THE EVALUATION OF QUARTZ RESONATORS VIA X-RAY DIFFRACTION TOPOGRAPHY

K. G. Lipetzky and R. E. Green, Jr.
Center for Nondestructive Evaluation
The Johns Hopkins University
Baltimore, MD 21218

R. W. Armstrong
Department of Mechanical Engineering
University of Maryland
College Park, MD 20742

W. T. Beard, Jr.
Laboratory for Physical Sciences
College Park, MD 20740

INTRODUCTION

While quartz resonators have been the mainstay of the ultrasonics industry for some time, intricacies exist in the production of quality resonators and therefore fabrication remains somewhat of an art form. Recently the Johns Hopkins University Center for Nondestructive Evaluation has initiated a research program to investigate methods to analyze quartz single crystals for the purpose of assessing the relative crystal quality of the raw material as well as material which has been manufactured into resonators. One technique which has proven to be useful in a variety of single crystal inspection applications is x-ray diffraction topography. X-ray diffraction topography is the name given to several x-ray diffraction techniques which permit the imaging of strains and lattice misorientations associated with surface and internal defects as small as dislocations to be examined. Because the topographic techniques are based on Bragg diffraction from a periodic crystal, the images are extremely sensitive to crystal imperfections, strains, and rotations since any alteration to the interplanar position and spacing of the crystal will effect a corresponding change in the Bragg condition. The dynamical theory of interaction between transmitted and reflected wave fields at, or near to, the Bragg condition is intimately connected with spatial contrast in the topographic images. This paper describes experimental details of the x-ray diffraction topographic system used and shows topographic images illustrating the utility of the system as applied to quartz crystals in raw material and resonator form.

BACKGROUND

Conventional methods for examination of single crystal specimens rely on the Laue technique of x-ray diffraction whereby patterns of individual spots (reflections) are obtained from individual planes. In the Laue method, a continuous (white radiation) x-ray source is utilized. The x-ray source is collimated (usually with a pin-hole collimator) and then incident upon the sample when the transmission configuration is used, or through a hole in a film followed by the sample when the back-reflection configuration is used. Figure 1 shows a schematic of the Laue x-ray diffraction method in both the (a) transmission and (b) back-reflection configurations and Figure 2 shows actual Laue transmission and back-reflection x-ray diffraction patterns obtained from a quartz crystal. Each of the spots seen on the film corresponds to diffraction from a particular set of "parallel" lattice planes within the quartz crystal and contains information about the perfection of the crystal lattice. The Laue method is an excellent means to determine the crystallographic orientation of a crystal although it is not very conducive to the evaluation of the overall perfection of a crystal due to the small size of the "probe" x-ray beam and resulting diffraction spots. While it is possible to use a larger collimator in order to investigate a large region on a single crystal specimen, overlapping of the diffraction spots may occur (particularly when a short film to specimen distance is used, i.e. 3 cm). By using a monochromatic x-ray source it is possible to eliminate the overlapping of the diffraction spots, however, beam divergence associated with the Laue method remains. One technique which has proven to be useful in a variety of single crystal inspection applications is x-ray diffraction topography [1-10].

EXPERIMENTAL PROCEDURE

X-ray diffraction topography is the name given to several x-ray diffraction techniques which permit close examination of possible spatial variations in diffracted intensity over relatively large crystal areas that can be matched on a point-by-point basis with the physical crystal dimensions. It must be emphasized that the term topography literally means "to describe a place" (topos = place, graphein = to write) and the diffraction

