Recording of phase fluctuations of stimulated scattered light

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The phase fluctuations were recorded experimentally and the spectral line width of the Stokes wave produced as a result of stimulated Mandel'shtam-Brillouin scattering (SMBS) was measured for the first time. A method of heterodyning the light of two mutually incoherent SMBS Stokes waves excited by neodymium laser radiation in optically isolated liquids, was used to do this. The oscillations of the current from the photodetector illuminated by these waves was recorded with a high-speed oscilloscope. The spectral line width of the stimulated scattered light in carbon tetrachloride was determined from the average time of the oscillations.

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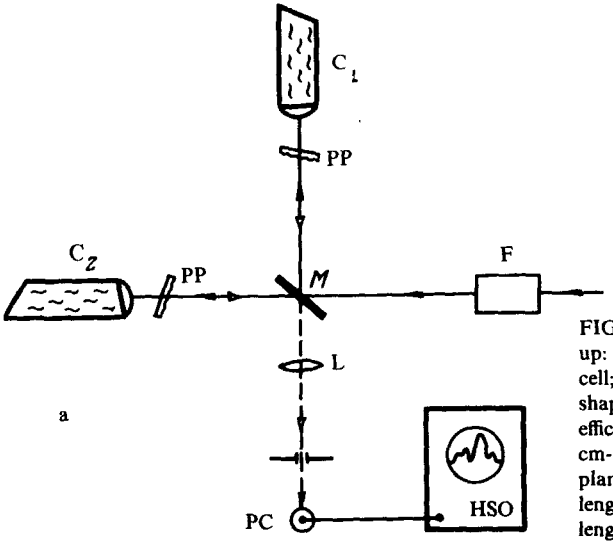
As is known, stimulated scattering is a process for amplifying spontaneously scattered light via the nonlinear interaction of excitation radiation with the medium. Spontaneously scattered light is *a noise* field that fluctuates in amplitude and phase with a characteristic frequency determined by the line width of the spontaneous scattering. It follows from the theoretical work[1,2] that the fluctuations should be greatly retarded and acquire a phase during amplification. However the phase fluctuations of stimulated scattered light heretofore have not been observed experimentally.

In our work we have recorded for the first time the phase fluctuations in the presence of stimulated Mandel'shtam-Brillouin scattering (SMBS) and on this basis determined the width of the SMBS radiation spectrum.

The recording was done by using the light heterodyning method,[3] which in our case consisted of recording the current by a square-law detector illuminated by two mutually incoherent beams of scattered light. No amplitude fluctuations were present in each of these beams, but the phase fluctuations produced photocurrent fluctuations as a result of mixing of the beams in the detector.[4] For such an experiment to be successful, the current fluctuations from different parts of the photodetector must be synchronized (see Ref. 3); to do this, the wavefronts of the beam must be identical. It was possible to achieve this by splitting the exciting light into two parts with identical wavefronts and their stimulated scattering with a wavefront inversion (WFI)[5] in two, optically isolated volumes of a nonlinear medium.

The experimental setup is shown schematically in Fig. 1. The linearly polarized radiation of a neodymium laser (one longitudinal mode, pulse length at half-height of 35 nsec, peak power of 20 MW, divergence of 3×10^{-4} rad), which was divided into two parts with equal power (in the first version—Fig. 1a—by the semitransparent mirror M, and in the second version—Fig. 1b—by a polarization wedge made from a birefringent material) and then directed to two identical cells filled with carbon tetra-



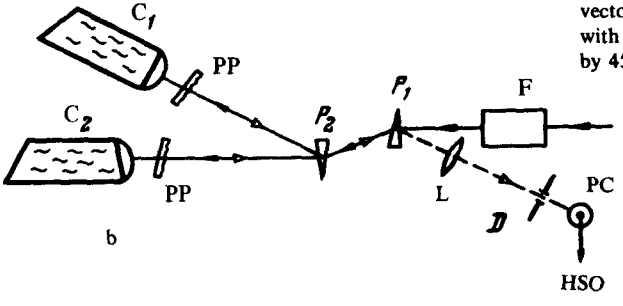


FIG. 1. Schematic of the experimental set-up: F, optical isolator based on a Faraday cell; M, plane dielectric mirror on wedge-shaped substrate with a 50% reflection coefficient; PP, phase plates; C_1 and C_2, 30-cm-long, 2-cm-diam cells; cell windows are plane-convex lenses with a 10-cm focal length (in air); L, lens with a 150-cm focal length; D, 0.7-mm diam diaphragm; PC, FK-19 coaxial photocell; HSO, I2-7 high-speed oscilloscope; P_1, Iceland spar wedge with a 8° vertex angle and an optical axis parallel to the direction of the polarization vector of laser radiation; P_2, spar wedge with a 15° angle and an optical axis rotated by 45° with respect to the wedge axis P_1.

