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Field and concentration dependence of chain formation in magnetic fluids

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Abstract. The field and concentration dependence of chain formation in magnetic fluids is investigated. The results show that the chain length increases rapidly with the field and then continues to increase gradually at higher fields approaching saturation. Moreover, the results show that the chain length at a given field is linear with the concentration of the fluid. The correlation between chain formation and some magneto-optic effects is discussed.

1. Introduction

Magnetic fluids, often called ferrofluids, are colloidal suspensions of single domain ferromagnetic particles (Fe_3O_4, iron or cobalt) in a suitable liquid carrier. It is well established that magnetic fluids exhibit magneto-optic effects such as birefringence, dichroism, Faraday rotation and Faraday ellipticity.

It has been suggested that chain formation has an important role in the optical properties of magnetic fluids [1–4]. Rousan *et al* [4] have studied the role of chain formation in the transmission of light in magnetic fluids and also have investigated the role of chain formation in Faraday rotation [5]. Transient studies of light transmission and Faraday rotation in magnetic fluids [6] have indicated the effects of chain formation on magneto-optic effects in magnetic fluids.

Colloidal suspensions of ferromagnetic particles in magnetically passive liquid carriers have been found in different states of aggregation depending upon temperature, density and external magnetic fields. These systems have been observed to form long chains as well as large compact clusters [7–10].

Theoretical studies of chain formation in magnetic fluids based on particle–particle interaction by De Gennes and Pincus [11] and by Jordan [12, 13] predicted a particle chain length, l, proportional to C or $C^{1/2}$ where C is the concentration of the magnetic fluid.

Some experimental observations of chain formation have been indirect; Haas and Adams [14] concluded that chains had formed from the diffraction pattern characteristic of a grating found when light is passed through a sample of magnetite-based ferrofluid perpendicular to the applied field. Popplewell *et al* [15] have investigated the chain formation in magnetic fluid composite thin films and have shown both experimentally and by Monte Carlo simulation that extensive chain formation occurs in relatively weak magnetic fields. They have also shown that the chain length varies with the concentration according to the empirical formula $l \propto C^{3/2}$.

Recently, a detailed study on the field-induced agglomeration in thin films of water-based magnetic fluids has been reported by Jones and Niedoba [16]. In their work, a drop of undiluted fluid was sandwiched between two parallel glass cover slips and placed normal to the optic axis of the microscope. The magnetic field was applied parallel to the axis of the microscope, i.e. perpendicular to the plane of the thin film sample. In this experimental arrangement Jones and Niedoba observed the number of chains per unit volume, n_v, and found it to increase rapidly with the applied field.

In this paper the field and concentration dependence of chain formation in Fe_3O_4 particle magnetic fluid is presented. The saturation magnetisation of the samples are 5, 10, 15, 20 and 25 G. Magnetic fields up to 900 Oe are used in this work.

2. Experiment

The magnetic fluids used in this work were contained in a 0.2 mm thick rectangular cell. The cell was placed between the poles of an electromagnet specially made to fit onto an optical microscope. Magnetic fields up to 900 Oe were generated using this electromagnet. The chains formed in the fluid were viewed normal to the applied magnetic field and the plane of the fluid. Optical micrographs of the sample were taken at different magnetic fields ranging from ~150–900 Oe.

Micrographs were also obtained by viewing the sample parallel to the magnetic field and normal to the plane of the fluid. In this case a field (200 Oe) was generated by a coil. The viewing arrangement of the sample is shown in figure 1. By shining a laser beam perpendicular to both the magnetic field and the plane of the magnetic fluid a diffraction pattern was obtained.

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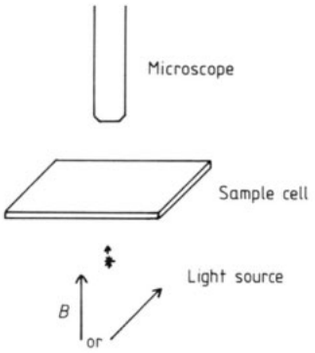

Figure 1. Viewing arrangement of the chains formed in magnetic fluids.