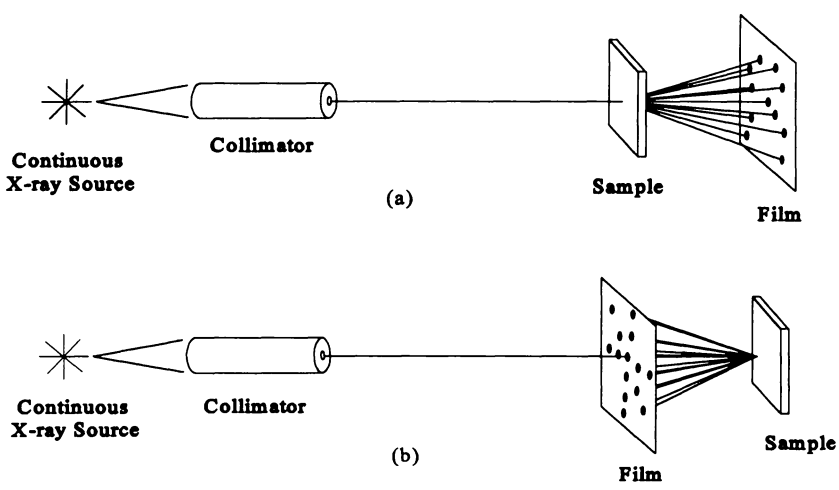
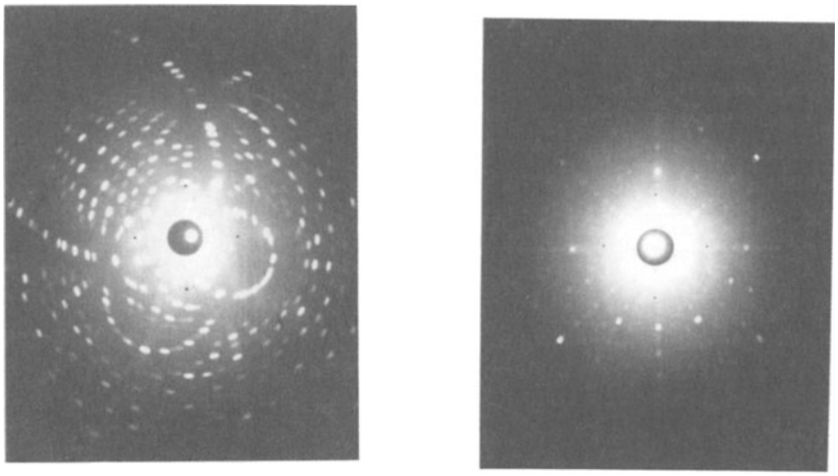


Figure 1. Schematic diagram of the Laue x-ray diffraction method in the (a) transmission configuration and the (b) back-reflection configuration.



(a) (b)

Figure 2. Typical Laue x-ray diffraction patterns obtained from a quartz crystal in the (a) transmission configuration and the (b) back-reflection configuration.

information which is obtained by x-ray topographic methods may be bulk (transmission) or surface (back-reflection) in nature. The topographic techniques based on Bragg diffraction from a periodic crystal are extremely sensitive to imperfections and strains in the crystal, since any alteration to the interplanar spacing of the crystal will effect a corresponding change in the Bragg condition. In the topographic set-up used in this investigation, called asymmetric crystal topography (ACT) [11], a slit collimated white radiation x-ray source was incident upon a high quality asymmetrically cut silicon crystal. The silicon crystal served as both a monochromator of K_α and possibly K_β wavelengths, and a beam expander, resulting in an x-ray beam of approximate dimensions 2 inches high by 3/4 inch wide.

A crystal specimen of interest was mounted in a goniometer on a Newport rotation stage and placed in the path of the monochromated and expanded x-ray beam. The sample was rotated in the monochromated and expanded x-ray beam until a Bragg condition was satisfied for some plane reflection as detected using an image intensifier with a fluorescent screen faceplate placed near the specimen. The beam diffracted from the specimen crystal and captured by the image intensifier is ensured to be monochromatic when the stringent simultaneous diffraction conditions of the first silicon crystal and the quartz sample crystal are satisfied. Diffraction images of interest were permanently recorded on x-ray film (Kodak occlusal DF-50 dental film) with exposure times averaging a half-hour for back-reflection topographic images and five hours for transmission topographic images obtained through plates of about 1/16 inch thickness. Due to the relatively weak x-ray source available in the laboratory (copper target tube operated at 50 kV and 32 mA), as well as the thickness of the majority of the specimens examined, the back-reflection configuration was that which was predominantly used. However, in the examination of the thinner quartz resonators, both transmission and back-reflection configurations were utilized. A schematic of the ACT system showing positions of the highly perfect, asymmetrically cut silicon first crystal (monochromator and beam expander), the second crystal (specimen under investigation), and the x-ray image intensifier (for direct real-time viewing of topographic images) is shown in Figure 3.

