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Observation by Neutron Correlation Spectroscopy of a Nonlinear Soft Mode at the Rayleigh-Bénard Instability in Para-Azoxyanisole

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By use of neutron correlation spectroscopy a soft mode has been found for the Rayleigh-Bénard instability with frequency $\omega \sim (R/R_c - 1)^{0.5}$, R denoting the Rayleigh number. The power spectrum shows periodic states split into a narrow band, and some low, perhaps solitary, states. Guided by the autocorrelation function the main features are modeled by phase modulation of the soft mode by the slower modes. A connection is made to the central-mode problem in structural phase transitions.

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Recently critical fluctuations near the convective instability in para-azoxyanisole (PAA) were reported.[1] The fluctuations manifested themselves as excursions from the time average of the intensity of scattered neutrons. In this paper we report on the dynamics of these fluctuations. For the experiment we used a slab-type aluminum vessel of dimensions $38\times 38\times 5$ mm^3 filled with fully deuterated PAA. Of the long dimensions one is vertical; the other is horizontal and along the common direction of the scattering vector and a magnetic field. The temperature at midheight of the aluminum side walls was kept constant at 121 °C. PAA is then a nematic liquid crystal susceptible to alignment by a magnetic field. A vertical temperature difference ΔT is obtained by setting the difference of the power fed to the heating elements at the top and bottom of the vessel; positive ΔT denotes a warmer bottom than top. Convection sets in at a predictable value ΔT_c, the Rayleigh-Bénard instability point.[2] ΔT_c is proportional to R_c, the critical value of the dimensionless Rayleigh number R. In isotropic liquids $R = \beta g l^3 \Delta T (\nu\kappa)^{-1}$. Here β is coefficient of the thermal expansion, g the gravitational acceleration, l the vertical layer thickness, ν the kinematic viscosity, and κ the heat diffusivity. For our cell only one convection roll is predicted[3] and observed,[4] but higher harmonics develop gradually at increasing R.[5] Neutron scattering can explore these phenomena in nematic liquid crystals.[4] This method rests on the coupling between molecular orientation and flow, and on the anisotropic scattering power of the molecules. In the absence of flow the molecules are aligned horizontally by the field. The convection makes molecules tilt towards the vertical direction, which in turn gives increased neutron intensity

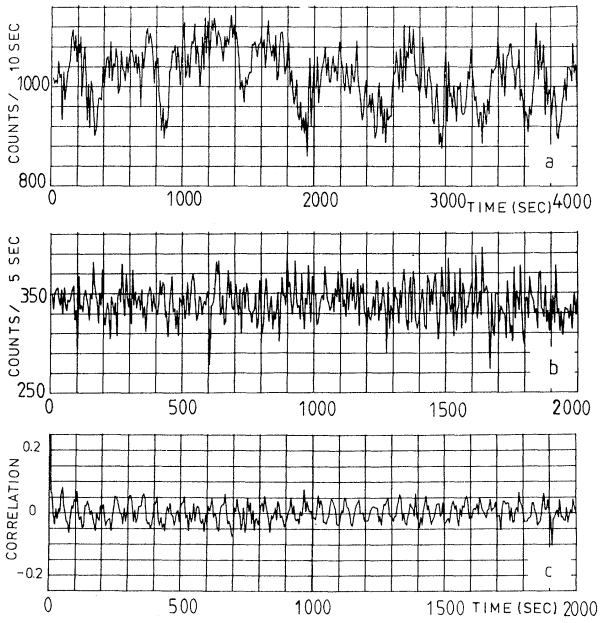

FIG. 1. (a), (b) Portions of data records showing time variation of neutron intensity in a liquid diffraction peak in nematic PAA. Intensities should not be directly compared. In (a) $\epsilon = 0.02$, in (b) $\epsilon = 10.7$. (c) Autocorrelation function of (b).

