thermionic emission was determined as a function of the temperature. A Richardson plot of these data gave an average value for the work function of 5.24 electron volts. Consistent values for A were not obtained, but its value appeared to be in the neighborhood of 50.

The value of the photoelectric work function, 5.05±0.05 electron volts obtained below the Curie temperature agrees well with the value, 5.01 electron volts, obtained photoelectrically at room temperature by Glasoe.[12] The photoelectric work function of 5.20±0.05 electron volts determined at 1108°K is slightly lower than the value 5.24 electron volts determined thermionically at temperatures of 1150°K and above. This indicates that the photoelectric work function might further increase slightly with temperature before reaching a constant value, and thus before giving a typical Fowler variation of photocurrent with temperature. The above values are higher than the thermionic value 5.03 electron volts determined by Fox and Bowie[13] and 4.61 electron volts reported by Wahlin.[4]

V. ACKNOWLEDGMENT

The author wishes to express his indebtedness to Miss Helen Peterson and Mr. Donald E. Findley whose services have been valuable in carrying out the experimental work.

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[12] G. N. Glasoe, Phys. Rev. 38, 1490 (1931).

[13] G. W. Fox and R. M. Bowie, Phys. Rev. 44, 345 (1933).

PHYSICAL REVIEW VOLUME 76, NUMBER 1 JULY 1, 1949

Breakdown of Ionic Crystals by Electron Avalanches*

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(Received March 14, 1949)

In gases at moderate pressures breakdown occurs when the avalanche produced by a primary electron regenerates with certainty a new starting electron (Townsend condition). In ionic crystals the breakdown strength appears to be determined by the probability of exciting lattice vibrations, without reference to the electron supply. The theory as formulated to date predicts an increase of the breakdown strength with temperature. Confirming our previous results we find for both d.c. and a.c. an increase followed by a decrease. By extending our experiments to transient voltages of rise times down to 10^{-8} sec., and by adopting evaporated metal electrodes and various kinds of irradiation, the influence of field emission and of space charges on the breakdown strength and on the scatter of the breakdown values has been investigated. The occurrence of directional breakdown paths seems to be connected with the Brillouin zones of the crystal structure as the outcome either of electron diffraction or of collisions between electrons and directed lattice vibrations. The possibility of using crystals as Geiger counters is briefly discussed.

INTRODUCTION

While the sequence of events leading to electric breakdown in gases is today fairly well understood, in the case of solids basic questions are still asked concerning the mechanisms causing the destruction of these dielectrics. This lag in advance is partly due to the experimental difficulties of handling very high field strengths and partly inherent in the complexities of the solid state. In solids, electrons and ions do not lead a transitory existence as in gases, but appear as stable building stones of the structure in various forms of bonding. They may be mobilized under a variety of conditions, but remain, even in motion, strongly coupled to their surroundings. Space is at a premium; reaction products, once formed, are left behind as localized debris instead of being swept out in short order. Hence breakdown may occur in various ways: by thermal activation of charge carriers or by chemical decomposition, by dendrite formation or electron trapping, and finally, when these types of destruction are avoided, by electronic impact ionization.[1]

This last form of electronic breakdown has been of special interest to us for a number of years[2] because it holds the key to the development of a "physics of high field strength." It is the purpose of the present paper to re-assess the situation in the light of new evidence.

The major part of our experiments up to the present has been carried out with alkali halide single crystals since they may be grown and handled with ease, form a variety of compounds of cubic structure, and have been investigated in much detail at lower field strength, especially by Pohl and his co-workers.[3]

1. ORIGIN OF THE STARTING ELECTRONS

Statistical considerations as well as experimental evidence show that the concentration of conducting electrons in these crystals is negligible at room tem-

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* Sponsored by the ONR, the Army Signal Corps, and the Air Force under ONR Contract N5ori-07801.

[1] A. von Hippel, J. App. Phys. 8, 815 (1937).

[2] A. von Hippel, Trans. Faraday Soc. 42, 78 (1946).

[3] R. W. Pohl, Physik. Zeits. 39, 36 (1938).

