Bethe theory of stopping incorporating electronic excitations of partially stripped projectiles

R. Cabrera-Trujillo,[1,2] S. A. Cruz,[2] Jens Oddershede,[1,3] and John R. Sabin[1,3]
[1]*Department of Chemistry, Odense University, DK-5230, Odense M, Denmark*
[2]*Departamento de Física, Universidad Autónoma Metropolitana, Apartado Postal 55-534, Código Postal 09340 México Distrito Federal, Mexico*
[3]*Department of Physics, University of Florida, Gainesville, Florida 32611*
(Received 12 August 1996)

The Bethe theory for the energy loss of swift charged particles colliding with atomic targets is extended to treat explicitly the case when the projectile has bound electrons. The theory is characterized by taking into account the excitation and ionizations (but not charge exchange) of both projectile and target. As a consequence of the Coulombic character of the interaction between projectile and target, the electronic stopping cross section S_e is split into contributions arising from the electronic structure of the projectile and of the target. In the lower part of the velocity region where this theory is applicable, the electronic structure of the projectile makes an important contribution, of the order 10–20 %, to S_e. At high projectile velocities, however, this contribution decreases, leaving only the standard Bethe term. The number of electrons bound to the projectile, N_1, as a function of the velocity of the projectile is calculated using the adiabatic Bohr criterion in conjunction with the Thomas-Fermi model of the atom. We obtain an analytic expression for the total stopping cross section using the Bethe approximation, and we compare the results of calculations of He, Li, and B ions on C and Al targets with experiment. [S1050-2947(97)06903-5]

PACS number(s): 34.50.Bw, 34.10.+x

I. INTRODUCTION

The energy loss of swift ions moving through matter has been the subject of extensive theoretical and experimental study since the beginning of the century [1]. In recent years, the stopping of partially stripped heavy ions in matter has received increased attention both experimentally and theoretically [2]. However, a first-principles description of the influence of the projectile electronic structure on the stopping has not yet been formulated. This is the subject of the present paper.

For fully stripped projectiles with velocity v much greater than the orbital velocity v_e of the target electrons, the Bethe-Bloch theory [3,4] accounts for the electronic stopping cross section with good accuracy. According to Bethe's theory [3], the electronic stopping cross section for a swift stripped ion with charge Z_1 is given by

$$S_e(v) = -\frac{1}{n_2}\frac{dE}{dx} = \frac{4\pi e^4}{m_e v^2} Z_1^2 L(v), \tag{1.1}$$

where $L(v) = Z_2 \ln(2mv^2/{}_2I_0)$ is the stopping number, ${}_2I_0$ is the mean excitation energy of the target, $-dE/dx$ is the energy loss per unit path (stopping power), and n_2 is the number of atoms per volume of the target. Here and below, the subscripts 1 and 2 refer to the projectile and target, respectively. However, while penetrating matter, the projectile ions may carry, at least temporarily, some bound electrons even if $v > v_e$ [5,6]. Furthermore, capture and loss of electrons by the projectile are processes with a high probability, in particular in the velocity range $v_0 < v < v_0 Z_1^{2/3}$, v_0 being the Bohr velocity, leading to important contributions to the energy loss [7]. Thus, in order to obtain a proper description of the stopping cross section for a wide range of velocities, one must consider the electronic structure of both projectile and target.

The first study along these lines was carried out in the mid 1950s by Bates and Griffing [8], who calculated the electronic cross section for the simplest collision process: two hydrogen atoms. This work formed the basis for systematic studies of more general atom-atom interactions [9].

During the last few years, the charge-state dependence of electronic stopping has become an active research topic. Previous treatments based on the average scaling rules such as the effective charge theory [10], although useful for estimating the mean stopping power of heavy ions, still represent an incomplete description of the electron capture and loss processes involved in a collision. Recently, more realistic and sophisticated models based on the numerical solution of the time-dependent Schrödinger equation [11,12], time-dependent variational principle [13], or density-functional theory [14] has been suggested. However, these methods are currently only feasible for H- and He-like projectiles, so there is still a need for theories such as the one proposed here treating more general projectiles.

Within the spirit of the Bethe theory, there have been various efforts to account for the stopping of partially stripped ions. A scheme was suggested by Kim and Cheng [5] where the electronic stopping cross section S_e is obtained from the original Bethe formula by replacing the projectile nuclear charge Z_1, and target mean ionization energy ${}_2I_0$ by Z_{eff} and I_{eff}, respectively. Both Z_{eff} and I_{eff} are derived from properties of the projectile and target that can be calculated from first principles.

Similarly, Arnau and Echenique [15] proposed a way to introduce the electronic structure of the projectile into the dielectric theory of stopping. More recently, Moneta and Czerbniak [16] have used the first-order Born approximation

for estimating the stopping power of partially stripped ions based on the semiclassical impact parameter description. In their model, the projectile electrons are allowed to be excited or ionized but no electron exchange is considered.

The purpose of this work is to report a first Born approximation Bethe-like theory for partially stripped projectiles carrying a velocity-dependent number of bound electrons. In contrast to the treatment of Moneta and Czerbniak [16], our formulation does not depend on the impact parameter, and the number of electrons bound to the projectile is obtained using the Bohr adiabatic criterion [17]. Furthermore, it naturally separates the contributions to the stopping originating from the projectile and target.

Section II A provides the general first Born approximation theory of stopping power for an ion with electronic structure impinging on an atomic target. The basic formula for energy loss including excitation and ionization of the compound projectile-target system is developed. In Sec. II B we perform the standard Bethe approximation for the stopping power, assuming a random, isotropic distribution of target atoms, i.e., we do not consider directional dependence [18]. In Sec. II C, we invoke the Bohr criterion [17] for the number of electrons stripped from the projectile, thus obtaining an analytical expression for the ionization fraction (i_f) of the ion as a function of its velocity. In Sec. II D we combine the results of Secs. II A, II B, and II C to obtain a final expression for the stopping cross section. In Sec. III we discuss and compare our results with other theories and experiment, and, finally, in Sec. IV we present our conclusions.

