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OSCILLATORY AND STEADY CONVECTION IN A NEMATIC LIQUID CRYSTAL

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1. INTRODUCTION

Thermal convection is a phenomenon that occurs in a liquid, heated from below, when the vertical temperature gradient, ΔT, exceeds a certain threshold value, ΔT_c [1]. The study of convection was traditionally a field mostly reserved for the hydrodynamicists, but has recently also become a relevant topic in physics[2]. The main reason for this is the upsurge of interest in nonlinear dynamics and chaotic phenomena, and the development of turbulence from stationary flow has been one of the main testing grounds for the theoretical predictions.

Another reason for the interest of the physicists is the similarity of convection, and the driven systems in general, to phase transition problems. As predicted by Landau[3], and experimentally tested by several people, the convection threshold at control parameter R_c (proportional to ΔT_c, see below) is similar to a phase transition. Thus the amplitude v of the convection velocity behaves as $v \propto (R-R_c)^{0.5}$ and relaxes as $\tau \propto (R-R_c)$. v therefore plays the role of the order parameter in an equilibrium system obeying mean field theory.

Presently, in equilibrium physics, much interest attaches to systems described by two or more order parameters. Such systems display complex critical behaviour with multicritical points. On general grounds one expects similar phenomena to occur also in nonequilibrium systems, as recently worked out for the two-mode laser by Agarwal and Dattagupta[4]. The existence of competing relaxation times then leads to the possibility of breaking also of time symmetry. Thus, in addition to the time average of the order parameter, its time behaviour or attractor character is of great interest.

Among the convective systems for which one expects multicritical phenomena to occur are binary liquids, thermohaline systems, metallic liquids or nematic liquid crystals in a magnetic field. In all of these systems one expects steady convection to be preceded by a regime of oscillatory convection. By varying the temperature gradient and another suitable parameter, the bifurcation points for stationary and oscillatory convection move along critical lines and intersect at a multicritical point. As recently pointed out by Brand et al.[5], this point is a codimension-two bifurcation.

It is the purpose of this paper to demonstrate that a nematic liquid crystal in a vertical magnetic field is a convenient physical system for studying these phenomena, and that neutron scattering provides us with an ideal experimental tool. The motivation of our studies and some results have been presented in an earlier paper[6]. In the experiments that we report on now we have used a setup that has allowed us to study a wider range of temperature gradients and magnetic fields, which has enabled us to disentangle most of the intricate phase diagram. The information that we have obtained on the time behaviour of the order parameters and of the attractors in phase space is still incomplete.

2. THEORETICAL SURVEY

In a liquid layer, heated from below, the threshold for steady convection is given by a critical value R_c^s of the Rayleigh number R^s defined by[2]

$$R^s = \frac{\beta g \ell^3 \Delta T}{\nu \kappa} \qquad (1)$$

β: volume thermal expansion

g: gravitational acceleration

ℓ: vertical layer thickness of the liquid
ΔT: vertical temperature difference
ν: kinematic viscosity
κ: heat diffusivity.

For rigid boundaries and a high aspect (width to depth) ratio of the sample, the calculated value of R_c^s is 1708. For an isotropic liquid crystal of layer thickness 1 cm this typically corresponds to $\sim 1\ ^oC$. At a lower aspect ratio R_c^s will be higher[7], thus ΔT_c^s will for aspect ratio 2 be more than a factor 10 higher.

Our system is a nematic liquid crystal in a vertical magnetic field (H). Lekkerkerker[8] was the first to predict that steady convection should be preceded by an oscillatory regime, and Guyon et al.[9] gave the first experimental verification of its existence. The calculated threshold gradient ΔT_c^o for the oscillatory phase is, when $H \to 0$, more than a factor two lower than ΔT_c^s, ΔT_c^o is predicted to increase as

$$\Delta T_c^o(H) = \Delta T_c^o(0)\left[1+(H/H_c)^2\right]. \qquad (2)$$

Here $H_c \sim 10$ G for $\ell = 1$ cm. An oscillatory phase is expected only below a field $H^{max} \sim 350$ G.

The theoretical predictions about the nature of the bifurcations at R_c^o and R_c^s are somewhat conflicting. Dubois-Violette and Gabay[10], using a Landau-type expansion, find it to be an inverted bifurcation (i.e. similar to a first-order transition), which probably changes to a normal bifurcation above a cross-over field $H_1 \sim H^{max}$. In their theory R_c^s decreases with the magnetic field, but the associated bifurcation to the stationary regime is always of the normal (i.e. of the second-order) type.

