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Efficiency of various mechanisms of the laser damage in transparent solids

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Abstract. **Criteria for the laser damage in transparent solids are compared for different mechanisms of radiation absorption. It is shown that the most efficient mechanism of laser damage in a broad range of pulse durations (from millisecond to femtosecond laser pulses) is the photoionisation thermal explosion of absorbing inclusions. It is found that upon irradiation by long laser pulses, the damage proceeds through a melting stage. A fundamental feature of the damage is that, to produce a crack in a transparent solid, the size of an absorbing region should exceed a critical size.**

Keywords: *laser damage, damage criterion, mechanical damage, crack formation, absorbing inclusion, ablation.*

1. Introduction

The damage of transparent solids can be caused by various mechanisms of absorption of laser radiation energy. Among them are the so-called intrinsic mechanisms related to the properties of a matrix itself (impact and multiphoton ionisation) and mechanisms caused by absorbing inclusions (thermoelastic, thermal explosion, and photoionisation mechanisms) [1]. Realisation of one or another of the damage mechanisms depends on many factors such as the sample purity, the radiation wavelength, the laser pulse duration, etc.

Upon irradiation of solids by nanosecond or picosecond laser pulses, the most probable damage mechanism is thermal explosion caused by absorbing inclusions [2]. The damage morphology substantially changes upon irradiation by femtosecond pulses [3 – 5]: instead of the crack formation, ablation occurs. Such a change in the damage type suggests that the damage of solids irradiated by subpicosecond pulses is caused by another absorption mechanism of laser radiation. The authors [3 – 5] assumed that the damage induced by nanosecond – picosecond pulses is due to absorbing inclusions, whereas the damage produced by subpicosecond pulses is caused by intrinsic mechanisms.

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This point of view has not been consistently substantiated. It was based exclusively on heuristic reasoning. In particular, the dependence of the damage threshold on the laser pulse duration has not been conclusively explained. At the same time, a consistent analysis of this experimental dependence based on the thermal explosion mechanism showed that this dependence is well described by this mechanism in all pulse width range investigated, including femtosecond pulses [2, 6].

To reveal the conditions for realisation of a laser damage mechanism and, hence, its domination under certain conditions, it is necessary to formulate and compare criteria for different damage mechanisms. This question is important both for finding the dominant damage mechanism and for a practical estimate of the ultimate laser damage resistance of transparent solids under various irradiation conditions.

The aim of this paper is to compare the efficiencies of different laser damage mechanisms and to find out the dominating mechanism using consistently formulated criteria for their realisation.

2. Laser damage criteria for different mechanisms

The existing laser damage criteria can be divided into two classes: criteria based on the existence of a critical electron concentration in the conduction band of a transparent solid and criteria based on the existence of the threshold temperature of a crystal lattice.

In the first case, it was assumed that the damage occurs when the electron concentration in the conduction band of a transparent solid achieves the critical concentration n_{cr}, which is sufficient for 'strong' absorption of laser radiation. It follows from this definition that the choice of n_{cr} is quite arbitrary in this case. In one of the earlier papers [7] devoted to impact ionisation, it was assumed that $n_{cr} = 10^{18}$ cm^{-3}, whereas the authors of recent paper [3] set n_{cr} equal to the plasma concentration $n_{cr} \sim 10^{21}$ cm^{-3}.

In the second case, it was assumed that the damage occurs when the lattice temperature of a solid achieves the threshold temperature T_{th} in the interaction region. The value of T_{th} is determined by the damage mechanism. For example, it was assumed within the framework of the thermal model proposed in [8] that the damage occurs if the temperature of an inclusion absorbing radiation achieves the melting temperature of a transparent solid. The criterion of 'fourteen generations' for the impact ionisation mechanism [9] was based on a similar approach (the attainment of the melting temperature).

The authors [10] assumed that the damage occurs when thermoelastic stresses in the vicinity of a heated inclusion in a solid achieve the ultimate strength of the material. The order of magnitude estimate of T_{th} obtained in [10] is 10^4 K.

The disadvantage of papers [8–10] is the neglect of the temperature dependence of the parameters of materials. Such an approximation is not valid in a broad temperature range up to the melting temperature, all the more up to temperature 10^4 K. The consideration of this dependence in paper [11] showed that an increase of absorption in the material with increasing temperature results in a sharp increase in the temperature of an inclusion upon laser heating. It is important that within the framework of this approach, the temperature T_{th} is not arbitrary but is determined by the thermal properties of materials of inclusions and a transparent solid.

