The Electrical Breakdown Strength of Ionic Crystals as a Function of Temperature*

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It has been generally believed that the breakdown strength of an insulator is *constant* in the lower temperature range and decreases rapidly at high temperatures, indicating thermal breakdown. The authors show that, at least in ionic crystals like KBr, a very steep *increase* of breakdown strength with temperature is recorded in the lower range. This new phenomenon in insulators is apparently analogous to the temperature coefficient of the resistance in metals. It allows us to decide between the different theories of breakdown proposed.

SINCE Wagner,[1] Hayden and Steinmetz,[2] and Güntherschulze[3] proposed almost simultaneously that the electrical rupture of solid dielectrics is a phenomenon of overheating by current, the existence of a "thermal breakdown" has been accepted as one cause of the destruction of insulating materials. The insulator in this case is treated as a conductor of high resistance having a strongly negative temperature coefficient. Its breakdown voltage has been reached when the heat production by current begins to surpass the heat dissipation. This theory of thermal instability has been developed further by Rogowski,[4] v. Kármán,[5] and Dreyfus.[6] Fock[7] finally gave the complete mathematical solution, and Moon,[8] its formulation convenient for application.

Appropriate cooling or impulse testing will prevent the thermal decomposition of the dielectric; the material breaks down as a result of some other mechanism of electrical destruction. According to Inge, Semenoff and Walther,[9] the two fields of breakdown may be easily distinguished by the shape of the voltage-temperature characteristic (Fig. 1). Below a critical temperature the breakdown strength stays constant ("electrical" breakdown); above this point a steep decrease indicates the thermal nature of the process. Moon and Norcross[10] concluded later from extensive tests evaluated statistically that a third intermediate region of slower decay exists, but the independence of the "electrical" breakdown of temperature has become an axiomatic belief of the literature.

For several years one of us, in studying the properties of dielectrics under high field strength, has become increasingly doubtful of the simple conception presented above.[11] Not only is the numerical agreement between thermal theory and experiment in many cases insufficient, but the visual observation of the development of breakdown in ionic crystals and glass reveals directly that even at high temperatures the

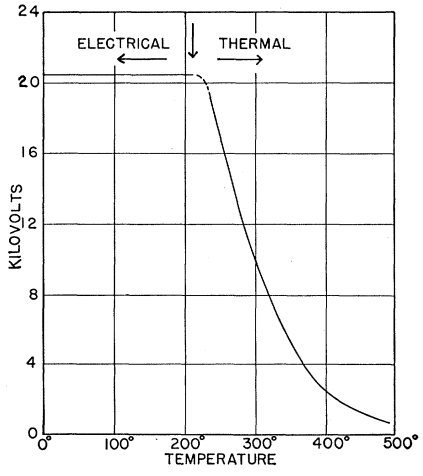

FIG. 1. Temperature dependence of the breakdown voltage through a 1-mm plate of NaCl. (Inge, Semenoff and Walther.)

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[1] K. W. Wagner, J. A.I.E.E. **41**, 1034 (1922).

[2] J. L. R. Hayden and Ch. P. Steinmetz, Elec. World **80**, 865 (1922).

[3] A. Günther-Schulze, *Jahrb. d. Radioakt. u. Elektr.* **19**, 92 (1922).

[4] W. Rogowski, Arch. f. Elektrot. **13**, 153 (1924).

[5] Th. v. Kármán, Arch. f. Elektrot. **13**, 174 (1924).

[6] L. Dreyfus, Schweiz Elektr. Verein Bull. **15**, 321, 577 (1924).

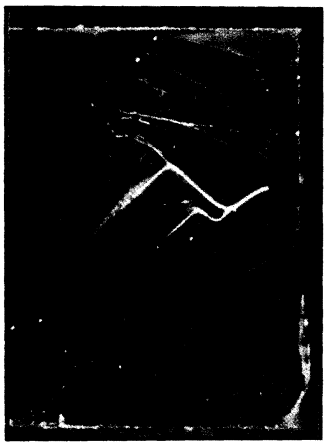
[7] V. Fock, Arch. f. Elektrot. **19**, 71 (1927).

