SURFACE DAMAGE OF RUBY CRYSTALS BY LASER RADIATION

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The processes of surface damage of ruby crystals by the radiation of a ruby laser are investigated experimentally. The dependence of damage threshold on the duration of laser pulses ranging from 3×10^{-7} to 4×10^{-4} s and the effect of structure-optical properties of the surfaces on the damage threshold are studied. A theory of thermal damage at absorbing surface defects is developed. An expression is obtained for the dependence of damage threshold on pulse duration. Good agreement is found between the experimental data and the proposed theory.

1. INTRODUCTION

RECENTLY there have appeared a large number of papers (see, for example,[1-6]) devoted to the investigation of the processes of damage of optically transparent dielectrics by laser radiation. Particular attention has been given to the investigation of the processes by which crystals of ruby and white sapphire are damaged.[7-11] However, an adequate mechanism has not been established. In particular, the question of the significant difference between the thresholds for surface and volume damage remains unclear. This difference obviously points to a connection between the damage mechanism and the structural (in the sense of the presence and number of various defects) and optical properties of the surface and volume of the crystals.

A large number of papers (e.g.,[12,13]) are devoted to structural investigations of ruby and white sapphire crystals; however, the connection between the structure and the damage mechanism has not been investigated in depth. The available data on the processes of damage in ruby and white sapphire have been obtained for pulses of radiation not longer than 10^{-7} s. The investigation of these processes over a wide range of pulse lengths would be of obvious help in explaining the destruction mechanism.

In this paper we investigate the dependence of the surface damage threshold for ruby crystals on the pulse duration and the relation between the structural properties of the surface and the damage threshold. In this work, we used laser pulses (λ = 6943 Å) of rectangular shape with durations in the range 3×10^{-7} to 4×10^{-4} sec. We examined samples with mechanically polished surfaces treated in various ways, as well as with crystal surfaces formed by mechanical cleavage.

Our data is discussed on the basis of a proposed mechanism for thermal damage at absorbing surface defects.

2. EXPERIMENTAL ARRANGEMENT

A schematic diagram of the experimental arrangement for investigating surface damage on ruby crystals is given in Fig. 1. The laser system consisted of a high-quality ruby laser followed by a two-stage amplifier. The basic laser was operated in the spikeless mode with a pulse length of ~1 msec. This mode was achieved through the use of a complex resonator[14] consisting of two plane mirrors and two spherical lenses (F = 25 cm), between which the ruby crystal was located.

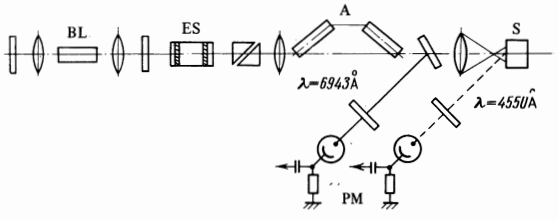

FIG. 1. Block diagram of the experimental arrangement for investigating surface damage of crystals: BL – basic laser, ES – electrooptic shutter, A – amplifier, S – sample, PM – photomultipliers with interference filters.

A Pockels electrooptic shutter was placed between the basic laser and the amplifier in order to obtain rectangular pulses and variation of pulse length. The electrical system regulating the shutter provided pulses with length variable from 3×10^{-7} to 4×10^{-4} sec, flat tops, and rise times not worse than 30–40 nsec. The power density of the radiation at the amplifier output reached 10^5 W/cm^2. A fast oscillographic scan of the output rectangular pulse with a time resolution of $\sim 10^{-9}$ sec demonstrated the absence of fast modulation of the radiation intensity which could have arisen as a result of mode-locking.

In order to attain radiation densities high enough for damage, the radiation was focused by a lens (F = 5.5 cm) onto the surface of the sample, which was placed at the lens focus in such a way that the diameter of the light spot on the surface was ~1 mm. The fact that damage had occurred was determined visually or with a photomultiplier to register the appearance on the crystal surface of the light flashes which always accompany damage.

