

Impact-parameter dependence of electronic energy loss and straggling of incident bare ions on H and He atoms by using the coupled-channel method

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A first-principles calculation based on an expansion of the time-dependent electronic wave function in terms of atomic orbitals (coupled-channel method) has been applied to evaluate the impact-parameter dependence of the electronic energy loss and the fluctuation in energy loss of swift ions colliding on H and He atoms at energies of 10 to 500 keV/amu. The results have been compared with experimental data as well as with other existing models, e.g., the local-density approximation in an electron-gas target, the harmonic-oscillator target treatment, and the first-order plane-wave-Born approximation. Our results show a nearly exponential shape of the mean electronic energy loss for small impact parameters, in contrast to the Gaussian shapes obtained by Mikkelsen and Sigmund [Nucl. Instrum. Methods B 27, 266 (1987)].

I. INTRODUCTION

The phenomenon of energy loss of ions in matter has been studied for several decades. Although its overall properties are well known, there is no general theory that can accurately describe the energy loss in the whole energy range. Most of the existing models are based either on classical and semiclassical theories [1–3] or on first-order quantum perturbation theory [4–9], and for this reason their applicability is limited. However, only recently, some effort has been made to attack the stopping-power problem starting from first principles [10]. In this treatment, the electronic and nuclear energy losses of an incident bare ion and the corresponding stopping powers are obtained by solving the electronic time-dependent Schrödinger equation coupled with the classical motion of the nucleus.

Besides the stopping-power and energy-straggling information, in some cases it is necessary to go into more detail in the energy-loss phenomena. In particular, channeling experiments and measurements of energy loss as a function of scattering angle demand the knowledge of the impact-parameter dependence of the electronic energy loss. Moreover, the full impact-parameter dependence of the energy loss is a basic quantity to describe spatially correlated collisions that play an important role in structured targets, e.g., crystalline and solids and diatomic gases.

There are few descriptions of the average energy loss as a function of the impact parameter in the literature. Most of those models adopt either first-order perturbation theory [7–9] or local-density approximation in an electron-gas treatment of the target [11–15] or other approaches [2,16,17] in which it is hardly possible to determine the uncertainty due to the approximations involved.

In this work we will evaluate the average energy loss as a function of impact parameter $\bar{Q}(b)$ as well as the average of the square of energy loss $\overline{Q^2}(b)$ (related to the energy-loss straggling process) by using the improved

quantum calculation of Ref. [10] for very light systems (p, \bar{p} in H and He). The obtained $\overline{Q^2}(b)$ values will be used to compute the straggling in energy loss for hydrogen ions in H_2 and He considering the projectile charge states and the structure of the target, for the H_2 case. Some of the stopping-power calculations and a comparison with experimental data have already been discussed in a previous paper [10]. The same notation as in Ref. [10] is used in this work.

In the following, we shall give a brief description of the theoretical procedure used to evaluate $\bar{Q}(b)$ and $\overline{Q^2}(b)$. In Sec. III, numerical results are presented and discussed in connection with other energy-loss models. Finally, in Sec. IV, we provide a comparison of the calculated energy-loss straggling results with some existing experiments. If not indicated otherwise atomic unit (a.u.) will be used throughout the paper.

II. THEORY

It is well known that the impact parameter b , the asymptotic transversal distance of the incoming ion trajectory from the target particle, is a meaningful concept for heavy colliding particles ($m \geq m_{\text{proton}}$) even for thermal energies [1,18].

In this approach we assumed that the nuclear motion can be described by classical paths and the electronic motion is governed by a time-dependent Schrödinger equation given by

$$H(\{\mathbf{r}_e\}, \mathbf{R})\Phi_e = i \frac{\partial}{\partial t} \Phi_e \quad (1)$$

where $\{\mathbf{r}_e\}$ are the electron coordinates and \mathbf{R} is the internuclear distance which is assumed to be only a time-dependent parameter. $\mathbf{R}(t)$ is extracted from the classical trajectory calculated by using the averaged Hamiltonian for the heavy particles,

$$H_{\text{av}} \equiv \langle \Phi_e(t) | H(\{\mathbf{r}_e\}, \mathbf{R}) | \Phi_e(t) \rangle. \quad (2)$$

Then, for each impact parameter b , we have $\mathbf{R}=\mathbf{R}(t,b)$. However, for the cases considered so far we found no influence of the dynamic curved trajectory, given by Eq. (2), on the electronic energy loss. An influence on the nuclear energy loss at large impact parameter was found in Ref. [10]

The many-body Hamiltonian in Eq. (1) is treated in the framework of the independent electron model for an active electron, as described by McGuire and Weaver [19]. Of course, correlation effects are not taken into account. Furthermore, we will only analyze cases where the projectile is fully stripped. For these cases Eq. (1) can be solved quite well by expanding $\Phi_e(t)$ in terms of the eigenfunctions of the target without the projectile perturbation (single-center atomic wave functions φ_i). Then Eq. (1) can be replaced by a set of first-order coupled differential ordinary equations for the coefficients $a_i(t)=\langle\varphi_i|\Phi_e(t)\rangle$ originating from this expansion. These equations are so-called coupled-channel equations [20].

