STIMULATED MANDEL'SHTAM-BRILLOUIN SCATTERING, DAMAGE, AND SELF-FOCUSING IN GLASSES

V. M. RYSAKOV

Institute of Semiconductors, USSR Academy of Sciences

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Stimulated Mandel'shtam-Brillouin scattering (SMBS) and damage in glasses are studied. It is suggested that the diameter of filamentary damage is determined by the intensity of a collapsing elastic wave due to electrostriction. A consideration of the results from this viewpoint permits one to explain the development of SMBS and damage processes in glasses induced by laser-pulse irradiation. The experimental results are in good agreement with Kerr's theory[7], according to which the self-focusing threshold is determined by electrostriction under nonstationary conditions.

STUDY of the processes of stimulated Mandel'shtam-Brillouin scattering (SMBS), damage, and self-focusing (SF) has recently attracted the attention of many investigators[1-7], but the essential details of these processes have not yet been satisfactorily explained. To obtain some additional information, we have investigated the processes of SMBS, damage, and SF in the glasses K-8 and LK-5, in fused quartz (KU), in one type of heavy flint (TF), and in organic glass (plexiglas). We used a single-stage Q-switched ruby laser of power ~25 MW and pulse duration ~40 nsec. The operating regime was close to single-mode, the beam divergence ~5′, and the line half-width ~0.3 cm^{-1}. The SMBS was observed with the aid of a Fabry-Perot interferometer in the backward direction (θ_S = 180°), and the spectrum was recorded photographically.

EXPERIMENTAL RESULTS

1. It was observed that the character of the damage depends strongly on the focal length of the lens employed. Let us examine this dependence in greater detail using K-8 glass as an example. For a lens with F = 20 mm, the damage was in the form of the usually observed ellipsoids. When lenses with focal lengths F = 90, 200 and 500 mm were used, the damage was in the form of filaments with different structures. With a lens having F = 90 mm, the filament (Fig. 1a) was short but large in diameter. With a lens having F = 200 mm, the structure of the filament (Fig. 1b) was different, namely, the filament had a relatively large diameter in the region of the focus (~0.03 mm), after which it decreased jumpwise by approximately one order of magnitude and such a thin filament extended on both sides of the focus, gradually decreasing in diameter[1)]. With an F = 500 mm lens (Fig. 1c), very thin, practically uniform filaments were observed (usually several at a time). The averaged parameters of the observed tracks are shown in Table I.

The obtained data can be interpreted in the following

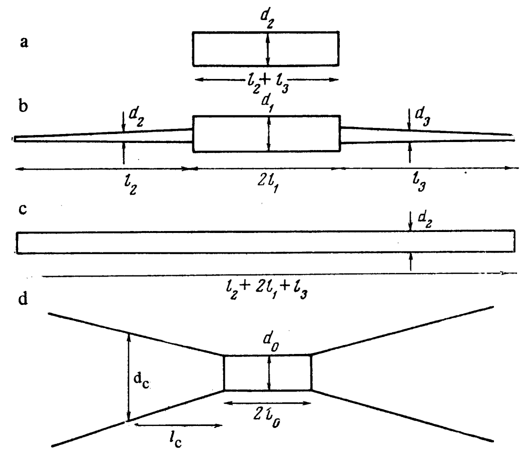

FIG. 1. Schematic diagram of damage observed with lenses of F = 90 mm (a), F = 200 mm (b), and F = 500 mm (c), and schematic diagram of light beam in the region of the lens focus (d).

manner. As already noted a number of times, there are no sufficient grounds for assuming that the observed tracks are true tracks of waveguide channels, and that their diameter is equal to the diameter of the self-focusing-beam channel. It can be assumed that the track diameter is determined by the intensity of the compressing elastic wave which is shocklike in character and is due to electrostriction. Damage occurs when this wave collapses on the beam axis.

For a quantitative estimate we shall use the result of the theoretical paper[7], which considers the nonstationary problem of self-focusing due to electrostriction. It is shown in the paper that if the self-focusing threshold power in the stationary case is K, then in the nonstationary case it increases sharply. The time of establishment of the stationary state is determined by the time of flight of the acoustic wave from the boundary of the initial beam to its center. In the nonstationary case the threshold SF power depends on the parameter

$$\beta = d/2\tau v, \qquad (1)$$

where d is the diameter of the initial beam, τ the pulse duration, and v the speed of sound.

