Non-Linear Optical Studies of Colloidal Nanofluids

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Abstract

We report non-linear optical effects observed in colloidal nanofluids due to transmission of laser beam. A 10mW 632nm He-Ne Laser beam is passed through separate dispersions of magnetic and non-magnetic nanoparticles. Thermal lens induced diffraction patterns are observed in case of Fe_3O_4 nanoparticles dispersed in hexane and kerosene. In case of CdS nanoparticles dispersed in methanol, no such patterns were observed. This is useful in studying non-linear optical properties of nanofluids arising due to local heating of the medium by the laser establishing a refractive index gradient. Variation in non-linear optical properties is observed to be based on magnetic and non-magnetic nature of particles. In case of magnetic nanoparticle dispersions, the effect of magnetic field on the non-linear optical properties is also examined.

Introduction

Suspensions of nanoparticles in liquid media known as "nanofluids" have generated interest in researchers. These applications include wide areas ranging from mass transport, absorption and conversion of radiation, optics, consumer goods and catalysts[1-4]. The area of research in nanofluids is of interdisciplinary nature involving physics, chemistry, material science and their engineering and biomedical applications. In case of nanofluids, reduction in particle size resulting into higher surface area enhances thermal conduction and changes the optical properties compared to base fluid. Non-linear optical properties in nanofluids are studied to understand the effect of particle size, composition, concentration with respect to base fluid by using standard experimental techniques like z-scan.

Motivation for the present study arises from our past studies involving optical scattering, magneto-optic and thermal lensing phenomena in nano ferrofluid samples[5-8]. Optical transmission and optical scattering studies in nano ferrofluids have potential applications to develop sensors[9]. Optical methods provide non-invasive way to probe thermal properties of nanofluids. These studies have applications in nanofluid based optical filters, optical switches[10].

We report comparative study of thermal lensing phenomena in colloidal nanofluids especially considering magnetic and non-magnetic nature of particles. When laser passes through a fluid it undergoes a spatial phase modulation due to a phenomenon called as "Thermal Lensing". Laser beam undergoes self-focussing or defocussing based on a change in non-linear refractive index caused due to local heating by laser. In some cases, thermal lens induced diffraction patterns are reported. These include carbon nanotubes, metallic nanoparticles[11,12]. In case of a ferrofluid, formation of thermal lens induced diffraction patterns has been studied in presence of magnetic field as well[13-17].

Non-linear optical phenomena in nanofluids

The transmission of a laser beam through a section of fluid causing a refractive index gradient has been established theoretically[17]. The formation of thermal lensing induced diffraction patterns is shown in the fig. 1. Based on the intensity of incident laser beam, a refractive index gradient develops in the medium. The change in local refractive index determines self-defocussing or self-focussing of a laser beam. This local heating gives rise to temperature and concentration diffusion of particles. The local change in refractive index is given by,

$$\delta n(r,z) = \left(\frac{\partial n(r,z)}{\partial T}\right)\delta T + \left(\frac{\partial n(r,z)}{\partial c}\right)\delta c \quad (1)$$

In case of an applied magnetic field, there is an additional component influencing local change in refractive index. Hence the local change in refractive index is given by,

$$\delta n(r,z) = \left(\frac{\partial n(r,z)}{\partial T}\right)\delta T + \left(\frac{\partial n(r,z)}{\partial c}\right)\delta c + \left(\frac{\partial n(r,z)}{\partial H}\right)\delta H$$

(2)

Thermal conduction in the sample is given by,

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

(3)

where σ is the absorption coefficient of the ferrofluid that depends on the concentration of particles, I is the intensity of the laser beam, ρ and C_p the density and the heat capacity of the system and χ the thermal diffusivity. Concentration diffusion obeys the following equation,

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

(4)

where c is the concentration of particles, T is the temperature, D and D_T the mass and thermal diffusion constants respectively. Eqn. (3) and (4) play important role to establish temperature profile in the sample given by,

$$\Delta T(r,t) = \frac{Aw_o^2}{8k}\left\{ \exp i\left(-\frac{2r^2}{w_o^2}\right) - \exp i\left(-\frac{2r^2}{8\left(\frac{k}{\rho c_p}\right)t + w_o^2}\right)\right\}$$

(5)

Temperature distribution profile ΔT(r,t) is a function of radial distance from optical center r and time t respectively. In finite time this reaches a steady state solution,

$$\Delta T(r) = \frac{Aw_o^2}{8k}\left\{ \ln\left(\frac{2\gamma a^2}{w_o^2}\right) - \frac{2r^2}{w_o^2}\right\}$$

(6)

where γ and *a* are constants as discussed by Gordon et al[18]. This paper discusses the transient phenomenon in nanofluid samples based on eqn. (4) and eqn. (5).

