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Diffraction of partially coherent beams on three-dimensional periodic structures and the angular shifts of the diffraction maxima

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The scattering of electromagnetic fields of arbitrary state of coherence and polarization and of arbitrary spectrum by three-dimensional periodical structures is described in the first-order Born approximation and under assumptions of statistical stationarity and quasihomogeneity of the fields. It is shown that, in general, the angular dependence of the radiant intensity of scattered radiation is influenced and modified by the coherence properties of the incident radiation. The results are illustrated by examples that indicate that when x rays are scattered by a crystal, shifts of diffraction maxima due to partial coherence of incident beam may appear, both in the Laue method and in the powder method.

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I. INTRODUCTION

In recent years a good deal of effort was paid to the study of spectral properties of a partially coherent radiation [1–9]. Closely related to this is scattering of a partially coherent beam on a diffraction grating, which has been examined theoretically in [10] with the aim to take into account the process of spectra measurement. Diffraction of coherent waves on a grating is a well-known process. If the grating is illuminated by a polychromatic plane wave, the angular distribution of the radiation intensity behind the grating is proportional to the spectrum of radiation; i.e., the components of different frequencies are deflected in different directions (neglecting aperture effects). Different situations may occur when the incident radiation is spatially not completely coherent as assumed in [10]. In that paper the diffraction grating was assumed to be illuminated by a beam generated by a secondary quasihomogeneous source of any state of coherence and the scalar approximation was used. It has been shown there that under these circumstances the angular dependence of the intensity of radiation in the far zone behind the grating is given by an integral relation in which there is, beside the spectrum, another frequency-dependent term due to the state of coherence of the radiation. The fact that in the case of the partially coherent beam the grating does not have to "give" the spectrum like in the coherent case has two physical reasons, which cannot, however, be separated in general: The Wolf effect, i.e., the change of spectrum of partially coherent radiation during propagation, and an inherent angular divergence of partially coherent beam. So, it may happen that when a suitably correlated light beam with the spectral line centered around some fixed frequency is incident on the grating, the diffraction maxima will be shifted with respect to those pertaining to the fully coherent beam. This result is of interest in connection with x-ray structure analysis, which determines the internal composition of crystals just from the location of diffraction maxima of the scattered x rays. It is worth mentioning that shifts of x-ray diffraction lines dependent on the degree of coherence of radiation have been already observed [11]. However, diffraction on three-dimensional (3D) gratings corresponding to scattering of x rays on crystals could, in principle, be different from diffraction on two-dimensional periodic structures that we already mentioned. The present paper deals with this problem, i.e., with the theory of scattering of an electromagnetic radiation of any state of coherence on three-dimensional periodic structures within the accuracy of the first-order Born approximation and with the question of coherence-induced angular shifts of diffraction lines in the context of x-ray structure analysis.

II. SCATTERING OF ELECTROMAGNETIC FIELDS FROM PERIODIC STRUCTURES

To describe the scattering of statistically stationary electromagnetic fields of an arbitrary state of coherence and polarization, and with an arbitrary spectrum from three-dimensional quasiperiodic media (with dispersion and space fluctuations in general) within the accuracy of the first-order Born approximation, we modify the theory of Wolf and Foley [12] developed for scattering from spatially and temporally fluctuating media [4,12,13]. The first-order Born approximation means that the scattered radiation does not itself scatter.

Using the method of Hertz vectors and imposing the conditions of statistical stationarity of the radiation as well as the constraints for the applicability of the first-order Born approximation the authors of Ref. [12] have shown that the spectrum of the scattered electric field in the far zone takes the following form [Eqs. (4.8) and (4.11) in [12]]:

$$S(r\boldsymbol{u},\omega) = \int_{-\infty}^{\infty} d\omega'\,(2\pi)^6\,\frac{k^2k'^2}{r^2}\,\exp[i(k'-k)r] \times(\delta_{lm}-u_lu_m)\langle\langle\mathcal{P}_l^*(\boldsymbol{k},\omega)\mathcal{P}_m(\boldsymbol{k}',\omega')\rangle_R\rangle_M. \quad (1)$$

Here $r\boldsymbol{u} = \boldsymbol{r}$ is a position vector with $|\boldsymbol{u}| = 1$, ω is a frequency, $\boldsymbol{k} = k\boldsymbol{u} = (\omega/c)\boldsymbol{u}$ with c being the speed of light in vacuum, δ_{lm} denotes the Kronecker symbol, $l, m = 1, 2, 3$ (or x, y, z), symbols $\langle\rangle_R$ and $\langle\rangle_M$ represent averaging over ensembles of fluctuations of the incident radiation and of the scattering medium, respectively, summation over repeated suffixes being implied. The vector function $\boldsymbol{\mathcal{P}}(\boldsymbol{k},\omega)$ is given by the expression,

$$\boldsymbol{\mathcal{P}}(\boldsymbol{k},\omega) = \frac{1}{(2\pi)^4}\int_V d^3r\,\exp(-i\boldsymbol{k}\cdot\boldsymbol{r}) \times \int_{-\infty}^{\infty} dt\,\exp(i\omega t)\boldsymbol{P}(\boldsymbol{r},t). \tag{2}$$

