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Diffuse Scattering of X-Rays from Sylvine. II

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The absolute intensity of the diffuse scattering of x-rays of wave-length 0.71A from sylvine has been measured for a range of angles from 5° to 110°. Homogeneous radiation obtained by the use of balanced filters was used. Woo's formula taking account of the incoherence of part of the scattering was used to calculate the average atomic structure factors of the ions K^+ and Cl^-. The results are in good agreement with Wollan's values for argon as well as theoretical values calculated from a Schrödinger charge distribution by Hartree's method.

I. INTRODUCTION

It has been shown[1, 2] that the diffuse scattering from a simple cubic crystal consisting of atoms of one kind should be given classically by

$$S_{\text{class.}}=1+(Z-1)f'^2/Z^2-F^2/Z, \qquad (1)$$

where $S_{\text{class.}}$ is the scattering per electron in terms of the Thomson[3] value in the same direction, Z the atomic number, F the atomic structure factor including the effect of thermal agitation and f' a quantity related to[1] the true atomic structure factor f. Woo[4] has independently deduced a similar formula, taking account of the fact that part of the scattering is incoherent. Woo's formula may be written

$$S=(f'^2-F^2)/Z+(1-f'^2/Z^2)(1+\alpha\ \text{vers}\ \phi)^3, \qquad (2)$$

where ϕ is the angle of scattering and $\alpha=h/mc\lambda$. The first term in Eq. (2) represents the coherent and the second the incoherent part of the scattering.

Several investigators[5, 6, 7, 8, 9] have reported measurements on the diffuse scattering of x-rays from crystals but in no case has homogeneous radiation been used. Although previous measurements on sylvine[7] gave values of the structure factors f' in good agreement with Wollan's values[10] for argon it was thought that this agreement was somewhat fortuitous in that no correction was made for incoherent radiation (Woo's paper[4] had not then appeared) and the wave-length range, though apparently narrow as measured by absorption, must have been rather broad. Further, it does not seem to the writer that an *average* wave-length as measured by absorption is the correct average to use for the

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[1] G. E. M. Jauncey, Phys. Rev. **37**, 1193 (1931).

[2] G. E. M. Jauncey and G. G. Harvey, Phys. Rev. **37**, 1203 (1931).

[3] J. J. Thomson, *Conduction of Electricity through Gases*, 2nd edition p. 325.

[4] Y. H. Woo, Phys. Rev. **38**, 6 (1931).

[5] G. E. M. Jauncey, Phys. Rev. **20**, 405 (1922).

[6] G. E. M. Jauncey and H. L. May, Phys. Rev. **23**, 128 (1924).

[7] G. G. Harvey, Phys. Rev. **38**, 593 (1931). The values of s_ϕ/ρ on p. 601 of this paper must all be multiplied by 0.024.

[8] G. E. M. Jauncey and P. S. Williams, Phys. Rev. **41**, 127 (1932).

[9] G. E. M. Jauncey and Paul Ehrenfest II, Phys. Rev. **42**, 907 (1932).

[10] E. O. Wollan, Phys. Rev. **37**, 862 (1931).

process of scattering. For if $\bar{\lambda}_a$ is the average wave-length as measured by absorption and $\bar{\lambda}_s$ the correct value to use for scattering and if $I(\lambda)$ is the spectral distribution then

$$e^{-\mu(\bar{\lambda}_a)t}\int_{\lambda_0}^{\infty} I(\lambda)d\lambda \approx \int_{\lambda_0}^{\infty} I(\lambda)e^{-\mu(\lambda)t}d\lambda$$

for some particular range of values of t and

$$S(\bar{\lambda}_s)\int_{\lambda_0}^{\infty} I(\lambda)d\lambda = \int_{\lambda_0}^{\infty} I(\lambda)S(\lambda)d\lambda,$$

where $\mu(\lambda)$ is the absorption coefficient of the substance used in determining $\bar{\lambda}_a$, λ_0 the minimum wave-length and S is given by Eq. (2). It is obvious that in general $\bar{\lambda}_a \neq \bar{\lambda}_s$, though of course they may not differ greatly if the spectrum is not too extended. It is to be noted, however, that $\bar{\lambda}_s$ will also depend on ϕ. Recently in the case of sodium fluoride Jauncey and Williams[8] have found it necessary to take account of the fact that part of the scattered radiation is incoherent and although this effect is smaller for sylvine it is not negligible. It was hence thought desirable to repeat the measurements on sylvine using homogeneous radiation obtained by the use of a balanced filter.[11]

