Intercomparison of atomic models for computing stopping parameters from the Bethe theory: Atomic hydrogen

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The stopping cross section, the straggling parameter, and three higher moments over the energy-loss cross section have been evaluated for a moving point charge interacting with atomic hydrogen. The aim was to estimate the accuracy of predictions based on four commonly used models for computation of stopping parameters. The standard of reference was a straight evaluation of the Born approximation. All considered models, i.e., the dielectric theory, the binary-encounter model, the kinetic theory, and the harmonic-oscillator model, have been shown to be capable of providing reliable predictions within their respective regimes of feasibility, but distinct variations show up from lower to higher moments and from higher to lower projectile energy.

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I. INTRODUCTION

Bethe [1] established the quantum theory of stopping for a point charge penetrating through a disordered medium. He evaluated the mean energy loss per path length, i.e., the stopping power, within the Born approximation, and found that it can be expressed by a sum of integrals over the generalized oscillator strengths,

$$f_{n0}(Q)=\frac{1}{Q}(E_n-E_0)|F_{n0}(\mathbf{q})|^2 \ . \tag{1}$$

Here, the E_n are electronic energy levels of the atoms or molecules of the penetrated medium ($n=0,1,2,\ldots$), and the matrix elements $F_{n0}(\mathbf{q})$ are defined by

$$F_{n0}(Q)=Z^{-1/2}\left\langle n \left| \sum_j e^{i\mathbf{q}\cdot\mathbf{r}_j} \right| 0 \right\rangle \ , \tag{2}$$

where the sum runs over all Z electrons of the target atom or molecule. The quantity $\hbar\mathbf{q}$ is the momentum transfer in an individual interaction, $Q=\hbar^2q^2/2m$, and m the electron mass.

The information contained in Eq. (1) is adequate in principle to completely determine the mean electronic energy loss of a point charge in a slice of matter as well as all higher moments over the energy-loss spectrum and the spectrum itself [1–3]. However, for all targets except atomic hydrogen [4], difficulties occur which make the direct computation of stopping powers from Eq. (1) a formidable task. The main problem is providing a reliable set of matrix elements involving sufficiently many states, including the continuum, over a sufficiently wide range of Q values. Even for a comparably simple system like helium, a recent attempt based on Green-function techniques [5] suggests that generalized oscillator strengths cannot readily be computed for Q values far enough into the continuum to allow evaluation of the stopping power without introduction of simplifying assumptions [6].

Most often, only the high-velocity limit of the Bethe theory is considered. In that case, sum rules can be applied [1], and the mean energy loss reduces to Bethe's well-known logarithmic expression which contains only one material parameter in addition to Z, the mean excitation energy I defined by

$$\ln I=\sum_n f_n \ln(E_n-E_0) \ , \tag{3}$$

where $f_n=f_{n0}(0)$ denotes a dipole oscillator strength. Similar parameters enter higher moments [2,3]. This reduces the necessary computational effort substantially [7]. Nevertheless, even for the best-studied systems [8], I values extracted empirically from stopping-power measurements [9] have generally been considered superior to computed ones.

The asymptotic Bethe formula for the stopping power needs to be supplemented by shell corrections at all but the highest velocities [2,10]. These corrections increase in importance with decreasing speed and increasing atomic number of the target. They are crucial near the stopping maximum, and for heavy target atoms they are significant at all projectile speeds. For atomic hydrogen, these shell corrections have been evaluated rigorously [11]. All other existing estimates involve atomic models. The models that have been designed and utilized for this purpose differ in complexity, in their respective ranges of applicability, and in *a priori* accuracy.

It is the purpose of the present paper to present a comparison of four frequently used atomic models with respect to their capability of predicting stopping parameters. Atomic hydrogen is the only atomic system for which a complete set of exact generalized oscillator strengths is available. An accurate evaluation of the stopping cross section for a point charge is available [11]. We have reproduced this calculation and in addition determined the straggling parameter and three higher moments over the energy-loss cross section. This set of stopping parameters, all based on the Born approximation, serves as the standard of reference.

