Geiger-Müller Counter Measurements of Reflected Mo $K\alpha$ X-Rays from Powdered Zinc

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Volz has used the formula $n=n_0 \exp(-n_0/2.78n_m)$ where n is the observed counting rate of the system, n_0 is the rate of production of pulses in the counter and n_m is the maximum possible random counting rate. It was found that n_0, calculated from n and corrected for background count, varied as the inverse square of the distance from a gamma-ray source. Further, the absorption coefficient of Mo $K\alpha$ rays in aluminum was found to have the correct value when measured by means of this formula. Thus the formula was satisfactory. The absolute F values were obtained by comparison of the $(11\bar{2}2)$ reflection from zinc with the (420) reflection from KCl. This gives $F(11\bar{2}2)=13.9$. Brindley, using Cu $K\alpha$ rays, obtains $F(11\bar{2}2)=12.0$. The difference is caused by dispersion. The F values for other reflections very closely agree with those found by Brindley for Cu $K\alpha$ rays, when his values are corrected for dispersion. The very weak (0004) reflection, which was not obtained by Brindley, was found to have an F value of about 10.6. Jauncey and Bruce's values of the amplitudes of the thermal vibrations parallel and perpendicular to the c axis fit these results better than do Brindley's.

1. INTRODUCTION

THE problem of the anisotropy of the thermal vibrations of the atoms in zinc crystals has recently been studied by Brindley,[1] Jauncey and Bruce[2] and Zener.[3] Brindley measured the reflection of Cu $K\alpha$ rays ($\lambda=1.54$A) from powdered zinc, Jauncey and Bruce measured the diffuse scattering of x-rays (average $\lambda=0.435$A) from single zinc crystals at different orientation angles, while Zener made a theoretical investigation of the Debye-Waller temperature factor for zinc. Since Jauncey and Bruce's experiments were made with wave-lengths on the short wave-length side of the Zn K absorption edge ($\lambda=1.28$A) while Brindley's experiments were made on the long side, it was considered worthwhile to obtain F values for powdered zinc on the short wave-length side of the Zn K edge. For this purpose Mo $K\alpha$ rays ($\lambda=0.71$A) were used. Moreover, in Zener's formula[3]

$$M=(a\cos^2\psi+b\sin^2\psi)\cdot\frac{\sin^2\phi/2}{\lambda^2}. \qquad (1)$$

Brindley uses the values[4] $a=1.27\text{A}^2$ and $b=0.495\text{A}^2$, whereas Jauncey and Bruce prefer the respective values 2.34 and 0.68. This situation gave further reason for this present research, and especially so since Jauncey and Bruce were forced to correct Brindley's values for dispersion. Still again Brindley used a photographic method and it was thought worthwhile to make the measurements by another method. On account of the recent success of Locher, LeGalley[5] and others in using Geiger-Müller tubes for x-ray measurements, it was decided to use the G-M tube in the present research.

2. APPARATUS

The Geiger-Müller tube which was used had a 100-mesh stainless steel screen cathode of 1.5 cm diameter and 15.0 cm length and was filled with methyl bromide at 8 cm pressure. The anode wire was made of 22-mil nickel. The x-rays entered the tube through a thin glass window whose effective area was defined by means of a slit in a lead sheet. The rays passed along the tube at a small angle with the anode wire and crossed this wire. A lead cylinder 1.5 cm thick surrounded the tube to reduce the number of stray counts. Methyl bromide was found to be quite sensitive to x-rays, but to deteriorate with use so that the counter had to be refilled daily. The anode was held at 1400 volts, about 175 volts above the threshold.

[1] G. W. Brindley, Phil. Mag. [7] **21**, 789 (1936).
[2] G. E. M. Jauncey and W. A. Bruce, Phys. Rev. **50**, 408 (1836).
[3] C. Zener, Phys. Rev. **49**, 122 (1936).
[4] The English have lower room temperatures than we do. These values of a and b are for 290°K. Calculated for 298°K they become $a=1.29\text{A}^2$ and $b=0.51\text{A}^2$.
[5] G. L. Locher and D. P. LeGalley, Phys. Rev. **46**, 1047 (1934); D. P. LeGalley, Rev. Sci. Inst. **6**, 279 (1935).

