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PRECISE DETERMINATION OF STOKES SHIFTS IN STIMULATED-BRILLOUIN-SCATTERING-ACTIVE SUBSTANCES WITH THE USE OF BRILLOUIN LOOP LASERS PUMPED BY GAUSSIAN MONOCHROMATIC BEAMS

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Abstract

A method for determination of Stokes shifts in stimulated Brillouin scattering is suggested and implemented; the method is based on the jumplike behavior of the spatial structure of the emission of a Brillouin loop-scheme laser upon change in the loop length in the vicinity of $\Delta k_B L = \pi(2n+1)$, where Δk_B is the Brillouin shift, L is the loop length, and n is an integer.

1. Introduction

The development of nonlinear-optics methods makes it possible to suggest unconventional techniques for determination of characteristics of various materials. Thus, as far back as 1980 [1], we employed an interferometer equipped with a phase-conjugate mirror to determine the stimulated Brillouin scattering (SBS) shift $\Omega_B = 0.125\ \text{cm}^{-1}$ at a temperature $t = 20°$ C in carbon disulfide at the wavelength of a neodymium laser; this value of Ω_B is consistent with results of measurements by the methods of active laser spectroscopy [2]. We analyzed the possibilities of increasing considerably the accuracy of determination of SBS shifts without introducing any serious complications of experimental technique; we found the following major sources of errors in the corresponding measurements:

(I) the presence of an nonconjugated component in the signal reflected from the SBS mirror, with the amplitude of this component varying randomly from one laser burst to another;

(II) the sinusoidal character of the interference pattern, which makes it difficult to determine the positions of maxima (minima) with adequate accuracy owing to the gradual variation of the intensity.

As early as the first study of a loop-SBS laser [3] a periodic dependence of the loop-laser characteristics on the loop length was pointed out; this dependence is related to a non-reciprocal phase advance $\Delta\varphi$ in the resonator loop of the loop-SBS laser due to the difference in frequencies between the pump and the Stokes radiation. In fact, $\Delta\varphi = \Delta k_B L$, whence it follows that the characteristic period is $T = 1/\Omega_B$, where Δk_B is the difference in the magnitudes of the wave vectors of the interacting waves, Ω_B is the corresponding shift in reciprocal centimeters, and L is the loop length. It is this dependence that can be used for determination of the corresponding SBS shifts Δ_B in various active substances because $\Omega_B \simeq 0.1\ \text{cm}^{-1}$ for typical liquids at the wavelength of a neodymium laser and the loop length is $L \simeq 0.5 - 1$ m. With an integer number of periods fitting in the loop length, we obtain $\Omega_B = n/L$, where n is the number of periods. We studied such a laser [4] and showed that the radiation of the loop-scheme laser pumped by a Gaussian beam can include not only the main (central) mode but also angular-dependent modes; this hampers precise determination of the period T and, consequently, of the Brillouin shift. In this work, we employed the technique developed by us to measure experimentally the Brillouin shifts in carbon disulfide and titanium tetrachloride at a pump wavelength $\lambda = 1.064\ \mu$m and to determine $d\Omega_B/dt$.

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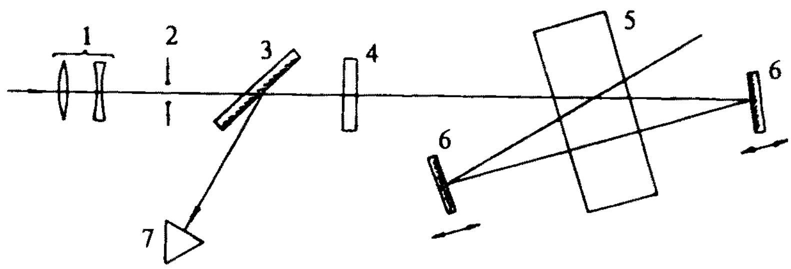

Fig. 1. Layout of the experiment: 1) positive–negative telescope; 2) soft diaphragm; 3) polarizer; 4) quarter-wave plate; 5) cell filled with an SBS-active liquid; 6) mirrors with $R = 100\,\%$; 7) sensor.