chloride or acetone, was used to excite the SMBS. The SMBS Stokes waves from the two cells propagate in the opposite direction, are combined with each other beyond the beamsplitter element and strike the detector. In version 2, an elliptically polarized wave, whose one component is fed to the detector by using a polarization wedge P_1, is formed as a result of combining of waves with orthogonal polarization and a fluctuation difference in their phases. The photodetector was a coaxial photocell whose signal was recorded by means of a high-speed oscilloscope (the time resolution of the recording system was 1 nsec). The inverted component of the stimulated scattered light, into which $\approx 10\%$ of the energy of the exciting radiation was converted, was isolated with an angular selector (lens L and diaphragm D). To achieve WFI, we placed in front of the cells glass plates of nonuniform thickness, which introduced a phase distortion.

In view of the high amplification typical of SMBS, there is a real danger of the development of oscillations in each of the cells and of the interaction between them, which can lead to an attenuation of the fluctuations. To avoid this, special measures

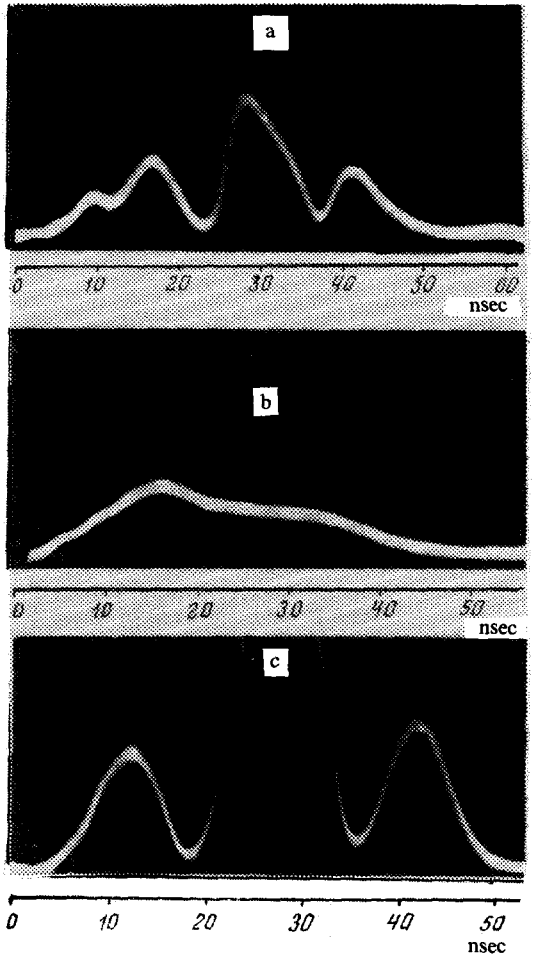


FIG. 2. Photocurrent oscillograms: a, the scattering is excited in carbon tetrachloride; b, the scattering is excited in acetone; c, beats of two Stokes waves with different average frequencies (the temperature of the cells with acetone differs by 3.8 °C).

were taken to eliminate the feedback in each cell as well as between them. There was no mirror coupling between the cells which were separated from each other by a distance of 5 m; this prevented a coupling between them during the time of the SMBS pulse ($\sim$30 nsec). The focusing lenses, which were the front windows of the cells, were canted at 5°; the rear windows were tilted at the Brewster angle. Glass absorbing filters (not shown in the figure) were placed behind the cells and tilted at the Brewster angle. Phase plates, which were also tilted, were placed 5 cm in front of the cells. The presence of dust in the scattering materials was monitored.

