EXPERIMENTAL INVESTIGATION OF THE ELASTIC QUASI-MOSAIC EFFECT

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The elastic quasimosaic effect[4] arising upon diffraction of x-rays or gamma rays by elastically deformed anisotropic single crystals is investigated experimentally. This effect was previously investigated theoretically[5,6,9] in connection with the explanation of the features of focusing crystal-diffraction spectrometers[6,7]. Satisfactory agreement with the theory has been found. It is pointed out that the effect can be employed in the design of diffraction instruments. As shown in the present work, the resolving power can thus be considerably increased (by 7—8 times).

INTRODUCTION

It was observed experimentally already back in the early 30's that the diffraction of x-rays by a number of single crystals (particularly quartz) depends on the elastic stress applied to the crystal[1-3]. At that time the effect was qualitatively explained as being due to bending of the reflecting planes and to change in the interplanar distances due to the inhomogeneous elastic deformation[2].

In 1950, in a study of the diffraction of x-rays from single-crystal quartz plates elastically bent into a cylinder (under the conditions of the focusing Cauchois-Du Mond diffraction gamma spectrometer), Lind, West, and Du Mond observed[4] that the dependence of the integral reflection coefficient R on the wavelength λ is quadratic, whereas the same dependence for the same unstressed plates is linear. Lind, West, and Du Mond proved the geometrical identity of the cylindrical focusing variant and the plane-parallel variant with unstressed (flat) plates and were therefore forced to explain the observed phenomenon by postulating a special elastic mosaic effect (i.e., one which vanishes after the stress is removed), the mechanism of which remained unexplained.

In 1957, the observed effect was explained quantitatively and turn out to be the consequence of violation, during the bending of the anisotropic plate, of the so-called Bernoulli hypothesis, i.e., elastic bending of the normal transverse cross sections (reflecting planes)[5].

This phenomenon is of interest because it can occur in focusing diffracting-crystal gamma spectrometers, which have found extensive use in the 50's and 60's, and can thus influence strongly such characteristics of these instruments as the resolution, transmission, sensitivity to low energy shifts of the investigated lines[6-8]. The theory of γ-ray diffraction by elastically bent anisotropic single crystals was therefore subsequently developed in greater detail[6-9].

A comparison of the experimentally obtained parameters of diffraction instruments with the theoretically calculated ones, with allowance for the foregoing effect[6,7], as well as a direct utilization of certain consequences of the theory of the effect for the purpose of improving these parameters[8,10,11], have shown in all cases good agreement with theory; however, insofar as we know, no special experimental investigations of the phenomenon or an experimental verification of its most distinguishing feature have been performed to date.

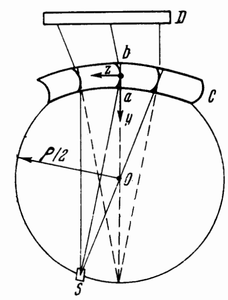

FIG. 1. Schematic diagram of diffraction in the cylindrical geometry characteristic of focusing Cauchois–Du Mond spectrometers. C – bent crystal, S – linear source (Du Mond geometry) or receiving slit of detector (Cauchois geometry), D – extended detector (Du Mond geometry) or source (Cauchois geometry).

THEORY OF THE EFFECT

We present, following[5,6], the main relations of the theory of this effect. Let us consider (see Fig. 1) a plane-parallel single-crystal plate elastically bent along a cylinder of radius ρ. The solution of the corresponding theoretical problem of the elasticity of an anisotropic body (see, for example,[12]) has shown that the reflecting planes ab, which coincided prior to the bending with the normal transverse sections of the plate, become curved after the bending, assuming the form of parabolic surfaces:

$$z = ky^2, \quad k \approx \left(a_{34} - \frac{a_{45}}{a_{55}} a_{35}\right) \Big/ 2\rho\left(a_{33} - \frac{a_{35}^2}{a_{55}}\right), \qquad (1)$$

where a_{ij} are the components of the elastic tensor.

