SELF-FOCUSING OF LASER RADIATION IN SOLID DIELECTRICS

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Submitted May 8, 1969

Self-focusing of the second harmonic radiation ($\lambda = 530$ nm) of a single-mode neodymium laser resulting in the formation of filamentary fractures in glasses, sapphire, and ruby is investigated. Compression of the laser beam prior to filamentary fracture was observed. The threshold radiation powers leading to the formation of filamentary defects in materials with different absorption are measured as functions of the conditions of laser—radiation focusing on the samples. The rate of “growth” of the filamentary fractures towards the laser beam ($\sim 10^8$ cm/sec) is determined. The dependence of the filament lengths on the incident power is investigated. The magnitude of nonlinear changes in the refraction index $n_2$ is estimated on the basis of the experimental results, and possible self-focusing mechanisms in solid dielectrics are discussed.

INTRODUCTION

For many years, the phenomenon of self-focusing of laser radiation has been attracting the attention of many investigators. In most experiments, this phenomenon was investigated in liquids (see, for example, [1]). At the same time, self-focusing of laser radiation takes place also in many solid dielectrics. In such materials as glass, quartz, ruby, leucosapphire, or nonlinear crystals, the passage of a laser beam gives rise to characteristic filamentary damage, which can be explained only as a result of self-focusing. Similar damage can be observed directly in the active elements of Q-switched lasers, namely ruby and neodymium glass. Self-focusing in laser materials can influence also the formation of the laser radiation field, and determine the limiting endurance of these materials under the action of powerful optical radiation.

It was shown in [1, 2] that the filamentary damage observed in solid dielectrics is not connected with the waveguide propagation of light in the medium. This damage occurs when the region in which the light beam collapses as a result of the increase of the refractive index of the medium propagates in the medium during the time of the laser pulse in a direction opposite to the incident radiation.

We report here further investigations of the self-focusing of laser radiation in solid dielectrics, namely transparent and colored glasses, leucosapphire, and ruby. We show that the appearance of filamentary damage is preceded by light-beam contraction caused by the self-focusing. We measure the threshold laser radiation power, at which filamentary damage occurs in materials with different absorption, as a function of the conditions for the focusing of the laser radiation in the samples. We studied the dynamics of development of the filamentary damage, determine the rate of “growth” of the filament-like damage in a direction opposite to the incident beam. We investigate the dependence of the length of the filaments on the incident power.

On the basis of the experimental results, we estimate the values of the nonlinear additions to the refractive index of the medium in the field of the light wave, and consider possible mechanisms of self-focusing in the investigated materials.

EXPERIMENTAL PROCEDURE

The self-focusing investigations were carried out in transparent dielectrics—optical glass K-8 and leucosapphire. We also investigated colored materials—glass ZHS-11, sapphire crystals with small additions of chromium, and ruby crystals. In the experiments we used second-harmonic radiation of a Q-switched neodymium laser operating with one transverse mode. The second-harmonic radiation ($\lambda = 0.53 \mu$) led to the formation of longer filamentary flaws with smaller threshold than the radiation from a neodymium laser ($\lambda = 1.06 \mu$). In this case the filaments were not accompanied by strong cracking of the material. The single-mode laser operation made it possible to obtain one filamentary flaw per flash with good reproducibility of the results.

The laser consisted of a generator and one amplifier with active elements of LGS-5 glass, measuring 10 mm in diameter and 130 mm in length. The concentric resonator of the generator was made up of a spherical dielectric mirror and a flat glass substrate. The Q switch was a phototropic liquid. To obtain stable generation on one transverse mode, a diaphragm of 2.0 mm diameter was placed in the resonator.

The experimental setup is shown in Fig. 1. The radiation from the master generator 1 was amplified in amplifier 2 and directed to KDP crystal 3 to obtain the second harmonic. The S2S-21 filter 4 absorbed the radiation of the fundamental frequency of the neodymium laser, while the neutral filters 5 made it possible to vary the energy of the harmonic. The radiation passed by the filters was focused by a lens 6 into the interior of the investigated polished samples 7. Part of the

FIG. 1. Diagram of experimental setup. 1 — Neodymium-glass laser ($\lambda = 1.06 \mu$), 2 — amplifier, 3 — KDP crystal, 4 — S2S-21 filter, 5, 15 — neutral filters, 6 — focusing lens, 7 — investigated sample, 8 — glass plates, 9 — calorimeter, 10, 11 — FEK-09 photodiodes, 12 — S1-11 oscilloscope, 13 — photographic objective, 14 — microscope with camera.
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radiation was diverted by glass plates 8 to a calorimeter 9 in order to measure the energy and to a photodiode 10 in order to trigger the sweep of the S1-11 oscilloscope 12. A high-speed FEK-09 photodiode 11 was used to register the scattered laser radiation on the developing filamentary flaw. The field distribution of the laser radiation was investigated with the aid of photographic lens 13 and a microscope with photographic camera 14, placed behind the output surface of the sample. The neutral filters 15 made it possible to obtain photographic images of normal density.

