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INFLUENCE OF THERMODIFFUSIVE PARTICLE TRANSPORT ON THERMOMAGNETIC CONVECTION IN MAGNETIC FLUIDS

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Thermomagnetic convection is driven by a temperature difference and a magnetic field applied to a layer of fluid. Its onset is characterized by the so-called Rayleigh number Ra. Since a temperature gradient also drives thermal diffusion in binary fluids characterized by the Soret coefficient $\mathrm{S_T}$, the impact of the two transport phenomena on each other have to be investigated with the focus on the point of transition from enhanced to hindered or even suppressed convection. A linear stability analysis provides the functional context of $\mathrm{Ra(S_T)}$. Suppression of the convective motion is thereby reached for $\mathrm{S_T} < -0.001$, which implies a negative Soret coefficient. The zero magnetic field coefficient in ferrofluids is positive, but measurements of the magnetic Soret coefficient in an applied magnetic field prove that its sign is sensitive to the strength of the magnetic field. In fields higher than 75 kA/m, the Soret coefficient switches from positive to negative.

1. Introduction. Buoyancy-driven convection in a layer of fluid can establish when heating the lower and cooling the upper layer's boundary. The additional application of a magnetic field can enhance the convective motion what is referred to as thermomagnetic convection [1] which was experimentally investigated in [2, 3]. The experiments of [3] were carried out using different ferrofluids, which showed in one case the enhancement and in the other, against the expectations, the complete suppression of convective motion. Since the application of a magnetic field can have a major impact on the magnitude of the fluid's parameters, it was first expected to find a strong field-dependence of the viscosity, for example, which was not the case and could be excluded as a reason for the suppression [3]. It is, therefore, supposed that the applied temperature gradient leads to a second transport phenomenon in the layer, which opposes the direction of convective transport. Thermal diffusion, also called thermodiffusion, is a transport process driven by a temperature gradient leading to a separation of particles and fluid in a colloidal suspension, such as a ferrofluid [4]. Its intensity and direction of separation are characterized by the so-called magnetic-field-dependent Soret coefficient. A positive (negative) value moves the particles to the colder (hotter) side of the layer. The high influence of thermodiffusion on convective motion in zero magnetic field is theoretically and experimentally known [5, 6] and therefore its influence in the non-zero case is reasonable to be investigated.

2. Theoretical investigations on thermomagnetic convection. The onset of convection is investigated in a horizontal layer of ferrofluid, with the z-axis being aligned to the layer's height. For the first investigations, the layer is considered infinite in the x- and y-directions and bounded by free boundaries in the z-direction. The governing equations for this system are derived from [1] and [7] and contain the balance equations for mass, momentum, heat, and concentration as well as the Maxwell equations for the magnetic field and flux. To investigate the transition from conductive to convective heat transport in the layer, the hydrodynamic variables of the system, namely, velocity (v), temperature (T), concentration (c), and magnetic potential (Φ) are regarded perturbed linearly by w, Θ, δc, and φ. Also considering the ground state, when the fluid is at rest, leads to the following system of dimensionless linear differential equations for the deviations [8]

$$\mathrm{div}(\mathbf{v}) = 0, \tag{1}$$

$$\frac{\partial \triangle w}{\partial t} = \mathrm{Pr}\left\{\mathrm{Ra}\nabla_h\left[\Theta - \psi\delta c - \frac{\mathrm{M}_1}{1+\chi}\left(\frac{\partial_z\varphi}{1+\chi} - \Theta + \frac{\mathrm{M}}{\partial c}\delta c\right)\right] + \triangle^2 w\right\} \tag{2}$$

$$\frac{\partial\Theta}{\partial t} - w = \triangle\Theta, \tag{3}$$

$$\frac{\partial\delta c}{\partial t} = \mathrm{Le_m}\left(\triangle\delta c + \triangle\Theta\right), \tag{4}$$

$$\mathrm{div}(\mathbf{B}) = 0, \tag{5}$$

with $\nabla_h = \partial_{xx} + \partial_{yy}$, Pr being the Prandtl number, χ the susceptibility, $\mathbf{B}$ the magnetic flux, and $\mathrm{Le_m}$ denoting the magnetic Lewis number. Considering the boundary conditions for the deviations

$$\partial_{zz}w = w = \Theta = \partial_z\delta c + \partial_z\Theta = \partial_z\varphi = 0,$$

