SOME EXPERIMENTS IN PIEZO ELECTRICITY WITH ROCHELLE SALT CRYSTALS

THESIS FOR THE DEGREE OF
MASTER OF SCIENCE

HORACE V. CRANDALL

1933

Pyro - [illegible] - electricity

Tide, Rivers, and crystals

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Thesis submitted to the faculty of Michigan State
College of Agriculture and Applied Science

by

Horace V. Crandall

Candidate for degree of Master of Science

June 1933

CONTENTS

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INTRODUCTION

The selection of the topic of piezo electricity in Rochelle salt was not made because of its research possibilities. Neither was it because the phenomena are something new, for piezo electricity has been known for well over a hundered years. Rather, was it selected to bring to the attention of many the possibilities which lie in the everyday application of these phenomena.

In this field much credit is due the Brush Development Company of Cleveland Ohio, to whose work many references will be made in this paper. It was this company that first successfully commercialized the production of Rochelle salt crystals for such applications and who are now making splendid advancements in the field.

To the many scientific investigators of the phenomena is due much credit for making possible this new field of industry. They are mentioned with full credit in the historical presentation to follow.

May those who read this paper find in it, in some measure, the satisfaction and pleasure which has been the author's upon the successful completion of this work.

INTRODUCTION

HISTORY OF PIEZO ELECTRICITY

GENERAL TREATMENT

The Curies are accredited with having discovered the phenomena of piezo electricity in 1880 - 1883. However,these phenomena have been known for well over a century. In 1820 - 1833 A.C. Becquerel made many experiments with piezo electricity and tested a large number of substances,crystalline and otherwise,for these effects. Even as far back as 1703 we find records of a Dutch jeweller heating tourmaline crystals on embers and finding that they attracted small bodies to them upon cooling. This discovery, the first recorded,was,as many such findings are,purely accidental. And yet it would be interesting to know just what the man was actually searching for at the time.

The discovery of the Dutchman,coming as it did before the existance of two kinds of electricity was known,had no significance attached to it and went unpublished except for the mere mention,anonymously,in a German publication,a small pamphlet published in 1707 under the title of "Curious Speculations in Sleepless Nights". Notice was given to the extent that from then on tourmaline became known as the electric stone or as the Ceylon magnet.

From 1880 to 1883 the Curies,later of Radium fame,made many experiments with piezo electricity. They demonstrated that tension and compression have the same effect as heating and cooling respectively. A squeezed crystal of tourmaline behaves electrically as a cooling crystal,and the expanding crystal resembles a heated one. To distinguish the various

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phenomena the electrification due to pressure was designated as piezo electricity; piezo from the Greek piezein meaning to press or pressure. The generation of electricity by heating was termed pyro electricity; pyro,also,from the Greek word pur meaning fire. It is generally believed, however,that piezo and pyro phenomena are due to the same or at least to similar reactions in nature.

The Curies published in 1880 the results of their experiments with quartz. These included a quantitative measurement of the amount of electricity generated by unit pressure along various axes of the substance.

In 1881 Lippman predicted from mathamatical considerations that if quartz was subjected to an electric field, deformation of the quartz would result. This was tested by the Curies and found to be the case. They pointed out that from the law of conservation of energy any piezo electric substance,which acts as a generator of electricity in response to mechanical motion,will act conversely upon application of electric potential. The first practical application of this converse principle was made by the Curies in the form of an electrostatic voltmeter.

The honor of being the first to suggest acoustic applications of piezo electric substances goes to Roentgen of X-ray fame. This came in 1890 as a result of his study of torque as produced in several quartz cylinders when subjected to electrical charges.

In their investigations the Curies made determinations of the piezo electric constant (the charge per unit stress) for many crystalline substances,among which Rochelle salt possessed by far the greatest.

Little was actually done in this direction until the Great War came, with its submarine warfare. Then many investigators took up the task of finding some means of detecting these death dealing monsters from the depths in time to thwart their plans. The application of the piezo electric properties of these crystals,coupled with the newly invented three element vacuum tube as an amplifier of the small potentials from the crystals,was the contribution of the brilliant French investigator,Langevin. With the crystal-tube combination at both input and output ends on the circuit,he was able to detect the presence of submarines by the supersonic vibrations given off by the boat as it progressed through the water.

Although he was interested mainly in super-audible frequencies,he demonstrated the possibilities of these crystals in the audible range.

PIEZO ELECTRIC ROCHELLE SALT

Some of the first investigations of piezo electricity and its possible applications in Rochelle salt crystals was made by the Western Electric Company about 1919. A.M. Nicolson, writing on the subject of "The Piezo Electric Effect in the Composite Rochelle Salt Crystal" in the Proceedings of the American Institute of Electrical Engineers for October 1919, reached very definate conclusions about the possibilities of commerdial application of such crystals. The crystals of his investigations were characterized by their lack of homogeneity and the inclusion of mother liquor. Experience has shown that, due to deterioration and lack of uniform properties in such crystals, only clear and homogeneous crystals can be used with anything near success.

Let us consider some of the early investigators of these crystals, beginning with the work of Joseph Valasek, of the University of Minnesota, and his associates. This work was published as early as 1921 in the Physical Review of that year. Various other articles have since appeared in the same publication giving further advancements and findings of these men. Let us consider first Mr. Valasek's article as published in the 1922 Physical Review in which he says in effect:

We shall begin by reviewing some of the general facts concerning Rochelle salt. This substance is a double tartrate of sodium and potassium, having a chemical formula $Na\ K\ C_4\ H_4\ O_6\ \cdot\ 4\ H_2O$. It has a molecular weight of 282.19 and a specific gravity of 1.77. It crystalizes in the

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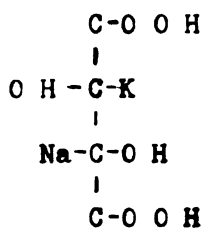
ortho-rhombic system,showing sphenodial hemehedrism. The crystalline form appears as trimetric prisms. It is optically active both as a crystal and in solution. The structural formula appears as:



This form has the usual type of symmetry occuring in optically active carbon compounds. The crystal is brittle and soluble in water,thus permitting cutting and polishing with water. Since it cracks easily due to too rapid or unequal temperature changes,it must be handled carefully. It cannot be heated above 53 degrees centigrade as a crystal,since at this temperature it transforms into water and single tartrates of sodium and potassium,with the absorption of heat.

With this brief and general knowledge of Rochelle salt let us see what has been done with it during the recent investigation of its properties. Assuming we have successfully grown the crystal,a not altogether easy task,the preparation of it for such tests as we may see fit to make is quite well described by Mr. Frank Isley in the Physical Review of 1924. A block of crystal is cut as shown in figure 1. Since it has been known for some time that the maximum activity is in the plane of the (b) axis perpendicular to the (a) axis we will select this plane for our examination. We will then employ the usual method of cutting with a wet string and grinding by means of emery powder and water. The finishing polish is

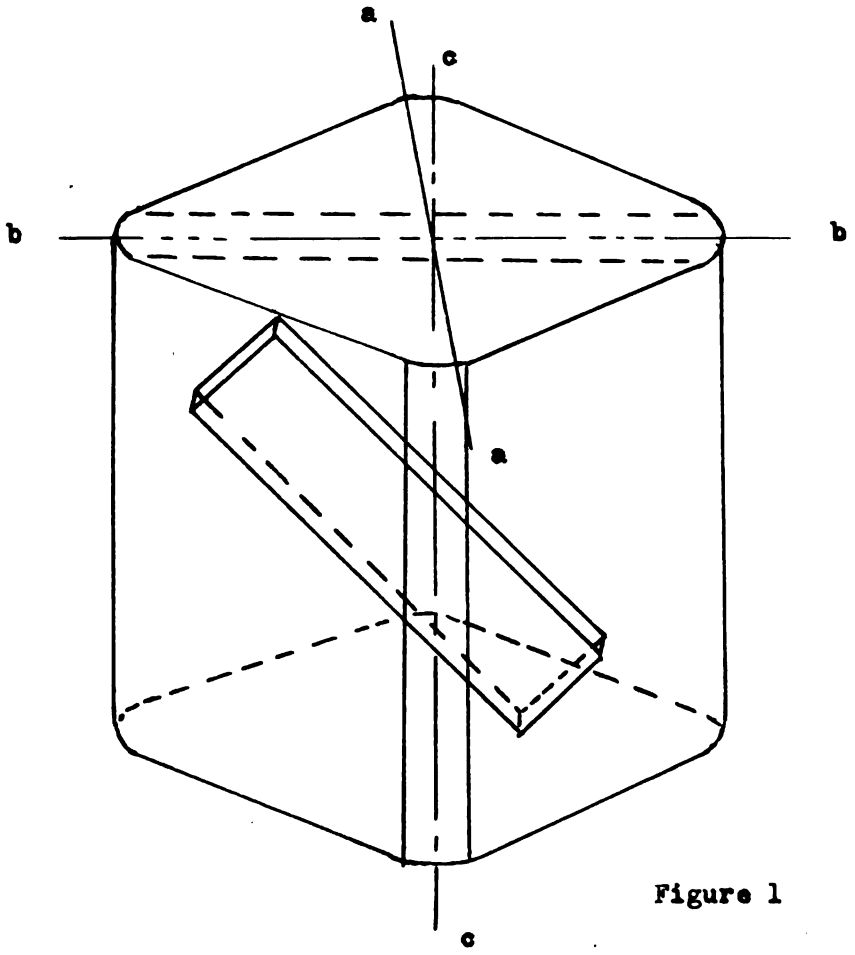


Figure 1

given by water and a ground glass plate. After shaping,the crystal is coated with asphaltum cement,which is made from asphaltum gum and benzine. This cement in no way alters the crystalographic structure of the crystal and reduces to a minimum the change in vapor tension. A piece of tin foil or lead foil is also cemented to each side of the crystal to which is attached a lead-in wire for applying or removing the potential. With such a prepared crystal and a delicate weighting instrument the crystal is subjected to pressure between two brass plates. An electrometer can be used to measure the potential generated. A vacuum inclosed chamber with heating and cooling facilities is necessary for temperature determinations. With the apparatus here indicated,Mr. Isley obtained data for the set of curves as shown in figures 2,3,and 4.

