Spin-dependent electron emission from metals in the neutralization of He^+ ions

M. Alducin,[1,*] J. I. Juaristi,[2,3] R. Díez Muiño,[3] M. Rösler,[1] and P. M. Echenique[1,2,3]
[1]*Donostia International Physics Center DIPC, P. Manuel de Lardizabal 4, 20018 San Sebastián, Spain*
[2]*Departamento de Física de Materiales Facultad de Químicas, UPV/EHU, Apartado 1072, 20080 San Sebastián, Spain*
[3]*Unidad de Física de Materiales Centro Mixto CSIC-UPV/EHU, Facultad de Químicas, UPV/EHU, Apartado 1072, 20080 San Sebastián, Spain*
(Received 14 January 2005; published 18 August 2005)

We calculate the spin-polarization of electrons emitted in the neutralization of He^+ ions interacting with metals. All stages of the emission process are included: the spin-dependent perturbation induced by the projectile, the excitation of electrons in Auger neutralization processes, the creation of a cascade of secondaries, and the escape of the electrons through the surface potential barrier. The model allows us to explain in quantitative terms the measured spin-polarization of the yield in the interaction of spin-polarized He^+ ions with paramagnetic surfaces, and to disentangle the role played by each of the involved mechanisms. We show that electron-electron scattering processes at the surface determine the spin-polarization of the total yield. High energy emitted electrons are the ones providing direct information on the He^+ ion neutralization process and on the electronic properties of the surface.

DOI: 10.1103/PhysRevA.72.024901 PACS number(s): 34.50.Dy, 79.20.Rf, 72.10.−d

In recent years, a large amount of experimental work has been devoted to study spin effects in the interaction of atomic particles with metal surfaces [1–13]. Low velocity spin-polarized He^+ ions and He^* metastable atoms are especially appealing as projectiles, due to the absence of resonant electron capture processes from the valence band of the metal to the $1s$ bound state of the incident particle [1–4,9–13]. In their interaction with the target, He projectiles are neutralized or deexcited via Auger processes that involve excitation and emission of electrons from the surface. Analysis of the emitted electrons provides information on the electronic properties of the surface, as well as on the local magnetization induced in the target by the spin-polarized projectile. The singularity of this technique relies on its extreme surface sensitivity. For instance, it allows one to probe surface magnetism in ferromagnetic targets [1,3,4,10,11,13]. Modification of the magnetic properties of surfaces as a function of the number of deposited adlayers can also be investigated [1,3,4,13].

In spite of the large amount of knowledge already acquired from this technique, its development has been somehow limited by the lack of theoretical support for some of the measured key quantities, such as the spin-polarization of the electrons ejected as a result of the Auger process [2,3,9,12]. For paramagnetic surfaces, the energy-dependent spin-polarization of the emitted electrons has been recently measured for 10–500 eV He^+ projectiles on Al (001), Au (001), and Cu (001) [9,12]. For all these paramagnetic surfaces, a similar average polarization of emitted electrons of around a 30% was found, always parallel to the spin-orientation of the electron bound to the incoming He^+ ion. Larger values of the polarization are measured at higher emission energies. The similar behavior of the spin-polarization for all these different surfaces is not fully understood yet.

From a general point of view, no theoretical model has yet given a reliable estimation of the spin polarization of emitted electrons. The aim of this paper is to present such a model for paramagnetic targets, and to show that it is able to explain the measurements quantitatively. We consider all stages in the emission process and analyze separately their contribution to the spin-polarization of the electron spectra. We present specific calculations for the neutralization of He^+ ions inside a paramagnetic metal. First, the spin dependent perturbation represented by the ion embedded in the system is modelled using density functional theory (DFT). As a second step, the Auger neutralization rates are obtained, distinguishing the spin orientation of the electrons excited in the process. Next, a transport calculation is performed in order to obtain the cascade of secondary electrons created by the initially excited electrons. Finally, the escape of the electrons through the surface potential barrier is calculated in order to obtain the final spin-polarization of the ejected electrons. Our most important result is that the spin orientation of the emitted electrons is governed by a highly spin-dependent Auger neutralization process, but also strongly modulated by the creation of a cascade of unpolarized secondary electrons.

