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59

MAPPING DAUPHINÉ AND BRAZIL TWINS IN QUARTZ BY X-RAY TOPOGRAPHY

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Much effort has recently been devoted to precise measurement of intensities of x-ray reflections from single crystals of α quartz.[1–3] A difficulty that besets this work is the prevalence of interpenetration twinning following the Dauphiné Law. This Law mutually relates twinned parts by a 180° rotation about the c axis. Their lattices remain parallel, but the plane $(hkil)$ of one part coincides with the plane $(\overline{hki}l)$ of the other. Many $hkil$, $\overline{hki}l$ pairs of reflections have greatly differing structure factors. Hence measurement of a weak reflection $hkil$ may be vitiated even if only a small fraction of the crystal volume is Dauphiné-twinned with respect to the major part, when the reflection $\overline{hki}l$ is strong. In a recent study[3] it was found necessary to introduce a correction for Dauphiné twinning to explain discrepancies in the observed intensities of weak reflections. The best fit to the data was obtained by assuming that 1.5% of the crystal volume was Dauphiné-twinned with respect to the matrix. In this Letter it is pointed out that small amounts of Dauphiné-twinned material can be detected by x-ray topography, even when totally enclosed within the specimen. Their positions within the specimen, and their shapes, can be determined by stereo pairs of topographs. It is therefore suggested that any quartz crystal upon which it is planned to make careful x-ray reflection intensity measurements should be surveyed by transmission topography to establish its freedom from Dauphiné twinning. Etching can only detect Dauphiné twins that outcrop at the surface: x-rays probe the whole volume.

Figure 1(a) is a projection topograph of a single-crystal quartz plate which has five regions of Dauphiné twinning around its margins. In this topograph the major part of the plate is set to give the strong $10\bar{1}1$ reflection.[5] The twinned regions give the $\bar{1}011$ reflection whose structure factor is only 2/3 of that of reflection $10\bar{1}1$: their weaker reflecting power is evident on the topograph. The specimen is fairly perfect. Some inclusions and about a score of dislocations can be seen. Mottling on the topograph arises from the roughness of the deeply etched specimen surfaces. Pendellösung fringes[6] appear at twin boundaries.

The large difference between structure factors of some $hkil$, $\bar{h}\bar{k}\bar{i}\bar{l}$ pairs may be exploited to give excellent discrimination between Dauphiné twins. In Fig. 1(*b*) the major part of the specimen is set for the strong $30\bar{3}1$ reflection. The peripheral twinned regions give the very weak $\bar{3}031$ reflection: their images appear only faintly on the original topograph plate, and do not show up on the positive print Fig. 2(*b*). On the other hand, in Fig. 1(*c*), the peripheral twinned regions are brought into prominence by being set for the strong $30\bar{3}1$ reflection, in which case the major part of the specimen gives only the very weak $\bar{3}031$ reflection. Note the small region of Dauphiné twinning in the top right corner. This could easily be overlooked in an examination of the etched specimen surface. In a crystal of fairly uniform texture, free from gross misorientations, it may reasonably be estimated that a Dauphiné-twinned region as small as 50 μ in diameter should not fail to be detected by comparing topograph pairs such as $30\bar{3}1$ and $\bar{3}031$. Such a region would comprise only 1/2% of the volume of the specimen used in the work cited.[3] Mapping of Dauphiné twins by this method does not require the specimen to be highly perfect; it has been applied to relatively imperfect amethyst quartz.[7]

The second common type of parallel-lattice twinning in quartz follows the Brazil Law, a reflection in $(11\bar{2}0)$. Two structures related by this symmetry operation form an enantiomorphic pair and have opposite optical rotations. Their differentiation by diffraction methods is not so simply performed as in the case of Dauphiné twinning. It can be done using x-ray wavelengths which are scattered by silicon with appreciable anomalous dispersion, so that for certain reflections Friedel's Law fails. Owing to the

(a)
(b)
(c)

