Response of colloidal liquids containing magnetic holes of different volume densities to magnetic field characterized by transmission measurement*

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This paper systematically investigates the response of colloidal liquids containing magnetic holes of different volume densities to magnetic field by conventional transmission measurements. It finds that the enhancement in the transmission of such a colloidal liquid under a magnetic field exhibits a strong dependence on the volume density of magnetic holes. A linear increase in the maximum enhancement factor is observed when the volume density of magnetic holes is below a critical level at which a maximum enhancement factor of ∼150 is achieved in the near infrared region. Once the volume density of magnetic holes exceeds the critical level, a sharp drop of the maximum enhancement factor to ∼2 is observed. After that, the maximum enhancement factor increases gradually till a large volume density of ∼9%. By monitoring the arrangement of magnetic holes under a magnetic field, it reveals that the colloidal liquids can be classified into three different phases, i.e., the gas-like, liquid-like and solid-like phases, depending on the volume density of magnetic holes. The response behaviour of colloidal liquids to magnetic field is determined by the interaction between magnetic holes which is governed mainly by their volume density. A phase transition, which is manifested in the dramatic reduction in the maximum enhancement factor, is clearly observed between the liquid-like and solid-like phases. The optical switching operations for colloidal liquids in different phases are compared and the underlying physical mechanisms are discussed.

Keywords: magnetic holes, phases change, optical switching operations

PACC: 7550M, 7865M, 4550M

1. Introduction

Magnetic fluids or ferrofluids, which are formed by magnetic nanoparticles uniformly dispersed in a liquid carrier, have attracted great interest in the past decade because the formation of magnetic chains or clusters under an external magnetic field gives rise to many interesting phenomena such as birefringence,[1−3] modulation of optical transmission,[4−9] magnetochromatics,[10−12] etc. These phenomena can be employed to realize some important magneto–optical devices such as optical switches,[13−15] tunable gratings,[16,17] magnetic field sensors,[18−20] etc. In comparison, nonmagnetic particles of micrometer size immersed in a magnetic fluid, which are generally referred to as magnetic holes (MHs), have received less intensive studies. The first investigation on the self-assembly of MHs under a magnetic field was carried out more than twenty years ago by Skjeltorp.[21] Based on the arrangement of MHs under a magnetic field, he indicated that three phases (i.e., gas-like, liquid-like and solid-like phases) may exist in such colloidal liquids, depending on the vol-

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ume density of MHs.[21] A physical model concerning the interaction between MHs was presented to explain the arrangement of MHs under a magnetic field.[22,23] However, the images obtained by microscope observations provide only the information on the local ordering of MHs induced by the magnetic field. From the viewpoint of fundamental research and device application, it is necessary to know the ordering of MHs in a large volume and its dependence on magnetic field strength. In addition, it is interesting to find out the response behaviour of such colloidal liquids in different phases to the magnetic field.

Modification of the optical transmission through magnetic fluids in the presence of magnetic field has been extensively studied because of its potential applications in the fabrication of magneto–optical devices such as optical switches.[12−14] Due to the formation of large size magnetic clusters, the incident light suffers from a stronger scattering which leads to a reduction in its transmission.[24−26] In comparison, a magnetic fluid doped with nonmagnetic particles or MHs responds to a magnetic field in the form of self-organization of MHs through the interaction between the induced magnetic moments. The MHs are randomly distributed in the liquid which begin to form dimmers, chains, and even crystallites in response to the magnetic field.[21] In other words, the ordering of the colloidal liquid is improved, more or less, upon the application of the magnetic field. It is well known that conventional transmission measurements act as a simple and effective way to characterize the ordering of a scattering system. Since the redistribution of MHs in the presence of a magnetic field is accompanied by an enhancement in the transmission through the colloidal liquid, the evaluation of the transmission enhancement can be used to examine the ordering of MHs from which the information on the interaction between them may be extracted. It is also expected that the enhancement in transmission for such a colloidal liquid induced by a magnetic field can also be utilized to construct a magneto–optical switch.

In this article, we present a systematic study on the response of colloidal liquids containing MHs of different volume densities to magnetic field by making use of conventional transmission measurements. It is organized as follows. In Section 2, we describe details of sample preparation and experimental setup. Then, the response of colloidal liquids containing MHs of different sizes to magnetic field is compared in Section 3. After that, we examine the dependence of the response behaviour of colloidal liquids on the volume density of MHs in Section 4. The three different phases observed in colloidal liquids with different volume densities of MHs are investigated in detail in Section 5. The switching behaviour of several typical colloidal liquids is studied in Section 6 and the physical mechanism responsible for the response behaviour is discussed in Section 7. Finally, a summary of our research work is given in the conclusion.

