# Optical binding between dielectric particles 

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#### Abstract

We report observation of optical binding between two dielectric particles with dimensions less than the wavelength of the interacting light. The observed dependence of the separation of optically bound particles on the polarization of the trapping beam is in agreement with earlier theoretical predictions.


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OCIS Codes: (140.7010) Trapping; (290.5850) Scattering, particles.

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## 1. Introduction

Under intense optical field, as available in a line optical tweezers, existence of a force between dielectric microspheres of size larger than the wavelength has been reported [1, 2]. This results in discrete values for particle spacing. This optical binding is of considerable interest as it can be used for construction of two or three-dimensional arrangement of objects.

Theoretical treatments [1, 3, 4] available for interaction of point dielectric particles illuminated by a plane optical wave predict a periodic potential well for particle sizes satisfying the condition $k a \leq 1$ ( $k$ is wave number and a is size of the particle). For particle sizes such that $k a>1$, the dipole approximation is not valid and therefore the interaction potential should be different from that predicted for particle satisfying dipole approximation. Further, while the theory developed is for plane wave incident on two adjacent dipoles, the tightly focused interacting beam used in the experiment can not be treated as plane wave. These facts as well as the possibility of some artifacts in the experimental reports due to proximity of the trapped particles with a glass surface has led to some skepticism about the observation of optical binding [5]. So far optical binding for particles with size $<\lambda$ has not been reported. A major reason for this could be that as the size of the particle decreases, for inter particle separations larger than the particle sizes the depth of potential decreases [4]. This as well as the larger Brownian motion of smaller particles makes the measurements more difficult.

We report in this paper investigation of the motion of two polystyrene spheres with radius $\sim 150 \mathrm{~nm}$ trapped in line tweezers with 1064 nm Nd: YAG laser beam. The motion of the two particles was tracked for $\sim 25 \mathrm{~s}$ at video rate ( 25 frames per second) and was found to be
correlated. The histogram for particle separation showed distinct peaks, which correspond to separation of particles at approximately multiple of the wavelength of the interacting light. The dependence of these discrete separation states on the plane of polarization of the interacting beam was also investigated and the results are in qualitative agreement with earlier theoretical predictions based on interaction of plane wave with point dipoles [3].

## 2. Experimental methods

The line tweezers set-up used for this study comprised of an inverted microscope (Axiovert 135 TV, Carl Zeiss, Germany) to which a plane polarized 5W CW Nd:YAG laser (Solid State Laser Division, C.A.T., Indore, India) was coupled via the base port. A combination of cylindrical lenses and 100X microscope objective was used to generate the elliptic beam profile $(\sim 40 \mu \mathrm{~m} \times 0.8 \mu \mathrm{~m})$ at the sample. The long optical tweezers was created to ensure a weak optical gradient so that the optical binding force should dominate the optical gradient. The particles were trapped in three dimensions and the plane of trapping was kept $\sim 10 \mu \mathrm{~m}$ away from the surface of the lower coverslip so as to avoid the undesired frictional forces near the surface. This also minimizes the possibility of particles being influenced by the interference of the incident beam with the light scattered from the particles getting back reflected from the lower coverslip. The upper coverslip was positioned 5 mm away from the trap plane and should therefore have no influence on the trapped particles. Rotation of the elliptical trap beam profile via rotation of the focusing cylindrical lens was used to change the orientation of the E vector of the interacting light with respect to the major axis of line tweezers.


Fig. 1. Line scan of the intensity profile along the major axis of the line tweezers.
Experiments to observe optical binding were carried out on polystyrene particles with diameter 600 and 300 nm . These are smaller than the wavelength of Nd: YAG laser in water $(800 \mathrm{~nm})$. The microspheres were suspended in water with $100 \mu \mathrm{M}$ Phosphate Buffered Saline (PBS), having Debye length of $\sim 30 \mathrm{~nm}$. This was done to avoid the possibility of static monopole forces arising from any static charge on the spheres. The buffer has no effect on the long range interactions observed in optical binding. In order to eliminate any undesirable gradient forces along the axis of the line tweezers, the intensity profile of the trapping beam was recorded on a CCD and any spatial modulation of the laser beam intensity profile was minimized by careful alignment of the optics. The measured intensity profile (Fig. 1) was reasonably free of spatial modulations. The smoothness of the potential well of the line tweezers was further monitored by tracking the position of a single polystyrene sphere trapped in the tweezers for an extended period of time. The histogram obtained for the particle position showed no preferred spatial positions in the trap conforming smoothness of the potential well.

