Ruby breakdown by pulse trains and single ultrashort pulses

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We investigate the breakdown of ruby by ultrashort laser pulses of wavelength 1.06 μ. When a train of pulses acts on the ruby, a cumulative effect that lowers the optical strength of the crystal is observed. The dependence of this effect on the pulse repetition period in the train is investigated. It is shown that a decisive role in the optical breakdown of ruby is played by direct ionization of the crystal-lattice atoms in the field of the strong electromagnetic wave, and not by impact cascade ionization.

INTRODUCTION

Breakdown of transparent dielectrics by giant laser pulses of 10^{-8} sec duration has by now been thoroughly investigated. Molchanov [1] gives the theoretical value of the threshold field intensity at which this breakdown takes place:

$$E_{\text{thr}}^2 = \frac{160\pi}{a}\frac{n^2\hbar^2\sqrt{I_i}}{e^2\sqrt{2m}}\frac{\rho u_*^2(\hbar\omega)^2}{\tau kT\varepsilon_i^2}.$$

The formula gives a threshold field that agrees with experiment [2-6]. This agreement gives grounds for assuming that the laser radiation is absorbed by the conduction-band electrons produced by impact ionization of the lattice atoms. This is confirmed by the presence of photoconductivity in ruby during the pre-breakdown stage [3]. When breakdown is produced in transparent dielectrics by ultrashort laser pulses, the relation $J_{\text{thr}}\tau = \text{const}$ ceases to hold at lower values of the threshold field intensity [7,8]. This indicates that impact ionization ceases to be the decisive mechanism that transfers electrons into the conduction band, first because only several electron generations can be produced in such a short time, and second because of the decrease in the cross section for impact ionization by fast electrons [9]. The main contribution to the transfer of conduction electrons to the conduction band is made by multiphoton ionization of the lattice, the probability of which $p^{(n)} \sim J^n\tau$ exceeds the probability $p^{(1)} \sim J\tau$ of single-photon impact ionization on going to giant pulses to ultrashort laser pulses of 5×10^{-12} sec duration.

EXPERIMENT

We investigate here the breakdown of ruby by a train of ultrashort pulses as well as by single pulses.

The apparatus for the investigation of the breakdown is shown in Fig. 1. The source of the train of high-power ultrashort laser pulses was a neodymium-glass laser (5–8), operating in the axial-mode locking regime. The diaphragm (7) selected the angular modes. Its diameter ranged from 1 to 3 mm, depending on the length of the resonator. The axial modes were locked by a saturable filter constituting a solution of polymethine dye in nitrobenzene. Parasitic frequency selection has been eliminated from the resonator. The cell with the saturable filter was placed on a mirror with 100% reflection coefficient. This prevented the filter position from influencing the quality of the mode locking. The mirror (5) could move freely along the optical axis. This varied the distance between the spikes in the train.

To separate a single pulse from a train of ultrashort pulses we used an electro-optical shutter consisting of

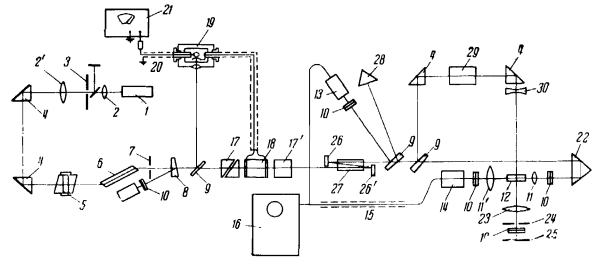

FIG. 1. Diagram of setup for the investigation of ruby breakdown by ultrashort pulses.

a Pockels cell (18), two crossed Glan prisms (17 and 17′) and a discharge gap (19) with a segment of driving cable (20). The Pockels cell was a Z-cut KDP (KH_2PO_4) crystal with silvered electrodes deposited on its two ends. To open the Pockels cell completely, a square pulse of amplitude 12 kV and duration 10 nsec was applied to it; this pulse was produced by an open cable segment of one meter length, charged to 25 kV by a VSIP-25 high-voltage source (21). The pulse was synchronized with the lasing by a discharge gap, breakdown of which was produced by a light pulse diverted from the main laser radiation by means of glass plate (9). The radiation was focused on one of the electrodes of the discharge gap through a hole in the second electrode. The rise and fall times of the resultant pulse did not exceed 1–2 nsec. The discharge gap was filled with nitrogen at 15 atm pressure.

To permit the passage of at least one radiation pulse through the opened Pockels cell, the electric pulse duration T = 10 nsec was chosen to be longer than the repetition period of the ultrashort pulses. In this case either one or two pulses were cut out from the train. The transmitted pulse energies, measured with a calorimeter (28), were 0.007 and 0.015 J following passage of one and two pulses, respectively. This indicates that the background makes a small energy contribution and its influence on the breakdown can be neglected.

