Energy-loss distributions of 50–250-keV protons traversing thin solid foils: Determination of the skewness coefficient and influence of foil roughness

N. E. Capuj, J. C. Eckardt, G. H. Lantschner, and N. R. Arista
Instituto Balseiro and Centro Atomico Bariloche, C.N.E.A., RA 8400 San Carlos de Bariloche, Argentina
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From new measurements of energy-loss distributions of proton beams in thin solid foils we determine the second- and third-order moments of the energy spectra, and evaluate the skewness coefficient γ, which is of interest in studying the asymmetry of the energy-loss spectra. We present calculations of γ based on electron gas and Thomas-Fermi models; we compare these calculations with the experimental results and with the values derived from Bohr's energy-loss treatment. We present a theoretical analysis of the effects of surface roughness on the skewness of the energy-loss distributions.

When a monoenergetic ion beam traverses a thin solid foil, the ions are slowed down and dispersed in energy. A well-studied quantity pertaining to this process is the average energy loss, or stopping power. To obtain further information about the energy-loss process it is of interest to analyze not only the mean energy of the distribution, but also its shape,[1–4] which can be represented through the higher-order moments of the distribution. Studies of the energy spread (straggling) have already given interesting information on energy-loss fluctuations. Moreover, recent theoretical and experimental work[5–7] stimulated further interest in studies of higher-order moments. In particular, the difference between the mean and the most probable energy loss is related to the third-order moment of the distribution.[5–7]

We present in this letter first experimental results of the skewness coefficient of energy-loss distributions for proton beams traversing thin solid foils, as well as theoretical results based on existing models. We finally analyze the influence of foil roughness on the value of this coefficient.

According to the Landau-Vavilov theory[1,2] the shape of the energy-loss spectrum for thin targets should be asymmetric towards the side of higher energy losses. For thicker targets, the shape should approach the Gaussian form as predicted by Bohr[3] (however, if the thickness is sufficiently large, so that the energy lost in the medium is not a small fraction of the initial energy, the shape of the distribution becomes again asymmetric[8]). The case considered in this work corresponds to the thin-target case, with low deviations from the Gaussian form. The asymmetry of the distribution can be well represented by the "skewness coefficient"

$$\gamma = \langle \Delta E^3 \rangle / \langle \Delta E^2 \rangle^{3/2} , \tag{1}$$

where $\langle \Delta E^2 \rangle$ and $\langle \Delta E^3 \rangle$ are the second and third moments of the energy-loss distribution, defined as $\langle \Delta E^n \rangle = \langle (\langle E \rangle - E)^n \rangle$.

The experimental setup has been described in a previous paper.[9] An ion beam from a 300-kV Cockroft-Walton accelerator is magnetically selected in mass and collimated by two 1-mm-diam. diaphragms to $\pm 0.05°$. Immediately behind the target holder a pair of electrostatic deflection plates permits an angular analysis of the beam. The particles are detected by a plastic scintillator and a photomultiplier tube, the angular acceptance of the detector being 0.05°. The energy analysis is performed by a 90° cylinder-sector electrostatic analyzer. The measured energy resolution of the whole system, including the energy spread of the incoming beam, was maximal 0.5%. The targets are self-supporting, 2-mm-diam. thin films prepared in this laboratory by evaporation, under clean vacuum conditions, on a plastic film that is later dissolved. To avoid foil thickening effects by ion bombardment,[10] fresh foils and low ion doses (10^{-8} C/mm^2) were employed, and the target chamber was trapped by liquid-nitrogen traps at its entrance and exit. The foil thickness was estimated from the measured energy loss, using standard stopping-power tables.[11]

To obtain the values of the second and third moments of the distribution from the primary experimental data, one must take into account the finite spread of the incident beam. This was accomplished through the analysis of the moments of the incident and emergent energy distributions, $F_1(E)$ and $F_2(E)$, of the proton beams. If we denote by $F_0(E,E')$ the distribution corresponding to the "ideal" experiment using a monoenergetic incident beam of energy E', then F_1 and F_2 are related by

$$F_2(E) = \int dE' F_1(E') F_0(E,E') . \tag{2}$$

Using this relation we can deduce the moments of F_0, after careful evaluations of the moments of F_1 and F_2. To evaluate the moments of F_1 and F_2 from the experimental distributions, the spectra were smoothed by a five-neighbor geometric weight function and noise was subtracted. To correct for the fact that the moments were determined from a finite-energy-range spectra (not ranging from $-\infty$ to $+\infty$), we analyzed the incidence of this truncation using Edgeworth distributions of similar parameters; the corrections to the moments obtained in this way were applied to the experimental data. In all the cases we found that the experimental spectra could be well represented by the Edgeworth distributions corresponding to the corrected moments.