An analysis of the diffraction of γ radiation emitted by a line source S located on a focal circle (circle with radius $\rho/2$ centered at the point 0), i.e., in the geometry characteristic of the focusing Cauchois-Du Mond diffraction instruments, has shown that a single crystal which is ideal prior to the bending reflects under these conditions like a mosaic crystal with an angular mosaic-distribution width equal to 2kL, where L is the thickness of the plate (see[6], p. 27). This conclusion, as shown recently by Alekseev[13], is particularly obvious within the framework of the model of an ideal mosaic crystal. Indeed, assume that prior to bending the plate was an ideally-mosaic single crystal with a mosaic-block angular distribution described by the re-

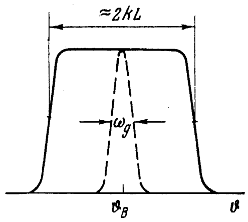

FIG. 2. Angular distribution of the mosaic blocks at k = 0 (dashed) and at $2kL > \omega_g$. At high energies of the diffracted radiation, i.e., when $2W_\Sigma(\vartheta - \vartheta_B) R << 1$ (see relation (4)), the curves represent the shapes of the diffraction lines.

lation

$$dn / d\vartheta = W_1(\vartheta). \quad (2)$$

it is customary to use for $W_1(\vartheta)$ a Gaussian distribution (see Fig. 2, dashed curve). Flexure superimposes on the initial distribution a distribution $W_2(\vartheta)$ due to the bending of the reflecting planes. The function $W_2(\vartheta)$ has the form of a rectangle of width 2kL; from (1) it follows that

$$dz / dy = \vartheta = 2ky,$$
$$W_2(\vartheta) \equiv \frac{dy}{d\vartheta} = \begin{cases} 1/2k - \text{const} & \text{if } |\vartheta| \leq kL \\ 0 & \text{if } |\vartheta| > kL \end{cases}. \quad (3)$$

The total distribution $W_\Sigma(\vartheta)$ is shown in Fig. 2 by the solid line. The model of an ideal mosaic crystal, by definition, presupposes that the radiation scattered by the individual blocks is incoherent (randomness of phases), and therefore within the framework of this model the crystal with a composite distribution $W_\Sigma(\vartheta)$ (Fig. 2) due to both the natural mosaic structure and to the bending of the reflecting planes is indistinguishable from a crystal having only natural mosaic structure, but with the same distribution as the total distribution in Fig. 2. Consequently, the process of diffraction by the considered rather specific system can be described exactly, within the framework of the model of an ideal mosaic crystal, by means of an equation derived for an ordinary ideal mosaic crystal[14,15] 1):

$$I(\vartheta - \vartheta_B) = {}^1/{}_2[1 - \exp\{-2W_\Sigma(\vartheta - \vartheta_B)R\}],$$
$$R = \frac{p^2}{\cos^2\vartheta} r_0^2 \left(\frac{F}{V}\right)^2 \lambda^2 Ld. \quad (4)$$

here $I(\vartheta - \vartheta_B)$—coefficient of reflection from the crystal, for radiation incident at an angle ϑ close to the Bragg angle ϑ_B; p—factor taking into account the polarization of the incident radiation; $r_0 = e^2/mc^2$—classical radius of the electron; F—structure factor; V—volume of crystal unit cell; d—interplanar distance between reflecting planes; λ—wavelength of diffracted radiation.

Starting from the tabulated values of the elastic constants of the crystals, referred to the coordinates xyz of the plate, which are congruent with the crystallographic coordinates, and transforming the constants by rotation from the crystallographic coordinates to those of the chosen cut in accordance with the usual rules for tensor-component transformation (see, for example,[12], p. 36), we can easily calculate the value of the flexure coefficient k. Such calculations, performed in particular for a number of quartz plates, have shown that k can range from 0 to 1×10^{-3} cm^{-1} (ρ = 200 cm)[6,9]. The width of the distribution $W_\Sigma(\vartheta)$ changes in this case from $\omega_g = 0.3-1''$ to $\omega_\Sigma = 2$ kL = 50—100″ (for plates 1—2 mm thick). Thus, a crystal possessing a narrow angular distribution of the mosaic blocks prior to flexure, i.e., a nearly ideal crystal, will begin to behave after flexure (in the case when $2kL \gg \omega_g$) like a crystal with a much more developed mosaic structure. Such a change of the properties of crystals as a result of elastic flexure can be called the effect of elastic quasimosaic structure.

