

Dependence of the damage threshold of polymethylmethacrylate on the duration of laser pulses and dimensions of irradiated region

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An investigation was made of the bulk optical strength of polymethylmethacrylate subjected to smooth ruby laser pulses. Characteristic dependences of the critical power density on the diameter of the illuminated region and pulse duration were observed. The experimental curves were explained by analytic expressions obtained by a further development of the concept of thermal instability around absorbing inhomogeneities.

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INTRODUCTION

Studies of the damage of transparent dielectrics (such as glasses) by laser radiation have established that the critical power density is a function of the diameter of the irradiated region ("size effect") and of the duration of the laser pulses. The size and time dependences of the damage of transparent dielectrics are among the most important characteristics of the optical strength of materials in high-power optics. Much work has been done on the size and time effects (see, for example,^[1-4]). The main materials investigated so far have been optical glasses. Studies of polymer materials irradiated with laser pulses also started relatively long ago.^[5,6] The time characteristics of the damage of polymethylmethacrylate (PMMA) by free-oscillation laser pulses are reported in^[7].

However, the size effect has not yet been investigated in the case of PMMA. In the present study, the experiments were carried out in such a way as to determine the size and time (pulse-duration) dependences of the critical power density causing damage, i. e., for each damage event, we determined the coordinates, time of appearance, and critical power density.

Many phenomena which precede and accompany damage in inorganic solid transparent dielectrics can be explained on the assumption that the damage occurs at inhomogeneities. However, the local nature of the damage in inorganic materials is difficult to demonstrate. On the other hand, such local damage is easily observed in PMMA and is clearly due to damage nuclei, i. e., the damage is undoubtedly initiated at absorbing inhomogeneities and microinclusions.^[8,9]

Thus, PMMA is a good model object for experimental investigation of the damage at nuclei and its relationship to size and time dependences.

DESCRIPTION OF EXPERIMENTS

We used apparatus shown schematically in Fig. 1. A quasicontinuously operating ruby laser 1 emitted smooth pulses of $\tau_p \sim 10^{-3}$ duration and the radial distribution of the power density in the beam was uniform.^[10] Beam splitters 2 and 3 deflected some of the light toward an energy meter 4 (IMO-2) and a photocell

5 (F-5). The signal from the photocell 5 was applied to an oscillograph 6 (S1-37). The laser pulses were focused in a transparent sample 8 which was a PMMA block and this was done using a converging lens 7 (focal length in air ~ 42 mm). The samples were cubes of ~ 43 mm edge. A rotatable prism 9 and a streak camera 10 (SFR) were used to record the beginning of a laser pulse. The laser and the camera were synchronized by a unit 11.

The objective of the streak camera made it possible to view a small part of a sample (~ 17 mm) in the focal region of the lens 7. This was done using a tube attachment with a Gelios-44 objective. Damage nuclei resulting from irradiation were recorded with the aid of their own luminescence using a photosensitive film in the SFR camera. Information on the coordinate of a damage nucleus and the time of its appearance relative to the beginning of a laser pulse was contained in photographic streak records. Part of the pulse energy transmitted by a sample at the moment of appearance of a damage nucleus was calculated from information in the form of oscillograms, readings of the energy meter, and streak records.

The influence of the previous history of a sample, i. e., of previous irradiation, on the state of the investigated material was avoided because each PMMA sample was irradiated only once and not used again even when no damage appeared in the irradiation zone.

An investigation of the damage outside the focal region at some distance from it in the direction of the fo-

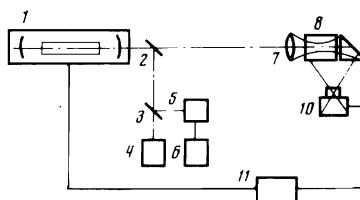


FIG. 1. Schematic diagram of the apparatus: 1) ruby laser; 2), 3) plane-parallel transparent plates (beam splitters); 4) energy meter (calorimeter); 5) photocell; 6) oscillograph; 7) lens; 8) sample; 9) rotatable prism; 10) SFR camera; 11) unit for synchronization and control of SFR camera and laser.

