

(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 authors⁴ 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.

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

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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 Müller⁹ 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 $+23^\circ\text{C}$. 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 Kurtchatov,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 Kurtchatov, Zeits. f. Physik **66**, 192 (1930).

⁹ Müller, Phys. Rev. **44**, 854 (1933).

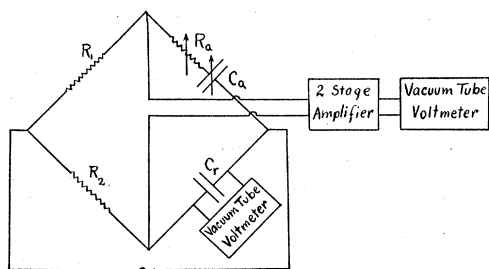


FIG. 1. Capacitance bridge.

with the newer type of electrodes to very high frequencies. The crystal was cut so that its faces were at right angles to the a axis, since it is only in this direction that the unusual dielectric properties exist.

METHOD OF MEASUREMENT AND APPARATUS

Two types of measuring circuits were used, a capacitance bridge for the lower frequencies (30–75,000 cycles) and a resonance circuit for the higher frequencies (75,000–30,000,000 cycles).

In the capacitance bridge, Fig. 1, R_1 and R_2 are equal resistances, C_a is a standard variable condenser (50–1,000,000 μmf), R_a is a variable non-inductive resistance (0–100,000 ohms) and C_r a condenser whose dielectric is a plate of the crystal under observation.

It has been shown by Kobeko and Kurtschakov⁸ that the potential gradient within a Rochelle salt crystal when used as the dielectric of a condenser is approximately uniform. The crystal condenser and its leakage, therefore, may be represented by a pure capacitance C in parallel with a non-inductive resistance R . Since R_1 at balance equals R_2 , the impedance Z_p of the crystal, that is, of C and R in parallel, must equal the impedance Z_s of R_a and C_a in series. The impedance of R and C in parallel is

$$Z_p = (R/j\omega C)/(R+1/j\omega C).$$

The impedance of R_a and C_a in series is

$$Z_s = R_a + 1/j\omega C_a.$$

Since at balance Z_p equals Z_s ,

$$R_a + 1/j\omega C_a = (R/j\omega C)/(R+1/j\omega C).$$

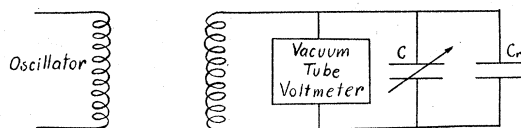


FIG. 2. Resonance circuit.

Equating the real and the imaginary parts of the above, and solving for C and R , it is found that:

$$C = C_a / (1 + \omega^2 R_a^2 C_a^2)$$

and

$$R = R_a + 1/\omega^2 C_a^2 R_a^2.$$

The open-circuit voltage across the crystal, measured by a vacuum-tube voltmeter, gives the electrostatic field strength to which the crystal is subjected during the measurements.

The resonance circuit method is a substitution method of the type used by Frayne.⁶ The difference at resonance, Fig. 2, in the two settings of the standard air condenser, C , when C_r is disconnected and connected, is the capacitance C_r . In this circuit the vacuum-tube voltmeter indicates the voltage across the crystal as well as resonance.

The types of electrodes used were (1) the mercury-Rochelle salt solution type, Fig. 3, similar to those used by Kobeko and Kurtschakov⁸ and (2) the silver-plated type, Fig. 4, in which the silver was evaporated and allowed to condense on the surface of the crystal in a vacuum. The crystal plates varied in thickness from 2 to 8 mm and the surfaces were usually 3 cm square. The electrodes were several millimeters smaller. After preparing the condenser, the crystal was given an impervious coating of celluloid dissolved in amyl acetate.

The temperature of the crystal was maintained at 0°C because at this temperature the dielectric constant is high and has a low temperature coefficient.^{1, 6}

RESULTS

Fig. 5 shows typical curves for the dependence of the dielectric constant on frequency, for each of the two types of electrodes used, at 0°C and a potential gradient of 8.75 volts/cm.

