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Aleksandra A. Bozhko
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Convection in Ferro-Nanofluids: Experiments and Theory

Physical Mechanisms, Flow Patterns, and Heat Transfer

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Physical Mechanisms, Flow Patterns, and Heat Transfer

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*To the memory of Professor G. F. Putin,
outstanding experimentalist, colleague and
teacher*

Preface

Scope

This book is based on the results of experimental and theoretical studies of hydrodynamic stability and heat and mass transfer processes in ferrofluids that we have been involved with over the past several decades. The main motivation for such studies has been the growing interest in the use of magnetically controllable media as heat carriers in various thermal management systems. Along with other non-gravitational mechanisms capable of inducing the motion of initially quiescent fluid such as vibrational and electroconvection thermomagnetic convection can be used to enhance heat transfer in conditions where natural convection is impossible, for example, in microgravitation conditions. The experimental investigation that is reported in this book was conducted at Perm State University, Russia, under the license from the Russian State Corporation for Space Research (RosCosmos) and formed the ground-based component of a larger programme involving experiments on board autonomous and piloted spacecrafts, the orbital station "Mir" and the International Space Station. From a fundamental point of view the book considers an intricate interaction of non-isothermal and electrically nonconducting magnetopolarisable fluid with gravitational and magnetic fields. In the absence of a magnetic field ferrofluids behave similarly to other nonmagnetic nanofluids, studies of which have been growing exponentially over the past two decades due to their ever expanding applications in modern technology.

Audience

We hope that this book will be of interest to researchers and practitioners working in the areas of fluid mechanics, hydrodynamic stability and heat and mass transfer with the view of perspective applications of ferrofluids in heat management systems, in particular, in microelectronics and space technologies. The main emphasis of the book is on the influence of a uniform magnetic field on flows of non-isothermal ferrofluids and the associated heat transfer. However, we also discuss peculiar features of ferrofluid flows occurring in the absence of a magnetic field, which are shown to be drastically different from those of ordinary fluids and need to be taken into account by practitioners working with magnetic and non-magnetic nanofluids.

Content

Invention of ferrofluids, their industrial synthesis and numerous studies at micro and macro levels have been primarily motivated by their magnetic properties that are many orders of magnitude stronger than those of natural paramagnetic and diamagnetic fluids and gases. The composition of ferrofluids that defines their magnetic properties and the related mechanisms of heat and mass transfer in them are briefly reviewed in Chapter 1.

The main equations describing motion of non-isothermal ferrofluids by treating them as magnetopolarisable continuous media are summarised in Chapter 2. While such a description has its limitations that become evident when the theoretically obtained results are compared with those of experimental observations, currently, such an approximation offers the most robust way of modelling ferrofluid flows. The reasons for this are outlined in the subsequent chapters of the book. The major governing non-dimensional parameters are also defined and their physical meaning is discussed in Chapter 2.

Results of a theoretical analysis of thermomagnetic convection in geometrically simple yet practically relevant domains are presented in Chapter 3. Such an analysis sheds light on physical processes taking place in the bulk of ferrofluid offering the insight that is successfully used to guide experimental observations and measurements. In particular, the existence of thermomagnetic waves associated with the thermally induced non-uniformity of fluid magnetisation and of oscillatory regimes of convections caused by the nonlinear variation of magnetisation across a ferrofluid layer was discovered theoretically first and then was confirmed in specialised experiments. A comprehensive analysis of magnetoconvection arising in the arbitrarily oriented magnetic field in gravity-free conditions is another example of a practically important situation considered in Chapter 3 that is out of reach for ground-based laboratory experiments.

Chapter 4 contains a detailed description of experimental setups specifically designed for a comprehensive study of buoyancy and thermomagnetically driven ferrofluid flows. The details distinguishing experimental chambers and flow visualisation and heat flux measurement techniques used for working with magnetically active media from those used in experiments with non-magnetic fluids are emphasised. In particular, it is shown that the shape and size of the working chamber have a defining influence on the type of convection patterns arising in a magnetic field.

The features of thermogravitational and thermomagnetic convection arising in finite flat layers and spherical cavities filled with ferrofluids and placed in uniform gravitational and magnetic fields are detailed in Chapters 5 and 6, respectively. Notably, a strong influence of gravitational sedimentation of solid particles and their aggregates contained in ferrofluids is demonstrated experimentally. It changes qualitatively the character of convection compared to that observed in ordinary single-phase fluids. Specifically, it is shown in Chapter 5 that in the vicinity of convection threshold in ferrofluids flows become oscillatory and chaotic both in space and time. A hysteresis is observed when the onset of convection in the initially density-stratified ferrocolloid is delayed compared to that recorded for the same but pre-mixed fluid. Convection is found to arise and decay spontaneously and irregularly and this found to be related to the concentration of solid phase in experimental fluids.

The influence of magnetic fields of various orientations on ferrofluid convection and heat transfer is discussed next in Chapter 6. It is shown that such an influence is not monotonic. Depending on the values of the governing gravitational and magnetic parameters, the application of magnetic field can either enhance or suppress convection drastically changing the observed flows and offering a not-intrusive means of controlling them. The experimental evidence of the fact that conditions of a particular laboratory run, storage and past usage of a ferrofluid strongly affect its flows and performance as a heat carrier. These factors should be taken into account when interpreting physical observations of a non-isothermal ferrofluid behaviour and when using it in practical applications. Overall, the book is intended to provide a guidance to a very rich and frequently ambiguous behaviour of non-uniformly heated ferrocolloids caused by their complex composition and influenced by an external magnetic field.

