Phase Effect in the Energy Loss of H Projectiles in Zn Targets: Experimental Evidence and Theoretical Explanation

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The energy loss of H projectiles in solid and gaseous Zn has been measured for the first time down to 20 keV/u, showing large differences in the stopping cross section depending on the state of aggregation of the target. For 25-keV protons, the stopping cross section of Zn in the gas phase is found to be 60% higher than that in the solid phase. A charge-state approach to the stopping power of ions in the solid and in the gas is successful in explaining this effect.

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The electronic stopping power of matter for swift protons with energies below, and of the order of, 100 keV is dominated by outer-shell electron excitation and ionization. The stopping power has its maximum in this energy region for almost every target. As a result of the different electronic structure of the outer shells of a material in different states of aggregation, especially in the case of metals, one would expect large differences between solid and gas phase stopping for the same substance. Up to now, the few reported measurements either deal with materials for which the electronic structure does not change appreciably between the two phases [1,2], or are just measurements of the ratios of the He to H stopping cross sections [3]. These data do not permit the testing of theoretical predictions since they are restricted to simple elements [4–6]. In the low-energy range (below 100 keV for protons) accurate first-principles calculations are only possible for light elements like H and He targets [7,8].

From the experimental side, it is not easy to find an element for which the two phases show a marked difference in the valence structure in a temperature range where the experiment is feasible. Compounds would not be suitable because they inevitably add other aggregation effects due to the chemical bonds. We have chosen Zn as a target as it has a relatively low boiling point and is easy to handle in the solid phase.

The experiment was performed at the University of Linz using the 700-keV Van de Graaff accelerator. The energy loss in the gas phase was measured by transmission of the ion beam through a vapor cell in a way similar to our earlier measurements on water vapor [2]. Protons and deuterons in the energy range $E_{\min}=15$ keV/u to $E_{\max}=720$ keV/u were used as projectiles. After transmission through the gas cell, some of the ions impinged on a scattering target (a thin platinum layer evaporated onto carbon). The energy of the scattered ions was detected at an angle of 90° with respect to the beam axis by means of a particle-implanted silicon detector. The detector and the whole amplification system were connected to a thermostat in order to minimize thermal drifts. The vapor cell had a length of 30 cm with small apertures of 1.5 mm diameter for the ions to enter and leave. In the cell, the zinc vapor was in thermal equilibrium with the condensed phase (usually the liquid). Because of the low dissociation energy of Zn clusters the vapor consisted essentially of Zn atoms. The temperature of the gas cell was chosen and kept constant via heating, which was applied at the outer surface of the cell. A homogeneous temperature profile was obtained. The temperature was controlled by a thermocouple to keep the vapor pressure constant. Reproducibility was possible within $\pm 5\%$. Hence we measured the energy losses, $\Delta E(E,n)$, at a constant ion energy E for a set of vapor densities n in the range 8×10^{14} to 6×10^{15} atoms/cm^3. The slope of the linear regression of $\Delta E(E,n)$ vs $\Delta E(E_{\max},n)$ at constant energy E yielded the ratio of the stopping cross sections $S(E)/S(E_{\max})$. By fixing $S(E_{\max})$ equal to the theoretically calculated value at 700 keV (see below) we obtain the stopping cross section at all energies.

In order to measure the charge-state fractions of protons and neutral hydrogen atoms, ϕ^+ and ϕ^0, in the ion beam after exiting the vapor cell, a magnetic deflection field was applied, preventing the charged projectiles from hitting the scattering target. The scattered intensities I_0 and I_{tot} with and without applied magnetic field, respectively, were measured during equal times (100 s) under stable current conditions. This yielded $\phi^0=I_0/I_{\text{tot}}$, and $\phi^+=1-\phi^0$ (ϕ^- was found to be negligible). The charge-state fractions obtained follow a common curve with uncertainties of at most 2%.

