Dielectric Breakdown in Ionic Crystals

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A critical consideration of the theories of dielectric breakdown in ionic crystals proposed by Seeger and Teller and by Fröhlich is given. It is shown that the main difference in the formulae for the breakdown field is due to the fact that Seeger and Teller make the unjustified assumption that an electron transfers energy to the lattice vibration without changing its direction of motion. A mistake in the interaction between an electron and an ion, used by Seeger and Teller, is corrected. The author's condition for breakdown is discussed in greater detail than has been done hitherto.

§1.

A THEORY of dielectric breakdown in polar crystals has recently been given by the author.[1] More recently, Seeger and Teller[2] have presented a theory which differs in important respects from that of Fröhlich. The purpose of this paper is to show where, in the author's opinion, the theory of Seeger and Teller is incorrect, and to discuss the fundamentals of Fröhlich's theory in rather greater detail than has been done hitherto.

In both theories it is necessary to consider the interaction of free electrons in a polar lattice with the lattice vibrations. This leads to expressions for the mobility of electrons which, in the case of slow electrons, can be compared with experiments on photoconductivity. This will be done in another paper[3] in which it will be shown that Fröhlich's theory, modified for slow electrons, leads to a fair agreement with experiment.

Both authors follow von Hippel's original ideas in considering an electron with an arbitrary energy E in the conduction band of the crystal (i.e., the first empty band of allowed energy levels). The energy E is measured from the lowest state in the conductive band. There is evidence, as we shall see, that for high fields a certain number of electrons find their way into the conduction band.

In an external field F such an electron will lose energy to the lattice vibrations of the solid and gain energy from the field. If the field is strong enough, the energy of the electron will increase indefinitely until it is great enough to produce a secondary. For small fields, on the other hand, the electron will be slowed down until it reaches thermal energies. Both authors agree that breakdown will take place when the field is strong enough to accelerate electrons having energy greater than a critical value E_c. They disagree however as to (1) the value to be taken for E_c, and (2) the method of calculating the rate of loss of energy to the lattice.

§2. THE RATE OF LOSS OF ENERGY OF AN ELECTRON IN A POLAR LATTICE

The rate of loss of energy may be calculated either per unit time $-dE/dt$ or per unit path *in the direction of the field F*. We denote this by $-dE/dx$. They are connected by

$$dE/dx = (dE/dt)(1/\omega F), \tag{1}$$

where ω is the mobility of the electron. Instead of ω, the conception of the time of relaxation τ may be introduced, defined by $\omega = (e/m)\tau$ as, for instance, by Fröhlich.[1] The magnitude

$$\bar{v} = \omega F = e\tau F/m \tag{2}$$

is the average velocity over several collisions. Seeger and Teller, on the other hand, consider an electron moving always in a straight line. They obtain, therefore

$$dE/dx = dE/vdt \tag{3}$$

where $\frac{1}{2}mv^2 = E$, forgetting that an electron will continually change its direction of motion.* This seems to be entirely unjustified.

It is the neglect by Seeger and Teller of the variation in direction which is responsible for the

[1] H. Fröhlich, Proc. Roy. Soc. **160**, 230 (1937).
[2] R. J. Seeger and E. Teller, Phys. Rev. **54**, 515 (1938).
[3] H. Fröhlich and N. F. Mott, Proc. Roy. Soc. (in press).

* The motion of an α-particle in a gas, to which Seeger and Teller refer, is entirely different, because of the much greater mass of an α-particle.

main differences in their and the author's final formulae for the breakdown field. Moreover, there seems to be also a mistake in the interaction which they take between an electron and an ion. The field at the distance r from the electron is $e/\kappa r^2$, and we may take the force, acting on an ion to be

$$e^2/\kappa r^2.$$

This neglects the Lorentz-Lorenz terms, which seems correct.[4] κ is the effective dielectric constant and will be defined more carefully below.

Now the following approximation is made in the calculation of Seeger and Teller. It is assumed that all ions at distances greater than $v/2\pi\nu$ (ν = lattice frequency) can follow the force acting on them adiabatically. At smaller distances, however, the interaction between electron and an ion is treated as an impulse of infinitely short duration. This then may lead to a loss of energy. According to this assumption only ions at distances greater than $v/2\pi\nu$ from the electron give a contribution to the polarization of the lattice by moving from their equilibrium positions, whereas inside this range, only the deformation of the ions may contribute. For the interaction of the electron with ions inside this range, therefore, κ must have values within the limits

$$1 \leqslant \kappa \leqslant \epsilon_0. \tag{4}$$

Here $(\epsilon_0 - 1)/4\pi$ is the polarizability of the ions.

