Aging of α-$LiIO_3$ laser-irradiated single crystals

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The intensity of light scattering by microinhomogeneities in α-$LiIO_3$ crystals and the number of pulses which the crystal can endure without breakdown are investigated experimentally as functions of the radiation power per pulse at wavelengths of 1.06 and 0.53 μm. It is shown that under some auxilliary assumptions (supplementing those made by Liberman and Tribelskiĭ [Sov. Phys. JETP **47**, (1978)] and Genkin *et al.* [All-Union Conf. on the Use of Physical Nondestructive Research and Monitoring, Khabarovsk, 1981, p. 76] regarding the mechanism of aging, the experimental data can yield the activation energy of the processes responsible for aging and also the absorption coefficients (linear or nonlinear) of the microinhomogeneities. Similar assumptions also permit an interpretation of the published data on fatigue failure of glasses.

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1. INTRODUCTION

The physical mechanisms of optical breakdown of transparent dielectrics are being widely discussed in the literature.[1–4] This is due to the importance of the problem, both for the understanding of the physics of the interaction of high-power radiation with matter, and for use of optical materials in laser systems. It is shown in a number of recent papers that the threshold of optical breakdown of many materials (glasses,[5,6] polymers,[7,8] α-$LiIO_3$ crystals[9–11]) the optical-breakdown threshold depends on the preliminary irradiation dose. In particular, in Ref. 9 was proposed a phenomenological description of the process of aging of α-$LiIO_3$ crystals, based on the assumption of thermal generation of crystal defects around the absorbing microinclusions. The results of the experiments reported in Refs. 9 and 11, for the one-shot regime of operation, at a small number N of flashes ($N \leqslant 100$), agrees qualitatively with the dependence, predicted in Ref. 9, of the number of flashes required to destroy the crystal on the pulse energy. We report in this paper the results of experimental investigation of the optical strength of α-$LiIO_3$ crystals, jointly with investigations of the change in the light scattering in them following multiple ($N \leqslant 10^4$) irradiation by pulses with $\lambda = 0.53$ and 1.06 μm.[1)] We show that the experimental results can be explained if, besides the assumptions made in Ref. 9, we take into account the following: (a) an important role is played at $\lambda = 1.06\mu$ by the nonlinear absorption of the microinclusions; (b) the temperature rise near the microinclusions is comparable with the initial temperature. The latter assumption makes it possible, on the basis of the obtained experimental data, to estimate the activation energy of defect formation and the absorption coefficient of the microinclusions.

2. EXPERIMENTAL TECHNIQUE

The optical endurance and the photo-aging of the α-$LiIO_3$ crystals was investigated with the laser setup illustrated in Fig. 1.

The laser operated in a single (zeroth) transverse mode. The selection was effected by using a diaphragm in the resonator. No selection of longitudinal modes was made. The frequency doubling was with $LiNbO_3$ or α-$LiIO_3$ crystals. The laser radiation was focused into the investigated samples by lenses with $F = 75$ cm when working at $\lambda = 0.53\,\mu$m and $F = 50$ cm at $\lambda = 1.06\,\mu$m. The beam cross section on entering the crystal was, at the e^{-1} level, $s \approx 0.17$ mm^2 at $\lambda = 1.06\,\mu$m and $s = 0.2$ mm^2 at $\lambda = 0.53\,\mu$m.

We investigated α-$LiIO_3$ crystals with length ~ 1 cm along the beam and with polished end-surface. The crystals were grown by evaporation from salts of "chemically pure" and "special purity" grade from acid (pH $\sim$ 2) and neutral (pH $\sim$ 6.5) solutions. The stability and aging were investigated with an ordinary beam.

