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Trap stiffness modification of an optically trapped microsphere through directed motion of nanoparticles

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We report an enhancement in the corner frequency of an optically trapped non-magnetic microsphere in the plane perpendicular to the laser propagation direction on addition of ferrofluid to the suspension medium. We conjecture that a directed motion of the nanoparticles toward the trap in this plane is responsible for the augmentation. Changes in the corner frequency in the presence of external magnetic field gradients lend credence to this conjecture. Corner frequency augmentation is also observed when zinc oxide nanoparticles are used. Here, however, no further changes are seen in the presence of magnetic field gradients. © 2020 Optical Society of America

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1. INTRODUCTION

Ferrofluids (FFs) are suspensions of magnetic nanoparticles in carrier fluids like water or organic solvents [1–3]. To prevent agglomeration of these nanoparticles in the suspension, a surfactant is usually used [1,3]. The use of the surfactant leaves the otherwise neutral nanoparticles either positively or negatively charged on the surface. The Coulombic repulsion between the nanoparticles then prevents aggregation and a well synthesized FF has a characteristic feature of being highly mono-disperse.

Optical tweezers (OTs) are capable of trapping micrometer to submicrometer sized particles with the use of highly focused laser beams, exerting forces of the order of a few piconewtons [4,5]. Conventional OTs can trap particles with dimensions between 0.2 μm to 100 μm [6,7] and are used extensively to study a wide variety of soft and living material.

Stable optical trapping of nanoparticles requires consideration of certain other criteria. The spot size of the focused laser beam must be comparable to the size of the particle for there to be sufficient gradient force for trapping. This implies that the diffraction limit determines the smallest particle that can be stably trapped [8,9]. Further, the optical trapping of nanoparticles depends critically on the particle material that influences the stability of trapping through the polarizability [10]. Techniques to trap nanoparticles optically thereby involve measures such as nanoparticle design with enhanced polarizability [10], use of plasmonic tweezers/near-field optical trapping [8,9,11,12], or the use of ultrashort laser pulses [13–16]. Without a deliberate

attempt to design for the trapping of nanoparticles, there is a strong possibility that conditions for stable trapping are never satisfied in a conventional OT. Thus, one may not observe the formation of clusters within the trap volume unlike that reported in Ref. [3].

A study on the behavior of a non-magnetic probe trapped using a conventional OT while suspended in an aqueous medium modified with the presence of FFs was carried out to investigate whether the dynamics of the nanoparticles present in the suspension could be inferred from the fluctuation behavior of the probe. If this was indeed the case, it would be possible to distinguish between various FFs on the basis of the dynamics of the nanoparticles. Further, it would also be possible to detect alterations in the dynamics due to the presence of any object that could elicit a magnetic response from the nanoparticles. In fact, this study could be thought of as a precursor to understanding the dynamic response of an FF system to a wide variety of magnetic structures present in the vicinity in differing concentrations, each eliciting different magnetic responses from the nanoparticles. These studies could then ultimately pave the way to a method of magnetically characterizing a microvolume with a combination of a FF and other types of magnetic probes.

In this study, we report on the trapping of silica microspheres suspended in a dilute water-based FF with magnetite nanoparticles in a conventional OT. We demonstrated that at FF concentrations greater than a certain threshold concentration, the trapped probe showed an increased corner frequency

in the plane perpendicular to the laser propagation direction when compared to that in plain water for the same laser power. The increase appeared to depend on the FF concentration for the range of concentrations studied by us. Further, the change in the corner frequency was found to be sensitive to the presence of magnetic field gradients at the trap location. Along the laser propagation direction, the variance of the positional fluctuations of the probe was found to increase with increase in FF concentration. These studies were repeated with suspensions of zinc oxide (ZnO) nanoparticles at concentrations identical to that of the FFs. The results with the ZnO nanoparticle suspension showed a trend identical to that with the FF except for a complete lack of response to external magnetic field gradients.

2. EXPERIMENTAL DETAILS

The OT setup employed by us is described in detail in Ref. [17]. Briefly, it consists of an 830 nm diode laser (Thorlabs Inc, USA) which is passed through an oil immersion objective (Olympus PLCN100XO/1.25, Japan) to form the trap. Backscattered light from the trapped particle is detected using a quadrant photo-detector (QPD) (Newport Inc., USA) whose output is logged on a computer after digitization through a data acquisition (DAQ) card (National Instruments). A Kohler illumination arrangement allows for visualization of the trapped micro-object using a camera (Dragonfly, PointGrey Inc.). Figure 1 gives a schematic of the trap.

