Velocity dependence of the energy loss of very slow proton and deuteron beams in Cu and Ag

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We present energy loss measurements of protons and deuterons down to velocities of 0.1 a.u. in very thin polycrystalline Cu and Ag foils, using the transmission technique. As in previous experiments in Au, a transition between two energy loss regimes is observed, i.e., from a stopping due to the excitation of only the free conduction band electrons at the lowest velocities, to a slowing down with the participation of both the free electrons and the loosely bound *d* electrons. This transition causes a change of the slope in the energy loss versus ion velocity curve at a characteristic threshold velocity related to the binding energy of the *d* electrons. As shown by model calculations these threshold velocities for Cu and Ag are shifted according to their density of states distributions.

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A basic process involved in the interaction of ion beams with matter is the energy loss due to the excitation of target electrons. This phenomenon is usually characterized by the electronic stopping power $\frac{dE}{dx}$ or the electronic stopping cross section $S=\frac{1}{N}\frac{dE}{dx}$, being N the atomic density of the target. The corresponding values are fairly well known for energies above 25 keV/amu, however at very low ion velocities the knowledge is incomplete due to a lack of accurate measurements. This scarcity is caused by some experimental difficulties, mainly problems in constructing smooth self supporting films with thicknesses below 30 nm required for the transmission technique, the most straightforward method for these experiments.

The standard energy loss theories [1–4] for low velocity light ions in metals, which consider the target electrons as a free-electron gas, predict a proportionality of the stopping power with the ion velocity. The existing data tabulations [5–7] generally confirm this prediction. However low-energy experiments with protons in Au, Ag, and Cu [8,9] showed significant departures from the predicted proportionality for velocities in the range $0.25 \lesssim v \lesssim 0.4$ a.u. At these low velocities the energy losses are lower than expected according to the density-functional theory (DFT) [4]. In Ref. [9] this departure was explained by including a minimum energy transfer for the excitation of the nearly free *d* electrons of the targets.

Recent experiments [10–12] at yet lower velocities with H^+ in Au polycrystals [10] and monocrystals [11] showed a transition from a velocity-proportional stopping power for $0.1 \lesssim v \lesssim 0.15$ a.u. to a steeper behavior for larger velocities. This result was explained in Ref. [11] extending the model of Ref. [9] by considering a regime at very low velocities with participation of only the free *sp*-electrons, and above a certain threshold velocity v_{th}, another regime that includes also the excitation of the nearly free *d* electrons [11]. A variation of this model has been also successfully applied to the energy loss of protons in the insulators LiF and AlF_3 [13].

The aim of this paper is the investigation of the very low and low velocity regimes for Group-11 metals, in particular Cu and Ag, with the purpose of obtaining further knowledge on the low-energy behavior and studying possible deviations from the velocity proportionality prediction [1–4].

The energy loss determinations were made by the straightforward transmission technique, complemented by an electrostatic ion energy analysis. The proton and deuteron beams were produced with a low-energy accelerator located at the Centro Atómico Bariloche, Argentina. The ions were generated on a hot-cathode ion source; accelerated; focused and filtered in mass and charge before entering on an hydrocarbon free vacuum collision chamber. The energy dispersion of the incident beams was of few eVs.

The polycrystalline Cu and Ag self-supported foils were made by evaporation over a very smooth plastic substrate that is subsequently dissolved [14]. The thicknesses Δx were 157 ± 8 Å for the copper target and 250 ± 12 Å for the silver one. These values were obtained by comparison with previous stopping power determinations [8,9]. The stated uncertainties arise from our energy loss determinations and do not include possible errors of the reference stopping power values. The thickness inhomogeneities of the targets were characterized by upper bounds for the roughness coefficients, being $\rho_{Cu} < 14\%$ and $\rho_{Ag} < 12\%$. These bounds were obtained assuming that all the energy loss straggling of the energy spectra is due to these inhomogeneities.

The targets were mounted perpendicular to the incoming beam direction, followed by a 127° cylindrical electrostatic energy analyzer in the forward direction with an angular acceptance of 0.5° in one direction and 1.6° in the perpendicular one.

