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Inelastic proton-solid collisions

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A first-principles calculation of charge states of moving protons in Al is presented. The many-body self-energy approach combined with ordinary atomic physics has been used. We find that at high velocities, $V > 2V_0$ or $3V_0$ (Bohr velocity), the processes are atomiclike, while at intermediate velocities, $0.7V_0 < V < 2V_0$ or $3V_0$, solid-state effects are responsible for the proton charges.

Since the pioneering work by Phillips[1] the charge states of moving protons emerging from condensed-matter medium have been experimentally measured over a wide range of proton energies, ranging from 25 keV to the megaelectronvolts region.[2–4] The physical processes determining the charge states are of complicated nature and difficult to treat theoretically, even in gases, particularly in the intermediate energy region.[5] In solids an important extra difficulty arises, namely the role of screening associated with electrons in the valence or conduction band. *At low velocities the electron gas-screening is the determinant factor fixing the charge state.* In this regime results obtained[6,7] using nonlinear density-functional theory of Hohenberg and Kohn[8] and Kohn and Sham[9] show, contrary to earlier speculations[10] based on linear theory, that static protons in an electron gas binds two electrons in a very shallow level over the whole metallic density range. When the proton is moving several mechanisms can result in an electron being captured into a bound state of the proton: (a) An electron can make a transition from a bound state of an atom in the lattice to a bound state of the moving proton. This is an atomiclike process. We shall refer to it as a *shell process.*[5] (b) The proton moving through the lattice effectively feels a time-dependent potential of frequency $\omega \approx V/a$ (a is the lattice constant) which can result in a transition to a bound state of the proton; we call this a *resonant* process.[11] (c) In addition direct capture from the valence band assisted by a third "body," i.e., plasmon on electron-hole pair emission, can occur.[12] We denote this process by electron gas capture or *Auger* capture. On the other hand Auger and resonant processes are responsible for the loss of an electron bound to a proton moving inside a crystal target. In a lucid paper, Cross has pointed out[13] that at high enough velocities neither screening by the valence electrons nor the detailed structure of the crystal should matter and therefore the charge states in solids, at those velocities, are simply given by scaling gas-target results to solid densities: the cross section per atom remaining unchanged. At intermediate velocities, however, screening and effects associated with the crystal structure are important.[10] Still the capture and loss processes can be described in terms of capture to (and loss from) bound states of the proton.[12] At low velocities screening effects are crucial.[7] A detailed analysis of the screening effects is necessary in order to achieve a good degree of quantitative agreement with experiment. In this letter we address the problem of the charge state of a proton moving in a solid, over the whole velocity range of interest, i.e., $(0-6)V_0$. We present, for the first time, the results of a detailed study of all processes described above for the case of a proton moving in an aluminum target. We concentrate mainly in the most difficult region to treat theoretically, i.e., the intermediate-energy region (15–100 keV) where the pioneering work of Phillips[1] was first performed. We also, briefly, comment on the high-velocity limit where, indeed, the ideas of Cross[13] are quantitatively confirmed. Our results, in the intermediate-energy region, also confirm the validity and insight of the work of Brandt[10,14] and co-workers[15] in stressing the relevance and importance of solid-state effects in this problem.

To make the above ideas more quantitative we begin by writing the Auger cross sections for capture and loss, per atom, σ_A^C and σ_C^L, in terms of the ion-electron composite self-energy.[12] We work in atomic unit throughout:

$$\sigma_A^{C,L} = -D^{C,L} 2\,\mathrm{Im}\Sigma_A^{C,L}/(nV) , \tag{1a}$$

where

$$\mathrm{Im}\Sigma_A^{C,L} = \int_0^\infty d\omega \int [d^3q/(2\pi)^3](4\pi/q^2)\,\mathrm{Im}[\epsilon^{-1}(q,\omega)] \sum_{k \lessgtr k_F} |\langle U_0 | e^{\mp i\mathbf{q}\cdot\mathbf{r}} | k \rangle|^2 \delta(\omega_0 - \tfrac{1}{2}(\mathbf{k}-\mathbf{q})^2 \mp \mathbf{q}\mathbf{v} \pm \omega) , \tag{1b}$$