The stopping cross section (SCS) for the solid phase was determined by the well established Rutherford backscattering (RBS) technique [9,10]: Zinc was evaporated onto a carbon backing following Ref. [11]; purity, stabili-

ty under ion bombardment, and thickness homogeneity were checked by RBS. The stopping cross section was obtained from the RBS spectra measured using protons and deuterons in the energy range 20 to 720 keV/u [10]. The areal mass density of the zinc layer was measured relative to that of a copper target by applying charge collection and making use of the precisely known stopping cross section of copper for 500-keV/u hydrogen ions [12]. The uncertainty of the thickness determination amounts to $\pm 5\%$ while the statistical uncertainty of the stopping cross-section measurements is at most $\pm 2\%$.

In Fig. 1 we show our experimental results together with a calculation based on a charge-state approach to the stopping power. As one might expect the phase effect increases when going down in projectile energy; it is 60% below 30 keV/u.

A very brief outline of the main ingredients of the calculation is explained below. A complete description of our work together with comparison, when appropriate, with available calculations for each charge state, and a critical evaluation of the approximations invoked will be presented elsewhere [13].

We use a charge-state approach [14] to the stopping power of ions in matter to calculate the energy loss of H projectiles in the solid and gas phases of Zn. This is a crucial step due to the fact that in this energy range the hydrogen projectiles appear in different charge states depending mainly on ion speed. To explain the stopping process one has to take into account the energy loss due to projectile and target excitations, including charge-changing events (capture and loss of electrons).

The SCS of atomic Zn for H projectiles has been calculated in the energy range 50 to 700 keV/u. The total SCS is a weighted sum of partial SCS of each of the charge states in the hydrogen beam. The weighting factors are the experimentally measured charge-state fractions. The target atom is described in the Hartree-Fock-Slater (HFS) approximation, i.e., each electron in the initial and final state is considered to move in the HFS potential [15,16] of neutral Zn. Excitation and ionization cross sections are calculated in the first Born approximation (FBA) [17]. In the energy range considered here, the largest contributions come from the outer shells ($4s$ and $3d$); for those we expect the FBA to be fairly accurate above 50 keV. Comparison with the calculations by McGuire [18] of the subshell stopping power [19] of Zn for bare protons shows that above 50 keV the $4s$ and $3d$ electron contribution that we obtain is always within 10% of his.

We have evaluated approximately the contribution from capture and loss processes to the energy loss using the experimentally measured charge-state fractions and calculated capture cross sections in the continuum distorted wave (CDW) approximation [20].

The stopping power of solid Zn for hydrogen projectiles in the energy range 10 to 700 keV/u is calculated evaluating the equilibrium charge-state distributions, the energy loss in charge-changing events, and the partial stopping powers for each of the charge states as described in Refs. [21] and [22] with the proper parameters needed to describe the solid Zn target instead of Al. These include a different valence electron density and the inclusion of $3d$ electrons in the description [23].

In Fig. 2 we plot the contributions of the outer shells of the target to the energy loss of protons for both the gas and the solid phase of Zn to show the contribution of the target excitation to the phase effect. It is clear from the figure that the different excitation spectrum of the $4s$ electrons in the gas and solid phases is the main source of the effect.

In Fig. 3 we plot the contributions from the different charge states and capture and loss processes to the energy

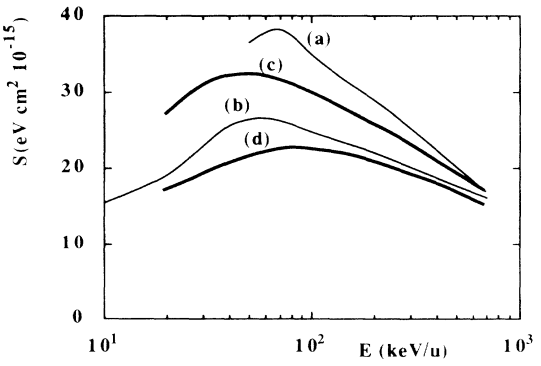

FIG. 1. Stopping cross section (in 10^{-15} eV cm^2) of Zn for H projectiles as a function of the projectile energy (in keV/u). The thin lines are the result of our calculation for the gas (curve a) and the solid phase (curve b). The thick lines are the best fit to the measured data for the gas (curve c) and the solid phase (curve d).

