THE REVERSIBLE INDUCTIVITY OF ROCHELLE SALT CRYSTALS AND ITS RELATION TO FREQUENCY AND TEMPERATURE.

By John G. Frayne.

Abstract.

Reversible inductivity of Rochelle salt crystals is defined as $k_r = -dD/dE$ or the limit of the ratio of the change of induction to the change of electric field in the dielectric as these approach zero. *The method of measurement* used involved determining the capacity of a condenser with a plate of Rochelle salt crystal, cut normal to the ă-axis, as dielectric, by adjusting the oscillating circuit containing it to resonance with a loosely coupled vacuum tube circuit. *Variation with frequency of alternating field* was studied up to 4×10^7 cycles, for the crystal at 0° C (Fig. 3). Above the natural periods of vibration of the plate, 60,000 and 156,000 cycles, k_r was constant at 70, while below them it was constant at about 112 to 8,000 cycles and then began increasing. The lower frequency values are affected by piezo-electric effects due to mechanical vibration in certain ranges and by polarization effects which increase as the frequency decreases; hence the high frequency values have a simpler physical significance. *Variation with temperature, − 80° to 50° C:* Curves obtained for frequencies of 10^6, 50,000 (near resonance), and 8,000 cycles (Fig. 4), all show maxima for about − 16° and 24° C. The minimum at about 6° may be associated with the maximum piezo-effect which occurs in this range. *Hysteresis loop:* The previous results are for an applied electric field of zero. As the field is increased, k_r reaches a maximum, then a minimum, then increases to infinity as conduction begins for 2,700 or 3,000 volts/cm, depending on the direction. *Natural polarization* may be estimated from the asymmetry of the hysteresis loop. Two samples gave 125 and 175 volts/cm. A comparison of the temperature curves for three samples cut from the same crystal shows marked differences which indicate that the natural polarization is far from uniform even in the same crystal. *Relation of k_r to high frequency resistance* is probably close. The two properties vary in general in much the same way. This seems to indicate that the energy absorbed in the dielectric is very simply related to the process of doublet formation.

Mechanical vibrations of Rochelle salt condenser in oscillating circuit.—At the natural frequencies of the crystal plate, abnormal values of k_r and of the resistance were found. In the case of the transverse resonance, the computed wave-length was four times the width of the plate.

In a paper on the theory of ferromagnetism in 1908 R. Gans[1] introduced a quantity which he called the reversible permeability of iron. The name was suggested to him by the fact that iron can be taken through a reversible hysteresis cycle, provided the field intensity is varied at the start in a direction *opposite* to the existing field in the

---

[1] R. Gans, Annalen der Physik, 27, pp. 1–6, 1908.

iron. The resulting loop may be made as small as desired, and Gans defines the limiting value of the ratio of the change in the induction to the change in the field strength as the *reversible permeability* of iron.

When an alternating field is applied to iron the induction undergoes a hysteretic cycle for each current cycle, and if the alternating field be made very small, the ratio of the maximum amplitude of the induction per cycle to the maximum amplitude of the field strength per cycle will become the reversible permeability of Gans. By measuring the self-induction of an iron core solenoid in a resonating circuit Fritz Erhardt[1] has measured the reversible permeability under various conditions.

In a recent paper Valasek[2] has shown that the behavior of Rochelle salt crystals in an electric field is analogous to that of iron in a magnetic field. He found that a marked dielectric hysteresis loop existed when a Rochelle salt crystal was carried through a cycle of electric field intensities. The dielectric constant of this substance, consequently, depends on the field strength when measured by direct or alternating current methods.

Carrying the analogy further I have denoted by the term "*reversible inductivity*" the quantity which corresponds to reversible permeability in ferromagnetic substances. If we take a point on the normal induction curve (Fig. 1) of a sample plate of Rochelle salt and gradually decrease the field to B, then reverse the direction of the field, the point A will again be reached. If the field had only been reduced to B' and increased again to A, the area of the loop would have been reduced, and in the limiting case AB will become a straight line. The value of

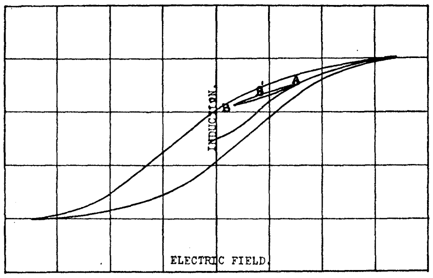

Fig. 1.

$$\text{limit } (\Delta E \doteq 0) - \frac{\Delta D}{\Delta E} = - \frac{dD}{dE} = k_r,$$

which we shall call the reversible inductivity.

