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E. Kittinger, E. Bertagnolli. A method for the visualization of secondary Dauphiné twinning in α -quartz. *Revue de Physique Appliquée*, 1979, 14 (5), pp.601-605. 10.1051/rphysap:01979001405060100 . jpa-00244635

HAL Id: jpa-00244635

<https://hal.science/jpa-00244635>

Submitted on 1 Jan 1979

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REVUE DE PHYSIQUE APPLIQUÉE

Classification

Physics Abstracts

78.20Hp — 42.80Nw — 77.60+v

A method for the visualization of secondary Dauphiné twinning in α -quartz

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(Reçu le 5 décembre 1978, accepté le 16 février 1979)

Résumé. — On montre que l'effet piézooptique permet, en utilisant la strioscopie, l'observation directe des macles du Dauphiné sous l'action d'une contrainte uniaxiale appliquée le long de l'axe polaire (X). Cette méthode peut être utilisée avec une mesure simultanée de la charge induite par l'application de la force pour l'étude du mouvement des macles du Dauphiné. Des photographies, montrant la formation et le développement d'une macle du Dauphiné sont présentées.

Abstract. — It is shown that the piezooptic effect allows direct observation of Dauphiné twins in α -quartz under uniaxial stress in X direction by use of the Schlieren technique. In conjunction with measurements of the piezoelectric polarization charge as a function of applied force this method may be used to study the dynamics of twinning. A sequence of pictures is presented showing the formation and growth of a twin domain.

1. Introduction. — The existence of Dauphiné-twins proved troublesome to the production of resonators from natural α -quartz. Primarily for this reason the possibility of detwinning by mechanical means was studied on the basis of thermodynamical considerations [1]. Natural quartz since has been largely replaced by synthetic crystals and the problem of twins has lost its importance in this respect.

In the course of the development of piezoelectric measurement techniques it turned out however that secondary twinning at elevated pressure and temperature imposes a severe limit on the application of quartz elements in pressure and force transducers. This practical aspect again prompted rising interest in this topic. Furthermore experimental work on this problem should help to provide the basis for the formulation of a microscopic theory of this phenomenon. So far theoretical considerations have been confined to the general framework of the theory of ferroic crystals [2, 3, 4]. In this context Dauphiné-twins are seen as domains of a ferroelastic crystal.

Among the methods employed for investigating the extent of twinning in a crystal etching techniques (for a survey and further references see [5, 6]) appear to have been most widely used. Together with diffraction experiments [7] and electron microscopy [8] they are basically of static nature and are used in general to study stable twin regions which extend out to the surface of the crystal.

A further group of methods is based on the difference in certain material properties in the two twin orientations. Experiments making use of the piezoelectric, the piezooptic or the electrooptic effect may be

used. Optical techniques appear particularly attractive as they allow (at least in principle) the direct observation of the formation and disappearance of twins, the determination of their extent and give information on the kinetics of domain wall motion. Aizu [2] and Dolino [9] both used the piezooptic effect for the visualization of twin domains using polarized light and differing crystal cuts. We describe here a method suitable for optical observation of Dauphiné-twins in a crystal orientation which differs from that used by either [2] or [9] and makes use of natural light.

2. The piezooptic effect in X -cut α -quartz. — Under mechanical stress the dielectric impermeability tensor B_{ij} of a crystal is altered in linear approximation according to the relation [2, 9]

$$B_{ij} = B_{ij}^0 + \Pi_{ijkl} T_{kl} \quad (1)$$

where Π_{ijkl} is called the piezooptic tensor, B_{ij}^0 is the impermeability tensor at zero stress, T_{kl} the stress tensor and where the Einstein summation convention is used. Referred to the crystal axes X, Y, Z which will be denoted below by x_1, x_2, x_3 respectively the tensor B_{ij}^0 for α -quartz has only diagonal elements and may be written as

$$B_{ij}^0 = \begin{pmatrix} 1/n_o^2 & 0 & 0 \\ 0 & 1/n_o^2 & 0 \\ 0 & 0 & 1/n_e^2 \end{pmatrix} \quad (2)$$

where $n_o = 1.544$ and $n_e = 1.553$ are the ordinary and extraordinary refractive indices respectively [10].

