Energy losses and their straggling of H^+ and He^+ ions of several hundred keV energy after passing through metal and polystyrene films

A. A. Bednyakov, Yu. V. Bulgakov, V. S. Nikolaev, and V. L. Chernov

Nuclear Physics Research Institute
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We measured the energy losses and their straggling for H^+ ions with energies from ~120 to 480 keV and for He^+ ions with energies from ~150 to 1300 keV in thin films of Al, Cu, Ag, Au, and polystyrene $(C_8H_8)_m$. The data obtained on the energy-loss straggling are discussed on the basis of the existing theories. It is shown that in the investigated ion-energy regions an important role (and in the case of the He ions, a dominant one) is played by additional straggling mechanisms due to fluctuations of the ion charges and to the correlations of the energy losses when the ion interacts simultaneously with several electrons of the medium. Allowance for these mechanisms improves considerably the agreement between the calculation results and the experimental data.

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1. INTRODUCTION

Extension of the possible use of beams of ions having energies of several dozen or several hundred keV as a tool for investigating and altering the structure of the surface layers of solids calls for more accurate and deeper knowledge of the processes that accompany the penetration of such ions through matter. Of particular interest in this respect is the slowing down of ions, and it is important to know not only the average energy losses but also their straggling. Many experimental studies were therefore made recently of the energy straggling of light ions in both gaseous[1,2] and solid[3-9] targets, and attempts were made to make a more detailed theoretical analysis of this phenomenon. Thus, Refs. 1 and 10 contain a consistent refinement of the traditional theory[11,12] based on the model of independent pair collisions of the ions with the electrons of the medium. Other workers have considered the effect exerted on the energy straggling by additional mechanisms connected with the fluctuations of the charge of the decelerating ions[7,13] or with the ion-loss correlation that occurs when the ion interacts simultaneously with several electrons.[14] On the whole, the performed calculations and estimates account correctly for the main regularities of the energy straggling of the light ions, but do not provide a quantitative description of the straggling. To improve the theory we need both more accurate calculations of the contributions of the various mechanisms to the total energy-loss straggling, and new systematic experimental data.

The present study was made for the purpose of obtaining experimental data on the energy straggling of H^+ and He^+ ions in solids at energies near the maximum of the stopping power, i.e., under conditions when the contribution of the above-cited additional mechanisms of the energy-loss straggling is most noticeable. Preliminary results of measurements made on some of the targets were published earlier.[7,9]

2. EXPERIMENTAL METHODS AND TECHNIQUE

The measurements were made with H^+ and He^+ ions in the energy ranges 120–480 and 150–1300 keV, respectively. The ions were normally incident on the target film surface and were registered, after passing through the target, with a surface-barrier silicon detector. The latter was placed on the beam axis and ensured registration of the particles scattered within an angle $\theta \sim 1°$, so that the contribution of the elastic collisions to the energy-loss straggling was negligible.

The source of ions with energy up to 500 keV was an accelerator with a cascade high-voltage generator. The system for the analysis and shaping of the beam made it possible to apply to the target an ion beam with a diameter from 0.1 to 0.5 mm, with intensity $\sim 1 \cdot 10^3$ particles/sec, and with an approximate energy straggling 0.5 keV. The energy spectra of the primary-beam ions and of the ions that have passed through the target were measured two or three times in succession for each value of the initial energy E_0. The spectrometer resolution was 7–8 keV for protons and 11–13 keV for He^+ ions.

The measurements with the higher-energy He^+ ions were made with a 72-cm cyclotron by a previously described procedure.[6] The spectra of the ions of the primary beam and of the beams passed through the target were registered in this case simultaneously. The overall resolution of the apparatus was 25–30 keV.

The targets were unsupported films of aluminum, copper, silver, gold, and polystyrene $(C_8H_8)_m$. The metallic films were made by evaporating the pure metals in vacuum (at a pressure $\sim 5 \cdot 10^{-5}$ Torr) on cleaved surfaces of rock-salt crystals, followed by dissolution of the substrate in distilled water. The polystyrene films were obtained from a solution of the polymer in dichlorethane on the surface of well cleaned polished glass and were removed from the latter likewise with distilled wa-

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ter. The films selected for the tests were those without visible defects observable in an optical microscope with magnification 56×. An analysis of the metallic film samples by electron microscopy revealed no noticeable preferred orientation of the crystalline blocks and showed that the depths of the surface roughnesses could range from several dozen (Al, Au) to two or three hundred (Cu, Ag) angstrom.

