

STIMULATED ENTROPY (TEMPERATURE) SCATTERING AND ITS EFFECT ON STIMULATED MANDEL'SHTAM-BRILLOUIN SCATTERING

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Stimulated temperature scattering of light (STS) and stimulated Mandel'shtam-Brillouin scattering (SMBS) are studied experimentally in liquids. In pure liquids, STS due to the electrocaloric effect is observed; in light-absorbing liquids, STS due to direct light absorption is observed. The effect of STS on the positions of the SMBS components is also studied. It is shown that, due to this effect, the shift in the SMBS components may differ from that determined on the basis of thermal scattering data.

INTRODUCTION

IN TENSE light in a material medium produces various nonlinear optical effects; among them is stimulated entropy or temperature scattering of light (STS),^[1,2] the features of which have not yet been sufficiently studied.

In the STS phenomenon, nonlinear interaction of intense exciting light of frequency ω_0 with initially weak light of thermal entropy scattering of frequency ω_1 gives rise to "temperature waves" at the difference frequency $\Omega = \omega_1 - \omega_2$. As a result of the nonlinear interaction of the waves mentioned, the energy of the incident light is "pumped over" into the scattered light and into the "temperature wave." There exist two different physical causes of the nonlinearity that produces the stimulated entropy scattering. The first is the electrocaloric effect; the second is the absorption of light by the medium and thus the transformation of light energy into heat. If the fundamental role in the scattering process is played by the electrocaloric effect, and the light absorption is so small that it can be neglected, then the maximum intensity in the scattered light spectrum will be displaced in the Stokes direction relative to the frequency of the exciting radiation ω_0 . We shall call such scattering STS - I. If light absorption plays a dominant role in the STS process in comparison with the electrocaloric effect, the maximum intensity will be displaced in the anti-Stokes direction relative to ω_0 , and we shall call such a process STS - II.

Zaitsev and the authors of this article^[1] have previously shown the existence of the STS phenomenon in pure benzene, but it was not demonstrated which of the two types of STS just described was observed by them. Both phenomena were observed in our subsequent researches. The phenomenon of STS-I was detected in gases^[3,4] and pure liquids.^[5] We also first observed STS-II in pure liquids.^[5] The first observation of STS-II in absorbing liquids was made by Rank et al.^[6], and in absorbing gases by Wiggins et al.^[7]

We report here the results of an experimental study of STS in liquids following variation in the intensity and absorption coefficient of the scattering light. It is shown that an increase in the light absorption coefficient leads to a transition of the phenomenon STS-I into

STS-II. The effect of STS on stimulated Mandel'shtam-Brillouin scattering (SMBS) is also studied. It has been established that, as a result of such an STS effect, the maximum intensity of the SMBS line can change its position. Consequently, the speed of hypersound, which is determined from the location of the SMBS components, can differ from its real value under certain experimental conditions, on either the larger or the smaller side.

A study of the causes of a shift in the position of the SMBS components is important also from the purely practical viewpoint, inasmuch as the SMBS phenomenon finds ever increasing application to the measurement of the speed of hypersound in various media.

1. FUNDAMENTAL THEORETICAL RELATIONS

On the basis of the coupled nonlinear equations of the electromagnetic field, hydrodynamics and temperature conductivity, the phenomena of STS-I, STS-II, and SMBS and their mutual effect in the steady state, and also the character of the development of STS-I and STS-II for large amplification coefficients in the non-stationary regime, have been considered by one of us.^[8-10] The amplification coefficient of STS-I in the steady state was obtained previously by one of the authors,^[11] and the amplification coefficient of STS-II and SMBS by Herman and Gray.^[12] Some problems of STS-II and its effect on SMBS were developed in other investigations (see^[13-21] for example). Formulas will be given below which follow from the theory developed in^[8-10] in a form that is suitable for discussion of the experimental results of the present research.

In the steady state, the amplification coefficients of STS (g_T) and SMBS (g_{MB}) have the following form:

$$g_T = g_{T1} + g_{TII} = \frac{|k_1| Y^2 \beta_s (\gamma - 1)}{16\pi n^2} \frac{\Omega \delta \Omega_c}{\Omega^2 + (\delta \Omega_c + \delta \omega_0)^2} \left(1 + \frac{1}{h}\right) |E_0^m|^2,$$

$$g_{MB} = g_{MB}^0 + g_{TMB} = \frac{|k_1| Y^2 \beta_s}{32\pi n^2} \left\{ \frac{\Omega_{MB} (\delta \omega_0 + \delta \Omega_{MB})}{(\Omega - \Omega_{MB})^2 + (\delta \Omega_{MB} + \delta \omega_0)^2} \right\} \quad (1)$$