The energy of the neodymium-laser harmonic reached 0.02 J at a pulse duration of approximately 10 nsec. The beam diameter in the plane of the lens was 1.5 mm. The length of the filamentary flaw following application of the total radiation power and using a focusing lens with f = 110 cm fluctuated for different samples from 15 to 30 mm. The longest filaments in the colored glass ZhS-11 greatly exceeded the focal length of the lens. The diameter of the filament at the region of the focus of the lens was approximately 5 μ, increasing to 10–12 μ towards the end the diameter increased, and the filament usually terminated in a crack measuring 0.1–0.5 mm. As a rule, practically continuous filaments were observed (Fig. 2a).

However, in many cases intermittent filaments with a clearly pronounced periodic structure were observed. The most distinct periodicity (with a distance of approximately 20 μ between the flaws) was observed in leucosapphire crystals (Fig. 2b). This character of the damage may be connected with the high-frequency modulation of the laser pulse, with approximate frequency 5 × 10\(^{10}\) Hz, due to beats of the neighboring axial modes.

Filamentary flaws induced by the fundamental frequency of the neodymium laser reached up to 20 μ in diameter and was accompanied by appreciable cracking of the material.

2. CHANGE OF TRANSVERSE LASER BEAM STRUCTURE PRECEDING THE FILAMENTARY DAMAGE

The 0.53–μ radiation was focused with a lens of f = 40 cm into a sample of ZhS–11 glass. Over the length of the sample (l = 60 mm), the cross section of the laser beam remained practically constant (at low intensities) at about 10\(^{-2}\) cm. The ZhS–11 glass has the lowest threshold of filamentary damage among all the investigated materials, making it possible to use long-focus lenses. Under such conditions, it is possible to observe most clearly the contraction of the light beam.

Figure 3 shows photographs of the transverse cross section of the laser beam passing through the sample at power levels close to the threshold of the filamentary damage \(P_{\text{thr}}\). At a power \(P \approx 0.5P_{\text{thr}}\), the radiation field has a Gaussian character (Fig. 3a). Raising the power to \(0.9P_{\text{thr}}\) leads to the appearance of a bright central region in the beam, with diameter 30–50 μ. At a power corresponding to the threshold of the filamentary damage, the diameter of the bright region is close to the diameter of the filament (approximately 10 μ).

These results show that filamentary damage in solid dielectrics is directly connected with the phenomenon of self-focusing of the laser radiation. We note that the action of a single-mode laser leads to the appearance of one central spot in the distribution of the radiation field, and consequently, to one damage filament. At the same time, in the case of a multimode regime, a number of filaments per flash is produced; the most intense regions of the laser beam are focused separately into filaments.

3. THRESHOLDS OF FILAMENTARY DAMAGE. DEPENDENCE OF THE FILAMENT LENGTH ON THE RADIATION POWER

We measured the threshold of filament-like damage with a lens (f = 10 cm) at a radiation wavelength 0.53 μ in the K-8 and ZhS-11 glass, in sapphire with varying absorption, and in ruby. The table lists the thresholds for the investigated materials, and also the values of the absorption coefficient \(κ\) at the generation wavelength.

