Predischarge Phenomena in Liquids

I. E. BALYGIN

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The results of an investigation of prebreakdown phenomena in transformer oil, castor oil, xylene and distilled water are presented. At voltages sufficient for breakdown, it is shown that ionization by collision occurs during the statistical delay period, resulting, in a time of approximately 10^{-8} sec, in the production of electron avalanches and small streamers. The latter do not always lead to cumulative breakdown, but are a preparatory stage. It has been established that breakdown can occur when the streamer current is greater than $5 - 8 \times 10^{-5}$ amp.

It is also shown that in distilled water with highly inhomogeneous fields the breakdown mechanism depends on the polarity of the point. If the latter is positive, then during the statistical delay period a positively charged column of very small radius is formed in about 5×10^{-6} sec.

IT is well known that both a definite voltage and sufficient time are required before a discharge can occur in a dielectric. The time interval between the application of a voltage, adequate for breakdown, to the electrodes and the appearance of a free electron, capable of initiating a cumulative discharge, is usually called the statistical delay time. It is virtually impossible to determine experimentally the moment at which such an electron appears, and consequently, for experimental purposes, the end of the statistical delay period is taken to be the moment at which there occurs either a noticeable voltage drop across the electrodes or a cumulative increase in the electrode current.

The question of what occurs in a dielectric during the statistical delay time is of great interest. To shed light on these phenomena we have performed a number of experiments in liquids. The results of these experiments are reported below.

In solid or liquid dielectrics, it is well known that the energy of free electrons, moving under the influence of an applied field, is dissipated in excitation of oscillations in nearby atoms and molecules, electronic excitation in these atoms and molecules, molecular dissociation, and collisions of the second kind[1,2]. The dielectric, as it were, hinders the development of discharges at low field intensities. Another barrier to the development of a discharge is the capture of free electrons by molecules of the material[3]. It is reasonable to believe that these processes depend on the physico-chemical properties of the dielectric. If ionization takes place during the statistical delay period, but a cumulative discharge does not occur, these physico-chemical properties should have some effect on the character of the predischarge phenomena. The experiments reported here indicate that such is the case ,at least in certain liquid dielectrics.

According to present-day ideas of the structure of the amorphous phase, a liquid consists of separate crystallite aggregates comprising several tens of molecules in a group[4]. These groups, as might be supposed, do not consist of rigidly bound molecules but are constantly being dissolved and reformed. In certain liquids at least, there is some justification for the assumption that ions of opposite sign are contained in these crystallite groups. This assumption is based on the experimental finding that the electrical conductivity σ of heptane is very small at low field intensities. With increasing intensities, however, there is a pronounced rise in σ. It is believed[5] that this increase results from the destruction of electrically neutral ion pairs by the externally applied field. It is also possible to explain the increase in σ on the basis of crystallite ion groups.

It is well known that a wave function has a strong maximum at any defect of a perfect crystal, including those close to the surface of the crystal.[6] Consequently, it may be expected that extensive trapping takes place at the boundary surfaces of a crystal. In this connection it may be assumed that the size of the crystallite aggregates, the stability of the bonds, the nature of the levels, and various other characteristics, pertinent to the formation and

[1] A. von Hippel and G. Lee, Phys .Rev. 59, 824 (1941).

[2] G. Hurd, Gen. Elec. Rev. 51, 26 (1948).

[3] A. Von. Hippel, J. Appl. Phys. 8, 815 (1937).

[4] P. P. Kobeko, *Amorphous Materials*, Academy of Sciences Press (USSR), 1952, p. 17.

[5] K. A. Reiss, Ann. Physik 28, 325 (1937).

[6] I. E. Tamm, Z. Physik Sowjetunion 1, 733 (1932).

maintenance of an electric discharge, should also depend on the physico-chemical constitution of the liquid.