The circuit described by Neher and Harper[6] was used, because of the lower resistances necessary, and because the counting rate is independent of the supply voltage over a range greater than in the conventional circuit. The voltage supply was a conventional one with a Medicus[7] control. For recording the counts, the frequency meter described by Hunt[8] was tried, but was found unsatisfactory for slow counting rates, even when using the tank circuit described by Gingrich, Evans and Edgerton.[9] A mechanical counter consisting of a stopwatch actuated by a magnetic relay was finally used. The over-all resolving time of the system (amplifier plus counter) for irregularly spaced pulses was found to be of the order of 0.1 second.

3. Tests of Counting System

The observed counting rate of the mechanical counter is only proportional to the rate of production of pulses in the G-M tube when the rate is small. For brevity we shall call the rate of production of pulses in the G-M tube the true counting rate.

Volz[10] has used the formula

$$n = n_0 e^{-n_0/en_m}, \tag{2}$$

where n and n_0 are the observed and true counting rates, respectively, and the parameter n_m is the maximum random counting rate which can be recorded by the mechanical counter. n_m can be easily determined by bringing a source of gamma-rays up to the G-M tube; it is found that the counting rate increases to a maximum and then diminishes as the distance between the gamma-ray source and the G-M tube is lessened. For this apparatus, $n_m = 670$ counts per minute.

Formula (2) was subjected to the following tests: (a) A source of gamma-radiation was placed at various distances from the tube, and it was found that, correcting for the background counts, the true counting rate as obtained by (2) from the observed counting rate[11] was proportional to the inverse square of the distance from the counter. (b) The $(10\bar{1}1)$ reflection from zinc of Mo $K\alpha$ rays, which is relatively very intense, was allowed to enter the G-M tube. In order to remove the Mo $K\beta$ rays a ZrO_2 screen was placed over the counter window admitting the x-rays. This screen also removed the fluorescent zinc radiation. Various thicknesses of aluminum were placed in the beam and it was found that when $\log n_0$ is plotted against thickness, a reasonably good straight line was obtained. The value of the mass absorption coefficient of Mo $K\alpha$ rays in aluminum was found to be 5.30. This is in good agreement with the value 5.22 given by Compton and Allison.[12] (c) With the $(10\bar{1}1)$ reflection entering the window of the G-M tube, first the upper half and then the lower half of the window was closed. The sum of the observed counting rates was found to be considerably greater than the counting rate for the completely open window. When the corresponding true counting rates were found by means of (2), the sum of the true rates for the two halves was equal to the true rate for the whole window.

In order to measure the intensity of a zinc reflection at one setting of the G-M tube, it was necessary to make the width of the window admitting the rays of the reflection to the tube wide enough to take account of the angular divergence of the beam. With a wide window it was further necessary to measure the sensitivity of the G-M tube to rays entering various parts of the window. This was done by placing a fixed slit one-quarter the width of the window just in front of the window and measuring the counting rate as the tube with its window was moved across the beam. Such a test showed that the tube was equally sensitive to rays entering all parts of the window.

4. Experimental Arrangement

The collimating slit and the G-M tube window were each placed at 16.8 cm from the powdered crystal so that partial focusing was obtained. The collimating slit was 0.1 cm wide and 0.8 cm high. The tube window was 0.2 cm wide by 0.5 cm high. The ZrO_2 filter was placed in front

[6] H. V. Neher and W. W. Harper, Phys. Rev. **49**, 940 (1936).
[7] G. Medicus, Zeits. f. tech. Physik **8**, 304 (1933).
[8] F. V. Hunt, Rev. Sci. Inst. **6**, 43 (1935).
[9] N. S. Gingrich, R. D. Evans, H. E. Edgerton, Rev. Sci. Inst. **7**, 450 (1936).
[10] H. Volz, Zeits. f. Physik **93**, 540 (1935).
[11] It is not possible to express n_0 explicitly in terms of n so a graph of n *versus* n_0 was plotted. From this, n_0 corresponding to a given value of n can be obtained immediately.
[12] Compton and Allison, *X-Rays in Theory and Experiment* (1935).

