

# Magnetic field and concentration dependence of light scattering from a ferrofluid

ZHANG QI, WANG JIANHUA, ZHU HESUN\*

*Department of Mechanical Engineering, and \*Research Center of Materials Science, Beijing Institute of Technology, Beijing 100081, People's Republic of China*

Measurements of light scattering from a ferrofluid were undertaken as functions of both magnetic field and particle concentration. The results show that the distribution pattern of light intensity in space is a continuous banding perpendicular to the field direction. The light intensity weakens with increasing scattering angle. The experiments also indicate that the scattering coefficient increases both with the magnetic field and with the particle concentration and tends to saturate at higher field strengths. Finally, the experimental results are discussed in terms of an expanded theory of light scattering established by considering the widths of the chains formed in the ferrofluid as functions of both the magnetic field and the particle concentration.

## 1. Introduction

Light scattering from a ferrofluid was first investigated by Haas and Adams [1], who found that light transmitting through a ferrofluid subjected to a magnetic field became scattered. Different from the distribution pattern of light intensity due to a normal grating, the distribution of light intensity scattered from a ferrofluid in a magnetic field was a continuous banding perpendicular to the field direction. The results suggested that there existed a diffraction grating with a continuum of grating spacings in the sample.

By microscope observation of association in a ferrofluid, Hayes [2] observed a series of needles which were approximately  $0.9 \times 10^{-4}$  cm in width. These needles, caused by the interactions between the colloidal particles in an applied magnetic field, were large enough to be observed with an optical microscope. Moreover, Hayes explained the experimental distribution of light intensity in terms of the diffraction theory of one slit.

Tasker *et al.* [3] calculated light transmittance through the paramagnetic particles of a ferrofluid as a function of applied magnetic field, taking into account the interference between the particles by means of an effective extinction cross-section. The transmission of plane polarized light through  $\text{Fe}_3\text{O}_4$  ferrofluid of different particle concentrations, was measured by Rousan [4]. The results show that the transmission coefficient increases with the field strength and tends to saturate with a Langevin-type behaviour similar to the magnetization of magnetic fluids.

We also investigated the light scattering from a ferrofluid in a strong magnetic field [5], where the theory of light scattering was established by taking into account the interferences of the chains, whose widths were believed to follow a normal distribution pattern in a definite area,  $W_{\min}$ ,  $W_{\max}$ , where  $W_{\min}$  and  $W_{\max}$

are the minimum and maximum widths of the chains. The results showed that the normal distribution followed by the widths of the chains greatly influences the light scattering from a sample. The wider the distribution of the chain widths, the weaker the light scattering from a sample became, and only when the expected chain widths approached the wavelength of the incident light did the light scattering become obvious.

In this work, the light scattering from a  $\text{Fe}_3\text{O}_4$  ferrofluid was measured as functions of both magnetic field and particle concentration. The results obtained from the experiments have been discussed by expanding the theory of light scattering given previously [5] and considering the widths of the chains as functions of both the magnetic field and the particle concentration.

## 2. Experimental procedure

Measurements of the light scattering from a ferrofluid were undertaken using an optical arrangement shown elsewhere [5]. A He–Ne laser was used as the light source, with a wavelength of  $0.6328 \mu\text{m}$ . The electromagnet was placed so that its field direction is perpendicular to the optical axis. Its strength can vary from  $0$ – $2.4 \times 10^5 \text{ A m}^{-1}$  with the change in the input electric current. An apparatus with a detector, which can rotate around a permanent point in the plane scattered, was used to detect light intensities from different directions. The detector, together with a phase-locking amplifier, could accurately determine light intensity by transferring light intensity received into voltage. The accuracy of the amplifier can reach  $\pm 1 \mu\text{V}$ .

The ferrofluid used here was  $\text{Fe}_3\text{O}_4$  particles coated with oleic acid and dispersed in an oil. Samples suitable

for measurement in the experiments were prepared by diluting the ferrofluid and coating the diluted solution on the surface of a transparent ribbon. The thickness of a sample determined by a Minitest 2000 thickness detector was about 8.0  $\mu\text{m}$ .

