Influence of macroscopic cracks on optical strength of solid transparent dielectrics

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Impact of pulsed laser radiation on dielectrics with macroscopic cracks has been studied. Mechanisms of the crack influence on the dielectric optical and mechanical strength are determined. The dependence of the optical strength on the crack lifetime is discussed. It is shown that the effect of radiation of pre-threshold intensity on the sample with cracks can improve its mechanical and optical strength.

The condition of material near the crack surfaces depends on the lifetime of the crack, the environment, material characteristics, and other factors. The crack growth in a dielectric is accompanied by electron emission, plastic deformation, and by the occurrence of strain regions in the material.[1–3] The impact of a cracked layer on the optical strength of material was investigated in a series of papers.[4–9] However, many phenomena were studied only qualitatively, and there are no investigations, which would consider the entire complex of processes determining the optical strength of materials with cracks.

We have considered in detail the impact of a surface cracked layer on the surface strength in case of irradiation with laser. However, the technology of producing cracked layer is such that in addition to cracks the surface layer contains microroughness of the surface, foreign inclusions, structure defects, and so on. Physical processes, resulting in the optical breakdown on a crack, are not separated from other mechanisms of the optical breakdown.

The primary goals of this study were 1) studying the interconnections among the processes accompanying the crack growth and the optical strength of transparent materials; 2) closer understanding of causes of low optical strength of transparent materials with cracks; 3) investigation of the phenomena accompanying the optical breakdown on a crack.

Experimental procedure

The investigations have been carried out using single crystals of NaCl, KCl, LiF, $CaCO_3$. In different experiments, samples with different content of impurities were used. Several work parts of 15×30×16-mm^3 size have been cleaved from a large single crystal. Each work part was cleaved into two equal parts of 15×30×8-mm size. In one of them, in the crystal plane (100), a macroscopic crack of ~6 mm length was introduced. The second part remained undamaged. Then both samples, with and without the crack, were irradiated. The number of radiation pulses before destruction was recorded. The samples were placed so that the crack was in the irradiation zone. A series of experiments was carried out, in which the orientation of input sample surface (100) with respect to the direction of radiation propagation changed from the normal one to the parallel direction in a 5° step.

Neodymium glass laser of GOS 1001 type was used that delivers pulsed radiation at the wavelength of 1060 nm and pulse duration ~1 ms. The pulse energy varied from 40 to 500 J and the irradiated surface area varied from 12 to 100 mm^2. Crystals were irradiated with pulses having energy starting from $E = 0.7E_{\text{th}}$ (E_{th} is the threshold energy) and the energy of every next pulse increased by 10%, up to the sample destruction. Detailed data on radiation can be found in Ref. 10.

The other technique used to irradiate the crystals assumed focusing the radiation pulse into the volume inside a sample for obtaining nonlinear optical effects.

Experimental results and discussion

Destruction of crystals with an original macroscopic crack due to the optical breakdown on the absorbing inclusions

In irradiating crystals of calcite with large absorbing impurities (up to ~50 μm in size and larger) by the radiation pulses of pre-threshold intensity, optically observed areas of 0.3-mm size damages occurred in the sample volume on the spot of large absorbing impurities. In the vicinity of these centers the calcite was decomposed. The pulse energy increase resulted in the sample destruction because of the emergence, at absorbing impurities, of the main cracks in the cleavage planes. Thus, the cause of the calcite crystal destruction and growth of the initial macroscopic crack is the heating of absorbing impurities located in the material volume.

The alkali-halide crystals (AHC), as a rule, were destructed due to the occurrence of main laser-induced cracks on the absorbing impurities, often these cracks combined and merged with the initial macrocrack. A

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macroscopic laser-induced crack can be produced in a plane parallel to the plane of the initial crack and can combine and merge with it due to transitions in the planes {110} and {100}.

The combination of a developed laser-induced crack in the plane (001) with the initial one often resulted in slivering of a part of the crystal. In some cases when no large sources of destruction existed the initial crack grew up and became the main crack. The main cracks occurred in the planes {100}. The cracks located in the planes {110} being well developed only in the region of optical breakdown.

