behavior but indicates that the electron component extends to appreciably higher values than that given by Wilson.

It must be emphasized that this experiment, using a high field, was not primarily intended for work in this transition energy region. More accurate information, not involving the necessity of so stringent a correction, could be obtained by using an experimental set-up with a lower field such that the magnetic correction would set in at an energy lower than the one of primary interest.

It is not only a pleasure but a privilege for me to acknowledge at this point the ever-present help and inspiring guidance of Professor A. H. Compton which have made this work possible. Dr. Haydn Jones has been directly responsible for the construction of most of the apparatus used, and I wish to express my great indebtedness to him. In all the work of taking the photographs and measuring the tracks Mr. F. Leslie Code has been of very great assistance. The help of Mr. Ralph Meagher is also gratefully acknowledged.

APRIL 1, 1940 PHYSICAL REVIEW VOLUME 57

A Theoretical Study of the Diffuse Scattering of X-Rays by Crystals*

W. H. Zachariasen
Ryerson Physical Laboratory, University of Chicago, Chicago, Illinois

(Received January 22, 1940)

A new derivation is made of the intensity expression for the coherent scattering of x-rays by a small crystal. The accepted intensity formula for the diffuse scattering—the well-known Debye formula—is shown to be incorrect and is replaced by a more complicated expression. According to the revised theory the intensity of the diffuse scattering varies much more rapidly with the scattering direction and exhibits a series of diffraction maxima. These maxima are found in the directions $\mathbf{k}_m$ satisfying equations of the type $(1+\lambda\tau_{\min})\mathbf{k}_m=\mathbf{k}_0+\mathbf{B}_H$, where $\mathbf{k}_0$ is the direction of incidence (with $k_m=k_0=1/\lambda$, the reciprocal wave-length) and $\mathbf{B}_H$ a reciprocal lattice vector. The consequences of the theory are discussed in detail.

Introduction

When a beam of parallel and monochromatic x-rays falls on a crystal scattering processes take place. Disregarding the thermal agitation of the atoms in the crystal lattice the intensity of the coherent part of the scattered radiation is given by the well-known Laue expression

$$J_0=Sf^2\prod_i \frac{\sin^2[N^{\frac{1}{3}}\pi(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{a}_i]}{\sin^2[\pi(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{a}_i]}. \quad (1)$$

The equation holds for a small crystal (linear dimensions of the order 10^{-4} cm, for instance) containing N atoms with one atom per unit cell. S is the familiar J. J. Thomson formula for the intensity of scattering from a single free electron, f is the atomic scattering power and $\mathbf{a}_i$ $(i=1,2,3)$ are the edges of the unit cell. The vectors $\mathbf{k}_0$ and $\mathbf{k}$, where $k_0=k=1/\lambda$, represent the directions of incidence and of scattering. According to Eq. (1) there are sharp intensity maxima in the Laue directions which are given by

$$\mathbf{k}-\mathbf{k}_0=\mathbf{B}_H\equiv H_1\mathbf{b}_1+H_2\mathbf{b}_2+H_3\mathbf{b}_3, \quad (2)$$

where H_i designate three integers, $\mathbf{b}_i$ the vector set reciprocal to $\mathbf{a}_i$. For other directions the intensity is zero.

In the famous paper of P. Debye[1] the effect of the thermal vibrations was studied theoretically, and it was found that Eq. (1) had to be replaced by

$$J=J_1+J_2,\quad J_1=J_0e^{-2M},\quad J_2=NSf^2(1-e^{-2M}), \quad (3)$$

where M is a function of temperature and scattering angle. While the first term like the original Laue expression exhibits sharp maxima

---

* The results given in this article were presented in a paper before the American Physical Society at the Chicago meeting December 1, 1939.

[1] P. Debye, Ann. d. Physik **43**, 49 (1914).

in the Laue directions and is zero elsewhere, the part J_2 changes slowly with the scattering angle and represents diffusely scattered x-rays.