The influence of the magnetic field on light scattering from a ferrofluid was measured experimentally for  $\text{Fe}_3\text{O}_4$  ferrofluid having a particle concentration  $C = 0.02$ . The field strengths were selected as  $0.32 \times 10^5$ ,  $0.67 \times 10^5$ ,  $0.83 \times 10^5$ ,  $1.05 \times 10^5$ ,  $1.53 \times 10^5$  and  $2.0 \times 10^5 \text{ A m}^{-1}$ . For each value of field strength, the corresponding distribution of light intensity was measured and thus the scattering coefficient was calculated.

The measurements of the dependence of particle concentration on light scattering were carried out for three particle concentrations  $C = 0.0145$ ,  $0.0207$  and  $0.0306$ . The field strength was selected as  $H = 2.0 \times 10^5 \text{ A m}^{-1}$ . The distributions of light intensity in space were measured for the three cases and the scattering coefficients corresponding to the three cases were calculated.

### 3. Results

A scattering coefficient is defined to describe the distribution of light intensity scattered from a ferrofluid in space

$$F = 1 - \frac{I_{\theta=0}}{I_{\text{tot}}} \quad (1)$$

where  $I_{\text{tot}}$  and  $I_{\theta=0}$  are the total light intensity and the light intensity at the centre, respectively.

Fig. 1 shows the scattering coefficient, for the sample with constant particle concentration  $C = 0.02$ , plotted as a function of the applied magnetic field. The experiments indicate that the distribution pattern of the rays scattered from the ferrofluid in space is a continuous banding perpendicular to the magnetic field direction. The light intensity scattered decreases with increasing scattering angle. From Equation 1 the scattering coefficient is calculated for different magnetic field strengths. The results in Fig. 1 show that the scattering coefficient increases with increasing applied magnetic field and tends to saturate at higher magnetic fields.

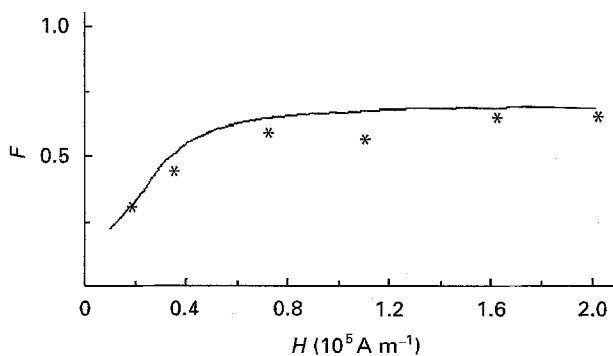


Figure 1 The influence of the magnetic field on the light scattering from a  $\text{Fe}_3\text{O}_4$  ferrofluid under the condition  $C = 0.02$ . (\*) Experimental results, (—) theoretical calculations.

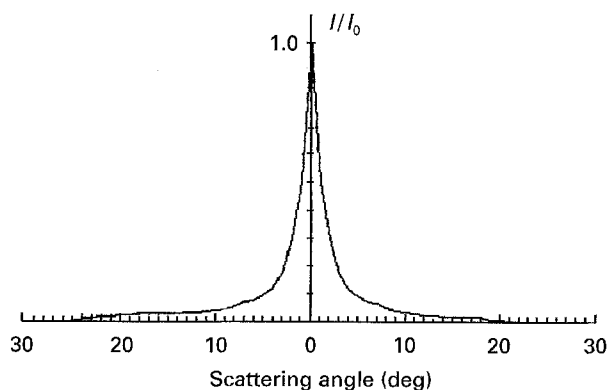


Figure 2 The distribution of light intensity scattered from a  $\text{Fe}_3\text{O}_4$  ferrofluid under the conditions  $C = 0.0145$ ,  $H = 2.0 \times 10^5 \text{ A m}^{-1}$ .

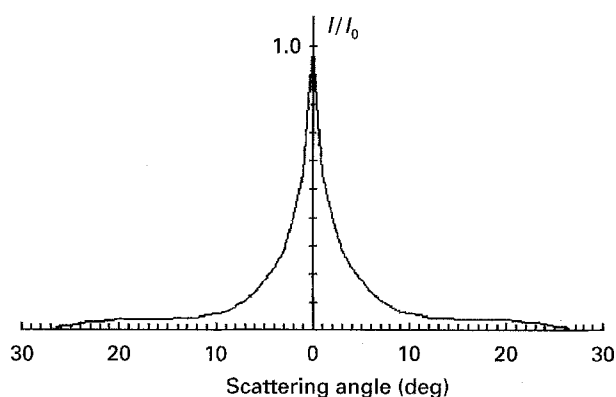


Figure 3 The distribution of light intensity scattered from a  $\text{Fe}_3\text{O}_4$  ferrofluid under the conditions  $C = 0.0207$ ,  $H = 2.0 \times 10^5 \text{ A m}^{-1}$ .