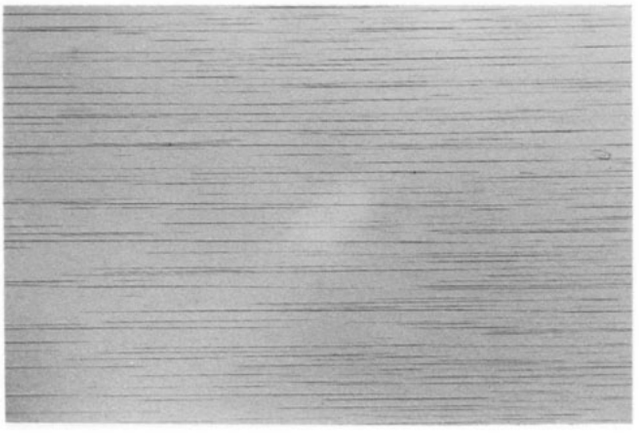

Figure 2. An optical micrograph for 15 G saturation magnetisation magnetic fluid under 420 Oe magnetic field parallel to the plane of the fluid.

The samples used in this work were Fe_3O_4 particle magnetic fluids with Isopar M as a liquid carrier. The saturation magnetisation of the samples were 5, 10, 15, 20 and 25 G.

3. Results

A typical optical micrograph is shown in figure 2 for a 15 G saturation magnetisation sample in the presence of a 420 Oe magnetic field. Because the sample is thick compared to a thin film, a contrast between chains well in focus and others at different depths in the sample which are out of focus is seen. Although the chain length is not uniform, an average was obtained by measuring the length of a relatively large number of chains.

In figure 3 the average length of chains for the 15 G saturation magnetisation is plotted versus magnetic field. The results in the figure show a rapid increase in the chain length with the field for low fields, and then a slow gradual increase at higher fields with a definite trend towards saturation.

In figure 4 the average length of chains at a magnetic field of ~420 Oe is plotted versus the saturation

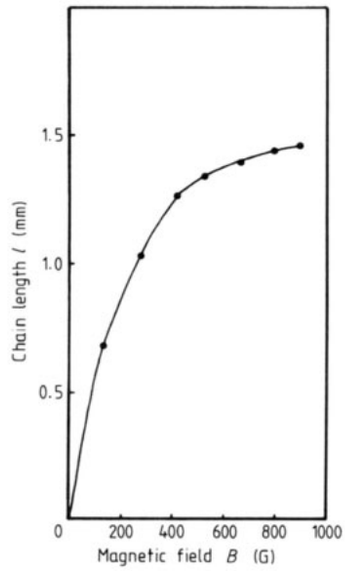

Figure 3. The average chain length l in a 15 G saturation magnetisation magnetic field versus applied magnetic field B.

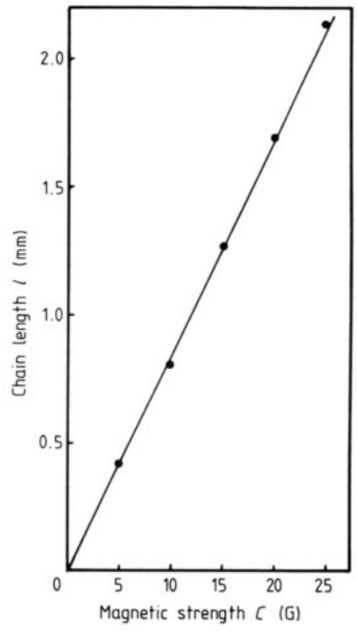

Figure 4. The average chain length at $B = 420$ Oe versus the saturation magnetisation I_s of the fluid.

magnetisation of the sample. The results show a linear relationship with the saturation magnetisation of the sample in the range of concentration used in this study.