Generally speaking, this effect should take place in elastic flexure of a single crystal of any symmetry (any of the nine classes of elasticity). It vanishes identically only when the reflecting plane (normal cross section of the plate) coincides with the plane of the elastic symmetry of the crystal, or when the longitudinal z axis of the plate coincides with the elastic symmetry axis of any order, owing to the vanishing of the elastic-constant tensor components responsible for the bending of the cross sections (see[12], p. 22).

After choosing the reflecting plane and aligning it with the normal cross section of the cut plate, there always remains one more degree of freedom—the possibility of rotating the plate around the longitudinal axis z[9]. Under such a rotation, the chosen crystallographic plane remains congruent with the normal cross section; the parameters characterizing the diffraction by the unbent crystal, namely d, F, and V (see relation (4)), and apparently also ω_g, will likewise remain unchanged, but the flexure coefficient k will change in accordance with the transformation of the elasticity-tensor components.

The dependence of k on the angle of rotation φ around the z axis is given the general case by the expression[6,9]

$$k = \frac{1}{2\rho} \frac{(a_{34}a_{55} - a_{35}a_{45})\cos\varphi + (a_{45}a_{34} - a_{44}a_{35})\sin\varphi}{(a_{33}a_{44} - a_{34}^2)\sin^2\varphi + (a_{45}a_{33} - a_{34}a_{35})\sin 2\varphi + (a_{33}a_{55} - a_{35}^2)\cos^2\varphi} \quad (5)$$

where a_{ij} are the components of the elasticity tensor, calculated for the initial position of the plate (the position from which the angle φ is reckoned). When the coefficients of cos φ and sin φ vanish (their vanishing corresponds to the aforementioned cases of the identical vanishing of k), the equation

$$(a_{34}a_{55} - a_{35}a_{45})\cos\varphi + (a_{45}a_{34} - a_{44}a_{35})\sin\varphi = 0 \quad (6)$$

always has the solution, i.e., it is always possible to choose the angle φ such that the elastic quasimosaic effect vanishes. Figure 3 shows by way of an example plots of $k(\varphi)$ calculated for quartz plates having differ-

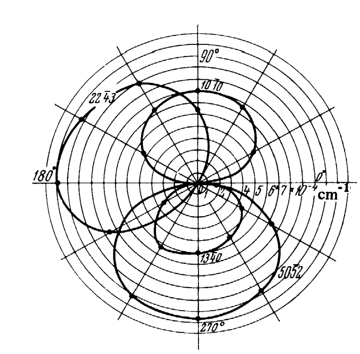

FIG. 3. Dependence of the flexure coefficient k on the angle for a number of quartz plates. The curves indicate the indices of the planes coinciding with the normal cross sections of the plates.

1)Insofar as we know, Eq. (4) was first used for the calculation of the integral reflection coefficient from a bent crystal in [16]. However, they used for $W_\Sigma(\vartheta)$ a Gaussian distribution with a width chosen to get best agreement with experiment.

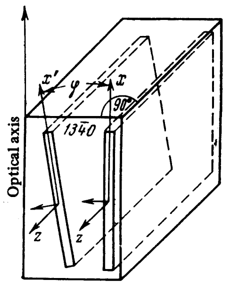

FIG. 4. Position of compared plates in the single-crystal quartz block.

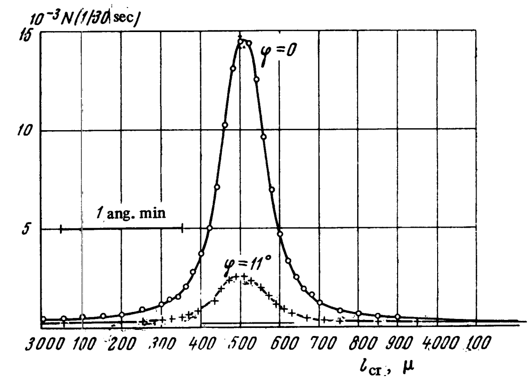

FIG. 5. K_{α_1} Sn x-ray line obtained with a two-meter Cauchois spectrometer using compared plates of 1 mm thickness. Receiving slit ≈ 250μ; N – number of γ quanta registered in 30 sec; l_{cr} – linear displacement of the end of the one-meter arm of the crystal drive. The vertical scale is doubled for the = 11° line.

ent orientations (the indices of the planes coinciding with the normal cross sections are indicated on the curves)[9].

