Characteristic X-Ray Production in Single Crystals (Al,Cu,W) by Proton Bombardment. II. Protons of 250 to 1560 keV*

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The study of the effect of proton channeling on the production of characteristic x rays in thick, metallic single crystals has been extended by the use of protons with energy between 250 and 1560 keV. The x-ray yields of Al K, Cu K, Cu L, W K, W L, and W M were measured as a function of the relative orientation of the crystal with respect to the direction of the incident beam. As in the previous study (Part I) minimum values of the x-ray yield were observed when the beam direction lies in crystal planes or along crystal directions having low indices. The relative x-ray yield is presented as a function of the orientation of the crystal planes with respect to the direction of the incident beam. The data indicate that the widths of the resulting curves (yield versus orientation in a plane) depend on the proton energy E_P as 1 $(E_P)^{1/2}$. However, for a given element, the widths do not depend on the particular shell ionized.

I. INTRODUCTION

MANY investigators[1,2] have measured reaction yields that show a dependence on the relative orientation of a single crystal with respect to the direction of the positive-ion beam. This effect is attributed to the confinement (or channeling) between crystal planes of positive ions in their passage through the crystal. The distribution of the trajectories within the crystal depends on the relative orientation of the crystal with respect to the beam. As a result, the reaction yield shows a corresponding dependence. The production of characteristic x rays by protons incident on thick, metallic single crystals is one such reaction, and this paper presents part II of a continuing study of the dependence of the x-ray yield on the relative orientation of a single-crystal target with respect to an incident proton beam.

In Part I,[3] protons having an energy between 70 and 100 keV were used to study the effects of channeling in aluminum and copper. The x-ray yield was measured as a function of the orientation of the [011] direction of the crystal, with the result that a minimum value of the yield was obtained at that direction. A characteristic width, measured at the midpoint between the maximum and the minimum values of the yield, was associated with the curves. It was found that this width exhibited a dependence on the proton energy E_p. In addition, the change of the curves with a change in the contamination of the target was studied.

In this paper (Part II), the investigation of the dependence of the widths on E_p is continued and a study is made of the dependence of the widths on the particular atomic shell initially ionized. Measurements were made with protons of energy up to 1560 keV. A contour of the Cu L x-ray yield for orientations of the [011] direction with respect to the beam for an E_p of 1560 keV is presented for comparison with the contour (E_p= 70 keV) shown in Fig. 5 of Part I. Cu K and Cu L x-ray yields for orientations of the [011] direction and (100) and (111) planes with respect to the beam are presented for a number of proton energies. The Al K x-ray yield for orientations of the [011] direction with respect to the beam is presented for an E_p of 1000 keV. W K, W L, and W M x-ray yields for orientations of a {011} plane with respect to the beam are also shown at various proton energies.

II. APPARATUS AND METHODS

The target chamber, target holder, and general method have been described previously[3,4]; only features specific to the current experimental problems are presented here.

A Van de Graaff accelerator was used to supply a beam of magnetically analyzed H_1^+, H_2^+, H_3^+ ions in the energy range from 750 to 1560 keV; however, the shapes of the yield curves were found to be a function *only* of the energy per nucleon. The beam collimation was defined by the exit slits of the analyzing magnet, which established a maximum beam divergence of $\pm 0.05°$. The target current was always below 1.0 μA.

In contrast to I, the effect from carbonaceous films

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[1] G. R. Piercy, M. McCargo, F. Brown, and J. A. Davies, Can. J. Phys. **42**, 1116 (1964); J. A. Davies, G. C. Ball, F. Brown, and B. Domeij, *ibid.* **42**, 1979 (1964); M. T. Robinson and O. S. Oen, Phys. Rev. **132**, 2385 (1963); L. Lehmann and G. Leibfried, J. Appl. Phys. **34**, 2821 (1963); A. L. Southern, W. R. Willis, and M. T. Robinson, *ibid.* **34**, 153 (1963); O. Almen and G. Bruce, Nucl. Instr. Methods **11**, 279 (1961); R. S. Nelson and M. W. Thompson, Phil. Mag. **8**, 1677 (1963); C. Erginsoy, H. E. Wigner, and W. M. Gibson, Phys. Rev. Letters **13**, 530 (1964).

[2] E. Bogh, J. A. Davies, and K. O. Nielsen, Phys. Rev. Letters **12**, 129 (1964).

[3] J. M. Khan, D. L. Potter, R. D. Worley, and Harold P. Smith, Jr., Phys. Rev. **148**, 413 (1966), hereafter referred to as I.

