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Manifesting the Effects of Thermal Nonlinearity in Optical Trapping for Rayleigh Regime

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Abstract

Since long the thermal effects have not been much explored in the ***optical trapping*** theory, in this paper, we are ***establishing the effects*** of optically induced thermal nonlinearity in the medium of optical trapping in the Rayleigh regime for both continuous wave and a pulsed laser. For a single beam, optical tweezers with high numerical aperture (N.A.) objectives are used as a routine. In such a tight focusing scenario, both optical nonlinearity and thermal effects may prevail in the cases of continuous wave (C.W.) and pulsed laser-mediated optical trapping events. In this paper we will introduce the effects sequentially, starting from optical nonlinearity and methods to implement this effect and subsequently introduce the thermal nonlinearity in the medium. The effects are significantly different when compared between ***CW and pulsed optical tweezers*** and will be discussed in detail in this paper.

1. Introduction

Nobel prize for the year 2018 made the discovery of ***Optical Tweezer*** (OT) by Arthur Ashkin more prominent and brought his important discovery worldwide accolade. At the same time, the shared Nobel prize for ultra-fast lasers made it obvious for groups like ours who are using ultra-fast lasers in optical trapping to take the conjugation of two discoveries to the next level. Theoretical development for OT had been started years ago to simulate real experimental scenario. On this regard optical (Kerr) nonlinearity and its effect on OT have been discussed by some scientists. Discovery of optical non-nonlinearity has not only revitalized the study of light-matter interaction but also has provided a new basis for many exciting applications such as nonlinear optical modulation, Optical switching, optical delays, etc. But since so long, the effects of thermal nonlinearity were being subdued in optical tweezers theory. The inclusion of nonlinear optics in the study of forces on optically trapped particle before this work introduced the effects of electronic nonlinearity (Kerr Effect) [1,2], but this electronic nonlinearity is not the only nonlinear effect present in a medium, and thermal nonlinearity also plays a major role [3]. We show here that thermal effects in the medium play a prominent role in both CW and pulsed laser-mediated optical traps for Rayleigh range particles. In our numerical method, we have used a pulsed laser with a central wavelength of 780nm, a repetition rate of 76 MHz with a pulse width of 160 fs. The CW laser source is also set at 780nm, and the average power for both cases are kept the same for thorough comparison for the 60nm particle.

2. Measuring Thermal Nonlinearity

Interaction of high-intensity light with the material through which it propagates results in changing the properties of the material which in turn results in the generation of higher order harmonics. For the case of nonlinear interaction, induced polarization in the material is given as:

$$P(t) = \varepsilon_o[\chi^1 E(t) + \chi^2 E^2(t) + \chi^3 E^3(t) \ldots.$$

Here $\chi^n$is the higher order nonlinear susceptibility of the material with n representing the order of nonlinearity. E(t) is the incident electric field which in our case is the electric field for a Gaussian TEM_{00} beam. Here second order nonlinearity is zero for the materials showing inverse symmetry however third order nonlinearity is present irrespective of material showing inverse symmetry or not. Contribution of third order nonlinearity is given by:

$$P^3(t) = \chi^3 E^3(t)$$

Now total refractive index in the presence of optically induced nonlinearity can be given as:

$$n = n_o + n_2 I$$

here, n_o is the constant linear refractive index of the material and n_2 is the strength of refractive index contributed by the nonlinearity in the material. Where n_2 is given by:

$$n_2 = \frac{.0395}{{n_o}^2}\chi^3(esu)$$

The more detailed theory about optical nonlinearity can be found in reference [4]. Due to this intensity-dependent refractive index of the material, the material under the illumination of light now acts as the self-focusing and self-defocusing lens. When n_2 is positive, the material starts to show self-focusing and for negative n_2 material starts showing self-defocusing. Although we are not considering

any nonlinear absorption in our case, one should notice that there are materials which also shows nonlinear absorption property [3]. Nonlinearity in a material can be introduced electronically as well as non-electronically [5]. Electronically induced nonlinear responses are very fast as compared to non-electronically induced nonlinear responses. There are multiple methods available to measure the nonlinearity in materials, but none of these methods can be used to separate the various nonlinear responses unambiguously. Both of the nonlinearity mentioned above (fast and slow nonlinearity) are simultaneously present in the material, but there are ways of finding purely electronic nonlinearity [6]. Although we agree to the different techniques present for measuring nonlinearity we are using the Z-scan method to determine the nonlinearity in our case due to the simplicity of this method.

