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## Laser induced breakdown of alkali-halide crystals

B. G. Gorshkov, Yu. K. Danileiko, A. S. Epifanov, V. A. Lobachev, A. A. Manenkov, and A. V. Sidorin

*P. N. Lebedev Physics Institute, USSR Academy of Sciences*  
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The frequency and temperature dependences of the laser-induced breakdown thresholds of a number of alkali halide crystals are investigated. In the samples with the highest thresholds the dependences agree qualitatively with the avalanche ionization theory and this mechanism is the probable cause of their destruction. Possible causes of the quantitative discrepancy between theory and experiment are discussed.

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### 1. INTRODUCTION

The mechanism of laser-induced breakdown of solid transparent dielectrics remains to this day a complicated and debatable problem in the physics of the interaction between high-power electromagnetic radiation and matter. For pure materials, in the case of pulses in the picosecond range, the most widely discussed mechanism is the electron avalanche due to impact ionization.<sup>[1]</sup> It is still unclear, however, whether this mechanism predominates in the breakdown of real materials, since the latter always contain various kinds of defects and inclusions that can influence strongly the breakdown process.

It was stated in<sup>[2-5]</sup> that the electron-avalanche mechanism was realized in the alkali-halide crystals investigated by the authors. The proof advanced in the cited papers, however, can hardly be regarded as convincing. In fact, the statements there were based mainly on the following observed experimental facts: a) the independence of the laser-breakdown threshold of the frequency

in a wide range, and the fact that these thresholds agreed with those observed in constant electric fields; b) the correlations of the thresholds in the series of the alkali-halide crystals. It was also stated that no effects of the absorbing inclusions were accounted for in the experiments. All this, however, cannot be regarded as fully convincing proof in the favor of a breakdown mechanism that is connected with the development of an electron avalanche. In fact:

1. The independence of the thresholds of the frequency may also be a property of other breakdown mechanisms, including the thermal one. And if this fact is analyzed within the framework of the electron avalanche mechanism, then it means that the relaxation time  $\tau$  of the longitudinal component of the momentum of the "hot" electrons turns out to be shorter than  $6 \times 10^{-16}$  sec, which is, in the very least, unexpected and requires careful corroboration. The comparison that was carried out between the thresholds of laser breakdown and

of dielectric breakdowns in constant fields is not quite correct, since the experiments were performed in these two cases under conditions that are difficult to compare with one another. In addition, the question of the breakdown mechanism in a constant field has not been resolved unequivocally.<sup>[6]</sup>

2. A correlation of the breakdown thresholds in the series of alkali-halide crystals can be observed also in the case of the thermal breakdown mechanism, since the same sequence is followed also by the changes of such parameters as the heat capacity, the thermal conductivity, and the melting temperature, which determine the thermal endurance of the crystals.

3. The conclusion that the absorbing inclusions exert no influence was based only on a study of the morphology of the damage, which is patently insufficient.

The information obtained in<sup>[2-5]</sup>, in our opinion, is therefore insufficient to draw reliable conclusions concerning the role of avalanche ionization in laser-induced breakdown of solid dielectrics.

As shown in<sup>[7]</sup>, the most distinctive features of the electron-avalanche mechanism are the frequency and temperature dependences of the damage thresholds, and these dependences are closely connected with each other; the temperature dependence is essentially determined by the field frequency  $\Omega$ , or more accurately by the quantity  $\Omega\tau$ . These relations were investigated by us<sup>[8]</sup> for a number of alkali-halide crystals at neodymium- and ruby-laser frequencies, were found to vary strongly from crystal to crystal, and did not conform at all to the predictions of the theory of avalanche ionization. It appears that these variations mean that the mechanism of "pure" avalanche ionization was not realized even in crystals whose threshold fields greatly exceeded the constant critical fields.

In the present study, to assess in detail the role of impact ionization in the damage of transparent dielectrics, we have investigated the connection between the frequency and temperature dependences of the damage thresholds of a large number of samples of alkali-halide crystals in a wide range of laser-radiation frequencies.

The investigations were made on the crystals NaCl, KCl, KBr, LiF and NaF at the frequencies of the CO<sub>2</sub> laser (wavelength  $\lambda = 10.6 \mu\text{m}$ ), neodymium laser (1.06  $\mu\text{m}$ ), and ruby laser (0.69  $\mu\text{m}$ ), and also at the second harmonic of a neodymium-garnet laser ( $\lambda = 0.53 \mu\text{m}$ ) in the temperature interval from 100 to 800 K.

