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Statistical features of avalanche ionization of wide-gap insulators by laser radiation under conditions of shortage of initiating electrons

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A theoretical investigation is made of the characteristics of the process of laser damage to transparent insulators as a result of development of an electron avalanche when this avalanche is retarded by a shortage of initiating electrons. The process of simultaneous formation of initiating electrons and of an avalanche is considered from the statistical point of view. The expected dependence of the breakdown threshold on the volume of the interaction zone is analyzed.

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There are several mechanisms of photoconductivity in wide-gap solid insulators. Free carriers may appear as a result of interband transitions involving the absorption of one or more photons (photoionization of the matrix), as a result of ionization of impurities and defects, and also because of development of impact avalanche ionization. This last mechanism has a number of important features. Firstly, an avalanche-like increase in the number of free carriers practically always results in optical breakdown of an insulator. Secondly, this mechanism requires the presence of a certain initial number of free carriers in the zone of interaction of radiation with matter. These carriers may form in wide-gap insulators at reasonable temperatures only as a result of photoionization of the matrix or defects.

The process of impact avalanche ionization is usually considered beginning from a certain initial density $N_0 \approx 10^7 - 10^{12}$ cm^{-3}, the actual value depending on the laser radiation frequency. It is found that the breakdown threshold field depends very weakly on N_0 (Refs. 1 and 2). However, it is obvious that this conclusion can only be valid if a sufficient number of initiating carriers is present in the interaction zone of volume V. Since in many laser damage experiments[3,4] the size of the interaction region is very small, $V \lesssim 10^{-10}$ cm^3, there may be a situation when the threshold field is governed not so much by the rate of development of an avalanche but by the rate of creation of initiating electrons. However, it should be stressed particularly that the situation under discussion here is radically different from the usual case of breakdown as a result of multiphoton ionization, although in both cases the damage threshold is governed by the photoionization rate. In fact, in the case under discussion, we have

$$\gamma > W,$$

where γ is the avalanche growth constant and W is the frequency of formation of carriers as a result of photoionization, whereas in the case of damage as a result of multiphoton ionization the opposite inequality is satisfied.

We shall consider the influence of an insufficient number of initiating electrons on the threshold (critical) damage field and the associated statistical features of the optical breakdown process. In particular, we shall discuss the temperature and size dependences of the breakdown threshold under these conditions.

1. ELECTRON AVALANCHE IN THE PRESENCE OF A SMALL NUMBER OF INITIATING ELECTRONS

Let us assume that a sequence of impact ionizations by some specific electron is a Poisson process, namely that the probability of a certain electron making k ionizations in a time t is

$$P_k(t) = \frac{(t/\tau)^k}{k!} \exp\left(-\frac{t}{\tau}\right),$$

where $\tau \approx \gamma^{-1}$. An analysis of the usual conditions for the process to be of the Poisson type leads to the conclusion that this is a reasonable assumption if

$$\tau \gg 1/\nu,$$

where ν is the frequency of electron–phonon collisions.

Let us assume that initially, at $t=0$, the interaction zone contains l initiating electrons. We shall use $R_n^l(t)$ to denote the probability that at a moment t there are n electrons. The functions $R_n^1(t)$ obey the following recurrence relationship:

$$R_{n+1}^1(t)=\int_0^t R_n^1(t')\frac{n}{\tau}\exp\left[-\frac{(n+1)(t-t')}{\tau}\right]dt',$$

whose solution is the function

$$R_n^1(t)=[1-e^{-t/\tau}]^{n-1}e^{-t/\tau}.$$

The above can be proved easily by induction. Next, we obviously have

$$R_n^l(t)=\sum_{k=0}^{n-l}R_{l-1+k}^{l-1}(t)R_{n-l-k+1}^1(t),$$

so that

$$R_n^l(t)=C_{n-1}^{l-1}[1-e^{-t/\tau}]^{n-l}e^{-lt/\tau}. \qquad (1)$$

We know that breakdown appears if during a laser radiation pulse the electron density in the conduction band N reaches a certain critical value $N_{cr}\approx 10^{17}-10^{18}$ cm^{-3}, which should be such that the absorbed energy is sufficient for irreversible processes to take place in the matrix. Thus, the probability of breakdown is the function

$$G_n^l(t)=\sum_{k=n}^{\infty}R_k^l(t)=1-\frac{\Gamma(n)}{\Gamma(l)\Gamma(n-l)}B_{1-\xi}(l,n-l), \qquad (2)$$

where $n=N_{cr}V$; $\xi=1-e^{-t/\tau}$; $B_x(a,b)$ is an incomplete B function. In particular, if the interaction zone contains initially one electron, the probability of breakdown in a time t is given by

