Threshold effect in the energy-loss straggling of protons channeled in Au⟨100⟩ at very low velocities

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The energy-loss straggling of protons channeled in gold crystals in the ⟨100⟩ direction has been experimentally and theoretically studied at the low-velocity limit. The experimental results show a threshold effect in the straggling values, which is similar to that observed earlier in the energy loss. This effect is described by a theoretical model which explains the velocity dependence by considering the individual features of the contributions of free electrons in the conduction band and the nearly free ($5d^{10}$) electrons which generate a threshold effect at low energies due to their binding energy. The model predicts a deviation of proportionality with ion velocity of the energy-loss spread and yields a good description of the obtained experimental data.

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The energy-loss straggling of point particles in metals has been studied experimentally and theoretically in the past [1–8]. In a recent work [9] we showed the deviation of the energy loss ΔE from the proportionality with ion velocity for protons and deuterons channeled in Au ⟨100⟩ in the very-low velocity range, and interpreted this result with a theoretical model that takes into account the properties of the electronic band structure of the medium. This analysis has now been extended to the second moment of the energy-loss distribution showing also interesting deviations of the energy spread from the proportionality with ion velocity. The behavior is theoretically explained by separating the contributions of the ($5d^{10}6s^1$) electrons in the outer shells of Au into a free-electron (conduction) band, of mixed s,p character, and a nearly free band formed by the localized d electrons [10].

The experimental data were obtained with the equipment described in Ref. [11]. The energy-loss spread values have been determined with a dispersion of ±5 eV using the transmission technique. This small dispersion leads to a good definition of the velocity dependence. In these experiments a monocrystalline gold target with a thickness of 12 nm oriented in the ⟨100⟩ direction (mounted on a 3 mm transmission electron microscopy grid) was used, and the energy spectra of the channeled protons were determined using a rotatable energy analyzer followed by an electron multiplier.

The experimental straggling Ω^2 was corrected for instrumental resolution by

$$\Omega_0'^2 = \Omega^2 - \delta^2\varepsilon_m^2, \tag{1}$$

where δ is the detector resolution and ε_m the mean energy of ions after crossing the gold foil. The effect of foil roughness is taken into account through the well-known correction

$$\Omega_0^2 = \Omega_0'^2 - \rho^2\Delta E^2, \tag{2}$$

where Ω_0^2 is the intrinsic straggling and ρ is the roughness coefficient that in this case is approximated by 0.05. The systematic errors of the resulting Ω_0 values are of about ±15%.

In Fig. 1 we show the measured values of the energy-loss spread as a function of the average velocity $\langle v\rangle$ of the ions in the foil $[\langle v\rangle = \frac{1}{2}(v_{\text{in}}+v_{\text{out}})]$, covering the velocity range from 0.15 to 0.58 a.u. The aim of the dashed line is to show the threshold effect and to visualize the deviation of proportion-

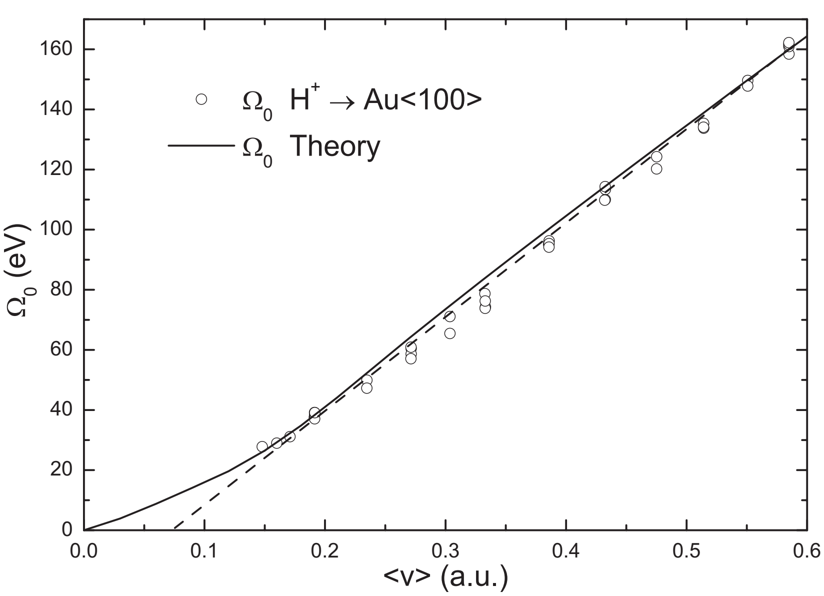

FIG. 1. Measurements of the energy-loss spread Ω_0 for protons as a function of the average velocity $\langle v\rangle$ of the ions in the gold crystal. The aim of the dashed line is to show the deviation of proportionality with ion velocity. The solid line represents the theoretical results.