The essence of the present calculations is to solve numerically in time, step by step, Eq. (1) and the classical trajectory of the nucleus in order to obtain the coefficients a_i after the collision ($t=\infty$) since the probability of exciting (or ionizing) the active electron from the target in a collision with impact parameter b [$P_i(b)$] is given by

$$P_i(b)=\lim_{t\rightarrow\infty}|a_i(t)|^2. \quad (3)$$

The probability $P_i(b)$ should be highly accurate as long as electron capture by the projectile is of minor importance. The description of even a single projectile state requires an infinite number of single-centered target states as basis set, which is impossible to include in any numerical calculation. However, by choosing an appropriate basis set, with bound and continuum states of high orbital angular momenta, electron capture can be simulated for small internuclear distances. The advantage of this method, called AO (atomic-orbital expansion), is the good description of the time evolution of the electronic wave function at small internuclear distances, from where the projectile energy loss originates.

Each excited or continuum state corresponds to a well-defined energy transfer ΔE_i . Then the average electronic energy loss \bar{Q} in a given impact parameter b can be written as

$$\bar{Q}(b)=\sum_i P_i(b)\Delta E_i \quad (4)$$

and the mean squared of the transferred electronic energy (\bar{Q}^2) is given by

$$\bar{Q}^2=\sum_i P_i(b)\Delta E_i^2. \quad (5)$$

It is noted that the above sums have to be replaced by integrals in the case of continuum states.

The electronic stopping power S_e and energy straggling W per atom can be computed directly from

$$S_e=\sum_i \sigma_i \Delta E_i \quad (6a)$$

$$=2\pi \int_0^\infty b db \bar{Q}(b), \quad (6b)$$

$$W=\sum_i \sigma_i \Delta E_i^2 \quad (7a)$$

$$=2\pi \int_0^\infty b db \bar{Q}^2(b) \quad (7b)$$

where σ_i is the cross section for excitation (or ionization) from the ground state to a state i .

In this work we are only interested in electronic energy-loss processes. However, the kinetic energy transferred from the ion to the target core, the nuclear stopping process, can be obtained directly from the calculated classical trajectories.

We can also restrict the present computer code to the so-called semiclassical approximation (SCA) [7–9,18]. In this approach, the nuclear trajectories are assumed to be straight lines and the coupled-channel equations are solved by neglecting all matrix elements, except those which lead to transition from initial state, $1s$ or $1s^2$ in the He case, to one of the final states. This model yields the same cross section as the first-order plane-wave-born-approximation (PWBA) [21]. It is emphasized that the present SCA calculation is in disagreement with the one performed by Kabachnik, Kondratev, and Chumanova [9] for high-energy $H^+ + H$ collisions. The corresponding stopping cross section derived from Ref. [9] at 625 keV is about 20% less than the one predicted by PWBA. Therefore, in what follows, comparisons with first-order theory will be provided only through the present SCA code.

It was verified that the coupled-channel results agree with the predictions of the first-order perturbation theory (SCA) in the case of a small perturbation. Small perturbations correspond to either fast projectiles, large impact parameters, or small projectile charges. Thus the advantage of coupled-channel calculations compared to first-order theories show up especially at intermediate incident energies and for small impact parameters. However, the range of validity of the present single-centered atomic-orbital model should be $v_p/Z_p \geq 1$ due to the insufficient simulation of projectile-centered states, which dominate the collision process at low energies.

In the present work we have used a basis set with 10 gerade bound states and 80 gerade continuum wave packets [10]. The energies of the continuum states were chosen, depending on the projectile energy, from 0 to 500 eV and the largest angular momentum used was $l=7$. With this basis set, the estimated error for the probabilities (3) is less than 1% for 10 keV and roughly 0.01% for higher energies (e.g., 200 keV). For impact parameters smaller than 3 a.u., the accuracy of the calculation depends weakly on the selected impact parameter b . For larger b , our evaluation is much more precise since only dipole transitions are important. The computation time is less than one hour per impact parameter without counting the time required to generate all matrix elements (around 20 h).

Further details of the calculation, e.g., the numerical

treatment of continuum states and the adopted basis set, may be found in Ref. [10].