[8] P. H. Moon, Elec. Eng. **50**, 676 (1931).

[9] L. Inge, N. Semenoff and A. Walther, Arch. f. Elektrot. **17**, 433 (1926).

[10] P. H. Moon and A. S. Norcross, J. A.I.E.E. **49**, 125 (1930).

[11] A. v. Hippel, Zeits. f. Physik **98**, 580 (1936).



Cathode← →Anode

FIG. 2. Spark proceeding in NaCl.

material is not destroyed primarily by a melting process.[12] In rocksalt, for instance, a spark path may be observed stepping forward from the anode towards the cathode in the [110] direction (Fig. 2). Furthermore, dendrite formation takes place as well as electron emission from sensitive spots activated by ionic conduction, effects depending on time and temperature and influencing the destruction of the material. Walther and Inge, even in their latest publication,[13] fail to recognize these phenomena published in clear photographs which make the calculation of the breakdown strength by the simple theory of thermal instability impossible. A clear insight into the mechanism of destruction is necessary before a mathematical treatment can be undertaken. A really convincing example of thermal breakdown has been reported recently by Fuoss,[14] in polyvinyl chloride.

Similar reservations have to be made against the claim that the breakdown characteristic (Fig. 1) in the lower temperature region shows independence of temperature. The absolute value of the breakdown strength given for rocksalt, for instance, is much too low (385 kv/cm[13] instead of 1500 kv/cm) indicating difficulties in the measuring technique. Furthermore if electronic impact ionization is responsible for the breakdown as seems probable from the work of one of us,[11, 12, 15] an *increase* in breakdown strength with increasing temperature might be expected in the low temperature field. This effect would be the analog of the temperature coefficient of metals as particularly pointed out recently by Fröhlich:[18] the interaction between conducting electrons and lattice rises if the regularity of the structure becomes damaged by heat vibrations. An effect pointing in the same direction was found by one of us several years ago[16] (Fig. 3): the breakdown strength of a mixed crystal plotted as a function of the composition passes over a maximum like the resistivity of metal alloys.

This parallelism of effects in metals and crystals does not necessarily imply, that in both cases an identical mechanism is acting. In metals the quantum-mechanical scattering of the electron waves belonging to the crystals as a whole is responsible. In insulators the disorder of the lattice produced by foreign matter, stresses or temperature creates deeper potential holes and this may increase the probability of capture.[12, 15]

How large the temperature dependence should be has not yet been calculated by the theory of Seeger and Teller.[17] The physical background of this theory is in accordance with our point of view. Fröhlich,[18] assuming an electronic ionization process without avalanche formation, gives a quantitative formula of the temperature dependence. Frenkel,[19] on the other hand, proposes a *decrease* of breakdown strength with

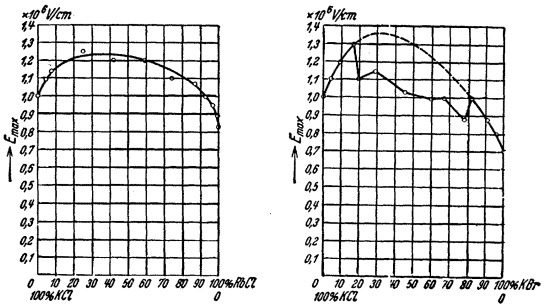


FIG. 3. Breakdown strength as function of the composition. (a) KCl-RbCl; (b) KCl-KBr.