$D^c=2$, $D^L=1$, n is the mean-target atomic density, and V the atomic velocity. In Eq. (1b), ω_0 is the binding energy of the bound state, $U_0(r)$, while $\epsilon(q,\omega)$ is the dielectric function of the medium, and $|\,k\,\rangle$ the wave function of the valence orbital, here approximated by a plane wave orthogonalized[12] to $U_0(r)$. Equations (1) have been evaluated with no approximation in the matrix element between states $|\,k\,\rangle$ and $|\,U_0\rangle$, this representing a substantial improvement over the results presented in Ref. 12.

The capture and cross sections for the resonant process can be described by similar equations. The corresponding $\mathrm{Im}\Sigma_R^{C,L}$ is given by:[11]

$$\mathrm{Im}\Sigma_R^{C,L}=\pi \sum_{k \lessgtr k_F} \sum_{G} |\,V_G\,|^2\,|\,\langle\, U_0(\mathbf{r})\,|\,e^{i\mathbf{G}\cdot\mathbf{r}}\,|\,k\,\rangle\,|^2 \times \delta(\omega_0-\tfrac{1}{2}(\mathbf{k}-\mathbf{G})^2 \mp \mathbf{G}\mathbf{V}) \;, \qquad (2)$$

where G is the reciprocal-lattice vector and V_G the Fourier transform of the effective crystal potential acting on the incoming electron. For $V \rightarrow 0$, V_G can be approximated by an Ashcroft pseudopotential,[11] while for V larger than V_0 we can expect that the incoming electron can penetrate the shell of the target ion. We have taken into account these effects by using, at a given velocity, the effective potential that can be deduced from the phase shifts calculated in LEED.[16]

The capture-shell process is evaluated by using an Oppenheimer-Brinkman-Kramers approximation.[5] This is known to overestimate the corresponding cross reactions by a factor of 4 or 5 in the range of velocities we are interested in.[5] Following Ref. 11 we take a factor of 4.5 to reduce the OBK results.

We have evaluated the capture and loss cross sections for all the processes quoted above using the self-consistent bound state, $U_0(r)$, and the bound level, ω_0, calculated in Ref. 12. In Figs. 1 and 2, to establish the energy region where the capture and loss processes are atomlike, we compare the results of our calculation of aluminum with available experimental data of other gases. Differences arise due to the fact that there are different atoms but we are mainly interested in the trends.

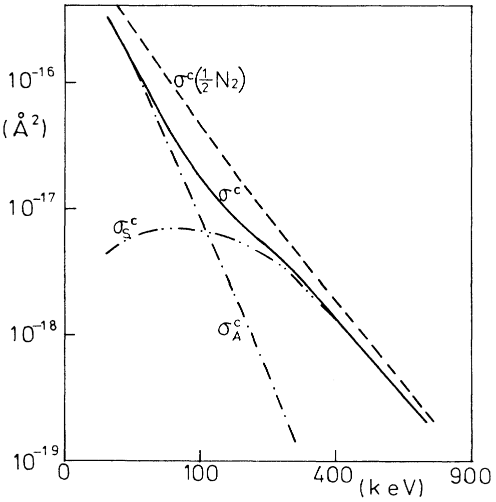

FIG. 1. Shell, — · · · —, Auger, — · —, and total ——, capture cross section for H^+ colliding with Al. For comparison, we show the capture cross section (normalized to one atom of the target) for the gas collision H^+-N_2, — — —.

