the closest-packed direction, and measured perpendicular to the surface (solid line). The fit to the exponential-like tail, according to $\exp(E/A_1)E^{A_2}$, with $E$ the energy of the electrons and $A_1$ and $A_2$ fit parameters, is shown by the dashed curve. The shape of the peak is shown in the inset.

## III. EXPERIMENTAL APPROACH

The experiments presented here were performed with a 2–6-keV proton beam incident on a single crystalline Al(111) target. The ions were incident on the surface at a polar angle of incidence of 5° with respect to the surface plane and at an azimuthal angle of incidence parallel to the closest-packed direction (unless stated otherwise). The ion beam has an energy spread of approximately 10–20 eV, and an angular spread of approximately 0.5°. Since a grazing angle of incidence was used, the spot size on the crystal was large (about 13 mm in the direction of the beam), and therefore acceleration of the electrons toward the analyzer could not be applied. The target was mounted in an UHV chamber with a residual gas pressure of around $2\times10^{-10}$ torr, and was prepared by cycles of sputter cleaning and annealing. The electron energy spectra were taken by means of a hemispherical analyzer with an energy resolution of 3% and with a solid acceptance angle of $7.8\times10^{-5}$ sr. All spectra presented were corrected for the transmission function of the analyzer. Due to stray magnetic fields of about 25 mG, the spectra are only trustworthy at electron energies above around 5 eV. Except for the spectra shown in Fig. 5, all spectra were measured in a direction normal to the surface. As an extra validity check of the Al measurements, we have also performed experiments on a Cu(110) surface. The results of one of these experiments, performed at similar experimental conditions as the experiments with Al(111), will also be discussed. As an inset of Fig. 3, a schematic drawing of the experimental situation used in the Cu experiments is shown.

## IV. RESULTS

In Fig. 2 an electron energy distribution is shown that was obtained with 4-keV protons scattering off the Al(111) surface. The dominant part of the energy distribution consists of low-energy electrons with an exponential-like tail toward higher energies. In addition, a peaklike structure is observed at energies around 11 eV. Upon application of a small voltage to the crystal, the peak shifts accordingly, thereby showing that the peak is not caused by an experimental artifact of the analyzer and that it consists of electrons emitted from the crystal itself. The shape of the peak is shown as an inset of Fig. 2, and was obtained by subtracting a fit to the exponential-like tail from the experimental spectrum, and normalizing the obtained distribution to the electron intensity at the maximum of the peak. Most of the emitted electrons have energies well within the regime of the excitations of single electrons via the interaction with the screened protons, a regime that extends up to energies of approximately 20 eV [5]. However, these excitations are expected to lead to rather smooth electron energy distributions and not to a peaklike structure at 11 eV. The energy position of this peak coincides with the energy at which the plasmon shoulder is observed in experiments, and calculations performed with higher energy protons [5,18,19]. This exact agreement in energy and the absence of any other known mechanism that could possibly lead to a peaklike structure at the observed energy strongly suggests that the peak in our experiments is also caused by the excitation and decay of bulk plasmons. This is a rather unexpected result, since the presently used energy was approximately an order of magnitude below the calculated threshold of 40 keV. To our knowledge excitation of plasmons by protons with energies of only a few keV has only been invoked in one publication before [21], in order to explain a shoulder at the appropriate energy on top of an exponential-like background. This shoulder is much less pronounced than the peaklike structure we have observed in Fig. 2, a difference that is most probably caused by the much larger angle of incidence used in the experiments discussed in Ref. [21].

