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Convective instability of magnetized ferrofluids

Mark I. Shliomis[a,*], Boris L. Smorodin[b]

[a] *Department of Mechanical Engineering, Ben Gurion University of the Negev, P.O.B. 653, Beer-Sheva 84105, Israel*
[b] *Department of Theoretical Physics, Perm State University, 15 Bukirev str., 614000 Perm, Russia*

Abstract

Convective instability in a flat ferrofluid layer subject to a transverse uniform magnetic field is investigated theoretically. A temperature gradient imposed across the layer induces a concentration gradient of magnetic grains owing to the Soret effect. Both these gradients cause a spatial variation in magnetization that establishes a gradient of magnetic field intensity within the fluid layer. The field gradient induces in its turn a redistribution of magnetic grains due to magnetophoresis. The resulting self-consistent magnetic force tries to mix the fluid. Linear analysis performed for the case of realistic boundary conditions on confined horizontal planes predicts double-diffusive oscillatory instability in a certain region of parameters, whereas if the particle diffusion had been not operative, then only stationary instability would occur.

Keywords: Ferrofluids; Convective instability; Magnetophoresis; Soret effect

1. Introduction

Mechanical equilibrium of a nonisothermal ferrofluid in a magnetic field H is in general impossible. At the basis of the mechanism of thermomagnetic convection [1–5] lies the temperature dependence of the magnetization M; under otherwise equal conditions, the colder elements of the fluid are more strongly magnetized, and therefore they are subject to a larger magnetic force in the direction of ∇H. Interestingly, this convection can arise even in a *uniform* applied magnetic field [4]. Let such a vertical external field act upon a ferrofluid confined between two horizontal planes at the temperature $T(z)$. Then the dependence $M(T)$ provides the field nonuniformity within the fluid layer. Indeed, from the Maxwell equation $\operatorname{div}\mathbf{B}=0$ where $\mathbf{B}=\mathbf{H}+4\pi\mathbf{M}$, it follows that

$$\frac{dH}{dz}=-4\pi\frac{dM}{dz}=-\frac{4\pi}{\hat{\mu}}\left(\frac{\partial M}{\partial T}\right)_H\frac{dT}{dz}, \tag{1}$$

where $\hat{\mu}=1+4\pi(\partial M/\partial H)$ is the differential magnetic permeability. As the field gradient (1) is directed *opposite* to the gradient of magnetization, the magnetic force $\mathbf{F}=M\nabla H$ always tries to mix the ferrofluid. The magnetic mechanism of convection predominates over the buoyancy mechanism in a sufficiently thin fluid layer, $d\sim 1$ mm. Actually, as it is shown below, the magnetic Rayleigh number $Rm\propto(\Delta T\, d)^2$ whilst the gravitational Rayleigh number $Rg\propto\Delta T\, d^3$, i.e. $Rm/Rg\propto\Delta T/d$. With a decrease in d, the critical temperature gradient $\Delta T_c/d$ increases, so that it becomes $Rm\gg Rg$.

Ferrofluids should be treated as *binary mixtures* whose magnetization depends on the concentration ϕ of magnetic grains. An imposed ∇T induces $\nabla\phi$ owing to the *Soret effect*; the magnetic force $\mathbf{F}$ leads to an additional redistribution of magnetic grains due to *magnetophoresis*. As it was recently demonstrated [6], these thermo- and magnetodiffusion processes give rise to *oscillatory instability*, which cannot occur in a hypothetic pure (i.e., single-component) magnetized fluid [4,5,7,8].

The flux of magnetic grains in a ferrofluid is

$$\mathbf{j}=\phi\mathbf{u}-D(\nabla\phi+S_T\nabla T), \tag{2}$$

*Corresponding author. Fax: +972-8-647-7100.
E-mail address: shliomis@bgumail.bgu.ac.il (M.I. Shliomis).

