Stopping Power for Helium in Aluminum

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The relative contributions to the stopping power of the different charge states for a beam of helium ions moving with velocity v in aluminum are shown, for the first time, from a first-principles calculation. Combining dielectric and density-functional results in the appropriate velocity range with the energy lost in the capture and loss processes we have obtained good agreement with experiment.

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The problem of the energy loss of ions moving through solids has attracted the attention of physicists since the beginning of the century. In the high-velocity regime the ion is stripped of its electronic charge and the theories of Bethe[1] and Bloch[2] of electronic stopping give a good description of the energy-loss process. At low velocities the ion is surrounded by a cloud of polarization charge and one must take into account the perturbation introduced in the medium by the incoming ion. After the pioneering work of Fermi and Teller,[3] some calculations were done within the framework of dielectric theory,[4] the binary encounter approximation,[5] and with different model potentials.[6] The density-functional theory allows one to calculate the induced screened potential, the density fluctuation, and the energy loss at low speeds in a self-consistent way. The first explicit calculations of the stopping power in this formalism were performed by Echenique, Nieminen, and Ritchie[7] for hydrogen and helium, and later were extended to higher ionic charges.[8]

At intermediate velocities there is no rigorous theory of the stopping power for ions moving through solids, even in the case of light ions. Some calculations have been done[9] with different degrees of agreement with the available, scattered, experimental data in the energy region near the stopping-power maximum. Effective-charge theories have been proposed[10] to explain the stopping data. This effective charge is related to the mean occupation number of the ion's bound states. But in the effective-charge approach nothing is said either about the energy loss in charge-changing events or about the fractional stopping power associated with each charge state. Separating the different contributions to the stopping power has, besides its intrinsic theoretical and experimental interest,[11] relevance to fields such as secondary-electron emission.[12]

We have calculated the stopping power for He in aluminum as a function of the intruder velocity with explicit inclusion of solid-state effects to calculate the charge-state distribution inside the medium. In our approach, linear-response theory in the dielectric formalism is combined with many-body techniques to calculate the energy loss per unit path length. The main assumptions of our model are as follows.

(i) A bound level is well defined over the whole range of He velocities. In the static case bound levels appear at metallic densities, and at high speeds, where the stripping probability is high, atomiclike states may be occupied since electrons in the conduction band do not have enough time to screen the ion.[13] This can be easily seen by calculating the energy of an electron bound to a He nucleus through a screened Coulomb potential with a velocity-dependent screening parameter that reproduces the static and high-velocity screening limits. One finds that the extension of the wave function is not very different from the $1s$ wave function of a He atom in vacuum and it is smaller than the lattice constant of Al ($a \cong 4$ Å).

(ii) The equilibrium charge-state fractions of bare ions (ϕ^{++}), singly ionized ions (ϕ^{+}), and neutral atoms (ϕ^{0}) as determined by the different mechanisms discussed below are given in terms of the probabilities per unit time of capturing and losing the electrons as

$$\phi^{++} = \Gamma_{\mathrm{loss}}(\mathrm{He}^{+})\Gamma_{\mathrm{loss}}(\mathrm{He}^{0})D^{-1}, \tag{1a}$$

$$\phi^{+} = \Gamma_{\mathrm{loss}}(\mathrm{He}^{0})\Gamma_{\mathrm{capt}}(\mathrm{He}^{++})D^{-1}, \tag{1b}$$

$$\phi^{0} = \Gamma_{\mathrm{capt}}(\mathrm{He}^{+})\Gamma_{\mathrm{capt}}(\mathrm{He}^{++})D^{-1}, \tag{1c}$$

where

$$D = \Gamma_{\mathrm{loss}}(\mathrm{He}^{+})\Gamma_{\mathrm{loss}}(\mathrm{He}^{0}) + \Gamma_{\mathrm{loss}}(\mathrm{He}^{0})\Gamma_{\mathrm{capt}}(\mathrm{He}^{++}) + \Gamma_{\mathrm{capt}}(\mathrm{He}^{+})\Gamma_{\mathrm{capt}}(\mathrm{He}^{++}).$$

In these equations $\Gamma_{\mathrm{loss}}(\mathrm{He}^{0})$ and $\Gamma_{\mathrm{loss}}(\mathrm{He}^{+})$ are the probabilities per unit time of losing an electron by the

neutral atom and by the single ionized ion, respectively, while $\Gamma_{\text{capt}}(\text{He}^{++})$ and $\Gamma_{\text{capt}}(\text{He}^{+})$ are the corresponding probabilities per unit time of capturing an electron by the bare helium ion and the single ionized one. We have neglected double-electron capture by the bare ion and double-electron loss by the neutral atom.