The spatial integral is taken over the domain occupied by the scatterer and the polarization $\boldsymbol{P}(\boldsymbol{r},t)$ induced by incident (real) electric field $\boldsymbol{E}(\boldsymbol{r},t)$ [14] is defined as follows [15]

$$\boldsymbol{P}(\boldsymbol{r},t) = \frac{1}{2\pi}\int_0^{\infty} dt'\eta(\boldsymbol{r},t')\boldsymbol{E}(\boldsymbol{r},t-t'), \tag{3}$$

where $\eta(\boldsymbol{r},t)$ is the dielectric susceptibility. The "correlation" term in Eq. (1) is given by the expression

$$\langle\langle\mathcal{P}_l^*(\boldsymbol{k},\omega)\mathcal{P}_m(\boldsymbol{k}',\omega')\rangle_R\rangle_M = \frac{1}{(2\pi)^6}\delta(\omega-\omega')\int_V d^3r_1\int_V d^3r_2\,\exp[i\boldsymbol{k}\cdot(\boldsymbol{r}_1-\boldsymbol{r}_2)] \times\langle\tilde{\eta}^*(\boldsymbol{r}_1,\omega)\tilde{\eta}(\boldsymbol{r}_2,\omega)\rangle_M W_{lm}(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega), \tag{4}$$

where $W_{lm}(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega)$ is the cross-spectral tensor of the incident radiation, i.e., the Fourier transform of its cross-correlation tensor (assuming statistical stationarity),

$$W_{lm}(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\tau\,\exp(i\omega\tau) \times\langle E_l(\boldsymbol{r}_1,t)E_m(\boldsymbol{r}_2,t+\tau)\rangle_R, \tag{5}$$

and

$$\tilde{\eta}(\boldsymbol{r},\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\,\exp(i\omega t)\eta(\boldsymbol{r},t). \tag{6}$$

Let us suppose now that the cross-spectral tensor of the incident radiation is quasihomogeneous (at least in the region of scattering medium), i.e., that

$$W_{lm}(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega) = s(\omega)I^{1/2}(\boldsymbol{r}_1)I^{1/2}(\boldsymbol{r}_2)\,\mu_{lm}(\boldsymbol{r}_2-\boldsymbol{r}_1,\omega), \tag{7}$$

where $s(\omega)$ is the normalized spectrum, $I(\boldsymbol{r})$ is the intensity distribution, and $\mu_{lm}(\boldsymbol{R},\omega)$ is the tensor of degree of spectral coherence.

Let us assume that the dielectric susceptibility is a quasiperiodic function of position vector,

$$\tilde{\eta}(\boldsymbol{r},\omega) = T(\boldsymbol{r})\sum_{\boldsymbol{G}}\gamma_{\boldsymbol{G}}(\omega)\,\exp(i\boldsymbol{G}\cdot\boldsymbol{r}). \tag{8}$$

Here $T(\boldsymbol{r})$ is the form factor that characterizes the macroscopic geometric shape of the scatterer (crystal) and $\boldsymbol{G}$ are vectors of the reciprocal lattice. The coefficients $\gamma_{\boldsymbol{G}}(\omega)$ characterize the structure of the lattice. It is assumed that the form factor varies with $\boldsymbol{r}$ much more slowly than the exponentials. In order to shorten the analysis we introduce the tensor function $\Xi_{lm}(\boldsymbol{k},\omega)$ so that

$$S(r\boldsymbol{u},\omega) = (2\pi)^6\,\frac{k^4}{r^2}\,(\delta_{lm}-u_lu_m)\,\Xi_{lm}(\boldsymbol{k},\omega). \tag{9}$$

According Eqs. (4), (7), and (8),

$$\Xi_{lm}(\boldsymbol{k},\omega) = \frac{1}{(2\pi)^6}\int_{R^3} d^3r_1\int_{R^3} d^3r_2\,\exp[i\boldsymbol{k}\cdot(\boldsymbol{r}_1-\boldsymbol{r}_2)] \times X^*(\boldsymbol{r}_1,\omega)X(\boldsymbol{r}_2,\omega)s(\omega)\mu_{lm}(\boldsymbol{r}_2-\boldsymbol{r}_1,\omega). \tag{10}$$

Here the integrations extend over entire space (R^3) because the finite size of the crystal appears in the form factor. Further,

$$X(\boldsymbol{r},\omega) = A(\boldsymbol{r})\sum_{\boldsymbol{G}}\gamma_{\boldsymbol{G}}(\omega)\,\exp(i\boldsymbol{G}\cdot\boldsymbol{r}), \tag{11}$$

with $A(\boldsymbol{r}) = I^{1/2}(\boldsymbol{r})T(\boldsymbol{r})$ being a real function. Setting

$$\mathcal{A}(\boldsymbol{f}) = \frac{1}{(2\pi)^3}\int_{R^3} d^3r\,\exp(-i\boldsymbol{f}\cdot\boldsymbol{r})\boldsymbol{A}(\boldsymbol{r}), \tag{12}$$

we can introduce the quantity

$$\mathcal{X}(\boldsymbol{f},\omega) = \frac{1}{(2\pi)^3}\int_{R^3} d^3r\,\exp(-i\boldsymbol{f}\cdot\boldsymbol{r})X(\boldsymbol{r},\omega) = \sum_{\boldsymbol{G}}\gamma_{\boldsymbol{G}}(\omega)\,\mathcal{A}(\boldsymbol{f}-\boldsymbol{G}). \tag{13}$$

