# Laser-induced damage in solids

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The current state of studies of laser-induced damage (l.i.d.) in dielectrics and semiconductors in the wavelength range from the infrared to the ultraviolet, and pulse lengths in the range $10^{-3}$–$10^{-12}$ s, is discussed. Experimental methods used in l.i.d. studies are presented. The influence of nonlinear effects, such as self-focusing, self-defocusing, and stimulated scattering, is also discussed. Principal l.i.d. mechanisms are examined, including the heating of absorbing inclusions and defects (extrinsic mechanism) and collisional and multiphoton ionization (intrinsic processes). Statistical aspects of l.i.d. due to the probabilistic nature of the entry of absorbing defects into the interaction region and the creation of seed electrons are analyzed. The nature of the accumulation effect in l.i.d. under multiple-pulse illumination is examined. Experimental results are reported on l.i.d. in wide-gap dielectrics by infrared, visible, and ultraviolet radiation, and in semiconductors by infrared radiation. Absorbing defects play a dominant part in the damage produced in most real optical materials. Data confirming the avalanche l.i.d. mechanism in high-purity crystals are reproduced.

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## 1. INTRODUCTION

The phenomenon of optical breakdown in transparent solids was discovered in 1964 (Refs. 1 and 2) soon after the advent of the giant-pulse laser. A very large number of investigations has since been carried out in different materials (wide-gap dielectric crystals, glasses, polymers, and semiconductors). Interest in this subject has been stimulated above all by the practical needs of laser technology, since breakdown leads to irreversible damage in optical components of lasers and, essentially, is one of the basic physical factors restricting the ultimate laser output power.

It is therefore obvious that the elucidation of the factors governing laser induced damage is an important problem in the physics of interaction between high-intensity electromagnetic radiation and matter.

The aim of this paper is to present the current state of l.i.d. research with emphasis on physical aspects of the subject. Because of lack of space, we shall not attempt to provide an exhaustive review of all papers in this field (this would be practically impossible), but will try to present the main lines of research and advances made so far in the understanding of the mechanisms responsible for laser induced damage. For the same reason, theoretical and experimental results will be reviewed in a compressed form. Readers requiring more detailed information are referred to original papers. We note that a number of review type publications has already appeared. They contain extensive information on the individual aspects of the l.i.d. problem. A series of brief reviews (see Ref. 3 and the references therein) of papers presented at symposia has been published, covering optical materials for powerful lasers.[4] There is a review of papers on l.i.d. in thin film optical coatings,[5] and also a review of experimental work.[6]

Ever since l.i.d. has come under scrutiny, a relatively large number of possible mechanisms and models of l.i.d. in different transparent optical materials has been under discussion in the literature. This discussion has covered effects such as light pressure, electrostriction, hypersound generation during stimulated Mandel'shtam-Brillouin scattering (SMBS), collisional (electron avalanche) and multiphoton ionization, and various types of thermal effect induced in optical materials by absorbing inclusions and defects (thermoelastic stresses, thermal ionization, photoionization by radiation emitted by heated inclusions, thermochemical and mechanochemical reactions, and so on). These thermal effects give rise to the so-called extrinsic breakdown, since its characteristics are largely determined by various types of defect and inclusion in the material, in contrast to the processes mentioned in the beginning of this section, which are characteristic of the host material itself and give rise to the

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so-called intrinsic breakdown.

It has been known for considerable time[7] that absorbing inclusions (metallic platinum particles) contribute to damage produced in laser glass. Subsequent studies showed that absorbing defects and inclusions, which are always present to some extent in optical materials, are usually the dominant source of surface and bulk l.i.d. in optical components made from different materials, and reduce (sometimes very substantially) the threshold intensity at which damage begins.

There is particular interest in intrinsic mechanisms governing the ultimate radiation stability of optical materials exposed to laser radiation. Research has shown that the most probable of the above intrinsic l.i.d. mechanisms is collisional and multiphoton ionization,[1)] the efficiency of the former (electron avalanche) being more reliably confirmed by experimental l.i.d. studies in very pure alkali halide crystals.[8-10]

An important aspect of the l.i.d. problem is the probabilistic character of both the extrinsic and intrinsic damage mechanisms. In the former case, this is due to the random spatial distribution of absorbing defects with different characteristics (and therefore producing different thermal breakdown thresholds), whereas in the second case it is due to the probabilistic character of the creation of nucleating electrons that induce the ionization avalanche. One of the characteristic features of the l.i.d. process, namely, the dependence of the damage threshold on the size of the interaction region (the so-called size effect) is closely related to this probabilistic behavior.

It follows that studies of statistical properties (such as spatial variations in the damage threshold, the size effect, and damage probability as a function of laser intensity) are of considerable interest both from the fundamental point of view and for practical reasons, since they provide information on the mechanisms responsible for l.i.d. and, at the same time, provide data for estimates of the laser stability of optical components (for example, in studies of the reliability of laser devices for given laser intensity and laser beam diameter).

The cumulative effect plays an important part in laser induced damage. Thus, macrodamage on the surface or within the body of an optical material is produced as a result of the action of a series of laser pulses with intensities below the single-pulse damage threshold.

Research has shown that the cumulative effect is very well defined in certain materials, especially in polymers.[14] It is observed even for intensities that are lower by two orders of magnitude than the single-pulse damage threshold. Hence it is clear that studies of the nature of the cumulative effect are of great importance for the understanding of the mechanisms responsible for the interaction between laser radiation and various optical materials, both from the practical point of view, in estimating the life of optical components of lasers, and in searches for effective methods of suppressing this effect which tends to restrict the life of these components.

Self-interaction effects such as self-focusing, self-defocusing and stimulated scattering in the medium under investigation can play an essential part in l.i.d. processes. The first of these effects gives rise to a strong reduction in the axial intensity of a beam of radiation passing through the medium and is responsible for damage to many optical materials, including laser glass and crystals. It has been investigated in some detail in the literature. In this case, the damage threshold is determined not by the intrinsic optical stability of the material, but by the self-focusing threshold (see for example Ref. 15).

On the contrary, self-defocusing and stimulated scattering give rise to a reduction in the intensity in the medium and prevent the onset of damage.[13,16]

Hence it is clear that self-focusing, self-defocusing, and stimulated scattering must be taken into account in l.i.d. studies if threshold intensities are to be correctly estimated and an adequate interpretation of the mechanisms of damage is to be achieved.

We note one further aspect of l.i.d. that is of considerable importance, namely, it is essential to perform experiments under carefully controlled and reproducible conditions. This means that, among other things, it is essential to use lasers with specified spatial, temporal, and spectral characteristics, the purity of samples under investigation must be carefully analyzed, and so on.

Reliable conclusions must always be based on carefully designed experiments capable of producing results that can be adequately analyzed both qualitatively and quantitatively (in terms of well-founded theoretical models).

Research performed in this way has led in recent years to considerable advances in the understanding of laser induced damage in transparent solids, and to the development of various materials and components for optical technology, which are highly stable under exposure to powerful laser radiation.

## 2. EXPERIMENTAL METHODS AND CONDITIONS FOR l.i.d. STUDIES

The threshold laser intensities for which laser induced damage is observed in transparent solids are very different for different materials and even different samples of the same material. They depend on the frequency $\Omega$ and pulse length $\tau_p$ as well as on the linear dimensions $r$ of the illuminated region. Certain definite conditions must be satisfied in any correct determination of these threshold intensities $I_d$ and of the basic l.i.d. properties (the dependence of $I_d$ on $\Omega$, $\tau_p$, $r$, the sample temperature $T_0$, the statistical properties, and so on) with a view to elucidating the physical basis of damage and of identifying effective ways of increasing the laser stability of optical materials and components. The most important of these conditions are the following: the lasers employed must have stable frequency, temporal, and spatial characteristics, these characteristics must be monitored in the course of the experiment, self-interaction effects such as self-focusing, self-defocusing, and stimulated scattering as well as the cumulative effect must be taken into account, sample parameters must be monitored, and adequate methods must be used to record the damage.

The source of radiation commonly used in l.i.d. experiments is the pulsed laser operating at a fixed fundamental

frequency or its harmonics produced by frequency conversion in nonlinear crystals. The most frequently used lasers in these experiments are the ruby laser ($\lambda = 0.69\ \mu m$), the yttrium aluminum garnet laser activated with $Nd^{3+}$ ions ($Nd^{3+}$:YAG, $\lambda = 1.06\ \mu m$), lasers using $Nd^{3+}$ activated glass ($\lambda = 1.06\ \mu m$), and $CO_2$, HF, and DF gas lasers ($\lambda = 10.6\ \mu m$, $2.7\ \mu m$, and $3.8\ \mu m$, respectively). The second, third, and fourth harmonics of the $Nd^{3+}$:YAG laser are used ($\lambda_2 = 0.53\ \mu m$, $\lambda_3 = 0.35\ \mu m$, and $\lambda_4 = 0.27\ \mu m$). A number of experiments have been carried out using $Er^{3+}$:$CaF_2$ ($\lambda = 2.76\ \mu m$) and $Er^{3+}$:YAG ($\lambda = 2.94\ \mu m$) lasers. The laser pulse length can be varied in a wide range, depending on the particular experimental conditions: $\tau_p = 10^{-2}$–$10^{-4}$ s in free generation, $\tau_p = 10^{-7}$–$10^{-9}$ s under $Q$-switching, and $\tau_p = 10^{-10}$–$10^{-12}$ s under mode-locking conditions.

It is very important to use lasers with given and controllable temporal and spatial beam parameters if correct measurements of laser intensity are to be performed, and reproducible and comparable l.i.d. data are to be obtained. These conditions are well satisfied by single transverse and longitudinal mode lasers with a Gaussian lateral intensity distribution:

$$I(x,\ y) = I_0 \exp\left(-\frac{x^2+y^2}{a^2}\right), \tag{1}$$

where $I_0$ is the maximum intensity on the beam axis ($x = 0$, $y = 0$), $x$, $y$ are the lateral coordinates, and $a$ is a typical lateral beam dimension.

The temporal parameters of laser radiation in the millisecond and nanosecond pulse length ranges are relatively simply and reliably analyzed by the usual oscillographic methods, whereas the analysis of picosecond pulses requires more complicated methods, including high-resolution electrooptic cameras.[17] In the case of nanosecond pulses generated by $Q$-switched lasers, the pulse shape is usually Gaussian,[18] whereas pulses produced under free-running conditions (millisecond range) and under mode-locked conditions (picosecond range) may have a different shape. In particular, pulses generated by free-running lasers have a complicated spike structure. Since the amplitudes of the individual spikes are randomly distributed, such pulses are unsuitable for l.i.d. studies. Special methods for generating structure-free laser pulses must then be employed. Such pulses can be produced by lasers incorporating cavities bounded by flat mirrors and spherical lenses.[19]

It is important in l.i.d. experiments to be able to vary the pulse length $\tau_p$ continuously in as wide a range as possible without varying the spatial intensity distribution or the spectrum. This type of variation of $\tau_p$ is most simply produced by using an external electrooptic shutter to cut out a portion of a long enough laser pulse, followed by amplification in a laser amplifier. This method was used in Ref. 19 where ruby-laser pulses of nearly rectangular shape (width of rising and falling edges $\Delta t = 30$–$40$ ns) and smooth variation of pulse length in the range $\tau_p = 3\times10^{-7}$–$4\times10^{-4}$ s were produced.

This method of varying the pulse length is difficult to use for shorter pulses because the electrooptic shutter must then produce high-voltage control pulses with short rising and trailing edges.

A different method of producing nanosecond laser pulses (1–10 ns) whose length could be varied continuously was proposed in Ref. 20. It is based on time-variable extraction of radiation from a $Q$-switched cavity.

A spatially uniform laser intensity distribution can be produced by selecting one of the transverse cavity modes (the lowest $TEM_{00}$ mode) and using spatial filtration of the output beam. The latter is particularly important whenever multistage laser amplifiers are employed in which the active medium is optically inhomogeneous (usually a laser crystal).