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This book would not be possible without the hard work of our colleagues and technical staff who were invaluable in building experimental equipment and maintaining it in working order over many months during which individual experimental runs were performed and over decades during which this research was conducted. We are especially grateful to our students T. Pilugina, D. Shupeinik, P. Bulychev, A. Sidorov, M. Krauzina, P. Krauzin, H. Rah-

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Perm, Russia
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April 2018

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Acronyms

CTO	Centrifugally purified transformer oil
FF	Ferrofluid
FF-PES	Ferrofluid based on polyethylsiloxane
FF-TO	Ferrofluid based on transformer oil
GCC	Gravi-concentrational convection
GS	Gravitational sedimentation
MCC	Magneto-concentrational convection
MPH	Magnetophoresis
TD	Thermodiffusion
TGC	Thermogravitational convection
TMC	Thermomagnetic convection
TO	Transformer oil

Chapter 1
Ferrofluids: Composition and Physical Processes

Abstract A brief history and an overview of the current state of knowledge of ferrofluids (also known as ferrocolloids or ferro-nanofluids) are given. Applications of ferrofluids as advanced heat carrier media in heat management systems are emphasised. It is discussed that in the absence of a magnetic field, ferrofluids can be considered as a type of synthesised nanofluids or ordinary colloids. However, when they are placed in an external magnetic field, they behave as magneto-polarisable media, the magnetic susceptibility of which is several orders of magnitude larger than that of natural fluids and gases. Various physical mechanisms of heat and mass transfer in ferrofluids are identified. It is shown that the macroscopic behaviour of ferrofluids is strongly affected by their microstructure that depends on the way they are synthesised, stored and used.

1.1 Brief History and Composition of Ferrofluids

First colloids (termed later as ferrofluids[1]) containing single-domain ferromagnetic particles with a characteristic size of 10 nm suspended in a carrier liquid were synthesised in the 1930s [85]. The interest to their technological applications was significantly boosted in the 1960s when their industrial production became possible [22, 178, 184, 244]. The presence of ferromagnetic nanoparticles with magnetic moments that are 10^3–10^4 times larger than those of ions of paramagnetic materials enables achieving ferrofluid magnetisation of up to $\sim 100\,\mathrm{kA/m}$ using external magnetic fields created by ordi-

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[1] Not to be confused with magnetorheological fluids containing much larger, of the order of a micron, particles.

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nary permanent and electromagnets. When taken out of an external magnetic field, ferrofluids lose their magnetisation due to the disorientation of magnetic moments of individual particles by Brownian motion. Because of this ferrofluids are classified as superparamagnetics [120] and are referred to as magneto-polarisable media. Fluid behaviour of electrically non-conducting ferrocolloids is similar to that of natural dia- and paramagnetic fluids but is characterised by much larger (10^4–10^6 times) magnetic susceptibility and thus magnetic forces. It is studied in a special division of hydrodynamics that is known as ferrohydrodynamics [209]. The main motivating factor for a rapid development of this area of research is a wide range of ferrofluid applications that include vibration damping, magnetic sealing, species separation as well as their use in various sensors, actuators modulators of laser radiation, MEMS and NEMS, and cancer treatment to name a few. Of the main interest in this book is the application of ferrofluids as heat carrier media in thermal management systems, for example, in power transformers and converters and solar collectors. Comprehensive reviews of heat and mass transfer processes taking place in ferrofluids can be found, for example, in [13, 26, 88, 173, 209, 215, 222].

While the main property of ferrofluids that defines their numerous applications is their ability to respond to external magnetic field, the specifics of their composition put them in a larger class of synthetic fluids known as nanofluids—fluids that contain solid particles with sizes ranging from 1 to 100 nm. The term "nanofluid" was coined relatively recently (in 1995) to denote artificial fluids created to drastically improve the performance of traditional heat carrier liquids [64] by adding solid particles. However, the field of colloidal chemistry dealing with fluids containing such small particles existed for a much longer time [268] being motivated by the existence of natural nanomaterials [107] and their use in arts, trades and industry [181, 226]. Therefore ferrofluid research benefits greatly from the knowledge and experience accumulated over decades of studies of other similar media.

Ferrofluids typically contain nanoparticles of cobalt, magnetite, hematite and various ferrites. Nanoparticles are usually obtained by ball-milling macroscopic materials [184] or by chemical precipitation [22, 139]. The latter method is frequently preferred as it results in particles with more uniform sizes. In this method magnetite particles are obtained via a chemical reaction between iron salt solutions and concentrated alcali. The magnetite sediment then is mixed with a surfactant such as oleic acid that prevents the particles from forming aggregates in a carrier fluid. Magnetic properties of such nanoparticles are defined by their size and shape, the type of a crystal lattice and its defects and the interactions of particles with molecules of a carrier liquid [49, 107].

The stability of a magnetic colloid depends on the balance between attraction and repulsion forces acting between the particles. Closely located particles can coagulate under the action of van der Waals forces. The strength of these forces reduces in inverse proportion to the sixth power of the distance between particles. Single-domain magnetic particles also experience the attraction due to magnetic dipole-dipole interaction [47, 222] that decreases with the distance slower than van der Waals forces. However, at room temperature, the dipole-dipole interaction between particles of the size of the order 10 nm is negligible [192, 222]. Electrostatic or steric repulsion is used to prevent particles from forming aggregates. Depending on the stabilisation method, ferrofluids are categorised into two main groups: ionic, where particles are surrounded by an electrically charged shell, or surfacted, where particles are coated with amphiphilic molecules. Magnetite particles are typically surfacted with oleic acid. It has long (1.8 nm) molecules that are bent in the middle due to a double bond and that attach to the particle surface with one end due to adsorption. Long molecular tails extending away from particles create steric repulsion between them.