Material and Methods

This paper discusses the results of a series of experiments carried out using three types of nanofluid. Two of the nanofluids comprise of Fe_3O_4 (40-60 nm) nanoparticles prepared by chemical co-precipitation method. The first sample using Fe_3O_4 is dispersed in hexane (30 mg/ml) while the second is dispersed in kerosene (30 mg/ml and 7 mg/ml) separately. The third sample is that of non-magnetic nanoparticles like CdS prepared by solvo-thermal method. Nanofluid dispersion is made using CdS (5-10 nm) in methanol (5 mg/ml). Two types of experiments were carried out using the setup. One set of experiments involved the study of transient phenomenon using manual shutter with and without magnetic field and the other was on the intensity dependence of thermal lens patterns.

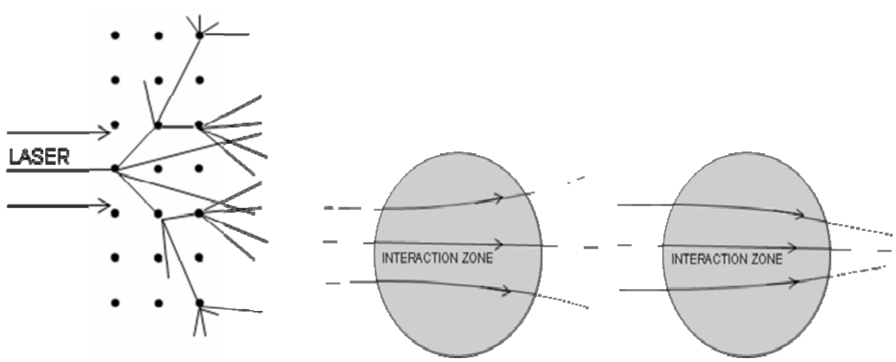

Figure 1: Formation of thermal lensing in a liquid

The setup consists of He-Ne laser (632 nm, 10 mW) beam which was passed through a cuvette (path length 1mm) containing a sample of the nanofluid. Schematic of setup is shown in fig. 2. The intensity dependent study was carried out by using a polarizer to vary the intensity of the incident beam. The time transient was studied by initiating the response using a manual shutter. Time transient study was carried out with and without magnetic field. In later case, a pulse of magnetic field (1.7kG) perpendicular to the beam was generated using electromagnet. This was carried out for Fe_3O_4 in hexane. For the samples of Fe_3O_4 in kerosene and CdS in methanol, a convex lens of 10cm focal length was used to focus the laser beam on the cuvette. Diffractions patterns formed on a screen were recorded using a CCD camera. Relevant images were extracted and processed using open source software packages Virtualdub and ImageJ.

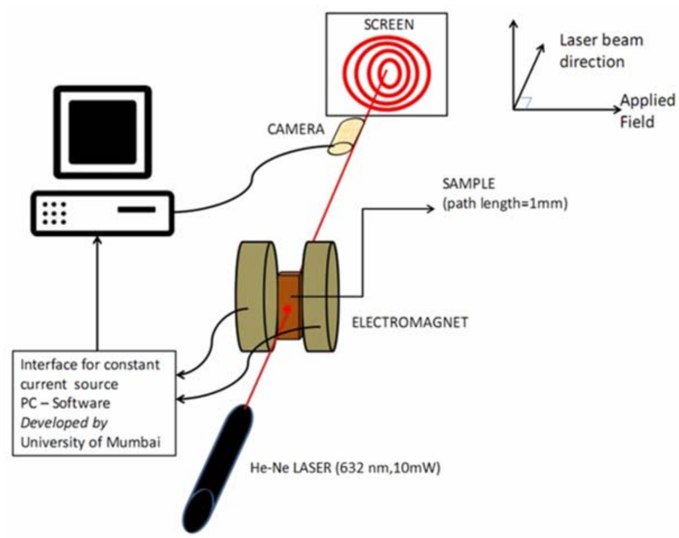

Figure 2: Schematic of the setup

Results and Discussions

Thermal and concentration diffusion of particles in nanofluid is initiated due to heating by a laser beam. This results in establishing a refractive index gradient. In case of magnetic nanoparticles like Fe_3O_4 in hexane and kerosene, it leads to a formation of diffraction patterns. No such patterns were observed in case of non-magnetic nanoparticles like CdS in methanol. This is probably due to the fact that absorption is less for CdS nanoparticles compared to Fe_3O_4 nanoparticles at 632 nm.

