Comparison of theoretical models for the electronic stopping power of low-velocity heavy ions

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Calculations of the electronic stopping power for low-velocity ($v < Z_1^{2/3} v_0$, $v_0 = e^2/\hbar$) heavy ions, based upon three models, are performed for two systems for which experimental data are available: 800-keV $^{14}N^+$ ions incident on amorphous solid targets from carbon to tellurium and 100-keV $^7Li^+$ ions incident on amorphous targets from carbon to selenium. The results of the models are compared with each other and with the experimental data. The models are found to offer qualitatively better fits to the oscillatory experimental data than the smooth curves of the Lindhard-Scharff theory, with a particular modification of the Firsov theory favored for predictive calculations. All the models, as implemented here, required a parameter to be determined by fitting the calculated curves to the experimental points.

I. INTRODUCTION

Knowledge of the electronic stopping power for heavy ions at low velocity is an important requirement in many applications, such as ion implantation and studies of material composition and defects and of radiation damage, particularly in nuclear reactors. In view of the absence of a comprehensive theory of electronic stopping power S_e at "low" velocity ($v \lesssim Z_1^{2/3} v_0$, where v and Z_1 are the velocity and atomic number of the projectile and $v_0 = e^2/\hbar \simeq c/137$), values for S_e must be obtained either from experiment or from models of a semiempirical or phenomenological nature. In this paper we discuss three such models which are well known and widely used and apply them to two systems for which extensive experimental data are available. Our purpose is to see how well these models can either predict or correlate such data. Discussions involving electronic stopping power are usually considered in terms of the dependence on energy and the dependence on projectile atomic number Z_1 or target atomic number Z_2. The present paper deals with the dependence of S_e on Z_2 for amorphous targets.

In this paper our principal goal is to correlate with the theoretical models some recent data for the electronic stopping power of 800-keV $^{14}N^+$ ions incident on numerous solid targets from carbon to tellurium, results obtained by Simons *et al.*[1] With the exception of studies which consider the very light ions (p or α) as projectiles, this is the most extensive study of the Z_2 oscillatory behavior of S_e. We also apply these models to 100-keV $^7Li^+$ ions incident on selected targets from carbon to selenium, results obtained by Bernhard *et al.*[2] and by Apel *et al.*[3] and previously analyzed in a single model by Pietsch *et al.*[4] We find that all models (i) agree qualitatively with the experimental data over the entire region for which the data are available, (ii) show similar behavior, but differ quantitatively from each other for certain target materials, and (iii) depend on at least one parameter, basically a scale factor to be determined by experiment.

Models for S_e at low velocity have been proposed by Lindhard and Scharff[5] and by Firsov.[6] Both use the Thomas-Fermi statistical model to describe atomic structure and both provide reasonable estimates for S_e if these are considered as average values over Z_1 or Z_2. However, a characteristic feature of the experimental data, observed for the past decade, is a periodic dependence at constant projectile velocity of S_e on either Z_1 or Z_2 when the other is fixed.[1-3,7,8] Indeed, in some cases the oscillation is so large as to produce a 100% modulation. It has been proposed[9-11] that the oscillatory behavior arises from periodic variations in atomic structure which the Thomas-Fermi model cannot describe, and that this behavior can be accounted for by the use of more realistic atomic models, such as Hartree-Fock-Slater (HFS) atomic wave functions. And indeed, during the past decade the models of Lindhard and Scharff and of Firsov have been modified in numerous ways to incorporate such atomic structure.

In the present work we consider three models: the first, a modification of the Lindhard-Scharff model; the second, a modification of the Firsov model; the third, a modification of the original theory of electronic stopping developed by Lindhard and his coworkers. This particular choice is intended to be representative of models of current interest. A significant point relating to the present study of the variation of S_e with Z_2 is that calculations are compared to sets of data for S_e developed by single experimental groups. This procedure is especially reasonable here insofar as systematic experimental errors should tend to play a minimal role. It should also be mentioned

that all of the calculational procedures employed here predict a behavior for S_e that is linear in velocity. Deviations from a strict velocity-proportional dependence have frequently been observed experimentally. Little theoretical progress on this problem has been made to date, although results are beginning to emerge.[12,13]

