Neutron-Scattering Study of Transitions to Convection and Turbulence in Nematic Para-azoxyanisole

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The critical temperature gradients for the onset of convection and turbulence were measured in nematic para-azoxyanisole. The transition to turbulence was found to occur through two successive instabilities. The first of these clearly signals the entrance to a periodic-motion regime, and the second, although less clearly, to a regime with nonperiodic, time-dependent motion. A typical phase-transition behavior was observed at the transitions to turbulence.

The behavior of fluids near hydrodynamic instabilities, a classical field of study in fluid mechanics,[1] has recently become a topic of general physics. A number of theoretical papers have called attention to the fluctuations near the instabilities which mark the onset of convection[2,3] and turbulence,[4,5] and to the phase-transition analogies of these instabilities. The possibility of using light scattering to probe such transitions has been pointed out[6] and realized experimentally for the convective instability[7] and for the related Taylor instability.[8] We have used the neutron-scattering technique to locate the convective instability and to study the time dependence of fluctuations near the onset of turbulence in a nematic liquid.

When a vertical temperature gradient ΔT is imposed on a liquid, the quiescent state breaks down when ΔT exceeds a critical value ΔT_c. This phenomenon is usually referred to as the Rayleigh-Bénard problem. The convective flow pattern for $\Delta T > \Delta T_c$ is one of rolls, the specific form of which is influenced by the geometry of the sample container. By further increase of ΔT the steady-state convective motion becomes unstable to time-dependent, turbulent motion. Theory predicts that the transition to turbulence involves at least two instabilities and a regime of periodic motion with wavy convection rolls.[5] Local temperature measurements and visual observations on mercury and air showed the existence of such periodic motion.[9] Recent high-resolution thermal measurements on liquid He^4 revealed the broad frequency spectrum of fully developed turbulence, but not the periodic motion.[10]

Nematic liquid crystals are good candidates for experiments on convective instabilities, mainly because ΔT_c is lower by a factor of 10^2–10^3 compared with isotropic liquids. In addition they present the possibility of studying the stabilizing effects of external fields. Convective instabilities in nematics have been extensively studied in a series of papers by the Orsay group.[11] To the best of our knowledge, turbulence has not earlier been investigated in such substances.

It has earlier been demonstrated[12] that the intensity of neutrons scattered coherently from a nematic substance depends strongly on the orientation of the molecules with respect to the scattering vector. In the present experiment on para-azoxyanisole (PAA) a field of 50 G along the horizontal scattering vector was sufficient to extinguish the intensity of the liquid diffraction peak at 1.7 $Å^{-1}$, in the absence of flow. Hence a field of 50 G is sufficient to introduce a Fréedericksz transition in the nematic range for the geometry of our experiment. At the onset of flow, because of the coupling that exists between orientational and translational motions in a nematic substance,[13] the coherent intensity reappears and increases with the flow. At any instant the molecules are at an angle defined by the resultant of the viscous torques and the magnetic torque.

Our sample was fully deuterated PAA. The solid-nematic and the nematic-isotropic transition temperatures were measured to be 119 and 135°C, respectively. The sample was contained in a cylindrical aluminum vessel with inner diameter 5 mm and height 25 mm. Controlled heating from above and below makes it possible to

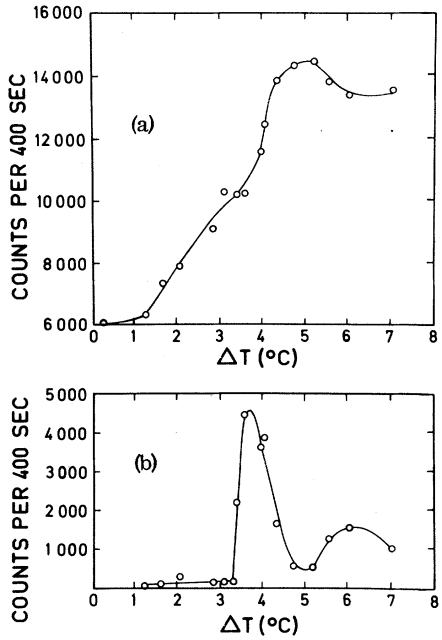

FIG. 1. (a) Average neutron intensity (I_0) observed as a function of the vertical temperature difference (ΔT) across the sample. (b) Variance $\langle \Delta I^2 \rangle = \langle (I - I_0)^2 \rangle$, i.e., mean square of fluctuations of intensity about the time average, as a function of ΔT.

keep a mean temperature and a chosen gradient constant within ± 0.02°C for days. The temperatures were measured with Pt resistance thermometers and printed out at 10-min intervals. Neutron data were printed out every 400 sec and the total measuring time was on the average 24 h for each gradient setting. The data included in the present paper were all taken at sample temperatures near 104°C, in the supercooled nematic range, in order to get a large difference in counting rate as one passed from the conduction to the convection region. A field of 50 G was all the time applied along the scattering vector.

