COHERENCE OF THE MOSAIC BLOCKS IN X-RAY DIFFRACTOGRAPHY

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It is shown that the coherence properties of mosaic blocks can be studied both qualitatively and quantitatively. The idea behind the calculation is as follows: the mosaic blocks are considered to be a secondary source of X-rays and their coherence properties are then investigated by the usual methods used in the theory of coherence properties of light. The considerations given in this paper allow a more detailed examination as to the complexity of the origin of the diffraction by real crystals and also theoretically give reasons for the hitherto intuitively presented assumptions that mosaic blocks or powder crystal particles are mutually incoherent. From these calculations it also follows that the coherence of mosaic blocks depends not only on the crystallographic point of view, but also on the coherence properties of the X-rays.

1. INTRODUCTION

In paper [1] the influence of the coherence properties of X-rays on diffraction by a small ideal crystal is studied. In this paper we shall consider X-ray diffraction by real crystals, i.e. those where the primary ideal lattice is decomposed, due to crystal defects of various kinds, into smaller parts of the ideal crystal lattices which are slightly misoriented and shifted with respect to the primary lattice. This simple model of the real crystal lattice is in accordance with existing presuppositions concerning mosaic crystals.

The basic problem when calculating the intensity of X-ray diffractions by a mosaic crystal is the question of the coherence of mosaic blocks. This was discussed for the first time in 1914 by Darwin [2] whose calculations of the intensity of diffraction were carried out under the assumption that all mosaic blocks are incoherent. The same presumption was expressed for powder crystal particles. The experimental results were in good agreement with calculations and therefore further attention was not paid to this problem. In X-ray literature there appears the term "the coherent domains of the crystal lattice" used for those parts of a crystal without defects.

In connection with the X-ray studies of cold-worked metals, methods for the measurement of the mean value of the particle size (coherent domains) [3, 4] have been elaborated. The question then arose as to how to interpret the measured mean value of the coherent domains crystallographically and particularly from the viewpoint of the theory of dislocations. Wilkens [5] dealt with this problem in detail and ascertained that the mean value of the particle size cannot be identified with either the distance of small-angle or large-angle grain boundaries. He says that it is a quantity of no unambiguous crystallographic significance in plastically deformed crystals. Nevertheless, in calculations based on the kinematic theory the mean value

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of the particle size is considered as that of the ideal crystal over which the summation of the secondary waves is carried out.

The question as to the value of the particle size of a couple of mosaic blocks has also been studied by Khachaturjan [6]. He has shown that the value of the particle size L appearing in the relation $B = \lambda / L \cos \vartheta$ depends on the angle of the mutual misorientation of the lattice of the mosaic blocks: the smaller the angle of misorientation, the larger the coherent domain. Also in a paper by Warren [3] we find a note that two mosaic blocks can be coherent but neigher Khachaturjan nor Warren clarify this question more closely.

It will be shown here that the coherence of mosaic blocks may be clarified not only qualitatively but also quantitatively from the aspect of the optical principles of diffractions. From calculations of the intensity of diffraction by the couple of mosaic blocks it follows that the coherence of mosaic blocks cannot be considered from the crystallographic aspect only, but it is also necessary to take into consideration the coherence properties of X-rays.

2. CALCULATION OF THE INTENSITY OF DIFFRACTION BY A COUPLE OF MOSAIC BLOCKS

The idea behind the calculation of the intensity of diffraction by a couple of mosaic blocks is as follows: each of the blocks is considered as a separate source of secondary waves and their coherence is studied by the help of the classical theory of coherent light.

Let us consider two mosaic blocks of which the mutual position, the position of the X-ray source σ and that of the point of observation Q is evident from Fig. 1. By respecting the coherence properties of X-rays an expression for the intensity of diffraction by an ideal crystal was derived in paper [1], which may be written in the simplified form:

$$I(Q) = I_0 \sum_{P,P'} \Gamma(P, P') \cos \mathbf{K}(\mathbf{r}_P - \mathbf{r}_{P'}) \tag{1}$$

I_0 – is a factor which for a given diffraction changes slowly with the angle ϑ.

