DESTRUCTION OF KDP, ADP, AND $LiNbO_3$ CRYSTALS BY INTENSE LASER RADIATION

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Volume destruction of KDP, ADP, and $LiNbO_3$ crystals due to focused radiation from a single-mode Q-switched laser is investigated. The nature of the destruction of KDP or ADP crystals does not depend on orientation of the sample or frequency of the incident radiation. In $LiNbO_3$ crystals, and under some conditions, in KDP and ADP crystals thread-like defects are observed. The absolute destruction thresholds for KDP, ADP, and $LiNbO_3$ crystals at 1.06 and 0.53 μm are given. A strong dependence of the threshold power density on the focal distance of the lens is noted. The decrease of the destruction threshold is ascribed to self-focusing and to the dynamics of thermoelastic stresses. It is shown that multiphoton absorption and the stimulated Mandel'shtam-Brillouin scattering effect are not decisive in destruction processes in KDP and ADP crystals; the important absorption mechanism is linear.

1. INTRODUCTION

THE nonlinear crystals potassium dihydrogen phosphate (KDP), ammonium dihydrogen phosphate (ADP), and lithium niobate ($LiNbO_3$), are used in systems for frequency conversion and modulation of laser radiation. In parametric oscillators, frequency multipliers, and modulators (shutters), nonlinear crystals work under high power densities of the laser radiation, which leads, under some conditions, to destruction of these crystals. Destruction of KDP and ADP was reported in[1-3]. In[3] the threshold power density for destruction at 1.06 μ was estimated to be 190–100 MW/cm^2 for KDP and ~500 MW/cm^2 for ADP. However, a special study of the destruction process has not been made before.

Our goal was a systematic investigation of the processes of volume destruction of nonlinear crystals by laser radiation under different conditions (at different frequencies, beam size at the lens focus, impurity content, etc.), in an attempt to establish the possible destruction mechanisms.

2. EXPERIMENTAL RESULTS

1. Experimental arrangement and samples. The principal investigations were conducted with the experimental arrangement shown schematically in Fig. 1. A neodymium-glass laser (rod 8 mm in diameter, 130 mm long) with plane mirror was Q-switched by a dye solution. The dye cuvette was placed at the Brewster angle to the beam axis; this produced linear polarization of the output. The laser was made to operate in a single transverse mode by means of a diaphragm 1.5 to 2 mm in diameter placed in the resonator. The radiation was attenuated by calibrated neutral filters and focused into the body of the sample by a spherical lens. In a number of the experiments the radiation at 1.06 μ was doubled to 0.53 μ by means of a KDP crystal.

The use of a single-mode laser permitted obtaining a regular distribution of intensity at the lens focus. The output of the laser was measured with a calibrated calorimeter. The output energy of each laser pulse was monitored. The pulse length was measured with

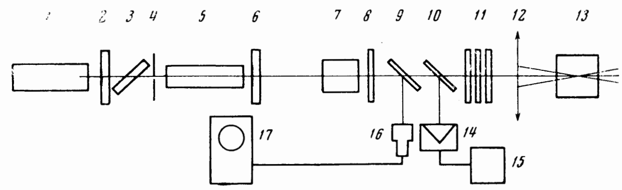

FIG. 1. Block diagram of the experimental arrangement: 1–gas laser (λ = 6328 Å); 2, 6–dielectric mirrors; 3–passive shutter; 4–diaphragm, 1.5 to 2 mm; 5–neodymium-glass rod, 8 mm diameter, length 130 mm (glass LGS-5); 7–KDP frequency doubler ($\varphi = 0°$, $\theta = 57°$); 8–SZS-21 filter; 9, 10–plane-parallel glass plates; 11–neutral filters; 12–spherical lens; 13–sample; 14, 15–calorimeter; 16–FÉK-09 photoelement; 17–S1-11 oscilloscope.

an FÉK-09 photoelement and S1-11 oscilloscope.

In certain cases, the radiation of a ruby laser was used. The radiation of the multimode Q-switched laser was amplified by means of a ruby rod 10 mm in diameter and 120 mm long.

The laser parameters are given in Table I.

