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Enhancement of the field modulation of light transmission through films of binary ferrofluids

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$CoFe_2O_4$ nanoparticles are ferrimagnetic and p-$MgFe_2O_4$ nanoparticles are paramagnetic. Binary ferrofluids can be synthesized by mixing $CoFe_2O_4$ ferrofluids and p-$MgFe_2O_4$ fluids in such a way that the magnetic interaction of the $CoFe_2O_4$ particles is large enough to form field-induced chainlike aggregates. The field modulation of light transmission through films of $CoFe_2O_4$–p-$MgFe_2O_4$ binary ferrofluids with different values of applied magnetic field is compared with pure $CoFe_2O_4$ ferrofluids. The experimental results revealed that the light transmission coefficient of binary ferrofluids can be more intensely modulated by an external magnetic field than pure $CoFe_2O_4$ ferrofluids. These show that in the binary ferrofluids, the field-induced structure mainly arises from the $CoFe_2O_4$ nanoparticle system and the p-$MgFe_2O_4$ nanoparticles introduce a nonlinear modulation effect, even though the microstructure of p-$MgFe_2O_4$ fluids is not affected by an applied magnetic field. Using a model of magnetic bidispersal, the enhanced field modulation of the light transmission through binary ferrofluids is explained by the coupling of geometric shadowing effects from both the $CoFe_2O_4$ and p-$MgFe_2O_4$ particle systems.

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I. INTRODUCTION

Ferrofluids generally consist of colloidal suspensions of single-domain ferromagnetic or ferrimagnetic nanoparticles (of size about 10 nm) dispersed in a carrier liquid. Arising from the pioneering work by De Gennes and Pincus in the 1970s about chain formations [1], it has been well known that when an external magnetic field is applied, the nanoparticles can form chainlike aggregates parallel to the field direction [2–5] producing a corresponding change to the properties of the ferrofluids. In addition, droplets of the concentrated phase inside the more dilute one can exist or appear when the ferrofluids are submitted to a magnetic field due to an enhancement of attractive interaction between the particles, and can deform, move and coalesce [6–9]. These deforming droplets behave as chains aggregates. The important physical properties of ferrofluids, for instance, the light transmission through a thin ferrofluid film, can be modified by an applied magnetic field. This field modulation effect has attracted the interest of many researchers because it not only has potential physical applications, but can also be used to probe the microstructure of samples of ferrofluids exposed to low magnetic fields (<1500 Gs) [10–19].

Polydispersity is a natural property of ferrofluids since the particles in real ferrofluids always possess a size distribution [20]. A theoretical model of a bidispersed ferrofluid containing "large" and "small" particles has been advanced [21–24]. This study indicated that the large particles constitute the main structure of the ferrofluids under an external magnetic field, and the small particles, depending on relative content, either suppress or enhance the formation and variation of the field-induced structure. However, such a bidispersed system is difficult to achieve experimentally. The magnetic moment m of single domain particles is proportional to their volume and magnetization. For a spherical particle, this can be expressed as

$$m = \pi a^3 M/6, \tag{1}$$

where M is the magnetization of the particle and a is its diameter. Therefore, a bisystem consisting of both large and small particles can also be regarded as a system consisting of stronger and weaker magnetic particles with different m, arising from different a [25]. Ferromagnetic or ferrimagnetic nanoparticles have intrinsic magnetic moments. So in formula (1) M is the saturation magnetization M_s [26]. Paramagnetic particles have induced magnetic moments. So, for paramagnetic particles, in formula (1) M expressed by χH, where χ is the susceptibility and H is the strength of the applied magnetic field [27]. Accordingly, the difference in magnetization (different M) between two pure systems can be used to synthesis a bidispersed system. That is, one can use a mixture of two pure systems with different magnetizations, such as ferrimagnetic and paramagnetic, to produce a magnetically bidispersed system instead of a size bidispersed system [28]. Fluids based on mixtures of two different magnetic nanoparticles are known as binary ferrofluids and can have different behavior compared with pure ferrofluids. In previous work, field modulation of light transmission through films of single Fe_3O_4 ferrofluids has been explored by continually switching on and off the magnetic field [17]. In this work, the response of magnetic field modulated light transmission through $CoFe_2O_4$–p-$MgFe_2O_4$ binary ferrofluids film has been investigated.