The lowest threshold was observed in colored glass

<table>
<thead>
<tr>
<th>Material</th>
<th>(κ), cm(^{-1})</th>
<th>(P_{\text{thr}}), MW</th>
<th>(P_{\text{cr}}), MW</th>
<th>(\delta_f) from measurements lengths</th>
<th>(\delta_f) from threshold measurements</th>
<th>(\delta_f), cm</th>
<th>(\delta_f), theory (10^{-14})</th>
</tr>
</thead>
<tbody>
<tr>
<td>K-8 glass</td>
<td>(-10^8)</td>
<td>0.3</td>
<td>0.7</td>
<td>3 × 10(^{-14})</td>
<td>1.6 × 10(^{-14})</td>
<td>1.5 × 10(^{-14}) (striction)</td>
<td></td>
</tr>
<tr>
<td>Leucosapphire Sapphire</td>
<td>1.4 × 10(^{-9})</td>
<td>0.5</td>
<td>0.6</td>
<td>0.6 × 10(^{-13})</td>
<td>1.2 × 10(^{-16})</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ruby</td>
<td>1.5</td>
<td>0.14</td>
<td>0.15</td>
<td>1.7 × 10(^{-13})</td>
<td>4 × 10(^{-16})</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ZhS-11 glass</td>
<td>4.2 × 10(^{-9})</td>
<td>0.1</td>
<td>0.15</td>
<td>1.7 × 10(^{-13})</td>
<td>4 × 10(^{-16})</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
ZhS-11. In sapphire, the threshold dropped with increasing absorption coefficient. In a ruby crystal with absorption coefficient $\kappa = 1.5 \text{ cm}^{-1}$ at a wavelength 0.53 $\mu$, the threshold depended on the position of the focal region in the sample. If the distance from the input surface of the sample to the focus $l_f$ exceeds $1/\kappa$, then the threshold in ruby is higher than in sapphire, owing to the strong absorption in the sample. In the opposite case, when $l_f < 1/\kappa$, the threshold in ruby is lower than in leucosapphire. The table lists the value of the threshold in ruby for $l_f = 0.5$ cm. At the same time, the thresholds in materials with small absorption ($l_f \ll 1/\kappa$) remain practically unchanged when $l_f$ varies in a wide range.

Fig. 6 shows oscillograms of laser pulses at a power 10–20% below the damage threshold (Fig. 6a) and at a power slightly exceeding the threshold (Fig. 6b). The distortion of the pulse, observed in Fig. 6b, is due to absorption and scattering of the beam, which has been compressed by self-focusing, by the produced filamentary flaw. If the radiation is attenuated by a neutral filter in such a way that no self-focusing of the beam occurs, and the beam is passed again through a region of the sample where filamentary flaw is already present, then no distortion of the laser waveform will be observed. At low radiation intensity, a thin filamentary flaw (5–10 $\mu$ diameter) has little effect on the propagation of the light beam through a focal region of about 50 $\mu$ diameter. The time during which the change in the pulse waveform is observed corresponds to the time of existence of the compressed state of the light beam, when its diameter is close to the diameter of the filamentary flaw.

As seen from the oscillogram on Fig. 6b, the distortion appears after the maximum of the laser pulse (2–3 nsec later) and is observed practically to its end. From the considerable depth of the dip in the pulse it follows that a large section of the beam cross section is involved, carrying up to 70% of the power. The delay of the dip relative to the maximum of the laser pulse offers evidence that the mechanisms that lead to self-focusing are subject to inertia.

5. DYNAMICS OF DEVELOPMENT OF FILAMENTARY-LIKE DAMAGE

As shown in[1], filamentary damage in solid dielectrics develops in a direction opposite to the incident radiation as a result of motion, in this direction, of a region of collapse of the light beam. This is due to the
growth of the refractive index of the medium in the field of the light wave during the laser pulse.

We have investigated the dynamics of the development of filamentary damage and determined the rate of motion of the filament in a direction opposite to the laser beam. 0.53-μm radiation at full power was focused into a ZnS-11 glass sample, in which the filamentary flaws are longest. An FŠK-09 photodiode located near the sample, perpendicular to the laser beam (Fig. 1), registered the laser radiation scattered from the developing filamentary flaw. After each flash, the sample was moved 1–2 mm in a direction perpendicular to the light-beam axis. The signal from the photodiode was fed to an SI-11 oscilloscope (the resolution of the oscilloscope with the amplifier was 4 nsec). Figure 7a shows an oscillosgram of the pulse scattered by the filamentary flaw. The oscillosgram of Fig. 7b shows the laser pulse scattered only by the ends of the filament. Its central part was covered with an opaque screen 15 mm wide, and the ends projected approximately 5 mm from each side of the screen. It is seen from this oscillosgram that the damage in the filament occurs during the time of the laser pulse, and that the delay between the time of formation of the start and the end of the filamentary flaw is 8–10 nsec, corresponding to a growth of the refractive index of the medium in the field vector. At the entrance to the nonlinear medium we have

\[ \frac{dz}{d\omega} = \frac{1}{\alpha_0 R_{n\ell}} + \frac{1}{\alpha_0 R_n} \]

where \( \alpha_0 \) is the growth rate of the light wave during the laser pulse. \( o(z) = o(0) \) is the diameter of the light wave during the laser pulse.

