X-Ray Extinction in Piezoelectrically Oscillating Crystals

GERALD W. FOX AND WALTER A. FRASER, *Physics Department, Iowa State College*

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The original work of Fox and Carr on the effect of piezoelectric vibrations of quartz crystals on the Laue patterns has been continued and extended to include tourmaline and Rochelle salt. The effect of different surface conditions on the Laue patterns has also been studied. The results obtained for all three types of crystals were similar, though the effects were most pronounced in quartz. Quartz is undoubtedly the best material for the study of x-ray extinction in piezoelectric crystals. The increased blackening of the spots in a Laue pattern resulting from piezoelectric oscillation is explained as a consequence of decreased extinction brought about by the warping of the lattice planes during the vibration.

INTRODUCTION

IN 1931,[1] it was observed that the intensity of the Laue diffraction patterns of quartz was greatly increased when the crystal slip was oscillating piezoelectrically. Since that time, a number of investigators[2–11] have studied various phases of the general problem of the reflection of x-rays from crystals subjected to mechanical or piezoelectric strain. The results reported here further contribute to an understanding of the relations between x-ray reflection, surface conditions, and piezoelectric oscillation in crystals.

EXPERIMENTAL

(1) The law of blackening

The law of photographic blackening for the x-rays used in this experiment was determined in the usual way. The opacity of the spots was measured by the microphotometer and the density of each was calculated and plotted against the logarithm of the corresponding time. The blackening proved to be linear above a density of 0.3.

(2) The effect of piezoelectric oscillation on Laue patterns

The increased blackening of the spots in a Laue pattern which results from piezoelectric oscillation of a quartz specimen is very striking, but, for tourmaline and Rochelle salt, it is barely noticeable. The examination of a large number of corresponding pictures involving specimens of quartz, tourmaline and Rochelle salt, oscillating and nonoscillating, leads to the following observations.

(a) The pattern of the oscillating specimen is in every case uniformly increased in intensity over that of the nonoscillating specimen.

(b) Where the crystal slip is thick enough to show double spots, each spot of a pair appears to have the same intensity. Fig. 1 shows typical microphotometer traces of symmetrically located pairs of spots in the pattern produced by a Y-cut quartz plate.

(c) In every instance, individual spots in the Laue patterns are better resolved and sharper for the nonoscillating case. This applies to all

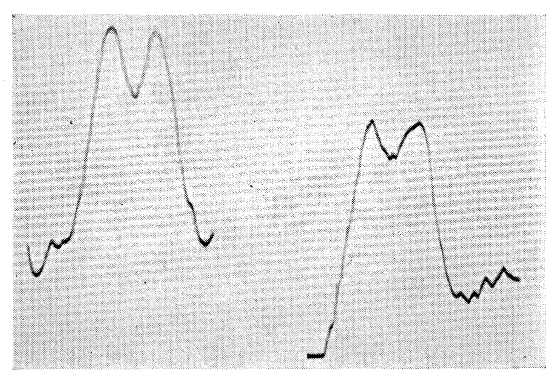

FIG. 1. Microphotometer traces of double spots.

[1] G. W. Fox, and P. H. Carr, Phys. Rev. **37**, 1622 (1931).
[2] C. S. Barrett, and C. E. Howe, Phys. Rev. **39**, 889 (1932).
[3] M. Y. Colby, and S. Harris, Phys. Rev. **42**, 733 (1932).
[4] M. Y. Colby, and S. Harris, Phys. Rev. **43**, 562 (1933).
[5] J. M. Cork, Phys. Rev. **42**, 749 (1932).
[6] G. W. Fox, and J. M. Cork, Phys. Rev. **38**, 1420 (1931).
[7] S. Nishikawa, Y. Sakisaka and I. Sumoto, Phys. Rev. **43**, 363 (1933).
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[9] Y. Sakisaka, Jap. J. Phys. **4**, 171 (1927).
[10] Y. Sakisaka, Proc. Phys.-Math. Soc. Japan **12**, 189 (1930).
[11] Y. Sakisaka, and I. Sumoto, Proc. Phys.-Math. Soc. Japan **13**, 211 (1931).

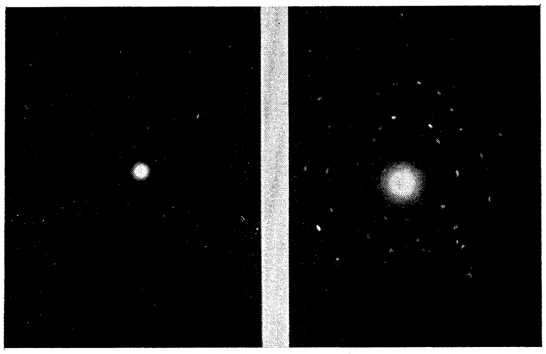

FIG. 2. Laue photographs of quartz, nonoscillating (A), oscillating (B).