The screening of the spin-polarized He^+ ion and the Auger capture rates are calculated as explained in Ref. [14]. A charged impurity in a metal induces a strong rearrangement of the electrons in its vicinity, providing total screening of the charge. Since the He^+ ion constitutes a spin polarized object, the induced screening cloud is spin unbalanced even in a paramagnetic metal. We use the Kohn-Sham (KS) scheme of DFT, within the local spin density approximation, to obtain the spin-dependent electron density around the ion embedded in a three-dimensional free electron gas. The latter is described by an electron density $n_0=3/(4\pi r_s^3)$. The He^+ ion is modeled by populating just one of the KS $1s$ wave functions of the system (there is one for each spin orientation). As a convention, in the following, the spin orientation of the electron bound to the He^+ ion will be spin up. Our

*Email address: wapalocm@sq.ehu.es

results show that the screening provided by the electron gas is preferably due to spin-up electrons, as exchange effects favor spin alignment for electronic levels well separated in energy and space.

Next, we calculate the Auger capture (AC) rate, i.e., the probability of filling the unoccupied 1*s* spin-down state. The Auger process is due to the Coulomb interaction between two continuum electrons. One electron decays to the empty bound state of the He^+ ion, whereas the second electron is promoted to an excited state. Although the spin of the decaying electron is fixed (spin-down), both spin orientations are allowed for the one that is excited. Therefore, two different channels contribute to the total AC probability $\Gamma=\Gamma^{\uparrow}+\Gamma^{\downarrow}$: that associated to the excitation of a spin-up electron $\Gamma^{\uparrow}$ and that associated to the excitation of a spin-down electron $\Gamma^{\downarrow}$. The AC probabilities are calculated in first order perturbation theory. In the excitation of a spin-down electron, the two electrons participating in the process have the same spin and are indistinguishable. As a consequence, a destructive interference term appears in the calculation of $\Gamma^{\downarrow}$. This term can also be understood from the reduced probability of finding two electrons with the same spin close to each other in the conduction band of the metal.

The difference between $\Gamma^{\uparrow}$ and $\Gamma^{\downarrow}$ rules the spin-polarization of the excited electrons in the AC process. This difference is the result of two joint effects: the spin-dependent screening and the interference term. Both favor the excitation of spin-up electrons. As a result, the spin-polarization of the excited electrons is very large (70–90% in the range $r_s=2-5$ a.u.). The interference term plays the dominant role in this strong spin dependence. In this respect, a surface calculation presented in Ref. [9] underestimates the polarization of the excitation due to the neglect of this interference term. The large spin polarization of the Auger rates is not related to the bulk nature of the model. Surface calculations of the Auger rates including the spin-dependent perturbation induced by the ion [15] and without including it [16] showed similar high values of the spin polarization.

Although the calculated spin orientation of the excited electrons is consistent with that measured in the experiments of Refs. [9,12], the measured value is much lower. Only at high emission energies, calculated and measured values of the polarization roughly coincide. Therefore, the lower measured spin polarization must be related to different mechanisms not related to the Auger process itself. Next, we show that a cascade of secondary electrons is the main responsible for the drop of spin polarization in the yield.

We use the theoretical model of Ref. [17] to calculate the transport of electrons in bulk from the energy distributions of the initially excited Auger electrons for each spin direction σ, $S^{\sigma}(E)=d\Gamma^{\sigma}/dE$. The final distribution of inner-excited electrons $N^{\sigma}(E)$ is different from $S^{\sigma}(E)$ due to electron-electron scattering events. We neglect any spin dependence in the electron-electron scattering potential. In a single scattering of an excited electron with energy E and spin σ with the target electrons there are two ways in which an electron with energy E' may appear. First, the electron (E,σ) may loss the amount of energy $E-E'$, decaying to the state (E',σ) with probability $W^{sc}(E',E)$. Second, the electron (E,σ) decays and may excite an electron from the conduction band of the target to the state E', with probability $W^{xc}(E',E)$. In this second case, the electron with energy E' can have any spin orientation with equal probability. $W^{sc}(E',E)$ and $W^{xc}(E',E)$ are calculated using the Lindhard dielectric function of the medium. The distribution $N^{\sigma}(E)$ is finally calculated using the Boltzmann transport equation formalism. In our case, this involves to solve self-consistently the following set of equations:

$$\frac{v(E)}{l(E)}N^{\sigma}(E)=S^{\sigma}(E)+\int_{E}^{E_{max}}dE'\left(W^{sc}(E,E')+\frac{1}{2}W^{xc}(E,E')\right)N^{\sigma}(E')+\frac{1}{2}\int_{E}^{E_{max}}dE'\,W^{xc}(E,E')N^{-\sigma}(E'),\quad \sigma=\uparrow,\downarrow, \tag{1}$$

where E_{max} is the maximum energy of initially excited Auger electrons, and $l(E)$ and $v(E)$ are the inelastic mean free path and the velocity of the electron, respectively. These equations are solved iteratively so that we can also obtain the energy distribution of excited electrons after a fixed number of scattering events.

