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Nondestructive Evaluation of Synthetic Quartz Crystals

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Abstract – On synthetic quartz crystals, quantitative evaluation about the correlation between peaked cobbles and dislocations with screw component was investigated.

This correlation provides capability of nondestructive evaluation for dislocation density by cobble density. The authors developed a system to observe the cobbles effectively. It can be used for industrial field.

Ⅰ.INTRODUCTION

It is known that the peaked cobbles on Z-surface of as-grown synthetic quartz crystals connect dislocations and etch-channels. In the F.C.S. 1996, Kurashige et al. reported the quantitative evaluation about correlation between cobbles and dislocations, and also between dislocations and etch-channels. The correlation suggested the capability of nondestructive evaluation for defect density in synthetic quartz crystal. [1]

In this study, the authors investigated the correlation between cobbles and dislocations again by different points, taking account of the scattering data in the previous study [1]. Cobbles and dislocations were correlated clearly. The character of dislocation type which forms cobble was discussed through an observation of detailed cobble texture by differential interference contrast microscopy.

Additionally, nondestructive evaluation system of synthetic quartz crystals and its method are proposed.

Ⅱ.EXPERIMENTAL PROCEDURE

A. *Sample Selection*

Ten Z-bars were prepared from industrially produced synthetic quartz crystals for sample bars as shown in the figure 1. It was paid attention for sample bars to have variety of the Z-surface texture from smooth to rough. These Z-bars' growth conditions were the same, and the specific data appears in the table 1.

These growth conditions are normally applied to the Z-bars sized 22 to 26 mm in Z-dimension.

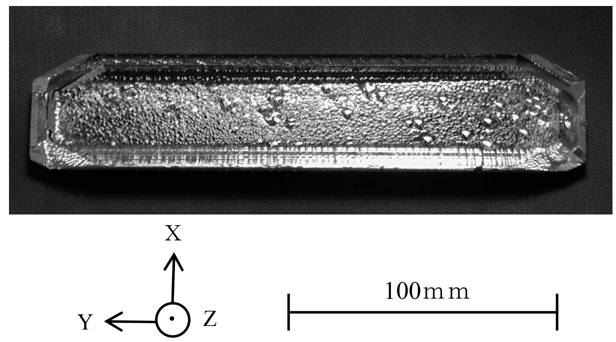

Figure 1. Z-bar of synthetic quartz crystal

Table 1. Specific data of sample crystal bars

Seed dimensions	X=31.0mm Y=210.0 mm Z=1.0mm
Growth Temperature	330 deg.C
Pressure	100Mpa
Solution	1.0 N NaOH doped Li salt
Growth duration	73 days
Autoclave	ϕ 600 × 15m

B. *Cobble Density*

In the previous study, Kurashige et al. defined cobble as "large-cobble" that was 3mm in diameter with peaked shape. But the definition of size restricted evaluation.

The authors used differential interference contrast microscopy (DICM). Using DICM, the Z-surface texture appears clearly. A small peaked cobble and large peaked cobble can be seen on the Z-surface. We gave an eye to peaked cobble irrespective of size as shown in the figure 2, and counted only peaked cobble to evaluate cobble density.

These specimens were cut from as-grown Z-bars, as shown in the figure 3(a)-1, 3(a)-2. Two growth surface sides were decided to be different sample bar, because if seed surface condition on each side changes, the number of dislocations and cobble texture will be different on the each growth Z-region. Inclusion on the seed surface affects the generation of dislocations. Twenty sample bars were cut from ten Z-bars.

Kagami and Takahashi suggested that increase of dislocation make change of cobble texture. [2] Two specimen plates for the cobble density evaluation were cut from one sample bar. Cobble density was total cobble number on the Z-surface in square centimeters. The cobble density of each sample bar was defined as average value of the two specimen places which is 5 cm square centimeter (2.5 x 2.0) in area on the Z-surface.

In this paper 'Cobble' means cobble which has peaked shape after here.

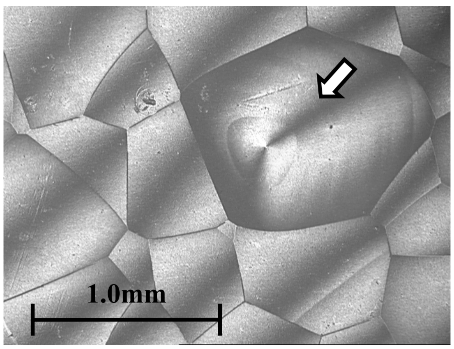

Figure 2. Differential interference contrast micrograph of Z-surface: The peaked cobble is indicated by the arrow

C. *Dislocation Density*

Dislocation density was observed by X-ray topography. The topographies were taken with $MoK\alpha_1$ reflected by (0003) and $(11\bar{2}0)$. The contrast of dislocations in growth Z-region by (0003) reflection and $(11\bar{2}0)$ reflection means the dislocations which have screw component and edge component, respectively.

