

Electronic energy loss of helium ions in aluminum using the extended-sum-rule method

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Using a recently derived extension of the Friedel sum rule to finite velocities we calculate the energy loss of helium ions in aluminum. The model includes the contribution due to the excitation of valence electrons according to a quantum-mechanical picture based on scattering theory (transport cross-section approach) and gives a self consistent representation of the velocity-dependent screening for swift ions. The method provides a direct evaluation of the nonlinear effects in the more complex range of energies around the stopping power maximum. In the corresponding limits, the present results converge to the existing low- and high-velocity approximations. The stopping power values calculated with this method are in good agreement with various experimental results. [S1050-2947(98)00209-1]

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I. INTRODUCTION

The problem of the energy loss of ions in solids has been for many years a subject of intense experimental and theoretical research [1,2]. One of the main theoretical problems is to formulate a quantum description of the nonlinear effects that arise from the strong perturbation that the ion produces in the solid. A consistent solution to this problem has so far been given only for the case of very slow ions moving in jellium [3–5], which represents the conduction electrons of metals. These calculations were based on the density functional (DF) formalism and in the transport cross-section (TCS) approach. On the other hand, a simplified self-consistent model based on the Friedel sum rule was also proposed [6,7]. The model emulates the DF results and has the advantage of a much simpler calculation. The extension of the method to the case of inhomogeneous electronic distributions was shown to explain very well the experimental data on the energy loss of slow ions in solids, including both the Z_1 and Z_2 dependences [8,9]. These models provide a good description of nonlinear scattering and stopping coefficients but they are all restricted to the case of slowly moving ions.

More recently, the possibilities of extending the Friedel sum rule to finite velocities were investigated [10,11]. In particular, the self-consistent model based on the Friedel sum rule was reformulated and a quantum calculation was carried out for arbitrary (nonrelativistic) velocities of the moving ions [11]. It was shown that this approach reproduces the maximum in the stopping power at intermediate velocities and merges with the perturbation results in the high-velocity limit, thus providing a complete description of the transition from the low- to the high-velocity regimes.

On the other hand, the problem of the energy loss of helium ions in metals has been theoretically investigated by several authors using different methods. A first self-consistent quantum model was proposed by Ferrell and Ritchie for slow ions [12] and the case of swift ions was considered by Arnau *et al.* [13] using a combination of dielectric and density-functional models. Further calculations by Kaneko [14], and Wang and Nagy [15], showed the possibilities of stopping-power evaluations on a wide range of

velocities using a local-density representation of the dielectric models.

In the present work we consider the application of a recently developed model [11], based on an extension of the Friedel sum-rule method, to calculate the energy loss of a beam of helium ions in aluminum. With this model we evaluate the stopping power for a wide range of velocities, including the low- and high-velocity regimes and the region around the stopping maximum. A good agreement with experiments is obtained in all cases. A description of the model is given in Sec. II. The results are reported in Sec. III, and the main conclusions are summarized in Sec. IV.

II. THEORETICAL MODEL

A. Partial stopping power

In order to represent the various charge states of the projectile we assume conditions of charge equilibrium and we calculate the average energy loss, or *stopping power* $\langle dE/dx \rangle$, as a sum of the contributions from each possible charge state, namely,

$$\left\langle \frac{dE}{dx} \right\rangle = \sum \Phi_n(v) \left\langle \frac{dE}{dx} \right\rangle_n, \quad (1)$$

where $\Phi_n(v)$ (with $n=0,1,2$) denotes the equilibrium charge-state fractions, and $\langle dE/dx \rangle_n$ is the stopping power for each charge state (i.e., the *partial stopping power* [16]). Additional contributions from capture and loss processes and inner-shell excitations are to be considered later.

The values of $\langle dE/dx \rangle_n$ will be calculated according to our previous formulation [11] using the following expression (atomic units are used):

$$\left\langle \frac{dE}{dx} \right\rangle_n = \frac{1}{4\pi^2 v^2} \int_0^{v_F} u \, du \int_{|v-u|}^{v+u} dk \, k^4 \sigma_{tr}^{(n)}(k, v) \times \left[1 + \frac{v^2 - u^2}{k^2} \right], \quad (2)$$

where v_F is the Fermi velocity of the metal and $\sigma_{\text{tr}}^{(n)}(k, v)$ is the transport cross section corresponding to the scattering of electrons in the moving-ion potential [11]. One should note the various velocities that appear in this expression: v is the ion velocity, u is the electron velocity relative to the metallic environment ($0 < u < v_F$ for a distribution of electrons within a Fermi sphere), and k is the *relative* velocity of the scattered electron with respect to the ion (the use of the variable k in this case is adopted to maintain the usual notation of scattering theory). The integrations over the angular variables have already been taken into account.

