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Energy straggling of 5.486-MeV alpha particles in Al

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Energy straggling of 5.486-MeV α particles in Al foils has been measured as a function of energy loss using foil thicknesses corresponding to the range $0.01 < \Delta E/E < 0.1$. The thickness uniformity of the foils has been investigated by proton backscattering, electron microscopy, and by mechanical surface profiling. The estimated effect of foil nonuniformity on the straggling results is small. The results are in excellent agreement with the Bethe-Livingston theory.

I. INTRODUCTION

The energy straggling of alpha particles passing through thin solids is a subject that has, until recently, received some theoretical attention but comparatively little experimental consideration. Currently, interest in this subject has been revived due to its importance in determining depth resolution in ion-backscattering analysis of materials.[1,2,3]

The various theories of energy straggling in general give very similar predictions for high-velocity alpha particles traversing thin targets ($0.01 < \Delta E/E < 0.1$). High velocity in this context refers to alpha-particle velocities in excess of the velocity of K-shell electrons of the target atoms ($v/Zv_0 > 1$). In the case of 5.5-MeV alpha particles ($v/Zv_0 \approx 0.56$) traversing thin Al foils, the Bohr,[4] Bethe-Livingston,[5] and the Tschalär[6] theories give straggling results which differ by no more than 15%. Furthermore, the Lindhard and Scharff[7,2,3] theory gives results identical to those of the Bohr theory for this incident energy although predicting lower straggling values than the Bohr theory for lower-velocity α particles.

Sykes and Harris[8] have reported the anomalous energy straggling of 5.486-MeV α particles on their passage through Al foils. They found that for a given energy loss the α particle straggling in Al was greater than in Ag or Cu, contrary to the target atomic-number dependence of straggling predicted by any of the theories referred to above. They also noted that a similar result was obtained by Comfort *et al.*[9] The energy-straggling values reported by both these groups are, in fact, greater than the predictions of the Bohr theory by about a factor of 2.

Furthermore, Harris and Nicolet[1] have commented that in their straggling study of 1- to 2-MeV α particles incident on various materials, only Al showed an energy straggling greater than the Bohr-theory prediction. At this velocity, however, they expected the Lindhard and Scharff[7] theory, which predicts lower straggling values than the Bohr theory, to be in better agreement with their observations.

On the other hand, the straggling of 7.7-MeV α particles in Si has been shown by Avdeichikov *et al.*[10] to be in accord with theory. These authors suggest that as the Z-dependent straggling in Si should be nearly the same as for Al, the previously reported anomalous straggling of α particles in Al probably arose from target nonuniformities. More recently, Strittmatter and Wehring[11] have made a study of the energy straggling of 6.112-MeV α particles in evaporated foils of Al, Ag, and Au. They observed straggling in Al that was much lower than the values obtained by Sykes and Harris and Comfort *et al.* Both of these groups had used commercially rolled Al foils. The Z dependence of Strittmatter and Wehring's result was in reasonable accord with theory. Nevertheless, in some cases their measured straggling exceeded the predictions of the Bethe-Livingston theory, and from this they concluded that their foils had significant nonuniformities.

It is clear from the above discussion that a study of α-particle straggling in Al, using foils whose thickness uniformity has been tested by an independent method, is required to establish reliable measured values of α-particle straggling and hence to test the accuracy of theoretical predictions.

It is generally believed on the basis of energy straggling studies that, at least on a scale less than 1mm^2, foils produced by vacuum evaporation are more uniform than rolled foils. Nevertheless, Abele *et al.*[12] have obtained straggling results which suggest that self-supporting vacuum-evaporated foils have areal density nonuniformities that contribute significantly to straggling. They also found that the magnitude of this contribution depended on the release agent used in the preparation of the foils.

A study of the thickness uniformity of commercially rolled and locally made evaporated Al foils has been carried out at Harwell.[13] The commercially rolled foils were similar to those used in previous measurements of α-particle straggling at about 5.5 MeV.[8,9] This study indicated that nonuniformities in rolled Al foils could account for the previously reported[8,9] anomalous α-particle straggling and that our evaporated Al foils were free from nonuniformities of the magnitude and areal scale found in the rolled foils. Using this information, supplemented by additional foil uniformity studies, we have carried out a further investigation of the straggling of 5.486-MeV α particles in Al foils.

II. EXPERIMENTAL TECHNIQUES

The measurements were carried out using self-supporting commercially rolled Al foils,[14] as used in previous work,[8,9] and vacuum-evaporated Al foils.

