# *Breakdown of Transparent Dielectrics by the Radiation from Mode-Locked Lasers*

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Breakdown of K-8 glass, ruby, leucosapphire, and fused and crystalline quartz by a train of ultrashort pulses with wavelengths $\lambda = 1.06\ \mu$ and $\lambda = 0.53\ \mu$ is considered. A characteristic feature observed is the presence of filamentary defects in the materials, which indicate that self-focusing of the radiation occurred. Threshold energy densities for the volume defects are presented. Possible breakdown mechanisms are discussed. The surface resistance of glass and ruby and also the bulk resistance of materials with microscopic inhomogeneities (such as ruby with color centers) are much higher on destruction by a train of ultrashort pulses than on destruction by a single pulse of duration of 10 nsec. This is due to thermal conductivity in the gap between the ultrashort pulses.

## 1. INTRODUCTION

A number of investigations have been devoted by now to the study of the damage produced in transparent dielectrics by radiation from free-running and Q-switch lasers. The mechanisms that lead to breakdown in the interior of the dielectric can be caused by absorption of the radiation by microscopic inhomogeneities of the material, and also by impurities, inclusions, etc.[1,2]. In homogeneous materials of high purity, the breakdown may be due to impact ionization in the field of the intense light wave[3,4]. In many cases, a decisive role in the breakdown process is played by self-focusing of the laser radiation[5,6].

Recently, the interaction of ultrashort laser pulses with matter, including transparent dielectrics, has attracted much interest[7,8]. It is of interest also to investigate the damage produced in transparent dielectrics by ultrashort laser pulses. Application of radiation with a power greatly exceeding the single-pulse power, and of duration $10^{-11}$–$10^{-12}$ sec to a substance can lead to a different evolution of the breakdown processes connected with the nonstationary character of the interaction between the radiation and the substance.

We have investigated breakdown in a number of solid dielectrics (glass, crystalline and fused quartz, ruby, and leucosapphire) by a sequence of picosecond pulses in one transverse mode. The values of the energy densities leading to volume and surface breakdown in the indicated materials at wavelengths $\lambda = 1.06$ and $\lambda = 0.53\ \mu$ are obtained. These values are compared with the breakdown thresholds by a laser operating in the single-pulse regime ($\lambda = 1.06\ \mu$). The character of the resultant breakdown is considered and possible breakdown mechanisms are discussed.

## 2. EXPERIMENTAL PROCEDURE

We used in the experiment a neodymium-glass laser with axial-mode locking. The laser generated a sequence of 30 pulses with time interval 11 nsec between pulses. The duration of each pulse, measured by the two-photon luminescence excitation method[9], was 4 psec. The output energy of the sequence of pulses reached 0.05 J in one transverse mode. For second-harmonic generation we used a KDP crystal. The output energy at $\lambda = 0.53\ \mu$ was 0.005 J. The radiation was attenuated by calibrated neutral filters and focused inside the volume of the sample or on its surface by a lens having a focal length f = 30 mm.

For temporal monitoring of the radiation, part of the light flux was diverted by glass plate to an FÉK-09 coaxial photocell and registered with a high-speed oscilloscope. Part of the energy incident on the lens was diverted by a glass plate to a calorimeter. The distribution of the radiation field at the focus of the lens was photographed through a microscope and measured with a MF-4 microphotometer. The diameter of the focal region at the 1/e intensity level was 28 $\mu$ for $\lambda = 1.06\ \mu$ and 17 $\mu$ for $\lambda = 0.53\ \mu$.

To measure the endurance and the monopulse regime, we used a neodymium-glass laser with energy 0.1 J in a pulse of 10 nsec duration in one transverse mode. The light-spot diameter at the focus of a lens with f = 30 mm was 30 $\mu$. The volume breakdown was investigated in polished rectangular samples of ruby, leucosapphire, K-8 glass, and fused and crystalline quartz, measuring $10 \times 10 \times 20$ mm. We investigated ruby crystals of three types: with and without color centers, with essentially different thresholds and different characters of the breakdown in the single-pulse regime[10]. The surface breakdown was investigated in rectangular samples of ruby and in plane-parallel PN-15 plates made of K-8 glass.