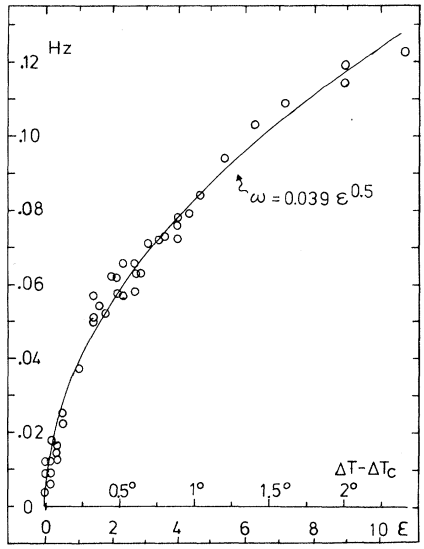

FIG. 2. Observed frequencies of concective oscillations vs reduced vertical temperature difference ϵ. The drawn curve is parabolic with fitted prefactor.

in the liquid diffraction peak at 1.8 $Å^{-1}$. Large-scale fluctuations of the convective velocity give fluctuations of the intensity. Liquid crystals are anisotropic liquids and the formula for R is slightly more complicated[6] than for isotropic liquids. An effect of this is that ΔT_c also depends on the magnitude of the field through the formula $T_c(H) = \Delta T_c(0)(1 + H^2 H_c^{-2})$. H_c is the critical field above which molecular alignment is achieved. We have earlier[4] found $\Delta T_c(0) = 2.4 \times 10^{-3}$ K and $H_c = 24$ Oe. In the present experiment the field was 240 Oe. The neutron intensity was sampled 2048 or fewer times at Δt intervals; Δt was chosen in the range 5 to 40 sec. Each data record was stored in a computer memory and the subsequent processing gave the autocorrelation function $C(\tau)$ and the power spectrum $P(\omega)$. Figures 1(a) and 1(b) show portions of raw data for $\epsilon = 0.02$ and 10.8, where $\epsilon = R/R_c - 1$. In Fig. 1(a) even the raw data display oscillations. For Fig. 1(b) the calculated $C(\tau)$, shown in Fig. 1(c), also reveals an oscillating state. Through $C(\tau)$ or $P(\omega)$, or both, we arrive at Fig. 2. The curve drawn is $\omega = A\epsilon^{0.5}$ with a fitted A of 0.039. When $\epsilon \lesssim 5$ the power spectra at first sight look chaotic; as an example see Fig. 3. $C(\tau)$ is amplitude modulated with an easily distinguishable frequency at $\omega_0 = 0.062$ Hz. $P(\omega)$ has only a relatively weak line at ω_0, but a surrounding multiplet of equidistant lines. The multiplet can be explained by phase modulation of ω_0 by the lowest frequencies in the spectrum, of the type $\cos[\omega_0 t + a \sin(\omega t)]$. The amplitude of the nth line in the multiplet is given by $J_n^2(a)$, the square of a Bessel function.[7] Taking $2\omega_1$ and $6\omega_1$ in Fig. 3 as phase-modulating frequencies we reproduce the main features of the multiplet. If to this modulated wave we add the

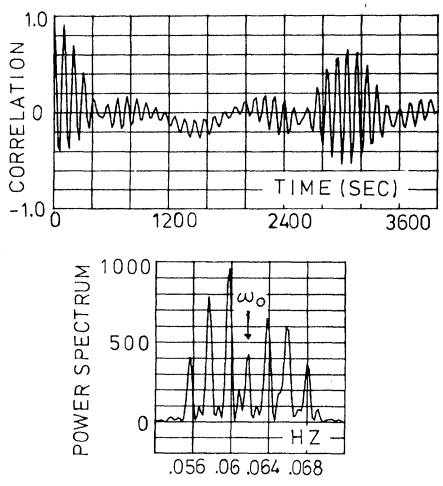

FIG. 3. Processed data from observations at $\epsilon = 2$. Top panel: portion of autocorrelation function. Lower panels: power spectrum.

slow, modulating waves, we also reproduce the main features of $C(\tau)$. Modeled portions of $C(\tau)$ and $P(\omega)$ are shown in Fig. 4, which should be compared with Fig. 3. Phase modulation also has the desired effect of producing oscillations asymmetric with respect to time reversal, an aspect noticed in earlier data[1] and also seen in Fig. 1(a) above.