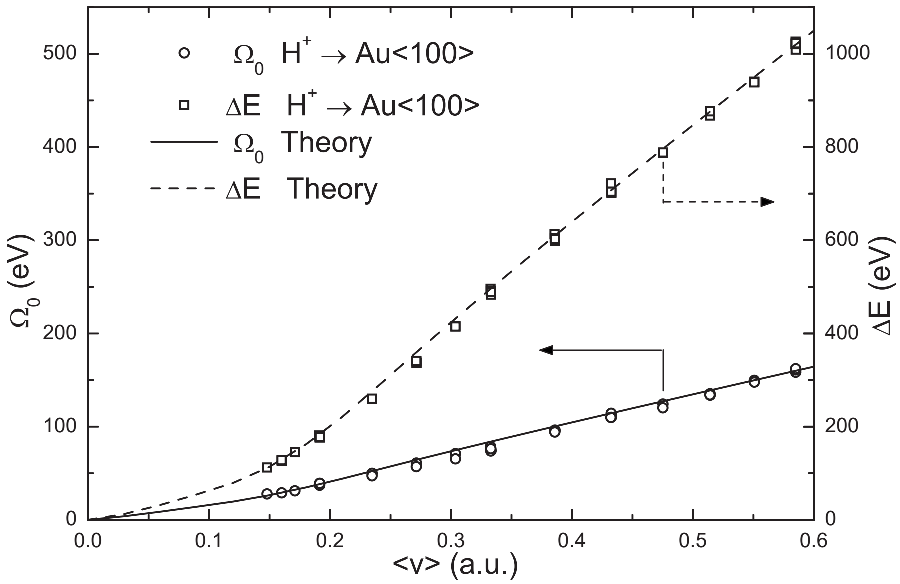

FIG. 2. The energy-loss spread Ω_0 and mean energy loss ΔE for protons as a function of the average velocity $\langle v\rangle$ of the ions in the foil. The solid line corresponds to the calculated energy-loss spread and the dashed line corresponds to the calculated mean energy loss.

ality with ion velocity. We observe from this figure a clearly displaced straight-line dependence of the data, pointing to an apparent threshold at $v\sim 0.08$. The solid line is the theoretical result to be described in the following.

According to Refs. [3,4] the energy-loss straggling for slow ions may be expressed as

$$\frac{\Omega^2}{\Delta x} = K v^2, \tag{3}$$

where Δx is the thickness of the foil and K is given by

$$K = \sum_i v_{F_i}^2 n_i \bar{\sigma}_{\mathrm{str}_i}. \tag{4}$$

In the last equation n_i ($i=1,2$) are the densities of (s,p) free electrons and nearly free d electrons, v_{F_i} are the corresponding Fermi velocities, and $\bar{\sigma}_{\mathrm{str}_i}$ are the respective straggling cross sections which are calculated extending our previous model [9],

$$\bar{\sigma}_{\mathrm{str}_i}(v) = \int_0^{\varepsilon_F} \sigma_{\mathrm{str}}(v,\varepsilon_F-\varepsilon) g_i(\varepsilon) d\varepsilon, \tag{5}$$

where $g_i(\varepsilon)$ is the corresponding density of states [12] and σ_{str} is the straggling cross section calculated including the requirement of a minimum excitation energy $U_\varepsilon=\varepsilon_F-\varepsilon$, as

$$\sigma_{\mathrm{str}}(v,U_\varepsilon) = 3\int_{\theta(v,U_\varepsilon)}^{\pi} |f(\theta)|^2 \sin^3(\theta/2) d\tilde{\Omega}, \tag{6}$$

where $d\tilde{\Omega}$ is the solid angle element. The lower limit in this integral is determined by the relation between the center of mass (c.m.) scattering angle (θ) and the energy transfer in the laboratory system [9],

$$\cos[\theta(v,U_\varepsilon)] = 1 - \frac{U_\varepsilon}{m v v_r} \tag{7}$$

and $f(\theta)$ is the scattering amplitude given by

$$f(\theta) = \frac{1}{v_r}\sum_l (2l+1)e^{i\delta_l}\sin(\delta_l)P_l(\cos\theta), \tag{8}$$

where v_r is the electron velocity in the c.m. system and δ_l is the phase shift corresponding to the scattering of waves with angular momentum l.

