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Stefan Odenbach

Magnetoviscous Effects in Ferrofluids

Springer

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Preface

Within the scope of this work we've investigated the magnetoviscous effects – i.e. the changes of viscous properties due to the action of magnetic fields – in so-called ferrofluids. These fluids, suspensions of magnetic nanoparticles in appropriate carrier liquids, show a pronounced increase of viscosity in the presence of moderate magnetic fields with strengths of the order of several tens of mT. Classically this effect is explained by the hindrance of the free rotation of magnetic particles – with a magnetic moment spatially fixed in the particle – in a shear flow due to magnetic torques trying to align the particles' magnetic moments with the magnetic field direction.

Starting from the classical theory by Mark Shliomis (Shliomis, 1972) we've performed a couple of experiments to validate the predictions of the theory. The use of relatively concentrated commercial magnetic fluids lead to the conclusion that the mentioned theory – developed for highly diluted fluids – is not able to give a quantitative description of the behavior of commercial fluids. The discrepancies have been attributed to the appearance of interparticle interactions between the magnetic particles.

Since the microscopic make-up of commercial ferrofluids is relatively complicated, and in particular parameters like the size distribution of the magnetic particles are not known precisely, a theoretical description of the microscopic reasons for the fluids' macroscopic behavior is impossible without further information. Therefore we've started a series of investigations shedding light on the viscous behavior of magnetic fluids in the presence of magnetic fields, stepwise reducing the number of relevant microscopic parameters to prepare a basis for sufficient modeling of concentrated ferrofluids.

As a first step in this development a specialized rheometer for the investigation of magnetic fluids has been designed. With this rheometer, allowing well-defined application of a magnetic field to a rheometric flow of ferrofluids, we've investigated the shear dependence of the magnetoviscous effect in commercial ferrofluids. These investigations showed that the field-dependent increase of viscosity reduces with increasing shear rate. On the basis of this result we developed a model, assuming that the formation of chains of magnetic particles dominates the magnetoviscous properties of magnetic fluids. The chains themselves represent large magnetic structures which lead to pronounced changes of viscosity if a field is applied. Furthermore, the rupture of the chains in a shear flow and the resulting reduction of the size of the magnetic structures is a starting point for the explanation of the observed shear thinning.

Since chains of magnetic particles can only be formed by particles exhibiting a sufficient interparticle interaction, and since this interaction depends furthermore on the size of the particles, the next step had to be a clarification, whether the relatively small fraction of large particles in the suspension used is of major importance for magnetoviscosity in ferrofluids. These large particles exhibit – in contrast to the majority of particles with diameters of about 10 nm – sufficiently strong interaction to explain at least the appearance of chain formation.

To get an insight into these questions, we've performed experiments using ferrofluids with variable contents of large particles. In these experiments it was clearly shown that the magnetoviscous effect rises with an increasing amount of large particles. This leads to further input for the theoretical modeling. In an extended approach the ferrofluid is assumed to be a bidisperse system containing a large fraction of small particles, which do not directly contribute to magnetoviscosity, and a small fraction of large particles which form chains determining the field-dependent changes of viscosity. On the basis of these assumptions the magnetoviscous properties could be fitted quantitatively to the experimental data using methods of statistical physics. Thus, a first quantitative description of the microscopic reasons for the rheological behavior of ferrofluids was found, taking into account the effects to the formation of magnetic particle chains. The conclusion that chains exist in the fluids gives rise to the assumption that these fluids should exhibit viscoelastic effects too. To prove this, we finally carried out experiments on the Weissenberg effect, i.e. the climb of a free surface of magnetic fluids at a rotating axis, showing the field-dependent existence of normal stress differences in ferrofluids. Again, the experimentally found behavior could be explained by the formation and rupture of chains of magnetic particles in the fluid.

Thus – within the scope of this work – we've been able to develop a microscopic model of ferrofluids allowing a quantitative description of their rheological behavior, and to prove this model with numerous experimental results on field-dependent effects in ferrofluids rheology. On the basis of these results, information for the optimization of ferrofluids with respect to their magnetoviscous behavior can be obtained, leading to the synthesis of new ferrofluids. Such fluids with enhanced magnetoviscous properties may be used in the future development of devices using the magnetically induced control of viscous properties as an active part in technical applications like dampers or clutches.

