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Avalanche ionization produced in solids by large radiation quanta and relative role of multiphoton ionization in laser-induced breakdown

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The generalized solution of the diffusion kinetic equation for avalanche ionization is generalized to include the region of short radiation pulses of 10^{-9}–10^{-11} sec. A solution of the differential-difference quantum kinetic equation is obtained and it is shown that in the case of large radiation quanta the dependences of the critical field on the frequency and on the pulse duration are substantially altered. The relative roles of avalanche and multiphoton ionization in laser-induced breakdown of transparent dielectrics are analyzed.

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We have previously investigated,[1-3] by solving the quantum-kinetic equation for the conduction-band electrons in the diffusion approximation, the process of avalanche impact ionization as one of the principal mechanisms of breakdown of transparent dielectrics in a strong electromagnetic field. We determined thereby the principal regularities of this process, namely, the dependence of the critical field E_c on the initial lattice temperature T_0 on the frequency Ω, and on the duration of the radiation pulse, in a wide range of the indicated parameters. It was shown in particular that the diffusion approximation provides a rather good description of the process up to field frequencies satisfying the condition $\hbar\Omega \ll I$, where I is the effective ionization potential, and that this approximation leads to a relation of the type

$$E_c^2 \propto \Omega^2 + \nu_{eff}^2, \tag{1}$$

where ν_{eff} is the effective frequency of the hot-electron collisions. The frequency dependence of $E_c(\Omega)$ was subsequently[5] refined for high frequencies by solving a quantum-kinetic difference equation. It turned out that the $E_c(\Omega)$ dependence can be quite weaker in the indicated region than in the diffusion case described by relation (1).

The derived regularities[1-4] have made possible purposeful experimental investigations[4,5] aimed at determining the role of the electron avalanche in laser-induced breakdown of real crystals. It was shown that the dependences of the breakdown thresholds on the radiation frequency and on the temperature, observed for a number of the optically most durable samples of alkali-halide crystals agree with the theoretical predictions and lead to a reasonable estimate, $\nu_{eff} \approx 6\times10^{14}$ sec^{-1}, of the frequency of the electron–phonon collisions. Some experimental results, however, particularly on the temperature dependence of the breakdown thresholds at low and high radiation frequencies, were not fully explained. It was assumed, in particular, that the disparity between the observed temperature dependences of the breakdown threshold and that predicted by the theory of avalanche impact ionization for high-temperature laser radiation ($\lambda = 0.53\ \mu m$) may be due to the effective inclusion of another carrier-generation mechanism, namely multiphonon ionization.

These facts have prompted us to investigate theoretically in greater detail the development of avalanche impact ionization for large radiation quanta and to analyze the relative role of the mechanisms of impact and multiphoton ionization in laser-induced breakdown of transparent dielectrics. The relative contribution made to the carrier generation by these two processes, characterized respectively by the electron-avalanche development constant γ and by the rate W_n of n-photon carrier generation, should be essentially determined by the dependence of γ and W_n on the intensity and frequency of the electromagnetic radiation. It follows from an analysis and multiphoton transitions (see, e.g., Ref. 6), that in the entire investigated range of pulse durations t_p (10^{-7}–10^{-11} sec) we have $W_n \propto E^{2n}$ if $n \lesssim 10$. As to the avalanche development constant γ, it can have a stronger dependence on the frequency for wide-band dielectrics. Thus, in the case of laser breakdown in the nanosecond pulse range, as shown in Refs. 1–3, γ depends exponentially on the radiation intensity. On going to the picosecond band, the $\gamma(E)$ dependence can be weaker.

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Therefore, to determine which of the two carrier generation mechanisms—impact or multiphoton ionization—can predominate, it is necessary first to study in detail the dependence of the avalanche-development constant on the intensity in a wide range of pulse durations. Since this dependence can be significantly different at relatively low radiation frequencies (when the diffusion approximation is valid) and at high frequencies, we must investigate the singularities of the development of the process of avalanche ionization for strong electromagnetic-field quanta.

To answer these questions, we consider in Secs. 1 and 2 the dependence of the avalanche development constant on the intensity at low and high radiation frequencies, and determine more accurately the region in which the diffusion approximation is valid. In Sec. III we consider the dependence of the avalanche-breakdown critical field on the radiation frequency. Finally, using the results obtained in Secs. I–III, we analyze,the breakdown criterion and the relative role of the mechanisms of impact and multiphoton ionization at various durations of the electromagnetic-radiation pulse.