## 3. EXPERIMENTAL RESULTS AND DISCUSSION

In all the investigated materials, with the exception of the ruby crystals with color centers, radiation from a mode-locked laser at the fundamental frequency and at the second harmonic produced filamentary breakdown (Fig. 1) due to the self-focusing of the radiation[1)]. The filament diameter was 3 $\mu$, and the length at the breakdown threshold was approximately 1 mm. With increasing radiation power, the filaments grew longer in the direction towards the incident beam and terminated in volume breakdown regions measuring up to 0.2 mm. Many local breakdowns of the material, spaced 0.2–0.3 mm apart, were observed along the filaments. When the same materials were damaged by a mono-

[1)]The fact that the filamentary damage is due to self-focusing of the radiation was demonstrated earlier[6].

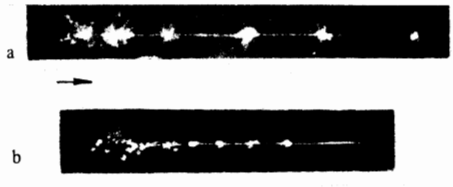


FIG. 1. Characteristic form of volume breakdown in glass, leucosapphire, ruby without color centers, and fused and crystalline quartz under the action of ultrashort pulses ($\lambda = 1.06\mu$). The arrow shows the direction of radiation propagation; a) magnification 28X; b) 23X.

pulse laser with $\lambda = 1.06\ \mu$ focused by the same lens, the breakdown regions were cracks in the focal region of the lens; no filaments were observed under these conditions.

In ruby crystals with color centers, the damage produced by radiation with mode synchronization constituted a track consisting of separated microcracks. A characteristic feature of the damage in this case is a small transverse dimension of the track, which remains the same along the entire track (Fig. 2). The displacement of the centers of the microcracks from the axis of the laser beam does not exceed 10 $\mu$. In the case of breakdown by means of a single pulse, the microscopic cracks fill the caustic of the lens, forming a conical track expanding in a direction opposite to that of the beam.

When the radiation of a mode-locked laser was focused on the input surfaces of glass and ruby, funnel-like craters were produced on these surfaces. In the case of damage by radiation of a monopulse laser, only burns were produced on the surfaces of the glass and ruby.

The table lists the values of the threshold energy densities leading to breakdown of the indicated materials, for the mode-synchronization regime ($\lambda = 1.06\ \mu$ and $\lambda = 0.53\ \mu$) and for the monopulse regime ($\lambda = 1.06\ \mu$). It should be noted that the threshold energy densities in these two regimes were close, and furthermore, for all materials, with the exceptions of ruby with color centers, the threshold energy densities in the mode-locking regime were lower by 1.5--2 times than in the single pulse regime. At the same time, the threshold power density in the mode-locking regime is larger by approximately three orders of magnitude than the threshold power density in the single pulse, and reaches $10^{14}$ W/cm$^2$. For ruby with color centers, to the contrary, the threshold energy

Threshold energy densities of volume breakdown ($10^2$ J/cm$^2$)

| Material | Train of picosecond pulses, $\mu$ | | Single pulse (T = 10 nsec, $\lambda = 1.06\mu$) |
|---|---|---|---|
| | $\lambda = 1.06$ | $\lambda = 0.53$ | |
| K-8 glass | 10 | 5,3 | 21 |
| Crystalline quartz | 14 | 7 | — |
| Fused quartz | 31 | 7,5 | — |
| Leucosapphire | 21 | 8 | 27 |
| Ruby without color centers | 15 | 4,4 | 26 |
| Ruby with color centers | 3,4 | 2,6 | 0,6 |

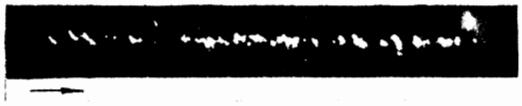

FIG. 2. Damage produced in ruby with color centers by ultrashort pulses ($\lambda = 1.06\mu$); magnification 23X.

density in the mode-locking regime is 5—6 times higher than with the pulse regime.