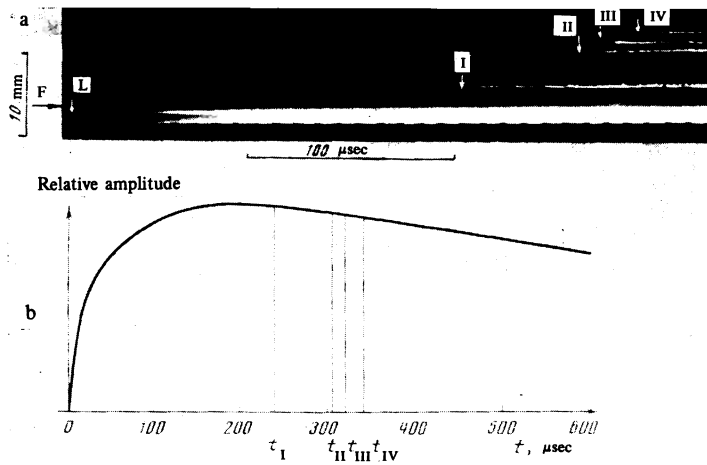


FIG. 2. a) Typical streak pattern of the appearance of micro-damage (L is the beginning laser emission and I-IV are discrete damage nuclei which appear at different points and at different times). b) Oscillogram of a laser pulse showing times of appearance of four damage nuclei.

cusing system made it possible, in principle to eliminate the influence of self-focusing frequently observed in the bulk of a sample made of an inorganic transparent material; in this way, it was possible to solve the problem of the role of self-focusing in the size effect.^[11]

Figure 2a shows streak records of the appearance of damage in the irradiated zone. Away from the focus F, i.e., when the power density decreased, the appearance of damage nuclei was subject to increasing delay. The arrow L denotes the beginning of a laser pulse. Figure 2b is an oscillogram of a laser pulse showing the moments of appearance of damage nuclei I, II, III, IV ($t_I, t_{II}, t_{III}, t_{IV}$).

The fraction of the pulse energy corresponding to the event I, II, III, or IV was estimated from an oscillogram and energy meter (calorimeter) readings, i.e., from the total energy per pulse and the moment of appearance of damage. This information and the geometry of irradiation of PMMA enabled us to calculate easily the critical power density in that section of the light cone where damage took place.

The experimental results were in the form of sets of values of the critical power density q_{ij} , diameter of the irradiated region d_i , and moment of appearance of a damage nucleus t_{ij} . The value of q_{ij} was found to be a random function of the arguments d_i and t_{ij} (here, $i, j = 1, 2, \dots$, where i is the number of the section in which damage was observed and j is the number of a laser shot). These results were analyzed as follows. The investigated ranges of the values of d_i and t_{ij} were divided into intervals $d'_k = d_1 + \Delta d(k-1)$ and $t'_m = t_1 + \Delta t(m-1)$, where d_1 and t_1 are the minimum values of d_i and t_{ij} ; Δd and Δt are the intervals used in the division; $k, m = 1, 2, \dots$. We next calculated the average values of q_{ij} , whose arguments d_i and t_{ij} were inside a square defined by the inequalities

$$d_1 + \Delta d(k-1) < d_i < d_1 + \Delta dk, \quad t_1 + \Delta t(m-1) < t_{ij} < t_1 + \Delta tm,$$

and the parameters d_i and t_{ij} were attributed the values

$$d_k = d_1 + \Delta d(k-1/2), \quad t_m = t_1 + \Delta t(m-1/2).$$

The resultant average values $\langle q_{ij} \rangle = q(d_k, t_m)$ were the

experimental points plotted in Figs. 3 and 4.