Transient study was carried out to understand how refractive index gradient is developed over time. In the transient study it was observed that as the particles diffuse, it takes some time to attain dynamic equilibrium and to subsequently form stable thermal lens induced diffraction patterns. In case of CdS nanoparticles, there is a variation in diameter of transmitted laser beam. This is shown in fig. 3 and fig. 4. Time transient was also studied in magnetic field for Fe_3O_4 nanoparticles in hexane to compare the difference in patterns obtained in zero magnetic field. This is shown in fig. 5 and fig. 6. In the case of Fe_3O_4 nanoparticles in hexane in zero field, a compression along the vertical direction is clearly seen with passage of time in seconds. This is attributed to effect of gravity[8]. Contrary to this effect, in the presence

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of transverse magnetic field, no such compression is observed. This effect may be attributed to the aggregation of the nanoparticles forming chains along the direction of the magnetic field as has been reported earlier.

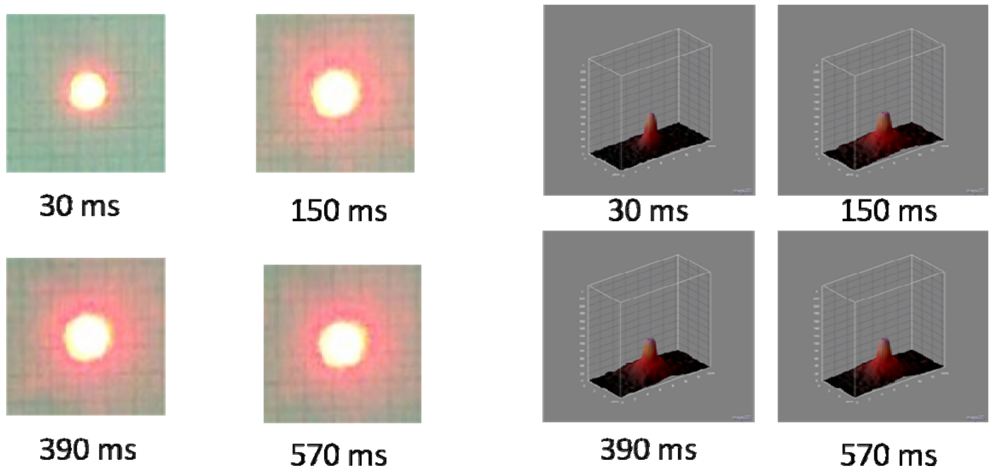

Figure 3: Time transient response of CdS in methanol and corresponding intensity profile

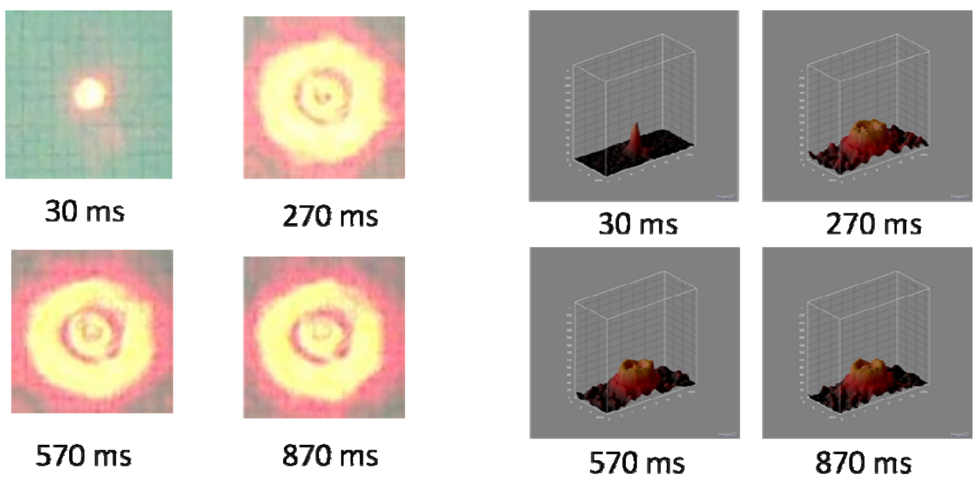

Figure 4: Time transient response of Fe_3O_4 in kerosene and corresponding intensity profile