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where $D = k_B T/6\pi\eta a$ is the diffusion coefficient of grains of radius a, S_T the Soret coefficient, and the regular grain velocity $\mathbf{u}$ with respect to the liquid is determined by the Stokes drag coefficient $6\pi\eta a$ and the magnetic force acting upon a particle:

$$6\pi\eta a\mathbf{u} = m\mathscr{L}(\xi)\nabla H. \tag{3}$$

Here $m = M_s V$ is the magnetic moment of the particle of volume V, M_s is the saturation magnetization of a dispersed ferromagnet, and $\mathscr{L}(\xi) = \coth\xi - \xi^{-1}$ is the Langevin function of the parameter $\xi = mH/k_B T$. Eliminating $\mathbf{u}$ from Eqs. (2) and (3), we obtain

$$\mathbf{j} = -D[\nabla\phi + S_T\nabla T - (\phi/H)\xi\mathscr{L}(\xi)\nabla H]. \tag{4}$$

At the equilibrium ($\mathbf{j}=0$) in a uniform applied magnetic field, only the temperature gradient can cause gradients of ϕ and H. Their *self-consistent* values are determined by Eqs. (1) and (4):

$$\frac{dH}{dz} = \frac{4\pi M}{\sigma T}\left(\Psi + \frac{\xi\mathscr{L}'}{\mathscr{L}}\right)\frac{dT}{dz}, \tag{5}$$

$$\frac{d\phi}{dz} = -\frac{\hat{\mu}\phi}{\sigma T}\left(\Psi - \frac{\hat{\mu}-1}{\hat{\mu}}\xi\mathscr{L}\right)\frac{dT}{dz}. \tag{6}$$

Here $\Psi = (T/\phi)S_T$ plays the role of the *separation ratio* in the mechanism of thermomagnetic convection, $\sigma = \hat{\mu} + 12\pi\chi_0\mathscr{L}^2(\xi) = 1 + 12\pi\chi_0(\mathscr{L}' + \mathscr{L}^2)$, and $\chi_0 = \phi M_s m/3k_B T$ is the initial magnetic susceptibility. Note that $d\phi/dz \neq 0$ even if $\Psi = 0$. The non-Soret $\phi - T$ coupling originates from the magnetophoresis, which is manifested as a negative separation ratio.

2. Eigenvalue problem

Small perturbations of a standing mode are characterized by velocity $\mathbf{v}$, pressure p, temperature θ, concentration φ, magnetic field $\mathbf{h}$, and magnetic induction $\mathbf{b}$. The latter reads

$$\mathbf{b} = (1 + 4\pi M/H)\mathbf{h} + 4\pi\left[\frac{\partial M}{\partial T}\theta + \frac{\partial M}{\partial \phi}\varphi + \left(\frac{\partial M}{\partial H} - \frac{M}{H}\right)h_z\right]\mathbf{e}, \tag{7}$$

where $\mathbf{e}$ is the unit vector along axis z. Let us introduce the potential Φ of magnetic field perturbations: $\mathbf{h} = \nabla\Phi$. Substituting $\mathbf{b}$ from Eq. (7) and $M = \phi M_s\mathscr{L}(\xi)$ into $\operatorname{div}\mathbf{b} = 0$, we obtain the equation for Φ:

$$\mu\nabla^2\Phi - (\mu - \hat{\mu})\frac{\partial^2\Phi}{\partial z^2} = -4\pi M\left(\frac{1}{\phi}\frac{\partial\varphi}{\partial z} - \frac{\xi\mathscr{L}'}{T\mathscr{L}}\frac{\partial\theta}{\partial z}\right), \tag{8}$$

where $\mu = 1 + 4\pi(M/H) = 1 + 12\pi\chi_0\mathscr{L}(\xi)/\xi$.

The perturbation $\mathbf{f}$ of the magnetic force density $\mathbf{F} = M\nabla H$ is

$$\mathbf{f} = -h_z\nabla M + \left(\frac{\partial M}{\partial T}\theta + \frac{\partial M}{\partial\phi}\varphi + \frac{\partial M}{\partial H}h_z\right)\nabla H.$$

On substitution of dH/dz from Eq. (5) and $h_z = \partial\Phi/\partial z$, the last formula takes the form

$$f_z = \frac{M}{\sigma T}\left(\Psi + \frac{\xi\mathscr{L}'}{\mathscr{L}}\right)\left[\hat{\mu}\frac{\partial\Phi}{\partial z} + 4\pi M\left(\frac{\varphi}{\phi} - \frac{\xi\mathscr{L}'}{\mathscr{L}}\frac{\theta}{T}\right)\right]\frac{dT}{dz}. \tag{9}$$

