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MECHANISMS OF DESTRUCTION OF RUBY AND LEUCOSAPPHIRE CRYSTALS BY POWERFUL LASER RADIATION

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The mechanisms of volume destruction of ruby and leucosapphire single crystals under the influence of powerful laser radiation are investigated. The destruction thresholds of these crystals are obtained as functions of the temperature, duration of the generation pulse, and the wavelength of the laser radiation. It is shown that the mechanism of crystal destruction by radiation with $\lambda = 1.06\ \mu$ and $\lambda = 0.7\ \mu$ is connected with impact ionization and with the development of an electron cascade. An approximate calculation of this mechanism agrees with the experimental results.

INTRODUCTION

A large number of recent papers is devoted to the destruction of transparent dielectrics under the influence of intense laser radiation. Studies were made on glass[1-7], alkali-halide crystals[4,5,8], transparent polymers[4,9], fused and crystalline quartz[6,10], and ruby and leucosapphire crystals[11,12,14]. Different possible destruction mechanisms were also considered, such as damage to the material by a powerful hypersonic wave excited during the course of stimulated Mandel'shtam-Brillouin scattering[1,4,6,10], damage due to multiphoton ionization[11], due to self-focusing of the laser radiation[13], and others. However, the mechanism of the destruction of transparent materials by light is still not sufficiently clear.

Particular interest attaches to a study of the damage produced by the light in the laser active media themselves, since this process limits the power of Q-switched lasers.

In our preceding investigation[12] we established a connection between the endurance of ruby crystal to the action of ruby-laser radiation and the presence of color centers in the ruby. The destruction thresholds of ruby crystals with color centers turned out to be 20–30 times lower than the destruction threshold of leucosaphire and "pure" ruby.

In this paper we present the results of an investigation of the destruction of optically homogeneous leucosapphire and ruby crystals without color centers. Crystals containing unfused inclusions, bubbles, and other visible defects are easier to damage; the destruction of such crystals is not considered in the present paper. A study of the light-induced damage in homogeneous pure crystals makes it possible to establish more distinctly the destruction mechanisms and to determine the maximum permissible powers that can be withstood by laser material. We investigated the dependence of the destruction thresholds on the duration of the light pulse, on the temperature of the investigated samples, and on the wavelength of the laser radiation.

INVESTIGATION PROCEDURE AND EXPERIMENTAL RESULTS

We used in the experiments ruby and leucosapphire samples in the form of rectangular polished bars measuring 20 × 20 × 30 mm.

A block diagram of the experimental setup is shown in Fig. 1. Radiation from a laser was focused into the volume of the samples by a spherical lens (f = 45 mm). The radiation intensity was attenuated by neutral light filters calibrated for large light fluxes. Part of the

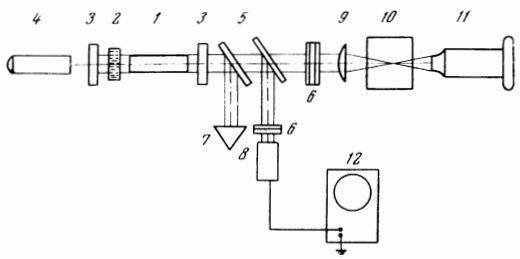

FIG. 1. Block diagram of experimental setup: 1–ruby rod 10 mm diameter × 120 mm, 2–saturating filter, 3–dielectric mirrors, 4–gas laser (λ = 6328 Å), 5–plane-parallel quartz plates, 6–neutral light filters, 7–calorimeter, 8–photomultiplier, 9–spherical lens (f = 45 mm), 10–investigated sample, 11–microscope with camera, 12–high-speed oscilloscope.

