Phase fluctuations of the Stokes wave produced as a result of stimulated scattering of light

N. G. Basov, I. G. Zubarev, A. B. Mironov, S. I. Mikhaĭlov, and A. Yu. Okulov
P. N. Lebedev Physics Institute, USSR Academy of Sciences

(Submitted May 6, 1980)
Pis'ma Zh. Eksp. Teor. Fiz. **31**, No. 11, 685–689 (5 June 1980)

The presence of a phase irregularity during a laser pulse in the Stokes component radiation produced as a result of stimulated Mandel'shtam-Brillouin scattering in active media with a short acoustic phonon lifetime was directly demonstrated by using an interferometer with mirrors for wavefront inversion.

PACS numbers: 78.35. + c, 78.45. + h, 07.60.Ly

1. Investigation of the phase fluctuations of the Stokes wave is important not only for understanding the mechanism of stimulated scattering of light and, in particular, the reconstruction of the time-dependent structure of quasi-monochromatic pumping, but also for a whole series of practical applications, since this process determines the limiting monochromaticity of laser radiation frequency converters that use stimulated Raman scattering of light.[1-3]

A theoretical examination of the noise-induced stimulated scattering[4] shows that the line width of the Stokes component $\Delta\nu_S$ is determined by the narrowing of the spontaneous line $\Delta\nu_l$ is a result of amplification

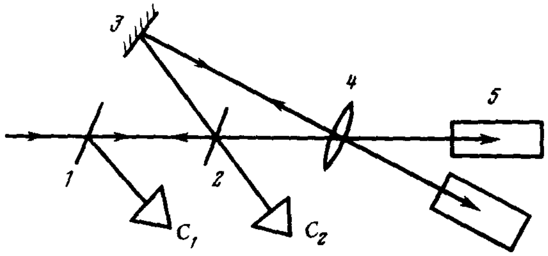


FIG. 1. Experimental setup: 1, glass plate; 2, semitransparent mirror; 3, mirror with $R = 100\%$; 4, lens; 5, cells with active material.

$$\Delta\nu_S \approx \Delta\nu_l/\sqrt{G}\,, \tag{1}$$

where G is the amplification increment at the threshold power of the exciting radiation equal to ~ 25. This process has not been adequately investigated experimentally. Only several papers, in which the line widths for stimulated Raman scattering were measured, have been published. However, the conditions under which these studies were made did not always permit unambiguous interpretation of the results. We shall, therefore, refer to Ref. 5 which showed that the relation (1) is not satisfied; the line width of the Stokes component, which was found to be narrower than that obtained by using Eq. (1), approximately reproduced the line of the exciting radiation. In contrast to Eq. (1), this additional line narrowing is attributable to a parasitic feedback due to a variety of reasons, which were mentioned in the literature.[6]

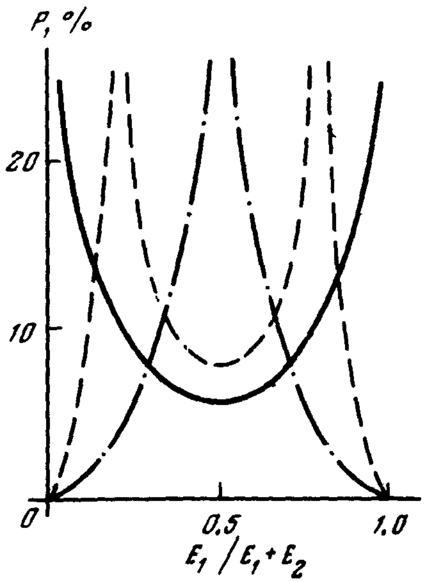
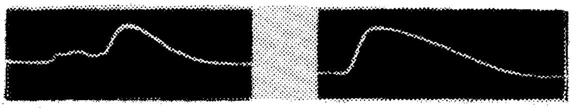


FIG. 2. Probability densities of the normalized pulse energy in one of the recording channels for different models of the phase behavior: phase difference of the scattered waves during the pulse is constant ———, one phase irregularity at the end of the pulse ----; one phase irregularity in the middle of the pulse -·-·-·-



