

Quartz Reflectivity and Piezoelectricity*

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Abstract

It has been observed that the reflectivity of the (110) planes of a quartz lamina can be increased by submitting it to a static or alternating electric field. Measurements have been performed on 2.5 and 6 mm thick plates using a curved-crystal spectrometer with DuMond geometry. Photon energies were in the range 50 to 300 keV. Under the proper conditions, the increase in reflectivity is accompanied by negligible loss in resolution. The effects are explained by the occurrence of a quasi-mosaicity.

1. Introduction

A drawback of crystal spectrometers is their low luminosity, which can be improved by several means. The easiest would be to use large crystals. Unfortunately, these are very expensive and the crystal orientation is generally not sufficiently constant over the entire area. Another way is to juxtapose several crystals whose relative orientation can be adjusted (Beer, Dousse, Kern, Piller & Schwitz, 1976). In both cases we have to contend with increased geometrical aberrations (Schwitz, Kern & Lanners, 1978). Yet another solution is to use highly reflecting crystals. The problem then is to obtain a crystal with good resolution. It is known that germanium and silicon crystals have good reflectivities. However, large specimens of sufficient quality are seldom available. In contrast, it is possible to obtain large quartz plates of good quality. Unfortunately, the reflectivity of quartz is not large under normal circumstances.

In this paper techniques are described by which the reflectivity of quartz can be markedly increased. Under the proper conditions the line broadening is very small, as noted in a preliminary account of our results (Dousse & Kern, 1976).

Measurements were performed with a DuMond curved-crystal spectrometer (DuMond, 1947) of the transmission type entailing Laue diffraction. The bending radius was 5 m and the bending blocks had an aperture of 50×50 mm.

The reflectivity enhancement is obtained by applying a static or an alternating electric field across the plate:

(i) With an alternating field, the frequency can be tuned to a resonant frequency of the crystal. The required voltage is then only a few volts. The phenomenon, which we call the AC effect, takes place instantaneously.

(ii) With a static field, the voltage has to reach a few kilovolts. The reflectivity increases only slowly with time. We have given the name DC effect to this new phenomenon.

Fox & Carr (1931) first observed the change in intensity of X-rays diffracted from piezoelectrically vibrated quartz plates. Considerable interest has since been exhibited in this phenomenon. A comprehensive review of the subject is given by Spencer (1968). The change of the diffracted intensity is a dynamical effect and relies essentially on primary-extinction variations. In particular, this phenomenon has been used for studying the vibrating modes of crystal lamina by X-ray topography. An extensive list of references on this particular aspect can be found in Spencer (1968). As is shown in this paper (§2.3), the overall reflectivity of a plate can also be enhanced, the increase depending on the diffracted photon energy and on the reflection order. It is stressed, however, that in our application the peak reflectivity and not the integrated reflectivity is important since the diffraction lines must have narrow widths. These characteristics are considered in §2.4.

Thermal-neutron diffraction using vibrating quartz crystals was also studied. Large enhancements in diffracted intensities were obtained (Kenney, Jefferies & Jacobs, 1974).

On the other hand, the DC effect is new. As far as we know, one group only has recently and independently observed a similar effect on X-rays (Lapin, Samsonov, Sumbayev & Tunis, 1976).

2. The AC effect

2.1. Preliminary remarks

Preliminary experiments on the AC effect have shown that the fundamental frequency was

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unfavourable since the increase in peak reflectivity was accompanied by a marked loss in resolution. The use of 5th, 7th and 9th harmonics, on the other hand, gave very encouraging results, as can be seen in Fig. 1, which shows an example where the diffracted intensity is more than doubled while the widening is only about 17%. It was also observed that for a given photon energy and order of diffraction, the integrated reflectivity R is a function of the applied voltage U . There is an optimum for which the peak reflectivity Γ is largest. The variation of the peak width ω (FWHM) and of R and Γ as a function of U is shown in Fig. 2.

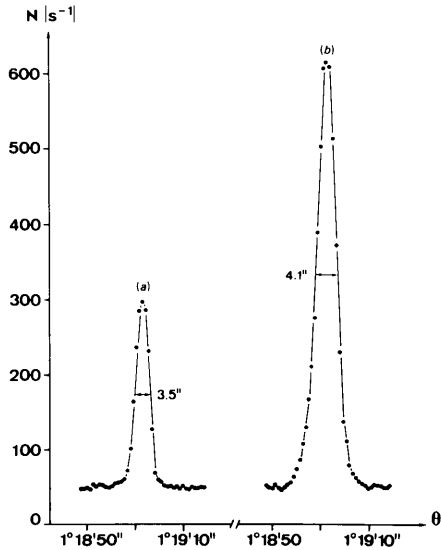


Fig. 1. Reflection line obtained from 109.78 keV photons diffracted in first order by the (110) planes of a 2.5 mm thick quartz lamina: (a) without electric field; (b) with the AC effect ($f_7 = 5452.5$ kHz, $U = 2$ V).

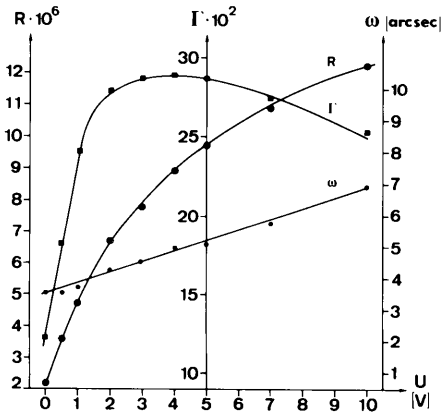


Fig. 2. Increase in integrated reflectivity R , peak reflectivity Γ and FWHM ω of a reflection line as a function of the applied voltage U . The data were collected with the 2.5 mm plate and 109.78 keV photons in first order.

