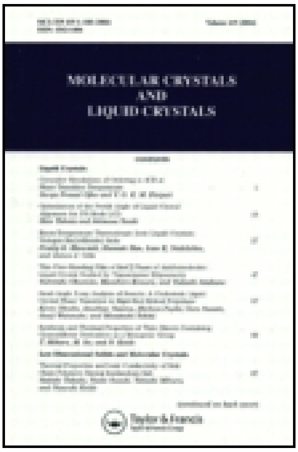

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ORIGIN OF THE FLUCTUATIONS OF THE NEUTRON INTENSITY SCATTERED BY A NEMATIC LIQUID CRYSTAL IN A RAYLEIGH-BENARD FLOW.

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Abstract The sample is a nematic liquid crystal (PAA) in a Hele-Shaw cell. This cell is submitted to a vertical temperature difference ΔT and a magnetic field H. The temperature difference destabilizes the system, producing the well-known Rayleigh-Bénard rolls. The magnetic field stabilizes the system. The combination of both produces a phase diagram. T. Riste et al. showed that, for moderate temperature difference and fields, periodic fluctuations of the neutron scattered intensity appear. Moreover the squared frequencies are linear versus ΔT over adjacent ΔT intervals (frequency quantification). The aim of this experiment was to determine the origin of these fluctuations by simultaneous neutron scattering time series measurements and optic observation thanks to a transparent cell.

In our first experiment with a cell similar to Riste's we have been able to reproduce the first part of his observations ; our conclusion is that the fluctuations do not arise from the rolls themselves but from oscillators which periodically perturb the rolls structure.

INTRODUCTION

After having demonstrated the existence of "frequency quantification" in rectangular samples as in square samples Riste et al.[1,2,3] pointed various possible origins for these periodic fluctuations. Namely there are two schools : some propose[4] that the time dependence arises from boundary layers eruption from the bottom, others put forward[5,6] the existence of particle like excitations when coming close to the turbulence.

The aim of this experiment was to look to the overall Rayleigh-Bénard rolls structure simultaneously with the neutron scattering time series recording, in order to attribute unequivocally the origin of the fluctuations first observed by Riste et al.

EXPERIMENTAL

In order to make easier the comparison with Riste's work we built a sample cell very similar to the one where the frequency quantification was first observed, filled with the same sample, deuterated para-azoxyanisole (d-PAA). The inner dimensions of the cell are 30 mm x 30 mm for the large lateral faces and 3 mm for the thickness. The large faces are vertical, the thickness direction is in the horizontal plane (so called Hele-sShaw configuration). The large faces are made from 1 mm thick sapphire, kept apart by sapphire spacers. The top and bottom are closed by copper blocks. All parts of the cell are glued together by polysiloxane high temperature glue (CAF 730 from Rhône-Poulenc which avoids significant acetic acid or gaz discharges during polymerisation). The upper copper piece is connected to a small tank ended by a flexible tube (Kapton 200XP919 from Dupont de Nemours). The cell is filled through this tube which can eventually be closed or sealed. The small tank accomodates the liquid crystal thermal expansion.

The experimental cell is placed in between four permanent magnet bars. Their assembly produces a vertical magnetic field of 250 Oerstedt which does not vary more than 5% over the sample area.

The whole set itself stands in the center of a large vessel which it is possible to evacuate. This vessel is placed on a neutron diffractometer. The neutrons enter it through a small quartz window. The beam is limited by a channel of absorbing material before impinging on the sample, the transmitted beam is absorbed by a neutron beam catcher located inside the vessel : that way the two usually main background sources (windows in and out) are strongly limited. The neutron beam scattered by the sample leaves the vessel through an aluminium window. The periodic recording of the intensity of the scattered neutrons (time serie) may start once the sample plane is aligned parallel to the scattering vector and the detector is set at the angular position where the maximum intensity is measured (scattering from the first liquid ring). We shall not detail here the theory of neutron scattering by condensed matter, which has been done for this precise case several times[1]. Let us merely remind that the position of the first liquid ring of a nematic liquid crystal is correlated to the transverse distance between the first neigbour molecules ; it follows easily that the neutron flux falling into the detector will depend upon the number of molecules having their equatorial plane close to the horizontal scattering plane. The time serie intensity will allow to appreciate qualitatively the amount of ordering.

The sample is illuminated from behind by diffuse light. On its main axis is placed a semi transparent mirror which allows a video camera to superpose the sample image and an image of the controlling computer screen. The images from the camera can be recorded on a VHS tape recorder or digitalized and stored in the computer.

