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Stopping Power for Protons in Aluminum.

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Abstract. – The electronic stopping power of aluminum metal for protons has been calculated with explicit account taken of the different charge states of the photon inside the medium. The fraction of negative ions (H^-) neutral atoms (H^0) and bare protons (H^+) in the beam as a function of ion speed are derived from the capture and loss cross-sections. The total stopping power is then obtained by weighting the calculated partial stopping powers with the appropriate charge state fractions. The energy loss per unit path length due to electronic exchange processes is also evaluated. Our calculations show that the relative contribution to the stopping power from capture and loss processes is of the order of 15% for the case of protons moving in aluminum. Good agreement with experimental data is found.

The energy loss of ions in solids is well understood in the high [1] and low [2] velocity regimes. At intermediate velocities, things are much more complicated due to the important charge-exchange processes. In a simplified approach empirical, effective charge theories [3] have been proposed to account for the mean charge state of the ion as a function of its velocity. The processes by which a partially stripped ion interacts with a target are, however, numerous and complex, and the development of first principles methods to calculate the ion charge state is only reasonable for light atoms [4]. The knowledge of proton stopping power over a wide range of velocities, besides its intrinsic theoretical interest, provides a helpful tool in the effective charge description of heavier-ion stopping power. Note that the calculation of ion charge states says nothing about energy loss in charge-changing events, a contribution that must be included in the calculation of the ion stopping power.

In a recent publication [5], we evaluated the stopping power for He in Al over the whole range of nonrelativistic velocities. The energy losses for the different He charge states were determined together with the probabilities of finding the different charge states; charge-changing contributions to the stopping power were also evaluated. In the present

paper, we present theoretical values of the stopping power for protons in Al. This theory is more complicated than that for the He projectile. On the one hand, the H charge states are not as well defined as for He, and on the other hand the calculation of the partial stopping powers of the different H charge states is more complex. Atomic units are used in this paper unless otherwise stated.

The underlying physics of our model can be described as follows:

i) We assume that a bound level that can be doubly occupied is well defined over the whole range of proton velocities. This is based on several different calculations for a static proton inside a metal. Local Density Approximation (LDA) theory [6] shows the existence of a H^- ion, sorrounded by positive charge localized in the metal conduction band. It has been argued that in the LDA the energy levels are meaningless, and that only the total density has a physical meaning. However, a self-energy approach [7] has shown that an H^- state exists for a proton inside an electron gas. Moreover, Nørskov [8] has found using a LDA approach that the H^- configuration in Al has an energy 9 eV lower than the one for H^0, with a single electron bound to the proton. Recent Linear Combination of Atomic Orbitals (LCAO) calculations [9] have also shown that this is the case and proved that the main mechanism for hydrogen adsorption on simple metals is associated with the lowering of the hydrogen affinity level due to its electrostatic interaction with the metal atoms. As the ion velocity increases, the atomiclike states for H are not so strongly screened by the conduction band electrons. This increases their binding energies and yields an atomic wave function which is more contracted the larger the velocity is. In our calculation we shall use the binding energies and wave functions given in ref. [10].

ii) The equilibrium charge state fractions are calculated in terms of the capture and loss rates that involve all the mechanisms described below. We neglect double-electron capture by H^+ and double-electron loss by H^-.

iii) A weighted sum of the partial stopping powers of the different charge states plus the energy loss due to the charge exchange processes gives the total stopping power.

Three processes that can result in an electron transition are considered here [11]:

i) The periodic potential of the lattice is seen by the incident ion as a time-dependent perturbation of characteristic frequency $\omega \simeq v/a$, where a is the lattice constant and v is the ion-speed. This perturbation can induce transitions between states in the conduction band of the solid and bound states of the ion. We refer to these as resonant processes. In the case of protons our calculations show that the probability of resonant capture can be neglected compared with other capture processes.

ii) Transitions can occur between states of the conduction band and bound states of the ion, assisted by a third body (creation of a plasmon or an electron-hole pair). We call these Auger processes and find them to dominate at speeds $v < 1.0$.

iii) When the velocity of the ion is high, an atomiclike transition can take place from a bound state of a lattice ion to a bound state of the projectile. These are termed shell capture processes.

