

# Euromech Colloquium 526



# Patterns in Soft Magnetic Matter

Book of abstracts



# **Euromech Colloquium 526**

# "Patterns in Soft Magnetic Matter"

held in Dresden from the 21st to the 23rd of March 2011

is organized with the kind support of the



# Sunday, 20th of March 2011

# 18:00–21:00 registration, get together

# Monday, 21st of March 2011

09:00–09:15	opening		
<b>plenary talk</b> 09:15–09:50	JF. Berret	Controlling the shape of supracolloidal assemblies using magnetic fields: towards the generation of highly persistent paporods	page 3
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10:40–11:30	coffee break		
11:30–11:55	<i>A. Lange</i> , S. Odenbach	Patterns of thermomagnetic convection in magnetic fluids subjected to spatially modulated magnetic fields	26
11:55–12:20	<i>A. Pöhlmann</i> , I. Rehberg, R. Richter	Measuring the onset of the Rayleigh- Taylor instability in rotating magnetic fields	31
12:20–12:45	<i>M. Reindl</i> , S. Odenbach	Influence of magnetic fields on some flow patterns of ferrofluids in a Taylor-Couette system	32
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15:15–15:40	M. Klinkigt, Taisia Prokopyeva, R. Weeber, <i>S.</i> <i>Kantorovich</i> , C. Holm	Suspension of particles with shifted magnetic dipoles	24
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# Taming magnetic snakes in an annular cage

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## Introduction

At a liquid-air interface magnetic microparticles self-assemble to patterns supported by capillary surface waves. Driven by a vertical alternating magnetic field, the microparticles spontaneously form snakelike structures [1]. Large-scale vortex flows propel asymmetric snakes to move around erratically [2]. In the present study the complexity of the pattern is reduced by imposing cyclic boundary conditions in a quasi 1-D geometry.

## Experimental setup

Spherical nickel particles of  $\approx 90 \ \mu \text{m}$  diameter are dispersed on the surface of deionized water. They float due to surface tension. For 2-D experiments a circular glass dish of 80 mm diameter and 9 mm depth is used. The 1-D experiments are carried out in an annular channel milled into a Macrolon disc. The channel has an outer diameter of 74 mm, a width of 5 mm, and a depth of 10 mm. The channel width is designed to be roughly half of the pattern wavelength.

Either the dish or the channel is placed in the midplane of a Helmholtz pair of coils. The frequency of the magnetic field can be adjusted between 50 and 100 Hz, with an amplitude of up to 14 mT. In the experimental area the field is homogeneous with 97 % accuracy. The structures are observed from above by a charge-coupleddevice video camera (Lumenera) which is connected to a computer. For illumination we utilize an electroluminescent film mounted below the transparent vessel.



Figure 1: Sketch of the experimental setup.

### 2-D pattern

A variety of complex and time dependent arrangements of interacting snake patterns can be observed in two dimensions. Figure 2 shows a snapshot. The complexity



Figure 2: Snake pattern for a driving amplitude (RMS) of B = 11 mT, and a driving frequency of f = 60 Hz. Exposure time 10 seconds.

of the patterns is likewise fascinating, and a hindrance for a quantitative description of the basic properties.

# Reduction to 1-D

Due to the channel dimensions (circumference  $\approx 45 \times$  channel width) a quasi 1-D system with cyclic boundary conditions is obtained. This reduces the complexity of the system considerably and enables the determination of the pattern wavelength as a function of the frequency of the magnetic field.



Figure 3: An annular snake pattern in the channel for a driving amplitude (RMS) of B = 9 mT, and a driving frequency of f = 50 Hz. The inset shows an enlarged view of a section in the fourth quadrant.

# Results in 1-D

For driving frequencies ranging from 50 Hz to 130 Hz pictures similar to Fig. 3 have been recorded. For each frame the annular channel is divided into 720 segments. Next the spatially averaged intensity of each segment is plotted versus the azimuthal angle. This yields a cyclic closed variation of the gray scale. From the maximum in the power spectrum of this curve we determine the wave number k. Plotting the driving frequency f versus the wave number k gives the dispersion re-

lation of the magnetically excited surface waves. We compare it with the dispersion relation of water waves in an infinitely deep container

$$\omega^2 = g \, k + \frac{\sigma}{\rho} \, k^3. \tag{1}$$

Here g denotes the acceleration of the earth,  $\sigma$  the surface tension, and  $\rho$  the density of the carrier fluid. In our contribution we discuss the implications and conclusions from this modeling.

# Acknowledgments

We would like to gratefully acknowledge fruitful discussions with A. Snezhko and J. Vollmer.

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# Controlling the shape of supracolloidal assemblies using magnetic fields: towards the generation of highly persistent nanorods

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The emergence of novel materials and processing at the nanoscale has set the conditions for the fabrication of a wide range of nano-objects and multilevel nanostructured networks. In this communication we report a simple and versatile waterborne synthesis of magnetic nanorods following the innovative concept of electrostatic "desalting tran-Highly persistent supermagnetic sition". nanorods are generated from the controlled assembly of oppositely charged nanoparticles and polymers [1, 2]. The rods have diameters around 200 nm and lengths comprised between 1 mm and 1/2 mm, with either positive or negative charges on their surface. The rods are rigid (their persistence length is of the order of 1 meter) and able to reorient via the application of a magnetic field. We also review recent results on the toxicity and uptake of these rods by murine fibroblasts and human lymphoblasts. Our studies reveal that the physico-chemical characteristics of the aggregates play an important role in the interactions with living cells. The rods are finally tested as probes for passive and active microrheology experiments. The mechanical responses of the intracellular medium of cells is presented and compared to that of model viscoelastic liquids.

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# Ferrofluid and Ferroelastomer Based Locomotion Systems – Theory, Experiments and Prototypes

V. Böhm<sup>1</sup>, R. Gärtner<sup>1</sup>, V.A. Naletova<sup>2</sup>, J. Popp<sup>1</sup>, I. Zeidis<sup>1</sup>, K. Zimmermann<sup>1</sup>

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# Introduction

Starting with macro experiments in 2001 first approaches of modelling of peristaltic motion systems, which have the earthworm as a living prototype, were made at the Ilmenau University of Technology. A deficit of these first biologically inspired robots was the low level of autonomy of the systems [1]. In order to work efficiently in pipelines or areas that have been hit by an earthquake, the communication and power supply both need to be wireless. The use of ferrofluids and ferroelastomers whose properties can be controlled using magnetic fields present a new solution to this problem. The initiator of motion in such devices is an alternate controllable magnetic field, which forms to exterior sources (electromagnetic system or motion permanent magnets).



Fig.1 A ferrofluid filled capsule (left) and an artifical ferroelastomer worm (right) at different moments in a travelling magnetic field

The type of locomotion realized in 2004 with the elastic capsule filled with ferro-

fluid [2] and the magnetic elastomer [3] is a snake-like motion called 'concertina motion' (Fig. 1). We like to emphasize that our work follows first mechanical methods and mathematical framework and after that practical experiments. Hence, we start our research with a well-founded mathematical theory, including numerical simulation, which give us the answer to the question: Why does it moves?

Thus, in connection with the issue of peristaltic motion, the problem of a plane flow of an incompressible viscous magnetic fluid layer along a horizontal surface in a non-uniform magnetic field is currently analyzed [4].

# Ferroelastomer Based Locomotion Systems with integrated micro-coils

The use of ferroelastomers allows the realization of simple locomotion systems without internal moving parts, wear and friction (Fig. 2).



Fig.2 Selected realized ferroelastomer based locomotion systems - a: prototype for locomotion in the plane with 6 electromagnets; b, c: uniaxial locomotion systems with one micro-coil

The locomotion of these systems is caused by a periodic deformation (generated by integrated micro-coils made by Würth Elektronik GmbH & Co. KG) of a compliant magneto-sensitive elastomeric body. A periodically alternating magnetic field causes an oscillation of the compliant body, the friction force between the supporting surface (ground) and the system changes periodically and locally repeating. The oscillation mode can be variied in dependence of the pulsing frequency in a wide range.

The qualitative description of the mechanical performance of the systems was provided by transient dynamical analyses by means of the Finite Element Software package ANSYS. Fig. 3/a shows exemplarily the obtained results focusing on the velocity of the centre point "vm" of the system in Fig. 2/a for symmetric electromagnetic excitations and Fig. 3/b selected results for the displacement of the centre of mass of the system in Fig. 2/b in dependence on the actuating frequency.



Fig. 3 Average velocity of the system's centre point in Fig. 2/a and displacement of the centre of mass of the system in Fig. 2/b in dependence on the actuating frequency

The numerical simulations prove the locomotion of the prototypes.

The results of the theoretical and experimental analyses show, that it is possible to realize ferroelastomer based uniaxial and planar locomotion systems where the direction of locomotion can be controlled on a simple way by means of the actuating frequency.

# Conclusion

Based on theoretical investigations, several prototypes of ferroelastomer based uniaxial and planar locomotion systems with integrated micro-coils were realized. The prototypes achieve maximal velocities up to 15 mm/s.

# Acknowledgments

This work was supported by the Deutsche Forschungsgemeinschaft (DFG, Zi 540/11-1 and Zi 540/11-2).