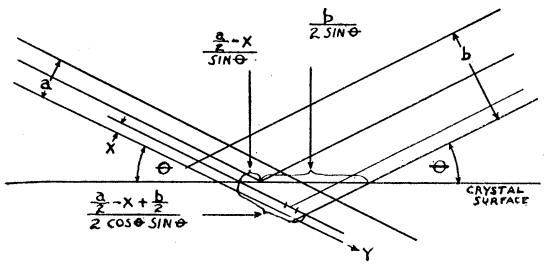

FIG. 1.

of the counter window to absorb the Mo $K\beta$ line and the zinc fluorescent radiation.

The zinc powder was prepared by the sublimation of zinc filings heated in vacuum, as described by Brindley and Spiers.[13] The x-ray tube with a water-cooled molybdenum target was operated at 32 kv peak and 28 ma. The briquet of zinc powder was always placed so that α, the angle between the primary beam and the briquet face, was equal to the angle between the reflected beam and the face.

For convenience the notation used in Brindley's[13] paper will be followed. The quantity I/A is the intensity for a given reflection when corrected for background. Values of I/A were determined on both sides of the primary beam and the average taken. Each of the reflections was compared with the $(11\bar{2}2)$ reflection as a standard. Ten minute readings were taken in the order—standard reflection, standard background, test reflection and test background. A total of two hours of observation was taken for the stronger reflections. The weaker ones were observed for a total of three hours. As an example of the number of counts recorded, the $(11\bar{2}2)$ reflection will serve. The true background (counter plus x-rays) rate was about 80 per minute, while that of the reflections was 115 per minute.

To get the absolute value of the $(11\bar{2}2)$ reflection, it was compared with the (420) reflection of KCl. The KCl was finely powdered and passed through a 200-mesh screen. The comparison was made by alternately taking readings, first from a KCl briquet and then from the zinc. A comparison was tried with MgO, but because of its very small density, when powdered, and its small absorption coefficient, the x-ray beam penetrated the briquet a considerable distance. The scattered beam was so broad that accurate measurements were impossible. The same effect occurs to a much smaller degree with KCl, and it becomes necessary to find a formula which will indicate the inaccuracy due to this effect.

In Fig. 1, let $\phi=2\theta$ be the scattering angle, a be the width of the incident x-ray beam, b the width of the detector window, μ' the effective linear absorption coefficient of the powder, and let y be measured from the crystal surface along the direction of propagation of the x-rays. Let $P(x)dx$ be the intensity distribution across the beam between x and $x+dx$. We shall consider the case of no angular divergence. When the incident and reflected beams are at the same angle, the fraction of the rays traveling between x and $x+dx$ which are scattered by an element dy at y and which emerge from the crystal in the direction of the detector window is $\exp(-2\mu' y)s'dy$, where s' is a linear scattering coefficient. By reference to Fig. 1, it is seen that the total intensity scattered is

$$\int_0^a P(x)dx \int_0^{(a-2x+b)/(4\cos\theta\sin\theta)} e^{-2\mu' y}s'dy. \quad (3)$$

For a beam of uniform cross-sectional intensity $P(x)dx=I_0dx/a$. In this case the total intensity becomes $s'I_0/2k\mu'$, where

$$k=\frac{1}{1-(\sin\phi/2a\mu')[e^{-\mu'(b-a)/\sin\phi}-e^{-\mu'(a+b)/\sin\phi}]}. \quad (4)$$

The final result is that μ' in Eq. (2) of Brindley and Spiers' paper[13] must be replaced by $k\mu'$. When the method of standardization by separate powders is used, μ_1 and μ_2 in Brindley and Spiers' Eq. (5) must be replaced by $k_1\mu_1$ and $k_2\mu_2$ where the subscripts refer to the two powders. For MgO this correction amounted to about 30 percent. In the case of KCl, however, it was less than $\frac{1}{2}$ percent and so was neglected.

From data given by James and Brindley[14] the absolute F value for the (420) reflection from KCl was estimated to be 13.5. From this it

[13] G. W. Brindley and F. W. Spiers, Phil. Mag. [7] **20**, 865 (1935).

