Understanding femtosecond optical tweezers: the critical role of nonlinear interactions

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Abstract. Typical single-beam optical tweezers use continuous wave (CW) lasers, which can be explained through force balancing the light pressure from a tightly focused laser beam used for trapping microscopic particles. Recent years have also seen a surge in single-beam optical trapping research with high-repetition-rate femtosecond lasers that has shown certain differences from the CW tweezers, one of which is its sensitive detection capability of the ultrashort pulse induced background free two-photon fluorescence signals. The high peak power of each laser pulse is enough to provide instantaneous trapping potential, while the high repetition rate ensures sustained stable trapping from the successive pulses. Though the capability and usefulness of the optical-tweezers are well established, for both CW and pulsed lasers, simulating real-time scenarios to predict optical trapping behaviour remains a challenging problem. This is especially true for femtosecond laser tweezers since high peak powers are involved when the laser is tightly focused for achieving the tweezing action. The nonlinear optical effect and thermal nonlinearity become much more significant for femtosecond optical trapping. We demonstrate the importance of including these nonlinear interactions for femtosecond pulsed laser mediated optical trapping via their effect in scattering and gradient forces in the Rayleigh regime. Our optical-tweezers model includes thermal and optical nonlinear interactions, making it easier to predict the optical-trap stability in real optical trapping scenarios for both CW and pulsed lasers. Our model provides predictive metrics for choosing solvents, probes, and several optical parameters, which can be validated from our experiments.

1. Introduction
The capabilities and utility of the single-beam optical tweezer technique are well recognized [1,2]. Optical trapping is a technique, which is used for trapping small particles over a modest range of sizes, from tens of microns to tens of nanometers. A token of the method’s remarkable efficiency is the fact that only a few milliwatts of laser power is sufficient to trap a particle in a single-beam optical tweezer setup. This corresponds to radiation forces, which are in the pico-Newton range. However, numerically and theoretically accessible models to simulate real-time prediction of optical trapping behaviour are challenging and typically incorporate unrealistic simplifications like ignoring the localized heating of the medium surrounding the laser focus during optical trapping experiments [3]. In fact, there have been almost no efforts towards uncovering the thermal effects on the force and potential of trapped particle systems. Recent years [4,5] have seen an uptick in the usage of high-repetition-rate (HRR) femtosecond lasers for the single beam optical trapping research that has certain differences from the continuous wave (CW) tweezers. One of the differences that has often been utilized is the enhanced sensitivity of HRR femtosecond laser tweezer setup compared to traditional CW laser tweezers arises from the ultrashort
pulse, which induces background free two-photon fluorescence signals [5]. The CW optical tweezers mostly rely on light-scattering techniques [6] for measurements that often have considerable background noise to contend with. The study of thermal effects in optical trapping becomes even more relevant, as it is an omnipresent effect across both CW and pulsed setups, albeit with varying contributions. A previous study [7] also showed that the scattering force exerted by femtosecond pulsed laser was much higher than the CW tweezers.

For a tightly focused beam as used in optical tweezers, cumulative heating can occur despite the minimum absorption cross-section of the trapping medium or the trapped particle, which reaches its maximum near the focus [8]. A temperature gradient from the laser focal spot is thus generated outwards from the laser focus in the medium, which, in turn, creates a refractive index gradient across the focusing region. The refractive index attains a minimum at the focus, gradually increasing as a function of distance away from it. Since the trapping force and potential depend on the refractive index of the medium [6], the thermal effect impacts the force and potential of the trapped particle significantly. With CW lasers, computational evidence of temperature rise at the focus of optical tweezers has been measured [9], which, unfortunately, is not a feasible approach for ultrafast lasers, given the computational complexities. A better understanding of high photon flux induced processes and a working model of the single-beam optical tweezers that could address both CW and pulsed lasers would be ideal for uncovering the effects of this inherent thermal gradient of the optical tweezers.

In the theoretical approach, that we present here, we have included all possible nonlinear effects arising due to high photon flux interactions. This allows a coherent treatment of both CW and ultrafast cases. We have the purely thermal type nonlinear effects for the CW laser case, while for the ultrafast laser case, we include both the thermal and the Kerr type nonlinearities [10].

