## **Onset of convection in colloids stratified by gravity**

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Settling of the equilibrium (barometric) distribution of colloidal grains in a liquid requires a very long time in comparison with that necessary for temperature equilibrium establishment. This mismatch enables different scenarios for the onset of thermal convection. If the stratified state has had no time to be settled, the colloid behaves as a pure fluid where only stationary convection arises. On the contrary, if the barometric concentration profile has been reached before the temperature gradient is imposed, only oscillatory convection occurs. Thereafter, the oscillations last as long as it takes the convection to wash away the initial concentration profile. Then the fluid becomes homogeneous and stationary convection is eventually established. The influence of the Soret effect upon the onset of convection is also taken into account and discussed.

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Since the middle of the 1980s convection in binary fluid mixtures has been the subject of extensive research. A variety of bifurcation phenomena and convective states with different structural and temporal properties have been displayed in many theoretical  $\begin{bmatrix}1-5\end{bmatrix}$  and experimental  $\begin{bmatrix}6-10\end{bmatrix}$  works. The experiments were carried out with true (molecular) solutions such as water-ethanol and water-methanol mixtures where the coupling between the fields of velocity, temperature, and concentration is provided by the Soret effect, which is the mechanism by means of which an externally imposed temperature gradient causes a certain concentration gradient.

Colloidal solutions significantly differ from molecular ones. The size of ions or/and molecules dissolved in true solutions never exceeds 1 nm while colloidal grains are approximately in the range from 10 to 100 nm. The grains are still too small to settle out of solution due to gravity but they are large enough to form a gravity induced concentration profile  $\phi(z)$ —a Boltzmann (barometric) particle distribution with a marked vertical gradient. Thus, the colloid proves to be stratified even in the absence of the Soret effect.

Another important distinction between colloidal and true solutions is also due to the difference in the sizes of the dispersed particles. According to the Einstein formula, the diffusion coefficient of particles is inversely proportional to their diameter *d*:

$$
D = k_B T / (3 \pi \eta d), \qquad (1)
$$

where  $\eta$  is the fluid viscosity. Hence, as colloidal grains are about 100 times the molecule size (we are here not dealing with polymer solutions), the characteristic diffusion time in colloids  $\tau_D \sim D^{-1}$  is about 100 times longer than that in true solutions. As a consequence, this mass-diffusion time appears to be 10 000 times longer than the temperature-

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diffusion time  $\tau$  during which an imposed temperature gradient settles (for true solutions the ratio  $\tau_D / \tau_T$  does not exceed 100).

The great difference between  $\tau_D$  and  $\tau_T$  enables different scenarios of convection onset in colloids. Actually, one can excite convection *before* any concentration stratification is settled, and then the colloid behaves as a pure (i.e., singlecomponent) liquid, where only stationary instability occurs. Otherwise, a supercritical temperature difference can be applied to the layer after the establishment of the barometric distribution. As will be shown below, in this case only oscillatory instability arises. The oscillatory convection washes out the initial concentration profile thereby eliminating the very reason for oscillations. Thus, in the long run, stationary convection sets in.

Let a suspension of grains of diameter *d* and density  $\rho_s$  in a carrier liquid of density  $\rho_f$  fill up a horizontal layer. The equilibrium concentration of grains in the gravity field obeys the barometric formula

$$
\phi(z) = \overline{\phi}e^{-z/l}, \quad l = k_B T/(\Delta \rho V g), \tag{2}
$$

where *l* stands for the *sedimentation length*,  $\overline{\phi}$  is the mean volume fraction of the grains,  $\Delta \rho = \rho_s - \rho_f$ , *V* is the particle volume, **g** is the gravity acceleration, and the *z* axis is directed upward. All estimates will be carried out for a waterbased suspension of silica  $(\rho_s = 2.255 \text{ g/cm}^3, d = 22 \text{ nm})$ used earlier in  $[11]$ . The data yield the spatial scale of sedimentation  $l \approx 60$  mm. For the layer of thickness  $h = 2$  mm the ratio *h*/*l* is sufficiently small to allow replacement of the barometric distribution (2) by the linear relationship  $\phi(z)$  $\approx \phi(1-z/l)$ , so the concentration gradient across the layer is almost constant:

$$
d\phi/dz \simeq -\,\overline{\phi}/l.\tag{3}
$$

For solutions of molecules with diameter  $d \leq 1$  nm one has  $l \sim 1$  km; thus the gradient (3) is negligible.]

