Detection of subhertz fluctuations of the anisotropy in small-angle scattering of light

V. P. Tychinskiĭ, V. L. Pankov, A. G. Daugel'-Dauge, and A. V. Karpun'kin
Moscow Institute of Radio Engineering, Electronics, and Automation

(Submitted 1 July 1986)
Pis'ma Zh. Eksp. Teor. Fiz. **44**, No. 4, 197–200 (25 August 1986)

A new method is proposed for studying fluctuations of the anisotropy in the small-angle scattering of light. In nematic liquid crystals and several biological objects, this new method has revealed fluctuations with a spectral power density of 10^{-3}–10^{-2} Hz^{-1} in the frequency interval 0.1–1 Hz.

Critical and cooperative processes are characterized by fluctuations of the order parameter with large correlation radii $(r_K \gg \lambda)$, which correspond to scattering through small angles $(\alpha \sim \lambda / r_K)$ (Ref. 1). The light intensity fluctuations due to these fluctuations in the order parameter are thus almost impossible to observe against the background of the unscattered component. The difficulty can be overcome comparatively simply by polarization methods, however, if the object is anisotropic. Such methods look promising primarily for studying liquid crystals, in which orientation fluctuations are the controlling fluctuations.[2] We would also naturally expect a strong anisotropic scattering in biological objects, since they contain liquid-crystal structures and similar structures.[3]

In this letter we report a study of fluctuations of the anisotropy by a new method. We have confirmed the existence of strong equilibrium fluctuations of the orientation in nematic liquid crystals in the hertz and subhertz ranges. We have discovered some anomalous low-frequency fluctuations in biological objects.

0021-3640/86/160250-05$01.00

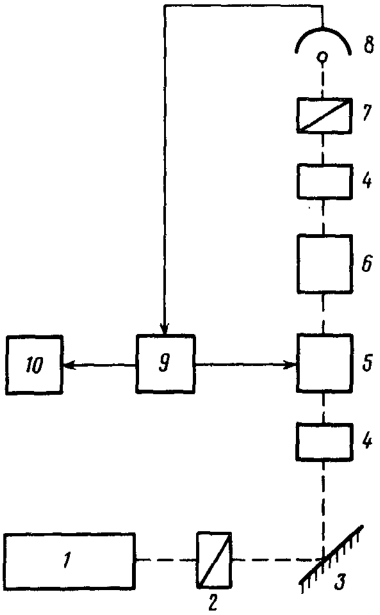

FIG. 1. Block diagram of the dynamic laser polarimeter. 1—Laser; 2—polarizer; 3—rotating mirror; 4—$\lambda/4$ plate; 5—object; 6—electrooptic modulator; 7—analyzer; 8—photodetector; 9—electronic signal processing; 10—computer.

Figure 1 is a block diagram of the dynamic laser polarimeter which can carry our real-time absolute measurements of the dispersion, spectral power density, and other properties of fluctuations.

The apparatus has a modulator-object-compensator-analyzer layout. The beam from the laser (block 1 in Fig. 1; $\lambda = 0.63$ μm, $P = 0.1$ mW) passes in succession through a polarizer (2), a compensator (4) (a $\lambda/4$ plate), and an electrooptic modulator (5) and strikes the object (6). The scattered light after the second compensator and the analyzer (7) is measured by a photodetector (8). Information on the fluctuations is extracted from the variable component of the photocurrent in an electronic system (9) by a time-interval method.[4] This method involves a modulation of the polarization of the probing beam. The results are analyzed statistically by an Élektronika-60 microcomputer (10).

In the approximation of the model of a thin anisotropic screen,[5] the measured quantity—the phase Φ of the variable component of the photocurrent—and its fluctuations are related to the orientation of the optical axis, θ, and the birefringence δ by $\tan\Phi = -\cos 2\theta \tan\delta$, $\delta = 2\pi/\lambda(n_0 - n_e)h$, where h is the sample thickness.

