DECEMBER 15, 1932 PHYSICAL REVIEW VOLUME 42

Laue Patterns from Thick Crystals at Rest and Oscillating Piezoelectrically

By J. M. Cork
University of Michigan

(Received October 26, 1932)

Laue patterns have been observed for thick crystals of quartz and Rochelle salt oscillating piezoelectrically and for the same crystals as well as for calcite, rocksalt and other materials at rest. Patterns have been obtained when the crystalline faces were the natural cleavage surfaces and when they were polished and when etched by suitable solvents. Regular Bragg reflections have been observed and compared for a quartz crystal with polished face and for the same crystal with the face etched with hydrofluoric acid. In the Laue patterns, the individual spots show a multiple structure in every case. The inner spots are double in nature. For the crystal oscillating piezoelectrically or at rest with polished faces the doubling of each spot is radially symmetrical. For the crystal with etched faces and at rest the inner component of the double spot is the stronger particularly for those spots near the center. The Bragg reflection from a quartz crystal with etched face shows the absence of any layer with reduced extinction at the surface. Further elaboration of the Laue diffraction theory seems necessary to account for the multiple structure of the spots formed by an ideally perfect crystal.

THE reflection of x-rays from quartz crystals oscillating piezoelectrically and at rest have been observed by means of the Bragg method and the Laue method in many investigations.[1,2,3,4] In the transmission method the piezoelectric oscillation leads to an intensity of the reflected x-ray beams much greater than that found for the crystals at rest. Moreover Barrett and Howe have observed that each Laue spot for the oscillating crystal appears to have a fine structure. To observe this fine structure the present investigation employing a more finely collimated beam of x-rays and crystals of greater thickness was undertaken.

Apparatus and Results

Two Laue cameras were constructed. In one the distance from the crystal to the photographic plate was 20 cm while in the other the corresponding distance was only 7 cm. It was thus possible to observe the complete pattern or only the central spots in greater detail. In both cases the x-ray beam was collimated by a fine slit system formed by two pinholes each 0.05 cm in diameter and spaced 12 cm apart. The first pinhole was 30 cms from the focal spot of the x-ray tube. A standard 200 kv Coolidge tube with a target of tungsten was employed. It was operated continuously at only 110 kv with a current of 2 milliamperes.

---

[1] Y. Sakisaka, Jap. Jour. Phys. **4,** 171 (1927).

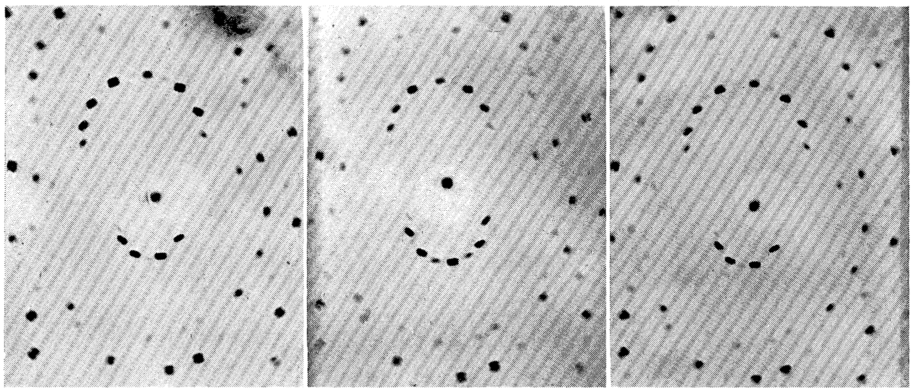
[2] G. W. Fox and P. H. Carr, Phys. Rev. **37,** 1622 (1931).

[3] G. W. Fox and J. M. Cork, Phys. Rev. **38,** 1420 (1931).

[4] C. S. Barrett and C. E. Howe, Phys. Rev. **38,** 2290 (1931); **39,** 889 (1932).

Exposures were first taken with crystals of quartz ranging in thickness from 4 mm to 7 mm. In the beginning considerable difficulty was experienced in maintaining these thick crystals in the vibrating state continuously. A vacuum tube oscillator with two tubes having capacitative feed-back was arranged which accomplished this result satisfactorily.

Typical patterns obtained with the longer camera are shown in Fig. 1, A, B and C. For the quartz crystal with polished faces the spots were symmetrically double as were also the spots for the oscillating crystal regardless of the nature of the faces. When the crystal with polished faces was etched by immersion in hydrofluoric acid so that the thickness was reduced by not over 0.005 mm a pattern as shown in C, Fig. 1 was obtained. It is apparent that although the spots are now double the component parts are not of equal intensity. The inner part (i.e., that portion usually associated with reflection from the side of the crystal nearest the photographic plate) is much blacker than the outer component. This dissymmetry is not so apparent in the outer spots of the pattern as shown in the photograms obtained with the shorter camera.



