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Thermal conductivity measurements on ferrofluids with special reference to measuring arrangement

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ABSTRACT

Material properties like viscosity and sound propagation in colloidal suspensions of magnetic nanoparticles, so-called ferrofluids, are known to depend on external magnetic fields due to structure formation of the magnetic particles. In this experimental study we investigate the effect of magnetically driven structure formation on heat flux in ferrofluids on the basis of thermal conductivity measurements in variation of an external magnetic field. Therefore an improved measuring device based on the plane heat source instead of the standard hot wire method is used to enable both parallel and perpendicular orientation of magnetic field and heat flux. Thermal conductivity measurements are carried out in variation of strength and direction of an external magnetic field relative to heat flux. Unlike former experimental investigations for the first time the results show qualitative consistency with theoretical predictions for both orientations.

1. Introduction

1.1. Thermodynamics and theoretical analysis

Ferrofluids are colloidal suspensions of magnetic nanoparticles and may be considered a subclass of nanofluids. Nanofluid is a comprehensive term for colloidal suspensions of particles in the nanoscale range [1]. Usually the particle size varies from 1 nm to 100 nm in diameter [2]. There are no restrictions on the suspension's carrier liquid or the dispersed particles. Accordingly, the material properties are highly variable, so nanofluids cannot be termed a class of substances.

The thermal conductivity of a suspension differs from the thermal conductivity of its carrier liquid due, first of all, to the higher thermal conductivity of the colloidal particle fraction. The effective medium theory, particularly the Maxwell Garnett Equation [3], which is derived from Maxwell et al. [4] and applied analogously to several material properties such as electric conductivity and dielectric constant, describes the effective macroscopic properties of a composite material as a function of the particle fraction and the material properties of the components. Results of thermal conductivity measurements on nanofluids diverge from the effective medium theory very often. An overview of possible reasons and explanations is given by Vadasz et al. [5] and Keblinski et al. [2]. One possible reason for the discrepancies between effective medium theory and experimental results is interparticle interaction, which can result in the formation of structures, such as clusters or chains [6]. These structures may have the effect of heat bridges. The form and magnitude of the structures formed varies and depends not only on the material of the carrier medium and the particles, but also on the shape and size of the particles [6,7].

In ferrofluids, the interparticle interaction has an even more important impact on the fluid's properties, since the magnetic dipole interaction forces active agglomeration of the particles, which can be controlled by applied magnetic fields. The effect of this interaction can be seen, for example, in changes in the viscosity of ferrofluids [8], which depends on interparticle interaction and differs depending on the material of the components and the size and shape of the particles [9].

To avoid irreversible particle agglomeration due to van der Waals interaction in ferrofluids, the magnetic particles are often synthesized with a surfactant. Hence, the particles are suspended colloidally and homogeneously in the liquid in the absence of a magnetic field [10]. Hence, structure formation in ferrofluids is constrained to the presence of a magnetic field and depends on particle size and shape, as well. Thus, in ferrofluids, the impact of particle interaction on material properties such as thermal conductivity can be investigated separately by varying the magnetic field.

Blums et al. [11,12] predict anisotropy of thermal conductivity in ferrofluids in the presence of a magnetic field. This is explained by the formation of chain-like structures in ferrofluids due to particle interaction. For parallel alignment of the magnetic field and heat flux, an increase in thermal conductivity is predicted, and a decrease for perpendicular alignment.

Magnetically-induced structure formation only arises if the magnetic energy of the particles is larger than their thermal energy. Rosensweig [13] describes this ratio as the interparticle interaction

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parameter, which is given as $\lambda = \mu_0 M^2 V/(24 k_B T)$. However, since most ferrofluids are synthesized with a surfactant surrounding the magnetic particles, the magnetic components cannot be in contact, and thus, the magnetic forces have to be strong enough to bridge the surfactant layer. Thurm et al. [14] apply a term which takes into account the thickness s of the surfactant. One can calculate the modified interparticle interaction parameter as:

$$\lambda^* = \frac{\mu_0 M_{sp}^2 V}{24 k_B T}\left(\frac{d}{d+2s}\right)^3 \tag{1}$$

For $\lambda^* < 1$, the thermal energy is dominant and no particle interaction occurs in a magnetic field. For $\lambda^* > 1$, the magnetic energy is stronger than the thermal motion of the particles, and structures can form in magnetic fields. Fig. 1 shows the modified interparticle interaction parameter as a function of the particle diameter and the thickness of the surfactant.

We assume that the thermal conductivity of ferrofluids is influenced by homogeneous, external magnetic fields if the modified interparticle interaction parameter is $\lambda^* > 1$.