The spectra of the scattering by equilibrium fluctuations of the orientation in a planar cell (MBBA; $h = 30$ μm) are shown in Fig. 2 for voltages $U = 0$ (a) and $U = 5.3$ V (b). Curve c is the level of intrinsic noise of the apparatus. The dashed curves are Lorentzian approximations

$$S_f(F) = \frac{\sigma_f^2\ \Gamma/2\pi}{(2\pi F)^2 + \Gamma^2}\,.$$

The spectral width $\Gamma/2\pi$ for curve 2a agrees satisfactorily with the relaxation time $\tau = \eta/Kq^2$ for $q = 10^3$ cm^{-1}, $\eta = 10$ Ps, and $K = 10^{-6}$ dyn.

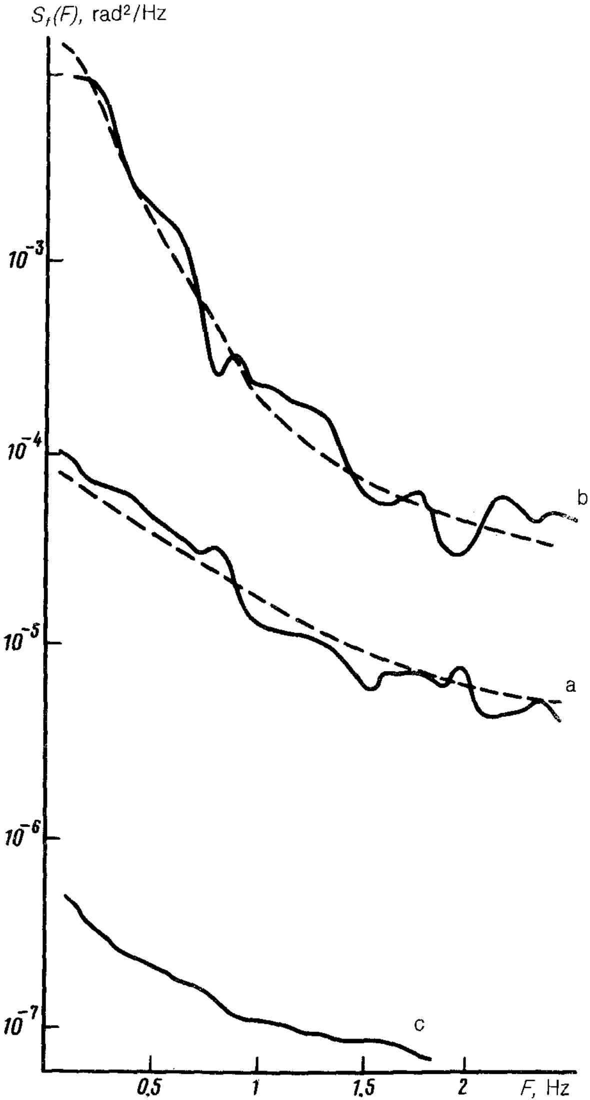

FIG. 2. Anisotropy fluctuation spectra $S_f(F)$ in a nematic-liquid-crystal cell. a—$U = 0$ V; b—$U = 5.3$ V; c—intrinsic noise level. The dashed curves are Lorentzian approximations. The values of the variance σ_f^2 and the width $\Gamma/2\pi$ are, respectively: a) $\sigma_f^2 = 4 \times 10^{-5}$, $\Gamma/2\pi = 0.76$ Hz; b) $\sigma_f^2 = 3.4 \times 10^{-3}$, $\Gamma/2\pi = 0.14$ Hz; c) $\sigma_f^2 = 6 \times 10^{-7}$.

The variance $\sigma_f^2 = \int S_f(F)dF$ is proportional to the intensity of the equilibrium fluctuations, which is given by the following expression, according to Ref. 2:

$$\sigma_\theta^2 = \frac{k_B T}{VKq^2} ,$$

where V is the correlation volume, K is the elastic constant, and **q** is the scattering vector. Long-wave fluctuations with $q \lesssim \pi/h$ dominate σ_θ^2.

The narrowing of the spectrum and the intensification of the fluctuations[6] at the Fréedericksz transition ($U_t = 5.3$ V) agree qualitatively with the theory of phase transitions.

