Excitation of Cascade Ionization in Transparent Dielectrics by a Strong Alternating Electromagnetic Field

P. M. MEDNIS AND V. M. FAÍN

Institute of Solid State Physics, USSR Academy of Sciences

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We analyze the possibility of formation of cascade ionization in a transparent dielectric as a result of a series of single-photon transitions of electrons in the conduction band. The following alternate mechanisms of cascade-ionization excitation are proposed: 1) as a result of a multiphoton transition of the electron into a state with energy higher than the ionization energy, and 2) as a result of electron-electron collisions with simultaneous multiphoton transition. Comparative estimates of the different mechanisms are presented.

1. INTRODUCTION

CASCADE ionization is one of the main mechanisms determining the breakdown of transparent dielectrics under the influence of a powerful optical wave from a laser (see, for example[1-3]). The development of cascade ionization under the influence of an electromagnetic field can be visualized in the following fashion. The "priming" electron in the conduction band of the dielectric acquires, as a result of the action of the light wave, an energy higher than the ionization energy I, and knocks an electron from the valence band out into the conduction band. As a result there are two electrons in the conduction band, which again acquire an energy I under the influence of the light wave, etc. To estimate the breakdown field produced after the time τ of the action of the light pulse, we can use the "40-ionization" criterion[4]

$$\gamma\tau = 40, \tag{1}$$

where γ^{-1} is the average time necessary to acquire an energy I in the field of the light wave. (The "priming" electron and the electrons knocked out from the valence band are assumed to be "cold.")

Molchanov[5] proposed that an electron can acquire energy in the field of a light wave as a result of a series of collisions with photons and acoustic phonons (the latter do not influence the energy balance but take up the excess momentum). He did not take into account the intraband scattering of the electrons by the acoustic phonons. Allowance for this process can greatly increase the threshold of the cascade ionization by means of an alternating field. As we shall show, an important role is played here by multiphoton processes in the conduction band.

We propose in this paper alternate mechanisms for the excitation of cascade ionization via multiphoton transition of a low-energy electron into a state with energy $I \approx n\hbar\omega$, where $\hbar\omega$ is the energy of the field quantum and n is the order of the multiphoton process. In Sec. 2 we calculate the probability of the multiphoton transition with participation of phonons, and in Sec. 3 we determine the probability of the electron-electron collision with simultaneous n-photon transition leading to impact ionization. In Sec. 4 we consider the influence of intraband scattering by acoustic phonons on the process whereby an electron acquires energy by a series of single-photon transitions, and obtain comparative estimates of the different mechanisms.

2. MULTIPHOTON TRANSITIONS IN THE CONDUCTION BAND WITH PARTICIPATION OF PHONONS

In the calculation of the multiphoton transition probability in the conduction band, we start from the Hamiltonian

$$H = \sum_{\mathbf{p}} \varepsilon\left(\mathbf{p} - \frac{e}{c}\mathbf{A}(t)\right) a_{\mathbf{p}}^{+} a_{\mathbf{p}} + \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}} b_{\mathbf{k}}^{+} b_{\mathbf{k}} + \sum_{\mathbf{pk}} C_{\mathbf{k}} a_{\mathbf{p}+\mathbf{k}}^{+} a_{\mathbf{p}} (b_{\mathbf{k}} + b_{-\mathbf{k}}^{+}), \tag{2}$$

where $\epsilon(\mathbf{p})$ is the energy of an electron with momentum $\mathbf{p}$, $\hbar\omega_{\mathbf{k}}$ is the energy of a phonon with momentum $\mathbf{k}$, $C_{\mathbf{k}}$ is the electron-phonon interaction constant (we shall henceforth have in mind interaction with acoustic phonons), $a_{\mathbf{p}}^{+}$ and $b_{\mathbf{k}}^{+}$ are the production operators of the electrons and phonons, respectively, and the electric field is equal to

$$\mathbf{E} = -c^{-1}\partial\mathbf{A}/\partial t = \mathbf{E}_0 \cos\omega t. \tag{3}$$