2. Theoretical Formulation

Stable optical trapping is essentially the result of a balancing act between the two types of optical forces that act in tandem on a trapped particle. There is the scattering force that operates in the direction of the propagating beam. The gradient force, however, pulls the particle towards the focus if its refractive index is higher than the trapping medium and repels particles with lower refractive index (relative to the trapping medium). In order to achieve tight focusing for optical tweezers, a high numerical aperture (NA) microscope objective is used. For the case of any particle with a higher refractive index than the trapping medium, this ensures that the gradient force effectively negates the scattering force, which results in the formation of a stable trap.

A smooth beam profile is an additional necessity as it is desirable for a continuous and smooth gradient force to act on the trapping particle. The magnitude of force transfer to the particle is described in terms of the momentum, \( p = \frac{\hbar}{\lambda} \) from the laser radiation, and as discussed in the earlier paragraph, the direction of the force depends on the relative refractive index of the particle and the trapping medium [6]. The effective force calculation, therefore, depends on the particle size. Here, we consider the Rayleigh regime of optical trapping for which the particle size is much smaller than \( \lambda \), the wavelength of the trapping laser. The particle then acts like a dipole, with forces along the axial direction consisting of the scattering force (\( F_{sc} \)) and the gradient force (\( F_{gr} \)). Mathematically, \( F_{sc} \) is proportional to the laser intensity (\( I \)) while \( F_{gr} \) is proportional to the gradient of the laser intensity (\( \nabla I \)) [7,10,11]:

\[
F_{sc} = \frac{128\pi r^6 n_m}{3\lambda^4 c} \left[ \frac{m^2-1}{m^2+2} \right]^2 I
\]

\[
F_{gr} = \frac{2\pi n_m r^3}{c} \left[ \frac{m^2-1}{m^2+2} \right] \nabla I
\]

Where the ratio of the refractive index of the particle, \( n_p \), to that of the medium, \( n_m \), is defined as \( m \). The particle radius is \( r \), and \( c \) is the speed of light. Typically, the refractive indices, \( n_p \) and \( n_m \), are the linear refractive indices, which are characteristic properties of a material. However, in our formalism here, we include all possible nonlinear effects, and this modifies the effective overall refractive index (\( n_{Total} \)) to be a function of the incident laser intensity (\( I \)) as [12]:

\[

\]
\[ n_{\text{Total}} = n_0 + n_2 I \]  

In this case, \( n_0 \) is the linear refractive index, and \( n_2 \) is the nonlinear refractive index coefficient (unit in \( \text{m}^2/\text{W} \)) that arises from the consideration of nonlinear effects that could be optically and/or thermally induced. The \( n_2 \) can be either positive or negative, depending on the type of nonlinearity. Thus, an electronically induced nonlinearity results in a positive \( n_2 \) that acts at a fast timescale [13]. On the other hand, a negative \( n_2 \) may signify nonlinear heating occurrence due to thermal effects that are nonelectronic in nature and have slow timescales [14]. While thermal effects are omnipresent in both CW and ultrafast laser systems, the Kerr type electronically induced nonlinearity gains prominence for femtosecond laser pulsed tweezers where the peak intensities are large enough for this to be of significance. In this paper, we consider electronically induced optical nonlinearity to result in the modified refractive index of the trapped particle \((n^p_{\text{Total}})\) while the thermally induced nonlinearity to be the reason for the modified refractive index of the trapping medium \((n^m_{\text{Total}})\), since a trapped particle is essentially a solid in a fluid medium. Thus, we have three distinct conditions, which we refer as:

a) Pure: when no nonlinear effects are considered, i.e., calculations are based on \( n_p \) and \( n_m \),

b) NLO: when we consider only the optical nonlinear effect, i.e., calculations are based on \( n^p_{\text{Total}} \) and \( n_m \), and

c) NLO+Thermal: when we consider both the optical nonlinear effect as well as the thermal effect, i.e., calculations are based on \( n^p_{\text{Total}} \) and \( n^m_{\text{Total}} \).