We calculate the probability of an electron capture or loss event by a resonant or Auger process from the imaginary part of the self-energy associated with the proton-bound electron composite [12], calculated in a GW approximation [13]. The capture cross-sections for shell processes have been calculated in the Brinkman-Kramers approximation [14] with a reduction factor $\beta = 4.5$ to produce agreement with Eichler's results [15].

The stopping power for hydrogen projectiles is written as

$$\frac{dE}{dR}(H) = \phi^+ \frac{dE}{dR}(H^+) + \phi^0 \frac{dE}{dR}(H^0) + \phi^- \frac{dE}{dR}(H^-) + \\ + \left[\phi^+ \frac{dE^C}{dR}(H^+) + \phi^0\left(\frac{dE^C}{dR}(H^0) + \frac{dE^L}{dR}(H^0)\right)\phi^- \frac{dE^L}{dR}(H^-)\right]. \quad (1)$$

In eq. (1) ϕ^+, ϕ^0 and ϕ^- are the equilibrium charge state fractions. dE^C/dR and dE^L/dR are the energy losses per unit path length in the capture and loss processes, respectively (see eqs. (4), (5) and (6) below). dE/dR (H^+), dE/dR (H^0) and dE/dR (H^-) are the energy losses of bare protons, neutral atoms and negative ions, respectively (also called partial stopping powers). In general, we consider for each case the total charge, nuclear plus electronic, moving with the atom. The electronic stopping power of Al for H projectiles with energies below 100 keV/u is dominated by valence electron excitation and ionization. In our model this corresponds to electron-hole pair and plasmon excitations of a free-electron gas with one electron radius $r_s = 2$. We have calculated the energy losses due to electron-hole pair and plasmons excitations using different approximations. Plasmons can only be excited at velocities $v > 1.3$ in Al; a regime where linear response theory is quite accurate for a unit charge. At lower velocities only pair excitations can be created in the metal, and nonlinear effects in the screening are important. dE/dR (H^-) is obtained by using its low-velocity limit (energy loss proportional to ion speed) with a transport cross-section calculated from the LDA phase shifts at the Fermi level [5]. The justification for this approximation is that the H^- charge state fraction is only relevant for $v < 1.0$, a velocity for which plasmons are not yet excited. We have calculated (dE/dR) (H^0) and (dE/dR) (H^+) by obtaining independently contributions from the electron-hole pairs and from the plasmons. We have also included the L-shell contribution to the stopping power of bare protons calculated in the first Born approximation. The stopping power associated with close collisions (electron-hole pairs excitation) is obtained using a binary encounter approximation [16]. Thus

$$\frac{dE}{dR}(v) = \frac{1}{4\pi^2}\frac{1}{v^2}\int_0^{v_F} u\,du \int_{|v-u|}^{v+u} dv_r\, v_r^4\, \sigma^{tr}(v_r)\left[1 + \frac{v^2 - u^2}{v_r^2}\right], \quad (2)$$

where v_F is the Fermi velocity, v is the ion velocity and σ^{tr} is the transport cross-section evaluated at the relative velocity v_r. We describe the interaction between an electron and the atom in terms of a spherically symmetric potential. This potential is obtained by solving the Poisson equation for a point charge with a spherically symmetric charge density around it, screened in the Thomas-Fermi way with a velocity-dependent screening parameter that interpolates between the high- and low-velocity limits. Using this short-range potential, we calculate the transport cross-section and the close collision contribution to the stopping power. The transport cross-section is given by

$$\sigma^{tr} = \frac{4\pi}{v_r^2}\sum_{\lambda=0}^{\lambda_{max}} (\lambda+1)\sin^2(\delta_\lambda - \delta_{\lambda+1}), \quad (3)$$

where $\lambda_{max} = 2vv_F/\omega_p$ and δ_λ are the phase-shifts at the relative velocity. The plasmon contribution is obtained in linear theory using the Random Phase Approximation (RPA) dielectric function (nonlinear effects are only important at low ion velocities). This contribution verifies the equipartition rule at high velocities.