The following atomic models have been considered: (i) the dielectric theory [10,12], (ii) the harmonic-oscillator

model [13], (iii) the kinetic theory [14], and (iv) the binary-encounter model [15]. These models have all been described in detail in the literature. The survey that follows below (Sec. III) serves mainly to specify the distinct features that enter the computations.

Stopping parameters have frequently been determined on the basis of Eq. (1) by insertion of hydrogenlike atomic wave functions [4,6,16,17]. Testing the validity of this procedure obviously requires to go beyond atomic hydrogen and is, therefore, outside the scope of the present paper.

The restriction to atomic hydrogen has a double advantage: Not only can exact moments be computed within the Born approximation, but accurate and reliable input is also available for all parameters required in the model calculations. Therefore, possible conclusions regarding the quality of the atomic models are not likely to be blurred by inadequate input.

On the other hand, atomic hydrogen is a one-electron system, and penetrating particles may excite collective degrees of freedom [12]. Therefore, some caution is indicated in the generalization of our conclusions to higher-Z atoms or molecules.

One major limitation inherent in the Bethe theory is the neglect of higher terms in the Born series in powers of the projectile charge. That aspect is a topic in its own right which involves an assessment of the accuracy of "exact" evaluations [18] and detailed consideration of how the various models can be generalized such as to account for higher-order Born corrections. That aspect will be left out of consideration here.

II. EXACT CALCULATIONS

We evaluate the first five moments over the excitation cross section σ_n,

$$M^{(\nu)}=\sum_n (E_n-E_0)^\nu \sigma_n \ , \quad \nu=1,2,3,4,5 \ . \tag{4}$$

Here, $M^{(1)}$ is the stopping cross section and $M^{(2)}$ the straggling parameter; the moment $M^{(3)}$ is related to the skewness of the energy-loss spectrum, and $M^{(4)}$ to the curtosis [19].

Within the Born approximation, σ_n is given by [3]

$$\sigma_n=\frac{2\pi e_1^2 e^2}{mv^2}\frac{1}{E_n-E_0}\int_{Q_{\min}}^{\infty}\frac{dQ}{Q}f_{n0}(Q) \ , \tag{5}$$

with

$$Q_{\min}=\frac{(E_n-E_0)^2}{2mv^2} \ . \tag{6}$$

Here, e_1 and v are the projectile charge and speed. Expressions for $f_{n0}(Q)$ are well known for atomic hydrogen [1,3], both for the discrete spectrum and the continuum.

The moments specified in Eq. (4) have been evaluated numerically. The integration over Q was performed with an upper limit equal to at least twice the Bethe ridge. When necessary, the integration interval was extended to ensure an error below 0.1%. The sum over the discrete portion of the spectrum was truncated after 400 terms, and the integration over the continuum portion was transformed into an integration over a finite interval. A numerical accuracy of about 0.1% was aimed at for all moments.

TABLE I. Comparison of numerical evaluation of the stopping cross section of atomic hydrogen. The quantity tabulated is the stopping number $L=(mv^2/4\pi e_1^2 e^2)M^{(1)}$, and $R=13.6$ eV.

$2mv^2/R$	Ref. [11]	Present work
0.4	0.012 53	0.012 51
4	0.834 5	0.832 9
40	3.480	3.477

The results for the first moment have been compared with those of Ref. [11]. It is seen from Table I that there is agreement to better than 0.2%.

III. ATOMIC MODELS

A. Dielectric theory

The stopping power of a homogeneous electron gas for a penetrating point charge is given by [12]

$$-\frac{dE}{dx}=\frac{4\pi e_1^2 e^2}{mv^2}\rho L_e(\hbar v_F/e, 2mv^2/\hbar\omega_p) \ , \tag{7}$$

where ρ is the electron density, $\omega_p=(4\pi\rho e^2/m)^{1/2}$ the plasma frequency, v_F the Fermi speed, and L_e a function that can be expressed as an integral over the inverse dielectric function. In the high-velocity limit, L_e becomes independent of the ratio $\hbar v_F/e^2$ and approaches the asymptotic Bethe expression $L_e\simeq\ln(2mv^2/\hbar\omega_P)$.