In order to further check the validity of our results, we have performed experiments with keV protons scattering off Cu(110). Although Cu is not a nearly free-electron metal like Al, it is known that collective excitations exist: Electron-energy-loss spectroscopy has shown that electrons transmitted through a thin film of Cu may lose an energy of approximately 20 eV [22]. In Fig. 3 an electron energy distribution is shown that was obtained with 4-keV protons scattering off the Cu(110) surface at the same experimental conditions as for Fig. 2. The experimental situation is shown as an inset of the figure. Again a high intensity is observed at low electron energies with an exponential-like tail toward higher energies. Also, a peaklike structure is observed which now seems to be centered at an energy of approximately 14–15 eV. This energy is consistent with the energy of the collective excitation at 20 eV minus the work function of the surface (($\phi$ $=4.48$ eV [20]). Again, the results strongly suggest that collective excitations, subsequently decaying in single-electron-hole pairs, are produced by keV protons scattering off a single-crystal surface.

The question now arises, what is the real kinetic energy threshold for plasmon excitation by protons? We have therefore performed a series of experiments on Al(111), in which the electron energy distributions were measured as a function of the proton energy. The results, i.e., the peaklike structures obtained by subtraction of the exponential-like tails as well as their integrated intensities, are shown in Fig. 4. The peak intensity clearly decreases with proton energy, leading to a threshold at an energy of approximately 2 keV. Also, the shape of the peaklike structure changes: With increasing proton energy the width becomes larger and the maximum shifts to higher energies. For 6-keV protons an additional structure at higher energies (ranging from approximately 15 to 20 eV) seems to appear. The general trends of these phenomena are realistic, and are not just caused by the subtraction procedure, since they are even visible in the raw spectra. They suggest that with increasing proton energy plasmons with higher momenta are excited. We presently do not understand the origin of the additional higher-energy structure for 6-keV protons and whether or not it can be understood in terms of

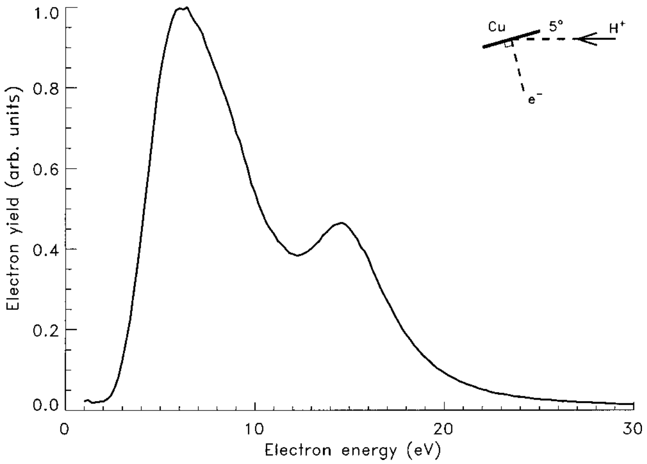

FIG. 3. Electron energy distribution obtained with 4-keV protons incident at 5° on Cu(110) in the closest-packed direction, and measured perpendicular to the surface. A schematic drawing of the experimental situation is shown as an inset of the figure.

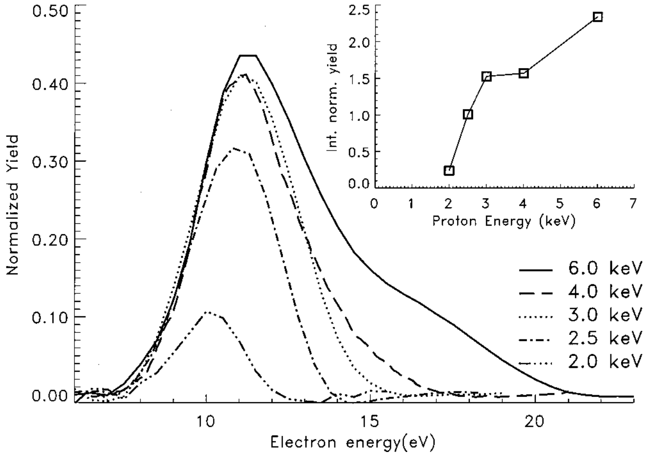

FIG. 4. Shapes of the peaklike structures for five different proton energies, obtained by subtraction of fitted exponential-like tails from experimental spectra (similar as discussed for Fig. 2). The experimental conditions were the same as the ones for Fig. 2. In the inset, the integrated intensity is shown as a function of the proton energy.

