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MAGNETOVISCOUS AND VISCOELASTIC EFFECTS IN FERROFLUIDS

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Suspensions of magnetic nanoparticles, commonly called ferrofluids, exhibit a broad variety of viscous effects induced by the action of magnetic fields. The related phenomena are in focus of research since about 30 years and have recently reached new dynamics due to the discovery of viscoelastic effects induced by magnetic fields. Here, effects due to the influence of magnetic fields on the rotation of single magnetic nanoparticles as well as cooperative phenomena and their importance for viscous effects in ferrofluids will be reviewed.

1. Introduction

Before discussing the influence of magnetic fields on the viscosity of magnetic fluids some of their basic properties, important for magnetoviscous phenomena in ferrofluids, should be shortly summarized. The interested reader is referred to the books of Rosensweig[1] and Blums[2] as well as to various review articles[3–6] for extended information on ferrofluids.

The system in focus of this article is a suspension of magnetic nanoparticles in an appropriate carrier liquid. The magnetic nanoparticles are commonly made of magnetite Fe_3O_4 but experiments have also been made using e.g mixed ferrites[7] or cobalt[8] particles. Furtheron magnetite particles will generally be assumed in the discussion, since these are usual for the most often used ferrofluids. In special cases, where other magnetic material is used, this will be explicitly mentioned. The magnetic particles have a mean diameter of about 10 nm and can thus be treated in the following as magnetic single domain particles[9] which are thermally agitated in the carrier liquid.

The volume concentration of the magnetic material reaches up to 15 vol.% and is usually about 7 vol.% in commercially available concentrated ferrofluids. Due to the small size of the particles, the suspensions are stable against sedimentation in the gravitational field as well as in strong magnetic field gradients. Furthermore agglomeration of the particles due to magnetic dipole–dipole interaction is negligible for this particle size. Only van der Waals interaction between the particles would

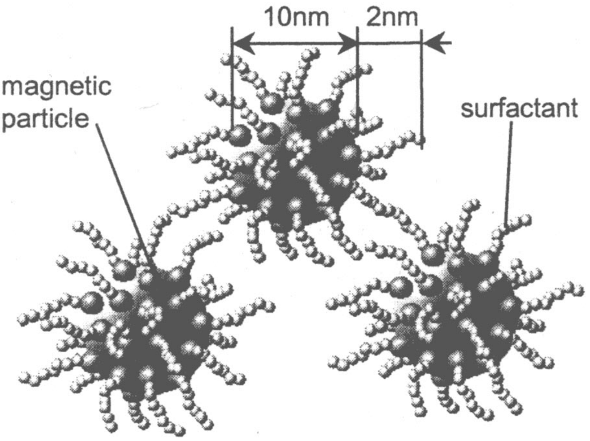

Fig. 1. Schematic sketch of the magnetic particles with surfactant. For reasons of clearness, particles and surfactant are not drawn in scale.

affect the stability of the suspensions. Therefore they have to be covered with a surfactant from long chained molecules (Fig. 1). The steric repulsion of these molecules prevents agglomeration and thus stable suspensions can be produced, showing long term stability over decades.

The special feature of these nanoparticle suspensions is the combination of normal liquid behavior with superparamagnetic properties. Assuming the particles as non-interacting, thermally agitated magnetic dipoles, the magnetization of the suspension as function of an applied field H can be described using the well known Langevin equation[10]

$$\begin{aligned} M &= \phi M_0 \left(\operatorname{ctgh} \alpha - \frac{1}{\alpha} \right) \\ \alpha &= \frac{\mu_0 m H}{kT} \end{aligned} \tag{1}$$

where ϕ denotes the volume fraction of the magnetic component, M_0 its spontaneous magnetization, m the magnetic moment of a single particle, k Boltzmann's constant and T the absolute temperature.

In contrast to paramagnetic salt solutions, the initial suszeptibility of a ferrofluid is about four orders of magnitude higher and reaches thus values of about 1. This bases on the fact that the particles, which have to be treated as the magnetic units affected by the field, contain about 10^4 Bohr magnetons.

Since the magnetic force acting on the fluid in a magnetic field gradient is proportional to the magnetization of the fluid, small fields in the order of 50 mT can produce forces comparable to gravity as shown in Fig. 2.

This controllable magnetic force acting on the fluids gave rise to the development of numerous technical applications[11] partly gaining high commercial importance and entering everydays live.

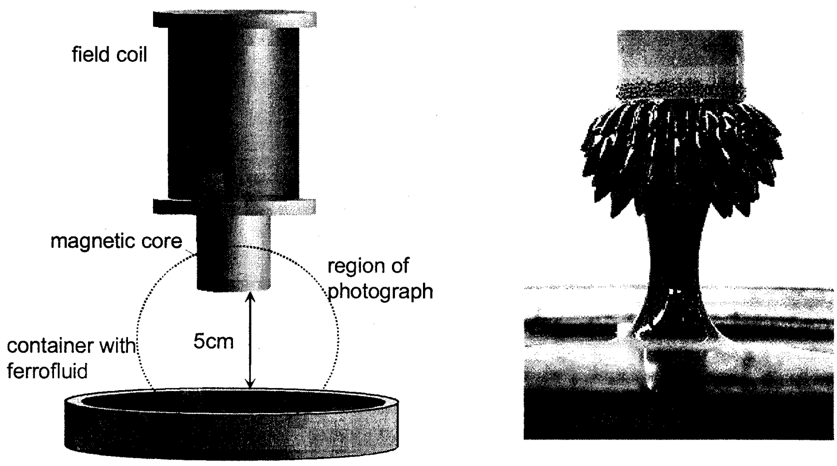

Fig. 2. Attraction of a magnetic fluid by the pole of an electromagnet.

