# Spectral and temporal characteristics of stimulated scattering of light of the Rayleigh wing in an external transverse resonator

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Anomalously large spectral shifts and broadenings of the line of stimulated Rayleigh-wing (SRW) scattering in an external transverse resonator have been observed and investigated, as well as a periodic temporal structure in the SRW radiation. This temporal structure is attributed to mode locking of the SRW radiation in the resonator as a result of the quadratic Kerr effect.

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The influence of stimulated Rayleigh-wing light scattering (SRW) was observed in[1] and investigated repeatedly under various experimental conditions (e.g., [1–6]), including studies in an external transverse resonator.[8, 9] This phenomenon is the result of orientation of anisotropic molecules of the liquid (the quadratic Kerr effect) in the combined field of two waves, the one scattered by the fluctuations of the anisotropy (initially weak), and the pump wave. It follows from the linearized theory[5–7] that the intensity of the scattered light should increase exponentially with increasing pump intensity in the region of the nonlinear interaction, with a growth rate

$$g = 2A\frac{\pi\omega_s K}{cn^2}\,\frac{\Omega\tau}{1+\Omega^2\tau^2}\;;\qquad K = \frac{1}{15}\,\frac{N(a_\parallel - a_\perp)^2}{k_B T}. \tag{1}$$

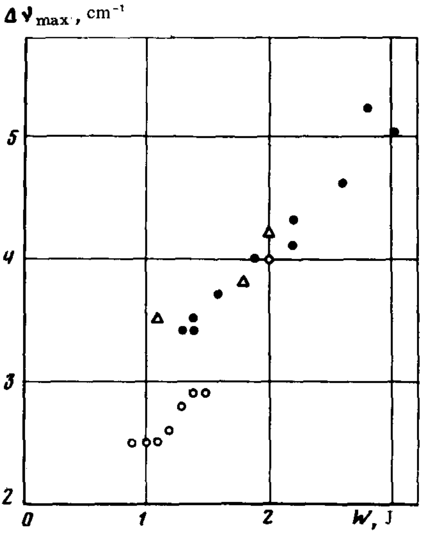

FIG. 1. Dependence of the position and of the maximum of the SRW line $\Delta\nu_{\max}$ on the pump energy $W$: ●—mirror reflection coefficients $R_1 \sim 100\%$ and $R_2 \sim 80\%$, region of nonlinear interaction $l_{nl} \approx 3$ cm, optical resonator length $L = 30$ cm; ○—$R_1 \sim 100\%$, $R_2 \sim 80\%$, $l_{nl} = 18$ cm, $L = 30$ cm; Δ—$R_1 \sim 100\%$, $R_2 \sim 80\%$, $l_{nl} \approx 18$ cm, $L = 30$ cm.

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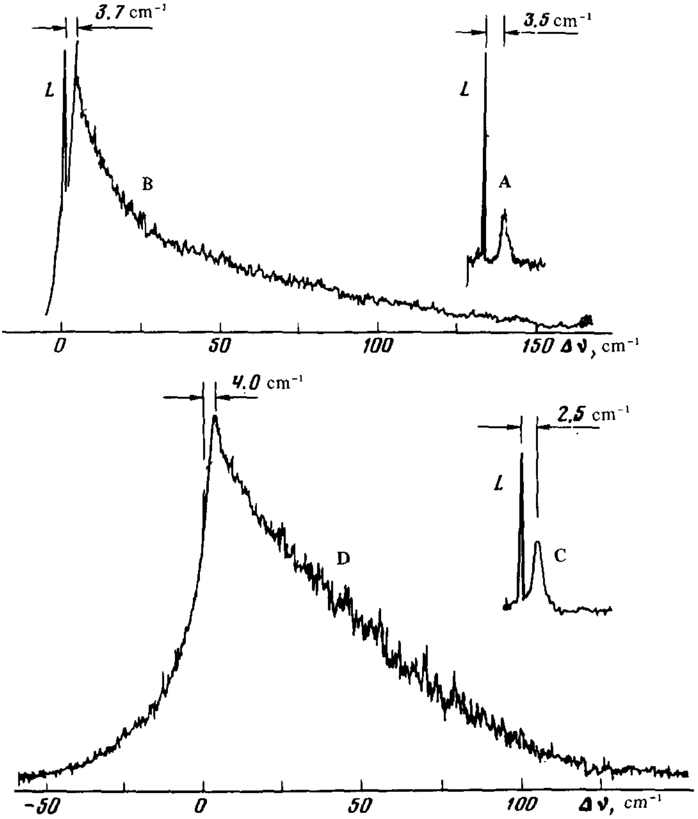

