

DOUBLE REFLECTIONS OF X RAYS IN CRYSTALS

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1. INTRODUCTION

DOUBLE reflection of x-rays is the term referring^[1-3] to one of the diffraction phenomena in which a ray reflected in accordance with the Bragg representation from some set of parallel planes in a crystalline specimen, is reflected again in the same specimen from the planes of the same or another set. The direction of propagation and the intensity of the doubly reflected rays, as in the case of ordinary single reflections, are determined by the experimental conditions and the structure of the investigated object. Consequently, double reflections can be used in principle for structural research, and at any rate the possibility of their occurrence must be taken into account when other diffraction effects are investigated. Since diffraction takes place in crystals not only for x rays but also for particles, above all electrons and neutrons, it is obvious that double reflections can occur also when particle beams traverse crystals.

The possibility of the occurrence of double reflections in crystals was pointed out by Bethe^[4] as far back as in 1928 in an analysis of electron diffraction. They were likewise observed experimentally for the first time in the case of electrons^[5,6]. The probable reason for this is that for electron diffraction the probability of occurrence of double reflections is very large, and the relative intensity of the diffraction is much higher than for x-rays. Subsequently, these reflections were observed in many investigations for x rays and in recent years also for neutrons.

In the first studies, the double reflections were investigated in single crystals. The fact that such reflections should also occur in polycrystalline specimens was pointed out by Renninger^[7,8] in 1937. In the case of polycrystals, however, the effects connected with this phenomenon could not be observed for a long time in as distinctive a fashion as for single crystals. Yet it is probably double reflection which accounts for the diffuse scattering of x rays by deformed metals at small scattering angles, observed by many investigators since 1939^[9]. The first to call attention to this were Neynaber, Brammer, and Beeman^[10-12] in 1955 and Webb and Beeman^[13,14] in 1956. This was followed by many investigations of the nature of small-angle scattering by deformed metals and alloys and by further investigations of double reflection in both single crystals and polycrystalline samples. In accordance with the

results obtained, doubts were cast on the possibility and principle of measuring the submicroscopic porosity of some crystalline materials by small-angle scattering methods^[15]. The results of the work discussed in 1958 at the Conference on Small-Angle X-ray Scattering [the transactions of the conference were published in the Journal of Applied Physics 30, 601 (1959)] have shown that the intensity of double reflections can indeed in many cases not be neglected in the study of weak diffraction effects, and, particularly, in the interpretation of the "true" small-angle scattering by metals, which is due to the presence in the specimen of electron-density inhomogeneities.

Brief reviews of work devoted to double reflection (essentially work discussed at the aforementioned conference) are contained in^[16-18]. The present review is devoted to a more systematic exposition of the results obtained up to 1963. We consider in detail the geometry of double reflections, we cite the main results of theoretical calculations and experimental measurements of their intensity, methods of separating their effects in cases when they are parasitic and interfere with the observation of other diffraction effects, and possible fields of application of double reflections in structure research.

2. GEOMETRY OF DOUBLE REFLECTIONS

a) Double Reflections in Single Crystals

The geometrical conditions for the reflection of x-rays in crystals are determined by the Bragg equation

$$2d_1 \sin \vartheta_1 = \lambda, \quad (2.1)$$

where d_1 is the interplanar distance for the set of reflecting planes (h_1, k_1, l_1) , ϑ_1 is the glancing angle for the incident and reflected rays, and λ is the wavelength of the radiation incident on the crystal; the order of reflection is assumed to be accounted for by the indices of the reflecting planes. If this condition is satisfied, then the ray incident on the crystal in a direction determined by the unit vector S_0 is reflected in a direction determined by the unit vector S_1 (Fig. 1). At some orientation of the crystal relative to the incident ray there can, in addition to the reflection from the planes (h_1, k_1, l_1) , also occur reflection from a different set of planes (h_2, k_2, l_2) . Such a reflection occurs in the direction S_2 if the following condition is satisfied

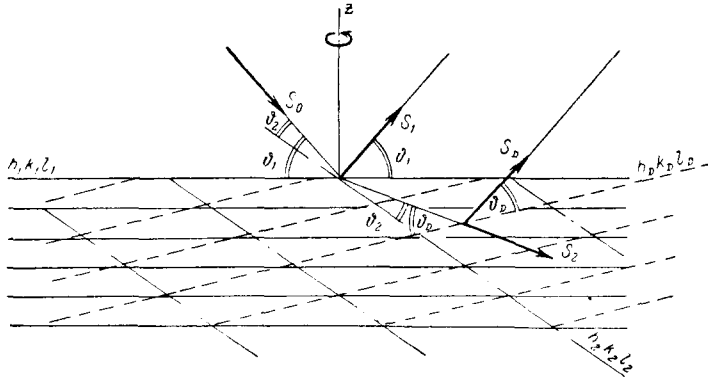


FIG. 1. Double reflection in a perfect single crystal.

$$2d_2 \sin \vartheta_2 = \lambda, \quad (2.2)$$

where d_2 is the interplanar distance for the set of planes (h_2, k_2, l_2) and ϑ_2 is the glancing angle reckoned from the planes of this set.

Conditions (2.1) and (2.2) can be satisfied simultaneously in experiments by rotating, for example, the crystal about the z axis, which is parallel to the normal to the reflecting planes (h_1, k_1, l_1) [7,8,19]. As a result of such a rotation, the planes (h_1, k_1, l_1) remain in reflecting position, so that the reflected ray is constantly observed in the S_1 direction, while reflection occurs in the S_2 direction only when condition (2.2) is satisfied.

It can be shown that if conditions (2.1) and (2.2) are satisfied, some third set of planes in the crystal $(h_D k_D l_D)$ turns out to be in the reflecting position for the reflected ray S_2 , i.e., the condition

$$2d_D \sin \vartheta_D = \lambda, \quad (2.3)$$

is satisfied, where d_D is the interplanar distance for the planes $(h_D k_D l_D)$ and ϑ_D is the corresponding glancing angle. In other words, in this case one observes double reflection: the ray which is first reflected from the planes $(h_2 k_2 l_2)$ in the S_2 direction is then reflected from the planes $(h_D k_D l_D)$ in the S_D direction. The S_D direction then turns out to be parallel to S_1 . This can be readily verified by considering the reflection conditions, using the concept of the reciprocal lattice and the sphere of reflection. Figure 2 shows part of a plane of the reciprocal lattice of the crystal. The solid circle corresponds to the intersection of the sphere of reflection with the plane of the figure. The center C of the sphere is on a line parallel to the direction of the incident ray and passing through the origin O of the reciprocal lattice, with $OC = 1/\lambda$. Since we are considering a case when reflection conditions (2.1) and (2.2) are satisfied simultaneously, the sphere of reflection passes through the points P_1 and P_2 corresponding to the planes $(h_1 k_1 l_1)$ and $(h_2 k_2 l_2)$.

The ray reflected in the direction S_2 will be regarded as the primary ray. Then the intersection between the plane of the figure, and the associated

sphere of reflection is shown by the dashed circle whose center C' is located on the line $C'O$ parallel to S_2 , with $C'O = CO$. From the center C' we draw a radius $C'P_D$ parallel to CP_1 . Then it follows from simple geometrical considerations that

$$\vec{P_D P_1} = \vec{O P_2}, \quad \vec{O P_1} = \vec{O P_2} + \vec{O P_D}. \quad (2.4)$$

But $\vec{O P_1}$ and $\vec{O P_2}$ are vectors of the reciprocal lattice, and consequently $\vec{O P_D}$ is also a vector of the reciprocal lattice. This means that on the (dashed) sphere of reflection connected with the ray S_2 there is located at the point P_D a reciprocal-lattice point and a doubly reflected ray propagates in the S_D direction. It is also obvious that $S_D \parallel S_1$. It is easy to obtain relations between the indices of the points $P_1(h_1 k_1 l_1)$, $P_2(h_2 k_2 l_2)$, and $P_D(h_D k_D l_D)$.

Since

$$\begin{aligned} \vec{O P_1} &= h_1 \mathbf{a}^* + k_1 \mathbf{b}^* + l_1 \mathbf{c}^*, \\ \vec{O P_2} &= h_2 \mathbf{a}^* + k_2 \mathbf{b}^* + l_2 \mathbf{c}^*, \\ \vec{O P_D} &= h_D \mathbf{a}^* + k_D \mathbf{b}^* + l_D \mathbf{c}^*, \end{aligned}$$

where \mathbf{a}^* , \mathbf{b}^* , and \mathbf{c}^* are the basic reciprocal-lattice vectors, it follows from (2.4) that

$$h_1 = h_2 + h_D, \quad k_1 = k_2 + k_D, \quad l_1 = l_2 + l_D. \quad (2.5)$$

Thus, a doubly reflected ray S_D is produced in the

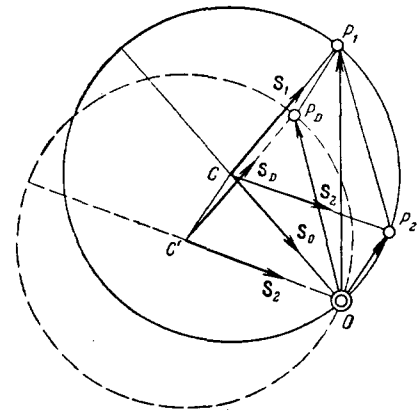


FIG. 2. Scheme of double reflection in reciprocal space.

same direction as the ray S_1 , which has experienced ordinary single reflection, with the indices of the reflections related by (2.5).

Since the condition for the occurrence of a doubly reflected ray is, in terms of the reciprocal lattice, the simultaneous presence of two (or more) points of the reciprocal lattice on the sphere of reflection, not counting the origin, we can use this condition and determine for a crystal with known structure the corresponding relations between the indices of the Bragg reflections, the wavelength of the employed radiation, and the orientation of the crystal relative to the primary beam. The general solution of such a problem is given in [20]. In the case of cubic crystals this relation is given by the formula

$$\left[\left(\frac{a}{\lambda} \right)^2 - \frac{p^2}{4} \right] \cos^2 \beta = K^2,$$

where a is the lattice parameter, $p^2 = h_1^2 + k_1^2 + l_1^2$,

$$K^2 = \frac{1}{4} \frac{[(h_2^2 + k_2^2 + l_2^2) - (h_1 h_2 + k_1 k_2 + l_1 l_2)]^2}{h_2^2 + k_2^2 + l_2^2 - \frac{h_1 h_2 + k_1 k_2 + l_1 l_2}{h_1^2 + k_1^2 + l_1^2}},$$

and β is the angle between the plane passing through the center of the sphere of reflection and the vector \vec{OP}_1 (Fig. 2) and the plane passing through the reciprocal-lattice point P_2 and the vector \vec{OP}_1 (in Fig. 2 this angle is equal to zero).

For crystals with complicated structures, double reflections should occur quite frequently [2]. Usually, however, these reflections are weak and therefore indistinguishable against the "background" of the stronger $(h_1 k_1 l_1)$ reflection, all the more since the intensification of the $(h_1 k_1 l_1)$ reflection due to double reflection $(h_2 k_2 l_2) - (h_D k_D l_D)$ is partially or completely compensated by the attenuation due to the Aufhellung phenomenon [1], i.e., the decrease in the intensity of one reflection as a result of simultaneous appearance of another. An experimental investigation of this phenomenon was made, for example, in [21] for quartz. In some cases, however, the intensity of the reflections $(h_2 k_2 l_2)$ and $(h_D k_D l_D)$ may be so high that the intensity of the reflection $(h_1 k_1 l_1)$ changes noticeably as a result of the double reflection, and this gives rise to experimental errors in the determination of the reflection intensities [2]. In the case of neutron diffraction this is demonstrated in [22-25].