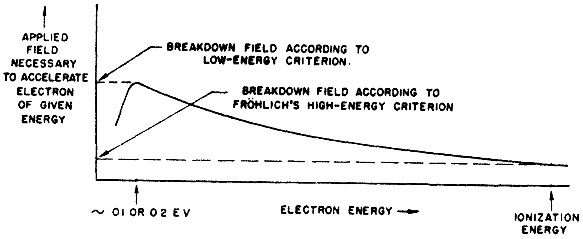

FIG. 1. The field necessary to accelerate an electron of given energy (schematic).

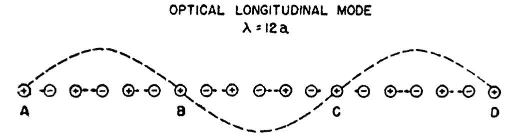

FIG. 2. Concentration of charges by a longitudinal mode. The arrows indicate the displacements of the ions from their equilibrium positions. *A* and *C* are regions of concentrations of negative charge, *B* and *D* of positive charge.

perature. A thermal activation energy of about 4 ev, required for their liberation from the halogen ions, would lead to a concentration of about 10^{-12} per cm^3. Release from the more shallow "Schottky defects" of the real crystal, i.e., from the *F*-centers where halogen ions are missing,[4] may be accomplished by activation energies of 1 to 2 ev, but still the concentration of conducting electrons should be much smaller than 10^{-5} per cm^3 in uncolored crystals (density of *F*-centers $\ll 10^{12}$).

This argument rules out any breakdown theory proceeding from the assumption that a large number of conducting electrons are present in these crystals in thermal equilibrium without field. However, it does not exclude offhand the possibility that the crystal itself can provide the starting electrons for breakdown as soon as the voltage has reached a limiting value. Any electron, while free momentarily, may be accelerated by the electric field; hence the equilibrium concentration multiplied by the vibration frequency causing thermal liberation corresponds to the number of conducting electrons actually available to the field per second and unit volume. This number may be about 10^{11} times larger than the equilibrium concentration, but its influence is counteracted by the fact that only a small volume (*ca.* 10^{-4} cm^3) is, in general, subjected to the breakdown field. Hence it seems that the starting electrons will normally be made available from external sources, as in gases.

That, in general, an ample electron supply exists, is illustrated by two experiments. When the concentration of electrons trapped in *F*-centers is raised to 10^{16} by strong irradiation with *X* or cathode rays, the breakdown strength of such colored crystals, in the dark, remains equal to that of colorless crystals in spite of the fact that the concentration of thermally-released conducting electrons must have increased by a factor of at least 10^4. Furthermore, when, by illumination of such colored crystals, an electronic photo-current of the order of the dark current is produced, no instantaneous lowering of the breakdown strength results.

In gases at moderate pressures breakdown occurs when a starting electron creates by impact ionization an avalanche of such height that it regenerates, with certainty, the initiating carrier (Townsend condition). At pressures of many atmospheres we find that electrons begin to be provided by field emission,† and similarly in the alkali halides the electron supply at breakdown field strengths seems, in general, so ample that is regeneration does not enter into the breakdown condition.

2. THE BREAKDOWN CRITERION

To produce breakdown, the conducting electrons must be accelerated to impact ionization. When slow, these electrons are not free in the usual sense, but are surrounded by a halo of counter-charges due to lattice polarization.[5] The mobility of such slow electrons, aptly named "polarons" by Pekar,[6] is being calculated at present by H. B. Callen.

In order to accelerate the electrons, the external field must replace the energy lost by them in excitation processes. In gases at lower field strengths the electrons are prevented from ionizing effectively by the electronic states of the gas molecules which dissipate their energy in light emission. In solids like the alkali halides, these electronic states have practically disappeared and the energy seems to be predominantly lost by the excitation of lattice vibrations.**

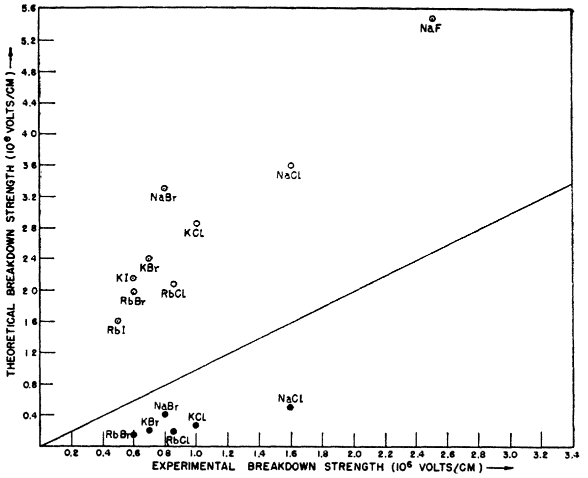

FIG. 3. Comparison of theoretical and experimental breakdown strength of alkali halides crystals at room temperature. Open circles are low energy criterion; black dots are Fröhlich's high energy criterion.

