The Transition from Glow Discharge to Arc

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(Received November 10, 1938)

INTRODUCTION

THE fundamental difference between glow and arc discharges lies in the mechanism of electron supply. The glow discharge requires a high cathode fall, because electrons must be liberated from the cathode by the comparatively inefficient agencies of ionic bombardment, photoelectric effect, or the impact of excited atoms generated in the gas. In the arc discharge, the cathode emits electrons by more efficient processes and the cathode fall is correspondingly lower.

If the cathode is made of a highly refractory material, the electron emission can be thermionic. Wehrli[1] has observed the gradual decline in the glow discharge cathode fall which accompanies the increase in temperature of cathodes of this kind. Arcs also occur, however, with cathodes of materials which, because of thermal disintegration, could never reach the high temperatures necessary for effective thermionic emission. Furthermore, glow discharges have been observed to change into arcs even when the cathodes were efficiently cooled, leading to the suggestion that the electrons are liberated from the cathode in this case by high electric fields. This hypothesis has, however, never been completely proved.

Transitions from glow to arc discharge can be produced either by an increase in current under constant gas pressure, or by an increase in pressure at constant current. The highest pressure under which glow discharges have been observed heretofore is of the order of one atmosphere.[2] This paper describes some further experiments on glow discharges in air, oxygen and nitrogen at one atmosphere pressure, and in hydrogen under pressures ranging from 1 to 13 atmospheres, using various combinations of electrode materials. These experiments have shown that glow discharges between copper electrodes may exist in hydrogen with currents as large as 14 amperes and, though not simultaneously, under pressures as high as 13 atmospheres; that with the other gases and electrode combinations tested the glow was less stable than in hydrogen; that continuous stability of the glow over long periods may not be expected even with small currents. Additional evidence was found for the hypothesis of electron emission in arcs under high surface field strengths, and characteristic changes of the electrodes in glow discharges have been observed due to sputtering and chemical reactions.

APPARATUS

The electrodes used consisted of disks approximately 12 mm in diameter and from 0.3 to 3.0 mm thick fixed to the ends of brass tubes. Very effective cooling was obtained by directing a jet of cold water against the center of the back of each electrode disk. The cooling was sufficiently effective that a tenfold increase over normal in the flow of water produced no change in the behavior of the discharge. The current for the discharge was supplied in most cases by two 2000-volt, one-ampere, d.c. generators which could be connected in series or parallel. For the largest currents one 550-volt and two 230-volt generators of large capacity were connected in series. In all cases the discharge was initiated by a high frequency, high voltage impulse, which was kept out of the generators by a suitable filter.

In the experiments at atmospheric pressure a chamber of about $3\frac{1}{2}$ liters volume housed the electrodes. The gas was caused to stream through the chamber at a rate sufficient to carry off products of the discharge quickly. The hydrogen used at atmospheric pressure was prepared electrolytically, great care being taken to keep it

* The material included in this paper is taken from a thesis submitted to the Electrical Engineering Department of the Massachusetts Institute of Technology in May 1937 in partial fulfillment of the requirements for the degree of Doctor of Science. Publication has been delayed by the author's return to China, where he is now connected with Tsing Hua University.

[1] M. Wehrli, Helv. Phys. Acta **1**, 323 (1928).

[2] H. Thoma and L. Heer, Zeits. f. tech. Physik **13**, 464 (1932); **14**, 385 (1933); **15**, 186 (1934); R. Seeliger and K. Bock, Physik. Zeits. **34**, 767 (1933); O. Becken and R. Seeliger, Ann. d. Physik **24**, 609 (1935).