Finally, the escape process is described using the standard model of a planar surface barrier [17]. The surface barrier height $W=E_F+\Phi$ is defined by the Fermi energy E_F and the work function Φ. The final energy distribution of electrons emitted after crossing the surface barrier is denoted by $I^{\sigma}(E)$, with E referred to the vacuum level.

Figure 1 shows the results of our complete calculation for the distribution $I^{\sigma}(E)$ of electrons emitted when a spin polarized He^+ ion is neutralized. The value of the work function used is $\Phi=4.25$ eV. Different values of the embedding-medium electronic density are considered. Results obtained after one and two scattering events, and also from the self-consistent solutions of Eqs. (1) are presented. Additionally, we provide also results for the directly emitted electrons $[W^{sc}(E,E')=W^{xc}(E,E')=0$ in Eqs. (1)]. The inclusion of transport has very different consequences in the spectra depending on the energy range considered. At low energies, the number of spin-up and spin-down electrons increases with the number of considered scattering events. Information on the neutralization process is therefore masked by the cascade of secondary electrons excited in electron-electron scattering processes. At high emission energies the number of emitted spin-up and spin-down electrons hardly changes with the transport process. High energy electrons are directly emitted after excitation without suffering further scattering events and they provide direct information on the ion neutralization process.

The change in the spin-polarization of the emitted electrons due to the transport process can be further analyzed by means of the quantity

$$P(E)=\frac{\int_{E}^{E_{max}}dE'[I^{\uparrow}(E')-I^{\downarrow}(E')]}{\int_{E}^{E_{max}}dE'[I^{\uparrow}(E')+I^{\downarrow}(E')]}, \tag{2}$$

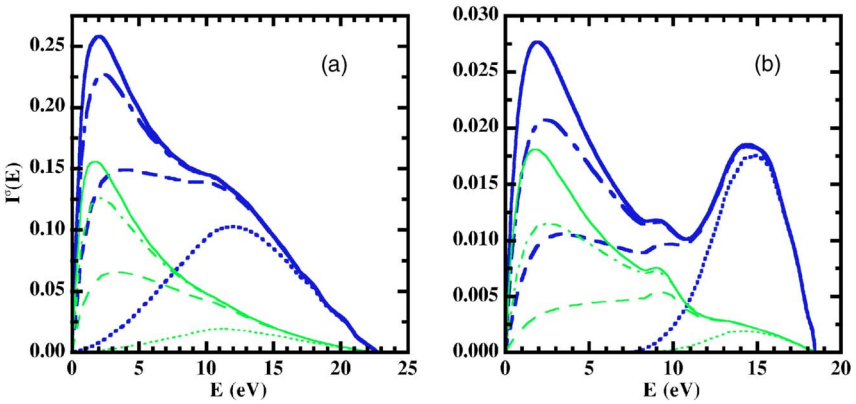

FIG. 1. (Color online) Energy distribution of emitted electrons in the neutralization of a spin polarized He^+ ion embedded in a free electron gas with electronic density: (a) $r_s=2$ and (b) $r_s=3$. Electron energy is measured from the vacuum level. Thick blue (thin green) lines correspond to electrons with spin parallel (antiparallel) to that of the electron bound to the He^+ ion. Dotted lines correspond to the distributions of directly emitted electrons, dashed lines to a single-scattering approach, dash-dotted lines to the twofold scattering approximation, and solid lines to the complete self-consistent transport calculation.

shown in Fig. 2. $P(E)$ is the polarization of the electrons emitted with energy higher than E. In the high energy range, for which the transport plays a negligible role, the polarization is high reflecting the high efficiency of the Auger process for exciting spin-up electrons. For $E=0$, $P(E=0)$ is the total polarization of the electron yield. Due to the transport process, $P(0)$ is much reduced as compared to the polarization of the initially excited electrons. For the sake of comparison, we show in Fig. 2 the experimental data of Ref. [12] obtained for Al (001). The agreement between our results and the experimental data is remarkable, showing that the theoretical model captures well the main features of the problem.

The surface of a metal consists in a region of varying electronic density whereas in a bulk calculation like ours the background electronic density is uniform. Reasons for which such a theoretical approach is valid and accurately reproduces the experimental data follow. In the experiments of Refs. [9,12] the perpendicular energy of the projectile is high enough so that the ion probes a region very close to the topmost layer. Based on different calculations for the distance dependent Auger neutralization rates for this system [18–20], it was estimated that neutralization should typically occur at 2–3 a.u. from the topmost atomic layer. This is very close to the jellium edge. At these distances, the relative electronic density variations are small, and a local model based on a uniform density background is reasonable.