The dependence of the transport cross section σ_{tr} on the ion velocity v in Eq. (2) is an implicit result of the optimization of the scattering potential, which contains a screening parameter that is adjusted in a self-consistent way for each velocity, and for each charge state, as described in Ref. [11].

B. Velocity-dependent sum rule

The generalization of the Friedel sum rule to finite velocities was developed earlier, and we will give here only a brief description of the model. In the general case of an ion with nuclear charge Z_1 carrying a number N_b of bound electrons, the sum rule may be expressed as follows [11]:

$$Z_1 = \frac{2}{\pi} \sum_{l=0}^{\infty} (2l+1) G_l(v, v_F) + N_b, \quad (3)$$

where $G_l(v, v_F)$ takes into account the contribution of each l -wave component to the screening charge, and may be expressed as an integral of phase-shift contributions for a displaced Fermi sphere (DFS), in the form

$$\begin{aligned} G_l(v, v_F) &= \frac{1}{4\pi} \int_{\text{DFS}} \left[\frac{d\delta_l(k)}{dk} \right] d\Omega dk \\ &= \int_{k_{\min}}^{k_{\max}} \left[\frac{d\delta_l(k)}{dk} \right] g(k, v) dk, \end{aligned} \quad (4)$$

where $k_{\min} = \max\{0, v - v_F\}$, $k_{\max} = v + v_F$, and the function $g(k, v)$ is defined in Ref. [11]. Explicit expressions for $G_l(v, v_F)$ are also given in Ref. [11], and some useful analytical results for $v > v_F$ and $v < v_F$ are included in the Appendix.

The basis of this method consists of using the extended sum rule as a condition to adjust a parametrized model potential by changing the screening parameter and recalculating numerically the phase shifts until the sum rule is satisfied with enough accuracy. In this way, we adjust both the potential and the values of the phase shifts δ_l . The model provides an extension of the nonlinear transport cross section approach to all velocities of the projectile. The main assumption of this model is the limitation to the spherical average of the scattering potential; however, the self-consistent method used to adjust the potential reduces the influence of this assumption on the final results, showing a good agreement with the expected values in all cases [11].

C. Model potentials

The potentials used in the present description should satisfy at least two basic conditions:

(1) Near the nucleus the potential must have a Coulomb-like dependence, $V(r) \simeq -Z_1/r$ (for $r \rightarrow 0$). (2) Asymptotically it should be consistent with the conditions of complete screening (i.e., the total induced charge should be equal to $Z_1 - N_b$), and so it must decay faster than $1/r$ [$rV(r) \rightarrow 0$, for $r \rightarrow \infty$]. These conditions must be fulfilled for any physical values of the parameters. In the case of helium, with three possible charge states: He^0 , He^+ , and He^{2+} , we propose simple model potentials to represent each of these cases. We use combinations of hydrogenic potentials of the form

$$V(r) = -Ze^{-\alpha r} \left(\frac{1}{r} + \frac{\alpha}{2} \right). \quad (5)$$

The electronic density corresponding to this potential is given by

$$n = -\frac{1}{4\pi} \nabla^2 V = Z \frac{\alpha^3}{8\pi} e^{-\alpha r}. \quad (6)$$

In the case of ions carrying bound electrons (He^0 and He^+) the total electronic density around the moving ion will be composed of a core term n_{core} , due to the bound electrons, and a screening term n_{scr} due to the scattering of the free electrons, namely, $n = n_{\text{core}} + n_{\text{scr}}$, and the corresponding potential will be of the form $V = V_{\text{core}} + V_{\text{scr}}$.

Let us then consider each of these cases.

(1) He^{2+} : This is the simplest case, where the screening density is provided wholly by the scattering of free electrons, and hence the potential will be assumed of the form of Eq. (5), with $Z=2$. The value of the parameter α in this case will depend on the ion velocity v , since it will be determined using the velocity-dependent sum rule for each velocity.

