Anisotropy-induced right–left asymmetry in diffraction by an elastically bent single crystal

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(Submitted 19 January 1978)
Zh. Eksp. Teor. Fiz. **74**, 2280–2285 (June 1978)

The contribution of the elastic anisotropy of crystals [O. I. Sumbaev, Sov. Phys. JETP **5**, 170 (1957); Kristall-difraktsionnye gamma-spektrometry (Crystal Diffraction Gamma Spectrometers), Atomizdat, M., 1963; Sov. Phys. JETP **27**, 724 (1968)] to the right–left asymmetry in the diffraction of x and γ rays by elastically bent absorbing single crystals is considered using the published theoretical and experimental work [P. Penning and D. Polder, Philips Res. Rep. **16**, 419 (1961); N. Kato, J. Phys. Soc. Jpn. **19**, 971 (1964); B. Okkerse and P. Penning, Philips Res. Rep **18**, 82 (1963); L. I. Datsenko, Sov. Phys. Crystallogr. **21**, 447 (1976)]. Contrary to the published work, this contribution does not disappear when the reflecting surfaces coincide with normal transverse cross sections of the investigated crystal plate. It is shown theoretically [in terms of the eikonal theory of Penning and Polder (1961) and Okkerse and Penning (1963)] and experimentally (using the geometry of the Cauchois diffraction spectrometer) that the difference—due to this effect—between the integrated intensities of the diffraction lines to the left and right may be considerable (the intensity on the "strong" side may be several times greater than the intensity on the "weak" side). This effect should be allowed for, in particular, in practical use of diffraction equipment since the symmetry assumed in the usual comparison of the right–left profiles may not apply. The condition for the retention of this symmetry is the additional requirement of zero value of the coefficient responsible for the anisotropic bending of the reflecting planes.

PACS numbers: 78.20.Hp

INTRODUCTION

Penning and Polder[1] and Kato[2] developed a dynamic theory of the diffraction of x rays, γ rays, electrons, or neutrons by weakly deformed perfect crystals: this is known as the ray approximation. The basic features of the theory have been confirmed by experimental investigations (see, for example, the work of Okkerse and Penning[3] and of Datsenko[4]).

One of the interesting effects in this very attractive and interesting theory is the right–left asymmetry effect.[1,2,4] The problem considered in the two fundamental theoretical papers is practically identical with the geometry of the Cauchois focusing diffraction spectrometer (Fig. 1), which is closest to the geometry employed in our investigation. The notation adopted in Fig. 1 is as follows: S is an extended radiation source; C is a plane-parallel single-crystal plate with a family of reflecting planes perpendicular to the plane of the instrument (i.e., to the plane of the figure) but inclined at an angle φ to the right (φ_r in Fig. 1) or to the left (φ_l) relative to the front face. After elastic bending of a crystal to form part of a cylindrical surface with its center at the point O, the continuations of the fan-like reflecting planes intersect at the points R or L, respectively, on the focal circle. The problem is to determine the intensities (integrated values proportional to the areas under the relevant diffraction lines) to the right and left of the direct beam, i.e., in the regions r and l in Fig. 1, for instruments with the R or L geometry. In other words, a comparison is made of the quantities J_{Rr} and J_{Rl}, and of J_{Lr} and J_{Ll}.

In the case of an absorbing crystal we generally have $J_{Rr}\neq J_{Rl}$ and, correspondingly, $J_{Lr}\neq J_{Ll}$, i.e., in the case of initially asymmetric R- and L-type instruments the intensities on the right and left are no longer equal (this is known as violation of the Friedel law). It would seem that it would be natural for the following relationships to be obeyed:

$$J_{Rr}=J_{Ll},\quad J_{Rl}=J_{Lr}$$

and, in particular, if

$$\varphi=0, \tag{1}$$

then

$$J_{Or}=J_{Ol}. \tag{2}$$

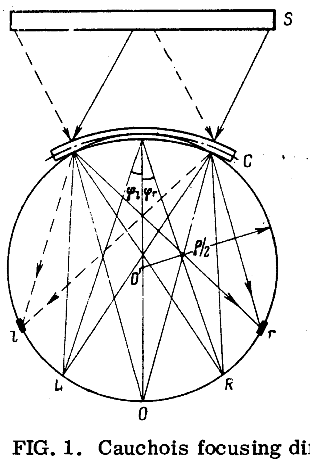

FIG. 1. Cauchois focusing diffraction spectrometer whose right–left asymmetry is investigated. Here, S is the radiation source, C is a single-crystal plate bent to form a cylindrical surface with its center at the point O and radius ρ. The focusing occurs on a focal circle whose center is O' and radius is $\rho/2$; O is the point of intersection of continuations of the reflecting planes coinciding with the normal transverse cross sections ($\varphi=0$ case); L and R are the corresponding intersection points for $\varphi=\varphi_l$ or φ_r; l and r are the regions of focusing of the diffracted radiation.

