

Optical breakdown of transparent media containing microinhomogeneities

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A model of a transparent medium with randomly distributed absorbing inhomogeneities is used to analyze optical breakdown of transparent dielectrics and to explain the experimentally observed dependence of the breakdown threshold on pulse length and focusing conditions. A statistical approach to the breakdown problem is formulated; the distribution of breakdown probabilities is obtained and the averages characterizing the threshold are calculated.

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1. INTRODUCTION

In most cases the experimentally measured thresholds for optical breakdown of transparent dielectrics^[1–3] turn out to be substantially lower than calculated theoretically taking into account processes of impact ionization and multiphoton absorption.^[3–5] The cause of this lack of correspondence is apparently the extraordinary simplification of the theoretical model of a transparent dielectric, in which the material is assumed to be optically uniform. At the same time it is known that the optical breakdown threshold is substantially affected by spatial inhomogeneity of the absorption coefficient. A well known example of an optically inhomogeneous medium is the platinum-containing glasses, for which optical breakdown is found experimentally to be initiated by the strongly absorbing platinum inclusions.^[6] Another example of the strong influence of absorbing inhomogeneities on optical breakdown is given by the experimental results of Butenin and Kogan,^[7] from which it follows that the optical breakdown threshold of a liquid dielectric increases monotonically as it is purified of microscopic solid particles.

In the examples presented the existence of absorbing inhomogeneities in the medium is not in question and the properties of the inhomogeneities are known. In most optical media the existence of absorbing inhomogeneities is much less obvious—they are difficult to observe as the result of the smallness of the absorption produced by them. Recently, however, direct experimental proofs have appeared of the existence of local regions of increased absorption in a number of dielectric coatings^[8] and glasses.^[9,10] In addition, the literature contains data which indicate that absorbing inhomogeneities associated directly with structural features of the material exist in compensated and amorphous semiconductors.^[11]

Recently several studies appeared in which attempts are made to develop a theory of the optical breakdown of gases,^[12] films,^[13] semiconductors, and glasses^[14] containing absorbing inhomogeneities of small size. In the present work we have also attempted to construct a theory of optical breakdown of condensed transparent

media with absorbing inhomogeneities which permits explanation from a unified point of view of a large number of experimental data relating to the optical breakdown of crystals and glasses under the influence of laser radiation.

2. EXPERIMENTAL RELATIONS

Before proceeding to the theoretical analysis of optical breakdown of media with absorbing inhomogeneities, we shall enumerate some basic regularities which have been established in the experimental study of the breakdown of the surface layer of transparent dielectrics (we discuss the results on surface breakdown in order not to involve the question of the possible effect of self-focusing of the radiation). We note the following experimental facts which the theory of breakdown must explain.

1) The dependence of the threshold intensity of radiation on the duration of the laser pulse. This dependence is observed for all materials studied, both crystalline and amorphous. The results of a systematic study carried out for ruby crystals^[15] and K-8 optical glass^[16] are shown in Fig. 1. A characteristic feature is the existence in the graphs of points near which a change in the form of the time dependence occurs.

2) The dependence of the threshold for optical breakdown of the surface of transparent dielectrics on the size of the region irradiated. This dependence apparently is universal in nature; in addition to optical glasses, it has been observed, for example, in the action of radiation on the retina of the eye.^[13] The existence of a sharp size dependence explains, in particular,

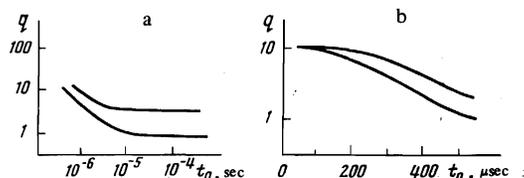


FIG. 1. Dependence of breakdown threshold on duration of radiation action: a) for ruby crystals according to the data of Ref. 15; b) for K-8 glass according to the data of Ref. 16.

