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Neutralization mechanisms in He$^+$–Al surface collisions

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Abstract

From a quantum mechanical calculation where the populations of He ground and first excited states are properly taken into account, we can identify for the first time the neutralization to the He first excited state as an operative mechanism in He$^+$–Al surface collisions. This identification allows us to understand the presence of high energy electrons in the ion induced electron emission spectra, through the inclusion of Auger deexcitation as an electron emission source, as well as to suggest a possible cause for the disagreement still found between theory and experiments in low energy ion scattering (LEIS) for this system.

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Keywords: Ion scattering; Electron emission; Auger; Aluminum

1. Introduction

The interaction of low energy ions with solids is a complex process where several different basic physical mechanisms are involved. It is the basis of some of the most important surface characterization techniques, being the quality of such techniques determined by the balance of these mechanisms. For instance, the analysis of the mass of the sputtered ions is the basis of secondary ion mass spectrometry (SIMS), probably the most sensitive surface technique and the basis in all related microelectronic devices analysis. On the other hand, the analysis of the energy of the scattered projectiles is the basis of ion scattering spectrometry (ISS), a very surface sensitive technique. In both these techniques, the ion survival probability is related to their sensitivity and the knowledge of the neutralization mechanisms is related to our capability of quantification.

Auger process is the most usual mechanism of neutralization of the impinging ions [1]. In this process, the Coulomb repulsion between two electrons in the solid forces one of them to tunnel, through the surface barrier, to fill the incoming hole. If the energy of this hole is larger than twice the solid work function, this process is accompanied by the emission of one electron. When the incoming ion level falls within the valence band, the neutralization is performed through a tunneling mechanism without energy exchange, known as resonant neutralization [2].

Being aluminum (Al) a prototypical free-electron metal and helium (He) the simplest noble gas, it is natural that the He$^+$/Al surface collision system is usually chosen as a model system for ion surface collision problems [3–6]. Since the empty He$^+$ ground state level falls below the bottom of the Al valence band, no resonant neutralization to this level is expected, turning the Auger process in the obvious neutralization mechanism. However, a full quantum mechanical calculation of the neutralization probability applied to the LEIS regime based only on AN had shown major discrepancies with the experiments [7,8]. The inclusion of resonant neutralization (RN) to He ground state and reionization (RI) processes, operative at close distances due to the He-1s level promotion [9], improves the agreement between theory and experiment, but it is still insufficient to completely accounts for the large neutralization of He$^+$ at the Al surface [8].

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In this work, a time-dependent quantum mechanical calculation including the neutralization to the ground and first excited states of He in a correlated way accordingly to a Coulomb blockade effect [10] is performed. We found that the resonant neutralization to the He first excited state can improve the agreement between theory and ISS experiment in the He^+/Al ISS experiments. On the other hand, the electron emission produced by the Auger deexcitation, following the neutralization to the He excited state, can account for the presence of energetic electrons in ion induced secondary electrons emission in the same system.

2. Experiments and theory

The experiments were done in a commercial surface analysis system (Perkin-Elmer SAM 590A) equipped with a single cylindrical mirror analyzer, and the base pressure in the low 10^{-10} Torr range. The sample is obtained by evaporation of pure (99.999) Al in UHV conditions. Contamination is controlled by means of Auger electron spectroscopy (AES), and the repetitiveness of secondary electron emission measurements.

In order to determine the importance of the neutralization to He excited states in the He^+/Al collision, as compared to the usually accepted mechanisms, we perform a quantum mechanical calculation that includes both neutralization channels, ground and first excited He states. Including the resonant neutralization to the first excited state requires, on the other hand, an appropriate calculation where only one electron transference (either to the He-1s or to the He-2s) is allowed, inhibiting in this way the He negative charge configuration. This kind of calculation is achieved by using a developed formalism based on the infinite-correlation approach to the Anderson Hamiltonian [10]. In a simplified picture and by considering frozen the spin component of the first electron in He^+, a second electron with the same spin component is responsible of the neutralization to the excited state ($1s{\uparrow}2s$), while a second electron with the opposite spin component is responsible of the neutralization to the ground state ($1s{\uparrow}2s{\downarrow}$). The Hamiltonian can be written as:

$$H = \sum_{k,\sigma} \varepsilon_k n_{k\sigma} + \sum_{\sigma} E_\sigma n_\sigma + \sum_{k,\sigma} [V_{k\sigma} c^{+}_{k\sigma} b^{+} c_\sigma + h.c.] + \{\text{Auger neutralization terms}\}$$

Here k denotes the solid states (the valence and the core ones), and the energies, corresponding to the 1s and 2s neutralization channels, are defined as total energy differences:

$$E_\uparrow = E_{\text{tot}}(1s^1 2s^1) - E_{\text{tot}}(1s^1), \quad E_\downarrow = E_{\text{tot}}(1s^2) - E_{\text{tot}}(1s^1)$$

The boson operators b^+, b ensure the projection on the correct subspace through the constraint relation:

$$b^+ b + \sum_\sigma n_\sigma = 1;$$

and the Auger terms refer only to the ground state neutralization. The Auger deexcitation is then calculated in a second order step by using a semiclassical approximation.