In the case of  $h \ll l$  the characteristic mass-diffusion time

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is  $\tau_D \simeq h^2 / (\pi^2 D)$  [12]. Substituting from [11] the measured value  $D=2.2\times10^{-7}$  cm<sup>2</sup>/s and  $h=2$  mm gives  $\tau_D \approx 5$  h, while the temperature-diffusion time  $\tau_T \simeq h^2 / (\pi^2 \kappa)$  on substitution of the coefficient of temperature diffusivity  $\kappa$  $=1.48\times10^{-3}$  cm<sup>2</sup>/s hardly reaches 3 s. Thus, the equilibrium temperature gradient  $dT/dz = -\Delta T/h$  settles about 6000 times faster than the concentration gradient  $(3)$ .

The convection in colloids is described by the set of equations for the fluid velocity **v**, pressure *p*, temperature  $\theta$  $-\left(\frac{\Delta T}{h}\right)z$ , and concentration  $\varphi-\left(\frac{\overline{\phi}}{l}\right)z$ :

$$
d\mathbf{v}/dt = \nabla^2 \mathbf{v} + (\theta - \varphi)\mathbf{e} - \nabla p,\tag{4}
$$

$$
Pd\theta/dt = \nabla^2 \theta + R(\mathbf{v} \cdot \mathbf{e}),\tag{5}
$$

$$
P d\varphi/dt = L[\nabla^2 \varphi + r^{-1} \partial \varphi/\partial z] + B(\mathbf{v} \cdot \mathbf{e}), \tag{6}
$$

where  $d/dt = \partial/\partial t + P^{-1}(\mathbf{v} \cdot \nabla)$ ,  $r = l/h$ , and the unit vector **e** is aligned with the *z* axis. The dimensionless equations contain two material parameters, the Lewis number  $L = D/\kappa$  and the Prandtl number  $P = \nu / \kappa$  (here  $\nu$  is the kinematic viscosity), and two control parameters

$$
R = \frac{\alpha g \Delta T h^3}{\nu \kappa}, \quad B = \frac{\beta g \bar{\phi} h^4}{\nu \kappa l}, \tag{7}
$$

which are the Rayleigh number and what we call the Boltzmann (or barometric) number;  $\alpha = -\rho^{-1}(\partial \rho/\partial T)$  and  $\beta$  $=\rho^{-1}(\partial \rho/\partial \phi)$  are coefficients of thermal and solutal expansion. Units of length, time, velocity, temperature, and concentration are *h*,  $h^2/v$ ,  $\kappa/h$ ,  $v\kappa/\alpha gh^3$ , and  $v\kappa/\beta gh^3$ , respectively. In these units, the gradients of temperature and concentration  $(3)$  in the steady state look like

$$
dT/dz = -R, \quad d\phi/dz = -B. \tag{8}
$$

Let us perform a linear stability analysis of the steady state. Assuming the dependence of small perturbations on time and horizontal coordinates to be  $exp[i\omega t + i(k_x x + k_y y)]$ , we reduce the set of equations  $(4)$ – $(6)$  to

$$
(D2 - k2)(D2 - k2 - i\omega)v - k2(\theta - \varphi) = 0,
$$
 (9)

$$
(D2 - k2 - i\omega P)\theta + Rv = 0,
$$
\n(10)

$$
\underline{\mathcal{L}}(\underline{\mathcal{D}}^2 - k^2 + \underline{\mathcal{D}}/r)\varphi - i\omega P\varphi + Bv = 0, \qquad (11)
$$

where  $D = d/dz$ ,  $v = (\mathbf{v} \cdot \mathbf{e})$  is the vertical component of velocity, and  $k^2 = k_x^2 + k_y^2$ . The solutions of these equations should satisfy the boundary conditions on the confining rigid impervious planes  $z = \pm 1/2$ :

$$
v = Dv = \theta = D\varphi + \varphi/r = 0.
$$
 (12)