From theoretical side objections were raised to Debye's treatment[2] and on the basis of later investigations[3] it was claimed that the diffuse radiation was concentrated under the maxima due to the term J_1 of Eq. (3).

Extensive experimental studies of the diffuse scattering from crystals have been made by G. E. M. Jauncey[4] and collaborators and their results were found to be in general agreement with Debye's formula. It seems that the theoretical conclusions reached by Brillouin, Faxén and Waller never have been seriously considered while the Debye formula on the other hand has been confirmed by more recent theoretical derivations due to Jauncey and Harvey[5] and to Woo.[6]

The writer has made a simplified derivation of the intensity formula for the scattering of x-rays by a small crystal. It is found that the Debye expression for the diffuse scattering is not correct. The new form for J_2 leads to intensity maxima in certain specific directions. These diffraction maxima may coincide with the Laue maxima (as claimed by Brillouin, by Faxén and by Waller). However, under proper conditions the maxima will not coincide with the Laue directions, thus giving rise to new diffraction spots in x-ray photographs. In the following sections the theory of this new effect will be developed and discussed while a preliminary experimental study will be reported upon in a following article.

1. Derivation of the Intensity Expression for the Coherent Scattering of X-Rays by a Small Crystal

For the sake of convenience let it be supposed that the small crystal is a parallellepiped with edges $(N^{\frac{1}{3}}-1)\mathbf{a}_i$ (where $N^{\frac{1}{3}}-1$ is assumed to be an even number) and that there is one atom per unit cell. The equilibrium positions of the atoms may consequently be described by the vectors

$$\mathbf{r}_l=\sum_i l_i\mathbf{a}_i,\quad -\tfrac{1}{2}(N^{\frac{1}{3}}-1)\leqq l_i\leqq\tfrac{1}{2}(N^{\frac{1}{3}}-1),\quad (4)$$

where $l_1l_2l_3$ is a set of integers. If $\mathbf{\Delta}_l$ is the displacement of the atom at $\mathbf{r}_l$ the intensity of the coherent scattering is

$$J=Sf^2\sum_l\sum_{l'}\exp\left[i2\pi(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}_{ll'}\right]\times\langle\exp\left[i2\pi(\mathbf{k}-\mathbf{k}_0)\cdot(\mathbf{\Delta}_l-\mathbf{\Delta}_{l'})\right]\rangle_{\mathrm{Av}}\quad (5)$$

with

$$\mathbf{r}_{ll'}=\sum_i(l_i-l_i')\mathbf{a}_i.$$

One finds readily

$$\langle\exp\left[i2\pi(\mathbf{k}-\mathbf{k}_0)\cdot(\mathbf{\Delta}_l-\mathbf{\Delta}_{l'})\right]\rangle_{\mathrm{Av}}=\exp\{-s^2[\langle(\mathbf{u}\cdot\mathbf{\Delta}_l)^2\rangle_{\mathrm{Av}}-\langle(\mathbf{u}\cdot\mathbf{\Delta}_l)(\mathbf{u}\cdot\mathbf{\Delta}_{l'})\rangle_{\mathrm{Av}}],\quad (6)$$

where $s=2\pi|\mathbf{k}-\mathbf{k}_0|=4\pi\sin\theta/\lambda$ while $\mathbf{u}$ is a unit vector along $\mathbf{k}-\mathbf{k}_0$. With the abbreviations

$$2M=s^2\langle(\mathbf{u}\cdot\mathbf{\Delta}_l)^2\rangle_{\mathrm{Av}},\quad P_{ll'}=s^2\langle(\mathbf{u}\cdot\mathbf{\Delta}_l)(\mathbf{u}\cdot\mathbf{\Delta}_{l'})\rangle_{\mathrm{Av}}\quad (7)$$

and with $e^{P_{ll'}}\approx1+P_{ll'}$ (which is justifiable except for very high temperatures) Eq. (5) becomes

$$J=J_1+J_2,\quad J_1=J_0e^{-2M},$$
$$J_2=Sf^2e^{-2M}\sum_l\sum_{l'}P_{ll'}\exp\left[i2\pi(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}_{ll'}\right].\quad (8)$$

The two terms J_1 and J_2 correspond to Laue scattering and diffuse scattering; but while J_1 agrees with the expression found by Debye (compare Eq. (3)) a different formula is obtained for J_2.

