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Sigmund, Peter; Schinner, Andreas

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Is electronic stopping of ions velocity-proportional in the velocity-proportional regime?

P. Sigmund\textsuperscript{a}, A. Schinner\textsuperscript{b}

\textsuperscript{a}\textit{Department of Physics, Chemistry and Pharmacy, University of Southern Denmark, DK-5230 Odense M, Denmark}
\textsuperscript{b}\textit{Department of Experimental Physics, Johannes Kepler University, A-4040 Linz, Austria}

Abstract

We have studied systematic deviations of reported electronic stopping cross sections from velocity proportionality predicted by Lindhard and Scharff. Most pronounced is an apparent drop-off at beam energies below $\sim 0.01 \text{ MeV/u}$. In the frequently-used transmission technique, only a narrow cone of the beam is analysed in order to minimize the contribution from nuclear stopping. This has the consequence that a reduced instead of the full electronic energy loss is recorded. The magnitude of this effect has been computed on the basis of a recently developed extension of the scheme of Fastrup et al. to correct for nuclear stopping. In a detailed study of the C-Ar system based on our PASS code we find close agreement between the derived reduced electronic stopping cross section and the experimental result reported by Ormrod. Results are reported for eight ions on argon, for O in C and Cl in C and for Ni in Ag. For Ni in Ag as well as for one set of reported stopping cross sections for N in Ar a particularly sharp drop-off is shown to be caused by an inadequate nuclear-stopping correction. Our findings do not support Paul’s assertion of a gas-solid difference in electronic stopping of low-energy heavy ions.

Keywords: Stopping power, Threshold effects, Transmission measurement, PASS code

1. Introduction

This study addresses the slowing-down of ions in the velocity range below the stopping maximum (or Bragg peak). According to Lindhard & Scharff \cite{1} the Bragg peak lies close to a projectile speed $v$ equal to the Thomas-Fermi speed $v_{TF} = v_0 Z_1^{2/3}$, where $v_0$ denotes the Bohr speed and $Z_1$ the atomic number of the projectile ion. While in early theories \cite{2, 3, 4} the electronic stopping force was expected to rapidly approach zero below $v = v_0$, explicit studies of stopping in a Fermi gas \cite{5, 6} and of atomic-collision theory \cite{7, 8} suggested a linear dependence on the beam velocity. The very appearance of a linear dependence is a rather general result since it reflects the first term in a Taylor expansion \cite{9}, while the multiplying factor is model-dependent.

The validity of the Lindhard-Scharff formula for the electronic stopping cross section \cite{1},

$$S_e = 8\pi \xi e Z_1 Z_2 / (Z a_0 e^2 v / v_0), \quad (1)$$

where $Z^{2/3} = Z_1^{2/3} + Z_2^{2/3}$, $a_0$ the Bohr radius 0.0529 nm and $\xi e \approx Z_1^{1/6}$ an empirically determined parameter, was supported by an analysis of measured ranges of fission fragments \cite{10}. However, direct measurements \cite{11, 12, 13, 14, 15} revealed that when $S_e$ is matched to a power law

$$S_e = C E^p, \quad (2)$$

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the proportionality factor $C$ shows an oscillatory structure as a function of $Z_1$ which is absent in Eq. (1). Moreover, the exponent $P$ was found to differ significantly from $P = 1/2$ \cite{11, 12} and was likewise found to oscillate with $Z_1$ \cite{13, 14}.

A few years later, direct stopping measurements in the fission-fragment range \cite{16} suggested a linear dependence of $S_e$ on $v$, but with an extrapolated threshold velocity at $v \approx 2v_0$. Recent studies \cite{17, 18}, however, led to the conclusion that there was no contradiction: Stopping cross sections were found to change slope mainly due to variation in equilibrium ion charge \cite{18}.

In the meantime a large amount of experimental data has accumulated and been compiled in the IAEA database \cite{19}. Even a superficial glance at those data shows clear evidence supporting velocity-proportionality for many ion-target combinations. This is surprising, not the least because deviations from strict proportionality in $v$ have been looked for, especially threshold effects caused by bandgaps \cite{20}. On the other hand, significant deviations from velocity proportionality have been found which have not only been unexplained but even have served as guidance for empirical interpolation programs such as SRIM \cite{21} or MSTAR \cite{22}.

In this paper we demonstrate that a major source of such deviations from typical behavior lies in the analysis of raw experimental data, which typically is based on the assumption of electronic stopping acting as a mere friction force. The possible significance of this has been mentioned repeatedly in the past, especially in early work by the Linz group \cite{23}, although a quantitative estimate was not given.

