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Transmitted light relaxation and microstructure evolution of ferrofluids under gradient magnetic fields

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ABSTRACT

Using light transmission experiments and optical microscope observations with a longitudinal gradient magnetic field configuration, the relationship between the behavior of the transmitted light relaxation and the microstructure evolution of ionic ferrofluids in the central region of an axisymmetric field is investigated. Under a low-gradient magnetic field, there are two types of relaxation process. When a field is applied, the transmitted light intensity decreases to a minimum within a time on the order of 10^1–10^2 s. It is then gradually restored, approaching its initial value within a time on the order of 10^2 s. This is type I relaxation, which corresponds to the formation of magnetic columns. After the transmission reaches this value, it either increases or decreases slowly, stabilizing within a time on the order of 10^3 s, according to the direction of the field gradient. This is a type II relaxation, which results from the shadowing effect, corresponding to the motion of the magnetic columns under the application of a gradient force. Under a magnetic field with a centripetal high-gradient (magnetic materials subjected to a force pointing toward the center of the axisymmetric field), the transmitted light intensity decreases monotonously and more slowly than that under a low-gradient field. Magnetic transport and separation resulted from magnetophoresis under high-gradient fields, changing the formation dynamics of the local columns and influencing the final state of the column system.

1. Introduction

Ferrofluids are stable colloid systems containing magnetic nanoparticles (about 10 nm in diameter) suspended in a liquid carrier [1]. Without a magnetic field, there are some self-assembled structures such as monomers, dimers, trimers and short chains of nanoparticles in ferrofluids. When an external magnetic field is applied, the self-assembled structures undergo Néel and Brownian rotation, forming long chains, parallel to the field direction. Because of the lateral coalescence among the long chains under a certain critical magnetic field, the chains transform into columns, whose diameters can reach 0.1–1 μm. A great deal of theoretical, experimental and simulational research on chain/column structures under uniform magnetic fields has been carried out (including the research in the chapters edited by Ilg and Odenbach in Ref. [1]). Recently, tunable technologies with magnetic columns have exhibited the potential for application in optical devices [2–5]. The magneto-optical relaxation behavior, including relaxation of the transmitted light under a magnetic field, is a type of dynamic characteristic of ferrofluids and it is inevitable in research in the field of the optical applications of ferrofluids.

In most of the previous works reported, the external magnetic field is usually uniform and static. However, in some experimental studies, the magnetic field may change with time, for example a field of ramping rate [6] and an alternative field [7], or the field may change with space, such as a static gradient field [8]. Recently, the magnetophoresis in ferrofluids under a gradient magnetic field has raised concerns. Magnetophoresis changes the distribution of the magnetic particles in ferrofluids and effects the concentration distribution [9,10], diffusion [11], the transport of the microstructures [12] and the magnetization [13]. These changes could affect the magneto-optical relaxation behavior of ferrofluids.

Under a gradient magnetic field, regular variations in the relaxation behavior of the transmitted light through ferrofluid films, which have valley and peak in the T_n-t curves, have been observed [14–16] (see Fig. 1). Li et al. [17] divided the variations in the light transmission (T_n-t curves) under a centripetal gradient field into four stages, t_0–t_1 for stage I, t_1–t_2 for stage II, t_2–t_3 for stage III and t_3–t_4 for stage IV, as shown in Fig. 1. T_n is the normalized transmission. $T_n=I(t)/I(t_0)$, where $I(t)$ is the intensity of the transmitted light and t_0 is the time when the magnetic field is turned on. According to the Bouguer–Lambert–Beer law [10], the

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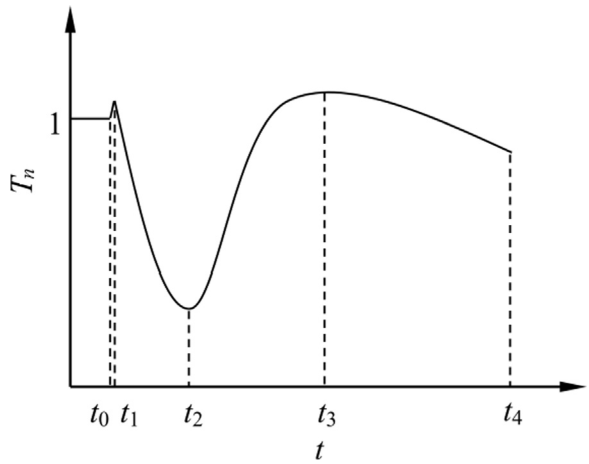

Fig. 1. General transmitted light relaxation curve of ferrofluids under a centripetal gradient magnetic field.

normalized transmission can be written as: $T_n = [I_0 e^{\psi(t)}]/[I_0 e^{\psi(t_0)}]$, which is dependent on the microstructure state of the ferrofluid ($\Psi(t)$) and the reference state ($\Psi(t_0)$), and is independent of the incident light intensity (I_0). T_n is directly proportional to the light transmittance, T_r ($=I(t)/I_0$). After the magnetic field is turned on at t_0, T_n increases quickly during the first stage and reaches a maximum at t_1. It then decreases to a minimum at t_2. T_n begins to restore gradually from t_2 and reaches a maximum at t_3. It then decreases slowly until t_4, which is the time when the field is turned off. Generally, the evolution of the microstructures in ferrofluids can be inferred from the light transmitted and scattered through the ferrofluid films [18–22]. For a mixture of small and large particles, valley and peak in the magneto-optical transmission have been observed [23,24], which could be attributed to scattering from the large particles ($\sim$0.5 μm).