We then find that

$$\Xi_{lm}(\boldsymbol{k},\omega) = s(\omega)\int_{R^3} d^3f\,\mathcal{X}^*(\boldsymbol{f},\omega)\mathcal{X}(\boldsymbol{f},\omega)\hat{\mu}_{lm}(\boldsymbol{k}-\boldsymbol{f},\omega), \tag{14}$$

where

$$\hat{\mu}_{lm}(\boldsymbol{f},\omega) = \frac{1}{(2\pi)^3}\int_{R^3} d^3r\, \exp(-i\boldsymbol{f}\cdot\boldsymbol{r})\mu_{lm}(\boldsymbol{r},\omega). \quad (15)$$

Equation (14) can be easily obtained by the use of the 6D-convolution theorem (for the 4D analogy see [10]). The radiant intensity measured in the far zone in the direction $\boldsymbol{u}$ is proportional to the integral of the spectrum over the whole frequency range, i.e., to the quantity

$$I(r\boldsymbol{u}) = \int_0^\infty d\omega\, S(r\boldsymbol{u},\omega) = (2\pi)^6 \int_0^\infty d\omega\, \frac{k^4}{r^2}\,(\delta_{lm} - u_l u_m) \sum_{\boldsymbol{G}_1}\sum_{\boldsymbol{G}_2} \gamma^*_{\boldsymbol{G}_1}(\omega)\,\gamma_{\boldsymbol{G}_2}(\omega) \times s(\omega)\int_{R^3} d^3f\, \mathcal{A}^*(\boldsymbol{f}-\boldsymbol{G}_1)\mathcal{A}(\boldsymbol{f}-\boldsymbol{G}_2)\,\hat{\mu}_{lm}(\boldsymbol{k}-\boldsymbol{f},\omega), \quad (16)$$

where Eqs. (9), (14), and (13) have been used.

Let us add a few remarks about the function $\hat{\mu}_{lm}(\boldsymbol{f},\omega)$. Since the incident field satisfies the homogeneous wave equation [12] the appropriate electric cross-spectral tensor has to satisfy the "double Helmholtz" equation [8,17,18]. Consequently the function $\hat{\mu}_{lm}(\boldsymbol{f},\omega)$, which represents the spatial Fourier transform of the degree of spectral coherence, must have the form

$$\hat{\mu}_{lm}(\boldsymbol{f},\omega) = \psi_{lm}\left(\frac{\boldsymbol{f}}{|\boldsymbol{f}|},\omega\right)\delta\left(|\boldsymbol{f}| - \frac{\omega}{c}\right). \quad (17)$$

We will use the fact that we are dealing with a beamlike field, i.e., with radiation that propagates close to some chosen direction $\boldsymbol{s}_0$ ($|\boldsymbol{s}_0| = 1$). In such a case $|\psi_{lm}(\boldsymbol{s},\omega)|$ is negligible except when $|\boldsymbol{s} - \boldsymbol{s}_0| < \epsilon \ll 1$ [19], where $|\boldsymbol{s}| = 1$ and 2ϵ represents the angular divergence of the beam. It is worth mentioning that this is an inherent divergence of partially coherent radiation arising from imperfect transverse correlations in wave fronts and has no direct connection with aperture diffraction effects.

III. DIFFRACTION ON CRYSTALS

If we suppose, as it is usually done, that the scattering by the crystal is essentially due to point atoms we can approximate the coefficients $\gamma_{\boldsymbol{G}}(\omega)$ by unity for all vectors of the reciprocal lattice $\boldsymbol{G}$. If, in addition, the particular diffraction orders corresponding to different vectors $\boldsymbol{G}$ are well separated one can also restrict oneself to studying a single diffraction line only. Thus, instead of the quantity $I(r\boldsymbol{u})$ we will focus our attention on the following one:

$$I_{\boldsymbol{G}}(r\boldsymbol{u}) = \frac{(2\pi)^6}{r^2}\,(\delta_{lm} - u_l u_m)\int_{R^3} d^3f\,|\mathcal{A}(\boldsymbol{f}-\boldsymbol{G})|^2 \times \int_0^\infty d\omega\, k^4\, s(\omega)\,\hat{\mu}_{lm}(\boldsymbol{k}-\boldsymbol{f},\omega). \quad (18)$$

To emphasize the correlation-induced effects we neglect the influence of the finite size of the sample; i.e., we will assume a relatively large crystal and a homogeneous distribution of intensity of the incident beam so that the function $|\mathcal{A}(\boldsymbol{f})|^2$ will be very narrow in comparison with $\hat{\mu}_{lm}(\boldsymbol{f},\omega)$ when considered a function of $\boldsymbol{f}$. Equation (18) can be then written in the approximate form

$$I_{\boldsymbol{G}}(r\boldsymbol{u}) = \alpha\frac{(2\pi)^6}{r^2}\,(\delta_{lm} - u_l u_m) \times \int_0^\infty d\omega\, k^4\, s(\omega)\,\hat{\mu}_{lm}(\boldsymbol{k}-\boldsymbol{G},\omega), \quad (19)$$

where α is a real constant proportional to the volume of the crystal and still $\boldsymbol{k} = k\boldsymbol{u}$.