2. Examples

Figures 1 and 2 show pairs of typical but quite different examples. Stopping cross sections reported for oxygen and chlorine ions in carbon (Figure 1) from 22 and 9 sources, respectively, follow the expected behavior, as is seen by comparison with the predictions of the PASS code \cite{26, 24} which approaches a $\sqrt{E}$ dependence at low velocity. For carbon and nitrogen ions in argon gas (Figure 2), only few data have been reported altogether, as is typical for gas targets, and those data show a noticeably steeper slope below $\sim 0.025\text{MeV/u}$, i.e., for $v \lesssim v_0$. Moreover, two conflicting data sets are found for nitrogen in argon.

The behavior of the stopping cross section of carbon in argon is typical not only for that of other ions in argon \cite{14} but also for other gas targets. From the fact that PASS output appears to overestimate stopping cross sections in several gas targets it was asserted \cite{27} that there must be a gas-solid difference in the stopping of low-energy ions amounting to up to 60% as compared to PASS data reported in ref. \cite{28}.

We first note that, if real, such a gas-solid difference would show a strong energy dependence and would, at the lowest energies studied, exceed a factor of two for C in Ar and even more for N in Ar. While we rejected \cite{29} the suggestion brought forward in ref. \cite{27} of this being caused by a gas-solid difference in the equilibrium charge, we did not offer an explanation for the change in slope for $v < v_0$.

Our recent analysis of stopping measurements in the transmission geometry \cite{30} has resulted in what we find a more adequate explanation, to be discussed in the following section.

3. Measurement in transmission

Electronic stopping cross sections may be measured by a variety of techniques. Most direct is the transmission method, where projectiles pass through a target of a well-defined thickness and the stopping
Figure 1: Stopping cross section of oxygen (upper graph) and chlorine (lower graph) ions in amorphous carbon. Experimental data from IAEA database [19]. Red solid lines: PASS [24].
Figure 2: Same as figure 1 for carbon (upper graph) and nitrogen (lower graph) ions in argon gas. Low-energy data for C-Ar (filled triangles) from Ormrod [14], for N-Ar from [14] (filled circles) and [25] (filled squares). Also included is output from SRIM [21].
cross section is determined from the energy spectrum of the transmitted ions.

In transmission measurements in the velocity-proportional regime, care needs to be taken to eliminate the contribution from nuclear stopping. This is accomplished by making use of the fact that nuclear stopping is accompanied by angular deflection. By analysing only a narrow beam of penetrating ions, most of the nuclear energy loss is eliminated, and only a small fraction of multiply-scattered ions enters the detector. However, deflected ions also experience electronic energy loss. Since electronic energy loss increases with decreasing impact parameter, i.e., increasing scattering angle, a reduced electronic energy loss will be recorded in the detector.

A theoretical scheme dealing with this effect has been developed in ref. [30] as an extension of a formalism developed by Fastrup et al. [13] on the basis of Bohr-Williams theory of multiple scattering [4]. According to this theory the angular distribution of a beam after penetrating a thin target is composed of a gaussian core with the width defined by the multiple-scattering angle \( \alpha_1 \) given by

\[
\alpha_1^2 = N x \int_0^{\alpha_1} \alpha^2 d\sigma(\alpha)
\]

and a tail defined by the differential cross section,

\[
F(\alpha)2\pi\alpha d\alpha = N x d\sigma(\alpha),
\]

where \( x \) denotes the target thickness. Within the gaussian part of the beam, both electronic and nuclear energy loss are assumed independent of \( \alpha \). At larger angles either quantity is given by the respective differential cross section. The border line between the two regimes is given by the cross-over between the two expressions for the angular distribution. Reduced nuclear and electronic stopping depend on the opening angle \( \phi \) of the detector and the multiple-scattering angle \( \alpha_1 \) which in turn depends on the target thickness in terms of a parameter

\[
\xi = N x a_0^2
\]

and the beam energy \( E \).

In ref. [13] the reduced nuclear energy loss was determined from the relation

\[
S_{n,\text{red}} = \int_0^{T_1} T(p)d\sigma(T),
\]

where \( T_1 = T(\alpha_1) \), and the electronic energy loss was determined by subtracting the reduced nuclear energy loss from the maximum in the recorded energy-loss spectrum.

Our treatment in ref. [30] adds three aspects to the Fastrup scheme,

- Instead of operating with the peak energy loss we determine average energy losses as a function of the detector opening angle \( \phi \).
- We allow the detector opening angle \( \phi \) to exceed the multiple-scattering angle \( \alpha_1 \).
- The dependence of electronic energy loss \( T_e(p) \) on impact parameter \( p \) is a central part of the description.