1. DEPENDENCE OF THE ELECTRON-AVALANCHE DEVELOPMENT CONSTANT ON THE RADIATION INTENSITY FOR RELATIVELY WEAK ELECTROMAGNETIC-FIELD QUANTA ($\hbar\Omega \ll I$)

We consider here avalanche ionization at radiation frequencies in the region of validity of the diffusion approximation of the kinetic equation for the electron energy distribution function $\tilde{f}(\varepsilon)$; this equation takes in the case of high-temperature scattering of electrons by acoustic phonons the form

$$\frac{\partial}{\partial x}(x^2 f'(x)+\alpha x^2 f(x))=\gamma_0 \alpha x^{1/2} f(x) \tag{2}$$

and must be solved for the boundary conditions

$$S(0)=2S(1), \tag{3}$$

$$f(1)=0. \tag{4}$$

Here $x=\varepsilon/I$, $S(x)=-[x^2f'(x)+\alpha x^2 f(x)]D(1)$ is the electron flux along the energy axis

$$D(x)=\frac{4(1+q)mv_s^2x^{3/2}}{(2mI)^{1/2}l_{ac}}$$

is the diffusion coefficient, $Q(x)=D(x)/\delta(1+q)$ is the spontaneous energy-loss power, E is the maximum amplitude of the electromagnetic field,

$$q=e^2E^2/6m^2v_s^2\Omega^2, \quad \delta=kT/I,$$

l_{ac} is the mean free path in scattering by acoustic phonons, v_s is the speed of sound, m and e are the effective mass and charge of the electron, $\gamma_0=\gamma Q^{-1}(1)$, $\alpha=[(q+1)\delta]^{-1}$. Since the dependence of the avalanche-development constant γ on the field E (see Sec. III below) determines, via the breakdown criterion, the dependence of the critical field on the pulse duration, while this duration, since we wish to determine the relative roles of impact and multiphoton ionization, is of interest to us in a wide range including the picosecond region, we solve Eq. (3) without the limitation $\alpha\gamma_0 \ll 1$.[1)]

We obtain for Eq. (2) an approximate solution such that the behavior of the function $\gamma(E)$ is described with sufficient accuracy in the intermediate region of variation of the parameter α and goes over into the exact solution in the limits $\alpha \gg 1$ and $\alpha \ll 1$, which correspond to the cases previously considered in Refs. 1–3 and in Ref. 7, respectively.[2)] We note that the case considered in Ref. 7 was used without justification to analyze an electron avalanche at nanosecond radiation pulses (for more details see Refs. 1–3). To simplify the notation we introduce a new function $u(x)=x^{1/2}f(x)$. Since the solution of Eq. (2) as $\alpha\to 0$ is expressed in terms of modified Bessel functions,[7] we can change over to the following Volterra-type integral equation

$$u(x)=G(x,1)-\alpha\int_1^x d\xi\, G(\xi,x)\left\{\xi u'(\xi)+\frac{3}{2}u(\xi)\right\}, \tag{5}$$

where

$$G(\xi,x)=4\{K_2(4(\gamma_0\alpha)^{1/2}\xi^{1/4})I_2(4(\gamma_0\alpha)^{1/2}x^{1/4}) -I_2(4(\gamma_0\alpha)^{1/2}\xi^{1/4})K_2(4(\gamma_0\alpha)^{1/2}x^{1/4})\}. \tag{6}$$

Using (3), we obtain an equation for γ_0:

$$8\gamma_0\alpha-2I_2[4(\gamma_0\alpha)^{1/2}]=\alpha\int_0^1 d\xi\, I_2(4(\gamma_0\alpha)^{1/2}\xi^{1/4})\left\{\xi u'(\xi)+\frac{3}{2}u(\xi)\right\}. \tag{7}$$

At $\alpha \gg 1$ we can expand the Bessel functions in the integrands and confine ourselves to the first term of the expansion. In the other limiting case $\alpha \ll 1$, the error due to such an expansion is also insignificant, since the decisive terms in (7) are those which do not contain the factor α. The error in the intermediate region can be easily estimated by taking into account the next higher terms of the Bessel-function expansion.

Using the integration formulas

$$\int_0^\xi dx\, x^m K_2(\alpha x^{1/4})=\frac{4}{\alpha^{4m+4}}\left\{2^{4m+4}(2m+2)!(2m)! -\sum_{p=0}^{2m}(\alpha\xi^{1/4})^{4m+3-p}\frac{(2m)!2^p}{(2m-p)!}K_{3+p}(\alpha\xi^{1/4})\right\},$$

$$\int_0^\xi dx\, x^m I_2(\alpha x^{1/4})=\frac{4}{\alpha^{4m+4}} \times\left\{\sum_{p=0}^{2m}(-1)^p(\alpha\xi^{1/4})^{4m+3-p}\frac{(2m)!2^p}{(2m-p)!}I_{3+p}(\alpha\xi^{1/4})\right\}$$

and the indicated expansion when necessary, we easily obtain recurrence relations for the fractional moments of the functions $u(x)$ and $u'(x)$, so that (7) can be rewritten in the form