The structure of the crystal surfaces was studied by electronography, optical microscopy, and light scattering. The electronograph had an electron energy of 70 keV and the microscope a magnification of 500.

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3. STRUCTURE-OPTICAL PROPERTIES OF THE CRYSTAL SURFACES

In order to elucidate the effect of structural and optical properties of the crystal surface on the radiation damage threshold, we used a large number of mechanically polished ruby crystals with subsequent treatment by various methods (chemical polishing and thermal monocrystallization[13]). Figure 2 shows electronograms of several types of investigated surfaces obtained by various treatments. As is seen, type-a crystals have an amorphous surface structure, whereas type-b surfaces are close to being monocrystalline.

Microrelief photographs of these same surfaces, obtained with the optical microscope, are shown in Fig. 3. It can be seen that the surfaces of samples a and b have a grainy structure, whereas that of sample c shows the step structure characteristic of etch figures.

Observation of interferograms of the investigated samples showed that the contrast of the interference fringes depends markedly on the degree of surface roughness. Analysis of the contrast led to the conclusion that the height of the grains on the surface of samples a and b was much less than the wavelength (λ = 6328 Å), but was comparable to it in sample c.

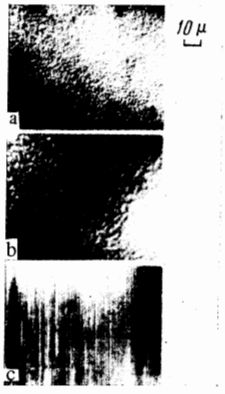

FIG. 3. Microphotographs of the surface relief of the same samples of ruby as in Fig. 2.

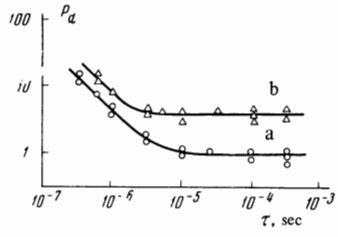

FIG. 4. Dependence of threshold power for surface damage (in relative units) on the length of laser pulses for two types of ruby samples.

4. SURFACE DAMAGE OF THE CRYSTALS

Using the apparatus described in Sec. 2, we measured the surface damage threshold as a function of laser pulse length. The experimental data obtained for samples of type a and b are presented in Fig. 4. It is seen that in the region of short pulses the damage threshold power $P_d \sim 1/\tau$, whereas for long pulses P_d = const (independent of τ). The location of the transition region depends on the structure of the investigated surface: for sample a, with amorphous surface structure, a change in the character of the dependence is observed at $\tau \approx 5 \times 10^{-6}$ sec, and for sample b at $\tau \approx 2 \times 10^{-6}$ sec. It is also important to notice that the difference in the damage thresholds of samples a and b is greater for long pulses ($\tau > 5 \times 10^{-6}$ sec) than for short pulses ($\tau < 2 \times 10^{-6}$ sec). Approximate measurements of the power for damage showed that for sample a in the region of long pulses $P_d \approx 10^6$ W/cm^2. We remark that the scatter of the experimental points in Fig. 4 is not due to fluctuations in the power and duration of the laser pulses from flash to flash, but to differing abilities of different points of the sample surface to resist damage. A particularly strong scatter in P_d (sometimes by factors of two or three) is observed on cleaved surfaces.

These data indicate a definite connection between the mechanism of surface damage and the structure-optical properties of the surface. This connection is obviously rather complex and depends on many factors. Evidence for this is the fact that the threshold for damage of surface d, formed by cleavage of the crystal and having a monocrystalline structure (which is clear from the character of the electronogram), was found to be close to the threshold for sample a, which has an amorphous surface. This suggests that an electronogram (obtained by reflection from the surface) does not give information about the defects which may be responsible for the damage process. And as far as the optical characteristics (microphotography, light scattering) are concerned, these reflect the structure of the surface relief. However, electronographic and optical methods nevertheless