In the Z-scan technique, focused light is allowed to pass through a moving sample, and the light beyond the sample is allowed to fall on two detectors via a beam splitter for measuring open aperture and closed aperture transmittance through the sample. Open aperture is mainly used for determining the nonlinear absorption coefficient of the material, and the closed aperture is used to determine the nonlinear refraction in the material. In our case since we are taking a mixture of ethanol and water (1:4), we have no nonlinear absorption, and the only nonlinearity present is in the form of nonlinear refraction. The experimental setup details can be found in the earlier work [7]. As the focused beam of laser passes through the material, there is a nonlinear phase shift. The peak nonlinear phase shift is given by:

$$\Delta\varphi_o = k_o n_2 I L_{eff}$$

Here k_o is the wave propagation constant, n_2 is the nonlinear refractive index, I is the incident intensity given as $(2P/(\pi\omega_o^2))$for CW laser and $(2P/(\pi\omega_o^2 f\tau))$ for pulsed laser and L_{eff} is the effective thickness of the material sample. Since we are taking a thin sample, $L \leq Z_o$ where Z_o is the Rayleigh range given by $\pi\omega_o^2/\lambda$. The transmittance at the detector is given by:

$$T(z) = \frac{\int_{-inf}^{inf} P_T(\Delta\varphi_o(t))dt}{S\int_{-inf}^{inf} P_i(t)dt}$$

Where $P_i(t)$ is the instantaneous input power, and S is the aperture linear transmittance. For the case of Gaussian beam transmittance is:

$$T(z) = 1 - \frac{4\Delta\varphi_o \frac{z}{Z_o}}{\left[\left(\frac{z}{Z_o}\right)^2 + 9\right]\left[\left(\frac{z}{Z_o}\right)^2 + 1\right]}$$

As $\Delta\varphi_o$ is a function of n_2, fitting the data obtained in z-scan to the above equation gives us the value of n_2 (n_2 can be both positive as well as negative). As discussed earlier that in a sample there are both electronic and non-electronic nonlinearity present., and generally non-electronic

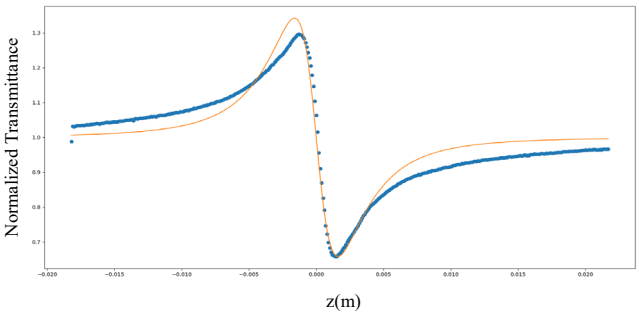


Figure 1 Transmittance obtained by experiment and fitting curve for pure ethanol

nonlinearity is due to thermal effects in media um which takes place as a result of heating of material as light propagates through it. In general cases, thermal nonlinearity is greater than the electronic nonlinearity but in the case of the pulsa ed laser, the two may be comparable. The general way of distinguishing between electronic and thermal nonlinearity is the sign of n_2. If the sign is positive, then it is an electronic nonlinearity and if the sign is negative, it is thermal nonlinearity. A typical graph for close aperture single beam z-scan data for ethanol is shown in **Figure 1**.

3. Theory of Optical Tweezer

CW lasers have been conventionally used for a long time now for trapping and manipulating the particle of various sizes, in the recent years effective trapping using low power high repetition femtosecond lasers have been developed and delved thoroughly by researchers. Recently effects of Kerr effects were observed for particles of an arbitrary size and studied extensively. We now add to it the effects of thermal nonlinearity in the Rayleigh regime. As described by Arthur Ashkin there are mainly two forces acting on a particle in Rayleigh regime. These are gradient and scattering forces. We are going to discuss the forces in the longitudinal (axial) direction. For a pulsed laser these forces can be given by [8, 9]:

$$F_{scat.}(z=0,r) = \frac{8\pi n_m (ka)^4 a^2}{3c}\left(\frac{m^2-1}{m^2+2}\right) I_o \frac{1}{1+2\tilde{z}^2} \exp\left(\frac{-2\tilde{x}^2}{1+4\tilde{z}^2}\right)$$

$$F_{grad.}(z=0,r) = -\frac{2\pi n_m a^3}{c}\left(\frac{m^2-1}{m^2+2}\right) I_o \frac{1}{1+2\tilde{z}^2}\left(\frac{\frac{8\tilde{z}}{k{\omega_o}^2}}{1+2\tilde{z}^2}\right)\left(1 - \frac{-2\tilde{x}^2}{1+4\tilde{z}^2}\right)$$

