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# Understanding and Optimizing Laser-induced Thermoelectric Forces for Enhanced Trapping and Manipulation of Colloidal Particles

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## ABSTRACT

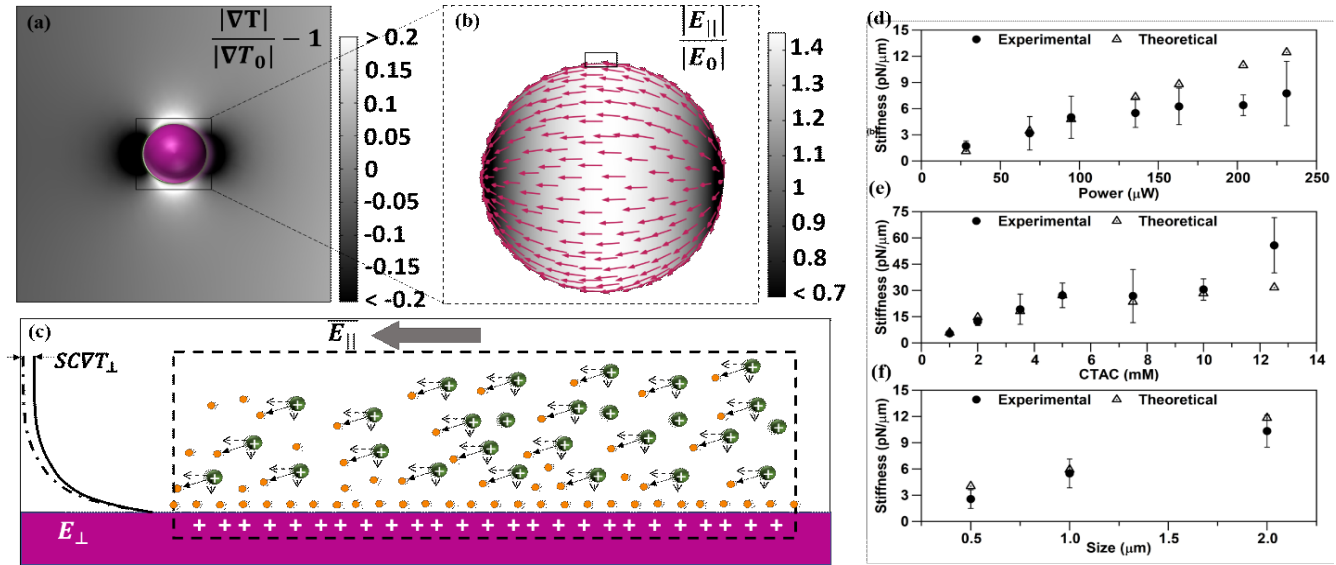
Several studies have been proposed to control particle trajectory in liquid solutions using optically induced thermal gradient. Upon introducing different solutes such as salts and surfactants along with microparticles in these solutions, an additional optically induced thermoelectric trapping force is generated due to the differential motion of ions in the solution under thermal field. As the complexity of the solution increases, it becomes increasingly difficult to understand particle response towards laser irradiance. More importantly, the existing models to study the thermoelectric behavior of the particle assumes a constant temperature gradient across the particles, which becomes obsolete in the micro-regime due to discontinuity of thermal conductivity at the particle-solution interface. For a better understanding of trapping and manipulation behavior of particles under light induced thermoelectric field, the temperature gradient distortion must be considered. In this work, full-scale finite-element solver model has been proposed to determine the temperature variation around a microparticle under laser heating. The resultant temperature distribution is utilized to numerically evaluate the thermoelectric field and the trapping potential of the laser induced opto-thermoelectric trap. To experimentally validate this methodology, polystyrene micro-particles are trapped opto-thermoelectric-ally in CTAC solution and compared the experimental trapping stiffness to theoretical estimates obtained from the model. It is observed that trapping stiffness saturates as surfactant concentration increases which can be optimized by choosing the lowest CTAC concentration at the onset of saturation. The model implemented here can be easily extended to arbitrarily shaped particles, particles with non-uniform surface morphology, different combinations of core-shell particles and electrolyte solutions, which can be implemented to study different phenomenon such as optical pulling, rotation and translation.