An additional point is that, since the distance from the surface at which the ions are neutralized may vary, there exists some uncertainty about the value of the r_s parameter to be used. An important outcome of our calculation is that the spin-polarization of emitted electrons does not depend significantly on the choice of r_s. This is shown in Fig. 2, in which we observe that a similar good agreement with the experimental data can be obtained for different values of r_s. Therefore, we conclude that the knowledge of the exact neutralization distance and of the corresponding electronic density probed by the projectile is not so important in order to explain the measured spin-polarization, provided that the Auger neutralization takes place at short distances from the surface and that the surface electronic density is not too low. One may even invoke a better agreement with the data for Al (001) using $r_s=3$, which is a reasonable value for the electronic density in the surface region.

The weak dependence of the electron-yield spin polarization on the electronic density explains as well the similar experimental results obtained for different surfaces such as Al (001), Au (001) and Cu (001) [12]. The measured polarization of the yield is of the order of 30% in all three cases. In Fig. 3 we show the calculated total polarization of the yield [$P(E=0)$ in Eq. (2)] for several values of the density parameter r_s. Experimental uncertainty for this quantity is shown as well. Our calculations give values of the polarization consistent with the experiment in the range $r_s=1-4$. If the transport process were neglected, the value of the calculated spin polarization would be much larger in general. Finally, we remark the similar results obtained for the spin-polarization of the emission when considering two scattering events or all orders of scattering in the transport calculation

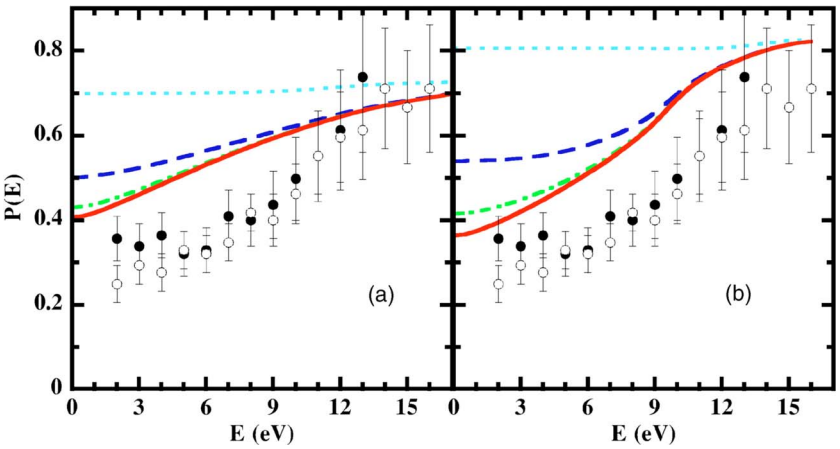

FIG. 2. (Color online) Polarization of the electrons emitted with energy larger than E as defined by Eq. (2). The electron density of the embedding medium is (a) $r_s=2$ and (b) $r_s=3$. Results obtained for directly emitted electrons (cyan dotted lines), for a single-scattering approach (blue dashed lines), for a twofold scattering approach (green dash-dotted lines), and for the complete self-consistent transport calculation (red solid lines) are shown. Experimental data of Ref. [12] for the polarization of the electrons emitted in the neutralization of 15 eV (black circles) and 500 eV (white circles) He^+ ions incident on Al (100) are shown in both panels.

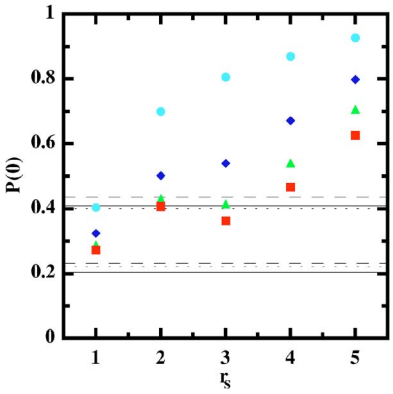

FIG. 3. (Color online) Polarization of the total yield of emitted electrons [$P(0)$ from Eq. (2)] as a function of the density parameter r_s. Cyan circles correspond to the directly emitted electrons, blue diamonds to a single scattering approach, green triangles to the twofold scattering approximation and red squares to the complete transport calculation. The lines limit, according to the error bars of Ref. [12], the experimental uncertainty for He^+ incident on Al (100) (solid lines), Cu (100) (dotted lines), and Au (100) (dashed lines).

(see Figs. 2 and 3). Electrons excited in the surface region may undergo a limited number of scattering events before being emitted, though it is not completely clear the exact number of scattering events one should consider. The similarity of our results taking either two orders or all orders of scattering allows us to overcome this uncertainty when interpreting experimental spectra.