For the discussion of magnetoviscous effects, the question of relaxation of magnetization has major importance. In principle two different relaxation processes are possible. On the one hand the particles themselves can reorient in the field, while the magnetic moment is fixed within the particle. This process, called Brownian relaxation,[12] is described by a corresponding relaxation time.

$$\tau_{\mathrm{B}} = \frac{3\tilde{V}\eta}{kT} \tag{2}$$

where $\tilde{V}$ denotes the particle volume including the surfactant and η is the dynamic viscosity of the fluid.

On the other hand the magnetic moment can change its direction inside the particle and thus magnetization can relax without movement of the particle itself. The process of Néel relaxation takes place when the thermal energy is high enough to enable the magnetic moment to overcome the energy barrier given by the anisotropy of the particle. It is characterized by the Néel relaxation time[13]

$$\tau_{\mathrm{N}} = f_0^{-1} \exp\left(\frac{KV}{kT}\right), \tag{3}$$

where f_0 is the Larmour frequency of the magnetization vector in the anisotropy field of the particle and K is the anisotropy constant of the magnetic particle. The relaxation of magnetization in a ferrofluid will take place by the process with the shorter relaxation time. From Eqs. (2) and (3) one can see, that both times increases with the size of the particles. While τ_{B} scales only linear in V, τ_{N} increases exponential with particle size. Thus the Néel process will dominate for small particles, which are called magnetically weak, while large particles will relax by Brownian relaxation. Such particles are called magnetically hard. The critical size for which the change between the processes occurs depends on the anisotropy characteristics of the magnetic material as well as on the viscosity of the suspension. For normal magnetite based ferrofluids the critical size is about 13 nm, i.e. particles larger than 13 nm will follow the Brownian process while smaller ones will relax by the Néel process.

2. Magnetoviscous Effects in Colloids Containing Single Particles

2.1. *Rotational viscosity*

In 1969 J. P. McTague[14] investigated the influence of a magnetic field on the viscosity of a suspension of cobalt nanoparticles. Therefore a highly diluted suspension of 6 nm cobalt particles was examined using a capillary viscometer in a magnet gap. The field was directed either in direction of the flow or perpendicular to it. He found that the viscosity increases with increasing magnetic field strength and that the increase for field parallel to the flow is twice as high as for the perpendicular direction.

To explain this phenomenon one has to consider a magnetic particle in a shear flow. Due to viscous friction the particle will rotate in the flow, having the axis of rotation parallel to vorticity of the flow (see Fig. 3). If a magnetic field is applied to the system, the magnetic moment of the particle will tend to align with the field direction. Assuming that the particle is magnetically hard, this will provide an alignment of the particle itself in the flow.

If the field is aligned with the vorticity of the flow, the particles align with the field and will rotate around the field direction. In contrast, if the field is perpendicular to vorticity, the rotation of the particle induced by viscous friction in the flow will cause a disalignment of the magnetic moment and the magnetic field direction. This disalignment will give rise to a magnetic torque trying to align the moment with the field. Since the moment is fixed in the particle, a counteraction of mechanic torque acting on the particle and magnetic torque will appear, resulting in a hindrance of free rotation of the particle in the flow. Macroscopically this provides an increase of viscosity of the fluid. Obviously the increase of viscosity is anisotropic and depends strongly on the angle between magnetic field direction and vorticity. This explains the difference between different relative arrangements of flow and field observed by McTague. In the case of parallel alignment of flow and field, vorticity is always perpendicular to the field. In the other case, a distribution of angles between vorticity and field occurs, resulting in a mean angle which gives rise to half of the increase in viscosity observed in the parallel arrangement.

As a first step to a theoretical explanation, Hall and Busenberg[15] assumed a system of particles totally aligned with the magnetic field direction. For this system they extended Einsteins[16] well known relation for the viscosity of a fluid with suspended particles by introduction of a magnetic term

$$\eta = \eta_c \left(1 + \frac{5}{2}\phi_1 + \frac{3}{2}\phi' \sin^2 \varphi_s\right) . \tag{4}$$

