

Scattering of x and optical radiation in large-scale quasiperiodic structures

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The existence of a translational order in the blue phase of a cholesteric liquid crystal has been demonstrated by the methods of small-angle scattering of x radiation and optical scattering. Macroscopically porous glasses are observed to exhibit the external manifestations of the blue phase. Their correlation functions are analogous.

The particular features of the blue phase of cholesteric liquid crystals are seen most vividly in optical experiments.¹⁻⁶ It follows from those experiments and from a theoretical analysis¹ of the blue phase that the tensor order parameter characterizing the correlation in the orientations of the molecules has a three-dimensional spatial periodicity^{1,2} with a period on the order of 2000 Å. Whether this periodicity is accompanied by a modulation of the density has remained unclear. To resolve this problem, we have now carried out a study of small-angle x-ray scattering and of the optical scattering characteristic in the blue phase of a cholesteric liquid crystal and of macroscopically porous glasses. The method of x-ray structural analysis is a direct method for detecting density inhomogeneities.

The x-ray measurements were carried out in a small-angle Kratki chamber in a helium atmosphere at angles between 1.5' and 120' at the wavelength $\lambda = 1.54 \text{ \AA}$ (the $\text{CuK}\alpha$ line), corresponding to characteristic dimensions ranging from 3500 Å to 40 Å. The angular resolution was no worse than 0.35'. The error in the intensity measurements did not exceed 1%. The optical scattering intensity was studied on the apparatus described in Ref. 7. As light sources we used a He-Ne laser ($\lambda = 6328 \text{ \AA}$) and an argon laser ($\lambda = 5145 \text{ \AA}$, $\lambda = 4658 \text{ \AA}$). The scattering angle θ ranged from 30° to 160°. We studied polydomain samples of cholesterylolate at a temperature 0.4 °C below the point of the transition to the isotropic phase. This position corresponds to the temperature of the existence of blue phase II. During the recording of the x-ray diffraction patterns, the liquid crystal was placed between polyester films 15 μm thick; the background from these films was taken into account. The thickness of the sample layer did not exceed 1 mm. In the optical measurements we used an evacuated cylindrical cell with an inside diameter of 16 mm and a height of 40 mm, filled with a dust-free liquid crystal.

Figure 1 shows an x-ray diffraction pattern of blue phase II (line 1). We see a clearly defined peak with a maximum at the wave number $q = 3.7 \times 10^5 \text{ cm}^{-1}$ on this pattern. Shown for comparison here are lines for the isotropic phase (2) and the cholesteric phase (3), on which there is no peak, and the intensity falls off monotonically. It follows that the peak in the blue phase stems from a periodic variation of the

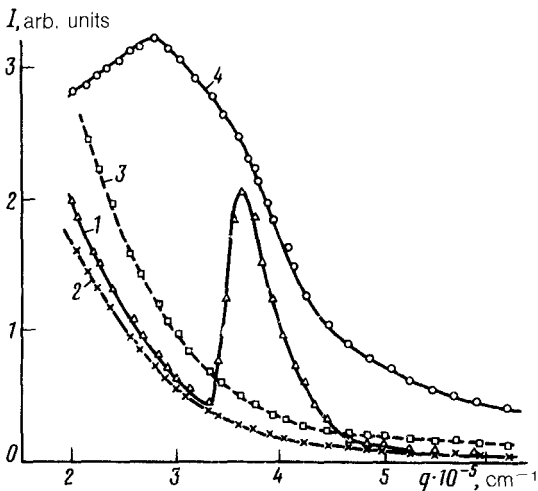


FIG. 1. Intensity of the scattered x radiation as a function of the wave number. 1—Blue phase II; 2— isotropic phase; 3—cholesteric phase ($T - T_c = 3^\circ\text{C}$); 4—macroscopically porous glass (the entrance slit is $20\ \mu\text{m}$ wide; the detector slit is $40\ \mu\text{m} \times 8\ \text{mm}$).

density (the lattice constant is $d \approx 1700\ \text{\AA}$) rather than from diffraction by the cholesteric helix.