2.2. Determination of the resonant frequencies

For (110) quartz plates (Fig. 3a), an alternating electric field produces thickness shear oscillations (see also §3.2). The eigenfrequencies are given by Mason (1943)

$$\Omega_l = 2\pi l / (2s)(c_{66}/\rho)^{1/2}, \quad (1)$$

where c_{66} is a component of the elasticity tensor (matrix notation), ρ the crystal density and s the plate thickness. Only odd harmonics are possible ($l = 1, 3, \dots$).

Formula (1) is valid for a crystal plate of infinite extension in the directions x_1 and x_3 . In reality, the finite width b of the plate also plays a role, as shown by the following formula (Ekstein, 1945)

$$\Omega_{l,m} = \pi(c_{66}/\rho)^{1/2}(l^2/s^2 + k_l^2 m^2/b^2)^{1/2}, \quad (2)$$

where k_l is a constant depending on the harmonic l and on the elastic constants, and $m = 1, 2, \dots$

We have reported in Fig. 4 (bottom curve) the voltage measured across the plate as a function of frequency for the 2.5 mm thick plate and in a frequency range close to the 7th harmonic. Important fluctuations are apparent and the function jumps abruptly from a minimum value (series resonance) to a maximum value (parallel resonance). With this procedure, it is possible to determine experimentally the eigenfrequencies. The counting rate at the peak of a Bragg reflex is then observed as a function of frequency. At resonance a maximum is registered (Fig. 4, top curve). Comparison of the upper and lower curves in Fig. 4 shows that the reflectivity is a maximum for frequencies intermediate between series and parallel resonances. It is interesting to note that the AC effect is more sensitive than electric measurements, and displays minor resonances not predicted by formula (2). These may arise from the finite height of the plate which has been neglected in

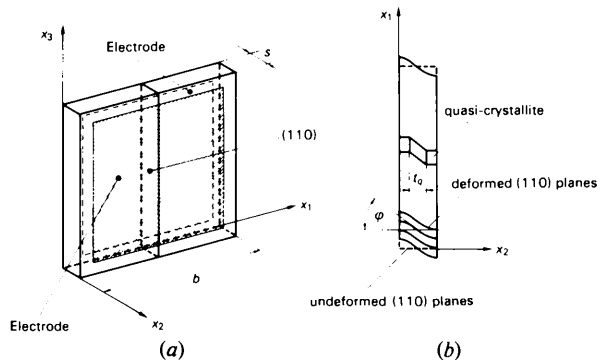


Fig. 3. (a) Orientation of the crystal plate and of the (110) planes with respect to the piezoelectric axes (x_1, x_2, x_3). The optical axis is parallel to axis x_3 . (b) Deformation of the (110) planes by shearing stress. Definition of the quasi-crystallites.

Ekstein's calculations. Other authors (Sykes, 1944) take all three dimensions of the plate into account and predict resonance frequencies $\Omega_{l,m,n}$. For further discussion see Spencer (1968, and references therein).

2.3. Increase in integrated reflectivity and AC quasi-mosaicity

The shearing oscillations in the plate cause some distortion of diffracting planes. At resonance frequency, the deformation becomes large enough to decouple the waves in the crystal: the extinction effects are reduced and the diffracted intensity increases.

To take this misalignment into account, let us suppose that the plate can be divided into slices of thickness t_q representing the distance over which the orientation of the diffracting planes varies by an angle equal to the Darwin breadth ω_D . The expression for ω_D is given by (James, 1967)

$$\omega_D = (2/\pi)FNr_0(\lambda/n)d_{hkl} \cos \theta_B, \quad (3)$$

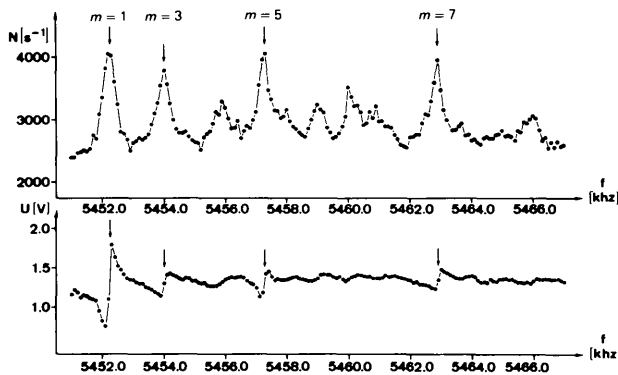


Fig. 4. Variation of the counting rate at the top of a reflection (109.78 keV, first order of diffraction) and of the voltage across the plate as a function of frequency near the seventh harmonic.

where F is the structure factor, N the number of elementary cells per unit volume, r_0 the classical electron radius, λ the wavelength of the diffracted photons, n the diffraction order, θ_B the Bragg angle and d_{hkl} the interplanar spacing.

In other words, one can say that the AC effect induces quasi-mosaicity in the crystal, t_q in this representation being the average dimension of the above-mentioned slices which we refer to as quasi-crystallites (Fig. 3b).

