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Problems of the physics of high-power ultrashort laser pulse interaction with transparent solids

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***Abstract*. The contribution of A.M. Prokhorov to quantum electronics is briefly reviewed. The recent experimental data in the field of laser damage (LD) of transparent solids by ultrashort pulses are analysed. The dependence of the LD threshold on the pulse duration and the damage morphology are discussed. The experimental data are interpreted within the framework of the theoretical concepts of the LD mechanisms. In particular, the criteria for a change in the damage morphology from cracks to ablation are formulated at transition of an irradiation regime from long pulses to ultrashort ones.**

Keywords: *laser damage, transparent solids, ultrashort pulses, crack formation, ablation.*

1. Introduction

This paper is based on the materials of the invited report at the Memorial session devoted to A.M. Prokhorov and N.G. Basov of the International Conference on Quantum Electronics (A.A. Manenkov, 'Physics of High-Power – Transparent Solids Interaction in Ultrashort Time Domain', IQEC'2002, 22 – 28 June, 2002, Moscow, Conference Program, p. 4). In addition to the subject corresponding to the title of this report, it also includes a brief review of certain works by A.M. Prokhorov in the field of quantum electronics and laser physics.

A significant interest in the physics of the interaction of high-power laser radiation with solids in a time domain of ultrashort pulses ($10^{-12} - 10^{-15}$ s) is initiated by the recent achievements in the development of high-power femtosecond lasers and the potentialities of their application in various fields. Therefore, the problem of revealing the interaction mechanisms determining the damage of solids in this range of pulse durations becomes extremely important. The knowledge of such mechanisms is important for the development of ultrahigh-power laser systems, since their maximum power is limited by a damage of optical elements, and for various studies in the areas of laser physics and technological applications.

The recent investigations of laser damage (LD) of transparent solids exposed to ultrashort pulses [1 – 4] revealed substantial distinguishing features of the LD characteristics compared to those observed for longer (nanosecond and picosecond) laser pulses. In this paper, these results are analysed on the basis of contemporary theoretical concepts of the LD mechanisms; topical unsolved problems are also discussed.

2. The contribution of A.M. Prokhorov to quantum electronics and laser physics

Let us briefly review the contribution of A.M. Prokhorov to quantum electronics and laser physics. It should be mentioned that, in view of a limited size of this paper and inevitably subjective selection of works, this review is far from being complete. We tried to present only the basic (in our opinion) works by A.M. Prokhorov in these fields.

The following characteristics of A.M. Prokhorov as a scientist should be emphasised:

An extremely wide scope of scientific interests and an outstanding erudition.

A combination of fundamental and applied investigations in his scientific activity. It can even be said that his attitude to the division of sciences into basic and applied ones was critical, and he emphasised their tight relation. His activity had a great effect on the formation of many scientific trends and putting of the results of research into practice.

Very wide contacts with scientists, engineers, and managers in the scientific and technological fields of activity in Russia and many countries all over the world. A great number of researchers of all ranks, from young to leading scientists, working in basic and applied sciences were eager to discuss research and practical problems with him. These discussions were always very fruitful and stimulating.

A.M. Prokhorov published many papers on numerous problems of physics, electronics, and other fields. Let us mention only some of his works in quantum electronics and laser physics.

The pioneering works that formed the basis of quantum electronics:

The proposal of a new principle of amplification and generation of electromagnetic radiation, which is based on a stimulated emission upon quantum transitions in atoms and

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called the maser – laser principle (1953 – 1954, together with N.G. Basov [5]).

The proposal of a new method for producing an inverted population of quantum states, the so-called three-level method based on the application of external (auxiliary) electromagnetic radiation (1955, together with N.G. Basov [6]). This method usually named the electromagnetic pumping turned out to be a universal and the most efficient technique for inverting the populations in various atomic systems, especially in solids and liquids and allows the creation of quantum oscillators and amplifiers over a very wide spectral range of electromagnetic waves (from microwaves to UV radiation).

The proposal of a new type of electromagnetic resonators, the so-called open resonator (1958 [7]). This type of the resonator formed by two plane – parallel mirrors became the basic prototype for other open resonators of various configurations, which are currently widely used in lasers.