exponential ansatzes for the deviations in the x- and y-direction, and ansatzes fulfilling the boundary conditions in z-direction are chosen. The resulting system of equations can be solved and a functional context for the Rayleigh number, which denotes the threshold for convection, can be calculated. The former Rayleigh number [1]

$$\mathrm{Ra_{Fin}} = \frac{(\pi^2+k^2)^3}{k^2\left(1+\mathrm{M}_1\left(1-\dfrac{\pi^2}{\mathrm{M}_3k^2+\pi^2}\right)\right)}$$

can be directly compared to

$$\mathrm{Ra} = \frac{(\pi^2+k^2)^3}{k^2\left(1+\psi+M_1\left(1+\dfrac{\partial\mathrm{M}}{\partial c}\right)\left(1-\dfrac{\pi^2}{M_3k^2+\pi^2}\right)\right)} \tag{6}$$

by [8] including thermal diffusion, with k denoting the wave number. $\mathrm{M}_1 \approx 0.5$ and $\mathrm{M}_3 \approx 1.1$ are the factors containing fluid properties and magnetic field as well as factors from its scaling. The separation ratio ψ and $\partial\mathrm{M}/\partial c$ are proportional to $\mathrm{S_T}$.

Figs. 1 and 2 plot the resulting critical Rayleigh number, being the minimum of $\mathrm{Ra}(k)$ with respect to k, and the critical wave number in dependence on the Soret coefficient. Only $\mathrm{S_T} > -0.001$ leads to positive Rayleigh numbers so that a threshold for the onset of convective motion represented by the critical Rayleigh number can only be determined starting at that value. For $\mathrm{S_T} = 0$, the plotted Rayleigh number corresponds to the one calculated in [1]. Small negative Soret coefficients lead to a hindrance of the onset of convection if compared to the case neglecting thermal diffusion; a positive coefficient leads to an enhancement of convective motion in any case. The corresponding critical wave number increases strongly for small Soret coefficients up to 0.02 1/K, which is the point where saturation of the wave number's magnitude sets in. Experiments on thermodiffusion, as described in the following paragraphs, determine the actual magnetic-field-dependent Soret coefficient for the ferrofluid used.

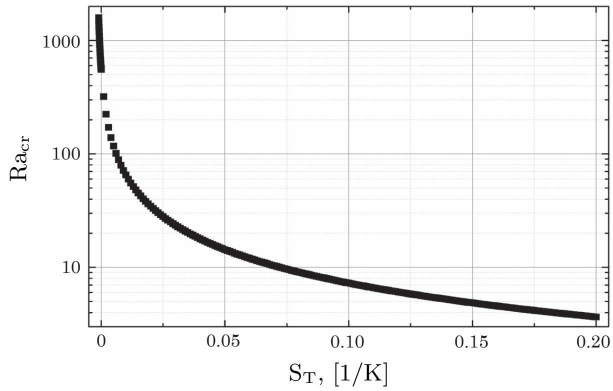

Fig. 1. Plot of the critical Rayleigh number dependent on the Soret coefficient.

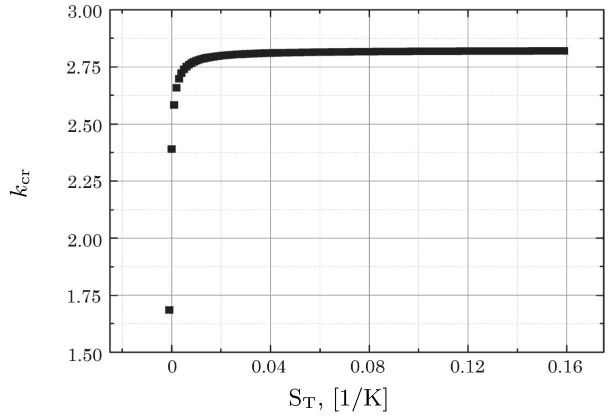

Fig. 2. Plot of the critical wave number dependent on the Soret coefficient.

