Scaling Behavior for the Onset of Convection in a Colloidal Suspension

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We investigate the early stages of mass convection in a colloidal suspension at high solutal Rayleigh number $\widetilde{\mathrm{Ra}}_s$. From the time evolution of shadowgraph images and by assuming a diffusive growth of the boundary layers we obtain an indirect measurement of the concentration boundary layer thickness δ^* at the onset of convection. We show that the dimensionless boundary layer thickness $\delta = \delta^*/d$ scales as Ra_s^{-p}, where $\mathrm{Ra}_s = \widetilde{\mathrm{Ra}}_s \delta$ is a modified solutal Rayleigh number for convection which accounts for the actual density unbalance and d is the thickness of the sample layer. This scaling behavior is analogous to that reported at steady state for turbulent convection in simple fluids. We find $p = 0.35$, a value compatible with the exponent $1/3$, reported for turbulent heat convection in simple fluids at steady state.

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The heat transfer in fluids at high Rayleigh numbers is a widely debated problem whose basic features are still unclear [1–4]. At large Rayleigh numbers the transferred heat scales algebraically with the applied temperature difference ΔT. The dimensionless Rayleigh number $\mathrm{Ra} = \alpha g \Delta T d^3/\kappa\nu$ is the ratio of the temperature-gradient-generated buoyancy forces and viscous forces (here g is the gravity acceleration, d is the sample thickness, α is the thermal expansion coefficient of the fluid, ν its kinematic viscosity, and κ its thermal diffusivity). The heat transferred is expressed by the dimensionless Nusselt number Nu, which is the ratio between the actual heat flow to the flow generated by heat conduction alone under the same temperature difference.

At high Rayleigh numbers the convective flow becomes turbulent and scaling applies: $\mathrm{Nu} \sim \mathrm{Ra}^p$. The value of the scaling exponent p is still questioned. Reported values of p range from $2/7$ up to $1/3$ (see for example [4] and references therein). The imposition of a large temperature difference creates two thermal boundary layers (BLs) near the thermalizing plates, where the temperature is strongly nonuniform. The heat transport properties in a simple fluid at high Ra are completely determined by the ratio δ between the BL thickness δ^* and the sample height d, as $\mathrm{Nu} \simeq 1/\delta$. Up to now measurements of scaling have been performed under steady state conditions, when the planform of the instability has been fully established.

In 1966 Howard, in a seminal paper on convection at high Rayleigh numbers, described a *gedanken experiment* to investigate the scaling of Nu at the onset of convection [5]. He assumed that, after the imposition of a temperature difference at $t = 0$, the initial stages are dominated by the growth of the BL by means of heat conduction. The determination of the conductive time t^* needed to trigger convection provided an estimate of δ, which in turn provided an estimate of Nu.

The actual execution of this experiment is rather cumbersome, as it is very difficult to fulfill the ideal condition where a temperature difference is imposed instantaneously. Near critical fluids represent an exception to this, because the thermal diffusivity becomes small as the critical point is approached. Close to the critical point, however, the fluid becomes very compressible, and the time scale for the onset of the instability is strongly influenced by the piston effect [6,7].

In this work we have investigated the convective mass transfer at high solutal Rayleigh numbers. By exploiting the small diffusion coefficient of complex fluids we have performed Howard's experiment in a colloidal suspension to investigate the convective destabilization of concentration BLs. Here the imposition of a temperature difference induces a uniform mass flow due to the Soret effect [8]. The formal analogy between heat convection in the classical Rayleigh-Bénard instability and mass convection in a Soret driven instability is dealt with in detail in [9,10]. The mass flow promotes the diffusive growth of concentration BLs, mass being accumulated at one boundary and depleted from the other one. Because of the small diffusion coefficient of the colloidal suspension used, the time scale for the diffusive growth of the BLs is much larger than that needed to establish the conductive temperature profile across the sample. In this way, the initial phase where the temperature profile is created almost instantaneously is followed by a slower phase where the concentration BLs grow diffusively. Therefore, by using a quantitative shadowgraph technique we are able to obtain a reliable determination of the concentration BL thickness δ^* at the onset of convection from the diffusive time t^* needed to trigger the instability.