The investigations were carried out on oriented polished samples of KDP and ADP of dimensions $20 \times 20 \times 20$ mm, and $LiNbO_3$ measuring $8 \times 8 \times 30$ mm. In $LiNbO_3$ Sample No. 1, there were scattering centers that could be distinguished visually at λ = λ = 0.63 μ. There were no such centers in Sample No. 2, which was prepared by a more sophisticated technique. The crystals were single-domain.

2. Nature of the destruction of KDP, ADP, and $LiNbO_3$ crystals. Volume destruction of the KDP and ADP crystals at laser powers close to threshold occur as cracks along planes of the form $\{110\}$ (Fig. 2,a), which correspond to the minimum values of the components of the elasticity tensor in these crystals. As the power is increased, chains of defects extended along the axis of the laser beam appear, the sizes of the splits increase, and cracks appear along $\{012\}$ as well as two cracks close to $\{001\}$ (Fig. 2,b). Often, an opaque nucleus is observed at the center of the damaged region (Fig. 2,c). The character of the destruction of crystals of the KDP group does not depend on crystal orientation.

At 0.53 μ, by focusing with lenses of f > 7.5 cm, we

Table I

Wavelength, μ	Output energy, J	Pulse length nsec
1.06 (single-mode operation)	0.11	14
0.53 (single-mode operation)	0.03	10
0.69 (multimode operation)	3 (after amplification)	20

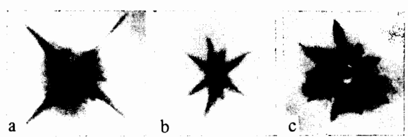

FIG. 2. Characteristic damage of KDP and ADP crystals. Magnification 34 times. a—cracks along {110}planes; b—cracks along {012}planes and two cracks close to {001}; c—opaque nucleus frequently observed in the center of the damaged spot.

were able to obtain threadlike defects in the KDP and ADP crystals; this is evidence that there is self-focusing of the laser radiation.[4] The threads were accompanied by splitting of the material (Fig. 3). In $LiNbO_3$, Sample No. 1, the damage was in the form of a track consisting of microcracks (Fig. 4,a). In the purer crystal (Sample No. 2), threads always appeared at 1.06 μ (Fig. 4,b).

The ruby laser produced threadlike damage in the ADP crystal. To compensate for the divergence of the laser and the weak compression of the beam, a lens of focal length f = 60 cm was used. When an ADP sample, 68 mm long, was placed so that the power density on its front face was greater than on the back face (i.e., when it was placed at the focus of the lens), the damage was in the form of a bundle of threads close to the back surface of the sample. The number of threads in the bundle increased with increasing laser power. Under the same conditions, a KDP crystal of the same length displayed only a track consisting of characteristic microcracks.

3. Measurement of the destruction threshold. Dependence of the destruction threshold on the frequency of the incident radiation and the laser beam cross section. The destruction threshold was taken to be the minimum power density at the lens focus that leads to defects in the sample that are visually distinguishable in the beam of a gas laser. At this power level one also observes strong scattering of the incident radiation and an intense white light accompanying the destruction.

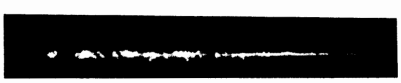

FIG. 3. Threadlike formations in KDP crystals, obtained by focusing radiation at 0.53 μ by a lens f = 7.5 cm. Magnification 7 times.

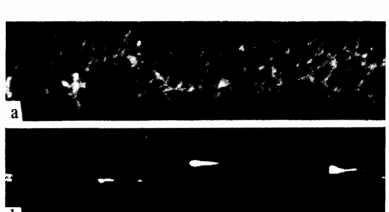

FIG. 4. Damage in $LiNbO_3$ crystals at 1.06 μ. a—Sample No. 1 (with scattering centers). The damage has the form of a track consisting of microcracks. Magnification 58 times. b—pure Sample No. 2. Damage in the form of a long thread. Magnification 30 times.

To determine the power density at the lens focus it is necessary to know the field distribution in the focal region. The spatial field distribution was photographed through a microscope and studied by photometry. The intensity distribution was close to Gaussian: $I = I_0 \exp(-r^2/r_0^2)$. We took the beam diameter $2r_0$ to be its size at the level $I = I_0/e$, which is equivalent to replacing the Gaussian distribution by a uniform one with intensity I_0 and radius r_0.