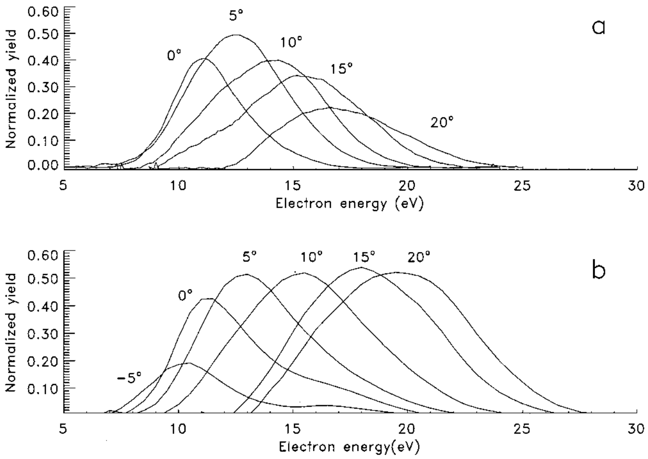

FIG. 5. Shapes of the peaklike structures for 4-keV (a) and 6-keV (b) protons incident at 5° in the closest-packed direction on Al(111) for different observation angles (given with respect to the surface normal, positive angles are in the direction of the proton beam).

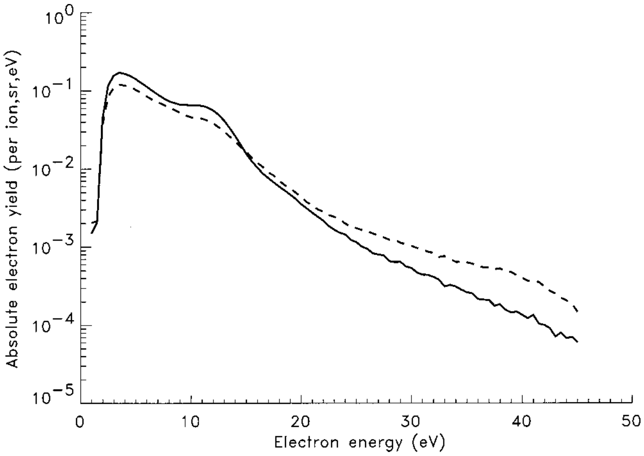

FIG. 6. Absolute electron energy distributions measured perpendicular to the surface obtained with 3-keV protons incident at 5° on Al(111) for two different azimuthal angles of incidence (solid curve: closest packed direction; dashed curve: ''random'' direction).

the excitation and decay of plasmons.

To further test the above ideas of excitation and decay of plasmons with finite momenta, we have performed two series of experiments in which the observation angle was varied: One for 4-keV protons and one for 6-keV protons. The results are shown in Figs. 5(a) and 5(b), respectively. The angles indicated in the figures denote angles with the surface normal (positive angles denote angles in the direction of the proton). With an increasing observation angle the peaklike structure shifts to higher energies and significantly broadens. These effects are more pronounced for the 6-keV protons leading for the two largest observation angles to energy distributions reaching up to at least 25 eV. This energy is in fact too high to be simply understood in terms of the excitation and subsequent decay of plasmons with momenta smaller than the cutoff momentum $k_c$ (compare to Fig. 1). This will be discussed in more detail in Sec. V.

For the Al spectra, one might be surprised that the spectra do not show any indication of the excitation and decay of surface plasmons. The energy position of the corresponding electrons would be at about 6 eV, which would be very close to the energy of the maximum intensity and to the range of energies that are affected by stray magnetic fields. Therefore, even in the derivative of the spectrum, excitation and decay of surface plasmons would not be discernable. Of course this does not imply that the excitation and decay of surface plasmons does not contribute to the emission of electrons with energies of around 6 eV.