(iii) The stopping power is calculated by summing the stopping powers for each charge state weighted by the respective charge-state fractions, and adding the energy loss per unit length in capture and loss processes. Thus we write for the stopping power

$$\left.\frac{dE}{dx}\right|^{\text{total}} = \phi^{++}\frac{dE}{dx}(\text{He}^{++}) + \phi^{+}\frac{dE}{dx}(\text{He}^{+}) + \phi^{0}\frac{dE}{dx}(\text{He}^{0}) + \phi^{++}\frac{dE^{C}}{dx}(\text{He}^{++}) + \phi^{+}\left[\frac{dE^{L}}{dx}(\text{He}^{+}) + \frac{dE^{C}}{dx}(\text{He}^{+})\right] + \phi^{0}\frac{dE^{L}}{dx}(\text{He}^{0}). \tag{2}$$

In Eq. (2), dE^{C}/dx and dE^{L}/dx are the energy losses per unit path length in the capture and loss processes [see Eqs. (4) and (5) below]. The stopping power for bare ions $[dE/dx(\text{He}^{++})]$ and singly ionized ions $[dE/dx(\text{He}^{+})]$, relevant at high and intermediate velocities, is calculated in linear theory.[4] The contribution of inner-shell excitations ($2p$ and $2s$ shell electrons of Al) to the stopping power for bare ions has been taken into account using standard methods.[14] Energy loss by the He^{+} ion is computed accounting for the interaction of the charge densities of the electron and the He nucleus with the medium. For the He^{0} charge state (relevant at low velocities) we have calculated the stopping power $[dE/dx(\text{He}^{0})]$ using the phase shifts at the Fermi level $[\delta_{l}(E_F)]$ for scattering of electrons by the self-consistent potential, created by the He, calculated in density-functional theory. It is given by (atomic units are used in this paper)[7,8]

$$\frac{dE}{dx}(\text{He}^{0}) = \frac{3v}{k_F r_s^3}\sum_{l=0}^{\infty}(l+1)\sin^2[\delta_l(E_F) - \delta_{l+1}(E_F)], \tag{3}$$

where v is the ion speed, r_s is the one-electron radius, and k_F is the Fermi momentum of the electrons ($k_F = 1.92/r_s$). This way of calculating the stopping power is strictly valid in the $v \to 0$ limit, but a comparison by Mann and Brandt[15] of experimental data of the stopping power for protons in twenty different elemental solids shows that the linear dependence on velocity holds up to $v \cong v_F$ ($\cong 1$ a.u. in Al). It could be argued that our calculation overestimates the stopping power of He^{0} for intermediate velocities ($1 < v < 2$), while at the same time it underestimates the stopping power of He^{++} and He^{+}. One could expect, however, that both effects tend to compensate, as the results of the total stopping power (see Fig. 1) suggest.

Several mechanisms can result in an electron being captured into a bound state of the moving ion: (i) An electron can make an atomiclike transition from a bound state of an atom in the lattice to a bound state of the moving ion. We shall refer to it as a shell process henceforth. It is relevant at high velocities. (ii) The ion moving through the lattice feels a time-dependent potential of characteristic frequency $\omega \cong v/a$ (a being the lattice constant) which may result in a transition of an electron from the conduction band to the bound level of the ion; we call these resonant processes. (iii) A direct capture of an electron from the conduction band assisted by creation of a plasmon or an electron-hole pair may also occur. These, denoted as Auger processes, give the greatest contribution to the capture cross section at low velocities. Auger and resonant processes may also result in the loss of an electron bound to the moving singly ion-

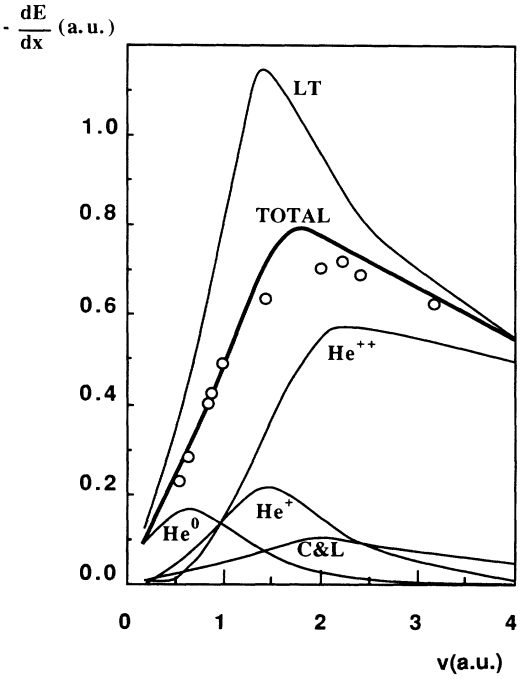

FIG. 1. Stopping power in atomic units of Al for helium ions as a function of ion speed. The thick solid line (TOTAL) is the result of our calculation and the curve labeled LT is obtained from linear-response theory for a bare ion. Both of them include inner-shell corrections. The circles are the experimental data. The different contributions to the curve labeled TOTAL from the fractions of bare ions (He^{++}), singly ionized ions (He^{+}), neutral atoms (He^{0}), and capture and loss processes (C&L) are shown separately.

ized ion (He^+) or neutral atom (He^0). The shell capture cross sections are calculated in the Brinkman-Kramers approximation with a reduction factor fixed by the experimentally measured charge-state fractions at high velocities.[16] The reduction factor that we have used is 6, a value consistent with Eichler's calculation.[17] The Auger and resonant capture and loss cross sections are found from the imaginary part of the self-energy associated with the He-nucleus–bound-electron composite.[18]

