Evolution of Nonlinear Optical Characteristics of Magnetic Nanoparticle Colloidal Suspensions after Laser-Induced Clusters

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ABSTRACT: In this paper, the nonlinear optical properties of magnetic nanoparticle colloidal solutions were studied by the Z-scan technique using 25 ps laser pulses at a wavelength of 1064 nm. Our results reveal that the formed magnetic nanoparticle clusters under high incident laser intensity will greatly affect the nonlinear optical characteristics of the solution. As the intensity of the pulsed laser decreases, the reverse saturable absorption coefficient and nonlinear refractive index of the sample tend to increase. The evolution of this nonlinear characteristic only occurs in liquid suspension. This is confirmed by fixing particles on a substrate upon which the responses observed in the liquid medium are no longer present. Besides, the possibility of generating optical trapping in the focus of the laser pulses is proposed to explain our experimental results.

1. INTRODUCTION

In the past years, nonlinear optical materials have made significant development in laser technology. Various kinds of nonlinear effects arise from the interaction between laser and nanoparticles. The search for these materials is one of the defining characteristics of nonlinear optics.1

Recently, nanoparticles suspended in a solution have attracted much attention. As we know, liquid suspensions of nanoparticles can show large optical nonlinearities.2 This has been amply demonstrated in a variety of experiments including optical limiting,3 Mie scattering,4 optical spatial soliton propagation,5,6 and nonlinear optical absorption and refraction.7 In addition to chemical synthesis methods, laser ablation also provides a new method to prepare nanoparticle colloidal solutions.8–11 With the help of new technologies, different kinds of nanoparticle colloidal solutions were prepared for research in the field of nonlinear optics. Two factors may affect the nonlinear optical characteristics of the colloidal solutions: first, the properties of the material from which the nanoparticles are made; second, the influences of Mie scattering, thermal lens effects, optical forces, and other factors in solutions.12–14 It is possible to achieve large enhancements of nonlinear optical parameters by changing the size and shape of the nanoparticles.15 The interaction between laser and nanoparticle colloidal solutions will open up a new field of research.

Ferrofluids are colloidal suspensions of Fe3O4 nanoparticles in a liquid carrier. As a transition metal oxide, Fe3O4 nanoparticles have a large third-order optical nonlinear magnetic χ(3) and fast nonlinear optical response.16–18 The recovery time of particles is about 18–30 ps.19 In addition, the nonlinear optical properties of Fe3O4 nanoparticle suspension were also studied, including two-photon absorption,20,21 optical limiting,22 and nonlinear scattering.23 Therefore, ferrofluids are known to be a promising nonlinear optical material. However, there is no systematic study on the effect of self-assembled clusters of magnetic nanoparticles under irradiation of the pulsed laser.

In this paper, we present our recent work on evolution of nonlinear optical characteristics of magnetic nanoparticle colloidal suspensions after forming laser-induced clusters. The nonlinear characteristics of nanoparticle colloidal solutions are largely determined by the intensity and sequence of the incident laser. To verify that nanoparticle clusters can only be formed in
liquid suspensions, the nonlinear absorption coefficient and nonlinear refractive index of solid films were compared with that in suspensions. Besides, the role of the laser trap is used to explain the formation of nanoparticle clusters.

2. RESULTS AND DISCUSSION

Water-based ferromagnetic fluid was selected to prepare Fe₃O₄ nanoparticle colloid solutions in our experiment. Different types of surfactants were used for protecting the Fe₃O₄ nanoparticles from self-aggregation. The relatively low viscosity of water facilitates the movement of particles under the action of light.

The X-ray spectrum of the Fe₃O₄ nanoparticles is shown in Figure 1. The peaks at 2θ of 30.1, 35.5, 43.2, 53.6, 56.9, and 62.6 degree are assigned as the X-ray diffraction from (220), (311), (400), (422), (511), and (440) planes, respectively.

The TEM image (Figure 2a) shows the detailed structure of the Fe₃O₄ nanoparticles. Statistical analysis (in Figure 2a) of the image revealed a normal distribution of particles with a mean size of 11 nm. The ferrofluid was diluted to different concentrations and placed in a quartz colorimetric dish with a 1 mm path length, as shown in Figure 2b. The concentrations of the two samples were 59 mg/mL (sample A) and 11.8 mg/mL (sample B), respectively.