Transverse-mode selection is achieved by placing a pin hole (1–2 mm diameter) in the laser cavity, whilst spatial filtration at the output of the amplifying system is performed by passing the beam through a set of pin holes[20] or confocal spherical lenses with a pin hole placed at the common focus (the pin hole diameter is of the order of the diffraction diameter of the beam in the focal plane).[21,22]

Longitudinal-mode selection is achieved in lasers by inserting additional Fabry-Perot etalons into the laser cavity. These are usually in the form of a stack of plane parallel plates (see, for example, Ref. 23). The selection of a single longitudinal mode is important not only from the point of view of producing a single-frequency generation spectrum, but also in order to exclude possible power fluctuations due to mode beating.

Various methods are available for measuring the radiation intensity and its spatial distribution in the laser beam (both collimated and focused). Methods based on the measurement of the fraction of energy transmitted when a flat screen or wire is partially inserted into the beam, and then examining the size of the burn on a photographic plate for different incident radiation power levels,[24,25] are relatively simple and reliable.

When a flat screen (usually a razor blade) is inserted into the beam, the transmitted power

$$F(x) = \int_{-\infty}^{x}\int_{-\infty}^{+\infty} I(x,\ y)\,dx\,dy \tag{2}$$

for a Gaussian intensity distribution $I(x, y)$ over the beam cross section is given by

$$F(x) \approx P_0\left(\frac{1}{2} + \frac{3a^2x - x^3}{3\pi^{1/2}}\right), \tag{3}$$

where $P_0 = \pi a^2 I_0$ is the total beam power.

By measuring $F(x)$ for different positions of the screen, it is possible to determine the true distribution $I(x, y)$ and to establish relatively simply the characteristic beam radius $a$.

It is common in l.i.d. experiments to use an external lens to focus the laser radiation on the surface or in the interior of the sample. This results in high enough intensity to produce damage at the focus of the lens for relatively moderate laser output power levels, and to separate surface from volume damage effects. When samples with a flat entrance surface are used, the aberration of the beam by this surface must

then be taken into account when volume damage is being investigated. For samples with low refractive index (most transparent dielectrics such as glasses, polymers, and ionic crystals), this aberration effect produces only a slight distortion of the beam in the focal region. However, in the case of media with a high refractive index (for example, semiconductors such as germanium), aberrations due to the flat surface produce a considerable distortion of the intensity distribution in the focal region in the interior of the sample. They can be excluded by giving the surface a spherical shape.

Apart from the above aberration effect, which is linear, i.e., it does not depend on the laser-beam intensity, it is also important in l.i.d. studies to take into account possible distortion of the spatial intensity distribution due to nonlinear effects in the specimen (self-focusing, self-defocusing, stimulated scattering, and multiphoton absorption). Although the influence of these effects on the l.i.d. process has frequently been analyzed (see Refs. 13, 15, and 16, and the references therein), quantitative allowance for them in measurements of damage thresholds is in most cases practically impossible because of the absence of adequate theoretical models as well as the lack of information on the mechanisms responsible for these effects and on the necessary material constants (nonlinearity coefficients in the refractive index, and so on). The common procedure in l.i.d. research is therefore to resort to qualitative estimates of these nonlinear effects and to procedures designed to minimize their influence on the measured degree of damage.

For example, the frequently observed filamentary structure of damage is usually due to self-focusing and is explained by the motion of the nonlinear focus in the medium if the laser pulse is bell shaped.[26]

However, point damage can also be observed in the presence of self-focusing if the laser pulse is rectangular in shape[27] which is explained in the multifocus model of self-focusing[28] by a standing pattern of nonlinear foci. Short focal length lenses are used to exclude the influence of self-focusing in l.i.d. experiments. This is justified[29] by the fact that, in this case, the intensity in the focus that is necessary for damage is reached when the input power $P_i$ is less than the critical self-focusing power $P_s$. For the Kerr mechanism of nonlinearity in the refractive index, which is usually the dominant factor in many transparent solid dielectrics is the case of short optical radiation pulses, it can be shown that[30]

$$P_s = \frac{0.942\, c\lambda^2}{8\pi^2 n_2}; \tag{4}$$

where $c$ is the velocity of light, $\lambda$ is the wavelength, and $n_2$ is the nonlinearity coefficient in the refractive index.

At the initial stage of self-focusing (before the catastrophic collapse of the beam and the appearance of the multifocus structure), the intensity at the focus of the lens in the nonlinear medium is given by[29,30]

$$I \approx I_L \left(1 + \frac{P_1}{P_s}\right); \tag{5}$$

where $I_L$ is the intensity in the linear focus of cross section $S_L$

The formula given by (5) has frequently been used (see, for example, Refs. 25, 29, 30, 31, and 32) to determine $P_s$ from the measured threshold damage power $P_i$ for different focal lengths $f$ (i.e., different $S_L$). It is assumed in this procedure that the threshold damage intensity $I_d$ is independent of the size of the region of interaction (focal spot). However, experiment shows that, in most transparent optical materials, the threshold intensity $I_d$ is size dependent even in the absence of self-focusing. As noted above, this can be traced to spatial fluctuations in the damage threshold, which are due to the presence of absorbing defects that initiate damage. Because of this, and also because of the uncertainty about the dominant self-focusing mechanisms and the possible influence of other self-interaction effects, the above procedure for determining $P_s$ turns out to be incorrect.

In contrast to self-focusing which facilitates breakdown in the medium, self-defocusing due to, say, the generation of nonequilibrium carriers by some particular mechanism (multiphoton and collisional ionization, etc.) should impede breakdown. Theoretical analysis[16] does actually show that, in a medium with a negative nonlinearity, self-defocusing produces a broadening of the beam and an intensity saturation in the focal region of the focusing lens (Fig. 1).

The maximum saturation intensity is determined by the focusing angle:

$$I_{\max} \approx \frac{5r_0}{\varepsilon_2}\left(\frac{a_0}{f}\right)^2; \tag{6}$$

where $\varepsilon_2$ is the coefficient of nonlinearity in the permittivity of the medium ($\varepsilon = \varepsilon_0 + \varepsilon_2|E|^2$) and $a_0$ is the radius of the Gaussian beam at the focusing lens of focal length $f$. This is accompanied by a substantial distortion of the radial and axial intensity distributions: the radial distribution becomes highly non-Gaussian, and acquires a flat top, whereas the axial distribution is also broadened, but its maximum shifts beyond the geometric focus.

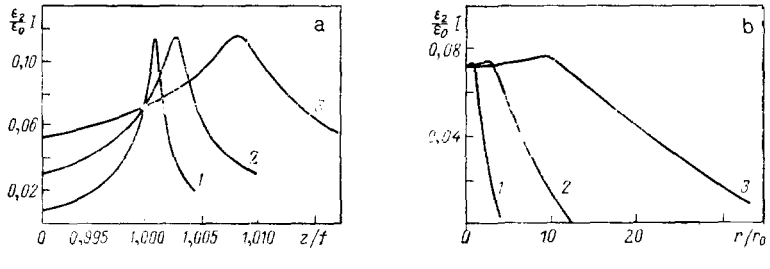


FIG. 1. Axial, $r = 0$ (a), and radial, $z = f$ (b), distributions of laser-beam intensity in the focal region of the lens in a medium with a negative nonlinearity of the refractive index, due to the generation of nonequilibrium carriers, plotted for different intensities of the incident Gaussian beam: $I_1$ (MW/cm$^2$) = 0.3(*1*), 3(*2*), and 30(*3*). The intensity $I$, the distance $z$ along the beam axis, and the distance $r$ along the radius are normalized, respectively, to the nonlinearity parameter of the medium $|\varepsilon_2/\varepsilon_0|$ [$\varepsilon_0$ is the linear part of the permittivity $\varepsilon = \varepsilon_0 + \varepsilon_2 I$ and $\varepsilon_2 < 0$ is the nonlinearity coefficient], the focal length $f$ of the lens, and the linear size $r_0$ of the focal spot in the linear medium. The calculations were performed for germanium, assuming $f = 1.5$ cm and $r_0 = 3\mu$m (Ref. 16).

The theoretically predicted intensity saturation in the focal region, due to self-defocusing, explains the absence of damage in germanium crystals when pulsed $CO_2$-laser radiation (pulse length 100 ns) is focused in these crystals. This occurs even for very high incident powers for which the intensity in the linear focus is $I_L \simeq 10^{12}$ W/cm$^2$ (Ref. 16) for which damage does definitely occur in other semiconductors and dielectrics. Self-defocusing of $CO_2$—laser radiation in germanium has been explained[16] in terms of the generation of nonequilibrium carriers. The maximum carrier concentration and maximum intensity at the focus of the lens with $a_0/f = 0.13$ were found to be $N_{e,\max} \simeq 10^{18}$ cm$^{-3}$ and $I_{\max} \simeq 3 \times 10^8$ W/cm$^2$, respectively.

The above results illustrate the extent to which self-defocusing effects are important in studies of laser damage. However, as in the case of self-focusing, it is difficult to perform a quantitative estimate of the influence of self-defocusing on the intensity distribution because the mechanism responsible for this phenomenon in the media under investigation is uncertain.

Experimental methods for investigating self-defocusing are not well developed. In a recent paper,[33] the role of self-defocusing was investigated by a two-beam method in which the mutual effect of two closely-spaced beams on the damage threshold was studied.

Stimulated scattering can also substantially modify the intensity distribution in the focal region of the focusing lens. It has been found[13] that, when l.i.d. in glass is investigated, back scattering due to SMBS must be taken into account because it can substantially reduce the intensity in the focus. This effect is very dependent on the spectral width $\Delta\nu$ of the laser radiation and the focal length $f$ of the focusing lens: it is a maximum for small $\Delta\nu$ and large $f$.

SMBS and self-focusing effects have been found to compete in laser damage processes at particular values of $\Delta\nu$ and $f$: for large $f$ and small $\Delta\nu$, SMBS is the dominant process in many glasses and prevents damage even for high input power levels, well above the critical self-defocusing power level. These results can be explained qualitatively in terms of existing theoretical ideas, although quantitative estimates of the damage threshold for given $\Delta\nu$ and $f$ can only be made on the basis of theoretical calculations of the propagation of a laser beam in the medium, including both effects, which is obviously a very complicated problem. It has been found[13] that the influence of SMBS on l.i.d. can be excluded by using a lens with a short focal length.

A variety of methods is being used in l.i.d. studies to detect damage. They are described in detail in the literature (see, for example, Refs. 4, 14, 34, and 35). The simplest method involves the observation of the emission of visible radiation (spark) and the scattering of the probing He-Ne laser radiation ($\lambda = 0.63$ $\mu$m), which occur immediately after the threshold intensity of the damaging radiation has been reached. As a rule, the damage threshold can be established clearly and accurately in this way. The morphology of the resulting damage is usually examined by optical microscopy, including aftereffects (residual damage) and dynamic observations (during the damage process).[34,35] Quite useful information on l.i.d. processes is obtained by studying the scattering of the incident (damaging) radiation[35] (including measurements of the intensity, polarization, and the spatial distribution) and the characteristics of visible and ultraviolet emission (intensity, spectrum, and emission kinetics). The latter methods can be used to investigate processes occurring in the medium when it is addressed by a powerful laser beam, both below and above the damage threshold. In particular, these methods can be used to investigate in detail the damage dynamics, to establish the presence of subthreshold changes in the material, and to follow the process of the accumulation of this damage that eventually results in visible macrodamage (see Refs. 35 and 14, and the references therein).

Preliminary studies of the characteristics of particular samples are of considerable value in the elucidation of the l.i.d. mechanisms in optical materials. This involves above all the estimation of the degree of optical purity of the sample, i.e., the presence of impurities, inclusions, and defects, which may have a considerable effect on the damage process.