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[4] J. H. De Boer, Rec. trav. chim. 56, 301 (1937).

† From experiments on CO$_2$ by D. R. Young, to be published elsewhere.

[5] A. von Hippel, J. Chem. Phys. 8, 605 (1940).

[6] S. Pekar, J. Phys. U.S.S.R. 10, 341 (1946).

** There exists an excited electronic state in the alkali halides corresponding to the transfer of an electron from halogen to alkali

This energy loss, considered as a function of the electron energy, traverses a maximum because very slow electrons have insufficient energy and fast electrons insufficient time for such excitation. Consequently, the external field balancing this loss traverses also a maximum; it is this maximum field strength, allowing the acceleration of electrons of any energy, which we have identified with the breakdown strength of these crystals.[7] The slow secondaries produced by the primary electrons may be speeded up immediately, whereupon destructive avalanche formation results. Because the maximum of the vibration barrier is traversed when the electrons are still slow (*ca.* 0.1 to 0.2 ev), this breakdown condition may be called the "low energy criterion."

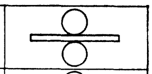
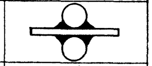
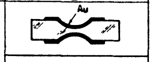
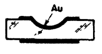
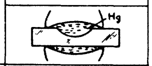
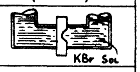
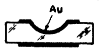
An alternative "high energy criterion," based on the same vibration barrier, has been proposed by Frölich[8] who postulates that the breakdown field strength corresponds to a field just capable of accelerating the electrons which have already acquired ionizing velocities (Fig. 1). Frölich assumes that the barrier is practically transparent, hence the conducting electrons will reach ionizing velocities by favorable impact with the phonons of the lattice; the crystal survives because of a delicate balance between ionization and recombination processes. Once this equilibrium is upset by the applied field, breakdown occurs.

Unfortunately, in Frölich's case, the secondary electrons formed by impact ionization would have to go through the same tortuous acceleration as the primary ones, that is, they would have to take the energy from the lattice vibrations instead of from the field. Thus the concentrated, rapid destruction produced by the avalanche mechanism of the "low energy criterion" is missing. Callen[9] has therefore recalculated the interaction between electrons and lattice vibrations on the basis of both criteria, following, in principle, Frölich's mathematical approach.

3. CONDUCTING ELECTRONS AND LATTICE VIBRATIONS

The calculation was made under the following simplifying assumptions. The electron is represented by a plane wave in a constant potential field, propagating, without scattering, through a perfect periodic lattice; scattering results from the disturbance of this periodicity by thermal vibrations. Of these vibrations only the longitudinal optical modes are strongly coupled to the electron, as Fröhlich and Mott[10] first pointed out. They cause a deviation from the normal charge density by a bunching of the positive and negative ions (Fig. 2), which leads to a polar perturbing potential. Ascribing the same amplitude to all optical modes and a frequency equal to the maximum optical frequency of the longitudinal modes, and assuming spherical symmetry of the wave-vector lattice, Callen obtained the breakdown field strengths of the alkali halides according to the "low energy criterion." They are compared in Fig. 3 with the experimental values.

TABLE I. Dielectric strength of KBr for various electrodes and treatments.