$$\frac{1}{\gamma_0\alpha}\left(2-\frac{I_2[4(\gamma_0\alpha)^{1/2}]}{2}\right)=F(\alpha)-\frac{4}{3}, \tag{8}$$

where the function $F(\alpha)$ is specified by the series

$$F(\alpha)=\sum_{m=0}^{\infty}\frac{2^{m+2}}{(2m+3)!!(2m+1)}\alpha^m$$

and admits of the following integral representation

$$F(\alpha)=\frac{1}{\alpha^{1/2}}\int_0^\alpha \xi^{-2}\gamma\left(\frac{3}{2},\xi\right)e^{\xi}d\xi,$$

where $\gamma(\frac{3}{2},\xi)$ is the incomplete gamma function. This representation and Eq. (8) lead asymptotically as $\alpha\to\infty$ directly to the results of Refs. 1 and 3:

$$\gamma_0\approx\frac{2}{\pi^{1/2}}[(1+q)\delta]^{-1/2}\exp\left(-\frac{1}{(q+1)\delta}\right).$$

To estimate the error due to the expansion of the Bessel functions, we must use the formula for the residual term of the series and perform calculations similar to those made above. It turns out that the largest error of γ_0 is obtained in the range $\alpha = 0.8-4.0$, where it does not exceed 70%, as against 20% outside these limits.

Figure 1 shows the function $\gamma_0(q\delta)$ calculated in accordance with (8). Here and hereafter we use throughout the parameter $\delta = 1/350$, which corresponds to a lattice temperature $T = 300$ K and to an ionization potential $I \approx 9$ eV. The same figure shows for comparison a plot of $\gamma_0(q\delta)$ calculated from the equation

$$2\Delta e^{-\alpha/2}=\Delta \operatorname{ch} \Delta/2-\alpha \operatorname{sh} \Delta/2, \quad \Delta=(\alpha^2+4\gamma_0\alpha)^{1/2}, \tag{9}$$

which is obtained in the solution of the kinetic equation with constant coefficients[5] [after replacing the coefficients of (2) by their values at $x = 1$]. It is seen that the curves differ very little from each other, and at any rate the uncertainty in the quantity

$$\gamma=\gamma_0 Q(1)$$

is much larger, inasmuch as $Q(1)$ is known only accurately to one order of magnitude.

This conclusion has also a much more general character. Calculations similar to those made above were performed for other types of kinetic coefficients [the coefficient $D(x)$ of diffusion along the energy axis and the coefficient $Q(x)$ of the spontaneous energy loss power], and in particular for the case of polar and nonpolar scattering by optical phonons. The results allow us to state that not only the character of the function $\gamma_0(q)$ but also the numerical values depend little on the concrete form and on the behavior of the kinetic coefficients as $x \to 0$, i.e., in final analysis, on the concrete dispersion laws and on the energy dependence of the frequency of the electron–phonon collisions. A peculiar exception occurs in cases when electron "runaway" is possible; since, however, the essential role in wide-band materials at high energies is played by scattering by acoustic phonons, which hinders the runaway, consideration of this effect is outside the scope of the present paper.

2. DEPENDENCE OF THE ELECTRON-AVALANCHE DEVELOPMENT CONSTANT ON THE RADIATION INTENSITY FOR LARGE LIGHT QUANTA

We start from the differential-difference kinetic equation with constant coefficient, derived in Ref. 5 directly from the general quantum equation[8,9]:

$$\gamma_0\delta^{-1}f(x)=f''(x)+\delta^{-1}f'(x)-2sf(x)+sf(x-x_0)+sf(x+x_0),$$
$$x_0=\hbar\Omega/I, \quad s=q/x_0^2.$$

On the basis of the results and discussion in Ref. 5, we omit here the terms that contain the derivatives of the distribution function in the displaced points $x' = x \pm x_0$, as well as the terms that describe multiphoton intraband transitions with participation of phonons, since these terms are not essential at the electromagnetic-radiation frequencies and intensities of interest to us.

In this section we consider only cases when $I/\hbar\Omega = n$ is an integer. Introducing the notation

$$f_k(x)=f((n-k)x_0+x), \quad 0\leqslant x\leqslant x_0, \quad k=1, 2,\ldots, n, \tag{10}$$

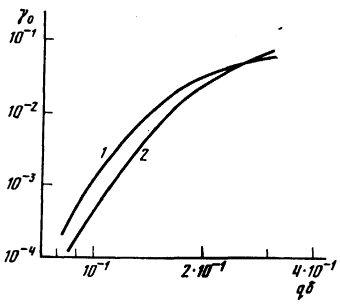

FIG. 1. Dimensionless avalanche development constant γ_0 vs the parameter $q\delta$ (at $\hbar\Omega \ll 1$): 1—calculated from (8); 2—obtained by solving the kinetic equation with constant coefficients.

