

# Theory of avalanche ionization induced in transparent dielectrics by an electromagnetic field

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A diffusion approximation of the quantum kinetic equation for conduction-band electrons is obtained and the region of its applicability is investigated. The main regularities of development of the electron avalanche are investigated on basis of the solution of the derived equation, and critical field values are determined by taking electron scattering by acoustic and optical phonons into account.

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## 1. INTRODUCTION

One of the possible mechanisms of damage to transparent solids by high-power laser pulses is the electron avalanche, a process most widely discussed of late. The interest in this mechanism is due primarily to the fact that the electron impact ionization is an inherent property of pure substances, and by the same token established the maximum endurance of materials to laser radiation. On the other hand, if the experimentally observed damage is indeed due to a sharp increase in the absorption of electromagnetic energy as a result of an avalanche-like increase of the carrier density, then it becomes possible to obtain, albeit indirectly, very valuable information on the dynamics of the electrons in the conduction band of dielectrics. To realize this possibility, and also to ascertain whether an electron avalanche is the dominant mechanism of laser damage, it is obviously necessary to have a consistent theory of this process, capable not only of giving numerical estimates of the damage threshold but also of helping to ascertain the main regularities, such as the dependence of the threshold on the electromagnetic field frequency  $\Omega$ , the pulse duration  $t_p$ , and the initial lattice temperature  $T$ . Such a theory should be based on the solution of the quantum kinetic equation.

For the case when the electromagnetic field frequency exceeds appreciably the frequency  $\nu = 1/\tau$  of the electron-phonon collisions ( $\tau$  is the relaxation time of the longitudinal component of the electron momentum),

$$\Omega \gg \nu, \quad (1)$$

and the multiphoton processes inside the conduction band are insignificant,

$$H_\infty = \frac{eE\Delta p}{\hbar m \Omega^2} \ll 1, \quad (2)$$

the problem of cascade ionization in solid transparent dielectrics was solved in<sup>[1]</sup>.<sup>1)</sup> The process of cascade ionization in a wide range of electromagnetic-field frequencies, from the visible band to constant electric fields, was considered in<sup>[2]</sup>, where the dependence of the breakdown threshold on frequency and temperature was analyzed with allowance for the scattering of electrons only by acoustic phonons.

The present paper is aimed at a complete analysis of the breakdown of transparent dielectrics by avalanche ionization, on the basis of a solution of the quantum kinetic equation for the electrons of the conduction band in the diffusion approximation.

In Sec. 2 we derive the diffusion equation cited in<sup>[2]</sup> and obtain general expressions for the connection between the avalanche development constant and the electric field strength.

In Sec. 3 we investigate the regions of applicability of the diffusion approximation. Next, in Secs. 4 and 5, we study the role of the optical phonons in cascade breakdown under the influence of an electromagnetic field. Finally, in the analysis of the results in Sec. 6, we discuss the dependence of the critical field on the duration of the electromagnetic radiation, a dependence typical of avalanche ionization, and discuss experiments that can determine the role of the considered mechanism in laser breakdown of optical materials.

## 2. DIFFUSION APPROXIMATION FOR THE QUANTUM KINETIC EQUATION

We start from the quantum kinetic equation

$$\frac{\partial f(\mathbf{p}, t)}{\partial t} = \frac{2\pi}{\hbar} \sum_{\mathbf{q}} B^2(\mathbf{q}) \sum_{n=-\infty}^{\infty} J_n^2 \left( \frac{eE\mathbf{q}\tau(\mathbf{p})}{\hbar m \Omega (1 + \Omega^2 \tau^2(\mathbf{p}))^{1/2}} \right) \{ [f(\mathbf{p}+\mathbf{q})(N_{\mathbf{q}}+1) - f(\mathbf{p})N_{\mathbf{q}}] \delta(\varepsilon(\mathbf{p}+\mathbf{q}) - \varepsilon(\mathbf{p}) - \hbar\omega_{\mathbf{q}} - n\hbar\Omega) + [f(\mathbf{p}+\mathbf{q})N_{\mathbf{q}} - f(\mathbf{p})(N_{\mathbf{q}}+1)] \delta(\varepsilon(\mathbf{p}+\mathbf{q}) - \varepsilon(\mathbf{p}) + \hbar\omega_{\mathbf{q}} - n\hbar\Omega) \}. \quad (3)$$

This equation is analogous to that derived in<sup>[4]</sup>, except that the arguments of the Bessel functions contain an additional factor  $\Omega\tau/(1 + \Omega^2\tau^2)^{1/2}$ , introduced to take into account the longitudinal component of the momentum resulting from electron-phonon collisions.

