CRITICAL PHENOMENA NEAR BIFURCATIONS IN NONEQUILIBRIUM SYSTEMS*

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The usual deterministic description of spatially-extended nonlinear dissipative systems far from equilibrium yields a sharp bifurcation point at a critical value $R = R_c$ of the control parameter where the system makes a transition from the spatially uniform ground state to a state with spatial variation. However, when the effect of thermal noise is considered, then even below the bifurcation there are fluctuations of the macroscopic variables away from the uniform state and the relevant fields, although they have zero mean, have a positive mean square. Here we review measurements of the properties of these fluctuations. In the case of Rayleigh-Bénard convection (RBC) in common fluids, fluctuation amplitudes are small and the exponent of the powerlaw which describes their mean square has its classical (mean-field) value $\gamma_{MF} = 1/2$ in experimentally accessible parameter ranges. However, for RBC of a fluid near its liquid-gas critical point fluctuation amplitudes are much larger and nonlinear interactions between them yield a first-order transition as predicted by Swift and Hohenberg. Electroconvection in nematic liquid crystals (NLC) does not belong to the same universality class as RBC, and fluctuation interactions leave the bifurcation supercritical; but the critical behavior is renormalized.

1. Introduction

Bifurcations in spatially extended dissipative systems are usually discussed in terms of deterministic equations for the macroscopic variables which neglect thermal noise. Many such “ideal” systems undergo a sharp bifurcation at a critical value of a control parameter, where a spatially uniform state loses stability and a state with spatial variation appears. However, if noise is present, it will drive the system to fluctuate away from the uniform state, even below the bifurcation. As near a thermodynamic critical point, the fluctuation amplitudes grow as the bifurcation is approached because the susceptibility diverges there. Using the stochastic hydro-

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dynamic equations introduced by Landau and Lifshitz,\textsuperscript{1} this problem was considered theoretically three decades ago\textsuperscript{2,3,4} for the case of Rayleigh-Bénard convection (RBC), which is the buoyancy-induced motion in a shallow horizontal layer of fluid heated from below. For RBC the deterministic model predicts pure conduction until the temperature difference $\Delta T$ exceeds a critical value $\Delta T_c$. In the presence of noise, time-dependent fluctuating flows are predicted to occur even for $\Delta T < \Delta T_c$. They have zero mean, but their mean-square (ms) amplitude is finite. Near the bifurcation and when nonlinear saturation is neglected, this ms amplitude is proportional to $|\epsilon|^{-1/2}$ where $\epsilon \equiv \Delta T/\Delta T_c - 1$, i.e. it diverges at $\Delta T_c$. In spite of the divergence, these fluctuations induced by thermal noise were expected to be unobservably weak because the thermal energy $k_B T$ ($k_B$ is Boltzmann’s constant) which drives them is many orders of magnitude smaller than the typical kinetic energy of a macroscopic convecting fluid element. Nonetheless, it has now become possible to observe the fluctuating convection patterns below the bifurcation directly and to make quantitative measurements of their rms amplitudes.

Fig. 1. Root mean square director angle fluctuations $\langle \theta^2 \rangle^{1/2}$ as a function of $\epsilon \equiv V^2/V_c^2 - 1$. The three lines represent three different models for the fluctuations (see Ref. \textsuperscript{5}). The solid line corresponds to the prediction Eq. 1. After Ref. \textsuperscript{5}.

2. Results in the linear range

When the fluctuation amplitudes are small enough, their interactions with each other can be neglected and the amplitudes can be described well by stochastic linearized hydrodynamic equations. The first system for which quantitative measure-
ments were made was electroconvection (EC) in a nematic liquid crystal (NLC). This system consists of a thin layer of a NLC confined between parallel glass plates, with the director aligned in a predetermined direction parallel to the plates. When an alternating electric voltage of amplitude $V$ is applied between electrodes deposited on the insides of the glass plates, convection occurs above a threshold voltage $V_c$. Even though that system is "macroscopic", it is particularly susceptible to noise because the physical dimensions are only of order $10$ µm and because the elastic constants (which determine the macroscopic energy to which $k_B T$ has to be compared) are exceptionally small. Measurements for this system of the mean square director angle fluctuations $\langle \theta^2 \rangle^{1/2}$ for $V < V_c$ are shown in Fig. 1. They can be described very well by the prediction

$$\langle \theta^2 \rangle = F_{EC} / |\epsilon|^{1/2}$$

of linear theory, as shown by the solid line in the figure (for electroconvection $\epsilon \equiv V^2/V_c^2 - 1$). Only the coefficient $F_{EC}$ was adjusted to fit the data; its best value was found to be of the same size as the approximate theoretical estimate

$$F_{EC} \approx k_B T / \bar{k}d$$

where $\bar{k}$ is an average elastic constant of the LC. Quantitative calculations of $F_{EC}$ based on fluctuating hydrodynamics have not yet to been carried out.

