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X-ray Topographic study of Brazil-twin inserts in natural quartz

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ABSTRACT. — Natural quartz crystals were studied by X-ray topography to identify the nature of some defects that appeared as small inserts in the shape of right or equilateral triangles, when basal slices were studied with symmetrical reflections $\{10\bar{1}0\}$. Optical tests in polarized light and chemical etching of basal plates suggested that they were inserts of Brazil twins. Analysis of the topographic contrasts confirmed the optical tests and it was possibile to determine the fault; vectors $\underline{D}$ of twin-boundaries of two sets of twin inserts parallel to $(0\bar{1}11)$ and $(\bar{1}101)$. In addition, a third planar defect was observed and characterized at the intersection of the two sets.

Key words: Quartz, X-ray topography, Brazil twins.

Introduction

When studying the growth history of quartz crystals from natural druses by X-ray topography (SCANDALE et al., 1983; SCANDALE & STASI, 1985) various growth defects, mainly dislocations, inclusions, growth bands and growth-sector boundaries, were observed. Some other defects, appearing as right or equilateral triangles in the peripheral parts of basal slices when projected by symmetrical reflections $\{10\bar{1}0\}$, were also found.

Optical tests in polarized light and chemical etching of basal plates suggested that these defects might be inserts of Brazil twins (FRONDEL, 1962).

In accordance with literature (MCLAREN & PHAKEY, 1966; PHAKEY, 1969; MCLAREN & PITKETHLY, 1982) our initial observations showed that topographic contrast from Brazil twins could not be worked out as ordinary surface defects.

Nevertheless, twinning is a subject of continual interest among mineralogists and natural quartz crystals are so frequently twinned, according to Brazil, Dauphiné and Japan laws, that twinning is the rule rather than the exception. Brazil twins are difficult to recognize by classical mineralogical and crystallographic tools when they are thin plates or bodies of restricted size (satellites) inserted in a large crystal. In this case X-ray topographic studies, besides TEM observation, are available. A careful X-ray topographic investigation was therefore carried out using Lang's technique (LANG, 1959a; 1959b) which allows study of even small twin-inserts, without any loss of information on the whole crystal. In addition, it is possible to characterize the interface between the structures of the enantiomorphous pair that forms the twin.

Brazil twins were studied using different research tools and approaches and also by X-ray topography and T.E.M.

Brewster fringes, corresponding to the boundaries between right-handed and letf-handed quartz (polysynthetic twins) have been studied (MCLAREN & PITKETHLY, 1982). Fringe patterns originating from Brazil twin boundaries (LANG, 1967; LANG & MIUSKOV, 1969; MCLAREN & PHAKEY, 1966; PHAKEY, 1969; MCLAREN et al., 1967) were observed and fault vectors determined. Different models, involving the nature of the Brazil twin boundary and reflecting on the diffraction-contrast origin and the fault-vector characterization, have been proposed. Some authors (VAN GOETHEM et al., 1977) studied crystallographic aspects of Brazil twins and concluded that the model proposed by McLAREN & PHAKEY (1966) is correct.

In this paper attention is focused on the complex triangular contrasts of defects that so far have been assumed to be Brazil twins but without any conclusive analysis and fault-vector determination.

Experimental

Crystals of two quartz druses of different origins (Brazil, Italy) were studied by X-ray topography (SCANDALE et al., 1983; SCANDALE & STASI, 1985).

The diffraction contrasts studied in this paper were observed in a number of thin basal plates about 1 mm thick obtained from a quartz crystal of the Brazilian druse.

The slices, mechanically polished and chemically etched in 50% hydrofluoric acid for about 20 minutes, were observed under polarizing microscopes, both in transmitted and reflected light. In the peripheral parts of the slices, triangular regions of different polarization state were observed in transmitted light; rotatory power tests showed they were left-handed inserts in a right-handed matrix in accordance with Biot's convention (FRONDEL, 1962). Reflected light observations of etch-pits on basal planes confirmed the previous hypothesis. In fact, upon rotation of the microscope stage, different reflection powers could easily be appreciated between matrix and inserts.

