Spectral structure of stimulated Brillouin scattering

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The behavior of the phase of the light scattered in stimulated Brillouin scattering in CCl_4 has been studied experimentally. The fundamental shape of the scattering spectral line in the buildup regime has been determined. Instead of the theoretically predicted Gaussian emission spectrum, the results reveal a reproducible fine structure of the line in the form of an asymmetric triplet in the Stokes region under ordinary experimental conditions. © *1995 American Institute of Physics.*

The shape and width of the emission spectrum resulting from stimulated scattering was studied theoretically in Refs. 1 and 2. It was shown that the spectral line of spontaneous scattering, having a Lorentzian shape with a width at half-maximum $2\delta\nu$, transforms in the course of the amplification into a Gaussian curve

$$I_s(\nu) \propto e^{-G[(\nu-\nu_0)/\delta\nu]^2}, \tag{1}$$

where ν_0 is the frequency of the scattered light, which is shifted in the Stokes direction with respect to the exciting light, and G is the gain. In the experiments of Ref. 3 it was found that the line of stimulated Raman scattering detected in a single event consists of a set of a narrow lines. Only when the observed spectrum was averaged over numerous realizations was the result predicted theoretically found.[4] There has been no corresponding test of the theory for simulated Brillouin scattering (or "stimulated Mandel'shtam–Brllouin scattering"). Such an experiment is complicated by the relatively narrow spectral lines. Estimates put the widths of the lines in liquids in the interval $(0.3–3)\times 10^{-3}$ cm^{-1} (for exciting light with a wavelength of 1.06 μm). These values are right at the limiting resolution of standard spectral apparatus, and they are furthermore close to the linewidths of pulsed single-mode lasers.

The first measurements of the width of the fundamental spectrum of stimulated Brillouin scattering were carried out by the Ragul'skiĭ group.[5] The method they used was based on a mixing of signals of identical frequency which were scattered in CCl_4 and which underwent independent phase changes. In interpreting beats in the phase at a frequency of 50 ± 11 MHz, the authors made a deliberate effort to reconcile them with the existing theory.[1,2] In later studies, various types of broadening of the stimulated-Brillouin line were investigated. A broadening can be caused by, for example, a self-modulation resulting from absorption of the light in the medium[6] or a relaxation in instability of the process.[7]

In this letter we are reporting measurements of not only the intrinsic width, but also the shape of the spectral line of stimulated Brillouin scattering.

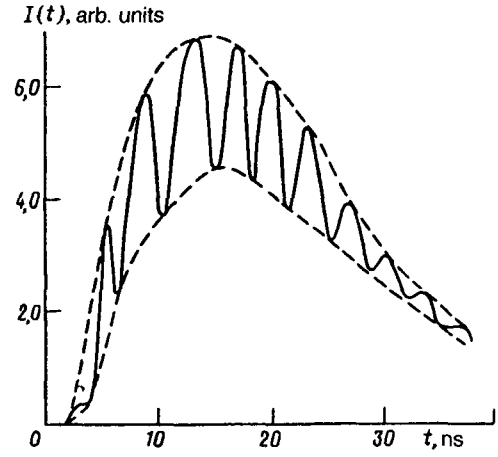

FIG. 1. Oscilloscope trace of the interference of two stimulated-Brillouin-scattering processes, in CCl_4 and $TiCl_4$. The dashed curves are the upper envelope $A(t)$ and the lower one $B(t)$ of the temporal modulation of the signal.

