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CRITICAL SLOWING DOWN NEAR THE RAYLEIGH-BÉNARD CONVECTIVE INSTABILITY

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Résumé. — On a mesuré, à l'aide de la technique de diffusion Rayleigh forcé, le temps de relaxation des fluctuations de température dans une mince couche horizontale de fluide soumise à un gradient de température orienté vers le bas. On a trouvé que ce temps de relaxation augmente beaucoup lorsque le gradient de température s'approche de la valeur critique correspondant à l'instabilité de convection de Rayleigh-Bénard, et que cette augmentation est la plus grande pour des longueurs d'onde spatiales proches du double de l'épaisseur de la lame de liquide.

Abstract. — The decay time of temperature fluctuations in a thin horizontal fluid layer subjected to a downward temperature gradient has been studied by forced Rayleigh scattering. The decay time was found to increase markedly as the temperature gradient approached the critical value for the onset of the Rayleigh-Bénard convective instability and was most pronounced for spatial wavelengths close to twice the thickness of the fluid layer.

In recent years there has been a marked resurgence of interest in hydrodynamic instabilities [1]. Laser light scattering experiments have been particularly effective in probing the velocity fields in both the Rayleigh-Bénard and circular Couette flow instabilities in the heterogeneous regimes above the primary bifurcation where spatially ordered hydrodynamic flow structures exist [2, 3, 4].

It has also been widely recognized that there should be interesting pretransitional phenomena in the homogeneous regime preceding the first transition as these systems become unstable against particular hydrodynamic modes. In analogy to equilibrium phase transitions, theory predicts both an increase in the magnitude of thermally excited fluctuations of the unstable modes and a divergence in their relaxation times, i.e. critical slowing down [5, 6, 7].

Direct observation of these pretransitional effects has proven to be elusive, however, primarily because of the characteristically long wavelengths of the critical modes. In principle, the critical fluctuations near the Rayleigh-Bénard instability can be studied directly by light scattering since they should produce a marked increase in the scattered light intensity. But the very small angle light scattering measurements

required are technically difficult because of inevitable stray light problems as discussed by Lastovka [8]. Therefore, various indirect approaches have been proposed. For example, Riste [9] has looked for enhanced neutron scattering associated with orientational fluctuations in a liquid crystal layer near the instability. Clark [10] has examined the refractive index changes in a fluid layer near the transition when heated by a fine wire, and Lekkerkerker, Boon and Lastovka [11] have proposed studying the diffusion constant of suspended Brownian particles which should be modified near the transition by the enhanced velocity fluctuations.

In this letter we report a preliminary observation of critical slowing down in a horizontal fluid layer approaching the Rayleigh-Bénard convective instability from below. We have employed a variant of the forced Rayleigh scattering technique [12] in which a spatially periodic horizontal temperature variation is induced in the fluid by a brief pulse of intense argon ion laser light (which is absorbed by the fluid), and the decay of the resulting refractive index variation following the pulse is monitored by observing the diffraction of a weak cw He-Ne laser beam (to which the fluid is transparent) by the resulting thermal grating. We have observed a marked increase in the decay time of the diffracted intensity as the constant vertical temperature gradient is increased towards

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its critical value, and we find that this critical slowing down is most pronounced for spatial wavelengths of the thermal grating close to the critical wavelength of twice the cell height.

1. **Theory.** — Zaitsev and Shliomis [5] and Lekkerkerker and Boon [6, 7] have analysed the hydrodynamic fluctuations in a horizontal fluid layer subjected to a vertical temperature gradient. The usual separation of transverse modes (viscous shear modes) and temperature modes (thermal diffusion modes) found for homogeneous fluids no longer occurs. Instead, for each wavevector $\mathbf{k}$, one finds a pair of coupled modes, each of which is a combination of transverse velocity and temperature. The relaxation rates for the two coupled modes of wavevector $\mathbf{k}$ are given by : [7]

$$\lambda_{\pm} = (k^2/2) [(v + \kappa) \pm \{ (v + \kappa)^2 - 4 v \kappa [R_c(a) - R]/R_c(a) \}^{1/2}] \quad (1)$$

where v is the kinematic viscosity, κ is the thermal diffusivity, R is the Rayleigh number which is proportional to the temperature gradient, and $R_c(a)$ is the critical Rayleigh number for instability against a fluctuation whose wavevector $\mathbf{k}$ has horizontal component a . (We will use reduced units for $a = 2 \pi d/l$ where d is the height of the fluid layer and l is the horizontal wavelength of the fluctuation.)