$$G_n^1(t)=[1-e^{-t/\tau}]^{n-1}.$$

We shall assume that the damage threshold is defined experimentally as the radiation intensity at which the probability of breakdown is $P=1/2$. Then, bearing in mind that $n\gg 1$, we obtain the relationship

$$t/\tau=\ln(n/\ln 2),$$

which is practically identical with the usual breakdown criterion.[1] Moreover, variation of P within the reasonable limits $P=0.1-0.9$ does not alter significantly the critical intensity. Hence, we can draw two important conclusions. Firstly, even one initiating electron in the interaction zone is sufficient for the development of an avalanche in spite of the statistical nature of the process. Secondly, the scatter of the breakdown thresholds in the case of pure avalanche breakdown remains very slight if

$$n_0=N_0V\gg 1.$$

Thus, all the statistical properties appear only if this last inequality is not obeyed. We shall study these properties in detail by considering the process of avalanche ionization which is simultaneous with impurity photoionization.

2. AVALANCHE IONIZATION INITIATED BY MULTIPHOTON IONIZATION OF IMPURITIES OR MATRIX

Let us assume that the probability of appearance of m electrons in a time t as a result of multiphoton ionization is

$$Q_m(t)=\frac{(t/\tau_1)^m}{m!}\exp\left(-\frac{t}{\tau_1}\right).$$

We shall use $S_n(t)$ to denote the probability that at a moment t this ionization process, which occurs simultaneously with impact avalanche ionization, produces n electrons in the interaction zone in the case when initially (at $t=0$) there are no free carriers in this zone. We then find that the functions $S_n(t)$ obey

$$S_n(t)=\int_0^t dt'\,S_{n-1}(t')\left(\frac{n-1}{\tau}+\frac{1}{\tau_1}\right)\exp\left[-\frac{n(t-t')}{\tau}\right]\exp\left(-\frac{t-t'}{\tau_1}\right)$$

[clearly $S_0(t)=e^{-t/\tau_1}$] and hence

$$S_n(t)=\frac{1}{n!}\left[\prod_{k=0}^{n-1}\left(k+\frac{\tau}{\tau_1}\right)\right][1-e^{-t/\tau}]^n\exp\left(-\frac{t}{\tau_1}\right)$$
$$=\frac{\Gamma(n+\tau/\tau_1)}{\Gamma(\tau/\tau_1)\Gamma(n+1)}[1-e^{-t/\tau}]^n\exp\left(-\frac{t}{\tau_1}\right).$$

The probability of breakdown in a time t can be written as follows:

$$G_n=\sum_{k=n}^{\infty}S_k(t)=\xi^n\frac{(1-\xi)^\alpha\Gamma(n+\alpha)}{\Gamma(\alpha)\Gamma(n+1)}F(1,n+\alpha;n+1;\xi)=1-\frac{B_{1-\xi}(\alpha,n)}{B(\alpha,n)}, \qquad (3)$$

where $\alpha=\tau/\tau_1$; $F(a,b;c;x)$ is the hypergeometric Gauss function ${}_2F_1$. A comparison of Eqs. (3) and (2) shows that the parameter τ/τ_1 corresponds physically to the number of initiating electrons l.

Using the asymptotic form of an incomplete B function in the limit $n\to\infty$, we obtain the following relationship which is more convenient in calculations:

$$P=G_n\approx\frac{\Gamma[\alpha,ne^{-t/\tau}]}{\Gamma(\alpha)}, \qquad (4)$$

where $\Gamma(\alpha,x)$ is an incomplete Γ function. For a given breakdown probability P any of the relationships (3) or (4) can be regarded as the equation which, together with the expressions that give the dependences of the quantities τ and τ_1 on the radiation intensity I, can be used to calculate the breakdown threshold I_{cr}.

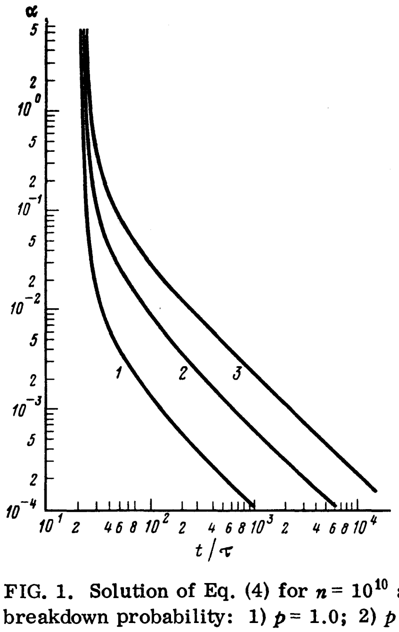

FIG. 1. Solution of Eq. (4) for $n=10^{10}$ and three values of the breakdown probability: 1) $p=1.0$; 2) $p=0.5$; 3) $p=0.9$.