2. Sample preparation and experimental details

The colloidal liquids used in the experiments were obtained by mixing a water-based Fe_3O_4 magnetic fluid fabricated by the chemical co-precipitation technique with the aqueous solutions of polystyrene (PS) spheres (Duke Scientific) in different volume ratios. The diameter and volume fraction of magnetic nanoparticles in the water-based magnetic fluid are ∼12 nm and 7.9%. Monodisperse PS spheres with three different diameters (1.9, 4.3 and 11 μm) were used and their volume fractions in the aqueous solutions are 10%. In order to study different phases in colloidal liquids, colloidal liquids with different mixing ratios were prepared and the volume densities of PS spheres (or MHs) in the resulted colloidal liquids are given in Table 1. Finally, the colloidal liquids were sealed into rectangular glass cells with a thickness of 150 μm for transmission measurements. During the measurements, a homogonous magnetic field was applied perpendicularly on the samples. The magnetic field was provided by an electromagnet whose strength can be controlled by adjusting the current. A collimated incandescent light from a halogen lamp was incident normally on the samples and the spectrum of the transmitted light was detected by a spectrometer equipped with an InAs detector. In addition, the distribution and arrangement of MHs in colloidal liquids under different magnetic fields parallel to the samples were monitored and recorded by making use of an inverted microscope (Zeiss Axion Observer A1) and the attached charge coupled device (CCD). In this case, the magnetic field applied on the samples was supplied by a solenoid. In order to observe the enhanced transmission through a sample induced by a magnetic field, the 632.8-nm light from an He–Ne laser was incident on the sample and the transmitted light beam was recorded by using another CCD.

3. Response of colloidal liquids containing MHs of different sizes to magnetic field

Physically, the repulsive interaction energy between two MHs can be expressed as follows:[21]

$$E_{\rm rep} \propto MH^2V^2/r^3. \quad (1)$$

Here, M is the magnetization of the magnetic fluid, H is the strength of the magnetic field, V is the volume of the MHs and r is the separation between two MHs. It means that the interaction energy between MHs is proportional to the square of the volume of MHs. In other words, the larger the MHs the stronger the interaction between them. Therefore, it is expected that the strongest interaction should occur for 11-μm MHs. Unfortunately, a significant modification in the transmission spectrum for 11-μm MHs is expected to appear in infrared region which is beyond the wavelength range covered by the InAs detector. If we choose 1.9-μm MHs, the response of the colloidal liquid to magnetic field in this case may not be conspicuous because of the relatively weak interaction between MHs.

In order to seek suitable MHs which can render a significant change in the wavelength range covered by the InAs detector (i.e., in visible or near infrared region), we have measured the transmission spectra for colloidal liquids composed of MHs with different sizes and the results are compared in Fig. 1. The mixing ratio between the magnetic fluid and the aqueous solution of PS spheres is chosen to be 1:3. In order to see the enhancement in transmission induced by the applied magnetic field, the transmission spectra obtained at different magnetic fields have been normalized with respect to that obtained in the absence of magnetic field. In Fig. 1(a), a maximum enhancement factor of 4.5 is observed at 700 and 900 nm for 1.9-μm MHs. No obvious change is found in the transmission spectrum for magnetic fields higher than 450 Oe (1 Oe=80 A/m). Similar measurements for 11-μm MHs are shown in Fig. 1(c). In this case, a maximum enhancement factor of $\sim$2.8 is achieved at $\sim$500 nm under a magnetic field of 1980 Oe. Unlike the 1.9-μm case, a monotonic increase in the relative transmission is observed with increasing magnetic field.

Now let us examine the evolution of the transmission spectrum with increasing magnetic field for 4.3-μm MHs, as shown in Fig. 1(b). As expected, a much larger enhancement factor of $\sim$23 is observed at $\sim$1000 nm under a magnetic field of 360 Oe. In this case, the enhancement factor increases first with increasing magnetic field. When the magnetic field exceeds 360 Oe, it begins to decrease with a further increase of magnetic field. However, the spectral shape remains nearly unchanged. Anyway, this experiment indicates clearly that the colloidal liquids containing 4.3-μm MHs are suitable for investigating the enhancement of transmission induced by a magnetic field.

