Distorted-wave calculation of stopping powers for light ions traversing H targets

Pablo D. Fainstein and Victor H. Ponce
Centro Atómico Bariloche and Instituto Balseiro, Av. E. Bustillo 9500, 8400 Bariloche, Argentina

Alejandra E. Martinez*
Instituto de Física Rosario and Departamento de Física, Escuela de Ciencias Exactas y Naturales, Facultad de Ciencias Exactas, Ingeniería y Agrimensura, Universidad Nacional de Rosario, Av. Pellegrini 250, 2000 Rosario, Argentina
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The continuum-distorted-wave–eikonal-initial-state model is applied to the calculation of charged charge-state electronic stopping powers. The contributions from electron capture, excitation, and ionization are evaluated for $\bar{p}$, H^+, He^{2+}, and Li^{3+} projectiles impinging on H targets at energies of 10–1000 keV/amu. With the neutral H charge-state electronic stopping power obtained from the first Born approximation, charge-state fractions and total mean electronic stopping powers for H^+ beams traversing H are calculated. Present results are compared with calculations using the first Born approximation, a one-center atomic-orbital coupled-channel code, and experimental data for H_2 targets.

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I. INTRODUCTION

One of the oldest problems in quantum collision theory is that of the calculation of the energy loss of ions traversing dense matter. For gaseous targets, calculations were performed by Bethe [1] in the first Born approximation (B1). Using a multipole expansion of the perturbation, and keeping only the dipole term, he was also able to obtain closed formulas of the cross sections for single excitation and ionization by bare ion impact. These two reactions are the most important mechanisms for energy loss at high energies. As a consequence, the Bethe formula gives the high-energy behavior of the stopping power. As the energy decreases, other reaction channels become important and the contribution from different charge states must be included. These calculations can also be done within the first Born approximation. This task was undertaken by Bates and Griffing [2,3] during the 1950s. In pioneering works, performed without computers, total cross sections for single ionization and excitation of H by H^+ and H impact were calculated between 10 keV and 3.5 MeV. For the latter case the contributions from double transitions were also calculated without using closure [3]. It was shown that these reactions, where the neutral projectile is excited or ionized simultaneously with the target, give important contributions to the cross sections. These results were later shown to be in good agreement with experimental data above 50 keV [4,5]. Dalgarno and Griffing [6], using the charge-state approach, applied the first Born approximation to calculate the stopping power and the charge-state fractions of protons in H targets between 10 keV and 3.5 MeV. Their results show the typical behavior of first Born calculations; they give good agreement for high energies, while at intermediate and low energies it overestimates the experimental results. For proton impact on H it is well known that the first Born approximation overestimates the total cross section for single ionization [7] and electron capture [8], which are the main mechanisms of energy loss for the H^+ charge state at intermediate and low energies, respectively. Another feature where it fails is in the projectile charge dependence of the stopping power. Experiments with antiprotons or highly charged ions are in disagreement with the Z_p^2 scaling law of B1, where Z_p is the projectile nuclear charge.

To improve the description of this process, Schiwietz [9] introduced a one-center coupled-channel calculation using atomic orbitals (AO). The main problem with the method is that it requires very large basis sets in order to describe the capture channel and a large number of pseudostates for the continuum in order to account for ionization. These limitations imply that the AO method will be valid at intermediate and high energies. An extension to lower energies can only be done in practice using two-center coupled-channel calculations [12,13] or even three-center ones [14]. This was recognized in a recent work [10], where AO is used for energies higher than 25 keV and a two-center coupled-channel atomic-orbital expansion (AO+) [13] is used for lower energies. In [10] the contribution from the neutral charge-state fraction was also calculated with AO but without explicit inclusion of double transitions.

An alternative approach is to use perturbation expansions based on the distorted-wave (DW) method. Calculations with approximations based on this method are much less expensive than the coupled-channel ones in computer time, and they have been shown to yield excellent agreement with experiments for ionization [7], excitation [15–17], and capture [18] in a broad range of impact energies starting at 10 keV. The main limitation is that coupling between the different channels is neglected. A coupled-channel calculation using two-center distorted-wave functions has not been attempted yet.