For projectiles transmitted in the forward direction the contribution of the nuclear (or elastic) energy loss is reduced, and was estimated by computer simulations to be less than the experimental data dispersion. The energy loss data were taken as the difference between the incident and the most probable energy of the transmitted ions. In Fig. 1 we show as an example an energy spectrum of deuterons with incident energy $E_0 = 1.5$ keV/amu after emerging from the Cu foil.

We measured the energy loss of protons and deuterons in copper and silver in the very low-energy regime, between 0.2 and 9 keV/amu. In Figs. 2 and 3 we show the stopping cross section as a function of the mean velocity $\langle v \rangle = \frac{1}{2}(v_{in} + v_{out})$ of the ions together with the data of Valdés *et al.* [8,9]. The agreement between the velocity dependence of the present stopping power curves and the previous publications in the overlapping range is excellent.

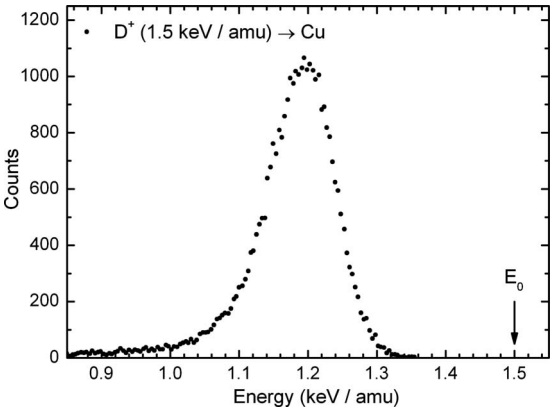

FIG. 1. Energy spectrum of 1.5 keV/amu deuterons transmitted through a 157 Å Cu foil.

For both targets the data of protons and deuterons are coincident within the experimental uncertainties, showing the absence of an isotopic effect on the stopping power of Cu and Ag for H^+ and D^+ in this energy range.

The experimental data plotted in Fig. 2 clearly show that at the lowest energies ($\langle v\rangle \lesssim 0.15$ a.u.) the stopping power of Cu is proportional to the projectile velocity. For higher energies the slope of the curve is increased and the proportionality relationship is broken. The stopping curve for the Ag target (Fig. 3) shows a similar behavior, with the low-energy regime holding up to velocities of about 0.2 a.u.

To describe the experimental data presented in this work we applied the theoretical model used in Ref. [11] for the stopping power of Au for protons and deuterons in the range of very low velocities. The model considers a stopping force dE/dx for slow ions with a friction coefficient Q that depends on the projectile velocity v

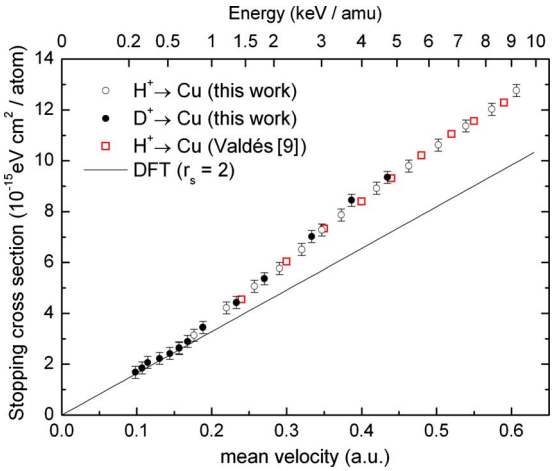

FIG. 2. (Color online) Stopping cross section for hydrogen ions in Cu, as a function of the mean projectile velocity (with the equivalent energy shown in the upper scale). Open circles show proton data, full circles correspond to deuterons. Open squares are previous data of Valdés *et al.* [9]. The high energy value of this data was used as reference stopping power in this work. Solid line corresponds to a DFT calculation for a free-electron gas with $r_s = 2$ a.u.