		Treatment	Dielectric strength megavolts/cm	Maximum spread megavolts/cm
1		None	0.38	0.33–0.46
2		None	0.88	0.75–1.1
3		Heated to 375°C in vac.	0.54	0.50–0.58
		Heated to 375°C in vac.	0.55	0.50–0.65
		Heated to 375°C in vac.	0.54	0.45–0.65
4		None	0.84	0.71–0.98
5		Stored in moist atmosphere for 2 days	0.79	0.56–1.1

In the same figure are shown the breakdown values predicted by the "high energy criterion." To produce the best possible agreement between theory and experiment in this case, Callen followed Fröhlich in setting the frequency of the vibration modes equal to the reststrahl frequency, the ionization energy equal to the first excitation, and including the longitudinal acoustical modes on equal footing with the optical modes in the perturbation calculation. In spite of this, the breakdown strengths predicted by the "high energy criterion" fall appreciably below the actual values. On the other

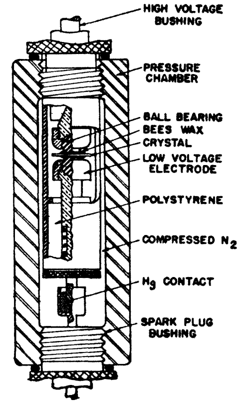


FIG. 4.

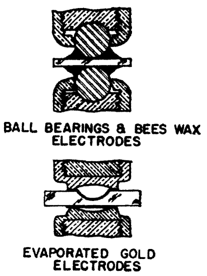


FIG. 5.

FIGS. 4 and 5. Ball-bearing electrodes.

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ion (see R. Hilsch and R. W. Pohl, Zeits. f. Physik **59**, 812 (1930)). Lying near the ionization limit, its effect has so far not been discerned.

[7] A. von Hippel, Zeits. f. Physik **75**, 145 (1932); Zeits. f. Elektrochemie **39**, 506 (1933).

[8] H. Fröhlich, Proc. Roy. Soc. (London) **A160**, 230 (1937).

[9] H. B. Callen, Technical Report XIII, Laboratory for Insulation Research, October, 1948.

[10] H. Fröhlich and N. F. Mott, Proc. Roy. Soc. (London) **A171**, 496 (1939).

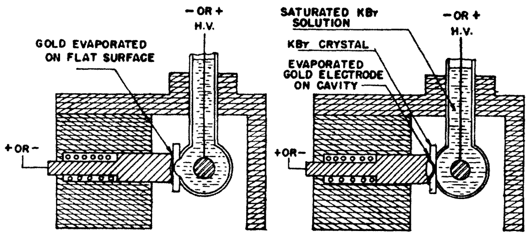

FIG. 6. Electrode arrangements for measuring the polarity effect.

hand, the breakdown values obtained with the "low energy criterion" are appreciably higher than those observed.

Notwithstanding other uncertainties in the present theory, such as the use of a perturbation method beyond its legitimate range of application,[3] there are reasons why the calculated values should fall too high. The theory assumes a completely ionic structure, while actually the heteropolar character of the crystals decreases rapidly with increasing ion sizes. Hence the polar disturbing potential, and with it the coupling between electron and lattice, has been overestimated. Furthermore the "low energy criterion" should not be read as identifying the extreme maximum of the vibration barrier with the breakdown strength. The secondary electrons are receiving already some kinetic energy from the ionizing electrons, and the primary electrons have a good chance to take over some vibration quanta from the lattice, hence the criterion may be satisfied by a field strength somewhat below the maximum. However, this uncertainty due to initial electron velocities may not be appreciable, because, as Callen shows,[9] the barrier is relatively flat near its maximum.

Seitz,[11] on the other hand, has recently calculated the barrier due to non-polar lattice vibrations and finds that it has a maximum at appreciably higher energies, and a magnitude which cannot be neglected. Thus, one should not expect close agreement with any theoretical calculation at the present stage of our knowledge, especially since a number of complicating experimental phenomena enter the picture also.

4. ELECTRODE EFFECTS

Our earlier experiments were, in general, carried out with thin crystal plates that were clamped with light spring pressure between properly rounded and highly polished metal electrodes (lately, ball bearings). A dielectric guard ring of wax or a plastic of the proper dielectric constant was built around the contact area (Fig. 4) and the sample holder placed in a pressure bomb under 50 to 100 atmospheres of nitrogen to avoid edge breakdown. Microscopic inspection made certain that the breakdown had taken place in the center of the electrode area.

