Energy loss of MeV α particles in the palladium-hydrogen system in planar channeling

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The energy loss of MeV α particles moving between (111) planar channels of the palladium-hydrogen system has been calculated for the case when the hydrogen-atom concentration is only a few at.% (α phase). The contribution due to additional electrons of hydrogen atoms is estimated by assuming that these go to the $4d$ band, and that due to protons is obtained by considering α-particle–proton scattering. The proton contribution is found to be very small compared to that due to additional electrons for well-channeled particles, which itself is about $0.001x$ of the electronic stopping power in the (111) planar channel for pure Pd, x being the atomic percentage of H in Pd.

Recent experiments[1] on dechanneling of α particles by a low concentration of hydrogen atoms in palladium have suggested that it is reasonable to describe the dechanneling in this system as "obstruction" type,[2] and that this obstruction can be described by a Rutherford-type interaction in which an α particle is scattered by the proton sitting in the channel. We calculate here the effect of these hydrogen atoms on the energy loss of those α particles which are not dechanneled but suffer an additional energy loss due to electrons and protons of hydrogen atoms.

To calculate the energy loss due to additional electrons of hydrogen atoms, we assume that these go to the d band of palladium. While for higher concentration of H atoms (in β phase), photoemission studies[3] show that the additional electrons go to H-induced low-lying states[4] as well as to the $4d$ band, for low H concentration (α phase) all of the additional electrons can be assumed to fill the holes of the d band.[5]

Next, we simplify the situation and avoid the use of complicated band structure by saying that in these calculations of channel stopping power it is enough to use the atomic wave functions. This approximation is implicit in most of the channel-stopping-power calculations to date and in any case it is not bad for nearly filled d-band transition metals. In fact the use of hydrogenlike wave functions has given very good quantitative agreements with experimental results on channel stopping power in semiconductors[6] for energetic protons. For low-velocity heavy ions also, the atomic wave functions have been used with reasonably good success in connection with Z_1 and Z_2 variations,[7] and recently for calculating the position dependence of the stopping power in planar channels of gold.[8]

The electronic stopping power in the high-velocity region is given by[9,10]

$$S_e = \frac{4\pi Z_1^2 e^4}{m v_1^2} L \quad , \tag{1}$$

where

$$L = \rho_{cond} \ln \frac{2mv_1^2}{\hbar \omega_p} + \rho_{val} \ln \frac{v_1}{v_F}$$

$$+ \rho_{loc} \ln \frac{2mv_1 v_F}{I} + \sum_j \rho_j \ln \frac{2mv_1^2}{I_j} \quad , \tag{2}$$

where e and m are electronic charge and mass, respectively, and Z_1 and v_1 are the charge and velocity of the incident particle. The first term in Eq. (2) is the conduction-electron contribution, ρ_{cond} and ω_p being the corresponding density and plasma frequency. The last term includes contribution of inner shells, ρ_j and I_j being corresponding densities and binding energies. The second and third terms in Eq. (2) represent the valence electron (d-shell) contribution. The second term corresponds to the contribution of collective excitations which use the total density of valence electrons, while the third term gives the contribution of the local electron density in the channel to single-particle excitations; v_F is the Fermi velocity, and I the binding energy of the d shell. This separation of valence-electron contribution in two parts, as suggested by Appleton et al.,[9] has been found to give good quantitative agreement with experiment for the well-channeled energetic α-particle stopping power in (111) planar channels of gold, as well as for the variation of channel stopping power with the distance from the channel wall.[10] In fact, one can easily see from Eqs. (1) and (2) that, when ρ_{loc} becomes equal to ρ_{val}, i.e., when the incoming particle encounters all the valence electrons, one gets the formula of Dettman and Robinson[10] for the valence shell.

For Pd, it is now well established, both experimentally[11] and theoretically,[12] that $Z_{cond} = 0.36$ electrons per atom and $Z_{val} = 9.64$ electrons per atom. Thus $\rho_{cond} = 0.36N$ and $\rho_{val} = 9.64N$, where N is the atomic concentration per unit volume. The contribution ρ_{loc} and that due to other inner shells ρ_j is obtained by making a planar average of the corresponding shells,[8,10] and using the one-term Slater orbitals with optimized orbital ex-

ponents given by Clementi et $al.$[13] Thus we get the electron density due to one plane at a distance y from the plane as:

$$\rho_j^{(1)}(y) = \frac{Nd_p}{2} \frac{(2\xi_j)^{2n_j}}{2n_j} \omega_j e^{-2\xi_j y}$$

$$\times \sum_{k=0}^{2n_j-1} \frac{y^{2n_j-1-k}}{(2\xi_j)^k (2n_j - 1 - k)!} , \quad (3)$$

so that the total electron density due to both planes becomes

$$\rho_j(y) = \rho_j^{(1)}(y) + \rho_j^{(1)}(d_p - y) . \quad (4)$$

Here N is the atomic concentration per unit volume, d_p the interplanar distance (so that Nd_p is atomic density per unit area of the plane); n_j, ω_j, and ξ_j are the principal quantum number, the occupation number, and the orbital exponent of jth shell, respectively.

