STRUCTURE OF A SPARK PRODUCED BY A PICOSECOND LASER PULSE FOCUSED IN A GAS

F. V. BUNKIN, I. K. KRASYUK, V. M. MARCHENKO, P. P. PASHININ, and A. M. PROKHOROV

P. N. Lebedev Physics Institute, USSR Academy of Sciences

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A study has been made of the formation of points of strong nonlinear scattering of laser radiation and of breakdown in air, nitrogen, and argon due to a focused 20–100 psec pulse of ruby radiation of about 2×10^9 W. The mechanism of this phenomenon, which is connected with self-focusing of laser radiation in a gas, is discussed. Experimental data on the breakdown thresholds obtained with short and long focal length lenses are analyzed. The role of self-focusing of laser radiation in the development of breakdown at optical frequencies is evaluated.

A relatively large number of experimental and theoretical papers has appeared in recent years on the development of breakdown in gases at optical frequencies.[1-8] Although, under certain definite experimental conditions, the overall development of optical breakdown resembles microwave breakdown in gases, there is still no satisfactory quantitative agreement between theory and experiment. Moreover, some experimental data cannot be explained within the classical microwave breakdown theory even in a qualitative fashion. It would appear, therefore, that a complete explanation of the phenomenon will require more careful inclusion of the particular features of the optical region. For example, transition from nanosecond to picosecond laser pulses in a certain range of gas pressures ensures that the multiphoton ionization of atoms and molecules becomes the dominant mechanism.[9] The dependence of the breakdown threshold on the size of the focused beam in the nanosecond region, where all the estimates indicate that diffusion losses can be neglected, has not as yet been explained.

There have been recent suggestions in the literature that self-focusing may be possible during breakdown in gases.[4,5] These experimental data, however, do not provide unambiguous indication as to whether self-focusing appears prior to the spark and, consequently, whether it has an essential effect on the breakdown threshold. It is also not clear what is the nonlinear mechanism leading to self-focusing.

In this paper we report experimental studies of breakdown in air, nitrogen, and argon at atmospheric pressures, using picosecond laser pulses. The structure of the laser spark and the dynamics of the formation of breakdown points were investigated using a fast image converter with high light sensitivity.

The experimental setup is illustrated in Fig. 1. The traveling-wave ring ruby laser[10] generated a series of picosecond pulses. One of these pulses was isolated by an electro-optical shutter placed in a resonator. The length of the pulse thus isolated was 20–100 psec. The necessary power was produced by using the optical amplifier 2 consisting of two ruby elements, 240 mm long and 15 mm in diameter. The radiation energy and its time dependence were monitored by a calibrated

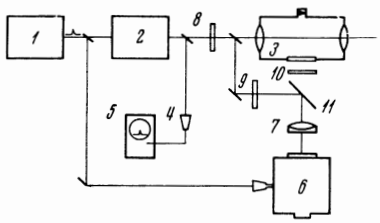

FIG. 1. Experimental setup: 1) ruby laser, 2) optical amplifier, 3) chamber with focusing lens, 4) coaxial photocell, 5) fast oscillograph, 6) image converter, 7) objective, 8)-9) optical filters, 11) glass plate.

photocell 4, working in combination with a fast oscillograph 5 to a resolution of 0.2 nsec. The laser radiation was focused in the chamber 3 by a lens having a focal length of 15 cm. The transparent lateral window of the chamber could be used to observe the focal region at right-angles to the direction of propagation of the laser beam. The objective 7 imaged this region on the photocathode of the image-converter 6. A proportion of the laser radiation was, at the same time, directed onto the photocathode to determine the time structure of the pulse with a high resolution.

The intensity distribution of the optical radiation along the caustic surface of the focusing lens was determined as follows. The laser radiation was focused in a chamber containing a solution of rhodamine 6G in ethyl alcohol. The glow of the solution due to two-photon absorption of the laser beam produced in this way was recorded on a photographic film. Analysis of these photographs showed that the beam had a Gaussian shape in each cross section. Figure 2 shows a photograph of the beam (b), the measured dependence of the laser beam diameter d (full width at half height), and its intensity I as functions of distance (c). It is clear that the minimum transverse beam diameter in the focusing region is 0.2 mm, which corresponds to a beam divergence of 4′. The intensity falls by a factor of two over a distance of 2.2 mm from the focal plane. The laser beam power was chosen so as to ensure that the caustic surface was not distorted by possible self-focusing in the liquid.

