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Dynamics of laser damage in KDP crystals

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Photoresponse investigation has revealed the dynamics of laser damage in KDP crystals. It is established that in sufficiently pure crystals the damage develops in the volume at the speed of sound and terminates in formation of cracks characterized by a substantial surface-charge density. Characteristics of the damaged region, such as pressure, temperature, absorbed-energy density, and absorption coefficient are determined. The role played by the pressure wave in the process of damage to dielectrics by nanosecond laser pulses is discussed.

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1. INTRODUCTION

The mechanisms whereby materials are optically damaged have been intensively studied for more than a decade, and it has been established by now that the development of sizeable (10^{-3}–10^{-2} cm) damage in pure substances is not the direct consequence of heating of the absorbing inhomogeneities, but is due to the absorption by the medium itself under the influence of the laser radiation. [A substance is regarded is here as pure if the absorbing inclusions are small enough and their heating cannot cause directly any noticeable damage in the medium, the distance between the inclusion being $l_0 \gg (\tau_p \chi)^{1/2}$, where τ_p is the duration of the laser pulse and χ is the thermal diffusivity of the medium.] The nature of this absorption and the mechanisms of its onset in real crystals are still the subjects of debates. Many papers have dealt with absorption of laser by free carriers, whose appearance is attributed, for example, to a thermal absorption wave[1] or to ultraviolet preionization of the matrix.[2] These mechanisms differ substantially both in the values of the parameters (e.g., the matrix temperature) needed for their realizations, and in the rate of propagation of the absorption produced by them. One should therefore expect an analysis of the dynamics of damage development to cast light on the roles of the different mechanisms in the breakdown.

Most frequently, attempts to track the dynamics of the damage are made by studying the emission that accompanies the damage, and the scattering of the light from the damaged region (see, e.g., Refs. 3 and 4). Experiments have shown, however, that both the light and the scattering appear during later stages of the damage, frequently after the termination of the laser pulse.[1)] It seems promising therefore to investigate the damage dynamics by using the photoelectric response. In fact, the large amount of information provided by this method has been well demonstrated with photoelectric spectroscopy of semiconductors as the example, but the methods developed for semiconductors cannot be directly applied to dielectrics. Owing to the low volume conductivity, the surface and contact phenomena in dielectrics are relatively strong and impede seriously the interpretation of the results, especially in the study of damage. This may be the reason why hardly any investigations of photoconductivity in transparent have been made until now, while Belikova *et al.*[6] attributed the damage in corundum, whose strength is 10^{10} W/cm^2, to the formation of an electron avalanche, an assumption that does not seem to be reliable enough.[2)]

We report below the results of an investigation of the dynamics of the photoresponse in damage of KDP crystals, which were used as an example to develop a technique[8] that eliminated the influence of contact and surface phenomena. It was established with the aid of this technique that the generation of free carriers and nonstationary heating of the crystal lattice produce photoresponses of opposite polarity, J_σ and J_ε, respectively. The current

$$J_\sigma \sim \int_{V_0} \sigma(t,r)\,dV$$

(σ is the light-induced conductivity in the volume V_0) corresponds to an increase of the sample conductivity, while the thermoelectric current

$$J_\varepsilon \sim P_{abs}\, d\varepsilon/dT$$

(P_{abs} is the radiation power absorbed in the sample, ε is the low-frequency permittivity of the crystal, and T is the lattice temperature) decreases the conductivity.

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In addition, nonstationary heating gives rise to generation of sound waves that introduce into the photoresponse current pulses $J_{ac} \sim W_{abs}$, where W_{abs} is the radiation energy absorbed in the sample.

It must be emphasized that in KDP the currents J_σ and J_ε have opposite polarities, so that it is possible to separate reliably in the experiments the processes connected with the lattice heating from those due to generation of free carriers.