## 2. EXPERIMENTAL PROCEDURE AND CONDITIONS

The laser damage of alkali-halide crystals was investigated using the following single-mode lasers:

1) CO<sub>2</sub> laser with transverse discharge. The fundamental transverse mode was separated by using an unstable semi-concentric resonator,<sup>[9]</sup> in which the total-reflection mirror was a concave spherical metallic mirror with curvature radius  $R = 5 \text{ m}$ , and the output mirror was a silicon plate. The distance between mirrors was somewhat more than 5 meters and was adjusted experimentally. The short lasing pulse was shaped by us-

ing a CO<sub>2</sub>-He working mixture without adding N<sub>2</sub>, and by using a low-inductance discharge-supply circuit. The result was a smooth bell-shaped radiation pulse of near-Gaussian form and of duration  $\sim 60 \text{ nsec}$  at half the maximum power.

2) YAG:Nd<sup>3+</sup> laser with passive Q-switching. The selection of the transverse and longitudinal modes, which ensured a single-frequency lasing regime, was effected by using a standard scheme, viz., by placing a small-diameter ( $d \approx 1 \text{ mm}$ ) diaphragm and a quartz stack in a resonator with flat mirrors. The duration of the smooth bell-shaped pulse was 10 nsec.

3) A ruby laser, the arrangement and space-time characteristics of which were perfectly analogous to those given for the YAG:Nd<sup>3+</sup>.

4) A YAG:Nd<sup>3+</sup> laser with a frequency doubler using a lithium iodate crystal.

The output energy was measured with an IKT-1M calorimeter or with a graphite calorimeter checked against the former. Particular attention was paid to the study of the spatial distribution of the intensity in the caustics of the lenses used to focus the radiation into the interior of the investigated samples. We used lenses of focal lengths  $f = 100, 38, \text{ and } 15 \text{ mm}$  for the CO<sub>2</sub> laser and at the other frequencies, respectively. For a correct determination of the beam radius at the minimal section of the lens caustic we used the following two methods,<sup>[10,11]</sup> which yielded identical results.

a) A light-struck photographic film was placed in the focal plane of the lens in such a way that only the very maximum of the spatial distribution of the radiation was printed on the film. The power was then gradually increased, producing spots of larger diameter with sufficiently sharp boundary. From the measured values of the spot diameters and of the radiation power at the entrance to the lens we plotted the distribution of the intensity in the cross section of the beam.

b) A razor blade was displaced in discrete steps along the  $y$  coordinate in the focal plane in such a way that it covered partly the laser beam. The transmitted fraction  $G$  of the radiation energy was recorded with a photomultiplier. The radiation was attenuated beforehand with filters to prevent production of a plasma on the edge of the blade. The  $G(y)$  curve obtained in this manner was used to calculate the spatial distribution of the radiation intensity in the lens caustic.

Measurements by the two described methods have shown that the intensity distribution in the cross sec-

TABLE I. Diameters of the caustics of the employed lenses,  $\mu\text{m}$ .

$f, \text{ mm}$	$\lambda, \mu\text{m}$			
	0.53	0.69	1.06	10.6
15	4.0	3.6	4.6	—
38	11	10	12	—
100	—	—	—	51

tions of the caustics of all the employed lenses is close to Gaussian at all the employed laser frequencies. The dimension of the caustics practically coincide in this case with the diffraction limit, as demonstrated by the absence of noticeable lens aberrations. Table I lists the measured caustic diameters (at the level  $1/e$  of the maximum intensity).

To prevent breakdown from occurring on the surfaces of the investigated samples, the radiation was focused into the volume of the crystals with a lens of  $f=15$  mm to a depth of 1 mm, with  $f=38$  mm to a depth 5 mm, and with  $f=100$  to a depth 10 mm. Estimates show that at the indicated distances from the lens foci to the sample surfaces the aberrations due to the surface are small, and the focal-spot radius measured in air remains practically constant when the radiation is focused in the interior of the sample. This circumstance, and also the fact that the inhomogeneity of the refractive index of the crystal did not affect the radiation intensity at the focus, were confirmed by an experiment in which the damage thresholds remained unchanged when the depth of focusing in the interior of the samples was varied.