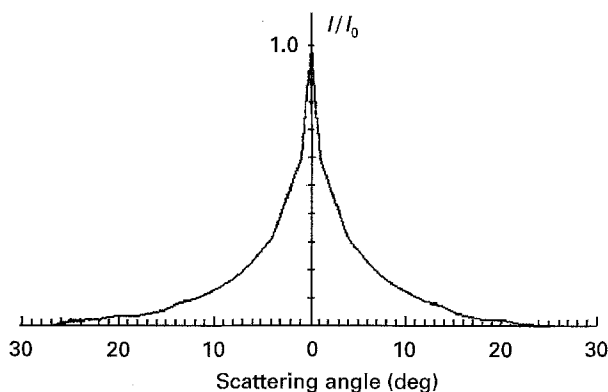


Figure 4 The distribution of light intensity scattered from a  $\text{Fe}_3\text{O}_4$  ferrofluid under the conditions  $C = 0.0306$ ,  $H = 2.0 \times 10^5 \text{ A m}^{-1}$ .

Figs 2–4 show the distributions of light intensity scattered from the ferrofluids having particle concentrations  $C = 0.0145$ ,  $0.0207$  and  $0.0306$ , respectively. Fig. 5 shows the relationship between the scattering coefficient and the particle concentration. From Fig. 5 it can be seen that the scattering coefficient increases with increasing particle concentration.

### 4. Discussion

Light scattering from a ferrofluid is linked closely to the chain structure formed under the influence of a magnetic field. In the presence of a magnetic field, the

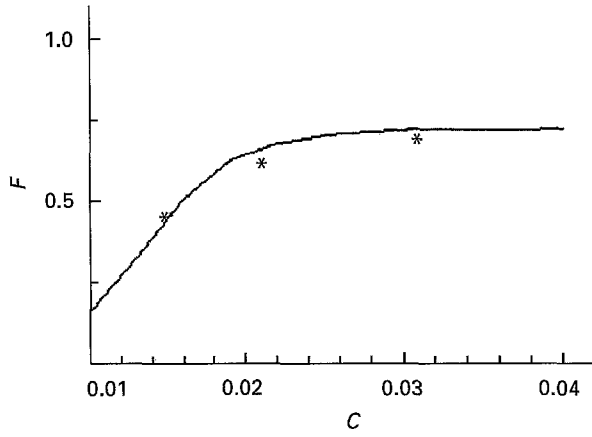


Figure 5 The influence of the particle concentration on the light scattering from a  $\text{Fe}_3\text{O}_4$  ferrofluid under the condition  $H = 2.0 \times 10^5 \text{ A m}^{-1}$ . (\*) Experimental results, (...) theoretical calculations.

particles in the sample tend to align along the field direction, forming chains. The width of the chain is expected to vary depending on both the applied magnetic field and the particle concentration. Therefore, the light scattering from the sample is both magnetic-field and particle-concentration dependent.

The expected width,  $\bar{W}$ , of the chain in a given magnetic field can be described as follows [6]

$$\bar{W} = N P d \exp(-U_i/KT) \quad (2)$$

where  $N$  is the total number of the particles,  $P$  is a constant and  $d$  is the mean diameter of the particles  $U_i$  is the particle potential energy due to the dipole-dipole interactions. According to the model of the mean field approximation, the dipole-dipole interactions are

$$U_i = -m H_i = -m k M \quad (3)$$

where  $m$  is field magnetic moment of a particle,  $k$  is the coefficient of the mean field and  $M$  is the magnetization of the ferrofluid. By combining Equations 2 and 3, we have

$$\bar{W} = \frac{V_{\text{samp}}}{v} C P d \exp(m k M/KT) \quad (4)$$

where  $V_{\text{samp}}$  and  $v$  are the volumes of the sample and one particle, respectively;  $C$  is the particle concentration. Equation 4 shows that both the magnetic field and the particle concentration have an influence on the chain formation in a ferrofluid. Their influences (shown in Figs 6 and 7, respectively) indicate that the expected width of the chain decreases with the increases in both the field strength and the particle concentration and tends to saturate at higher field strengths.