FIG. 2. Microphotograms of the SRW under various experiment conditions: $L$—laser-emission line, A,B) $R_1 \sim 100\%$, $R_2 \sim 80\%$, $l_{nl} = 18$ cm, $L = 30$ cm; C,D) $R_1 \sim 100\%$, $R_2 \sim 8\%$, $l_{nl} = 18$ cm, $L = 30$ cm.

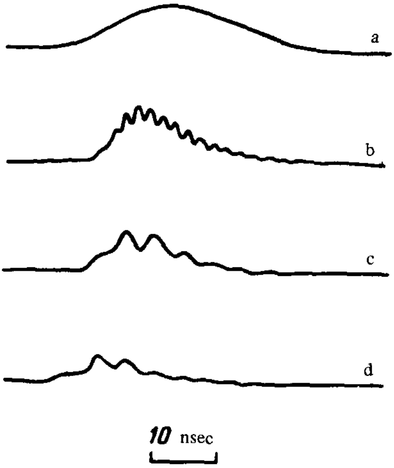

FIG. 3. Oscillogram of pump radiation (a) and of SRW radiation at optical-resonator lengths $L = 30$ cm (b) and $L = 60$ cm (c,d). (a,c—obtained with coaxial photocells FÉK-15; c,d—with FÉK-09).

In (1) $\Omega = \omega_L - \omega_S$, where $\omega_L$ and $\omega_S$ are the pump and scattered-light frequencies, $\tau$ is the anisotropy relaxation time, $N$ is the number of molecules per $cm^3$, $\alpha_\parallel$ and $\alpha_\perp$ are the principal polarizabilities of the molecule, $c$ is the speed of light, $n$ is the refractive index, and $A$ is a quantity of the order of unity. It follows from (1) that the SRW radiation should have a maximum at a frequency $\Omega = 1/\tau$. Thus, SRW provides a method of measuring the anisotropy relaxation time by determining the line shift of the stimulated scattering of light. A convenient procedure for such measurements is the excitation of SRW in an external transverse resonator, as confirmed by experiments[9] on several liquids with not too small values of $\tau(\frac{1}{2}\pi c\tau < 0.3\ cm^{-1})$. It was recently observed,[10] however, that under the same experimental conditions in carbon disulfide and in other media with small values of $\tau(\frac{1}{2}\pi c\tau > 0.6\ cm^{-1}$ the SRW line is shifted much more than expected from (1) and from the data on the value of $\tau$. That study yielded new data on the SRW spectrum in an external transverse resonator, and conditions were obtained under which the shift of the SRW line yields the correct value of $\tau$ also for the case when $\tau$ is small. In experiments where the SRW radiation density in the resonator was large enough, a periodic temporal structure in its radiation was observed.

The experimental setup was the same as described in[9,10]. The vessel with the carbon disulfide was placed inside a resonator made up of spherical mirrors; one mirror had a reflection coefficient 100%, and the other 80 or 8%. A ruby-laser pulse of power ~ 50—150 MW and duration ~ 20 nsec was focused into the investigated liquid with a cylindrical lens (focal length 6 cm) whose generatrix was directed along the resonator axis. The pump light was polarized in the scattering plane, so that no stimulated Mandel'shtam-Brillouin scattering was excited along the resonator axis by the pump light. To analyze the spectrum of the scattered light we used a diffraction spectrograph with linear dispersion 3.6 $cm^{-1}$/mm. The temporal character of the radiation was analyzed with the aid of an FÉK-09 or FÉK-15 coaxial photocell and an I2-7 oscilloscope.