we obtain the following system of n linear differential equations of second order:

$$\delta^{-1}\gamma_0 f_1=f_1''+\delta^{-1}f_1'+sf_2-2sf_1, \tag{11}$$

$$\cdots\cdots\cdots\cdots$$

$$\delta^{-1}\gamma_0 f_k=f_k''+\delta^{-1}f_k'+sf_{k-1}+sf_{k+1}-2sf_k,$$
$$\cdots\cdots\cdots\cdots \tag{11'}$$
$$\delta^{-1}\gamma_0 f_n=f_n''+\delta^{-1}f_n'+sf_{n-1}-sf_n.$$

The solution of this system is of the form

$$f_k(x)=\sum_{p=1}^{2n} A_k^p e^{\lambda_p x}. \tag{12}$$

The values of λ_p are obtained in the usual manner from the characteristic equation

$$\Delta_n=0, \tag{13}$$

where Δ_n are the corresponding determinants of order $2n$, for which the following recurrence relation holds:

$$\Delta_n=[\lambda(\lambda+\delta^{-1})-(2s+p)]\Delta_{n-1}-s^2\Delta_{n-2}, \tag{14}$$

with

$$\Delta_0=1, \quad \Delta_1=\lambda(\lambda+\delta^{-1})-s-p, \quad p=\gamma_0\delta^{-1}.$$

Thus,

$$\Delta_n=\frac{1}{y_1-y_2}\{(y_1+s)y_1^n-(y_2+s)y_2^n\},$$

where y_1 and y_2 are the roots of the characteristic equation for the difference relation (14). Solving (13), we get

$$\lambda(\lambda+\delta^{-1})^{(p)}=\gamma_0\delta^{-1}+4s\cos^2\frac{\pi p}{2n+1}, \quad p=1,2,\ldots,n.$$

and for the coefficients of $\exp(\lambda_p x)$ in the distribution functions we have

$$A_k^p=G_k^p A_0^p,$$

where

$$G_k^p=(-1)^{k+1}\sin\frac{2\pi pk}{2n+1}\Big/\sin\frac{2\pi p}{2n+1}.$$

The sought value of the avalanche development constant γ_0 is obtained from the solution of the equation

$$|u_{lm}|=0. \tag{15}$$

In the determinant $|u_{lm}|$, $2n-1$ rows are the result of the conditions that the distribution functions and the flux be continuous at the points $x_k = kx_0$, $k = 1, 2, \ldots, n-1$, while the remaining two rows correspond to boundary conditions similar to (3) and (4), but with allowance for the fact that x_0 is finite:

$$f_1(x_0)=0,$$

$$2\left\{f_1'(x_0)+\delta^{-1}f_1(x_0)-s\int_0^{x_0}f_1(x)\,dx\right\}-f_n'(0)-\delta^{-1}f_n(0)=0. \qquad (16)$$

The elements of the determinant equation are thus

$$u_{1,p}=e^{\lambda_p x_0},$$

$$u_{2,p}=G_n^p(\lambda_p+\delta^{-1})-2\lambda_p e^{\lambda_p x_0}+\frac{2s}{\lambda_p}(e^{\lambda_p x_0}-1),$$

$$u_{2+k,p}=G_{k+1}^p e^{\lambda_p x_0}-G_k^p,\quad k=1,\ldots,n-1,$$

$$u_{n+1+k,p}=\lambda_p u_{2+k,p},\quad k=1,\ldots,n-1,\quad p=1,2,\ldots,2n.$$

We note that in the considered frequency region

$$x_0/\delta\gg 1 \qquad (17)$$

and the terms containing exponentials of $\lambda_i x_0$ with $\lambda_i<0$ can be neglected in the calculation.

We consider first the case when

$$s\delta x_0\ll 1, \qquad (18)$$

and all the more, according to (17),

$$s\delta^2\ll 1. \qquad (19)$$

The general solution of (15) in the lowest orders in the indicated parameters is of the form[3)]

$$\gamma_{0,n}\approx\frac{n^{n+2}}{n!}(q\delta)^{n+1}+C_{2n-1}^{n-1}\frac{n^{2n}}{\delta}(q\delta^2)^n,\quad n>1. \qquad (20)$$

We note that since the inequality (17) is satisfied with a large margin, the second term of (20) is negligible at $n=3-10$:

$$\frac{(2n-1)!}{q(n-1)!}(\delta n)^{n-2}\ll 1$$

for all reasonable values of q ($q>10$) and δ ($\delta<\frac{1}{250}$).

Thus, in contrast to the diffusion solution, in the limit (18), when $\gamma_0\ll 1$, the avalanche-development constant is a power-law function of the field

$$\gamma\infty E^{2(n+1)}. \qquad (21)$$

In the expansion of (20) in powers of $s\delta x_0$, the coefficients a_k of some of the terms are of order n^k, so that if the inequality (18) is not satisfied rigorously enough this expansion cannot be used: with increasing n, the region where $\gamma_0(E)$ is given by a power law such as (21) shifts into the region of pulses of increasing duration. This explains how the transition to the diffusion solution takes place.