II. DESCRIPTION OF THE MODELS

A. Lindhard-Scharff model and modification

The model proposed by Lindhard and Scharff[5] for the electronic stopping power of low-velocity ions is based upon a general treatment of stopping by Lindhard[14] in which the incident particle is treated as an electric pulse which perturbs the target material considered as an electron gas. The application to atomic systems was made by using the Thomas-Fermi model for atomic structure to determine the density of the electron gas. The final result is summarized by their formula

$$S_e = (8\pi e^2 a_0 \xi Z_1 Z_2 / Z)(v/v_0), \tag{1}$$

where $\xi = Z_1^{1/6}$, $Z = (Z_1^{2/3} + Z_2^{2/3})^{3/2}$, and $a_0 = \hbar^2/me^2$.

A modification of the Lindhard-Scharff model to include atomic structure was proposed by Pietsch, Hauser and Neuwirth[4] who noted that the quantity $Z^{-1/3}$ related to the factor Z in the denominator of Eq. (1) plays the role of an atomic screening length. [Recall that in the Thomas-Fermi (TF) model, length scales as $Z^{-1/3}$.] Pietsch *et al.* suggested that this fixed factor be replaced by a variable one obtained by matching the Thomas-Fermi potential with a potential derived by the use of Hartree-Fock-Slater wave functions at some fixed point. Thus, if we write the atomic potential in the form $V(r) = (Z_1 Z_2 e^2/r) U(r/a)$, we equate

$$U_{\mathrm{HFS}}(r/a) = U_{\mathrm{TF}}(r/a') = U_{\mathrm{TF}}(\alpha r/a)$$

to determine a value of α which appears as the ratio of the HFS screening length to the TF screening length. The TF screening-length factors $Z_{1,2}^{1/3}$ should be replaced by the HFS screening-length factors $\alpha Z_{1,2}^{1/3}$ in the denominator of Eq. (1). As the variation in the HFS potential from one atom to the next reflects the variation in the atomic structure, such variation is thus introduced into the model for the stopping power. The position at which the potentials are matched appears as a parameter. In addition, a second parameter is introduced to account for the difference between ionic and atomic wave functions by fitting to experimental data.

B. Firsov model and modification

A second model was proposed by Firsov[6] and considers the electronic stopping to arise from the work involved in the transfer of momentum mv from the projectile to target electrons as the target electrons cross an imaginary surface S, located at the position of the potential minimum between the colliding particles, and are picked up or captured by the projectile. The electron flux is calculated from the expression $\frac{1}{4} v_e \rho_e$, where v_e and ρ_e are the electronic velocity and density, respectively. In the original work of Firsov this model was used to calculate the average excitation energy in a collision as a function of impact parameter b by evaluating v_e and ρ_e from the Thomas-Fermi atomic model and choosing the surface S to be a plane (called the Firsov plane) midway between the particles. The stopping power was first calculated from this model by Teplova *et al.*[15] by integrating the excitation energy over all impact parameters. The result may be written

$$S_e = mv \int_{b_0}^{\infty} 2\pi b\, db \int_{-\infty}^{\infty} dx \int S\, dS\, \tfrac{1}{4} v_e \rho_e.$$

The integration over x is along the trajectory of the particle, assumed to be a straight line.

A modification to this model was introduced by Cheshire *et al.*[9] and by Bhalla and Bradford[10] in which the electronic velocities and densities in this formula were calculated from HFS wave functions. The surface S was taken to be a plane, but positioned at the potential minimum. A nonzero value of the minimum impact parameter b_0 was used to be consistent with the basic assumption of a straight-line trajectory for the projectile. In practice, b_0 may be determined by fitting the theoretical curves to experimental data.