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# High magnetization magnetizable fluids in rotating seals

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Magnetic fluids for sealing applications have to be tailored in such a way to ensure high magnetization, low viscosity, low or very low vapor pressure and excellent colloidal stability in intense and strongly nonuniform magnetic field. These conditions are difficult to be satisfied all at the same time. In some cases to overcome large pressure differences, the magnetization of the sealing fluid has to be increased an order of magnitude compared to usual magnetic fluids.

Extremely bidisperse magnetizable fluids, in particular micron range iron particles dispersed in a ferrofluid [1,2], due to increased magnetorheological effect compared to usual MR fluids [3], offer new solutions for magnetorheological dampers. and in some particular cases also for low rotation speed magnetic seals. Very high magnetization nano-micro composite fluids were prepared [4], which show good stability in magnetic field [5]. The magnetization curves for a high magnetization nanofluid (MF/UTR40 Fe3O4), as well as for a series of CMFs obtained from this magnetic fluid used as carrier for micron sized Fe particles, are given in Fig.1.



Fig.1. Magnetization curves of MF and CMF samples, fitted with Fröhlich-Kennelly relationship;

In Fig. 2. the sealing capacity, i.e. the burst pressure measured for a single stage MF seal, is given for increasing values of the saturation magnetization Ms.



Fig. 2. sealing capacity of various sealing magnetic fluids and CMFs (D1, D3)

In Figs. 3 and 4 two examples of applications of high magnetization fluids are represented for a combined mechanicalmagnetofluidic rotating seal and for a pressurized gas valve.





Fig. 3. Mechanical- magnetic fluid combined seal for liquefied gas pump

1 - shaft; 2 - mechanical seal; 3 - magnetic fluid seal; 4 - inlet for cooling and lubrication fluid; 5 - system for escaped process fluid evacuation



Fig. 4. Gas valve equipped by magnetic fluid or CMF seal; 1 - Body; 2 - Valve seat;

3 - Guide; 4 - Magnetic liquid (or CMF)
seal; 5 - Bar (Valve stem); 6 - Stuffing box;
7 - Valve Handle; 8 - Screw; 9 - Spiral
elastic suspension pin;

# Acknowledgments

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# Competing spikes in the Rosensweig instability

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The horizontal free surface of a magnetic liquid pool turns unstable when the strength of a vertical and static magnetic field exceeds a threshold. The instability, usually called the normal field instability or the Rosensweig instability, is accompanied by the formation of liquid spikes arranged in hexagonal or square patterns [1]. In large diameter pools the main issue is to illuminate the mechanism of selection between regular hexagonal or square patterns. In smaller diameter pools, which can only accommodate a few spikes, the question under study is their number and their relative position. Single spikes compete against 2-, 3-, or 4-fold spike arrangements and their appearance is affected by the pools size, the field strength and its variation speed, as experiment showed - cf. Fig. 1.

The underlying mechanisms of single or multi-spike formation as well as the symmetry of the entire deformation are studied by solving the equations of capillary magnetohydrostatics. They give rise to a nonlinear free boundary problem, which is discretized with the Galerkin/finite element method in two- or three- dimensional domains [3]. The field distribution and the free surface shape are computed simultaneously in a compact numerical scheme based on Newton iteration. Realistic predictions require at least medium scale computations which reveal the multiplicity of the emerging equilibrium deformations near the onset of the instability – a case of multiple bifurcation [3]: branches of solutions which carry deformations shapes with 2-, 3-fold and 4-fold symmetry all bifurcate at approximately the same critical value of the applied field strength from a solution branch corresponding to axisymmetric deformations. Computational predictions are tested against experimental measurements showing remarkable agreement.



Fig. 1. Free surface deformation of cylindrical magnetic liquid pools of different sizes; D is the diameter of the pool.

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# Wave turbulence on the surface of a ferrofluid

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The archetype of wave turbulence is the random state of ocean surface waves, but it appears in various systems over a wide range of length scales: Alfvén waves in solar winds, atmospheric waves, spin waves in magnetic solids. Wave turbulence theory, also called weak turbulence, assumes that the transfer of energy is governed by resonant interactions between waves leading to an energy cascade from large (forcing) scales up to small (dissipative) ones. Although this weakly nonlinear theory provides analytical solutions for the wave energy spectrum as a power law of the spatial scale [1], few well controlled laboratory experiments exist. Recent experimental studies of wave turbulence have shown new observations such as intermittency, fluctuations of the energy flux, and finite size effect of the system [2]. Several theoretical questions are open, notably about the validity domain of the theory and the possible existence of solutions for non dispersive systems. In this context, finding an experimental system where the dispersion relation of the waves could be tuned by the operator should be of primary interest to test the wave turbulence theory.

In contrast with usual fluids, the dispersion relation of surface waves on a ferrofluid depends on the amplitude of the applied magnetic field [3]. Thus, one can easily tune the dispersion relation of surface waves from a dispersive to a non dispersive one with just one single control parameter. We will talk about the first observation of magnetic wave turbulence on the surface of a ferrofluid submitted to a normal magnetic field [4]. We have shown that magnetic surface waves arise only above a critical field. Their power spectrum displays a frequency-power law which is experimentally shown to involve a 4-wave interaction process. The existence of the regimes of gravity, magnetic and capillary wave turbulence is reported in the phase space parameters as well as a triple point of coexistence of these three regimes. Most of these features are understood using dimensional analysis and the dispersion relation of the ferrohydrodynamics surface waves.

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# Experimental study of the behavior of a thin horizontal ferrofluid layer on a liquid substrate under the action of magnetic fields

C. A. Bushueva<sup>1</sup>, K. G. Kostarev<sup>1</sup>

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# Introduction

The work presents the results of experimental investigation of a two-laver liquid system with a stable rupture of the upper layer caused by the action of ponderomotive forces. Such a system has only recently been explored [1]. A distinguishing feature of the system is that the layer rupture exists in the absence of any external force and has the shape of a circle, inside which the fluid of the lower layer comes into contact with gas. Due to the absence of contact with solid surfaces the rupture is highly sensitive to the external effects and therefore can deform and move even under the action of arbitrarily small forces. Thus, in the case when the upper layer is formed by a ferrofluid, the application of the external magnetic fields to the system can cause a displacement of the rupture and a change in its shape. In this case the magnetic field orientation and the degree of its homogeneity influence on the surface deformation of a thin horizontal ferrofluid layer placed on a liquid substrate [2].

The preliminary experiments revealed the dependence of the diameter of rupture on the thickness of the breaking layer and the dependence of their critical values on the cuvette size in the absence of the magnetic field. Due to the dependences received we came to a conlusion that the behavior of ruprure of ferrofluid is comparable to that of other homogenious liquids.

The subsequent experiments showed, that the magnetic field had a twofold effect: it could generate a rupture by deforming the integrity of the layer and could close a stable rupture of the ferrofluid layer.

## Horizontal magnetic field

The use of the magnetic field applied parallel to the layer surface caused a redistribution of ferrofluid along the field direction. This led to a displacement of the rupture with respect to the cuvette center which was accompanied by the change of its shape from a circle to that of an ellipse. With a further increase in the field intensity the rupture rapidly approached the cuvette wall and took the form of a trapezoid provided that the initial diameter of the rupture was larger than the cuvette radius. In the opposite case, with increase of the field intensity the rupture took the form of a "drop" and then closed.

The results of our studies are well consistent with data obtained in [3], describing the deformation of a single gas bubble in the volume of magnetic fluid in the transparent thin duct under the action of drug and magnetic forces. Unlike this experiment two-layer horizontal system let to avoid the effect of gravity in our experiment.

# Vertical magnetic field

The non-uniform magnetic field directed perpendicular to the layer surface could have a twofold effect: it could deform the stable rupture by increasing its diameter or provoke rupture formation in the case of layer integrity. In this situation, application of the magnetic field caused first a redistribution of the ferrofluid in the upper layer. Then, as the field intensity increased to the critical value, the layer deformation spread deep into the layer until it reached the surface of the liquid substrate. At this point there occurred a threshold rupture of the layer in the form of a circle, the diameter of which is determined by the initial layer thickness, the ferrofluid density and the difference in the surface tension of the fluids.

The use of the uniform magnetic field directed perpendicular to the layer surface provokes the instability [4] of the ferrofluid layer. In the case of an entire ferrofluid layer the application of the vertical magnetic field leads to the appearance of the relief in the form of hexagonal cells, which, as soon as the magnetic field reaches the critical intensity, form separate drops. A further increase of the field intensity transforms the drops into the cones submersed into the fluid of the substrate. The wave number of the arising drop system decreases monotonically with the growth of the initial layer thickness.

Application of the magnetic field to the layer with already existing stable rupture also leads to the formation of periodic structures. However, in this case the evolution of the layer deformation depends on the dimensions of the rupture.

Note, that along with the initial size of the rupture the value of ferrofluid magnetization is also of considerable significance for evolution of the layer deformation. Thus, the formation of drop structures in a ferrofluid with lower magnetic susceptibility is observed at much higher values of the field intensity.

During our investigation we determined the dependence of different characteristics of ruptures and drops on the field intensity and the value of ferrofluid magnetization.

