MATHEMATICAL MODELS OF SOFT MAGNETIC MATERIALS AND THEIR VERIFICATION

Dr. Andrejs Cēbers, Dr. Kaspars Ērglis, Dr. Andrejs Tatuļčenkovs, MSc. Oksana Petričenko

1.Magnetic microconvection.

A magnetic fluid subject to an external magnetic field induces an inhomogenous magnetic field dependent on the configuration of the external field. An example of such a situation is the case when a homogenous magnetic fluid in a Hele-Shaw cell borders a pure carrier fluid. A diffuse boundary layer in which the particle concentration decreases to zero forms between the two mixing fluids. If the layer is affected by an orthogonal magnetic field, an inhomogenous magnetic field is induced. Because of this the field acting on the magnetic colloid increases in the direction of the carrier fluid. Since the ponderomotive force acting on the magnetized environment is proportional to its magnetization and therefore the concentration of magnetic particles, a convective instability analogous to the observed in inhomogenously heated fluids is formed. The critical field value is determined by the magnetic Rayleigh number -- the ratio between the characteristic fluid motion timescale and the timescale of particle diffusion. This phenomenon was discovered at the Institute of Physics in 1983 [1*] and an adequate model was developed in 1997 [2*]. During that time, the thresholds of microconvection development at various field configurations had been explored [3*, 4*], however very little experimental research had been done. During this project, the microconvection velocity fields and their time evolution were measured for the first time utilizing the experimental and magnetic fluid synthesis capabilities of the Laboratory of Magnetic Soft Matter (LMSM). The dynamics of microconvective motion can be seen in Fig. 1.

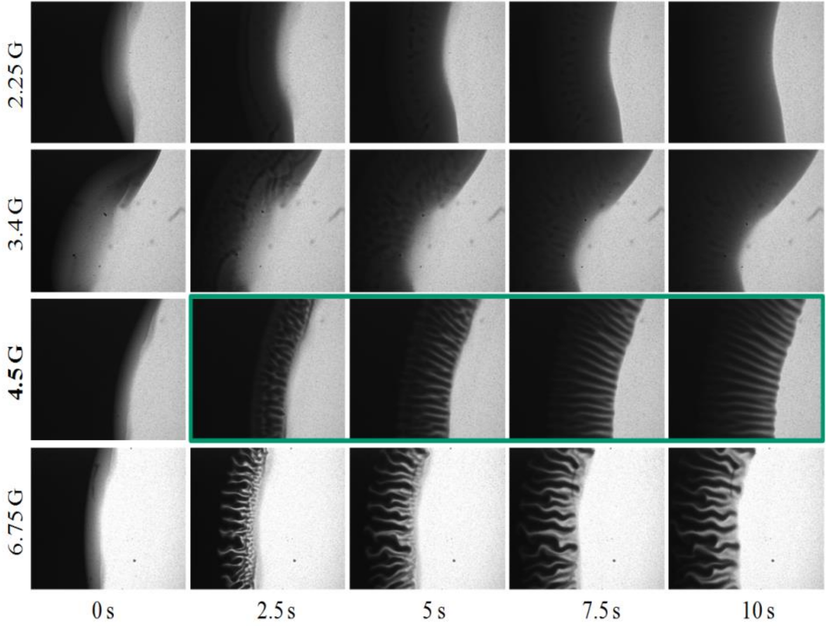

Fig.1

Fig.1 clearly shows that microconvective motion starts as the magnetic field exceeds a threshold value of roughly 3 G. The microconvection velocity fields were measured using the PIV device (Dantec) available at the LMSM. A representative measurement of a velocity field is shown in Fig. 2.

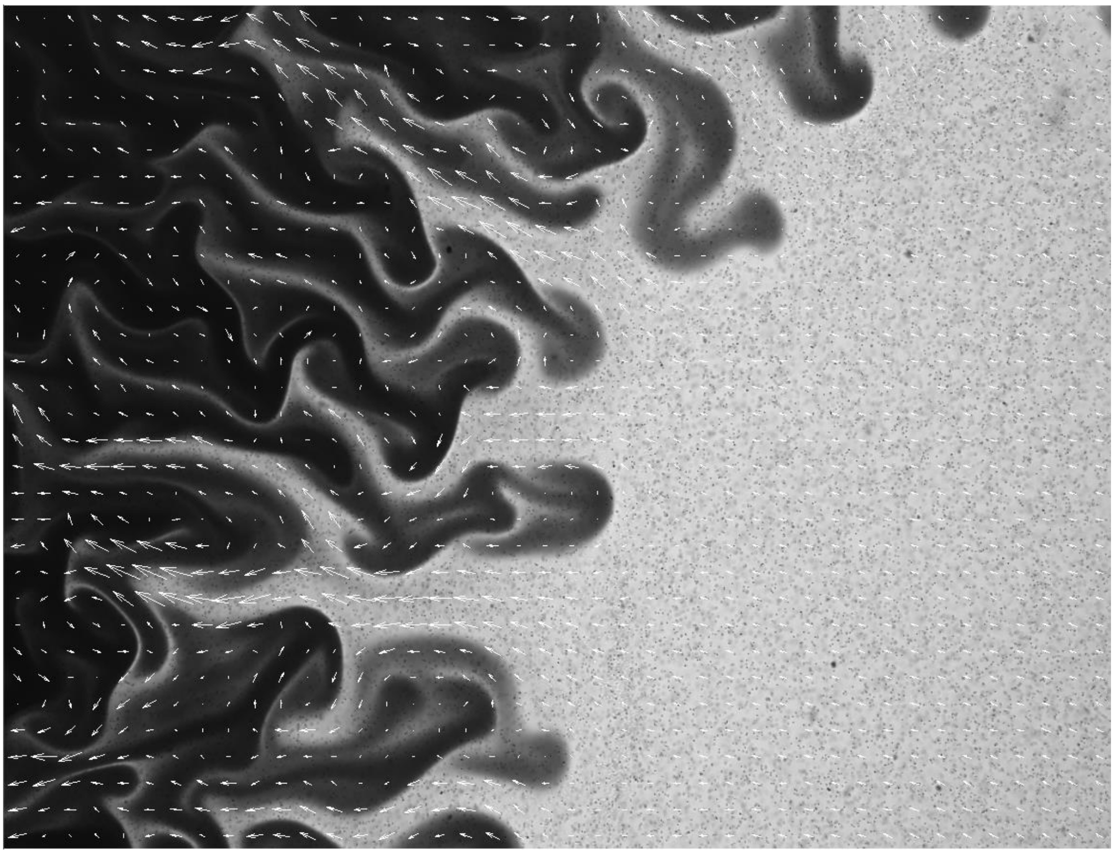

Fig.2

The measurement of velocity fields enabled the determination of vorticity evolution in time, which is shown in Fig. 3. It is clearly seen from Fig. 3 that microconvective motion reaches a maximum after which the concentrations of magnetic particles in evolving magnetic fingers are homogenized by particle diffusion. This illustrates the potential use of magnetic microconvection in the field of microfluidics since the development and subsequent subsiding of magnetic microconvection entails a significant mixing between the magnetic particles and the carrier fluid. The mixing of different fluids is a significant problem in microfluidics. Numerical simulations of this phenomenon show qualitative agreement with experiments, as shown in Fig. 4. Despite this, there are some discrepancies as well, for instance, using the Darcy approximation one cannot reproduce the characteristic hat-like shapes observed at the ends of the magnetic fingers. Further research in this direction will be continued using better approximations of fluid motion (the Brinkman model).

When conducting the measurements of velocity fields it was also found that gravitationally induced motion also plays a significant role despite the exceedingly small density differences between the magnetic colloid and the carrier fluid. This motion causes an almost horizontal boundary to be formed between magnetic and non-magnetic fluids under the influence on an orthogonal magnetic field. The dynamics of this boundary formation were numerically calculated using the

COMSOL environment and the results are shown in Fig. 5. As a consequence of this, a new problem was formed -- the instability of a horizontal boundary layer between mixing fluids in a vertical magnetic field. The instability of a horizontal magnetic fluid surface in a vertical field when the characteristic length scale is in the centimeter range is determined by the so called capillary length, this situation is well known in the literature. The new problem, however, is qualitatively different since there is no surface tension between the mixing fluids. In this case, the characteristic length scale is determined by particle diffusion and is on the order of tens of microns. The theoretical model developed for a sharp diffusion front can explain phenomena caused by gravitation during the microconvection process. It would be interesting to also explore the case of a smeared rather than sharp diffusion front. In addition to the aforementioned tasks, it would also be useful to explore magnetic microconvection in an environment where the fluid density difference is compensated for by an appropriate water solution. The glycerin water solution currently in use is not the most ideal because of different fluid viscosities.