Furthermore, the absorption spectra of samples A (black curve) and B (red curve) are shown in Figure 3. The absorption of sample B is almost negligible after 800 nm. The small values of nonlinear optical parameters of the sample were caused by the low concentration of Fe₃O₄ nanoparticles. Therefore, sample B was selected in the following experiment.

The next step is to study the third-order optical nonlinearities of magnetic colloid solutions, including nonlinear absorption and nonlinear refraction. In our experiment, all data were measured by the Z-scan system, which is similar to that in ref 24. The pump source is a mode-locked Nd:YLF laser with a wavelength of 1064 nm and a pulse width of 25 ps. The pulse repetition rate was set to be 1000 Hz. We also verified that the quartz colorimetric dish containing water and surfactant did not show any nonlinear response under the same experimental conditions. All these data were measured at room temperature (300 K).

The on-axis peak intensity at focus varies from \( I₀ = 0.1 \text{ GW/cm}^2 \) to \( I₀ = 15 \text{ GW/cm}^2 \), and the beam waist is \( ω₀ = 45 \mu\text{m} \).

Figure 4 shows the open aperture (OA) Z-scan result of sample A at a density of 12.2 GW/cm² in the focal plane.

Figure 1. X-ray spectrum of Fe₃O₄ nanoparticles corresponds to the standard JCPDS card no. [19-0629].

Figure 2. (a) Transmission electron micrograph (TEM) image of Fe₃O₄ nanoparticles. The result of the particle size distribution is shown in (a). (b) Different concentration of the sample was placed in quartz plates.

Figure 3. Evolution of the optical absorption spectra of two samples.

Figure 4. Open-aperture (OA) Z-scan result at a density of 12.2 GW/cm² in the focal plane.
B with higher incident excitation intensity at the focal point (12.2 GW/cm²). It is observed that there was a sudden decrease in the transmittance when the sample was located near the focal point. The transmission dropped dramatically to 10%, almost opaque. While this open curve has no symmetry, it cannot simply be explained by the nonlinear absorption process but also due to the effect of magnetic nanoparticle clusters.

Figure 5a shows typical open aperture Z-scan results of the Fe₃O₄ nanoparticle colloid solutions with different excitation energy densities at the focal point. The corresponding energy densities are 8.7 GW/cm² (1st), 8.7 GW/cm² (2nd), 0.1 GW/cm² (3rd), and 0.1 GW/cm² (4th), respectively.

At the beginning of the experiment, two identical densities (8.7 GW/cm²) were sequentially applied to the sample. The transmission at focus drops to 30% under the second-incident power density of I₀ = 8.7 GW/cm². Under two laser excitations of the same energy density (8.7 GW/cm²), the influence of clusters on the nonlinear characteristics of colloidal solution was increasing. To explore the stability of clusters, a very low excitation energy density (0.1 GW/cm²) was used to scan the sample at the same location as the third and fourth Z-scans. In Figure 5a, it can be clearly seen that when the incident energy was reduced to 0.1 GW/cm² the nonlinear characteristics of the suspension still exist.

The plot of the normalized transmittance at focus as the function of time is shown in Figure 5b. The normalized transmittance decreases from 95 to 30% when two densities (8.7GW/cm²) were sequentially applied to the sample. The interval between two excitations is 10 min. Since the third and fourth excitation densities (0.1GW/cm²) are very low, such a low energy density is not enough to maintain the continued existence of nanoparticle clusters. The transmittance increases from 40 to 70% under the same excitation energy density (0.1 GW/cm²). Clusters gradually disappear. Figure 5c shows that there is no nonlinear phenomenon under the excitation energy density of I₀ = 0.1 GW/cm² after the sample position changed.

After changing the position of the sample, the variation laws of the nonlinear variation with lower incident power were compared. Figure 6a shows the Z-scan results at the same position of the sample measured with three laser energy densities of 6.8, 6.2, and 2.8 GW/cm². As the excitation energy of the laser decreases, the transmission dropped to 95, 90, and 85%, respectively. The above results suggest that the formation process of nanoclusters can be controlled by adjusting the energy density of the incident laser and the incident laser sequence. By changing the sample position again, the incident energy density (2.8 GW/cm²) is no longer able to excite the nonlinear characteristics of the sample and is shown in Figure 6b.