More general $\gamma_0(q)$ dependences can be easily calculated subject to the rather weak limitation (19), which is satisfied at $n=3-6$ all the way to $\gamma_0\approx 0.1$.[4)]

The results are given in Fig. 2, which shows for comparison also the plot of $\gamma_0(q\delta)$ obtained in the diffusion approximation (for a kinetic equation with constant coefficients). It is seen that at $n\geq 5$ the diffusion solution describes quite well the function $\gamma_0(q\delta)$ in the entire pulse-duration region of interest to us.

The boundary condition (16) and the differential equation (11′) were written down for the case when both electrons are in the vicinity of the point $x=0$ after the impact-ionization act. Of course, this does not correspond fully to the real situation. Since it is impossible to obtain the solution of the complex distribution of the electrons that take part in the impact ionization, we confine

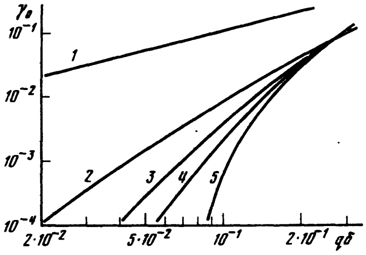

FIG. 2. Dimensionless avalanche development constant γ_0 vs the parameter $q\delta$: 1—for $n=1$ quantum, 2—for $n=2$, 3—for $n=3$, 4—for $n=4$; 5—dependence obtained in the diffusion approximation.

ourselves to another case that is limiting in a certain sense: if the ionization is produced by an electron with energy $(n-1)\hbar\Omega+\varepsilon_0$, $0\leq\varepsilon_0\leq\hbar\Omega$, then electrons with energy $\varepsilon=0$ and $\varepsilon=\varepsilon_0$ result. Equation (11′) and the boundary condition (16) take the form

$$\delta^{-1}\gamma_0 f_n=f_n''+\delta^{-1}f_n' +sf_{n-1}+sf_1-sf_n,$$

$$2(f_1'(x_0)+\delta^{-1}f_1(x_0)) -s\int_0^\infty f_1(x)\,dx-f_n'(0) -\delta^{-1}f_n(0)=0.$$

Calculations similar to those given above show that the character of the function $\gamma_0(q\delta)$ changes insignificantly, and in particular, the relation (20) remains valid. The coefficients of the higher powers of $sx_0\delta$ are insignificantly altered. By way of example we present the simple calculation relation for $n=3$:

$$\gamma_0(2u_1-3u_2+u_3-3u_1u_2-3u_1u_3+6u_2u_3+6u_1u_2u_3) =s\delta[-4u_1u_2+8u_1-5u_2+1+6x_0s\delta(u_1u_2+u_2-u_1)],$$

where

$$u_p=\exp(\lambda_p x_0),\quad \lambda_p=\{-1+[1+4(\gamma_0+g_ps\delta)]^{1/2}\}/2\delta,$$
$$g_1=3,\quad g_2=2,\quad g_3=0.$$

3. DEPENDENCE OF THE AVALANCHE-DEVELOPMENT CONSTANT ON THE LASER-RADIATION FREQUENCY

In the preceding section we confined ourselves to the case $I/\hbar\Omega=n$ with integer n. The method employed can be easily generalized also to the case when I and $\hbar\Omega$ are commensurate:

$$I/\hbar\Omega=n+p/m,$$

where n, p, and m are integers. We must subdivide I into $nm+p$ parts, after which we determine f_1 in the region $1-p/(mn+p)\leq x\leq 1$, f_2 in the region $1-m/(mn+p)\leq x\leq 1-p/(mn+p)$, and so forth, and obtain thus a system of equations similar to (11) for $2n+1$ functions, in which only functions with even (odd) indices are coupled with one another in the kinetic equations.

The expansion similar to (20) takes the form (without the terms $\sim q\delta^2$):

$$\gamma_{0,n,\ldots,n+1}=s\delta\left\{\frac{(s\delta z_2)^n}{n!}+\frac{(s\delta z_1)^{n+1}}{(n+1)!}+\eta_n(s\delta z_2)^{n+1}+\text{cross terms}\right\},$$

where z_1 and z_2 are respectively the lengths of the even and odd segments into which the unit-length interval was broken down.