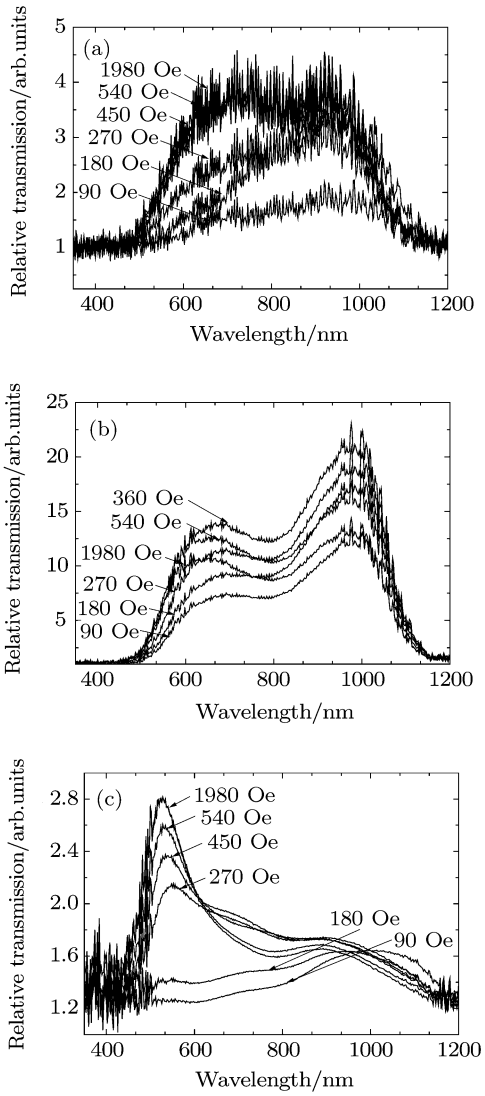

Fig. 1. Relative transmission spectra for colloidal liquids composed of magnetic holes with different sizes: (a) 1.9 μm; (b) 4.3 μm; and (c) 11 μm.

4. Response of colloidal liquids containing MHs of different volume densities to magnetic field

In order to find out the response behaviour of colloidal liquids with MHs of different volume densities to

magnetic field, we have prepared totally eight samples in which the mixing ratios and the volume densities of MHs ($\Phi_{\rm MH}$) are given in Table 1. For convenience, they are denoted in sequence as S_1 to S_8 with increasing $\Phi_{\rm MH}$. Then the transmission spectra for these samples were measured under different magnetic fields and the results are presented in Fig. 2. Similarly, all the spectra have been normalized with respect to that obtained in the absence of magnetic field and the enhancement factor in transmission is denoted as η.

Table 1. Mixing ratios and volume densities of magnetic holes in different samples.

sample	mixing ratio	$\Phi_{\rm MH}$/ml^{-1}
S_1	9:1	0.3×10^9
S_2	2:1	1.0×10^9
S_3	1:1	1.5×10^9
S_4	1:3	2.3×10^9
S_5	1:4	2.4×10^9
S_6	1:5	2.5×10^9
S_7	1:9	2.7×10^9
S_8	1:19	2.9×10^9

For sample S_1 in which $\Phi_{\rm MH}$ is quite small, we can see a slight enhancement of transmission to ∼1.5 at 1100 nm under a magnetic field of 90 Oe. When the magnetic field is raised to 270 Oe, the maximum enhancement factor is increased to ∼2.0. In this case, it is observed that an enhancement factor smaller than one (∼0.3) appears at ∼700 nm. A further increase in magnetic field results in a decrease of enhancement factor. When the magnetic field is increased to 810 Oe, the enhancement factors over the entire spectral range become smaller than one. It implies that the transmission of the sample is reduced upon the application of the magnetic field. This behaviour is quite similar to that of a pure magnetic fluid.[27]

The situation is dramatically changed when $\Phi_{\rm MH}$ is increased to 1.0×10^9 ml^{-1}, as manifested in sample S_2. From Fig. 2(b), it can be seen that the relative transmission increases significantly with increasing magnetic field. Maximum enhancement factors of ∼24 and ∼36 are observed at 90 and 180 Oe, respectively. After that, the relative transmission begins to decrease with increasing magnetic field. At 1980 Oe, the maximum enhancement factor is ∼8 while the minimum enhancement factor is less than one. For sample S_3 in which $\Phi_{\rm MH}$ is 1.5×10^9 ml^{-1}, the evolution of the relative transmission spectrum with increasing magnetic field is similar to that observed in sample S_2. However, the maximum enhancement factor, which is also observed at 180 Oe, is almost doubled.

In sample S_4, we observe similar evolution of the transmission spectrum due to the increase of magnetic field. However, an impressive and remarkable enhancement factor as large as ∼150 is achieved at 270 Oe. Even at the highest magnetic field (1980 Oe), an enhancement factor of ∼80 is still attainable.