Kvernvold[5] has predicted oscillations for $\epsilon \sim 0$ in convection cells of the present geometry, but gives no formula for ω. Degiorgio[8] predicts $\omega = A(\epsilon - \epsilon_t)^{0.5}$, where $A = 73\kappa l^{-2}$ and $\epsilon_t = 0.3$. For our sample and geometry this gives $\omega = 4\times 10^{-3}(\epsilon - 0.3)^{0.5}$. The discrepancy between calculated and observed values of A and ϵ_t could possibly be due to uncertainties in the data and ϵ scale for $\epsilon \sim 0$. To extract reliably the low soft-mode frequencies we need better knowledge of the resolution problem and of the mode coupling. Consequently the data at $\epsilon < 1$ may have systematic errors. Unfortunately the sample was ruined by accidental overheating as we were exploring this important region and we had to rely on the data and the ϵ scale established in earlier experiments. Degiorgio's predictions[8] are for *transient*, relaxational oscillations following a sudden change of ϵ. We are suggesting that these hydrodynamic oscillations are sustained by coupling to spontaneous, microscopic fluctuations. In nematics orientational fluctuations represent an extremely strong source of agitation. In isotropic liquids one may also expect to observe the same phenomenon, and Bergé and Dubois[9] have, in fact, observed stimulated oscillations with $\omega \sim \epsilon^{0.5}$ for $\epsilon > 5$ in measurements of the local velocity field.

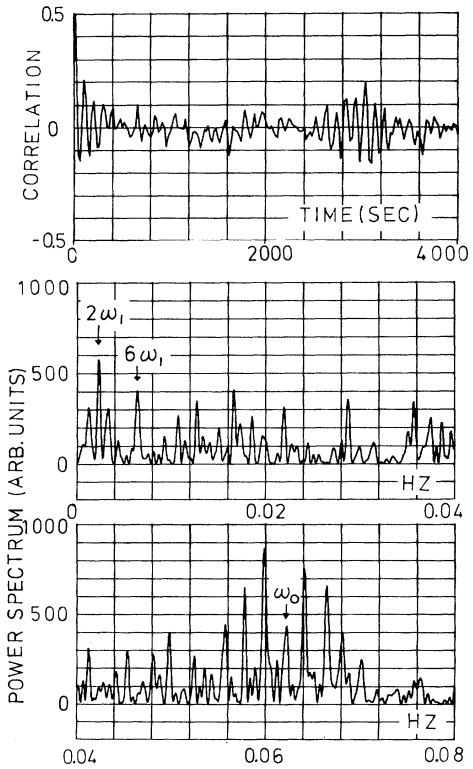

FIG. 4. Autocorrelation function and portion of power spectrum at $\epsilon = 2$ obtained by modulating phase of ω_0 with frequencies $2\omega_1$ and $6\omega_1$ of Fig. 3.

We can offer no detailed model for the mode coupling that we invoke. Modulation, subharmonics, and higher harmonics are, however, common characteristics of nonlinear phenomena[10] and have received renewed interest in the current upsurge of soliton physics. We plan a new series of experiments with improved thermometry to deduce the temperature variation of the frequencies and coupling constants involved. The main mode observed has, however, the characteristics of soft modes commonly observed in solid-state phase transitions, and lends strong support to the phase-transition analogy of hydrodynamic instabilities. The simplicity of the system, a single cell with essentially single-mode dynamics, makes it an attractive one for phase-transition studies. The power spectra above may, e.g., contain a clue to the long-standing central-mode problem[11] in structural phase transitions.

The oscillatory states reported here are one or two orders of magnitude faster than those reported before[12-14] at higher ϵ. While writing this paper we became aware of a recent paper by Maurer and Libchaber[15] in which they study the onset of turbulence in liquid helium. In a two-mode regime they find, as we do, that the phase of one frequency is modulated by the other.

Correlation spectroscopy, first used in neutron scattering by Møller and Riste,[12] can measure frequencies a factor of 10^{10} lower than conventional neutron spectroscopy. We foresee several other applications of this technique in liquid crystals, and possibly also in solids.

In conclusion, the present work relates the fluctuating component[1] of neutron scattering for $\epsilon \sim 0$ to a previously unobserved, nonlinear soft mode of the convective state. We suggest that the mode is driven by spontaneous, microscopic fluctuations coupled to the convective velocity.

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