The separated pulse was amplified by triple passage through a neodymium-glass amplifier (27). Mirrors (26 and 26′) were used for spatial separation of all three passages of the radiation through the rod. After going through the amplifier, part of the radiation was deflected by a wedge-shaped glass plate to a photocell, and another part to an IÉK-1 calorimeter (28). A second glass plate diverted the radiation to a system of prisms (4) and the KDP crystal (29), where it was broadened by negative lens (30) to equal the sample dimensions. The broadened radiation of the second harmonic passed laterally through the ruby sample, through the lens (23), and through diaphragm (24) and was incident on photographic film (25).

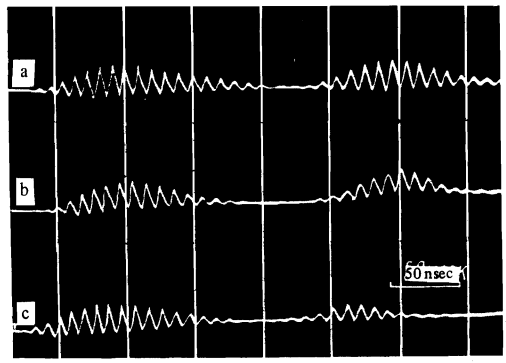

FIG. 2. Oscillograms showing the breakdown of ruby by a train of ultrashort pulses with different intensities: a) $J = 0.2\,J_{thr}$, b) $J = J_{thr}$, c) $J = 1.5\,J_{thr}$.

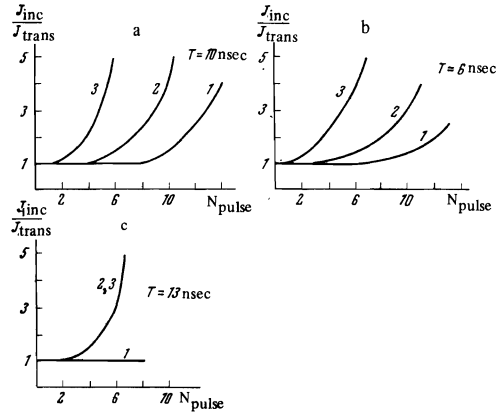

FIG. 3. Decrease of intensity of ultrashort pulses following passage through ruby with different repetition periods in the train and with different intensities, as a function of the number of pulses in the train. Curves: 1) $J < J_{thr}$, 2) $J = J_{thr}$, 3) $J > J_{thr}$.

The use of neodymium radiation to sound second harmonics made it possible to increase the resolution of this system, first because a diffraction limit of the resolution is increased and second because it was possible to register the shadow picture with a film having a higher sensitivity and resolution.

The remaining radiation passed through a prism (22) and was focused by a lens (11) into the interior of the sample (12). The light passing through the sample was incident on an FÉK-09 photocell (14), the signal from which was fed, with a delay of 30–200 nsec relative to the signal from another photocell (13), to type I2-7 oscilloscope (16). The delay of the sounding pulse relative to the breakdown pulse was scanned by moving the prism (22).

The adjustment was with the aid of a helium-neon laser 1, the beam divergence of which was reduced to the divergence of the neodymium laser with the aid of a telescopic system (2–2′). The same laser was used to monitor the onset of damage inside the sample. The intensity of the light incident on the sample was varied by moving the filters (10) from the front of the sample to its rear.

The investigations were carried out with breakdown produced either by a train of picosecond pulses or by single pulses. In the former case the system for separating the single pulses was excluded from the setup.

Photographs of oscillograms produced by breakdown with a train of pulses are shown in Fig. 2. They comprise two trains of pulses, follow each other with a time delay 200 nsec produced by a cable (15). The duration of the spikes on the oscillograms is determined by the time resolution of the apparatus. The first train corresponds to radiation incident on the sample, and the second to the transmitted radiation. The sensitivities of the photocells were balanced by filters (10). The oscillograms were obtained at different radiation intensity incident on the sample. In Fig. 2b, the intensity is equal to the threshold at which breakdown takes place, while $J = 0.2J_{thr}$ and $J = 1.5J_{thr}$ in Figs. 2a and 2c, respectively. It is easily seen that in all three cases the first spikes of the train pass through the sample without change, whereas the last spikes are considerably attenuated. At $J = 0.2J_{thr}$, the attenuation begins with the tenth spike, and at $J = 1.5J_{thr}$ it begins even with the third spike.