---

[1] Fritz Erhardt, Ann. der Phys., 54, p. 41, 1917.

[2] J. Valasek, Phys. Rev., 4, p. 475, 1921.

The piezo-electric activity of Rochelle salt crystals tends further to complicate the dielectric properties of the substance when studied from the resonance standpoint. Cady[1] has shown that the dielectric constant of quartz, another piezo-electrically active substance, changes very abruptly when the period of the exciting field approaches the natural period of vibration of the crystal plate. The counter piezo-electromotive force produces a displacement in the dielectric which becomes very large at resonance, and being practically 180° out of phase with the applied e.m.f. makes the actual displacement negative over a narrow band of frequencies at the resonance point.

Valasek[2] has shown that the piezo-electric properties and dielectric constant of Rochelle salt are markedly dependent on temperature, the piezo response rising to a maximum between $-20°$ C and $+20°$ C, but dropping sharply to zero below and above these temperatures. He also found that the dielectric constant when measured by direct current methods increased rapidly between the same temperatures.

In this paper I have studied the effect on the dielectric constant of changing the frequency of the exciting field from 1,000 cycles to 4×10^7 cycles per second. At the same time the temperature-inductivity graphs have been plotted for different frequencies. Curves have also been obtained showing how the inductivity depends on the state of polarization of the crystal plate, and finally the properties of different plates cut from the same crystal have been examined.

EXPERIMENTAL PROCEDURE.

The crystal plate which formed the dielectric of the condenser used in this research was cut so that its face was perpendicular to the ă-axis, so as to obtain the greatest piezo-electric effect. Tin foil was fastened with shellac to either side, and leads were brought out from the foil through the stopper of the glass tube in which the condenser was contained. Phosphorous pentoxide was always inserted in the bottom of the tube while the condenser was in use, in order to keep the sides of the plate free from moisture. The glass tube itself was placed in a Dewar flask, and the temperature of the plate was usually kept at 0° C, except when temperature variation was desired. Arrangements were also made, see Fig. 2, for applying a steady electric field to the crystal, the size and direction of the latter being capable of adjustment.

The capacity of the condenser gives immediately the value of the dielectric constant, and when a weak alternating field is used the value

---

[1] Cady, P. I. R. E., 10, p. 83, 1922.

[2] Valasek, PHYS. REV., 19, p. 478, 1922.

obtained for this constant coincides with the reversible inductivity of the material under the conditions in question.

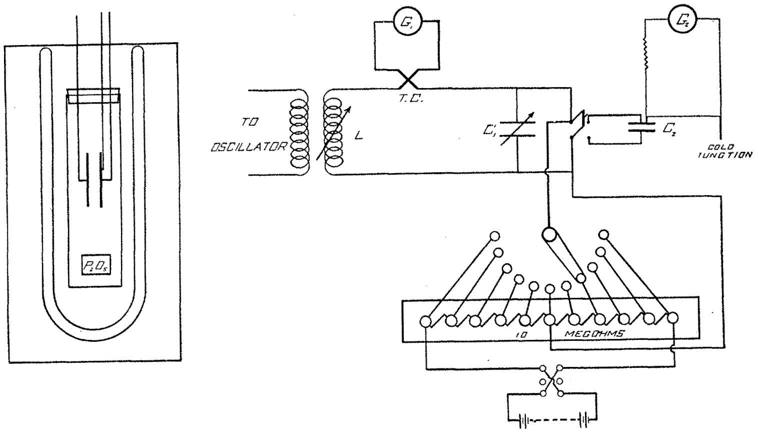

Fig. 2.

