Observation of magneto-optical effect in extremely dilute ferrofluids under weak magnetic field

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Received December 9, 2011; revised January 17, 2012; accepted January 18, 2012; posted January 18, 2012 (Doc. ID 159752); published March 26, 2012

We propose a novel liquid-core waveguide using water-based ferrofluids as the guiding layer in a symmetrical metal-cladding waveguide structure to investigate the magneto-optical effect of extremely dilute ferrofluids. Owing to the high sensitivity of the ultra-high-order modes, the reflection intensity can be effectively tuned even by a weak magnetic field and the modulated reflectivity exhibits no threshold behavior. Furthermore, by properly adjusting the transmission axes of the polarizer, the detected laser intensity can be magnetic-field independent because the refractive indices for ordinary and extraordinary rays vary oppositely under the external field. © 2012 Optical Society of America

OCIS codes: 160.3820, 230.7390, 310.6860.

1. INTRODUCTION

Ferrofluids, which refer to stable colloids consisting of suspended magnetic nanoparticles in liquid carriers, combine Newtonian fluid behavior with the magnetic properties of superparamagnets [1]. It is already well established that an externally applied magnetic field will orient the magnetic moment of each particle along its direction and result in aggregates consisting of nanoparticles [2]. Consequently, the hydrodynamic, magnetic, and optical properties of the fluids can be effectively magnetically modulated [3–6]. In the past decade, much attention has been focus on its versatile magneto-optical effects, such as static magnetic birefringence, dichroism, and magnetically tunable refractive index, because of their broad applications in various functional optical devices, e.g., sensors, switches, modulators, and photonic crystal fiber [7–11]. On the other hand, investigation on ferrofluids is also nontrivial from the theoretical point of view. As typical dipolar fluids, the phase behavior and microstructure formation of ferrofluids with or without an external field are longstanding issues of great interest [12–14]. Using ferrofluids as a nonequilibrium pattern forming system driven by an ac magnetic field, various superlattice patterns such as parametrically excited standing surface waves or stable solitonlike structures have been observed [15–17]. Recently, it has even suggested that ferrofluids are suitable to investigate thermal ratchet behavior [18].

The magneto-optical phenomena of ferrofluids, especially the issue of optical transmission of a ferrofluid film under an applied magnetic field, have been extensively studied recently [19,20]. However, due to limited experimental accuracy, few works addressed the issue of extremely dilute ferrofluids under weak field experimentally, which might shed some light on the formation of complex structures containing very few particles with permanent dipole moments [12,13]. In our recent work, instead of using the extensively studied ferrofluid film, we sealed the ferrofluids in a symmetrical metal-cladding optical waveguide (SMCW) structure of millimeter scale and illustrated a millisecond-scale switching time [21]. In this paper, we take a step further by carrying out magneto-optical measurements of extremely dilute water-based Fe_3O_4 ferrofluids via the excited ultra-high-order modes. As described in [10], the refractive index of water-based Fe_3O_4 with concentration of 0.85 emu/g does not change until the applied field exceeds the critical field strength (about 22 Oe). Owing to the high sensitivity of the SMCW structure, we found that the field strength responsible for a notable change in the reflectivity is only several oersted (Oe), even when the sample is diluted to a low concentration of only $6.46 \times 10^{-3}\%$. Optical anisotropy of the extremely dilute ferrofluids is also observed.

2. THEORY

As shown in Fig. 1, the SMCW structure in our experiment is composed of three parts: (i) a thin glass slab of 0.3 mm and a sample cell of 0.7 mm sandwiched between two metal films work as the guiding layer, (ii) a thin gold film (about 30 nm) is coated on the top side of the thin glass slab to enable better coupling, and (iii) a relatively thick gold film (>300 nm) is deposited on a thick glass slab to prevent light leakage. In this scheme, we do not need to rely on the widely used grating-coupling or prism-coupling method to couple the light source into the guiding layer, because the so-called free space coupling technology can be applied [22]. The guided modes excited under the phase-matching condition, which are also