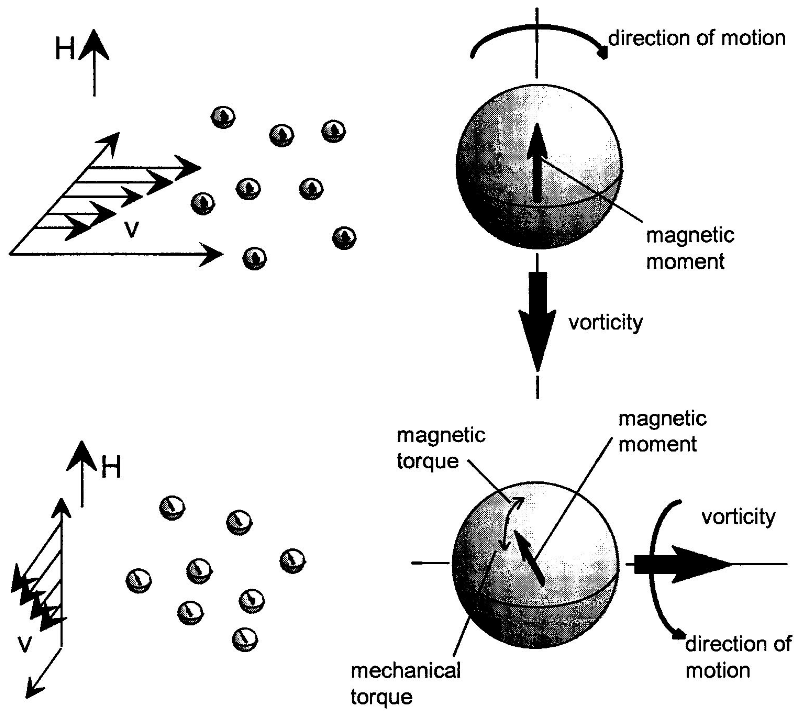

Fig. 3. On the generation of field induced viscosity changes in a ferrofluid. Explanation see text.

Here ϕ_1 denotes the volume concentration of all suspended material, i.e. particles with surfactant, free surfactant etc. and ϕ' means the volume concentration of the particles with surfactant, η_c is the viscosity of the carrier liquid. The term $\sin^2 \varphi_s$ contains the magnetic effects and is a function of the angle between field and vorticity as well as of the relation of magnetic and mechanic torque. While this approach is able to explain the factor 2 between the two different field directions, it does not give a reasonable result for the absolute value of the viscosity increase since a total alignment of the particles is assumed and the action of Brownian motion is completely neglected.

These backdraws of the theory in Ref. 15 forced Shliomis to set up a new approach for the description of the magnetic field effect on particle rotation in a ferrofluid.[17] First of all he showed by an energy argument, that Brownian motion is essential for the description of the process. The simple comparison of magnetic and mechanic energy, as it was used in Ref. 15 leads to a saturation field for the total alignment of the particles of about 1 kA/m; approximately three orders of magnitude smaller than the values found experimentally.

Shliomis introduced a new variable, the density of internal angular momentum

$$S = I\omega \tag{5}$$

with I the density of the moments of inertia of the particles and ω their angular

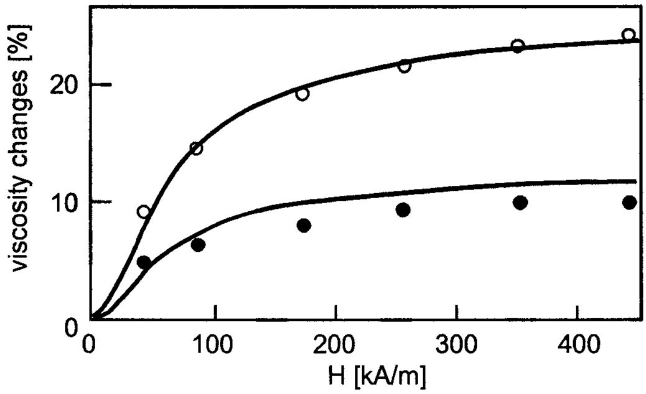

Fig. 4. The comparison of Shliomis' theory on rotational viscosity with the results by McTague[14] (after Ref. 17).

velocity. Putting this into the equation for the relaxation of magnetization of the system he obtains an expression that is to be used in the equation of motion of the fluid. With a specialization of the problem to a one-dimensional Couette flow, he ended up with an expression for the components of the stress tensor allowing to extract an equation for the viscosity of the suspension. This equation contains a quantity summed to the usual viscosity of the suspension in absence of a magnetic field, describing the field dependent viscosity changes. This part of viscosity is commonly called rotational viscosity $\eta_{\mathrm{r}} = \eta_{(H)} - \eta_0$

$$\eta_{\mathrm{r}} = \frac{3}{2}\phi' \eta_0 \frac{\alpha - \tanh \alpha}{\alpha + \tanh \alpha} \langle \sin^2 \beta \rangle \tag{6}$$

where η_0 is the viscosity of the suspension in absence of a magnetic field and β denotes the angle between magnetic field direction and vorticity ($\langle \cdots \rangle$ means a spatial average). This equation did not only fit with the factor 2 difference of the two different field directions but does also quantitatively well agree with the experimental data as shown in Fig. 4.

It should be recalled that the assumptions made for the theoretical approach, in particular the postulation of a suspension of non interacting particles which are magnetically hard, are well fulfilled by the experiment. The 6 nm Co-particles used in Ref. 14 show relaxation after the Brownian process, and the magnetic volume fraction of about 0.02 vol.% is small enough to avoid an influence of magnetic interaction.