3. Criteria for the appearance of a mechanical damage

3.1 Criteria for the crack formation

The above criteria for the critical concentration and threshold temperature are indirect. They ascertain a substantial increase in absorption in the interaction region if the radiation intensity in this region exceeds the threshold intensity I_{th} rather than the appearance of a damage. However, to solve the problem of laser damage correctly, it is necessary to formulate criteria for the final damage stage of a transparent solid. This final stage can be melting, crack formation or ablation. Numerous experimental studies have shown that upon irradiation of transparent solids by laser pulses of duration from milliseconds to picoseconds, the crack formation occurs at the final stage. Deviations from this damage mechanism were observed (almost for all transparent solids) upon irradiation by ultrashort pulses (shorter than 10 ps) [3–5], when the damage was mainly caused by ablation. Upon irradiation of some materials by longer (millisecond) laser pulses, a melting was observed [12].

The criterion for formation of a mechanical damage caused by local laser heating was consistently formulated in paper [13], where it was shown that the crack formation is independent of the mechanism of absorption of laser radiation. This process is the same for intrinsic absorption mechanisms (impact and multiphoton ionisation) and mechanisms involving absorbing inclusions. It was found in [13] that the crack formation can be described by the break-off mechanism, and the condition necessary for its formation has the form

$$\max \sigma_\phi(r,t) \geqslant \sigma_{th}, \tag{1}$$

where $\sigma_\phi(r,t)$ is the tangential component of the stress tensor and σ_{th} is the ultimate strength of a transparent solid.

However, the fulfilment of inequality (1) is not sufficient for the crack formation. As shown in [13], the produced crack always has a finite size, so that a finite energy is required for its formation. In the case of a spherical interaction region with the radius R, this energy is $E_m = 39R^2\gamma$ (where γ is the surface energy density of a transparent solid). This means that the crack will be formed if only the strain-field energy in the vicinity of local heating exceeds E_m:

$$\eta\vartheta E_p \geqslant E_m, \tag{2}$$

where E_p is the laser pulse energy; η is a parameter determining a fraction of the absorbed energy; $\vartheta = (T_0/9c_h) \times [(1+\nu_h)/(1-\nu_h)]^2\alpha_h^2 c_l^2$ is the connectivity coefficient; c_h is the heat capacity; α_h is the linear expansion coefficient; ν_h is the Poisson ratio; c_l is the longitudinal sound speed; and T_0 is the temperature before irradiation by a laser pulse. Hereafter, the subscripts h and i denote the material parameters of a transparent solid and an inclusion, respectively.

Therefore, to produce a mechanical damage in a transparent solid upon local laser heating, both the local strength criterion (1) and energy criterion (2) should be fulfilled.

The energy criterion (2) can be written in other equivalent forms, which are of interest from a physical point of view. Taking into account the relation between the pulse energy and the radiation power F_p in the interaction region [$E_p = (\sqrt{\pi}/2)F_p\tau_p$, where τ_p is the pulse duration], we transform expression (2) to the form

$$\tau_p \geqslant \tau_{cr} \simeq \frac{78R^2\gamma}{\eta\vartheta F_p}. \tag{3}$$

Inequality (3) means that cracks cannot be produced upon irradiation of solids by short pulses ($\tau_p < \tau_{cr}$) with the threshold intensity I_{th}.

The numerical estimate of τ_{cr} for fused silica in experiments [3] gives the value 50 ps [13], in good agreement with experimental data which show that the transition from the crack formation to ablation occurs at $\tau_p = 20$ ps.