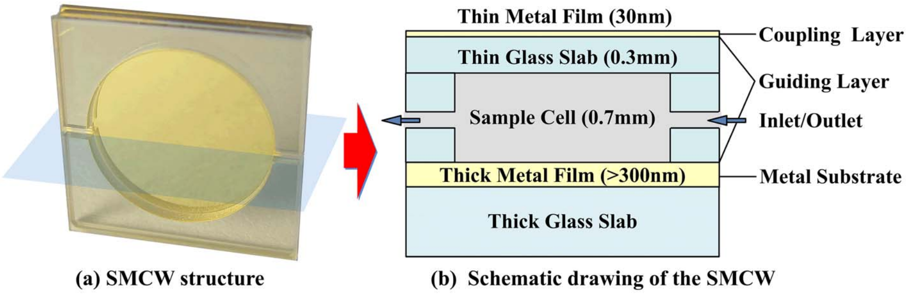

Fig. 1. (Color online) Configuration of the SMCW structure.

known as the ultra-high-order modes, result in a serial of sharp dips in the reflection spectrum. And the variation in the refractive index of the ferrofluids upon the application of the magnetic field will give rise to a shift of the resonance angles of these sharp dips: an increase in the refractive index will shift the resonance angles right, and vice versa. When the operation incident angle is selected at the middle area of the falling edge or rising edge, good linearity can be achieved. Different from those conventional sensors based on waveguide structure or surface plasmon resonance (SPR), in which an evanescent field is applied for detection, the proposed SMCW structure utilizes an oscillating field instead of the widely applied evanescent field. Based on our previous works [21,23], the excited modes propagate along the guiding layer with high intensity. Thus a significant sensitivity enhancement can be obtained owing to the strong concentration of the electromagnetic field [23,24]. In this case, even a slight perturbation in the formation structure of the nanoparticles will lead to a significant change in the reflection intensity. To make this point clear, a more specific theory is needed. In order to obtain explicit analytical expressions, here we consider a three-layer optical waveguide consisting of a thin metal coupling layer, a guiding layer, and a metal substrate. Different from the actual scheme used in our experiments, the guiding layer in this simplified SMCW structure model contains only one single homogeneous layer.

A simple theoretical formula to analyze the reflectivity of this optical system can be expressed as [25]

$$R = |r_{12}|^2\left(1 - \frac{4\,\mathrm{Im}(\beta^0)\mathrm{Im}(\Delta\beta^{\mathrm{rad}})}{[k_0N - \mathrm{Re}(\beta^0) - \mathrm{Re}(\Delta\beta^{\mathrm{rad}})]^2 + (\mathrm{Im}(\beta^0) + \mathrm{Im}(\Delta\beta^{\mathrm{rad}}))^2}\right), \tag{1}$$

where $r_{ij} = \frac{\kappa_i - \kappa_j}{\kappa_i + \kappa_j}$ denotes the Fresnel reflection coefficient, k_0 is the wavenumber in vacuum, $N = \sqrt{\varepsilon_1}\sin\theta$ is the effective index of the guided mode, and θ is the angle of the incident beam from the air space. The normal component of the wave vectors in each medium κ_i is given by $\kappa_i = k_0(\varepsilon_i - N^2)^{1/2}$. The subscripts 1, 2, 3, and 4 here represent the free space, coupling layer, guiding layer, and metal substrate, respectively. Here only the TE mode is considered.

The term β^0 represents the eigenpropagation constant of a guided mode in a simplified metal–dielectric–metal model where the two metal films are both considered to be semi-infinite. This term can be numerically calculated through the dispersion equation in the SMCW structure, and its imaginary part $\mathrm{Im}(\beta^0)$ is the intrinsic damping, which represents the Joule loss in the metal when complex dielectric constants of metal are considered. $\Delta\beta^{\mathrm{rad}}$ represents the propagation constants difference between the simplified metal–dielectric–metal model and the proposed three-layer waveguide coupling system and is given by [25]

$$\Delta\beta^{\mathrm{rad}} = \frac{\kappa_3 r_{12}[r_{23} + r_{34}\exp(2i\kappa_3 h_3)]\exp(2i\kappa_2 h_2)}{2\beta^0\left(\frac{1}{\kappa_2} + \frac{1}{\kappa_4} - ih_3\right)}. \tag{2}$$

The imaginary part of $\Delta\beta^{\mathrm{rad}}$ is called radiative damping and represents the leakage loss of the guided mode back into the free space.