The purpose of the present paper is to present a comparison of the experimental and calculated parameters of the diffraction process for different values of the flexure coefficient k(φ), and particularly a verification of the behavior of the elastic quasimosaic effect at the critical angle φ calculated from (6), i.e., a comparison of the most distinguishing characteristic properties of the effect with experiment.

DESCRIPTION OF EXPERIMENTS AND RESULTS

We cut from a single crystal of optical α-quartz a parallelepiped (Fig. 4) whose front face coincided accurate to ≈ 1′ with the 1340 plane, and the side edge coincided with the optical axis. We cut from the block two plates measuring 50 × 30 × 1 mm, such that one of them was parallel to the side face of the parallelepiped (the vertical edge was parallel to the optical axis), and the second was rotated around the z axis by an angle φ = 11.0°. (This value is the solution of (6) when the tabulated values of the elastic constants of quarta are used[2].) The side faces of the plates were made flat accurate to 2–3 optical interference fringes, and were made mutually parallel with accuracy ≈ 1 μ. The plates were alternately clamped between the steel mirrors of a focusing Cauchois diffraction spectrometer[8], and were bent elastically into a cylindrical surface with radius ρ = 200 cm.

Figure 5 shows the K_{α_1}Sn x-ray lines obtained using the compared plates. All the remaining conditions of the experiment remained in this case completely unchanged. The width of the receiving slit of the detector was ≈ 250 μ.

Rotation of the plate by ≈ 11°, on approaching the critical value (for the same reflecting planes $13\bar{4}0$ and for the same orientation of these planes in the plates), changed the reflectivity of the crystals radically. The ratio of the areas under the lines, which equals the ratio of the integral coefficients of reflection, turned out to be

$$S_{\varphi=0} / S_{\varphi=11^\circ} \equiv R_0 / R_{11} = 11 \pm 1. \qquad (7)$$

Integrating (4) and choosing as $W_\Sigma(\vartheta - \vartheta_B)$ for the plate with φ = 11° (k ≈ 0) a Gaussian distribution with ω_g = 0.3–0.5″ (see footnote[2]) and for the plate with φ = 0 a rectangular distribution of width 2kL = 8″ [3], and using the tabulated values of the remaining parameters, namely F = 21, V = 112 Å³, d = 1.12 Å, L = 0.1 cm, $r_0 = 2.82 \times 10^{-5}$ Å, and λ = 0.49 Å, we get[4] R_0/R_{11} = 6–11, depending on the assumed value of the width of the mosaic of the initial crystal.

Thus, the experiment has confirmed the presence of an elastic quasimosaic effect and the satisfactory agreement between the value of the critical angle φ and the observed change in the reflectivity, on approaching the critical angle, with estimates based on the theory proposed above for the phenomenon. The obtained data, however, concerned only the integral characteristic of the reflectivity (the integral reflection coefficient). Undoubtedly, more information can be gained from an experiment that yields directly the shapes of the diffraction lines in the comparable cases (see Fig. 2 above and relations (1) and (4)). The main difficulty lies in this case in the need for eliminating or, reducing as much as possible, the influence exerted on the line shape by other factors, such as the aperture aberration and the finite width of the receiving slit of the spectrometer, and the proper (physical) width of the investigated spectral lines[6]. This problem is particularly difficult in the case of a plate with k ≈ 0. (φ = 11°), since the expected line width should be close to ω_g (dashed in Fig. 2), i.e., of the order of one second of angle.