Two specimen plates were cut from one sample bar perpendicularly to the seed plate, and etched lightly in ammonium bifluoride NH_4HF_2 after lapping to a thickness of 1.0mm using green silicon carbide #1000, as shown in the figure 3(b)-1, 3(b)-2.

The number of dislocations were counted at the middle between the seed surface and as-grown surface of growth Z-region on X-ray picture. The dislocation density was evaluated with the dividing the number of dislocations by cross section. The dislocation density of the each sample bar was defined as average value of the two specimen plates in similar cobble density evaluation.

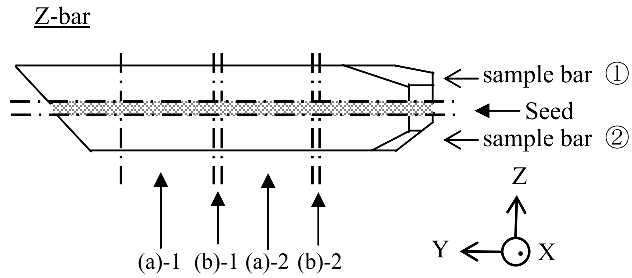

Figure 3. The location of specimen slice
(a)-1,(a)-2 specimens for cobble density evaluation
(b)-1,(b)-2 specimens for dislocation density evaluation

III.EXPERIMENTAL RESULT

A. *Cobble and Dislocation*

The correlation between cobble density and the dislocation density with screw component was plotted about twenty sample bars as shown in the figure 4. There are only nineteen points in this figure, lack in one point, because 2 points overlap each other. It is observed from this plot that the sample bar which has higher cobble density tends to have higher dislocation density.

The figure 5(a)-1 and 5(a)-2 show the X-ray topograph of Y-plate by (0003) reflection and $(11\bar{2}0)$ reflection, respectively. The figure 5(a)-1 was taken at the same part of figure 5(a)-2. The figure 5(b)-1 shows the differential interference contrast micrograph from the Z direction of the same sample plate taken topography. The figure 5(b)-2 shows enlarged photograph of the cobble in the figure 5(b)-1.

Dislocation "A" in the figure 5(a)-1, 5(a)-2 existing at cobble did not change contrast in the both cases reflected from (0003) and $(11\bar{2}0)$.

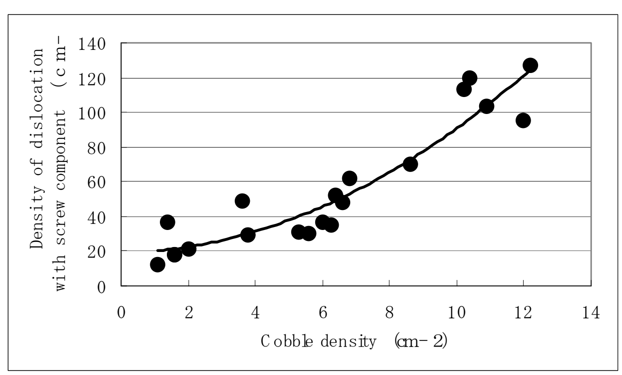

Figure 4. The correlation between cobble density and density of dislocation with screw component