A relatively simple test of the optical purity of materials is the scattering of light by foreign inclusions and defects (Rayleigh scattering).

Danileĭko *et al*[34] have suggested that a comparison between Rayleigh scattering and Mandel'shtam-Brillouin scattering in the same medium, or in some standard medium, could be used as a quantitative criterion for optical purity. This comparison should obviate difficult absolute measurements of the scattered intensity. We note that such tests would detect the presence of inclusions and defects with very small linear dimensions (10–100 nm), which are always present in optical materials to some extent and are shown by experiment to influence laser damage. Large inclusions, on the other hand, which play the most undesirable part in laser damage, can readily be detected by simpler methods as well. It has been shown[34] that there is a definite correlation between the Rayleigh scattered intensity and volume l.i.d. in a number of media (crystalline quartz, sapphire, and glass).

Light scattering can also be used to estimate the optical quality of the surface of optical components because different methods of polishing (abrasive, ion, laser, and so on) give rise to different surface topologies which effect laser stability.

It is also important to note that laser scattering of low-intensity light does not always yield unambiguous information about the optical quality and the correlation with laser stability because the latter is sensitive to highly absorbing defects whereas scattering is determined by all defects. There is therefore considerable interst in studies of the scattering of light at high (but nondamaging) incident intensities.

Attempts have been made[36] to use high-intensity nonlinear-scattering data to estimate the absorption coefficients of scattering defects, which determine the l.i.d. threshold. However, further studies will be necessary to elucidate reliably whether or not the absorption coefficients and other defect parameters can actually be determined from nonlinear-scattering data.

In view of the exceptional practical importance of predictions of laser stability of optical materials, urgent searches are being undertaken for other nondestructive methods of determining the nature of defects.

There is considerable interest in the development of different methods of measuring low absorption (including laser calorimetry,[37-39] photoacoustics,[40] and modulation spectroscopy[41]), which could be used to determine the absorption coefficient of defects. So far, these methods have been successful only in measuring the volume or area averages of the absorption coefficient ($\overline{\alpha}$), whereas the absorption properties of *localized* defects ($\alpha_{\text{loc}}$) are necessary in studies of pulsed laser damage. Typical values of $\overline{\alpha}$ for sufficiently pure optical materials, such as those used in laser optics, turn out to be very low ($\overline{\alpha} = 10^{-3}$–$10^{-5}$ cm$^{-1}$) and cannot give rise to heating sufficient for damage in the case of pulses of length $\tau_p \lesssim 10^{-7}$ s. In l.i.d. processes, the local temperature at individual points in the interaction region may be $T \gtrsim 10^3$ K, which requires $\alpha_{\text{loc}} \gtrsim 100$ cm$^{-1}$ (see below).

## 3. LASER-INDUCED DAMAGE DUE TO THE HEATING OF ABSORBING DEFECTS

Transparent solids can contain a variety of defects, namely, foreign phases (metal, semiconductor, ceramic, etc.), impurity and bulk clusters, particles of abrasive materials, and surface adsorbed materials. All these defects can have substantial absorption coefficients, and give rise to local heating and, hence, to damage of the material in the neighborhood of the defect. Possible processes in the host material surrounding the absorbing defect include melting, thermoelastic stresses that damage the material through mechanochemical reactions, thermochemical reactions, photoionization by radiation emitted by the heated defect, and the formation of plasma. The relative contribution of a particular process to l.i.d. will of course depend both on the parameters of the defect and on the physicochemical and mechanical parameters of the host medium.

### 3.1. Models of thermal damage

The first theoretical model of thermal damage by absorbing defects was examined in Ref. 42 in connection with the analysis of l.i.d. on the surface of ruby crystals. A solution of the heat conduction equation was used with this model to examine laser heating of absorbing defects with mean linear dimensions $q$ and mean separation $l$ in the surface layer of a crystal, taking into account heat transfer to the ambient medium.

An approximate solution of the equation of heat conduction was obtained on the assumption that the coefficient of absorption $\alpha$ of a defect and the thermal parameters of the defect and medium (thermal conductivity, etc.) were independent of temperature (although it was pointed out that the temperature dependence of these parameters could be allowed for).

The damage criterion was taken to be the attainment by the defect region of some critical temperature $T_c$ for which irreversible surface changes took place (melting, cracking, and so on). The following simple expressions were obtained for the threshold power:

$$P_{\text{d}} \approx (T_c - T_0)\left[\frac{\alpha V}{4\pi k q} + \frac{\alpha V}{l^2}\left(\frac{\tau_p}{c_0\rho_0 k_0}\right)^{1/2}\right], \quad \tau_p > \tau_{\text{x}}, \qquad (7)$$

$$P_{\text{d}} \approx \frac{c_0\rho_0 (T_c - T_0)}{\alpha \tau_p}, \quad \tau_p < \tau_{\text{x}}, \qquad (8)$$

where $\tau_x = c\rho q^2/k$ is the average characteristic time for the propagation of heat within the absorbing centers. $c$ is the specific heat, $\rho$ is the density, $k$ is the thermal conductivity of the medium forming the center (the subscript 0 labels the host crystal), $V$ is the volume of the absorbing center, and $T_0$ is the initial temperature of the crystal.

These formulas provide a qualitative explanation of the observed function $P_d(\tau_p)$ and also the variation of $P_d$ with the surface treatment (dependence of $P_d$ on the defect parameters $\alpha$ and $q$; Fig. 2). We note that the results reported here apply to volume damage as well.

A similar approach to the analysis of thermal damage by absorbing inclusions was developed in Refs. 43 and 44 in relation to volume damage in glass containing metal or ceramic particles. An analysis was made of the heating of a particle with allowance for heat transfer to the ambient host medium and the onset of thermoelastic stresses within it. As in Ref. 42, it was assumed that the thermophysical parameters of the particles and medium were temperature dependent.

It was found[44] that for laser energy flux densities of 20 J/cm$^2$ and pulse lengths of 30 ns, the temperature of the absorbing particle could reach $10^4$ K, and this produced stresses in the ambient glass that exceeded its limiting strength. It was also found that there was a characteristic particle size for which the maximum temperature was reached for given radiation intensity. This means that, within the framework of this particular model, there should be a "most hazardous size" of the absorbing inclusions, which corresponds to the minimum threshold for laser induced damage.

This model has frequently been discussed (see Ref. 45 and the references therein) in connection with the volume and surface laser l.i.d. in different materials containing inclusions of various kinds (metal, ceramic, etc.). It was found[45] to provide a good explanation of experimental data on the damage threshold of thin-film dielectric coatings as a function of pulse length and film thickness.

Although reported results show that the above thermal damage models[42-44] provide a qualitative explanation of some important trends in l.i.d., they are not of course com-

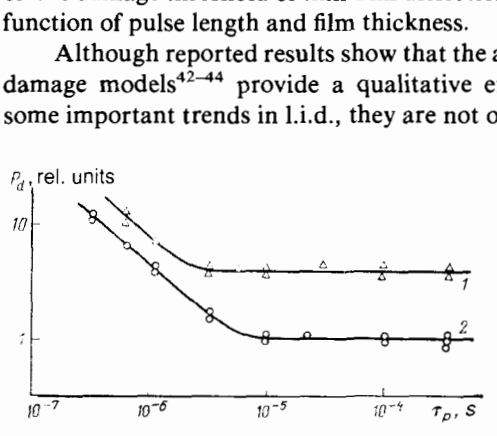


FIG. 2. Threshold power for surface damage as a function of the laser pulse length for two typical ruby samples.[42]

pletely satisfactory because they do not take into account the temperature dependence of the thermophysical parameters of particles and medium. Actually, parameters such as the coefficient of optical absorption of inclusions, the specific heat, and the thermal conductivity of an inclusion and of the ambient host can hardly be regarded as temperature independent at about $10^4$ K. Moreover, it is important to note that the critical temperatures and stresses remain undefined in these models because of the dynamic nature of the damage process (especially in the case of short laser pulses) and, hence, the threshold criterion for laser induced damage is also undetermined.

An analysis of the thermal mechanism of laser induced damage, taking into account the above functions for both absorbing inclusions and the ambient medium, was carried out in Refs. 46 and 47. The heating of the absorbing particle was described by the heat conduction equation

$$\frac{\partial}{\partial t}(c\rho) = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 k \frac{\partial T}{\partial r}\right) + Q(I, T), \tag{9}$$

where $c$, $\rho$, $k$ are respectively the specific heat, the density, and the thermal conductivity (which are different for the inclusion and the ambient medium, and are functions of temperature) and $Q(I, T)$ is the heat source function of the particle, which depends on the laser intensity $I$ and temperature $T$.

As an example, the functions $Q(T)$ and $k(T)$ were taken in the following well-known form which there are approximations that hold for a number of dielectrics and metals at temperatures in excess of 300 K:

$$Q(I, T) = Q(I)\exp\left(\xi\frac{T-T_0}{T_0}\right), \qquad k(T) = \frac{d}{T}, \tag{10}$$

where $T_0$ is the initial temperature of the sample, $\xi$ is a parameter representing the temperature nonlinearity of the absorption coefficient of the inclusion, and $d$ is a constant for the material of the inclusion or the host medium.

It was shown by solving (9) that the heating of inclusions is highly nonlinear (in fact explosive) in character. The asymptotically rapid increase in the temperature of the inclusion was then used to determine the damage threshold:

$$Q_{th} = \frac{2d}{a^2\beta}\frac{\ln\xi}{\xi-1}, \tag{11}$$

where $a$ is the particle size,

$$\beta = \frac{1}{3}\left(1 + 2\frac{d_1}{d_2}\right),$$

and the subscripts 1 and 2 label the particle and the ambient medium, respectively. This expression is valid for rectangular laser pulses of length $\tau_p \gg \tau_x$ where $\tau_x = \tau_0 c_1\rho_1 a^2/d_1$ is the characteristic time for the propagation of heat in the particle.

For short laser pulses ($\tau_p \ll \tau_x$), the solution of (9) was found to yield a different expression, namely,

$$Q_{th}(I) = \frac{c_1\rho_1 T_0}{\xi\tau_p}\left\{1 - \exp\left[-\xi\left(\frac{T_c}{T_0} - 1\right)\right]\right\}, \tag{12}$$

where $T_c$ is the critical temperature for damage.

Since $T_c \gg T_0$, we find that $Q_{th}$ is practically independent of $T_c$ provided the nonlinearity parameter $\xi$ is not too small (measurements show[47] that for metals $\xi \simeq 0.1$).

The threshold intensity $I_d$ of laser radiation that produces damage in the material can be determined from the relation

$$Q_{th} = \frac{\sigma I_d}{V}, \tag{13}$$

where $\sigma$ is the absorption cross section of a particle of volume $V$.

The absorption mechanism must be known for specific estimates of $I_d$. For semiconducting or metal inclusions with linear (single photon) absorption at the laser frequency, the cross section is given by the following well-known expressions[48] ($a \ll \lambda$):

$$\sigma = \frac{12\pi\Omega a^3\varepsilon''}{c\left|(\varepsilon'+2)^2 + \varepsilon''^2\right|} \tag{14}$$

for the semiconducting particle,

$$\sigma = \frac{12\pi\Omega a^3\varepsilon''}{c}\left(\frac{1}{|\varepsilon|^2} + \frac{\Omega^2 a^2}{90c^2}\right) \tag{15}$$

for the metal particle with low conductivity, and

$$\sigma = 6\pi a^2(\varepsilon')^{-1/2} \tag{16}$$

for the metal particle with high conductivity. In these expressions, $\Omega$ is the frequency of the absorbed radiation and $\varepsilon = \varepsilon' + i\varepsilon''$ is the permittivity of the medium.