II. STOPPING POWER

A. The first Born approximation

Let us consider a process in which an ion moving with a velocity v (in the laboratory frame), mass M_1, nuclear charge Z_1e, and carrying N_1 bound electrons described by the electronic eigenstate $|n_0\rangle$ collides with a stationary target with mass M_2 and N_2 bound electrons in an initial state denoted $|m_0\rangle$. The projectile is deflected into the solid-angle element $d\Omega$ along a direction with polar angle (θ,φ) measured in the laboratory frame. Suppose the projectile-target system undergoes a transition to final states $|n\rangle$ and $|m\rangle$, respectively, with energies E_n and E_m. Then the kinetic energy of the projectile, when electron transfer is not permitted, is thereby reduced by $(E_n-E_{n_0})+(E_m-E_{m_0})=\hbar(w_{nn_0}+w_{mm_0})$.

The stopping cross section [19] is given by

$$S_e(v)=\sum_{n,m}\int\frac{d\sigma_{n,m}}{d\Omega}[(E_n-E_{n_0})+(E_m-E_{m_0})]d\Omega,$$

where $d\sigma_{n,m}/d\Omega$ is the differential cross section.

When the projectile is sufficiently fast but still nonrelativistic, the differential cross section calculated in the first Born approximation for the interaction V between the projectile and target becomes [20]

$$\frac{d\sigma_{n,m}}{d\Omega}=\frac{M_1^2}{(2\pi)^2\hbar^4}\frac{k}{k_0}|\langle nmk|V|n_0m_0k_0\rangle|^2, \qquad (2.1)$$

where

$$\begin{aligned}\langle nmk|V|n_0m_0k_0\rangle=\int e^{(-i\mathbf{q}\cdot\mathbf{R})}&\psi_m^*(\mathbf{y}_1,\ldots,\mathbf{y}_{N_2})\\&\times\phi_n^*(\mathbf{x}_1,\ldots,\mathbf{x}_{N_1})V(\mathbf{x},\mathbf{y},\mathbf{R},)\\&\times\psi_{m_0}(\mathbf{y}_1,\ldots,\mathbf{y}_{N_2})\phi_{n_0}(\mathbf{x}_1,\ldots,\mathbf{x}_{N_1})\\&\times d\mathbf{x}_1\cdots d\mathbf{x}_{N_1}d\mathbf{y}_1\cdots d\mathbf{y}_{N_2}d\mathbf{R}. \qquad (2.2)\end{aligned}$$

Here, $\mathbf{R}$ is the position of the projectile relative to the center of mass of the target, $\mathbf{x}_i$ the position vector of electron i of the projectile with respect to the center of mass of the projectile, $\mathbf{y}_j$, the position vector of the electron j of the target with respect to the center of mass of the target. The momentum transfer is $\hbar\mathbf{q}=\hbar(\mathbf{k}-\mathbf{k}_0)$, with $\hbar\mathbf{k}_0$ being the momentum of the projectile before the collision and $\hbar\mathbf{k}$ the momentum of the projectile after the collision. The ψ's and ϕ's are the electronic eigenfunctions $|m\rangle$ and $|n\rangle$ in the coordinates $\mathbf{y}_i$ and $\mathbf{x}_i$ for target and projectile, respectively.

Since the interaction between projectile and target is Coulombic, the interaction potential is given by

$$\begin{aligned}V(\mathbf{x},\mathbf{y},\mathbf{R})=\frac{Z_1Z_2e^2}{R}-\sum_{i=1}^{N_2}\frac{Z_1e^2}{|\mathbf{R}-\mathbf{y}_i|}-\sum_{i=1}^{N_1}\frac{Z_2e^2}{|\mathbf{R}+\mathbf{x}_i|}\\+\sum_{i,j=1}^{N_1N_2}\frac{e^2}{|\mathbf{y}_j-\mathbf{R}-\mathbf{x}_i|}. \qquad (2.3)\end{aligned}$$

Following Bethe's classic derivation, Eqs. (2.1) and (2.2) can be simplified when we perform the integration over $\mathbf{R}$. The first term of Eq. (2.3) does not contribute to S_e owing to the orthogonality of the ground- and excited-state electronic wave functions for the projectile and for the target. Using the Bethe integral [21] and some straightforward manipulation, we find that the stopping cross section can be written as

$$\begin{aligned}S_e(v)&=\frac{2e^4}{m_ev^2}\sum_{n,m}\left\{\int_{q_{\min}}^{q_{\max}}|Z_2\delta_{m_0m}-{}_2M_{m_0m}(\mathbf{q})|^2{}_1F_{n_0n}(\mathbf{q})\frac{d\mathbf{q}}{q^2}\right.\\&\left.+\int_{q_{\min}}^{q_{\max}}|Z_1\delta_{n_0n}-{}_1M_{n_0n}(\mathbf{q})|^2{}_2F_{m_0m}(\mathbf{q})\frac{d\mathbf{q}}{q^2}\right\}\\&=S_{e,1}(v)+S_{e,2}(v), \qquad (2.4)\end{aligned}$$

where

$$\begin{aligned}{}_1M_{n_0n}(\mathbf{q})&=\left\langle n_0\left|\sum_{j=1}^{N_1}e^{-i\mathbf{q}\cdot\mathbf{x}_j}\right|n\right\rangle,\\{}_2M_{m_0m}(\mathbf{q})&=\left\langle m_0\left|\sum_{j=1}^{N_2}e^{-i\mathbf{q}\cdot\mathbf{y}_j}\right|m\right\rangle\end{aligned} \qquad (2.5)$$

are the atomic form factors [1,22] of the transition between the initial state and the final state of the projectile and target, respectively, and where we have made use of the definition $\mathbf{q}=\mathbf{k}-\mathbf{k}_0$ to write $d\Omega=q\,dq\,d\varphi/kk_0=d\mathbf{q}/kk_0$. The generalized oscillators strengths (GOS) for projectile and target are defined as [3]

$$ {}_1F_{n_0n}(\mathbf{q}) = \frac{2m_e}{\hbar q^2} |{}_1M_{n_0n}(\mathbf{q})|^2 w_{nn_0}, $$
$$ {}_2F_{m_0m}(\mathbf{q}) = \frac{2m_e}{\hbar q^2} |{}_2M_{m_0m}(\mathbf{q})|^2 w_{mm_0}. \tag{2.6} $$

