INVESTIGATION OF BREAKDOWN PRODUCED IN DIELECTRICS BY ULTRASHORT LASER PULSES

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Breakdown in air, KDP crystal, LGS-228 glass, and water due to a pulse train or single picosecond pulse from a neodymium laser or its second harmonic is investigated. The breakdown threshold for the second-harmonic frequency is lower than the breakdown threshold for the fundamental radiation. Nonlinear absorption in the pre-breakdown state is also observed. These observations, as well as numerical estimates, indicate that multiphoton processes make a major contribution to optical breakdown in picosecond-pulse fields. Investigation of breakdown, and in particular, study of the time variation of the optical absorption during breakdown, may yield valuable information regarding the structure of picosecond pulses.

INTRODUCTION

IN powerful fields of ultrashort light pulses radiated by a laser with self-synchronized modes, practically all dielectrics exhibit nonlinear polarizability (due to the behavior of the optical electrons) and a number of phenomena associated with it, such as stimulated scattering, self-focusing, and multiphoton absorption. An experimental investigation of these phenomena is limited, however, by optical breakdown, which arises in strong light fields. The nature of the breakdown determines whether it is possible to increase this limit in some way or another.

When giant laser pulses ($\tau = 3 \times 10^{-8}$ sec) act on dense media and gases at atmospheric pressure, the breakdown is caused as a rule by cascade ionization[1,2]. In cascade ionization, the breakdown power density depends on the pulse duration τ like $I_{cr} \sim 1/\tau$. With decreasing τ, however, the relative role of the multiphoton processes increases with increasing I_{cr}; as a result, the formula presented remains valid only for not too high values of I_{cr}[3] [1)].

The multiphoton mechanism was registered experimentally in investigations of breakdown of rarefied gases by ultrashort (5×10^{-11} sec) pulses[4,5]. Compared with optical breakdown of gases, the breakdown of condensed media by ultrashort (picosecond) pulses of light has been investigated to a much lesser degree.

In the present paper we report an experimental investigation of breakdown in the field of picosecond pulses in different media at different light-field frequencies. The results are compared with the characteristics obtained in the same media and under the same experimental conditions for breakdown by a nanosecond pulse.

1. EXPERIMENTAL RESULTS

1. Optical breakdown was investigated in the field of a picosecond-pulse neodymium-glass laser and its second harmonic. The experimental setup is shown in Fig.

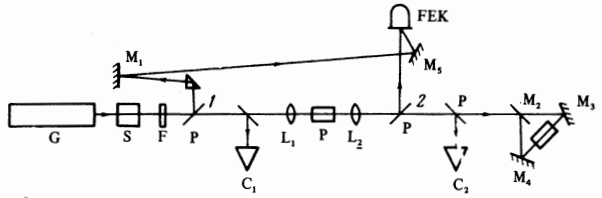

FIG. 1. Schematic diagram of setup: G–picosecond-pulse generator, S–system for separating one pulse, F–neutral filters, P–beam-splitting plates, C–calorimeters, L–lenses, D–dielectric, M–mirrors, FEK–coaxial photocell, 1–beam incident on the dielectric, 2–beam passing through the breakdown region.

1. We used in the laser a hemispherical resonator 165 cm long with a total-reflection mirror of 6m radius, and a four-lamp illuminator, while the end surfaces of the neodymium-glass active element (l = 305 cm, diameter 12 mm) were inclined at the Brewster angle. The laser emission (λ = 1.06 μ) consisted of 20 pulses of duration $(3-4) \times 10^{-12}$ sec with a repetition period 11×10^{-9} sec, and pertained to one transverse high-order mode (usually TEM_{55}) inherent in a resonator with right-angle mirrors.

To obtain the second harmonic of the neodymium laser (λ = 0.53 μ), we used a KH_2PO_4 (KDP) crystal 3 cm long, which ensured conversion of the light pulses without any appreciable change in their duration.

The breakdown was investigated in focused beams. The cross section of the focal spot was determined by photography through a microscope and amounted to 10^{-4}, 3×10^{-5}, and 3×10^{-6} cm^2 for lenses with focal lengths 70, 35, and 5 mm, respectfully. Principal attention was paid to measurements of the energies of the beam incident on the dielectric (1) and of the beam passing through the breakdown region (2). When working with picosecond pulses, the breakdown was investigated in the field of single pulses and of a train of picosecond pulses. In the latter case, an interesting possibility was afforded for the investigation of the temporal evolution of the pre-breakdown nonlinear absorption and of the absorption under breakdown conditions at times on the order of 10^{-8}–10^{-7} sec. To this end, both beams (1 and 2) are aimed, with a small relative delay, at the FEK-15

[1)] The indicated values were apparently reached in our experiments (see Sec. 2 of the present article).