[4] J. M. Khan and D. L. Potter, Phys. Rev. **133**, A890 (1964); J. M. Khan, D. L. Potter, and R. D. Worley, *ibid.* **139**, A1735 (1965).

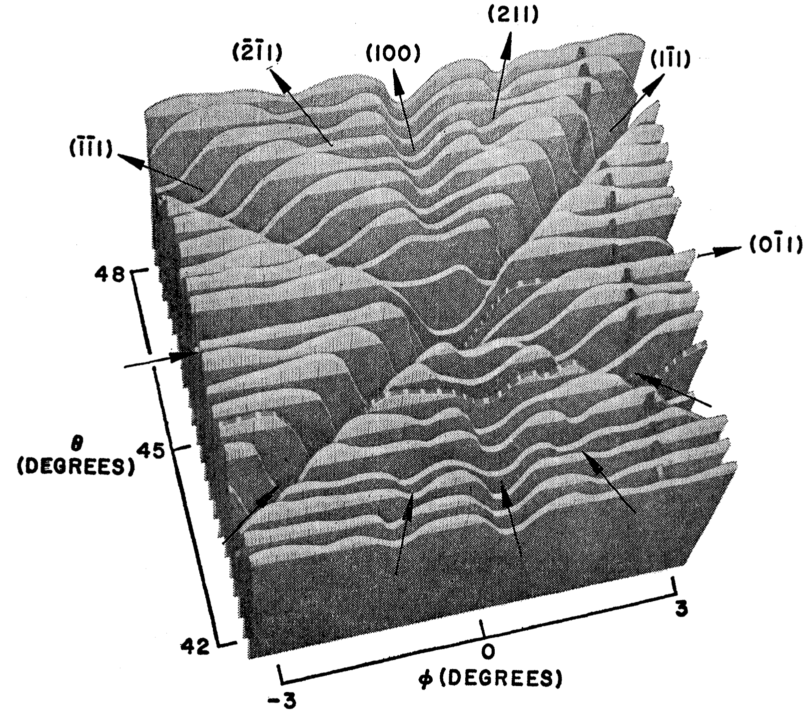


FIG. 1. Copper L-shell x-ray-yield contour for the direction of the proton beam in the region of the [011] direction at $E_p=1560$ keV. Horizontal lines represent the polycrystalline yield. The dashed curves are also presented separately in Figs. 2 and 3 as a function of energy.

was negligible, making periodic removal and cleaning of the target unnecessary. This resulted from two consequences of the use of protons of higher energy: (1) The higher x-ray yields permitted the use of lower beam currents, which reduced the rate of film formation,[4] and (2) the effectiveness of the film in scattering the protons and in degrading the energy was lower.

The copper and aluminum crystals were cut with the [001] direction normal to the surface. The tungsten crystal was cut with the direction normal to the surface.

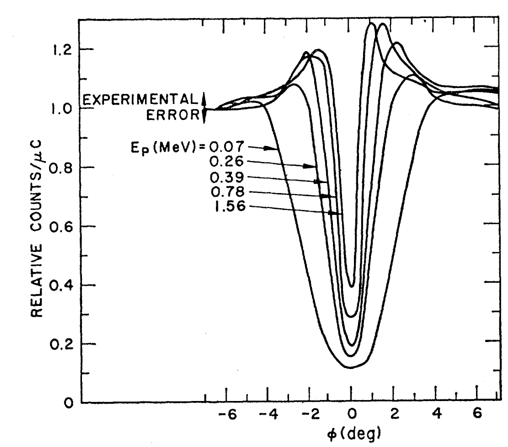


FIG. 2. Copper L-shell x-ray yield in the [011] direction normalized to unity at $\phi=-7°$.

III. OBSERVATIONS

Copper (fcc)

The similarity of the contour shown in Fig. 1 ($E_p=1560$ keV) and that of Fig. 5, of I ($E_p=70$ keV) is apparent. The polycrystalline value appears to be the average of the single-crystal values. The ($\bar{2}\bar{1}1$) and (211) planes can be observed in Fig. 1, and are more distinct than those in Fig. 5, of I. The 1560-keV contour, which

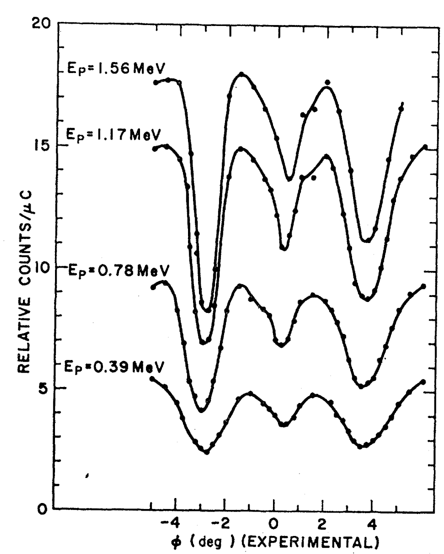


FIG. 3. Copper L-shell x-ray yield near the [011] direction ($\Delta\theta=1.5°$) showing the (111)-plane asymmetry.