While computations in ref. [13] were performed assuming the standard Thomas-Fermi interatomic potential [31], we mostly used the Thomas-Fermi-Moliere potential for analytical convenience. Having made several checks we find the difference insignificant compared to calculated differences between full and reduced electronic stopping cross sections.
4. Restricted stopping cross sections

4.1. C in Ar

Figure 3 shows an expanded version of Figure 2, where restricted electronic stopping cross sections computed for several values of $\phi$ and $\xi$ have been included. These curves are compared with results reported by Ormrod [14].

It is seen that for $\xi = 1$ (upper graph) the restricted stopping cross section is independent of the detector opening angle for $E < 0.01$ MeV/u. The multiple-scattering angle is large in this energy range, so that only multiple-scattered projectiles are recorded. With increasing energy, i.e., decreasing multiple-scattering angle, an increasing portion of singly-scattered ions will enter the detector, with the result that the restricted electronic stopping depends on the detector opening and, eventually, approaches the full electronic stopping cross section.

From the three graphs we conclude that for $\phi = 0.02$, calculated restricted stopping cross sections agree with the measurements for $\xi = 0.4$ and $\xi = 0.1$ but not for $\xi = 1$.

We have not found information on $\phi$ and $\xi$ in Ormrod’s paper. However, restricted nuclear stopping cross sections were tabulated, so that it is possible to extract the parameter $\xi$ uniquely.

Figure 4 shows reduced nuclear stopping cross sections calculated for the angles $\phi$ in Figure 3 and three values of $\xi$. We note that also this quantity is independent of $\phi$ at low energies. Moreover, it is $\propto 1/E$, as found by Fastrup et al. [13] for the multiple-scattering regime. At the transition to single-scattering the $1/E$ dependence turns over into a slow increase with a gradual approach toward full nuclear stopping. The single-scattering regime was ignored in the Fastrup scheme and, hence, did not enter Ormrod’s data treatment.

Figure 4 shows that the actual $\xi$-value pertaining to the data must lie between $\xi = 0.1$ and 1. The actual value $\xi = 0.38$ found by trial and error is included in the bottom graph.

Figure 5 shows the results of our analysis. Experimental parameters in Ormrod’s setup must have been close to $\xi = 0.38$ and $\phi = 0.020$. The nuclear-stopping correction applied by Ormrod is correct for the lowest energy value. At higher energies the proper nuclear-stopping correction is no longer $\propto 1/E$, but the actual error made is seen to lie in the 1-2% range.

The reported values are fully compatible with predictions of PASS, if interpreted as reduced stopping cross sections. The reduced electronic stopping cross section underestimates the full electronic stopping cross section by a factor of 1.5 at the highest of five measured values and by a factor of 2.8 at the lowest point.

As a word of caution we emphasize a major difference between the parameters $\xi$ and $\phi$:

- We have determined $\xi$ solely from the nuclear-stopping correction given in the original paper [14]. We used exactly the same formalism as Ormrod, except for interchanging input with output. Therefore, we are confident that our extracted $\xi$-value, which also enters figures 6 and 8, is a reliable estimate of the actual value.

- On the other hand, the determination of $\phi$ in figure 5 hinges on a comparison between PASS predictions and quoted experimental values corrected for nuclear stopping. Evidently, this fit is not very sensitive to the adopted value of $\phi$. This, taken together with a built-in uncertainty of the PASS code itself, indicates considerable uncertainty about our extracted values of $\phi$. 
Figure 3: Upper graph in Figure 2 amended by restricted electronic stopping cross sections computed for detector angles $\phi = 0.02, 0.01, 0.005, 0.002$ and $\zeta = 1, 0.4$ and 0.1 (top to bottom graph). ASCS is the name of the code which computes reduced stopping cross sections from PASS [30]. Filled triangles: Measurements of Ormrod [14].
Figure 4: Restricted nuclear stopping in C-Ar. Filled triangles: From [14]. Detector opening angles as in Figure 3. \( \xi = 1, 0.1 \) and 0.38 (top to bottom). The abscissa scale has been expanded to illustrate the behavior of nuclear stopping.
Figure 5: Electronic and nuclear stopping of C in Ar in transmission geometry. Full (solid lines) and reduced (broken lines) stopping cross sections for $\xi = 0.38$ and $\phi = 0.025, 0.020$ and 0.015. Solid triangles: [14].

