Thermodiffusive effects in magnetic fluids: application of the ferrofluid dynamics theory

Adrian Lange

Chair Magnetofluiddynamics, Institute of Fluid Mechanics, Technische Universität Dresden,
01062 Dresden, Germany

E-mail: `adrian.lange@tu-dresden.de`

Abstract. The ferrofluid dynamics theory is applied to thermodiffusive problems in magnetic fluids in the presence of magnetic fields. The most general expression of the mass flux are given. By employing these results to experiments, global Soret coefficients in agreement with measurements are determined. Also an estimate for so far unknown transport coefficient is made.

1. Introduction

The thermodiffusive or Soret effect describes the establishment of concentrations gradients in response to temperature gradients for a two- or multi-component fluid. Since the motion of the ferromagnetic nanoparticles in magnetic fluids (MFs) can be influenced by external magnetic fields, the Soret effect in MFs shows a strong dependence on any nonzero magnetic field strength [1–3]. The changes of the Soret coefficient can be up to six times its zero field value [2] and even a change of its sign was measured [2; 3]. Previous theoretical approaches [4; 5] need as input the magnetophoretic velocity of the nanoparticles. Therefore certain microscopic properties are assumed as e.g. spherical particles of equal size and the applicability of the Stokes hydrodynamic drag. A comparison with the experimental results show great differences: the microscopic approach gives changes of the Soret coefficient which are three orders of magnitude smaller than those in the experiment. That means that concerning thermodiffusive processes in MFs there is a wide gap between experiment and theory. Therefore as an alternative approach, a macroscopic theory, called ferrofluid dynamics (FFD) theory, is tested to describe the measured phenomena.

2. Ferrofluid dynamics theory

The principal structure of the ferrofluid dynamics theory was laid down in [6]. It is "... a general, strictly macroscopic approach relying solely on symmetry considerations, conservation laws, and thermodynamics." [7]. An essential part of the theory is the set of material-dependent parameters like susceptibilities and transport coefficients. They can be determined by well design experiments [8] and used here as fit parameters to describe thermodiffusive processes in magnetic fluids under the influence of magnetic fields.

As usual in thermodynamic theories, one starts with the thermodynamic energy density u. It is taken as a function of the entropy density s, the density $\rho^{(1)}$ of the magnetic part of the fluid, the momentum density $\mathbf{g} = \rho\mathbf{v}$, the total density ρ, and the magnetic induction $\mathbf{B} = \mu_0(\mathbf{M}+\mathbf{H})$

[6],

$$du = Tds + \tilde{\mu}_c d\rho^{(1)} + v_i dg_i + \mu^{(2)} d\rho + H_i dB_i \ , \tag{1}$$

where $\tilde{\mu}_c = \tilde{\mu}^{(1)} - \tilde{\mu}^{(2)}$ is the difference in the chemical potentials of the two constituents and $\rho = \rho^{(1)} + \rho^{(2)} = \phi\rho_{\rm m} + (1-\phi)\rho_{\rm cl}$. The density of the magnetic particles (carrier liquid) is denoted by $\rho_{\rm m}$ ($\rho_{\rm cl}$) and ϕ is the volume fraction of magnetic particles in the fluid. It is assumed that the magnetic part of the chemical potential can be separated, $\tilde{\mu}_c = \mu_c(\rho, \rho^{(1)}, T, \mathbf{v}) + \mu_c^m(\rho, \rho^{(1)}, T, \mathbf{v}, \mathbf{H})$. This assumption guarantees a nonzero chemical potential for zero magnetic field and is confirmed by calculations for MFs, where the magnetic part contributes additively to the total chemical potential [9]. The nonmagnetic part is given in [10]. The dissipative mass flux $\mathbf{j}^D$ is proportional to the gradients of the the temperature and the chemical potential,

$$j_i^D = \xi_1 \nabla_i T + \xi \nabla_i \tilde{\mu}_c + \xi_{\|} M_i M_j \nabla_j \tilde{\mu}_c + \xi_{\times} \varepsilon_{ijk} M_j \nabla_k \tilde{\mu}_c \ . \tag{2}$$

Whereas the first two terms characterize isotropic mass fluxes, the last two terms describe anisotropic mass fluxes, namely parallel and perpendicular to the direction of $\mathbf{M}$. The first two transport coefficients, ξ_1, ξ, are related to the classical equation of diffusion including the Soret effect (see below). The last two coefficients, $\xi_{\|}$, $\xi_{\times}$, reflect that the magnetic field may influence the mass flux for any nonzero strength. Starting with these equations, one gets an analytical result for the magnetic part of the chemical potential,

$$\mu_c^m = -\mu_0 \int_0^H \frac{\partial M}{\partial \rho^{(1)}} dH' \ . \tag{3}$$