(2) He^+ : In this case we have one bound electron in a $1s$ orbital; hence, the total electronic density and potential will be assumed of the form

$$n^{(1)}(r) = \frac{8}{\pi} e^{-4r} + \frac{\alpha^3}{8\pi} e^{-\alpha r}, \quad (7)$$

$$V^{(1)}(r) = -e^{-4r} \left(\frac{1}{r} + 2 \right) - e^{-\alpha r} \left(\frac{1}{r} + \frac{\alpha}{2} \right). \quad (8)$$

The first term in these equations represents the contribution of the bound electron (for a $1s$ state with nuclear charge $Z_1=2$), while the second term represents the additional contribution of the screening electrons corresponding to a total screening charge of unit value. As before, the value of α will be fixed by the velocity-dependent sum rule, and it will take a different value than for He^{2+} .

(3) He^0 : In this case we observe that the core term, $n_{\text{core}} = (2\gamma^3/8\pi) \exp(-\gamma r)$, already contains two electrons, which provide enough screening charge to assure the electric neutrality of the composite. However, this is not the self-consistent solution to the problem of a neutral system immersed in a screening medium, as it may be checked simply by noting that the Friedel sum rule is generally not satisfied. Then, in order to achieve self-consistency we will consider

an additional screening charge distribution, but in order to maintain the overall neutrality the total screening charge should be zero.

Therefore, we propose a convenient form for the total electronic density and potential, given by

$$n(r) = 2\frac{\gamma^3}{8\pi}e^{-\gamma r} + q\frac{\alpha^3}{8\pi}e^{-\alpha r} - q\frac{\beta^3}{8\pi}e^{-\beta r}, \quad (9)$$

$$V(r) = -2e^{-\gamma r}\left(\frac{1}{r} + \frac{\gamma}{2}\right) - qe^{-\alpha r}\left(\frac{1}{r} + \frac{\alpha}{2}\right) + qe^{-\beta r}\left(\frac{1}{r} + \frac{\beta}{2}\right). \quad (10)$$

Where, in addition to the core contribution (first term), we have included an additional screening term (with parameter α) and a compensating *antiscreening* component (with parameter β).

The values of these parameters are now taken as follows: the parameter γ will be taken equal to 3.375, which is the value corresponding to the variational calculation of He^0 in vacuum. The value of q is somewhat arbitrary and was fixed to 0.5; this might be interpreted as an indication that the screening involves a charge of order unity, similar to the previous cases, although we note that the results were not very sensitive to this value once the sum rule was satisfied since the variation of the free parameter usually compensates the change in the q value (several calculations using q values in the range 0.3–1.5 support this conclusion). Therefore we do not attribute any significant meaning to this particular value. The parameters α and β are to be adjusted using the Friedel sum rule. We note that this provides only one condition and is therefore not enough to determine both parameters. Then, we used as an auxiliary condition that the value of the phase shift δ_0 obtained with this method should coincide with the value determined from density functional theory.

III. CALCULATIONS

In Fig. 1 we show the values of the parameter α for (a) He^+ and (b) He^{2+} adjusted for each velocity using the extended Friedel sum rule of Eq. (3) with $N_b=1$ and $N_b=0$, respectively, for the case of aluminum targets ($r_s=2.07$). By comparison, the analytical values from the Born approximation to the Friedel sum rule [see Appendix, Eqs. (A10), (A11)], and the asymptotic high-velocity limit, $\alpha \approx \sqrt{2}\omega_p/v$ [Eqs. (A6), (A9)], are also shown. We observe important differences in the low-velocity range ($v \ll v_F$) due to the failure of the perturbative Born approximation, and a rapid convergence for $v \gg v_F$. The velocity dependence of α shows a relaxation in the screening conditions with increasing ion velocity. In particular, we observe a fast drop of α when $v \sim v_F$, corresponding to a sudden expansion of the screening charge around the ion. The behavior of α at larger velocities shows the characteristic dependence ($\sim \omega_p/v$) expected in the asymptotic range, as first pointed out by Bohr [17].

