Diffuse Reflection of X-Rays by Anisotropic and Isotropic Solids

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THE elastic vibrations of a single crystal cause an extension of reflecting power around each reciprocal lattice point, the intensity of reflection along a given direction depending essentially upon the frequencies of the vibrations propagated in that direction and upon the angular relation of the directions of oscillation to the directions of reciprocal lattice vectors.

The expression for the frequencies introduces the elastic constants of the crystal, as has been shown by Waller in his dissertation;[1] and one of us has shown[2] that for a monatomic cubic crystal the general Waller expression can be used to obtain the shapes of the isodiffusion surfaces in the close neighborhood of the reciprocal lattice points (L, M, N) in the following form:

$$
\begin{aligned}
r^2(l, m, n) = [&L^2\{c_{44}{}^2+c_{44}(c_{11}-c_{44})(m^2+n^2)+(c_{11}+c_{12})(c_{11}-c_{12}-2c_{44})m^2n^2\} \\
+&M^2\{c_{44}{}^2+c_{44}(c_{11}-c_{44})(n^2+l^2)+(c_{11}+c_{12})(c_{11}-c_{12}-2c_{44})n^2l^2\} \\
+&N^2\{c_{44}{}^2+c_{44}(c_{11}-c_{44})(l^2+m^2)+(c_{11}+c_{12})(c_{11}-c_{12}-2c_{44})l^2m^2\} \\
-&2MNmn(c_{12}+c_{44})\{c_{44}+(c_{11}-c_{12}-2c_{44})l^2\}-2NLnl(c_{12}+c_{44})\{c_{44}+(c_{11}-c_{12}-2c_{44})m^2\} \\
&\qquad\qquad -2LMlm(c_{12}+c_{44})\{c_{44}+(c_{11}-c_{12}-2c_{44})n^2\}] \\
\div [&c_{11}c_{44}{}^2+c_{44}(c_{11}+c_{12})(c_{11}-c_{12}-2c_{44})(m^2n^2+n^2l^2+l^2m^2) \\
&\qquad\qquad +(c_{11}+2c_{12}+c_{44})(c_{11}-c_{12}-2c_{44})^2l^2m^2n^2],
\end{aligned}
$$

where r is the radius of the isodiffusion surface measured from the reciprocal lattice point along the direction (l, m, n); L, M, N, l, m, n being direction cosines. For particular values of $(L, M, N)(l, m, n)$ this expression is readily simplified; a useful way of expressing the results graphically for any particular crystal (that is, for any particular relative values of c_{11}, c_{12}, c_{44}) is to do so by means of a stereographic projection of the isodiffusion surface for each lattice point (L, M, N), a method which reveals very sharply the differences between the reciprocal lattice points of a given anisotropic solid.[3]

Measurement of the diffuse reflecting regions for various planes of sodium single crystals (for which $c_{11}=0.52$, $c_{12}=0.40$, $c_{44}=0.41\times10^{11}$ dynes/cm²)[4] are found to be in excellent agreement with the above formula. The observations were made, with radiation from a copper target, on a succession of Laue photographs taken at small angular intervals in various crystal orientations, over ranges covering the observable regions of

reflecting power for all possible planes.[5] Similar photographs of lithium single crystals, which are isomorphous with those of sodium (body-centered cubic structure), indicated that the relative values of the elastic constants of this very light metal are similar to those of sodium. The formula given above applies to all cubic lattices, and in proof of this it has been found that lead single crystals, which are face-centered cubic in structure, but elastically anisotropic in the same sense as sodium $(c_{11}-c_{12}<2c_{44})$ give photographs leading to similar reflecting regions.

For all cubic crystals so far examined $c_{11}>c_{44}$; for some ionic compounds, such as KCl, NaCl, the Cauchy relation $c_{12}=c_{44}$ is fulfilled and $c_{11}\gg c_{44}$. This implies a very small extension of reflecting power along the reciprocal lattice vector itself, for all values of (L, M, N). This and other more detailed consequences of the theory are confirmed by the observations and measurements of Knaggs, Lonsdale and Smith upon single crystals of KCl and NaCl, using the radiation from molybdenum and silver targets.[6]

[1] Waller, Dissertation, Uppsala (1925).

