Magnetic-field-induced birefringence and particle agglomeration in magnetic fluids

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Birefringent effect dependent on magnetic intensity and wavelength in a water-based Fe_3O_4 magnetic fluid over the wavelength range of 400–700 nm is studied in this letter. The results are compared with the theory proposed by Llewellyn [J. Phys. D **16**, 95 (1983)], in which the optical anistropic properties of the magnetic fluid due to the aggregation of the particles at a relatively low density is suggested. It is shown that the degree of aggregation corresponds to the value of the axial ratio of the aggregated particles. So does the magnitude of the birefringence. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2392824]

Magnetic fluid (MF) is a colloidal suspension of single domain ferromagnetic particles in a suitable carrier liquid. It has attracted a great deal of attention from researchers because of its remarkable optical properties, such as birefringence,[1–5] magnetochromatics,[6,7] and optical transmittance.[8–11] MF exhibits strong optical birefringence under a magnetic field,[12] and the birefringence is believed to be the result of spatial anisotropy caused by the alignment of the particles under a magnetic field.[13] Some experimental results have revealed that the ferrite particles agglomerate to form short chains as a magnetic field is applied on the sample film.[14,15]

Although extensive experimental investigations of magneto-optical effects in MFs have been presented, the studies of the wavelength dependences of these effects are few.[16] Further work should be done to clarify the origin of these effects and stimulate research on their applications in the fields of biology and biotechnology.[17] The theme of this letter is to demonstrate a method of investigating the magnetic-field-induced birefringence and particle agglomeration in magnetic fluids. The method we propose here is to use a theory model[18] to compare our experiment results with the calculation curves. The experimental setup and calculation results will be shown and discussed.

Figure 1 shows the schematic diagram of the experimental setup for measuring the birefringence effect. The propagation direction of the light is normal to the applied magnetic field. The monochromator was used to select the various wavelengths in the range of 400–700 nm. The samples investigated were water-based MFs with an average diameter of 10 nm, and the volume concentrations are 3%, 1.5%, and 0.75% separately. At a given magnetic field and a selected wavelength for a given sample, the transmittance of light was investigated by rotating the analyzer and determining the maximum and minimum transmitted intensities $I_{\max}$ and $I_{\min}$, respectively. The value of birefringence Δn is determined to be[19]

$$\Delta n = \sin^{-1}\frac{2\sqrt{I_{\min}/I_{\max}}\,ch(h_1-h_2)}{1+I_{\min}/I_{\max}} \times \lambda/(2\pi d),$$

where d equals 7 μm and $h_i(i=1,2)$ is the absorption coefficient along two directions which can be obtained by solving the equation $I_i=I_{0i}e^{-2h_i(H)}$ (I_{0i} is the intensity of the output in zero field).

It is known that an ordered array of elongated particles of refractive index n_p in a liquid of refractive index n_l will exhibit a birefringence effect, if n_p is not equal to n_l.[20] Llewllyn[18] obtained the expression of the birefringence

$$\Delta n = n(L_\alpha) - n(L_\beta),$$

where L_α and L_β are depolarizing coefficients corresponding to light polarized parallel and perpendicular to the symmetry axis of the particles, respectively. L_α and L_β have been tabulated by Stoner,[21] who got the relationship $L_\alpha+2L_\beta=1$, and L_α is inversely proportional to m, the axial ratio of the aggregated particles. Based on Llewllyn's theory model, the values of Δn as a function of wavelength for water-based Fe_3O_4 MF can be calculated. For a magnetic fluid, the optical transmission properties of Fe_3O_4 play an important role in the magneto-optical effects.[22–24] Figure 2 shows the simulation results. The results strongly suggest that the values of Δn are quite sensitive to the values of L_α, and a small change of L_α can result in a quite different value of Δn.

Figure 3 shows the experiment results of the birefringence Δn as a function of magnetic field at different wavelengths for a sample with a concentration of 1.5%. It is

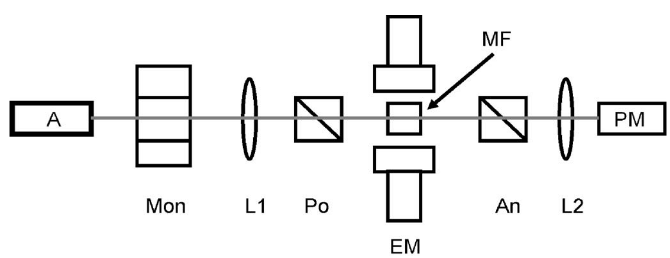

FIG. 1. Schematic diagram of the experimental setup: A, white light source; Mon, monochromator; L1 and L2, lenses; Po, polarizer; MF, water-based Fe_3O_4 magnetic fluid; EM, electromagnet; An, analyzer; and PM, power meter.