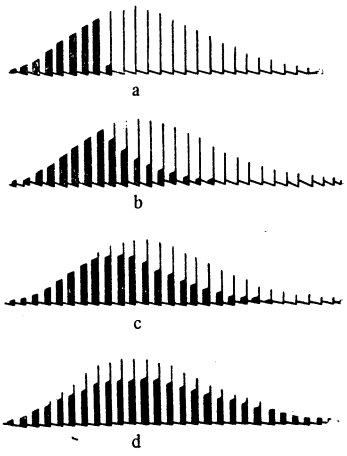

FIG. 2. Trains of picosecond pulses. Thick lines–simultaneous incidence of beams (1) and (2) on the photocell; a, b, c–breakdown, $\lambda = 1.06\mu$, a–air, b–KDP, c–glass and water, d–pre-breakdown absorption in air, $\lambda = 0.53\mu$.

photocell, which is connected to an I2-7 oscilloscope.

For comparison with picosecond breakdown, we used in the same setup a giant-pulse neodymium-glass laser (τ = 30 nsec). The breakdown was investigated in air, a KDP crystal 1 cm long, LGS-228 glass 3 cm long, and a cell with water 10 cm long.

2. We first consider the qualitative picture of the breakdown in a train of picosecond pulses, as registered by an oscilloscope. Figure 2 shows the most typical oscillograms, which depict a set of pulses ahead of and behind the breakdown region, based on the reduction of 90 oscillograms. The pulse train passing through the breakdown region characterizes, in essence, the time evolution of the optical absorption. The role of scattering and reflection, as shown by observations, is slight in this case.

It is seen from Fig. 2a that breakdown in air is produced in practice by one pulse, and is accompanied by the appearance of strong absorption that leads to complete suppression of all the succeeding pulses. The subsequently discussed energy relations also show that the air breakdown is produced by a single pulse that reaches a certain critical amplitude. Breakdown in condensed media such as KDP crystals and particularly in water and glass differs in that the absorption increases slowly. A threefold attenuation of the incdient beam is attained in KDP after 3—6 periods, in glass after 5—7 periods, and in water after more than 10 periods.

3. Useful information concerning the character of the breakdown can be obtained by measuring the dependence of the energy transmitted through the breakdown region on the incident energy. Such a dependence was measured for all the aforementioned media in the field of a train of picosecond pulses at 1.06 and 0.53 μ, in the field of single picosecond pulses at 1.06 μ, and in the field of nanosecond pulses at 1.06 μ. The corresponding results are shown in Figs. 3 and 4 and in the table.

Let us turn to the results of breakdown by a train of pulses. At the threshold of the breakdown of air (and of the KDP crystal) by radiation of 1.06 μ wavelength, the transmitted-energy curves have a sharp descent. This is followed by a horizontal or slightly inclined section (Figs. 3a and 3b). Such a dependence, as shown by calculation, should obtain also for a train of pulses with exponential or, respectively, a Gaussian envelope of the leading front of the train, if the breakdown arises as soon as the amplitude of one of the pulses reaches a certain critical power and if all the remaining pulses are completely absorbed in the resultant plasma. The

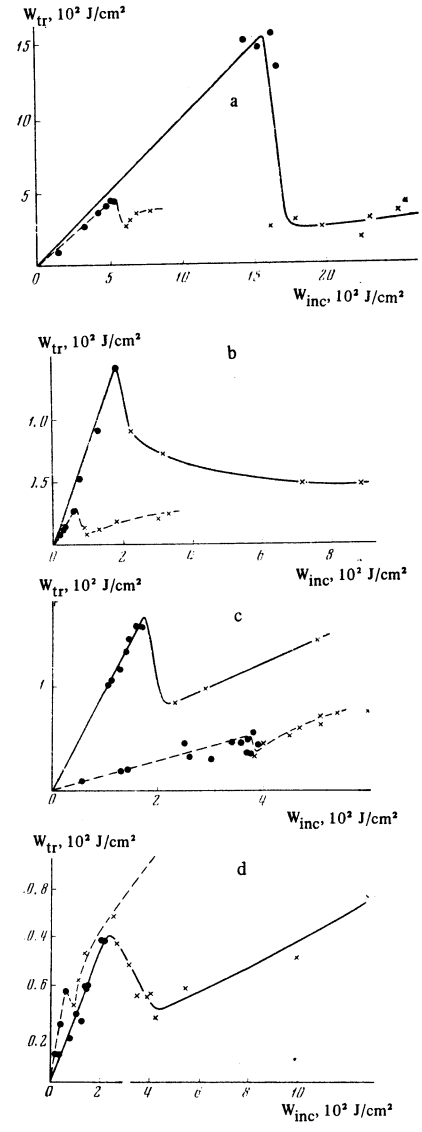

FIG. 3. Breakdown by a train of pulses: a–air, b–KDP, c–glass, d–water. Solid lines $\lambda = 1.06\mu$, dashed–$\lambda = 0.53\mu$; ●–no breakdown, X–breakdown. W_{inc}–density of incident energy, W_{tr}–density of transmitted energy.