## V. DISCUSSION

To explain this remarkable observation of plasmon excitation below the commonly accepted threshold, several mechanisms are possible. One possibility has been proposed in Ref. [21] as the most plausible one: plasmon excitation by electrons with energies higher than the plasmon energy that have been excited by the protons. The authors came to their conclusion by making a comparison between their experiments and calculations of the probability for plasmon excitation. We will return to this mechanism later. Another possibility for subthreshold plasmon excitation is that the surface acts as a ''third collision partner,'' and that additional momentum is transferred to the surface: Since the proton scatters off the Al atoms at the surface, it may be that during plasmon excitation excess energy and momentum are transferred via the recoiling of a single Al atom or via the excitation of one or more phonons. A process in which a single recoil atom is involved is, of course, equivalent to the excitation of a plasmon during a proton Al-atom collision. When the momenta of the proton and the Al atom are considered in the center-of-mass frame, one can easily show that on grounds of energy and momentum conservation excitation of a plasmon is allowed. Since the proton mass is small compared to the mass of an Al atom, this process mainly involves the transfer of momentum. When, instead of a single Al atom, one or more phonons are excited, a similar conclusion is reached: The energy of the phonon(s) is small compared to the energy of the plasmon, and the phonon(s), therefore, mainly carries (carry) away excess momentum. Since the number and the impact parameter distribution of the ion-atom collisions at the surface are expected to play an important role in case momentum transfer to the surface is involved in the excitation process, one would expect that the measured structure should depend on the azimuthal angle of incidence. Therefore, we have measured electron spectra from 3-keV protons incident at 5° on the Al surface for two different azimuthal angles of incidence, i.e., in the closest packed direction and in a direction 22° off the closest packed direction (we will call this the ''random'' direction). To make an absolute comparison possible, the spectra were normalized to the ion-beam flux as measured with a Faraday cup (see also Ref. [23]). The results are shown in Fig. 6. Obviously, in the random direction, the intensity of the low-energy electrons including the peaklike feature at an energy of around 11 eV is drastically smaller than the corresponding intensity measured in the closest-peaked direction, whereas

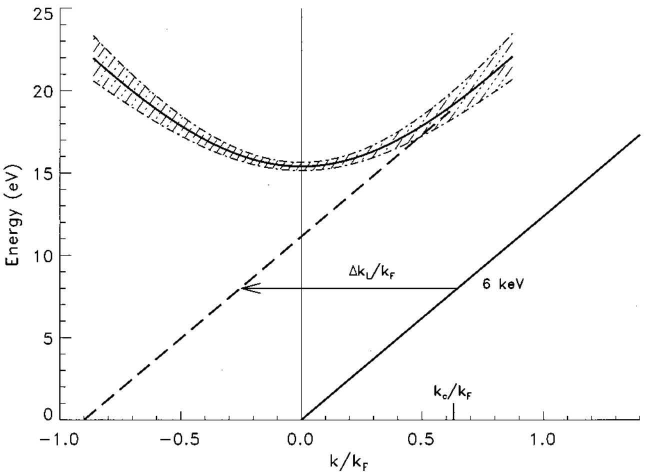

FIG. 7. Dispersion relation for a bulk plasmon in Al according to Eq. (1) with the width of the curve indicated by the shaded area. Also shown is the energy-loss function of a proton of 6 keV that induces an excitation without exchanging additional momentum with the target (solid curve). If the proton transfers additional momentum of $\Delta\hbar k_L$ to the target, the energy-loss function has to be shifted in the horizontal direction. Plasmon excitation is allowed only if the shifted curve (dashed curve) intersects with the plasmon dispersion relation. $k_c$ and $k_F$ denote critical and Fermi wave vectors for Al, respectively.