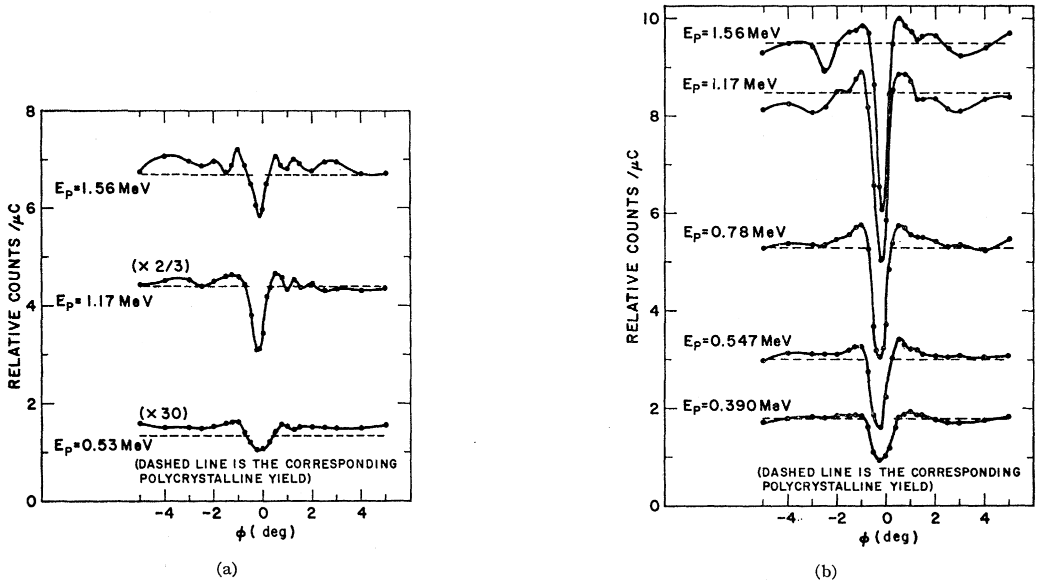


FIG. 4. Copper K-shell (a) and L-shell (b) x-ray yields for the (100) plane at $\theta=30°$.

covers the range of 6°×6°, shows that almost all features are reduced in angular scale from the 20°×20° contour at 70 keV.

Figure 3 shows relative yields at various proton energies for orientations of the [011] direction with respect to the direction of the proton beam, with the additional condition that the beam direction lies in the (011) plane. The curve at 70 keV is taken from I. The width and depth monotonically decrease with increasing proton energy. The yield values on either side of the [011] direction rise above and then drop to relatively constant values. These local maximums are hereinafter referred to as "wings." Equal widths are obtained from the K- and L-shell data; however, no wings are observed in the case of the K shell.

Figure 4 shows curves for orientations of the crystal with respect to the beam such that the beam direction is always in a plane which is (i) 1.5° from the [011] direction and (ii) perpendicular to the (100) plane. The curves show two outer dips in {111} planes which intersect in the [011] direction, and a center dip in the (100) plane.

At the highest energy (1560 keV), the two {111}-plane minima differ by 35%, but as the energy is decreased this difference is reduced. At 390 keV the sweep is nearly symmetric about the (100) plane, and at 70 keV (Part I) no asymmetry was observed. Four ⟨011⟩ directions were studied at 1560 keV, and curves were obtained at 1.5° from each of these ⟨011⟩ directions. In all cases, the two {111}-plane minima exhibited asymmetry of differing degrees. Asymmetries may indicate large domains within the target, with the beam overlapping some of these domains.

Figures 4(a) and 4(b) present, respectively, K- and L-shell x-ray yields for orientations of the crystal with respect to the beam such that the beam direction lies in

TABLE I. Characteristic full widths (deg). Values of the ratio R are given in parentheses.