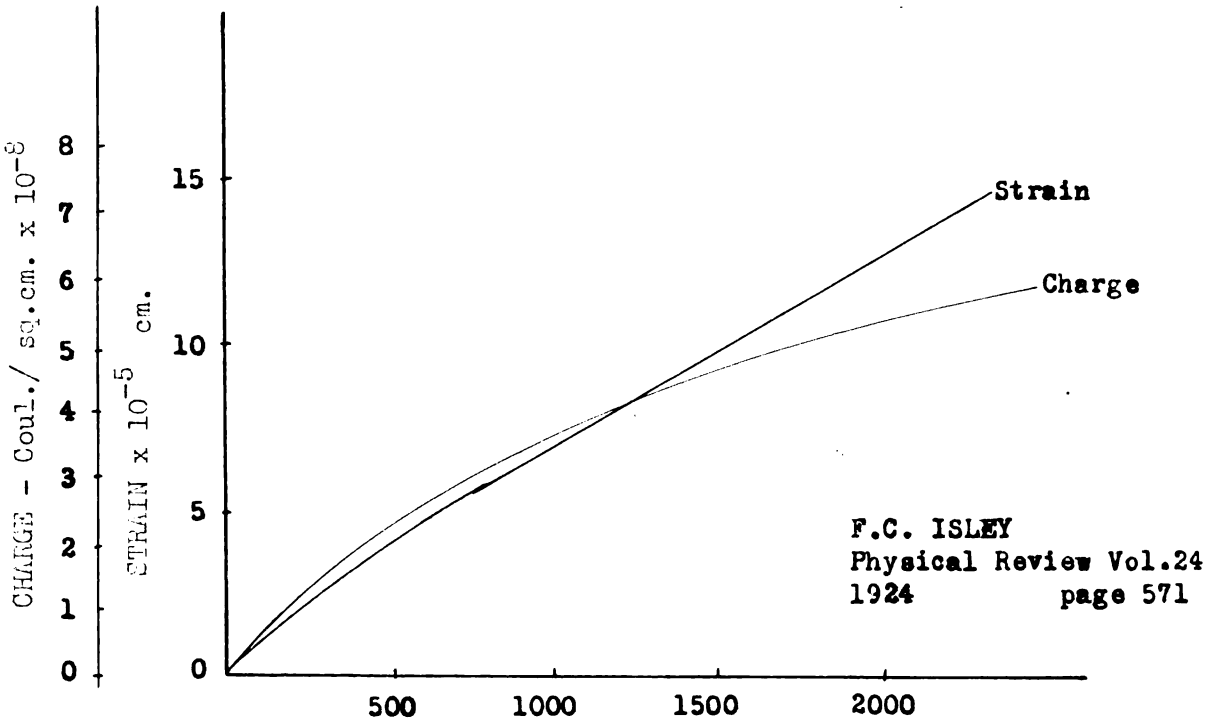


Figure 2

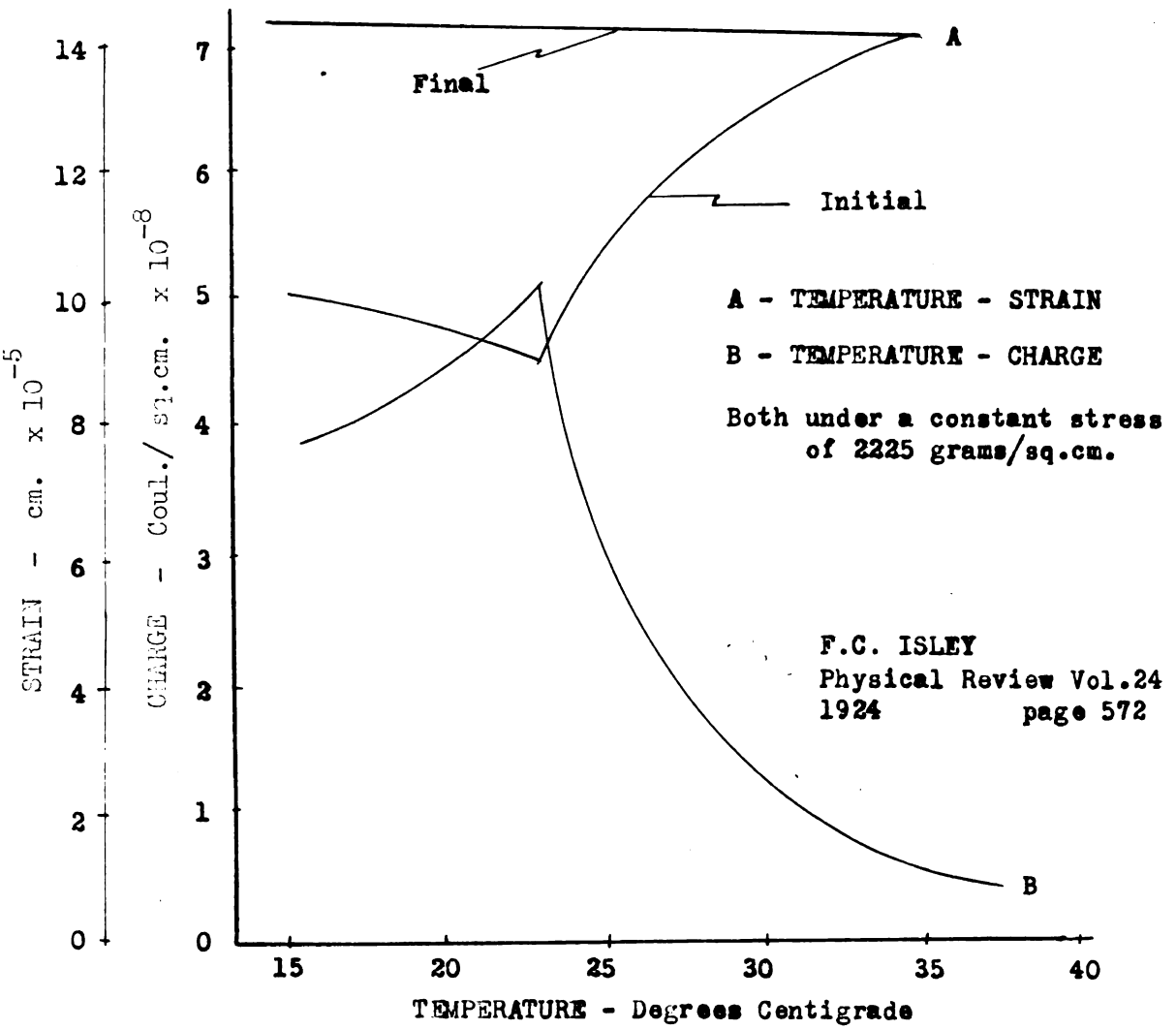
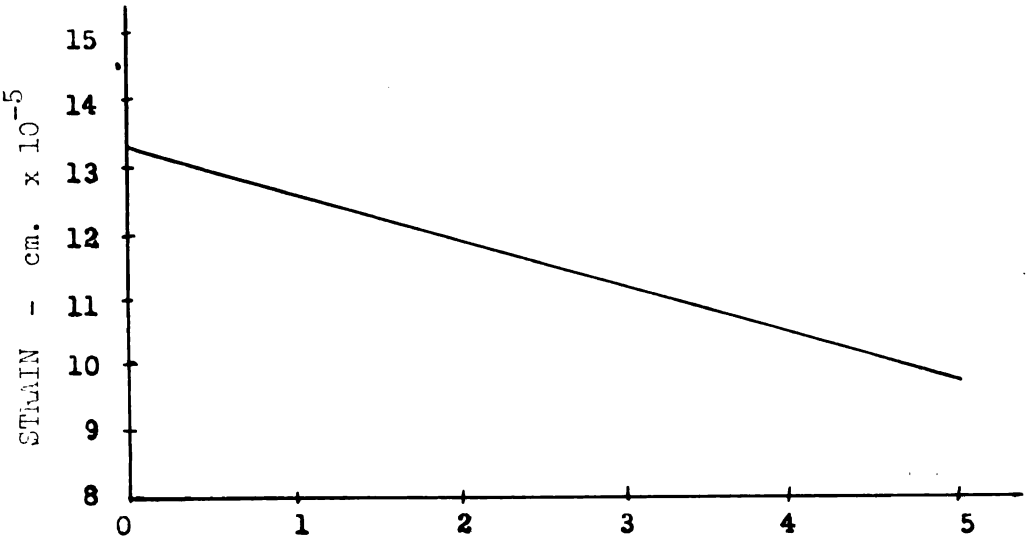


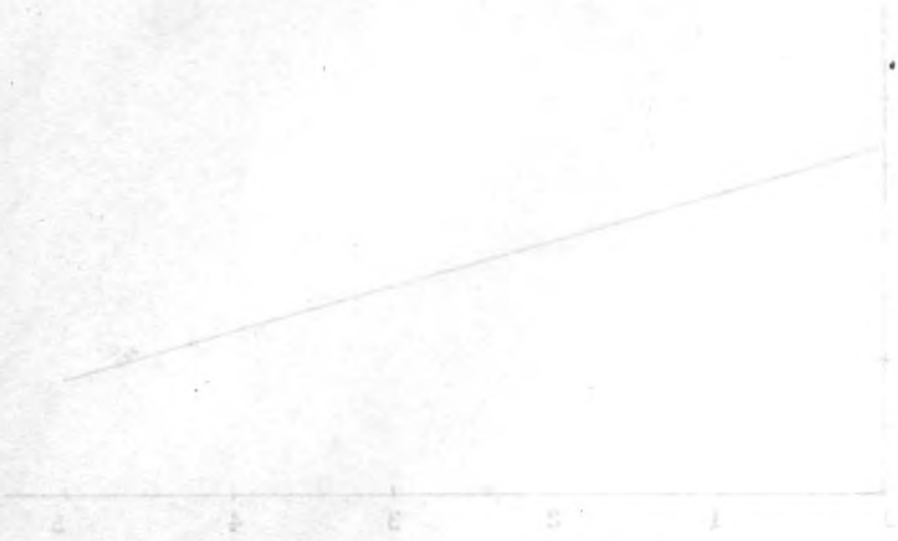
Figure 3



F.C.ISLEY
Physical Review Vol.24
1924 page 573

Figure 4

Following a very similar procedure Mr. John G. Frayne made another series of investigations using,instead of direct potential polarization, a high frequency alternating potential. He termed the resulting response the "Reversible Inductivity of Rochelle Salt Crystals Under High Frequency Fields" and published it in the Physical Review of 1922. Mr Frayne defines his reversible inductivity as the limit of delta (D) over delta (E) as delta (E) approaches zero,where (D) is the induction and (E) the electric field in the dielectric. The plate used for his examination by Mr. Frayne was the conventional cut in the (b) plane perpendicular to the (a) plane to obtain maximum activity. It was mounted between the two plates of a condenser as the dielectric. Capacity determinations were made by the resonance method using a field oscillating at two mega-cycles per second. He found the reversible inductivity (K_r) was about one tenth the dielectric



constant of the crystal as measured by the direct potential methods. Variations of (K_r) with temperature was obtained between -80° C. and 50° C. This relation is shown in figure 5.

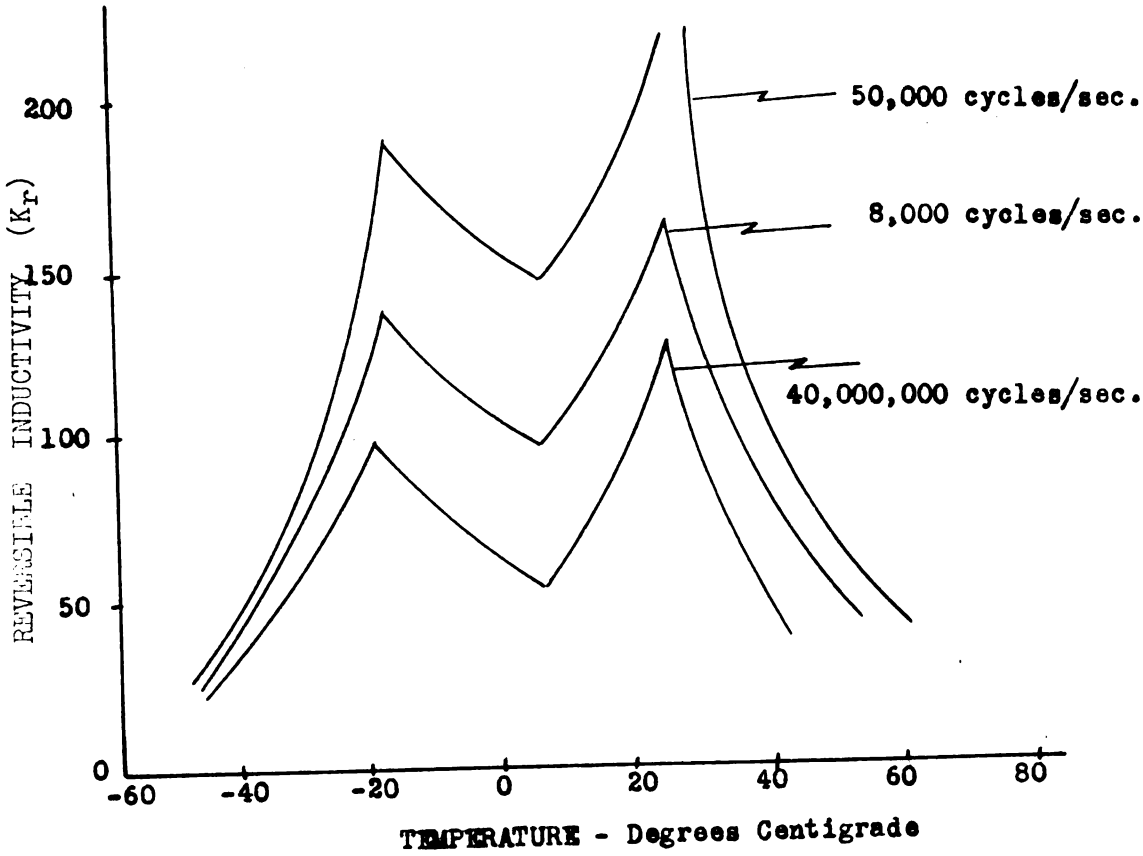


Figure 5

J.G.FRAYNE
Physical Review Vol.21
1923 page 355

It will be noted that the value of (K_r) increases with temperature directly from -80° to -20°. Then from -20° to 4° it decreases. From 4° to 23° it again increases and from 23° to the melting point of the crystal it decreases. The concave portion of the curve between -20° and 23° corresponds to the range of greatest piezo electric activity. The dielectric constant as measured by the direct potential method varies in the opposite

manner between -20° and 23°, being a maximum at about 4°, as shown in figure 6, which was taken from an article by Joseph Valasek in the Physical Review for 1922.

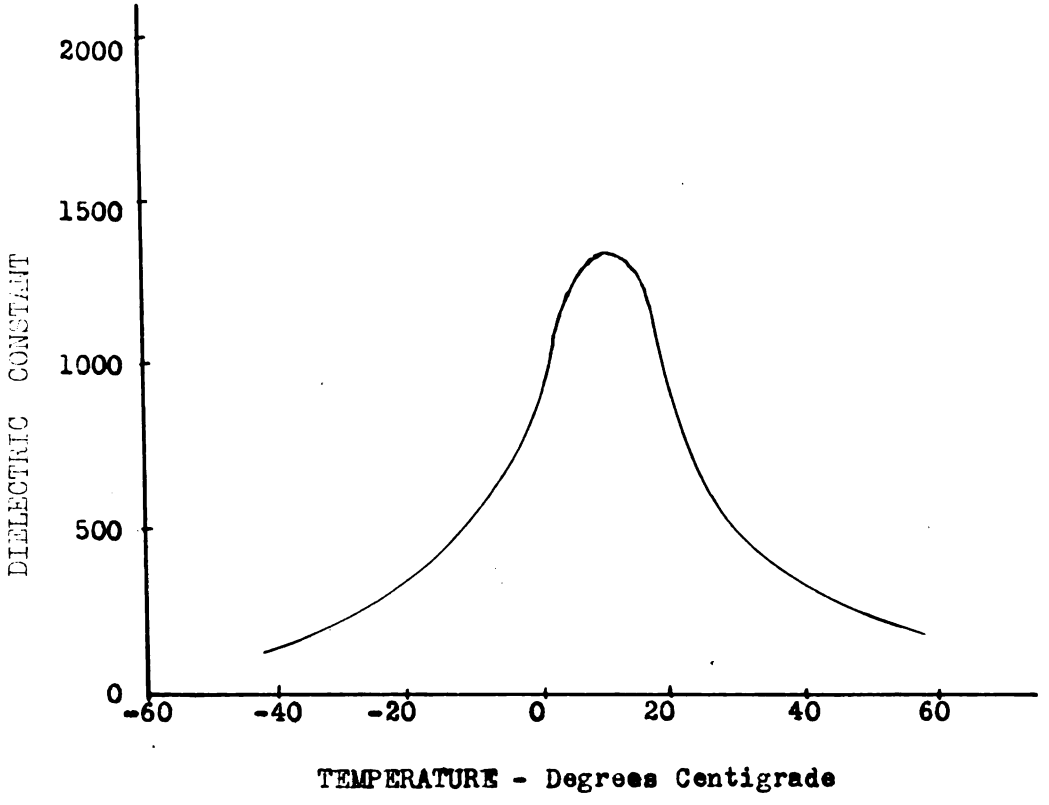


Figure 6

JOSEPH VALASEK
Physical Review Vol.19
1922 page 488

The value of (K_r) was measured for different states of polarization of the crystal. The high frequency potential was superimposed on a direct field. Different samples of crystals used showed permanent polarization effects. The crystals became conducting and the capacity infinite for smaller values of field strength in one direction than in the other. Curves showing the variation of (K_r) with polarized fields are shown in figure 7.

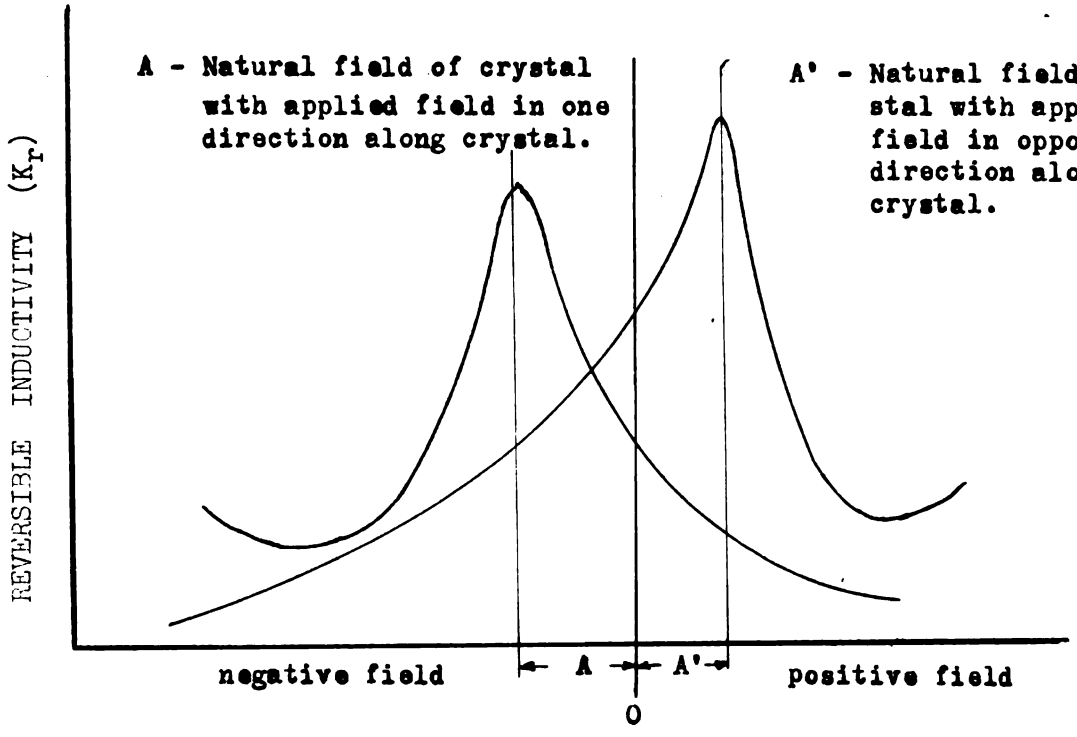


Figure 7

J.G.FRAYNE
Physical Review Vol.21
1923 page 357

The fact that Rochelle salt crystals experience fatigue has been observed by various experimenters. W.G. Cady in his report to the National Research Council in May 1918 described these effects as not of a permanent nature but existing for many hours. Valasek finds that this may account for the capricious manner in which the piezo electric modulus varies in response to a number of conditions such as temperature,humidity,and previous history with regards to electrical and mechanical treatment.

