Thin-film inhomogeneities studied by energy-loss measurements using ion beams

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A method to evaluate thin-film thickness inhomogeneities is presented. It is based on the measurements of the first two moments of the energy-loss distributions of swift ions traversing thin foils at two different beam incidence angles. We apply the method to a set of thin (~200-Å) aluminum foils, resolving thickness fluctuations of the order of 10%.

I. INTRODUCTION

It is known that in various beam-foil experiments the inhomogeneities in foil thickness and atomic density may have a distorting influence on a series of measurements. This occurs mainly when very thin target foils are used. As examples, we mention the appearance of increased angular effects in the energy loss of swift light ions in thin solid films,[1,2] or the appearance of an additional straggling[3–6] on the ion energy distributions after traversing the foils.

It is well known that the foil inhomogeneity plays an important role in a series of other areas in physics. Some techniques that can provide information on this aspect of thin films are the following.

(a) Scanning electron microscopy, yielding surface topography information with resolutions of about 10 Å in height and 100 Å in a lateral sense.

(b) High-resolution transmission electron microscopy and related techniques, such as microdensitometer analysis of electron micrographs of surface replicas, yielding information about both lateral and height distributions of surface structures.[7]

(c) Glancing monochromatic x-ray reflection,[8,9] which gives information on electronic density, mean thickness of the film, and both height and lateral dimensions of roughness distributions.

(d) Tunneling electron microscopy; this technique gives the best information about surface topography but provides no information on bulk inhomogeneities.

(e) Stylus techniques; these give information about topography with a resolution of a few tens of angstroms in height, although thickness variations that occur over distances smaller than the stylus size (~10 000 Å) cannot be detected.

Most of these techniques make the foils unusable for transmission beam-foil experiments. This is due to the analysis procedure itself and also because of the mounting requirements.

Here we present an alternative method of analysis specially suited for ion-beam experiments. It is based on the measurement of the mean energy loss and the energy-loss straggling of ions after traversing thin foils.

II. THEORETICAL CONSIDERATIONS

As is well known, when an ion beam traverses a thin solid foil, the ions are slowed down and dispersed in energy due to the statistical nature of the energy-loss mechanism. So, even for a monoenergetic incident beam, one obtains an energy distribution at the exit of a foil. If the foil presents inhomogeneities, such as thickness and atomic-density fluctuations, an additional energy broadening and change of the distribution shape will arise because different ions of the beam may traverse different foil sections, leading to different energy losses.

The energy straggling $\langle \Delta E^2 \rangle$, defined as the second moment of the energy-loss distribution, can be written in terms of the straggling coefficient Ω_0^2, the stopping power $S=\langle \Delta E \rangle / \langle t \rangle$, and the foil thickness fluctuations $\langle \delta t^2 \rangle$ as[3,4]

$$\langle \Delta E^2 \rangle = \Omega_0^2 \langle t \rangle + S^2 \langle \delta t^2 \rangle \ . \tag{1}$$

The first term of Eq. (1), which corresponds to a *uniform* foil of thickness $\langle t \rangle$, will be referred to as the *intrinsic* straggling.

If one places the foil at different angles θ_f with respect to the ion beam, the intrinsic and inhomogeneity contributions to the straggling vary in different ways with θ_f, providing information about the relative contributions of each one.

Let us write the formulas corresponding to this situation, i.e., ions of energy E, traversing a rough foil of mean thickness $\langle t \rangle$ and roughness coefficient ρ, mounted with its normal at an angle θ_f with respect to the ion beam axis. The roughness coefficient ρ is a parameter introduced to characterize the foil thickness inhomogeneity, and is defined as $\rho = \sigma_t / \langle t \rangle$, where $\sigma_t = \langle \delta t^2 \rangle^{1/2}$ is the variance of the thickness distribution. Since the average thickness traversed by the beam is now $\langle t \rangle / \cos\theta_f$, the mean energy loss and the energy straggling for inhomogeneous foils tilted at an angle θ_f, are given by

