Bénard-Marangoni Convection in Two-Layered Liquids

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We describe experiments on Bénard-Marangoni convection in horizontal layers of two immiscible liquids. Unlike previous experiments, which used gases as the upper fluid, we find a square planform close to onset which undergoes a secondary bifurcation to rolls at higher temperature differences. The scale of the convection pattern is that of the thinner lower fluid layer for which buoyancy and surface tension forces are comparable. The wave number of the pattern near onset agrees with the linear stability prediction for the full two-layer problem. The square planform is in qualitative agreement with recent two-layer weakly nonlinear theories, which fail however to predict the transition to rolls.

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Convection in fluids has been a fruitful system for the study of nonlinear, nonequilibrium patterns for nearly 100 years [1–3]. Bénard's original experiments [1] used shallow layers of whale oil, heated from below and open to the air above. Above a threshold temperature difference, the oil became unstable to the now classic pattern of hexagonal flow cells. Many years passed before it was conclusively shown that surface tension gradients, or "Marangoni" driving forces, were involved [4,5]. Beginning with Pearson [5], theories of this instability have traditionally neglected the dynamics of the overlying gas, replacing it by a constant heat flux boundary condition on the liquid's upper surface. This condition is experimentally ill posed, however. Well-controlled experiments [6–11] always involve two fluid layers: the overlying gas forms a second layer bounded by a plate on which a constant temperature is maintained. The heat flux across the gas/liquid interface results from conduction and convection in both layers and is not uniform once flow begins. The interface can deform, and therefore its distance from the upper plate changes the local heat flux [11].

In general, for thick layers, both Marangoni and buoyancy forces are significant [12,13]. Buoyancy in the upper layer actively assists or impedes convection in the lower via surface stresses [13–15]. Until recently, most experiments have addressed only the limiting case of a gas as the upper fluid. In this Letter, we present a precise experimental study of convection in a system of two immiscible liquids sharing a deformable interface. When the upper fluid is also a liquid, a much richer range of pattern phenomena become possible. This much more complex, experimentally well-posed, two-layer convection system promises to be an interesting new arena for the quantitative study of spatially extended patterns [3].

There have been several previous experiments on two-layer convection in restricted geometries [16], but to our knowledge, only one previous experiment has been published on convection in a laterally extended system [14]. It was restricted to locating the onset of instability, without visualization. More recent experiments have revealed new effects, including instabilities on heating from above and oscillatory convection at onset [17]. On the theoretical side, the two-layer problem has been the subject of a complete linear stability analysis [13–15] and several weakly nonlinear analyses [13,18,19]. The general problem is well within the range of modern weakly nonlinear theory and of numerical simulations.

In our experiment the Marangoni forces were comparable to the buoyancy forces at the onset of convection. The dimensionless heat flux across the interface is $\sim 40\times$ larger than in previous studies. This is expected to have important effects in the nonlinear regime [18,20]. We used shadowgraph imaging to visualize the pattern, measured its mean amplitude and wave number using Fourier techniques, and studied its secondary instabilities. The first pattern found above onset had a square planform at $\epsilon = 0.05$. Here $\epsilon = (\Delta T/\Delta T_c) - 1$, where ΔT is the temperature difference across *both* layers and ΔT_c is its value at onset. The mean wave number of this pattern is in good agreement with the critical value predicted from linear theory [15]. We did not resolve any hysteresis at onset. The square planform persisted up to $\epsilon \sim 0.7$, where it underwent a transition to a roll pattern. Up to the maximum value reached ($\epsilon \sim 1.4$), the scale of the convection cells was determined by the depth of the lower liquid. We compare our results with recent nonlinear theory [19] and find qualitative agreement.

The lower fluid was FC-75, a low-viscosity, perfluorinated hydrocarbon [21], while the upper was water. An important parameter is the depth fraction, $L = d^+/d$, where d^+ (d) is the depth of the upper (lower) fluid. In this paper, we report results for $L = 2.18 \pm 0.04$. The dimensionless heat flux across the interface, which is uniform below onset, is given by the Biot number $\mathcal{B} = \Lambda^+/\Lambda L$, where Λ^+ (Λ) is the thermal conductivity of the upper (lower) fluid. In contrast to previous liquid/gas experiments [6–11] for which $\mathcal{B} \sim 0.1$, in our experiment $\Lambda^+ > \Lambda$ and $\mathcal{B} = 4.31 \pm 0.08$, so that most of the total temperature drop ΔT falls across the thinner lower layer.