# Conclusion

It has been found that a non-uniform magnetic field normal to the layer surface can cause the deformation and rupture of the layer. As soon as the action of the field ceases the rupture can collapse or remain open if the initial layer thickness is below the critical value. However, the existing stable rupture of the layer can be closed by a homogeneous magnetic field applied to the layer in the longitudinal direction. Application of the normal uniform magnetic field leads to the development of the ordered structures due to destruction of a horizontal layer of.

# Acknowledgments

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# The effects of magnetic fields on the microscopic structures of ferrofluids and ferrogels

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The effects of magnetic fields on the microscopic structures of ferrofluids and ferrogels are examined using computer simulations. Simple microscopic models capture the essential characteristics of magnetic nanoparticles and the suspending medium (fluid or gel). Monte Carlo and molecular dynamics simulations of these models yield macroscopic properties in good agreement with experiment and simultaneously offer unique insights on the microscopic organisation of the particles. Three situations will be discussed: the generation of highly complex structures in a ferrocolloid monolayer using a perpendicular magnetic field, the determination of particle-size polydispersity in a ferrofluid using the magnetisation curve, and the mechanical anisotropies of ferrogels induced by uniform magnetic fields.

# Synchronization phenomena in dipolar systems

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Synchronization plays an important role in nature [1]. Far from full list includes pacemaker cells, colonies of fireflies [1], magnetic dipoles in rotating field [2,3]. Recently it was found that synchronization phenomena play an important role in a swimming of microorganisms [4].

There are similarities which exist between the synchronization phenomena in living and dipolar systems. So, for example, infinite period bifurcation [1] observed for magnetic dipoles in a rotating magnetic field [2,3] leads to a step-like behavior of the dipole phase angle near the critical frequency if the thermal fluctuations are present [5], similarly to that observed in the beating of flagella [4]. Synchronization of the rotation of dipolar particles causes a liquid flow due to the "negative viscosity effect" in the suspension of dielectric particles in a liquid with low conductivity [5] or magnetic liquid in an AC magnetic field [6]. Synchronized rotations of dipoles cause liquid flow [7] similarly to ciliated microorganisms [8].

Here we study the synchronization of two interacting dipoles floating on the surface of liquid in an AC magnetic field. Equations of motion of two magnetic dipoles  $\vec{m}_{1,2} =$  $m\vec{n}_{1,2}$  with radius vectors  $\vec{r}_1$  and  $\vec{r}_2$  in the plane with normal  $\vec{\nu} = (-\sin \alpha, 0, \cos \alpha)$  in the vertical AC field are

$$-\zeta \vec{n}_1 \times \dot{\vec{n}}_1 + m\vec{n}_1 \times \vec{H} + m\vec{n}_1 \times \vec{H}_{12} = 0 ,$$
(1)
$$-\zeta \vec{n}_2 \times \dot{\vec{n}}_2 + m\vec{n}_2 \times \vec{H} + m\vec{n}_2 \times \vec{H}_{21} = 0 .$$
(2)

Here

$$\vec{H}_{12,21} = -\frac{\vec{m}_{2,1}}{r_{12}^3} + \frac{3\vec{r}_{12}(\vec{m}_{2,1}\cdot\vec{r}_{12})}{r_{12}^5}$$

is the magnetic field strength acting on the particle and created by its neighbor and  $\vec{r}_{12} = \vec{r}_1 - \vec{r}_2$ .

The behavior of dipoles besides the inclination angle of the liquid interface  $\alpha$  is controlled by two parameters  $\varepsilon = (\omega \tau)^{-1}$ , which characterizes the ratio of the period of the AC field and the characteristic relaxation time of the dipoles  $au\,=\,\zeta(mH_0)^{-1}$  and the parameter  $\lambda = m(r^3H_0)^{-1}$  that characterizes the ratio of the magnetic field of neighbor particle to the amplitude of an applied external field. For small  $\lambda$  there are two time scales in the problem and time averaging may be applied. Analysis of the slow variables in dependence on time in the case of simple model ( $\vec{n}_{1,2}$  are confined to the plane defined by vectors  $\vec{\nu}$  and  $\vec{H}$ ) shows that for small  $\varepsilon$  dipoles synchronously oscillate with the frequency of the AC field. At  $\varepsilon \simeq 1.265$ ( $\alpha = \pi/4$ ) the behavior changes qualitatively and regime of synchronous rotation of dipoles by the infinite period bifurcation appears. The time dependence of the azimuthal orientation angle of dipoles is shown in Fig.1. Period of synchronous rotation in dependence on the parameter  $\varepsilon$  is shown in Fig.2. By comparison Fig.1 and Fig.2 one may see that a duration of oscillations is in agreement with the period of synchronous rotation regime.

Analysis of the general case shows that the regime of confined synchronous rotation of the dipoles is unstable in some intermediate range of the parameter  $\varepsilon$ . In this regime the azimuthal angles are synchronized as for particular initial conditions it is illustrated in Fig.3.

The developed approach is applied to an ensemble of the dipoles and arising flow of a liquid is analyzed.



Figure 1: Synchronous rotation regime at  $\varepsilon = 2$  and  $\lambda = 0.01$ . Step-like oscillations are due to slow change of angle in the bottlenecks between the null isoclines for slow variables.



Figure 2: Period of synchronous rotation in dependence on the parameter  $\varepsilon$ . Dashed line - the simple model of the confined synchronous rotation of the dipoles. Solid line according to the full set of equations for the slow variables ( $\alpha = \pi/4, \lambda = 0.01$ .)

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Figure 3: Synchronization of the azimuthal orientation angles for some arbitrary initial conditions.  $\varepsilon = 1.4$  and  $\lambda = 0.01$ .

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# Mechanical properties of magnetic nanowires

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The dialysis of maghemite nanoparticules ( $\gamma$ -Fe<sub>2</sub>O<sub>3</sub>) and oppositely charged polymers, under a magnetic field, leads to the formation of one-dimensional anisotropic aggregates. These aggregates are nanowires with diameters around 400 nm and lengths comprised between 1 and 100 µm. It was shown recently that the nanowires are superparamagnetic, *i.e.* they acquire a macroscopic magnetic moment in the presence of an external magnetic field [1,2]. The wires are also characterized by enhanced mechanical properties, which are investigated in detail in the present communication.



Figure 1: Superimposition of two microscopic images (top view) of a nanowire stuck on a cover slip in presence and in absence of magnetic field. With the magnetic field applied (B = 28.3 mT), the wire bent toward the field direction.

The flexural rigidity of nanowires was studied by optical microscopy using a home-made set-up mounted on an inverted microscope. The wires are fixed by one extremity on a glass substrate, the other part dangling in the solvent. A constant magnetic field (B = 10 - 50 mT) is then applied to the wire, and the deflection is recorded as a function of the field orientation and intensity (figure 1). By analogy with polymers, the bending modulus of the nanowire expresses as : C = EI, where E is the Young modulus and I the second moment of inertia. The persistence length  $L_P$  is then calculated from the equation  $L_P = C/k_BT$ . Writing the equality between the curvature and magnetic energies of a bent nanowire at equilibrium, an expression of the contour length is determined and compared to the experimental data. The agreement between theory and experiments is excellent and allows to retreive values of the persistence lengths ( $LP \sim 2 \text{ m}$ ) and Young modulus (E = 300 MPa). This high rigidity has been confirmed by fluctuation measurements. In conclusion, we anticipate that these wires represent a promising class of nanomaterials for cell manipulation and microrheology.

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# Numerical simulation of the conical meniscus of a magnetic fluid in the field of a current-carrying wire

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### Introduction

Placing a vertical current carrying wire in a horizontal layer of magnetic fluid forms a simple, but striking setup to test the capabilities of a magnetic field to influence the free surface of the magnetic fluid. Analytical [1] as well as experimental studies [2] have already been done. These studies indicate the strong influence of the contact angle  $\phi_1$ , formed by the fluid at the rim of the wire, on the shape of the meniscus.

The analytical study was done without taking into account the conservation of mass because an unlimited size of the system was considered in [1]. The experimental study [2] in turn did not show the whole surface profile of the meniscus due to the experimental setup. Both drawbacks can be circumvented in a numerical study presented here. Additionally, the influence of the contact angle  $\phi_2$  of the fluid at the wall of the beaker can be studied numerically, too. This quantity has neither been examined in [1] nor in [2].

### **Numerical Model**

The numerical simulation was done by the commercial software FLUENT. The used numerical model is based on the experimental setup in [2]. The Kelvin force density

$$\mathbf{F}_{\mathbf{K}} = \mu_{\mathbf{0}} \left( \mathbf{M} \operatorname{grad} \right) \mathbf{H} \tag{1}$$

was implemented as a source term in the Navier-Stokes equation by using so called "User Defined Functions". Figure 1 shows a surface profile of the meniscus for two different contact angles  $\phi_2$ . Particularly the profile for  $\phi_2 = 40^\circ$  indicates the far reaching influence of  $\phi_2$  over the entire range of the surface.



Figure 1: Surface profile  $\xi(r)$  against the distance from the wire with R = 0.95 mm for the magnetic fluid EMG 909. The dotted line shows the profile for  $\phi_2 = 40^\circ$  and the triangles the profile for  $\phi_2 = 90^\circ$ .