In our study, we used commercially procured water-based FFs (Chemicell GmbH) with magnetite (Fe_3O_4) core nanoparticles of hydrodynamic diameter of 100 nm and anionic surface charge. Appropriate quantities of FF along with silica beads (Cospheric, USA) of 1 μm diameter were suspended in de-ionized water to make a total sample volume of 410 μl and held in a sample chamber made by gluing an O-ring of inner diameter 0.8 cm and thickness 1 mm to a glass coverslip. The final concentration of the FF in the O-ring is reported in units of microgram/microliter ($\mu\text{g}/\mu\text{l}$). The ZnO nanoparticle aqueous suspension was sourced from Sigma Aldrich. The nanoparticles had an average particle size of 40 nm.

Each experiment was comprised of trapping a single silica bead, which was used as the probe particle, and recording its Brownian fluctuations from which the power spectral density (PSD) [18–22] and autocorrelation function (ACF) [22–24] were computed. The experiment was carried out using plain water and different concentrations of FF as well as ZnO in the suspension medium at three different laser powers. The symmetry of the trap was verified visually with the camera.

The Brownian fluctuations of the trapped bead were recorded as differential voltages from the QPD and converted from analog to digital through the DAQ card. The data was recorded using a code written on LabView software. We recorded for a time of about 13 s with a sampling time of 50 μs , yielding a total of 262,144 data points. Ten such datasets were recorded at each concentration at three different laser powers.

3. RESULTS AND DISCUSSIONS

A. Fluctuation Behavior of a Trapped Probe in the XY Plane

The QPD detects fluctuations of the trapped bead in the XY plane perpendicular to the laser propagation direction (Z), as shown in Fig. 1. The QPD resolves the intensities of the laser backscattered from the trapped probe onto the four quadrants in terms of voltages (inset of Fig. 1). The normalized fluctuations of the trapped particle in two orthogonal displacements, along the X and Y axes, in units of volts, are given by [25]

$$X_V = \frac{(V_1 + V_4) - (V_2 + V_3)}{V}, \quad (1)$$

$$Y_V = \frac{(V_1 + V_3) - (V_2 + V_4)}{V}, \quad (2)$$

where V_1 , V_2 , V_3 , and V_4 are the voltages from the four quadrants of the QPD, respectively, and V is their sum. Note that the voltage recorded is proportional to the intensity of the backscattered light on each of the quadrants. We have used the calibration method outlined in Refs. [17,25] to obtain the calibration factor to convert the fluctuations obtained in voltages to displacements.

We computed the PSD [17,26] of the trapped probe from the fluctuations recorded in the XY plane and estimated the corner frequency from the fit to the PSD. The reported value of the corner frequency is the mean extracted from the 10 such measured datasets while the standard deviation is reported as the error bar. Figure 2 shows typical PSD plots for a bead trapped in the absence and presence of the FF.

When the probe was trapped in plain water at room temperature, the corner frequencies along two orthogonal directions in the XY plane matched to two decimal places, further confirming the symmetry of the trap.

Figure 3 shows the variation of the corner frequency as a function of FF concentration at three different laser powers.

It can be seen from the figure, that up to a certain concentration, the value of the corner frequency does not change. A change was observed only for concentrations greater than

Fig. 1. Schematic of the optical tweezer setup. Inset: a schematic of the QPD.

Fig. 2. Comparative PSD plots for the trapped bead, trapped at 12 mW, in the absence and presence of the FF.

Fig. 3. Variation of the corner frequency of the probe as a function of FF concentration at different laser powers. Lines are a guide to the eye.

this threshold value. Note, also, that certain close-lying concentrations above the threshold value showed similar values of the corner frequency. This was because of the finite sampling time of 50 μ s which rendered the measurement insensitive to any change involving a time scale smaller than this.

Figure 4 shows the variation of corner frequency with FF concentration for each of the laser powers along with straight

Fig. 4. Variation of corner frequency with FF concentration at laser powers of (a) 4 mW, (b) 8 mW, and (c) 12 mW. Additionally, straight line fits for (i) initial values of FF concentrations where no change in the corner frequency is observed and (ii) only those concentrations where measurable changes in the corner frequency are observed are shown for each laser power. The concentration corresponding to the point of intersection of these two straight lines is the threshold concentration at that laser power.

line fits. The point of intersection of the two linear fits yielded the threshold value of FF concentration. The threshold value is reported in the plot. Note that the threshold concentration increases with increase in laser power.

The PSD analysis of fluctuations recorded in the X and Y directions established that the corner frequencies in the two directions were the same (within error bars) for the entire range of FF concentrations studied. Figure 5 shows the variation of the corner frequency as a function of the FF concentration in the X and Y directions for a laser power of 8 mW.