The piezooptic tensor is symmetrical with respect to an interchange of indices i with j and k with l which allows the use of the more compact matrix notation.

There is however no symmetry with respect to an interchange of the pairs ij with kl . The matrix of $\Pi_{\lambda\mu}$ has the following form

$$(\Pi_{\lambda\mu}) = \begin{pmatrix} \Pi_{11} & \Pi_{12} & \Pi_{13} & \boxed{\Pi_{14} & 0} & 0 \\ \Pi_{12} & \Pi_{11} & \Pi_{13} & \boxed{-\Pi_{14} & 0} & 0 \\ \Pi_{31} & \Pi_{31} & \Pi_{33} & 0 & 0 & 0 \\ \boxed{\Pi_{41} & -\Pi_{41}} & 0 & \Pi_{44} & 0 & \boxed{0} \\ 0 & 0 & 0 & 0 & \Pi_{44} & \boxed{2\Pi_{41}} \\ 0 & 0 & 0 & \boxed{0 & \Pi_{14}} & \Pi_{11} - \Pi_{12} \end{pmatrix} \quad (3)$$

where elements reversing their sign on twinning (reversal of the direction of the x_1 -axis) are indicated. The values of the independent coefficients are in units of $10^{-12} \text{ m}^2/\text{N}$ [11] :

$$\begin{aligned} \Pi_{11} &= 1.110 & \Pi_{31} &= 2.770 \\ \Pi_{12} &= 2.500 & \Pi_{33} &= 0.183 \\ \Pi_{13} &= 1.970 & \Pi_{44} &= -1.015 \\ \Pi_{14} &= 0.097 & \Pi_{41} &= 0.320. \end{aligned}$$

For the present experiments the optical properties under uniaxial stress in x_1 -direction and for light propagation in a perpendicular direction are of importance. The following considerations are therefore confined to the $x_2 x_3$ -plane. In order to find the refractive indices in an arbitrary propagation direction in this plane the usual procedure of determining the section of the representation quadric of B_{ij} (indicatrix) with a plane perpendicular to the wave normal is followed. To this end the B_{ij} are transformed to a coordinate system x'_1, x'_2, x'_3 which is rotated about the x_1 -axis through an angle φ in such a way that the x'_3 -axis corresponds to the incident wave normal.

The section of the indicatrix surface at zero stress with the $x'_1 x'_2$ -plane has the general form of an ellipse :

$$B_{11}^{0'} x_1'^2 + B_{22}^{0'} x_2'^2 + 2 B_{12}^{0'} x_1' x_2' = 1 \quad (4)$$

where

$$\begin{aligned} B_{11}^{0'} &= \frac{1}{n_o^2} \\ B_{22}^{0'} &= \frac{1}{n_o^2} \cos^2 \varphi + \frac{1}{n_e^2} \sin^2 \varphi \\ B_{12}^{0'} &= 0. \end{aligned} \quad (5)$$

From (4) two refractive indices are found :

$$\begin{aligned} n_o^{0'} &= n_o \\ n_e^{0'} &= \left[\frac{1}{n_o^2} \cos^2 \varphi + \frac{1}{n_e^2} \sin^2 \varphi \right]^{-1/2} \end{aligned} \quad (6)$$

$n_o^{0'}$ being independent of direction is the refractive index of the ordinary ray while $n_e^{0'}$ pertains to the extraordinary ray in the absence of mechanical stress.