According to Rosensweig [13], the thermodynamic properties of ferrofluids show another peculiarity as well. The magnetocaloric effect is described as the increase in a magnetic substance's temperature when it is exposed to a magnetic field and the decrease when the magnetic field is removed. Rosensweig derives the effect of a magnetic field on the specific heat capacity c_p as:

$$\left.\frac{\partial c}{\partial H}\right|_T = \mu_0 T \left.\frac{\partial^2 M}{\partial T^2}\right|_H \tag{2}$$

This equation shows that the change in specific heat capacity due to a magnetic field depends on the material, by the temperature T, and the pyromagnetic coefficient $\partial M/\partial T$. We obtained the pyromagnetic coefficient measuring the magnetization curve for temperature values of $260\ \mathrm{K} \leq T \leq 380\ \mathrm{K}$ in steps of $\Delta T = 10$ K. One can calculate $\partial c/\partial H$ for the magnetite-based commercial ferrofluid APG513A at laboratory conditions ($T = 293.15$ K; $H = 25$ kA/m; $\partial^2 M/\partial T^2 = -355$ A/(m K)) $\partial c/\partial H|_T = 0.45$ J/(kg K) which is a change of -0.3% of $c_0 = 1510$ J/(kg K). Thus, the magneto-caloric effect can be neglected for the investigation if thermal conductivity of ferrofluids is influenced by magnetic fields.

1.2. State of the art

The first published studies on magnetic field-dependent thermal conductivity of ferrofluids are from Kronkalns [15] and Blums et al. [16]. In these efforts, no effect of the magnetic field on thermal conductivity was found in magnetite liquids stabilized with oleic acid in kerosene. Popplewell et al. [17] carried out measurements with four kinds of liquids: diester, hydrocarbon, water and fluorocarbon-based ferrofluids with suspended magnetite particles. No explanation concerning a surfactant is given. In addition, no influence of the magnetic field on the thermal conductivity was measured. Particle size distributions were not stated for the liquids used in [15–17].

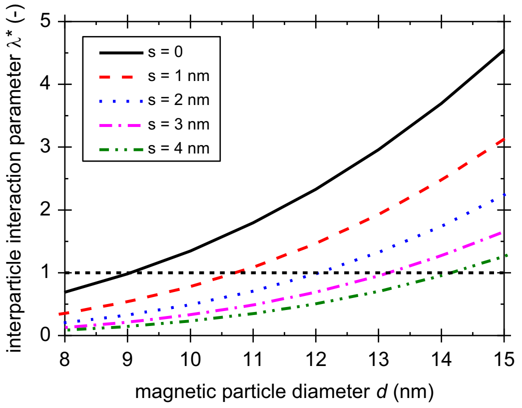

Fig. 1. Diagram of the interparticle interaction parameter as function of the spherical magnetic particle diameter d and the thickness s of the surfactant.

Recent experimental studies on thermal conductivity from Li et al. [18], Djurek et al. [19] and Philip et al. [20–23] show different dependencies on an applied external magnetic field. The investigation of Li et al. [18] was performed with water-based ferrofluids with iron particles and a dodecylbenzenesulfonate surfactant. The particle size distribution is given as log-normal with a mean diameter of $d = 26$ nm. The ferrofluid was used with various volume fractions from one to five percent. The time until sedimentation takes place is given, ranging from a few hours to one week. For a magnetic field aligned perpendicular to the heat flux, no effect on thermal conductivity was measured. However, for parallel alignment, an increase in thermal conductivity of 11% for $1\%_{\mathrm{vol}}$ and 25% for $5\%_{\mathrm{vol}}$ was observed for a magnetic field strength of 19 kA/m.

Djurek et al. [19] used water and n-decane based ferrofluids with maghemite or cobalt ferrite particles stabilized with oleic acid or ethylene glycol. There is no particle size distribution given. The alignment of the magnetic field with respect to heat flux is unclear. However, the observed decrease of thermal conductivity is between 5 and 50%.

Philip et al. [20–23] investigated kerosene-based ferrofluids with suspended magnetite particles of mean diameter 6.7 nm stabilized with oleic acid. The particle size distribution is not precise because it is shown in a logarithmic diagram. But it may be interpreted that there are some volume per cent of magnetic material with a size larger than 13 nm. The liquid was studied at various volume fractions from 0.03% to 6.3%. For a perpendicular alignment of magnetic field and heat flux, no effect on thermal conductivity was found. However, for parallel alignment, an increase in thermal conductivity was discovered. For a volume fraction of 0.03% the observed increase of thermal conductivity is 50% at a magnetic field level of $H = 30$–35 kA/m. For a volume fraction of 6.3% the increase is 300% at $H = 7$ kA/m field strength.

From this review, it is evident that experimental results have been heterogeneous, and that none of them agrees with the theoretical prediction completely.

2. Experimental setup and material

2.1. Thermodynamics of thermal conductivity measurements

Heat conduction in homogeneous media is described by Fourier's law (Eq. 3). Herein thermal conductivity k occurs as a linear coefficient connecting heat flux $\vec{q}$ to the temperature gradient grad T

$$\vec{q} = -k \operatorname{grad} T \tag{3}$$

To measure thermal conductivity, either the heat flux or the temperature gradient, must be given as a boundary condition and the other has to be measured to calculate thermal conductivity.

The temporal and spatial temperature distribution for heat conduction is described by Fourier's differential equation of temperature distribution. For homogeneous and isotropic media it can be written as:

$$\frac{\partial T(\vec{x}, t)}{\partial t} = \frac{k}{\rho c_p} \operatorname{div}(\operatorname{grad} T) \tag{4}$$

where ρ is the density and c_p is the specific heat capacity of the medium. Although ferrofluids are inhomogeneous at a mesoscopic