

Stimulated concentration scattering of light in stratifying solutions

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A stimulated scattering line, which has no frequency shift relative to the lasing line (within the measuring error $\cong 0.003 \text{ cm}^{-1}$) has been observed near the phase transition temperature in aqueous solutions of dimethylpyridine (2,6 and 3,4 lutidines). The scattering is interpreted as a stimulated concentration scattering.

Virtually all forms of stimulated scattering (SS) of light, except stimulated concentration scattering (SCS) characteristic of solutions, have been observed and studied in stratifying solutions and other condensed matter. This type of scattering must, as we know, have a small growth rate in comparison with, for example, the stimulated Mandelstam-Brillouin scattering (SMBS). Interest in this matter was expressed in Ref. 1, where estimates were given for the gain for the steady-state excitation of SMBS and SCS in various media

$$\frac{g_c}{g_B} = \frac{k\nu_s \eta_T (\partial \epsilon / \partial c)_{P,T}^2}{2 \rho_0^3 (\partial \epsilon / \partial \rho)_c^2 T (\partial \mu / \partial c)_{P,T}}, \quad (1)$$

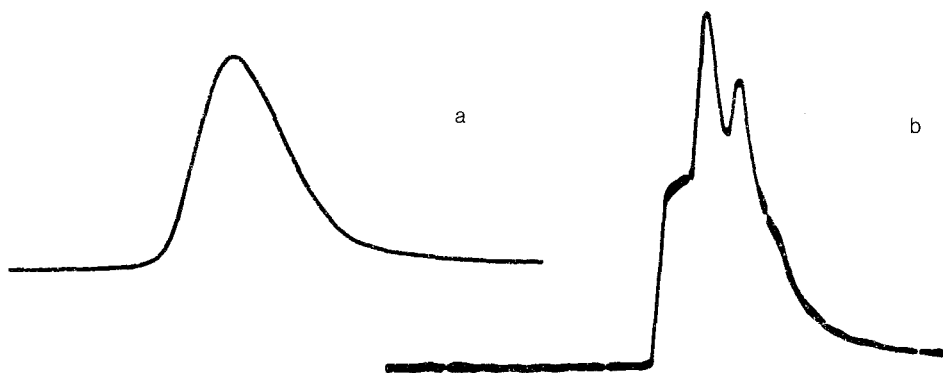


FIG. 1. Oscilloscope traces of the stimulated scattering (the sweep is 20 ns/div). a— β -Picoline solution; the spectrum contains only the SMBS line; b—3,4 lutidine solution; the spectrum has SMBS line and unshifted line ($t_s = 16.5^\circ\text{C}$).

where k is the wave number, v_s is the hypersound velocity, ρ_0 is the density, μ is the chemical potential, ϵ is the dielectric constant, c is the concentration, and η_T is the isometric viscosity.

We present here the results of an experimental study of the stimulated scattering in stratifying aqueous solutions. We chose 2,6 and 3,4 lutidine solutions of critical concentration and a β -picoline solution, which has a singularity.^{2,3} The 2,6 lutidine solution had a large temperature-dependent capacity for light absorption. At 20°C the absorption at the wavelength of the excited light was $\alpha \approx 0.0261 \text{ cm}^{-1}$. Substituting into (1) the quantity $\delta\mu/\delta c \approx 10^6 \text{ cm}^2 \text{ s}^{-2}$, which was obtained in Ref. 4 for the nitromethane-*n*-amylalcohol solution, and the quantity $\delta n/\delta c \approx 3 \times 10^{-3}$, which we measured using the Abbe refractometer, we find $g_c/g_B \approx 0.25$. We can assume on the basis of the value that the stimulated concentration scattering develops in the solution near its stratification temperature.

To excite the stimulated scattering, we used a second-harmonic light from a single-frequency Nd:YAG laser (the radiation wavelength was $\lambda = 0.532 \text{ nm}$, the pulse length at half-maximum was $\tau \approx 20 \text{ ns}$, the maximum energy was $E \approx 5 \text{ mJ}$, and the lasing linewidth was $\Delta\nu \approx 0.003 \text{ cm}^{-1}$). The laser light with a smooth spatial distribution was focused by a spherical lens ($f = 20 \text{ cm}$) into a thermally stabilized quartz cell which contained the test liquid. The temperature was stabilized within $\Delta T \approx 0.1^\circ\text{C}$. The spectral composition of the pump light and the stimulated scattering light were analyzed using Fabry-Perot interferometers with dispersion regions 0.5, 0.25, and 0.033 cm^{-1} . The temporal component of the processes was recorded by LFD-2A photodiodes, whose signals were sent to a C7-19 oscilloscope. During the experiment, the absence of an optical breakdown of the liquid was controlled at the focal point of the lens.

It was established that at lasing energies up to 5 mJ the temporal component (Fig. 1a) and the spectral component of the stimulated scattering in the β -picoline