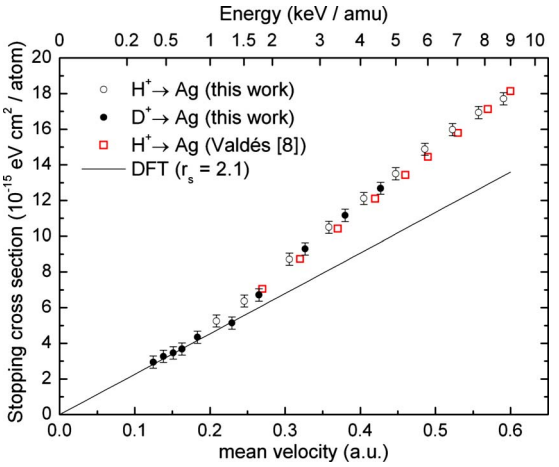

FIG. 3. (Color online) Stopping cross section for hydrogen ions in Ag, as a function of the mean projectile velocity (with the equivalent energy shown in the upper scale). Open circles show proton data, full circles correspond to deuterons. Open squares are previous data of Valdés *et al.* [8]. The high energy value of this data was used as reference stopping power in this work. Solid line corresponds to a DFT calculation for a free-electron gas with $r_s = 2.1$ a.u.

$$\frac{dE}{dx} = Q(v)v. \tag{1}$$

In this work $Q(v)$ is separated in two components for the electronic stopping: one represents the excitation of the conduction or free-electron band (of mixed s, p character) and the other is produced by the nearly free d electrons. This friction coefficient is calculated using the theoretical framework of the transport cross section, incorporating a minimum energy transfer U for the d electrons in order to take into account their binding energy. Using atomic units, its value is given by

$$Q(v) = n^{sp} v_F^{sp} \sigma_{tr}^{sp} + n^d v_F^d \sigma_{tr}^d, \tag{2}$$

where n^{sp} and n^d are the densities of free and d electrons, v_F^{sp} and v_F^d are the corresponding Fermi velocities, and σ_{tr}^{sp} and σ_{tr}^d are the respective momentum-transfer cross sections (MTCS). The MTCS's are calculated as

$$\sigma_{tr}(v,U) = 2\pi \int_{\theta_{\min}(U,v)}^{\pi} |f(\theta)|^2 (1-\cos\theta)\sin\theta d\theta, \tag{3}$$

$f(\theta)$ being the scattering amplitude and $\theta_{\min}(U,v)$ the minimum scattering angle given by [11]

$$\cos(\theta_{\min}) = 1 - \frac{U}{v v_r}. \tag{4}$$

In this equation v_r is the relative (electron-ion) velocity. Following [3], in this work the mean relative velocity was approximated by $v_r \cong (3/4)v_F$. For the free electrons $U_{sp}=0$ and their contribution to the stopping coefficient Q does not depend on v. On the other hand, for the nearly-free d electrons $U_d > 0$, so they contribute to Q only for velocities

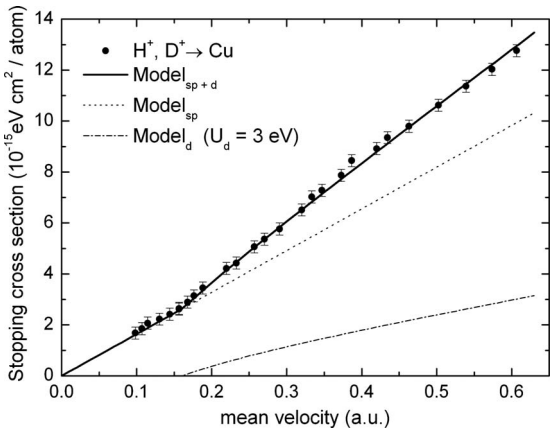

FIG. 4. Stopping cross section for protons and deuterons in copper. Full circles: present experimental results. Curves: solid line, theory total result; dotted line, free electron contribution; dash-dotted line, nearly-free-electron contribution. The minimum energy transfer of the d electrons in this calculation is U_d=3 eV.

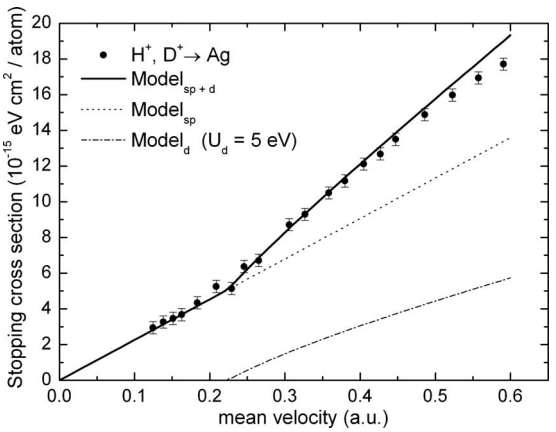

FIG. 5. Stopping cross section for protons and deuterons in silver. Full circles: present experimental results. Curves: solid line, theory total result; dotted line, free-electron contribution; dash-dotted line, nearly-free-electron contribution. The minimum energy transfer of the d electrons in this calculation is U_d=5 eV.

greater than a threshold velocity v_{th}, with a velocity dependence arising from Eqs. (2)–(4).