If x is the atomic percentage of H in Pd, we get $0.01x$ electrons per additional atom in the d shell. This means that ρ_{val} changes from 0.097 (in a.u.) to $(0.097 + 0.0001x)$. Using Eqs. (3) and (4) to calculate ρ_{loc} at a point halfway between (111) planar channels, one gets $\rho_{loc} = 0.008\,57$ without H atoms and $\rho_{loc} = 0.008\,57 + 9 \times 10^{-6}x$, with H atoms. Using Eqs. (1) and (2) to calculate the corresponding stopping powers we get for 4-MeV α particles:

$$-\frac{dE}{dx} = 1.331 \ (\text{conduction electrons})$$

$$+ 14.59 \ (\text{valence electrons})$$

$$+ 0.015x \ (\text{from } x\% \text{ H atoms})$$

$$+ 0.112 \ (4p \text{ electrons})$$

$$+ 0.015 \ (4s \text{ electrons})$$

$$= (16.048 + 0.015x) \ \text{eV/Å}. \quad (5)$$

The contribution due to protons, which are known to take octahedral positions in Pd and hence to be situated halfway between (111) planes, can be obtained by considering the scattering of protons from the incoming ion (α particle in this case). As expected this contribution will be shown to be about three orders of magnitude smaller than the contribution from the additional electrons (see the Appendix).

Thus we see from Eq. (5) that the energy-loss contribution from the additional H atoms is extremely small and can not be detected for low concentration. This also shows that it is quite safe to neglect effects of additional energy loss due to H

atoms in the calculations for dechanneling. However, if by suitably adjusting temperature and pressure one introduces a high concentration of H atoms within the α phase, the additional stopping power of well-channeled particles could be measured. Such an experiment would consist in measuring the minimum energy loss of well-channeled particles [Pd (111) planar channels in the present case] without and with H atoms. It should be equally interesting to study the more complicated β phase with higher H concentration, in which case the theory will also have to be extended, because the electrons of these H atoms will start filling the new H-induced bonding states,[4] the number depending upon the H/Pd ratio and approaching 0.5 electron state per Pd at the H/Pd ratio of 1. In addition, the interaction between the protons themselves will also have to be taken into account.

APPENDIX

Let us consider the scattering of protons from the incoming α particle. Assuming that any proton velocity due to its jumping around in palladium is negligible compared to the velocity of the incoming α particle and that protons do not interact with each other (for the α phase), the energy lost to the protons (mass m_2) by the incoming α particles (mass m_1) is given by[14]

$$-\frac{dE}{dx} = \frac{n_2 m_1^2 m_2 v_1^2}{(m_1 + m_2)^2} Q_d ,$$

where n_2 is the density of protons (i.e., H concentration), Q_d is the momentum-transfer cross section for scattering of protons from the α particle, and is given by

$$Q_d = 2\pi \int_0^\pi I_0(\theta)(1 - \cos\theta) \sin\theta \, d\theta .$$

For fast α particles, the scattering is essentially from the bare charge $Z_1(= 2$ for the α particle) so that

$$I_0(\theta) = \frac{Z_1^2 e^4}{m_2^2 v^4} \frac{1}{(1 - \cos\theta)^2}$$

and one finally gets

$$-\frac{dE}{dx} = \frac{2\pi n_2 m_1^2 Z_1^2 e^4}{(m_1 + m_2)^2 m_2 v_1^2} \ln \frac{2m_1^2 m_2 v_1^2}{(m_1 + m_2)^2 I_2} ,$$

where the term in the logarithm is obtained by recalling[14] that $\theta = \pi$ in the expression for Q_d corresponds to maximum energy transfer from α particle to proton, $T_2^{max} = 2m_1^2 m_2 v_1^2/(m_1 + m_2)^2$, and the minimum θ is determined by $T_2^{min} = I_2$, where I_2 is

minimum energy that the proton can take during the scattering process (equal to the diffusion energy, 0.23 eV, for the Pd-H system). For x at.% H in Pd, one gets from the above expression for 4-MeV α particles:

$$-\frac{dE}{dx} = 1.84x \times 10^{-5} \text{ eV/Å} ,$$

which is about three orders of magnitude smaller than the corresponding electronic contribution given in Eq. (5).

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