From Eq. (1), it is clear that when the phase-matching condition $k_0N = \mathrm{Re}(\beta^0) + \mathrm{Re}(\Delta\beta^{\mathrm{rad}})$ is satisfied, the reflectivity reaches its minimum value, namely,

$$R_{\min} = |r_{12}|^2\left[1 - \frac{4\,\mathrm{Im}(\beta^0)\mathrm{Im}(\Delta\beta^{\mathrm{rad}})}{(\mathrm{Im}(\beta^0) + \mathrm{Im}(\Delta\beta^{\mathrm{rad}}))^2}\right], \tag{3}$$

and the half-width of the resonance dip, which is completely determined by the intrinsic and radiative dampings, can be roughly approximated as

$$W_\theta = \frac{2[\mathrm{Im}(\beta^0) + \mathrm{Im}(\Delta\beta^{\mathrm{rad}})]}{k_0\sqrt{\varepsilon_1}\cos\theta_{\mathrm{res}}}. \tag{4}$$

Finally, it is easy to deduce from the dispersion equation of the guided modes the following relation:

$$d\theta = \frac{1}{\sqrt{\varepsilon_1}\cos\theta}dN \approx \frac{n_3}{\varepsilon_1\sin\theta\cos\theta}dn_3. \tag{5}$$

So based on the above equations, a rather rough estimation of the intensity variation of the reflected beam can be written as

$$\Delta R \approx \frac{1 - R_{\min}}{W_\theta}\frac{d\theta}{dn_3}\Delta n_3. \tag{6}$$

Inserting the following parameters: $\varepsilon_2 = \varepsilon_4 = -20 + 1.5i$, $\varepsilon_3 = 2.25$, $h_2 = 30$ nm, $h_3 = 1$ mm, $\theta = 8.657^\circ$, the calculated ratio of ΔR to Δn is about 3.27×10^4, the simulated results confirm the high sensitivity of the proposed structure.

3. EXPERIMENT AND DISCUSSION

The water-based Fe_3O_4 ferrofluid samples were provided by Anhui Jingke Magnetic Liquids Co., Ltd., and the distribution of the magnetic nanoparticle size ranges between 8 and 12 nm and follows the normal distribution with a central size of 10 nm. The density of the ferrofluids and nanoparticle are 1.20 g/ml and 5.18 g/cm^3, respectively, and the mass of particles per volume fraction is 0.248 g/ml. In the present investigation, the volume concentrations of the dilute ferrofluids are 0.079%, 0.053%, 0.0395%, 0.0316%, 12.9×10^{-3}%, and 6.46×10^{-3}%, respectively. Figure 2 is the absorption spectrum of one diluted sample, and it shows that less light is absorbed in the wavelength ranges from 600 to 900 nm. The refractive indices of different concentrations of the ferrofluids under zero fields were determined via SPR technique and plotted in the inset of Fig. 2.

An 850 nm continuous wave laser (Toptica DL 100) of 100 mW in power was used as a light source in our experiments. In order to obtain a well-collimated, polarized laser beam, a polarizer and two apertures with a diameter of 1 mm were set between the laser and the SMCW structure. A solenoid that can be placed at fixed positions marked as "I" or "II" was used to provide a uniform magnetic field either perpendicular or parallel to the surface of the SMCW structure. The magnetic field was measured by an HT100G Gauss meter provided by Shanghai Hengtong Magneto Electric Technology Co., Ltd. Figure 3 illustrates the experiment setup, and its inset gives the Poynting vector and electric displacement of the incident laser and the guided modes, respectively, when the incident light is TM/TE polarized. Based on basic waveguide theory, if any light is coupled into a guided mode of a planar waveguide, it will continue to propagate along the guiding layer. So the electric displacement vector of the TM-polarized incident light will be perpendicular to the structure surface, while the electric displacement vector of the TE-polarized light will be parallel to the structure surface. From now on, we will refer to the rays whose electric displacement vector is perpendicular/parallel to the magnetic field as ordinary/extraordinary rays. Figure 3 reveals that the TM-polarized laser is an extraordinary ray for magnetic field "I" and an ordinary ray for magnetic field "II," and the opposite situation occurs for the TE-polarized laser.