2. EXPERIMENT AND ITS RESULTS

The experimental setup is illustrated in Fig. 1. To study the damage induced by the light in the volume we used a neodymium-laser system ($\lambda = 1.06$ μm) in which the driving generator produced single pulses and was Q-switched by a saturable filter. (Surface damage was produced and led to a large-amplitude photocurrent due to the appearance of free carriers in the surface region. The results reported below pertain only to volume damage.) The radiation pulse waveform was close to Gaussian with duration $\tau_p = 4 \times 10^{-8}$ sec at half-maximum and with a single longitudinal and a single transverse (zero) mode. The output radiation was focused by a lens of focal length 70 cm, and the investigated z-cut samples with dimensions $5 \times 5 \times 10$ mm were placed 60 cm away from the lens. The diameter of the light beam was practically constant over the length of the crystal and equal to 300 μm at the half-intensity level. Just as in Ref. 8, we used an external voltage $U_0 \approx 2 \times 10^3$ V of frequency $F = 50$ Hz, synchronized with the laser in such a way that the lasing pulse coincided with the maximum voltage on the sample. To prevent photocarrier injection from the metallic contacts (for example, when the contacts were illuminated by stray light), a teflon ribbon was placed between the electrodes and the crystal, and in front of the sample we placed a diaphragm of 2 mm diameter, which prevented the contacts from being directly illuminated by the spatial wings of the laser pulse.

To exclude the contribution made to the photoresponse by the surface photoconductivity, the illuminated surfaces of the investigated samples were purified (photodesorbed). This was done by repeated prior irradiation with laser pulses having a power density somewhat lower than the optical-damage threshold p^*, and by

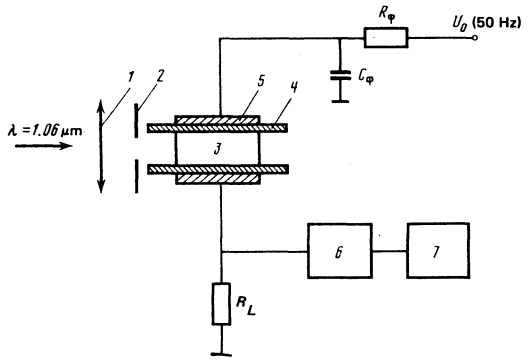

FIG. 1. Photoconductivity measurement setup: 1—lens, 2—diaphragm, 3—crystal, 4—teflon ribbon, 5—metallic contacts, 6—amplifier, 7—S1-11 oscilloscope.

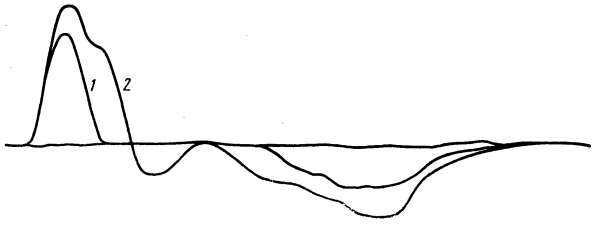

FIG. 2. Oscillograms of photoresponses: 1—following the action of a damaging laser pulse with $p = 0.9^*$, 2—following the action of a damaging pulse with $p \approx p^*$.

gradually increasing the power as the threshold was approached (the power increment from pulse to pulse did not exceed 10%). After this treatment, the photoresponse was independent of the external electric field near the illuminated surfaces of the sample.

The damages were observed and their morphology studied with a microscope of 100× magnification, which showed a clear image of the surfaces as well as of any section in the interior of the sample. The photoresponse was recorded with a system having a sensitivity $J_{min} \approx 5 \times 10^{-8}$ A and a time resolution $\tau_{st} \lesssim 10^{-8}$ sec.

The study of the dynamics of the laser damage and of the interaction of the radiation with the substance in the course of the damage was based on a combined analysis of the photoresponses at $p < p^*$ and $p \gtrsim p^*$ and of the damage morphology. Figure 2 shows a typical oscillogram of a KDP crystal at $p \approx p^*$ ($p^* = 10^{10}$ W/cm^2), obtained when a fault was produced in it in the form of a sphere of diameter $2a = 130$ μm, from which radial cracks propagated with length $l \approx 200$ μm from the center of the sphere. The same figure shows an oscillogram obtained in the absence of damage at $p \approx 0.9p^*$. At $p < p^*$ the photoresponse consists of a thermal current pulse J_ε and of an acoustic pulse J_{ac}, the shape of J_ε being a duplicate of that of the laser pulse. (At $p < p^*$ the amplitudes of the currents J_ε and J_{ac} increased linearly with increasing laser power up to the damage threshold.) At $p \approx p^*$ a rather abrupt kink of duration $\tau_1 \approx 14$ nsec appears on the trailing edge of the thermal pulse and attests to an abrupt increase of the absorbed power P_{abs}. This is followed, at $\tau_2 \approx 35$ nsec, by a reversal of the photocurrent polarity from that of J_ε to that of J_σ, and J_σ reaches a maximum already after the termination of the laser pulses, after which it falls off with a characteristic damping time $\tau_3 \approx 100$ nsec. After a certain delay, determined by the sound-wave propagation time from the beam-heated region to the contact surface of the crystal, an acoustic current pulse appears, with an amplitude J_{ac}^* markedly higher (by an approximate factor 1.5) than that before the breakdown.