If the intensity of the reflection $(h_1 k_1 l_1)$ is very small or the reflection is generally absent, being forbidden by the structure factor or by the space group of the crystal, while the two other reflections are sufficiently intense, then double reflection in the S_1 direction will be observed in more or less "pure" form. By way of an example of such a case, Fig. 3 shows the scheme for double reflection $(111) - (\bar{1}\bar{1}\bar{1})$ in a diamond crystal [26]. The figure shows the plane of the reciprocal lattice of the diamond, passing through the origin and the lines $[100]$ and $[011]$. The black circles denote those reciprocal-lattice points

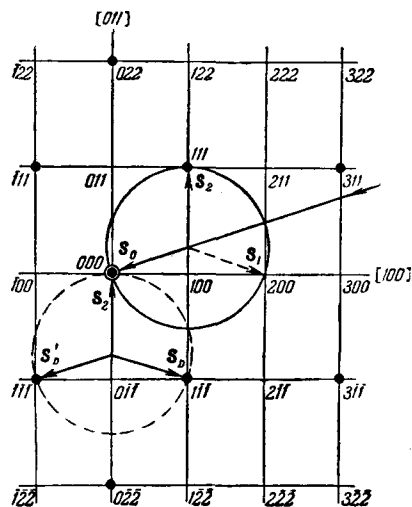


FIG. 3. Double reflection $(111) - (\bar{1}\bar{1}\bar{1})$ in diamond.

which correspond to a nonzero structure factor. For the remaining points, the structure factor is zero; these points correspond to hkl indices of mixed parity, and also to those for which $h + k + l = 4n + 2$, where n is an integer. The orientation of the crystal relative to the primary beam S_0 and the wavelength of the radiation incident on the crystal are chosen such that the sphere of reflection passes through the 111 and 200 points of the reciprocal lattice. This produces in the S_2 direction a ray corresponding to the (111) reflection. No (200) reflection occurs in the S_1 direction, since the corresponding structure factor is equal to zero. The dashes denote the intersection of the sphere of reflection connected with the ray S_2 and the plane of the figure. This sphere passes through the 111 point of the reciprocal lattice. Consequently, a doubly reflected ray is produced in the S_D direction. Since $S_D \parallel S_1$, the double reflection occurs in a direction corresponding to the forbidden (200) reflection. A double reflection $(111) - (\bar{1}\bar{1}\bar{1})$ is also produced in the direction S'_D , which coincides with the direction of the primary beam S_0 .

Double reflection in diamond in the direction of the forbidden (200) reflection was observed experimentally in [7, 8, 26]. Other investigations, in which the concept of double reflection was used to explain the occurrence of forbidden reflections, are also known. For example, in [4-6] this notion was used to study the diffraction of electrons in crystals.

Double reflections of x rays were observed by Kossel and Voges [27, 28]. A systematic study of these reflections was first made by Weigel et al. [29-30] and especially by Renninger [7, 8]. Renninger measured the intensity of the forbidden (222) reflection of diamond with $\text{CuK}\alpha$ and $\text{MoK}\alpha$ radiation for several crystals turned at different angles relative to the normal to the (111) plane. A weak (222) reflection was observed for arbitrary orientation of the crystal

relative to the normal to the reflecting planes.*

Against the background of this reflection, Renninger, working with $\text{CuK}\alpha$ radiation, observed 36 intensity maxima, occurring when the crystal is fully rotated; of these 12 were particularly strong with intensity exceeding the "background" by a factor of 10–20. Intensity maxima were also observed against the background of the forbidden (200) reflection. In the case of $\text{MoK}\alpha$ radiation, the lowest intensity of the (200) reflection was 1/6th of the average intensity of the (222) reflection, while the intensity of the maxima exceeded the minimum value by a factor of 70. The intensity of these maxima, and also their relation to the orientation of the crystal, can be attributed only to double reflection of the x rays. Renninger called this phenomenon "indirect excitation" (Umweganregung). In this author's experiments, the most intense maxima in the direction of the (222) reflection were due to the successive reflections (331) and $(\bar{1}\bar{1}1)$ or $(\bar{1}\bar{1}1)$ and (331), and also due to reflections which correspond to such cyclic permutations of these indices for which the sum remains equal to 222.

Renninger made an approximate theoretical calculation of the intensity of the double reflections with allowance for the interaction between the primary and secondary rays passing through the crystal, but without account of absorption. It turned out, in particular, that the intensity should depend strongly on the size and quality of the crystal. A more accurate calculation of the intensity on the basis of the model of the mosaic crystal, usually used in calculations of secondary extinction, with allowance for the absorption of reflected rays, was recently made by Schermer^[37].

Pabst^[38], in a study of the structure of ralstonite, observed on the x-ray rotation photographs obtained with $\text{CuK}\alpha$ and $\text{FeK}\alpha$ radiation, the 442 reflection, which is allowed by the space group, but forbidden in connection with the special positions of the atoms in this structure. It was shown, first graphically with the aid of the reciprocal lattice projection, and then by direct calculation, that this reflection is a result of the double reflection (333)—(11 $\bar{1}$).

Davisson and Havorth^[39] have attributed to double reflection the spots which were obtained on x-ray patterns when a quartz crystal was rotated together with the film about an axis parallel to the normal to the (001) plane, set at the Bragg angle corresponding to the forbidden (002) reflection of $\text{CuK}\alpha$ radiation.

*The causes of the occurrence of this reflection, and also a theoretical and experimental determination of its intensity, have been the subject of many papers^[31–36]. For example, Brill^[31] and Konobeevskii and Mamedov^[32] attributed the presence of a weak (222) reflection to the asymmetry in the distribution of the electrons in the carbon atoms in the diamond lattice: the valence electrons form in the gaps between the neighboring atoms "bonding" charges which ensure a homopolar bond.

The number of such reflections on the x-ray pattern as the crystal was rotated through 360° reached 180; the most intense of these were due to double reflections of the type (011)—(0 $\bar{1}$ 1) and (013)—(0 $\bar{1}$ 3). The total number of double reflections, calculated from the density of the points in the reciprocal lattice of quartz, was on the order of 700. However, the intensity of the majority of them was below the sensitivity of the method, and many reflections were not obtained on the film even after 72 hours of exposure.

Collin and Lipscomb^[40] have likewise attributed to double reflections of x rays the origin of the weak 302 reflection on x-ray patterns of groutite which they obtained with $\text{MoK}\alpha$ radiation, and for which reflections of the type $h0l$ with $h + l$ odd are forbidden.

Double reflection of $\text{MoK}\alpha$ radiation was claimed to be the cause of the anomalous reflections on x-ray rotation patterns of Al-Cu alloys in the work of Silcock^[41]. These were due to two successive reflections, first from one set of planes in the aluminum lattice and then from another set of planes in the lattice of the θ' phase, which is coherently bound with the lattice of the matrix (or in reverse sequence). This paper apparently reports for the first time an example of such double reflections, for which the sets of planes which produce the successive reflections belong to different structures. Because of this, the indices of the anomalous reflections due to the double reflection calculated for any one of the structural components (in^[41] they were calculated for the aluminum lattice), turn out to be neither integer nor equal to the sum of the indices of the successive reflections (if the latter are determined by the usual method for the lattices of the two structural components separately).

Bland^[42] investigated the reflections forbidden by the space group for the σ phase of the system Ni - V, the occurrence of which was previously attributed to the partial ordering of the structure of this phase. He showed that such a treatment is incorrect, since it is incapable of explaining many experimental facts. For example, it could not be understood why the forbidden reflections are observed in the case when Cr radiation was used, but were not observed in Mo radiation; why the forbidden reflections, whenever they appear not on the zero-layer of the x-ray rotation pattern, are observed only on the upper or on the lower halves of the photograph and never appear above and below simultaneously, etc. The double-reflection interpretation has made it possible for Bland to explain completely both his own observations and those of others.

Renninger^[43], continuing the investigation of reflections in diamond, showed that double reflections occur both in diamond of type I (in the classification of Robertson et al.^[44]) and in diamonds of type II, although their intensity is different. This difference

is due to the different degree of perfection of crystals of types I and II.

Fraenkel^[19] constructed a special camera in which the investigated crystal and the film were rotated in synchronism. Double reflections were registered in this camera in the form of intense spots against a weak background of forbidden reflections, obtained in the form of lines. Using $\text{CuK}\alpha$ radiation, Fraenkel observed double reflections in the direction of the forbidden (222) reflection for single crystals of diamond and germanium, but did not observe them for silicon, although, as is well known, all these crystals have the same structure. The author believes that this is due to the low degree of perfection of the investigated silicon crystal. Fraenkel and Kalman^[45] obtained with the aid of the same camera^[45] double (222) reflections for germanium not only with $\text{CuK}\alpha$ but also with $\text{MoK}\alpha$ radiation, and also for diamond with $\text{MoK}\alpha$ radiation.

Lang^[26] used for the observation of double reflections in diamond a microfocuss x-ray tube (with effective focus dimensions $30 \times 30\mu$) and recorded the reflections photographically. The practically point-like radiation source enabled him to investigate the connection between double reflections and other diffraction effects such as Aufhellung and extinction lines in the direct beam. Using $\text{AgK}\alpha$ radiation, Lang, as already noted above, observed rather intense (111)–(1 $\bar{1}\bar{1}$) double reflections.

In an already cited paper^[20] reporting experiments performed in accordance with the Renninger scheme^[8] with a setup using ionization registration of the radiation, the authors were able to observe in the course of one complete rotation of a germanium crystal about the [222] axis a total of 204 intensity peaks due to double reflections of $\text{CuK}\alpha$ radiation. Some of these peaks correspond to double (in the language of the authors, "simultaneous") reflections of "second order," when three reciprocal-lattice points lie simultaneously on the sphere of reflection.

It is easy to understand that double reflections cannot occur in crystals having a primitive Bravais lattice, in directions corresponding to forbidden reflections. Let us consider, for example, a face-centered cubic lattice. The reflections forbidden for this lattice are all those corresponding to indices $h_1k_1l_1$ of mixed parity. In order for double reflection to occur in a direction corresponding to a forbidden reflection, it is necessary that there exist such allowed reflections ($h_2k_2l_2$) and ($h_Dk_Dl_D$) which satisfy the condition (2.5), i.e., the indices $h_2k_2l_2$ and accordingly $h_Dk_Dl_D$ should have the same parity. Consequently, the indices $h_1k_1l_1$ will also have the same parity [see (2.5)], i.e., double reflection can occur only in the direction of the allowed reflection.

The direction of double reflection in such crystals can differ from the direction of single reflection only if the first and second reflections occur in different

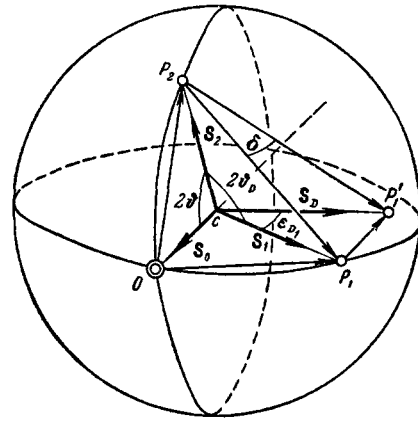


FIG. 4. Double reflection in a deformed single crystal.

slightly disoriented coherent regions (mosaic blocks) of the crystal^[46,47]. Such regions can appear, for example, under deformation.

Let O (Fig. 4) be the origin of the reciprocal lattice of such a crystal, and let P_1 and P_2 be the points on the sphere of reflection corresponding to the planes ($h_1k_1l_1$) and ($h_2k_2l_2$) in one of the coherent regions, S_0 —direction of the primary beam, and S_1 and S_2 —directions of the two rays singly reflected in this region. Then

$$\frac{S_2 - S_0}{\lambda} = \vec{OP}_2. \quad (2.6)$$

Let P' be the reciprocal-lattice point corresponding to the reflection $h_Dk_Dl_D$ in another region of the crystal, the orientation of which differs somewhat from the orientation of the first. Then the ray reflected in the first region in the direction of S_2 will be reflected again in the second region in the direction of S_D , with

$$\frac{S_D - S_2}{\lambda} = \vec{P}_2P'_1. \quad (2.7)$$

From (2.6) and (2.7) it follows that

$$\frac{S_D - S_0}{\lambda} = \vec{OP}_2 + \vec{P}_2P'_1$$

or, since $\vec{P}_2P'_1 = \vec{P}_2P_1 + \vec{P}_1P'_1$,

$$\frac{S_D - S_0}{\lambda} = \vec{OP}_2 + \vec{P}_2P_1 + \vec{P}_1P'_1.$$

But

$$\vec{OP}_2 + \vec{P}_2P_1 = \vec{OP}_1 = \frac{S_1 - S_0}{\lambda}.$$

Consequently

$$\frac{S_D - S_1}{\lambda} = \vec{P}_1P'_1,$$

which follows also directly from the figure.