$$\langle \Delta E(\theta_f) \rangle = S \frac{\langle t \rangle}{\cos\theta_f} \ , \tag{2a}$$

$$\langle \Delta E^2(\theta_f) \rangle = \Omega_0^2 \frac{\langle t \rangle}{\cos\theta_f} + \rho^2 S^2 \left[\frac{\langle t \rangle}{\cos\theta_f} \right]^2 , \tag{2b}$$

while the intrinsic straggling $\langle \Delta E_0^2(\theta_f) \rangle$ varies with angle θ_f as

$$\langle \Delta E_0^2(\theta_f) \rangle = \Omega_0^2 \frac{\langle t \rangle}{\cos\theta_f} \ . \tag{3}$$

Rewriting Eq. (2b) in terms of the full width at half maximum $W(\theta_f)$ of the energy-loss distribution,

$$W^2(\theta_f)=(8\ln 2)\langle \Delta E(\theta_f)^2\rangle \ ,$$

and of the energy loss $\langle \Delta E(\theta_f)\rangle$, one obtains

$$W^2(\theta_f)=(8\ln 2)\left[\Omega_0^2\frac{\langle t\rangle}{\cos\theta_f}+\rho^2\langle \Delta E(\theta_f)\rangle^2\right] . \tag{4}$$

Evaluating Eq. (4) for two different foil angles θ_1 and θ_2 and using the relation

$$\frac{\langle \Delta E(\theta_1)\rangle}{\langle \Delta E(\theta_2)\rangle}=\frac{\cos\theta_2}{\cos\theta_1} \ , \tag{5}$$

which arises from the stopping power definition, one obtains the following expression for ρ^2:

$$\rho^2=\frac{1}{8\ln 2}\frac{W^2(\theta_1)\langle \Delta E(\theta_2)\rangle-W^2(\theta_2)\langle \Delta E(\theta_1)\rangle}{\langle \Delta E(\theta_1)\rangle\langle \Delta E(\theta_2)\rangle[\langle \Delta E(\theta_1)\rangle-\langle \Delta E(\theta_2)\rangle]} . \tag{6}$$

This equation yields the roughness coefficient in terms of experimental measurements of the energy losses ΔE and the widths W at two foil tilt angles θ_f. Notice that it is not necessary to measure θ_1 and θ_2 in order to evaluate this expression.

We will now derive some relations which show in a general way the effect of inhomogeneities on the energy straggling when measuring at two different foil tilt angles. From Eq. (3), the intrinsic widths $W_0(\theta)$ for angles $\theta=\theta_1,\theta_2$ satisfy the relation

$$\frac{W_0^2(\theta_1)}{W_0^2(\theta_2)}=\frac{\cos\theta_2}{\cos\theta_1} . \tag{7}$$

According to Eqs. (2a), (2b) and (4) we have

$$\frac{W^2(\theta_1)}{W^2(\theta_2)}=\frac{1/\cos\theta_1+\rho^2S^2\langle t\rangle/(\Omega_0^2\cos^2\theta_1)}{1/\cos\theta_2+\rho^2S^2\langle t\rangle/(\Omega_0^2\cos^2\theta_2)} . \tag{8}$$

Regarding Eq. (8) as a function of $\rho^2S^2\langle t\rangle/\Omega_0^2$, where $0\le\rho^2S^2\langle t\rangle/\Omega_0^2<\infty$, it can be readily seen that

$$\frac{\cos\theta_2}{\cos\theta_1}\le\frac{W^2(\theta_1)}{W^2(\theta_2)}\le\frac{\cos^2\theta_2}{\cos^2\theta_1} \tag{9a}$$

or, in terms of the measured energy losses,

$$\frac{\langle \Delta E(\theta_1)\rangle}{\langle \Delta E(\theta_2)\rangle}\le\frac{W^2(\theta_1)}{W^2(\theta_2)}\le\left[\frac{\langle \Delta E(\theta_1)\rangle}{\langle \Delta E(\theta_2)\rangle}\right]^2 , \tag{9b}$$

provided that $\cos\theta_2\ge\cos\theta_1\ge 0$.