In the present work we will present calculations of the

stopping power of proton beams impinging on a H target. The charge-state approach will be used and the stopping power of the H^+ and H fractions will be calculated using distorted-wave models and first-order Born, respectively. For the neutral fraction double transitions are taken into account. Previous results using the one-center atomic-orbital expansion model [9] will be corrected to include these effects. Results from the distorted-wave models for the H^+ fraction will be compared with the coupled-channel and first Born calculations. The capture cross sections obtained with the distorted-wave model, and the loss cross section calculated with the first Born approximation are used to obtain charge-state fractions, which are compared with those of [6] and with experimental ones from [19]. The sum of the stopping power of H^+ and H averaged over the corresponding charge-state fraction will be compared with experimental data, first Born [6], and AO [9] calculations.

II. THEORETICAL MODELS

A. Distorted-wave models for charged projectiles

We are concerned with the calculation of the stopping power of protons impinging on H but the present results can be extended to more complex atomic targets. The only reactions that will give contributions to the energy loss are single excitation:

$$H^+ + H(1s) \rightarrow H^+ + H(nlm) , \quad (1)$$

where nlm indicate the quantum numbers of the final state; single ionization:

$$H^+ + H(1s) \rightarrow H^+ + H^+ + e^- ; \quad (2)$$

and single electron capture:

$$H^+ + H(1s) \rightarrow H(nlm) + H^+ . \quad (3)$$

The common feature of these three reactions is that the initial- and final-channel potentials are Coulomb potentials. This potential has an infinite range, so that the initial or final bound states are distorted even at infinite distances. For reaction (2) the situation is even more complicated because the ejected electron travels in the presence of two Coulomb potentials, and it is not possible to neglect either of them [7]. The distortion can be incorporated using the DW method. Usually the initial and final unperturbed states as defined by the first Born approximation are distorted using multiplicative factors that take into account part of the perturbation. These are the initial and final distorted waves. The remainder of the perturbation can be calculated from them and it is expected to be smaller than the full perturbation from B1. In this way the perturbation expansion will converge faster than the Born series and the first order will contain higher orders of the Born expansion. A full description of the method with application to reaction (3) can be found in [8] and its application to reaction (2) in [7].

For the initial state we will use what is known as the eikonal distortion. The multiplicative factor is chosen as an eikonal phase which represents the projectile-electron interaction at large distances. The main reason behind this election is that the distortion preserves the normalization of the initial state [7].

For the final state the distortion is chosen as the full Coulomb wave representing the projectile-electron interaction for reactions (1) and (2), and the target-electron interaction for reaction (3). The model that results from this approximation is the continuum-distorted-wave–eikonal-initial-state (CDW-EIS) [7]. For reaction (1) we will make a further approximation to the final state. The Coulomb wave will be approximated by its asymptotic form, i.e., an eikonal phase as used for the initial state. We make this approximation because for this reaction the electron remains bound and does not change center, so a symmetric model is more appropriate. This model is called the symmetric-eikonal (SE) approximation and the differential and total cross sections obtained from it are in very good agreement with experiments [15,16]. For asymmetric systems the CDW-EIS model may give better results [17].

With these distorted initial and final wave functions the transition amplitudes can be obtained in closed form. To calculate the stopping power in reactions (1) and (3) a one-dimensional numerical integration is necessary, while for reaction (2) the numerical integration is more cumbersome because it is necessary to perform an additional three-dimensional integration over the final electronic momentum. Although these calculations take more computer time than the corresponding ones using first Born, they can be easily done on a PC-486.

B. First Born calculations for neutral projectiles

For neutral hydrogen impact the incoming nucleus is screened by its electron. The perturbation is then of short range and weaker than in the case of a bare proton. There is no distortion of the initial bound state. The first Born approximation is then useful to treat this problem.