More recently, thermally driven fluctuations were observed also for RBC, and quantitative measurements of their amplitudes were made by Wu et al. In part these measurements were made possible by the development of experimental techniques for the study of RBC in compressed gases. There it is possible to use sample spacings an order of magnitude smaller than for conventional liquids and kinematic viscosities are relatively small, thus making the systems more susceptible to noise. In addition, maximizing the sensitivity of the shadowgraph method and careful digital image analysis have enhanced the experimental resolution.

In the left part of Fig. 2 we show a processed image of a layer of CO$_2$ of thickness 0.47 mm at a pressure of 29 bars and at a mean temperature of 32.0°C. The sample was at $\epsilon = -3 \times 10^{-4}$, very close to but just below the bifurcation point. The fluctuating pattern is barely detectable by eye. The right half of the figure shows the average of the structure factors (squares of the moduli of the Fourier transforms) of 64 such images. It demonstrates clearly that the fluctuations have a characteristic wavenumber $q$. The value of $q$ is in quantitative agreement with the critical wavenumber $q_c = 3.117$ for RBC. The ring in Fourier space is azimuthally uniform, reflecting the continuous rotational symmetry of the RBC system.

The power contained within the ring in Fourier space can be converted quantitatively to the mean-square amplitude of the temperature field. Results for the temporal and spatial averages $\langle \delta T^2 \rangle$ of the square of the deviations of the temperature from the local time average (pure conduction) as a function of $\epsilon$ at two different sample pressures are shown in Fig. 3 using logarithmic scales. The data can
Fig. 2. Left: Shadowgraph snapshot of fluctuations below the onset of convection ($\epsilon = -3 \times 10^{-4}$). Right: The average of the square of the modulus of the Fourier transform of 64 images like that on the left. After 7.

Fig. 3. Mean square amplitudes of the temperature fluctuations below the onset of convection of a layer of CO$_2$ of thickness 0.47 mm and a mean temperature of 32°C. The solid (open) circles are for a sample pressure of 42.3 (29.0) bars. The two lines are the theoretical predictions. Note that there are no adjustable parameters. After 7.

be described quite accurately by straight lines with slopes close to -1/2, consistent with the powerlaw $\langle \delta T^2 \rangle \propto \epsilon^{-1/2}$ as predicted by theory.

The amplitudes of the fluctuating modes below but close to the onset of RBC were calculated quantitatively from the linearized stochastic hydrodynamic equations$^1$ by van Beijeren and Cohen$^{10}$, using realistic (no-slip) boundary conditions at the top and bottom of the cell. For the mean square temperature fluctuations their
results give\(^11,7\)

\[
\langle \delta T^2(\epsilon) \rangle = \tilde{c}^2 \left( \frac{\Delta T_c}{R_c} \right)^2 \frac{F}{4\sqrt{-\epsilon}},
\]

with \(\tilde{c} = 3q_c\sqrt{R_c} = 385.28\). Here \(R_c = 1708\) is the critical Rayleigh number, and the noise intensity \(F\) is given by

\[
F = \frac{k_B T}{\rho \nu^2} \times \frac{2\sigma q_c}{\xi_o \tau_o R_c},
\]

with \(\xi_o = 0.385\) and \(\tau_o \simeq 0.0796\). One sees that \(F\) depends on the density \(\rho\) and kinematic viscosity \(\nu\), as well as on the Prandtl number \(\sigma = \nu/\kappa_T\) (\(\kappa_T\) is the thermal diffusivity). Using the fluid properties of the experimental samples,\(^9\) one obtains the straight lines in Fig. 3. Since there are no adjustable parameters, the agreement between theory and experiment can be regarded as excellent. This agreement lends strong support to the validity of Landau’s stochastic hydrodynamic equations\(^4\).

