observed. As a matter of fact this experiment was first undertaken in the hope of detecting a rapid decrease in scattering at very small angles, due to statistical fluctuations. A system of Soller slits was constructed giving a maximum angular divergence of less than 15′ and it was possible to come within 30′ of the primary beam and yet not have the direct radiation enter the ionization chamber, but the scattered radiation was collected throughout such a small solid angle that measurement was hopeless and the experiment had to be revised.

Conclusion

The present experiment has shown that the effect of intermolecular interference is not negligible even in the case of a gas under moderate pressures. The scattering curves are very similar to those obtained for liquids and solids, the principal difference being that the main diffraction peak occurs at a smaller angle due to the greater mean molecular distance. We would thus conclude with Warren,[17] and with Gingrich and Warren[18] that such a decrease in scattering at small angles is due merely to the fact that there are a large number of scattering centers within a small space, rather than to any preferred grouping of the molecules, since the existence of such groups would be very unlikely in a gas so far from the liquid state.

In conclusion the author wishes to express his appreciation to Professor Arthur H. Compton for his interest in the work and the use of the Ryerson Physical Laboratory as well as for numerous suggestions and discussions during its progress.

An X-Ray Study of a Long X-Cut Quartz Crystal Vibrating Under the Transverse Piezoelectric Effect*

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The sixth order reflection from the (110) set of planes of a long X-cut quartz crystal of the $K\alpha$ radiation of Mo was photographed for the following conditions: (1) with the crystal non-oscillating, (2) with the crystal oscillating at its fundamental frequency under the transverse piezoelectric effect, and (3) with the crystal oscillating at its second harmonic frequency. From the microphotometer analysis of these lines it is shown that no increase in width of the lines reflected from the crystal oscillating at its fundamental frequency over the width of the lines reflected from the non-oscillating crystal is observed. This fact shows that there is no elastic deformation of the spacing of the (110) set of planes greater than 1.45×10^{-5} per unit dimension. This value is the limit of the sensitivity of the spectrograph and microphotometer used. However, an appreciable increase in intensity in the lines reflected from the crystal oscillating at its fundamental frequency over the lines reflected from the non-oscillating crystal is shown. A microphotometer analysis of Laue spots obtained by passing a rectangular beam of x-rays through the middle of the crystal when it was oscillating at its fundamental frequency confirms the prediction made by the authors in an article recently published in this journal. The results seem to indicate that any increase in intensity of the reflected x-rays from any portion of a quartz crystal produced by piezoelectric oscillations of the crystal is due almost entirely to a reduction of secondary extinction.

Introduction

Since Fox and Carr[1] reported a decided increase in the intensity of Laue spots obtained from a quartz crystal oscillating piezoelectrically along the direction of thickness of the plates caused by the longitudinal effect,[2] a number of investigators have conducted experiments with plates oscillating under this "thickness vibration" or more properly longitudinal effect; however, the authors are the only ones who have experimented with a long crystal vibrating under the transverse effect.[3,4] From the interferometer experiments of Osterberg[5,6] we now know that the strains in the plates

* This paper is an extract from the thesis presented for the degree of Doctor of Philosophy by Sidon Harris.

[1] Fox and Carr, Phys. Rev. **37**, 1622 (1931).

[2] The terminology here used is taken from *Piezoelectric Terminology* by W. G. Cady, Inst. Radio Eng. **18**, No. 12 (1930).

[3] Colby and Harris, Phys. Rev. **42**, 733 (1932).

[4] Colby and Harris, Phys. Rev. **43**, 562 (1933).

[5] Osterberg, Phys. Rev. **43**, 819 (1933).