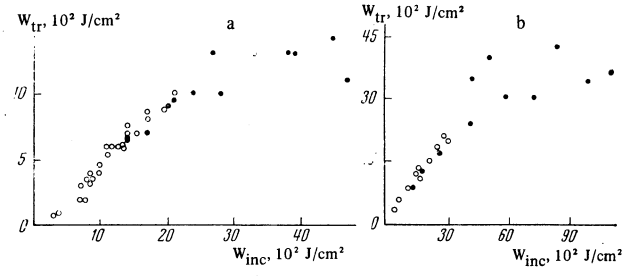

FIG. 4. Breakdown by one pulse: a–air, b–KDP, O–no breakdown, ●–breakdown.

Threshold energy densities W_{inc}, 10^2 J-cm^{-2}

	f of lens	Monopulse, τ = 30 nsec	Train of pulses N_{eff} = 10, τ_{pulse} = 3 psec		One pulse, τ = 3 psec
		λ = 1.06 μ	λ = 1.06 μ	λ = 0.53 μ	λ = 1.06 μ
Air	5		170		20
	35		20—30	12	
	70	25—35	20	6	
KDP	70	5—10	2	0.6--0.9	0,2—0.3
Glass	70	10—25	1.4—2	1.1	0.3
Water	70	>50	1.5	0.8	0.15

described picture agrees with the time evolution of the breakdown shown in Fig. 2a. In glass or water, and at 0.53 μ also in all the mentioned media, the picture is different: the transmitted energy again increases with increasing incident energy at a slight excess above the breakdown threshold.

The possibility of transmission of the breakdown-producing pulse without significant energy absorption in the breakdown region is demonstrated also by experiments on the breakdown by one selected pulse (see Fig. 4). We see that some of the pulses causing the breakdown (marked with crosses) lie on a straight line passing through the points at which there is no breakdown. This corresponds to breakdown without noticeable energy absorption in the pulse. The points deviating from this straight line correspond to partial absorption of energy. In the latter case, there is apparently a fine structure of the pulse, which cannot be resolved by the oscilloscope, when some of the pulses are transmitted and some are absorbed in the breakdown region.

A comparison of the thresholds for breakdown by a pulse train and by a single pulse (see the table) shows that they differ by 7—10 times not only in air but also in condensed media. This is evidence in favor of inertialess breakdown by the very first pulse that has reached threshold power. Indeed, as shown by observations, this is indeed the ratio of the energy of the entire train to the single largest pulse of the train (at the breakdown threshold, it is precisely this pulse which excites the breakdown). Accordingly, in the subsequent estimates of the threshold power, wherever there were no data on breakdown by a single pulse (air, s = 70 mm), we assumed $I_{cr} = 0.1 W_{cr}/\tau$, where W_{cr} is the threshold energy density for breakdown by a train of pulses.

4. It is perfectly obvious that the evolution of the breakdown and the form of the energy dependences can be influenced by self-focusing. For example, when the beam becomes laminated during the self-focusing process, the transmission of energy beyond the breakdown region (Figs. 3c and 3d) may be connected with the nonsimultaneity of the occurrence of the focal points over the cross section of the beam. To get around this difficulty, we used lenses with short focal lengths, shorter than the self-focusing length under our conditions[2)].

The main experimentally obtained material is summarized in the table. We took the breakdown threshold to be the minimum incident energy at which a flash of radiation was observed in the focal region of the lens. In the case of breakdown by a train of pulses, this quantity coincides with the start of the decrease on the transmitted-energy curve (see Sec. 3). In solid dielectrics, breakdown is usually (but not always) accompanied by damage, which can be observed in the bright beam of a helium-neon laser. In calculating the breakdown threshold of water at the fundamental frequency and of LGS glass at the second harmonic, we took into consideration the linear absorption in these media. The relative accuracy of the measurements was 20%.

2. DISCUSSION

An analysis of the table shows that in breakdown of dielectrics by picosecond pulses of light there appear a number of singularities that distinguish it from breakdown by pulses of nanosecond duration.

The threshold energy densities for picosecond pulses are smaller by one or two orders of magnitude than for nanosecond pulses. This indicates that at the available durations and powers of the picosecond pulses, the relation $I_{cr} \sim 1/\tau$ is no longer satisfied and is violated in the direction of decreasing I_{cr}. Nonetheless, the threshold power density of picosecond pulses exceeds the threshold power of nanosecond pulses by several orders of magnitude. Thus, as follows from the table, this excess amounts to three orders of magnitude for air and two orders for glass.