It must be noted that it is quite difficult to obtain sufficiently simple oscillograms with clearly pronounced sharp kinks when the damage occurs near the threshold, and in our experiment we obtained only approximately one such oscillogram out of ten. Thus, when the excess above threshold is increased, the photoresponse becomes much more complicated, while the dynamic range and the temporal resolution of the employed apparatus were insufficient to obtain the photocurrent

data needed for a reliable interpretation of the results. On the other hand, when the size of the damage region is decreased, its contribution to the photoresponse is significantly smaller, inasmuch as this contribution is proportional to the laser-radiation energy absorbed in the damaged volume.

It can be assumed, in accord with the described dynamics of the photoresponse at $p \approx p^*$, that the damage process is initiated by a region of size much smaller than a, and the increase of the absorption in this initial region starts out with a prolonged heating over the time from the start of the laser pulse to the appearance of a kink in the current J_ε($\approx$60 nsec). The dimensions of the region of increased absorption (damage) increase at a speed close to that of sound ($a/\tau_1 = 4.7 \times 10^5$ cm/sec, as against the speed of sound $c \approx 5 \times 10^5$ cm/sec in KDP), so long as the laser radiation energy ensures a sufficient thermal-energy density in the absorbing volume. When the laser pulse terminates, the energy density in the damaged region decreases rapidly and the volume-damage process ceases. There are left, however, large tangential stresses on the boundary of the damage region, due to heating of this region, and lead to formation of cracks accompanied by appearance of free carriers (of a current J_{cr}) in the photoresponse. The rise time of J_{cr} is determined by the crack growth time and is close to the ratio of their length to the speed of sound. We note that the product of the carrier density by carrier mobility on the crack surface is $n_{sur}\mu_{sur} = 2 \times 10^{14}$ cgs esu, and the lifetime is $\sim 10^{-7}$ sec, i.e., the same as on the surface of the crystal surface.[8]

On the basis of the oscillograms of Fig. 2 and on the basis of the dynamics that follows from their analysis we can obtain certain characteristics of the medium in the course of its damage. To this end we use first the acoustic current pulses J_{ac}. The presence of a sufficiently soft teflon ribbon between the bulky contact and the crystal makes the sample boundary free. Therefore the acoustic waves lead to a change of the capacitance of the contact gap and of the sample capacitance on account of the change of its dimensions. In our experiments there were satisfied the conditions $\varepsilon_c h \gg \varepsilon x$, $\varepsilon \gg \varepsilon_c$, and $\Delta x \gg x$, where ε_c and x are respectively the low-frequency permittivity and the height of the gap of the capacitive contact, Δx is the change of the gap height, and h is the dimension of the sample in the direction between the contacts. The expression for the acoustic current then takes the simple form

$$J_{ac}(t) = -\frac{U_0 C_0}{h}\frac{C_\Sigma^2}{C_0^2}\frac{\varepsilon}{\varepsilon_c}\left\langle \frac{dx}{dt}\right\rangle, \tag{1}$$

where C_0 is the capacitance of the sample, C_Σ is the series capacitance of the contacts and the sample, and $\langle \ldots \rangle$ is the change of the gap height averaged over the contact area.