For all crystal samples studied, there was a strong dependence of the destruction threshold on the lens focal length, i.e., on the radius of the laser beam (Fig. 5). For example, for KDP crystals, as r_0 increased from 1.5×10^{-3} to 7×10^{-3} cm (for lenses with f = 7.5 cm and f = 33 cm), the destruction threshold at 1.06 μ fell by an order of magnitude. Further increase in beam size hardly changed the threshold. The relative stability of KDP and ADP remained approximately constant.

In Table II we give the thresholds for crystals of KDP, ADP, and $LiNbO_3$ at 1.06 and 0.53 μ for two lenses: f = 7.5 cm and f = 33 cm.

The experimental accuracy of determining the threshold was 30%. Note, however, that the local stability of different portions of a sample, as well as of different samples, can vary. The observed scatter of the results is indicated in Table II. No dependence of threshold on orientation of the KDP and ADP samples was observed. The destruction threshold for KDP and ADP did not change by more than a factor of two in going from 1.06 to 0.53 μ.

We were unable to damage the $LiNbO_3$ crystals with 0.53-μ radiation because of the optical inhomogeneities induced by this radiation.[5]

The observed scattering centers in $LiNbO_3$ No. 1 lower the threshold by one order of magnitude. The damage threshold for Sample No. 2 was higher than in KDP. The threadlike defects in the KDP and ADP crystals arise at 0.53 μ when the incident power is elevated to several times above the threshold. In the purer $LiNbO_3$ sample, the threads appear right at the threshold.

4. Additional experiments. In order to clarify the mechanisms of absorption of energy that lead to de-

Table II. Absolute destruction thresholds for nonlinear crystals in a focused laser beam

Sample	f = 7.5 cm		f = 33 cm
	$\lambda = 1.06\ \mu$	$\lambda = 0.53\ \mu$	$\lambda = 1.06\ \mu$
	I, GW/cm²	I, GW/cm²	I, GW/cm²
KDP	17 — 35	34 — 57	2.5 — 5.2
ADP	68 — 125	115 — 221	9.6 — 18
$LiNbO_3$ № 2	36	—	9.8 — 13.1
$LiNbO_3$ № 1	3.5	—	—

struction of the crystals, we carried out a few additional investigations.

a) The effect of stimulated Mandel'shtam-Brillouin scattering (SMBS).

We were able to observe SMBS in all the crystals[6]1) We observed SMBS in the KDP and ADP crystals at 0.53 μ, and in the $LiNbO_3$ crystals at 1.06 μ. In the KDP and ADP crystals, the destruction threshold was higher than the SMBS threshold with long-focus lenses (f = 15 to 30 cm), and lower with short-focus lenses (f $<$ 5 cm). Since it was possible to obtain damage without SMBS, this phenomenon does not play a decisive role in the process of destruction of KDP and ADP.

b) Effect of impurity absorption.

We investigated the threshold of green-colored ADP and KDP crystals. The colored ADP crystals had an additional absorption band in the region of 14 500 cm^{-1}. Hence, for relative measurements of the destruction thresholds of the colored and clear portions of the crystal we used the ruby laser described in Sec. 1. The linear absorption coefficient of the ADP crystal, averaged over the thickness, at the frequency of the ruby laser (ν = 14 400 cm^{-1}) was ~0.012 cm^{-1} for the colorless portion and ~0.018 cm^{-1} for the colored region.

The crystal was placed at the focus of a lens f = 60 cm in such a way that the radiation passed through both the colored and colorless regions of the crystal. Destruction of the colored region was more intense and set in at a power 1.2 to 1.7 times less than the colorless region.

These results show the important role played by linear impurity absorption in the processes of destruction of KDP and ADP crystals.

For the $LiNbO_3$ crystals, we compared the thresholds of Sample No. 1 with scattering centers (contaminated by the crucible material during preparation) and of the pure, homogeneous Sample No. 2. As Fig. 4 and Table 2 show, the character and threshold of destruction for these two samples differ.

3. DISCUSSION OF RESULTS

The most important result of this investigation is the discovery of the strong dependence of the threshold power density leading to destruction on the transverse section of the laser beam at the lens focus. We have observed this effect, not only in KDP, ADP, and $LiNbO_3$, but also in other transparent materials (NaCl, LiF, ruby, glass). This dependence of the threshold on the focusing conditions must definitely be taken into account when comparing the results of different authors, who were unaware of it (see, for example,[8]).