E_p (MeV)	Cu (100) L shell	Cu (100) K shell	Cu (111) L shell	Cu (111) K shell	Cu [011][a] L shell	W (110) M shell	W (110) L shell	W (110) K shell	Al [011] K shell
0.070	1.6 (2.5)				4.6				
0.26			1.0 (2.2)		1.7	1.6[b] (3.0)			
0.39	1.0 (2.0)				1.4	1.3 (3.2)			
0.55	0.70 (2.0)	0.65[c] (1.2)	0.65 (2.3)	0.80 (1.9)					
0.78	0.68 (1.9)		0.60 (2.4)	0.60 (1.5)	0.95	0.92 (3.1)	0.90 (2.3)		1.0 (1.5)
1.17	0.65 (1.8)	0.60 (1.5)	0.55 (2.6)		0.90	0.8 (2.6)	0.85 (2.0)	0.85 (1.8)	
1.56	0.5 (1.6)	0.65 (1.2)	0.45 (2.4)	0.45 (1.3)	0.65	0.72 (2.4)	0.75 (1.8)	0.75 (1.3)	

[a] In the (0$\bar{1}$1) plane. [b] 0.25 MeV. [c] 0.53 MeV.

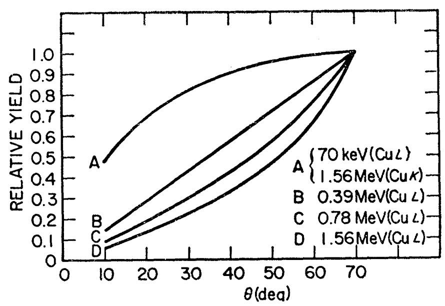


FIG. 5. Relative copper polycrystalline yield showing the effect of self-absorption. The curves were normalized to unity at $\theta=70°$.

a plane which is perpendicular to the (100) plane 30° from the crystal normal. Polycrystalline values are shown as dashed lines in the figures. The low yield from the K shell, combined with the small decrease observed at the (100) plane, makes quantitative interpretation

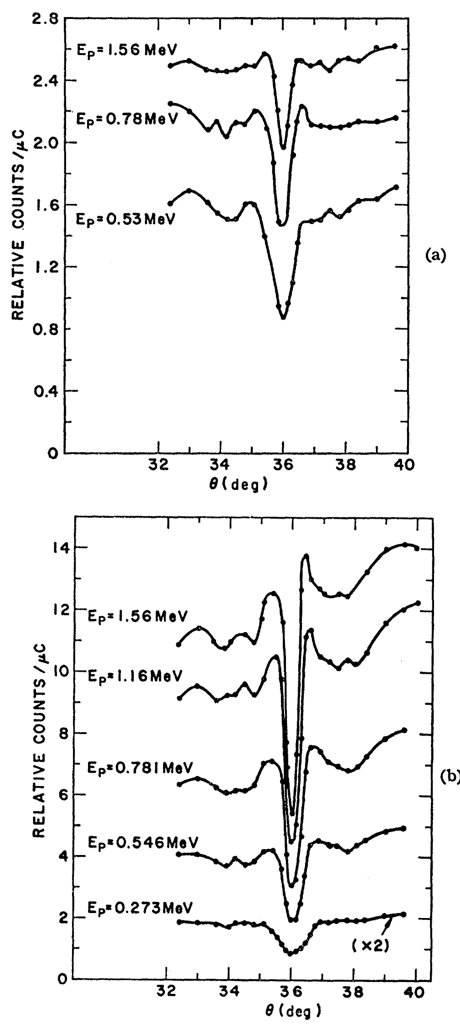


FIG. 6. Copper K-shell (a) and L-shell (b) x-ray yields as the tilt angle θ is varied across a {111} plane.

of the structure in Fig. 4(a) uncertain. Secondary structure in Fig. 4(b) (which becomes more pronouned as the energy is increased) may reflect the emergence of higher-index planes. In Fig. 4(b) the distinct maxima on either side of the (100)-plane dip resemble the wings observed in Fig. 2.

Figure 5 shows the effect of self-absorption of K- and L-shell x rays as a function of θ. Note that the curve for the L-shell x-ray yield at $E_p=70$ keV coincides with the curve for the K-shell yield at $E_p=1560$ keV.

