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On distinctive growth marks in quartz druses

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ABSTRACT. — Crystals from two quartz druses of different origins (Brazil, Italy) have previously been studied by X-ray topography (SCANDALE et al., 1983; SCANDALE and STASI, 1985). In this paper, their growth histories are compared and observed defects related to growth stages. In particular, dislocations were found to be directly related to changes in chemico-physical growth conditions and may be seen as growth marks that characterize and differentiate the druses studied.

Key words: quartz druses, growth marks, X-ray topography.

RIASSUNTO. — Cristalli prelevati da due druse di quarzo di differenti origini (Brasile, Italia) sono stati studiati mediante topografia R. X (SCANDALE et al., 1983; SCANDALE e STASI, 1985). In questo lavoro vengono confrontate le loro storie di crescita ed inoltre si correlano i difetti osservati agli stadi di crescita. In particolare, si è trovato che le dislocazioni sono direttamente correlate alle variazioni di condizioni chimico-fisiche di crescita e che possono essere considerate come marchi di crescita che caratterizzano e differenziano le druse studiate.

Parole chiave: druse di quarzo, difetti di crescita, topografia R. X.

Introduction

While the earliest studies of natural crystals by X-ray topography (LANG, 1959) focused on structural defects (LANG, 1963; AUTHIER and SAUVAGE, 1966; LANG, 1967), topographic study of defects was later applied to reconstruct the growth history of individual minerals (LANG, 1974; GIACOVAZZO et al., 1975; ISOGAMI and SUNAGAWA, 1975) and to allow comparative study of single crystals of different origins (CORNEY et al., 1976; SCANDALE et al., 1979 a and 1979 b). This type of study has proved to be very useful from a mineralogical point of view since it provides important information about the genesis of minerals (GRAZIANI et al., 1981), the origin of the defect (SCANDALE and ZARKA, 1982), and the nature of optical anomalies (TANNER, 1972; SCANDALE et al., 1984) even when there is a total lack of geological and petrographic data concerning the origin of sample.

Since the ultimate goal of mineralogy is to contribute to knowledge of the chemico-physical history of the Earth, it was quite natural to study crystals grown in the same conditions so as to find possible growth marks characterizing the growth history and genetic medium of the minerals.

This was in fact the aim of the defect study of crystals from a Brasilian quartz druse (SCANDALE et al., 1983), results of which indicated intrinsic unit defects of crystals from a druse. A complete growth-history reconstruction has allowed us to state that since different defects correspond to different stages of growth history, defects may be considered as growth marks for the druse.

In particular, inclusion absorptions indicated growth-condition changes and new dislocation sets were detected in the growth stages following the impurity absorption.

The study of a second quartz druse from Rocca dei Cristalli, Val Malenco, Italy (SCANDALE and STASI, 1985) confirmed the role of inclusions and dislocations as growth marks, even in a druse from a different geological site, and supplied more information on growth marks. In fact, prominent $x\ \{5161\}$ growth bands were observed and related to the line orientation of dislocations, indicating that growth bands corresponding to minor forms must also be taken into account in growth-mark research,