It is well-known that non linear systems are very sensitive to perturbations, which is the case here. Therefore we tried especially to avoid temperature fluctuations (the stability of each copper plate is about 0.02°C) and moreover thermal losses. Indeed a previous experimental set up showed some flow even at $\Delta T=0$. Here the whole cell and the surrounding holding parts were wrapped in aluminised mylar, except on the neutron and observation trajectory. This last part was thermally screened by two sapphire plates coated with gold. Meanwhile, in order to achieve the no movement at $\Delta T=0$ condition, we had to reduce the pressure of air inside the vessel. This is in no way trivial as one may pump out the sample if it is not properly sealed ; nevertheless, if the sample is sealed, a too high pressure may happen inside the sapphire cell and break it. Moreover adsorbed gazes or PAA partial pressure vapour usually create bubbles which are very difficult to remove. It does not seem that the presence of a limited bubble on top of the cell changes much the oscillation frequencies ; meanwhile that may broaden the line or most probably change the transient which will allow the oscillating mode to establish itself or disappear. Finally we decided to seal the sample, to pump the vessel down to 80 mbar, and to get rid of the bubbles ; all measurements reported below were made according to these conditions.

The temperature of the lower copper block was kept at 132°C. The temperature of the higher block was smaller. As PAA can be easily supercooled down to 92°C, large ΔT can be reached.

SPATIO-TEMPORAL PATTERN IN A SQUARE CONTAINER

In their papers Riste et al. showed that all their observations could be summed up in the follwing formula [1]:

$$\omega^2 = A\left(\frac{h^2}{a^2}+\frac{k^2}{b^2}\right)(\Delta T-\Delta T_c^{h,k})(T-T_0)$$

which lead to speak about "quantification of frequencies". The reason for the h,k integer indexes remains obscure while it was proposed that they correspond to a number of rolls versus the horizontal (size of the cell a) and the vertical (size of the cell b).

We report our main results in the same way as Riste on the Figure 1. Most often several modes are apparent, which are not always (sub)harmonics. The most prominent modes are shown as circles. Three regions can be distinguished.

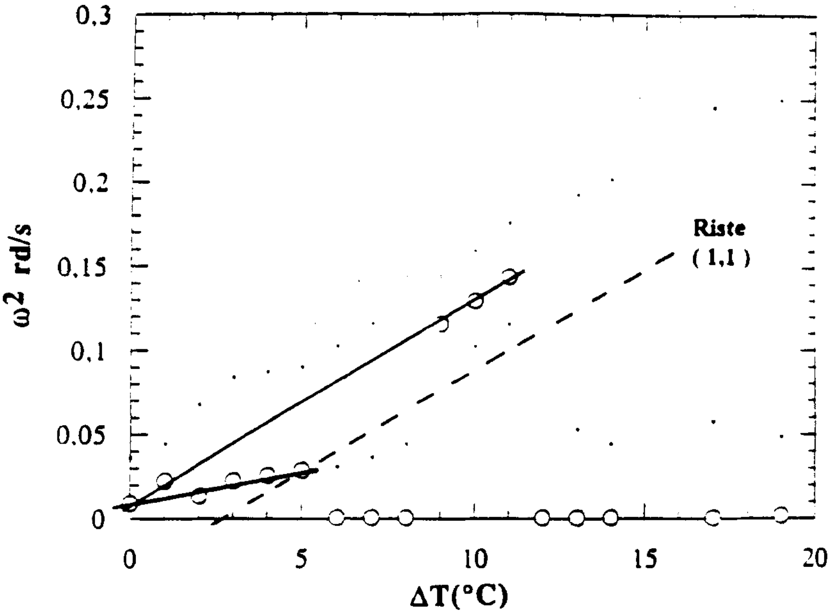

FIGURE. 1 Frequencies quantification.

The circles o correspond to the most prominent frequencies. The • to minor ones. The dashed curve represents the (1,1) mode of Riste et al.[3]

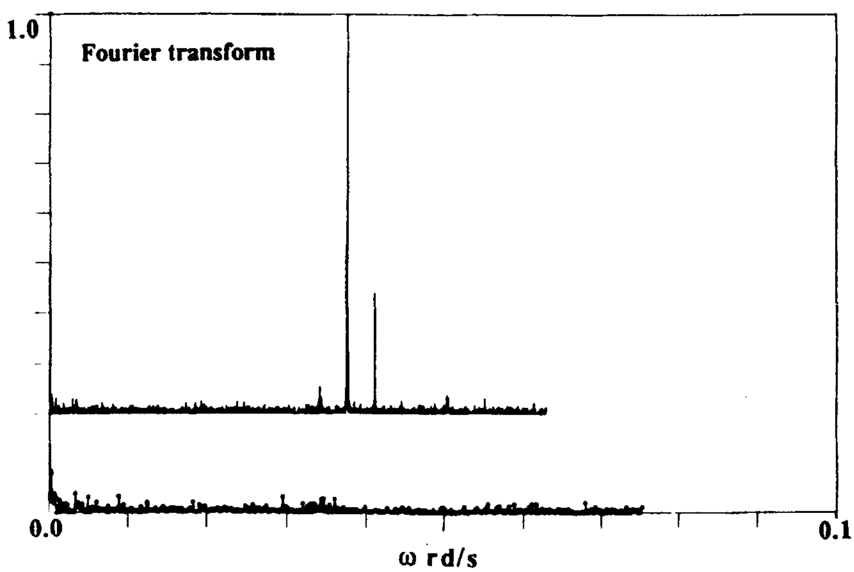

FIGURE. 2 Fourier transform of a time serie for ΔT=11°C.

