Fundamental mechanisms of laser-induced damage in optical materials: today's state of understanding and problems

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Abstract. Theoretical models of laser-induced damage mechanisms in optical materials are reviewed: inclusion-initiated thermal explosion (extrinsic mechanism) and impact ionization (II) and photoionization (intrinsic mechanisms). Different approaches to II theory based on quantum kinetic equation, Boltzman equations, and rate equations are briefly described. A relative contribution of II and photoionization predicted by these models at different laser pulse durations, including femtosecond-range, are discussed and compared with available experimental data. Basing on an analysis of published theoretical and experimental results, a today's state of understanding fundamental laser damage mechanisms is concluded. © *The Authors. Published by SPIE under a Creative Commons Attribution 3.0 Unported License. Distribution or reproduction of this work in whole or in part requires full attribution of the original publication, including its DOI.* [DOI: 10.1117/1.OE.53.1.010901]

1 Introduction

Laser-induced damage (LID) in optical materials plays a significant role in laser-matter interaction phenomena. This role can be both negative (a factor which limits laser intensity in high-power laser systems or material under study) and positive (providing a tool for material processing and modification). In this context, understanding of fundamental mechanisms of LID is very important for both laser science and its applications.

On this reason, research in this field is so active and nature of the LID mechanisms is a subject of many laser conferences and publications. A number of models for the LID mechanisms were suggested and investigated during almost 50 years of research since the first observation of laser damage.[1–3]

The LID mechanisms, related mostly to nanosecond-picosecond laser pulse duration range, have been earlier reviewed rather comprehensively in Ref. 4. During last two decades, major interests in LID studies were focused mostly to sub-picosecond pulse duration range due to development of ultra high-power lasers based on chirped-pulse amplification concept.[5] A huge number of papers were published, aimed at elucidating the mechanism of LID in this pulse duration range. However, there are still no publications that review experimental and theoretical results and attempt to critically analyze suggested models of LID in this range.

This article, based on an invited talk of the author presented at Pacific Laser Damage Symposium (PLD 2013, Shanghai, China, May 19-22, 2013), is perhaps a first attempt in this direction.

Although the LID phenomenon consists of several stages, we concentrate our analysis on mechanisms of initial stages: nonlinear absorption of laser radiation in initially transparent lattice of a material and temperature rise of the lattice to levels high enough to produce phase transitions. Initial stages are major processes that determine LID resistance of the material, and final stages (melting, crack-formation, or ablation) determine morphology of the damage sites.

A review of morphological studies is out of a scope of this article. We can refer to only two recent publications on this topic,[6,7] where some aspects of the problem have been investigated rather comprehensively.

2 Extrinsic and Intrinsic LID Mechanisms: General Survey

It seems reasonable to start describing the LID mechanisms that were proposed in the literature with a general survey. These mechanisms may be divided into two classes: extrinsic mechanisms associated with absorbing inclusions, and intrinsic ones which are inherent to intrinsic processes in pure defect-free materials.

At earlier stages of research, various models of extrinsic LID mechanisms were proposed:[8–10] they are based on consideration of laser-produced heating of absorbing inclusions and heat transfer to surrounding media (surface or bulk host materials). All these models were similar in physical principles: they assumed a temperature independence of optical, thermal, and elastic properties of materials. Although these models explain some damage characteristics, they are not quite adequate because the assumption of temperature independence is questionable. Indeed, a temperature during a laser damage process can be very high, $>10^4$ K, and dependence of material properties should be essential.

A more realistic model was suggested[11,12] that took into account the temperature dependence of material properties. Such an account changes the LID concept principally: inclusion heating assumes an explosion character. As discussed below, this thermal explosion (TE) model explains pretty well experimentally observed LID features (in particular, pulse-width dependence of damage threshold).

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Among the possible intrinsic mechanisms there were proposed the following: electrostriction, hyper-sound generated at stimulated Brillouin scattering, electron impact ionization (II), and photoionization. However, only II and photoionization (PI) were considered most effective, especially at ultrashort laser pulse duration. In the next section, grounds of the above mentioned extrinsic and intrinsic damage mechanisms and some of their predictions are briefly described.

