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Roll, square, and cross-roll convection in ferrofluids

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Abstract

Using the Galerkin technique we calculated different kinds of convection patterns in the Rayleigh–Bénard system and investigated their stability for very small Lewis numbers and very high separation ratios as typical for colloidal systems like ferrofluids.

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The Rayleigh–Bénard system [1] is a standard setup to study pattern formation outside of equilibrium. It consists of a fluid layer confined by two parallel plates perpendicular to the direction of gravity that are kept at a constant temperature. When the temperature difference between lower and upper plate is positive and large enough, buoyancy forces lead to the destabilization of the quiescent state and a convective pattern appears in the fluid layer.

For a pure fluid like water there are two relevant parameters that describe the system, namely the dimensionless temperature difference given by the Rayleigh number R and the ratio between momentum and temperature diffusion constants given by the Prandtl number σ. When the conducting state becomes unstable, the first stable convection pattern takes the form of rolls, i.e., two-dimensional structures with roughly circular streamlines in alternating directions. Fig. 1A shows a schematic top view.

Studying convection in a ferrofluid instead of a simple fluid is of interest since it allows the use of a magnetic field as a second control mechanism next to the temperature difference [2]. But even without magnetic field, the stability and bifurcation behavior of a ferrofluid is different from that of a simple fluid: as colloidal fluid it has to be described as a binary mixture of the carrier fluid and the magnetic particles [3].

The Rayleigh–Bénard setup with molecular binary mixtures like e.g. ethanol–water has already been studied extensively [4]. A binary mixture behaves differently from a pure fluid whenever concentration variations are present inside the layer. This is for example the case when temperature gradients generate concentration currents via the Soret effect. The Soret effect is quantified by the separation ratio ψ. A positive ψ means that the lighter component of the fluid is driven into the direction of higher temperature which enhances the thermal density variations and further destabilizes the fluid layer heated from below. A fourth relevant parameter is the Lewis number L giving the ratio between concentration and thermal diffusion.

Ferrofluids have a very small Lewis number of about 10^{-3}–10^{-4} that is up to two orders of magnitude lower than for ordinary liquid mixtures. The Soret effect, on the other hand, is orders of magnitude larger than for molecular mixtures: $|\psi| = 10-100$, where the Soret effect seems to be positive (negative) for sterically (ionically) stabilized ferrofluids.

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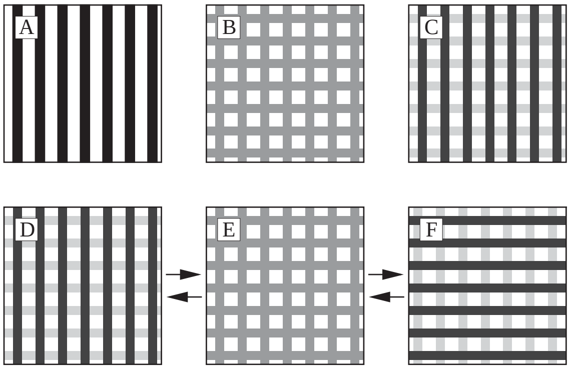

Fig. 1. Schematic top view of convection patterns of rolls (A), squares (B), cross-rolls (C), and oscillating cross-rolls (D,E,F).

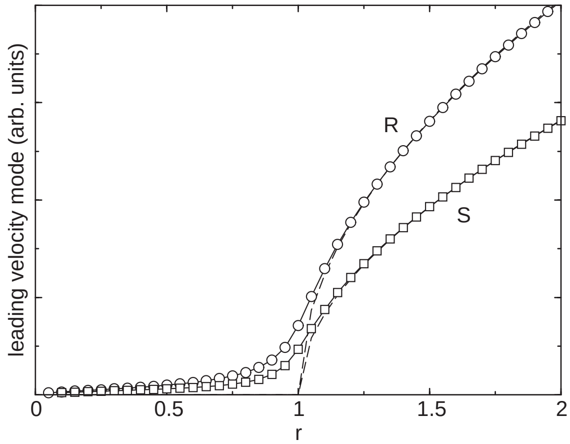

Fig. 2. Amplitude of the leading velocity mode versus reduced Rayleigh number r for rolls (circles) and squares (squares) for $\psi = 10$ and $L = 2 \times 10^{-4}$. The dashed curves refer to pure fluids.