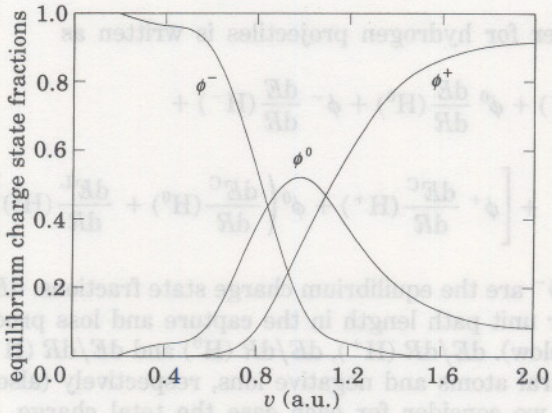

Fig. 1. – Equilibrium charge state fractions of bare protons (ϕ^+), neutral atoms (ϕ^0) and negative ions (ϕ^-) in the beam as a function of ion speed (v).

The energy loss due to the charge exchange Auger and resonant processes is given by [5]

$$\frac{\mathrm{d}E^{\mathrm{C,L}}_{\mathrm{Auger}}}{\mathrm{d}R} = \frac{2D_{\mathrm{S}}}{v} \sum_{|\boldsymbol{k}+\boldsymbol{v}| \gtrless k_{\mathrm{F}}} \int \mathrm{d}\omega \int \frac{\mathrm{d}^3\boldsymbol{q}}{(2\pi)^3} (\boldsymbol{q}(-,+)\boldsymbol{k})\cdot\boldsymbol{v}\,\frac{4\pi}{q^2}\cdot$$

$$\cdot \mathrm{Im}\left\{-\frac{1}{\varepsilon(\boldsymbol{q},\omega)}\right\} |M_{k,0}(\boldsymbol{q})|^2\,\delta^{\mathrm{C,L}}(\omega - \boldsymbol{q}\cdot\boldsymbol{v}(-,+)E_{k,0}), \tag{4}$$

$$\frac{\mathrm{d}E^{\mathrm{L}}_{\mathrm{Resonant}}}{\mathrm{d}R} = \frac{2\pi D_{\mathrm{S}}}{v} \sum_{\boldsymbol{G}} \sum_{|\boldsymbol{k}+\boldsymbol{v}| > k_{\mathrm{F}}} |V(\boldsymbol{G})|^2 (\boldsymbol{G}+\boldsymbol{k})\cdot\boldsymbol{v}\,|M_{k,0}(\boldsymbol{G})|^2\,\delta(\boldsymbol{G}\cdot\boldsymbol{v} - E_{k,0}) \tag{5}$$

and for the shell capture process:

$$\frac{\mathrm{d}E^{\mathrm{C}}_{\mathrm{Shell}}}{\mathrm{d}R} = n_{\mathrm{at}} \sum_n \left(E_n - E_{\mathrm{b}} + \frac{1}{2}v^2\right)\sigma_{n,0}\,. \tag{6}$$

In these equations $\boldsymbol{qv}$ and $\boldsymbol{Gv}$ are the contributions to the change in the kinetic energy of the centre of mass of the ion-bound electron composite and $\boldsymbol{kv}$ is the energy transferred to the exchanced electron. D_{S} takes into account the spin degeneracy. $V(\boldsymbol{G})$ is the Fourier transform of the crystal pseudopotential [17] and $\boldsymbol{G}$ is a reciprocal lattice vector. $E_{k,0} = E_{\mathrm{b}} + k^2/2$, $M_{k,0}(\boldsymbol{q}) = \langle s|\exp[i\boldsymbol{qr}]|\boldsymbol{k}\rangle$ and $-E_{\mathrm{b}}$ and $|s\rangle$ are the binding energy and wave function of the bound electron. $|\boldsymbol{k}\rangle$ is the state vector of a plane wave orthogonalized to $|s\rangle$ and $\varepsilon(q,\omega)$ is the RPA dielectric function. E_n is the energy of the electron bound to the n-th shell of the Al atom, and $\sigma_{n,0}$ is the capture cross-section.