To determine t_q , we calculate the average value $\overline{\Delta x_2}$ for which the disorientation amounts to ω_D . After some calculation we obtain

$$t_q(t) = s/(2l)\omega_D/|\varphi(t)|, \quad (4)$$

where $\varphi(t) = \varphi_0 \sin(\Omega_l t)$, φ_0 being proportional to the deformation amplitude in the direction x_1 normal to the diffraction planes. In order to reproduce the integrated reflectivities observed with the vibrating crystal, we have used the following formula, similar to that given for a flat mosaic crystal (James, 1967),

$$R_{AC} = \kappa Q \bar{f}(A_q) s \exp[-\bar{g} \kappa Q \bar{f}(A_q) s], \quad (5)$$

where κ is an adjustable parameter ($\kappa < 1$) taking into account the fact that crystallite diffraction occurs over an effective angular range wider than ω_D and where Q is defined as

$$Q = (FNr_0)^2 \lambda^3 / \sin 2\theta_B. \quad (5a)$$

$\bar{f}(A_q)$ which represents the mean primary extinction is given by

$$\bar{f}(A_q) = 1/T_l \int_0^{T_l} f[A_q(t)] dt, \quad (6)$$

$$A_q(t) = FNr_0 \lambda t_q(t). \quad (6a)$$

$T_l = 2\pi/\Omega_l$, where Ω_l is given by (1). f is a complicated function which can be found, for example, in Zachariasen (1946).

Table 1. Comparison between the observed integrated reflectivities of the 2.5 mm thick plate in resonance ($f_7 = 5452.5$ kHz), and the theoretical values computed with the quasi-mosaic model (equation 10)

The following parameters have been used:

$$\begin{aligned} U = 2 \text{ V: } & \varphi_0 = 0.7 \text{ arc sec, } \bar{g} = 5.05 \times 10^4, \kappa = 0.75 \\ U = 5 \text{ V: } & \varphi_0 = 1.8 \text{ arc sec, } \bar{g} = 2.80 \times 10^4, \kappa = 0.80 \\ U = 10 \text{ V: } & \varphi_0 = 2.7 \text{ arc sec, } \bar{g} = 1.70 \times 10^4, \kappa = 0.82. \end{aligned}$$

E (keV)	U = 2 V		U = 5 V		U = 10 V	
	$R_{obs} \times 10^6$	$R_{AC} \times 10^6$	$R_{obs} \times 10^6$	$R_{AC} \times 10^6$	$R_{obs} \times 10^6$	$R_{AC} \times 10^6$
63.12	7.21 ± 0.30	7.24	12.75 ± 0.64	13.12	19.69 ± 0.91	20.47
93.62	7.05 ± 0.80	7.01	11.48 ± 0.70	11.04	14.13 ± 0.80	14.25
109.77	6.61 ± 0.25	6.54	9.88 ± 0.40	9.54	12.15 ± 0.50	11.48
130.51	5.48 ± 0.23	5.60	7.31 ± 0.33	7.66	9.19 ± 0.37	9.01
177.18	3.76 ± 0.16	3.87	4.74 ± 0.19	4.76	5.35 ± 0.22	5.35
197.97	3.29 ± 0.15	3.28	4.02 ± 0.16	4.06	4.49 ± 0.18	4.38
261.05	1.96 ± 0.11	2.14	2.41 ± 0.12	2.48	2.57 ± 0.12	2.62
307.68	1.60 ± 0.07	1.62	1.73 ± 0.09	1.84	1.93 ± 0.09	1.92

Table 2. Comparison between experimental and theoretical integrated reflectivities of the 6 mm quartz plate in resonance ($f_7 = 2272.0$ kHz, $U = 6$ V)

The fitted parameters used in formula (10) are:

$$n = 1: \varphi_0 = 1.7 \text{ arc sec}, \bar{g} = 2.80 \times 10^4, \kappa = 0.75$$

$$n = 2: \varphi_0 = 1.7 \text{ arc sec}, \bar{g} = 3.23 \times 10^4, \kappa = 0.80.$$

E (keV)	n = 1		n = 2	
	$R_{\text{obs}} \times 10^6$	$R_{\text{AC}} \times 10^6$	$R_{\text{obs}} \times 10^6$	$R_{\text{AC}} \times 10^6$
63.12	11.35 ± 0.52	11.51	10.22 ± 0.42	10.25
109.77	13.46 ± 0.55	12.89	10.72 ± 0.45	10.41
130.51	11.59 ± 0.49	11.81	8.97 ± 0.38	9.02
177.18	8.42 ± 0.35	8.78	6.25 ± 0.26	6.21
197.97	7.59 ± 0.31	7.67	5.17 ± 0.21	5.28
307.68	4.02 ± 0.18	3.88	2.65 ± 0.12	2.52

As the crystal oscillates at higher harmonics ($l > 1$), the deformation across the plate is periodic so that there is a significant probability for the photon to be diffracted by parallel quasi-crystallites. We therefore assume that the secondary extinction can be described by the expression $\exp(-gQ's)$ (Darwin, 1922), where $Q' = \kappa Q f(A_q)$. Since the secondary-extinction parameter is now time dependent, we use its mean value \bar{g} .

The observed integrated reflectivities have been fitted by adjustment of the parameters φ_0 , \bar{g} and κ . This fit was performed for the 2.5 mm thick plate in the first order of diffraction for three different voltages U . For the 6 mm thick plate, we have performed the measurements with one value of U at two diffraction orders. In all cases the frequency corresponding to the 7th harmonic with $m = 1$ was used. The observed and calculated reflectivities shown in Tables 1 and 2 are in good agreement, the difference between calculated and observed values being in general smaller than the experimental uncertainty. It can be noted that \bar{g} decreases for increasing values of φ_0 while κ is approximately constant. The deformation φ_0 increases almost linearly with U .