When the Y displacements are plotted against the X displacements, we get scatter plots. For a symmetric trap and a spherical

Fig. 5. Corner frequency of the probe along X (Red) and Y (Black) directions. The equivalence of these values demonstrates the isotropy of the optical trap in the XY plane for plain water and in the presence of FF. The dotted lines are a guide to the eye.

probe, the scatter plots ought to be a circle. Figure 6 shows the scatter plot of the trapped probe in (a) plain water (red) and (b) in the presence of FF particles (green) at the highest concentration studied by us ($125 \mu\text{g}/\mu\text{l}$) along with position histograms for the two cases.

Fig. 6. Scatter plots of the trapped particle in (a) plain water (red) and (b) with $125 \mu\text{g}/\mu\text{l}$ FF (green) and the corresponding displacement histograms. Trapping power is 12 mW.

Fig. 7. Variation of the FWHM of the trapped probe displacements in the XY plane as a function of FF concentration. The lines are a guide to the eye. The values are averaged over both X and Y values owing to isotropy.

From the scatter plots we observed that there was a reduction in the fluctuation amplitudes of the trapped probe on addition of FF. Figure 7 shows the variation of the full width at half-maximum (FWHM) of the position histograms with increase in the FF concentration. The plot shows a decrease in the FWHM as the FF concentration is increased. This ascertained that the probe was constrained to execute smaller fluctuations in the XY plane with increase in FF concentration.

B. Autocorrelation Function Analysis

A change in the corner frequency in the PSD of an object trapped in an OT can arise out of various factors, such as a change in the NA of the objective, change in the laser power, change in the wavelength of the laser used, change in the refractive index of the suspension medium, or an alteration of the viscosity of the suspension medium. For a microparticle of a fixed size and material in an OT setup with fixed laser wavelength and an objective of a fixed NA, the only means of varying the corner frequency is by either altering the viscosity of the suspension medium or the laser power. We do not expect a change in the refractive index of the suspension on addition of FF as the concentrations used are extremely low and these nanoparticles do not dissolve in water.

Our studies showed an increase in the corner frequency in the XY plane with increase in FF concentration when the laser power was held constant. A change in the viscosity of the suspension fluid with the addition of FF could be a possible reason for the augmentation of the corner frequency. An ACF analysis was carried out to shed further light on this.

We computed the normalized position ACF of the probe particle. The fit to this variation with lag time (Δt) is given by [19,22,23]

$$\langle x(t)x(t + \Delta t) \rangle = \frac{Ae^{-\frac{t}{\tau}}}{\sigma_x^2}, \quad (3)$$

for $\Delta t > \tau$, where τ is the relaxation time, σ_x^2 is the variance of the x fluctuations of the probe, and A is given by

Fig. 8. Autocorrelation function for (a) plain water and (b) 125 µg/µl concentration. The relaxation times τ are shown alongside. The laser power is 4 mW.

$$A = \frac{K_B T}{\kappa}, \quad (4)$$

κ being the trap stiffness, K_B the Boltzmann constant, and T the temperature of the suspension. The relaxation time, τ , is that for the diffusive motion of the trapped probe which depends on the drag felt by the probe and the trap stiffness [19]. At much smaller lag times, the behavior of the probe is ballistic, often referred to as fast Brownian motion [19,27], which the measurement process used here, with a sampling time of 50 µs, is completely insensitive to.

Figure 8(a) shows the ACF (and fit) for plain water while Fig. 8(b) shows the same for an FF concentration of 125 µg/µl. The fit yields τ , which is shown in the plots.

Figure 9 shows the variation of τ with FF concentration at different laser powers.

As is evident from Eq. (3), the ACF of a particle trapped in an OT shows an exponential drop with time. The characteristic time at which the value reduces to $1/e$ of the initial value, τ , is dependent on the trapping force. One can regard τ as a characteristic time in which the optical trap force restores the probe back to the equilibrium position. A weak trap will have a large relaxation time while stronger traps will have small values [19,20]. We found that the relaxation time decreased with

Fig. 9. Variation in relaxation time, τ , extracted from the exponential fit to the ACF, with FF concentration at different laser powers. The dotted lines are a guide to the eye.

increase in FF concentration, indicative of an increase in the trap strength.