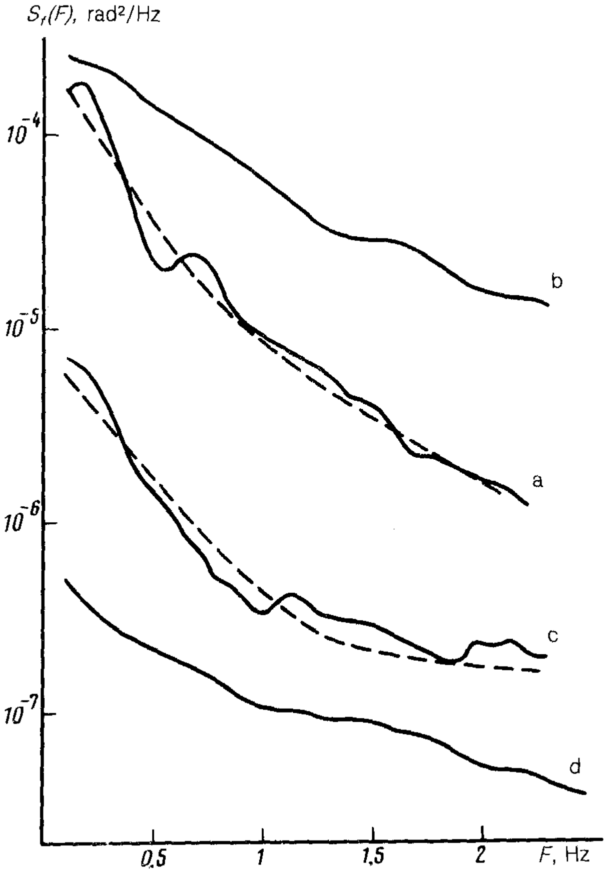

FIG. 3. Anisotropy fluctuations spectra $S_f(F)$ for the alga *Nitella*. a—Fluctuations in the living cell ($\sigma_f^2 = 1.2\times 10^{-4}$); b—biogenic fluctuations in a cell in which flow of the protoplasma has been suppressed ($\sigma_f^2 = 5\times 10^{-5}$); c—dead cell ($\sigma_f^2 = 2\times 10^{-6}$); d—intrinsic noise level. The dashed curves are Lorentzian approximations.

Figure 3 shows spectra $S_f(F)$ for a cell of the alga *Nitella*, which is commonly used in biophysics experiments.[7,8] Curve a represents the living cell; b again represents the living cell, but motion of the protoplasm has been suppressed by an electric pulse; c represents the dead cell; and d is the noise level. The dashed line is a Lorentzian approximation. It follows from a comparison of spectra a, b, and c in Fig. 3 that the intensity of the fluctuations in the living cell is exceeded by a factor of ~ 15, while the correlation time is conserved, and there is a fourfold broadening of the spectrum, with a significant increase in the variance, where there is a translation motion of the protoplasm in the normal cell. The apparent reason for the decrease in the correlation time to ~ 0.2 s is a change in the particular realization in the volume being probed; with the known[7,8] velocity $v = 50$–$70\ \mu$m/s, we can estimate the correlation length to be ~ 10–$15\ \mu$m.

The contribution from the motion of the protoplasm is equal to the difference between spectra c and b in Fig. 3. A better model for it would apparently be a model of a steady-state random process consisting of bounded pulses arriving at random, including the case of Poisson distribution of pulse lengths,[9,10] which gives a flicker ($\sim F^{-2}$) or Lorentzian spectrum[9] with an intensity in the wings proportional to the variance σ_f^2 and an average pulse frequency Γ. The flicker nature of the spectra agrees with the results of measurements at lower frequencies.

Confirmation that the fluctuations due to vital activity in biological objects can be detected even in objects with small cells, without any apparent flow of the protoplasm, comes from the results of measurements carried out with plant leaves. The fluctuations in a living leaf are about an order of magnitude higher than those in a leaf after an inhibitor or heating has stopped biogenic processes.

To the best of our knowledge, these results are the first indication of the existence of intense low-frequency anisotropy fluctuations of biogenic origin.

These results demonstrate that the method of dynamic laser polarimetry is highly effective for studying low-frequency fluctuations of an anisotropy with large correlation radii in small-angle scattering of light.

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Translated by Dave Parsons