In the equation for charge (q) on a crystal of length (l),breadth (b), and thichness (d),for a total force (F) on the end of the crystal we have:

$$q = -S_{14} F (l / 2d)$$

Here $-S_{14}$,the piezo electric modulus,varies from practically zero to 4.0×10^{12} esu/dyne. He finds that humidity increases the response directly and that temperature varies the activity as shown in figure 8.

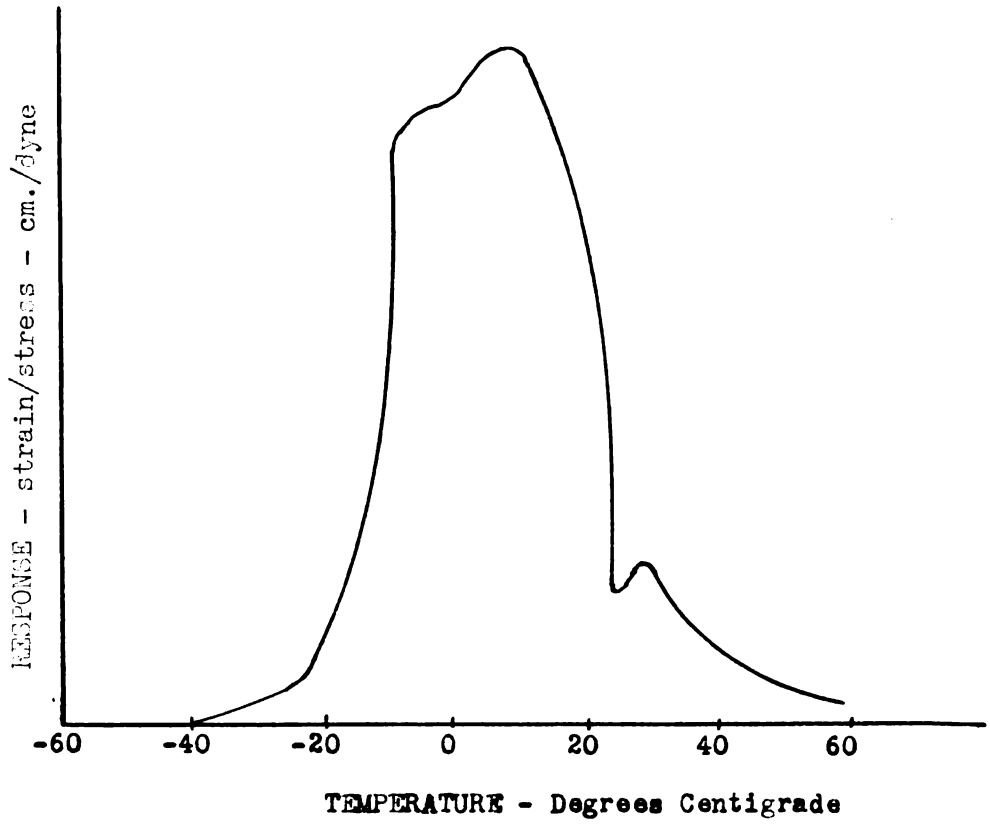


Figure 8

Joseph Valasek
Physical Review Vol.19
1922 page 485

The actual fatigue and recovery of a crystal is shown in figure 9 with accompanying explanations.

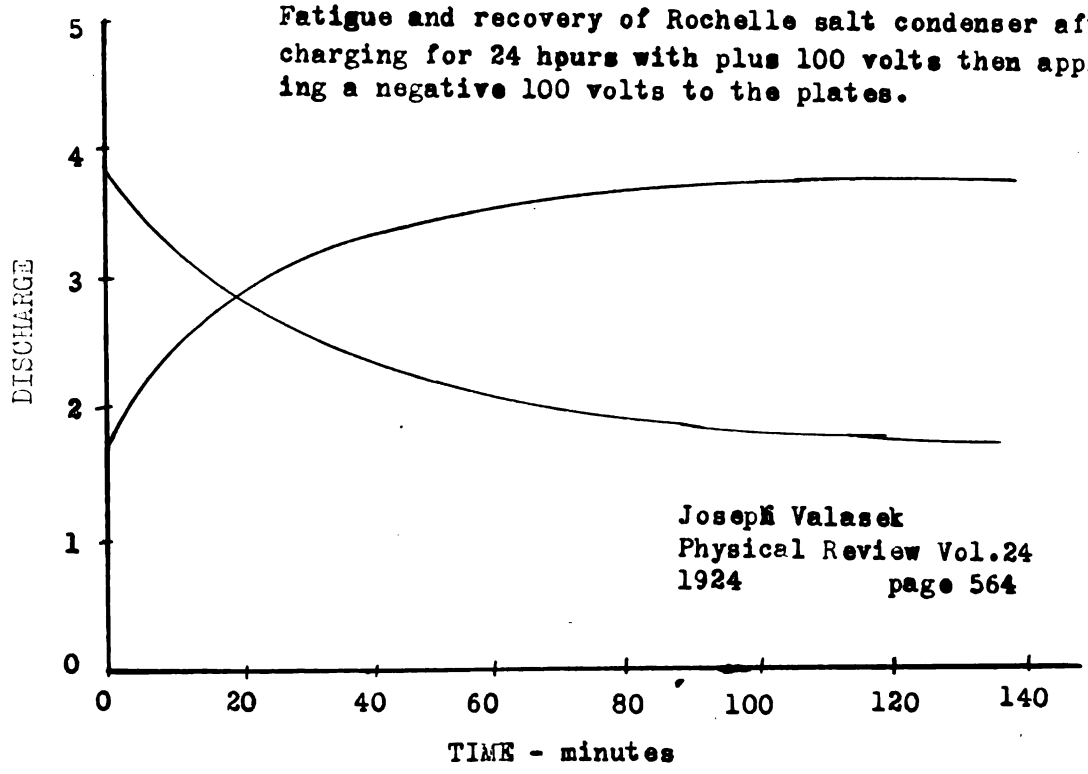


Figure 9

The variation of the piezo electric constant,that is the charge resulting per unit applied force,is shown in figure 10,which curve is due to Mr. Valasek and was taken from an article by him as published in the Physical Review for 1922.

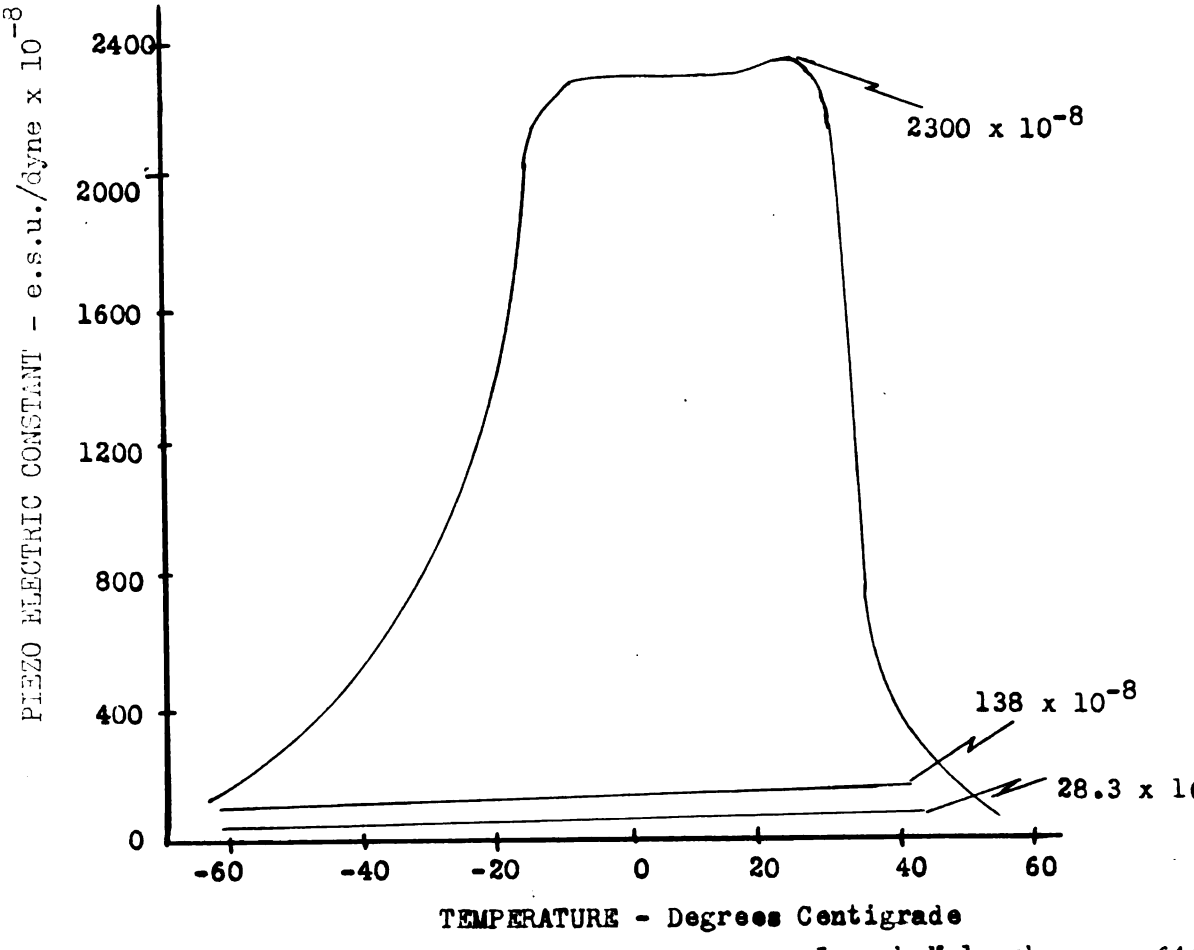


Figure 10

Joseph Valasek page 64
Physical Review Vol.20

Many attempts at explanation of the phenomena of piezo electricity have been made by various authorities. Perhaps the best presented to date is that of Joseph Valasek as it appeared in the Physical Review for 1924. Mr. Valasek states: "It is interesting to note that the high dielectric constant of Rochelle salt can be attributed to the water of crystal-

ization without requiring greater displacement than the distances between molecules."

"The maximum polarization obtainable is less than 15,000 e.s.u./cm.^{3} There are four molecules of water for every molecule of Rochelle salt. The crystal lattice of each molecule of water may be regarded as equivalent to three ions,two hydrogen and one oxygen. Suppose the restoring force/ unit displacement/unit charge is the same. Then:

$$P_{max} = 16\ N\ E\ R$$

Where: P_{max} - the maximum polarization

E - the electronic charge

R - the displacement

N - the number of molecules of salt / cm.^{3} which is equal to 3.76×10^{21}.

"This gives (R) a value of 5×10^{-11} cm. However the value of (R) from a consideration of the number of molecules present (N) is only (R) = 3×10^{-7} cm. as the distance between molecules. Thus it is possible for the water of crystalization to furnish the ions for internal conduction and yet not move out of the molecule"

In support of this theory Mr. Valasek offers the experimental results shown in figure 11,as taken from the Physical Review for 1922. In all fairness,however,Mr. Valasek states: "The decrease in maxima and also their displacement is in the same direction as,and may be due entirely to, the effect of the different dielectric properties of the crystal,and of the dehydrated layer. In other words the presence of a layer of inactive

dielectric of relatively low specific inductive capacity will diminish the charge on the plates due to polarization of the active layer, and thus decrease the piezo electric response. It will also diminish the effective field across the active layer making it necessary to increase the potential difference between plates to produce the same field across the inner layer, thus shifting the position of the maximum activities."

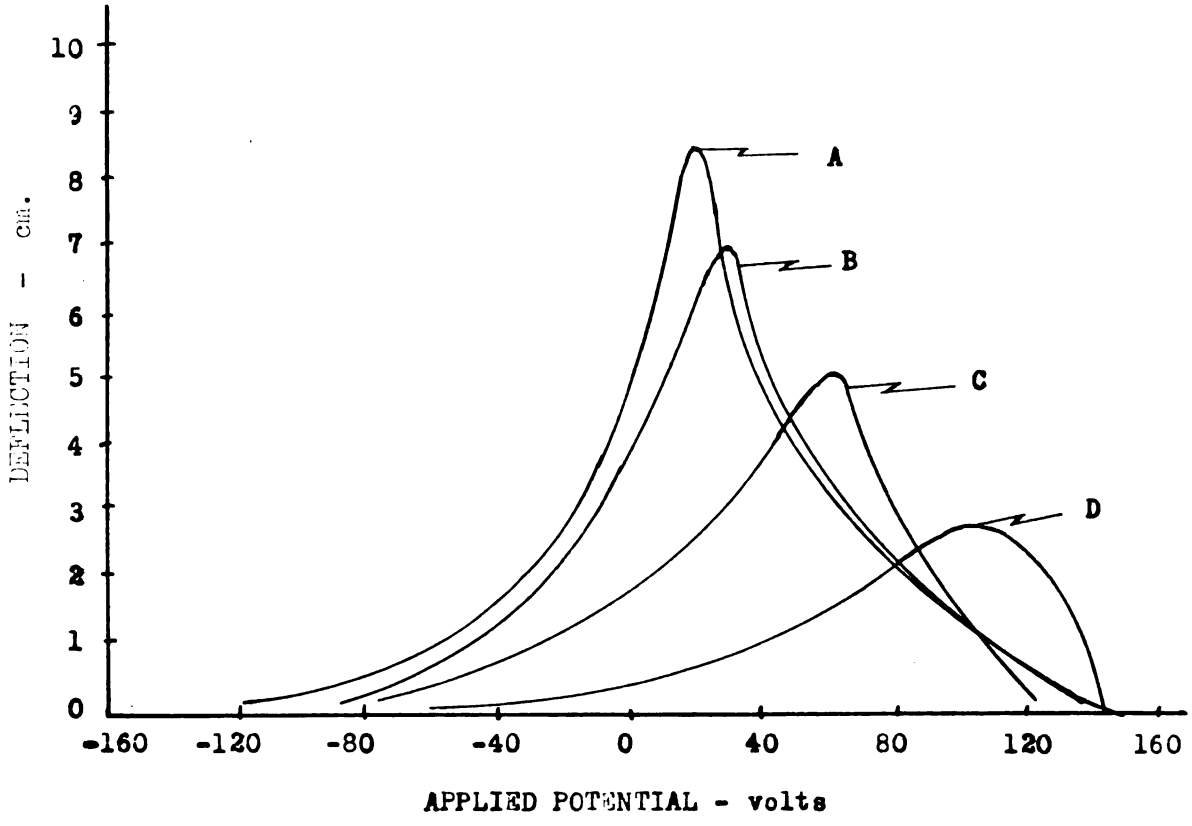


Chart showing the effect of drying upon Rochelle salt crystal.

A - response before drying.
B - response after drying in chamber with P_2O_5 for 1 day.
C - " " " " " " " " 3 days.
D - " " " " " " " " 12 " .