When the resonator was made up of mirrors with reflection coefficients 100 and 80% and the optical length of the resonator was $L = 30$ cm, the SRW line shift was always larger than expected from (1) and from the data on the anisotropy relaxation time. Instead of the expected shift 2.3—2.5 $cm^{-1}$, a shift ~ 3.5 $cm^{-1}$ was observed at the lasing threshold and this shift increased with increasing pump energy regardless of the length of the region of the nonlinear interaction (see Fig. 1). With increasing pump energy, the SRW line spectrum also changed (see Figs. 2A and 2B); whereas at the lasing threshold the line was sufficiently narrow and symmetrical (Fig. 2A), with increasing pump energy it became asymmetrical (Fig. 2B) with a sharp edge on the anti-Stokes and a broad wing (up to 150 $cm^{-1}$) on the Stokes side. Although the time scan of the pump radiation was smooth and bell shaped (see Fig. 3a), the SRW radiation contained a periodic temporal structure (Fig. 3b) with a period $T = 2L/c = 2$ nsec. When the optical length of the resonator increased to 60 cm, the temporal period increased also to 4 nsec (Figs. 3c and 3d). The apparent depth of modulation was ~ 50%, but when account is taken of the temporal resolution of our apparatus it should actually be no less than 70—80%.

When the resonator was made up of mirrors with reflection coefficients 100 and 8%, the theoretically expected SRW line shift ~ 2.5 $cm^{-1}$ was observed at the lasing threshold and when the pump threshold intensity was raised to 30%, but with further increase of the pump energy the shift of the maximum began to increase (Fig. 1). With increasing pump energy, the SRW line is strongly broadened in the Stokes direction (Figs. 2C and 2D), and also, albeit not so strongly, in the anti-Stokes direction. Under these experimental conditions, the oscillogram of the SRW radiation has a smooth form similar to that of the pump pulse (Figs. 3a). In the case when one of the resonator mirrors was completely removed, no SRW could be observed at our pump power. It has thus been shown that excitation of stimulated scattering and a determination of the correct relaxation time $\tau$ from the shift of the SRW line are possible for liquids with small values of $\tau$, by using a resonator in which one of the mirrors has a small reflection coefficient. If the resonator is made up of mirrors with large reflection coefficients, then the high SRW intensity can result in repeated scattering and in temporal-structure singularities that distort the spectrum.

We have observed for the first time in a resonator a periodic of SRW-radiation temporal structure that is caused in our opinion by mode locking of this radiation in the resonator as a result of a Kerr effect quadratic in the field. The experimental grounds for this conclusion are the following: the modulation period is equal to double the time of passage over the optical length of the resonator; the large modulation depth (not less than 70—80%); the reproducibility of the temporal radiation picture in several dozen experiments; the presence of a temporal structure only under the condition $1/\tau \gg \Omega_m$, where $\Omega_m$ is the intermode spacing; the increase of the time elapsed prior to the appearance of the first maximum in the temporal structure with decreasing SRW radiation density in the resonator.

An analysis of the nonlinear Maxwell equation together with the relaxation equation for the Kerr susceptibility has shwon that the SRW radiation modes in the resonator are stably locked if the intensity of this radiation is strong enough. Radiation pulses of duration $\sim\tau$ should then be observed at the exit from the resonator.

Liquids consisting of anisotropic molecules were used earlier for mode locking of a multimode solid-state laser.[11,12]

The locking conditions for such an experimental setup will be optimal if the gain band of the active element is $\Delta\nu_L \approx 1/\tau$. Under the conditions of our experiments the gain band is always $1/\tau$. Since, in addition, the active and phasing elements coincide and the active element (liquid) is homogeneous, it follows that the conditions for the locking are also improved. Finally, by varying the liquid or its viscosity it is possible to vary the relaxation time $\tau$ in a wide range[5] and by the same token vary the SRW pulse duration (from $10^{-10}$ to $10^{-12}$ sec).

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