Within the local-density approximation, the stopping cross section of an atom or molecule can be tentatively connected to the electron density $\rho=\rho(\mathbf{r})$ by the relation

$$S=\frac{4\pi e_1^2 e^2}{mv^2}\int d^3r\,\rho(\mathbf{r})L_e\left[\hbar v_F(\mathbf{r})/e^2, \frac{2mv^2}{\hbar\omega_p(\mathbf{r})}\right] \ , \tag{8}$$

where $\omega_p=\omega_p(\rho(\mathbf{r}))$ and $v_F(r)=v_F(\rho(\mathbf{r}))$.

From the high-speed limit of Eq. (8) one finds the mean excitation energy I in this approximation:

$$\ln I=\int d^3r\,\rho(\mathbf{r})\ln[\hbar\omega_p(\mathbf{r})] \ . \tag{9}$$

In Ref. [10], an additional factor $\sqrt{2}$ was inserted under the logarithm on the right-hand side of Eq. (9) to qualitatively account for atomic binding forces. Including this factor more generally in Eq. (8) would affect also the behavior of S at low projectile speed, where such a correction is not indicated. In Ref. [20], this problem was handled by use of a low-speed and a high-speed representation of L_e, the former being uncorrected and the latter corrected. In order to illustrate the difference, Eq. (8) was evaluated both as it stands and in the version outlined in Ref. [20].

According to Ref. [10], the binding correction should actually become much smaller than $\sqrt{2}$ for light target materials. This is easily seen to be true for helium but not so for atomic hydrogen.

For the direct evaluation of Eq. (8), L_e has been evaluated by straight numerical integration on the basis of the dielectric function given in Ref. [12]. The results agree well with previous numerical evaluations [12,21]. Equation (8) was then integrated numerically by insertion of the electron density of atomic hydrogen in the ground state.

In the evaluation of the corrected stopping power we followed exactly the procedure described in Ref. [20].

The expression given in Ref. [12] for the straggling parameter of a free electron gas was evaluated similarly, with no binding correction. The local-density approximation has previously been applied to straggling in Refs. [22,23]. Higher-order moments were treated correspondingly.

The dielectric theory is the most frequently used model for the computation of stopping cross sections. Early experience [20,24] indicated that the model is more successful in the evaluation of shell corrections than in the evaluation of I values.

B. Harmonic-oscillator model

Within the Born approximation, the stopping section of a spherical harmonic oscillator can be written in the form

$$S_{\rm osc}=\frac{4\pi e_1^2 e^2}{mv^2}L_{\rm osc}(2mv^2/\hbar\omega_0)\ , \tag{10}$$

where ω_0 is the classical resonance frequency and $L_{\rm osc}(\xi)$ a function that has been computed accurately [13]. In the high-speed limit, that function approaches the Bethe logarithm $L_{\rm osc}(\xi)\sim\ln\xi$.

It was proposed in Ref. [13] to determine the stopping cross section of an atom from the expression

$$S=\frac{4\pi e_1^2 e^2}{mv^2}\sum_n f_n L_{\rm osc}\left[\frac{2mv^2}{E_n-E_0}\right]\ . \tag{11}$$

This was evaluated in Ref. [13] for atomic hydrogen. Those results were accurately reproduced in the present computations.

The corresponding expression for straggling was also suggested in Ref. [13], and the values obtained for atomic hydrogen were confirmed in our computations. Higher-order moments were evaluated correspondingly.

The expression (11) goes over into the asymptotic Bethe formula at high projectile speed, i.e., the mean excitation energy I takes on the exact value given by Eq. (3) in this model. Application of the proper sum rule shows that also the leading term in the shell-correction expansion agrees with the exact value for any target, and for the specific case of atomic hydrogen, the same was found to be true for the second term in the shell-correction expansion in powers of v^{-2} [13]. Moreover, the dependence of the mean energy loss on impact parameter that is implied by the oscillator model agrees asymptotically with Bloch's exact result in the dipole limit [25,26]. The oscillator model has also been shown to correctly describe the asymptotic behavior of the straggling parameter at high velocities in the two leading terms [13].