In the meantime, the nonlinear refraction properties of the sample were also investigated. The energy density corresponds to the density of the open-aperture curves (Figure 6a). Several physical mechanisms contributed to the nonlinear index of refraction, such as electronic polarization, electrostriction, population redistribution, and so on. It has been proven that the distortion of the electron cloud by the optical field is the...
main source of the nonlinear optical (NLO) refraction of noble metal nanocomposites. However, the source of the nonlinear optical (NLO) refraction of magnetic nanoparticle colloidal suspensions has not been studied in depth. Figure 7a shows the closed-aperture results of the sample under different pulse energies. The appearance of valley-peak shapes in Figure 7a means that the sample has a self-focusing effect. As the intensity of the incident light was changed from high to low, the distance between the peaks and valleys of the closed-aperture curve increased. This distance is an important parameter for calculating the nonlinear refractive index of a material. Figure 7b shows that once the excitation position on the sample is changed the incident energy density (2.8 GW/cm²) cannot excite the nonlinear characteristics of the sample.

The nonlinear absorption coefficient and refractive index of the film were measured for comparison. (thickness = 15 um, concentration Φ = 59 mg/mL). After the Fe₃O₄ nanoparticles were dried on the thin film (the movement of particles was frozen), the phenomena in liquid samples could no longer be observed. The corresponding nonlinear absorption coefficient and refractive index were entirely determined by the properties of the Fe₃O₄ nanoparticles themselves. Moreover, the conclusion is similar to other literature.

Eqs 1 and 2 were used to fit the $T_N(z)$ and $T_{NO}^{CA}(z)$ data to obtain the nonlinear absorption coefficient $\beta$ and nonlinear refractive index $n_2$, respectively:

$$T_N(z) = 1 - \frac{\beta I_0 z_{eff}}{2\sqrt{2}(1 + x^2)}$$ (1)

$$T_{NO}^{CA}(z) = 1 + \frac{4\eta_2 k I_0 z_{eff} x}{(1 + x^2)(9 + x^2)}$$ (2)

where $x = z / z_{02}$, $n_2$ is the nonlinear refractive index, $I_0$ is the on-axis peak intensity at the focus, $L_{eff} = (1 - e^{-\alpha L})/\alpha$, $L_{eff}$ is the effective interaction length, $\alpha$ is the linear absorption coefficient, $z$ is the longitudinal displacement of the sample from the focus ($z = 0$), $L$ is the sample length, and $z_{02}$ is Rayleigh diffraction length. $T_{NO}^{CA}$ is the normalized transmittance for open-aperture (OA) results. $T_{NO}^{CA}$ is the normalized transmittance for closed-aperture (CA) results.

Table 1 below shows the calculated data obtained by combining the Z-scan eqs 1 and 2 based on the curves in Figure 6a, Figure 7a, and Figure 8. At the bottom, the table shows the Z-scan data for the thin film in comparison with the liquid sample.

Table 1. Nonlinear Optical Parameters of Nanoparticle Colloidal Solution and Thin Film

| Solution          | $I_0$ (GW/cm²) | $\beta$ (cm/GW) | $n_2$ (10⁻²⁰ cm²/W) |
|--------------------|----------------|-----------------|----------------------|
| 1st                | 6.8            | 0.21            | 2.72                 |
| 2nd                | 6.2            | 0.46            | 6.81                 |
| 3rd                | 2.8            | 1.52            | 21.68                |
| Thin film          | 4.3            | 0.34            | 9.6                  |

excitation ($I_0 = 6.8$ GW/cm²) with a value of ~0.21 cm/GW. The same comparison is also applicable to the nonlinear refractive index of the sample. With the decrease of the excitation energy density from $I_0 = 6.8$ GW/cm² to $I_0 = 2.8$ GW/cm², the nonlinear refractive index of the sample increased from ~2.72 × 10⁻¹⁴ to ~2.168 × 10⁻¹⁴ cm²/W, which is about 8 times larger.

At the bottom of Table 1, the nonlinear parameters of the film were calculated to be $\beta = 0.34$ cm/GW and $n_2 = 9.6 \times 10^{-14}$ cm²/W. As a comparison, we find that the excitation energy density of the film ($I_0 = 4.3$ GW/cm²) is greater than that of the liquid ($I_0 = 2.8$ GW/cm²); the nonlinear coefficient is much smaller than that of the liquid sample (Table 1). Once the nanoparticles were fixed on the film, particle clusters cannot form under the action of laser, and the nonlinear characteristics of the film will not evolve similarly like liquid samples.