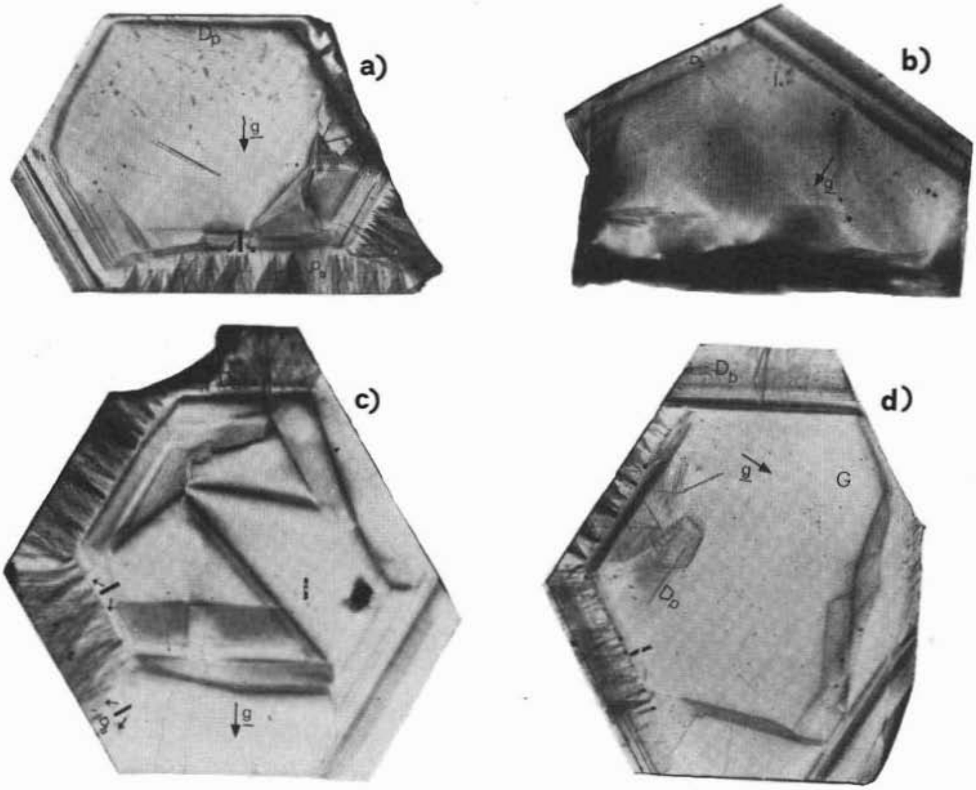

Fig. 1 *a-b*. — *BD* quartz druse (Brazil). X-ray topographs. D_b: basal dislocations. *G*: prismatic growth horizons. D_p: pyramidal dislocations. *I*: inclusions. - *a*) BDI crystal. Slice 2. $g = 10\bar{1}0$. *b*) BDI crystal. Slice 8. $g = \bar{1}100$. *c*) BDII crystal. Slice 2. $g = 10\bar{1}0$. In BDI crystal, starting from S5 (fig. 3) and more so up to S10, a deterioration of the crystalline quality can be seen. In BDII crystal, evidence of crystalline quality deterioration is present only at level of S5 (not reported here).

The purpose of this paper is to compare the growth histories of the studied druses as an initial step towards a generalized study of growth defects in crystalline aggregates. The study of a third quartz druse, from a well-know geological site, is now underway as a further generalization and in order to connect structural defects to geological data.

Materials and methods

Crystals of two quartz druses were studied by X-ray topography (Scandale et al., 1983; Scandale and Stasi, 1985). The sample sites are not available: it is only known that the first druse, labelled BD, comes from Brazil while the second, labelled RC, is from Rocca dei Cristalli, Val Malenco, Italy. Two single crystals were cut from each druse and labelled (BDI, BDII) and (RCA, RCB) respectively. In order to obtain general results, the crystals of each druse were selected with different sizes and development of crystal faces. Successive series of thin plates about 1 mm thick were cut parallel to basal plane, from the bottom to the top of the crystals: ten plates were obtained from each BD crystal and six successive plates from each RC crystal, a number considered necessary to give a careful reconstruction of the growth histories.

The slices were then mechanically polished and chemically etched. X-ray topographs were taken by MoKα_1 radiation.

