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STRING EFFECT WITH 5 MeV PROTONS AND 20 MeV ALPHA PARTICLES ON BISMUTH

BY

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Synopsis

The yield of backward elastic scattering (150°) of 5 MeV protons and 20 MeV α-particles from a single crystal of bismuth was found to depend on the orientation of the crystal with respect to the beam direction. When the latter coincides with the trigonal axis of the crystal the yield has a deep minimum, the angular width of which agrees with Lindhard's formula. Smaller and narrower minima in the yield are found when the beam is parallel to low index crystal planes. The effect is found to increase when the crystal is cooled to liquid nitrogen temperature. The string effect decreases with the depth below the surface, at the low temperature, with a factor of two in about 10 μm. The variation is found to correspond to an almost linear increase in Ω^2, the mean square angular width of the beam, which may be understood as a result of the thermal vibrations of the atoms.

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1. Introduction

As first pointed out by LINDHARD[1)], when a narrow beam of charged particles enters a single crystal, the yield of nuclear reactions and scattering processes will have a minimum when a low index crystal axis is parallel to the beam direction. Looking at the crystal in the direction of the axis, one could imagine (with high! magnification) to see the end of the "strings" on which the atoms are sitting like pearls, and between the strings would be empty channels. Rather, the channels would not be completely empty; only the nuclei and some of the inner atomic electrons are located in or near the strings, whereas the outermost electrons may also be found in the channels. One could further imagine that, if charged particles were shot into the crystal in a direction almost exactly parallel to the strings, only nuclei in the front layer could be hit and the particles having passed this layer would travel along the channels without striking the nuclei. Such "channelling" effects may be present if the angle ψ between the beam direction and the axis is very small. However, LINDHARD has shown that even when particles have much larger ψ-values, for which no proper channelling takes place, the Coulomb fields around the nuclei in a string prevent the particles from striking the nuclei, provided only that ψ is smaller than a certain critical angle ψ_1. In fact, the particles cannot even come close enough to the nuclei to undergo large angle Coulomb scattering.

ψ_1 is given approximately by

$$\psi_1 \sim \sqrt{\frac{2Z_1Z_2e^2}{Ed}}, \tag{1}$$

where Z_1 and Z_2 are the charge numbers of the projectile and the target nucleus, respectively, e is the electronic charge, E the C.M. energy, and d the distance between neighbouring atoms in a string. For an incoming particle beam having an angular width smaller than ψ_1 one observes, by varying the crystal orientation, a minimum in the yield of nuclear reactions when the beam direction coincides with the string direction. This was first demonstrated by Bøgh, Davies and Nielsen[2], who studied the yield of the resonant (p,γ) reaction in aluminum using protons with a kinetic energy slightly higher than the 411 keV resonance. In the (1, 1, 1) direction they found a decrease in the γ-yield to about 20 % of the normal value, the angular half width of the dip being in rough agreement with (1).

It was the purpose of the present experiments to investigate whether the string effect could be observed also in the elastic scattering with higher energy particles such as could be obtained from our cyclotron, and furthermore to look for a possible temperature effect.

Studies of the string effect have meanwhile been continued in Aarhus[3] and elsewhere. Dips in yield of γ-radiation and in Rutherford scattered particles have been observed not only for string directions, but smaller dips also when the beam is parallel to a plane in the crystal. For higher energy particles the effect has been observed with 1–2 MeV protons at the Van de Graaff of this Institute and with naturally occurring α-particles in Stockholm[4].

Particles moving parallel to string directions will tend to move at some distance from the nuclei, hence where the electron density is relatively low. Such particles will therefore have a longer range than particles moving at some angle with the crystal axis, an effect which was observed by Davies et al.[5] long before Lindhard predicted the reduction in nuclear reaction yield. Even for particles which are predominantly stopped by electronic encounters, the specific energy loss will be smaller for string directions than normal, an effect which has also been directly observed[6].