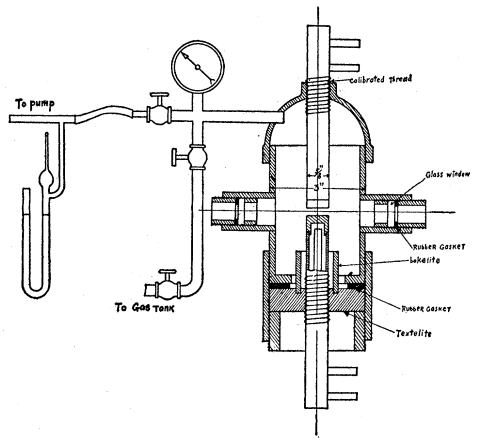
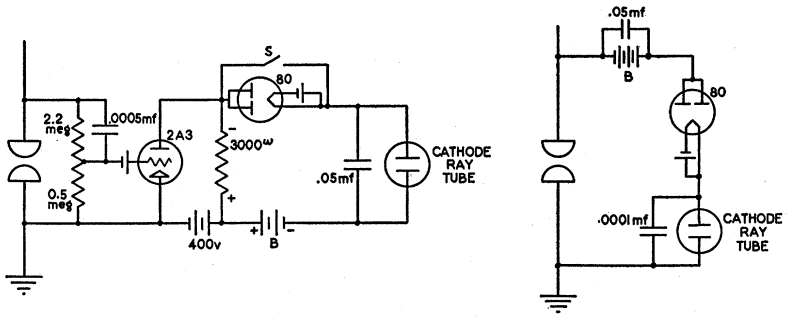


FIG. 1. Section of chamber used for studying discharges at pressures above atmospheric.

as pure as possible. The other gases were supplied from tanks, and the oxygen used was of ordinary commercial purity. The nitrogen and hydrogen were obtained especially, and had about one part in a thousand of impurity. The pressure chamber shown in Fig. 1 was used for the measurements with hydrogen above atmospheric pressure.

There are regions of current and pressure where the glow type of discharge is comparatively steady, and other different regions where the arc is steady. The experiments described in this paper pertain to the intermediate region where transitions from glow to arc take place. When the current is sufficiently large, temporary transitions from glow to arc can be detected. These produce a momentary increase in brightness of the discharge, particularly at the cathode, and are accompanied by a momentary drop in discharge voltage. When the discharge current is small the transition period is usually too brief to be observed by simple methods, and a fast detecting circuit must be used. The circuits of Fig. 2 were devised for this purpose, that of Fig. 2(a) having been developed first and serving for voltage drops of duration down to about 10^{-3} second, and that of Fig. 2(b), a modification of 2(a), serving for drops of duration as small as 10^{-6} second. Both circuits act on the principle that a condenser is charged when the voltage across the discharge falls, the charge being trapped by means of a thermionic rectifier when the voltage recovers. The presence and magnitude of the trapped charge is indicated by means of an electron oscillograph. In the circuit of Fig. 2(a) the deflection of the cathode-ray beam caused by a fall in discharge voltage was comparatively permanent, and it was necessary to return the beam to its original position by means of the switch S. In the circuit of Fig. 2(b) the internal leakage of the cathode-ray tube was sufficient, because of the small charge stored in the small capacitance across the tube, to return the beam in about one second to its equilibrium position. Observations with this circuit were facilitated by the simultaneous use of the horizontal sweep. The circuit of Fig. 2(b) gave a fairly accurate measure of the magnitude of the temporary change in voltage drop across the discharge, but the amplifier of Fig. 2(a) was not sufficiently carefully constructed to give more than qualitative information on the magnitude of the voltage changes. Decreases in glow discharge voltage drops were observed for all currents, and most of these decreases were in excess of 250 volts.



(a) (b)

FIG. 2. Circuits used in the study of voltage disturbances of short duration.

PROCEDURE

The systems investigated are shown in Table I.

Air

In Fig. 3 the dotted curve shows the volt-ampere characteristic of the discharge in air at atmospheric pressure with copper electrodes. The glow becomes increasingly unstable as the current is increased, and the arc becomes increasingly unstable as the current is decreased. The voltage of the glow is constant over a wide range as is the rule for "normal" glow discharges, in which the surface of the cathode is not entirely covered by the negative glow. At very low currents the voltage increases sharply, but the curve could be followed over only a short range because of the limited voltage of the generators, and the need of using a large series stabilizing resistance.