A. Foil production and uniformity studies

The evaporated Al foils were produced on glass slides coated with a release agent, RBS-25,[15] which is detergent based. The release agent was applied sparsely over the slide and any excess wiped off with a lint-free tissue. After the evaporation the Al foils were floated off the slides onto distilled water and picked up to cover a 9-mm hole in an Al frame. The foils were not perfectly taut over the holes, but subsequent measurements established that the degree of wrinkling was insufficient to contribute significantly to apparent thickness nonuniformity.

The uniformity of these foils has been studied using several different approaches:

(1) In an earlier investigation of self-supporting Al foils, both rolled and evaporated,[13] relative thickness variations had been obtained from measurements of the relative yield of protons backscattered from them. An areal resolution of about 5×10^{-6} cm^2 was achieved by use of the Harwell microbeam facility. The evaporated Al foils were found on this scale to be uniform to better than ±1% of their thickness. In contrast, the rolled Al foils showed fluctuations varying from 10% to 5% depending on their mean thickness. The influence of slight but visible wrinkling in these self-supporting foils was effectively included by the scanning procedure. It may be noted incidentally that the backscattering technique indicated the presence of oxide layers contributing up to 1% of the foil thickness.

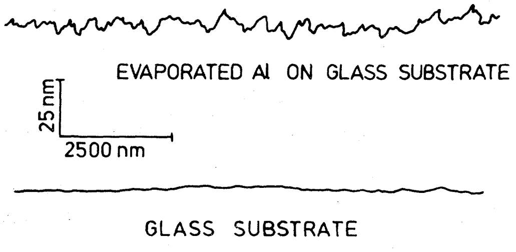

FIG. 1. Surface topography of Al evaporated to 160 μg/cm^2 onto a glass substrate. The substrate surface topography is also shown for comparison. These traces are typical of several made at different points on the surfaces. Estimated visible nonuniformity is ± 30 Å in this example.

(2) The surface topography of evaporated Al foils has also been studied with a Tallystep[16] instrument equipped with a stylus whose tip dimensions were 0.1 by 2 μm, i.e., covering an area of about 2×10^{-9} cm^2. A stylus loading of ≈ 2 mg was used. The topography of the foil surface was measured for Al evaporated onto a clean glass slide free of release agent. The profile of a glass slide was measured separately and found to be sufficiently uniform for its effect to be neglected, as can be seen from Fig. 1. The Tallystep traces indicated surface features with horizontal linear dimensions of about 100 nm while their average vertical size varied from sample to sample between ± 2 and ± 3 nm in foils of thickness $\approx 160\,\mu$g/cm^2 (600 nm). Irregularities less than 100 nm wide were not resolved, and the dimensions of the stylus were such as to lead also to some attenuation for surface features in the range 0.1 to 2 μm. It is therefore estimated that the degree of nonuniformity could be a factor of 2 larger than indicated by the Tallystep traces. This would be equivalent to about ±2% to ±3% mean thickness variation for a 160-μg/cm^2 Al foil if one assumes the structure at both surfaces to be similar but uncorrelated. For the thicker foils used in the straggling studies the percentage thickness fluctuations would be expected to be smaller.

(3) Electron micrographs of the evaporated Al foils were obtained and showed[17] that minute holes occurred in these foils, but that these amounted to at most 2% of the area of the foils. The Al crystallites had faces with mean widths of about 100 nm, which is similar to the length over which the foil topography was found to vary in the Tallystep measurements. There was some evidence of tungsten oxide contaminants arising from the evaporation boat used to prepare the foils, but these only covered about 0.1% of the foil surface.

(4) Measurements of the relative intensities of x-ray diffraction lines showed evidence of a small amount of preferred orientation in the Al crystallites, somewhat stronger for the rolled than for the evaporated foils. The small departure from random orientation was not expected to give rise to any significant channelling effects.

B. Straggling measurements

A vacuum-sublimed ^{241}Am α-particle source, obtained from the Radiochemical Centre at Amersham, was used to provide 5.486-MeV α particles for this straggling study. A self-supporting Al foil was mounted with its surface normal to and intersecting the axis formed by the α source and a 60-μm-thick surface-barrier detector in a vacuum chamber kept at pressures $\leq 2\times 10^{-5}$ Torr. The foil could be interchanged with others or removed from the α-particle beam by driving the mounting rack with a lead screw. A ^{244}Cm α source mounted on the rack was used in conjunction with the ^{241}Am source to provide an energy calibration. The ^{241}Am source and the detector were separated by 5 cm and both were collimated to 2-mm diameter, sufficient to limit the solid angle contribution[18] to straggling to less than 2 keV.