The filamentary character of the breakdown in the materials under the influence of radiation from a mode-locked laser indicates that the self-focusing process is in operation. Experiments show that when a series of short powerful pulses is applied, the self-focusing is much easier to produce than in the single-pulse regime. It follows from the character of the breakdown of ruby with color centers that in this case the breakdown is preceded by self-focusing of the radiation, but the formation of the filaments is hindered by breakdown on the inhomogeneities of the material.

The decrease of the threshold energy density at the second-harmonic frequency, in comparison with the fundamental frequency of the neodymium laser, is obviously connected with the dependence of the self-focusing threshold on the radiation frequency[11]. It should be noted that self-focusing for the mode-locking regime is not thermal, since the conditions for heating a small-diameter channel in the substance are more favorable for a single-pulse of 10 nsec duration, than for a series of short pulses with an envelope of approximately 300 nsec. In the latter case, an important role is played by the thermal conductivity from the light channel compressed to a diameter less than 5 $\mu$. However, self-focusing is observed only in the mode-synchronization regime, and the threshold energy density is in this case lower than for the monopulse. Apparently the self-focusing is due to the action of the electronic polarizability with short relaxation times.

The process of direct breakdown of materials in the field of powerful short pulses, whose intensity is furthermore magnified by the self-focusing of the radiation, can be due both to multiphoton ionization and to impact ionization in the field of the light wave. Estimates carried out in accordance with the work by Keldysh[2] show that even at light fluxes of $5 \times 10^{12}$ W/cm$^2$ at a wavelength $\lambda = 1.06\ \mu$ the probability of 6-photon ionization in leucosapphire reaches a value $2 \times 10^{29}$ cm$^{-3}$ sec$^{-1}$, which ensures an electron concentration of $10^{18}$ cm$^{-3}$ in the conduction band during the time of action of one pulse of 4psec duration. At laser intensities close to the breakdown threshold ($10^{14}$ W/cm$^2$) one can expect complete ionization of the matter in the focal region[2)].

Pulses of power $10^{14}$ W/cm$^2$ and duration $4 \times 10^{-12}$ sec can also produce in the materials impact ionization in the field of the light wave. Owing to the considerable intensity of the light-wave field, the electron energy loss to scattering by phonons becomes negligible and the breakdown free path is determined by the appear-

[2)]It is impossible to estimate the ionization probability in accordance with [12] for a radiation intensity higher than 5–$10^{12}$ W/cm$^2$, since the approximation does not hold for such intensities.

ance of a definite number of generations of the electron cascade during the time of the pulse. Estimates based on the classical equation of motion of the electron in the field of a light wave[13], with allowance for the dependence of the collision frequency $\nu_{eff}$ on the electron energy $\epsilon$ ($\nu_{eff} \sim \epsilon^{-3/2}$)[14] show that when a pulse of intensity $10^{14}$ W/cm$^2$ acts in a time $4 \times 10^{-12}$ sec, the electron concentration in the cascade reaches $10^{19}$ cm$^{-3}$. This suffices to breakdown the material if the initial electron concentration in the conduction band is $10^4$—$10^5$ cm$^{-3}$. Most probably the multiphoton and impact-ionization processes act simultaneously and lead to breakdown of the material within the time of action of the ultrashort pulse.

Let us examine the breakdown of ruby with color centers and the surface breakdown of ruby and glass by radiation from a mode-locked laser. As already noted[10], in the single pulse regime ruby with color centers has a much lower breakdown threshold than ruby without color centers. In the mode-locking regime, ruby without color centers also has a lower endurance than the remaining investigated materials (see the table), but the threshold energy is in this case somewhat higher than in the single-pulse regime. This circumstance can be attributed to the thermal conductivity in the interval between the ultrashort pulses. With decreasing dimension of the absorbing region, the cooling by heat conduction proceeds more rapidly and at a certain dimension of the absorbing region the action of each ultrashort pulse can be regarded as independent.