To produce a more definite contact and to vary the electrode metal at will, we adopted the use of evaporated electrodes. Edge breakdown was avoided by hollowing the crystal on one or both sides by evaporation, lapping or grinding (Fig. 5). It now became apparent that the various possible electrode combinations had a distinct influence on the breakdown strength of the material. Table I summarizes the situation for KBr. In the pressure bomb the ball-bearing electrodes without the dielectric guard ring gave edge breakdown as expected and a low dielectric strength (1). The guard ring increased the breakdown strength about 230 percent (2). Evaporated gold or liquid mercury electrodes, had the effect of lowering the breakdown strength from about 0.85 to 0.55×10^6 v/cm (3). If cavity electrodes filled with saturated KBr solution were used, the dielectric strength increased again to about that observed with the ball-bearing electrodes (4). A similar increase was observed when crystals, after evaporation, were stored for two days in a moist atmosphere (5).

Consequently one may cite a d.c. breakdown strength 0.55 to 0.85×10^6 v/cm for KBr at room temperature, and similarly for NaCl 1.35 to 1.65×10^6 v/cm, depending on the electrodes and the pretreatment chosen.

By observing the effect of polarity on the breakdown strength in an arrangement employing one metal and one liquid electrode, we found that the low value results when the evaporated metal electrode is the cathode, while the high breakdown strength is obtained with the solution electrode as the cathode (Fig. 6). Field emission is only possible from a metal cathode; the likelihood of its occurrence will depend on the intimacy of the contact between metal and crystal. Hence the data of Table I and Fig. 6 seem to bear out the assumption that field emission is the cause of the lowered breakdown strength and that the scatter in the breakdown values of solids is partly due to the uncertainty in the onset of this emission.

Our experiments on colored crystals (see Section 1) indicate that electrons are available for producing breakdown, but that it occurs only when the proper field strength has been reached. The effect of the field emission, therefore, may be to create this critical gradient earlier by field distortion. A copious electron supply at the cathode produces a negative space charge and

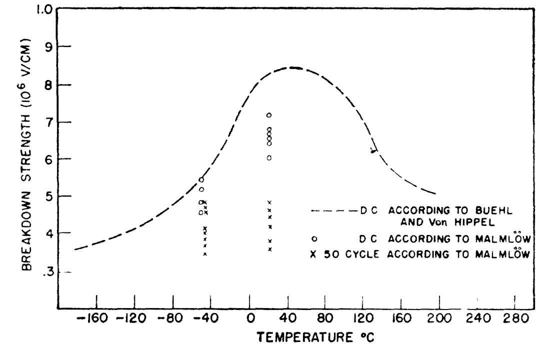

FIG. 7. Breakdown strength of KBr as function of temperature.

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[11] F. Seitz, Phys. Rev. 75, 1283 (1949).

thus raises the field strength in the anodic part of the crystal until the transport velocity of the charges reaches a balance with the supply.

5. TEMPERATURE DEPENDENCE

We previously established that the d.c. breakdown strength of the alkali halides increases with rising temperature to a maximum not far above room temperature and then falls.[12] Malmlöw,[13] investigating the breakdown strength of mica and KBr with d.c. and a.c., believes that he found temperature independence of the breakdown strength at 50 cycles/sec. (Fig. 7), and concludes that the d.c. temperature dependence is due to polarization. Seitz and Malmöw[14] have based an "exciton" theory of breakdown on this finding.

A careful recheck with d.c. and a.c., using the cavity electrodes, confirmed our previous results; identical values were obtained for d.c. and 60 cycle a.c. voltage (Fig. 8). We believe that Malmlöw's results, as their wide scattering indicates, were affected by edge effects that are difficult to avoid at a.c. if pressed-on electrodes are used. The change-over to cavity electrodes, while not altering the general trend of the temperature dependence, varies the shape of the characteristics appreciably. Figure 9 shows the corresponding situation in the case of NaCl.

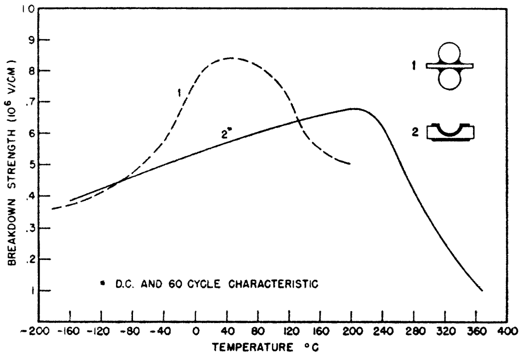

FIG. 8. D.c. breakdown strength of KBr as function of temperature.