The problem of finding the normal coordinates for the vibrations of a simple lattice is well known[7] and needs but little discussion. By means of a Fourier series expansion the displacement function $\mathbf{\Delta}_{(r,\,t)}$ (which is given physical significance only at the lattice points since an atom is assumed to be displaced as a whole) is represented as a superposition of elastic waves

$$\mathbf{\Delta}_{(r,\,t)}=\sum_j\sum_p{}'\mathbf{A}_{p,\,j}(t)\exp\left[i2\pi\boldsymbol{\tau}_p\cdot\mathbf{r}\right].\quad (9)$$

In this expansion the index j refers to the three waves associated with each propagation vector

---

[2] E. Schrödinger, Physik Zeits. **15**, 79 and 497 (1914); H. Faxén, Ann. d. Physik **54**, 615 (1918).

[3] L. Brillouin, Ann. de physique (9) **17**, 120 (1922); H. Faxén, Zeits. f. Physik **17**, 266 (1923); I. Waller, Diss. Uppsala (1925), Zeits. f. Physik **17**, 398 (1923).

[4] A great many articles in *The Physical Review*, particularly during the last 10 years.

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

[6] Y. H. Woo, Phys. Rev. **38**, 1 (1931) and **41**, 21 (1932).

[7] M. Born and Th. v. Kármán, Physik. Zeits. **13**, 294 (1912); *ibid.* **14**, 15 and 65 (1913). M. Born, *Dynamik d. Kristallgitter* (Leipzig, 1915). I. Waller, reference 3.

$\boldsymbol{\tau}_p$. The vectors $\boldsymbol{\tau}_p$ are given by

$$\boldsymbol{\tau}_p=\sum_i \frac{p_i}{N^{\frac{1}{3}}-1}\mathbf{b}_i, \tag{10}$$

where p_i denotes an integer. Because of the periodicity the integers p_i may be restricted to lie within the limits $-\frac{1}{2}(N^{\frac{1}{3}}-1)\leq p_i \leq \frac{1}{2}(N^{\frac{1}{3}}-1)$. Accordingly all propagation vectors $\boldsymbol{\tau}_p$ lie within a corresponding region—the Brillouin zone—which in the present case is the unit cell of the reciprocal lattice. There are two standing waves for every set of values $(\boldsymbol{\tau}_p, j)$ and one verifies readily that the combination $(-\boldsymbol{\tau}_p, j)$ gives the same two waves. It suffices consequently to consider the $\boldsymbol{\tau}_p$-values lying in one-half of the Brillouin zone. (The summation over half the Brillouin zone is indicated in Eq. (9) by the symbol $\sum_p{}'$, while $\sum_p$ will be used for summation over the entire Brillouin zone. Clearly $\sum_p{}'=\frac{1}{2}\sum_p$.)

Since Eq. (9) corresponds to the introduction of normal coordinates it follows that

$$\begin{aligned}\langle(\mathbf{u}\cdot\boldsymbol{\Delta}_l)^2\rangle_{\text{Av}}&=\tfrac{1}{2}\sum_j\sum_p \gamma_{p,j}{}^2\langle A_{p,j}{}^2\rangle_{\text{Av}},\\ \langle(\mathbf{u}\cdot\boldsymbol{\Delta}_l)(\mathbf{u}\cdot\boldsymbol{\Delta}_{l'})\rangle_{\text{Av}}&\\ =\tfrac{1}{2}\sum_j\sum_p &\gamma_{p,j}{}^2\langle A_{p,j}{}^2\rangle_{\text{Av}}\cos 2\pi\boldsymbol{\tau}_p\cdot\mathbf{r}_{ll'}.\end{aligned} \tag{11}$$