Let δ be the angle between the vectors \vec{P}_2P_1 and $\vec{P}_2P'_1$, i.e., the angle between the normals to the reflecting planes ($h_Dk_Dl_D$) in the first and second region of the crystal. Then for small δ the vector $\vec{P}_1P'_1$ will be approximately perpendicular to the vector \vec{P}_2P_1 , and its length will be

$$P_1P'_1 = \frac{2 \sin \vartheta_D}{\lambda} \delta, \quad (2.8)$$

where ϑ_D is the Bragg angle for the secondary reflection ($h_D k_D l_D$).

If ϵ_{D1} is the angle between the directions S_D and S_1 , then we obtain from the triangle $P_1CP'_1$ (recognizing that the angle ϵ_{D1} is small)

$$P_1P'_1 = \frac{1}{\lambda} \epsilon_{D1}. \quad (2.9)$$

Comparing (2.8) with (2.9) we find the following relation between the angle ϵ_{D1} , which determines the deflection of the direction S_D of the doubly reflected ray from the direction S_1 of single reflection, and the angle δ which characterizes the disorientation of the coherent regions in the single crystal:

$$\epsilon_{D1} = 2\delta \sin \vartheta_D. \quad (2.10)$$

In the particular case when $\vec{P}_2\vec{P}'_1 = -\vec{OP}_2$ and S_1 coincides with S_0 , we have

$$\frac{S_D - S_0}{\lambda} = \vec{P}_1\vec{P}'_1,$$

and the angle between the direction of the doubly reflected beam and the direction of the primary beam is

$$\epsilon_{D0} = 2\delta \sin \vartheta. \quad (2.11)$$

where ϑ —Bragg angle, which is in this case equal to ϑ_D . Obtaining from the experimental data the values of ϵ_{D1} or ϵ_{D0} , we can estimate the disorientation angle δ . This was done, for example, in [48] for a deformed single crystal of copper, where the $(11\bar{1}) - (\bar{1}13)$ double reflection was observed at an angle $\epsilon_{D1} = 2.4^\circ$ from the ordinary (004) reflection, and in [46,47] for a deformed single crystal of aluminum. In the latter investigations, the maximum angle of disorientation of the coherent regions was $\delta_{\max} \approx 5.6^\circ$; it was calculated from the experimentally determined value of $\epsilon_{D0} \approx 4^\circ$ for the $(111) - (\bar{1}\bar{1}\bar{1})$ double reflection of $\text{CuK}\alpha$ radiation.

The doubly reflected ray, the direction of which is determined by Eq. (2.11), propagates in a direction close to that of the primary beam, since usually the angle δ is small (on the order of several degrees), i.e., in the region of small scattering angles. This form of double reflection is of greatest interest, and we shall therefore analyze it in greater detail.

Figure 5 shows double reflections occurring in single crystals whose structures have different degrees of perfection [49]. In all cases the plane of the figure passes through the primary and singly-reflected beams.

Figure 5a corresponds to the undeformed single crystal. If the crystal is oriented relative to the primary beam in such a way that a ray is reflected from some set of planes, this ray is reflected again from the same set of planes (but from its other side) and a beam of doubly reflected rays parallel to the primary beam is produced. In this case the doubly

reflected rays will produce in the plane of observation perpendicular to the primary beam a streak which stretches from the primary spot towards the Bragg reflection corresponding to the single reflection. The size of the streak can obviously not exceed the dimensions of the single crystal and does not depend on the distance between the crystal and the plane of observation.

Figure 5b corresponds to the same single crystal but deformed in such a way that the planes in the lattice are curved and variations in the interplanar distances are observed. In this case the doubly reflected rays will not be parallel to the primary beam and the scattering will be less intense than in case a), owing to the angular spread of the reflections. In case b) the size of the double-reflection streak near the primary spot depends on the distance between the crystal and the plane of observation. In [50] this type of double reflection is called a single-domain process.

When the single crystal consists of many slightly disoriented blocks, an x ray reflected from some set of planes in one block may be reflected again from the same set of planes in different blocks (Fig. 5c). These doubly reflected rays will travel along the generatrices of a Debye cone, the axis of which is the first reflected ray and which passes through the primary beam that has passed through the crystal. When this cone intersects the plane of observation, a Debye ring is produced passing through the spot due to the primary beam. If the region where the blocks are disoriented is small, only part of a ring is produced in the form of a streak in the region of small scattering angles. It is easy to see that this streak is stretched out in the plane of observation in a

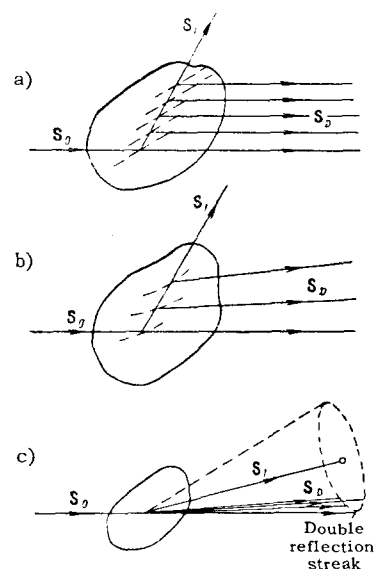


FIG. 5. Double reflections in the region of small scattering angles: a) In a perfect crystal; b) in a deformed single crystal with distorted lattice; c) in a single crystal with mosaic structure [49].

direction perpendicular to the line joining the spot due to the primary beam with the Bragg spot. In [50] this type of double reflection is called a two-domain process.

Double reflections of the type shown in Fig. 5a were observed in [51,52] for large diamond crystals and for single-crystal calcite plates.

These reflections were thoroughly investigated by Guinier and Guyon [53] in single crystals of lithium fluoride, diamond, Al-Cu alloy, and organic single crystals such as pentaerithritol and stilbene. They considered theoretically the case when a monochromatic beam of parallel rays passes through a single-crystal plate of thickness h (Fig. 6a). The primary beam strikes the point A_1 at the Bragg angle ϑ for planes which are not parallel to the surface of the plate. A_1A_2 is the path of the primary beam in the crystal. A_1B is the path of the ray reflected at the point A_1 , and β is the angle between the normal to the surface and the reflected ray. Since the first reflection can occur at an arbitrary point M on the segment A_1A_2 , while the second can occur at an arbitrary point N along the path of the reflected ray in the crystal, one should expect a continuous streak OB' , forming the trace of the doubly reflected rays, to appear in the plane of observation; this streak extends from the trace of the primary beam O towards the reflection C_1C_2 corresponding to the rays which are singly reflected at an angle ϑ . If we neglect the absorption of the rays in the crystal, then the length of the streak will be

$$a = \frac{h}{\cos \beta} \sin 2\theta = l_2 \sin 2\theta, \quad (2.12)$$

where $l_2 = A_1B$. For strongly absorbing crystals the length of the streak should be the smaller, the larger the absorption coefficient. It follows from (2.12) that the streaks will be the longer, the closer the angle ϑ is to 45° . The longest streaks for a given substance and for a given radiation will be obtained when the reflecting planes in a thin single-crystal plate are located at an angle 45° to the surface of the plate and when the Bragg angle is also 45° (Fig. 6b)

In the most favorable cases the path length l_2 can be of the order of 1–10 mm. In such cases the streak of double reflections can be readily obtained outside the beam spot. Streaks up to 5 mm long were obtained in [53] (for diamond, lithium fluoride, pentaerithritol, using $\text{MoK}\alpha$ radiation). In accordance with the predictions of the theory, the length did not change when the distance from the sample to the plane of observation was varied.

Double reflections in the region of small scattering angles, from mosaic crystals (Fig. 5c), were investigated by many workers [60,46,47,49,54–57]. The geometry of such reflections is described in greatest detail in the papers of Atkinson [54,57] for deformed single crystals containing a sufficiently large number of disoriented blocks. Atkinson assumes that the

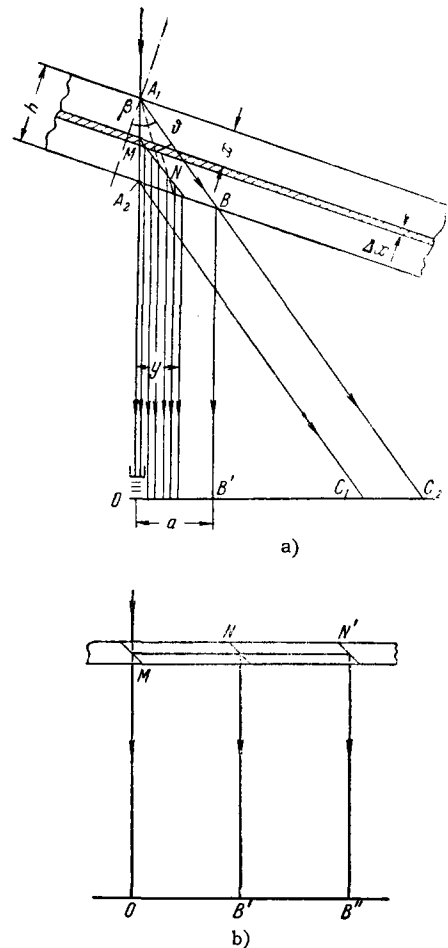


FIG. 6. a) Double reflections $(hkl) - (\bar{h}\bar{k}\bar{l})$ in a perfect single crystal; b) optimal case for obtaining a long double-reflection streak [43].

broadening of the reflections due to the dispersion of the blocks can be neglected in a theoretical analysis of the problem. In this case the details of the geometry can be calculated if one knows the orientation distribution of the coherent regions. The calculation was carried out with the aid of a stereographic projection of the reflections on a plane perpendicular to the primary beam O (Fig. 7). It is assumed that the projections of the normals to some system of reflecting planes are uniformly distributed inside the circle covered by the dots in Fig. 7a. The beam of the once-reflected rays is represented by the line pr . Each ray from this beam can be reflected once more in different regions of the crystal. For example, the ray h produces after second reflection a streak of twice-reflected rays $H'H''$. This streak is obtained in the following fashion. In order for reflections to be produced, the normals must be located on the circles marked $(\pi/2 - \vartheta)$. The reflected rays will then be located on the circles marked 2ϑ . The numbers 1 and 2 on the figure pertain respectively to the first and second reflections. To construct the pro-

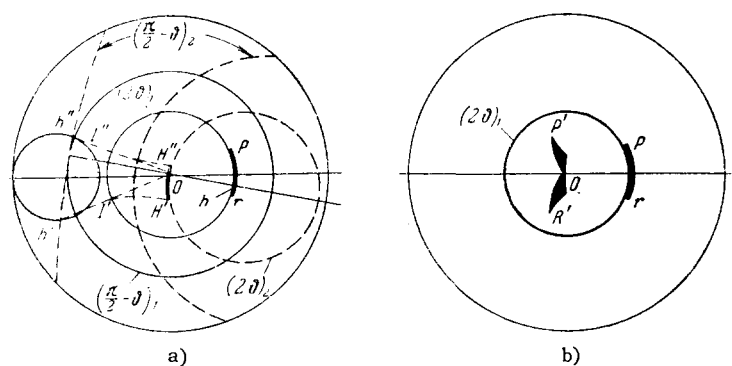


FIG. 7. Stereographic projections; a) Of double reflections $H'OH''$ occurring in the second reflection of the first-reflected ray h ; b) of all secondary reflections of the first-reflected rays pr ^[57].

jection of the rays which are produced in the second reflection of the ray h from the regions corresponding to the normals on the line $h'h''$ the projection plane must be turned in such a way as to make the ray h perpendicular to this plane. In the new projection plane, the doubly-reflected rays are determined in the same way as the singly-reflected rays were determined for the primary beam; this will be the streak $I'I''$. Rotation of the new plane of projection towards the original plane gives the sought double-reflection streak $H'H''$. These rotations are carried out with the aid of a Wulff net, the equation of which lies along the line Oh .

To obtain the projection of all the double reflections it is necessary to sum the effects produced by each once reflected ray from the beam pr . This results in the picture shown in Fig. 7b. Obviously, if the blocks have a preferred orientation, discontinuities or intensity peaks should be observed in the streak $R'OP'$ ^[49].