Surprisingly, a sharp decrease of enhancement factor from 150 to 2.3 occurs when $\Phi_{\rm MH}$ is slightly raised from 2.3×10^9 to 2.5×10^9 ml^{-1}, as shown in Figs. 2(d) and 2(f) for samples S_4 and S_6. This is the most important and intriguing feature we observed in the experiments. Figure 2(f) shows that the maximum enhancement factor is obtained at a much higher magnetic field of 720 Oe. In addition, the spectral shape is also changed.

As $\Phi_{\rm MH}$ is further increased to 2.7×10^9 ml^{-1}, the relative transmission increases again (see Fig. 2(g)). The maximum enhancement factor obtained at 1260 Oe is increased to ∼14. In Fig. 2(h), we can see that the maximum enhancement factor becomes ∼22 and the spectral shape is changed back to the original one. Besides, it is noticed that the maximum enhancement factor is obtained at the highest magnetic field (1980 Oe).

One of the remarkable features observed in Fig. 2 is the significant enhancement in transmission achieved at $\Phi_{\rm MH} = 2.3\times10^9$ ml^{-1}. It implies that the sample which is originally opaque to an incident light will become transparent to it by the application of a magnetic field. This property is completely different from that observed in pure magnetic fluids where a reduction in transmission is generally observed after applying a magnetic field. In order to gain a deep insight into this phenomenon, we show in Fig. 3 the evolution of the transmitted light through sample S_4 when a magnetic field is imposed, increased and finally withdrawn. The 632.8-nm light from an He–Ne laser was used and the transmitted intensity was recorded by using a CCD. It can be seen that the transmitted intensity through sample S_4 is too weak to be detected in the absence of a magnetic field due to

the strong scattering of the incident light by randomly distributed MHs (see Fig. 3(a)). As a magnetic field of 40 Oe is applied on the sample, randomly distributed MHs begin to organize into a relatively ordered structure. As a result, the transmitted intensity becomes detectable (see Fig. 3(b)). When the magnetic field is raised to 120 Oe, a strong transmitted light is observed and its intensity remains nearly unchanged for magnetic fields up to 360 Oe (see Figs. 3(d)–3(f)). When the magnetic field is removed, the transmitted intensity decreases rapidly and completely disappears after some time, as shown in Figs. 3(g) and 3(h). It implies that the random distribution of MHs is recovered due to Brownian motion.