A vacuum tube was used as a source of undamped oscillations, and frequencies from a few thousand to 40,000,000 cycles a second were obtained. The oscillator was very loosely coupled to the receiving circuit, the coils being placed about a meter apart. With the plate condenser out of the circuit the variable capacity C was adjusted for resonance, the latter being indicated by the maximum deflection of the galvanometer G connected to the vacuum thermo-couple which was inserted in the resonating circuit. The crystal plate condenser was then inserted in parallel with C and the latter adjusted again for resonance. The capacity and effective resistance of the crystal plate condenser were then calculated as follows:

Let C_x = capacity of plate condenser.
R_x = H.F. resistance of plate condenser.
C_1 = capacity of C at first resonance.
C_2 = capacity of C at second resonance.
L_1 = inductance of coil L.
I_1 = current through C.
I_2 = current through C_x.

From first resonance condition, if the mutual inductance between oscillator and receiving circuit be neglected, $LC_1 = \omega^2$, ω being $2\pi \times$ the frequency. In order to write an analytical statement of the second condition for resonance, it is necessary to find an expression for the vector impedance of two condensers in parallel, one of which has an equivalent series resistance. If we denote by E the instantaneous e.m.f. across the condensers, then

$$I_1 = E \Big/ \frac{j}{C_2\omega},$$

j being $\sqrt{-1}$,

$$I_2 = E \Big/ \left(R_x + \frac{j}{C_x\omega}\right) = \frac{E(R_x - j/C_x\omega)}{R_x^2 + 1/C_x^2\omega^2}.$$

The total current is therefore

$$\begin{aligned} I = I_1 + I_2 &= E\left\{\frac{R_x}{R_x^2 + 1/C_x^2\omega^2} - j\left(C_2 + \frac{C_x}{R_x^2C_x^2\omega^2 + 1}\right)\right\} \qquad (1) \\ &= E(A - jB) \end{aligned}$$

or

$$I = E \Big/ \left\{\frac{A + jB}{A^2 + B^2}\right\}.$$

Hence the impedance of the divided circuit is

$$\frac{A}{A^2 + B^2} + j\frac{B}{A^2 + B^2}.$$

The impedance of the entire resonant circuit is, therefore,

$$-jL\omega + A/(A^2 + B^2) + jB/(A^2 + B^2).$$

The condition for resonance is that the coefficient of j becomes zero. Hence

$$L\omega = B/(A^2 + B^2).$$

As a first assumption, which is supported by experiment, we shall neglect A^2 in comparison with B^2. Then $L\omega = 1/B$ or

$$\omega^2 = \frac{1}{L\{C_2 + C_x/(R_x^2C_x^2\omega^2 + 1)\}}. \qquad (2)$$

As a second assumption, if we consider $R_x^2C_x^2\omega^2$ negligible in comparison to unity, then

$$\omega^2 = \frac{1}{L(C_x + C)},$$

which is the ordinary condition for resonance. From (2) we obtain

$$C_x = (C_1 - C_2)(R_x^2C_x^2\omega^2 + 1).$$

From this relation C_x can be determined, provided R_x is known. The latter can best be found by inserting the plate condenser in series with the air condenser and noting the resonance deflections before and after insertion. The value of R_x can very easily be calculated from these observations. Except at very high freuqencies, or near the natural period of the crystal where the absorption of energy becomes very large, $R_x^2C_x^2\omega^2$ is very small compared to unity, and we may, with a fair degree of accuracy, write $C_x = C_1 - C_2$ which is the ordinary law for adding capacities in parallel.

The coil L was gradually decreased by taking off turns of wire as the frequency of the field was increased. When the frequency reached 4×10^7 cycles per second, the coil consisted of only one turn. In order to get accurate results at this frequency, all the connecting wires were made as short as possible, thereby reducing their inductance to a negligible quantity.

In order to measure k_r it was necessary to use very weak alternating fields, and in no instance did the alternating field in the dielectric exceed 3 volts per cm. It was found by trial that a variation from .5 to 5.0 volts per cm produced no measurable change in k_r. Above 5 volts k_r increased with increasing field strength up to about 100 volts per cm. Of course, the quantity as measured at these high field strengths is no longer the reversible inductivity of the substance.

Frequency Variation.