In the lower ranges of current this discharge appears to the eye to be perfectly stable, a fact which has perhaps led some observers to postulate a lower limit of current below which transitions do not occur. It is true that the frequency of the transitions decreases with decreasing current, but with the fast detecting circuits used, transitions were observed down to the limit of current where further decrease became impractical because of the sharp increase in voltage (cf. Fig. 3). Substitution of platinum for copper in the electrodes did not change appreciably the frequency of the disturbances, which indicates

TABLE I.

GAS	CATHODE	ANODE	PRESSURE (ATMOS.)
Air	Cu	Cu, Pt	1
Air	Pt	Pt	1
O_2	Cu, Pt	Cu	1
N_2	Cu, Pt, Al	Cu	1
H_2	Cu, Al	Cu	1
H_2	Cu, Pt, Mo	Cu	1 to 13

that oxidation of the cathode is not the main cause of their occurrence.

In addition to the large voltage drops described above, small voltage drops (10–20 volts), following each other in rapid succession, were observed when current had been passed between the electrodes for a long period of time. At the higher currents these disturbances can be recognized immediately as being associated with the

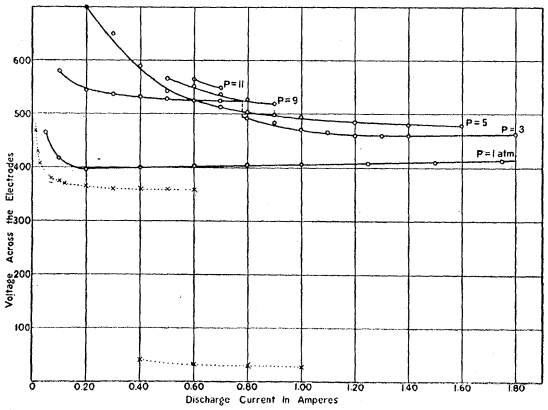


FIG. 3. Volt-ampere characteristics of a discharge in air at atmospheric pressure between water-cooled copper electrodes separated 1.0 mm (dotted curve), and of a glow discharge in hydrogen between water-cooled copper electrodes separated 0.9 mm at several pressures.

unsteady behavior of the anode spot and vaporization of the anode.

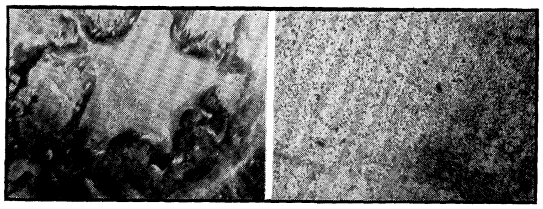
On the anode, the discharge leaves a trace consisting of a clean center surrounded by a black deposit. Once the anodic spot has moved out of the center it falls on the deposit, which begins to evaporate, thus causing further displacements of the spot and the voltage unsteadiness described above. The trace on the cathode shows a smooth spot corresponding in size to the negative glow. This spot, which is perfectly smooth even when enlarged 750 times, is surrounded by a deposit of fine black powder probably removed from the cathode by sputtering. Discharge traces on platinum electrodes do not differ essentially from those on copper electrodes.

Oxygen

Voltage disturbances are more frequent in oxygen than in air. The discharge traces on the electrodes have the same appearance, but the center of the cathode spot on platinum is rougher than in air.