From the above discussion, different choices of the polarization of incident light and the direction of the magnetic field will result in a total of four cases. When an external magnetic field is applied to the ferrofluids, a variation in the refractive

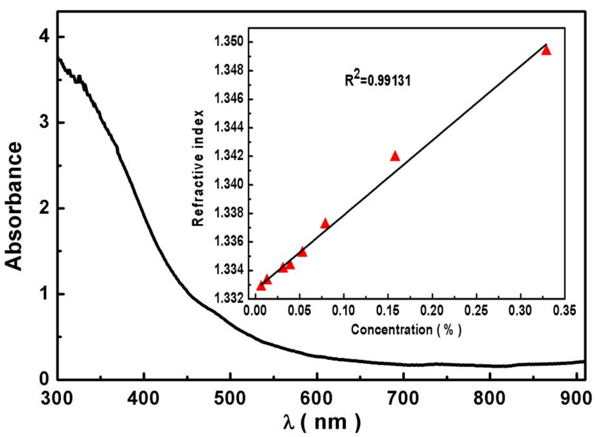

Fig. 2. (Color online) Linear absorption spectra of dilute ferrofluids with volume concentration of 6.46×10^{-3}% and (inset) calibration curve of the refractive index of ferrofluids with different volume concentrations (triangles) fitted with a linear function ($R^2 = 0.99131$).

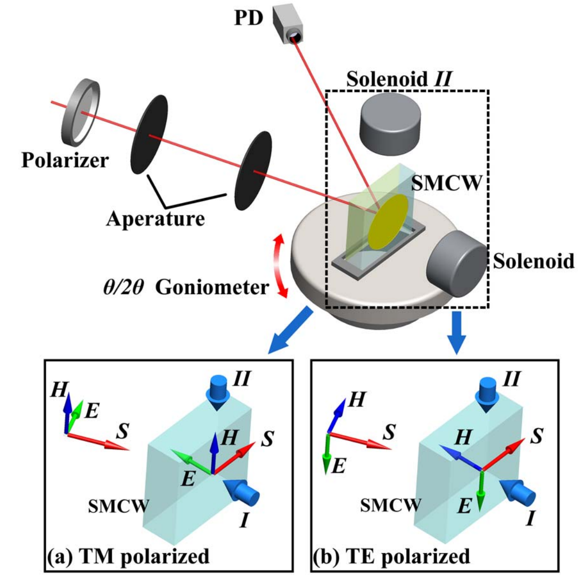

Fig. 3. (Color online) Schematic of the experimental setup. The details at the bottom show the Poynting vector and electric displacement of the excited modes when the incident light is TE/TM polarized.

index that results from the agglomeration of magnetic nanoparticles will occur. We experimentally measured the reflectivity variations as a function of the applied field strength corresponding to all four cases and plotted the dependences of the normalized reflectivity on the field strength in Fig. 4.

The concentration of the measured ferrofluids was 0.053%; the incident angle was fixed at the middle area of the falling edge, and the maximum magnetic field strength used was only 24 Oe. Combined with the SMCW structure, no such threshold behavior as described in [10] was observed in these magneto-optical measurements, even for extremely diluted ferrofluids. This may probably be due to the optical trapping effect, which is induced by the periodic distribution of ultra-high-order modes in the guiding layer [21]. As a result, the refractive index changes Δn of the ferrofluids are determined by the light field in the guiding layer as well as the external field. Research is currently ongoing in our laboratory to investigate the laser field induced Δn under zero magnetic fields. Furthermore, the magnetic field dependent reflectivity of the liquid-core waveguide structure shows different trends for ordinary

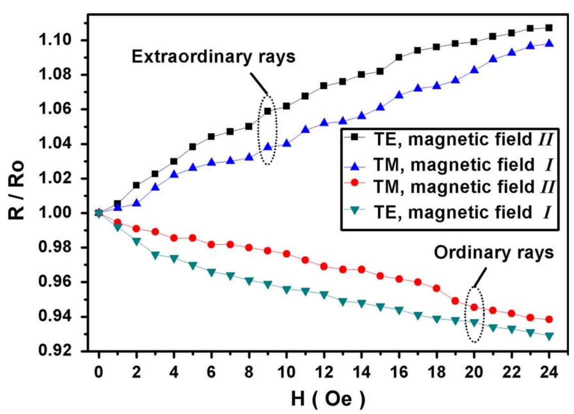

Fig. 4. (Color online) Normalized reflectivity as a function of applied field for four different cases. The concentration of the ferrofluids measured was 0.053%.