If the balance between particle repulsion and attraction is broken, they can start forming aggregates. Particle aggregation can also be triggered by the variation of the shape of nanoparticles from spherical [49, 155], by the presence of large particles and of surface coating defects [53, 224, 266] as well as by the increase of the particle concentration [8, 118]. The appearance of aggregates strongly influences magnetic and transport properties of ferrofluids. Depending on the number of participating particles, nano- and microscopic aggregates are distinguished with characteristic sizes of tens of nanometres and microns, respectively [53, 63, 118]. Nano-sized aggregates can be quasi-spherical [53, 118, 142, 228] or chain-like [8, 192, 193]. Microscopic aggregates also known as droplet aggregates are more likely to form in a magnetic field [53, 119, 187], when the fluid is cooled or if the concentration of nanoparticles is increased [61].

Properties of ferrofluids also depend on the choice of a carrier liquid. Most common ferrofluids are based on kerosene, silicon or transformer oil and water. Organic carrier fluids themselves have a complex composition and contain molecules of different weights and sizes as well as contaminants that could lead to the formation of insoluble sediment. Given that ferrofluids also can contain unbound surfactant (i.e. free oleic acid), ferrofluids are essentially multiphase systems. Their composition is schematically shown in Figure 1.1.

Single-domain magnetic particles suspended in ferrofluids can be of spherical (1, 2) or nonspherical (3) shapes with complete (1, 2) or deficient (3) coating. The size of magnetic particles determines the prevailing mechanism of magnetic relaxation (the alignment of magnetic moments of individual particles with the applied magnetic field). For large particles (1 and 3), Brownian mechanism [222] dominates when a particle rotates in the magnetic field as a whole. In small particles (2) Neel's mechanism [166] is preferred when the magnetic moment reorients within a particle. Quasi-spherical aggregates (4) with the size of 40–90 nm are typical for ferrofluids used in the majority of experiments described in this book. They can have both Brownian and Neel's mechanisms of relaxation [53, 194].

At present even more advanced two-phase (particle-fluid) models of ferrofluids [31, 122] are incapable of fully representing their microscopic structure. Therefore, simpler models treating ferrocolloids as monofluids with continuously varying properties are still used for a theoretical description of their macroscopic flows [13], and this approach will be taken in Chapter 3 here.

1.2 Physical Processes Taking Place in Ferrofluids

Despite containing ferrous particles, ferrofluids with organic bases have a low electric conductivity. When they are nonuniformly magnetised, bulk forces can arise that are capable of exciting convective motion of a nongravitational nature as well as forces that prevent such a motion [13]. The former are due to the so-called magnetic buoyancy, while the latter appear as a consequence of magnetic field distortion by the moving magnetised medium. In this context ferrohydrodynamics can be considered as the limit opposite to the inductionless limit in classical magnetohydrodynamics of electrically conducting fluids. The other distinction between magneto- and ferrohydrodynamics is that the main driving mechanism in the former is Lorentz force acting on a conducting fluid moving in a magnetic field, while in the latter it is ponderomotive Kelvin force driving stronger magnetised non-conducting fluid to regions with a stronger magnetic field. The magnetisation of ferrofluids decreases with temperature because of three reasons: Curie effect leads to demagnetisation of individual particles, more intense Brownian motion disorients their magnetic moments and thermal expansion reduces the effective concentration of magnetic phase in the fluid. Because of that ponderomotive force drives stronger magnetised cooler fluid to regions with stronger magnetic field displacing warmer fluid. Such a motion is called thermomagnetic convection. Its mechanism is shown schematically in Figure 1.2 for the cases of nonuniform and uniform external magnetic field.

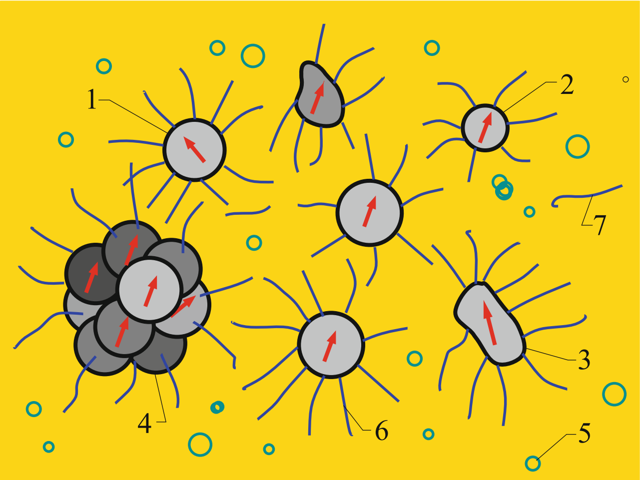

Fig. 1.1 Schematic composition diagram of a magnetic fluid: 1–3, single-domain magnetic particles of different sizes and shapes; 4, quasi-spherical aggregates; 5, molecules of a carrier liquid; 6 and 7, adsorbed and free surfactant, respectively. Arrows show magnetic moments of individual particles.