The experimental system that is being modelled consists of spherical polystyrene beads dispersed in pure water. Our particle sizes for this theoretical study, 5 nm and 80 nm have been chosen to span the Rayleigh range criteria, which correspond to laser powers of 5 mW and 100 mW for both the CW and femtosecond pulsed laser cases. The linear refractive indices of polystyrene particle and water medium are 1.578 and 1.329, respectively, and the nonlinear refractive index coefficient of the polystyrene particles are \( 5.17 \times 10^{-17} \text{ m}^2/\text{W} \) [15]. We have earlier conducted experiments with 50 nm radius latex beads [4,5] with a HRR femtosecond laser operating at 800 nm. We perform axial force calculations for various particle sizes as a function of displacement from the mean position. For stable trapping to occur, not only the gradient force should dominate but also the corresponding Boltzmann factor resulting from the potential \((U)\) of the gradient force \((F_{\text{gr}})\), should follow: \( \exp \left( -U/k_BT \right) \ll 1 \), at temperature \( T \), where \( U \propto n_m I \) and \( k_B \) is the usual Boltzmann constant. The refractive index of the medium, temperature and a low number density of the target particles to be trapped pay a significant role. We use a Gaussian beam profile for the intensity distribution for a given laser power, \( P \). For a CW laser, \( P \) represents the average power while for a pulsed excitation, it is replaced by the peak power: \( P_{\text{peak}} = \frac{P}{f_t} \). For the femtosecond pulsed laser experiment, the repetition rate \((f)\) is chosen as 76 MHz, and pulse width \((\tau)\) is taken to be 150 fs. For both the CW and femtosecond lasers, we use the central wavelength of 800 nm that is tightly focused with a numerical aperture (NA) of 1.2, to determine the Gaussian beam of the TEM\(_{00}\) mode as: \( w_0 = \frac{0.82 \times \lambda}{n_m \text{NA}} \). The scattering and gradient forces (eqs. (1) and (2)) can be found for the Rayleigh region as a function of displacement from its mean position, while their integrals correspond to the respective potentials [10] for all the three cases, viz. (a) Pure, (b) NLO and (c) NLO+Thermal described earlier.

A typical representative plot of the axial force and the corresponding potential energy calculated based on this model presented here is shown in Figure 1 for a Rayleigh particle of 75 nm radius bead in water trapped with a CW laser operating at 800 nm. On careful inspection of Figure 1, we note that there is no impact of NLO but there is indeed an impact of thermal effect, which is as per expectation for the CW tweezers. The trapped particle can show motion in the potential inside the laser focus, which can be visualized as the total potential plotted in Figure 1. If the particle has energy greater than that of the total potential well depth as provided by the optical trapping, then the bead can exit the trap and become un-trapped. The escape potential or the potential depth is the minimum energy required by the
particle to get un-trapped from the potential. Being a differential measure, escape potential is often a more convenient way to understand the characteristics of a trapping process, which we will use hereafter.

![Figure 1](image)

**Figure 1.** Plot of (a) axial force and the corresponding (b) potential for a 75 nm radius bead size for a CW laser trap at 800 nm at 10 mW average power. The plot labels Case A, Case B and Case C represent, respectively: (A) Without incorporating any nonlinear effects (pure), (B) Considering optical (Kerr) nonlinearity only, (C) Including both thermal and optical (Kerr) nonlinearity.

3. Results and Discussions

We inspect the variation of axial forces and trapped particle escape potentials as we increase the trapping particle size at several fixed average powers first for CW laser trapping followed by the femtosecond laser trapping. As discussed in the earlier section, we consider the three situations, which are demarcated by the inclusion of nonlinear effects, that is (a) not incorporating any nonlinear effects (i.e., the pure or the ideal case) versus (b) when we introduce the Kerr nonlinearity only, and finally (c) when we consider the inclusion of all thermal effects in addition to the Kerr nonlinearity (both nonlinear effects are included). As expected, for the CW laser, case (a) and case (b) are essentially indistinguishable, while both of them differ from (c). The effects are more pronounced at higher average laser powers. It is also important to note that for Rayleigh regime particles, since the particle size is very small, the thermal characteristics of the solvent is very important, as it occupies a major portion of the focal volume.