Shliomis[17] notes that the relatively simple flow geometries used in Ref. 14 and for the calculation in Ref. 17 are favorable for the comparison of experimental data with the respective theory, and that more complicated flow geometries may be influenced by numerous additional magnetic field effects. A proof of Shliomis' theory in a complex flow geometry has been performed using the transition from Couette to Taylor vortex flow.[18,19] In these experiments the determination of the

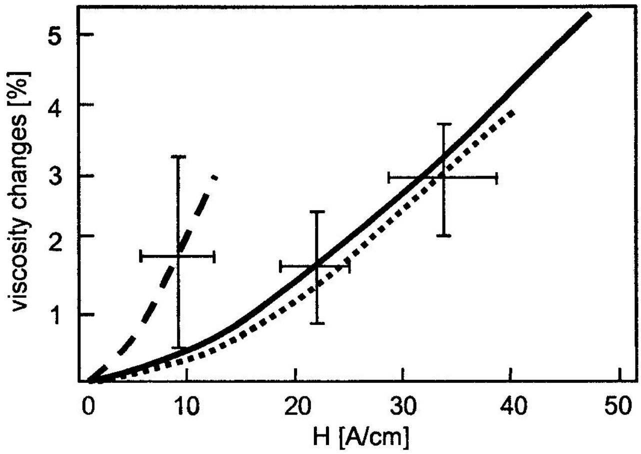

Fig. 5. The comparison of the relative change of viscosity for various angles between field direction and vorticity as obtained in Refs. 18 and 19.

field dependent shift of the critical frequency for the transition between the laminar flow state and the vortex flow has been used as a measure for the viscosity changes. This is possible, since the transition frequency depends only on the viscosity of the fluid as long as the geometry of the system is fixed. Using the general symmetry of the system, three different magnetic field directions can be realized, axial, azimuthal and radial fields. All three fields have a different relative angle with respect to the mean direction of vorticity of Taylor vortex flow.

Thus the term $\langle \sin^2 \beta \rangle$ could be varied and a comparison of the results was performed. In Fig. 5 the relative change of viscosity normalized by $\langle \sin^2 \beta \rangle$ is plotted for the various field directions. Within the margin of error all three curves coincide and thus the $\langle \sin^2 \beta \rangle$ law — and therefore the description for the anisotropy of rotational viscosity — are confirmed. Discrepancies appearing in a quantitative comparison of Eq. (6) with the results in Refs. 18 and 19 will be discussed later in the context of the influence of agglomerate formation on the viscosity of magnetic fluids.

2.2. *Negative viscosity*

It has been discussed above, that the equilibrium of magnetic torque, exerted by a magnetic field to the particles and viscous torque due to differences between the angular velocities of particles and flow

$$M \times H = 6\pi\phi\,(\omega_{\mathrm{p}} - \Omega) \tag{7}$$

results in a hindrance of free rotation of the particles in a static field.

We will now discuss the effect of an alternating magnetic field on particle rotation and thus on the viscosity of the fluid. Assuming a fluid on rest and an applied rotating magnetic field, the particles will start to rotate due to the action of the magnetic torque. Nevertheless, this will not lead to macroscopic effects, since there is no preferable direction for the particle rotation, and thus the mean rotation will be zero. If a shear flow is applied, the degeneracy of the direction of rotation of the particles is broken and the field induced rotation will affect the viscosity of the flow. The dependence of rotational viscosity on the angular velocity of particle rotation can be written in the form[20]

$$\Delta\eta = \frac{3}{2}\eta_0\phi\frac{\Omega - \omega_{\mathrm{p}}}{\Omega}\,. \tag{8}$$

For small frequencies of the magnetic field the angular velocity of particle rotation ω_{p} will remain smaller than Ω and thus the free particle rotation will be impeded, resulting in the well known viscosity increase. For large field frequences ω_{p} may become larger than Ω and the particles will spin up the flow. This will finally give rise to a reduction of viscosity due to a negative field dependent term in the expression for viscosity.

By an extension of the original concept of rotational viscosity[17] to alternating magnetic fields, Shliomis and Morozov were able to predict the discussed phenomenon of field dependent reduction of viscosity quantitatively.[21] For small magnetic field they found

$$\Delta\eta = \frac{1}{4}\eta_0\phi'\alpha^2\frac{1-\omega^2\tau_{\mathrm{B}}^2}{(1+\omega^2\tau_{\mathrm{B}}^2)^2} \tag{9}$$

where ω denotes the frequency of the magnetic field. For a static field ($\omega = 0$) Eq. (9) gives the same result for the field induced increase of viscosity as it is obtained in small field approximation of Eq. (6). For increasing ω the rotational viscosity decreases, and for $\omega\tau_{\mathrm{B}} = 1$ $\Delta\eta$ becomes zero. Higher frequencies will result in a spin up of the flow due to the rotation of the particles and thus in a negative $\Delta\eta$. Furthermore in Ref. 21 a detailed analysis of the field and the frequency dependencies of rotational viscosity for arbitrary field strength has been performed, allowing a comparison of theory with experiments.

The first experimental evidence for this so called "negative viscosity" effect in ferrofluids has been given by Bacri *et al.*[22] To fulfill the condition of magnetic hardness of the particles they used a Co-ferrite fluid containing 10 nm particles. Figure 6 shows the relative rotational viscosity as a function of magnetic field strength for various frequencies of the alternating field.

The data for static field ($\omega = 0$) shows the well known increase of rotational viscosity with field strength. In qualitative agreement with theory,[21] this increase reduces with increasing frequency of the field and becomes even negative at very high frequencies.