Eq. (4) under the replacement $\phi \to \phi + \varphi, T \to T + \theta$, and $H \to H + \partial\Phi/\partial z$ determines the matter flux perturbation. So, making allowance for Eq. (6), we obtain the diffusion equation

$$\frac{\partial\varphi}{\partial t} = D\nabla^2\left(\varphi + \frac{\phi}{T}\Psi\theta - \frac{\phi}{H}\xi\mathscr{L}\frac{\partial\Phi}{\partial z}\right) + \frac{\phi\Delta T\hat{\mu}}{\sigma T d}\left(\Psi - \frac{\hat{\mu}-1}{\hat{\mu}}\xi\mathscr{L}\right)(\mathbf{ve}). \tag{10}$$

Pass over to dimensionless variables by choosing a unit of length d, time d^2/κ (κ is the fluid thermodiffusivity), velocity κ/d, temperature ΔT, concentration $(\Delta T/T)\phi$, and field potential $4\pi M(\Delta T/T)d$. Then inserting the force given by Eq. (9) in the equation of fluid motion and assuming all perturbations to be dependent on time and horizontal coordinates as $exp[\imath(\omega t + k_x x + k_y y)]$, we arrive at the following system of linear equations for z-dependent amplitudes of the vertical component of velocity $W = (\mathbf{ve})$, temperature θ, matter flux potential $\Omega = \varphi + \Psi\theta - (\sigma - \hat{\mu})D\Phi$ (it is introduced as an independent variable instead of the concentration perturbation φ), and magnetic field potentials inside (Φ) and outside (Φ_e) the fluid layer [6]:

$$(D^2 - k^2)(D^2 - k^2 - \imath\omega/P)W = k^2 RmG[\Omega - (\Psi + \xi\mathscr{L}'/\mathscr{L})\theta + \sigma D\Phi], \tag{11a}$$

$$(D^2 - k^2 - \imath\omega)\theta - W = 0, \tag{11b}$$

$$L(D^2 - k^2 - \imath\omega/L)\Omega + \imath\omega[\Psi\theta - (\sigma - \hat{\mu})D\Phi] = -\sigma^{-1}[\hat{\mu}\Psi - (\hat{\mu} - 1)\xi\mathscr{L}]W, \tag{11c}$$

$$(\sigma D^2 - \mu k^2)\Phi = -D[\Omega - (\Psi + \xi\mathscr{L}'/\mathscr{L})\theta], \tag{11d}$$

$$(D^2 - k^2)\Phi_e = 0. \tag{11e}$$

Here $P = \eta/\rho\kappa$ is the Prandtl number, $L = D/\kappa$ is the Lewis number, $D \equiv d/dz$,

$$Rm = \frac{[\phi M_s\Delta T d]^2}{\eta\kappa T^2},$$

$$G = \frac{4\pi\mathscr{L}^2}{\sigma}\left(\Psi + \frac{\xi\mathscr{L}'}{\mathscr{L}}\right). \tag{12a, b}$$

Note that the magnetic Rayleigh number Rm is always *positive*.

To specify the solutions of Eqs. (11), one needs 12 boundary conditions (b.c.) at confined surfaces $z = \pm 1/2$. For the case of rigid, perfectly heat conducting and completely impervious boundaries, the b.c. on velocity, temperature and matter flux perturbations read as

$$W = DW = \theta = D\Omega = 0 \quad \text{at} \quad z = \pm 1/2. \qquad (13)$$

B.c. of continuity of tangential components of magnetic field and the normal component of magnetic induction (7) on the layer borders take the form [6]

$$\Phi = \Phi_e,\ \sigma D\Phi + \Omega = D\Phi_e \quad \text{at} \quad z = \pm 1/2, \qquad (14)$$

respectively. Eigenvalue problem (11)–(14) can be simplified by means of exception from consideration of the value Φ_e. According to Eq. (11e), $\Phi_e \propto \exp(\pm kz)$. As the perturbation must decay far from the layer, we find $D\Phi_e = \mp k\Phi_e$ at $z = \pm 1/2$. This relation permits to eliminate Φ_e from Eq. (14):

$$\sigma D\Phi \pm k\Phi = -\Omega \quad \text{at} \quad z = \pm 1/2. \qquad (15)$$

Thus, we have obtained the closed b.c. on Φ, so there is no necessity to find the magnetic potential outside the layer.

