Experimental and theoretical study of the energy loss of C and O in Zn

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We present a combined experimental-theoretical study of the energy loss of C and O ions in Zn in the energy range 50–1000 keV/amu. This contribution has a double purpose, experimental and theoretical. On the experimental side, we present stopping power measurements that fill a gap in the literature for these projectile-target combinations and cover an extended energy range, including the stopping maximum. On the theoretical side, we make a quantitative test on the applicability of various theoretical approaches to calculate the energy loss of heavy swift ions in solids. The description is performed using different models for valence and inner-shell electrons: a nonperturbative scattering calculation based on the transport cross section formalism to describe the Zn valence electron contribution, and two different models for the inner-shell contribution: the shellwise local plasma approximation (SLPA) and the convolution approximation for swift particles (CasP). The experimental results indicate that C is the limit for the applicability of the SLPA approach, which previously was successfully applied to projectiles from H to B. We find that this model clearly overestimates the stopping data for O ions. The origin of these discrepancies is related to the perturbative approximation involved in the SLPA. This shortcoming has been solved by using the nonperturbative CasP results to describe the inner-shell contribution, which yields a very good agreement with the experiments for both C and O ions.

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The study of the energy loss of ions in solids is a problem of interest for basic and applied research in many areas, such as ion implantation, radiation damage, and space research. Although a large number of experiments and calculations have been produced over the years, there is a demand for new data for various projectile-target combinations. Additionally, the development of a consistent theoretical framework is a subject of great current interest [1,2]. In recent years, we have performed a set of studies that combine experimental and theoretical research with the aim of providing a theoretical framework that could serve as a basis for more accurate predictions of the energy loss of swift ions in various solid materials. It is worthwhile to mention that for ions heavier than Li, the experimental stopping power data are scarce except for some particular materials (C, Al, Si, Ag, Au) [3]. In zinc, no energy loss measurements for C and O ions have been reported in the literature.

In previous papers, we have studied in a systematic way the stopping coefficients for a series of ions of increasing atomic numbers: H, He, Li, Be, and B in zinc [4–8], and with our theoretical formulation, we have been able to explain changes in the stopping power curves, reaching a very good agreement between the experimental and theoretical results. This agreement was achieved by a detailed theoretical study of the contribution of each individual charge state of the projectile and each electronic shell of the target. This was made by separately calculating the contribution of valence electrons and inner target shells through a combination of two theoretical models: a nonperturbative scattering model using the transport-cross-section–extended-Friedel-sum rule (TCS-EFSR) approach [9], and a shellwise local plasma approximation (SLPA) [10].

In the present work, we study experimentally and theoretically the energy loss of C and O in Zn targets with the aim of exploring how far we can extend the application of the previous theoretical schemes. The choice of these ions is particularly appropriate, since the maximum energy we can measure (1 MeV/amu, or an impact velocity of 6.32 a.u.) means that we deal with the nonperturbative regime ($Z/v \gtrsim 1$).

In this contribution, we show that for projectiles heavier than C, the theoretical approach based on the SLPA calculations is no longer adequate due to the growing influence of higher-order or nonlinear effects on the whole range of energies. We show also that the combination of a nonperturbative description, such as the convolution approximation for swift particles (CasP) model [11], with the TCS-EFSR for the valence electrons leads to a very good agreement between experiment and theory.

The energy loss determinations were done by the Rutherford backscattering technique (RBS) at the 3 MV Tandetron of the Instituto de Fisica da Universidade Federal de Rio Grande do Sul, Brazil. In the present experiments, the energy range between 50 keV/amu and 1.2 MeV/amu was studied.

The targets were made of a multilayered Au-Zn-Au film deposited on Si wafers, as in previous works [7,8]. Several target thicknesses were used according to the different energies and projectiles used in the present experiment. The employed Zn films were of 81, 244, and 350 nm, with a typical uncertainty of 3%. The last values were determined by energy loss measurements using proton beams. The stopping power values used to obtain the thicknesses were taken from Refs. [12,13].