It is obvious that this result can be extended, from continuity considerations, to include the case when the ratio $I/\hbar\Omega$ is arbitrary. By way of example we present a formula for the avalanche development constant, which is valid only if $\frac{1}{3} \leqslant x_0 \leqslant \frac{1}{2}$:

$$\gamma_{0,2,\ldots,3}=s\delta\{{}^1/_2(s\delta)^2(3x_0-1)^2+{}^1/_6(s\delta)^3(1-2x_0)^3 + {}^1/_2(s\delta)^3(3x_0-1)(1-2x_0)(5x_0-1)\}+6s^2\delta^3(3x_0-1)^{5)}. \tag{22}$$

Figure 3 shows the calculated plots of the critical field against the frequency of the laser radiation for two values of the avalanche development constant: $\gamma_0=10^{-5}$ (for nanosecond pulses) and $\gamma_0=3\cdot10^{-2}$ (picosecond pulses). The diffusion solution was used in the region $x_0 \lesssim 0.2$ (see Sec. I).

It is seen from the figure that the character of the frequency dependence of the critical field is determined essentially by the duration of the electromagnetic-radiation pulse. At nanosecond durations the breakdown threshold can decrease with increasing frequency of the electromagnetic field even at $x_0 \gtrsim 0.3$, as already noted previously,[5] and $E_c(\Omega)$ is an oscillating function. The oscillations are due to the presence of sharp maxima of the distribution function $f(x)$. Allowance for the exact boundary conditions can lead to the appearance of additional shallow maxima and to a certain smoothing of the frequency dependence of the critical field.

In concluding the analysis of the singularities in the development of the electron avalanche at large laser-radiation quanta, it must be noted that the dependence of the critical field on the initial sample temperature remains the same also in this case as in the diffusion approximation, under conditions when the field frequency exceeds the frequency of the electron–phonon collisions: $E_c^2 \propto T^{-1}$.

4. RELATIVE ROLE OF THE IMPACT-IONIZATION AND MULTIPHOTON-IONIZATION MECHANISMS IN LASER-INDUCED BREAKDOWN

The change of the free-carrier density N in the crystal conduction bandband as a result of impact and multiphoton ionization is described by the equation

$$\frac{dN}{dt}=\gamma N+W_n-R(N), \tag{23}$$

where $R(N)$ is the term that describes the carrier recombination.[6)]

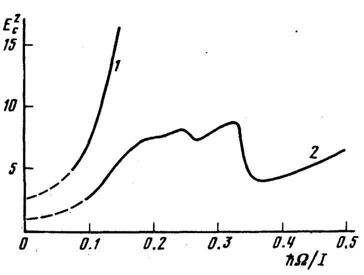

FIG. 3. Dependences of the square of the critical field E_c^2 (in arbitrary units) on the laser-radiation quantum for different values of the avalanche development constant: 1—$\gamma_0=3\cdot10^{-2}$; 2—$\gamma_0=10^{-5}$.

It is obvious that to determine which of these two carrier-generation mechanisms predominates in laser-induced breakdown we must not only know the values of γ and W_n, but also investigate the breakdown criterion that connects the rate of free-carrier generation sufficient to produce breakdown with the duration of the radiation pulse or with the recombination time. A criterion for breakdown due to impact ionization was obtained[1] for the case when the pulse duration is shorter than the carrier recombination time τ_r. It was shown that such a criterion is a rapid explosive-like increase of the crystal-lattice temperature T, due to absorption of the radiation energy by the free carriers in the course of the avalanche impact ionization.

It is of interest to analyze this "breakdown temperature criterion" in greater detail with account taken of the variation of the avalanche-development constant in the course of the process, for example,because of the change in the lattice temperature, and also with account taken of the carrier recombination.

We discuss first the case when the carrier recombination during the time of the field-pulse action can be neglected ($\tau_r \gg t_p$). The equations that describe the avalanche generation of carriers in the conduction band and the heating of the lattice as a result of the electron-phonon collisions can then be written in the form

$$\frac{dN}{dt}=\gamma^0\varphi(\Theta)N,\quad \frac{d\Theta}{dt}=\beta\Theta^{\varkappa}N, \tag{24}$$

where $\Theta=T/T_0$, T_0 is the initial lattice temperature; if scattering by acoustic phonons predominates, the parameters are[1,3]:

$$\beta=4Q(1)(q\delta_0)^{3/2}I/\pi^{1/2}C\rho T_0,\quad \delta_0=kT_0/I,\quad \varkappa={}^3/_2.$$

Here C is the specific heat of the lattice, ρ is the density,

$$\gamma=\gamma^0\varphi(\Theta),\quad \gamma^0=\gamma(T_0).$$

The solution of the system (24) is

$$\gamma^0 t=\int_1^{\Theta}\frac{d\Theta'}{\Theta'^{3/2}A(\Theta')},\quad A(\Theta')=\frac{\beta N_0}{\gamma^0}+\int_1^{\Theta'}d\Theta''\Theta''^{-3/2}\varphi(\Theta''), \tag{25}$$