Figures 6(a) and 6(b) show curves for the K- and L-shell x-ray yield as a function of θ. The minimum occurs at a {111} plane, 2° from a ⟨112⟩ direction. Curves for the K shell show relatively little effect of self-absorption, whereas curves for the L shell show an increasing effect as the proton energy, and hence depth of production, are increased. Again we see the appearance of wings on either side of the plane dips for the L shell, whereas larger uncertainties for the K shell preclude quantitative interpretation of the secondary structure. Note that R (the ratio of the left-side maximum value to the minimum value) for the L-shell yield at the (111) plane increases with increasing proton energy. This behavior is the opposite of what is observed in all the other cases.

Aluminum (fcc)

Figure 7 shows a curve for the K shell at $\theta=45°$. The minimum occurs when the [011] direction is along the direction of the incident beam. Comparison with Fig. 7(a) of I shows that the ratio R for the [011] direction at 1000 keV ($R=1.5$) is one-third the ratio at 70 keV. At 1000 keV, the yield in the (100) plane and 30° from the normal decreased from its maximum value by only 15%. This was approaching the experimental uncertainty, and therefore plane channeling in Al was not investigated further.

Tungsten (bcc)

Figures 8(a), 8(b), and 8(c) show curves for orientations of the crystal such that the beam is confined to a plane perpendicular to the ($\bar{1}10$) plane and 30° from the crystal normal. The M-shell curves show extensive

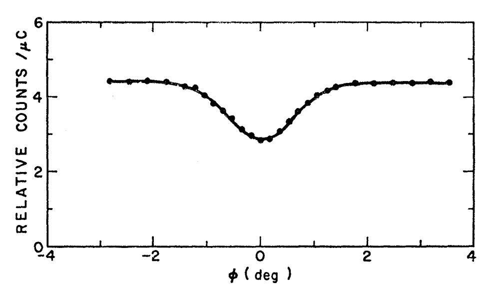


FIG. 7. Aluminum K-shell x-ray yield for the [011] direction at $\theta=45°$ and $E_p=1$ MeV.

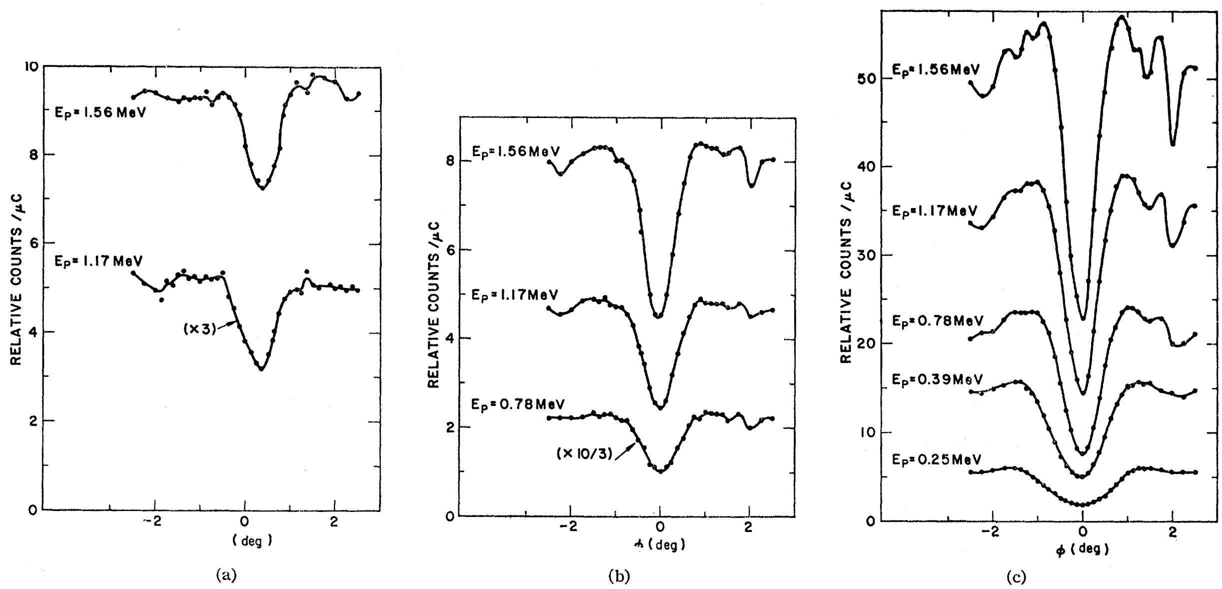


FIG. 8. Tungsten K-shell (a) L-shell (b), and M-shell (c) x-ray yield profiles for the (Ī10) plane at $\theta=30°$.

structure which is less prominent in the K- and L-shell curves. This structure may be due to nearby planes. No attempt has been made to explain the variation of structure with the shell excited.