In this case there are several open channels because both collision partners can be excited and/or ionized. The final state of the incoming hydrogen atom defines the projectile-elastic and projectile-inelastic channels if it remains in the initial ground state or is excited to a bound or continuum state, respectively. The final state of the target defines the symmetric target-elastic and target-inelastic channels. The application of first Born to study these reactions was introduced by Bates and Griffing [2,3]. Two basic assumptions are made: (i) exchange is neglected, and (ii) when the projectile or target electron is ionized it is assumed to travel only in the field of its parent nucleus. The first approximation is always valid at high impact energies, while the second one is also correct because electrons are usually emitted with low velocities. In the first work [2] only the projectile-elastic channel was considered. In successive works [3] the contributions from all double transition reactions were summed to get the total contribution from the projectile-inelastic channel. This approach was also taken for the stopping-power calculations [6] and is the one we will take here. There are other authors that have used the closure relation to sum the contributions from the different channels. To do this it is necessary to suppose

that the minimum momentum transfer is the same for all channels [20] or to introduce a mean minimum momentum transfer [21]. This approach overestimates the total cross section [22]. A detailed analysis of the application of closure shows that corrections must be introduced [21], but the corrected closure method yields results which are similar to those from [3].

The projectile-elastic and target-elastic channels are single excitation:

$$\mathrm{H}(1s)+\mathrm{H}(1s)\rightarrow\mathrm{H}(1s)+\mathrm{H}(nlm)$$
$$\rightarrow\mathrm{H}(nlm)+\mathrm{H}(1s)\ , \tag{4}$$

and single ionization:

$$\mathrm{H}(1s)+\mathrm{H}(1s)\rightarrow\mathrm{H}(1s)+\mathrm{H}^{+}+e^{-}$$
$$\rightarrow\mathrm{H}^{+}+e^{-}+\mathrm{H}(1s)\ . \tag{5}$$

The projectile-inelastic and target-inelastic channels are double excitation:

$$\mathrm{H}(1s)+\mathrm{H}(1s)\rightarrow\mathrm{H}(n'l'm')+\mathrm{H}(nlm)\ , \tag{6}$$

with $n,n'\neq 1$; double ionization:

$$\mathrm{H}(1s)+\mathrm{H}(1s)\rightarrow\mathrm{H}^{+}+e^{-}+\mathrm{H}^{+}+e^{-}\ ; \tag{7}$$

and simultaneous excitation and ionization:

$$\mathrm{H}(1s)+\mathrm{H}(1s)\rightarrow\mathrm{H}(n'l'm')+\mathrm{H}^{+}+e^{-}$$
$$\rightarrow\mathrm{H}^{+}+e^{-}+\mathrm{H}(nlm)\ . \tag{8}$$

We will consider $n,n'\leq 3$, because contributions from higher excited states are negligible [3,6]. The formulas for the computation of the stopping power in reactions (5)–(9) are given in [6]. It must be noted that the contributions from the H^{-} charge-state fraction and from the fraction of neutrals in excited states are also neglected. While the first one is usually very small [19], the second one may give an important contribution. In this case the energy loss is less than for projectiles initially in the ground state but the cross sections are much higher [5].

III. RESULTS AND DISCUSSIONS

A. Electronic stopping power of protons on H targets

First of all we study the proton charge-state contribution to the electronic stopping power S_e^{+}. In all the calculations we neglect the contribution from the nuclear stopping power because it is very small in the range of impact energies that we study in the present work. S_e^{+} is calculated as described in Sec. II A from the cross sections and energy transfers ΔE_{ij} for the different channels

$$S_e^{+}=\sum_{f=1}^{\infty}(\Delta E_{if}^{E}\sigma_{if}^{E}+\Delta E_{if}^{C}\sigma_{if}^{C})+\int_0^{\infty}dE_k\,\Delta E_{ik}\frac{d\sigma_{ik}^{I}}{dE_k}\ , \tag{9}$$

where σ_{if}^{E} and σ_{if}^{C} are the total cross section for reactions (1) and (3), respectively. $d\sigma_{ik}^{I}/dE_k$ is the single differential cross section for ionization [reaction (2)] as a function of the final electron energy $E_k=(\hbar k)^2/2m$, with $\hbar\mathbf{k}$ the momentum and m the mass of the ejected electron. For the first term we use the SE approximation, while for the other two we use the CDW-EIS. It must be noted that the cross sections for each reaction are in excellent agreement with corresponding experiments. We have also compared our results for capture and ionization with calculations using different two-center coupled-channel models [12,13]. All theoretical results are in very good agreement at all impact energies considered here (from 10 keV/amu to 1 MeV/amu).