The experiments were performed using the apparatus shown in Fig. 1. The voltage from a pulse

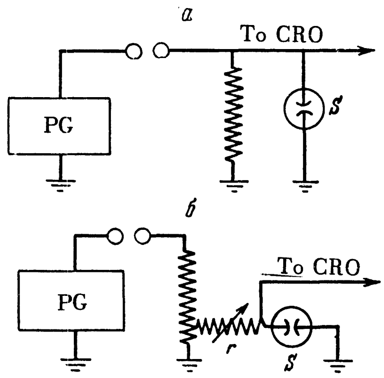

FIG. 1. Experimental set-up

generator *PG* was applied directly to the sample under observation *S* (diagram *a*). This voltage was also applied, through a capacity divider, to the plates of a high-voltage oscilloscope which had been calibrated for the measurements. Certain of the experiments were performed using the scheme shown in diagram *b*. In these cases an ohmic resistance *r* was connected in series with the discharge gap in order to limit the discharge current.

It has been stated in the literature that relatively small variations of the voltage across the spark gap are to be associated with the onset of stepped leaders or streamers[7,8] and that the production of streamers causes a sharp drop in the voltage[9] even within so short time as 4×10^{-9} sec. Hence impact ionization and the resulting production of avalanches and streamers can be observed through changes in the pulse amplitude as seen on the oscillographs.

In Fig. 2 are presented oscillographs recorded with scheme *a* (I-III) and scheme *b* (IV-VI) of Fig. 1. The period of the reference sinusoidal oscillations along the abscissa axis in Oscillographs I-III is 5×10^{-7} sec. Explanatory data for these oscillographs are given in Table 1.

[7] T. E. Allibone and J. M. Meek, Proc. Roy. Soc. (London) **169A**, 246 (1938); **166A**, 97 (1938).

[8] N. A. Kaptsov, *Electrical Phenomena in Gases and Vacuum*, GITL , 1950, p. 582.

[9] F. Dunnington and H. J. White, Phys .Rev. **46**, **99**, (1934).

Inspection of Oscillograph I reveals that the statistical delay time in transformer oil was greater than 30 μsec. During this period there was intense ionization by impact with resulting avalanche production; this can be seen from the nicks (1, 2, 3, 4) in the top of the pulse. Similar results were obtained in the tests performed in castor oil (Oscillograph II). In the latter case it is worthy of note that a small avalanche was formed immediately after the application of the voltage (1). During the statistical delay time, which lasted for 50μsec, three partial discharges were recorded; as in the transformer-oil test, two of these occurred immediately prior to the terminal discharge of the gap.

In the transformer-oil test, as can be seen from Oscillograph I, the avalanches appeared within small time intervals (1-3.5 μsec) and the cumulative discharge was initiated within 3 μsec after the the last avalanche. It would seem that the avalanches prepare the way for the formation of the discharge.

In the castor-oil test the avalanches appeared at considerably longer time intervals (3.5-28 μsec), but again the cumulative discharge appeared within 3.5 μsec after the last avalanche.

For strongly inhomogeneous fields, it appears that ionization processes occur during the so-called statistical delay time; these do not cause direct breakdown but constitute a preparatory stage in that ionization processes in the discharge gap lead to the production of volume charges. From a knowledge of the prebreakdown time (statistical delay time) for strongly inhomogeneous fields it is possible to ascertain the time during which impact ionization occurs with the resulting production of avalanches and small streamers. The latter processes do not initiate a breakdown but act to set the stage for it. It should be remembered that at the same time there are processes taking place in the dielectric which tend to inhibit the development of a discharge. In the main, these same results were found in the tests with homogeneous fields; a typical case (transformer oil) is shown in Oscillograph III of Fig. 2. At intensities of 315-350 kv/cm, breakdown did not occur in spite of the fact that the prebreakdown phenomena were relatively strong. Almost immediately after the pulse reached its highest amplitude (0.5 μsec) 350 kv, nicks can be seen in the top of the pulse; these are similar to those which were observed in the tests with inhomogeneous fields. In certain cases the nicks appeared somewhat later (2). A large number of rather weak lines — short-term fluctuations in the voltage which are indicated by arrows — can be seen in Oscillograph III. These all appeared within a time interval less than a microsecond. These small voltage fluctuations are undoubtedly caused by electron avalanches which result from impact ionization in the oil. Small streamers, which are formed at the very beginning, ionize the interelectrode gap and produce a number of relatively low-intensity avalanches with lifetimes less than 10^{-8} sec. Because of the various electron-energy loss mechanisms and electron capture in the potential within the crystallites, a discharge did not occur.