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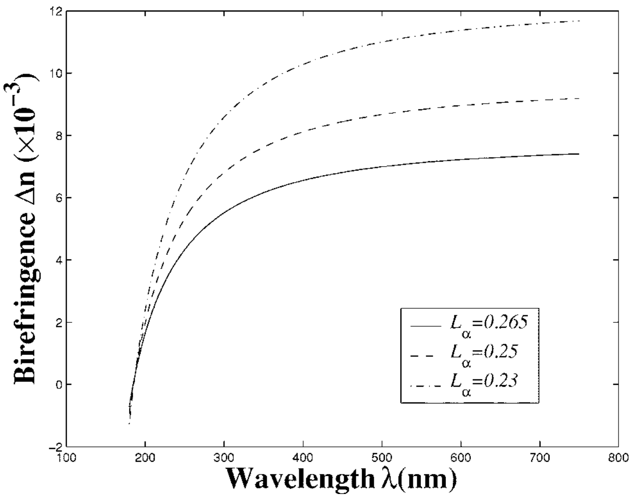

FIG. 2. Simulation results of birefringence (Δn) as a function of wavelength λ for water-based Fe_3O_4 MF. The curves shown were calculated with L_α=0.265, 0.25, and 0.23. The volume concentration is f=3%.

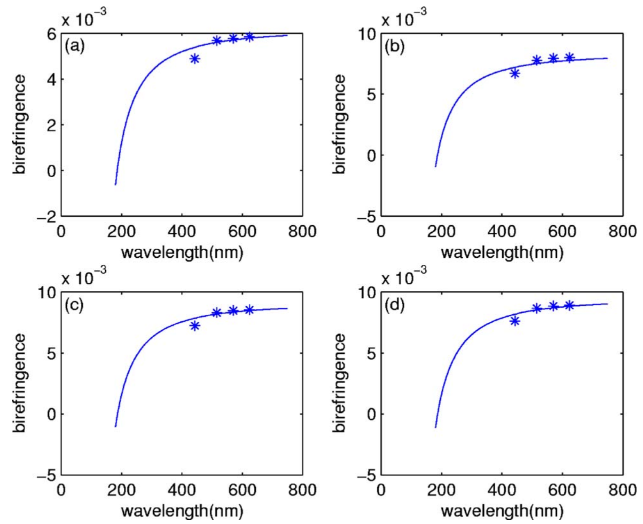

FIG. 4. Experimental and theoretical birefringence as a function of wavelength under four different magnetic fields: (a) 220 Oe, (b) 500 Oe, (c) 750 Oe, and (d) 960 Oe. Solid lines are calculated curves by choosing the value of depolarizing coefficient L_α: (a) 0.23, (b) 0.20, (c) 0.19, and (d) 0.185, respectively, and the volume concentration of the sample is 1.5%.

shown in Fig. 3 that the discrepancy of Δn between different wavelengths decreases with the increase of wavelength. There is a general agreement with the previous calculated results shown in Fig. 2.

It is well known that the MF's birefringence effect is due to the spatial anisotropy caused by the alignment of the magnetic chains within the MF under a magnetic field.[25,13] It is interesting to establish a quantitative relation between the magnetic chains and birefringence. The experimental data of the birefringence of MF as a function of wavelength at a certain magnetic field are employed to investigate this correlation. By choosing the specific values of L_α, the experimental data can be fitted with the above theory model. The results are depicted in Fig. 4. It is shown in Fig. 4 that the values of L_α are inversely proportional to the magnetic fields and Δn. These changes tend to saturate at high fields. This process is exactly expressed in Fig. 5. It is worth mentioning that L_α is direction dependent in nonspherical particles, corresponding to m (the axial ratio of the aggregated particles), and L_α is inversely proportional to m. Therefore, the decrease of L_α in Figs. 4(a)–4(d) means the increase of m, which indicates the presence of the particle agglomeration.

When the external magnetic field is applied, the magnetic energies of the magnetic particles increase within the MF. The larger the magnetic field strength, the larger the magnetic energies. After the field is applied over some critical value, the magnetic energies of the magnetic particles exceed their thermal energies, and the magnetic particles agglomerate to form chains and tend to saturate at high fields. Since the effect of birefringence of MF is induced by the alignment of the magnetic chains within the MF under a parallel magnetic field and the values of Δn also correspond to the changes of L_α, the information of agglomeration in MFs under the applied magnetic field can be acquired from the value of the measured birefringence.