$\Gamma(P, P')$ – is a coherence function, the value of which depends on the mutual position of the points P and P', on the frequency composition of the X-rays, on the size of the source and its distance from the crystal. This function will be discussed below.

$\mathbf{K}$ – is the difference between the wave vectors of the incident and diffracted radiation.

$\mathbf{r}_P, \mathbf{r}_{P'}$ – are the position vectors of the latitce points P and P'. These lattice points form the diffracting volume of the crystal where summation over all possible couples of the lattice points P, P' is carried out (see Eq. (1)).

The set of lattice points P, however, may be divided into two parts:

1. The points U forming the first mosaic block $\{U\}$.
2. The points V forming the second mosaic block $\{V\}$.

If we denote the phase difference of the secondary waves scattered by the lattice points P and P'

$$\mathbf{K}(\mathbf{r}_P - \mathbf{r}_{P'}) \equiv \alpha(P, P'), \tag{2}$$

then Eq. (1), due to the distribution of the diffracting volume on two mosaic blocks, may be written as follows

$$I(Q)\, I_0^{-1} = \sum_{U,U'} \Gamma(U, U') \cos \alpha(U, U') + \sum_{V,V'} \Gamma(V, V') \cos \alpha(V, V') + \\ + \sum_{U,V} \Gamma(U, V) \cos \alpha(U, V) + \sum_{V,U} \Gamma(V, U) \cos \alpha(V, U). \tag{3}$$

Taking into account

$$\alpha(U, V) = -\alpha(V, U)\,; \quad \Gamma(U, V) = \Gamma(V, U) \tag{4}$$

Eq. (3) may rewritten in another form:

$$I(Q)\, I_0^{-1} = I_U(Q) + I_V(Q) + 2I_{UV}(Q). \tag{5}$$

Let us now pay greater attention to the significance of the different terms of this equation.

The first two terms, $I_U(Q)$ and $I_V(Q)$, are the relative intensities of diffraction in the point Q from the individual mosaic blocks.

The third term

$$I_{UV}(Q) = \sum_{U,V} \Gamma(U, V) \cos \alpha(U, V) \tag{6}$$

has the significance of an interference term of the intensity of diffraction by the couple of mosaic blocks. Applying the classical theory of coherence, the coherence of mosaic blocks may be discussed as follows:

1. The considered mosaic blocks are incoherent (optically independent) if $I_{UV}(Q) = 0$. In this case the resulting intensity of diffraction is the sum of the intensities of the individual mosaic blocks. Here the mosaic blocks appear as optically independent sources of X-rays.

2. If $I_{UV}(Q) \neq 0$ the two mosaic blocks are optically dependent, i.e. wholly or partly coherent. The resulting intensity of diffraction is then not a simple sum of intensities from the separate blocks.

As is evident from the calculations (see Eq. (6)), for the determination of the coherence properties of mosaic blocks we can confine ourselves to the calculation of the interference term of the intensity $I_{UV}(Q)$ [1]) only. In the next part of this article we shall carry out its calculation for some special cases of the situation of the blocks $\{U\}$ and $\{V\}$.

3. CALCULATION OF THE INTERFERENCE TERM OF THE INTENSITY OF DIFFRACTION

For simplicity and with respect to the application of the results of paper [1], let us consider a primitive and orthorhombic lattice which in block $\{U\}$ is determined by the vectors $\boldsymbol{a}_i$ and in block $\{V\}$ by the vectors $\boldsymbol{a}'_i$; (see Fig. 1). Then, let the following transformation hold for these vectors

$$\boldsymbol{a}'_i = \sum_k \alpha_{ik} \boldsymbol{a}_k \quad i, k = 1, 2, 3 \tag{7}$$

where α_{ik} is the cosine of the angle formed by the vectors $\boldsymbol{a}'_i$ and $\boldsymbol{a}_k$. The position of the lattice point U is determined by the lattice vector $\boldsymbol{r}_U = u_1\boldsymbol{a}_1 + u_2\boldsymbol{a}_2 + u_3\boldsymbol{a}_3$. The position of the point V in the dashed reference system is determined by the lattice vector $\boldsymbol{r}_V = v_1\boldsymbol{a}'_1 + v_2\boldsymbol{a}'_2 + v_3\boldsymbol{a}'_3$ whilst in the undashed reference system the point V is determined by the vector

$$\boldsymbol{r}_V = \sum_k [z_k + \sum_i \alpha_{ik} v_i] \, \boldsymbol{a}_k \tag{8}$$