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# Magnetic spatial forcing of a ferrofluid layer

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### Introduction

Historically, spatial forcing of a pattern forming system was first studied experimentally in electroconvection [1]. More recently, inclined layer convection was measured under the influence of lamellar surface corrugations [2]. In both cases, stripes are the first convection pattern beyond a threshold.

The Rosensweig instability in a layer of ferrofluid can provide a primary instability to hexagons if a homogeneous magnetic field normal to the flat surface is applied [3, 4]. In case of a tilted magnetic field, a primary instability to stripes can be observed [5]. As a consequence, switching between these two basic types is possible in one single system. We explore how both configurations respond to a stripe like modulation of the magnetic induction.

### Setup

To apply the spatial modulation, a grid of live wires (see Fig. 1) generates an inhomogeneous magnetic field, which can be superimposed to the homogeneous field of a Helmholtz pair of coils. This way we get a sinusoidal magnetic field with independently tunable offset and amplitude. The wavelength is given by the design of the lattice. Figure 2 shows an example of the spatially modulated field measured using a hall probe, moved along the arrow sketched in Fig. 1.

To detect the fluid's response to the different configurations of magnetic fields, an x-ray imaging technique [6] is used. The recorded data is analysed by means of a 2D-FFT.



Figure 1: Photograph of the grid of wires to produce the spatially modulated magnetic field. The arrow indicates the path of the hall probe for the field measurement shown in Fig. 2.



Figure 2: Measured spatial modulation of the magnetic field.

## Stripes vs. Hexagons

The impact of stripe-forcing on a system with a primary instability to hexagons [7] has not been measured so far. We apply a static magnetic stripe-forcing to the Rosensweig instability and demonstrate first experimental results.

# Stripes vs. Stripes

The tilted field instability which causes a stripe pattern of liquid ridges, can also be forced with another kind of stripes, enabling various scenarios. For example, the ridges of the tilted field instability can be forced with a spatially modulated magnetic field with a wavevector parallel to the ridges, but different from the intrinsic one. In this way comensurate to incomensurate transitions can be explored.

Moreover the direction of forcing can be oriented perpendicular to the liquid ridges. This will favour a rectangular pattern instead of a hexagonal one.

All in all, surface instabilities in ferrofluids can conveniently be forced in space. In this way the impact to patterns of different symmetry can be explored quantitatively for the first time.

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# Spin-coating of magnetic colloids in a magnetic field

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Fabrication of magnetic thin film devices is gaining research and commercial interests [1, 2]. By varying the size of the magnetic particles, their applications can be classified. Of many ways, self assembly of magnetic particles is interesting due to less complexity during fabrication and comparatively high quality structures [3,4]. Spin coating of colloidal dispersion has a great advantage in producing resultant morphologies at a short span of time [5–7].

By controlling the spinning rate and the initial concentration of the dispersion the kind of final morphologies can be tuned. In this work, we will focus first on the study the spin-coating of micron sized magnetic colloids with external axial magnetic field. The superparamagnetic colloids are obtained from Polysciences, Inc. of concentration 2.52%. They are polydisperse with diameters ranging from 1 to 2  $\mu$ m.

We experimentally investigate the coating process by pouring 120  $\mu$ l of the suspension on a spinning substrate at 2000 rpm. We performed the experiments with magnetic field (0.033 T and 0.066 T) and without it. At these experimental conditions (for applied field), deposits of colloidal particles are submonolayer, mainly formed by clusters of particles. Figure 1 shows the average area  $\langle A \rangle$ (in fact, the area of projection on the substrate) of clusters at several distances from the center of rotation. It can be seen that the mean area of clusters is much bigger than the case of not having an applied field. Also, in the cases where we apply a field, there is a relative maximum due to the deflection of the poured suspension. Thus, it does not hit

onto the center of the rotating substrate and gives a relatively depleted region around the center.



Figure 1: Average area of clusters as a function of distance from the center of rotation.



Figure 2: Number of clusters per unit area as a function of distance from the center of rotation.

Also, we measured the density of clusters at the above mentioned distances (figure 2). In

this case, we also observe a different behavior in the case of having field compared to that without field.

Microscopic image of the dried substrate at an applied field of 0.033 T is shown in figure 3. To compare this situation, we include the corresponding image without applying magnetic field (figure 4). The other experimental conditions are unchanged.



Figure 3: Image of the dried substrate at 4 mm from the center. During the spinning, the applied field was 0.033 T. Scale bar is 0.1 mm.



Figure 4: Image of the dried substrate at 4 mm from the center. Without applied field. Scale bar is 0.1 mm.

We also study the effect of the rotation rate of the substrate in the corresponding formation of colloidal patterns, and we characterize the geometry and orientation of the clusters.

# Acknowledgments

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# Magnetic Properties of Ferrofluid Emulsions: Model of Noninteracting Droplets

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Ferrofluid emulsion is a colloidal suspension of ferrofluid droplets suspended in an immiscible liquid [1,2]. An applied magnetic field induces a magnetic dipole moment in each droplet, since the ferrofluid becomes magnetized. So, the induced droplet magnetic moment is dependent on the droplet volume and the magnetization behavior of the bulk ferrofluid. In case of weak external field, the droplet magnetic moment is a linear function of applied field strength  $H_0$ .

A droplet shape is greatly dependent on the value of the surface tension; all droplets are spherical in zero magnetic fields. For ferro-fluid emulsions with strong surface tension the droplets could be regarded as spherical even in rather strong magnetic field [1,2]. Here the emulsion magnetic permeability  $\mu_e$  might be constant in a wide range of magnetic field strengths. Really, the emulsion magnetic permeability could be defined as

$$\mu_e = 1 + 4\pi \frac{M_e}{H_0} \,. \tag{1}$$

Here  $M_e$  is the emulsion magnetization, which is the product of the droplet concentration  $n_d$  and the induced droplet magnetic moment  $m_d$ :  $M_e = n_d m_d$ , for simplicity we consider the surrounding liquid as totally nonmagnetic. Assuming a linear magnetization, for a spherical droplet we get

$$m_d = \chi_f V_d H_d = \frac{3\chi_f V_d H_0}{3 + 4\pi\chi_f}, \qquad (2)$$

where  $\chi_f$  stands for a ferrofluid initial magnetic susceptibility,  $V_d$  has a meaning of the droplet volume, and  $H_d$  is an internal magnetic field inside the ferrofluid droplet.

Combining expressions (1) and (2), we easily obtain:

$$\mu_{e} = 1 + 4\pi \frac{n_{d}m_{d}(H_{0})}{H_{0}} =$$

$$= 1 + \varphi \frac{12\pi\chi_{f}}{3 + 4\pi\chi_{f}} \qquad (3)$$

This well-known expression for the permeability of a mixture is valid for low concentrated emulsion, and we may call this approach as "the model on non-interacting droplets" because we suggest that droplets do not influence each other.

The behavior is much more interesting for emulsions, the interfacial tension  $\sigma$  on the droplet surfaces of which is rather weak. Here the surface forces cannot stabilize the spherical shape of droplets, and an applied magnetic field tends to elongation of droplets along the filed direction. In a simplest way the droplet shape might be considered as elongated ellipsoid of revolution with major semiaxes *a* and *b*, a > b,  $a \parallel H_0 \parallel Oz$ . The expression (2) converts to

$$m_d = \chi_f V_d H_d = \frac{\chi_f V_d H_0}{1 + 4\pi \chi_f n_z}, \qquad (4)$$

where  $n_z$  is the demagnetization factor of an ellipsoid in the magnetic field direction. Thus, the effective emulsion magnetic permeability is determined as

$$\mu_{e} = 1 + 4\pi \frac{n_{d}m_{d}(H_{0})}{H_{0}} =$$

$$= 1 + \varphi \frac{4\pi\chi_{f}}{1 + 4\pi\chi_{f}n_{z}}$$
(5)

The demagnetization factor of stretching ellipsoid varies from the value  $n_z = 1/3$  (for a sphere, a = b) to zero (for an infinitely elongated ellipsoid,  $b/a \rightarrow 0$ ). For this kind of ferrofluid emulsions the magnetic field induces both the droplet magnetic dipoles and the droplet elongation. And the demagnetization factor is a decreasing function of applied field strength. It means that the effective emulsion permeability increases with magnetic field.

The ellipsoid elongation is determined by the minimization condition for the droplet free energy [3-5], as a sum of surface energy  $F_s$  and magnetic energy  $F_m$ , with respect to semiaxes ratio c = a / b. The resulting elongation is governed by the dimensionless magnetic Bond number  $B_m = (3V_d/4\pi)^{1/3}H_0^{2/4}\pi\sigma$  and the ferrofluid magnetic susceptibility  $\chi_f$ . The well-known distinctive feature of curves  $c(B_m)$  is the occurrence of hysteresis loops in the case of sufficiently high values of  $\chi_f \sim 10$ . In this area the ferrofluid droplet undergoes a jump-like elongation; the main features of such process have been experimentally studied in Refs. [3,4,6]. The ellipsoidal droplet becomes high-elongated (c >> 1)when the Bond number  $B_m$  exceeds a certain value, less or approximately equal to unity. For micron-sized droplets and low surface tension the jump-like elongation could be realized even in weak magnetic fields. This elongation results in rapid growth of the emulsion magnetic permeability (5) due to a decay in droplet demagnetization factor  $n_{z}[c(H_{0})] \rightarrow 0$ .