¹⁾In [8] one should have $(\partial \epsilon / \partial T)_\rho$ and $\gamma \chi$ in Eqs. (5) and (10) for $(\partial \epsilon / \partial T)_\rho$ and χ , respectively. Equation (16) in [8] is incorrect; the correct formula is given in [9, 10].

$$+ \left(1 + \frac{1}{h}\right) \frac{\delta\Omega_c(\Omega_{\text{MB}} - \Omega)(\gamma - 1)}{(\Omega - \Omega_{\text{MB}})^2 + (\delta\Omega_{\text{MB}} + \delta\omega_0)^2} \} |E_0^m|^2. \quad (2)$$

The following notation is used in (1) and (2):

$$\gamma = \frac{C_p}{C_v}, \quad \delta\Omega_c = \chi q^2, \quad h = \left(\frac{\partial e}{\partial T}\right)_p \frac{T_0 \delta\Omega_c}{4k_\omega n c}, \quad Y = \left(\rho \frac{\partial e}{\partial \rho}\right)_T,$$

ρ is the density, $\beta_S = (\rho V_S^2)^{-1}$, V_S is the adiabatic sound speed, $q = k_0 - k_1$; k_0, k_1 are wave vectors corresponding to the excited and scattered light waves, χ is the temperature conductivity coefficient, $2k_\omega$ the light absorption coefficient, T_0 the equilibrium value of the temperature, c the speed of light in a vacuum; $2\delta\omega_0, 3\delta\Omega_{\text{HB}}$ are the half-widths of the lines of exciting radiation and thermal Mandel'shtam-Brillouin scattering. The steady state of STS and SMBS is reached within the times

$$t \geq j_T \tau_T, \quad t \geq j_{\text{MB}} \tau_{\text{MB}},$$

where t is the time elapsed after the beginning of stimulated scattering, $j_T = mg_T L$, $j_{\text{MB}} = g_{\text{MB}} L$ are the increments of the corresponding form of scattering in the steady state, L is the length of the region of non-linear interaction, τ_T and τ_{MB} are the damping times of the intensity of the temperature and acoustic waves, $\tau_T = 1/2\chi q^2$, and $\tau_{\text{MB}} = 1/2\alpha V_S$.

The expression for g_T consists of two components. One of them owes its origin to the electrocaloric effect ($g_{\text{TI}} \sim (\partial\epsilon/\partial T)_p^2$), and the other to the effect of light absorption ($g_{\text{II}} \sim k_\omega (\partial\epsilon/\partial T)_p$). If the liquid is so transparent that

$$k_\omega \ll |\epsilon| (\partial\epsilon/\partial T)_p |T_0 \delta\Omega_c / nc,$$

i.e., $g_{\text{II}}/g_{\text{TI}} \equiv 1/h \ll 1$ and, consequently, light absorption can be neglected, then the maximum intensity of STS will be located on the Stokes side relative to ω_0 (STS-I). In the opposite case of large k_ω and for $(\partial\epsilon/\partial T)_p < 0$ (for example, liquids), the maximum intensity of STS will be on the anti-Stokes side relative to ω_0 (STS-II). For $(\partial\epsilon/\partial T)_p > 0$, the effect of STS will always be shifted to the Stokes side.

We also represent the amplification coefficient g_{MB} in the form of two components. The maximum of one of them (g_{MB}^0) is identical with the frequency of the thermal Mandel'shtam-Brillouin scattering. The maximum of the second component (g_{TMB}) is shifted, depending on the value of k_ω , to the Stokes (k_ω large) or anti-Stokes (k_ω small) side relative to the frequency of the thermal scattering components by the amount $\pm(\delta\Omega_{\text{MB}} + \delta\omega_0)$.

The amplification coefficient g_{TMB} and the corresponding type of scattering can be called "mixed". Such scattering for large k_ω was observed in the work of Pohl et al.^[19]

The ratios of the maximum values of the absorption coefficients indicated above are expressed by the following formulas:

$$\frac{g_T}{g_{\text{rMB}}} = 2 \frac{\delta\Omega_{\text{MB}} + \delta\omega_0}{\delta\Omega_c + \delta\omega_0}, \quad (3)$$

$$\frac{g_r}{g_{\text{MB}}^0} = \left|1 + \frac{1}{h}\right| (\gamma - 1) \frac{\delta\Omega_c (\delta\Omega_{\text{MB}} + \delta\omega_0)}{\Omega_{\text{MB}} (\delta\Omega_c + \delta\omega_0)}, \quad (4)$$

$$\frac{g_{\text{TMB}}}{g_{\text{MB}}^0} = \frac{1}{2} \left|1 + \frac{1}{h}\right| (\gamma - 1) \frac{\delta\Omega_c}{\Omega_{\text{MB}}}. \quad (5)$$

The critical value $(2k_\omega)_{\text{cr}}$ is determined from the condition $g_T = g_{\text{MB}}^0$.^[12] Since $|h| \ll 1$ for this case, we get from (1) and (2)

$$(2k_\omega)_{\text{cr}} = \left| \frac{\partial e}{\partial T} \right|_p \frac{T_0 \Omega_{\text{MB}} (\delta\Omega_c + \delta\omega_0)}{2n^2 c (\gamma - 1) (\delta\Omega_{\text{MB}} + \delta\omega_0)}. \quad (6)$$

Consequently, for $2k_\omega = (2k_\omega)_{\text{cr}}$, the amplification coefficients of STS-II and SMBS should be equal.