The conclusion (2) that in the case of an initially symmetric instrument (an instrument with $\varphi = 0$, where the reflecting planes intersect at the point O) the right and left reflections are identical is essentially reached by the authors of the two fundamental papers (see also Okkerse and Penning[3]).

Our aim is to shown that the equality (2) may not be satisfied, i.e., that the condition (1) is insufficient.

THEORY AND EXPERIMENT

The terms which do not disappear in the limit $\varphi \to 0$ and are due to anisotropy are omitted from the relationships for the shape of the reflecting planes of an elastically bent single crystal [Eq. (34) given by Penning and Polder[1] and Eq. (24a) given by Kato[2]]. These terms are well known in the theory of elasticity[5] and have often been considered in connection with the process of diffraction by bent single crystals[6-8]; they are responsible for the bending of normal transverse cross sections (Fig. 2). The shape of the reflecting planes is then described by the following relationship[6] which applies in the $\varphi = 0$ case:

$$z = ky^2, \tag{3}$$

where

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

a_{ij} are the components of the elasticity tensor of the crystal in question, and ρ is the radius of the cylindrical surface to which the crystal plate is bent.

We can easily show [see Eq. (3) in Kato's paper[2]] that the expression for the effective Kato force now becomes

$$f = \frac{2\pi}{d} \left(\frac{\cos\varphi \sin 2\varphi}{\rho \sin 2\vartheta} + \frac{\cos\vartheta}{\sin\vartheta} k \right); \tag{4}$$

here, ϑ is the Bragg angle and d is the distance between the reflecting planes. Thus, we have for $\varphi = 0$ an additional nonvanishing term which is proportional to the anisotropic bending coefficient k.

Since the force is constant (it is independent of the coordinates y and z of the problem), we can apply directly Kato's solution (27).[2] The ratio of the integrated intensities for the right and left positions of the instrument should be written in the form

$$\eta = \left(\int_{-1}^{1} I_g^{(1)} e^{2S^i} dt + \int_{-1}^{1} I_g^{(2)} e^{-2S^i} dt \right) \left(\int_{-1}^{1} I_g^{(1)} e^{-2S^i} dt + \int_{-1}^{1} I_g^{(2)} e^{2S^i} dt \right)^{-1}, \tag{5}$$

where $I_g^{(1)(2)}$ are the intensities of the essential and inessential modes [see Eq. (44b) in Kato's paper[2]], and S^i is the imaginary eikonal [Eq. (40a)]. Integration is carried out over the exit face of the crystal $y = T/2$ (where T is the thickness of the crystal plate) within the Borrmann delta, where

$$-1 \leqslant t = z/T \operatorname{tg}\vartheta \leqslant 1.$$

The quantity η depends on the parameters[1)]

$$\alpha = \frac{\cos\vartheta}{\cos\pi^*} \frac{\pi}{\lambda} \frac{V}{r_e d F_{gr}} kT \tag{6}$$

and

$$P = \frac{\cos\pi^*}{\cos\vartheta} \frac{\mu\epsilon T}{\alpha}, \tag{7}$$

where λ is the wavelength of the radiation, r_e is the classical radius of an electron, F_{gr} is the real structure factor of the reflecting planes,[2)] V is the volume of a unit cell in the crystal, μ is the linear absorption coefficient of the radiation in the crystal, ϵ is the ratio of the imaginary structure factors (imaginary scattering amplitudes at the Bragg angle and in the forward direction), and $\cos\pi^*$ is the factor depending on the polarization and equal to either unity or $\cos 2\vartheta$.