3 Extrinsic and Intrinsic Mechanisms: Grounds and Predictions

3.1 Inclusion-Initiated Thermal Explosion Model

The TE model is based on the heat transfer equation:

$$\frac{\partial}{\partial t}(C\rho T) = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 k \frac{\partial T}{\partial r}\right) + Q(I,T), \qquad (1)$$

where c, ρ, and k are, respectively, heat capacity, density and thermal conductivity of inclusion, and host material, $Q(I,T)$ is power of heat sources associated with laser radiation absorption by inclusions, I is the laser beam intensity, and T is the temperature

The key point of the TE model is the temperature dependence of Q. In accordance with experimental data, it can be taken in the form:[11]

$$Q(I,T) = Q(I) \exp\left(\xi \frac{T - T_0}{T_0}\right), \qquad (2)$$

where ξ is a material parameter, and T_0 is the initial temperature.

A solution of Eq. (1) showed that heating of inclusions has an explosive character: the inclusion temperature increases infinitely at the laser intensity approaching the threshold ($T \to \infty$ at $I \to I_{\text{th}}$). This allows one to determine damage characteristics [in particular, pulse-width dependence of the damage threshold, $T_{\text{th}}(\tau_p)$] without analyzing phase transitions in a host material (crack formation, melting, etc).

An important development of the TE model consisted of a suggestion of the inclusion-initiated photoionization mechanism of laser damage.[12] The concept of this mechanism is the following. The inclusion temperature at the damage threshold can be rather high (estimated as $\sim 7 \times 10^4$ K). At such a temperature, a thermal radiation is emitted in UV-spectral range ($\lambda_{\text{max}} \approx 50$ nm). This UV thermal radiation is an efficient source for photoionization of surrounding dielectric host material creating free carriers (FC). So, UV-excited FC becomes an additional absorption source of laser radiation producing expansion of the inclusion-initiated TE. Besides the UV-thermal radiation, other sources of inclusion-initiated absorption in the surrounding matrix are also possible: photochemical reactions,[13] thermoionic emission of electrons from inclusions, and matrix band-gap collapse.[6] However, these mechanisms can be realized only in rather specific materials and laser irradiation regimes. In contrast, the UV-thermal radiation is the most universal mechanism which can be effective in many types of inclusions and host materials, and at different irradiation regimes, including a variety of inclusions (metallic, semiconductor, ceramic), wide band-gap dielectrics, laser pulse lengths, and operation regimes.

A comprehensive analysis of the TE model[14] resulted in deriving pulse-width and pulse-shape dependence of LID threshold. In particular, for rectangular and Gaussian pulse-shapes at $\tau_p \geq \tau$ (τ_p is pulse width, τ is heat relaxation time in a medium) the pulse-width dependence of laser-induced damage threshold (LIDT) is approximated by the following formulas:

$$I^{\text{root}}(\tau_\rho) = \frac{I_{\text{th}}}{1 - \exp(-\tau_p/2\tau)}. \qquad (3)$$

$$I^{\text{gauss}}(\tau_\rho) = I_{\text{th}} \exp[(\tau/\tau_p)^2]. \qquad (4)$$

Pulse-width dependence of LIDT, predicted by the TE model, has been proved by comparison with experimental data obtained by two research groups that investigated LID in nanoseconds-picoseconds pulse-width range on different materials.[15,16] Results of this comparison are shown in Figs. 1(a) and 1(b).

A good agreement between predicted and experimental data (for Gaussian pulse-shape), as seen in Figs. 1(a) and 1(b), confirms adequacy of the TE model in a wide range of laser pulse widths.