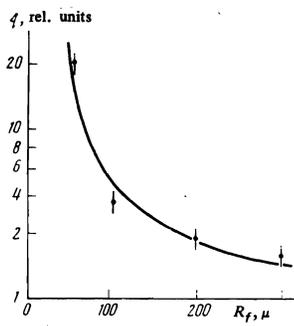


FIG. 2. Size dependence of optical breakdown threshold for K-8 glass. [17]

the significant spread in the value of the threshold intensity of optical breakdown given by different authors for the same materials. Typical dependences of the threshold for breakdown on the size of the focal spot are given in Fig. 2. [17]

3) Recent studies have shown that in a number of cases optical breakdown of dielectrics does not have a threshold nature, [18,19] but that a finite probability exists for occurrence of breakdown over a wide range of intensity of the incident radiation. It is obvious that in this case optical breakdown can be characterized by specifying the threshold intensity of light only with an accuracy equal to the width of this interval. The size of the interval in which the principal change of breakdown probability occurs changes from material to material, but it is still not completely clear just what parameters of the material are responsible for these changes. In Fig. 3 we have shown an example of the dependence of the probability for optical breakdown of the surface of glass on the intensity of the incident light flux. [19]

3. THEORETICAL ANALYSIS OF BREAKDOWN

In analysis of the optical breakdown of transparent media containing absorbing inhomogeneities, we must keep in mind two possible paths of development of breakdown. In the presence of inhomogeneities of sufficiently large size the most probable breakdown mechanism is local melting of the material near inhomogeneities and formation of cracks in the surrounding material as the result of mechanical stresses which arise. An analysis of the heating of inhomogeneities and the material surrounding them under the influence of light has been carried out in Refs. 15, 16, and 20, and one of these studies [20] took into account the temperature dependence of the absorption coefficient of the inhomogeneity. If the size of the inhomogeneities is sufficiently small, the experimentally observed breakdown pattern cannot be interpreted as the direct consequence of mechanical stresses due to heating of the inhomogeneities. In this case the damage would have to have primarily a local nature and would resemble local melting and formation of microcavities in solid materials under the action of fast particles. [21] It is possible, however, to have another breakdown mechanism due to the fact that heating of the medium in the vicinity of an absorbing inhomogeneity should produce some additional absorption of

light, which is equivalent to an increase in the size of the inhomogeneity. Under certain conditions this process represents the propagation of a wave of heating from the surface of the inhomogeneity to the volume of the medium. The experimentally observed macroscopic breakdown can be related to a wave of this type arising as the result of instability in the thermal behavior of the absorbing inclusion.

In order to obtain a quantitative criterion for breakdown, we shall discuss a small absorbing inclusion located in a transparent medium placed in a radiation field. We shall assume that the laser pulse is rather long. In a stationary regime the quantity of energy received by the inhomogeneity in absorption of light is equal to the quantity given up by it to the surrounding medium. We designate by q the radiation energy flux density near the inhomogeneity. The thermal flux density given up by an inhomogeneity of size R to the surrounding medium can then be set equal to $\alpha(R)q$, where the coefficient $\alpha(R)$ is easily obtained from solution of the problem of diffraction of light by the inhomogeneity. We will not dwell on discussion of the form of α , since the analysis given below does not depend on the particular form of $\alpha(R)$. Detailed calculations of the function $\alpha(R)$ for metallic and dielectric inclusions of various size have been carried out by several groups. [22-24]

As we have already noted, heating of the medium near an inhomogeneity will lead to appearance of an additional absorption of light. It is easy to show that under the conditions realized experimentally in the medium near an inhomogeneity, thermodynamic equilibrium can be established, so that the local absorption coefficient is determined by the local temperature. Usually the temperature dependence of the absorption coefficient of the media considered here can be described by the formula

$$\chi(T) = \chi_0 \exp[-E/T],$$

where $2E = E_g$ is the width of the forbidden zone of the material.

The stationary temperature distribution $T_0(r)$ near an inhomogeneity of size R is determined from solution of the heat conduction equation [1]

$$\chi \nabla^2 T_0 + \chi(T_0)q = 0 \quad (1)$$

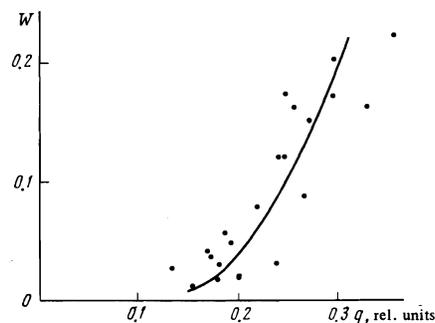


FIG. 3. Dependence of optical breakdown probability for the surface of F-2 glass on the intensity of radiation. [19]

with the boundary condition

$$-\chi \nabla T_0 = \alpha q \quad \text{for } r=R. \quad (2)$$

It has been shown previously^[25] that the stationary temperature distribution $T_0(r)$ turns out to be unstable to small perturbations if the flux density q exceeds some critical value. Without dwelling on the details of investigation of the stability of the solution, we shall point out that the condition for loss of stability with any sufficiently rapid dependence $\kappa(T)$ has the following form:

$$R\kappa(T_0(R))/\alpha(R) \geq 1. \quad (3)$$

The breakdown criterion (3) relates the inhomogeneity size R with the mean free path of the radiation in the heated medium at its surface κ^{-1} . It can be seen from Eq. (3) that the stationary temperature distribution is unstable when the mean free path of the radiation is still significantly greater than the size of the inhomogeneity, i. e., the additional absorption due to heating of the medium remains small.