The damage was revealed visually by a spark observed through a light filter that cut off the main illumination and by the scattering in the beam of a helium-neon laser, and was investigated with a microscope. We note that the damage observed under the microscope in samples having sufficiently high thresholds always occurred simultaneously with the appearance of the spark and of the scattering. By critical (threshold) intensity we mean here the minimal peak laser intensity, at the center of the lens caustic, necessary to produce damage in a single flash.

### 3. RESULTS OF EXPERIMENTS

In the investigation of the temperature dependences of the damage thresholds, we have observed the following effect. After a heat-treatment cycle consisting of heating, soaking for a certain time ( $\sim 30$  min) at a temperature  $T'$ , and cooling (with a time constant  $\sim 5$  min), most NaCl, KCl, and KBr crystals had appreciably higher damage thresholds at the emission frequencies of the ruby and neodymium lasers and at the

second harmonic of the latter. This effect is most noticeable in the case of crystals having low initial thresholds. On the whole, the scatter of the thresholds from sample to sample, using the indicated heat treatment, decreased appreciably, and crystals of different quality acquired an optical quality closer to that of the better ones. Table II lists some data on the variation of the thresholds as a result of heat treatment at various temperatures  $T'$ . It is seen that appreciable strengthening sets in if  $T'$  is close to the lattice melting temperature  $T_{\text{melt}}$ .

We note that among the crystals that increase greatly in optical strength as a result of the heat treatment there are also some (see, e.g., the data for KCl and KBr in Table II) whose initial thresholds are close to those measured by Fradin *et al.*<sup>[3,4]</sup> This circumstance casts doubts on the conclusion drawn by Fradin *et al.*, that an intrinsic damage mechanism was realized in their experiments.

In the investigation of crystal damage at the  $\text{CO}_2$ -laser frequency, no noticeable effect of heat treatment on the damage thresholds was recorded.

To check on the possible existence of a correlation between the change in the laser-induced damage thresholds and the optical characteristics of the crystals we measured the scattering (at  $\lambda=0.63 \mu\text{m}$ ) and the absorption of light (at  $\lambda=1.06 \mu\text{m}$ ), and also the spectrum of the transmission of the samples in the range  $\lambda=0.2-0.8$  nm before and after the heat treatment. The results are listed in Table II. It is seen that no definite correlation whatever is observed between the optical characteristics and the damage thresholds.

The following fact is also of interest: The results obtained in the study of the temperature dependences of the damage threshold of crystals subjected to preliminary heat treatment were not reproducible. When a certain temperature interval was passed through slowly, the damage threshold decreased. In heat treatment consisting of heating to a temperature close to  $T_{\text{melt}}$  followed by slow cooling (the cooling rate in the interval from  $T_{\text{melt}}$  to 400 K was  $\sim 50$  deg/hr), the endurance of the previously strengthened crystals decreased to the initial value. Naturally, effects of this kind can greatly influence the observed temperature dependences of the damage thresholds. An attempt was therefore made to find samples not affected by heat treatment.

By careful selection, we found such samples of NaCl, KCl, and KBr crystals, and it turned out that they had the highest damage thresholds, just as the best heat-treated crystals. These crystals were used to study the temperature dependences of the damage thresholds at various laser-radiation frequencies (Fig. 1).

In addition, we investigated the temperature dependences of the damage thresholds at  $\lambda=0.69 \mu\text{m}$  and  $\lambda=1.06 \mu\text{m}$  of the crystals NaF and LiF. It was observed that the thresholds of these crystals do not depend on the temperature in the interval 300–900 K and amount respectively to  $1.4 \times 10^{11}$  and  $3.6 \times 10^{11}$  W/cm<sup>2</sup> at both frequencies. We note that such high values of

TABLE II. Effect of heat treatment on the damage threshold and on the optical characteristics of the crystals\*

Material	$T', ^\circ\text{C}$	$W \cdot 10^{-10}$ at $\lambda=1.06$	$J$ at $\lambda=0.63$	$k$ at $\lambda=0.25$	$\alpha \cdot 10^4$ at $\lambda=1.06$
NaCl I ( $T_{\text{melt}} = 801^\circ\text{C}$ )	—	1.0	4	0.12	1.7
	680	1.1	2	0.15	—
	730	1.5	2	0.14	—
	800	6	2.5	0.18	2.0
NaCl II	—	12	730	0.4	1.3
	800	12	130	0.11	1.3
KCl ( $T_{\text{melt}} = 776^\circ\text{C}$ )	—	0.5	420	0.32	4.0
	680	0.9	100	0.22	—
	770	2.5	120	0.22	4.5
KF ( $T_{\text{melt}} = 730^\circ\text{C}$ )	—	0.25	16	0.24	2.0
	730	3	2.5	0.24	1.7

\* $T'$ —heat-treatment temperature,  $W$  [W/cm<sup>2</sup>]—damage threshold,  $J$  (rel. un.)—scattering intensity,  $k$  [cm<sup>-1</sup>]—extinction coefficient,  $\alpha$  [cm<sup>-1</sup>]—absorption coefficient; the values of  $\lambda$  are in microns.