In fig. 1 we show the equilibrium charge state fractions for a proton in aluminum as a function of the proton velocity. The H^- charge state is significant up to velocities around v_{F}. This can be understood using the Bohr stripping criterion. Even when the bound state is very shallow the effective barrier is the Fermi energy, so the Pauli exclusion principle (PEP) prevents an efficient stripping up to the Fermi velocity. At high velocities ($v \approx 2.0$) the fraction of bare ions has the largest value but there are still 10% neutral atoms. The highest fraction of H^0 ($\approx 50\%$) is reached at velocities roughly equal to the Fermi velocity. Our equilibrium charge states are appropriate only inside the solid. Charge states are measured outside the solid; this means that the crossing of the surface can be important in determining

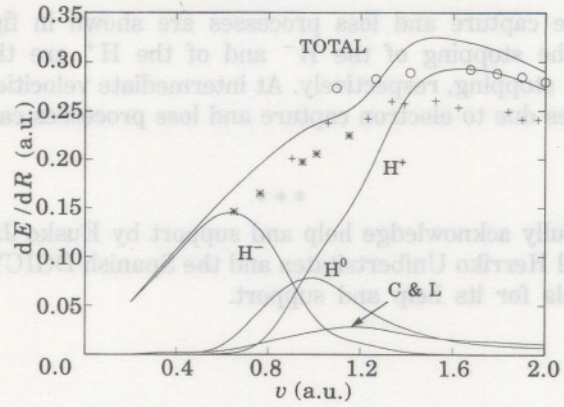

Fig. 2. – Stopping power for protons moving in aluminum as a function of ion speed. The different contributions from the fractions of bare protons (H^+), neutral atoms (H^0), negative ions (H^-), and capture and loss processes (C&L) are shown separately. The curve labeled TOTAL is the sum of all the others. The crosses are data from ref. [20], the dots are from ref. [21] and the stars from ref. [22].

the measured distributions especially at low ion velocities. We find that in the solid the mean charge of the proton is lower than in gaseous targets [18] at the same velocity. This is the opposite of the density effect found for heavy ions [19], in which the excitation of electrons bound to the projectile leads to losses in the solid with a high probability, unlike the situation in gases. However, in the case of light ions (H or He) at low and intermediate velocities, screening prevents the existence of excited states inside the solid and electron transitions between weakly bound levels of the projectile and the continuum states in the valence band of the solid are more sensitive to the restriction of the PEP.

Figure 2 shows in the curve labeled TOTAL results calculated from eq. (1) for the stopping power of protons in aluminum *vs.* the ion velocity. The partial contributions of the different charge states (H^-, H^0, H^+) and that of the charge exchange processes (C&L) are presented as well. Some experimental data [20-22] are also shown. Our results are in good agreement with the experimental data considering the rather large dispersion in the measured values. The relative contributions to the total stopping power from the different

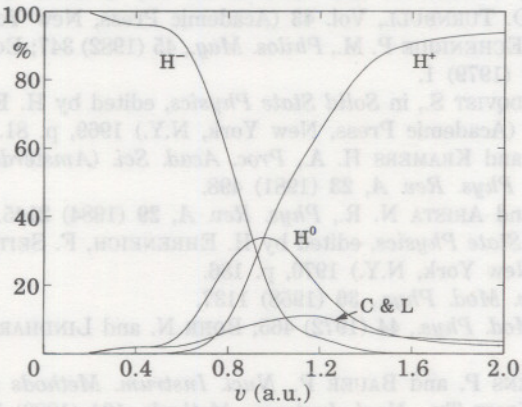

Fig. 3. – Relative contributions to the stopping power as a function of ion speed from bare protons (H^+), neutral atoms (H^0), negative ions (H^-), and capture and loss processes (C&L).

charge states and the capture and loss processes are shown in fig. 3. In the low- and high-velocity limits, the stopping of the H^- and of the H^+ are the ones which mainly contribute to the total stopping, respectively. At intermediate velocities all contributions are relevant. Energy losses due to electron capture and loss processes can be about 15% of the total at $v \approx v_0$.

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