One can also estimate viscosity of the suspension medium through the ACF analysis. In terms of the corner frequency, f_c , the trap stiffness κ can be written as [28,29]

$$\kappa = 2\pi\gamma f_c, \quad (5)$$

where γ is the drag coefficient. The drag coefficient for a spherical particle is given by

$$\gamma = 6\pi\eta r_p, \quad (6)$$

η being the dynamic viscosity of the surrounding medium and r_p , the radius of the trapped probe. The relaxation time and the trap stiffness are related as follows [19]:

$$\tau = \frac{\gamma}{\kappa}. \quad (7)$$

From Eqs. (4)–(7), one can write the viscosity in terms of τ and the fit parameter A as

$$\eta = \frac{K_B T \tau}{6\pi r_p A}. \quad (8)$$

Taking $T = 296$ K (room temperature), the value of the dynamic viscosity was estimated using Eq. (8) for various FF concentrations. Figure 10 shows the variation of viscosity with FF concentration averaged over all laser powers. No significant variation of viscosity with change in FF concentration was observed for the range studied by us.

Figure 11 shows the variation in the corner frequency with FF concentration as estimated from the viscosity values arrived at from the ACF analysis using Eq. (6). The measured values of corner frequency are also shown. It is evident that viscosity change alone cannot explain the large variation in corner frequency that is measured with change in FF concentration. The corner frequency change thereby is attributed solely to the change in the trap stiffness experienced by the probe [Eq. (5)].

Fig. 10. Variation of viscosity as a function of FF concentration (black squares). Note that the viscosity values shown here are estimated at a room temperature of 296 K and averaged over all laser powers. The viscosity values of some common liquids are also shown to contrast with the minor variation seen here. Inset shows the variation of the estimated viscosity on an enlarged Y scale.

Fig. 11. Variation of the measured corner frequency values for 4 mW laser power and those estimated from viscosity values as a function of FF concentration. The dotted lines are a guide to the eye.

C. Effect of an External Magnetic Field

In order to study the effect an external magnetic field would have on the trap enhancement measured due to the presence of FF in the suspension medium, we placed two bar magnets with field strengths of about 150 Gauss, 1.5 cm from the outermost edge of the O-ring in the various configurations shown in Fig. 12. The sample contained 125 $\mu\text{g}/\mu\text{l}$ of FF and 10 μl of the probe particles in a total volume of 410 μl .

We observed that the corner frequency values in the X and Y directions were no longer coincident in the presence of the external magnetic field. The exact manner of the change depended on the particular spatial configuration of the permanent magnets. An increase in the laser power, while increasing the individual corner frequencies in both the X and Y directions, did not alter the manner of the change. In other words, the presence of external magnetic fields altered the isotropy of the trap in the XY plane, the resulting anisotropy depending on the exact nature of the external magnetic field and field gradients.

Fig. 12. Schematic of the arrangement of the bar magnets around the sample. The magnets were placed with their (a),(b) like poles pointing in the same direction and (c),(d) like poles pointing in opposite directions.

Fig. 13. Variation of the corner frequency for the external bar magnet spatial configurations (a), (b), (c), and (d) as shown in Fig. 12 as a function of the laser power. The dotted lines are a guide to the eye. Legends: black – X (plain water), red – Y (plain water), blue – X (plain water + FF), cyan – Y (plain water + FF), magenta – X (plain water + FF in the presence of magnetic fields), dark yellow – Y (plain water + FF in the presence of magnetic fields).

For purposes of comparison, the fluctuations of the trapped probe were studied in the presence of each of the bar magnet configurations when there was no FF present in the suspension media. No significant change in the corner frequency was observed from that with plain water without external magnets. Figure 13 shows the plots of the corner frequency as a function of laser power for the various external magnet configurations (a), (b), (c), and (d) as shown in the schematic in Fig. 12.

Table 1. α Values for the Three Different Laser Powers in Each of the Magnet Configurations^a

Power (mW)			
Magnet Configuration	4	8	12
(a)	0.916	0.946	0.931
(b)	1.078	1.063	1.073
(c)	1.414	1.353	1.313
(d)	0.721	0.756	0.773

^aWhen there are no external magnetic field gradients, α is 1.

In configurations (a) and (b), a higher corner frequency was recorded in both X and Y directions compared to the situation with just plain water or FF without external magnets. However, while the corner frequency in the X direction was consistently higher than that in the Y direction for all powers in configuration (a), the reverse was true for configuration (b). In configurations (c) and (d), we found that while the corner frequency in one direction was elevated over the value for FF without external magnets, it was suppressed below this value in the orthogonal direction.

We define an asymmetry factor, α , that is the ratio of the corner frequency in the X direction to the corner frequency in the Y direction. The α value for the three different laser powers in each of the configurations has been tabulated in Table 1. We found that the α value was different for different configurations at a given laser power reflecting the changes in the magnetic neighborhood of the trapped probe.