The target thicknesses t were determined from the average energy losses $\overline{\Delta E}$ of the protons and He^+ ions with respective energies ~300–500 and ~800–1000 keV on the basis of the known data on the stopping powers dE/dt of the investigated materials.[15-21] The relative errors in the values of t (without allowance for the errors of dE/dt) were 1–2%. The differences between the values of t obtained for one target from the energy losses of the protons and of the He ions did not exceed 2–4%.

The measurements were made with films of thickness from ~0.1 to 1.1 μm, satisfying the "intermediate target thickness" criteria[11]: $\overline{\Delta E} \gg T_{max} = 4(m_e/M_1)E_0$ and $\Omega \ll E_0 - \overline{\Delta E}$, where $\overline{\Delta E}$ and Ω are the mean values of the energy losses and of their variances, while T_{max} is the maximum energy imparted by an ion of mass M_1 to an immobile electron of mass m_e. The energy distributions $I(E)$ of the ions passing through such targets should have an approximately Gaussian shape, as was in fact observed in the measurements. Only in the spectra of the He ions having the very lowest energies was a small "tail" observed on the lower-energy side, attesting to an increased contribution of the elastic collisions; their influence, however, was not noticeable in the main part of the spectrum. No distortions capable of indicating the presence of particle channeling in the spectrum shape, was observed at all on the high-energy side.

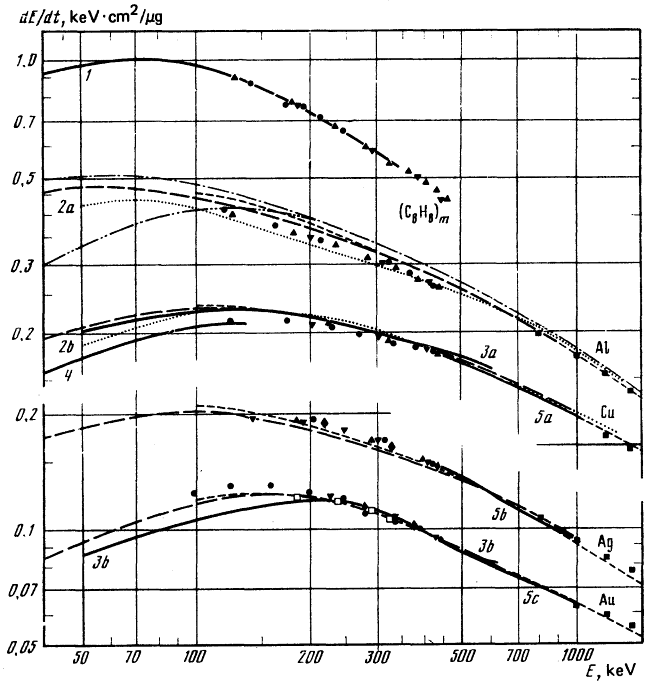

FIG. 1. Specific energy losses of hydrogen ions: 1 and □—Ref. 18, 2a and 2b—Ref. 15, 3a and 3b—Ref. 16, 4—Ref. 22, 5a, 5b, and 5c—Ref. 17, ■—Ref. 23, ●, ▲, ▼, ◆—present work (various targets). Curves with long and short dashes—results of the calculations of Refs. 30 and 29, respectively. Dash-dot curves with single and double dots—results of calculations of Ref. 32 without and with allowance for the change of the effective charge of the proton, respectively.

The obtained spectra were approximated by the Gaussian functions $I = I_{max} \cdot \exp[-(E-E_m)^2/2\Omega^2]$. The ion energy losses in the target ΔE_{en} and their variances Ω^2_{en} were calculated from the formulas

$$\overline{\Delta E}_{en} = E_{m\,0} - E_{m\,1}, \quad (1a)$$

$$\Omega_{en}^2 = \Omega_1^2 - \Omega_0^2, \quad (1b)$$

where the subscripts 1 and 0 pertain to spectra obtained with and without the target, respectively. The errors in the values of $\overline{\Delta E}_{en}$ and Ω^2_{en} amounted respectively from 0.5 to 2–3% and from ~5 to 20–30%, and were determined principally by the statistical accuracy of the measurements and by the relative values of the differences in (1a) and (1b). The results of measurements performed with He ions on one and the same target with a cyclotron and with a cascade generator, at close values of the energy, were in good agreement.