C. Lindhard-Scharff-Winther (LSW) model

The third method we discuss (the LSW method) follows closely upon the original model of Lindhard for S_e developed by Lindhard and Scharff,[16] Lindhard and Winther,[17] and Bonderup,[18] in which the stopping power is obtained from the equation

$$S_e = \frac{4\pi Z_{1,\mathrm{eff}}^2 e^4}{mv^2} \int 4\pi r^2\, dr\, \rho_e L(\rho_e, v);$$

here $L(\rho, v)$ is a specific function of the electronic density and the projectile velocity. To include the atomic structure of the target, one again chooses for ρ_e, HFS wave functions. The effective charge of the projectile $Z_{1,\mathrm{eff}}$ can be estimated from the Bohr formula[19]

$$Z_{1,\mathrm{eff}} = Z_1^{1/3} v/v_0, \tag{2}$$

which is based on considerations of capture and loss processes in which the projectile participates. Alternatively, defining $Z_{1,\mathrm{eff}}^2 = (\gamma Z_1)^2$, one could set

$$(\gamma Z_1)^2 = \sum_{i=1}^{Z} i^2 P_i, \quad \sum_{i=1}^{Z} P_i = 1,$$

where P_i is the population of the ith charge state. The first implementation of this approach to a study of Z_2 dependence was done by Rousseau, Chu, and Powers[20] in a calculation of the stopping power for α particles, extended by Chu and Powers[21] for targets up to $Z_2 = 103$ at α energies from 800 keV to 20 MeV. Studies involving this approach have recently been made by Latta and Scanlon[22,23] in order to extend this model to low-velocity projectiles. While the factor $Z_{1,\mathrm{eff}}$ can be approximated by the ideas we discussed above, the method does not seem to take account of the atomic structure of the incident projectile and hence $Z_{1,\mathrm{eff}}$ may also be determined by a fit to experimental data for a given projectile at a given energy.

From a quick reflection on the basic mechanisms for electronic stopping developed by Lindhard and by Firsov it may appear that they are quite unrelated. However, Latta and Scanlon[23] have suggested that these methods are not really different but are two approaches to the same physical mechanism for energy loss by the transfer of momentum from projectile to target electrons.

III. RESULTS

Using these three models we have calculated the electronic stopping power as a function of Z_2 for two systems: nitrogen ions incident on targets up to xenon ($Z_2 = 54$), and lithium ions incident on targets up to molybdenum ($Z_2 = 42$). Data for 800-keV $^{14}N^+$ ions incident on numerous solid targets have been obtained from the work of Simons *et al.*[1] Data from Porat and Ramavataram[24] have also been included. Somewhat more limited data for 100-keV lithium ions are available from Bernhard *et al.*[2] and from Apel *et al.*[3] For each model we have chosen the free parameter appropriate to that model by fitting the theoretical curves to the experimental points in the region from titanium ($Z = 22$) through germanium ($Z = 32$) for both the nitrogen and lithium projectiles. Our calculations employ the Hartree-Fock-Slater wave functions tabulated by Clementi and Roetti.[25] We have used ground-state wave functions of the neutral atom to describe the target and, for the Firsov model, ground-state wave functions of the singly charged ion to describe the projectile. The theoretical results along with the experimental points are shown in Figs. 1–4.

For the method based on the Firsov model, we have considered both cases of holding the Firsov plane fixed at the midpoint between atoms (fixed plane), and of positioning the plane at the potential minimum (variable plane). A comparison between these two cases is shown in Figs. 1 and 3. The results for the fixed plane are also shown in Figs. 2 and 4. The minimum impact parameter was taken as the free parameter. For nitrogen we find $b_0 = 2.11$ and 2.5 a.u. for the two cases of placing the Firsov plane at the potential minimum or at the midpoint, respectively, while for lithium we find $b_0 = 2.7$ and 4.8 a.u. for the corresponding cases.

For the modification of the Lindhard-Scharff equation proposed by Pietsch *et al.*[4] we have matched the potentials for all cases at $r = 2.0$ a.u. The resulting curves display systematic differences from the experimental data. It was suggested by these authors that these differences arise because the scaling of the TF potential was performed for the projectile treated as an atom rather than as an ion, and that an additional scaling of the projectile should be performed. The parameter so introduced by this procedure may be characterized as the ratio of the ionic to atomic screening length. We have therefore readjusted the contribution to the screening length arising from the incident ion in order that the theoretical curves fit the experimental points. These results are displayed in Figs. 2 and 4 for nitrogen and lithium ions, respectively. This method had previously been used by Pietsch *et al.*[4] in a study of the Z_2 dependence for lithium projectiles. We find for the ratio of ionic to atomic screening lengths the values of 0.735 and 0.312 for 800-keV nitrogen and 100-keV lithium ions, respectively. These values imply that the nitrogen ion is slightly smaller, and the lithium ion considerably smaller, than the corresponding atoms, which is in accord with experimental observations.