2.4. Increase in peak reflectivity and reflection broadening

The increase in peak reflectivity is a function of the diffracted photon energy and of the diffraction order. The angular width of the source may also play a role as shown later in this section. In Fig. 5 are presented the values of Γ/Γ_0 observed with the 2.5 mm thick crystal for several energies and for the first three orders of diffraction. Γ_0 is the peak reflectivity for the crystal at rest ($U = 0$) and Γ is the peak reflectivity at resonance conditions using a voltage U such that the line broadening remains less than 5%.

Γ is larger than Γ_0 for the following reasons:

(a) Integrated reflectivity measurements on the two quartz lamina under normal conditions, *i.e.* without application of any electric field, have shown that the

observed values can be reproduced by the mosaic-crystal model including primary and secondary extinctions (Dousse & Kern, 1980). From this study we deduced that the mean crystallite size is more than large enough to ensure equilibrium diffraction conditions. However, the crystallite-orientation distribution is locally narrower than the apparent angular source, especially if one takes into account the focal aberration due to imperfect crystal curvature. Therefore the peak reflectivity Γ_0 remains smaller than the limit value of 50%. At 109.78 keV, for example, in the first order of diffraction, it is only about 12% for the 2.5 mm thick plate.

(b) For small applied voltages, the induced deformation φ_0 is small and the quasi-crystallite size is still large enough for equilibrium diffraction conditions. Their distribution is, however, wider than originally and therefore better adapted to the source width. This produces an increase in peak reflectivity.

(c) When φ_0 increases, Γ also increases until the quasi-crystallite size becomes too small for equilibrium diffraction conditions. For large values of U , the quantity A_q defined by (6a) will be much smaller than unity. Under these circumstances the peak reflectivity can be described by (Knowles, 1959)

$$\Gamma \simeq (1/2)\{1 - \exp[-W(0)R]\}, \quad (7)$$

where R is the integrated reflectivity and $W(0)$ the angular distribution of the quasi-crystallites for $\theta = \theta_B$. The variation $\Delta\Gamma$ can be deduced from (7)

$$\Delta\Gamma = (1/2) \exp[-W(0)R] [W(0)\Delta R + R\Delta W(0)]. \quad (8)$$

Since the integrated reflectivity R increases with the applied voltage and tends toward an asymptotic value, the variation ΔR tends toward zero but remains positive. We will see that $W(0)$ is a decreasing function of the deformation φ_0 (Fig. 6), so that $\Delta W(0)$ is negative. Therefore, there exists a value of U for which $\Delta\Gamma$ vanishes. Under this condition $\Gamma = f(U)$ reaches a maximum, as observed experimentally (see Fig. 2).

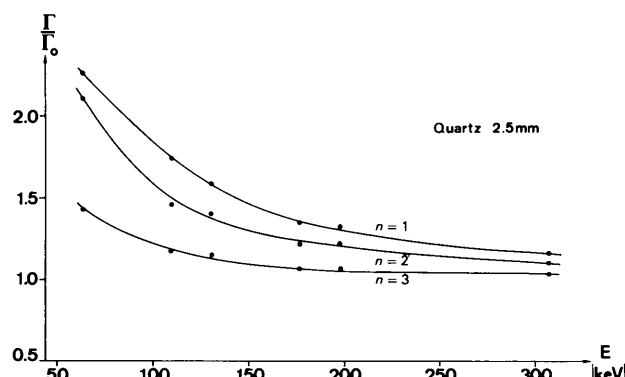


Fig. 5. Variation of the ratio Γ/Γ_0 as a function of the diffracted photon energy and of the diffraction order. The data have been taken with the 2.5 mm thick plate using the seventh harmonic. The line widening is negligible.

We have computed the angular quasi-crystallite distribution $W(\varphi)$ for different values of φ_0 with the assumption that $w(\varphi)$, the distribution at zero field, can be represented by a Gaussian distribution having a FWHM equal to 3 arc sec. The results are presented in Fig. 6, where the FWHM of $W(\varphi)$ and the ratio $W(0)/w(0)$ are plotted as functions of φ_0 . We have found experimentally (Fig. 2) that ω does not increase appreciably for small voltages of the order of one volt. This feature is reproduced by the calculation (Fig. 6). For quantitative comparison, φ_0 must be calibrated as a function of U . As shown in Table 1, a voltage $U = 5$

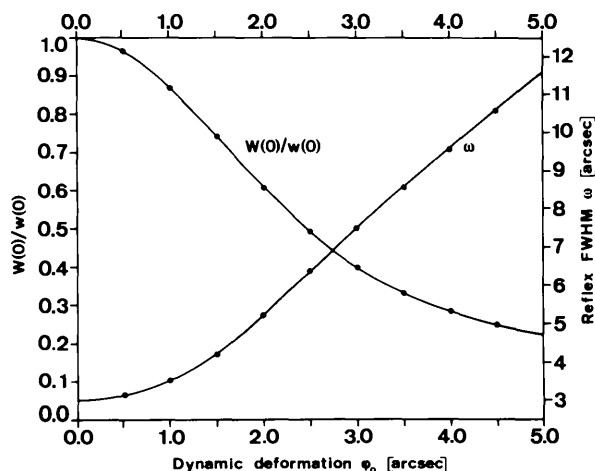


Fig. 6. Effect of the deformation φ_0 on the FWHM ω of the distribution $W(\varphi)$ and on its normalized height $W(0)/w(0)$. The dots represented calculated values. It has been assumed that the intrinsic mosaicity of the crystal can be represented by a Gaussian distribution having a FWHM of 3 arc sec.