Two particles were trapped in the central region of the line tweezers and it was ensured that the sample was sufficiently dilute so that no third particle could come inside the trap for
the time duration of the observation. The time for which the motion of the two particles was tracked was 180 seconds for 600 nm particles and about 25 seconds for smaller particles. The maximum tracking time was constrained by the need to ensure that none of the two particles in the trap escape the trap or a third particle enters the trap because of their thermally induced Brownian motion. Motion of the trapped particles was recorded at video rate and digitized. Software performing centroid detection and thresholding to improve image contrast was used to monitor position of particle(s). For 600 nm diameter particles the position could be determined with a precision of about 25 nm . However, the precision in determination of the position of 300 nm diameter particle was $\sim 60 \mathrm{~nm}$ because of the poorer contrast of the image for smaller particle. For estimating the precision of positional measurements we fixed the particles on the coverslip by drying the solution and then recorded their images. The value of $2 \sigma$ ( $\sigma$ being the standard deviation) for position measurements from these images was taken as the precision for the measurement. The pixel size of the CCD used was 6.7 micron, magnification was 500 and the illumination intensity was $\sim 300 \mu \mathrm{~W} / \mathrm{cm}^{2}$.

It is pertinent to note that in contrast to typical point tweezers where the stiffness of trap along the axial direction is weaker [6], for the line tweezers used in the present experiment the stiffness of trap along the axial direction is about an order of magnitude larger than that along the major axis of the trap. Therefore, thermal fluctuations in the position of the particle in axial direction are expected to be much smaller than that along the major axis. Further, if the particle moves axially the contrast of the image of the particle decreases significantly and determining the position of the particle becomes difficult. Since we have investigated the optical binding forces only along the major axis of the trap where the gradient force is minimal, such events involving axial movement of the two particles were discarded by rejecting frames for which contrast was below some prefixed threshold. For 300 nm diameter particles of the total of about 625 events recorded in a scan of 25 s , the number of discarded events was less than 100 .

## 3. Results and discussion

For our experimental situation of a plane wave propagating along Z direction with wave vector $k$ normal to the vector separation $R$ of the two particles on the $x$-axis, the binding force along x between the point dipoles can be expressed as [3]:

$$
F_{x}\left(E_{x}, E_{y}, R\right)=\frac{\alpha^{2}}{2 R^{4}}\left[\begin{array}{l}
2 \cdot E_{x}^{2}\left\{-3 \cdot \operatorname{Cos}(k R)-3 k R \operatorname{Sin}(k R)+(k R)^{2} \operatorname{Cos}(k R)\right\} \\
+E_{y}^{2}\left\{3 \cdot \operatorname{Cos}(k R)+3 k R \operatorname{Sin}(k R)-2(k R)^{2} \operatorname{Cos}(k R)-(k R)^{3} \operatorname{Sin}(k R)\right\}
\end{array}\right]
$$

Here $\alpha$ is the polarizability of the particles given by the expression $\alpha=a^{3} \frac{\left(n^{2}-1\right)}{n^{2}+2}$, where a is the radius of the spherical particle, $n$ its relative refractive index i. e. the ratio of the refractive index of the particle with respect to the value of refractive index of the surrounding medium. $\mathrm{E}_{\mathrm{x}}$ and $\mathrm{E}_{\mathrm{y}}$ represent the component of the electric field of the wave along x and y .

Although for 300 nm particles interacting with 800 nm light, the point dipole approximation is not exactly valid, in absence of more accurate theoretical models, we use the available treatment to work out the optical force between two 300 nm particles for the conditions of our experiment. The results are shown in Fig. 2(a). For these calculations the value of relative refractive index was taken as $n=(1.57 / 1.33)$, and the power of the trapping beam at the specimen plane was taken as 255 mW , which corresponds to an intensity of $\sim 0.8$ $\mathrm{MW} / \mathrm{cm}^{2}$ in a spot size of $\sim 40 \mu \mathrm{~m} \times 0.8 \mu \mathrm{~m}$. Three orientations of electric vector of the trap beam with respect to the long axis of trap were investigated: a) the E vector perpendicular to the long axis of trap $\left(\theta=90^{\circ}\right)$, b) the $E$ vector parallel to the long axis of trap $\left(\theta=0^{\circ}\right)$ and $\left.c\right) E$ vector at an angle of 45 degree to the long axis of $\operatorname{trap}\left(\theta=45^{\circ}\right)$.