The experimental procedure was similar to that described in Ref. [8]. In this case, the detector angle was of 120° with respect to the beam direction. The data analysis was performed using the mean energy approximation with the energy-loss ratio method [14]. This experimental method requires two measurements at different sample angles in order to obtain the stopping power at the inward and outward path. In order

to improve the precision, for each energy we measured four RBS spectra at different sample angles. The overall resolution of the detector plus electronics is of the order of 25 keV.

The theoretical approaches used to calculate the energy loss of ions in solids have been described in detail in previous publications [6–8].

In the equilibrium regime, the mean energy loss will be the weighted average of the stopping cross sections $S_q(v)$ corresponding to each ion charge state q, namely,

$$S(v) = \sum_{q=0}^{Z_P} \phi_q(v)\, S_q(v). \tag{1}$$

The stopping cross sections $S_q(v)$ are calculated for each individual case, considering an ion dressed with $N = Z_p - q$ bound electrons frozen in its ground state.

Each partial stopping cross section $S_q(v)$ is calculated as

$$S_q(v) = S_{q,\text{valence}}(v) + \sum_{n\ell} S_{q,n\ell}(v), \tag{2}$$

where $S_{q,\text{valence}}(v)$ and $S_{q,nl}(v)$ are the contributions of valence and inner-shell excitations, respectively. The screening of the nuclear charge by the N bound electrons was considered in the form of an effective charge function that depends on the momentum transferred. To this end, the atomic structure of the incident ion was represented by the Hartree-Fock wave functions (for the neutral or the different positive ions) tabulated by Clementti and Roetti [15]. The effective charge function has an analytical expression in terms of solutions of the Flannery-Levy integrals [16] as shown in Ref. [17].

The stopping power due to valence electrons was calculated considering them as a free electron gas (FEG) using the TCS-EFSR approach [9], as described in previous papers [6–8]. The TCS-EFSR is a *nonlinear* or nonperturbative model, since it contains no linearizing or perturbative assumptions, and, therefore, it applies to all orders in the interaction. This scheme has been found to be very important in previous studies to get an appropriate description of the energy loss for the low range of ion velocities, below the stopping power maximum. It converges to the value obtained with Lindhard dielectric formalism for energies around 1 MeV.

The contribution of inner shells was calculated by two different methods: the SLPA [10] and the CasP [11]. The SLPA is a general method to calculate the contribution of the ionization of target bound electrons to the different moments of the energy loss [10,18–21] within the dielectric formalism. The dielectric function employed is the Levine-Louie one [22], which includes the binding energy of each shell within the known Lindhard dielectric function, keeping the validity of the f-sum rules. The Lindhard dielectric function [23] includes screening among electrons and possible collective effects. It is a linear response approach (first order in the projectile charge), and so it works within the perturbative limit, i.e., for $Z/v \ll 1$.

In Figs. 1 and 2, we show the experimental results for the energy loss of C ions in Zn, together with theoretical calculations. In Fig. 1, we display the theoretical curves corresponding to the partial contributions to the stopping cross sections: line v shows the contribution of the valence-band electrons according to the TCS-EFSR model calculated using the HISTOP program as described in Ref. [9]. The curve