The results of this project in conjunction with the theoretical description of this phenomenon and relevant numerical simulations have been accepted for publication in the Journal of Fluid Mechanics [1]. This may be the first case when a collaborative experimental as well as theoretical work fully carried out in Latvia has been accepted for publication in a scientific journal of this stature.

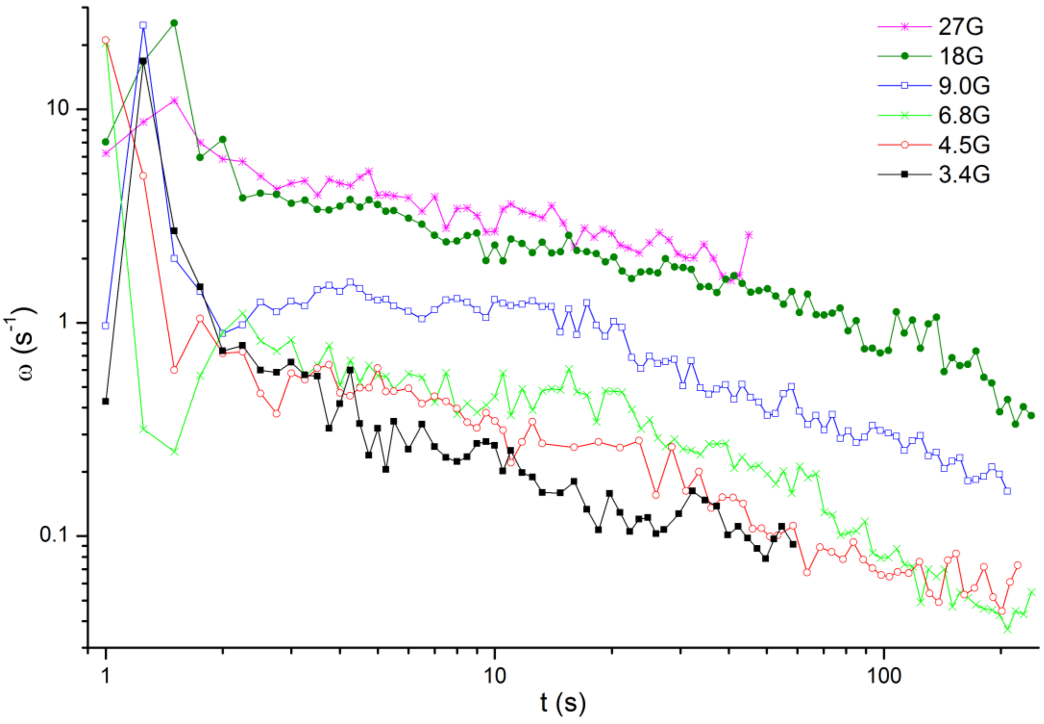

Fig.3

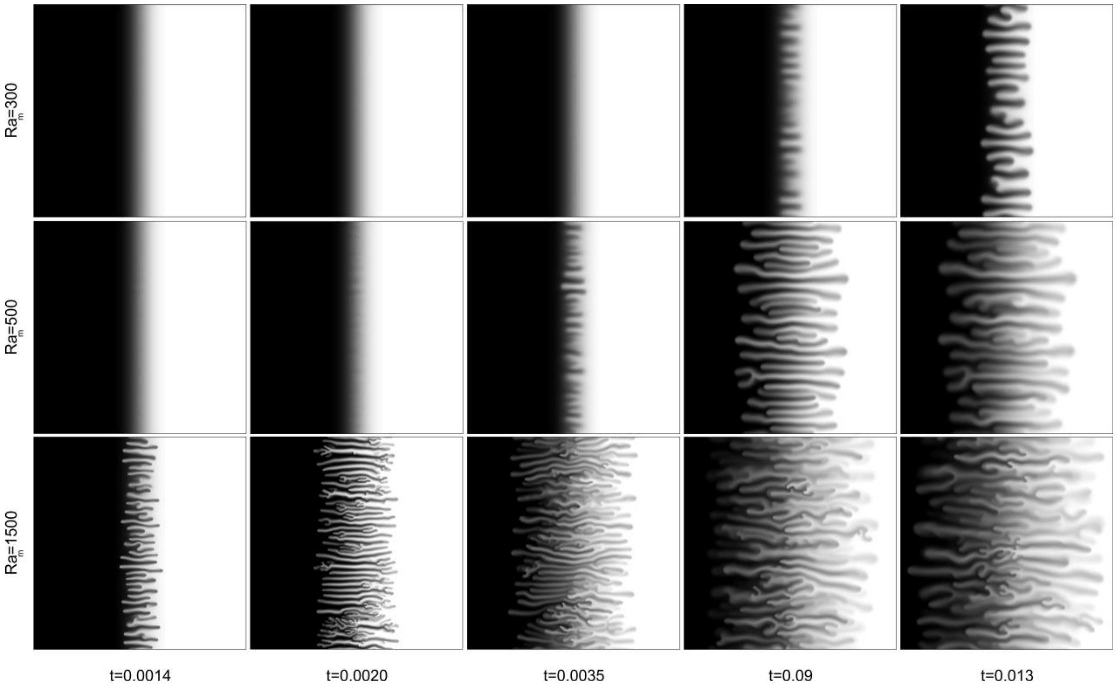

Fig.4

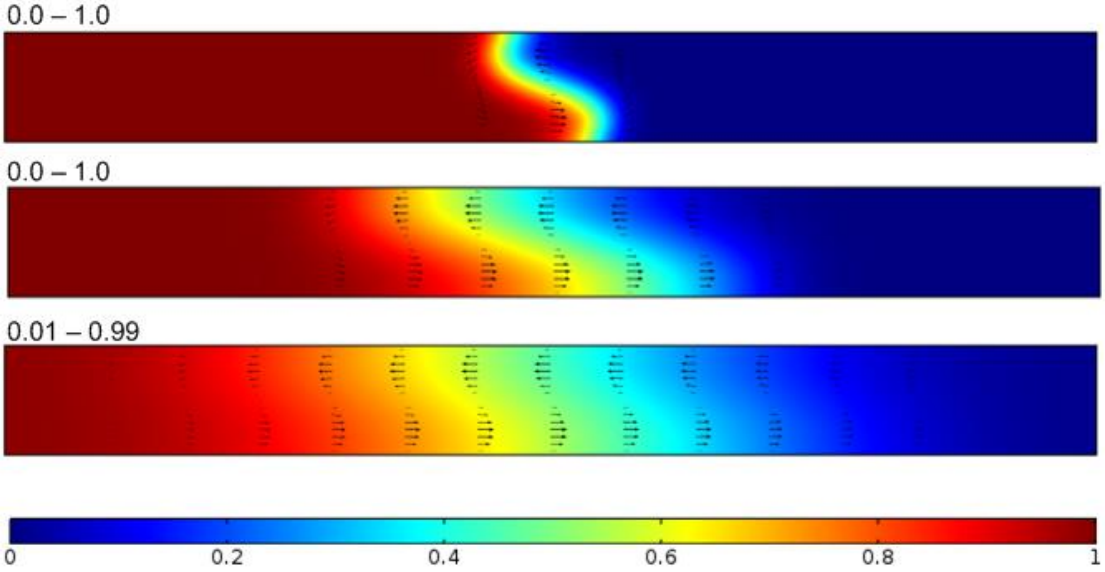

Fig.5

2.Synthesis and properties of magnetoliposomes.