Line 1 in Fig. 2 shows the correlation function $\varphi(q)$ as a function of the wave number according to measurements of the polarized component of the intensity of the scattered light. For this purpose, we wrote the measured intensity $I_v^v(\theta, \lambda)$ in the form

$$I_v^v(\theta, \lambda) = \text{const } \varphi(q) / \lambda^4,$$

where $\mathbf{q} = \mathbf{k}_i - \mathbf{k}_s$, $q = (4\pi n/\lambda) \sin \frac{\theta}{2}$ and \mathbf{k}_i and \mathbf{k}_s are the wave vectors of the incident and scattered light.

The maxima on lines 1 in Figs. 1 and 2 coincide. It follows from this coincidence

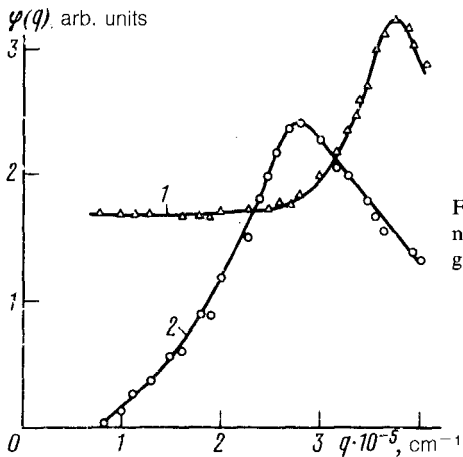


FIG. 2. Correlation function as a function of the wave number. 1—Blue phase II; 2—macroscopically porous glass impregnated with benzene.

that in the x-ray and optical measurements we are studying structures having the same lattice constant, although the reasons for the scattering are different. The x-ray scattering is a scattering by electron density fluctuations, while the intensity of the scattered light is dominated by fluctuations in the tensor order parameter. Evidence for this conclusion comes from the value of the depolarization coefficient, which is 0.75, after corrections for the rotation of the polarization plane. The data on the optical scattering can be used to estimate the size of the uniformly ordered regions; it turns out to be 8–10 lattice constants.

We do not carry out corresponding estimates for the x-ray results, since the half-width of the peak for line 1 in Fig. 1 is seriously distorted by the instrumental function of the apparatus.

It follows from the behavior of the function $\varphi(q)$ that for scattering angles $\approx 90^\circ$ the intensity of the optical scattering (the factor of $1/\lambda^4$ is taken into account) for the blue light is nearly an order of magnitude greater than that for the red light. This is the reason for the characteristic color of the blue phase.

It is reasonable to suggest that any transparent structure having a correlation function of this sort should exhibit the external manifestations of the blue phase. A convenient object for testing this suggestion is macroscopically porous glass,⁸ in which it is possible to create inhomogeneities with prespecified average dimensions. The particular sample which we used was synthesized by leaching a sodium borosilicate glass with a pore dimension $\approx 1000 \text{ \AA}$. The distance between pores was $\approx 1500 \text{ \AA}$, and their volume fraction was 0.38. An electron micrograph revealed some order in the arrangement of the elements of the solid phase. For the optical measurements, the pores were filled with an immersion fluid. The samples became transparent and were indistinguishable from the blue phase of cholesteric liquid crystals in visual observations.

The optical scattering characteristic was measured on cylindrical samples 5 mm in diameter and 4 cm high, which were mounted in the cell in such a way that the cell and the sample were coaxial. As the immersion fluid we used benzene at 53.7°C , which we poured into the cell. For the x-ray measurements we used wafers ranging in thickness from 0.1 to 0.2 mm.

Line 4 in Fig. 1 and line 2 in Fig. 2 show the results of the x-ray and optical measurements for the macroscopically porous glass. These lines, like the lines for the blue phase II, have maxima; the positions of these maxima coincide and correspond to a characteristic dimension $\approx 2200 \text{ \AA}$. The degree of depolarization of the scattered light turns out to be 0.1. In other words, in contrast with the blue phase, the scattering is caused primarily by scalar irregularities of the refractive index. Another distinction is that in the porous glasses the scattering component, which is unrelated to the spatial structure, is negligible.

In summary, these experiments show that, regardless of the molecular nature of the substance, the presence of spatially correlated, large-scale irregularities leads to similar optical manifestations. Generating a detailed description of such structures requires carrying out two types of experiments jointly: optical scattering and small-angle x-ray scattering.

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