The energy loss per unit path length in the shell capture process is obtained[19] in terms of the target atomic density, the energy of the electron bound in the nth shell of the Al atom e_n, the binding energy of the captured electron e_0, and the cross section σ_{n0} for the $n \rightarrow 0$ transition. In the resonant capture and loss processes it is given by

$$\frac{dE_{\text{res}}^{C,L}}{dx} = \frac{2\pi D_s}{v} \sum_{\mathbf{G}} \sum_{|\mathbf{k}+\mathbf{v}| \lesssim k_F} (\mathbf{G}\cdot\mathbf{v}) |V(\mathbf{G})|^2 |\langle s|e^{i\mathbf{G}\cdot\mathbf{r}}|\mathbf{k}\rangle|^2 \delta(E_{\mathbf{k}0} \pm \mathbf{G}\cdot\mathbf{v}) , \tag{4}$$

and for the Auger capture and loss processes we obtain

$$\frac{dE_{\text{Auger}}^{C,L}}{dx} = \frac{2D_s}{v} \sum_{|\mathbf{k}+\mathbf{v}| \lesssim k_F} \int d\omega \int \frac{d\mathbf{q}}{(2\pi)^3} (\mathbf{q}\cdot\mathbf{v}) \frac{4\pi}{q^2} \text{Im}\left\{\frac{-1}{\varepsilon(q,\omega)}\right\} |M_{\mathbf{k}0}|^2 \delta(\omega \mp \Delta E) . \tag{5}$$

In Eq. (4), $\mathbf{G}$ is a reciprocal-lattice vector, $V(\mathbf{G})$ the Fourier transform of the effective potential acting on the incoming electron that we have approximated by a Hartre screened pseudopotential, and $E_{\mathbf{k}0} = E_b + k^2/2$. In Eq. (5), $M_{\mathbf{k}0} = \langle s|e^{i\mathbf{q}\cdot\mathbf{r}}|\mathbf{k}\rangle$ and $\Delta E = \mathbf{q}\cdot\mathbf{v} + E_b + k^2/2$. $-E_b$ and $|s\rangle$ are the binding energy and wave function of the bound electron that we have calculated variationally by a minimization procedure. We denote by $|\mathbf{k}\rangle$ a plane wave orthogonalized to the state $|s\rangle$, thus describing electron states in the conduction band accounting for the high nonlinearity of the electron-gas–ion interaction $\varepsilon(q,\omega)$ is the response function of the electron gas that we have approximated by a random-phase-approximation (RPA) response function.[4] D_s takes into account the spin degeneracy of the electron states; i.e., in the capture of an electron by the bare ion or in the loss by the neutral atom, $D_s = 2$, while in the capture and loss of an electron by the singly ionized ion, $D_s = 1$.

The results obtained from Eq. (2) for the stopping power of He ions moving with velocity v in Al are shown in Fig. 1 as the thick solid line (labeled TOTAL). The curve labeled LT is the stopping power for bare ions calculated in linear theory using an RPA dielectric function[4] to represent valence-electron excitations in aluminum, with $r_s = 2$. Inner-shell corrections from the $2s$ and $2p$ electrons of the Al ions have also been included.[14] The different contributions to the curve labeled TOTAL have been separated to show the relevance of the various terms as a function of ion speed. When $v \approx 1$ a.u. the contributions from the neutral-atom (He^0) fraction, from the singly-ionized-ion (He^+) fraction and bare-ion (He^{++}) fraction are comparable, each being about 30% of the total, while that of charge-exchange (capture and loss) processes is about 10%. The contribution from the inelastic processes of capture and loss is about 15% at $v = 2$. At high velocities the stopping of the bare ion dominates, as it must, and at low velocities the neutral-atom energy loss is the biggest contribution. The circles are the experimental data from Refs. 20 and 21.

A final comment related to the choice of helium as the projectile rather than the more obvious choice of an apparently simpler projectile, namely, hydrogen, should be made. In the case of helium the bound states are well defined and so it is a clear case to illustrate our new perspective in the difficult problem involving the stopping of ions having low to moderate velocities. For protons, some difficulties arise related to the existence of the H^- state. The existence of a state in the solid having the character of H^- does not imply that the state is similar to the free negative ion; it is a state that can exist as a result of the interaction of the projectile with the solid. In the metallic density range, capture and loss cross sections for the H^- have to be included in the model. We have performed such a calculation for the case of protons moving in aluminum. The results of our calculation agree well with the experimental data and will be published elsewhere. For higher electron-gas densities the bound-state character disappears and the modeling becomes much more complicated since one has to include the treatment for scattering from continuous resonant states.

In conclusion, the electronic stopping power of aluminum metal for He has been calculated with explicit inclusion of the different charge-state populations inside the medium. The total stopping power is obtained by weighing the appropriately calculated stopping powers with the respective charge-state fractions and adding the energy loss per unit path length due to electronic-exchange processes. The relative contribution to the stopping power from capture and loss processes may be 15% for the case of He moving in aluminum. Good agreement with experimental data is found.

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