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# Electrokinetic patterning of colloidal particles with optical landscapes†

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We demonstrate an opto-electrokinetic technique for non-invasive particle manipulation on the surface of a parallel-plate indium tin oxide (ITO) electrode that is biased with an alternating current (AC) signal and illuminated with near-infrared (1064 nm) optical landscapes. This technique can generate strong microfluidic vortices at higher AC frequencies (> 100 kHz) and dynamically and rapidly aggregate and pattern particle groups at low frequencies (< 100 kHz).

## Introduction

The ability to control the arrangement of particle groups, including biological cells, is important for the development of artificial architectures including colloidal crystals,<sup>1</sup> MEMS devices,<sup>2</sup> bioengineered tissues, and other products from similar assembly processes.<sup>3</sup> Non-contact particle-handling methods such as optical trapping,<sup>4–8</sup> electrophoresis,<sup>9–11</sup> and dielectrophoresis<sup>12–16</sup> have been widely accepted by the scientific community. Each of these techniques has their respective limitations. High resolution optical trapping methods suffer from limited throughput while electrokinetic techniques are limited to platform-specific applications due to permanently patterned electrode features. In addition, significant Brownian motion for extremely small particles and substantial drag on larger particles impose additional restrictions on transport speed.

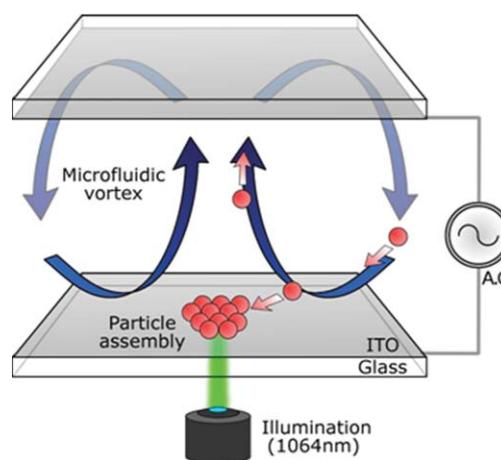
Although optically-controlled massive parallel transport of particles with dielectrophoretic forces has been achieved,<sup>15</sup> adaptation of the technology for large-scale close-packed assembly processes has not yet been demonstrated. Arbitrary electrokinetic colloidal aggregation at low AC frequencies have also been explored,<sup>17–20</sup> though creating specified patterns was not observed. Patterns have been generated with a DC electric field coupled with broad near-ultraviolet illumination geometries applied to a photosensitive platform,<sup>21</sup> however this patterning process can take hours to complete. Moreover the use of a rigid illumination mask inhibits dynamic control of these microstructures. Illumination has also been used to induce thermophoresis and convection to form two-dimensional crystalline structures.<sup>22</sup> However, this technique has yet to produce complex particle-assembly geometries.

Our new rapid electrokinetic patterning (REP) technique addresses these limitations combining a variety of electrokinetic

mechanisms including fluid manipulation, rapid particle assembly, and selective particle capture; each of these mechanisms can be induced on the platform mentioned here. These mechanisms can be operated independently or in concert to improve its overall efficiency. In addition, REP is optically induced offering dynamic control for its intended application. REP utilizes near-infrared optical landscapes coupled with a simple parallel plate ITO platform that is biased with an appropriate AC frequency. We demonstrate dynamic particle manipulation with polystyrene, latex, and silica particles (300 nm to 3.0  $\mu\text{m}$ ) at applied AC signals of 1 kHz–100 kHz and 0–20 volts peak-to-peak ( $V_{pp}$ ). The accumulated particles can be readily assembled, translated and patterned anywhere on the surface of the ITO substrate by dynamically changing the shape and intensity of the optical landscape.