To understand the evolution of nonlinear characteristics of liquid samples, a comparison is diagrammed in Figure 9. As the excitation energy density at the focal point changes from high to low, the characteristic data of the liquid sample increase. This abnormal nonlinear optical characteristic evolution is due to the influence of nanoparticle clusters.

According to experimental results, it is found that there are three processes that affect the nonlinear properties of nanoparticle solutions. The schematic diagrams of these processes are shown in Figure 10a–c as follows:

(i) The nanoparticles are dispersed in the colloidal solution.

The concentration of the colloidal solution is low, so there are nonlinear characteristics under lower power excitation energy density, as shown in Figures 5c, 6b, and 7b.

(ii) As laser power becomes higher, nanoparticles start to approach each other. At this stage, the nonlinear optical characteristics dominated by Fe₃O₄ nanoparticles can be obtained in the Z-scan curves like Figure 5a (1st), 6a (1st), and 7a (1st).

(iii) Higher laser power produces more compact assemblies consisting of nanoparticles. Clusters are formed at this
stage, resulting in more complex nonlinear optical characteristics of the colloidal solution. The Z-scan curves are affected by the clusters of nanoparticles, as shown in Figure 5a (3rd), 6a (3rd), and 7a (3rd).

Figure 10d shows that in the case of low power laser energy density the nanoparticles are only constrained in the propagation direction of the Gaussian beam with the help of the optical trapping. Under the action of high power laser energy density, the optical trapping is considered to be stable. Clusters of nanoparticle will appear at the focal point when the potential trapping well is deep enough, as shown in Figure 10e. With the influence of nanoparticle clusters, the Z-scan curves become more complicated, including the influence of nonlinear absorption and nonlinear scattering, etc.

The formation process of nanoparticle clusters is shown in Figure 10.

According to optical tweezers theory, the trapping potential of the particles was calculated and compared with the thermal energy, as show in eqs 3 and 4

\[ U_{\text{trap}} = -\pi \epsilon_0 \epsilon_r R_p^3 \left( \frac{\epsilon'_p - \epsilon_m}{\epsilon'_p + 2\epsilon_m} \right) \text{Re}(\mathbf{E} \cdot \mathbf{E}) \]  

(3)

The relationship between light intensity and electric field intensity can be determined by eq 4

\[ I_0 = \frac{1}{2} \epsilon_0 \eta_0 E_0^2 \]  

(4)

where \( R_p \) is the particle radius, \( \epsilon'_p \) is the real part of the particle dielectric permittivity, and \( \epsilon_m \) is the dielectric permittivity of the ambient medium that is assumed non-absorbing, \( I_0 = 10 \text{ GW/cm}^2 \). The thermal energy: \( k_B T = 4.14 \times 10^{-21} \text{ J} \) (room temperature is 300 K). The lower limit of the trapping potential.
magnitude for our system can be readily estimated given the intensity of the focused Gaussian. The energy comparison is shown in Figure 11.

Figure 11. Curve of the trapping potential as the size of the nanoparticle changes. The line is the thermal energy (negative for comparison purposes).

3. CONCLUSIONS

In this work, we have studied the evolution of nonlinear optical properties of magnetic nanoparticle colloidal suspensions after laser-induced clustering. Results reveal that the magnetic nanoparticle clusters formed in the potential trapping well have obvious optical nonlinear evolutionary characteristics, which can be reflected in the change of the open and closed aperture curves. The reverse saturable absorption coefficient and nonlinear refractive index of the colloidal suspensions were measured and compared with the film sample. In addition to the Z-scan results, a laser potential trapping well model is used to explain the formation of nanoparticle clusters. At the same time, the cluster process of magnetic nanoparticles under pulsed laser needs further study.

4. EXPERIMENTAL SECTION

Water-based ferrofluids were prepared in a laboratory by using a previously reported method. The colloidal solution was made by diluting a water-based ferrofluid. Corresponding surfactant and deionized water were added during the dilution process. Samples were placed in a 1 mm-thick quartz cuvette for Z-scan experiments. The pump source was a mode-locked Nd:YLF laser with a wavelength of 1064 nm and a pulse width of 25 ps. The experiments were carried out with a wavelength of 1064 nm and a pulse width of 25 ps. The on-axis peak intensity for our system can be readily estimated given the intensity of the focused Gaussian. The energy comparison is shown in Figure 11.

Figure 11. Curve of the trapping potential as the size of the nanoparticle changes. The line is the thermal energy (negative for comparison purposes).

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Notes
The authors declare no competing financial interest.

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