IV. LAUE METHOD

In the case of the Laue method a fixed monocrystal is illuminated by a collimated x-ray beam with a broad ("white") spectrum. For the sake of simplicity we will assume that the spectrum in the location of the sample is constant [let $s(\omega) = 1$ on the effective frequency range]. It is well known that when the incident radiation is fully coherent the lattice selects the directions of diffraction maxima and the proper frequencies so that $\boldsymbol{k} = \boldsymbol{G} + k\boldsymbol{s}_0$ ($k\boldsymbol{s}_0$ represents the wave vector of the incident wave). In the coherent limit the function $\hat{\mu}_{lm}(\boldsymbol{f},\omega)$ tends to the δ function $\delta(k\boldsymbol{s}_0 - \boldsymbol{k} + \boldsymbol{G})$ and thence $I_{\boldsymbol{G}}(r\boldsymbol{u})$, given by Eq. (19), is nonzero only when $\boldsymbol{k} = \boldsymbol{G} + k\boldsymbol{s}_0$. From Eq. (19) it is clear that for a partially coherent incident beam the situation is somewhat more complicated.

Let us choose such a coordinate system so that the incident beam will propagate in the direction of the z axis and the vector $\boldsymbol{G}$ under consideration is located in the x-z plane. Further, let us confine ourselves to directions of observation $\boldsymbol{u}$ that are also in the x-z plane. Thus $\boldsymbol{s}_0 \equiv (0,0,1)$, $\boldsymbol{G} \equiv (G\sin\beta, 0, G\cos\beta)$, and $\boldsymbol{u} \equiv (\sin\theta, 0, \cos\theta)$, where β and θ are the angles that $\boldsymbol{G}$ and $\boldsymbol{u}$, respectively, make with the z axis (with $\boldsymbol{s}_0$).

To show that the shift of the diffraction maxima caused by coherence properties of the incident radiation occurs even in very common situations, we will assume that the radiation is generated by a circular chaotic nonpolarized planar source (e.g., the radiant of a tungsten x-ray tube) of radius ρ_0 placed perpendicularly to the z axis at the distance z_0 from the sample. We will assume that the tensor of degree of spectral coherence of the radiation has, even in the region of the scattering crystal, only two (same) nonzero components $\mu_{xx} = \mu_{yy} = \mu$ [20]. To determine $\mu(\boldsymbol{r}_2 - \boldsymbol{r}_1, \omega)$ we will employ the scalar, paraxial, and far zone approximations. As it is shown in Appendix A, for the spatial Fourier transform of this degree of spectral coherence we then obtain the following expression,

$$\hat{\mu}(\boldsymbol{f}, \omega) = \frac{1}{2\pi} \frac{1}{\epsilon^2 k^2} \delta(k - f_z) \operatorname{circ}\left(\frac{\boldsymbol{f}_\perp}{\epsilon k}\right), \tag{20}$$

where $\boldsymbol{f}_\perp$ is the projection of $\boldsymbol{f}$ perpendicular to the z axis and $\epsilon = \rho_0 / z_0$ [21]; the function circ is defined in Appendix A.

After substituting from Eq. (20) into Eq. (19) one obtains for $I_{\boldsymbol{G}}(r\boldsymbol{u})$ the expression

$$I_{\boldsymbol{G}}(r\boldsymbol{u}) = \mathcal{I}(\theta) = \alpha c \frac{(2\pi)^5}{r^2 \epsilon^2} (1 + \cos^2\theta) \int_0^\infty dk\, k^2 \delta(k - k\cos\theta + G\cos\beta) \operatorname{circ}\left[\frac{1}{\epsilon}\left(\sin\theta - \frac{G\sin\beta}{k}, 0\right)\right]. \tag{21}$$

From the positiveness of the frequency it follows that the function $\mathcal{I}(\theta)$ is zero unless β is in the range $(\pi/2, 3\pi/2)$. Further we will consider the angle θ to be greater than ϵ and less than $2\pi - \epsilon$ (then we need not consider pathological cases corresponding to $G/k = 0$ and limiting cases with $\beta = \pm\pi/2$). One can than show (see Appendix B), that

$$\mathcal{I}(\theta) = \alpha c G^2 \frac{(2\pi)^5}{r^2 \epsilon^2} \cos^2\beta \, \frac{1 + \cos^2\theta}{(1 - \cos\theta)^3} \tag{22}$$

for $\theta \in (-\pi + 2\beta - \epsilon, -\pi + 2\beta + \epsilon) \cap (\epsilon, 2\pi - \epsilon)$ and zero otherwise in the interval $(\epsilon, 2\pi - \epsilon)$ (2π periodicity is not displayed here for brevity); see Fig. 1. The function $(1 + \cos^2\theta)/(1 - \cos\theta)^3$ is increasing in the interval $(0, \pi)$ and it is decreasing in the interval $(\pi, 2\pi)$. Thus if the "coherent position" of the diffraction line $\theta_{\text{coh}} = 2\beta - \pi$ lays between 0 and π (0° and 180°) the maximum of $\mathcal{I}(\theta)$ is shifted, with respect to θ_{coh}, toward smaller angles and if $\theta_{\text{coh}} \in (\pi, 2\pi)$ it is shifted toward larger angles [22]. Similarly, it can be shown that the angular shift appears also in the centroid

$$C = \frac{\int \theta \mathcal{I}(\theta)\, d\theta}{\int \mathcal{I}(\theta)\, d\theta}. \tag{23}$$

(The shift of the centroid decreases with increasing θ_{coh} [on the interval $(\epsilon, \pi - \epsilon)$].)