Joseph Valasek
Physical Review Vol.19
1922 page 482

Figure 11

Despite Mr. Valasek's frank presentation of the possibility of the results of figure 11 being due to some other causes than the actual loss of piezo electric properties, by loss of the water of crystalization, the fact still remains that the loss of this water does decrease the activity, whereas the increase of water present, in the form of increased humidity, gives rise to greater activity. This in itself is a fair proof that the water of crystalization is, in a great measure, responsible for the phenomena of piezo electricity, as observed in its maximum activity in Rochelle salt.

EXPERIMENTAL AND CONSTRUCTIONAL DATA

GROWTH OF CRYSTAL

In order to carry on any experiment with Rochelle salt crystals, some source of crystal supply must be at hand. Since the whole procedure was one of experimentation it was decided to do some work on the growing of crystals at home,so to speak,with what facilities there were at hand. Being quite unfamiliar with the chemistry of crystal growth,the author was at a loss to know just how to begin. All articles on the subject dealt very briefly,or not at all,upon this very essential phase of the work. After exhausting all available articles without finding anything of practical value,a review of local possibilities was made. Upon the suggestion of some of the students majoring in chemistry,Mr. Eck,of the Chemistry Department of the College,was interviewed on the subject. His suggestions were of a quite general nature. He stated that at no time had he found it necessary to produce crystals as large as those required for this work. His suggestion was that an attempt be made to grow then from a slightly supersaturated solution of the salt by the process of slow evaporation,or by a temperature gradient method with a more concentrated solution. As for the latter method no data was available as to just what the temperature gradient should be in degrees per hour or day,nor as to how saturated the solution should be.

A review of some of the early literature on the subject,however,led to the discovery of some previously overlooked suggestions on this latter method of growth in an article by A.M. Nicolson in the Transactions of the A.I.E.E. for 1919. This article stated,in effect,that Rochelle salt

crystals are grown from perfect nuclei possessing definate form. The nuclei or seed crystals are immersed in a saturated solution of salt under identical conditions of temperature. A density of 1.33 at 50° C. can be used conveniently and the seed crystal, previously warmed to the same temperature as the liquid, should be inserted between the temperature of 38°C and 35° C. Continuing, Mr. Nicolson says, "The crystals may be grown by application of temperature gradients to a saturated solution of the salt, or by concentration brought about by slow evaporation. The former, producing a specific type under the conditions of rapid cooling, is the method preferred. The crystals may be grown in the mother liquor by suspension from a clean thread, by flotation on mercury, or by being laid on a glass plate, the latter two methods being preferred."

With this material as a basis, the production of some Rochelle salt crystals of suitable nature was attempted. A solution was made of the recommended density. Having no seed crystals at first, the solution was set aside and left to cool over night. The next day a plentiful supply of crystals was found on the bottom of the jar. Some of the more perfect of these were removed with care and set aside for future use. The solution was then reheated to the proper temperature, about 50° C. , and allowed to cool to 37° C. At this point one of the small crystals, which had been warmed to the same temperature in an improvised oven made from a hinged cover wooden box with an electrical heating element of the common variety in it, was placed in the solution and the whole allowed to cool as rapidly as it would in the air of the room, until the next day. The inspection of the jar the next day, however, was very disappointing; for not only had the seed crystal grown

but also numerous others had formed and grown so that the entire bottom of the jar was filled with crystals,all intersecting one another and no perfect ones to be found.

The same general procedure was followed on the next trials with the slight variations of incidentals usually made. The temperature of setting the crystal was varied,as was the density of the solution. The temperature gradients were decreased by placing the whole jar in the oven and allowing it to cool more slowly than before. Much care was exercised to prevent jarring and thus forming extra crystals from the supersaturated solution. The results were always the same with the exception of the number of crystals forming at onesetting. The smallest number formed at onetime was three. These were arranged symmetrically about the bottom of the jar,the (c) axes of the crystals forming the spokes of a wheel 120°apart and all intersecting at the center. Of course even this was unsatisfactory for our needs.

The next variation of method was to try the second suggestion of Mr. Nicolson and grow the crystals on mercury placed in the bottom of the jar. This,however,met with a similar fate and no single crystals were grown by this method.

Next the growth of the crystals,as a complete type,was attempted by suspension from a fine wire. This was tied about the crystal and then suspended from a small rod placed across the top of the jar,as shown in the photograph of figure 12. It often happened,however,that the crystal would dissolve a little when first placed in the solution and would thereby become small enough to fall from the retaining loop of wire. It then

FIGURE 12

became necessary to reset the jar with a new seed crystal. Here was a point to be improved upon and several other methods of fastening the seed crystal to the wire were tried. The best and most satisfactory of these was that of melting a small hole in the end of the crystal with a hot pin, inserting the suspension wire, and allowing the crystal to cool and harden about it. By so doing, and inserting the wire sufficiently deep into the crystal, it became impossible for the crystal to fall from the wire until it was practically all dissolved, a thing which rarely happened. This method of suspension also made a smaller hole in the crystal, when sectioned, than did the first method. This was a great help later in being able to use more of the crystal, since large crystals seemed very difficult to obtain.

This suspension method of growth proved the best of all methods tried, as it kept the seed crystal away from the growth which invariably took place at the bottom of the jar, the thickness of which can be seen in figure 12.

It was found necessary, in some cases, to place a piece of copper plate just under the surface of the solution, suspended by wires from the top of the jar. This was to catch the little crystals which are continually forming on the surface of the solution and growing until they become large enough to sink, for in so doing they often strike and adhere to the seed crystal, producing a parasite which grows with it and prevents formation of perfect specimens.

After having a fair degree of success with this method of growth, and having accumulated some twelve or fifteen crystals, from one and one half to two and one half inches long and an inch to two inches thick, the next problem was some manner of cutting. Several methods were tried, which will

be discussed in detail later on. Upon the first cutting it was found that what had appeared to be a clouded portion of the crystal was really a smallpocket including some of the mother solution. Naturally this spoiled the whole crystal,since the pocket had a peculiar faculty of always forming at right angles to the plane of greatest piezo electric activity,namely the (b) plane,thus separating the section into two or more parts,which were then too small to be of use. Besides being small,these sections were filled with tiny cracks and crevices due to the unequal growth of the crystal. Since all crystals grown in this manner were more or less clouded or cracked it was necessary to find some other method of growth which could be better controlled.

Luckily,at this point,the attention of the author was called to an article in the Journal of the American Chemical Society for 1919,entitled "A Method of Growing Large Perfect Crystals from Solution", by R.W. Moore. In this article Mr. Moore put forth a method which he had used with great success for growing Rochelle salt crystals of large size and perfect clearness and symmetry. This method was,in effect,as follows: Prepare a saturated solution of Rochelle salt at a temperature of between 35°C.and 40° C. Then,after removing the excess salt from solution,heat to about 7° or 8° above this temperature and filter,being careful that the temperature at no time falls below 4° or 5° above saturation temperature. The seed crystal should then be inserted and the jar placed in a large container of water at .5° above the saturation temperature of the solution. The water is allowed to cool to saturation temperature,and the thermo-regulator and heater arrangement then maintains it at this temperature until read-

justed. The cooling gradient recommended was .1° the first day, .2° the second day,and from .3° to .6° each succeeding day. If more than one crystal is grown at a time in the same jar,this gradient may be increased. Mr. Moore furnished with his article a chart,of specific gravity of the saturated solution of Rochelle salt plotted to temperature,as a guide in preparing the solution for a certain temperature range. This chart is reproduced here,figure 13,with all credit given Mr.Moore for its origin.

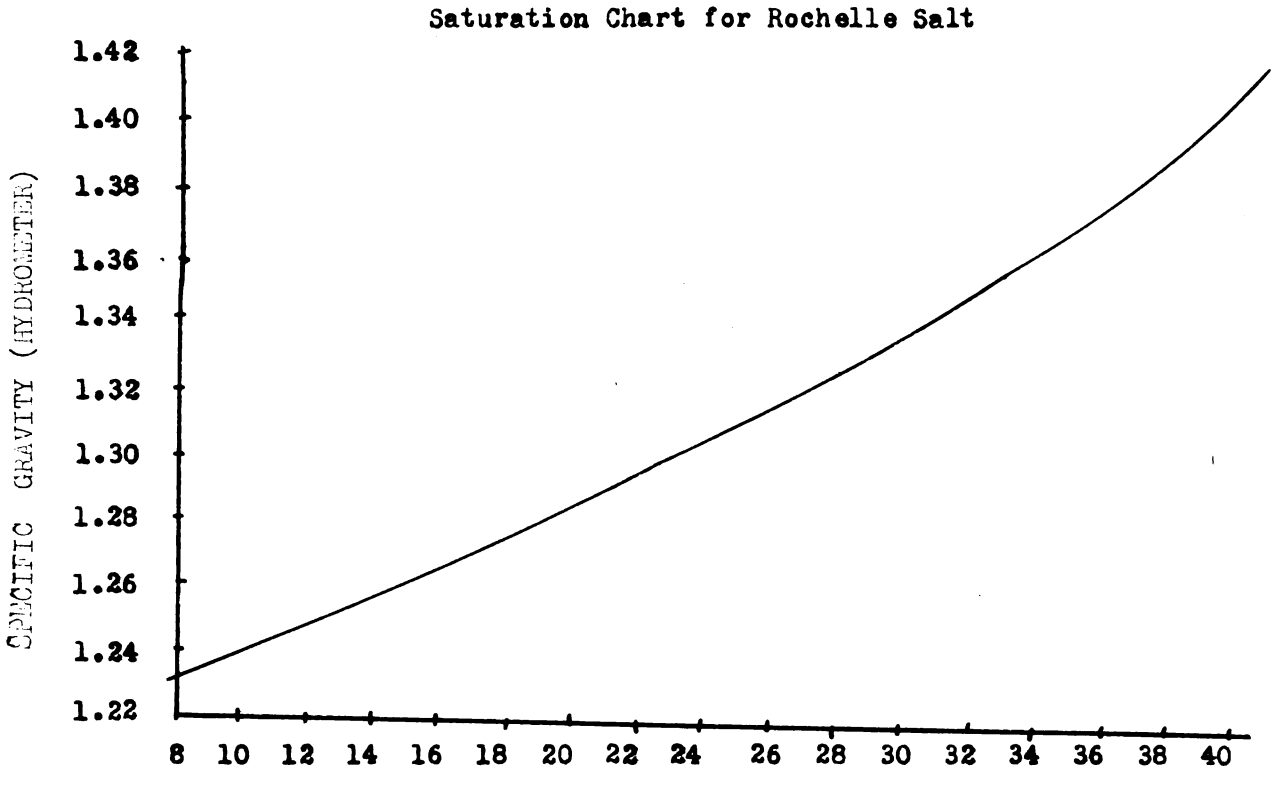


Figure 13

It will be noted that for the range indicated as best by Mr. Moore,namely between 35° and 40° C. ,the specific gravity of the saturated solution is between 1.37 and 1.42 as compared with the value of 1.33 at 50° as given

by Mr. Nicolson. It has been the experience of the author that the values as given by Mr. Moore work much better than those suggested by Mr. Nicolson.

Having no facilities for the control of temperature to an accuracy of .5° available in the Engineering Department, the assistance of the Chemistry Department was again solicited. In this department were found so called 'mercury regulators' which would maintain a constant temperature to .005°, provided the bath was kept in constant motion. The jarring caused by this agitation of the bath, however, would have proved fatal to the growth of single perfect crystals. As the necessary temperature regulation was only .5° it was thought probable that this instrument would maintain the desired regulation by means of convection currents alone, withoutagitation, providing the heating coil was placed in the bottom of the tank. Through the generous cooperation of Dr. Ewing and Mr. Eck the apparatus was assembled, as shown in figure 14, and a trial run was made.

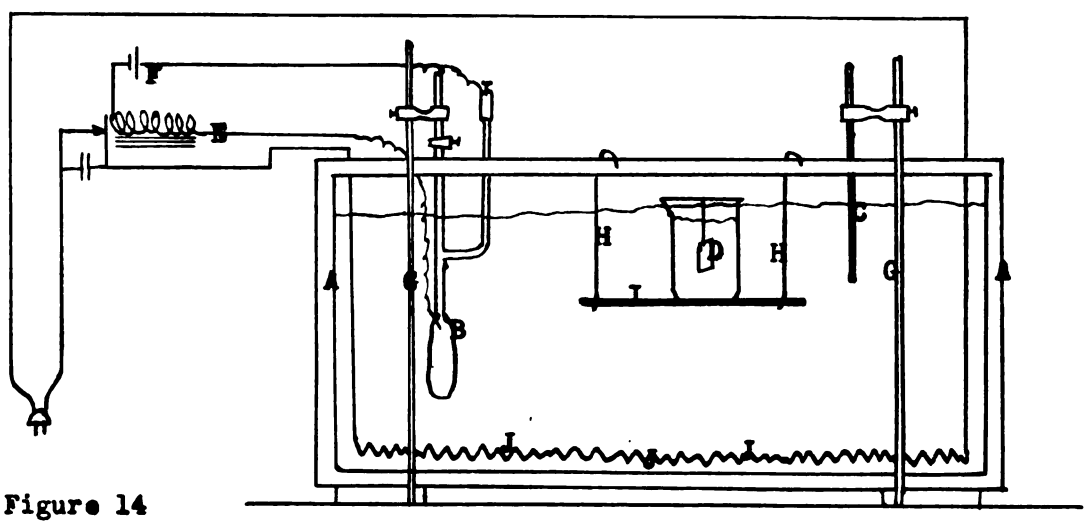


Figure 14

A - Glass Aquarium
B - Mercury Regulator
C - Thermometer
D - Beaker containing solution and crystals
E - Relay
F - Dry Cell
G - Ring Stands
H - Copper strips supporting I
I - Sheet Rock plate holding beakers
J- Electric Heating Element

For a setting of 37° the temperature range was from 36.5° to 37.5°. This was thought to be satisfactory. The crystals were then set after the method described by Mr. Moore,and developments eagerly awaited.

The crystals grew very well for the first few degrees drop. Then the limit of adjustment of this regulator was reached. In attempting to readjust the regulator the capillary tube at the side was broken off. This appeared quite a catastrophe in view of the fact that it was borrowed equipment. It must be repaired! Having had a little experience in glass blowing at the Physics Department,the author decided to attempt the repair of the broken regulator. The attempt was so successful that it was decided to make a regulator with a greater range of adjustment. This was done very nicely with the generous assistance of Professor Miller of the Physics Department. The finished regulator is shown at the right in figure 15. At this point a brief description of the regulator,and its more sturdy companion on the left of the same figure,is not out of place.

The bulb at the bottom was made from a 50 cc. round bottom flask. To it was attached a short piece of 1/2 inch glass tubing by drawing down the neck of the flask. The L tube on the right was then inserted. It was made by sealing a four inch piece of 1/16 inch capillary tubing into an eight inch length of 1/2 inch tubing leaving about one and one half inches at one end and about three and one half inches at the other. The long end was bent at right angles just below the end of the capillary tube and sealed into the neck of the flask just above the bulb. This completed the glass work.

The regulator was now thoroughly cleaned,first with gasoline to remove all grease,then with the conventional glass cleaning solution of caustic potash,and finally rinsed with distilled water and dried in an air blast.

FIGURE 15

Then the clean mercury was placed in the bulb filling it to a height of about one inch in the tube proper. The work was then ready for the adjustment device.