Based on the results of all the studies carried out, we hypothesize that the gradient force acting in the XY plane of the OT attracts the charged dipole on the nanoparticles toward the trap center. The particles are accelerated toward the trap center and are moving with large velocities as they approach it. The nanoparticles collide with the trapped probe and transfer momentum to it, always in the direction toward the trap center. Under these circumstances the trapped probe particle is constrained to execute smaller fluctuations in the XY plane. This is evident from the reduction in the area of the scatter plot of the trapped probe with increase in FF concentration and an augmented corner frequency in the power spectral density of the probe at the same laser power.

We further argue that in the presence of the laser trap alone, the movement of the FF particles with respect to the trap position is isotropic in the XY plane. This is evident from the fact that the corner frequencies in two orthogonal directions in the XY plane at a given power are practically the same for the entire range of FF concentrations studied and also true at every laser power studied by us. Figures 14(a) and 14(b) are representations of the FF particles, the trapped probe, and the isotropic movement of the FF particles toward the trapped probe in the XY plane, effectively constraining the amplitude of fluctuations executed by the probe, causing it to experience a higher trap stiffness.

Also, we find that the stronger the trap, the higher the threshold concentration of FF before an enhancement in the corner frequency is seen. This can also be explained, as a probe that is held more strongly would require a larger rate of momentum transfer toward the center of the trap for the enhancement in

Fig. 14. (a) Representation of the FF nanoparticles and the probe particle in the suspension in the absence of the trap (not to scale). (b) Representation of the movement of the FF nanoparticles in the XY plane when the laser is switched on.

Fig. 15. Variance of the probe along the Z direction with FF concentration when it is trapped at 4 mW. The dotted line is a guide to the eye.

corner frequency to be evident. This would only be possible at larger concentrations of the FF particles.

When bar magnets are placed in various spatial configurations around the sample, an anisotropy in the XY fluctuations of the probe is observed, the exact nature of which is linked with the particular spatial configuration in place. This is due to alteration of the movement of the FF particles under the influence of the external magnetic field/field gradients.

While the movement of the FF particles toward the trap, in the absence of an external magnetic field, is isotropic in the XY plane, these particles do not stay in the trap volume [6,7]. The variance in the Z direction showed a trend of increasing with increase in FF concentration, as seen in Fig. 15. The FF particles are thereby likely to be making their way out of the trap volume primarily along the Z axis.

D. Estimation of Velocity of the FF Particles

The Langevin equation for an optically trapped probe particle suspended in plain water is [22]

$$m\ddot{x} + \gamma\dot{x} + \kappa x = (\sqrt{2\gamma K_B T}) \eta(t), \quad (9)$$

where m is the mass of the probe particle, κ is the trap stiffness, K_B is the Boltzmann constant, T is the temperature, and $\eta(t)$ is a Gaussian stochastic process satisfying

$$\langle \eta(t) \rangle = 0, \quad (10)$$

$$\langle \eta(t)\eta(t') \rangle = \delta(t - t'), \quad (11)$$

δ being the Dirac delta function. Note, while it is a common approach to solve for Brownian trajectories using the Langevin equation, alternate approaches like Fokker Planck analysis outlined in Ref. [30] allow for a direct estimation of the position probability distribution function of the Brownian particle, eliminating the need for analyzing a large number of computed trajectories to understand the trapping dynamics. In the presence of FF, the Langevin equation for the trapped probe is modified to

$$\gamma\dot{x} + (\kappa + \Delta\kappa)x = (\sqrt{2\gamma K_B T}) \eta(t). \quad (12)$$

The inertial term in Eq. (9) is dropped here as low Reynold's number conditions apply. Further, no change in the drag coefficient is expected as ACF analysis established that addition of FF, in the concentrations relevant to this study, does not alter the viscosity of the suspension. The change in the trap stiffness, $\Delta\kappa$, is attributed to the interaction between the trapped probe and the FF nanoparticles.

Owing to the large mass disparity between the probe and individual FF particles, the momentum transfer for every FF nanoparticle–probe interaction can be written as $2 \times \langle m_{FF} \rangle \times \langle v_{FF} \rangle$. Here $\langle m_{FF} \rangle$ is the average mass of a FF nanoparticle and $\langle v_{FF} \rangle$ is its average velocity within the trap volume. We assume that the velocity of the probe is much smaller than $\langle v_{FF} \rangle$ due to its much larger size that results in a much higher drag coefficient. Also, the fact that the probe is confined in a weak trap implies that its maximal attainable velocity remains small. On the other hand, the primary force acting on the FF nanoparticle is the gradient force which can yield much higher magnitudes of $\langle v_{FF} \rangle$. While the gradient force on

the FF nanoparticles is the dominant force, electrostatic forces due to surface charges on the probe and other nanoparticles as well as the hydrodynamic interactions of the probe and other nanoparticles are also likely to have influence on $\langle v_{FF} \rangle$.