The theoretical analysis is based on the electron gas formalism, in terms of Lindhard's dielectric function $\epsilon(q,\omega)$. Using this approach, the energy-loss moments Q_l for a particle of charge Z_1e and velocity v can be written as an integral over the wave vector q and the frequency ω, in the form[12,6]

$$Q_l \equiv \frac{(Z_1 e\omega_p)^2}{v^2} L_l(n,v) = \frac{2Z_1^2e^2}{\pi\hbar v^2}\int \frac{dq}{q}\int_0^{qv} d\omega(\hbar\omega)^l \operatorname{Im}\left[\frac{-1}{\epsilon(q,\omega)}\right] . \quad (3)$$

The calculation of these integrals, using suitable approximations for $\epsilon(q,\omega)$,[12] permits us to obtain the following analytical expressions for the cases of low velocities ($v \ll v_F$):

$$L_l(n,v) \cong \frac{3}{l+2}(4E_F)^{l-1} \times \left[\frac{v}{v_F}\right]^{l+2}\int_0^1 \frac{z^{l+2}dz}{[z^2+\chi^2 f_1(z)]^2} \quad (4)$$

(where the integrals can be expanded analytically), as well as for high velocities ($v \gg v_F$) (cf. also Refs. 6 and 12)

$$L_1(n,v) \cong \ln\left[\frac{2mv^2}{\hbar\omega_p}\right] - \frac{3}{5}\left[\frac{v_F}{v}\right]^2 ,$$
$$L_k(n,v) \cong (mv^2)^{k-1} \quad \text{for } k>1 . \quad (5)$$

Here v_F and ω_p are the Fermi velocity and plasma frequency.

The treatment of an atomic system with nonuniform electron density $n(r)$ follows the local-density approach (LDA) first introduced by Lindhard and Scharff[13] for stopping-power calculations and used later in various energy-loss studies[14–18] (we remark, however, that the use of the LDA in these cases should be considered as a phenomenological approach). The local atomic average of the electron gas moments are then given by

$$\langle Q_l \rangle = \frac{(Z_1 e)^2}{v^2}\int d^3r\, \omega_p^2(r) L_l(n(r),v) , \quad (6)$$

where $\omega_p^2(r) = 4\pi n(r)e^2/m$, and $n(r)$ was approximated using the Thomas-Fermi model. Finally, the expected values of the first three moments of the energy loss, for a foil with atomic density N and thickness $\bar{x}$, are given by the relation[3,5] $\langle \Delta E^l \rangle = N\bar{x}\langle Q_l \rangle$, $l = 1,2,3$. The linearity of $\langle \Delta E^2 \rangle$ and $\langle \Delta E^3 \rangle$ with the thickness $\bar{x}$ has the immediate consequence that the skewness coefficient γ, Eq. (1), decreases with $1/\sqrt{\bar{x}}$ (the linearity with $N\bar{x}$ applies only for the first three moments as already shown in Ref. 5).

We show in Fig. 1(a) the results for the energy straggling for proton beams transmitted through a 180-Å foil of Cu (the values are represented here, for experimental convenience, in terms of the width at half maximum W). Figure 1(b) shows the results for the skewness coefficient γ as given by Eq. (1).

As a reference we write down the values of the second and third moments according to Bohr's high-energy approximation[3]

$$\langle \Delta E_B^2 \rangle \equiv \alpha_B \bar{x} = 4\pi Z_1^2 Z_2 e^4 N\bar{x} , \quad (7)$$

$$\langle \Delta E_B^3 \rangle \equiv \beta_B \bar{x} = 4\pi Z_1^2 Z_2 e^4 N\bar{x} mv^2 , \quad (7')$$

and hence the skewness coefficient in this approximation becomes

$$\gamma_B = \frac{\beta_B}{{\alpha_B}^{3/2}} = \frac{mv^2}{(4\pi Z_1^2 Z_2 e^4 N\bar{x})^{1/2}} . \quad (7'')$$