We use the following notation:  $e$  and  $m$  are the charge and mass of the electron,  $\mathbf{p}(\varepsilon)$  is the momentum of an electron with energy  $\varepsilon$ ,  $B(\mathbf{q})$  is the matrix element of the electron-phonon interaction,  $N_{\mathbf{q}}$  is the number of phonons with wave vector  $\mathbf{q}$ ,  $\hbar\omega_{\mathbf{q}}$  is the energy of the phonon,  $\tau_1(\varepsilon)$  is the relaxation time of the longitudinal momentum component of an electron with energy  $\varepsilon$  in the absence of a field. The relaxation time  $\tau(\varepsilon)$  is defined by the equation

$$\frac{p^2}{\tau(\mathbf{p})} = \frac{2\pi}{\hbar} \sum_q B^2(q) q \mathbf{p} \sum_{n=-\infty}^{\infty} J_n^2 \left( \frac{eE q \tau(\mathbf{p})}{\hbar m \Omega (1 + \Omega^2 \tau^2)^{1/2}} \right)$$

$$\times \{N_q \delta(\epsilon(\mathbf{p}-\mathbf{q}) - \epsilon(\mathbf{p}) - \hbar\omega_q - n\hbar\Omega) + (N_q + 1) \delta(\epsilon(\mathbf{p}-\mathbf{q}) - \epsilon(\mathbf{p}) + \hbar\omega_q - n\hbar\Omega)\} \quad (4)$$

We assume, bearing in mind a derivation of the diffusion approximation, that

$$\tau(\mathbf{p}+\mathbf{q}) \approx \tau(\mathbf{p}).$$

The distribution function  $f(\mathbf{p})$  can be obtained, generally speaking, only by simultaneously solving Eqs. (3) and (4) as well as the corresponding equation for the phonons. To be able to disregard the latter, we assume an equilibrium phonon spectrum, an assumption justified in most cases because of the high frequencies of the phonon-phonon collisions at a lattice temperature 50°K and higher.

We seek the solution of the kinetic equation in the so-called diffusion approximation. To this end, we must make the following assumptions:

1. The anisotropic scattering described by Eq. (3) is replaced by some isotropic scattering:

$$J_n^2 \left( \frac{eE q \tau}{\hbar m \Omega (1 + \Omega^2 \tau^2)^{1/2}} \right) \rightarrow \int_0^1 J_n^2 \left( \frac{eE q \tau}{\hbar m \Omega (1 + \Omega^2 \tau^2)^{1/2}} x \right) dx.$$

This assumption seems to be equivalent to assuming smallness of the asymmetrical part of the distribution function in comparison with the symmetrical part.

2. The symmetrical part of the distribution function is expanded in the form

$$\tilde{f}(\epsilon + \Delta\epsilon) \approx \tilde{f}(\epsilon) + \Delta\epsilon \frac{\partial \tilde{f}(\epsilon)}{\partial \epsilon} + \frac{1}{2} (\Delta\epsilon)^2 \frac{\partial^2 \tilde{f}(\epsilon)}{\partial \epsilon^2},$$

where

$$\Delta\epsilon = \pm n\hbar\Omega \pm \hbar\omega_q.$$

It is assumed here, obviously, that

$$|\Delta\epsilon \partial \tilde{f}(\epsilon) / \partial \epsilon| \ll \tilde{f}(\epsilon). \quad (5)$$

It is clear that this inequality cannot be satisfied for all energies and parameters of the  $n$  quanta. For the error due to this assumption to be negligible, it is necessary to satisfy the condition

$$n_{\max} \hbar\Omega |\partial \tilde{f}(\epsilon) / \partial \epsilon| \ll \tilde{f}(\epsilon), \quad (6)$$

where  $n_{\max}$  is the maximum number of quanta whose probability of simultaneous absorption remains significant. Relation (6) will be analyzed in Sec. 3.