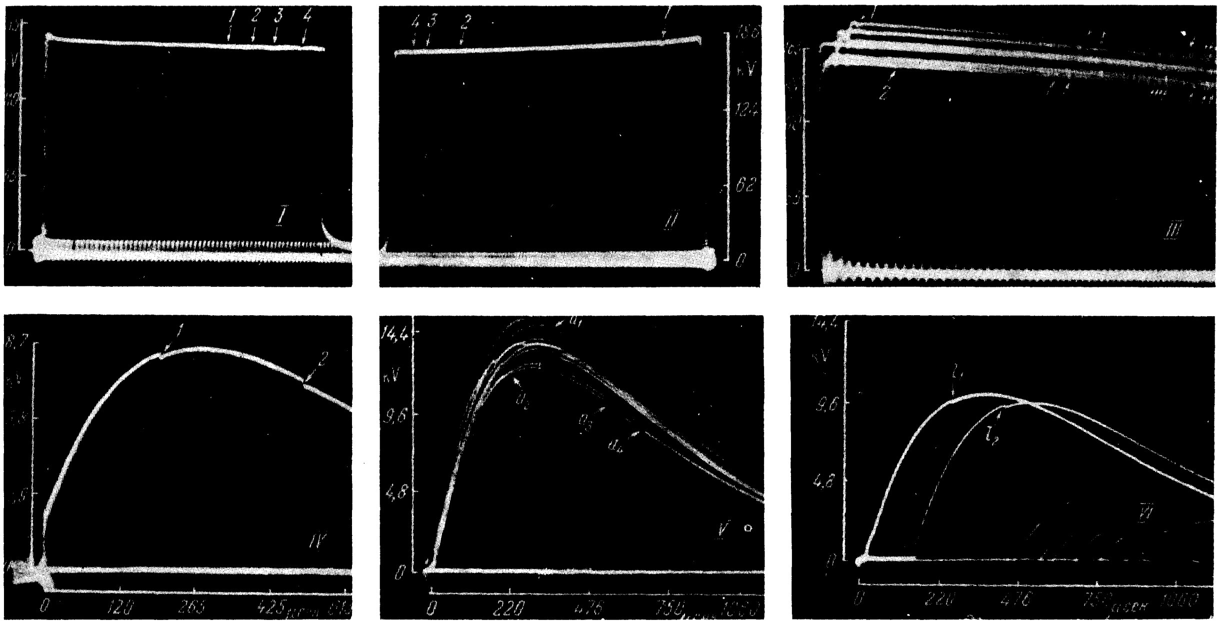

FIG. 2. Oscillographs of the voltage behavior at breakdown and during the prebreakdown period in transformer oil, castor oil, xylene, and paper impregnated with transformer oil.

It has been shown experimentally that small electron avalanches in CCl_4, mineral oil and certain other liquids can also be produced in an almost uniform ac field at very low field intensities (several kilovolts per millimeter)[10].

Oscillographs IV-VI of Fig. 2 were also made with uniform electric fields but using small spark gaps and limiting of the discharge current. For comparison purposes, tests were carried out on condenser paper saturated under vacuum with transformer (condenser) oil (Oscillograph IV). This oscillograph was made using a limiting resistance (see *b*, Fig. 1) of 3×10^6 ohms. The partial discharges could not be seen when smaller values of *r* were used. Two well-defined incipient discharges (1 and 2) can be seen. In both cases these were limited to partial discharges because the formation of a complete discharge was inhibited. It may be noted that this type of oscillograph could not always be reproduced in subsequent experiments.