Four kinds of convection structures that exist for positive ψ will be discussed here, namely rolls (R), squares (S), stationary cross-rolls (CR), and oscillatory cross-rolls (O-CR). All these structures are periodic in two directions x and y perpendicular to the direction of gravity z. Rolls as 2D structures are independent of y. These four different structures are solutions of the nonlinear equations of motion for the velocity field $\mathbf{u}$, the temperature field T, and the concentration field C.

We used these basic equations in a dimensionless form given in Refs. [4,5]. To evaluate the fixed-point and time-dependent solutions and to perform the stability analysis for these solutions a Galerkin method was applied. A detailed explanation can be found in Refs. [4,5]. We solved the equations for laterally periodic convection structures with periodicity length of about twice the layer thickness. To be precise, we always fixed the wave number to $k = 3.117$. That is the wave number of the first unstable mode in pure fluids and approximately the wave number observed in experiments in mixtures for a wide range of parameters. The Prandtl number was fixed to $\sigma = 10$, a typical value for liquid mixtures. As long as $\sigma \gg 1$ the solutions are not very sensitive to this parameter.

Calculating the solutions and investigating their stability for the very low L and very high ψ values that are typical for ferrofluids requires much numerical effort: these parameter combinations favor the assembly of very small boundary layers in the concentration field requiring very large sets of modes to resolve them. For the 2D roll solutions we used up to 40 modes in each spatial direction; for all 3D structures we used up to 24 modes. For details concerning the selected modes we again refer to Refs. [4,5].

For positive ψ, rolls and squares always exist as forward bifurcating solutions above the onset of convection. The squares can approximately be thought as superpositions of two perpendicular sets of convection rolls with an equal velocity amplitude (see. Fig. 1B) that is smaller than that of the actual roll solution. This can be seen in Fig. 2 where the amplitudes of the leading modes as a function of the reduced Rayleigh number $r = R/R_c(\psi = 0)$ [$R_c\,(\psi = 0) = 1707.76$ being the critical Rayleigh number for the pure fluid] are compared for rolls and squares at $\psi = 10$ and $L = 2 \times 10^{-4}$. The convective solutions for the pure fluid existing at $r > 1$ are also shown in Fig. 2. Here, convection is strong enough to mix and homogenize the fluid almost perfectly except for very narrow concentration boundary layers near the plates. The amplitudes for pure fluids and mixtures are therefore semiquantitatively—and for these parameters even quantitatively—the same. For $r < 1$ convective solutions exist only in the mixture due to the destabilizing positive Soret effect but the amplitude is much smaller. The convective threshold for these extreme parameter combinations is practically zero. The transition between both regions becomes more distinct for smaller L but smoother for larger ψ. The bifurcation diagram looks therefore quite similar to that of common liquid mixtures with much smaller ψ but larger L.

It has been shown that in binary mixtures rolls are the primary stable form of convection for positive ψ only when L is large and ψ is small enough [6]. Otherwise, and especially for ferrofluid parameters, squares are the first stable pattern to appear. But at higher Rayleigh numbers where the binary mixture behaves qualitatively like a pure fluid, rolls will finally gain stability.

The transfer of stability from squares to rolls happens via an intermediate cross-roll pattern that resembles the square pattern but lacks the $x \leftrightarrow y$ symmetry of squares since the amplitudes of the two roll sets are different (Fig. 1C). By increasing the Rayleigh number, one of the amplitudes increases while the other one decreases until they end as a simple roll pattern at another bifurcation point.

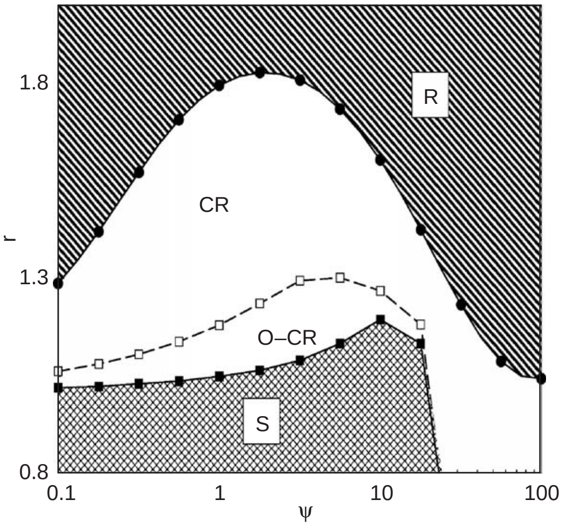

Fig. 3. Stability boundaries and bifurcation thresholds in the $r-\psi$ plane for $L = 0.002$. Rolls and squares are stable in the shaded domains marked by R and S, respectively. Filled circles: stability boundary of rolls against stationary cross-roll perturbations—here a CR solution bifurcates out of the R solution. Filled squares: stability boundary of squares against oscillatory cross-roll perturbations—here an O-CR solution bifurcates out of the S solution. Open squares: stability boundary of squares against stationary cross-roll perturbations—here a stationary CR solution bifurcates out of the S solution. The specific existence and stability ranges of CR and O-CR solutions have not been determined.