It is therefore possibile to attribute Space Group $P3_221$ (structural left-hand screw axis: LS; right-handed coordinate system: RHCS) to the matrix, and Space Group $P3_121$ (structural right-hand screw axis: RS) to the inserts. Besides $30\bar{3}1$ X-ray reflection, which is less intese than the $\bar{3}031$ reflection, shows that the matrix is in Z(+) setting in accordance with Lang's (1965) mnemonic rule (DONNAY & LE PAGE, 1978).

X-ray topographs taken during this study are similar to those shown in Fig. 1. The results presented are concerned with the slice shown in Fig. 1. All the slices being similar, this slice was selected on the basis of the lower defect density in the region containing Brazil twins. X-ray topographs were taken with $MoK\alpha_1$ radiation for reflections $10\bar{1}0$, $01\bar{1}0$, $\bar{1}100$, $\bar{1}1\bar{2}0$, $\bar{2}110$, $20\bar{2}0$, $0\bar{2}20$, $\bar{2}200$, $10\bar{1}1$, $0\bar{1}11$, $\bar{1}101$, $\bar{1}01\bar{1}$, $0\bar{1}1\bar{1}$, $01\bar{1}\bar{1}$, $\bar{1}10\bar{1}$, $0\bar{2}21$, $02\bar{2}2$, $\bar{3}031$, $30\bar{3}\bar{1}$, $03\bar{3}1$, and $\bar{3}30\bar{1}$.

This number of reflections was considered necessary in order to know the exact crystallographic orientation of fault vectors and of the different composition planes defining the inserts.

Phase relations for the structure factors of the corresponding reflections in left-handed (LS) and right-handed (RS) structures, calculated by the SHELX76 (SHELDRICK, 1976) program using the atom positions given in literature (LE PAGE & DONNAY, 1976; DONNAY & LE PAGE, 1978), are reported in Table 1.

X-ray topographic observations

In Fig. 1 triangular diffraction contrasts can be seen along the (1010) slice edge. Their

TABLE 1

Phase relations for structure factors of corresponding reflections, φ_{LS} in $P3_221$ and φ_{RS} in $P3_121$. LS: structural left-hand screw axis; RS: structural right-hand screw axis

hkl	Φ_{LS}	Φ_{RS}	$\Phi_{RS}-\Phi_{LS}$
$10\bar{1}0$	π	π	0
$01\bar{1}0$	π	π	0
$\bar{1}100$	π	π	0
$20\bar{2}0$	0	0	0
$02\bar{2}0$	0	0	0
$\bar{2}200$	0	0	0
$10\bar{1}1$	$(4/3)\pi$	0	$(2/3)\pi$
$0\bar{1}11$	0	$(4/3)\pi$	$(4/3)\pi$
$\bar{1}101$	$(2/3)\pi$	$(2/3)\pi$	0
$01\bar{1}\bar{1}$	0	$(2/3)\pi$	$(2/3)\pi$
$01\bar{1}1$	0	$(4/3)\pi$	$(4/3)\pi$
$0\bar{1}1\bar{1}$	0	$(2/3)\pi$	$(2/3)\pi$
$0\bar{2}21$	π	$(1/3)\pi$	$(4/3)\pi$
$03\bar{3}1$	0	$(4/3)\pi$	$(4/3)\pi$