Having in mind the case of a weak electron-phonon interaction, we shall calculate the transition probability between electronic states accurate to terms quadratic in the energy of the electron-phonon interaction (the third term in (2)), without imposing any limitations on the magnitude of the electric field, except that it be small compared with the atomic field, so as to be able to neglect the interband transitions. Thus, the unperturbed Hamiltonian $H_0(t)$ (the first two terms in (2)) is explicitly dependent on the time. In this case the transition probability per unit time, averaged over a time much larger than the period of the radiation field, can be written in the form

$$w_{\mathbf{p}_0 \to \mathbf{p}} = \frac{2}{\hbar^2}\left\langle \operatorname{Re} \lim_{\delta \to +0} \int_{-\infty}^{t} e^{\delta t_1} V_{\mathbf{p}_0\mathbf{p}}(t_1)\, V_{\mathbf{p}\mathbf{p}_0}(t)\, dt_1 \right\rangle, \tag{4}$$

where the angle brackets denote averaging over the time, V is the operator of the interaction energy (the third term in (2)), and the dependence of the matrix elements on the time is determined by a unitary operator satisfying the equation

$$i\hbar\partial S/\partial t = H_0(t)S; \quad i\hbar\partial S^{-1}/\partial t = -S^{-1}H_0(t). \tag{5}$$

It follows therefore that the operators $a_{\mathbf{p}}$ have a time dependence in the form

$$a_p(t) = a_p \exp\left\{-\frac{i}{\hbar}\int_0^t \varepsilon\left(\mathbf{p} + \frac{e\mathbf{E}_0}{\omega}\sin\omega t_1\right)dt_1\right\}. \quad (6)$$

This causes formula (4) to differ from the usual expression for the transition probability per unit time in that the δ functions expressing the energy conservation laws are replaced by the quantity

$$\lim_{\delta\to+0}\frac{1}{\pi}\left\langle\int_{-\infty}^{t} e^{\delta t_1}\exp\left\{\frac{i}{\hbar}\int_{t_1}^{t}\left[\varepsilon\left(\mathbf{p}_0+\frac{e\mathbf{E}_0}{\omega}\sin\omega t_2\right)\right.\right.\right. \\ \left.\left.\left. -\varepsilon\left(\mathbf{p}+\frac{e\mathbf{E}_0}{\omega}\sin\omega_2 t\right)-\hbar\omega_{\mathbf{p}_0-\mathbf{p}}\right]dt_2\right\}dt_1\right\rangle. \quad (7)$$

As $\mathbf{E}_0 \to 0$, this quantity goes over into

$$\delta[\varepsilon(\mathbf{p}_0) - \varepsilon(\mathbf{p}) - \hbar\omega_{\mathbf{p}_0-\mathbf{p}}].$$

In the case of a quadratic dispersion law $\epsilon(\mathbf{p}) = \mathbf{p}^2/2m$ we obtain from (4) and (7)[1)]

$$w_{\mathbf{p}_0\to\mathbf{p}} = \frac{2\pi}{\hbar^2}|C_{\mathbf{p}_0-\mathbf{p}}|^2\sum_l J_l^2\left[\frac{e\mathbf{E}_0(\mathbf{p}_0-\mathbf{p})}{\hbar m\omega^2}\right]\left[n_{\mathbf{p}_0}(N_{\mathbf{p}_0-\mathbf{p}}+1)\right. \\ \cdot\delta\left[\frac{\varepsilon(\mathbf{p}_0)-\varepsilon(\mathbf{p})}{\hbar}-\omega_{\mathbf{p}_0-\mathbf{p}}-l\omega\right]+n_{\mathbf{p}_0}N_{\mathbf{p}-\mathbf{p}_0}\delta\left[\frac{\varepsilon(\mathbf{p}_0)-\varepsilon(\mathbf{p})}{\hbar}\right. \quad (8) \\ \left.\left.+\omega_{\mathbf{p}-\mathbf{p}_0}-l\omega\right]\right]\approx\frac{2\pi}{\hbar}|C_{\mathbf{p}_0-\mathbf{p}}|^2\sum_l J_l^2\left[\frac{e\mathbf{E}_0(\mathbf{p}_0-\mathbf{p})}{\hbar m\omega^2}\right] \\ \times n_{\mathbf{p}_0}(2N_{\mathbf{p}_0-\mathbf{p}}+1)\delta[\varepsilon(\mathbf{p}_0)-\varepsilon(\mathbf{p})-l\hbar\omega],$$