$\gamma_{p,j}$ is cosine of the angle between $\mathbf{A}_{p,j}$ and $\mathbf{u}$. The average energy associated with one vibrational mode is $\frac{1}{2}Nm_a\omega_j{}^2\langle A_{p,j}{}^2\rangle_{\text{Av}}$, m_a being the atomic mass. (The factor of $\frac{1}{2}$ enters because there are two standing waves for every term of Eq. (9).) Accordingly one has

$$\langle A_{p,j}{}^2\rangle_{\text{Av}}=\frac{Q_j}{2\pi^2Nm_a\nu_j{}^2},\quad Q_j=\frac{h\nu_j}{e^{h\nu_j/kT}-1}+\tfrac{1}{2}h\nu_j. \tag{12}$$

The density of points $\boldsymbol{\tau}_p$ in the Brillouin zone is NV, where $V=(\mathbf{a}_1\mathbf{a}_2\mathbf{a}_3)$ is the volume of the unit cell. The summation over the discrete points in the Brillouin zone may conveniently be replaced by an integration over a continuous distribution having the same density. One finds then

$$2M=\frac{s^2V}{4\pi^2m_a}\sum_j\int\gamma_j{}^2\frac{Q_j}{\nu_j{}^2}dv, \tag{13}$$

$$P_{ll'}=\frac{s^2V}{4\pi^2m_a}\sum_j\int\gamma_j{}^2\frac{Q_j}{\nu_j{}^2}\cos[2\pi\boldsymbol{\tau}\cdot\mathbf{r}_{ll'}]dv. \tag{14}$$

Upon substitution of Eq. (14) in Eq. (8) and summation over all pairs of atoms in the crystal the intensity J_2 becomes

$$J_2=Sf^2e^{-2M}\frac{s^2V}{4\pi^2m_a}\sum_i\int\gamma_j{}^2\frac{Q_i}{\nu_j{}^2}\times\left\{\prod_i\frac{\sin^2[N^{\frac{1}{3}}\pi(\mathbf{k}-\mathbf{k}_0+\boldsymbol{\tau})\cdot\mathbf{a}_i]}{\sin^2[\pi(\mathbf{k}-\mathbf{k}_0+\boldsymbol{\tau})\cdot\mathbf{a}_i]}\right\}dv. \tag{15}$$

The integrand in Eq. (15) is zero unless $\mathbf{k}-\mathbf{k}_0+\boldsymbol{\tau}\approx\mathbf{B}_H$ and it is therefore convenient to introduce a small vector $\boldsymbol{\varepsilon}=\sum_i\epsilon_i\mathbf{b}_i$ defined by

$$\boldsymbol{\varepsilon}=\boldsymbol{\tau}-\boldsymbol{\tau}_0,\quad \mathbf{k}-\mathbf{k}_0+\boldsymbol{\tau}_0=\mathbf{B}_H. \tag{16}$$

Noting that $\nu_j=\tau v_j$ (where v_j is the propagation velocity) and that $Vdv=d\epsilon_1d\epsilon_2d\epsilon_3$ Eq. (15) may be given the form

$$J_2=Sf^2e^{-2M}\frac{s^2}{4\pi^2m_a}\sum_i\int\int\int\frac{\gamma_j{}^2Q_j}{v_j{}^2|\boldsymbol{\tau}_0+\boldsymbol{\varepsilon}|^2}\times\prod_i\frac{\sin^2[N^{\frac{1}{3}}\pi\epsilon_i]}{\sin^2\pi\epsilon_i}d\epsilon_i. \tag{17}$$