Fig. 2: (a) Optical binding force between two Rayleigh particles for three different angles of orientation $(\theta)$ of the electric vector of the interacting light with respect to the X -axis, the long axis of the optical tweezers, (b) Interaction potentials for the three different cases

In Fig. 2(b), we show for these three cases, the calculated interaction potentials between the particles, normalized with respect to $\mathrm{K}_{\mathrm{b}} \mathrm{T}$. Here, $\mathrm{K}_{\mathrm{b}}$ is the Boltzmann constant and T the ambient temperature, taken as $300{ }^{0} \mathrm{~K}$. Both the depth of the potential wells as well as the values of inter particle separations leading to potential minima can be seen to depend on the polarization of the interacting beam.

The two particles trapped inside the line tweezers undergo thermally induced Brownian motion, which is more vigorous for the smaller particles. However, since the potential energy of the two particles depends on their separation (Fig. 2(b)), the particles are more likely to be found with their separations corresponding to the potential minima. The relative probability of finding a given separation (R) between the particles will be proportional to $\exp (-V(R))$. The resulting probability distribution for the case with electric vector of the trap beam being perpendicular to the major axis of the trap $\left(\theta=90^{\circ}\right)$ is shown in Fig. 3(a). The experimentally measured histogram of inter particle separation obtained from a record of 550 frames (~ 22 sec ) shown in Fig. 2(b) is in reasonable agreement with the theoretical estimates shown in Fig. 2 (a). The bin size for the histograms was taken to be 125 nm , the $2 \sigma$ values for measurement of particle positions.

Several distinct bound states corresponding to particle separation of approximately integral multiples of wavelength can be observed in Figs. 3(a) and (b). Since the optical binding potential well is strongest for lower inter particle separations, frequency of finding particles with separation of a lower integer multiple value of $\lambda$ is observed to be more than that for higher multiple of $\lambda$. Further since the width of the histogram peaks in Fig. 3 (b) can be attributed to thermal fluctuations as well as the weak gradient force, for bound states with higher inter particle separation; the ratio of the peak height to width of the peaks is significantly lower than that for bound states with lower inter particle separation. We also used the experimental histograms for the distribution of particle separation to estimate the potential in the trap [7]. Potential wells corresponding to the position of histogram peaks were observed with the largest potential well depth being about $2 \mathrm{~K}_{\mathrm{b}} \mathrm{T}$.


# Distance between centre of the particles (in units of $\lambda$ ) 

Fig. 3. (a) Probability distribution of separation between two 300 nm particles for the case when the electric vector of the trapping beam was orthogonal to the long axis of the optical tweezers $\left(\theta=90^{\circ}\right.$ ), (b) Measured histogram of the distance between center of two particles for $\theta=90^{\circ}$.

In Fig. 4(a), we show the relative probability distribution of the separation between two 300 nm particles for electric vector of the trap beam oriented parallel to the major axis of the trap $\left(\theta=0^{0}\right)$ and in Fig. 4(b), the experimentally measured histogram of inter particle separation. It is important to note that here the frequency of the first two bound states is considerably larger than that for Fig. 3(b). Further, the two peaks corresponding to these bound states are broader and have significant overlap. This is because for this polarization configuration the optical binding potential wells are much shallower compared to the case when electric vector is perpendicular to the trap major axis (see Fig. 2). Therefore, the gradient and diffusive forces would be able to overcome the rather weak potential wells corresponding to larger inter particle separation leading to an increase in the frequency of the bound states with inter particle separation of approximately $\lambda$ or $2 \lambda$. We should also emphasize that in agreement with the theoretical predictions, the positions of the peaks in Fig. 4(b) (where E is along $x$ ) occur at different inter particle separation compared to that for the
case when where E is along y (Fig. 3(b)). However, the separation of the peaks in both the cases is roughly an integral multiple of $\lambda$.