The following magnetic field strengthening is accompanied by a decrease of ferrofluid susceptibility. And we get the changeover in emulsion permeability behavior: the field induced growth of permeability changes to its decrease. The correct theoretical description of this non-monotone dependence needs the solution of droplet elongation problem with account for nonlinear droplet magnetization.

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# Structure formation and dynamics of dipolar colloids in rotating fields

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Dipolar particles and in particular induced dipolar particles can be brought to self-assemble in a variety of ways. In static fields such particles form chains, in biaxial rotating fields they tend to form layers [1], and triaxial fields can result in a multitude of different structures [2, 3].

We focus on suspensions of colloidal particles with permanent dipoles that are exposed to external rotating fields. We are mainly

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Figure 1: Snapshot of a layered system.

interested in the dynamics of the colloidal particles and the question for which field strengths and frequencies layer formation occurs. To explore these matters, we primarily employ computer simulations, most notably Brownian (Langevin) dynamics simulations, which we supplement by semi-analytical considerations. In particular, we propose a simple theory that describes layer formation in a density functional framework [4].

# Suspensions of particles with shifted magnetic dipoles

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In previous study we investigated the ground state for systems of particles, magnetic moment of which was shifted from the centre of particle, and pointed always outwards radially, using the combination of Monte Carlo simulations and analytical calculations. We showed that antiparallel orientation of moments becomes the most favorable configuration when the shift approaches the particle radius. Thus shift influences greatly the microstructure at low temperatures [1]. Our preliminary analysis of relatively small systems at room temperatures also showed a lot of interesting features.

To study larger systems one have to use molecular dynamic simulation to reach better performance. Therefore, we implemented our model in ESPResSo [4], that allows us to use the  $P^3M$  algorithm for dipolar long range interactions in systems with periodic boundary conditions in three dimensions [2]. Recently implemented MDLC [3] gives us the possibility to simulate monolayers with periodic images in two dimensions.

### Magnetization and cluster formation

The behavior of a suspension of particles with shifted magnetic dipoles in the presence of an external magnetic field is strongly influenced by the shift factor. The tendency of parallel orientation of the magnetic moments for low shift leads to a magnetization curve with large initial susceptibility. Whereas the initial slope decreases for larger shift parameters. For higher magnetic interaction parameter  $\lambda$  for particles with shifted dipole we find initial slopes lower then the associated Langevin curve. To understand the microscopic properties of our system better, we are developing new methods to analyze the clusters which magnetic particles form for different shift values. In the present work we develop the algorithm which allows for the presence of antiparallel pairs and triangular conformations.

### Outlook

Currently, analytical calculations are performed in order to obtain magnetization curves for different shifts using perturbation theory in both 2 and 3 dimensions. We also work on the cluster formation theory. The tunable anisotropy of such a system will allow to control the process of cluster formation at room temperatures and crystallization at low temperatures.

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# Magnetophoretic transport in thermoreversible ferrogels

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Figure 1: Two coils are arranged in a Maxwell configuration, creating a field of constant gradient (symbolized by the dashed line). The sample cell and temperature bath are made of two coaxial plexiglass cylinders.

### Introduction

The viscoelastic properties of thermoreversible ferrogels [1] can be tuned via the temperature, making such gels an exciting type of magnetic soft matter [2]. Preliminary studies with these materials showed a separation effect of gel and ferrofluid. So far magnetophoresis was studied by optical means, and limited to dilute magnetic fluids [3]. With our x-ray based technique [4], we are able to investigate magnetophoretic transport in any kind of magnetic fluid as well as in ferrogels.

# **Experimental Methods**

The setup shown in Fig.1 is used to characterize the magnetophoretic behaviour, such as the transport velocity, the mobility and the saturation effects. The constant field gradient created by the coils can be raised to a maximum value of about 0.75 T/m. In order to measure the density of magnetic particles the absorption of x-rays is used [4]. The sample cell is made of a plexiglass tube with an inner diameter of 0.73 cm. This allows for convenient preparation and non-destructive extraction of the sample after the measurement. The outer plexiglass tube enables temperature control of the setup via a water bath.

### **Ferrogel samples**

Most recently, we were able to synthesize cobalt ferrite hydrogels [5]. These thermoreversible hydrogels have cubic and hexagonal mesophases which are tunable via the temperature. As a consequence, the magnetophoretic transport in different phases can be studied as a function of the viscoelasticity. For a further characterization of the samples, magnetometric and magneto-rheometric measurements have been carried out.

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# Patterns of thermomagnetic convection in magnetic fluids subjected to spatially modulated magnetic fields

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Fluidic systems which are driven out of equilibrium by the temperature dependence of the density of the fluid have been long studied with respect to the emerging patterns and their stability and belong now to the set of classical pattern forming systems [1,2]. By using a magnetic fluid (MF) as working substance an additional ways opens up to generate patterns. Thus, in a horizontal layer of MF subjected to a vertical gradient of temperature and a vertical magnetic field convection can be initiated in two different ways.

The temperature dependent magnetization, whose gradient is antiparallel to the temperature gradient, is that other cause for triggering convective motion. If now a fluid element with magnetization M (hot region) is adiabatically moved to the cold region, where the magnetization  $M - \Delta M$  is present, a difference in the magnetization between the fluid element and the surrounding fluid exist. This difference interacts with the gradient of the inner field to a resulting magnetic force. That force can generate a destabilization of the fluid layer which leads to a convective motion, called thermomagnetic convection. The strength of the force is controlled by the strength of the magnetic field.

An analysis of the thermomagnetic convection in a layer of MF subjected to a spatially and symmetrical modulated magnetic field is presented. For any nonzero magnetic field the base state is a convective one formed by a double vortex which reflects the symmetrical modulation. The influence of long- as well as short-wavelength modulations is discussed. The linear stability analysis [3] reveals that the threshold for the stability of the base state increases with increasing magnetic driving. An analytical expression for the threshold is derived which shows the characteristic feature of sudden jumps of the threshold at particular values of the magnetic driving.



Figure 1: Plot of the double vortex flow field.

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# **Thermal Convection in Viscoelastic Magnetic Fluids**

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The purpose of the present work is to analyze the influence of the viscoelasticity on the convective thresholds in magnetic fluids. To this aim an Oldroyd viscoelastic magnetic fluid heated from below is considered [1,2]. The description of the system involves many parameters whose values have not yet been determined accurately. Therefore, we will consider a broad range of possible parameter values. In order to be as exhaustive as possible, we will analyze the linear regime for two different sets of boundary conditions. For the idealized boundary case, one can explicitly calculate the threshold as function of the parameters of the fluid. In the case of realistic boundary conditions, an analytical calculation is not tractable and we numerically solve the linearized system using a collocation spectral method in order to determine the eigenfunctions and eigenvalues and consequently the convective thresholds. Close to the bifurcation, using the standard nonlinear analysis, we calculate the corresponding amplitude equations for the idealized boundary conditions.

Figure 1 shows the external field dependence of the threshold for realistic boundary conditions. We observe that for strong fields,  $M_1 >> 1$  the slope is -1 (dotted line). Finally, we remark that the onset of the oscillatory instability (not possible for Newtonian fluids) also strongly depends on the viscoelastic properties.



Figure 1: The critical oscillatory Rayleigh number,  $Ra_{oc}$ , as a function of  $M_1 \sim H^2$ .

### Acknowledgments

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# Numerical study of the Rosensweig instability in a magnetic fluid subject to diffusion of magnetic particles

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The present study is devoted to the Rosensweig instability, taking into consideration the particle inhomogeneity in the magnetic fluid. The spatially varying particle concentration is an unknown quantity of the model, dependent on the magnetic field inside the fluid with a free surface.

The theoretical model with no interparticle interactions is considered.

# **Mathematical Model**

We consider a semi-infinite magnetic-fluid layer with a horizontal plane free surface bounded from above by air. The particles are uniformly distributed inside of the flat fluid layer with the constant volumetric concentration  $C_0$ . The system is regarded under the action of gravity (0, 0, -g) and a uniform magnetic field normal to the plane free surface of the layer  $(0, 0, H_0)$ .

A simplified mathematical model can be derived for a single peak in the pattern under the assumption of axial symmetry. It allows us to formulate the model in cylindrical coordinates in a domain  $[0, a] \times [-\infty, +\infty]$ , where *a* denotes half of the distance between two nearest peaks in the pattern.

The magnetic field in the fluid is described by the Maxwell's equations

$$\nabla \times \mathbf{H} = \mathbf{0}, \, \nabla \cdot \left[ \left( 1 + \frac{M(H,C)}{H} \right) \mathbf{H} \right] = 0$$

with the use of the one-particle model for the equilibrium ferrocolloid magnetization

$$M(H,C) = \frac{C}{C_0} M_s L(\gamma H), \ \gamma = \frac{3\chi}{M_s}.$$

Here  $L(t) = \operatorname{coth}(\xi) - 1/\xi$ . This model is valid for dilute ferrofuids [1].