Changing in (3) and (4) to integration with respect to  $q$  and using the summation formulas

$$\sum_n J_n^2(x) = 1, \quad \sum_n n^2 J_n^2(x) = \frac{x^2}{2},$$

we obtain an equation in the form

$$-\frac{\partial}{\partial \epsilon} \mathcal{S}(\epsilon, t) = \tilde{g}(\epsilon) \frac{\partial \tilde{f}(\epsilon, t)}{\partial t},$$

$$\mathcal{S}(\epsilon, t) = -\tilde{g}(\epsilon) \left[ \tilde{D}(\epsilon) \frac{\partial \tilde{f}(\epsilon, t)}{\partial \epsilon} + \tilde{Q}(\epsilon) \tilde{f}(\epsilon, t) \right] \quad (7)$$

with the following coefficients:

$$\begin{aligned} \tilde{Q}(\epsilon) &= \tilde{Q}^0(\epsilon), \quad \tilde{D}(\epsilon) = \tilde{D}_E(\epsilon) + \tilde{D}^0(\epsilon), \\ \tilde{D}_E(\epsilon) &= \frac{e^2 E^2 p^2(\epsilon) \tau(\epsilon)}{6m^2 (1 + \Omega^2 \tau^2)}, \end{aligned} \quad (8)$$

where  $\tilde{D}^0(\epsilon)$  and  $\tilde{Q}^0(\epsilon)$  are respectively the diffusion coefficient and the power loss in the absence of an electromagnetic field, and  $\tilde{g}(\epsilon)$  is the density of the number of states.

We note that under our conditions Eq. (4), which is in fact an equation defining  $\tau_1$  i.e., the relaxation time of the longitudinal component of the electron momentum, remains unchanged in the diffusion approximation when the electromagnetic field is turned on. The derived Eq. (7) with coefficients (8) goes over in the limit as  $\Omega \rightarrow 0$  into the known equation for the symmetrical part of the distribution function in a constant electric field.<sup>[15]</sup>

We shall use henceforth the dimensionless variable  $x = \epsilon/I$  ( $I$  is the ionization potential) and the coefficients of the kinetic equations  $\mathcal{D}(x)$  and  $\mathcal{Q}(x)$  without the tilde, which have the dimensionality  $\text{sec}^{-1}$ :

$$\mathcal{D}(x) = \tilde{D}(\epsilon)/I^2, \quad \mathcal{Q}(x) = \tilde{Q}(\epsilon)/I.$$

To solve the obtained equation we use the successive-approximation method developed in<sup>[1]</sup>. According to that paper we have in first order in the ratio  $\gamma/Q$  the following expression for the avalanche-development constant:

$$\gamma_1 = Q(1) g(1) f_0(1) / \int_0^1 g(x) f_0(x) dx, \quad (9)$$

$f_0$  is the stationary distribution function. Numerical calculations show that up to values  $\gamma \sim 0.2Q(1)$  we have

$$(\gamma_2 - \gamma_1) / \gamma_1 < 0.3,$$

i.e., the first approximation is quite adequate if  $\gamma < 10^{11} \text{sec}^{-1}$ . We shall use (9) subsequently to obtain concrete expressions for the critical field in Secs. 4 and 5.

### 3. REGION OF APPLICABILITY OF THE DIFFUSION APPROXIMATION

In fields close to critical, the following relation holds:

$$D_E \gg D_0.$$

In the quasi-stationary approximation ( $\gamma \ll Q$ ) the function  $\tilde{f}(\epsilon)$  and its derivative  $\tilde{f}'(\epsilon)$  are connected, according to (7), by the following approximate relation:

$$\tilde{f}'(\epsilon) \approx -\frac{6\hbar\omega_0 (1 + \Omega^2 \tau^2)}{e^2 E^2 p^2 (2N_0 + 1)} \tilde{f},$$

where  $\hbar\omega_0$  is the average energy of the phonons that take

part in the absorption of the electromagnetic quanta by electrons. (We have used the known expressions for  $Q^0$  and  $D^0$ ; see, e.g., [5,8].)

Since the Bessel function  $J_n^2(x)$  decreases exponentially with increasing  $n$  in the region  $n > x$ , it follows that

$$n_{\max} \sim eE/\hbar\Omega(1+\Omega^2\tau^2)^{1/2}$$

and the condition (6) takes the form

$$eE/(1+\Omega^2\tau^2)^{1/2} \gg \hbar\omega_0/(2N_0+1) \sim \hbar\omega_0. \quad (10)$$

In scattering of electrons by acoustic phonons, the concept of "average phonon energy" can be made more precise, as a result of which we obtain

$$eE/(1+\Omega^2\tau^2)^{1/2} \gg mv_s^2 I/kT, \quad (10a)$$

$v_s$  is the speed of sound.

