INTRODUCTION
With the recent advancements in the field of fine particle engineering, it has become possible to manufacture particles of various homogeneous and composite materials in near uniform distribution sizes with diameter as small as several nanometers. Many are already widely available commercially and are considered to be directly useful as materials for bottom-up fabrication of devices such as photonic crystals, flat panel displays, and biomedical sensors. Accompanying the advancement in particle synthesis, there is an increasing interest in manipulating and ordering micron- and submicron-sized particles into patterns and structures.

The success of trapping micron-sized dielectric particles in a single beam gradient force optical trap by A. Askin et al has unfolded the possibility of manipulating particles using forces induced by optical fields [1]. Notably, S. Kawata et al have demonstrated that optical near-fields can also produce forces to move micron-sized particles [2]. Optical near-field has an advantage that it can be created on a localized area that is not exposed to the Rayleigh criterion [3]. Therefore, there are possibilities to create near-field enhanced patterns which are not subject to the diffraction limits of light. The work reported in this paper deals with manipulating and ordering submicron-sized particles dispersed in liquid using forces induced from optical near-fields. Researches involving direct manipulation of submicron-sized particles in liquid using optical near-fields have been reported by other groups; focused beam with a high numerical aperture objective lens and lithographically defined wave guides have been proposed for manipulating single or several particles [4][5]. We propose a method to order extensive particles into patterns using optical near-fields formed by multi-beam interference fringes exerted at a total internal reflection (TIR) interface. Our objective is to study the mass ordering of submicron-sized particles in a patterned optical near-field. In this paper we; 1) provide numerical estimations to derive the influence of the optical near-field pattern upon particles and 2) show experimental results of particle propulsion and ordering directed by optical near-field.

EXPERIMENTAL SETUP
The schematic of our setup is provided in Fig. 1. Samples of mono dispersed micron- and submicron-sized polystyrene particles or gold particles in water were each sealed between 100μm spaced borosilicate glass plates, and coupled to a BK7 prism using immersion oil with near matching refractive index. The glass plates were cleaned before being assembled into a target cell; they were sonicated with acetone and then with ethanol for 5 minutes respectively, dried in ambient air, and baked in an oxygen plasma oven for 10 minutes. The mono dispersed particles were sealed immediately after the cleaning of the glass plates. All experiments were done within 3 hours after the preparation of the target cell. For observations, the target cell was illuminated with incoherent white light and monitored through an objective lens. A color filter was placed after the objective lens to cut off scattering of the optical near-field by particles. Sequential images were recorded with a charged coupled device (CCD) camera. The incident angle at the glass-water TIR interface was set to an angle larger than the critical angle.

For experiments on particle propulsion, ø500nm polystyrene spheres (Polyscience Inc. Polybead® 07307) and ø250nm gold spheres (BBInternational Ltd. EM.GC250) were used. They were diluted with distilled water to 1.82×10^9 particles/ml and 3.60×10^8 particles/ml respectively. A linear polarized continuous wave laser with a TEM00 Gaussian beam intensity profile and wavelength of 1064nm (Coherent Inc.
Compass 1064) was converged into an elliptical spot of 100×240 \( \mu \)m at the TIR interface of the target cell without using the beam splitters. P-polarization was used.

For experiments on particle ordering, \( \phi 1 \mu \)m polystyrene spheres (Polyscience Inc. Polybead® 07310) diluted with distilled water to \( 2.28\times10^8 \) particles/ml was used. A linear polarized continuous wave laser with a TEM00 Gaussian beam and wavelength of 1064nm (Keopsys Inc. KPS-BT2-SLM-YFL-1064-30-PM-COL) was split into 4 beams, converged, and merged into an elliptical spot of 50×120 \( \mu \)m at the TIR interface of the target cell. The beams were arranged to create 5 \( \mu \)m pitched interference fringes. S-polarization was used. The fringes were visualized, as shown in Fig. 2, by integrating sequential images of light scattered from particles without white light illumination and color filter.

**NUMERICAL ESTIMATIONS**

To derive the influence of the optical near-field pattern upon the particles above the TIR interface of our setup, we have performed estimations based on dipole approximation. It is known that a dielectric or metallic particle which is smaller than the wavelength of light and is located in an optical field gradient can be considered as a dipole [6][7]. If the particles are suspended in liquid, the force \( F \) induced upon a particle by the optical field is the composition of the gradient force \( F_{\text{grad}} \), scattering force \( F_{\text{scat}} \), and absorbance force \( F_{\text{abs}} \) given by [8],

\[
F = F_{\text{grad}} + F_{\text{scat}} + F_{\text{abs}},
\]

\[
F_{\text{grad}} = \frac{1}{2} \epsilon |\nabla \langle E^2 \rangle|,
\]

\[
F_{\text{scat}} = n_n (S) k_m |\alpha|^2 / (6\pi),
\]

\[
F_{\text{abs}} = n_m (S) k_m \text{Im} |\alpha| / c,
\]

where \( \alpha \) is the polarizability of the particle, \( \langle E^2 \rangle \) is the time averaged electrical field strength of the optical field, \( n_n \) and \( k_m \) are the index of refraction and wave number of the surrounding medium, \( \langle S \rangle \) is the time averaged Poynting vector, and \( c \) is the speed of light in vacuum.

For spherical particles, polarizability \( \alpha \) is given by [9],

\[
\alpha = 3V \epsilon_0 \epsilon_m \left( \epsilon_p - \epsilon_m \right) / (\epsilon_p + 2\epsilon_m),
\]

where \( V \) is the volume of the particle, \( \epsilon_0, \epsilon_p, \) and \( \epsilon_m \) are the dielectric constants of the

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**FIGURE 1:** Schematic of the experimental setup; (a) overview and (b) detail of the target.