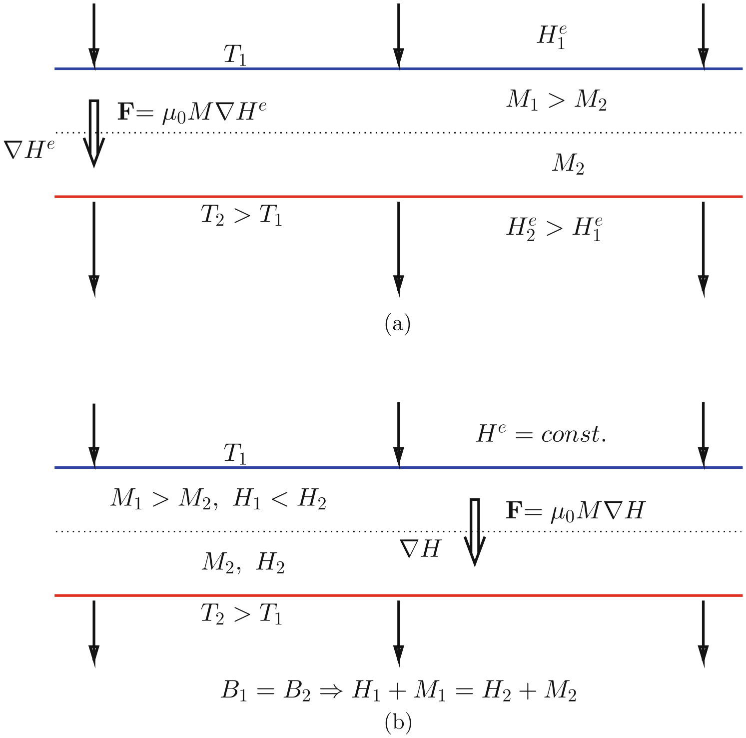

Fig. 1.2 Schematic diagram of forces driving thermomagnetic convection in (a) nonuniform and (b) uniform external magnetic field.

In the case of a nonuniform external magnetic field (such as the one created by a standard bar magnet), the motion of fluid is induced primarily by the gradient of this field (see Figure 1.2(a)). It plays the role of effective gravity, while fluid magnetisation is analogous to fluid density. Cooler fluid that has larger magnetisation (M_1) is drawn to the regions with a larger magnetic field that is in the direction of the external magnetic field gradient (∇H^e). When the nonuniformity of external magnetic field is sufficiently strong, perturbations of a magnetic field caused by the variation of fluid temperature are assumed to be negligible. Therefore, thermomagnetic convection in nonuniform external magnetic field is qualitatively similar to gravitational convection.

When the external magnetic field is uniform (e.g. magnetic field inside a solenoid), the mechanism of thermomagnetic convection is more subtle. The rigorous mathematical formulations describing forces acting in this case will be given in Chapter 2, but qualitatively their origin can be seen from Figure 1.2(b). According to Maxwell's boundary conditions at the surface separating differently magnetised media, the normal component of the magnetic induction vector **B**, which is proportional to the sum of the normal components of the magnetisation vector **M** and the magnetic field vector **H** inside the fluid, remains constant. Therefore, the magnitude of internal magnetic field (H_1) must be smaller wherever fluid is stronger magnetised. Thus, given the dependence of fluid magnetisation on the temperature discussed above, we conclude that even if the externally applied magnetic field is uniform, a cooler and stronger magnetised fluid will have a stronger "field-blocking" effect. Therefore, the internal magnetic field H there will be weaker. Hence placing a non-isothermal ferrofluid in a magnetic field parallel to the applied temperature gradient creates an inherently unstable situation, which is a necessary condition for the occurrence of thermomagnetic convection. The two approaches used to model a motion of non-isothermal magnetic fluid in an external magnetic field in the limiting cases of $\nabla H^e \gg \nabla H$ and $\nabla H^e \ll \nabla H$ are termed as inductionless (zero order) and induction (first order) approximations, respectively [12, 13].

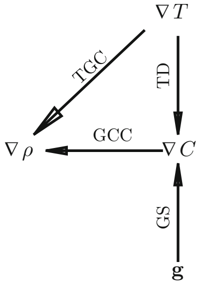

Fig. 1.3 Mechanisms of heat and mass transfer in ferrofluids in the absence of magnetic field. The acronyms denote TGC, thermogravitational convection driven by gravity **g**; GCC, gravi-concentrational convection; TD, thermodiffusion; and GS, gravitational sedimentation caused by gravity **g**. Various gradients existing in the fluid are of ∇T, temperature; $\nabla\rho$, density; and ∇C, concentration of solid phase.

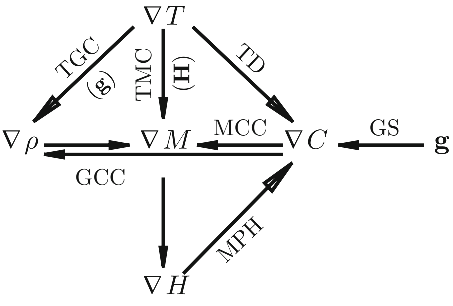

Fig. 1.4 Mechanisms of heat and mass transfer in ferrofluids in magnetic field. See caption of Figure 1.3 for the meaning of various acronyms. The additional acronyms appearing here denote TMC, thermomagnetic convection driven by the application of magnetic field **H**; MCC, magneto-concentrational convection; and MPH, magnetophoresis; ∇M and ∇H denote gradients of magnetisation and magnetic field, respectively.

It will be shown in Chapters 5 and 6 that thermoconvective instability arising in non-isothermal ferrofluids and caused by the combined action of gravitational buoyancy and ponderomotive forces is strongly influenced by the nonuniformity of solid-phase distribution in the bulk of fluid [23, 37, 38, 200]. In the absence of magnetic field, the gradient of the solid-phase concentration can be established as a result of thermodiffusion (TD) (or Soret effect) [23, 24, 75, 162, 230, 231] and gravitational sedimentation (GS) of particles and their aggregates [41, 84, 100, 141] as well as of insoluble residues [112, 161, 213] present in organic carrier fluids (see Figure 1.3). The size and weight variation of organic molecules contained in carrier fluids as well as the presence of unbound surfactant molecules can also lead to thermodiffusion [159, 190] resulting in the appearance of the concentration and thus fluid density gradient. Such a gradient can lead to gravi-concentrational convection (GCC) that may either enhance or suppress thermogravitational convection (TGC) arising due to nonuniform thermal expansion of a non-isothermal fluid.

