Rayleigh-Bénard convection in a homeotropically aligned nematic liquid crystal

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We report experimental results for convection near onset in a thin layer of a homeotropically aligned nematic liquid crystal heated from below as a function of the temperature difference \( \Delta T \) and the applied vertical magnetic field \( \vec{H} \). When possible, these results are compared with theoretical calculations. The experiments were done with three cylindrical cells of aspect ratios (radius/height) \( \Gamma = 10.6, 6.2, \) and 5.0 over the field range \( 8 \lesssim h \equiv H/H_F \lesssim 80 \) (\( H_F = 20.9, 12.6, \) and 9.3 Gauss are the Fréedericksz fields for the three cells). We used the Nusselt number \( \mathcal{N} \) (effective thermal conductivity) to determine the critical Rayleigh number \( R_c \) and the nature of the transition. We analysed digital images of the flow patterns to study the dynamics and to determine the mean wavenumbers of the convecting states. For \( h \) less than a codimension-two field \( h_{ct} \approx 46 \) the bifurcation is subcritical and oscillatory, with travelling- and standing-wave transients. Beyond \( h_{ct} \) the bifurcation is stationary and subcritical until a tricritical field \( h_t = 57.2 \) is reached, beyond which it is supercritical. We analysed the patterns to obtain the critical wavenumber \( k_c \) and, for \( h < h_{ct} \), the Hopf frequency \( \omega_c \). In the subcritical range we used the early transients towards the finite-amplitude state for this purpose. The bifurcation sequence as a function of \( h \) found in the experiment confirms the qualitative aspects of the theoretical predictions. Even quantitatively the measurements of \( R_c \), \( k_c \) and \( \Omega_c \) are reproduced surprisingly well considering the complexity of the system. However, the value of \( h_{ct} \) is about 10% higher than the predicted value and the results for \( k_c \) are systematically below the theory by about 2% at small \( h \) and by as much as 7% near \( h_{ct} \). At \( h_{ct} \), \( k_c \) is continuous within the experimental resolution whereas the theory indicates a 7% discontinuity. The theoretical tricritical field \( h_t^{cb} = 51 \) is somewhat below the experimental one. The fully developed flow above \( R_c \) for \( h < h_{ct} \) has a very slow chaotic time dependence which is unrelated to the Hopf frequency. For \( h_{ct} < h < h_t \) the subcritical stationary bifurcation also leads to a chaotic state. The chaotic states persist upon reducing the Rayleigh number below \( R_c \), i.e. the bifurcation is hysteretic. Above the tricritical field \( h_t \), we find a bifurcation to a time independent pattern which within our resolution is non-hysteretic. However, in this field range, there is a secondary hysteretic bifurcation which again leads to a chaotic state observable even slightly below \( R_c \). We discuss the behavior of the system in the high-field limit, and show that at the largest experimental field values \( R_c \) and \( k_c \) are within 6% and 1%, respectively, of their values for an infinite field.

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I. INTRODUCTION

Convection in a thin horizontal layer of an isotropic fluid heated from below by a heat current \( Q \) is well known as Rayleigh-Bénard convection (RBC) [1,2]. When the fluid is a nematic liquid crystal (NLC), this phenomenon is altered in interesting ways. [3] NLC molecules are long, rod-like objects which are orientationally ordered relative to their neighbors, but whose centers of mass have no positional order [4]. The axis parallel to the average orientation is called the director \( \hat{n} \). By confining the NLC between two properly treated parallel plates [5], one can obtain a sample with uniform planar (parallel to the surfaces, i.e. in the x-y plane) or homeotropic (perpendicular to the surfaces, or parallel to the z-axis) alignment of \( \hat{n} \). The alignment can be re-inforced by the application of a magnetic field \( \vec{H} \) parallel to the intended direction of \( \hat{n} \). This is so because the diamagnetic susceptibility is anisotropic, usually being larger in the direction parallel to the long axis of the molecules. The phenomena which occur near the onset of convection depend on the orientation of \( \hat{n} \) and \( \vec{H} \). [6] In this paper we are concerned with a horizontal layer of a homeotropically aligned NLC in a vertical magnetic field \( \vec{H} = H\hat{z} \) and heated from below. In that case, \( Q = Q\hat{z} \) is parallel to \( \hat{n} \) when the system is in the conduction state. At a critical temperature difference \( \Delta T_c(H) \) the fluid will undergo a transition from conduction to convection. The precise value of \( \Delta T_c(H) \), the nature of the bifurcation at \( \Delta T_c(H) \), and the pattern-formation phenomena beyond \( \Delta T_c(H) \) are expected to depend in interesting ways upon \( H \). [7,8,9,10]

A feature common to the homeotropic NLC and to an isotropic fluid heated from below is that the system is...
isotropic in the horizontal plane. Thus the convection pattern may form with no preference being given to a particular horizontal axis unless the experimental apparatus introduces an asymmetry. In both cases, the convection is driven by the buoyancy force. However, the mechanism in the NLC case is more involved.\cite{5} The usual destabilization due to the thermally-induced density gradient is opposed by the stiffness of the director field which is coupled to and distorted by any flow. Since relaxation times of the director field are much longer than thermal relaxation times, it is possible for director fluctuations and temperature/velocity fluctuations to be out of phase as they grow in amplitude. The existence of two very different time scales and this phase shift typically lead to an oscillatory instability (also known as overstability), i.e. the bifurcation at which these time-periodic perturbations acquire a positive growth rate is a Hopf bifurcation.\cite{1}$^-$\cite{3} This case is closely analogous to convection in binary-fluid mixtures with a negative separation ratio.\cite{2}$^-$\cite{4} In that case, concentration gradients oppose convection, and concentration diffusion has the slow and heat diffusion the fast time scale. As in the binary mixtures, the Hopf bifurcation in the NLC case is subcritical.\cite{1}$^-$\cite{3} For the NLC the fully developed nonlinear state no longer is time periodic. Instead, the statistically stationary state above the bifurcation is one of spatio-temporal chaos with a typical time scale which is about two orders of magnitude slower than the theoretical inverse Hopf frequency.\cite{4} However, it is possible to actually measure the Hopf frequency by looking at the growth or decay of small perturbations which are either deliberately introduced\cite{1} or which occur spontaneously when the system is close to the conduction state and near the bifurcation point.

A linear stability analysis of this system was carried out by several investigators\cite{1}$^-$\cite{3},\cite{5}$^-$\cite{7}. A very detailed analysis was presented by Feng, Decker, Pesch, and Kramer (FDPK)\cite{7}. These authors also provided a weakly-nonlinear analysis, which allowed the distinction between sub- and supercritical bifurcations. Quantitative bifurcation diagrams were predicted for the nematic liquid crystal MBBA. In the present work we repeated and slightly extended the calculations for the material 5CB (see below) used in our experiments. Since the material parameters of MBBA and 5CB are similar, we found qualitatively the same bifurcation sequences as a function of the field. Here we outline briefly the theoretical results and their relationship to our experimental results.

As the magnetic field is increased, a subcritical Hopf bifurcation-line is expected to terminate at $H_{ct}$ in a codimension-two point (CTP) beyond which the perturbations which first acquire a positive growth rate are at zero frequency. Close to but beyond the CTP this stationary bifurcation is predicted to also be subcritical. At an even higher field $H_{t}$ a tricritical point (TCP) is predicted to exist beyond which the primary bifurcation is expected to become supercritical. To our knowledge there are no predictions about the patterns which should form beyond either the subcritical Hopf bifurcation below the CTP or the subcritical stationary bifurcation between the CTP and the TCP. Although there are no explicit predictions of the patterns for $H > H_{t}$, in analogy to isotropic fluids one might expect convection rolls above the supercritical bifurcation, unless non-Boussinesq effects\cite{15}$^-$\cite{17} yield a transcritical bifurcation to hexagons.