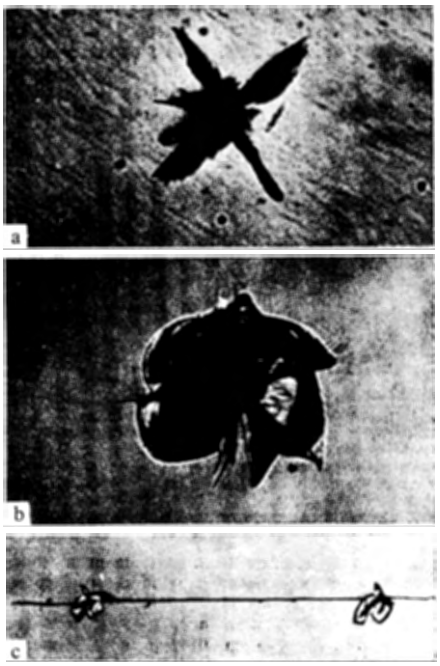

FIG. 2. Characteristic damage in ruby and leucosapphire crystals under the influence of radiation with different wavelengths. Magnification 40×. a—$\lambda = 0.7\ \mu$ and $\lambda = 1.06\ \mu$ (viewed along the axis of the laser radiation); b—the same (perpendicular to the radiation axis); c—$\lambda = 0.53\ \mu$ and $\lambda = 0.35\ \mu$ (perpendicular to the radiation axis).

radiation was diverted to a calorimeter to measure the pulsed energy, and part to a photomultiplier. The pulse duration was registered with a high-speed oscilloscope S-1-11.

By crystal destruction threshold is meant the minimum power (or energy) flux from the laser, causing damage to the sample in the focal region of the lens; this damage could be visually distinguished by passing through this region the radiation from a gas laser, with 6328 Å wavelength. The light from the same laser was used to select optically homogeneous crystals or individual homogeneous sections of crystals.

a) Dependence of the Ruby Crystal Destruction Threshold on the Duration of the Laser Radiation Pulse

In this experiment we used a monopulse laser with Q-switching by means of a solution of vanadium phthalocyanine in nitrobenzene. The resonator was made up of two external dielectric mirrors with reflection coefficients 99.5 and 40%. The pulse duration was regulated from 15 to 60 nsec (at half intensity) by varying the resonator length from 60 to 490 cm.

The dimension of the focal spot was calculated from

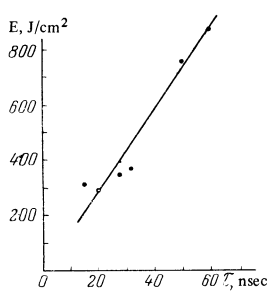

FIG. 3. Destruction threshold energy vs. duration of laser pulse.

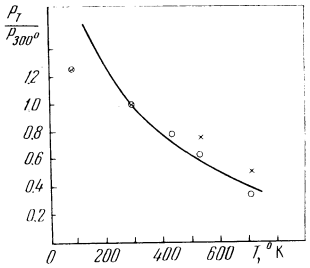

FIG. 4. Temperature dependence of destruction thresholds: × — leucosapphire, ○—ruby. Solid curve—calculated from formula (7).

the divergence of the laser beam, with allowance for the spherical aberration of the lens. The divergence was estimated from the pattern of the radiation at the focus of a long-focus lens (f = 1 m).

The character of the destruction in the crystals was the same for all pulse durations, taking the form of flat cracks, either round or elongated along the beam, intersecting one another along the laser beam direction (Figs. 2a, b). The dimension of the damaged section at threshold was ~0.1 mm and increased with increasing incident power. The results of the measurements of the destruction energy threshold are shown in Fig. 3. The straight line on this figure corresponds to the constant-power level. The experimental values of the thresholds for durations longer than 15 nsec are close to this straight line. The observed scatter in the experimental values of the threshold can be connected with variations of the distribution of the generation field at the focus of the lens with changing pulse duration. An additional confirmation of the fact that the destruction threshold is determined by the power (and not by the energy) of the radiation was obtained from the following experiment. The operating regime of the ruby laser was modified: the laser produced two identical short pulses (τ = 20 nsec) spaced ~100 μsec apart. During this time interval, the effective thermal conductivity does not cause a noticeable loss of energy absorbed in the focal region during the time of action of the first pulse[1)].

It was established that when destruction was produced by two pulses, the threshold energy of each pulse corresponds (accurate to 15%) to the threshold energy of destruction by one pulse, and the total threshold energy doubles in this case.

b) Temperature Dependence of the Destruction Threshold of Ruby and Leucosapphire Crystals

The destruction thresholds of the crystals were investigated in the temperature interval from 77 to 700°K. The samples were cooled by placing them on the cold finger of a vacuum cryostat. A ruby laser pulse of 20 nsec duration was focused into the crystal through the quartz window of the cryostat. Damage was observed through the second window perpendicular to the direction of the incident radiation. The crystals were heated in air with a constantan heater. A quartz

[1)] The characteristic time of thermal relaxation from the focal region $t = a^2/4\kappa$ (where a—diameter of the focal spot, κ—coefficient of temperature conductivity) was in our case 250 μsec. Photometry of the focal spot revealed the absence of considerable fluctuations of the radiation intensity, and therefore we chose the focal spot diameter a as the characteristic dimension.