[6] Osterberg, Rev. Sci. Inst. **5**, 183 (1934).

oscillating under the longitudinal effect are extremely complex and flexural, and that it is even doubtful if any standing waves parallel to the thickness dimension exist at all. It then seems probable that the large increased intensities of the Laue spots obtained under these conditions are produced by reduction of secondary extinction caused by the large inhomogeneous strains set up in the crystal, as first suggested by Langer.[7] Fox and Cork[8] failed to note any change in the lines reflected from the face of a plate oscillating under the longitudinal effect, but the authors[3] have succeeded in obtaining an effect on the lines reflected from the face of a long quartz crystal oscillating under the transverse effect. The failure of the Fox-Cork experiment to get any effect was due probably to the fact that the mode of oscillation of their plate was such that there existed approximately a node of pressure at the surface of the plate from which the x-ray beam was reflected. Hence, even if elastic deformations of the atomic plane spacings were taking place in other regions of the crystal, no widening of the lines obtained from this experiment could be expected. What reduction of secondary extinction that might have occurred at the point of reflection, caused by slight inhomogeneous strains in this region set up by the flexural vibrations, was probably obscured by an imperfect surface layer which had already reduced secondary extinction to nearly a minimum. In the authors' experiment,[3] a long crystal was used vibrating under the transverse effect; hence, a sinusoidal standing wave may be assumed with reasonable certainty. In the middle of the crystal where the x-rays were reflected, a loop of pressure existed clear through the crystal; hence, if any elastic deformations of the plane spacings were taking place, they would exist through this region of the crystal; also there would be a homogeneous strain through the crystal at this point, and therefore some displacement of the Zwicky blocks should be expected in the middle region of the crystal, if such blocks exist at all. Now if there is merely an elastic deformation of the spacings of the (1 1 0) set of planes and no rocking of the proposed blocks, the reflected line should show an increase in width but no appreciable increase in intensity. And if no appreciable elastic deformations are taking place, but a rocking of the blocks is taking place, then an increase in intensity and no appreciable increase in width of the lines should be expected. In the earlier experiment,[3] the authors detected both a widening of the lines and an increase in intensity. One purpose of the present work was to investigate further this result. By oscillating the crystal at its second harmonic, thereby producing a node of pressure through the middle of the crystal (the region from which the x-ray beam was reflected), it was found that the widening of the line is due solely to the jumping of the crystal in its mounting; however, the appreciable increase in intensity in the line reflected from the crystal when it was oscillating at its fundamental frequency was again observed. Thus it seems that the effect is due entirely to a reduction of secondary extinction produced by the rocking of the proposed Zwicky blocks.

In another paper,[4] the authors obtained practically no increase in the intensity of Laue spots obtained by passing an x-ray beam through the middle of the long crystal when it was oscillating at its fundamental frequency under the transverse effect; that is, no increase as far as the eye could detect. This result is due to the fact that in this experiment, homogeneous strain is present which does not reduce secondary extinction nearly as much as inhomogeneous strain which occurred in the other experiments in which the longitudinal effect was employed. Another purpose of this work was to examine these spots with a microphotometer, and see if the prediction made in the earlier paper[4] was correct.

Experimental Procedure

The crystal is an X-cut crystal of $100 \times 6 \times 2$ millimeter dimensions, with the 100 millimeter dimension parallel to the Y or mechanical axis, and the 2 millimeter dimension parallel to the X or electric axis. The electric field was applied across the X direction. The fundamental frequency of oscillation of the crystal is 27,400 cycles per second. With the spacing of the (1 1 0)

[7] Langer, Phys. Rev. **38**, 573 (1931).
[8] Fox and Cork, Phys. Rev. **38**, 1420 (1931).

set of planes given by Bradley and Jay[9] and the accepted value of the $K\alpha$ radiation of Mo, it was found that the sixth order reflection of this radiation from this set of planes could be obtained at a Bragg angle of 60.3°. By simple trigonometry it can be shown that the displacement ΔS of a line on the film caused by a change Δd in the spacing of the reflecting planes may be expressed as follows:

$$\Delta S = 2r \cdot \tan\theta \cdot \Delta d/d, \qquad (1)$$

where r is the film to crystal distance, which was 200 millimeters in this case, and θ is the usual Bragg angle. It was possible to detect a change of 0.01 millimeter in the width of a line from the microphotometer analysis; therefore a change as small as 1.45×10^{-5} per unit dimension of the spacing of the (1 1 0) set of planes could be detected.