As will be shown below, the critical field at a fixed pulse duration has (apart from small corrections) the following dependence on the frequency of the electromagnetic field:

$$E_{cr} \propto (1+\Omega^2\tau^2)^{1/2}. \quad (11)$$

Comparison of (10) with (11) shows that in fields close to critical the condition for the applicability of the diffusion approximation is satisfied in the entire frequency region if it is satisfied at any one frequency. Since, as shown in [7], the diffusion approximation for the electrons of the conduction band of dielectrics can be used in constant fields, the presented analysis shows that it can be used also in the study of the breakdown of optical materials under the influence of laser radiation. We note that at first glance it appears that the diffusion equation may not hold if the energy of the light quantum is of the same order of or larger than the average electron energy  $\langle \epsilon \rangle$ . This statement, which to all appearances is valid when many processes connected with the dynamics of electrons that are not too hot in the conduction band (e.g., transport processes), becomes meaningless when it comes to consideration of an electron avalanche. The avalanche-development constant  $\gamma$  is determined in this case by the ratio of the number of electrons capable of effecting ionization to the total number of electrons in the conduction band. It is therefore important only that the approximate electron energy distribution function  $f(\epsilon)$  be close to the real one in the greater part of the range of energies lower than the ionization energy  $I$ . From this requirement we obtain directly the second basic condition for the applicability of the diffusion approximation:

$$\hbar\Omega \ll I. \quad (12)$$

In spite of the fact that in the electron energy region  $\epsilon \lesssim \hbar\Omega$  the number of the electrons can be large if

$$\hbar\Omega \sim \langle \epsilon \rangle,$$

the error in the calculation of the cascade development

constant by the method of [1] will be contained, if condition (10) is satisfied, only in the pre-exponential factor, which is of little importance.

An additional limitation can arise at high energies as a result of avalanche development due to single absorption of  $n_0$  electromagnetic-radiation quanta, so that

$$n_0 \hbar\Omega \approx I - \langle \epsilon \rangle.$$

The avalanche-development constant  $\gamma'$  in such a process is estimated from the formulas of [6]. Comparison of  $\gamma$  and  $\gamma'$  shows that for transparent dielectrics, in fields close to critical, the electron avalanche described by (7) is more effective if

$$n_0 > 5,$$

and less effective if

$$n_0 < 4.$$

In the intermediate case  $n_0 = 4-5$  it is necessary to carry out a comparison in each concrete case.

#### 4. DEVELOPMENT OF ELECTRON AVALANCHE IN SCATTERING BY ACOUSTIC AND NONPOLAR OPTICAL PHONONS

In scattering by acoustic and nonpolar optical phonons in the absence of an electromagnetic field, the coefficients of the kinetic equation (7) take the form

$$\begin{aligned} D_{ac}^0(x) &= \frac{4v_s^2 m}{(2mI)^{1/2} l_{ac}} x^{1/2}, \\ Q_{ac}^0(x) &= \frac{2v_s^2 (2mI)^{1/2}}{kT l_{ac}} x^{1/2}, \\ D_{op}^0(x) &= \frac{(\hbar\omega_{op})^2}{l_{op}(2mI)^{1/2}} x^{1/2}, \\ Q_{op}^0(x) &= \frac{2\hbar\omega_{op}}{(2N_{op}+1)l_{op}(2mI)^{1/2}} x^{1/2}. \end{aligned}$$

The quantities pertaining to scattering by acoustic, polar, and nonpolar optical phonons are labeled by the subscripts ac, po, and op, respectively:  $\omega_{op}$  is the frequency of the optical phonon,  $l_{ac}$  and  $l_{op}$  are the electron mean free paths in scattering by acoustic and nonpolar optical phonons, respectively.