We shall assume that the dimension of the damage

[1)]In these thin filaments, the damage was usually not continuous, but consisted of several segments with lengths up to 20 mm. Discontinuities in the tracks were also observed with a lens having F = 90 mm.

Table I. Averaged parameters of observed tracks and calculated dimensions of self-focusing regions (in mm)

F	$2l_1$	d_1	l_2+l_3	$d_2(\approx d_3)$	$2l_0$	d_0	β	$2l_c$
90			10—20	0.1—0.3	0.7	0.2	0.4	30
200	4—7	0,03—0.04	40—60	0.008—0.003	3.8	0.4	0.8	70
500			80—100	0.001	20	1.0	2	150

depends on the intensity of this elastic shock wave, and the intensity depends on the excess above threshold power. Let us consider the results from this point of view.

As is well known, the cross section of the beam in the focal region of the lens has approximately the form shown in Fig. 1d, with

$$d_0 = \vartheta F, \qquad (2)$$

$$2l_0 = (\sqrt{2}-1)F^2\vartheta/D, \qquad (3)$$

where ϑ is the total divergence of the laser beam and D is its diameter.

The results of a calculation of d_0 and $2l_0$ for the lenses employed are shown in Table I. We consider first the data for the lens with F = 200 mm. As seen from Table I, in this case the parameter in the focal region is $\beta = 0.8$, i.e., according to[7], a stationary state has time to become established towards the end of the triangular pulse.

The self-focusing threshold power K[7] is determined in the stationary case by the expression

$$K = \frac{c\lambda^2\rho_0 v^2}{8\pi n_0(\rho_0 \partial n/\partial\rho)^2}, \qquad (4)$$

where c is the speed of light, λ the wavelength, ρ_0 the density, and n the refractive index.

For glass of the crown type[2)], calculation yields $K \approx 1$ MW. Since in our case the exciting power is ~25 MW, an intense shockwave should develop in the focal region at such a large excess over threshold, leading to heavy damage. Indeed, as seen from Table I, the length $2l_1$ of the region of intense damage is close to the length $2l_0$ of the focal region, and exceeds it slightly. The latter can be attributed to the fact that for a given lens the stationarity condition ($\beta \leq 1$) is satisfied in a region somewhat exceeding the focal region. In the regions l_2 and l_3, the initial beam diameter is larger, the stationary SF state does not have time to become established, and consequently the threshold power increases. At a fixed laser power, this leads to a decrease of the shock-wave intensity. In addition, when this wave reaches the beam axis (at a time after the termination of the laser pulse), its intensity has been decreased by attenuation. It is therefore natural to expect less damage in these regions, as is indeed observed in the experiments. The abrupt change in the track diameter apparently separates the region where the wave collapses even during the time of the pulse from the region where it collapses already after the termination of the pulse. At the employed laser power, the SF threshold due to electrostriction sets in at a parameter value $\beta \approx 3.5$, which in our case gives a permissible value of the beam diameter $d_c = 1.7$ mm. Table I gives the calculated values of the lengths $2l_c$ corresponding to this threshold value (see Fig. 1). As seen from Table I, the value of $2l_c$ is close to the experimentally observed $l_2 + l_3$.

In the case of the lens with F = 500 mm in the focal region we have $\beta = 2$, and consequently the stationary state is not reached. The total length where the power exceeds threshold (i.e., the length up to $d_c = 1.7$ mm), which equals $2l_0 + 2l_c$, amounts to 170 mm. Since in this case the excess of the power over the threshold value is small and varies little, we can expect a thin filament to appear (the shock wave is weak and, in covering the long path to the axis after the end of the pulse, it attenuates). As seen from Table I, the experimental results agree with those expected, but the reason for the formation of several filaments is not yet clear. It is possible that this is due to the presence of higher modes which are not completely suppressed.

In the case of a lens with F = 90 mm, in the region of the focus the parameter is $\beta = 0.4$, i.e., the stationary state is reached at approximately half of the pulse duration, and since the excess of power above threshold is large, damage of larger diameter is produced. An estimate of the length of the SF region $2l_c$ (to a value $d_c = 1.7$ mm) also gives a result close to that obtained by experiment (Table I).

2. It has been found that the divergence of the laser beam greatly influences the shape of the damage, and this must be taken into account when comparing the experimental results of different workers. For example, at a divergence ~10′ damage produced with a lens of F = 90 mm took the form shown in Fig. 1b, while that with lens F = 200 mm took the form of Fig. 1c, and in the latter case only one filament was produced. If it is recognized that the parameter β assumes for such a divergence the values 0.8 and 1.6 respectively, then these changes can readily be explained from the point of view considered above. The observed track lengths are also in good agreement with the calculations.