When the above formulas are used for the ruby crystal and for glass containing metal inclusions (nickel and platinum), the estimated values of $I_d$ are found to be in good agreement with observations.[47]

It is clear from the foregoing that, when the above model of nonlinear thermal damage in media with absorbing inclusions is used to determine the damage threshold, the specific critical temperatures and stresses in the ambient medium need not be known, and the damage threshold can be determined from the condition of a rapid increase in the temperature of the inclusion (thermal explosion).

However, a particular amount of energy must, of course, be released if irreversible changes, which can be looked upon as macrodamage, are to take place in the ambient medium (melting, crack formation, and so on). An additional mechanism for higher absorption outside the limits of the inclusion is necessary for this condition to be satisfied even for very small inclusions ($a \sim 10^{-6}$ cm).

It is shown in Ref. 49 that photoionization of the host medium by radiation from the laser-heated inclusion is an effective mechanism of this kind. Simple estimates based on the assumption that the small particle behaves like a black body[48] do actually show that the temperature of the particle heated by radiation of frequency $\Omega$ and intensity $I_d$ is given by

$$T = k^{-1}\left(\frac{\hbar^4\pi^2 c^2\Omega I_d}{25}\right)^{1/5} \tag{17}$$

so that, when $\Omega = 2.73\times10^{15}$ s$^{-1}$ (ruby laser frequency) and $I_d = 10^{10}$ W/cm$^2$, the temperature is $T = 7\times10^4$ K.

At this temperature, the maximum in the spectral distribution of black-body radiation emitted by the particle lies at $\Omega_m = (3.92/\hbar)kT = 3.9\times10^{16}$ s$^{-1}$ ($\lambda_m \simeq 500$ Å) and the

power radiated at this frequency is $P_m = 4\pi c\sigma(\Omega_m)e_0(\Omega_m) \simeq 3\times 10^{-2}$ W [$\sigma(\Omega_m)$ is the cross section and $e_0(\Omega_m)$ is the spectral density of the radiation]. This ultraviolet radiation is absorbed in the adjacent layer of the dielectric (linear dimensions $l \sim 2\times 10^{-5}$cm), thereby effectively generating free carriers at the rate $dn/dt \simeq 10^{30}\text{s}^{-1}\,\text{cm}^{-3}$ which in turn is the source of absorption. The heating of this plasma layer is qualitatively similar to the heating of the original absorbing inclusion, and leads to the photoionization of the next layers of the dielectric. Calculations show that the propagation of this photoionization wave is quite rapid (the characteristic time for a thermal explosion in the ionized layer under the above conditions is $\sim 5\times 10^{-11}$ s; Ref. 49), and should lead to macrodamage.

Other mechanisms of additional absorption by the medium surrounding the inclusion have been discussed in the literature and include thermal ionization,[50,51] thermochemical reactions producing absorbing products (Ref.52 and the references therein), and mechanochemical reactions producing excited particles.[53]

In the thermal ionization mechanism in which absorption results in the appearance of equilibrium free carriers in the conduction band of the solid heated by the absorbing inclusion, the rate of propagation of absorption waves and the onset of thermal instability (damage threshold condition) are governed by the thermal conduction mechanism. It has been shown[54] that the electronic component of thermal conductivity plays the dominant part in laser breakdown.

It is clear that the thermal ionization mechanism in the above "semiconductor model"[50,51] can be effective only in l.i.d. in narrow-gap materials because this mechanism is relatively unimportant in wide-gap materials, even at high temperatures (close to the melting point).

There is a number of thermochemical processes that take place when a powerful laser beam interacts with a solid. They include decomposition of complex compounds (semiconductors, multicomponent inorganic gases, and polymers), formation of the oxides of atomic semiconductors such as germanium and silicon, and so on. The relative importance of these processes in l.i.d. in transparent media containing absorbing inclusions is determined by many factors[52] including, above all, the absorption coefficients of reaction products at the laser frequency, their temperature dependence, the rate of the corresponding chemical reactions, the diffusion coefficient of the decomposition products, and so on.

The part played by thermochemical processes in these phenomena has been extensively discussed in connection with l.i.d. in polymers. It seems that it is now well established that the thermochemical mechanism, involving pyrolysis and the formation of highly-absorbing soot particles, is responsible for l.i.d. in polymethylmethacrylate (PMMA) under illumination by continuous laser radiation.[55,56] As far as damage by short (nanosecond and picosecond) pulses of radiation is concerned, the thermochemical mechanism leading to the formation of soot particles does not appear to be significant.[53] This is so because the pyrolysis reaction requires a relatively long period of time (the corresponding rate constant is $\nu = 0.1$–$0.02\ \text{s}^{-1}$ and the activation energy is[56] $E \simeq 3000$ K).

The mechanochemical mechanism has also been proposed[53] for the observed laser induced damage in polymers, including the cumulative effect. The essence of this mechanism can be summarized as follows. The heating of the absorbing defects is accompanied by the appearance of thermoelastic stresses in the host medium, which lead to chemical bond breaking and the formation of "hot" (vibrationally excited) radicals which absorb the laser radiation by undergoing transitions between excited vibrational sublevels of the ground and higher-lying electronic states. The rate constant for these mechanochemical reactions, which produce hot radicals, is very dependent on the pulsed ($\sigma$) and residual ($\sigma_s$) thermoelastic stresses[53]

$$k_{\text{mch}} = k_0 \exp\left[-\frac{E}{A\tau_0(1+\delta\sigma_s)\sigma}\right], \tag{18}$$

where $E$ is the activation energy, $A$ and $\delta$ are parameters that depend on the thermophysical characteristics and structure of the polymer, and $\tau_0$ is the vibrational relaxation time of particular chemical bonds in macromolecules (the so-called active oscillators), the breaking of which results in the appearance of the hot radicals. Thermoelastic stresses are found to increase during the laser heating of absorbing defects which, by virtue of the function $k_{\text{mch}}(\sigma)$, leads to an increase in the concentration of hot radicals and, consequently, to an increase in the absorption coefficient. This essentially implies the appearance of a mechanochemical absorption wave which is responsible for the loss of thermal stability in the host medium, and damage sets in. When the intensity of the incident laser radiation is insufficient to ensure that this instability will develop in the course of a single pulse, the instability may appear after a *series* of successive pulses because of the accumulation of residual stresses.

We note that, in contrast to the thermochemical mechanism which involves the accumulation of absorbing products (such as soot), the particles, i.e., the hot radicals, appear in the above mechanochemical process only at the time of the laser pulse and do not accumulate from pulse to pulse.

The mechanochemical mechanism can be used as a basis for an explanation of the principal observed features of laser induced damage in polymers, namely, the dependence of the damage thresholds under single-shot ($I_1$) and multiple-shot ($I_N$) illumination on the viscoelastic properties of the host and the concentration of low-molecular impurities introduced into it. The viscoelastic properties are characterized by the tensile strength of the polymer and influence directly the rate of formation of hot radicals in the mechanochemical reaction [in accordance with (18)]. As far as the low-molecular impurities are concerned, they play a twofold role: on the one hand, they influence the viscoelastic properties (plastification effect) and, on the other, they influence the vibrational relaxation time $\tau_0$ of active oscillators (AO) as a result of their interaction with the impurity molecules, which in turn affects the rate of formation of hot radicals [see (18)].

Figures 3 and 4 show experimental data on the l.i.d. threshold of polymethylmethacrylate, and illustrates the

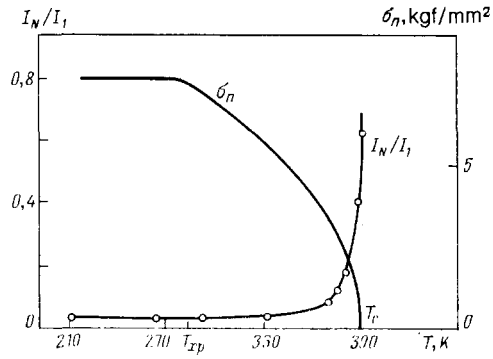


FIG. 3. Threshold for $N$-pulse damage $I_N$ ($N = 200$) and the strength $\sigma_s$ of polymethylmethacrylate as functions of temperature[53]: $I_1$—single-pulse threshold, $T_b$ and $T_g$—brittle and glass transition temperatures.

above features, namely, the effect of viscoelastic properties and of low-molecular impurities.[53] Figure 3 shows that there is a clear correlation between the temperature dependence of the threshold for $N$-pulse damage ($I_N$) and the viscoelastic parameter ($\sigma_p$), whereas Fig. 4 shows the selectivity of the effect of low-molecular impurities on $I_N$.

The above brief details of the mechanochemical mechanism of laser induced damage has been used as a basis for synthesizing polymers with high stability to laser radiation, comparable with the stability of other optical materials such as inorganic glasses and crystals. High-strength components for polymer optics for laser applications have been developed in this way (they include passive $Q$-modulators, active components for dye lasers, and so on[57–59]).

### 3.2. Statistical properties of laser induced damage

The dependence of the damage threshold on the parameters of defects (the coefficient of absorption, its temperature dependence, and the linear dimensions of defects) ensures that l.i.d. is statistical in character and is governed by the spatial distribution of the defects within the region of interaction and the probability that one or other defect will enter this region. Even a qualitative analysis of these effects

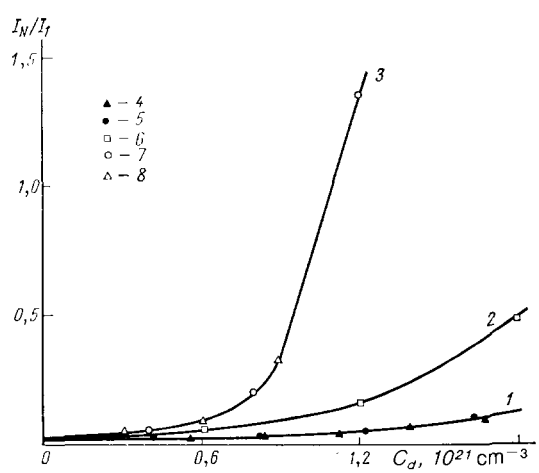


FIG. 4. $N$-pulse damage threshold $I_N$ ($N = 200$) in polymethylmethacrylate as a function of the concentration $C_d$ of the additive: 4-cyclohexanol, 5-butyronitrile, 6-ethanol, 7-butanol, 8-hexanol.

will show that the l.i.d. threshold $I_d$ must depend on the linear dimension $r$ of the interaction region in which the radiation intensity is $I$, and the damage probability $P$ for given laser-beam dimensions must depend on intensity.

Quantitative relationships for these effects have been examined in the relatively extensive literature devoted to this problem (see Refs. 60–64 and the references therein). The aim of all this research was to explain the properties of the functions $I_d(r)$ and $P(I)$ which are of interest not only for elucidating the damage mechanism, but also as a way of determining the parameters of the defects (the concentration and the $I_d$ distribution), which can be extracted by comparing theory with experiment.

The basic results of these investigations are summarized below.

In the simplest statistical l.i.d. model, examined in Ref. 60, it is assumed that the sample under investigation contains absorbing defects of a particular species, and all the defects have the same damage threshold $I_1$. The damage probability $P$ is then the probability that a defect will enter the volume $V_{\text{eff}}$ in which the intensity exceeds $I_1$, and can be written in the form

$$P = 1 - \exp(-\rho V_{\text{eff}}), \tag{19}$$

where $\rho$ is the mean concentration of defects.

The evaluation of $V_{\text{eff}}$ in the region of the caustic of the focused Gaussian beam with spatial intensity distribution

$$I(r, z) = I_L \left(1 + \frac{z}{z_R}\right)^{-1/2} \exp\left\{-\frac{2r^2}{\omega_0^2[1+(z/z_R)]}\right\}, \tag{20}$$

leads to the following expression:

$$P = 1 - \exp\left\{-\rho \frac{(\pi\omega_0^2)^2}{\lambda}\left[\frac{2}{9}(\gamma-1)^{1/2}(\gamma+5) - \frac{4}{3}\operatorname{arctg}(\gamma-1)^{1/2}\right]\right\}, \tag{21}$$

where $\omega_0$ is the minimum beam size in the caustic, $z_R = \pi\omega_0^2/\lambda$ is the Rayleigh length, and $\gamma = I_L/I_1 \gg 1$.