We introduce the additional notation

$$\begin{aligned} l^{-1} &= l_{op}^{-1} + l_{ac}^{-1}, \quad \eta = 2I/mI^2(I)\Omega^2, \\ q_{ac} &= \frac{e^2 E^2}{6m^2 v_s^2 \Omega^2}, \quad q_{op} = \frac{2e^2 E^2 I}{3m(\hbar\omega_{op})^2 \Omega^2}, \\ \delta_{ac} &= \frac{kT}{I}, \quad \delta_{op} = \frac{\hbar\omega_{op}(2N_{op}+1)}{2I}, \\ Q &= Q_{ac}(1) + Q_{op}(1), \quad s = Q_{ac}(1)/Q. \end{aligned}$$

Equation (3) then takes the form

$$\begin{aligned} \frac{\partial}{\partial x} \left\{ \left( \frac{[q_{ac}\delta_{ac}s + q_{op}\delta_{op}(1-s)]x^2}{1+\eta x} + \delta_{ac}s x^2 + (1-s)\delta_{op}x \right) f'(x) \right. \\ \left. + (s x^2 + (1-s)x) f(x) \right\} = \frac{\gamma}{Q} f(x), \quad f(x, t) = f(x) e^{\gamma t}. \end{aligned}$$

(We use the high-temperature approximation for scat-

tering by acoustic phonons.) The solution of the equation is determined by relation (9). To simplify the form of the subsequent expressions, we note that in all realistic cases the following conditions are satisfied:

$$q \gg \eta, q \gg 1.$$

For the avalanche-development constant we obtain

$$\gamma = \Theta^{-1} \exp \left\{ -\frac{\eta s}{2q\delta} - \frac{[\eta(1-s)+s]}{q\delta} - \frac{(1-s)}{q\delta} \ln [1+q\delta/(1-s)\delta_{op}] \right\},$$

where (13)

$$q\delta = q_{op}(1-s)\delta_{op} + q_{ac}s\delta_{ac},$$

$$\Theta = Q^{-1} \int_0^1 dx \sqrt{x} \exp \left\{ -\frac{\eta s x^2}{2q\delta} - \frac{[\eta(1-s)+s]x}{q\delta} \right\} \left[ 1 + \frac{q\delta x}{\delta_{op}(1-s)} \right]^{-(1-s)/q\delta} \quad (14)$$

In many cases, the integral of (14) admits of simple estimates. Thus, if

$$\frac{[\eta(1-s)+s]\delta_{op}(1-s)}{(q\delta)^2} \ll 1, \quad (15)$$

then

$$\Theta = Q^{-1} \frac{\delta_{op}^{3/2}(1-s)^{3/2} \Gamma(3/2) \Gamma((1-s)/q\delta - 3/2)}{(q\delta)^{3/2} \Gamma((1-s)/q\delta)}, \quad \frac{1-s}{q\delta} > \frac{3}{2}, \quad (14a)$$

$$\Theta = Q^{-1} \Gamma\left(\frac{3}{2}\right) \left(\frac{q\delta}{\eta(1-s)+s}\right)^{3/2}, \quad \frac{1-s}{q\delta} < \frac{3}{2}, \quad \frac{\eta s q\delta}{(\eta(1-s)+s)^2} \ll 1, \quad (14b)$$

$$\Theta = Q^{-1} \frac{\Gamma(3/4)}{2} \left(\frac{2q\delta}{\eta s}\right)^{3/4}, \quad \frac{1-s}{q\delta} < \frac{3}{2}, \quad \frac{\eta s q\delta}{(\eta(1-s)+s)^2} \gg 1. \quad (14c)$$

If  $s=1$ , then

$$\Theta = Q^{-1} \left(\frac{q\delta}{\eta}\right)^{3/2} \Gamma\left(\frac{3}{2}\right) \exp\left(\frac{1}{4\eta\delta q}\right) D^{-3/4} \left(\frac{1}{\eta\delta q}\right)^{3/4}. \quad (16)$$

The inequality (15) is obviously satisfied at  $s=0$  and  $s=1$ , so that we can expect it to be valid also in the intermediate cases.

To obtain an expression for the critical field we must specify a criterion for the breakdown. We use the criterion obtained in<sup>[1]</sup>:

$$\gamma t_p = L. \quad (17)$$

If  $\Omega\tau \ll 1$ , then

$$L = \ln \left\{ 1 + \frac{4\gamma}{3\beta_0 n_0} (T_{cr}^n - T_0^n) \right\}, \quad (17a)$$

$$\beta_0 = \frac{QI \Gamma(3/2)}{C\rho \Gamma(3/4)} \left(\frac{2q\delta}{\eta T_0}\right)^{3/4};$$

if  $\Omega\tau \gg 1$ , then

$$L = \ln \left\{ 1 + \frac{\gamma}{\beta_\infty n_0} \frac{1}{T_0^{3/4}} \right\}, \quad \beta_\infty = \frac{2QI}{\sqrt{\pi} C\rho} \left(\frac{q\delta}{T_0}\right)^{3/4}. \quad (17b)$$