As was mentioned previously, since g_{MB} consists of two components whose maxima are not of identical frequency, the SMBS line of frequency Ω_{MB} can have a satellite $\Omega_S = \Omega_{\text{MB}} - (\delta\Omega_{\text{MB}} + \delta\omega_0) \cdot \text{sign}(1 + 1/h)$.²⁾ For $|h| \ll 1$, when k_ω is large, this satellite can be even more intense than the fundamental SMBS line of frequency $\omega_0 - \Omega_{\text{MB}}$ if the condition

$$2k_\omega > (2k_\omega)_{\text{cr}} = \left| \frac{\partial e}{\partial T} \right|_p \frac{T_0 \Omega_{\text{MB}}}{n^2 c (\gamma - 1)}. \quad (7)$$

is satisfied.

In the case when the intensity of the SMBS lines and the lines of the mixed form of scattering T-SMBS exceed the detecting threshold, but the same lines are not resolved by the spectral apparatus, the appearance of T-SMBS should lead to a decrease, of the shift of the maximum of the total contour SMBS and T-SMBS at $(1 + 1/h) > 0$, and to an increase in this shift at $(1 + 1/h) < 0$ (k_ω large). Consequently, the speed of hypersound determined from the location of the maximum of the total contour of SMBS and T-SMBS will differ from the true value.

The intensity of the SMBS can be so great that the SMBS light will undergo repeated scattering as a consequence of STS. This leads to the appearance, along with the SMBS line of frequency Ω_{MB} ; of a line of frequency $\Omega_S = \Omega_{\text{MB}} + (\delta\Omega_{\text{MB}} + \delta\omega_0) \cdot \text{sign}(1 + 1/h)$. For STS-I ($|h| > 1$), this line will be displaced in the Stokes direction and for STS-II, in the anti-Stokes direction relative to the frequency Ω_{MB} . Consequently, in this case, for the same sign $(1 + 1/h)$, the STS effect will lead to a displacement of the visible (unresolved by the apparatus) total contours of the lines of frequency Ω_{MB} and Ω_S , in the opposite direction to the case when SMBS and T-SMBS are observed.

The lines that arise as a result of repeated scattering because of STS of the SMBS components will be more intense than the T-SMBS line when $g_{\text{TE}}^2 \text{E}_{\text{MB}}^2 > g_{\text{TMB}}^2 \text{E}_0^2$, which corresponds, with account of (3), to the condition

$$\frac{E_{\text{MB}}^2}{E_0^2} > \frac{1}{2} \frac{\delta\Omega_c + \delta\omega_0}{\delta\Omega_{\text{MB}} + \delta\omega_0}. \quad (8)$$

In the nonstationary regime, at large amplification coefficients and a rectangular shape of the pulse of

²⁾It follows from (5) that the electrocaloric effect ($h \rightarrow \infty$) has an insignificant effect on the value of the amplification coefficient in liquids. All the formulas given here were obtained with neglect in the initial equations of the quantity $(\partial\epsilon/\partial T)_p$.^[8-10] With account of terms containing $(\partial\epsilon/\partial T)_p$ practically the same quantitative values are obtained for the amplification coefficients as in the formulas given here, because of the smallness of $(\partial\epsilon/\partial T)_p$ relative to $(\partial e/\partial T)_p$. The conclusion of Enns and Rangnekar^[22] of the significant (~50%) effect of the electrocaloric effect on the value of g_{MB} is the result of an incorrect inference that $(\partial\epsilon/\partial T)_p \approx |(\partial e/\partial T)_p|$ for liquids. At the same time, for all liquids except water, $|\partial\epsilon/\partial T|_p$ is greater by an order of magnitude than $(\partial e/\partial T)_p$ (see^[23]).

exciting radiation, the STS intensity changes according to the law^[8]

$$I_1(L, t) \sim \exp \{ (2|B_T|E_0^2 L t)^{1/2} \}, \quad (9)$$

where

$$B_T = \frac{|k_x| Y \beta_s (\gamma - 1)}{16\pi n^2} \left(Y \delta \Omega_c - \frac{4k_a n c}{T_0 \sigma} \right).$$

Broadening of the scattered light in this regime is determined by the expression^[8]

$$\Omega'(t) \approx \frac{1}{2} (|B_T| L t^{-1} |E_0|^2)^{1/2} \text{sign } B_T. \quad (10)$$