THEORETICAL ANALYSIS AND DISCUSSION OF RESULTS

The dependences of the critical power density q_{cr} on the diameter d of a laser spot in the section in which damage is observed (Fig. 3) can be interpreted on the basis of the fluctuation theory,^[4,12,13] which explains the dependence of q_{cr} on d if the damage is observed in the focal region. According to the fluctuation theory, developed for inorganic solid transparent dielectrics, this is due to the fact that an increase in the volume of the focal region $V = \gamma d^4$ ($\gamma = \text{const}$)^[1] results in an increase in the probability that this volume contains a larger absorbing inhomogeneity of radius R ; this corresponds to a lower value of $q(R)$ for which a thermal instability appears in the medium surrounding the inhomogeneity and an avalanche begins to grow.^[16,17] It is assumed that, during the action of laser radiation for a time τ , one center may give rise to avalanche ionization over a considerable part of the focal volume.^[18] Here, the time τ is understood to be the time needed for the appearance of a damage nucleus.

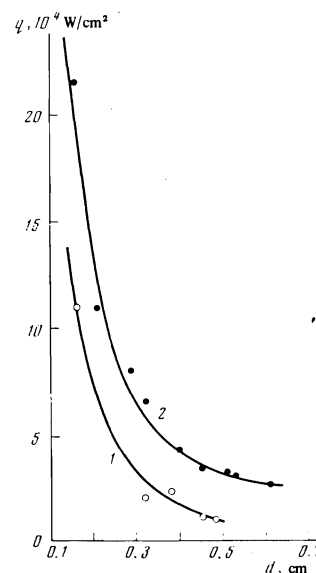


FIG. 3. Dependences of the critical power density q_{cr} on the diameter of the irradiated region d plotted for two different moments τ during a laser pulse: 1) $\tau = 500 \mu\text{sec}$; 2) $\tau = 275 \mu\text{sec}$.

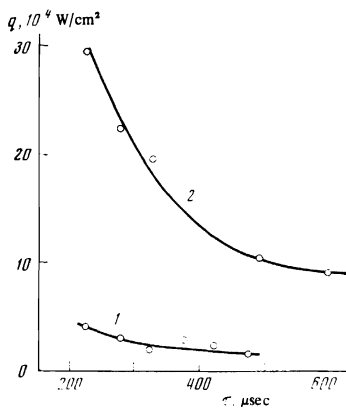


FIG. 4. Dependences of the critical power density q_{cr} on the laser pulse duration τ for two different diameters of the irradiated region: 1) $d = 0.5$ cm; 2) $d = 0.16$ cm.

The above analysis also applies to organic transparent dielectrics such as PMMA. However, in this case, the fairly low values of q_{cr} may give rise to a situation in which avalanche ionization is preceded by avalanche dissociation producing particles such as carbon black, which absorb strongly laser radiation.

The value of q_{cr} is known to be a function not only of the diameter d but also of the pulse duration τ .^[3,4,7] Our experiments confirm this dependence (Fig. 4). The theory developed in^[4,12,13] ignores this point. The dependence of q_{cr} on τ can be allowed for within the framework of the fluctuation theory, as follows.

We shall assume that the condition for optical breakdown is the equality of the induction time $\tau_i(q, R)$, representing the rate of growth of a thermal instability around an absorbing inhomogeneity, and the laser pulse duration τ . Hence, we can find $q = q(R, \tau)$. The average critical power density $q_{cr} = \langle q(R, \tau) \rangle$ can be obtained by averaging over the radii R of inhomogeneities in the focal volume V . This can be done if we know the distribution function $F(R)$ of the radii of absorbing inhomogeneities. We shall assume that, in the region of interest to us, the distribution function can be approximated by a power function of R . Specifically, we shall employ the Junge distribution

$$F(R) = \frac{\beta-1}{2R_m} \left(\frac{R_m}{R} \right)^\beta,$$

where R_m is the median radius.^[19] As in^[12], we find that

$$q_{cr} = \int_0^\infty q(R(x), \tau) e^{-x} dx \quad (1)$$

where $x = (R_m/R)^{\beta-1} VN/2$ is the average number of particles of radius R in the focal volume V ; N is the average density of inhomogeneities in a sample and it is assumed that $VN \gg 1$.^[2]