Figure 1(a) shows average intensities (I_0) recorded for several values of the gradient. The first increase of the intensity at $\Delta T = 1.25$°C marks the onset of convection. The second increase is accompanied by strong intensity fluctuations, as seen from Fig. 1(b), which gives $\langle I^2 \rangle = \langle (I - I_0)^2 \rangle$ at each gradient value. The fluctuations exhibit a large maximum at $\Delta T = 3.6$°C and a smaller maximum at $\Delta T \sim 6$°C. We shall suggest below that these correspond to transitions to turbulent periodic and nonperiodic regimes, respectively.

In Fig. 2 we give in more detail raw and processed intensity data for seven selected values of the gradient. For each value we give (on the top two lines) the directly recorded time dependence $I(t)$ of the intensity, the autocorrelation function $\langle \Delta I(0) \Delta I(t) \rangle$ of the fluctuations, and the spectral function obtained by taking the Fourier transform of the latter function. The following tendency is observed: The increase in $\langle \Delta I^2 \rangle$ marks the passage to time dependence of the fluctuations; the first maximum falls in a region of periodic motion and the second maximum marks the passage to a less periodic and probably time-dependent regime. Unfortunately the maximum temperature difference obtainable was ~ 7°C which prevented us from investigating this last point more thoroughly.

In the valley between these two peaks the autocorrelation function has very little structure, while I_0 reaches a rounded maximum. It is instructive to follow the development of the spectral function as we pass through the first maximum of ΔI^2. Initially, at $\Delta T = 3.4$°C, there are two sidebands at relatively high frequencies. As we increase ΔT they move to lower frequencies. Near the maximum a wide central peak is clearly seen, and perhaps also a narrow one. The observation of a narrow central peak is of course difficult, since zero frequency corresponds to an infinitely long observation time. McLaughlin and Martin[5] have predicted four modes at the instability which marks the onset of periodic motion. Although we have not so far been able to distinguish as many as four modes clearly, it is very tempting to ascribe the maximum at $\Delta T = 3.6$°C to an instability of this type. The second instability that they predict is the lower limit of a time-dependent, nonperiodic regime. This transition takes place when the fourth mode becomes large. Although, as remarked above, we have not been able to establish clearly the nonperiodic character of the motion, it is tempting to identify our second maximum with their second instability.

The phase-transition character of our data is very striking, in particular that of the first maximum and its spectral functions. The observation of softening sidebands and an increasing central mode is a well-known phenomenon in structural phase transitions.[14] A pattern of coexisting sidebands and two central peaks, as indicated from our data at $\Delta T = 3.6$°C, would correspond closely to what is predicted for liquids in mode-mode coupling theory, i.e., sound-wave sidebands and two central peaks due to viscous flow and heat flow.[14]