Consequently, the spectral broadening of the scattered light in the nonstationary regime is Stokesian for $B_T > 0$ ($|h| > 1$) and anti-Stokesian for $B_T < 0$ ($|h| < 1$) and depends on the intensity of the exciting radiation, and at $|h| < 1$ also on the light absorption coefficient. Inasmuch as the real pulse, beginning at the moment $t = 0$ ($\Omega' \rightarrow \infty$), terminates at $t = t_0$ ($\Omega' = \Omega'(t_0)$), it follows that the spectrum of scattered light in the nonstationary region is a band, and the edge of which that is nearest the exciting line is distant by an amount $\sim \Omega'(t_0)$.

2. EXPERIMENTAL SETUP

Stimulated scattering was excited by a giant pulse from a ruby laser with maximum power ~ 180 MW. The distance between the laser and the scattering volume was so large that there was no feedback between them. The experimental setup for the study of the spectral composition of light of stimulated scattering is shown in Fig. 1.

Linearly polarized light with vector \mathbf{E} , directed along the vertical z axis, was focused by the cylindrical lens L_0 with a focal length $f = 3$ cm into a cell filled with the liquid under study. To find the small ($5 \times 10^{-3} - 10^{-2} \text{ cm}^{-1}$) spectral shift of the STS line relative to the frequency of the exciting light, the spectra of the exciting and scattered light were photographed simultaneously on a single plate so that they could easily be compared with each other. For this purpose, part of the exciting light was diverted from the main beam by the divider S and passed through a crystalline quartz plate $P_k(\lambda/2)$ and polarizer N . Thus the direction of polarization of this comparison beam made an angle of 90° with the direction of polarization of the fundamental beam of the exciting laser radiation.

The combined beams of light scattered at an angle of 90° (z polarization) and of light of the comparison beam (x polarization) were directed through a light filter and frosted glass plate P_m into the interfero-

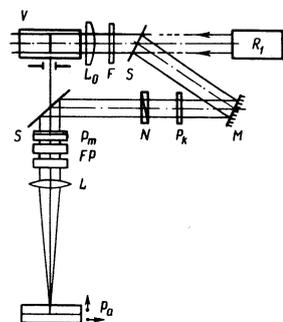


FIG. 1. Scheme of apparatus for study of STS and SMBS spectra at the scattering angle of $\vartheta = 90^\circ$: R_1 —ruby laser, L_0 —cylindrical lens, V —cell with liquid, F —central light filter, P_k —half-wave plate, N —polarizer, P_m —frosted plate and interference light filter, FP —Fabry-Perot interferometer, L —objective, P_a —analyzers, S —divider, M —mirror.

metric apparatus. A Fabry-Perot interferometer was used as the spectral apparatus, with dispersion ranges 0.25 and 0.5 cm^{-1} . Two analyzers P_a were placed in front of the photoplate, so oriented that vertically polarized scattered light was incident on one half of the interferogram and horizontally polarized light of the comparison beam was incident on the other.

It should be specially noted that, in order to avoid a "false" shift of the STS line from the line of the exciting light, all the experimental results in the STS-I and STS-II radiation were obtained by us for a fixed geometry of the setup. Furthermore, it should be kept in mind that the system of analyzers worked satisfactorily only in the case of not very high intensity of the light. Therefore, for comparison of the location of the STS line and the line of the exciting radiation, it is necessary to choose such an intensity that only exciting light was recorded on one half of the interferogram and only scattered light on the other. In those cases in which the SMBS intensity was very large, weak components of SMBS were also recorded on that half of the interferogram which corresponded to the exciting light. Consequently, we could carry out measurements of the shifts of the SMBS components in such a case, both relative to the exciting light and relative to the STS line.

3. INTENSITY DISTRIBUTION OF THE LASER RADIATION AT THE FOCUS OF THE CYLINDRICAL LENS

As has been mentioned above, the maximum power of radiation of the laser was ~ 180 MW, the duration 15 nanosec and the spectral width $\sim 10^{-2} \text{ cm}^{-1}$. If this radiation had been smoothly distributed over the cross section of the beam (for example, in Gaussian fashion), then one would have expected a mean intensity $\sim 10^{10} \text{ W/cm}^2$ at the focus of the cylindrical lens. Here, under the conditions of our experiment (the STS process is nonstationary) the STS-I intensity could be less than the recording threshold. But actually, the radiation of the laser was nonuniform over the cross section; therefore, several regions of elevated intensity could be isolated in the cross section, with several square millimeters in each. Intensities in these regions of the focus ought to be several times as large as the mean intensity.