In the formation of a breakdown and the increase of the current in the discharge gap, a definite voltage appears across the resistance *r*. Since the dominant resistance in the circuit is that of the resistance *r*, the major part of the voltage drop appears across this resistance when a breakdown occurs. For a drop of 180 volts and $r = 3 \times 10^6$ ohms the current at the instant of discharge was approximately 6×10^{-5} amp. With this current and the given intensity of the applied field, a cumulative breakdown can be cut off by inhibitory processes.

Oscillographs V in Fig. 2 were obtained in experiments with xylene from which the volatile component had been distilled off at a temperature of 110-129°. These experiments were carried out with steel spheres 11 mm in diameter, a gap of 0.2 mm and a limiting resistance of 5×10^6 ohms. The pulse voltage was increased gradually to the value at which breakdown occurred, which was 7×10^5 kv/cm; at smaller intensities, however,

[10] S. Yamanka and T. Suita, J. Phys. Soc. Japan 8, 277 (1953).

numerous breakdowns were observed even before the time at which maximum amplitude of the pulse was reached. The current for partial breakdowns $a_1 \ldots a_4$ was roughly 5×10^{-5} amp. This type of partial breakdown was not observed in subsequent experiments if r was less than 3×10^6 ohms. When the current is limited, a highly conducting bridge is not always formed when the gap breaks down. After some deionization of the discharge column, the latter continues to break down but at considerably lower voltages.

The appearance of partial discharges $a_1 \ldots a_4$ is an indication that impact ionization, with the production of electron avalanches and possibly streamers, takes place up to the moment of breakdown. Complete breakdown of the gap, however, occurs only at a definite intensity of the applied field.

TABLE 1

Reference data for oscillographs in Fig. 2.

Oscillograph no.	Liquid	Electrodes	Distance between electrodes in mm	Size of resistance used to limit discharge current.	Time in μsec required for first avalanche to appear after pulse reached greatest amplitude.
I	Transformer (condenser) oil	Negative (steel) point and brass plane	50	—	19.5
II	Castor oil	Positive point and plane	150	—	6.5
III	Transformer (condenser) oil	Brass spheres (nickel-plated) $d = 62.5$ mm	10	—	0.5—3
IV	Four layers of condenser paper-total thickness 40μ. Saturated under vacuum with condenser oil.	Steel sphere $d = 11$ mm and brass plane in transformer oil	4×10^{-2}	3×10^6	at maximum amplitude
V	Xylene with volatile fraction distilled off at 110-129°.	Steel spheres $d = 11$ mm	0.2	5×10^3	"
VI	Vacuum dried castor oil.	"	0.2	5×10^6	"

In Fig. 2 VI are shown oscillographs obtained in tests with vacuum-dried castor oil. The limiting resistance was 5×10^6 ohms. Inspection of these oscillographs reveals that breakdown of a gap with $l = 0.2$ mm occurred at about 480 kv/cm, but that with a constant pulse the discharge began to form at somewhat lower intensities (points l_1 and l_2). In these partial discharges the current was $4 - 6 \times 10^{-5}$ amp.

The data on current strengths in partial breakdown given here indicate that total breakdown can occur for currents greater than $5 - 8 \times 10^{-5}$ amp.

Several other predischarge processes were observed in the distilled water experiments. For a voltage pulse this liquid may be assumed to be an insulator. The oscillographs in question were made using the arrangement shown in Fig. 1 (scheme *a*) and are shown in Fig. 3, while the explanatory data are given in Table 2. The period of the reference sinusoidal oscillations in Oscillographs V and VI was also 5×10^{-7} sec. The reproducibility of these oscillographs was found to be excellent.