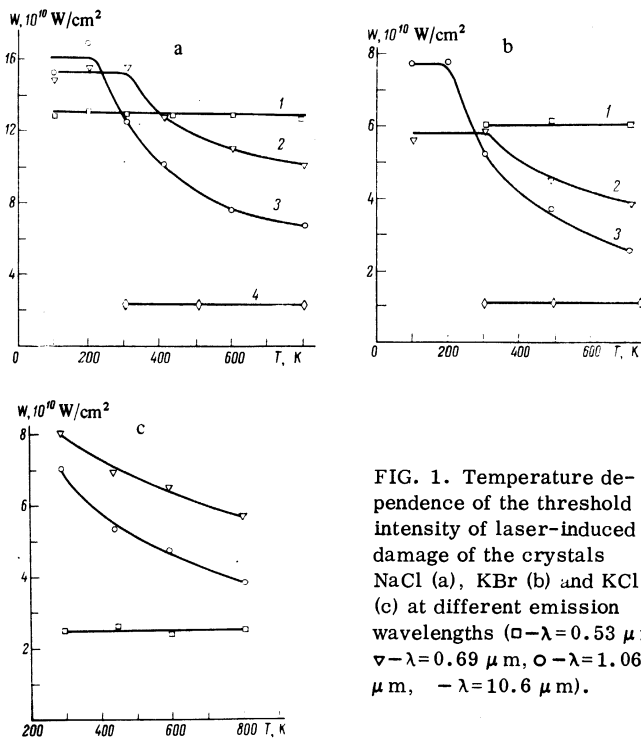


FIG. 1. Temperature dependence of the threshold intensity of laser-induced damage of the crystals NaCl (a), KBr (b) and KCl (c) at different emission wavelengths ( $\square - \lambda = 0.53 \mu\text{m}$ ,  $\nabla - \lambda = 0.69 \mu\text{m}$ ,  $\circ - \lambda = 1.06 \mu\text{m}$ ,  $- \lambda = 10.6 \mu\text{m}$ ).

the optical strength were obtained for exceptional samples.<sup>1)</sup>

The experimental results shown in Fig. 1 indicate that in specially selected NaCl, KCl, and KBr crystals the character of the dependences agrees qualitatively with the predictions of the avalanche ionization theory.<sup>[1,7]</sup> For a correct comparison of the results of the experiments with the theory it must be borne in mind, however, that the diffusion approximation used in<sup>[1,7]</sup> may not be valid at high electromagnetic-radiation frequencies. Therefore, before proceeding to a discussion of the results, we examine the features of avalanche development under the influence of radiation in the visible region of the spectrum.

#### 4. THEORY

We consider the process of the development of an electron avalanche in the case when

$$\hbar\Omega > \langle \epsilon \rangle, \quad (1)$$

where  $\langle \epsilon \rangle$  is the average electron energy in the conduction band.

The differential-difference equation describing this process can be derived from the general quantum-kinetic equation for the electrons of the conduction band,<sup>[12]</sup> by using the customary expansion of the distribution function in the displaced point:

$$\tilde{f}(\epsilon + n\hbar\Omega \pm \hbar\omega) \approx \tilde{f}(\epsilon + n\hbar\Omega) \pm \hbar\omega f'_\epsilon(\epsilon + n\hbar\Omega) + 1/2(\hbar\omega)^2 f''_\epsilon(\epsilon + n\hbar\Omega), \quad (2)$$

where  $\hbar\omega$  is the phonon energy,  $n = 0, 1, \dots$

In the frequency region of interest to us the parameter

$$H = eE\Delta p / \hbar m \Omega^2$$

( $e$  and  $m$  is the charge and effective mass of the electron,  $\mathbf{E}$  is the amplitude of the electric field, and  $\Delta p$  is the change of the electron momentum as a result of the electron-phonon collision), which characterizes the role of the interband multiphoton processes, turns out to be less than unity, and therefore, to avoid excessively cumbersome formulas, we shall take into account below only single-photon transitions.