In the field of picosecond pulses, the thresholds of breakdown at the second harmonic (λ = 0.53 μ) are lower than at the fundamental frequency (λ = 1.06 μ), whereas in experiments on breakdown by nanosecond pulses the inverse relations are observed[6-8]. Notice should also be taken of the strong nonlinear absorption observed in air at the second-harmonic frequency and at powers close to breakdown (Fig. 2d).

These singularities enable us to speak of an appreciable contribution made by multiphoton ionization to the development of breakdown of dielectrics by picosecond light pulses[2,3].

One cannot exclude, however, the possibility that multiphoton ionization affects only the components with the lower ionization potential, which serve as electron sources, and that the subsequent development of the breakdown proceeds via impact ionization. In air, disregarding small impurities, this component is made up of the oxygen molecules[9] (I_0 = 12.15 eV), and in glass—of the oxygen ions which are bound only to one SiO_4 tetrahedron[10] (I_0 = 5.4 eV). It can be assumed that in KDP crystals, too, the oxygen ions in the PO_4 tetrahedra have a close value of the ionization potential.

Taking the ionization potentials indicated above to be the ionization potentials of the dielectrics themselves, we find that at the neodymium-laser frequency ($h\nu$ = 1.17 eV) the ionization of air is an 11-quantum process, and that of glass or KDP a 5-quantum process; at the second-harmonic frequency ($h\nu$ = 2.34 eV), the ionization of air is a 6-quantum process and that of glass and KDP a three-quantum process.

Estimates performed in accordance with[11] show that the ionization of air by pulses of 3×10^{-12} sec duration at neodymium-laser frequency occurs at a power 2.3×10^{13} W-cm^{-2}, and at the second harmonic at a power 4×10^{13} W-cm^{-2}. Ionization of glass or of the KDP crystal requires 7.6×10^{12} and 6.8×10^{12} W-cm^{-2} at the

[2)]To the contrary, when using long-focus lenses (f = 50 cm) a spectrum broadening (up to 1000 cm^{-1}) was seen to accompany self-focusing in glass.

fundamental frequency and at the second harmonic, respectively.

Assuming that the train of a picosecond laser contains approximately 10 pulses of equal magnitude, we find that at the air breakdown threshold, the average power densities in the experiment were 7×10^{13} and 3×10^{13} W-cm^{-2} at the fundamental frequency and at the second harmonic; for breakdown of KDP and of glass, the average power densities were $(7-10) \times 10^{12}$ and $(4-6) \times 10^{12}$ W-cm^{-2} at the fundamental frequency and at the second harmonic. These figures are in satisfactory agreement with the results of the calculation, and thus confirm the qualitative conclusion that multiphoton ionization takes part in the development of the breakdown.

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[1] G. M. Zverev, T. N. Mikhaĭlova, V. A. Pashkov and N. M. Solov'eva, Zh. Eksp. Teor. Fiz. 53, 1849 (1967) [Sov. Phys.-JETP 26, 1053 (1968)].

[2] Yu. P. Raĭzer, Usp. Fiz. Nauk 87, 29 (1965) [Sov. Phys.-Uspekhi 8, 650 (1966)].

[3] F. V. Bunkin and A. M. Prokhorov, Zh. Eksp. Teor. Fiz. 52, 1610 (1967) [Sov. Phys.-JETP 25, 1072 (1967)].

[4] I. K. Krasyuk, P. P. Pashinin and A. M. Prokhorov, ZhETF Pis. Red. 9, 581 (1969) [JETP Lett. 9, 354 (1969)].

[5] I. K. Krasyuk, P. P. Pashinin and A. M. Prokhorov, Zh. Eksp. Teor. Fiz. 58, 1606 (1970) [Sov. Phys.-JETP 31, 860 (1970)].

[6] S. A. Akhmanov, A. I. Kovrigin, M. M. Strukov and R. V. Khokhlov, ZhETF Pis. Red. 1, No. 1, 42 (1965) [JETP Lett. 1, 25 (1965)].

[7] H. T. Busher, R. G. Tomlinson and E. K. Damon, Phys. Rev. Lett. 15, 847, 1965.

[8] G. M. Zverev, E. A. Levchuk and É. K. Maldutis, Zh. Eksp. Teor. Fiz. 57, 730 (1969) [Sov. Phys.-JETP 30, 400 (1970)].

[9] Énergii razryva khimicheskikh svyazeĭ (Rupture Energies of Chemical Bonds), a Handbook, M. Kondrat'ev, Editor, Moscow, 1962.

[10] B. S. Sharma and K. E. Rieckhoff, Canad. J. Phys. 45, 378, 1967.

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

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