Seeger and Teller's expression for κ, however, may be written in the form

$$\kappa = (\epsilon\epsilon_0)^{\frac{1}{2}},$$

making use of Born's lattice theory,[5] according to which

$$\epsilon - \epsilon_0 = e^2/2\pi a^3 M\nu^2. \tag{5}$$

Here, ϵ is the static dielectric constant, M is the resultant mass of the ions and a is the lattice distance. Since $\epsilon > \epsilon_0$, Seeger and Teller find $\kappa > \epsilon_0$ which is in contradiction with Eq. (4).

In the author's treatment any polarization would appear only in the second order of approximation. The first-order processes include only scattering and correspond to a value of κ equal to unity. It should be noted, however, that in contrast to a remark by Seeger and Teller, the interaction energy W between an electron and a dielectric medium is entirely correct in Fröhlich's treatment since W was given as

$$W = e\varphi$$

$$\nabla^2\varphi = 4\pi \operatorname{div} \mathbf{P}(r)$$

($\mathbf{P}$ = polarization of the dielectric). This is equivalent with

$$W = -e\int \frac{\operatorname{div} \mathbf{P}}{r} d\tau,$$

an expression which follows immediately from Maxwell's equations.

It is easy to see that in the case of fast electrons it is correct to assume that any possible screening of the electronic charge (i.e., a value of $\kappa > 1$) is an effect of a higher order. According to Fröhlich's treatment, the contribution of polarization waves of a wave number w to the time of relaxation τ is given by

$$\frac{1}{\tau} \sim \int_0^{w_0} w dw, \quad w_0 = 2^{\frac{1}{3}}\pi/a,$$

if the energy of the electron $E > \hbar^2 w_0^2/8m$. This means that the main contribution to the scattering is due to the ions in the immediate surrounding of the electron, where there can hardly be any screening. The effect on the energy loss dE/dt is greater since $dE/dt \sim \int dw/w$ which means that ions at greater distances contribute too. But even so the effect on the breakdown field is not greater that one would expect from an effect of a higher order (<30 percent).

For slower electrons one might expect a more important influence of a screening. For very slow (thermal) electrons, however, this effect should again be unimportant (as for fast electrons), as will be shown by Fröhlich and Mott.[3]

§3. The Condition for Breakdown

As we have seen, both authors assume that breakdown takes place when the field is so strong that all electrons with energy greater than a critical value E_c gain energy from the field more quickly than they lose it to lattice vibrations.

[4] Cf. N. F. Mott and M. J. Littleton, Trans. Faraday Soc. **34**, 485 (1938).

[5] Cf. M. Born and M. Göppert-Mayer, *Handbuch der Physik*, 2nd ed. 24/2, 623 (1933).

The condition for breakdown is thus

$$eF=(dE/dx)_{E=E_c}.$$

It may easily be seen that, as E is decreased, dE/dx rises to a maximum at a value of E in the neighborhood of several $h\nu$. Seeger and Teller assume with von Hippel[6] that E_c is equal to this value of E; thus *all* electrons in the conduction band will be accelerated by the field. Thus any electron in the conduction band will produce a secondary; the two electrons will be accelerated again so that finally an electron avalanche is built up.

In contrast to this it was considered by Fröhlich to be sufficient that electrons with an energy only a little below the ionization energy I should gain more energy from the field than they lose to the lattice. Thus the field would cause, it was argued, irreversible ionization processes so that no stationary state could exist. In the following we hope to go more deeply into the details of this question.

We shall discuss first the difficulties connected with the behavior of an electron gas in a constant external electric field. Assume a gas of N electrons which are in interaction with lattice vibrations. We shall assume that the interaction of the electrons with each other may be neglected and that N is so small that in the absence of a field the electrons are distributed over the energies according to the Maxwell distribution law. Now apply an external electrical field F. The electrons take up energy from the field in an irreversible way. To obtain a stationary state for the electrons in which a steady current flows, it must be possible for them to transfer this energy to the lattice. It has been shown previously (Fröhlich[1]) that in the case of an ionic lattice this is only possible for electrons with an energy $E<E'$, where the energy E' still depends on the field: $E'\sim 1/F$. For $E>E'$, the electrons will gain more energy from the field than they can transfer to the lattice. Thus, a stationary state is impossible unless a new kind of collision process is considered.

Now, for energies $E>I$ the distribution function is certainly determined rather by the collisions leading to ionization than by collisions with the lattice vibration, for the probability of ionization should be nearly 100 times bigger than the probability for scattering by the lattice vibrations. To obtain an equilibrium, we have, of course, to consider the inverse processes too, which means that as many electrons as leave the energy region $E>I$ by ionization, will come back by the reverse process. Now, as long as the field strength F is so small that $E'>I$ it seems to be possible to obtain a stationary state by means of the ionization processes. The energy which the electrons for which $E>E'$ obtain from the field must, of course, still finally go to the lattice. Such electrons spend, however, only a very short time in the energy region $E>E'$. As long as $E'>I$ they will, after the ionization, have an energy $E>E'-I>0$ and, therefore, there will be an opportunity to lose energy to the lattice.