II. EXPERIMENT AND RESULTS

A. Sample description

The binary ferrofluids are synthesized by mixing acidic $CoFe_2O_4$ aqueous ferrofluids and p-$MgFe_2O_4$ aqueous fluids

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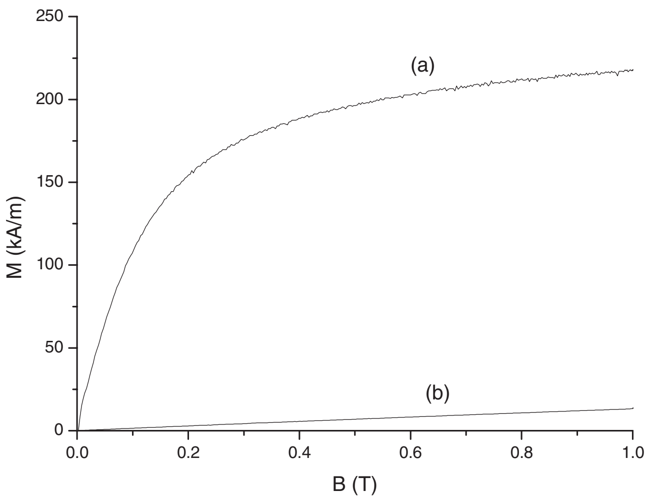

FIG. 1. The magnetization curves of (a) $CoFe_2O_4$ nanoparticles and (b) p-$MgFe_2O_4$ nanoparticles,

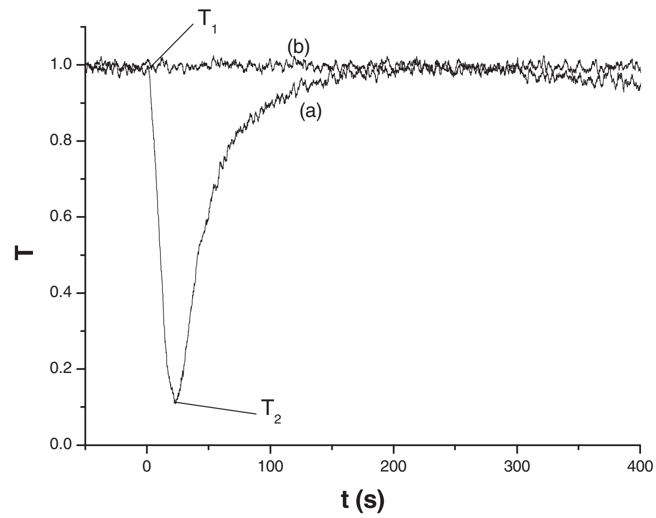

FIG. 2. T-t curves for (a) pure $CoFe_2O_4$ ferrofluids and (b) pure p-$MgFe_2O_4$ fluids with particle volume fraction 0.2% with 400 Gs applied magnetic field. The magnetic field is switched on at $t=0$. It should be noted that T_1 may be too weak to be measured, as discussed in Ref. [11].

prepared by the Massart method [29]. Their pH values were tested with pH monitor and are about 2.08. In the binary ferrofluids, total particle volume fraction is 0.2%, and the particle volume fraction of both the $CoFe_2O_4$ ferrofluid and p-$MgFe_2O_4$ fluids is 0.1%, respectively. The $CoFe_2O_4$ particles, and p-$MgFe_2O_4$ particles which is the hydroxide precursor to produce $MgFe_2O_4$ particles are prepared by coprecipitation. From electron micrographs, the particles size and standard deviation which describes the size distribution are obtained [30]. The $CoFe_2O_4$ particles have a median diameter of 12.76 nm and the standard deviation of 0.35, the p-$MgFe_2O_4$ particles have a median diameter of 5.58 nm and the standard deviation of 0.37. The magnetization curve of the particles are measured at room temperature using a vibrating sample magnetometer (VSM), as shown in Fig. 1. Obviously, the $CoFe_2O_4$ particles are ferromagnetic, whose saturation magnetization is estimated as about 220 kA/m according to the relation of $M \sim 1/B$ under high field, and the p-$MgFe_2O_4$ particles are paramagnetic whose magnetization at 1 T is 11.48 kA/m. The dipolar coupling constant λ is a important parameter to characterize the magnetic feature of a system and is defined as

$$\lambda = -\frac{u_{m-m,\max}}{k_B T} = \frac{\mu_0 m^2/2\pi d^3}{k_B T}, \qquad (2)$$