An important feature of pulse-width dependence of LIDT, predicted by the TE model, is a significant influence of the laser pulse-shape. Note in this context that such an influence is not taken into account in many LID-related publications (for more detailed discussion of this subject, see Ref. 7). Note also that the linear approximation model of inclusion-initiated LID, not accounting for temperature dependence of inclusion material parameters, predicts quite different pulse-width dependences of the LID threshold.[17,18] In particular, "diffusion rule" for the threshold fluence $F_{\text{th}} = \sqrt{D}\,\tau_p$ where D is the thermal diffusion coefficient[17] that is used in many publications on pulse-width dependence of the LID threshold, does not explain experimental data. Furthermore, a deviation of the experimentally observed dependence $F_{\text{th}}(\tau_p)$ from this "diffusion rule" was erroneously attributed for a change of the damage mechanism (from extrinsic inclusion-initiated mechanism to an intrinsic one).[19]

3.2 Electron Impact Ionization

Different theoretical approaches have been used in theoretical studies of II as intrinsic LID mechanism. They were based on the quantum kinetic equation (QKE) for energy distribution function of electrons in a conduction band of dielectric material,[19,20–23] Baltzman equations for energy distribution functions of electron and phonon systems,[24] and phenomenological rate equations for free electron density in the conduction band.[25,26]

A most comprehensive theoretical investigation of LID based on the QKE was carried out by Epifanov et al.[21–23] The general form of the QKE is written as

$$\frac{\partial f(P,t)}{\partial t} = \frac{2\pi}{h}\sum_q^r B(q) \sum_{n=-\infty}^{\infty} J_n^2 \left\{\frac{eEq\tau(p)}{hm\Omega[1 + \Omega^2\tau^2(p)]^{1/2}}\right\}$$

$$\times \{[f(p+q)(N_q+1) - f(p)N_q]\delta[\varepsilon(p+q) - \varepsilon(p)$$

$$- h\omega_q - nh\Omega] + [f(p+q)N_q - f(p)(N_q+1)]\delta$$

$$\times [\varepsilon(p+q) - \varepsilon(p) + h\omega_q - nh\Omega]\}, \qquad (5)$$

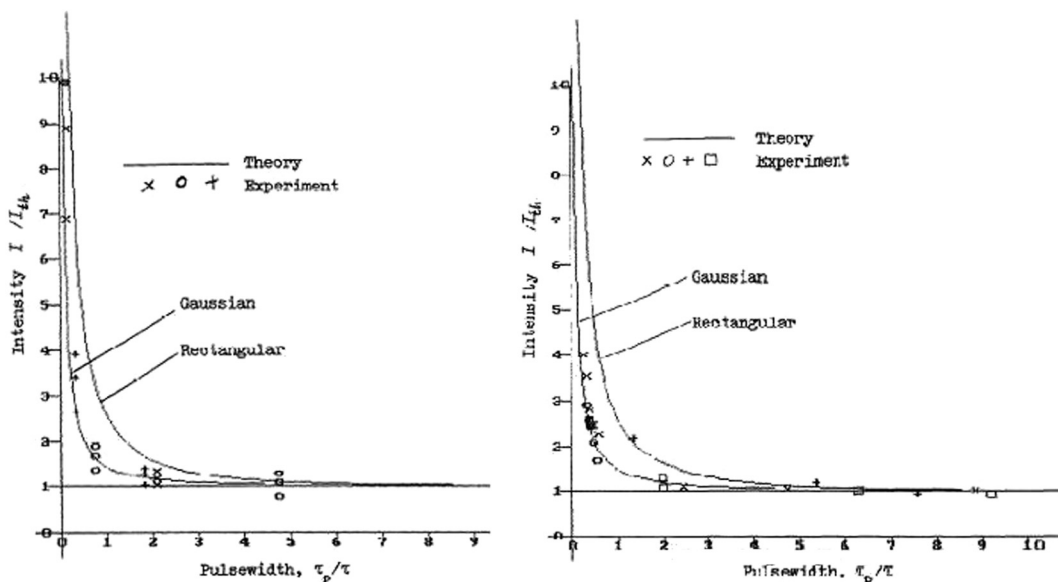**Fig. 1** (a) Damage threshold versus laser pulse width in alkali—halide crystals at 1.06 μm. Experimental data:[15] x- NaCl, $I_{th} = 10$ GW/cm^2, $\tau = 1$ ns, o-KCl, $I_{th} = 5$ GW/cm^2, $\tau = 3$ ns, $\pm$KBr, $I_{th} = 4$ GW/cm^2, $\tau = 8$ ns. (b) Damage threshold versus laser pulse width in SiO$_2$ at different wave lengths, λ. Experimental data:[16] x – $I_{th} = 0.8$ TW/cm^2, $\tau = 20$ ps, $\lambda = 1.05$ μm, o – $I_{th} = 1.5$ TW/cm^2, $\tau = 20$ ps, $\lambda = 1.05$ μm, $\pm I_{th} = 0.4$ TW/cm^2, $\tau = 20$ ps, $\lambda = 0.53$ μm, $- I_{th} = 0.75$ TW/cm^2, $\tau = 20$ ps, $\lambda = 0.53$ μm.

