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Observation of the dynamics of magnetic nanoparticles induced by a focused laser beam by using dark-field microscopy



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ABSTRACT

The dynamics of Fe_3O_4 magnetic nanoparticles under the irradiation of a tightly focused laser beam was investigated by using a high-intensity dark-field microscopy. A depletion region of magnetic nanoparticles was found at the center of the laser beam where the dissipative force (absorption and scattering forces) dominated the dynamics of the magnetic nanoparticles. In contrast, the dynamics of magnetic nanoparticles was dominated by thermal and mass diffusions at the edge of the laser beam where the dissipative force was negligible. In addition, the transient variation in the concentration of magnetic nanoparticles was characterized by recording the transient scattering light intensity. The coefficients of thermal diffusion, mass diffusion and the Soret effect for this kind of magnetic nanoparticles were successfully extracted by using this technique.

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1. Introduction

In the past decades, Magneto-optical effects of magnetic fluids have attracted great interest due to the potential applications in the construction of diverse functional devices such as magnetic field sensors [1–3], tunable gratings [4–6], optical filters [7,8], modulators [9-11], and switches [12,13]. Recently, a new technique that can be used to realize multi-sized nanospheres assembly or to form ordered three-dimensional optical crystals in magnetic fluids under magnetic fields has been reported [14-17]. Based on this technique, we have proposed a completely different method to assemble silica microspheres suspended in magnetic colloid into a three-dimensional crystal by utilizing the giant nonequilibrium depletion force induced by a single tightly focused laser beam [18]. The physical mechanism responsible for these phenomena is ascribed to the interaction of magnetic nanoparticles (MNPs) suspended in magnetic fluids with the tightly focused laser beam.

Early in 1998, Tabiryan et al. theoretically studied the interactions of MNPs with a laser light and experimentally investigated the Soret feedback effect in the thermal diffusion of MNPs by monitoring the transmission of the laser light [19]. Meanwhile, Lenglet et al. showed that the forced Rayleigh scattering could be used as an experimental tool to study the thermal dynamics of

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http://dx.doi.org/10.1016/j.jmmm.2014.03.080 0304-8853/© 2014 Elsevier B.V. All rights reserved. MNPs suspended in colloids and to measure the Soret coefficient of MNPs covered with different surfactants [20]. Since the particle size of MNPs is generally in the range of several and several tens of nanometers, the direct observation of the dynamics of MNPs under the irradiation of tightly focused laser beam remains to be a challenge. In addition, the effect on the dynamics of MNPs induced by optical forces (e.g. scattering and absorbing forces) were not taken into account for simplicity in previous reports [19,20].

In this article, the dynamics of Fe_3O_4 MNPs under the irradiation of focused laser beam was investigated by using dark-field microscopy. It is shown that a simple description of the interaction of focused laser beams and complex media based on thermal diffusion and mass diffusion is not adequate in many cases. It is revealed that the dynamics of MNPs is dominated by different mechanisms when MNPs are located at different areas of the focus laser beam. In addition, it is demonstrated that the Soret coefficient of MNPs can be extracted by recording the transient scattering light intensity.

2. Theory

The dynamics of MNPs after considering both mass and thermal diffusions can be written as follows [19]:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial r^2} - \frac{\sigma}{\chi \rho C_P} D_T c(1-c) I(r), \tag{1}$$

where *c* denotes the concentration of MNPs; *D* and *D*_T are the coefficients of mass diffusion and thermal diffusion, respectively; χ is the thermal conductivity coefficient; *I*(*r*) is the intensity of the laser beam as a function of the displacement *r* from the laser spot center, ρC_p is the heat capacitance of the MNPs, and σ is the absorption constant on the concentration of the MNPs. For small *c*, it can be simplified as

$$\sigma = \alpha c, \tag{2}$$

where α is a constant. In this case, Eq. (1) can be rewritten as

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial r^2} - \frac{\alpha}{\chi \rho c_P} D_T c^2 (1 - c) I(r).$$
(3)