3. Results in the nonlinear range

Sufficiently close to the bifurcation, where fluctuation amplitudes become large, nonlinear interactions between them play a role and linear theory breaks down. In this regime the nonlinear interactions induce deviations from the usual mean-field behavior implied by Eqs. (1) and (3).\(^4,11\) In that case genuine critical phenomena which differ from mean-field predictions are expected, and the precise critical behavior should depend on the symmetry properties and the dimensionality of the system.

Deviations from the prediction of linear (mean field) theory have indeed been observed recently for electroconvection in nematic liquid crystals.\(^12,13\) In Fig. 1 we show the results for the mean square director angle fluctuations \(\langle \theta^2 \rangle\) of the NLC I52 (4-ethyl-2-fluoro-4′-[2-(trans-4-pentylcyclohexyl)ethyl]-biphenyl) as a function of \(\epsilon \equiv V^2/V_c^2 - 1\). The open symbols correspond to data for \(V < V_c\) or \(\epsilon < 0\). At large \(|\epsilon|\) (near the right of the figure) the data are consistent with mean-field theory which is given by the short dashed line. However, the mean-field prediction diverges close to \(\epsilon = -0.02\) which is indicated by the vertical dashed line. One sees that the actual onset voltage \(V_c\) for the ordered state has been shifted by the fluctuations to values higher than the mean-field onset at \(V_{c,MF}\). The growth of the fluctuation intensities upon approach toward \(|\epsilon| = 0\) can be described by an effective powerlaw over a range of about two decades, with an exponent close to 0.25 (solid line through the open symbols), to be compared with the corresponding mean-field exponent of 1/2. Of course this apparent powerlaw growth of the fluctuations can not continue indefinitely since \(\langle \theta^2 \rangle\) must remain finite at the transition. Thus, actually there must be a continuous and finite crossover function which describes the evolution of \(\langle \theta^2 \rangle\) from below to above onset. The quantitative nature of this function is not known at this time. The solid symbols in the figure correspond to the ordered state.
for $\epsilon > 0$. They can be described well by the classical exponent $2\beta = 1$, and were used to determine $V_c$. Clearly this fascinating phenomenon deserves more detailed theoretical attention.

![Fig. 4. The mean square director angle fluctuations $\langle \theta^2 \rangle$ as a function of $|\epsilon|$. Open symbols: $\epsilon < 0$. Solid symbols: $\epsilon > 0$. Short dashed line: mean field (or linear) prediction. Vertical dashed line: mean field onset at $V_{c,M_F}$. Solid line: an effective powerlaw with an exponent of 0.25. Dotted line through the solid symbols: a powerlaw with exponent $2\beta = 1$.](image)

For RBC, Swift and Hohenberg were able to show that the system belongs to the same universality class as one considered by Brazovskii.\textsuperscript{4,11,14} Equilibrium systems belonging to this class include the crystallization of di-block co-polymers.\textsuperscript{18} For this universality class the transition is of second order at the mean-field level, but the fluctuations induce a first-order transition. A common feature of all the systems belonging to this class is that the order parameter near the bifurcation has a relatively large volume of phase space accessible to it. In the RBC case this is reflected in the rotational invariance of the system as demonstrated by the ring in Fourier space shown in Fig. 2. On the basis of this qualitative consideration one would not expect the electroconvection system discussed above\textsuperscript{12,13} to belong to the Brazovskii universality class because the anisotropy due to the director leads to only one or two pairs of spots in Fourier space.

For RBC in ordinary liquids one can estimate\textsuperscript{4} that nonlinear fluctuation effects should be observable typically only for $|\epsilon| \leq 10^{-6}$, which has not been accessible to experiments so far. For RBC in compressed gases the critical region is a bit wider, reaching as far out as $|\epsilon| \simeq 10^{-5}$; but as can be seen from Fig. 3, this too has been beyond experimental resolution. However, the situation is much more favorable near a liquid-gas critical point (CP). Part of the reason for this can be seen by inspecting Eq. 4. and the phase diagram of SF$_6$ shown in Fig. 5. In that figure we show the temperature-density plane near the CP. The vertical dotted line
Fig. 5. The temperature-density plane near the critical point of SF<sub>6</sub>. The dashed line is the coexistence curve separating liquid and vapor. The vertical dotted line is the critical isochore. The solid line is the critical point \( T_c = 45.567^\circ C, \rho_c = 37.545 \text{ bars} \) and \( \rho_c = 0.742 \text{ g/cm}^3 \).