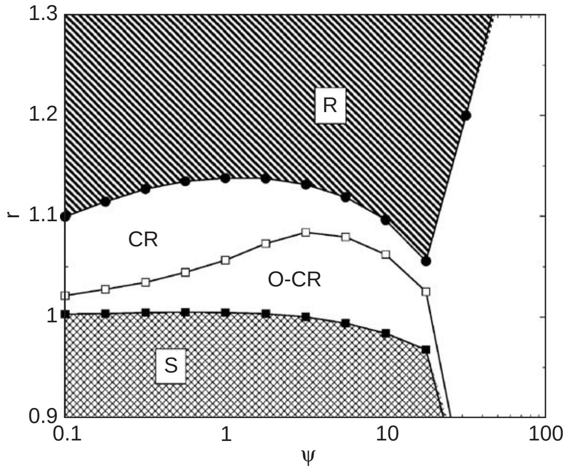

Fig. 4. Same as Fig. 3 but for $L = 0.0002$.

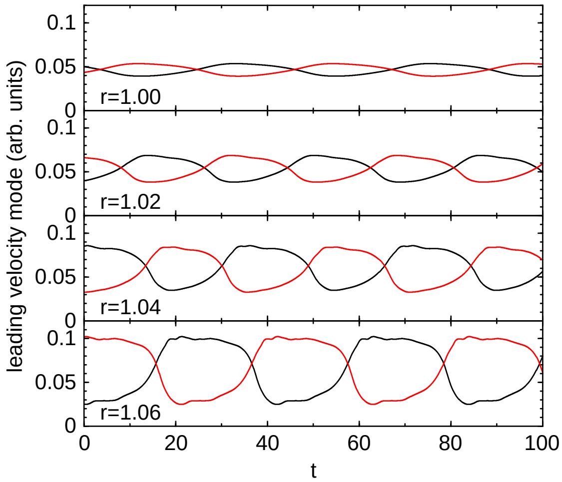

Fig. 5. The time-dependent leading velocity modes of oscillatory cross-rolls for $L = 0.0002$ and $\psi = 10$.

The range of r-values where these cross-roll patterns exist for two values of L is the interval between the open squares and the filled circles in Figs. 3 and 4. The apparent divergence of the stability boundary of squares—and for $L = 0.0002$ also for rolls—in the interesting range $\psi > 10$ may be an artifact of insufficient mode numbers, although in smaller models it happens at roughly the same parameter combination.

As can be seen from Figs. 3 and 4, the square structures lose their stability already at smaller r, before the bifurcation point that marks the appearance of the stationary cross-roll branch. In molecular mixtures, this earlier instability appears for Lewis numbers smaller than $L \approx 0.01$ and leads to a fourth kind of pattern, the time-dependent oscillatory cross-rolls. Here the amplitudes of the two roll sets slowly oscillate in counter phase, sweeping through a square state twice in a period such that the rolls in x or y direction alternate as dominant set. In the schematic picture in Fig. 1, the oscillatory cross-rolls undergo the cycle of states D → E → F → E → D periodically.

The oscillatory cross-rolls exist also for typical ferrofluid parameters. We simulated them through time integration of the mode equations starting from a slightly perturbed square solution. Fig. 5 shows the two leading velocity modes for this pattern over 100 diffusion times for $L = 0.0002$ and $\psi = 10$. Here the oscillatory instability takes place at about $r = 0.985$. With growing r the amplitude and frequency of the oscillation grow, and the oscillation becomes more and more anharmonic. Between $r = 1.02$ and 1.04 the time-averaged $x \leftrightarrow y$ symmetry is broken. One roll set now stays the dominant one for a longer time than the other. At $r = 1.06$, this symmetry is restored again but the frequency is now lower. At $r = 1.08$ (not shown), we found a chaotic behavior that may be transient. The evolution of the oscillatory cross-rolls with growing r is qualitatively different from what we found for larger L [5] and not fully understood yet.

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