Carriers for the targeted delivery of drugs and genetic information are an object of considerable interest in modern biomedicine. Liposomes with cation type lipid bilayers are potentially interesting for this purpose. During the period covered by this report the LMSM has developed a process for synthesizing large vesicles using cation type lipids created by the LIOS (Latvian Institute of Organic Synthesis). The characteristic sizes of such vesicles are on the order of tens of microns. This enables optical microscope measurements of lipid bilayer curvature elasticity which is known to play a significant role in endocytotic processes in cells. In order to determine this elasticity using the method of spontaneous engorgement, magnetic liposomes were synthesized and their curvature elasticity was determined by measuring liposome deformations in an external magnetic field. This phenomenon is illustrated in Fig. 6. By analysing the liposome eccentricity as a function of the external field, as shown in Fig. 7, it was found that the vesicles in question are characterised by a particularly small modulus of curvature elasticity. This may be explained by the polar heads of the lipid being particularly large.

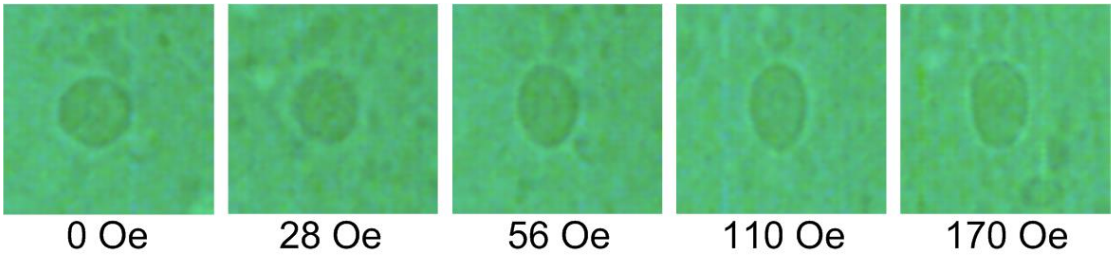

Fig.6

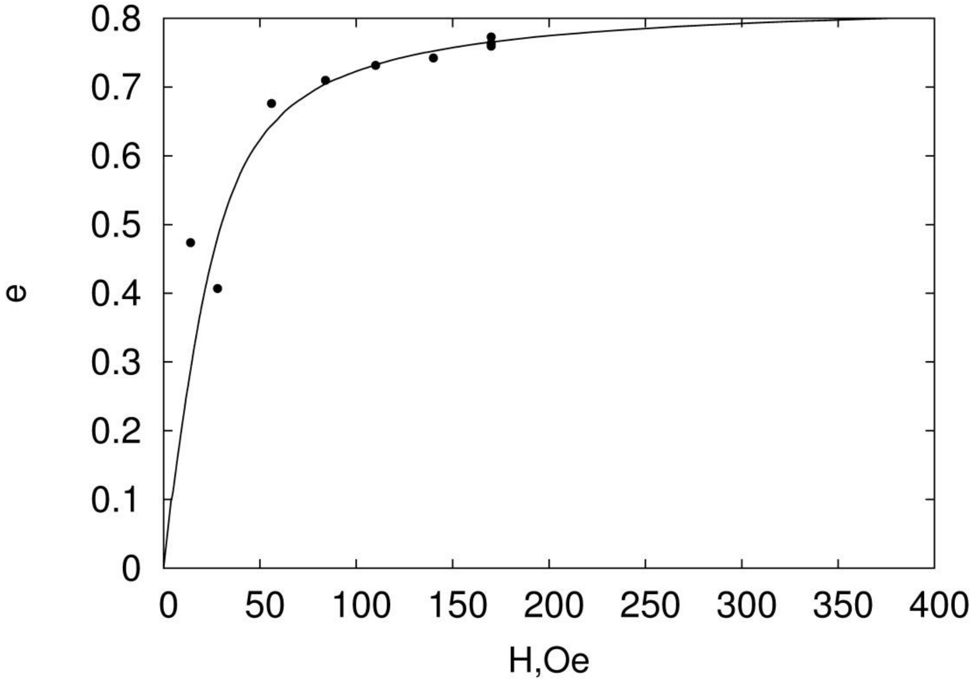

Fig.7

The magnetic properties of liposomes have been determined using magnetophoresis. It has been found that the concentration of particles within the liposomes is approximately 10 times smaller than that of the particles within the magnetic colloid used for the synthesis. This research has been submitted for publication [2] in the European Physical Journal E.

3.Microrheology.

One of the research directions that will continue to be developed in the Department of Theoretical Physics and the LMSM is microrheology, with a particular focus on magnetic microrheology. Methods in microrheology are typically divided into two groups. Active methods measure environmental response to external stimuli, whereas passive methods concern the properties of the thermal motion of a neutral particle in the environment of interest, which contains information about the properties of the environment. Active microrheological methods were used to measure the viscoelastic properties of a bacteriophage Pf1 gel for the first time, the results were published in [3]. An example of the Pf1 gel electronmicrography is shown in Fig. 8.

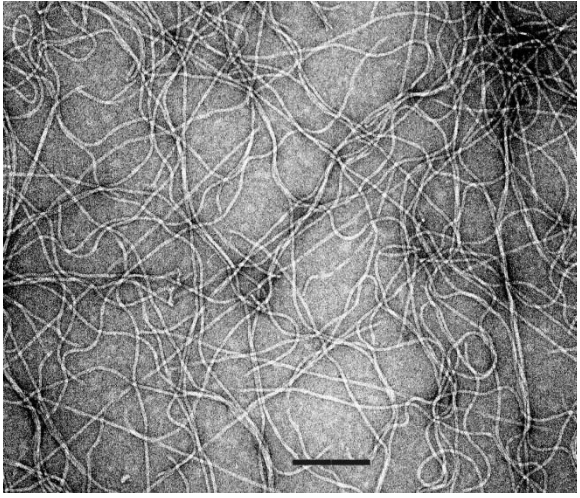

Fig.8

An important gel parameter to determine in order to further explore its properties using microrheology is the ratio between the size of the holes in the net and the size of the probing particle. In this work [3] a ten micron long superparamagnetic filament synthesized by the LMSM was used as the probe, this size considerably exceeds the size of the typical net hole. By determining the oscillation amplitude and phase of the filament subject to an external magnetic field, it was also possible to obtain the viscoelastic properties of the Pf1 gel as functions of frequency. The resulting values for the elastic modulus at different gel concentrations are shown in Fig. 9.

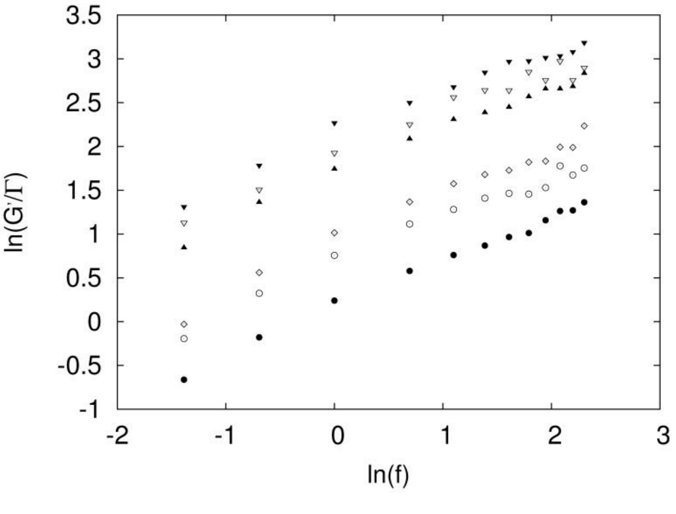

Fig.9

Research into the viscoelastic properties of Pf1 gels is also interesting due to the role these viruses play in the creation of biolayers of *Pseudomonos aeroginosa* bacteria. It is known that the accumulation of bacteria in biolayers, a serious pathology, is due to Pf1 viruses which develop by expressing the genes of the relevant bacteria. By developing the passive method described in [4] for the measurement of viscoelastic properties a new phenomenon was found [5]: a change in the viscoelastic properties of Pf1 gels under the influence of polyvalent cations. This can be seen in the Fig. 10, where the time dependence of the mean square displacement of a Brownian particle is altered by a change of concentration of the added salts. It is worth noting that, unlike classical Brownian motion, the mean square displacement is proportion to a power of time smaller than one. This is characteristic of subdiffusive processes and requires further theoretical work to be carried out in the future.