The lower two oscillographs in I of Fig. 3 are for cases in which breakdown did not occur. These give an indication of the length of the applied pulse (343 μsec from maximum to half amplitude). In the other pulses, in which breakdown did take place, the length of the pulse is found to be shortened. This results from the fact that during the predischarge period there is an uninterrupted flow of the ever-increasing prebreakdown currents which prepare the gap, so to speak, for breakdown. Since the form of the pulse remains the same, the duration of all these pulses in I of Fig. 3 should also be the same. These prebreakdown processes in distilled water have also been observed with homogeneous fields[11]. It is evident from examination of the oscillographs in Fig. 3 that a more rapid decay of the pulse is to be associated with higher amplitudes, i.e., a greater current flows across the spark gap during the prebreakdown time. In a paper which has been published earlier, the assumption has been made that electrons which move from the negative point into the region of smaller field intensity are captured by water molecules and form volume charges during the formative lag period; these charges lead to an increase in the field intensity and are responsible for the cumulative breakdown[12].

In addition to the ionization processes, the point,

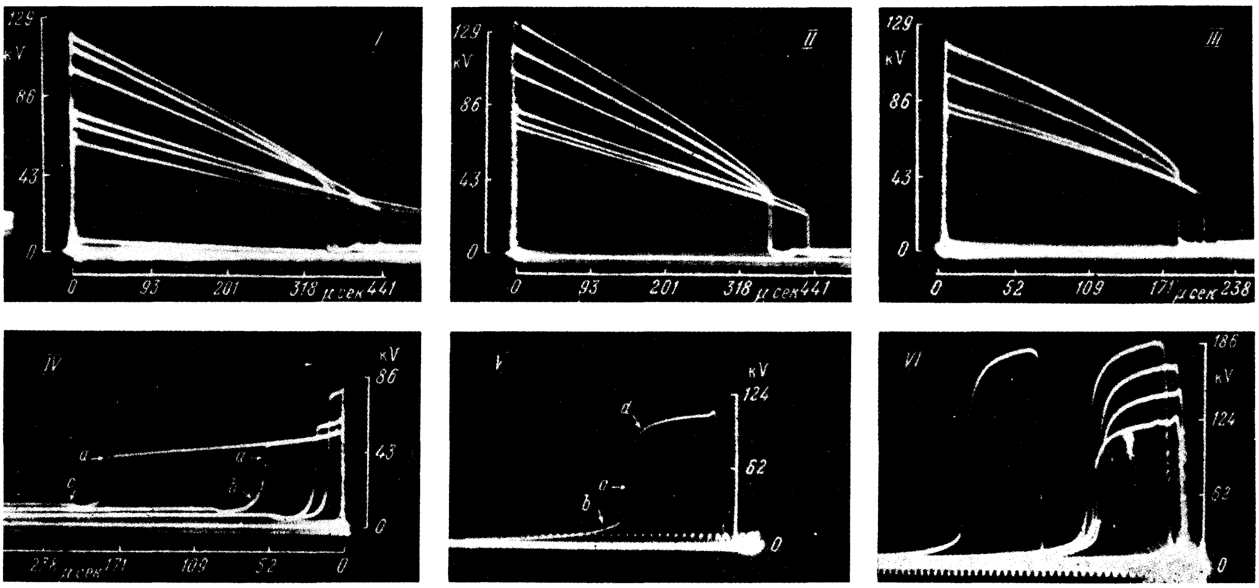

FIG. 3. Oscillographs of the voltage behavior at breakdown and during the prebreakdown period in distilled water.

itself, can act as an electron source[13-15]. It follows from Oscillographs I -III of Fig. 3 that the magnitude of the overvoltage plays a relatively small role in the general dynamics of the cumulative discharge. The length of the formative lag period, however, is strongly affected by the length of the spark gap; this lag is decreased in a pronounced way when the gap is reduced. For example, with a gap spacing of 100 mm and maximum amplitude $U = 77$ kv for the applied pulse, the formative lag period before breakdown occurs is 202 μsec; with a gap of 150 mm, however, and the same value of U, the time lag is 423 μsec. This large difference may be attributed to the length of time required for electrons and negative ions to travel to the positive electrode.