Here, the lower limit occurs when $\rho=0$, i.e., a perfect uniform foil. While the upper limit is approached when the intrinsic straggling becomes negligibly small compared to that introduced by inhomogeneities.

This criterion will be useful for the analysis of the experimental data, inasmuch as the measured values are expected to lie in the range predicted by Eqs. (9a) and (9b).

III. EXPERIMENTAL PROCEDURE

The experimental system consists of a 25–250-kV electrostatic accelerator producing ion beams which are mass analyzed and collimated before striking the targets. These are mounted in a multiple target holder, perpendicular to the beam direction or with a tilt angle of $\sim 30°$ with respect to the former position.

After passing the foil, the ions are energy analyzed by an electrostatic analyzer (of 0.5% resolution) and detected with a plastic scintillator in combination with a photomultiplier tube.

The spectra were obtained by sweeping the analyzer plate voltage with a step function, synchronously with the channel of a multiscaler into which the signal pulses were stored.

The energy loss $\langle \Delta E\rangle$ and energy full width at half maximum W were determined after smoothing and substracting the background from the spectra following the method described in Ref. 5.

The targets analyzed through this method were self-supporting, 2-mm-diam films, prepared under clean vacuum conditions following a previously described technique.[10] The thicknesses were evaluated using standard stopping power tables.[11]

We applied the method to approximately 180-Å-thick Al foils. Since at the time of this measurements the target holder did not allow a variation of the foil tilt angle during the experiment, we worked with ten foils from a unique production batch; half of them were mounted perpendicular to the beam, and the other five, tilted near 30° from this position.

IV. RESULTS

Figures 1 and 2 show the $\langle \Delta E\rangle$ and W data obtained for a foil set provided by the same evaporation batch. In

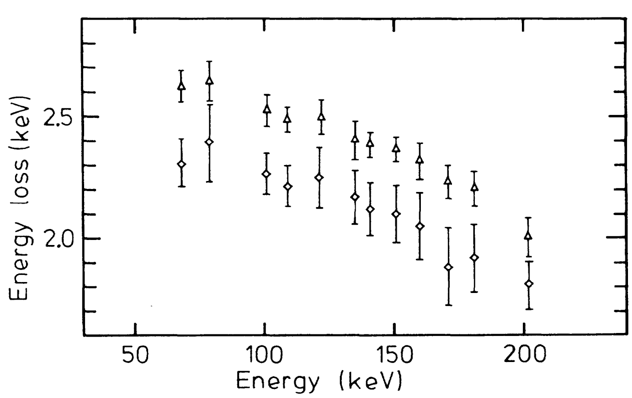

FIG. 1. Energy losses ΔE of protons as a function of projectile energy: $\Delta E(0)$,$\Diamond$; $\Delta E(\theta)$,$\triangle$.

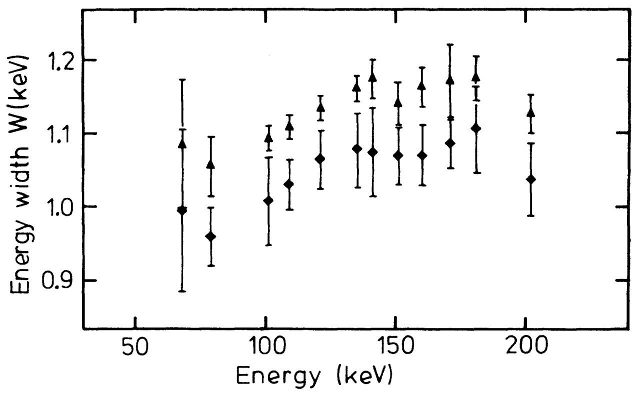

FIG. 2. Full widths at half maximum W of the energy spectra as a function of projectile energy: $W(0)$,♦; $W(\theta)$,▲.