The velocities v_j are in general functions of the vibration direction; but (unless the anisotropic character of the crystal is pronounced) the range of variation is rather small, so that the velocities to a first approximation may be treated as constants. As a rule the three velocities v_j are not greatly different from one another, and as an admittedly rough approximation it will be assumed therefore that $v_1=v_2=v_3=v=$ a constant. With these assumptions the integration of Eq. (17) may be carried out approximately[8] and the following result is obtained

$$J_2\approx I_0\left(\frac{e^2}{mc^2R}\right)^2\frac{1+\cos^2 2\theta}{2}f^2e^{-2M}N\frac{4\sin^2\theta}{\lambda^2m_av^2}\times\frac{2\pi(NV)^{\frac{2}{3}}Q(\tau_0)}{1+2\pi(NV)^{\frac{2}{3}}\tau_0{}^2}. \tag{18}$$

The corresponding approximate expression for $2M$ is obtained from Eq. (13) when the Brillouin zone is replaced by a sphere of equal volume. The radius of this sphere is $\tau_m=(3/4\pi V)^{\frac{1}{3}}$ and

---

[8] Each of the three sine functions $\sin^2[N^{\frac{1}{3}}\pi\epsilon_i]/\sin^2\pi\epsilon_i$ of the integrand was replaced by smoothing functions $N^{\frac{2}{3}}\exp[-\pi N^{\frac{2}{3}}\epsilon_i{}^2]$.

the formula for $2M$ becomes

$$2M=\frac{s^2V\tau_m kT}{\pi m_a v^2}\{\phi_{(x_m)}+\tfrac{1}{4}x_m\}, \tag{19}$$

where

$$x_m=\frac{hv\tau_m}{kT}\quad\text{and}\quad \phi_{(x)}=\frac{1}{x}\int_0^x\frac{d\xi}{e^\xi-1}.$$

The customary form is obtained when the characteristic temperature Θ is introduced by means of the relation $hv\tau_m=k\Theta$.

2. Discussion

The intensity of the diffuse scattering changes according to Eq. (18) rapidly with the scattering angle and exhibits maxima in the directions for which τ_0 assumes minimum values. This result contrasts sharply with the slow variation with scattering angle predicted on the basis of the Debye formula, but agrees with the statements made by Faxén and Waller[3] that the main part of the diffuse scattering is to be found in the Laue directions. The lack of confirmation of Debye's equation is not surprising since Debye at one stage in the derivation incorrectly assumed neighboring atoms to scatter independently. (The same mistake was made in the later derivations of the Debye formula by Jauncey and Harvey and by Woo.) In the notation of this article Faxén's expression for the diffuse scattering becomes

$$J_2=Sf^2e^{-2M}N\frac{kT}{V\tau_0^4}\left\{\frac{(\mathbf{B}_H\times\boldsymbol{\tau}_0)^2}{c_{44}}+\frac{(\mathbf{B}_H\cdot\boldsymbol{\tau}_0-\tau_0^2)^2}{c_{11}}\right\},$$

where c_{44} and c_{11} are elastic constants. It is readily seen that Faxén's equation agrees rather well with the first approximation to Eq. (18) when $2\pi(NV)^{\frac{2}{3}}\tau_0^2\gg 1$ and when $hv\tau_0/kT\ll 1$. Faxén's discussion of the physical significance of the formula he obtained is unfortunately incomplete and some of the most interesting consequences escaped his notice.