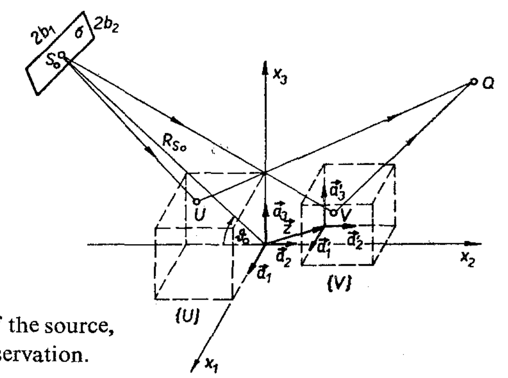

Fig. 1. Scheme of the spatial arrangement of the source, couple of mosaic blocks and point of observation.

where the vector $\boldsymbol{z} = z_1\boldsymbol{a}_1 + z_2\boldsymbol{a}_2 + z_3\boldsymbol{a}_3$ determine the position of the origin of the dashed reference system in the undashed one. The difference of the position

[1]) The fact that we have considered a couple of mosaic blocks only does not restrict the application of this method to an arbitrary number of mosaic blocks. The statement that if block $\{A\}$ is partly coherent with block $\{B\}$ and $\{B\}$ with block $\{C\}$, then block $\{A\}$ is partly coherent with $\{C\}$ does not hold generally. A proof of the invalidity of this statement is the case when all three blocks are not simultaneously incident on a coherent region of 0·1 degree (for the definition of a coherent region of k degree see [1]).

vectors may then be written as follows:

$$\mathbf{r}_U - \mathbf{r}_V = \sum_k [(u_k - v_k) - z_k - \sum_i (\alpha_{ik} - \delta_{ik})\, v_i]\, \mathbf{a}_k \tag{9}$$

where δ_{ik} is Kronecker's symbol. If we denote the difference of the wave vectors in the undashed reference system in terms of $\mathbf{K} = K_1\mathbf{a}_1 + K_2\mathbf{a}_2 + K_3\mathbf{a}_3$, the phase difference may then be expressed in the form

$$\alpha(U, V) = \alpha_0(U, V) - \beta(\mathbf{z}, V) \tag{10}$$

where

$$\alpha_0(U, V) = \sum_k a_k^2 K_k (u_k - v_k) \tag{11}$$

has the significance of the phase difference but only in the case when the two reference frames have mutually paralell axes and the vector $\mathbf{z}$ has such a value that the blocks $\{U\}$ and $\{V\}$ form one block again and

$$\beta(\mathbf{z}, V) = \sum_k K_k a_k^2 [z_k + \sum_i (\alpha_{ik} - \delta_{ik})\, v_i] \tag{12}$$

has the significance of a change of the difference due to the shift and misorientation of the dashed reference system of block $\{V\}$. The interference term of the intensity of diffraction is then given by the expression

$$\begin{aligned} I_{UV}(Q) = &\sum_{U,V} \Gamma(U, V) \cos \beta(\mathbf{z}, V) \cos \alpha_0(U, V) + \\ &+ \sum_{U,V} \Gamma(U, V) \sin \beta(\mathbf{z}, V) \sin \alpha_0(U, V)\,. \end{aligned} \tag{13}$$

If we confine ourselves to the immediate surroundings of the maximum diffraction only, then the angle $\alpha_0(U, V) = n(U, V)\, 2\pi + \varepsilon(U, V)$, where $n(U, V)$ is an integer and $\varepsilon(U, V) \ll 1$. Therefore, the second term on the right side of Eq. (13) may be neglected in comparison with the first one.

The factor $\cos \beta(\mathbf{z}, V)$ has the physical meaning of the coherence function respecting the crystallographic properties of blocks $\{U\}$ and $\{V\}$. We therefore denote analogically

$$\Gamma_4(\mathbf{z}, V) \equiv \cos \beta(\mathbf{z}, V)\,. \tag{14}$$

The value of this coherence function depends then on the mutual position of the two mosaic blocks and on the mutual misorientation of their crystal lattices (see Eq. (12)). In the next part of this paper we shall show the coherence functions $\Gamma(U, V)$ and $\Gamma_4(\mathbf{z}, V)$ for some special cases and then we shall analyze the coherence properties of mosaic blocks.