The determination of δ^* provides an indirect measurement of the solutal Nusselt number Nu_s associated with the mass transfer (also called Sherwood number) [11,12], which mirrors the Nusselt number defined for simple fluids. Nu_s is defined as the ratio between the mass transferred due to convection and the mass transferred due to diffusion in the absence of convection under the same concentration difference. Here also the mass transfer properties are completely determined by δ, and $\mathrm{Nu}_s \simeq 1/\delta$ [11].

We find that δ scales as $\delta \sim \mathrm{Ra}_s^{-0.35}$ in the solutal Rayleigh number range $3.8 \times 10^5 < \mathrm{Ra}_s < 5.8 \times 10^7$, where Ra_s is a modified solutal Rayleigh number which accounts for the fact that convection is induced by imposing a constant mass flow.

The scaling exponent is compatible with the value $1/3$ originally suggested by Howard for the onset of convection and with the exponent $1/3$ reported under steady state conditions for simple fluids [4] and for convection induced by electrochemical mass transfer [13].

The imposition of a temperature difference to a binary mixture induces a mass flow by means of the Soret effect [14]. The Soret mass flow is $J_S = -\rho D S_T c(1-c)\nabla T$, where ρ is the density of the mixture, D its mass diffusion coefficient, S_T is the phenomenological Soret coefficient and c is the weight fraction concentration of the denser component. The Soret coefficient can be either positive or negative, according to whether the denser component migrates towards the colder or hotter plate. The overall mass flow due to diffusion and to the Soret effect is $J = -\rho D \nabla c + J_S$. Thereby at steady state, in the absence of convection, a concentration gradient $\nabla c = -S_T c(1-c)\nabla T$ is generated within the mixture. The density gradient associated with ∇c is $\nabla \rho = \rho \beta \nabla c = -\rho \beta S_T c(1-c)\nabla T$, where $\beta = \rho^{-1}(\partial \rho / \partial c)_T$ is the solutal expansion coefficient. The mutual relevance of density variations generated by thermal dilation and by concentration is expressed by the separation ratio $\Psi = \beta S_T c(1-c)/\alpha$. The Rayleigh number for the Soret driven convection is the solutal Rayleigh number

$$\widehat{\mathrm{Ra}}_s = \frac{\beta g S_T c(1-c)\Delta T d^3}{\nu D} = \mathrm{Ra}\frac{\Psi}{L} \qquad (1)$$

where $L = D/\kappa$ is the Lewis number. The threshold condition for mass convection in the presence of no-slip impermeable boundaries is $\widehat{\mathrm{Ra}}_s > 720$ [15].

The solutal Rayleigh number $\widehat{\mathrm{Ra}}_s$ does not account for the actual density imbalance present in the sample. In general the choice of the appropriate Rayleigh number depends on the boundary conditions. In the case of a simple fluid, convection can be induced either by applying a constant temperature difference ΔT or by letting a constant heat flow through the fluid. The condition of constant ΔT becomes relevant in the presence of boundaries whose thermal conductivity is much higher than that of the layer of fluid. Under this condition the appropriate Rayleigh number is Ra, which is defined under a density unbalance $\Delta \rho = \rho \alpha \Delta T$. The condition of convection induced by applying a constant heat flow is relevant when the thermal conductivity of the thermalizing plates is smaller than that of the layer of fluid, as it becomes difficult to control the temperature difference at the boundaries of the fluid. In this case the Rayleigh number $\widehat{\mathrm{Ra}} = (\alpha g \nabla T|_{z=0,d} d^4)/\kappa\nu$ is proportional to the heat flux at the boundaries $J_b \propto \nabla T|_{z=0,d}$. The relation between the two Rayleigh numbers, recently derived by Otero *et al.* [16] is $\widehat{\mathrm{Ra}} = \mathrm{RaNu}$.