[12] A. v. Hippel, J. App. Phys. **8**, 815 (1937).
[13] A. Walther and L. Inge, Techn. Phys. U.S.S.R. **5**, 335 (1938).
[14] R. M. Fuoss, J. Am. Chem. Soc. **60**, 456 (1938).
[15] A. v. Hippel, Phys. Rev. **54**, 1096 (1938).
[16] A. v. Hippel, Zeits. f. Physik **88**, 358 (1934).
[17] R. J. Seeger and E. Teller, Phys. Rev. **54**, 515 (1938).
[18] H. Fröhlich, Proc. Roy. Soc., London **160**, 230 (1937).
[19] J. Frenkel, Phys. Rev. **54**, 646 (1938); Tech. Phys. U.S.S.R. **5**, 685 (1938).

increasing temperature, assuming that the material is destroyed by thermal ionization, facilitated by the high field strength applied. And Franz,[20] following a theory of Zener[21] that the high voltage transforms the insulator into a conductor by destroying the Bragg reflections of the electron waves, claims nearly independence of temperature for the breakdown strength.

The authors have sought an experimental decision among these contradictory opinions; their methods and first results are reported in this paper.

The Breakdown Apparatus

A main source of error in testing insulating materials is the electrical field at the edge where the electrode bends away from the insulator. If a discharge starts at this boundary before the breakdown voltage of the sample has been reached, the originally homogeneous stress through the sample becomes distorted by surface charges, thus destroying the regularity of the shape of the electrodes. An early breakdown results as a consequence of the inhomogeneous field distribution produced. This edge effect is responsible for most of the erroneous data reported. It can be avoided,[22] by protecting the edge between insulator and electrode by a dielectric guard ring which by perfect contact fills the interspace between both boundaries. Beeswax, for instance, melted under vacuum in the interspace and then solidified and maintained under high pressure may be used in one temperature range. The application of pressure fulfills the second purpose of preventing corona from unprotected parts of the electrodes. Our apparatus was therefore constructed as a high pressure vessel.

Important information about the process of breakdown and the reliability of a measurement can be gained by recording the current through the sample during a test. This provision was made and a temperature range from about $-200°C$ to $+400°C$ became accessible by the following design (Fig. 4).

A copper block A containing the crystal holder

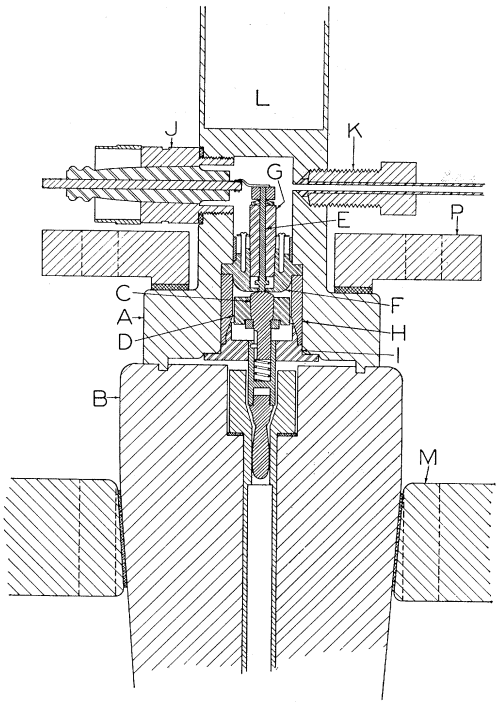


FIG. 4. Cross section through the breakdown apparatus.

in a central cavity serves as an electrical shield at ground potential and transfers heat from a temperature bath in L and around A to the sample. A metal pipe carries the high voltage up to the test piece through the porcelain insulator B, fitting a contact plug of the high voltage electrode C. A and B can be clamped together by a steel ring P anchored with bolts to the steel plate M which supports the porcelain cone. A groove cut into the upper surface of the porcelain insulator takes the gasket ring sealing the apparatus. At K a nitrogen tank can be connected for raising the pressure inside up to 100 atmospheres if necessary. The chamber can be evacuated through the hollow high voltage line.