The force acting on the probe due to the FF is the rate of change of momentum for probe–nanoparticle interactions. The nanoparticles are only locally influenced by the trap in the spatial region around the center of the focused optical field and as a result will have an average velocity of $\langle v_{FF} \rangle$ only in the limited spatial range of the trap volume. Let us assume that the probe moves a distance δx in a small time interval δt . We take δx as comparable to the spatial range over which the average nanoparticle velocity magnitude is indeed $\langle v_{FF} \rangle$. The number of collisions in the small time interval δt can be written as

$$\beta = \rho_{FF} \times A_{\text{probe}} \times \delta x, \quad (13)$$

where ρ_{FF} is the number density of FF particles and A_{probe} is the cross section of the probe. The momentum transfer to the probe in the direction of the trap center for this interval of time δt can then be written as

$$\Delta p_c = 2 \langle m_{FF} \rangle \langle v_{FF} \rangle \times \beta. \quad (14)$$

The rate of change of this momentum will show up as the augmentation of the trap force. Identifying δx as the displacement of the probe from the trap center, as displacements here are very small, we use Eq. (12) and write

$$\Delta\kappa\delta x = \lim_{\delta t \rightarrow 0} 2 \langle m_{FF} \rangle \langle v_{FF} \rangle \times \rho_{FF} \times A_{\text{probe}} \times \frac{\delta x}{\delta t}, \quad (15)$$

$$\Rightarrow \Delta\kappa = 2 \langle m_{FF} \rangle \langle v_{FF} \rangle \times \rho_{FF} \times A_{\text{probe}} \times \left\langle \frac{\dot{x}}{x} \right\rangle_{\text{plain water}}, \quad (16)$$

where $\left\langle \frac{\dot{x}}{x} \right\rangle_{\text{plain water}}$ is the average of the ratio of the velocity of the probe particle and displacements of the probe particle in plain water. If we plot $\Delta\kappa$ as a function of ρ_{FF} , one could express in terms of the slope as

$$\langle v_{FF} \rangle = \frac{\text{slope}}{2 \langle m_{FF} \rangle \times A_{\text{probe}} \times \left\langle \frac{\dot{x}}{x} \right\rangle_{\text{plain water}}}. \quad (17)$$

We take the number density (concentration) to be the same as that throughout the sample volume. This is unlikely to be the case as FF particles will aggregate near the trap. This is because the laser field acts on the dipole on the nanoparticles and as the

Fig. 16. Variation of $\Delta\kappa$ as a function of FF concentration for the three different laser powers. The dotted line is a guide to the eye. The solid lines are linear fit to the data. Max $\langle v_{FF} \rangle$ at the three different laser powers are — 4 mW: 194.53 ms⁻¹; 8 mW: 237.76 ms⁻¹; and 12 mW: 256.59 ms⁻¹.

field strength increases near the vicinity of the trap, the number of particles will also increase. Therefore, a density gradient will develop when the laser is switched on. However, as we do not have an independent measure of this density gradient, we assume that the concentration of the particles in the vicinity of the trap is the same as elsewhere in the sample volume. Thereby the value of velocity estimated is an upper limit to the average velocity of the nanoparticles in the trap volume. This quantity could help quantify the effect that addition of an FF to a suspension medium has on an optically trapped probe. Further, it could help in distinguishing and comparing different types of FFs.

We plotted the variation of $\Delta\kappa = 2\pi\gamma\Delta f/c$ [in analogy with Eq. (5)] with change in concentration for concentrations greater than that of the threshold concentration for each laser power. Concentration was taken to scale in direct proportion with ρ_{FF} .

Figure 16 shows the variation of $\Delta\kappa$ with FF concentration with straight line fits to the data for all three laser powers. The maximum value of the average velocity attributed to each FF nanoparticle within the trap volume is reported in the plots. As was expected, the value of $\langle v_{FF} \rangle$ increased with laser power.

E. Studies with ZnO Nanoparticles

The entire sets of studies carried out with the FFs were repeated for identical concentrations of ZnO nanoparticles. The variation of the corner frequency as a function of ZnO concentration is shown in Fig. 17. An isotropic enhancement in the corner frequency in the XY plane beyond a threshold concentration was observed as with the FFs. Here too the threshold concentration increased with increasing laser power while the corner frequency enhancement at a given laser power was found to depend on the nanoparticle concentration.

In contrast with the FFs, however, no change in trapping isotropy was observed in the presence of the magnets. Figure 18 provides the consolidated results which show that the corner frequencies in the two orthogonal directions in the XY plane remain unaffected in the presence of magnets.