An explicit analytical solution for the steadystate particle concentration problem was constructed in [3]

$$C = \frac{\psi(\gamma H)V}{\int\limits_{\Omega} \psi(\gamma H)d\Omega}, \ \psi(t) = \frac{\sinh(t)}{t} \quad (1)$$

for the fluid domain  $\Omega$  of volume V. Another way to get (1) is an application of the Boltzmann distribution, valid for a gas of non-interacting particles [2].

Equilibrium surfaces are defined by the generalized Young-Laplace equation

$$\sigma \mathcal{K} = -\rho g z + \frac{\mu_0}{2} \left( M \frac{H_n}{H} \right)^2 + \mu_0 \int_0^H M dH + c.$$

Here  $\sigma$  is the surface tension coefficient,  $\mathcal{K}$  the sum of principal curvatures,  $\rho$  is the fluid density,  $\mu_0$  the magnetic constant.

The concentration, defined by relation (1), influences to other unknown quantites only through the magnetization. It brings additional nonlinearities to the field und surface subproblems.

## **Numerical Treatment**

An initial free-surface configuration is chosen as a small perturbation of the plane surface with an amplitude of around 1 % of the wavelength. A critical value of the pattern wavelength, predicted by the linear stability analysis [4],  $\lambda = 2\pi/\sqrt{\rho g/\sigma}$ , is used in the model for any applied field intensity  $H_0$ . The magnetostatic problem is solved by a finite element method in a bounded domain for the given field far from the fixed fluid-air interface. The next step is a solution of the Young-Laplace equation for the last computed field. This is realized by a finite-difference method, based on strategy in [5].

Switching iteratively from the field to surface computations, the process can show a damping of the initial perturbation or can converge to a solution with a curved surface. The latter case is interpreted as the onset of the Rosensweig instability.

## Numerical Results

Numerical calculations were performed for the magnetic fluid EMG 901 ( $\chi = 2.2$ ) with the magnetic phase volume fraction of 10 %. A dimensionless applied field  $\gamma = 3\chi H_0/M_s$ is chosen as a control parameter.



Figure 1: Free-surface shapes for nonuniform (solid) and uniform (dashed) concentration and isolines of the potential.

The Figure shows that the non-uniform particle distribution results in a 20% higher peak amplitude in comparison to a homogeneous fluid. The main inhomogeneity of the particle distribution occurs in the peak region. The relative concentration takes a value at the peak top which is about 25% greater than in the fluid bulk. The concentration increases at places with higher magnetic intensities and takes the smallest value at the peak foot. The magnetic fluid becomes uniform over the fluid volume for z/a < -1. The particle diffusion mechanism is absent there and the relative concentration is constant.

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# A Theoretical Understanding of Two-Dimensional Localised Patterns: Challenges and Mysteries of Ferrosolitons and Localised Ferropatterns

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In this talk, we will explore the theoretical understanding of two-dimensional localised patterns seen in the Swift-Hohenberg equation and other reaction-diffusion systems; see [1, 2, 3, 4, 5]. In particular, we will look at how these results translate to the ferrofluid problem to understand ferrosolitons and cellular hexagon patches of ferrofluid. However, it is found there is a problem with such a straight forward translation and I will present the theoretical challenges these structures present.

### Acknowledgments

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# Measuring the onset of the Rayleigh-Taylor instability in rotating magnetic fields

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If a dense fluid is supported by a less dense fluid, the flat interface separating them is subject to the Rayleigh-Taylor instability [1–3]. The interface tension between the fluids suppresses the growth of all unstable modes with wavenumbers greater than the critical one. This gives rise to a maximum experimental interface diameter (size of boundary) at which the flat interface is stable.

In the case of one of the fluids being magnetic, an azimuthally rotating magnetic field can be used to stabilize modes with wavenumbers smaller than the critical wavenumber [4]. This allows for the preparation of the flat interface with greater interface diameters. When switching off the field, all unstable modes, which are not suppressed by the size limitation of the experiment, start to grow. Consequently, a precise study of the Rayleigh-Taylor instability should be possible.

In our experiment a magnetic fluid is covered by a more dense transparent one. The flat interface is stabilized by a rotating magnetic field (rotation in x-y-plane). The size of the interface is restricted to a circular shape of certain diameters. For different diameters the field strength is lowered to a threshold level, so that the stability boundaries can be observed. In the instance of switching off the magnetic field, the dynamics can be measured.

The stability boundaries (Fig. 1) can be calculated by feeding our material parameters



Figure 1: Stability diagram at f=50Hz for different magnetic fields and wavenumbers of surface modes. Gray color marks the unstable regime.

in the calculation of Ref. [4]. We measure the onset of the instability as a function of driving frequency and amplitude and compare it with the predictions.

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# Influence of magnetic fields on some flow patterns of ferrofluids in a Taylor–Couette system.

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### Motivation

The flow behaviour of ferrofluids under influence of magnetic fields was subject to numerous rheological investigations already. However, a typical rheometric setup, featuring a cone-plate measuring cell which is exposed to a magnetic field, offers only a very basic flow structure [2]. The question of a direct influence of magnetic fields on more complex flows of ferrofluids is thereby left open. In this paper, this question is highlighted by the investigation of typical flow structures appearing in a Taylor-Couette system. A Taylor-Couette system, which consists of two rotary concentric cylinders, where the gap in between is filled with a fluid, offers many different flow regimes which can be studied [1].

### Experimental

The fluid cell of our setup is subjected to homogeneous axial and transverse magnetic fields and the axial velocity components of the flow are measured using ultrasound— Doppler velocimetry. The ultrasound transducer is mounted on top of the fluid cell near the outer cylinder wall [4].

If the inner cylinder rotates slowly, below a critical rotation rate  $\omega_{1c}$ , the fluid near the vertical center of the fluid gap may be considered as moving in an azimuthal direction around the inner cylinder. With the axial velocity components being absent, this results in a "zero signal" at the measurement device. This is the ground state flow, called circular Couette flow (CCF).

If then the inner cylinder rotates faster than  $\omega_{1c}$ , TVF is present. Here, the fluid flows in helical lines around the inner cylinder. The axial velocity components of the vortices then yield a sinusoidal velocity profile, due to the flow moving upwards and downwards at the outer cylinder wall.

The flow pattern of a Taylor vortex flow of finite length always consists of pairs of vortices [5]. Hence, the wavenumber k, as a dimensionless measure for the axial elongation of such a pair of vortices, is an important property of such a flow pattern like the Taylor vortex flow.

# Influence of an axial magnetic field on the wave number

Normally, the vertical dimension of one vortex should be close to its horizontal one, which is the gap width d [5]. Since the system presented in this work has an aspect ratio of 20, we would expect a flow pattern consisting of 10 vortex pairs for a non-rotating outer cylinder and zero magnetic field. According to equation (1), this yields a wave number of k = 3.14.

Here, the wavenumber k was obtained by counting the vortex pairs over the depth of the gap and using equation (1), which scales the wavelength with the gap width. A dimensionless wave number

$$k = \frac{2\pi d}{h} \cdot n \tag{1}$$

is thus obtained, where n is the number of vortex pairs and h is the height of the gap. However, a wave number that is stable at a given (or zero) axial magnetic field and certain rotation rates of the cylinders, did not necessarily remain stable if the field was subsequently increased or decreased. Generally speaking, with an increase in axial magnetic field strength, lower wave numbers (i.e. a flow pattern with less vortices) showed to be preferred over higher ones [4].

This connection between the axial wave number and an axial magnetic field is qualitatively the same for a resting or rotating outer cylinder, but gets shifted to higher wave numbers (counterrotating outer cylinder) or lower wave numbers (corotating outer cylinder). The surfaces in Fig. 1 give a qualitative idea which wave numbers are stable at certain combinations of the Reynolds–number of the outer cylinder  $R_2$  and magnetic field.



Figure 1: (Color online) Surface plot of possible stable wave numbers k at different axial magnetic field strengths H and rotation rates of the outer cylinder  $R_2$ . The black circles denote those experimental data points that were recorded to file.

# Outlook

Beside the influence of magnetic fields on the basic Taylor–vortex flow, the influence on axially propagating spiral vortices and azimuthally rotating wavy vortices are presented in detail at the conference.

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# The growth of localized states on the surface of magnetic liquids

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### Introduction

Static liquid spikes are emerging on the free surface of a magnetic fluid (MF), when a critical value  $B_c$  of the vertical magnetic induction is surpassed. This instability, first reported by Cowley and Rosensweig [1] has become a landmark for ferrofluid research. Moreover it is of particular interest for the field of pattern formation, as demonstrated in a recent review of Rosensweig patterns [2]. The formation of a hexagonal pattern of spikes is commonly explained as a resonant interaction of three degenerate wave modes [3]. A different, more "atomistic" approach to pattern formation regards the hole lattice as a crystal where an individual Rosensweig spike stands for an atom [4]. From this the question arises, whether a spike can also exist independently from the extended pattern.