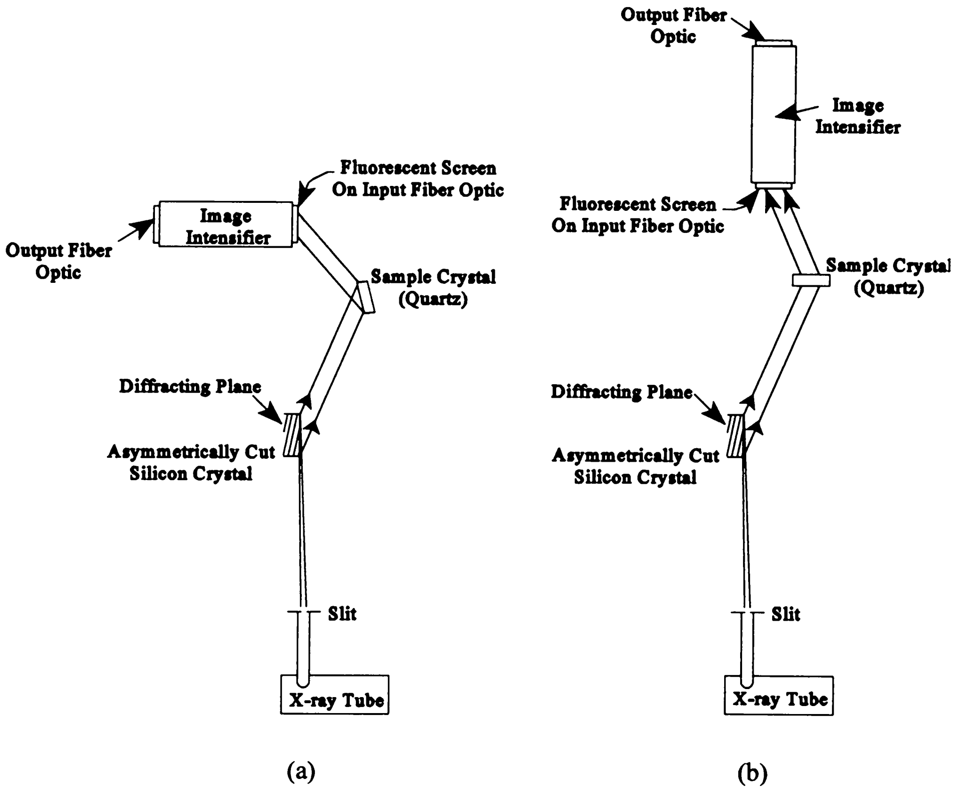


Figure 3. Schematic of the asymmetric crystal topography (ACT) system in the (a) back-reflection configuration and the (b) transmission configuration.

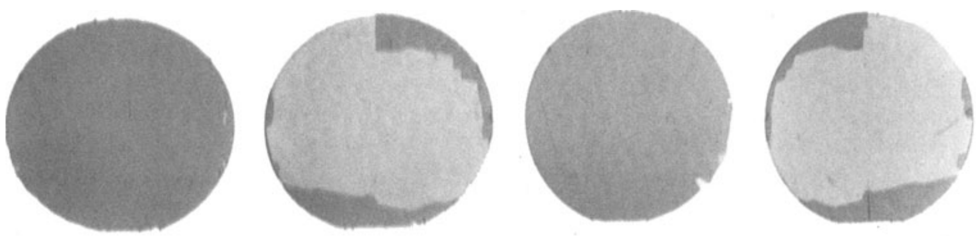
In the ACT technique thus employed, each individual topographic image is essentially a large Laue "spot" generated by diffraction from a particular set of "parallel" lattice planes covering a large area of a given sample crystal. In this investigation the incident monochromated and expanded x-ray beam illuminated the entire quartz sample (i.e. the size of the quartz crystals both in the raw material form and the resonator form were smaller than the dimensions given above for the monochromated and expanded x-ray beam) unlike conventional Laue pin-hole techniques, and because of the special beam expanding monochromizing silicon crystal, this large incident beam experienced minimal horizontal divergence.

