Variation of peak energy for energy loss with angle of observation

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(Received 22 December 1986)

The energy loss for protons on Al, Cu, and Ag thin foils around the maximum of the stopping have been measured at two different exit angles. The bombardment energy $E_{\rm max}$ where such a maximum occurs is found to depend on the angle of observation, $E_{\rm max}$ being greater when the detector is positioned off beam than when measured downstream in the beam direction. These results are discussed in terms of a varying participation of different electrons in the target. The effect of foil inhomogeneities on $E_{\rm max}$ is also evaluated.

INTRODUCTION

Recent experiments carried out at intermediate and high bombardment energy showed that the inelastic energy loss for light ions traversing thin solid films depends on the angle on which the ions are detected.¹⁻⁶ This phenomenon can be explained by an impact-parameter variation of the mean energy loss in the scattering of the projectile by target atoms.^{7,8}

It is well known, on the other hand, that the stopping of ions in matter shows a single maximum when plotted as a function of the bombardment energy. The position of such a maximum, i.e., $E_{\rm max}$, has been an object of various theoretical speculations, linking it with the electronic properties of the target material.⁹

The purpose of this work is to bring these two subjects together, by studying the angular dependence of $E_{\rm max}$ for protons traversing thin Al, Cu, and Ag foils. Our results indicate that the corresponding $E_{\rm max}$ do exhibit angular variations. A thesis is put forward regarding such an effect as being of a more universal character, intimately related to the very nature of the origin of the maximum of the energy loss.

ENERGY-LOSS MAXIMUM AND ITS DISPLACEMENT WITH OBSERVATION ANGLE

To begin, we will briefly outline various concepts and mathematics associated with the theory of the maximum of the stopping power. In 1975, Brandt⁹ showed that the stopping-power maximum for protons in matter can be calculated from the approximate expression for the energy loss

$$\Delta E = -\Delta x \frac{4\pi e^4}{mv^2} N \int_0^{2mv^2/\hbar} g(\omega) \ln\left[\frac{2mv^2}{\hbar\omega}\right] d\omega , \quad (1)$$

where e and m are the electronic charge and mass, respectively, v is the ion velocity, N is the atomic density, Δx is the distance traveled by the ion, \hbar is the Planck

constant, and $g(\omega)$ is the oscillator strength distribution of the target where ω is the plasma frequency given by $\omega = (4e^2\rho/m)^{1/2}$, ρ being the electronic density.

By maximizing Eq. (1), assuming the Thomas-Fermi model for the atom, he obtained an equation for E_{max} as a function of v_{max} which compares remarkably well with experiments. Here $E_{\text{max}} = E(v_{\text{max}})$ and

$$\frac{d(\Delta E)}{dv}\Big|_{v=v_{\max}}=0.$$

Unfortunately, as soon as one includes the angle of observation into the energy-loss formula (1), as we shall see later, the resulting expression is not as amenable to maximum calculations as Eq. (1). To circumvent this difficulty, we have found it more convenient to follow a path which, although less accurate, will allow us to analyze the influence of the observation angle upon $E_{\rm max}$ without having to recourse to a cumbersome algebra.

Let us follow Brandt once again and rewrite Eq. (1) in terms of the electronic density

$$\Delta E = -\Delta x \frac{4\pi e^4}{mv^2} N \int_{2mv^2 > \hbar\omega} \rho(\mathbf{r}) \ln \left[\frac{2mv^2}{\hbar\omega} \right] d\mathbf{r} . \qquad (2)$$

According to this, ΔE may be viewed as a result of making an average of the stopping cross sections S over all electronic densities in the target atom as

$$\Delta E = -\Delta x \frac{1}{V} \int d\mathbf{r} \rho(r) S(\rho, v) , \qquad (3)$$

where

$$S(\rho, v) = \begin{cases} \frac{4\pi e^4}{mv^2} \ln\left(\frac{mv^2}{\hbar\omega}\right) & \text{for } 2mv^2 > \hbar\omega\\ 0, & \text{otherwise} \end{cases}$$
(4)

and V is the atomic volume, i.e., V = 1/N.

As can be easily seen, S becomes maximum at $v = v_{\max}(r) \cong (\hbar \omega/m)^{1/2}$ which can be also written as

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 $v_{\max} \sim \chi^{1/2} v_F$, v_F being the Fermi velocity of the electronic distribution and $\chi = (e^2/\pi \hbar v_F)^{1/2}$. A clear connection between v_{\max} and either v_F or ρ thus becomes apparent. This result, though of no help to find the maximum of Eq. (2), allows us to draw the following conclusion.

C1: In computing v_{max} for a nonuniform electron system, those electrons placed in a low-density region will tend to shift v_{max} toward lower velocities. The opposite effect is expected from high velocity electrons, i.e., those in a higher electronic density.