In Fig. 2 we show the stopping powers of the He^+ and He^{2+} components obtained from the present formulation,

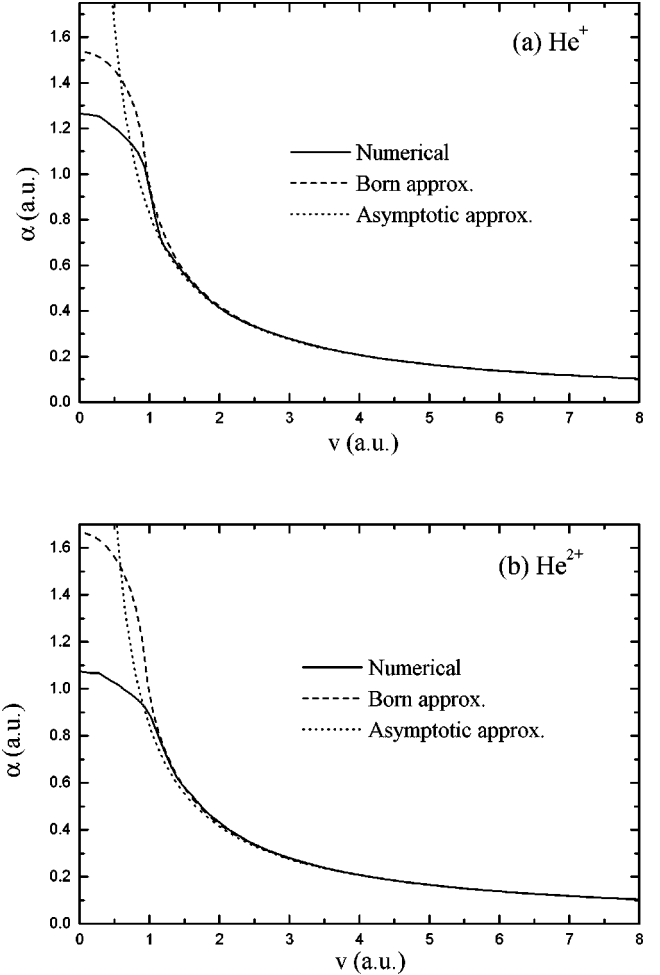


FIG. 1. Values of the screening parameter α for (a) He^+ and (b) He^{2+} in aluminum ($r_s=2.07$), as a function of ion velocity. Solid lines: α values numerically adjusted for each velocity using the extended Friedel sum rule of Eq. (3) with $N_b=1$ and $N_b=0$, respectively. Dashed lines: analytical values from the Born approximation to the Friedel sum rule [see Appendix, Eqs. (A10), (A11)]. Dotted lines: asymptotic high-velocity limits [Eqs. (A6), (A9) in the Appendix].

Eq. (2), using the α values previously determined. The ratio between these stopping values is shown in the inset. We find that this ratio is about 2 not only near the maximum but also at high velocities. This is a rather striking result since the expected Z^2 behavior at high velocities would predict a value close to four. To explain this behavior we note that the He^+ ion is not a simple point charge but a nucleus with charge 2 surrounded by the charge density of the bound electron. According to the partition rule [1]—which would apply at high velocities—the contributions to the energy loss due to close and distant collisions, in the case of pointlike charges, are of the same order; but the effective charge of the He^+ ion for close collisions should be close to two, while the effective charge for distant collisions should approach one. Therefore, the stopping power for He^+ at high velocities may be roughly approximated by an average between the stopping powers of a proton and an alpha particle, that is, about half the stopping power of He^{2+} .

In order to calculate the total stopping power for a beam of helium ions in solids we should take into account the