In magnetic field, in addition to the above mechanisms, the concentration of solid phase can be affected by magnetophoresis (MPH) of magnetic particles and their aggregates [23, 197]. This may influence the local magnetisation of the fluid and lead to magneto-concentrational convection (MCC) (see Figure 1.4). These mechanisms act simultaneously with those causing thermomagnetic convection (TMC) schematically illustrated in Figure 1.2. As will be discussed in this book, the interaction of these processes leads to very complex macroscopic spatio-temporal dynamics including oscillatory convection, intermittency of convection patterns and suppression or promotion of convection onset and hysteresis.

A complex composition of ferrofluids and a wide range of microscopic processes taking place in them result in a large and frequently unmeasurable variation of fluid transport coefficients. Their values depend strongly on the conditions and history of storage and use of ferrofluids. This may lead to the unquantifiable uncertainty of the values of nondimensional control parameters that are used for identifying various flow regimes and thus to ambiguity of interpretation of experimental results. Several main reasons for that are discussed in Section 1.3.

Despite these difficulties the use of ferrofluids as magnetically driven heat carriers in conditions where gravitational convection is impossible due to the extreme congestion (microelectronics) or in the absence of gravity (spacecrafts) offers an efficient alternative to conventional heat management strategies. Laboratory experiments with ferrofluids placed in an external magnetic field are also used for physical modelling of processes taking place in the Earth's mantle, of oceanic currents and of flows arising in crystal growth applications.

1.3 Physical Properties of Ferrofluids

As noted in Section 1.1, in the absence of magnetic field, ferrofluids behave similarly to other non-magnetic nanofluids or colloids. Their effective thermal conductivity increases with concentration of nanoparticles because the nanoparticle material is a much better heat conductor than the base fluid. The thermal conductivity of a nanofluid is defined by the material and morphology of particles, the degree and type of their aggregation as well the type of the carrier liquid and the temperature. Its numerical value is well predicted by formulae first suggested by Maxwell [157] and Tareev [245] and later refined by various researchers by accounting for the shape of the particles and their aggregates, Brownian motion and thermodiffusion [103, 139, 246, 257]. The thermal conductivity of ferrofluids containing around to 10% by volume of nanoparticles increases up to 50% compared to that of a carrier liquid [26, 88, 134, 149, 188, 209]. However, in magnetic field, the value of the thermal conductivity of ferrofluid becomes a function of the field orientation relative to the direction of the temperature gradient. Depending on it the increase could vary between a few percent and several hundred percent [5, 94, 137, 149, 167, 186, 188].

Since the base liquids used for manufacturing ferrofluids are Newtonian, ferrofluids with concentration of nanoparticles up to 10% also remain Newtonian. However, as was shown by Einstein [83], the viscosity of fluids seeded with solid particles increases. Finite-size particles experience a rotating moment in a shear flow, which leads to the appearance of the so-called rotational viscosity. At present there exist around a dozen of rheological models of nanofluids that account for their viscosity dependence on nanoparticle concentration [11, 77, 122, 139, 204]. Experiments also show that the rotational viscosity depends on the size and shape of nanoparticles and their aggregates, fluid temperature and surfactants [11, 64, 77, 204, 257]. Moreover, the viscosity of ferrofluids depends on the magnitude and direction of the applied magnetic field [23, 158, 170]. The reason for that is that magnetic moments of particles with Brownian mechanism of relaxation [48] tend to align with the applied magnetic field by turning as a whole[2]. If the field is perpendicular to the local vorticity, it will hinder particle rotation, which is perceived as the increase of the fluid viscosity at a macroscopic level [108]—the so-called magnetoviscous effect. Similar to the rotational viscosity, magnetoviscosity is influenced by the local flow shear and the concentration and size of nanoparticles and their aggregates. In addition, it depends on the magnitude and direction of a magnetic field. Therefore, the value of magnetoviscosity measured in a given fluid using a viscosimeter can only give an approximation of the actual fluid viscosity in convection experiments because of the variation of flow velocity profiles from one experiment to another.

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[2] Particles with Neel [166] relaxation where magnetic moments align with the field within a particle not causing its overall rotation do not lead to magnetoviscous effect.

A species separation known as Soret effect or thermodiffusion can occur in non-isothermal mixtures. In colloids this effect is two orders of magnitude stronger than in binary liquid mixtures [23, 24, 75, 232, 252]. It is characterised by the Soret coefficient, which is the ratio of the thermal and molecular diffusion coefficients. The sign of the Soret coefficient in ferrofluids depends on the concentration of nanoparticles, properties of a carrier liquid, the magnitude of the applied magnetic field and its orientation with respect to the temperature gradient [25, 231, 232, 253], and its magnitude is hard to determine due to the difficulties with measuring the coefficient of molecular diffusion [232].

One of the most important characteristics of ferrofluids placed in a magnetic field is the initial magnetic susceptibility determining the magnetic response of the ferrofluid to an applied weak magnetic field. Its magnitude also depends strongly on the microstructure of the colloid: concentration, material, morphology and size distribution of nanoparticles, types and sizes of particle aggregates, composition of the carrier fluid and the temperature of ferrofluid [104, 142, 154, 195].