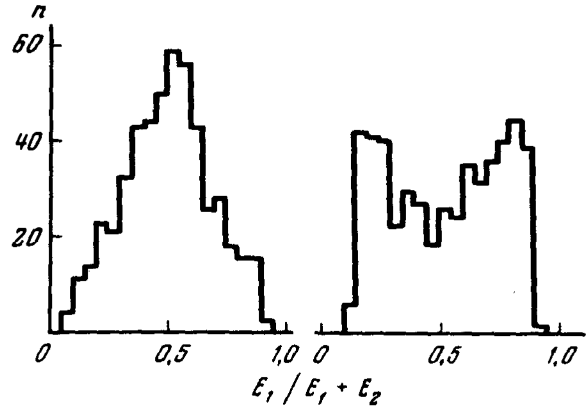


FIG. 3. Experimental distribution of the pulses with a given normalized energy for carbon tetrachloride (on the left-hand side) and compressed nitrogen (on the right-hand side). Oscillograms of the scattered radiation for these active media are shown in the insets.

We have directly observed the phase fluctuations of the Stokes radiation due to stimulated Mandel'shtam-Brillouin scattering. To investigate the line structure of the scattered light for monochromatic pumping, we developed a basically new procedure for direct measurements of the characteristic time of the phase irregularity in the Stokes wave.

2. The proposed method is based on the use of a Michelson-type, doubled-beam interferometer with wavefront inverting SMBS mirrors. The experimental apparatus is shown schematically in Fig. 1. The exciting radiation from a single-frequency laser source (an yttrium-aluminum garnet laser was used with passive Q-switching and mode selection) with a lasing line width $\Delta\nu_p \approx 1\times 10^{-3}$ cm^{-1} was split by a semitransparent mirror into two beams, which were reflected from the independent SMBS mirrors with an inversion of the radiation wavefront. A wavefront inversion ensures an ideal adjustment of the interferometer even when the pumping beams are spatially nonuniform. During the experiments, we measured the energies of the Stokes radiation that was reflected and transmitted by the semitransparent mirror, and the time-dependent structure of the Stokes signal was also recorded before and after it passed through the semitransparent mirror. The experiments were performed at a constant energy of the laser pumping pulse, which corresponded to a four-fold increase above the threshold power for the SMBS process. The energies of the beams directed to different calorimeters C_1 and C_2 were determined by the initial phase difference of the beams and by its time-dependent behavior. In the normalized form for the first calorimeter, for example, we obtain:

$$\frac{E_1}{E_1 + E_2} = \frac{1}{2T} \int_{-T/2}^{T/2} \{ 1 + \cos[\Delta\phi(t)] \} dt, \quad T \to \infty . \tag{2}$$

Since the reflection occurs from independent SMBS mirrors, the initial phase difference $\Delta\phi(0)$ of the distribution is completely random. Figure 2 shows the calculated probability density distributions of $E_1/(E_1 + E_2)$ for three different models of the time-dependent behavior of the phases $\Delta\phi(t)$. Thus, by measuring the relative number of $E_1/(E_1 + E_2)$, we can determine which model matches the experimental case. We note that the quantity (2) ceases to fluctuate almost completely from one laser flash to another when the number of irregularities of the relative phase difference is large during a pulse and becomes equal to 0.5 (the distribution approaches a Gaussian shape with a dispersion $[2(1 + k)]^{-1}$, where k is the number of irregularities per pulse). It can be shown that in the saturation mode which was realized in these experiments, the line width of the reflected Stokes signal is determined by the random phase modulation,[4] and the characteristic phase variation time, according to Eq. (1), is:

$$\tau_{\Delta\phi} \approx \sqrt{G}\, \tau_{\text{hyp}} , \tag{3}$$

where τ_{hyp} is the decay time of the hypersound. In our case $G \approx 25$ and $\tau_{\Delta\phi} \approx 5\tau_{\text{hyp}}$.

The cases in which the length of the laser pulse $\tau_p \approx 30$ nsec was less than the indicated time (the active medium was nitrogen with $\tau_{\Delta\phi} \approx 200$ nsec compressed at a pressure of 300 atm) and greater than the indicated time (the active medium was liquid carbon tetrachloride with ($\tau_{\Delta\phi} \approx 10$–20 nsec) were obtained experimentally.

