Energy loss of protons in Zn: Measurements between 2 and 200 keV

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The energy loss of H^+ ions in polycrystalline Zn films has been determined by the transmission method in the energy range between 2 and 200 keV. Experiments in this energy range are of special interest for the study of the energy-loss mechanism in the region of the stopping-power maximum where important phase effects have been recently determined by other authors, as well as in the low-energy range where we have previously found deviations from the predicted velocity proportionality for other metallic elements. The data obtained here allow an experimental evaluation of the phase effect and provide a test of the velocity dependence for low energies at a very good level of accuracy. In addition, the experiment contributes to filling an important gap in the data due to the lack of low-energy measurements for Zn targets.

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I. INTRODUCTION

Data on energy loss for ions in solid Zn are especially scarce because of experimental difficulties in preparing good quality targets. This is due in part to the condensation properties of the vapors of this metal. Additionally, there exist general difficulties, such as beam dispersion and detector responses, arising at energies below $\sim$ 10 keV. Thus newer stopping-power tabulations of various elements for protons [1, 2] do not include Zn data at all. Other tabulations or semiempirical fits at low velocities only contain indicative values arising from generalization formulas [3, 4] and show appreciable differences between them. On the other hand, interesting experimental and theoretical comparisons of proton energy losses in solid and vapor phases of Zn have been recently published [5, 6] for proton energies around and above the stopping-power maximum. In these references, important differences (phase effect) of up to 50–60 % between solid and vapor Zn targets have been found.

Another question of considerable interest in the light of current research is the study of the velocity dependence of the low-energy stopping power in metallic targets. As is well known, the dominant mechanism for the energy loss of protons below the stopping-power maximum is the energy transfer to the electrons in the outer shells of target atoms. In the case of metallic targets this includes the electrons in the conduction band and possibly other close-lying electronic bands. Based on free-electron gas properties, and even within the scope of nonlinear theories, the theoretical models predict a velocity proportionality for the low-energy stopping power of metallic elements [7–9].

We have found [10, 11] that this dependence is experimentally confirmed for some elements (Al, Sb, and Bi), whereas other group of elements (Cu, Ag, and Au) show deviations from this dependence. The theoretical interpretation of these differences [11] is based on studies of the electronic structure of each element or group of elements. Thus, in the case of the transition metals, the deviations from the velocity proportionality (expected to apply only for free-electron metals) can be explained by the existence of d electrons whose energy bands are shifted by only a few eV with respect to the Fermi energy; this introduces a minimum energy transfer necessary for an excitation process (threshold effect).

Since the vicinity of the $4s$ and $3d$ shells in Cu, the $5s$ and $4d$ shells in Ag, and the $6s$ and $5d$ shells in Au are crucial for the quantitative estimation of the threshold effect, the properties of Zn, with a larger energy shift (of about 8 eV) between the $4s$ and $3d$ shells, point to this element as a good candidate to test the characteristics of the velocity dependence.

In this work we report measurements of energy losses of proton beams on an extended energy range (from 2 keV up to 200 keV) after transmission in the beam direction through very thin foils of Zn. The significance of the results is discussed in relation to the two main problems mentioned before, namely, the phase effect in the energy loss of protons in Zn and the velocity dependence in the lower-energy range. The results obtained here are compared with experimental and theoretical results by other authors.

II. EXPERIMENTS

The extended energy range from 2 to 200 keV was obtained by measurements performed with two ion accelerators of our laboratory: a low-energy accelerator operated in the range from 2 to 10 keV and a medium-energy accelerator operated from 10 to 200 keV. The measuring equipment has been described elsewhere [12, 10]. A similar experimental setup for the transmission method was employed in both accelerators. The energy analysis

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was performed by electrostatic analyzers collecting the ions emerging from the target in the forward direction. The energy resolution was 2% at low energies and 0.5% at energies above 10 keV. The angular acceptance was $\pm 0.34°$ at low energies and $\pm$ 0.05° at energies above 50 keV. After this energy and angular selection, the protons were detected with open electron multipliers, followed by pulse electronics and a multiscaler analyzer.