The limits of the q integration in Eq. (2.4) are determined by the kinematics of the collision process. The minimum momentum transfer is calculated assuming that there is at least one internal excitation for the projectile-target system. The maximum momentum transferred to an electron of the target in a collision is calculated by conservation of momentum and energy. Therefore for heavy ions [19]

$$ q_{\min} = \frac{w_{nn_0} + w_{mm_0}}{v}, \quad q_{\max} = \frac{2m_e v}{\hbar}. \tag{2.7} $$

Equation (2.4) is a first Born approximation expression applicable to the electronic stopping power of ions with electronic structure. The first term on the right-hand side of Eq. (2.4) contains the quantity ${}_1F_{n_0n}$, and thus gives the contributions to the stopping cross section from the atomic transition of the projectile modulated by the electronic structure of the target. Similarly, the second term containing ${}_2F_{m_0m}$ describes the contributions from the transitions in the target modulated by the electronic structure of the projectile. Hence, the two terms in Eq. (2.4) represent the contribution to the stopping power related to electronic transitions in the projectile and target, respectively. Knowing the GOS's for both projectile and target, it is a straightforward task to compute $S_e(v)$ from Eq. (2.4). However, it is necessary to know the full excitation spectrum of both the projectile and the target, and it may be necessary to resort to approximate methods.

B. The Bethe approximation

In order to derive an approximate formula for the stopping cross section $S_e(v)$ involving the mean excitation energies of projectile and target, it is convenient to introduce the Bethe sum rule [3] for projectile and target:

$$ \sum_n {}_1F_{n_0n}(\mathbf{q}) = N_1, \quad \sum_m {}_2F_{m_0m}(\mathbf{q}) = N_2. \tag{2.8} $$

Assuming Hartree-Fock atomic wave functions, we may show that

$$ \sum_n |Z_1\delta_{n_0n} - {}_1M_{n_0n}(\mathbf{q})|^2 = |Z_1 - {}_1M_{n_0n_0}(\mathbf{q})|^2, $$
$$ \sum_m |Z_2\delta_{m_0m} - {}_2M_{m_0m}(\mathbf{q})|^2 = |Z_2 - {}_2M_{m_0m_0}(\mathbf{q})|^2, \tag{2.9} $$

where ${}_1M_{n_0n_0}(\mathbf{q})$ and ${}_2M_{m_0m_0}(\mathbf{q})$ are the electronic atomic form factors of the ground state of projectile and target, respectively.

Following Bethe [19], we interchange the summation over (m,n) in Eq. (2.4) with the integration over $\mathbf{q}$, and we replace $q_{\min}$ by a suitable average value, which is independent of the excited state of the system:

$$ q_{\min_{\mathrm{AV}}} = \frac{(w_{nn_0} + w_{mm_0})_{\mathrm{AV}}}{v} = \frac{\epsilon}{\hbar v}. \tag{2.10} $$

Using this approximation and the sum rules [Eqs. (2.8) and (2.9)], we obtain

$$ S_e(v) = \frac{2e^4}{m_e v^2} \int_{\epsilon/\hbar v}^{2m_e v/\hbar} \{N_2[Z_1 - {}_1M_{n_0n_0}(\mathbf{q})]^2 + N_1[Z_2 - {}_2M_{m_0m_0}(\mathbf{q})]^2\} \frac{d\mathbf{q}}{q^2}. \tag{2.11} $$

Thus, we have expressed the electronic stopping power in terms of a yet unspecified quantity ϵ, which depends on the projectile-target system in question.

We may obtain an expression for ϵ by recalling that it is defined in order to allow the interchange of integration and summation in Eq. (2.4). Following the procedure outlined in the Appendix, we obtain

$$ \epsilon = {}_2I_0^{1/(1+\alpha)} {}_1I_0^{\alpha/(1+\alpha)}, \tag{2.12} $$

where

$$ \alpha = \frac{(Z_2 - N_2)^2 N_1}{(Z_1 - N_1)^2 N_2}. \tag{2.13} $$

Equation (2.11) is the resulting stopping formula in the Bethe approximation when excitations and ionization of projectile and target are taken into account, that is, assuming nonrelativistic projectile velocities v much larger than orbital velocities of both target and projectile bound electrons.

C. Adiabatic assumption for $N_1(v)$

We have proposed a modification to the Bethe theory of stopping power for a projectile with bound electrons. In order to apply this theory, we need to know the number of electrons bound to the projectile N_1.

Until now, we have tacitly assumed N_1 to be constant for all velocities v. In fact, it is a fluctuating quantity, and the momentary distribution of electrons is determined by the electronic capture and loss processes that take place along the trajectory of the ion. Often, a dynamic equilibrium between capture and loss is established, and it makes sense to define $N_1 = N_1(v)$, the equilibrium number of electrons bound to the projectile as a function of the velocity of the projectile. We shall make this approximation here.

In general, the calculation of an equilibrium distribution of electrons in the projectile requires detailed estimates of the cross sections for electron capture and loss. However, the number of electrons can be estimated by making the assumption that the projectile electrons are stripped if their orbital velocity v_e is less than the projectile velocity, while electrons with $v_e > v$ are kept since the collisions—seen, for example, from the system moving with the projectile—are adiabatic. This is the adiabatic Bohr criterion [17]. Such a criterion can be rewritten in the form of a potential-energy condition [23]

$$\tfrac{1}{2}mv^2+U(r_c)=0, \tag{2.14}$$

where r_c is the distance from the nucleus of an electron bound to the projectile for which its velocity is equal to the velocity of the projectile ($v_e=v$). An electron for which $r>r_c$ is moving with $v_e<v$ and therefore is stripped from the projectile.

The number of electrons stripped from the projectile is

$$Z_1-N_1(v)=\int_{r_c(v)}^{\infty}\rho_{e,1}(r)d\mathbf{r}, \tag{2.15}$$

where r_c is the solution of Eq. (2.14) and $\rho_{e,1}(r)$ is the electron density of the neutral projectile.