The phenomena described above were previously explored only partially by experiment. Except for recent measurements at relatively small fields\cite{4}, the experiments have been qualitative or semi-quantitative\cite{18}$^-$\cite{20}. In the present paper we report the results of an extensive systematic experimental investigation of this system which covered a wide range of magnetic fields $H$. In agreement with previous work\cite{19}$^-$\cite{20} we find a subcritical Hopf bifurcation at relatively small $H$ which terminates in a codimension-two point (CTP). The CTP is located at a slightly higher field than the theoretical prediction. We measured the Hopf frequency $\omega(H)$ from visualizations of the spontaneous small-amplitude early transients just above $\Delta T_{c}$. Except for the influence of the small shift of the CTP, we found $\omega(H)$ to be in quantitative agreement with the theory. From these transients, we also determined the critical wavevector $k_{c}(H)$, and found it to be typically a few percent smaller than the theoretical value. The reason for this small difference between theory and experiment is not known. As reported previously\cite{13} we found the convecting nonlinear state for $\Delta T$ above $\Delta T_{c}$ to be one of spatio-temporal chaos (STC). Except at very small $H$, its characteristic wavenumber was smaller than $k_{c}$ and insensitive to $H$ and $\Delta T$. A long-time average of the structure factor of the system was investigated for two cells with aspect ratios $\Gamma = 6$ and 5.01. We refer to them as cells 5 and 6 respectively (for details see Sect. III below).

Beyond the CTP we found a subcritical stationary bifurcation, as had been predicted\cite{10}. The finite-amplitude state which evolved was also a state of STC, but beyond a certain field value greater than $H_{ct}$ it had distinctly different properties from the chaotic state at lower $H$. This difference was clearly evident from a discontinuity (as a function of $H$) of the characteristic wavenumber of the nonlinear state, which was larger at the larger fields. There is no theoretical guidance for the interpretation of these experimentally observed phenomena.

The range $H \geq H_{t}$ was investigated for two cells with aspect ratios $\Gamma = 6.15$ and 5.01. We refer to them as cells 5 and 6 respectively (for details see Sect. III below).

We found a primary bifurcation to a state with a hexag-
onal flow pattern. Within our resolution this bifurcation was non-hysteretic and the Nusselt number $N$ grew continuously from zero. The appearance of hexagons rather than rolls is attributable to non-Boussinesq effects which occur when the up-down symmetry is lifted by variations of the fluid properties over the cell height. Theoretically the bifurcation to hexagons is transcritical, and there should also be hysteresis associated with it. However, the hysteresis is often so small that it is unobservable even though hexagons are found over a substantial range. At sufficiently large $\Delta T/\Delta T_c$ the hexagons become unstable with respect to rolls. Since $\Delta T_c$ decreases with the cell thickness $d$ ($\sim d^{-3}$), the range of stability of hexagons should depend on the thickness of the fluid sample. However, for the thinner fluid layer of cell 5, the existence range of hexagons was limited by a different secondary instability and grew from zero very near the TCP to $\epsilon = \Delta T/\Delta T_c - 1 \approx 0.1$ at the highest fields available to us. At this stability limit a hysteretic transition yielded the chaotic state, and stationary rolls were never found. With decreasing $\Delta T$, the chaotic state persisted down to $\Delta T_c$ somewhat smaller than $\Delta T_c$. For the thicker fluid layer of cell 6, typically $\Delta T_c$ was about 2 °C, and hexagons were found only up to $\epsilon \approx 0.015$ even at high fields. For $\epsilon > 0.015$, a pattern of rolls not exhibiting STC was observed, as expected for a weakly non-Boussinesq system. At even higher $\epsilon$ and consistent with the measurements in cell 5, a hysteretic secondary bifurcation again yielded the chaotic state.

II. PARAMETER DEFINITIONS AND VALUES

The quantitative aspects of the instabilities are determined by four dimensionless parameters which are formed from combinations of the fluid properties. They are the Prandtl number

$$\sigma = \frac{(\alpha_4/2)}{\rho k} ,$$

the ratio between the director-relaxation time and the heat-diffusion time

$$F = \frac{(\alpha_4/2)k}{k_{33}} ,$$

the Rayleigh number

$$R = \frac{\alpha g \rho d^4 \Delta T}{(\alpha_4/2)\kappa ||} ,$$

and the dimensionless magnetic field

$$h = H/H_F$$

with the Fréedericksz field

$$H_F = \frac{\pi}{d} \frac{\sqrt{k_{33}}}{\rho \chi_a} .$$

In these equations $\alpha_4$ is one of the viscosity coefficients, $\kappa$ is the thermal diffusivity parallel to $\hat{n}$, $k_{33}$ is one of the elastic constants of the director field, $\chi_a$ is the anisotropy of the diamagnetic susceptibility, $\alpha$ is the isobaric thermal expansion coefficient, and $g$ is the gravitational acceleration. The time scale of transients and pattern dynamics is measured in terms of the thermal diffusion time

$$t_v = d^2/\kappa || .$$

Both $h$ and $R$ are easily varied in an experiment, and may be regarded as two independent control parameters. The availability of $h$ in addition to $R$ makes it possible to explore an entire line of instabilities. The parameters $F$, $\sigma$, and $t_v$ are essentially fixed once a particular NLC and temperature range have been chosen, and even between different NLCs there is not a great range at our disposal. For 5CB at 25.6°C (the material and mean temperature used in this work), we have $\sigma = 263$ and $F = 461$. The value of $t_v$ is typically several minutes, but depends on the thickness of the fluid layer. It is given in the next Section for each of our cells. The critical value $R_c(h)$ of $R$ and the fluid parameters determine the critical temperature difference $\Delta T_c$ for a sample of a given thickness $d$. The realistic experimental requirement that $\Delta T_c$ is a few °C dictates that the sample thickness should be a few mm. Typical values of $H_F$ are 10 to 20 Gauss. Thus modest fields of a kGauss or so are adequate to explore the entire range of interest.

In order to evaluate $R_c$ from $\Delta T_c$, $h = H/H_F$, and the theoretical values for $R_c(h), k_c(h)$, and $\omega_c(h)$, we used the material properties given in Ref. [22]. We followed closely the calculational methods of FDPK. In order to insure a sufficient resolution of any boundary layers we used Chebycheff modes in the Galerkin method (no more than 20 were required).

III. EXPERIMENTAL APPARATUS AND SAMPLE PREPARATION

The apparatus used in this work was described previously. We made measurements using three circular cells of different thicknesses, identified as cells 4, 5, and 6. The thickness and radius were $d = 3.94 \text{ mm}$, $r = 41.9 \text{ mm}$ for cell 4, $d = 6.60 \text{ mm}$, $r = 40.6 \text{ mm}$ for cell 5, and $d = 8.88 \text{ mm}$, $r = 44.5 \text{ mm}$ for cell 6. The corresponding radial aspect ratios $\Gamma = r/d$ were 10.6, 6.15, and 5.01. The fluid was 4-n-pentyl-4’-cyanobiphenyl (5CB). All experiments were performed at a mean temperature of 25.6°C. The vertical thermal diffusion time was $t_v = 139$, 383, and 694 s, and $H_F$ was 20.1, 12.6, and 9.34 Gauss for cells 4, 5, and 6 respectively. Despite the longer time scales involved for experiments in the thicker cells, cells 5 and 6 had an advantage over cell 4 due to the smaller field strengths and temperature.
differences required to perform the measurements. To ensure homeotropic alignment near the surfaces of the top and bottom plates of both cells, a surface treatment with lecithin was applied.