We calculated the scattering amplitudes for the sp and the d electrons by using the DFT phase shifts of Puska and Nieminen [15] for an electron gas characterized by the usual parameter r_s. The contribution of the sp electrons of each target was obtained by adjusting the r_s value fitting the low-energy slope of the stopping power data. These results are shown with solid lines in Figs. 2 and 3. The effective density of the d electrons was adjusted to obtain a total electronic stopping power of the theoretical model in agreement with the experimental results.

Using the DFT results of Ref. [15] we calculated the r_s value for the very low stopping power energy regime, obtaining r_s^{Cu}=2 and r_s^{Ag}=2.1 for Cu and Ag respectively. These values correspond to densities of roughly 2.4 and 3 free sp-electrons for each target atom. It may be noted that although the very low velocity behavior of the stopping power measurements in these metals is produced by the excitation of the free sp-electrons, also the nearly free electrons participate in the interactions. These combined effects are here represented by the effective r_s values indicated above.

The theoretical calculations are shown in Figs. 4 and 5 together with the experimental results. The most striking features of the measurements, i.e., the change of the slope of the stopping cross section curves at very low velocities and also the different position of the threshold velocity v_{th} for both targets are very well reproduced by the model. The stopping contribution of the free sp electrons is proportional to v as was previously stated. However, the d electrons only contribute to the stopping power of ions faster than a certain threshold velocity v_{th}. The agreement between the experimental data and the model is very good for both targets, with only some discrepancies at the highest energies for Ag.

Using a minimum energy transfer to d electrons of U_d^{Cu} =3 eV for the Cu calculations, the model gives a threshold velocity $v_{th}^{\mathrm{Cu}} \simeq 0.16$ a.u. In the case of silver, the corresponding values are U_d^{Ag}=5 eV and $v_{th}^{\mathrm{Ag}} \simeq 0.22$ a.u. It is worth to mention that the mean binding energies of d electrons from Cu and Ag, according to density of states calculations [16], are 3.0 and 4.9 eV, respectively, which agree very well with the above mentioned U_d^{Cu} and U_d^{Ag} values. So according to the present theory, the observed higher threshold velocity for Ag respect to Cu is a consequence of the deeper position of the silver d electron band [16].

An aspect that can also be observed in Figs. 4 and 5 is the slight bending of the energy loss vs v curve beyond $v=v_{th}$. This is also a consequence of the contribution of the d electrons, which has a nonlinear dependence on v due to the minimum excitation energy U_d.

One feature that is not predicted by the model and was adjusted to fit the experimental data is the value of the effective densities of both the sp and the nearly free d electrons. This aspect is important to characterize the relative contribution to the stopping cross section of each electronic component, and requires further development.

To summarize, measurements of the stopping power of Cu and Ag for very low-energy hydrogen ions are presented. No isotopic effects between H^+ and D^+ were found for both targets within the experimental uncertainties.

The energy dependence of the stopping power does not follow the predictions of standard theories, in a similar way to what has been observed for Au. The measured curves show a transition from a velocity proportionality with a reduced slope at very low energies, to a steeper dependence above a certain threshold velocity v_{th}.

The experimental results are well reproduced by the model of Ref. [11] which considers a transition from a stopping regime due to the excitation of only the conduction electrons to a more complex behavior at higher energies where the contribution of the nearly free d electrons excitation is also present.

The values of the minimum energy transfer to d electrons U_d used in the theoretical model, for both Cu and Ag, are in good agreement with the mean binding energies of these electrons given by density of states calculations. The corresponding values of v_{th} (larger for Ag than for Cu) are therefore explained by those binding energy values.

Considering also the previous results for Au [11], this work extends the applicability of the theoretical model for the low velocity behavior of the stopping power of the group 11 metals: Cu, Ag, and Au.

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