The values of the dielectric constant changed from about 62,000 at 30 cycles to approximately

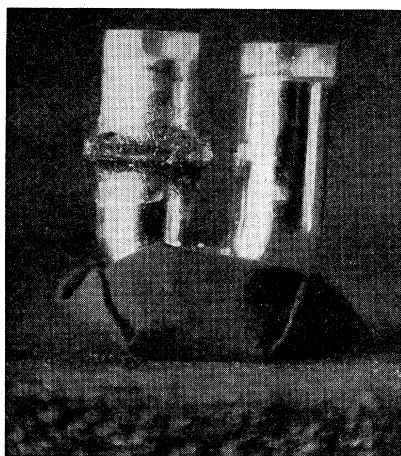


FIG. 3. Rochelle salt crystal condenser with mercury-salt solution electrodes.

220 at 10^7 cycles when the mercury-salt solution electrodes were used and from about 3000 at 30 cycles to 100 at about 10^7 cycles with the silver-plated electrodes.

At a frequency of about 14×10^6 cycles a critical point was reached, after which the reactance of the crystal became inductive and remained that to the experimental limit of 30×10^7 cycles. In some cases it rose and fell again very steeply as shown in Fig. 5 before remaining negative. This critical point was independent of the physical dimensions of the crystal and of the type of electrodes used. It is accordingly of a different nature from those observed by Frayne and Errera at about 60,000 cycles.

These latter variations in the dielectric constant are due to the well-known fact that in a piezoelectric crystal the polarization produced by an applied alternating field is very appreciably altered by the accompanying piezoelectric polarization due to the deformation of the crystal. Especially large variations which resemble optical anomalous dispersion curves occur at the resonance frequencies. These frequencies depend on the dimensions of the crystal and on the velocity of sound in the crystalline medium. In our experiment the large damping and low field strength apparently prevented appreciable resonance effects of this type. According to the dimensions of our crystals these should have

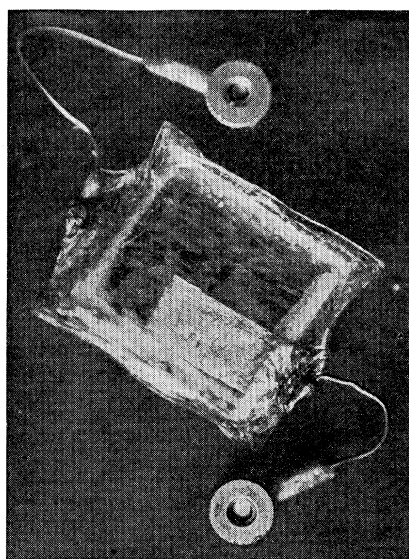


FIG. 4. Rochelle salt crystal condenser with silver plated electrodes.

been located at about 50,000 cycles. The critical frequency we observed at 14×10^6 cycles was independent of the dimensions of the crystal. If it is due to any resonance effect, it must be ascribed to an internal structure of about 0.17 mm in width.

The rather large decline of the dielectric constant in the frequency range 30 to 100,000 cycles is no doubt related to the large residual-charge effect discussed by Valasek.^{1b}

The large decrease in the values obtained for the dielectric constant when using the silver-plated electrodes and other electrodes with the probable exception of mercury, may be explained by assuming that during the process of silver-plating the surface of the crystal may become slightly dehydrated and that this thin dehydrated film has a low dielectric constant. From a well-known formula in electrostatics, the dielectric constant of two dielectrics in series is

$$K = K_1 K_2 (a_1 + a_2) / (a_1 K_2 + a_2 K_1),$$

where K_1 and a_1 are the dielectric constant and thickness, respectively, of one of the dielectrics, the crystal in this case, and K_2 and a_2 are the dielectric constant and thickness, respectively, of the other dielectric, the film. If the ratio of