It was desirable that the regulator be adjustable over a rather wide range. To accomplish this an adjustable plunger was inserted in the main tube to regulate the height of the mercury in the side tube at a given temperature. This plunger was a leather washer,held between two brass washers,and attached to the end of a small threaded rod by a little bolt inserted in a tapped hole in the end of the rod. The long threaded rod extended out the top of the glass tube through a cap made from a small copper cylinder into which was soldered a brass nut of the same thread as the rod. The cap was then slipped over the end of the glass tube and held firmly in place by a heavy wax. A cast off radio dial was then placed at the end of the threaded rod to afford an easy means of turning it and of marking its setting.

A similar procedure was carried out for the side tube except that the plunger extended only into the one and one half inch space at the top of the capillary. A small iron wire had been soldered on to the threaded rod and this continued on about three inches into the capillary tube. The threaded rod was attached as before to the top of the tube and a smaller radio knob served for its adjustment. The purpose of this fine wire in the capillary was that of completing the circuit through the regulator and as a micrometer adjustment for the plunger.

The regulator was designed to work in conjunction with a relay in the power circuit. The method of operation is similar to that of an ordinary

thermometer with the exception of the electrical circuits involved. Since mercury has a relatively high coefficient of expansion for temperature change,is a liquid at ordinary temperatures,and is an electrical conductor, it is ideal for our purpose. The large quantity of mercury contained in the bulb gives ample expansion for small temperature changes. This expansion may take place in two directions,up the main tube or up the capillary tube. By means of the adjustable plunger in the main tube the amount of expansion in that direction can be controlled at will. Thus by turning down the plunger the mercury is forced up the capillary tube . When it comes in contact with the small iron wire,projecting down into the tube,there is a completed electrical circuit between the cap of the main tube and that of the capillary tube. Thus the caps of the two tubes become the terminals of the regulator which is connected in series with the control relay and a power supply. At this temperature and plunger setting,the relay circuit is closed thereby opening the power or heating circuit. Leaving the plunger set at this point,the mercury will remain in contact with the iron wire until the surrounding media has cooled sufficiently to reduce the temperature and volume of the mercury to a point where the thread of mercury in the capillary tube breaks contact with the wire. The electrical circuit is then broken and the relay opens,closing the heating circuit. When enough heat has been supplied to the system to raise the temperature sufficiently to expand the mercury up the capillary tube to contact with the wire,the whole cycle is repeated and the temperature oscillates between two points. The value of this swing is determined by three factors,the amount of mercury in the bulb,the size of the capillary tube,and the rapidity with which

the heat is distributed through the water in the tank,or in other words whether the water is agitated ot not.

After the regulator was completed it was roughly calibrated by placing it in a dish of water with a thermometer and heating the water. After contact had been made between the caps,the plunger was raised one turn,and a note was made of the number of degrees rise in temperature required to expand the mercury until contact was again made. This was found to be about 5° per turn,and,as the dial was etched,almost any value could be selected by turning the dial only a few divisions. The micrometer adjustment of the side tube could be added to this and very fine regulation maintained.

Being made of glass,and containing so much mercury,the regulator was quite sensitive to jar. It was so delicate,in fact,that several were broken before one was completed and put in operation. To minimize the chances of the regulator getting a damaging bump while in operation,it was placed in a perforated metal container and packed with excelsior. This,of course,lengthened the time required for the temperature change in the water to reach the regulator and thus increased the swing of the temperature between its limits.

To overcome the difficulties involved in the first regulator a second one was constructed. This was more sturdyly made throughout,having all possible glass parts replaced with similar ones of iron piping,as shown at the left of figure 15. It was built along the same pattern as the original one where ever possible. The only glass used in the construction was the capillary tube,which served two purposes. It magnified the effect of the expansion in the mercury and served as an insulator between the terminals of the regulator.

It was found necessary to place a rubber washer at the end of the plunger, despite the fact that the threaded rod fitted tightly into the inside of the pipe,which had been machined to a smooth surface with only six threads left at the top. Most of the other variations are visable in the photograph. The capillary tube was waxed into the tube with the same heavy wax as was used in the first case. This regulator replaced the original for a short time. For some reason it was difficult to keep a temperature setting with it,presumably due to the leakage of mercury past the washer in the tube. Because of this it was eventually set aside in favor of its more delicate but reliable predecessor,which was used throughout the rest of the experiment.

Having now a regulator of our own,so to speak,it was decided to return to the greater convenience of the Engineering Department to continue the experiments. Here an arrangement of apparatus,similar to that used at the Chemistry Department,was made. A large fifteen gallon crock was substituted for the glass aquarium and a small 10 volt toy transformer and an A.C. relay for the batteries and D.C. relay previously used. The general idea of the arrangement can be obtained from the photograph of figure 16. The crock was insulated with two layers of Balsum Wool on the sides and bottom. The jars of solution were set in the crock on a shelf of sheet rock board which was supported on two hangers hooked over the top of the crock. The heating element was the wire from an ordinary 500 watt heater which had been unwound and coiled back and forth at the bottom of the crock. The regulator was placed in its metal container and rested on the shelf along with the jars of solution. A thermometer was hung in the crock to keep record of the temperature at all times.

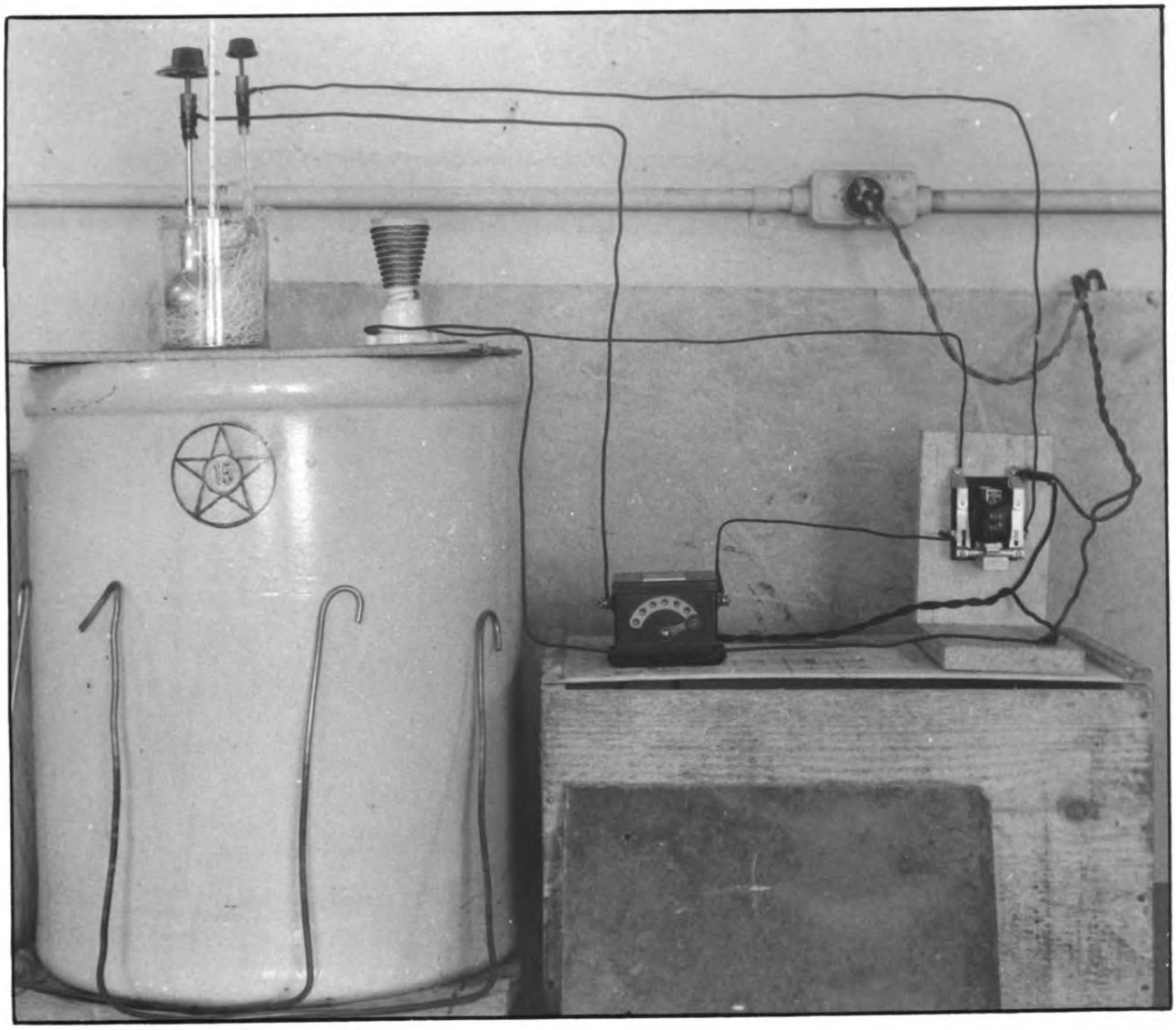


FIGURE 16

With this apparatus, and a plentiful supply of Rochelle salt, some very fine crystals were grown. They were perfect in symmetry and as clear as glass although somewhat smaller than the ones that had been grown before.

Of course accidents will happen, and one of these cost the loss of a large jar of solution. This happened as the waste crystals, which had formed at the bottom of the jar, were being remelted in preparation for a new setting. This was being done by placing the jar in a pan of cold water and gradually warming it by admitting live steam into the water. It seems that the crystals expand a great deal more than glass for a given change in temperature, and, in so doing, invaribly break the container. As this happened when no one was around, the entire jar of solution ran out into the pan of water and was lost. Experience was a very good teacher, but an expensive one, and after that the crystals were broken out of the bottom of the jar with a pick before trying to redissolve them.

SECTIONING THE CRYSTAL

Having grown several crystals of the proper size and clearness,some means of cutting or sectioning these crystals must be found,since the complete crystal is of little value piezo electrically. It was decided to try several methods of doingthis and to use the first grown crystals,since they were more or less clouded and therefore of little value even if successfully cut.The method which had been previously used by experimenters in the field was that of a wet string which was moved back and forth across the crystal, thus dissolving a strip as wide as the string through the crystal. This was a tedious process and not applicable to quantity production. The Brush Development Company of Clevelend Ohio had successfully used machine methods of sectioning these crystals quite similar to those used in woodworking, and with very critical cutting speeds. Exactly what the procedure was,and the value of the critical speed,was naturally omitted in their article in the November 1931 issue of the Proceedings of the Institute of Radio Engineers.

The first woodworking machine tried was the band saw. A jig was used to hold the crystal and this rested on the table of the saw. After cutting half way through the crystal it suddenly cracked the remaining distance and fell apart. As was to be expected,however,the break was not in the desired direction and the crystal was spoiled. Several other trials were made, holding the crystal in different positions,but all with the same result. The crystal always broke apart due to the vibration of the saw or from inability to hold the crystal properly. Since the speed of the saw could not

be changed,or the size of the blade varied,it was decided to try a small hand saw of the coping variety,using a very fine blade. The same trouble was still present. It was impossible to hold the crystal solidly without exerting sufficient pressure to crack it. Holding it loosly permitted it to slip and this invaribly kinked the saw and snapped out a piece of crystal. From the adverse results of these trials it was apparent that some other means of cutting must be found.

The next attempt was to cut the crystal with a wire. The wire soon became covered with the salt and stuck in the crystal. To remedy this the wire was heated by passing an electric current through it. This dissolved the adhering salt and cut the crystal but in so doing it cracked it,due to the unequal expansion near the wire. This eliminated the hot wire as a cutting tool.

Returning again to the idea of a saw for cutting the crystal,a high speed paper disc was tried. This disc was about four inches in diameter and made from the paper cover of a laboratory folder. It was attached directly to the shaft of a motor designed to drive a refacing wheel for lathe use,and ran 17,500 r.p.m. The motor was mounted in the tool rest of the lathe and the crystal was held firmly between two wooden jaws clamped between the jaws of the chuck. The disc was advanced to the crystal by the lathe screw to keep it in perfect alignement. The cutting was satisfactory,from that point only,but the heat developed by the speed of the disc was as disastrous as that from the wire in the previous trial.

Having exhausted the available supply of tools for cutting purposes, the last resort was the old method of using a wet string. This was slow

and tedious,but at least it gave satisfactory results in the way of clear and uncracked sections. A photograph of the improvised cutter is shown in figure 17.

The crystal was held between two soft pine blocks which slid in a groove cut in the supporting piece. These blocks were firmly held together by adjustable thumbscrews through angle irons attached to the base. They were soft enough to prevent breaking the crystal yet rigid enough to hold it securely. The thread,which was used to cut the crystal,was moved back and forth across it by hand. Guides were placed in front and behind the crystal to keep the thread in the correct cutting position. The thread was kept wet by passing it through a dish of water as shown in the photograph.

After properly orienting and securing the crystal in place the thread was passed over the pulley,through the water,and across the crystal. Attached to the pulley end of the string was a small weight which hung over the edge of the table. The string was pulled across the crystal,lifting the weight,which always kept it taut. Upon releasing the string,the weight pulled it back across the crystal again,and all was ready for the next cut. One precaution should be taken particularly,and that is to be sure that the water used in cutting is the same temperature as the crystal. This is to prevent cracking due to too rapid temperature changes in the crystal if it is exposed to cold water.

With this apparatus,about five minutes is required to cut a crystal. The usual cut is to halve the crystal along the (b) axis. The remaining portion of the crystalnear the outside is either cut away or ground off.

The crystal is then polished by using powdered carborundum and water on a smooth surface,finished by ground glass and water. This left a thin slab of crystal from the center of the piece,perpendicular to the (a) axis and in the (b) plane,as shown in figure 18. This is the most active plate that canbe cut from the crystal unless the one cut at 45° to this one is considered,as shown in the dotted lines of figure 18.

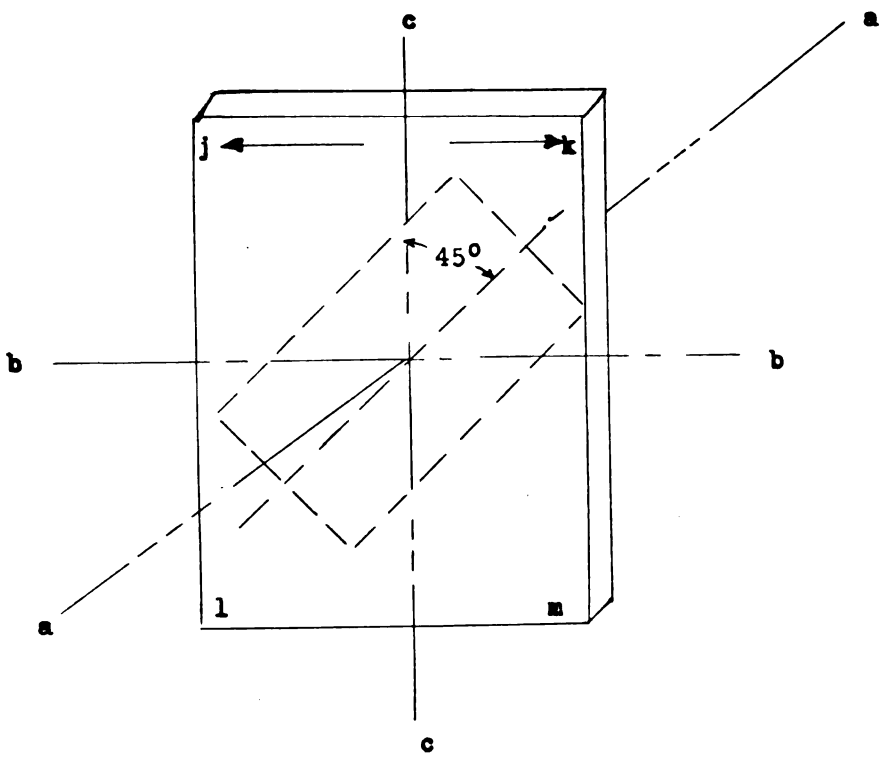


Figure 18

MOUNTING THE CRYSTAL

After the crystal had been sectioned,some form of mounting must be considered. As before mentioned there are two possible arrangements of the crystal for best results. If the crystal is as first cut,shown in the solid lines of figure 18,the expansion,upon application of potential,is in the (b - c) plane parallel to the (b) axis. If the crystal is held firmly at (l - m) the (j - k) portion will move back and forth in synchronism with the applied voltage. Obviously this is one way of mounting the crystal. It may be clamped firmly at one end and the desired movement taken from the other end,as described above. This might be classed as arrangement "A",for simplicity. Another arrangement of almost equal simplicity is that of a section cut from the crystal at 45° to the (b) and (c) axes in the plane of these axes,as shown in the dotted lines of figure 18. Such a plate will be acted upon in compression and extension where the first mentioned plate responds to shear. Let us denote the latter plate as arrangement "B". Obviously either of these methods would produce a piezo electric bar,or plate,which,when energized from either electrical or mechanical sources, would function as the simplest form of electro-acoustical instrument it is possible to conceive.