EXPERIMENTAL RESULTS

The first application of the ACT system to quartz crystals was to determine if the x-ray topographic system could distinguish between a "good" resonator versus a "bad" resonator and furthermore, the possible cause(s) for failure of the "bad" resonator. Two quartz resonators each of 5/8 inch diameter and approximately 1/16 inch thickness were examined in the ACT system. A feature which was present on each sample, a major flat (for crystallographic purposes), was used as a fiduciary marker to determine whether a sample was in the up or down position. Since there was no way to distinguish the front from the back of a given sample, an arbitrary designation was made.

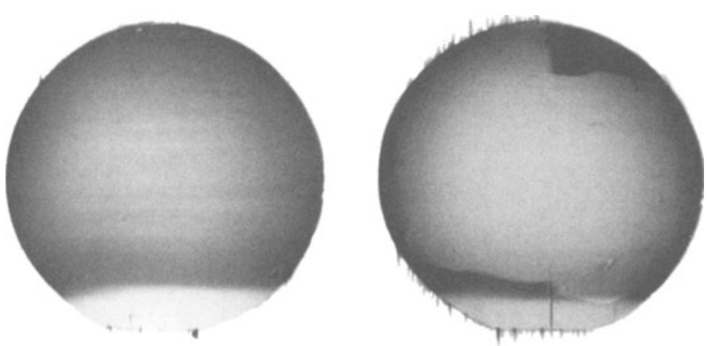
Each sample was examined individually using the back-reflection configuration of the ACT system. In order that direct comparisons could be made between the front and back of a given sample and between the two different samples it was necessary to ensure that the same Bragg reflection was selected. This was done by recording a selected diffraction pattern from one sample and then, without changing the Bragg angle, removing the first sample and either flipping the sample over to examine the other side or replacing it with the second, making sure that the position of the fiduciary flat coincided. Minor adjustments in the Bragg angle ($< 1°$) needed to be made occasionally in order to optimize the diffraction conditions and for a given Bragg condition a total of four topographs were obtained ("good" sample/front, "good" sample/back, "bad" sample/front and "bad" sample/back). Figure 4 shows typical topographic images acquired in the back-reflection configuration from the two samples.

As can be seen in Figure 4, distinct differences can be discerned between the topographs of the "good" sample versus those acquired from the "bad" sample; the most notable being the jagged region (of darker contrast) which appears around the circumference of the "bad" quartz resonator. From experience, this region is known to be indicative of twinning which has occurred within the sample and is the most probable reason for the piezoelectric failure



(a) (b) (c) (d)

Figure 4. Back-reflection, x-ray diffraction topographs obtained from a (a) "good" quartz resonator (front view), (b) "bad" quartz resonator (front view), (c) "good" quartz resonator (back view) and (d) "bad" quartz resonator (back view).



(a) (b)

Figure 5. Transmission, x-ray diffraction topographs obtained from a (a) "good" and a (b) "bad" quartz resonator.

mechanism observed in the "bad" sample. The dimpled appearance which is observed in all the topographs is termed "cobbling" in the quartz growth industry. Cobbling is a consequence of cells of dislocations forming during the crystal growth process and normally is revealed on the (0001) surface.