Since $R_c(a)$ has a minimum for $a = 3.116$ [13], the decrease in the slow root λ_- with increasing temperature gradient should be most pronounced for values of a close to 3.116, or of l close to $2 d$. Because of the large Prandtl number (v/κ) which is about 16.6 in ethanol, the fast mode λ_+ decays ~ 16.6 times faster than the slow mode at $\Delta T = 0$ and is only slightly changed as ΔT is varied, while the relaxation rate of the slow mode λ_- decreases nearly linearly from $\lambda_- = \kappa k^2$ at $\Delta T = 0$ to $\lambda_- = 0$ at $R = R_c(a)$.

We note that eq. (1) contains an ambiguity in the relation between k and a . The horizontal component of the wavevector a will be determined by the experimental geometry, but the vertical component is not well defined. Lekkerkerker and Boon took $k_{\perp} = \pi/d$ which is a good first approximation, but which describes modes that are inconsistent with the boundary conditions imposed on the flow by the rigid upper and lower surfaces of the Rayleigh-Bénard cell.

In order to obtain more precise theoretical predictions, we have made a normal mode analysis of the linearized hydrodynamic equations, with the appropriate rigid boundary conditions, following the variational method discussed by Chandrasekhar [13]. A detailed discussion of this work, which is a generalization of that of Weisfred *et al.* [4] will be published elsewhere, but we give here the results which are useful for the interpretation of the experimental data to be presented in this paper.

We have solved, by successive approximation, the linearized hydrodynamics equations describing the fluid with a constant vertical gradient applied to it and two horizontal rigid and perfectly conducting plates, assuming solutions of the form

$$e^{-st} e^{iax} w(z)$$

where x and z are respectively the horizontal and vertical coordinates in reduced units. We find that $w(z)$ is by no means a simple exponential. We have determined the Rayleigh number $R(s, a)$ for each value of s and a . From there we deduce the roots of $s(R, a)$, and we find two values : s_- , the smallest root and s_+ , the largest root. If we let R go to zero, $s_-(R, a)$ is equal to λ_- from eq. (1) provided we set

$$k^2 = \left(\frac{\pi}{d}\right)^2 + \left(\frac{a}{d}\right)^2$$

in that equation, but $s_+(R, a)$ differs significantly from λ_+ (for the same value of k). Actually we obtain $s_+ = f(a) \lambda_+$. It is interesting to note that $f(a)$ does not depend upon the value of the Prandtl number (calculations have been performed for three values of the Prandtl number 16.6 (ethanol), 3.6 (acetone), 0.7 (gas)). We show $f(a)$ in figure 1.

FIG. 1. — Value of the rate constant of the fast mode $s_+(0, a)$ for a vanishing value of the Rayleigh number expressed in terms of $f(a) = s_+(0, a)/(v(a^2/d^2 + \pi^2/d^2))$.

Using these roots $s_-(0, a)$ and $s_+(0, a)$ we find that $s(R, a)$ is given to a good approximation by solving the equation :

$$R = (y - s_-(0, a))(y - s_+(0, a)) \frac{R_c(a)}{s_-(0, a) s_+(0, a)} \quad (2)$$

where $R_c(a)$ is the critical value of R for a given value of a (as is eq. (1)).