The results of the LSW method are also shown in Figs. 2 and 4. For this method we adjusted the effective charge $Z_{1,\mathrm{eff}}$ to fit the experimental data, finding $Z_{1,\mathrm{eff}}$ to be 2.5 for 800-keV nitrogen projectiles and 1.1 for 100-keV lithium projectiles. These values should be compared to the Bohr values obtained from Eq. (2) of 2.89 for 800-keV nitrogen and 1.10 for 100-keV lithium.

Shown for comparison in all the figures is the electronic stopping power obtained from the Lindhard-Scharff model of Eq. (1). The smooth variation predicted by this model contrasts with the general oscillatory nature of the experimental data, and for the nitrogen projectiles is consistently lower than the data.

One notes that all methods give rise to qualitative agreement with the experimental data, at least as regards the position of the maxima and minima of the stopping-power curves. However, there clearly are instances of quantitative disa-

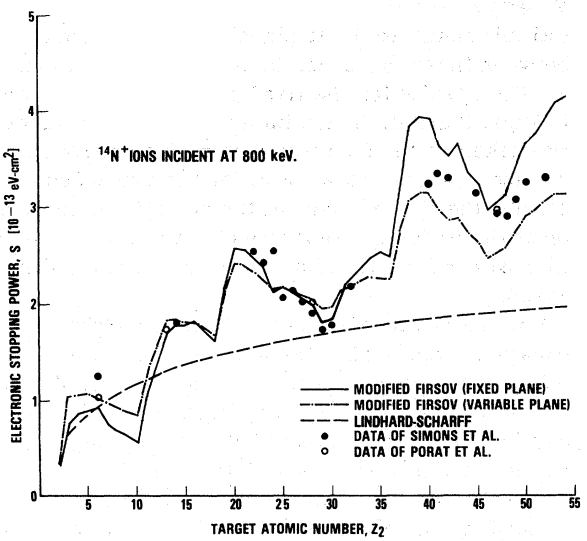

FIG. 1. Comparison of theoretical results for the electronic stopping power of 800-keV $^{14}N^+$ ions based upon two implementations of the modified Firsov method. Experimental data are included.

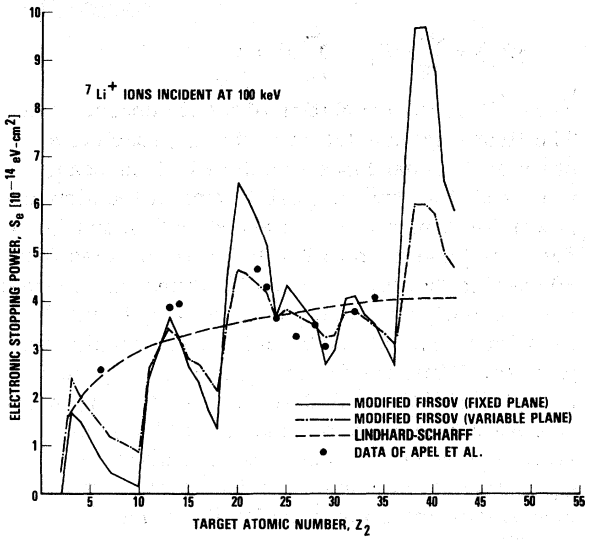

FIG. 3. Comparison of theoretical results for the electronic stopping power of 100-keV $^7Li^+$ ions based upon two implementations of the modified Firsov method. Experimental data are included.

greement. From Figs. 1 and 2 we see that for the nitrogen data[1] all methods agree quite closely with the data for targets from titanium ($Z = 22$) through zinc ($Z = 32$). This is hardly unexpected insofar as the theories were normalized in this region. However, for targets from zirconium ($Z = 40$) through tellurium ($Z = 52$), we find that either Firsov method is in closer quantitative agreement with the data than either the Pietsch or LSW methods. In addition, the data for both aluminum and silicon ($Z = 13, 14$) also agree more closely with the Firsov models. The two experimental points shown for the carbon target differ from each other, but straddle at least one theoretical curve. It has been noted[26] that the stopping power of carbon may depend on the details of the chemical composition of the particular type of target material used. From Figs. 3 and 4 we see that for the