The signals from the surface barrier detector were amplified and processed in a 4096-channel analyzer using a conversion gain of 8192 channels and a 4096-channel back bias. A pulser was used to check that gain drifts in the system were negligible during the periods in which related calibration and straggling measurements were made. The sources used were sufficiently weak to avoid dead-time losses in the electronics which might affect the apparent α-particle energy distribution.

The mean thicknesses of the foils were in each case estimated from the measured α-particle energy loss and the dE/dx values obtained from the compilation of Northcliffe and Schilling,[19] and ranged from about 100 to 1000 μg/cm^2.

III. RESULTS

A typical spectrum of the straggled and nonstraggled ^{241}Am α particles is shown in Fig. 2. In addition to the 5.486-MeV α group, the ^{241}Am source produced weak α groups at 5.443 MeV ($\approx$12%), 5.389 MeV ($\approx$1%), and 5.513 MeV (<1%). In the experiment these were not, in general, resolved from the main group which was used to determine the energy straggling. In order to compare the straggling data with theoretical predictions, it was necessary to estimate the shape of the energy distribution of the 5.486-MeV α group after passage through the Al foil, free from the folded contribution of the instrument function. The latter was assumed to be adequately represented by the 5.486-MeV group of the unstraggled spectrum. Therefore the straggled and unstraggled distributions were analyzed into their various components using line shapes chosen in accordance with theoretical predictions.

A criterion presented by Fano[20] for the occurrence of Gaussian straggling distributions is satisfied for 5.5-MeV α particles traversing foils of the thickness range used for these measurements. The Tschalär theory,[6] which takes account of the increased spread in energy distribution due to change in stopping power across the distribution, predicts a distribution which is very close to a Gaussian in the range of energy loss encompassed by this study ($\Delta E/E\approx 0.01$ to 0.1). The detector response function which includes energy straggling in the source and the detector dead layer, as well as electronic noise, is also expected to be Gaussian.

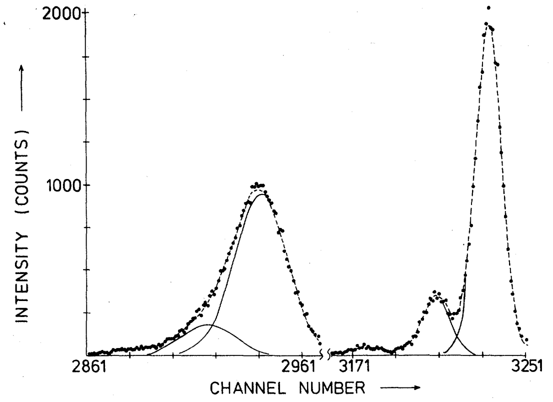

FIG. 2. ^{241}Am α-particle energy distributions measured with a surface barrier detector: on the left is the distribution after passage through an 834-μg/cm^2 Al foil; on the right, without traversing a foil. The dashed curve is the fitted sum of the three components of the distribution. The unbroken curves show the individual components where these differ significantly from the summed dashed curve.

In view of these considerations, Gaussian line shapes were used to fit the observed α-particle distributions, both straggled and nonstraggled. This was done using a nonlinear least-squares program.[21] In each case Gaussians were included for the main α-particle group and for the two more intense secondary groups. The weak 5.513-MeV group was found to give a negligible effect and was disregarded in the analyses. The widths of the main groups thus extracted from the straggled and unstraggled spectra were expressed in terms of their full width at half maximum (FWHM) and the instrument response function was unfolded from the straggled distribution by the subtraction of these widths in quadrature.

The fitting procedure allowed free variation in the height, width, and position of each component Gaussian. An example of the resulting fit to the data is shown in Fig. 2. As a check on the adequacy of the fitting it was noted that the known energy separations of the unstraggled groups were reproduced satisfactorily, as were the somewhat larger separations predicted for the straggled groups in accordance with the energy dependence of stopping power.[19] Moreover, the widths of the three Gaussians required to fit the straggled distribution were found to be approximately the same, and likewise

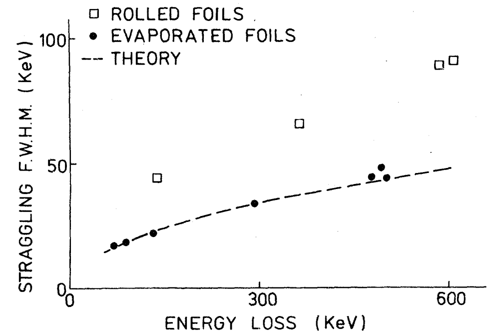

FIG. 3. Energy straggling as a function of energy loss in various thicknesses of rolled and evaporated Al foils for 5.486-MeV α particles. The predictions of the Bethe-Livingston theory are also shown.

for the unstraggled distribution. The effects of tails due to slit scattering could account for the residual difference found in the widths of the small α groups compared with those of the main groups. For these reasons the Gaussian line shape was considered adequate as well as being in reasonable agreement with theory.