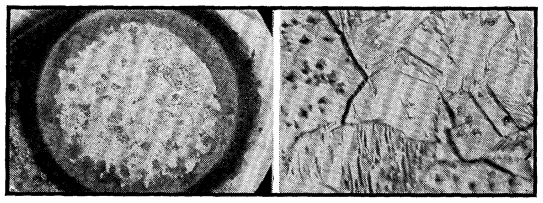
Nitrogen

Voltage disturbances are less frequent and less violent in nitrogen than in oxygen or air, and a comparatively stable glow can be maintained at much higher currents, sometimes up to two amperes. However, the discharge is restless, and to keep the anodic spot stationary the anode must be cone-shaped. Even when the anodic spot is



(a) (b)

FIG. 4. Photomicrographs of surface of copper cathode after glow discharge in nitrogen at 1 atmosphere. (a) magnification of 50 diameters showing irregular shape. (b) center of spot magnified 750 diameters.



(a) (b)

FIG. 5. Photomicrographs of surface of copper cathode after glow discharge in hydrogen at 5 atmospheres pressure. (a) magnification of 50 diameters. (b) center of spot magnified 750 diameters.

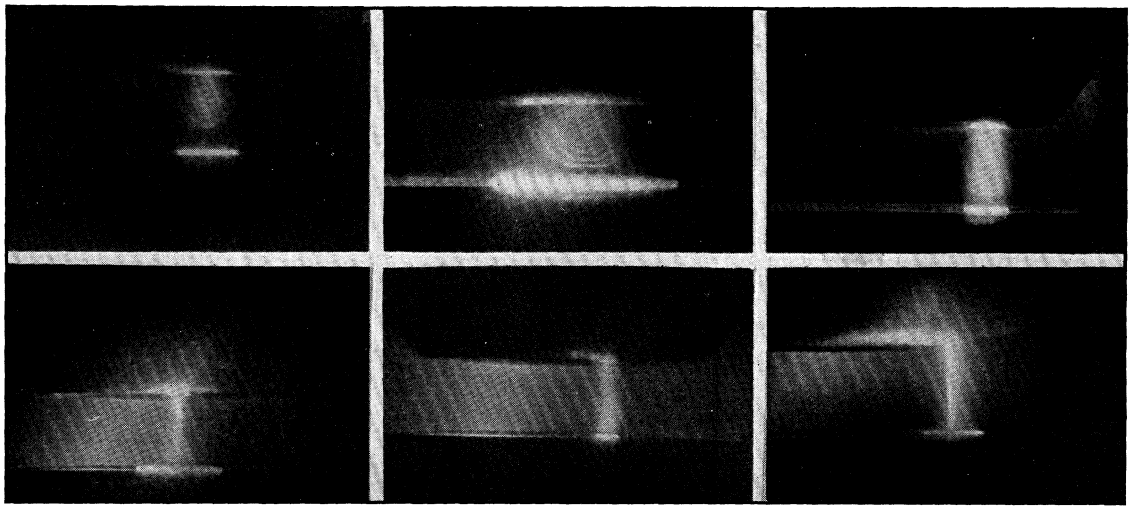
fixed on the top of the cone the irregularly shaped cathode glow refuses to stay still. The anode is not appreciably affected by the discharge, but Fig. 4(a) shows the irregular shape of the spot produced on the copper cathode and the destruction of the surface polish at its center. With aluminum cathodes (three mm thick) the discharge was much less stable than with copper cathodes. Disturbances were more frequent, and the discharge often broke down to a continuous arc running rapidly around the electrode. This arc quickly roughened and destroyed the polished cathode surface until it was no longer capable of supporting a glow discharge.

Hydrogen

The glow between copper electrodes is very stable in hydrogen, with no disturbances occurring at two amperes for several hours. In two tests at five amperes the glow broke down to a continuous arc after 40 and 25 minutes. In a test at ten amperes, the breakdown occurred after 15 minutes. To reach these large values without breakdown, the current must be increased slowly, and by this procedure the current was raised in one case to 14 amperes before the glow changed to an arc.

