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Nonlinear optical effects in ferrofluids induced by temperature and concentration cross coupling

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Both experiment and numerical calculation were performed to study the nonlinear optical (NLO) effects in ferrofluids consisting of magnetic particles suspended in an organic solvent. We show that, in contrast to all the mechanisms responsible for NLO phenomena known so far, the novel NLO effects in ferrofluids in the zero applied field can be explained by the cross coupling between the temperature of the system and the concentration of particles through *thermophoresis*, which in turn leads to the spatial distribution of the refractive index. This NLO effect is enhanced by applying a moderate magnetic field whose mechanism is unclear so far. © 1998 American Institute of Physics. [S0003-6951(98)02003-8]

The discoveries of nonlinear optical (NLO) effects have greatly advanced our knowledge of the interaction between light and matter and, at the meantime, created a revolution in optical technology. Searching for new mechanisms for such effects will not only lead to better understanding of this interaction but also have great economic impacts in the information age. Recently, current authors have observed NLO effects in the diffraction patterns from a ferrofluid/magnetic fluid.¹ In this letter, we report new experimental and numerical results that demonstrate that a new mechanism—the thermophoresis induced coupling between the temperature and concentration, which leads to spatial distribution of phase and the index of refraction—is responsible to the NLO effect in ferrofluids reported earlier. We found that this NLO effect is significantly enhanced even in a small applied magnetic field. These results will lead to a new class of optical materials whose optical parameters can be easily controlled by external parameters such as gradient forces from temperature, field, and concentration.

Ferrofluid samples used in this study consist of magnetic particles suspended in kerosene.² The mean diameter of particles is 9 nm. The volume fraction of the magnetic particles is 6%. Figure 1 is the schematic of the experimental setup. The wave length of the He-Ne laser is $\lambda = 0.6328 \mu\text{m}$. The number of diffraction rings appeared at the far field after the laser is switched on, N , depends on both the thicknesses of the sample and the intensity of the laser. The time scale for the ring-formation process is in the order of 1 s for the focal length of 1 cm. The details about the setup and the sample have been published elsewhere.¹

Because of the large absorption coefficient of our sample (550/cm), there is a temperature increase in the liquid. The temperature change follows the thermal conduction equation

$$\frac{\partial T}{\partial t} - \chi \Delta T = \frac{\sigma I}{\rho C_p}, \quad (1)$$

where σ is the absorption coefficient of the ferrofluid that depends on the concentration of particles, I the intensity of the laser beam, ρ and C_p the density and the heat capacity of the system, and χ the thermal diffusivity. The time for ther-

mal conduction can be estimated by: $\tau_{\text{con}} \sim a^2/\chi$, a is the distance that the heat flux travels in time τ_{con} . For focal length of 1 cm, $a \sim 10 \mu\text{m}$ if we take the diameter of the focused beam as a . We get $\tau_{\text{con}} \sim 0.1$ ms for our ferrofluid, several orders of magnitude faster than the ring-formation time observed in Fig. 1. Apparently, the *temperature increase in the system originated from the thermal conduction alone cannot account for the diffraction rings observed*.

In order to explore the physics for the observed nonlinear phenomenon, we measured the time dependence of output power. In Fig. 2, the total transmission P_t , which is equal to the ratio of the output power to input power, plotted against time in zero field. The increase of P_t with time, we believe, is due to the change in the number of particles cross the light beam. We realize that the laser intensity has the following spatial dependence: $I(r, z) = I_0 f(z) \exp(-r^2/w^2)$, where $f(z) = \exp(-\sigma z)$, the temperature inside the sample will also have a spatial distribution with the highest temperature at the optical axis along the radial direction and somewhere below the center of the cell along the z direction. This temperature gradient should lead to thermophoresis—particle migration along the temperature gradient, which was first described by Tyndall³ in dust particles in gases. There are several mechanisms for thermophoresis.⁴ For small particles, the thermal diffusion of particles due to the temperature gradient in the carrier dominates. We found that, by assuming the thermal diffusion mechanism, we could produce main feature of our experimental results. Taking both the thermal and mass diffusion into account, the mass flux obeys the following equation:^{5,6}

$$\frac{\partial c}{\partial t} = D \Delta c + D_T \nabla [c(1-c) \nabla T], \quad (2)$$

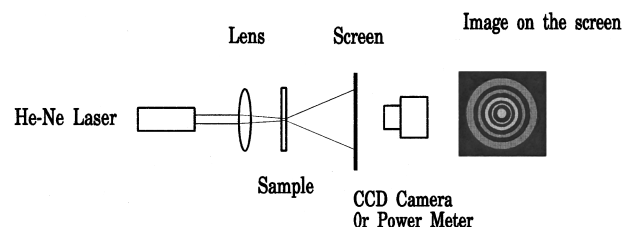


FIG. 1. The schematics of experimental setup. The right side is the diffraction rings observed in far field.

