PHYSICAL REVIEW VOLUME 79, NUMBER 3 AUGUST 1, 1950

Notes on the Theory of Dielectric Breakdown in Ionic Crystals

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(Received April 17, 1950)

A comparative discussion is given of the theories proposed by the authors on dielectric breakdown in ionic crystals.

In the literature bearing on the subject of dielectric breakdown in crystals we have expressed somewhat different viewpoints concerning the details of the process.[1–3] This note is written with the intention of expressing these viewpoints in a somewhat coordinated manner.

I.

We both agree that in the range of velocity in which a free electron does not have sufficient energy to stimulate excitation waves or to ionize the atoms of the lattice, so that interaction with the lattice vibrations is the principal means by which the electron dissipates its energy, the interaction energy contains three prominent terms.

(a) An interaction with a dipole field which is described by treating the ions as if rigid, that is by neglecting any polarization or distortion of the electron shells. This interaction was first discussed by Fröhlich. It extends over a long range and is very large for longitudinal polarization waves of long wave-length.

(b) An interaction with the dipole component of the field arising from the distortion of the electron cloud. This component produces a long-range effect, very similar to the interaction (a) for long polar waves. If combined with (a) it decreases the influence of the latter. In the case of long polarization waves, the interaction resulting from the sum of (a) and (b) is similar in mathematical form to that derived for (a) alone, with the ionic charge replaced by an effective charge e^* of the form introduced by Lyddane, Sachs, and Teller.[4]

(c) An additional "short-range" interaction associated with electron distortion which excludes the dipole component (b). This component is the only one which occurs in metals and non-polar insulators.[5]

Interactions (a) and (b) are largest for long polar waves and decrease with decreasing wave-length. Both are small for acoustical waves of long wave-length and increase with decreasing wave-length.

Interaction (c) is negligible compared with the sum of (a) and (b) for long polar waves. On the other hand, (a) and (b) are negligible compared with (c) for long acoustical waves.

Fröhlich has, by definition, called interaction (a) the polar interaction. Although (b) is closely related to (a) for long polar waves, since the most important part of the electron polarization arises from the long-range electrostatic field associated with ionic displacement, Fröhlich believes that this interrelation between (a) and (b) ceases to be highly significant when the waves become short. For this reason, he believes it to be reasonable to regard (b) and (c) as a unit for purposes of discussion. Moreover, he believes that the sum of (b) and (c) will be relatively small for short waves since each will have opposite sign and nearly equal magnitude. As a result, (a) is the dominant term for short waves and it becomes permissible, for short waves, to treat the scattering with the use of Fröhlich's original theory in which electron polarization is neglected.

Seitz has called, by definition, the sum of (a) and (b) the polar interaction, and has termed (c) the non-polar interaction. He has assumed that the sum of (a) and (b) can be extrapolated to the range of short waves with the use of Fröhlich's equation for interaction (a) taken in the form in which the ionic charge is replaced by e^*. Interaction (c) was estimated with the use of equations developed for non-polar materials, and was assumed to dominate in the range of short waves.

These two viewpoints are not inconsistent with one another. In fact, both seem to lead to the same total interaction in the range of short waves which are of interest for the scattering of electrons with an energy of the order of several electron volts, which have wave number vectors lying close to the edge of a Brillouin zone.

To summarize, we have made different intuitive approaches to estimating the interaction for short waves and in these approaches have used different definitions of the polar interaction. Both approaches seem to lead to essentially the same conclusion, namely that the collision frequency of the conduction electrons

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[1] H. Fröhlich, Proc. Roy. Soc. **A160**, 230 (1937); Reports Prog. Phys. **6**, 411 (1939).

[2] H. Fröhlich, Proc. Roy. Soc. **A188**, 521, 532 (1937).

[3] F. Seitz, Phys. Rev. **76**, 1376 (1949).

[4] Lyddane, Sachs, and Teller, Phys. Rev. **59**, 673 (1941).

[5] F. Seitz, Phys. Rev. **73**, 550 (1948).

is high both when the electron has an energy of the order of $\hbar\omega$, where ω is 2π times the frequency of the longitudinal polar waves of long wave-length, and when the electron has an energy in the vicinity of the Brillouin zone.