With cathodes of Pt (0.3 mm), Mo (0.375 mm) and Al (3 mm) disturbances were more frequent in hydrogen than with copper cathodes. The combination Pt-H_2 gave, however, less frequent disturbances than that Pt-N_2 combination. Re-

(a) (b) (c)



(d) (e) (f)

FIG. 6. Appearance of the discharge in hydrogen with copper electrodes separated 3 mm and at a variety of pressures and currents. (a) 1 atmosphere and 0.25 ampere, (b) 1 atmosphere and 2.0 amperes, (c) 2 atmospheres and 0.25 ampere, (d) 2 atmospheres and 1.0 ampere, (e) 4 atmospheres and 0.25 ampere, (f) 4 atmospheres and 1.0 ampere.

sults with molybdenum were similar to those with platinum; aluminum showed the same behavior in hydrogen as it did in nitrogen. The anodes were made of copper in all these experiments and they were not appreciably affected by the discharge.

The stability of the glow discharge in hydrogen with copper electrodes permitted increasing the pressure above one atmosphere, and the glow characteristics under different pressures are shown in Fig. 3. The region of comparatively

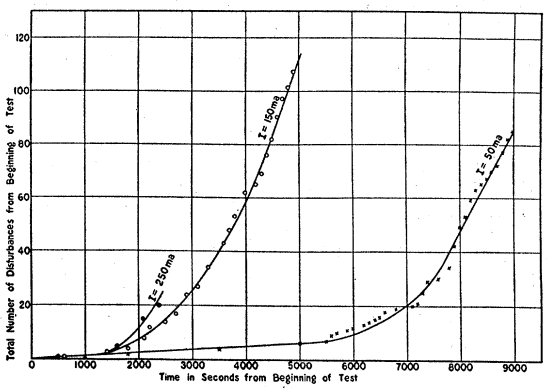


FIG. 7. Total number of disturbances from the beginning of a typical test (copper electrodes in air at atmospheric pressure) as a function of elapsed time from beginning of test.

stable glow becomes narrower with increasing pressure. The lower limit of this region, where the voltage begins to increase at low currents, shifts to higher currents, and at the same time the upper limit is shifted down by an increase in the frequency of disturbances. By using a very high voltage and a very large series resistance the glow can be stabilized with somewhat lower currents, but the limit is soon reached where it degenerates into a Townsend discharge.

Figure 5 shows the effects on the copper cathode of the discharge in hydrogen at a pressure of five atmospheres. The circular spot shown in the photograph of low magnification is surrounded by a circle of reddish deposit. The larger magnification shows how the crystal structure is exposed through sputtering.

Figure 6 shows the appearance of the glow in hydrogen at various pressures and currents. At the lower values of current and pressure the positive column shows well-defined striations. With increasing values the striations become diffused and eventually (Fig. 6(f)) the column concentrates abruptly into a narrow line, with a simultaneous drop in the voltage of the discharge of about 70 volts. It seems likely that this change results from temperature ionization in the column.

If a few percent of oxygen are added to hydrogen some additional striations are observed and the measuring circuit indicates frequent drops of voltage. The oxygen is soon cleaned up, as indicated by the disappearance of the additional striations, and the disturbances gradually disappear.

TIME EFFECTS

With any combination of gases and freshly polished electrodes the time interval before the first disturbance is usually longer than subsequent intervals between disturbances, and the frequency of disturbances gradually rises until it reaches a more or less constant value. The curves of Fig. 7 illustrate this behavior, though these curves must be considered as only qualitative, since the data on which they are based vary widely from trial to trial under apparently identical conditions. In these curves the total number of disturbances taking place from the beginning of the test up to any particular time is plotted as ordinate, with the time in question as abscissa. Consequently the slope of the curve in any region gives the frequency of occurrence of transitions. This is seen to reach a fairly constant value, with the time required to reach constancy less for the larger currents.