Some critical parameter values obtained from the numerical solution of the eigenproblem  $(9)$ – $(12)$  are presented in Table I. For calculations, we have taken the material constants of a suspension earlier used in [11]:  $L=0.00015$ , P=5.5, and *r*  $=30.$ 

As was shown above, the particle diffusion plays the decisive role in forming the equilibrium concentration profile. Nonetheless, dealing with the convection excitation, one can

TABLE I. Critical parameters for the Prandtl number  $P = 5.5$  and some values of the Boltzmann number B.

$\bar{\phi}$ (%) B			$R_c$ $k_c$ $\omega_c$	$\Delta T_c$ (K) $\qquad$ $T_c$ (s)	
			$0 \t 0 \t 1708 \t 3.117 \t 0 \t 8.7$		
1.5	399	2079 3.136 1.675		10.6	18.4
$\mathcal{R}$		783 2437 3.156 2.360		- 12.4	13.1

omit the underlined diffusion term in Eq.  $(11)$  because of the smallness of the Lewis number. The Lewis number represents none other than the ratio  $\tau_T / \tau_D$ , and its smallness illustrates the fact that the diffusion process does not manage to alter the particle distribution during the development of convection: it has simply no time to do that. The remaining part of Eq. (11),  $i\omega P\varphi=Bv$ , allows us to eliminate  $\varphi$  from Eq. (9), and thus the set of equations  $(9)$ – $(11)$  reduces to

$$
[i\omega P(D^2 - k^2)^2 + \omega^2 P(D^2 - k^2) + k^2 B]v - i\omega Pk^2 \theta = 0,
$$
  
 
$$
Rv + (D^2 - k^2 - i\omega P)\theta = 0.
$$
 (13)

For the case of so-called *free boundaries*, the equations admit an exact solution of the kind  $v \sim \theta \propto \cos \pi$ . This ansatz leads to a dispersion relation for the frequency of neutral (critical) oscillations  $\omega$ :

$$
i\omega P[\omega^2 P(\pi^2 + k^2) + k^2 (R - B) - (\pi^2 + k^2)^3]
$$
  
+ 
$$
\omega^2 P(P + 1)(\pi^2 + k^2)^2 = k^2 (\pi^2 + k^2)B.
$$
 (14)

It can be seen that the *stationary* instability  $(\omega=0)$  occurs only if  $B=0$ , i.e., when the concentration gradient (8) has had no time to be formed. Then Eq.  $(14)$  has the well-known solution  $R^{st}(k)$  with the minimum  $R_0$  at  $k = k_0$ :

$$
R^{st} = \frac{(\pi^2 + k^2)^3}{k^2}, \quad R_0 = \frac{27\pi^4}{4}, \quad k_0 = \frac{\pi}{\sqrt{2}}.
$$

Otherwise, i.e., if the concentration gradient has been formed, only the *oscillatory* instability occurs:

$$
Rosc = \frac{(\pi^2 + k^2)^3}{k^2} + \frac{PB}{P + 1},
$$
  

$$
\omega^2 = \frac{k^2B}{P(P + 1)(\pi^2 + k^2)},
$$
 (15)

or, after minimization of R over *k*,

$$
R_c^{osc} = R_0 + \frac{PB}{P+1}, \quad \omega_c^2 = \frac{B}{3P(P+1)}.
$$
 (16)

The mechanism of convective oscillations can be illustrated in the following way. Consider a small perturbation of the steady state  $(8)$  resulting in a displacement of an element of the suspension's volume upward. As long as the element rises, it loses both momentum (due to viscosity) and warmth (due to thermal conductivity) but maintains the number of grains since the diffusion is negligible. The motion upward stops when the element reaches a position in which it is surrounded by fluid of the same density: the fluid is cooler but less concentrated in heavy grains than the element is. So in the new position the element is still warmer than its surroundings and hence it continues to cool down. As a result, it becomes heavy—because of its heightened concentration than the surroundings, and then it begins to sink. Thus, the gradient of concentration (8) gives rise to a *restoring force*; therefore perturbations are oscillatory. Their subsequent fate depends on the Rayleigh number: the oscillations die out if  $R \leq R_c^{\text{osc}}$  and continue to grow at  $R > R_c^{\text{osc}}$ .