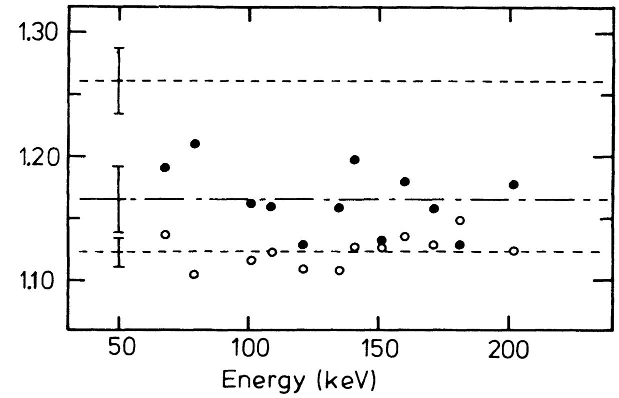

FIG. 3. Energy loss and energy straggling ratios as a function of projectile energy: $\langle \Delta E(\theta)\rangle/\langle \Delta E(0)\rangle$,○; $W(\theta)^2/W(0)^2$,●. The lower and upper bounds for the straggling ratios, Eq. (10), are indicated by the dashed lines. As a visual guide we represent with a dashed-dotted line the mean value of $W(\theta)^2/W(0)^2$ over all energies.

Fig. 3 we can observe that the straggling ratios $W(\theta)^2/W(0)^2$ lie above the ratios $\langle \Delta E(\theta)\rangle/\langle \Delta E(0)\rangle$ for the energy losses; however, following Eqs. (5) and (7) these ratios should be the same in the case of uniform foils. The larger values of $W(\theta)^2/W(0)^2$ are a clear indication of the presence of roughness, as described by the ρ^2 term of Eq. (8).

Additionally, we notice that the straggling ratios $W^2(\theta)/W^2(0)$ in Fig. 3 fall within the range predicted by Eqs. (9a) and (9b), for $\theta_1=\theta$, $\theta_2=0$, viz.,

$$\frac{\langle \Delta E(\theta)\rangle}{\langle \Delta E(0)\rangle} \le \frac{W^2(\theta)}{W^2(0)} \le \left[\frac{\langle \Delta E(\theta)\rangle}{\langle \Delta E(0)\rangle}\right]^2 . \tag{10}$$

Introducing the values of $\langle \Delta E\rangle$ and W for normal and tilted targets in Eq. (6), we obtain ρ values for each projectile energy. Finally, we calculate the weighted average of the roughness coefficient $\langle \rho\rangle$, obtaining $\langle \rho\rangle = 0.12\pm0.06$.

V. COMMENTS AND CONCLUSIONS

An alternative method to evaluate foil roughness was described, suitable for beam-foil experiments and sensitive in a scale of very low roughness. The additional equipment requirement is an ion-energy analyzer and a target holder with provisions for foil tilting. The method has the advantage of allowing nondestructive *in situ* roughness evaluations.

The method yields information about the combined thickness and density fluctuations because the energy loss as much as the energy-loss straggling depend on the amount of matter traversed by the ions. The passages of ions through matter are noncorrelated events, and because the sensing is performed by ion beams of very large diameter compared to atomic scales, the information obtained is an average over the foil region struck by the beam. Therefore, no information can be extracted about the lateral inhomogeneity distribution.

Since ρ is defined as $\rho=\sigma_t/\langle t\rangle$, the resolution is proportional to the thickness, so that we cannot specify an absolute thickness resolution. For instance, for very thin foils ($\sim$200 Å), the analysis is sensitive to foil roughness in the order of $\sim$10 Å.

The best resolution is achieved at projectile energies of maximum stopping power because the increase of W due to roughness increases with ΔE as can be seen from Eq. (1b). It is noticeable that this analysis is independent of theoretical energy-loss models.

The precision of this method can be improved by a full analysis of the tilting-angle dependence for each individual foil. The construction of a new target holder which allows a finite tilting of the foils is currently under way in our laboratory.

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