If we take $I_d = I_L$ at $P = 0.5$ and examine the function $I_d(\omega_0)$, we can use the above formula to find the two characteristic parameters of the absorbing defects, namely, $I_1$ and $\rho$. If the sample contains more than one species of defect, and the damage thresholds are different, the statistical description of the laser induced damage is formulated in a similar way except that instead of the mean concentration of defects of a particular species we have a distribution of defects over the thresholds.[61,63,64] The damage probability as a function of intensity is then given by

$$P(I) = 1 - \exp\left[-\int_0^I V(I, x)\, c(x)\, dx\right], \tag{22}$$

where $c(x)$ is the concentration of inclusions in the medium, which initiate the breakdown for radiation intensity $x$, $V(I,x)$ is the volume of the region containing the caustic, in which the intensity exceeds $x$, and $I$ is the intensity at the center of the caustic. Since $c(x)$ is unknown, it is interesting to consider whether it can be found from experimental data on the breakdown probability $P(I)$. The procedure for find-

ing $c(x)$ involves the evaluation of the integral equation (22), which gives[64]

$$c(x)=\frac{2}{\pi}\frac{\mathrm{d}}{\mathrm{d}x}\left[x^{5/2}\frac{\mathrm{d}}{\mathrm{d}x}\int_0^x\frac{\Phi'(x)}{I(x-I)^{1/2}}\,\mathrm{d}I\right], \tag{23}$$

where

$$\Phi(I)=\frac{1}{2\pi\rho_0 z_0}\ln[1-P(I,\ c(x))]^{-1},$$

and $\rho_0$, $z_0$ are the parameters of the caustic of the focused Gaussian beam.

This so-called inversion problem in l.i.d. statistics has been examined in a number of papers (see Refs. 63 and 64 and the references therein). It was shown that the solution given in (23) is unstable against small changes in $P(I)$ and, consequently, the inverse problem is one of a class of incorrectly posed problems.[63,64]

A number of methods can be used[63,64] to obtain a suitable solution and, consequently, physically correct distributions $c(x)$, from the inversion formula (23). In particular, it was shown in Ref. 64 that this requires that the measured function $P(I)$ must satisfy the conditions of smoothness.

Determinations of $c(x)$ from specific l.i.d. data are of considerable interest both from the point of view of l.i.d. mechanisms in different optical materials and from the point of view of improved fabrication technology for optical materials.

### 3.3. Cumulative effect

One of the most important problems in l.i.d. physics is the nature of the cumulative effect. This effect has now been found in different materials (silicate glasses, alkali halide crystals, ferroelectric crystals and polymers; see Ref. 14 and the references therein). It is particularly well-defined in polymers where it is observed below the damage threshold for between 1 and 100 pulses.

All the available experimental data on l.i.d. indicate that the cumulative effect is due to irreversible changes in the host medium around the initiating absorbing defect which accumulate from pulse to pulse. These irreversible changes can arise as a result of photochemical, thermochemical, and mechanochemical reactions, and different phase transitions.

All these processes were included in the analysis[14] of experimental data on l.i.d. in different materials. However, reliable data on the presence of any particular mechanism are still few and far between, and further specialized studies will be necessary to elucidate the relative importance of these mechanisms in specific materials and under particular experimental conditions (e.g., dependence on wavelength and laser pulse length). All that can be said at present is that the most extensively investigated question is that of the nature of the cumulative effect in polymers[14,53] in which the formation of hot radicals in mechanochemical reactions is the most likely reason for it (under illumination by short pulses). These reactions occur as a result of thermoelastic stresses that accumulate between pulses in the vicinity of absorbing defects.

Experiments show that l.i.d. under multiple-shot illumination, in which a well-defined cumulative effect occurs, is statistical in character just as in the case of single-pulse damage.

In view of this, it has been suggested[14] that the cumulative effect can be described statistically by analogy with single-pulse damage. This description involves the following basic concepts that adequately characterize the accumulation process: the critical number $\overline{N}_c(I)$ of pulses, averaged over a large number of irradiated points, which is necessary for macrodamage in the specimen for given intensity $I$; the threshold intensity $I_N$ for damage after the $N$th pulse, for which damage occurs after a given number $N$ of pulses with given probability $P_N(I)$; and the distribution function $f(I_N)$ for the damage thresholds. The quantities $P_N(I)$ and $f(I_N)$ are related by a formula analogous to (22):

$$P_N(I)=1-\exp\left[-\int_A \mathrm{d}A\int_0^I f(I_N)\,\mathrm{d}I_N\right], \tag{24}$$

where $A$ is the irradiated surface area of the sample.

This relation can be used to find the size dependence of $I_N$ or to determine the distribution function $f(I_N)$ by analogy with the above analysis of single-pulse damage.

Because of the close similarity between processes producing damage under single and multiple shot irradiation, the l.i.d. characteristics in these two cases may be expected to be related. To identify this relationship, we must know the specific mechanism responsible for the cumulative effect, which gives rise to the evolution of the initiating absorbing defects in the course of the successive laser pulses [and, hence, the evolution of the function $f(I_1)\rightarrow f(I_N)$]. A consistent statistical theory of the cumulative effect which takes into account (24) and the evolution of $f(I_1)$ has not yet been developed. Moreover, we note that experimental data on l.i.d. in polymers[14] definitely indicate that the distribution function $f(I_1)$ evolves in the course of multi-shot damage (a transformation of the size and statistical dependence of the damage thresholds was observed).

We note in conclusion of this section that all the mechanisms discussed above and all the features of volume l.i.d. due to absorbing defects will in general apply to l.i.d. of surface and transparent thin-film coatings on optical components. The small quantitative and functional differences between volume and surface l.i.d. thresholds may be due to the characteristics of defects and the particular laser-beam intensity distribution in the two cases. Thus, for surfaces and for thin-films, adsorbed films of absorbing oxides or other materials (for example water) play an important part in addition to the point defects present in surface films. Multilayer thin-film coatings (dielectric mirrors and antireflective coatings) are more defective which, naturally, produces a lowering of l.i.d. thresholds as compared with materials in bulk (a more detailed discussion of l.i.d. in thin-films will be found in Ref. 5). The laser-beam intensity distribution on the surface of a solid component may be very different from the distribution within its volume because of effects associated with the reflection and interference of waves at the boundary between optical media with different refractive indices. In particular, this is responsible for the difference between

the l.i.d. thresholds of the front and back surfaces of optical components and multilayer half-wave and quarter-wave coatings (cf., for example, Refs. 65 and 66).

## 4. INTRINSIC l.i.d. MECHANISMS DUE TO COLLISIONAL AND MULTIPHOTON IONIZATION

Ionization by electron impact has been extensively discussed in the literature (cf., Refs. 67–77 and the references therein), since it is one of the most probable l.i.d. mechanisms in ultrapure transparent solids. A consistent theory of this mechanism has now been developed[2)] in detail[70–73,75,76] and describes l.i.d. in a wide range of frequencies (from dc to the ultraviolet) and for various pulse lengths ($10^{-7}$–$10^{-11}$ s). The predictions of this theory have been used as a basis for specific experimental studies[7–10] of l.i.d. in different dielectric crystals, and it has been shown that the electron avalanche mechanism of l.i.d. does in fact occur in ultrapure samples.

Multiphoton ionization as a l.i.d. mechanism has been studied to a lesser extent. There are only Refs. 76 and 78–81 in which this question is analyzed, and it is shown that, under certain definite conditions (especially at high laser frequencies) multiphoton ionization can be the dominant l.i.d. mechanism.

We shall now present the basic results of theoretical and experimental studies reported in the papers cited above on the relative importance of collisional and multiphoton ionization in l.i.d.

### 4.1. Collisional ionization

It has been shown[70–73] that the most rigorous analysis of the avalanche breakdown in transparent dielectrics involves the solution of the quantum-mechanical transport equation for conduction electrons, which describes the multiplication of electrons by collisional ionization. This equation is

$$\frac{\partial f(P,t)}{\partial t}=\frac{2\pi}{\hbar}\sum_{q}^{r}B(q)\sum_{n=-\infty}^{\infty}J_n^2\left(\frac{eEq\tau(p)}{\hbar m\Omega\,[1+\Omega^2\tau^2(p)]^{1/2}}\right)$$
$$\times\{[f(p+q)(N_q+1)-f(p)N_q]$$
$$\times\delta(\varepsilon(p+q)-\varepsilon(p)-\hbar\omega_q-n\hbar\Omega)$$
$$+[f(p+q)N_q-f(p)(N_q+1)]$$
$$\times\delta(\varepsilon(p+q)-\varepsilon(p)+\hbar\omega_q-n\hbar\Omega)\},\qquad(25)$$

where $e$ and $m$ are, respectively, the electron charge and mass, $p(\varepsilon)$ is the momentum of an electron of energy $\varepsilon$, $B(q)$ is the matrix element of the electron-phonon interaction, $N_q$ is the number of phonons with wave vector $q$, $\hbar\omega_q$ is the phonon energy, and $\tau(\varepsilon)$ is the relaxation time of the longitudinal component of the momentum of an electron of energy $\varepsilon$ in the absence of the field. The criterion for breakdown is found as a result of the simultaneous solution of the equations for the time-dependent electron density $n$ and lattice temperature $T$:

$$\frac{dn}{dt}=\gamma n,\quad \frac{dT}{dt}=\beta nT^{3/2};\qquad(26)$$

where $\beta$ is a coefficient that depends on the parameters of the electron-phonon interaction, the ionization potential, and other lattice parameters, and $\gamma$ is the avalanche development constant determined by solving (25):

$$f(x,t)=e^{\gamma t}f(x).\qquad(27)$$

The development of an avalanche has been investigated in detail in the literature,[70,71,73,75,76] taking into account the scattering of electrons by acoustic and optical phonons. Different lattice temperatures $T_0$, electromagnetic-field frequencies $\Omega$ [related to the effective electron-phonon collision frequency $\nu_{\text{eff}}$ ($\Omega/\nu_{\text{eff}}\ll 1$, $\Omega/\nu_{\text{eff}}\gtrsim 1$)], and ionization potentials $I$ ($\hbar\Omega/I\ll 1$, $\hbar\Omega/I\lesssim 1$) were considered. By finding the avalanche development constant $\gamma$ as a function of the field $E$, and by determining the breakdown criterion

$$\gamma\tau_p=\ln\left(1+\frac{2\gamma}{\beta n_0}\right),\qquad(28)$$

from (26), where $\tau_p$ is the pulse length and $n_0$ the initial electron density, formulas were obtained for the critical breakdown field $E$. These formulas can be used to calculate both the field strength and its dependence on radiation parameters ($\Omega$ and $\tau_p$) and lattice parameters (in particular, the crystal temperature $T_0$). These functional relationships are particularly valuable when experimental data are compared with theory because they lead to specific conclusions on the l.i.d. mechanism operating under specific experimental conditions.

We now reproduce some of these formulas and functional relationships.

When acoustic phonons [high-temperature region, $kT>\frac{1}{2}v_s(2mI)^{1/2}$ where $v_s$ is the velocity of sound] are scattered in the so-called diffusion approximation to the quantum-mechanical transport equation, i.e., when $\hbar\Omega/I\lesssim 0.2$, the critical breakdown field is given by [71,73]

$$E_c^2=\frac{Im^2v_s^2}{2kTe^2}\left(\Omega^2+\frac{I}{ml_{ac}^2}\right),\qquad(29)$$

where $l_{ac}$ is the electron mean free path.