Another limitation, which follows from inequality (2), is the requirement of a finite size of the local heating region for the damage to occur. Indeed, the quantity η determines a fraction of the absorbed pulse energy. For a spherical interaction region, we obtain $\eta E_p \simeq (4/3)\pi R^3 k_a W_p$ (where W_p is the pulse energy density and k_a is the absorption coefficient). Then, inequality (2) takes the form

$$R \geqslant R_{cr} \simeq \frac{10\gamma}{\vartheta k_a W_p}. \tag{4}$$

Expression (4) has a clear physical meaning, which is as follows. The absorbed laser radiation energy is proportional to the volume of the interaction region, i.e., to R^3, whereas the crack energy is proportional to R^2. As R decreases, the energy required for the crack formation decreases slower than the absorbed energy, so that the energy absorbed in a quite small interaction region will be insufficient for the crack formation. It is important that, as follows from numerical estimates, inequality (1) is also satisfied for a small interaction region, whereas condition (4) [or equivalent condition (2)] is not valid in this case. Therefore, criteria (1) and (2) are independent, and both these criteria should be satisfied simultaneously to produce a crack.

Let us estimate the value of R_{cr}. By using the material constants for fused silica from [14], we obtain $\vartheta \simeq 5 \times 10^{-5}$ and $\gamma \simeq 2.4 \times 10^{-5}$ J cm^{-2}. Under the experimental conditions [3], the energy density $W = 5$ J cm^{-2}, so that expression (4) yields $R_{cr} \simeq 0.1 - 0.7$ μm for $k_a \simeq 10^5 - 10^4$ cm^{-1}.

Therefore, it seems that the thermoelastic damage of fused silica by submicron inclusions assumed in [10] cannot occur. The energy absorbed in the region of these inclusions is insufficient to cause the damage.

3.2 Restrictions caused by the stress-growth dynamics

The strength criterion (1) is inconvenient for analysis of the laser damage because it is represented in terms of the stress. Because the stress dynamics in a local heating region is different for the intrinsic and extrinsic mechanisms of radiation absorption, we will study these mechanisms separately.

3.2.1 Intrinsic mechanism of laser radiation absorption

As shown in [13], upon the local laser heating of a solid, the tangential component of the stress tensor becomes maximal to the instant of time $t = 1.15\tau$ ($\tau = c_h\rho_h R^2/\chi_h$, where ρ_h and χ_h are the density and heat conduction, respectively) after the beginning of absorption of laser radiation, and then decreases despite increasing temperature. Such a change in the stress is caused by its proportionality to the temperature gradient. A sharp increase in the temperature at the initial stage of radiation absorption causes an increase in the stress; however, later the diffusion of heat to a transparent solid reduces the temperature gradient and, hence, pressure is also reduced.

Taking into account this time dependence of the stress, criterion (1) is equivalent to the fulfilment of two inequalities [13]

$$\theta \geqslant \theta_{cr} = \varepsilon_{th}/\alpha_h, \tag{5}$$

$$\frac{d\theta}{dt} \geqslant 0.9\frac{\theta_{cr}}{\tau}, \tag{6}$$

where $\theta = (T - T_0)$; θ_{cr} is the critical temperature of the damage formation; and ε_{th} is the ultimate strain. In the elastic approximation,

$$\varepsilon_{th} = \frac{3\sigma_{th}(1 - \nu_h)}{E_h}, \tag{7}$$

where E_h is the Young modulus. By assuming $\sigma_{th} = 0.1E_h$, we obtain, according to the theoretical limit, the critical temperature for fused silica $\theta_{cr} > 10^4$ K. It seems that such high temperatures cannot be achieved. Such a high value of θ_{cr} is possibly explained by the use of the theoretical ultimate strength.

In the case of linear absorption, we have $d\theta/dt \sim \theta/\tau_p$, so that it follows from inequality (6) that

$$\tau_p < \tau. \tag{8}$$

Inequality (8) means that upon irradiation by long pulses (for the interaction region of size of several micrometers in silica, τ is a few tenth of nanoseconds), the melting will occur instead of the crack formation. This conclusion is also applied in the case of nonlinear absorption, which is characterised by a low threshold intensity. Indeed, by neglecting the heat diffusion from the interaction region, we write the relation $d\theta/dt \simeq k_a I_{th}/(c_h\rho_h)$, which, taking into account (6), yields the condition

$$I_{th} \geqslant I_{cr} \simeq \frac{c_h\rho_h\theta_{cr}}{k_a\tau} \tag{9}$$

for the crack formation. When I_{th} is lower than I_{cr}, a crack is not produced even if $\theta > \theta_{cr}$ because the heat diffusion from the heated region reduces stresses in the region vicinity. The temperature in the interaction region will be higher than the melting temperature of the material, so that the damage will develop through the melting stage. Upon melting, the volume of a transparent solid drastically increases (for a typical case of transition to a liquid state, $\Delta V/V \sim 0.01 - 0.03$, where V is the melt volume and ΔV is the volume change upon melting), and the stress in the melt region sharply increases due to the phase transition. Such a jump of the stress also can produce cracks; however, this problem requires a special treatment.