Here  $C$  is the lattice specific heat,  $\rho$  is the density,  $n_0$  is the initial number of electrons per unit volume,  $T_0$  is the initial lattice temperature, and  $T_{cr}$  is the critical temperature at which damage sets in. Strictly speaking, it is necessary to take into account also the  $\gamma(T)$  dependence, then  $L$  is decreased somewhat in compari-

son with the values calculated by formulas (17), and the dependence of  $L$  on  $T_{cr}$  becomes even weaker. We note also that for dielectrics in the case of breakdown in nanosecond pulses, the value of  $L$  ranges from 13 to 20 and has a negligible effect on the critical field, so that in practical calculations we can approximately choose as the breakdown criterion the relation

$$\gamma t_p = 15. \quad (17c)$$

From (13), using (17), we obtain formulas for the critical field in the case of scattering by acoustic phonons only:

$$E_{cr}^2 = \Lambda \frac{Im^2 v_s^2}{2kTe^2} \left( \Omega^2 + \frac{I}{ml_{ac}^2} \right), \quad (18)$$

$$\Lambda^{-1} = \frac{1}{12} \ln \left( \frac{t_p}{L\Theta} \right) \approx 1.$$

As the first example, we consider an electron avalanche in an NaCl crystal. We assume the following parameters: the electron-phonon coupling constant<sup>[8]</sup>  $\xi_1 = 10$  eV,  $D = 10^{-3}$  erg/cm,  $T_0 = 300$  °K,  $\hbar\omega_{op} = 3.7 \times 10^{-2}$  eV. Then

$$s = 0.97; \quad q_{op}/q_{ac} \approx 2.3.$$

We can use the asymptotic form (14b) at the frequency corresponding to the neodymium laser ( $\Omega \approx 2 \times 10^{15}$  rad/sec), formula (16) at the CO<sub>2</sub> laser frequency, and the asymptotic form (14c) at  $\Omega = 0$ . Allowance for the non-polar scattering by optical phonons changes the critical field insignificantly in comparison with the value calculated from (18):

$$E_{cr} = 0.97 E_{cr \text{ ac}}, \quad \Omega = 0,$$

$$E_{cr} = 1.05 E_{cr \text{ ac}}, \quad \Omega = 2 \cdot 10^{15} \text{ rad/sec}$$

where  $E_{cr \text{ ac}}$  is the corresponding threshold obtained when account is taken of scattering only by acoustic phonons.

The energy of optical phonons is much lower, e.g., in RbI ( $\sim 1.3 \times 10^{-2}$  eV). In this case we have

$$q_{op}/q_{ac} \approx 17$$

and the critical field is given by  $E_{cr} = 0.8 E_{cr \text{ ac}}$ .

The value  $D = 10^{-3}$  erg/cm is a very crude estimate. At best it is of the right order of magnitude. Our results, however, depend very strongly on  $D$ . If we assume a value of  $D$  only three times larger than above, then, for example, for the breakdown of RbI we obtain

$$E_{cr} = 0.44 E_{cr \text{ ac}}.$$

It is clear from the foregoing analysis that in this case, if experiment confirms that the cause of damage by high-power laser radiation is an electron avalanche, then comparison of experiment with the present theory will make it possible to estimate such an interesting quantity as the electron-phonon coupling constant  $D$  for deformation scattering by optical lattice vibrations.

## 5. EFFECT OF OPTICAL POLAR PHONONS ON THE DAMAGE THRESHOLD

The kinetic equation that takes into account scattering by both acoustic and polar optical phonons is very complicated. Its solution is determined by formula (9), but in general it is impossible to obtain concrete estimates without resorting to numerical calculations. We confine ourselves therefore to one particular case, which incidentally corresponds to many real situations.