Light scattering from a ferrofluid in a strong magnetic field was investigated previously [5]. The distribution of light intensity in space is given by

$$I_\theta = \frac{N W_0^4 (\epsilon_{11}^2 \sin^2 \beta \cos^2 \theta + \epsilon_{33}^2 \cos^2 \beta) \cos^2 \theta}{\pi^2 \epsilon_0^2 c^4 D^2} \times \int_{W_{\min}}^{W_{\max}} \frac{V^2 [(x - \sin x)^2 + (1 - \cos x)^2]}{x^4} f(W) dW \quad (5a)$$

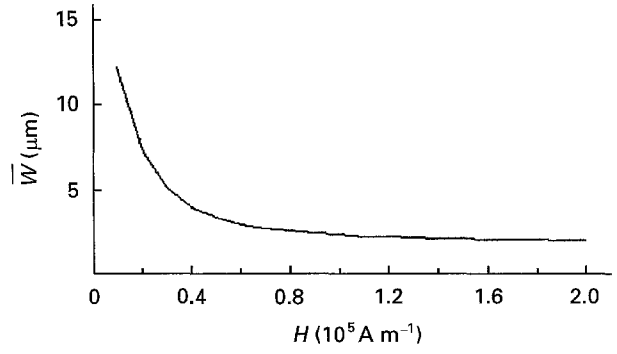


Figure 6 Magnetic field dependence of chain formation in a ferrofluid under the condition  $C = 0.02$ .

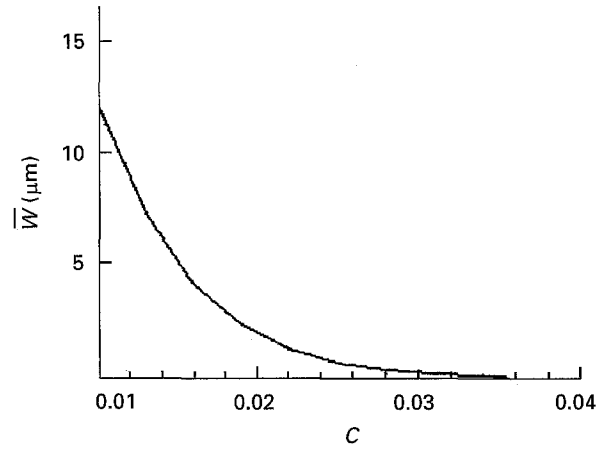


Figure 7 Particle concentration dependence of chain formation in a ferrofluid under the condition  $H = 2.0 \times 10^5 \text{ A m}^{-1}$ .

and

$$\frac{I_\theta}{I_{\theta=0}} = \frac{(\epsilon_{11}^2 \sin^2 \beta \cos^2 \theta + \epsilon_{33}^2 \cos^2 \beta) \cos^2 \theta}{\epsilon_{11}^2 \sin^2 \beta + \epsilon_{33}^2 \cos^2 \beta} \times \frac{\int_{W_{\min}}^{W_{\max}} \{V^2 [(x - \sin x)^2 + (1 - \cos x)^2]/x^4\} f(W) dW}{\int_{W_{\min}}^{W_{\max}} V^2 0.25 f(W) dW} \quad (5b)$$

with

$$x = 2\pi \sin(\theta/2) W/\lambda \quad (6)$$

In Equations 5 and 6,  $\theta$  is the scattering angle,  $\lambda$  is the wavelength of the incident light,  $W$  is the width of one chain and  $V$  is its volume.  $\epsilon_{11}$ ,  $\epsilon_{22}$  and  $\epsilon_{33}$  are the three elements of the dielectric constant matrix of the chain.  $f(W)$  is the probability density of the distribution which the chain widths follows, that is

$$f(W) = \frac{1}{(2\pi)^{1/2} \sigma} \exp\left[-\frac{(W - \mu)^2}{2\sigma^2}\right] \quad (7)$$

where  $\mu$  and  $\sigma$  are the expected and the deviation of the normal distribution, respectively. Moreover, Equations 5 and 6 become general if the parameters  $\epsilon_{11}$ ,  $\epsilon_{22}$  and  $\epsilon_{33}$  are considered as a function of the magnetic field and the expected  $\mu$  is replaced by