It allows a direct determination of μ_c^m if the magnetization $M(H, \rho^{(1)}, T)$ is known without any assumption about the properties of the MF in contrast to [9; 11], where the knowledge of the volume concentration of the nanoparticles [11] or the strength of the magnetodipole interaction [9] is needed. One gets also a general expression for the mass flux,

$$\begin{aligned} \frac{\mathbf{j}^D}{\rho} &= \left(\frac{\xi_1}{\rho} + \frac{\xi}{\rho}\frac{\partial \mu_c}{\partial T}\right)\nabla T + \frac{\xi}{\rho}\frac{\partial \mu_c^m}{\partial T}\nabla T + \frac{\partial \tilde{\mu}_c}{\partial T}\left[\frac{\xi_{\|}}{\rho}\mathbf{M}\left(\mathbf{M}\nabla T\right) + \frac{\xi_{\times}}{\rho}\left(\mathbf{M}\times\nabla T\right)\right] \\ &\quad + \xi\frac{\partial \mu_c}{\partial \rho^{(1)}}\nabla c_1 + \xi\frac{\partial \mu_c^m}{\partial \rho^{(1)}}\nabla c_1 + \frac{\partial \tilde{\mu}_c}{\partial \rho^{(1)}}\left[\xi_{\|}\mathbf{M}\left(\mathbf{M}\nabla c_1\right) + \xi_{\times}\left(\mathbf{M}\times\nabla c_1\right)\right] \\ &\quad - \frac{\mu_0}{\rho}\frac{\partial M}{\partial \rho^{(1)}}\left[\xi\nabla H + \xi_{\|}\mathbf{M}\left(\mathbf{M}\nabla H\right) + \xi_{\times}\left(\mathbf{M}\times\nabla H\right)\right] \ , \end{aligned} \tag{4}$$

which contains terms proportional to thermophoresis ($\sim \nabla T$), to diffusiophoresis ($\sim \nabla c_1$), and to magnetophoresis ($\sim \nabla H$). For zero magnetic field, Eq. (4) reduces to the classical result (see Eq. (227), Chapt. XI in [10])

$$\frac{\mathbf{j}^D}{\rho} = \left(\frac{\xi_1}{\rho} + \frac{\xi}{\rho}\frac{\partial \mu_c}{\partial T}\right)\nabla T + \xi\frac{\partial \mu_c}{\partial \rho^{(1)}}\nabla c_1 = c_1 c_2 D_T \nabla T + D_c \nabla c_1 \tag{5}$$

with (D_T) D_c the (thermal) diffusion coefficient and $c_2 = 1 - c_1$. The concentration of the magnetic particles $c_1 = \rho^{(1)}/\rho$ is defined by the mass fraction of the total density ρ.

3. Application to experiments

According to the experiments [3], a horizontal unbounded layer of MF of thickness h sandwiched between two plates is considered, where the lower one is cooled to T_1 and the upper one is heated to $T_2 > T_1$ (Fig. 1). These boundary conditions together with the equation of head conduction yield the temperature profile of the quiescent conductive state, $T(z) = T_0 + (T_2 - T_1)(z/h)$, with $T_0 = (T_1 + T_2)/2$. The diffusion equation, $\partial c_1/\partial t = \operatorname{div}\left(\mathbf{j}^D/\rho\right)$, and the boundary condition for impenetrable plates, $j_z^D(z = \pm h/2) = 0$, in rearranged form,

$$\frac{-h}{c_1 c_2 (T_2 - T_1)} \left.\frac{\partial c_1}{\partial z}\right|_{z=\pm h/2} = \frac{D_T}{D_c} = S_T \ , \tag{6}$$

allow the determination of the Soret coefficient in the zero field case. In the same way the global Soret coefficient in the presence of a magnetic field can be determined by using Eq. (4). Depending on the orientation of the magnetic field $\mathbf{H}$ to the temperature gradient ∇T (see Fig. 1), the global Soret coefficients $S_T^{\|}$ and $S_T^{\perp}$ are functions of the material parameters D_c, ξ, $\xi_{\|}$, and $\xi_{\perp}$ if $M(H)$, μ_c, S_T, and $\rho^{(1)}$ is known [12].

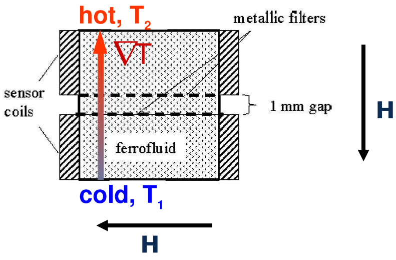

Figure 1. Sketch of the experimental setup with the vertical temperature gradient ∇T inside the ferrofluid layer and the external field $\mathbf{H}$ which is either parallel or perpendicular to ∇T. Courtesy of [3].