Let an incident beam of parallel, monochromatic x-rays fall on a stationary crystal. The vector $\mathbf{k}_0$ is then sharply defined relative to the crystal lattice and Eq. (18) gives the intensity of the diffuse scattering as function of the scattering direction $\mathbf{k}$. The quantity τ_0 is defined by the relation $\mathbf{k}-\mathbf{k}_0+\boldsymbol{\tau}_0=\mathbf{B}_H$, where as stated the vector $\mathbf{B}_H$ is unique. Consider a particular scattering direction $\mathbf{k}$. The vector $\mathbf{B}_H$ associated with it is readily found from a construction in the reciprocal lattice as shown in Fig. 1. (Cf. the article by Faxén.[3]) It is seen that the sought vector $\mathbf{B}_H$ is the reciprocal lattice vector having the shortest separation from the vector $\mathbf{k}-\mathbf{k}_0$, this separation being the vector $\boldsymbol{\tau}_0$. Consider next the direction of scattering as variable. The terminus of $\mathbf{k}$ describes then a sphere of radius $1/\lambda$ about the terminus of $-\mathbf{k}_0$; this is the "sphere of reflection." The points on the surface of the sphere of reflection which lie closer to $\mathbf{B}_H$ than to any other lattice point define a sector of the solid angle 4π, such that the same vector $\mathbf{B}_H$ is associated with all directions $\mathbf{k}$ within the sector. In this manner all scattering directions are grouped into a finite number of sectors, each sector being characterized by a different vector $\mathbf{B}_H$. The number of different sectors is approximately

$$n\approx 8\pi V\tau_m/\lambda^2=15.6\ V^{\frac{2}{3}}/\lambda^2. \tag{20}$$

As $\mathbf{k}$ varies within a given sector, τ_0 varies with it by virtue of $\mathbf{k}-\mathbf{k}_0+\boldsymbol{\tau}_0=\mathbf{B}_H$ and the intensity changes accordingly. It is important to note that τ_0 can become zero only by proper adjustment of the incident wave vector $\mathbf{k}_0$. Since $\mathbf{k}_0$ and $\mathbf{B}_H$ are considered fixed vectors while $\mathbf{k}$ has the definite length $1/\lambda$, τ_0 assumes a

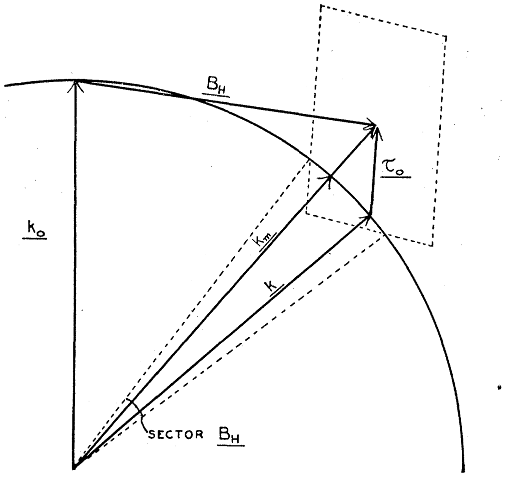

FIG. 1. Showing a section of the reciprocal lattice and illustrating the geometrical interpretation of Eqs. (16) and (22).

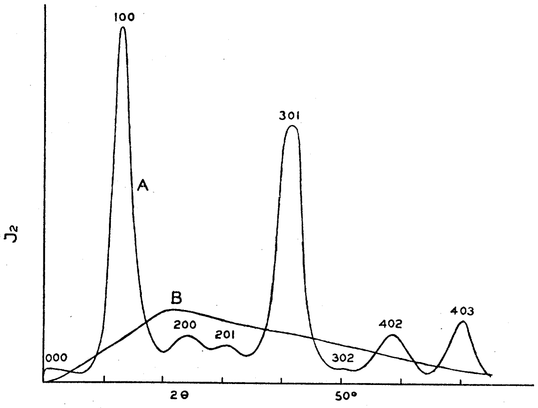

Fig. 2. Showing the intensity distribution of the diffuse scattering for KCl. The crystal is treated as a simple cubic structure with lattice constant $a=3.14$A and one atom per unit cube. The direction of incidence is assumed to be along $\mathbf{a}_3$, while the intensity distribution in the plane $\mathbf{a}_2=0$ is given in the figure. The wave-length is taken to be $\lambda=0.71$A. The curve A is calculated from Eq. (18), curve B from the Debye formula. The three integers affixed to the maxima of curve A are the Miller indices of the active lattice planes. The different behavior of the two intensity curves as the scattering angle goes to zero is of interest. As the scattering angle decreases to small values the intensity according to Eq. (18) approaches first the constant value $J_2=NSZ^2kT/m_av^2$ (Z being the atomic number) and drops then rapidly to zero in the exact forward direction.