where $u_{m-m,\max}=-\mu_0 m^2/2\pi d^3$ is the maximum dipole-dipole interaction potential of two particles in contact with each other, μ_0 is the vacuum permeability, d is distance of the particles center, $k_B T$ is thermal energy, k_B is the Boltzmann constant and T is the absolute temperature [26]. The magnetic interaction between particles plays an important role in determining the properties of ferrofluids. When $\lambda > 2$, aggregation is stimulated, and for $\lambda < 2$ the aggregation cannot form [31]. From the magnetization measured, it can be known that two p-$MgFe_2O_4$ particles ($d=a=5.58$ nm), linked by contact, the great interaction energy ($B=1$ T) is about 4.78×10^{-24} J, the interaction energy of two $CoFe_2O_4$ particles ($d=a=12.76$ nm) is about 1.71×10^{-20} J, and while one $CoFe_2O_4$ particle, and one p-$MgFe_2O_4$ particle are linked by contact [d=(diameter of p-$MgFe_2O_4$ particles +diameter of $CoFe_2O_4$ particles)/2] the interaction energy ($B=1$ T) is about 2.76×10^{-22} J. At room temperature (T =300 K), the thermal energy $k_B T=4.14 \times 10^{-21}$ J. Thus, it can be given that $\lambda_{Mg}=1.15 \times 10^{-3}$, $\lambda_{Co}=4.13$ and λ_{Mg-Co} $=6.67 \times 10^{-2}$. So, the magnetic interaction between $CoFe_2O_4$ nanoparticles alone is large enough to form field-induced aggregates.

B. Magneto-optical experiment

The fluids were inject into a thick glass cell of 0.3 mm to form the fluids film and size of the cell is $l \times l = 15 \times 15$ mm^2. In the magneto-optical experiment, apart from replacement of the light source by a He-Ne laser of 10 mW power, the remaining experimental setup is as in Ref. [16] with the incident light parallel to the applied magnetic field and normal to the ferrofluid film. In experiment, the field was controlled by directly taking on/off electric current. The delay-time of the field switching on/off is less than 0.5 s and the relaxation time of T variation is order of 10 s, so the speed of the field switching on/off is fast enough to characterize to the field modulation effect of light transmission through films of ferrofluids.

Figure 2 shows the variation of the relative transmission coefficient T in the presence of a 400 Gs magnetic field for a pure $CoFe_2O_4$ ferrofluid and a pure p-$MgFe_2O_4$ fluid with particle volume fractions 0.2%. $T=I'/I$, $I(I')$ is the measured intensity of the transmitted light before (after) the field was turned on at $t=0$. It can be seen that the light transmission through the film of $CoFe_2O_4$ ferrofluid exhibited a field-induced relaxation behavior. This is normal for ferrofluids and is explained by the “geometric shadowing effect” in relation to chain motion [32]. However, the light transmission through the film of p-$MgFe_2O_4$ was not affected by the magnetic field. Experiments showed that light transmission