The transmitted light relaxation behavior in ferrofluids under a gradient magnetic field has been explained using two possible models for the microstructure variations. One is a model of the formation of chains/columns [15] and the other is a model of the motion of the chains/columns [16]. In spite of this, there are different opinions as to which model corresponds to each stage of the relaxation of the transmitted light. It is unclear to what extent the magnitude and direction of the gradient influence the relaxation of the transmitted light. In this paper, by analyzing the transmitted light relaxation curves and the optical microscope images of ferrofluids, the corresponding relationship between the variations of the microstructures and the relaxation of the transmitted light in ferrofluids under a gradient magnetic field is clarified and the influence of the magnitude of the gradient and direction on the transmitted light relaxation is determined. Magneto-optical effects in ferrofluids are complicated, and interpreting the experimental data depends on the type of ferrofluid used [25]. Thus, ionic $CoFe_2O_4$ ferrofluids, prepared by Massart's method [26,27] and aged over one month, were used here for the comparability of the experiments on the transmitted light relaxation [14,15,17]. The size of $CoFe_2O_4$ colloidal particles is order of 10 nm [27], so that the scattering from individual particles can be neglected [28].

2. Experiments

2.1. Experimental setup

The experimental setup shown in Fig. 2 was used to measure the relaxation behavior of the light transmission through ferrofluids under a magnetic field. The light signals were received by a photoelectric detector, and gathered and recorded on a computer in real time. The magnetic field had a preset value that was turned on and off by a computer-controlled current source. The times that it was turned on and off were preset in the computer program. An electromagnet (equipped with one of two alternative pairs of pole shoes with central hole diameters of 5 mm) and a solenoid (with a length and inner diameter of 300 mm and 35 mm, respectively) or a permanent magnet (NdFeB, the diameter of the central hole was 2.5 mm) were used to generate the magnetic fields. A current source and a teslameter are not required when a permanent magnet is used to generate the field. In the experiments, the sample was placed between the pole shoes of the electromagnet, or at a depth of 150 mm in the solenoid, or on the plane, 7.5 mm above the upper surface of the permanent magnet. A cylindrical coordinate system was established, as shown in Fig. 2. The light source is a He–Ne laser with a central wavelength of 632.8 nm, whose power is tunable (the maximum is 5 mW). The light travels through the sample with the wave vector (**k**) parallel to the z-axis. The light is approximately Gaussian beam (divergence angle 1 mrad). The diameter of the spot on the sample is 2.9 mm. The electromagnet or solenoid was equipped with a mechanical device so that they could be set either parallel or perpendicular to the direction of gravity.

The magnetic field generated by the equipment is deemed to be axisymmetric. Its symmetry axis is parallel to the z-axis, as shown on the right in Fig. 2. The radial component (B_r) of the magnetic field along the sample plane is neglected because its magnitude is much smaller than that of the axial component (B_z). The axial component of the field at the cross-point of the symmetry axis and the sample plane is called the magnetic field at the symmetry center (MFSC).

Before the light transmission experiments were implemented, the radial distributions of the axial components of the magnetic field were measured. By attaching the Hall probe of the teslameter to a micrometer caliper, the magnetic flux density data was sampled at regular intervals along an arbitrary radial orientation (coordinate x-axis). Data were measured every 0.1 mm for the field generated by an electromagnet and every 0.2 mm for those generated by a solenoid and a permanent magnet. The symmetry point of the magnetic field on the x-axis was treated as the origin of the coordinates. The gradient of magnetic field (∇B) was calculated by fitting the data with polynomials. If the field intensity at the center of symmetry is larger than that of the ambient, ∇B directs from the ambient to the center along a radial axis and this magnetic field is called the centripetal gradient field. For the contrary direction of ∇B, the field is called centrifugal gradient field.

B_1, B_2, B_3 and B_4 denote the magnetic fields at the sample plane generated by the electromagnet with two dissimilar pairs of pole shoes, the solenoid and the permanent magnet respectively. The gradient distributions of the magnetic fields with a given MFSC are shown in Fig. 3. The maximum B_1 appears at about ± 3 mm, and the minimum B_1 appears at the origin. Therefore, B_1 can be regarded as a centrifugal gradient magnetic field in the range $-2\ \text{mm} < x < 2\ \text{mm}$, and the gradient of B_1 is estimated to be on the order of 10^{-2} T/m. B_2 is a maximum at the origin, and it is a centripetal gradient magnetic field. The gradient of B_2 is on the order of 10^{-1} T/m in the range $-2\ \text{mm} < x < 2\ \text{mm}$. The gradient of B_3 is constant on the order 10^{-2} T/m because the symmetry axis of B_3 deviates dramatically from the geometric symmetry axis of the solenoid. The distribution of B_4 is similar to that of B_2 and it is also a centripetal gradient magnetic field. The order of the gradient of B_4 is 10^0T/m in the range $-2\ \text{mm} < x < 2\ \text{mm}$. Except for the solenoid, the symmetry axes of the magnetic fields generated by the other pieces of equipment deviate slightly from their geometric axes (less than 0.4 mm). Because of the spatially