Let us now go over the angular variation on the energy loss of ions traversing thin solid films due to impactparameter variation of the mean energy loss in the scattering of the projectile by target atoms. The energy loss for ions transmitted into a well-defined angle with respect to the beam is given by 5^{-7}

$$\Delta E = -\Delta x N \int Q(\mathbf{p}) d\mathbf{p} \frac{F_{\rm MS}(\theta - \phi, \Delta x)}{F_{\rm MS}(\theta, \Delta x)} , \qquad (5)$$

where $F_{\rm MS}$ is the multiple-scattering angular distribution and $Q(\mathbf{p})$ represents the average energy loss in a single scattering, characterized either by the impact parameter \mathbf{p} or the scattering angle $\boldsymbol{\phi}$.

Equation (5) can be cast into a form more suitable to compare with Eq. (2). To this end, we can calculate $Q(\mathbf{p})$ by integrating the stopping cross section along a straight line with impact parameter p, as¹⁰

$$Q(\mathbf{p}) = \int_{-\infty}^{\infty} dz \,\rho(\mathbf{r}) S(\rho, v) , \qquad (6)$$

with $r = (p^2 + z^2)^{1/2}$, where $S(\rho, v)$ can be replaced by approximation (4), thus obtaining

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$$\Delta E = -\Delta x \frac{4\pi e^{4}}{mv^{2}} N$$

$$\times \int_{2mv^{2} > \hbar\omega} \rho(\mathbf{r}) \ln\left[\frac{2mv^{2}}{\hbar\omega}\right] d\mathbf{r} \frac{F_{\rm MS}(\theta - \phi, \Delta x)}{F_{\rm MS}(\theta, \Delta x)} \quad .$$
(7)

One can easily see now that this expression would give the same result as (2) if $F_{\rm MS}(\theta - \phi, \Delta x) / F_{\rm MS}(\theta, \Delta x) = 1$. This equation, however, does not hold in general. According to well-known features of the functions $F_{\rm MS}$ and $\phi(\mathbf{p})$, we can anticipate that $F_{\rm MS}(\theta - \phi, \Delta x) / F_{\rm MS}(\theta, \Delta x) =$ 1 only asymptotically as $|\mathbf{p}| \rightarrow \infty$. Meanwhile, as $|\mathbf{p}| \rightarrow 0$ the corresponding scattering angle becomes increasingly large so that $F_{\rm MS}(\theta - \phi, \Delta x) / F_{\rm MS}(\theta, \Delta x) \rightarrow 0$.

As pointed out in Ref. 7, $F_{\rm MS}(\theta - \phi, \Delta x)/F_{\rm MS}(\theta, \Delta x)$ weights each impact parameter according to the probability for the ion to leave the target with angle θ after undergoing a scattering angle ϕ somewhere in the foil. It thus follows that if $\theta = 0$, small impact parameters would be strongly disfavored while as θ increases, more and more violent collisions will become allowed. These conclusions, which were already announced in previous publications,^{6,7} can be rephrased in terms more suitable for the purpose in this work as follows.

C2: As the observation angle θ increases, there will be inner electrons contributing to the stopping which are

rarely or almost not active at all when $\theta = 0$.

Notice that C2 does not imply linearity. According to our model, the number of inner electrons excluded of being active on the stopping in the case of $\theta = 0$ are those located within the relatively small spatial region defined by the smallest impact parameter accessible by the ions emerging at $\theta = 0$. As soon as θ turns greater than, say a few degrees, the excluded region becomes so small that further variation can hardly have an observable effect on the energy loss. At this point we may present a third conclusion, also a thesis of this work, which is nothing else but a straightforward consequence of C1 and C2.

C3: Due to an increasing number of inner electrons involved with the stopping of the ion when increasing the observation angle, on one hand, and according to the effect that inner electrons have on the stopping-power maximum on the other, one can expect that the energy $E_{\max}(\theta)$ at which the stopping curve turns maximum for a given observation angle θ will increase when going from $\theta = 0$ to $\theta > 0$.

EXPERIMENTAL PROCEDURE

The experimental setup has been described in detail elsewhere.^{5–7} Nevertheless, let us mention that the whole system consists of a 25-300-kV Cockroft-Walton accelerator producing a proton beam which is mass analyzed and highly collimated before striking the foil. Just behind the foil, two electrostatic deflector plates bend the ion trajectory back to the original beam direction. Finally, the ions are detected after being energy analyzed. Figure 1 shows a sketch of the experiment.

The angles are measured with an uncertainty of 0.05° while the energy is determined with an accuracy of 0.05%. The foil thicknesses were estimated from the energy losses using stopping-power tables,¹¹ being approximately 210, 170, and 140 Å for Al, Cu, and Ag, respectively.