where $n_{\mathbf{p}_0}$ is the occupation number of the electrons with momentum $\mathbf{p}_0$, $N_{\mathbf{p}_0-\mathbf{p}}$ is the number of phonons, J_l is a Bessel function, and in the last equation we have neglected the phonon energy compared with the other energies $\epsilon(\mathbf{p})$ and $l\hbar\omega$. At

$$|e\mathbf{E}_0(\mathbf{p}-\mathbf{p}_0)/\hbar m\omega^2| \ll 1 \quad (9)$$

expression (8) coincides with the usual electron-phonon scattering probability. Expression (8) coincides with the kinetic coefficient in the equations obtained by Épshteĭn[6] and Mel'nikov[7].

The probability of interest to us, that of the transition to a state with energy

$$\varepsilon(\mathbf{p}) = \varepsilon(\mathbf{p}_0) + l\hbar\omega; \quad l > 0$$

is equal to

$$\gamma_l = \frac{2\pi}{\hbar}\sum_{\mathbf{p}}|C_{\mathbf{p}_0-\mathbf{p}}|^2 J_l^2\left[\frac{e\mathbf{E}_0(\mathbf{p}_0-\mathbf{p})}{\hbar m\omega^2}\right](2N_{\mathbf{p}_0-\mathbf{p}}+1)\delta[\varepsilon(\mathbf{p})-\varepsilon(\mathbf{p}_0)-l\hbar\omega]. \quad (10)$$

When $l > \mathrm{I}/\hbar\omega$, where I is the ionization potential, using the condition (1), we obtain the criterion of cascade ionization in the form

$$\gamma_l\tau = 40. \quad (11)$$

We have used here the fact that the probability of ionization by an electron that has reached an energy $\epsilon \gtrsim \mathrm{I}$ is much larger than γ_l. In Sec. 4 below we shall estimate the ionizing fields satisfying (10) and (11).

3. PROBABILITY OF IMPACT IONIZATION IN THE PRESENCE OF AN ALTERNATING ELECTROMAGNETIC FIELD

A strong electromagnetic field can not only produce a multiphoton transition in the conduction band of the dielectric, but also change the very probability of the impact ionization. Whereas without a field the energy conservation law admits of ionization only with the aid of "hot" electrons of the conduction band, having $\epsilon \gtrsim \mathrm{I}$ and momenta $p \gtrsim \sqrt{m\mathrm{I}}$, allowance for the field can lead to multiphoton processes that allow the "cold" electrons ($\epsilon \ll \mathrm{I}$, $p \ll \sqrt{m\mathrm{I}}$) to knock out electrons from the valence band. The probability of such a pure electronic process (without participation of phonons) can easily be obtained in analogy with (4), (6), (7), and (8). Thus, assuming in the two-band model[8] that the conduction-band electrons are quasi-free and that the valence electrons are described by a Bloch wave function of the type

$$\varphi_{\mathbf{k}} = V^{-1/2}e^{i\mathbf{k}\mathbf{r}}\sum_{\mathbf{n}} a(\mathbf{n}+\mathbf{k})e^{i\mathbf{n}\mathbf{r}},$$

where $\mathbf{n}$ are the reciprocal-lattice vectors, $\hbar\mathbf{k}$ is the quasimomentum, and the quantities $a(\mathbf{n}+\mathbf{k})$ satisfy the normalization conditions