The first thing of interest in studying reversible inductivity is the effect of changing the frequency of the exciting field. In the method used by Valasek for determining the statical dielectric constant, he measured the charge on the condenser by the throw of a ballistic galvanometer, but it is doubtful if the value he obtained in that manner really represents the true dielectric constant. The hysteresis property of the substance would indicate that a considerable time would elapse before the condenser was really discharged, it being well known that Rochelle salt crystals retain their polarization for some time after the field has been removed. On the other hand when an alternating field is used, provided the alternations are sufficiently rapid, it is conceivable that the material would scarcely have sufficient time to become polarized in any one direction. The curve in Fig. 3 shows that as the frequency

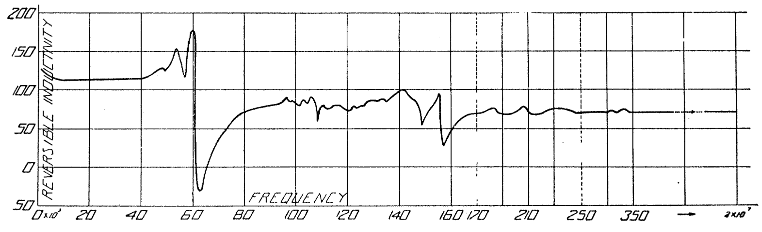

Fig. 3.

is increased above zero k_r drops until a frequency of about 8,000 cycles per sec. is reached. From this frequency up to about 50,000 cycles per sec. very little change is apparent. However, at 61,000 cycles per sec. the natural longitudinal period of vibration is reached, and the dielectric constant becomes negative over a narrow range of frequencies. Just below and above this frequency the absorption of energy by the plate is very pronounced, the effective resistance becoming about several thousand ohms. Between 80,000 and 140,000 cycles per sec. the curve shows irregularities which are probably due to harmonics of the natural vibration or interference between the latter and sub-harmonics of the higher resonance vibration. At 156,000 cycles per sec. another resonance point is reached, but the changes in the inductivity here are small compared with the previous values. Between 170,000 and 350,000 cycles per sec. a few small variations are found which are probably due to excitation of the crystal by frequencies bearing a relation to the natural period of vibration.

If we take Young's Modulus for the substance as 4×10^{11} dynes per cm^2 and the specific gravity as 1.76 grams per c.c., the velocity of sound in the crystal is given by $v = \sqrt{M/\rho} = 4.77 \times 10^5$ cms per sec. The wave-lengths corresponding to the resonance frequencies above are respectively 7.70 and 3.30 cms. From the manner in which the plate is mounted the transverse vibration should be the greatest, and the greatest changes in the inductivity would be expected there. If the plate were vibrating freely the wave-length should be twice the breadth of the plate, but it is actually four times the breadth. The wave-length of the longitudinal vibration is no multiple of the length 2.34 cm, and this is probably caused by the mounting which tends to damp the longitudinal vibrations.

Above 350,000 cycles per sec. the value of k_r remained constant up to 4×10^7 cycles per sec. which was the maximum frequency used. The constancy of the inductivity over such a large range of frequency would seem to indicate that this value of 70 is correct for this specimen of the salt. Below the resonance point the inductivity may be influenced by polarization effects or the proximity of the resonance vibrations. At frequencies of ten million cycles or more, however, it is difficult to conceive of permanent polarization effects. As it was experimentally difficult to use a resonance method at frequencies above 4×10^7, further investigation above this frequency has been left over until a better method has been found for this purpose.

The high frequency resistance remained constant with k_r. At the resonance points, as stated above, the absorption increased before and after the natural periods of vibration of the plate. At frequencies approaching zero the resistance increased in about the same ratio as the inductivity.

Temperature Variation.

The curves in Fig. 4 show that it would be impossible to work with Rochelle salt at room temperatures and obtain any consistent results. Hence the measurements carried out in the last section were all carried out at 0° C, the tube containing the Rochelle salt being entirely surrounded by melting ice. It will be noticed that all the temperature inductivity curves show the same characteristics. The value of k_r rises from about 12 at − 80° C to 100 or more, depending on the frequency, in the neighborhood of − 16° C. Then it takes a sharp dip dropping by about 50 per cent at + 6° C. Here it rises to a sharp maximum at about 24° C, where it starts to drop again. The temperature was not carried above 50° C, as the substance melts at about 51° C. The curve A represents conditions from the highest frequency used down to the resonance region. The curve B represents conditions at a frequency of 50,000 cycles per sec., where Fig. 3 indicates a large value of k_r at zero Centigrade. It is peculiar that throughout the entire range of temperature, the values of k_{r_1} in B are greater than at the corresponding temperatures in A. The same condition holds good in C which represents conditions below resonance and above 8,000 cycles per sec.

