Investigation on configuration of self-assembly in magnetic fluid film under a magnetic field

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We experimentally study transmittances of water-based magnetic fluid (MF) film and the behaviors of nanoparticles under an external magnetic field. The transmittances of two orthogonally polarized rays exhibit different curve shapes. The experimental data are used to determine the values of three important parameters that are related to the intensity of the field. Based on the relationship between these parameters and the field, the self-assembly of particles is discussed. The method introduced in this paper can be employed as an available means to inspect the behavior of the particles in MF.
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Since magnetic-optical effects of magnetic colloidal solutions were found, the magnetic-optical properties of magnetic fluid (MF) have been studied for a long time.[1–4] Up to now, the studies on the optical absorption and transmittance are mainly concerned in the situation in which the emitted rays are parallel to the external magnetic field.[5–7] The theoretical investigations and the experimental reports on transmittance when rays are perpendicular to the field, however, are rare.[8–10]

The chain forming of particles in MF film could be considered as the configuration of self-assembly under an applied magnetic field. The process is rather complicated and it is affected by many factors.[4] Probing into the self-assembly in MF is necessary in order to grasp the nature of this process and approach to achieving progresses in applications since all the magneto-optical effects origin from the self-assembly of the nanoparticles. Usually the chain forming can be observed directly with the measuring devices such as microscope. However, precise results may not be provided by the method because the chains are often difficult to be identified due to their overlap, especially in weak fields. The focus of this paper is on the research of chain forming by measuring the polarized light transmittances.

In our experiment, the sample is water-based Fe_3O_4 with the volume concentration of 4.8%. The average diameter of the particles is 10 nm. A 4 mW He–Ne laser with a wavelength of 632.8 nm and a 0.34 mW neodymium-doped yttrium aluminum garnet laser with a wavelength of 1064 nm are used as light sources. A polarizer is set between the laser and sample to acquire the polarized light. The rays whose electric displacement vector is perpendicular/parallel to the magnetic field are defined as ordinary/extraordinary rays. Samples of 5.6-μm-thick and 7-μm-thick films in glass cells are used in the experiment. The surface of the MF films is perpendicular to the incident beam and parallel to the external magnetic field.

Figure 1 shows the measured transmittances that are corresponding to the wavelength of 632.8 nm. For the 1064 nm wavelength, the curves have similar shapes but higher values. It is seen that the transmittances of the rays reduce with

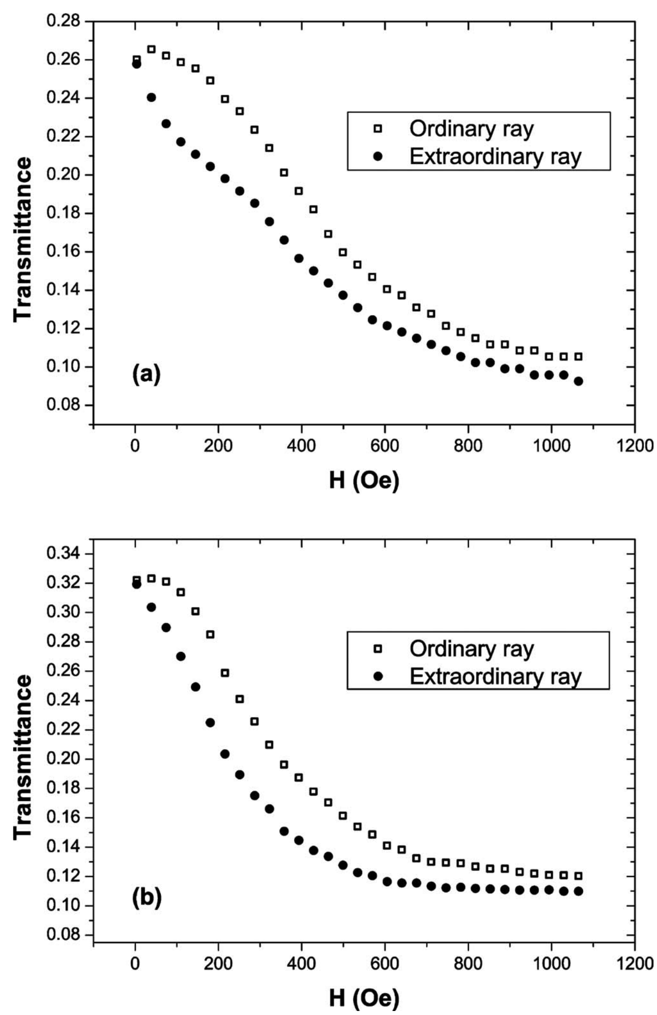

FIG. 1. Optical transmittances of the MF as a function of magnetic field. The film thickness and the laser wavelength are (a) 7 μm, 632.8 nm and (b) 5.6 μm, 632.8 nm.

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the enhancement of the magnetic field, and tend to the minimum. In these cases the ordinary rays increase slightly under the weak fields and then, with the increasing intensity of the magnetic field, turn to decrease after reaching a maximum. Each of their curvatures has an inflection point. The same phenomenon cannot be found in the extraordinary rays.