We shall calculate $q(x, \tau)$ in a model which allows us to consider all possible situations. In this model, we analyze the growth of a thermal instability around a transparent inhomogeneity, which is an aggregate of

impurity centers^[20] whose preliminary heating is due to nonradiative electron transitions. The question of the growth of a thermal instability then reduces to an analysis of an equation for the temperature $T(\mathbf{r}, t)$ of a medium which contains an absorbing inhomogeneity^[17]:

$$\frac{\partial T}{\partial t} = \chi \Delta T + \frac{q\kappa_0}{c} f\left(\frac{r}{R}\right) + q\kappa(T) f\left(\frac{r}{R}\right). \quad (2)$$

Here, χ is the thermal diffusivity and c is the specific heat per unit volume, which are assumed to be the same for an inhomogeneity and the medium around it; $\kappa_0 = n\nu\epsilon/2(q+\bar{q})$, where n is the concentration of impurity centers in an impurity aggregate, ν is the rate constant of nonradiative deactivation of the impurity centers, $\bar{q} = \epsilon\nu/2\sigma$, and σ is the cross section for the absorption of a photon of energy ϵ ; $f(x) = 1$ for $x \leq 1$ and $f(x) = 0$ for $x > 1$;

$$\kappa(T) \approx Ae^{-E/T}, \quad (3)$$

where A and E are constants; A is a quantity of the order of the linear absorption coefficient in the case of complete single ionization of the medium and E is half the ionization potential of the dielectric. However, in the case of organic transparent dielectrics such as PMMA with fairly low values of q , the quantity A is of the order of the linear absorption coefficient corresponding to complete dissociation of chemical bonds which produces carbon black or graphite, and E is the dissociation energy of these bonds; \bar{R} is the characteristic size of the focal volume³⁾ and $V \gg R$.³

Bearing in mind that, in the region of importance to us, we have $E/T \gg 1$ in Eq. (3), we shall seek the solution of Eq. (2) in the form

$$T(r, t) = T_1(r, t) + T_2(r, t),$$

where $T_1(r, t)$ satisfies Eq. (2) if the last term on the right-hand side is omitted. Then, introducing a dimensionless quantity

$$\theta(t) = \frac{E}{T_1^2(r=0, t)} T_2(r=0, t),$$

we obtain the following equation for $\theta(t)$:

$$\left. \begin{aligned} \theta(t) &= \frac{E}{T_1^2(r=0, t)} qA \int_0^t dt' G(t') e^{\theta(t-t')}, \\ G(t) &= \int_0^{\bar{R}} dr' G(r=0, r', t) \exp\{-E/T(r', t)\}, \end{aligned} \right\} \quad (4)$$

where $G(r, r', t)$ is the Green function of Eq. (2), from which all the heat source functions are omitted.

Equation (4), subject to $R^2/4\chi\tau \ll 1$ and $\delta/\delta_{cr} - 1 \ll 1$, where $\delta = 3\kappa[T_1(r=0, t=\infty)]/\kappa_0$, and $\delta_{cr} = 1/e$,^[16] gives the induction time τ_i . Since $\tau_i = \tau$, we obtain the following equation for $q(R)$:

$$\tau \approx \tau_0 \left(\frac{R}{\bar{R}} \right)^4 \frac{q}{q+\bar{q}} \left[\frac{q+\bar{q}}{\bar{q}} \exp\left\{ - \left(\frac{R}{\bar{R}} \right)^2 \frac{q+\bar{q}}{q} \right\} - 1 \right]^{-1}. \quad (5)$$

Here,

$$\tau = \frac{3\pi^2 R^2}{4\sqrt{2} \chi} \quad R^2 = \frac{4\chi cE}{nev} \quad \bar{q} = \frac{nve}{6eA}$$

If $R^2/4\chi\tau \leq 1$ and $\delta/\delta_{cr} \gg 1$, we find from Eq. (4) that

$$\tau \approx \tau_0 \frac{\bar{q}q}{(q+\bar{q})^2} \left(\frac{R}{R}\right)^4 \exp\left\{\left(\frac{R}{R}\right)^2 \frac{q+\bar{q}}{q} \left(1 + \frac{2}{3} \frac{R}{\sqrt{\pi}\chi\tau}\right)\right\}. \quad (6)$$

For $R^2/4\chi\tau \gg 1$, we obtain

$$\tau \approx \tau_0 \frac{2}{3} \frac{\sqrt{2}}{\pi^2} \frac{q+\bar{q}}{q} / \ln \frac{q+\bar{q}}{3e\bar{q}}. \quad (7)$$

We shall now consider in turn several cases when the power density q_{cr} is calculated using the integral (1) to which the chief contribution is made by one of the expressions for q found from Eqs. (5)–(7).