where $f(p, t)$ is the energy distribution function of electrons, $p(\varepsilon)$ is the electron impulse with energy ε, $B(q)$ is matrix element of electron–photon interaction, N_q is a number of phonons with wave vector q and energy $h\omega_q$, e and m are the electron charge and mass, respectively, $\tau(\varepsilon)$ is the electron relaxation time, and E and Ω are the electric field strength and frequency of laser radiation, respectively.

This equation is approximated by the diffusion Focker-Planck equation at relatively low laser frequencies, Ω, as compared with the ionization potential, I (at $h\,\Omega \ll I$), and is transformed into the differential kinetic equation at $h\,\Omega < I$.

A solution of these equations, supposed in the form $f(x, t) = f(x) \exp(\gamma t)$, allowed us to investigate dependence of the avalanche rate, γ, on the laser field strength, E, and

frequency, Ω. Different behaviors of $\gamma(E)$ in different frequency and pulse-duration ranges were predicted:

$$\gamma \infty \exp(E), \quad \text{at } h\Omega \ll I, \quad \tau_p = 10^{-7} \div 10^{-11} \text{ s}$$

$$\gamma \infty E^2, \quad \text{at } h\Omega \ll I, \quad \tau_p < 10 \text{ ps},$$

$$\gamma \infty E^{2(n+1)}, \quad \text{at } I/h\Omega = n, \quad n = 1 \div 4$$

Such a behavior of $\gamma(E)$ leads to a different frequency dependence of the critical breakdown field at various pulse widths, as shown in Fig. 2.

4 Laser Damage Criterion

Damage criterion is a significant aspect of the LID theory for any damage mechanism. For the electron avalanche mechanism, such a criterion has to be determined from a solution of combined equations describing kinetics of avalanche electrons in a conduction band and material lattice heating due to electron–phonon collisions. In particular, when carrier recombination is disregarded, these equations are:[23]

$$\frac{dN}{dt} = \gamma^0 \varphi(\Theta)N, \qquad \frac{d\Theta}{dt} = \beta\Theta \times N, \qquad (6)$$

where $\theta = T/T_0$, T_0 is the lattice initial temperature, and β and χ are the parameters that depend on a type of electron-phonon interaction.

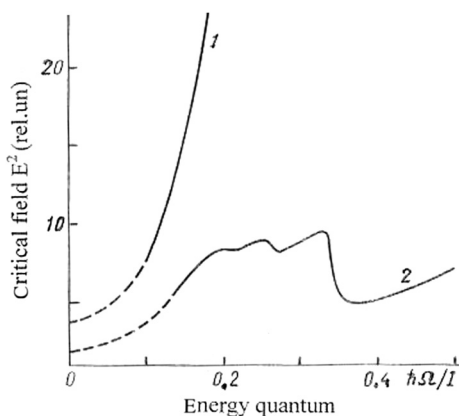The solution of these equations gives a breakdown criterion in the form:

Fig. 2 Frequency dependence of critical field for avalanche breakdown at different pulse widths: 1 – $\tau_p = 30$ ps, 2 – $\tau_p = 100$ ns.

$$\gamma\tau_p = \ln(1 + 2\gamma/\beta N_0) \qquad (7)$$

where τ_p is the laser pulse duration (rectangular pulse-shape is assumed), N_0 is the initial electron concentration.

Use of this criterion with the avalanche rate, γ, determined from a solution of Focker–Plank or differential equations, allows one to determine critical breakdown field, E, and its dependence on laser radiation parameters (Ω, τ_p) and material parameters.