Nevertheless, since the assumption of small fields used for Eq. (9) is violated in the experiment, a quantitative comparison of the data with theory can only be made with the extended model. Figure 7 shows the isolines for rotational viscosity normalized to it's static high saturation value $\eta_{\mathrm{r}(H,\omega\tau_{\mathrm{B}})}/\eta_{\mathrm{r}(\infty,0)}$ from the experimental data as well as from the calculation in Ref. 21. Obviously qualitative agreement

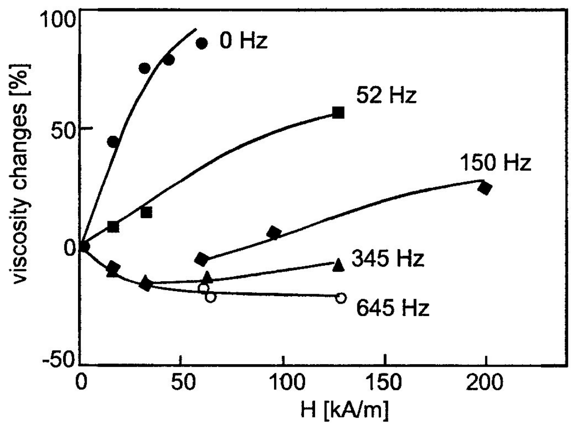

Fig. 6. The reduced rotational viscosity of a cobalt ferrite ferrofluid in an alternating magnetic field for various frequencies (after Ref. 22).

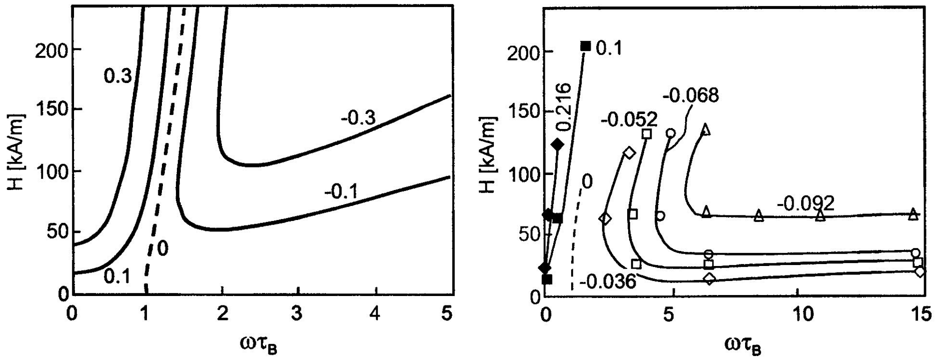

Fig. 7. Map of rotational viscosity as function of field strength and frequency of field oscillation. The curves show isolines of $(\eta_r/\eta_0)\,/\,(3\phi'/2)$. Left side: theoretical prediction[21]; right side: experimental results.[22]

between theory and experiment is found here. In particular η_r changes its sign for $\omega\tau_B = 1$ as predicted by the theory.

The disagreement concerning the absolute values has been related to cooperative phenomena, since a highly concentrated fluid has been used. More significant differences between experimental results and theory have been found using commercial ferrofluids.[23] This may also be related to cooperative phenomena like aggregation or chain formation, like they'll be discussed in the next section.

3. Magnetoviscous Effect in Presence of Interparticle Interaction

3.1. *Effects in commercial magnetic fluids*

In the previous chapter most of the discussed experimental data has been obtained using cobalt or cobalt ferrite particles. In these cases the high anisotropy of the magnetic material ensures that even particles with diameters about 10 nm are magnetically hard and can thus contribute to viscous effects basing on the influence of a magnetic field on rotation of the magnetic particles. In addition in the case of McTagues[14] experiments the concentration of magnetic material was extremely low — enabling a direct comparison with theory. In contrast high concentrated fluids as used in Ref. 22 show quantitative disagreement which may be related to cooperative phenomena. In addition the major part of the nowadays used ferrofluids is based on magnetite particles with a diameter about 10 nm. As known, these fluids are concentrated up to volume concentrations of magnetic material of about 15 vol.% and have usually a relatively broad particle size distribution. Thus interaction of the particles may not be *a priori* neglected and since the particle size enters the rotational viscosity by the sixth power, the particle size distribution may give rise to significant changes in the absolute values of rotational viscosity. Besides this, for magnetite particles it must be observed, that the critical size, above which the particles are magnetically hard, is about 13 nm. Thus a majority of particles in a normal ferrofluid should relax by the Néel-process and should not contribute to effects like rotational viscosity.