With a high intensity field, it may be assumed that impact ionization occurs within a region of radius R_1 during the prebreakdown period. This process leads to the production of electron avalanches; simultaneously, however, electron capture is taking place, especially in the region $R_2 > R_1$. Thus avalanche production should occur in accordance with the well-known relation

$$n = n_0 \exp\left\{\int_0^{R_1} (\alpha - \alpha_1)\, dx\right\} \quad \text{for} \quad x < R_1,$$

[11] I. E. Balygin, J. Exptl. Theoret. Phys. (U.S.S.R.) 25, 738 (1953).

[13] D. W. Goodwin and K. A. Macfadyen, Proc. Phys. Soc. (London) B66, 85 (1953).

[14] T. J. Lewis, Proc. Phys. Soc. (London) B66, 425 (1953).

[15] Bragg, Sharbaugh and Crowe, J. Appl. Phys. 25, 382 (1954).

[12] I. E. Balygin, J. Tech. Phys. (U.S.S.R.) 24, 338 (1954).

where, as usual, α is the number of ionization events per centimeter of path length due to one electron and α_1 is the number of electron attachments in the path length. In the region $R_2 > R_1$, electrons due to incipient avalanches will be captured by water molecules or at the surfaces of the crystallites. The rate at which this process proceeds is proportional to the average number of capture centers K and the number of electrons n' in an avalanche. Thus we can write

$$dn'/dx = -\mu n'k \text{ and}$$

$$n' = ne^{-\mu k(x-R_1)} \text{ for } x > R_1,$$

since $n' = n$ when $x = R_1$. Hence, the avalanche is attenuated; on the other hand, the negative ions with captured electrons, moving toward the anode, form an ever-increasing field up to the moment at which breakdown occurs.

In the case of a positive point and plane, an altogether different kind of effect is observed in distilled water. At threshold voltages, it can be seen from Oscillographs IV of Fig. 3 that the drop in the amplitude of the pulse does not occur. The decay of the voltage becomes noticeable only in strongly overvolted gaps (d, Oscillograph V).

This decay may be called the initial (prebreakdown) stage in the development of the breakdown. Immediately afterwards there is a second stage – a sharp drop in the voltage (a, Oscillograph IV and V). The third stage is characterized by a relatively slow drop in the voltage (b, Oscillograph IV) and the fourth by a slight increase (c, Oscillograph IV). It is assumed that in the first stage a positive streamer column is formed which propagates from the anode to the cathode. In the effective short-circuiting of the electrodes due to this column, the current increases and the voltage "collapse" occurs. It is evident that the magnitude of this "collapse" depends on the amplitude of the applied pulse; this is apparent on inspection of the curves in Fig. 4 (U_1 . . . U_6) which have been drawn from Oscillographs IV-VI of Fig. 3. After a time, which is taken as zero in Fig. 4, there is a pronounced drop in the amplitude of the pulse (beginning of breakdown). In the absence of an overvoltage, the first stage of the development of the discharge remains invisible because of the low intensity of the process and the "collapse" seems to occur suddenly. It would seem that the formation of a positively charged column occurs during the predischarge period, which as we have indicated, is called the statistical delay. In oscillographs IV, this time is 184 and 52 μsec. Judging from the size of the "collapse", which is very small, it appears that the radius of the column is also very small. When the amplitude of the applied pulse is increased, this radius increases and there is a corresponding growth in the "collapse" U_1 . . . U_6 of Fig. 4. In the third stage the radius of the column grows slowly because of photoioniza-

TABLE 2

Reference data for oscillographs in Fig. 3

Oscillograph No.	Electrodes	Distance between electrodes in mm	Maximum pulse amplitude in kv	Voltage at which voltage "collapse" occurred in kv.	Formative lag period in μ sec	Statistical delay time in μsec
I and *II*	Negative steel point and brass plane	150	60—132	25,5—34.0	423—348	—
III	"	100	77—115	30—43	202—180	—
IV—*VI*	Positive steel point and brass plane	150	53—185	39—125	0—10	184—0