Water, at a constant temperature T_0, bathed the top surface of a sapphire window which formed the upper

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boundary of the cell [22]. The baseplate was maintained at a temperature $T_1 > T_0$ using a thin-film electric heater attached to its bottom surface. Both temperatures were controlled to ± 1 mK. The experimental control parameter is $\Delta T = T_1 - T_0$. The cell's plastic sidewall included a radial fin [23] to pin the two-fluid contact line. The round cell had thickness $D = d^+ + d = 4.06 \pm 0.01$ mm and radius $r = 41.15 \pm 0.05$ mm. The relevant aspect ratio is that of the lower fluid, $\Gamma = r/d = 32.3$. The uniformity of D was ± 8 μm, checked interferometrically. The top surface of the window was leveled to ± 100 μrad, using an electronic bubble level.

Both fluids wet all of the surfaces, so configurations with a single fluid bridging the gap between the top and bottom plates were difficult to avoid. To prepare the two-layer configuration, the horizontal cell was filled with FC-75 which was then displaced by water introduced above the fin. The interface was made level with the fin by visually eliminating the meniscus. The resulting layering was slightly imperfect due to the pinning of small deformations of the contact line and because the interface was difficult to see. The filling fraction L was determined destructively at the end of the experiment by tipping the cell on edge and measuring the interface position.

The onset of convection and the nonlinear patterns were visualized by the shadowgraph method [22], using a beam of light reflected off the mirrored bottom plate of the cell. The shadowgraph signal therefore contained contrast due to temperature gradients in both fluids, plus a component due to deflections at the deformed interface. The latter effect is small since the fluids are nearly index matched. We observed that the scale of the pattern in the upper layer was slaved to that of the lower. Thus, it was not necessary to disentangle the various contributions to the images in order to understand the basic planforms.

A typical run consisted of slowly increasing and decreasing ΔT while recording images. The onset of convection occurred at $\Delta T_c = 0.999 \pm 0.025$ °C. Convection entered from one side as a disordered pattern and eventually filled the cell. We did not observe hysteresis in the onset. Nonuniformities in d, linked to cell leveling and contact line deformations, caused a slight rounding of the bifurcations. The first patterns that emerged at $\epsilon = 0.05$ had a square planform. Square patterns have been observed previously in Marangoni convection [9], but only at much higher ϵ as a secondary bifurcation from a hexagonal pattern. Squares at onset are found in binary fluids [24] and in rotating convection [25]. Our observations are consistent with a square planform emerging right at onset, but, due to rounding effects, we cannot exclude the possibility that the more usual hexagonal pattern exists within the narrow range $0 < \epsilon < 0.05$.

Typical patterns are shown in Fig. 1. For increasing ϵ, a patchy, time-dependent square planform, as shown in Fig. 1(a), was found between onset and $\epsilon_{SR} = 0.67 \pm 0.05$, where a transition to rolls was observed. This transition could easily be located by a significant increase in the amplitude of the shadowgraph signal shown in Fig. 2. Near the transition, we found a dynamical coexistence of squares and rolls [Fig. 1(b)]. The rolls appeared where squares merged along shared edges, and vice versa. For $\epsilon > \epsilon_{SR}$, the pattern was dominated by rolls with squares appearing only at grain boundaries [Fig. 1(c)]. The more ordered roll patterns were very slowly time dependent. The finned sidewalls had only weak orienting effects on the rolls. As ϵ was decreased, an ordered square pattern re-emerged at $\epsilon_{RS} = 0.67 \pm 0.05$ [Fig. 1(d)]. In all cases, the patterns had a scale $\sim d$, the dimension of the thinner lower layer.

The characteristic time scale for convection in the lower fluid is the vertical thermal diffusion time $\tau_v = d^2/\kappa = 47$ s, where κ is the thermal diffusivity. We ramped ΔT slowly compared to τ_v, but it was impractical to increase it more slowly than the much-longer horizontal diffusion time $\tau_h = \Gamma^2 \tau_v = 14$ h. Thus, some of the disorder we observe on increasing ϵ may be due to the finite ramp rate. However, we also performed experiments in which ϵ was ramped quickly to a value $< \epsilon_{SR}$ and then held constant for $\sim 6\tau_h$. The resulting square pattern did not anneal significantly and remained time dependent. Thus, the disorder may be partially dynamical in origin.

We determined the wave number of the patterns using Fourier analysis. We found the azimuthally averaged structure function of a 256^2 pixel region of the center of the cell. The peak in the structure function gave the mean wave number $\langle k \rangle$. Figure 3 compares $\langle k \rangle$ with the results of linear stability theory [15]. The dimensionless wave number k is scaled by the depth of the bottom fluid d. The value of $\langle k \rangle$ at small ϵ agrees with the critical value $k_c = 2.38$ from the linear theory.

