

Registration of central peak of light scattering in liquids with a spectral resolution 10^{14}

V. P. Tykhinskii and V. L. Pankov

Institute of Radio Engineering, Electronics, and Automation

(Submitted 30 May 1978)

Pis'ma Zh. Eksp. Teor. Fiz. **28**, No. 7, 437-439 (5 October 1978)

Anomalous low-frequency fluctuations of the amplitude and of the phase of the scattered light were observed in two-component solutions of CCl_4 and H_2O in measurements by the photodisplacement and by the time-interval methods (N. N. Evtikhiev *et al.*, *Soviet Journal of Quantum Electronics* **7**, 36, 1977). The spectra of the fluctuations in the low-frequency region were obtained with a resolution of 1 Hz.

PACS numbers: 78.35. + c, 42.60.He

The spectra of Rayleigh scattering of light contain information on the kinetic processes in the medium, including the relaxation of the bulk viscosity, diffusion, and the change of the structure.^{2,3} The registration of slow processes with a correlation time 10^{-3} -1 sec by the usual photodisplacement methods is made complicated by the large technical width of the emission line of the laser (10^6 - 10^6 Hz).

In this communication we report the preliminary results of measurements of the spectrum of the central peak of Rayleigh scattering (CPRS) in liquids by a new method, that ensures a spectral resolution limited by the natural line width of the emission (0.1-1 Hz for helium-neon lasers) and the instrumental function F_{IF} of the recording instruments. The use of the time-interval method, which was developed for precision interferometry,¹ has made it possible to carry out independent measurements of the phase of the scattered wave with a sensitivity on the order of $\langle \Delta\phi \rangle \sim 10^{-3}$.

In our method, the influence of the technical fluctuations of the laser frequency on the photocurrent spectrum is eliminated by the use of a stable compensated Michelson interferometer,¹ and the lowering of the contribution of the low-frequency noise is due to modulation of the reference arm. The relatively large spatial resolution ($V_{\text{pr}} \sim 10^{-9}$ cm³) was attained by focusing the probing beam into the interior of a flat cell with axial length $l = 3$ mm, mounted in the measurement arm of the interferometer. The spectral composition of the photocurrent was investigated with a spectrum analyzer having $F_{\text{IF}} \sim 10$ Hz, or with a panoramic multichannel analyzer SG-1 with $(F_{\text{IF}})_{\text{min}} \sim 1$ Hz.

In weakly scattering liquids (water, CCl_4 , alcohol, acetone), the CPRS spectrum is of the form shown in Figs. 1a and 1b. On the wings of these spectra ($10 < F < 100$ Hz) the fluctuation level did not exceed noticeably the shot-noise level. At a higher resolution, weak fluctuations of the CPRS were observed in a band $\Delta F = 1$ -10 Hz with a ratio of the signal to the shot-noise level equal to 1.5 (see Figs. 2a and 2b).

The next measurement run was performed on a mixture $(\text{CCl}_4)_{1-x}(\text{H}_2\text{O})_x$ at $X = 0.007$ - 0.008 and revealed an anomalous dependence of the power and of the spectrum of the fluctuations on the water concentration X . To this end, stirring was

used to produce a solution with a concentration $X_{\max} = 0.008$ higher than the equilibrium value $X_0 = 0.007$ at room temperature. Equilibrium was established in a time $t \approx d/4D = 30$ min, and the concentration of the water changed in this case monotonically in the range from X_{\max} to X_0 . Here d denotes the distance from the boundary of the liquid surface to the axis of the probing beam, and D is the diffusion coefficient. The concentration $X(t)$ was determined from the experimental $n(t)$ dependence (see Fig. 3) measured by the time-interval method.

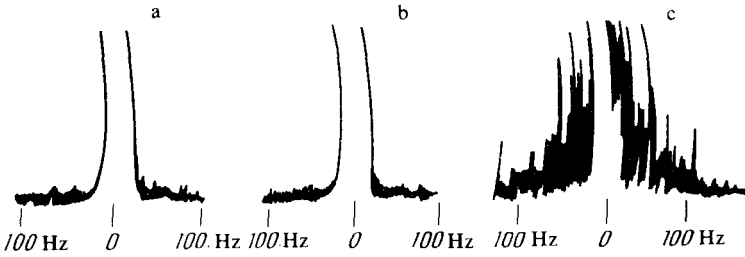


FIG. 1. CPRS spectra for light scattering in a— H_2O , b— CCl_4 , c— $(\text{CCl}_4)_1 - x(\text{H}_2\text{O})_x$ at $X = 0.0075$ and at a resolution $F_{\text{IF}} \sim 10$ Hz.