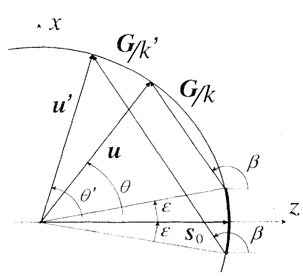

FIG. 1. The notation and the layout of vectors $\boldsymbol{u}$, $\boldsymbol{s}_0$, and $\boldsymbol{G}$ is shown. The function $\hat{\mu}_{lm}(\boldsymbol{k} - \boldsymbol{G}, \omega)$, given by Eq. (20), is nonzero when $(\boldsymbol{u} - \boldsymbol{G}/k)$ lays on the bold arc of the circle.

As an example let us consider a situation when the parameter $\epsilon = 0.01$. This value determines the shift of diffraction maxima in radians. It is approximately 0.5°. The width of the line (2ϵ) is about 1° [23]. For the vector $\boldsymbol{G}$ for which $\theta_{\text{coh}} = 30°$ the centroid of the diffraction profile is $C = 29.977°$. However, the presence of apertures between the source and the crystal improves the coherence of the radiation and the shifts may decrease.

Hence even in the case of the usual thermal source, the profile of the diffraction line differs from those for the fully coherent illumination. Not only the broadening of the lines appears but also the angular shifts both of the diffraction maxima and of the centroids. With a more complicated form of the function $\psi_{lm}(\boldsymbol{f}/|\boldsymbol{f}|, \omega)$, which realizes the coherence properties of the radiation in the place of the sample, the diffraction profile may be, of course, modified in more complex ways. Especially the explicit frequency dependence can play an important role. Nevertheless, the shift of the maxima and of the centroids never exceeds the value of the half "coherence" divergence of the beam (i.e., ϵ). For practically applicable estimates of shifts it probably will be necessary to conceive a more realistic description of correlations in the incident radiation, taking into account the influence of possible monochromators, apertures etc., to consider the effect of the finite size of the crystal [thus to evaluate the convolution in Eq. (18)], and to know the actual spectrum of the incident radiation.

We can see that the physical cause of the shifts lies in the inherent divergence of partially coherent beams [which is, in fact, a consequence of a finite size of the (transverse) coherence area] and on the inhomogeneous modification of the intensity of particular frequency components during propagation (Wolf effect) [1,2]. The first effect is connected with dependence of $\hat{\mu}$ on $\boldsymbol{f}/|\boldsymbol{f}|$ and the second one with its total dependence on ω [together with the factor k^4 in Eq. (19)].

V. POWDER METHOD

In the case of the powder method the sample is in the form of a powder or a polycrystalline material. The incident radiation is quasimonochromatic. To find the intensity distribution of scattered radiation we have to include the contributions from crystals of all present orientations [each contribution is given by Eq. (18), in general]. We can suppose that the distribution of the orientations of component crystals is almost continuous. Let us introduce the following parametrization of the unit sphere,

$$x = \sin\beta\cos\phi, \quad y = \sin\beta\sin\phi, \quad z = \cos\beta, \quad \beta \in (0,\pi), \quad \phi \in (0, 2\pi). \tag{24}$$

Starting again from Eq. (19) and using the same assumptions regarding polarization of the incident radiation and regarding the source as in the previous section [Eq. (20)], one has

$$I_G(r\boldsymbol{u}) = \mathcal{I}(\theta) = \alpha\frac{(2\pi)^5}{r^2\epsilon^2}(1+\cos^2\theta)\int_0^\infty d\omega\, k^2 s(\omega)\int_0^{2\pi} d\phi \int_0^\pi d\beta\,\sin\beta \times\delta(k - k\cos\theta + G\cos\beta)\,\mathrm{circ}\left[\frac{1}{\epsilon}\left(\sin\theta - \frac{G\sin\beta\cos\phi}{k}, \frac{G\sin\beta\sin\phi}{k}\right)\right]. \tag{25}$$

Here $\boldsymbol{u} \equiv (\sin\theta, 0, \cos\theta)$, $\boldsymbol{s}_0 \equiv (0,0,1)$; $k = \omega/c$ and G is the magnitude of the chosen vector of the reciprocal lattice. Integration over ϕ and β covers all orientations of component crystals.

A kind of "degeneration" appears here: we cannot distinguish contributions from vectors of the same magnitude G and hence, to obtain an actual value of intensity, we have to multiply the function $I_G(r\boldsymbol{u})$ by the number of vectors of the reciprocal lattice having magnitude equal to G. Since the space lattice is real, at least the vector $-\boldsymbol{G}$ must occur together with $\boldsymbol{G}$.