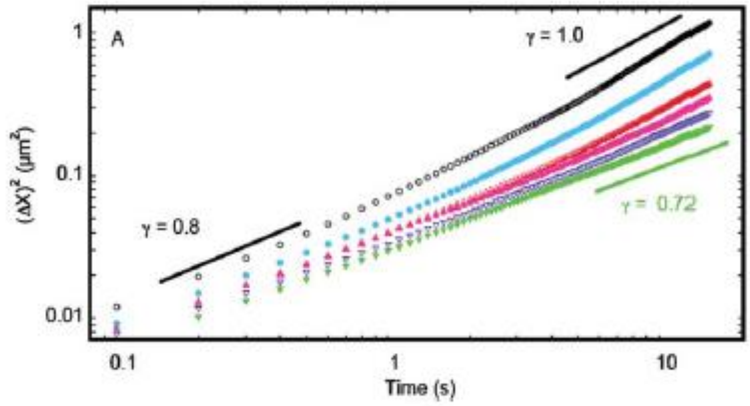

Fig.10

4.Mechanics of magnetic filaments.

The LMSM has successfully synthesized flexible ferromagnetic filaments by using commercially available micron-sized functionalized ferromagnetic particles and carried out several experiments [6] exploring their properties. An interesting phenomenon can be seen when a ferromagnetic filament situated in an external magnetic field reverses its orientation and forms a loop, as can be seen in Fig. 11.

By numerically modelling the filament dynamics and comparing the results with experiments the modulus of curvature elasticity has been evaluated. A three dimensional numerical algorithm was developed for carrying out the numerical computations. Results of numerical calculations at a magnetoelastic number chosen to closely mimic experimental observations can be seen in Fig. 12. The observed phenomenon of filament loop formation led to the idea of creating a biomimetic magnetic micro-engine described in the next section of this report.

A model describing the dynamics of a viscous string in a rotating magnetic field was developed [5*]. It was found that when the field frequency exceeds a critical value, a tangent angle shockwave spreads along the string.

The model described in [7]

Fig.11

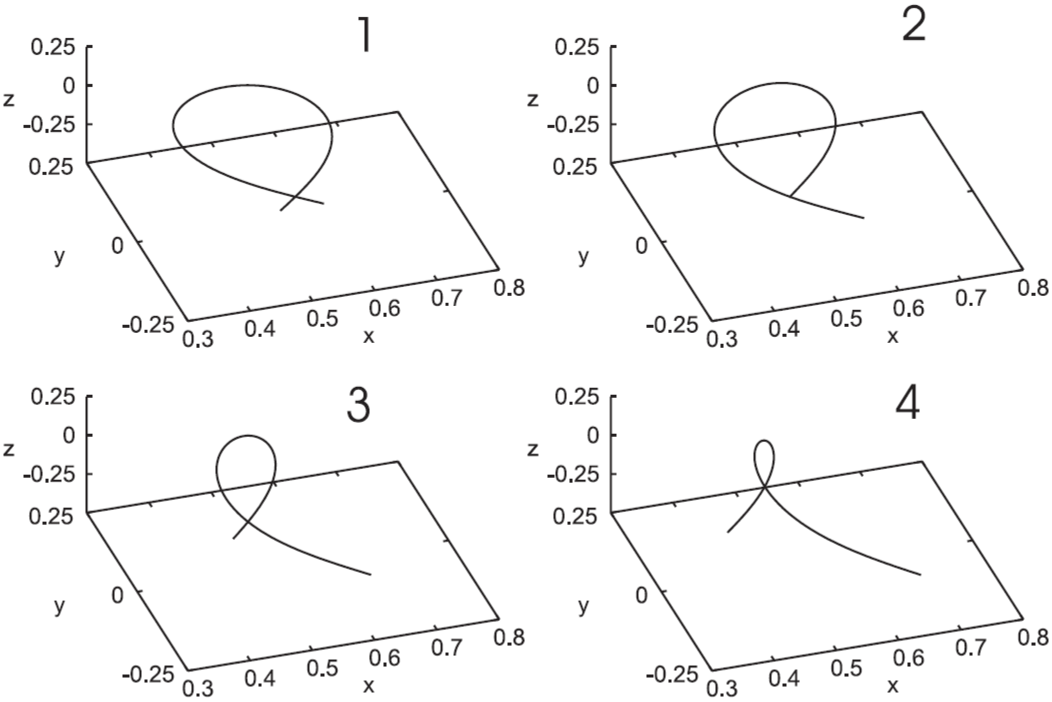

Fig.12

was used to explore the phenomenon described in [6*] -- the formation of a spiral at the ends of a chain of magnetic particles subject to a rotating field. Taking into account the dissipative torque induced on the filament if the particle magnetization has a finite relaxation time, results shown in Fig. 13 were obtained [7]. It is shown that spirals are formed at the ends of the filament which then propagate towards the center of the filament with a constant velocity. It is worth noting the such spiral formation can only be seen when repulsive forces between individual elements of the filament are taken into account. Otherwise, the formations induced at the ends of the filament propagate towards the center of the filament and annihilate. This can be seen in Fig. 14. In order to carry out numerical computations a regularization approach based on physical considerations was proposed [5*]. The question of regularization was further explored in [8] and it was found that a regularization function with a piecewise smooth derivative could be used.

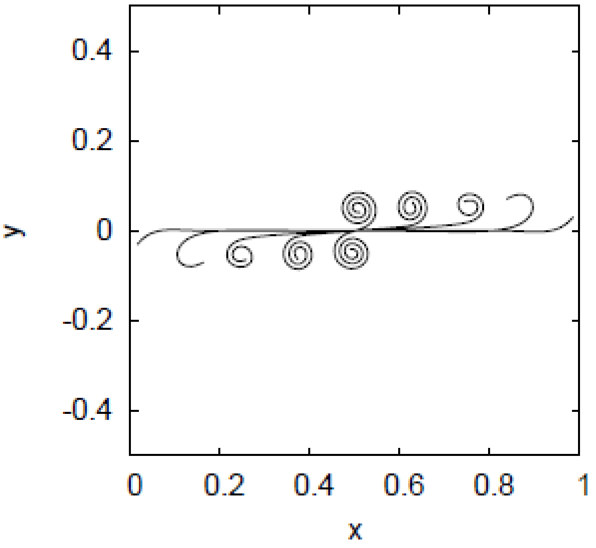

Fig.13

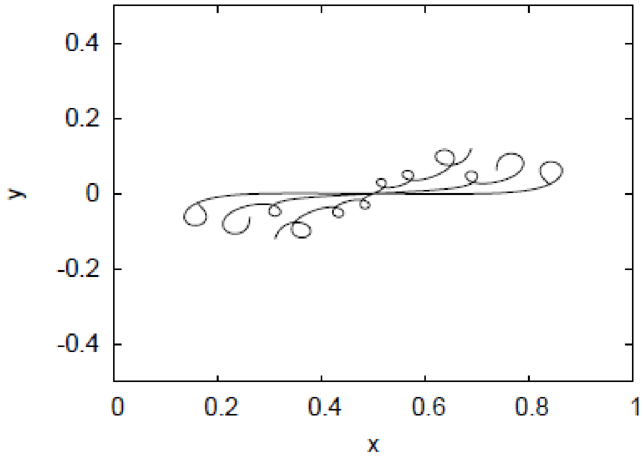

Fig.14

A new approach for describing elastic filaments was developed in [9]. Equations were formulated and solved in terms of the intrinsic variables of the filament -- the curvature and the tension on the filament. An interesting modifications to this class of problems was proposed involving tangential forces acting on the filament. This situation can be realised by, for instance, having an actin filament moving on a surface covered by molecular motors. In this case a circle-like filament configuration is formed, similar to experimental observations [7*]. This configuration is shown in Fig. 15. By comparing these configurations with the model solution, a previously undetermined constant was fixed; a result that corresponds will with experiments [7*]. The data used for determining this constant are shown in Fig. 15.