minimum value when $\mathbf{k}$ is parallel to $\mathbf{B}_H+\mathbf{k}_0$. If $\tau_{\min}$ is this minimum value one has consequently

$$\tau_{\min}=|\mathbf{B}_H+\mathbf{k}_0|-|\mathbf{k}|$$

or

$$\lambda\tau_{\min}=\left[1+8\sin\theta_B\cos\frac{\theta_B+\theta_i}{2}\sin\frac{\theta_B-\theta_i}{2}\right]^{\frac{1}{2}}-1\approx-\Delta\sin 2\theta_B, \qquad (21)$$

where θ_B is the Bragg angle and $\theta_i\equiv\theta_B+\Delta$ the glancing angle of incidence relative to the sequence of lattice planes normal to $\mathbf{B}_H$. The intensity of scattering reaches a maximum value when τ_0 is a minimum and the direction of the intensity maximum, $\mathbf{k}_m$, is therefore given by

$$[1+\lambda\tau_{\min}]\mathbf{k}_m=\mathbf{B}_H+\mathbf{k}_0. \qquad (22)$$

Thus the direction of maximum intensity lies in the plane of incidence, but the glancing angle of scattering is in general not equal to the glancing angle of incidence. When $\theta_i-\theta_B\equiv\Delta$ is small the scattering angle, $2\theta_m$, corresponding to the maximum becomes

$$2\theta_m\approx 2\theta_B+2\Delta\sin^2\theta_B. \qquad (23)$$

For scattering directions $\mathbf{k}$ near a maximum, $\mathbf{k}_m$, one may set

$$(\lambda\tau_0)^2=(\lambda\tau_{\min})^2+\chi^2, \qquad (24)$$

where χ is the angle between $\mathbf{k}$ and $\mathbf{k}_m$. According to Eqs. (18) and (24) the intensity distribution is symmetrical about a maximum (in the immediate neighborhood of the maximum). The half-width at half-maximum is evidently

$$\chi_{\frac{1}{2}}=\lambda[\tau_{\min}{}^2+1/2\pi(NV)^{\frac{2}{3}}]^{\frac{1}{2}}\approx\lambda|\tau_{\min}|$$
$$=|\Delta|\sin 2\theta_B. \qquad (25)$$

The number of different intensity maxima of J_2 corresponding to a fixed incident wave is given by Eq. (20) since there is one maximum for every sector $\mathbf{B}_H$.

The difference between the Debye formula and ours is well illustrated in Fig. 2 which shows the diffuse (coherent) scattering for a concrete example as calculated according to the two formulas. It becomes clear from this figure that the Debye formula cannot even be considered as a fair approximation. With this fact in mind it is surprising that the extensive experimental studies of the diffuse scattering by Jauncey and collaborators not only failed to reveal the presence of the intensity maxima but even led to confirmation of the Debye expression which we now know to be incorrect. However, reference to the articles describing these experiments shows that the observations were made with highly divergent incident beams and that in all but a few cases continuous rather than monochromatic radiation was used. The aperture of the ionization chamber corresponded furthermore to a large solid angle and it seems that the precaution to remove the warped surface layers of the crystals by etching was not taken. Under these circumstances neither $\mathbf{k}_0$ nor $\mathbf{k}$ are well defined, and the observations should accordingly be compared with the average value of J_2 over large variations of the vector $\mathbf{k}-\mathbf{k}_0$. Suppose that the variation in $\mathbf{k}-\mathbf{k}_0$ is so large that the vector $\boldsymbol{\tau}_0$ traces out the entire Brillouin zone. The average value J_2 is then obtained when the quantity

$$\frac{2\pi(NV)^{\frac{2}{3}}Q(\tau_0)}{1+2\pi(NV)^{\frac{2}{3}}\tau_0{}^2}$$

in Eq. (18) is replaced by its average value.