2. Experimental

As mentioned above, a competition between the main and angular-dependent modes hampers precise determination of the characteristic period T. Hence, as distinct from [4], the experiments were conducted with a resonator length of the loop laser $L \simeq 1.5$ m and a pumping beam close to a Gaussian one with a typical size $d = 2$ mm. In this case, the cross section of the pump beam defines the cross section of the characteristic aperture diaphragm of the loop resonator. Furthermore, the Fresnel number is approximately equal to unity and the diffraction losses for the angular-dependent modes are high so that these modes are not observed.

Figure 1 shows the schematic layout of the experiment. A linearly polarized single-frequency ($\Delta\nu \leq 10^{-3}$ cm^{-1}) pumping beam formed by a positive lens–negative lens telescope 1 and a soft diaphragm 2 was introduced, via a polarizer 3 and a quarter-wave plate 4, into the active volume 5, which was kept at a constant temperature to within 0.1 K. The optical path was closed by mirrors 6. The angle of convergence of the pump beams was not larger than 10^{-2} rad in all the experimental runs. In order to preserve the geometry of the experiment when the loop length L was changed, we carried out these changes by simultaneous displacements of the mirrors 6 in opposite directions. Radiation reflected from the SBS laser and passed through the quarter-wave plate was removed from the optical scheme by the polarizer 3.

In the course of the experiments, we measured the temporal characteristics of the incident, transmitted, and reflected pulses and also the spatial structure of the pump beam and the generated radiation in the near field. Figure 2 shows photographs of a reflected beam at a distance of 1.2 m from the exit face of an optical cell 5 (at the point conjugated to the diaphragm 2) for various loop lengths L. In this case, carbon disulfide served as the active medium. As is evident, with the condition $\Delta k_{\mathrm{B}} L = 2\pi n$ fulfilled, the structure of the generation beam is closest to that of the pump beam. Then with an increase in $\Delta k_{\mathrm{B}} L$, the size of the beam decreases. For $\Delta k_{\mathrm{B}} L = \pi(2n+1)$, a jumplike increase in the beam size is observed; the characteristic uncertainty in the determination of the length L corresponding to a jumplike change in the beam structure amounts to $\Delta L \simeq 2$ mm for a total length $L \simeq 150$ cm. Then as L increases, the beam size decreases until the condition for the next jump is fulfilled. It is this narrow range of change in the length L corresponding to a jump that allowed us to determine the magnitude of the Brillouin shift with high accuracy using the formula

$$\Omega_{\mathrm{B}} = \frac{2n+1}{2L}\left(1 + \frac{\Delta L}{L}\right),$$

which yields $\Omega_{\mathrm{B}} = 0.12537$ cm^{-1} or $\Omega_{\mathrm{B}} = (3761 \pm 5)$ MHz for carbon disulfide at 20° C with a pump-radiation wavelength $\lambda = 1.064$ μm.

In order to check the accuracy of the technique, we determined Ω_{B} in CS_2 at the same temperature for a wavelength $\lambda = 1.060$ μm. Since $\Omega_B \sim \omega_p \sim 1/\lambda_p$, the ratio of the frequency shifts obtained is equal to the