The first investigation of the microwave spectrum of ruby crystals and the proposal of using them as an active material for masers (1955 – 1956, jointly with A.A. Manenkov [8, 9]).

The first demonstration of the maser effect in ruby at microwaves in the decimetre and centimetre ranges (1958, jointly with G.M. Zverev, L.S Kornienko, A.A. Manenkov [10, 11]).

Subsequent works on masers and lasers:

The development of practical layouts and designs of ruby masers in a broad wavelength range (from decimetres to millimetres) and their application in radio astronomy and remote space communications (1958 – 1965, with G.M. Zverev, N.V. Karlov, L.S Kornienko, A.A. Manenkov, V.B. Shteinshleiger, et al. [12]).

The proposal and implementation of a new type of the laser, the so-called gas-dynamic laser (1966, with V.K. Konyukhov [13]).

The development of new materials (glasses and crystals activated with rare-earth and group irons) for solid-state lasers of IR, visible, and UV wavelength ranges (1970 – 1986, with E.M. Dianov, V.V. Osiko, I.A. Shcherbakov, et al. [14 – 16]).

Detailed studies of physical processes in solid-state lasers that resulted in discoveries of many important effects (a compensation for thermal distortions in multicomponent laser glasses [14], the role of cross relaxation in multilevel laser media [17], etc.).

Investigations in laser physics and nonlinear and fibre optics:

The proposal, substantiation, and confirmation of adequate models of self-focusing of laser beams in nonlinear media: models of a multifocal structure and moving foci (1967 – 1970, with V.N. Lugovoi (theory), V.V. Korobkin, A.A. Manenkov, et al. (experiment) [18 – 21]).

The development of the theory of long-distance interaction of solitons in optical fibres (1992, with E.M. Dianov et al. [22]).

The proposal of new efficient materials for Raman fibre lasers (2000, with E.M. Dianov [23]).

Comprehensive studies of laser breakdown in gases, a so-called laser spark (1964, with P.P. Pashinin et al. [24]).

Detailed studies of fundamental mechanisms of laser damage (LD) in transparent solids (1970 – 1980, with Yu.K.Danileiko, A.S. Epifanov, A.A. Manenkov, et al. [25]). A.M. Prokhorov paid much attention to this field of investigations, in particular, to the problems of laser damage under ultrashort pulses.

Before analysing these problems that are considered in the subsequent sections, we briefly review the results of studies of LD mechanisms related to ‘long’ pulse (nanosecond) range.

3. History of optical breakdown in transparent solids

The phenomenon of optical breakdown (laser damage) in transparent solids was discovered soon after giant-pulse lasers were devised in 1964 by two groups of scientists (C. Guiliano [26] and G. Cullom, R. Waynant [27]). Studies of this effect aimed at the elucidation of its nature and the development of highly radiation-resistant optical materials and components of high-power lasers were performed since that time. A great number of papers on this problem have been published, including detailed reviews (e.g., a review by A.A. Manenkov and A.M. Prokhorov [25]). The main results of these investigations can be summarised as follows:

(i) It was established that absorbing inclusions play a dramatic role in the optical breakdown by initiating the LD and significantly reducing its threshold;

(ii) An intrinsic damage whose threshold is maximum for a given type of material is observed only in extremely pure inclusion-free materials;

(iii) The basic LD mechanisms of extrinsic (caused by absorbing inclusions) and intrinsic types have been revealed: a thermal explosion of absorbing inclusions, photoionisation of the material matrix surrounding an inclusion by thermal UV radiation of this inclusion heated by laser radiation, impact ionisation, and multiphoton ionisation of matrix atoms.

The basic characteristics of the damage processes were determined in the studies aimed to develope theoretical models (by Yu.K. Danileiko, A.S. Epifanov, M.F. Koldunov, A.A. Manenkov, A.M. Prokhorov, et al. [25, 28 – 33]). For extrinsic LD mechanisms, such characteristics are statistical LD properties caused by a random distribution of inclusions in the interaction region, the dependence of the LD threshold on the laser-pulse width, and also the features of the LD for the surface and optical coatings. For the intrinsic LD mechanisms, these are the dependences of the LD threshold on the laser-radiation frequency and pulse width and the temperature of the irradiated solid, and statistical LD properties determined by a lack of electrons initiating the impact ionisation (an electron avalanche). The relative role of impact and multiphoton ionisation has been also determined.