Defect-free homeotropic samples were prepared by applying a magnetic field while cooling the bath, and thus the sapphire top plate of the sample, from above the isotropic-nematic transition temperature $T_{NI}$ to $T < T_{NI}$. During this process, the bottom plate naturally lagged behind, and thus an adverse density gradient existed. In the nematic/isotropic two-phase region even the relatively small thermal gradients associated with small cooling rates induced convection. When the cooling was too rapid and the field too small this led to a nematic sample with defects which remained frozen. By using cooling rates of 1°C/hour in the presence of a field of $h \geq 17$ over the temperature interval 36 to 34°C ($T_{NI} = 35.1^\circ C$) and annealing at 34°C for an hour or two the defects healed and a defect-free homeotropic sample could be prepared. Further cooling could then be at least ten times as rapid without introducing new defects because the threshold for convection in the nematic phase is large. Before each experimental run, the procedure was repeated.

The critical temperature differences for the onset of convection were determined from heat-transport measurements. These are usually expressed in terms of the Nusselt number

$$\mathcal{N} \equiv \lambda_{eff}/\lambda_{\parallel}$$

where $\lambda_{\parallel}$ is the conductivity of the homeotropically aligned sample, and

$$\lambda_{eff} \equiv -Qd/\Delta T$$

is the effective conductivity and contains contributions from diffusive heat conduction and from hydrodynamic-flow advection. Measurements of $\mathcal{N}$ were made by determining the heat current $Q$ required to hold $\Delta T$ constant. At each $\Delta T$, the heat current and temperature of the bath and bottom plate were measured at one-minute intervals for three to five hours, when typically all transients had died out.

In addition to heat-flow measurements, we also visualized the convective flow patterns. The homeotropic samples were translucent even for $d$ as large as several mm. It was just about possible to see features of the bottom plate in typical ambient lighting. Any director distortion by convection rolls or domain walls generated opaque regions with enhanced diffuse scattering which were easily visible. It should be kept in mind that the optical signal in the images has a complicated relationship to the hydrodynamic flow fields, and that quantitative information about velocity- or temperature-field amplitudes could not be obtained. Such quantities as the wavevector of the patterns or frequencies of traveling convection-rolls could of course be determined quantitatively.

The samples were illuminated from above by a circular fluorescent light. Digital images were taken from above by a video camera which was interfaced to a computer. Typically 50 to 200 images were averaged to improve the signal-to-noise ratio. Averaged images were divided by an appropriate reference image to reduce the influence of lateral variations in illumination and of other optical imperfections. Some images were processed further by filtering in Fourier space.

FIG. 1. Examples of the Nusselt number as a function of $\Delta T$ for cell 5. The upper figure is for $h = 15$ and the lower one is for $h = 50$. Open circles were taken with increasing and solid circles with decreasing $\Delta T$. The transitions between conduction and convection are indicated by the arrows.

IV. RESULTS

A. Nusselt Numbers and Critical Rayleigh Numbers

Figure 1 shows $\mathcal{N}$ for cell 5 as a function of $\Delta T$ for two field strengths $h = 15$ and $h = 50$. The open circles were obtained with increasing, and the solid circles with decreasing $\Delta T$. For the lower fields ($h < 20$) $\mathcal{N}$ decreased below one when convection started. This can be understood because the convecting sample has a distorted director with a component perpendicular to $\vec{Q}$. The contribution from this component to the conductivity corresponds to $\lambda_{\perp}$, which is less than the conductivity $\lambda_{\parallel}$ of the homeotropic case. It turns out that for small fields the direct hydrodynamic contribution to the heat flux is smaller than the decrease in the heat flux due to the director distortion by the flow. For the higher fields ($h > 35$), $\mathcal{N}$ remained above one in the convect-
ing state. Thus with the higher fields the hydrodynamic contribution to the heat flux is greater than the decrease in the heat flux due to any distortion of the director. Both examples in Fig. 1 demonstrate the predicted and previously observed [9,14,20] hysteretic nature of the bifurcation, i.e. as $\Delta T$ was decreased, the conduction state was reached at a value of $\Delta T < \Delta T_c$.

From data like those in Fig. 1, critical temperature differences $\Delta T_c$ were determined with an uncertainty of less than one percent. The corresponding Rayleigh numbers are shown in Fig. 2 as a function of $h^2$. The open circles were obtained in cell 4, the filled ones in cell 5. The good agreement between the two data sets confirms the expected scaling of the field with $H_F$. It also shows that using the fluid properties at the mean temperature does not lead to systematic errors in $R_c$ even for cell 4 where $\Delta T_c$ is over 10°C. One sees that $R_c$ is quadratic in $h$ at small $h$, as is expected because the system should be invariant under a change of the field direction. The solid line follows the theoretical prediction and agreement between theory and experiment is excellent.

Results for $R_c$ over our full experimental field range are shown as a function of $h$ in Fig. 3. Here we include data taken with cell 6 as open squares. The data for $R_c$ reveal a sharp maximum at $h = 44.3$. We interpret this field value as the codimension-two point $h_{ct}$ and indicate it in Fig. 3 by the dashed vertical line. The solid line in the figure is the theoretical prediction for $R_c$, evaluated for the properties of our sample. For the entire range $h < h_{ct}$, the theoretical result is in excellent agreement with the data. However, the theory gives $h_{ct} = 41.8$ which is slightly lower than the experimental value. Above $h_{ct}$ the measurements of $R_c$ are systematically larger than the calculation, although the largest discrepancy is only about 4%.

The triangles in Fig. 3 show the lower limit of existence (the “saddle-node” Rayleigh number $R_s$) of the finite-amplitude convecting state as determined from data like those in Fig. 1. They suggest that the tricritical bifurcation is located near $h = 59$, which is larger than the theoretically calculated value $h_t \simeq 51$. However, we will return later to the best estimate of $h_t$.

Measurements similar to those shown in Fig. 3 were made by Salán and Fernández-Vela [20] (SF), using the nematic liquid crystal MBBA. Their results are shown in Fig. 4, together with the theoretical curve for that case [26]. The data and the curve illustrate that there are significant quantitative differences between the bifurcation lines of different nematics. In Fig. 4 the experimental points lie on average about 25% above the theoretical curve, and the lower hysteresis limit is further below the bifurcation line than we found for 5CB.

FIG. 2. Critical Rayleigh numbers for the onset of convection as a function of $h^2$. Open and filled symbols were obtained in cells 4 and 5, respectively. The line is the theoretical prediction.

FIG. 3. Critical Rayleigh numbers $R_c$ and saddle-node Rayleigh numbers $R_s$ over the experimentally accessible field range. The open circles, filled circles, and open squares are $R_c$ for cells 4, 5, and 6 respectively. The open and filled triangles are $R_s$ for cells 4 and 5 respectively. The dashed line indicates the location of the codimension-two point as found experimentally. The solid line is the theoretical prediction for $R_c$. The plusses at $h = 35$ were obtained with short equilibration times and cell 5 (see text).

FIG. 4. Results for $R_c$ and $R_s$ from Ref. [20] obtained with MBBA. The theoretical curve for $R_c$ was computed from typical fluid properties of MBBA [26].