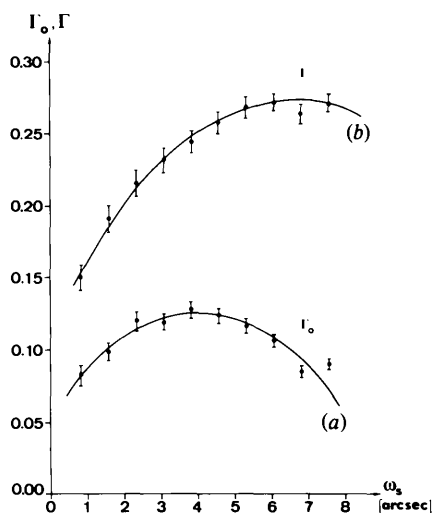


Fig. 7. Effect of the apparent angular source width ω_s on the crystal peak reflectivity: (a) in normal conditions (Γ_0); (b) with the AC effect (Γ). The measurements have been performed with 67.8 keV photons diffracted in first order by the 6 mm thick crystal.

volts produces a deformation $\varphi_0 = 1.8$ arc sec. We calculate that the corresponding peak widening is 1.5 arc sec (Fig. 6), in good agreement with observed value of 1.6 arc sec (Fig. 2).

In the above results the source width has not been taken into account. To illustrate the effect of this parameter, we have observed the peak reflectivities Γ and Γ_0 as a function of the apparent source angular width ω_s (Fig. 7). One can deduce from this figure that the ratio Γ/Γ_0 is approximately 1.9 for source widths smaller or equal to 3.5 arc sec, which represents the effective line width contributed by the crystal and the bending blocks. For larger values of ω_s this ratio increases. It is, for instance, 3 for $\omega_s = 7$ arc sec. This phenomenon has the following explanation. The peak reflectivity is largest when the line width of the crystal matches the apparent source width (see Fig. 7, bottom curve). Because of the wider angular distribution induced by the AC effect, the optimal conditions can be obtained even if the source width surpasses the crystal resolution at rest. In other words, the peak reflectivity Γ in the AC mode can retain its maximum value even when the source thickness becomes large. Since the reflectivity Γ_0 of the crystal at rest decreases under these conditions, the ratio Γ/Γ_0 becomes larger.

2.5. Final remarks

Our study of the AC effect on two quartz plates can be summarized as follows:

- The effect is instantaneous. The increase of R and Γ occurs as soon as the crystal is excited by the electric field. There is no remanence.
- The crystal is not damaged by the oscillations.
- The effect only occurs if the deformation relating to the x_1 direction (see Fig. 3) does not vanish. For a particular plate orientation, application of the piezoelectric equations (see §3.2) will verify if this condition is fulfilled.
- The centre of gravity of the diffraction line is not shifted because the deformation alternates in sign.
- The increase in integrated reflectivity depends on the photon energy and on the order of diffraction. For

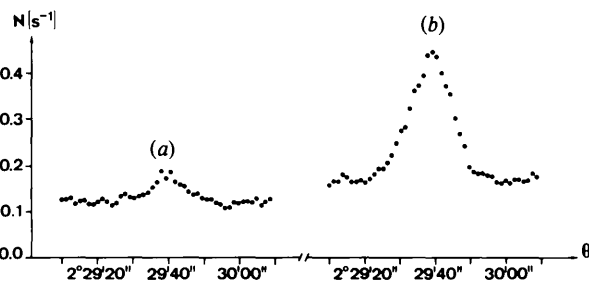


Fig. 8. Reflection observed with 57.98 keV X-rays diffracted in first order by the 6 mm thick plate: (a) without electric field (normal conditions); (b) with the AC effect ($f_7 = 2272.0$ kHz, $U = 15$ V).

the first and second orders, and for energies below 100 keV, this increase may be quite significant. The AC effect is useful whenever a slight loss in resolution can be accepted. This would apply for the first crystal of a double flat crystal spectrometer.

– The increase in peak reflectivity also depends on the energy and order of diffraction, along with the source thickness. For a thin source, at low energy and of first order, the height of a peak can be more than doubled without appreciable loss in resolution, even for FWHM's as small as 3 arc sec. For thick sources, or in the observation of X-rays with a sizeable natural width, the peak reflectivity may be multiplied by a factor of five or more. Fig. 8 illustrates the power of the method. Curve (a) shows a weak peak nearly submerged in background fluctuations. Its intensity is strongly enhanced by the AC effect (curve b) so that the peak clearly emerges.

3. The DC effect

3.1. Preliminary remarks

Under a direct high voltage, a rotation of the diffraction planes is observed. The peaks are shifted by an angle proportional to the applied voltage (see §3.2). When high tension is maintained, it is observed that after some time the reflectivity begins to increase until it reaches a maximum value which depends on the voltage, the energy of the diffracted photons, and the order of diffraction. When the voltage is switched off, the reflectivity decreases very slowly, approaching its original (normal) value asymptotically (see Fig. 9).

If the high voltage does not exceed a critical value (about 6000 volts for our 2.5 mm crystal) and if the

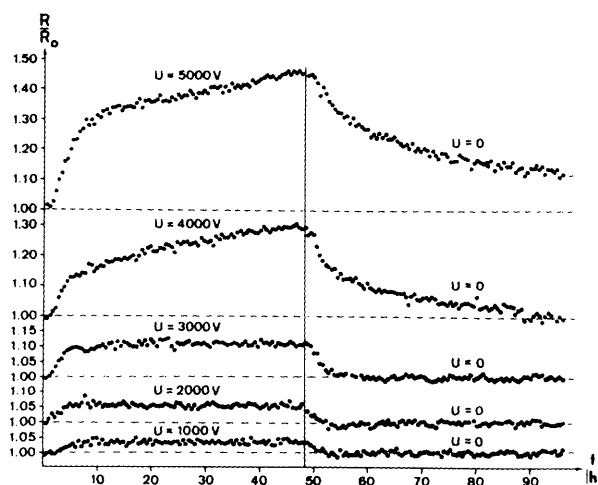


Fig. 9. Relative increase in integrated reflectivity of the (110) planes of a 2.5 mm thick quartz plate as a function of time and of the applied voltage. The measurements have been performed with 197.97 keV photons diffracted in first order.