Convective instability of magnetized ferrofluids in the Bénard configuration has been analyzed recently [6] for the case of ideal (so-called "free") boundaries at which $W = D^2W = \theta = \Omega = D\Phi = 0$. These b.c. permit to obtain an exact solution, $W,\ \theta,\ \Omega \propto \cos \pi z$, $\Phi \propto \sin \pi z$, whose properties guide our present analysis produced below.

3. Solution for realistic boundaries

We have obtained critical values of the Rayleigh number for stationary ($\omega = 0$) and oscillatory ($\omega \neq 0$) instabilities using the Galerkin method by expanding the velocity, temperature and matter flux potential in the series containing only *even* functions of z:

$$W = \sum_{n=0}^{N-1} a_{2n} W_{2n},\ \theta = \sum_{n=0}^{N-1} b_{2n}\theta_{2n},\ \Omega = \sum_{n=0}^{N-1} c_{2n}\Omega_{2n}. \qquad (16)$$

As the W_{2n} basis we have chosen even solutions of the eigenvalue problem

$$(D^2 - k^2)^2 W_{2n} = -\lambda_{2n}(D^2 - k^2)W_{2n},$$
$$W_{2n}(\pm 1/2) = DW_{2n}(\pm 1/2) = 0,$$

which describes velocity perturbations in the fluid layer when $Rm = 0$—see Eq. (11a). The eigenfunctions W_{2n} are

$$W_{2n} = \frac{\cosh kz}{\cosh k/2} - \frac{\cos\sqrt{\lambda_{2n} - k^2}z}{\cos\sqrt{\lambda_{2n} - k^2/2}}$$

and eigenvalues λ_{2n} are determined by the equation

$$\sqrt{\lambda_{2n} - k^2}\tan\sqrt{\lambda_{2n} - k^2/2} = -k\tanh k/2.$$

Functions θ_{2n} and Ω_{2n} are determined by the corresponding eigenvalue problems:

$$(D^2 - k^2)\theta_{2n} = -\nu_{2n}\theta_{2n},\quad \theta_{2n}(\pm 1/2) = 0$$
$$(D^2 - k^2)\Omega_{2n} = -\rho_{2n}\Omega_{2n},\quad D\Omega_{2n}(\pm 1/2) = 0$$

and have the form

$$\theta_{2n} = \cos(2n+1)\pi z,\quad \nu_{2n} = (2n+1)^2\pi^2 + k^2;$$
$$\Omega_{2n} = \cos 2n\pi z,\ \rho_{2n} = 4n^2\pi^2 + k^2,\quad n = 0, 1, 2, \ldots.$$

Trial functions (16) satisfy b.c. (13). Substituting θ and Ω from Eq. (16) in field perturbation equation (11d) and satisfying b.c. (15), we find an exact *odd* solution of the equation:

$$\Phi = \sum_{n=0}^{N-1} b_{2n}\Phi_{2n}^{(1)} + \sum_{n=0}^{N-1} c_{2n}\Phi_{2n}^{(2)}, \qquad (17)$$

$$\Phi_{2n}^{(1)} = \frac{(2n+1)\pi(\Psi + \xi\mathscr{L}'/\mathscr{L})}{(2n+1)^2\pi^2\sigma + \mu k^2} \times \left[\sin(2n+1)\pi z - \frac{\cos n\pi \sinh \gamma kz}{\sigma\gamma\cosh\gamma k/2 + \sinh\gamma k/2}\right],$$

$$\Phi_{2n}^{(2)} = -\frac{1}{(2n\pi)^2\sigma + \mu k^2} \times \left[2n\pi\sin 2n\pi z + \frac{\mu k\cos n\pi\sinh\gamma kz}{\sigma\gamma\cosh\gamma k/2 + \sinh\gamma k/2}\right],$$

where $\gamma = \sqrt{\mu/\sigma}$. Functions (16) and (17) are substituted into Eqs. (11) and then Eqs. (11a)–(11c) are required to be orthogonal to each of the functions W_{2n}, θ_{2n}, and Ω_{2n}, respectively. This way we arrive at the $3N$-rank determinant of the coefficients a_{2n}, b_{2n}, c_{2n},