A comprehensive analysis [30] of the experimental data on LD in optical materials of different kinds under various experimental conditions performed on the basis of the developed theoretical models has shown the following:

(1) Many characteristics of LD (the statistical properties, characteristics of the damage under the action of single and multiple laser pulses, features of LD of optical coatings, the dependence of the LD threshold on the pulse width, etc.) in most of optical materials agree with the characteristics predicted by the theory of the inclusion thermal explosion mechanism. For example, Fig. 1 demonstrates a good agreement between the theory and experiment for the pulse-width dependence of the LD threshold over a wide range (10 ns – 20 ps).

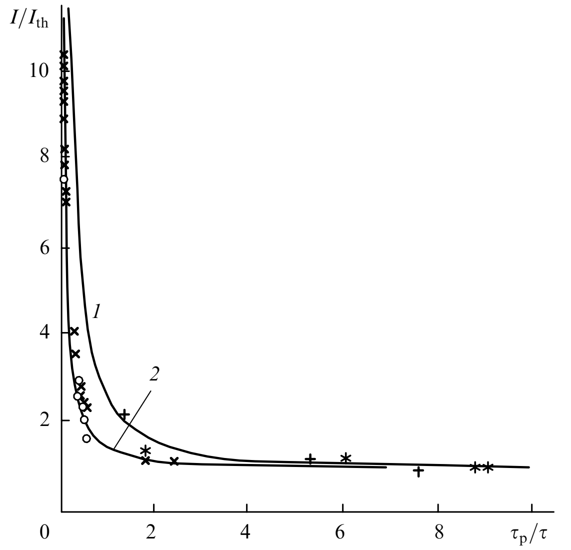


Figure 1. Laser-damage threshold I in SiO_2 as a function of the pulse duration $\tau_{\rm p}$ at $\lambda = 1.05$ μm, $w = 7.2$ μm, and $I_{\rm th} = 0.8$ TW cm^{-2} (×), $\lambda = 1.05$ μm, $w = 5.0$ μm, and $I_{\rm th} = 1.5$ TW cm^{-2} (○), $\lambda = 0.53$ μm, $w = 7.2$ μm, and $I_{\rm th} = 0.4$ TW cm^{-2} (+), and $\lambda = 0.53$ μm, $w = 5.0$ μm, and $I_{\rm th} = 0.75$ TW cm^{-2} (∗) (experimental data [34]). Solid curves are calculated on the basis of the theory of thermal explosion of absorbing inclusions [30] for (*1*) rectangular and (*2*) Gaussian laser pulses, $I_{\rm th}$ is the threshold LD intensity at $\tau_{\rm p} \to \infty$, λ is the laser wavelength, w is the size (diameter) of the focal spot of focused radiation at a level of 1/e of the maximum intensity at the beam centre, and τ is the temperature relaxation time of an inclusion (the theoretical and experimental data agree at $\tau = 20$ ns).

(2) Some LD characteristics (the dependences of the LD threshold on the laser frequency, the pulse width, and the sample temperature and the lack effect of seed electrons) agree with the predictions of the theory of the electron-avalanche mechanism only for a few specially selected (extremely pure) alkali-halide crystals.

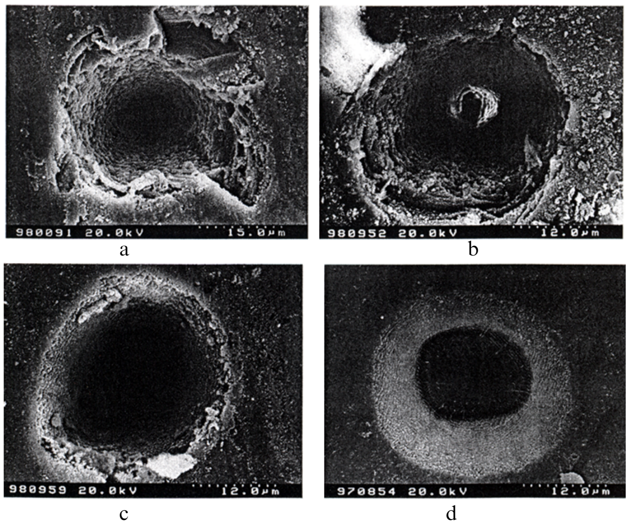
Hence, this analysis allows us to conclude that the extrinsic LD mechanism dominates in the range of nano- and picosecond laser pulses. The question as to what LD mechanism predominates in the ultrashort (femtosecond) regime requires a special consideration (see Section 4).