If the block disorientation is on the order of 1° , which in the case of single crystals of many metals corresponds to a deformation of several percent^[58], one must expect the streaks $R'OP'$ to extend up to scattering angles on the order of several degrees. In practice the effects connected with the small magnitude and deformation of the blocks cause the doubly-reflected beams to broaden. Consequently, the scattering picture due to the double reflections is not distinct in the region of very small scattering angles ($< 1^\circ$) and it becomes quite difficult to distinguish these reflections from other small-angle scattering effects by means of characteristic streaks similar to those shown in Fig. 7b. Frequently, however, the indicated crumbling and deformation of the blocks is accompanied by large disorientation (for example, for deformed nickel the first effects produce for $\lambda = 1.5 \text{ \AA}$ a reflection broadening by approximately 1° , when the disorientation reaches $\sim 10^\circ$ ^[58]). Consequently when the scattering angles are not too small ($\gtrsim 1^\circ$) the main features of the double-reflection picture (Fig. 7b) should remain. If on the other hand the distortions of the structure are not connected with large disorientation of the blocks (such

distortions can occur, for example, during hardening or irradiation), then even in the presence of double reflections the crystals may not produce the "small angle" streak which is characteristic of these reflections^[57].

Concluding thus the discussion of the geometry of double reflections in single crystals, let us present some conclusions pertaining to the conditions under which they occur and are experimentally observed.

1. Double reflections can occur in single crystals only if the crystal is in reflecting position, i.e., if at least one primary reflection is produced.

2. If the crystal is perfect, so that there is no difference whatever between the orientations of individual sections of its volume, then if the crystal has a Bravais lattice other than primitive, double reflections can be observed in the directions of the forbidden reflections. For a crystal with a primitive Bravais lattice, the directions of the doubly reflected rays always coincide with the direction of one of the allowed reflections.

3. For imperfect (say, deformed) crystals double reflections occur in directions close to the direction of one of the primary reflections. The crystal should then be in a position such that in addition to satisfying the condition for one primary reflection, noted in 1, there should also be satisfied, exactly or to some approximation, the condition for the second reflection. In this case the doubly-reflected rays are distributed over the surface of a circular cone (with vertex in the sample), the axis of which is the primary reflected ray, with the cone passing through the secondary ray.

4. For double reflections to occur in the region of small scattering angles, it is sufficient that at least one set of crystal planes be in reflecting position. In an observation plane perpendicular to the primary beam, these reflections form a characteristic streak, which for perfect crystals extends in the direction of the line joining the trace of the primary beam and the spot corresponding to the ordinary reflection, and which in the case of imperfect or mosaic crystals stretches out in a direction perpendicular to this line.

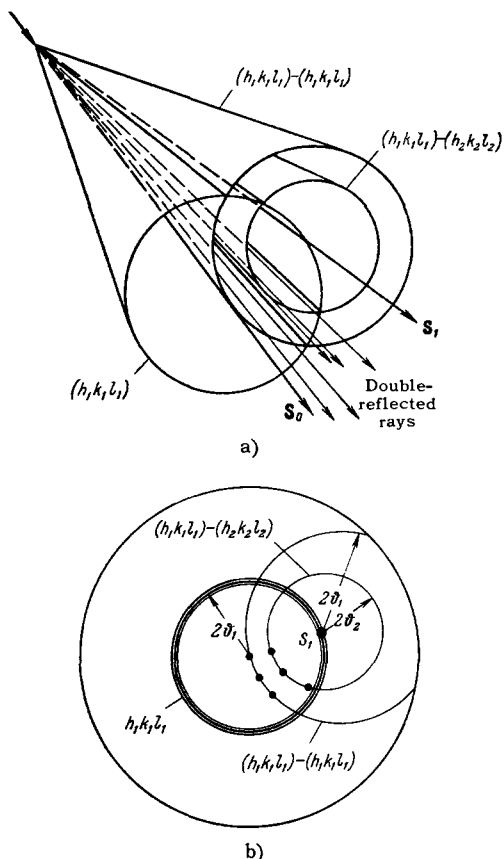


FIG. 8. Double reflections in polycrystalline samples. a) Diagram; b) stereographic projection.

b) Double Reflections in Polycrystalline Samples

For polycrystalline samples a distinction is made between internal or intragrain, and external or intergrain double reflections [46,49,60]. Double reflections are called internal if the first and second reflections occur within the confines of the same grain, and external if the first reflection is in one grain and the second is in another. The conditions for the occurrence of the internal reflections and their geometry remain the same in the case of polycrystalline samples as for single crystals [46]. The character of the external reflections is determined by the orientation distribution of the normals to the reflecting planes.

Let us consider first the geometry of external double reflections in polycrystalline specimens, when the orientation of the grains is completely disordered, and their number is sufficiently large [54]. If a monochromatic beam of x rays traveling in the S_0 direction strikes the specimen (Fig. 8a), then the resulting beams of primary reflected rays S_1 are distributed over the generatrices of the Debye cones, whose axis is the direct beam. The figure shows one such cone corresponding to the reflections $(h_1k_1l_1)$. Each of the primary reflected rays passing through the specimen can be reflected once more in other grains. The

second-reflected rays are also distributed over Debye cones, the axis of which is no longer the direct beam, but the corresponding beam of the first-reflected rays. The figure shows two cones of such twice-reflected rays; the first corresponds to reflections of the beam S_1 from the same planes $(h_1k_1l_1)$ from which the first reflections were produced (this cone passes through the direct beam), and the second is due to reflections from other planes $(h_2k_2l_2)$. The arrows indicate the individual rays corresponding to the double reflections $(h_1k_1l_1)-(h_1k_1l_1)$ and $(h_1k_1l_1)-(h_2k_2l_2)$. On the stereographic projection of the reflection pattern (Fig. 8b) the same doubly reflected rays, as well as the direct and first-reflected beams, are designated by dots. The projection plane is perpendicular to the direct beam. Therefore the direct beam is represented by a point in the center of the projection circle, while the Debye cone corresponding to the first reflections is represented by a circle centered about this point.

When the direct beam passes through the sample, several Debye cones are produced, on the generatrices of which are distributed the beams of the first-reflected rays, each of these beams producing upon second reflection a system of cones of the doubly-reflected rays. Therefore, generally speaking, the doubly-reflected rays can be directed at arbitrary angles to the direct beam, i.e., their scattering angles can vary between zero and 180° . Indeed, the region of scattering angles, at which the double reflections of the type $(h_1k_1l_1)-(h_1k_1l_1)$ occur, covers the range of angles from 0° to $4\vartheta_1$ (see Fig. 8b), while for the reflections of the type $(h_1k_1l_1)-(h_2k_2l_2)$ the range is from $2|\vartheta_1 - \vartheta_2|$ to $2|\vartheta_1 + \vartheta_2|$, where ϑ_1 and ϑ_2 are the Bragg angles corresponding to the reflections $(h_1k_1l_1)$ and $(h_2k_2l_2)$. Consequently, the reflections corresponding to angles ϑ larger than 45° are sufficient to cover the region of all the possible scattering angles. This is a feature wherein double reflections differ from the single reflections for which the scattering angles, under the same conditions, have definite discrete values. To be sure, the intensity of double reflections discussed in the present section is usually much lower than the intensity of the single reflections. Double reflections are therefore frequently difficult or even impossible to observe either photographically or by counter methods against the background of the primary reflections and the diffuse background of the x-ray pattern, the nature of which is due to other effects. However, under certain conditions these reflections are quite clearly pronounced. This is primarily true of double reflections which occur in the region of small scattering angles. It is easy to understand that the "density" of the double reflections or, in other words the probability of their occurrence, is maximal in that region, and for sufficiently small angles it is inversely proportional to the scattering angle [61]. This purely

geometric effect is due to the fact that all the cones of the double reflections of the type $(h_1k_1l_1)-(h_1k_1l_1)$ pass through the primary beam.

The observation in [10-12] of double reflections at arbitrary scattering angles was the key to the correct interpretation of the mechanism of small-angle scattering of x rays by cold-deformed metals. In these investigations, in studying the scattering by annealed metal foils (copper, nickel, aluminum) the x-ray patterns sometimes showed individual reflections at various scattering angles, most frequently between $30'$ and $4-5^\circ$. For deformed metals, such reflections never appeared. The occurrence of the reflections and their positions on the x-ray pattern were determined by the orientation of the specimen, a change of which by several degrees caused the reflections either to appear or to disappear. No regularity in their position or in their relative intensity was observed. Collimation of the primary beam has shown that only an insignificant fraction of the irradiated volume of the foil participated in the formation of these reflections. The angular width of the reflections was usually 10-15% of the scattering angle at which the reflection appeared. A check was made to disclose whether the $K\alpha$ and $K\beta$ radiations are not intermixed in these reflections (the tube had an iron target). This was done by measuring the coefficient of absorption of a known material. It was found that in most cases this coefficient corresponded to a rather pure $K\alpha$ radiation, and sometimes to almost pure $K\beta$ radiation. Attempts made to explain, at least qualitatively, the foregoing peculiarities of the isolated peaks by attributing them to some single-scattering process turned out to be inconsistent. The double-reflection mechanism, proposed for the polycrystalline foil, made it possible to explain convincingly all the observed effects. The reflections in the region of small scattering angles are produced when two relatively large grains are accidentally in favorable orientation relative to each other and relative to the direct beam, so that the ray reflected in one grain is reflected once more in the second grain and produces a reflection observed on the x-ray pattern. A statistical estimate of the number of reflections, with account of the dimensions and the divergence of the primary beam, and also the grain size [61], turned out to be in good agreement with experiment. The absence of such reflections from deformed metals is due to the distortion and the considerable decrease in the dimensions of the coherent regions following the deformation. Because of this, the intensity of the individual reflection turns out to be too small to be observed. On the other hand, the total number of double reflections increases following deformation, and this leads to an increase in the intensity of the diffuse scattering, particularly in the small-angle region.

These, and also a few other deductions of [10-14],

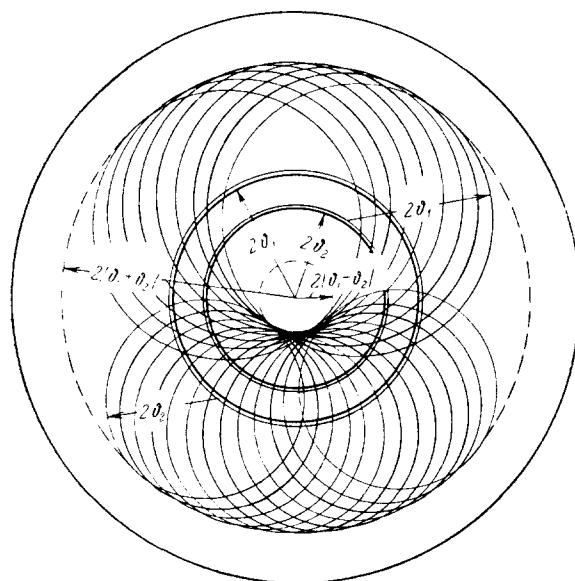


FIG. 9. Double reflections $(h_1k_1l_1)-(h_2k_2l_2)$ and $(h_2k_2l_2)-(h_1k_1l_1)$ in polycrystalline samples (stereographic projection).

connected with a study of the temperature dependence of the intensity and polarization of the scattered radiation, afforded a new interpretation of the results of earlier research (for example [9,62-70]) on small-angle scattering by deformed metals. Usually, scattering from metals was explained on the basis of notions of submicroscopic inhomogeneities in the distribution of the electron density. Such inhomogeneities may be surface defects [71-74], regions surrounding dislocations [75-83], submicroscopic pores, and cracks. The presence of so many causes makes it difficult to interpret the experimental data. This has sometimes caused some authors to refrain from any interpretation [84]. To explain the results of his own experiments, carried out for aluminum, nickel, copper and zinc, Blin [63,64] used the notion of submicroscopic pores and cracks produced during deformation. By the methods described in [85], the author determined the form and the average parameters of these pores. However, the results thus obtained appeared doubtful [86] in connection with the concept of double reflection.

In addition to the increase in intensity of double reflections with decreasing scattering angle, noted above, another likewise geometric effect is the possibility of a radical change in the intensity at scattering angles $\epsilon = 2|\vartheta_1 - \vartheta_2|$. The origin of this effect can be understood by referring to Fig. 9, which shows the stereographic projection of several cones of doubly-reflected rays $(h_1k_1l_1)-(h_2k_2l_2)$ and $(h_2k_2l_2)-(h_1k_1l_1)$. All these cones have a common enveloping cone with a half-apex angle $2|\vartheta_1 - \vartheta_2|$.

*The change in intensity should also occur at scattering angles $\epsilon = 2(\vartheta_1 + \vartheta_2)$. In this case, however, the intensity of double reflections is usually too small for experimental observation.