3. Experiments on thermodiffusion.

3.1. Experimental setup. The Soret coefficient is measured indirectly by detecting the separation of particles and carrier liquid in a horizontal thermodiffusion cell based on a design presented in [4]. The particles' concentrations at the two ends of a cylindrical fluid container (*1*) (see Fig. 3) is determined by measuring the inductance of the two sensor coils (*2*) wrapped around the cell. The coils' parameters, such as diameter and number of layers, are important to adjust the sensitivity of the measurement. For that reason, numerical simulations of the magnetic field of the coils have been carried out to define the ideal set of parameters. Since the inductances of the coils are not detected simultaneously in the experiment, the interaction between the magnetic fields of both can be neglected in the simulations, which are executed using COMSOL Multiphysics Modeling. The coil's inductance detects all magnetic material inside the area spanned by the magnetic field induced by the current applied to the coil. However, since this magnetic field spans broader than the geometric dimensions of the coil, its magnitude has to be adjusted in a way that mainly the magnetic material inside the coil influences the inductance. For that reason, the field ideally has to have a strong gradient in the axial direction of the coil. Fig. 4 plots the magnetic field component in the z-direction normalized by its maximum value over the z-axis of the fluid container. The continuous vertical lines mark the height of the container,

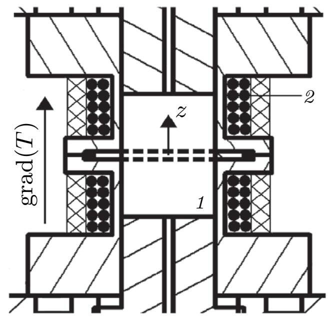

Fig. 3. Fluid container as a separation chamber of the horizontal thermodiffusion cell: *1* – fluid container, *2* – sensor coil.

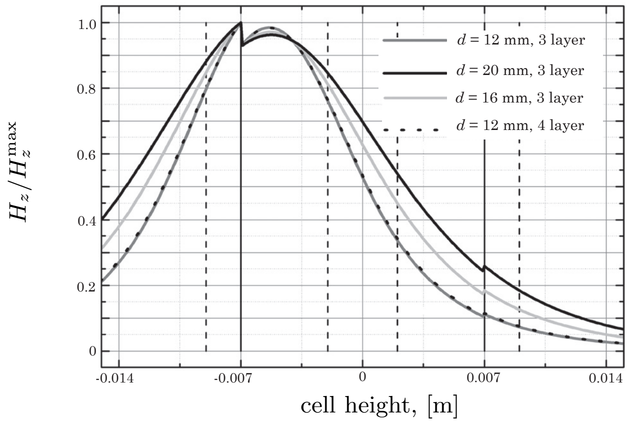

Fig. 4. Axial component of the magnetic field strength along the axis of the fluid container for different coil diameters and number of coil layers. Solid vertical line: position of the fluid container; dashed vertical line: position of the coils.

the dashed vertical lines mark the position of the two sensor coils. It can be concluded that a small coil's diameter, if compared to a larger one, leads to a more distinct peak descending faster towards the ends of the coil. An additional layer results in an increase of the absolute magnetic field strength, but is indifferent regarding the shape of the plotted normalized component. Since the coil has to be winded by hand, this fact then restricts the number of layers possible. This is why the coils are chosen with a diameter of 12 mm and four layers of a 0.2 mm copper wire.

The inductance signal measured then provides the time-dependent change in concentration difference between the lower and the upper part of the fluid volume, which leads to the so-called separation curve. For the amount of fluid used in these experiments as well as for the chosen temperature gradient 1 K/mm and the total separation time of approximately 72 hours, the separation of the fluid will still be in a linear regime described by

$$\frac{c_{\mathrm{lo}} - c_{\mathrm{up}}}{c_0} = \frac{4}{h - h_{\mathrm{grid}}} \frac{\mathrm{S_T} D \triangle T\, t}{h} \tag{7}$$

as suggested by [4], c_{lo} denotes the mass concentration of magnetic material in the upper (lower) fluid chamber, c_0 is the homogenous initial concentration, h denoting the cell's height, h_{grid} is the distance of the double layer grid in the cell's centerpart, $\mathrm{S_T}$ is the Soret coefficient, D is the molecular diffusion coefficient, $\triangle T$

denotes the temperature difference at the cell's boundaries, and t denotes time. Since the molecular diffusion coefficient for the ferrofluid used in the experiments could not be measured so far, it is calculated via the Batchelor relation [9]

$$D = \frac{k_{\mathrm{B}}T}{6\pi(r+s)\eta}(1 + 1.45\varphi_{\mathrm{c}}), \tag{8}$$

with k_{B} being the Boltzmann constant, φ_{c} is the particle volume concentration, η denoting the viscosity of the carrier liquid, and $r+s$ is the sum of the magnetic particles' radius, taking the size distribution in consideration, and the thickness of the surfactant layer. Since the dependence of the molecular diffusion coefficient on the applied magnetic field cannot be determined in the experiments so far for the certain ferrofluid used, the coefficient will be held constant.