4. Recent data on LD caused by ultrashort pulses and discussion

Several works [1–4] published in the recent eight years are of greatest interest for understanding the physics of the LD phenomenon in the ultrashort pulse width range. These studies were devoted to the LD in fused silica (SiO_2), CaF_2 crystals, and multilayer dielectric coatings in a wide range of pulse durations (1 ns–5 fs). A significant difference in the LD characteristics for long (nano- and picosecond) and short (femtosecond) pulses has been revealed:

(i) The LD morphology in the case of long pulses is characterised by cracks and melted regions, while an ablation character of LD is observed for femtosecond pulses (Fig. 2).

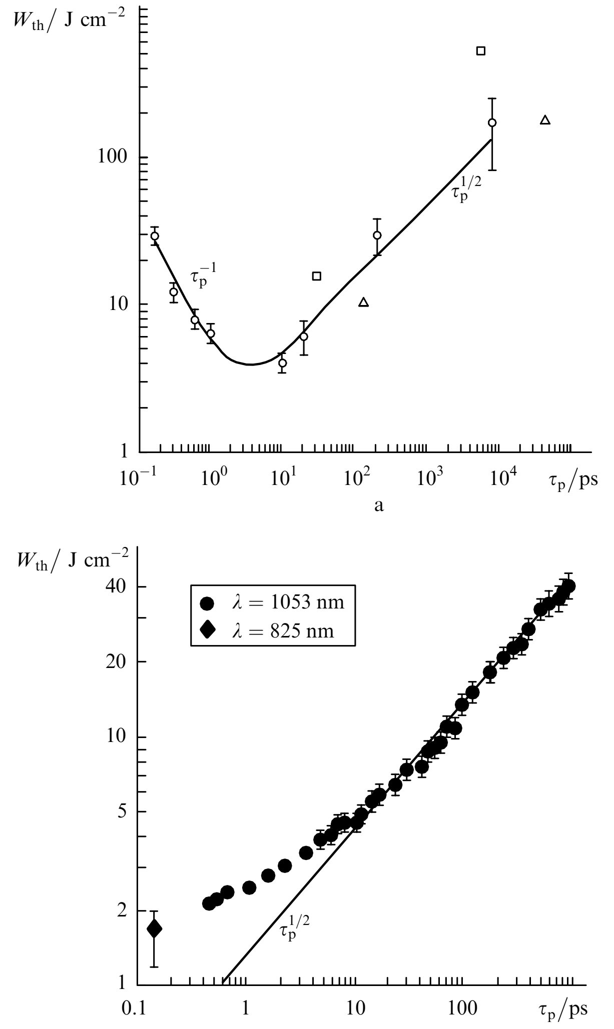
(ii) The LD threshold energy $W_{\rm th}$ as a function of pulse duration $\tau_{\rm p}$ is (Fig. 3): $W_{\rm th} \sim \tau_{\rm p}^{1/2}$ for $\tau_{\rm p} = 1$ ns–20 ps, $W_{\rm th} \to$ const (independent of $\tau_{\rm p}$) for $\tau_{\rm p} < 20$ ps (Stuart B.C. et al. [1] and Lenzner M. et al. [4]), and $W_{\rm th}$ increases for $\tau_{\rm p} < 20$ ps (Mourou G. et al. [2]).



a b

c d

Figure 2. The LD morphology of fused silica, which was irradiated with 80 780-nm pulses, observed at various pulse durations τ_0 and laser energy densities F_0 in the irradiated region: (a) $\tau_{\rm p} = 3$ ps, $F_0 = 19.9$ J cm^{-2}, (b) $\tau_{\rm p} = 220$ fs, $F_0 = 10.7$ J cm^{-2}, (c) $\tau_{\rm p} = 20$ fs, $F_0 = 11.1$ J cm^{-2}, and (d) $\tau_{\rm p} = 5$ fs, $F_0 = 6.9$ J cm^{-2} (the data from [4]).



a

b

Figure 3. The LD threshold $W_{\rm th}$ in SiO_2 as a function of the pulse duration $\tau_{\rm p}$ obtained in (a) [2] and (b) [1].