The equilibration times after each temperature step used by SF were 30 minutes, which is a factor of six to ten shorter than those of our experiments. In addition the
temperature steps of SF were a factor of two larger than ours, yielding a difference in the average rate of change of the temperature of a factor of 12 or more. Looking for an explanation of the difference between the experimental and theoretical $R_c$ revealed in Fig. 4, we conducted one run with equilibration times similar to those of SF, but using our 5CB sample. It gave the plusses in Fig. 4. As can be seen, these results do not differ significantly from the data taken with our usual longer equilibration times. Thus we have no explanation for the difference between the SF data for $R_c$ and the theoretical curve. However, the agreement between our runs with the different equilibration times implies that our usual experimental procedure yielded quasi-static results.

FIG. 5. A sequence of images of the travelling/standing wave transients for $h = 32$ and $\epsilon = 0.015$ in cell 5. The number in each image corresponds to the time, in units of $t_v$, that elapsed since flow first became visible. A time series of the pixel intensity was taken at the location marked in the top left image.

B. Hopf Frequency and Critical Wavevector

Once the critical Rayleigh numbers were measured, a detailed analysis of the patterns could be undertaken. At first we will characterize the Hopf bifurcation for $h$ below $h_{ct}$. Since in that field range the bifurcation is subcritical, we had to use the small-amplitude transients to determine $\omega_c$ and $k_c$. Figure 6 shows images from cell 5 which are characteristic of these patterns. They were taken at the times (in units of $t_v = 383$ s) indicated in each figure after the pattern initially became visible. This typically occurred around one hour after $\epsilon$ was raised from below zero to around 0.015. Inspection of successive images revealed that the transients could be either traveling or standing waves, sometimes with both occurring at different locations in the same cell. In the top left image of Fig. 6, a location is indicated at which a time series of the pixel intensity was acquired. This time series is shown in Fig. 6 along with its corresponding power spectrum. The length of the time series was limited by the rapid growth of the pattern to its finite-amplitude steady state. Because of this, only a small number of periods could be obtained before the finite-amplitude state was reached. Thus to avoid errors associated with incommensurate sampling, the data was windowed before its Fourier transform was evaluated. This process was repeated at several pixel locations in the cell. The signal from the second harmonic was often found to be stronger than that from the fundamental. Thus it was used to calculate the frequency. The frequencies at different locations generally were within a percent of each other, and were averaged to determine the critical Hopf frequency $\omega_c$.

FIG. 6. The upper figure is the time series of the pixel intensity at the location shown in the top left image of Fig. 5. The lower figure is the power spectrum of that time series. The mean frequency calculated from the solid circles was used to determine the Hopf frequency.

The dependence upon $h$ of the measured $\omega_c$ is compared with theory in Fig. 7. The arrow indicates the location of the theoretical codimension-two point while the dashed line represents the experimental determination of $h_{ct}$. As can be seen, away from the codimension-two point the agreement with the measurements is excellent. In accordance with theory, the experimental $\omega_c$ changes
discontinuously to zero at \( h_{ct} \), above which the bifurcation is stationary.

By evaluating the Fourier transforms of images such as those in Fig. 1, the critical wavenumber \( k_c \) of the patterns could be measured. The transforms were based on the central parts of the images by using the filter function \( W(r) = \left\{ 1 + \cos[(\pi)(r/r_o)] \right\}/2 \) for \( r < r_o \) and \( W(r) = 0 \) for \( r > r_o \). Here \( r_o \) was set equal to 85% of the sample radius. Time averaging the square of the modulus of the transforms over the length of the time series yielded the structure factor \( < S(k) > \). Figure 2 shows the azimuthal average \( < S(k) > \) of \( < S(k) > \) for the run at \( h = 32 \). We used a weighted average of the three points nearest the peak of the second harmonic of \( < S(k) > \) to calculate \( k_c \).

![FIG. 7. The Hopf frequency \( \omega_c \) as a function of \( h \). Open and filled circles are for cells 4 and 5, respectively. Open squares are for cell 6. The solid line is the theoretical prediction for \( \omega_c \). The dashed line (arrow) indicates the location of the codimension-two point as found experimentally (theoretically).](image1)

![FIG. 8. The azimuthal average of the time-averaged structure factor \( < S(k) > \) of the travelling/standing wave transients at \( h = 32 \) for cell 5. The mean wavenumber calculated from the solid circles was used as the critical wavenumber.](image2)

Figure 3 displays the results for \( k_c \) for all \( h \) together with the theoretical analysis. For \( h < h_{ct} \) the measured critical wavenumber of the transients is systematically smaller than the theoretical one. When the codimension-two point is approached, the experimental wavenumbers make a smooth rather than discontinuous transition to those associated with the stationary bifurcation, whereas the theory predicts a 7% discontinuity of \( k_c \) at \( h_{ct} \). The reason for these discrepancies is as yet unknown. Above \( h_{ct} \), the agreement between the experimental and theoretical wavenumbers is excellent.

As shown explicitly for \( R_c \) in Fig. 4, \( R_c, \omega_c, \) and \( k_c \) are proportional to \( h^5 \) for small \( h \).

![FIG. 9. The characteristic wavenumbers of the observed patterns as a function of \( h \). Circles: The wavenumber of the small-amplitude transients. Triangles: The wavenumber \( k_n \) of the fully developed spatially and temporally chaotic flow, as measured close to the onset of such flows. Open and filled symbols were obtained in cells 4 and 5, respectively.](image3)

C. Nonlinear State below the tricritical field \( h_{ct} \)

Because of the subcritical nature of the bifurcation for \( h < h_{ct} \) a finite-amplitude state develops directly at onset. The time dependence and spatial structure of this state are very different from that of the small-amplitude transient state. The first two rows of Fig. 5 show typical images of the patterns from cell 5 which are characteristic of the fully developed flow. They are from a single experimental run with constant external conditions. They were taken at the times indicated in each image, in units of \( t_v = 383 \) s, which had elapsed since \( \epsilon \) had been raised from below zero to 0.014. The convection rolls of the fully developed flow show an irregular time dependence, with typical time scales around a hundred times longer than the inverse Hopf frequencies of the transients. One can see that the “chaotic” behavior is associated with the formation of defects and the continuous reorientation of the convection rolls. This continuous reorientation of the rolls is evident in the rightmost image in the bottom row of Fig. 5 (labeled “Avg”). It shows the time average of the structure factor. The average involved 75 images taken over a total time period of \( 724 t_v \) (over three days). It is seen to contain contributions at all angles, consistent with the idea of a statistically stationary process of non-periodic pattern evolution and with the expected rotational symmetry of the system. Similar results for cell
4 have been shown previously. 

FIG. 10. Top two rows: a sequence of images taken with constant external conditions \((h = 50, \epsilon = 0.014)\) for cell 5. The time elapsed since \(\epsilon\) was raised from below zero (in units of \(t_v = 383s\)) is given in the top left corner of each image. Bottom row: The structure factor of two of the images shown above, and the average of the structure factor of 75 images spanning a time interval of 724\(t_v\). The structure factor was obtained using a Hanning window, and thus is dominated by the patterns near the cell center.

FIG. 11. A temporal succession of images during the transient leading from conduction to convection when \(\Delta T\) was raised slightly above \(\Delta T_c\) for cell 5. The field was \(h = 50\). The numbers are the elapsed time, in units of \(t_v\), since the threshold was exceeded.

When \(h\) was increased above \(h_{ct}\), the nature of the pattern at first did not change noticeably. For instance, as evident in Fig. 11, the characteristic wavenumber of the pattern (as denoted by the triangles) remained close to 3.4 for \(h \leq 55\). Over this field range the patterns of the fully developed flow look similar to those illustrated in Fig. 10, i.e. they exhibit spatio-temporal chaos.