Radiation parameters and destruction thresholds of crystals at different frequencies

Radiation wavelength, μ	Total pulse power, MW	Pulse duration, nsec	Destruction thresholds, 10^{10} W/cm^2	
			Leucosapphire	Ruby
1.06	30	30	2.5	2.3
0.69	50	20	3.8	2.8
0.53	2.5	20	1.5	0.4
0.35	3.5	15	2.0	1

plate, immitating the cryostat window, was placed between the lens and the crystal.

The threshold power flux was measured in relative units (the threshold at room temperature was taken as unity). Such measurements do not require allowance for the distribution of the laser generation field, and therefore are more accurate.

The character of crystal destruction did not change in the entire temperature interval. The experimental results are shown in Fig. 4. The destruction thresholds are noticeably lowered with increasing temperature.

c) Frequency Dependence of the Thresholds and of the Character of the Crystal Destruction

We investigated the destruction of ruby and leucosapphire crystals by radiation from Q-switched ruby and neodymium lasers and their second harmonics. The frequency doubling was with KDP crystals.

In calculating the threshold power flux, the dimension of the focal spot was determined at all frequencies at half intensity by photographing the focal region through a microscope with magnification 14×. The radiation was attenuated with neutral filters to obtain normal blackening of the photographic film. The blackening curves were constructed with the aid of a nine-step attenuator. The energy and the duration of each generation pulse were controlled. The parameters of the radiation at different frequencies and the measured destruction thresholds of the investigated crystals are listed in the table. In calculating the destruction thresholds, the radiation intensity was averaged over the cross section of the focal spot. The thresholds listed for the ruby crystals take into account the absorption of the radiation by the chromium impurity.

We note the following features of the results.

1. The character of the destruction of ruby and leucosapphire crystals by radiation of 1.06 μ wavelength is similar to the destruction by ruby-laser radiation, but the dimension of the threshold destruction is approximately twice as large.

2. Extended damage regions were observed following the action of the harmonic radiation. Microscopic studies of such damaged regions has shown that they consist of very thin (~10 μ) and long (up to 5 mm) filaments, usually emerging from the focus and directed along the laser-beam axis (Fig. 2c).

Additional investigations of the temperature dependence of the destruction threshold of ruby crystals at 0.35 μ wavelength have shown that the endurance is practically constant in the interval from 77 to 700°K.

These results show that the ruby and leucosapphire crystal destruction mechanisms at the fundamental frequencies of the lasers are different from those at their second harmonics.

DISCUSSION OF RESULTS

The dependence of the threshold crystal breakdown energy on the duration of the generation pulse shows that the destruction mechanism cannot be attributed to thermal effects only. The destruction is determined by mechanisms that depend on the field intensity of the light wave. As shown earlier[12], mechanisms that depend on the field intensity - multiphoton ionization of the chromium impurity and stimulated Mandel'shtam-Brillouin scattering - are not decisive in the destruction of ruby and leucosapphire crystals.

The destruction of ruby and leucosapphire crystals under the influence of ruby and neodymium-glass laser radiation can be explained from the point of view of the mechanism of impact ionization and the development of an electron cascade in the field of the light wave.

This mechanism is connected with the acceleration of the conduction electrons in the crystal under the influence of the electric field, up to an energy exceeding the width of the forbidden band. Such electrons can cause further ionization of the crystal and increase the concentration of the conduction electrons. The impact ionization mechanism, as is well known, determines the electric breakdown of solid dielectrics in static fields[15].

Let us consider the action of the light field on an electron appearing in the conduction band of a dielectric as a result of thermal ionization of shallow impurities or multiphoton ionization in the intense field of laser emission. The conduction electron is accelerated by the field of the light wave only in the presence of scattering from impurities of phonons. On the other hand, scattering of electrons by phonons decreases the electron energy.