If Eq. (5) is valid, we find $q(R, \tau)$ and, substituting this expression in Eq. (1) on the assumption that $\beta \gg 1$, we obtain

$$q_{cr} \approx \bar{q} \left(\frac{d}{d}\right)^{\beta/(\beta-1)} / \ln \frac{\bar{q}}{q} \quad (8)$$

for $q_{cr} \ll \bar{q}$, $(\bar{\tau}/\tau)(d/\bar{d})^{8/\beta-1} \ll 1$, and

$$q_{cr} \approx 2 \left(\frac{\pi}{\beta}\right)^{1/2} \bar{q} \left(\frac{d}{d}\right)^{4/(\beta-3)} \exp\left[\left(\frac{d}{d}\right)^{\beta/(\beta-3)}\right] \quad (9)$$

for $q_{cr} \gg \bar{q}$ and $(\tau_0/\tau)(d/\bar{d})^{16/(\beta-3)} \ll 1$.

Here,

$$\bar{d} \approx (R/R_m)^{(\beta-1)/4} (2/\gamma N)^{1/4}, \quad \bar{\tau} = \tau_0 / \ln(\bar{q}/\bar{q}), \\ \bar{d} = \bar{d} (2/\beta)^{1/4}.$$

If Eq. (6) is valid, we find that

$$q_{cr} \approx \bar{q} \left(\frac{d}{d}\right)^{\beta/(\beta-1)} \left[1 + \frac{4}{9\pi} \left(\frac{d}{d}\right)^{4/(\beta-1)} \left(\frac{\tau_0}{\tau}\right)^{1/2}\right] / \ln \left[\frac{\bar{q}}{q} \frac{\tau}{\tau_0} \left(\frac{d}{d}\right)^{\beta/(\beta-1)}\right] \quad (10)$$

if

$$q_{cr} \ll \bar{q}, \quad \frac{\bar{\tau}}{\tau} \left(\frac{d}{d}\right)^{\beta/(\beta-1)} \gg 1, \quad \frac{4}{9\pi} \left(\frac{d}{d}\right)^{4/(\beta-1)} \left(\frac{\tau_0}{\tau}\right)^{1/2} \ll 1$$

and

$$q_{cr} \approx 2 \left(\frac{\pi}{\beta}\right)^{1/2} \bar{q} \frac{\tau_0}{\tau} \left(\frac{d}{d}\right)^{12/(\beta-3)} \exp\left\{\left(\frac{d}{d}\right)^{\beta/(\beta-3)} \left[1 + \frac{4}{9\pi} \left(\frac{d}{d}\right)^{4/(\beta-3)} \left(\frac{\tau_0}{\tau}\right)^{1/2}\right]^{\beta/(\beta-3)}\right\}, \quad (11)$$

if

$$q_{cr} \gg \bar{q}, \quad \frac{\tau_0}{\tau} \left(\frac{d}{d}\right)^{16/(\beta-3)} \gg 1, \quad \frac{4}{9\pi} \left(\frac{\tau_0}{\tau}\right)^{1/2} \left(\frac{d}{d}\right)^{4/(\beta-3)} \ll 1.$$

It should be noted that Eqs. (8)–(11) correspond to a situation in which there is on the average one particle in the focal volume around which a thermal instability develops in a time τ .