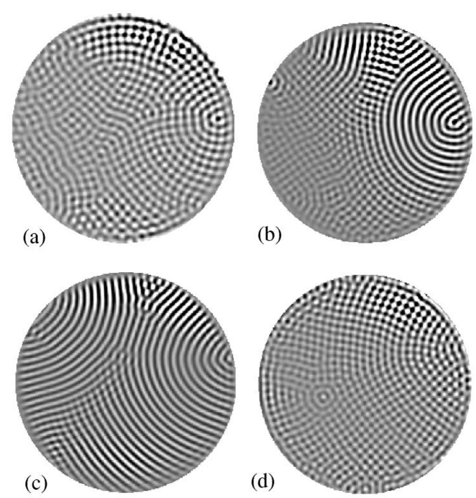

FIG. 1. Shadowgraph images of the patterns observed. (a) A disordered square planform at $\epsilon = 0.48$. (b) Coexistence near the transition from squares to rolls at $\epsilon = 0.78$. (c) Roll planform at $\epsilon = 1.38$. (d) A more ordered square pattern obtained from (c) upon slowly decreasing ϵ to 0.50.

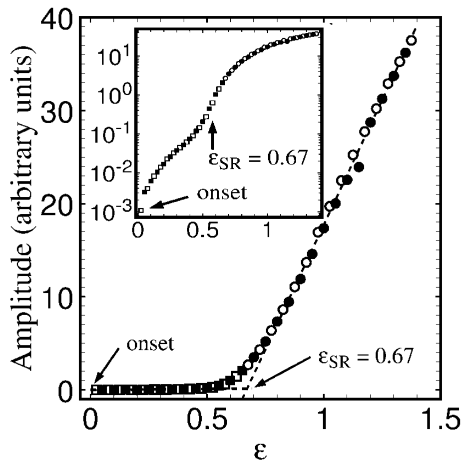

FIG. 2. The mean convective amplitude vs the dimensionless control parameter, showing the secondary bifurcation to rolls at $\epsilon_{SR} = \epsilon_{RS} = 0.67$ (inset on a log scale). The dashed lines are linear fits from which $\epsilon_{SR}/\epsilon_{RS}$ was determined. In all figures, square (circular) symbols indicate the square (roll) planform was observed, and open (solid) symbols indicate increasing (decreasing) ϵ.

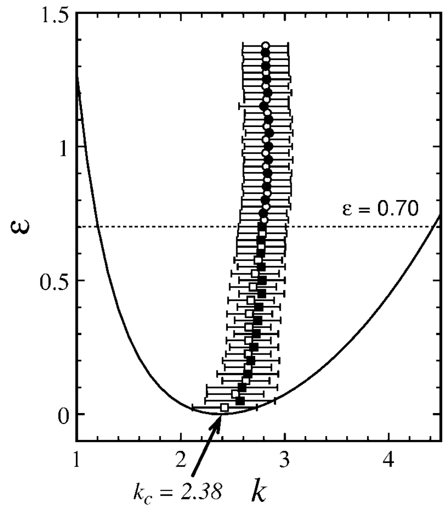

FIG. 3. The mean wave number $\langle k \rangle$, showing the linear stability boundary from Ref. [15] (solid line). Bars indicate the width of the azimuthally averaged structure function. We find a square planform for $\epsilon < 0.7$, and rolls otherwise.

To evaluate the linear stability boundary shown in Fig. 3, all of the material parameters are needed. These are, for the lower fluid, the kinematic viscosity ν, the density ρ, and $\kappa = \Lambda/\rho C_P$, where C_P is the specific heat, as well as the corresponding quantities for the upper fluid and the interfacial surface tension σ. All of these are assumed to be independent of T, according to the usual Boussinesq approximation, except σ and the two densities, which are linearly temperature dependent. Thus, we also require the thermal expansion coefficient $\beta = (1/\rho)\partial\rho/\partial T$ for each fluid and the temperature derivative of the interfacial surface tension $\gamma = -\partial\sigma/\partial T$. The negative sign is included so that $\gamma > 0$. All of the parameters are known, except σ and γ. Using Antonow's rule [26], we estimated σ to be equal to the difference of the surface tensions measured against air. The neutral stability boundary is very insensitive to σ, which enters only into the small surface deformations. On the other hand, the remaining parameter γ is crucial to the Marangoni mechanism. We fixed γ by requiring that the linear theory reproduce the correct measured ΔT_c. The result is $\gamma = 0.047 \pm 0.003$ dynes/cm K. This is consistent within a factor of 2 with Antonow's rule [26] applied to the known values of γ measured against air.