Perturbation theory may be applied to obtain quantitative results only if $E<\hbar\omega$, and leads to the formula derived by Fröhlich and Mott[6] (modified according to Fröhlich, Pelzer, and Zienau[7] and by Callen[8]). The formal procedure of perturbation theory also converges in the region of higher energies, that is, when $E\gg\hbar\omega$, if applied to the form of the polar matrix components used in the low energy range. It then leads to the result

$$1/\tau=(\epsilon_\infty^{-1}-\epsilon^{-1})(\omega/ka_0)(1+2n_\omega) \qquad (1)$$

for the collision frequency, in which n_ω is the average number of vibrational quanta associated with an oscillator having frequency ω at temperature T, ϵ is the dielectric constant, ϵ_∞ is the square of the refractive index, $2\pi/k$ is the de Broglie wave-length of the electron, and a_0 is the Bohr radius. This equation can be obtained from Eq. (6.6) of reference 7 by replacing $2kT/\hbar\omega$ by $1+2n_\omega$. For energies near a Brillouin zone this equation yields a value of the collision frequency of $2\cdot10^{13}$ sec.$^{-1}$ in the case of NaCl at thc absolute zero of temperature. In this case $\omega\simeq5\cdot10^{13}$ sec.$^{-1}$, $\epsilon\simeq5.6$, $\epsilon_\infty=2.3$. Since application of the perturbation theory requires that $1/\tau>\omega/2\pi$, this is about the lowest energy in the high energy region for which the foregoing equation may be used.

We both believe, however, that the actual value of the collision frequency is higher than that given by Eq. (1). From Fröhlich's viewpoint, an approximation to the true value is to be computed by taking interaction (a) alone into account. The result is obtained by replacing (1) by an expression having similar form, but a larger coefficient [see Eq. (22) of reference 5]. Seitz has replaced (1) by an expression derived from a treatment of non-polar materials which takes into account the interaction (c) alone.

In this connection, Seitz desires to correct an ambiguity which appears in his paper[3] in the section following Eq. (22). The value of the collision frequency for an electron having an energy near the Brillouin zone which is given, namely $1.5\cdot10^{13}$ sec.$^{-1}$, is not computed from Eq. (22) which is the equation Fröhlich would employ to estimate the total collision frequency, but is obtained from the collision frequency ($4.3\cdot10^{14}$ sec.$^{-1}$) for electrons having energy in the range near $\hbar\omega$, given in a preceding paragraph, by multiplying by a factor $k(\hbar\omega)/k_z$ where $k(\hbar\omega)$ is the wave number associated with an electron having an energy $\hbar\omega$ and k_z is the wave number of an electron near the edge of the Brillouin zone. This multiplicative factor is suggested by the energy dependence contained in Eq. (22).

II.

We both agree that the increase in electrical conductivity in strong fields (but below breakdown strength) arises from a strongly fluctuating increase in the number of free electrons, and may be described in terms of avalanche formation. Seitz has related breakdown to the formation of avalanches by assuming that breakdown occurs when these avalanches exceed a certain size. He has adopted as a criterion for breakdown the condition that a primary produce 40 progeny in traversing the specimen. It was assumed that these progeny would produce further free electrons, so that $2^{40}=10^{12}$ electrons would result. This was regarded as a reasonable upper limit for the number required to induce changes that might be regarded as constituting breakdown.

The actual value of the breakdown field will depend upon the number of electrons required to bring about the onset of breakdown through the logarithm of this number and hence will not be highly sensitive to it.

We agree that considerations of the above type would lead to an upper theoretical limit for the breakdown strength, but that breakdown may occur through other processes connected with the existence of an increased number of free electrons. Experimentally, breakdown would then appear not as a continuation of pre-breakdown currents, but rather as a phenomenon abruptly interrupting the increase of pre-breakdown currents. Experimental work in this field is somewhat ambiguous; however it appears that von Hippel[9] has found this to be the case in his experiments on alkali halides. Fröhlich's low temperature theory[1] is based on the assumption of such an effect; in the high temperature theory[2] it follows as a consequence of the basic assumptions.

III.

Finally, Seitz wishes to thank Professor A. Nordsieck for indicating an error in connection with Section 8 of reference 3, which deals with excitons. Nordsieck has pointed out that a consistent treatment of the excitons with the use of the hydrogen-atom model requires, in analogy with the treatment of electron-positron states, that one identify the first exciton state with total quantum number $n=1$ instead of $n=2$, as was done in reference 3. This increases the ionization energy of the excitons by a factor 4, that is, changes the energy for diamond and sodium chloride from 0.049 and 0.30 ev to 0.20 and 1.2 ev, respectively. The change in the value for diamond does not alter the discussion appreciably: One still expects the excitons to be dissociated by the field near breakdown. On the other hand, the larger value for NaCl is in much closer agreement with Mott's value of about 1.7 ev discussed in the paper and makes it seem very dubious that the excitons are decomposed by fields as weak as the breakdown value in the alkali halides.

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[8] H. Callen, Phys. Rev. **76**, 1394 (1949).

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