[2]Control calculation performed during the writing of this paper have shown that the critical value of φ changes noticeably when data of different authors are used for the elastic constants of quartz. The values assumed to be the most reliable (see, for example, [17]) correspond to φ = 12 – 13°, so that the orientation error of all our plates with respect of φ was on the order of 1.5°, corresponding to a residual quasimosaic effect 2kL ≈ 1″. However, in view of the approximate nature of relation (1) and accordingly (5) (see [6,18], in view of possible errors in the orientation when the plates are prepared, and also in view of the inaccurate knowledge of the shape and width of the distribution of the most natural mosaic of the unstressed crystal, we preferred not to take this correction into account and to consider henceforth the total mosaic effect at k ≈ O as a parameter that is free to a certain degree, is somewhat arbitrarily identified with ω_g, and is chosen to obtain best agreement with experiment.

[3]The value of the flexure coefficient k(0) calculated from relation (1), using tabulated values of the elasticity tensor components (expressed in terms of the coordinates of the plate in Fig. 4), is (1.83 – 2.02) × 10^{-4} cm^{-1}, depending on which set of constants was employed, In our calculations we assumed $k_{theor} = 2 \times 10^{-4}$ cm^{-1}.

[4]In these and subsequent calculations by means of (4) we used the table and graphs from [13].

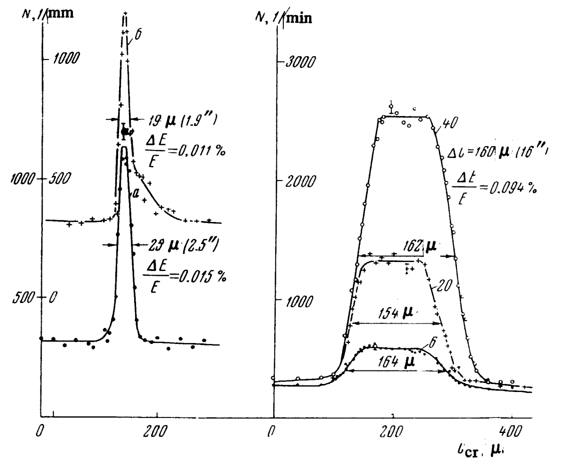

FIG. 6. 63-keV gamma line of Tm^{169}, obtained using compared plates 1.7 mm thick. Right – plate with $\varphi = 0$, receiving slit equal to 40, 20, and 6μ respectively; left – plate with $\varphi = 11°$, receiving slit 6μ, N – number of γ quanta registered in one minute.

New cylindrical mirrors were prepared, whose surfaces ($\rho \approx 200$ cm) were verified by means of cylindrical test glasses. The maximum distance between the test glass and the steel mirror when the two were in optical contact did not exceed 0.6 μ (approximately two interference fringes). New plates were prepared with $\varphi = 0$ and $\varphi = 11°$, each 1.7 mm thick (when the thickness is larger it is easier to ensure sufficient flatness of the large faces). These measures should have decreased the aperture aberration due to the deviation of the large faces of the bent plate from an ideal cylinder. In order to reduce the aperture aberration further, the working surface of the crystals (35 × 11 mm) was covered with a lead diaphragm with an aperture (5 mm diameter) over the center of the working surface.

The edges of the receiving slit of the detector were made of tungsten plates 1 mm thick, the working surfaces of the "knife edges" were polished on a plane with accuracy to 1 μ. The source was a tablet of Yb^{168} oxide (23% enrichment), irradiated by a flux of thermal neutrons ($\approx 5 \times 10^{13}$ neutron/cm^2-sec). We investigated the 63-keV gamma lines emitted upon transformation into the daughter Tm^{169}.