For comparative purposes the same two quartz samples were examined using the transmission configuration of the ACT system. Because the dimensions of the "probe" x-ray beam (monochromated and expanded) were larger than the sample being examined, a sample mount of lead was designed to block the monochromated and expanded x-rays except for those which were incident on the sample. Again, each sample was examined individually; and, in order that direct comparisons could be made between the two samples, it was necessary to ensure that the same Bragg reflection was selected. This was done by recording a selected diffraction pattern from one sample and then, without changing the Bragg angle, removing the first sample and replacing it with the second, making sure that the position of the fiduciary flat coincided. As before, minor adjustments in the Bragg angle ($< 1°$) needed to be made occasionally in order to optimize the diffraction conditions. Because transmission topographic techniques examine the volume (bulk) of a given sample crystal it was only necessary to take two topographs for a given Bragg condition ("good" sample and "bad" sample). Figure 5 shows typical topographic images acquired in the transmission configuration from the two samples. As with the back-reflection topographs, distinct differences between the "good" resonator and the "bad" resonator can be seen.

Another application of the ACT system to quartz crystals was to evaluate the overall crystal quality of two samples supplied after growth by the Suzuki method [12]. Figure 6 shows an identified Laue back-reflection pattern from a sectioned ($01\bar{1}0$) crystal plate. In brief, the Suzuki method is a hydrothermal crystal growth process (with a seed crystal) designed at producing cultured quartz crystals with large areas of low defect population [13]. The two samples were grown in the same manner, with the exception that the

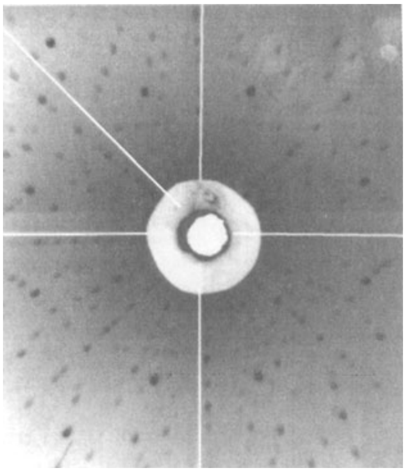
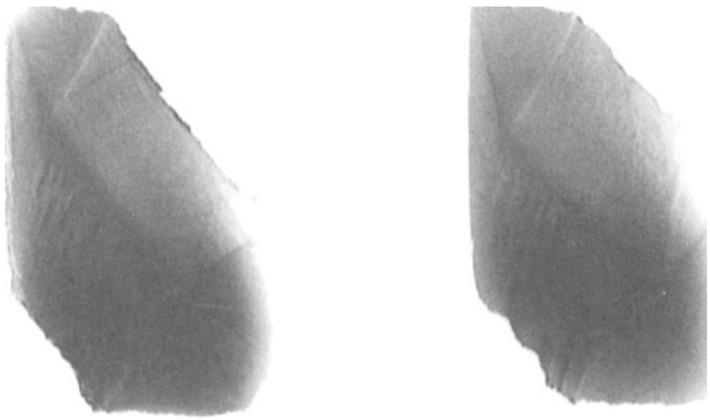


Figure 6. Back-reflection, Laue x-ray diffraction pattern from a sectioned ($01\bar{1}0$) cultured quartz crystal grown by the Suzuki method.



(a) (b)

Figure 7. Back-reflection, x-ray diffraction topographs obtained from two different cultured quartz crystals grown by the Suzuki method.

crystallographic cuts of the seed crystals were different. Due to the thickness of the samples (approximately 1/4 inch), it was only possible to examine the samples topographically using the back-reflection configuration. Figure 7 shows typical topographic images acquired from the two samples. Diffraction contrast from the seed crystal can be seen in both samples (rectangular shaped region in the "center" part of the crystal lying at an approximate 45° angle, upper left to lower right) along a $[2\bar{1}\bar{1}\bar{1}]$ direction and distinct growth sector boundaries consistent with previous observations [12] are recognized as well. The $[\bar{2}110]$ direction is vertical in Figure 7. It appears that an additional growth sector boundary is present in sample (a) compared to (b). [See the top right portion of sample (a), Figure 7]. In this case, the occurrence of new growth sector regions, designated ξ-regions, are of interest because of impurity segregation influences that produce strong lattice distortions. The case provides an illustrative example of the power of x-ray topography to reveal atomic scale effects.