2. Experimental apparatus

The arrangement is shown in fig. 1. Part of the analyzed beam from the Copenhagen cyclotron was passed through two lead stops, *Pb*1 with a 2 mm diameter hole and *Pb*2 with a 1 mm diameter hole. The two diaphragms were 420 mm apart, and thus the transmitted beam had an angular half width of 0.0036 radians or 0°.2. A small piece of a *Bi* single crystal, about 3 mm thick, was glued to a hollow copper cylinder *C* and placed close to the axis of the scattering chamber. The crystal was made by cleaving a larger single crystal cooled to liquid air temperature; the surface was parallel to the trigonal plane, as was found by *X*-ray analysis. The copper cylinder was supported by an insulating teflon holder *D* screwed to the brass ring *E*. To increase the heat insulation of the copper cylinder the two axles *F* (only one shown in the figure) were thin-walled stainless steel

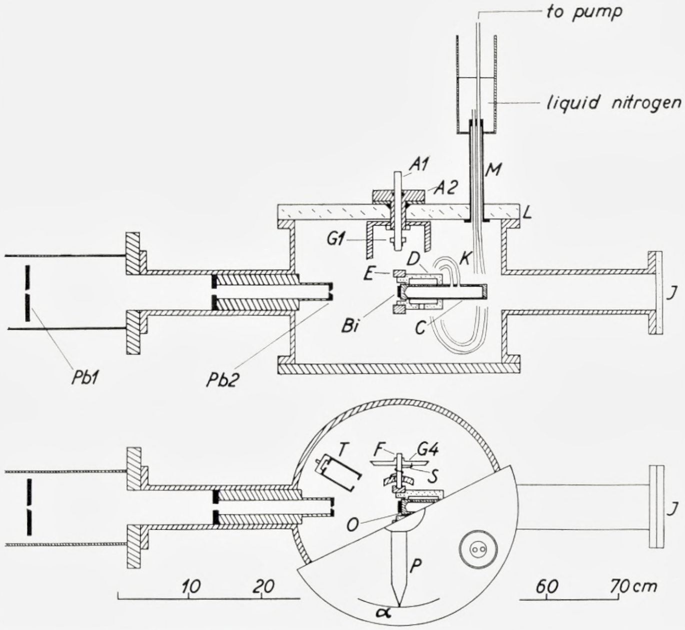

Fig. 1. Upper figure: Vertical section through scattering chamber. Lower figure: To the left, horizontal section and, to the right, top view.

tubes. By rotating the axle $A1$ the copper cylinder could be tilted about F, the connection being made through four gear wheeles, only two of which are shown ($G1$ and $G4$). The setting of the axle $A1$ was read on a duodial; one revolution = 100 divisions would produce a tilt of 12° of the copper cylinder. The axles F rotated in ball bearings and a spring S minimized the uncertainty in the tilting angle β. The holders for the ball bearings were stiffly connected to the outer axle $A2$, the setting of which could be read with an accuracy of 0°.2 on the scale α by means of the pointer P. Thus, the crystal could be rotated about a vertical axis (angle α) and a horizontal axis (angle β). By holding a small lamp at I and looking at the light reflected from the end of the copper cylinder one could predetermine the setting where the crystal surface is perpendicular to the beam with an accuracy of about one degree.

The copper cylinder could be filled with liquid nitrogen from a well-insulated reservoir (insulation not shown). The copper tubes K were thin (1.5 mm inner diameter) and long (each about 50 cm) in order not to impede the movements of the copper cylinder. A thin-walled stainless steel tube M

served to avoid cooling the lucite lid L of the scattering chamber. A copper-constantan thermo-element (not shown) measured the temperature of the copper at O.

The current to the copper cylinder was of the order of $10^{-9} A$. An integrating device determined the dose of α-particles or protons. The particles scattered at 150° were measured by a solid state counter T with an opening angle of $2 \cdot 10^{-3}$ steradians.