3.2. Experimental results. The experiments were carried out using the fluid EMG905 from Ferrotec. Its particle volume concentration has been determined to 8.7 vol.% by VSM measurements with an average particle diameter of approximately 10.4 nm. The fluid viscosity was measured by an Anton Paar rheometer and has been determined to 0.011 Pas at 298 K. In zero magnetic field, the molecular diffusion coefficient assumed for this fluid is then $3.9 \cdot 10^{-12}\,\mathrm{m}^2/\mathrm{s}$. In all experiments, the fluid was exposed to a temperature gradient of 1 K/mm. When applying the magnetic field, two different alignments have been considered. First, the magnetic field is directed parallel to the temperature gradient, second, the field is directed perpendicular to it. For each direction, three different field strengths were applied, in particular, 40 kA/m, 100 kA/m, and 320 kA/m. The separation curves measured for these setups are presented in Figs. 5 and 6. Independent of the positioning of the magnetic field, an increase in field strength leads to a hindrance of the separation process represented by a decline of the separation curve's inclination. A further increase in magnetic field strength leads to a change in separation direction, i.e. an enhancement of the transport recognized by a growing negative inclination.

Considering the separation curve, the temperature gradient and the geometric dimensions of the cell, the Soret coefficient can be calculated from Eq. (7) to 0.169 1/K in zero magnetic field, to 0.167 1/K, –0.061 1/K and –0.152 1/K in the parallel and to 0.084 1/K, –0.512 1/K and –0.257 1/K in the perpendicular case. The change in coefficients sign can be detected.

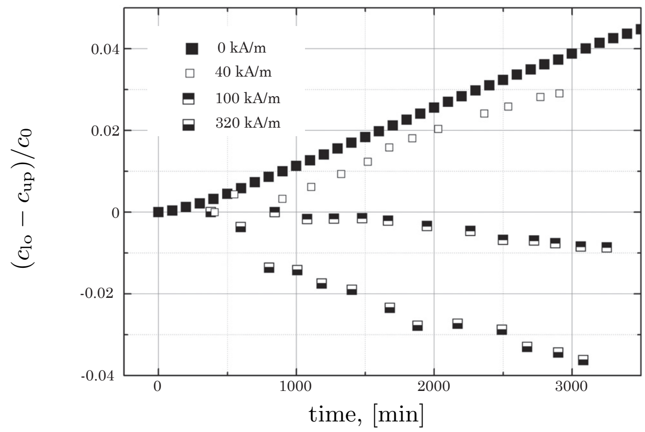

Fig. 5. Separation curve for the parallel positioning of the magnetic field to the temperature gradient of 1 K/mm.

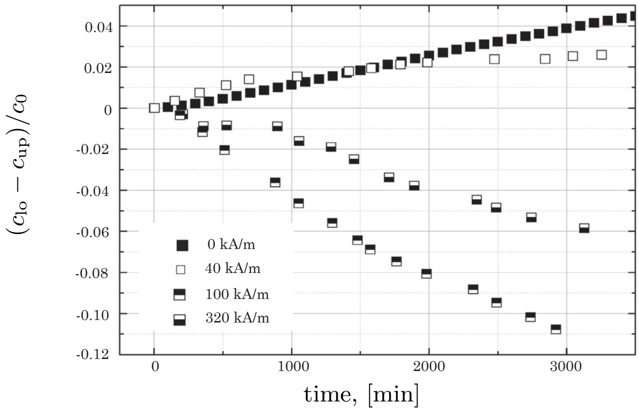

Fig. 6. Separation curve for the perpendicular positioning of the magnetic field to a temperature gradient of 1 K/mm.

4. Results and discussion. From the linear stability analysis, a general dependence of the critical Rayleigh number on the Soret coefficient can be derived. Convective motion is enhanced by $S_T > 0$, hindered in the interval $0 > S_T > -0.001\,1/K$ and even suppressed at $-0.001\,1/K > S_T$. The experimental data give an overview of the magnetic Soret coefficient. The variation of the value from small positive (0.16 1/K) to small or large negative (–0.5 1/K) proves that the suppression of convection due to thermal diffusion is possible and motivates further detailed investigations on the impact of thermodiffusion on convective motion.

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