The experimental procedure which we used is based on a mixing of signals from two sources. The only difference between our approach and that proposed in Ref. 5 is that the frequencies of the signals to be mixed differ by an amount which is several times as great as the presumed width of the stimulated Brillouin line. In our experiments, the stimulated scattering was excited by light from a single-mode, single-frequency Nd-YAG laser ($\lambda = 1.064$ μm) with a pulse length ~45 ns at half-maximum and with an energy up to 50 mJ. The laser was decoupled from backscattered light by a Faraday shutter; the optical decoupling was ~40 dB. Some 60% of the energy was sent to one arm of a Michelson interferometer, which contained the test cell holding the CCl_4. The light was focused into this cell by lenses with focal lengths ranging from 5 to 15 cm. The reference wave was generated in the other arm of the interferometer, in a glass waveguide ~0.5 mm in diameter 7 cm long filled with $TiCl_4$. The stimulated-Brillouin signals from the two cells were mixed in a beam splitter and filtered by means of a diaphragm. They were then detected by an FK-26 photodetector and an S7-19 oscilloscope with a total bandwith well above 1 GHz. The beat signal was photographed and then digitized at a step of 0.2 ns and stored in computer memory.

Figure 1 shows the typical time evolution of the intensity of the detected signal. To determine the time evolution of the phase of the optical signal we used a method like that described in Ref. 8. The signal to be detected was described by

$$\frac{A-B}{2}\cos(\Omega t+\Psi(t))+\frac{A+B}{2}, \tag{2}$$

where A and B are the amplitudes of the upper and lower envelopes of the beats. The value of Ω was taken to be 1.6 rad/ns, the same in all cases. The value of $\Psi(t)$ was chosen by fitting the expression written above to the experimental data. For each value of t, this fitting was carried out over an interval of ~2.5 ns. The slight nonlinearity (~10%)

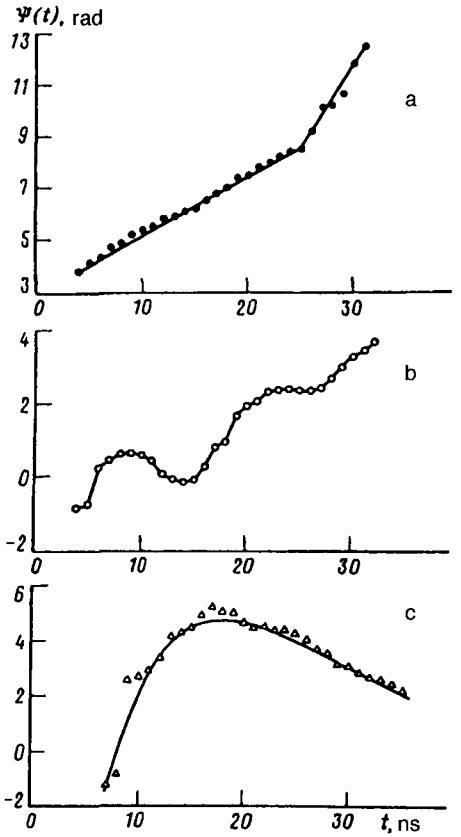

FIG. 2. Various versions of the dynamics of the phase of the reflected light. The probability that these versions occur decreases from a to c; part b shows the behavior of the phase $\Psi(t)$ in Fig. 1.

of the oscillscope sweep could have introduced some perceptible distortions in the measured phase. To eliminate errors of this sort, we calibrated the sweep with the help of a sine-wave generator at a frequency Ω_1. The phase of the calibration sine wave, $\varphi(t)$, multiplied by the frequency ratio Ω/Ω_1 was subtracted from the measured value of $\Psi(t)$ in each case.

Figure 2 shows a typical time evolutions of the phase. Most frequently the phase is proportional to the time over the entire pulse, although the proportionality factors vary from realization to realization. The slope of the $\Psi(t)$ curve often change abruptly during the pulse (Fig. 2a). Figure 2b corresponds to damped oscillations of the phase, which usually arose at the beginning of the stimulated Brillouin scattering and also after the change in the slope on the phase curve. Much less frequently, we observed a bell-shaped behavior (Fig. 2c), often accompanied by laser breakdown in the CCl_4.