C. Kinetic theory

In Ref. [14], one of us suggested evaluating the stopping cross section of an atom from the expression

$$S=\int d^3v' f(\mathbf{v}')\frac{\mathbf{v}\cdot(\mathbf{v}-\mathbf{v}')}{v|\mathbf{v}-\mathbf{v}'|}S_0(|\mathbf{v}-\mathbf{v}'|)\ , \tag{12}$$

where $S_0(v)$ is the stopping cross section in the high-speed limit, i.e., the asymptotic Bethe formula. Here, $f(\mathbf{v}')$ is the velocity spectrum of the target electrons which is determined via Fourier transformation of the wave function. For a ground-state atom, the velocity spectrum is isotropic, $f(\mathbf{v}')\equiv f(v')$, so that Eq. (12) reduces to a one-dimensional integral. In this model, the mean excitation energy I enters as an input parameter, i.e., the kinetic theory is primarily a scheme to determine shell corrections.

Equation (12) has been evaluated on the basis of the exact I value and the velocity spectrum of atomic hydrogen in the ground state. The integration includes only target velocities for which S_0 is positive. The corresponding expression for straggling was also suggested in Ref. [14] and has been evaluated along the same line.

The validity of this model rests on the assumption that major deviations from the asymptotic Bethe formula are of a kinematic nature: with decreasing projectile speed, the fact that v becomes comparable to the electron speed v' is considered more important than the fact that $2mv^2$ becomes comparable with electron binding energies. It was demonstrated Ref. [14] that this model correctly predicts the first term in the shell-correction expansion of the stopping cross section as well as that of the straggling parameter.

D. Classical binary-encounter model

The classical binary-encounter model operates on the basis of free Coulomb scattering between moving particles and an effective binding energy which can be treated as an adjustable parameter [15]. The stopping cross section arising from this model is a factor of 2 too small in the limit of high velocities [27]. In the straggling parameter, binding forces become significant beyond the leading order in v^{-2} [28]. Higher moments are known to be less sensitive to binding forces [19].

In order to avoid ambiguities we have only computed third- to fifth-order moments from this model. These results complement the first and second moment computed from the kinetic theory. The same electron velocity spectra have been utilized in the two types of computation.

IV. RESULTS

A. The stopping cross section

Figure 1 shows the first moment, i.e., the stopping cross section. It is seen that within the Born approximation, the stopping cross section is quite accurately reproduced by the harmonic-oscillator model at projectile speeds down to $2mv^2/R\sim 3$, where $R=13.6$ eV. The agreement is quantitative in the energy range above the stopping maximum. The error near the maximum is

$\sim 1\%$. At lower velocities, the oscillator model leads to a slight overestimate of the stopping cross section except near threshold where it crosses the exact result. Note that at those low projectile speeds, higher-order Born terms overshadow these discrepancies [29,30].

The agreement found here between the oscillator model and the exact result is better than reported in Ref. [13]. The reason for this is that in Ref. [13], comparison was made with a stopping cross section computed by Turner [4] and reported in Ref. [2]. The comparison suggested that a single harmonic oscillator should provide a better estimate for the stopping cross section of atomic hydrogen that the ensemble specified by Eq. (11). This finding was highly implausible. A subsequent reevaluation of the "exact" result revealed a slight inaccuracy in the latter [11].

The dielectric theory yields good agreement with the exact result down to $2mv^2/R \sim 10$, i.e., almost down to the stopping maximum. Inclusion of a factor $\sim 1/\sqrt{2}$ in L_e, Eq. (8), appears indeed essential in this regime. Below the stopping maximum, the dielectric theory predicts the well-known velocity-proportional stopping cross section that is characteristic of the free-electron gas. This behavior is very different from that predicted by the Bethe theory.

The kinetic theory has been tailored to yield the exact asymptotic behavior at high projectile speed. Figure 1 shows that down to the stopping maximum, this model yields results that are intermediate between those obtained from the corrected dielectric theory and the oscillator model. At low velocities, the prediction of the kinetic theory is intermediate between the uncorrected dielectric theory and the straight Born result.