Using matrix notation the relevant B_λ^0 are altered under the action of T_1 as follows :

$$B_\lambda = B_\lambda^0 + \Pi_{\lambda 1} T_1. \quad (7)$$

On account of the symmetry of α -quartz B_5 and B_6 vanish and the remaining terms take the following form :

$$\begin{aligned} B_1 &= B_1^0 + \Pi_{11} T_1 \\ B_2 &= B_1^0 + \Pi_{21} T_1 \\ B_3 &= B_3^0 + \Pi_{31} T_1 \\ B_4 &= \Pi_{41} T_1. \end{aligned} \quad (8)$$

Transformation of these B_λ to the x'_1, x'_2, x'_3 system leads to the following equation for the elliptic central section :

$$\begin{aligned} \left(\frac{1}{n_o^2} + \Pi_{11} T_1 \right) x_1'^2 + \left[\left(\frac{1}{n_o^2} + \Pi_{21} T_1 \right) \cos^2 \varphi + \left(\frac{1}{n_e^2} + \Pi_{31} T_1 \right) \sin^2 \varphi + \Pi_{41} T_1 \sin 2\varphi \right] x_2'^2 = 1 \end{aligned} \quad (9)$$

from which expressions for n_o' and n_e' are obtained :

$$\begin{aligned} n_o' &= n_o (1 + n_o^2 \Pi_{11} T_1)^{-1/2} \\ n_e' &= \left[\left(\frac{1}{n_o^2} + \Pi_{21} T_1 \right) \cos^2 \varphi + \left(\frac{1}{n_e^2} + \Pi_{31} T_1 \right) \sin^2 \varphi + \Pi_{41} T_1 \sin 2\varphi \right]^{-1/2} \end{aligned} \quad (10)$$

n_o' does not depend on the propagation direction in the $x_2 x_3$ -plane and therefore represents the stress dependent ordinary refractive index. Because of the smallness of $n_o^2 \Pi_{11} T_1$ (which does not exceed a value of 2.65×10^{-3} at $T_1 = 1 \text{ GPa}$) n_o' may be approximated by

$$n_o' = n_o \left(1 - \frac{1}{2} n_o^2 \Pi_{11} T_1 \right) \quad (11)$$

n'_e is dependent on φ and represents the stress dependent extraordinary refractive index for the particular propagation direction. Using (10) n'_e takes the form

$$n'_e = n_e^{0'} \left\{ 1 + n_e^{0'2} [\Pi_{31} + (\Pi_{21} - \Pi_{31}) \cos^2 \varphi + \Pi_{41} \sin 2 \varphi] T_1 \right\}^{-1/2}. \quad (12)$$

Again making use of the smallness of the second term in the bracket (5×10^{-3} at $T_1 = 1$ GPa) this may be written as

$$n'_e = n_e^{0'} - \frac{1}{2} n_e^{0'3} \times [\Pi_{31} + (\Pi_{21} - \Pi_{31}) \cos^2 \varphi + \Pi_{41} \sin 2 \varphi] T_1. \quad (13)$$

Of the components present in (11) and (13) only Π_{41} changes its sign as a consequence of twinning. Therefore n_0 has the same value in both twin orientations (+) and (-) :

$$n'_e(+)-n'_e(-)=0. \quad (14)$$

On the other hand n'_e differs in the two twin orientations depending on the propagation direction

$$n'_e(+)-n'_e(-)=n_e^{0'} \Pi_{41} T_1 \sin 2 \varphi. \quad (15)$$

Due to the fact that the angular dependence of $n_e^{0'}$ (6) is extremely weak for quartz a maximum of this difference is obtained on propagation under 45° to the x_2 and x_3 axes, while no difference occurs for rays propagating in either x_2 or x_3 direction.

The above analysis presumes a uniaxial stress T_1 . In an actual experiment pure uniaxial stress is hardly obtainable. For the present purpose it would not even be desirable since in this case no secondary twinning should occur for thermodynamical reasons. Since additional stress components are not known quantitatively a more general treatment of the optical problem is not warranted. What has been shown is the feasibility of visualizing twin domains by the Schlieren technique. For this the existence of a T_1 component (which will be dominant under the experimental conditions considered here) is sufficient to produce a refractive index gradient along the twin boundaries.