Figure 3 shows plots of the ratio of the incident-spike intensity to the transmitted intensity against the number of the spike in the train, obtained by reducing the oscillograms. Figure 3a corresponds to a distance T = 10 nsec between spikes. Curve 2 was plotted for breakdown of the crystal at threshold intensity $J = J_{thr}$. Curves 1 and 3 were obtained with the ruby exposed to light of intensity $J < J_{thr}$ and $J > J_{thr}$. Figures 3b and 3c correspond to intervals T = 6 and T = 13 nsec between spikes. The marking of the curves is the same as in Fig. 3a.

It can be noted from Fig. 3 that when the distance between the spikes in the train is increased to T = 13 nsec, the absorption of the last spikes vanishes if there is no damage. The decrease of the amplitude of the last pulses in the train can be attributed only to their absorption, since light scattered through angles up to 10° still was incident on the photocell, and scattering through larger angles in the absence of damage can be neglected. Thus, relaxation of the absorption of light by the electrons of the conduction band of ruby takes place during the time between the pulses. The relaxation time is $\tau = 13$ nsec, inasmuch at this interval between pulses there is no absorption at below-threshold intensity. If the axial period of the laser is shorter, however, there is absorption and, as seen from Fig. 2, it increases towards the end of the train, i.e., there is a cumulative effect that decreases the optical strength of the crystal.

The damage produced in ruby by either a single pulse or by a train comprises a quasiperiodic array of points. It can be attributed to the onset of self-focusing in the sample, constituting a system of traveling foci [10]. The damage occurred at the turning points of these foci, i.e., where the radiation interacts longest with the medium. In damage of a medium by a train of ultrashort pulses, the turning points of the first pulses move towards the radiation source and consequently the preceding damage points will not screen the radiation. The first pulses for which $P_{cr} < P < 2P_{cr}$ have one turning point each. For the following pulses, the damage will occur in one large region, owing to the screening of the radiation by the preceding breakdown.

Shadow photographs of ruby breakdown by two pulses are shown in Fig. 4, where a corresponds to a delay t = 100 psec of the sounding radiation relative to the breakdown. One can clearly see the absorption produced at the turning points of the first pulse, while at the turning points of the second pulse there is practically

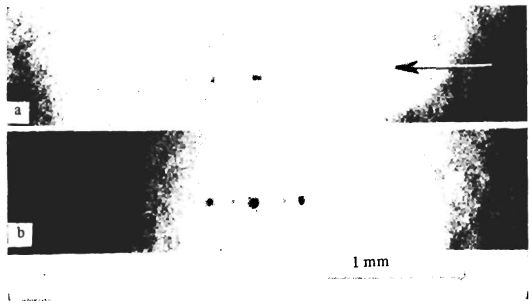

FIG. 4. Shadow photographs of ruby breakdown: a) delay of the sounding radiation relative to the breakdown radiation t = 100 psec (the arrow shows the direction of the radiation), b) produced damage.

no absorption. Figure 4b shows the onset of the damage in the sample. The direction of the breakdown radiation is shown by the arrow.

For crystals of poor optical quality and with more impurities, the damage took place over the entire track of the traveling foci. Breakdown of these crystals makes it possible to trace the dynamics of development of the region of increased absorption, in which the breakdown takes place subsequently. This region occurs first in the focus of the lens and then "travels" towards the radiation source. In the case of poor self-synchronization, the pulses that follow the principal ultrashort pulse are absorbed as they pass through the sample, and this absorption is stronger the closer the pulse is to the principal ultrashort pulse. The results of experiments with delay of the breakdown relative to the sounding light showed total absence of its absorption, and when the sounding pulses were delayed relative to the breakdown by $t=(60-800)\times 10^{12}$ sec, the dimensions of the absorption region were in conformity with the subsequently produced damage.

The threshold radiation intensity at which the breakdown of ruby by one ultrashort pulse takes place amounted in our experiments to 10^{14} W/cm^2. The cross section of the beam was assumed to be the absorption region corresponding to the pre-breakdown state of the ruby, and the pulse duration was assumed to be equal to 5×10^{-12} sec. Exact measurements of the duration of the ultrashort pulses were not carried out in this study, so that the calculations presented below can be only approximate.

DISCUSSION

As noted earlier, on going from nanosecond laser pulses to ultrashort pulses, an ever increasing role is assumed, in addition to the cascade ionization of the lattice at breakdown intensities, also by multiphoton absorption. The role of cascade ionization decreases with increasing intensity, since the cascade-development constant becomes a decreasing function of the flux density. Physically this is connected with the fact that the effective energy of the electron oscillations in the field of a light wave with amplitude $\mathcal{E}_0 = e^2E^2/2m\omega^2$ becomes comparable with or larger than the ionization potential. As a result, the electron falls rapidly into the energy region where the ionization probability of the crystal-lattice atoms decreases with increasing field amplitude [9].