FOIL INHOMOGENEITIES

When the foil does not have a well-defined thickness, there is an additional contribution Ω_r to the energy-loss straggling which is proportional to the stopping-power and the foil thickness variations as demonstrated elsewhere.¹² This term <u>can be written</u> in the form $\Omega_r = \Delta E R_g$, with $R_g = \frac{[\delta(\Delta x)]^2}{\Delta x^2}$, $\overline{\Delta x}$ being the aver-



FIG. 1. Schematic experimental setup. IB represents the mass-selected ion beam; D1,D2,D3, the diaphragms; BM, the beam monitor; T, the target; Sh, the electron shield; DP, the deflection plates; FC, the Faraday cup; S, the slits; Sc, the scintillator; PM, the photomultiplier; and ESA, the electrostatic analyzer.

age thickness, and $\overline{\delta(\Delta x)^2}$ the standard deviation of the (unknown) thickness distribution of the foil, i.e., $p(\Delta x)$. The measured energy-loss straggling Ω_{ex} can be thus written as

$$\Omega_{ex} = [\Omega_0^2 + (\Delta E R_g)^2]^{1/2} , \qquad (8)$$

where Ω_0 is the straggling corresponding to a foil without any roughness.

In order to estimate the uniformity of the foils used in this experiment, we compare our measured energy-loss straggling with Eq. (8), where Ω_0 is replaced by the commonly accepted values calculated by Chu.¹³ This leads to a straightforward determination of R_g , which amounts to 0.10, 0.05, and 0.15 for Al, Cu, and Ag, respectively.

RESULTS

Figure 2 depicts the results of our energy-loss measurements for protons on Al, Cu, and Ag foils at emer-



FIG. 2. Energy loss for protons traversing (a) Al, (b) Cu, and (c) Ag foils, respectively, as a function of the ion energy for two projectile emergence angles, $\theta \neq 0^{\circ}$ (solid circles), and $\theta = 0^{\circ}$ (open circles).

gence angles $\theta = 1.42^{\circ}$, 1.50°, and 1.50°, respectively, and at $\theta = 0$ for the same foils. The $\theta \neq 0$ angles have been chosen so that in all cases the energy loss at those angles was well in the "plateau" of the energy-loss versus angle curve.¹⁻⁷ There we can observe that, for all targets, the energy at which the maximum of the stopping takes place is slightly greater for $\theta \neq 0$ than for $\theta = 0$.

In Fig. 3 we plot the difference $\Delta E(\theta \neq 0) - \Delta E(\theta = 0)$ as a function of the ion energy. There we can see such differences increasing almost linearly around the corresponding E_{max} . This way of plotting clearly indicates that the maximum of the energy-loss results shifted toward higher energy for $\theta \neq 0$ as compared with that of the forwardly transmitted ions. This will occur everytime there is an increase of $\Delta E(\theta \neq 0) - \Delta E(\theta = 0)$ with E in the region of the maximum, independently of the origin of this increase.

Theoretical results are also depicted in the same figure. They are obtained by using Eq. (5), where the en-



FIG. 3. Difference between the energy loss at $\theta \neq 0$ and $\theta = 0$, respectively, for (a) Al, (b) Cu, and (c) Ag targets. Solid line, theoretical calculations.

ergy loss Q is calculated assuming both the local-density approximation and a Thomas-Fermi atom.¹⁰ Moreover, the Thomas-Fermi atom has been confined into a sphere of radius r_0 in order to take into account the restricted volume in the solid.¹⁴ The value of r_0 is determined by $4\pi r_0^3/3 = m_p A/\rho_0$, m_p being the proton mass, A the atomic mass, and ρ_0 the mass density of the target. If this solid-state effect is not taken into account, the calculated E_{max} 's are much lower than the real ones. It can be seen in Fig. 3 that measurements and calculations compare remarkably well.

In order to evaluate the effect of foil roughness on $E_{\max}(\theta=0)$ and $E_{\max}(\theta\neq0)$, firstly we calculate the incidence of thickness inhomogeneities on the angular effect. A similar study¹⁵ has recently been done by using a Monte Carlo technique, without inclusion of correlation between scattering angle and energy loss.

Here we follow an analytic approach consisting in the calculation of the $\Delta E(\theta, \Delta x)$ [Eq. (5)] averaged over a thickness distribution $p(\Delta x)$ with mean value $\overline{\Delta x}$, through the expression

$$\Delta E(\theta, \overline{\Delta x}, R_g) = \frac{\int P(\Delta x') F_{\rm MS}(\theta, \Delta x') \Delta E(\theta, \Delta x') d\Delta x'}{\int P(\Delta x') F_{\rm MS}(\theta, \Delta x') d\Delta x'},$$
(9)