FIG. 12. Nusselt-number measurements for cell 5 illustrating the variation of the size of the hysteresis loop between conduction and convection with \(h\). The number in the upper left corner of each plot is the field \(h\). Open circles were taken with increasing and solid circles with decreasing \(\Delta T\). The arrows show the values of \(\epsilon_s \equiv R_s/R_c - 1\).

It is instructive to examine the transients which lead from the small-amplitude to the finite-amplitude statistically-stationary state. This is done in Fig. 11. Here the number in each image gives the time, in units of \(t_v\), which had elapsed since \(\Delta T\) was raised slightly (1.4\%) above \(\Delta T_c\). At \(t = 4.7\) small-amplitude transients like those in Fig. 8 are evident in part of the cell. By \(t = 9.4\) these had filled the cell and grown to a saturated amplitude. At this stage they formed nearly-straight parallel rolls with a wavenumber which was smaller than \(k_c\). However, these straight rolls turned out to be unstable to a zig-zag instability. In the end, this instability led to the spatially and temporally disordered pattern as shown in Fig. 10. Thus, we see that a
secondary instability led to a chaotic state rather than to a new time-independent pattern. This phenomenon most likely is similar to the one encountered in very early experiments on spatio-temporal chaos using liquid helium [27,28], where ordinary RB convection became chaotically time dependent, most likely because the secondary skewed-varicose instability [29] was crossed.

When the Nusselt numbers in Fig. 12 are plotted against the Rayleigh number  \( R_{\text{avg}} \) for several field values as a function of \( \epsilon \). They illustrate the evolution with \( \epsilon \) of the hysteretic nature of the bifurcation. As can be seen also in Fig. 8, the hysteresis \( |\epsilon_s| \) increased with \( \epsilon \) for \( \epsilon < \epsilon_{c1} \), from about 10% at the low fields to nearly 25% close to the codimension-two point. Above this point the hysteresis decreased and suggested the existence of a tricritical point near \( \epsilon_{c1} \approx 59 \) (see below for more detail about the tricritical region).

When the Nusselt numbers in Fig. 13 are plotted against the Rayleigh number \( R \), they fall on or approach a single curve independent of \( \epsilon \). This suggests that the convection in the chaotic state is sufficiently vigorous to achieve nearly complete randomization of the director orientations, regardless of \( \epsilon \). In that case one would expect that the system should behave approximately like an isotropic fluid, with an averaged conductivity \( \lambda_{\text{avg}} = (2\lambda_\perp + \lambda_\parallel)/3 \). Thus we plot in Fig. 13 a modified Nusselt number \( \tilde{N} \) given by the ratio of the effective conductivity of the convecting state to \( \lambda_{\text{avg}} \) as a function of \( R_{\text{avg}} \), where \( R_{\text{avg}} \) is computed using \( \kappa_{\text{avg}} = \lambda_{\text{avg}}/\rho C_P \) in Eq. 3 rather than \( \kappa_{\parallel} \). At all but the highest fields (where the primary bifurcation is supercritical) the data reach the common curve. At small \( R_{\text{avg}} \), this curve extrapolates to \( \tilde{N} = 1 \) near \( R_{\text{avg}} = 1708 \) (the cross in the figure), which is the critical Rayleigh number of an isotropic fluid. An analogous behavior has been observed in binary-mixture convection with negative separation ratios \( \Psi \), [30] where the bifurcation is also subcritical. In that case the convective flow achieves thorough mixing of the concentration field and \( \tilde{N} \) approaches a curve which is independent of \( \Psi \). In both cases the mixing achieved by the flow can persists because of the existence of a slow time scale, namely that of director or concentration relaxation.

Further support for the idea that the chaotic flow in some respects can be approximated by isotropic-fluid convection is found in Fig. 14, where for \( h \approx 55 \) the wavevectors (triangles) are independent of \( \epsilon \) and much closer to the critical value \( k_{\epsilon, \text{iso}}^c = 3.117 \) than to the critical values \( k_c(h) \) of the anisotropic system (circles in Fig. 14). Exact agreement with \( k_{\epsilon, \text{iso}}^c \) would of course not be expected even for a genuine isotropic fluid because of the finite flow amplitude and various wavenumber-selection processes.

Lastly we note that an extrapolation of the data in Fig. 13 to \( \tilde{N} = 1 \) and \( R_{\text{avg}} = 1708 \) yields an initial slope \( S_1 \) of \( \tilde{N} - 1 = S_1(R_{\text{avg}}/1708 - 1) \) of about 0.6. For a laterally infinite system of straight rolls in an isotropic fluid with a large Prandtl number one expects \( S_1 \approx 1.43 \). [31] However, experiments in finite systems with modest aspect ratios [32] have always yielded smaller values, usually in the range of 0.6 to 1. Particularly when many defects are present, as in our case, one would expect the heat transport to be suppressed relative to that of a perfect straight-roll structure.

D. Tricritical Region and beyond

This section is devoted to the phenomena which occur near the tricritical field \( h = h_t \). At first the Nusselt numbers and the patterns are described and the corresponding bifurcation diagram is given. Further subsections deal with the precise determination of \( h_t \) and with hexagons observed near threshold for \( h > h_t \).

1. Nusselt Numbers and Patterns

From the measurements of the Nusselt number (see Fig. 13) there is clear evidence of a tricritical field \( h_t \), above which the primary bifurcation is supercritical. For instance for \( h \approx 60 \), measurements of \( \tilde{N} \) revealed no hysteresis at the primary bifurcation and within our resolu-
tion $\mathcal{N}$ grew continuously from one beyond $\Delta T_c$. This is exemplified for cell 5 and $h = 63$ in Fig. 14. The open (solid) circles correspond to the stable states reached by increasing (decreasing) $\Delta T$. This behavior of $\mathcal{N}$ stands in sharp contrast to that shown in Fig. 12 for lower fields.

![Image of hexagonal flow in cell 5 for $h = 65$ and $\epsilon = 0.01$. The pattern was essentially the same over the entire existence range of hexagons.](image1)

At first we will focus on the wedge labeled Rolls/Hex where time-independent convection is stable. For both cell 5 and 6, a seemingly supercritical primary bifurcation led to a hexagonal pattern. For cell 5 this pattern is shown in Fig. 14. The range of $\epsilon$ over which the hexagons were stable differed in the two cells. In cell 5, hexagons remained stable up to $R_n(h)$ (solid squares in Fig. 15) for the entire range of $h$. At $R_n(h)$ a transition to a spatially and temporally chaotic roll pattern with a lower characteristic wavenumber occurred. For $h < 75$ this transition was distinguished by a jump in $\mathcal{N}$ as well as the onset of time dependence of $\mathcal{N}$, as illustrated in Fig. 14. The upper figure gives the steady-state $\mathcal{N}$ and shows the jump at $\epsilon_n \equiv R_n/R_c - 1$. The lower figure is the time series of $\mathcal{N}$ obtained in the same run. Here $\epsilon$ was held constant for a five-hour period at each of the eleven successively increasing values. The data show that $\mathcal{N}$ is steady below and time dependent above $\epsilon_n(h)$. For $h \geq 75$ the discontinuity in $\mathcal{N}$ was no longer pronounced, but a transition to time dependence still occurred at $\epsilon_n(h)$. Thus, depending on the field, either of these two indicators was used to determine the location of $\epsilon_n(h)$. In the thicker cell 6, a transition from hexagons to rolls occurred near $h \approx 0.015$, independent of field strength. This transition was not associated with a measurable change in the wavenumber, a jump in $\mathcal{N}$, or a time dependence of $\mathcal{N}$. A further increase of $\epsilon$ again led to a transition at $\epsilon_n(h)$ from steady rolls to the chaotic state, consistent with the cell 6 experiments. The results for $\epsilon_n(h)$ obtained in cells 5 and 6 are shown in Fig. 15 as circles and triangles respectively.