In Fig. 1 our present DW results for S_e^{+} are compared with first Born from [6] and coupled channel with atomic orbitals (AO) from [9]. At high energies our results agree with first Born and hence with Bethe's formula while AO gives lower values [9]. At intermediate and low energies the three models show different behaviors: distorted-wave values of S_e^{+} are lower than those from first Born, both reach a maximum at different impact energies and then decrease as the energy increases towards Bethe's limit. The close-coupling values of S_e^{+} always increase as the impact energy decreases and are lower than those from the other models at high energies. From these results one would expect that the AO and B1 calculation will overestimate the total mean energy loss because first Born always overestimates the total cross sections. This may not be the case, because the proton stopping power has to be weighted with its corresponding charge-state fraction. In the figure we also show these results, labeled AO*, B1*, and DW*, which show a similar behavior. Although the ratio between the results from these models does not change, the difference between them decreases by a factor equal to the charge-state fraction. From these results we can expect that the calculated total stopping power from the three models will be similar. As indicated in [9] the incorrect behavior of AO at intermediate en-

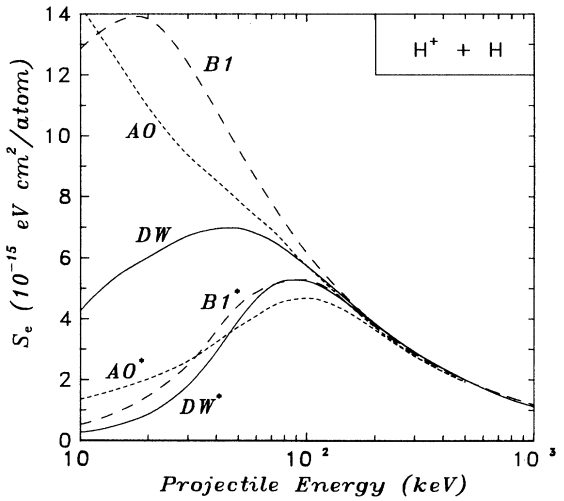

FIG. 1. Electronic stopping power for the H^{+} charge-state fraction as a function of projectile energy. Present DW calculations, solid line; one-center AO from [9], short dashed line; B1 from [6], dashed line. Present DW multiplied by calculated charge-state fraction (DW*), solid line; AO from [9] multiplied by experimental charge-state fraction [19] (AO*), short-dashed line; B1 multiplied by calculated charge-state fractions [6] (B1*), dashed line.

ergies arises from the truncation of the one-center expansion. This behavior was corrected in subsequent work by considering a two-center atomic-orbital expansion at energies lower than 25 keV [10].

B. Projectile-charge dependence of the electronic stopping power

First-order perturbative approximations like B1 for excitation or ionization, predict that the cross sections scale with the square of the projectile charge. Deviations with respect to this simple scaling are usually associated with higher orders of the Born series. Negative charged projectiles like antiprotons have received great attention because differences in the cross sections that depend on the sign of the projectile charge yield information about what are usually called Z_p^3 corrections. In the Born series they appear in the second order [23]. In the distorted-wave series they are already considered in the first order due to the correct treatment of the Coulomb potentials [7]. The close-coupling calculations contain, in principle, contributions from all orders of Z_p.

The projectile charge dependence of S_e^+ is studied by making the ratio with the result obtained from first Born, where only the contribution from single ionization and excitation are considered. These ratios are shown in Fig. 2 for different projectiles: protons, antiprotons, $^4He^{2+}$, and $^7Li^{3+}$. Also included are the ratios for protons and antiprotons calculated with the atomic-orbital expansion [9]. For antiprotons, $^4He^{2+}$ and $^7Li^{3+}$ impact, the close-coupling and distorted-wave calculations give the same behavior although the ratios calculated from DW are smaller. The distorted-wave calculations for capture will probably underestimate experiments because only final states with $n \leq 2$ are considered. If higher values of n were included the calculated stopping power would be higher and the ratios would increase bringing both theories in closer agreement. For proton impact the results are very different as could be expected from Fig. 1.