The oscillogram of Fig. 7b shows the damage in the filament occurring during the time of the laser pulse, and that the delay between the time of formation of the start and the end of the filamentary flaw is 8–10 nsec, corresponding to a growth of the refractive index of the medium in the field vector. At the entrance to the nonlinear medium we have

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in mind here that the theory does not take into account the experimentally observed inertia of the nonlinearity.

Figure 4 shows the solid curves describing the dependence of $P_{th}$ on $d_0$ in accordance with expression (9). The values of $l_{th}$ and $P_{cr}$ are the parameters of these curves. The curves were plotted for the following values of $l_{th}$ and $P_{cr}$: $1.3 \times 10^6$ W/cm$^2$ and $1.5 \times 10^5$ W for ZhS-11 glass, $2.5 \times 10^5$ W/cm$^2$ and $5 \times 10^5$ W for sapphire, and $3 \times 10^5$ W/cm$^2$ and $7 \times 10^5$ W for K-8 glass. The values of $l_{th}$ and $P_{cr}$ can be determined by writing (9) in the form

$$l_{th} = l_{th} + d_0 \frac{d}{P_{cr}}$$  \hspace{1cm} (10)$$

(where $l_{meas} = \frac{dP_{th}}{dP}$), and by plotting the experimental curve of $l_{meas}$ against $d_0$. Then the intersection of the experimental lines with the ordinate axis, as follows from (10), gives the value of $l_{th}$, and the slope relative to the abscissa axis gives $P_{cr}$. The values of $P_{cr}$ and the corresponding values of $d_0$ are listed in the table.

It is seen from Fig. 4 that the experimental results are well described by expression (9) for ZhS-11 glass and sapphire. However, for K-8 glass at $d_0 > 5 \times 10^{-3}$ cm, the experimental values of $P_{th}$ increase with $d_0$ more strongly than would follow from (9). This discrepancy may be connected with the fact that $P_{cr}$ increases in this region like $\sim d_0$. The reason for the dependence of $P_{th}$ on $d_0$ will be considered below.

The most probable mechanisms of self-focusing in solid dielectrics may be connected with thermal effects and with electrostriction. According to L. Zernike, heating of a medium in the light channel can lead to self-focusing, if the refractive index of the medium increases with temperature (i.e., $dn/dT > 0$). Sapphire and glass are substances of this kind. A characteristic feature of thermal self-focusing is the dependence of $n_2$ on the time $t$ and on the absorption coefficient $\kappa$. The experimentally observed decrease of the threshold $P_{th}$ for the formation of self-focusing filaments with increasing absorption for sapphire and ZhS-11 glass shows that self-focusing in these materials is determined by the thermal mechanism.

The value of $P_{cr}$ calculated in accordance with $\kappa = 2 \times 10^{-6}$ cm$^{-1}$ and $0.7 \times 10^5$ W for ZhS-11 glass. These values are approximately half the values of $P_{cr}$ calculated from the experimental data using expression (10). Taking into consideration the approximate character of the stationary theory, from which expression (10) is derived, we can regard the agreement between the calculated and experimental values as satisfactory. The differences between the calculated values of $P_{cr}$ and the experimental ones may be connected also with the large relaxation time of the excited states, compared with the duration of the pulse, and with the saturation of the absorption. We note that for thermal self-focusing the increment of the refractive index increases with time and reaches a maximum at the end of the pulse. The values of $n_2$ estimated from experiment pertain to the second half of the laser pulse, when the power density at the focus reaches a maximum.

In K-8 glass, where the absorption coefficient is small ($\kappa = 10^{-3}$ cm$^{-1}$), an important role may be played, besides the thermal mechanism, also by the mechanism of electrostriction, which was considered in $^{12}$. The striction self-focusing can be regarded as a stationary process, if a quasiequilibrium density of matter, corresponding to the striction pressure produced in the light field, is established in the light channel during the time of the laser pulse. The nonlinear increment $n_2$ of the refractive index, calculated in accordance with $\kappa = 6 \times 10^{-3}$ cm, amounts to $1.5 \times 10^{-14}$ cgs emu, while the thermal effects yield $n_2 \approx 1.0 \times 10^{-14}$ cgs emu. The critical power determined by the joint action of the striction and thermal mechanisms amounts to $9 \times 10^5$ W, which is close to the experimental $7 \times 10^5$ W. If the beam diameter at the focus is $d_0 > 5 \times 10^{-3}$ cm, then the density of the matter in the channel does not reach the equilibrium value during the pulse time $t_{pulse}$, since $d_0 > \tau_{pulse} V_{ac}$ (vac velocity of sound in the medium). In this case it is necessary to take into account the nonstationary character of the striction self-focusing, which leads to a dependence of the critical power on the beam diameter. According to $^{12}$, $P_{cr} = \kappa d_0$. This apparently explains the deviation of the dependence for the K-8 glass in Fig. 4 from the calculated value at a diameter $d_0 > 5 \times 10^{-3}$ cm, starting with which $P_{cr}$ increases approximately quadratically with increasing $d_0$.