Note that in many publications, a plasma critical density is assumed as breakdown criterion. However, such a criterion has not been grounded by any consistent theoretical model. Therefore, its correctness is questionable and special analysis is required to elucidate this problem.

Damage criteria considered in this section determine threshold intensity of laser radiation in laser interaction region for formation of any phase transition in a material. Determination of laser energy density threshold for realization of particular phase transitions (melting, crack formation, or ablation) requires a special analysis of laser material interaction. In particular, such an analysis has been performed for laser-produced melting and ablation (of SiO_2 surface)[27] and for crack formation (in bulk materials).[7]

5 Deterred Avalanche

The above consideration of II related to a case when initial concentration, N_0, of seed electrons in an interaction volume, V, is high enough: $N_0 V > 1$. In this case, the breakdown threshold only slightly depends on N_0, as seen in Eq. (7). In contrast, when $N_0 V < 1$, characteristics of breakdown significantly depend on N_0: the electron avalanche is deterred because of the lack of seed electrons.[28,29]

In this case, the birth of seed electrons becomes a bottleneck of LID. The damage process takes a statistical character: the breakdown threshold is determined by the probability of seed electron appearing. An effective mechanism of seed electron generation can be associated with photoionization of host material or impurity atoms by any radiation source.

6 Experimental Confirmation of Impact Ionization

Realization of the II in LID was a subject of many experimental studies. Comprehensive investigations of some LID characteristics, indicative for this mechanism (temperature and frequency dependences, in particular), were carried out in 1970s on a large variety of alkali-halide crystals in a wide range of laser wavelengths (10.6, 1.06, 0.53, 0.69, 2.76 μm).[30,31] It was found that LID threshold in the majority of samples varied significantly from sample to sample indicating influence of impurities and defects. However, in some very pure samples of NaCL, KCl, and KBr, observed dependence corresponded to theoretical predictions. Figure 3 demonstrates some of those results.

These experimental results were interpreted as follows. At ruby and Nd:YAG laser wavelengths, 0.69 and 1.06 μm, the observed temperature dependence of LIDT correlates quite well with that predicted by the "normal avalanche" theory (sufficient number of seed electrons). At second harmonic Nd:YAG wavelength, LID is due to multiphoton (MP) ionization. At CO_2-laser wavelength, 10.6 μm, the avalanche is deterred by the lack of seed electrons. At Er:CaF$_2$-laser wavelength, 2.76 μm, "normal avalanche" is realized at temperatures $T \leq 600$ K, whereas it is deterred at $T \geq 600$ K.
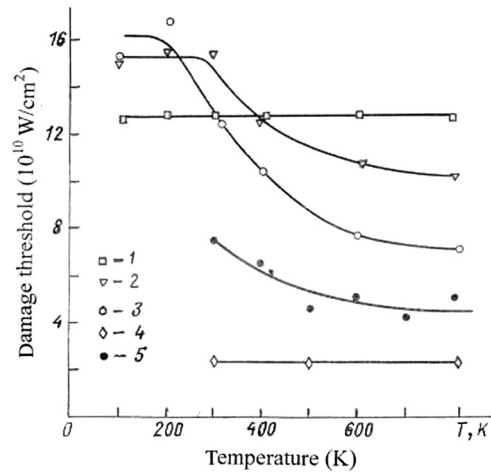

Fig. 3 Temperature dependence of LIDT in NaCl at various laser wave lengths: 1–0.53 μm, 2–0.69 μm, 3–1.06 μm, 4–10.6 μm (Gomelauri and Manenkov[31]), 5–2.76 μm (Gorshkov et al.[30]).

"Deterred avalanche" at 10.6 μm was directly confirmed in two-frequency experiment:[32] when a NaCl crystal was additionally irradiated with low-power N$_2$-laser ($\lambda = 0.337$ μm), the damage threshold was reduced significantly (6 times). This effect definitely indicates that N$_2$-laser irradiation creates a sufficient number of seed electrons to initiate the avalanche.

7 Photoionization: Relative Contribution of Impact Ionization and Photoionization

A general theory of photoionization in solids under strong electromagnetic fields was developed by Keldysh[33] who formulated probability of ionization at various frequencies and field strengths. It was shown that at low frequencies and high field strengths the ionization probability coincides with that of tunnel autoionization, whereas at high frequencies it corresponds to MP ionization.