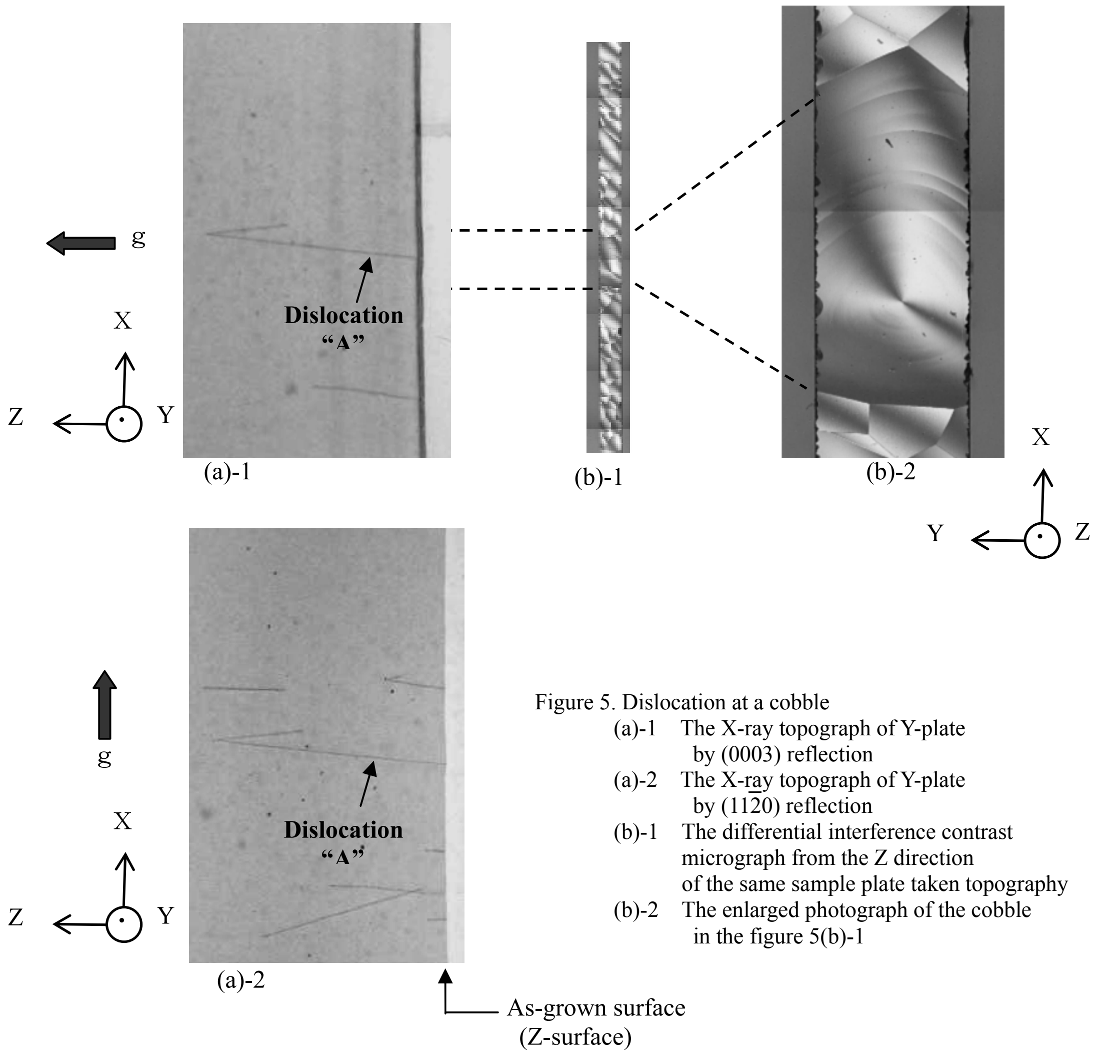

Figure 5. Dislocation at a cobble
(a)-1 The X-ray topograph of Y-plate by (0003) reflection
(a)-2 The X-ray topograph of Y-plate by $(11\bar{2}0)$ reflection
(b)-1 The differential interference contrast micrograph from the Z direction of the same sample plate taken topography
(b)-2 The enlarged photograph of the cobble in the figure 5(b)-1

B. *Total dislocation and the dislocation with screw component*

The correlation between the density of dislocations which have screw component and total dislocation density is plotted in the figure 6. The dislocations with screw component plus the dislocations with edge component is the total dislocation.

It is observed from this plot that the sample bar which has higher density of dislocations which have screw component tends to have higher total dislocation density.

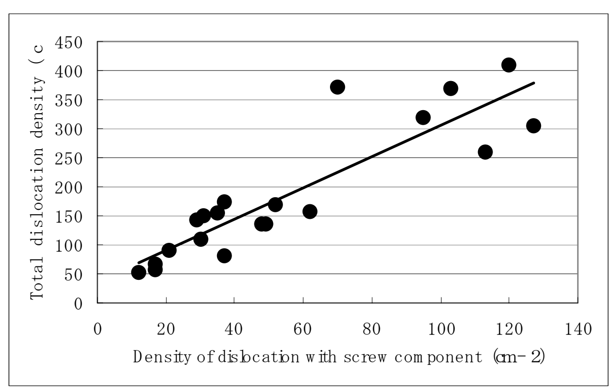

Fig6: The correlation between density of dislocation with screw component and total dislocation density

Ⅳ.DISCUSSION

A. *Peaked Cobble*

The dislocations in synthetic quartz crystals have been studied with many investigators by X-ray topography. It is known that the dislocations existing at the cobble did not change contrast in the both cases reflected from (0003) and $(11\bar{2}0)$. The figure 5 was the same result as many investigators' result. This means that these dislocations have both screw and edge components. Similar results are obtained at many other cobbles in this study.