To conclude, ferrofluid studies have to consider a wide range of microscopic physical processes that can take place in the bulk of a fluid and lead to its de-homogenisation. Given that convection is one of the most sensitive natural phenomena, it can easily be affected by the so-created nonuniformities so that experimental results have to be interpreted very carefully. The presence of multiple interacting microscopic transport mechanisms also makes theoretical analysis of magnetoconvection a very challenging problem. At present it can only be solved approximately by employing a number of simplifying assumptions as will be detailed in Chapter 2. The obtained theoretical and computational results thus have to be viewed in the context of the validity of the adopted simplifications, which puts an even stronger emphasis on experimental observations and quantitative measurements.

Chapter 2
Governing Equations

Abstract In this chapter the equations describing flows of non-isothermal ferrofluids and the corresponding boundary conditions are summarised. The main physical assumptions under which these equations are valid are discussed, and references to further reading are given. The constitutive equations for ferrofluid magnetisation are also reviewed. It is emphasised that commonly used Langevin's magnetisation law may be inaccurate in the case of non-isothermal ferrofluids, and thus the second-order modified mean-field model is preferred. Subsequently, the nondimensional form of equations is introduced, and the major governing nondimensional parameters in terms of which the results are presented throughout the manuscript are defined, and their physical meaning is identified.

2.1 Simplifying Physical Assumptions and Basic Equations

Equations describing flows of a ferrofluid placed in an external magnetic field were first given in [89]. If the temperature variation in the flow domain is sufficiently small, the Boussinesq approximation of the continuity, Navier-Stokes and thermal energy equations that are complemented with Maxwell's equations for the magnetic field written in the magnetostatic form due to the negligible electrical conductivity of ferrofluids [13, 209] read:

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$$\nabla \cdot \mathbf{v} = 0\,, \tag{2.1}$$

$$\rho_* \frac{\partial \mathbf{v}}{\partial t} + \rho_* \mathbf{v} \cdot \nabla \mathbf{v} = -\nabla p + \eta_* \nabla^2 \mathbf{v} + \rho \mathbf{g} + \mu_0 M \nabla H\,, \tag{2.2}$$

$$\frac{\partial T}{\partial t} + \mathbf{v} \cdot \nabla T = \kappa_* \nabla^2 T\,, \tag{2.3}$$

$$\nabla \times \mathbf{H} = 0\,,\ \nabla \cdot \mathbf{B} = 0\,, \tag{2.4}$$

where

$$\mathbf{B} = \mu_0(\mathbf{M} + \mathbf{H})\,, \quad \mathbf{M} = \frac{M(H,T)}{H}\mathbf{H}\,. \tag{2.5}$$

In the above equations, $\mathbf{v}$ is the velocity vector with the respective components (u, v, w) in the x, y and z directions, t is time, T is the temperature, p is the pressure, $\mathbf{g}$ is the gravity acceleration, $\mathbf{B}$ is the magnetic flux density, ρ_* is the fluid density, η_* is the dynamic viscosity, κ_* is the thermal diffusivity of the fluid and $\mu_0 = 4\pi \times 10^{-7}$ H/m is the magnetic constant. The internal magnetic field inside the fluid domain is $\mathbf{H}$ such that $|\mathbf{H}| = H$. It induces fluid magnetisation $\mathbf{M}$ such that $|\mathbf{M}| = M$, which is assumed to be codirected with the magnetic field: $\mathbf{M} = \chi_* \mathbf{H}$, where χ_* is the integral magnetic susceptibility of the fluid. As discussed, for example, in [172] and references therein, this is true if the magnetic particle size does not exceed $d_p \sim 13$ nm. In this case the ratio of the Brownian particle magnetisation relaxation time $\tau_B = (4\pi d_p^3 \eta_*)/(k_B T)$, where $k_B = 1.38 \times 10^{-23}$ J/K is Boltzmann constant, to the viscous time $\tau_v = \rho_* d^2/\eta_*$ characterising the macroflow development is $\tau_B/\tau_v \sim 10^{-5}$. Thus it is safe to assume that the orientation of the magnetic moments of individual particles and thus of the fluid magnetisation follows the direction of a local magnetic field. However, the orientation of magnetic particle aggregates can in principle be affected by the mechanical torque due to the local shear of the flow so that they can misalign with the local magnetic field. Yet the experiments reported in [174] show that such a misalignment only becomes noticeable for shear rates exceeding 15 s^{-1}, while the shear rate for typical convection flows that are of interest here is of the order of 0.1 s^{-1} or smaller. Therefore the misalignment of the magnetisation and magnetic field vectors can be safely neglected. It is also common to assume that the fluid magnetisation depends only on field and temperature, which is the case when the concentration of magnetic particles remains uniform in experiments (this assumption is not always valid in reality due gravitational sedimentation, thermo- or magnetophoresis of solid particles, which we discuss in detail in Section 6.3.4).

The subscript $*$ in the governing equations denotes the values of the fluid properties evaluated at the reference temperature T_* and reference internal magnetic field H_*. In writing Equation (2.2), we assume that the fluid remains Newtonian. It has been found in experiments of [29] that this is a reasonable approximation for fluids with the concentration of solid phase not exceeding

$f = 0.1$. The more recent measurements reviewed in [171, Ch. 4] have indicated that ferrofluids placed in the magnetic field can also behave as Bingham fluids with a non-zero yield-stress that increases approximately quadratically with the applied magnetic field. However, the yield-stress magnitude remains very small for the field strength range relevant to thermomagnetic flows of interest here so that the Newtonian fluid approximation is well justified.