In the course of sample irradiation, we observed a change in the scattering of the radiation in the sample. An optical system consisting of a lens 5, diaphragm 6, and light pipe 7 made it possible to receive at a photomultiplier (FÉU 53 at $\lambda = 0.53\,\mu$m and FÉU-22 at $\lambda = 1.06\,\mu$m) the radiation scattered from the sample volume, and discriminated the scattering from the end faces. The crystal breakdown was revealed by an abrupt growth of the scatted radiation, and was subsequently monitored visually.

3. EXPERIMENTAL RESULTS

It was noted in the observations of the radiation scattering that it depends on the crystal irradiation dose. At low

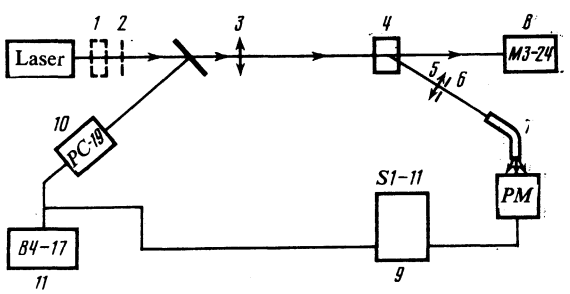

FIG. 1. Block diagram of experimental setup: 1) frequency doubler; 2) filter; 3) lens; 4) sample; 5) objective; 6) diaphragm; 7) light pipe; 8) power meter; 9) high-speed pulsed oscilloscope; 10) photo-electric converter; 11) pulsed voltmeter.

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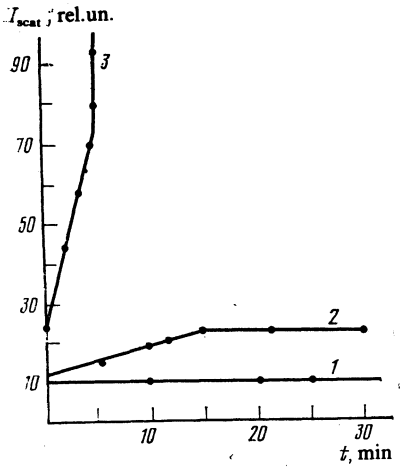

FIG. 2. Dependence of the light-scattering intensity in an α-$LiIO_3$ crystal on the irradiation time at different radiation power inthe pulse: 1) 110 MW/cm^2; 2) 140 MW/cm^2; 3) 165 MW/cm^2; $\lambda = 0.35\,\mu$m.

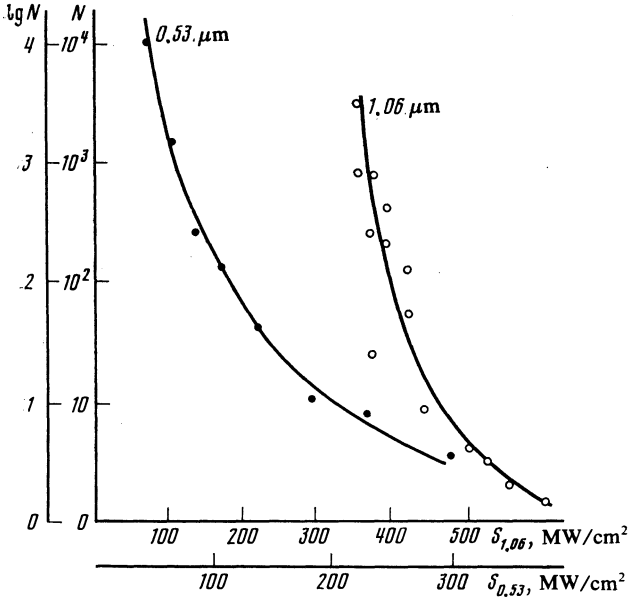

FIG. 4. Typical form of the fatigue relations for an α-$LiIO_3$ crystal at the wavelengths 0.53 and 1.06 μm.