We find from Fig. 1(a) a very good agreement between

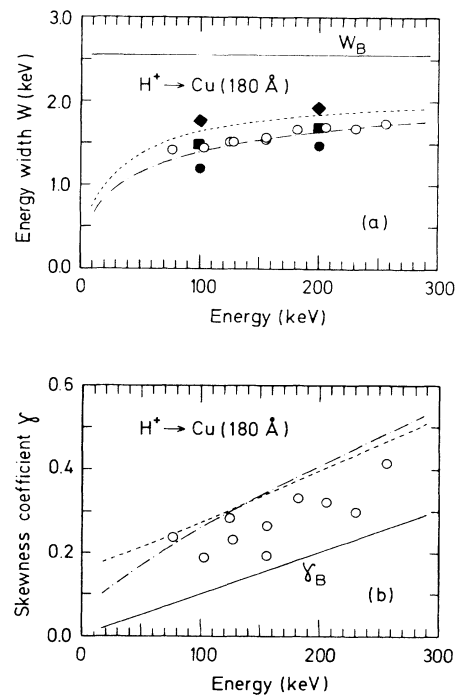

FIG. 1. (a) Energy with W ($W^2 = 2.355 \langle \Delta E^2 \rangle$) of the energy-loss distribution for protons in Cu as a function of projectile energy. Present measurements, ○. Calculations by Chu (Ref. 16) for the cases of (1) uniform foil, ●; (2) Ref. 16, including 10% foil roughness effect, ■; (3) Ref. 16, including 15% foil roughness effect, ♦. Prediction of the Bohr high-energy approximation, ——. Present calculations, - - - -; and same including 10% foil roughness effect, — — —. (b) Skewness coefficient $\gamma = \langle \Delta E^3 \rangle / \langle \Delta E^2 \rangle^{3/2}$ for the same case. Present measurements, ○. Calculations using Bohr's high-energy approximation, ——. Present calculations, - - - -; same, including 10% foil roughness effect, — — —.

the values of the energy width W obtained from the experiment, from our calculations, and also from the calculations by Chu.[16] At these energies the straggling is much smaller than in Bohr's approximation, Eq. (7), which corresponds to a width $W_B = 2.55$ keV.

Our calculations of both energy straggling and skewness coefficient approach the Bohr values at very high energies as should be expected, but in our energy range the model yields a smaller straggling and larger asymmetries ($\gamma > \gamma_B$) than obtained from Bohr's approximation. The experimental values for the skewness coefficient γ, Fig. 1(b), are also larger than γ_B. We also observe that the experimental results for γ fall between the predictions of these models, as Fig. 1(b) illustrates. This same behavior was found for the various foils used in this experiment.

Let us now analyze the influence of foil thickness fluctuations[19,20] on the values of $\langle \Delta E \rangle$, $\langle \Delta E^2 \rangle$, and $\langle \Delta E^3 \rangle$. For a discussion of these effects we follow a previous approach by Chen, Laubert, and Brandt.[20] Let $f(E,x)$ be the energy distribution of emerging protons for an ideal foil of uniform thickness x, and let $P(x)dx$ denote the actual fraction of particles traversing a thickness between x and $x+dx$. Then we can calculate the following combined averages:

$$\langle \Delta E \rangle = \int \int E f(E,x) P(x) dE\, dx = S\bar{x} \tag{8}$$

$$\langle \Delta E^2 \rangle = \int \int [\langle E(\bar{x}) \rangle - E]^2 f(E,x) P(x) dE\, dx = \alpha \bar{x} + S^2 \langle \delta x^2 \rangle \,, \tag{8'}$$

$$\langle \Delta E^3 \rangle = \int \int [\langle E(\bar{x}) \rangle - E]^3 f(E,x) P(x) dE\, dx = \beta \bar{x} + 3\alpha S \langle \delta x^2 \rangle + S^3 \langle \delta x^3 \rangle \,, \tag{8''}$$

where $\bar{x} \equiv \langle x \rangle = \int x P(x) dx$, $\langle \delta x^l \rangle = \int (\bar{x} - x)^l P(x) dx$, and we have used the relation $\langle E(\bar{x}) \rangle = \langle E(x) \rangle + S\delta x$ to separate the terms of the integrals conveniently. The terms $S\bar{x}$, $\alpha\bar{x}$, and $\beta\bar{x}$ are the expected values of $\langle \Delta E \rangle$, $\langle \Delta E^2 \rangle$, and $\langle \Delta E^3 \rangle$ for a uniform foil of thickness $\bar{x}$. We consider now the case of low thickness fluctuations and, from the previous relations, we expand the skewness coefficient as follows:

$$\gamma = \frac{\langle \Delta E^3 \rangle}{\langle \Delta E^2 \rangle^{3/2}} \cong \gamma_0 \left[1 + \left[\frac{3\alpha S}{\beta} - \frac{3}{2} \frac{S^2}{\alpha} \right] \frac{\langle \delta x^2 \rangle}{\bar{x}} \right] , \tag{9}$$