One finds readily

$$\left\langle\frac{2\pi(NV)^{\frac{2}{3}}Q(\tau_0)}{1+2\pi(NV)^{\frac{2}{3}}\tau_0{}^2}\right\rangle_{\text{Av}}\approx 4\pi kTV\tau_m[\phi_{(x_m)}+\tfrac{1}{4}x_m] \quad (26)$$

and hence with reference to Eq. (19)

$$\langle J_2\rangle_{\text{Av}}=NSf^2e^{-2M}2M\approx NSf^2[1-e^{-2M}], \quad (27)$$

which is the Debye expression. The experimental results of Jauncey and co-workers are consequently useless as a means of distinguishing between Eq. (18) and the Debye formula. Further detailed studies of the diffuse scattering with well defined x-ray beams are therefore desirable and have been begun in this laboratory.

According to Eq. (22) the directions of the intensity maxima of J_2 are uniquely fixed only if the incident wave is monochromatic and if the crystal is stationary. The directions $\mathbf{k}_m$ vary with the rotation angle when the crystal is rotating and wave-length held fixed, so that the diffuse scattering maxima will be recorded as streaks on rotating crystal photographs.[9]

If the crystal is held stationary while the wave-length is varied continuously (as in the Laue method of crystal analysis) the directions $\mathbf{k}_m$ remain in the fixed planes defined by $\mathbf{k}_0$ and the active vectors $\mathbf{B}_H$, but the scattering angles, $2\theta_m$, change with the wave-length. The diffraction maxima J_2 accordingly appear as sections of radial straight lines on a photographic plate placed normal to the incident beam. Such radial streaks have been observed in Laue photographs of various crystals[10] and different interpretations have been suggested. The radial streaks should according to our theory be a general feature of all Laue photographs; but whether they are observed or not is a question of exposure times since the intensity of the streaks is small, particularly for crystals with high characteristic temperatures. The suggestion that the radial streaks of Laue photographs might be due to the temperature effect was first made by Faxén in his paper of 1923.[3] Quite recently this suggestion was experimentally confirmed by Preston[10] who showed that the intensity of the streaks increased with the temperature. Preston's conclusion that the thermal vibrations of the lattice breaks the crystal up into groups consisting of an atom and its next neighbors cannot be accepted, however.[11]

The exact treatment of the theory as applied to crystals containing different kinds of atoms becomes very complicated and will not be attempted in this article. A first approximation may, however, be obtained by assuming that the atoms of a unit cell vibrate as a single particle, and one finds then

$$J_2\approx I_0\left(\frac{e^2}{mc^2R}\right)^2\frac{1+\cos^2 2\theta}{2}|F|^2e^{-2M}N \times\frac{4\sin^2\theta}{\lambda^2v^2\sum m_a}\frac{2\pi(NV)^{\frac{2}{3}}Q(\tau_0)}{1+2\pi(NV)^{\frac{2}{3}}\tau_0{}^2}, \quad (28)$$

where

$$F=\sum f\exp[i2\pi(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}]$$

is the structure factor, $\sum m_a$ the mass of the unit cell and N the number of unit cells contained in the crystal, while the expression for M must be correspondingly modified.

---

[9] These streaks of rotating crystal photographs are not to be confused with another set of streaks due to Laue scattering (J_1) of a continuous component of the incident radiation.

[10] See for instance: W. Friedrich, Physik. Zeits. **14**, 1082 (1913); A. P. R. Wadlund, Phys. Rev. **53**, 843 (1938); G. D. Preston, Proc. Roy. Soc. **172**, 116 (1939).

[11] In a few instances the observed radial streaks must be attributed to disorders in the lattice tending to produce essentially two-dimensional super-lattices. The present writer suggested (Phys. Rev. **53**, 844 (1938)) that Wadlund's observations on rocksalt might be explained as two-dimensional lattice effects. In view of the results of this article it is quite certain that the streaks observed by Wadlund are due to diffuse scattering.