Thermodiffusion effects in convection of ferrofluids

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Thermal convection in ferrofluids, the colloidal suspension of ferromagnetic particles, is investigated theoretically. Ferrofluids are treated as binary liquid mixtures with weak solutal diffusivity but large separation ratio. Due to the pronounced Soret effect of these materials in combination with a considerable solutal expansion, the resulting solutal buoyancy forces are dominant and the concentration dynamics cannot be disregarded for thermal convection. In principle, convective motion sets in at Rayleigh numbers well below the critical threshold for single-component liquids. But only far above this (hypothetical) threshold the growth dynamics of the amplitude is fast enough to be detectable and a nonlinear analysis demonstrates that there it quickly saturates in a state of stationary convective motion.

1. Introduction A typical property of binary mixture convection is the formation of concentration boundary layers [1]. This is a consequence of the fact that the concentration diffusivity D_c in mixtures is usually much smaller than the heat diffusivity κ. For molecular binary mixtures the dimensionless Lewis number $L = D_c/\kappa$ adopts typical values between 0.1 and 0.01 [2]. If colloidal suspensions are under consideration, the time scale separation is even more dramatic. In this context magneto-colloids, also known as ferrofluids, are a canonical example. These materials are dispersions of heavy solid ferromagnetic grains suspended in a carrier liquid [3]. With a typical diameter of 10 nm the particles are pretty large on molecular length scales, resulting in an extremely small particle mobility. This feature is reflected by Lewis numbers as small as $L = 10^{-4}$ [4]. The smallness of L leads to a situation where de-mixing effects (if any) take place on time scales far beyond any reasonable observation time. Thus, in most experiments ferrofluids can safely be treated as single-component fluid systems.

However, ferrofluids are also known to exhibit a very large separation ratio ψ. This observation is due to the pronounced thermo-diffusivity (Soret effect) of these materials in combination with the fact that the specific weights of the two constituents (magnetite and water/oil) are quite distinct. Following investigations of Blums et al. [4], who carried out experiments with a thermo-diffusion chamber, $|\psi|$ can adopt values up to about 100. Recent light scattering investigations of Bacri et al. [5], reveal ψ-values between around -200 (for ionic ferrofluids) and up to $+30$ (cyclohexane carrier) at a volume concentration of 10%. Meanwhile the Soret effect in ferrofluids has also been studied under the influence of an external magnetic field [6–8].

A fairly small number of papers deals with convection in ferrofluids. Most of them treat these liquids as single-component fluids, focusing on the extra drive associated with the temperature dependence of the magnetization (pyro-magnetic effect) [9–11]. Quite recently Shliomis and Souhar [12] studied the influence of the concentration field on thermal convection in ferrofluids without an external magnetic field. Using linear arguments they predicted a novel kind of relaxation-oscillation convection to appear at Rayleigh numbers below the single-component threshold Ra_c^0. Meanwhile, magnetic field related effects have also been investigated in this problem [13]. The purpose of the present work is to investigate in detail the role of the concentration field within a nonlinear treatment.

Provided no magnetic field is applied, thermal convection in a perfectly intermixed ferrofluid is usually believed [12] to behave as a single-fluid system. However, our investigation reveals that this is not correct. Rather it is the combination of both, the weak solutal diffusivity and the pronounced solutal buoyancy force, which renders the convective dynamics distinct from the pure fluid case. A Rayleigh-Bénard setup will already become unstable at Rayleigh numbers well below Ra_c^0, but perturbations will grow extremely slowly (i.e. on the creeping solutal diffusion time scale). On the other hand, near Ra_c^0 convective perturbations are found to grow and saturate into a stationary convective state on a much faster, experimentally relevant, time scale.