In order to determine r_c, and thus N_1, it is necessary to know v_e as a function of r. Even though this is possible in Hartree-Fock theory, the approximate nature of the present approach does not warrant such a complicated procedure. We therefore make use of Thomas-Fermi theory (TF) [24], which describes an ion with N_1 bound electrons by the radially symmetric electron density

$$\rho_{e,1}(r)=\frac{N_1}{4\pi\Lambda_1^2 r}\frac{d^2\Phi(x)}{dx^2}, \tag{2.16}$$

where $x=r/\Lambda_1$, $\Phi(x)$ is the screening function that is solution of the TF equation, and the screening length, Λ_1 is treated as a variational parameter [see Eq. (2.18)] obtained by minimizing the total energy

$$E=E_{ne}+\lambda E_{ee}+E_{\rm kin} \tag{2.17}$$

as a function of the number of electron N_1. Here, the parameter λ is introduced in order to include both correlation and exchange [10]. The total energy is evaluated subject to the conditions

$$\frac{\partial E}{\partial\Lambda_1}=0, \quad \left.\frac{\partial E}{\partial N_1}\right|_{Z_1}=0, \tag{2.18}$$

where the second condition ensures that the neutral atom has lower energy than its ions.

We seek forms of $\Phi(x)$ that permit us to obtain analytical results for atoms without extensive recourse to numerical methods. To this end, we choose for $\Phi(x)$ an approximate solution to the TF equation. According to Tietz [25], the screening function for the TF atom can be written as

$$\Phi(x)=\frac{b^2}{(x+b)^2}, \tag{2.19}$$

where we choose $b=(8/\pi)^{2/3}$ to make sure that the electronic density is normalized [25]. Inserting Eq. (2.19) in Eq. (2.16), and following the standard procedure [26] for calculation of the total energy [Eq. (2.17)], we obtain

$$E=-\frac{Z_1^{7/3}}{c}\left(\frac{N_1}{Z_1}\right)^{1/3}\left(1-\frac{\lambda}{5}\frac{N_1}{Z_1}\right)^2\frac{e^2}{a_0} \tag{2.20}$$

and

$$\Lambda_1=\frac{ca_0}{Z_1^{1/3}b[1-(\lambda/5)(N_1/Z_1)]}\left(\frac{N_1}{Z_1}\right)^{2/3} \tag{2.21}$$

with $\lambda=\frac{5}{7}$ and $c=0.969376$.

From Poisson's equation, we can then obtain the electronic potential, and using Eq. (2.14), we solve for $x_c=r_c/\Lambda_1$, obtaining

$$x_c(v)=-2\left(\frac{b}{3}\right)+\frac{1}{[a/2+(b/3)^3+\sqrt{(a/2)^2+a(b/3)^3}]^{1/3}}\left(\frac{b}{3}\right)^2+\left\{\frac{a}{2}+\left(\frac{b}{3}\right)^3+\left[\left(\frac{a}{2}\right)^2+a\left(\frac{b}{3}\right)^3\right]^{1/2}\right\}^{1/3}, \tag{2.22}$$

where $a(v)=b^2/0.60647y^2(v)$, and $y(v)=v/(v_0Z_1^{2/3})$ is the effective ion velocity. This is the radial distance, measured from the nucleus of the projectile, for which the electronic orbital velocity is equal to the projectile velocity.

Finally, substituting Eqs. (2.16) and (2.19) into Eq. (2.15), the following relation for the number of average electrons kept by the projectile moving with a velocity v is obtained:

$$N_1(v)=Z_1\left(1-\frac{b^2[3x_c(v)+b]}{[x_c(v)+b]^3}\right). \tag{2.23}$$

D. Analytic expressions for $S_e(v)$

In order to obtain an analytical expression for the stopping cross section $S_e(v)$ in the first Born approximation, we also need the atomic form factors for both projectile and target [see Eq. (2.11)]. Due to the assumed spherical symmetry of the ground state $|0\rangle$ of both projectile and target, Eq. (2.5) becomes

$${}_iM_{00}(q)=4\pi\int\frac{\rho_{e,i}(r)\sin(qr)}{qr}r^2dr, \quad i=1,2, \tag{2.24}$$

where $\rho_{e,i}(r)$ is the ground-state electronic density. Introducing Eqs. (2.16) and (2.19) in Eq. (2.24) we obtain

$${}_iM_{00}(q)=N_i[1-(q\Lambda_i b)^2 g(q\Lambda_i b)], \tag{2.25}$$

where

$$g(z)=-\cos(z)Ci(z)-\sin(z)si(z) \tag{2.26}$$

is the auxiliary function for the sine and cosine integrals [27]. Using the approximate analytical representation for $g(z)$ [28],

$$g(z)=\frac{0.37093}{z^2+13.88129}+\frac{0.62907}{z^2+0.967375}, \tag{2.27}$$

we obtain an analytical expression for the atomic form factor.

Introducing Eqs. (2.25) and (2.27) in Eq. (2.11), we find that the contributions to the stopping cross section from the electronic excitations in the projectile becomes

$$S_{e,2}(v)=\frac{4\pi e^4}{m_e v^2}N_2 Z_1^2\left\{ i_f^2\ln\left(\frac{q_{\max}}{\overline{q}_{\min}}\right)+(1-i_f)\left[0.448685[i_f+0.402031]\ln\left(\frac{(q_{\max}\Lambda_1)^2+0.27819}{(\overline{q}_{\min}\Lambda_1)^2+0.27819}\right)\right.\right.$$
$$+0.0513151[i_f+6.22848]\ln\left(\frac{(q_{\max}\Lambda_1)^2+3.99187}{(\overline{q}_{\min}\Lambda_1)^2+3.99187}\right)\Bigg]+(1-i_f)^2\Bigg(0.0550439\left[\frac{1}{(q_{\max}\Lambda_1)^2+0.27819}\right.$$
$$\left.\left.-\frac{1}{(\overline{q}_{\min}\Lambda_1)^2+0.27819}\right]+0.274619\left[\frac{1}{(q_{\max}\Lambda_1)^2+3.99187}-\frac{1}{(\overline{q}_{\min}\Lambda_1)^2+3.99187}\right]\Bigg)\right\}, \tag{2.28}$$

with $q_{\max}=2mv/\hbar$, $\overline{q}_{\min}=\epsilon/\hbar v$, and the ionization fraction [10] i_f is defined as $i_f=1-N_1(v)/Z_1$. In the same way, we obtain the contribution to the stopping cross section from the electronic structure of an ionic target as