Data were obtained using foils from the same evaporation batch in both accelerators. The consistency of the measurements made with the different machines was checked by comparison of the values at 10 keV, accessible to both. Each data point is the mean value over two or more foils of the same batch.

The self-supported targets were made by evaporation under clean vacuum conditions on a very smooth plastic substrate [13]. To overcome the condensation difficulties, a very small amount ($\sim$ 2%) of Ag was previously deposited on the substrate [14]. The effect of this deposit on the measured energy loss values is well below the statistical uncertainties, as it was estimated using tabulated values [3] and previous measurements made at this laboratory [10].

The foil thicknesses were determined by fitting the energy-loss measurements at 200 keV to the stopping cross section given by the Andersen-Ziegler tables [3], which are based on experimental data at this energy and above. The resulting thickness values were between 194 Å and 205 Å. The foil roughness was evaluated by a beam technique [15] (which gives an upper limit to it) being at most 17% of the mean foil thickness. Foil thickening by beam bombardment was held within negligible limits by using a very low ion current density of $\sim 10^{-9}$ A/cm^2 and irradiation times of the order of 2 min per spectrum. We noticed no changes in the energy loss during the measurements, even repeating them after many months. The statistical errors of the present measurements are of the order of $\pm$2%.

III. RESULTS

Figure 1 shows the energy-loss measurements together with the experimental and theoretical stopping cross sections of Ref. [6], he experimental values of Ref. [16], and the semiempirical curve of Ref. [3]. The way the experimental energy losses can be compared with the theoretical calculations at low velocities (where the energy loss in the foil amounts to a finite fraction of the initial energy) is described in Ref. [10] and will be briefly reviewed below. The result is that one should represent the energy-loss values vs the average $\langle v \rangle$ of the velocities at the entrance and the exit of the foils (or the equivalent energy). This average procedure is specially significant at the lowest energies of this experiment. Thus the lowest point in Fig. 1 is located at 1.61 keV instead of 2 keV (the incident energy). At higher energies, where the energy lost in the foil is a small fraction of the incident energy, these differences become negligible.

The data from lower (2–10 keV) and higher (10–200 keV) energies show an excellent matching, in absolute values and slopes, at 10 keV. Both sets of measurements

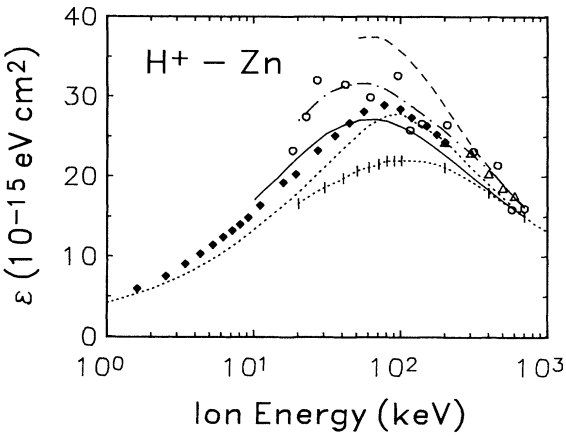

FIG. 1. Energy-loss values (in units of stopping cross section) of Zn for protons vs the energy corresponding to the average velocity of the projectiles in the foil. Experimental values for solid Zn: diamonds, this work (zero-angle transmission method); dotted line with vertical bars, stopping cross sections (RBS method) from Ref. [6]; triangles, data from Ref. [16]. Dotted line, semiempirical values from the Andersen and Ziegler tabulation [3]; solid line, calculations for solid targets from Ref. [6]; circles, experimental values for gases from Ref. [6]; dot-dashed line, a fitting curve for these values; dashed line, calculations for gases from Ref. [6].

are independent, but a common calibration procedure (by energy-loss measurements at 200 keV) was applied to all the foils. Therefore, the nearly exact coincidence of the data at 10 keV gives a proof of the consistency of both experiments.