Figure 1 shows the diagrams of characteristic macropatterns of laser-induced destruction of samples with the initial macroscopic crack. The growth of the initial macroscopic crack in samples destructed following the diagram shown in Fig. 1*c* can be the mechanical stresses that can appear in the crystal due to the effect of a high-power laser pulse propagating in it. For samples, destructed according to diagram shown in Fig. 1*b*, these stresses are of thermal nature caused by heated absorbing impurities and mechanical stresses accompanying the growth of a laser-induced crack. The decisive cause of the growth of the initial macroscopic crack in the samples shown in Figs. 1*d*–*j* is, evidently, its interaction with the laser-induced crack.

As the admixture content in the alkali-halide crystal (AHC) decreases, the role of the macrocrack in the material degradation at pulsed irradiation increases.

Peculiarities of the optical breakdown initiation on the crack plane and output surface of calcite samples by self-focused radiation

The irradiation causes the appearance of two channels of burnt out material, corresponding to the ordinary and extraordinary beams. Channels are directed from the output surface to the sample volume, with the maximum length up to 3 mm.

As a rule, self-focusing resulted in the optical breakdown at a focal point. At the same time, at lower pulse energy the self-focusing radiation may travel through the crystal. In this case, the presence is necessary of an absorbing impurity, crystal surface or other optical inhomogeneity on the way of self-focusing radiation for generating the optical breakdown.

The optical breakdown on the output surface leads to the formation of plasma flare and material burning out deep into the sample toward self-focused radiation (Fig. 2).

The plasma pressure does not exceed a critical value, necessary for sample destruction, because the material burning out starts at the output surface, and plasma escapes from the channel being formed. The absence of defects before the channel top (on the way of a self-focused radiation propagation) confirms the proposed mechanism of their formation.

Optical strength of the output surface is lower than of the input one. Two main reasons can be recognized, which determine low value of the optical breakdown for the output surface as compared with the input surface and the sample volume. The one can be the interference between the incident radiation and the radiation reflected from the output surface.[6] The other one can be that the self-focused radiation results in an increase of the power density of incident radiation at the output surface.

The optical breakdown on the sample surface gives rise to the formation of a damaged layer. In this case, the mechanical integrity of the sample can keep. For mechanical destruction of a large sample (with the size much greater than the thickness of the damaged layer) the formation of mechanical waves in the sample is necessary, whose interference could cause the sample destruction.[11] Only a fraction of energy of the plasma formed at the optical breakdown at the crystal surface is transferred to the sample. The origination of plasma of optical breakdown results in the laser radiation absorption and screening of sample volume from it.

In the case a macroscopic crack is on the propagation of self-focusing radiation, the optical breakdown causes the mechanical destruction of the sample.

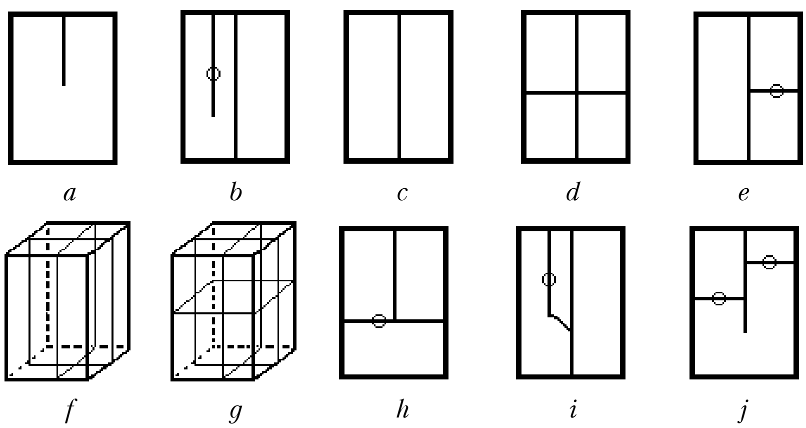


Fig. 1. Macropatterns of the laser-induced destruction of samples having initial cracks: the initial sample with a macroscopic crack (*a*); (*b*) through (*j*) show the destruction patterns after irradiation. Open circles in Figs. *b, e, h, i,* and *j* show the regions of the optical breakdown of the absorbing inclusions.