A B C

Fig. 1. Laue patterns obtained with quartz. A, etched faces oscillating 12 hr. exposure; B, polished faces non-oscillating 20 hr. exposure; C, etched faces non-oscillating 20 hr. exposure.

To observe the effect of this etching upon the surface of the crystal the following test was carried out. The quartz crystal was mounted in a Siegbahn vacuum spectrograph and employed to measure the wave-length of the $K\alpha_1$ line of molybdenum ($\lambda = 5.41\text{A}$). The crystal was adjusted so that the desired radiation fell on the photographic plate in one position and then both crystal and plate were rotated through predetermined angles so that the same line was reflected to the photographic plate on the opposite side of the direct beam. One should obtain therefore upon the plate the segments of two inverted circles overlapping exactly at the center if the angle of displacement were chosen correctly. With the use of the quartz crystal with polished faces these lines are very diffuse so that the two segments are indistinguish-

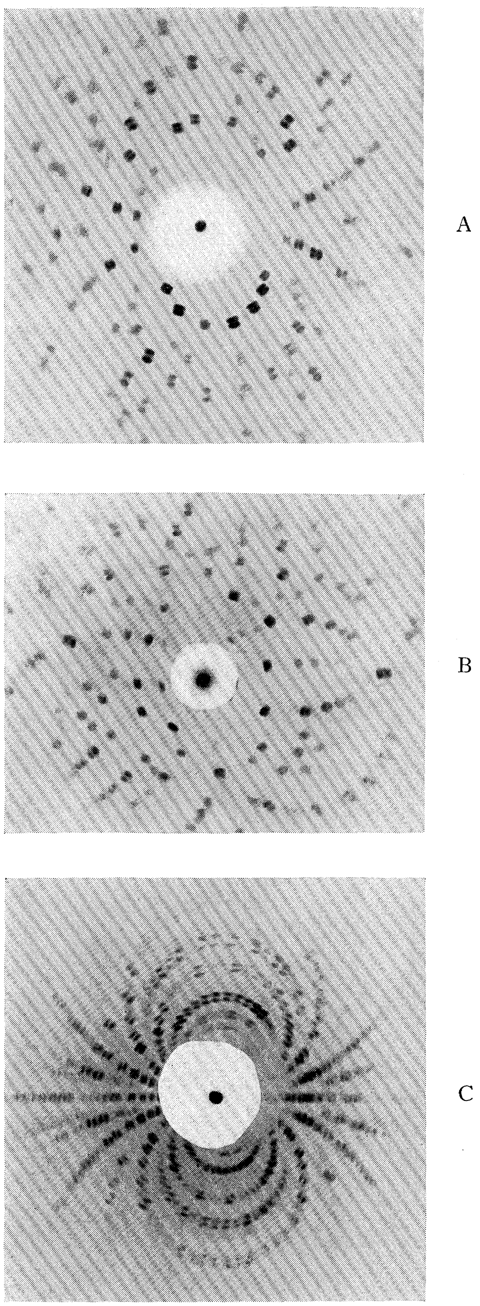
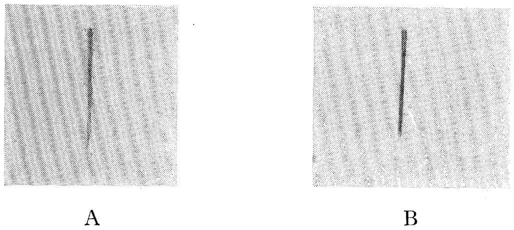


Fig. 3. Laue patterns from thick crystals at rest, A, quartz, B, calcite, C, Rochelle salt.

able as shown in Fig. 2A. Upon lightly etching the face of the quartz crystal with hydrofluoric acid these two lines become very sharp and are clearly distinguishable in Fig. 2B. For radiation of this wave-length the penetration into the crystal is exceedingly small and the sharpness of the photographic lines might safely be interpreted as indicating the almost complete absence of any disarranged particles on the surface.

It would appear therefore that the doubling of the spots was not due to a lack of extinction at the surfaces as has been generally assumed. To investigate this phenomenon further, Laue patterns were obtained with freshly cleaved slabs of several other materials. For some exposures the faces were etched by appropriate solvents while for others the specimen was left with its natural cleavage faces. Only in the case of a very bad specimen of rocksalt was a pattern obtained in which the Laue spots were not clearly multiple in structure. The spots toward the center of the pattern were double while those farthest removed from the central position were often triple in nature. Typical photograms of this sort are shown in Fig. 3 A, B and C which are for quartz, calcite and Rochelle salt, respectively.



A B

Fig. 2. Double exposures of molybdenum $L\alpha_1$ line, A, polished face, B, etched face.