With further increase of the divergence, the picture changed qualitatively, this being apparently connected with the large role of the transverse higher-order modes, and accordingly with the complication of the field distribution in the region of the focus. In particular, with the F = 90 mm lens, an increase of beam divergence up to ~15′ has led to the occurrence in the focus of an ordinary ellipsoidal damage with relatively short filament (10—15 mm), elongated in the direction of the laser. At a divergence ~30′ (as in[2]), only the usual ellipsoidal damage was observed. In some cases, at a divergence 15′—30′, the damage produced in the focal region had the form of individual points (from one

[2)]The calculation was made in [7] for VK-7 crown (American nomenclature). We do not know the exact values of the elastic constants for K-8 crown, but we may expect the properties of these glasses to be similar.

to three or four). This is apparently connected with the fact that in this case some special relation arose between the higher transverse modes (only the divergence was monitored in the experiments, and not the fine structure of the beam).

3. A similar damage picture was also observed in the other investigated glasses. Their tendency to self-focusing, however, i.e., the possibility of observing filament-like tracks at a large beam divergence, or at a lower power, or else with a shorter-focus lens, was different. In accordance with the tendency to self-focusing, the investigated samples can be arranged in the following order: fused quartz (no SF is observed), LK-5, K-8, TF, and plexiglas. The theoretically calculated threshold powers given in[7] for three types of glass—fused quartz, crown, and heavy flint—follow the same sequence.

4. SMBS was observed in all the investigated samples. The results of the determination of the displacement of the SMBS components and of the hypersound velocities calculated from them are given in Table II. As seen from this table, the velocities coincide in all the cases, within the limits of experimental error, with direct ultrasonic-measurement data.

The results confirm the conclusion drawn in[2] that the velocity of the hypersound does not depend on the presence or absence of SF or on the focal length of the lens, and the results of[8] are apparently in error[3)]. We must stop to discuss specially fused quartz. The results of our measurements (both SMBS and ultrasonic) coincide with the universally accepted value (5980 m/sec), and also with the ultrasonic measurements in[2]. At the same time, the velocity determined from SMBS in[2] turned out to be different, 5585 m/sec. The discrepancy greatly exceeds the possible measurement error and calls for further investigation.

5. We estimated the intensity of the SMBS lines. It turned out that the SMBS intensity depends strongly on the width of the laser line. With increasing line width from ~0.03 to ~0.05 cm^{-1}, the intensity of the SMBS line, other conditions being equal, decreased by almost one order of magnitude. No amplitude modulation of the pulse was observed at the resolving power of our control apparatus (~15 nsec)[4)].

If it is assumed that the attenuation α of the hypersound in glass is of the same order as in quartz (and this is evidenced, in particular, by the proximity of the SMBS "thresholds" in them), i.e., $\alpha \sim 300$ cm^{-1}, then the total line width $2\Delta\nu$ of spontaneous scattering

$$2\Delta\nu = \alpha v \qquad (5)$$

will be of the order of 0.001 cm^{-1}, i.e., much narrower than the excitation line. In this case we can expect a strong dependence of the SMBS intensity on the excitation line width, as was apparently indicated for the first time by[11]. A detailed quantitative study of this question is worthy of special consideration.

Table II. Shift of SMBS component and speed of sound in glasses

Sample	$\Delta\nu$, cm^{-1}	v_{MB}, m/sec	v^*_{us}, m/sec	v from published data
Fused quartz	0.824 ± 0.013	5930 ± 90	6000 ± 30	5980 [14] 5804 [18] 5585 [2]
K-8	0.870 ± 0.013	6040 ± 90	6090 ± 50	5961 [2]
TF	0.642 ± 0.010	4090 ± 60	4020 ± 40	—
LK-5	0.846 ± 0.015	5950 ± 100		—
Plexiglas 10	0.400 ± 0.015	2820 ± 100		2870 [15] 2680 [14]

*v_{us}—velocity as determined from ultrasonic measurements.