The different behavior of the AO ratios for proton impact as compared with the other projectiles was attributed to the possibility of resonant capture in the case of H^+ impact [9]. This is true if only the ground state is considered. For He^{2+} and Li^{3+} there are resonant conditions for final states with $n=2$ and $n=3$, respectively, while for antiprotons the capture channel is always closed. Also, there are other mechanisms that come into play for multiply charged ions. For example, first Born total capture cross section scale as Z_p^3 at intermediate energies [24] and as Z_p^5 at higher energies [25]. Hence, the different reaction channels have different behaviors as a function of the projectile charge. To see this we calculate the fractional contribution of each channel to the total charge-state stopping power as the ratio between each contribution with S_e^+. These ratios are plotted in Fig. 3. We see that at low energies the importance of capture *increases* while that of ionization and excitation *decrease* with the projectile charge. Capture remains very effective to the energy loss because there is resonant capture to highly excited states of the projectile and because the binding effect makes the ionization and excitation cross sections to fall down very quickly as the energy decreases [7].

Another feature of the ratios shown in Fig. 2 is that they are frequently used to search for high-order effects in the Born series. As the first order for excitation and ionization scale with the square of the projectile charge, all deviation in the ratios are related to terms in the Born series proportional to higher powers of Z_p. The second-order contribution of order Z_p^3 is called the Barkas correction and it accounts for the differences in the ranges of particles of opposite charge such as protons and antiprotons. Several experiments with antiprotons have been done in the last years to compare with theoretical

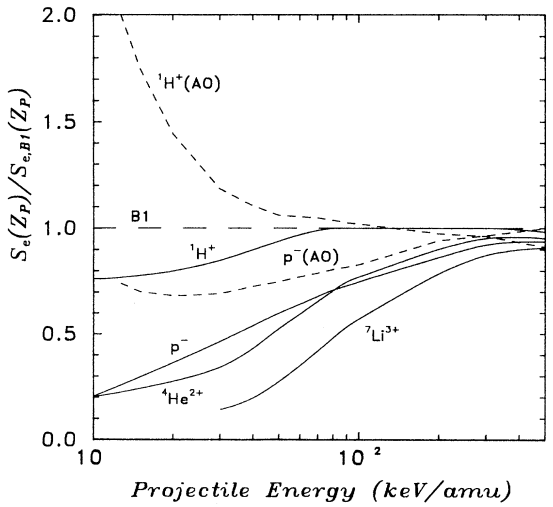

FIG. 2. Ratio of stopping powers, calculated with present DW model (solid line), and with one-center AO [9] (dashed line), with B1 calculations including only excitation and ionization channels as a function of projectile energy.

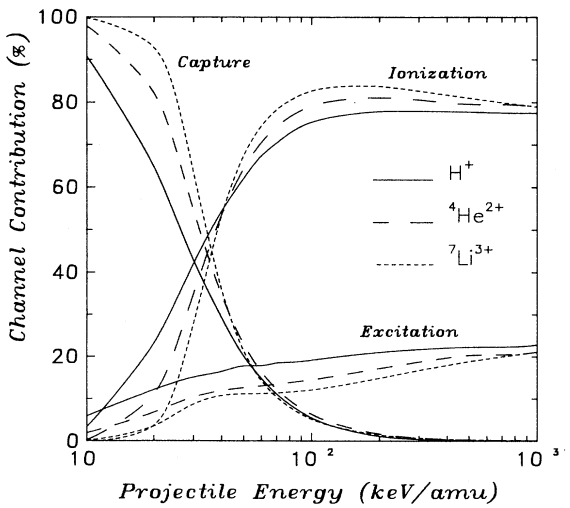

FIG. 3. Present DW calculations of the contribution from the different channels to S_e^+ for H^+ impact (solid line), $^4He^{2+}$ impact (dashed line), and $^7Li^{3+}$ impact (short dashed line) as a function of projectile energy.

