Effects of Electrostatic Field on the Diffraction Contrast of Imperfections in Quartz

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AT-cut oscillator plates were examined by X-ray diffraction topography under an applied electrostatic field of more than 5 kV/mm. Growth layers, cellular structures and some misfit boundaries, which are hardly observable without the field, can be detected with remarkable contrast. After the field has been switched off, the effects decrease with a relaxation time of about 10 hours. Not only the converse piezoelectric effect due to an instantaneous field, but also ionic or electronic charges account for the contrast change. The present technique is useful for studying crystal imperfections.

1. Introduction

The X-ray diffraction topography originated by Lang (1959) is a useful non-destructive technique for studying crystal imperfections. Since then, several studies have been reported on quartz (Spencer & Haruta, 1966; Spencer & Smith, 1966; Lang, 1967; Lang & Miuscov, 1967; McLaren, Osborne & Saunders, 1971; Homma & Iwata, 1973; Kume & Kato, 1974). Another type of study has been carried out by Spencer & Hunt (1966) and Young & Wagner (1966). In the latter studies, an alternating electric field was applied to quartz oscillators and the oscillation modes were studied through the diffraction contrast in the topographs.

The present authors found in quartz that static fields also give rise to a drastic change in the diffraction contrast. Here, some of the preliminary results concerning crystal imperfections, particularly defects pertinent to crystal growth, are reported.

2. Experimental

The specimens were AT plates used as electric oscillators, which were prepared from synthetic quartz as circular discs of 10 mm in diameter and 0.1 mm in thickness. The peripheries were tapered by several degrees. The main part of the sample was cut from the Z growth sector of the crystal as shown in Fig. 1. Natural quartz was also studied.

Circular silver electrodes, 6 mm in diameter, were plated by vacuum evaporation onto the discs. Two wire rings were attached to the electrodes as illustrated in Fig. 2. A static voltage of up to a few kilovolts was gradually applied to the specimen. Usually it took about one hour to attain the final voltage without electrical breakdown.

The diffraction topographs were obtained by the conventional method, but under the applied electric field. Ag Ka1 radiation and nuclear emulsion plates were used for the topographs. The exposure time was about 40 h. For studying the time dependence of the field effects, Mo Ka1 and conventional X-ray film were used. The exposure time was one or two hours in this case.

3. Results

(a) The effects of the electric field

Up to about 500 V no essential effect was observed. The appreciable changes usually occurred in the topographs when the potential was more than several hundred volts. The voltage required, however, depended upon the specimen. With increasing voltage, the changes became noticeable.

Fig. 3(a) shows a topograph before applying the voltage and Fig. 3(b) is a topograph of the same specimen obtained at 2500 V. Figs. 4(a), (b) and (c) are similar ones for another specimen. The reflexion vector seed

Fig. 1. Cutting of specimens from synthetic quartz.

Fig. 2. The application of the electrostatic field.
The diffraction contrast of imperfections in quartz is shown in the figures. The circular contrast corresponds to the edges of the electrodes. The contrast depended upon the specimens and to some extent on the sign of the applied field with respect to the incident direction of the X-rays.

The growth layers (L) and cell boundaries (C) (Lang & Miuscov, 1967) which could hardly be seen in the original specimens became clearly observable after the voltage had been applied. Also, the contrast of various kinds of defect, such as line defects (L), cell boundaries (C'), growth sector boundaries (S), became either clearer than or different from the original contrasts. The labels of the defects are given in Figs. 3 and 4.

The effects of the field for the defects extend over the

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(a) (b)

Fig. 3. (a) Before application of the electrostatic field. (b) With a voltage of 2500 V.

(a) (b)

Fig. 4. (a) Before application of the voltage. (b) With a voltage of +1500 V.
whole of the specimen across the contrast of the edge of the electrode as seen in Fig. 3(b), where the edge contrast is relatively weak. In Fig. 4(b) and (c), where the edge contrast is very high, the effects for the defects outside the edge are not as noticeable as in Fig. 3(b).

The phenomena mentioned above were also observed in natural quartz.

(b) The time dependence of the field effects

Fig. 5 is a series of topographs under the conditions of (a) 500 V for 1 h, (b) 2500 V for 1.5 h, (c) switching off and (d) 18 h after switching off. The topograph (a) is essentially identical to the topograph of the original specimen. The topograph (c) shows clearly the growth layers and other defects but with less contrast than in topograph (b). In the last one (d), the contrast returns to its original value. The decay time depended on the specimens and the time for which the specimen was kept at high voltage.

4. Discussion and conclusions

(a) The origin of the contrast changes

Obviously, the contrast in diffraction topographs is caused by strain gradients. The changes in the contrast, therefore, must be due to the converse piezoelectric effect in the present case and electrostriction in general. The strain induced by the electric field must be locally different from position to position, even though the field is homogeneous on a macroscopic scale, because the original specimen is already inhomogeneously strained around the defects on a scale of a few microns. The relatively slight changes in the contrast around defects such as C', I and S can be explained by assuming a homogeneous average field.

The remarkable changes in the contrast of the growth layer (L) and cell boundaries (C), however, seem difficult to explain in terms of such a homogeneous field. Presumably, an inhomogeneous field will be induced by applying the voltage to the crystal. By electrostriction, then, an inhomogeneous strain is introduced and this causes changes in the contrast.

The field which causes the diffraction contrast is not however only the applied static field itself nor the instantaneously induced inhomogeneous field. The fact that the contrast is observed for about 10 h after
switching off indicates that some residual field remains during the relaxation time. It seems that ionic or electronic charges are induced within the crystal and they remain with an inhomogeneous distribution, particularly along the growth layers and cell boundaries. The diffusion from the electrodes of silver ions, which remain as neutral atoms, seem unlikely to be the cause of the remarkable change of contrast, because the relaxation time is rather short for the diffusion of neutral atoms at room temperature. The definitive explanation, however, must wait for further detailed studies.

(b) Application to the study of crystal imperfections

The present method is very useful for studying imperfections in synthetic and natural quartz. As shown in Figs. 3 and 4, the growth layers and cell boundaries become observable simply by application of the electric field. In the following, a few observations which are relevant to crystal growth are described.

In Figs. 3(b) and 4(b), it turns out that the growth layers in the Z growth sector continue the growth layers in the S and $\pm X$ sectors. A similar example is seen in Figs. 6(a) and (b). Curiously, in Fig. 6(a) no
growth layer is observed in the $Z$ growth sector. By applying a voltage to the specimen, however, one can see that a growth layer connecting the growth layers in the $s$ and $-X$ sectors was really formed at some stage of the growth process.

Another interesting thing is that some areas of weak contrast, appearing as line defects in Fig. 3(a), are revealed to be cell boundaries in Fig. 3(b). Thus, it is concluded that the main part of the $Z$ growth sector has a cell structure which could hardly be deduced if only Fig. 3(a) were available.

Examination of the details of the growth layers after the contrast has been intensified reveals that the growth layers between the cell boundaries are convex toward the growth direction. It is clear that the network of grooves often observed on the cobble surface of the $Z$ sector is due to the cell boundaries (Lang & Miuscov, 1967; Homma & Iwata, 1973).

In conclusion, it is expected that the present technique of intensifying the diffraction contrast increases the usefulness of Lang's method.

References