In application to LID processes, photoionization can play a double role, as a source of seed electrons for II, and as a dominating mechanism of damage, if a photoionization rate exceeds an II rate.

Relative contribution of photoionization and II in LID was investigated by several research groups that used different approaches.

An analysis conducted by Gorshkov et al.[23] was based on a solution of combined rate equations for electrons in a conduction band, generated by II and MP ionization, and for the lattice temperature:

$$\frac{d\theta}{dt} = \beta \theta^{\chi} N, \tag{8}$$

$$\frac{dN}{dt} = \gamma N + W_n - R(N). \tag{9}$$

Here, N is the electron concentration, γ is the II rate, W_n is the n-photon ionization rate, $R(N)$ is the electron recombination term, $\theta = T/T_0$, T_0 is the initial lattice temperature, β and κ are the electron-phonon interaction parameters
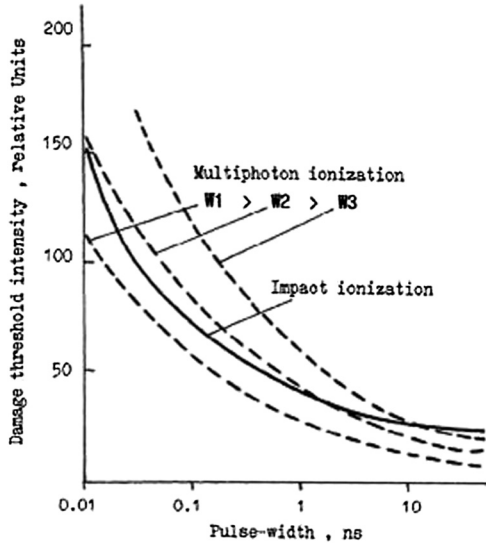
Fig. 4 Pulse-width dependence of damage threshold in NaCl: predicted[23] for impact and multiphoton (MP) ($n = 5$) ionization mechanisms. W1, W2, and W3—MP ionization rates.

$$\left(\frac{\partial}{\partial t}\right)f(k,t) = \left(\frac{\partial f(k,t)}{\partial t}\right)_{\text{e-e}} + \left(\frac{\partial f(k,t)}{\partial t}\right)_{\text{e-ph}}$$
$$+ \left(\frac{\partial f(k,t)}{\partial t}\right)_{\text{e-ph-pt}} + \left(\frac{\partial f(k,t)}{\partial t}\right)_{\text{imp}}$$
$$+ \left(\frac{\partial f(k,t)}{\partial t}\right)_{\text{sefi}} \tag{10}$$

$$\left(\frac{\partial}{\partial t}\right)g_\beta(q,t) = \left(\frac{\partial g_\beta(q,t)}{\partial t}\right)_{\text{ph-e}}, \tag{11}$$

where terms labeled by e-e, e-ph, e-ph-pt, imp, sefi, ph-e, and ph-e-pt describe electron-electron, electron-phonon, phonon-assisted photon interactions, II, and strong electric field ionization (SEFI), i.e., photoionization, respectively.

A numerical solution of Boltzman integro-differential equations allowed authors to analyze temporal evolution of occupation numbers of electrons, $f(\varepsilon)$, and of phonons, $g(\varepsilon)$, as functions of kinetic electron energy, ε. This analysis was especially addressed to femtosecond pulse duration breakdown.

The most important results of this solution respecting a relative contribution of II and SEFI (MP/ tunneling) in LID of SiO_2 are shown in Fig. 5.

As seen in this figure, the contribution of II becomes important only at $t > 50$ fs. The contribution of photoionization (SEFI) is dominant at all pulse lengths. Only at $\tau_p = 200$ fs the contribution of II is comparable with that of photoionization.