To determine whether the interval between the initiation of the discharge and the first transition was entirely random in length, a series of measurements of this interval was made, the electrode surfaces being carefully polished after each breakdown. The result of a typical series of one hundred tests, made in this case with copper electrodes in air at atmospheric pressure and a current of 0.31 ampere, is shown in Fig. 8. The block curve of this figure, showing the distribution in length of intervals between initiation of the discharge and the first disturbance, indicates that the length of this interval is not random, but has a most probable value, a sort of "mean formation time" during which the

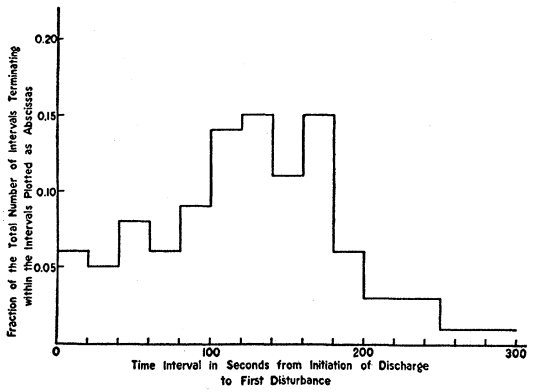


FIG. 8. Distribution in length of intervals between initiation of the discharge and first disturbance, using copper electrodes in air at atmospheric pressure, and a current of 0.31 ampere.

electrode surfaces undergo some kind of progressive change which finally disturbs the stability of the glow.

CURRENT DENSITY AT THE CATHODE

In all of the gases used except nitrogen the discharge formed a well-defined spot on the cathode. From the area of this spot and the total current, a cathode current density can be calculated, and the results of such calculations for discharges in air and in hydrogen are shown in Fig. 9. The current density is constant in air over the range studied, but in hydrogen it increases substantially in the region of small currents.

DISCUSSION

Calculation of the temperature distribution within the materials of the cathode, taking into account the effective cooling of the back surface, shows that the exposed surface could not reach the temperatures required to give appreciable thermionic emission. Since field emission is then the most likely source of cathode electrons during the intervals when the glow breaks down to an arc, the field strengths in the discharges studied were calculated in a manner similar to that used by Engel and Steenbeck.[3] The values found lie between 10^5 and 10^6 volts per cm indicating that field emission from small surface irregularities may be expected. Since the field strength at the cathode surface is determined by space charge, which is in turn dependent on current density, no lower limit of current for the occurrence of disturbances should exist. This is in accord with the experimental results. The increase in frequency of the disturbances with increasing current is to be expected, since the increased current spreads the cathode glow over a larger area, thus increasing the probability of the inclusion in this area of microscopic rough spots.

The effect of sputtering of the cathode, which is evident in some of the photomicrographs, is to uncover sharp points and edges at crystal boundaries and thus increase the probability of the occurrence of large surface field strengths.

The increase in the frequency of disturbances with time, and the differences in the behavior of the glow in different gases seem to point to chemical processes taking place at the cathode. These processes may reduce the work function or cause gas or vapor evolution, both effects facilitating field emission. Güntherschulze and Fricke[4]

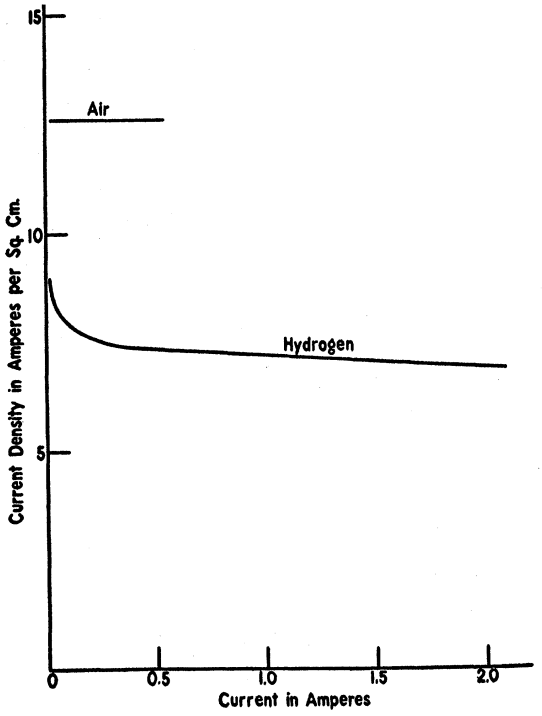


FIG. 9. Current density at the cathode surface as a function of total current in the discharge.