As evidenced by numerous studies [13, 86, 170–172, 174, 175, 193, e.g.], the viscosity of concentrated ferrofluids depends on the applied magnetic field, and the local flow shear that influences the concentration of aggregates formed as a result of a dipole interaction between magnetised particles. In general, both the average and local values of viscosity can vary. Even though multiple experiments aiming at quantifying such a dependence have been reported in literature [171, 174, 193, e.g.], the data collected in these measurements cannot be used directly to model flows in geometries and conditions that are significantly different from those of rheological experiments. However, in theoretical studies the reference average fluid viscosity only enters the nondimensional governing equations in combination with other fluid properties forming magnetic Rayleigh or Grashof numbers (e.g. see Section 2.2). In parametric studies their values are typically allowed to vary over a wide range, which effectively includes all experimental conditions even though the exact value of magnetoviscosity remains unknown. The unknown variation of the local viscosity and other fluid properties subject to the action of the locally varying magnetic field and shear presents a more daunting problem. It is well known [239, 240, e.g.] that if sufficiently large such a variation can strongly influence the structure of the flow and its stability. Yet to make analytical progress in absence of a quantitative rheological model, one is forced to neglect these spatial variations of fluid properties in Equation (2.2). This is consistent with a widely used Boussinesq thermal approximation adapted for magnetic fluids [13] and is reasonable if the temperature and magnetic field variation across the domain occupied by fluid remain small. The qualitative agreement between the computational results and the experimental observations reported in [238, e.g.] indicates that indeed such a simplification preserves sufficient accuracy of the model and makes it tractable. Further discussion and a quantitative justification of this simplification will be provided for the specific case of a vertical fluid layer in Sections 2.2 and 3.2.

The last term in Equation (2.2) represents a ponderomotive (Kelvin) force that acts on a magnetised fluid in a nonuniform magnetic field and drives it towards regions with a stronger magnetic field as discussed in [13, 144]. To close the problem, thermal and magnetic equations of state are required, which are assumed to be in the simplest linear form that is valid for small temperature and field variations within the layer,

$$\rho = \rho_* - \beta_*(T - T_*)\,, \tag{2.6}$$

$$M = M_* + \chi\Delta H - K\Delta T\,, \quad \Delta H \equiv H - H_*\,, \quad \Delta T \equiv T - T_*\,. \tag{2.7}$$

Here $\beta_* = -\left.\frac{\partial\rho}{\partial T}\right|_P$ is the coefficient of thermal expansion of the fluid at $T = T_*$, H_* and $M_* = \chi_* H_*$ are the magnitudes of the magnetic field, and the magnetisation at the location with temperature T_*, $\chi = \partial M/\partial H|_{(H_*,T_*)}$ is the differential magnetic susceptibility and $K = -\partial M/\partial T|_{(H_*,T_*)}$ is the pyromagnetic coefficient. Using Equation (2.7) it is possible to rewrite (2.5) as

$$\mathbf{M} = \frac{\chi H + (\chi_* - \chi)H_* - K\Delta T}{H}\mathbf{H}\,. \tag{2.8}$$

Subsequently, eliminating the magnetisation in favour of the magnetic field, one obtains from the second of Equation (2.4)

$$(1+\chi)\nabla\cdot\mathbf{H} + \frac{(\chi_* - \chi)H_* - K\Delta T}{H}(\nabla\cdot\mathbf{H} - \nabla H\cdot\mathbf{e}) - K\nabla T\cdot\mathbf{e} = 0\,, \tag{2.9}$$

where $\mathbf{e} = (e_1, e_2, e_3) \equiv \mathbf{H}/H$ is the unit vector in the direction of the magnetic field. This equation shows that thermomagnetic coupling occurs mostly when the magnetic field and the temperature gradient have components in the same direction.

It is convenient to redefine pressure p entering the momentum equation (2.2) so that it includes both a hydrostatic component and a Kelvin force potential (see also [170, pp. 86, 87]). Upon using Equation (2.7), one writes

$$\begin{aligned}\mu_0 M\nabla H &= \mu_0[M_* + \chi\Delta H - K\Delta T]\nabla H \\ &= \mu_0\nabla[M_* H + \frac{1}{2}\chi\Delta H^2] - \mu_0 K\Delta T\nabla H\,.\end{aligned}$$

Upon introducing the modified pressure

$$P = p - \rho_*(\mathbf{r}\cdot\mathbf{g}) - \mu_0\left[M_* H + \frac{1}{2}\chi\Delta H^2\right]\,, \tag{2.10}$$

where $\mathbf{r} = (x, y, z)$ is the position vector equation (2.2) is written as

$$\rho_*\frac{\partial\mathbf{v}}{\partial t} + \rho_*\mathbf{v}\cdot\nabla\mathbf{v} = -\nabla P + \eta_*\nabla^2\mathbf{v} - \rho_*\beta_*(T - T_*)\mathbf{g} - \mu_0 K\Delta T\nabla H\,. \tag{2.11}$$

The governing Equations (2.1)–(2.4) require a set of appropriate boundary conditions. At the solid boundaries limiting the domain occupied by fluid, its velocities vanish and the temperature is assumed to be known:

$$\mathbf{v} = \mathbf{0}\,, \quad T = T_b\,. \tag{2.12}$$

The applied magnetic field must satisfy magnetic boundary conditions:

$$(\mathbf{H}^e - \mathbf{H})\times\mathbf{n} = \mathbf{0}\,,\ (\mathbf{B}^e - \mathbf{B})\cdot\mathbf{n} = 0\,, \tag{2.13}$$

where superscript e denotes external fields and $\mathbf{n}$ is the normal vector to the boundaries. Using Equation (2.9), the second of the conditions in Equation (2.13) can be rewritten as

$$[((1+\chi)H + (\chi_* - \chi)H_* \pm K\Theta)\mathbf{e} - \mathbf{H}^e] \cdot \mathbf{n} = 0 \tag{2.14}$$

where it is assumed for definiteness that the temperatures at the opposite surfaces bounding a ferrofluid layer deviate from the average value T_{av} and are $T_{av} \pm \Theta$. This completes the formulation of a main set of governing equations.