Although an equilibrium is possible, it should be noted that the distribution function may be very different from a Maxwell distribution.

Now, if the field F is so strong that $E'<I$ an equilibrium should no longer be possible. For in this case, an electron with the energy E' will, on the average, gain the energy $I-E'$ from the field and then carry out an ionization process after which it will have an energy $E\sim 0$; thus it cannot transfer the energy $I-E'$ to the lattice.

It might be possible, of course, that the probability of finding an electron with an energy $E>E'$ is extremely small so that the deviation from a stationary state would not be serious. For $E'\sim I$ this would, in fact, be the case if an electron with the energy E' could be obtained only by thermal fluctuations, as Seeger and Teller suppose. Under the influence of an electrical field of about 10^6 volts/cm, however, there is a rather large chance for an electron to attain a high energy. Let $\phi(E)$ be the probability that an electron makes a collision per second. Then

$$W=\exp\left[-\int_0^t \phi(E)dt\right]$$

will be the probability that after t seconds it has *not* yet made a collision. During the whole time it is under the influence of the external field. It takes about $t=10^{-13}$ second to accelerate an electron from $E=0$ to $E=I$, i.e., to a velocity $v\sim 10^8$ cm/sec. The mean free path l of such electrons is $\sim 10^{-6}$ cm, for slow electrons ($v\sim 10^7$ cm/sec.) it is $\sim 10^{-7}$ cm. Thus $\phi=v/l\sim 10^{14}$ sec.$^{-1}$

[6] A. v. Hippel, Ergebn. exakt. Naturwiss. **14**, 79 (1935).

for all energies and $W \simeq e^{10} \sim 10^4$. It thus follows that it will take only about $W \cdot t \sim 10^{-9}$ second to bring an electron up to energies $E' \sim I$. This shows that in strong fields (but below the breakdown strength where a stationary state is still possible) the equilibrium distribution of electrons is such that a considerable fraction of electrons have energies of the order $E \sim I$.

It should be emphasized that by a direct acceleration of an electron, which moves in the direction of the field, an ionization process may be caused even at field strengths below breakdown. Since, however, the reverse process exists (i.e., as electrons may as well be retarded by the field) the field will, in a first approximation, not change the number of ionization processes since it accelerates as many electrons as it retards. Thus it has to be decided by the second approximation, i.e., by the energy transfer A, whether or not a net increase of ionization processes is caused by the field.

There is still a possibility of obtaining a stationary state if we consider the recombination of an electron. Very little is known about recombination in solids, but it seems to be rather certain that such a process will take much longer* than 10^{-9} sec. Therefore, even if in principal a stationary state may eventually be reached, this state would be entirely different from the state where $E' > I$.

* The cross section for recombination with emission of light is $<10^{-20}$ cm.

We shall now connect the above model with the actual case with which we have to deal in the breakdown problem. We note the experimental fact that for field strengths near the critical field of breakdown, but below it, there exists a stationary current in the dielectric medium. This means that in fact we deal with an electron gas in a stationary state. We shall not, at present, investigate theoretically how these electrons come into the conductive levels of the dielectric but accept it as an experimental fact. Then, it seems that the above considerations about the possibilities of reaching a stationary state may be applied.

The experimental evidence too seems to justify this condition. Hippel's condition (used by Seeger and Teller) would yield theoretical values for the breakdown field which are too high by a factor ~ 5 in the case of the alkali halides. The fact that Seeger and Teller obtained better values is due to a compensation by the different mistakes, discussed in §§2 and 3.

Finally I should like to mention in this connection that experiments by S. Whitehead, A. E. W. Austen and W. Hackett carried out at the laboratory of the British Electrical and Allied Industries Research Association (kindly communicated to the author before publication) have now confirmed most of the conclusions of the author's theory.*

* Cf. Austen and Hackett, Nature **143**, 637 (1939).

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Remarks on the Dielectric Breakdown

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IN the foregoing paper Fröhlich touches upon a number of questions that are of importance in the theory of breakdown of insulators. Since we do not find ourselves in agreement with his conclusions, we should like to clarify our point of view.

In the first place Fröhlich correctly remarks that in our treatment[1] of the interaction between an electron and a lattice the variation of direction in the motion of the electron has been neglected. Such variation of direction may, in fact, become very important either for electrons of high speed

[1] R. J. Seeger and E. Teller, Phys. Rev. **54**, 515 (1938).