[3] A. von Engel and M. Steenbeck, *Elektrische Gasentladungen II*.

[4] A. Güntherschulze and H. Fricke, Zeits. f. Physik **86**, 451 (1933); H. Fricke, Zeits. f. Physik **92**, 728 (1934).

have observed that glow discharges without cathode-fall are obtained with cathodes made of semi-conductors covered with a thin layer of insulating material. Metal cathodes covered in the same way have a cathode fall but the discharge is accompanied by small sparks on the cathode surface. In a similar way insulating layers formed chemically on the cathode surfaces in the experiments described might give rise to the flashes observed at large currents.

The perfectly smooth surfaces of the cathode spots in air and oxygen and the restlessness of the negative glow in nitrogen are direct evidences of complicated phenomena on the cathode surface. More definite statements about the chemical effects cannot be made, however, until the disturbances have been studied under very pure conditions.

APRIL 15, 1939 PHYSICAL REVIEW VOLUME 55

A Dynamic Measurement of the Elastic, Electric and Piezoelectric Constants of Rochelle Salt

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(Received November 12, 1938)

The elastic, electric and piezoelectric constants of Rochelle salt have been measured at low field strengths by measuring the resonant frequencies and impedance of vibrating crystals. It is shown experimentally that the resonant and antiresonant frequencies of the crystal are both considerably below the natural mechanical resonant frequency of the crystal in disagreement with the usual derivation of the frequencies of a piezoelectric crystal. By assuming that the piezoelectric stress is proportional to the charge density on the electrodes rather than the potential gradient as usually assumed, theoretical frequencies are obtained which agree with those found experimentally. This theoretical derivation together with the measured frequencies supply values for the piezoelectric constants. The elastic constants measured dynamically show some differences from those measured statically. A large difference is found for the dynamically measured piezoelectric constants from those statically measured, which may be attributed to the finite relaxation time for the piezoelectric elements.

I. INTRODUCTION

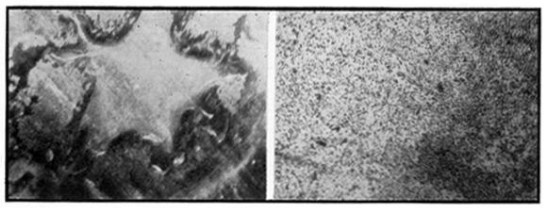
THE static properties of Rochelle salt crystals have formed the subject for a number of measurements. W. Mandell[1] has measured the nine elastic constants of Rochelle salt by a static method, and a number of measurements have been made on the dielectric constants and the piezoelectric constants. However, very few measurements[2] have been made on the dynamic characteristics of Rochelle salt, which may differ considerably from the static characteristics due principally to the large relaxation time of the piezoelectric elements of the crystal. It is the purpose of this paper to present dynamic measurements on the elastic, dielectric and piezoelectric constants of Rochelle salt crystals. These indicate a considerable difference from those determined statically, particularly the piezoelectric and the dielectric constants. Because of the precision obtainable in frequency determination it is felt that the measurements made in this way should be more accurate than those obtained by other methods.

In measuring the elastic constants of a crystal, the frequency of resonance of some known mode of motion is measured electrically and the elastic constant is calculated from this measurement and the known density of the crystal. The usual method of deriving the equations of motion[3]

[1] "The Determination of the Elastic Modulii of the Piezo-Electric Crystal Rochelle Salt by a Statical Method," W. Mandell, Proc. Roy. Soc. London **116**, 623 (1927).