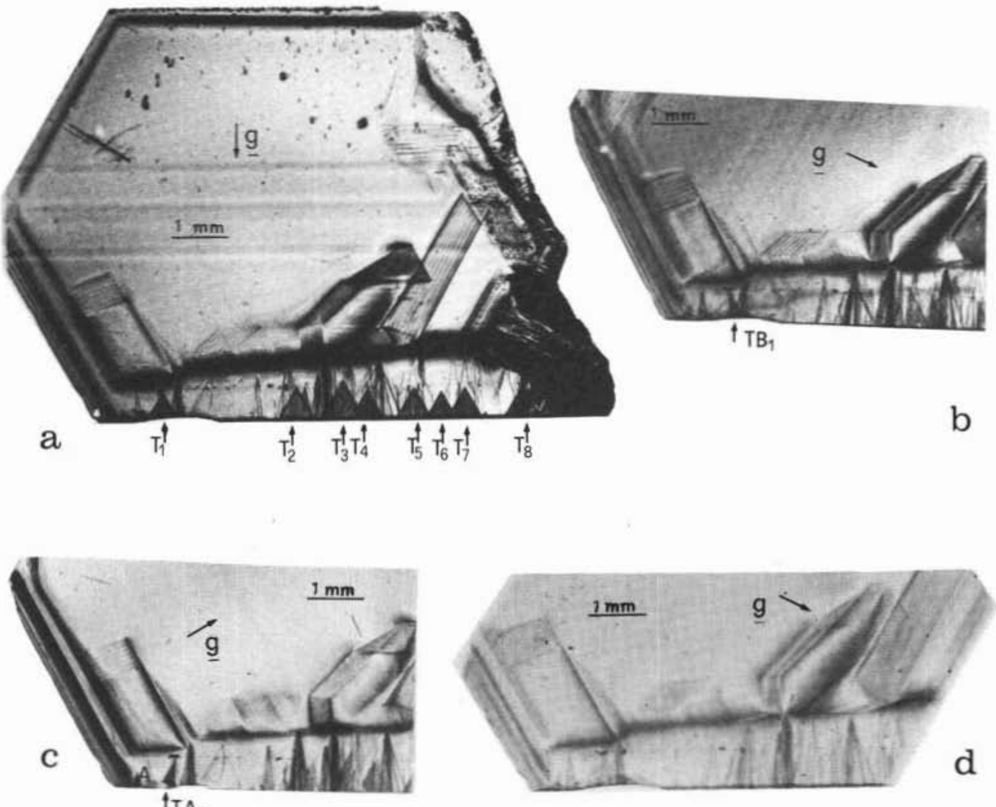

Fig. 1. — Natural quartz crystal from Brazil. Basal slice. X-ray diffraction topographs, $MoK\alpha_1$ radiation. T: Inserts of Brazil twins. TA: Brazil-twin insert parallel to $(0\bar{1}11)$. TB: Brazil-twin insert parallel to (1101). a) $\underline{g} = 10\bar{1}0$. Complete view of slice. TA and TB both in contrast. b) $\underline{g} = 01\bar{1}0$. Partial view of slice. TA out of contrast, TB in contrast. c) $\underline{g} = \bar{1}100$. View like in b). TA in contrast. TB out of contrast. d) $\underline{g} = 0\bar{2}20$. Inserts out of contrast.

shape changes with the reflections: in Fig. 1a) the contrasts, labelled Ti (i = 1, 2,...), appear as equilateral triangles with all sides parallel to traces of prism faces; in Figs 1b) and 1c) they are labelled respectively TB_1 and TA_1 and appear as right-angle triangles with bases parallel to prism face $(10\bar{1}0)$; the hypotenuse is parallel to a prism face at 120° to $(10\bar{1}0)$ and the third side parallel to [210], normal to the slice edge. From now on we will refer only to T_1, TA_1, TB_1, using the quoted capital letters without any suffix, as the contrast changes of all triangular-shaped defects have the same appearance.

TA defects are in strong contrasts for $g = 10\bar{1}0$ and $g = \bar{1}100$ (figs 1a and 1c); TB defects for $g = -10\bar{1}0$ and $g\ 01\bar{1}0$ (Figs. 1a and 1b). Both are out of contrast for the third equivalent reflection $\{10\bar{1}0\}$ and for all equivalent reflecions $\{20\bar{2}0\}$ (see, for example, $g = 0\bar{2}20$ in Fig. 1d) and in weak contrast for all the equivalent reflections $\{11\bar{2}0\}$ (not reported here).