The present theory of the interaction between electrons and lattice vibrations predicts only an increase of the breakdown strength with rising temperature (Fig. 10).[9] That the breakdown strength remains finite at 0°K according to this perturbation theory, is due to the zero-point vibrations.

It is not possible to discount the falling trend of the breakdown characteristic at higher temperature by assuming a change-over in the mechanism to thermal breakdown. The crystals under discussion have a heat conductivity approximating that of iron; the thin samples should therefore not be destroyed by local melting in the early phases of the process. The breakdown is abrupt, as before, and the breakdown paths are oriented in crystallographic directions. Hence, either the coupling strength between electron and lattice changes with increasing temperature in a way not yet foreseen by the theory, or some parameter, such as the field distribution across the crystal, varies with temperature. To discern this situation more clearly, the breakdown strength of KBr and NaCl was investigated with impulse voltages of increasing steepness.

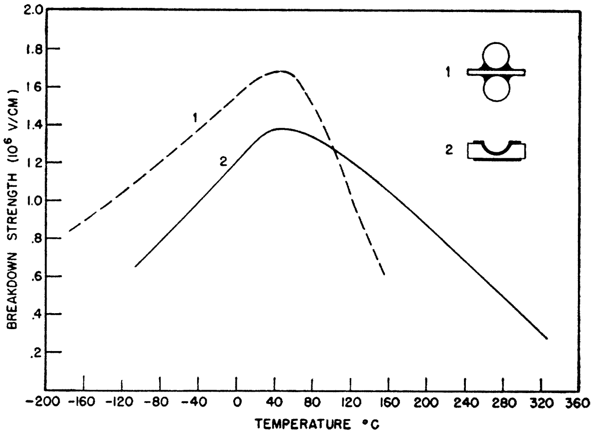

FIG. 9. D.c. breakdown strength of NaCl as function of temperature.

6. IMPULSE BREAKDOWN

The impulse equipment for producing and recording transients of rise times between 10^{-2} and 10^{-8} sec. was developed by D. R. Young and has been described elsewhere.[15]

A preliminary survey for KBr at room temperature showed that the static breakdown value is obtained as long as the linearly-rising transient reaches the breakdown field in times $\gtrsim 10^{-2}$ sec. At shorter rise times the breakdown strength first drops slightly with a minimum

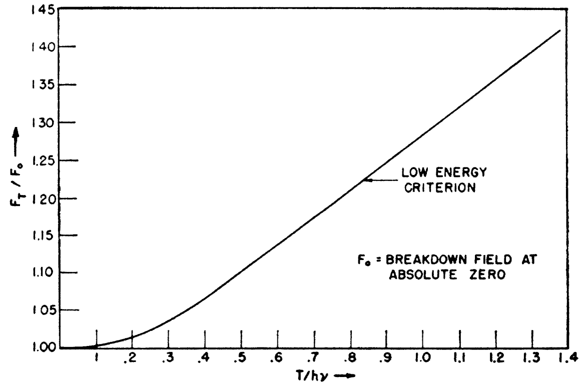

FIG. 10. Temperature dependence of breakdown field strength.

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[12] R. C. Buehl and A. von Hippel, Phys. Rev. **56**, 941 (1939); A. von Hippel and G. M. Lee, Phys. Rev. **59**, 824 (1941); A. E. Austen and W. Hackett, Nature **143**, 637 (1939).

[13] G. Malmlöw, Arkiv. f. Mat., Astr. o. Fys. **30B**, 1 (1944).

[14] G. Malmlöw and F. Seitz, Phys. Rev. **71**, 125 (1947).

[15] D. R. Young, Technical Report XIV, Laboratory for Insulation Research, December, 1948.

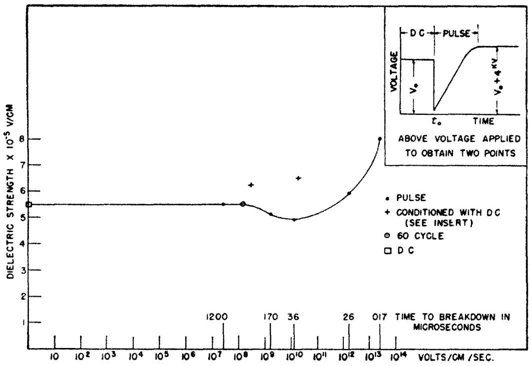

FIG. 11. Dielectric strength of KBr *vs.* rate of rise of applied field.