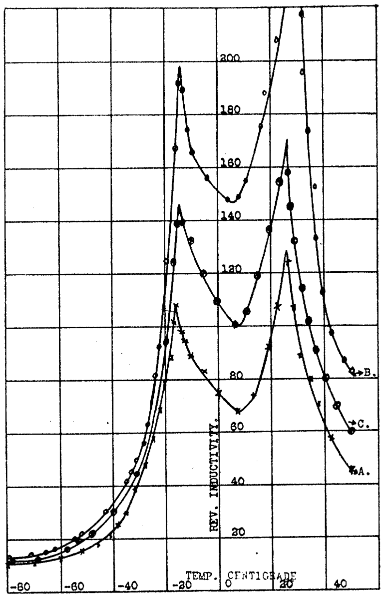

Fig. 4.

The temperature inductivity curves, by dropping to a minimum at + 6° C, vary in exactly the opposite manner to the values of the dielectric constant as determined by Valasek[1] using the ballistic galvanometer method. He did, however, find that the piezo-electric response reached a maximum at about 0° C, rising from practically nothing at − 20° C and descending sharply again at + 23° C. The stresses that are set up in the crystal due to the alternating field produce a counter piezo-electromotive force, which in turn produces a displacement which may be almost

---

[1] Loc. cit.

180° out of phase with the displacement caused by the exciting field. Hence in a region where the piezo-response is a maximum, we might expect a maximum counter displacement in the dielectric and this will in turn cause a diminution in k_r at that place.

Below − 16° C and above + 24° C the curves are of interest in that we have a sharp variation of k_r with temperature. These regions lie outside the piezo-electric range of activity, and the effect of temperature seems to increase or decrease the polarization depending on the particular part of the temperature range in question. An increase in the polarization is connected very closely with an increase in the high frequency resistance. This seems to indicate that the energy absorbed in the dielectric is very simply related to the process of doublet formation. High frequency resistance, as a rule, has no counterpart in direct current measurements, owing its existence to the dissipation of electrical energy in the dielectric, as the latter charges and discharges with the alteration of the field. It is quite natural, then, to expect a large high frequency resistance to accompany a large polarization on account of the energy required to separate the electric doublets. I have avoided using the term high frequency conductivity altogether, as it could at best be defined as the reciprocal of the high frequency resistance. The latter, as we have seen, has a physical significance, but we can only attach an indirect meaning to the former.

Effect of Electrostatic Field.

As k_r is defined as limit $(\Delta E \doteq 0) - \dfrac{\Delta D}{\Delta E}$, it will be seen that if the crystal be acted on by an electrostatic field, the value of k_r will in general depend on the value of E and the previous history of the electric field in the crystal. In fact k_r will very nearly be the slope of the D, E curve for any given value of E. In order to measure the value of k_r as suggested above the crystal was carried through a regular hysteresis cycle in the manner described by Valasek.[1] At several points on the normal induction curve, as well as on the hysteresis loop proper, the inductivity was determined as before, and the results obtained for two different plates are shown in Figs. 5 and 6. The dotted line (Fig. 5) shows how k_r varies with increasing field. It first reaches a maximum and then gradually declines until a field strength of 2,200 volts per cm is reached where it begins to increase again. When the field reaches a value of 2,750 volts per cm the crystal begins to conduct, the capacity approaching infinity. The field is then gradually decreased and the capacity reaches a maximum slightly above

---

[1] Loc. cit.

zero field. Below zero field it gradually decreases down to the largest negative field used. On increasing the field again towards zero the capacity increases in a rather irregular fashion up to a maximum for a positive field of 250 volts per cm. For higher field strengths the capacity gradually decreases until the region of conductivity is reached, where it again starts to ascend. Fig. 6 shows a similar curve for another plate of the material, the normal induction curve being obtained this time by applying the electric field in the opposite direction along the ă-axis. The condenser became conducting after the field had been reversed and had reached a value of slightly over 3,000 volts per cm. At 3,800 volts per cm the capacity was increasing indefinitely.