The time-integrated photograph of breakdown in the visible region at atmospheric pressure in air and nitrogen is shown in Fig. 2a, which is quite typical for these

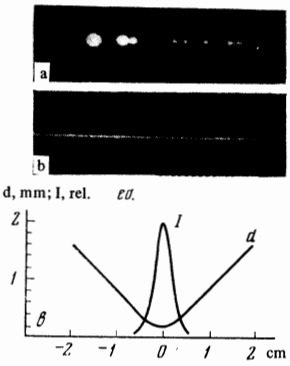

FIG. 2. Time-integrated photograph of breakdown in air (a), photograph of the glow from rhodamine 6G in ethyl alcohol (b), and the distribution of the laser intensity and beam diameter near the focal spot of the lens (c). The scale along the abscissa axis is the same in all cases. A reducing neutral light filter is placed in front of the breakdown region behind the focal point in case a. The laser beam travels from left to right.

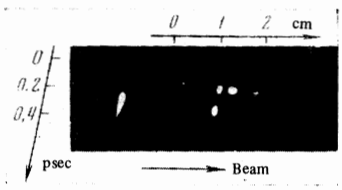

FIG. 3. Photograph taken from the image-converter screen showing the time and space distribution of points of nonlinear scattering of laser radiation in air at atmospheric pressure. An interference filter transmitting at the ruby laser wavelength was placed in front of the image-converter objective. The laser pulse is on the left of the photograph.

gases. It is clear that the longitudinal size of the spark is substantially greater than the size of the region occupied by the high-intensity field in the case of linear focusing. The breakdown points are usually located at a relatively large distance from one another. Quite frequently there is a regular structure with mean distance between the points of 1.5–2 mm. Photographs with higher magnification showed that the minimum size of an individual breakdown point could be less than 15 μ.

The breakdown was investigated with the image-converter both in the light of the scattered laser radiation, using an interference filter with a bandwidth of 70 Å, and in the intrinsic light of the spark. It was found that when the laser radiation was focused down at a peak power of about 1.5×10^9 W in air or in nitrogen at atmospheric pressure, there was strong nonlinear scattering at points which was unaccompanied by breakdown. Breakdown occurred at one or more of these points when the laser pulse power was slightly increased.

It is interesting to note that, in the case of the long focal length lenses, the threshold intensity for nitrogen and air, $I_{th} = 3.5 \times 10^{12}$ W/cm^2, was substantially lower than in the case of the short focal length lens (f = 2 cm and $d = 1.7 \times 10^{-3}$ cm[9]), when it was 1.5×10^{14} W/cm^2. A similar effect was observed in the case of breakdown produced in gases by nanosecond pulses.[3] An attempt was made in[3] to explain this result by anomalous diffusion-like losses. In our case, however, the effect of diffusion losses can be entirely neglected for picosecond pulses.

The points of scattering in the gases which we have investigated appear in temporal succession in the direction of propagation of the laser beam. Figure 3 shows a photograph taken from the image-converter screen, showing the appearance of these scattering points in air. The slope of the straight line passing through the points plotted on the time-position plane along the beam axis corresponds to the velocity of propagation of the laser pulse. The length of the laser pulse for this particular case is 70 psec and the length of the scattered pulse is 30 psec. An additional pulse of scattered light occasionally appears in air and in nitrogen at one or a number of points at a certain finite time after the first pulse. This is illustrated in Fig. 3. Here, the additional pulse of scattered light appears after 160 psec following the first pulse, i.e., it appears on the tail of the laser pulse.

The experimental data show that the discrete breakdown structure which we have observed is associated with self-focusing of the laser beam. This is supported by a comparison of the characteristic size of the breakdown region with the transverse and longitudinal size of the focal caustic, by the appearance of nonlinear scattering of laser radiation near the breakdown threshold but without the appearance of the laser spark, and by the dynamics of the development of points of nonlinear scattering or breakdown, both in time and space.

The experimental data can be explained if we assume that the nonlinearity leading to self-focusing occurs in two stages. During the first stage, it is the nonlinearity of the electronic polarizability of the molecules which is important, and this is followed by the appearance of nonlinearity due to the presence of excited molecules. Calculations of the nonlinear polarizability of atoms and molecules based on the data reported in[11,12] have shown that the laser radiation power P employed in our experiments was close to the critical power P_{crit} necessary for self-focusing.

The possibility of self-focusing using excited molecules was considered in[13,14]. Under our experimental conditions this may occur as follows. Cascade ionization begins to develop in the strong field of laser radiation near the threshold, and the electron density rapidly increases. According to the data reported by Chalmeton,[15] even at intensities lower by an order of magnitude than the breakdown threshold value there is an appreciable density of free electrons near the focus. Electrons accelerated by the laser field take part in inelastic collisions and can excite gas molecules and atoms. Since the polarizability of the excited molecules and atoms is much greater than the polarizability of unexcited particles, the attendant changes in the refractive index turn out to be sufficient for the appearance of the self-focusing effect at electron densities much lower than the threshold value. Approximate calculations show that the excited-state lifetimes of air and nitrogen molecules may be sufficient to ensure self-focusing during the time of the laser pulse.