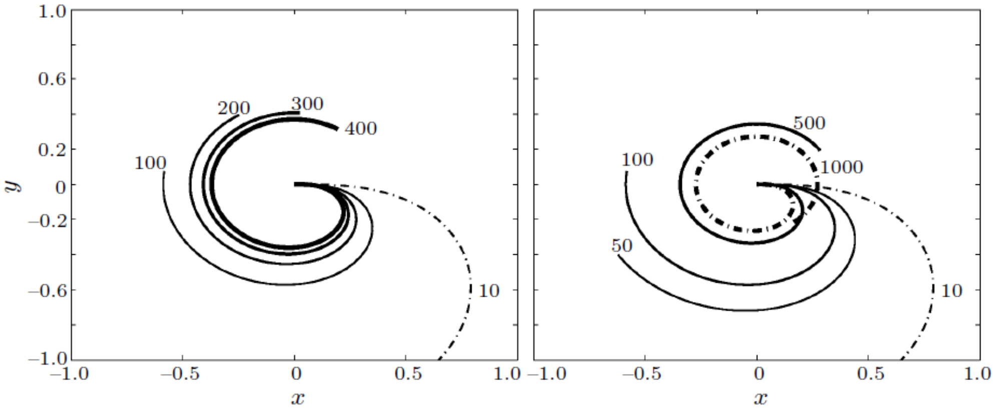

Fig.15

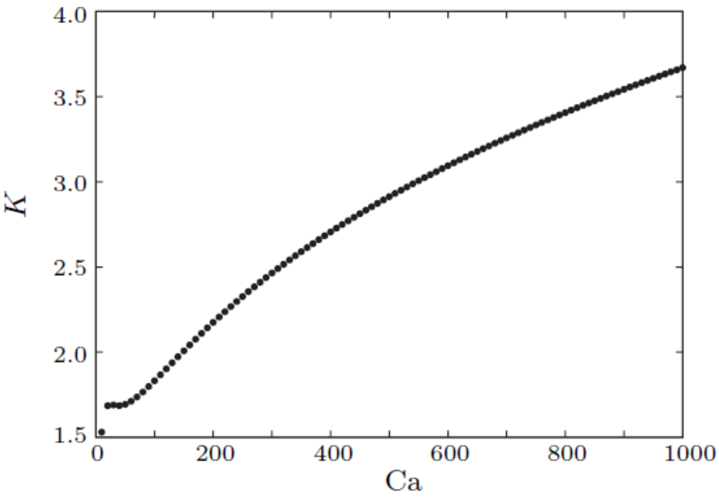

Fig.16

5.Magnetic micro-engines and micro-mixers.

Since the development of the model [8*] describing elastic magnetic filaments, work on the creation of magnetic field driven micro-engines and mixers has been carried out in the Department of Theoretical Physics. Some of the proposed micro-engine configurations have also been explored experimentally, although most of the results remain unpublished.

The proposition to create a biomimetic magnetic micro-engine based on the phenomena described in the previous section was considered in [10]. When a filament is subject to a time dependent magnetic field, self-propulsion is generated by the periodic formation and dissolution of loops. The time dependence of the center of mass coordinate of the filament (obtained numerically) is shown in Fig. 17. It can be seen that the forward motion of the center of mass of the filament is also accompanied by a smaller period of backward motion similar to the observed movement of *Chlamydomonas* flagellates in nature. It may be noted that after a certain period of time the filament finally orients perpendicularly to the magnetic field and the motion ceases. This process was characterized using the writhe number, the time dependence of which is shown in Fig. 18.

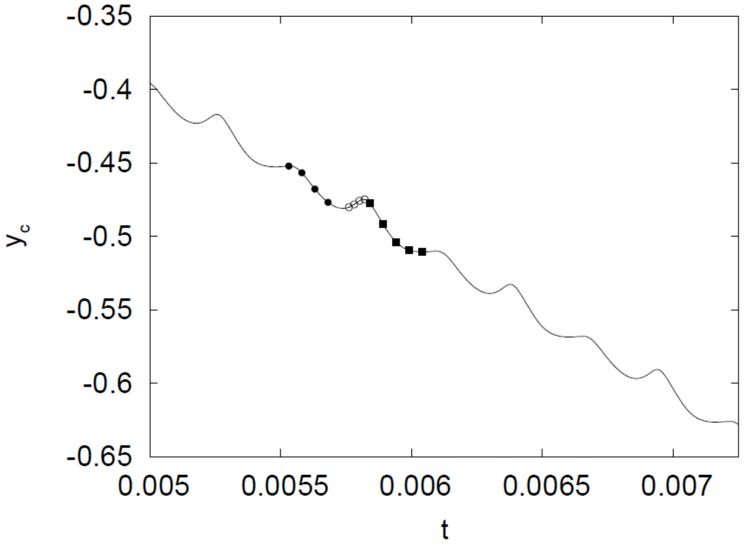

Fig.17

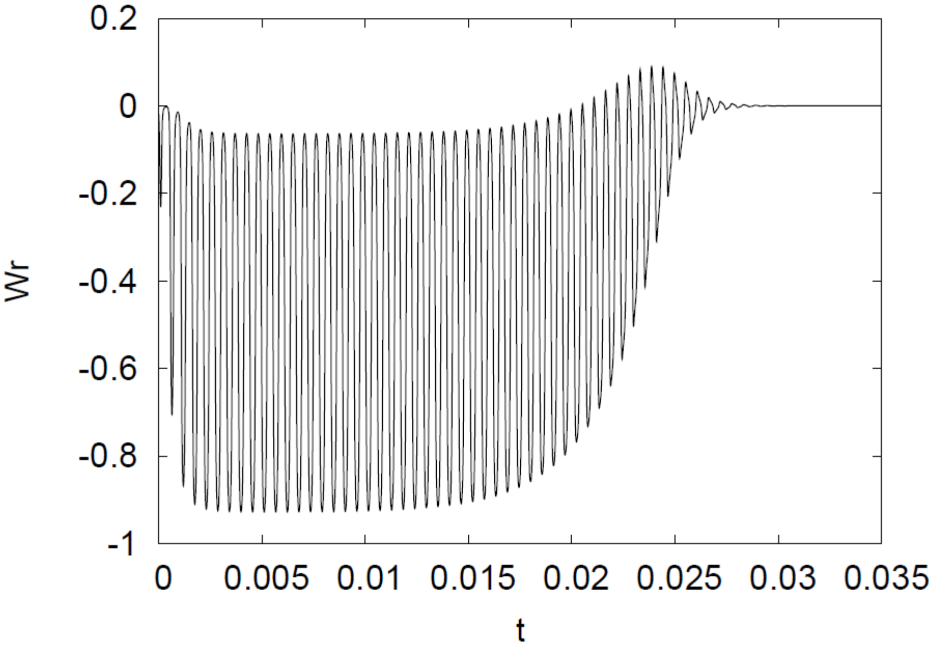

Fig.18

A new configuration - a magnetic dipole with an elastic filament attached to it - was proposed as a potential magnetic field driven micro-engine in [11]. By inducing dipole oscillations with a linear time dependent magnetic field the flexible tail deforms and causes the fluid-immersed object to move. It is interesting to note that the motion of this dipole also exhibits the back-and-forth motion characteristic of the aforementioned natural flagellates as well as the previous configuration. In contrast, only forward motion can be seen when using a rotating external magnetic field. The latter also appears to be faster. These results are shown in Fig. 19.

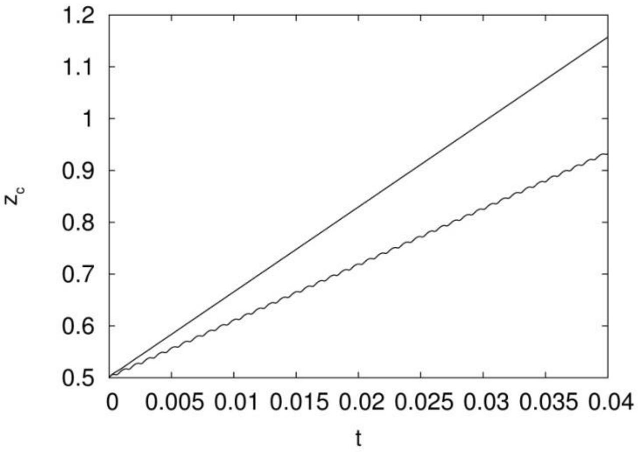

Fig.19

Projections of the micro-engine configurations on the plane of the rotating magnetic field during a single period are shown in Fig. 20. It is apparent that filament deformations cause its oriented motion.