3.2.2 Damage caused by inclusions

Upon heating of an absorbing inclusion, whose elastic parameters strongly differ from those of a transparent solid, the dynamics of stress variation will also differ from the dynamics considered above. Once the stress has achieved its maximum value, it will decrease only slightly, and the ultimate strain in the elastic approximation will be [15]

$$\varepsilon_{th} = \frac{3\sigma_{th}(1 - \nu_i)}{E_i(1 - \xi)}. \tag{10}$$

The parameter ξ tends to zero with decreasing the pulse duration, and $\xi = \xi_{ih} = \alpha_h E_h(1 - \nu_i)/[\alpha_i E_i(1 - \nu_h)]$ for long pulses. This means that, when the damage is caused by absorbing inclusions for which $\xi_{ih} \ll 1$, inequality (6) is not valid. Then, the damage criterion has the form (5), where α_h should be replaced by α_i, and ε_{th} is defined by expression (10).

The requirement $\xi_{ih} \ll 1$ is valid for Pt, Pb, Fe, Sb, etc. metal inclusions in fused silica. For these inclusions, ξ_{ih} lies between 0.01 and 0.04. For non-metal, as well as some metal inclusions, ξ_{ih} is much greater (for example, $\xi_{ih} = 2.3$ for Ce in fused silica), so that the damage criteria, as in the case of the intrinsic absorption mechanism of laser radiation, have the form (5) and (6). In these expressions, however, α_h should be replaced by α_i, and the ultimate strain has the form (10), where one should set $\xi = 0$.

The estimate of the critical temperature of the silica damaged by a metal inclusion yields typically a value of no more than 6×10^3 K. Such temperatures of inclusions can be probably attained at the laser radiation intensity close to the threshold. However, as follows from criterion (4) and estimates of the critical size of the interaction region, the damage will occur only in materials containing inclusions of a sufficiently large (micrometer) size.

4. Thermal explosion of an absorbing inclusion

4.1 Thermal instability and thermal explosion

The thermal instability of materials is caused by the dependence of their parameters (first of all, the absorption coefficient k_a) on temperature. Because $k_a(T)$ of many materials increases with temperature, absorption increases nonlinearly, leading in turn to the temperature increase, etc. As a result, when the temperature of inclusions exceeds the threshold temperature T_{th}, thermal instability appears due to a positive feedback. The threshold temperature of thermal instability can be determined from equation (9) of paper [16] assuming that the heat conduction coefficient

is independent of temperature. In this case, this equation has the form

$$(T - T_0)\frac{\mathrm{d}\sigma_a(T)}{\mathrm{d}T} = \sigma_a(T), \tag{11}$$

where $\sigma_a(T)$ is the absorption cross section, which is proportional to $k_a(T)$ for small inclusions. The threshold intensity of the thermal-instability initiation is [16]

$$I_{th} = 4\pi R\chi_h \left[\frac{\mathrm{d}\sigma_a(T)}{\mathrm{d}T}\right]^{-1}_{T=T_{th}}. \tag{12}$$

Thermal instability will dominate over a direct mechanical damage when $\theta_{th} \leqslant \theta_{cr}$ and inequality (6) is satisfied. By comparing expressions (5) and (11) and taking into account that $\sigma_a(T) \sim k_a(T)$, we transform the inequality $\theta_{th} \leqslant \theta_{cr}$ to the form

$$\frac{\mathrm{d}k_a(T)}{\mathrm{d}T} \geqslant \frac{\alpha_i k_a(T)}{\varepsilon_{th}}. \tag{13}$$

For a strongly absorbing inclusion, $k_a(T) = 10^5$ cm^{-1}, and inequality (13) yields $\mathrm{d}k_a(T)/\mathrm{dT} \geqslant 10$ cm^{-1} K^{-1}. The latter inequality is satisfied for some metals [17]. In the case of a weakly absorbing inclusion, $k_a(T) = 10^2$ cm^{-1}, we obtain from (13) $\mathrm{d}k_a(T)/\mathrm{dT} \geqslant 10^{-2}$ cm^{-1} K^{-1}, which is also satisfied for many metals.