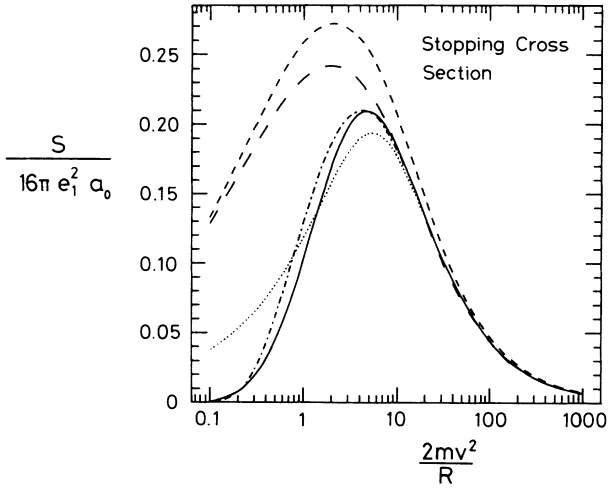

FIG. 1. Stopping cross section $S \equiv M^{(1)}$ of a point charge in atomic hydrogen. Solid line: Born approximation, direct integration; short-dashed line: dielectric theory, uncorrected [Eq. (8)]; long-dashed line: dielectric theory, corrected for binding according to Ref. [20]; dotted line: kinetic theory; dash-dotted line: oscillator model. $R = 13.6$ eV; $a_0 = 0.529$ Å.

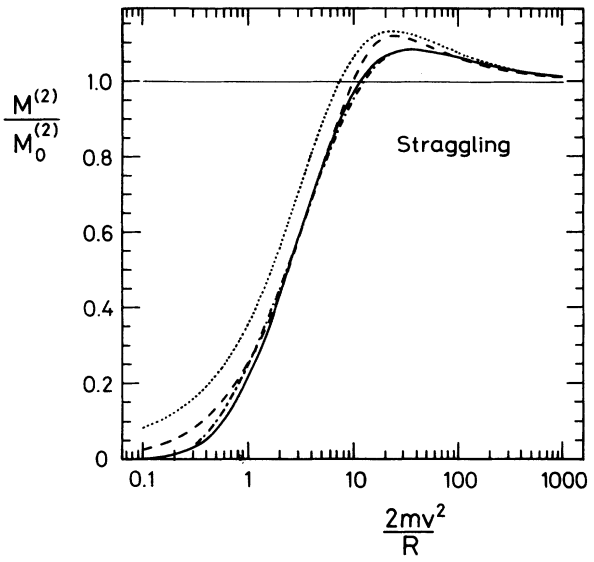

FIG. 2. Straggling parameter of a point charge in atomic hydrogen. The quantity plotted is the ratio between the computed second moment $M^{(2)}$, Eq. (4), and Bohr's result, $M_0^{(2)} = 4\pi e_1^2 e^2$. Notation as in Fig. 1.

B. Straggling

Figure 2 shows second moments. All curves approach Bohr's result [28] at high velocities, and a Bethe-Livingston-type maximum [31] is observed at intermediate velocities. All models yield good overall agreement with the straight Born result. However, the prediction of the kinetic theory is consistently high. The result from the dielectric theory is high around the maximum as well

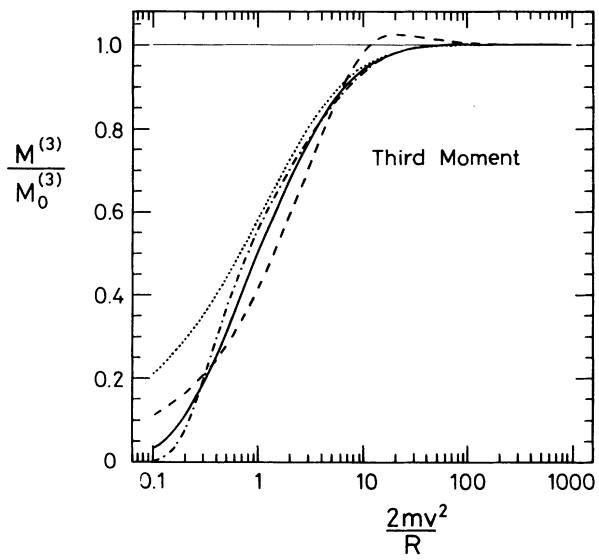

FIG. 3. Third moment over the energy-loss spectrum, normalized to the free-Coulomb value for stationary target electrons, $M_0^{(3)} = 4\pi e_1^2 e^2 mv^2$. Notation as in Fig. 1, except that the dotted line refers to the binary encounter model.