The figure 7-A shows the differential interference contrast micrograph of the Z-surface in early grow phase. The figure 7-B shows enlarged photograph of the spiral growth hillock at the sign of arrow in the figure 7-A. A spiral growth hillock can be observed clearly in the figure 7-B.

The figure 2 shows the typical micrograph of the Z-surface of synthetic quartz crystal. The Z-surface is covered with cobbles and convex hills. The spiral growth has never started without screw dislocation. The comparison between the figure 2 and the figure7 suggests that the spiral growth hillocks in early grow phase correspond with the cobbles after growth.

B. *The correlation between cobbles and dislocations*

The authors presume that the formation of cobbles will concern screw component of dislocations according to the figure 2 and the figure 7. For this reason the authors investigated the correlation between peaked cobbles and dislocations which have screw component in this study. The dislocations which have screw component can be obtained using X-ray topography by (0003) reflection.

The previous study investigated about correlation between cobbles and the dislocations had edge component.

The correlation that is shown in the figure 4 reinforces above presumption.

The reason why the dislocation density was nearly ten times as high as the cobble density is thought that the number of dislocations were counted at the middle between the seed surface and the as-grown surface on X-ray picture. If the cobble is next to each other, these will integrate with increasing growth time. The cobble on the Z-surface decreases, but the existing dislocation at the cobble does not disappear. The dislocations can be counted on X-ray picture.

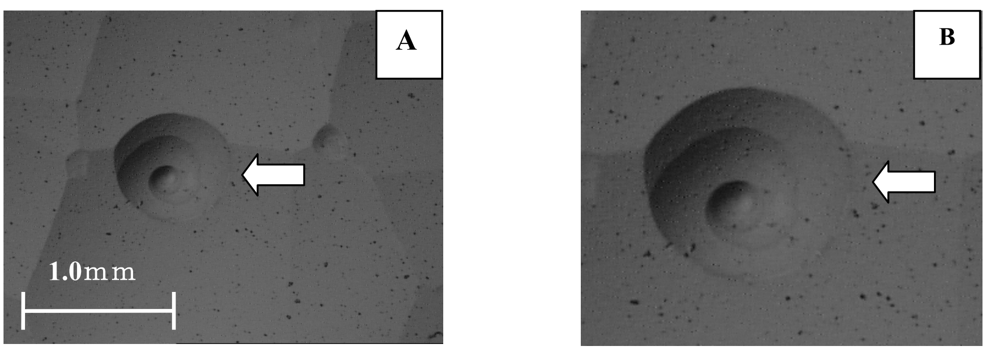

Figure 7. The spiral growth hillock on Z-surface of synthetic quartz crystal is indicated by arrow
A: The differential interference contrast micrograph of Z-surface of early growth, and the spiral growth hillock.
B: The differential interference contrast micrograph enlarged from the figure 7-A

C. *Presumption of total dislocation density*

Most studies have concluded that the character of dislocations in the Z-region of synthetic quartz crystals are generally all type of edge, screw, and combined both components. [3] [4] [5] et al.

The correlation between the density of dislocation with screw component and the total dislocation density is plotted in the figure 6.

Increasing of total dislocation density will proportionally provide increasing of the density of dislocations which have screw component. Therefore the sample bars which have higher total dislocation density can be estimated to show many cobbles formed by the dislocation with screw component on their appearance.

D. *Nondestructive evaluation*

Dislocations are often observed using techniques such as X-ray topography and etching method. In the latter technique, if a sample is etched in NH_4HF_2 solution, narrow etch-channels along dislocations will be seen with the unaided eye. However, we can not use those techniques as in-process quality checking, because both techniques are destructive evaluation.

In this study, the correlation between cobbles and dislocations which have screw component was investigated. Increasing of total dislocation density will provide increasing of the density of dislocations which have screw component, and the total dislocation density can be expected by the cobble density. Therefore we can evaluate dislocation density roughly of as-grown quartz by the cobble density evaluation, which is non-destructive, without cut and etch procedures.

A great number of nearby growth hillocks over the Z-surface integrate into bigger one with increasing growth time. The number of hillocks get to decrease, but peaked cobble stays in the Z-surface until the growth end. Growth units arrive on peak of cobble preferentially, this is thought to be provided by following reason. (1) A peak of cobble containing screw dislocation has higher free energy than other place. (2) Concentration gradient at a peak of cobble is higher than the other place.