Fig. 1. — Specimen geometry. Specimen dimensions are $10 \times 10 \times 5$ mm³.

3. **Experiment.** — Based on the simplified treatment presented above sample geometry was so chosen as to yield the maximum effect (Fig. 1). Synthetic right-handed quartz crystals were used. Their side faces were ground and polished optically flat and parallel. The effect should be observable using white unpolarized light thus obviating the need for polarizers and compensators. The arrangement used is sketched in figure 2 and corresponds to the conventional

Fig. 2. — Schematic representation of experimental arrangement.

bright-field Schlieren-method. An incandescent lamp is used to illuminate after collimation a pinhole which serves as a point source S. Lens L_1 with a focal length of 0.5 m renders the light parallel. Lens L_2 produces an image of the source at the focus (focal length of 0.4 m) where the Schlieren diaphragm SD is placed. All light deviated due to inhomogeneities in refractive index is blocked by this diaphragm thus producing the typical Schlieren image revealing regions exhibiting a refractive index gradient.

Twin formation is a dynamical phenomenon. It was therefore desirable to provide for continuous observation. Consequently the Schlieren image (the position of which depends on the distance between sample and L_2) was projected directly onto the light sensitive screen of a video-camera VC which was connected to a video-recorder VR. Reproduction from the magnetic tape then provides a further contrast enhancing capability.

The mechanical stress necessary to create Dauphiné-twins as well as to render them visible was produced by means of a hydraulic press. The pressing pistons P were machined from especially hardened steel. The faces which were in contact with the crystal Q were ground flat anew before each experiment. Particular care was taken that the specimen was well centered in the press in order to insure that stress was evenly distributed. All experiments were conducted at room temperature.

The total force F exerted on the quartz crystal was measured with the aid of a piezoelectric force transducer T . Its signal was amplified by means of a charge amplifier and used to drive the X-axis of an XY-recorder. It is well known [1, 5, 6] that the existence of twins manifests itself by a reduced piezoelectric sensitivity. This effect has not yet been widely used as a

means of investigating the dynamics of twin formation. We have employed this method [12] simultaneously with optical observation. Using the pressing pistons as electrodes the polarization charge of the crystal specimen was measured and recorded along the Y -axis of the XY -recorder. This electrical measurement provides information on the stress threshold necessary to induce twinning, on the volume of the twin domains formed and on the role of the duration of the applied stress. Simultaneous optical observation confirms these findings and yields additional information on the spatial distribution of twins, on preferred sites of their formation and on the speed and direction of the motion of their boundaries.

4. Results and discussion. — The pictures reproduced below have been photographed from a TV screen. The apparent lack of parallelism of sample side faces as well as their curved appearance are due to distortions arising from the reproduction from the video tape. Since there was no possibility to synchronize the photographic shutter with the electron beam of the TV set exposure is not completely uniform giving rise to the slanting lighter bands which appear in some of the pictures.

An example which demonstrates very clearly the

capabilities of this method is shown in figure 3. Here an internal defect not visible in the unstressed state (Fig. 3a) apparently serves as starting point of twin formation (Fig. 3b) which sets in at an average stress (total applied force divided by crystal cross section) of $T_1 = 0.6$ GPa. At 0.8 GPa rapid growth in X -direction starts (Fig. 3c) which continues at nearly constant stress until the twin domain extends out to a surface of the crystal which in this set up is in contact with the pressing piston (Fig. 3d). Only after this breakthrough which is completed within one or two seconds further growth occurs in a perpendicular direction (Z -axis). This pattern of growth is observed quite generally.