For real boundary conditions  $(12)$  on the confining rigid planes the pair of equations  $(13)$  can be easily solved by the Galerkin method. The simplest Galerkin approximation satisfying the conditions  $(12)$  has the form [13]

$$
v = v_0 \left(z^2 - \frac{1}{4}\right)^2
$$
,  $\theta = \theta_0 \left(z^2 - \frac{1}{4}\right)$ . (17)

This leads to the dispersion relation for the frequency  $\omega$  of neutral oscillations:

$$
i\omega P \left[ \omega^2 P(f_1 + 2) + \frac{27}{28} k^2 R - k^2 B - f_1 f_2 \right] + \omega^2 P f_3 = k^2 f_1 B,
$$
\n(18)

where we introduced the *k*-dependent functions

$$
f_1 = 10 + k^2
$$
,  $f_2 = 504 + 24k^2 + k^4$ ,  $f_3 = f_1(f_1 + 2) + Pf_2$ .

In the absence of the concentration gradient  $(B=0)$ , Eq. (18) predicts a stationary convective instability at

$$
R^{st} = 28f_1f_2/(27k^2). \tag{19}
$$

If the concentration equilibrium settled *before* imposing the temperature gradient, then only the *oscillatory* instability arises. In this case Eq.  $(18)$  gives

$$
Rosc = \frac{28f_2}{27} \left( \frac{f_1}{k^2} + \frac{PB}{f_3} \right), \quad \omega^2 = \frac{k^2 f_1}{P f_3} B. \tag{20}
$$

Minimization of  $R^{st}$  and  $R^{osc}$  over *k* yields the approximate critical values of R,  $k$ , and  $\omega$ . The relative error of those estimates with respect to the exact values presented in Table I does not exceed 2.5%. For instance, in the case of  $B=0$  our Galerkin approximation (17) gives  $R_c^{st} \equiv R_0 = 1750$ ,  $k_c$  $=$  3.116 instead of R<sub>0</sub> $=$ 1708,  $k_c$  = 3.117. Thus the solutions obtained for  $L=0.00015$  and 0 practically coincide. In the two last columns of Table I there are adduced dimensional estimates for the critical temperature difference  $\Delta T_c$  and the period of neutral oscillations  $T_c$  obtained for the layer thickness  $h=2$  mm and the unit of time  $h^2/v=4.91$  s (we take the kinematic viscosity coefficient  $\nu=8.15\times10^{-3}$  cm<sup>2</sup>/s [11]).

The critical Rayleigh number for oscillatory instability is always higher than that for stationary instability—cf. Eqs.  $(19)$  and  $(20)$ . Therefore, after some oscillations, when the initial concentration profile is washed away by convection, the stationary finite-amplitude convection is eventually settled. This prediction is confirmed by the numerical solution of nonlinear equations  $(4)$ – $(6)$  with the boundary conditions  $(12)$ . Numerical results are presented in Fig. 1. The calculations were performed for the mean particle concentration  $\bar{\phi}$ =1.5% (the volume fraction), and B=399 if the baro-



FIG. 1. Evolution of stream function  $\Psi(t)$  after the onset of convection in homogeneous  $(B=0)$  and stratified  $(B=399)$  colloids. Excess  $\Delta$  of Rayleigh number over its critical value is (a) 13%, (b) 19%, and (c) 25%.

metric distribution has been formed, or  $B=0$  if the equilibrium stratification has had no time to settle. In the latter case, the colloid behaves like a pure fluid where the stationary convection sets in at  $R > R_c^{st} = 1708$ . In the former case, i.e., when the colloidal solution was preliminarily stratified by gravity, there occurs oscillatory convection at  $R > R_c^{osc}$ =2079; no subcritical oscillations have been detected.