The crystal holder inserted into the copper block is a small tubular box. A copper tube H forms its wall, and is closed at the bottom by an insulating piece I which carries the high voltage electrode. A copper guard electrode is screwed to the other end, holding the glass insulator G in place, which insulates the measuring electrode E from the guard ring. Fig. 5 gives a front view of this ground-electrode arrangement with a crystal plate in place after a breakdown test. The

[20] W. Franz, Verh. d. deutsch phys. Ges. **19**, 113 (1938); **20**, 27 (1939).
[21] Ch. Zener, Proc. Roy. Soc., London **145**, 523 (1934).
[22] A. v. Hippel, Zeits. f. Physik **75**, 145 (1932).

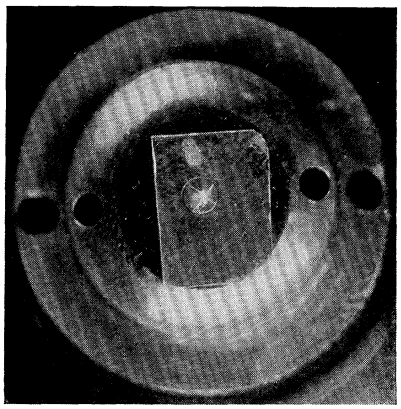


FIG. 5. Front view of ground-electrode arrangement with crystal (enlarged 1 : 2.5).

puncture in the center caused two cracks in the [110]-direction, progressing about to the edge of the center electrode. Guard and center electrode are separated by a spacing of only 1/1000 of an inch but are insulated from each other by a resistance of more than 10^{12} ohms. A spring contact transfers the current from the measuring electrode through a spark plug J into the current amplifier connected outside by a coaxial cable.

After the crystal has been properly adjusted on the ground electrode a weak spring is released, pressing the high voltage electrode against the test sample. The sample is then surrounded by some drops of an appropriate insulating material, or the whole little chamber may be filled under vacuum with some embedding dielectric. The temperature of the sample is controlled by a thermoelement in the copper block A very near to it, and by a thermometer in L, both readings being calibrated against a thermocouple which was inserted at the place of the test piece.

THE ELECTRICAL CIRCUIT

D.c. high voltage up to 75 kilovolts, steady and smoothly controllable, is delivered by a condenser charged from a power pack through the resistor r_1 (Fig. 6). This voltage reaches the insulator through a protecting resistor r_2 of 100 megohms. Here it is measured by the current drained out through the precision resistor r_3 of 1000 megohms. An amplifier provides the different voltage ranges and the power necessary to record the voltage by a 5-ma Esterline-Angus recorder.

The current flowing through the central section of the test piece is amplified by a feedback micromicroammeter and fed into a second current recorder. The recorders are coupled together; therefore current-voltage-time characteristics are obtained. The range of the amplifier extends from 10^{-11} amp. to $5 \cdot 10^{-6}$ amp. full scale; the instruments have an accuracy of one percent of full scale. The amplifiers have been developed and built in our laboratory by Mr. S. Roberts who has published a detailed description recently.[23]

When breakdown of the insulator occurs, a glow discharge lamp connected across the current amplifier lights up, protecting the instrument and indicating the failure. Simultaneously the voltage falls back to the resistor r_2, and the voltage recorder notes the breakdown voltage by the point where a steep break of the characteristic occurs (Fig. 7). The reading accuracy thus achieved makes full use of the high sensitivity of our equipment.

Our resistors, r_1 to r_3, are of the spiral-wound MV type of I.R.C. Resistor r_3 has been carefully checked several times against a high voltage wire-wound precision resistor, and the lower ranges of the voltage amplifier have been calibrated by d.c. precision instruments. Our results should therefore be reliable to one percent of the full-scale reading of our instruments; that is, to between one percent and two percent of the values observed, so far as the electrical circuit is concerned.

RESULTS

The dielectric strength of defined materials cannot be found by piling up many measure-

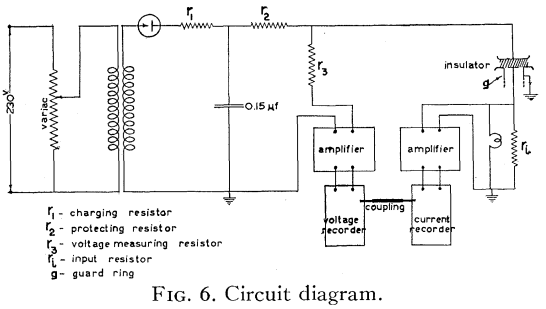


FIG. 6. Circuit diagram.