At small surface displacements ($\Delta x \ll x$) we have

$$\left\langle \frac{dx}{dt}\right\rangle = \frac{1}{S_c}\int \mathbf{v}\, dS, \tag{2}$$

where S_c is the contact area, $\mathbf{v}$ is the surface-velocity component in the direction of the surface normal, and the integration is over that part of the surface where $\mathbf{v} \neq 0$.

The sample surface is acted upon by a cylindrical sound wave due to linear absorption of the radiation in the crystal ($\alpha_0 = 5 \times 10^{-2}$ cm^{-1}), as well as a spherical wave produced by the energy absorbed in the damage region (the pressure cannot escape the damage region during the time of its formation, since the dimensions of the region increase at the speed of sound) and leads to the appearance of an additional current $A_{ac}^* = J_{ac}^* - J_{ac}$. To determine the connection between A_{ac}^* and the average pressure p_0 in the damage region we solve the problem of the motion of a free crystal boundary under the influence of a spherical sound wave. The potential φ of the radial velocity of the material particles, defined as $v = \partial\varphi/\partial r$, is given by[9]

$$\varphi = \frac{p_0}{4\rho c}\frac{(ct-r)^2 - a^2}{r}, \tag{3}$$

where ρ is the density of the substance, c is the speed of sound, r is the distance from the center of the damage region to an arbitrary point on the sample surface, and $\varphi \neq 0$ at $r - a < ct < r + a$.

Using (2) and (3) we obtain from (1) the connection between A_{ac}^* and p_0:

$$A_{ac}^* = \frac{U_0 \varepsilon}{4h^2}\frac{C_\Sigma^2}{C_0^2}\frac{\varepsilon}{\varepsilon_c}\frac{p_0 a^2}{\rho c}. \tag{4}$$

Substituting in (4) the experimental values $A_{ac}^* = 7 \times 10^{-7}$ A, $U_0 = 1.8 \times 10^3$ V, $\varepsilon = 44$, $h = 0.5$ cm, $C_\Sigma^2/C_0^2 = 0.6$, $\varepsilon_c = 2$, $a = 5.6 \times 10^{-3}$ cm, $\rho = 2.3$ g/cm^3, and $c = 5 \times 10^5$ cm/sec, we obtain the average pressure $p_0 \approx 2 \times 10^{10}$ dyn/cm^2 in the damage region. From the relation $p^0 = \Gamma w_T$, where $\Gamma = \alpha c^2/c_v$ is the Grüneisen coefficient, α is the temperature coefficient of volume expansion, and c is the specific heat, we obtain for the density of the thermal energy in the damage volume a value $w_T = 10^3$ J/cm^3 (in the calculation of w_T we took no account of the temperature dependence of the specific heat, this being justified at $T \lesssim 10^4$ °C,[10]). This value of w_T corresponds to a temperature $T = w_T/c_v = 500$ °C, which is higher than the melting point of KDP under normal conditions ($T_{m0} = 250$ °C at $p_0 = 0$).

The value of T_m of KDP at high pressure is unknown and can be estimated only from Johnson's empirical formula.[11] At $p_0 = 2 \times 10^4$ atm, according to estimates, $T_m \sim 2 \times 10^3$ °C so that the melting of the damage region occurs apparently only after all the pressure waves leave this region. It can be assumed on this basis that the absorbed-energy density is $w_{abs} = w_T = 10^3$ J/cm^3. From the current pulse J_ε in the photoresponse we can determine the radiation energy incident on the damage volume during the damage development time (within a time $\tau_1 = 14$ nsec) and calculated the coefficient of absorption by the damage region, averaged over this time, namely $\alpha^* = 25$ cm^{-1}. We note that a value of α^* close to this is obtained by comparing the corresponding values of J_ε in the absence and in the presence of damage.

3. DISCUSSION

We have proposed above an interpretation of the macroscopic damage as a process that starts in a

TABLE I. Values of the pressure P_{cr} of the deformation phase transition and of the optical-damge threshold p^* for several alkali-hadide crystals.

	Substance						Reference
	KI	KBr	KCl	NaCl	NaF	LiF	
P_{cr}, kbar	27	38	49	100	330	680	[16]
p^*, GW/cm^2 ($\lambda = 1.06$ μm)	22	50	70	120	140	360	[17]

Note. At $P_{cr} \lesssim 100$ kbar the pressure density is directly proportional to the transition pressure.

small region and propagates with the speed of sound. It is obvious that at $p \approx p^*$ the process can be maintained only by energy from the laser beam, and this is possible if the damage is accompanied by an increase of the absorption. This raises the question of the mechanism of absorption in the volume of the crystal and the method of its initiation.