Evolution of the stream function  $\Psi(t)$  in the center of a convective roll is shown in Fig. 1. As is seen from the figure, the evolution depends on the excess of R over  $R_c^{osc}$ : *supercriticality* is defined as  $\Delta = (R/R_c^{osc}) - 1$ . The higher  $\Delta$  the sooner oscillatory convection washes away the initial concentration profile removing thereby the reason for the oscillations. As a result, the oscillatory regime of convection changes into the stationary one: both the amplitude and frequency of oscillations decrease with time, and the function  $\Psi(t)$  coincides finally with the stationary solution taken for the case of a nonstratified colloid  $(B=0)$ .

There is still a very narrow interval  $\Delta_{\omega}$  where the oscillatory instability produces undamped supercritical oscillations so weak that they do not destroy the concentration profile completely. (Such oscillations were observed for the first time [14] in water-ethanol mixtures with  $L \sim 10^{-2}$  stratified by the Soret effect.) The width of  $\Delta_{\omega}$  can be estimated as follows. Mixing occurs only when there is relative motion between various parts of the colloid. Hence, the mixing rate  $\dot{\gamma}$  should depend on the space derivatives of the fluid velocity. Near the convection threshold, the velocity and its gradients  $\partial v_i / \partial x_k$  are small, so  $\dot{\gamma}$  is supposed to be a linear function of the gradients:  $\dot{\gamma} \sim \partial v / \partial x \sim k_c^2 \Psi_{\text{max}}$ . Substituting here  $\Psi_{\text{max}} \simeq a \kappa \sqrt{\Delta_{\omega}}$  and  $k_c \simeq \pi/h$  gives  $\dot{\gamma} \sim \pi^2 a \sqrt{\Delta_{\omega}}(\kappa/h^2)$ . Undamped oscillations exist if the concentration profile is restored by diffusion faster than it (the profile) is destroyed under convection. Since  $\tau_D \approx h^2 / (\pi^2 D)$ , one gets  $\dot{\gamma} \tau_D$  $\sim (a/L)\sqrt{\Delta_{\omega}}$  so the last inequality reduces to  $\Delta_{\omega} \leq L^2/a^2$ . The amplitude *a* of the pattern depends on the Prandtl number (see [15]). For the roll pattern and P=5.5 one gets *a* =0.846. Thus, for L  $\sim$  10<sup>-4</sup> the interval  $\Delta_{\omega}$  is negligible.

Figure 2 presents the streamlines, isotherms, and concentration isolines in 9.5 units of time after imposing the temperature difference corresponding to  $R = 2640$ . It is seen that the streamlines and isotherms are *smooth* while the field of concentration looks *disordered*. This difference can be explained by the fact of a very large mismatch in diffusion coefficients in the equations of velocity and temperature, on the one hand, and concentration, on the other hand. Actually, small vortices and localized temperature perturbations rapidly disappear owing to the diffusion of velocity (the viscous term  $\nu \nabla^2$ **v** in the equation of fluid motion) and temperature (the term  $\kappa \nabla^2 T$  in the thermal conductivity equation), while the mass-diffusion term  $D\nabla^2 \phi$  in the diffusion equation is negligible. Therefore, even small islets of increased and decreased concentration do not dissolve due to diffusion but only spread by convection.

The pictures shown in Fig. 3 present a succession of snapshots of concentration isolines at a certain time after imposing the temperature difference on the fluid layer. They illustrate how the field of concentration is breaking into small pieces and then becoming more and more fragmentary.