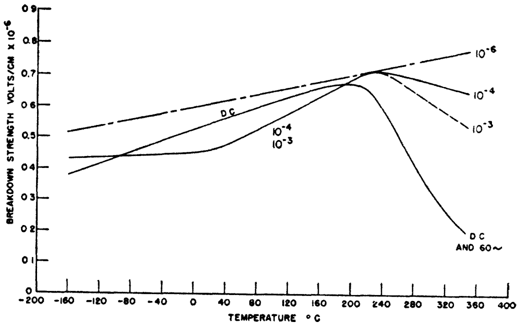

FIG. 12. Average breakdown strength of KBr as function of rise time.

near 10^{-5} sec. and then rises again, exceeding at 2×10^{-8} sec. the static value by about 50 percent[15] (Fig. 11). When a d.c. voltage of about 90 percent of the expected breakdown strength was applied and followed immediately by a fast-rising transient in the minimum region, the breakdown strength was increased above the static value and the maximum scatter of the breakdown value in successive experiments was reduced from about ±10 to ±2 percent.

Next the electric strength was investigated as a function of temperature with transients rising to breakdown in 10^{-3}, 10^{-4} and 10^{-6} sec. respectively (Fig. 12). A more complex situation appears: the 10^{-3} and 10^{-4} sec. characteristics, nearly indistinguishable in the lower temperature region, intersect the d.c. curve near 160°C. They then fall more steeply towards room temperature and flatten out below 0°C; above 200°C they diverge, the 10^{-3} sec. curve dipping more steeply as the temperature is raised. The 10^{-6} sec. transients produce higher breakdown values and a characteristic that rises nearly linearly with temperature.

This involved situation may result from three effects which superpose their temperature dependence on the predicted characteristic (see Fig. 10) and thus blur the effect of the thermal scattering of the electrons by lattice vibrations. The macroscopic breakdown strength is reduced by field emission, which causes a negative space charge near the cathode and raises the field strength towards the anode (see Section 4). Similarly, the breakdown voltage will be lowered by the migration of positive ions which are mobilized with increasing temperature, to form a positive space charge and raise the field strength in front of the cathode (Fig. 13). At low temperatures the negative, and at high temperatures the positive, space charge will dominate the situation, in both cases depressing the macroscopic breakdown voltage, while in an intermediate region the combination of both effects may give a relatively undistorted breakdown field. With increasing transient speed the positive space charge has less and less time to develop, hence the steep fall of the macroscopic breakdown strength at high temperatures tends to disappear (see Fig. 12).

Not only these space charge effects, but also the onset of the field emission, are time- and temperature-dependent. This one can see directly by observing the migration of electron clouds into crystals at higher temperatures.[16] Electron emission tends to set in after a formative period during which a surface layer of alkali metal appears to activate the cathode. Hence, for very fast transients at low temperature, the negative space charge may not have time to develop and the breakdown strength rises towards that observed for electrodes without field emission (see Figs. 8 and 12, and Section 4).

7. EFFECTS OF ILLUMINATION

The coloration of crystals by *F*-centers has no marked effect on the breakdown strength in the dark as mentioned above (Section 1), but strong illumination of such colored crystals or intense irradiation of uncolored crystals with x-rays (Fig. 14) during the breakdown experiment lowers the d.c. breakdown strength appreciably. This influence diminishes for transient voltages of increasing speed and disappeared in our experiment for breakdown times $<10^{-5}$ sec.

This phenomenon is closely connected with the space charge effects discussed in the previous section. Illumination in a strong field produces a positive space charge due to the emigration of electrons. The building-up of this space charge can be directly observed by current-time characteristics. Figure 15 gives one example. In consequence, the field gradient near the cathode increases and the breakdown voltage is lowered.

A more detailed investigation of the space charge effects is in progress.

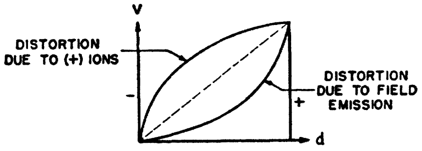

FIG. 13. Schematic view of field distortion.