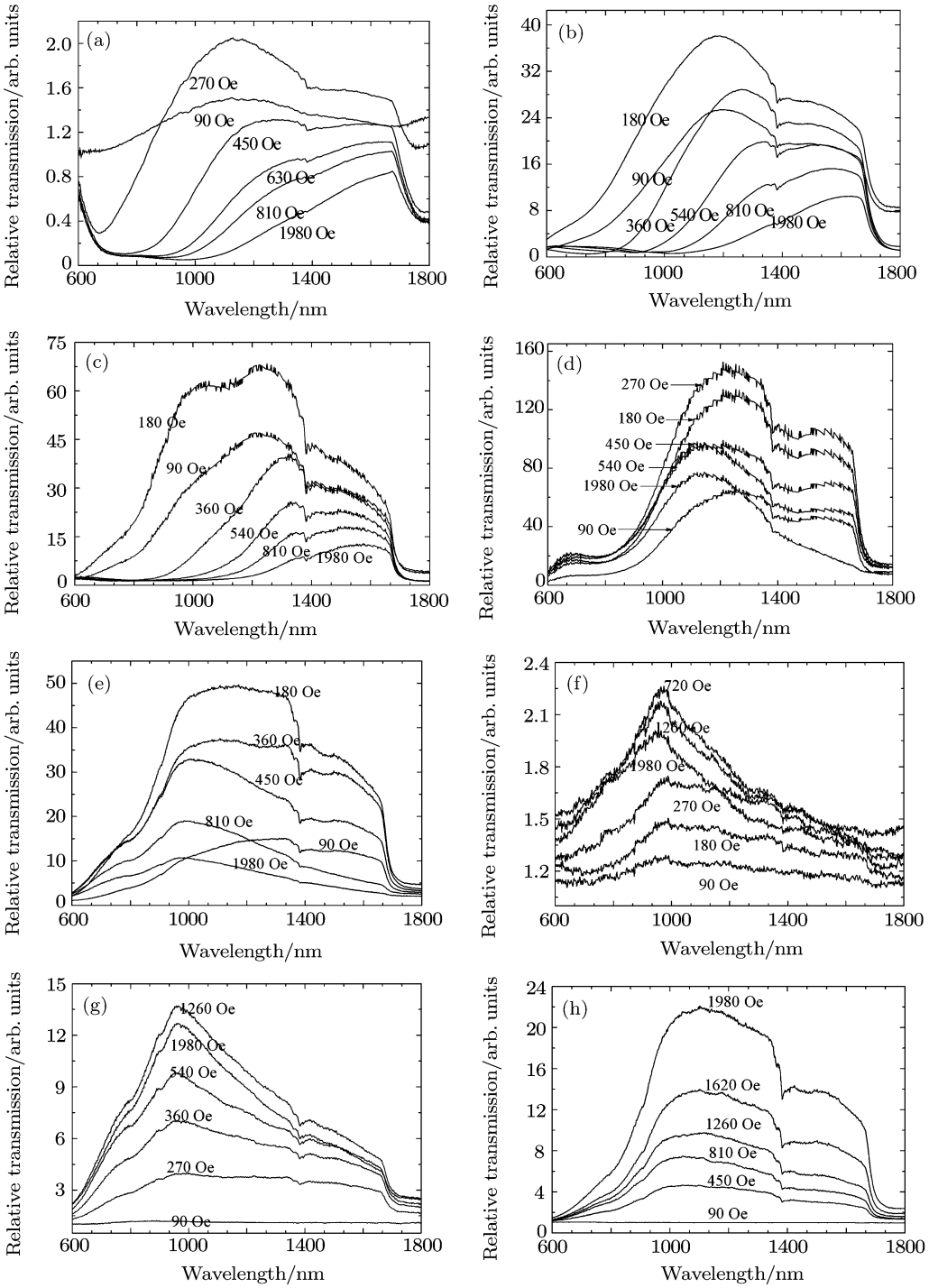

Fig. 2. Relative transmission spectra measured under different magnetic fields for samples S_1 to S_8 (figures (a)–(h)).

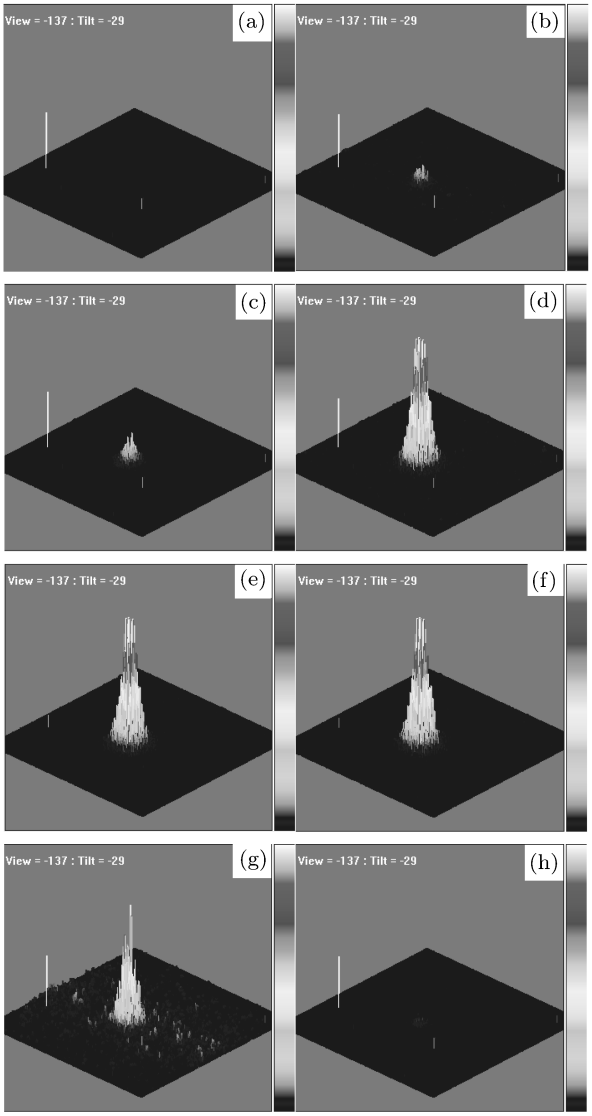

Fig. 3. Evolution of the transmitted light through sample S_4 when a magnetic field is imposed, increased and finally withdrawn: (a) $H = 0$ Oe; (b) $H = 45$ Oe; (c) $H = 90$ Oe; (d) $H = 180$ Oe; (e) $H = 270$ Oe; and (f) $H = 360$ Oe. Images (g) and (h) show the intensity distributions of the transmitted light 2 and 4 s after the switching-off of the magnetic field from 360 Oe.