2. Setting up the problem Let us consider a laterally infinite horizontal layer of an incompressible ferrofluid (density ρ, kinematic viscosity ν) bounded by two rigid impermeable plates. The setup is heated from below with a temperature difference ΔT between the plates, i.e. along the z-direction. In the present paper we do not consider magnetic field related effects, thus the evolution equations for non-magnetic binary mixtures can be adopted. Taking $C(\boldsymbol{r}, t)$ as the concentration of the solid constituent of the suspension, the dimensionless equations for the Eulerian fields of velocity $\boldsymbol{v}(\mathbf{r}, t)$, temperature $T(\boldsymbol{r}, t)$, and $C(\boldsymbol{r}, t)$ read in Boussinesq approximation [14–16]

$$\begin{aligned} \boldsymbol{\nabla} \cdot \boldsymbol{v} &= 0, & (1)\\ \partial_t \boldsymbol{v} + (\boldsymbol{v} \cdot \boldsymbol{\nabla})\boldsymbol{v} &= -\boldsymbol{\nabla} W + Pr\, Ra \left[(T - \bar{T}) - \psi(C - \bar{C})\right] \hat{\boldsymbol{e}}_z + Pr\, \boldsymbol{\nabla}^2 \boldsymbol{v}, & (2)\\ \partial_t T + (\boldsymbol{v} \cdot \boldsymbol{\nabla})T &= \boldsymbol{\nabla}^2 T, & (3)\\ \partial_t C + (\boldsymbol{v} \cdot \boldsymbol{\nabla})C &= L(\boldsymbol{\nabla}^2 C + \boldsymbol{\nabla}^2 T). & (4) \end{aligned}$$

where gravity is directed along the negative z-axis ($-\hat{\boldsymbol{e}}_z$). Here we have scaled length by the layer thickness h, time by the characteristic heat diffusion time h^2/κ, temperature by ΔT, and the concentration by $(D_T/D_c)\Delta T$. The scale for the pressure W is $\kappa^2\rho/h^2$. Thereby κ, D_C, D_T are the coefficients for heat, concentration and thermo-diffusion, respectively. The quantities $\bar{T}$ and $\bar{C}$ are reference values defined as the mean values for temperature and concentration. Apart from the Prandtl number $Pr = \nu/\kappa$ and the Lewis number $L = D_c/\kappa$ there is a third dimensionless material parameter, the separation ratio $\psi = D_T\beta_c/(D_c\beta_T)$, where $\beta_T = -(1/\rho)\partial\rho/\partial T$ and $\beta_c = (1/\rho)\partial\rho/\partial c$ are the thermal and solutal expansion coefficient. The dimensionless Rayleigh number $Ra = \beta_T g h^3 \Delta T/(\kappa\nu)$ is the control parameter measuring the strength of the thermal drive. In Eq. (4) we have suppressed the Dufour-effect (heat current driven by a concentration gradient) as it is significant in gas mixtures, only.

The equations of motion are to be completed by boundary conditions: Taking no-slip conditions for the velocity and assuming the bounding plates to be highly heat conducting and impermeable for concentration currents, we have at the upper ($z = 1/2$) and the lower ($z = -1/2$) plates

$$\begin{aligned} \boldsymbol{v}|_{z=\pm 1/2} &= 0, & (5)\\ T|_{z=\pm 1/2} &= \bar{T} \mp \frac{1}{2}, & (6)\\ (\partial_z C + \partial_z T)|_{z=\pm 1/2} &= 0. & (7) \end{aligned}$$

Eq. (7) guarantees that a concentration current cannot penetrate the plates. Owing to the Soret effect the applied temperature difference enforces a finite concentration gradient at the boundaries. The dynamic equations (2-4) together with the boundary conditions (5-7) serve to determine $\boldsymbol{v}, T, C$, while Eq.(1) allows to eliminate the pressure W.

