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GENERAL THEORY OF X-RAY DIFFRACTION IN REAL CRYSTALS*

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The use of counter techniques in the study of x-ray diffraction by a single crystal makes it possible to measure the integrated intensities of the diffracted beams to a precision of 1% on a relative scale. However, much physical information contained in experimental data of this accuracy has been inaccessible because a satisfactory theory of diffraction in a real crystal has not been available.

A new theoretical study has led to an approximate formula for the integrated intensity applicable over the entire range from the perfect to the ideally imperfect crystal. This universal intensity expression is based on the usual model of a real crystal as an aggregate of perfect crystal domains in imperfect alignment. It is assumed that the orientation of the domains obeys a Gaussian distribution law $W(\Delta) = g \times \exp(-\pi g^2\Delta^2)$ with Δ the angular deviation from the mean orientation.

The results presented below are given specifically for the case of x-ray diffraction, but they hold also for neutron or electron diffraction if the expression for the quantity Q is appropriately modified.

The universal formula for the integrated intensity, P, is

$$P = I_0 QvAy, \tag{1a}$$

$$Q = |e^2F/mc^2V|^2\lambda^3K^2/\sin 2\theta, \tag{1b}$$

$$A = v^{-1}\int \exp(-\mu T)dv, \tag{1c}$$

$$y = (3x)^{-1/2}\tanh(3x)^{1/2}, \tag{1d}$$

$$x = \alpha Q\{\bar{t} + (\bar{T}-\bar{t})/[1+(\alpha/g)^2]^{1/2}\}, \tag{1e}$$

$$\alpha = 2\bar{t}_\perp/3\lambda. \tag{1f}$$

I_0 is the incident intensity, v the irradiated crystal volume, λ the wavelength, 2θ the scattering angle, F/V the structure factor per unit volume, and μ the linear absorption coefficient. K^2 is the polarization factor so that $K = 1$ for normal and $K = \cos 2\theta$ for parallel polarization. T is the path length through the crystal, and $\bar{T} = A^{-1}dA/d\mu$ its mean value. $\bar{t}$ is the mean path length through a perfect crystal domain, and $\bar{t}_\perp$ the mean thickness of the domain measured normal to the incident beam and parallel to the plane of incidence.

For most purposes it is reasonable to assume the perfect domain to be a sphere of radius r, in which case $\bar{t} = \bar{t}_\perp = 3r/2$, and $\alpha\lambda = r$.

If x and $\mu\bar{T}$ are greater than unity, Eq. (1d) tends to make y too small for given x, and it is possible that $y = (3x)^{-1/2}\tan^{-1}(3x)^{1/2}$ is a better approximation.

The kinematical theory approximation which neglects extinction (i.e., absorption due to energy transfer by diffraction) is the asymptotic form of Eqs. (1) as x goes to zero. It is seen that this approximation is accurate to 1% for $x < 0.01$, which implies either an extremely weak reflection or a highly imperfect crystal.

The dynamical theory is valid only for a perfect crystal.[1,2] The equations of this theory are complicated and have been solved only for an infinite plane-parallel crystal plate. Furthermore, very few crystal specimens exhibit the required degree of perfection. In our model a perfect crystal corresponds to a single-domain situation, so that $t = T$ and $x = \alpha Q\bar{t}$. Equations (1) applied to a perfect crystal plate are, indeed, in good agreement with the predictions of the dynamical theory.

For a strong reflection ($Q \approx 10^{-2}$ cm^{-1}) and the commonly used crystal size of $\bar{T} \approx 10^{-2}$ cm, the value of y will range from 1 to 10^{-2} depending upon the state of perfection of the crystal.

The first term of Eq. (1e), $x = \alpha Q\bar{t}$, represents primary extinction (extinction within a given domain), and the second term, secondary extinction (extinction suffered during passage through other domains). Primary extinction becomes negligible, even for strong reflections, if $\bar{t} < 10^{-4}$ cm.

Experiments indicate that $g > \alpha$ for all crystals exhibiting large extinction effects. In this case $x = \alpha Q\bar{T}$, and values of $x > 1$ will occur for strong reflections if $\bar{t} \geq 10^{-4}$ cm. If α is measured experimentally for the same reflection and the same crystal specimen, the condition $g > \alpha$ requires $\alpha_1\lambda_1 = \alpha_2\lambda_2$.

It has generally been assumed that crystals showing small extinction effects, i.e., $y \approx 1-x$, $x \ll 1$, can be interpreted in terms of the theory of secondary extinction. This theory was developed by Darwin[1] for plane-parallel plates and was recently amended and extended to include crystals of any shape.[3] The theory is based on the assumptions that primary extinction is negligible and that $g \ll \alpha$. According to Eq. (1e) one has then $x = gQ\bar{T}$, and it is indeed true that $x \ll 1$. Since g is a material constant for the specimen, measurements with two wavelengths must give $g_1 = g_2$.

The two cases $x = gQ\bar{T}$ and $x = \alpha Q\bar{T}$, which correspond to rather different physical properties of the crystal medium, are experimentally distinguishable only if different wavelengths are used.

It is believed that the results given above will make it possible to extract more extensive and more accurate physical information from diffraction data.

A detailed account of the rather lengthy derivation which led to Eqs. (1) will be published later elsewhere.

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[2]P. P. Ewald, Ann. Physik. 54, 519, 577 (1917); Z. Physik 2, 232 (1920); 30, 1 (1924); Physik. Z. 26, 29 (1925).

[3]W. H. Zachariasen, Acta Cryst. 16, 1139 (1963); Trans. Am. Cryst. Assoc. 1, 33 (1965).

STANDING WAVES IN SELF-TRAPPED LIGHT FILAMENTS

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Intense light tends to be trapped in very small filaments in an optical material.[1,2] It is shown here that under some conditions of sufficiently high intensity, light waves of approximately equal intensity travel in the backward and forward directions in these filaments, and the resulting standing waves produce visible spatial beats. This effect allows a rather direct detection of substantial changes in the index of refraction within the trapped filaments.

The spatial beats, producing bands or striations, are shown in Fig. 1 for two filaments in CS_2, one of diameter 37 μ and the other of diameter about 2 μ. Examination shows that the striations tend to be spaced at distances which are close to integral multiples of a unit distance, about 5 μ. Not all trapped filaments of light produce these intense striations, but characteristically the more intense filaments do.

It is proposed that laser light is scattered backwards in the filament, probably largely by stimulated Rayleigh scattering.[3] Furthermore, stimulated Raman light corresponding to the first Stokes line is generated also, in both forward and backward directions. Hence,