The experimental values of the per-unit energy losses ε_{en} and of the per-unit straggling D_{en} of these losses were calculated from the formulas

$$\varepsilon_{en} = dE/dt\,|_{E=\bar{E}} \approx \overline{\Delta E}_{en}/t, \quad (2a)$$

$$D_{en} = d\Omega^2/dt\,|_{E=\bar{E}} \approx \Omega_{en}^2/t, \quad (2b)$$

where $\bar{E} = E_0 - \overline{\Delta E}_{en}/2$. Estimates based on the known relations[16,20] $dE/dt = f(E)$ have shown that the additional error due to the finite thickness of the film did not exceed 1–2% in the most unfavorable cases.

3. RESULTS OF ENERGY LOSS MEASUREMENTS

The experimental values of $\varepsilon_{en}(E)$ for protons and He ions in the investigated materials are shown in Figs. 1 and 2, respectively. It is seen that on the whole they agree with the best known data from other studies,[15-28] which are also shown in the same figures. The obtained $\varepsilon_{en}(E)$ dependences agree, naturally, with those given in Refs. 15–21 at the energies used there to determine the target thicknesses. Outside these intervals they do not always agree with the results of some particular paper, but do not go beyond the limits of the aggregate of the available data. An exception are the results obtained for protons in Au, where the good agreement with the results by others[16-18] in the energy region $E \gtrsim 250$ keV becomes rapidly worse and the discrepancy reaches +17% at $E \sim 100$ keV. The reason for this is not clear and lies apparently in the differences in the microstructures of the employed targets.

Figures 1 and 2 show (dashed) also the results of semi-empirical calculations from the latest papers[29-31] of this kind. It is seen that the values of $\varepsilon(E)$ obtained in Refs. 29 and 30 for protons are in sufficiently good agreement (within 2–5%). They agree also within approximately the same limits with the results of our present measurements in the region $E \gtrsim 250$ keV; at

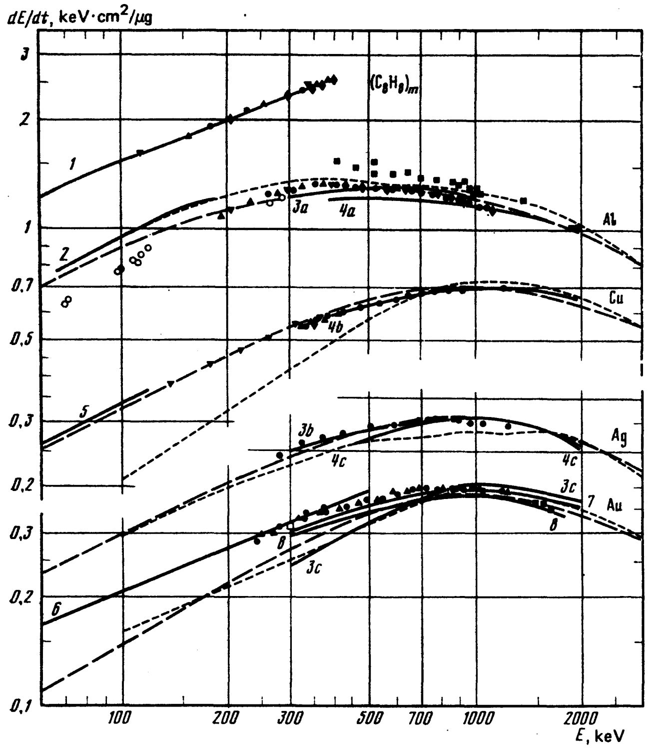

FIG. 2. Specific energy losses of helium ions: 1 and □—Ref. 18, 2—Ref. 26, 3a, 3b, and 3c—Ref. 19, 4a, 4b, and 4c—Ref. 20, 5—Ref. 22, 6—Ref. 24, 7—Ref. 27, ○—Ref. 25, ●,▲,▼,◆—present work. Curves with long and short dashes—results of calculations of Refs. 31 and 29, respectively.

lower energies the difference increases to +9% for Cu and to −7% for Au. In the case of the He ions the agreement between the results of the calculations in Refs. 29 and 31 is worse than for protons. A better agreement with experiment is found in Ref. 31: the difference between the values of $\varepsilon(E)$ cited there and our present results is within ~5, 3, and 6% for Al, Cu, and Ag, respectively. Satisfactory agreement for Au (in the range 4–8%) is obtained only in the region $E \gtrsim 600$ keV; at lower energies the difference reaches ~20%.