Let us take into consideration the Soret effect. The flux density of the matter,

$$
\mathbf{j} = \phi \mathbf{u} - D(\nabla \phi + S \nabla T), \tag{21}
$$

consists of the diffusion part caused by concentration and temperature gradients (S is the Soret coefficient), and the regular flux provided by the particle sedimentation under gravity. The Archimedean force  $\Delta \rho V g$  and the Stokes drag coefficient  $3\pi\eta d$  determine the downfall velocity of the particle in the liquid:

$$
\mathbf{u} = \Delta \rho V \mathbf{g} / (3 \pi \eta d) = -(D/l) \mathbf{e};\tag{22}
$$

we have used definitions  $(1)$  and  $(2)$  for the diffusion coefficient *D* and the sedimentation length *l*. Substituting **u** from Eq.  $(22)$  into Eq.  $(21)$  gives

$$
\mathbf{j} = -D[\nabla \phi + S \nabla T + (\phi/l)\mathbf{e}]. \tag{23}
$$

Here the first two terms describe the diffusion and thermo-



FIG. 2. Streamlines (a), isotherms (b), and lines of constant concentration (c) for  $R = 2640$  and  $B = 399$  at the moment  $t = 9.5$ .

diffusion fluxes, while the third term may be treated as a *barodiffusion* flux [16]:  $-D(\phi/l)\mathbf{e}=D(\phi/p)\nabla p$ .

If the steady state  $(j=0)$  has been formed in the presence of a temperature gradient  $dT/dz = -\Delta T/h$ , the equilibrium value of the concentration gradient is

$$
\frac{d\phi}{dz} = S\frac{\Delta T}{h} - \frac{\phi}{l}.
$$

So, in the dimensionless form, instead of Eq.  $(8)$  one gets

$$
d\phi/dz = \psi \mathbf{R} - \mathbf{B},\tag{24}
$$

where  $\psi=(\beta/\alpha)S$  is the *separation ratio*. For colloidal solutions used in [11], this value is negative  $(\psi \approx -4$  at  $\overline{\phi}$  $=1.5\%$ ), while the Boltzmann number is always positive. In fact, substituting *l* from Eq. (2) and  $\beta = \Delta \rho / \rho$  into the definition  $(7)$  yields



FIG. 3. Evolution of the concentration field for B=399 and R =2350 (the supercriticality  $\Delta$ =13%).



FIG. 4. Stability diagram in the  $(R, \psi)$  plane for B=399. Oscillatory marginal stability, solid line; stationary Soret instability, dashed lines; frequency of neutral oscillations $\times$ 300, dash-dotted line;  $\bullet$ , codimension-2 point.

$$
B = \frac{(\Delta \rho \overline{\phi} g)^2 h^4}{\eta \kappa \overline{n} k_B T} > 0,
$$

where  $\bar{n}$  is the mean number density of colloidal grains. Replacing B in Eq. (18) by  $(B - \psi R)$ , we arrive at the dispersion relation

$$
i\omega P \left[ \omega^2 P(f_1 + 2) + \frac{27}{28} k^2 R - k^2 (B - \psi R) - f_1 f_2 \right] + \omega^2 P f_3
$$
  
=  $k^2 f_1 (B - \psi R).$  (25)

Interestingly, in contrast to Eq.  $(18)$  which has a stationary solution only if  $B=0$ , Eq. (25) has such a solution for any nonzero B value:

$$
R^{st} = B/\psi.
$$
 (26)

More precisely, restoring the diffusion term in Eq.  $(11)$ , instead of Eq.  $(26)$  we obtain

$$
R^{st} = \left[ B + \frac{10}{7}L(504 + 24k^2 + k^4) \right] / \psi.
$$

This function has the minimum  $R_c^{st} = (B + 720L)/\psi$  in the long-wavelength limit  $k=0$ . For  $L \sim 10^{-4}$ , however, the term 720L does not exceed 0.1 and hence is negligible compared to any reasonable value of B. Thus Eq.  $(26)$  describes two branches of the stationary Soret instability shown in Fig. 4, one of which corresponds to positive values of  $\psi$  and R, and the other fits their negative values. The dispersion relation  $(25)$  also determines the oscillatory Rayleigh instability:

$$
Rosc = \frac{28f_2(f_1f_3 + k^2PB)}{k^2(27f_3 + 28\psi Pf_2)},
$$
  

$$
\omega^2 = \frac{f_1(27k^2B - 28\psi f_1f_2)}{27f_3 + 28\psi Pf_2}.
$$
 (27)