We estimate the ratio  $Q_{po}/Q_{ac}$  (the subscript po will henceforth label quantities pertaining to polar scattering) for NaCl, i.e., for one of those crystals where this ratio is the largest as a result of the high energy of the optical phonon and the large difference  $\nu_{\omega}^{-1} - \nu_0^{-1}$  (the dielectric constants at  $\Omega = 10^{14} - 10^{15} \text{ sec}^{-1}$  and  $\Omega = 0$ ). At an electron energy equal to the ionization energy we have  $Q_{po}/Q_{ac} \approx 10^{-2}$ , and this ratio becomes of the order of unity at  $\epsilon < 1 \text{ eV}$ . On the other hand, the energy acquired during the average time between the collisions with the phonons  $\Delta\epsilon$  is of the order of  $eEl$ , which is of the order of 0.5 eV even at the minimal threshold fields. It is clear from the results of Sec. 2 that from the quantum point of view this means that the multi-photon processes that cause the electron to acquire an energy  $\Delta\epsilon$  have a high probability. All this seems to indicate that the additional energy losses connected with the scattering of the electrons by the polar optical phonons play a negligible role. We shall consider therefore the case when such corrections can be neglected, and take into account only that kinetic-equation term which is connected with scattering by polar optical phonons and enters in  $D_E$ .

In analogy with the procedure used in Sec. 4, we obtain the following expression for the critical field:

$$E_{cr}^2 = \Lambda \frac{Im^2 v_s^2}{2kT\epsilon^2} f_1(\alpha) \left\{ \Omega^2 + \frac{I}{ml^2(1)} \frac{f_2(\alpha)}{f_1(\alpha)} \right\}.$$

Here

$$f_1(\alpha) = 1 - \alpha \ln \left( \frac{1+\alpha}{\alpha} \right), \quad f_2(\alpha) = 1 - 2\alpha + 2\alpha^2 \ln \left( \frac{1+\alpha}{\alpha} \right),$$

$$\alpha = \frac{1-s}{s} \left( \frac{q_{po}\delta_{po}}{q_{ac}\delta_{ac}} \right), \quad s = \frac{Q_{ac}(1)}{Q_{po}(1) + Q_{ac}(1)}, \quad q_{op} = q_{po}.$$

The value of  $\Lambda$  can be estimated from the formulas of the preceding section. Since

$$0 < f_1(\alpha) < 1, \quad f_2(\alpha)/f_1(\alpha) > 1,$$

it follows that the critical field decreases somewhat in the entire range of electromagnetic-field frequencies, but this change itself is more pronounced at high frequencies and depends quantitatively on the value of  $\alpha$ .

By way of examples, we consider cascade ionization in NaCl and in sapphire. For NaCl we have  $\alpha = 0.016$  and

$$E_{cr}^2 = 0.93\Lambda \frac{Im^2 v_s^2}{2kT\epsilon^2} \{ \Omega^2 + 1.03I/ml^2 \},$$

i.e., the critical field is practically unchanged in com-

parison with the case when scattering by only acoustic phonons is taken into account. An analogous result is obtained also for RbI crystal (in contrast to scattering by nonpolar optical phonons). The critical field in polar scattering does not depend on the energy of the optical phonon in the high-temperature limit.

In sapphire, the speed of sound is large, and this leads to a certain lowering of the critical field as a result of polar scattering:

$$f_1(\alpha) \approx 0.8, \quad f_2/f_1 \approx 1.1.$$

## 6. DISCUSSION OF RESULTS

Our theoretical investigation of avalanche ionization in solid transparent dielectrics has explained the main regularities of this process, and these can serve as a basis for an experimental determination of its role in laser breakdown of optical materials. Notice must be taken, above all, of the essentially different temperature dependence of the critical field in those cases when the field frequency is higher or lower than the electron-phonon collision frequency. This fact is discussed in<sup>[2]</sup>. For the sake of completeness we present here an expression for the critical field in the case of scattering by acoustic zero-point vibrations only:

$$E_{cr}^2 = \Lambda_s \frac{mv_s(2mI)^{1/2}}{2e^2} \left( \Omega^2 + \frac{2I}{5ml_{ac}^2} \right),$$

where

$$\Lambda_s^{-1} = \frac{1}{15} \ln \left( \frac{t_p}{L\Theta} \right) \approx 1,$$

$$\Theta = Q^{-1} 4q^3 \delta_0^3, \quad \Omega\tau \gg 1,$$

$$\Theta = Q^{-1} \frac{2}{5} \Gamma \left( \frac{3}{5} \right) \left( \frac{q\delta_0}{\eta_0} \right)^{3/5}, \quad \Omega\tau \ll 1,$$

$$\delta_0 = 4mv_s/5(2mI)^{1/2}.$$

A second essential feature of damage due to the development of an electron avalanche is the very weak dependence of the critical field on the pulse duration, as follows from (17) and (18). Unfortunately, there are at present practically no data on the dependence of  $E_{cr}$  on the pulse duration in a wide range of the latter. An exception is<sup>[9]</sup>, where it is indicated that the dependence on the pulse duration is much stronger than predicted by the theory presented here.