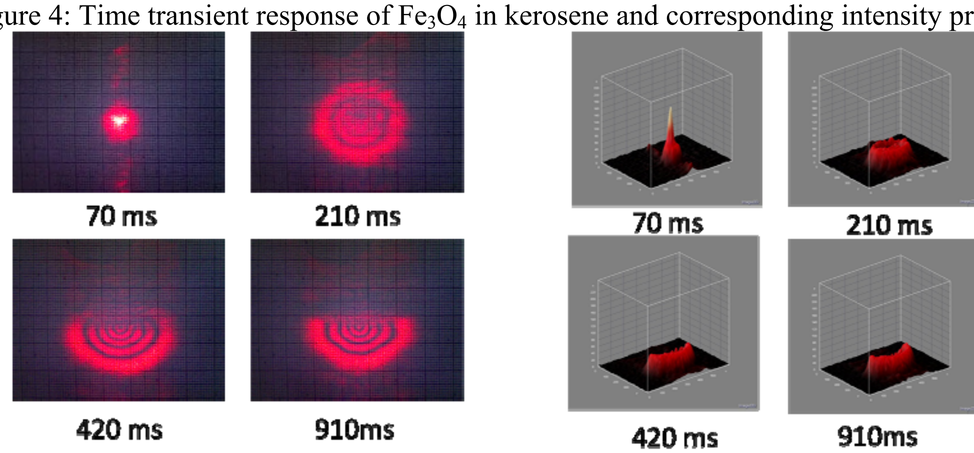

Figure 5: Time transient response of Fe_3O_4 in hexane (B = 0 G) and corresponding intensity profile

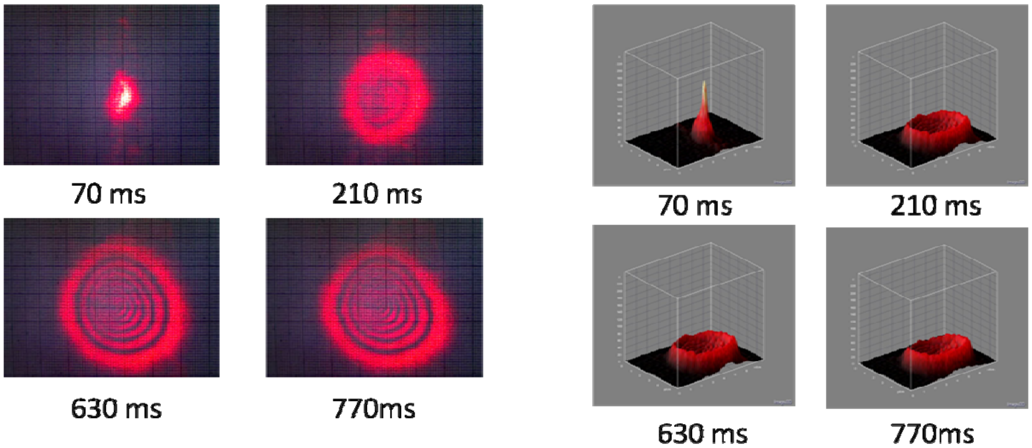

Figure 6: Time transient response of Fe_3O_4 in hexane (B = 1.7kG) and corresponding intensity profile