The ordinates is in arbitrary units. The lower curve is a10x enlargement of the low frequencies. The full abscissa scale of the upper curve is 1 rd/s

In the low temperature region not well defined modes exist, showing always very broad lines. The frequencies seem to align over a straigth line passing through the origin. Metastability is observed at the larger ΔT. The video observation shows very instable rolls, mainly two large ones, with fast varying borders.

The intermediate region is centered over $\Delta T=10°C$. It includes few points. It is only for that region that we find very narrow frequencies (Fig.2) as reported before by Riste et al.. These frequencies also seem to align over a straigth line passing through the origin. We have drawn on the same diagram a broken line showing the mode [1,1] from Riste. The parallelism of it with the line for our intermediate region is striking : the common slope must be characteristic of the PAA sample at this temperature. One should not be bothered with the 2.5°C gap between the two lines. Riste et al. mention that they had to introduce thresholds in their quantification formula. Once we took care of thermal losses as explained above, the possible threshold in our experiment became a small fraction of a degree. The video observation shows now stationnary rolls which are perturbed by oscillators (Fig. 3) ; these are small rolls, coming in general in alternance from top and bottom. Each of these perturbs either the right or the left side of the cell without mixing. They exhibit a droplet shape. It may happens that two of them pertub simultaneously one side of the cell, usually one is ascending the other descending and they tend to roll around one another building a S-shaped border between them. The most important point is their periodic appearance which reflects in the observed neutron frequencies.

The ΔT temperature larger or equal to 12°C form the last region. Some very low, badly defined frequencies can be observed. Mostly for $\Delta T=12$ or 13°C the time series exhibit metastability which results in transitions between various regimes ; the data treatment is made difficult by the relatively small number of points in each coherent interval. The video still shows small rolls perturbing the main ones as in the chaotic metastable sequence in the first regime (Fig 4) ; the difference is that the appearance of the perturbators is now chaotic.

DISCUSSION

The observed frequencies values are robust. We found already some of the displayed points during a previous measurement where bubbles remained. Meanwhile one may ask oneself about the difficulty to reproduce the full extension of Riste's results. Certainly our cell is much more insulated from outside then his. A major discrepancy comes from the nature of the main wall of the cell. While sapphire has a good heat conduction, the temperature of the sapphire walls is far from being linear versus the height, which is more

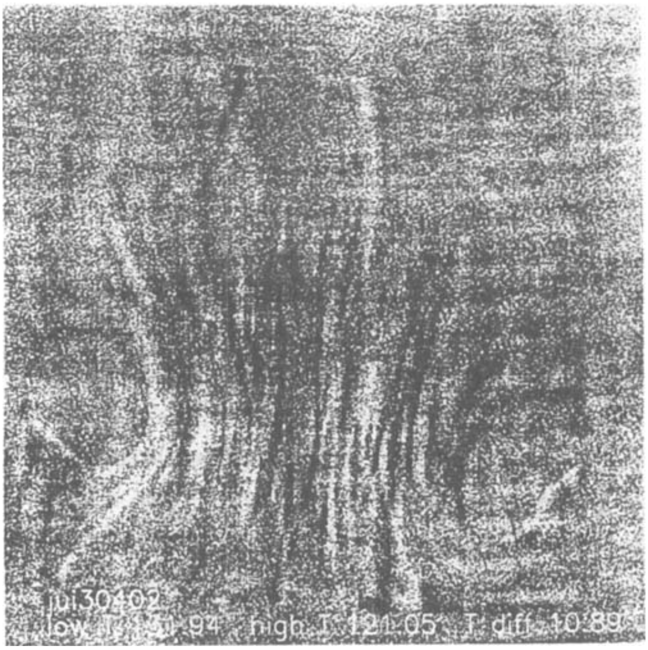

FIGURE. 3 Video for ΔT=11°C. Periodic behaviour

FIGURE 4 Video for ΔT=9°C. Chaotic behaviour

probable to be achieved with Riste's metallic cell. All this means that, while all the care taking, the boundary conditions may still differ resulting in our higher instabilty and even intermittencies.

Another discrepancy lays in the applied magnetic field 250 Oe here instead of 100 Oe or less in Riste's experiment. We do not expect that this feature is really significant.

CONCLUSION

Apart from a very small ΔT threshold we find again Riste's observations, while limited to a few temperature differences which build a very narrow segment. The origin of the perodic fluctuations arises in oscillators which perturb the stationnary rolls structure. These perturbations exist as well in the chaotic sequences, the difference is that their occurence is not more periodic.

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