If we do not assume anomalously short electron mean free paths, then the theoretical dependence of the critical field on the frequency differs from the experimental one. It is interesting to note, however, that the maximum increase of the threshold with increasing field frequency was obtained in precisely such crystals as RbI, for which one can expect such a behavior, according to Sec. 4, in the case when electron cascade is responsible for the damage.

Thus, even though it is impossible at present to draw final conclusions concerning the role of the electron cascade as the laser-damage mechanism, our results make it possible to organize task-oriented experiments for this purpose. If the dominant role of this damage mechanism is experimentally demonstrated, it will become possible to estimate a number of interesting

parameters, such as the mean free path, the electron-phonon coupling constant, etc.

<sup>1)</sup>The diffusion equation was solved for the first time for laser breakdown of solids in<sup>[3]</sup>, but no account was taken there of the energy lost in electron-phonon collisions, which can not be neglected, as shown in<sup>[1]</sup>, for the considered range of pulse durations ( $10^{-11}$ – $10^{-7}$  sec).

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## Weakly bound excitation states in a crystal

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It is shown that the formation in crystals of bound states by two elementary excitations (two phonons, a phonon and an electron) is possible for an arbitrarily weak interaction between them. This occurs near points in the quasi-momentum space of the excitations at which the two effective masses for the relative motion of the excitations become infinite. The mathematical situation here is similar to the situation that obtains in superconductors during the formation of Cooper pairs. The binding energy turns out to be exponentially small in the coupling constant.

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### 1. INTRODUCTION

It will be shown in this paper that the formation in crystals of bound states by two elementary excitations (two phonons, a phonon and an electron) interacting arbitrarily weakly with each other is possible. This occurs near certain special points in the quasi-momentum space of the excitations.

The question of the formation of bound states by excitations has attracted considerable attention in recent years. In Wortis's paper<sup>[1]</sup> two-magnon bound states were investigated. Cohen and Ruvalds,<sup>[2]</sup> Ruvalds and Zawadowsky,<sup>[3]</sup> and Agranovich<sup>[4]</sup> have studied the bound states of phonons. In the cases considered by these authors, however, in order for the formation of bound states to be possible, it is necessary that the phonon interaction energy exceed some threshold value. Another situation obtains, as is well known, in liquid He<sup>4</sup>. There, two rotons with an arbitrarily weak attraction between them can form a bound state.<sup>[5–7]</sup> This is explained by the fact that the energy of the roton as a function of its momentum has a minimum on a whole sphere in momentum space. Thus, this phenomenon is closely connected with the isotropy of the liquid. Kozhushner<sup>[8]</sup> has discovered that two excitons form a bound state when they interact arbitrarily weakly with each other in a special model in which only the interaction with the nearest neighbors is taken into account.

Some examples of bound-state formation in crystals under conditions when the interaction is weak have been considered by Rashba and Levinson.<sup>[9,10]</sup> These authors were, however, concerned either with phenomena that occur when the optical-phonon dispersion is neglected or with electrons in a magnetic field, when the situation becomes one-dimensional. Meanwhile, as we shall see, the formation of weakly-bound excitation states is possible in crystals with the most general properties and, thus, should be the rule, rather than the exception.

To see this, let us consider the simpler case when two identical excitations form a bound state. (Phonons belonging to one and the same branch.) Let the dispersion law for the excitations forming the bound state have the form  $\omega(\mathbf{k})$ , where  $\omega$  is the excitation energy and  $\mathbf{k}$  is the quasi momentum. Henceforth, instead of quasi momentum, we shall always speak of momentum. In this case the question determining the bound-state energy has, as will be shown in the following section of the paper, the form

$$\lambda \int \frac{d^3q / (2\pi)^3}{\epsilon - \omega(\mathbf{p}/2 - \mathbf{q}) - \omega(\mathbf{p}/2 + \mathbf{q}) + i0} = 1. \quad (1)$$

Here  $\lambda$  has the meaning of an effective coupling constant,  $\epsilon$  is the bound-state energy, and  $\mathbf{p}$  is the bound-state momentum. The function  $\epsilon(\mathbf{p})$  determined by this equation is the dispersion law for this bound state.