When the laser beam passes through MF, its attenuation is caused by several reasons including the Rayleigh scattering and the absorption. Both of them are related to the shape of the self-assembly chains. According to the studies by Taketomi *et al.*,[8] the transmittances of ordinary and extraordinary rays can be expressed as

$$\mathrm{Tr}_{\perp} = \exp\left\{-2\pi\left(\frac{d}{\lambda}\right)\phi_M\left[\frac{4(4\pi\sigma/\omega)}{[(\varepsilon+1)-\langle N\rangle(\varepsilon-1)]^2} + \frac{C_2}{2}\left(\frac{\langle a\rangle}{\delta}\right)^4\right]\right\}, \quad (1)$$

$$\mathrm{Tr}_{\parallel} = \exp\left\{-2\pi\left(\frac{d}{\lambda}\right)\phi_M\left[\frac{(4\pi\sigma/\omega)}{[1+\langle N\rangle(\varepsilon-1)]^2} + C_2\left(\frac{\langle a\rangle}{\delta}\right)^4\right]\right\}, \quad (2)$$

where $\langle N\rangle$ is the average of the depolarization factor in the field direction and $\langle a\rangle$ represents the average radius of chains. They are the only two variables in the equations which depend on the external field H. The meanings of other parameters can be found in Ref. 8. In our experiment, $\phi_M = 4.8\%$, $\varepsilon_1=80$, and $\varepsilon_2=4.5$. The value of $4\pi\sigma/\omega$ is 0.1993 and $\varepsilon=\varepsilon_2/\varepsilon_1=0.056\,25$.

Taketomi *et al.* thought that $\langle a\rangle$ can be neglected for alkylnaphthalene-based or alkylbenzene-based MF. When the sample of water-based Fe_3O_4 MF is concerned, this item should be considered. This is proved by our experimental results which show the transmittance of the ordinary ray increases initially before the peak appears, and then decreases along with the enhancement of the field.

As we know, the average depolarization factor $\langle N\rangle$ is related to the shape of the self-assembly chains. In other words, the value of the factor depends on the axis ratio of the chains. Here the axis ratio refers to the ratio of the length to the width. Without external field, the magnetic moments fixed in the particles are randomly oriented, thus $\langle N\rangle$ is 1/3. Affected by an enhancing field, the particles aggregate together gradually and form chains. Then $\langle N\rangle$ decreases with the increase in the axis ratio of the chains in the field direction. It is believed that the average radius of chains $\langle a\rangle$ grows with the field.[8] However, if the radius means the radius of a single chain, the trend conflicts with some observations which show that with the increase of H and chain number, the width of chains decreases.[4] Actually, it is often not necessary to know the behavior of a single chain because the average effect plays a more important role. We define $\langle r\rangle^4 = C_2(\langle a\rangle/\delta)^4$ and $\langle r\rangle$ here reflects the area ratio occupied by the chains in a unit area of the MF film which is perpendicular to the incident light. Assume the number of the chains in a unit area remains a constant. It is reasonable that $\langle r\rangle$ is regarded as the average equivalent radius of chains. Thereupon, these two variables above can be employed to analyze the self-assembly chain-forming circumstance by using the measured data of light transmittances.

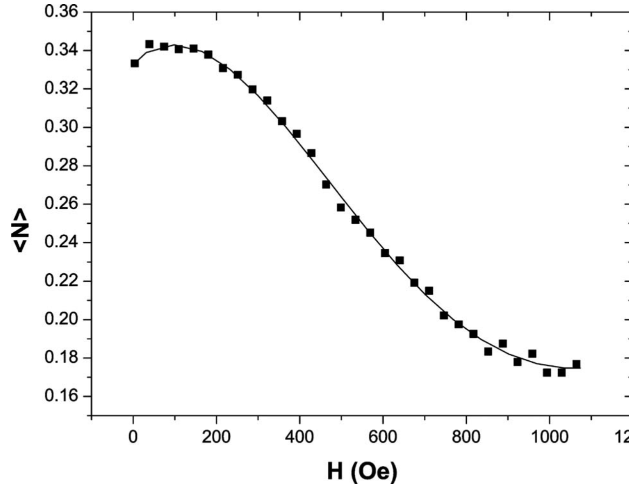

FIG. 2. Average depolarization factor in the field direction $\langle N\rangle$ as a function of magnetic field.

Two sets of the measured transmittances about ordinary and extraordinary rays correspond to a given set of fields. Therefore, based on Eqs. (1) and (2), we can uniquely determine the values of $\langle N\rangle$ and $\langle r\rangle$ by substituting the measured values about transmittances into the equations. Thus, the relationships between the two variables and field intensity can be shown clearly.

Using three sets of data measured in the same condition as shown in Fig. 1(a), we calculate the values of $\langle N\rangle$ and $\langle r\rangle$. The averaged results are shown in Figs. 2 and 3. In Fig. 2 the curve of $\langle N\rangle$ exhibits a trivial increase in the weak fields (about 0–200 Oe). When the intensity of the field exceeds 200 Oe, $\langle N\rangle$ decreases gradually along with the growing of chains, and approaches to a minimum about 0.18 eventually.