radiation power S_1 the scattering is independent of irradiation time: at $S_2 \sim S_{cr}$ the scattering increases with time, but no crystal damage takes place during the observation time; at powers $S_3 > S_{cr}$ the scattering first increases slowly, followed by an abrupt growth of the scattering (Fig. 2). The crystal damage takes place at that instant. This behavior of the scattering indicates that in the course of the sample irradiation microdefects accumulate in the crystal at $S \gtrsim S_{cr}$. The variation of the scattering with gradual increase of the laser radiation power is shown in Fig. 3. The laser operated at a frequency 6.25 Hz, and the observation at each point lasted 10 sec. At high powers (curve 1), a "nonlinear" scattering of sorts was observed, i.e., an increase in the scattering coefficient. This phenomenon, however, is due not to the nonlinearity of the scattering, but to the gradual accumulation of defects in the course of the measurement. Indeed, on returning to low powers (curve 2) it was observed that the scattering coefficient at these powers also increased. An interesting fact is that after a certain time (on the order of several days) the scattering from the same crystal region decreases somewhat, thus indicating a partial "spreading" of the defects in the crystal and the possibility of their annealing.

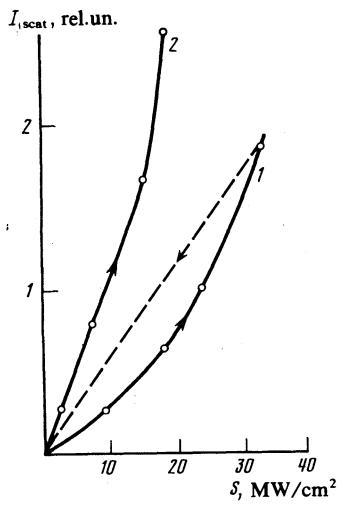

FIG. 3. Dependence of the light-scattering in an α-$LiIO_3$ crystal on the pulse power as the later is continuously increased.

We investigated in detail the dependence of the number N of shots that the crystal can withstand without damage on the power density S in the pulse. Typical plots obtained at $\lambda = 0.53$ and $1.06\,\mu$m are shown in Fig. 4. The experimental results for a number of crystals are shown also in Fig. 5.

It must be noted that each "point" on the plots of Figs. 4 and 5 was obtained, naturally, for an individual point in the crystal. And since such crystals are not always optically homogeneous, this manifests itself in the scatter of the experimental results, so that regular dependences could be obtained only for the most homogeneous samples. The scatter of the experimental points is stronger at $\lambda = 1.06\,\mu$m than at $\lambda = 0.53\,\mu$m.

Just as in the preceding studies devoted to optical endurance of $LiIO_3$ (e.g., Ref. 12), our experiments have shown that the breakdown threshold as well as the rates of aging depend on the quality of the crystal. The best from this point of view turned out to be the experimental crystals[13] grown from neutral solutions of specially pure grade salts. These crystals, however, were noticeably inhomogeneous optically at relatively small dimensions, therefore regular relations of the type shown in Fig. 4 could not be obtained for them.

4. DISCUSSION OF EXPERIMENTAL RESULTS

It follows from the experimental data that the aging of the α-$LiIO_3$ crystal upon irradiation is due to the development of microdefects in it. This development can be attributed to thermal activation of the defects near the absorbing microinhomogeneity, in analogy with the analysis in Ref. 7