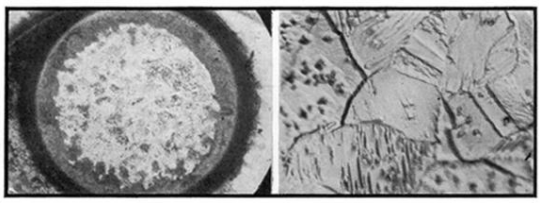
[2] One of the dynamic piezoelectric constants was recently measured by G. Mikhailov (Tech. Phys. U. S. S. R. **4**, 461 (1937)) using a different method. He does not obtain as high a piezoelectric constant as measured by this method.

[3] See for example *Quartz Resonators and Oscillators*, P. Vigoureux (H.M. Stationery Office, London), Chapter III, or *Piezo-electrizitat Des Quarzes*, Adolf Schiebe (Theodor Steinkopff, 1938), p. 85.



(a) (b)

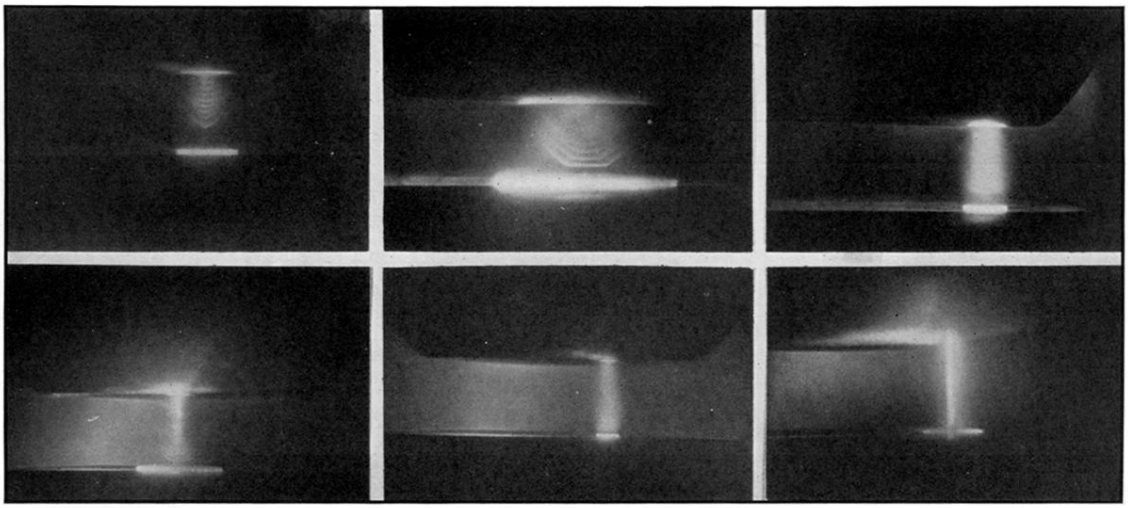
FIG. 4. Photomicrographs of surface of copper cathode after glow discharge in nitrogen at 1 atmosphere. (a) magnification of 50 diameters showing irregular shape. (b) center of spot magnified 750 diameters.



(a) (b)

FIG. 5. Photomicrographs of surface of copper cathode after glow discharge in hydrogen at 5 atmospheres pressure. (a) magnification of 50 diameters. (b) center of spot magnified 750 diameters.

(a) (b) (c)



(d) (e) (f)

FIG. 6. Appearance of the discharge in hydrogen with copper electrodes separated 3 mm and at a variety of pressures and currents. (a) 1 atmosphere and 0.25 ampere, (b) 1 atmosphere and 2.0 amperes, (c) 2 atmospheres and 0.25 ampere, (d) 2 atmospheres and 1.0 ampere, (e) 4 atmospheres and 0.25 ampere, (f) 4 atmospheres and 1.0 ampere.