3. Nonlinear behavior We solve the nonlinear problem (1-7) by use of numerical methods. To that end we make the following ansatz of a 2-dimensional pattern, which is laterally (in x-direction) periodic with wave number k

$$
\begin{aligned}
C\left(x,z,t\right)-\bar{C} &= C_0(z,t)+c_1(z,t)\cos kx, &(8)\\
T\left(x,z,t\right)-\bar{T} &= -z+\theta_0\left(z,t\right)+\theta_1\left(z,t\right)\cos kx, &(9)\\
\boldsymbol{v}(x,z,t) &= -\hat{\boldsymbol{e}}_x(1/k)\partial_z w_1(z,t)\sin kx+\hat{\boldsymbol{e}}_z w_1(z,t)\cos kx. &(10)
\end{aligned}
$$

with incompressibility already built in. We adopt vertical profiles w_1, θ_0, θ_1, C_0, and c_1 in the form

$$
\begin{aligned}
w_1(z,t) &= A\left(t\right)\cos^2\left(\pi z\right), &(11)\\
\theta_1(z,t) &= B\left(t\right)\cos\pi z, &(12)\\
\theta_0(z,t) &= F\left(t\right)\sin 2\pi z, &(13)\\
C_0\left(z,t\right) &= z-\theta_0(z,t)+\sum_{n=0}^{n=N} a_n\left(t\right)\sin\left(2n+1\right)\pi z, &(14)\\
c_1\left(z,t\right) &= -\theta_1(z,t)+\sum_{n=0}^{n=N} b_n\left(t\right)\cos 2n\pi z, &(15)
\end{aligned}
$$

which satisfy the boundary conditions (5-7) identically. They describe two-dimensional convection in the form of parallel rolls along the y axis in an infinite slab of thickness 1. We point out that for $\psi = 0$, the concentration fields decouple from temperature and velocity. This reduces Eqs. (11-13) to the 3-mode model introduced by Lorenz [17] to mimic the dynamics of convective rolls in single-component Rayleigh-Bénard convection. At non-zero ψ, convection is modified by the concentration field, but we can adopt the above few-mode expansions for temperature and velocity without modifications, because the diffusivities for heat and momentum are large enough to prevent the appearance of strong gradients. By way of contrast, owing to the small Lewis number, the concentration field does build up steep boundary layers, which we account for by multi-mode Fourier series as given in (14,15). For C_0 the modes are antisymmetric in z, while for c_1 symmetric modes are appropriate. The number N of contributing modes are to be taken large enough to ensure that the results are insensitive against a further increase of N. For the parameter values considered here, $N = 20$ turned out to be sufficient. The equations for the mode amplitudes A, B, F, a_n, b_n have been solved by a Runge-Kutta integration. The wave number k, usually taken to be the mode of maximum linear growth rate $\lambda(k, Ra)$ varies between 3 and 3.5 within the investigated Rayleigh number regime. However, since the final predictions of our model do not depend sensitively on the k-value chosen, we have adopted $k = \pi$ in our simulations. All runs were started form an initial configuration characterized by a undisturbed linear temperature profile $T = \bar{T} - z$, a uniform concentration distribution $\partial_z C_0 = c_1 = 0$, and small random velocity fluctuations. In all of our runs the convective motion was found to settle in a state of *stationary* convection. A relaxation oscillation behavior as predicted in Ref. [12] could not be observed. The times necessary to reach the saturation values are several thermal diffusion times and increase with decreasing ε. However, they are still much shorter than the evolution time of the creeping concentration profile.