II. Apparatus and Procedure

A diagram of the apparatus used is shown in Fig. 1. T is a molybdenum target x-ray tube immersed in transformer oil in a lead box A. The tube was operated at 30 m.a. at a potential of about 42.5 kv peak with full wave kenotron rectification. The x-rays left the box through a small celluloid window after passing through about 2 mm of oil. The primary beam was defined by the "slits" s_1 and s_2 consisting of small circular holes. s_3 served to eliminate scattering from the slit s_2 and was slightly larger than the x-ray beam. The angular width of the beam was about one degree. The balanced ZrO_2–SrO filter was placed in front of s_1 and the radiation scattered by the crystal C was measured by an ionization chamber and a Compton electrometer operating at a sensitivity of 5000 mm/volt. The ionization chamber was built of copper and fitted with thin celluloid windows the absorption in which was too small to be measured. All insulation was of amber. The chamber was 7.5 cm long and filled with krypton at a pressure of 57 cm of mercury N.T.P. This results in an absorption of 85 percent for the Mo $K\alpha$ lines. The use of such a small chamber not only greatly reduces the residual ionization due to cosmic radiation and α-particles but results in a rather low capacity electrometer system with a consequent high charge sensitivity. Further details on the construction of such ionization chambers have been given by Compton elsewhere.[12]

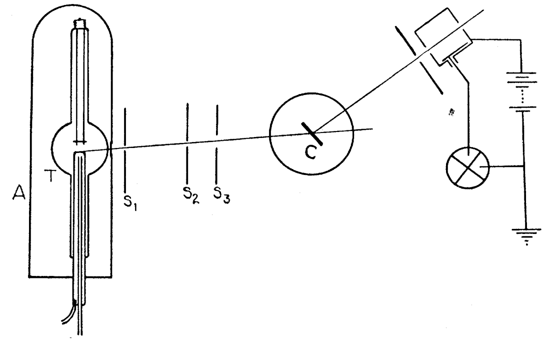

Fig. 1. Diagram of apparatus.

The transmission by the filters is shown in Fig. 2, the unshaded portion representing the part used. It is not to be supposed that this represents the energy distribution in the spectrum as no correction for absorption or second order reflection was made and a rather broad primary beam was used. This accounts for the peak at 0.8A and the very broad $K\alpha$ lines, though of course does not affect the correctness of the balance.

The procedure was the same as in previous experiments except that readings were taken with first one filter and then the other and the difference of the currents taken as a measure of the scattering for the Mo $K\alpha$ lines alone. Due to the fact that it is the difference of the readings that is required, it is necessary to take a large number of readings and in fact several thousand were taken. With the chamber set at an angle ϕ the ionization current was measured for a series of values of θ, the angle between the primary beam and the normal to the crystal face. At $\theta=\phi/2$ the Laue spot was not entirely absent as this would necessitate

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[11] P. A. Ross, Phys. Rev. **28**, 425 (1926).

[12] A. H. Compton, Rev. Sci. Inst. **2**, 365 (1931).