In this work we have used the δ_l values obtained by Puska and Nieminen [13] which arise from the density functional theory. A full integration over the densities of states corresponding to (s,p) and d electrons has been performed by using Eq. (5). The densities corresponding to the free (s,p) and d electron components in Eq. (5) were calculated using fixed values $r_s=3.01$ and $r_s=2$, respectively, as in our previous calculations [9].

As a result of the minimum excitation energy U_ε introduced here, the straggling coefficient K in Eq. (3) becomes a function of the ion velocity. The theoretical results provided by this model are shown by the solid line in Fig. 1. In order to allow a close comparison of the shape of the curve, the calculations have been normalized to the experiments at the velocity $v=0.58$ a.u. The differences between the measured values and unnormalized values calculated with this model are of the order of 20%. We observe that the characteristic displaced-straight-line behavior and the apparent threshold at $v\sim 0.08$ are very well explained by the theoretical model. The model predicts a change in the slope of the straggling curve, indicating a transition between a very low-velocity region where only free electrons (conduction band) are excited, to a second low-velocity range where also nearly free electrons are excited. This behavior is consistent with our previous observations for the energy loss [9]. In Fig. 2 we compare the behavior of both the spread Ω_0 and the mean energy-loss values ΔE [9] including also the corresponding (normalized) theoretical calculations obtained with the same model. We observe that the lowest-velocity data of Ref. [9] were not included in this figure because it was not possible to determine the corresponding straggling values with an acceptable precision due to the higher statistical requirements for this magnitude.

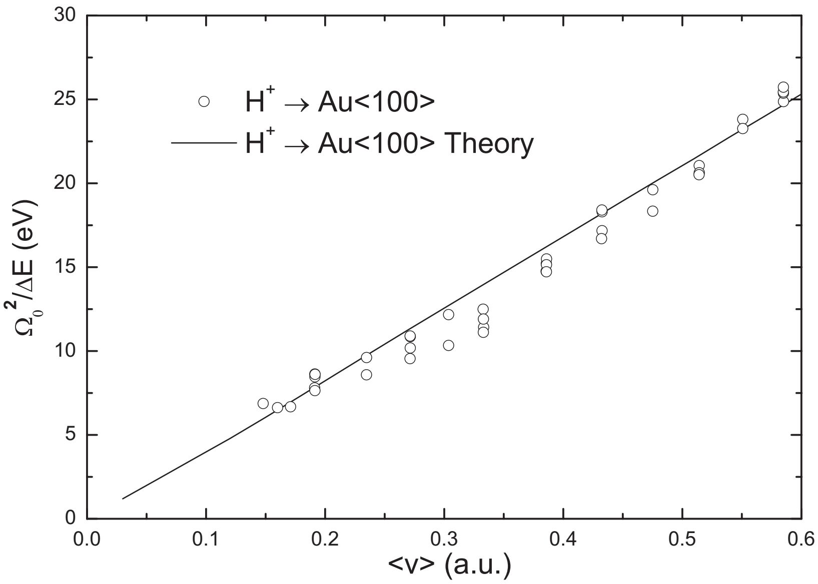

FIG. 3. The relative energy-loss straggling data for protons as a function of the average velocity of the ions channeled in a Au ⟨100⟩ crystal. The solid line is the calculated relative energy-loss straggling.

To show common features in the straggling and stopping curves, we represent in Fig. 3 the measured data in the form of a relative energy-loss straggling, defined as $\Omega_0^2/\Delta E$, plotted as a function of the mean velocity of the ions in the foil. We observe here a straight-line behavior which is also reproduced by the theoretical calculations.

In summary, we present energy-loss straggling data for low energy protons in monocrystalline gold. A threshold behavior in the dependence on ion velocity is observed. Extending our previous energy-loss [9] model we provide a theoretical explanation for this behavior taking into account the band structure properties of solid Au [12] in the calculation of the energy transfer. The theoretical model explains this particular effect by considering the individual features of free (s,p) and nearly free (d) electrons, showing a threshold behavior that appears at very small energies. We obtain a very close agreement for both the stopping and straggling dependence with ion velocity by applying the same theoretical approach.

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