4.2. N in Ar

Figure 6 shows the analogous case of N in Ar. Reduced nuclear and electronic stopping cross sections were computed with the values for $\xi$ and $\phi$ deduced from the C-Ar system. This appears appropriate since the pressure was kept at $\sim 10\mu = 1.33$Pa in the measurements of [14] and is confirmed by the fact that the reduced nuclear stopping cross sections calculated for $\xi = 0.38$ agree with the values given in ref. [14].

While there is found excellent agreement between the calculated reduced stopping cross section and the data given in ref. [14], we have not succeeded in finding a pair of $\xi, \phi$ parameters that produced a similar match for data from [25]. Neither values of $\phi$ or $\xi$ nor the applied nuclear-stopping correction were specified in the paper. Instead, we assert that data from [25] were determined by subtracting the full nuclear stopping cross section from the measured signal.

In support of this assertion we added the nuclear stopping cross section to the reported electronic stopping cross section. This leads to the filled red circles in Figure 7, which are very close to the data from [14]. This suggests that parameters $\xi, \phi$ have been similar in the two setups. We also note that the actual (restricted) nuclear-stopping correction is negligible in comparison with the difference between the two sets of reported data.

4.3. Other ions in argon

Figure 8 shows similar graphs for ions from H to Ne in Ar. Since all measurements – with the possible exception of H and D bombardment – were performed on the same apparatus, we assumed the
Figure 6: Stopping of N in Ar gas. Low-energy data from Ormrod [14] (triangles) and Fukuda [25] (squares).
same value for $\phi$ in the analysis. As mentioned above, also $\xi$ is kept at the value 0.38 assuming constant pressure.

While there is found excellent agreement between calculations and data from for He, B and O ions, there are noticeable differences for H, F and Ne.

For Ne-Ar a significant difference had to be expected, because Ne lies close to a minimum in $Z_1$ oscillations, not only in Ar gas but also in C and Al targets [12, 13]. This feature, although less pronounced, comes up already for F ions.

Hydrogen and deuterium are the only projectiles for which no nuclear-stopping correction is listed in ref. [14], yet Ormrod lists data for H and D bombardment separately. There is found nearly-complete agreement between the two sets, despite the fact that nuclear stopping and scattering differ. While the nuclear-stopping correction is small, Figure 8 shows that reduced electronic stopping cross sections should differ significantly both from each other and from the unrestricted stopping cross section, if the measurements had been done with the same set of parameters as the other $Z_1$–Ar data. Therefore we conclude that measurements with H and D were done with a different choice of parameters.

4.4. Ni in Ag

Figure 9 shows the case of Ni in Ag. Experimental data, all from one group [32, 33], show a rapid decrease from 0.2 MeV downward, which cannot be explained as a geometry effect. However, a result compatible with general behavior is obtained when the full nuclear stopping cross section is added to the quoted results, as is shown in the lower graph in Figure 9. Thus, as in Figure 7, the full nuclear stopping cross section was subtracted from the measured values, as is actually stated in the paper [33]. The upper
Figure 8: Stopping cross sections for $Z_1$-Ar with $Z_1 = 1, 2, 6, 8, 9$ and 10. Notation as in Figures 5 and 6.
Figure 9: Stopping cross section for Ni in Ag. Experimental data from [32, 33]. Upper graph: Reduced stopping cross section for $\phi = 0.01, \xi = 1.0$. Also included is output from [21]. Lower graph: Sum of full $S_n$ and reported $S_e$ included (filled triangles).
Figure 10: Electronic stopping of Cl in C. Lower graph of Figure 1 amended by reduced electronic stopping cross section. See text

graph also demonstrates that the resulting electronic stopping cross section, which underestimates the actual value by a factor of 4 - 8, was taken as a basis for the SRIM tabulation [21].

4.5. Cl in C

Finally we return to Figure 1. In Figure 10 we have added a reduced stopping cross section computed on the basis of the values of $\phi = 0.02$ and $\xi \simeq 4.9$ extracted from [12]. It is seen that the calculated curve deviates significantly from the experimental data, whereas the data agree well with the full PASS stopping cross section. This feature – which is likewise found for O in C – has several possible explanations, one of which being a systematic error in the PASS code. We shall come back to this example in a companion study devoted to the reciprocity principle [34].