III. IMPACT-PARAMETER DEPENDENCE $\bar{Q}(b)$ AND $\bar{Q}^2(b)$

The numerical results calculated according to the preceding section are presented in Figs. 1–6. Figure 1 shows the energy dependence of $\bar{Q}(b)$ for protons incident on He. For impact parameter smaller than 2 a.u. the shape of $\bar{Q}(b)$ is approximately described by an exponential $e^{-\alpha b}$ function (dashed line). In fact, this behavior was found for almost all cases studied in this work. In addition we have found that the coefficient α depends only weakly on the projectile energy. For protons on H, $\alpha \approx 0.8$, and for p on He, $\alpha \approx 1.2$, for $E > 20$ keV. For lower energies, the energy loss $\bar{Q}(b)$ cannot be fitted with an exponential function anymore. The exponential behavior of the impact-parameter dependence of the electronic energy loss was already proposed, as an ansatz, by Oen and Robinson [22]. They have suggested that the α parameter is a function of interatomic screening length [3] and independent of the projectile energy.

From Fig. 1 we can also observe that the curves become flatter at large impact parameters with increasing ion energy. This is in harmony with results of first-order perturbation theories [23], where it is known that the mean impact parameter for excitation and ionization processes is proportional to the ion velocity v_p . It is emphasized that the contribution due to excitation becomes dominant at large impact parameters. We can also note that $\bar{Q}(0)$ follows the energy dependence of the electronic stopping power, showing a maximum around 80 keV [10].

In Fig. 2 the calculated impact-parameter dependence of the electronic energy loss for protons on H at 30 and 300 keV is displayed. The dashed lines represent a simple model where it is assumed that the energy loss is propor-

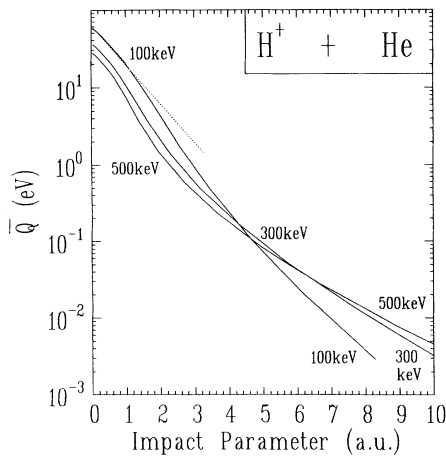


FIG. 1. Theoretical results of the mean electronic energy loss for proton incident on He at 100, 300, and 500 keV. Dashed line represents an exponential curve with $\alpha = 1.2$ (see text).

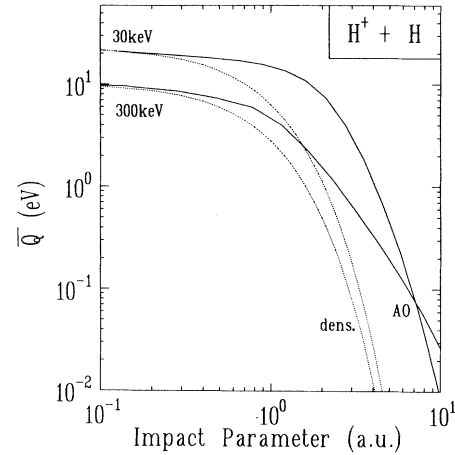


FIG. 2. Comparison between our theoretical result of $\bar{Q}(b)$ (AO) and the one obtained by assuming that the electronic energy loss is proportional to the electronic density integrated along the ion trajectory (dens.), for $H^+ + H$ collisions at 30 and 300 keV.

tional to the electronic density integrated along the ion path [24]. As can be seen from the figure, only for very small impact parameters ($b < 0.3$ a.u.) may the electronic energy-loss processes be considered as a local quantity. As a matter of fact, for larger impact parameters the energy loss $\bar{Q}(b)$ falls off very slowly. This tendency, indicating a breakdown of the local-density model, was observed for all cases (H and He targets). This result, which was already pointed out in Ref. [16], is very important, since most of the energy-loss models used in channeling experiments and theories are based on the assumption that the electronic stopping power is proportional to the local electron density [22,24].

Figure 3 displays the present calculation for 100-keV $H^+ + H$ collisions in comparison with other $\bar{Q}(b)$ models existing in the literature. The models (O-1) and (O-2) correspond, respectively, to the first- and second-order quantum perturbation theory in a harmonic-oscillator target, as proposed by Mikkelsen and Sigmund [16,17]. In this model, they have obtained, for not too large impact parameters, a near Gaussian dependence of $\bar{Q}(b)$ in contrast to the exponential shape found by us. In spite of this, the (O-2) model predicts quite good energy-loss values at impact-parameter zero. In this figure, $\bar{Q}(b)$ is also presented as evaluated by using a local-density electron-gas treatment [15]. The agreement with all other calculations is bad. However, for the present cases, the applicability of such a model is rather dubious.