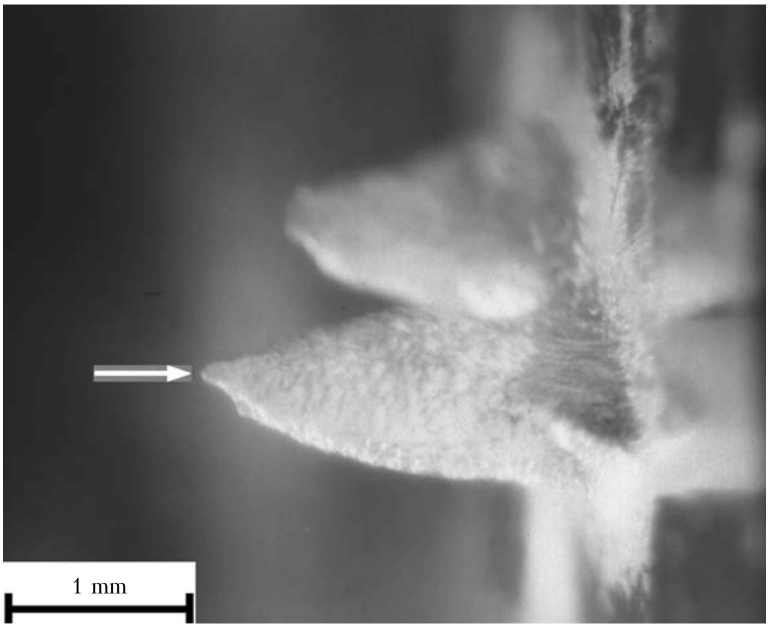


Fig. 2. Channels of the burnt out material directed from the back surface of a sample toward its center. The arrow shows the top of a channel and the direction of incident radiation propagation.

Mechanical destruction at the optical breakdown in the sample volume takes place due to the pressure of heated gas (plasma) on the ambient material. Thus, the optical breakdown on the crack is more dangerous for crystal than the optical breakdown on the crystal surface. The crack top, especially of not aged one, is potentially dangerous place of the destruction development. In all the recorded cases of optical breakdown on a crack caused by an absorbing impurity being located on the crack surface or the breakdown due to self-focusing of radiation the breakdown initiated mechanical destruction of samples.

The effect of the lifetime of cracks on the optical strength of solid transparent materials

By now it has been established that the growth of cracks in approximately one half of solid transparent bodies is accompanied by electron emission. The decrease of optical strength, due to free electrons, is most pronounced in materials with high threshold of optical breakdown. The free electrons play, in such materials, a part of seed electrons initiating the development of the electron avalanche.

In the literature[7,12] it is shown that the illumination by ultraviolet light, leading to the appearance of free electrons, causes the decrease in the optical strength (for laser radiation with the wavelength of 10.6 μm).

The free electrons can appear due to plastic deformation and/or emitted by a growing crack. The threshold of optical breakdown of deformed crystals is about twice as low as that of the same crystals without deformations.[8] Evidence of the role of free electrons in the initiation of optical breakdown in solid transparent materials, having high optical strength, is an experimentally recorded optical breakdown at laser radiation propagation along the surface of a crystal being deformed.[8] Thus, during the crack growth the limiting optical strength is determined by the presence of free electrons generated on the crack surfaces.