On this cone the "density" of the double reflections is largest. Consequently, an intense ring with a sharp inside edge, corresponding to the scattering angle $\epsilon = 2 |\vartheta_1 - \vartheta_2|$, should appear on the x-ray pattern. A change in the intensity of the double reflections in the region of small angles is also possible at scattering angles $\epsilon = 2(\pi - 2\vartheta)$, if there is a Bragg angle ϑ close to $\pi/2$. For example, for copper and aluminum with $\text{CuK}\alpha$ radiation, in the region of small scattering angles, the first ring for the double reflections (211)–(311) should be observed at scattering angles 5° and, respectively, 3.5° [57].

Such rings were observed on x-ray patterns of annealed copper powders [46, 47]. Figure 11 (see below) shows the most important theoretically-calculated peaks of intensity of double reflections at large scattering angles. Their origin is connected with the aforementioned geometric effect.

Let us consider the geometry of external double reflections for samples which have a texture. In this case the distribution of the double reflections relative to the primary beam does not have circular symmetry and depends on the distribution of the normals to the reflecting planes of the grains. In other words, the double-reflection scattering should be determined by the corresponding pole figure [57].

By way of an example, Fig. 10 shows the origin of double reflections (200)–(111) for copper foil with a rolling texture [46, 47, 56]. In the right half of the figure, the shaded area determines the position of the normals to the (111) planes that corresponds to their maximum density. The ends of the normals emerge on the front part of the projection sphere. To the left, a similar shading indicates the section of the densest distribution of the normals to the (200) planes, the ends of which emerge on the rear hemisphere. The primary beam P_1 in the direction of the observer perpendicular to the plane of the foil and to the projection plane, is reflected by the (200) planes, the normals to which are located at the point N_1 from the opposite side of the projection plane, and gives a reflected ray at the point R_1 on the front side of the projection plane. This ray is again reflected from the (111) planes, the normals to which are located on the line N_2 . This results in doubly-reflected rays in directions lying on the line R_2 . Since actually the first reflections should be mapped not by the point R_1 but by the line passing through this point, we obtain in place of the line R_2 a double-reflection streak whose boundary is marked by the dots around the line R_2 . Owing to the symmetry of the pole figure, four such streaks should be produced symmetrically relative to the point P_1 ; the locations of the three other streaks are marked by the dashed lines.

Change in the direction of the primary beam should result in a change in the double-reflection scheme. This case is considered in [47]. Experiments

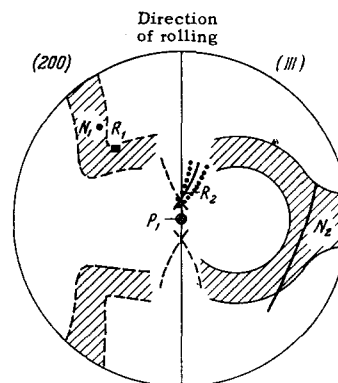


FIG. 10. Stereographic projection of double reflections (200)–(111) in polycrystalline copper having a rolling texture ($\text{CuK}\alpha$ radiation [46]).

have fully confirmed the presence of such double-reflection streaks for strongly textured specimens [46, 47, 56].

In conclusion let us specify the main features of the geometry of double reflections in polycrystals.

1. The conditions for the occurrence of internal double reflections were similar to those already noted for single crystals.
2. External double reflections in specimens with disordered orientation of the grains produce an x-ray pattern which has circular symmetry relative to the primary beam, and can occur at arbitrary scattering angles.
3. As a result of the purely geometric effects, the intensity of double reflections is maximal near the primary beam and varies in inverse proportion to the scattering angle. At small scattering angles, the values of which depend on the values of the Bragg angle for single reflections, the intensity of the double reflections changes in discontinuous fashion.
4. The pattern due to the external reflections in specimens having a texture does not have circular symmetry relative to the primary beam, and can be calculated if the texture is known.

3. INTENSITY OF DOUBLE REFLECTIONS

Theoretical calculations of the intensity of double reflections have been carried out in [8, 46, 47, 53, 59, 87–90]. In some of the papers [8, 46, 47], however, only semi-quantitative estimates were made of the order of magnitude of the expected intensity. Without dwelling on the details of the calculations, we present the most interesting results and compare them with the experiments.

We note first that for ideal crystals a dynamical theory has been developed, which takes into account multiple reflections in the calculations of the intensities of the diffracted radiation. We shall not consider the deductions of this theory, since they can be found in several books (for example [1, 91, 92]). Experiment confirms the predictions of the dynamical

theory only as an exception^[91] since crystals with a structure close to ideal are encountered very rarely. As a rule, the experimental results are close to the deductions of the kinematic theory.

Starting from the concepts of the kinematic theory, Guinier and Guyon^[53] calculated the intensity of double reflections in a single-crystal slab with negligibly small block disorientation. The calculation was made for crystals whose structure can be represented schematically by a series of parallel and slightly unevenly distributed lamellae, so that there is no interference between waves reflected by different lamellae. The authors have assumed that the beam incident on the crystal has considerable convergence and an intensity that is uniformly distributed over the entire cross section, and that all the reflections of the waves in the crystal, occurring after the second reflection, can be neglected. Under these assumptions, the integrated intensity of the once-reflected rays from a lamella of thickness Δx (Fig. 6a) is equal to

$$E_1 = E_0 Q_1 \Delta x,$$

where E_0 is the integrated intensity of the incident beam and Q_1 is the reflecting power of the lamella relative to the incident beam.

The integrated intensity of the doubly reflected beam is equal to

$$E_2 = E_1 Q_2 \Delta x = E_0 Q_1 Q_2 (h - x) \Delta x,$$

where Q_2 is the reflecting power of the lamella relative to the first-reflected beam, x is the distance from the surface of the crystal to the first-reflecting layer, and h is the thickness of the crystal. The factor $(h - x)$ takes into account the contribution made to the second reflection by all the layers located below the one in which the first reflection took place. If the intensity E_2 in the observation plane on the photographic film is uniformly distributed over a streak of length y , then the intensity of the second reflection from the individual layer, determined by the energy flux per unit length of the film, is equal to

$$\Delta I = E_0 Q_1 Q_2 \frac{h-x}{y} \Delta x = E_0 Q_1 Q_2 \frac{h}{a} \Delta x,$$

where a is the size of the double-reflection streak in the plane of observation, determined by formula (2.12). Since the reflected rays which strike the point with abscissa y after second reflection are those which were reflected for the first time from all layers, located at distances from zero to x from the surface of the crystal, we obtain for the total intensity, with allowance for absorption, the expression

$$I = E_0 Q_1 Q_2 \frac{h^2}{a^2} (a - y) \exp \left\{ -\mu \left[l_1 + (l_2 - l_1) \frac{y}{a} \right] \right\}, \quad (3.1)$$

where $l_1 = A_1 A_2$ and $l_2 = A_1 B$.

To solve the problem completely it is necessary

to know the reflecting powers Q_1 and Q_2 . These quantities have been calculated in^[53], where it is shown that for the photography geometry used by the authors Q_2 exceeds Q_1 . This is due to the difference in the structure of the beam issuing from the bent-crystal monochromator and the beam obtained by first reflection in the crystal: the angular width of the first reflection is much smaller than the angular aperture of the direct beam. Therefore the intensity of the double reflections, for sufficiently perfect crystals, can be comparable with the intensity of the primary reflections. For crystals with a clearly pronounced mosaic character, the intensity of the double reflection should be weak.

The qualitative agreement between theory and experiment was found to be good. The distribution of the intensity along the double-reflection streak, for a lithium fluoride crystal, agreed with the variation of the function (3.1). For diamond and lithium fluoride, the intensity of the double reflections was of the same order of magnitude as the intensity of the primary reflections. However, no quantitative agreement is obtained between theory and experiment. The calculated intensity turned out to be approximately 10 times larger than the observed one, apparently owing to primary extinction, which is not accounted for in the calculations, and also owing to errors in the estimate of the intensity of the direct beam. In addition, considerable fluctuations were observed on the experimental intensity-distribution curve, owing to local inhomogeneities in the structure of the crystal; it was impossible to allow for these inhomogeneities in the calculations.

The conditions for which the intensity calculation was made in^[53] are rarely realized in actual experiments, so that this work is not so much of practical interest as helpful in the understanding of the nature of those phenomena which can occur in ordinary experiments. In practice, in the region of small angles, one usually investigates scattering from thin mosaic crystals (on the order of 20–30 microns thick in the case of metals). In such crystals all the blocks cannot be in a reflecting position simultaneously (Fig. 7a), and the radiation entering into the small-angle region is that scattered from only an insignificant fraction of the volume of the investigated crystal. Owing to the small thickness of the sample, the double-reflection streaks directed towards the Bragg maximum should be very short and could hardly be observed experimentally. In this case, as we have already seen, streaks are produced which are elongated in a direction perpendicular to the line joining in the plane of observation the trace of the primary beam with the first Bragg maximum. The intensity of the double reflections should have the order of magnitude of the product of the intensity for a perfect crystal and the probability, averaged over the entire illuminated volume, of two blocks having

an orientation at which a doubly-reflected ray is produced [17]. It can be shown that this probability W depends on the average angle of block disorientation, δ_{av} , and the natural angular width φ of the Bragg reflection by an individual block, in the following fashion:

$$W(\delta_{av}, \varphi) = \frac{2\varphi}{\pi\delta_{av}}.$$

For not too strongly deformed metals, δ_{av} is on the order of $1-2^\circ$ [58]. If for the most intense reflections we assume that the order of magnitude of φ is $10''$, then $W(\delta_{av}, \varphi) \approx 10^{-2}-10^{-3}$. Since the energy of the primary reflections does not exceed several percent of the energy of the incident beam even for nearly perfect crystals, it must be concluded that the intensity of the double reflections from a deformed single crystal is not more than $10^{-4}-10^{-5}$ of the intensity of the direct beam. Consequently, the intensity of the double reflections is much smaller than the intensity of the maximum of the first reflections. Apparently, this is why the double reflections from single crystals with strongly pronounced mosaic character were not observed in [53].

The scattering produced by the double reflections in polycrystalline specimens is a superposition of reflections due to individual pairs of suitably oriented grains or blocks. The relative intensity of the external double reflections from polycrystalline samples at large scattering angles has been roughly estimated by Fricke and Gerold [46,47]. The estimate was made under the assumption that the intensity of the double reflection is proportional to the expression

$$\frac{p_1 L_1 f_1^2 p_2 L_2 f_2^2 P_{12}}{\tan \epsilon}.$$

Here p_i are the multiplicities

$$L_i = \frac{\cos \theta_i}{\sin 2\theta_i}$$

are the Lorentz factors, f_i are the atomic scattering factors, including the temperature factor, and

$$P_{12} = \frac{1 + \cos^2 2\theta_1 \cos^2 2\theta_2}{2}$$

is the polarization factor of double reflection, corresponding to a scattering angle $\epsilon = 2|\theta_1 - \theta_2|$ (i.e., to the scattering angle on the "enveloping" cone; see Fig. 9); the subscripts 1 and 2 pertain to the first and second reflections respectively, while the factor $(\tan \epsilon)^{-1}$ allows for the angular distribution of the intensity on the flat film. Calculation with the aid of this formula has shown that for annealed copper filings and for $\text{CuK}\alpha_1$ radiation, the most intense are the (111)-(200) reflections; the intensity of the double reflections of other types is smaller by at least a factor of 10. The experimental data were in general in qualitative agreement with the theory. No absolute measurements of the intensities were made in [46,47]. However, on the basis of the presented

results we can conclude that in the region of large scattering angles the intensity of the double reflections is much lower than the intensity of the ordinary Bragg reflections from powder specimens. To obtain visible double-reflection "rings" on the film, when using a vacuum camera with a focusing monochromator and with a specimen-to-film distance of ~ 25 mm, the exposure time was approximately 100 hours.

A detailed calculation of the intensity of double reflections in polycrystals at large scattering angles was made by Warren [87]. It was shown that along with Compton and thermal diffuse scattering, double reflections make a definite contribution to the diffuse background of the x-ray photograph. At scattering angles larger than $\sim 10^\circ$, this contribution is small: the intensity of the double reflections for copper powder and for $\text{CuK}\alpha$ radiation is in the mean equal to 5 electron units per atom and depends little on the scattering angle (Fig. 11). Nonetheless, double reflections can explain (at least in part) the disparity between the experimentally measured and theoretically calculated values of the intensity of the diffuse background. An example of such an explanation is given in [93] for copper polycrystalline samples.