3. Results and discussion

In fig. 2 some proton spectra are shown. When the beam enters in a direction *not* coinciding with a string, the shape of the spectrum may be calculated. Neglecting finite energy resolution and straggling, $N(E)$ should be proportional to $\left[(E')^2 \frac{dE}{dx}\right]^{-1}$, where E' is the energy of the protons just before scattering, E is the energy of the protons when escaping from the surface of the crystal, and $\frac{dE}{dx}$ is the stopping power of the crystal for the latter energy. Using the range-energy relations for Pb given by WILLIAMSON and BOUJOT[7] we find that the average decrease in $N(E)$ in the range from 3 to 5 MeV is about 6 per cent per MeV, in agreement with the experimental curves.

$N(E)$ is considerably smaller when the beam is parallel to the trigonal axis. This effect is more pronounced for the cooled target; not only is the difference larger for the cold than for the warm target, but the effect also extends to larger depths in the crystal (smaller E) in the former case. For the upper curves, 4 MeV (3 MeV) corresponds to protons scattered at a depth of 17 μm (32 μm); thus, one channel roughly corresponds to one μm. For the lower curves, the relation between energy and scattering depth is not the same (and not known), since the stopping power for protons moving parallel to the trigonal axis is smaller than the normal value. Neglecting this difference, we find roughly that the string effect decreases by a factor of two in 3 μm or in 10 μm for the warm and the cold crystal, respectively. If the stopping power in the string direction has half the normal value these figures are reduced by about 25 per cent. However, it may be mentioned here that the reduced stopping power produces a change in the curve, which is not just a multiplication of the abscissae, but somewhat more complicated. For example, if a layer of the target were a perfect crystal, whereas for greater

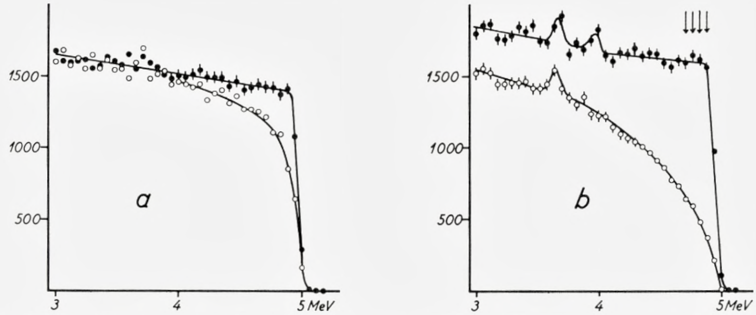

Fig. 2. Spectra of scattered protons, $\vartheta = 150°$. a) Bi-crystal at room temperature. b) Crystal at liquid nitrogen temperature. The upper curves and full circles are obtained with the incoming beam a few degrees off the (1, 1, 1) direction; the lower curves and open circles are obtained when the beam coincides with the (1, 1, 1) direction (the trigonal axis).

depths the atoms were randomly arranged, then the two curves of fig. 2a (or 2b) would never meet, but show a "string effect" for any depth, simply because the particles having passed the crystalline layer in the string direction have the higher energy.

The curves in figs. 2a and 2b correspond to the same measured dose of incoming protons. The ordinates of the two upper curves should therefore be expected to be equal. The reason for the observed difference is unknown. One possible explanation may be that the changing of surface conditions gives rise to different amounts of secondary electrons from the target and, hence, influences the beam dose measurement. There does not seem to be any connection with the thickness of the surface layer of carbon and oxygen, since the corresponding two peaks in the proton spectrum, at energies about 3.6 and 4 MeV, varied quite irregularly in intensity from day to day, whereas the yield difference between a cold and a warm target was reproducible.