In Fig. 4 the same calculation is shown as in Fig. 3 for 300-keV $H^+ + He$. The (O-2) calculation was performed with two harmonic oscillators with the same frequency $\omega = 1.54$ a.u. [17]. In this figure an experimental point from Ref. [25] is also presented for $b = 0.02$ a.u. The agreement between the experimental value and the calculated one is reasonable. However, a much better agreement can be achieved by increasing the size of the basis set used or adapting the basis set to the physical situation

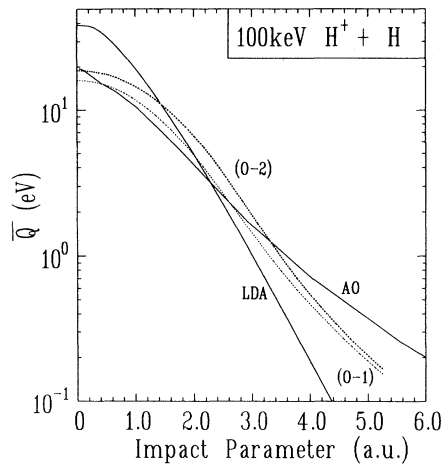


FIG. 3. Theoretical mean energy loss as a function of impact parameter for 100-keV $H^+ + H$ collisions. Present theory is denoted by (AO). Dashed lines (O-1) and (O-2) correspond to a calculation of $\bar{Q}(b)$ in a harmonic-oscillator target, with $\omega=0.55$ a.u., from Refs. [16] and [17]. The solid line (LDA) represents the evaluation by using local-density approximation in an electron-gas target [15]. $H(1s)$ ground-state density was used.

[10,25]. For small impact parameters, e.g., only states with angular momentum projection numbers $m=0$ need to be considered. It should also be pointed out that double-ionization processes become important for a He target mainly for very small impact parameters and the present calculations cannot properly take into account these processes because of the independent electron model which was employed in the calculations. Again we can

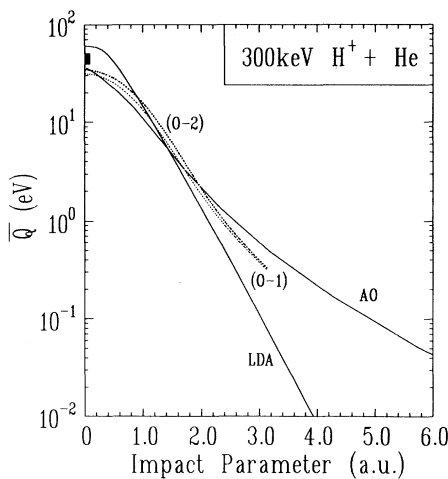


FIG. 4. The same as in Fig. 3 for 300-keV $H^+ + He$ collisions. The experimental point in $b=0.02$ was taken from Ref. [25]. Calculations (O-1) and (O-2) were performed by assuming two harmonic oscillators with $\omega=1.54$ a.u. [17]. LDA was calculated by using the $He(1s^2)$ ground-state density.

observe from this figure that the local-density models, i.e., theories based on a local behavior of the energy loss, cannot be applied for large impact parameters.

The impact-parameter dependence of the mean square of the electronic energy loss for protons and antiprotons on H at 10 and 30 keV is shown in Fig. 5. These values are compared with first-order SCA results as described in Sec. II. In a first-order calculation it is known that the excitation and ionization probabilities are proportional to Z_p^2 , the squared projectile charge. Therefore it predicts the same behavior for incident p and \bar{p} . However, as can be seen from the full $\bar{Q}^2(b)$ curves calculated using the coupled-channel method, they are quite different. For large-impact parameters this difference comes from the polarization effect, which is not included in the SCA calculation. The projectile can attract or repel the electrons, depending on its sign of charge. This implies an enhancement or reduction of the electronic density around the projectile trajectory and consequently, the of electronic energy loss. For small impact parameters the presence of the projectile charge may increase or decrease the electron binding energy. This can enhance or reduce the probability to excite or ionize the target electron [see Fig. 5(b)].

In the case of 10-keV protons, Fig. 5(a) shows a remarkable peak around $b=2$ a.u. This is due to resonant electron capture by the projectile. For low velocities v_p , it is well known there is an oscillatory behavior of the capture process probability as a function of $1/v_p$ (proportional to the collision time). This can easily be seen from a quasimolecular description, where this oscillation is re-

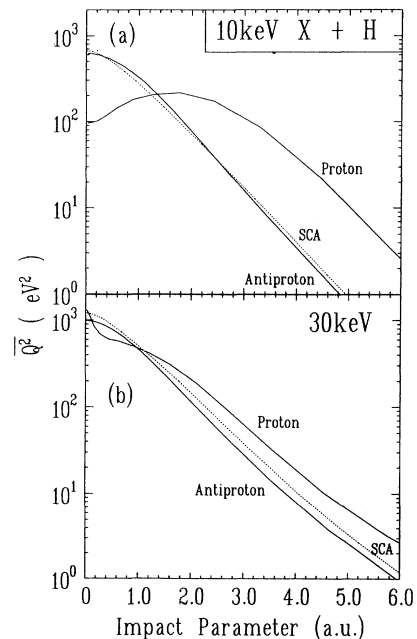


FIG. 5. Mean square of the electronic energy loss as a function of impact parameter. Solid lines represent the present calculation for p and \bar{p} on H at 10 and 30 keV. Dashed lines are the first-order perturbation theory SCA.