Therefore we can evaluate the as-grown quartz by the cobble density.

E. *Nondestructive evaluation system*

When we evaluate the cobble density, it is not easy to distinguish the cobbles from the other smooth convex hills on the Z-surface with the unaided eye. That is why the synthetic quartz crystal is transparent.

If the Z-surface is observed using DICM, it is easy to distinguish these each other. However this method is impractical, because an observation view is so narrow.

The authors developed a nondestructive evaluation system which can be used for industrial field.

The figure 8 shows the construction of nondestructive evaluation system. The Z-surface of as-grown quartz is taken by CCD camera, and taken portrait of the Z-surface can be observed on the monitor after image processing. The cobble size, a number, combined both data and the cobble density can be displayed on the monitor.

The figure 9 shows the Z-surface which was taken by this system. The cobbles are indicated by the arrows in the figure 9. It is found that the distinction between the cobbles and smooth convex hills are easy. An observation view get to be tenfold compared to the view with microscopy.

The dislocation density could be evaluated as reproducible result using this system.

This system is designed to observe the cobbles simply without recourse to the way to light and various kinds of samples. It is important thing for a process control.

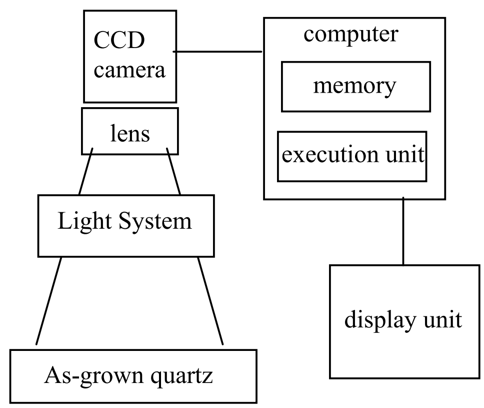

Figure 8. The construction of nondestructive evaluation system.

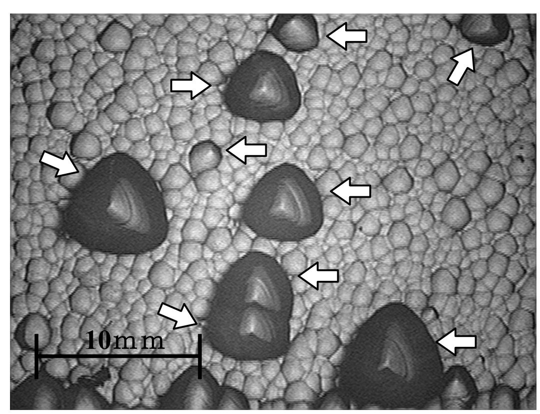

Figure 9. The Z-surface which was taken by nondestructive evaluation system
: The cobbles are indicated by the arrows

F. *The method of Nondestructive evaluation*

A practical example, using the nondestructive evaluation system as shown in the figure 8, is reported.

Firstly, evaluate the cobble density using this nondestructive evaluation system instead of DICM. Next, plot the correlation between the cobble density and dislocation density like the Figure 4.

The dislocation density having a screw component that corresponds to the cobble density is found. The total dislocation density increases in the proportion of the density of dislocations which have screw component. Consequently, evaluation of the dislocation density is obtained by evaluating cobble density.

Each of Z-growth size and seed size has to need the graph like the figure 4 to evaluate more precisely, because the cobble becomes bigger, and area of the Z-surface becomes smaller as r and R faces grow.

G. *Industrial Contribution*

Nondestructive cobble density evaluation can be used to a quality control of the seed crystals.

The dislocation density is evaluated by this system before as-grown quartz is sliced to seed crystals. When we choose only seed crystals which have low dislocation density, it will be possible to grow high quality quartz which have low dislocation density.

The dislocations in the seed crystals affect the grown crystals, because the dislocations propagate from the seed crystal surface generally. For this reason, the quality control of the seed crystals is important.

Additionally, for users who do not have costly X-ray topography cameras, this method may be usable as substitution for dislocation density estimation.

V .CONCLUSION

Quantitative evaluation about correlation between peaked cobbles and dislocations with screw component was investigated. This correlation provides capability of nondestructive evaluation for the dislocation density by cobble density.

Cobble observation gets to easy by the developed nondestructive evaluation system.

For example, this nondestructive evaluation method can be used to a quality control of the seed crystals.

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VII. REFERENCE

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