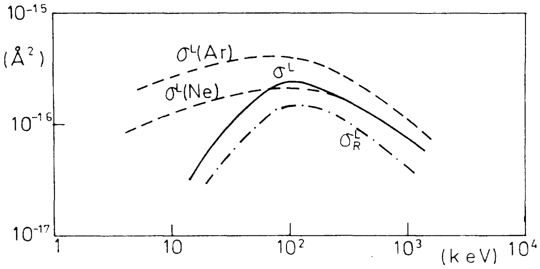

FIG. 2. Resonant, — · —, and total loss cross section for H^0 colliding with Al. For comparison, we show the loss cross section for the gas collision H^0-Ne and H^0-Ar.

Figure 1 shows the Auger and the shell-capture cross sections and the total one for H^+ (a proton) on Al (the resonant process gives a negligible contribution to this cross section). Comparison is made[17] with the case of H^+ on N_2: Figure 1 shows the loss cross section of this case normalized to a N atom. Our results show that the capture processes for H^+ on Al are atomiclike for $V > 3V_0$: in this limit, the calculated cross section is very similar to the one measured for H^+ on $\frac{1}{2}$ (N_2). For smaller velocities, the shell cross section decreases and screening processes typical of the crystal structure take over. This explains the small bump appearing in the capture cross section between $1.5V_0$ and $3V_0$ (see Fig. 1). We should comment that, in spite of those solid-state effects, the general behavior of the total cross section for H^+ on Al is very similar to the one found for collisions of H^+ on gases.

Figure 2 shows the resonant-loss cross section and the total one, as calculated for H^0 (neutral) on Al. Comparison is made[18] with the loss cross section of H^0 on Ne and Ar. We find a good agreement with the case of H^0 on Ne for $V \geq 2V_0$ showing that in this range of velocities the loss processes are atomiclike. For smaller velocities, however, the loss cross section per atom in the solid state is very much reduced with respect to the atomic case: This result is due to the Pauli principle applied to the electron gas of the conduction band. An electron bound to the moving ion cannot jump to the occupied states of the crystal conduction band.

From these comparisons we deduce that specific crystalline effects only appear in the different cross sections for $V < 2V_0$ or $3V_0$. This is, on the other hand, intimately related to the ion charge states at intermediate velocities, $V_0 < V < 2V_0$, that are very much dependent on screening and crystal effects.

Figure 3 shows our results for $n(H^+)/n(H^0) = \sigma^L(H^0)/\sigma^c(H^+)$ for velocities lower than $2V_0$. The agreement with experiments[1] for $V > V_0$ is excellent improving very much on Guinea *et al.*[12] results. This is due to a better calculation of the screening effects and to introducing shell and resonant processes.

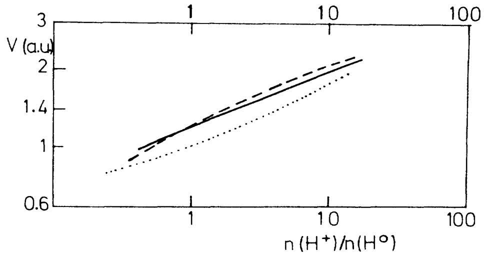

FIG. 3. Calculated values, ——, of $n(\mathrm{H}^+)/n(\mathrm{H}^0)$ as a function of the proton velocity. Experimental values, — — —, and the theoretical results of Ref. 15, · · · ·, are also shown.

For $V < V_0$, our agreement is not so good: this is related to the appearance of surface effects. Indeed, the mean free paths, associated with the total cross sections, for $V < V_0$, become very small and comparable to the surface width[19] (for $V = 0.8V_0$, the capture mean free path is only ≈ 9 a.u.).

In conclusion we have presented, for the first time, a complete calculation of the charge state of a proton moving in a solid which agrees well with the experimental data even in the intermediate-energy region where electronic screening and crystal effects are important. At very low energies ($V < 0.7V_0$) surface effects are very important and quantitative evaluation of the charge state by the methods presented here becomes a cumbersome task; qualitative trends, however, go in the same direction as the experimental data.

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