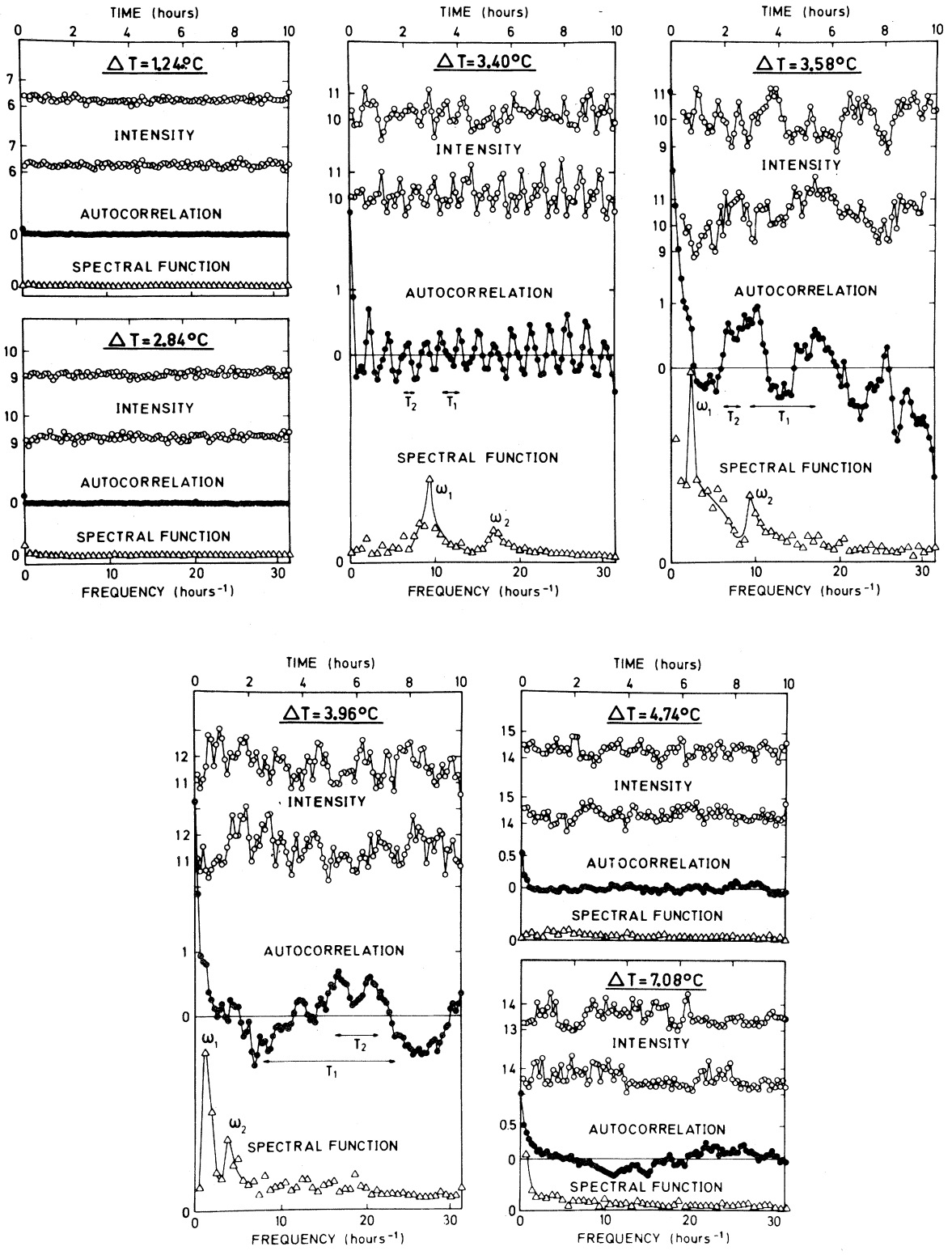

FIG. 2. For seven different values of ΔT are shown (1) the time variation of the neutron intensity $I(t)$ on two lines, (2) the autocorrelation function of the fluctuations $\langle \Delta I(0) \Delta I(t) \rangle$, and (3) the spectral function $|\int e^{i\omega t} \langle \Delta I(0) \times \Delta I(t) \rangle dt|$. Some characteristic frequencies, ω_1 and ω_2, and the corresponding periods, T_1 and T_2, are indicated.

An alternative statistical analysis of the fluctuations was tried by calculating $\langle \Delta I^3 \rangle$ and $\langle \Delta I^4 \rangle$ in addition to $\langle \Delta I^2 \rangle$. The ratio $\langle \Delta I^4 \rangle / \langle \Delta I^2 \rangle^2$ was calculated with the hope of finding a high value in the periodic regime.[5] The actual values found were always close to 3, as expected for a Gaussian distribution of fluctuations.

It should be noticed that we have left out a few data points, some of which do not fit to our curve for $\langle \Delta I^2 \rangle$. Some of them were left out because of high values for the third moment. Others were left out because they did not represent the final, stationary state to which the system passed when waiting long enough (several hours). These points evidently correspond to local minima of the thermodynamic potential and some of them have autocorrelation functions that display a very clear damped-harmonic-oscillator behavior.

The experiments will be continued with the aim of presenting more quantitative data. At present we know only qualitatively how the data of Fig. 1 are connected with the order parameters that have been defined for convection[2] and for periodic[5] and strongly turbulent flow.[4] This work represents a novel application of neutron scattering and of neutron-data analysis. We know of no other case where the neutron intensity oscillates on a time scale longer than the period of measurement. An effect of this is that the problem of energy resolution is converted to one of time and patience.

We first observed such instabilities when investigating the nematic-isotropic transition of PAA. The complicated time behavior and field dependence of the intensities caused us initially to draw some unjustified conclusions about the nature of this transition.[15] The instability at this transition, where two phases coexist, is probably of the type found in binary mixtures.[1]

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Antonoff's Rule and the Structure of Interfaces near Tricritical Points*

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The Van der Waals theory of surface tension and interface structure is extended to three-phase equilibrium. The theory predicts that near a tricritical point the three interfacial tensions will satisfy "Antonoff's rule" in the form $\sigma_{\alpha\gamma} = \sigma_{\alpha\beta} + \sigma_{\beta\gamma}$, where β is the distinguished phase, and that the composition profile of the $\alpha\gamma$ interface will be qualitatively different from that of the $\alpha\beta$ and $\beta\gamma$ interfaces.

Van der Waals's theory of the liquid-vapor interface[1-4] is summarized graphically in Fig. 1. In Fig. 1(a) is shown the analytic chemical-potential–density isotherm, $\mu(\rho)$, and the equal-areas