Moreover, TA defects are out of contrast for $g = 01\bar{1}\bar{1}$, $g = 03\bar{3}1$ and $g = 0\bar{2}21$, reflections in which TB are in contrast (Fig. 2). TB defects are out of contrast for $g = \bar{1}101$ (Fig. 2c).

Determination of fault vectors

The contrast origin of Brazil twins has been studied in electron microscopy (McLaren & Phakey, 1966; Van Goethem et al., 1977), and in X-ray topography (Phakey, 1969; Lang, 1967). It was proved that a Brazil twin introduces a phase-shift into the X-ray wavefield due to the boundary between the two opposite structural LS and RS regions. Then the total phase-shift introduced is given

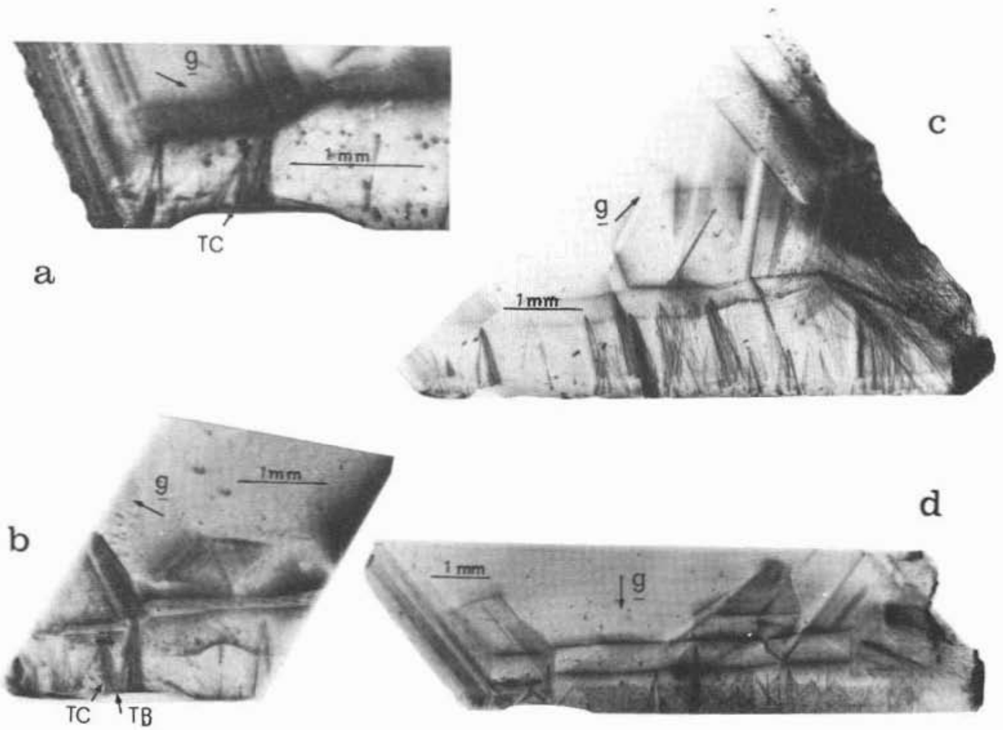

Fig. 2. — Same quartz basal plate as in Fig. 1. X-ray topographs. Characterization of defects. TA and TB as in Fig. 1. TC: boundary plane at intersections of TA and TB.
a) $\underline{g} = 03\bar{3}1$. TA out of contrast, TC in contrast. b) $\underline{g} = 0\bar{2}21$. TA out of contrast, TB and TC in contrast. c) $\underline{g} = \bar{1}101$. TB out of contrast. d) $\underline{g} = 10\bar{1}1$. TA and TB in contrast, TC out of contrast.

by

$$\alpha = 2\pi \underline{g} \cdot \underline{D} + \Delta\varphi$$

where $\underline{g}$ is the reciprocal lattice vector of the operating reflection, $\underline{D}$ the fault vector which relates the two regions relative to one another, and $\Delta\varphi = \varphi_{RS} - \varphi_{LS}$ where φ_{RS} and φ_{LS} the phase angles of the structure factors F_{RS}(hkil) and F_{LS}(hkil).