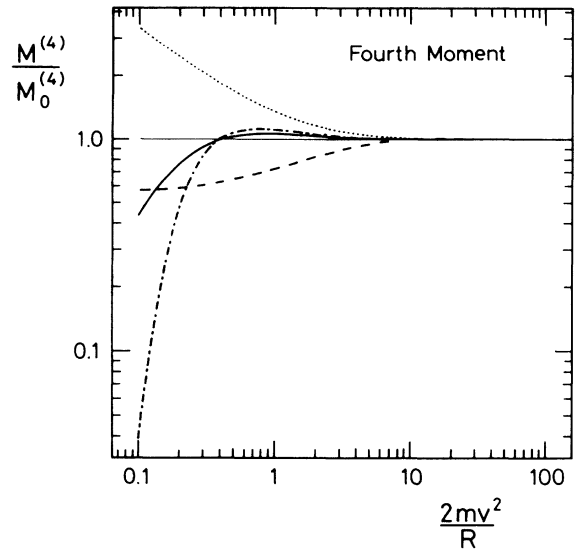

FIG. 4. Same as Fig. 3 for the fourth moment, with $M_0^{(4)}=4\pi e_1^2 e^2(2mv^2)^2/3$.

as at very low velocities. The oscillator model shows very good agreement ($\sim 1\%$) with the straight Bethe result down to $2mv^2/R \sim 2$, and acceptable agreement down to $2mv^2/R \sim 0.3$.

C. Higher moments

Figure 3, which shows third-order moments, does not indicate major changes in comparison to the behavior of the second moments. The errors of both the oscillator model and the dielectric theory have become only slightly larger. Predictions of the kinetic theory have not been included, but even the straight binary-encounter model, assuming free-Coulomb scattering and neglecting any binding energy [19], is seen to yield very satisfactory results except at the lowest velocities.

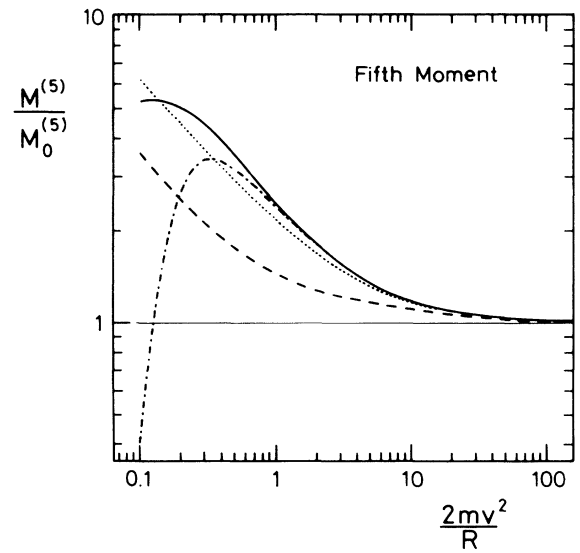

FIG. 5. Same as Fig. 3 for the fifth moment, with $M_0^{(5)}=4\pi e_1^2 e^2(2mv^2)^3/4$.

Discrepancies increase in the fourth and fifth moment (Figs. 4 and 5). Note the logarithmic scale in the ordinate. Down to somewhat below $2mv^2/R \sim 1$, the oscillator model is seen to show excellent agreement with the straight Born integrals while the dielectric theory is consistently low for $2mv^2/R \lesssim 10$, with the error increasing with decreasing energy. Drastic errors show up in the predictions of the oscillator model for $2mv^2/R < 0.2$. This is not unexpected. More surprising appears the fact that such discrepancies do not show up at considerably higher projectile velocities.

V. DISCUSSION

As a main conclusion, one may note that three of the considered models, the dielectric theory, the oscillator model, and the kinetic theory, all provide reasonable estimates of the stopping cross section almost down to the stopping maximum. For the dielectric theory, this requires inclusion of a binding correction of the order of $\sqrt{2}$ even for an atomic hydrogen target. The harmonic-oscillator model shows more accurate agreement with the straight Born result in this velocity range than the other two approximations. At velocities below the stopping maximum, major differences occur. Here, surprisingly close agreement is found between the prediction of the oscillator model and the straight Born result.