lated to the energy difference between the gerade and ungerade molecular states formed during the collision. Then, for small impact parameters $b \ll 2$ (short collision time, $\Delta t \approx b/v$), the electron of the target remains basically at the same initial state since the evolution time of the electronic wave function is very small. On the other hand, for $b \gg 2$, the interaction time is large enough to "pull out" the electron from the target via capture process. For this reason the proton electronic energy loss is very high when compared with the SCA calculation, which describes only excitation and ionization mechanisms. For the antiproton case, since there are no projectile-centered bound states, the shape of the electronic energy loss is similar to the one calculated in first-order perturbation theory SCA.

The computation of $\overline{Q^2}(b)$ [Eq. (5)] requires a basis set with high-energy components, i.e., eigenfunctions for large ionization energies. For example, for 50 keV $H^+ + H$ with $b = 0.218$ a.u., the basis set was truncated at $\epsilon = 400$ eV, whereas the mean energy loss is 21 eV and the root mean square of energy loss is 34 eV. Therefore the calculation of the mean energy loss is more accurate than $\overline{Q^2}(b)$. We estimate an uncertainty of 10–15% for the coupled-channel $\overline{Q^2}(b)$ results at incident energies below 100 keV.

Figure 6 shows the average of the squared and the square of the mean electronic energy loss as a function of the impact parameter for 10 and 100 keV $H^+ + H$. The mean energy-loss fluctuation, for a fixed impact parameter, is just $\Delta Q^2(b) = \overline{Q^2}(b) - \overline{Q}^2(b)$. In general, we have $\overline{Q^2}(b) \gg \overline{Q}^2(b)$ so that the fluctuation, for a given impact

parameter b , is basically determined by $\overline{Q^2}(b)$. However, for low energies ($E \leq 10$ keV) the fluctuation falls down very drastically around $b = 2$ a.u. [Fig. 6(a)]. This means that the excitation and ionization probability behaves like a δ function. As a matter of fact, this is a signature of resonant electron capture, which is very important at these energies [10].

IV. STRAGGLING CALCULATION AND COMPARISON

The fluctuation in energy loss of a monoenergetic incident beam passing through matter is not only due to the $\Delta Q^2(b)$, calculated in the preceding section, but also due to the statistical fluctuation in the number of collisions suffered by the penetrating ion. Assuming statistical independence of the collision events, the variance of the energy-loss distribution (Ω^2) can be calculated, for a thin penetrated layer δx , as [1]

$$\Omega^2 = N \delta x 2\pi \int_0^\infty b db [\overline{\Delta Q^2}(b) + \overline{Q}^2(b)], \quad (8a)$$

$$\Omega^2 = N \delta x W \quad (8b)$$

where N is the target density (atm/cm³) and W is given by Eq. (7b). The last term in Eq. (8a) arises after assuming Poisson statistics to describe the fluctuations in the number of ion-target collisions.

For an extremely thin penetrated layer, the energy-loss spectrum will exhibit a single collision energy-loss structure. On the other hand, if many more collisions happen, the energy-loss distribution will be broadened. If additionally the mean energy loss is much less than the initial projectile energy, the final energy-loss distribution will tend to a Gaussian profile [1].

According to Bohr [1], the straggling of a high-velocity particle with atomic number Z_1 , penetrating matter with atomic number Z_2 , is given by

$$\Omega_B^2 = N \delta x 4\pi Z_1^2 Z_2, \quad (9)$$

which is independent of the ion velocity v_p .

For not too high projectile energies, Fano evaluated the asymptotic straggling using quantum-mechanical perturbation theory [26]. The following expression was derived by Fano and corrected later [27]:

$$\frac{\Omega_{\text{Fano}}^2}{\Omega_B^2} = 1 + \frac{(1/Z_2)S(1)}{v_p^2} \left[\ln \frac{2v_p^2}{I_1} - \frac{3}{2} \right] \quad (10)$$

where I_1 and $(1/Z_2)S(1)$ are given in terms of the atomic dipole oscillator strength f_{0i} related to the transition from the ground state (E_0) to an excited state (E_i). For H targets we can take $(1/Z_2)S(1) = 0.67$ a.u. and $I_1 = 0.88$ a.u. [33]. Both formulas (9) and (10) assume a fully stripped projectile.