In summary, we have analyzed all different mechanisms involved in the emission of electrons after neutralization of spin-polarized He^+ ions in paramagnetic metals. This study has allowed us to characterize unambiguously the sources of spin-polarization of the emission and its energy dependence. We have shown that the transport process is responsible of reducing the average spin-polarization to values of around 30%. This result depends weakly on the target electronic density, and it is expected to be valid for those paramagnetic metal surfaces with typical electronic densities $r_s<4$, provided the Auger process takes place close enough to the surface. Additionally, the strong spin-dependence of the Auger process is reflected in that of the electrons emitted at high energies. Finally, let us mention that the transport may play a different role in ferromagnetic surfaces, in which the excitation depends as well on the spin-orientation of the incoming projectile relative to that of the target. Nevertheless, electrons emitted at high energies are expected to enclose information on the spin-dependent properties of the ion/surface interaction in the same way they do in paramagnetic surfaces.

We acknowledge partial support by the Basque Departamento de Educación, Universidades e Investigación, the University of the Basque Country UPV/EHU (Grant No. 9/UPV 00206.215-13639/2001) and the Spanish MCyT (Grant Nos. BFM2001-0076 and FIS2004-06490-CO3-00). M.A. acknowledges financial support by the Gipuzkoako Foru Aldundia. The authors want to express their gratitude to Dr. C. Sarasola.

[1] M. Onellion, M. W. Hart, F. B. Dunning, and G. K. Walters, Phys. Rev. Lett. **52**, 380 (1984).
[2] M. W. Hart, M. S. Hammond, F. B. Dunning, and G. K. Walters, Phys. Rev. B **39**, R5488 (1989).
[3] M. S. Hammond, F. B. Dunning, G. K. Walters, and G. A. Prinz, Phys. Rev. B **45**, 3674 (1992).
[4] M. Salvietti, R. Moroni, P. Ferro, M. Canepa, and L. Mattera, Phys. Rev. B **54**, 14758 (1996).
[5] M. Schleberger, M. Dirska, J. Manske, and A. Närmann, Appl. Phys. Lett. **71**, 3156 (1997).
[6] A. Närmann, M. Dirska, J. Manske, G. Lubinski, M. Schleberger, and R. Hoekstra, Surf. Sci. **398**, 84 (1998).
[7] T. Igel, R. Pfandzelter, and H. Winter, Phys. Rev. B **59**, 3318 (1999).
[8] R. Pfandzelter, T. Bernhard, and H. Winter, Phys. Rev. Lett. **86**, 4152 (2001).
[9] D. L. Bixler, J. C. Lancaster, F. J. Kontur, P. Nordlander, G. K. Walters, and F. B. Dunning, Phys. Rev. B **60**, 9082 (1999).
[10] R. Moroni, E. Oliveri, and L. Mattera, Nucl. Instrum. Methods Phys. Res. B **203**, 29 (2003).
[11] M. Kurahashi, T. Suzuki, X. Ju, and Y. Yamauchi, Phys. Rev. B **67**, 024407 (2003).
[12] J. C. Lancaster, F. J. Kontur, G. K. Walters, and F. B. Dunning, Phys. Rev. B **67**, 115413 (2003).
[13] M. Kurahashi, T. Suzuki, X. Ju, and Y. Yamauchi, Phys. Rev. Lett. **91**, 267203 (2003).
[14] M. Alducin, R. Díez Muiño, and J. I. Juaristi, Phys. Rev. A **70**, 012901 (2004).
[15] N. Bonini, G. P. Brivio, and M. I. Trioni, Phys. Rev. B **68**, 035408 (2003).
[16] L. A. Salmi, Phys. Rev. B **46**, 4180 (1992).
[17] M. Rösler and W. Brauer, *Particle Induced Electron Emission I*, Springer Tracts in Modern Physics Vol. 122 (Springer, New York, Heidelberg, 1991), p. 1.
[18] N. P. Wang, E. A. García, R. Monreal, F. Flores, E. C. Goldberg, H. H. Brongersma, and P. Bauer, Phys. Rev. A **64**, 012901 (2001).
[19] N. Lorente, M. A. Cazalilla, J. P. Gauyacq, D. Teillet-Billy, and P. M. Echenique, Surf. Sci. **411**, L888 (1998).
[20] M. A. Cazalilla, N. Lorente, R. D. Muiño, J. P. Gauyacq, D. Teillet-Billy, and P. M. Echenique, Phys. Rev. B **58**, 13991 (1998).