The crystal U can be seen in its hard rubber mounting J in Fig. 1. When the crystal was oscillated at its fundamental frequency, the two top plates R' and R were connected to the binding post N, and the two similar bottom plates were connected in like fashion to the post N'. These terminals were connected across a large air condenser which was in a tuned circuit inductively coupled to the out-put of an amplifier, which was connected across the out-put of a standard Western Electric oscillator. When the crystal was oscillated at its second harmonic the plate R' was connected with the opposite bottom plate to one binding post, and the plate R was similarly connected with the other bottom plate to the remaining binding post.

Three exposures were made on the same film, one with the crystal not oscillating, one with the crystal oscillating at its fundamental frequency, and one with the crystal oscillating at its second harmonic frequency. Any effect on the line reflected from the crystal when it was oscillating at its second harmonic frequency may be attributed to the motion of the crystal as a whole in its mounting parallel to the 2 millimeter direction, as a node of pressure exists clear through the region from which the x-rays are reflected, and no deformations of the plane spacings or movements of the supposed blocks

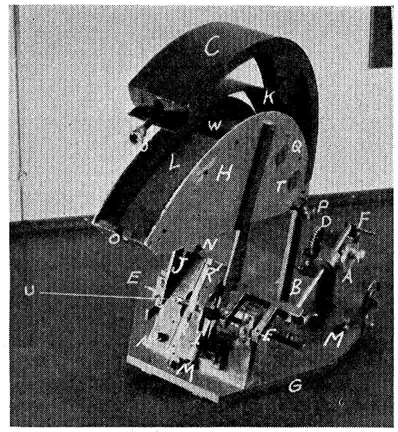

FIG. 1. The backward reflecting Bragg spectrograph shown with the quartz crystal U in its mounting J. R and R', brass plates forming the condenser through which the electric field is applied; N and N', terminals of the condenser; EE, shaft on which the crystal is rocked; I, protractor for setting the crystal; G, base of spectrograph; M, set screws for height adjustment. Arm B, follower F, cam A, and worm and pinion D, part of the arrangement for rocking the crystal during exposure. H, film holder; L, lead screen; W, window in screen; K, spring for holding film; G, light tight cover. P, screws for holding H in position; Q and T, threaded holes for other positions of H.

could possibly occur in this region. The motion of the crystal parallel to the 100 mm dimension in its mounting will have no effect on the width or intensity of the spectral line reflected from the face of the crystal. Thus, by comparing the widths of the lines reflected from the crystal when it is oscillating at its fundamental frequency and when it is oscillating at its second harmonic frequency, the true effect on the line produced by either the motion within the crystal of the grating space, or by the motion of the proposed Zwicky blocks may be determined accurately.

The crystal mounting J (Fig. 1) was accurately centered on the pointed ends of the rods E. The crystal was rocked through an angle of 5° during exposure by means of the shaft B, follower F, cam A, worm and pinion D, and a shaft (not shown in the figure) which connected the worm gear arrangement with the armature of a motor through a reduction gear. The film was placed in the holder H, over the lead screen L. The window W permitted the reflected radiation to strike the film at the proper position. The film was held fast by the spring K, and the cover C fitted over the holder, thus shielding

[9] Bradley and Jay, Proc. Roy. Soc. London **A45**, 507 (1933).