In Fig. 7 we show Ω_B^2 , Ω_{Fano}^2 , for p in H in comparison to straggling values (Ω_{AO}^2) calculated to the procedure described in Sec. II [Eqs. (7b) and (8a)]. In addition we have included the straggling results obtained by the plane-wave-Born approximation (Ω_{PWBA}^2) [16]. The difference between Ω_{Fano}^2 and Ω_{PWBA}^2 is attributed to the

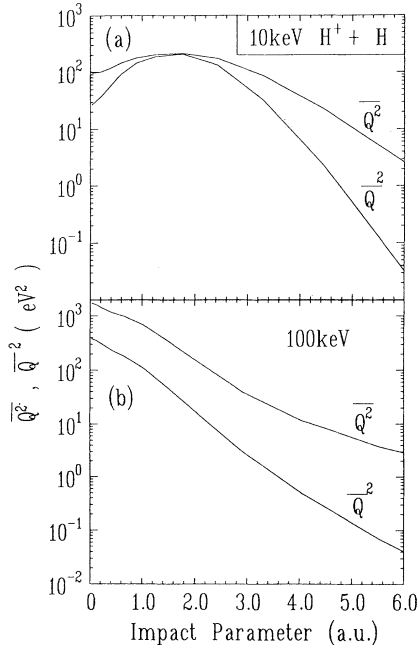


FIG. 6. Comparison between the squared and the square of the mean electronic energy loss for $H^+ + H$ collisions at 10 and 100 keV. For 10 keV the two curves do not cross over.

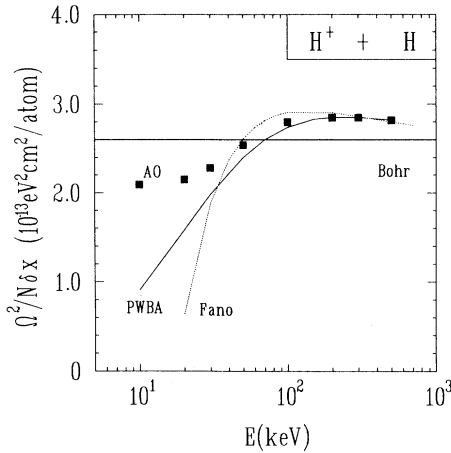


FIG. 7. Theoretical straggling values for H^+ charge state in collisions with hydrogen atoms. Present theory (AO) is indicated by squares. Both solid and dashed lines represent the straggling in energy loss Ω_{Bohr} and Ω_{Fano} from Eqs. (9) and (10), respectively. The solid line PWBA corresponds to the straggling results obtained by plane-wave-Born approximation [21].

dipole approximation used to obtain Eq. (10). For energies higher than 200 keV, the quantum results give basically the same result. The full calculation AO, as expected, converges to the first-Born results for energies $E > 200$ keV.

The existing measurements of straggling in energy loss of H ions in gases were performed in an energy range where there are two equilibrated charge states of the projectile H^+ and H^0 [28]. In order to supply a comparison with these experiments we also have to evaluate the straggling for a neutral incident hydrogen. For this case we can calculate the excitation and ionization cross section within the PWBA, since the H^0 projectile corresponds to a relatively small perturbation of the target. With the cross sections, the straggling can easily be computed through Eq. (7a). Then, an averaged straggling can be evaluated as

$$\Omega^2 = F_1 \Omega_1^2 + F_0 \Omega_0^2 \quad (11)$$

where Ω_1^2 , Ω_0^2 are the straggling in energy loss for penetrating p and H^0 , respectively, and F_0 , F_1 are the equilibrium charge-state fraction for hydrogen beams. These values may be found for H_2 and He target in the review of Allison [28].

Special attention must be drawn to the molecular gas target. According to Sigmund [29], the molecular geometry influences the spectral distribution of energy loss, deflection angles, and excitation phenomena. From a purely geometrical point of view, disregarding the differences between the electronic states of the constituent atoms in a molecule and in a free atom, Sigmund ob-

tained an additional positive-definite term to the expression (8a) for binary molecules:

$$\Omega_M^2 = N \delta x \left\langle \int d^2 b \overline{Q_1}(b_1) \overline{Q_2}(b_2) \right\rangle_\Omega \quad (12)$$

where the $\langle \rangle_\Omega$ means an average over all possible orientations of target molecules, $\overline{Q_i}$ is the mean energy loss in a collision with target atom i of the molecule, and b_i is the impact parameter relative to the i atom. For a homonuclear molecule, separated by a distance d , this expression can be written as [29]

$$\Omega_M^2 = N \delta x \frac{1}{2\pi d} \int_0^\infty dk \sin(kd) \Xi^2(k) \quad (13)$$

with

$$\Xi(k) \equiv \int_0^\infty 2\pi b db \overline{Q(b)} J_0(kb) \quad (14)$$

where J_0 is the zero-order Bessel function of the first kind.