Fig. 3a shows the low scattering yield for protons entering the crystal in a direction close to the trigonal axis. When the beam coincides with the axis, the yield for the cold target is reduced to less than half the normal value. A decrease in yield, though smaller, was also observed when the beam was parallel to a low index plane of the crystal (see later). When the angle β is kept constant at the proper value while the angle α is varied (open circles), the beam direction passes through the trigonal axis, but for all values of α the beam is nearly parallel to a $(0, \bar{1}, 1)$ plane, and hence the yield is low also outside the dip. When gluing the crystal to the supporting copper cylinder we aimed at such a position that the α-axis should be parallel to a $(0, \bar{1}, 1)$ plane. Since we were off by some fraction of a degree, the

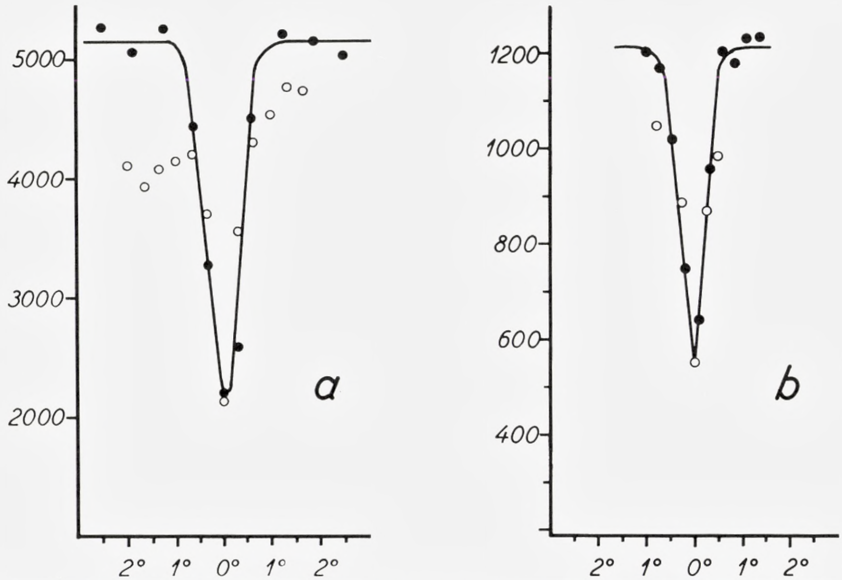

Fig. 3. a) Yield of elastic scattering of 5 MeV protons as a function of the angle between the trigonal axis and the beam direction, for a Bi-crystal at liquid nitrogen temperature. Ordinates: Number of scattered particles in four energy channels (approximately the arrow marked channels in fig. 2 b) per 200 nCoulomb of incoming beam. Full drawn curve and full circles correspond to tilting the crystal in the β-direction, i. e., perpendicular to a $(0, \bar{1}, 1)$ plane. Open circles correspond to a tilting in the α-direction, i. e., parallel to a $(0, \bar{1}, 1)$ plane. b) Similar curves for 20 MeV α-particles.

varying of α does not correspond exactly to travelling along the bottom of the $(0, \bar{1}, 1)$ valley, but it corresponds to a path slantingly climbing the valley side and this accounts for the lack of symmetry indicated by the open points.

In fig. 3b, similar measurements for 20 MeV α-particles are shown. For α-particles the obtainable statistics is poor, and the attention was therefore focussed on the proton measurements.

For the full drawn curves, the angle α is kept constant and the varying of β corresponds to moving perpendicularly to the $(0, \bar{1}, 1)$ plane. As seen in the figure, the shape of the dip is nearly identical for α- and β-tilting, apart from the difference in yield outside the dip. The half widths agree roughly with the theory; the full width at half minimum depth is about $0\overset{\circ}{.}8$ for protons and $0\overset{\circ}{.}6$ for α-particles. Neglecting the finite half width $(0\overset{\circ}{.}2)$ of the beam, we find for the measured half widths Δ of the dips $\Delta = 1.3\ \psi_1$ for both α-particles and protons, where ψ_1 is given by (1).