5. Different phases observed in colloidal liquids with MHs of different volume densities

In order to understand the response behaviour of colloidal liquids to magnetic field, we present in Fig. 4(a) the evolution of the maximum enhancement factor ($\eta_{\max}$) with increasing $\Phi_{\rm MH}$. As $\Phi_{\rm MH}$ increases, the value of $\eta_{\max}$ increases monotonically from ∼2 for sample S_1 to ∼150 for sample S_4. Surprisingly, it drops sharply to ∼2.3 for sample S_6 when $\Phi_{\rm MH}$ exceeds a critical value which is $\sim 2.5\times10^9$ ml^{-1}. However, a further increase in $\Phi_{\rm MH}$ leads to a recovery of $\eta_{\max}$ and it reaches 22 in sample S_8.

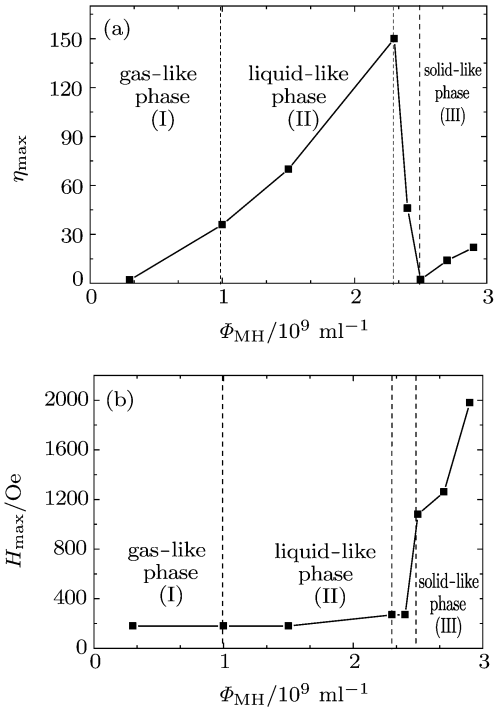

Fig. 4. (a) Evolution of the maximum enhancement factor ($\eta_{\max}$) with increasing $\Phi_{\rm MH}$. (b) Dependence of the magnetic field at which the maximum enhancement factor is achieved ($H_{\max}$) on $\Phi_{\rm MH}$.

In the presence of a magnetic field, the interaction between MHs originating from the induced magnetic moments results in a redistribution of MHs in the magnetic fluid. In general, the attraction of vertically aligned magnetic moments leads to the formation of MH chains along the direction of the magnetic field. In addition, a repulsive interaction exists between these MH chains. As a result, the colloid may exist in three different phases, namely the gas-like, liquid-like and solid-like phases, depending on the value of $\Phi_{\rm MH}$. In order to view the distribution and arrangement of MHs in different phases, we have prepared several samples by filling thin sample cells of ∼20 μm with the colloidal liquids listed in Table 1. Then, we used the microscope and CCD to observe and record the arrangement of MHs under a magnetic field parallel to the sample cells, as shown in Fig. 5. For small $\Phi_{\rm MH}$ ($\Phi_{\rm MH} < 1.0\times10^9$), the interaction between MHs is too weak to form any magnetic chains. Only dimmers are observed in the colloid. Thus, the transmission of the colloid is not affected too much by the magnetic field. In this case, we can say that the colloid is in a gas-like phase and appears in samples S_1 and S_2 as shown in Fig. 5(a). In this case, a

quite small $\eta_{\max}$ is observed. With increasing $\Phi_{\rm MH}$, the mean separation between MHs is reduced and the interaction between them is enhanced. Consequently, the possibility to form MH chains is increased significantly and we can observe many long MH chains in the colloid when a magnetic field is applied. This situation is clearly distinct from the gas-like phase and we say that the colloid is changed to a liquid-like phase. Such a phase occurs in samples S_3 and S_4 in which the formation and alignment of MH chains are shown in Fig. 5(b). Obviously, a significant change in the transmission of the colloid is expected in this case when a magnetic field is imposed. Accordingly, we obtain a $\eta_{\max}$ as large as $\sim$150 in sample S_4. In this case, MHs which are randomly distributed in the colloid are self-organized into MH chains by the application of a magnetic field. As a result, the incident light which suffers from strong scattering in the absence of the magnetic field becomes much less scattered, leading to a high transmission.