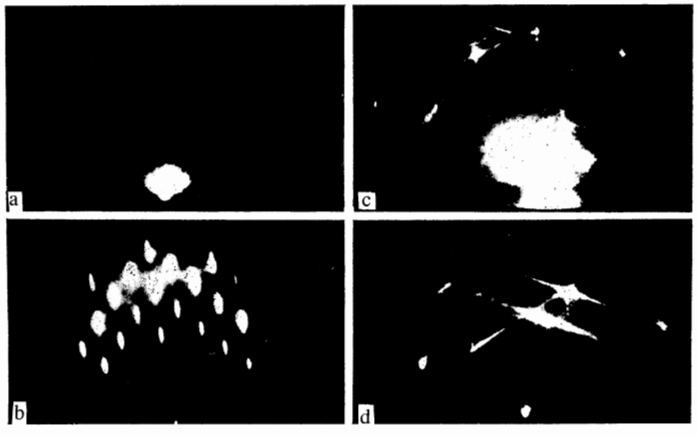

FIG. 2. Electronograms of the surfaces of crystals of ruby obtained by various methods.

give information on the properties of the crystal surfaces, which in certain cases can directly characterize their resistance to damage, as was noted in[10].

5. INTERPRETATION OF EXPERIMENTAL RESULTS. MECHANISM FOR SURFACE DAMAGE.

In many papers (e.g.,[2,4,5,11]) the most probable mechanisms discussed for the damage of transparent dielectrics by radiation are multiphoton ionization and electronic breakdown associated with nonlinear (multiphoton) absorption of the light. However, our experimental data on the surface damage of ruby crystals presented in the preceding section are difficult to explain on the basis of these mechanisms. In fact, the observed dependence of the threshold power for damage in the region of short pulses $P_d \sim 1/\tau$ is inconsistent with multiphoton absorption. In addition, the level of power at which damage is observed is obviously too small for multiphoton absorption to be sufficiently strong.

We propose that the observed dependence $P_d(\tau)$ can be explained on the basis of a mechanism of thermal damage during ordinary single-photon absorption of light at local surface defects. Examples of such defects are microcracks, boundaries between mosaic blocks, twin boundaries, and aggregates of dislocations in cleavage planes. Processes of diffusion of impurities both from the surrounding medium (e.g., by mechanical treatment of the samples) and also from the bulk of the crystal can lead to a marked increase in the absorption of light by these defects.

Let us consider the thermal mechanism of surface damage. Let there be local absorbing centers of mean size q and mean separation l in some surface layer of the crystal. Suppose that the absorption coefficient of these centers $\alpha \gg \alpha_0$, where α_0 is the absorption coefficient of the bulk crystal. As a result of absorption of light by surface defects, they will be heated to some critical temperature T_{cr} at which damage sets in (irreversible structural changes of the surface like fusion, cracking, etc.). This temperature obviously depends on the magnitude of the absorbed power P and on the processes of heat removal, which determine the dependence of T_{cr} on the duration of the light pulse. In the region of short pulses $\tau < \tau_{char}$, where $\tau_{char} = c\rho q^2/k$ is the mean characteristic time for establishment of thermal conductivity within the absorbing centers (c is the heat capacity, ρ the density, k is the coefficient of thermal conductivity of the substance forming the center[2)]), it is obvious that $T_{cr} \sim P\tau$, i.e., it is given by the absorbed energy.

In the region of long pulses ($\tau > \tau_{char}$) it is necessary to consider the processes of heat transfer between the absorbing centers. In the point-defect approximation the heat conduction equation is written

$$\Delta u = \frac{c\rho_0}{k_0}\frac{\partial u}{\partial t} - \frac{\psi(t)}{k_0}\sum_i Q_i\delta(\mathbf{r}-\mathbf{r}_i), \qquad (1)$$

where $u = T - T_0$, T is the temperature of the sample at point r at time t, T_0 is its initial temperature, $\mathbf{r}_i$ is the coordinate of the i-th thermal source (an absorbing center), and Q_i its intensity, $\psi(t)$ is a function of the shape of the light pulse. A solution of Eq. (1) with the initial condition $u(\mathbf{r}, 0) = 0$ and in the absence of heat removal from the surface to the medium surrounding the crystal, for a rectangular pulse of length $\tau > \tau_{char}$ can be easily found in approximate form[15]:

$$u(\mathbf{r}, t) \approx \frac{Q}{4\pi kq} + \frac{Q}{k_0 l^2}\sqrt{\frac{k_0 t}{c_0\rho_0}}. \qquad (2)$$