the intensity of the high-energy electrons is larger. These observations are consistent with the interpretation that plasmons are excited with the surface involved as a ‘‘third collision partner’’: It is known that when the ions are incident in the closest-packed direction they undergo many more ion-atom collisions than in the random direction (see, e.g., Ref. [23]), thereby being better able to transfer momentum to the surface. On the other hand, the observations are clearly not consistent with the interpretation that plasmons are excited by higher-energy electrons: The number of high-energy electrons is not at all proportional to the probability for plasmon excitation. Although the measurements shown in Fig. 6 are clearly in favor of the mechanism in which excess momentum is transferred to the surface, we can of course not exclude some contribution from high-energy electrons. It should be noted that in the spectra shown in Ref. [21], that were taken at much larger angles of incidence, the contribution of the plasmon feature to the electron emission is much smaller. It may very well be that the relative contributions of the different mechanisms strongly depend on the angle of incidence.

Not taking into account angular scattering of the proton, and assuming that both the plasmon and third collision partner have momenta parallel to the proton momentum, the condition for plasmon excitation to occur can simply be illustrated by shifting the energy-loss function of the ion. This is shown schematically in Fig. 7 for a 6-keV proton. The energy-loss function of the proton that induces an excitation without exchanging additional momentum with the lattice is shown by the solid line. If the proton transfers an additional momentum of $\Delta(\hbar k_L)$ to the surface, the energy-loss function of the ion has to be shifted in the horizontal direction. Plasmon excitation is allowed only if this shifted curve intersects with the plasmon dispersion curve, as shown in the figure. Since the slope of the energy-loss function of the ion is proportional to the ion velocity [see Eq. (3)], the smallest amount by which the curve has to be shifted for plasmon excitation to occur $[\Delta(\hbar k_L)_{\min}]$ increases with decreasing ion velocity. For the experimentally observed threshold energy of 2 keV, $\Delta(\hbar k_L)_{\min}$ amounts to approximately $2(\hbar k_F)$. The corresponding momentum of the plasmon, on the other hand, decreases with decreasing ion velocity. This is consistent with the observations shown in Fig. 4, which suggest that with increasing proton energy plasmons with higher momenta are excited. As already discussed for the angular distributions shown in Fig. 5, however, the observed energy shifts are too large to be solely accounted for by excitation and decay of plasmons with momenta smaller than the cutoff momentum $k_c$. These larger shifts may be understood if one assumes that the emitted electrons are accelerated in the field of the receding protons. It also may be that plasmons with momenta larger than $k_c$ are excited. In fact, the excitations then should not be understood as plasmons but as a kind of a resonance that enhances the probability for electron-hole pair excitations. These different possible contributions to the energy shifts are presently under study.

From the above considerations on energy and momentum conservation, the occurrence of a threshold at a proton energy of 2 keV cannot be explained: When either a single recoil atom or one or more phonons are involved in the excitation process, in principle there is no limitation on the amount of momentum that can be transferred. It therefore seems that for understanding the threshold behavior a careful study of the excitation process itself, including angular scattering of the proton, has to be performed. Possible contributions of other excitations of the electron gas, like electron-hole pair excitations, should also be considered. This is beyond the scope of the present publication. It should be noted, however, that the value for the minimum momentum transfer at the threshold energy of 2 keV is rather close to $\hbar G$, with $G$ the reciprocal-lattice vector in the closest-packed direction of Al(111). This also suggests that exchange of a reciprocal-lattice vector during plasmon excitation may play a role. Finally, one should realize that we have only considered minimum momentum transfer and that the above simple constructions only yield reasonable values for $\Delta(\hbar k_L)$ if virtually no momentum is transferred perpendicular to the proton momentum.

In conclusion, we have performed experiments that strongly suggest that plasmon excitation on Al(111) occurs by protons with energies down to 2 keV. This energy is more than an order of magnitude below the generally accepted threshold of 40 keV. We have given an explanation which implies that the excess momentum is transferred to the Al target. In order to understand the mechanism in more detail as well as its threshold behavior, more experimental as well as theoretical studies are needed.

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