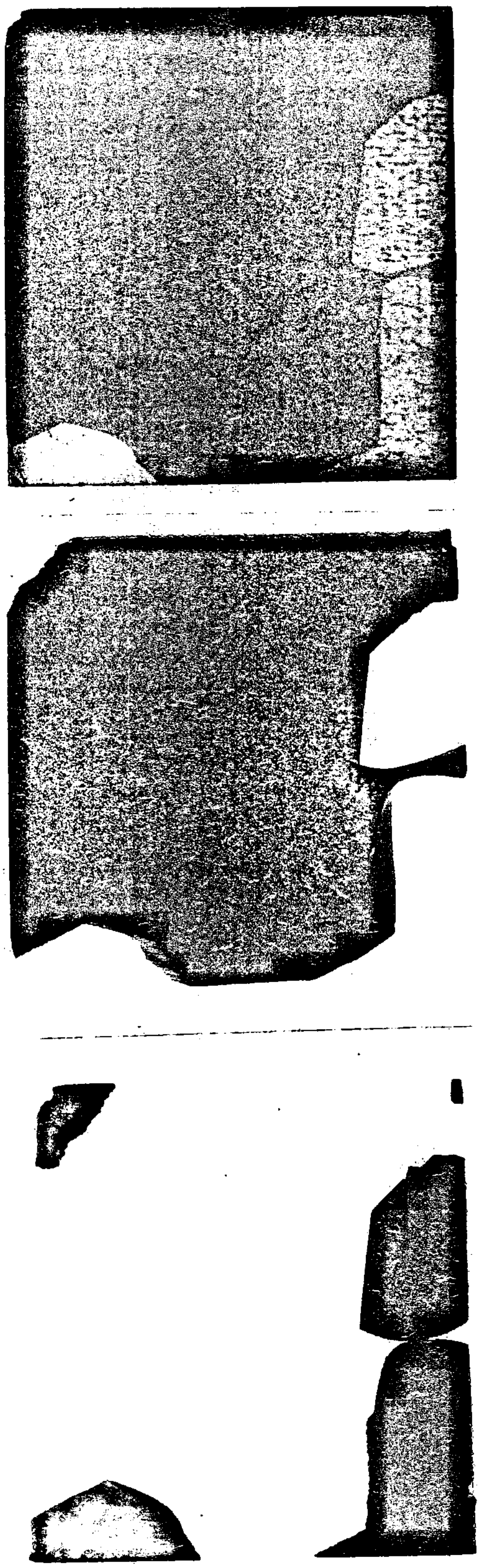

Fig. 1. Transmission topographs of Dauphiné-twinned quartz, Ag $K\alpha$ radiation. The specimen is 15 mm square in area, 1 mm thick, and makes about 11° with a rhombohedral plane. The direction [$2\bar{1}\bar{1}0$] is vertical. The central region gives a $10\bar{1}1$ reflection in (a) and a $30\bar{3}1$ reflection in (b). In (c) the peripheral twinned regions give a $30\bar{3}1$ reflection.

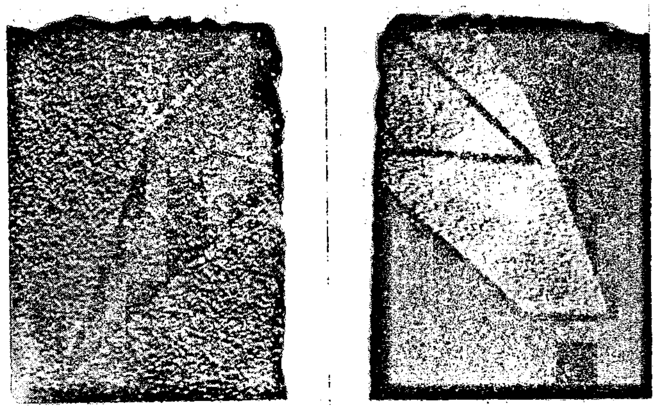

Fig. 2. Reflection topographs of both faces of a Brazil-twinned quartz plate making 11° with the major rhombohedron. Field area 6 mm × 8½ mm. Co $K\alpha$ radiation. Reflections are $11\bar{2}1$, $\bar{1}\bar{1}21$ and their inverses. Twin composition surfaces are parallel to a major and to a minor rhombohedral plane, and to a prism plane.

rather high absorption of such wavelengths by quartz, a reflection rather than a transmission technique is the more easily employed. These reflection topographs do not provide more information than is obtainable by etching, but they can be used as an alternative method for mapping outcrops of Brazil twins, applicable, for example, when it would be undesirable to destroy a polished surface by etching. A fairly strong reflection which is also strongly affected by anomalous dispersion is $11\bar{2}1$. The plane ($11\bar{2}1$) in one member of the twin pair lies parallel to ($\bar{1}\bar{1}21$) in the other member. When Friedel's Law is obeyed the reflections $11\bar{2}1$ and $\bar{1}\bar{1}21$ will be equally strong; this follows because the reflections $11\bar{2}1$ and $11\bar{2}\bar{1}$ are structurally equivalent in α quartz, through operation of the twofold symmetry axis parallel to [$11\bar{2}0$]. As a result of anomalous dispersion, the squares of the structure factors of reflections $11\bar{2}1$ and $\bar{1}\bar{1}21$ differ by as much as 43% with Co $K\alpha$ radiation[8] and by 20% with Cu $K\alpha$.[9] Figure 2 shows reflection topographs from both surfaces of a plate 1 mm thick which has a wedge-shaped insert of Brazil-twinned material. This insert itself contains a couple of Brazil-twinned lamellae, but one of them does not persist throughout the whole specimen thickness. In one topograph the reflections by the two components are $11\bar{2}1$ and $\bar{1}\bar{1}21$, respectively, and in the other topograph, taken by reflection from the back surface of the plate, they are their inverses $\bar{1}\bar{1}2\bar{1}$ and $11\bar{2}\bar{1}$. The intensities are clearly complementary and the delineation of the twinned regions outcropping on each surface is sharp.

Transmission topography of Brazil-twinned specimens using the softer radiations would be feasible with thinner specimens. With fairly perfect crystals one would expect a modification of the Borrmann effect by anomalous dispersion, with the possibility of strong discrimination between Brazil twins.

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[1] R. A. Young and B. Post, *Acta Cryst.* **15**, 337 (1962).

[2] G. S. Smith and L. E. Alexander, *Acta Cryst.* **16**, 462 (1963).

[3] W. H. Zachariasen and H. A. Plettinger, *Acta Cryst.* **18**, 710 (1965).

[4] A. R. Lang, *Acta Cryst.* **12**, 249 (1959).

[5] In this Letter the indices refer to the accepted morphological Miller-Bravais axes of α quartz. These differ by a 60° rotation about the c axis from those used in the diffraction studies cited.[1–3] This difference has been explained [A. R. Lang, *Acta Cryst.* **18**, (1965), in press] as arising from the work of P.-H. Wei[*Z. Krist.* **92**, 355 (1935)] who plotted in right-handed axes the coordinates which Wyckoff had derived in left-handed axes.

[6] N. Kato and A. R. Lang, *Acta Cryst.* **12**, 787 (1959).

[7] H. H. Schlössin and A. R. Lang, *Phil. Mag.* **12**, (1965), in press.

[8] A. de Vries, *Nature (Lond.)* **181**, 1193 (1958).

[9] W. H. Zachariasen, *Acta Cryst.* **18**, 714 (1965).