Such localized states find currently great interest [5]. Examples comprise oscillons in shaken fluids [6] and granular materials [4], or cavity solitons in optics [7]. For the stability of these 2D-structures a balance of dissipation and energy gain is essential [4], while the localized spikes, or "ferrosolitons" [8], persist without permanent energy dissipation.

Apart from a fundamental interest in their being and stability, "ferrosolitons" open also up a new way to investigate growth dynamics of surface structures. Whereas for regular Rosensweig patterns one has to develop special techniques [9], here a lateral optical observation is not hindered by surrounding spikes. Thus the growth



Figure 1: Growth of a ferrosoliton after switching on the probe coil at t = 0 ms.

of "ferrosolitons" can conveniently be recorded by a high speed camera, as shown in Fig. 1.

# Scaling of the delay times

Applying an overcritical induction  $B_{\rm p} > B_{\rm p,c}$  to a probe coil mounted beneath the vessel [8] ignites a "ferrosoliton" in the bistable range of the Rosensweig instability. As shown in Fig. 2(b) we observe a delayed growth, characterized by the delay time  $t_{\rm d}$ .

We record a series of amplitude curves for increasing values of  $B_{\rm p}$ . In Fig. 3 we plot  $t_{\rm d}^{-2}$  versus the rescaled probing induction  $\hat{B} = (B_{\rm p} - B_{\rm p,c})/B_{\rm p,c}$ . The data can well



Figure 2: Growth measurement of a "ferrosoliton". (a) Pulse sequence of the probe coil. (b) Time resolved amplitude of a spike resulting from the pulse sequence in chart (a) with  $B_{\rm H} = 9.0 \,\mathrm{mT}$ and  $B_{\rm p} = 1.1 \,\mathrm{mT}$ . The delay time  $t_{\rm d}$  is indicated by horizontal arrows.

be fitted by the scaling law

$$t_{\rm d} = a \cdot \hat{B}^{-1/2},\tag{1}$$

where *a* is a fit parameter. The delayed growth and its scaling (1) is characteristic near a saddle-node bifurcation. Such a bifurcation can be induced by an imperfection added to the transcritical bifurcation describing the instability of a rotationally symmetric spike. In the experiment the imperfection is created by  $B_{\rm p}$ . In numerical investigations [10] we corroborate this bifurcation scenario together with the scaling law (1) and the measured stability diagram  $B_{\rm p,c}(B_{\rm H})$ .

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Figure 3: The delay times determined according to Fig. 2 for  $B_{\rm H} = 8.71 \,\mathrm{mT}$ . A fit of the data with Eq. (1) yields the solid line, with  $a = (45.0 \pm 0.3) \,\mathrm{ms}$ .

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# On the estimation of the ferrofluids particle size distribution on the acoustomagnetic effect base

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The technique considered is based on the analysis of the field dependence of the acoustomagnetic effect (AME), which lies in the investigation of an electromagnetic wave using a column of a magnetized ferrofluid (FF), in which the sound wave propagates in [1–3]. The EMF induced in the circuit is proportional to the amplitude of oscillations of fluid magnetization due to variations in concentrations of dispersed phase particles.

According to the theory of superparamagnetism the amplitude of the induced EMF can be represented in relative units as a function of the parameter [4]:

$$\beta_{\xi} = \frac{L(\xi) - \xi^{-1} - k'\xi D(\xi)}{1 + k''\xi^{-1}D(\xi)},$$

where  $D(\xi) = (\xi^{-1} - \xi s h^{-2} \xi)$ ,  $L(\xi)$  is the Langevin function,  $\xi$  is the Langevin parameter,  $k' = qc^2C_p^{-1}$ ,  $k'' = N_d \mu_0 n m_*^2/k_0 T$ , qis the thermal expansion coefficient, Tdenotes absolute temperature, c is the speed of acoustic wave propagation,  $C_p$  is the specific heat capacity at constant pressure,  $N_d$  is the dynamic demagnetizing factor defined by the shape parameter P (the wavelength-to-sound beam diameter ratio), n is the ferroparticles concentration,  $m_*$  is the ferroparticle magnetic moment.

The given expression allows the estimation of only the limiting values of the physical parameters of the disperse phase nanoparticles and does not consider the particle size distribution and interpartial interactions.

We used widely known theory MMF2 [5] for the account of this moments. Expres-

sions from work [6] were also used for definition of parametres of allocation. The expression for EMF, directed in the measuring contour, gained taking into account the data [5,6] will register as:

$$\beta_{\xi e} = \frac{\int_{0}^{\infty} L(\xi_{e}) f(x) dx - k' \int_{0}^{\infty} D(\xi_{e}) f(x) dx}{1 + \frac{N_{d} \mu_{0}}{k_{0} T} n_{0}^{\infty} m_{*}^{2}(x) \xi_{e}^{-1} D(\xi_{e}) f(x) dx},$$

where  $\xi_e$  is the parameter of the effective field featured in [5].

In extreme cases this formula becomes: when  $H \rightarrow 0$ 

$$\beta_{He} = \frac{\left(1 - k'\right) \cdot \chi}{M_S \left(1 + N_d \chi\right)} H ,$$

where  $\chi$  is the FF initial magnetic susceptibility [5].

when  $H \to \infty$ 

$$\beta_{He} = \left(1 - \frac{(1+k')nk_0T}{\mu_0 M_S H}\right) \left(1 + \frac{(1+k')nk_0T}{3\mu_0 H^2}\right)$$

where  $M_s$  denotes MF saturation magnetization.

The given expressions with the use of the known relations published in [5,6] allow finding the FF particle size distribution from the data examination of the AME amplitude dependence from the exterior magnetic field value.

The experimental installation in detail featured in [4] has been created for examination of the FF disperse composition. Examinations were spent on samples of the «magnetite in the kerosene» type, stabilized by the olein acid, with various concentrations.

The FF particle size distribution curves, gained by the atomic force microscopy

(AFM), magnetogranulometry analysis (MFA) and the estimation on the AME basis, are presented on Fig. 1.



Fig. 1. The FF particle size distribution: AME (continuous line), MGA (dashed line), AFM (dash-dotted line)

The data was obtained for the FF sample with the density  $\rho$ =1360 kg/m<sup>3</sup>, the saturation magnetization  $M_s$  =57.3 kA/m and the initial magnetic susceptibility  $\chi$ =4.2. The difference of the mean particle diameter value  $\langle x \rangle_{AFM}$ =11.6 nm measured by AFM and the magnetic core diameter  $\langle x \rangle$ =, spotted by estimation on the AME basis is possible to explain by the presence of the stabilizing shell and the thin antimagnetic stratum on the magnetic particle surface

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# Cobalt Particles with Tuned Pattern Formation with and without External Fields

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Ferromagnetic single-domain particles are of interest to combine with soft matter matrices due to their strong interparticle ineraction and high magnetic moment.

However, highly magnetic nanoparticles such as iron or cobalt suffer from low stability against oxidation.

Various reports show that cobalt particles are readily accessible by hot injection methods<sup>1</sup>, resulting in highly monodispersed cobalt nanoparticles in the presence of a surfactant. We recently found that by using polystyrene with a carboxylic acid group as coordination surfactant after Pyun et al., <sup>2</sup> the obtained cores show an unexpected oxidation stability.

In this work we investigated the influence of reaction conditions on particle properties.



**Fig. 1:** Synthesis of cobalt nanoparticles decorated with polystyrene in a one step reaction.

More precisely, we studied the influence of tri-n-octylphosphineoxid (TOPO), and of polymer with different molecular weight on the properties of the cobalt particles. For analyzing the magnetic properties of the resulting magnetic fluids, vibrating sample magnetometry (VSM) has been used, and the magnetic stability of the cobalt particles under different storage conditions has been recorded over time. A core-shell architecture has been proposed by using light scattering and AC susceptometry experiments. The formation of organogels from the particles is subject to ongoing examinations.



**Fig. 2:** TEM image of cobalt based particles, a) prepared with PS-COOH without TOPO. b) Prepared in the presence of 6,5 mmol/L of TOPO.

### Acknowledgments

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# Magnetically induced phase condensation in an aqueous magnetic colloid

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### Introduction

In this paper we present the investigation of the magnetically induced "gas-liquid" condensation in a water based magnetic nano-colloid with magnetite nanoparticles of about 8nm mean diameter and 2.3% particle volume fraction, sterically stabilized with a double surfactant layer of myristic acid and dodecylbenzenesulphonic acid (MA+DBS) [1]. The results from DLS, light extinction and dichroism experiments are presented and discussed.

### **Results and discussions**

In Fig.1 is presented the temperature dependence of the scatterer's mean hydrodynamic diameter from DLS experiments. One can notice the sharp decrease of the clusters' diameter in the temperature range  $30^{\circ}$ C- $60^{\circ}$ C.



Fig.1 Temperature dependence of the primary particle clusters hydrodynamic diameter.

The comparison between the hydrodynamic and physical diameter of the magnetite particle indicates that the particles spontaneously agglomerate to form primary, rather soft, agglomerates.

From light extinction experiments it was found that the sample undergoes magnetically induced "gas-liquid" condensation with critical point: below the critical temperature  $t_c$  there's a temperature dependent critical field ( $H_c(t)$ ) below which no condensation occurs, and above which the amount and size (in the microns range) of the nano-particle aggregates increases with the field intensity. Above  $t_c$  no phase condensation was observed up to the highest available field intensity of 3500 Oe.