[23] S. Roberts, Rev. Sci. Inst. **10**, 181 (1939).

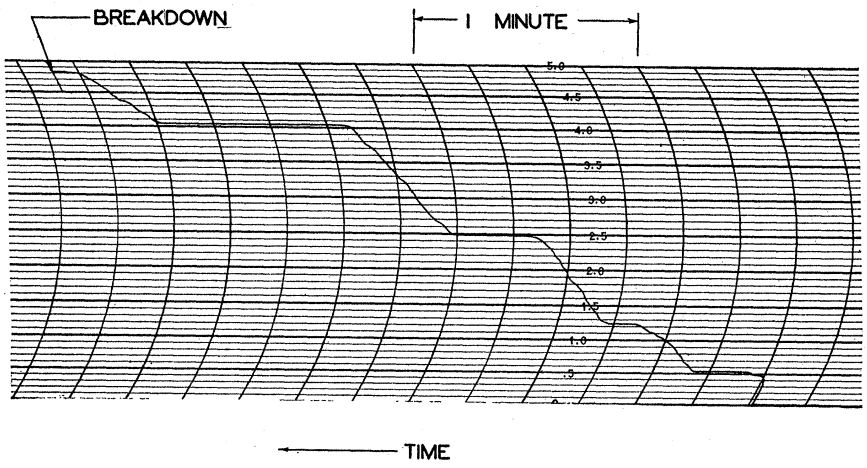


FIG. 7. Voltage record noting the breakdown voltage.

ments and averaging their results statistically. The current-voltage record during the test and the microscopic examination of the insulator afterwards give together the decisive evidence of whether or not an experiment was successful. We have not found a single case yet where the highest breakdown strength recorded was not at the same time the most reliable one. On the other hand, even the greatest care in preparing electrodes, sample, and guard ring did not prevent occasional erroneous low values produced by unnoticed faults in the test sample or in the arrangement. An extensive study of KBr single crystals revealed the principal aspects of the situation.

CURRENT-VOLTAGE-TIME CHARACTERISTICS

Figure 8 gives a characteristic sequence of sections of a current-time record for a KBr crystal at room temperature. The charging resistor r_1 was omitted in order to demonstrate the effect of fluctuating line voltage. Between the first five sections less essential intermediate parts of the long record have been left out. The last three sections present one continuous curve with the corresponding voltage record above it. The voltage setting and time scale are indicated below.

Four facts are demonstrated very clearly: After each voltage rise the current drops rapidly.

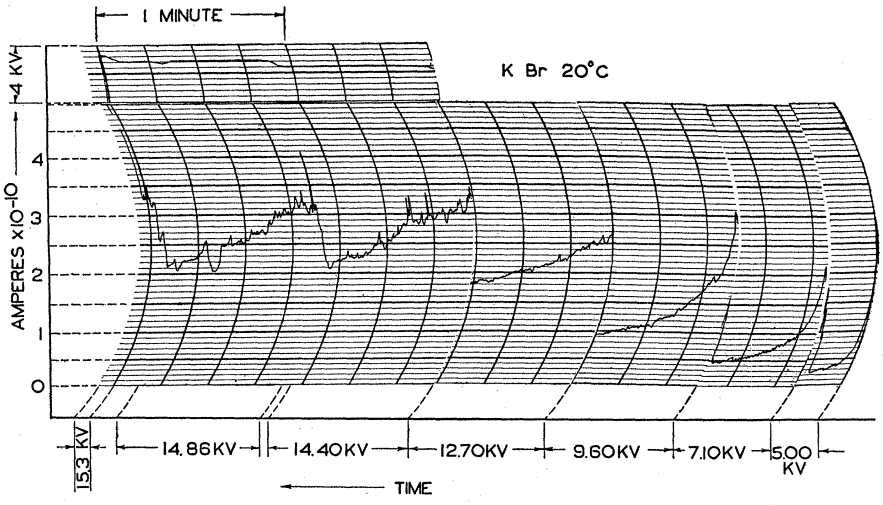


FIG. 8. Current-time record up to breakdown.