In the case of scattering by zero-point lattice oscillations [low-temperature region, $kT<\frac{1}{2}v_s(2mI)^{1/2}$] the critical field in the same approximation is given by

$$E_c^2=\frac{mv_s(2mI)^{1/2}}{2e^2}\left(\Omega^2+\frac{2I}{5ml_0^2}\right).\qquad(30)$$

Equations (29) and (30) lead to a very characteristic frequency and temperature dependence of the laser breakdown threshold due to avalanche ionization. Since the mean free path $l_{ac}$ is inversely proportion to temperature, the temperature dependence of $E_c^2$ is very different for $\Omega>\nu_{\text{eff}}$ and $\Omega<\nu_{\text{eff}}$: in the former case $E_c^2\sim T^{-1}$ whereas in the latter case $E_c^2\sim T$. For $\hbar\Omega/I>0.2$, when the diffusion approximation is not valid and the difference-differential transport equation[8,76] must be solved, the frequency dependence of $E_c$ turns out to be different from (29) or (30), and its nature is essentially determined by the radiation pulse length. Figure 5 shows the calculated dependence of the critical field on the radiation frequency, obtained from the difference-differential transport equation, for two values of the avalanche constant $\gamma_0$ corresponding to pulses in the nanosecond and picosecond ranges of $\tau_p$ [by virtue of the breakdown criterion

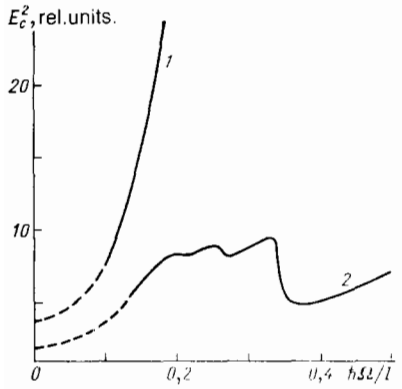


FIG. 5. Square of the critical field as a function of $\hbar\Omega/I$ for different avalanche constants[76]: 1—$\gamma_0 = 0.02$ (picosecond pulses, $\tau_p = 30$ ps), 2—$\gamma_0 = 1^{-5}$ (nanosecond pulses, $\tau_p = 100$ ns).

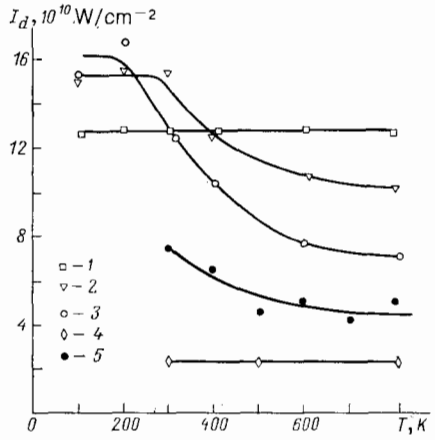


FIG. 6. Threshold intensity $I_d$ in NaCl crystal as a function of temperature for different wavelengths $\lambda(\mu m) = 0.53(1)$, $0.69(2)$, $1.06(3)$, $10.6(4$, Ref. 8), and $2.76(5$, Ref. 9).

$\gamma\tau_p = \gamma_0 Q(1)\tau_p = L \simeq$ const, $Q(1)$ being the characteristic rate of electron-phonon process $\sim 10^{12}$–$10^{13}$ s$^{-1}$]. It is clear that for nanosecond pulses for which $\hbar\Omega/I > 0.3$, the frequency dependence of the damage threshold is appreciably reduced and may even fall with increasing frequency, showing an oscillatory dependence. Analysis[8,76] of the solution of the difference-differential transport equation has shown that the temperature dependence of the critical field in this case remains the same as in the diffusion approximation: $E_c^2 \sim T^{-1}$ when $\Omega > \nu_{\text{eff}}$.

The theoretically predicted characteristic frequency and temperature dependence of the critical field for the avalanche breakdown mechanism have been used[8,9] in an experimental verification of the validity of this mechanism of laser induced damage in alkali halide crystals.[8,9] The experiments were performed with a large number of NaCL, KCl, KBr, LiF, NaF, etc., crystals in a wide range of laser frequencies [$CO_2$ laser ($\lambda = 10.6$ $\mu$m), $Nd^{3+}$:YAG laser ($\lambda = 1.06\,\mu$m and second harmonic $\lambda = 0.53\,\mu$m), ruby laser ($\lambda = 0.69$ $\mu$m), $Er^{3+}$:$CaF_2$ laser ($\lambda = 2.76$ $\mu$m)] and temperatures in the range 100–900 K.

Considerable variations were found in the damage thresholds. There were also variations in the temperature dependence of the thresholds between different specimens for each type of crystal. This indicates that impurities and defects influence the l.i.d. process, but highly radiation-stable specimens of NaCl, KCl, and KBr were found to have reproducible frequency and temperature dependence of $I_d$, which at some radiation frequencies was in agreement with the theoretical predictions for the avalanche mechanism of breakdown. As an example, Fig. 6 shows these results for one of these NaCl specimens. Analysis of these experimental data has shown that the frequency dependence in the wavelength range 10.6–0.69 $\mu$m is in adequate agreement with the above theoretical dependence for the electron avalanche mechanism if the effective electron-phonon collision frequency is $\nu_{\text{eff}} = 6\times 10^{14}$ s$^{-1}$. The temperature dependence of the l.i.d. threshold observed at $\lambda = 1.06$ and 0.69 $\mu$m is also satisfactorily explained in terms of the avalanche mechanism. However this theory does not explain the observed function $I_d(T)$ for $\lambda = 10.6$, 2.76, and 0.53 $\mu$m).

This discrepancy has led to the proposal that for long-wavelength radiation (10.6 and 2.76 $\mu$m), the avalanche ionization process is delayed by a shortage of seed electrons[82] (the above theory of the avalanche mechanism of l.i.d. was constructed on the assumption that there was a sufficient density of initial electrons), which should be reflected both in the magnitude of the breakdown threshold and its frequency and temperature dependence. As far as the observed absence of a temperature dependence of the breakdown threshold at $\lambda = 0.53$ $\mu$m is concerned, this may be due to the other l.i.d. mechanism, namely, multiphoton ionization.

In view of the above anomalies in the temperature dependence of the breakdown threshold, a theoretical analysis has been carried out of the avalanche ionization of dielectrics by laser radiation in the situation where there is a shortage of seed electrons,[82,83] and the relative contributions of collisional and multiphoton ionization mechanisms to l.i.d. were examined.[76] We shall now briefly consider the results of this analysis.

#### *4.1.1. Delay of the avalanche by a lack of seed electrons*

It is generally accepted that seed electrons, which are necessary for the development of the electron avalanche due to collisional ionization, arise from the photoionization of impurities and defects, or the multiphoton ionization of the host atoms in the crystal lattice. If these processes give rise to an electron density $n_0$ within the interaction region that is high enough even during the initial stages of the laser pulse, so that $n_0 V > 1$, the critical breakdown field is determined by the rate of development of the electron avalanche which is a very slowly-varying function of $n_0$ (Refs. 70 and 73). In the opposite case, i.e., when $n_0 < 1$, the critical field is determined not so much by the rate of development of the avalanche as by the rate of production of seed electrons. In other words, the bottleneck in the l.i.d. process due to collisional ionization is in this case the creation of seed electrons. The l.i.d. process then becomes essentially statistical in character, i.e., the breakdown threshold is determined by the probability of appearance of a seed electron.

The basic statistical properties of breakdown can be established in this case by considering simultaneously the production of seed electrons and the evolution of the avalanche.

It is assumed that the former process is due to multiphoton ionization of host atoms in the lattice, impurity atoms, and defects. Then, assuming that the two processes are Poissonian, and that each of them is described by a probability of the form

$$p_k = \frac{t/\tau}{k!} \exp\left(-\frac{t}{\tau}\right), \tag{31}$$

it can be shown that the probability of appearance of $n$ electrons in the region of interaction with radiation as a result of the simultaneous effect of multiphoton and collisional ionization is given by[82]

$$S_n = \frac{\Gamma(n+(\tau/\tau_1))}{\Gamma(\tau/\tau_1)\,\Gamma(n+1)}\left[1-\exp\left(-\frac{t}{\tau}\right)^n \exp\left(-\frac{t}{\tau_1}\right)\right], \tag{32}$$

where $\tau$ and $\tau_1$ are the characteristic times of collisional and multiphoton ionization, respectively. The probability that breakdown will occur in a time $t$ as a result of the growth of the electron avalanche is given by

$$P = \sum_{k=n}^{\infty} S_n(t) = \frac{\Gamma(\alpha+n\exp(-t/\tau))}{\Gamma(\alpha)}, \tag{33}$$

where $\Gamma(\alpha, x)$ is the incomplete $\Gamma$-function and $\alpha = \tau/\tau_1$. If the relationships governing the dependence of $\tau$ and $\tau_1$ on the radiation intensity $I$ are known, the relation given by (33) enables us to calculate the breakdown threshold $I_d$ for given probability $P$. The function $\tau(I) \simeq \gamma^{-1}(I)$, where $\gamma$ is the avalanche development constant, is determined from the theory of the electron avalanche when the latter is not delayed by a lack of seed electrons:[76]

$$2\Delta \exp\left(-\frac{\beta}{2}\right) = \Delta \operatorname{ch}\frac{\Delta}{2} - \beta \operatorname{sh}\frac{\Delta}{2}, \tag{34}$$

where $\Delta = (\beta^2 + 4\gamma_0\beta)$, $\gamma_0 = \gamma/Q$, $Q$ is the characteristic rate of electron-phonon processes, $\beta = 1/QqS[1 + (v^2\theta^2/\Omega^2)]$, $\delta_0 = kT_0/\varepsilon_1$, $q = e^2E^2/6m^2v_s^2\Omega^2 \sim I$, $\theta = T/T_0$, $\varepsilon_1$ is the ionization potential, $v_s$ is the velocity of sound, $m$ and $e$ are the electron mass and charge, respectively, and $T_0$ is the initial temperature of the crystal.

The function $\tau_1(I)$ is determined by the well-known relation for multiphoton ionization

$$\frac{1}{\tau_1} = Aq^k, \tag{35}$$

where $A$ is a constant and $k$ is the number of photons (multiphoton ratio). Analysis of (33) shows[82] that the range $\alpha < 1$ corresponds to a considerable delay of the avalanche by the absence of seed electrons. The breakdown probability is then given by

$$P = 1 - \exp\left(-\frac{t}{\tau_1}\right), \tag{36}$$

i.e., as can be seen from (32), by the probability that at least one seed electron will appear as a result of multiphoton ionization during the presence of the pulse of radiation.

In the range $\alpha > 1$, the l.i.d. threshold corresponds to the usual avalanche breakdown mechanism and is determined as described above.

An important consequence of the above statistical analysis of avalanche breakdown is that the delay of the avalanche is accompanied not only by an increase in the breakdown threshold $I_d$, as compared with the usual threshold of undelayed avalanche, but also by a change in the characteristic dependence of $I_d$ on the temperature of the sample and the volume of the irradiated region. Actually, if the seed electrons are produced by multiphoton ionization, it follows from (35) and (36) that, in the case of undelayed avalanche, the threshold $I_d$ does not depend on temperature for any ratio of the frequency $\Omega$ of the electromagnetic radiation and the electron-phonon collision frequency $\nu_{\text{eff}}$.