CONCLUSIONS

X-ray diffraction topography can easily detect and image the presence of defects within a crystal, making it a powerful nondestructive evaluation tool for characterizing industrially important single crystal specimens such as quartz. Whether looking for gross defects (crystallographic misorientations) or smaller defects such as dislocations or even atomic (point defect) scale influences, the x-ray diffraction topographic techniques are well suited to a variety of applications. In the examples shown, x-ray topography was utilized to distinguish between "good" and "bad" resonators and evaluate the crystalline perfection of cultured, raw quartz samples produced in a new seeded crystal growth orientation. Further use of x-ray diffraction topography would aid in the optimization of such crystal growth and processing of quartz as well as other single crystal applications.

ACKNOWLEDGEMENTS

This work was supported by the Center for Nondestructive Evaluation at The Johns Hopkins University. The authors wish to thank Jonathan W. Foise of Sawyer Research Products for providing some of the quartz samples which were utilized in this study.

REFERENCES

1. W.T. Beard, K.G. Lipetzky and R.W. Armstrong, "Dynamical Effects in High Resolution Topographic Imaging of Electronic Devices, (to be published *in Advances in X-ray Analysis*, **41**).
2. Kirsten G. Lipetzky, Robert E. Green, Jr., and Paul J. Zombo, "Development of X-ray Diffraction Methods to Examine Single Crystal Turbine Blades," (to be published in *Nondestructive Characterization of Materials*, **VIII**).
3. R.W. Armstrong, W.T. Beard, K.A. Green and X.J. Zhang, "High-Resolution Imaging of Electronic Devices Using Line Modified-Asymmetric Crystal Topography (LM-ACT)," *Il Nuovo Cimento* **19 D** (2-4), 147 (1997).
4. W.T. Beard, Jr., K.A. Green, X.J. Zhang and R.W. Armstrong, "High Resolution Imaging of Electronic Devices via X-ray Diffraction Topography," *Applied Physics Letters*, **69**(4), 488 (1996).
5. K.A. Green, W.T. Beard, X.J. Zhang, and R.W. Armstrong, "Application of Line Modified-Asymmetric Crystal Topography for Qualitative and Quantitative Evaluation of Integrated Circuits," *Advances in X-ray Analysis*, **38**, 227 (1995).
6. W.T. Beard, Jr., K.A. Green, X.J. Zhang and R.W. Armstrong, "In-Depth Resolution of Integrated Circuits via X-ray Based Line Modified Asymmetric Crystal Topography," *1994 IEEE International Reliability Physics Proceedings*, IEEE Catalog No. 94CH3332-4, 425.
7. Kirsten A. Green and Robert E. Green, Jr., "Application of X-ray Topography for Nondestructive Inspection of Industrial Materials, " *Review of Progress in Quantitative Nondestructive Evaluation*, **13**, 571 (1994).
8. Kirsten A. Green and Robert E. Green, Jr., "Nondestructive Characterization of Metals Subjected to High-Power Ultrasound," *Nondestructive Characterization of Materials* **VI**, 147 (1994).
9. John M. Winter, Jr., Robert E. Green, Jr., and Kirsten A. Green, "Application of Synchrotron and Flash X-ray Topography to Improved Processing of Electronic Materials," *Advances in X-ray Analysis*, **35A**, 239 (1992).
10. Kirsten A. Green and Robert E. Green, Jr., "Application of X-ray Topography to Improved Nondestructive Inspection of Single Crystal Turbine Blades," *Proceedings of the 16th Symposium on NDE* (Southwest Research Institute, San Antonio, Texas), 13 (1987).
11. W. J. Boettinger, H. E. Burdette, M. Kuriyama and R. E. Green, Jr., "Asymmetric Crystal Topographic Camera," *Rev. Sci. Instru.*, **47**, 906 (1976).
12. Armando H. Shinohara and Carlos K. Suzuki, "Study of S- and ξ-Bar Synthetic Quartz by X-ray Topography," Proceedings of the IEEE International Frequency Control Symposium, 72 (1996).
13. G. R. Johnson and J. W. Foise, "Quartz," *Encyclopedia of Applied Physics*, **15**, 365 (1996).