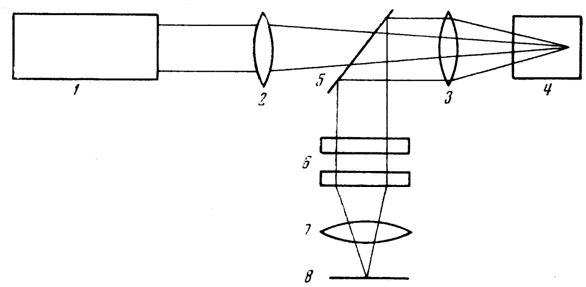

FIG. 2. Block diagram of modified setup. 1—Ruby laser, 2—lens F_1, 3—lens F_2, 4—sample, 5—beam-splitting plate, 6—Fabry-Perot interferometer, 7—objective, 8—photographic film.

6. We estimated the change of the SMBS line intensity I_{MB} as a function of the focal distance of the employed lens. It turned out that I_{MB} changes little and is in most cases maximal at F = 90 mm (with the exception of fused quartz, when the intensity is maximal at F = 20 mm). It could be assumed that for long-focus lenses, owing to the increase of the divergence in SF, only part of the energy of the scattered SMBS component is returned to the focusing lens. To verify this assumption, the experimental setup was modified somewhat (Fig. 2). The laser beam was focused into the sample by two lenses in tandem, long-focus F_1 and short-focus F_2. The distance between the lenses was somewhat smaller than F_1. With such an arrangement, the optical strength of the entire system was determined mainly by F_1, but the scattered diverging beam was completely gathered by F_2 and entered the recording system. The observed intensity of the SMBS components indeed increased, but usually did not exceed the intensity with F = 90 mm.

As is well known[12,13], for SMBS in the stationary case (at a pulse duration 40 nsec the process can certainly be regarded as stationary) we have

$$I_{MB} = I_{eq} \exp I_0 g l, \qquad (6)$$

where I_{eq} is a certain coefficient, I_0 the intensity of the exciting radiation, g the gain determined by the parameters of the medium, and l the length of the interaction region.

If we disregard SF and assume in first approximation that the interaction occurs only in the focal region, then $l = 2l_0$, $I_0 = P/S$, where P is the laser power, and

[3)]We note, incidentally, that the ultrasound velocity in glasses determined in [9], with the results of which the data of [8] are compared, has no bearing on the problem in question. In [9] the velocity was measured at 60 kHz, i.e., the so-called rod velocity was measured, and not the velocity of sound in free space.

[4)]The comparatively large line width in the near-single-mode regime is possibly partly connected with the change of the frequency during the pulse. As noted in [10], in a ruby laser the change of frequency amounts to ~ 10 MHz/nsec, i.e., more than 0.01 cm^{-1} over the entire pulse duration. At a spectrum width 0.05 cm^{-1}, apparently, several close longitudinal modes were generated and were not resolved by our recording apparatus.

$S = \pi d_0^2/4$ is the area of the focal region. Using (1) and (2), we can readily show that

$$I_{MB} = I_{eq} \exp \frac{8(\sqrt{2}-1)}{\pi} \frac{gP}{\vartheta D}. \quad (7)$$

Consequently, in this approximation the SMBS intensity does not depend on the focal distance of the lens and this, in general, is in agreement with the experimental results.

On the other hand, if we assume that the SMBS process occurs in an interaction region determined by the track dimensions (i.e., for example, for F = 500 mm, l = 100 mm, $d_0 = 10^{-3}$ mm), the exponent increases by several orders, and this should lead to a strong increase of the line intensity (even if the saturation is disregarded). This, however, is not observed in experiment.

7. When short-focus lenses were used (F ≤ 200 mm), the observation of the SMBS was always accompanied by damage to the sample. With a lens having F = 500 mm, it was sometimes possible to register SMBS without damaging the sample, using a weaker beam. But such a regime is very critical, a small increase causing damage to the object, and a small decrease causing a drop in the SMBS level below the registration threshold. Using the two-lens system (Fig. 2), we succeeded in reliably registering SMBS with a weakened beam without damaging the sample. The results are in qualitative agreement with the theoretical calculations. Indeed, as shown by (7) above, the intensity (or the experimental "threshold") of the SMBS is independent in first approximation of the focal distance of the lens, whereas the threshold for the occurrence of the self-focusing shock wave increases with increasing focal distance, owing to the increase in the diameter of the focal region[7]. It is clear that when the focal length of the lens increases, an instant should arise at which the SMBS is still registered, but the SF threshold is not reached, and consequently no filament-like damage is produced. Some quantitative deviation from the results of[2], where the SMBS was registered without damage even with a lens of F = 180 mm, can readily be explained as being due to the fact that the short pulse duration in[2] greatly raised the SF threshold, while lowering the SMBS intensity only insignificantly.