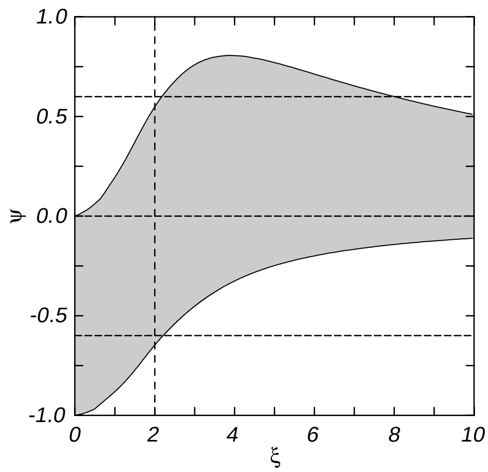


Fig. 1. The corridor of oscillatory instability (*dashed*) in ξ, Ψ plane. Dashed lines correspond to $\Psi = 0, -0.6, +0.6$ and $\xi = 2$. Cross-sections of the surface $Rm_c(\xi, \Psi)$ along these lines are presented below in Figs. 2–5, respectively.

which leads to a characteristic equation to be solved for the eigenvalue $Rm(k)$.

Our analysis has shown that there exists an area in the (ξ, Ψ)-plane

$$-\frac{\xi\mathscr{L}'(\xi)}{\mathscr{L}(\xi)} < \Psi < \frac{\hat{\mu}-1}{\hat{\mu}}\xi\mathscr{L}(\xi) \tag{18}$$

(see Fig. 1), where oscillatory instability occurs. Interestingly, the area location does not depend on the boundary conditions of the problem. The same corridor (18) of oscillatory instability has been obtained for unrealistic "free" boundaries [6]. Actually, on the lower border of the area, the critical Rayleigh number for oscillatory instability, Rm_c^{osc}, turns into infinity since the field gradient (5) (and the magnetic force too) vanishes on this border. On the upper border of the interval (18), the frequency of neutral oscillations, ω_c, turns into zero together with the concentration gradient (6). Indeed, if ϕ= const, only stationary instability can occur [4].

Note that ferrofluids are characterized by two very different time scales [9]: the long mass-diffusion time $\tau_D = d^2/\pi^2 D$ and the short thermodiffusion time $\tau_D = d^2/\pi^2\kappa$. The ratio τ_T/τ_D is equal to the Lewis number, $L = D/\kappa$, which is very small ($\sim 10^{-4}$) due to the low diffusion coefficient for magnetic grains. Therefore, there are two possible scenarios of the convection onset. If the temperature difference ΔT between the layer boundaries is increased from zero till a supercritical value for a time $\tau \ll \tau_D$, diffusion processes have no time to evolve; hence magnetic colloid behaves like a pure fluid. Stationary convection starts in this case at a certain $Rm_c^{(0)}$. If however ΔT is formed for a time $\tau > \tau_D$, the concentration gradient is built up undisturbed by convection. Then in the dashed area of Fig. 1 there arises oscillatory instability at Rm_c^{osc}. Thus, both the critical Rayleigh numbers, Rm_c^{osc} and $Rm_c^{(0)}$, have a direct physical meaning. Depending on the conditions of a real experiment, either stationary convection above $Rm_c^{(0)}$ or oscillatory convection above Rm_c^{osc} set in.

4. Numerical results and conclusions

Results of applying the Galerkin method for the case $\Psi = 0$ and realistic ("rigid") boundaries (subindex r) are indicated in Table 1 and Fig. 2 in comparison with an exact solution for "free" boundaries (subindex f). In the figure, the logarithms of Rm_c^{osc} and $Rm_c^{(0)}$, and critical cyclic frequency $f = \omega_c/2\pi$ are presented as functions of the magnetic field ξ. The calculations have been performed for a ferrofluid with the initial magnetic permeability $\mu_0 = 1 + 4\pi\chi_0 = 3$. The critical Rayleigh number for oscillatory instability in the fluid (see curve a_r in Fig. 2) reaches a minimum, min[Rm_c^{osc}] = 9340.7, at $\xi_c = 1.44$; corresponding wave number and frequency of neutral oscillations are $k_c = 3.72$ and $\omega_c = 15.14$. As seen from Table 1, these critical values weakly depend on the number $3N$ of trial function that we used.