On the basis of the usual Landau equation for a tricritical bifurcation, one would expect the hysteresis to grow gradually as $h$ is reduced below $h_t$. However, within our resolution this was not the case and a hysteretic primary bifurcation to a chaotic state occurred immediately below $h_t$. Indeed, the secondary bifurcation line $\epsilon_n(h)$ for $h > h_t$ met the primary bifurcation line at $h_t$ within experimental resolution, as can be seen already in Fig. 14.

Besides the Nusselt number the analysis of the patterns gives important additional insight in particular with respect to secondary transitions. In Fig. 15 all the available information has been condensed in a bifurcation diagram for the vicinity of the tricritical point.

![Bifurcation diagram in the region of the $R - h$ plane close to the tricritical point. Solid circles: primary bifurcation. Solid triangles: $R_t$. Solid squares: hysteretic secondary bifurcation to chaotic convection. Shaded areas labeled $k_n = 3.4$, 3.6, and 3.9 correspond to chaotic regimes with different mean wavenumbers. The wedge-shaped area labeled Rolls/Hex shows the parameter range over which time-independent convection is stable. For cell 5, the pattern is hexagonal in this entire region. For cell 6, the pattern is hexagonal in this region for $\epsilon \leq 0.015$. For larger $\epsilon$ but still in this region it consists of time-independent rolls.](image2)
hours, i.e. the time between steps in \( \Delta T \). Time is measured in units of 5 hours, i.e. the time between steps in \( \epsilon \).

FIG. 18. Values of \( \epsilon_n(h) \) where a transition to a spatially and temporally chaotic state occurred. Circles and triangles were obtained in cells 5 and 6, respectively. The lines represent a fit of a quadratic polynomial in \( (h/h_n - 1) \) to the data. The fits extrapolate to zero at \( h_n = 58.3 \pm 0.7 \) for cell 5 and \( h_n = 58 \pm 3 \) for cell 6.

It is shown more explicitly in figure [18], where the range \( \epsilon_n \) of time-independent patterns vanishes near \( h = 58 \). Fitting a quadratic polynomial in \( (h/h_n - 1) \) to \( \epsilon_n(h) \) yields \( h_n = 58.3 \pm 0.7 \) (cell 5) and \( h_n = 58 \pm 3 \) (cell 6) for the field where \( \epsilon_n \) vanishes. Within error, these values agree with the tricritical field obtained from the slope of the Nusselt number (see the next Section). The bifurcation for \( h > h_t \) is supercritical but the amplitude at constant \( \epsilon > 0 \) diverges as \( h \) approaches \( h_t \) from above. Since secondary bifurcations occur at finite values of the amplitude, we expect \( \epsilon_n(h) \) to vanish at \( h_t \). Therefore the \( \epsilon_n \) measurements provide a relatively precise lower limit \( h_t = 57.6 \) for the tricritical field.

FIG. 19. Nusselt-number measurements for \( h = 64 \) in cell 5. Open circles were taken with increasing and solid circles with decreasing \( \Delta T \). The dotted line indicates \( \epsilon \).

The secondary bifurcation at \( \epsilon_n \) is strongly hysteretic. As illustrated in figure [19], when \( \epsilon \) was decreased from \( \epsilon > \epsilon_n \), a transition back to hexagons did not occur. Instead, the chaotic state persisted to values of \( \epsilon \) slightly below zero, at which point the conduction state was reached (see also the solid triangles in Fig. [15]). In a separate section we will come back to the hexagons.

2. Determination of the Tricritical Point

As \( h \) approaches the tricritical point from above, the initial slope \( S_{1,r} \) of \( \mathcal{N} \) for rolls is expected to diverge as \( 1/(h-h_t) \). For cell 6, we estimated \( S_{1,r}(h) \) from data for \( \epsilon \geq 0.015 \) where rolls were observed. At a given \( h \), \( S_{1,r} \) was determined by fitting the polynomial

\[
\mathcal{N} = 1 + S_{1,r} \epsilon + S_{2,r} \epsilon^2
\]

with \( \epsilon = \Delta T/\Delta T_c - 1 \) to the data. The parameters \( \Delta T_c \), \( S_{1,r} \), and \( S_{2,r} \) were adjusted in the fit. Figure [23] shows the dependence of \( 1/S_{1,r} \) upon \( h \) as solid circles. The fitting procedure did not yield highly accurate values because the Nusselt data for \( \epsilon < 0.015 \) had to be excluded; thus the error bars for \( S_{1,r} \) are relatively large. The line is a fit of a quadratic polynomial in \( (h/h_t - 1) \) to the results for \( 1/S_{1,r}(h) \). This fit indicates the tricritical point to be at \( h_t = 57.2 \pm 2.6 \).

In order to compare the theory with the measurements above \( h_t \), we calculated \( S_{1,r} \) using the properties of 5CB. The results for \( 1/S_{1,r} \) are shown as a dashed line in Fig. [24]. There is quite reasonable agreement with the
experimental data for $h \gtrsim 60$, particularly when it is considered that the initial slope of $N$ in finite systems is usually smaller than the theoretical value for the infinite system. However, the theory yields a tricritical field $h_{t1}^{tr}$ = 51 which differs significantly from the experimental estimates. We note that this difference is in the same direction as and somewhat larger than the corresponding one for the codimension-two point. We have no explanation for this difference.

![Graph](image)

**FIG. 20.** The dependence on $h$ of the reciprocal of the initial slope $S_1$ of the Nusselt number as obtained from a fit of the data to Eq. $3$. The open circles are $1/S_{1,h}$ for cell 5, and the filled ones are $1/S_{1,r}$ for cell 6. For cell 5, data over the range $0 < \epsilon < \epsilon_n$ were used, and the pattern was hexagonal. For cell 6, data over the range $0.015 < \epsilon < \epsilon_n$ were used, and the pattern was one of rolls. The solid line represents a fit of a quadratic polynomial in $(h/h_t - 1)$ to the cell 6 $S_{1,r}$ data. The field $h_t = 57.2 \pm 2.6$, where $1/S_{1,r}$ for cell 6 extrapolates to zero, is interpreted to be the tricritical point. The theoretical results for $1/S_{1,r}$ are given by the dashed line. They yield a tricritical point at $h_t^{tr} = 51$. The theoretical results for $1/S_{1,h}$ are given by the dash-dotted line. The location of the codimension-two point is given by the solid (experimental) and dashed (theoretical) short vertical lines.

### 3. Hexagons

In this section we will discuss in more detail the hexagonal patterns. Hexagonal patterns at onset may be attributable to departures of the physical system from the Oberbeck-Boussinesq (OB) approximation $[8,9]$, i.e., to a variation of the fluid properties over the imposed temperature range. For isotropic fluids it has been shown that non-OB effects lead to hexagons at a transcritical (hysteretic) primary bifurcation $[10,21]$. Below onset, for $\epsilon_a \leq \epsilon \leq 0$, both hexagons and the conduction state are stable. Above onset, hexagons are stable for $0 \leq \epsilon \leq \epsilon_r$. For $\epsilon_r \leq \epsilon \leq \epsilon_h$ hexagons and rolls are both stable, while for $\epsilon \geq \epsilon_h$ only rolls are stable. When the thickness of the fluid layer is increased, $\Delta T_c$ is reduced and thus departures from the OB approximation become smaller. Thus the range of $\epsilon$ over which hexagons are stable is reduced when the thickness of the fluid layer is increased, as seen in the experiment by comparing cells 5 and 6.