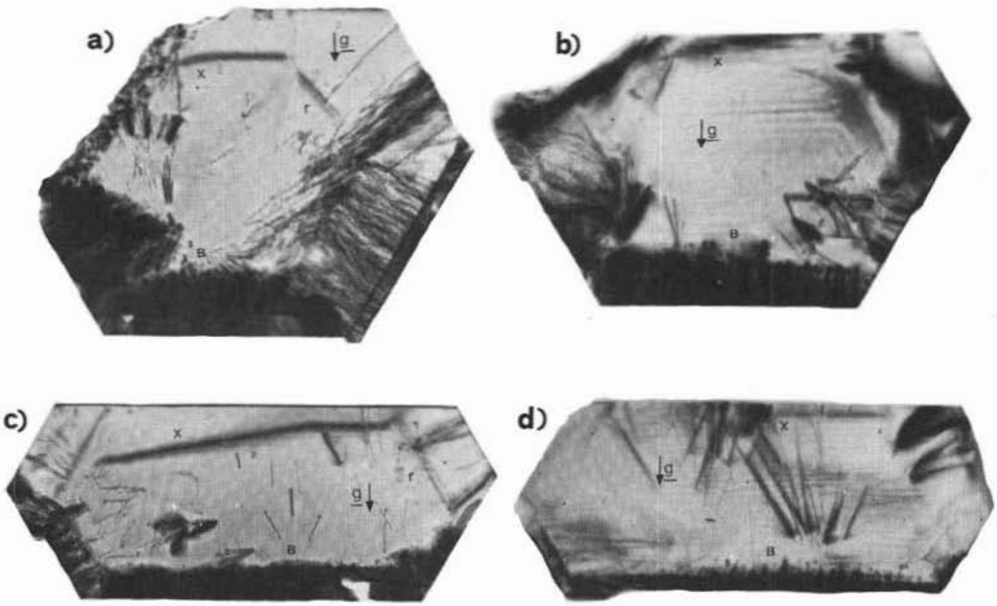

Fig. 2 *a-d*. — *RC* quartz druse (Rocca dei Cristalli, Val Malenco, Italy). X-ray topographs. D_b: basal dislocations. *B*: contrast due to impurity absorption. *r* and *x*: growth bands parallel to $\{10\bar{1}1\}$ and $\{5161\}$ respectively. - *a*) *RCA* crystal. Slice 1. $g = 10\bar{1}0$. *b*) *RCA* crystal. Slice 6. $g = 10\bar{1}0$. *c*) *RCB* crystal. Slice 2. $g = 10\bar{1}0$. *d*) *RCB* crystal. Slice 6. $g = 10\bar{1}0$. In both samples, effects of impurity absorption can be seen near the crystal edges and in the central parts, starting from S4 slices (fig. 4).

X-ray topographic observations

This paper deals with the role of lattice defects as growth marks in crystals from quartz druses. Information about defects was obtained by the authors in the course of extensive investigation into growth histories of the previously quoted druses. Thus in the following account, defects will only be described briefly, after which their role as growth marks will be discussed in detail.

Figs. 1 and 2 show two series of topographs corresponding respectively to BD and RC druses. Although distribution, density and type of the observed defects-inclusions, dislocations and growth bands-often differ, features common to the druses can be recognized, with the following points characterizing growth in both cases:

1) absorption of impurities marks a boundary between the first and the second half of the growth process. Topographs of BDIS5 and RCBS4 slices clearly show a deterioration of crystalline quality (figs. 3 and 4);

2) after the quoted absorption of impurities new sets of dislocations were nucleated. In BD druse $<a+c>$ pyramidal dislocations (fig. 3) were characterized (SCANDALE et al., 1983) and in RC druse $<a>$ dislocations, whose lines form an angle of about 10° with basal plane, were observed (fig. 4) and labelled as pseudo-basal dislocations (SCANDALE and STASI, 1985). To date these dislocation sets have not been characterized in as-grown crystals.

Different growth bands were also found in both druse crystals: they are mainly parallel to *m* $\{10\bar{1}0\}$ faces in BD and to *r* $\{10\bar{1}1\}$ and *x* $\{5161\}$ in RC druse (figs. 3 and 4). The importance of the latter *x* bands lies in their strict relation to pseudo-basal dislocations (p.b.d.); their role will be examined later on.

Discussion

From the comparative analysis of defects of the two druse it is possible to state that although their growth histories were on the whole quite similar, an interal unit may be stressed that permits their differentiation and characterization to be made.

In fact the observed defects allow two growth stages, separated by a precipitation period, to be distinguished in both druses. The first growth stage was quite perfect and in lower slices defects are mainly found near the crystal edges (figs. 1 and 2). Basal dislocations with 1/3 $<11\bar{2}0>$ Burgers vectors may be recognized.

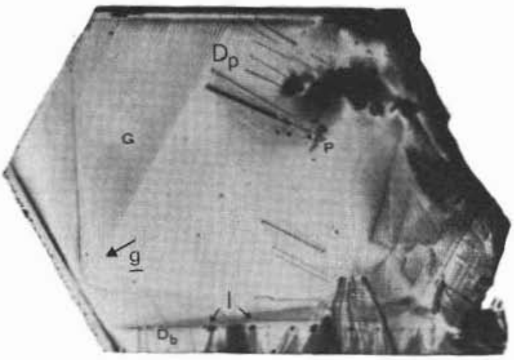

Fig. 3. — BDIS5. X-ray topograph. $g = 1\bar{1}00$. D_p: pyramidal dislocations. D_b, G, I and P as in fig. 1.

The precipitation period affects both druses almost at one half of the crystal developments along the c axis. The second growth half is characterized by a deterioration of crystalline quality (see f.e. the top slices in figs. 1 and 2), but especially by nucleation of dislocations not detected in the first growth half. In BD druse, pyramidal dislocations 1/3 $<11\bar{2}3>$ in $\{10\bar{1}1\}$ planes were characterized, whereas in RC druse 1/3 $<11\bar{2}0>$ in (0001) and p.b.d. dislocations were observed. The full dislocation characterization (l, v_b, and (l, v_b) plane) and complete growth-history reconstruction of the crystals allows us to correlate defects and growth. Due to their importance in characterizing and differentiating the druses, it is worthwhile to devote some time to dislocations observed in the different druses.