[2] H. A. Jahn, Nature **147**, 511 (1941); Proc. Roy. Soc. London A**179**, 326 (1941).

[3] H. A. Jahn, Proc. Roy. Soc. London (in the press).

[4] S. L. Quimby and S. Siegel, Phys. Rev. **54**, 299 (1938); values extrapolated to room temperatures. See also Fuchs, Proc. Roy. Soc. London A**157**, 444 (1936).

[5] K. Lonsdale and H. Smith, Nature **148**, 628 (1941).

[6] K. Lonsdale and H. Smith, Proc. Roy. Soc. London A**179**, 8 (1941); see also W. H. Zachariasen, Phys. Rev. **59**, 207 (1941) and other papers in the same volume.

One way of simplifying the general formula given above is to use solids for which the condition of isotropy $c_{11}-c_{12}=2c_{44}$ is fulfilled. The expression then becomes

$$r^2(l, m, n; L, M, N)$$

$$=\frac{1}{c_{11}}+\left(\frac{1}{c_{44}}-\frac{1}{c_{11}}\right)[1-(Ll+Mm+Nn)^2]$$

$$=\frac{\cos^2\theta}{c_{11}}+\frac{\sin^2\theta}{c_{44}},$$

where θ is the angle between the directions $l, m, n)$ and (L, M, N). For pure transverse waves (the case originally considered by Faxén) this reduces to $r^2=(1/c_{44})\sin^2\theta$, and the isodiffusion surfaces have the shape of an anchor ring of vanishing internal diameter, obtained by rotating about the reciprocal lattice vector (L, M, N) two equal circles touching at the reciprocal lattice point, their line of centers being *perpendicular* to the reciprocal lattice vector. The surfaces corresponding to pure longitudinal waves alone, given by $r^2=\cos^2\theta/c_{11}$, would be pairs of equal spheres touching at the reciprocal lattice point, the line of centers being *along* (L, M, N). It is to be noted that even for isotropic solids the formula

$$r^2=(1/c_{11})\cos^2\theta+(1/c_{44})\sin^2\theta$$

leads to a whole series of possible shapes according to the ratio $c_{11}:c_{44}$. Tungsten, for which $c_{11}=51.3$, $c_{12}=20.6$, $c_{44}=15.3\times10^{11}$ dynes/cm²,[7] is an example of a hard, isotropic, body-centered cubic structure. It gives small, elliptical diffuse spot reflections which fade out quickly as the orientation changes, quite unlike the detailed, persistent diffuse spots and streaks obtained for the soft, anisotropic alkali metals and lead.

While Faxén[8] gave a correct formula for the scattering by the longitudinal and transverse waves of an isotropic solid, he also gave an approximate formula $a(\sin\theta+\cos\theta\tan\theta_1)=\lambda$, to which much reference has been made and which has been obtained in various forms by various theoretical processes. This formula is in fact a very poor approximation to the truth. It implies that in the neighborhood of the reciprocal lattice points the isodiffusion surfaces are spheres concentric with those points. This can never really be the case, according to the above theory. For r^2 to be independent of θ, c_{11} must be equal to c_{44}. Since $c_{11}-c_{12}=2c_{44}$, that would mean $c_{12}=-c_{11}=-c_{44}$. Either the compressibility $(c_{11}+2c_{12})$ or the shear coefficient (c_{44}) would thus have to be negative, which is clearly impossible. In fact, for positive compressibility and positive shear coefficient, c_{12} can never have a negative value which is numerically greater than $c_{11}/2$, or a positive value which is greater than c_{11}, and therefore the maximum value of c_{44} which fulfills the isotropy condition $c_{11}-c_{12}=2c_{44}$ is $3c_{11}/4$. This sets an upper limit to the extension of the surfaces of isodiffusion, along the direction of the radius vector for each reciprocal lattice point. It can never be more than $\frac{3}{4}$ of the extension in a perpendicular direction, and is usually considerably less.

[7] P. Bridgman, Proc. Am. Acad. Sci. 60, 305 (1925); see also Wright, Proc. Roy. Soc. London A126, 613 (1930).

[8] Faxén, Zeits. f. Physik 17, 266 (1923).