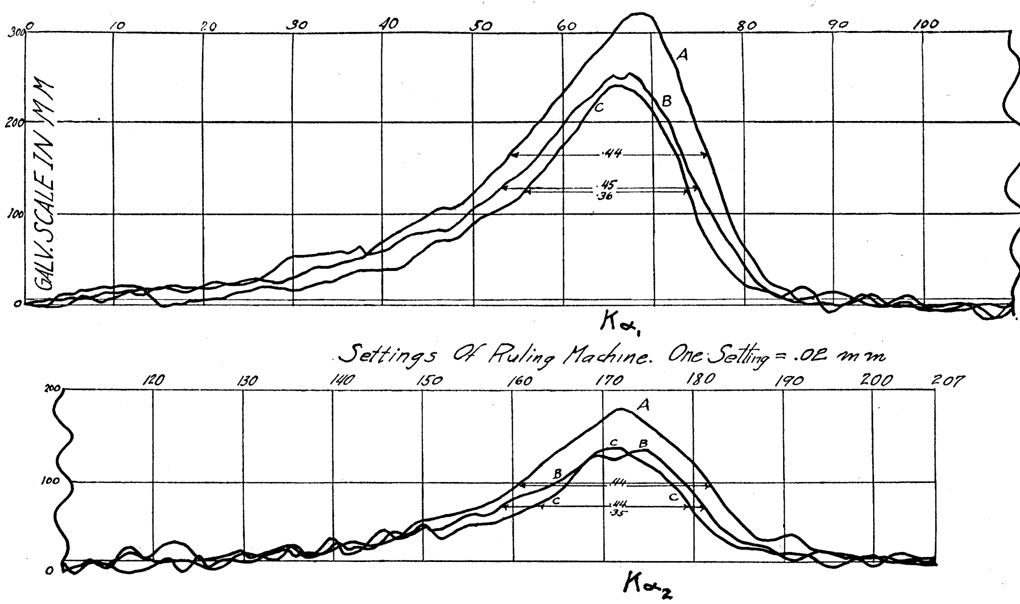

FIG. 2. Photograms of the three doublets obtained by the sixth order reflection from the (1 1 0) set of planes of the $K\alpha$ radiation of Mo. A, photogram of doublet obtained when crystal was oscillating at its fundamental frequency under the transverse effect; B, photogram of doublet obtained when the crystal was oscillating at its second harmonic frequency; C, photogram of doublet obtained when crystal was not oscillating.

the film from light. The holder was held in position by the screws P. The holes Q and T are for other positions of the holder. The spectrograph fits onto a track in a standard G.E. diffraction apparatus. The height of the spectrograph is adjusted by the screws M, shown on the base G of the instrument.

An exposure of sixteen hours was necessary to make each picture. The doublet obtained was highly resolved but quite weak. However, very satisfactory photograms were obtained by running the pictures through a fine structure photometer which was especially designed to reduce grain effects. This instrument has been described elsewhere.[10]

Experimental Results

Fig. 2 shows the photograms of the three doublets. Curve A is the photogram of the doublet reflected from the crystal when oscillating at its fundamental frequency, curve B is the photogram of the doublet reflected from the crystal when it was oscillating at its second harmonic frequency, and curve C is the photogram of the doublet reflected from the crystal when it was not oscillating. It is seen that the curve B is 0.09 mm wider than curve C, but of approximately the same amplitude. This result shows that the increase in width of the line is due solely to the jumping of the crystal in its mounting. Any difference in the amplitude of the curves B and C may be attributed to the variations in the operation of the x-ray tube. It is clearly shown that the width of curves A and B are approximately the same within the accuracy of the experiment. Hence, no variation of the grating constant takes place during oscillation; at least no variation which is greater than 1.45×10^{-5} per unit dimension. The fact that the amplitude of the lines shown in curve A are greater than the amplitude of the lines shown in curves B and C, shows that secondary extinction was reduced on the face of the crystal in the vicinity of the middle when the crystal was oscillating at its fundamental frequency, but was not reduced in this region when the crystal was oscillating at its second harmonic. This fact shows that strain existed through the middle of the crystal when it was vibrating at its fundamental frequency, but that no appreci-

[10] Sidon Harris, Rev. Sci. Inst. **4**, 598 (1933).