The experimental results of Δn as a function of magnetic field at a wavelength of 632.8 nm for different concentrations are presented in Fig. 6. It is shown that, for a given concentration, Δn increases with applied fields and tends to saturate at high levels. Additionally, the higher the concentration, the higher the magnitude of Δn. As we know, higher concentration will result in more absorption. For practical

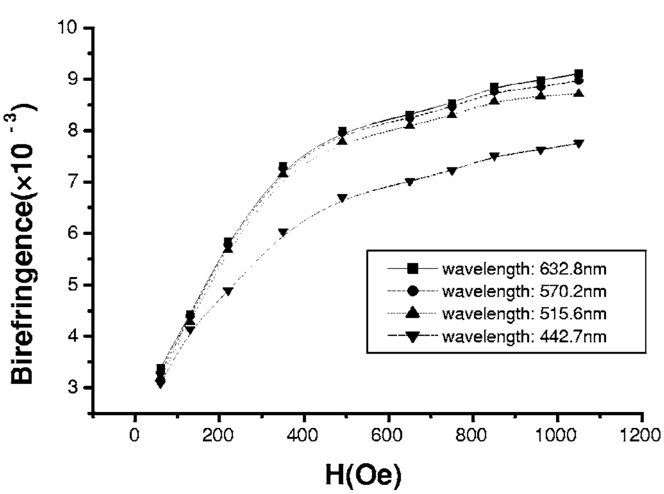

FIG. 3. Measured values of magnetic birefringence vs applied magnetic fields at four different wavelengths (632.8, 570.2, 515.6, and 442.7 nm) for water-based Fe_3O_4 MF, with a volume concentration of 1.5%.

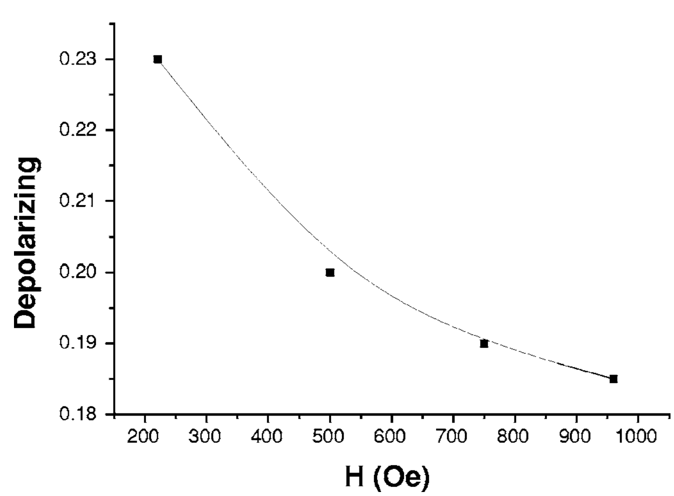

FIG. 5. Depolarizing coefficient L_α vs applied magnetic field. The data are from Fig. 4.

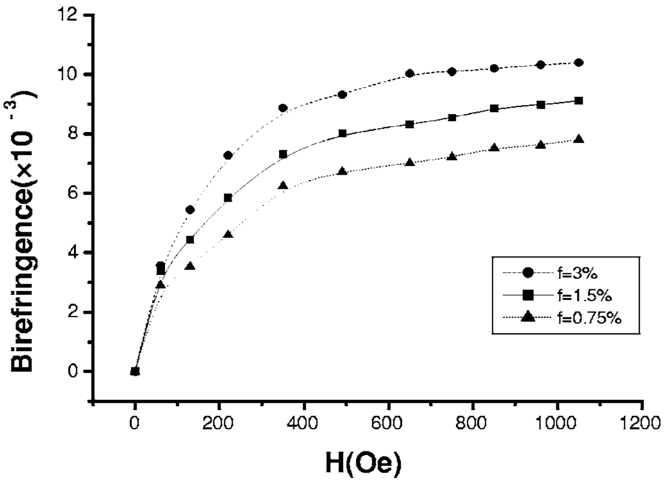

FIG. 6. Magnetic birefringence vs applied magnetic field for three water-based Fe_3O_4 MFs samples with different concentrations (3%, 1.5%, and 0.75%).

applications in photonics devices of MFs, the concentration should be carefully chosen to meet the special requirements.

Conclusively, the magnetodielectric anisotropy effect in MFs has been determined from magneto-optical measurements. The field, concentration, and wavelength dependence of the effect have been investigated. The role of the magnetic field in the agglomeration formation is investigated, which can be quantitatively expressed by the value of L_α and the magnitude of birefringence.

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