The oscillator model has been designed to correctly describe soft interactions. Discrepancies must be expected under conditions where mostly violent interactions contribute, i.e, for higher moments and, generally, for all moments at velocities approaching the threshold for excitation. Figure 1 demonstrates that this breakdown occurs at much lower velocities than could have been anticipated. Similar conclusions emerge from considering the behavior of the higher moments which increasingly hinge on low-impact-parameter collisions. Moreover, the zeroth moment, i.e., the total cross section, is exclusively determined by soft interactions and, therefore, very accurately approximated by the harmonic-oscillator model. For most practical purposes, these six moments characterize the overall behavior of the differential cross section very well.

The dielectric theory was designed with the aim of estimating stopping parameters for many-electron atoms. While the model appears intuitively appealing, no limiting case is known where it is exact. An illuminating discussion [32] gave no clues on the range of validity of the model. It is well established that for the homogeneous electron gas, about half the stopping power goes into collective excitations in the high-speed limit. Applying such a model to a one-electron atom could be expected to lead to artifacts. Indeed, there is a noticeable error in its predicted impact-parameter dependence of the electronic energy loss [33]. In consideration of these features, the observed deviations of the predicted moments from the straight Born approximation are surprisingly small. The biggest disparities are actually observed in the first moment, i.e, the stopping power, where the necessity of in-

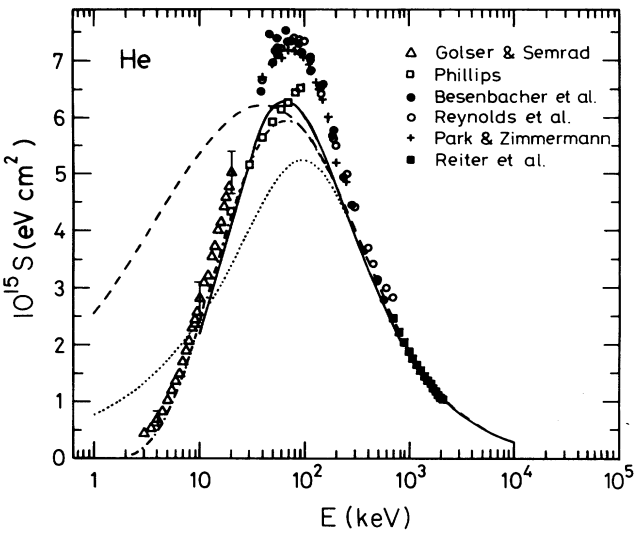

FIG. 6. Same as Fig. 1 for helium target. Full drawn curve from Ref. [6]. Experimental data: Semrad and Golser [34]; see also Ref. [36].

cluding a binding correction at high velocities, and omitting it at lower velocities, has long been a cause of considerable concern.

Predictions of the kinetic theory have been included only for the first and second moment. These predictions are known to compare favorably with more accurate calculations at not too low projectile speeds. For higher moments, only straight binary-encounter expressions have been evaluated, i.e., expressions that do not contain the mean excitation energy. Its predictions become exceedingly accurate with increasing order of the moments, as is seen by inspection of Figs. 3–5.

In view of recent stopping measurements on hydrogen and helium gas at very low energies [34], a comparison with some of the present results is of interest. The stopping cross section of helium for a penetrating point charge was evaluated similarly to the case of hydrogen, yet with approximate input. The results of Ref. [6] served as a reference standard. Dipole oscillator strengths needed in the oscillator model were taken from Ref. [7], and electronic charge densities and velocity spectra were found from hydrogenic wave functions with standard parameters [35]. A comparison of the theoretical stopping cross sections shown in Fig. 6 shows a qualitatively similar behavior as atomic hydrogen. There is very good agreement between the results of Ref. [6] and the oscillator model. No correction was applied to the dielectric theory, in accordance with what has been said above. The behavior of the curves predicted by the kinetic and the dielectric theory is very similar to what was found for hydrogen.

A number of experimental results were included in addition to the data reported in Ref. [34]. There is strikingly good agreement with the results of Ref. [6] as well as the oscillator model at energies well above and well below the maximum. Also the position of the maximum is well predicted, but the height is slightly underestimated.

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