Owing to purely geometrical effects (Sec. 2b), the intensity of double reflections in polycrystals is relatively large in the region of small scattering angles. For such angles, the calculations of the intensity of both the intergrain and the intragrain scattering have been carried out in a different paper by Warren [59]. The calculations were made for a thin powder specimen having small grains of disordered orientation, so that the beam incident perpendicular to the surface of the specimen encounters a sufficiently large number of grains.

The intensity of the intergrain scattering was

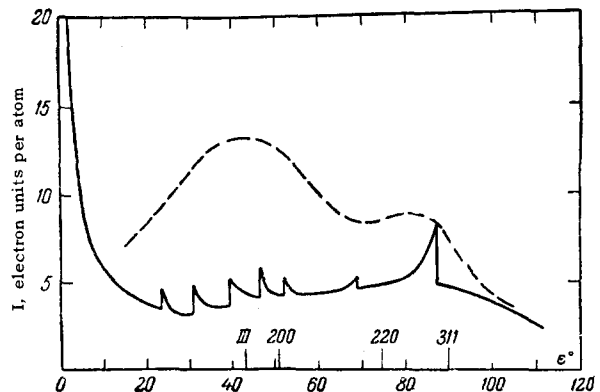


FIG. 11. Intensity of diffuse background on x-ray pattern of copper powder ($\text{CuK}\alpha$ radiation). The continuous line shows the theoretically calculated scattering intensity due to double reflections [87]. The dashed line is obtained by subtracting from the experimentally measured intensity the background of the Compton and thermal scattering intensity [94]. The abscissa axis indicates the positions of the diffraction lines.

calculated by using the ordinary intensity formulas first for the primary and then for the secondary scattering, without allowance for the temperature factor, since the main contribution to the scattering comes from low-order reflections. After some simplifications, which are permissible for small scattering angles ϵ , the following formula was obtained for the intensity of the intergrain scattering $(h_1k_1l_1)-(h_1k_1l_1)$ in absolute units:

$$I(\epsilon) = \frac{\Phi_0 K_i^2 t \exp(-\mu t) (1 + \cos^4 2\theta_i)}{16\pi^2 \mu R^2 \sin^2 \theta_i \sin 2\theta_i \sin \frac{\epsilon}{2}} \left\{ 1 - \frac{1 - \exp[-g_i]}{g_i} \right\},$$

where Φ_0 —energy flux in the incident beam,

$$K_i = \frac{e^4}{m^2 c^4} \frac{\lambda^3 p_i F_i^2}{4V^2 \sin \theta_i},$$

$$g_i = \frac{2\mu t \sin^2 \theta_i}{|\cos 2\theta_i|},$$

V —volume of the unit cell, t —specimen thickness, R —distance from the sample to the plane of observation; the remaining symbols are standard.

The intensity, in electron units per atom, with allowance for superposition of the reflections with different indices $h_1k_1l_1$, is equal to

$$I_{\text{el. un. at}}(\epsilon^\circ) = \frac{1}{\epsilon^\circ} \frac{e^4}{m^2 c^4} \frac{45\lambda^6 V_0}{32\pi^3 V^4 \mu} \sum_i \frac{p_i F_i^4 (1 + \cos^4 2\theta_i)}{\sin^4 \theta_i \sin 2\theta_i} \times \left\{ 1 - \frac{1 - \exp(-g_i)}{g_i} \right\}, \quad (3.2)$$

where V_0 is the volume per atom; the summation is over all possible reflections $(h_1k_1l_1)$, the angle ϵ is expressed in degrees. In this case the connection between $I(\epsilon)$ and $I_{\text{el. un. at}}(\epsilon)$ is determined by the expression

$$I(\epsilon) = \Phi_0 \frac{e^4}{m^2 c^4 R^2} \frac{1 + \cos^2 \epsilon}{2} I_{\text{el. un. at}}(\epsilon) \frac{t}{V_0} \exp[-\mu t]. \quad (3.3)$$

The intensity of the intragrain scattering was calculated first for a sample with a primitive cubic lattice, with allowance for second scattering by each atom of the undeformed reflecting grain. The result was then generalized to include the case of a crystal with arbitrary lattice. Rigorous calculations lead to rather cumbersome formulas. Therefore many simplifications were made, and an approximate expression was obtained for the intensity of the intragrain scattering in electron units per atom

$$I_{\text{el. un. at}}(\epsilon^\circ) = \frac{e^4}{m^2 c^4} \frac{\lambda^3 V_0 D^4}{24V^4} \sum_i \frac{p_i F_i^4 (1 + \cos^4 2\theta_i)}{\sin^2 \theta_i} \times \exp \left[- (1 - 0.20 \cos^2 \theta_i) \left(\frac{D\epsilon^\circ}{31.3\lambda} \right)^2 \right], \quad (3.4)$$

where D —average grain dimension.

According to (3.2), the intensity of the intergrain scattering varies in inverse proportion to the scattering angle, and is independent of the grain dimension. Apparently the latter must be attributed to the

fact that the intensity of both the primary and the secondary reflections is directly proportional to the volumes of the grains and the number of reflections is directly proportional to the number of grains in the illuminated section of the sample, i.e., inversely proportional to the grain volumes. The intensity of the intragrain scattering, determined by (3.4), decreases much more rapidly with increasing scattering angle than the intensity of the intergrain scattering, and depends on the grain dimensions.

Figure 12 shows curves calculated in [59] by means of formulas (3.2) and (3.4), showing the dependence of the intensity (in electron units per atom) on the scattering angle. The dashed lines correspond to the contribution from the intergrain scattering for samples of different thicknesses, while the continuous lines correspond to intragrain contribution for samples with different grain dimensions. The plot has been calculated for a copper sample and $\text{CoK}\alpha$ radiation.

For a scattering angle $\epsilon = 1^\circ$ and a sample of thickness $t = 2/\mu$, the intensity of the intergrain scattering is 40 electron units per atom. Such an intensity, although small, is perfectly adequate for experimental measurements with the aid of ordinary apparatus for small-angle scattering research. Its order of magnitude is the same as that of the intensity of scattering from deformed and annealed nickel, measured in [66].

The theory proposed is not applicable to deformed metals, since it does not take into account the considerable increase in the contribution from the intragrain scattering resulting from the development of a block structure during the course of deformation. When the grain breaks up into small subgrains or

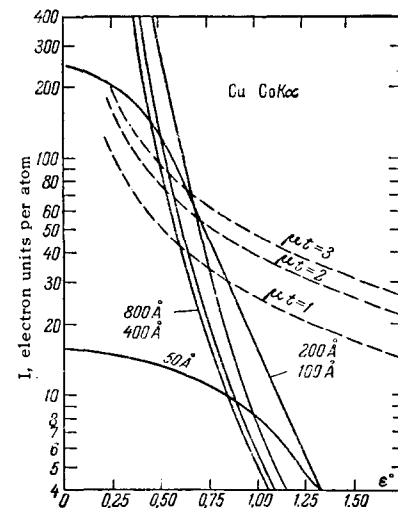


FIG. 12. Intensity (in electron units per atom) of small-angle scattering due to double reflections in powdered copper ($\text{CoK}\alpha$ radiation). The dashed lines correspond to the intergrain scattering for samples of different thicknesses. The continuous lines correspond to the intragrain scattering for different grain sizes [59].

blocks with slightly different orientations, the ray first reflected in one subgrain has a great probability of encountering other subgrains in a position favorable for a second reflection. The theory corresponding to this case is developed in [88-90].

Webb and Beeman [88] calculated approximately the total flux of the energy scattered because of intragrain double reflections in a deformed sample with initial grains having a disordered orientation. The authors have started from the following expression for the scattered flux:

$$\Phi = \Phi_0 W_1 n_1 W_2 n_2 f_1 f_2. \quad (3.5)$$

Here W_1 is the probability that a subgrain illuminated by a beam of parallel rays produces a first reflection; it is equal to $p \cos \vartheta/2$, where p is the sum of the total multiplicities and $\overline{\cos \vartheta}$ is the average cosine of the Bragg angle for all possible reflections, φ —angular width of the reflection; n_1 —number of illuminated subgrains, equal to At/l^3 , where A is the illuminated area of the surface, t the thickness of the specimen, and l is the average subgrain dimension; W_2 is the probability that the grain in the first-reflected beam produces a second reflection, and is equal to $\varphi/\sqrt{\omega}$, where ω is the solid angle of the disorientation of the normals to the reflecting planes of the subgrains; n_2 is the number of subgrains in the first-reflected beam, equal to $D(2l)$, where D is the average grain dimension; f_1 is the fraction of the incident flux, first-reflected by the subgrain, and equal to $r l^2/A$, where r is the reflecting power of the subgrain; f_2 is the fraction of the once-reflected flux, which is reflected again; its value is r .

Taking the foregoing expressions into account and substituting them in (3.5) we get

$$\Phi = \Phi_0 \frac{p(r\varphi)^2 D t \overline{\cos \vartheta}}{4 l^2 \sqrt{\omega}}. \quad (3.6)$$

For the photography geometry and the other experimental conditions employed, with $\Phi_0 = 10^9$ quanta/sec, formula (3.6) with FeK α radiation scattered by nickel, yields

$$\Phi_{\text{calc}} = 1.5 \cdot 10^5 \text{ quanta/sec,}$$

while experiment yields

$$\Phi_{\text{expt}} = 6.5 \cdot 10^4 \text{ quanta/sec.}$$

Such agreement between theory and experiment is satisfactory, allowing for the measurement errors. The experiment has shown that the total scattered flux is independent of the degree of reduction by rolling over a rather wide range (from 15 to 50%). This is ascribed to the relatively small changes occurring during the deformation in the average subgrain dimension l and the subgrain disorientation ω ; with decreasing l an increase takes place in ω , and this should in accordance with (3.6) make Φ approximately constant.

According to the cited data, the scattered flux is 10^{-4} — 10^{-5} of the flux incident on the sample. This corresponds in general with the estimate made in [17] for a deformed single crystal.

It is also shown in [88] that the distribution function of the subgrains by orientation, $p(\delta)$, and the angular distribution $\Phi(\epsilon)$ of the scattered flux, which would be observed were the contribution to the scattering due to only one set of reflecting planes, are related by

$$p(\delta) = K \epsilon \Phi(\epsilon), \quad (3.7)$$

where K is a parameter having different values for different Bragg reflections (for FeK α radiation and a nickel specimen, its values change by less than a factor of 2 for different reflections); $p(\delta)d\delta$ is proportional to the number of subgrains, for which the normals to the reflecting plane are inclined to their average direction by an angle ranging from δ to $\delta + d\delta$; $\Phi(\epsilon)$ is the energy flux per unit solid angle, scattered at an angle ϵ to the primary beam. Experiment shows that $\Phi(\epsilon)$ varies more rapidly than ϵ^{-1} , and therefore $p(\delta)$ can have a maximum at $\delta = 0$. The half-width of $p(\delta)$ is on the order of 1 — 2° and increases with increasing degree of deformation.

Since the function $\Phi(\epsilon)$, which represents a superposition of all double reflections possible for the given sample and the used radiation, is determined in the experiment, the form of the function $p(\delta)$ coincides qualitatively with the form of the function $\Phi(\epsilon)$ only at scattering angles that are not too large.

A more detailed calculation of the intensity of the intragrain scattering, with allowance for the relative contribution of each reflection ($h_i k_i l_i$)—($\bar{h}_i \bar{k}_i \bar{l}_i$) to the over-all pattern of small-angle scattering is given in the paper of Ogier et al [89]. The authors assume that a large number of grains participates in the scattering and that the initial grains break up during the deformation process into such small and disoriented subgrains, that extinction effects can be neglected.