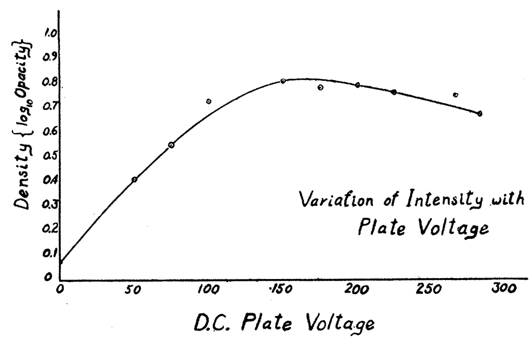

FIG. 3. Density of Laue spots *vs.* voltage on crystal plate.

crystals studied: quartz, Rochelle salt, tourmaline. The boundaries of the spots appear fuzzy and somewhat indistinct when the crystal is oscillating, and, for thick quartz specimens in which complete resolution of the double spots occurs when the plate is not oscillating, a slight additional darkening appears between the spots of a pair under conditions of oscillation.[12]

(d) A more complete pattern results when the crystal is oscillating. This does not mean that any additional spots appear for the oscillating case but that spots representing high orders which ordinarily require long exposures, come out in a shorter time.

(e) A curious observation is that, when a specimen is oscillating, the central spot caused by the undeviated primary beam appears to be more intense. At least the image covers a larger area on the film than in cases where the specimen is not oscillating (Fig. 2). This has been observed for all the crystals studied.

(3) The effect of amplitude of vibration on the pattern of the oscillating crystal

In the preliminary investigation of Fox and Carr,[1] a change in the intensity of the Laue pattern was observed when the piezoelectric amplitude of oscillation of the diffracting crystal was altered. This was investigated more thoroughly through a series of exposures in which the input to the x-ray tube (75 kv, 13 mils) and the time of exposure were constant but the d.c. voltage on the plate of the pliotron was changed.

To insure uniform treatment, the exposures were all developed, and likewise fixed, simultaneously. The opacities of certain corresponding spots in the patterns were determined by the microphotometer and the densities were plotted against the corresponding plate voltages. Fig. 3 shows the data from a series of such exposures. It may be observed that in all cases a maximum exists. Beyond the maximum, the amplitude of crystal oscillation decreases, probably because of partial short circuiting resulting from the high potentials developed across the crystal.

(4) The effect of etching the crystals

For this problem, a plate of quartz and a plate of Rochelle salt were ground with No. 400 carborundum and Laue pictures were taken for each specimen under oscillating and nonoscillating conditions. The quartz plate was then etched for 48 hours in a three percent solution of hydrofluoric acid and the Rochelle salt plate was placed for a short time in water. Laue pictures were then taken of the etched crystals oscillating and nonoscillating. The quartz plate was again etched for the same period and pictures taken. This process of etching and picture taking was repeated for the quartz until the total period of etching reached two weeks. Photomicrographs of the surface of the plate were made at intervals so that the smoothing process could be observed. The results of the study are as follows:

(a) Etching does not change any of the relationships previously observed for the unetched quartz crystal.

[12] C. C. Murdock, Phys. Rev. 45, 117 (1934).

(b) In the case of Rochelle salt, the Laue photographs of the etched crystal oscillating show slightly increased intensity and a more complete pattern with better spot resolution than do those for the unetched crystal oscillating. For the nonoscillating Rochelle salt crystal, the intensity of the pattern for the unetched crystal is greater than for the etched crystal. There is, however, better resolution of individual spots and the pattern is more complete for the etched crystal.

(c) The effect of etching is more evident with quartz than with Rochelle salt. After the first period of etching, a great increase in intensity of the Laue pattern of the oscillating crystal is evident, and the pattern is more complete than that of the unetched oscillating crystal. Additional etching, however, results in no further increase but actually in a decrease of both the intensity and completeness of the patterns. The surfaces eventually become passive to further action of the acid. For nonoscillating quartz crystals, etching appears to bring about decreased intensity of the Laue pattern and to cause first an increase and then a decrease in the completeness of the pattern. The resolution of the individual spots is better for the etched surfaces, though the first period of etching produces most of the effect.

(d) Rochelle salt plates are so active that the effect of etching on their oscillating properties is not very apparent. The effect on quartz plates, however, is to increase considerably the strength of oscillation. Perhaps this treatment may find commercial application.

Discussion

The unusual diffraction of x-rays by oscillating piezoelectric crystals seems to be intimately tied up with the phenomenon of x-ray extinction and a plausible explanation of the results herein stated may be given on this basis.

By extinction is meant the unusually great absorption of particular frequencies in an x-ray beam of white radiation, the wavelengths and glancing angles of which are such that multiple reflections occur within each nearly perfect crystal fragment. The extent to which extinction occurs is dependent upon the size of the individual fragments.

An ideally perfect crystal is one in which the atoms throughout its entire extent are accurately placed in accordance with the requirements of an undistorted lattice. In such a perfect crystal, there should be no reflection of a particular x-ray wavelength from a set of planes until the grazing angle approaches the particular value given by the Bragg law ($n\lambda = 2d \sin \theta$), at which the reflection should be complete over an angular interval $\Delta\theta$, after which it again falls abruptly to zero. Quartz appears to approximate the conditions representing a perfect crystal.