After introducing the dimensionless energy

$$x = \epsilon / I,$$

where  $I$  is the effective ionization potential,<sup>[13]</sup> the differential-difference equation takes in the parabolic-band approximation the following form ( $x_0 = \hbar\Omega / I$ ):

$$\begin{aligned} \bar{v}_x \frac{\partial f(x, t)}{\partial t} = \frac{\partial}{\partial x} \{ \bar{v}_x [D_0(x) f'_x(x, t) + Q_0(x) f(x, t)] \} \\ + w_1(x) f(x + x_0) + w_2(x) f(x - x_0) - [w_1(x) + w_2(x)] f(x) + w_{11}(x) f'_x(x + x_0) \\ + w_{21}(x) f'_x(x - x_0) + w_{12}(x) f''_x(x + x_0) + w_{22}(x) f''_x(x - x_0). \quad (3) \end{aligned}$$

We introduce in addition the following notation:  $v_s$  is the speed of sound,  $\rho$  is the density,  $\epsilon_1$  is the energy constant of the deformation potential,  $T$  is the lattice temperature, and  $l_{ac}$  is the mean free path of the electrons in deformation scattering by acoustic phonons.

For the case of predominant scattering the phonons of the acoustic branch we have in the high-temperature approximation:

$$\begin{aligned} D_0(x) = \frac{4v_s^2 m}{(2mI) l_{ac}} x, \quad Q_0(x) = \frac{I}{kT} D_0(x), \quad l_{ac} = \frac{\pi \rho \hbar^2 v_s^2}{m^2 \epsilon_1^2 kT}; \\ w_1(x) = \frac{4I\alpha}{(2mI) l_{ac}} x^2 \left(1 + \frac{x_0}{x}\right)^{1/2} \left(1 + \frac{x_0}{2x}\right), \\ w_2(x) = \begin{cases} \frac{4I\alpha}{(2mI) l_{ac}} x^2 \left(1 - \frac{x_0}{x}\right)^{1/2} \left(1 - \frac{x_0}{2x}\right), & x \geq x_0 \\ 0, & x < x_0 \end{cases} \\ w_{11}(x) = \frac{I}{6kT} x w_0(u_+^e - d_+^e) + w_0 \frac{u_+^e - d_+^e}{(1 + x_0/x)^{1/2}}, \\ w_{21}(x) = \begin{cases} \frac{I}{6kT} x w_0(u_-^e - d_-^e) + w_0 \frac{u_-^e - d_-^e}{(1 - x_0/x)^{1/2}}, & x \geq x_0 \\ 0, & x < x_0 \end{cases} \\ w_{12}(x) = 1/6 x w_0(u_-^e - d_-^e), \\ w_{22}(x) = \begin{cases} 1/6 x w_0(u_-^e - d_-^e), & x \geq x_0 \\ 0, & x < x_0 \end{cases} \\ w_0 = \frac{m v_s^2 \alpha x^2}{(2mI) l_{ac}}, \quad \alpha = \frac{e^2 E^2 I}{6(\hbar\Omega)^2 m \Omega^2}, \\ u_\pm = 1 + (1 \pm x_0/x)^{1/2}, \quad d_\pm = \mp 1 \pm (1 \pm x_0/x)^{1/2}. \end{aligned}$$

It is interesting to note that Eq. (3) has distinctive terms containing  $f'(x \pm x_0)$  and  $f''(x \pm x_0)$ . Their appearance is connected with the fact that the triple electron-phonon collision cause the electron to go over into the state with energy  $\epsilon$  not from a state with  $\epsilon \pm \hbar\Omega$  (this process would be described only by terms containing  $f(x \pm x_0)$ ), but from a state with an energy shifted additionally by an amount  $\hbar\omega$ , which is the phonon energy.

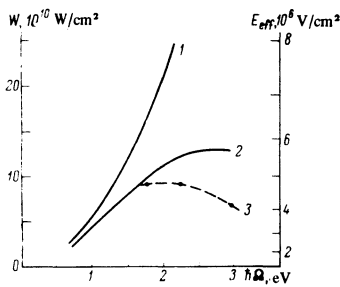


FIG. 2. Theoretical plots of the damage-threshold intensity of NaCl crystals vs. the radiation frequency at  $T = 300$  K: 1) in the diffusion approximation, 2, 3) from the differential-difference equation: 2) using relation (7), 3) numerical calculation.