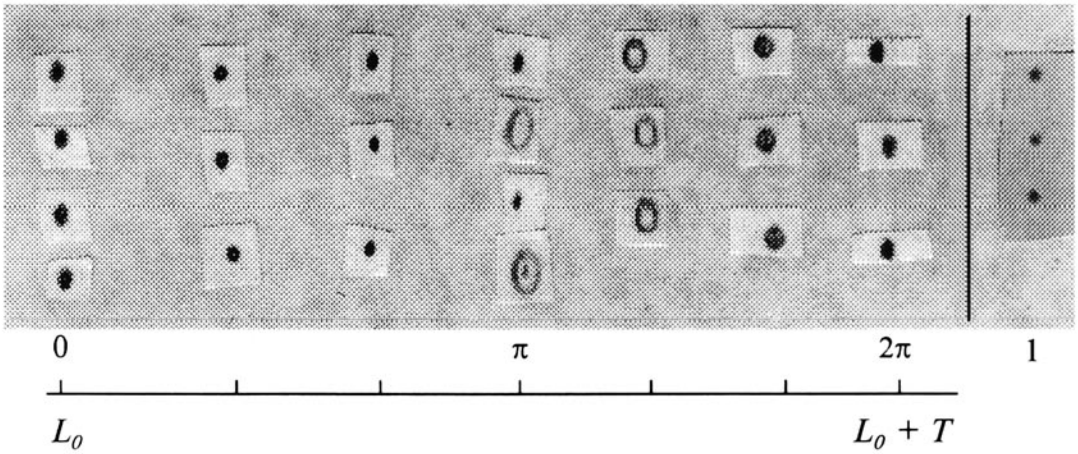

Fig. 2. Photographs taken in the near field of loop-laser generation for several values of the loop length L. The beginning corresponds to a length that is a multiple of the period T: $L_0 = n/\Omega_B$, and the end of the horizontal scale corresponds to the length $L = L_0 + T = (n+1)/\Omega_B$. A series of runs amounting to 10–20 laser bursts were performed for each value of the length. Three or four laser bursts are shown for each length. The length of the jump region at the point π where the spatial structure of the beam is unstable is approximately 2 mm. 1 denotes the near field of the pump radiation.

reciprocal ratio of the wavelengths,

$$\frac{\Delta\nu_B^{1.06}}{\Delta\nu_B^{1.064}} = \frac{1.064}{1.060} = 1.0038.$$

It is this value that was obtained experimentally.

The length L corresponding to a jump can also be determined calorimetrically, i.e., from the change in the efficiency of generation of the loop-scheme laser; in this case, a space filter placed in the detection channel in front of the calorimeter and precisely in the backscattering direction is required to enhance the detection accuracy. Figure 3 shows the experimental dependence of the generation efficiency for the backscattered radiation on the loop length at the active-medium temperatures $t_1 = 19°$ C and $t_2 = 21°$ C. Since the jump occurs for the same magnitude of $\Omega_B L = \pi(2n+1)$, we can easily obtain

$$\Delta\Omega_B L + \Omega_B \Delta L = 0; \qquad \Delta\Omega_B = -\frac{\Omega_B \Delta L}{L},$$

whence, knowing the temperature change, the length, the length change ΔL, and the shift Ω_B (e.g., for CS_2 at $t = 20°$ C), we find $d\Omega_B/dt = (-10.15 \pm 0.3)$ MHz/(° C). Similar measurements were performed in the case of $TiCl_4$. The results obtained are given in Table 1, together with published data for comparison.

3. Results and Discussion

As demonstrated in Fig. 2, with the use of a long resonator, the single mode generation is maintained for any values of $\Delta k_B L$; however, the mode structure changes as $\Delta k_B L$ increases from $2\pi n$ to $2\pi(n+1)$, with the spatial structure of the mode changing abruptly at the point $\Delta k_B L = \pi(2n+1)$. A qualitative explanation of this can consist in the following: the condition of phase balance required for generation is fulfilled at the center of the stimulated-scattering line for $\Delta k_B L = 2\pi n$. In this case, $\omega_s = \omega_p - \Omega_B$. When $\Delta k_B L$ exceeds $2\pi n$