For metals with cubic face-centered lattice, the following expression was obtained for the flux of scattered energy $d\Phi$, per element of solid angle $d\Omega$:

$$d\Phi = 8.0 \cdot 10^{-11} \Phi_0 \frac{t \exp(-\mu t) d\Omega}{\epsilon} \times \sum_i D_{\text{av}} \frac{p_i f_i^4 \lambda^6 (1 + \cos^4 2\vartheta_i)}{a^3 \sin^3 \vartheta_i \cos \vartheta_i} P_2 \left(\pm \frac{\epsilon}{2 \sin \vartheta_i} \right). \quad (3.8)$$

Here D_{av} is the average distance traversed by the first-reflected beam in the grain, equal in order of magnitude to the grain dimension; f_i is the absolute value of the atomic scattering factor with allowance for the temperature factor, a is the lattice parameter,

Table. Values of the double-reflection factor

$$\frac{\rho_i f_i^4 \lambda^6 (1 + \cos^4 2\vartheta_i)}{a^{12} \sin^3 \vartheta_i \cos \vartheta_i}$$

for aluminum and nickel ($\lambda = 1.54 \text{ \AA}$)^[89]

$h_i k_i l_i$	111	200	220	113	222	400	331	420	422	$\frac{333}{511}$
Al	1.09	0.40	0.11	0.083	0.018	0.006	0.012	0.010	0.007	0.018
Ni	39.3	11.8	2.7	2.0	0.55	0.21	0.83	1.3		

$$p_2\left(\frac{\epsilon}{2 \sin \vartheta_i}\right) = \iint p\left(\alpha + \frac{\epsilon}{2 \sin \vartheta_i}, \beta\right) p(\alpha, \beta) d\alpha d\beta,$$

$p(\alpha, \beta)$ is a function which determines the probability of the orientation distribution of the subgrains; α and β are the deflections of the normals to the reflecting planes in the subgrains from the normal in the initial undeformed grain; α and β are measured in two mutually perpendicular planes, and the summation is over all possible reflections. Here α and λ are expressed in Angstrom units and t and D_{av} are in microns. The factor

$$\frac{\rho_i f_i^4 \lambda^6 (1 + \cos^4 2\vartheta_i)}{a^{12} \sin^3 \vartheta_i \cos \vartheta_i}$$

characterizes the contribution of the double reflection of each type to the total scattered flux. The table gives the values of this factor, calculated for $\text{CuK}\alpha$ radiation, scattered by aluminum and nickel at 293°K . It is obvious that the $(\bar{1}\bar{1}\bar{1})-(111)$ and $(200)-(200)$ reflections, which determine essentially the intensity of the double reflections in the region of small scattering angles, predominate.

The scattering effect, determined by formula (3.8), is very sensitive to the angular distribution of the subgrains by orientations $p(\alpha, \beta)$. For further estimates, the authors assume that the normals to the reflecting planes are uniformly distributed over a cone with a small angle δ at the vertex; the axis of this cone is the normal corresponding to the undeformed grain. The scattered flux turns out to depend on the degree of deformation and has a maximum at some value of the latter (for nickel deformation in tension by approximately 4%). An approximate estimate yields (for not too small scattering angles) the following expression for the ratio of the maximum scattered flux to the flux in the direct beam passing through the sample:

$$\frac{\Phi_{\max}}{\Phi_0 \exp(-\mu t)} = \frac{K_{\max} D_{av} t \Omega}{\epsilon^3}. \quad (3.9)$$

Here Φ_{\max} is the scattered flux contained in the solid angle Ω , at an effective angle ϵ to the direct beam. At very small scattering angles, this formula is meaningless, for $\Phi_{\max} \rightarrow \infty$ as $\epsilon \rightarrow 0$.

The calculated values of the coefficient K_{\max} for $\text{CuK}\alpha$ radiation turn out to be 1.7×10^{-11} for alumi-

num and 7.4×10^{-10} for nickel, if D_{av} and t are expressed in microns, Ω is in steradians, and ϵ is in radians.

The experiments performed by the authors on the determination of the scattering intensity and its variation as a function of the grain dimension, degree of deformation, and a few other factors, yield results which can be well explained by the theory. In particular, for a 4% deformation in tension of nickel specimens, the measured ratio

$$\frac{\Phi_{\max}}{\Phi_0 \exp(-\mu t)}$$

turned out to be of the order of 10^{-5} , as is also obtained from formula (3.9), and confirms estimates of this kind made by others.

The theory presented above has been developed in somewhat greater detail in^[90]. For the intensity of the intragrain scattering by not too strongly deformed metals with initial grains having a disordered orientation, the following expression was obtained:

$$I_{\text{el. un. at}}(\epsilon^\circ) = \frac{1}{\epsilon^\circ} \frac{4\delta}{4\pi^2} \frac{e^4}{m^2 c^4} \frac{\lambda^6 V_0 D' p(\delta)}{V^4} \sum_i \frac{\rho_i f_i^4 (1 + \cos^4 2\vartheta_i)}{\sin^2 \vartheta_i \sin 2\vartheta_i}, \quad (3.10)$$

where

$$D' = D_{av} \exp[-\mu D_{av} \sin^2 \vartheta_j].$$

The symbol j corresponds to the reflection $(h_j k_j l_j) - (\bar{h}_j \bar{k}_j \bar{l}_j)$, which makes the largest contribution to the scattering; $p(\delta)$ is a function characterizing the distribution of the subgrains by orientation; the connection between ϵ and δ for all reflections is assumed to be determined by (2.11), where $\sin \vartheta$ corresponds to the reflection $h_j k_j l_j$. In (3.10) the product $D' p(\delta)$ is taken outside the summation sign. This is permissible in those cases when the contribution of the reflection $(h_j k_j l_j) - (\bar{h}_j \bar{k}_j \bar{l}_j)$ to the overall scattering pattern is larger than the contribution of all the remaining reflections taken together.

For a specimen of copper and for $\text{CoK}\alpha$ radiation, Eq. (3.10) takes the following form

$$I_{\text{el. un. at}}(\epsilon^\circ) = \frac{49.2}{\epsilon^\circ} [D' p(\delta)], \quad (3.10a)$$

where

$$D' = D_{av} \exp[-133D_{av}],$$

$\delta^0 = 1.16\epsilon^0$, and D is in centimeters.

By experimentally obtaining a curve ($I_{el.un.at.} \epsilon^0$), it is possible to calculate by means of (3.10) or (3.10a) the value of $D'p(\delta)$ as a function of the angle δ . By assuming with some approximation that D_{av} is of the order of half the grain dimension D , and that D is sufficiently small, we can write

$$D' = \frac{D}{2}.$$

If the grain dimension is determined by some independent method (for example metallographically), then the function $p(\delta)$ can be readily determined from the experimental data. On the basis of only the measurements of $I_{el.un.at.} \epsilon^0$ we can also determine the effective grain dimension D and the function $p(\delta)$. To do this, it is essential that the value of the expression $D'p(\delta)\delta^0$, obtained from the experimental data for $I_{el.un.at.} \epsilon^0$ with allowance for (3.10) tend sufficiently rapidly to zero with increasing δ . We can then determine without much error the area under the curve of $D'p(\delta)\delta^0$ against δ^0 . In accordance with the normalization condition for $p(\delta)$ we have

$$\int p(\delta) d\omega = \int p(\delta) 2\pi\delta^0 \left(\frac{\pi}{180}\right)^2 d\delta^0 = 1,$$

therefore

$$D' = \frac{1}{540} \int D'p(\delta) \delta^0 d\delta^0. \quad (3.11)$$

Consequently, the value of the integral in (3.11) determines D' . Knowing D' , we can easily obtain D and the absolute values of the function $p(\delta)$.

To illustrate the possibilities of the method, an example of an approximate determination of the average grain dimension and of the function $p(\delta)$ is given in [90] for deformed copper. It is shown that in this case the contribution of the intergrain scattering is insignificant and almost all the scattering is due to intragrain double reflections.

Generalizing the results of investigations involving theoretical calculations and experimental measurements of the intensity of double reflections, we can draw the following main conclusions:

1. In large single crystals with high degree of perfection, the intensity of double reflections can be comparable in order of magnitude with the intensity of single reflections. For single crystals with a clearly expressed mosaic character, the intensity of the double reflections is smaller and depends on the degree of disorientation of the mosaic blocks. For not too strongly deformed metallic single crystals, the intensity of double reflections is not more than 10^{-4} – 10^{-5} of the primary-beam power.

2. For polycrystalline samples in the large-angle scattering region, the intensity of the double reflections is relatively small. Therefore it is pos-

sible to observe effects due to these reflections only in special experiments (this calls for powerful radiation sources, long exposures, etc.).

3. At low angles, the intensity of the double reflections is much larger and can be readily detected experimentally.

4. For powder specimens with undeformed grains, the contributions from the intergrain and intragrain reflections in the small angle region are comparable in order of magnitude. At scattering angles on the order of 0.5 – 1° , their intensity can reach several dozen electron units per atom. The role of the intergrain and intragrain reflections in the general scattering pattern can be determined by investigating the dependence of the intensity on the scattering angle. The intensity of the intergrain reflections does not depend on the grain dimensions, while that of the intragrain reflections does.

5. In the case of deformed metals, the contribution from the intragrain double reflections increases. The contribution from intergrain reflections can in this case usually be neglected. The intensity of the intragrain double reflections at a given scattering angle is very sensitive to the disorientation of the coherent regions in the grain and has a maximum in the region of small scattering angles at not too high degrees of deformation. The intragrain reflections may account for up to 10^{-5} of the energy flux incident on the sample. The decisive factor in the overall scattering pattern is the contribution of one or two most intense reflections ($h_1k_1l_1$)–($\bar{h}_1\bar{k}_1\bar{l}_1$). In the case of face-centered cubic crystals, such reflections are usually the (111) –($\bar{1}\bar{1}\bar{1}$) and (200) –($\bar{2}00$).

4. SOME METHODS OF SEPARATING THE EFFECTS DUE TO DOUBLE REFLECTIONS FROM OTHER DIFFRACTION EFFECTS, AND EXAMPLES OF THE USE OF DOUBLE REFLECTION IN STRUCTURE RESEARCH

Effects due to double reflections, as already noted, can be observed at various scattering angles most frequently in the small-angle region. Therefore, in the interpretation of the experimental results of structure analysis, it is necessary to check whether the observed effects are connected with these reflections. We intend here to summarize the information on the most characteristic features of double reflections, allowance for which can make it possible to solve such a problem unambiguously in many cases.

1. Owing to double reflections, the x-ray patterns of single crystals may disclose reflections which are forbidden by the space group or by the structure factor. A change may also occur in the intensity of the allowed reflections. We have indicated in Sec. 2 some investigations in which this effect was observed and used to study double reflections, and also de-

veloped the principles of the geometrical theory of this effect, according to which double reflections can be determined from the indices of the most intense single reflections [formulas (2.5)]. If we disregard the possibility of appearance of spots due to double reflections, we can arrive at an incorrect interpretation. An example of an erroneous interpretation of this kind is given in the already cited paper^[42], in which the inconsistency of attributing forbidden reflections to partial ordering in a Ni-V alloy was demonstrated.

2. Forbidden reflections whose origin is due to double reflection can be distinguished on x-ray patterns of single crystals by their intensity and their peculiar form. Usually such reflections are weaker than others, connected with single reflection, and for crystals with weakly pronounced mosaic structure they are less smeared out^[2,41,42].

3. Owing to double reflections one observes on x-ray patterns of single crystals in the small-angle region some characteristic streaks. For weakly absorbing perfect crystals these streaks extend in the direction from the trace of the primary beam to the spot corresponding to the intense Bragg reflection, while for imperfect single crystals with mosaic structure they extend perpendicularly to the indicated direction.

4. In the case of polycrystalline samples with sufficiently large undeformed grains, double reflection may give rise to individual clearly pronounced reflections. The number and position of the reflections on the x-ray pattern are random (if we disregard the fact that they are observed predominantly at small angles) and depend on the orientation of the specimen. If the wavelength of the employed radiation and the lattice parameter of the investigated specimen are such that it is impossible to connect single-reflection acts with these reflections, their connection with double reflections can usually be established unambiguously.

5. Owing to double reflections, x-ray patterns of polycrystalline samples with small grains frequently display additional rings, the origin of which was explained in connection with the discussion of Fig. 9. Distinguishing attributes of these rings are their weak intensity (compared with the intensity of the Debye rings) and the connection between their scattering angle and the Bragg angles, a connection given by the formula $\epsilon = 2 | \vartheta_1 - \vartheta_2 |$.

6. The origin of the scattering due to double reflections can be established from the characteristic dependence of the intensity of this scattering on the temperature^[61]. Such a procedure was used, for example, by Pines et al.^[95-97] From the increase in the intensity of the small angle scattering after annealing of electrolytically deposited films of copper, nickel, and iron, the authors concluded that the observed effects cannot be due to double reflections. If

after annealing one observes a decrease in the intensity of the scattering, then final deductions concerning the connection between the scattering and double reflection call for more complicated research on the temperature dependence—measurements of the scattering intensity from samples whose temperature is varied over a wide range during the exposure. In this case, at temperatures not exceeding the recrystallization temperature, the intensity of scattering which is connected with the double reflections should vary in proportion to the temperature factor of these reflections $\exp(-4M)$ ^[61]. In calculations of this factor for polycrystalline samples account must be taken of the superposition of several double reflections.