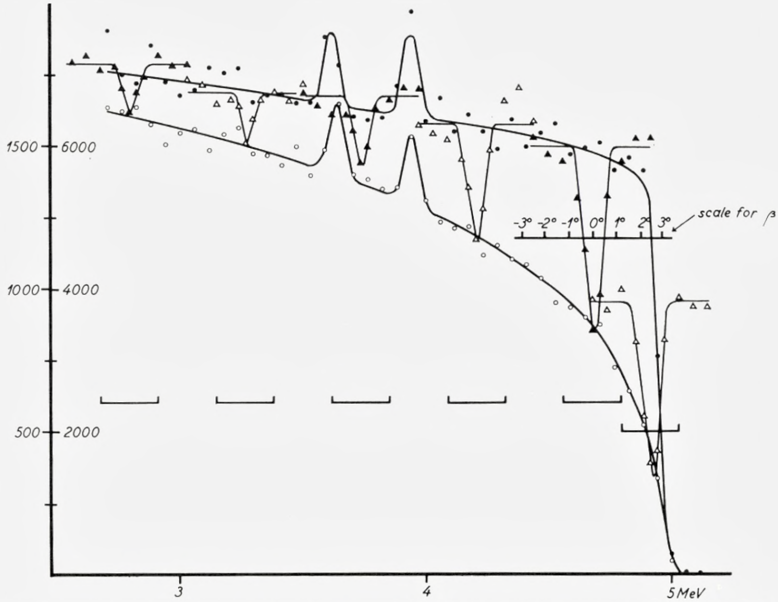

Fig. 4. Spectra of protons scattered 150° from a cold Bi-crystal with the beam coinciding with the trigonal axis (lower curve and open circles) and 2°.4 off this direction (upper curve and closed circles); scale of ordinates to the left of the axis. The variation in yield when tilting the crystal in the β-direction, i.e., perpendicular to the $(0, \bar{1}, 1)$ plane, is shown for several values of the depth of the scattering layer; in each case the number of protons in four energy channels, indicated in the figure, was counted for different β-settings in the vicinity of the trigonal axis; scale of ordinate to the right of the axis.

Fig. 4 shows the result of another measurement which illustrates the declining of the string effect as the protons penetrate deeper into the crystal. Measuring the effect by the percentage decrease in cross section, we again find a reduction by a factor of two in roughly 10 μm. However, the half width of the dip is constant, $\sim$ 0°.8, at least to depths greater than 20 μm; only for the two last curves corresponding to depths of 30–40 μm the half width may be slightly smaller.

Fig. 5 illustrates some planar effects. The curve to the right shows the variation in scattering yield when the crystal is tilted in such a way that the beam direction passes normally across a $(1\ \bar{1}\ 0)$ plane. The three biggest dips correspond to traversing planes of the same order $(1\ \bar{1}\ 0)$, but the two outer dips are wider than the middle one, because the outer planes are crossed at angles of 60° from the normal to the planes. The half widths are 0°.5 for the middle dip, 0°.9 for the other two, the ratio thus being close to

