Sub-threshold plasmon excitation in free-electron metals by helium ions

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Abstract

In this work, we re-examine sub-threshold plasmon excitation by ions measuring the energy distributions of electrons emitted from Al and Mg under the impact of 0.16–4 keV single charged Helium ions. Electron energy distributions for the case of He⁺ projectiles contain well distinguished structures that confirm the excitation of multipole surface plasmons, due to the potential energy released by the neutralization of incoming ions. It is shown that electron emission from multipole plasmon decay is more intense than from Auger neutralization (AN). Our experiments indicate that scattering and cascade in the solid of electrons excited by plasmon decay and by AN may give an important contribution to electron emission.

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1. Introduction

Plasmon excitation in ion-solid interaction, for projectile velocities below the threshold for direct Coulomb excitation, is currently the subject of intense scrutiny and has been recently reviewed [1]. Plasmon excitation has been experimentally identified by the characteristic structure produced in the energy distribution of electrons emitted from the solid, when the plasmon decays by valence electron excitation. Plasmon structures have been observed for the first time by Baragiola and Dukes in the energy distributions of electrons emitted by the free-electron metals Al and Mg under slow noble gas ions impact [2]. Since then, several investigations [3–7] have been performed to study plasmon excitation by slow ions. The main conclusion of these studies is that the dominant mechanism for plasmon production is indirect excitation by fast secondary electrons travelling inside the solid. For slow projectile ions that do not penetrate inside the bulk, plasmon excitation is attributed to the transfer of the potential energy released upon neutralization of the incoming ions.
and leading to production of multipole surface plasmon [1,8]. This mechanism thus competes with the well known Auger neutralization (AN) process [9] and plasmons of energy $E_{pl}$ can be excited if $E_n \approx E_{pl}$, where $E_n$ is the potential energy released by ion neutralization. An important question, that has not yet been answered, is the relative contribution of these processes to electron emission. In recent experiments of electron emission in the interaction of spin-polarized He$^+$ ions with Al surfaces [2], potential plasmon excitation has been discarded as an important mechanism for ion neutralization.

In this work, we report on experiments of electron emission from Al and Mg surfaces under the impact of 0.16–4 keV He$^+$ ions, aimed at establishing the relative importance of different electron emission mechanisms. We show that in the spectra of electron emitted from Al by slow He$^+$ projectiles the plasmon and the AN structures are distinguishable, though strongly overlapped. The situation is clearer in the case of the spectra of electron emission from Mg surfaces, where the plasmon and the AN structure are clearly separated in energy. Our experiments indicate that electron emission from plasmon decay is more important than AN and the observed plasmon structures are consistent with the attribution to a multipole surface plasmon excited by potential energy transfer. Above $\sim$0.8 keV impact energy, we observe bulk plasmon excitation by fast electrons travelling inside the solid.

2. Experiments

The experiments were performed in a UHV (base pressure $\sim 1 \times 10^{-10}$ Torr) system described in [2]. The double pass cylindrical mirror energy spectrometer was operated at a constant pass energy of 50 eV and a resolution of 0.2 eV. Electron energy are referred to the vacuum level of the sample. The surface of the sample was normal to the axis of the spectrometer and at 12° with respect to the ion beam direction. Ions were produced in an electron impact source, which was operated at an electron energy of 58 eV. The polycrystalline Al and Mg surfaces were sputter cleaned by 4 keV Ar$^+$ ion bombardment and cleaning was monitored by ion and electron induced Auger electron spectroscopy.

3. Results and discussion

In Fig. 1, we report energy distributions $N(E)$ of electrons emitted by the Al surface bombarded by...
160–360 eV He$^+$ ions. The energy spectra are compared with those induced by 1 keV electrons. The spectrum induced by electron bombardment shows the two shoulders attributed to decay of low momentum $q$ bulk and surface plasmons [11]. These structures are better visualized in the derivative of the spectrum, $dN/dE$, where they result in the minima at energies $E_m = E_{pl} - \Phi$, 6.3 and 11.2 eV, corresponding respectively to surface and bulk plasmon ($\Phi = 4.3$ eV is the work function for polycrystalline Al). The ion induced spectra compare well with previous results [2]. The derivative of the spectra show two structures labelled $a$ and $b$. The energy of the structure $a$ is lower than the bulk plasmon decay structure observed in the electron induced spectrum and it is close to the analogous structure observed in the case of low energy Ne$^+$ impact on Al [7]. For this reason, it is attributed to decay of multipole plasmons, excited by the potential energy released when the incoming ions are neutralized by electron capture. The energy of the structure $b$ is consistent with the energy $E_b = I' - 2\Phi$ (here, $I'$ is the ionization potential of the parent atom shifted by the image interaction) expected for the high energy edge of the spectrum of the electrons emitted from the Al surface by AN. This attribution is confirmed by the observation that the electron energy distributions $N(E)$ broaden with increasing incident ion energy (see inset in Fig. 1). This broadening is typical of neutralization via Auger processes and results from the atomic energy level shift near the surface and incomplete adiabaticity caused by the ion velocity normal to the surface [12]. Furthermore, the spectra shown in Fig. 1 intersect at a *magical* point [13], another characteristic of velocity broadened AN spectra, that corresponds to the structure $b$ observed in the derivative.