Figure 1 shows the dependences of α on t/τ for three values of the damage probability. The range

$$\alpha<1$$

corresponds to a strong delay of an avalanche because of the absence of initiating electrons. In this case the breakdown probability is given by

$$G_n=1-\exp(-t/\tau_i), \tag{5}$$

i.e., it is the probability of the appearance of at least one free electron during a pulse. In the range

$$\alpha>1$$

the damage threshold corresponds to the conventional avalanche breakdown and is found in the usual way.[2]

It should be noted that the results of the present section showed if there is a shortage of initiating electrons, one can expect that experiments will demonstrate a considerable increase in the spatial scatter of the laser damage threshold.

3. DEPENDENCE OF THE CRITICAL FIELD ON THE LATTICE TEMPERATURE

It follows from recent direct experiments[5] that the case of an avalanche delayed by a shortage of initiating electrons, discussed above, is not observed at least at the neodymium laser and higher frequencies ($\lambda \lesssim 1\ \mu$). However, when the laser radiation frequency is reduced, the probabilities of multiphoton ionization of impurities and the matrix decrease strongly so that, in principle, we may have the situation of avalanche delay discussed above.

Attempts to explain damage caused by radiation of wavelengths 10.6 and 2.76 μ to even specially selected samples have met with a number of difficulties when the electron avalanche mechanism is involved.[3,4] First of all, the falling temperature dependence of the threshold, followed by a plateau at higher temperatures, observed at the wavelength of 2.76 μ (Ref. 4) should, according to the theory, correspond to a rising temperature dependence at the wavelength of 10.6 μ, whereas it is found experimentally[3] that the damage threshold is independent of temperature. Some possible explanations of this disagreement have been put forward earlier.[3] We shall show here that, in certain cases, the anomalous temperature dependence can be explained by the delayed avalanche mechanism discussed above.

We shall assume that throughout the investigated range of lattice temperatures the damage mechanism is associated with electron avalanches and the frequencies of laser radiation Ω and of the electron–phonon collisions obey $\Omega>\nu$. If at the same time the number of initiating electrons in the interaction zone is $n_0=N_0V \geqslant 1$, it follows from Sec. 1 that the critical intensity should exhibit a temperature dependence characteristic of the pure avalanche breakdown mechanism:

$$I_{cr}\propto T^{-1}.$$

In the other limiting case when

$$n_0<1,$$

the avalanche delay results in an absence of a temperature dependence. In this case this absence of a temperature dependence is observed also for $\Omega<\nu$. It seems reasonable to assume that this is the situation in the case of damage to alkali halide crystals at the wavelength of 10.6 μ.

We shall consider in greater detail the case when the breakdown mechanism is of purely avalanche origin and the inequality $\Omega>\nu$ is obeyed and the initial lattice temperature T is less than a certain value T', but an avalanche is delayed at temperatures[1)] $T>T'$. We shall use the recent results[6] demonstrating the existence of short recombination times of electrons in crystals ($\tau_r \sim 10^{-11}$ sec) assuming that the parameter α in Eq. (4) can be represented by[2)]

$$\alpha=1\Big/\tau_i\left(\frac{1}{\tau}-\frac{1}{\tau_r}\right).$$

We shall next use the dependence $\tau(I)$ given by[7]

$$2\Delta e^{-\beta/2}=\Delta\,\mathrm{ch}\frac{\Delta}{2}-\beta\,\mathrm{sh}\frac{\Delta}{2},$$

where

$$\Delta=(\beta^2+4\gamma_0\beta)^{1/2},\quad \gamma_0=\gamma/Q,$$

Q is the characteristic rate of the electron-phonon processes ($\sim 10^{12}-10^{13}$ sec^{-1}),

$$\beta=\frac{1}{Qq\delta_0}\left(1+\frac{\nu^2\theta^2}{\Omega^2}\right),$$

and

$$\delta_0=\frac{kT_0}{\varepsilon_i},\quad q=\frac{e^2E^2}{6m^2v_s^2\Omega^2}\propto I,\quad \theta=\frac{T}{T_0}.$$

Here, ε_i is the ionization potential; v_s is the velocity of sound; m and e are the mass and charge of an electron; $T_0=300$ °K.