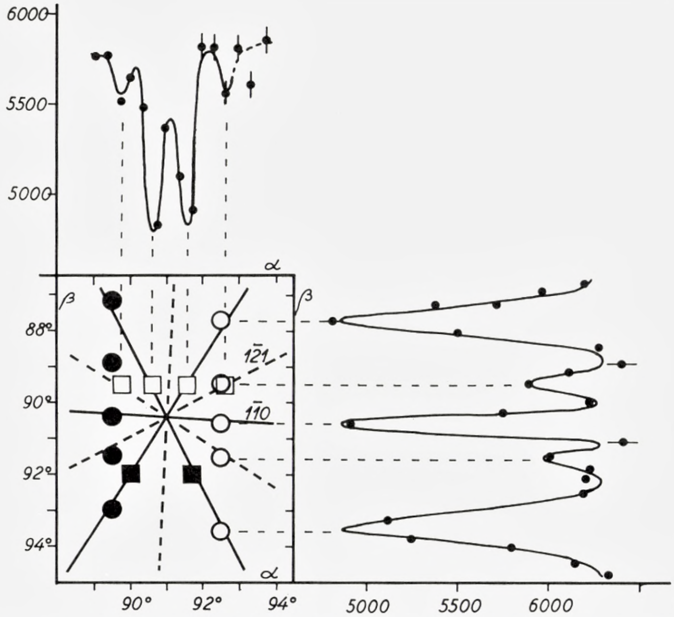

Fig. 5. Map showing positions of some relative minima in scattering yield due to plane effects. When keeping $\alpha = 92^\circ.5$ and varying β, we obtained a yield *vs.* β curve shown to the right, giving minima represented by the open circles in the map. The upper curve corresponds to β constant at $89^\circ.5$ and α being varied; the minima of this curve are represented by open squares. The size of the "points" represents the estimated uncertainty. The absolute values of the ordinates in the two graphs should not be compared, see text.

the expected value cos 60°. As can be seen, the planar dips are narrower than the string dip, in accordance with the theory. However, the ratio is not quite as high as the estimated theoretical factor[8] $2Z_2^{1/6} \approx 4$. In between the big dips are seen smaller dips corresponding to planes of order $(1\ \bar{2}\ 1)$.

The upper curve shows the yield variation when tilting in a direction parallel to a $(1, \bar{1}, 0)$ plane but 3/4 of a degree away from this plane. Again the $(1, \bar{1}, 0)$ planes are clearly seen, and there are also indications of the $(1, \bar{2}, 1)$ planes. At the center, for $\alpha = 91^\circ$, the curve does not rise to "normal" yield, because we are here on the edge of the string hole (cf. fig. 3a).

The two yield curves were not obtained on the same day and since there might be slight changes in beam energy, etc., the absolute ordinates of the two curves are not directly comparable.

Similar curves were obtained for α = const. = $89^\circ.5$, yielding minima shown in the map by full circles and for β = const. = 92° yielding the minima shown by full squares. When drawing straight lines in the proper way through the minima thus located, we get a picture of the planes intersecting in the (1, 1, 1) direction.

For the plane marked $(1, \bar{1}, 0)$ in the figure, the dip observed by perpendicular crossing was followed as far away ($\sim 30°$) from the string as permitted by our apparatus; no appreciable change in the dip was seen.

Each point in the curves of fig. 5 was obtained from a spectrum like those in fig. 2. From these spectra we can obtain the variation of the planar effect with the depth in the crystal in the same way as shown for the string effect in fig. 4. The planar effect decreases with depth somewhat faster than does the string effect, the half length being $\sim 3.5\ \mu$m.

4. Discussion of influence of lattice vibrations

According to LINDHARD, a beam of particles entering a single crystal will be divided into a random and an aligned beam, the latter consisting of the particles having $\psi < \psi_1$ inside the crystal. Particles with $\psi < \psi_1$ entering the crystal at points far away from strings will never come closer to any string than $\sim a = a_0 \cdot 0.88\ [Z_1^{2/3} + Z_2^{2/3}]^{-\frac{1}{2}}$, which in our case is $a = 10^{-9}$ cm. When approaching a string, the particles lose part of or the whole velocity component perpendicular to the string, but after reflection ψ again obtains its original value. If all the incoming particles in the beam have $\psi < \psi_1$, only those striking the surface within the distance a from string positions will go into the random beam and have the possibility of suffering large angle scattering or initiate nuclear reactions. The yield at the dip minimum should therefore be expected to be $Nd\pi a^2 \approx 10^{-2}$ times the normal yield. We shall discuss why we do not observe so low a yield and why the minimum yield increases with depth.