[14] R. W. James and G. W. Brindley, Proc. Roy. Soc. **A121**, 154 (1928).

follows that the F value for the standard $(11\bar{2}2)$ reflection from zinc is 13.9. The F values for all the zinc reflections are expressed in terms of this.

5. The Effect of Diffuse Scattering on the Background

During this work the question of the proper background from which to measure the reflection maxima arose. As a result of a discussion with Professor Jauncey, the following answer to this question is presented.

Diffuse scattering consists of a coherent and an incoherent part. When monochromatic rays fall upon a single crystal, reflections only occur at those values of θ which satisfy Bragg's law and nowhere else. The incoherent part of the diffuse scattering occurs at all except very small values of the scattering angle, while the coherent part occurs between the Bragg reflections. As a Bragg reflection is approached the coherent part changes into the Bragg reflection at a Bragg value of θ. Hence for a single crystal the background which must be subtracted from a reflection maximum is that due to the incoherent part of the diffuse scattering. In a recent experiment in this laboratory, the intensity of the (200) reflection of a slightly divergent beam of Mo $K\alpha$ rays from rock-salt was 2000 as against 1 for the diffuse scattering. Of the value of 1 for diffuse scattering, an amount 0.5 was probably due to the incoherent part. Whether one subtracts 1 or 0.5 from 2000 makes very little difference, and the subtraction of whatever background there is on either side of the reflection maximum for a single crystal gives a sufficiently accurate value of the true reflection intensity.

In the case of reflection from a powdered crystal the intensity of the background which is due to diffuse scattering and is on either side of the reflection is of the same order of magnitude as the intensity of the reflection itself. The probability that a crystallite of the powder[15] will be lined up to give a certain reflection of x-rays of a given wave-length depends upon the divergence of the primary beam and upon the values of $\cos\theta$ and of the multiplicity factor p. The fraction of the crystallites taking part in reflection is so small that almost 100 percent of these may be considered as giving rise to diffuse scattering. For this reason practically no error is made when the whole of the diffuse scattering background is subtracted from a reflection maximum in order to obtain the true intensity of the reflection.

Table I. *F values for various reflections of Mo Kα x-rays from powdered zinc.*

Reflection	$\sin\theta/\lambda$	ψ	$pS^2\phi(\theta)$	I/A	F
0002	0.203	0.0	375	212	22.3
$10\bar{1}0$	.218	90.0	242	161	24.2
$10\bar{1}1$	.240	65.0	1187	630	22.0
$10\bar{1}2$	.298	47.0	251	100.1	18.7
$10\bar{1}3$	.374	35.6	455	217	15.9
$11\bar{2}0$	.378	90.0	303		
0004	.406	0.0	85	11	10.6
$11\bar{2}2$	.428	61.7	454	100	13.9
$20\bar{2}1$	.447	76.9	309	68	13.9
$20\bar{2}3$	.532	55.0	206	24.5	10.2
$10\bar{1}5$	.552	23.2	189	33.5	8.2
$11\bar{2}4$	.554	42.8	250		
$21\bar{3}1$	.584	80.0	328	34.9	9.7
0006	.609	0.0	32.8	9.2	7.85
$21\bar{3}2$	.611	70.6	98.5		

These values of I/A have been corrected for the $K\beta$ ray which was 0.0075 times the intensity of the $K\alpha$. The only value which it changes is the $(10\bar{1}0)$ reflection.

6. Discussion of Results

The final results are shown in Table I. By comparison with Brindley's F values for Cu $K\alpha$ rays it is seen that the above F values are all larger than his values. This is because of dispersion. According to Hönl[16] the difference of f values for $\lambda/\lambda_K=1.20$ and $\lambda/\lambda_K=0.55$ is 2.36. By comparison with the (220) and (311) reflections from aluminum Brindley obtained $F=12.0$ for the standard $(11\bar{2}2)$ reflection from zinc. The value of F for the $(11\bar{2}2)$ reflection from zinc found by the author was 13.9. The relation between Δf and ΔF is

$$\Delta f=\Delta F e^{-M}, \qquad (5)$$

provided that F and $F-\Delta F$ refer to the same temperature. From diffuse scattering measurements, Jauncey and Bruce[2] find $a=2.34\text{A}^2$ and

[15] Compton and Allison, *X-Rays in Theory and Experiment* (1935), pp. 415–420.