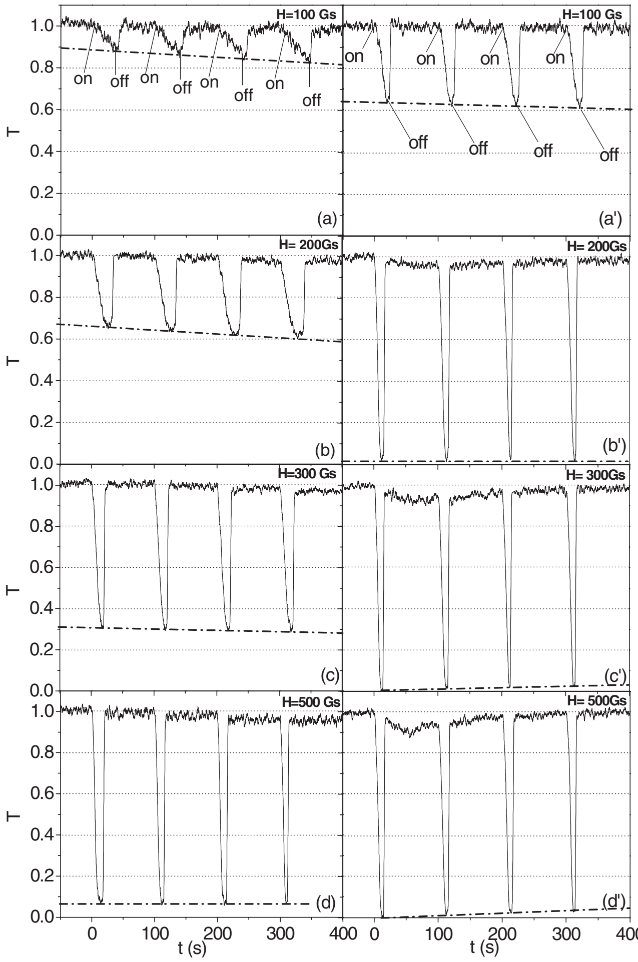

FIG. 3. The T sequence from $CoFe_2O_4$ ferrofluids [(a)-(b)] and $CoFe_2O_4$–p-$MgFe_2O_4$ binary ferrofluids [(a′)-(b′)] with different strengths of the applied magnetic field. The particle volume fraction of the $CoFe_2O_4$ ferrofluids is 0.2%. For the binary ferrofluids, total particle volume fraction is 0.2% and the particle volume fraction of both $CoFe_2O_4$ and p-$MgFe_2O_4$ particles is 0.1%, respectively.

through the binary ferrofluid film also have relaxation behavior similar to pure $CoFe_2O_4$ ferrofluids. To investigate the effect of p-$MgFe_2O_4$ on binary ferrofluids, the process from T_1 to T_2 was continuously repeated by switching on and off the magnetic field and successive T sequences were measured for films of both pure $CoFe_2O_4$ ferrofluid and the $CoFe_2O_4$–p-$MgFe_2O_4$ binary ferrofluid. The real time intensity of the transmitted light was monitored, and once it started to increase from its lowest value at T_2, the magnetic field was switched off immediately. And, after T increased and tended to stabilize, the magnetic field was taken on again. The variation with T for different values of the central field strength is given in Fig. 3.

III. RESULTS AND DISCUSSION

From Fig. 3, it can be known that under the same applied field, the minimum at T_2 of the binary ferrofluids is lower than the pure $CoFe_2O_4$ ferrofluid. This indicates that the field-induced variation of the light transmission through the binary ferrofluid film arises from the $CoFe_2O_4$ system, and the p-$MgFe_2O_4$ system can produce a enhancement effect. This can be explained as follows.

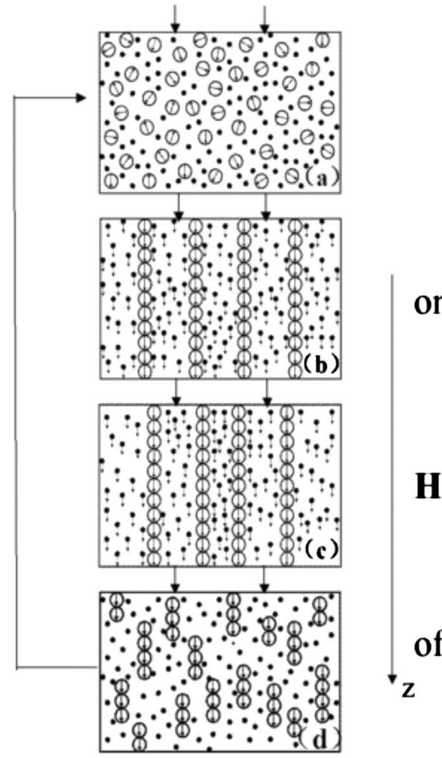

FIG. 4. Scheme of microstructure change in the binary ferrofluid film. Both applied magnetic field and optical path are along the z direction. ⊖, $CoFe_2O_4$ nanoparticles; ●, p-$MgFe_2O_4$ nanoparticles.