Due to the δ function we can eliminate the integral over the variable β in Eq. (25). Then

$$G\cos\beta = k(\cos\theta - 1). \tag{26}$$

Since $\cos\beta > -1$ it is clear that k has to be less than $G/(1-\cos\theta)$, otherwise the result of integration over β is zero. Let us confine ourselves, for the sake of simplicity, to the angles θ of the range $(\epsilon, \pi-\epsilon)\cup(\pi+\epsilon, 2\pi-\epsilon)$. The remaining integral over the variable ϕ represents, in fact, the length l of the bold arc in Fig. 2 given by intersecting circles. If the circles do not intersect the integral is zero. Consequently,

$$\mathcal{I}(\theta) = \frac{\alpha}{G}\frac{(2\pi)^5}{r^2\epsilon^2}(1+\cos^2\theta) \times\int_0^{Gc/(1-\cos\theta)} d\omega\, k^2 s(\omega) l_G(\theta,\omega), \tag{27}$$

where

$$l_G(\theta,\omega) = \mathrm{Re}\left\{\frac{2G}{k}\sqrt{1-\frac{k^2}{G^2}(\cos\theta-1)^2}\arccos\left[\frac{k}{2G}\frac{\sin^2\theta + (G/k)^2 - (\cos\theta-1)^2 - \epsilon^2}{|\sin\theta|\sqrt{1-(k/G)^2(\cos\theta-1)^2}}\right]\right\}; \tag{28}$$

we have also used Eq. (26). The value of this function is equal to the length of the arc emphasized in Fig. 2, when the circles intersect, and is zero otherwise.

From Eqs. (27) and (28) it can be seen that even in the case of monochromatic incident illumination [$s(\omega) = \delta(\omega - \omega_0)$] the diffraction line is broadened— the width is 2ϵ (see Fig. 3) — and the diffraction profile is described by a relatively complicated function. The reason for this is the coherence divergence of the input beam and the fact that in different directions different numbers of the orientations of component crystals contribute to the intensity of scattered radiation. If the spectrum of the incident x-ray beam has a small but finite width, the diffraction line is further broadened due to this (this contribution is responsible for increasing the breadth of the line with increasing G) and its profile is influenced by the form of the spectrum, modified due to the Wolf effect, in general.

An example: Let us consider the same configuration

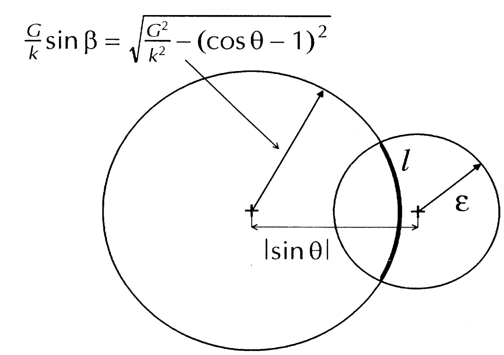

FIG. 2. Geometrical meaning of the integral over ϕ in Eq. (25). When the circles intersect the integral is equal to the length of the bold arc.

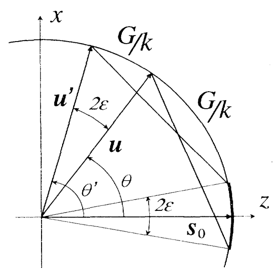

FIG. 3. Illustration of broadening of the diffraction line in the case of monochromatic illumination.

as in Sec. IV, i.e., let the parameter $\epsilon = 0.01$ again. Let us suppose, further, that the spectrum of the radiation at the position of the crystal is Gaussian,

$$s(\omega) \propto \exp\left[-\frac{(\omega-\omega_0)^2}{\sigma^2}\right], \tag{29}$$

with the width $\sigma = 0.001\,\omega_0$. In Fig. 4 there are results of the numerical analysis of Eq. (27) together with Eq. (28) under the conditions mentioned above. It shows the shifts of maxima of diffraction profiles and of their centroids [see Eq. (23)] with respect to Bragg angles (corresponding to the ideal diffraction of the monochromatic plane wave of frequency ω_0) for different magnitudes G of vectors of the reciprocal lattice. The values of G are from the interval between $0.1\,k_0$ and $1.9\,k_0$ ($k_0 = \omega_0/c$) and they are represented on the horizontal axis by the corresponding Bragg angles for frequency ω_0, $\theta_B = 2\arcsin[G/(2k_0)]$. All the quantities are given in degrees. The accuracy of the calculated values is better than 0.001°. From Fig. 4 we can see that the shifts of diffraction maxima may appear also in the powder method under commonly occurring circumstances. Nevertheless, in our example, the shifts are very small in comparison with the breadth of the profile, which is about 1° [24].

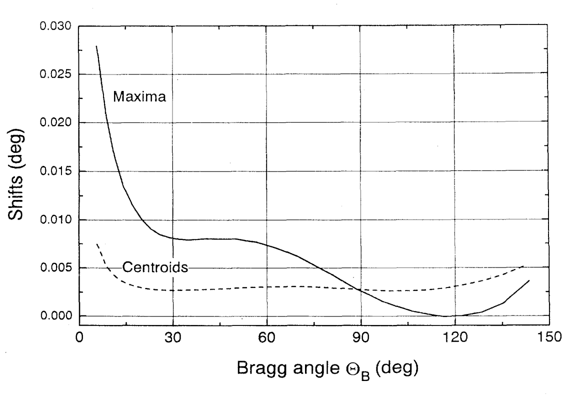

FIG. 4. The shift of the maximum of the diffraction profile and the shift of its centroid with respect to the corresponding Bragg angle vs the Bragg angle. For details, see the text.