Having analyzed the contribution of II at ultra-short times, the authors[24] came to a conclusion that simple rate equation
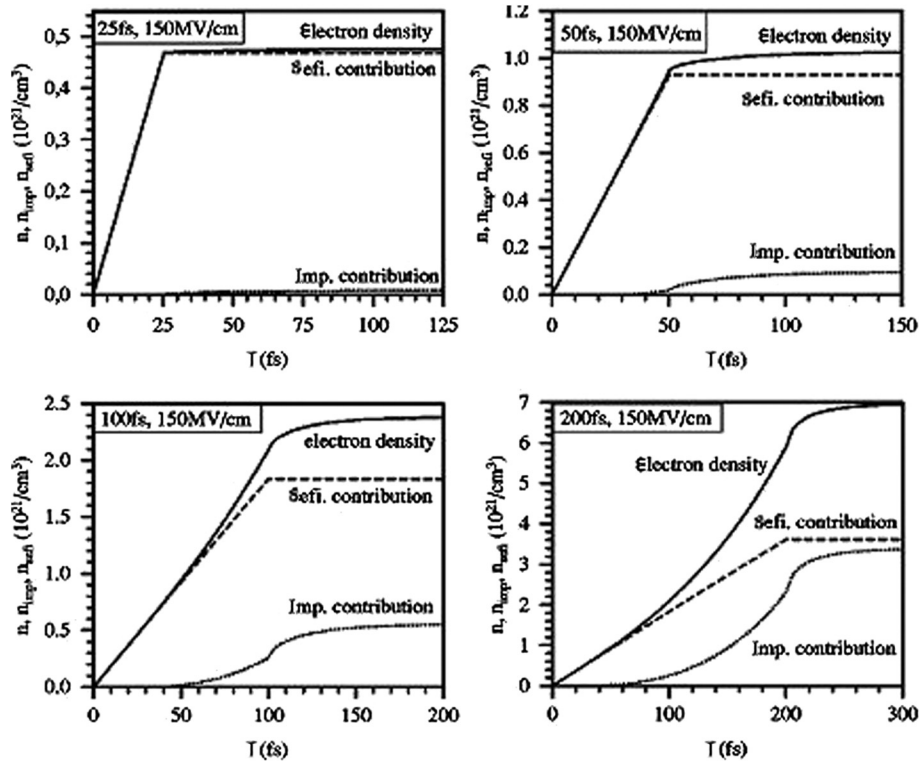
Results of this analysis are shown in Fig. 4, where pulse-width dependence of LIDT in NaCl is presented at various n-photon ionization rates and some fixed II rate.

A competition of MP and II mechanisms is seen in Fig. 4: in some pulse-width range MP dominates over II.

Another approach to elucidate a relative contribution of II and photoionization was used by Kaizer et al.[24] It was based on Boltzman equations for occupation numbers for electrons, $f(k,t)$, and phonons, $g(q,t)$:

Fig. 5 Time dependence of free-electron density and contributing processes in SiO_2 during and after laser pulse irradiation at various pulse lengths.[24]

(SRE) is not applicable to the analysis of breakdown in femtosecond-duration range, $\tau_p < 200$ fs. Detailed analysis of this problem led to a modification of SRE model: the multiple-rate equation (MRE) model was proposed.[25]

An application of MRE model allowed one to predict[26] that a definite transient time ($t \approx 100$ fs) is required for development of II. It also explained the results obtained in Ref. 24—that avalanche ionization does not appreciably contribute to free electron generation at ultra-short laser pulse durations, $\tau_p < 100$ fs.

For elucidating a relative contribution of II and photoionization, a new approach was proposed by Chimier et al.[27] Their approach was based on the rate equation and took into account photoionization, $\omega_{pi}(U_g)$, and II, $\omega_{ii}(U_g)$, terms of valence electrons, and similar terms of self-trapped excitons. Photoionization term $\omega_{pi}(U_g)$ was analyzed using Keldsysh formulation. For II term $\omega_{ii}(U_g)$, Fermi-liquid theory was used to describe free electron evolution in a conduction band.

This model was applied to investigation of damage and ablation in SiO_2 under femtosecond-laser pulse irradiation (results of these studies are briefly presented in Sec. 8).

8 Relative Contribution of II and Photoionization: Comparison with Experiment

Different experimental approaches have been used to elucidate a relative contribution of II and photoionization in femtosecond LID.