To fit $S_T^{\|}$ to the experimental data, the combined quantities ξ/D_c and $\xi_{\|}/D_c$ are used as fit parameters, since $S_T = 0.15\ \mathrm{K}^{-1}$ was measured in the zero field case [3] but not D_c. The blue solid line in Fig. 2 gives the best two parameter fit, yielding $\xi/D_c = 8.2\ \mathrm{kg\,s^2/m^5}$ and $\xi_{\|}/D_c = -1.41 \cdot 10^{-7}\ \mathrm{kg\,s^2/(m^3\,A^2)}$. To underline the relevance of $\xi_{\|}/D_c$ even for small fields, the cyan dot-dashed line in Fig. 2 displays $S_T^{\|}$ for $\xi_{\|}/D_c = 0$ and all other parameters as before. Now the theoretical curve misses the measured data (□) clearly. That underlines that the transport coefficients of the anisotropic terms in Eq. (2) are necessary and nonzero for *all* magnetic strengths.

In contrast to the parallel setup, in the perpendicular setup the following coarse approximation is made: $(\partial c_1/\partial y)_{z=\pm h/2}$ shall be a constant C for all H-values tested here. Then the global Soret coefficient $S_T^{\perp}$ depends solely on $F = (\xi_{\perp} C)/D_c$ since ξ/D_c was determined in the parallel setup. The best fit yields $F = 3.75 \cdot 10^{-2}\ \mathrm{kg\,s^2/(m^5 A)}$. The inferior match with the experimental data (see △ and blue dashed line in Fig. 2) in comparison with the parallel setup is due to the approximation that $(\partial c_1/\partial y)_{z=\pm h/2}$ is constant. In the real system it will depend on the magnetic field since the solution for c_1 depends on the magnetic field.

4. Conclusion

The ferrofluid dynamics theory is applied to thermodiffusive problems in magnetic fluids in the presence of magnetic fields. A general expression for the mass flux is given which is independent of the fluid properties, temperature distribution and assumptions about the concentration of the nanoparticles. Applying these results to the experiments [3], it is shown that for thermodiffusive problems in magnetic fluids anisotropic mass fluxes are relevant and no small contributions for any nonzero strengths of the magnetic field. To elucidate this insight, more well designed experiments and further theoretical as well as numerical analyses are needed.

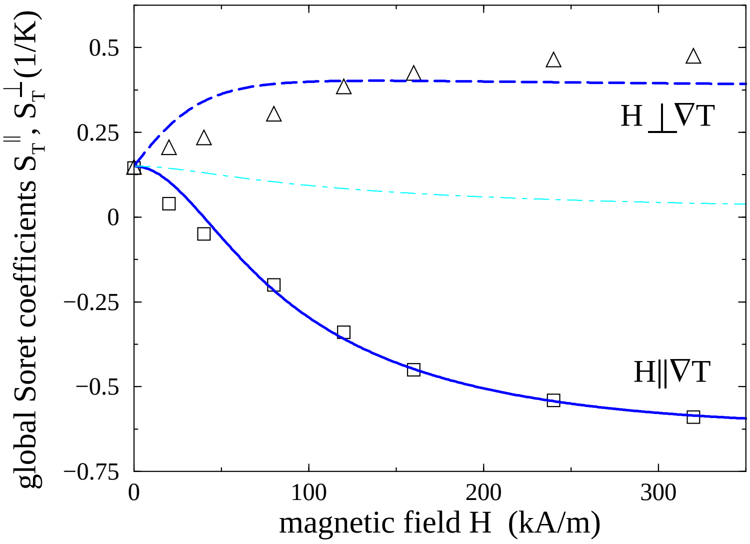

Figure 2. Global Soret coefficients $S_T^{\|}$ and $S_T^{\perp}$ against the magnetic field strength. The blue solid line shows the best fit of $S_T^{\|}$ with $\xi/D_c = 8.2$ kg s^2/m^5 and $\xi_{\|}/D_c = -1.41 \cdot 10^{-7}$ kg s^2/(m^3 A^2) to the experimental data (□). The cyan dot-dashed line displays $S_T^{\|}$ for the same parameters but $\xi_{\|}/D_c = 0$. The blue dashed line indicates the best fit of $S_T^{\perp}$ with $\xi/D_c = 8.2$ kg s^2/m^5 and $F = 3.75 \cdot 10^{-2}$ kg s^2/(m^{5}A) to the experimental data (△) taken from [3].

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