Distance between centre of the particles (in units of $\lambda$ )

Fig. 4. (a) Probability distribution of separation between two 300 nm particles for the case when the electric vector of the trapping beam was parallel to the long axis of the optical tweezers $\left(\theta=0^{\circ}\right.$ ), (b) Measured histogram of the distance between center of two 300 nm particles $\theta=0^{0}$.

The results of measurements with electric vector at an angle of 45 degrees to major axis of the trap are shown in Fig. 5. We note that in agreement with the theoretical predictions, the positions of the peaks in Fig. 5(b) occur at inter particle separations in between the case where $E$ is along $x$ or $y$.

Larger particles (diameter ~ 600 nm ) trapped inside the line tweezers show a less vigorous thermally induced Brownian motion ( $\sim 2 \mu \mathrm{~m} / \mathrm{s}$ ) as compared to that of smaller particle (diameter $\sim 300 \mathrm{~nm}$ ). This and the better image contrast obtained for the bigger particles makes tracking their motion considerably easier. The tracked motion of these particles inside the trap is shown in Fig. 6(a) for the case where the electric vector of the trapping beam was perpendicular to the major axis of the trap. Two distinct bound states with separation of $\sim 0.6 \mu \mathrm{~m}(0.75 \lambda)$ and $\sim 1.8 \mu \mathrm{~m}(2.25 \lambda)$ can be seen. The histogram of inter particle separation is shown in Fig. 6(b). One can note the larger disagreement with the
theory for Rayleigh particles (see Fig. 1(b)) in that the bound states are not periodic in wavelength.


Fig. 5. (a) Probability distribution of separation between two 300 nm particles for the case when the electric vector of the trapping beam was parallel to the long axis of the optical tweezers $\left(\theta=45^{\circ}\right)$, (b) Measured histogram of the distance between center of two 300 nm particles $\theta=45^{\circ}$.

Results presented in Fig. 6 suggest that in the experiments of Burns et al performed using $\lambda=0.387 \mu \mathrm{~m}$ and 1430 nm sized particles ( $k a>2 \pi$ ), the measured inter particle separation of bound states should deviate significantly from the predictions of theory based on interaction of plane wave with Rayleigh particles. It is pertinent to note that Burns et al. used the approximate expression for interaction potential $\left(W_{\text {approx }}(x, a)=-\frac{1}{2} \alpha \cdot E^{2} \cdot k^{2} \cdot \frac{\operatorname{Cos}(k x)}{x}\right.$, Eq. (2) of Ref. [1]) to predict periodicity of $\lambda$ in the inter particle separation. However, the approximate expression is valid for very large inter-particle separations [3] and for their experimental conditions, the estimates for potential using their exact expression (Eq. 1 of reference 1) deviates significantly from that obtained using the approximate expression and does not predict the periodicity of $\lambda$ in the inter particle separation. For making a valid comparison between experimental results, the theoretical treatment should be extended to deal with larger dimension particles and also take account of the fact that in the focusing geometry
used in the experiments the incident wave is not plane but consists of a broad spectrum of plane waves


Fig. 6. (a) A representative track of two 600 nm particles for 14 sec , (b) Histogram of distance between center of two 600 nm particles for the case when the electric vector of the trapping beam was orthogonal to the long axis of the optical tweezers $\left(\theta=90^{\circ}\right)$.

## 4. Conclusions

To conclude, optical binding of dielectric particles with dimensions less than the wavelength of the interacting light has been demonstrated using a line tweezers with Nd: YAG laser as the trapping beam. The measured inter particle separations for 300 nm particles are in qualitative agreement with the theoretical treatment developed under dipole approximation for interaction of dielectric objects with plane wave. However, significant differences were observed for 600 nm particles. The observed dependence of the separation of optically bound Rayleigh particles on the polarization of the trapping beam was also in qualitative agreement with earlier theoretical predictions. This dependence provides an additional control on optical interaction forces and can help organize microscopic objects with sub wavelength accuracy.

## Acknowledgments

Authors would like to thank Shri T.P.S. Nathan, Head, Solid State Laser Division and members of his division for providing the Nd: YAG laser and their help in smooth running of the laser. Authors would also like to thank H.S. Patel and N. Ghosh for useful $r^{\prime}$ 'cussions.