$$\sum_{\mathbf{n}}|a(\mathbf{n}+\mathbf{k})|^2 = 1,$$

we obtain

$$\gamma_l^{(i)} = \frac{8\pi}{\hbar^2}\left(\frac{4\pi e^2}{V}\right)^2\sum_{\mathbf{k}\mathbf{k}_2'\mathbf{k}_1\mathbf{n}}|a(\mathbf{n}+\mathbf{k}_2')|^2\delta_{\mathbf{k}+\mathbf{k}_1,\,\mathbf{k}_2'+\mathbf{k}_1'+\mathbf{n}} \\ \times\left[\frac{1}{(\mathbf{k}-\mathbf{k}_1')^4}+\frac{1}{(\mathbf{k}_1-\mathbf{k}_1')^4}+\frac{1}{(\mathbf{k}-\mathbf{k}_1')^2(\mathbf{k}_1-\mathbf{k}_1')^2}\right] \\ \times J_l^2\left[\left(1+\frac{m_c}{m_v}\right)\frac{e^2E_0^2}{8\hbar m_c\omega^3}\right]\delta\left[\frac{\hbar}{2m_c}\left(k_1'^2-\frac{m_c}{m_v}k_2'^2-k^2-k_1^2\right)\right. \quad (12) \\ \left.-\omega_g-\left(1+\frac{m_c}{m_v}\right)\frac{e^2E_0^2}{4\hbar m_c\omega^2}+2l\omega\right].$$

Here $\hbar\mathbf{k}_1'$ is the momentum of the electrons in the conduction band prior to the collision, m_c and m_v are the effective masses of the electron in the conduction band and of the valence electron, and $\hbar\omega_g$ is the width of the forbidden band. In the derivation of (12) we assumed also that

$$|(1+m_c/m_v)e\mathbf{E}_0\mathbf{k}_2'/m_c\omega^2| \ll 1. \quad (13)$$

The last condition can be satisfied mainly as a result of smallness of $\mathbf{k}_2'$. Indeed, the energy and momentum conservation limits the maximum $|\mathbf{k}_2'|$ to a value on the order of $(m_v/m_c)^{1/2}|\mathbf{k}_1'|$, which we choose to be small.

In weak fields, when

$$(1+m_c/m_v)e^2E_0^2/8\hbar m_c\omega^3 \ll 1,$$

the obtained expression (12) goes over into the well-known expression for the probability of impact ionization without a field[8,9].

In analogy with (11), we can estimate the field producing cascade ionization by a given process in the form

$$\gamma_l^{(i)}\tau = 40; \quad 2l\hbar\omega > \hbar\omega_g + (1+m_c/m_v)e^2E_0^2/4m_c\omega^2. \quad (14)$$

4. DISCUSSION OF DIFFERENT IONIZATION MECHANISMS. ESTIMATES

Let us stop first to discuss the mechanism whereby an electron acquires energy through a series of single-photon transitions, with allowance (unlike in[5]) for the intraband scattering by acoustic phonons. Recognizing that the photon energy $\hbar\omega$ is much smaller than the ionization energy I, we can describe the change of the energy distribution $n = n(\epsilon)$ resulting from a series of

[1)] It is meaningful to emphasize that the introduction of the effective mass and the use of the ordinary matrix elements of the electron-phonon interaction for electrons with sufficiently large kinetic energy is purely arbitrary.

single-photon transitions and collisions with acoustic phonons in the form

$$\frac{\partial n}{\partial t}=\frac{1}{g}\frac{\partial}{\partial \varepsilon}\left\{g(D+D_1)\left[\frac{\partial n}{\partial \varepsilon}+\beta_1 n\right]\right\}, \qquad (15)$$

where D and D_1 are respectively the coefficients of energy diffusion as a result of collisions only with phonons and with photons and phonons, respectively, g is the density of states, $\beta = 1/kT$, and $\beta_1 = \beta D/(D + D_1)$.

Changing over in the usual fashion to an equation of the Fokker-Planck type[10], we can easily show that in the case of scattering by deformation acoustic phonons at $kT \gg sp(\epsilon)$, where s is the speed of sound and $p(\epsilon) = (2m\epsilon)^{1/2}$, we have

$$D=s^2p^2(\varepsilon)\gamma(\varepsilon), \qquad (16)$$

$$D_1=D\left(\frac{\hbar\omega}{sp(\varepsilon)}\right)^2 2\int_0^1\int_0^1 dx\,dy\,y J_1^2\left[\frac{eE_0 2p(\varepsilon)}{\hbar m\omega^2}xy\right]. \qquad (17)$$

The variable y in the integral of (17) determines the phonon momentum divided by the quantity $2p(\epsilon)$. The integration with respect to x is due to the fact that in the derivation of (15) we made the substitution

$$J_1^2\left[\frac{e\mathbf{E}_0\mathbf{k}}{\hbar m\omega^2}\right]\to\int_0^1 dx J_1^2\left[\frac{eE_0k}{\hbar m\omega^2}x\right].$$

This means in fact that the anisotropic scattering produced in the field has been replaced by a certain angle-averaged isotropic scattering. Obviously, such an averaging is accurate only to within a factor on the order of unity.