able strain existed in this section when the crystal was oscillating at its second harmonic. The crystal was resonating vigorously during both exposures; the total average change in length per unit length along the 100 mm direction was calculated from microscope readings and found to be 6×10^{-4}. If the strain along X through the middle of the crystal can be taken as equal to the strain along Y as the authors[11] have found to be the case in static conditions, an increase in line width of 0.42 mm would be expected from the line reflected from the crystal when oscillating at its fundamental frequency, if the spacing of the (1 1 0) set of planes changed in a one to one ratio with the outside dimension of the crystal in the X direction. As no increase, not even as small as 0.01 mm was detected, it is to be supposed that most of the strain inside the crystal along the X direction must surely occur in the π-planes of Zwicky, and not in the p-planes, whose spacing effects the position of the reflected line on the film. This evidence would be a strong support of Zwicky's theory if it were definitely known that the dynamic ratio of X strain to Y strain were unity. Experiments to determine this dynamic ratio are now in progress in this laboratory. However, until this ratio for dynamic conditions is determined, the result presented immediately above is added as merely an interesting possibility. As the strain through the middle of the crystal described in this experiment is homogeneous when the crystal is oscillating at its fundamental frequency, naturally the reduction of secondary extinction in the middle region is not as great as in the cases where inhomogeneous strain acts on the reflecting surfaces of the crystal. In order to observe this relatively small decrease in secondary extinction, the surface of the crystal irradiated by x-rays, was etched lightly with hydrofluoric acid in order to remove the imperfect surface layer produced by polishing.

X-ray photographs were also made by sending a beam of x-rays from a slit source through the middle of the crystal when it was oscillating at its fundamental frequency, and when it was not oscillating. The photograms of the transmission lines thus obtained are shown in Fig. 3. Curve A

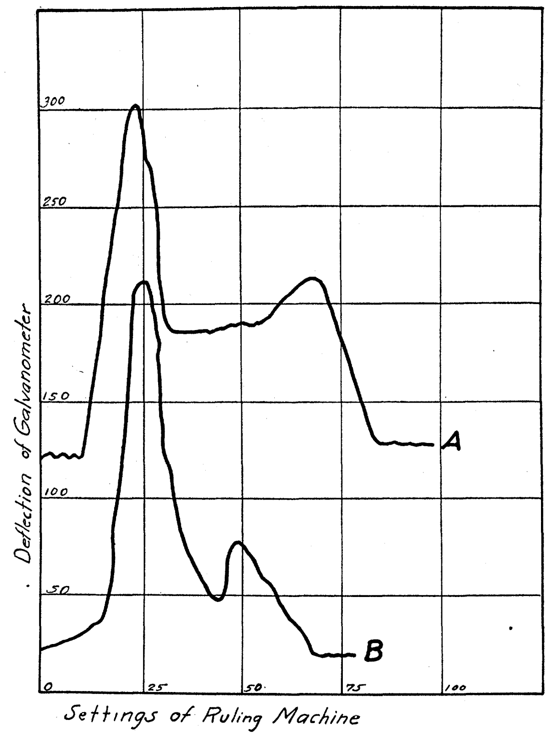

FIG. 3. Photograms of transmission lines obtained by passing a rectangular beam of x-rays through the middle of the crystal. A, photogram of line obtained when crystal was oscillating at its fundamental frequency; B, photogram of line obtained when crystal was not oscillating.

is the photogram of the line obtained from the crystal when it was oscillating at its fundamental frequency, and curve B is the photogram of the line obtained from the crystal when it was not oscillating. Curve A is wider than curve B because the lines were not reflected from the same set of planes. The crystal was in a horizontal position when the line corresponding to curve A was taken, and in a vertical position when the line corresponding to curve B was taken. The films were given the same exposure and development. As the source of x-rays was a slit instead of the usual pinhole used in making transmission pictures, only the lines of the principal spectrum were sharp. The sharpest line was chosen for the photogram in each case. The fact that one of the spots is wider than the other should have no effect on the comparison of the relative intensities of the components. It is seen that the component reflected from the polished face of the crystal

[11] Colby and Harris, J. Opt. Soc. Am. **24**, 217 (1934).

(the component of greatest intensity) was not increased by the oscillation of the crystal, and that the component reflected from the etched face of the crystal (the weak component) was increased by the oscillation of the crystal, while the middle of the spot which was reflected from the interior of the crystal was approximately doubled in intensity by the oscillation of the crystal. This result can be explained readily by the reduction of secondary extinction through the middle of the crystal. The polished surface already had secondary extinction reduced to a minimum, and therefore could be reduced no further by the oscillation; while on the surface of the etched face, secondary extinction existed to a higher degree and was appreciably reduced by the oscillation; and in the interior, secondary extinction existed to such a marked degree that the intensity of the reflected radiation was approximately doubled by the oscillation of the crystal. This result was predicted by the authors[4] in a recent publication in this journal.