Although experimentally a drop in the optical breakdown threshold at the surface of a deformed material was observed on AHC, this effect should take place practically for all solid transparent dielectrics. Actually, free electrons cause a decrease in the laser strength while free electrons themselves appear at destruction of about one half of the known solid bodies. Electric discharges between the banks of growing cracks were observed in semiconductors,[13] alkali-halide crystals,[14–15] piezo-, pyro-, and ferroelectric crystals.[2]

The decrease of optical breakdown threshold, caused by electron emission, can be observed not only at crack formation, but under the action of plastic deformation as well. Intense electron emission and bursts of luminescence were observed at material deformation.[16]

The decrease of the optical breakdown threshold caused by absorption of radiation by free electrons is not limited by the time of crack growth. Free electrons (formed because of the crack growth and accompanying plastic deformation) can survive during several hours and even days.[17] However, for most of transparent materials under normal conditions, free electrons can affect markedly the material radiation strength during ~100 s after termination of the deformation.

For materials, having the optical breakdown threshold of the order of 10^{11} W/cm^2 and higher, laser strength decreases markedly (by about 6 times) when seed electrons occur in the radiation zone.[7]

Using the materials with the optical breakdown threshold of the order of 10^6 W/cm^2 and lower, irradiated with the pre-threshold intensity radiation, single electrons will not produce a marked effect on the laser strength. For such materials the limiting optical strength, as a rule, is restricted by absorbing inclusions. The optical breakdown will be initiated due to heating by laser radiation of absorbing inclusions, and the appearing cracks and free electrons make an impact on the following stages of the development of optical breakdown and mechanical destruction.

The greatest charge density corresponds to the time of crack propagation. The dynamic charge density can be several orders higher than the value of residual charges and can reach ~ 10^3–10^4 μC/m^2 that corresponds[18] to the electron concentration of 10^{11}–10^{12} per cm^2.

Thus in the first period of existence of the crack a dominating factor, determining the limiting value of the optical strength, is the electron emission generated at the crack growth and accompanying its plastic deformation.

The time, during which free electrons make an impact on the laser strength, depends on the material characteristics and the environment.

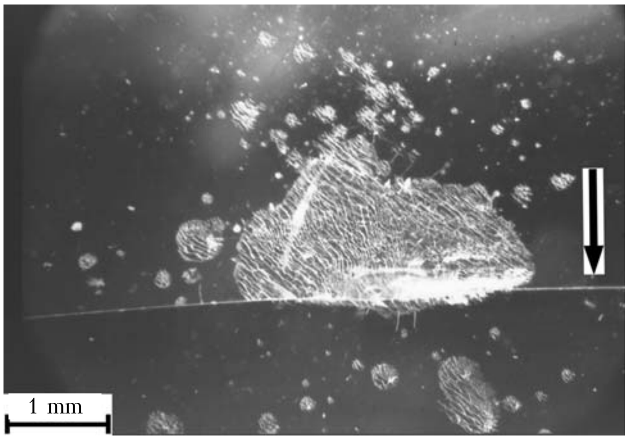
The contribution of free electrons to the decrease of optical strength of transparent material depends on the optical perfection of the material. The higher is the optical breakdown threshold in the absence of free electrons, the more significant is the decrease of optical strength at their occurrence.

In this study the samples were irradiated in 5 min after the introduction a macrocrack when the electron emission and relaxation of high mechanical stress practically terminates. At the same time, the effect of aging of the crack surfaces (due to absorbing inclusions and chemical reactions on the surface) did not affect the laser strength.[5] The highest optical strength corresponds to this period, when the contribution of crack to the decrease of optical strength is lowest and being caused by the interference phenomena only.