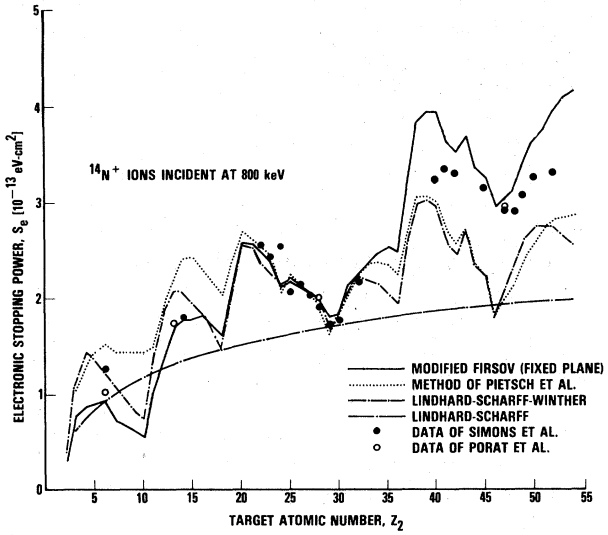

FIG. 2. Comparison of theoretical results for the electronic stopping power of 800-keV $^{14}N^+$ ions based upon the modified Firsov method, Lindhard-Scharff-Winther method, and the method of Pietsch *et al.* Experimental data are included.

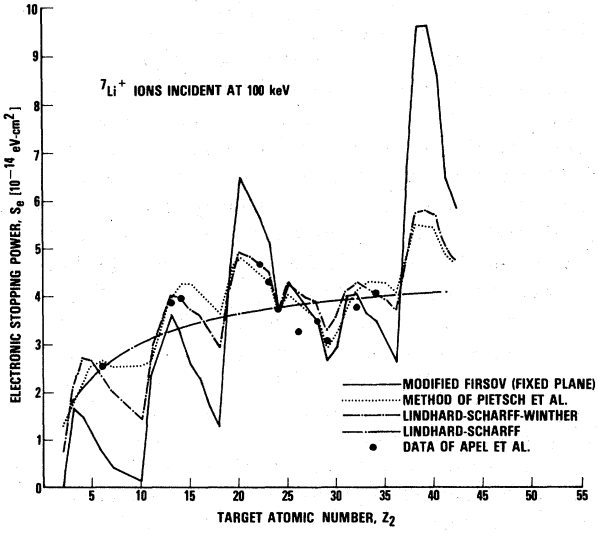

FIG. 4. Comparison of theoretical results for the electronic stopping power of 100-keV $^7Li^+$ ions based upon the modified Firsov method, Lindhard-Scharff-Winther method, and the method of Pietsch *et al.* Experimental data are included.

lithium data[2,3] we again obtain not unexpectedly good agreement for all models with the data in the region from titanium through selenium ($Z = 34$). The points at aluminum and silicon are also satisfactory. There are, unfortunately, no corresponding data for targets with atomic numbers above selenium. However, the rather unphysical character of the stopping-power curve corresponding to the fixed-plane modified Firsov method from krypton ($Z = 36$) through molybdenum ($Z = 42$) would seem to favor the variable-plane Firsov method in the correlation with the experimental data.

We summarize in Table I the parameters and their values for each method for both the nitrogen and lithium systems. The parameters for both the Pietsch and LSW method seem quite reasonable on physical grounds, as discussed above. The minimum impact parameter required by the modified Firsov models would seem at first sight to be unphysically large. Indeed, arguments based upon the exponential atomic model of Bohr would suggest that impact parameters as small as 0.01 a.u. are possible. The large values required here can be made plausible by the following consideration suggested by Denkin[13] and applied by him to the Firsov model. The energy loss in the inelastic stopping occurs by collisions with the atomic electrons. However, those electrons having orbital velocities much greater than the projectile velocity can readjust their orbits adiabatically with little energy loss. Most of the energy loss arises from those electrons having velocities equal to or less than the projectile velocity. The contribution from those inner shells having orbital velocities that are too large can be effectively eliminated by choosing the minimum impact parameter b_0 to be approximately equal to the orbital radius with $v \sim v_{\text{proj}}$. This leads to a value of b_0 in the case of copper of about 2 a.u. for the 800-keV nitrogen projectiles and 3.5 a.u. for the 100-keV lithium projectiles, values in qualitative agreement with those required here for the nitrogen and lithium data, respectively.