4. THE COURSE OF COHERENCE FUNCTIONS

On the assumption that the radiation is quasimonochromatic and the spectral line can be approximated by the Gaussian curve, and also that the plane source of X-rays is in coherent with a constant distribution of intensity of X-rays, the following expression has been derived for the coherence function $\Gamma(U, V)$ in paper [1]

$$\Gamma(U,V) = \Gamma_1(\Delta V, N_3) \,.\, \Gamma_2(b_1, N_1) \,.\, \Gamma_3(b_2, N_2, N_3) \tag{15}$$

where the individual factors are given by the functions

$$\Gamma_1(\Delta\nu, N_3) = \exp\left[-\{(2\pi\, \Delta\nu a_3 N_3 \sin \vartheta_0)/1{\cdot}66c\}^2\right] \tag{16}$$

$$\Gamma_2(b_1, N_1) = \frac{\sin(Kb_1 a_1 N_1/R_{S_0})}{Kb_1 a_1 N_1/R_{S_0}} \tag{17}$$

$$\Gamma_3(b_2, N_2, N_3) = \frac{\sin\left[Kb_2(a_2 N_2 \sin \vartheta_0 + a_3 N_3 \cos \vartheta_0)/R_{S_0}\right]}{Kb_2(a_2 N_2 \sin \vartheta_0 + a_3 N_3 \cos \vartheta_0)/R_{S_0}} \,. \tag{18}$$

The symbols here have the following significance:

K – is the absolute value of the wave vector

$\Delta\nu$ – is the width of the spectral line

c – is the phase velocity of the X-rays

a_1, a_2, a_3 – is the magnitude of vectors of the orthorhombic lattice

$N_1 = v_1 - u_1$, $N_2 = u_2 - v_2$, $N_3 = u_3 - v_3$ are the differences of the coordinates of the points $U(u_1, u_2, u_3)$ and $V(v_1, v_2, v_3)$

$2b_1$ – is the size of the rectangular source in the direction of the x_1 axis (see Fig. 1)

$2b_2$ – is the size of the rectangular source in the direction perpendicular to the primary beam

R_{S_0} – is the distance of the centre of the source S_0 from the origin of the coordinate system

ϑ_0 – is Bragg's diffraction angle (it lies in the x_2x_3 plane).

It is clear that the coherence function depends on all three spatial coordinates. For our considerations concerning the coherence of mosaic blocks it is important to have an idea about the graph of this function in the plane of the angle ϑ. Figs. 2a,b c and d show the loci of points where the product of the function $\Gamma_1(\Delta\nu, N_3) \,.\, \Gamma_3(b_2, N_2, N_3)$ has the same values which are plotted in the figures in multiples of a hundred. These plots are calculated for the following experimental values: $R_{S_0} = 10$ cm, $a_2 = a_3 = \lambda = 1{\cdot}54$ Å, $\vartheta_0 = 30°$ and for the width of the copper spectral lines $K\alpha_1$. The plots differ from one another by the size of the source b_2, which is given in the eight upper part of the figures. The direction of the primary beam is indicated in the left lower part.

The physical importance of these plots consists in determining the value of the coherence function in the point V with regard to the point U situated in the origin of the reference frame. As is evident from Figs. 2a,b,c, and d, the greater the width of the source the longer is the coherent region. As the size of the source increases the coherent region enlargers in the x_2 direction only.