We consider first the nature of the absorption. The moderate damage conditions ($p^* = 10^{10}$ W/cm^2, $p_0 = 2 \times 10^{11}$ dyn/cm^2, $T = 500$ °C) allow us to assume that the absorption is more readily due to a redistribution of the populations of the electronic states than to a substantial restructuring of the energy spectrum. In particular, the absorption can be due to fundamental transitions between a pair of valence bands or between a pair of conduction bands. The band structure of KDP (Ref. 12) admits of either possibility. The absorption-coefficient value observed in the experiment reported above (25 cm^{-1}), at typical values of the transition matrix elements, is ensured by a free-carrier density of the order of 10^{18} cm^{-3}. (This density is related to the start of the electron avalanche. But an avalanche in its usual sense[13] is hardly possible in KDP, in which the width of the upper valence bands and of the lower conduction bands is substantially less than the width of the forbidden band.[12]) This value of the density does not contradict the experimental results on the photoresponse of KDP (Ref. 8) (the average conductivity in the damage region is $\sigma < 2 \times 10^8$ sec^{-1}), if we assume for the carrier mobility a value $\mu \lesssim 10^{-3}$ cm^2/V-sec, which is perfectly realistic under breakdown conditions.[3)]

The question of generation of free carriers, with account taken of the high propagation velocity of this process in space, is more complicated. We associate the carrier generation process with the pressure wave[4)] and we can point out immediately two possibilities. First, the deformation potential produces on the pressure wave a displacement of the levels, and as a result a change in their populations. For pressures corresponding to laser-induced damage of KDP ($p_0 = 2 \times 10^{11}$ dyn/cm^2) this displacement is of the order of the Fermi energy, if a value 10 eV is assumed for the deformation potential. On the other hand the density of the defect states on the order of 10^{18}–10^{19} cm^{-2} in KDP single crystals is perfectly feasible.

Second, a pressure wave can generate defect states simply on account of the fluctuations that appear in a fast (10^{-9}–10^{-8} sec) transition with increasing pressure through the region of the deformation phase transition, and the increase of absorption can be due in this case also to defect–band carrier transitions.[5)] There are no direct data on this question in the case of KDP, but for a large number of alkali-halide crystals, for example, one can trace a distinct correlation between the pressure of the deformation phase transition,[16] on the one hand, and the threshold of the optical damage, on the other[17] (see Table I).

As for the properties of the initial center (which initiates the damage), a slow heating should be followed in it by an abrupt increase of the laser-emission absorption within a time on the order of the ratio of its dimension to the speed of sound. This is a necessary condition for the production of a pressure that ensures the start of absorption in the volume of the crystal. The required value of the thermal-energy density $w_T = 10^3$ J/cm^3 in a center of size $R = 10^{-5}$ cm is reached when the absorption coefficient is increased to $\alpha^*_{cent} = 5 \times 10^3$ cm^{-1}.

Thus, the volume damage induced in sufficiently pure dielectric crystal by nanosecond laser pulses can be due to a pressure wave.

In conclusion, the authors thank A. A. Manenkov for a helpful discussion.

1) In a number of recent papers (see, e.g., Ref. 5), the emission is associated with crack formation.

2) According to contmeporary ideas (see, e. g., Ref. 7), an electron avalanche is possible only in extremely pure optical materials with strength of the order of 10^{12} W/cm^2.

3) See, e.g., Ref. 14, where the free-electron mobility in sapphire is found to the 2×10^{-3} cm^2 V/sec under normal conditions.

4) We note that neither impact ionization nor thermal excitation can ensure the required propagation velocity of the process. In addition, the thermal energy ($T = 500$ °C) is insufficient for a substantial change of the population at a Fermi energy of the order of 2 eV.[8]

5) An absorption increase amounting to several orders of magnitude in the transparency band was observed for high-temperature phase transitions in a direct experiment.[15]

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