Equation 4. According to the model of dynamic chain-forming in a ferrofluid with a magnetic field [7], the expressions of the three parameters  $\varepsilon_{11}$ ,  $\varepsilon_{22}$  and  $\varepsilon_{33}$  can be given by

$$\varepsilon_{11} = \varepsilon_{22} = \left(1 - \frac{La}{a}\right) \varepsilon_{\xi} + \frac{La}{a} \varepsilon_{\zeta} \quad (8a)$$

$$\varepsilon_{33} = 2 \varepsilon_{\xi} \frac{La}{a} + \left(1 - 2 \frac{La}{a}\right) \varepsilon_{\zeta} \quad (8b)$$

with

$$\varepsilon_k = \frac{\varepsilon_e \varepsilon_i}{\varepsilon_e (1 - n^{(k)}) + \varepsilon_i n^{(k)}} \quad k = \xi, \zeta \quad (8c)$$

where  $\varepsilon_e$  and  $\varepsilon_i$  are the dielectric constants of the carrier and the particles, respectively.  $n^{(\xi)}$  and  $n^{(\zeta)}$  are the electric depolarization factors of the chain in the  $\xi$  and  $\zeta$  axes, respectively.  $La$  is the Langevin function and  $a = (mH)/(KT)$ .

The experimental results in Fig. 1 can be explained as follows: because the expected width of the chain decreases with increasing applied magnetic field (shown in Fig. 6), according to the conclusions derived from the former work [5], we can expect that the light scattering from a ferrofluid becomes obvious as the expected width of the chain decreases. Therefore, the scattering coefficient in Fig. 1 increases gradually with magnetic field and tends to saturate at higher fields. The theoretical calculations made under the same conditions as those of the experiments are also shown in Fig. 1. It can be seen that both curves agree well.

The influence of the particle concentration on the light scattering can be explained theoretically in terms of the decrease in the expected width of the chain as the particle concentration of the ferrofluid increases, which is shown in Fig. 7. Thus, the scattering coefficient can be expected to increase with increasing particle concentration, according to the conclusions given elsewhere [5]. Fig. 5 gives the theoretical curve of the relationship between the scattering coefficient and the

particle concentration under the same conditions as those of the experiments. From Fig. 5 we can see that the experimental curve contradicts this.

## 5. Conclusion

Our measurements of the light scattering from a ferrofluid show that the scattering coefficient increases with increase in both the magnetic field and the particle concentration and tends to saturate at higher magnetic fields. The experimental results can be explained in terms of an expanded theory of light scattering, which suggests that the expected width of the chain decreases both with magnetic field and particle concentration, and therefore the light scattering from the ferrofluid becomes obvious with them. The experimental results are in good accord with the theoretical analysis. For future work, we intend to study other influences on the light scattering from a ferrofluid.

## References

1. W. E. L. HASS and J. E. ADAMS, *Appl. Phys. Lett.* **27** (1975) 571.
2. C. F. HAYES, *J. Coll. Interface. Sci.* **52** (1975) 239.
3. A. TASKER, R. W. CHANTRELL, J. J. MILES, M. R. PARKER and A. BRADBURY, *IEEE Trans. Magn.* **24** (1988) 1671.
4. A. A. ROUSAN, N. A. YUSUF and H. M. EI-GHANEM, *ibid.* **18** (1988) 1653.
5. ZHANG QI, WANG JIANHUA and ZHU HESUN, *J. Coll. Interface Sci.* **172** (1995) 155.
6. H. ABU-SAFIA, I. ABU-ALJARAYESH, H. M. EI-GHANEM and N. A. YUSUF, *J. Magn. Magn. Mater.* **103** (1992) 19.
7. ZHANG QI, WANG JIANHUA and ZHU HESUN, in "Proceedings of the '94 Chinese Symposium on Materials '94C-MRS", Part 2 edited by Hengde Li, Beijing, China (The Chinese Material Society, 1994) p. 518.

Received 22 June 1995

and accepted 24 April 1996