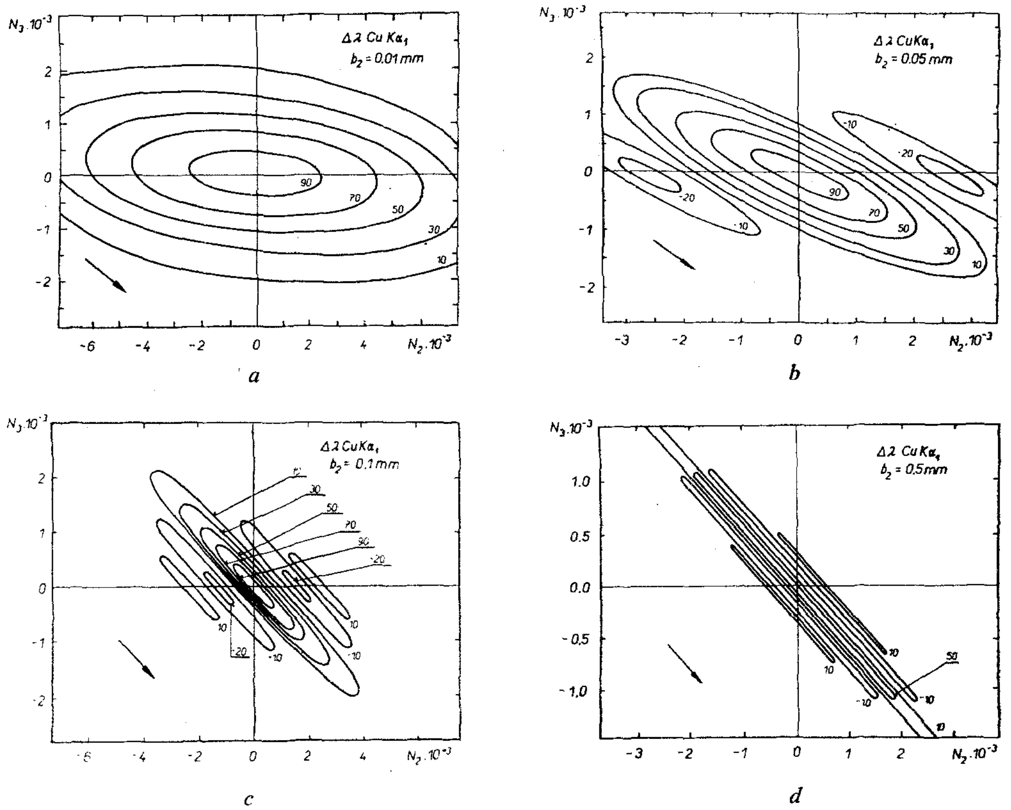

Fig. 2. A map of coherent regions $\Gamma(U, V)$ in the x_2x_3 plane for various sizes of the source.

Example: What is the value of the function $\Gamma(U, V)$ for a source of size $b_2 = 0.5$ mm and for $b_2 = 0.01$ mm, if the lattice points have the coordinates U (300, 500, 1000), V (300, 1500, 2000). For these points $N_2 = 1000$, $N_3 = 1000$. From Fig. 2d it follows that for $b_2 = 0.5$ mm $\Gamma(U, V) = 0$ and from Fig. 2a that for $b_2 = 0.01$ mm $\Gamma(U, V) = 0.5$.

An analysis of the coherence function $\Gamma_4(\mathbf{z}, V)$ will be carried out for the case when block $\{V\}$ is misoriented with respect to block $\{U\}$ by a small angle ε lying in the plane of the angle ϑ_0. If we consider diffraction in the planes $(00l)$, in our geometric arrangement the difference of the wave vectors is given by the relation $\mathbf{K} = 4\pi\, \mathbf{a}_3 \,.\, \sin \vartheta_0/\lambda a_3$ and if we apply Bragg's equation it holds that

$$\beta(\mathbf{z}, V, \varepsilon) \doteq 2\pi\, l[z_3 - \varepsilon v_2]\,. \tag{19}$$

5. INFLUENCE OF THE POSITION OF MOSAIC BLOCKS ON THEIR MUTUAL COHERENCE

Let us assume that $\varepsilon = 0$ and investigate how the coherence of mosaic blocks can influence their mutual situation determined by the vector $\mathbf{z}$. For simplicity, we consider the two blocks to be of the same size. Let us divide the discussion into three parts:

1. Let us assume that both blocks $\{U\}$ and $\{V\}$ are simultaneously in one coherent region of high degree, i.e. that for all the couples of points $\Gamma(U, V) = 1$. The value of the interference term now depends only on the vector $\mathbf{z}$.