The Brazil twin boundary is out of contrast when $\alpha = 2\pi n$, with $n = 0, \pm 1, \pm 2...$

It was also shown that fault vectors $\pm 1/2$ [110], 1/6 [03$\bar{2}$] and 1/6 [302] are necessary to provide continuity between the left- and right-handed structures, as the disruption of the oxygen bond across the boundary is to be discarded.

These vectors bring into register (PHAKEY, 1969); VAN GOETHEM et al., 1977) two of the three Si atoms at the boundary, while the third becomes an interstitial atom lying over the $\underline{c}$-axis channel.

Comparison of the phase relationships of Table 1 with the extinction observations, reported in the previous paragraph, fault vectors $\underline{D}_A$ and $\underline{D}_B$ of boundaries TA and TB were determined to be $\underline{D}_A = 1/6$ [302] and $\underline{D}_B = \pm\ 1/2$ [110]: the composition planes of Brazil twin boundaries associated to these vectors are respectively (0$\bar{1}$11) and ($\bar{1}$101). The traces of these planes on (0001) are the sides of the equilateral triangles shown in Fig. 1a.

Discussion and conclusions

The contrast observation shows no fringe patterns (Fig. 1) of the twin boundary and right triangles (Figs. 1b and 1c) with one side normal to the crystal edge, i.e., parallel to [210]. It is therefore necessary to take into account that the actual structure of the body twinned inserts consists of two systems of Brazil twin-inserts parallel to (0$\bar{1}$11) and ($\bar{1}$101). In fact, in this case, projection topographs cause overlapping of the fringe patterns of each system of inserts that, in turn, give rise to an uniform contrast, due to the poor resolution of the topographic technique. Also taking into account the fact that the intersection of $(0\bar{1}11)$ and $(\bar{1}101)$ is parallel to [211], whose projection into the basal planes is [210], the overlapping explains the right-triangle shape of contrast of one system when the other is out of contrast, in place of the elongated obtuse triangle with the shortest side parallel to [210] that can be observed in the case of a single twin lamella (PHAKEY, 1969).

Careful examination of the topographic contrast where TA and TB systems are both out of contrast shows no stair-rod dislocations at the intersection of the twin boundary, like those observed in a simple case (PHAKEY, 1969). These dislocations, with line orientations parallel to [211] and Burgers vectors given by the sum $\underline{D}_A + \underline{D}_A$, are expected because the two systems have different fault vectors.

A solution to this problem is given by the observation of a triangular contrast, marked TC in Figs. 2a and 2b, which may represent the interface between the two sets of Brazil twins. The interface containing directions [211] and [210] may be parallel to plane $(1\bar{2}10)$. Fault vector $\underline{D}_C$ associated to the TC defect may be related the sum of the fault vectors associated to the intersecting twins by:

$$\underline{D}_A + \underline{D}_B + \underline{D}_C = \text{lattice vector}$$

from which it follows that $\underline{D}_C = 1/6\ [03\bar{2}]$ or $1/6\ [0\bar{3}\bar{2}]$.

Extinction observations ($\underline{g} = 10\bar{1}1$ in Fig. 2d and all equivalent reflections $\{20\bar{2}0\}$; Fig. 1d) confirm that the determined $\underline{D}_C$ vector is the fault-vector of the boundary.

As a last remark, we notice that three different families of Brazil twins have been observed and characterized in the studied regions. Until now three different families have never been encountered in one twinned region of the same crystal (VAN GOETHEM, 1977).

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