Let us assume that the beam inside the target has a Gaussian shape, the angular distribution being

$$W(\theta)d\theta = \frac{2}{\Omega^2}\theta e^{-\frac{\theta^2}{\Omega^2}} d\theta. \tag{2}$$

The fraction of the particles *not* belonging to the aligned beam is—neglecting the very small contribution $Nd\pi a^2$—

$$\int_{\psi_1}^{\infty} W(\theta)d\theta = e^{-\frac{\psi_1^2}{\Omega^2}}. \tag{3}$$

Furthermore, assume that Ω^2 increases linearly with the distance z traversed, say

$$\Omega^2 = \frac{z}{L} \cdot \psi_1^2, \tag{4}$$

where L is a constant. Then, the fractional scattering yield in the dip minimum is

$$Y = e^{-\frac{L}{z}}. \tag{5a}$$

In figure 6 we have plotted experimental points obtained from the same measurements as the points in figure 2. The ordinate for a point in figure 6 is the number of protons counted in a channel when the incoming beam is parallel to the axis divided by the number counted in the same channel for a "random" crystal orientation (ratio between the two curves in, for example, fig. 2a). The errors shown are the statistical errors; no corrections have been applied for the carbon and oxygen peaks, and they are the cause for the increased scattering exhibited by the points corresponding to the cold target in the region $2.5L < z < 3.5L$.

The points for the cold target can be well fitted by a curve of the type (5a) if we put $L = L_c = 7$ channels $\sim 7\ \mu$m. For reasons which will be outlined in the following, we have chosen to compare the points with a curve of the form

$$Y = S + (1 - S)e^{-L/z}, \tag{5b}$$

where $S = 0.15$, and again a good fit is obtained for $L_c = 7$ channels, as shown by the curve in figure 6.

The points for the warm target fit reasonably well to a curve of the same type (5b), but with $S = 0.50$ and $L_w \sim 1.96$ channels $\sim 2\ \mu$m.

In an amorphous *Bi*-target the nuclear small angle scattering would produce an increase in Ω^2 of the order of ψ_1^2 in one μm, thus giving $L = L_0 \sim 1\ \mu$m. The experimental value $L_c \sim 7\ \mu$m illustrates the reduction in multiple scattering in the cold crystal. In an amorphous target, the electrons would give a negligible contribution to the multiple scattering; the length L_{0e} needed to give an increase of ψ_1^2 in Ω^2 by electronic encounters alone would be $L_{0e} \sim Z_2 L_0 \sim 80\ \mu m$, and in the crystal the corresponding length is probably at least a factor of two larger[8]. The electronic multiple scattering is therefore insufficient to explain the increase in the minimum scattering yield with depth in the crystal.

The difference between a cold and a warm target may suggest that thermal vibrations of the atoms play some role. If, for simplicity, we assume

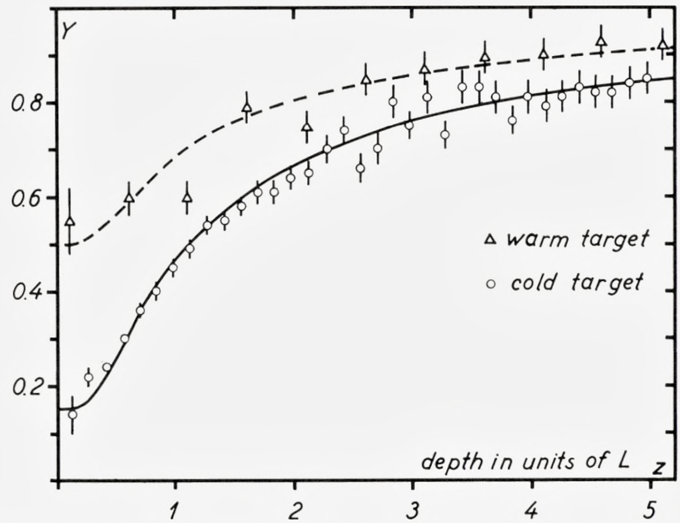

Fig. 6. Scattering yield in string direction relative to the normal yield as a function of the depth in the crystal. The circles and the full drawn curve correspond to the cold target, the triangles and the dotted curve to the crystal at room temperature. The distance between successive points is about one μm. The curves correspond to a linear increase in the mean square angular width Ω^2 of the beam.