It has already been mentioned in Chapter 2, that experiments with commercial magnetite based magnetic fluids have shown significant differences in comparison with theory. Figure 8 shows the relative magnetoviscous effect $\Delta\eta/\eta_0$ for a commercial ferrofluid containing magnetite particles.[18] The fluid — APG 513 A from Ferrofluidics — is based on a Di-ester liquid and contains about 7.2 vol.% of magnetic material. The mean size of the partuicles is about 10 nm and the size distribution

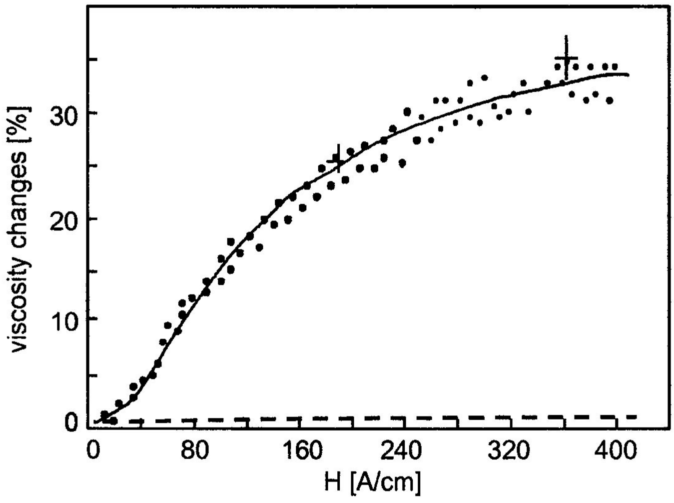

Fig. 8. The relative magnetoviscous effect $\Delta\eta_{(H)}/\eta_0$ as a function of magnetic field strength measured in APG 513 A for shear rate $\dot{\gamma} = 500\ \mathrm{s}^{-1}$. The dashed line gives the values as calculated from Eq. (6) using the fluids data. The solid line is a fit of Eq. (6) to the experimental data with $\bar{d} = 16$ nm.

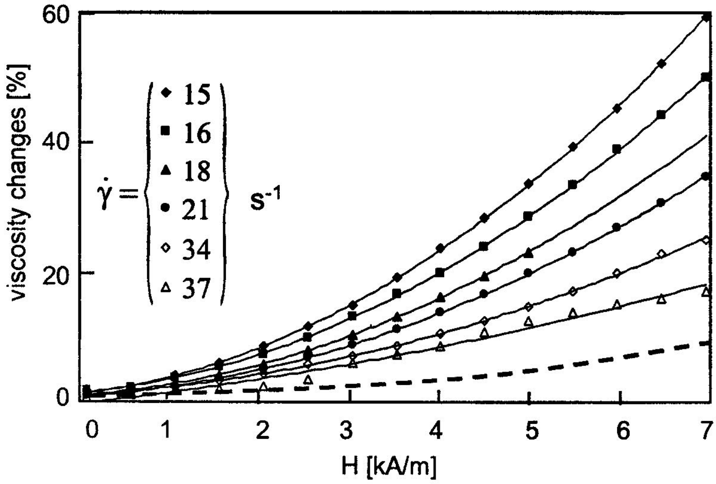

Fig. 9. Shear dependence of the magnetoviscous effect in APG 513 A.[26] The dashed line gives the data for the same fluids as obtained for $\dot{\gamma} = 500\ \mathrm{s}^{-1}$ in Ref. 18.

as obtainbed from a magnetogranulometric analysis[24,25] shows only a small portion of particles with diameters above the critical diameter of 13 nm. A comparison of the experimental data in Fig. 8 with the theoretical curve for rotational viscosity calculated from Eq. (6) on the basis of the fluids data and using the sizes of particles from the size analysis, shows a quantitative difference of about one order of magnitude. On the order hand, it is possible to fit Eq. (6) to the experimental data, providing a relevant diameter of the magnetic core of the particles of about 16 nm. The qualitative agreement shows that the measured effect bases on the hindrance of rotation of magnetic structures in the fluid. On the other hand the quantitative disagreement leads to the assumption that interaction between the particles may play an important role. This is strengthened if one tries to vary the shear acting on the fluid. In Fig. 9 data for the same fluid obtained with a specialized rheometer for magnetic fluids[26] at low shear rates is shown together with the data from Ref. 18. First of all the absolute value of the magnetoviscous effect observed for small magnetic fields is even higher than the saturation value $\Delta\eta_{\mathrm{r}}^{\max}/\eta_0 = 3\phi'/2$ expected from Eq. (6). Futhermore, a clear shear dependence of the field induced change of viscosity is observed. This field dependent shear thinning was explained in Ref. 27 by the assumption of formation and breakage of chains of magnetic particles. The chain formation requires a sufficient dipole–dipole interaction between the particles which is not present for 10 nm magnetite particles. On the other hand it is well known, that agglomerates are formed during the production process of a ferrofluid.[28] While large agglomerates are separated from the fluid by centrifuging or magnetic separation, smaller ones may remain. Such agglomerates, usually dimers with a common surfactant, have significant influence on the magnetoviscous behavior of ferrofluids. This was shown e.g. in numerical simulations by Satoh *et al.*[29]

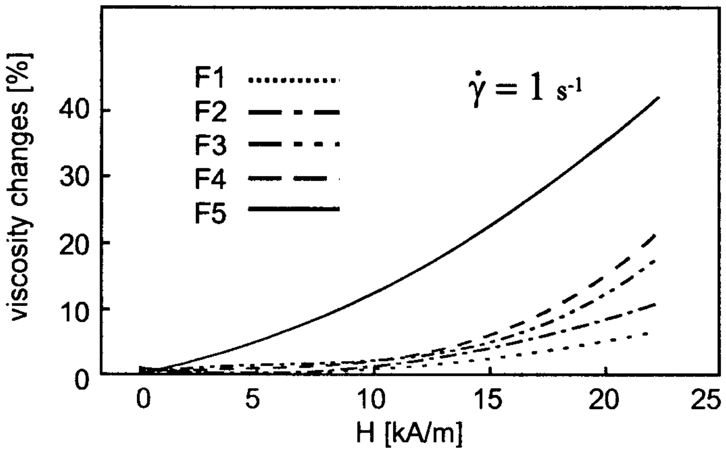

Fig. 10. The comparison of the relative magnetoviscous effect for fluids with different content of agglomerates for $\dot{\gamma} = 1\ \mathrm{s}^{-1}$. The content of agglomerates is indicated by the ordinal number, with F1 being the most purified liquid.[30]