[16] H. Hönl, Ann. d. Physik **18**, 625 (1932).

$b=0.68\text{A}^2$. Using these values in (1), we find from (5) that $\Delta f=1.9e^{0.193}=2.31$. This is remarkably close to the Hönl value of 2.36. This excellent agreement is probably more fortuitous than real.

The experimental values of F are shown as hollow circles in Fig. 2. The same main features are found with Mo $K\alpha$ rays as Brindley finds with Cu $K\alpha$ rays, *viz.*, the zigzag at the last four points and also the anomalous behavior of the first two points. This anomalous behavior of the first two points, the (0002) and $(10\bar{1}0)$ reflections, has been previously verified by Miller and Foster.[17] It has been suggested as being due to distortion of the electron atmosphere of the zinc atoms. That the zigzag of the last four points is due to anisotropy in the vibrations of the atoms is shown by the fact that, when the f values are calculated by means of

$$f=Fe^{M},$$

and when Jauncey and Bruce's values of a and b are used, the resulting f points fall upon a curve in which the zigzag has been almost entirely smoothed out. The black circles in Fig. 2 show the f values for this case. The crosses show the f values as calculated when Brindley's values[4] of a and b are used in the calculation.

In addition to the F values found by Brindley it was estimated that the F value for the (0004) reflection is 10.6. This is not to be taken as accurate because the counting rate for this reflection is very small, but it does show that it is another low point on Fig. 2. Evidently the F values for $\psi=0°$ are low and the amplitude of vibration along the c axis is large. This shows that a in (1) must be large.

It is believed that after correcting for the atomic vibrations the f values fall upon a smooth curve at large values of $(\sin\theta)/\lambda$ but that at small values of $(\sin\theta)/\lambda$ the f values fall between two curves, one for $\psi=0°$ and the other for $\psi=90°$, the particular position for an f point depending upon the value of ψ. The forking f curve is shown in Fig. 2. This phenomenon is caused by electron distortion. Since by reference to Fig. 2 it is seen that for large values of $(\sin\theta)/\lambda$ the black circles fall somewhat better on a smooth curve than do the crosses it seems that Jauncey and Bruce's values of a and b are better than Brindley's. Jauncey and Bruce's values correspond to 0.172A and 0.093A for the root-mean-square displacements parallel and perpendicular to the c axis.

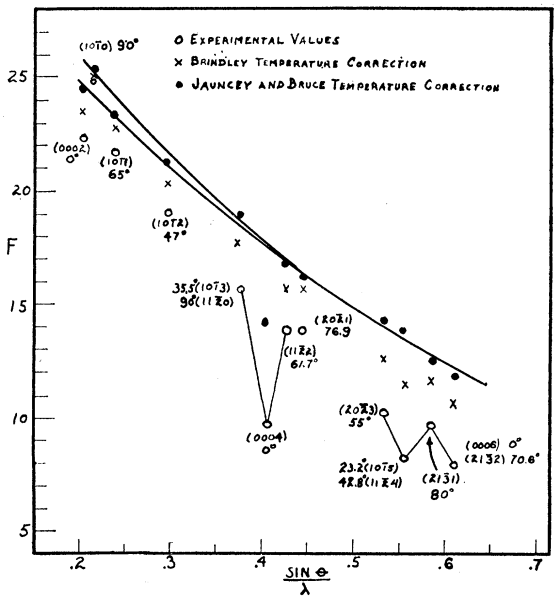

FIG. 2. Experimental F values and temperature corrections. (A better value for the (0004) reflection seems to be 10.6 instead of the value 9.6 indicated in the figure.)

The author wishes to thank Professor Jauncey for his untiring encouragement and help in the working out of this problem.

[17] R. D. Miller and E. S. Foster, Jr., Phys. Rev. **50**, 417 (1936).