Formula (17) determines the connection between the diffusion coefficients D and D_1. The collision frequency in the case under consideration is equal to[10]

$$\gamma(\varepsilon)=\frac{kT\varepsilon_1^2}{\rho s^2}\frac{\sqrt{2m^3\varepsilon}}{\pi\hbar^4}, \qquad (18)$$

where ρ is the density of the crystal, and ϵ_1 is the deformation-potential constant and has the dimensions of energy. When $eE_0 2p(\epsilon)/\hbar m\omega^2 \ll 1$, which corresponds to condition (19), formula (17) takes the form

$$D_1=D\left(\frac{\hbar\omega}{sp(\varepsilon)}\right)^2\frac{1}{6}\left[\frac{eE_0p(\varepsilon)}{\hbar m\omega^2}\right]^2. \qquad (19)$$

Accurate to the refractive index $n \sim 1$, which is not taken into account here, Eq. (19) goes over, upon substitution of (16) and (18), into the formula given for D_1 in Molchanov's paper[5]. In a field $E_0 \sim 10^4$ cgs esu, in which we can use the approximation (9) and which corresponds, in accordance with[5], to breakdown of the dielectric when the remaining parameters in (19) are equal to those in[5], we obtain the relation

$$D_1\lesssim D. \qquad (20)$$

As already noted in the Introduction, it was assumed in[5] that

$$D\ll D_1,$$

i.e., it is possible to neglect the intraband scattering by acoustic phonons. At the same time, the condition under which D is negligible by comparison with D_1 in Eq. (15) should take the form

$$D_1/D\gg I/kT. \qquad (21)$$

This condition has a clear physical meaning. The average time during which energy is acquired under the influence of the field is $I^2/D_1(I)$, and the time of energy acquisition by collision with acoustic phonons at $kT > sp(I)$ (which is satisfied in our case) is equal to[10] $kTI/D(I)$. Thus, the condition (21) is a condition that the time of energy acquisition under the influence of the field be small compared with the average time of energy loss by collisions with acoustic phonons. To satisfy the condition (21) it is necessary to increase the square of the breakdown field by a factor $I/kT \sim 10^3$. In this case, however, one can no longer use the approximation (9), and it is necessary to consider the general expression (17), in which the argument of the Bessel function is not small[2)]. An analysis of the solution of (15) is in this case beyond the scope of the present article. We note only that in the case when (20) holds, the average time required for the electron to acquire an energy I is exponentially large

$$\gamma^{-1}\sim\exp(\beta_1 I)/(D+D_1)\beta_1^2 \qquad (22)$$

and cannot lead to impact ionization within the time of action of the light pulse. In the same case, when

$$D_1/D\gtrsim I/kT,$$

the electric fields necessary to satisfy this condition are so large that ionization can take place both as a result of energy acquisition in a series of single-photon transitions and as a result of the multiphoton processes considered above (for which, obviously, the losses no longer play any role).

We now stop to estimate the fields necessary to realize the multiphoton processes. We consider first a multiphoton transition in the conduction band with participation of acoustic phonons. According to (10)

$$\gamma_l\approx J_l^2\left[\frac{eE_0(p_0-p)}{\hbar m\omega^2}\right]\gamma(I), \qquad (23)$$

where $\gamma^{-1}(I)$ is the average time of momentum relaxation by collision with acoustic phonons, given by formula (18) at $\epsilon = I$. The argument of the Bessel function can be written in the form

$$\varkappa=E_0/\tilde{E},\quad \tilde{E}=I(mI)^{1/2}/e\hbar l^2. \qquad (24)$$

We have taken into account the fact that $p \sim (mI)^{1/2}$ and $\hbar\omega = I/l$. Then, using the condition (11), we find from (23) that a breakdown should set in in fields satisfying the relation