(iii) Statistical LD properties also differ: a decrease in the variance of the LD threshold and its independence of the size of the irradiation region are observed for femtosecond pulses.

Contradictory data for the dependence $W_{\rm th}(\tau_{\rm p})$ for femtosecond pulses obtained by different research groups are evidently caused by different methods used to determine the ablation threshold. For example, the LD threshold was determined in Ref. [2] by the onset of a laser-plasma glow in the interaction region under single-shot irradiation of the surfaces of the samples under study, whereas in Refs [1] and [4], the LD threshold was found, respectively, from the damage morphology under multiple-shot irradiation and from measurements of the volume of the ablated materia $V_{\rm a}(W)$ by extrapolating it to $V_{\rm a} = 0$ in the single- and multiple-shot irradiation regimes.

The interpretation of the aforementioned experimental results in Refs [1–4] was based on the assumption that the LD produced by ultrashort pulses is caused by intrinsic mechanisms (impact and multiphoton ionisations). The extrinsic LD mechanisms, i.e., those associated with the effect of absorbing inclusions, were neglected, although the dependence $W_{\rm th}(\tau_{\rm p})$ for long pulses ($\tau_{\rm p} > 20$ ps) was explained in Ref. [1] by a thermal mechanism with a reference to works in which the diffusion law $W_{\rm th} \sim \tau_{\rm p}^{1/2}$ was attributed to the LD mechanism due to inclusions. The theoretical analysis in Ref. [1] was based on the quantum kinetic equation (QKE) for electrons excited in the conduction band by impact ionisation (a multiphoton ionisation was considered as a source of seed electrons). The QKE was solved in the so-called diffusion approximation developed earlier in Ref. [29]. The approaches to the interpretation of the experimental data on LD mentioned above seem to be doubtful for the following reasons.

The assumption of the predominance of intrinsic LD mechanisms in the investigated materials is not substantiated. Neglecting the influence of absorbing inclusions on the experimental regularities cannot be justified. Moreover, we have shown [31] that one of the most important LD regularities – the damage threshold as a function of the pulse width – is explained well by the mechanism of a thermal explosion of absorbing inclusions (Fig. 4). A good agreement between the experiment and a thermal explosion theory indicates that the absorbing inclusions play a significant role in the LD processes in a wide range of pulse widths, including ultrashort (femtosecond) pulses. This is also confirmed by the observation of an incubation effect in the laser ablation of $\rm SiO_2$ by 5-fs pulses [4], which can hardly be accounted for intrinsic LD mechanisms.

The dependence of the LD threshold on the pulse duration $W_{\rm th} \sim \tau_{\rm p}^{1/2}$, used in [1] for explaining the experimental data for long pulses has no theoretical grounds: a correct rigorously substantiated function $W_{\rm th}(\tau_{\rm p})$ for the thermal mechanism significantly differs from $W_{\rm th} \sim \tau_{\rm p}^{1/2}$ (Fig. 1).

The diffusion approximation of the QKE is correct only for $\tau_{\rm p} \geqslant 10$ ps [29] and requires a special analysis of the conditions for extending the region of applicability of this approximation to the range of ultrashort pulses ($\tau_{\rm p} <$ 10 ps). In particular, for ultrashort pulses with $\tau_{\rm p} < \tau_{\rm e-ph}$ ($\tau_{\rm e-ph}$ is the characteristic time of electron–phonon relaxation), one should take into account that electron–phonon collisions may be inefficient in impact-ionisation processes.

Within the framework of the interpretation offered in Refs [1–4], it is impossible to account for a change in the morphology from the formation of cracks or melting to an ablation-caused removal of the surface material, when the irradiation regime changes from long ($\tau_{\rm p} > 20$ ps) to ultrashort pulses. This fact was explained on the basis of the analysis of the thermoelasticity problem solution for laser-heated solids [32]. Let us briefly examine the results of this analysis. First, we present the arguments in favour of the applicability of thermoelasticity equations, whose solution forms the basis of this analysis [32], in a wide range of pulse widths, including ultrashort pulses.