It is now possible to perform an accurate computation of the molecular correlation term (12). We have calculated this term numerically using $\overline{Q(b)}$ from Eq. (4) and analytically assuming an exponential dependence of $\overline{Q(b)}$. Both methods give to within 20% the same result for Ω_M^2 , which shows that the exponential description of $\overline{Q(b)}$ is quite good (at least for a description of the molecular correlation term). By using $\overline{Q(b)} = (S/2\pi)\alpha^2 e^{-\alpha b}$, Ω_M^2 can be written as

$$\Omega_M^2 = N \delta x \frac{S^2}{2\pi d^2} g_{\text{exp}}(d\alpha) \quad (15)$$

where S is the electronic stopping cross section per atom [3,6] and $g_{\text{exp}}(x)$ ($x = d\alpha$) is shown in Fig. 8.

In Fig. 8 there is also $g_{\text{Gauss}}(x)$ obtained by assuming a Gaussian dependence $\overline{Q(b)} = (S/\pi)\alpha_{\text{Gauss}}^2 e^{-\alpha_{\text{Gauss}}^2 b^2}$ as proposed by Sigmund in Ref. [30]. Both curves have the same asymptotic limits [29], $\lim_{x \rightarrow \infty} g_{\text{exp, Gauss}}(x) = 1$,

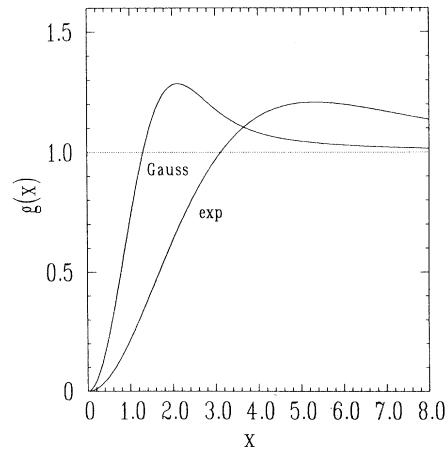


FIG. 8. Function $g(x)$, $x = d\alpha$ [Eq. (15)], which corresponds to the magnitude of the molecular correlation effect in the electronic straggling calculation.

however, they are different for realistic x values ($\alpha d \approx 1$, $d = 1.4$ a.u.) If we assume $\alpha_{\text{Gauss}} = \alpha/\sqrt{2}$ to make $\bar{Q}_{\text{Gauss}}(0) = \bar{Q}_{\text{exp}}(0)$ as shown in Figs. 3, and 4, the Ω_M^2 results calculated using an exponential and a Gaussian $\bar{Q}(b)$ function will be different by a factor of 2. Furthermore, Sigmund has proposed $g(\alpha d) \approx 1$, so his Ω_M^2 values, at least for the case $\text{H} \rightarrow \text{H}_2$, overestimate the effect by a factor of 4.

In the PWBA there is no information about the impact-parameter dependence of any quantity. Therefore it was not possible to calculate correctly the molecular correlation term for a neutral projectile. To overcome this situation we assumed an exponential dependence of the average energy loss for neutral H with the same coefficient α extracted from the p projectile cases. This is not strictly correct because we expect $\bar{Q}(b)$ for neutral projectiles to be of shorter range than in the case of bare ions. However, this approximation introduces only a very small error ($\ll 5\%$) because in the cases where the neutral component in Eq. (11) is dominant, the S values in (15) become very small.

We have also estimated the straggling produced by charge-state fluctuation according to Ref. [31]. But for the present cases this effect is of minor importance.

Finally, the total straggling in energy loss for H in H_2 and He gases can be obtained through Eqs. (7b), (11), and (15) and compared with experimental values from Refs. [32] and [33].

Figures 9 and 10 show the present straggling calculation in comparison with experimental data. The dashed lines correspond to the evaluation without the molecular term (12). For the He target, it is also necessary to evaluate the molecular correlation term for $d \rightarrow 0$ in (12) since we are assuming, according to the independent electron

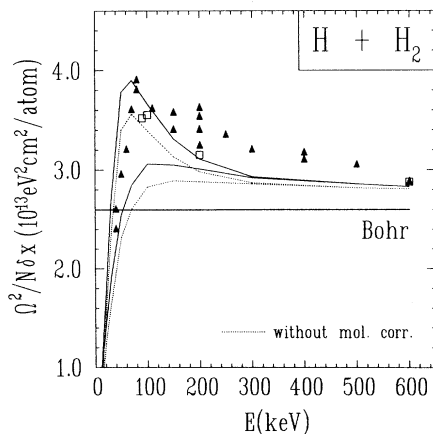


FIG. 9. Equilibrium mean total electronic straggling in energy loss per atom for incident H in H_2 target in comparison with experimental data from Refs. [32] (square) and [33] (triangle). Solid curves represent the calculation with the molecular term correction from Eq. (15). The difference between the two upper and lower curves is due to the treatment used to evaluate, in PWBA, the excitation and ionization cross sections for $\text{H}^0 + \text{H}^0$ collisions (see text).