$$S_{e,1}(v)=\frac{4\pi e^4}{m_e v^2}Z_1(1-i_f)Z_2^2\left\{\left(1-\frac{N_2}{Z_2}\right)^2\ln\left(\frac{q_{\max}}{\overline{q}_{\min}}\right)+\left(\frac{N_2}{Z_2}\right)\left[0.448685\left(1.402031-\frac{N_2}{Z_2}\right)\ln\left(\frac{(q_{\max}\Lambda_2)^2+0.27819}{(\overline{q}_{\min}\Lambda_2)^2+0.27819}\right)\right.\right.$$
$$+0.0513151\left(7.22848-\frac{N_2}{Z_2}\right)\ln\left(\frac{(q_{\max}\Lambda_2)^2+3.99187}{(\overline{q}_{\min}\Lambda_2)^2+3.99187}\right)\Bigg]+\left(\frac{N_2}{Z_2}\right)^2\Bigg(0.0550439\left[\frac{1}{(q_{\max}\Lambda_2)^2+0.27819}\right.$$
$$\left.\left.-\frac{1}{(\overline{q}_{\min}\Lambda_2)^2+0.27819}\right]+0.274619\left[\frac{1}{(q_{\max}\Lambda_2)^2+3.99187}-\frac{1}{(\overline{q}_{\min}\Lambda_2)^2+3.99187}\right]\Bigg)\right\}, \tag{2.29}$$

where Λ_2 is given by Eq. (2.21) with N_1 and Z_1 substituted for N_2 and Z_2, respectively.

These expressions are straightforward to apply, and together with Eq. (2.23) for the number of electrons kept by the projectile as function of v, give the stopping cross section calculated from Z_1, N_2, and the known mean excitation energies ${}_iI_0$, obtained either theoretically or experimentally (e.g., [29]).

III. ANALYSIS AND DISCUSSION

A. Comparison with other theories

Let us analyze Eq. (2.11) in some limiting cases in order to relate this general theory to well-known special cases of Bethe theory for stopping. If we consider a bare projectile with $N_1=0$ and charge Z_1 colliding with a target with mean excitation energy ${}_2I_0$, then from Eq. (2.13) $\alpha=0$, and $\epsilon={}_2I_0$, so Eq. (2.11) becomes

$$S_e(v)=\frac{4\pi e^4}{m_e v^2}Z_1^2 N_2\ln\left(\frac{2m_e v^2}{{}_2I_0}\right), \tag{3.1}$$

which is the standard Bethe result in the first Born approximation. For the inverse case, namely, a projectile with N_1 bound electrons colliding with a completely ionized target ($N_2=0$), $\alpha=\infty$ and $\epsilon={}_1I_0$, and Eq. (2.11) becomes

$$S_e(v)=\frac{4\pi e^4}{m_e v^2}Z_2^2 N_1\ln\left(\frac{2m_e v^2}{{}_1I_0}\right). \tag{3.2}$$

This is, however, just the standard Bethe theory result obtained for a bare projectile of charge Z_2 incident on a target atom with N_1 electrons and mean excitation energy ${}_1I_0$. The same results are obtained from Eq. (2.28) and Eq. (2.29) under the same set of assumptions.

Next, let us consider the special case of no electronic excitations of the projectile, i.e., we only consider an elastic process on the projectile ($n=n_0$). Then, from Eq. (2.4) the contribution from the electronic structure of the projectile disappears and we obtain

$$S_e(v)=\frac{2e^4}{m_e v^2}\sum_m \int_{w_{m_0 m}/v}^{2m_e v/\hbar}[Z_1-{}_1M_{n_0n_0}(\mathbf{q})]^2\,{}_2F_{m_0m}(\mathbf{q})\frac{d\mathbf{q}}{q^2}, \tag{3.3}$$

where ${}_1M_{n_0n_0}$ is the atomic form factor of the projectile [Eq. (2.5)]. This equation has been applied in recent studies to the slowing-down process of ions with electronic structure [6]. In this limit, and applying the same approximations as the ones used to derive Eq. (2.11), we find

$$S_e(v)=\frac{2e^4}{m_e v^2}N_2\int_{2I_0/v\hbar}^{2m_e v/\hbar}[Z_1-{}_1M_{n_0n_0}(\mathbf{q})]^2\frac{d\mathbf{q}}{q^2}. \tag{3.4}$$

However,

$$\rho_{ne,1}(\mathbf{q})=e[Z_1-{}_1M_{n_0n_0}(\mathbf{q})] \tag{3.5}$$

is the Fourier transform of the total charge density (electronic+nuclear) of a projectile of nuclear charge Z_1 moving with N_1 bound electrons. Therefore,

$$S_e(v)=\frac{2e^2}{m_e v^2}N_2\int_{2I_0/v\hbar}^{2m_e v/\hbar}|\rho_{ne,1}(\mathbf{q})|^2\frac{d\mathbf{q}}{q^2}. \tag{3.6}$$

Noticing that an electron gas of density $n=n_2N_2$ has a plasma frequency ω_0 given by

$$\omega_0{}^2=\frac{4\pi n_2 e^2}{m_e}N_2, \tag{3.7}$$

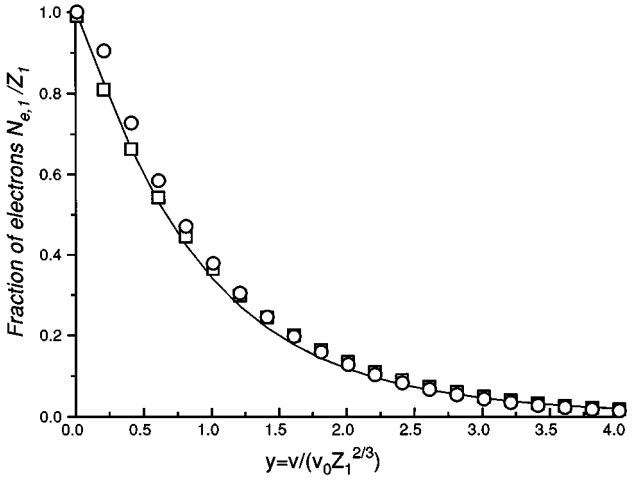

FIG. 1. Fraction of electrons remaining on the projectile N_1/Z_1 [see Eq. (2.23)] as a function of the velocity of the projectile. —, this work; ○, Anthony and Lanford [30]; □, Betz *et al.* [31].

