Abstract

There is presently an increased research activity in understanding the nature of optical force when ultrashort pulsed excitation is used to trap and manipulate objects with sizes ranging from micrometers down to nanometers. Such a "fem- tosecond laser tweezer" is peculiarly promising because the nature of the force can be dramatically tuned owing to optical nonlinearity. In this thesis, we have studied the role of optical Kerr effect in laser trapping theoretically as well as experimentally. In the first part of this thesis, we have shown numerical simulations using different theories such as dipole approximation formulation, geometric optics formulation, generalized Lorenz Mie theory (using localized approximation), and exact Mie theory under both continuous-wave (CW) and pulsed excitation for dielectric as well as metallic particles. It has been shown that owing to optical nonlinearity, the escape potential (the height of the axial trapping potential barrier along the beam propagation direction), not absolute potential (the absolute depth of the axial trapping potential), is the relevant parameter for stability of an optical trap created by a train of femtosecond laser pulses. We have optimized the average power and particle size by fixing the other parameters in numerical simulations for micron to nanometer-sized particles. Also, we have demonstrated that the optical trapping force/potential can be reversed (from repulsive to attractive), upon switching from CW to pulsed excitation. The results open up the possi- bility of utilizing optical nonlinearity for facile optical manipulation/sorting by controlled reversal of optical force. Later, we have extended our study from dielectric to metal nanoparticles and observed that the initial disappearance of trapping potential well along the axial direction with an increase in laser power but subsequent reappearance at higher laser power. These studies show how one can harness optical Kerr effect to fine-tune the stability of an optical trap and thereby have controlled optical manipulation. In the second part of this thesis, we have performed experiments to test the theo- retical results. We have designed and built a complete table-top optical tweezer set-up with versatile detection modalities: wide-field detection mode using camera xviirequired for spatial resolution and point detection mode using photomultiplier tubes for temporal resolution. To quantify the pulse-width at the sample posi- tion of optical tweezer set-up, we have used collinear two-photon fluorescence (TPF) autocorrelation. Firstly, we have explored the physics of the nonlinear nature of optical trapping force/potential under ultra-short pulsed excitation for micron-sized particles. Thus, we have presented the very first attempt in building a bridge between nonlinear optical phenomena and optical trapping by a combination of theory and experiment. Here, we have provided a model to elucidate sequential events (drag, adjustment, equilibration, fluctuation and ejection) in optical trapping dynamics and showed how we can map the highly asymmetric axial potential created by a femtosecond pulse-train. Later, we have extended our study from micron to nanometer-sized dielectric particles. We have shown that while TPF signal decays over time due to photobleaching but this signal is useful to know whether a particle is dragged towards the trap, in contrast, backscattered signal provides detailed information about the particle’s dynamics inside the optical trap. Therefore, a simultaneous detection set-up is essential to capture the trapping events of fluorescent particles. Considering fine-tuning of trap-stiffness through optical nonlinearity, we envision far-reaching applications of using ultra-short pulsed excitation in laser trapping and manipulation.