We have studied the intensity distribution in the focus of a cylindrical lens in transmitted light, projecting the different regions of its focus on a screen with a spherical lens. Shifting this lens together with the screen, we could establish the character of the intensity distribution along the direction of propagation of the laser radiation. Such investigations showed that there are several regions of increased intensity in the focal zone of a cylindrical lens of thickness (in the vertical direction) $\sim 10 - 20 \mu$, which are rectangles (located in the horizontal plane) with an area up to 10 mm^2 with approximately equal sides. Consequently, the intensity of the exciting light in these regions can reach $\sim 2 \times 10^{11} \text{ W/cm}^2$.

Observation of regions of increased intensity, with the dimensions given above, at the focus of a cylindrical lens gives an indication, in our opinion, of the development of one-dimensional self-focusing, under the conditions of our experiment.

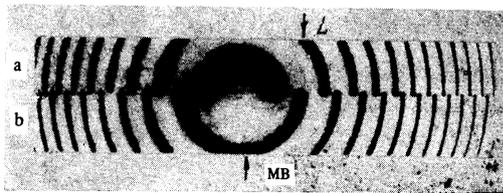


FIG. 2. Interferograms: a—exciting light, b—light scattered at an angle $\vartheta = 90^\circ$ in pure water; L—line of exciting radiation, MB—SMBS component. The dispersion range of the Fabry-Perot interferometer is 0.25 cm^{-1} .

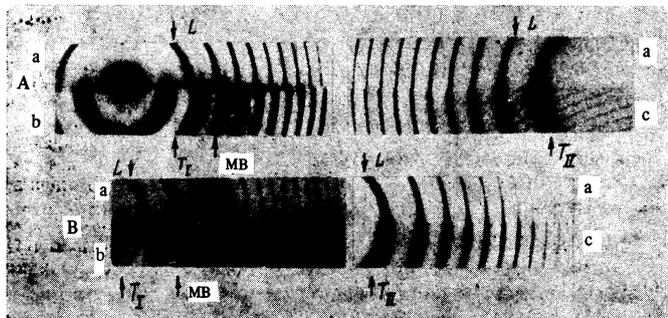


FIG. 3. Interferograms of exciting and scattered light: A—in benzene, B—in ethyl alcohol ($\vartheta = 90^\circ$): a—exciting radiation, L—line of laser radiation; b—scattered radiation in pure liquids, T_I —STS-I line (Stokes shift); b—scattered radiation for the same cases with the addition of light absorption (iodine), T_{II} —STS-II line (anti-Stokes shift). Dispersion range of interferometer— 0.25 cm^{-1} .

4. STIMULATED TEMPERATURE SCATTERING OF LIGHT IN LIQUIDS

The phenomenon of STS was studied in benzene, ethyl alcohol, and water by means of the arrangement described in Sec. 2. In pure water, an intense component of SMBS is observed in the spectrum of scattered radiation, corresponding to a scattering angle $\vartheta = 90^\circ$, and the central component is missing (Fig. 2). This result is easily understood if we take it into account that γ in water is close to unity and, in accord with (1) and (4), the amplification coefficient of STS should be near zero.

For a power of exciting radiation $W \sim 80 \text{ MW}$ in carefully purified benzene³⁾ and $W \sim 180 \text{ MW}$ in absolute ethyl alcohol, STS lines were observed that were shifted in the Stokes direction relative to the line of exciting radiation. This shift is quite evident in the interferograms, shown in Figs. 3a and 3b. Consequently, under these conditions, we observed STS-I, due to the electrocaloric effect.

Upon the addition of iodine into these liquids as an absorber and with other conditions unchanged, lines appeared in place of the exciting radiation, shifted in the anti-Stokes direction relative to the line of the exciting radiation; consequently, STS-II was observed in this case (Fig. 3c). For small light absorption coefficients, the shift of the STS-II line was of the same order as the shift in the STS-I line. For large light-absorption coefficients, close to critical, and high intensities of the exciting light, the STS-II line be-

³⁾Chemically pure benzene was subjected to additional purification by the method of Martin (see [23]).



FIG. 4. Interferograms of exciting (upper) and scattered (lower) light in pure benzene ($\vartheta = 90^\circ$) for high intensity of the exciting light. L—line of laser radiation, T_{II} —STS-II line. Dispersion range of interferometer— 0.25 cm^{-1} .

came wider and its shift (more properly, the location of the center of the line) increased several fold.

The STS-I line in pure benzene was observed at a power of the exciting radiation $\sim 50\text{--}80 \text{ MW}$. Upon increase in the power, the STS-II line with an anti-Stokes shift was observed in place of the STS-I line (Fig. 4). Evidently, at high intensities of the exciting radiation, nonlinear light absorption begins to grow and STS-II develops in pure liquids, replacing STS-I.