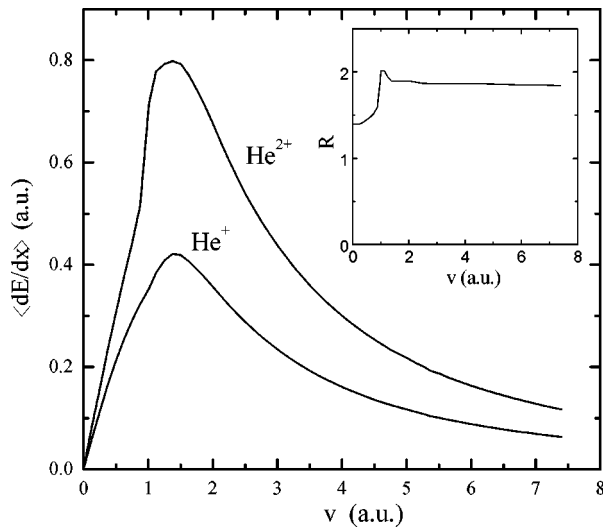


FIG. 2. Partial stopping powers for He^+ and He^{2+} in aluminum, calculated from Eq. (2) using the present method, with the values of α numerically determined for each velocity using the extended sum rule as in Fig. 1. The inset shows the stopping ratio $R = \langle dE/dx \rangle[\text{He}^{2+}] / \langle dE/dx \rangle[\text{He}^+]$.

population of each charge state, described by the equilibrium charge-state fractions $\Phi_n(v)$ (with $n=0,1,2$) as in Eq. (1). Since no direct experimental data of these fractions within the solid exist, we used the values calculated by Echenique *et al.* [18] for the case of aluminum targets. In Fig. 3 we show the partial stopping powers of He^0 , He^+ , and He^{2+} , taking into account the charge state fraction for each case. As expected, the He^0 fraction dominates the stopping process at low velocities while He^{2+} determines the high-velocity stopping. The figure also shows the total stopping power given by Eq. (1) corresponding to excitations of free electrons (FE).

The calculations already discussed describe the excitations of the free electrons in aluminum (corresponding to

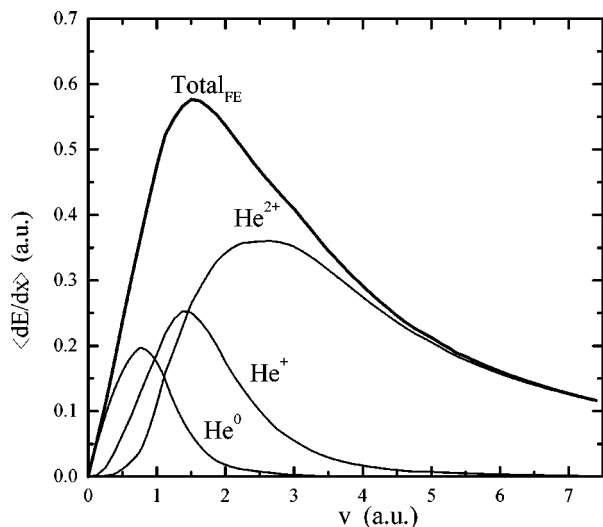


FIG. 3. Stopping power contributions for He^0 , He^+ , and He^{2+} in aluminum, calculated as in Fig. 2, and weighted by the corresponding charge state fractions from Ref. [18]. The upper curve is the sum of the three contributions and gives the total stopping power of the free-electron gas in the present method.

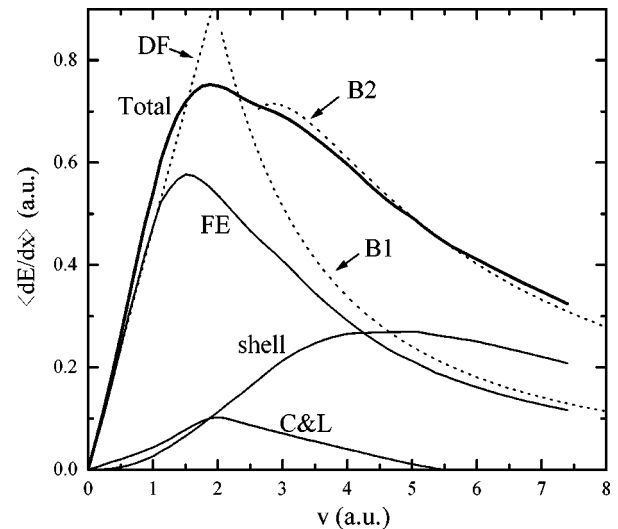


FIG. 4. Contributions to the stopping power from various processes. The curve denoted FE gives the free-electron gas contribution calculated with the present model. The C&L curve shows an estimation of capture and loss processes according to Ref. [13]. The contribution from inner shells was calculated following Ref. [19]. The upper solid curve shows the total stopping power values. The curve DF is the density functional prediction for low velocities and the curves denoted B1 and B2 are the asymptotic Bethe limits for the free electron gas and for the whole system.