IV. CONCLUSIONS

At the outset we stated as our purpose to determine how well current theoretical models can correlate or predict the electronic stopping power of low-velocity heavy ions in amorphous targets as a function of target material Z_2. Here we summarize our observations to answer this question.

From a comparison of calculations based on three models with experimental data we have found that all models agree qualitatively with each other and with the data, at least as regards the existence and position of the maxima and minima of the stopping-power curves. However, systematic quantitative differences do exist. While all models offer improvement over the results of either the basic Lindhard-Scharff or Firsov models, we have argued that the modified Firsov method with variable plane gives the closest correlation to the data. The quantitative fit of this model for either the fixed- or variable-plane methods is close for the nitrogen data, but the fixed-plane method was rejected on grounds of unphysical behavior for lithium projectiles. Two different values of b_0, the minimum impact parameter, are required to give agreement between the theory and the data 2.1 a.u. for 800-keV $^{14}N^+$ ions ($v = 1.51v_0$) and 2.7 a.u. for 100-keV $^7Li^+$ ions ($v = 0.76v_0$), values which were shown to be plausible.

In a previous calculation by Land and Brennan[27] the modified Firsov method was used to obtain S_e for a series of projectiles of velocity $v = 0.63v_0$ from carbon to germanium incident on carbon, and the results were compared to the experimental data of Hvelplund and Fastrup.[8] Satisfactory agreement was obtained with the same value of b_0

TABLE I. Values of the parameters associated with three methods of evaluating the electronic stopping power for incident 800-keV $^{14}N^+$ and 100-keV $^7Li^+$ ions.

Method	Parameter	Projectile 800-keV $^{14}N^+$	Projectile 100-keV $^7Li^+$
Modified Firsov	Minimum impact parameter (a.u.)		
	Variable-plane	2.11	2.7
	Fixed-plane	2.5	4.8
Pietsch *et al.*	Ratio of ionic to atomic screening length	0.735	0.312
Lindhard-Scharff-Winther	Effective projectile charge $Z_{1,\text{eff}}$		
	Experimental	2.5	1.1
	Bohr	2.89	1.10

as had been used for 800-keV nitrogen projectiles. In another calculation both the variable-plane Firsov and the LSW models were applied in calculations of the stopping power of 25- and 50-keV protons. Here the Firsov model requires large values of b_0, about 5 or 6 a.u., to obtain numerical values of S_e which have order-of-magnitude agreement with the data. Moreover, for both of these methods the effects of target structure, which are more pronounced in the lithium curves than the nitrogen curves, are even further enhanced for the proton curves and would seem physically unreasonable. Furthermore, no good correlation with the data could be obtained with either method.[28]

From the present studies we conclude that the variable-plane Firsov method with $b_0 \approx 2.1$ a.u. is favored to provide significant improvement in predictive calculations of the electronic stopping power over previous models for projectile ions and target atoms from carbon through tellurium, the region considered here. On the basis of the success in correlating the Z_1 oscillatory data, we had previously suggested this parametrization as a universal model for electronic stopping.[7] The unsatisfactory results for proton stopping power can probably be attributed to the fact that the Firsov model is basically a statistical one to apply in cases where both projectile and target atoms have many electrons. This condition certainly excludes application to protons. The requirement of the larger values of b_0 for proton and lithium projectiles may also be related to this condition.

It is clearly an undesirable feature of the modified Firsov model that the parameter b_0, on which the numerical results for S_e depend rather sensitively, remains to be determined in a rather *ad hoc* fashion. However, this method may also be used to interpolate S_e for target materials with atomic numbers that fall between elements for which experimental data are available and from which a value of b_0, more relevant to a particular system of interest, may be determined. Throughout this work we have emphasized that the models considered here require a parameter which appears as a scaling factor chosen by fitting the results of the models to the data. To the present authors' knowledge, there is no method applicable to amorphous targets which does not contain such a parameter. For the case of channeled particles, the channel size provides a natural value for the minimum impact parameter of the modified Firsov model if this quantity retains its classical interpretation. The parameter in the Pietsch method, the effective screening length of the projectile ion, can in principle be estimated by comparing the Thomas-Fermi atomic wave function with a realistic ionic wave function, although we are not aware of any attempt to implement this. Perhaps least dependent upon a scale factor is the LSW method, since the scale factor $Z_{1,\mathrm{eff}}$ can be estimated on the basis of average charge states of the projectile in the stopping material. Nevertheless, the apparent inability of this model to cope with the atomic structure of the projectile erodes to some extent confidence in the determination of $Z_{1,\mathrm{eff}}$ by this consideration.