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[16] A. von Hippel, Zeits. f. Physik **98**, 580 (1936).

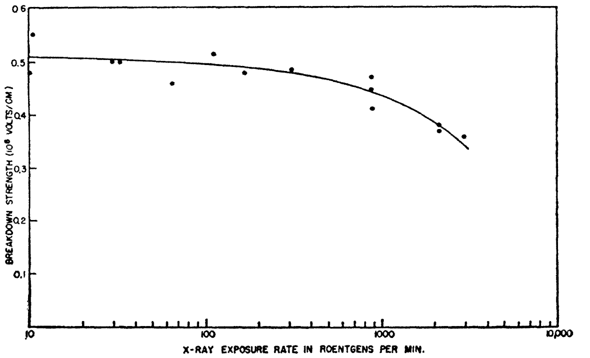

FIG. 14. D.c. breakdown strength of KBr crystals as function of x-ray exposure rate.

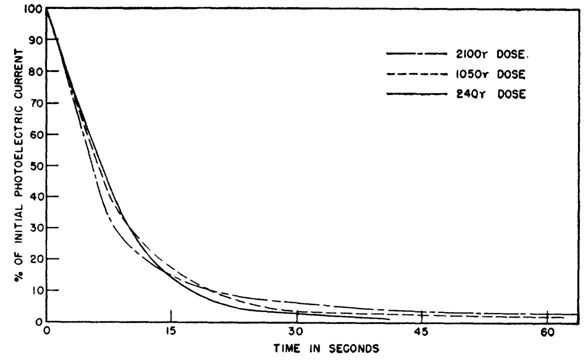

FIG. 15. Photoelectric current in KBr *vs.* time for constant voltage and illumination. *F*-band produced by three different x-ray exposures.

8. DIRECTIONAL BREAKDOWN

The breakdown in single crystals follows paths which are oriented in distinct crystallographic directions.[17] The possible directions are determined by the macroscopic crystal symmetry; the directions actually chosen depend, in addition, on temperature, voltage rise and crystal orientation.[18] The breakdown strength of the alkali halides seems to be independent of their orientation in the field.[19]

This independence of the breakdown strength from orientation is consistent with the "low energy criterion" (see Section 2). If the breakdown field strength determines the field, allowing the acceleration of very slow electrons, one should not expect much structure sensitivity, because the wave-length is long in comparison to the periodicity distance of the lattice and the electron interacts mainly with the long, isotropic virbations of the lattice.

In the later stages of acceleration the electrons, however, are bound to be affected by the crystal structure. The wave-length shrinks and electron diffraction becomes possible.[2] Simultaneously the electron begins to collide preferentially with the directed vibrations at the boundary of the Brillouin zones, and direction scattering may result because of the directed momentum transfer.[9] In both cases the same macroscopic symmetry relations are involved that govern the propagation of waves through periodic structures. Whether the diffraction of the electron waves, their interaction with directed elastic waves or some yet unknown phenomenon is the primary cause of the directional breakdown has to be decided by further experiments.

9. CRYSTALS AS GEIGER COUNTERS

Crystal counters at present make use of the fact that electrons can be liberated in solids by particle or photon bombardment and that these electronic currents can be measured directly or amplified as in the well-known primary or secondary photo-effects. If the signal could be amplified further by impact ionization as in the discharge of Geiger counters, much additional sensitivity could be gained.

Unfortunately, the prospect for the satisfactory operation of such counters does not appear too bright. It is true that Haworth and Bozorth,[20] from a careful analysis of the shot effect in glass and mica at high field strength, conclude they have measured electron avalanches below the breakdown strength. In our own experiments before the war,[21] we also found such avalanche noise in the alkali halides, were even able to produce it by illumination and pointed out the possibility of using these crystals as Geiger counters. However, we have so far no convincing evidence that these avalanches are not surface discharges. According to our present knowledge we would expect that an ionic crystal in a homogeneous field breaks down as soon as impact ionization sets in. Experiments to clarify this point beyond doubt are in progress.

The authors acknowledge the help of P. F. Ast, P. B. Kessel and D. O. Smith in carrying through the extensive measurements, and of V. Campbell in preparing crystal samples.

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