In figure 5 an optical micrograph obtained by viewing the sample parallel to the magentic field is shown. The micrograph shows a pattern of dark spots representing the cross sections of the already formed chains. In figure 6 the diffraction pattern observed when the laser beam is shined perpendicular to the magnetic field and the plane of the sample is shown. It is worth mentioning that this field-induced chain formation and

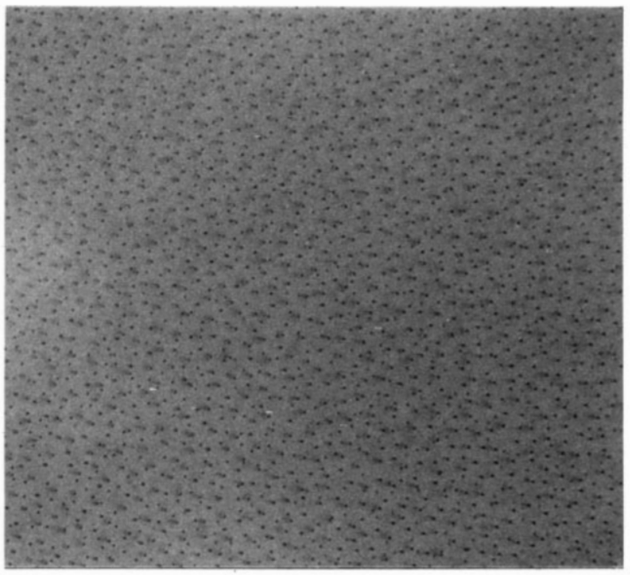

Figure 5. An optical micrograph obtained by viewing the sample parallel to an applied magnetic field $B = 200$ Oe.

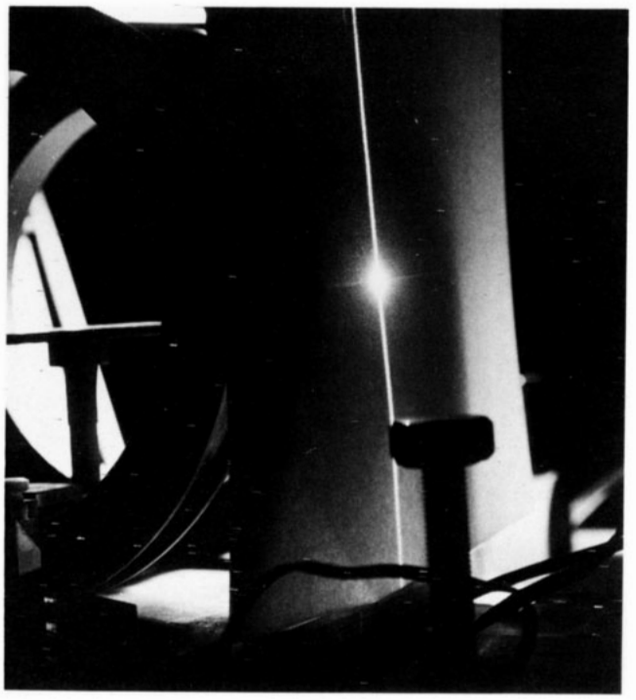

Figure 6. The diffraction pattern observed by shining a laser beam normal to the field and plane of the fluid.

the diffraction effect are temporary, and that the fluid reverses to its original colloidal state upon the reduction of the applied field to zero.

4. Discussion

In the presence of a magnetic field, the particles in the fluid tend to align along the field direction; and dipolar interactions lead to chain formation along the field direction.

The magnetic interaction between two particles separated by a distance r is given by

$$U = r^{-3}[\boldsymbol{\mu}_1 \cdot \boldsymbol{\mu}_2 - 3(\boldsymbol{\mu}_1 \cdot \boldsymbol{r})(\boldsymbol{\mu}_2 \cdot \boldsymbol{r})/r^2]. \qquad (1)$$

For a given average separation (given concentration) the interaction energy is a function of magnetisation. Because the magnetisation saturates at high fields, the interaction energy will also saturate. Consequently, chain formation is expected to saturate. Our results in figure 3 show that the average length of the chains along the field direction is indeed tending to saturate at high fields. The same behaviour has also been observed in the transmission of light [4] and Faraday rotation in magnetic fluids [5].