One- and two-layer theories of Bénard-Marangoni convection traditionally scale the problem using the temperature difference across the *lower* fluid $\Delta T^L = (1 + 1/\mathcal{B})^{-1}\Delta T$. This scaling is only approximate above onset, where convection increases the heat transport of each layer and produces temperature variations along the interface [13,27]. The Rayleigh and Marangoni numbers $\mathcal{R}$ and $\mathcal{M}$ of the lower layer are

$$\mathcal{R} = \left[\frac{g\beta d^3}{\kappa\nu}\right]\Delta T^L \qquad \mathcal{M} = \left[\frac{\gamma d}{\rho\kappa\nu}\right]\Delta T^L, \quad (1)$$

where g is the acceleration due to gravity. $\mathcal{R}$ and $\mathcal{M}$ are not independent, so it is more informative to define two related parameters [28],

$$\alpha = \left[1 + \frac{M}{M_0}\,\frac{R_0}{R}\right]^{-1} \qquad \lambda = \frac{R}{R_0} + \frac{M}{M_0}. \quad (2)$$

Here, $\mathcal{R}_0$ ($\mathcal{M}_0$) is the critical value of $\mathcal{R}$ ($\mathcal{M}$) in the absence of Marangoni (buoyancy) forces. In general, in the two-layer problem $\mathcal{R}_0$ and $\mathcal{M}_0$ will differ from the usual critical values for any one-layer model of the lower fluid. They can be determined from the full two-layer linear theory as described below. Fixed by the choice of fluids and depths, α is the fraction of the total forcing due to buoyancy, while λ is proportional to ΔT.

Figure 4 shows a plot of $\mathcal{M}$ vs $\mathcal{R}$. The linear stability boundary at constant L separates the conduction regime near the origin from the convecting regime. Its intercepts are the critical values $\mathcal{R}_0$ and $\mathcal{M}_0$. This boundary is calculated by holding L and the material parameters constant while varying d. α ranges from 0 on the $\mathcal{M}$ axis to 1 on the $\mathcal{R}$ axis. Above the line $\alpha = 1/2$, Marangoni forces dominate; below, buoyancy. For our experimental conditions $\alpha = 0.58$, so that the two effects are nearly equal. Increasing $\lambda \propto \epsilon$ corresponds to moving outward along this radial line. The symbols show the values of $\mathcal{M}$ and $\mathcal{R}$ obtained, and the secondary bifurcation point.

A weakly nonlinear theory of the full two-liquid problem has been completed recently by Engel and Swift [19]. They derived a set of amplitude equations, describing the stability of hexagonal, square, and roll planforms, from

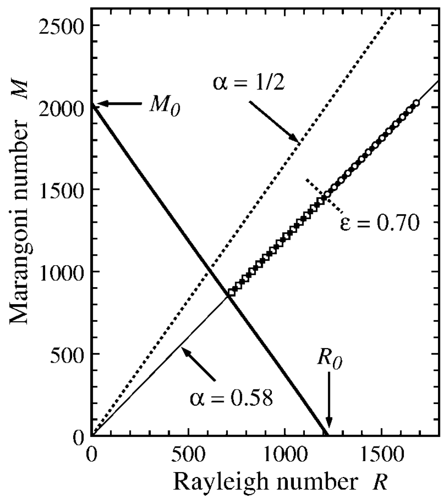

FIG. 4. The Marangoni number $\mathcal{M}$ vs the Rayleigh number $\mathcal{R}$ for the lower fluid, showing the neutral stability boundary (thick solid line) from the two-layer analysis of Ref. [15].

the complete hydrodynamic equations for both layers, including buoyancy and surface tension effects, but neglecting surface deformations. This theory generalizes previous results of Golovin *et al.* [18] and Regnier *et al.* [20]. For our experimental conditions, Engel and Swift obtained a very weakly subcritical bifurcation to hexagons at onset, with a hysteresis in ϵ of $\sim$0.01, and a secondary bifurcation to squares at $\epsilon_{hs} = 0.180$ [19]. They did not find a further bifurcation to rolls. Their results are thus only in qualitative agreement with our observations. The square planform we observed below ϵ_{hs} could be the result of inhomogeneous nucleation [29] occurring at boundaries or defects. Their amplitude equation model treated only unbounded patterns and omitted gradient terms that are important near defects. As well, a perturbative model such as theirs applies only in the limit of small ϵ and apparently fails to capture the secondary transition to rolls we observed at $\epsilon \sim 1$.

In summary, we have performed experiments on thermal convection in a system of two-layered liquids under conditions where buoyancy and Marangoni forces are comparable. We found a square planform of convection just above onset, which underwent a transition to a roll pattern at higher control parameter. We compared the pattern wave number to the linear stability boundary for the full two-layer coupled problem and found quantitative agreement between the wave number just above onset and the critical wave number. Recent weakly nonlinear theory, as applied to our experiment, is in qualitative agreement with the square patterns found near onset, but not the observed transition to rolls.

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