THE PHASE PROBLEM AND THE CORRELATION PROPERTIES OF X-RAY BEAMS

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Abstract

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THE PHASE PROBLEM AND THE CORRELATION PROPERTIES OF X-RAY BEAMS

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In coherent scattering of X-rays by the electron density of a crystal $\rho(\mathbf{r})$, the intensities of reflections I_{hkl} from planes hkl satisfying the Bragg angles are proportional to the squares of the structure amplitudes $I_{hkl} \sim |F_{hkl}|^2$. The coefficients F_{hkl} in the case of non-centrosymmetric crystals are complex quantities $F_{hkl} = |F_{hkl}|e^{i\alpha_{hkl}}$, and since in experiment one obtains data only on the squares of the moduli of the structure amplitudes, the problem arises of determining the phases α_{hkl}. The use of features of the interaction of electromagnetic radiation with an object sometimes makes it possible to solve the phase problem, for example, by the heavy-atom method ([6]). Below we shall give a justification for the use of two-beam interference of electromagnetic radiation in the X-ray wavelength range for determining phases in crystals.

Let two beams be incident on the recording instrument; according to the superposition principle, they create at the point of incidence a field equal to a linear combination of the fields

$$E = \xi_1^0 E_1 + \xi_2^0 E_2, \tag{1}$$

where $\xi_{1,2}^0$ are complex numbers characterizing the fields; $E_{1,2}$ is the field strength in the classical interpretation or the photon annihilation operator in the quantum sense, $E_{1,2} \simeq CE_{1,2}^{(-)}$.

Since in experiment the time-averaged intensity is recorded, from (1) we obtain

$$\langle |E|^2 \rangle = |\xi_1|^2 G^{(1)}(x_1 x_1) + |\xi_2|^2 G^{(1)}(x_2 x_2) + 2\,\mathrm{Re}\{\xi_1 \xi_2^* G^{(1)}(x_1 x_2)\}, \tag{2}$$

where $\langle ... \rangle$ denotes time averaging; $G^{(1)}(x_{ix}j) = \langle E_i^{(+)} E_j^{(-)} \rangle$.

The function $G^{(1)}(x_{ix}j)$ is called the first-order correlation function ([1]). The physical meaning of $G^{(1)}(x_1 x_2)$ is that it describes the correlation between the positions of photons at two space-time points x_1 and x_2.

Taking further into account that the radiation beams possess a certain thickness and a spectral distribution specified by the function $g(\omega)$, we obtain the basic expression for the integral intensity

$$I = \iint_{\omega\sigma} \{|\xi_1|^2 G^{(1)}(x_1 x_1) + |\xi_2|^2 G^{(1)}(x_2 x_2) + 2\,\mathrm{Re}[\xi_1 \xi_2^* G^{(1)}(x_1 x_2)]\} g(\omega)\, d\omega\, d\sigma, \tag{3}$$

σ is the cross-sectional area of the beam; a is the beam width, b its thickness.

Let us consider a specific experimental arrangement: two beams are incident on a synthesizer crystal in such a way that the diffracted beams emerge in the same direction.

Then the quantities entering into (3) have the following meaning: ξ_j describes the interaction of the radiation with the synthesizing crystal and is equal to [(2)]

$$\xi_j = P^{1/2} F_j(hkl),$$

$$P = HpL \exp(-2M) \exp(-2\mu l/\cos\theta);$$

$$\mathbf{F}_j = \sum_i f_i \exp 2\pi i \left(\frac{\mathbf{s}_j - \mathbf{s}}{\lambda} \mathbf{r}_i\right) = A_j + iB_j,$$

$j = 1, 2$; H is the multiplicity factor; p is the polarization factor; L is the Laue function; $\exp(-2M)$ is the Debye–Waller factor; $\exp(-2\mu l/\cos\theta)$ is the absorption multiplier; F_j is the structure amplitude of reflection from the j-th plane; $g(\omega)$ is the frequency distribution function, most often having a Lorentzian form: $g(\omega) = [(\omega - \omega_0)^2 + \gamma^2/4]^{-1}$; $|G^{(1)}(x_i x_i)|$ is the counting rate of the recording instrument, or simply the intensity of the synthesized beam, describing the interaction of coherent beams.

In the general case $G^{(1)}(x_i x_j) = |G^{(1)}(x_i x_j)| \exp(i\varphi)$. The appearance of the phase factor $\exp(i\varphi)$ is connected with the fact that the interfering waves traverse optically different paths and acquire a phase shift

$$\varphi = 2\pi \frac{\omega}{c} \frac{2l}{\cos\theta} \Delta,$$

where

$$\Delta = \frac{r_e^2 e^2}{mc^2} \sum_k 2\pi N_k f_k(0);$$

Δ is the difference in the refractive indices of the surrounding medium and the crystal [2]; $f_k(0)$ is the scattering factor of the k-th atom for forward scattering; N_k is the number of atoms of the k-th kind per unit volume.