The solid lines represent the isobars \( P = 38.10 \text{ bars} \) (lower line) and 39.58 bars (upper line) used extensively in experiments. The heavy solid lines, each ending in two circles, illustrate the density range spanned during measurements with \( \Delta T \approx \Delta T_c \) for a cell of spacing \( d = 34.3 \mu m \) (lower line) and \( d = 59.1 \mu m \) (upper line).

corresponds to the critical isochore, and the two solid lines are isobars. As the CP is approached on the critical isochore from higher temperatures, the viscosity \( \nu \) has only a mild singularity and remains finite, whereas the Prandtl number \( \sigma = \nu / D_T \) diverges because the thermal diffusivity \( D_T \) vanishes. Thus the divergence of \( \sigma \) at finite \( \nu \) leads to a divergence of \( F \). An equally important aspect is, however, that the fluid properties are such that typical sample spacings \( d \) which can be used are in the range of 10 to 100 \( \mu m \), thus increasing \( F \) by one or two orders of magnitude compared to liquids and compressed gases away from the critical point. Another factor which greatly increases the experimental shadowgraph resolution near the CP is the value of the temperature derivative of the refractive index \( dn/dT \). Typically we have \( |dn/dT| \approx 0.1 \), whereas for ordinary fluids it tends to be two or three orders of magnitude smaller.

In Fig. 6 we show shadowgraph snapshots of fluctuations and roll patterns for a cell of spacing \( d = 59.1 \mu m \) at a pressure \( P = 39.58 \text{ bars} \) corresponding to the upper isobar shown in Fig. 5. The mean temperature \( T = 48.000^\circ C \) was kept...
constant during the experiments and had a value which corresponded to the critical isochore. When the applied temperature difference was equal to $\Delta T_c$, the sample occupied the heavy section of the line representing the isobar. The theoretical value of $F$ was $5 \times 10^{-4}$ for this case. The images are for several $\epsilon$ values. The bottom row shows the moduli of their Fourier transforms. One sees that it was possible to visualize the fluctuations even well below $\Delta T_c$. Just above onset the pattern consisted of convection rolls, as predicted for the deterministic system. The images at $\epsilon = -0.001$ and +0.001 demonstrate the existence of a sharp transition from fluctuations to a roll pattern, as would be expected for a subcritical bifurcation (or first-order phase transition) in the presence of strong noise.

4. Future prospects

From the viewpoint of critical phenomena electroconvection in NLCs is particularly rich. There is a number of different cases, with different symmetry properties of the normal forms (Ginzburg-Landau equations) which describe them. For the planar alignment (director parallel to the confining glass plates) discussed above, the convection rolls can have their axes orthogonal to the director (normal rolls), or these axes can be at an oblique angle to the director (oblique rolls). In both cases, the bifurcation can be stationary, or it can be a Hopf bifurcation to a time-periodic state. As a function of the drive frequency, the angle of obliqueness changes, and the point where it vanishes (i.e. where oblique rolls turn into normal rolls) is known as a Lifshitz point. The Lifshitz point has its own distinct symmetries with different critical behavior even at the mean-field level. The systems become even richer when one considers homeotropic alignment (alignment of the director orthogonal
to the glass plates). In that case EC is usually preceded by a Freédericksz transition which removes the rotational degeneracy of the system; but under very special conditions\textsuperscript{25} it is possible to achieve electroconvection without a prior Freédericksz transition. In this last case EC should be in the same universality class as RBC and show a fluctuation-induced first-order transition. How many of the different systems mentioned here will actually yield different universality classes of critical behavior is not known at this time. So far only the Brazovskii case has been studied theoretically. Experimentally only the case discussed above and illustrated by Fig. 1 has been investigated thoroughly.\textsuperscript{12,13} It corresponds to a Hopf bifurcation to oblique rolls. However early measurements for a stationary bifurcation to oblique rolls suggest critical behavior which differs from the Hopf case and imply the existence of more than one universality class.\textsuperscript{26} Clearly a great deal of experimental and theoretical work remains to be done in this field.

References

15. This feature was noted also by M. Assenheimer and V. Steinberg, \textit{Europhysics News} \textbf{27}, 143 (1996).

20. Our results differ from measurements by Roy and Steinberg, who found hexagons just above onset for experimental parameters believed to be similar to ours.