Here I_o is the average intensity for CW and peak intensity for pulse laser given by:

$$I_o = \frac{2P}{\pi\omega_o^2} \; and \; I_o = \frac{2P}{\pi\omega_o^2 f\tau}$$

Also note here $\tilde{z}$ is z/(kω_o^2) and m is the ratio of refractive index of the particle to the refractive index of the medium. larger than the scattering force. Here refractive indices of the particle (n_p) and medium (n_m) are given by:

$$n_p = n_{po} + n_{p2} I(z,r,t)$$
$$n_m = n_{mo} + n_{m2} I(z,r,t)$$

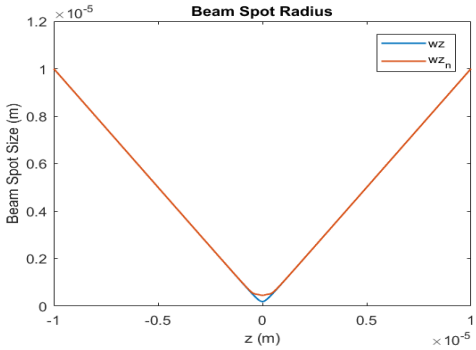
Here n_{po} and n_{mo} are the constant linear refractive index of the particle and medium respectively and n_{p2} and n_{m2} are the nonlinear refractive index of the particle and medium respectively. n_{p2} is due to the electronic nonlinearity (Kerr effect) while n_{m2} is due to the thermal nonlinearity. Thus n_{p2} is positive while n_{m2} is negative. In the case of CW case we have seen previously that researchers ignore the Kerr effect but thermal nonlinearity cannot be neglected in either case.

4. Results

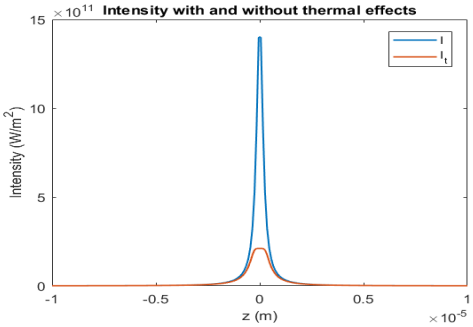
By the introduction of thermal nonlinearity for CW trapping case, the immediate effect that can be seen (figure 2a) is the change in beam waist .136um. The axial intensity distribution gets affected as a whole, which is shown in figure 2b. The apparently flat part is due to the Rayleigh region of focus.

In our case, we have taken a CW laser with a power of 82.45 mW at 780 nm and a numerical aperture of 1. We have taken a polystyrene particle of $n_{po} = 1.578$ and the medium is a mixture of water and ethanol with $n_{mo} = 1.329$, $n_{p2} = 5.9 \times 10^{-17}$ and $n_{m2} = -3.71 \times 10^{-12}$. The respective values have been used from references [10, 11]. Axial forces and axial potentials for CW laser trapping is shown in figure 3a and 3b respectively with color coding for each type of forces and potentials for the incorporation of Kerr effect only. For CW laser source, the Kerr effect is negligible and it basically shows the normal forces and potentials one can obtain from Rayleigh theory. It is observed that the nature of the scattering force changes completely due to addition of thermal effects. The gradient force is much higher compared to scattering force and is the major contributor towards the total force, responsible for stable trapping. When the thermal nonlinearity in the sample is added the results become significantly different. The nature of the scattering force changes completely due to change in refractive index of the medium. The gradient and total forces also decrease substantially. This is shown in figure 4(a). The potential and the escape potential also get affected as can be seen from figure 4(b).

For the case of ***pulsed excitation,*** Kerr effect already affects the total force without the incorporation of thermal effects alone (figure 5). The forces and potentials resulting after the inclusion of Kerr effect for pulsed trapping case is shown in figure 5(a). The next figure (figure 6) shows the effect of incorporation of both Kerr nonlinearity and thermal nonlinearity for the case of pulsed optical trapping.