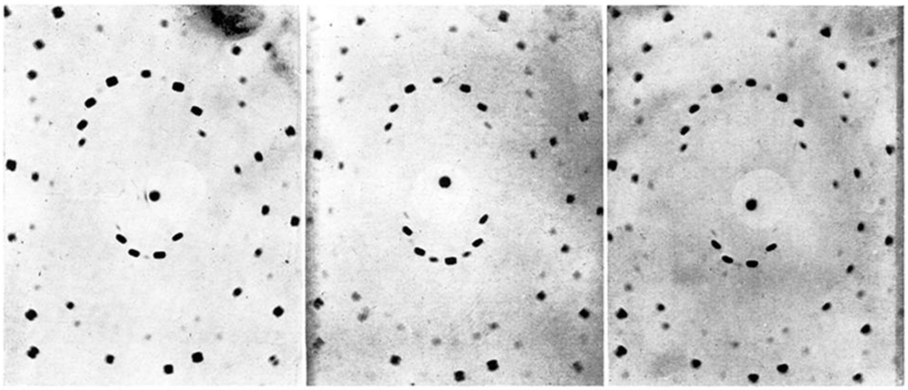
This doubling or tripling of the Laue spots was undoubtedly first observed by M. de Broglie[5] when he passed a beam of x-rays through thin slips of crystal (rocksalt and fluorite) at an angle of incidence of 80° or a grazing angle of 10°. In this way he had effectively a thick crystal. The result was explained by assuming that the crystal traversed by the x-ray beam was composed of as many granules separated by accidental parallel cleavage planes as there were components in the spots. This would of course not account for the regularity and radial symmetry always observed in the reflected beams.

Freshly cleaved surfaces of calcite have been found[6] to give by Bragg surface reflection, spectral lines of width approximating that to be expected for an ideally perfect crystal. From photograms such as Fig. 2B it may be concluded that at least certain specimens of quartz are also very regular. Yet these same crystals give Laue patterns in which the spots have a multiple structure. It would appear that further elaboration of the usual Laue theory introducing interference between non-parallel radiation and perhaps with reference to a secondary structure as proposed by Zwicky,[7] is necessary.

[5] M. de Broglie, Comptes Rendus **156,** 1153, also 1461 (1913).

[6] B. Davis and W. Stempel, Phys. Rev. **17,** 608 (1921).

[7] F. Zwicky, Phys. Rev. **41,** 400 (1932).



A B C

Fig. 1. Laue patterns obtained with quartz. A, etched faces oscillating 12 hr. exposure; B, polished faces non-oscillating 20 hr. exposure; C, etched faces non-oscillating 20 hr. exposure.

A B

Fig. 2. Double exposures of molybdenum $L\alpha_1$ line, A, polished face, B, etched face.

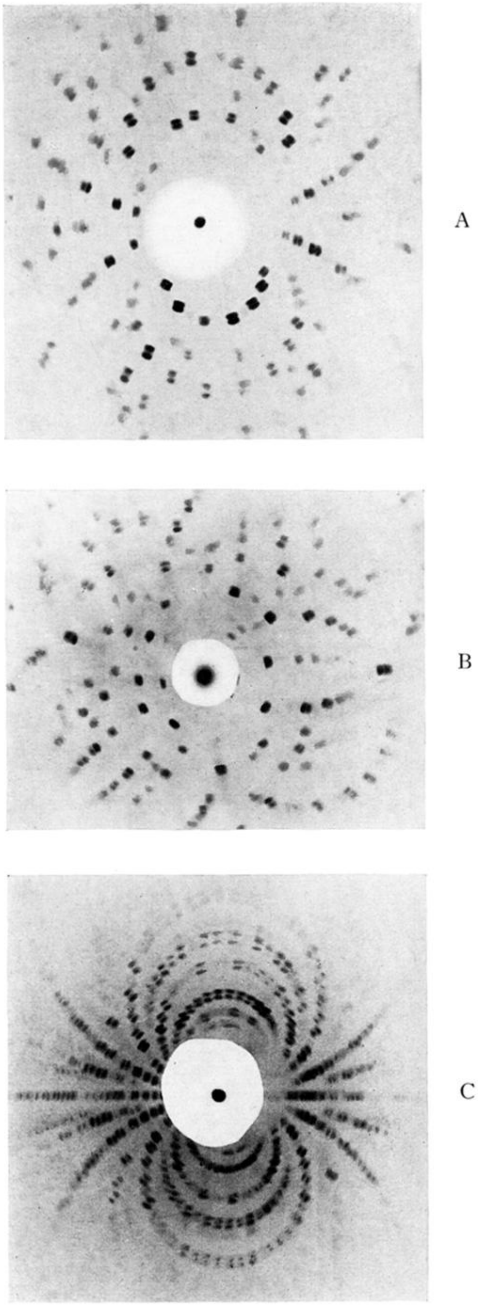


Fig. 3. Laue patterns from thick crystals at rest, A, quartz, B, calcite, C, Rochelle salt.