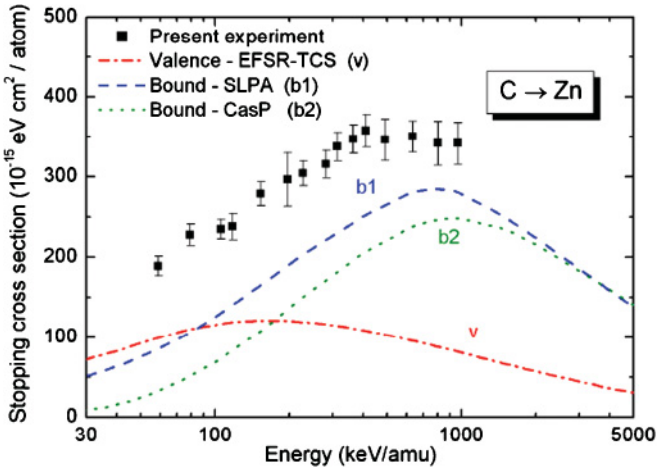

FIG. 1. (Color online) Stopping cross sections of C in Zn. Symbols: present experimental total stopping data. The curves show the separate contributions of the valence and the bound electrons. Dashed-dotted line: TCS-EFSR results for valence electrons. Dashed line: SLPA results for bound electrons. Dotted line: CasP results for bound electrons.

denoted b_1 is the contribution of the bound electrons of Zn (Ne-$3s^2 3p^6 3d^9$) calculated with the SLPA. In this figure, we also include the CasP results (curve b_2) only for the bound electrons [11]. The unitary convolution approximation (UCA) model was used to calculate the stopping cross section for each charge state of the ions. They were averaged considering the charge state fractions at each energy.

In Fig. 2 we show the experimental values and the theoretical results for the total stopping cross section obtained with the SLPA for the bound electron contribution (curve b_1 of Fig. 1) and with the CasP values for bound electrons (curve b_2 of Fig. 1). In both cases the contribution of the valence electrons calculated with the HISTOP code has been added (curve v of Fig. 1). Both theoretical curves describe the

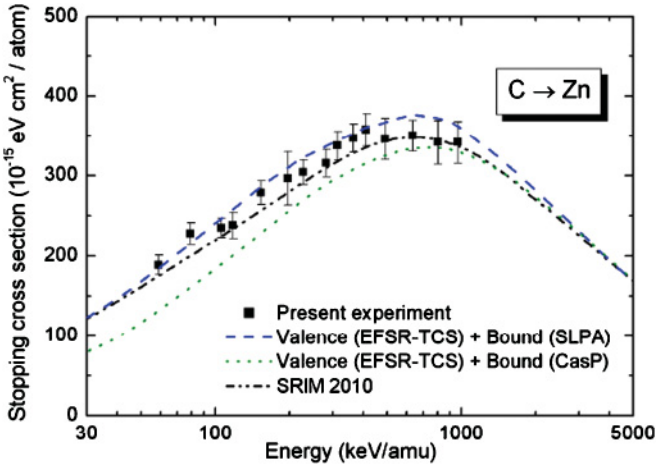

FIG. 2. (Color online) Total stopping cross sections of C in Zn. Symbols: present experimental total stopping data. Dashed line: addition of the SLPA for bound electrons and TCS-EFSR for valence electrons. Dotted line: addition of CasP values for bound electrons and TCS-EFSR for valence electrons. Dashed-dotted-dotted line: total stopping values of the SRIM package.

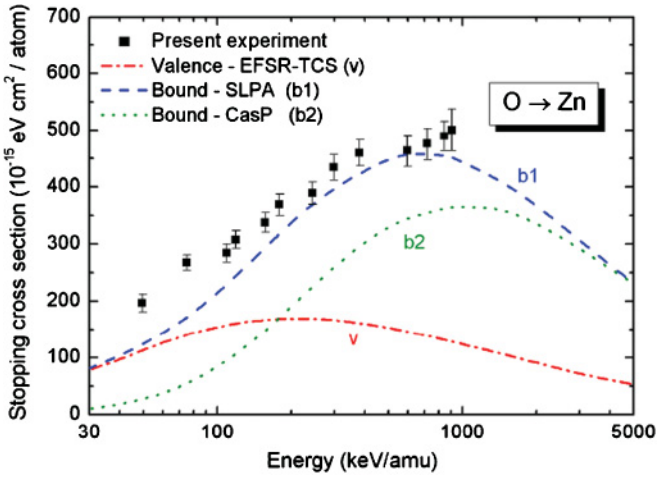

FIG. 3. (Color online) Stopping cross sections of O in Zn; separate contributions of the valence and the bound electrons. Curves and symbols as in Fig. 1.