The sensitivity of the width estimation to slight changes in line shapes was crudely estimated by fitting skewed Gaussians to the data. The difference in width obtained using Gaussian and skewed-Gaussian line shapes was found to be about ±1%. A calculation was also made of the change in the width parameter of the main component, corresponding to a change in height by the amount the fitted line shape appeared to mismatch the observed data at the peak of the distribution. This led to an estimate of less than 2% for the worst case. We conclude that errors due to line-shape uncertainties are unlikely to exceed ±2%.

The results of several measurements of the energy straggling of 5.486-MeV α particles in various thicknesses of rolled and evaporated Al foils are shown in Fig. 3. Table I lists the measured energy straggling and the estimated foil thickness for both the rolled and evaporated foils.

Errors on the straggling results were estimated as follows. The change in the width parameter of the main component required to produce a statistically significant change in the χ^2 of the fit to the data was estimated using an f test with the appropriate degrees of freedom. This fractional width change was typically about 0.3%, implying an error in the straggling small compared with those from other sources. The line-shape uncertainties considered above contributed about ±2%. The systematic error arising from the small nonuniformities of the evaporated foils discussed in Sec. II is difficult to quantify in general. In the case of the 133-μg/cm^2 evaporated foil in Table I the figures given in Sec. II imply a contribution of from 3 to 5 keV FWHM or a systematic error of +2% to +5% in this straggling result. For the thicker foils this error would be expected to be no greater.

The error in measurement of energy loss, arising from the location of the centroid of the major component in the straggled and unstraggled distributions, is estimated to be less than 1%. A systematic error due to oxide layers at the foil surfaces[19] is also expected to be $\leq 1\%$.

IV. DISCUSSION

It can be seen from Fig. 3 that the α-particle straggling in rolled Al foils considerably exceeds the straggling in evaporated Al foils of the same thickness. This result was not unexpected in view of our previous nonuniformity measurements,[13] which had shown that rolled Al foils were far less uniform than evaporated Al foils.

It is difficult to make a quantitative estimate of the effect on α-particle straggling of the measured nonuniformities in the rolled foils. A crude estimate can be made by assuming the foil thickness fluctuations are normally distributed. A standard deviation of the observed[13] thickness fluctuations in the rolled foils was calculated, and the energy spread (FWHM) was estimated from this standard deviation and the measured α-particle energy loss for the corresponding foil. The result was folded in quadrature with the FWHM α-particle straggling for a uniform foil of the same mean thickness, estimated from the evaporated foil results. The straggling thus predicted for a 258-μg/cm^2 rolled Al foil was 27 keV, about 30% greater than for an evaporated Al foil of the same thickness. The ob-

TABLE I. Energy loss and straggling of 5.486-MeV α particles traversing Al foils.

Energy loss (keV)	Straggling FWHM (keV)	Mean foil thickness (μg/cm^2)
	Rolled Al foils	
138.9	43.9	258
362.6	64.5	644
587.0	88.5	980
603.5	92.1	1013
	Evaporated Al foils	
71.3	16.1	133
91.4	17.9	170
132.2	21.5	247
292.6	32.7	523
479.4	41.4	801
493.6	47.4	829
498.3	43.7	834

served straggling for a 258-μg/cm^2 rolled foil was even larger (44 keV). However the distribution in the magnitude of the thickness fluctuations did not appear to be normally distributed, although it could not be determined accurately from the small sample observed. Moreover the finite resolution of the method may have smoothed the nonuniformities to some extent.

The anomalously high α-particle straggling values obtained by Sykes and Harris[8] using rolled Al foils obtained from the same source that we used are in close agreement with our rolled Al foil straggling results. For example, they obtained an energy-loss value of 215 keV and a straggling width of 48 to 55 keV for a 0.4-mg/cm^2 rolled foil, whereas interpolation of our results gives an energy-straggling value of ≈ 50 keV. One concludes that the so-called anomalous straggling of 5.486-MeV α particles in Al is most likely a spurious result arising from the use of nonuniform rolled Al foils. This conclusion is in agreement with that drawn by Strittmatter and Wehring[11] from a study of 6.112-MeV α particles traversing evaporated Al foils.