Figure 6 (right) shows the lines obtained with the plate having $\varphi = 0$ at 3 values of the width of the receiving slit of the detector, 40, 20, and 6 μ, respectively. The expected width of the image ($2k_{theor}L\rho = 140$ μ) on a focal circle with $\rho = 200$ cm exceeded the chosen values of the slit width. It is easy to see that in this case the line width at half height should not depend on the slit width and should equal[5)] $2kL\rho$. The experiment yielded a value 160 ± 6 μ. Thus, we get for the experimental value of the flexure coefficient k

$$2k_{exp}L\rho = 160 \pm 6\ \mu \qquad k_{exp} = 2.4 \cdot 10^{-4}\,\text{cm}^{-1}$$

which is in satisfactory agreement with the theory[6)]:

$$k_{theor} - k_{exp} / \bar{k} \approx 20\%.$$

The characteristic line shape, with a flat top, is satisfactorily reproduced (see Fig. 2). The latter circumstance is essential, since it serves as an experimental verification of the quadratic character of the bending of the reflecting planes (see relation (3)).

The left side of Fig. 6 shows the lines obtained with a plate with $\varphi = 11°$ ($k \approx 0$). The width of the detector slit was 6 μ. The line width fluctuates between 19 and 25 μ (1.9–2.5″), i.e., it is smaller than in the case $\varphi = 0$ by a factor 6.5–8.5. In the case $\varphi = 11°$, the influence of the aperture aberration is noticeable: the shape and width of the line change somewhat, depending on the choice of the position of the lead diaphragm (5 mm diameter) within the limits of the window of the crystal holder (profiles a and b of Fig. 6).

Comparing the lines with $\varphi = 0$ (receiving slit 6 μ) and $\varphi = 11°$, we get for the ratios of the line areas (the integral reflection coefficients) and the number of counts at the maxima (maximum reflection coefficients)

$$S_0 / S_{11} \equiv R_0 / R_{11} = 3.4 \pm 0.2,$$
$$\Gamma_{11} / \Gamma_0 \equiv I_{11}(0) / I_0(0) = 2.5 \pm 0.2.$$

Calculation by means of (4) with $E_\gamma = 63$ keV, $k = 2.4 \times 10^{-4}$ cm^{-1}, and $\omega_g = 1''$ yields

$$R_0 / R_{11} = 3.4, \qquad \Gamma_{11} / \Gamma_0 = 2.5;$$

for the total width of the line at half the height we then obtain $\omega_\Sigma = 1.9$.

The calculated and experimental parameters are in good agreement[7)].

CONCLUSION

The described experiments definitely confirm the presence of the quasimosaic effect and the correctness, in general outline, of the proposed theoretical description of the phenomenon. The theory offers satisfactory quantitative predictions of such rather specific properties as the value of the critical angle, the value of the flexure coefficient, and the quadratic character of the bending of the reflecting planes. It describes correctly not only the rather characteristic shape of the diffraction line, but also quantitative parameters such as their absolute width and the ratios of the integral and maximum reflection coefficients.

The elastic quasimosaic effect alters appreciably (by 1–2 orders of magnitude) the characteristics of the diffraction from elastically bent single crystals, and must undisputedly be taken into consideration, in particular, in the design and calculation of focusing diffracting x-ray or gamma spectrometers. An example of an improvement in the properties of these instruments, attainable when the elastic quasimosaic effect is taken into account, is the aforementioned experiment with the 63-keV gamma line of Tm^{169}, wherein replacement of the customarily employed cut ($13\bar{4}0$, $\varphi = 0$; see, for example,[4,6,16,19]) by a special cut ($13\bar{4}0$, $\varphi = 11°$)

[5)]Accurate to negligibly small corrections (at $\omega_g \approx 0.1 \times 2kL$) for the natural mosaic effect and the broadening due to the secondary damping.

[6)]We recall that relations (1) themselves are approximate [6,18].

[7)]If the integral reflection coefficients in the experiment with thin plates (1 mm) are calculated using the K_{α_1} Sn line (see above) with the experimental value $k = 2.4 \times 10^{-4}$ cm^{-1}, then the ratio R_0/R_{11} becomes equal to the experimental value 11 ± 1 at ($\omega_g = 0.4''$. The difference between the parameters ω_g for thin ($\omega_g = 0.4''$) and thick ($\omega_g = 1''$) plate can be due either to the actual difference between the mosaics (the thin and thick plates were prepared from different quartz single crystals), or to the difference in the errors upon orientation (see footnote[2]).

has made it possible to reduce the line width, and to obtain a spectrometer resolution better by a factor 7—8.

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