Finally, for the last of the possibilities considered here, corresponding to the case when the average num-

ber of such particles in the focal volume is much greater than unity, we find from Eq. (7) that, if $q_{cr} \ll \bar{q}$, then

$$q_{cr} \approx \frac{2\sqrt{2}}{3\pi^2} \bar{q} \frac{\tau_0}{\tau} / \ln \frac{\bar{q}}{3e\bar{q}}. \quad (12)$$

In this case, the condition $(\tau_0/\tau)(d/\bar{d})^{8/\beta-1} \gg 1$ should be satisfied.

The formulas obtained above allow us to explain the experimental dependencies in Figs. 3 and 4.

First of all, we note that, as indicated by estimates of the critical power density q_{cr} in our experiments, the inequality $q_{cr} \ll \bar{q}$ is obeyed. Therefore, we shall use Eqs. (8), (10), and (12) in dealing with the experimental results. The expressions (9) and (11) apply only in the case of giant pulses, i. e., when $q_{cr} \gg \bar{q}$.

It is clear from Fig. 3 that curves 1 and 2, representing the dependence of the critical power on d , tend to saturate for large diameters d . This is explained by Eq. (12), which shows that q_{cr} is independent of the diameter provided it is sufficiently large. Moreover, according to Eq. (12), the critical power density and high values of d obey $q_{cr} \propto 1/\tau$. This is in good agreement with the experimental curve 1 in Fig. 4.

In the range of small diameters, curve 1 of Fig. 3 is described by Eq. (10) with $\beta \sim 5-6$. It follows from Eq. (10) that, in this range of diameters, the value of q_{cr} varies weakly with τ . This is in good agreement with curve 2 in Fig. 4, which reaches saturation when τ is sufficiently long. The theoretical curve (1 in Fig. 3), corresponding to smaller diameters, is described by Eq. (8), i. e., by an expression which is independent of τ .

In the range of small diameters and short times, the curves denoted by 2 in Figs. 3 and 4 cannot be described by simple expressions. The experimental values of the parameters are such that Eq. (4) cannot be solved analytically and numerical calculations are required.

On further increase of d , curve 2 of Fig. 3 should theoretically reach the region where it is described successively by Eqs. (10) and (8). Thus, the theory predicts an approach of curves 1 and 2 in Fig. 3 when the diameter d is reduced and this is indeed observed experimentally. According to the theory, curve 2 of Fig. 4 should change to a $1/\tau$ dependence on further reduction of τ .

Thus, the experimental results indicate that PMMA behaves in the same way as inorganic solid transparent dielectrics and that, in addition to the time dependence of the critical power density, it exhibits also a dependence of the diameter of the irradiated region. The results obtained are explained satisfactorily by the fluctuation theory of the size effect, based on the assumption that a thermal instability appears around an absorbing inhomogeneity during the initial stage of the damage process. It follows from the experimental results that the simultaneous determination of the position and time of appearance of a damage nucleus is a necessary condition for the correct interpretation of the optical damage of materials.

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¹We shall confine our attention to the specific case of q_{cr} in the focal volume $V \propto d^4$.^{114,151} Since the volume V is simply the region in which, in a given section, the power density can be assumed to be approximately constant, the formulas obtained can easily be generalized to the case when damage occurs outside the focal volume. We then have to consider the volume $V \propto d^3$.

²It should be noted that, since $q(R, \tau) \rightarrow \infty$ in the limit $R \rightarrow 0$,¹¹⁷ a possible divergence of the integral in Eq. (1) limits the validity of this equation to certain distribution functions $F(R)$. A suitable analysis shows that it is more correct to average over R the quantity $1/\tau_i(R)$ and to find q_{cr} from $1/\tau = \langle 1/\tau_i(R) \rangle$. This approach avoids divergences. In our case, we are assuming that $F(R) = 0$ in the range $R \leq R_m$ so that the results obtained by either of these methods are practically identical. However, the averaging of $q(R, \tau)$ is preferred because of its simplicity.

³Equation (2) is similar to the equations solved in the theory of combustion when the exponential temperature dependence predominates and all other temperature dependences can be ignored compared with it. On this basis, Eq. (2) is derived ignoring the temperature dependence of the thermal diffusivity.¹²¹

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