For the binary ferrofluids, the p-$MgFe_2O_4$ nanoparticle system is the same as that of the p-$NiFe_2O_4$ nanoparticle system [28], that is, weakly magnetic. The field-induced interaction between the p-$MgFe_2O_4$ particles as well as that between the p-$MgFe_2O_4$ and $CoFe_2O_4$ particles is not enough to form field-induced aggregates. Thus, the "geometric shadowing effect" resulting in variation of transmitted light for the binary ferrofluid system (the AB system) may be equivalent to the coupling of both the $CoFe_2O_4$ system (A system) and p-$MgFe_2O_4$ system (B system). Before applying the magnetic field, the particles were well dispersed in the carrier liquid, as shown in Fig. 4(a). The "geometric shadowing effect" should be proportional to the reciprocal of the intensity of the transmitted, therefore, the intensity relationship of the transmitted light between the binary system (I_{AB}) and the single systems (I_A, I_B) can be described as

$$\frac{1}{I_{AB}} \propto \left(\frac{1}{I_A} + \frac{1}{I_B}\right). \tag{3}$$

By applying a magnetic field perpendicular to a thin film of ferrofluids, the particle column structure can be formed in the film. For a given final field strength, sweep rate of the field (dH/dt) decides the duration of the formative process and size of the column lessens with the sweep rate increasing [33]. Thus, when a magnetic field with given strength is turned on instantaneously $(dH/dt \to \infty)$, the A particle system will form thin column or chainlike aggregates as Fig. 4 showed, and the transmitted light is modified to I'_A. For the B particle system, as in pure *p*-$MgFe_2O_4$ fluids, $I'_B = I_B$ (as shown in Fig. 1), but in the model of the binary system, it is possible that $I'_B \neq I_B$ resulting from a coupling effect with the A particle system. This is because with the application of the magnetic field, paramagnetic B particles will induce magnetic moments orienting along the A particle chains in the direction of the applied magnetic field and act as magnetically polarized gas molecules restrained in the space between the A particle chains [see Fig. 4(b)]. Following the motion of the A particle chains toward the field/light axis, a local variation of the particle number density in the B particle system will take place, as shown in Figs. 4(b) and 4(c). Therefore, the light transmitted through the B particle system will vary, following the response of the A particle system to the magnetic field. Thus, while the magnetic field is applied, the intensity of the transmitted light can be described as

$$\frac{1}{I'_{AB}} \propto \left(\frac{1}{I'_A} + \frac{1}{I'_B}\right). \quad (4)$$

From the definition of the relative transmission coefficient, that of the binary system T_{AB} can be written as

$$T_{AB} = \frac{I'_{AB}}{I_{AB}} = T_A T_B \left(\frac{1 + I_B/I_A}{T_A + T_B \cdot I_B/I_A}\right), \quad (5)$$

where, $T_A = I'_A/I_A$, $T_B = I'_B/I_B$ are the relative transmission coefficients of the A and B particle systems, respectively, in the binary system. Formula (5) shows that before the field is applied, $T_A = T_B = 1$, so $T_{AB} = 1$. From formula (5), it can be seen that

$$T_{AB} = F T_A, \quad (6)$$

where $F = (1 + I_B/I_A)/(T_A/T_B + I_B I_A)$ is termed the modulating factor. Since F is related to T_A, this is a nonlinear modulation effect. For certain samples of binary ferrofluids film, I_A and I_B can be regarded as constants. While the A particle chains converge in the direction of the field to make T_A decrease, the B particle system will be compressed, causing T_B to decrease. Both the A and B particle systems have the same particle volume fraction, so before applying the magnetic field, the geometric shadowing effect of the A particle system is regarded as the same as the B particle system. After applying the magnetic field, the A particle system forms an ordered chainlike structure parallel to the light axis and the B particles, behaving as a gaslike molecular system, are compressed, so that the geometric shadowing effect of the A particle system is weaker than that of the B particle system. Therefore, T_B decrease faster than T_A, i.e., $T_B/T_A < 1$ and $F < 1$. Thus, it can be seen from formula (6) that $T_{AB} < T_A$. So, the minimum T of the $CoFe_2O_4$–*p*-$MgFe_2O_4$ binary ferrofluids is lower than that of the pure $CoFe_2O_4$ ferrofluids. I.e., the field modulation of light transmission through the $CoFe_2O_4$–*p*-$MgFe_2O_4$ binary ferrofluids is enhanced in comparison with the pure $CoFe_2O_4$ ferrofluids, as observed experimentally.

After the field was switched off, the $CoFe_2O_4$ particle chains diverged rapidly and broke up into short chains. The compressed *p*-$MgFe_2O_4$ particle system also spreads, as shown in Fig. 4(d). Thus, T_A and T_B as well as T_{AB} increased to a stable level which corresponded to the microstructure of binary ferrofluids in an equilibrium state.