The important question of whether Auger and resonant neutralization mechanisms should be treated coherently or independently has been answered for the case of considering only the ground state of He [11]. In that case, the different ion-surface distances at which both mechanisms take place allows us to treat them separately; the Auger process within a semiclassical approximation (SCA), while keeping the quantum character of the resonant process. The same conclusion is not evident in the present case, where a resonant neutralization to a more spatially extended state is involved. In this case, a full quantum calculation including all the processes is required, mainly for low kinetic energies where Auger neutralization becomes more important. In the present work, in order to explore if this mechanism may be the responsible of the main experimental features, we treat the Auger process within the SCA and perform the quantum calculation only for the resonant processes involving the ground and excited states. Within this model, the ion survival probability is given by:

$$P^+ = (1 - \langle n_\uparrow \rangle - \langle n_\downarrow \rangle) * P^+_{\text{Auger}}$$

3. Results and discussion

For a kinetic energy of 5 keV, considering a perpendicular trajectory and the scattering by only one surface atom, we obtain $\langle n_\uparrow = 0.12 \rangle$ and $\langle n_\downarrow = 0.14 \rangle$, which lead to a P^+ value around 0.45. The calculation involving only the resonant neutralization to the ground state gives $\langle n_\downarrow = 0.04 \rangle$. The interaction with Al-core states has a negligible effect for this kinetic energy, meaning that there is no time enough to allow for the promotion of projectile levels. From the point of view of ISS experiments [12], the calculation including both neutralization channels achieves a better agreement with the experimental trends. The interference between the 1s and 2s channels, originated in a Coulomb blockade effect, changes substantially the ground state neutralization when including the excited state channel. In the same way, interference effects changing the different contributions to the neutralization are expected when resonant and Auger mechanisms are simultaneously taken into account. This very complex calculation is left for a future work.

In Fig. 1 we show the population of the excited state for 5 keV He^+(1a), and the evolution of $E_\uparrow$, $E_\downarrow$ energies calculated as in Refs. [7,8], as a function of the distance from the surface (1b). We can see the downshift of the energy of the excited state below the Fermi level, which makes possible a significant probability of neutralization of this state. This channel is opened at distances larger than the ones operative for the ground state neutralization due to its more extended character, and the occupation becomes important at distances closer to the surface due to the energy resonance with the Al valence band. The excited state neutralization occurring at large ion distances from the surface limits the ground state occupation due to the Coulomb blockade effect associated to the energetically non-favorable negative configuration.

The appreciable population of the excited state obtained in our calculations predicts the existence of high energy electrons

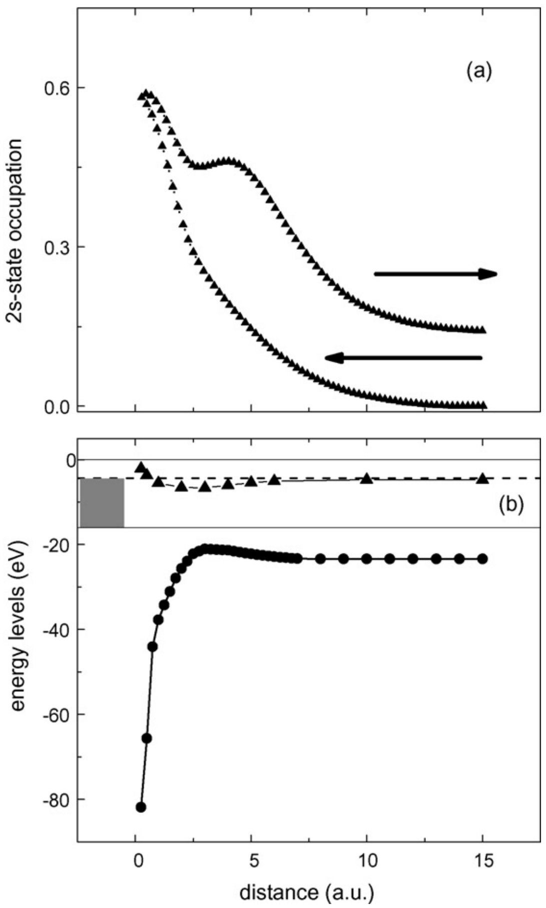

Fig. 1. (a) Evolution of neutralization probability to the first He excited state (upper panel). The arrows indicate the incoming and outgoing trajectories. (b) One-electron energies $E_\uparrow$ (full triangles) and $E_\downarrow$ (full circles). The shadowed area indicates the Al valence band.