In a penetrating series of articles da Costa, Knobloch, Proctor and Weiss[11] have calculated the velocity amplitudes, i.e. the order parameters, and their motion in phase space as a function of R for thermohaline and magnetoconvection. Although they do not treat our problem directly, their predictions are probably applicable. For magnetoconvection they find that both the oscillatory and the steady branches can be either subcritical or supercritical, i.e. R_c corresponds to either a first or a second order transition. They pay special attention to the junction between the two branches. When the steady branch is supercritical, the oscillatory branch ends on it via a Hopf bifurcation. When the steady branch is subcritical, the oscillatory branch ends on it via a heteroclinic orbit.

Da Costa et al.[11] also consider the behaviour of the oscillations. Their stability is predicted to depend on the bifurcations bounding the oscillatory regime, i.e. on the aspect ratio and the magnetic field, and on R.

All authors agree that the oscillatory period should become infinite as $H \to H^{max}$. Brand et al.[5] point out that, if in addition $R \to R_c^s$, the two regimes join in a multicritical point.

Experiments

Our present experimental setup differs from that used in our earlier report[6] in three ways: The aspect ratio is 2, the heat conductivity of the sidewalls of the sample cell is lower, and the magnetic field can be made stronger. The former means a better separation of the two flow regimes, while the latter two imply that we can investigate a wider range in the parameters R and H.

The sample cell is a parallelepiped of horizontal dimensions 50 and 4 mm and height 25 mm. In such a geometry one expects two convection rolls with axes along the shortest edge, but this point has so far not been checked. The sample is fully deuterated para-azoxy-anaisole (PAA), which is a nematic liquid crystal for $118 < T < 135$ °C. Pure samples may also be supercooled, and it was important in our experiments that we could supercool it to ~ 100 °C. The top, bottom and sidewalls are made of Al, Cu and stainless steel, respectively. A temperature gradient is obtained by setting the difference of the power fed to electrical heating elements at the top and bottom of the vessel. The temperature of the bottom plate is controlled and kept constant throughout the experiment. The temperature difference between the top and bottom plate is measured continuously by thermistors and thermoelements. With this arrangement ΔT stayed constant within $\pm$ 0.01^o for hours. The strength of the vertical magnetic field, which is the other parameter of the experiment, can be chosen up to 1.2 G, although only below 1 kG for extended periods.

The neutron wavelength was 1.25 Å. The scattered intensity was recorded at $Q \sim 1.8$ Å^{-1}, at the maximum of the first liquid diffraction peak of nematic PAA. The intensity of this peak is very sensitive to the molecular orientation, which in its turn depends on the convective flow. Hence we use the molecular orientation as an internal probe for the flow, and monitor it with a neutron beam. In the absence of flow the molecules are aligned parallel to the field. It can be shown that the intensity (i.e. its deviation from that of the fully aligned, flow-free state) is proportional to the order parameter. With a vertical magnetic field the deviation is negative, hence we use an inverted intensity scale in all the figures below.

The neutron beam covers the whole sample, and hydrodynamic fluctuations and oscillations of the relevant wavelength range show up as real-time intensity variations. The time-averaged intensity gives the order parameter.

Fig. 1 gives the time-averaged intensity as a function of ΔT. The curve at 1.2 kG gives ΔT_c^s for steady convection of isotropic PAA, and the threshold value is of the expected magnitude for the given sample geometry. At each gradient setting used when measuring the 1.2 kG-curve we also measured intensities for other field values, and data for 300 and 600 G in the steady flow regime are given in Fig. 1. The continuation of the latter two curves in the oscillatory regime is given below.

In Fig. 2 we give a complete curve for 300 G. Starting at the lowest gradient, the neutron intensity, i.e. the order parameter, follows the upper curve, crosses itself, and meets the 1.2 kG-curve at A. There it makes a dramatic fall, rises and joins the same curve at a higher point B. On its way back from higher gradients it goes towards C.