three free electrons per atom), and represent the main contribution to the energy loss process for light ions at low and intermediate velocities. This is illustrated in Fig. 4, where we compare the present results for free electron excitations (FE) with the other mechanisms that contribute to the stopping power, namely, shell excitations [19] and capture and loss (CL) processes [13], according to the evaluations given by previous authors for each case. The contribution from CL processes estimated in Ref. [13] amounts to about 15% of the total in the range of the maximum stopping, although a precise calculation of these terms is difficult due to the inaccuracy of the perturbative models in this energy range. Some authors have neglected or estimated smaller values for these terms [14,15,20]. The shell processes give the contribution from the excitation of the *L*- and *K*-shell electrons of aluminum [19]; these excitations become more important in the high-energy range, as expected from the Bethe theory. By comparison, we also show in the figure the results from the density-functional theory (in the low-velocity range), and the asymptotic results predicted by the Bethe theory for the free electron component (B1) and including the effects of inner-shell excitations (B2).

In Fig. 5 we show the final results of the present calculation, including the mentioned corrections, and compare them with all the available experimental results from different sources [21–27]. We observe a quite satisfactory agreement both in the low- and high-energy ranges, and a slight overestimation around the maximum. It does not seem possible to attribute a unique origin to these residual differences due to the mixed contributions from different processes that occur in the intermediate range of velocities. However, we observe a satisfactory representation of the shape of the stopping curve on the whole energy range.

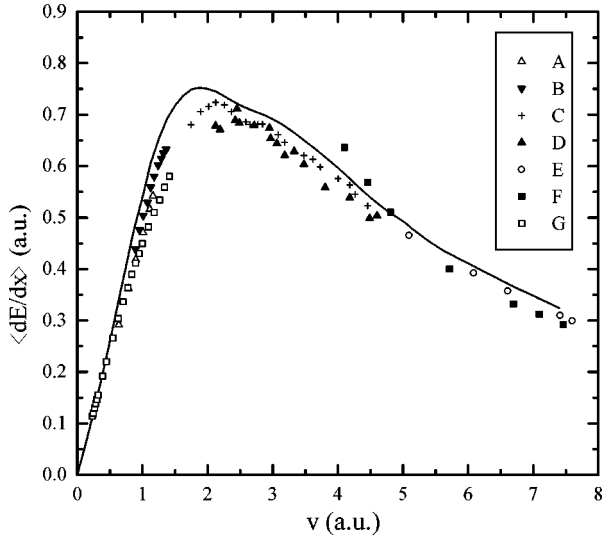


FIG. 5. Stopping power values for helium in aluminum as obtained from the present model, compared with experimental data from various sources: (a) Ref. [21], (b) Ref. [22], (c) Ref. [23], (d) Ref. [24], (e) Ref. [25], (f) Ref. [26], and (g) Ref. [27].

IV. SUMMARY AND CONCLUSIONS

The calculations reported here provide a nonperturbative quantum calculation of the stopping power for moving ions through the whole range of nonrelativistic energies, including the main region around the stopping-power maximum.

The model proposed is based on an extension of the Friedel sum rule to finite velocities of the ion, using the quantum scattering formulation and the transport cross-section approach. The model converges to the existing low- and high-velocity models in the corresponding limits, but in addition it describes the region of the stopping-power maximum where other approximations usually fail. The model provides a quantitative estimation of the role of nonlinear effects in the more complex range of intermediate energies. The present method describes also the transition from the nonlinear to the linear-screening conditions as the ion velocity increases.

In order to allow a comparison with experimental data, the present calculations for free-electron excitations must be supplemented with correction effects due to capture and loss processes and inner-shell excitations. These further corrections are evaluated using perturbative methods that are usually inaccurate for low and intermediate velocities, but there is no evidence so far that this may significantly affect the total results since, on one side, the contribution from capture and loss processes is relatively small, whereas inner-shell excitations become relevant only for high velocities where perturbative methods apply. It is perhaps due to these favorable conditions that the influence of the correction terms does not affect very much the final results.

From the comparison with the experimental results we may therefore infer that the present model describes in a satisfactory way the dynamical effects related to the velocity-dependent screening of moving ions, and the stopping-power due to the excitations of free electrons in metals, which yield the main contribution to the energy loss in the interesting range of intermediate velocities. We expect that further applications of the present method to other interacting elements will be of much help in providing a more complete under-

standing of the dominant processes around the stopping-power maximum.