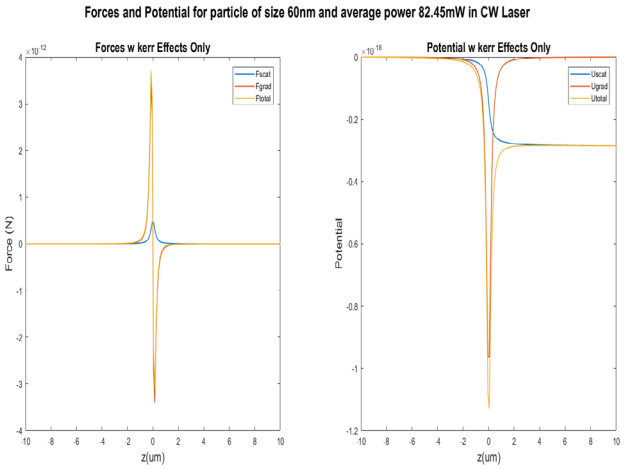


(a)



(b)

Figure 2 (a) Variation in intensity and (b) beam spot radius due to thermal effects Subscript t and n stands for thermal and thermal nonlinear effects respectively.



(a) (b)

Figure 3 Forces and Potential for a 60 nm particle with Kerr effects only in CW excitation

Here $n_{p2} = 5.9 * 10^{-17}$ *and* $n_{m2} = -2.26 \times 10^{-17}$ are taken.

5. Discussion

For the case when only Kerr effects are included, the plots for the forces and potential are different only in the case of pulsed laser optical trapping. But for the CW case the Kerr

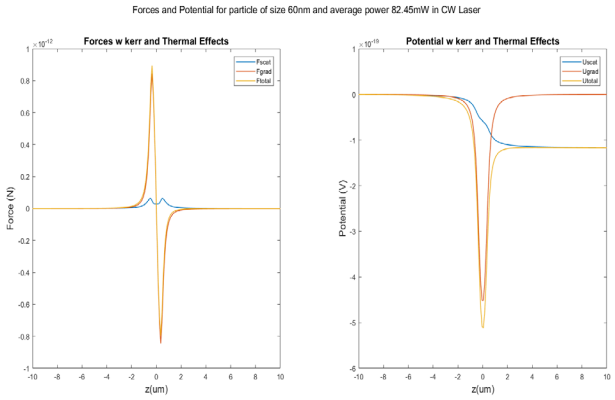
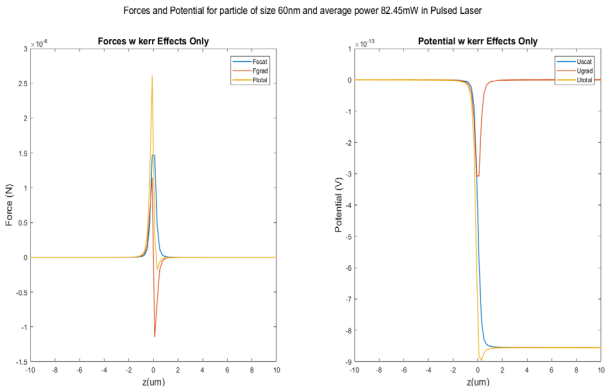


Figure 4 Axial force and potential including only Kerr and thermal nonlinearity for particles of size 60 nm in CW laser.



(a) (b)

Figure 5 Axial force and potential including only Kerr nonlinearity for particles of size 60 nm in pulsed laser.

nonlinearity is negligible and remains the same. When thermal nonlinearity in the medium is incorporated for the CW case the whole scenario changes significantly. Due to self-defocusing of the beam, the forces changes and the scattering force is undoubtedly different near the focal position of the tightly focused beam. The total force also decreases significantly in comparison to the Kerr-only case the escape potential is defined as difference between the potential minima and the potential value at maximum z-position (i.e. at infinity). The escape potential indicates the stability of the trapped bead. As can be seen from Table-1, the escape potential is almost halved.