As shown by estimates, the corrections that must be introduced in the critical field because of the presence of these terms in (3) turn out to be small, and will therefore be disregarded.<sup>2)</sup>

The free-electron concentration  $N$  increases in the case of avalanche ionization exponentially,  $N = N_0 e^{\gamma t}$ , and  $\partial f(\epsilon, t)/\partial t = \gamma f(\epsilon, t)$ .

We note that when condition (1) is satisfied we can neglect in (3) the terms containing  $f(x + x_0)$ , so that

$$f(x + x_0) \ll f(x) \ll f(x - x_0). \quad (4)$$

The latter inequality obviously takes place at  $x > x_0$ . The inequalities (4) allow us to obtain the quasistationary solution of Eq. (3) by the method of<sup>[12]</sup> if

$$x_0 \ll 1, \quad (\hbar\Omega < I/3), \quad (5)$$

with the aid of the following procedure. We obtain the solution of the equation in the region  $0 < x \leq x_0$ , we use the solution to obtain the distribution function in the region  $x_0 < x \leq 2x_0$ , etc., and later on join together the functions at the points  $nx_0$ ,  $n = 1, 2, \dots$ . The resultant relations for the avalanche-development constant  $\gamma$  are in this case quite cumbersome and are not illustrative. Numerical calculations, on the other hand, show that not only qualitatively but also quantitatively satisfactory results can be obtained from an approximate analysis of the differential-difference equation. For such an analysis, we put

$$\bar{\gamma} D_0(x) \approx 4v_s^2 m / (2mI)^{1/2} l_{ac}, \quad \bar{\gamma} Q_0(x) \approx 4v_s^2 m I / kT (2mI)^{1/2} l_{ac}, \\ w_1(x) \approx 4I\alpha / (2mI)^{1/2} l_{ac}, \quad w_2(x) = w_1$$

and introduce the notation

$$q = e^2 E^2 / 6m^2 v_s^2 \Omega^2, \quad q' = \delta q / x_0^2, \quad \delta = kT / I.$$

The resultant equation with constant coefficients has a stationary solution in the form

$$f(x) = A e^{-x/(s+1)\delta}, \quad (6)$$

the parameter  $s$  being determined from the equation

$$\frac{s}{(1+s)^2} = 2q'\delta \left( \text{ch} \frac{x_0}{(1+s)\delta} - 1 \right), \quad (7)$$

and the quantity  $(s+1)T$  playing the role of the effective electron temperature. We note that at

$$x_0 / (1+s)\delta \ll 1 \quad (\hbar\Omega \ll kT_s = (s+1)kT)$$

the obtained solution (6) goes over directly into the solution of the diffusion equation<sup>[13]</sup> with variable coefficients. This circumstance allows us to find the corrections that must be added to the critical field because of the large value of the light quantum, since it makes known the ratio

$$\xi = kT_s / I,$$

at which the damage sets in as a result of the development of an electron avalanche.<sup>[13]</sup> Thus, to damage NaCl crystals by laser radiation of duration  $10^{-8}$  sec we need  $\xi \approx 0.08$ .

We note some substantial results that follow from relation (7) which, if  $\xi$  is known, is in fact the equation for the determination of the critical field.

1. The temperature dependence of the damage threshold remains the same as in the diffusion approximation, and in particular

$$E_{cr} \propto T^{-1/2}$$

for the temperature region in which the high-temperature solution is valid ( $T > 250$  K for NaCl crystals), if

$$\Omega\tau \gg 1.$$

2. The frequency dependence turns out to be much weaker than in the diffusion approximation ( $E_{cr} \propto \Omega$  at  $\Omega\tau \gg 1$ ), and if

$$\hbar\Omega > 4kT_s,$$

then the damage threshold may even decrease with increasing electromagnetic-field frequency.

Figure 2 shows the theoretical dependence of the damage threshold on the radiation frequency for NaCl crystals at  $T = 300$  °K. In the calculations we assumed the following values of the parameters:  $m = m_e$  (the mass of the free electron),  $I = 10$  eV,  $v_s = 4 \times 10^5$  cm/sec,  $l_{ac} = 2 \times 10^{-7}$  cm. The threshold intensities were obtained from the relation

$$W = 2.6 \cdot 10^{-3} n E_{eff}^2 \quad (E_{eff} = E/\sqrt{2}),$$

where  $n$  is the refractive index, and  $W$  and  $W_{eff}$  are expressed respectively in  $W/cm^2$  and in  $W/cm$ . It is seen from Fig. 2, in particular, that the critical field is maximal at a quantum energy  $\hbar\Omega \sim 2$  eV ( $\lambda \sim 0.5$   $\mu$ m).