Chimier et al.[27] investigated pulse-width dependence of damage (melting) and ablation thresholds in SiO_2 in the wide pulse-width range, from 7 to 350 fs, using a laser source at 800-nm wavelength. Results of these studies, both observed and simulated on a base of the model described in the previous section, are shown in Fig. 6.

The observed dependences were well explained by the proposed model if assumed:

- both photoionization and II contribute to damage and ablation in whole laser pulse duration range, $\tau_p = 7 \div 300$ fs.
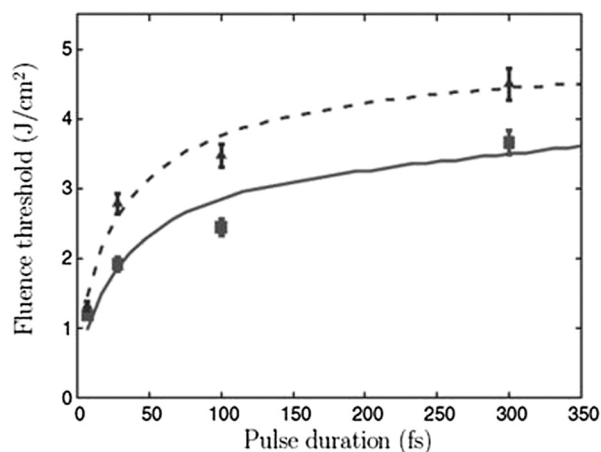

Fig. 6 Evolution of damage and ablation fluence thresholds as a function of laser pulse duration. Experimental data (squares and triangles) and numerical simulations (solid and dashed lines) correspond to damage and ablation, respectively. After Chimier et al.[27]

- Photoionization is a dominant mechanism at $\tau_p < 50$ fs, and II becomes predominant at $\tau_p > 50$ fs.

  It was also found that photoionization cannot be described by MP approximation at $\tau_p < 300$ fs: at $\tau_p < 50$ fs tunnel ionization becomes important.

Based on a double laser pulse excitation, and using a pump-probe interferometry technique for detecting free electrons generated at laser excitation, Mouskeftaras et al.[34] came to quite different conclusion on relative contribution of PI and II in laser ablation. The idea of their experiment was as follows. A sample (Al_2O_3 or SiO_2) was excited by two pulses: first 50-fs duration pulse at 400-nm wavelength, and second 800-nm pulse with variable duration and delay.

It was expected that first, 400-nm pulse creates free electrons in conduction band due to MP excitation, and second, 800-nm pulse creates additional number of free electrons due to II. However, no avalanche electrons in Al_2O_3 (and only small amount in SiO_2) were observed under 800-nm pulse.

Damage and ablation observed at double pulse excitation (400 + 800 nm) were interpreted as due to combined effects: MP ionization by 400-nm pulse plus heating of photoexcited electrons by 800-nm pulse without avalanche multiplication. So, the authors concluded that II is not a dominant damage mechanism at femtosecond-laser excitation.

9 Summary and Conclusions

Summarizing, we can make the following conclusions on fundamental mechanisms of LID, as understood today.

Absorbing inclusions in real optical materials play a significant role in LID. The most effective damage mechanism, associated with inclusions, is the TE due to nonlinear heating of inclusions and thermal UV-photoionization of the surrounding host material. Adequacy of the TE mechanism is well confirmed, in particular, by pulse-width dependence of the damage threshold in a wide range of laser pulse duration.

The II, as one of the intrinsic LID mechanism, was theoretically studied most comprehensively. Its dominant role in LID at long laser pulse irradiation (mostly in nanosecond-duration range) of many pure dielectrics was well confirmed experimentally. Its role in LID at ultra-short laser pulse irradiation (in femtosecond-duration range) is still under intensive theoretical and experimental investigation., Regarding effectiveness of this mechanism at ultra- short pulses (<100 fs), there are disagreements in predictions of different theoretical models.

The general theory of photoionization in transparent solids is well developed. A relative contribution of photoionization and II has been comprehensively studied both theoretically and experimentally. However, there are significant disagreements in theoretical predictions and experimental data regarding this contribution at femtosecond-pulse irradiation, which requires further studies to elucidate the problem.

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