It is found,however,that in this case,as in many cases,the simplest arrangement is not the best one for perfect results. As has been mentioned in the articles by Mr. Valasek,included in the first section of this paper, Rochelle salt crystals functioning piezo electrically experience fatigue and hysteresis loss,depending to a greater or less extent on several factors

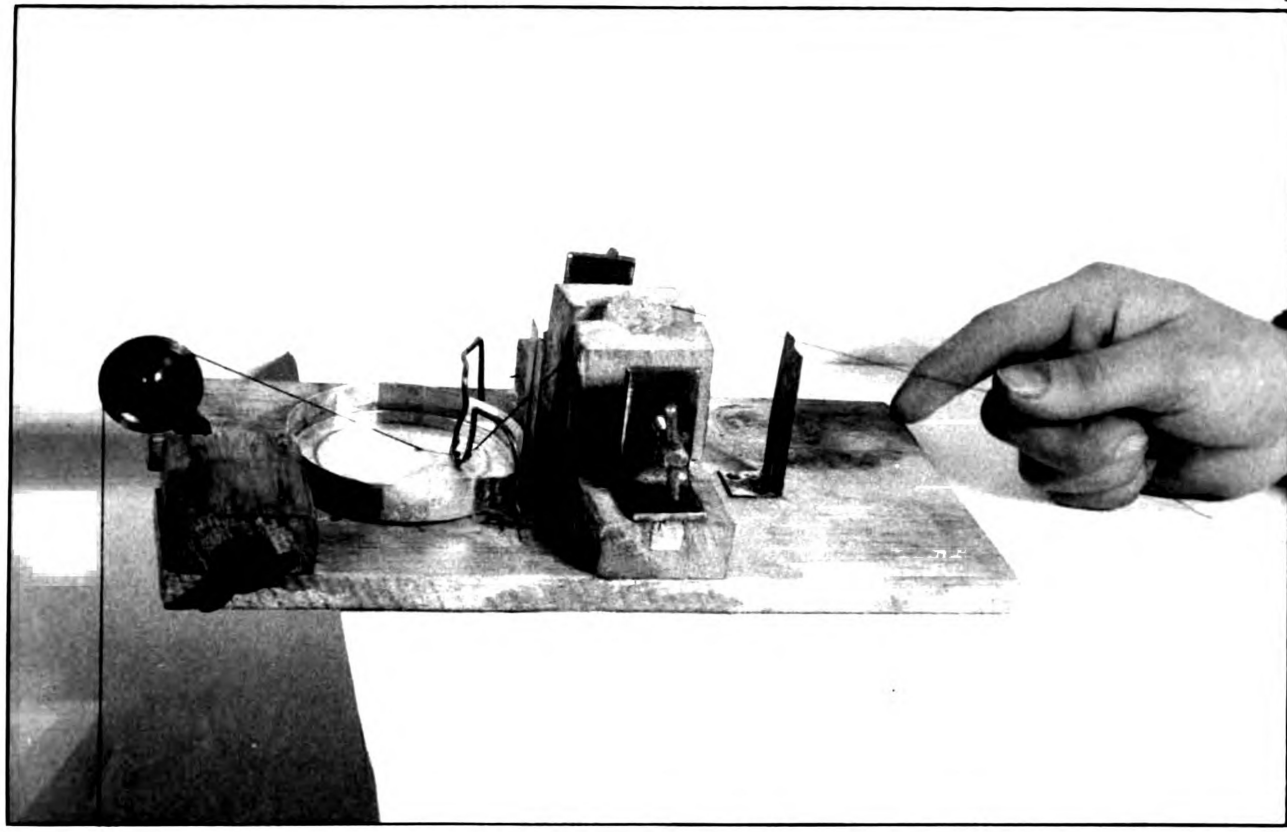


FIGURE 17

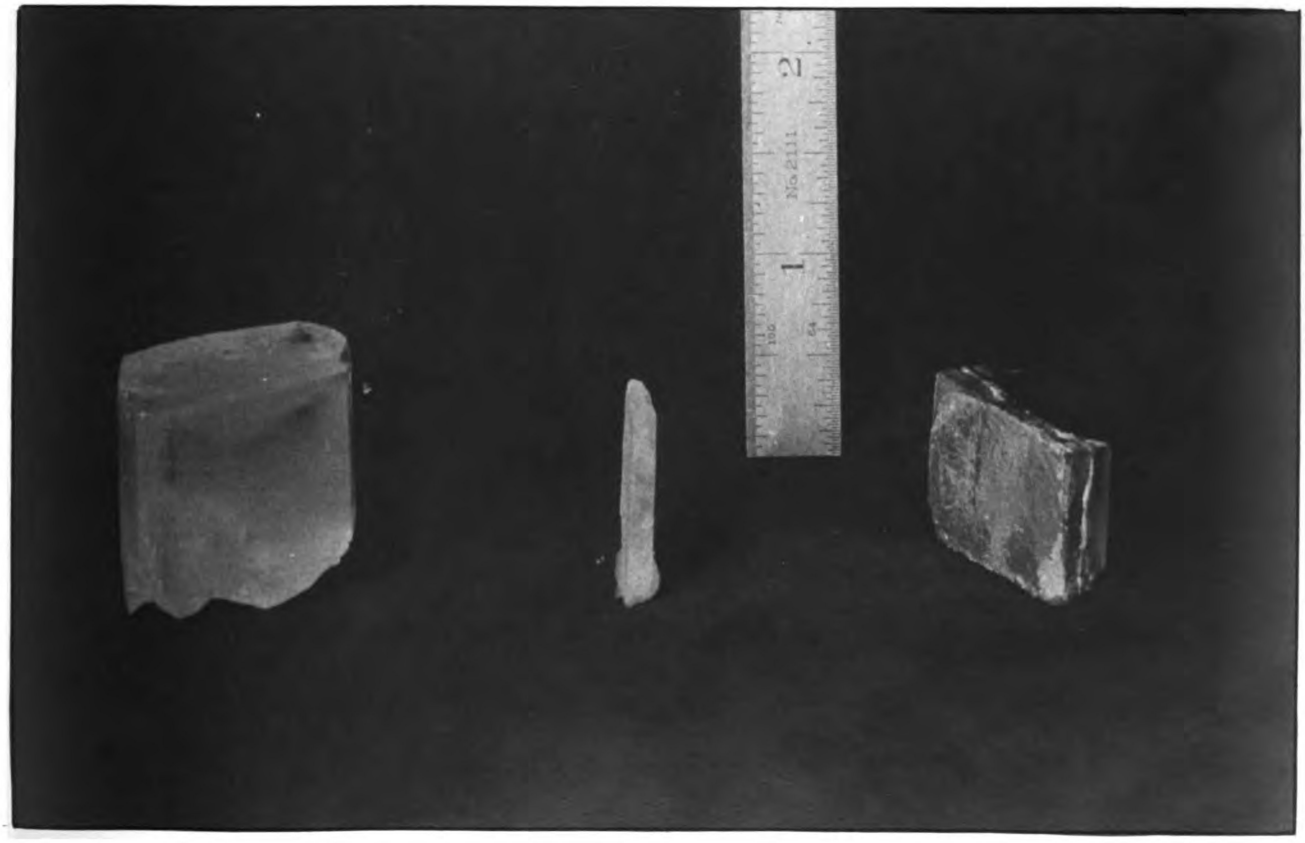


FIGURE 19

already discussed. To overcome this difficulty several remedies have been tried. It was found that if the entire plate was clamped and prevented from moving,to any great extent,upon application of potential,the hysteresis loss and saturation of the crystal disappeared. It would have been impossible to use a crystal so clamped,but,by cementing another similarly cut crystal to the first,in such a manner that the forces are always opposing each other,a similar result was obtained. At the same time great magnification of movement results,but in a direction at right angles to that experienced before.

Compare,if you will,such an arrangement to that of the bi-metallic thermostat. With two sections cut as in A,and cemented together in opposition,an element results which twists upon application of potential,or creates potential upon being twisted,the action being reciprocal. If cut as in B the movement is one of bending,very similar indeed to the bimetallic strip upon application of a temperature gradient. In either case the movement is proportional to the impressed voltage,or vice versa,and no hysteresis is present. The frequency of the applied force is also immaterial up to at least 500,000 cycles per second.

Owing to the small size of the crystals obtainable,all mountings were made after arrangement A. This gave an element sensitive in torsion, which must be used accordingly. Figure 19 shows the three stages of preparation of the crystals for use in these experiments. The steel scale, calibrated in inches,gives an idea of the size of the crystals.

THE PHONOGRAPH PICK-UP

The actual making up of one of the crystals into a usable unit took the form of a phonograph pick-up, for playing ordinary phonograph records through the radio or other electrical amplifiers. It was made from a piece of extremely clear crystal, which had unfortunately broken in two after having been used for some time in experiments with loudspeakers. The two pieces were cleaned and shellaced together again in opposition with a piece of lead foil between them. This center foil served as one terminal of the instrument. The other terminal was the two outer surfaces which had been foiled in a similar manner. To one corner of the crystal assembly was cemented a grooved piece of pressed fiber, which had been drilled and tapped to accomodate the phonograph needle and holding screw. The unit was then mounted in a flat circular head which was also made from a piece of pressed fiber. The needle box, as the receptacle for holding the needle is known, projected from the case through a hole in the side. The cover, which was machined to fit tightly into the head, was of the proper thichness to hold the crystal unit snugly in place and prevent motion, when once bolted together. This cover also had a cavity of the proper size cut in one side to accomodate the needle box, which, of course, must be free from all solid contact with the head, except through the crystal itself. Included in the head was a small .001 uf. condenser connected in parallel with the crystal unit to bipass the scratch of the needle on the record. Smaller details of the arrangement can be gathered from the photograph, figure 20, which is a close-up view of the pick-up head.