The distinction between AN and plasmon assisted neutralization appears clearer in the case of experiments on Mg samples. In Fig. 2, it is reported the energy distribution of electrons emitted by Mg under the impact of 260 eV He$^+$ ions. In this case, AN results in the broad distribution observed in the 10–20 eV energy range. This structure is well separated from the plasmon decay structure appearing in the 4–8 eV energy range.

The spectrum shows also a dominating low energy peak, which is indicative of an electronic collision cascade inside the solid.

The energy distribution $N(E)$ of emitted electrons can be written as $N(E) = T(E)N_0(E)$, where $N_0(E)$ is the internal energy distribution, i.e. the spectrum of electrons excited in the solid at energy $E$ above the vacuum level, and $T(E)$ is the surface
transmission function, giving the probability for an electron of excitation energy $E$ to be transmitted through the surface. As shown in Fig. 2, three processes contribute to $N(E)$: electron cascade, plasmon decay and AN. Thus, we have $N_0(E) = N_{0K}(E) + N_{0p}(E) + N_{0AN}(E)$, where $N_{0K}$, $N_{0p}$, $N_{0AN}$ are the internal energy distribution of electrons excited respectively by the three processes.

Fig. 2 shows that we are able to reproduce the experimental spectrum $N(E)$ using a suitable choice for $T(E)$ as Eq. (16) of [14] and by modelling the three internal distributions in the simplest fashion.

As in [15], for $N_{0p}(E)$ we have considered the convolution of a parabolic density of states with a lorentzian of $\sim 1.7$ eV FWHM, consistent with the width of the plasmon structures that we observed in electron energy loss experiments.

$N_{0AN}(E)$ has been modelled as the self-convolution of a parabolic density of states broadened by a lorentzian function [12] whose width was set in the range of a few tenths of electronvolts. This simple model is not able to reproduce the observed broadening of the AN structure, which would require a more sophisticated analysis. However, to keep things on the simplest ground, we have neglected this discrepancy, since it does not introduce a significant error in the estimation of the area of the structure.

For $N_{0K}(E)$, we have found that a simple exponential function of the type $a^{-bE}$ yields a good reproduction of the cascade peak, whereas other functional forms for $N_{0K}(E)$, as well for the surface transmission function $T(E)$, can also reproduce the experimental spectrum, without significant changes in the areas of the three structures.

The analysis of the spectrum shows that the area of the plasmon structure is about twice that of the AN structure. Furthermore, $N_{0p}(E)$ shows a high energy edge (at $E = E_m$) that is consistent with the expectation of multipole plasmon decay (the multipole plasmon energy is $\sim 0.9E_{vp}$, where $E_{vp}$ is the $q = 0$ bulk plasmon energy). This is also shown in Fig. 3, where we report the derivative of the spectra of electrons emitted by Mg under the impact of He$^+$ ions at different incident energy. At the lower incident energies, we observe that the plasmon shoulder results in the minimum in the derivative at about 6 eV, i.e about 1 eV less than the $q = 0$ Mg bulk plasmon (10.6 eV minus $\Phi = 3.7$ eV for Mg). At incident energies greater than 4 keV we observe bulk plasmon excitation, while at intermediate energies we observe the coexistence of both plasmon structures. The result shown in Fig. 3 is analogous to what has been observed in
the case of experiments of Ne$^+$ ions impact on Al [7], and confirms that bulk plasmons are excited by fast electrons traveling inside the solid.

4. Conclusions

This work reports advances in the understanding of the mechanisms for plasmon excitation at energies below the threshold for direct Coulomb excitation. Consistently with previous studies, low momentum bulk plasmons are observed for incident energies in the kinetic electron emission regime [14], in which electron excitation is mostly determined by the transfer of the kinetic energy of the projectiles. At lower energies, where electron emission is mainly determined by the transfer of the potential energy released when incoming ions neutralize, we observe the excitation of multipole surface plasmons. In contrast with a recent work [10], we find that our experimental data cannot be explained without considering the contribution of potential plasmon excitation to ion neutralization at free electron metal surfaces. For Mg, and likely also for Al, this contribution is more important than AN. An important issue, that emerges from this work, is the observation of a collision cascade peak in the energy distributions of emitted electrons, which in the case of Mg samples accounts for more than 60% of the total electron emission yield. Electronic collision cascade is generally neglected in potential electron emission, since electrons excited by potential energy transfer have low energies. It is possible that part of the cascade peak in the energy distributions of emitted electrons is due to kinetic electron excitation. On the other side, it was already suggested [11] that scattering and cascade in the solid of electrons excited by plasmon decay are significant in electron emission from free electron metal surfaces under electron impact. The role of the electronic collision cascade inside the solid in potential electron emission cannot thus be excluded and calls for detailed investigations.

References