A linear change in the phase in time corresponds to a certain frequency of the

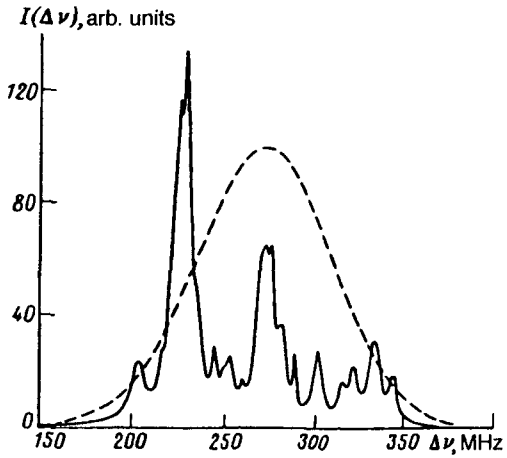

FIG. 3. Instantaneous spectrum of the beats, averaged over all realizations. The dashed curve is a theoretical prediction of the spectrum based on expression (3).

scattered light. A change in the slope of the plot of the phase as in Fig. 2a corresponds to a jump from one definite frequency of the scattered light to another. It thus turns out that the jumps in the phase which had been observed previously (see, for example, Refs. 5 and 8) reflect the abrupt nature of the change in the frequency shift of the stimulated scattering. The behavior of the phase in Fig. 2b apparently corresponds to a transition region in which the frequency approaches a quasisteady state (before the next abrupt change in frequency). In accordance with that interpretation, in those realizations in which it was meaningful to carry out a linear approximation of $\Psi(t)$ (if only a brief time interval) this approximation was indeed carried out. The results of all the experiments were summed, with allowance for the error in the determination of the frequency and the duration of the given approximation. These results are shown in Fig. 3.

To compare the spectrum of the heterodyne signal which was obtained with the theoretical spectrum, we will use some values of the half-width $2\delta\nu$ and of the Stokes shift $\Delta\nu_{si}$ of the lines of spontaneous scattering in the liquids used. According to the results of Ref. 9, the values of $2\delta\nu$ in CCl_4 and $TiCl_4$ are $\sim 528\pm 25$ and 216 ± 10 MHz, while the shifts of these lines are $\Delta\nu_{s1}\simeq 2770\pm 20$ and $\Delta\nu_{s2}\simeq 3070\pm 10$ MHz. Since the spectrum of the beat signal is determined by a convolution of two Gaussian spectra of different widths, this spectrum basically reflects the nature of the wider line, i.e., the line of the stimulated Brillouin scattering in CCl_4. Within some unimportant corrections, the spectral shape expected on the basis of Refs. 1 and 2 is

$$I_s(\Delta\nu)\propto e^{-G\frac{(\Delta\nu_0-\Delta\nu)^2}{\delta\nu^2}}, \tag{3}$$

where $\Delta\nu_0=|\Delta\nu_{s2}-\Delta\nu_{s1}|\simeq 300\pm 30$ MHz is the difference between the Stokes shifts in the two liquids, G is the total increment in the gain of the stimulated Brillouin scattering (usually ~ 25), and $\delta\nu$ determines the width of the spontaneous line in CCl_4. This line is shown by the dashed curve in Fig. 3 for the value $\Delta\nu_0=272$ MHz.

It would be natural to expect that, as in the case of stimulated Raman scattering, the frequency spectrum over many realizations would fill in the lineshape (1). However, this did not happen; the spectrum of stimulated Brillouin scattering found by the averaging procedure turned out to be triplet (Fig. 3). The intensities of the lines of the triplet are not distributed uniformly. A Fourier analysis of the behavior found shows that, in addition to the frequencies forming the triplet, which are separated by ~53 MHz, there are some weak modulations at frequencies ~24 and 15 MHz. It can be seen from Fig. 3 that the two latter frequency components are at the noise level, so we will not discuss them further.

A detailed study of the spectrum of the spontaneous line of the Brillouin doublet in liquids has revealed no fine structure of any sort in polarized light.[9] Accordingly, we should attempt to interpret such an unusual phenomenon on the basis of the nonlinear properties of stimulated Brillouin scattering. The explanation for the effect which we propose here incorporates the nonuniformity of the Doppler frequency shift of the light reflected from a nonlinear acoustic wave.