Substitution into the condition (3) of the solution of the boundary value problem (1) and (2) leads to the following expression for the threshold of thermal breakdown of a medium initiated by an absorbing inhomogeneity of size R :

$$q^*(R) = \frac{E\chi}{\alpha(R)R} \left[\ln \frac{\kappa R}{\alpha(R)} \right]^{-1} \approx \frac{E\chi}{\alpha(R)R}. \quad (4)$$

This formula is in reasonable agreement with the experimental data on optical breakdown of crystals and glasses.

As can be seen from Eq. (4), the threshold of breakdown initiated by an individual inclusion depends substantially on its size (absorption cross section). If several inclusions fall in the focal volume, a determining role in the origin of breakdown will be played by the most strongly absorbing of them. A nontrivial case arises in the presence in the focal volume of a large number of inclusions of approximately the same size. It is easy to understand that the breakdown threshold in this case should be lower than for an isolated inclusion. The reduction of the threshold can be calculated by using the analogy noted in Ref. 25 between the stability problem and the quantum-mechanical problem of a bound state in a given potential field. For simplicity we shall give calculations for the case of identical inclusions; generalization to an arbitrary distribution in the absorption cross sections presents no fundamental difficulties.

Let the intensity of laser radiation be constant over the focal volume and contain in a volume V a number of absorbing particles $N = nV$. The temperature field is described as before by Eq. (1) with the boundary conditions (2), which now are given at the surface of all inclusions contained in the focal volume. To investigate the stability of this solution of the boundary value problem formulated, we shall discuss the nonstationary heat-conduction problem. We set

$$\Theta(r, t) = T(r, t) - T_0(r) \quad (\Theta \ll T_0)$$

and, linearizing the heat conduction equation, we arrive at an equation for $\Theta(r, t)$:

$$\rho c \frac{\partial \Theta}{\partial t} = \chi \nabla^2 \Theta + \Theta F(r), \quad (5)$$

where the function $F(r) = q d\kappa(T_0(r))/dT$ has a very complicated form. However, for solution of the stability problem there is no need to know the function $F(r)$ in detail.

After separation of variables in Eq. (5) the stability problem reduces to the quantum-mechanical problem of the existence of a negative eigenvalue in the spectrum of the Schrödinger equation

$$\nabla^2 \bar{\Psi} + [E - U(r)] \bar{\Psi} = 0 \quad (6)$$

with potential

$$U(r) = -F(r)/\chi. \quad (7)$$

The qualitative features of the potential $U(r)$ are easy to establish without calculating $T_0(r)$. It is evident that the stationary temperature has maxima near the surface of all absorbing inhomogeneities. Since the function $\kappa'(T_0)$ is extraordinarily sharp, the potential turns out to be a smooth function far from the inclusions and has sharp minima near their surface. This real potential can be replaced by a model potential consisting of N narrow and deep potential wells located in spherical layers $R - \delta < r < R + \delta$ whose centers are located at random points \mathbf{r}_i . In view of the fact that the total volume of inclusions is much less than the size of the focal volume V , the potential (7) is equal to zero over practically the entire focal region; the solution of the Schrödinger equation in this case is logically sought in the form (see Ref. 26)

$$\Psi(r) = \sum_i \frac{\exp\{-k|r-r_i|\}}{|r-r_i|}. \quad (8)$$

The parameter k , which is related to the eigenvalue of Eq. (6) by the expression $E = -k^2$, must be found from the condition of matching the external solution (8) with the solution valid inside the spherical layers surrounding the particles. The latter can be easily obtained as follows. Write Eq. (6) in spherical coordinates with origin at the point \mathbf{r}_i ; make the substitution $\varphi = r\Psi$ and integrate the equation obtained for φ term by term over r inside the spherical layer with the boundary condition $\Psi'|_{r=R} = 0$. In carrying out the integration it is important that the wave function of the ground state with energy close to zero essentially does not change inside the potential layer. As a result of the integration we obtain an expression for the logarithmic derivative of the function φ at the outer boundary of the layer:

$$z = \frac{\varphi'(R+\delta)}{\varphi(R+\delta)} = R^{-1} + \int_R^\infty U_1(r) dr,$$

where $U_1(r)$ is the potential well near the point \mathbf{r}_i . Matching with the external solution is carried out in the following way. We place the origin of coordinates at one of the points \mathbf{r}_i , for example, at \mathbf{r}_1 . In the vicinity of this point

$$\psi \approx \frac{1}{|r-r_i|} - k + \sum_i \frac{\exp\{-k|r_i-r_i|\}}{|r_i-r_i|},$$

from which we obtain the relation

$$z = -k + \sum_i \frac{\exp\{-k|r_i-r_i|\}}{|r_i-r_i|}.$$

Since we are interested in small values of $k(k^3 \ll n)$, we can replace the summation by integration over the focal volume. Here the averaging operation is automatically carried out. After simple calculations we obtain an expression for k :

$$k = R^{-1} - \int_{\pi}^{\infty} U_i(r) dr + 2\pi n R_f^2,$$

where R_f is the radius of the focal volume, and also the instability condition:

$$\frac{R\alpha[T_0(R)]}{\alpha(R)} + 2\pi n R_f^2 R \geq 1.$$

For $n=0$ it goes over to the condition written out previously for breakdown at one particle, Eq. (3). In the case $nV > R_f/R$ the threshold is substantially reduced as a result of the fact that around each particle the average temperature turns out to be higher than the temperature beyond the limits of the focal volume, as a consequence of which the development of instability is facilitated.

It follows from what has been said that the experimentally measurable threshold for breakdown of a material containing absorbing particles should depend on the size of the particles which fall in the region with the greatest intensity of light. If the medium contains inhomogeneities of different sizes (different absorption cross sections), the breakdown threshold is no longer a completely defined quantity and we can speak only of the probability of breakdown under given conditions. Thus, breakdown of a medium containing absorbing inhomogeneities should be described by statistical methods.

Let the distribution of the intensity of radiation have the form $q(\mathbf{r}) = q_0 \varphi(\mathbf{r})$. To each intensity value there corresponds a critical size of inhomogeneity $R^*(\mathbf{r})$ which is related to $q(\mathbf{r})$ by a formula such as Eq. (4). Breakdown inside the volume V will occur if the volume contains at least one absorbing particle whose size exceeds the critical value $R^*(\mathbf{r})$ at the corresponding point. Assuming that the distribution of particles in size does not depend on the coordinates, it is easy to write out an expression for the breakdown probability in the volume V :

$$W[q(\mathbf{r})] = \frac{1 - \exp\left\{-n \int_{(V)} d^3r \int_{n^*(r)}^{\infty} f(\rho) d\rho\right\}}{1 - e^{-nV}}. \quad (9)$$

Here n is the average density of absorbing inhomogeneities, $f(R)$ is the distribution of particles in size, normalized to unity, and $R^*(\mathbf{r})$ is a function related to $\varphi(\mathbf{r})$ by Eq. (4). Usually in the experimental study of

breakdown of dielectrics the spatial profile of the intensity $\varphi(\mathbf{r})$ is kept unchanged and the maximum intensity q_0 is varied. The breakdown probability in this case is a function of q_0 and with the aid of Eq. (9) we can determine the average value, dispersion, and other statistical characteristics of the random value of breakdown threshold.

Let us consider the simplest case of a uniform distribution of light intensity inside the focal volume V . In this case

$$W(q) = \frac{1 - \exp[-N \int_0^{\infty} f(\rho) d\rho]}{1 - e^{-N}}, \quad (10)$$

where $R(q)$ is related to q by Eq. (4) and $N = nV$ is the average number of particles in the focal volume. From Eq. (10) it is easy to see that for a small concentration of absorbing particles ($N \ll 1$) all characteristics of the breakdown turn out to be independent of the conditions of focusing of the radiation, in particular, independent of the size of the focal volume. For $N \ll 1$ it is in fact easy to obtain an expression for the moments of the threshold intensity

$$\langle q^m \rangle = \int_0^{\infty} q^m(R) f(R) dR,$$

which does not contain N . The situation is different in the case in which the average number of absorbing inhomogeneities in the focal volume V is large ($N \gg 1$). In this case the average value of the threshold intensity turns out to be strongly dependent on the volume V and the asymptotic behavior of the inhomogeneity distribution function $f(R)$ at large R . In fact, the probability density is

$$w(q) = \frac{N}{e^N - 1} f(R(q)) \exp\left[N \int_0^{R(q)} f(\rho) d\rho\right] \left| \frac{dR(q)}{dq} \right|$$

and the average value of the threshold intensity is

$$\langle q \rangle = \frac{N}{e^N - 1} \int_0^{\infty} q(R) f(R) \exp\left[N \int_0^R f(x) dx\right] dR. \quad (11)$$