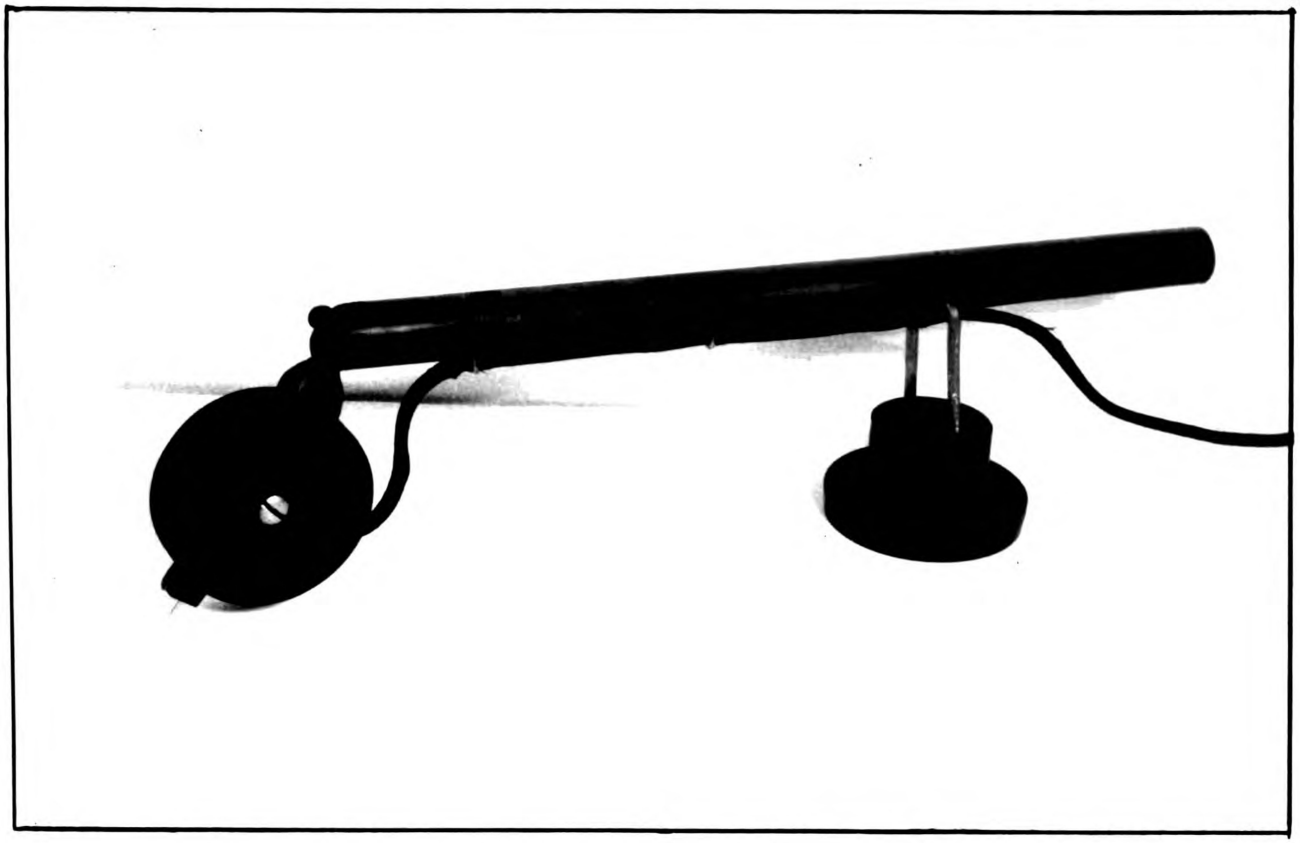


FIGURE 20

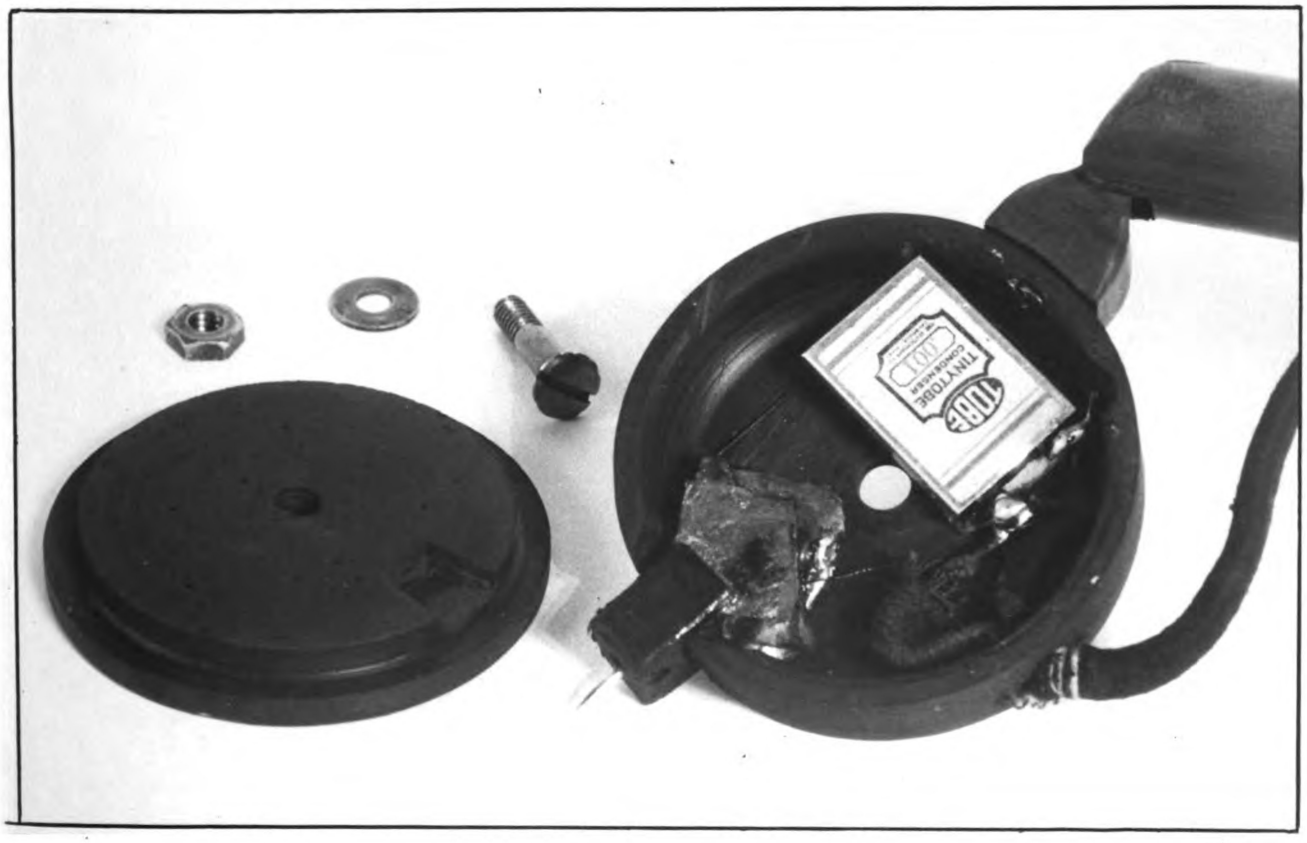


FIGURE 21

The complete unit was mounted on a pivoted arm which was free to rotate on its base,as shown in figure 21. The head was so constructed that it could be used to play either the conventional lateral cut record or the new type vertical cut recordings. All that is necessary,to change from one type to the other,is to loosen the retaining screw in the end of the arm, remove the head,turn it a quarter turn and reinsert it in the arm. The vertical position of the head,as shown in figure 21,is for playing lateral cut records. The horizontal mounting plays the vertical type. This seemingly reversed nomenclature results from the necessity of picking up the vibrations from the record in such a manner as to cause the proper torsion in the crystal unit,thus producing the piezo electric potential for the amplifier excitation.

In comparison with the magnetic type pick-up we find the crystal unit a very capable competitor. It is inherently simpler and under proper manufacturing facilities probably less expensive. No method of damping,as commonly employed,is necessary, since the resonant frequency of the crystal is well outside the audible range. Its response to high frequencies is better than the magnetic unit and here,perhaps,trouble is encountered,due to the high frequency scratch from the record,unless excellent records and proper needles are used. Impedance match must of course be made for best results. The impedance of the unit is such that it may be inserted directly in the grid circuit of the first amplifier either with or without a grid leak across it. Transformer coupling may be used but trnasformers tend to suppress the low frequencies and should be avoided whenever possible. The unit itself has the characteristic of a leaky condenser of about .01 uf. capacity the leakage ,however,being small.

The actual frequency response of the unit could not be determined due to lack of calibrated sound instruments in the laboratory. However, the response,as determined by the ear,the final criterian in any case, was entirely satisfactory to all.

THE CRYSTAL LOUDSPEAKER

Many experiments were preformed in an endeavor to produce a loudspeaker of suitable volume and fidelity. It must be admitted here that such aspirations were not entirely fulfilled. Due to the difficulty in producing crystals larger than about one and three quarter inches long by an inch or so thick, it was impossible to get sufficient motion to successfully drive the eight inch cone. The faithfulness of reproduction was good at low sound levels, but, in attempting to increase the volume, all quality was lost. Since the crystal itself could not be overloaded without great loss of fidelity, a mechanical lever was tried in an attempt to increase the movement. Any mechanical arrangement is inefficient, and as much was lost here as was gained from the lever ratio. That such a speaker can be constructed which will favorbly compare with the best dynamic units can be shown from the success of the Brush Development Company of Cleveland Ohio, who have produced such units for the past year or two. They used a crystal which measured two and three quarter inches by three and one half inches, so it is little wonder that the results obtained from the comparitively tiny one used in this experiment were so unsatisfactory.

THE CRYSTAL MICROPHONE

Since the crystals responded very well at low sound levels it was decided to try to construct a microphone from some of the crystals which had proved too small for a speaker. The crystal unit itself was made from the small crystal shown in figure 19. The unit was assembled for sensitivity in torque with terminals brought out as in the pick-up. It was mounted between two fiber pads,as shown in the photograph of figure 22,and securely held to the heavy pressed fiber base by two small bolts. The diaphram for the unit was made from a light manilla paper cover of the familiar laboratory report,in the absense of any better material. It was securely clamped between two rings made from the same fiber boerd as the base. The bottom ring served as a spacer,for the support of the diaphram at the proper level above the crystal,and provided ample space for mounting. A small threaded rod was cemented to one corner of the crystal at the center of the base. This served as a means of coupling the crystal to the diaphram,which was attached to the rod by two small nuts.

The ring was attached to the base by three bolts,which fitted into tapped holes in the ring,but did not project through it. Thus a neat unit was assembled. The view of the assembled microphone is shown in figure 23.

The microphone,while less sensitive than the conventional double button carbon type,is much more sensitive than the condenser microphone. It will furnish good volume with an amplifier consisting of a type '27 driver and two '47's in push pull,although another stage would reduce the noise level considerably. The quality,as determined by the only avail-

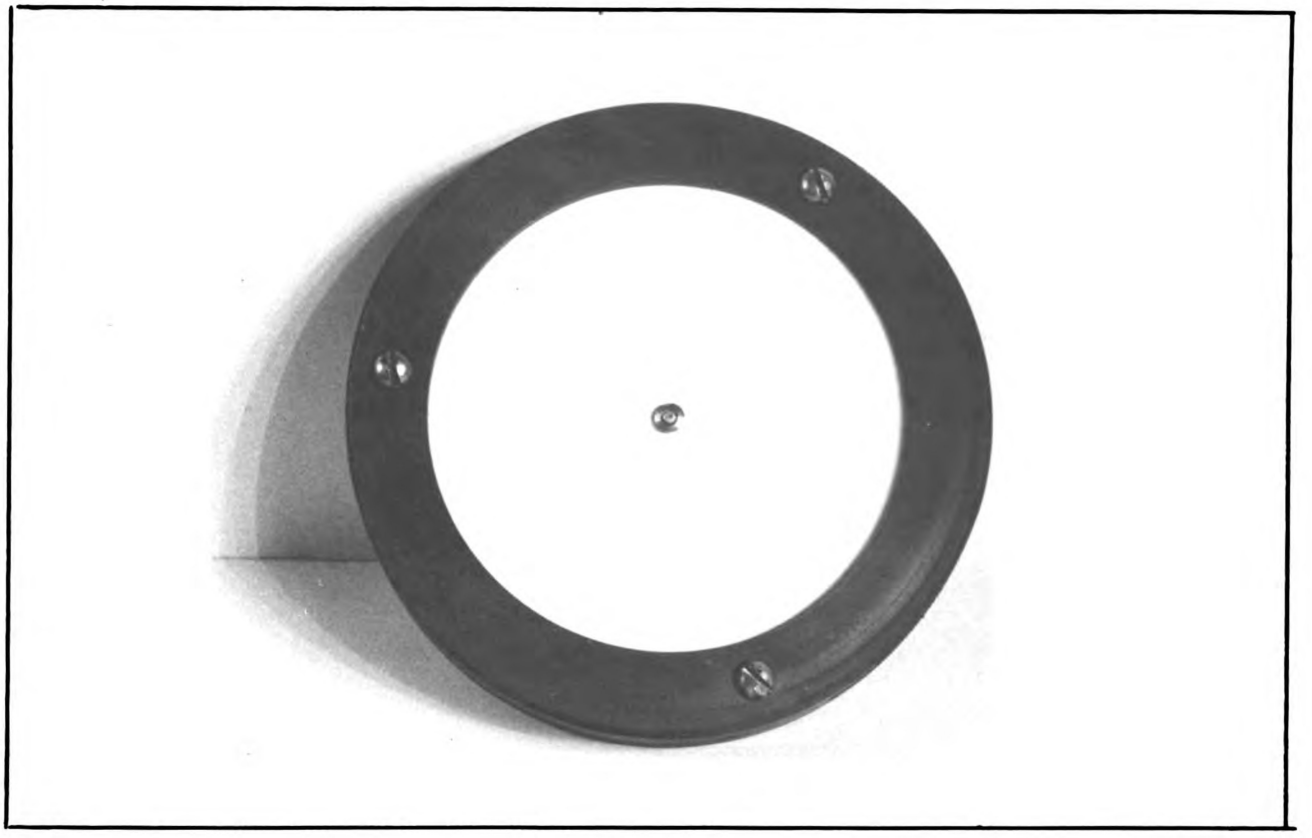


FIGURE 22

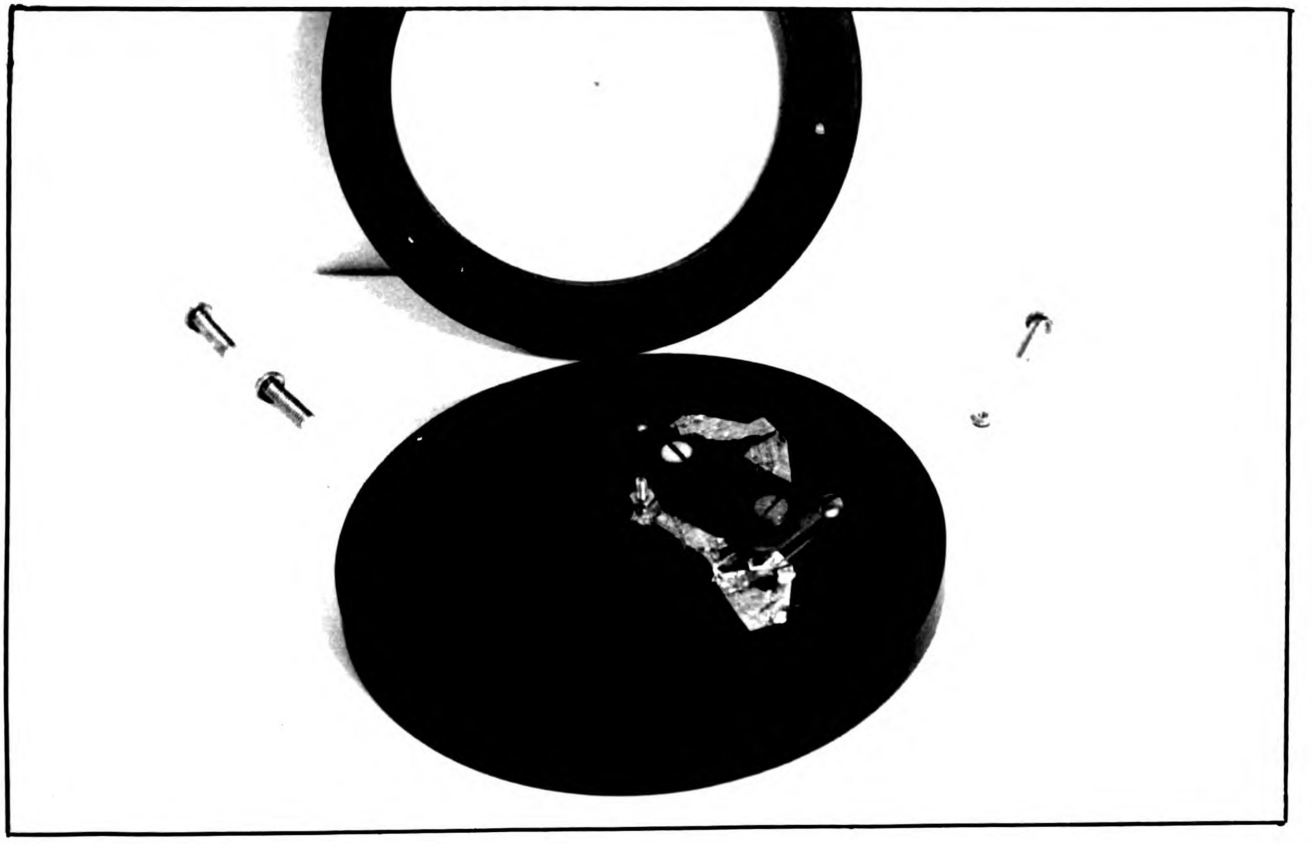


FIGURE 23

able means,the ear,was excellent. Absolute calibration,by the Brush Development Company,of one of their microphones,showed a straight line from 50 to 5000 cycles with a gradual drop below 50 and a similar rise above 5000. This was accomplished by addition of parallel resistance. However,the crystal unit alone varied less than 8 decibels over the entire range. Almost any impedance characteristic desired can be obtained by either parallel or series arrangement of several of the crystal units. In this case,of course, the units themselves act as the diaphram,with a corresponding decrease in sensitivity and increase in fidelity.

X - RAY AND CRYSTAL STRUCTURE

During the course of these experiments the author had occasion to become interested in some work which was being done in the X-ray field at the Physics Department,by Professor Snow. Having acquired some new equipment,a special room was being set aside for X-ray work alone,something previously impossible. In trying to work out a course in this field, Professor Snow invited several of the advanced students to participate in some experiments he proposed to preform. Being desirous of keeping up with the times as much as possible in all lines of endeavor the author at once availed himself of the opportunity. In the course of these experiments the investigation of crystal structure by X-ray analysis was mentioned and discussed. Having at hand a General Electric X-ray Spectrographic Outfit it was decided to actually preform the experiment and thereby get a little information 'first hand' ;so to speak.