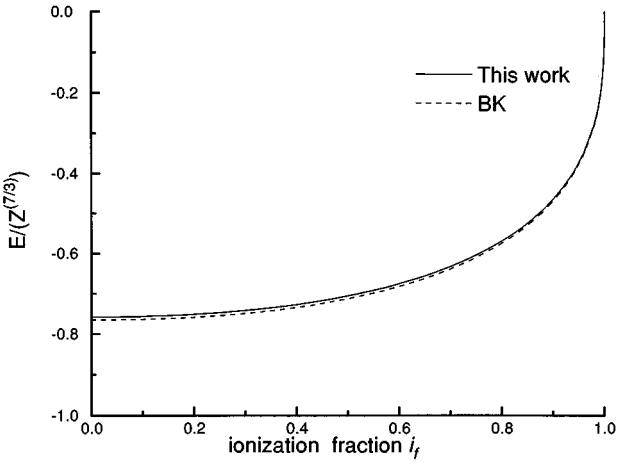

FIG. 2. Comparison of the total energy of the ion as a function of the ionization fraction ($i_f=1-N_1/Z_1$), calculated in the present model and in the Brandt-Kitagawa model [10]. —, this work; – – – –, BK model.

Eq. (3.6) may be written, for spherical symmetry, in the form

$$S_e(v)=\frac{{\omega_0}^2}{n_2 v^2}\int_{2I_0/v\hbar}^{2m_e v/\hbar}|\rho_{ne,1}(q)|^2\frac{dq}{q}. \tag{3.8}$$

This equation has precisely the same structure for the stopping power of solids as in a dielectric-response approximation [10] at high projectile velocity v.

B. The accuracy of the adiabatic assumption

In this section we will analyze the accuracy of the method suggested in Sec. II C for determining the number of electrons in the projectile as a function of the projectile velocity [see Eq. (2.23)] as well as the accuracy of the computed atomic form factors [Eq. (2.25)]. In Fig. 1 we show the fraction of electrons remaining with the projectile $[N_1(v)/Z_1]$ as a function of the effective ion velocity $y(v)=v/(v_0 Z_1^{2/3})$ compared with the experimental values of Anthony and Lanford [30] and of Betz *et al.* [31] for an Al target. It is a characteristic of the TF model that it does not represent the individual character of each atom (for example, shell structure, etc.), and Eq. (2.23) thus has a universal form when plotted as a function of the effective ion velocity. Also, we point out that the experimental data use the concept of ''effective charge'' [30], which is different from the mean charge of the ion [32] used here. This difference is noticeable in the low velocity region of Fig. 1 where the processes of electron capture and loss in the projectile as a function of the target ought to have been included in a more accurate way [32]. However, for higher projectile velocities, the Bohr criterion is more likely to be valid, and we find that the number of electrons remaining with the projectile is in close agreement with the experimental data.

We also find that our model for $\rho_{e,1}(r)$ performs well when compared with the Lenz-Jensen (LJ) [33] or the Brandt-Kitagawa (BK) models [10]. For example, as shown in Fig. 2, the energies [Eq. (2.20)] for different degrees of ionization, $i_f=1-N_1/Z_1$ calculated here and in the BK scheme are in close agreement. Thus, we are confident that calculation of N_1 with this approximation is sufficiently accurate for calculation of the stopping cross section for an ion with bound electrons.

In Fig. 3 we show the atomic form factor for several representative neutral atoms ($N_1=Z_1$) compared with the HF result of Hubbel *et al.* [34] and with the BK theory [10]. As we see, our method gives better overall agreement with HF than does the BK theory. This is probably due to the fact that our model is constrained to satisfy the TF model.

C. Comparison with experiment

The electronic stopping power calculated in this work includes the effect of transitions to all final states of the projectile and target that do not involve charge exchange. In order to compare to experimental data, we restrict our analysis to velocities high enough that the Bethe approximation is valid.

As examples, we calculate the stopping cross section for He, Li, and B projectile incidents on neutral C and Al targets

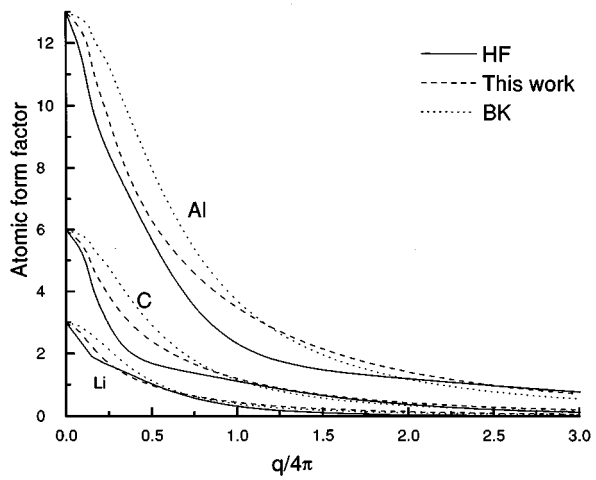

FIG. 3. Comparison of the form factor for neutral atoms as a function of the transfer moment q. —, HF calculations [34]; · · ·, BK model [10]; – – – –, this work.

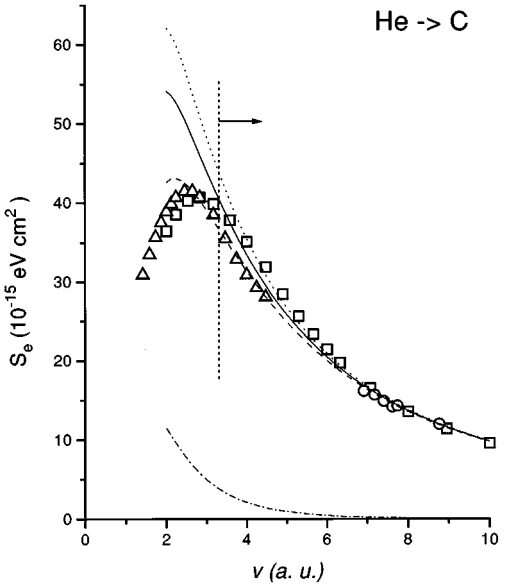

FIG. 4. Comparison of the computed electronic stopping cross section of He on neutral C with experimental results. – · – represents the contribution from the electronic structure of the projectile [Eq. (2.29)], – – – represents the contribution from the electronic structure of the target [Eq. (2.28)], — is the total contribution to the electronic stopping power, and ··· represents the normal Bethe logarithm term [Eq. (1.1)]. The ambit of applicability of the theory lies to the high velocities side of the vertical line. The symbols represent the experimental data from Ref. [35] (□), Ref. [36] (△), and Ref. [37] (○).