experiment does not last too long, the width of the lines remains unchanged. The explanation for the effect is similar to that for the AC effect. In this case, we also have an appearance of quasi-mosaicity. However, its origin is quite different. The high voltage induces a polarization in the crystal and an electric counter field builds up. This field is not homogeneous but its strength depends on the coordinate x_2 . The angle of rotation of the diffraction planes is then a function of this same coordinate, causing a deformation of the planes. The consequences are similar to that observed with the AC effect. An important difference is that the deformation is nearly time independent, and not fluctuating rapidly. This leads to an even greater increase in peak reflectivity.

3.2. Rotation of the diffraction planes

The rotation of the diffraction planes is due to the reciprocal piezoelectric effect, given in matrix notation by

$$\epsilon_n = \sum_{m=1}^3 d_{mn} E_m \quad (9)$$

where E_m is the m component of the electric field \mathbf{E} and d_{mn} a component of the piezoelectric matrix. In the coordinate system defined by Fig. 3(a), the piezoelectric coefficients of quartz have the following values at 293 K (Kleber, Meyer & Schönborn, 1968):

$$|d_{mn}| = \begin{pmatrix} -2.3 & 2.3 & 0. & 0.85 & 0. & 0. \\ 0. & 0. & 0. & 0. & -0.85 & 4.6 \\ 0. & 0. & 0. & 0. & 0. & 0. \end{pmatrix} \times 10^{-12} \text{ m V}^{-1}. \quad (10)$$

The angular rotation of the planes (110) is given by

$$\Delta\theta = \epsilon_6 = d_{26} E_2. \quad (11)$$

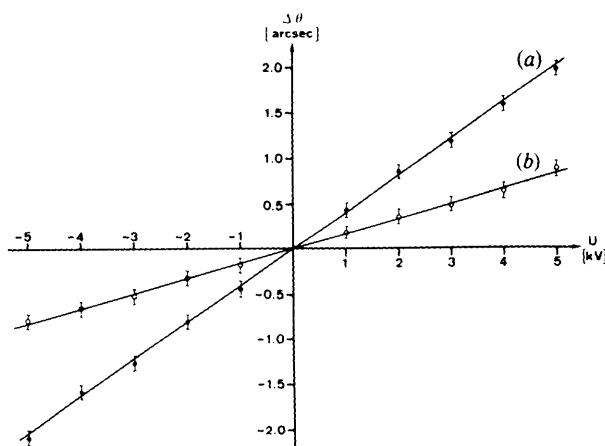


Fig. 10. Angular rotation $\Delta\theta$ of the (110) planes as a function of the high voltage on the electrodes of (a) a 2.5 mm, and (b) a 6 mm thick quartz plate.

Relation (11) predicts a rotation of 0.38 arc sec/1000 V for a 2.5 mm thick plate and of 0.16 arc sec/1000 V for a 6 mm thick plate. These values are in good agreement with the observed slopes of 0.40 and 0.17 arc sec/1000 V respectively (Fig. 10).

3.3. Crystal polarization and DC quasi-mosaicity

The electric potential due to the external voltage U_0 across the plate is given by

$$V(x_2) = -U_0 x_2/s. \quad (12)$$

The electric field \mathbf{E} is constant,

$$|\mathbf{E}| = E_2 = -dV/dx_2 = U_0/s. \quad (13)$$

Although quartz is a good insulator, free charge carriers are always present because of the unavoidable impurities and crystal defects in a real structure. When high voltage is applied to the electrodes, these charges begin to move and to accumulate near the electrodes. A counter field results from this polarization phenomenon. This field builds up only slowly. It is not a uniform field, so that (13) no longer applies. The electric potential is approximated by the following relation (Ioffe, 1915):

$$V(x_2) = -U_0 x_2/s + U_t \sin(2\pi x_2/s), \quad (14)$$

where U_t characterizes the degree of crystal polarization. This parameter depends on time, on the number of free carriers, on their mobility, and so on. The electric field corresponding to the potential (14) is obtained by differentiation:

$$E_2(x_2) = U_0/s[1 - u_t \cos(2\pi x_2/s)], \quad (15)$$

where $u_t = 2\pi U_t/U_0$.

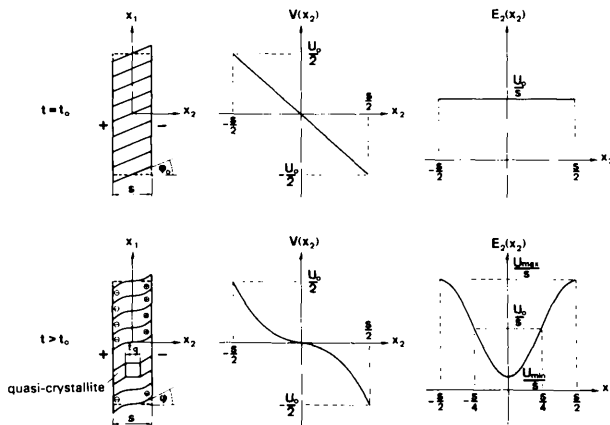


Fig. 11. Schematic shape of the (110) planes, potential $V(x_2)$ and electric field $E_2(x_2)$ distributions across the plate due to the polarization induced by the high voltage after a few hours. $U_{\max} = (1 + u_t)U_0$, $U_{\min} = (1 - u_t)U_0$.