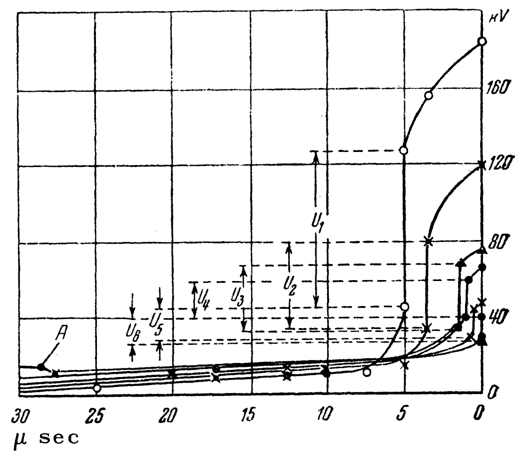

FIG. 4. Curves showing the decay of the voltage in the formation of a discharge in distilled water (positive point and plane); $U_1 = 84$; $U_2 = 44$; $U_3 = 34$; $U_4 = 22$; $U_5 = 16$; $U_6 = 12$ kV

tion and thermal ionization. During this stage the temperature of the column increases and gas is produced; this process is terminated with an "explosion" and the ejection of liquid[11]. At this point molecules of non-ionized liquid penetrate the column, the current diminishes, and there is a corresponding increase in the voltage (fourth stage; *A* of Fig. 4 and *C* of Oscillograph IV, Fig. 3).

It is evident from Oscillographs V and VI of Fig. 3 that even in the case of a very large overvoltage (3.5 times) a relatively long time (about 5 μsec) is reguired for the formation of the positive column which short-circuits the electrodes. During this period, as may be seen from the nicks in the front of the pulses in Oscillographs VI, small streamers are formed; these do not start breakdown but cause a certain amount of ionization in the gap. The development of the filamentary positive column from the positive point also depends on random factors. This is seen in Oscillographs IV of Fig .3 in which breakdown occurs after a lag of 184 μsec in one case and 52 μsec in the other although the amplitude of the applied pulse was the same in both cases.

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Energy Spectrum of the Penetrating Particles of Extensive Cosmic Ray Showers

I. I. SAKVARELIDZE
Physics Institute, Georgian Academy of Sciences, USSR

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The depth-intensity curve of penetrating shower particles absorbed in lead and rock of water equivalent of 3.7, 5.9, 27, 65.5 and 148 meters has been measured. The studies are carried out at an elevation of 400 meters above sea level. On the basis of the resulting path lengths, the integral energy spectrum of penetrating shower particles is found in the form of a power function. This function has the form $E^{-\gamma}$, where $\gamma = 0.62 \pm 0.05$ (for the energy interval from 5×10^8 to 3×10^{10} ev).

INVESTIGATION of the characteristics of penetrating shower particles is essential in connection with the mechanism of generation of extensive cosmic ray showers that is proposed by a group of Soviet students working under the guidance of Skobel'tsyn[1-4]. According to their idea, the formation of an extensive atmospheric shower appears to be the result of a nuclear-cascade process in which the main role is played by nuclear-active and penetrating components. The latter carry the major part of the energy of the whole shower, and determine the significant characteristics of the shower, its spatial structure, and the energy balance among its different components.

To this day, studies of the nature of extensive cosmic ray showers furnish the only key to understanding the interactions of high energy (of the order of 10^{15} - 10^{18} ev) particles with matter, and those physical processes which accompany these interactions. From this point of view, investiga-

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[2] D. V. Skobel'tsyn, Dokl. Akad. Nauk SSSR 67, 45 (1949).

[3] D. V. Skobel'tsyn, Dokl. Akad. Nauk SSSR 67, 225 (1949).

[4] G. T. Zatsepin, Dokl. Akad. Nauk SSSR 67, 993 (1949).