$$J_l^2(\varkappa)=40/\tau\gamma(I).$$

Let us consider the case of breakdown of leucosapphire, discussed in[5], and assume the same values of the parameters: $T = 300°K$, $\rho = 3.8$ g/cm^3, $s = 8 \times 10^5$ cm/sec, $\epsilon_1 = 11$ eV, $I = 9$ eV, $\hbar\omega = 1.17$ eV, and $\tau = 3 \times 10^{-8}$ sec. Then $l \approx 8$, $\gamma(I) \approx 8 \times 10^{13}$, and $J_8(\kappa) = 4 \times 10^{-3}$. Using the tables[11], we obtain $\kappa \approx 4$. As a result, the breakdown field is equal to

$$E_{cr}=4I(mI)^{1/2}/l^2e\hbar\approx 2\cdot10^5\ \text{cgs esu}\ \approx 6\cdot10^7\ \text{V/cm} \qquad (25)$$

Let us estimate now the fields necessary to realize impact ionization with the aid of the mechanism described in Sec. 3. According to (12)

$$\gamma_l^{(i)}\approx J_l^2\left[\left(1+\frac{m_c}{m_v}\right)\frac{e^2E_0^2}{8\hbar m_c\omega^3}\right]\gamma_0^{(i)}\left[\hbar\omega_g-2l\hbar\omega+\left(1+\frac{m_c}{m_v}\right)\frac{e^2E_0^2}{4m_c\omega^2}\right],$$

[2)]It should be noted that in this case D will also depend on the field.

where $\gamma_0^{(i)}$ is the probability of impact ionization (without a field) by an electron of energy

$$\varepsilon \geqslant \hbar\omega_g - 2l\hbar\omega + \left(1 + \frac{m_c}{m_v}\right)\frac{e^2E_0^2}{4m_c\omega^2}.$$

When $2l \gtrsim \hbar\omega_g/\hbar\omega$ (and the last term is neglected), the ionization will be produced by any electron from the conduction band. Owing to the multiphoton transitions, an effective overlap of the valence band with the conduction band ("metallization") takes place. Therefore the process of impact ionization in an alternating field has no threshold. Assuming for simplicity that $m_c = m_v = m$, where m is the free-electron mass, and $\gamma_0^{(i)} \approx 10^{16}$ sec^{-1} (see, for example,[8]), we obtain for the breakdown field the condition (14) in the form (at $\tau = 3 \times 10^{-8}$ sec and $2l = 8$)

$$J_4(y) = 3.6 \cdot 10^{-4}; \quad y = e^2E_0^2/4\hbar m\omega^3.$$

Hence[11] $y \approx 0.6$ and $E_{cr} = 2.4 \times 10^5$ cgs esu, which agrees approximately with the estimate (25).

It should be noted that each of the three considered mechanisms has a different dependence on the parameters, and that the realization of any particular mechanism is determined by the concrete values of the parameters. In particular, the considered multiphoton mechanisms depend strongly on the laser frequency. At a fixed ionization energy I, cascade ionization is made easier when the quantum multiplicity l is increased.

Let us stop now to discuss the conditions of the applicability of the main formulas (10) and (12). The indicated expressions were obtained in second order of perturbation theory in the electron-phonon and electron-electron interaction constants (the Born approximation). Since the characteristic electron energies in (10) are of the order of the ionization potential I, the validity of (10) is obvious. As already noted, the process of impact ionization in an alternating field has no threshold, i.e., the ionization of the valence electrons as a result of collisions with the electrons of the conduction band is possible at relatively low energies of these electrons. For such electrons, however, the Born approximation holds only if

$$e^2\alpha/\hbar v\mu \ll 1, \tag{26}$$

where v is the characteristic velocity of the electron in the conduction band, μ is the dielectric constant of the crystal, and α is of the order of the square of the Bessel function $J_l^2(\kappa)$. In the considered example of leucosapphire breakdown, the condition (26) is satisfied precisely because α is small. If $\alpha \sim 1$, then the criterion (26) is valid only for fast ($\epsilon \sim$ I) electrons.

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