3. Inasmuch as at a quantum energy  $\sim 1$  eV the more probable process is impact ionization with simultaneous absorption of a light quantum, the effective ionization potential in this frequency region is

$$I \approx \epsilon_g - \hbar\Omega,$$

where  $\epsilon_g$  is the width of the forbidden band. At the neodymium-laser frequency, the critical intensity is additionally decreased as a result of this effect by an approximate factor 1.5.

It must be noted that Eq. (7) can be used only if condition (5) is satisfied, for otherwise an important role is assumed in the formation of the distribution function by the electrons produced as a result of impact ioniza-

tion. This frequency region, however, seems to be of no particular interest from the point of view of the investigation of the properties of the electron avalanche, since the most probable cause of the damage at such large radiation quanta will be two- or three-phonon ionization processes.

## 5. DISCUSSION OF RESULTS

The experimental investigation of the frequency and temperature dependences of the laser-damage thresholds of a large number of samples of alkali-halide crystals has shown that for most crystals these dependences exhibit no singularities typical of the avalanche mechanism. Of particular interest, however, are crystals specially selected in accordance with the following two attributes:

- a) the highest and most reproducible damage threshold;
- b) independence of the optical endurance of the heat treatment.

NaCl, KCl, and KBr crystals possessing these properties have thresholds whose frequency and temperature dependences agree qualitatively with the theoretical ones obtained for damage as a result of the development of an electron avalanche at the frequencies of CO<sub>2</sub>, neodymium, and ruby lasers. There is, however, a quantitative disparity between theory and experiment. Thus, if it is assumed that the absence of a temperature dependence of the damage threshold at a wavelength 10.6 μm is due to the approximate equality of the CO<sub>2</sub>-laser frequency to the effective frequency  $\nu_{eff}$  of the electron-phonon collisions, then we obtain for the latter (at  $T = 300$  K)

$$\nu_{eff} \approx 1.2 \cdot 10^{14} \text{ sec}^{-1}.$$

On the other hand, an analysis of the frequency dependence leads to the value

$$\nu_{eff} \approx 6 \cdot 10^{14} \text{ sec}^{-1}.$$

To explain this disparity, we can advance two hypothetical explanations.

- 1) The avalanche-ionization theory was developed under the assumption that the electrons in the conduction bands have a parabolic dispersion law. However, owing to the complex structure of the real conduction bands of dielectrics, and the fact that they possess subbands, rather effective direct transitions between the subbands may be turned on, going from the CO<sub>2</sub>-laser frequency to the neodymium-laser frequency and above, with absorption of electromagnetic-radiation quanta. These processes, obviously, can lower appreciably the observed damage threshold if  $\hbar\Omega \gtrsim 1$  eV, and thereby decrease the effective ionization potential.

- 2) Atomic impurities and defects can exert just the opposite effect on the development of the avalanche ionization at the relatively low (CO<sub>2</sub> laser) and high (neodymium and ruby laser) frequencies. The electrons of

the conduction bands collide with the impurity atoms and excite the latter. If the radiation quantum is sufficient for the ionization of the excited atoms, then the damage threshold will be lowered, and in the opposite case the development of the electron avalanche may be halted.

This is also the possible cause of the influence of heat treatment on the damage threshold. When the crystals are slowly cooled, the atomic defects and impurities can form local clusters with a concentration  $\geq 10^{18} \text{ cm}^{-3}$ , which is sufficient for the development of an electron avalanche on the impurities at an electromagnetic-radiation quantum energy  $\geq 1$  eV. If the cooling is rapid, on the other hand, the impurities and defects are more uniformly distributed, and this raises the damage threshold. At the lower radiation frequencies (CO<sub>2</sub> laser) the influence of the impurities and of the defects, as noted above, can in principle be different, and from this point of view it becomes clear why heat treatment influences the optical endurance only at sufficiently high field frequencies.

Thus, the presence of characteristic frequency and temperature dependences of the laser-damage threshold of specially selected samples of the alkali-halide crystals NaCl, KCl, and KBr shows that their damage via an electron-avalanche development is probable, in contrast to most ordinary samples of the same crystals.