predictions of the Barkas effect [26]. As we have mentioned above the first Born capture cross sections scale as Z_p^3 at intermediate energies. This indicates that a first Born calculation of S_e^+ will also have this behavior in this energy range. Then the ratios shown in Fig. 2 are not adequate to highlight higher-order effects in a perturbation expansion, because even the first order does not follow the simple Z_p^2 scaling law. The reason for this is that the expansion of the Born series in powers of the projectile charge is valid only for asymmetric systems where $Z_p \ll Z_t$ (with Z_t the target nuclear charge). This was pointed out by Basbas, Brandt, and Laubert [27] in their original treatment of these higher-order effects in inner-shell ionization of heavy targets by light projectiles. The projectile charge, which represents the strength of the perturbation, must be smaller than the target nuclear charge in order to make the first-order perturbation treatment valid. For these asymmetric systems capture gives a very small contribution and so the stopping power calculated with first Born will follow a Z_p^2 dependence. In this case the ratios can be used to obtain information about the Barkas effect and other high-order effects like binding and polarization [7,27]. For one-center coupled-channel calculations the deviations with respect to first Born represent not only the contributions from these mechanisms but also from capture which can be considered as a higher-order contribution in this formalism.

C. Equilibrium charge-state fractions

The present method of calculation, where the cross sections for all the open channels are known, allows to calculate the charge-state fractions. If the charged and neutral components of the beam are in equilibrium, they are given by [6]

$$f(\mathrm{H}^+)=\frac{\sigma^L}{\sigma^C+\sigma^L} \tag{10}$$

and

$$f(\mathrm{H}^0)=\frac{\sigma^C}{\sigma^C+\sigma^L}\ , \tag{11}$$

where σ^C is the total capture cross section and σ^L is the total loss cross section. The first is calculated with the CDW-EIS approximation and the second with first Born taking into account double transitions as given in [6]. Our present results are shown in Fig. 4 together with the results from [6] where first Born was used to calculate all the cross sections, and experiments for H_2 targets from [19]. At high energies the theoretical results agree with experiments. At low energies first Born and CDW-EIS results for (11) are higher than experiments due to the possibility of resonant electron capture in H targets. At intermediate energies the differences between theory and experiment remain and we see that the slope of the theoretical results is higher than the experimental ones. These differences are indications of the breakdown of Bragg's rule for H_2 targets as suggested in [6] and confirmed in loss experiments [4,5].

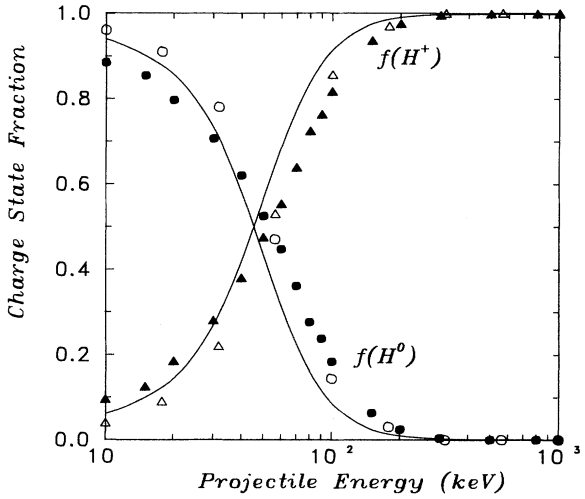

FIG. 4. Equilibrium charge-state fractions for proton beams traversing H as a function of projectile energy. Present DW calculation, solid line; B1 from [6], ○; experiments for H_2 targets [19], ●.

D. Total mean electronic stopping power of protons on H targets

From the stopping power for the charged fraction S_e^+ calculated with (9), the stopping power for the neutral fraction S_e^0 as given in [6] and the charge-state fractions calculated in the previous section we can calculate the total mean stopping power of the beam given by