3. Figure 3 shows the experimental distributions of the quantity (2). Each histogram is the result of a processing of 500 experimental points. It can be seen that in a compressed nitrogen the best approximation is provided by the model with a constant phase difference during the pulse, and in a liquefied carbon tetrachloride the best model is that for one phase irregularity in the middle of the pulse. The oscillograms of the backscattered pulse in any of the recording channels, shown in the insets of Fig. 3 confirm these conclusions. After reflection from nitrogen, the oscillogram of the scattered radiation remains smooth during the entire pulse. An average of one phase irregularity can be observed in carbon tetrachloride during this time. This oscillogram corresponds to the case when the beams that were initially reflected into a given recording channel are traveling nearly counterphase, and then the phase is shifted by π.

Therefore, the experimental value for the time of the characteristic phase irregularity is $\tau_{\Delta\phi} = 15$–20 nsec in the case of carbon tetrachloride, consistent with the value obtained from (3). Hence, for monochromatic exciting radiation the spectrum width of the Stokes component, which is determined by these phase discontinuities of the scattered wave, is numerically determined by Eq. (1). These results show that for stimulated scattering at the Stokes frequency these can be no reproduction of the spectrum of the monochromatic excitation of radiation in the media with a short phase irregularity time.

To determine the role of parasitic feedback, we modified to some extent the experimental setup in Fig. 1 by placing the substrates, which were adjusted in the normal direction to the pumping radiation in each of the two channels, between the focusing system and the carbon tetrachloride cells. We mounted the blind mirrors at the exit from the cells and placed the phase plates between these mirrors and the cells, which drastically increased the radiation divergence. The radiation loss during one complete traversal in these cavities was $I(2L)/I(0) \sim e^{-(6-7)}$. These cavities, nevertheless, reduced the threshold pumping intensity by a factor of 1.2 and increased the efficiency of reflection from the SMBS mirrors. A disruption of the pumping wavefront inversion mode was not observed. An analysis of the corresponding oscillograms and calorimetric measurement data showed that the expected narrowing of the Stokes radiation spectrum due to the feedback did not occur.

4. The results of the performed experiments enabled us to make the following conclusions: 1) for monochromatic pumping the spectrum width of the Stokes component of the SMBS is determined by the formula $\Delta\phi_S \approx \Delta\nu_l \sqrt{G}$, which describes the narrowing of the scattered radiation spectrum within the contour of the amplification line. 2) The mechanism for the line narrowing due to a weak feedback is missing for the SMBS process. 3) By using a weak feedback the SMBS threshold can be reduced and the reflection efficiency can be increased while preserving the inverting properties of the SMBS mirror.

In conclusion, we note that the interferometer with wavefront inverting mirrors used by us has a number of unique properties. In particular, if the light beams are inverted in different active media, then the oscillograms in the recording channel will have a sinusoidal modulation with a frequency equal to the difference between the frequency shifts of the Stokes components in each active medium. This can be utilized for a rapid recording of changes in the hypersound velocity during the laser pulse. A more detailed analysis of an interferometer with wavefront inverting mirrors will be published in another paper.

[1]A. Z. Grasyuk, I. G. Zubarev, A. V. Kotov, S. I. Mikhaĭlov, and V. G. Smirnov, Kvantovaya Elektron. **3**, 1062 (1976) [Sov. J. Quantum Electron. **6**, 568 (1976)].

[2]J. Cahen, M. Clere and P. Rigny, Opt. Commun. **21**, 387 (1976).

[3]S. M. Bierry, R. Frey, and F. Paradere, Rev. Sci. Instrum. **48**, 733 (1977).

[4]Yu. E. D'yakov, Pis'ma Zh. Eksp. Teor. Fiz. **10**, 545 (1969) [JETP Lett. **10**, 347 (1969)].

[5]A. N. Arbatskaya, K. A. Prokhorov, and M. M. Sushchinskiĭ, Zh. Eksp. Teor. Fiz. **62**, 872 (1972) [Sov. Phys. JETP **35**, 462 (1972)].

[6]B. Ya. Zel'dovich and V. V. Ragul'skiĭ, Kvantovaya Elektron. **4**, 927 (1977) [Sov. J. Quantum Electron. **7**, 525 (1977)].