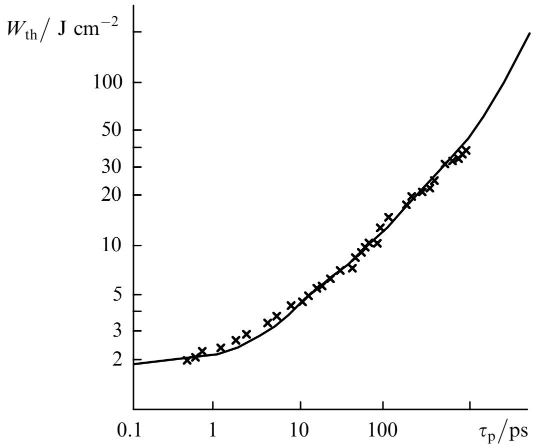


Figure 4. The LD threshold $W_{\rm th}$ in $\rm SiO_2$ as a function of the pulse duration $\tau_{\rm p}$: (dots) experimental data obtained at a laser wavelength of 1.053 μm and (solid curve) calculation [31] on the basis of the theory of absorbing inclusion thermal explosion (a model of three types of inclusions).

The characteristic times of the processes occurring in a solid exposed to pulsed laser heating have the following orders of magnitude: the electron–phonon relaxation time is $\tau_{\rm e-ph} \sim 10^{-12}$ s, the time in which thermoelastic stresses reach steady-state values is $\tau_{\rm s} \sim 10^{-9}$ s, and the crack-formation time is $\tau_{\rm f} \sim 10^{-8}$ s. Comparing these times shows that a mechanical damage of a solid resulting from a local laser heating is the slowest process. When femtosecond pulses heat a solid, a damage appears after their action is completed. This implies that the formation of a mechanical damage in solids exposed to a pulsed laser irradiation, including the action of ultrashort pulses, can be adequately analysed on the basis of the thermoelasticity equations. Such an approach used in Refs [32–34] allowed to establish the conditions for the onset (or absence) of a mechanical damage (a crack) in a solid under pulsed laser heating. In this case, an arbitrary (intrinsic or extrinsic) absorption mechanism of laser radiation, which caused the local heating, is assumed. The following two crack-formation criteria were established: the force criterion,

$$\max_{r,t} \sigma_{\phi}(r, t) \geqslant \sigma_{\rm th}, \qquad (1)$$

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

$$\eta\vartheta E_{\rm p} \geqslant E_{\rm m}, \qquad (2)$$

where $E_{\rm p}$ is the laser-pulse energy; η is the factor determining the fraction of the absorbed energy; $\vartheta =$

$(T_0/9c_V)[1+ v/(1-v)^2]\alpha^2 c_{\mathrm{long}}^2$ is the connectivity coefficient; c_V is the specific heat; α is the linear expansion coefficient; v is the Poisson coefficient; c_{long} is the longitudinal velocity of sound; $E_{\mathrm{m}} = 39R^2\gamma$ is the mechanical deformation energy, R is the size of the heated region; and γ is the surface-energy density.

When criteria (1) and (2) are satisfied, a mechanical damage in the form of a crack may arise, if the parameters of the laser pulse and the size of the irradiated region satisfy the relations

$$\tau_{\mathrm{p}} \geqslant \tau_{\mathrm{cr}} \simeq \frac{78R^2\gamma}{\eta\vartheta F_{\mathrm{th}}}, \tag{3}$$

$$R \geqslant R_{\mathrm{cr}} \simeq \frac{10\gamma}{\vartheta k_{\mathrm{a}} W_{\mathrm{th}}}, \tag{4}$$

where F_{th} is the threshold radiation power in the interaction region; W_{th} is threshold energy density; and k_{a} is the absorption coefficient.