This is very different from the temperature dependence in the case of the usual electron avalanche, as discussed above.[3)]

The dependence of $I_d$ on the volume of the interaction region in the case of the ordinary avalanche breakdown process is largely determined by spatial diffusion of electrons. The characteristic size of the interaction region for which the diffusion of electrons becomes important is given by[70]

$$d_k = 4\left(\frac{DG\tau_p}{E_c^2}\right)^{1/2}, \tag{37}$$

where $D$ is the coefficient of spatial diffusion, $\tau_p$ is the pulse length,

$$G = \begin{cases} \dfrac{6I}{kT}\left(\dfrac{mv_s\Omega}{e}\right)^2, & kT \gg v_s p(\varepsilon), \\ \dfrac{15(2mI)^{1/2}}{2mv_s}\left(\dfrac{mv_s\Omega}{e}\right)^2, & kT \ll v_s p(\varepsilon), \end{cases}$$

$I$ is the ionization potential, and $p(\varepsilon)$ is the quasimomentum of an electron of energy $\varepsilon$. All the other symbols have the same meaning as before. If the diameter of the laser beam in the sample under investigation is $d > d_c$, the avalanche threshold will not depend on $d$, whereas in the opposite case ($d < d_c$) it should increase with decreasing $d$, i.e., $E_c^2 \sim d^{-2}$. Estimates for wide-gap dielectrics such as NaCl crystals show that, when $\Omega \simeq 10^{15}\ \text{s}^{-1}$ and $\tau_p \simeq 10^{-8}\ s$, the quantity $d_s$ amounts to a few microns.

If the avalanche ionization process is delayed by a lack of seed electrons, the character of the size dependence of the breakdown threshold turns out to be essentially different: it is then determined by the probability that at least one seed electron will appear in the interaction region. The size dependence was analyzed in detail in Ref. 83, using a statistical approach similar to that employed in the statistical model of damage in absorbing defects. It was assumed in this analysis that the seed electrons were produced by ionization of impurities and defects, uniformly distributed in the sample with mean density $\rho_i$. The breakdown probability is then found to be

$$P = 1 - \exp\left[\sum_i \rho_i \int_V (e^{-t/\tau_i} - 1)\, dV\right], \tag{38}$$

where $\tau_i$ is the ionization time constant for impurity of type $i$ and $V$ is the volume of the interaction region in which the intensity $I(r, z)$ exceeds the critical value $I_A$ for the given breakdown. This expression has been analyzed to determine the nature of the size dependence in the following two limiting cases: (1) for readily ionizable impurities for which $\tau_p /$

$\tau_{iA} \gg 1$ [$\tau_{iA} = \tau_i(I_A)$] and (2) for impurities and defects that are difficult to ionize [$\tau_p/\tau_{im} \ll 1$, $\tau_{im} = \tau_i(I_m)$ where $I_m$ is the maximum intensity in the beam].

In the first of these two cases, it follows from (38) that the size dependence of the breakdown threshold is determined exclusively by the geometry of the beam (spatial distribution of intensity in the beam). For example, for the focused Gaussian beam, it is readily calculated from the relations

$$V\sum_i \rho_i = -\ln(1-P) \tag{39}$$

for given $P$ and $I_A$, since $V$ is a known function:

$$V = \frac{4}{3}\pi k b^4 \left[\frac{1}{3}\left(\frac{I_m}{I_A}+5\right)\left(\frac{I_m}{I_A}-1\right)^{1/2} - 2\arctan\left(\frac{I_m}{I_A}-1\right)^{1/2}\right], \tag{40}$$

where $k = 2\pi/\lambda$ and $b$ is the beam radius.

In the second case, the character of the size dependence is determined not only by the beam geometry, but also by the function $\tau_i(I)$ which depends on the mechanism responsible for the ionization of impurities. When this mechanism is multiphoton ionization, we can introduce the ionization rate $1/\tau = V_{\text{eff}}\Sigma_i \rho_i/\tau_{im}$ and show from (38) that the size dependence is determined by the effective volume

$$V_{\text{eff}} = \frac{4}{3}\pi k b^4 \int_0^{[(I_m/I_A)-1]^{1/2}} \xi^2 \frac{3+\xi^2}{(1+\xi^2)\varkappa - 1}\, d\xi, \tag{41}$$

where $\varkappa$ is the multiphoton ratio.

It follows from the foregoing results that by studying the size dependence of the breakdown threshold we can obtain information on the l.i.d. mechanism and, in particular, on the source of seed electrons. However, it must be remembered that, in some cases (for example, in the single-photon ionization of impurities or defects), the nature of the size dependence of the breakdown threshold in the case of delayed avalanche ionization may not be different from the corresponding quantity for breakdown by the usual thermal heating of absorbing defects by laser radiation (Section 3.2).

The idea that the avalanche ionization is delayed by a lack of seed electrons provides a qualitative explanation of observed anomalies in the temperature dependence of the l.i.d. threshold in alkali halide crystals at wavelengths of 10.6 and 2.76 $\mu$m (Fig. 6). Actually, the absence of a temperature dependence of the breakdown threshold $I_d$ at $\lambda = 10.6\,\mu$m and the descending form of the function $I_d(T)$ in the temperature range $T = 300$–$600$ K, which reaches a plateau for $T > 600$ K, can be explained in a consistent manner[82] in terms of the avalanche mechanism if we suppose that, for the 10-$\mu$m radiation, the avalanche is delayed throughout the above temperature range, whereas for $\lambda = 2.76\,\mu$m it begins to be delayed for $T > 600$ K. The latter effect is due to the temperature dependence of the parameter $\alpha$ of delayed avalanche theory (see above).

The avalanche delay effect was confirmed directly by the crossed-beam experiment described in Ref. 10 in which the presence of ultraviolet radiation was found to influence the l.i.d. threshold for $CO_2$ laser radiation ($\lambda = 10.6\,\mu$m) in NaCl. The source of the ultraviolet radiation, which produced the seed electrons in this experiment, was a nitrogen laser ($\lambda = 0.337\,\mu$m) locked to a $CO_2$ laser. The density of electrons excited by the nitrogen laser radiation, and determined from measured photoconductivity, was found to be $N_0 \simeq 5\times 10^9\ \text{cm}^{-3}$, which produced the necessary number of seed electrons $n_0 = N_0 V > 1$ within the volume containing the caustic of the short-focus lens (focal spot diameter $d = 23\,\mu$m) used to focus the $CO_2$ radiation.

The ultraviolet radiation produced a considerable reduction (by a factor of 6) in the l.i.d. threshold, down to $I_d = 5\times 10^{10}\ \text{W/cm}^2$. This clearly indicated that the avalanche breakdown was delayed until the ultraviolet radiation was introduced, and the intensity $I_d$ corresponded to the usual avalanche breakdown threshold.

A similar two-beam experiment has been performed[84] with NaCl in combined 1.6-$\mu$m ($Nd^{3+}$:YAG laser) and 0.266-$\mu$m (fourth harmonic of the same laser) radiation. It was found that the ultraviolet background did not affect the breakdown threshold at 0.266 $\mu$m. This shows that, for wavelengths $\lambda \lesssim 1\,\mu$m, avalanche ionization is not delayed by a lack of seed electrons (they are produced in sufficient quantity by this radiation itself without the auxiliary ultraviolet illumination). This conclusion is in agreement with the above (see Fig. 6) temperature dependence of the breakdown threshold at 1.06 $\mu$m, which suggests the ordinary avalanche breakdown mechanism for this case.

We note that the collisional ionization mechanism has frequently been used in the literature to explain experimental data on both volume and surface l.i.d. in wide-gap dielectrics in the infrared and visible ranges (see, for example, Refs. 69 and 85–89). However, reported conclusions about the relative importance of this mechanism were frequently based only on qualitative properties (correlation with breakdown thresholds in dc fields, threshold independent of radiation frequency in a wide range, damage morphology, and so on) and on comparisons with simple theoretical models. Analysis of the experimental data obtained in the above papers on l.i.d. in alkali-halide crystals and comparison with the data obtained later[7-8] for the same type of crystal (Fig. 7) have shown that the authors of Refs. 85–89 were probably dealing with samples that were not pure enough and the cor-

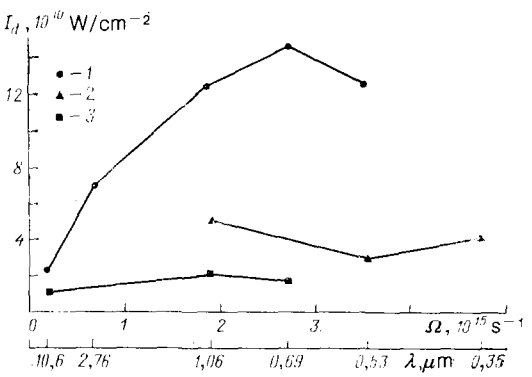


FIG. 7. Threshold intensity $I_d$ in NaCl as a function of radiation frequency at $T_0 = 300$ K: Refs. 8 and 9 (*1*), Ref. 89 (*2*), and Refs. 88 and 69 (*3*).

TABLE I. L.i.d. thresholds of transparent dielectrics at different wavelengths, GW/cm$^2$ (Ref. 92).

| Material | NaCl | KCl | KBr | CsJ | KJ | RbCl | CsBr | NaF | LiF | $SiO_2$ crystal | $SiO_2$ fused | $Al_2O_3$ | $CaF_2$ | KDP |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Wavelength $\mu$m: | | | | | | | | | | | | | | |
| 1,06 | 120 | 70 | 50 | 15 | 22 | 6 | 27 | 140 | 360 | 230 | 400 | 400 | 200 | — |
| 0,69 | 150 | 80 | 58 | 13 | 10 | 7 | 4 | 140 | 360 | 280 | 600 | 400 | — | — |
| 0,266 | 45 | 50 | 50 | 5,3 | 42 | 7 | 6 | 18 | 240 | 70 | 40 | 18 | 390 | 290 *) |

*Without taking into account induced optical inhomogeneity in the crystal.

responding l.i.d. in their samples was more likely to have been due to thermal breakdown on absorbing inclusions rather than avalanche ionization. This is indicated by the lower l.i.d. thresholds and the absence of a frequency dependence in the case of the samples investigated in Refs. 85–89, whilst the more stable samples investigated in Refs. 7–9 confirm a frequency dependence in the range 10.6–0.53 $\mu$m, as predicted by the theory of avalanche ionization.

#### *4.1.2. Ultraviolet* l.i.d. *in wide-gap dielectrics*

There is considerable interest in the possibility of an electron avalanche in ultraviolet l.i.d. The theory of avalanche ionization predicts, as described above, that the frequency dependence of the l.i.d. thresholds should exhibit some very specific features in this wavelength range. Moreover, photoionization of impurities and host atoms by multiphoton (in particular, two-photon) interband transitions may play a more important part in l.i.d. by ultraviolet radiation as compared with infrared and visible radiation. For example, the two-photon absorption coefficients measured at $\lambda = 0.266$ $\mu$m amount to $\beta \sim 1$ cm/GW (Ref. 90) for most alkali-halide crystals. These photoionization processes may have an important direct influence on l.i.d. through the absorption of radiation, as well as indirect influence through self-defocusing on the generated free carriers.

In addition to these processes, ultraviolet l.i.d. may also be influenced by different radiation defects produced under the influence of this radiation.[91]

Experimental data on l.i.d. at $\lambda = 0.266$ $\mu$m have been analyzed from this point of view for wide-gap transparent materials,[91] including the alkali-halides investigated in the infrared and visible ranges. It was found that, for most materials, the damage thresholds at $\lambda = 0.266$ $\mu$m were substantially lower than those at $\lambda = 1.06$ and 0.69 $\mu$m (Table I). This fact and the considerable variations in l.i.d. thresholds between different samples of the same material, as well as the influence of thermodynamic treatment on the optical stability of specimens, have led to the conclusion that, in most alkali-halide crystals such as NaCl, KCl, and so on, and for crystalline quartz, the l.i.d. thresholds at $\lambda = 0.266$ $\mu$m are determined by impurities and defects, even in optically very stable materials. Analysis of the frequency and temperature dependence of l.i.d. thresholds of fused quartz has shown that they are in agreement with the theory of avalanche ionization. For LiF and $CaF_2$ crystals, the damage thresholds at $\lambda = 0.266$ $\mu$m are close to the measured thresholds at $\lambda = 1.06$ and 0.69 $\mu$m (see Table I), which is not inconsistent with the theory of avalanche ionization for $\hbar\Omega/I = 0.3$–0.4 (see Fig. 5). However, studies of the temperature dependence of l.i.d. thresholds of these crystals at $\lambda = 0.266$ $\mu$m have shown[92] that this dependence is not in agreement with the theoretical prediction for the avalanche mechanism.