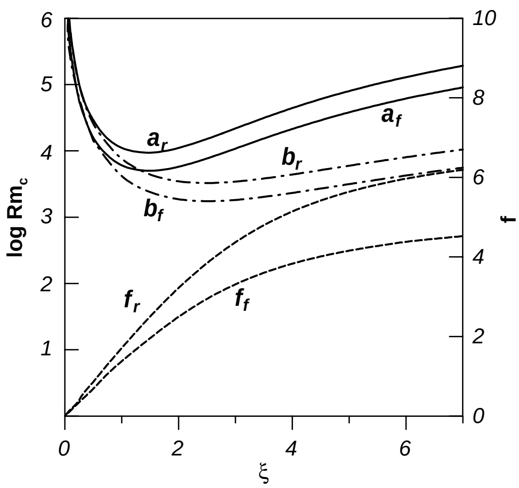


Fig. 2. Field dependence of Rm_c and circular frequency f in the case $\Psi = 0$. Curves (a)—oscillatory solution Rm_c^{osc}, curves (b)—stationary solution $Rm_c^{(0)}$ in the absence of particle diffusion. Solutions for realistic b.c. are marked by subindex r, for ideal ("free") boundaries—by subindex f.

Table 1
The dependence of critical parameters on numbers of trial functions in the case $\Psi = 0$

$3N$ (Rank of determinant)	min[Rm_c^{osc}]	ω_c	k_c	ξ_c
6	9380.193	15.148	3.755	1.439
9	9355.674	15.145	3.751	1.439
12	9347.244	15.142	3.750	1.439
15	9343.791	15.140	3.750	1.439
18	9342.140	15.140	3.749	1.439
21	9341.255	15.139	3.749	1.439
24	9340.740	15.139	3.749	1.439
Exact solution for "free" b.c.	4993.214	12.126	3.081	1.474

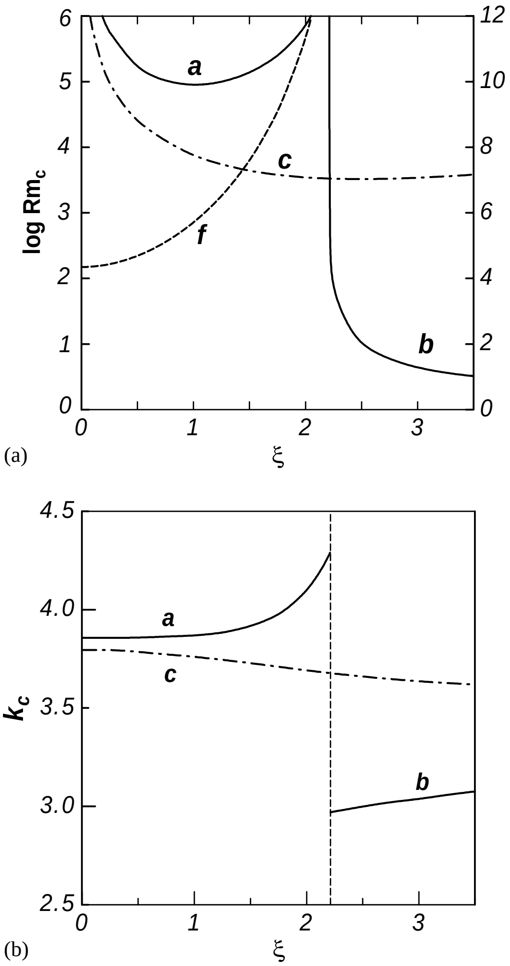
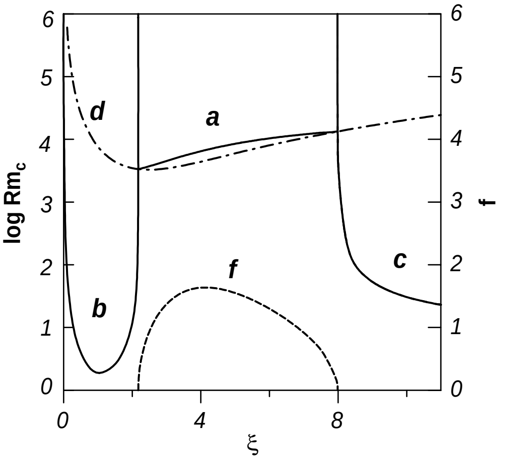


Fig. 3. Field dependence of Rm_c and f (a) and critical wave number k_c (b) for $\Psi = -0.6$. Curves (a)—oscillatory solutions Rm_c^{osc}, k_c and (b)—stationary solutions Rm_c^{st}, k_c; curves (c)—stationary solution $Rm_c^{(0)}$ in the absence of particle diffusion.