8. We note that in plexiglas the SMBS is extremely easy to observe, and frequently two or even three excitation components are observed in succession. As is well known[14], the damping of ultrasound in plexiglas is larger by many orders of magnitude than, say, in quartz, and, consequently, in accordance with the theory[12], the SMBS threshold should be high. The experimentally observed low threshold can apparently be attributed to the fact that the absorption relaxes. Such an explanation is confirmed also by observation of narrow lines in spontaneous Mandel'shtam-Brillouin scattering, as reported in[15].

DISCUSSION OF RESULTS

The obtained experimental results and their comparison with the results of other experimental and theoretical[7,12] investigations make it possible to describe, in first approximation, the main processes occurring in glasses under the influence of a laser pulse, as follows: At the initial instant of time, the cross section of the beam in the focal region has the form shown in Fig. 1d. A short time (~5 nsec) later, SMBS is produced in the region $2l_0$ of maximum flux density. At the same time, on the boundary of the volume occupied by the field, where the gradient is the largest, electrostriction produces a shock wave that tends to compress the beam. In the case of long-focus lenses, when $d_0/2\tau v \gg 1$, this process does not affect the SMBS strongly, and the observed thin filament-like damage is produced already after the end of the pulse, as a result of the collapse of the shock wave. Practically no self-focusing of the optical beam occurs here, having no time to develop, although the tracks have a diameter ~1 μ and a large length. At a low pulse power, the SF threshold is not reached at all, making it possible to register SMBS without damaging the sample.

In the case of lenses with medium focal length, when $d_0/2\tau v \sim 1$, the influence of the self-focusing is much stronger. A decrease in the area of the focal spot as a result of the contraction of the beam leads to an increase in the flux density and in the SMBS intensity. However, at the instant of the collapse of the shock wave, damage is produced, and the SMBS generation ceases. If photograph registration is used, this leads to a decrease in the observable intensity of the SMBS component, and the net change in the intensity, determined by the competition between these two processes, is negligible. Damage outside the focal region occurs after the termination of the pulse, and it is smaller, owing to the smaller amplitude of the collapsing shock wave.

In the case of short-focus lenses, $d_0/2\tau v < 1$, a similar process takes place, but the collapse occurs earlier, and this can stop the propagation of the energy beyond the focus and lead to development of a track mainly on the laser side. For lenses with very short focal lengths, $d_0/2\tau v \ll 1$, the length of the region where the energy exceeds the SF threshold is of the order of its diameter, the field gradients are large in all the directions, and the almost-spherical shock wave which is produced in this case causes, when it collapses, a strong ellipse-like damage. With such short-focus lenses, the SMBS intensity is decreased as a result of the short lifetime of the undamaged region.

The described mechanism apparently explains incontrovertibly the damage and SMBS processes observed in glass. It is much more difficult to explain the results from the point of view of the "moving focus" model[6]. In particular, major difficulties are encountered when attempts are made to explain tracks extending beyond the focus, and also when it comes to identifying the causes of the jump-like change in the track diameter. It is also difficult to explain the small change in the SMBS intensity on going from short-focus lenses, which produce no SF, to long-focus ones. Indeed, if, for example, F = 90 mm and $2l_0$ = 0.7 mm (and assuming that the latter quantity does not change when the focus moves, although in fact one can expect it to decrease), then at a track length $l \sim 20$ mm the duration of the irradiation of each section of the track is of the order of $(2l_0/l)(\tau/2) \sim 0.75$ nsec (for a triangular pulse). For such durations, the SMBS can no longer be regarded as stationary[12], and consequently the observed SMBS intensity should decrease sharply, as is observed experimentally in[16].

The results show also that the processes of multiphoton absorption can hardly play a decisive role in the damage process. These processes have practically no inertia, and therefore should greatly decrease the intensity of the SMBS component.

It can thus be assumed that both our results and the results of a number of other experimental investigations[2,3] can be explained, in main outline, as being due to self-focusing processes resulting from electrostriction, the dynamic theory of which has been developed in[7]. At the same time, the experimental results obtained in[17], which are easy to interpret from the point of view of the moving-focus model, are difficult to explain from the point of view of the mechanism described above, provided they are not the consequence of a complicated mode structure of the beam. A final clarification of this question calls for further experimental research.

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