where $\gamma_0 = \beta\bar{x}/(\alpha\bar{x})^{3/2}$ is the skewness expected for a uniform foil, and we assume that the term in $\langle \delta x^3 \rangle$ can be neglected for the present discussion. Thus we find that the skewness γ could be larger or smaller than γ_0, depending on the sign of the correction term. We can characterize this result by the parameter

$$\eta = \frac{2\alpha^2}{\beta S} = \frac{2\langle \Delta E^2 \rangle_0^2}{\langle \Delta E \rangle \langle \Delta E^3 \rangle_0} \,, \tag{10}$$

where the second expression gives η in terms of the values $\langle \Delta E^l \rangle_0$ corresponding to a perfectly uniform foil. The thickness fluctuations would then produce an increase in the asymmetry if $\eta > 1$ and a decrease if $\eta < 1$. It is interesting that this criterion is independent of both the thickness $\bar{x}$ and its fluctuation $\langle \delta x^2 \rangle$.

Let us evaluate the parameter η with the Lindhard model. For this purpose we write the values of S, α, and β in the form

$$S = (\alpha_B / mv^2) G_1(v), \quad \alpha = \alpha_B G_2(v), \quad \beta = \alpha_B mv^2 G_3(v) \,,$$

with $\alpha_B = 4\pi Z_1^2 Z_2 e^4 N$. In terms of some appropriate energy-loss functions $G_l(v)$ we readily find the expression

$$\eta = \frac{2G_2^2(v)}{G_1(v) G_3(v)} \tag{11}$$

(notice also that this is independent of the coefficient $4\pi Z_1^2 Z_2 e^4 N$). In particular, for the electron gas model we write $G_1 = L_1(v)$, $G_2 = L_2(v)/mv^2$, and $G_3 = L_3(v)/(mv^2)^2$ in terms of the L functions of Eq. (3), and we find again the simple relation $\eta = 2L_2^2/L_1 L_3$. It can be seen that this becomes a decreasing function of v. In fact, for low velocities, using the corresponding expressions for $L_l(v)$, Eq. (4), we find that $\eta > 1$ through the range of valence electron densities of all the solid elements, whereas for high velocities the corresponding approximations yield values $\eta < 1$. In particular, using the Bohr results for α_B and β_B, Eqs. (7) and (7′) and

$$S_B = 4\pi Z_1^2 Z_2 e^4 N \ln(2mv^2/I)$$

(with the quantum expression for the Bethe logarithm) we find

$$\eta_B \cong 2/\ln(2mv^2/I) < 1 \,.$$

Notice in Fig. 1(b) that the transition $\eta > 1$ to $\eta < 1$ (curve crossing) takes place in the energy range covered by our measurements. As a consequence, the skewness coefficient is not very sensitive to foil inhomogeneities in this region.

In order to estimate the magnitude of the roughness of our foils, we calculated the effects on the well-known straggling values of Chu[16] and those arising from our calculations following the above outlined procedure. In Fig. 1(a) one can observe that a good agreement of our W calculations with the experimental values is obtained for zero roughness, whereas the values of Chu fit well with $\langle \delta x^2 \rangle^{1/2}/\bar{x} = 0.1$, (i.e., 10% of foil roughness). The same analysis performed with the other foils yield also values between 10 and 15% of foil roughness.

In Fig. 1(b) we show our skewness coefficient calculations for foils with an estimated roughness of 10% (dashed line). One can observe that the inclusion of roughness effects improves the energy dependence as compared with the experimental data. The remaining discrepancy may indicate the limitation of this model, which is based on currently used approximations.

In conclusion, we present in this work the first experimental determination of the skewness coefficient γ, for proton beams traversing solid foils. Calculations of this coefficient in terms of higher-order moments for the electron gas, and Thomas-Fermi models, are in fair agreement with the experiments. Both experimental and theoretical results for the intermediate energy range (i.e., around the stopping-power maximum) covered in this work yield larger asymmetries than expected from Bohr's approximations. We analyzed the effect of surface roughness on the determination of the skewness coefficient and we find a parameter $\eta = 2\alpha^2/\beta S$ that describes the influence of roughness on this term. As a general behavior we obtain a relative increase of γ at low energies (when $\eta > 1$) and a decrease at higher energies ($\eta < 1$). We conclude that the analysis of the energy-loss distributions, together with the roughness effect evaluation formulas, could provide a useful tool for energy-loss data evaluations and for quantitative studies of foil roughness effects.

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