A stability analysis of RBC with non-OB effects in a homeotropically aligned NLC has not yet been carried out and would be very tedious. Thus, in order to obtain at least a qualitative idea of the expected range of stable hexagons, we used the theoretical results for the isotropic fluid with the fluid properties of 5CB. The values of $\epsilon_a$, etc. are determined by a parameter $\mathcal{P}$ which was defined by Busse $[10]$ and is given by $\mathcal{P} = \prod_{i=0}^{4} \mathcal{P}_i$, with $\gamma_0 = -\Delta \rho/\rho$, $\gamma_1 = \Delta \alpha/2\alpha$, $\gamma_2 = \Delta \nu/\nu$, $\gamma_3 = \Delta \lambda/\lambda$, and $\gamma_4 = \Delta C_P/C_P$. Here $\rho$ is the density, $\alpha$ the thermal expansion coefficient, $\nu$ the kinematic viscosity, $\lambda$ the conductivity, and $C_P$ the heat capacity. The quantities $\Delta \rho$, etc. are the differences in the values of the properties at the bottom (hot) and top (cold) end of the cell. For $\lambda$ we used $\lambda_{\text{h}}$, and for $\nu$ we used $\alpha_{\text{h}}/2\rho$. The coefficients $\mathcal{P}_3$ in the equation for $\mathcal{P}$ are given by Busse $[10]$. However, here we use the more recent results $[34]$ for large Prandtl numbers $\mathcal{P}_0 = 2.676$, $\mathcal{P}_1 = -6.631$, $\mathcal{P}_2 = 2.765$, $\mathcal{P}_3 = 9.540$, and $\mathcal{P}_4 = -6.225$ where $\mathcal{P}_3$ differs significantly from the earlier calculation.

At the fields where the hexagons were observed, the temperature difference across cell 5 (cell 6) was close to 5.05°C (2.07°C). At these temperature differences, we obtained $\mathcal{P} = -1.5$, $\epsilon_a = -1.7 \times 10^{-3}$, $\epsilon_r = 1.5 \times 10^{-2}$, and $\epsilon_h = 5.3 \times 10^{-2}$ for cell 5. For cell 6 the values are $\mathcal{P} = -0.6$, $\epsilon_a = -2.8 \times 10^{-3}$, $\epsilon_r = 2.5 \times 10^{-3}$, and $\epsilon_h = 8.7 \times 10^{-3}$. From these estimates, it follows that the hysteresis of size $\epsilon_a$ is too small to be noticeable in either cell with our resolution. The largest value of $\epsilon$ at which hexagons could exist in cell 6 would be $\epsilon = 8.7 \times 10^{-3}$. However, we observe hexagons to exist nearly twice this value. If the same is true for the thinner cell, the hexagon-roll transition attributable to non-OB effects should happen at a value of $\epsilon$ greater than $\epsilon_a(h)$ for the field range over which the experiments were performed. Thus, instead of leading to a time independent state, as observed in cell 6, the hexagon-roll transition is preceded by a transition to a state exhibiting spatio-temporal chaos at $\epsilon_a$.

The hexagonal pattern may be regarded as a superposition of three sets of rolls with amplitudes $A_i$, $i = 1, 2, 3$, corresponding to the three basis vectors at angles of 120° to each other. According to the Landau model $[13]$ the steady-state amplitudes $A_i$ are determined by

$$\epsilon A_1 + \frac{1}{2} b(A_2^2 + A_3^2) - g_{11} A_1^3 - g_{12} A_1 A_2^2 - g_{13} A_1 A_3^2 = 0$$

(10)

and the corresponding cyclic permutation for $i = 2, 3$. Since all amplitudes are expected to be equal in hexagons ($A_1 = A_2 = A_3 = A$), one has

$$\epsilon A + b A^2 - (g + 2\tilde{g}) A^3 = 0$$

(11)

where $g$ is the self-coupling coefficient $g_{11}$ and $\tilde{g}$ is the cross-coupling coefficient $g_{12}$. Because of the term $b A^2$, the bifurcation is transcritical and thus hysteretic.
However, as we discussed above and as is shown for instance by the data in Fig. 14, this effect is not resolved in the experiment because the coefficient $b$ (which is determined by $P$) is too small. Thus we neglect the term $bA^2$, and have to a good approximation

$$N - 1 = 3A^2 = \frac{3\epsilon}{g + 2\tilde{g}} \quad (12)$$

At the tricritical point $g$ vanishes as $g = g_0(h - h_t)$. However, there is no reason why $\tilde{g}$ should vanish also at $h_t$. Thus, one would expect the slope $S_{1,h} = 3/(g + 2\tilde{g})$ of $N$ near $\epsilon = 0$ to remain finite at $h_t$ and equal to $3/2\tilde{g}$. To test this idea, we fitted $N$ for cell 5 over the $\epsilon$ range where hexagons were observed (i.e. up to $\epsilon_n$) to an equation like Eq. 1. Since data quite close to threshold could be used, the results for $S_{1,h}$ are much more precise than those for cell 6. They are given in Fig. 20 as open circles. One can see that $S_{1,h}$ is non-zero at $h_t$. It extrapolates to zero near $h = 53$, which is well below $h_t = 57.2 \pm 2.6$. Unfortunately the relatively large uncertainty of $h_t$ and $S_{1,r}$ prevents the accurate determination of $\tilde{g}$. At $h_t$, we find $\tilde{g} = 3/2S_{1,h} \simeq 0.3$. With increasing $h$, $\tilde{g}$ also increases. For instance, the data in Fig. 20 suggest that $\tilde{g} = 3/2S_{1,h} - 1/2S_{1,r} \simeq 0.8$ for $h = 66$.

The experimental results for $1/S_{1,h}$ cannot agree quantitatively with the theory because we already know that $h^{th}_\alpha$ is lower than the experimental value. Nevertheless we calculated $1/S_{1,h}$, and give it in Fig. 20 as the dash-dotted line. We see that the relationship between $1/S_{1,h}$ and $1/S_{1,r}$ is quite similar in theory and experiment.

E. The High-Field Limit of $R_c$ and $k_c$

It is highly probable that there exists a high-field regime where the convection phenomena become independent of the field since the director is then frozen in the homeotropic configuration. It is instructive to examine whether the experimental data extend to sufficiently high fields to fully reveal this behavior. Building on the results of FDPK [10], one can show that in a one-mode approximation the neutral curve in the limit $h \to \infty$ is given by

$$R_{c,\infty}(k) = \frac{(\lambda_1/\lambda_||)k^2 + \pi^2}{2k^3I_1^2} \times \left[ -I_2k^2 \left( \frac{2\eta_1}{\alpha_4} + \frac{2\eta_2}{\alpha_4} + \frac{2\alpha_3}{\alpha_4} \right) + k^4\frac{2\eta_2}{\alpha_4} + b^2\frac{2\eta_1}{\alpha_4} \right] \quad (13)$$

Here $\eta_1 = (\alpha_4 + \alpha_5 - \alpha_2)/2$ and $\eta_2 = (\alpha_3 + \alpha_4 + \alpha_5)/2$ are Mieowicz viscosity coefficients, [4] and $I_1 = 0.69738, I_2 = 0.69738, I_2 = -12.3026$, and $b = 1.50567$. This leads to $R_{c,\infty}(k_c) = 3090.6$ and $k_{c,\infty} = 4.294$. Using many modes, one obtains numerically $R_{c,\infty}(k_c) = 3056.6$ and $k_{c,\infty} = 4.328$, close to the one-mode result. One can see that only the viscosities enter into $R_{c,\infty}$, and not the elastic constants. This is so because the director is held rigid by the field. $R_{c,\infty}$ is larger than the isotropic-fluid value because of the additional viscous interaction between the flow and the rigid director field.