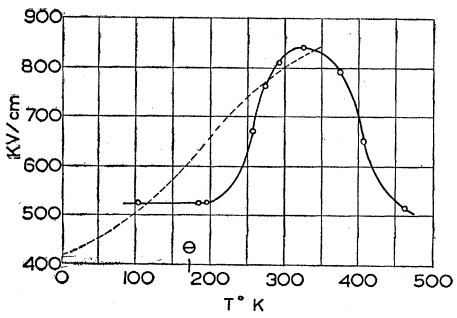


FIG. 9. The breakdown strength of KBr as $f(T)$. θ = Debye temperature.

With increasing field strength the decay becomes less steep and the curve shows more and more fluctuations. The breakdown itself appears as an abrupt discontinuity.

A comparison between current and voltage curves shows that the fluctuations are due partly to the line voltage. The deep hump about half a minute before breakdown, for instance, corresponds to a short voltage change of only one percent! Other fluctuations have their origin in the crystal itself, as one of us has found.[15] They are distorted in the record because our instrument needs about $\frac{1}{2}$ second for a full-scale deflection. The existence of fluctuations due to shot effect has already been proved by Haworth and Bozorth[24] several years ago.

The decrease of the current with time is mainly caused by polarization. An experienced observer can predict from the current slope and its ripples when the breakdown voltage is approached, and in this way can detect early failures of the material. A detailed study of the current curve, its true shape and its composition of electronic and ionic conduction, will be given in a later paper.

THE TEMPERATURE DEPENDENCE OF BREAKDOWN

In contradiction to the usual type of characteristic (Fig. 1) the temperature curve of KBr (Fig. 9) shows completely new and different aspects. The breakdown strength stays constant only below -80°C, increases sharply to a maximum at about $+50$°C, and decreases again. The current densities just before breakdown are plotted in Fig. 10.

These measurements give the decision we have sought (see p. 2): Frenkel's theory of thermal ionization can certainly not be true at temperatures below the maximum, because the slope of the voltage-temperature curve is contrary to his prediction. Also the current preceding breakdown has no direct connection to the breakdown phenomenon itself, as already stated recently.[15] The theory of Zener-Franz is ruled out, because a strong dependence of the breakdown strength on temperature has been found. The increase of breakdown strength with increasing temperature fits into the picture of electronic impact ionization given by one of us. The slope is much steeper than is predicted by Fröhlich's theory; the dotted line in Fig. 9 shows the temperature dependence according to his formula if the maximum is taken as reference point. It may be hoped that calculations of the temperature dependence on the basis of the avalanche theory results in a better agreement.

We have found a similar but even steeper characteristic for NaCl, while addition of AgCl to NaCl seems to quench the influence of temperature as should be expected. The measurements are not completed yet, because a new phenomenon was observed which delayed our program. At higher temperatures very low breakdown values with a clear indication of edge effect result if the voltage is not raised by small steps with large intervals of constant voltage between. Apparently some kind of progressive

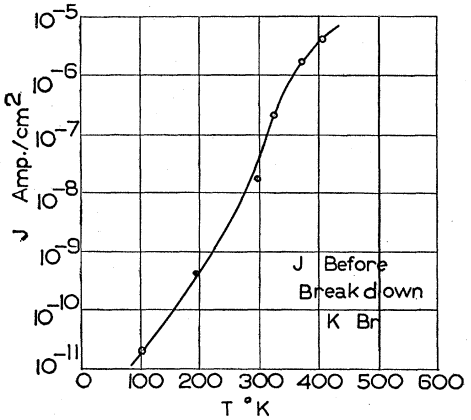


FIG. 10. Current density before breakdown of KBr.