The most difficult to explain is the absence of a temperature dependence of the damage threshold at the second harmonic of a neodymium laser. It is possible that the decisive role is played in this case either by direct ionization of the impurities or by multiphonon absorption processes.

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<sup>1</sup>These crystals were grown by G. F. Dobrzhanskii at the Crystallography Institute of the USSR Academy of Sciences.

<sup>2</sup>The kinetic-equation terms containing  $f'(x \pm x_0)$  and  $f''(x \pm x_0)$  may turn out to be significant in relatively weak fields and at sufficiently high phonon energies. In that case, however, the use of Eq. (3) may not be justified at all, since the expansion (2) becomes meaningless.

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# Tunnel effect in superconductors with nonequilibrium quasi-particle population under laser irradiation

O. P. Balkashin, I. K. Yanson, and A. V. Khotkevich

*Physico-technical Institute of Low Temperatures, Ukrainian Academy of Sciences*

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Small deviations of the volt-ampere characteristics of laser-irradiated Pb-PbO-Pb and Sn-SnO-Sn tunnel junctions from equilibrium are measured. The decrease of the junction conductivity due to nonequilibrium occupation of the quasi-particle states is investigated in detail for various temperatures and laser-radiation intensities. The tunnel current is calculated for a special model with a nonequilibrium excitation distribution function. The results of the calculation are in good agreement with the experiments. The role of thermal effects is analyzed.

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## 1. INTRODUCTION

A stationary energy distribution of the quasiparticles and phonons, which differs considerably from the equilibrium distribution, is established at sufficiently low temperatures ( $kT \ll \Delta$ ) and under the influence of laser radiation in thin superconducting films.<sup>[1]</sup> The properties of the superconductor in such a nonequilibrium state are determined both by the total number of excess excitations and by the actual form of their energy distribution. However, experiments carried out to date have been limited principally to measurements of such integrated characteristics as the energy gap,<sup>[2]</sup> the dc resistance,<sup>[3]</sup> or the reflection coefficient at microwave frequencies.<sup>[4]</sup> Therefore, their results can be sufficiently well explained by various model-dependent distribution functions<sup>[5,6]</sup> under the condition that the concentration of the nonequilibrium excitations is small and that their total number remains unchanged.

A decrease in the conductivity of a laser-irradiated tunnel junction was first observed experimentally in Ref. 7 in a narrow range of voltages  $eV \geq 2\Delta$ , the reason for this decrease is the occupation of part of the states over the energy gap by photo-excited nonequilibrium quasiparticles. The tunnel effect allows a direct measurement of the energy localization of the nonequilibrium occupation and, consequently, yields some information on the nonequilibrium contribution to the distribution function of the quasiparticles.

In the present we report here an experimental study of the behavior of the tunnel junctions of lead and tin irradiated by an He-Ne laser. The measurements were performed at various temperatures and radiation inten-

sities. The calculation of the volt-ampere characteristics of the junctions was performed with the use of the nonequilibrium quasiparticle distribution function proposed by Vardanyan and Ivlev<sup>[6]</sup> and agrees well with the experimental results.

## 2. EXPERIMENTAL METHOD

The measurements were carried out on Pb-PbO-Ob and Sn-SnO-Sn tunnel junctions prepared by the usual techniques (condensation in a vacuum and oxidation in a glow discharge). The area of the junctions amounted to 0.15-0.06 mm<sup>2</sup>, the resistance was 1-15 ohms. The thickness of the metallic films was within the range 1000-2000 Å. Crystalline quartz or sapphire was used as a substrate; in the initial states of the research, glass was also used. The sample was placed directly in liquid helium.<sup>[1]</sup>

Radiation from an He-Ne laser of wavelength 1.15 μ was chopped at a rate of 420 Hz by a disc modulator (Fig. 1), guided into the cryostat by a light pipe, and focused with the help of a cone on the upper film of the sample in the form of a spot with a diameter of 2.2 mm. Part of the radiation was diverted to a calibrated photodetector to record the power level in the measurement process. The power was regulated by an iris diaphragm and by grid attenuators. The optical system was tuned against the red emission line as the laser resonator tuning was varied. To prevent heating and formation of a spatially inhomogeneous nonequilibrium state in the film,<sup>[3]</sup> relatively low levels of radiation intensity were used.

After amplification and synchronous detection, the