In the high-field limit, we obtain

$$R_c = R_{c,\infty} + R_1/h^2 + O(h^{-4}) \quad (14)$$

The coefficient $R_1$ has not been calculated in detail, but is proportional to $k_{3,1}^{-1}$. The fact that at order $1/h^2$ elastic constants enter suggests the beginning of some director distortion by the flow.

![FIG. 21](image-url) The critical Rayleigh number as a function of $1/h^2$. The solid line was calculated using the parameters of Ref. 22. The open triangle is $R_{c,\infty}$ from Eq. 13. The solid triangle is the many-mode numerical result for $R_{c,\infty}$. The solid circles are from cell 5, and the open squares are from cell 6. The vertical bars indicate the location of the codimension-two point (right bar) and the tricritical point (left bar). The plusses, crosses, and dashed line show what happens to the data and the theoretical curve if the viscosity $\alpha_4$ is increased by 7.5%.

In Figs. 21 and 22 we show the experimental data and theoretical results as a function of $h^{-2}$. The one-mode high-field limits $R_{c,\infty}$ and $k_{c,\infty}$ are shown in the figures as open triangles. The corresponding numerical many-mode results are given as solid triangles. The experimental data for $R_c$ and $k_c$ are consistent with the expected dependence on $h$, but respectively fall about 4% above and 1% below the calculation. The data for $R_c$ and $k_c$ at the highest experimental field $h \simeq 80$ are already within about 6% and 1% respectively of the infinite-field value. Thus it seems unlikely that qualitatively new phenomena could be discovered by measurements at even higher fields.
FIG. 22. The critical wavenumber as a function of $1/h^2$. The solid line was calculated using the parameters of Ref. [22]. The open triangle is $k_{c,\infty}$ from Eq. [4]. The solid triangle is the many-mode numerical result for $k_{c,\infty}$. The vertical bars indicate the location of the codimension-two point (right bar) and the tricritical point (left bar). The dashed line shows what happens to the theoretical curve if the viscosity $\alpha_4$ is increased by 7.5% (the data for $k_c$ remain unchanged).

We examined whether the small difference between the theory and the experiment could be removed by small adjustments in the values of the fluid properties. We found that an increase by 7.5% of $\alpha_4$ yielded the plusses and crosses for the data in Fig. 21 and the dashed lines in Figs. 21 and 22 (the data for $k_c$ in Fig. 22 are not affected by changing $\alpha_4$). The adjustment of $\alpha_4$ produced an excellent fit for both $k_c$ and $R_c$. However, it spoiled the excellent agreement for $R_c$ along the oscillatory branch below the codimension-two point shown in Fig. 3 and did not significantly reduce the difference between calculation and experiment for $k_c$ at small $h$ which is shown in Fig. 9. Various other attempts to adjust the fluid properties used in the theoretical calculations were unsuccessful in yielding improved overall agreement between theory and experiment over the entire field range.

**F. Nonlinear States at High Fields**

Beyond the codimension-two point the finite-amplitude flow was split into three regions distinguished by their characteristic wavenumbers $k_n \simeq 3.4, 3.6$ and $3.9$. These regions are shown in Fig. 15. Figure 23 shows some characteristic patterns. Qualitatively the patterns appear similar, each exhibiting spatio-temporal chaos. The transitions between these state depended on both $h$ and $R$. They were determined by measuring the change in $k_n$ as $h$ was varied at a given Rayleigh number. As already mentioned above, to our knowledge there are no theoretical predictions for these patterns.

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[1] H. Bénard, Rev. Gen. Sci. Pure Appl. 11, 1261, 1309 (1900); and Ann. Chim. Phys. 23, 62 (1901).

[2] A large literature pertaining to this field has evolved. Particularly useful as introductions to early work are the reviews by E.L. Koschmieder, Adv. Chem. Phys. 26, 177 (1974); and in Order and Fluctuations in Equilibrium and Nonequilibrium Statistical Mechanics, XVIIth International Solvay Conference, ed. by G. Nicolis, G. Dewel, and J.W. Turner (Wiley, NY, 1981), p. 168; and by F. Busse, in Hydrodynamic Instabilities and the Transition to Turbulence, edited by H.L. Swinney and J.P. Gollub (Springer, Berlin, 1981), p. 97; and in The Fluid Mechanics of Astrophysics and Geophysics, Vol. 4, "Mantle Convection, Plate Tectonics, and Global Dynamics", edited by W.R. Peltier (Gordon and Breach, 1989).

[3] For reviews of convection in NLCs, see for instance E. Dubois-Violette, G. Durand, E. Guyon, P. Manneville, and
The properties were evaluated from the formulas in Appendix B of Ref. [13] for $T = 25.6^\circ$C and $T_{NI} = 35.1^\circ$C. For the elastic constants we have $(k_{11}, k_{22}, k_{33}) = (0.609, 0.403, 0.880)$ dynes. For the Lesley viscosity coefficients we used $(\alpha_1, \alpha_2, \alpha_3, \alpha_4, \alpha_5, \alpha_6) = (-0.0718, -0.804, -0.0426, 0.651, 0.651, -0.19)$ dyne s/cm$^2$. The density was 1.022 g/cm$^3$ and the expansion coefficient was \( \alpha = 0.895 \times 10^{-3} \text{ K}^{-1} \). We used \((k_{\perp}, k_{\parallel}) = (0.677 \times 10^{-3}, 1.136 \times 10^{-3}) \text{ cm}^2 / \text{s} \) and \( \chi_a = 1.132 \times 10^{-7} \text{ cm}^3 / \text{g} \).

For a recent review, see for instance, M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. 49, 545 (1994).

Cell 4 was used to obtain the results reported in Ref. [14], although since that work it had been disassembled and the sample had been changed.

We used typical fluid properties of MBBA, as cited by E. Bodenschatz, W. Zimmermann, and L. Kramer, J. Phys. (Paris) 49, 1875 (1988), to compute the theoretical curve for $R_c$.

For a summary of bifurcation types, see for instance Appendix A of P. Bergé, Y. Pomeau, and C. Vidal, Order within Chaos (Wiley, NY, 1986).

For a recent review, see for instance, M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. 65, 851 (1993).

For a recent review, see for instance, C.W. Meyer, G. Ahlers, and D.S. Cannell, Phys. Rev. A 23, 220 (1986).

See, for instance, C.W. Meyer, G. Ahlers, and D.S. Cannell, Phys. Rev. A 44, 2514 (1991).

For small $\epsilon > 0$ the unstable pure conduction state was extremely long lived because it had to evolve from spontaneous fluctuations. For $\epsilon \lesssim 0.015$ its lifetime exceeded the five-hour equilibration time used in the experiment, and thus there are no data with increasing $\Delta T$ in this $\epsilon$-range. With decreasing $\Delta T$, the decay of the amplitude to its new steady state was relatively fast and data very near $\epsilon = 0$ could be taken.

Tschammer, Ph.D. Thesis, University of Bayreuth, Bayreuth, Germany, 1997 (unpublished).