The calculations of η for the general case were carried out on a computer and the results (η_{theor}) are listed in Table I. For $P = 1$ and $P = 2$ it was possible to obtain exact analytic expressions which were then used to check the computer calculations (see Table I). The first of these analytic expressions was very simple:

$$\eta(\alpha, 1) = 1 + 2\alpha^2. \tag{8}$$

It seemed important to carry out a control experiment completely identical to the one carried out in the first basic studies, except that because of a special orientation of a cut plate (see below) the same reflecting planes (coinciding, as before, with the normal

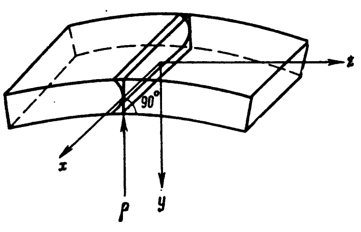

FIG. 2. Bending of normal transverse cross sections in an elastically bent anisotropic crystal plate; ρ is the radius of the cylindrical surface to which the plate is bent. The normal transverse cross sections are bent to form parabolic surfaces described by $z = ky^2$; the case illustrated in this figure corresponds to $k < 0$.

TABLE I.

Line and its energy, keV	μ, cm^{-1}	α	P	η_{theor}	η_{exp}
		Main experiment $k = (0.88 \pm 0.04) \times 10^{-4}$ cm^{-1}			
K_{α_1}Xe; 29.8	2.35	3.37	0.078	1.29	1.20±0.05
K_{α_1}Cd; 23.2	4.50	2.64	0.190	1.70	1.52±0.05
K_{α_1}Mo; 17.5	9.82	1.99	0.550	3.43	2.81±0.05
K_{α_1}Yb; 52.4*	–	–	–	≡1	1.04±0.05
K_{α_1}Rb; 13.4	20.8	1.52	1.525	12.4	9.2±0.2
K_{α_1}Sm; 40.1*	–	–	–	≡1	0.94±0.05
		Control experiment $k < 1.5 \times 10^{-7}$ cm^{-1}			
K_{α_1}Mo; 17.5	9.82	0.036	30.3	0.93÷1,07	0.91±0.05
K_{α_1}Yb: 52.4*	–	–	–	≡1	0.91±0.05
		Control calculation			
14.9**	15.3	1.70	1	6.78 (anal)	6.80 (comp)
12.5**	25.5	1.42	2	18.18 (anal)	18.18 (comp)

*These lines were used, in the third reflection order, as reference points for the $K_{\alpha 1}$ lines of Mo and Rb, respectively.
**The energies were determined from the $\mu(\lambda)$ curve to ensure the necessary values of $\mu(\lambda)$ for $p = 1$ and 2, respectively.

transverse cross sections) now had a bending coefficient k as close as possible to zero. It was then very easy to obtain a simple expression for η for the small values of α corresponding to this case; this was done using the well-satisfied approximate (with just the first term of the expansion) relationship [Eq. (40b) in Kato's paper] for the imaginary eikonal:

$$\eta \approx \frac{I_0(\alpha P)+2\alpha I_1(\alpha P)}{I_0(\alpha P)-2\alpha I_1(\alpha P)}, \tag{9}$$

where I_0 and I_1 are the Bessel functions.

Table I gives the values of the right–left asymmetry parameter η for the x rays used in our experiments, as well as the parameters α and P and the values of μ used by us. The experiments were carried out in the geometry of Fig. 1 on a crystal bent to a radius of $\rho = 200$ cm. We used natural quartz plates $T = 0.16$ cm thick cut so that the $01\bar{1}1$ reflecting planes coincided with the normal transverse cross sections. More specifically, the error in the orientation was such that $\varphi \leqslant 1'$ (within one angular minute). The plate in a control experiment was cut so that the large faces coincided with the $2\bar{1}\bar{1}0$ planes. In this case the bending coefficient was $k \approx 0$. (The small orientation error $\varphi < 1'$ ensured that $k < 1.5 \times 10^{-7}$ cm^{-1}.) The plate used in the main experiment differed by rotation through 11° about the longitudinal (normal to the reflecting planes) axis (the sense of the direction was unimportant), which ensured[3)] the value $k = (0.88 \pm 0.04) \times 10^{-4}$ cm^{-1} for the following parameters:

$$d=3.34\ \text{Å},\quad V=112\ \text{Å}^3,\quad F=37.5,\quad \varepsilon=0.695.$$