In the case of binary mixtures the boundaries are impermeable and convection is induced by a constant mass flow J_S, the concentration difference Δc across the sample being not determined a priori. By reexpressing the solutal Rayleigh number $\widehat{\mathrm{Ra}}_s$ in terms of the concentration difference at the boundaries $\Delta c \simeq S_T c(1-c)\nabla T \delta d$ we find $\widehat{\mathrm{Ra}}_s = \mathrm{Ra}_s/\delta$, which mirrors the relation obtained by Otero *et al.* for a simple fluid. Here $\mathrm{Ra}_s = \frac{\beta g \Delta c d^3}{\nu D}$ is a modified solutal Rayleigh number which accounts for the actual density unbalance $\Delta \rho = \rho \beta \Delta c$ present across the sample.

In this Letter we describe the Soret induced convection in a colloidal suspension of 22 nm diameter spherical silica particles (LUDOX® TMA) dispersed in water at a weight fraction concentration $c = 4.1\%$. This suspension is characterized by $\Psi = -3.41$ and $L = 1.48 \times 10^{-4}$ and consequently, as indicated by Eq. (1), large solutal Rayleigh numbers can be achieved.

The colloidal suspension is characterized by a negative Soret coefficient $S_T = -0.047\ \mathrm{K}^{-1}$. This feature allows convection to be induced by heating the fluid from above, a condition which prevents the onset of the Rayleigh-Bénard instability. Other physical properties are $D = 2.2 \times 10^{-7}\ \mathrm{cm}^2/\mathrm{s}$, $\kappa = 1.48 \times 10^{-3}\ \mathrm{cm}^2/\mathrm{s}$, $\alpha = 3.03 \times 10^{-3}\ \mathrm{K}^{-1}$, $\nu = 8.18 \times 10^{-3}\ \mathrm{cm}^2/\mathrm{s}$, and $\beta = 0.57$.

The experimental investigation of the onset of the instability requires the fast buildup of a large temperature difference across the sample and a good optical accessibility. Measurements were performed with sample thicknesses of 1.0, 1.3, 2.0, 2.3, 2.9, and 4.5 mm. The rise time of the applied temperature difference is about 35 s to reach 64% of the set point. The thermal stability at steady state is of the order of 1 mK/24 h for the average temperature and of the order of 3 mK/h for the applied temperature difference. The horizontal spatial uniformity of the thermal gradient applied to the sample is of the order of 3% across the observation window.

Images of the convecting fluid are generated by means of a shadowgraph setup [17]. Shadowgraph is a heterodyne technique where the field diffracted from concentration modulations in the sample is recombined with the transmitted beam onto a charge coupled device sensor. Since the incoming beam is sent orthogonal to the sample, optical gradients along the vertical axis do not produce any signal. However, when convection sets in, horizontal gradients are generated because of the three dimensional convecting structures. The variance of an image of the sample $\langle \delta I^2 \rangle = \langle [I(x,y) - \langle I \rangle]^2 \rangle$ is proportional to the mean square average of the concentration differences generated by convection. $\langle \delta I^2 \rangle$ provides a quantitative estimate of the turbidity of the sample which would be very difficult by using other techniques, due to the small wave vector of the excitations involved. Moreover, our measurement procedure relies on the estimate of dynamical changes in $\langle \delta I^2 \rangle$, and this allows a very efficient rejection of stray light at small wave

vectors. A typical measurement sequence involves the sudden imposition of a steady temperature difference to the sample, followed by the acquisition of a sequence of images at a rate of one image every five seconds. The variance $\langle \delta I^2 \rangle$ of the intensity of each image is then processed to obtain time sequences like those shown in Fig. 1. The curves in Fig. 1 represent some of the different runs obtained by changing the applied temperature difference. The temperature differences range from 3 K up to 38 K and the solutal Rayleigh numbers from $\widehat{\mathrm{Ra}}_s = 1.7 \times 10^6$ up to 1.6×10^9. At steady state the structure of the planform is that of a spoke pattern at all the investigated Rayleigh numbers [17,18].