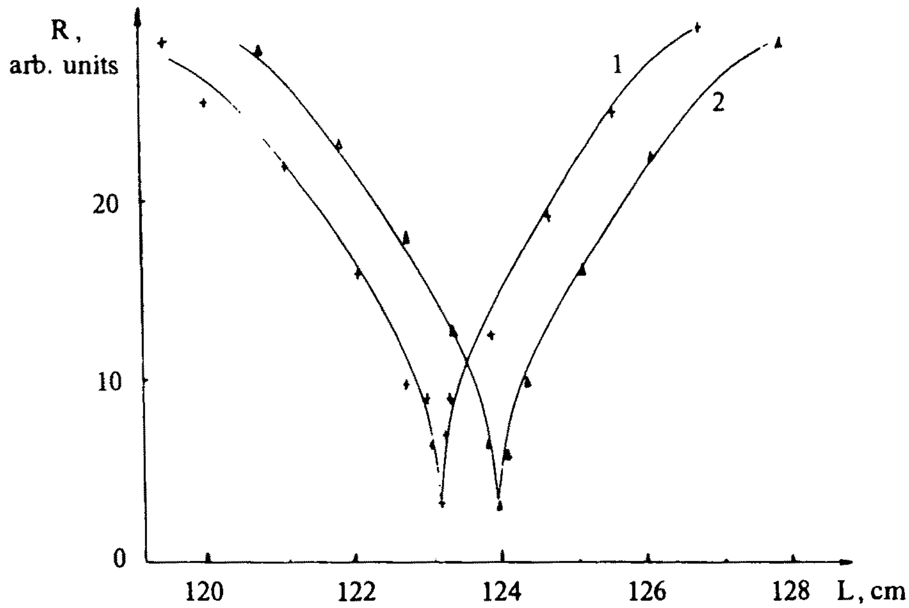

Fig. 3. Efficiency of scattering "exactly back" for two temperatures of carbon disulfide (the SBS-active medium) as a function of the laser loop length, which is varied within a single period: 1) 19° C and 2) 21° C.

TABLE 1

Substance	Temperature, °C	Brillouin shift Ω, MHz	$d\Omega_B/dt$, MHz/(° C)
CS_2	20	3761 ± 5 3761 ± 3 [2]	-10.15 ± 0.3 -9.9 ± 0.2 [2]
$TiCl_4$	20	3065 ± 5 3070 ± 10 [2]	-13.18 ± 0.3 no literature data

owing to an increase in the loop length L, the quantity Δk_B should be decreased so that the phase-balance condition is fulfilled. In this case, $\omega_s = \omega_p - \Omega_B + \delta$, where $\delta > 0$ is the detuning of the generation line with respect to the Brillouin resonance that is required for fulfillment of the phase-balance condition. Since the detuning $\delta > 0$, in view of the anomalous dispersion and the Gaussian pump-beam profile the nonlinear medium acquires the properties of a positive lens, which results in generation of a convergent mode. With $\Delta k_B L = \pi(2n+1)$, the mode that has $\delta > 0$ (with respect to the condition $\Delta k_B L = 2\pi n$) and the mode that has $\delta < 0$ [with respect to the condition $\Delta k_B L = 2\pi(n+1)$] feature the same Q. Similarly, generation of a divergent mode occurs for $\delta < 0$. The Q for the divergent mode with $\delta < 0$ is highest in the case of $\pi(2n+1) < \Delta k_B L < 2\pi(n+1)$. The pattern is reproduced completely with a further change in L.

It is evident that the properties of the active medium are manifested most clearly when $|\delta| \sim 1/\tau$, where τ is the lifetime of optical phonons, because in this case the nonlinear additions to the wave vector of the Stokes radiation are largest. With this in mind, it is a simple matter to derive an expression for the characteristic