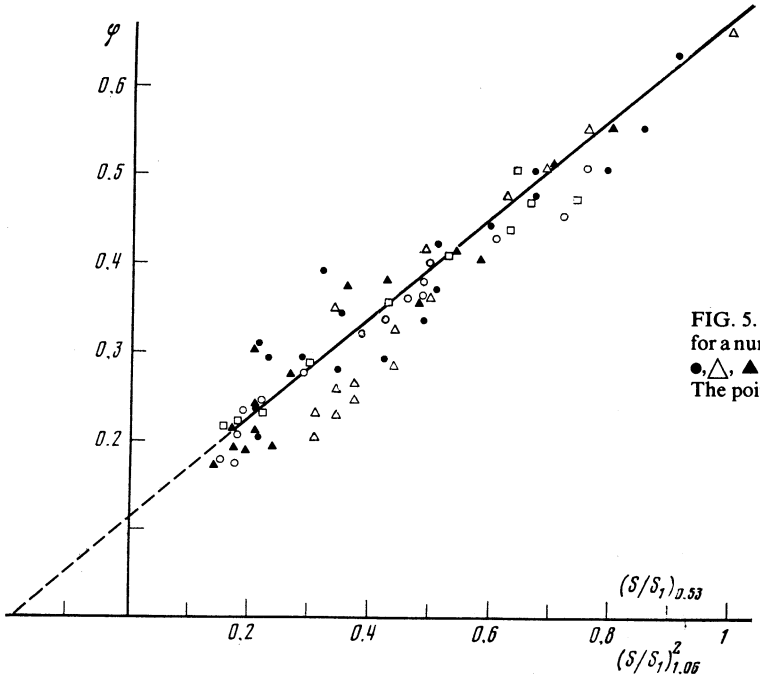

FIG. 5. Dependence of the function $\varphi(S/S_1) = 1/(\log N + a)$ at $a = 1.5$ for a number of α-$LiIO_3$ crystals: □,○ – $\lambda = 0.53\,\mu$m, $S_1 \approx 185$ MW/cm^2; ●,△, ▲ – $\lambda = 1.06\,\mu$m, $S_1 = 670$, 620, and 705 MW/cm^2, respectively. The points ○ and ● pertain to the same sample.

of data on optical breakdown in polymers. We shall assume that when a short light pulse ($\tau \ll R_0^2/\chi$, R_0 is the size of the inclusion and χ is the thermal diffusivity) the microinclusion is heated to a certain temperature $T_{in} + \Delta T$ (T_{in} is the initial crystal temperature) and generates crystal defects around itself in the course of cooling. If it is assumed that there exists a certain critical density of defects, at which the "induced" absorption becomes substantial and avalanche-like processes develop as a result[2-4] and lead to damage to the crystal, we easily obtain a relation between the number N of laser pulses that the crystal can withstand without damage and the power S in the pulse[2)]:

$$N(S) = \exp[-T_0/T_{in} + \Delta T^*] \exp[T_0/(T_{in} + \Delta T)]. \qquad (1a)$$

Here T_0 is the defect activation energy; $\Delta T = \alpha S\tau/c + \beta S^2\tau/c$ is the microinhomogeneity temperature rise, α and β are respectively the coefficients of linear and nonlinear absorption, c is the specific heat, ΔT^* is the heat rise accompanying damage by one shot. To compare the relation (1a) with the experimental data it is convenient to rewrite it in the form

$$\varphi\left(\frac{\Delta T}{\Delta T^*}\right) = \left[\lg N + \frac{T_0 \lg e}{T_{in} + \Delta T^*}\right]^{-1} = \frac{1}{\lg e}\frac{\Delta T^*}{T_0}\left(\frac{T_{in}}{\Delta T^*} + \frac{\Delta T}{\Delta T^*}\right). \qquad (1)$$

Analysis of the experimental results (see Fig. 5) shows that relation (1) describes well the aging of an α-$LiIO_3$ crystal if it is assumed that the absorption of the microinclusion is mainly linear at $\lambda = 0.53\,\mu$m and nonlinear at $1.06\,\mu$m. The activation temperature T_0 is in this case 6000–8000 K, and $\Delta T^* \approx 1500$–2000 K. From these data we can estimate also the absorption coefficient, namely $\alpha = 10^3$–10^4 cm^{-1} at $\lambda = 0.53\,\mu$m and $\beta \approx 1$ cm/MW at $\lambda = 1.06\,\mu$m.