We have given these results as they justify the procedure used to reduce the experimental data presented below. In addition the fact that $f(a) \sim 2$ for $a = 3.116$, allows us to make the following comment : Lekkerkerker and Boon have shown that the

correlation function of the temperature fluctuations, which turns out to be exactly proportional to what we measure in the version of the forced Rayleigh experiment, is the sum of two exponentials, with a small weight for the fast decaying mode (λ_+). Due to the fact that $s_+(R, a)$ is significantly larger than λ_+ given by eq. (1), we do not expect to be able to observe experimentally the contribution of the fast mode. (Its weight is probably smaller and our set up is not designed to observe the initial decay of the signal due to the finite duration of the heating part of the experimental cycle.)

2. Experiment. — The experimental arrangement is illustrated schematically in figure 2. The Rayleigh-Bénard cell is 12×70 mm with a gap of 1.65 ± 0.05 mm between the upper and lower copper plates. The upper plate was kept at 20°C by circulating water, while the lower plate was electrically heated. A $40 \mu\text{V/K}$ chromel-alumel thermocouple measured the temperature difference across the cell and also controlled the heating current, providing a temperature difference ΔT constant to ± 0.1 K. The cell was filled with ethanol containing a trace of red ink which partially absorbed the green (but not red) laser light. The critical ΔT for the appearance of convective rolls was $\Delta T_c = 5.25 \pm 0.2$ K ($210 \pm 10 \mu\text{V}$), in agreement with the anticipated value of 5.00 ± 0.50 corresponding to a critical Rayleigh number $R_c = 1\,707$.

FIG. 2. — Experimental apparatus : A, argon ion laser ; B-B', beam expander ; S, electrically activated shutter ; C, cylindrical lens ; G, brass grid ; M, red-reflecting mirror ; H, He-Ne laser ; HS, horizontal slit ; RB, Rayleigh-Bénard cell ; L, 4 meter focal length lens ; F, green-absorbing filter ; MS, movable slit ; PM, photomultiplier ; MSC, digital multiscaler ; SR, strip chart recorder.

The 800 mw (all lines) output of a Coherent Model 53 argon ion laser was enlarged to 5 cm diameter, collimated and then focussed vertically by a cylindrical lens. The laser output was normally blocked by an electrically actuated shutter which periodically opened briefly (≤ 0.1 second). Before reaching the cell, the light traversed a vertical brass grid which transmitted a horizontal square wave into the fluid. Grids of spatial wavelength $l = 1.5, 2, 2.5, 3, 3.5$ and 4 mm were used sequentially in the experiment.

The effective absorption coefficient of the colored ethanol for the argon laser beam was $\sim 0.3 \text{ cm}^{-1}$ leading to a maximum heating of the ethanol of 6 mK during a 0.1 second pulse.

The $6\,328 \text{ \AA}$ beam from a Spectra Physics Model 120 He-Ne laser was expanded, collimated and masked by a horizontal slit to produce a horizontal strip of red light which was continuously incident on the aperture of the cell colinearly with the green beam. A 4 m focal length lens and green-absorbing filter beyond the cell formed a vertical line image of the red beam on a movable vertical slit 4 m away, located directly in front of the photomultiplier.

When the shutter was kept open, a strong diffraction pattern containing many orders appeared on the slit surrounding the central maximum. The slit was positioned so that only the first-order diffraction maximum reached the photomultiplier. Thus, although the square wave temperature excitation in the fluid contains many spatial Fourier components, the detection optics select only the first Fourier component (horizontal wavelength $= l$) whose intensity determines the photomultiplier signal.

The pulse duration was set at the minimum consistent with homodyne detection. During a run, the photomultiplier output was digitized and stored in a multiscaler which provided signal averaging typically of between 10 and 50 pulses. The log output of the multiscaler was then recorded on a stripchart recorder from which the decay times could be directly determined by measuring the slope. Decay times were typically between 0.2 and 2 seconds and were not significantly affected by the number of pulses averaged in a run. In each complete experimental sequence, decay times were measured for each of the six grids with reduced temperature differences across the cell of $\Delta T/\Delta T_c = 0, 0.24, 0.48, 0.64$ and 0.81 . Approximately 8 separate measurements were performed for each combination of grid (l) and temperature (ΔT).