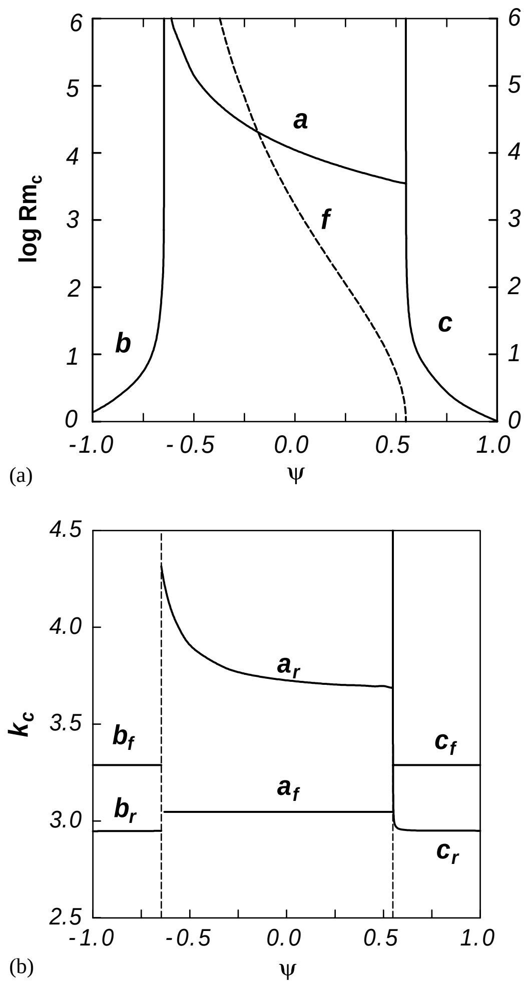


Fig. 5. Rm_c and f (a) and k_c (b) versus separation ratio Ψ at the fixed magnetic field strength $\xi = 2$. Curves (a)—oscillatory instability, curves (b, c)—stationary instability. Solutions for realistic b.c. are marked in Fig. 5b by subindex r, for free boundaries—by subindex f.

Therefore, all following calculations were carried out with 12 functions ($N = 4$).

Maps of stability in Rm_c, ξ plane are presented in Figs. 3 and 4 for $\Psi = -0.6$ and $+0.6$, respectively. For each of these magnitudes, there is a certain interval of ξ (see Fig. 1) in which oscillatory instability occurs. Out of the interval, there arises a stationary Soret convection above the critical Rayleigh numbers Rm_c^{st}. The latter are very small due to the small value of the Lewis number L. Having in mind a water-based ferrofluid, we carried out

Fig. 4. Field dependence of Rm_c and f for $\Psi = +0.6$. Curve (a)—oscillatory solution Rm_c^{osc} and (b, c)—stationary solution Rm_c^{st}; curve (d)—stationary solution $Rm_c^{(0)}$ in the absence of particle diffusion.

our calculations with $L = 1.9 \times 10^{-4}$ and the Prandtl number $P = 7$.

A diagram of stability on the (Rm_c, Ψ)-plane at the fixed magnitude of magnetic field is shown in Fig. 5. Two things distinguish the diagram from a traditional one for binary mixtures [10,11]. Firstly, the codimension-2 point (i.e., the intersection point of the oscillatory (a) and stationary (c) branches) is essentially shifted towards positive values of Ψ due to magnetophoresis. Secondly, the stationary branch b is usually located in the region of negative Rayleigh numbers (these determine the onset of convection in a binary mixture heated from *above*). However, thermomagnetic mechanism of convection has neither the top nor the bottom since $Rm \propto (\Delta T)^2$. Ψ dependence of Rm_c of f shown in the figures is basically similar to that found for the case of free boundaries [6], but the corresponding wave numbers vary sharply in the same range of Ψ values.

Thus, convective instability of magnetized ferrofluids is strongly effected by the magneto- and thermophoretic transfer of magnetic grains. The particle diffusion just gives an opportunity to observe oscillatory instability in a certain region of fluid parameters and in a certain experimental condition. Namely, an applied temperature difference must not be increased faster than the limit imposed by concentration diffusion. In the opposite case, ferrofluids behave like pure fluids, which results in stationary instability at $Rm_c^{(0)}$.

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