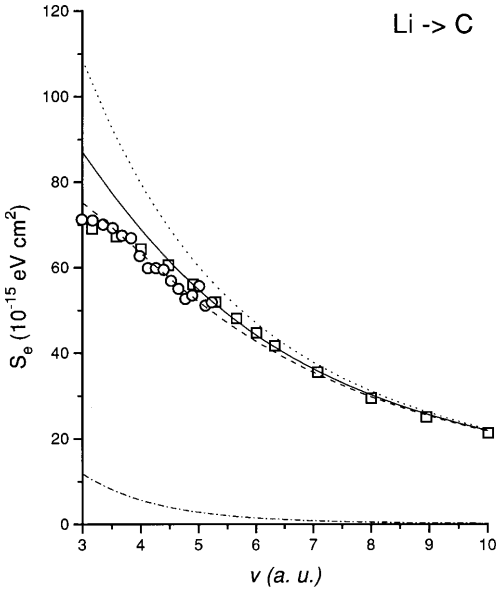

FIG. 5. Comparison of the computed electronic stopping cross section of Li on neutral C with experimental results. The legends are the same as in Fig. 4. The experimental data are taken from Refs. [35] (□) and [39] (○).

and compare to available experimental data [35–40]. The mean excitation energies of neutral C and Al, ${}_2I_0 = 73.8$ and ${}_2I_0 = 160.1$ eV, respectively, were taken from Janni's compilation [29]. The mean excitation energy for the projectile, as required by Eq. (2.12), is not needed in these calculations since the targets are neutral.

In Fig. 4 we compare the total stopping cross section for He incident on C with the experimental results of Northcliffe and Schilling [35] and of Santry and Werner [36,37]. For the region of high velocities (delineated by the vertical line) where this theory is valid, we find good agreement with experiment. In this case we have a light projectile colliding with a light target and the Bethe condition, $v > v_0$, is fulfilled.

In Fig. 5 we show the stopping cross section for Li projectiles incident on C, compared with the experimental data of Northcliffe and Schilling [35] and of Lin *et al.* [39]. Again, in the region of applicability of this theory, we find good agreement with experiment.

The comparison between calculation and experiment for Li on Al is made in Fig. 6 where we compare our theoretical result with the experimental results of Northcliffe and Schilling [35], Andersen *et al.* [38], and Lin *et al.* [39]. Similarly in Fig. 7, the calculated results for B on C are compared with the experimental results of Booth and Grant [40]. In all these cases, we see that for projectile velocities close to the maximum in $S_e(v)$, there is an overstimation of $S_e(v)$ for heavy projectiles on a heavy target. In these cases the velocities of the projectile and target electrons are high compared with the velocity of the projectile itself, hence we need consider corrections to the Bethe approximation for the low-velocity region. The most important corrections in this region are the shell correction and the Barkas correction. Both of these terms are negative and will bring our calculated result closer to the experimental result. The smaller Bloch correction has the opposite sign.

Also, in the same figures we plot the standard Bethe term [Eq. (1.1) for a bare projectile], and we show the behavior of contributions from both the projectile and the target. It is interesting to note the magnitude of the projectile contribution $S_{e,1}$ to S_e, of the order of 10–20%, in the lower part of

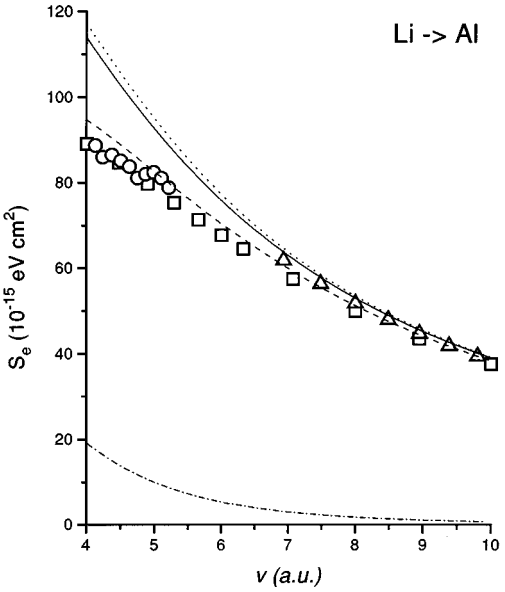

FIG. 6. Comparison of the computed electronic stopping cross section of Li on neutral Al with experimental results. The legends are the same as in Fig. 4. The experimental data are taken from Ref. [35] (□), Ref. [38] (△), and Ref. [39] (○).

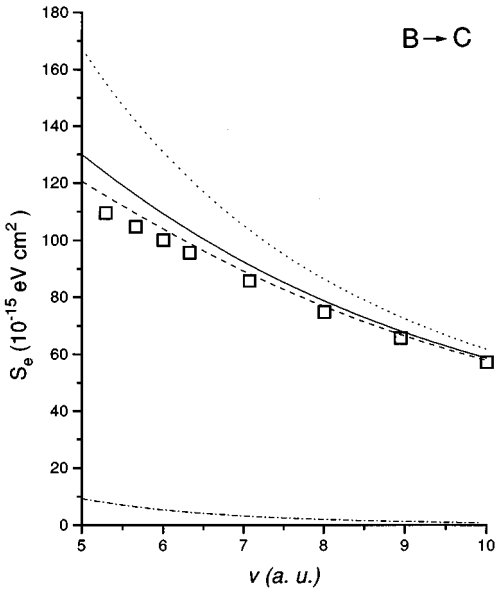

FIG. 7. Comparison of the computed electronic stopping cross section of B on neutral C with experimental results. The legends are the same as in Fig. 4. The experimental data are taken from Ref. [40] (□).

the velocity region where this model is applicable. This is observed in all the cases we have tested. This reflects the number of electrons bound to the projectile as a function of the velocity of the projectile (see Fig. 1), as $S_{e,1} \propto N_1$. This behavior has been found in other stopping theories at low velocities, such as that of Firsov [41], where the contribution to S_e is a sum of the contributions of the electronic structure of target plus the contribution of the electronic structure of the projectile [42].