A recent paper[32] reports values of the stopping power of Al for protons, calculated within the framework of the dielectric approximation and with allowance for the change of the effective charge Z_{eff} of the particles at low energies. The obtained values of $\varepsilon(E)$, shown in Fig. 1 by a dash-dot line, exceed somewhat the experimental and the semi-empirical values,[29,30] and the method used to take into account the change of Z_{eff} greatly underestimates the values of ε in the region $E < 100$ keV (dash-double dot curve in Fig. 1).

4. RESULTS OF MEASUREMENTS OF THE ENERGY LOSS STRAGGLING

The results of the measurements of the variances of the energy losses are shown in Figs. 3 and 4. The measurements were made with four or five targets of each material [$(C_8H_8)_m$, Al, Cu, Ag, and Au] in the respective thickness ranges 0.118–0.46, 0.22–1.09, 0.29–0.51, 0.17–1.60, and 0.10–0.23 μm. The obtained values of $D_{en} = \Omega_{en}^2/Nt$ revealed considerable straggling—largest for Cu and Ag and smallest for $(C_8H_8)_m$ and Al. The magnitude of this straggling was determined principally by the difference in the values of D_{en} in different targets of the same material (see the data for Al and Ag in Figs. 3 and 4). This is due in all probability to the differences between the degrees of thickness of homogeneity of films produced under approximately identical conditions (see Sec. 2). The smallest values of D_{en} for a given material should obviously correspond to targets that are most uniform in thickness, and should therefore be closest to the "true" values. [Figures 3 and 4 show data for only such $(C_8H_8)_m$, Cu, and Au targets.]

Figures 3 and 4 show also the experimental data obtained by others. It is seen that the results of measurements with protons in Ag[33] and Cu[34] and with He ions in Al and Au (Ref. 5) have a rather large straggling, within the limits of which they agree with our data. The values of D_{en} for He ions in Cu and Ag, cited in Ref. 8, are close at the lowest energy (~500 keV) to those obtained by us, but have a weaker dependence on the ion energy.[1)]

The same Figs. 3 and 4 show the results of calculation in accordance with the existing theories. The values of D_B and D_{LS} were calculated from the known Bohr formulas[11]:

$$D_B = 4\pi Z_1^2 Z_2 e^4 \quad (v_1 \gg v_0 Z_2), \tag{3}$$

and the Lindhard-Scharff formulas[12]:

$$D_{LS} = (m_e/M_1) E \varepsilon_e \quad (v_1 \leq v_0 Z_2), \tag{4}$$

where v_1 is the ion velocity ($v_0 = 2.19 \cdot 10^8$ cm/sec is the Bohr velocity), m_e and M_1 are the respective masses of the electron and ion, ε_e is the "electronic" stopping power of the medium. The values of D_C were taken

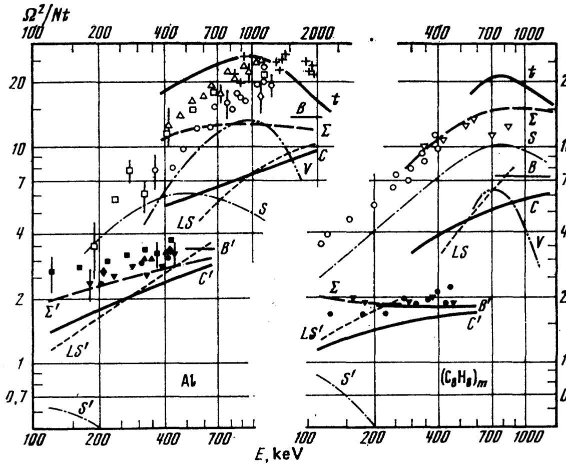

FIG. 3. Energy-loss straggling of ions in Al and $(C_8H_8)_m$. Dark and light symbols—results of present work for the H and He ions, respectively. For Al: $t = 59$ (▲△), 130 (▼), 216 (■□), 272 (◆◇) and 293 μg/cm² (●○); for $(C_8H_8)_m$: $t = 42.8$ (▼▽), and 47.5 μg/cm² (●○). +—data of Ref. 5. The calculated curves $D_B, D_{LS}, \ldots, D_t$ are designated respectively by the letters $B, LS, \ldots, t$ (primed—for protons, without primes—for He ions). The ordinate units are Ω^2/Nt in eV²/10¹² atom/cm².