The reasons for this dependence may be contained in the different stages of the destruction process. Conditionally, we might assume that the process goes through two stages: 1) absorption of light energy and heating of the lattice; 2) development of thermoelastic stresses up to the critical values for a given crystal.

In the first stage, the effect of thermal conductivity

1)We observed SMBS by the method described in [7], at 1.06 and 0.53 μ.

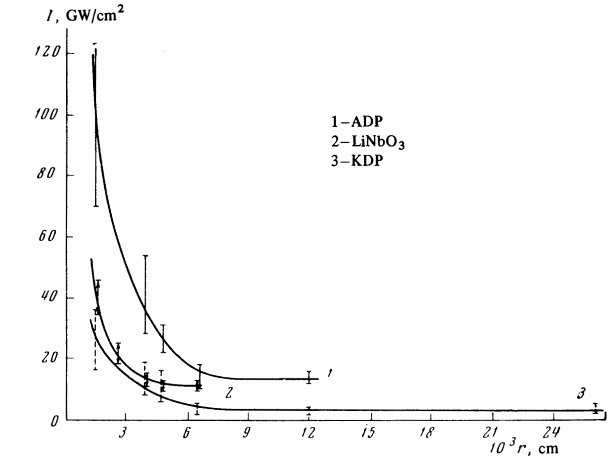

FIG. 5. Dependence of damage threshold on focal length of lens (focal radius of laser beam) at 1.06 μ.

and a change in the conditions for self-focusing can lead to a dependence of the thresholds on the cross section of the light beam. Estimates show that during the time of the pulse $\tau_p \sim 10^{-8}$ sec, thermal conductivity is effective only at radii of the focal region $\sim 10^{-5}$ cm, i.e., the thermal conductivity can be neglected in the conditions of our experiment.

As indicated above, we were able to observe thread-like damage due to self-focusing in KDP, ADP, and $LiNbO_3$ under certain conditions. In the long (68 mm) ADP samples threads were observed at the exit boundary of the sample, at the threshold. These facts point to the importance of self-focusing in the destruction process. The use of different lenses changes the conditions of self-focusing. Thus, the length of the focal region in which self-focusing occurs most effectively is changed: for long-focus lenses self-focusing is alleviated. Note that the turning point of the curve in Fig. 5 corresponds to the radius r_0, for which the characteristic time for sound evolution $\tau = r_0/v$ (where v is the velocity of sound) is equal to the pulse length τ_p. Hence the drop of the curve in Fig. 5 may also be due to the transience of self-focusing, but to take this into account requires a knowledge of the self-focusing mechanism.

Besides the effects described above, a decisive contribution to the dependence of destruction threshold on lens focus may be brought in by the process of development of thermoelastic stresses itself. When the laser radiation is focused into the body of the crystal by lenses with different focal lengths, the same power density gives the same heating, but different stresses. During the pulse in the case of long-focus lenses the stresses are unable to propagate out of the characteristic region ($\tau_p < r_0/v$), and their maximum value in this case will be greater than in the case of short-focus lenses, when the stresses propagate into a larger region during the pulse ($\tau_p > r_0/v$); hence to get damage we need more heat — higher energy density at the lens focus.

To have damage, we must have energy absorption. As was shown above, the linear processes of impurity absorption are of the highest importance in the destruction of KDP and ADP.[2)] It is seen from a comparison of the thresholds of these crystals at 1.06 and 0.53 μ that processes of multiphoton absorption do not play an important role: for three-photon processes (λ = 0.53 μm) the transition probability should be much higher than for six-photon processes (λ = 1.06)[9]; the experiment shows just the opposite dependence of the thresholds. Thus, our results do not confirm the supposed importance of multiphoton transitions in the radiation damage of crystals of the KDP type.[10] The roles of the processes of self-focusing and the SMBS effect were considered above.

In conclusion, we note that the absolute magnitudes of the threshold fluxes of radiation required for damage were found in our case to be considerably higher than in[3]. The difference is due to the fact that in using a single-mode laser we measured local power densities.

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[2)] Estimates show that the heating of the KDP crystals due to linear absorption amounts to a few degrees, which leads to the appearance of thermoelastic stresses of 1 to 10 kg/cm^2, insufficient for damage. Localization of absorption at the impurities, plus the increase of light intensity due to self-focusing, can elevate the stresses to values sufficiently high for destruction.

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