It is important that even a weak increase in absorption in the region of an inclusion is sufficient for producing thermal instability. Indeed, assuming that $k_a(T) = k_0(T/T_0)^{\delta}$ and taking into account that $\sigma_a(T) \sim k_a(T)$, we obtain from (11)

$$T_{th} = \frac{\delta}{\delta - 1} T_0 . \tag{14}$$

According to (14), thermal instability will appear if $k_a(T)$ increases with temperature faster than linearly.

The above estimates show that thermal instability appears when the absorption coefficient only slightly increases with temperature and inequality (13) is satisfied. Therefore, the thermal instability mechanism will dominate in the damage process when the rate of the temperature increase satisfies inequality (6) and, in addition, a sufficient amount of energy required for the damage development is absorbed in the region of inclusions [see inequality (2)].

It is assumed that thermal instability is accompanied by a drastic increase of temperature in the absorption region; however, saturation processes can slow down this increase and suppress thermal instability. The effect of absorption saturation processes on the development of thermal instability was studied in paper [18] using the Arrhenius model, in which the absorption coefficient has the form

$$k_a(T) = k_0 + k_1 \exp(-A/T), \tag{15}$$

where k_0 and k_1 are constants and A is the activation energy. For inclusions having $k_a(T)$ defined by expression (15), an increase in the temperature during the development of thermal instability is limited.

Analysis of the solution of equation (11) and the temperature growth kinetics performed in [18] showed that thermal explosion, i.e., a drastic increase in the temperature of inclusions up to θ_{cr} occurs when

$$k_0 \ll k_1, \tag{16}$$

$$A \gg T_0. \tag{17}$$

Therefore, in the case of weakly absorbing inclusions, whose size satisfies inequality (4) (i.e., of the order of one micrometer and larger), thermal explosion is a dominating damage mechanism of transparent solids.

4.2 Photoionisation thermal explosion

In the case of inclusions of a submicron size, the energy absorbed in the region of inclusions is insufficient for producing a damage. However, inclusions will initiate an absorption wave propagating into a transparent solid. The mechanism of the development of the absorption wave – photoionisation of a transparent solid by thermal radiation of the heated inclusion, was proposed in paper [19] and was analysed in detail in papers [16, 20, 21]. Here, we compare the efficiency of this mechanism with that of a direct mechanical damage.

Within the framework of the photoionisation thermal explosion model, the absorption cross section $\sigma_a(T)$ of an inclusion, taking into account the contribution from photoelectrons, has the form

$$\sigma_a(T) = \sigma_i + \sigma_h(T), \tag{18}$$

where $\sigma_i = (4/3)\pi R^3 k_i$ is the initial absorption of the inclusion and $\sigma_h(T)$ is the absorption cross section related to photoelectrons excited by radiation from the heated inclusion. According to [20], $\sigma_h(T)$ has the form

$$\sigma_h(T) = \frac{\sigma_{h0}}{2}\left(\frac{T}{E_g}\right)^3 \int_{(E_g/T)}^{\infty} \frac{s^2 \mathrm{d}s}{\mathrm{e}^s - 1}, \tag{19}$$

where $\sigma_{h0} = 16R^2\omega E_g^3 \tau_e c^{-3} N_h \mathrm{Im}[\beta_h(\omega)]$; ω is the laser radiation frequency; E_g and N_h are the energy gap and the refractive index of the transparent solid; c is the speed of light; τ_e and $\beta_h(\omega)$ are the recombination time and polarisability of electrons in the conduction band, respectively. Hereafter, we analyse photoionisation thermal explosion using the system of units in which the temperature and energy have the same dimensionality. It was shown in paper [16] that $\sigma_a(T)$ defined by expression (18) can provide the appearance and development of thermal explosion. It is important that, unlike the Arrhenius model, the photoionisation model does not exhibit the absorption saturation. This is explained by an increase in the geometric size of the absorption region caused by propagation of a photoionisation absorption wave.