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APPENDIX: HIGH-VELOCITY APPROXIMATIONS

We include in this appendix some useful approximations, corresponding to the perturbative (or first-order Born approximation), which apply in the limit of high-velocities.

By expanding the function $G_l(v, v_F)$ in the limit $v \gg v_F$ we get the high-velocity expression of the Friedel sum rule [11]:

$$Z_1 \cong \left(\frac{v_F^3}{3v^2} \right) \frac{2}{\pi} \sum_l (2l+1) \left[\frac{d\delta_l}{dk} \right]_{k=v} + N_b. \quad (\text{A1})$$

In this limit one can use the Born approximation to calculate the phase shifts, namely,

$$\delta_l^B(k) = -2k \int_0^\infty dr r^2 V(r) [j_l(kr)]^2. \quad (\text{A2})$$

Using this approximation we get for a simple hydrogenic potential [11],

$$\frac{2}{\pi} \sum_l (2l+1) \delta_l^B(k) = \frac{8k}{\pi} \frac{Z_1}{\alpha^2}, \quad (\text{A3})$$

and therefore,

$$Z_1 \cong \left(\frac{v_F^3}{3v^2} \right) \left[\frac{d}{dk} \left(\frac{8k}{\pi} \frac{Z_1}{\alpha^2} \right) \right]_{k=v} + N_b = \left(\frac{8v_F^3}{3\pi v^2} \right) \frac{Z_1}{\alpha^2} + N_b. \quad (\text{A4})$$

This expression yields the value of α that satisfies the sum rule in the Born approximation (for $N_b \neq Z_1$), namely,

$$\alpha_B = \sqrt{\frac{8v_F^3}{3\pi} \frac{Z_1}{Z_1 - N_b} \frac{1}{v}} = \sqrt{\frac{2Z_1}{Z_1 - N_b} \frac{\omega_p}{v}}. \quad (\text{A5})$$

In particular, in the case of He^{2+} (with $Z_1 = 2$, $N_b = 0$) we get

$$\alpha_B = \sqrt{\frac{8v_F^3}{3\pi} \frac{1}{v}} = \sqrt{2} \frac{\omega_p}{v}. \quad (\text{A6})$$

On the other hand, in the case of He^+ using the potential $V(r)$ of Eq. (8) in Eq. (A2) we obtain

$$\frac{2}{\pi} \sum_l (2l+1) \delta_l^B(k) = \frac{8k}{\pi} \left(\frac{1}{\alpha_B^2} + \frac{1}{\beta^2} \right) + N_b, \quad (\text{A7})$$

and in particular for $N_b = 1$ and $\beta = 4$, we get

$$\alpha_B = \left(\frac{3\pi v^2}{8v_F^3} - \frac{1}{16} \right)^{-1/2}. \quad (\text{A8})$$

The term $1/16$ in this expression is negligible for $v \gg v_F$, so that to a good approximation

$$\alpha_B \approx \sqrt{\frac{8v_F^3}{3\pi}} \frac{1}{v} = \sqrt{2} \frac{\omega_p}{v}, \quad (\text{A9})$$

which converges to the case of He^{2+} , Eq. (A6).

The previous considerations can be extended to the whole range of velocities (ignoring for the moment the breakdown of the Born approximation at low velocities), following the derivation given in Ref. [11] [cf. Eq. (19) of this reference]. By repeating those calculations we obtain

(a) for He^+ :

$$\alpha_B^2 = \left(\left\{ \frac{4v_F}{\pi} \left[1 + \frac{v_F^2 - v^2}{2v_F v} \ln \left| \frac{v + v_F}{v - v_F} \right| \right] \right\}^{-1} - \frac{1}{16} \right)^{-1} \quad (\text{A10})$$

(b) for He^{2+} :

$$\alpha_B^2 = \frac{4v_F}{\pi} \left[1 + \frac{v_F^2 - v^2}{2v_F v} \ln \left| \frac{v + v_F}{v - v_F} \right| \right]. \quad (\text{A11})$$

These approximations are compared with the exact numerical values of α in Fig. 1. It may also be observed that both values are nearly coincident for $v \gg v_F$.

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