Investigations like those described in this work require not only a certain time span to be performed but also the help and cooperation of numerous colleagues and the financial support enabling the research activities.

Thus I'd like to take the opportunity to express my gratitude to those helping me to do this research during recent years.

First of all I've to thank Prof. Dr.-Ing. H. J. Rath and Prof. Dr. K. Stierstadt for providing me with a working environment in Bremen as well as in former times in Munich that gave me the possibility of developing ideas and building up a research team able to explore this new and interesting field. Without these boundary conditions this wouldn't have been possible.

Furthermore my gratitude goes to my co-workers who were prepared to work even in difficult ways towards new scientific and technical goals: Dipl. Phys. H. Gilly for lively discussions during the time in Munich, Dipl. Phys. H. Störk who

built the first version of the ferrofluid rheometer in Wuppertal, and last but not least the members of the ZARM-ferrofluid team who participated in various experiments which led to the results presented, Dipl.-Ing. J. Fleischer, Dipl.-Ing. M. Heyen, Dipl.-Ing. K. Melzner, Dipl.-Ing. T. Rylewicz, Dipl.-Ing. S. Thurm and Dipl.-Ing. T. Völker.

Besides this I'm grateful to numerous colleagues and friends for fruitful and enlightening discussions. In this case it's nearly impossible to name all those who have been with me during the years, but I'd like to mention particularly: Prof. E. Blums, Prof. A. Zubarev and Prof. L. Vekas who were our guests in Bremen numerous times in the course of fruitful cooperations; Dr. K. Raj who provided us with the fluid series for the experiments concerning the influence of large particles; Prof. K. Stierstadt, Dr. H. W. Müller and Dipl.-Ing. Ch. Eigenbrod who helped me with deep and inspiring discussions; and numerous members of the German ferrofluid community who are helping to form a powerful research community on magnetic fluids.

As mentioned, financial support is also essential for the performance of research in general. In this respect I'd like to mention particularly the Deutsche Forschungsgemeinschaft (DFG) for granting most of the experimental work performed during the years in Bremen. In this context I'd like to express my gratitude to Dr. W. Lachenmeier from DFG for the excellent cooperation during the establishment of the DFG priority program on magnetic fluids focusing partly on the topics discussed here. Furthermore I've to thank the Deutsches Zentrum für Luft- und Raumfahrt (DLR), in particular Dr. H. Binnenbruck, for financial support over many years. In addition, the flight opportunities provided by DLR and ESA were of essential importance for the Weissenberg-effect experiments.

Since most of the work presented has an experimental character, the technical support provided by the workshop at ZARM and the Fallturm Betriebsgesellschaft was often of great importance to the success of our research. I'm especially grateful for this, since we often had to set extremely tight deadlines which were always observed.

Besides all the research work, these pages had finally to be written, and in this context I'd like to express my thanks to E. Renschen and C. Wieske for a lot of typing.

In general, the development of scientific activities is a part of life that can not be successful if it is not supported by an appropriate private environment. Many of the colleagues mentioned above have become real friends during the years, supporting me even in difficult times.

But particular gratitude in this respect goes to my parents and my wife Marlene, supporting me over all the years and understanding the difficulties and setbacks of this kind of life.

Bremen, 2001 *Stefan Odenbach*

Contents

1. Introduction

1.1 Magnetic fluids

Fluids which can be effectively controlled by magnetic fields of moderate strength are a challenging subject for scientists interested in the basics of fluid mechanics as well as for application engineers. For the basic research the introduction of a controllable force into the fundamental hydrodynamic equations opens a fascinating field of new phenomena.

Forces which can be varied over wide ranges in strength and direction relative to a flow are usually only applicable in theoretical treatments. For forces exhibited by magnetic field gradients the situation changes since magnetic fields can be varied quite well in strength and direction using different types of coils, pole shoes and permanent magnets. If the magnetic influence exerted by a magnetic field becomes strong enough to compete with gravitational forces, a new class of hydrodynamic phenomena becomes experimentally accessible.

Also the design of applications using fluids as relevant active or passive components gains new possibilities if the fluids can be positioned or moved by a force which can be produced by an electric current through a coil being controlled and switched electronically. Again – if the necessary forces can be produced by moderate fields which are generated with a relatively small technical effort – new design ideas using an additional control parameter can be realized.