$$\bar{S}_e = f(\mathrm{H}^+)S_e^+ + f(\mathrm{H}^0)S_e^0\ . \tag{12}$$

In Fig. 5 our present distorted-wave results are com-

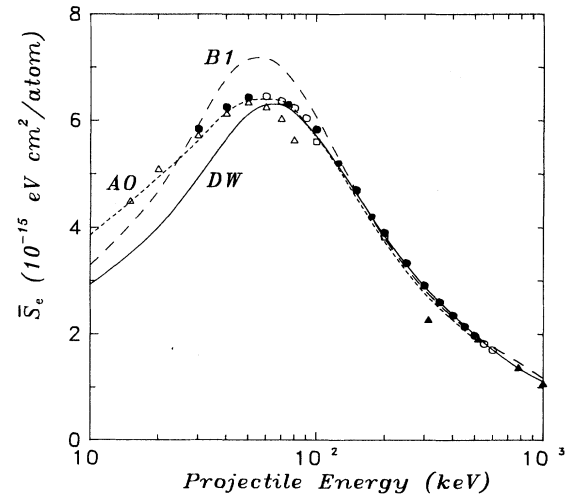

FIG. 5. Total mean electronic stopping power of proton beams on H targets as a function of projectile energy. Present DW calculations, solid line; B1 results from [6], dashed line; one-center AO results from [9] corrected for double transitions, short dashed line; experiments for H_2 targets, open triangles [28], closed circles [29], open circles [30], squares [31], closed triangles [32].

pared with full first Born and single-center atomic-orbital close-coupling calculations from [6] and [9], respectively. The AO results are obtained from (12) using the results from [9] for S_e^+ (shown in Fig. 1), while S_e^0 is taken from [6] as in our present distorted-wave calculations. However, in this case it is not possible to calculate the charge-state fractions because the contributions from different channels are not available. Experimental charge-state fractions from [19] for H_2 targets are then used. The main difference with previous results of the AO model [9] is that here we have included the contribution to S_e^0 from double transitions. It must be noted that our distorted-wave results do not change very much if the experimental charge-state fractions are used instead of the calculated ones. Experiments for the stopping of proton beams on H_2 targets [28–32] are included for comparison.

Above 70 keV the distorted-wave results are in excellent agreement with experiments. The same happens with first Born above 150 keV. In the region of the maximum first Born overestimates and distorted wave underestimates the experimental data, and at low energies both models go below the measurements. The single-center atomic-orbital results with explicit inclusion of double transitions are in excellent agreement with the experimental data over all the energy ranges. These results are also in very good agreement with recent calculations [10,11] where the AO model has been improved by using two-center atomic orbitals for the calculation of S_e^+ at low energies and a single-center atomic-orbital expansion for the calculation of S_e^0 instead of the first Born used here and in previous works [9]. The only difference between them appears at intermediate energies where the results from [10,11] are lower than the ones presented in Fig. 5.

The very good agreement with experiments at all impact energies considered here indicates that the AO model is very well suited to perform stopping-power calculations. Probably the new results where S_e^+ *and* S_e^0 are calculated with atomic-orbital expansions are more reliable than the ones presented here where S_e^0 is calculated with first Born corrected to account for double transitions. The better agreement with experiments of these calculations is a result of the fact that the one-center atomic-orbital calculation of S_e^+ is much higher than the distorted-wave and two-center atomic-orbital results. This difference remains after S_e^+ is multiplied by the charge-state fraction (see Fig. 1).

The distorted-wave results are also quite good. Some improvements are necessary for energies below 70 keV. As the cross sections for the different channels in the H^+ impact case are in very good agreement with experiments the problem arises in the first Born calculation of S_e^0. At low impact energies total cross sections calculated with this approximation for H impact on H are lower than the experimental data [4,5,10,33]. The first Born calculation of S_e^0 will underestimate this contribution. At intermediate energies where the stopping power reaches its maximum, the charge-state fraction for the neutral projectile calculated for an atomic target is lower than the experimental one for a molecular target. The inclusion of this *molecular* effect will also increase the calculated stopping power. An atomic-orbital calculation of S_e^0 together with the distorted-wave calculation of S_e^+ will probably be in much better agreement with experiments.

IV. CONCLUSIONS

We have used distorted-wave models to evaluate the stopping power of the charged fraction in collisions between protons and thick H targets. The neutral fraction is calculated with the first Born approximation. The cross sections obtained for the different reaction channels are used to calculate the charge-state fractions which are then used as weights for the calculation of the total mean energy loss. This full *ab initio* calculation is in excellent agreement with experiments at high energies and in good agreement at intermediate energies. Previous calculations with a one-center coupled-channel approach are in excellent agreement with experiments at intermediate and high energies when double transitions are considered in the loss cross sections. Calculations with the first Born approximation give good results only at high energies.

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*Present address: Centro Atómico Bariloche, Av. E. Bustillo 9500, 8400 Bariloche, Argentina.

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