An intense sound wave has different propagation phase velocities in compression and rarefaction regions. Monochromatic light reflected from positive and negative antinodes of the sound acquires different Doppler frequency shifts. To evaluate this splitting, we switch to a coordinate system which is moving at the average sound velocity, v_0. We restrict the discussion to the first approximation for a "simple-wave" perturbation of the density (Ref. 10, for example):

$$\rho/\rho_0 \propto e^{i(qx-\alpha t \sin qx)-t/\tau}, \tag{4}$$

where q is the wave number, α is a nonlinearity parameter, proportional to the wave amplitude, and τ is the sound attenuation time. Taking an average over the period of the density perturbation, we find the temporal modulation of the scattering light: $E_s(t) \propto J_0(\alpha t) e^{-t/\tau}$, where J_0 is the Bessel function, $\alpha = \epsilon v q M$, M is the Mach number, and ϵ is a nonlinear constant of the medium ($\epsilon = 6.8$ in the case of CCl_4). We thus find the spectrum of the scattered light to be

$$E_s(\omega) \propto \frac{1}{[(1/\tau + i\omega)^2 + \alpha^2]^{1/2}}. \tag{5}$$

We see that for values $\alpha > 1/\tau$ the spectrum of the reflected light splits by an amount $\approx 2\alpha$. The value of α for a given pump-wave intensity I and for the corresponding coefficient of the transformation of the pump into the reflected wave, η, can be calculated from the equations of the linearized theory. Using an expression from Ref. 11 for the density perturbation in the course of stimulated Brillouin scattering, we find

$$\alpha = \frac{16\pi^2 Y n \tau \epsilon}{\rho_0 \lambda^2 c} I\sqrt{\eta}, \tag{6}$$

where $Y = \rho(\partial n^2/\partial\rho)$, and n is the refractive index. Estimates show that a splitting of the spectrum by an amount $\sim 2\delta\nu/\sqrt{G}$ (~100 MHz for CCl_4) would require a light intensity $I \sim 2.5 \times 10^{10}$ W/cm^2 in the interaction region. Such values of I are achieved in our case when a lens with $f = 10$ cm is used to focus light with an energy ~30 mJ.

We might add that in the course of the experiments the intensity was varied by a factor of more than 3. This variation had no effect on the position of the fine-structure lines. The stability of the shift can be explained on the basis of a competition between the splitting of the line and a decrease in the amplification of the scattered light as the frequency deviates from resonance. The observation of temporal changes in the spectrum implies that the central part of the spectrum is stable and that there is a mutual pumping of the wings of the line. These experimental results suggest that a dynamic analysis of the process is necessary in order to find an adequate explanation of the experimental data. We explain the absence of scattered light, which would fill the frequency intervals between peaks, on the basis that there is a change in the frequency of this light, which absolutely must arise. The temporal instability accompanying the decay of these photons is reflected in phase oscillations of the type in Fig. 2b. We will take a more detailed look at the dynamics of the spectrum in a separate paper.

It can be seen from Fig. 3 that the spectrum of the stimulated Brillouin scattering consists of three lines $\sim$10 MHz wide. Such a spectral resolution cannot be achieved either in a single pulse 45 ns long or as the result of a summation of the spectra of a periodic-pulse process. The analysis procedure described by Eq. (2) makes it possible to combine in a coherent way processes which occur in different measurement events.

We note in conclusion that the spectrum of the stimulated Brillouin scattering which we found was determined from values of the instantaneous spectrum of the scattered signal. Mathematically, the concept of a spectrum may differ appreciably from that of an instantaneous spectrum. In the integral used by us, the shapes of these spectra are essentially the same, because of the use of the heterodyne (instead of homodyne) reception. For the particular experimental procedure used by us, the instantaneous spectrum carries additional information about the time evolution of the stimulated scattering. A condition for the validity of this concept is that the heterodyne frequency be well above the interval of frequencies under study.

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