It is worth mentioning one feature of the low-energy proton stopping data[28]: the stopping powers for the noble gases argon and krypton are larger than the values of neighboring elements which form solid targets, in contradiction to what is predicted by the present models. To be specific, we note from any of the stopping-power curves that there is a sharp increase in the stopping power for the several elements just beyond either argon or krypton. However, this behavior appears to be peculiar to the proton data. Values of S_e for 800-keV nitrogen projectiles are either available or can be inferred from the work of Teplova *et al.*[15] and Hvelplund[29]; they are (0.35, 0.80, and 1.60) $\times 10^{-13}$ eV cm^2 for He, Ne, and Ar targets, respectively. Furthermore, if we average the values for S_e available from the work of Denkin[13] for carbon and oxygen projectiles on Kr and Xe to estimate the values for nitrogen projectiles, we obtain (2.30 and 3.52) $\times 10^{-13}$ eV cm^2. These values are seen from Fig. 1 to be reasonably well fitted by the modified Firsov model for 800-keV nitrogen projectiles.

The present investigation points to certain features of the physics of electronic stopping power which can be accommodated in models such as those considered here and which should lead to improved numerical results. We have mentioned a possible relation between projectile velocity and the minimum impact parameter. This relation should be exploited as a means of eliminating b_0 as an arbitrary parameter from the theory, one on which the numerical results depend somewhat sensitively. We have also restricted the present calculations to projectiles that are singly ionized. However, it is well known that ionic projectiles interacting with a target material cannot be characterized by a single charge state, but change their charge states as a result of the interactions. Furthermore, while an equilibrium distribution of charge states may not exist in a strict sense, approximate distributions have been estimated as a function of ion velocity. Such distributions of charge states can be built into the present models.

These two effects would be expected, in general, to give rise to deviations from the strict velocity-proportional stopping intrinsic to the present models. Such deviations have, in fact, been ob-

served and can lead to a change in the amplitude of the Z_1 or Z_2 oscillations as a function of energy. A good example of this change for heavy ions can be found in the data of Hvelplund and Fastrup[8] for a series of projectiles incident on carbon. The calculations of Chu and Powers[21] for incident α particles also exhibit this rather strikingly. However, these are not strictly low-velocity calculations.

Another aspect of the physics of electronic stopping in solid targets which we have not considered here is the effect of the binding of the outermost electrons of the target atoms in the solid. This binding is crucial to the validity or nonvalidity of the Bragg rule for the stopping power of a composite material, but should also cause departures from the stopping power for solids as calculated for free atoms. These departures can be estimated from the fine structure of the stopping-power curve for elements for which an irregularity occurs as electrons fill the outer shells, for example at chromium ($Z = 24$). Latta and Scanlon[22] have introduced a representation of the atomic density function which approximates the change of the outer electron density from free to bound atoms by writing the electronic density

$$\rho'(r) = \rho(r) + Cr^p, \quad p = 0, 2,$$

where $\rho(r)$ is the density function for the free atom and C is a normalization constant determined such that $\rho'(r)$ is normalized to the total atomic charge Z_2 within a unit cell that encloses an atom in the solid. The factor r^p represents the density of the electrons bound in the solid. This method was applied in both the LSW[22,23] and modified Firsov models.[30] Latta and Scanlon reported limited success for protons and α projectiles with the LSW model, but no significant improvement within the Firsov model.

We conclude by stating that considerable success can be achieved in fitting experimental data for the electronic stopping power of low-velocity heavy ions to theoretical models. However, much work remains, and in particular it would be highly desirable to have available additional Z_2 data for several elements throughout the Periodic Table as projectiles to test the conditions of applicability of a universal model for electronic stopping such as suggested here.

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