**FIGURE 2:** Light scattering of interference fringes by \( \phi 80 \)nm gold particles (BBInternational Ltd. EM.GC80). A total of 100 consecutive frames were integrated at an exposure rate of 30ms/frame. Scale: 10 \( \mu \)m.

**FIGURE 3:** Computer simulated beam intensity profile of the near-field pattern above the TIR interface. Intensities are stronger in light colored areas.
vacuum, particle, and surrounding medium respectively. We assume a s-polarized incident light at the TIR interface and that the inclusive profile of the beam intensity of the optical near-field pattern above the TIR interface, as shown in Fig. 3, to be \( I_o \exp\left(-2\frac{y}{r_0^2}\right) \),
\( I = I_o \exp\left(-2\frac{x}{r_0^2}\right) \),
\( I_o = 4n_s^2 \cos^2 \theta / (n^2 - n_s^2) \),
\( d_p = \lambda_0 / 2\pi n_s^2 \sin^2 \theta - n_s^2 \),
(3)

where \( r_0 \) and \( r_0 \) are the semi major and semi minor axis of the intensity profile at the TIR interface, \( d_p \) is the penetration depth of the near-field radiation above the TIR interface, \( n_s \) is the index of refraction of the substrate, \( \theta \) is the incident angle which is larger than the critical angle, \( I_o \) is the intensity of incident light, and \( \lambda_0 \) is the wavelength of the incident light in vacuum. It is clear that the gradient force, scattering force, and absorption force become significantly larger as the particle approaches the vicinity of the TIR interface. The maximal gradient force \( F_{\text{grad}}^{\text{max}} \), the maximal scattering force \( F_{\text{scat}}^{\text{max}} \), and the maximal absorbance force \( F_{\text{abs}}^{\text{max}} \), at the TIR interface, are given by,
\( F_{\text{grad}}^{\text{max}} = \frac{I_o}{4\epsilon_0} |\alpha| \left| \nabla \left[ \frac{\epsilon}{\epsilon_m} \right] \right| \),
\( F_{\text{scat}}^{\text{max}} = 4\pi^2 I_o n_s^2 |\alpha|^2 \),
\( F_{\text{abs}}^{\text{max}} = 2\pi^2 I_o n_s^2 \ln(\alpha) / (\lambda c) \),
\( \lambda = \lambda_0 / (n \sin \theta) \). (5)

For \( \varnothing 250\text{nm} \) gold particles in water; using \( r_0 = 21.5 \text{ (m)} \), \( \lambda_0 = 1064 \text{ (nm)} \), \( \epsilon_s = 55.5 + 5.62 i \) \[11\], and \( \epsilon_m = 1.325 \) \[12\], we find that,
\( F_{\text{grad}}^{\text{max}} \gg F_{\text{scat}}^{\text{max}} \), \( F_{\text{grad}}^{\text{max}} \gg F_{\text{abs}}^{\text{max}} \) . (6)

Because the influence of the gradient force is dominant, the force induced upon a particle can be approximated as,
\( F \approx F_{\text{grad}} = \frac{1}{2} |\alpha| (\nabla \cdot E^2) \). (7)

The potential energy \( U \) induced by the optical field is then given by,
\( U \approx \frac{1}{2} |\alpha| (E^2) = I_o |\alpha|^2 / (2n_s \epsilon_0 c) \). (8)

Particles are pushed towards light intensive areas of the optical near-field pattern. To retain the particles in a pattern, the potential energy must be larger compared to thermal energy \( k_B T \), where \( k_B \) is the Boltzmann constant and \( T \) is the absolute temperature \[13\]. At \( T = 298.15 \text{ (K)} \), \( \theta = 61^\circ \), and using incident light of 500mW, the potential energy induced by the optical field within the full width at half maximum of \( I \) follows,
\( 4.1 \leq U / (k_B T) \leq 8.4 \). (9)

This indicates that the gold particles are directed onto the optical near-field pattern at the TIR interface.

**EXPERIMENTAL RESULTS**

Fig. 4 shows sequential images of particles propelled by optical near-field forces. The color has been converted to pseudo color so that particles show as light intensive spots. Back ground noise has been subtracted. Particles moved alongside the major axis of the elliptically illuminated spot at the TIR interface. Taking the losses of irradiance at the lenses, mirrors, and beam splitters into account, the calculated power of incident light at the TIR interface was 2.5W and 0.74W for \( \varnothing 500\text{nm} \) polystyrene spheres and \( \varnothing 250\text{nm} \) gold spheres respectively.

**FIGURE 4:** Sequential images of particles being propelled by optical near-field forces at the vicinity of the glass-water TIR interface. (a) \( \varnothing 500\text{nm} \) polystyrene spheres. (b) \( \varnothing 250\text{nm} \) gold spheres. Scales: 10\( \mu \text{m} \).
The speed of propulsion was 2.7 μm/s and 15 μm/s respectively.

Fig. 5 shows Φ1 μm polystyrene spheres directed onto an optical near-field pattern with 5 μm pitched line and space. The calculated power of incident light at the TIR interface was 0.58W.

CONCLUSIONS
In this paper we have provided numerical estimations to derive the influence of the optical near-field pattern upon particles and have shown experimental results of particle propulsion and ordering directed by optical near-field. It has been indicated that submicron-sized gold spheres can be directed onto a pattern by optical near-fields. In addition to the propulsion of submicron-sized polystyrene or gold spheres, ordering of polystyrene spheres with size affinity to the incident light’s wavelength have also been demonstrated.

REFERENCES