For pulsed case, the forces, as well as well as the escape potential changes even with the inclusion of Kerr nonlinearity (figure 5). Now, when thermal nonlinearity is added on top of that, the forces change dramatically which are shown in figure (6) and corresponding escape potentials are listed in Table-1. The escape potential is reduced by a factor of 2.75. It clearly shows that the thermal effect along

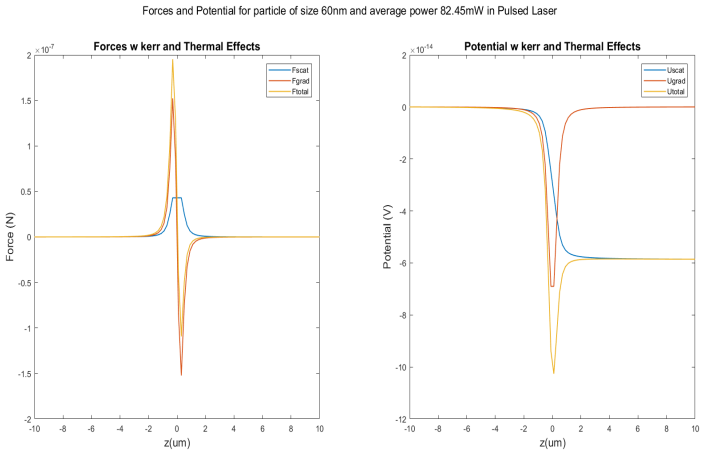


Figure 6 Axial force and potential including only Kerr nonlinearity for particles of size 60 nm in pulsed laser.

with Kerr nonlinearity is evident for pulsed case for stable optical trapping.

Overall the Rayleigh particles are more stably trapped in the case of pulsed laser than for CW. Also the thermally induced destabilization is also more prominent in the case of pulsed laser optical tweezers as for the case of 60nm particle.

Table 1 Comparison of escape potential for pulsed and CW laser including only Kerr and both Kerr and thermal effects.

Escape Potential in Volts for 60nm particle	
Pulsed Kerr only	1.43e-13
Pulsed Kerr and thermal	0.52e-13
(a) **rr only**	0 (b)
CW Kerr and thermal	0.4e-18

6. References

1. B. Agate, C. Brown, W. Sibbett, and K. Dholakia, "Femtosecond optical tweezers for in-situ control of two photon fluorescence," Opt. Express 12(13), 3011–3017 (2004).

2. R. Pobre and C. Saloma, "Radiation force exerted on nanometer size non-resonant Kerr particle by a tightly focused Gaussian beam," Opt. Commun. 267(2), 295–304 (2006).

3. H.J. Ravindra, A. John Kiran, K. Chandrasekharan, H.D. Shashikala and S.M. Dharmaprakash, "Third order nonlinear optical properties and optical limiting in donor/acceptor substituted 4'-methoxy chalcone derivatives" Applied Physics B 88, 105-110 (2007).

4. Rober W. Boyd, “Nonlinear Optics”, Elsevier Science, Academic Press 2008.

5. Qian Zhang, Xuemei Cheng, Bo He, Zhaoyu Ren, Ying Zhang, Haowei Chen and Jintao Bai, “Investigation on thermally-induced optical nonlinearity of alcohols”, Optics and Laser Technology 102, 140-146 (2018).

6. Sumit Singhal, Sirsendu Dinda and Debabrata Goswami, “Measurement of pure optical nonlinearity in carbon disulfide with high-repetition-rate femtosecond laser”, Applied optics 56, 644 – 648 (2017).

7. Eric W. Stryland and Mansoor Sheik-Bahae, “Z-Scan Measurement of Optical Nonlinearities”, Characterization Techniques and Tabulation for Organic Nonlinear Material M.G. Kuzyk and C.W. Dirk, Eds., Marcel Dekker Inc. 655-692 (1998).

8. A. Ashkin, J.M. Dziedzic, J.E. Bjorkholm and Steven Chu, “Observation of single-beam gradient force optical trap for dielectric particles”, Optics Letter 11, 288-290 (1986).

9. Anwar Usman, Wei-Yi Chiang and Hiroshi Masuhara, “Optical trapping of nanoparticles by ultrashort laser pulses”, Science Progress 96(1), 1- 18 (2013).

10. M. Jahja, “On nonlinear optical constants of polystyrene,” International Symposium on Modern Optics and Its Applications-2011 (2011).

11. J. M. Weber, CRC Handbook of Optical Material, Vol. III (CRC Press, 2003)