The inner parts of any crystal, provided it is homogeneous throughout, must be more regular than the surfaces where unbalanced forces between the atoms of the lattice will produce distortion. Just how much crystal should be included in the term "surface" is not entirely clear, but it seems reasonable to include all the outside region where unbalanced atomic forces are evident. If a crystal is roughened with an abrasive, its surface is rendered more imperfect by virtue of the additional strains introduced. Under such conditions, the "surface" must extend deeper into the body of the crystal. If extinction occurs in quartz, one would expect little or no contribution to the spots in the Laue pattern except from the surface region. The formation of double spots is in agreement with this point of view. As a beam of white radiation strikes the first surface of the crystal, it encounters a mosaic of small crystallites and in these occur the diffraction characteristic of the lattice. Because of the strains present in this region, small variations of the lattice constants and tilt of the atomic planes will exist so that the various $d\lambda$'s selected from the incident beam are wider than would be the case under unstrained conditions. Proceeding into the crystal, the lattice becomes more regular, until at a depth of a few thousand layers at the most, the "perfection" characteristic of the unstrained lattice is reached. The resolving power of the lattice depends on the degree of this "perfection." In the ideal case, the width of the $d\lambda$'s selected by this region would be extremely narrow, though finite, and, for those fulfilling the Bragg conditions, complete reflections should occur. Thus, the higher the degree of the lattice perfection, the less x-ray energy will it be able to contribute to the Laue diffraction images. Even if the reflection should be complete for certain wavelengths, the spectral intervals are so narrow that the actual energy contributions would be quite negligible.

Under piezoelectric oscillation, a number of mechanical pressure waves travel through the crystal simultaneously and a standing wave pattern occurs. As a result, the effective spacing throughout the crystal will vary somewhat and the central part of the crystal which, under non-oscillating conditions, can reflect only a number of very narrow wavelength intervals, now is able to reflect wider ones and so can contribute considerably to the reflection. It seems evident that, because no additional spots appear on the patterns of oscillating crystals, the variation in grating constants must be small. The increased intensity of each spot in the Laue pattern thus results from a slight increase in the width of the $d\lambda$ representing that spot. The fact that the spots are always sharper in the patterns for the non-oscillating crystal bears out this idea.

It is not at all evident why the area covered by the undeviated beam should be so much larger and apparently more intense for the oscillating cases. Perhaps increased scattering power results from the oscillation, though this is only conjecture.

On the basis of the foregoing, one would naturally expect increased amplitude of piezoelectric oscillation to bring about increased blackening. As the amplitude increases, the regularity of the lattice is further distorted and this results in reduced extinction so that each spot in the diffraction pattern represents an increasingly wider $d\lambda$ selected from the incident continuous x-ray spectrum. The fact that, in neither tourmaline nor Rochelle salt is there any great increase in spot blackening with increased amplitude of oscillation, indicates that, in these crystals, extinction does not play as important a role as in quartz. One might conclude that these crystals are not as "perfect" as quartz.

Etching, by removing to some extent the irregularities existing on the surfaces of crystals, tends to increase the extinction effects and so has an opposite effect from that of piezoelectric oscillation. The great increase in spot blackening observed after the first period of etching of the quartz specimen can be attributed to the unusual increase in amplitude of oscillation resulting from such treatment. Since subsequent periods of etching produce no further increase in blackening but actually a decrease, one can say that, after a certain time of etching, further treatment produces no further increased amplitude of oscillation but does have an important effect in decreasing the random orientation of the surface, thereby increasing the effects of extinction and hence decreasing the intensity of the spots in the pattern.

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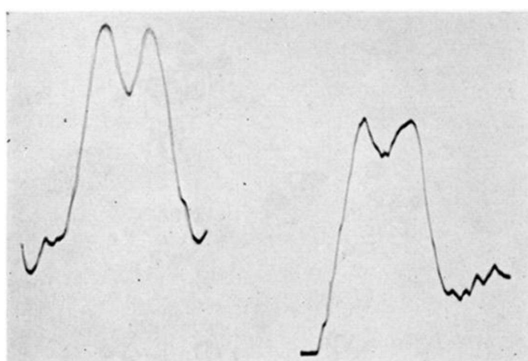

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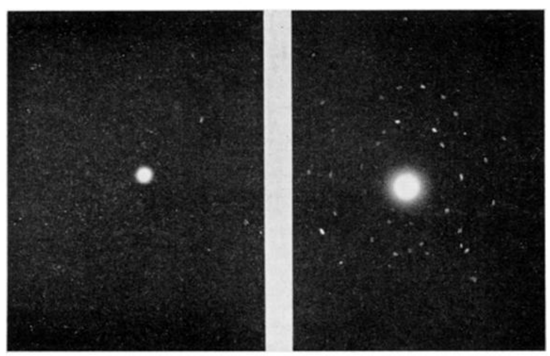

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