Assuming the existence of primary agglomerates, they simulated the formation of chains and rod-like structures in the fluid and calculated their influence on viscosity of the ferrofluid. Using data comparable to the experimental conditions in Ref. 27 they found good agreement of their simulation with experimental results.[29]

Nevertheless the importance of primary agglomerates for the appearance of strong magnetoviscous effects had to be proved experimentally. Therefore we recently carried out a series of experiments using five different ferrofluids with varying amount of primary agglomerates.[30] The fluids have been taken from a common production batch but have been purified to different degree. Thus they contain a decreasing amount of agglomerates, while the overall magnetic concentration has been fixed to avoid effects due to concentration induced changes of interaction. The results for the magnetoviscous effect for shear rate $\dot{\gamma} = 1\ s^{-1}$ for all five fluids is shown in Fig. 10. The amount of agglomerates is indicated by the ordinal number — the highest number corresponds to the highest amount of agglomerates.

Obviously the magnetoviscous effect decreases with a decreasing amount of agglomerates and for a highly purified fluid it nearly disappears even at extremely low shear rate. With increasing shear rate the magnetoviscous effect reduces and falls below the detection limit for the various fluids successively. Interesting is the comparison of the data for the most purified liquid (F1) with the theory from Ref. 17. Using the total volume fraction of magnetic material and a mean diameter of 10 nm as obtained from magnetic measurement, theory and experimental data for F1 seem to coincide for $\dot{\gamma} = 1\ s^{-1}$. This is obviously misleading since — from the rest of the data — it is clear that the small amount of agglomerates dominates the magnetoviscous behavior of the fluid. In additon such a comparison would neglect the fact that only a small portion of particles with sufficiently large diameter can contribute to rotational viscosity.

Nevertheless, this sheds a light on problems that may appear if highly purified commercial ferrofluids are used in experiments. In this case agreement between the classical theory of rotational viscosity[17] and experimental data may be identified while in reality only the small amount of agglomerates reduces the magnetoviscous effect to a value comparable with the theoretical prediction. This may also explain the disagreement between the result from Refs. 18, 19 and 26, 27 and those found for a commercial ferrofluid in a shear free investigation.[31]

In this case the damping of a shear free solid-body rotation of a ferrofluid, which is induced by internal rotational friction, is used to determine the magnetoviscous effect. This technique opens fascinating possibilities for the determination of the magnetoviscous effect for $\dot{\gamma} = 0$ — a situation preferable for theoretical investigations.

The question which kind of microstructure gives rise to the field induced changes in viscosity of suspensions of magnetic nanoparticles will require future efforts to be finally clarified. Nevertheless, the results discussed before strengthen the hypothesis that primary agglomerates and formation of chains of these agglomerates dominate the magnetoviscous behavior.

3.2. *Evidence for viscoelastic effects*

The formation of chains of magnetic particles in magnetic fluids will not only enhance the field dependent increase of viscosity in a ferrofluid. It will also provide the possibility to generate field dependent viscoelastic effects which may give rise to new classes of applications for ferrofluids.

The generation of viscoelasticity in suspensions of magnetic particles is known from the so-called magnetorheological fluids (MR fluids). These fluids contain micronsized magnetic particles which exbihit strong interaction providing drastic changes from viscous to viscoelastic behavior controlled by magnetic fields (see e.g. Ref. 32). Their backdraw is the sedimentation of the particles, reducing the stability of the suspensions. A liquid with viscoelastic properties basing on nanosized particles could thus be a promising development. An evidence, that such fluids are possible have been given by Kormann *et al.*[33] with investigations of so-called nano-MR fluids, containing particles in a size range about 30 nm.

In addition theoretical predictions have been made, lining out that chain formation should give rise to the appearance of viscoelastic effect in ferrofluids. In particular Zubarev *et al.* (see e.g. Refs. 34–36) analyzed the viscous properties of magnetic suspensions under the assumption of existence of chain like structures. One of their predictions,[36] the appearance of normal stress differences in ferrofluids has been the basis for an experimental search for the existence of a famous viscoelastic phenomenon — the Weissenberg effect[37] — in commercial ferrofluids. This effect, the rise of a free surface of a viscoelastic fluid at a rotating axis,