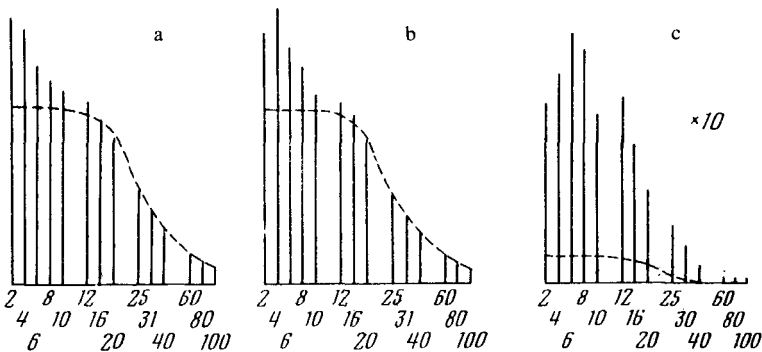


FIG. 2. CPRS spectra for light scattering in a— H_2O , b— CCl_4 , c— $(\text{CCl}_4)_1 - x(\text{H}_2\text{O})_x$ at $X = 0.0075$ and at a resolution $F_{\text{IF}} \sim 1$ Hz.

Figures 1c and 2c show typical CPRS spectra at $X = X_{\text{cr}}$ and at respective resolutions 10 and 1 Hz. Simultaneously with the registration of the spectra, we measured the amplitude of the fluctuations ΔI_{\sim} of the intensity and $\Delta\phi$ of the phase of the scattered wave. From the ΔI_{\sim} and $\Delta\phi$ curves on Fig. 3 follows a strong increase of the amplitude of the fluctuations of the phase and of the intensity, as well as of the power of the spectrum $P(t) = \int_0^{\infty} P(t, F) dF$ at a value $t = 2-3$ min, which corresponds to $X = X_{\text{cr}} = 0.0075$. From the measurements of the phase fluctuation we determined by the formula $\Delta n = \lambda \Delta\phi / 4\pi l$ the absolute value of the fluctuation of the refractive index, averaged over the volume V_{pr} . At $X = X_{\text{cr}}$ the value of Δn was 4.2×10^{-5} .

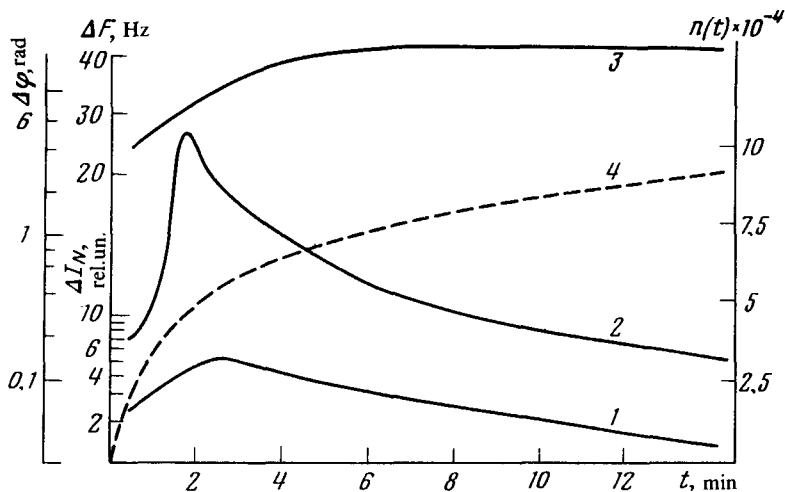


FIG. 3. 1—fluctuations ΔI_n of the intensity of the scattered light, 2—fluctuations $\Delta\phi$ of the phase of the scattered wave, 3—width ΔF of the CPRS spectrum, 4—average value of the refractive index $\bar{n}(t)$.

Results of the same order were obtained also with other binary mixtures, such as water–acetone and water–alcohol.

The foregoing results admit of the following interpretation:

1) At high spatial and spectral resolution fluctuations of the density, with characteristic frequencies 2–8 Hz, are observed in the CPRS, these fluctuations are apparently due to relaxation processes in the structure of the liquid (formation of clusters).

2) In binary solutions, near the points of critical concentration, an increase takes place in the fluctuations of the intensity, and a sharper increase in the fluctuations of the phase of the scattered wave; this agrees with the known data on critical opalescence in solutions² and with the assumption that the processes in structure fluctuations have a cooperative character.³ In contrast to the results of Ref. 3, however, the increase of the light scattering was observed in the region of low water concentration.

3) The large value of the relative modulation of the intensity and of the phase of the scattered wave offer evidence that the volume of the fluctuation and of the probing volume V_{pr} are commensurate, so that the correlation radius is of the order of $10 \mu\text{m}$.

The authors thank I.L. Fabelinskiĭ for a useful discussion and for critical remarks.

¹N.N. Evtikhiev, Yu. A. Snezhko, V.P. Tychinskiĭ, G.R. Levinson, and V.P. Zakharov, *Kvantovaya Elektron.* (Moscow) **4**, 69 (1977) [*Sov. J. Quantum Electron.* **7**, 36 (1977)].

²I.L. Fabelinskiĭ, *Molekulyarnoe rasseyanie sveta* (Molecular Scattering of Light), Nauka, 1965. [Plenum, 1958].

³M.F. Vuks, *Rasseyanie sveta v gazakh, zhidkostyakh i rastvorakh* (Light Scattering in Gases, Liquids, and Solutions), Leningrad University Press, 1977.