(N_0 is the initial carrier density) and is the sought breakdown criterion. Since the integral in (25) always converges, the function $\Theta(t)$ has a vertical asymptote, i.e., the temperature rise has an explosive character. The actual value of the energy needed for the breakdown is immaterial here, in other words, there is no need to calculate the effective lattice temperature at which the irreversible changes occur in the matrix, provided that it is not close to the initial value. It suffices to calculate the integral in (25) as $\Theta\to\infty$. Of course, this does not mean physically that an "infinite" temperature, giant carrier densities, and so on, will be reached. The accuracy of the value of $\gamma^0 t_p$ calculated in this manner is sufficient. In fact, we present the values of the product $\gamma^0 t_p$ calculated in accordance with (25) with allowance for the $\gamma(\Theta)$ dependence that follows from the results of Sec. I for different values of the critical temperature Θ_c:

$\gamma^0 t_p$:	10.70	12.08	12.80	12.88
Θ_c:	1.01	1.06	1.33	>2.4

We see that in a time exceeding $0.9t_p$ the heat rise is only approximately 10°. This is precisely the circumstance that allows us to regard the avalanche ionization development constant as independent of temperature and to obtain the simple breakdown criterion

$$\gamma t_p=\ln\left(1+\frac{2\gamma}{\beta N_0}\right), \tag{26}$$

whose accuracy is perfectly satisfactory.

We consider now the breakdown criterion for the avalanche breakdown mechanism in the presence of rapid carrier recombination: $\tau_r < t_p$. If the electromagnetic-field frequency exceeds the electron–phonon collision frequency, then the breakdown criterion is obvious: $\gamma\tau_r \approx 1$. The situation is more complicated if $\Omega < \nu_{\text{eff}}$, inasmuch as in this case the rate of avalanche ionization decreases with increasing lattice temperature, and if it becomes at some instant lower than the recombination rate, no further heating will take place. To analyze this phenomenon, we consider a solution of a system of equations analogous to (24)[7]:

$$\frac{dN}{dt}=\left(\gamma^0\varphi(\Theta)-\frac{1}{\tau_r(\Theta)}\right)N,\quad \frac{d\Theta}{dt}=\beta N. \tag{27}$$

This solution is

$$t=\int_1^\Theta\frac{d\Theta'}{\beta N_0+B(\Theta')},\quad B(\Theta')=\int_1^{\Theta'}d\Theta''\left(\gamma^0\varphi(\Theta'')-\frac{1}{\tau_r(\Theta'')}\right).$$

To make a final temperature Θ_c possible, it is necessary to satisfy the condition

$$\int_1^{\Theta_c}d\Theta''(\gamma^0\varphi(\Theta'')-\tau_r^{-1}(\Theta''))>0,$$

and the breakdown criterion can be written in the form

$$\int_1^{\Theta_c}d\Theta''(\gamma^0\varphi(\Theta'')-\tau_r^{-1}(\Theta''))=0. \tag{28}$$

In particular, if τ_r is independent of temperature and $\varphi(\Theta)\propto\Theta^{-2}$, we get

$$\gamma^0\tau_r=\Theta_c. \tag{29}$$

The breakdown criterion (29) at $\tau_r \lesssim 10^{-11}$ makes for a much weaker dependence of the critical breakdown field on the initial temperature of the sample, and in the case when the recombination rate increases with increasing temperature, complete absence of the temperature dependence of the breakdown threshold may be observed in the experiments.

If the main carrier generation mechanism is multiphoton ionization, the breakdown criterion, which is likewise obtained from a simultaneous solution of the system of equations for the rate of change of the carrier density and of the lattice temperature [the first equation of (24) must be replaced] takes the form

$$4(1-\Theta_c^{-1/4})=\beta W_n t_p^2. \tag{30}$$

It is seen that the critical intensity depends little on the final temperature in the interaction region.

The foregoing results yield the dependence of the critical intensity on the laser-radiation pulse duration for the two limiting mechanisms of carrier generation in impact and multiphoton ionization. By way of example, Fig. 4 shows these dependences for $n=5$, calculated using the results of the preceding section and the breakdown criteria (30) and (25).

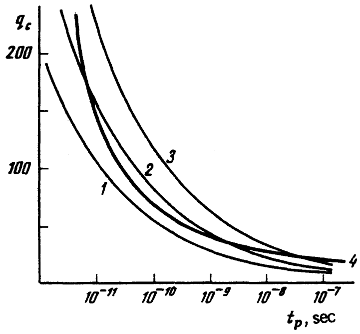

FIG. 4. Dependences of the parameter q_c, which determines the critical field ($q_c \propto E_c^2$) on the duration t_p of the laser-radiation pulse (NaCl crystal, $n=5$), for different laser-breakdown mechanisms: 1, 2, 3—for multiphoton ionization, $W_1 > W_2 > W_3$, 4—for impact ionization.