In order to understand and visualize these results better, we view our results in terms of the relative change in escape potential of the trapped particle, which also provides a measure of the stability of the trapped particle. Let us define:
$E_{esc}^{pure}$ = escape potential when no nonlinear effects are included in the calculation.
$E_{esc}^{Kerr}$ = escape potential when only Kerr nonlinear effect is included in the calculation.
$E_{esc}^{both}$ = escape potential when both Kerr and thermal nonlinear effects are included in the calculation.
$\Delta E_{esc}$ = $(E_{esc}^{both} - E_{esc}^{pure})$ = relative change in escape potentials.

Figure 2 shows the results for the CW laser trapping case. As per our discussions, only the thermal process dominates and there is no effect of the Kerr effect in this case and so it is not possible to distinguish between 2a and 2b. However, distinction occurs from the thermal effect, which makes Figure 2c distinct from the other two.
Figure 2. Trapping particle size-dependent variation of axial escape potentials in CW laser trapping for: (a) pure trapping case without Kerr or thermal effects ($E_{esc}^{pure}$) (b) with only Kerr effect ($E_{esc}^{Kerr}$) included, and (c) with both thermal and Kerr effect included ($E_{esc}^{both}$).

For the femtosecond laser tweezer, however, the effect of Kerr nonlinearity is significant, and since there is a change in the sign of the nonlinear refractive index coefficient on moving between the Kerr nonlinearity and the thermal nonlinearity, the results are quite striking and nonintuitive as shown in Figure 3.
\[ \Delta E_{\text{Esc}} = E_{\text{Kerr}} - E_{\text{Pure}} \]

Change in Escape Potential \((10^{-15})\)

Radius (nm)

(a)

\[ \Delta E_{\text{Esc}} = E_{\text{Both}} - E_{\text{Kerr}} \]

Change in Escape Potential \((10^{-15})\)

Radius (nm)

(b)
Figure 3. Trapping particle size-dependent variation of axial escape potentials for femtosecond pulsed laser trapping (point to note is that the pure trapping case without any nonlinear effect is identical to Fig. 1a). Here we present all the relative escape potentials including nonlinear effects: (a) decrease in escape potential due to Kerr effect relative to the pure one \( \Delta E_{esc} = \{E_{esc}^{Kerr} - E_{esc}^{pure}\} \), (a) decrease in escape potential due to thermal and Kerr effect relative to the Kerr only case \( \Delta E_{esc} = \{E_{esc}^{Both} - E_{esc}^{Kerr}\} \), and (c) decrease in escape potential due to thermal and Kerr effect relative to the pure one \( \Delta E_{esc} = \{E_{esc}^{Both} - E_{esc}^{pure}\} \).

Our theoretical exploration shows the important balancing act that occurs between the two nonlinearity inducing effects. Thus, we note that there is a bead size corresponding to a maximally stable trap for a given working power of the pulsed laser in the Rayleigh region.

4. Conclusions

We have demonstrated the importance of inclusion of the nonlinear interactions for both CW and femtosecond pulsed laser optical trapping in the Rayleigh regime. While the Kerr type nonlinearity has little impact on the CW trapping, the joint effect of the Kerr nonlinearity and thermal effects has a dampening effect on the escape potential values. The competing effect of the two nonlinear processes is even more pronounced for the femtosecond laser tweezer, and there exists a bead size of maximum stability for a given working power of the pulsed laser in the Rayleigh region. We demonstrate the existence of this inflection point by virtue of their differing signs, and this effect, arising from a full treatment of nonlinear effects and heating process has a pronounced effect on the trapping stability. Both the effects of the nonlinear processes can cancel each other so as to create a pure trapping scenario where the thermal effect is nullified by the Kerr effect. Further work on this is currently in progress in our laboratory for various bead sizes, and we have also found that the power range for maximum stability of 250 nm diameter trapped bead is also in agreement with the experimental observation in femtosecond pulsed laser operating at 800 nm centre wavelength.

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