From Fig. 1 we can note the following features. Our data show a more pronounced maximum than the experimental values for solids of Ref. [6] and the projectile energy E_{max} at which this maximum occurs is lower in our case. On the other hand, we observe a good matching with the calculations for solid Zn contained in Ref. [6].

The different shapes of the stopping curves and the displacement of E_{max} can be understood by taking into account the differences between both experimental situations. Whereas in our case the energy loss is measured by the transmission method, with a small angular acceptance in the forward direction (like in the vapor experiments of Refs. [5, 6]), the measurements for solid Zn in these references have been made using the backscattering technique. It should be noted that both (transmission and backscattering) types of experiments sample in different ways the interactions occurring inside the medium. In the transmission experiment a selection of projectile-target interactions with larger impact parameters is made, as compared with the backscattering case [17].

As is known from previous studies [12, 18, 19], there is a relation between the angular and the energy-loss distributions; the energy loss in the forward direction corresponds, on the average, to interactions with slower (external) electrons. Therefore, with increasing observation angles, a shift of E_{max} to higher energies is observed

[20]. So, a more comprehensive comparison of the results obtained with both methods should include a consideration of the combined angular–energy-loss distributions, a question that lies beyond the scope of the present work.

Our measurements yield values about 30% higher than those of Refs. [5, 6] in the region of the maximum. It may be noticed that this comparison depends on the criterion used for the foil thickness calibration in both experiments. Our data have been normalized at 200 keV according to the Andersen-Ziegler tables [3], whereas the values of Refs. [5, 6] for solid Zn have been normalized at 500 keV with respect to the tabulations by Paul, Semrad, and Seilinger [1] through measurements of the RBS yield ratios for H^+ in Zn and Cu.

We observe from Fig. 1 good agreement between the general behavior of our data and the semiempirical predictions of Andersen and Ziegler [3] through a wide range of energies. Good agreement is also observed with the experimental results from Bader *et al.* [16], with respect to the slope or energy dependence in the high-energy side.

When comparing our solid Zn data with the vapor data of Refs. [5, 6], the phase effect in the energy loss becomes smaller than the values reported earlier; the maximum effect that we obtain would be about 30%. Again we should note here that all these values depend on the absolute calibration of the energy-loss data. The Zn vapor data used in Refs. [5, 6] were normalized to their theoretical value at 700 keV. Irrespective of this, it should be remarked that the comparison of our data for solid Zn with those of Ref. [5, 6] for vapors of this element seems to be adequate, since both measurements have been made using the same (transmission) method.

In order to test the velocity dependence of the energy loss in the low-energy range, we shall briefly recall the way in which the data may be represented when the energy loss inside the medium is not a differential quantity [10]. As discussed earlier, if a simple velocity-proportional dependence for the differential energy-loss ratio is assumed

$$\frac{dE}{dx} = -kv \ , \tag{1}$$

then one can easily integrate the energy loss $\Delta E = E_0 - E_1$ in a foil of finite thickness Δx (here E_0 and E_1 denote the beam energies before and after being transmitted), so that one gets nearly the same expression for the ratio $\Delta E/\Delta x$, namely,

$$\frac{\Delta E}{\Delta x} = k\langle v\rangle \ , \tag{2}$$

in terms of an average velocity $\langle v\rangle$, defined by

$$\langle v\rangle = \frac{1}{2}(E_0^{\frac{1}{2}} + E_1^{\frac{1}{2}})(2/m_p)^{\frac{1}{2}} \ . \tag{3}$$

In Fig. 2, the data of this experiment are displayed vs the average velocity $\langle v\rangle$, together with the calculations of Nagy, Arnau, and Echenique [9] and those corresponding to solid Zn in Ref. [6]. These theoretical predictions are based on different models that apply either at very low velocities [9] or in a more extended range that includes energies around the maximum [6]. Both calculations show very good agreement with the present measurements.