5. Threshold Behavior

Here we mention an effect that is well-known in atomic-collision physics but usually ignored in stopping theory. Consider Firsov’s formula for the electronic energy loss $T(p)$ as a function of impact parameter $p$ [7],

$$T(p) = \frac{A v}{(1 + B R_{\text{min}})^{5/3}},$$  \hspace{1cm} (7)

where $A = 0.35h(Z_1 + Z_2)^{5/3}/a_0$ and $B = 0.16(Z_1 + Z_2)^{1/3}/a_0$, $R_{\text{min}}$ is the distance of closest approach defined by

$$1 - \frac{V(R_{\text{min}})}{E_{\text{rel}}} - \frac{p^2}{R_{\text{min}}^2} = 0,$$  \hspace{1cm} (8)
Figure 11: Effect of binding on full and reduced electronic stopping with $\phi = 0.02$ and $\xi = 0.38$ for C in Ar. Blue lines: Firsov; red lines: PASS. See text.

$E_{\text{rel}}$ is the relative energy in the c.m.s. frame and $V(R)$ the screened potential governing scattering of the projectile ion on a target atom.

In stopping theory it is customary to operate with a uniformly moving projectile, so that $R_{\text{min}} \equiv p$. This assumption must be expected to overestimate the electronic energy loss, since $R_{\text{min}} > p$ for repulsive interaction and $T(p)$ decreases with $p$. The difference must be expected to increase with increasing $R_{\text{min}}$, i.e., decreasing energy.

In order to estimate the magnitude of this near-threshold effect in PASS we have replaced $T(p)$ by $T(R_{\text{m}}(p))$. Figure 11 shows the situation for the C-Ar system with the parameters adopted in Figure 5. It is seen that the difference between the full electronic stopping cross sections for bent and straight trajectory, i.e., the difference between the solid and broken lines, is barely visible in the graph. Moreover, the difference between Firsov and PASS predictions is close to zero except at the upper end of the energy range, where the PASS curve bends over toward the stopping maximum. There is a marked difference between Firsov and PASS predictions for the reduced stopping cross sections. Since reduced stopping involves trajectories that have experienced small-angle scattering, bent trajectories are not expected to contribute. A minute difference is visible in case of PASS at the low-energy end. For Firsov the difference is less than 1% at the lowest energy in the graph.

From this and several other examples we conclude that for beam energies from 0.001 MeV/u upward, trajectory bending has a negligible influence on reduced electronic stopping cross sections. The effect is visible in the full electronic stopping cross section but far smaller than the difference between the full and reduced stopping cross sections.
Figure 12: Stopping cross sections for He-He, H-He and H-H (top to bottom). Measurements from Golser et al. [35, 36] and Raiola et al. [37]. Lines with markers referring to restricted stopping cross sections for $\phi = 0.0019$ and $\xi = 1.48$ (minimum) and 22.1 (maximum) based on figures given in [35].
6. Conclusions

In response to the question asked in the title, significant deviations from velocity-proportional stopping are well-documented in published data for stopping of ions heavier than protons in both gases and solid matter in the energy range below \( \sim 0.01 \text{ MeV/u} \). Most of those data originate in measurements in transmission geometry. We find that with one exception (see below) all data analysed in the present work are compatible with predictions of the PASS code after analysis by the tools developed in ref. [30], i.e., impact-parameter-dependent electronic stopping and angular (single and multiple) scattering on a Thomas-Fermi potential.

We have also identified cases where the correction for nuclear stopping was incorrectly performed by subtracting the unrestricted nuclear stopping cross section from the measured data.

In other words, in most of the examples discussed in this study, reported deviations from velocity-proportional electronic stopping do not reflect a behavior of the stopping cross section but are a characteristic feature of the transmission technique.

We note that our computed values of restricted electronic stopping cross sections in most cases are estimates based on feasible values of target thickness (or gas pressure) and detector opening angle, since at least one of these values is missing in most publications.

However, this is the point to mention a prominent exception. In a frequently-quoted paper, Golser et al. [35] reported measurements of the stopping cross section of He gas for protons and deuterons, which showed a very steep dependence on the beam velocity. Subsequent measurements on He-He and H-H [36] did not show this effect. Since both the dimensions of the scattering chamber, the slit width and the range of applied pressures are given in the paper, we were able to estimate a range of reduced stopping cross sections for all three systems. Figure 12 shows that the calculated stopping cross sections are insensitive to the target pressure, in agreement with the measurements, and that the calculations yield the same slope for all three ion-target combinations. Thus, taken the dimensions extracted from the paper we conclude that the behavior of the H-He points is not caused by inadequate analysis but a property of the unrestricted stopping cross section, as rationalized in the original paper [35].

Based on our analysis of data on stopping in argon, we do not see evidence to support Paul’s assertion [27] of a significant gas-solid difference in low-velocity stopping.

We do not wish to deny the existence of real threshold effects beyond what we discussed in Section 5, but we want to emphasize that treating electronic stopping as a mere friction force in the data analysis is likely to affect all reported experimental data on electronic stopping at low velocity.

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