IV. SUMMARY

We have derived a formula for the electronic stopping power of swift ions with $N_1(v)$ bound electrons colliding with an ionic target by considering the excitations and ionization of the bound electrons in both projectile and target. The formula follows from the same set of assumptions as involved in the normal Bethe theory of the stopping power in the first Born approximation. The general formulation requires knowledge of projectile and target GOS's. Due to the difficulties of evaluating these quantities, we have derived an approximation form of the general theory using TF atomic theory, thereby obtained analytic expressions for all the key quantities of the theory. The distribution of electrons in the projectile is calculated by making use of the Bohr criterion and the TF description for the atom, yielding an analytical expression for the ionization fraction i_f. The total stopping power is written as the contribution of the electronic structure of the projectile plus the contribution of the electronic structure of the target, and we found that the contribution of the projectile is important at intermediate velocities.

ACKNOWLEDGMENTS

One of the authors (R.C.T.) would like to thank the staff and faculty at the Kemisk Institute at the Odense University for providing him with such excellent working conditions. This work was supported by grants from the Danish Research Academy, the Danish Natural Science Research Council (Grants No. 11-0924 and 9313314 to J.O.), CONACyT-Mexico (Grant No. 1405-E9207 to S.A.C.), and the U.S. ARO (Grant No. DAA-H04-95-1-0326 to J.R.S.).

APPENDIX: LOWER LIMIT FOR THE Q INTEGRATION

We derive Eq. (2.12) by recalling that ϵ is defined in order to allow interchange of integration and summation in Eq. (2.4) viz., to ensure that

$$\sum_{n,m}\left\{\int_{q_{\min}}^{q_{\max}}|Z_2\delta_{m_0m}-{}_2M_{m_0m}(\mathbf{q})|^2\,{}_1F_{n_0n}(\mathbf{q})\frac{d\mathbf{q}}{q^2}+\int_{q_{\min}}^{q_{\max}}|Z_1\delta_{n_0n}-{}_1M_{n_0n}(\mathbf{q})|^2\,{}_2F_{m_0m}(\mathbf{q})\frac{d\mathbf{q}}{q^2}\right\}$$
$$=\left\{\int_{\epsilon/\hbar v}^{q_{\max}}\sum_{n,m}|Z_2\delta_{m_0m}-{}_2M_{m_0m}(\mathbf{q})|^2\,{}_1F_{n_0n}(\mathbf{q})\frac{d\mathbf{q}}{q^2}+\int_{\epsilon/\hbar v}^{q_{\max}}\sum_{n,m}|Z_1\delta_{n_0n}-{}_1M_{n_0n}(\mathbf{q})|^2\,{}_2F_{m_0m}(\mathbf{q})\frac{d\mathbf{q}}{q^2}\right\}.$$

This may be rewritten as

$$\sum_{n,m}\left\{\int_{\epsilon/\hbar v}^{(w_{n_0n}+w_{m_0m})/v}|Z_2\delta_{m_0m}-{}_2M_{m_0m}(\mathbf{q})|^2\,{}_1F_{n_0n}(\mathbf{q})\frac{d\mathbf{q}}{q^2}+\int_{\epsilon/\hbar v}^{(w_{n_0n}+w_{m_0m})/v}|Z_1\delta_{n_0n}-{}_1M_{n_0n}(\mathbf{q})|^2\,{}_2F_{m_0m}(\mathbf{q})\frac{d\mathbf{q}}{q^2}\right\}=0. \tag{A1}$$

Both limits of the integrals are small for velocities higher than the electronic velocities of projectile and target. Therefore, using the Bethe assumption ($v>v_e$) we expand Eq. (A1) for small momentum transfer. In the dipole approximation we obtain

$$\sum_{n,m}\left\{\int_{(w_{n_0n}+w_{m_0m})/v}^{\epsilon/\hbar v}(Z_1-N_1)^2\delta_{n_0n}\,{}_2f_{m_0m}\frac{d\mathbf{q}}{q^2}+\int_{(w_{n_0n}+w_{m_0m})/v}^{\epsilon/\hbar v}(Z_2-N_2)^2\delta_{m_0m}\,{}_1f_{n_0n}\frac{d\mathbf{q}}{q^2}\right\}=0, \tag{A2}$$

where

$${}_1f_{n_0n}=\frac{2m_ew_{n_0n}}{\hbar}\left|\left\langle n\left|\sum_i^{N_1}\hat{\mathbf{q}}\cdot\mathbf{x}_i\right|n_0\right\rangle\right|^2,$$

$${}_2f_{m_0m}=\frac{2m_ew_{m_0m}}{\hbar}\left|\left\langle m\left|\sum_i^{N_2}\hat{\mathbf{q}}\cdot\mathbf{y}_i\right|m_0\right\rangle\right|^2$$

are the optical dipole oscillator strengths and $\hat{\mathbf{q}}$ is a unit vector parallel to $\mathbf{q}$. The integration over q is now readily performed due to the independence of q in the dipole oscillator strength yielding

$$\sum_m (Z_1-N_1)^2{}_2 f_{m_0m}\ln\left(\frac{\epsilon}{\hbar w_{m_0m}}\right) + \sum_n (Z_2-N_2)^2{}_1 f_{n_0n}\ln\left(\frac{\epsilon}{\hbar w_{n_0n}}\right)=0. \quad \text{(A3)}$$

Using the standard definition of the mean excitation energy [19]

$$\ln_i I_0=\frac{1}{N_{e,i}}\sum_s f_{s_0s}\ln(\hbar w_{s_0s}), \quad i=1,2,$$

where the sum is over the excitated state, and inserting it into Eq. (A3) we obtain

$$N_2(Z_1-N_1)^2\ln\left(\frac{\epsilon}{{}_2I_0}\right)+N_1(Z_2-N_2)^2\ln\left(\frac{\epsilon}{{}_1I_0}\right)=0.$$

Defining

$$\alpha=\frac{(Z_2-N_2)^2N_1}{(Z_1-N_1)^2N)_2} \quad \text{(A4)}$$

and solving for ϵ results in

$$\epsilon={}_2I_0^{1/(1+\alpha)}{}_1I_0^{\alpha/(1+\alpha)}. \quad \text{(A5)}$$

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