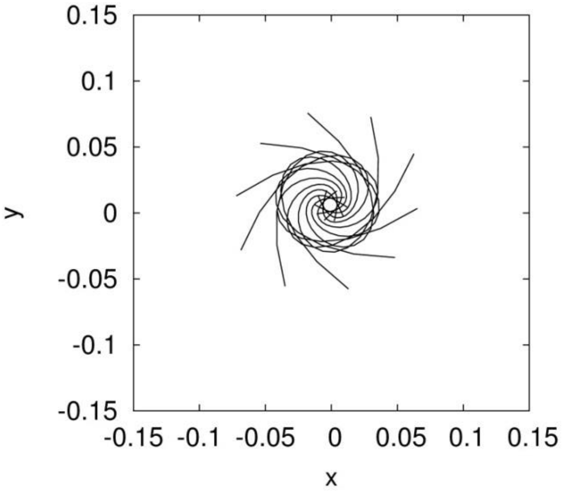

Fig.20

This engine exhibits several interesting properties related to the motion of the elastic filament. Since the magnetic torque of the swimmer's head exhibits phase lag with respect to the rotating magnetic field, a torque is induced which acts to compel the filament to move in the plane of the rotating field. As a result, the engine moves in a rotating trajectory similarly to the behaviour of magnetotactic bacteria in a rotating magnetic field [9*]. This trajectory is shown in Fig. 21.

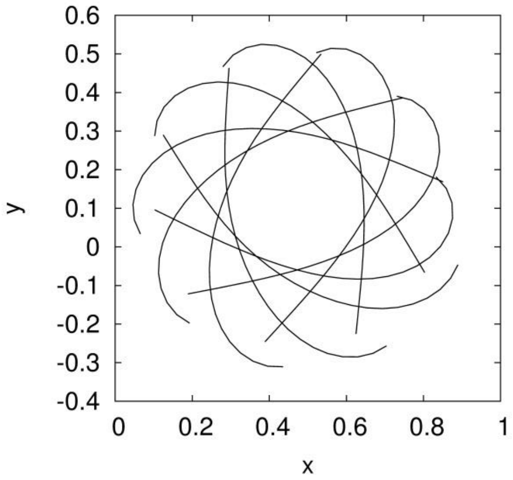

Fig.21

The trajectory shown in Fig. 21 is unstable and, if perturbed in a direction orthogonal to the plane of the rotating magnetic field, orients (in a time averaged sense) perpendicularly to the field and the self-propulsion shown in Fig. 19 may take place.

6.Synchronization.

Synchronizaton phenomena play a vitally important role in both biology and technology. Experiments have shown that synchronization phenomena also have a crucial role in various dipolar systems in time dependent magnetic fields. For instance, a system of two dipoles where the radius vector between the dipoles is oriented at a particular angle with the direction of the time dependent magnetic field was considered in [12]. It was found that the dipole motion exhibits synchronous motion in weak magnetic fields which is independent of the initial configuration. This phenomenon can be understood by considering the characteristic time scales of the system. In situations of interest the characteristic timescale determined by the dipole interaction is orders of magnitude larger than the period of the magnetic field. This implies that a qualitative understanding of the system can be attained by averaging the equations of motion over the field period and analyzing the so called slow variable dynamics. The isoclines of the slow variable differential equations are shown in Fig. 23 (for a supercritical field value).

22 (for a subcritical field value) and Fig.

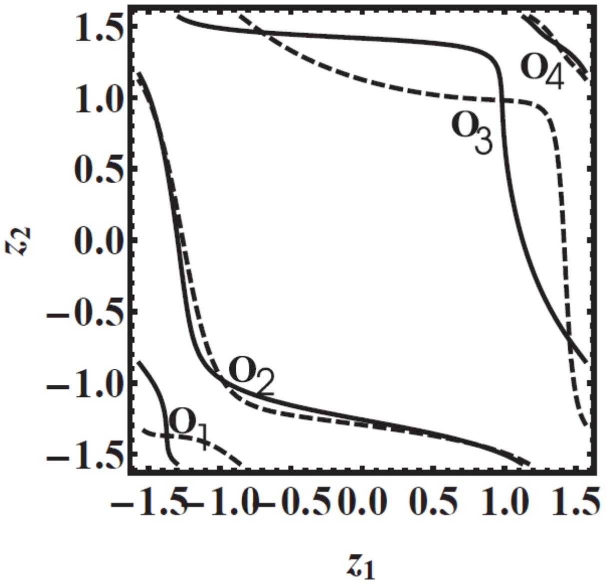

Fig.22

The stationary points of the system where the isoclines intersect are shown in Fig. 22. Stable stationary points correspond to O2 and O4. These states correspond to synchronous dipole oscillations. The stationary points are reached irrespective of the initial orientations. If the magnetic field strength is increased the stable stationary points disappear and there are in fact no stationary points for the system. This corresponds to synchronous dipole rotation in the same direction. During this rotation the dipole states reach the bottlenecks shown in Fig. 23 and the oscillation amplitude for both dipoles is almost constant. After a very gradual increase in the amplitude, the dipoles leave the bottleneck and quickly jump to the next one. This causes a non-zero dipole rotation velocity. This phenomenon can also be seen in numerical calculations in Fig. 24.

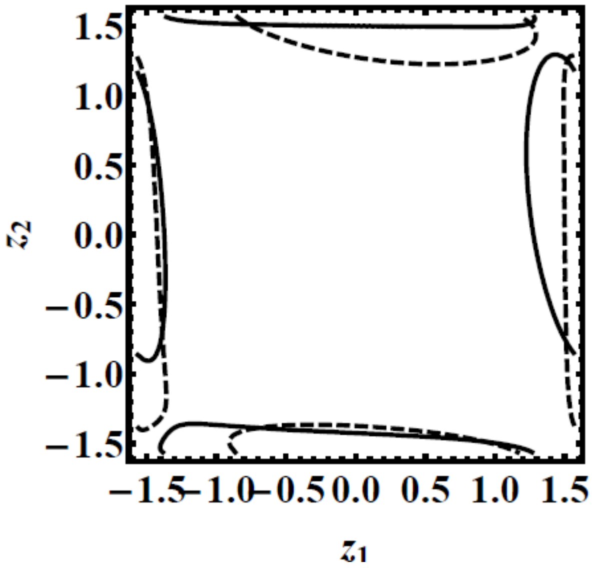

Fig.23

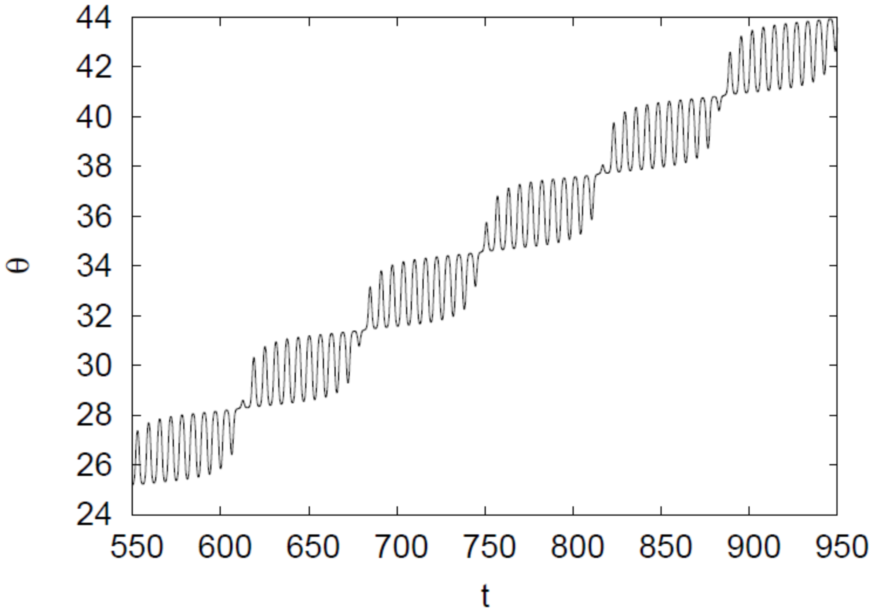

Fig.24

It is worth noting that the synchronization phenomena have a more complex character at intermediate values of the magnetic field strength. In the general case of dipole rotation, where the dipole rotation is characterized by two angles, the dipole equations of motion show a synchronization phenomenon illustrated in Fig. 25 and Fig. 26. These show that the dipoles synchronize by orienting in directions opposite to the plane of the magnetic field and the dipole radius vector. The trajectory of motion for this case is shown in Fig. 27.