Due to the fact that no natural liquids offer these features, the starting point of the field of magnetic fluid research can be found in theoretical treatments of magnetically controlled heat transfer machines (Resler and Rosensweig, 1964). Since these early ideas already showed that a liquid material with controllable magnetic properties would provide numerous development possibilities, strong efforts have been undertaken to synthesize a system enabling the mentioned magnetic control. As will be shown later on, suspensions of magnetic nanoparticles in appropriate carrier liquids are a sufficient realization of such a new class of smart materials. After their first stable synthesis in the early 1960s the development of these suspensions – called ferrofluids – proved the high potential of the new research field. Several hundred scientific publications per year and thousands of approved patents document the vitality of ferrofluid research as well as the close connection to applied engineering.

But not only engineers, experimental and theoretical physicists contribute to the development of the field called ferrohydrodynamics (Neuringer and Rosen-

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sweig, 1964). The complexity of the system and its difficult chemical make-up require distinct knowledge in chemistry and colloidal physics to synthesize new and improved liquids and to modify the basic properties of the suspensions. Moreover the utilization of the system is not only restricted to technical applications – a use is also possible for various medical treatment purposes. Thus, the overall field of ferrofluid research has a highly interdisciplinary character, bringing chemists, experimental physicists, engineers, theoretical physicists, applied mathematicians and physicians together.

The interdisciplinarity of the field leads to the necessity for strong cooperation between scientists from different research directions. In principle, basic research has to provide information about the relation between the microstructural make-up and the macroscopic field-dependent properties of the liquids. This knowledge has to be used to tailor special suspensions for new application ideas defining certain requests concerning the fluids behavior in the presence of magnetic fields. Obviously such an interconnected research forces a mutual fertilization of the involved research areas, making the whole field highly challenging from a scientific point of view.

The future development perspective and this interdisciplinary aspect has been the driving force in the establishment of various national research programs, e.g. in Japan and France. The most recent of these programs, a DFG priority program started in Germany in 2000, accounts especially for the interdisciplinarity of the field by combining the efforts of chemists and basic researchers with application engineers and scientists from medical research fields.

These programs are actually leading to a new concentration of efforts in the field, where the investigation of magnetoviscous effects is one of the core points of interest.

1.2 Magnetoviscous effects

Shortly after the publication of the first patent on the synthesis of stable suspensions of nanosized magnetic particles intense research efforts were started in the field, leading to the development of a theoretical background – the theory of ferrohydrodynamics based on early papers by M. Shliomis (Zaitsev and Shliomis, 1969; Shliomis, 1972) – as well as to patents for numerous applications which partly gained commercial importance forcing further development of the whole research area. While basic research covered nearly all areas of flow control and property changes in the fluids induced by the action of magnetic fields, commercially successful applications just used the possibility of the magnetic positioning of the liquids.

The principally predicted employment of the magnetic control of flow in the fluid, or the change of its properties under the influence of a field did not reach the stage of experimental realization since they require relatively high concentration of the suspended magnetic material to achieve a reasonable strength of the effects. The high concentration leads to an interaction of particles, which can not be neglected. The need to account for the interparticle interaction increases the complexity of the system essentially. Thus a well-founded understanding of phenomena observed in such suspensions is relatively hard to obtain. Nonetheless the knowledge about the microstructural properties and their importance for the fluids' macroscopic behavior is the background needed to synthesize application tailored suspensions and to design new devices based on magnetic liquids. Furthermore the influence of magnetic fields on changes in the microstructure of fluids of different make-up has to be taken into account in the prediction of their macroscopic properties.

These problems are of principal importance for the magnetically induced changes in the viscosity of magnetic fluids. The basic theories – formulated nearly three decades ago – model the microstructural make-up of the suspensions in an idealized way, neglecting any kind of interparticle interaction. Therefore these theories can only be used for quantitative predictions of the behavior of highly diluted fluids. In contrast "the promise of controllable fluids", as J.D. Carlson (Carlson, 1994) named the development of new applications of magnetorheological fluids, always requires highly interacting systems to obtain an order of magnitude of the relevant effects – e.g. the magnetoviscous effect – required for commercial needs.