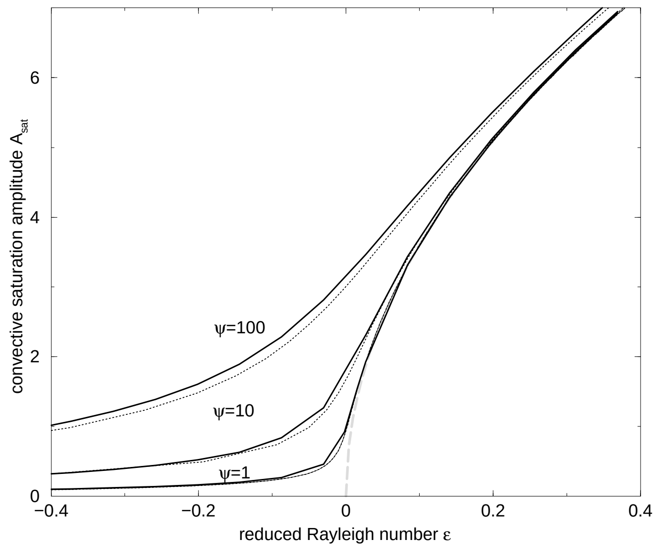

Figure 1: The saturation amplitude $A_{sat} = A(t \to \infty)$ as a function of $\varepsilon = Ra/Ra_c^0 - 1$ for $Pr = 7$ and $L = 7 \times 10^{-5}$. Ra_c^0 is the critical Rayleigh number for a single-component fluid ($\psi = 0$, dashed gray line). Dotted lines see text.

Fig. 1 shows the corresponding bifurcation diagram with the dependence of the saturation amplitude on the reduced Rayleigh number. At $\varepsilon > 0$ the amplitude saturates at a value, which does not significantly deviate from the single-component case. On the other hand, the influence of the concentration field is most pronounced for $Ra \leq Ra_c^0$. This is a consequence of the competitive interaction between the small Lewis number and the large separation ratio. Decreasing L makes the curve in Fig. 1 approaching the dashed reference line, whereas rising ψ has the opposite effect, since it amplifies the solutal buoyancy forces. For the sake of comparison the dotted lines in Fig. 1 show an analytical approximation for the saturated velocity amplitude based on a seven mode Galerkin approximation recently introduced by Hollinger et al. (Eq.(4.1b) in Ref. [18]).

When measuring a bifurcation diagram such as Fig. 1, one might conclude that the bifurcation is imperfect. Indeed, such a behavior was clearly observed in the experiments of Schwab et al. [11], who recorded the convective heat transport as a function of Ra. But we learn here that this phenomenon is to be attributed to the concentration dynamics: the very onset for convection is located at a much smaller Rayleigh number, $Ra_c = Ra_c^0 (1 + \psi)^{-1}$ [19], but at Rayleigh numbers slightly larger than Ra_c the linear growth rate of any disturbance remains extremely small. Thus, trying to detect Ra_c in such an experiment would be hopeless as it requires extremely long observation times.

In contrast, at $-0.2 < \varepsilon < 0.2$ the time necessary to wait for the equilibration of the nonlinear convective state amounts to only a few *thermal* diffusion times [19] for $\psi \gg 1$. This statement, which holds in particular also for the concentration field, demonstrates that the growth of convective perturbations is a fast process on the (creeping) time scale $1/L$ of solutal diffusion.

4. Conclusion Thermo-convection of binary mixtures with a weak concentration diffusivity and a large separation number has been investigated theoretically. By considering the classical Rayleigh Bénard setup it is shown that both the linear as well as the nonlinear convective behavior is significantly altered by the concentration field as compared to single-component systems. Starting from an initial motionless configuration with a uniform concentration distribution, convective perturbations are found to grow even at Rayleigh numbers well below the threshold Ra_c^0 of pure-fluid convection. It turned out that the actual critical Rayleigh number Ra_c is drastically smaller, but experimentally inaccessible due to the extremely slow growth of convection patterns for $Ra \gtrsim Ra_c$, requiring extremely large observation times. On the other hand, operating the ferrofluid convection experiment at Rayleigh numbers $Ra_c < Ra \lesssim Ra_c^0$, reveals considerable positive growth rates, which lead to a saturated nonlinear state almost as fast as pure-fluid convection does at $Ra > Ra_c^0$. This result is corroborated by earlier convection experiments. It does not comply with a recent prediction of convective self-oscillations conjectured from the interplay between short thermal and slow solutal diffusion time scales.

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