Formulas (3) and (4) yield the following estimates for $\mathrm{SiO_2}$ at $k_{\mathrm{a}} \simeq 10^5 - 10^4$ cm^{-1}: $\tau_{\mathrm{cr}} \simeq 50$ ps and $R_{\mathrm{cr}} \simeq 0.1 - 0.7$ μm. These values are in reasonable agreement with experimental data and account for a change in the LD morphology from cracking (melting) to an ablation (absence of cracks) at laser-pulse widths $\tau_{\mathrm{p}} < 20$ ps. These analytical results show that the LD morphology is independent of the absorption mechanism, being determined by the amount of the absorbed energy and mechanical parameters of materials.

In other words, a change in the LD morphology from cracking to ablation observed in experiments, when the irradiation regime changes from nanosecond to femtosecond pulses, does not testify to a change in the absorption mechanism (from extrinsic to intrinsic one) but is more likely a consequence of a decrease in the LD threshold due to the pulse shortening. The latter fact is naturally explained within the framework of the theory of thermal explosion of absorbing inclusions by a reduced effect of the heat conduction from the interaction region, although the intrinsic mechanisms (impact and multiphoton ionisations) also lead to a similar decrease in the LD threshold (due to an electron diffusion from the interaction region).

The change in the LD morphology from cracking to ablation physically means that, when the pulse width decreases to a certain value τ_{cr}, the energy absorbed in the interaction region becomes insufficient for forming a crack and ablation becomes the dominating LD mechanism. Obviously, an ablation is also present under the irradiation by long pulses, but it is masked by a more pronounced cracking process.

The consistent explanation of the observed regularities of the LD induced by ultrashort pulses (the morphology, the LD threshold as a function of the pulse width, etc.) within the framework of the thermal-explosion theory is a convincing argument in favour of a significant or even predominating role of absorbing inclusions in LD processes in solids in a wide range of pulse widths, including femtosecond pulses. However, an unambiguous solution to the problem of the dominating LD mechanism in the region of ultrashort pulses requires further studies of the role of the intrinsic mechanisms of laser-radiation absorption (impact and multiphoton ionisations), for which, as was mentioned above, there is no adequate theory applicable to ultrashort pulses. Moreover, the processes of laser ablation of transparent solids dominating at ultrashort pulses are far from being comprehensively studied. In particular, no precise definition (both theoretical and experimental) of the ablation threshold can be found in the literature, and its physical mechanisms have not been studied and even formulated.

5. Conclusions

The analysis of the recent experimental data on the LD of transparent solids in a wide range of pulse durations (including femtosecond pulses) presented in this paper and the interpretation of these data on the basis of the theoretical concepts of the LD mechanisms lead to the following conclusions. Precise and unambiguous theoretical and experimental definitions of the threshold of laser ablation, which dominates in the LD by ultrashort pulses, have not been formulated up to now. The methods of defining the ablation threshold and its criteria used by different research teams complicate the comparison of the experimental results. Nevertheless, certain important LD regularities observed in different pulse-duration ranges can be convincingly interpreted on the basis of the developed theoretical concepts and models. These are primarily the dependence of the LD threshold W_{th} on the pulse duration τ_{p} and the modification of the damage morphology caused by variations in the pulse duration. It is shown that the function $W_{\mathrm{th}}(\tau_{\mathrm{p}})$ is explained well on the basis of the mechanism of thermal explosion of absorbing inclusions in a wide range of pulse durations (from nano- to femtosecond pulses).

A change in the LD morphology from cracking to ablation upon changes in the irradiation regime, when nano- or picosecond pulses are replaced by ultrashort femtosecond pulses, are consistently explained by the model of thermoelastic damage. As is shown, the interpretation of the aforementioned regularities (the function $W_{\mathrm{th}}(\tau_{\mathrm{p}})$ and changes in the LD morphology) presented in several works, which is based on the impact ionisation mechanism, cannot be admitted as adequate. Although many facts and their explanation show a significant and even decisive role of absorbing inclusions in the LD for different pulse durations, solving the problem of the relative role of intrinsic (impact and multiphoton ionisations) and extrinsic (initiation by absorbing defects) LD mechanisms in the region of ultrashort pulses requires additional theoretical and experimental investigations. In particular, an impact-ionisation theory applicable to ultrashort pulses and a laser-ablation theory must be developed. Such studies are important for both the fundamental physics of the interaction of laser radiation with transparent solids and practical applications of high-power ultrashort pulses aimed, for example, at a further development of high-power laser engineering and high-precision processing of materials.

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