In the control experiment the effective force [see Eq. (4)] had terms due to the maximum deflections possible in the preparation of the plate. Using the above limiting values of φ and k, we found that the maximum force in the control experiment was

$$f < \frac{2\pi}{\lambda}(2.9\cdot 10^{-6}+3\cdot 10^{-7})\ \text{cm}^{-2}.$$

Thus, the main contribution was due to a possible projection bending. The value of η_{theor} calculated from Eq. (9) was also included in Table I. Since the sign of the deviation of φ from zero was unknown, the following range of the permissible values was established for η:

$$1/\eta_{max} < \eta < \eta_{max}.$$

The influence of possible inaccuracies of the alignment of the instrument or instability of the x-ray system during the main measurements was allowed for by the use of reference standards in the form of "harder" $K_{\alpha 1}$ x-ray lines of Yb and Sm, whose third-diffraction-order positions were very close to $K_{\alpha 1}$ of Mo and Rb, respectively. We readily established that the reference lines corresponded to a mosaic crystal (range corresponding to the random phase approximation) and that symmetric conditions were definitely expected ($\eta \equiv 1$).

DISCUSSION OF RESULTS AND CONCLUSIONS

The results (Table I) demonstrated that even at the "instrumental zero" (case $\varphi = 0$ in Fig. 1) there was a pronounced right–left asymmetry (the ratio of the integrated line intensities could reach ten) and the experimental values were in satisfactory agreement with the theory.[4)]

The physical factor responsible for the asymmetry is self-evident: clearly, a bent anisotropic crystal may be right- or left-handed even for $\varphi = 0$. A true symmetric situation is obtained on coincidence of the normal reflecting planes with one of the planes of the elastic symmetry of the crystal. We can easily see that this does take place[5)] since then the a_{34} and a_{35} components of the elasticity tensor of the crystal vanish identically and [see Eq. (3)] this is true also of the bending coefficient k of the planes.

The effect is observed in spite of a slight curvature of the crystal "space" (the bending radius of the reflecting planes in our main experiment was $\rho_g = 1/2k = 57$ m), which was a consequence of the high sensitivity of the asymmetry to small deformations, as pointed out earlier.[1,9]

The effect should be allowed for in the practical use of focusing diffraction instruments because in the frequently used method of comparison of the left–right profiles any difference between these profiles is attributed to aberrations (i.e., to poor alignment of the instrument). This method can, strictly speaking, be applied correctly only for a truly symmetric instrument and the symmetry condition is not just $\varphi = 0$ but also $k = 0$.

1)The parameter $2\alpha \equiv Z$ in Kato's paper[2]; see Eq. (30) in that paper for $z = T$.
2)We shall consider a centrosymmetric crystal when $F_g = F_{\bar{g}}$.
3)For details see Sumbaev's work.[7,8]
4)The reason for the slight (10–30%) but systematic excess of the calculated over the experimental values of η is not clear. It may be due to the inaccuracy of the coefficient k (the likely error is ≲5%) or due to the values μ and μ_a used by us (the latter were employed in the calculation of ϵ). We cannot exclude the possibility of the influence of the regions of intergration near $|t| = 1$ in Eq. (5), where—strictly speaking—the eikonal approximation is invalid.
5)For a homogeneous isotropic medium any plane is a plane of elastic symmetry and, therefore, the effect considered here is never exhibited by such a medium.

1P. Penning and D. Polder, Philips Res. Rep. 16, 419 (1961).
2N. Kato, J. Phys. Soc. Jpn. 19, 971 (1964).
3B. Okkerse and P. Penning, Philips Res. Rep. 18, 82 (1963).
4L. I. Datsenko, Kristallografiya 21, 788 (1976) [Sov. Phys. Crystallogr. 21, 447 (1976)].
5S. G. Lekhnitskiĭ, Teoriya uprugosti anizotropnogo tela (Theory of Elasticity of Anisotropic Bodies), Gostekhizdat, M., 1950.
6O. I. Sumbaev, Zh. Eksp. Teor. Fiz. 32, 247 (1957) [Sov. Phys. JETP 5, 170 (1957)].
7O. I. Sumbaev, Kristall-difraktsionnye gamma-spektrometry (Crystal Diffraction Gamma Spectrometers), Atomizdat, M., 1963.
8O. I. Sumbaev, Zh. Eksp. Teor. Fiz. 54, 1352 (1968) [Sov. Phys. JETP 27, 724 (1968)].
9R. Bubáková and Z. Trousil, Z. Naturforsch. Teil A 28, 1199 (1973).

Translated by A. Tybulewicz