The curves in Fig. 1 show some common features. After the imposition of the temperature difference, $\langle \delta I^2 \rangle \simeq 0$ for a long time. During this period the concentration gradients develop uniformly close to the boundaries and the concentration profile depends only on the vertical coordinate. Accordingly, the BLs do not generate any signal. Then the amplitude of the signal suddenly grows up to a maximum. After the maximum the signal exhibits damped oscillations and eventually attains a steady value.

The most prominent feature of Fig. 1 is that convection sets in within a time frame many orders of magnitude smaller than the diffusive time $t_m = d^2/D$ necessary to create a concentration gradient across the cell thickness. This is due to the fact that the mechanism for the onset of convection is the rapid diffusive growth of the BLs, which become unstable in a time much shorter than t_m. Another interesting feature of Fig. 1 is that $\langle \delta I^2 \rangle$ exhibits damped relaxation oscillations. A similar behavior has been also reported during the transient of Soret driven convection in a water-ethanol mixture [18] and during the transient to heat convection in compressible fluids [7,19]. The origin of these oscillations is still debated and its discussion is beyond the aim of this work.

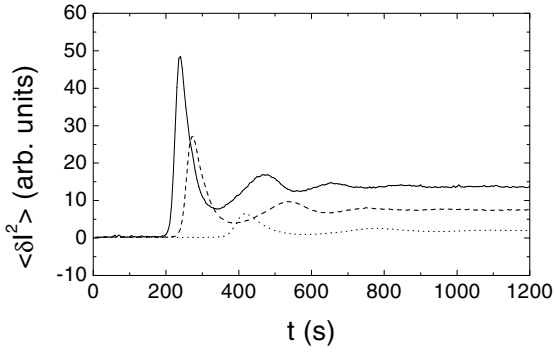

FIG. 1. Variance of shadow images sequences plotted as a function of time. The cell thickness is $d = 1.0$ mm. The applied temperature differences and solutal Rayleigh numbers correspond to $\Delta T = 34.8$ K and $\widehat{\mathrm{Ra}}_s = 1.84 \times 10^7$ (solid line), $\Delta T = 23.75$ K and $\widehat{\mathrm{Ra}}_s = 1.25 \times 10^7$ (dashed line), and $\Delta T = 9.6$ K and $\widehat{\mathrm{Ra}}_s = 5.08 \times 10^6$ (dotted line).

In this Letter we focus on the investigation of the very early stages of the instability where the BLs grow diffusively until they are destabilized by convection.

The analysis of the results in Fig. 1 involves the determination of the diffusive time $\tau^* = t^*/t_m$ needed to trigger the instability as a function of $\widehat{\mathrm{Ra}}_s$. The time τ^* corresponds to the point where the $\langle \delta I^2 \rangle$ signal in Fig. 1 starts to grow, due to the development of concentration differences generated by the convective flow. This indicates that the BLs have grown slightly above their steady state value and begin to develop bumps. The bumps grow due to the buoyancy force, thus giving rise to the destabilization of the BLs.

Figure 2 shows τ^* plotted as a function of $\widehat{\mathrm{Ra}}_s$. The continuous line represents the best power-law fit of the data, yielding the exponent $\alpha = 0.52 \pm 0.03$ [20].

By assuming that the initial stages of convection are dominated by the diffusive growth of the BLs, the concentration gradient at the boundaries after the imposition of the temperature difference is [21]

$$\nabla c(z, \tau) \simeq -S_T c(1 - c) \nabla T \left[1 - \mathrm{erf}\left(\frac{z/d}{2\sqrt{\tau}} \right) \right] \quad (2)$$

where the dimensionless time is $\tau = t/t_m$.

The characteristic length scale associated with this concentration gradient profile is the BL thickness and is given by [5]

$$\delta(\tau) = (\pi \tau)^{1/2}. \quad (3)$$

From Eq. (3) we are able to obtain an indirect estimate of δ from the latency time τ^*. The estimate of δ also allows us to derive the modified solutal Rayleigh number $\mathrm{Ra}_s = \widehat{\mathrm{Ra}}_s \delta$.