Since E_2 is not constant across the plate, the angle of rotation φ of the diffraction planes will also be dependent on x_2 . Using (11) we obtain

$$\varphi(x_2) = d_{26}E_2(x_2) = \varphi_0[1 - u_t \cos(2\pi x_2/s)]. \quad (16)$$

$\varphi_0 = d_{26}U_0/s$ is the angle of rotation of the diffraction planes without space-charge effects.

In Fig. 11 are presented schematically the (110) diffraction planes, the potential $V(x_2)$ and the electric field $E_2(x_2)$ at two different times. The first time corresponds to the moment t_0 when the tension has just been applied to the crystal. At the second time some polarization has already built up. The change of orientation of the diffraction planes with respect to their situation at the time t_0 is given by

$$\xi(x_2) = \varphi(x_2) - \varphi_0 = -u_t \varphi_0 \cos(2\pi x_2/s). \quad (17)$$

It is clear from (17) that the relative orientation depends on x_2 and that the diffraction planes are deformed. As with the AC effect, we define a quasi-mosaicity responsible for the observed increase of reflectivity. Use of a procedure similar to that proposed in §2.3 leads to the following results:

$$t_q = \omega_D s / (4u_t \varphi_0), \quad (18)$$

$$R_{DC} = \kappa Q s f(A_q) \exp[-g_{DC} \kappa Q s f(A_q)]. \quad (19)$$

A_q is obtained by replacing t_q in relation (6a) by its DC value. As before, the secondary-extinction parameter g_{DC} , which depends on the disorientation of the quasi-crystallites, will decrease with u_t .

The measurement of the integrated reflectivity of a crystal as a function of the energy and diffraction orders is a time-consuming operation. When the DC effect is used, one has to be especially cautious, since a high voltage cannot be applied for arbitrary lengths of time. A decrease in voltage is recommended once the

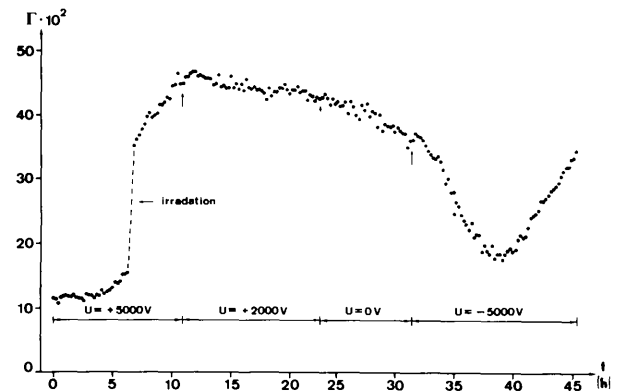


Fig. 12. Variation of the peak reflectivity as a function of time for various values and polarities of the high voltage applied on electrodes of the 2.5 mm thick crystal. The irradiation was performed with a 1 Ci (3.7×10^{10} Bq) ^{169}Yb source at a distance of 100 mm from the crystal and a time of 15 min. For these measurements, the 109.78 keV photons have been used in first order.

maximum reflectivity has been reached, thereby avoiding the occurrence of irreversible defects. The value of this reduced voltage has to be found empirically. As an illustration, it is shown in Fig. 12 that a voltage of 2000 V, used for test purposes after the crystal has been polarized under a 5000 V potential, was unable to maintain the optimal conditions. We therefore gave up quantitative verifications of relations (18) and (19). The following qualitative observations are offered:

– For some time after the application of the electric field, the polarization is low, so that $u_i \varphi_0 < \omega_D$. The induced quasi-mosaicity is too small to influence the reflectivity. The duration of this time lag depends on several factors which influence the number and the mobility of the charge carriers. In the case presented in Fig. 12, this time lag is about 3.5 h.

– Relation (17) shows that the disorientation ξ of the diffraction planes is proportional to φ_0 . Since φ_0 is itself proportional to U_0 , it is obvious that the DC effect will increase at high voltages. This is in agreement with the results shown in Fig. 9.

– One way to increase the number of free charge carriers is to subject the plate to intense ionizing radiation. The build up of polarization is then strongly enhanced and the reflectivity increases more rapidly, as demonstrated in Fig. 12.

– Heating the crystal increases both the number and the mobility of the free charge carriers, and speeds up the formation of space-charge effects and the increase in reflectivity. This hypothesis is borne out by experiment, as shown in Fig. 13.

– Finally, when the polarity of the external voltage on the electrodes is reversed, electric charges move in the opposite direction. This migration tends first to reduce (faster than voltage removal, see Fig. 9) the original polarization and the plane disorientation, until

a reverse polarization appears. This is in accordance with the observed variation of the reflectivity (Fig. 12).

3.4. Increase in peak reflectivity and reflection broadening

The DC effect permits larger increases in peak reflectivity than the AC effect. Moreover, the line widening is less important. These remarks are supported by a comparison of Figs. 1 and 14, where the two effects are compared under similar conditions. In both cases the mechanism which provokes the increase is similar so that the considerations given in § 2.4 are also valid for the DC effect. With regard to the

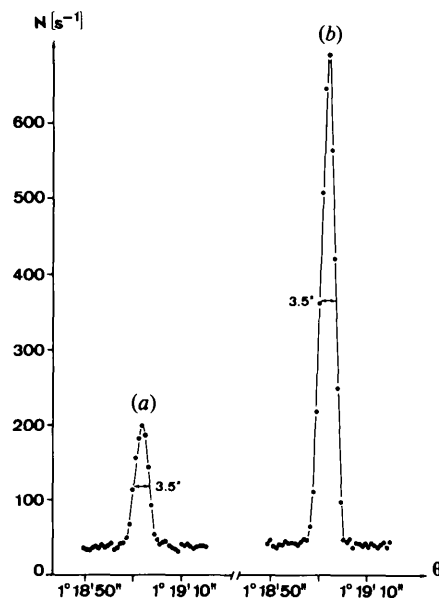


Fig. 14. Reflections due to 109.78 keV photons diffracted in first order by the 2.5 mm thick plate: (a) without electric field; (b) with the DC effect ($U_0 = 5000$ V, $t = 15$ h).