BD druse. Pyramidal dislocations $< a + c >$ in $\{10\bar{1}1\}$. The Burgers vector is the salient feature of these dislocations.

In fact, although dislocations in natural and synthetic quartz crystals have been extensively studied by various authors, their occurrence in as-grown crystals has only been suggested with no conclusive characterization (LANG and MIUSCOV, 1969; MC LAREN et al., 1971). In addition they were not observed in RC druse, and have only been characterized to date in experimentally deformed quartz crystals (TREPIED and DOUKHAN, 1982). On the other hand, by comparing the elastic energetic preference of one $< a + c >$ dislocation over two separated dislocations of Burgers vector $<a>$ and $<c>$, it may be concluded that $< a + c >$ dislocations can decompose spontaneously (KIRBY, 1975), although ΔE is moderate.

This being the case, their nucleation and stability can be justified only by assuming local changes of energy factor K depending on variations in elastic constants due to the sudden impurity absorption. Thus, pyramidal dislocations must be considered as distinctive growth marks.

RC druse · $<a>$ p.d.b.

The line orientation is the salient feature of these dislocations.

In fact, the numerous theoretical and experimental studies published on dislocations in quartz crystals indicate that the commonest (l, v_b) planes for $<a>$ dislocations are (0001) and $\{10\bar{1}0\}$ (see f. i., ARDELL et al., 1974). On the other hand, in agreement with the theoretical predictions observations confirm preferred orientations (TREPIED, 1978). As they are planes and directions, the line orientations of p.b.d. can only be explained by taking into account the growth bands parallel to x $\{5161\}$ planes (fig. 4).

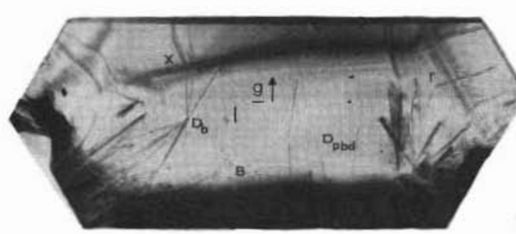

Fig. 4. — RCBS4. X-ray topograph. $g = 10\bar{1}0$. D_{pb}: pseudobasal dislocations. B, D_b, r and x as in fig. 2.

In fact, since it was experimentally determined that dislocation lines l form a 10° angle with (0001) plane and $(5161)\hat{}(0001)$ was calculated to be 81.95°, l is nearly perpendicular (92°) to (5161). In addition, as growth bands x correspond to well-developed faces observed in the actual habit of RC crystals, p.b.d. were forced to be nearly perpendicular to the growning faces in order to minimize their elastic energy (KLAPPER, 1972).

The strong relation between p.b.d. and growth conditions (impurity-absorption stage) is clearly supported by the observation that, in quartz morphology, x {5161} is the next important form after three major ones: m $\{10\bar{1}0\}$, r $\{10\bar{1}1\}$, and z $\{01\bar{1}1\}$.

However, the importance of this form is anomalous in terms of any growth theory (DOWTY, 1976); its prominence can be attributed to the retardation of growth due to absorption of foreign ions or molecules (HARTMAN, 1959). Indeed, since synthetic crystals show high concentrations of foreign ions in {5161} sectors (COHEN, 1963), the anomalous line orientations of p.b.d. may be attributed to impurity absorption in the second half of the druse growth. Thus, they are linked to growth history and must be taken as distinctive growth marks.

Conclusions

Defects and particularly dislocations observed in studied crystals must be seen as distinctive growth marks that distinguish and characterize the two druses.

In fact, as they have been shown to be closely linked to growth conditions, the anomalies of the observed dislocations can only be explained by considering the entire growth history of the crystals. In the BD druse growth-condition changes caused the nucleation of dislocations whose v_b is anomalous, while in the RC druse, dislocations occurred with anomalous line orientation l. Furthermore RC druse is characterized by the prominence of the x {5161} form in the second half of the growth. Taken as a whole, these observations indicate that impurity absorption is responsible for the nucleation of growth marks.

In this context, it would be interesting to analyse the foreign ions in the two druses in an attempt to determine the cause of these observed differences.

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