As was already mentioned above, STS-I was observed in benzene at a power of laser radiation $\sim 80 \text{ W}$ and in ethyl alcohol at a power $\sim 150\text{--}180 \text{ MW}$. Taking into account the intensity distribution in the focus of the lens, we estimated that the intensity of the exciting radiation was $\sim 10^{11} \text{ W/cm}^2$ for observation of STS in benzene, and $\sim 2 \times 10^{11} \text{ W/cm}^2$ in ethyl alcohol. In this case, the estimates for STS-I ($2k_\omega \sim 0$), from the formulas of Sec. 1, of the stationary value of the increment $j_{st} = gT L$, the nonstationary value of the increment j_{nst} under the conditions of our experiment, the time of establishment of the steady state t_{st} and the distance to the STS-I band $\Delta\nu' = \Omega'(t_p)/2\pi c$ for $L = 1 \text{ cm}$ and $t_0 = 15 \text{ nanosec}$, give the following values for benzene:

$$j_{st} = 25, \quad j_{nst} = 7, \quad t_{st} = 3.8 \cdot 10^{-7} \text{ sec}, \quad \Delta\nu' \sim 0.9 \cdot 10^{-3} \text{ cm}^{-1}$$

and for ethyl alcohol

$$j_{st} = 16, \quad j_{nst} = 4.6, \quad t_{st} = 3.6 \cdot 10^{-7} \text{ sec}, \quad \Delta\nu' \sim 0.6 \cdot 10^{-3} \text{ cm}^{-1}$$

Thus, in both liquids, for a pulse length $\sim 15 \text{ nanosec}$, the phenomenon of STS ought to be nonstationary. The calculated values of j_{nst} and $\Delta\nu'$ show that under our experimental conditions, the possibility of photographic recording of the STS-I line and of the determination of the direction of the shift of this line is at the limit of sensitivity of the apparatus. Actually, very weak STS-I lines were observed, while, as mentioned previously, there was not only a lower recording threshold, but also an upper (STS-II) was generated. The latter circumstance further complicates the observation of this phenomenon. The observed shift of the STS-I line $\sim 3\text{--}5 \times 10^{-3} \text{ cm}^{-1}$ is greater than the calculated distance to the band of scattered light, as it ought to be from the very definition of these quantities. However, this difference is greater than one ought to expect from the estimates, although we do not consider it possible to give a real value to this difference, in view of the low accuracy of the calculation and of the experimental determination of the position of the STS-I line.

For STS-II, the process of establishment of the temperature was determined not only by the temperature

conductivity of the medium, but also by the thermalization time τ .^[13] However, in liquids for not too small scattering angles $\tau \ll (\chi q^2)^{-1}$, the thermalization time can be neglected and the shifts of the STS-I and STS-II lines should be the same for equal amplification coefficients.

For an increase in the amplification coefficient of STS in the nonstationary state, the shift of the STS line ought to increase in accord with formula (10). In just this way, we explain the increase we observed in the shift of the STS line for increase in the intensity or light absorption coefficient. This effect was noted by us previously in^[5].

5. EFFECT OF THE PHENOMENON OF STIMULATED TEMPERATURE SCATTERING OF LIGHT ON STIMULATED MANDEL'SHTAM-BRILLOUIN SCATTERING

In our first researches^[24,25] and in many subsequent ones (see^[2]) it was noted that the shift of the SMBS components in liquids and gases is not identical with the shift calculated from the spectrum of thermal Mandel'shtam-Brillouin scattering (TMBS). Usually the shift of the SMBS components is less than that calculated from the spectrum of thermal scattering, and the reason for such a difference remained unclear. The initial assumption that this difference arises in liquids as a result of the change in the speed of hypersound for heating of the scattering medium by the intense laser light was not correct, inasmuch as a decrease in the shift of the SMBS components has also been observed in gases and in water, where $\partial V/\partial T > 0$.

In the observation of back scattering, there ought not to be aperture effects, for example, nonsymmetric intensity distribution of SMBS over the aperture, since, even in the observation of scattered radiation for an angle differing from $\vartheta = 180^\circ$ by $\Delta\vartheta = 15^\circ$, the displacement of the Mandel'shtam-Brillouin component ought to change by only 1%. It was shown experimentally by Brewer^[26] that in the case in which there is reverse coupling between the laser and the scattering medium, a "contraction" of the SMBS components takes place in the neighborhood of the mode of the laser resonator and, consequently, the location of the recorded components of SMBS can differ from the real by half the distance between the modes of the resonator.

However, this effect ought to lead to a decrease with the same probability as an increase in the shift of the SMBS components, while only a decrease is usually observed. Moreover, the decrease in the shift of the SMBS components has been observed even in the absence of reverse coupling between the laser and the scattering medium.