VI. CONCLUSIONS

We have developed a theory describing the scattering of electromagnetic radiation of any state of coherence and of arbitrary state of polarization on three-dimensional periodic structures, valid within the accuracy of the first-order Born approximation, i.e., for kinematic diffraction, in terms of x-ray structure analysis. The incident field may have an arbitrary spectrum. It is only assumed to be statistically stationary and quasihomogeneous (at least in the domain of the scattering medium).

We have shown that the diffraction profiles (i.e., the angular dependencies of the intensity of scattered radiation) are influenced, in general, by the coherence properties of incident radiation. It is caused partly by the Wolf effect, i.e., due to the modification of the spectrum of radiation during propagation as a consequence of the state of spatial coherence of the field, and partly by an inherent angular divergence of partially coherent beam-like fields, which is connected with the finite size of the (transverse) coherence area. The effect can have interesting consequences for x-ray structure analysis. We have demonstrated, by examples, that the coherence-induced angular shifts of diffraction maxima may appear even in common cases both in the Laue method and in the powder method.

A similar effect can probably appear in the particle diffraction (e.g., in the scattering of electrons or neutrons on crystals). The phenomenon described could perhaps occur also when a thin hologram is illuminated by a partially coherent light.

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APPENDIX A: CHAOTIC PLANAR SOURCE

Let us consider a planar circular spatially incoherent statistically stationary scalar secondary source with the radius ρ_0 and the center at the origin of coordinate system; its cross-spectral density

$$W(\boldsymbol{\rho}_1,\boldsymbol{\rho}_2,\omega) \to \delta(\boldsymbol{\rho}_1-\boldsymbol{\rho}_2)S(\omega)\,\mathrm{circ}\left(\frac{\boldsymbol{\rho}_1}{\rho_0}\right), \tag{A1}$$

where $\boldsymbol{\rho}_1, \boldsymbol{\rho}_2$ are position vectors laying in the source plane (2D vectors), which is perpendicular to the z axis, $S(\omega)$ is the spectrum (we suppose it to be the same throughout the whole source), and

$$\mathrm{circ}(\boldsymbol{\xi}) = \begin{cases} 1 & \text{for } |\boldsymbol{\xi}| < 1, \\ 0 & \text{otherwise.} \end{cases} \tag{A2}$$

The cross-spectral density in the far zone (i.e., when the distance from the source is much greater than the dimensions of the source) of the radiation generated by a planar secondary source is given by the formula (see, e.g., [25])

$$W^{(\infty)}(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega)=\left(\frac{k}{2\pi}\right)^2\cos\theta_1\cos\theta_2\,\frac{\exp[-ik(r_1-r_2)]}{r_1r_2}\int_{R^2}d^2\rho_1\int_{R^2}d^2\rho_2 \times W(\boldsymbol{\rho}_1,\boldsymbol{\rho}_2,\omega)\exp[ik(\boldsymbol{s}_{\perp 1}\cdot\boldsymbol{\rho}_1-\boldsymbol{s}_{\perp 2}\cdot\boldsymbol{\rho}_2)], \quad \text{(A3)}$$

where $\boldsymbol{r}_i = r_i\boldsymbol{s}_i$, $|\boldsymbol{s}_i| = 1$, $\cos\theta_i = s_{zi}$, and $\boldsymbol{s}_{\perp i}$ is the projection of vector $\boldsymbol{s}_i$ into the source plane, $i = 1, 2$.

We employ the paraxial approximation and assume $|r_1 - r_2| \ll r_i$ $(i = 1, 2)$. Near a point $(0, 0, z_0)$ in the far field (z_0 being the distance from the source obeying the constraint $|z_0 - z_i| \ll z_0$, z_i being the z components of $\boldsymbol{r}_i$), we obtain for the degree of spectral coherence the expression

$$\mu^{(\infty)}(\boldsymbol{r}_2-\boldsymbol{r}_1,\omega)=\exp[ik(z_2-z_1)]\frac{J(\epsilon k|\boldsymbol{r}_{\perp 2}-\boldsymbol{r}_{\perp 1}|)}{\epsilon k|\boldsymbol{r}_{\perp 2}-\boldsymbol{r}_{\perp 1}|}, \quad \text{(A4)}$$

where $\boldsymbol{r}_{\perp i} = r\boldsymbol{s}_{\perp i}$, $J(\,)$ is the Bessel function of the first kind and the first order, and $\epsilon = \rho_0/z_0$. Since we are interested in the cross-spectral density (tensor) only in the region of the sample crystal [elsewhere the function $T(\boldsymbol{r})$ is zero] we may regard, for the sake of simplicity (but without any loss of generality) the spectral degree of coherence as having the form given by Eq. (A4) in the whole space (with ϵ being constant). We can then determine its Fourier transform with respect to $(\boldsymbol{r}_2-\boldsymbol{r}_1)$:

$$\hat{\mu}(\boldsymbol{f},\omega)=\frac{1}{2\pi}\frac{1}{\epsilon^2k^2}\delta(k-f_z)\,\mathrm{circ}\left(\frac{\boldsymbol{f}_\perp}{\epsilon k}\right). \quad \text{(A5)}$$