a) If z_1, z_2, z_3 are all integers then from Eq. (13) it follows that $I_{UV}(Q) = I_U(Q) = I_V(Q)$, blocks $\{U\}$ and $\{V\}$ are wholly coherent and the total intensity (see Eq. (5)) is $4I_U(Q)$.

b) If z_1, z_2, z_3 are odd multiples of $1/2$ (this case can occur e.g. when blocks $\{U\}$ and $\{V\}$ are separated by a stacking fault) then the interference term $I_{UV}(Q) = -I_U(Q) = -I_V(Q)$ and the total intensity equals zero. The mosaic blocks are wholly coherent.

c) If z_1, z_2, z_3 take other values, the interference term can then take arbitrary values from the interval $\langle -I_U(Q), I_U(Q)\rangle$. In this case the blocks are partly coherent or incoherent for $I_{UV}(Q) = 0$.

2. Let us assume that both mosaic blocks are not simultaneously in one coherent region, i.e. that the function $|\Gamma(U, V)|$ for arbitrary couples of points V and U takes values between zero and one. In this case the value of the interference term is always in the interval $\langle -I_U(Q), I_U(Q)\rangle$ and the mosaic blocks are always partly coherent no matter which values are taken by the vector $\mathbf{z}$.

3. If block $\{V\}$ is outside the coherent region related to an arbitrary lattice point of block $\{U\}$, $(\Gamma(U, V) \doteq 0)$, then both blocks are incoherent whatever the magnitude of the vector $\mathbf{z}$.

6. THE INFLUENCE OF THE MISORIENTATION OF MOSAIC BLOCKS ON THEIR COHERENCE

Let us now consider two mosaic blocks which are shifted by the lattice vector $\mathbf{z}$ but their crystal lattices are mutually misoriented by an angle ε lying in the plane of the angle ϑ_0. We again divide the discussion into three parts.

1. For an arbitrary couple of points U and V let $\Gamma(U, V) = 1$. The value of the interference term then depends only on the angle ε. If $\Gamma_4(\mathbf{z}, \varepsilon) > 0$ for the whole block $\{V\}$ the interference term takes a value lower than $I_U(Q)$. The greater the angle ε, the nearer the absolute value of the interference term will approach zero. Otherwise, the greater the anlge ε, the less the degree of coherence of mosaic blocks.

2. If the function $|\Gamma(U, V)|$ takes values less than one, for arbitrary couples of points U and V, then the mutual misorientation of the crystal lattices causes additional decreasing of the degree of coherence of the mosaic blocks.

3. In the function $|\Gamma(U, V)|$ takes values equal to zero or slightly different from zero for arbitrary couples of points U and V, then such mosaic blocks are incoherent regardless of the angle ε.

7. COMPARISON WITH PREVIOUS CONSIDERATIONS ABOUT THE COHERENCE OF MOSAIC BLOCKS

An analysis of the interference term of the intensity of diffraction by couple of mosaic blocks permits a more detailed investigation of the origin of the diffraction profile on real crystalline samples and also gives reasons for the hitherto qualitative assumptions about the coherence of mosaic blocks and the crystal particles of a powder sample. Let us discuss these cases separately.

From the statistical distribution of the orientation of neighbouring crystal particles of powder samples it follows that the angle of mutual misorientation is large and the vector of the mutual shift is arbitrary. Therefore the average value of the interference intensity of all possible couples of powder particles equals zero and the sample behaves as though the separate powder particles were mutually incoherent.

Since in real crystals all mosaic blocks are separated from each other by a configuration of dislocations, it may be expected that the angles of mutual misorientation and the vectors of the shift need not obtain very different values. For this reason the mosaic blocks may appear as partly coherent.

It follows from our considerations that the degree of coherence can be changed experimentally. Here it is shown that the presumption of the incoherence of mosaic blocks and the crystal particles of powder will be better fulfilled, the worse is are the coherence properties of the primary beam of X-rays. We decrease the degree of coherence of X-rays when using a large source and a wide spectral line. At diffraction by a real monocrystal the average value of the interference term of the intensity of diffraction need not be equal to zero when using small sources of X-rays.

The problems of the crystallographic interpretation of the average value of the coherent regions measured by X-ray methods require further detailed analysis.

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