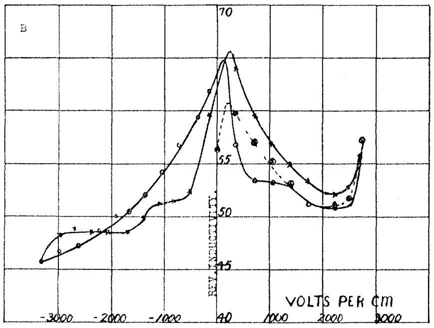

Fig. 5.

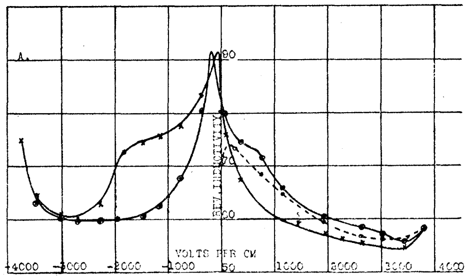

Fig. 6.

The increasing inductivity and conductivity occurring only in one direction of the electric field can be explained by the natural polarization of the crystal along the ă-axis. This natural electric field adds on to the external field in one direction and weakens it in the other direction. Hence the dielectric will break down in one direction for a smaller field than it will in the opposite direction. If there were no natural polarization the two maxima would be equally distant on either side of the axis. Hence the distance along the abscissæ out to the ordinate bisecting the distance between the maxima will give a measure of the natural field. For one plate the value is 125 and for the other 175 volts per cm.

Non-uniformity in Crystal.

In the last section the behavior of two different crystals was discussed and a glance at the curves will show a great difference in their characteristics. In Fig. 7 three inductivity-temperature curves are shown

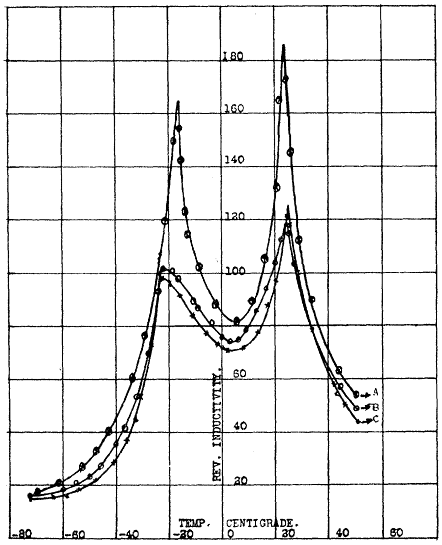

Fig. 7.

for three different plates. The frequency used was the same for the three plates, namely, two million cycles per sec. This frequency was sufficiently removed from the natural periods of any of the samples, so that effects due to different linear dimensions could be neglected. Two of the samples show very similar relations, but the third is radically different in that it reaches much higher maximum values and also has a higher value at 0° C than either of the other two. As these samples were cut from the same crystal and were used under exactly the same conditions we must attribute these results to the lack of uniformity in the crystal so far as the natural polarization is concerned.

In this paper I have not dealt with frequencies above 4×10^7 cycles per second. The resonance method used here is not very applicable to higher frequencies, but other methods are possible whereby the dielectric constant can be measured for wave-lengths down to one cm. As the natural periods of the doublets lie somewhere in the limiting radio frequency range, experimentation in this region ought to be very fruitful. The fact that the value of k_r remained constant up to the limiting frequency used implies that the grouping of doublets in micro-crystals apparently does not exist. In the case of reversible permeability measurements made on iron, resonance values were found in the neighborhood of 10 million cycles per second, in addition to the resonance values directly traceable to molecular vibrations at frequencies of about 10^{10} cycles per second.

I wish to express my thanks to Dr. W. F. G. Swann for his active interest in this research and to Dr. J. A. Valasek for his kindness in putting crystals and crystal-cutting apparatus at my disposal.

University of Minnesota,
September 16, 1922.