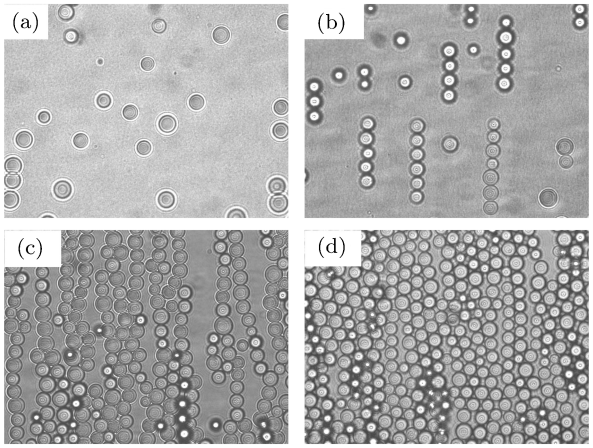

Fig. 5. Arrangements of magnetic holes in different samples under magnetic fields parallel to the sample cells: (a) S_1; (b) S_3; (c) S_5; and (d) S_7.

The most interesting and important feature observed in Fig. 4(a) is the sharp transition from the liquid-like phase to the solid-like one, which is reflected in the dramatic reduction of the enhancement factor. As shown in Fig. 4(a), $\eta_{\max}$ drops abruptly from 150 to 2.3 when $\Phi_{\rm MH}$ exceeds the critical value. As shown in Fig. 5(c), the image recorded by the CCD indicates an irregular distribution of MHs which is in sharp contrast to the ordered distribution of MHs in the liquid-like phase. We thought that the colloid with this $\Phi_{\rm MH}$ has entered into a solid-like phase. When $\Phi_{\rm MH}$ further increases, MHs tend to form a close-packed structure with better ordering, leading to the gradual increase in the enhancement factor, as shown in Fig. 5(d).

As mentioned above, the magnetic field at which the maximum enhancement factor is achieved ($H_{\max}$) exhibits a dependence on $\Phi_{\rm MH}$. The relationship between these two quantities is presented in Fig. 4(b). It is interesting to note that $H_{\max}$ remains nearly unchanged ($\sim$270 Oe) when $\Phi_{\rm MH}$ is below the critical level. After that, a rapid increase of $H_{\max}$ is observed with increasing $\Phi_{\rm MH}$. This phenomenon will be discussed in detail in Section 7.

6. Switching behaviour of colloidal liquids in different phases

In order to gain a deep insight into the response behaviour of the colloids with different $\Phi_{\rm MH}$, we have examined the switching operation of several typical samples (S_1, S_4 and S_7) under a magnetic field. The results are compared in Fig. 6. The times at which the magnetic field was switched on and off are indicated by dotted lines. The wavelength of the incident light and the magnetic field strength were fixed to be 1000 nm and 270 Oe, respectively. The transmitted intensity in the absence of the magnetic field is normalized to one. For each sample, well-defined and repeatable switching operations are observed. For sample S_1, we observed a reduction of the transmission after the magnetic field is switched on. It reaches a steady state with a constant transmission of $\sim$10 dB after some time. When the magnetic field is switched off, the transmission recovers rapidly to the original level. This response behaviour is quite similar to conventional magnetic fluids without any MHs.[12] For sample S_4 in which the colloid exists in the liquid-like phase, a completely different switching operation is observed. A significant increase in the transmission is found after the switching-on of the magnetic field. However, the response speed is obviously slower than that observed in sample S_1 and it takes a relatively longer time to reach the steady state. After the removal of the magnetic field, the recovery of the transmission to the original level occurs in a short time which is comparable to the recovery time of sample S_1. This behaviour indicates that the Brownian motion is still effective to destroy the MH chains formed by the attraction of magnetic moments. As for sample S_7, the switching behaviour is similar to that of sample S_4 but with a longer response time and a smaller

extinction ratio (∼10 dB).

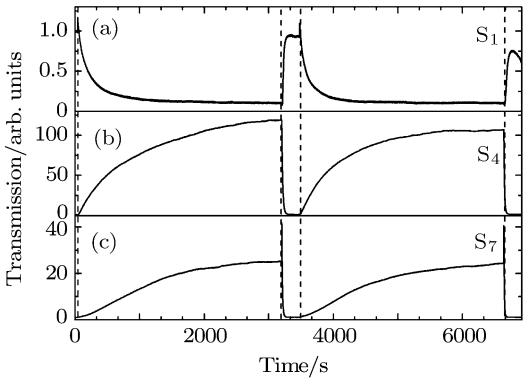

Fig. 6. Switching operations of several typical samples (S_1, S_4 and S_7) under a magnetic field of 270 Oe.