Since $Q = \alpha Pv$ (v is the volume of the absorbing center), we obtain a dependence of the threshold power for damage on the pulse length for $\tau > \tau_{char}$ of the form

$$P_d \approx (T_{cr} - T_0)\left(\frac{\alpha v}{4\pi kq} + \frac{\alpha v}{l^2}\sqrt{\frac{\tau}{c_0\rho_0 k_0}}\right)^{-1}, \qquad (3)$$

and for $\tau < \tau_{char}$ (in the absence of conductivity effects) it is obvious that

$$P_d \approx \frac{c_0\rho_0(T_{cr} - T_0)}{\alpha\tau}. \qquad (4)$$

Let us compare Eqs. (3) and (4) with the experimental data presented in Sec. 4 for the dependence of surface damage threshold of ruby crystals on the length of the laser pulses. From the character of these formulas it is seen that they can qualitatively explain the observed dependences for $\tau < \tau_{max}$, where

$$\tau_{max} = \frac{1}{16\pi^2}\left(\frac{l}{q}\right)^4\left(\frac{c_0\rho_0 k_0}{c\rho k}\right) \qquad (A)$$

is a time such that when $\tau > \tau_{max}$ processes of heat transfer between the absorbing centers begin to play an important role. Thus the dependence $P_d(\tau)$ for $\tau < \tau_{max}$ should be described by the formula

$$P_d \approx \frac{(T_{cr} - T_0)}{\alpha}\left(\frac{c_0\rho_0}{\tau} + \frac{3k}{q^2}\right). \qquad (5)$$

For a quantitative comparison of Eq. (5) with the values of P_d observed for ruby it is necessary to know the quantities q, k, and α for the absorbing centers. The dimension q can be estimated from the observed magnitude of $\tau_{char} \approx 5 \times 10^{-6}$ sec (region of transition from $P_d \sim 1/\tau$ to P_d = const). Setting $c = c_0 = 0.18$ cal/g-deg, $\rho = \rho_0 = 4$ g/cm^3, $k = k_0 = 0.06$ cal/cm-sec-deg, we find $q \approx 6 \times 10^{-4}$ cm. If we take T_{cr} as the melting temperature of ruby $T_m = 2 \times 10^{3\circ}$C, and $P_d = 3 \times 10^6$ W/cm^2 (the experimental value for $\tau = 5 \times 10^{-6}$ sec) as the surface damage threshold, then from (5) we obtain $\alpha = 10^2$ cm^{-1}.

Note that the coefficient of thermal conductivity k for the substance of the absorbing center can be much smaller than the magnitude of k_0 corresponding to the bulk crystal. In addition, the problem we are treating is linear, whereas in actuality the parameter c/k can markedly increase with heating to T_{cr}, and since $T_{cr} \gg T_0$, this must all serve to strongly diminish the magnitude of q.

Thus, the thermal damage theory developed here explains rather well the observed regularities and gives reasonable magnitudes for the coefficients of absorption of the absorbing centers in the surface layer of a ruby crystal. In addition, the proposed mechanism makes clear the difference in the magnitudes of the threshold power for damage for samples with different surface structures, as well as its difference for the surface and

[2)] The quantities c, ρ, and k may differ from the corresponding quantities c_0, ρ_0, and k_0 for the bulk crystal.

volume of the crystal. In fact, a change in the parameters α and q for surface defects (which is completely likely) ought to lead to a change in the magnitude of P_d, and one that is different in the region of short and long pulses, which was indeed observed experimentally (Fig. 4).

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