Fig.2 Temperature dependence of light extinction.

In Fig.2 is presented the temperature and magnetic field intensity dependence of the saturation extinction. Due to the light scattering on the condensed phase drops, elongated in the magnetic field direction with a size that can range from hundreds to thousands of nanometers, depending on temperature and magnetic field intensity, the larger the nanoparticle cluster size and number, the larger the extinction [2]. The sample presents a critical temperature of about 40°C above which no condensation occurs no matter how intense the external magnetic field.



Fig.3 The dependence of the critical field on the temperature.

In Fig.3 is plotted the temperature dependence of the critical field. One can notice the asymptotic increase of the critical field with increasing temperature. This behavior was theoretically predicted by Cebers [3]:

$$H_{c} = \frac{2 \cdot k_{B}}{\mu_{0} \cdot m} \cdot \frac{T_{c}^{2}}{T_{c} - T}$$
(1)

where  $k_B$  is Boltzmann constant,  $\mu_0$  is the magnetic permeability of vacuum, *m* is the magnetic moment of the magnetic nanoparticle and T<sub>c</sub> is the critical temperature. Using eq.(1) to fit the data from Fig.3, one obtains for the critical temperature a value of about 42°C and an estimate for the magnetic diameter of the particle primary clusters of D<sub>m</sub>=54nm.

From dichroism experiments in low magnetic fields at 30°C it was found that below the critical field the sample behaves like a typically stable ferrofluid. In Fig.4 is plotted the magnetic field dependence of normalized dichroism for the the (AM+DBS) water based colloid and for a sample with magnetite particles stabilized with oleic acid + dodecylbenzensulphonic acid (AO+DBS) double layer and dispersed in pentanol. One can notice that the normalized dichroism curves overlap, which is an indication that the rotation of the single prolate nano-particles is the main cause of the magnetically induced optical anisotropy [4].



Fig.4 Field dependence of the normalized dichroism.

Therefore, one may conclude that below the critical field the primary agglomerates are fairly isotropic and have little or no contribution to the magnetically induced optical anisotropy of the sample.

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# Computer models for the study of magnetic gels

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Magnetic gels, also called ferrogels, consist of magnetic nano-particles embedded in a cross-linked polymer network, and have potential applications ranging from medicine to engineering, e.g., drug release systems and artificial muscles. These applications rely on the combination of the mechanical properties of the polymer network – which can be tuned in a wide range during synthesis – with the ability to modify and control the sample with external magnetic fields due to the embedded magnetic nano-particles.

Today, many aspects of the synthesis of ferrogels are understood, and the properties of gels can be characterized and tailored. However, many aspects of the microstructure and its influence on macroscopic properties are still unknown. Computer simulations help in this area, because firstly, they allow complete control of the system under consideration, and secondly, because it is possible to study simplified model systems which focus on certain aspects of the material. In particular, an understanding of how a gel responds microscopically to an external magnetic field, and how mechanical properties change along with it, promise to help guide experimentalists toward building gels with desirable properties for applications.

In this contribution, we show two computer models, which stress two different aspects of the deformation mechanisms in a magnetic gel, namely, the re-arranging of magnetic particles and the deformation of the polymer matrix due to the alignment of magnetic particles. In this way, we identify two mechanisms which lead to a shrinking of the sample in an external magnetic field.



Figure 1: Upper part: Model of a magnetic gel in quasi two-dimensional geometry. Magnetic particles form the nodes of the network. The ends of flexible, non-magnetic chains are glued to a specific point on the magnetic particles' surface. Therefore, the network is deformed, once the magnetic particles align in an external magnetic field.

Lower part: boundary of a gel sample in external fields of different strengths. In the field, the gel contracts isotropically due to the deformation of the polymer matrix.

### Magnetic gels in magnetic fields: a fascinating alliance

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A magnetic gel is a chemically cross-linked polymer network swollen by a magnetic fluid. The finely divided colloidal particles couple the shape of the gel to the external magnetic field. The driving force of the deformation is the magnetic field gradient which can vary from point to point in the space resulting in non-homogeneous deformation. Direct observation of non-homogeneous deformation is reported and a theoretical interpretation based on the coupled magnetic and rubber elastic properties is provided [1,2].



Fig.1 Uniaxial elongation of a magnetic gel cylinder in non uniform magnetic field.

Synthesis of elastomers under uniform magnetic field can be used to prepare anisotropic samples. The imposed field orients the magnetic dipoles and if the particles are spaced closely enough, mutual particle interactions occur. Due to the attractive forces pearl chain structure develops. The resulting magnetic composite becomes anisotropic in terms of mechanical, swelling and magnetic properties. One can easily vary the direction of the particle chains by the direction of the applied magnetic field. The spatial distribution of the solid particles has a decisive effect on the stress-strain dependence as well as on the swelling kinetics [2,3]



Fig.2 Direction dependent elastic modulus (slope of the lines) of anisotropic magnetic polymer gel.

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# Inversion of Magnetic Interactions in Strongly Bidisperse Magnetic Fluids

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As compared with the traditional magnetorheological suspensions, compositions of the micronsized particles in ferrofluids are much more stable both with respect to sedimentation and irreversible aggregation. As a result, they are much more homogeneous, their properties are better reproducible and predictable. This is very important for precise technologies, which require high accuracy of the work media behavior.

In this work we present results of experimental and theoretical investigations of structural transformations and rheological properties of ferromagnetic microparticles dispersed in an initially isotropic ferrofluid (FF), which consists of nanometric magnets dispersed in a carried liquid. It is well known that upon magnetic field application, two particles of magnetizable materials which are approximately aligned in the direction of the field experience attractive magnetostatic forces. If these particles are free to move, e.g. they are dispersed in a liquid carrier, and the intensity of the applied magnetic field is large enough to overcome Brownian motion, these particles would attract in order to minimize their magnetostatic energy. The minimum is reached when the particles are in contact and aligned in the field direction. This is the basic phenomenon underlying the magnetorheological (MR) effect, which causes a quick and important change in the rheological properties of MR suspensions upon application of a magnetic field.

The classical theory of electrodynamics of continuum shows, that the force of

of magnetic attraction between two magnetizable particles immersed in a paramagnetic medium increases due to this medium. Thus one can expect increase of magnetorheological effect in suspension of magnetizable particles (MRS) in a ferrofluids as compared with the similar suspension in a nonmagnetic liquid.

Our experiments have demonstrated more rich set of internal transformations and rheological effects than the classical models predict. We used FF consisted of suspensions of oleate-covered magnetite nanoparticles dispersed in kerosene. In order to investigate the effect of FF particle size, the initial FF was centrifuged for 3 hours at 20000g. Particle size distribution of both the initial (non-centrifuged) FF and the centrifuged FF Results are shown in Fig.1



**Fig.1** Particle size distribution for the initial (non-centrifuged) FF (dashed line) and for the centrifuged FF (continuous line).

Microparticles of different materials (Fe, Ni, Ag-Ni) and average sizes (between 1 and  $10\mu m$ ) were used and a similar behavior was always found. In Fig.2a we see the

presence of a zone of more concentrated FF around the poles of the particles. In the stationary state (Fig.2b) this concentrated zone has the shape of a truncated ellipsoid and fills the gap between the particles maintaining them at a distance of about one diameter.



**Fig.2** Collision of Ni particles dispersed either in the centrifuged FF (a, b) or in the non-centrifuged FF (c, d), upon application of a magnetic field of 22kA/m in the indicated direction. (a, c) were taken during the approach, and (b, d) in the stationary state.

Microscopic observations were also performed in the non-centrifuged FF. In contrast to Fig.2a, in Fig.2c dense zones of FF are observed not only close to the microparticles but also far from them -it is possible to see dark lines along the field direction in all regions. In the stationary state, Fig.2d, the ellipsoidal nanoparticles cloud of between microparticles is considerably larger than that observed for the centrifuged FF, Fig.2b. Since the range of this magnetic barrier is strongly increased in the noncentrifuged FF, we conclude that the observed behavior is caused by the biggest nanoparticles inside the FF, which undergo a phase condensation, the especially zone in between microparticles, where the field is higher

Some results of measurements of rheological properties of the systems under study are presented in Fig.3.



Fig.3. Dynamical yield stress vs. applied magnetic field.

In contradiction with predictions, based on the classical theory of electrodynamics of continuum, and in contrast with the case of pure ferrofluids, in the compositions under study magnetorheological effect increases after centrifugation of the ferrofluid. The similar results have been obtained after measurements of storage and loss modulus of the composition.

We present a theoretical model of the equilibrium structures of the micronsized particles and the "stop-effect" at small distance between them. Analysis shows that the physical reason of this effect lies in the phase condensation of the biggest particles of the FF near poles of the micronsized particles. Appearance of the dense ferrofluid cloud in the gap between the micronsized particles leads to inversion of the force of magnetic interaction between them - at the relatively small distance instead of attraction it becomes repulsion. As a consequence, the presence of the biggest particles of ferrofluids, in the contrast with the case of a pure ferrofluid, decreases magnetorheological effect in these suspensions.

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# Maps



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