We shall assume that, for example, the multiphoton ionization is a six-quantum process (this applies to the experimental results on damage at $\lambda\approx 3\ \mu$):

$$1/\tau_i=Aq^6.$$

Figure 2 shows the temperature dependences of the critical intensity in the case under consideration. Curves 1, 2, and 3 correspond to three values of the damage probability ($P=0.1$; 0.5; 0.9) and the dashed curve represents a nondelayed avalanche; curve 4 is the

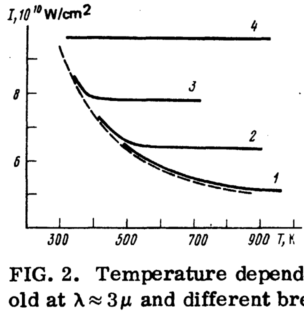

FIG. 2. Temperature dependences of the laser damage threshold at $\lambda\approx 3\mu$ and different breakdown probabilities P: 1) $P=0.1$; 2) $P=0.5$; 3) $P=0.9$. Line 4 corresponds to the case when an avalanche is delayed by a shortage of initiating electrons throughout the investigated temperature range. The dashed curve corresponds to the case of a sufficient number of initiating electrons.

case when the development of an avalanche is stopped by a deficiency of initiating electrons throughout the investigated temperature range.

Curves 1–3 show clearly that there is a range of temperatures in which an avalanche is held up. It is worth noting a considerable increase in the scatter of the threshold characterized by the divergence of curves 1–3. These curves are very close to the results obtained experimentally[4] for the temperature dependence of the damage threshold of NaCl at the wavelength of $\lambda = 2.76\ \mu$. Although this supports our explanation of the anomalous temperature dependence observed in the infrared range of laser frequencies, the final conclusions on the role of initiating electrons can only be made as a result of direct experiments similar to those reported in Ref. 5, i.e., by investigating the influence of additional ultraviolet illumination on the threshold of damage by infrared radiation.

In this case if the proposed avalanche decay mechanism does apply, ultraviolet illumination should produce the following results: at the wavelength of 10.6 μ the threshold should decrease throughout the investigated temperature range up to the usual avalanche threshold; at the wavelength of ~3 μ the threshold should decrease only at relatively high temperatures, where the experiments indicate that the threshold is independent of temperature.

4. DEPENDENCE OF THE BREAKDOWN THRESHOLD ON THE VOLUME OF THE INTERACTION ZONE (SIZE EFFECT)

Experimental investigations of laser damage to optical materials have often demonstrated the "size effect" which is the dependence of the damage threshold on the diameter of the focal spot produced by focusing lenses. This effect is usually attributed to the damage at absorbing inclusions and is explained by a reduction in the probability that sufficiently large inclusions will be found in the caustic region when the size of the caustic is reduced. The delay to the avalanche mechanism discussed here should also produce the size effect. In fact, when the damage threshold is governed by the probability of the appearance of the first electron and Eq. (5) is obeyed, we find that

$$1/\tau_1 = Vf(I),$$

where $f(I)$ is a function governed by the photionization mechanism [for example, in the case of multiphoton ionization we have $f(I)\propto I^k$], we find that for a given damage probability P,

$$Vf(I_{cr}) = \text{const.} \quad (6)$$

In particular, in the case of multiphoton ionization of the matrix, atomic impurities, or defects, we have

$$I_{cr} \propto V^{-1/k}. \quad (7)$$

It should be pointed out that the strongest dependence of the threshold on the dimensions of the caustic is observed when the most effective source of electrons are clusters (of atomic impurities) and dissolved inclusions, because in this case the dependence of the photoionization probability on the intensity may be quite weak ($k \approx 1-3$).

A weaker size effect should be observed when electrons are supplied by impurity levels lying quite deep in the band gap. In the limit of very small caustics the mechanism of creation of free carriers initiating an avalanche may be the multiphoton ionization of atoms of the matrix, i.e., multiphoton interband transitions. It then follows from Eq. (7) that the size effect will be practically unobservable in the infrared range because of the strong dependence of the ionization probability on the radiation intensity. At the same time and for the same reasons the scatter of the damage thresholds in recording the temperature dependence practically disappears (Fig. 2). The fact that the scatter of the thresholds (the difference between curves 1 and 3) and the size dependence are given by the same function $f(I)$ shows that investigations of this dependence will provide a good opportunity for identifying experimentally the nature of sources of initiating electrons.

In conclusion, we must make an important comment on the experimental method for the determination of the damage threshold in the case when avalanches are delayed. It follows from our results that in this case the damage is essentially of statistical nature so that the usual method[3] for the determination of the threshold becomes inadequate. It is necessary to carry out a statistical analysis of the results based on the determination of the damage probability at different illumination intensities. This applies particularly to studies of the size effect.

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