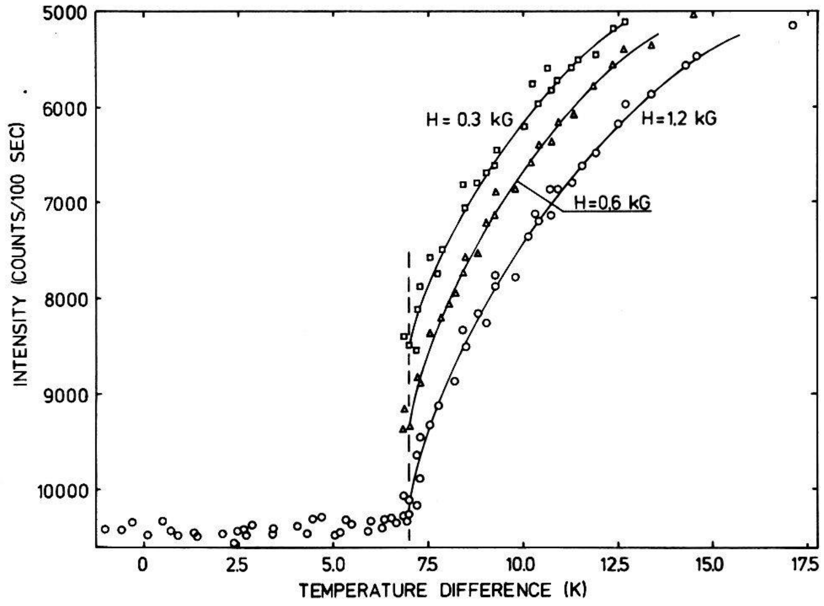

Fig. 1. Neutron intensity versus temperature difference between lower and upper plate, showing threshold to steady flow regime for 1.2 kG. Data points for the two lower fields in oscillatory regime are given in figures below.

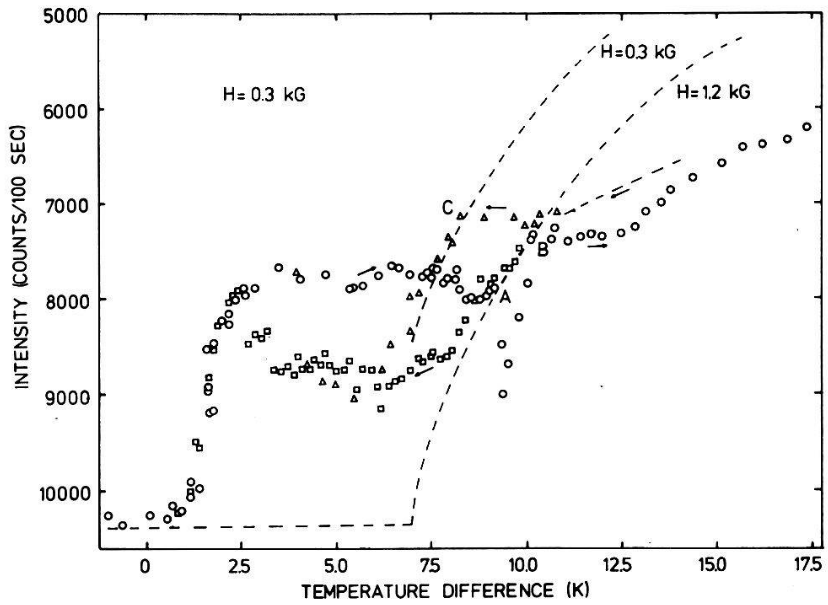

Fig. 2 Neutron intensity versus temperature difference for H = 300 G. Arrows indicate direction. Two different observed return paths are shown. The curves drawn for 1.2 kG and 300 G are taken from Fig. 1.

When it meets the 300 G-curve of Fig. 1 it joins it, after making a shallow dip. It steps down on the latter curve and follows a lower path back to the beginning of the loop. Other return paths are possible, and another route is indicated by the squares. The significance of some experimental points at levels between the upper and lower path will appear when we now discuss data for 600 G, shown in Fig. 3. Starting from left, the order parameter again passes through A, B and C, but from C it now moves