Here f_z is the z component of $\boldsymbol{f}$ and $\boldsymbol{f}_\perp$ is its projection on the source plane. This expression is only approximative and it does not satisfy condition (17). Nevertheless, since we assume that $\epsilon \ll 1$ (i.e., $\rho_0 \ll z_0$) the function $\hat{\mu}(\boldsymbol{f},\omega)$ is nonzero only for $|\boldsymbol{f}_\perp| \ll k$ and Eq. (A5) is nearly identical with

$$\hat{\mu}(\boldsymbol{f},\omega)=\frac{1}{2\pi}\frac{1}{\epsilon^2k^2}\,\delta(k-|\boldsymbol{f}|)\,\mathrm{rect}\left(\frac{1}{\epsilon}\left|\boldsymbol{s}_0-\frac{\boldsymbol{f}}{|\boldsymbol{f}|}\right|\right). \quad \text{(A6)}$$

Evidently this expression has the same form as Eq. (17). In this equation $\mathrm{rect}(\xi) = 1$ for $\xi \in (0, 1)$ and is zero otherwise, $\boldsymbol{s}_0$ is the unit vector in the direction of the z axis.

APPENDIX B: NOTES TO DERIVATION OF FORMULA (22)

Here we will inquire where the function $\mathcal{I}(\theta)$ appearing in Eq. (21) has nonzero values. The δ function in Eq. (21) selects only k fulfilling the equation

$$k(\cos\theta-1)=G\cos\beta. \quad \text{(B1)}$$

Because $k > 0$ the value of $\cos\beta$ must be negative and thus $\beta \in (\pi/2, 3\pi/2)$. The function "circ" is nonzero when

$$-1<\frac{1}{\epsilon}\left(\sin\theta-\frac{G\sin\beta}{k}\right)<1. \quad \text{(B2)}$$

Taking into account that $\epsilon \ll 1$ one can write $\epsilon \approx \sin\epsilon$ and $1 \approx \cos\epsilon$. Realizing this and using Eq. (B1) together with the negativeness of $\cos\beta$ one obtain the inequality

$$-\sin\epsilon\cos\beta>\sin\theta\cos\beta-\sin\beta\cos\theta +\sin\beta\cos\epsilon>\sin\epsilon\cos\beta, \quad \text{(B3)}$$

or, after some algebra,

$$\sin\left(\frac{\theta+\epsilon}{2}\right)\cos\left(\frac{\theta-2\beta-\epsilon}{2}\right)<0 <\sin\left(\frac{\theta-\epsilon}{2}\right)\cos\left(\frac{\theta-2\beta+\epsilon}{2}\right). \quad \text{(B4)}$$

Let us confine ourselves to angles of observation in the range $\theta \in (\epsilon, 2\pi - \epsilon)$. We then need not deal with elimination of nonphysical cases and with examinating the cases when β is close to $\pm\pi/2$, which need more careful analysis when θ is near zero. Under this constraint the sines in Eq. (B4) are positive and it is then straightforward to show that the inequality (B4) is satisfied when $\theta \in (-\pi + 2\beta - \epsilon, -\pi + 2\beta + \epsilon)$.

It should be noted that we do not distinguish between θ and $\theta + n\pi$, where n is an integer.

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[14] The scattering medium is supposed to be weak in the sense that for all values of its arguments the polarization induced by the scattered field is much smaller than polarization induced by the incident field and similar constrains hold for the first derivatives with respect to both the temporal and spatial arguments.
[15] This formula can be easily generalized for the case when the macroscopic properties of the medium vary slowly in the course of time, using a dielectric susceptibility function that depends on two time arguments $\eta(\boldsymbol{r}, t; t')$ [12] and also for the case of a spatially dispersive medium taking a susceptibility function with other spatial argument and adding the spatial convolution into Eq. (3) (see, e.g., [16]). Generalization for anisotropic media is also straightforward.
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[19] A transverse plane (perpendicular to $\boldsymbol{s}_0$) can be regarded as a secondary plane source and similar arguments apply to it as in Ref. [2].
[20] In general, a nonzero z component of the electric field vector appears but it can be neglected in the described configuration.
[21] In further calculations we will use this simple model function. However, note that the above-mentioned paraxial approximation may not be always well applicable in x-ray optics.
[22] The function $1/(1 - \cos\theta)^3$, which we would obtain for radiation (fully) polarized at the y direction, behaves in a similar way while the function $\cos^2\theta/(1-\cos\theta)^3$ corresponding to x polarization has minima at $\pm\pi/2$ so that the sign of shift of diffraction maxima changes in each quadrant.
[23] Let us recall again that we are neglecting the broadening of the line due to a finite size of the sample [more exactly from a finite width of the function $A(\boldsymbol{r})$]. If the linear dimensions of the sample in our example are, say, of order of the tenths of millimeters or greater this approximation is well applicable for x rays.
[24] Choosing more "exotic" correlations in the incident radiation the shifts could be probably increased. But the question is how to prepare such correlations in x-ray beams experimentally.
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