In the limit $T \ll E_g$, we obtain from (19)

$$\sigma_h(T) \simeq \sigma_{h0}(T/E_g)\exp(-E_g/T). \tag{20}$$

In the case of the photoionisation mechanism of thermal explosion, inequalities (16) and (17) have the form

$$\sigma_{h0}/\sigma_i \gg 1, \tag{21}$$

$$E_g/T_{ph} \gg 1, \tag{22}$$

where T_{ph} is the threshold temperature of the photoionisation instability.

The ratio σ_{h0}/σ_i can vary in a broad range. These variations are mainly caused by the uncertainty in the values of R and τ_e. In particular, τ_e ranges from 10^{-7} to 10^{-12} s, increasing with the material purity (see, for example, [22]). By using expressions for σ_{h0} and σ_i and separating their dependence on R and τ_e, which is most interesting from the physical point of view, we obtain for a transparent solid

$$\frac{\sigma_{h0}}{\sigma_i} \simeq 0.2\frac{\tau_e}{R}. \tag{23}$$

Because the numerical coefficient in expression (23) has the dimensionality cm s^{-1}, the radius R of the inclusion should be expressed in centimetres and the relaxation time τ_e in seconds.

Pure transparent solids have usually large τ_e. Assuming for the estimate that $\tau_e = 10^{-7}$ s and $R = 10^{-5}$ cm, we obtain from (23) that $\sigma_{h0}/\sigma_i \sim 20$, and for inclusions with $R = 10^{-6}$ cm, we have $\sigma_{h0}/\sigma_i \sim 200$. Therefore, inequality (21) is satisfied for pure materials, being better satisfied for smaller inclusions.

The threshold temperature of thermal instability is the solution of equation (11). In the case of the photoionisation mechanism, the absorption cross section is defined by expressions (18) and (20), so that expression (11) takes the form

$$\frac{\sigma_{h0}}{\sigma_i}\exp\left(-\frac{E_g}{T}\right)\left(1-\frac{T_0}{T}-\frac{T}{E_g}\right) = 1. \tag{24}$$

Taking inequalities (21) and (22) into account and assuming that $T_{ph} \gg T_0$, we obtain the estimate of T_{ph}:

$$T_{ph} \simeq E_g\left(\ln\frac{\sigma_{h0}}{\sigma_i}\right)^{-1}. \tag{25}$$

Expression (25) confirms the validity of inequalities $T_{ph} \gg T_0$ and (22) used in calculations. Moreover, it shows that inequalities (21) and (22) are not independent: if inequality (21) is satisfied, inequality (22) is also satisfied. By substituting expression (25) to (22), we find

$$\sigma_h(T_{ph}) = \frac{\sigma_i}{\ln(\sigma_{h0}/\sigma_i)} < \sigma_i. \tag{26}$$

A mechanical damage is caused by thermal instability if the parameters of an inclusion and a transparent solid satisfy criterion (13), which, taking into account (25), takes the form

$$\ln\left(\frac{\sigma_{h0}}{\sigma_i}\right) \geqslant E_g\frac{\alpha_i}{\varepsilon_{th}}. \tag{27}$$

The value of the right-hand side of inequality (27) is of about 0.5–2, and, since $\sigma_{h0} \gg \sigma_i$ [see (23)], inequality (27) is always satisfied for small inclusions. Therefore, the photoionisation thermal explosion precedes a mechanical damage.

5. Conclusions

A comparative analysis of criteria for the laser damage of transparent solids performed in this paper for different mechanisms of absorption of radiation revealed the following properties of the damage caused by local laser heating.

Upon irradiation of solids by long laser pulses (when the damage threshold is low), the melting should be observed. The irradiation by ultrashort pulses should cause ablation, while the irradiation by laser pulses with intermediate durations should produce cracks caused by the thermoelastic mechanism. The critical parameters for the crack formation are the laser pulse duration, the size of the irradiated region, and the energy absorbed. A dominating mechanism of the laser damage caused by inclusions is the photoionisation thermal explosion.

The range of laser-pulse durations in which one or another type of laser damage is observed is determined by the thermal and optical parameters of materials of solids and inclusions. For typical optical materials (fused silica, glasses, etc.), the melting is observed upon irradiation of solids by millisecond laser pulses, cracks are produced upon irradiation by nanosecond-picosecond pulses, and the irradiation by femtosecond pulses causes ablation.

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