isotropy in the oscillations, the displacements ϱ in a plane perpendicular to the string may be assumed to be normally distributed, i.e.,

$$W(\varrho)d\varrho = \frac{2}{\alpha^2}\varrho e^{-\frac{\varrho^2}{\alpha^2}}d\varrho, \tag{6}$$

where α^2 is the mean square displacement given by

$$\alpha^2 = 6T\hbar^2/(mk\theta^2) = 1.0\cdot 10^{-20}\cdot T. \tag{7}$$

Here $k = 1.38\cdot 10^{-16}$ erg/^0K is the Boltzmann constant, T is the absolute temperature, $\theta = 117$ is the Debye temperature for Bi, and m is the mass of the Bi-atom.[9)]

Again as a first crude approximation, the protons in the aligned beam passing through a layer of atoms may be assumed to be randomly distributed with respect to the location of the atoms, except that they do not come closer to any string than a. In their transverse vibrations the atoms will move far out from their equilibrium positions in the strings, and when they are displaced more than a, collisions between the atoms and the particles in the aligned beam may occur. The probability that an atom has a distance larger than a from a string is

$$S = \int_a^\infty W(\varrho)d\varrho = e^{-a^2/\alpha^2}, \tag{8}$$

and this is also the fraction, relative to an amorphous target, of collisions made by the protons in the aligned beam. Therefore, near the surface the minimum fractional scattering yield is $Y = S$ in accordance with (5b). Furthermore, $L = L_0/S$ and, according to (3), (4) and (5a) the fraction of the particles belonging to the aligned beam at the depth Z is $1 - e^{-L/z}$. At this depth we thus expect

$$Y = e^{-L/z} + S(1 - e^{-L/z}) = S + (1 - S)e^{-L/z}. \tag{5b}$$

As we have already seen, the experiments agree well with this formula, and it may be noted that also the ratio L_w/L_c agrees with theoretical expectations. From (7) and (8) we find

$$\frac{ln(1/S_w)}{ln(1/S_c)} = \frac{\alpha_c^2}{\alpha_w^2} = \frac{T_c}{T_w} \sim \frac{90}{300} = 0.3,$$

whereas the experiments give

$$\frac{ln L_w}{ln L_c} \sim \frac{ln 2}{ln 7} = 0.35.$$

In view of the crude procedure leading to (6), (7), (8) and (5b) it is not surprising that these formulae do not agree with the absolute experimental values of S. (7) gives $\alpha_c = 9.5 \cdot 10^{-10}$ cm and $\alpha_w = 1.7 \cdot 10^{-9}$ cm, whereas to obtain the observed S-values from (8) we must introduce the values $\alpha_c \sim 7 \cdot 10^{-10}$ cm and $\alpha_w \sim 1.4 \cdot 10^{-9}$ cm.

In an amorphous target, the Ω^2 corresponding to multiple scattering will increase somewhat faster than linearly with the distance z and, in addition, single and plural scattering will throw particles out of the beam, whose angular distribution therefore deviates slightly from a Gaussian. We may expect a similar behaviour in the crystal. Furthermore, it should be considered that the reduced stopping power in the axial direction influences the points; their ordinates are incorrect, because we compare protons with the same energy when coming out of the crystal, whereas we ought to compare protons having the same energy at the instant of scattering; the points should be corrected upwards.

Thus, although the measurements do not give the accurate yield *vs.* depth function, they do seem to show that the reduced string effect and its variation with the depth may be understood as a result of lattice vibrations. We remark that the finite energy resolution is of no significance, and that the carbon and oxygen layer on the surface also is unimportant, even if the original angular width of the beam is taken into account. Assuming a cross section of 30 times the Rutherford cross section—a value obtained by extrapolating from known data[10)]—the thickness of the layer, estimated from the peaks in fig. 2, is equivalent to about 0.1 mg/cm^2 of oxygen. Such a layer will produce an increase in Ω^2 by about 0.1 ψ_1^2 by small angle scattering.

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