The obtained estimate of the activation temperature of the defects in the crystal agrees in order of magnitude, for example, with the activation energy $W_3 \approx 0.5$–0.66 eV of the ionic conductivity (Ref. 14) and definitely contradicts the fact that the α-$LiIO_3$ crystal melts with gradual decomposition at $T > 435$ °C (Ref. 14).

As for the absorption coefficients, we note that at microinclusion sizes $\sim 1\,\mu$m and densities $\sim 10^3$ cm^{-3} the average absorption at $\lambda = 0.53\,\mu$m is 10^{-5}–10^{-6} cm^{-1}, in agreement with the high transparency of the crystal in the mean. Nonlinear absorption with $\beta > 1$ cm/MW was observed, for example, at $\lambda = 1.06\,\mu$m in GaAs.[15,16]

It follows from the foregoing analysis that different samples differ mainly in the character of the microinclusions (in their absorption coefficients), and since the character of the microinclusions can depend on the growth technology, it is clear that aging of crystals grown under other conditions can evolve in a somewhat different manner.

It is interesting to note here that analogous nature is possessed apparently also by photo-aging in optical glasses.[5,6] Indeed, the results of Refs. 5 and 6 agree qualitatively with the relation (1) if it is assumed that $\lambda = 0.53\,\mu$m absorption with $\alpha \approx 1$ cm^{-1} is substantial; at $\lambda = 1.06\,\mu$m in the giant pulse regime,[6] the nonlinear absorption with $\beta \approx 10^{-4}$ cm/MW is significant, and in the free lasing regime[5] at $\lambda = 1.06\,\mu$m linear absorption predominates; this is natural in view of the low radiation power in this case. The defect activation temperature, which can be estimated from the data of Refs. 5 and 6, is $\sim 6 \cdot 10^3$ K. An increase in the absorption by microinclusion in glasses was noted in Ref. 17.

To conclude the discussion of the experimental results, we must dwell on the possible role of self-focusing. The point is that as a rule lasers generate several longitudinal modes. The fluctuations of the power during the time of the pulse can in principle lead to self-focusing of the beam. Microscopic damage is then produced in the crystal. Such microscopic damage accumulates under periodic action and ultimately

leads to macroscopic damage. The character of the fatigue dependences is determined in this case by the statistics of the laser radiation.

Investigations of the temporal and spectral structure of the pulse of the employed YAG:ND^{+3} laser has shown that the power fluctuations in them are not large enough for this effect. Our additional experiments on the fatigue dependences when crystals are irradiated at different focusing ($F = 66$, 40, and 14 cm) have shown that, within the limits of experimental error, these dependences are not connected with the degree of focusing, and the crystal aging is determined only by the target density S. This also points to an insignificant role of self-focusing in our experiments.

5. CONCLUSION

The experimental results obtained on the fatigue damage of α-$LiIO_3$ crystals can be quite satisfactorily explained within the framework of a simple model of the growth of microinhomogeneities when they absorb (in linear or nonlinear fashion) laser radiation. The photoaging of optical glasses, where the damage is also due to the absorbing microinclusions, is of similar character. There is convincing evidence that optical damage (produced by one shot) of other optical materials (KPD and DKPD) is also due to absorbing microinclusions.[4] This allows us to conclude that the aging effect in the field of high-power laser pulses is a property possessed by a larger class of optical materials than heretofore assumed. Its observation was limited mainly by the high optical endurance of the materials, and consequently, by the need of using in the investigations high-power laser sources of multiple action. In addition, difficulties arise frequently also in observation of the photo-aging. Our results showed that observation of the light scattering in crystals during the course of their irradiation is quite suitable for this purpose.

[1)] Preliminary results of the investigations at $\lambda = 0.53\,\mu$m are presented in Refs. 10 and 11.

[2)] It is assumed that the cooling time of the microinclusion is much shorter than the pulse repetition period. In addition, it is assumed that some "freezing" process acts on the defects produced after the vicinity of the microinclusions is cooled, therefore the relations obtained can be used only at sufficiently large ΔT.

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