Experimentally it has been found that relatively strong field influence on viscosity can be induced not only in magnetorheological fluids, but also in ferrofluids with sufficient particle-particle interaction. But only recently a deeper understanding of these interactions led to microscopic models quantitatively explaining the experimentally found phenomena. This knowledge is actually used to find ways to optimize the magnetorheological effects in long-term sedimentation stable ferrofluids.

In this context new research concepts have been set up to accelerate the development process. Synthesis of the fluids, basic understanding of their properties, and the development of applications using magnetoviscous properties of the fluids are no longer addressed as isolated research fields. Moreover, programs have been established combining the expertise of the different fields of knowledge in ferrofluid research. The mentioned priority program of DFG is an example of such an integrated research activity. Fluids produced by several synthesizing groups are characterized and rheologically tested and from the understanding of the fluids' behavior steps towards optimization are undertaken. Parallel to this development new applications are designed, using in the beginning existing magnetorheological fluids to define the necessary properties of the fluids to be developed, and thus provide a guideline for the further synthesis steps.

1.3 Publications on ferrofluids

As already mentioned, the field of ferrofluid research is actually more than 30 years old. Thus it is clear that not only original publications in journals or conferences have been released, but also textbooks have been published giving overviews on certain areas of the investigation of fluids containing magnetic nanoparticles. In 1985 the famous book "Ferrohydrodynamics" by Ronald Rosensweig

(Rosensweig, 1985) was issued, and it is still the standard textbook for people entering the field of magnetic fluid research. Rosensweig's book leads the reader through all areas of the research field – from the synthesis and properties of magnetic fluids and the foundation of the theory of ferrohydrodynamics towards problems of experimental hydrodynamics in ferrofluids as well as the description of various applications. It features examples for flow control and magnetically driven surface and transport instabilities as well as some remarks concerning field-induced changes of the properties of the fluids.

Looking to magnetoviscous effects only the first results of McTague (McTague, 1969) and Rosensweig (Rosensweig et al., 1969) are briefly mentioned, and a glance at the related theory by Shliomis (Shliomis, 1972) is given.

A slightly more detailed treatment of the rheology of ferrofluids in a magnetic field was given in the second general textbook on "Magnetic Fluids" by Blums, Cebers and Maiorov (Blums et al., 1997). They include an extended theoretical discussion of rotational viscosity and deal also with questions like the dependence of the magnetoviscous effects on particle shape and the effect of variation of shear rate for weak shear. In addition this book also gives a good overview on ferrofluid research enlightening the related question from a more theoretical point of view.

Besides these two books no general treatment of the whole area of ferrofluid research is currently available. All other books have been published with a focus on certain sub-areas and refer to Rosensweig and Blums for the general questions. The field of heat and mass transfer was well treated by Blums, Mikhailov and Ozols in "Heat and Mass Transfer in MHD Flows" (Blums et al., 1986) which contains a special section on heat and mass transfer effects in ferrofluids – while the main part of the book is devoted to conducting fluids and thus to the action of Lorentz forces rather than of magnetic body forces.

Furthermore two books on applications of magnetic fluids are available. "Magnetic Fluids and Applications Handbook" by Berkovsky and Bashtovoy (Berkovsky and Bashtovoy, 1996) and "Engineering Applications of Magnetic Fluids" (Berkovsky et al., 1993) give an overview on numerous kinds of usage of ferrofluids in different fields, for example mechanical positioning, separation or even medicine. Besides the mentioned books, further monographs are available in Russian, Berkovsky and Polevikov's work on "Numerical Experiments in Ferrofluids" (Berkovsky and Polevikov, 1988). But since these have not been translated into English, the availability of the information contained is unavailable for an English-speaking reader, reducing their importance and rating.

1.4 The scope of this work

With the present work the field of magnetoviscous properties of ferrofluids will be addressed. As mentioned above, the standard textbooks give only a short treatment of the early findings concerning field effects on the rheological behavior of ferrofluids. Moreover no special treatment of this subject has existed till now. On the other hand the investigation of field-induced changes of the viscosity of suspensions of magnetic nanoparticles is one of the most vital areas in magnetic fluid research nowadays. The current research questions, focusing on the tailored design of fluids for new applications using the magnetoviscous effects, require a detailed understanding of the effect itself as well as of the influence of the microscopic make-up of the fluid on its macroscopic behavior. Since especially the latter mentioned question of the dependence of macroscopic effects on microscopic properties is based on experimental and theoretical results we obtained recently, no comprehensive description of the field exists yet.