The variation of $\langle r\rangle$ as a function of H is shown in Fig. 3. When $H=0$, $\langle r\rangle$ is close to zero. It increases quickly in the weak fields (under 200 Oe) and trends to saturation finally.

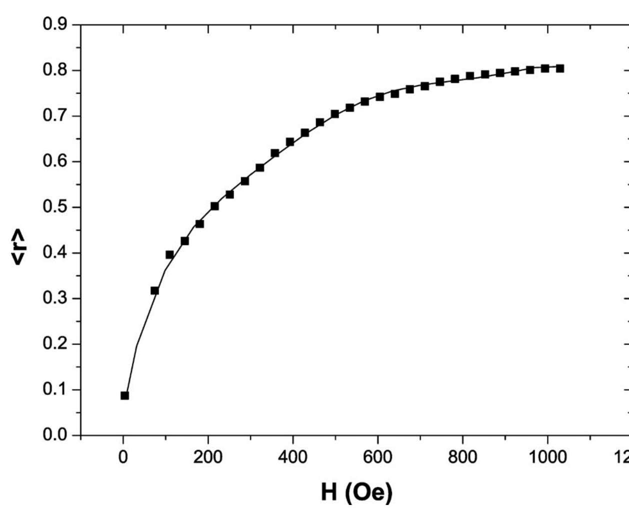

FIG. 3. Average equivalent radius of chains $\langle r\rangle$ as a function of magnetic field.

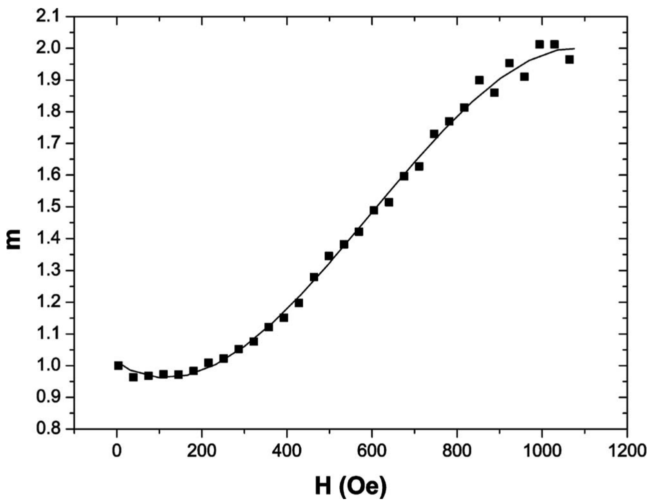

FIG. 4. Equivalent axis ratio of chains as a function of magnetic field.

For different wavelengths and film thicknesses, the similar variation trends of $\langle N \rangle$ and $\langle r \rangle$ are obtained.

According to Ref. 11 and the values about $\langle N \rangle$ calculated by the authors, the average axis ratio m of the chains can be obtained further, as shown in Fig. 4. We call it here the equivalent axis ratio because the values reflect the total effect of the whole MF. m decreases slightly to a value (<1) in the weak fields, but turns to increase rapidly when the magnetic field is larger than 200 Oe, which means the chains are forming and becoming more slender. When the magnetic field is large enough $(>1000$ Oe$)$, the value of m becomes saturated.

As we see in Fig. 2, $\langle N \rangle$ has a bit initial increase when the field is less than 200 Oe. From the experimental and calculated results, we speculate that two kinds of movements may take place after the magnetic particles are put in the magnetic field. The magnetic moment frozen in the particle will rotate to the direction of the field and aggregate with the nearby ones. In the weak fields, a small number of the particles congregate first, but there is still no enough magnetic force to pull them into chains. These particles can be seen as a cluster. Besides, the dimension of clusters in the horizontal direction (perpendicular to $\boldsymbol{H}$) increases quickly in the weak fields, which is displayed clearly in Fig. 3. This makes it possible for the transversal dimension of clusters exceeds the longitudinal dimension (parallel to $\boldsymbol{H}$), then $\langle N \rangle$ will increase from 1/3. This result shows the equivalent axis ratio of chain is less than 1 when the magnetic filed is less than 200 Oe (see Fig. 4).

Furthermore, our experiment also exhibits that wavelength of laser and thickness of film are almost without contribution in the trend of the transmittance versus magnetic field curves although they have influences on the values of transmittance. More experiments are needed to illustrate if this result is suitable for other conditions.

In conclusion, On the basis of the measured transmittances about polarized rays, the variation processes of the average depolarization factor $\langle N \rangle$, the average equivalent radius of chains $\langle r \rangle$ and the equivalent axis ratio of chains m are acquired. This method can be used as an indirect means to inspect the behavior of the particles in MF. Under an applied magnetic field, the magnetic moments of the individual particles tend to rotate to the field and assemble into chains. The curves of these parameters indicate that the assembly process relies on the intensity of the field and may be divided into two stages. In the weak field, rotation and aggregation are the main behaviors of the particles, and the chain-forming behavior becomes obvious under the influence of the strong field. It should be note that the formulas (1) and (2) are derived from a simplified model which only considering a single chain. As the concentration increases, the real chain-forming process may gradually become more complex.

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