from Chu's paper.[11] In the high-velocity limit ($v_1 \gg v_0 Z_2^{1/2}$) they go over int D_B. All these theories are based on the model of independent pair collisions of a fully ionized particle of charge $Z_1 e$ with the electrons of the stopping medium; the electrons were either assumed to be free[11] or regarded as an electron Fermi gas of homogeneous[12] or inhomogeneous[10] density.

To take into account the contributions of the additional mechanisms to the total energy-loss straggling, we used the results of Vollmer and Sigmund.[14] The former dealt with the influence of fluctuations of the average stopping power when the charge of the ion is changed as a result of capture or loss of electrons. The resultant additional energy-loss straggling D_V is defined as

$$D_V=\left[\frac{(q_a^2-q_b^2)\,\omega(A)}{(q_a^2-q_b^2)\bar{A}+q_b t}\right]^2 \varepsilon_e t, \tag{5}$$

where q_a and q_b are the dominant values of the ion charge at the given average ion energy, $\bar{A}$ and $\omega(A)$ are the average total path of the ion in the charged state a and the fluctuation of this quantity on a trajectory segment equal to the target thickness. The values of $\bar{A}/t$ and $\omega(A)/\bar{A}$ are tabulated in Ref. 13 as functions of the cross sections of the transitions between the states a and b and of the target thickness t.

The other paper[14] deals with the effect of the correlation of the energy losses when the ion interacts simultaneously with several electrons, and the value of the corresponding correction D_S is estimated in first approximation at

$$D_S=\zeta \varepsilon_e^2/4\pi Z_2 a_2^2, \tag{6}$$

where $a_2 = 0.8853 a_0 Z_2^{-1/3}$ is the Thomas-Fermi screening radius ($a_0 = 0.529 \cdot 10^{-9}$ cm is the Bohr radius), and ζ is a numerical factor that depends on the distribution of the electrons in the atoms of the medium and is close to unity in value.

In the calculations by formulas (3)–(6), a value $Z_2 = 7$ was assumed for the polystyrene $(C_8H_8)_m$. (For targets of intermediate thickness this is valid if it is assumed that $D \sim Z_4$ and that the collisions of the ion with the C and H atoms are independent.) For ε_e we used the experimental values of ε_{en} shown in Figs. 1 and 2, since the contribution of the elastic collisions to the energy losses was negligible under the conditions of our measurements. In view of the lack of data on the cross section for the loss and capture of electrons by ions in solids, the corrections D_V were calculated for the closest (in terms of Z_2) gas targets—nitrogen in the case of $(C_8H_8)_m$ and neon in the case of Al. The data needed for this purpose on the cross sections for the loss and capture of electrons by He ions in N and Ne were taken from Refs. 35. The value of the factor ζ in (6) was assumed equal to unity. The total straggling tendency was determined in each case in the form of a sum: $D_\Sigma = D_C + D_S$ and $D_t = D_C + D_S + D_V$.

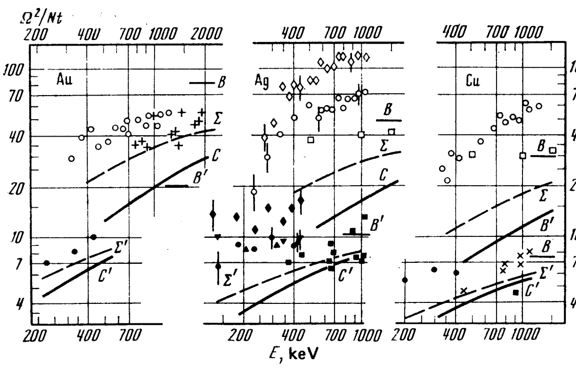

FIG. 4. Energy-loss straggling of ions in Au, Ag, and Cu. The symbols are the same as in Fig. 3. For Au—t = 192 (○) and 430 μg/cm^2 (●); for Ag—t = 176 (▾), 216 (◆), 280 (○), 460 (▴) and 625 μg/cm^2 (●); for Cu—t = 280 (○) and 454 μg/cm^2 (●), +, □, ×, and ■—the data of Refs. 5, 8, 33, and 34, respectively. The markings of the calculated curves are the same as in Fig. 3.