IV. SUMMARY AND DISCUSSION

In summary, the data indicate the following trends:

(a) The local maxima referred to as wings [(Fig. 2)] depend, for a given element, on the shell excited, but apparently are not related to nearby planes.

(b) Ratios (R) decrease with increasing proton energy.

(c) Characteristic widths are *independent* of the shell excited.

(d) Polycrystalline targets appear to give an x-ray yield which is the average of those given by a single crystal, throughout the proton energy range of 70 to 1560 keV.

(e) Asymmetry in the {111} planes in copper has been detected (Fig. 3) and is probably due to defects in the crystal.

The characteristic full widths and the ratios R for the elements studied are listed in Table I; Fig. 9 is a plot of the half-widths as a function of $1/E_p^{1/2}$ for copper and tungsten. The data are fitted by straight lines which extrapolate to a value of 0.1° at $E_p=\infty$. This indicates either that the widths approach an inherent limiting value (0.1°), or that the inherent widths are much smaller than this, with the single crystal being composed of a mosaic structure with a spread of 0.1°. In the latter case there would be a corresponding contribution to the widths observed at finite energies. If the connection between widths and mosaic spread could be established unambiguously, it would be possible to measure the degree of angular spread of the mosaic structure in the surface layers of the crystal.

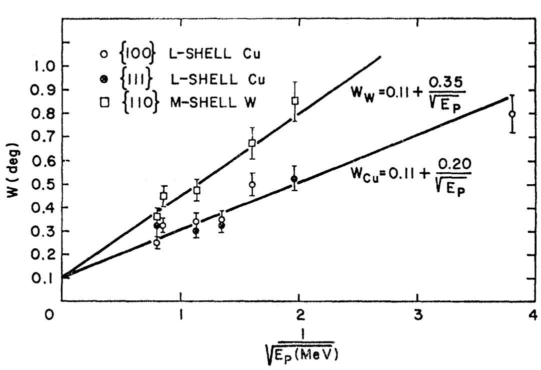
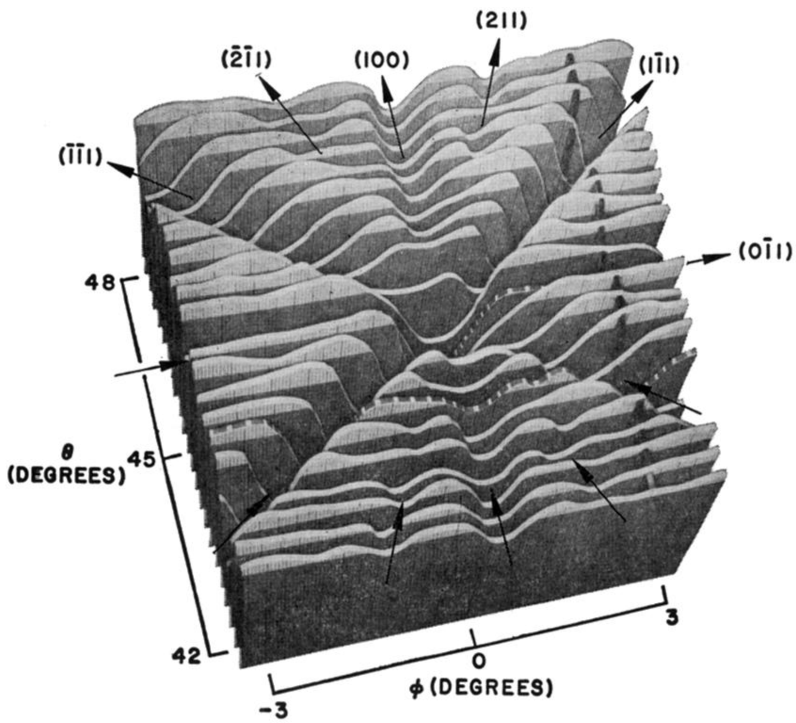


FIG. 9. Full widths divided by 2, plotted as a function of $1/E_p^{1/2}$.

The data may also reflect information on dechanneling,[2] which represents a loss of protons from the channel as the protons penetrate deeper into the crystal. These dechanneled protons are more effective in producing x rays than those remaining in the channel. The experimental results show that for a given element and proton energy, the ratio R increases with decreasing binding energy. Since the effective range for x-ray production (for these elements and proton energies) increases as the energy of the characteristic x ray increases (because of a decrease in self-absorption), a decrease in the ratio R indicates an increase in dechanneling. Therefore, protons become dechanneled as they penetrate deeper into the crystal.



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