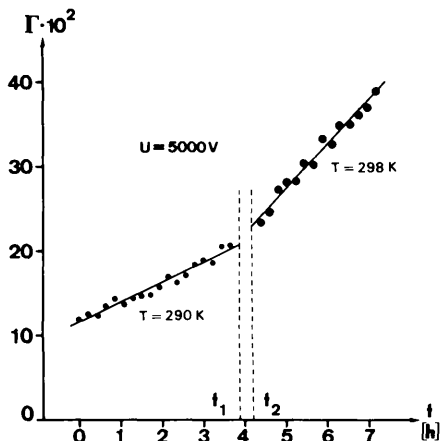


Fig. 13. Increase in peak reflectivity as a function of temperature observed with 109.78 keV photons diffracted in first order by the 2.5 mm thick plate. The time interval (t_1, t_2) was used for heating the crystal.

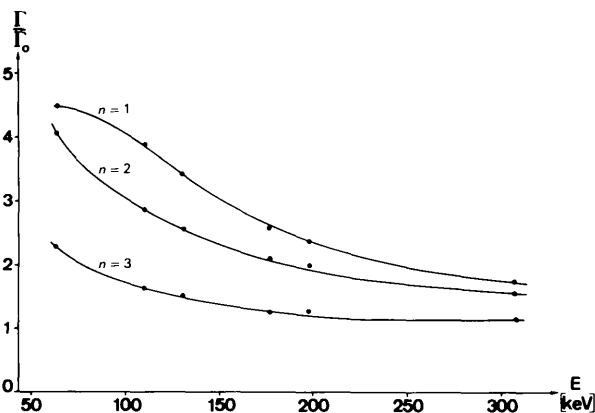


Fig. 15. Ratio Γ/Γ_0 as a function of the diffracted photon energies for three orders of reflection. The measurement was performed on the 2.5 mm thick plate. The polarization was obtained by application of a 5000 V high voltage for 15 h and maintained by 3000 V. Peak widening is negligible.

differences noted previously, they can be explained as follows:

The peak reflectivity can reach the maximum theoretical limit of 50% only if the following conditions are satisfied:

(a) The quantity A_q has to be sufficiently large to ensure the diffraction equilibrium.

(b) The width of the angular distribution of the quasi-crystallites must be large enough so that any photon from the source will pass through a properly oriented quasi-crystallite.

For equal deformations, *i.e.* for $\varphi_0^{\text{AC}} = u_t \varphi_0^{\text{DC}}$, and for all harmonics larger than first order, the AC quasi-crystallites will always be smaller than the DC quasi-crystallites. Condition (a) is therefore not as well satisfied for the AC effect as for the DC case. In addition, the local quasi-crystallite orientation cannot completely match the angular source width ω_s because φ_0^{AC} is modulated by the time dependence.

As for the AC effect, the relative increase in peak reflectivity I/I_0 is a function of energy and of diffraction order. These dependencies are illustrated in Fig. 15. Condition (a) is less well satisfied when E_v and n are increasing. This explains the observed trends.

To calculate the line widening due to the DC effect, we have proceeded as in § 2.4. The schematic representations in Fig. 6 are also valid here, but φ_0^{AC} must be replaced by $u_t \varphi_0^{\text{DC}}$. In our measurements the apparent angular source width was 2 arc sec. For the ideal case of a perfect crystal, the deformation $u_t \varphi_0^{\text{DC}}$ would need to be 1 arc sec to obtain a complete overlap of the quasi-crystallite angular orientation with the source. For a 5000 V high voltage, the rotation φ_0 of the (110) planes amounts to 2 arc sec (see Fig. 10). In this case, we would need to achieve a degree of polarization such that $u_t = 0.5$. According to Fig. 6, the peak widening should then amount to about 0.5 arc sec. No widening has been observed experimentally (see, for instance, Fig. 14). In fact a much smaller value of u_t was required since we are not dealing with an ideal crystal and since the lattice orientation has a non-vanishing distribution width.

3.5. Conclusion

Our results on the DC effect may be summarized as follows:

– The DC effect is basically more difficult to use than the AC effect. High voltages are required and the reflectivity increases only slowly. It is not easy to maintain the polarization over long periods of time. These problems do not appear in the AC effect. They are, however, not insoluble. As a counterpart, for thin sources, the relative increase in peak reflectivity is larger and the line widening less important.

– The reflection centre of gravity is not expected to drift if the polarization remains constant during the experiment. At low energies some line shift could

appear, since the front and rear parts of the crystal may contribute unequally. A measurement of the peak at positive and negative angles should, however, cancel out such an effect.

– For technical reasons it is difficult to work with very high voltages. Therefore, only crystals of limited thickness are usable and it is not possible to match very broad source widths.

– The peak widening should be negligible only if its original width is appreciably larger than $u_t \varphi_0$.

– As in the AC case, the DC effect will appear only if the electric field produces a rotation of the diffraction planes.

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