The temperature of the scattering liquid greatly influenced the average frequency of the scattered light (the frequency shift was 10–15 MHz/deg).[6] Therefore, to ensure average equal frequencies of the radiation scattered in the different cells, the cell temperatures were equalized within an accuracy of 0.1 °C. For the same reasons, we used the phase plates, which increased the divergence of the excitation beams to at least 2×10^{-3} rad, were used to reduce heating of the medium due to absorption of

light in it. The plates, fabricated from one hydrofluoric, acid-etched piece of glass, had identical characteristics. To prevent the frequency drift of the laser radiation, which occurred during a flash and amounted to 10 MHz/nsec,[7] from affecting the measurement results, we equalized the optical path lengths from the splitter to the cells within an accuracy of 2 cm. As a result of the path equalization, the influence of local heating of the medium during a flash was also eliminated. After each flash the system was thermally stabilized within a few minutes. SMBS occurred in the saturation mode (the energy reflection coefficient was 20%) in which, as is known, the total gain, which is stabilized, depends slightly on small fluctuations in the amplitude of the exciting radiation. These measures made it possible to prevent the appearance of false photocurrent pulses (such pulses, which were observed when the cell temperatures differed by 3.8°, are shown in Fig. 2c).

Figure 2a shows a typical photocurrent oscillogram for scattering in CCl_4. To interpret it properly, we must remember that the spectral density of the photocurrent fluctuations is a convolution of the spectral densities of the two waves illuminating the photocell (see, for example, Ref. 8). It follows from the estimates[1,2] that the spectral intensity of the stimulated scattered light is proportional to

$$\exp\left[\frac{G}{1+\left(\frac{2\Delta\nu}{\delta\nu_0}\right)^2}\right] \equiv e^G \exp\left[-G\left(\frac{2\Delta\nu}{\delta\nu_0}\right)^2 + \frac{G^2\left(\frac{2\Delta\nu}{\delta\nu_0}\right)^4}{G+G\left(\frac{2\Delta\nu}{\delta\nu_0}\right)^2}\right]. \tag{1}$$

Here G is the gain coefficient at the maximum of the SMBS line, where under typical conditions $G \approx 25$; $\Delta\nu$ is the frequency deviation from the center of the line, and $\delta\nu_0$ is the spectral line width of the spontaneous scattering. We can easily prove that the second term on the right-hand side of Eq. (1) affects only the far wings of the line, which contain only a small fraction of the scattered light. Therefore, we can assume that the shape of the spectral line of the stimulated scattered light is close to Gaussian $\exp[-G(2\Delta\nu/\delta\nu_0)^2]$ with a width at the 1/2 level

$$\delta\nu_S = \sqrt{\frac{\ln 2}{25}}\,\delta\nu_0. \tag{2}$$

Hence, the spectral density of the photocurrent fluctuations must also have a Gaussian shape with a width

$$\delta\nu_i = \sqrt{2}\,\delta\nu_S. \tag{3}$$

On the other hand, the width of the spectrum of the photocurrent fluctuations is related in a simple way to their average duration measured at half height (τ_i):

$$\delta\nu_i = \frac{2\sqrt{2}\ln 2}{\pi\tau_i}. \tag{4}$$

We obtain from Eqs. (3) and (4)

$$\delta \nu_S = \frac{2 \ln 2}{\pi \tau_i} . \quad (5)$$

The value of τ_i, which was determined from seven oscillograms, was 8.9 ± 2 nsec. Substituting this value in Eq. (5), we obtain $\delta\nu_S = 50 \pm 11$ MHz. A calculation according to Eq. (2), taking into account the known data for the width of the spontaneous line ($\delta\nu_0 = 420 \pm 60$ MHz for an excitation wavelength $= 1.06\ \mu m$[6]), gives a value of 70 ± 10 MHz for $\delta\nu_S$. Thus, our experimental results agree with the theoretical estimates. The photocurrent fluctuations were of longer duration as a result of scattering in acetone (Fig. 2b), because the spontaneous scattering line in acetone is 2.4 times narrower than that in CCl_4. A quantitative comparison with Eqs. (2) and (5) is difficult in this case, since the duration of the fluctuations is comparable to the duration of the excitation pulse.

For shorter excitation pulses

$$\left(< \frac{2\sqrt{25 \ln 2}}{\pi \delta \nu_0} \text{ in duration} \right)$$

special methods must be developed for recording the phase changes during one pulse. The phase instability in a series of such pulses with a large duty factor, should lead to a large spread in the energy measurements of the field produced as a result of superposition of the two, independently scattered waves, as observed in Ref. 9.

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