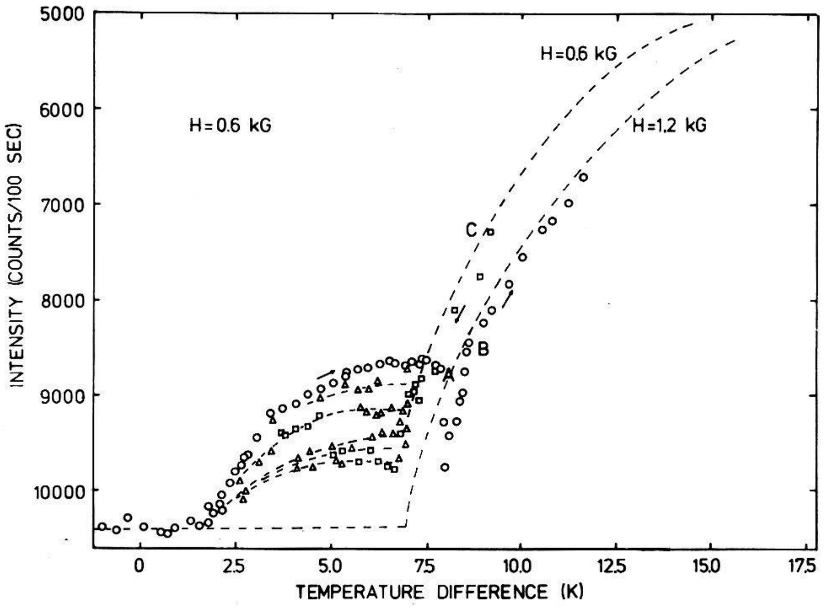

Fig. 3. Neutron intensity versus temperature difference for H = 600 G. Curves drawn for 1.2 kG and 600 G are taken from Fig. 1. Triangular symbols give experimental points at levels reached by manipulation of field and gradient.

back through the oscillatory regime along a lower level, but rejoins the upper curve after passing some intermediate levels. During this loop the field was kept constant. Additional data were taken in the oscillatory regime at many gradient settings where we let the magnetic field go through a sequence of values before setting it at 600 G again. The multitude of points obtained this way are also given in Fig. 3. It is evident that the order parameter is multistable in the oscillatory regime. In fact one may move to the right along any of these levels until one meets the 1.2 kG curve, from which point one goes through a loop similar to ABC. We have at least verified this for three of the levels.

Discussion

The data above are for three different values of the applied magnetic field. We have similar data also for four other fields in the range 60-900 G. In all of the cases do we find that the oscillatory regime is initiated by a continuous (normal, 2nd order) instability, with the possible exception of the lowest field, for which the threshold is masked by a tail extending to the steady convection regime at negative gradients, i.e. heating from above. The abscissa is thus a critical line of 2nd order transitions.

In the oscillatory regime there is multistability with five levels, possibly more, for the order parameter in the upper range. As far as we can judge, each level of the order parameter joins the curve for the same field of Fig. 1 when it intersects the vertical, broken line. Thus the latter line is another critical line, probably of first order. On this curve terminates a family of curves like in Fig. 2, each curve marks the lower boundary for steady flow regime for each field value. The upper boundary for the oscillatory regime is marked by the 1.2 kG curve, which is another critical curve. As seen in Figs. 2 and 3, the order parameter makes a dramatic drop during its encounter with this curve, when it enters the steady regime. A similar dip, but much more shallow, is seen when it again leaves the steady regime at the lower boundary.

The steady regime is probably chaotic: the order parameter may be led to almost any point by proper manipulation of field and gradient. The direction of its motion is much determined by the moving force, i.e. by the stepsize of the gradient. If we try to move the order parameter too fast out of the steady regime, it collides with the lower boundary and slides along it until it enters one of the possible levels of the oscillatory regime.

The threshold for the 1.2 kG-curve is a multicritical point at which three critical lines intersect: a second-order line for the onset of the oscillatory regime, and two first-order lines for the boundary between the oscillatory and the steady regimes.

The experiments reported on here do not give an exhaustive information on the dynamics of the order parameter. Such information requires additional data with counting periods shorter than the 100 seconds mostly used, and perhaps also more densely spaced in ΔT. Still it seems clear that regular oscillations are rare. The order-parameter levels in the oscillatory regime mostly look metastable and the dynamics is dominated by occasional excursions or transitions to other metastable states.

It is possible that the lack of stability of the oscillations is connected with the first-order character of the oscillatory-steady transition, as discussed by Da Costa et al.[11]. We are planning new experiments on a tall and narrow (low aspect-ratio) cell, for which they predict a second-order transition.

In earlier experiments on a cell of aspect ratio close to one[6] we observed an intensity peaking at the oscillatory threshold which we tentatively ascribed to critical fluctuations. In view of the present experiments this peak could possibly be due to a transition between two levels of the order parameter at a gradient slightly above ΔT_c^s.

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