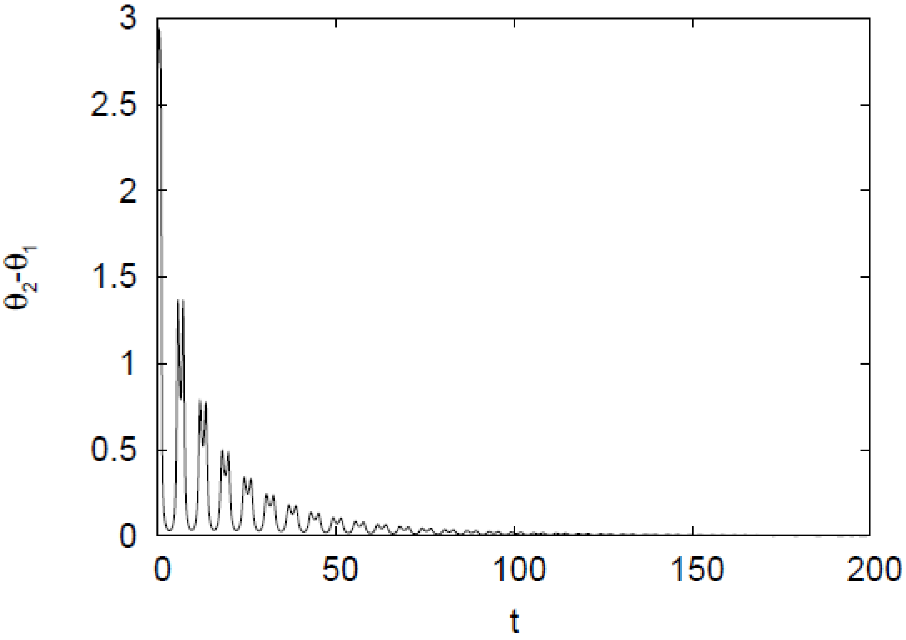

Fig.25

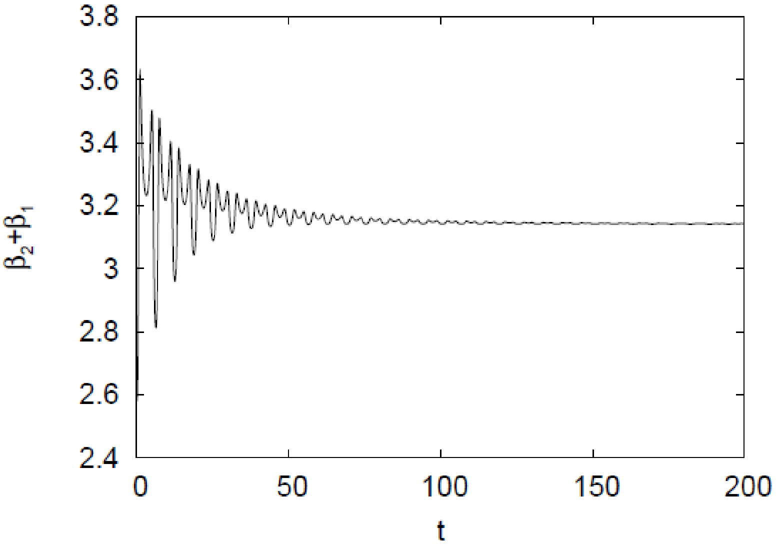

Fig.26

A host of interesting phenomena can be seen when considering the case of a large amount of dipoles. Fig. 28 shows the time dependence of the vectorial order parameter for 7 dipoles. The absolute value of the order parameter being equal to one means that the dipoles oscillate in phase and are oriented in the same direction. After a number of regular time intervals the synchronization is broken (as can be seen in Fig. 28) and the dipoles orient in different directions which leads to the order parameter taking a value close to zero. At the present time this phenomenon has only been reported in conferences. If further research leads to an understanding of the mechanism behind the characteristic synchronization time it could lead to an interesting publication as well.

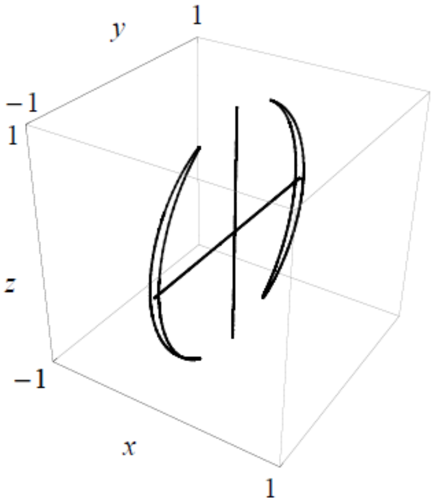

Fig.27

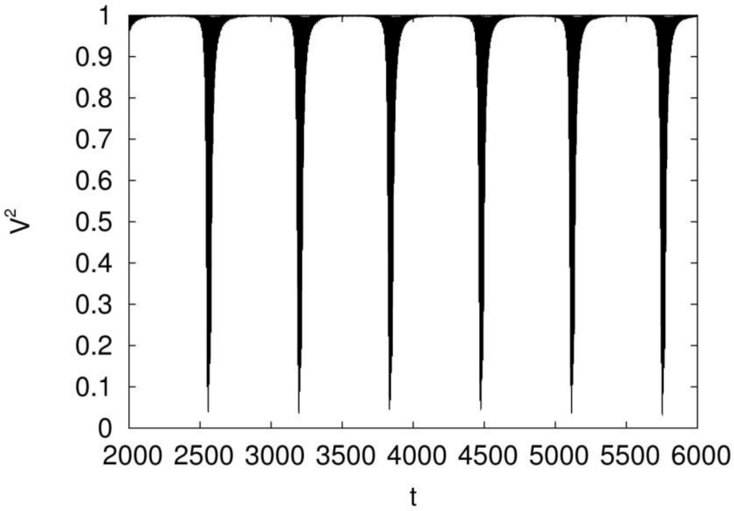

Fig.28

7.Magnetotactic bacteria.

Magnetotactic bacteria moving in a rotating magnetic field have been found [9*] to switch their directions of motion. Since this switching follows a probabilistic law, it leads to the center of curvature of the bacteria trajectory exhibiting a peculiar random walk pattern. This problem has been explored theoretically [13] by numerical modelling. Numerical calculations of this random bacterial motion were carried out in the case where the switching times follow a Poisson-type law and the characteristic diffusion coefficients of the resulting random walk were found. The numerically observed random trajectory is shown in Fig. 29. The random jumps between the circles of the bacteria trajectory can be clearly seen. By analyzing these random trajectories it was possible to ascertain the effective diffusion coefficients in the plane as functions of the rotating field frequency. A diffusion coefficient in one of the directions is shown in Fig. 30 as a function of the field frequency.

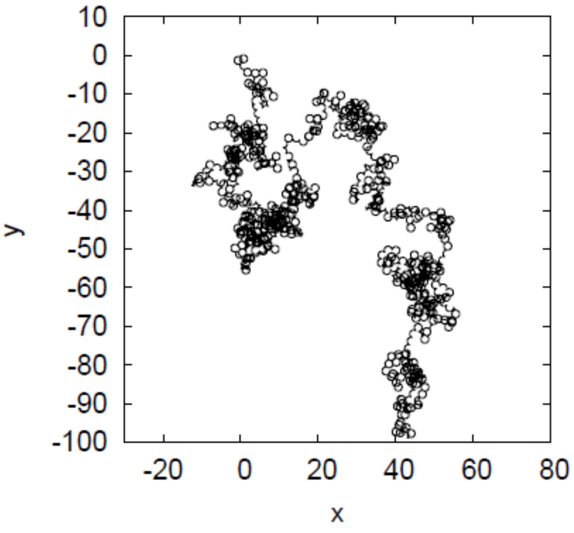

Fig.29

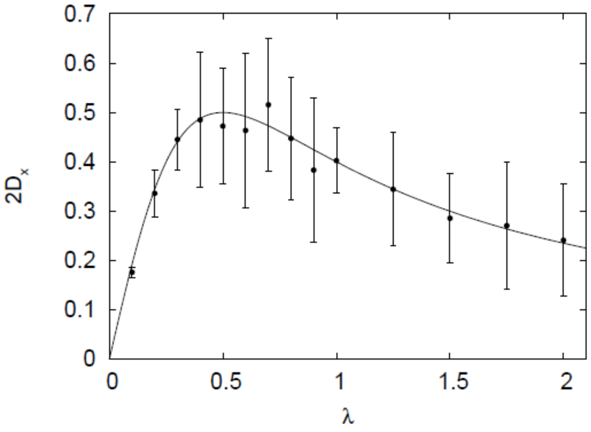

Fig.30

References.