So the idea of this work is to combine a description of the basics of magnetoviscous effects with a compilation of the most recent findings on the influence of structure formation on the viscosity of ferrofluids. To achieve this goal, the present work is organized in the following way.

Chapter 2 will introduce the material which is the focus of the discussion. Ferrofluids and their basic properties will be discussed to an extent that allows us to read the upcoming treatment of magnetoviscosity without further basic knowledge on suspensions of magnetic nanoparticles. Besides the discussion of basic properties, Chap. 2 will also contain a short glance on applications of ferrofluids. This part is thought to motivate the engineering aspect of the whole research field in general as well as to highlight the investigation of magnetoviscous effects for applications. This section does not claim to replace the standard textbooks mentioned in Sect. 1.3. Its scope is only to introduce those topics needed for the discussion of the main focus of this work. Thus a couple of references to the standard books are given to enable the reader to find more detailed information on topics from the field of ferrofluid research outside the focus of this work.

In Chap. 3 the basic phenomenon of rotational viscosity, i.e. the influence of a magnetic field on the viscosity of a suspension of noninteracting nanoparticles is discussed. Starting from an explanation of the basic physical background of the phenomena of field-induced viscosity changes in ferrofluids, the theoretical approach of Shliomis is reviewed. Particular interest is paid here to all aspects related to experimental proofs of the theory rather than to a deep theoretical discussion of the approach itself. Nonetheless, the derivation of the basic equation for rotational viscosity is briefly compiled to give the reader a general glance at one of the most fundamental theoretical developments of ferrohydrodynamics. Starting from the various theoretical predictions, experimental proofs of the theory are presented, leading to a discussion of the range of validity of the theory and in particular of the problems that appear if concentrated fluids are considered. Finally, for reasons of completeness, the phenomenon of viscosity reduction in alternating magnetic fields is briefly discussed to illustrate the wide range of phenomena based on the interaction of the magnetic field with the magnetic moment of the particles.

The magnetoviscous effects in concentrated suspensions, and thus in systems of interacting particles, are then discussed in Chap. 4. The starting points for this discussion are the discrepancies found in Chap. 3 in the comparison of Shliomis' theory with the experimental results for concentrated suspensions. Again the experimental investigation of magnetoviscous effects is the center of the discussion. The necessary experimental techniques, and the connected experimental problems are described in detail to form the basis for the discussion of the measured phenomena. The major part of this section is occupied with rheological investigations showing field and shear dependence of the magnetoviscous effect and providing the information necessary to construct a microscopic model explaining the phenomena observed. The related model is then briefly introduced and its results are compared with the experimental findings. Finally – as one of the consequences of the model – the question on magnetically induced viscoelasticity is discussed on the basis of a series of experiments on the Weissenberg-effect in a ferrofluid under the influence of a magnetic field. Again various results leading to a microscopic understanding of the appearance of the phenomena are presented, and the existence of normal stress differences and their dependence on magnetic field strength is experimentally verified. As in Chap. 2, this section is not thought to replace or rewrite the content of the standard textbooks, this time those dealing with rheology. The general rheological background is only mentioned to an extent that makes the description of the effects in ferrofluids understandable. For a deeper insight into rheology the interested reader will be referred to rheology and rheometry textbooks.

Finally Chap. 5 focuses on magnetorheological fluids, i.e. on suspensions of micron-sized magnetic particles. The scope of this section is mainly those effects which can in principle be achieved in a magnetoviscous system. Thus it is a glance at the future of the research on magnetic fluids and it shows how strong magnetoviscous effects can become if interparticle interaction becomes dominant in the behavior of a magnetic suspension. The principal differences between ferrofluids and magnetorheological fluids are highlighted, to motivate again the need for an improvement of the magnetoviscous effects in stable suspensions of nanosized magnetic particles.

2. Magnetic fluids

The material in the focus of this work are liquids which can be controlled by moderate magnetic fields. Presently no molecular liquids exhibit this property in a way that it has importance for technical applications in everyday life. Looking to ferromagnetic materials, it is well known, that their Curie temperature is always well below the melting point, and thus the materials loose their spontaneous magnetic ordering before they become liquid (Kittel, 1996).