where $F_{MS}(\theta, \Delta x')$ (Ref. 16) account for the transmission probability at exit angle θ , of a projectile through a foil section of thickness $\Delta x'$. Then we deduce the E_{max} shifts caused by this modified angular dependence. The results corresponding to the previously deduced roughness coefficients and assuming a Gaussian foil thickness distribution, are included in Table I. There one can observe that no E_{max} shift due to roughness results when measuring at $\theta=0$. Meanwhile, some additional shifts on $E_{max}(\theta\neq 0)$ do appear, although they can only account for no more than 25% of the experimental E_{max} shifts. In Table I we show the $E_{\max,expt}$ values obtained by finding the maximum of a fourth-degree polynomial which has been previously fitted to the experimental results. The maximum energy loss as well as the error of E_{\max} determinations are also shown in the same table. This latter quantity is obtained by propagating the uncertainties of the energy loss all along the fitting and maximizing procedure.

The results indicate that $E_{\rm max}$ clearly depends on the observation angle and, in our case, the foil roughness effect is, at the most, 25%. It should be mentioned that the influence of variations of the effective path with the angle, i.e., that is, actually followed by the ions and which is greater than the thickness of the foil,⁵ is negligibly small and has no practical effect on these measurements.

SUMMARY

It follows from our rather cursory theoretical development contained in the beginning of this work that when observing the energy loss for ions transmitted through a thin foil and within a well-defined angle of emergence, not all scattering angles are possible, or if preferred, not all impact parameters are as likely as if there were no such conditions. This certainly imposes a restriction over the "kind" of collisions undergone by the ions detected in such experiments. In particular, the occurrence of small impact parameters are highly diminished when observing the ions in the forward direction. Meanwhile, an increasing probability of small impact parameters are expected for those ions detected at larger emergence angles.⁷ In consequence, the ions transmitted off beam have followed trajectories where the electron density results in average slightly higher than that corresponding to the ions ejected around $\theta = 0$. It is therefore expected, according to the correlation existing between electronic density and E_{max} announced earlier in this work, that the maximum of the energy loss must be

TABLE I. $\Delta E(E_{\text{proj}} = E_{\text{max}})$ represents the energy-loss maxima; E_{max} , the projectile energies of maximum energy losses; $\sigma E_{\text{max,expt}}$, the standard deviations of the E_{max} determinations; $\delta E_{\text{max,expt}}$ ($\theta \neq 0, \theta = 0$), the differences between $E_{\text{max,expt}}$ ($\theta \neq 0$) and $E_{\text{max,expt}}$ ($\theta = 0$); $E_{\text{max,theor}}$, the present calculations of projectile energies of maximum energy losses for uniform foils; δE_{max} ($\theta \neq 0, \theta = 0$), the differences of calculated maximum positions for uniform foils; $E_{\text{max,theor}}^{\text{rough}}$, the calculated maximum positions for 10, 5, and 15% rough Al, Cu, and Ag foils, respectively; and $\delta E_{\text{max,theor}}^{\text{rough}}$ ($\theta \neq 0, \theta = 0$), the differences between calculated maximum positions for rough foils.

	Foil material								
		Al			Cu			Ag	
θ (deg)	0		1.42	0		1.50	0		1.50
$\Delta E(E_{\text{proj}}=E_{\text{max}})$ (eV)	2610		2720	3590		3750	2930		3130
$E_{\rm max, expt}$ (keV)	72		80	116		133	102		122
$\sigma E_{\rm max, expt}$ (keV)	1.6		4.0	4.0		4.0	3.9		9.3
$\delta E_{\max, expt}(\theta \neq 0, \theta = 0)$ (keV)		8			17			20	
$E_{\rm max, theor}$ (keV)	76		81	123		132	108		121
$\delta E_{\text{max,theor}}(\theta \neq 0, \theta = 0)$ (keV)		5			9			13	
$E_{\rm max,theor}^{\rm rough}$ (keV)	76		82	123		132	108		124
$\frac{\delta E_{\max,\text{theor}}^{\text{rough}}(\theta \neq 0, \theta = 0) \text{ (keV)}}{\delta E_{\max,\text{theor}}(\theta \neq 0, \theta = 0)}$		6			9			16	

shifted towards higher energies for ions transmitted off beam than for those exiting around the beam direction.

It is also worthwhile mentioning that our observations are in agreement with a previous publication where the maxima of the stopping power are found to be correlated with electronic structure of the stopping media, leading to oscillations of $E_{\rm max}$ with the atomic number of the target.¹⁷

In summary, when measuring the energy loss for well-collimated proton beams traversing thin foils, the resulting stopping curves have their maxima at energies which depend on the exit angle of the ions. Such an effect can be attributed to a slightly different electronic environment seen by ions exiting at different angles.

ACKNOWLEDGMENTS

This work has been partially supported by the Argentine Consejo Nacional de Investigaciones Cientifícas y Técnicas (CONICET), Grant No. PID 3-042700/85.

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