The introduction of the complicated self-focusing mechanism is necessitated by the fact that each of the component mechanisms will not explain all the experimental data. Because of the nonlinear electronic polarization of the molecules or atoms, the laser beam contracts in the gas near the linear focus down to a diameter d_c which is less than the diameter d of the caustic. Further contraction down to the diameter observed experimentally occurs as a result of self-focusing on excited molecules or atoms. The change in the refractive index which is connected with the increase in the number of excited molecules or atoms at time t is given by

$$\delta n_e = 2\pi(\varkappa_e - \varkappa_0)N_e = Ae^{\alpha t t}, \quad (1)$$

where κ_e and κ_0 are the polarizabilities of the excited and unexcited atoms or molecules, respectively, N_e is the number of excited atoms at time t, αI is the growth constant of the cascade, I is the intensity of the laser radiation, and A is a constant. It is well known[16] that the condition for self-focusing in a gas[1)] is

$$\delta n_{nl} \approx \delta n_e \geqslant 1/2 (0.61\lambda_0 / d)^2. \quad (2)$$

In this expression λ_0 is the laser radiation wavelength, d is the beam diameter, and δn_{nl} is the necessary nonlinear change in the refractive index.

By using Eq. (1) we can rewrite Eq. (2) in the form

$$A \exp\{\alpha I_c \tau\} \geqslant 1/2 (0.61\lambda_0 / d_c)^2. \quad (3)$$

In this expression the self-focusing condition is written for the time $t = \tau$, where τ is the length of the laser pulse, and I_c is the intensity of the beam contracted down to the diameter d_c. The quantity that one usually measures is $I_{exp} = P/d^2$, where P is the laser power and d the diameter of the focal spot in the case of linear focusing. It is readily seen that $I_c = I_{exp} d^2/d_c^2$ and hence condition (3) can be written in the form

$$A \exp\left\{\frac{\alpha I_{exp} d^2 \tau}{d_c^2}\right\} = \frac{1}{2}\left(\frac{0.61\lambda_0}{d_c}\right)^2. \quad (4)$$

If we suppose that d_c is not very dependent on the focal length f of the lens, and take the logarithm of both sides of Eq. (4), we obtain $I_{exp}^{th} = B/d^2$, where B is a constant and I_{exp}^{th} is the measured threshold intensity for breakdown, or a quantity close to it, at which the nonlinear scattering points appear. It follows that the above mechanism can, at least in principle, explain the dependence of the threshold intensity for breakdown on the diameter of the focal spot, which we have observed. In fact, this dependence is related to the method of determination of I_{exp}^{th}.

For breakdown in the case of nanosecond pulses it is possible that electrostriction is responsible for the nonlinearity during the first stage. The observed dependence of the threshold breakdown intensity on the focal-spot diameter can then again be explained.

Both in our experiments and in the experiments with nanosecond pulses,[4] the observed nonlinear scattering prior to breakdown appears to be connected with the excitation of atoms or molecules. The additional scattered-light pulse observed in our experiments may be due to the transition of the material from the excited state to the plasma state. In fact, experiment shows that the appearance of the additional pulse is accompanied by breakdown.

We have also investigated breakdown in argon under the above experimental conditions. Although the overall picture of the breakdown structure is the same as for air and nitrogen, there are some essential differences. It was found that the observed laser-radiation intensity in the focal region is lower by a factor of only two than the intensity for which breakdown is produced with the short focal length lens (f = 2 cm and d = 1.7 $\times 10^{-3}$ cm[9]). As in[9], the breakdown threshold is unaltered when the pressure is reduced down to 0.4 atm. Laser-radiation scattering in argon is usually accompanied by breakdown at the scattering points. The experimental data indicate that the spark structure development in argon is also determined by self-focusing of the laser radiation. However, the nonlinear mechanism leading to self-focusing in argon is not as yet clear. Although the threshold power necessary for the appearance of self-focusing points is close to the threshold power for self-focusing due to nonlinear electronic polarizability of the excited argon atoms, this mechanism alone will not explain our experimental data. The fact that the breakdown threshold is independent of pressure means that this mechanism cannot be the only one as far as self-focusing is concerned. The same argument excludes the strictional mechanism. It would, therefore, seem that more detailed experimental studies will be necessary before the true nature of the phenomenon can be established.

Since it can now be regarded as established that the self-focusing mechanism is important in the development of breakdown at optical frequencies, this phenomenon must be taken into account in estimating the threshold breakdown intensities, and also in the analysis of experimental data obtained earlier.

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[1)] The reduction in the refractive index due to the presence of free electrons will be small in comparison with δn_e if the ionization cross section is less than the excitation cross section, or if the laser frequency is close to the frequency of transition from the excited state.

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