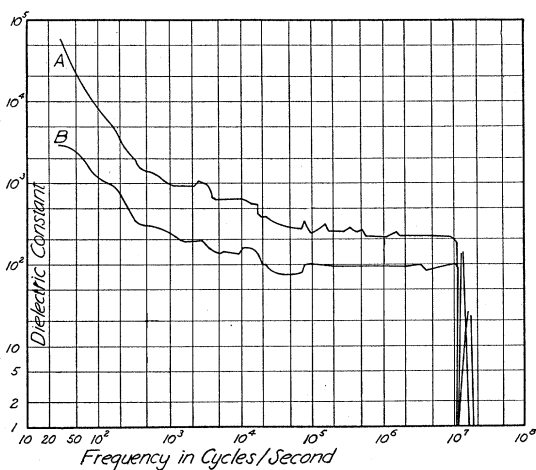


FIG. 5. Variation of dielectric constant with frequency of Rochelle salt crystal cut perpendicular to its a axis. Curve A is for mercury-salt solution electrodes and curve B for silver-plated electrodes; $t=0^{\circ}\text{C}$, potential gradient across crystal = 8.75 volts/cm.

K_1 to K_2 is very large, as is the case here, even with thin films the effective dielectric constant, K , will be considerably reduced from the large dielectric constant K_1 .

With low frequencies the mercury-salt solution electrodes give higher values for the dielectric constant than other electrodes that have been tested. When these contacts are new and properly made the film effect may be assumed to be nearly eliminated. The silver-film electrodes are of value in that they form a crystal condenser whose capacitance apparently remains constant if the condenser is imbedded in some moisture proof insulating material.

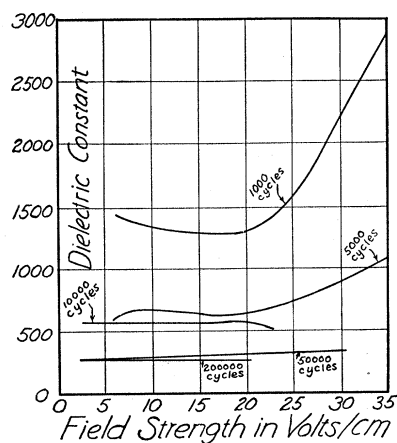


FIG. 6. Variation of dielectric constant with applied electrostatic field at various frequencies.

The dielectric constant is also a function of the field strength. For low field strengths and at low frequencies the observed variation is shown in Fig. 6. At higher frequencies and at low field strengths, the dielectric constant is very nearly independent of the field strength. At high field strengths such as 300 volts/cm or greater, and at low frequencies, Kobeko and Kurtschatov⁸ found that the dielectric constant decreased with an increase in field strength.

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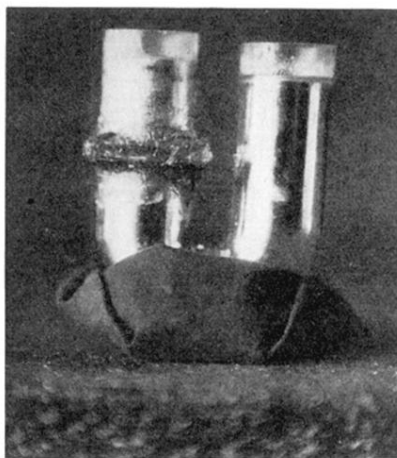


FIG. 3. Rochelle salt crystal condenser with mercury-salt solution electrodes.

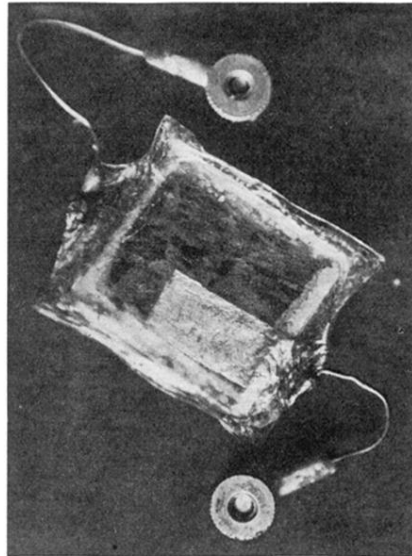


FIG. 4. Rochelle salt crystal condenser with silver plated electrodes.