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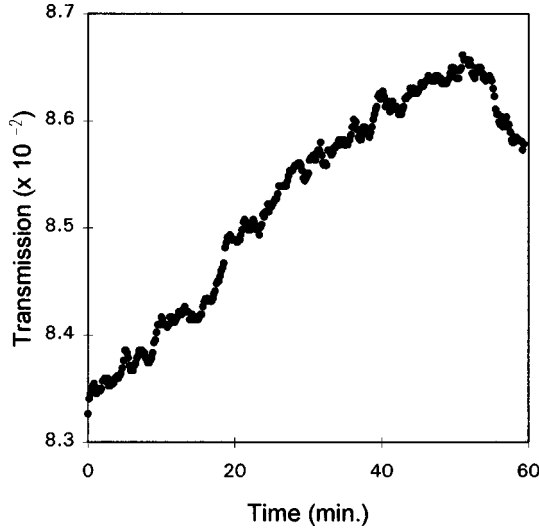


FIG. 2. The transmission vs time in zero applied magnetic field without the focal lens.

where c is the concentration of particles, T the temperature, D and D_T the mass and the thermal diffusion constants, respectively. $D = kT/6\pi\eta R$ with η the viscosity of the solvent, R the hydrodynamics radius which is twice of the particle radius. $\eta = 3 \times 10^{-3}$ Pa s for our ferrofluid. We get $D = 7 \times 10^{-8}$ cm²/s. In Eq. (2), D_T is the thermal diffusion coefficient with the unit of cm²/s K. From our experiment, we could do an order of magnitude estimate for D_T . The time dependence of the transmission, P_t , in Fig. 2 indicates that the particles migrate away from the optical axis, thus the increase in P_t , until a maximum is reached at t_{\max} . The decreasing of P_t with time for $t > t_{\max}$ suggests reverse flow of the particles towards the center of the light beam. We found that peak position t_{\max} shortens with increasing light intensity. For example, using a lens with a focal length of 1 cm will increase the intensity from 1 W/cm² for the unfocused beam to 10⁴ W/cm². This increase in intensity leads to a shift in the peak position in Fig. 2 towards shorter time.⁷ The increase in the transmission with time in Fig. 2 thus should be associated with the thermal diffusion of particles away from the optical axis. As time goes on, other processes will become increasingly important that will be addressed elsewhere. Furthermore, the temperature gradient cross the light beam will decrease. These mechanisms will reduce the thermal diffusion of particles. Therefore, we believe that it is reasonable to take the time to reach t_{\max} as the thermal diffusion time τ_{th} that satisfies: $D_T t_{\max} = r^2/T$, where r is the length scale particle traveled within time t_{\max} . Taking parameters from data in Fig. 2, i.e., the radius of laser spot $r \sim 1.5$ mm, and $t_{\max} = 3000$ s, we found $D_T \sim 2 \times 10^{-8}$ cm²/s K. In binary liquids, the thermal diffusion of species is referred as the Ludwig–Soret effect.⁸

Equations (1) and (2) are coupled through the absorption coefficient that is a function of the concentration c . We solve them numerically with the following boundary conditions: (a) temperatures at the boundaries are continuous, i.e.,

$$T(r=R) = T_0, \quad T(z=h+L) = T(z=-L) = T_0, \quad (3)$$

where R is the sample size from the center of the laser beam to the edge, h and L the thicknesses of the sample layer and

that of the substrates (glass plates), respectively, T_0 the room temperature. (b) Heat fluxes at the interfaces between the sample and the substrates are continuous:

$$\kappa_0 \left(\frac{\partial T_0}{\partial z} \right)_h = \kappa_1 \left(\frac{\partial T_1}{\partial z} \right)_h, \quad \kappa_0 \left(\frac{\partial T_0}{\partial z} \right)_0 = \kappa_1 \left(\frac{\partial T_1}{\partial z} \right)_0, \quad (4)$$

where $\kappa_0 = 7 \times 10^2$ W/mK and $\kappa_1 = 0.15$ W/mK are thermo-conductivities of glass and the ferrofluid, respectively; (c) At $t=0$, the temperature and concentration are uniform in space. We obtained temperature change $T(r,t) - T_0$ and relative concentration change $\Delta c(r,t) = (c - c_0)/c_0$ as functions of time shown in Figs. 3(a) and 3(c). We found that, at the steady state, the temperature change is 12 K within the laser beam ($r=0$ to $r/w=1$) and about 50 K to the boundary of the sample cell ($r \rightarrow \infty$) in the horizontal plane. We can see that temperature has a sudden increase at very short time (< 30 ms) after the laser is switched on but percentage change is much smaller for $t > 0.2$ s. In contrast, the concentration change, $\Delta c(r,t)$ is much slower at the short time as shown in Fig. 3(c) due to the slower responses of particle migration. Using the similar criterion for estimating the thermal conduction time, we found that characteristic time scales for both thermal and mass diffusions are in the order of 1 s. From Fig. 3(c) we can see that the concentration decreases as much as 70% on the optical axis when it reaches the steady state. This change in both temperature and concentration leads to the change in the local index of refraction $\delta n(r,z)$: $\delta n(r,z) = \delta n(r,z)/\delta T \delta T + \delta n(r,z)/\delta c \delta c$. Because $\delta n/\delta T < 0$, $\delta T > 0$ and $\delta n/\delta c > 0$, $\delta c < 0$, the results from both the thermal conduction and the thermal diffusion are *self-defocusing*. The phase change originated from δn seen by the laser beam traversing the medium of thickness h is

$$\delta\phi(r) = \frac{2\pi}{\lambda} \int_{-h/2}^{h/2} \delta n(r,z) dz,$$

λ is the wavelength of the light. Using the number of diffraction rings, N , from the experiment, we can estimate the maximum of $\delta\phi$ by: $\delta\phi_0 = 2\pi N = 2\pi h \delta n/\lambda$, where h is the thickness of sample layer and δn the corresponding change in the index of refraction. For $\lambda = 0.6328$ μm and $h = 50$ μm , we get $\delta n = 5 \times 10^{-2}$ for $N = 4$. The index of refraction is equal to: $n = n_0 + n_2 E^2$, E is the electric field and $|E|^2$ is proportional to the intensity of the light. Then the nonlinear part of the index of refraction can be obtained: $n_2 = \delta n/|E|^2 \sim 10^{-1}$ cm³/erg. This large change in the refractive index is larger than that of some liquid crystals.^{9,10}

The far-field interference patterns can be obtained by using the local phase change from the numerical calculation.¹¹ We plotted results in Figs. 3(b) and 3(d). In these calculations, the sample thickness is 50 μm and the focal length is 1 cm. Figure 3(b) represents diffraction at the far field at time $t = 1$ ms. At this early stage, thermal and mass diffusions are not appreciable. Apparently, the phase change due to temperature alone is small and the diffraction pattern is essentially invisible. Figure 3(d) shows the diffraction pattern at the far field for $t = 1$ s. Within this time, particle density has an obvious change as illustrated in Fig. 3(c) and reaches the saturation as time reaches the order of 1 s, agreeing with the ring-formation time in our experiment. To