If the field intensity of the light wave is sufficiently large, the increase of the electron energy will prevail over the energy loss due to scattering by phonons. In a field of such intensity, the electron can acquire an energy exceeding the width of the forbidden band and then, as shown in the theory of electric breakdown of dielectrics[15], it has a large probability of ionizing an atom or ion of the lattice. As a result of the ionization, two "slow" electrons appear in the conduction band in lieu of one "fast" electron; these, in turn, can be accelerated by the field. The cascade ionization produced in the crystal leads to a rapid increase of the conduction-electron density and to effective absorption of the laser-emission energy. The release of a large amount of energy in a small volume of the crystal leads to destruction of the latter.

The acceleration of the electron in the field of the light wave has a quantum character, since the electron energy varies discretely by an amount equal to the photon energy. The impact ionization mechanism should be described in this case by a quantum kinetic equation for the conduction-electron distribution function.

We present an approximate calculation of the impact-ionization mechanism, using the approximations of Frohlich's theory for the electric breakdown of dielectrics[15,16]. We consider a "fast" conduction electron with energy $\epsilon \approx I$, where I is the ionization potential or the width of the forbidden band of the crystal, and we assume $I \gg \hbar\omega$ ($\hbar\omega$ - photon energy).

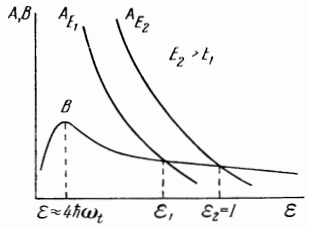

FIG. 5. Plots of A (rate of acquisition of energy by the electron) and B (rate of energy loss) vs. electron energy ϵ.

In this case the interaction of the electron with the light wave can be considered classically[17]. Then the increase of the electron energy per unit time under the influence of a light field of frequency ω will be

$$\left(\frac{d\varepsilon}{dt}\right)_1 = \frac{e^2E^2}{m^*\omega^2}\nu\frac{\omega^2}{\omega^2+\nu^2}, \tag{1}$$

where E is the intensity of the electric field of the wave, e and m* are the charge and effective mass of the electron, and ν is the effective frequency of electron collision with the phonons and impurity centers. Putting $\omega \gg \nu$, we get

$$\left(\frac{d\varepsilon}{dt}\right)_1 = \frac{e^2E^2}{m^*\omega^2}\nu = \frac{e^2E^2}{m^*\omega^2}\frac{1}{\tau}, \tag{2}$$

where τ is the time between collisions. The collision frequency is $\nu = \nu_{phon} + \nu_{imp}$, where ν_{phon} and ν_{imp} are the frequencies of the electron collisions with the phonons and impurities. For small impurity concentrations and sufficiently high temperatures we have $\nu \approx \nu_{phon}$.

The main contribution to the energy loss of the electron for a lattice with an ionic bond is made by the interaction between the electron and longitudinal optical phonons. As shown in Frohlich's theory[16], the frequency of collisions with such phonons is

$$\frac{1}{\tau} = \frac{1}{\tau_0}\left(1+\frac{2}{\exp(\hbar\omega_t/kT)-1}\right), \quad \frac{1}{\tau_0} = \frac{2^{2/3}\pi^3}{4\sqrt{2}}\frac{e^4\hbar}{m^{1/3}Ma^5\omega_t\varepsilon^{1/2}}, \tag{3}$$

where a - lattice constant, M - reduced mass of the ions, ω_t = frequency of optical phonons, and the energy losses per unit time are

$$\left(\frac{d\varepsilon}{dt}\right)_2 = \frac{\pi e^4\sqrt{2m}}{Ma^3\sqrt{\varepsilon}}\ln\frac{2\pi\sqrt{2\varepsilon}}{2^{2/3}a\omega_t\sqrt{m}}. \tag{4}$$

We put $(d\epsilon/dt)_1 = A$ and $(d\epsilon/dt)_2 = B$. Figure 5 shows the dependence of A and B on the electron energy ϵ.

To each value of the field intensity E there corresponds a critical electron energy ϵ_{cr}, for which

$$A(E, \varepsilon_{cr}) = B(\varepsilon_{cr}). \tag{5}$$

At the field intensity E_1 (Fig. 5), the electrons with energy $\epsilon < \epsilon_1$ are accelerated by the field, since $A > B$ for such electrons. The energy of electrons with $\epsilon > \epsilon_1$ will decrease. At a light-wave field intensity E_{thr}, the critical energy of the electron reaches the value of the ionization potential. Fields with larger intensity will accelerate the electrons to the ionization potential and lead to the development of an electron cascade.