Putting here  $\omega=0$  we find the coordinates of the *codimension-2 point* where the marginal curve of oscillatory

instability  $(27)$  branches from the curve of stationary instability (26):  $\psi_{c2} = B/R_0$ , R<sub>c2</sub>=R<sub>0</sub> (see Fig. 4). So, for B=399 one has  $\psi_{c2}$ =399/1708 $\approx$ 0.234.

The above estimates were made for colloids earlier used by Cerbino *et al.* [11]. We dare say the authors could have observed the oscillatory instability predicted in the present work if they had set themselves it as an object. However, they studied quite another convective problem. They prepared no steady state but imposed abruptly on the fluid layer a very strong heating from above. Then the concentration gradient induced by the negative Soret effect extends from the layer boundaries into the entire fluid as  $\lceil 13 \rceil$ 

$$
\frac{\partial \phi}{\partial z} = \psi \mathbf{R} \left[ 1 - \text{erf}\left(\frac{z}{2\sqrt{\mathbf{L}t}}\right) \right],
$$

where  $\text{erf}(\cdot)$  is the error function. Thus, over a certain time the concentration gradient is located within the diffusion skin layer  $\delta \sim \sqrt{L}t$  near the confined solid surfaces. During this time the characteristic length scale is  $\delta$  instead of the layer depth *h*, and hence the effective Rayleigh number grows proportionally to  $t^{3/2}$  as time goes on. As a result, the fluid velocity rapidly grows too and mixes the concentration, eliminating thereby the driving force of the Soret convection. The latter leads to a rapid damping of the motion, but then the concentration gradient is formed again and the process is repeated. A period of these self-sustained oscillations was estimated in [13], and the estimates were corroborated before long by experiments  $[11]$ . It is clear that there is a certain qualitative likeness between our problem and that of Cerbino et al. [11]. Nonetheless our predictions and their experimental results surely cannot be compared.

To summarize, the onset of convection in colloids depends on the way in which the initial steady state is prepared. Actually, depending on the *timing* in the temperature application, three scenarios are possible. If a supercritical temperature difference  $\Delta T > \Delta T_c$  is applied to the fluid layer before establishing the barometric particle distribution, the colloid behaves as a pure liquid; thus stationary convection sets in. Contrarily, if  $\Delta T$  is applied after stratifying the colloid by gravity, oscillatory instability occurs.

We should note the high sensitivity of the transient stage of convection in colloids to dissolved matter. In fact, according to our theory even negligible amounts of suspended grains may induce serious consequences. For example, adding only 0.2 vol % of silica grains into water provides a Boltzmann number  $B \approx 54$  that leads to oscillatory instability with frequency  $\omega \approx 0.62$  (for a fluid layer of 2 mm in thickness this dimensionless frequency corresponds to the period of convective oscillations  $T \approx 50$  s). The instability occurs at  $R_c^{\text{osc}} \approx 1760$ , exceeding the threshold of the stationary instability  $R_0 = 1708$  by 3%.

The last scenario implies that the subcritical temperature difference  $\Delta T < \Delta T_c$  is imposed from the very beginning and then increases gradually in order to build the concentration profile undisturbed by convection. In this case the profile is formed under the action of both gravity and the Soret effect; thus the character of the instability—stationary or oscillatory—proves to be dependent on the correlation between the Boltzmann number and the separation ratio.

Anyway, oscillatory convection lasts as long as it takes the convective motion to wash away the initial concentration profile. Then the fluid becomes homogeneous and stationary convection is eventually established. As we demonstrated, the more the Rayleigh number exceeds its critical value for oscillatory instability—and hence the higher the amplitude of convective oscillations—the sooner stationary supersedes oscillatory convection.

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