We now turn to a comparison of the measured straggling in the evaporated Al foils with the values predicted by the Bethe-Livingston theory,[5] the velocity range covered by this theory being appropriate[22] to 5.5-MeV α particles traversing thin Al foils.

The variance of a straggled energy distribution of Gaussian form is obtained from Bethe and Livingston's expression:

$$\frac{d}{dx}(\sigma^2) = 4\pi z^2 e^4 N\left[Z' + \sum_i k_i \frac{I_i Z_i}{mv^2} \ln\left(\frac{2mv^2}{I_i}\right)\right],$$

where z is the incident particle charge, e and m are the charge and mass of an electron, N is the number of atoms per unit volume of stopping material, x is the depth in the target, Z' is the effective number of atomic electrons of the stopping atoms, I_i and Z_i are the mean excitation energy and the number of electrons in the ith shell of the stopping atoms, and v is the incident particle velocity. The k_i are constants which, following Bethe and Livingston, are all set at $\frac{4}{3}$, a value expected to be too low for high-Z atoms. The summation extends over all shells for which $2mv^2 > I_i$.

The evaluation of a set of I_i follows the treatment of Comfort *et al.*[9] and Sternheimer.[23] The sum rule given by Bethe,[24]

$$Z \ln I = \sum_i f_i \ln I_i ,$$

is rewritten as

$$\sum_{i=1}^{j-1} f_i \ln I_i + f_j \ln(h\nu_p f_j^{1/2}) = Z \ln I ,$$

where the summation extends over all shells except the j orbit which contains the conduction electrons. The plasma frequency of conduction electrons is given by $\nu_p = (NZe^2/\pi m)^{1/2}$. The f_i are oscillator strengths and approximately equal to Z_i. The I_i are replaced by $\rho h\nu_i$, where the $h\nu_i$ are the x-ray critical-absorption energies[25] for the ith shell. The correction factor ρ is then evaluated from the above sum rule using the values of I, the mean ionization parameter, given by Fano.[20]

In the case of 5.486-MeV α particles incident on Al the evaluation of the effective number of electrons Z' is particularly simple as $2mv^2$ is larger than any of the I_i.

The measured values of FWHM straggling as a function of energy loss were compared with the Bethe-Livingston theory by use of the relation applicable to small energy loss,

$$\Omega = 2(2\ln 2)^{1/2}\left[\left(\frac{dE}{dx}\right)^{-1}_{\bar{E}} \frac{d}{dx}(\sigma^2)\right]^{1/2} (\Delta E)^{1/2} ,$$

where $(dE/dx)_{\bar{E}}$ is the stopping power evaluated at the mean energy $\bar{E}$ of the α particle in the Al foil and is obtained by interpolation from the tables of Northcliffe and Schilling.[19] As the straggling distribution is assumed Gaussian, the numerical factor converts the variance to the full width at half maximum. If the Northcliffe and Schilling tables are assumed to be accurate to better than ±5%, the $(dE/dx)_{\bar{E}}$ factor introduces an error of less than ∓2.5% to the predicted straggling.

The calculated values of straggling for 5.486-MeV α particles on Al are shown plotted in Fig. 3 and can be seen to be in excellent agreement with observation. The theory may be expected to diverge from observation for higher-Z targets, in which the α-particle velocity is no longer "fast" for most of the atomic electrons.

The other theories of straggling mentioned in the Introduction give, as would be expected from their neglect of separate shell contributions for this velocity regime, slightly lower values than the Bethe-Livingston theory. The Bohr and Tschalär theories give about 15% lower values than the observations over this energy-loss range.

The recently published results of Strittmatter and Wehring[11] for the straggling of 6.11-MeV α particles in Al, carried out using evaporated foils, may be compared to our results. The straggling is predicted by the Bethe-Livingston theory to be about 2 keV less than in the 5.486-MeV case. However, for an energy loss of 204 keV Strittmatter and Wehring observe a straggling of 41 keV, whilst interpolation of our results gives about 30 keV. They suggest that the discrepancy between their result and the Bethe-Livingston theory arises from foil nonuniformitites, although we note that their reported method of preparation of their evaporated foils seems to be similar to the one we used.

In conclusion, we have found the straggling of 5.486-MeV α particles in Al to be in agreement with the Bethe-Livingston theory. Comparison of our experimental values with other published values suggests that the problem of removing foil nonuniformity contributions to straggling measurements is very important and should be approached by measurements of foil uniformity that are independent of straggling.

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