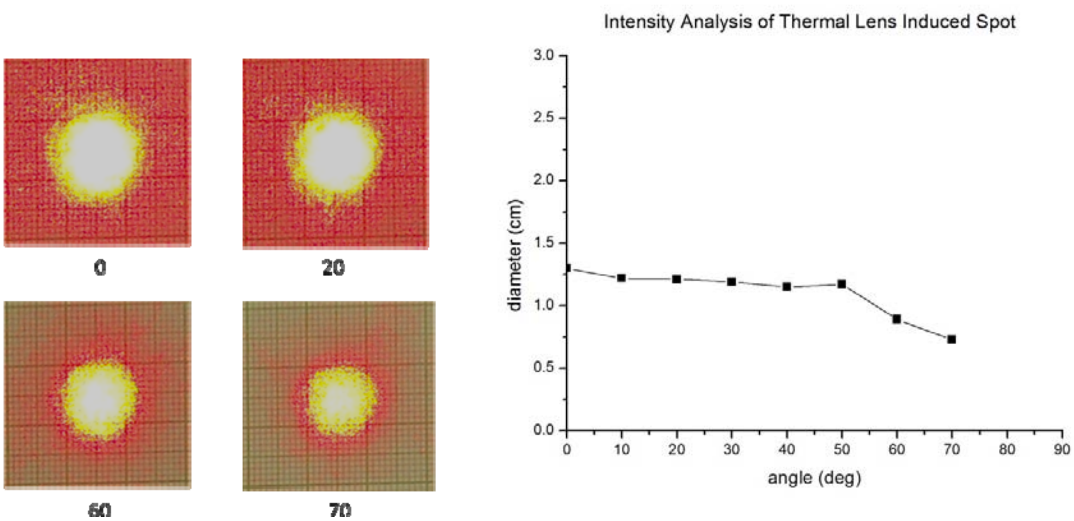

Figure 7: Intensity dependence of CdS in methanol and corresponding diameter variation at different polarizing angles

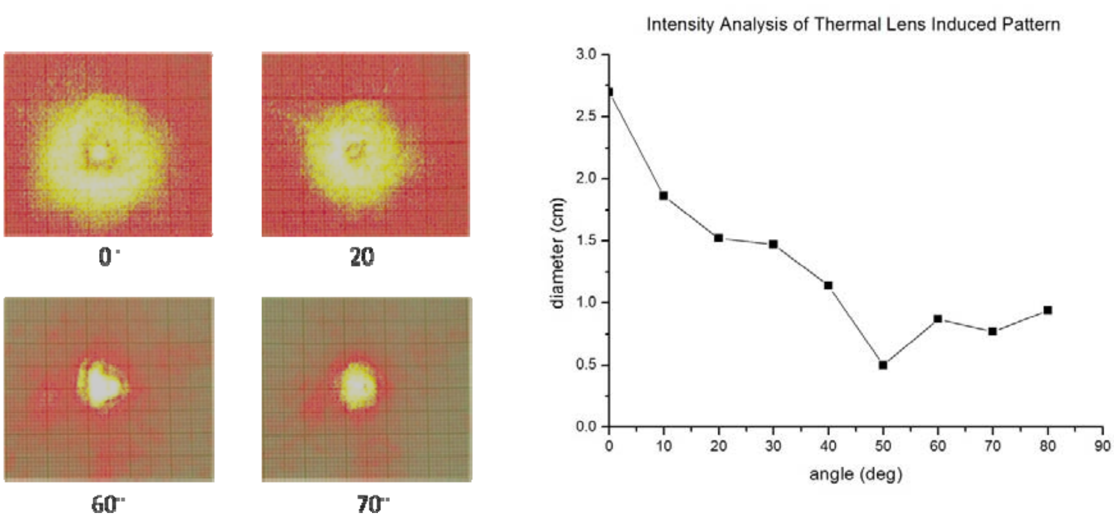

Figure 8: Intensity dependence of Fe_3O_4 in kerosene and corresponding diameter variation at different polarizing angles

Intensity dependence of thermal lensing shows that thermal lens induced diffraction patterns change as polarizer angle is changed. This is observed for magnetic nanoparticles. In case of non-magnetic nanoparticles, transmitted laser beam spot size changes with polarizer angle. This is shown in fig. 7 and fig. 8 respectively. For magnetic nanoparticles, below a threshold intensity of the incident beam, the diffraction patterns disappear.

Conclusions

Evolution of diffraction patterns in various nanofluids shows a definite thermal response time and correspondingly, changes in refractive index gradient relate to this thermal response. In case of non-magnetic particles like CdS, no such patterns were observed. This can be attributed to the fact that coefficient of absorption plays an important role to form diffraction pattern due to thermal lensing. This gives insight to understand heat transfer phenomena in different nanofluids and their potential application for developing nanofluid based sensors.

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