7. Physical mechanism determining the response behaviour

When the colloid is in gas-like phase, the mean separation between MHs is quite large and the possibility for them to form dimers or chains through magnetic moment attraction is quite small. In this case, the response of the colloid to a magnetic field is similar to that of a pure magnetic fluid. Magnetic nanoparticles aggregate into magnetic clusters or magnetic chains with a typical size of several hundred nanometers, resulting in a stronger scattering of the incident light in near infrared region. Consequently, a reduction in the transmission is observed with increasing magnetic field.

From Fig. 4(a), no obvious transition is observed between the gas-like and liquid-like phases. When the interaction between magnetic moments is considered, only the liquid phase is introduced. With increasing $\Phi_{\rm MH}$, the interaction between MHs becomes significant and many MH chains are created under a magnetic field. In this case, the regular alignment or distribution of MHs greatly reduces the scattering of the incident light, thus leads to a dramatic increase in the transmission of the incident light. Meanwhile, clustering of magnetic nanoparticles also occurs but the transmission of the colloid is dominated by the self-alignment of MHs. In sample S_4, it is expected that MH chains with lengths comparable to the thickness of the sample cell are well distributed in the magnetic fluid. In the transverse plane, MHs are self-organized into a hexagonal lattice due to the repulsive interaction between magnetic moments.[18]

As mentioned above, the colloid will enter into a solid-like phase when $\Phi_{\rm MH}$ exceeds the critical value. In this case, the mean separation of MHs is so small that stable MH chains cannot be obtained possibly because of the thermally driven and defect driven coarsening.[28] We can imagine this situation by adding more MHs into sample S_4 in which long and well-distributed MH chains have been achieved. The repulsive interaction between the extra MHs and those in the MH chains will distort or even destroy the MH chains, resulting in a disordered distribution of MHs. In addition, it has been suggested that the interaction between magnetic chains formed by magnetic nanoparticles can be repulsive or attractive, depending on the relative position of the two magnetic chains in the direction parallel to the chains.[29] We think that the similar situation occurs for MH chains. Therefore, the drastic reduction in the transmission of the colloid is easily understood. The boundary between the liquid-like and solid-like phases is thought to be the value at which the reduction of the transmission of the colloid occurs (i.e., the critical value). It can be determined by carefully measuring the dependence of the transmission of the colloid on $\Phi_{\rm MH}$ under the application of an external magnetic field. In our case, we think that the phase boundary between the liquid-like and solid-like phases appears in the range of $2.3 \times 10^9 < \Phi_{\rm MH} < 2.5 \times 10^9$. A more accurate determination of the phase boundary can be achieved by inserting more measurement points in this range.

With increasing $\Phi_{\rm MH}$, the distribution of MHs becomes complicated. Through the repulsive interaction between the MHs in the presence of a magnetic field, however, the random distribution of them will become more regular and an enhancement in the transmission of the colloid is expected. However, more experiments are needed to clarify the actual distribution of MHs in colloidal liquids with large $\Phi_{\rm MH}$.

Before summarizing the research work, we would like to address the effect of light-induced magnetic clusters on the evolution of the distribution of MHs. The formation of laser-induced magnetic clusters was reported previously[27] and investigated later by us from the viewpoint of magneto–optical switches.[15] Basically, magnetic clusters with small sizes can be created by irradiating a light (an incandescent light or a laser light) on a magnetic fluid. It was found by us that these small magnetic clusters can act as nuclei for the formation of large magnetic clusters when a magnetic field is applied.[15] In the experiments reported in this paper, small magnetic clusters may be created when we illuminate the colloid with an in-

candescent light (or a laser light) in the transmission measurements. The appearance of these small magnetic clusters can accelerate the formation of various structures (e.g., dimmers or MH chains) when a magnetic field is applied, this is similar to the formation of large magnetic clusters in a pure magnetic fluid.[15] In addition, it is thought that the small magnetic clusters induced by light may enhance the interaction between MHs but more experiments are needed to clarify this point.

8. Conclusion

In summary, we have investigated the response of colloidal liquids containing MHs of different volume densities to magnetic field by conventional transmission measurements and found that the enhancement in transmission under a magnetic field exhibits a strong dependence on the volume density of MHs. Three different phases with distinct distributions of MHs are identified in colloidal liquids with different volume densities of MHs. They exhibit completely different response to magnetic field and switching operations. With increasing volume density of MHs, phase transition from the gas-like phase to the liquid-like one and finally to the solid-like one occurs. The transition from the liquid-like phase to the solid-like one, which is reflected in the dramatic reduction of the enhancement factor, appears to be abrupt. Accordingly, the magnetic field at which the maximum enhancement factor is achieved also shows a rapid increase after the transition. Our finding may find applications in the design of magneto–optical devices based on MHs.

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