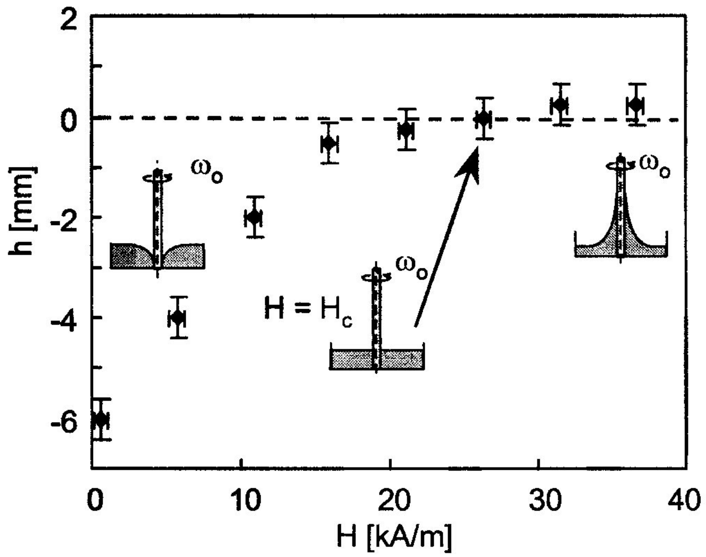

Fig. 11. The change of the height of the free surface of a magnetic liquid at a rotating axis due to the appearance of field induced normal stress difference.[39]

should be field and shear dependent in a ferrofluid with chain formation of magnetic particles. For vanishing field the liquid behaves Newtonian and thus the free surface lowers at the rotating axis due to the action of centrifugal forces. With increasing field strength chains are formed and give rise to normal stress differences, forcing the free surface to rise. At a certain critical field strength at which centrifugal forces and normal stress effects compensate, the fluid surface becomes flat. This is a preferable point for the determination of the combination of normal stress coefficients characterizing the Weissenberg effect, since from the relation for the height of the fluid surface h at the axis[38]

$$h_{(r_0)} = \frac{r_0^2\omega_0^2}{g}\left(-\frac{1}{2} + \frac{\nu_{10} + 4\nu_{20}}{\rho r_0^2}\right) \tag{10}$$

one can derive for the flat surface

$$\nu_{10} + 4\nu_{20} = \frac{1}{2}\rho r_0^2\,. \tag{11}$$

Here r_0 and ω_0 are the radius and angular velocity of the rotating axis, g denotes gravitational acceleration, ν_{10} and ν_{20} are the low shear limits of the normal stress coefficients of the fluid and ρ is its density. The normal stress coefficients ν_1 and ν_2 are defined via the normal stress differences $N_1 = \tau_{11} - \tau_{22}$ and $N_2 = \tau_{22} - \tau_{33}$ (with the main axis components of the stress tensor τ_{ii}) in the form $\nu_i = N_i/\dot{\gamma}^2$.

At higher field strength the known rise of the free surface at the axis will appear. With increasing shear rate the chains will be broken and higher field strength will be needed to induce normal stress effects sufficient to compensate the centrifugal forces. In usual commercial ferrofluids these effects are too small to be observed in normal terrestrial experiments. But Eq. (10) shows, that an amplification of the elongation effects of the free sufrace is possible by performing the experiments under reduced gravity conditions. This has been done[39] using parabolic flights providing a microgravity environment of about 20 s. Figure 11 shows the result for the height of the free surface at the rotating axis as a function of magnetic field strength for APG 513 A, the fluid discussed earlier in the context of rotational viscosity. It is clearly seen, that the free surface rises with increasing field strength, and that it finally exceeds over the original flat surface. From the transition point from negative to positive surface deformation one could determine $\nu_{10} + 4\nu_{20} = 4.10^{-2}$ kg/m for H = 30 kA/m in a shear flow with $\dot{\gamma} = 0.57\ \mathrm{s}^{-1}$.

This has been the first evidence for existence of normal stress differences and thus viscoelasticity in commercial magnetic fluids containing magnetite particles with a mean diameter about 10 nm.

4. Conclusion and Outlook

As it has been shown, suspensions of nanosized magnetic particles exhibit strong changes of their rheological properties under influence of magnetic fields. Generally the change of viscosity can be related to the change of the relative angular velocity of magnetic particles or agglomerates and the fluid due to the action of a magnetic field. In static fields and at a low frequencies of alternating fields a hindrance of the free rotation of the magnetic material appears resulting in an increase of viscosity, while high frequency field will force a faster rotation of the particles, spinning up the fluid and thus reducing it's viscosity.

It has been discussed that these effects are well understood in dilute suspensions of magnetically hard particles, e.g. cobalt ferrite particles. On the other hand high concentration induces effects due to interaction which are currently under investigation. In commercial ferrofluids with magnetite particles, effects have been observed which cannot be explained by a single particle model. The assumption of existence of magnetic particle dimers and of chain formation of these primary agglomerates leads to explanations for the strong magnetoviscous effects observed, as well as for the experimentally verified existence of viscoelasticity. Futhermore it has been shown that the working hypothesis of a dominance of the effect of the primary agglomerates on magnetoviscosity of commercial ferrofluids is supported by recent results from fluids with different agglomerate content.

To get deeper insight to magnetoviscous effects in ferrofluids and their microscopic reasons further experiments will be necessary. In particular, neutron small angle scattering experiments could provide an excellent tool to build up a relation between fields dependence of viscous behavior and relevant microstructural changes. A further development of the fluids, especially using the fact that the large amount of small particles provides a magnetic background enhancing the interaction between the larger particles or angglomerates,[40] may enable the development of applications using stable magnetoviscous fluids.

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