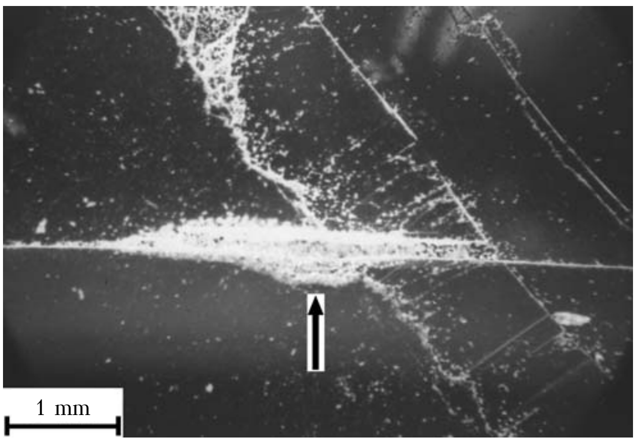
By now a series of mechanisms has been proposed responsible for the decrease of optical strength of transparent materials with cracks and pores.[12]

In this paper we have shown experimentally the effect of one macroscopic crack on the pattern of radiation intensity distribution.

Figure 3 shows the characteristic patterns of damage areas in calcite crystal with a macroscopic crack.



a



b

Fig. 3. Output surface of a sample. Destruction caused by the breakdown on the crack, the initial crack is shown by the arrow (*a*). Destruction on a crack oriented at a small angle with respect to direction of radiation propagation; arrow shows the region of destruction (*b*).

The crater of burned material is extended along the crack. The nature of such a shape is as follows:

1. Birefringence. Because of the birefringence, the effect of light separation into ordinary and extraordinary rays takes place. Depending on the sample thickness either complete or partial separation of ordinary and extraordinary rays occurs.

2. The crystal damage areas are extended along the direction [010] that happens due to the reflection of light from the crack sides. As a result, the size of irradiation area in the [010] direction remains invariable, while that in the direction [100] decreases (Fig. 4).

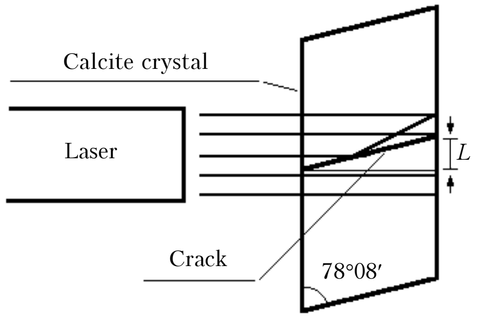


Fig. 4. Optical scheme showing the superposition of the incident light with the light reflected from the exit surface and from the crack plane. The size of the damage along the [100] direction will be smaller than along the [010] direction by the distance L. The scheme presented does not account the radiation focusing.

The probability of damaging the output surface of a transparent material is higher than that of the input one, and can be determined by the ratio $I_{exit}/I_{enter} = [2n/(n+1)]^2$, where n is the refractive index, I_{enter} is the light intensity inside the medium near the input surface, and I_{exit} is that at the output surface.[6] As the radiation propagates through calcite, the radiation intensity decreases due to birefringence in accordance with the Bouguer–Lambert–Beer law for the case of strong fields causing nonlinear phenomena. At the same time, the probability of optical breakdown on the output surface can increase because of superposition of the direct light and light reflected from the crack plane. Such a superposition of incident and reflected light can form on interference pattern. At the interference maxima the optical breakdown can occur, which then can spread into the entire volume irradiated.

The size of the irradiated spot on the output surface, in which the optical breakdown initiation is possible, strongly depends on the orientation of the crack plane relative to the direction of radiation propagation.

Depending on the angle of radiation incidence on the crack, the crack thickness, and the state of surfaces of the crack different processes are possible, including optical breakdown on the crack, partial radiation passage through the crack, radiation propagation along the crack plane, if it is oriented parallel to the direction of radiation propagation, or at an angle of total internal reflection. In the latter case, one can expect multiple increase of the radiation power density in the region of its exit from the crystal. Possibly, the pattern shown in Fig. 3*b* is exactly this case.

When in work the optical breakdown threshold of optical elements' surfaces decreases. The main cause of the optical strength decrease is the adsorption of impurities from the atmosphere, the deposition of dust, etc., on the surface of optical components.

In the materials, containing cracks, the impurity adsorption will also occur on the crack surfaces. In Ref. 5 a two to threefold decrease of optical strength (after 6 months of natural aging) was observed being caused by adsorption of moisture to the surface cracked layer. The adsorption of water took place not in the entire damaged layer but only in its cracked part. As a rule, recrystallization and thermal material decomposition develop on the surface.