7. A convincing criterion for the explanation of the nature of the diffuse scattering in the small-angle region is a measurement of the polarization of the scattered radiation. It is known that if the incident beam is not polarized, then in ordinary scattering at an angle ϵ to the incident beam, the radiation is partially polarized. The ratio of the intensities of the two components of the scattered beam is then equal to

$$\frac{I_{\perp}}{I_{\parallel}} = \frac{1}{\cos^2 \epsilon},$$

where I_{\perp} and I_{\parallel} are the intensities of the components connected with oscillations of the electric vector in the scattering plane and in the plane perpendicular to it, respectively (the scattering plane passes through the incident and scattered beams). For scattering angles $\epsilon \approx 1^\circ$, the ratio is

$$\frac{I_{\perp}}{I_{\parallel}} \approx 1,$$

i.e., there is practically no polarization.

If the scattering is the result of double reflection, then polarization occurs in both the primary and secondary reflections. Consequently, neglecting for the small-angle region the difference in orientation of the scattering planes for the primary and secondary reflections, we can write

$$\frac{I_{\perp}}{I_{\parallel}} = \frac{1}{\cos^4 2\vartheta},$$

since for both reflections the role of the scattering angle is played by the doubled Bragg angle 2ϑ . [This ratio was accounted for in formulas (3.2), (3.4), (3.8), and (3.10) by the polarization factor $(1 + \cos^4 2\vartheta)/2$.] Scattering due to double reflections is strongly polarized, and the degree of its polarization depends on the angle ϑ . Putting, for example, $2\vartheta = 45^\circ$ we find that $I_{\perp}/I_{\parallel} = 4$. In exact calculations it is necessary to take into account the superposition of different reflections, and also their relative contributions to the over-all scattering picture. Such calculations have been made in^[13,86] for deformed nickel. The ratio I_{\perp}/I_{\parallel} calculated theoretically for FeK α radiation with allowance for the finite dimensions of the measuring slit of the counter was 3.4. The same

ratio measured for not too high degrees of deformation is approximately equal to 3. For larger degrees of deformation this ratio decreased apparently owing to the physical broadening of the reflections accompanying the deformation, and because of the geometrical effect due to the finite height of the measuring slit.

In conclusion we note that in the case of double reflections the component I_{\perp} in the observation plane is connected with oscillations of the electric vector in the radial direction (i.e., in the direction of the line joining the trace of the primary beam with the point of observation), while I_{\parallel} is connected with oscillations of the electric vector in the tangential direction.

8. It was noted in Sec. 2 that doubly-reflected rays do not appear unless single Bragg reflections occur. For this reason, double reflections need not be taken into account in the investigation of gases, liquids, or amorphous substances. To eliminate the possibility of the appearance of double reflections in the case of crystalline materials, it is necessary to produce conditions under which no single reflections can arise. Then the effects connected with the double reflections should also disappear. In the case of single crystals this can be done by moving the crystal out of the reflecting position. For both single crystals and polycrystals this can also be done by using radiation of such a large wavelength that the reflection condition $2d \sin \theta = \lambda$ is not satisfied for the maximum value of d .

Thomas and Franks^[49], in an investigation of aluminum, used for this purpose $AlK\alpha$ radiation. However, the experimental difficulties connected with the use of this radiation did not enable them to draw any unambiguous conclusions from these experiments. Atkinson^[54,57,98] describes the use of a neutron beam, the wave length of which exceeds 6.5 Å. At such a wavelength, the Bragg reflections in the investigated samples of copper and aluminum cannot occur. By the same token, the possibility of double reflections is eliminated, and all the observed effects were due to "true" small-angle scattering by the inhomogeneities of the electron density. For deformed metals this scattering was quite weak. A comparison of the results of experiments on neutron and x-ray scattering has shown that not more than 1% of the total scattered energy goes into "true" small-angle scattering of x-rays by deformed metals, i.e., the observed scattering is almost completely due to double reflections. Since the neutron flux, even from powerful sources (nuclear reactors), is too weak compared with the flux of quanta in ordinary x-ray beams, it becomes necessary when working with neutrons to employ broad beams and to be satisfied with relatively poor collimation. Therefore, in the investigations mentioned above the experiment was carried out at much lower resolution

of the apparatus, and only the integral scattering of neutrons in a wide range of scattering angles, from 2-3 to 11-12°, was determined. Owing to the poor absorption of the neutrons, the linear dimensions of the investigated samples were several centimeters. For such dimensions, the contribution due to scattering by surface inhomogeneities of the investigated specimen is insignificant.

9. In many cases the intensity of the double reflections is very low, so that their contribution to the total scattering pattern can be neglected and it can be assumed that the scattering is due to other effects. This can be done, for example, for carbons. In^[99] small-angle scattering by graphite and lamp-black was investigated. It was shown that neither the form of the intensity curve nor the absolute measurements of the latter can be reconciled with the notion of double reflections. In the case of carbon, the classical interpretation of the experimental data, based on the concept of variations of the electron density, is valid. We have indicated above that this is true also for electrically deposited metals^[95-97]. For many aging alloys^[15,100] and apparently also for pure metals which were either hardened or irradiated by particles^[50], the role of double reflections in scattering is also small. In such cases the intensity of the small-angle scattering by inhomogeneities of the electron density can exceed by one or two orders of magnitude the intensity due to double reflections^[15]. However, even for alloys and hardened or irradiated metals it is desirable to study the nature of the small-angle scattering in each specific case, since there are investigations in which it is shown that the contribution of double reflections in the small-angle region can be appreciable for these materials^[101,102].

The authors of^[17,103-110], who studied submicro- porosity of metals and alloys by small-angle scattering of x-rays, neglected the phenomenon of double reflection. The conditions under which such neglect is allowed are spelled out in the paper of Kalikhman^[17]. On the basis of an analysis of previously published data, as well as those obtained in his own research, the author has concluded that an increase in the intensity of scattering by polycrystalline metals and alloys can hardly be attributed to double reflections at very small scattering angles (down to 3-4' from the edge of the primary beam). At such angles, the contribution from double reflections can be neglected if the dimensions of the submicroscopic pores are of the order of 200-2000 Å, and if their total volume amounts to not less than 0.02-0.03% of the total volume of the investigated sample.

We now proceed to consider the possibilities of employing double reflections in structure research. In the opinion of several authors^[15], a study of double reflections is usually of little interest, since the data obtained thereby can be acquired by simpler methods in the investigation of single reflections.

However, many researches offer evidence that double reflections can yield information on the structure of the investigated object, information which is difficult to obtain by other methods, or else make more precise the data obtained in the study of single reflections. Let us present examples of investigations of this type.

It was noted in Sec. 3 that scattering due to double reflections is very sensitive to the angular distribution of the coherent regions with respect to orientation. Therefore in some investigations double reflections were used to study the degree of perfection of crystals and the disorientation of the mosaic blocks.

The possibility of using double reflections in the study of the degree of perfection of single crystals was pointed out in a paper already cited above^[53]. In this investigation the imperfections of the crystal are related to inhomogeneities in the distribution of the intensity in the small-angle-scattering streak. This connection is not simple, since doubly reflected rays arrive at an identical point on the film from different sections of the crystals, and an integrated effect is observed, from which it is impossible to establish uniquely the distribution of the structural inhomogeneities. Therefore a study of the degree of perfection of single crystals by means of double reflections can be made only qualitatively.

Double reflections were used in^[111] to study the degree of perfection of diamonds of types I and II. The authors have found that double reflections in diamonds of type II are more intense than in diamonds of type I. In addition, for diamonds of type II the direction of double-reflected rays can differ somewhat from the direction of the forbidden (222) reflection, in which double reflections should occur for a perfect crystal. On the basis of this it was established that the mosaic blocks in diamonds of type II are larger in dimensions but are less ordered than those in diamonds of type I.

Betekhtin and Slutsker^[60] measured the intensity of double reflections in the region of small scattering angles, and determined the disorientation of mosaic blocks in aluminum. To this end, they used the connection between the distribution function of blocks by orientation, $p(\delta)$, and the angular distribution of the scattering intensity, determined by formula (3.7). A Gaussian distribution function $p(\delta)$ was assumed:

$$p(\delta) = C \exp(-k^2\delta^2),$$

where C is a coefficient determined from the normalization conditions and k is a parameter connected with the average disorientation angle by the relation

$$\delta_{av} = 0.88k^{-1}. \quad (4.1)$$

The scattering was assumed to be due to double reflections (111)–($\bar{1}\bar{1}\bar{1}$).

For the chosen type of the function $p(\delta)$, and with allowance for the features of the experimental array used in^[112], the angular dependence of the intensity of the scattering was given by the function

$$I(\epsilon) = M \exp(-3k\beta\epsilon), \quad (4.2)$$

where M is a coefficient which takes into account K from formula (3.7), the geometrical characteristics of the setup, etc., and $\beta = \frac{1}{2} \sin \vartheta_{111}$. The experimental data was used to plot $\log I(\epsilon)$ against ϵ , which according to (4.2) should be a straight line. The slope of the line yields the parameter k , while formula (4.1) yields δ_{av} . In spite of many approximations (the authors, for example, did not take into account the superposition of the double reflections of different types), it is convincingly shown in this work that the method of investigating double reflections can be successively used to determine the disorientation of mosaic blocks along with the microbeam and double crystal monochromator methods.

It was established in^[113] that a unique connection exists between the change in the strength characteristics of aluminum subjected to different heat treatment, hardening, and alloying and the disorientation angle δ_{av} determined by the method described above. The connection between hardening and the angular disorientation, determined also by the investigation of double reflections, was found in^[114] for nickel and iron.

Another promising field of application of double reflections is the study of the kinetics of recrystallization. This is due to the fact that the intensity of double reflections depends strongly on the grain dimensions. For intragrain reflections in powdered specimens this follows from (3.4). An analogous relation is valid also for annealed metal foils. In this case the intensity of the primary reflections is proportional to the volume of the grain, while the intensity of the secondary reflections is proportional to the grain diameter^[114]. Consequently, the data obtained by study of double reflections may be a useful supplement to the information concerning the mechanism of the recrystallization process, information obtained by other methods. Examples of recrystallization investigations by measuring the intensity of scattering due to double reflection are the investigations of Ogier et al.^[114-116] It is shown in these investigations, in particular, that rapid recrystallization in deformed nickel occurs by a growth of the blocks, the activation energy of this process being 2.2 eV.

We have mentioned in Sec. 2 that double reflections can be used to determine more precisely the texture in polycrystalline samples. Other regions of utilization of double reflections are also known, for example precision determinations of the lattice parameter of the crystal^[8,20] and a direct determination of the phase of the reflections^[117-120]. We

shall not dwell on these investigations, for so far they are isolated and their analysis calls for a special discussion.

CONCLUSION

Summarizing the foregoing, we must note that double reflections can occur quite frequently in structure investigations. The effects due to them are determined to a considerable degree by the fine structure of real crystals and are observed most frequently in the region of small scattering angles. These features of double reflections were successfully defined by Betekhtin and Slutsker^[60]: "The phenomenon of double reflection is based essentially on the fact that the volume of the mosaic crystal is a large aggregate of very small double-crystal monochromators (pairs of slightly disoriented regions) whose "action" produces the small-angle scattering."

The intensity of the double reflections at small angles is relatively high, and they can fully mask the "true" scattering by inhomogeneities of the electron density of crystalline materials. This must be taken into account both in cases when double reflections are parasitic, and in determining the possibility of their use for structure study.

The number of investigations devoted to the use of double reflections in structure research is small, and the possibilities of the method have been far from exhausted in this respect. It is therefore important to know these possibilities, since a study of double reflections may be a good supplement to other diffraction methods.

Very little was said in this survey about the features of the experimental setups used to study double reflections. This is due to the fact that in many cases the ordinary installations used for structure analysis, which ensure a sufficiently intense and well collimated monochromatic primary beam and eliminate parasitic scattering (scattering by air, edges of slits in the collimation system, etc.) are fully applicable for this purpose. For example, in the study of double reflections in the region of small angles of scattering, it is very convenient to use cameras with crystal monochromators, focusing the beam into a line or a point^[50,57,69,85], and also a high-intensity camera with "semi-infinite" primary beam^[112]. Some experimental schemes used for the study of double reflections are described in detail in^[8,19,46,61,88,121,122].

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