2.2 Nondimensionalisation and Governing Parameters

The governing equations and boundary conditions are nondimensionalised using

$$(x,y,z) = d(x',y',z')\,,\ \mathbf{v} = \frac{\kappa_*}{d}\mathbf{v}'\,,\ t = \frac{d^2}{\kappa_*}t'\,,\ P = \frac{\rho_*\kappa_*^2}{d^2}P'\,, \tag{2.15}$$

$$T - T_* = \Theta\theta'\,,\ \mathbf{H} = \frac{K\Theta}{1+\chi}\mathbf{H}'\,,\ H = \frac{K\Theta}{1+\chi}H'\,, \tag{2.16}$$

$$\mathbf{M} = \frac{K\Theta}{1+\chi}\mathbf{M}'\,,\ M = \frac{K\Theta}{1+\chi}M'\,, \tag{2.17}$$

where d is the characteristic size of the domain occupied by fluid and Θ is the characteristic temperature difference across the domain. Then omitting primes for simplicity of notation, one obtains nondimensional governing equations

$$\nabla \cdot \mathbf{v} = 0\,, \tag{2.18}$$

$$\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} = -\nabla P + \mathrm{Pr}\,\nabla^2 \mathbf{v} - \mathrm{Ra}\,\mathrm{Pr}\,\theta \mathbf{e}_g - \mathrm{Ra}_m\,\mathrm{Pr}\,\theta \nabla H\,, \tag{2.19}$$

$$\frac{\partial \theta}{\partial t} + \mathbf{v} \cdot \nabla \theta = \nabla^2 \theta\,, \tag{2.20}$$

$$\nabla \times \mathbf{H} = \mathbf{0}\,, \tag{2.21}$$

$$\begin{aligned}(1+\chi)(\nabla \cdot \mathbf{H} - \nabla\theta \cdot \mathbf{e})& \\ + \frac{(\chi_* - \chi)\mathrm{N} - (1+\chi)\theta}{H}(\nabla \cdot \mathbf{H} - \nabla H \cdot \mathbf{e}) &= 0\,,\end{aligned} \tag{2.22}$$

$$\mathbf{M} = [\chi H + (\chi_* - \chi)\mathrm{N} - (1+\chi)\theta]\mathbf{e} \tag{2.23}$$

with the boundary conditions

$$[((1+\chi)(H \pm 1) + (\chi_* - \chi)\mathrm{N})\mathbf{e} - \mathbf{H}^e] \cdot \mathbf{n} = 0\,, \tag{2.24}$$

$$\mathbf{v} = \mathbf{0}\,,\ \theta = \theta_b \tag{2.25}$$

along the solid boundary. The dimensionless parameters appearing in equations are

$$\mathrm{Ra} = \frac{g\beta\Theta d^3}{\eta_*\kappa_*}\,,\ \mathrm{Ra}_m = \frac{\mu_0 K^2 \Theta^2 d^2}{\eta_*\kappa_*(1+\chi)}\,,\ \mathrm{Pr} = \frac{\eta_*}{\rho_*\kappa_*}\,,\ \mathrm{N} = \frac{H_*(1+\chi)}{K\Theta}\,. \tag{2.26}$$

The gravitational and magnetic Rayleigh numbers Ra and Ra_m characterise the importance of buoyancy and magnetic forces, respectively, Prandtl number Pr characterises the ratio of viscous and thermal diffusion transport and parameter N represents the nondimensional magnetic field at the reference location. Note that while magnetic Rayleigh number is the main nondimensional parameter characterising pure magnetoconvection, in laboratory experiments, the influence of gravitational convection usually cannot be neglected, and in studies of mixed gravitational and magnetic convection flows, it is traditional to use a nondimensionalisation based on "viscous speed" $\frac{\eta_*}{\rho_* d}$ rather than on "thermal speed" $\frac{\kappa_*}{d}$ used in (2.15). In this case the nondimensional momentum and thermal energy equations (2.19) and (2.20) take a slightly different form and read

$$\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v}\cdot\nabla\mathbf{v} = -\nabla P + \nabla^2\mathbf{v} - \mathrm{Gr}\theta\mathbf{e}_g - \mathrm{Gr}_m\theta\nabla H\,, \tag{2.27}$$

$$\frac{\partial \theta}{\partial t} + \mathbf{v}\cdot\nabla\theta = \frac{1}{\mathrm{Pr}}\nabla^2\theta\,, \tag{2.28}$$

where gravitational and magnetic Grashof numbers are related to gravitational and magnetic Rayleigh numbers as

$$\mathrm{Gr} = \frac{\mathrm{Ra}}{\mathrm{Pr}} \text{ and } \mathrm{Gr}_m = \frac{\mathrm{Ra}_m}{\mathrm{Pr}}\,, \tag{2.29}$$

respectively.

Among other important physical quantities characterising the magnetic properties of the fluid are differential and integral magnetic susceptibilities χ and χ_* and pyromagnetic coefficient K that depend on the applied magnetic field and the temperature. The pyromagnetic coefficient K only enters the governing equations as an element of the nondimensional groups (2.26) so that its exact value is not required for the analysis. However, the magnitude of K (and thus of parameter N) can be conveniently used to distinguish between paramagnetic and ferromagnetic fluids. It is small in the former case and typically is of the order of 10^2 in the latter. At the same time, the values of magnetic susceptibilities χ and χ_* are important parameters entering the governing equations directly. It is a common practice to estimate them from Langevin magnetisation law that reads [144, 209]