1. **M.M.Mayorov, A.Cebers**. Magnetic microconvection on the diffusion front of ferroparticles//Magnitnaya Gidrod. (in Russ.). – 1983, v.19, N4 – P.36–40; Magnetohydrodynamics (Engl. translation) – 1983, v.19, N4 – P.376–380
2. **A.Cebers**. Stability of diffusion fronts of magnetic particles in porous media (Hele Shaw cell) under the action of external magnetic field//Magnitnaya Gidrodinamika – 1997, v.33, N1 – P.67–74; Magnetohydrodynamics (Engl. translation) – 1997, v.33, N1 – P.48–55.
3. **A.Cebers, M.Igonin**. Convective instability of magnetic colloid and forced Rayleigh scattering experiments. Magnetohydrodynamics – 2002, v.38 – P.265 – 270.
4. **A.Cebers, M.Igonin**. Convective instability of magnetic colloid and forced Rayleigh scattering experiments. Magnetohydrodynamics – 2002, v.38 – P.265 – 270.
5. **A.Cebers**. Dynamics of an elongated magnetic droplet in a rotating field. Phys. Rev. E – 2002, v.66 – P.061402-1-6.
6. **N.Casic, S.Schreiber, P.Tierno, W.Zimmermann, T.M.Fisher**, Europhysics Letters = 2010, v.90- P.58001
7. **L.Bourdieu, T.Duke, M.B.Elowitz, D.A.Winkelmann, S.Leibler, A.Libchaber**. Phys.Rev.Lett., 1995- v.75-P.176-179.
8. **A.Cebers**. Dynamics of a chain of magnetic particles connected with elastic linkers. Journal of Physics: Condensed Matter – 2003, v.15 – P.S1335–S1344
9. **A.Cebers,K.Erglis,Qi Wen,V.Ose,A.Zeltins,A.Sharipo,P.A.Janmey**. Dynamics of magnetotactic bacteria in a rotating field. Biophysical Journal. – 2007,v.93-1402-1412.

Publications

1. **K.Ērglis,A.Tatulcenkov,G.Kitenbergs,O.Petrichenko, F.G.Ergin,B.B.Watz, A.Cēbers.** Magnetic field driven microconvection in the Hele-Shaw cell. J.Fluid Mech., (accepted)
2. **O.Petrichenko, .K.Ērglis, A.Plotniece, K.Pajuste, G.Bealle, Ch.Menager, E.Dubois, R.Perzynski, and A.Cēbers.** Properties of bilayers formed by cationic pyridine amphiphile. European Physical Journal E (submitted)

3. **K.Ērglis, V.Ose, A.Zeltins, A.Cēbers**. Viscoelasticity of the bacteriophague Pf1 network measured by magnetic microrheology. Magnetohydrodynamics – 2010, v.46, P.23-29.
4. **G.Kitenbergs,K.Dzilna,K.Ērglis,A.Cēbers**. Thermal fluctuation effects in magnetophoresis of superrparamagnetic microbeads. AIP Conference Proc.- 2010, v.1311;141-145.
5. **E.M.Huisman, Qi Wen Yu-Hsiu Wang, K.Cruz, G.Kitenbergs, K.Ērglis, A.Zeltins, A.Cēbers, and P.A.Janmey**. Gelation of semiflexible polyelectrolytes by multivalent counterions. Soft Matter – 2011, v.7,7257-7261..
6. **K.Ērglis, R.Livanovičs, A.Cēbers**. Three dimensional instability of the flexible ferromagnetic loop. Magnetohydrodynamics -2010, v.46, 245-256
7. **A.Cēbers, H.Kalis**. Dynamics of superparamagnetic filaments with finite magnetic relaxation time. European Physical Journal E- 2011,v.34:30.
8. **A.Cebers,H.Kalis**. Mathematical modelling of an elongated magnetic droplet in a rotating magnetic field. Mathematical Modelling and Analysis – 2012, v.17, P.47-57.
9. **A.Cebers, and H.Kalis**. Intrinsic curve dynamics of magnetic filaments. Magnetohydrodynamics -2011,v.47, 223-236.
10. **K.Ērglis, R.Livanovičs, A.Cēbers**. Three dimensional dynamics of ferromagnetic swimmer. JMMM – 2011, v.323, 1278-1282
11. **R.Livanovics, and A.Cēbers**. Magnetic dipole with a flexible tail as a self-propelling microdevice. Phys.Rev.E-2012, v.85,041502
12. **M.Belovs,A.Cēbers**. Synchronization of magnetic dipole rotation in an ac magnetic field. J.Phys.A:Math.Theor., - 2011, v.44, 295101
13. **A.Cēbers**. Diffusion of magnetotactic bacterium in rotating magnetic field. JMMM – 2011, v.323, 279-282.

Conference theses

1. **K.Ērglis, R.Livanovičs,A.Cēbers.** Three dimensional dynamics of ferromagnetic swimmer. 8th International Conference on the Scientific and Clinical Applications of Magnetic Carriers, P.95,2010.
2. **G.Kitenbergs, K.Dzilna, K.Ērglis, A.Cēbers**. Thermal fluctuation effects in magnetophoresis of superparamagnetic microbeads. 8th International Conference on the Scientific and Clinical Applications of Magnetic Carriers, P.120,2010.
3. **K.Ērglis,R.Livanovičs,A.Cēbers.** Three dimensional dynamics of ferromagnetic swimmer. 12th International Conference on Magnetic Fluids. P.55-56, 2010.
4. **B.Frka-Petesic, K.Erglis, J.F.Berret, A.Cēbers, V.Dupuis, J.Fresnais, R.Perzynski, O.Sandre**. Dynamics of organic/inorganic magnetic microrods under rotating field. 12th International Conference on Magnetic Fluids. P.264-265,2010.
5. **A.Cēbers, R.Livanovičs**. Flexible ferromagnetic filaments as artificial cilia. Proceedings of the 12th international Conference electr-rheological fluids and Magneto-rheological suspensions. Philadelphia, USA,P.272-279,2010.
6. **M.Belovs,A.Cēbers**. Synchronization phenomena in dipolar systems. Euromech Colloqium 526:Patterns in soft matter,P.13-14, 2011.
7. **A.Cebers,M.Belovs**. Synchronization phenomena in systems with magnetodipolar interactions. APS March Meeting, Dallas,2011. Abstract X9.4.
8. **M.Belovs, A.Cēbers, and H.Kalis**. Dynamics of flexible magnetic microrods. 16th International Conference on Mathematical Modelling and Analysis, Sigulda,Latvia, P.13, 2011..
9. **A.Cēbers, and H.Kalis**. Intrinsic curve dynamics of magnetic filaments. 16th International Conference on Mathematical Modelling and Analysis, Sigulda,Latvia, P.25, 2011.
10. **A.Cēbers, M.Belovs**. Dipolar colloids in ac magnetic field. Moscow International Symposium on Magnetism. P.866-867,2011.

11. **A.Cebers, M.Belovs**. Synchronization phenomena in dipolar systems. 8th international pamir Conference on Fundamental and applied MHD., Borgo, France e, P.939-943, 2011.
12. **O.Petrichenko, G.Bealle, C.Menager, A.Cebers, R.Perzynsky, A.Plotniece, V.Ose**. Synthesis of magnetoliposomes and studies of their properties. Synchronization phenomena in dipolar systems. 8th international pamir Conference on Fundamental and applied MHD., Borgo, France,P.1015-1019, 2011.
13. **K.Erglis, O.Petrichenko, A.Cēbers**. investigation of magnetic microconvection with PIV method. 8th international pamir Conference on Fundamental and applied MHD., Borgo, France, P.1021-1025, 2011.
14. **A.Cēbers**. Dipolar systems in AC magnetic fields. XXXII Dynamic Days Europe, Gthenburg,Sweden, 2012, P.81
15. **R.Livanovičs, A.Cēbers**. Magnetic field driven mcrodevices. 23rd International Congress of Theoretical and Applied Mechanics, 2012, Beijing,China,P.41.