Goldblatt and Litovitz^[27] have shown that in research with a single-mode laser and in the absence of reverse coupling, shifts of SMBS components in gases are obtained which do not differ from those calculated. Madigosky et al.^[28] assumed that in the liquid under such conditions, the shifts of the SMBS components should be equal to that calculated. In these researches, the opinion was made clear that the reason for the change in the visual location of the SMBS components is usually the multimode structure of the laser radia-

tion. In the opinion of these authors, only the most intense modes stand out in the scattered radiation, and the rest do not appear; of the location of the SMBS components is produced from the total contour of the exciting radiation, which includes all modes, and which leads to an incorrect result.

Apropos of this, one can make the following remark. Reduction of the shift of the SMBS components was also recorded in experiments with single-mode lasers. Evidently, the single-mode structure of the radiation can actually lead to an incorrect result in the measurement of the shift of the SMBS components. However, it has been shown theoretically by D'yakov^[29] that weak modes of laser radiation can stand out in the scattered radiation, contrary to the opinion of the authors of^[27,28].

A large number of effects which owe their origin to the high intensity of the exciting radiation can influence the location of the SMBS components to a large or small degree (see, for example, ^[2,30,31]). A complete discussion of these problems goes beyond the limits of the present paper.

We have subjected to experimental test the supposition of one of the authors of this paper^[8] that the STS phenomenon has a significant effect on the location of the SMBS components (see Sec. 1). With this aim, we studied benzene, acetone, carbon tetrachloride and water. Scattering was observed at an angle of 90° to the direction of propagation of the exciting light. In all liquids, without exception, very intense SMBS components were recorded, the displacement of which corresponded to a scattering angle of 90° . The intensity of this component is such that in acetone, carbon tetrachloride, and water, it could excite subsequent SMBS components (multiple scattering), but with a displacement corresponding to a scattering angle of 180° . Increase in the light absorption coefficient leads to a decrease in the intensity of the SMBS components and to a simultaneous increase in the intensity of the STS-II line. These changes of intensity are clearly seen in the interferograms of the scattered light in benzene, shown in Fig. 5.

Figure 6 shows the values of the hypersonic speeds in benzene as a function of the light absorption coefficient $2k_\omega$, found from the shifts of the SMBS components, measured from the frequency of the STS line and from the frequency of the scattered light. In the same place is given the hypersonic speed, computed from the shift in the Mandel'shtam-Brillouin components, according to the thermal scattering spectrum. It

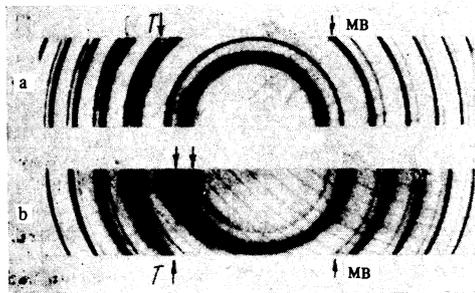


FIG. 5. Interferograms of scattered light in benzene ($\vartheta = 90^\circ$): a—scattering for $2k_\omega < 10^{-2} \text{ cm}^{-1}$; b—for $2k_\omega = 0.16 \text{ cm}^{-1}$; T—STS line, MB—SMBS line. Dispersion range of interferometer— 0.5 cm^{-1} .

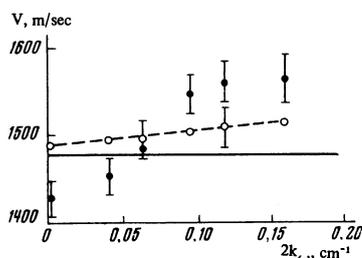


FIG. 6. Dependence of the hypersonic speed in benzene, determined from the SMBS spectrum, on the light absorption coefficient: ●—measurement made from the STS line, ○—measurement made from the exciting line; continuous curve—speed computed from the shift of the Mandel'shtam-Brillouin line in the thermal scattering spectrum.

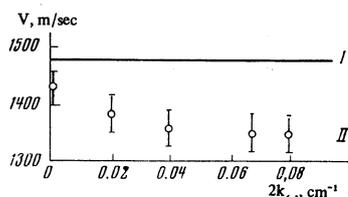


FIG. 7. Dependence of the hypersonic speed calculated from the shift in the SMBS components on the light absorption coefficient in water—circles, and of the speed calculated from thermal scattering spectrum—continuous line.

is seen from the drawing that the location of the SMBS line, measured from the scattering line within the limits of accuracy of the experiment, is identical with the results calculated from the thermal scattering spectrum. In measuring from the STS line, a shift is obtained that is smaller, and in an absorbing liquid, greater, than the shift of the Mandel'shtam-Brillouin line in thermal scattering. The resultant experimental data for benzene show that if an STS line appears in the data and the measurement of the shift of the SMBS component is made from the STS line or from the maximum of the total (unresolved) shape of the exciting line and the STS line, the result of such a measurement will differ from the measurement made from the thermal scattering spectrum.