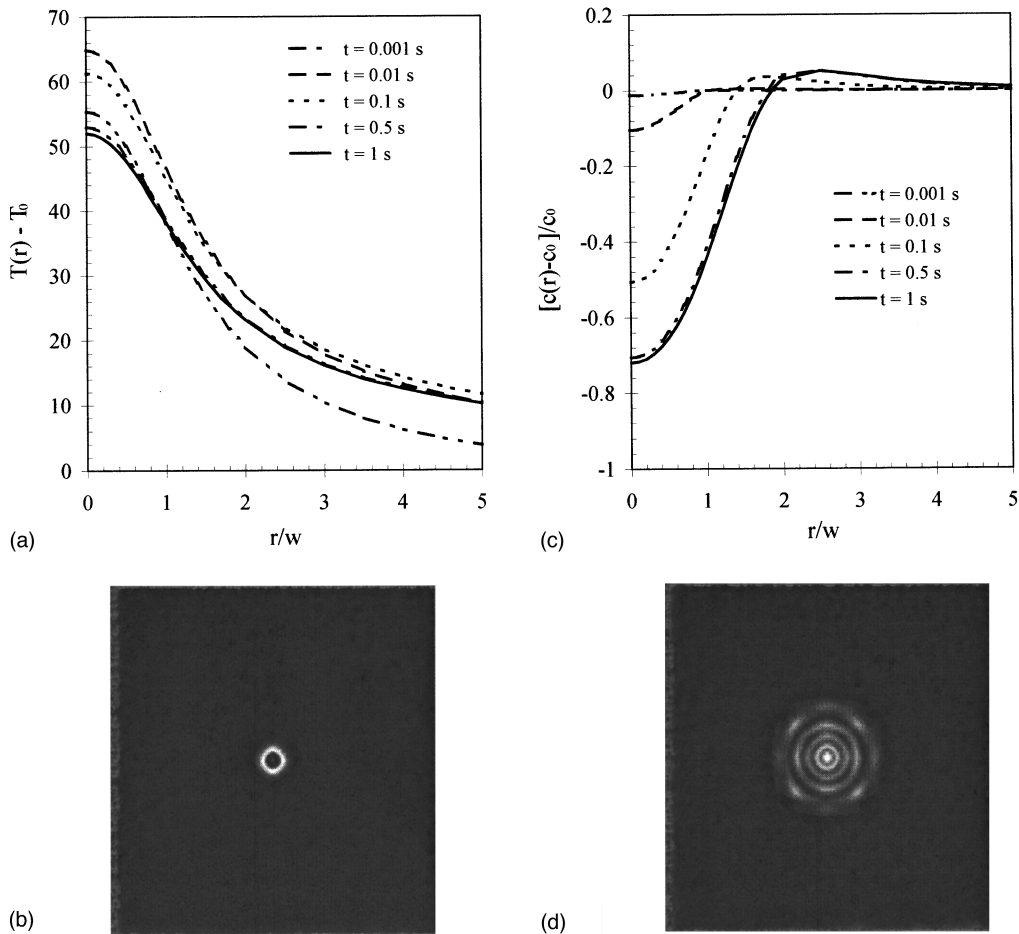


FIG. 3. Results from numerical calculations. (a) Temperature change as a function of radial distance, r/w , for different times, where w is the width of the laser beam. (b) Far field diffraction at short time, when only temperature change is appreciable. The small ring forms in less than a millisecond that is unobservable in our experiment. (c) The change in the particle concentration as a function of reduced radial distance, r/w , for different times. (d) For longer time, the change in the index of refraction due to concentration decrease becomes more important. This change leads to multiple rings in far field at $t = 1$ s.

reproduce the number of rings observed in experiment under the same condition, we need the thermal diffusion constant, $D_T \sim 1 \times 10^{-7}$ cm²/s K, 5 times larger than the experimental value, $D_T \sim 2 \times 10^{-8}$ cm²/s K obtained from Fig. 2. Apparently, to compare with experiment quantitatively, we have to take into account other processes in the future. When thermal dissipation through the substrates is neglected, the temperature increase in the sample will be around 10^3 K without the thermal diffusion. This huge temperature change will produce “thermal rings.” Then the particle migration will only change the distribution of light intensity among the rings, but not the number of rings.¹²

Because particles have permanent magnetic moments, there is an interaction between the ferrofluid and an applied magnetic field. We found that when the external field is switched on, the transmission has a dramatic increase in a short time. This effect is not understood yet and will be published elsewhere.¹³

In summary, NLO properties in a ferrofluid were studied by both experiment and numerical calculations. We have demonstrated that the cross-coupling between the concentration of particles and the temperature of the system, which originates from the thermophoresis, is likely the main mechanism for the NLO effects. A magnetic field enhances the NLO effect dramatically. These results should lead to a

new type of nonlinear optical materials for device applications.

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¹¹Detailed calculation will be submitted elsewhere by T. Du and W. Luo (unpublished).

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