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

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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 concentration 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 $g = \rho v$, the total density $\rho$, and the magnetic induction $B = \mu_0(M + H)$. 
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])

\[
\frac{\mathbf{j}^D}{\rho} = \left( \frac{\xi}{\rho} + \frac{\xi \partial \mu_c}{\rho \partial T} \right) \nabla T + \xi \frac{\partial \mu_c}{\rho \partial T} \nabla c_1 = c_1 c_2 D_T \nabla T + D_c \nabla c_1
\]

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 $\rho$.

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 heat 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, \( \partial c_1/\partial t = \text{div}(j^D/\rho) \), and the boundary condition for impenetrable plates, \( j^D_z(z = \pm h/2) = 0 \), in rearranged form,

\[
\frac{-h}{c_1 c_2 (T_2 - T_1)} \frac{\partial c_1}{\partial z} \bigg|_{z = \pm h/2} = \frac{D_T}{D_c} = S_T ,
\]

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 \( \nabla T \) (see Fig. 1), the global Soret coefficients \( S^\parallel_T \) and \( S^\perp_T \) are functions of the material parameters \( D_c, \xi, \xi^\parallel, \text{and} \xi^\perp \) if \( M(H), \mu, ST, \text{and} \rho^{(1)} \) is known [12].

To fit \( S^\parallel_T \) to the experimental data, the combined quantities \( \xi/D_c \) and \( \xi^\parallel/D_c \) are used as fit parameters, since \( S_T = 0.15 \text{ K}^{-1} \) 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 \( \xi/D_c = 8.2 \text{ kg s}^2/\text{m}^4 \) and \( \xi^\parallel/D_c = -1.41 \cdot 10^{-2} \text{ kg s}^2/(\text{m}^3 \text{A}^2) \). To underline the relevance of \( \xi^\parallel/D_c \) even for small fields, the cyan dot-dashed line in Fig. 2 displays \( S^\parallel_T \) for \( \xi^\parallel/D_c = 0 \) 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 \( S^\perp_T \) depends solely on \( F = (\xi^\perp C)/D_c \) since \( \xi/D_c \) was determined in the parallel setup. The best fit yields \( F = 3.75 \cdot 10^{-2} \text{ kg s}^2/(\text{m}^3 \text{A}) \). 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 $S^\parallel_T$ and $S^\perp_T$ against the magnetic field strength. The blue solid line shows the best fit of $S^\parallel_T$ with $\xi/D_c = 8.2$ kg s$^2$/m$^3$ and $\xi^\parallel/D_c = -1.41 \cdot 10^{-7}$ kg s$^2$/m$^3$ A$^2$ to the experimental data (□). The cyan dot-dashed line displays $S^\parallel_T$ for the same parameters but $\xi^\parallel/D_c = 0$. The blue dashed line indicates the best fit of $S^\perp_T$ with $\xi/D_c = 8.2$ kg s$^2$/m$^3$ and $F = 3.75 \cdot 10^{-2}$ kg s$^2$/m$^3$A to the experimental data (△) taken from [3].

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