In the bright field method employed here an inhomogeneity as produced by the boundary of a stressed twin domain should appear as a dark band in the bright field of view. This is confirmed by figure 3. However some parts of the crystal appear darkened while no twin is present there. In fact, although the force is applied uniaxially to the specimen, the resulting stress is by no means uniform. Deformation of the pressing piston faces is unavoidable leading to stress concentration around the edges. This also produces changes of index of refraction and a darkening of the respective areas in the Schlieren image.

Fig. 3. — Development of a twin under increasing stress.

Twins typically originate in this highly stressed region. Still, twins are observable there as well, appearing as bright bands in the darkened background. It seems that some of the light deviated due to stress concentration and therefore missing in the Schlieren image is refracted back by the twin boundary.

The difference in $n_e(+)$ and $n_e(-)$ as given by (15) probably is not the only reason for the observability of twin domains in a Schlieren arrangement. The fact that twinning occurs at all and calculations by the finite element method clearly show that stress components other than T_1 must be present as well. These components are relaxed by twin formation resulting in a lowering of the Gibbs free energy density. The state of stress in a twin domain then must be different from the surrounding crystal. In this case it is likely that there exists also a difference in ordinary refractive indices. If there were no contribution from the ordinary ray the use of polarized light corresponding to the extraordinary ray should greatly improve the contrast of the Schlieren image. No essential improvement was observed however confirming that the ordinary ray also contributes to the contrast.

In principle the precise extent and orientation of the domains could be determined by taking Schlieren images in two perpendicular directions. In practice however this was not possible since our experiments cannot really be termed *nondestructive*. To provoke twin formation at room temperature mean stresses from 0.5 to 0.8 GPa proved necessary. The stress concentrations in the vicinity of the edges then represent a rather rough treatment of the crystals. This almost invariably resulted in the breaking off of surface chips at two opposite faces. Once the optical polish is lost the surface is no more transparent in the Schlieren technique. The possibility of creating permanent twins by maintaining the maximum stress for hours however provides a means of studying the extent of those twins by etching techniques. This confirmed the findings of [2] and [3] that twins take the form of lamellae extending along the Z-direction.

5. Conclusions. — We have reported earlier [12] that our experiment resulted in reversible twin formation : the twin domains which formed during increasing load disappeared completely after removal of the load if the total duration of the stress cycle did not exceed the order of several minutes.

Since the change in refractive index is proportional to the stress T_1 it diminishes with decreasing load. The contrast in the image decreases in the same measure. This renders the observation of the disappearance of the twins from the crystal rather difficult. It could be established however that twin domains were still present and visible down to mean stresses as low as 25 MPa. Our concurrent electrical measurements indicate that a reduction in total twin volume begins at considerably higher stresses. This fact and our optical observation that the contrast just fades without revealing any moving twin boundaries lead us to assume that the twins disappear by first reducing the thickness of the lamellae (which has practically no effect on the Schlieren image) while maintaining their extension in Z-direction.

The advantages and shortcomings of our method may be summarized as follows : it may be used with X-cut crystals in an arrangement which resembles the conditions encountered in measurement applications. No special rotated cuts are necessary. In addition it yields direct information on preferred sites of twin formation, on the extent of twinning and on the time dependence of twin growth in a clear (typically magnified) image. The possibility of recording the observations allows to conveniently analyze results by replaying the tape even if the crystal is broken in the course of the experiments.

As shortcomings the difficulty of extracting quantitative information must be noted together with the fact that only a two dimensional projection of the crystal volume is obtained. Thus neither the location along the direction of light propagation nor the thickness of the twin lamellae may be determined. These disadvantages are shared with other optical methods, but may be circumvented if only stable twins are to be observed. In this case only moderate stress has to be used which does not result in damage to the crystal faces allowing (if necessary simultaneous) observation in two perpendicular directions. The limit of 25 MPa for the observability of twins indicates that our method provides about the same sensitivity as stated in [9].

Acknowledgment. — The authors are grateful to Prof. Dr. J. Kolb and to Doz. Dr. J. Tichý for their constant encouragement and for many helpful discussions.

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