If $|G^{(1)}(x_1x_2)| \neq 0$, then the beams are coherent, have the same or a similar space-time structure, and produce a stable interference pattern.

Carrying out the integration in (3), we obtain

$$I_{1,2} - (I_1 + I_2) = 2bP|G^{(1)}(x_1x_2)|\xi \int_0^a \{(A_1A_2 + B_1B_2)\cos k\omega_0 l +$$

$$+(A_1B_2 - A_2B_1)\sin k\omega_0 l\}\, dl, \tag{4}$$

where $I_{1,2}$ is the total intensity under the simultaneous action of both beams; I_1, I_2 are the intensities of each separately;

$$\zeta = -\frac{2\pi}{\gamma}\exp\left(-\frac{\gamma}{2}kl\right); \quad k = \frac{4\pi\Delta}{c\cos\theta}.$$

Analysis of expression (4) shows that appreciable interference occurs when ${}^1/_2\gamma kl \leqslant 1$. This condition means that the width of the spectral line must be much smaller than the reciprocal of the time of traversal by the beams of the optical path difference. The limiting path difference at which interference is still observed is called the coherence interval l_k. For characteristic x-ray radiation $l_k \sim 10^{-5} \div 10^{-6}$ cm. By reflection from absolutely ideal crystals, the coherence interval can apparently be increased to 10^{-4} cm. Monochromatic gamma radiation of Mössbauer transitions in nuclei has $l_k \sim 10^{-1} \div 10^4$ cm.

Bonse and Hart experimentally showed [3] that, using monochromatic radiation and splitting the primary beam with a crystal, one can observe a stable interference pattern consisting of a system of parallel fringes. According to expression (4), the interference pattern will contain only one maximum (minimum) at

$$a = \lambda\cos\theta/4\Delta.$$

Thus, in order to extract the magnitude of the contribution of the interference term in (4), one should use synthesizer crystals of such a size in a that only one interference extremum is contained within the diffraction maximum. Hence $a \sim 10^{-3} \div 10^{-1}$ cm. For $a\Delta \gg \lambda$, the interference term in (4) becomes zero. With a nonzero interference term in (4), the coordinates of the atoms in the unit cell can be determined directly.

Let the structure of the synthesizer crystal be unknown. Integrating in (4) over l and extracting the phase terms, we obtain

$$\sum_{i,j} f_i f_j \cos 2\pi[(\delta_i - \delta_j) + \psi] = Q^{-1}[I_{1,2} - (I_1 + I_2)], \tag{5a}$$

where

$$\delta_{i,j} = 2\pi \left(\frac{\mathbf{s} - \mathbf{s}_{1,2}}{\lambda} \mathbf{r}_{i,j} \right);$$

$$\sin \psi = (\cos k\omega_0 a - 1)[(\cos k\omega_0 a - 1)^2 + \sin^2 k\omega_0 a]^{-1/2};$$

$$\cos \psi = \sin k\omega_0 a[(\cos k\omega_0 a - 1)^2 + \sin^2 k\omega_0 a]^{-1/2};$$

$$Q = 2bP|G^{(1)}(x_1 x_2)|\xi[(\cos k\omega_0 a - 1)^2 + \sin^2 k\omega_0 a]^{1/2}(k\omega_0)^{-1}.$$

After simplification we find

$$\psi = -\frac{k\omega_0 a}{2}, \qquad Q = 4bP|G^{(1)}(x_1 x_2)|\xi \sin \frac{k\omega_0 a}{2}(k\omega_0)^{-1}.$$

If we now introduce a constant phase shift of $\pi/2$ for one beam (placing in its path, for example, a plate of suitable thickness), then one can obtain an expression for the antisymmetric part of the structural amplitude

$$\sum_{i,j} f_i f_j \sin 2\pi[(\delta_i - \delta_j) + \psi] = Q^{-1}[I_{1,2} - (I_1 + I_2)]. \tag{5 }$$

Formally, (5a) resembles the function of a squared crystal (or, correspondingly, the Patterson function) [4,5]. In essence, however, it is an entirely special function with its own specific properties.

Function (5a) is determined together with function (5), which makes it possible to identify its maxima in a unique way. Function (5a) has no origin maxima. All its maxima are resolved, which makes it possible to determine directly the coordinates of the atoms in the unit cell.

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Note: Figure translations are in progress. See original paper for figures.