[24] F. E. Haworth and R. M. Bozorth, Phys. Rev. **39**, 845 (1932).

formation of electrodes and sample takes place, the disturbances die down, and at last a reproducible highest breakdown value is reached, with the characteristics of a central breakdown. The formation once achieved seems to be permanent, because if a sample is treated in the way described and the voltage just before the expected breakdown value is cut down and after some time raised again very rapidly, the final high value results. It is hoped that a more detailed study of the current phenomena will give an understanding of this new effect. Until then the values published should be taken as preliminary results.

Simultaneously with our abstract for the Washington meeting a letter of Austin and Hackett appeared in Nature[25] reporting the breakdown strength of KBr as a function of temperature. The general shape of the curve given is in good agreement with our results. The absolute values lie lower than ours at the lower temperature end of the characteristic and have not been followed up above 350°K.

Some of the crystal samples used were kindly furnished by Professor D. C. Stockbarger, to whom the authors wish to express their gratitude.

[25] A. E. W. Austin and W. Hackett, Nature **143**, 637 (1939).

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On the Thermionic and Adsorptive Properties of the Surfaces of a Tungsten Single Crystal*

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The thermionic emission from a spherical tungsten single crystal has been observed. Photographs and diagrams show qualitatively the dependence of this emission on crystallographic direction. Nearly all the maxima and minima lie on a 110 zone. Similar observations have been made on the same crystal when caesium and barium were adsorbed thereon. Adsorption forces are largest for caesium on surfaces of highest work function. For barium the adsorption forces appear to be more dependent on surface structure as the force between the ion and its image contributes the major part of the adsorption energy. The behavior of adsorption as a function of crystallographic direction is such that the sphere surface may be approximated by that which would be obtained by carving the sphere from a perfect lattice. There is no evidence for a faceted or step-like microstructure. Emission from the spherical crystal when caesium is adsorbing on contaminated complex surfaces is more complicated in its dependence on crystallographic direction than when the surface is clean, and a map of the emission over the crystal changes its configuration with temperature. This behavior is not observed when the surface is clean or only slightly contaminated.

INTRODUCTION

IT has been realized for some time that thermionic and photoelectric emission (in an accelerating field) from a clean metal surface in good vacuum may vary by some orders of magnitude depending on which surface of the crystal is studied.[1–3] Ordinarily, polycrystalline aggregates have been used for experiment in the past, and consequently the data represent some sort of an average of the emissions from a great variety of surfaces.[4, 5] Theories thus far pro-

* Part of a thesis presented for the degree of Doctor of Science from the Department of Physics, Massachusetts Institute of Technology, June, 1938.

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[1] C. E. Mendenhall and C. F. deVoe, Phys. Rev. **51**, 346 (1937).

[2] E. W. Muller, Zeits. f. Physik **106**, 541 (1937).

[3] R. P. Johnson and W. Shockley, Phys. Rev. **49**, 436 (1936).

[4] L. H. Germer, Phys. Rev. **25**, 795 (1925).

[5] W. B. Nottingham, Phys. Rev. **49**, 78 (1936).

Cathode← →Anode

FIG. 2. Spark proceeding in NaCl.

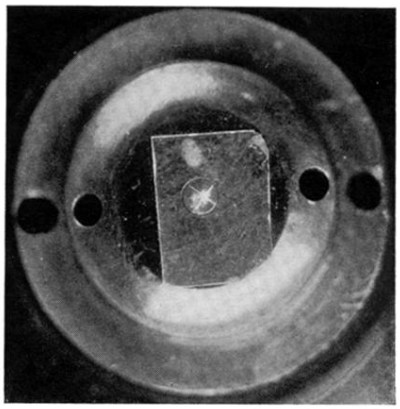

FIG. 5. Front view of ground-electrode arrangement with crystal (enlarged 1 : 2.5).