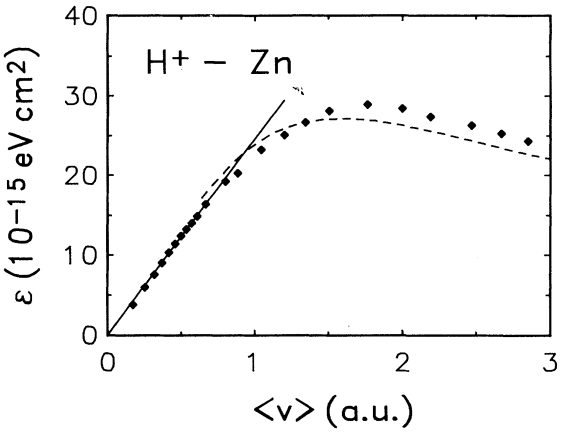

FIG. 2. Energy-loss values (in units of stopping cross sections) of solid Zn for protons vs the average velocity $\langle v\rangle$ in the foil. Diamonds, this experiment (zero-angle transmission method); solid line, calculations from Ref. [9]; dashed line, calculations for solid targets from Ref. [6].

The agreement is especially remarkable in the low-energy range, with the predictions of the density-functional theory [9] (which predicts a proportionality with ion velocity). The slight deviation from the velocity proportionality of the data at lowest velocities is very close to the statistical uncertainty, so that at this point we cannot attribute any special significance to this small effect. The density-functional calculations correspond to the effective r_s value of Zn ($r_s^{\text{eff}} = 1.97$) taken from Ref. [21].

The velocity dependence is similar to the one obtained earlier for Al, Sb, and Bi [10]. We can mention at this point that deviations with respect to the velocity proportionality were found earlier [10, 11] in the case of noble metals (Cu, Ag, and Au). These deviations can be explained due to the existence of d electron bands with energies shifted by few eV with respect to the Fermi energy of these metals. We refer to them as "nearly free" electrons. Therefore, for energies close to the stopping-power maximum the effective number N_{eff} of electrons that can be excited may be considerably larger than the number N_f of free electrons (those that contribute to the metallic conductivity). Thus, for instance, one finds N_{eff}:N_f values of 3.14:1.0 for Cu, 7.76:1.0 for Ag, and 8.21:1.0 for Au [21]. Then, when the ion velocity decreases, the contribution of the nearly free d electrons diminishes (threshold effect) and the velocity dependence shows a decline with respect to free-electron-like metals [11]. (Note also that these deviations can be experimentally distinguished only for rather large values of N_{eff}:N_f).

The N_{eff}:N_f value for Zn from Ref. [21] is 3.2:2.0,

i.e., closer to the ratios for Al (2.83:3.0), Sb (5.57:5.0), and Bi (5.57:5.0) than those for the other group of elements. The velocity dependence for Al, Sb, and Bi in the low-energy range was found to be consistent—within the limits of the experimental resolution—with a simple velocity-proportional behavior. Therefore, the low-velocity dependence in the case of Zn is consistent with the results obtained for these elements.

IV. CONCLUSIONS

We present energy-loss measurements for protons in Zn using the transmission method in the incident direction for beam energies between 2 and 200 keV. These measurements are in the low-energy range and extend to energies over the stopping-power maximum, with very low statistical fluctuations. The transmission method used here permits a straight comparison with experimental values taken in similar conditions for Zn atoms in the vapor phase. In this way, the existence of a phase effect previously demonstrated in Refs. [5] and [6] is confirmed, although the magnitude of the effect obtained here is smaller than the one previously reported.

We analyze the velocity dependence of the stopping cross section at low energies and find, within the experimental uncertainties, no significant deviations from the predicted velocity-proportional dependence. This is consistent with an interpretation based on the electronic structure of Zn and with previous results for other elements.

Finally, the present results provide a conclusive test to previous calculations for solid Zn made by Nagy, Arnau, and Echenique [9] and by Arnau *et al.* [6], in different energy ranges. Both calculations agree very well with the present experimental results.

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