For a single pulse of duration $T_p = 10$ nsec, the thermal conductivity can be neglected for regions measuring $a > (4\kappa T_p)^{1/2} \approx 1.5\ \mu$ ($\kappa$ is the temperature-conductivity coefficient). It can be shown that when an absorbing region with approximate dimension 2 $\mu$ is heated by a sequence of pulses with an interval of 11 nsec, and account is taken of the total conductivity during the time between pulses, the required threshold energy is six times larger than in the monopulse regime. Such a value of the threshold follows from the solution of the equation of heat conduction for heat sources acting in succession (a train of picosecond pulses) in the absorbing region. However, owing to self-focusing of the radiation, the energy density in the substance should be higher and therefore 2 $\mu$ should be regarded as the upper limit of the dimension of the absorbing region.

Thermal conductivity may be the reason for the increased surface endurance of glass and ruby in the mode-locking regime. Thus, the threshold energy densities for surface breakdown in the monopulse regime ($T_p = 10$ nsec, $\lambda = 1.06\ \mu$) amount to $3.2 \times 10^2$ J/cm$^2$ for K-8 glass and $10^2$ J/cm$^2$ for ruby. In the mode-locking regime, the same quantities for K-8 ruby are $5 \times 10^3$ and $9 \times 10^2$ J/cm$^2$, respectively ($\lambda = 1.06\ \mu$). The thickness of the absorbing surface layer, according to the solution of the heat-conduction equation and the ratio of the breakdown threshold in the single pulse and in the mode-locking regimes, is approximately 4.

The main results of our investigation are as follows.

1. Breakdown of ruby, glass, leucosapphire, and fused or crystalline quartz under the action of radiation with mode locking is facilitated by the self-focusing of the radiation.

2. In the mode-locking regime, materials containing absorbing microscopic regions, reveal a higher energy threshold for breakdown by a train of picosecond pulses than in the single-pulse regime; this is attributed to the heat conduction during the interval between the ultrashort pulses.

3. In the mode-locking regime, the surface endurance of the glass and ruby is much higher than in the single-pulse regime, this being possibly due to heat conduction on the absorbing surface layer.

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[1]G. I. Bamblat, N. N. Vsevolodov, L. I. Mirkin, and N. F. Pilipetskiĭ, Zh. Eksp. Teor. Fiz., Pis'ma Red. **5**, 85 (1967) [JETP Lett. **5**, 69 (1967)].

[2]R. A. Miller and N. F. Borelli, Appl. Opt. **6**, 164 (1967).

[3]G. M. Zverev, T. N. Mikhaĭlova, V. A. Pashkov, and N. M. Solov'eva, Zh. Eksp. Teor. Fiz. **53**, 1840 (1967) [Sov. Phys. JETP **26**, 1053 (1968)].

[4]A. G. Molchanov, Fiz. Tverd. Tela **12**, 954 (1970) [Sov. Phys. Solid State **12**, 749 (1970)].

[5]M. Hercher, J. Opt. Soc. Amer. **54**, 563 (1964).

[6]G. M. Zverev and V. A. Pashkov, Zh. Eksp. Teor. Fiz. **57**, 1128 (1969) [Sov. Phys. JETP **30**, 616 (1970)].

[7]R. R. Alfano and S. L. Shapiro Phys. Rev. Lett. **24**, 592 (1970).

[8]R. Yu. Orlov, I. B. Skidan, and L. S. Telegin Zh. Eksp. Teor. Fiz. **61**, 784 (1971) [Sov. Phys. JETP **34**, 418 (1972)].

[9]A. J. DeMaria, W. H. Glenn, M. I. Brienza, and M. E. Mack, Proc. IEEE **57**, 2 (1969).

[10]T. N. Mikhaĭlova, V. A. Pashkov, and G. M. Zverev, Trudy II Vsesoyuznogo simpoziuma po nelineĭnoĭ optike (Proc. Second All-union Symposium on Nonlinear Optics), Novosibirsk, 1966.

[11]R. Y. Chiao, E. Garmire, and C. H. Townes, Phys. Rev. Lett. **13**, 479 (1964).

[12]L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)].

[13]Ya. B. Zel'dovich and Yu. I. Raĭzer, Zh. Eksp. Teor. Fiz. **47**, 1150 (1964) [Sov. Phys. JETP **20**, 772 (1965)].

[14]G. I. Skanavi, Fizika diélektrikov (Physics of Dielectrics), Fizmatgiz, 1958.

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