Focusing on liquid metals, a well-established technique of magnetic control is given in the field of magnetohydrodynamics. An electric current is applied to the liquid metal and the Lorentz forces in strong magnetic fields can be used to control the flow of such systems (Davidson and Thess, 2000). Nevertheless, reasonable forces, providing significant changes of the liquid metal flow, require extremely high magnetic field strength in the order of several Tesla. In addition a liquid metal requires usually a high temperature environment and thus does not fit the requirements for a broad technical application. Even stronger problems for technical applications appear if undercooled metallic melts are considered. Such melts of certain Co-Pd alloys, undercooled to a high degree, have been found to show magnetic ordering even in the liquid state (Wilde et al., 1996a, Wilde et al., 1996b). But obviously these magnetic properties can not be used in the design of a technical device, since the undercooled state does not allow any handling of the liquid. Comparably a technical use of liquid ^{3}He showing magnetic ordering at temperatures below 3 mK (Mermin and Lee, 1976) seems to be not realistic at all.

As already mentioned, flow control using the Lorentz force requires extremely high magnetic field strength. Thus the only hope to obtain a real magnetic control of a liquid must concentrate on the question of magnetic body forces, commonly written as the Kelvin force (Landau and Lifschitz, 1985)

$$F_K = \mu_0 \int M \nabla H dV \tag{2.1}$$

for a magnetizable material with susceptibility proportional to density. Here M denotes the magnetization of the fluid, ∇H the magnetic field gradient, μ_o the vacuum permeability (μ_o=1.2566·10^{-6} Vs/Am) and the integration is carried out over the volume of the sample V. In paramagnetic salt solutions this force is negligible compared e.g. with the gravitational force even for strong magnetic fields, since their magnetization is too small. To make real use of the magnetic body force, a liquid material is required, having high magnetization even for small magnetic field strength. The way out of this situation was shown in 1964 by S. Papell (Papell, 1964) by producing stable suspensions of magnetic nanoparticles in appropriate carrier liquids. As will be discussed later on, these suspensions,

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commonly called ferrofluids, exhibit an extraordinary high initial susceptibility and thus show high magnetization for magnetic field strength in the order of about 50 mT. Thus their flow and properties can be controlled by such moderate magnetic fields. Before we will discuss the magnetic properties of these liquids and the resulting magnetic forces applicable to them, we will first have a glance on their basic make-up and the stability requirements they have to fulfill.

2.1 Basic structure and stability

As mentioned, the fluids we will have in focus from now on, are suspensions of magnetic particles in a liquid carrier medium. Obviously, the requirement of stability – being of outstanding importance if a technical use of the suspensions is considered – includes first of all the stability against sedimentation of the particles. Such sedimentation, and the connected demixing of the suspensions, can be driven by gravitational or magnetic forces. To ensure, that the particles do not sediment and that the suspension thus remains well dispersed, one has to observe, that the thermal energy of the particles $E_T=k_BT$ (k_B: Boltzmann's constant, T: absolute temperature) is high enough to provide sufficient mixing of the suspensions. Therefore it needs to be higher than the energy of the particles in the gravitational field or in a magnetic field respectively. As an example we will here shortly discuss this stability argument for the sedimentation in a magnetic field gradient. This sedimentation can be avoided, as long as the thermal energy is strong enough to enable the particles to move freely between a region with strong magnetic field and a field free region. Such a step in magnetic field strength can stand for an idealized magnetic field gradient. The energy of the particles in the field is given by (Landau and Lifschitz, 1985)

$$|E_H| = \mu_0 \, m \, H \quad , \tag{2.2}$$

where m is the magnetic moment of the particle. Using the spontaneous magnetization of the magnetic material of the particles M_0 one can rewrite their magnetic moment by

$$m = M_0 \frac{\pi}{6} d^3 \tag{2.3}$$

with d being the particles' diameter.

Therefore the energy argument for sedimentation stability in a magnetic field gradient $E_H<E_T$ becomes

$$k_B T > \mu_0 M_0 \frac{\pi}{6} d^3 H \quad , \tag{2.4}$$