At the same time, the probability of the tunnel effect and of multiphoton ionization increases rapidly with increasing intensity. The tunnel effect appears at high intensities but low frequencies, while multiphoton ionization occurs in the other limiting case at high frequencies and low intensities. The decisive parameter of these effects is

$$\gamma=\omega\sqrt{m\Delta}/eF,$$

where ω is the frequency of the incident field, m is the reduced mass of the electron and of the holes, e is the charge of the electron, and F is the intensity of the light-wave field. At $\gamma \gg 1$, the ionization is produced by a multiphoton effect, and at $\gamma \ll 1$ by tunnel ionization.

For the case of ultrashort pulses, in optical breakdown of ruby, we had in our case $\omega = 1.78\times 10^{15}$ sec^{-1}, $\Delta = 9$ eV, and $F = 6\times 10^5$ cgs esu, so that we get $\gamma = 0.7$. In this case it is impossible to use the formula for multiphoton ionization. It is necessary to use the general formula for the crystal-lattice ionization probability (see [11]):

$$W=\frac{2\omega}{9\pi}\left(\frac{\sqrt{1+\gamma^2}}{\gamma}\frac{m\omega}{\hbar}\right)^{3/2} Q\left(\gamma,\frac{\tilde{\Delta}}{\hbar\omega}\right)\exp\left\{-\pi\left\langle\frac{\tilde{\Delta}}{\hbar\omega}+1\right\rangle\right.$$
$$\left.\times\left[K\left(\frac{\gamma}{\sqrt{1+\gamma^2}}\right)-E\left(\frac{\gamma}{\sqrt{1+\gamma^2}}\right)\right]\Big/E\left(\frac{1}{\sqrt{1+\gamma^2}}\right)\right\}$$

where $\tilde{\Delta}$ is the effective ionization potential

$$\tilde{\Delta}=\frac{2}{\pi}\Delta\frac{\sqrt{1+\gamma^2}}{\gamma}E\left(\frac{1}{\sqrt{1+\gamma^2}}\right)$$

Substitution of our experimental values into this formula yields the following value of the ionization probability:

$$W=4.5\cdot 10^{32}\ \text{cm}^{-3}\ \text{sec}^{-1}.$$

Consequently, after a time $\tau = 10^{-12}$ sec the number of electrons in the conduction band is $n=4.5\times 10^{20}$ cm^{-3}, and the absorption coefficient amounts in this case to $K = 12$ cm^{-1}, leading to total absorption of the radiation and to optical breakdown.

Thus, the considered mechanism of direct ionization of the crystal lattice is decisive for the breakdown of ruby by ultrashort pulses. At intensities below threshold, the number of electrons in the conduction band is insufficient to produce breakdown.

When a train of ultrashort pulses acts on ruby, the electrons give up their energy to the lattice during the time between pulses, and roll down to the bottom of the conduction band. From the bottom of the conduction band the electrons can recombine with a hole in the valence band, or else go over to shallow donor levels whose lifetimes may turn out to exceed the time between the pulses. In this case, action of the next pulse causes the electrons situated in the shallow traps to go over much more rapidly to the conduction band than the electrons from the valence band. As a result the radiation becomes more strongly absorbed with each succeeding pulse, as was indeed observed in the experiment. This can explain also the decrease of the absorption of the last pulses in Fig. 3, curve 3, in comparison with curves 1 and 2. The distance between the spikes in this case 13 nsec, is the maximal in the given experiment. This means that during this time the donor levels have time to become depleted, and by the start of the next pulse the medium is in its initial state. Thus, the cumulative effects vanishes at $T > 13$ nsec. It appears that this type is typical only of the given type of crystals and is connected with the presence of uncontrollable dislocations and impurities. The observed cumulative effect must be

taken into account in the design of generators for ultrashort pulses. It can become manifest not only in crystals but also in glasses, and leads to a dependence of the optical strength of the working element and of the characteristics of the ultrashort pulses on the pulse repetition frequency and on their amplitude.

CONCLUSION

We have shown in this paper that in the absorption of radiation during optical breakdown of ruby by ultrashort pulses, the decisive role is played by direct ionization of the crystal-lattice atoms in the field of the strong electromagnetic wave, and not by impact cascade ionization. A cumulative effect was observed, due to the settling of the electrons from the conduction band on shallow donor levels with lifetimes $T = 13$ nsec, leading to a lowering of the optical strength of the crystal. When ultrashort pulses act on ruby, self-focusing is produced and leads to pointlike damage in the ruby at the turning points of the moving foci. A piercing of a self-focusing filament in a direction towards the radiation source was observed. The employed procedure makes it possible to determine the relaxation of the radiation absorption in times on the order of 10^{-11} sec.

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