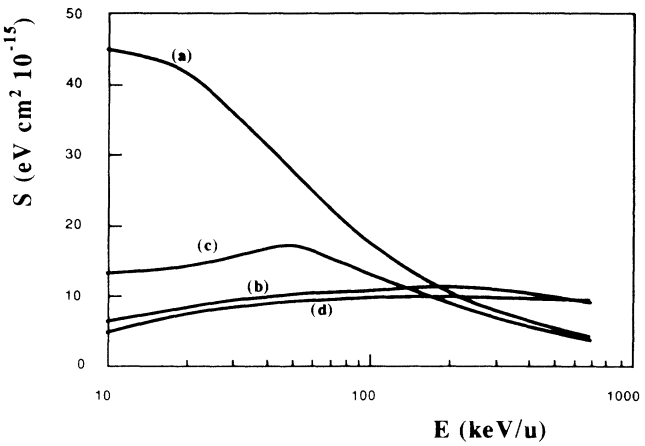

FIG. 2. Outer-shell electron contributions to the stopping cross section of Zn for bare protons. Curves a and b are the $4s$ and $3d$ contributions as obtained in our model for the gas-phase stopping cross section, while curves c and d are the $4s$ and $3d$ electron contributions that we obtain for the solid-phase stopping cross section.

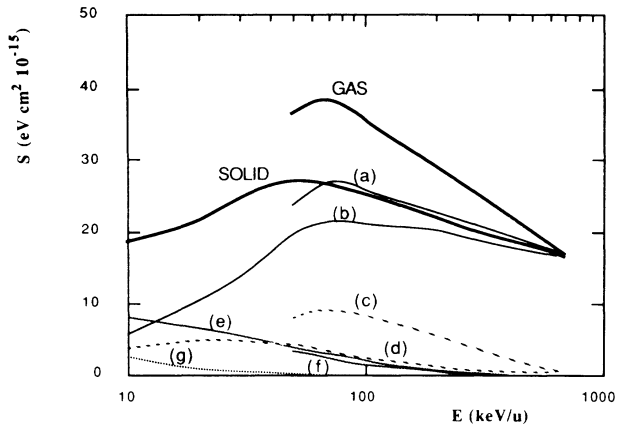

FIG. 3. Comparison between the theoretically calculated gas and solid phase stopping cross sections of Zn targets for H projectiles as a function of the projectile energy. The contributions to the total stopping from the different charge states and charge-exchange processes are shown separately. Curves *a* and *b* are the bare proton contribution to the energy loss in the gas and solid phases, respectively, while curves *c* and *d* are the contributions from capture and loss processes. The contributions from neutral hydrogen atoms are shown in curves *e* and *f* for the gas and solid phases, respectively, and curve *g* corresponds to the contribution from negative ions in the solid phase.

loss of hydrogen projectiles in Zn targets to show the charge state and projectile excitation effects. Except at very low energies, the main difference between the gas and solid phases is the contribution from capture and loss processes. This contribution is higher in the gas phase than in the solid phase. It is due to the fact that for light ions loss cross sections are higher for gases as compared to solids, since no final-state restriction exists [24] and transition energies are also higher in projectile ionization (no level shift due to screening in the gas phase).

In conclusion, we have reported measurements on the phase effect of Zn stopping for H projectiles at energies down to the stopping power maximum showing a 60% higher stopping cross section in the gas phase at 25 keV. New experiments with other targets and projectiles would give more insight into the phase effect problem. Alkali metals as targets and antiprotons as projectiles are expected to enhance this effect. Theoretical calculations based on a charge-state approach to the stopping power of ions in matter show that this difference between the two phases is due to both target (4*s* electrons) and projectile (capture and loss) excitations.

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