The SMBS components in the liquid, for not very small $2k_\omega$ (but also not too large, so that the SMBS intensity be large) can itself produce STS-II as a consequence of repeated scattering at the angle $\varphi = 180^\circ$ in the region of nonlinear interaction. This effect should lead to a decrease in the distance between the exciting lines and the maximum of the total (unresolved) shape of the line of "pure" SMBS and the SMBS line, undergoing repeated STS-II.

In benzene at the powers used (~ 80 MW), the SMBS intensity was evidently insufficient for this effect to appear (see Fig. 6, open circles). In acetone, water, and carbon tetrachloride, SMBS began at high intensities and the SMBS increment was greater than in benzene, which is indicated by the appearance of the components of repeated SMBS. The intensity of the SMBS components in these liquids was clearly sufficient to produce the recorded STS-II phenomenon for $2k_\omega \sim 10^{-2} \text{ cm}^{-1}$ at the scattering $\varphi = 180^\circ$. The experimental results obtained for all three liquids was identical and can be illustrated by the case of water (Fig. 7). The reading of the shift of the SMBS component was

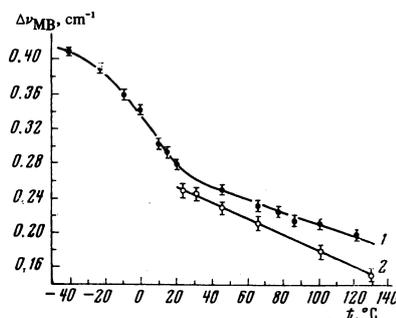


FIG. 8. Temperature dependence of the shifts of the components of 1—thermal and 2—stimulated Mandel'shtam Brillouin scattering in salol.

made from the line of the exciting radiation. In highly purified liquids, the shift of the SMBS components is almost identical with the data on the shift of the components in the thermal scattering spectrum. For an increase in $2k_\omega$, the shift of the SMBS components decreases, which is evidently connected with the superposition on the curve of the line of pure SMBS of the STS-II line produced by the SMBS. For further increase in $2k_\omega$, the intensity of the SMBS decreases sharply and it has not been possible to follow the change in the shift.

The results obtained in the measurement of the shifts of the SMBS components in salol and benzophenone can be explained in the same way.

Figure 8 shows the temperature dependence of the shifts of the components, calculated from the SMBS spectrum in liquid salol. In these experiments, focusing of the laser radiation was carried out by spherical lens ($f = 5$ cm), the SMBS was studied at an angle $\varphi = 180^\circ$ the reverse coupling between the laser and the scattering medium was not removed and special purification of the liquid was not carried out. Therefore, one can assume that the STS-II mechanism occurred in this liquid as a result of the large value of $2k_\omega$.

For comparison, the temperature dependence of the shifts of the components, calculated from the thermal scattering spectrum of $f^{[32]}$, is also shown in Fig. 8 for comparison.

As is seen from Fig. 8, the shifts of the SMBS components and the components of thermal scattering are practically identical for salol in the temperature region $t \sim 20^\circ \text{C}$.

It follows from the experimental data of $f^{[32]}$ that the hypersonic absorption coefficient α in salol is maximal in this range of temperatures; consequently, the minimum intensity of the SMBS components and the effect of STS on the location of the SMBS components in this case is evidently insignificant. For higher temperature, α is small, the intensity of SMBS is large and the STS produced by it leads to an apparent decrease in the shifts of the SMBS components. The lack of coincidence of the shifts of the components from the SMBS spectrum and the thermal scattering spectrum in benzophenone which we observed can be explained in the same fashion.

Thus, all our investigations of the departure of the shifts of the SMBS components from their value in thermal scattering in benzene (STS-I and STS-II), in water, acetone, carbon tetrachloride (STS-II) under

various conditions of experiment, can be understood if we take into account the effect of STS produced by the exciting radiation or by SMBS radiation.

We note one more circumstance. It follows from (2) that for SMBS, in addition to the line at frequency Ω_{MB} there can be a thermal Mandel'shtam-Brillouin line, shifted relative to Ω_{MB} by an amount $(\delta\Omega_{MB} + \delta\omega_0)$. Under the conditions of our experiment, the effect of thermal Mandel'shtam-Brillouin did not appear because of the fact that the amplification coefficient $g_{TMB} \ll g_{MB}^0$ and $g_{TMB} \ll g_T$. Under other circumstances, a complicated picture can arise because of the lines of different intensity (SMBS, STS and thermal Mandel'shtam-Brillouin) which are not resolved with an interferometer of average dispersion range, but the maximum of the unresolved contour is shifted one way or the other. In connection with the fact that a whole series of factors play a role (the values of the intensities I_0 and I_{MB} , the amplification coefficient, absorption and so on), the analysis of the final result can be seen to be difficult. As a result, although the effect of STS on the intensity and location of the SMBS components can be regarded as established, careful analysis of the experimental conditions is necessary in each specific case.

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