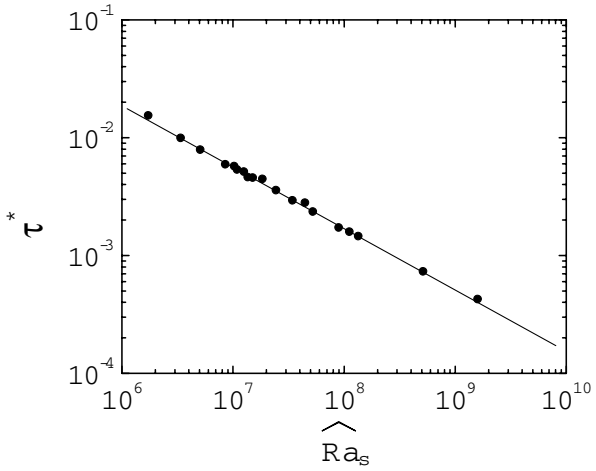

FIG. 2. Latency time τ^* for the onset of the instability plotted as a function of the solutal Rayleigh number $\widehat{\mathrm{Ra}}_s$. The solid line is the best fit of the experimental results yielding a power law $\tau^* = 25.3 \widehat{\mathrm{Ra}}_s^{-0.52}$.

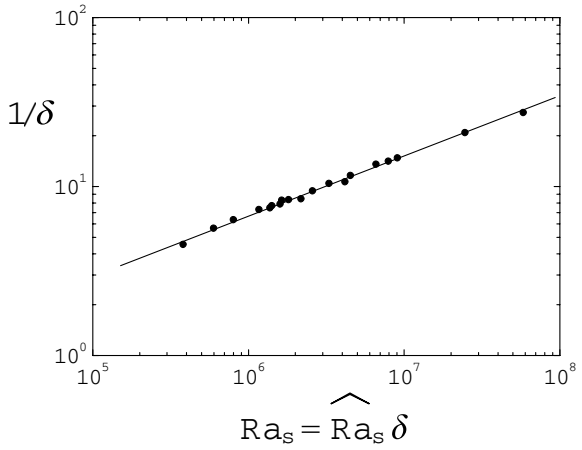

FIG. 3. Effective solutal Nusselt number $1/\delta$ plotted as a function of the modified solutal Rayleigh number Ra_s. The solid curve represents the best fit of the experimental results yielding a power law $1/\delta = 0.051\mathrm{Ra}_s^{0.35}$.

Figure 3 shows $1/\delta$ plotted as a function of Ra. The fit of the experimental data yields a scaling relation $1/\delta = (0.051 \pm 0.008)\mathrm{Ra}_s^{0.35\pm0.03}$, represented by the continuous line in Fig. 3. The power-law exponent is compatible with the value $1/3$ suggested in the original *gedanken experiment* by Howard.

The exponent 0.35 ± 0.03 is in good agreement with the exponent $1/3$ frequently reported for turbulent heat convection in simple fluids at steady state [4]. Our experiment was performed under extreme Schmidt number conditions ($\mathrm{Sc} = \nu/D = 37\,200$), which mirror the large Prandtl number ($\mathrm{Pr} = \nu/\kappa$) conditions in simple fluids. To our knowledge, the only investigation of scaling properties of the solutal Nusselt number Nu_s at very high Schmidt numbers has been performed by Goldstein *et al.* [13]. These authors used an electrochemical mass transfer technique to determine the asymptotic behavior of Nu_s at steady state in the solutal Rayleigh number range $10^8 < \mathrm{Ra}_s < 5 \times 10^{12}$ at a large Schmidt number, $\mathrm{Sc} = 2750$. They found the scaling relation $\mathrm{Nu}_s = 0.0659\mathrm{Ra}_s^{1/3}$. By recalling that, as suggested by Howard, $\mathrm{Nu}_s \approx 1/\delta$ we are in the position to compare this result with the scaling law determined by us at the onset. The exponents are compatible, both being in good quantitative agreement with the value $1/3$. The comparison of the prefactors is more difficult, as the Schmidt numbers used to perform the measurements differ by about a factor 14. However, by neglecting the dependence on Sc, the prefactors differ by about 30%. The estimate of Nu_s at the onset leads therefore to values bearing order of magnitude agreement with those estimated at steady state [22].

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