in the secondary emission, just due to the Auger deexcitation produced among the shifted levels shown in Fig. 1.

In Fig. 2 we compare the measured electron emission spectra for 5 and 2 keV He^+ impinging ions with the electron spectrum calculated within the SCA as

$$\frac{dN(E)}{dt} = [\Delta_s \times \rho_s(E_\uparrow - E_\downarrow + E) + \Delta_p \times \rho_p(E_\uparrow - E_\downarrow + E)] \times \langle n_\uparrow \rangle \times f_\prec(E_\uparrow - E_\downarrow + E)$$

where $\rho_{s(p)}(\varepsilon_k)$ is the partial density of states of the Al surface, $f_\prec(\varepsilon_k)$ is the Fermi function and $\Delta_{s(p)}$ is related with the transition matrix element. The vertical line included in the figure corresponds to the expected limit for the Auger Neutralization process to the ground state. It is clear, that neutralization to the excited state followed by the Auger deexcitation is needed in order to account for the presence of the energetic electrons in the ion induced spectra. The low energy peak (∼12 eV) appearing in the calculations, and not resolved experimentally, corresponds to transitions occurring at distances around 3 a.u., where an important occupation of the excited state is registered in the outgoing trajectory, and the energy difference between both levels is lower than the asymptotic value (Fig. 1b). The larger energy peak corresponds to deexcitations occurring in closer collisions where the level downshift is more pronounced. The weights of these peaks will depend on the distance dependence of $\Delta_{s(p)}$. A more rigorous calculation, where trajectory and velocity effects in the level shifts and widths are included, could justify the appearance of more energetic electrons. In the same way, smaller angle trajectories would involve longer times near to the surface increasing the relative weight of electron emission at larger energies. On the other hand, for lower ion incoming energies the increasing probability of Auger neutralization to ground state and the promotion of the He1s, which favors the resonant mechanism, will tend to suppress the emission of high energy electrons, in agreement with the experimental behavior observed in Fig. 2 when going from 5 to 2 keV.

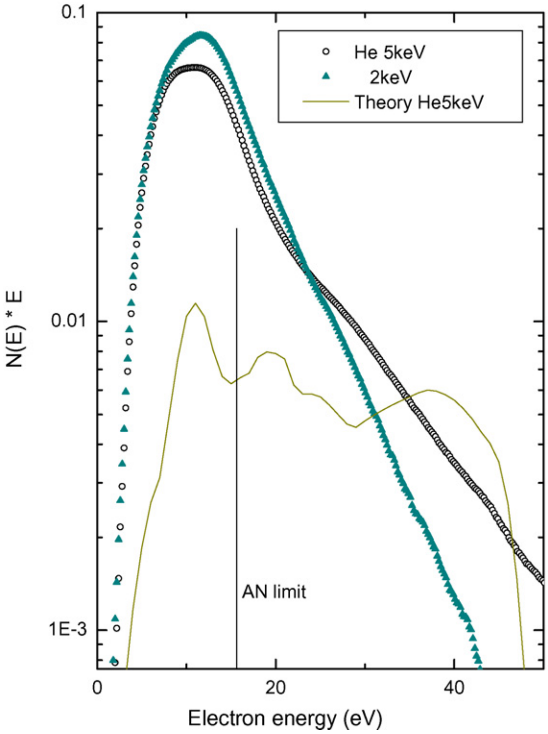

Fig. 2. Auger deexcitation electron emission (solid line) and secondary electron emission experimental results for a couple of He^+, showing the high energy electron contribution.

4. Conclusions

We found that a proper treatment of the resonant neutralization to the ground and first excited states can solve the discrepancies between theory and experiments in He^+/Al experiments on low energy ion scattering spectroscopy [7,8] and explain, through Auger deexcitation occurring at different ion surface distances, the presence of energetic electrons in the ion induced electron emission spectra of He^+ on Al.

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