The aging rate depends both on the type of material and on the conditions of its storage. The presence of active materials in the environment, the rise in temperature give rise to a faster aging processes. In our experiments we have observed the decrease of the radiation strength of the surface of single crystals of NaCl, KCl, and $CaCO_3$. Moreover, in the case of thermally treated crystals of $CaCO_3$ with a macroscopic crack a tenfold decrease of the optical strength was observed.

Thus in the third period of existence of the crack the limiting optical strength of material is determined by the effect of contamination and the increase of the absorption coefficient of the crack surfaces due to their contact with the environment.

The strain fields and defect structure of the crack top in a sample under the action of electromagnetic radiation

Normally it happens so that the fields of mechanical strain are formed near the top of a new macroscopic crack occurring in the cleavage plane. In samples that are apt to translational deformations, the dislocation density is enhanced near the crack top.

Similar inhomogeneities of structure and characteristics of AHC, subjected to the effect of high-power electromagnetic radiation, can more or less efficiently absorb radiation and evolve thus resulting in a decrease of the optical and mechanical strength of the material.

Depending on the intensity, irradiation time, and spectral composition of radiation, the processes of relaxation of mechanical stress can occur. Under certain conditions the annihilation of some defects is possible as well as partial healing of the macroscopic defects. By now the effect of infrared, visible, and ultraviolet radiation, as well as of small doses of X-rays on the processes of crack healing in AHC has been studied. The variation of the strain fields, integral density of dislocations at the crack top, beam length in the dislocation rosette at the crack top, and the length of healed crack region have also been studied quite well. The impact of the visible radiation on AHC as well as of small doses of ionizing radiation of UV and X-rays range of wavelengths leads to a decrease of the integral density of dislocations at the crack top (asymmetric shear), and the length of dislocation beams. Dependences of variation of the number of dislocations at the crack tops on the duration of radiation effect are of exponential character. Reversible motion of dislocations in the beams of dislocation rosette of the crack explains its plastic closing. The intensity of healing and relaxation of strain depends on the spectral composition of radiation and its intensity.[19]

Consequently, it is necessary to complete known mechanism of laser strengthening of optical materials by irradiating a sample with a prethreshold intensity radiation. As a result of such an irradiation the relaxation is possible of mechanical strain and the decrease of structural inhomogeneities in addition to burning out of small absorbing impurities. Hence, the processing of optical parts by a prethreshold intensity radiation may lead to an increase in their optical and mechanical strength.

Conclusions

1. A dominant reason of activation of growth of the initial macrocrack in AHC and calcite irradiated with a pulsed laser radiation is the heating of absorbing inclusions located in the material volume. The probability of activating the initial crack growth depends on the strength of a heat stress occurring at the absorbing inclusions, and on the location of the absorbing inclusions relative to the crack top. The initial macroscopic crack has a marked destabilizing effect on laser-induced defects occurring in the material volume, it provides high probability of their combining with it or among them only that favors for mechanical destruction of the crystal.

2. In the first period of crack development a dominant factor of reduction of the optical and mechanical strength is the electron emission and electric effects on the crack. In the second period, the crack contribution (whose duration depends on the material and the environment) to the reduction of optical strength of irradiated samples is determined by the interference phenomena on the crack. In the third period, the crack favors a decrease of the optical strength of samples because of the effect of pollution and increase of the absorption coefficient of crack surfaces due to their contact with the environment.

3. The probability of optical breakdown on macroscopic crack depends on the crack orientation relative to the direction of radiation propagation. Threshold density of laser radiation power can multiply decrease due to redistribution of the radiation intensity.

4. The impact of a prethreshold intensity radiation on the crystal with cracks favors the relaxation of mechanical strains, reduction of dislocation density, and partial healing of defects that results in an increase of the mechanical and optical strength of the material.

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