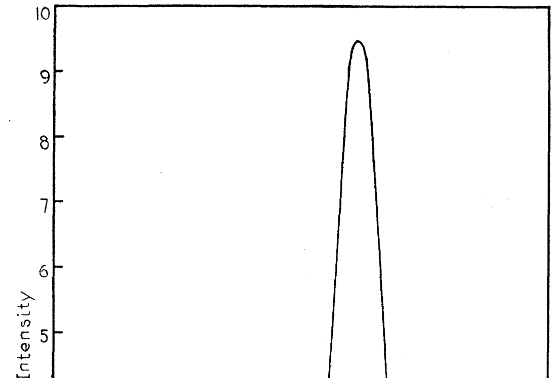

FIG. 2. Transmission by filters.

a more perfect balancing of the filters than is possible since the reflected and scattered intensities are of entirely different orders of magnitude. The Laue spot was never very intense, however (except of course for $\lambda = 0.71\text{A}$). The scattering from the crystal at an angle ϕ was then compared with that from a paraffin block at 90°. The ratio of the intensity transmitted by the crystal when set at $\theta = \phi/2$ to that transmitted by the paraffin at $\theta = 45°$ was also measured. Because of the rather high mass absorption coefficient of sylvine ($13.7\ \text{cm}^2\ \text{g}^{-1}$) it was necessary to use a very thin crystal, the surface density being $0.0843\ \text{g cm}^{-2}$. The surface density of the paraffin was $0.263\ \text{g cm}^{-2}$. The range of angles used extended from 5° to 110° and at the small angles it was necessary to correct for the height of the slit in front of the ionization chamber as it subtended an angle of about 5°.

It has been shown[13] that Crowther's formula[14] when modified to take account of the fact that part of the scattered radiation is incoherent because of the Compton effect, becomes

$$I_\phi = I_\phi' t(s_1 + s_2 T) \sec \phi/2, \qquad (3)$$

where I_ϕ is the scattered intensity per unit solid angle in the direction ϕ, I_ϕ' the transmitted intensity when the scattering block is set at $\theta = \phi/2$, t the thickness of the scattering block, s_1 and s_2 the spatial scattering coefficients for the unmodified and modified rays, respectively, and

$$T = (1 - e^{-(\mu' - \mu)t \sec \phi/2}) / (\mu' - \mu) t \sec \phi/2, \qquad (4)$$

μ' and μ being the linear absorption coefficients for the modified and unmodified rays. In Eq. (4) we have neglected the difference in the absorption of the modified and unmodified rays in the air and ionization chamber window. Since the absorption in the ionization chamber is not complete account must be taken of the fact that the incoherent portion is more completely absorbed than the coherent. As the derivation of the required equation has been given by Jauncey and Williams[8] it will not be repeated here. Jauncey and Williams assumed all the radiation scattered by paraffin at 90° to be incoherent, since they used a rather short wave-length (0.39A). For 0.71A, however, the percentage of coherent scattering becomes appreciable and this introduces a slight modification. Their equation, thus modified, becomes

$$\frac{K_0(f'^2 - F^2)}{Z_c} + K_\phi T_c \frac{1 - f'^2/Z_c^2}{(1 + \alpha \text{ vers } \phi)^3} = \frac{K_0 A + K_{90} B T_P/(1+\alpha)^3}{1 + \cos^2 \phi} \left(\frac{W}{Z}\right)_C \left(\frac{Z}{W}\right)_P \left(\frac{C_\phi}{P_{90}}\right) \left(\frac{P}{C}\right), \qquad (5)$$

where K_ϕ is the percentage absorption in the ionization chamber for radiation of wave-length $0.71 + 0.024 \text{ vers } \phi$, C_ϕ the ionization current for scattering from the crystal at an angle ϕ, P_{90} the same for paraffin at 90°, C and P the currents produced by the rays transmitted by the crystal and paraffin, $(\rho t)_C$ the surface density of the crystal, $(\rho t)_P$ the same for paraffin, W and Z the atomic weights and atomic numbers, A and B the percentages of coherent and incoherent scattering from paraffin at 90°, and the other quanti-

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[13] G. E. M. Jauncey and O. K. DeFoe, Phil. Mag. **1**, 711 (1926).

[14] J. A. Crowther, Proc. Roy. Soc. **A86**, 478 (1912).

ties have been previously defined. K_ϕ only varies from 0.85 to 0.88 from 0° to 110°.