As seen from Figs. 3 and 4, the experimental values of D_{en} generally exceed the values calculated by the traditional theory of the "single-electron" energy straggling in its most elaborate variant.[10] For protons in $(C_8H_8)_m$, Al, and Au this excess is relatively small (~25–50%) and decreases gradually with increasing particle energy. The values of D_{en} measured for the He ions in the same materials turn out to be larger than the theoretical ones (D_C) by 2–2.5 times, and in the cases of $(C_8H_8)_m$ and Al they exceed the "Bohr limit" D_B. The large difference observed in the cases of Cu and Ag for both protons (by 1.7–2.5 times) and He ions (by 4–5 times) is due apparently to the large thickness inhomogeneity of these targets (see Sec. 2).

Allowance for the corrections for the additional energy-straggling mechanisms in accord with Eqs. (5) and (6) improves appreciably the agreement between theory and experiment. The fact that these formulas yield only estimates does not permit a rigorous assessment of the degree of quantitative agreement, but does explain the behavior of the experimental $D_{en}(E)$ plots. As seen from Figs. 3 and 4, for protons in the investigated energy range the principal role is played by the mechanism of the "single-electron" energy straggling. The influence of the fluctuations of the ion charge is negligible here, since capture of an electron by a proton at these energies in unlikely. On the other hand, the contribution of the "correlation" mechanism of D_S becomes noticeable with decreasing proton energy, especially in the region $E \lesssim 200$ keV; when this contribution is taken into account, the theoretical $D_\Sigma(E)$ plots agree fairly well with the measured ones for the most homogeneous $(C_8H_8)_m$, Al, and Au targets.

The picture is different for the He ions. Both additional mechanisms—"correlation" and "charge-exchange"—play a substantial role in a rather wide range of energies in the region of the maximum of $\varepsilon(E)$, and may even be dominant [see the $D_S(E)$ and $D_V(E)$ curves for $(C_8H_8)_m$ and Al in Fig. 3]. Thus, the summary $D_\Sigma(E)$ and $D_t(E)$ dependences can have maxima in this energy region and can exceed the "Bohr limit" for high velocities (D_B). The existence of such a maximum in the energy region ~1200–1600 keV is indicated by the results of our measurements for Al and Au in conjunction with the data of Ref. 5.

5. CONCLUSION

Our measurements yielded data on the dependences of the specific energy losses $\varepsilon(E)$ and their variances $D(E)$ for H and He ions in polystyrene $(C_8H_8)_m$, Al, Cu, Ag, and Au in the important energy region near the maximum of the stopping ability of the material.

The obtained $\varepsilon_{en}(E)$ dependences agree generally well with the results of measurements made by other in overlapping ion-energy ranges, with the exception of the case of protons in gold at energies ≤ 200 keV.

The experimental data on the straggling of the energy losses of protons with energy < 400 keV and He ions with energy < 500 keV in solids were obtained here for the first time ever. The result obtained by others in the energy intervals investigated by us[23,24] or in adjacent intervals[5,8,33] agree with ours within the scatter of the experimental points. The obtained data offer evidence that in the region of the maximum stopping power of the material the energy straggling of the ions is strongly affected by the "charge-exchange" and "correlation" mechanisms. Whereas for the protons, in the investigated energy range, the contribution of the first of these mechanisms is small, while the contribution of the second constitutes a relatively small increment to the "usual" single-electron energy straggling, the influence of these mechanisms becomes dominant for the He ions. As a result, the value of D increases substantially and can exceed the Bohr limit at high velocities. The values of D_{en} for He ions turn out to be larger by a factor 6–8 than those for protons having the same velocity (in the region 150–300 keV/nucleon), i.e., D_{en} increases more rapidly than $\sim Z_1^2$ with increasing Z_1.

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1) These values of D_{en} are averages of several values for targets of various thicknesses; the scatter of these values is not indicated.

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