**Keywords:** Thermoelectric field, optical trapping, manipulation, colloids, dielectric particles

## INTRODUCTION

Opto-thermoelectric nanotweezers [1] were recently proposed to overcome the notable limitations of optical tweezers such as high laser power requirement and limited diversity in materials and the size of the nanoparticles. This technique uses the differential movement of cations and anions under laser-induced thermal gradient to create a thermoelectric field pointed towards the laser spot, enabling the trapping of positively charged particles at the laser center. The thermal gradient is created through the heating of a plasmonic substrate which has high photon-phonon conversion efficiency resulting in high temperatures for laser powers much lower than optical tweezers. The resultant thermoelectric field obtained because of the thermal gradient interacts with the charged particles in the colloidal solution resulting in an extremely stable trap at the laser focus. Although this technique has been excellently employed to trap and manipulate metallic, biological and dielectric particles [2], the underlying physics is not fully understood due to the complexity of multiple force fields such as optical, osmotic and thermoelectric forces interacting constructively. Also, the theory on thermoelectricity in colloidal solutions has been developed with a major assumption of a constant temperature gradient around the nanoparticles, which fails to explain several practical phenomena observed during the particles' interaction with lasers, thereby limiting their implementation in

applications [3]. Therefore, improvisation in thermoelectric field theory is essential for advancement of applications in nanorobotic studies. Herein, we prove that the introduction of the particle in an externally imposed constant thermal gradient distorts the thermal gradient. The distortion is decomposed into tangential and normal components on the surface of the colloidal particle. We show that the tangential component is responsible for the motion of the particle, while the normal component of the thermoelectric field after distortion dissipates as one approaches the surface of the particle. Since the particle distorts the thermal gradient, we can impose the same theory towards laser-induced spatially varying external thermal gradient. We numerically integrate the product of spatially-varying tangential component of the thermoelectric field and charge of the particle and extract the trapping stiffness of the dielectric particle under laser illumination and compare these values to the experimental trapping stiffness.



**Figure 1 Theory of thermoelectric trapping and experimental validation:** (a) Temperature gradient around the particle for a constant external temperature gradient. (b) Tangential component of the spatially varying thermoelectric field on the particle. (c) Schematic of ion distribution around the particle surface indicating the Stern layer and dipole alignment causing the tangential component of thermoelectric field. (d-f) Experimental and theoretical trapping stiffness of PS particles as a function of laser power, CTAC concentration and particle size. (Default values of power, concentration and size are 135  $\mu$ W, 1 mM and 1  $\mu$ m respectively).

## RESULTS

Herein, we proposed to bridge this theoretical deficit by numerically evaluating the thermoelectric forces and torques of nanoparticles by considering the spatial-variation of the field around the particle [4]. Using finite element analysis, we prove that the implementation of a constant temperature gradient across a particle would also result in a distortion of the temperature gradient around the particle due to discontinuity in the thermal conductivity at the interface (Fig. 1a). The resultant temperature gradient results in an ionic distribution which results in a thermoelectric field due to the electrolyte (in this case – a monovalent surfactant CTAC), formulated as

$$E_T = \frac{k_B T \nabla T}{e} \frac{\left[ \frac{Z_1 S_{T_1}}{N_{agg}} + S_{T_2} \right]}{1 - \frac{Z_1 S_{T_1} T}{N_{agg}}} = SC \nabla T \quad (1)$$

where  $E_T$  is the thermoelectric field,  $k_B$ ,  $e$ ,  $T$  and  $\nabla T$  are Boltzmann constant, electron charge, temperature and its gradient respectively.  $S_{T_1}$  and  $S_{T_2}$  are the Soret coefficients of surfactant micelle and chlorine ion, while  $N_{agg}$ ,  $Z_1$  and  $Z_2$  indicate the aggregation number, valency of CTAC micelle and valency of chlorine ion respectively.

Physical manifestation of charge separation is not observed in simulations due to low temperature gradients (10 K/ $\mu\text{m}$ ) utilized in this work. The parallel component of the thermoelectric field (Fig. 1b) interacts with the charge on the particle while the perpendicular component of the field is attenuated due to double layer screening (Fig. 1c), indicating that the response of the particle towards the temperature gradient is only due to the drag component of thermoelectric field.

Since the current theoretical method includes the changing temperature gradient around the particle, the same can be extended to laser heating of particles. The theoretical predictions have been experimentally substantiated for diverse phenomena such as opto-thermoelectric trapping, rotation and pulling of particles. For opto-thermoelectric trapping, thermoelectric forces have been estimated and trapping stiffness of PS particles is determined for varying laser powers, surfactant concentration and size which match well with the experimental values (Fig. 1d-f). The phenomenon of opto-thermoelectric pulling, obtained through self-heating of Silica particles, is well-explained from the balance between thermoelectric and optical forces. Also, the in-plane rotation of Janus particles under laser illumination was understood through explicit evaluation of thermoelectric and Stokes resistance torque.

## CONCLUSIONS

From the proposed theory, assisted with flexible modelling of the theory, we believe that this model has distinct advantages over mathematical models in terms of applicability to diverse experimental phenomenon. Moreover, shape-variations can easily be included in this theory by building the particle in any commercial 3D modelling software. Although the focus in this work has been on laser-induced thermoelectric fields, this model will also be valid for generic thermoelectric fields. With such flexibility in involving material, shape, size and composition of particles, and 3D spatial variation of electric fields around the particles to study particles' interactions, our computational model bridges down the gap considerably between the existing theory and experimental observations, thereby suitable for optimization studies in nanorobotics, nanophotonics and cellular biology.

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