3. Analysis. — Following each pulse of argon laser excitation, there will be a spatial wave of both temperature and pressure in the fluid. The pressure variation decays rapidly, in about $3 \mu\text{s}$ for the 3 mm grid, on a time scale set by the sound velocity. The remaining isobaric temperature wave, in the presence of a non-zero vertical temperature gradient, is a superposition of the two mixed normal modes and will decay with the two relaxation rates $\lambda_{\pm}$ given

by eq. (1). Since the fast mode always decays at least 17 times faster than the slow mode, we have analysed our data as a single exponential corresponding to λ_- .

At zero temperature gradient, $\lambda_- = \kappa k^2$ and the diffracted intensity (and photomultiplier output for homodyne detection) decays as :

$$I(\tau) = I_0 \exp(-2 \kappa k^2 \tau) = I_0 \exp(-\Gamma \tau). \quad (3)$$

In a preliminary experiment, we measured the decay times for each grid with a tall constant temperature cell ($d \sim 1$ cm) for which the wavevector $k = 2\pi/l$. The resulting scaled decay rates :

$$\Gamma^* = \Gamma/2 k^2 \quad (4)$$

for all the grids fell in the range $(9.2 \pm 0.6) 10^{-4} \text{ cm}^2/\text{s}$, in close agreement with the calculated thermal diffusion constant of pure ethanol, $\kappa = 9.34 \times 10^{-4} \text{ cm}^2/\text{s}$ [14].

With the 1.65 mm Rayleigh-Bénard cell, taking $k = 2\pi/l$ gave values of Γ^* (at $\Delta T = 0$) which increased rapidly with l because, with d and l comparable, the heat diffusion process is no longer one dimensional. If, following Lekkerkerker and Boon, we assume that the lowest-order Fourier component of the temperature pattern determines the observed decay, then the appropriate wavevector is given by :

$$k^2 = (2\pi/l)^2 + (\pi/d)^2 \quad (5)$$

with $d = 1.65$ mm.

Measured decay rates (at $\Delta T = 0$), for the 1.65 mm cell when scaled according to eq. (4) with k given by eq. (4), were again in agreement with κ and did not depend on l .

All of our data for each of the 30 combinations of l and ΔT were reduced this way. Average values of Γ^* are shown in figure 3 for the 1.5, 2, 3 and 4 mm grids. The vertical error bars indicate the statistical variation in the data. We also show theoretical curves along with the data. These are actually linear plots which extrapolate to zero at the critical Rayleigh number ($R_c(s, a)$ at $s = 0$) for each grid, determined in the normal mode analysis described earlier to be :

$$R_c(1.5 \text{ mm}) = 4761, \quad R_c(2 \text{ mm}) = 2585.$$

$$R_c(3 \text{ mm}) = 1735, \quad R_c(4 \text{ mm}) = 1788.$$

The linear approximation for this case does not depart significantly from the prediction of Lekkerkerker and Boon in eq. (1), nor from the exact calculation discussed above. In plotting the curves, we have taken $R_c = 1707$ to correspond to the observed threshold of $\Delta T = 5.25^\circ$.

FIG. 3. — Experimental results : scaled decay rate vs. temperature difference across the cell for different spatial wavelengths. ∇ -1.5 mm, Δ -2 mm, $\circ$ -3 mm, $\square$ -4 mm. Horizontal error bar indicates uncertainty in the temperature, vertical error bar indicates typical rms dispersion of experimental results. Solid lines are linear approximation to eq. (1) in the text for horizontal wavelengths shown with $R_c(3.3 \text{ mm}) = 1707$ taken as 5.25 K .

Qualitative agreement between the experimental results and theoretical predictions is seen in figure 3 in the decrease of decay rate with increasing ΔT , and in the fact that the effect is most pronounced for $l = 3$ mm. Because the copper plates were not precisely parallel and the cell was not isolated from the air, more detailed evaluation of this preliminary experimental result is not feasible. Additional experiments will be performed in the future with a more sophisticated cell. Furthermore it will necessary to avoid some difficulties near ΔT_c , as the large temperature gradient leads to a large index gradient which bends to beam towards the cold plate.

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