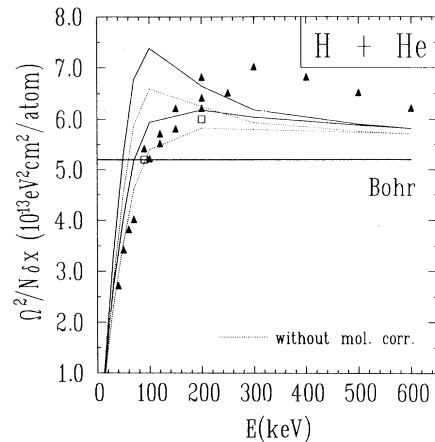


FIG. 10. The same as in Fig. 9 for incident H in He gas target.

model, the He target as being two full overlapping “effective one-electron atoms” each having the same one-electron wave function. It should be pointed out that for the first time this “bunching effect,” which was originally calculated by Besenbacher, Andersen, and Bonderup [31], is evaluated without using the electron-gas model [31,33].

The difference between the two upper and lower curves is due to the treatment used to obtain Ω_0^2 in (11). In a PWBA calculation, we can assume the neutral projectile to act as a screened Coulomb potential (lower curves), or we can take the dynamic motion of the projectile electron into account [34]. This implies, especially for small impact parameters, a so-called antiscreening effect (upper curves). Nevertheless, the treatment described in Ref. [34] usually overestimates the excitation and ionization cross sections. Therefore we expect that an exact two-electron treatment will yield a curve between the ones plotted as solid lines.

As can be observed from Fig. 9, for $E < 200$ keV the experimental points lie between the calculated values showing consistence of the theoretical results. But for the energy range between 200 and 600 keV, the theoretical predictions underestimate the experimental data by $\approx 10\%$. For these energies, there are basically $\text{H}^+ \rightarrow \text{H}_2$ reactions and the molecular correlation term (12) is negligible. In principle, we did not find any reasonable explanation for these discrepancies. However, as can be seen from the scatter of the data points at $E = 200$ keV, the experimental error seems to be larger than 5–7%, as quoted in Ref. [33].

In Ref. [33] the authors have claimed a very good agreement between their experimental values and the ones obtained by Fano [26] for H in H. This agreement was accidental since they have used the original Fano formula (10) which is uncertain by a factor 1.5 as noted by Sigmund and Haagerup in Ref. [27].

The same systematic deviation from the theory, for $E > 300$ keV, can be seen in Fig. 10, for the He target. In this case, the discrepancies are still higher (about 15%).

For lower energies, the agreement is also not good. However, in this energy range, the dominant contribution to the electronic straggling comes from $H^0 + He$ collisions for which we have computed the excitation and ionization cross sections in the PWBA. Besides the problems which may emerge from the description of projectile ionization, we have used hydrogenlike wave functions to describe the target excitation and ionization processes. It is well known that these functions deviate significantly from more sophisticated He target wave functions. It is emphasized that the solid curves in Fig. 10, i.e., "molecular correlation," stand for an independent particle treatment of double ionization. The double-ionization probabilities are known to be overestimated by a factor of 6 in the independent particle model [25]. However, it is not possible to determine from the existing experimental data the uncertainties due to approximations involved in our treatment.

V. CONCLUSIONS

In the present work we have presented for the first time a coupled-channel calculation of the mean and mean square energy loss as a function of the impact parameter for bare ions colliding with H and He atoms. We have obtained a near exponential shape of $\bar{Q}(b)$ for small impact parameters, in analogy to the one suggested by Oen and Robinson [22].

Comparisons with previous theoretical results have shown that the model (O-2), based on second-order quantum perturbation theory in a harmonic-oscillator target,

is more accurate than any other model based on local-density approximation. The (O-2) model reproduces quite well the mean energy loss at zero impact parameter but predicts a different behavior of $\bar{Q}(b)$ for nonzero b values.

The effect of a molecular target structure in the energy-straggling process was also exactly computed by using $\bar{Q}(b)$ calculated with the coupled-channel method. For the present cases, this effect is very small if we compare with the one proposed by Sigmund [30] (by a factor of 4).

By considering the charge states of the projectile and the target structure we have utilized the mean square energy transfer from the coupled-channel and PWBA calculations to evaluate the straggling in energy loss for hydrogen atoms in H_2 and He gases. The theoretical straggling values are in relatively good agreement (to within 10%) with the experimental ones. However, there is a need for more experimental data with improved accuracy.

This work shows the possibility of performing first-principles (coupled-channel) calculations of the energy-loss phenomenon, which until now was very scarce in the stopping-power literature.

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