In conclusion it may be said that the increased intensities of the components of the transmission lines, and the increased intensities of the reflected lines, together with the fact that the high resolving power spectrograph showed no increase in width of the reflection lines, show conclusively that any increased intensity in the radiation reflected from any portion of a quartz crystal oscillating piezoelectrically may be attributed almost entirely to a reduction of *secondary* extinction. This reduction of secondary extinction can be brought about only by the motion of some type of secondary structure block such as has been proposed by Zwicky as existing in crystals.

The authors wish to express their appreciation to Mr. L. H. Gruber, technician, for making the spectrograph used in this work.

Variation of the Dielectric Constant of Rochelle Salt Crystals with Frequency and Applied Field Strength

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(Received August 1, 1934)

The dielectric constant of Rochelle salt crystals perpendicular to the a axis at 0°C was measured over the frequency range of 30 to 30×10^6 cycles/sec. Following a fairly uniform decline from 62,000 at 30 cycles to 220 at 10^7 cycles, the dielectric constant drops abruptly to negative values (inductive reactance) at 14×10^7 cycles. This frequency is independent of the size and mounting of the crystal and thus appears to be critical for this crystalline material.

THE dielectric properties of Rochelle salt crystals are as unique as the magnetic properties of iron. In fact, the ferromagnetic analogy can be applied with considerable success, as has been pointed out by Valasek,[1] Müller[9] and others. Experimental measurements show a very high dielectric constant accompanied by hysteresis, and demonstrate the existence of two "Curie points," respectively, at −15°C and +23°C. Measurements of the dielectric constant of the substance are difficult because the results are very sensitive to the presence of even a slight layer of modified crystal or binding material between the crystal and the electrodes. The older measurements[1, 2, 3, 6] all give values which are too low because of this effect. In the more recent work of Sawyer and Tower,[5] Errera,[7] Kobeko and Kurtschatov,[8] and of Busch,[4] improved electrodes are described which give better results. The present paper describes the results obtained in extending the measurements

[1] Valasek (a) Phys. Rev. **19**, 478 (1922); (b) Phys. Rev. **23**, 114 (1924); (c) Phys. Rev. **15**, 537 (1920); (d) Science **65**, 235 (1927).
[2] Cady, Rep. Nat. Res. Council, May 1918.
[3] Anderson, Rep. Nat. Res. Council, April 1918.
[4] Busch, Helvetica Physica Acta **6**, 315 (1933).
[5] Sawyer and Tower, Phys. Rev. **21**, 269 (1930).
[6] Frayne, Phys. Rev. **21**, 348 (1923).
[7] Errera, Phys. Zeits. **32**, 368 (1931).
[8] Kobeko and Kurtschatov, Zeits. f. Physik **66**, 192 (1930).
[9] Müller, Phys. Rev. **44**, 854 (1933).

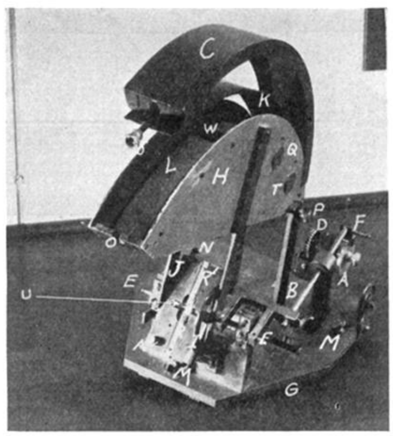

FIG. 1. The backward reflecting Bragg spectrograph shown with the quartz crystal U in its mounting J. R and R', brass plates forming the condenser through which the electric field is applied; N and N', terminals of the condenser; EE, shaft on which the crystal is rocked; I, protractor for setting the crystal; G, base of spectrograph; M, set screws for height adjustment. Arm B, follower F, cam A, and worm and pinion D, part of the arrangement for rocking the crystal during exposure. H, film holder; L, lead screen; W, window in screen; K, spring for holding film; G, light tight cover. P, screws for holding H in position; Q and T, threaded holes for other positions of H.