Let us calculate the asymptote of this quantity for large N . For this purpose it is necessary to specify the behavior of $f(R)$ at large R , since for large N , as can easily be seen, the main contribution to the integral (11) is from large R . Physically this statement is obvious; it means that if several particles are present in the focal volume the breakdown threshold is determined by the largest of them, since the latter have the smallest threshold for thermal instability.

We shall assume that as $R \rightarrow \infty$ the distribution function $f(R)$ falls off according to a power law: $f(R) = CR^{-h}$. This assumption, as we shall see below, is in agreement with the experimental data for the dependence of the breakdown threshold on the size of the focal spot. Calculating the integral in Eq. (11), we obtain

$$\langle q \rangle = \frac{\kappa E}{\alpha} B(k) [CN]^{-1/(h-1)}, \quad (12)$$

where

$$B(k) = \sqrt{\frac{2\pi}{k-1}} \left(\frac{k+1}{e^2} \right)^{1/2(k+1)/(k-1)}$$

In derivation of Eq. (12) we assumed for simplicity that the absorption cross section of a particle is $\sigma = 4\pi R^2 \alpha$, where α is a constant, and we neglected the effect of the logarithmic factor in Eq. (4). Note that for an exponential falloff of $f(R)$ as $R \rightarrow \infty$ the average value of the intensity falls off logarithmically with increase of the focal volume. If the distribution function goes identically to zero for $R > R_0$, then the average value $\langle q \rangle$ for $N \rightarrow \infty$ approaches a limit q_m related to R_0 by Eq. (4).

The expressions for the breakdown probability in the form in which they are written above are valid only in the case in which the critical size of the particle $R(q)$ is related to the intensity of light by Eq. (4). Here all particles whose size $R \geq R(q)$ will be critical, i.e., will produce breakdown inside the volume V , since the function $R(q)$ is monotonic. It is evident, however, that the actual calculation scheme for the breakdown probability remains unchanged for any specific form of dependence of the breakdown threshold of an individual particle on its size and on the duration of radiation action $q^*(R, t_0)$. In this case, like the above, particles for which the incident light intensity q exceeds the critical value $q^*(R, t_0)$ will be critical, with the difference that the dependence of q^* on R can be nonmonotonic and the number of critical particles is now defined as

$$N_{cr} = N \sum_{i=0}^{i_{max}} \int_{R_2(q, t_0)}^{R_1(q, t_0)} f(\rho) d\rho,$$

where $R_i(q, t_0)$ is the root of the equation $q^*(R, t_0) = q$. The breakdown probability in this case is determined by the expression

$$W(q, t_0) = \frac{1 - \exp[-N_{cr}(q, t_0)]}{1 - e^{-N}}$$

and goes over to Eq. (10) for $i_{max} = 0$; $R_0(q, t_0) = R(q)$, $R_1(q, t_0) = \infty$.

We shall discuss in somewhat more detail the time characteristics of breakdown. Proceeding from the expression for the induction time obtained by Makshantsev, Kondratenko, and Gandel'man,^[27] it is easy to write out the following formula which determines the nonstationary breakdown threshold of an individual absorbing particle:

$$q^*(R, t_0) = q^*(R) + Q(R, t_0), \quad (13)$$

where

$$q^*(R) \approx E\chi/\alpha R, \quad Q(R, t_0) \approx (\beta R^2/t_0)^{1/2}, \quad (14)$$

$$\beta = \frac{\pi}{27\sqrt{6}} \frac{(E^3 \chi^3 \kappa_0^3)^{1/2}}{\alpha^2 a},$$