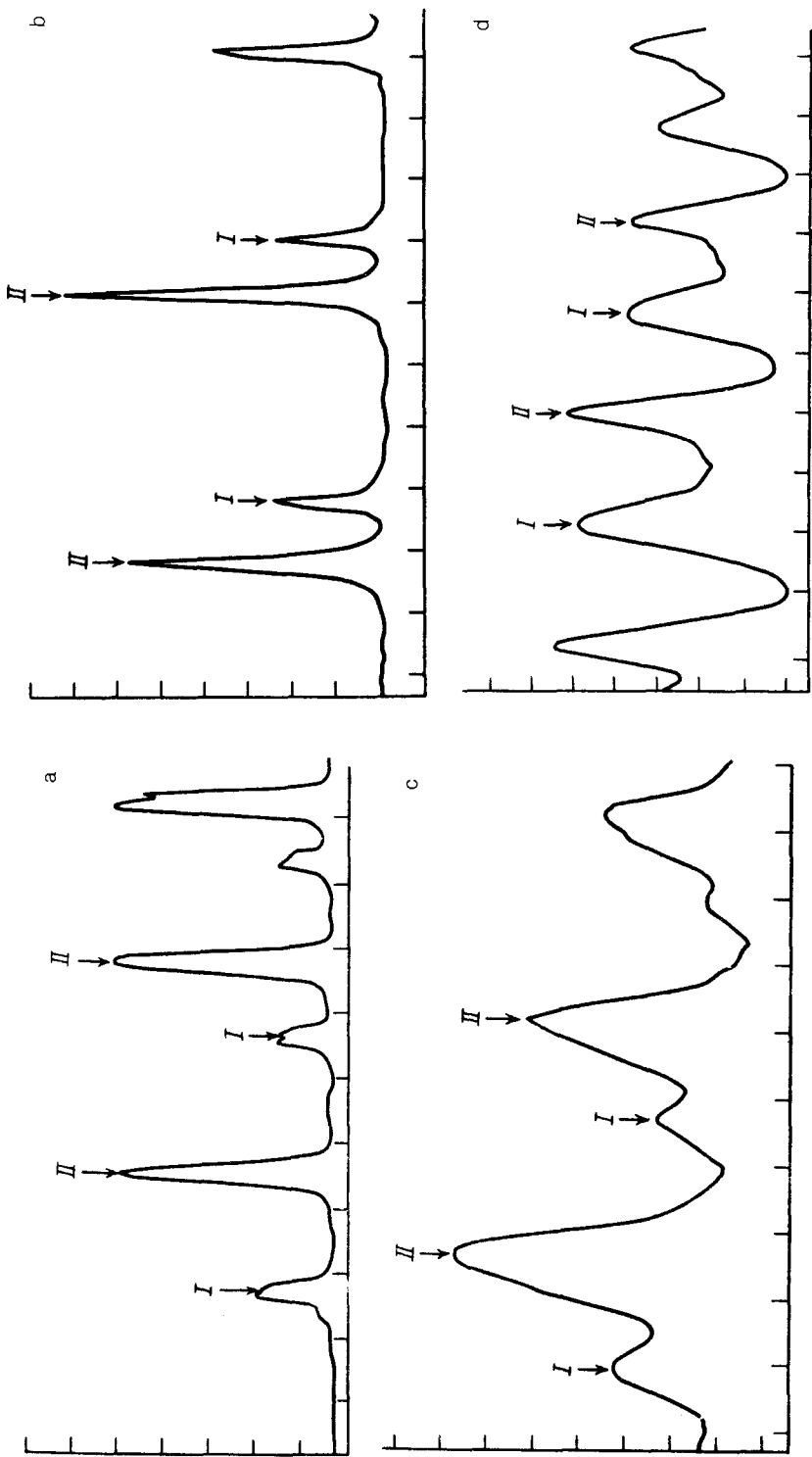


FIG. 2. Densitograms of SS spectra in a 3.4 lutidine solution near the critical point ($t_c = 16.5^\circ\text{C}$). I—Central line, II—SMBS line. a—Dispersion region of the Fabry-Perot interferometers is 0.5 cm^{-1} , the SMBS shift is $\Delta\nu \approx 0.19\text{ cm}^{-1}$; b—the dispersion region of the Fabry-Perot interferometer is 0.25 cm^{-1} , the SMBS shift is $\Delta\nu \approx 0.19\text{ cm}^{-1}$; c—the dispersion region of the Fabry-Perot interferometers is 0.033 cm^{-1} ; d—the dispersion of the Fabry-Perot interferometers is 0.033 cm^{-1} ; I— the central line and the pump line in Figs. 2c and 2d the distance between lines I and II is $\approx 0.017\text{ cm}^{-1}$.

solution had no structural features, in contrast with the stimulated scattering in pure liquids (we have disregarded here the fine structure of the SS lines^{5,6}), either at 20 °C or at a temperature of ≈ 70 °C, which is the temperature of the singularity.

The SS process in a 3,4 lutidine solution was studied at temperatures of 10, 16.5, and 50 °C, i.e., near the lower stratification point $T_c \approx 12.5$ °C and at some distance from it. At 50 °C the stimulated scattering light in this solution had no structural features. Near the stratification point (above and below it in terms of temperature) the SS spectrum revealed, in addition to the SMBS line, the presence of an emission line of a frequency which was not shifted (within 0.003 cm^{-1} error margin) relative to the pump light (Figs. 2a–2d). The threshold energy of the pump light, which is necessary in order to detect the unshifted SS line, was about 0.5–1 mJ. The oscilloscope trace in Fig. 1b characterizes the case in which two SS mechanisms compete with each other in the medium. Such a temporal behavior was observed previously (in Ref. 5, for example) in the case of the SMBS and STS-II excitation in homogeneous liquids.

The SMBS and SS lines at an unshifted frequency also were observed in 2,6 lutidine solution near the critical point (at 27.5 °C and 32 °C). The STS-II line, which was shifted in the anti-Stokes direction from the pump frequency by an amount $\Delta\nu \approx 0.01 \text{ cm}^{-1}$, was also observed. The time evolution of the SS processes in 2,6 lutidine solution is similar to that shown in Fig. 1b, although its modulation depth is smaller, which is apparently attributable to the STS-II. The STS-I process, which should have a Stokes frequency shift relative to the pump line, was not detected in any of the solutions that were studied.

In summary, we have observed a stimulated scattering in the solutions near the stratification points at a frequency which was not shifted relative to the pump frequency. This scattering can be interpreted as a stimulated concentration scattering which develops against the background of SMBS and STS-II processes.

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