Fig. 17. Variation of the corner frequency of the probe as a function of ZnO concentration at different laser powers. Lines are a guide to the eye.

Fig. 18. Variation of the corner frequency for the external bar magnet spatial configurations (a), (b), (c), and (d) as shown in Fig. 12 as a function of the laser power. The dotted lines are a guide to the eye. Legends: black – X (plain water), red – Y (plain water), blue – X (plain water + FF), cyan – Y (plain water + FF), magenta – X (plain water + FF in the presence of magnetic fields), dark yellow – Y (plain water + FF in the presence of magnetic fields).

4. CONCLUSION

We conclude that the addition of FF to a suspension of microparticles in water increases the trapping strength for the microparticle in an optical trap in the plane perpendicular to the laser propagation direction (XY plane). The trap strength enhancement is observed only above a certain threshold value of the FF concentration and the enhancement increases with increase in FF concentration in the range studied. The threshold concentration increases with laser power and the trap strength enhancement is isotropic in the XY plane. ACF analysis for the trapped probe shows that there is no measurable change in the viscosity of the suspension in the FF concentration range relevant to this study and we conclude that the corner frequency enhancement observed in the XY plane is indeed a trap strength enhancement.

All these studies lead us to believe that this is due to a directed movement of the nanoparticles toward the trap center in the XY plane. This movement leads to collisions between the FF particles and the trapped probe, leading to a momentum transfer that is always directed toward the trap center constraining the larger fluctuations of the probe. Strength is given to the above conjecture by demonstrating that the presence of external magnetic fields breaks the isotropy in the XY plane, and one observes different trap strengths in orthogonal directions.

The FF nanoparticles do not remain within the trap volume and an increase in the variance of the fluctuations of the trapped probe along the laser propagation direction leads us to believe that they exit the trap volume along this direction.

An upper limit to the average velocity of the nanoparticles within the trap volume when moving under the combined influence of the laser field gradient, hydrodynamic and surface charge related electrostatic effects is estimated from the slope of the variation of $\Delta\kappa$ with FF concentration. We propose that this maximum possible average velocity of the FF nanoparticles could be used as a factor to quantify the effect a particular FF has on a trapped probe.

Identical studies with suspensions of ZnO nanoparticles yield similar trends in the results except for a lack of response to external magnetic field gradients. Apart from lending further strength to our hypothesis, this could point out to a possible universality of being able to enhance the stiffness of an optical trap by adding any surface treated nanoparticles to the suspension medium. However, note that the surface charge on the nanoparticles as well as on the probe used could lead to varying results as the probes could affect the colloidal stability of the nanoparticle suspension.

We have clearly shown that a combination of a non-magnetic probe trapped in an optical tweezer while suspended in an FF with concentrations above a certain threshold can be used to sense the configuration of a magnetic field gradient around the sample space and this could underlie the development of a technique to magnetically map a small volume.