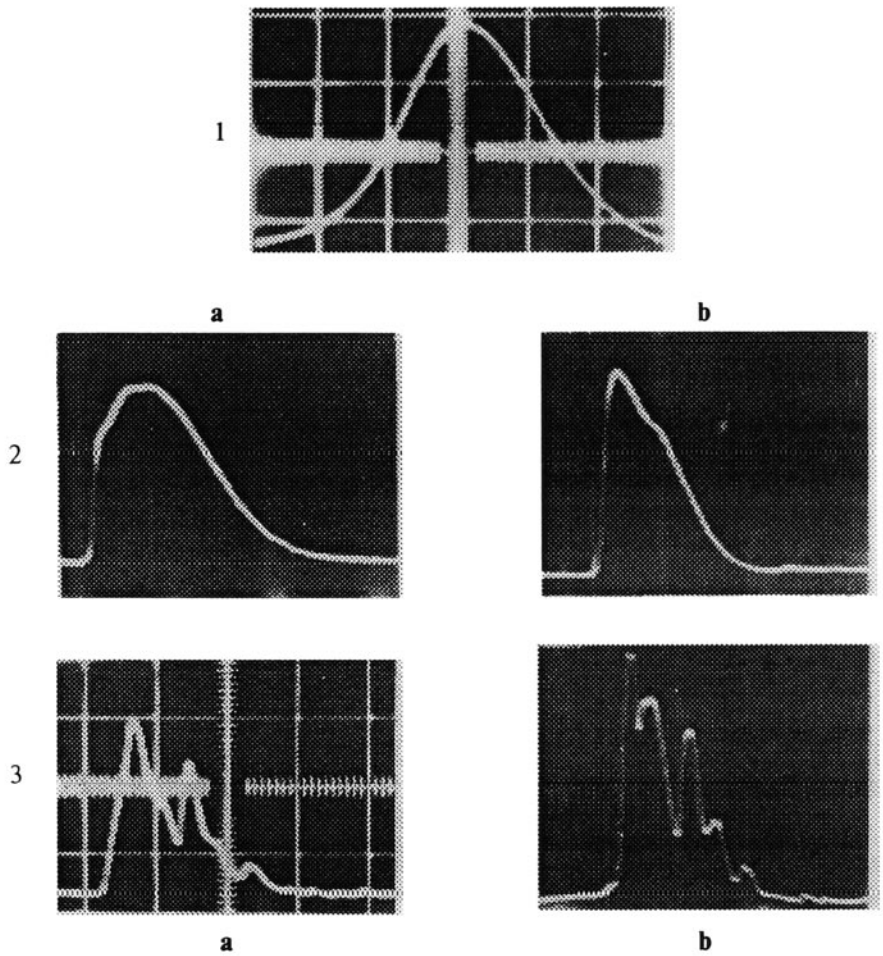

Fig. 4. Oscillograms of pump pulses (1) and loop-laser generation (2 and 3): 2a) for the loop length a multiple of the period $L = 2\pi n/\Delta k_{\rm B}$ and CS_2 as the active medium; 2b) for the loop length a multiple of the period and $TiCl_4$ as the active medium; 3a) for the length $L \simeq \pi(2n+1)/\Delta k_{\rm B}$ and CS_2; 3b) for $L \simeq \pi(2n+1)/\Delta k_{\rm B}$ and $TiCl_4$. The value of a horizontal-scale division is 20 ns.

loop length that corresponds to the case where these effects are pronounced: $L \leq \pi c\tau$, where c is the speed of light. In fact, when $TiCl_4$ possessing $\tau \simeq 2$ ns was used as the active medium, the discontinuity at the point $\Delta k_{\rm B} L = \pi(2n+1)$ was much less pronounced than in the case of CS_2 ($\tau \simeq 7$ ns), and for CCl_4 ($\tau \simeq 1$ ns) the effect was practically indiscernible.

An important feature of realization of various modes for a loop-scheme laser is that, with δ being nonzero, the condition of phase balance fulfilled in the approximation of a given pump field can be violated in the regime of saturation. This should result in pulsations of the loop-laser radiation for $\Delta k_{\rm B} L \neq 2\pi n$. We note that we observed such a regime [4]; however, results reported in [5] and related to realization of the regime for selection of angular-dependent modes are closest to the case under consideration.

Figure 4 presents oscillograms of pump and Stokes-signal pulses for several values of the quantity $\Delta k_{\rm B} L$;

pulsations are seen when the values of $\Delta k_B L$ are close to $\pi(2n+1)$.

4. Conclusion

Thus, we studied the spatial characteristics of loop-scheme lasers and identified resonator configurations that provide a jumplike change in the spatial characteristics of the laser as a function of the resonator length. This made it possible to develop a technique for precise determination of Brillouin shifts and their dependences on the active-medium temperature.

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