To calculate the coherent scattering from paraffin Wentzel's formula[15] in the form given by Compton[16] was used in a previous[17] paper giving

$$I_{\text{coh.}} = 4I_e/(1+0.352\ x^2)^4, \qquad (6)$$

where I_e is the Thomson value of the scattering from a single electron and $x = (\sin\phi/2)/\lambda$, as the coherent intensity scattered by each carbon atom in paraffin, it being assumed that only the K electrons scatter coherently. Likewise for the incoherent intensity from the K electrons of a carbon atom we obtain

$$I_{\text{incoh.}} = 2I_e[1 - 1/(1+0.352x^2)^4]. \qquad (7)$$

Taking the formula for paraffin as C_nH_{2n+2} and assuming all the scattering from hydrogen to be incoherent we obtain for $\phi = 90°$

$$I_{\text{coh.}}/I_{\text{total}} = 1.2n/(7.4n+2) = 0.16 \qquad (8)$$

for all values of n above 20. Thus in Eq. (5), $A = 0.16$ and $B = 0.84$. Inserting the experimental values of C_ϕ/P_{90} and P/C in Eq. (5) and using values of F as given by James and Brindley[18] we may calculate values of f'. The structure factors so calculated are given in Table I. The values in parentheses are rather doubtful because of the extremely weak intensity. These values of f' are also plotted in Fig. 3. The solid curve represents the average of f_{K^+} and f_{Cl^-} as calculated by James and Brindley[19] from a Schrödinger charge distribution by the method of Hartree. The crosses represent Wollan's values [10] for argon calculated from Compton's formula for the scattering from a monatomic gas[16] and the circles represent the present values for sylvine. Although it was not possible with the wave-length used here to go out to as large values of $(\sin\phi/2)/\lambda$ as previously[7] there is an indication that the present values give better agreement with theory at large angles. It may be mentioned that the f values for argon as given by James and Brindley[19] are slightly larger than the average for K^+ and Cl^- at small values of x but slightly smaller for large x. It is seen from Fig. 3 that at small angles Wollan's values for argon are above those for sylvine, though the

TABLE I. *Structure factors.*

ϕ	x	F	f'	$S_{\text{class.}}$
5°	0.061	17.2	(17.2)	(0.06)
10	0.123	14.3	15.15	1.80
15	0.184	11.8	12.95	2.07
20	0.245	9.9	11.61	2.62
25	0.306	7.85	10.00	2.82
30	0.365	6.45	9.03	2.97
35	0.424	5.48	8.15	2.82
40	0.482	4.57	7.61	2.87
50	0.596	3.40	6.65	2.67
60	0.704	2.45	5.95	2.53
70	0.808	1.73	5.6	2.47
80	0.905	1.12	4.7	2.09
90	0.995	0.75	4.3	1.98
100	1.080	0.50	(3.6)	(1.67)
110	1.154	0.35	(3.25)	(1.55)

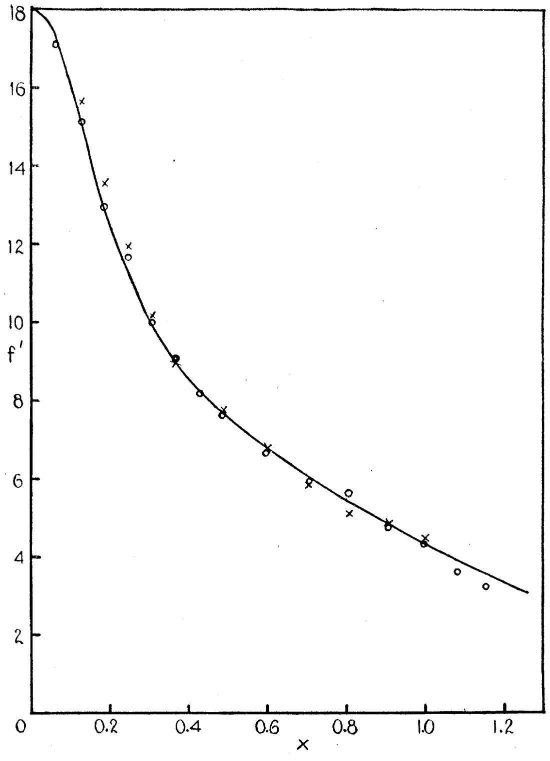

FIG. 3. Structure factors. Crosses, argon (Wollan); circles, sylvine; solid curve, $\frac{1}{2}(\text{K}^+ + \text{Cl}^-)$-wave mechanics.