It was noticed that for single $CoFe_2O_4$ ferrofluids, the minimum value of the light transmission coefficient decreased gradually with the time spent switching on and off the field and this was more obvious for low field than high field. For binary ferrofluids, the minimum value decreased gradually for a 100 Gs field, was unchaining for a 200 Gs field, and increased gradually for a 300 and 500 Gs field. Also, Fig. 3 shows that the stable value of the light transmission coefficient can be less than the initial value (≈ 1) for both pure $CoFe_2O_4$ ferrofluids and the binary ferrofluids. In addition, the stable value for the single $CoFe_2O_4$ ferrofluids decreased and for binary ferrofluids increased with the time spent switching on and off the field, a feature which is more obvious for the higher value of the applied magnetic field, as shown in Fig. 3. These results can be explained as follows:

After the magnetic field was taken off, the remanence of the magnet enhanced slightly with the times of switching and the broken chains would increase a little [17]. Thus, for single $CoFe_2O_4$ ferrofluids, when the magnetic field was applied again, the average length of the formed chains would be longer. These chains were affected by an interaction force along radial direction as

$$F_r = -M \left|\frac{\partial B}{\partial r}\right|, \quad (7)$$

where M is the magnetic moment of a chain which is directly proportional to the chain's length, and $\partial B/\partial r (< 0)$ is the gradient of the field. So, with times of taking on the field, the effect of the chains converging enhanced, and the value of the minima decreased. And, with the magnetic field increasing, the affection of the remanence weakened, so that the difference of the minimum values lessened in T sequences. In addition, some $CoFe_2O_4$ ferrofluids droplets, whose geometric shadowing effect is stronger than the ferrofluids particles since it's size is larger than the particles, can formed under the magnetic field of the remanence. And, with the times of the field switching, the number of the droplets could increase since the remanence will enhance slightly [17]. So, for the single ferrofluids, the values of stabilize would lessen gradually in T sequences.

For $CoFe_2O_4$–*p*-$MgFe_2O_4$ binary ferrofluids, *p*-$MgFe_2O_4$ particle system could suppress the formation of $CoFe_2O_4$ droplets, so that the remanence effect was very weak and can be neglected in the high field. Besides, after the field was taken off, the spreading process of *p*-$MgFe_2O_4$ particle gas could be faster than the $CoFe_2O_4$ particle system because the size of the *p*-$MgFe_2O_4$ particle is far less than the size of the $CoFe_2O_4$ short chains. Therefore, with the times of the field switching, the number density of *p*-$MgFe_2O_4$ particle would increase along the radial direction, so that strength of transmitted light in the spot of the light did so also, i.e., T_B increased correspondingly. From formula (6), it can be known that F will become large (still less than 1). Thus, both values of the minimum and of the stabilize increased all with the times of the field switching, and this behavior was more obvious with the field increasing for the binary ferrofluids.

In summary, these results indicate that for the $CoFe_2O_4$–p-$MgFe_2O_4$ binary ferrofluids, although the field-induced structure mainly arises from the $CoFe_2O_4$ nanoparticle system, the structure corresponding to the minimum value and the new stable value of the light transmission coefficient can be modulated by the p-$MgFe_2O_4$ nanoparticle system.

IV. CONCLUSIONS

For the binary ferrofluids based on ferrimagnetic $CoFe_2O_4$ nanoparticles and p-$MgFe_2O_4$ paramagnetic nanoparticles, only the magnetic interaction between $CoFe_2O_4$ particles is large enough to form chainlike structure. Experiments shows that under magnetic field applying, the field modulation effect of light transmission through the binary ferrofluids film is similar to one through pure $CoFe_2O_4$ ferrofluids, but the effect can be enhanced in the binary ferrofluids as compared to the $CoFe_2O_4$ ferrofluids. The enhancement is attributed to the behaviors of field-induced microstructure of the binary ferrofluids, in which the field-induced structure mainly arises from the $CoFe_2O_4$ nanoparticle system and the p-$MgFe_2O_4$ nanoparticle system induces a nonlinear modulation effection though pure p-$MgFe_2O_4$ fluids cannot form field-induced aggregates. The modulation of the field-induced microstructure results in the field modulation of light transmission through the binary enhancing by the coupling of geometric shadowing effects from both the $CoFe_2O_4$ and p-$MgFe_2O_4$ nanoparticle systems. Obviously, these results mean that for binary ferrofluids, other behaviors could have similar modulation effect since the macroscopical behaviors of the matter arise from the features of the microstructure.

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