It immediately occured to,the author to try some of the Rochelle salt crystals in an attempt to find out more about their actual construction and,possibly,the reason for their piezo electric action. To this end many trials were made. Between poor technique of handling the apparatus and films,and some bad accidents to these films,the first few trials were not very successful. However,by keeping at it,and gradually improving the methods of procedure,some very good Laue patterns were obtained. Three of these are shown in the accompanying photographs. They are respectively: Figure 24,with the crystal perpendicular to the (a) axis;figure 25,with the crystal perpendicular to the (b) axis;and figure 26,with the crystal

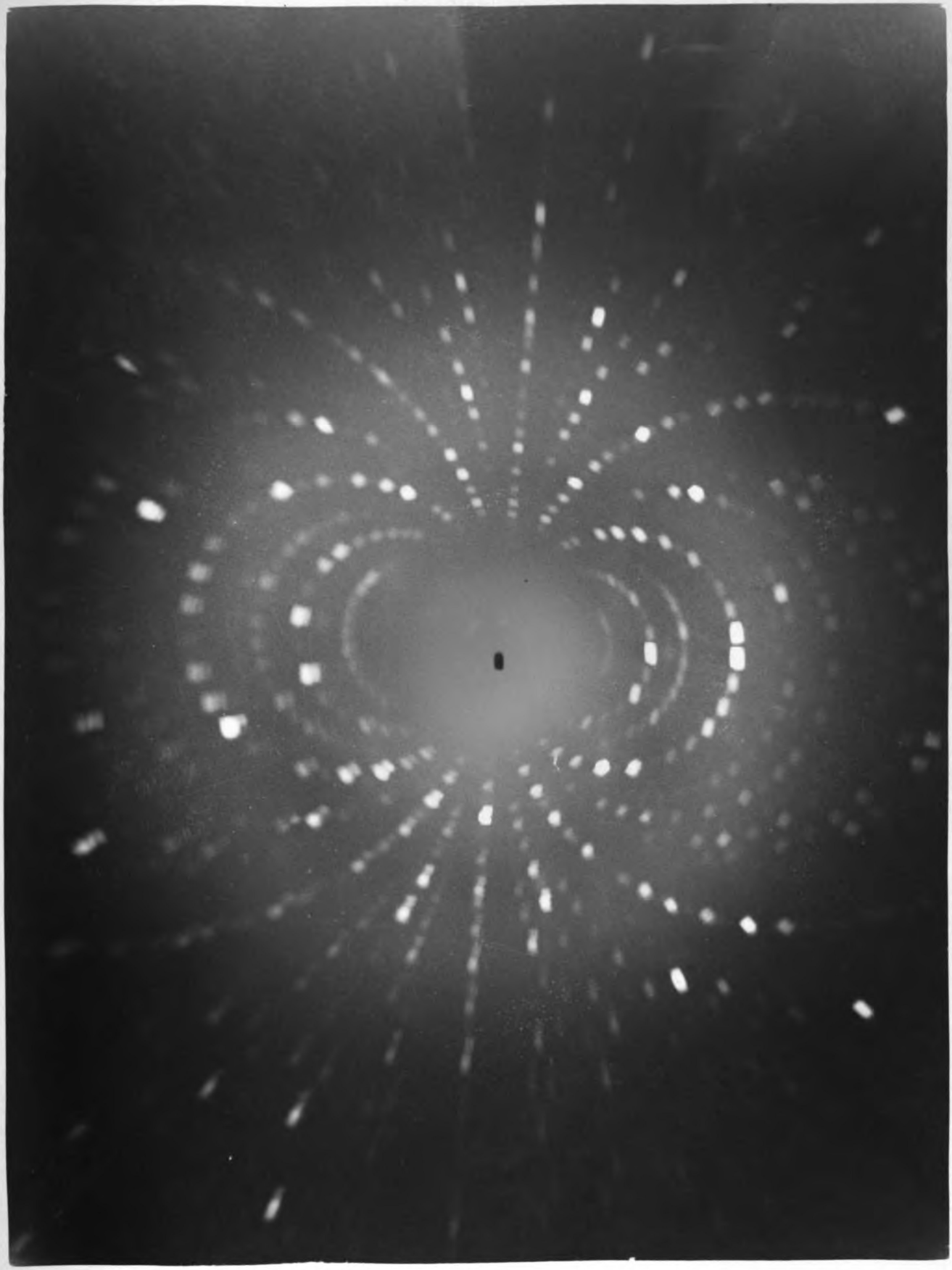


FIGURE 24

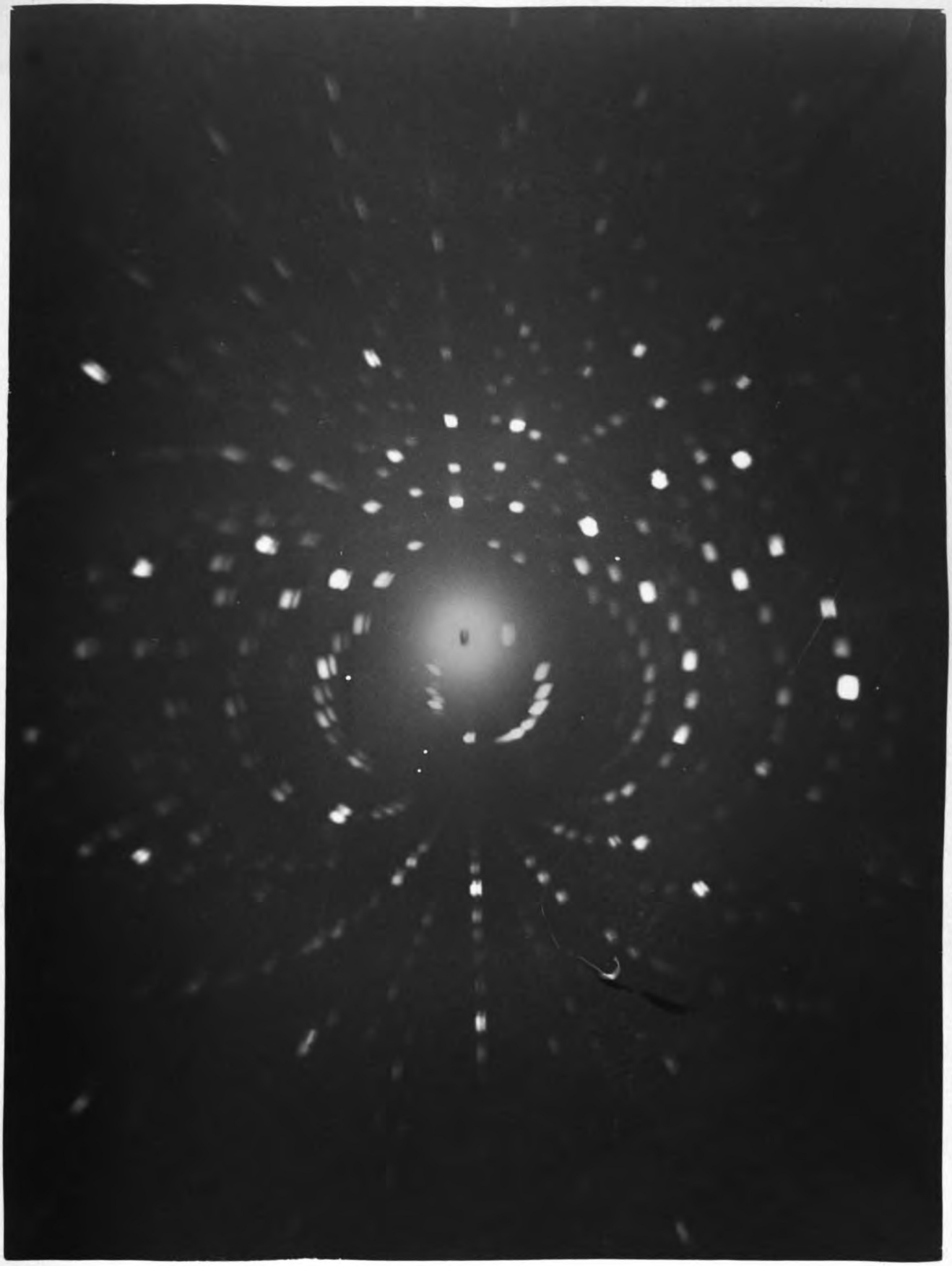


FIGURE 25

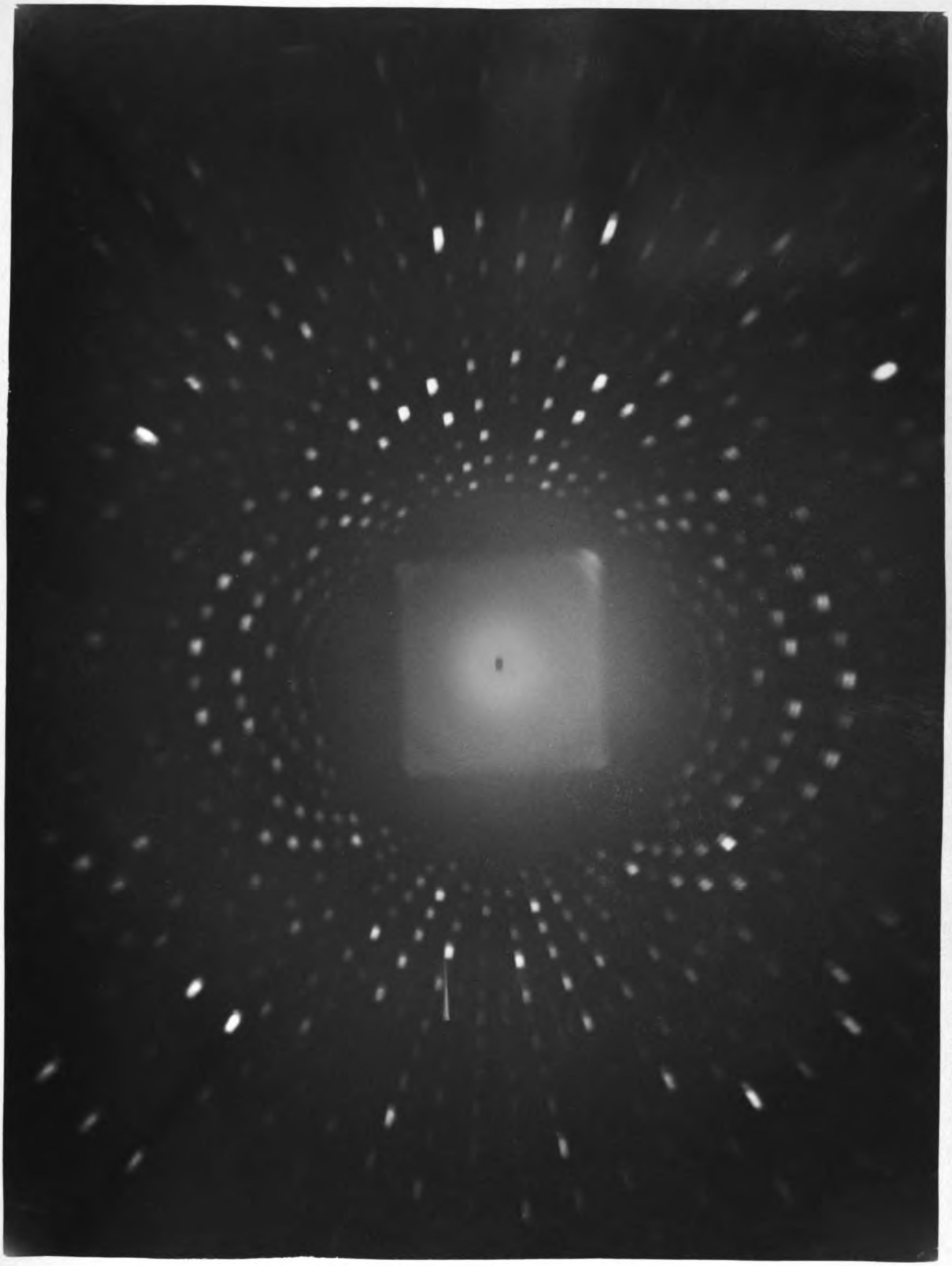


FIGURE 26

perpendicular to the (c) axis. The similarity of figures 24 and 25 indicate similarity of strusture in these planes,while the totally different figure 26 shows an extremely different arrangement of atoms here. Time did not permit the actual working out of the crystal structure,even if such were possible from these photographs,since such a problem is a thesis by itself. The lack of symmetry in figures 24 and 25 is due to inaccuracies in grinding the crystals and in mounting them in the machine. Each succeeding trial yielded much information as to better methods of procedure in taking the photographs,but this was costly to those taken in that trial. One example of this is the extremely light spot in the center of the plate due to the undeflected beam of X-rays. It was found later that this could be partially, if not,wholly,eliminated by placing a small piece of lead in front of the photographic plate at this point. This,of course,would result in a much clearer plate near the center by the elimination of scattering due to the emulsion on the plate.

The actual information yielded by these Laue patterns is not great. It is,however,interesting to note that X-ray furnishes a means of actually looking into the atomic structure of things. The use of these patterns,in conjunction with those made from mono-chromatic radiations in spectrographic work,provide very excellect methods of obtaining this priceless information about the internal arrangement of materials. Had time and facilities permitted,nothing could have pleased the author more than to have investigated the crystals more thoroughly by this means. As it is,the photographs serve only as a very crude example of what can be done with X-rays along the line of crystal structure,and to this end add a little color to the experiments with Rochelle salt crystals and their piezo electric properties.

CONCLUSION

In conclusion let us summarize the advantages posessed by Rochelle salt electro-acoustical apparatus over the older magnetic types,and the existing electro-static types,as well as cite some possibilities in application to other fields.

First,the use of Rochelle salt devices offers outstanding cheapness and simplicity coupled with a flexibility of design very difficult to find anywhere else. The life is extremely long,and failure from fatigue is unknown,under ordinary working conditions,with the sectionalized unit as herein described. Such apparatus as has been discussed,and perhaps many other applications,exhibit real individualism in that they,unlike all magnetic types,require no permanent magnets or heavy currents for field excitation. Unlike the carbon microphone,no batteries are required for power,and no polarizing bias is necessary as with electrostatic instruments. In all these cases Rochelle salt is indeed unique in that it carries its own field excitation.

When connected to an input circuit there is ample potential generated to swing the grid of almost any suitable tube sufficiently for complete utilization of that tube. In output circuits the impedance match is direct with most power tubes on the market today. For multi-unit installations the low impedance necessary can readily be obtained by the proper arrangement of units. Also,the force present in output units is indeed great,and can be used to good advantage in driving the heavy cones necessary for good reproduction at high sound levels.

The application of the piezo electric crystal in modern science is indeed just beginning. Besides the speaker,microphone,and pick-up these crystals have been successfully used in polarized relays,where potential is present but where very little power is available. They also find applications in oscillographs and oscilloscopes. Another very tempting application is in the matter of seismographs,for continuous automatic recording of earth tremors,such as might be caused by an earthquake several thousand miles away. It is apparent that such movements of the earth as would result from such a cause would indeed be small. But,small as they are,a device as sensitive as Rochelle salt crystals should pick them up with very little difficulty. That they were used successfully during the Great War to detect submarines,by means of minute water vibrations,is proof of their sensitivity. And in the light of modern developments this sensitivity has been increased many times.

There are undoubtedly scores of other applications,equally interesting,which are eagerly awaiting recognition and development. May they not be long in coming,and they will not if some of the gifted engineers,like those at the Brush labortories,get to work on the problem. The savings which are effected by such applications,both in labor and capital,should be a great incentive for the immediate development of these devices.

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Note: Parenthesis,(-),indicate classification in the Dewey Decimal system used in the Michigan State College Library.