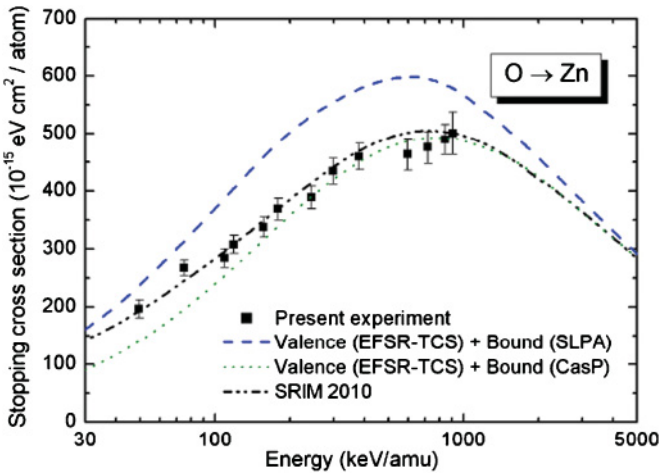

FIG. 4. (Color online) Total stopping cross sections of O in Zn. Curves and symbols as in Fig. 2

maximum of the stopping power around 600 keV/amu rather well and converge to similar values for high energies. The SLPA slightly overestimates the data within the experimental uncertainties, being always over the data. Additionally, we display the semiempirical stopping power values included in the SRIM 2010 package [24]. The SRIM values are in very good agreement with our data for the stopping power of these ions in Zn, which is basically a consequence of the semiempirical character of the SRIM package. It should be remarked that for other ion-target combinations, there are discrepancies between experimental data and SRIM values, in particular, for ions heavier that Li. (see Ref. [2]).

In Figs. 3 and 4, we show the experimental results for the energy loss of O ions in Zn, together with the theoretical calculations. The meaning of the different curves is the same as in the previous figures. In this case, we observe that the values of total stopping cross section obtained by the SLPA model for bound electrons are much higher than the experiment (20% above at the stopping maximum). This indicates the Z_1 limit of applicability of the perturbative SLPA approach for Zn targets. It is well known that the linear methods tend to overestimate the magnitude of the energy transfer since they do not contemplate possible saturation effects, a problem that becomes more relevant with increasing values of the projectile atomic number [25]. On the other hand, the combination of two nonperturbative models (TCS-EFSR for the valence electrons and CasP for bound electrons) gives a good description of the stopping power of O ions in Zn, as is shown here.

These new results and comparisons are of interest in order to test the validity range of the SLPA, which was successfully employed in the cases of H, He, Li, B, and Be ions studied previously. We find that, while C ions can still be described quite well by the SPLA, the values for O ions show instead a significant overestimation. The origin of these discrepancies is related to the linear response approximation (undistorted plane waves) used to calculate the Lindhard dielectric function. A nonlinear version of the SLPA may be possible by using a different dielectric function, such as the Coulomb-Lindhard one [26], but it is difficult to be implemented in the present case.

To summarize, a combined experimental and theoretical study of the energy loss of C and O ions in Zn has been made. Stopping cross sections data for C and O ions in Zn are presented. These measurements fill a gap in the stopping data available in the literature corresponding to the low-intermediate energy range, which includes the maximum of the stopping power.

The theoretical work presented here yields two main results concerning the Zn target:

(1) We determine the limit of applicability of the SLPA model, which was successfully applied to projectiles from H to B, and even to C ions but clearly overestimates the O case.

(2) By using the nonperturbative CasP calculations together with the TCS-EFSR method, we obtained a more accurate description of the excitation of bound electrons, which incorporates the property of saturation in the energy loss with increasing Z_1 values. In this way, we are able to explain the stopping power data for C and O in Zn, achieving a good general agreement, and we reproduce the positions of the corresponding maxima, which poses a real challenge for any theoretical description.

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