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REFERENCES

1. B. Hoffmann and W. Kohler, "Reversible light-induced cluster formation of magnetic colloids," *J. Magn. Magn. Mater.* **262**, 289–293 (2003).
2. R. R. Kellner and W. Köhler, "Short-time aggregation dynamics of reversible light-induced cluster formation in ferrofluids," *J. Appl. Phys.* **97**, 034910-8 (2005).
3. J. Masajada, M. Bacia, and S. Drobczyński, "Cluster formation in ferrofluids induced by holographic optical tweezers," *Opt. Lett.* **38**, 3910–3913 (2013).
4. A. Ashkin, J. M. Dziedzic, J. E. Bjorkholm, and S. Chu, "Observation of a single-beam gradient force optical trap for dielectric particles," *Opt. Lett.* **11**, 288–290 (1986).
5. A. Ashkin, J. M. Dziedzic, and T. Yamane, "Optical trapping and manipulation of single cells using infrared laser beams," *Nature* **330**, 769–771 (1987).
6. A. Ashkin, "Forces of a single-beam gradient laser trap on a dielectric sphere in the ray optics regime," *Biophys. J.* **61**, 570–582 (1992).
7. M. Sarshar, W. T. Wong, and B. Anvari, "Comparative study of methods to calibrate the stiffness of a single-beam gradient-force optical tweezers over various laser trapping powers," *J. Biomed. Opt.* **19**, 115001 (2014).
8. M. A. Zaman, P. Padhy, and L. Hesselink, "Near-field optical trapping in a non-conservative force field," *Sci. Rep.* **9**, 1–11 (2019).
9. M. L. Juan, M. Righini, and R. Quidant, "Plasmon nano-optical tweezers," *Nat. Photonics* **5**, 349–356 (2011).
10. P. M. Bendix, L. Jauffred, K. Norregaard, and L. B. Oddershede, "Optical trapping of nanoparticles and quantum dot," *IEEE J. Sel. Top. Quantum* **20**, 15–26 (2014).
11. O. M. Maragò, P. H. Jones, P. G. Gucciardi, G. Volpe, and A. C. Ferrari, "Optical trapping and manipulation of nanostructures," *Nat. Nanotechnol.* **8**, 807–819 (2013).
12. C. Min, Z. Shen, J. Shen, Y. Zhang, H. Fang, G. Yuan, L. Du, S. Zhu, T. Lei, and X. Yuan, "Focused plasmonic trapping of metallic particles," *Nat. Commun.* **4**, 1–7 (2013).
13. A. Usman, W. Y. Chiang, and H. Masuhara, "Optical trapping of nanoparticles by ultrashort laser pulses," *Sci. Progress.* **96**, 1–18 (2013).
14. D. Roy, D. Goswami, and A. K. De, "Exploring the physics of efficient optical trapping of dielectric nanoparticles with ultrafast pulsed excitation," *Appl. Opt.* **54**, 7002–7006 (2015).
15. D. Mondal, S. N. Bandyopadhyay, and D. Goswami, "Elucidating optical field directed hierarchical self-assembly of homogenous versus heterogeneous nanoclusters with femtosecond optical tweezers," *PLOS ONE* **14**, e0223688 (2019).
16. A. Devi and A. K. De, "Simultaneous detection of two-photon fluorescence and backscatter in laser trapping of dielectric nanoparticles," *J. Nanophotonics* **13**, 020501 (2019).
17. A. Raghu and S. Ananthamurthy, "Construction of an optical tweezer for nanometer scale rheology," *Pramana-J. Phys.* **65**, 699–705 (2005).
18. D. G. Grier, "Optical tweezers in colloid and interface science," *Curr. Opin. Colloid Interface Sci.* **2**, 264–270 (1997).
19. S. Henderson, S. Mitchell, and P. Bartlett, "Position correlation microscopy: probing single particle dynamics in colloidal suspensions," *Colloids Surf., A* **190**, 81–88 (2001).
20. P. Bartlett and S. Henderson, "Three-dimensional force calibration of a single-beam optical gradient trap," *J. Phys.: Condens. Matter* **14**, 7757–7768 (2002).
21. N. B. Viana, M. S. Rocha, O. N. Mesquita, A. Mazolli, P. A. Maia Neto, and H. M. Nussenzveig, "Towards absolute calibration of optical tweezers," *Phys. Rev. E* **75**, 021914 (2007).
22. P. H. Jones, O. M. Marago, and G. Volpe, *Optical Tweezers: Principles and Applications* (Cambridge University, 2015), Chap. 2, Chap. 7–9.
23. P. Bartlett, S. Henderson, and S. J. Mitchell, "Measurement of the hydrodynamic forces between two polymer-coated spheres," *Philos. Trans. R. Soc. London, Ser. A* **359**, 883–895 (2001).
24. M. Tassieril, F. D. Giudice, E. J. Robertson, N. Jain, B. Fries, R. Wilson, A. Glidle, F. Greco, P. A. Netti, P. L. Maffettone, T. Bicanic, and J. M. Cooper, "Microrheology with optical tweezers: measuring the relative viscosity of solutions 'at a glance,'" *Sci. Rep.* **5**, 1–6 (2015).
25. P. Praveen, Yogesha, S. S. Iyengar, S. Bhattacharya, and S. Ananthamurthy, "Two particle tracking and detection in a single Gaussian beam optical trap," *Appl. Opt.* **55**, 585–594 (2016).
26. M. W. Allersma, F. Gittes, M. J. deCastro, R. J. Stewart, and C. F. Schmidt, "Two-dimensional tracking of ncd motility by back focal plane interferometry," *Biophys. J.* **74**, 1074–1085 (1998).
27. T. Li and M. G. Raizen, "Brownian motion at short time scales," *Ann. Phys.* **525**, 281–295 (2013).
28. K. C. Neuman and S. M. Block, "Optical trapping," *Rev. Sci. Instrum.* **75**, 2787–2809 (2004).
29. M. A. Zaman, P. Padhy, P. C. Hansen, and L. Hesselink, "Extracting the potential-well of a near-field optical trap using the Helmholtz-Hodge decomposition," *Appl. Phys. Lett.* **112**, 091103-4 (2018).
30. M. A. Zaman, P. Padhy, and L. Hesselink, "Fokker-Planck analysis of optical near-field traps," *Sci. Rep.* **9**, 1–11 (2019).