Solving (5) and putting $\epsilon_{cr} = I$, we get E_{thr}[2]:

$$E_{thr} = \frac{2\sqrt{2}\,ma\omega\sqrt{\omega_t I}}{2^{1/3}\pi e\sqrt{\hbar}}\left[\ln\frac{2\pi\sqrt{2I}}{2^{2/3}a\omega_t\sqrt{m}}\right] \times\left[1+\frac{2}{\exp(\hbar\omega_t/kT)-1}\right]^{-1/2}. \tag{6}$$

Substituting the numerical values of the sapphire parameters: T = 300°K, I ≈ 6 eV, ω_t = 200 cm^{-1}, a = 5.13 Å, and ω = 14 400 cm^{-1}, we get $E_{thr} = 5 \times 10^7$ V/cm. The calculated value of E_{thr} exceeds by one order of magnitude the experimental value $E = 3.7 \times 10^6$ V/cm, corresponding to a flux 3.8×10^{10} W/cm^2. The discrepancy may be due to the difference between the peak values of the electric field at the focus of the lens and the experimentally measured mean values, and also due to imperfection of the theory.

Formula (6) determines the temperature dependence of the threshold field intensity, namely, E_{thr} decreases with increasing temperature. This circumstance is connected with the increased frequency of the electron collisions with the phonons with increasing temperature.

Figure 4 shows a plot, in relative units, of

$$E_{thr}^2 \sim \left(1+\frac{2}{\exp(\hbar\omega_t/kT)-1}\right)^{-1}. \tag{7}$$

This plot agrees well with experiment in the temperature region 300—700°K. The discrepancies observed for low temperatures can be connected with the process of electron scattering by impurities, which can no longer be neglected at low temperatures ($\nu_{imp} \approx \nu_{phon}$).

It follows from (6) that the threshold power increases with increasing frequency ($E_{thr}^2 \sim \omega^2$). Unfortunately, the different distribution of the generation field in the plane of the focus at wavelengths 0.7 and 1.06 μ does not make it possible to compare exactly the calculation with experiment. However, as seen from the table, the endurance of the crystals actually increases somewhat on going from the neodymium-laser frequency to that of the ruby laser. The dependence of the threshold on the ionization potential, which follows from (6), can explain the reduced endurance of ruby crystals with smaller width of the forbidden band, compared with leucosapphire.

For the development of an electron cascade, a certain finite time is required. If the duration of the laser pulse turns out to be smaller than this time, then the threshold intensity of the field should increase. This apparently explains the increase of threshold power at small durations (15 nsec) shown in Fig. 3.

Wasserman[18] considered the mechanism of conduction-electron acceleration by a light wave in interaction with longitudinal optical phonons, assuming approximately that the conduction electrons have in the stationary state (at field intensity below threshold) a Boltzmann distribution function with a temperature T_e larger than the equilibrium lattice temperature T_0.

On reaching the threshold field intensity, the equilibrium between the average rates of acquisition and loss of energy, and consequently also the stationary state, becomes upset and impact ionization develops. The threshold intensities were calculated with the aid of such a breakdown criterion for several alkali-halide crystals. The calculated values of the field intensities, just as in our case, turned out to be approximately one

[2] Frohlich's theory neglects the difference m* and m.

order of magnitude higher than the experimental ones.

The mechanism of impact ionization can play an important role in the processes of destruction of other transparent materials, too, such as fused and crystalline quartz or glass. The character of destruction in these materials is similar to the destruction in ruby and leucosapphire.

From the point of view of the mechanism under consideration, the materials having large endurance under the action of laser radiation should be those with a large forbidden-band width and a large electron free path time (or large mobility).

The mechanism of impact ionization determines the limiting endurance of ruby crystal against the action of laser radiation. It is interesting to note that the destruction threshold for ruby is lower by one order of magnitude than the maximum generation power determined by multiphoton absorption[19].

The characteristic destruction in ruby and leucosapphire crystals at second-harmonic frequencies can be attributed to self-focusing of the laser radiation[20]. This phenomenon is discussed in greater detail in[21]. The self-focusing effect increases the local density of the light-wave field and contributes to the development of damage in crystals.

The strong change in the character of the damage in ruby and leucosapphire crystals with changing wavelength of the laser radiation can serve as an example showing that essentially different destruction mechanisms have nearly equal thresholds. This circumstance greatly complicates the study of the optical breakdown of transparent dielectrics.

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