κ_0 is the absorption coefficient of the particle, and the remaining designations are as before. For simplicity we assume that $\alpha = \text{const}$. From Eqs. (13) and (14) it

is evident that for fixed length of radiation pulse t_0 there exists a size of particle \bar{R} which lies between the roots R_1 and R_2 of the equation $q^*(R, t_0) = q$, at which the minimum of the function $q^*(R, t_0)$ is achieved. Thus, in pulsed irradiation for each pulse length there are absorbing inhomogeneities of a most "dangerous" size, near which the breakdown threshold of the material is minimal. The location of the minimum of $q^*(R, t_0)$ is determined by the condition

$$\partial q^*(R, t_0) / \partial R = 0$$

and depends on the pulse length as a parameter. Keeping in mind Eq. (14), it is easy to obtain an explicit expression for the dangerous size:

$$\bar{R} = \left[\left(\frac{3}{8} \frac{E\chi}{\alpha} \right)^3 \left(\frac{t_0}{\beta} \right)^2 \right]^{1/4}$$

and the corresponding value of breakdown threshold of the material near the particle of dangerous size:

$$q^*(\bar{R}, t_0) = \frac{11}{8} \left[\left(\frac{8}{3} \right)^3 \left(\frac{E\chi}{\alpha} \right)^3 \left(\frac{\beta}{t_0} \right)^2 \right]^{1/4}. \quad (15)$$

The probability of breakdown of the focal volume V in the case considered is

$$W(q, t_0) = \frac{1 - \exp[-N \int_{\bar{R}}^{\infty} f(x) dx]}{1 - e^{-N}}$$

Obviously for $q \leq q^*(\bar{R}, t_0)$ the function $W(q, t_0) = 0$, so that Eq. (15) can be considered as the definition of the nonstationary breakdown threshold of the dielectric.

Note that in the limit of very long pulses, when the focal volume can be assumed uniformly heated, the analysis of breakdown in most cases can be carried out in the same way as in Ref. 28.

4. DISCUSSION

We shall give a brief summary of the material presented above and illustrate by examples the possibility and legitimacy of use of the proposed model for analysis of optical breakdown processes in transparent dielectrics.

As we have already noted, the breakdown of certain materials actually has the statistical nature predicted by the model. Thus, the experimental data for F-2 glass, which are given in Fig. 3, are quite satisfactorily described by Eq. (19) for a power asymptote of the function $f(R) \sim R^{-k}$ with an exponent $k = 5$ and a value $N = 25$.

In terms of the model proposed the dependence of the breakdown threshold of a dielectric surface on the diameter of the irradiated spot, which has remained a mystery for a long time, finds a natural explanation. To interpret this let us consider a near-surface layer of a dielectric of thickness h containing absorbing inhomogeneities with an average concentration n and a size distribution function CR^{-k} . It is obvious that the irradiated volume of the layer in this case is proportional to the square of the radius of the irradiated spot R_j . Here the average value of the breakdown intensity

$\langle q \rangle$ —the quantity measured experimentally—depends, according to Eq. (12), in power fashion on the radius of the focal spot:

$$\langle q \rangle \sim R_f^{-2/(k-1)} \quad (16)$$

The dependence (16) gives satisfactory agreement with the experimental data for K-8 glass, which are given in Fig. 2, for $k = 2.5$

The model of optical breakdown of a medium with absorbing inhomogeneities describes with qualitative correctness the behavior of the time dependences of the breakdown threshold. The characteristic times at which the form of the dependence changes are the time of heating of the inhomogeneity $\tau_1 = \langle R \rangle^2 / a$ ($\langle R \rangle$ is the average size of the inhomogeneity) and the time of heating of the irradiated volume $\tau_2 = l^2 / a$ (l is the average distance between absorbing inhomogeneities). The experimental results are in agreement with the concepts of existence in the medium of inhomogeneities of sizes 10^{-4} – 10^{-5} cm and an average distance between them of the order of 10^{-3} cm. A detailed comparison of the theoretical and experimental time dependences requires calculation of the thermal-breakdown induction time over a wide range of light intensities and is being carried out at the present time.

In summing up, we can say that the model proposed for optical breakdown of transparent dielectrics permits description of the main experimental relations. It is important to note that all of the facts enumerated apparently can be individually explained more or less satisfactorily without resorting to the assumption of absorbing inclusions. However, taking into account absorption in inhomogeneities gives the possibility of explaining the entire set of experimental facts from a unified point of view, and therefore the model proposed for optical breakdown of transparent dielectrics is extremely plausible.

¹For inclusion sizes $\sim 10^{-5}$ – 10^{-4} cm an analysis based on solution of the heat conduction problem will lead to a qualitatively correct picture of the development of instability.

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