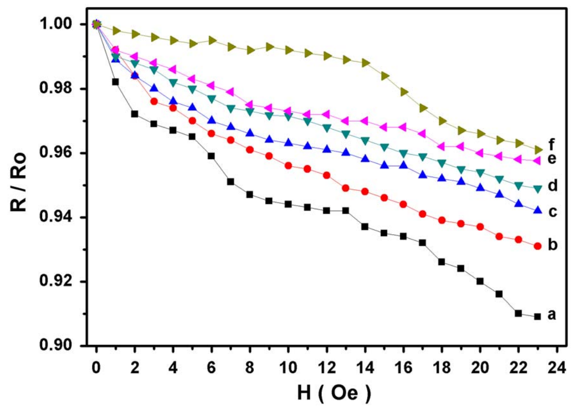

Fig. 5. (Color online) Normalized reflectivity of ordinary rays versus magnetic field for dilute samples with different concentrations: (a) 0.079%, (b) 0.053%, (c) 0.0395%, (d) 0.0316%, (e) $12.9 \times 10^{-3}\%$, (f) $6.46 \times 10^{-3}\%$.

and extraordinary rays due to the optical anisotropic property. The refractive index of the extraordinary (ordinary) ray increases (decreases) upon the application of the external field, no matter whether the magnetic field is perpendicular or parallel to the SMCW structure. Equivalently, the polarization of the incident light alone cannot determine how the refractive index varies. The difference between the top (bottom) two curves in Fig. 4 probably rises from the difference in the basic Fresnel formula governing the reflection characteristic of multilayer planar waveguide for TE (TM)-polarized light.

Figures 5 and 6 present the external field-modulated reflectivity of ordinary and extraordinary rays, respectively, when the SMCW structure was filled with diluted ferrofluids of different concentrations. Attributed to the high sensitivity of the ultra-high-order modes, the magneto-optical effect is observable even when the concentration of ferrofluids is as low as $6.46 \times 10^{-3}\%$. Because the intensities of ordinary and extraordinary rays vary oppositely under external field, it is theoretically and practically possible to make the total intensity invariant by properly adjusting the transmission axes of the polarizer. For example, when the field strength is 30 Oe and the concentration of the dilute ferrofluids is 0.053%, the variations in reflected intensity upon switching on and off the external field will not exceed 0.5% when the transmission axes are oriented by 44° to 46° relative to the vertical direction, because the variations in ordinary and extraordinary rays cancel each other out.

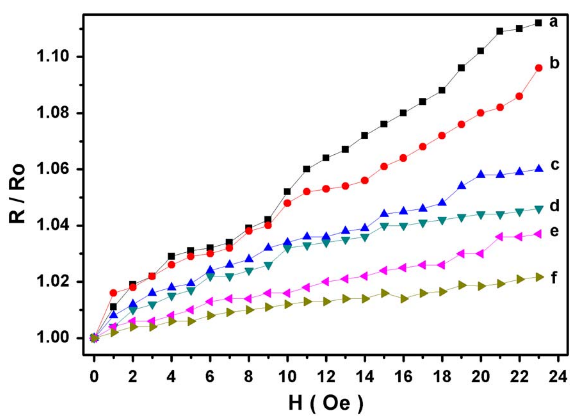

Fig. 6. (Color online) Normalized reflectivity of extraordinary rays versus magnetic field for dilute samples with different concentrations: (a) 0.079%, (b) 0.053%, (c) 0.0395%, (d) 0.0316%, (e) $12.9 \times 10^{-3}\%$, (f) $6.46 \times 10^{-3}\%$.

Based on the discussion above, there are many ways to tune the light intensity magnetically via this scheme for some practical applications, such as switching, modulation, and detection. Besides, this work may provide useful insight into the microstructure formation and phase diagram in ferrofluids with low densities and may be a new experimental approach to investigate systems containing very few particles with permanent dipole moments.

4. CONCLUSION

Although field modulation of light transmission through ferrofluid film has been intensively studied, it is difficult to observe the magneto-optical effect of extremely diluted ferrofluids, which is of great theoretical importance. Using ferrofluids as the guiding layer, the obtained liquid-core waveguide structure demonstrated a tunable reflectivity that depends strongly on the external field and no threshold behavior was observed in this scheme. Furthermore, we certified that the refractive index variation of dilute ferrofluids is determined by the relative angle between the electric displacement vector and the magnetic field direction.

ACKNOWLEDGMENT

This research was supported by the National Basic Research Program "973" of China (contract no. 2007CB307000), the National Natural Science Foundation of China (nos. 10874121, 10874119, and 10974044), and the Foundation for development of Science and Technology of Shanghai (grant no. 10JC1407200).

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