Generally speaking, the mass diffusion of MNPs is induced by the thermal diffusion of MNPs, which in turn weakens the thermal diffusion. Thus, Eq. (3) can be further simplified by the process dominating the dynamics of MNPs. When the laser light is turned on, the thermal diffusion dominates the dynamics of MNPs while the mass diffusion can be neglected. Eq. (3) can be simplified as

$$\frac{\partial c}{\partial t} = -D_T c^2 (1-c) \frac{\alpha I}{\chi \rho c_P} = \mu D_T c^2 (1-c) I(r), \tag{4}$$

where, $\mu = -(\alpha/\chi\rho c_P)$. Integration of Eq. (4) yields

$$-\frac{1}{c} + \ln \frac{c}{1-c} = \mu D_T I(r)t + C_{cst} \otimes \frac{c}{1-c} \exp(-1/c) = A_0 \exp[\mu D_T I(r)t],$$
(5)

where C_{cst} is the integration constant and we set $A_0 = \exp(C_{cst})$. By substituting $c(t = 0, r) = c_0$ in Eq. (5), we can get

$$C_{cst} = -\frac{1}{c_0} + \ln \frac{c_0}{1 - c_0},\tag{6}$$

as shown in Eq. (5), the concentration of MNPs decreases exponentially with time after the laser light is turned on. If the exponential decay time t_1 , which is defined as the time at which the concentration of MNPs is reduced to 1/e of its initial value, can be extracted from the experimental results, then one can derive the thermal diffusion constant D_T of MNPs by

$$t_1 = -\frac{1}{\mu D_T l(r)} \Rightarrow D_T = -\frac{1}{\mu l(r)t_1}.$$
 (7)

In the experiment, the scattering light intensity of MNPs can be used to characterize the concentration of MNPs. By recording the transient variation of scattering light intensity of MNPs, t_1 can be easily extracted by fitting the scattering light intensity with exponential decay.

As reported by Lenglet et al. [19], the temperature gradient formed by the absorption of laser energy will vanish when MNPs move to the boundary of the laser spot due to thermal diffusion effect. In this case, the dynamics of MNPs is dominated by mass diffusion and some MNPs will move back to the laser spot center. Thus, Eq. (3) is simplified as

$$\partial c/\partial t = D\Delta c.$$
 (8)

Here, our model considers only the two-dimensional (2D) case, the temperature gradient and the attenuation of the laser light intensity along the propagation direction are neglected. This assumption is reasonable and it allows us to capture the key feature of the dynamics of MNPs. Thus, we have

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial r^2}.$$
(9)

here, $c(0, r) = c_0(r)$, $c(t, 0) = p_0$ and $\lim_{r \to R} c(r, t) = c_0$. The solution of Eq. (9) is

$$c(r,t) = A\lambda_n \exp(\lambda_n Dt) \sum_{1}^{\infty} B_n \sin(\sqrt{-\lambda_n}r)$$

$$= \sum_{1}^{\infty} \left\{ C_n \exp[(-\pi^2 n^2 D t)/R^2] \sin\left[\frac{\pi n r}{R}\right] \right\},\tag{10}$$

here

$$c(r, 0) = \sum_{1}^{\infty} C_n \sin \left[(\pi n r) / R \right],$$

and

$$C_n = \frac{2}{R} \int_0^R c_0(r) \sin\left[\frac{\pi}{R}nr\right] dr$$

here, R denotes the size of the sample cell. For the right-side of Eq. (10), it is sufficient to consider the first term of the summation and one obtains

$$c(r,t) = C_1 \exp\left(-\frac{\pi^2 Dt}{R^2}\right) \sin\left(\frac{\pi}{R}r\right).$$
(11)

The concentration of MNPs varies exponentially with time. By recording the scattering light intensity of MNPs, the time constant t_2 for the mass diffusion of MNPs can be easily extracted by fitting the time-dependent scattering light intensity. Thus, we can derive the thermal diffusion constant *D* of MNPs by

$$t_2 = -\frac{R^2}{\pi^2 D} \quad \Rightarrow \quad D = -\frac{R^2}{\pi^2 t_2}.$$
 (12)

3. Sample preparation and experimental details

The magnetic colloid used in our study was water-based Fe₃O₄ fabricated by the chemical coprecipitation technique (Central Iron and Steel Research Institute, China). The average diameter of MNPs was measured to be ~12 nm and the weight fraction of MNPs was determined to be 25.7%. In our experiments, the magnetic fluid was first diluted with water and the volume concentration of MNPs was 1.75×10^{16} cm⁻³. Then the magnetic colloid was sonicated for half an hour and sealed into a sample cell with a thickness of 50 µm.

The 532-nm laser light from a solid-state laser (Verdi-5, Coherent) was focused on the sample cell by using a $63 \times$ objective lens (NA=1.43) and the power of laser light was chosen to be 8 mW, as schematically shown in Fig. 1. The dynamics of MNPs was monitored by using an inverted dark-field microscopy (Axio Observer A1, Zeiss) in combination with a charge-coupled device (CCD). The concentration of MNPs was characterized by the scattering light intensity and 500 images of the scattering light were captured with the continuous capture mode of the CCD after the laser light had been turned on. The exposure time for each image was set to be 200 ms.



Fig.1. Schematic showing the experimental setup used to investigate the dynamics of MNPs under the irradiation of focused laser light. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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