Changing the concentration will change the average separation between the particles. Because the sample used is three-dimensional, the average spacing between the particles is proportional to $C^{-1/3}$; and hence the interaction energy is proportional to C. Therefore, the chain length for low concentration (i.e. nearest-neighbour interaction) is expected to be linear with C.

Our results in figure 4 show a linear relation between the chain length and concentration. Noting that the magnetic fluid composites studied by Popplewell *et al* [15] are two-dimensional samples, one can explain their results $l \propto C^{3/2}$ by a similar argument.

The optical micrograph in figure 5 obtained by viewing the sample parallel to the applied field, similar to the micrographs obtained by Jones and Niedoba [16], shows a pattern of dark spots that represent the cross sections of chains already formed in the fluid.

The diffraction pattern shown in figure 6 is a result of chain formation and indeed is similar to what Haas and Adams have observed [14]. Similar to Haas and Adams, we feel that it is due to the thermal fluctuation and the non-uniform nature of the spacing between chains that only a straight line is seen and not maximum and minimum intensity positions typical of diffraction gratings.

The experimental arrangement used by Jones and Niedoba [16] with a 4 μm sample thickness put an upper limit on the chain length, which is supposed to be $\sim$4 μm. This chain length is easily reached even at low fields. When the first group of chains has reached this length new chains appear; therefore the number of chains per unit volume, n_v, increases. It is important to note that the rate of increase in n_v increases with the field in the range of fields used by Jones and Niedoba [16]. However, our results on chain length versus field in figure 3 show that the rate of increase in the chain length decreases with the field. These two results are consistent since a new group of chains will be formed only when the previous group is reaching saturation length. The micrograph in figure 2 shows chains with a distribution of chain lengths in support of the above argument. Furthermore, it has also been observed that the spacing between the chains for the same sample decreases with the applied magnetic field.

The longitudinal transmission coefficient of light through magnetic fluids has been studied experimentally by Yusuf *et al* [3] and by Rousan *et al* [4]. In both studies the transmission coefficient has been found to increase monotonically with the field and approach saturation at high fields. This behaviour has also been predicted by Tasker *et al* [17] using Monte Carlo simulation. The simulation is based on calculating the pair correlation functions in a dispersion of interacting particles as a function of applied field and including geometrical shadowing effects of the particles. Tasker *et al* found that the geometrical shadowing decreases with the field and consequently the transmission increases.

A comparison between our results on the transmission of light [3, 4] and our results on the chain length in figure 3 shows a striking similarity in the behaviour of the two effects. Thus the longitudinal transmission would be principally determined by the chain length for thick samples. Note that increasing the length of the chain, or increasing the number of chains per unit volume, has a similar effect, namely to decrease the geometrical shadowing of the system and consequently increase the transmission of light. Therefore, we feel that the transmission of light is principally determined by the number of chains per unit volume in the case of a thin film sample, while it is principally determined by the chain length in the case of a thick sample.

We now look briefly at the implication of chain formation for Faraday rotation. Yusuf and co-workers [5, 18] have reported measurements of Faraday rotation in magnetic fluids and provided some information on the effects of chain formation on Faraday rotation, concluding that chain formation plays an important role in Faraday rotation. In both works Faraday rotation increased monotonically with the field and approached saturation at high fields. Again, there is a striking similarity between the behaviour of Faraday rotation and chain length with the applied field.

Taketomi [2] and Taketomi *et al* [19] suggested that birefringence and dichroism are due to the anisotropy of the magnetic fluid caused by the formation of needle-like chains formed in the field direction. They also showed [19] that birefringence and dichroism are linear with concentration. In agreement with their work, our results on the chain length versus field also show a linear relationship.

It is of interest to study the chain formation at much higher concentrations and to investigate the validity of the linear relation in the case of strong interactions. Unfortunately, it has been difficult to take optical micrographs due to the low transmission of light through highly concentrated samples.

5. Conclusions

The field and concentration dependence of chain formation in magnetic fluids has been investigated. It is shown that the chain increases rapidly with the field at low fields, then gradually increases at high fields with a definite trend towards saturation. It is also shown that the chain length varies linearly with the concentration of the magnetic fluid in the range of concentrations used in this study.

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