The above data thus show that ultraviolet l.i.d. in wide-gap dielectrics is usually due to the influence of impurities and defects, although damage by collisional ionization is possible in some cases.

#### *4.1.3.* L.i.d. *in semiconductors*

The electron avalanche mechanism has been discussed in analyses[93] of experimental data on infrared l.i.d. in semi-

TABLE II. Threshold intensity $I_d$ and effective field $E_{eff}$ in volume l.i.d. in Si and GaAs (Ref. 93).

| Material | Wavelength $\lambda$, $\mu$m | | | | | |
|---|---|---|---|---|---|---|
| | 10,6 | | 2,76 | | 2,94 | |
| | $I_d$, GW/cm$^2$ | $E_{eff}$, MB/см | $I_d$, GW/cm$^2$ | $E_{eff}$, MB/см | $I_d$, GW/cm$^2$ | $E_{eff}$, MB/см |
| Si | 5,0 | 0,75 | 2,3 | 0,51 | 0,15 | 0,13 |
| Partially compensated GaAs | 16,5 | 1,4 | 0,82 | 0,32 | 0,083 | 0,1 |
| GaAs: | | | | | | |
| Optical № 1 | 1,27 | 0,39 | — | — | — | — |
| » № 2 | 14,5 | 1,3 | — | — | — | — |
| » № 3 | 10,3 | 1,1 | — | — | — | — |
| » № 4 | 12,4 | 1,2 | — | — | — | — |

conductors (Si, GaAs; $\lambda = 10.6$, 2.94, and 2.76 $\mu$m). The measured l.i.d. thresholds $E_c$ (Table II) at 10.6 $\mu$m were found to be higher than the critical dc breakdown fields $E_c^0(\text{Si}) = 5\times10^5$ V/cm, $E_c^0(\text{GaAs}) = 3.7\times10^5$ V/cm, which meant that laser breakdown data at this wavelength could be interpreted as being due to avalanche ionization (although there was a spread in the threshold value between different specimens, indicating the influence of defects on l.i.d.). Comparison of $E_c$ with $E_c^0$ in the light of the frequency dependence (29) was used to estimate the effective electron-phonon collision frequency $\nu_{\text{eff}}$. The results were: $1.6\times10^{14}$ s$^{-1}$ for Si and $5\times10^{13}$ s$^{-1}$ for GaAs. These are reasonable values, but more detailed experimental studies (including the temperature dependence of the l.i.d. thresholds) will be necessary for a more reliable conclusion. As regards l.i.d. at $\lambda = 2.76$ and 2.94 $\mu$m, the nature of the frequency dependence of the threshold (its decrease with increasing wavelength) has been interpreted as being due to three-photon absorption. We shall examine this question in the next Section.

### 4.2. Multiphoton ionization

Multiphoton ionization was discussed in Section 4.1.1. as an auxiliary process responsile for supplying the seed electrons necessary for the development of collisional ionization. However, if the multiphoton ionization probability $W_n$ is high, the rate of generation of electrons in the conduction band by this process may exceed the corresponding collisional rate, so that the former process may dominate l.i.d.

The conditions necessary for this to happen have been examined theoretically in a number of papers.[76,78–81] Different methods were employed, but we shall confine our attention to Ref. 76 where breakdown criteria were obtained for the collisional and multiphoton ionization mechanisms, and were then used as a basis for analyzing the relative contributions of the two processes to l.i.d.

The generation of free carriers in the conduction band by collisional and multiphoton ionization is described by

$$\frac{dN}{dt} = \gamma N + W_n - R(N), \tag{42}$$

where $R(N)$ describes the recombination of carriers.

A physically satisfactory l.i.d. criterion can be obtained by simultaneously solving this equation and the equation describing the heating of the lattice by electron-phonon collisions

$$\frac{d\theta}{dt} = \beta\theta^{\varkappa}N, \tag{43}$$

where $\theta = T/T_0$, $T_0$ is the initial temperature of the lattice, $\beta$ and $\varkappa$ are given by

$$\beta = \frac{4Q(1)(qS_0)^{3/2}I}{\pi^{1/2}c\rho T_0}, \quad \varkappa = \frac{3}{2},$$

if scattering by acoustic phonons is the dominant process. Here, $S_0 = kT_0/I$, $q = e^2E^2/6m^2v_s^2\Omega^2$, $I$ is the ionization potential, $c$ is the specific heat of the lattice, and $\rho$ is the density.

To elucidate the relative importance of collisional and multiphoton ionization in l.i.d., we have to derive the above "temperature criterion" for both processes, i.e., we must perform a simultaneous solution of (43) and (42), retaining the dominant term in the latter.

This procedure yields the following breakdown criteria:[4)]

$$\gamma\tau_p = \ln\left(1 + \frac{2\gamma}{\beta N_0}\right), \tag{44}$$

$$4(1 - Q_c^{-1/2}) = \beta W_p \tau_p^2; \tag{45}$$

where $\tau_p$ is the pulse length, $N_0$ is the initial carrier density, and $\theta_c$ is the critical temperature of the lattice at which l.i.d. sets in.

Since $\gamma$ and $W_p$ are known functions of the field $E$ and of the electromagnetic field frequency [$\gamma(E,\Omega)$ is determined by solving the transport equation, as described above, and $W_p(E,\Omega)$ is found from the theory of multiphoton transitions], we can use (44) and (45) to determine the l.i.d. thresholds for collisional and multiphoton ionization.

For $W_p$ we can use the Keldysh formula[81]

$$W_p = \frac{2}{9\pi}\Omega\left(\frac{m\Omega}{\hbar}\right)^{3/2}\Phi\left(\left(2\left\langle\frac{\tilde{\Delta}}{\hbar\Omega}+1\right\rangle - \frac{2\tilde{\Delta}}{\hbar\Omega}\right)^{1/2}\right) \times \exp\left[2\left\langle\frac{\Delta}{\hbar\Omega}+1\right\rangle\left(1 - \frac{e^2E^2}{4m\Omega^2\Delta}\right)\right] \times \left(\frac{e^2E^2}{16m\Omega^2\Delta}\right)^{\langle(\tilde{\Delta}/\hbar\Omega)+1\rangle}, \tag{46}$$

$$\tilde{\Delta} = \Delta + \frac{e^2E^2}{4m\Omega^2}, \quad \Phi(z) = \int_0^z dy\exp(y^2 - z^2),$$

where $\Delta$ is the band gap.

The range of validity of this formula was discussed in Ref. 45 where it was concluded that it yields good results for multiphoton processes of different order (up to single-photon processes).

It follows from (44) and (45) that the dependence of l.i.d. thresholds on the pulse length is different for collisional and multiphoton ionization mechanisms. As an example, Fig. 8 shows NaCl calculations based on (44)–(46) with $n = 5$, together with the analysis[76] of the critical field of avalanche ionization as a function of frequency and pulse length. The figure illustrates the competition between colli-

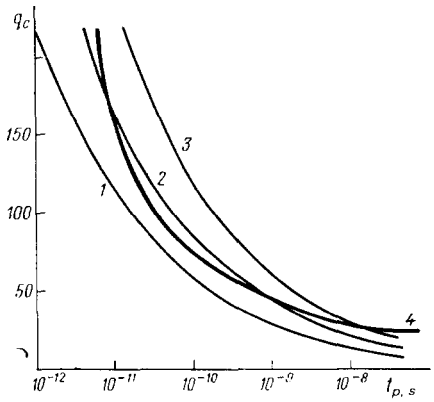


FIG. 8. The critical field parameter $q_c \sim E_c$ as a function of the laser pulse length $\tau_p$ (NaCl, $n = 5$) for different l.i.d. mechanisms: multiphoton ionization [*1–3*, $W_{(1)} > W_{(2)} > W_{(3)}$] and collisional ionization (*4*, Ref. 76).

sional and multiphoton ionization mechanisms, which depends both on $W_p$ and on the pulse length. Multiphoton ionization is the dominant breakdown process in a certain particular range of pulse lengths.

There have been no specific experimental studies of the validity of this theoretical prediction, although several attempts have been reported at an analysis of experimental data on l.i.d. in terms of the multiphoton ionization mechanism (see, for example, Refs. 45, 78–80, and 93).

In particular it has been shown[93] that the l.i.d. threshold of Si at $\lambda = 2.76$ $\mu$m is quite well described by three-photon ionization.

## 5. CONCLUSION

Our review shows that the basic l.i.d. mechanisms in transparent solids are now well understood, especially for nanosecond pulses.

The dominant part played by absorbing defects, which initiate damage in most real optical materials, has been demonstrated. Possible processes in the ambient host material, which participate in l.i.d. through thermal heating of absorbing defects, e.g., photoionization by the black body radiation from heated defects, the propagation of an ionization wave by means of electronic thermal conductivity, and mechanochemical processes, have now been more or less elucidated.

The statistical features of l.i.d. due to thermal breakdown on absorbing defects are now reasonably well understood. Basic features of the cumulative effect are also understood, but more detailed studies of this very important effect in l.i.d. under multiple irradiation are required.

The elucidation of the deleterious role of absorbing defects and, in some cases, of their nature, have led to effective methods for improving the laser stability of optical materials (removal of undesirable absorbing impurities from original materials, thermal treatment producing the decay of absorbing defects, suppression of mechanochemical reactions in polymers by special additives, development of new methods for polishing optical components, thus removing surface defects, and so on).

It has been shown that nonlinear effects such as self-focusing, self-defocusing, and stimulated Mandel'shtam-Brillouin scattering have been shown to have an important influence on the damage process.

It has been established that l.i.d. in ultrapure optical materials is due to collisional ionization. The decisive factor in establishing this fact has been provided by experimental studies of basic l.i.d. properties (frequency and temperature dependence of the threshold, and the influence of ultraviolet illumination on the threshold) as well as by comparison between the data obtained in this way and the rigorous theory of avalanche ionization.

As far as the other intrinsic breakdown mechanism is concerned, i.e., multiphoton ionization, there is only limited information at present on its relative contribution to l.i.d. A theory of this mechanism has been developed and can be used as a basis for specific experiments.

The advances achieved in the study of l.i.d. mechanisms have led to the development of different materials for laser optics (glass and crystal media for lasers, nonlinear frequency converters, electrooptic and passive modulators consisting of crystals and polymers, antireflective and reflective thin-film coatings, transmission-optics components, and so on).

There is also a number of unresolved questions in this field, which are of both fundamental and practical importance. Cumulative effects in l.i.d. in different optical materials, and l.i.d. mechanisms in the ultraviolet part of the spectrum, and for ultrashort pulses of radiation, have not been adequately studied. The last effect is particularly topical in connection with recent advances in the physics of ultrafast processes, involving the use of powerful picosecond and femtosecond lasers.

[1)]Other intrinsic mechanisms discussed in the literature involve light pressure, electrostriction, and hypersound generation in SMBS, and are not effective sources of damage.[11,12] Moreover, the last process (SMBS) will actually prevent damage because it leads to a reduction in the local intensity of the propagating laser beam, due to back scattering.

[2)]A brief review of this work is given in Ref. 17.

[3)]In the above analysis of the ordinary avalanche process, we did not take into account the recombination of electrons in the presence of the radiation pulse (i.e., we assumed that the recombination time $\tau_r$ was much greater than the pulse length $\tau_p$). In the reverse situation ($\tau_r < \tau_p$), the recombination of carriers leads to a substantial weakening of the dependence of $I_d$ on temperature.[76]

[4)]These criteria correspond to the situation in which the recombination of carriers in the presence of the pulse is neglected (i.e., it is assumed that $\tau_r \gg \tau_p$). Breakdown criteria, including recombination, are given in Ref. 76.


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