The possible action of the striction mechanism of self-focusing in K-8 glass is qualitatively confirmed also by the appearance in this glass of stimulated Mandelshtam–Brillouin scattering at a power approximately half as large as the threshold of the filamentary damage (for a focusing lens with $f = 10$ cm). This is evidence of the appreciable value of the constant $(\rho \kappa / \rho)$ for glass, which determines also the threshold of the striction self-focusing $^{12}$. At the same time, we were able to observe stimulated scattering in sapphire crystals, at a radiation power 4–5 times larger than the threshold of filamentary damage.

Let us examine now the development of filamentary damage during the time of the laser pulse at a radiation power exceeding the threshold. At the first instant, the collapse of the beam occurs in the focal region of the lens, where the filamentary damage is initiated. The subsequent increase of the radiation power leads to an increase of the refractive index of the medium and to a decrease of the self-focusing length. The region of collapse moves in a direction opposite to the incident beam. The position of the end of the filamentary flaw is
determined by the maximum value of the increment $\Delta n$ of the refractive index of the medium. For inertialess self-focusing, the maximum value of $\Delta n$ is reached at the maximum of the laser pulse. In the case of mechanisms with inertia, on the other hand, the maximum of $\Delta n$ can lag the pulse maximum.

The length of the filamentary flaw is determined thus as the difference between the focal distance of the lens in the sample $z_f$, where the filament-like damage begins, and the minimum length of self-focusing $z_{sf}$ which determines the position of the end of the filament.

In accordance with\(^{(10)}\) we have

$$\frac{1}{z_f} = \frac{1}{\Gamma_{ds}} + \frac{1}{|R|},$$

where

$$\Gamma_{ds} = R_{ds} \left( 1 - P_{\text{cr}} / P \right)^{-\beta}, \quad \tau_f = |R|.$$  

Then the length of the filament is $L = z_f - z_{sf} = R^2 / (|R| + R_{nd})$. For the case $R_{nd} \gg |R|$, we get

$$L \approx 4R^2 \left( \frac{\eta \gamma}{c n_0^2} \right)^{1/\beta} \left( \frac{R}{a} \right)^{1/2} \left( P - P_{\text{cr}} \right)^{1/\beta}.$$  

The inequality $R_{nd} \gg |R|$ corresponds to small changes of the focal distance of the lens due to the self-focusing, compared with the focal distance itself.

We note that $z_{sf}$ determines the position of the collapsed region, where the beam diameter $d$ vanishes. At the same time, from the preceding analysis it follows that filamentary damage occurs when $d > 0$. We take this circumstance into account by writing in place of (12) an approximate equation, in which $P_{\text{cr}}$ is replaced by $P_{\text{thr}}$:

$$L \approx 4\gamma^2 \left( \frac{\eta \gamma}{c n_0^2} \right)^{1/\beta} \left( \frac{R}{a} \right)^{1/2} \left( P - P_{\text{thr}} \right)^{1/\beta}.$$  

The filament length $L$, as follows from experiment (Fig. 5), depends linearly on $(P - P_{\text{thr}})^{1/2}$ in accordance with (13). From the slopes of the lines we determine the values of $n_0$ for the investigated materials. These values, which are listed in the table, are in satisfactory agreement with the values of $n_0$ obtained from the threshold experiments.

We note once more that the foregoing analysis, as well as the analysis of the results of the threshold measurements, is based on a stationary self-focusing theory, in which $n_0$ is a constant of the material and does not depend on the spatial coordinates or on the time. For real nonstationary processes, this analysis is approximate. At the same time, satisfactory agreement between the foregoing analysis and the experimental results offers evidence that the inertia of the nonlinearity does not change radically the course of the self-focusing in solids.

In conclusion, we are grateful to T. N. Mikhailova for help with the work.

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Translated by J. G. Adashko