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[15] G. Wentzel, Zeits. f. Physik **43**, 1, 779 (1927).

[16] A. H. Compton, Phys. Rev. **35**, 925 (1930).

[17] G. E. M. Jauncey and G. G. Harvey, Phys. Rev. **37**, 698 (1931). In this paper the factor 2 was omitted in the exponential occurring in $u(r)$, Eq. (11), and the exponent 4 in the denominator of Eq. (12). The correct form was used in the calculations, however.

[18] R. W. James and G. W. Brindley, Proc. Roy. Soc. **A121**, 155 (1928).

[19] R. W. James and G. W. Brindley, Phil. Mag. **12**, 81 (1931).

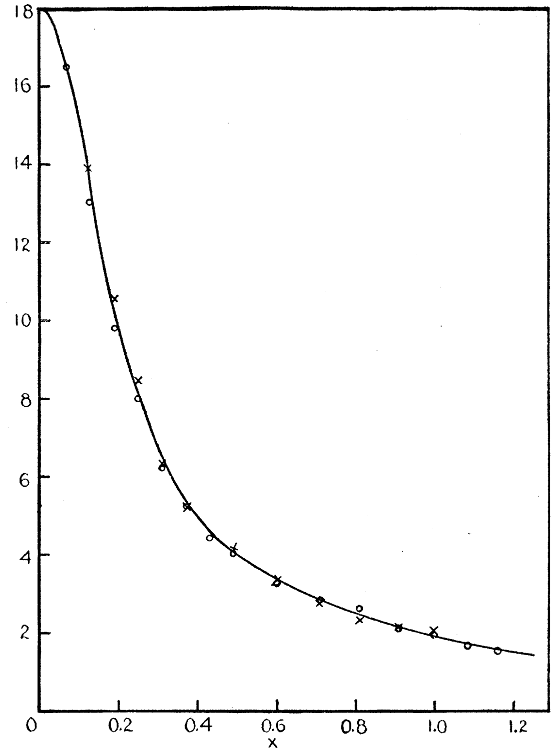

FIG. 4. Comparison with scattering from argon. Crosses $=S_{\text{gas}}$ (Wollan); circles $=(S+F^2/Z)_{\text{crystal}}$.

difference is perhaps within the combined experimental errors of the two sets of measurements. At large angles the difference is in the same direction but the intensity at large angles is extremely weak and this difference is less than the probable error of the sylvine measurements alone.

It has been pointed out[20] that the scattering from a crystal, reflection from a crystal, and scattering from a monatomic gas having the same atomic number as the crystal are related by the equation

$$S_{\text{gas}}=(S+F^2/Z)_{\text{crystal}}. \tag{9}$$

This relation holds whether the S values are classical or modified to take account of the Compton effect but as Wollan's values are tabulated as $S_{\text{class.}}$ we shall use this form. The values of S in the last column of Table I were calculated from Eq. (1) with the f' and F values given there. In Fig. 4 values of $S_{\text{class.}}+F^2/Z$ for sylvine are indicated by circles and Wollan's values of $S_{\text{class.}}$ for argon by crosses. It is seen that the agreement is very good on the whole but, again, at small angles there is a small difference.

III. CONCLUSION

Data on the diffuse scattering from sylvine when combined with results on reflection yield values of the structure factors which within the limits of experimental error agree with those calculated from a Hartree model. The difference between the structure factors for argon and sylvine, though hardly significant, is in general in the right direction.

The primary purpose of this investigation was to determine the effect of temperature on the intensity of the diffuse scattering. Experiments at the temperature of liquid air with monochromatic radiation are now in progress and will be reported later.

In conclusion the author wishes to express his appreciation to Professor Arthur H. Compton for his interest in the work and for the use of the Ryerson Physical Laboratory.

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[20] G. E. M. Jauncey and G. G. Harvey, Phys. Rev. **38**, 1071 (1931).