The parameters used in the calculations were $N_0=10^{12}$ cm^{-3}, $Q(1)=5\cdot 10^{12}$ sec^{-1}, $\rho=2.17\,g/\text{cm}^3$, $C\rho=2.86\cdot 10^{17}$ erg/deg (NaCl), and $T=300$ K.

Curves 1, 2, and 3 show the dependence of the critical field on the pulse duration for the generation mechanism connected with multiphoton ionization of the lattice at different values of W_n. We see that, depending on the rate of multiphoton ionization, two principally different cases are possible:

1. Curves 1 and 4 do not cross. This means that in the entire range of laser pulse durations it is impossible to observe the breakdown mechanism due to the electron avalanche, and the dominant limiting breakdown mechanism is the appearance of nonlinear absorption by the free electrons produced as a result of multiphoton interband transitions.

2. In the second case (curves 2 and 4), the breakdown threshold is determined by the development of the electron avalanche in the pulse-duration region

$$t_p^{(1)}<t_p<t_p^{(2)}. \tag{31}$$

Both at $t_p<t_p^{(1)}$ and at $t_p>t_p^{(2)}$ multiphoton ionization again predominates. The pulse duration region defined by the inequalities (31) can generally speaking be quite large and include both the nanosecond and the picosecond ranges (see curves 3 and 4).

[1]This condition is equivalent to $\alpha \gg 1$.
[2]It will be made clear later on that $\gamma_0\alpha \to$ const as $\alpha=0$
[3]$\gamma_0=s\delta$ at $n=1$.
[4]At $n=2$ the results can be easily obtained also without the restrictions (19).
[5]The last term of (22) corresponds to the second term of the expansion (20).
[6]We disregard here and below the carrier diffusion and the heat dissipation from the interaction region (see Ref. 1).
[7]When $\Omega<\nu_{\text{eff}}$, the exponent is $\varkappa=\frac{1}{4}$ (Ref. 1), and for simplicity we neglect the corresponding relation in (24). In addition, we confine ourselves to linear recombination, since the character of the recombination does not influence substantially the results that follow.

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Manifestation of the proximity effects in tunnel conductance of multilayer systems

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An experimental investigation was made of the tunnel characteristics of thin films of "poorly conducting" substances, CuCl, Bi–Sb, and normal metals, Cu and Ag, covered by superconducting lead. These characteristics were determined in a wide temperature range (1–300°K) at pressures up to 17 kbar. The changes in the tunnel conductance of these systems were explained qualitatively within the framework of the familiar ideas of the proximity effect. It was found that a copper chloride film had semiconducting properties and did not go over to the metallic and superconducting state.

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1. INTRODUCTION

The electron spectrum of a conducting material undergoes considerable changes when this material is brought into contact with a superconductor. An investigation of this phenomenon, known as the proximity effect, is of fundamental importance for the understanding of the nature of superconductivity[1] and it makes it possible to determine various characteristics describing the properties of the electron and phonon subsystems of the investigated objects.[2–4] The extensive experimental data on the proximity effect have been obtained mainly for contacts of a normal metal with a superconductor.[1] It seems interesting and desirable to investigate multilayer structures with semimetals and semiconductors because of the possibility of superconductivity mechanisms other than the electron-phonon effect, particularly the exciton mechanism near a semiconductor–superconductor interface.[5] In this sense, there is definite interest in systems such as CuCl and Bi–Sb, in which a transition to a metallic and even superconducting state is possible at high pressures.[6–8]

We carried out tunnel experiments on thin films of CuCl (10–200 Å) and Bi–Sb (1000 Å), which were in direct contact with superconducting lead. A study was made of changes in the tunnel conductance because of the proximity of these relatively "poorly conducting" substances with a superconductor, and because of the application of hydrostatic pressures up to 17 kbar.

We carried out additional experiments on normal metal–superconductor systems (Cu/Pb, Ag/Pb) composed of films of the same thickness as in the case of CuCl/Pb. In the majority of earlier investigations of these structures, attention was concentrated on the behavior of the tunnel conductance near the energy gap and the experiments were usually carried out on relatively thick (≳200 Å) films of normal metals.[1] Having thus extended the investigated energy range, we shall consider the applicability of various models of the proximity effect to the tunnel characteristic as a whole in the range of energies corresponding to the superconducting gap as well as in the range of phonon excitations of the superconducting (lead) coating.

2. EXPERIMENTS

1. We investigated cross-shaped structures of the superconducting aluminum–aluminum oxide–investigated substance/lead (Al–I–N/Pb) type, prepared by thermal evaporation of the various substances in a vacuum of ~10^{-6} Torr. A tunnel barrier was formed by oxidation of aluminum in a dry oxygen atmosphere or in air for 5–15 min at a pressure of 0.3–0.5 Torr in the chamber. Alu-