## Experimental setup and theory

Fig. 1 illustrates the REP process. A liquid sample containing the suspended particles to be patterned is introduced between two transparent, conductive ITO-coated glass substrates separated with a 50  $\mu\text{m}$  spacer. An AC signal is applied between these substrates. The illumination source for REP is a Nd:YAG laser-based holographic illumination system (Bioryx<sup>®</sup> 200 from Arryx Inc., Chicago, USA). Holographic optical landscapes



**Fig. 1** Particles suspended in DI water are introduced between two parallel indium tin oxide (ITO) electrodes biased with an AC signal. Optical landscapes are applied at 1064 nm. The illumination heats the fluid sample and induces an electrohydrodynamic microvortex, bringing particles towards its center. Further, this illumination increases local current densities on the surface of the photosensitive ITO and creates a trapping region used to capture and pattern particles.

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are implemented by a spatial light modulator, which bestows computer-controllability of the generated light patterns making them easily repeatable and reconfigurable.<sup>23</sup> These strongly focused illumination patterns were used to drive the electrokinetic mechanisms behind REP. For these experiments, particles were suspended in DI water.

There are three distinct electrokinetic phenomena that were observed during particle cluster formation in REP. The first is electrohydrodynamic and is responsible for generating the recirculating microfluidic vortex. Localized fluid heating generates gradients in electrical permittivity and conductivity which, in the presence of an applied electric field, induces a body force on the fluid.<sup>14,24–26</sup> Although the electric field by itself generates heat, the optical illumination is the dominant source of non-uniform heat due to the highly-focused light patterns that vary radically in intensity over a few micrometers. The optical axis is parallel to that of the electric field. The fluid flow profile is toroidal, with the center of the recirculation located at the laser focal point (Fig. 1). This microfluidic vortex exhibits a sink-type behavior in the plane normal to the electric field. The velocity of the microvortex depends on the illumination intensity, the dielectric properties of the medium, and the voltage and frequency of the applied AC signal.<sup>27–29</sup> This microfluidic vortex has been recently characterized and is reported elsewhere.<sup>27</sup> At higher frequencies (>100 kHz) no particle assembly was observed and REP could be operated in a purely microfluidic mode, where vigorous fluidic mixing was achieved.

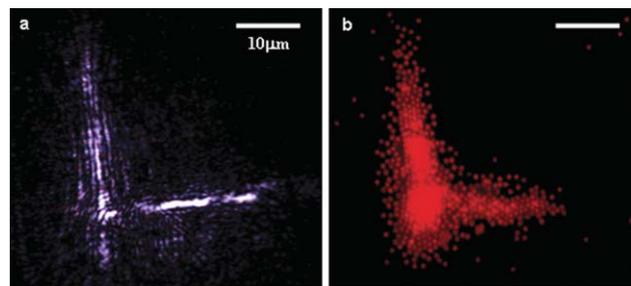
Low-frequency planar colloidal aggregation has been investigated previously<sup>17–21</sup> and their formation was attributed to induced electrohydrodynamic flows near electrodes that carry particles towards each other and into densely packed assemblies. This induced flow and particle aggregation occurs when the low-frequency electric field acts on the locally disturbed regions produced by the electrical double-layer of the particle. REP constructively uses the microfluidic vortex, which carries suspended particles towards the center of the optical illumination where they aggregate, leading to superlative particle collection rates.

These particle assemblies are patterned in and around the illuminated regions of the ITO. Aggregation shape is controlled by increasing the local current density on the electrode surface.<sup>19,21</sup> Previous experimenters have used a patterned oxide layer on top of the electrode<sup>19</sup> or selective illumination of a photoconductive surface<sup>15,21</sup> to create local variations in electric field density. Our optical landscapes increase the local current density of the electrode resulting in assembled colloidal patterns that resemble the illumination geometry. ITO is photosensitive in the infrared and in the ultraviolet<sup>30</sup> and optical patterning with this material has been investigated previously in the near-ultraviolet.<sup>21</sup> However, here we use its photosensitive nature in the near-infrared. To confirm the photoconductive nature of ITO, experiments were repeated with a 532 nm Nd:YAG laser. At this wavelength extremely weak microfluidic vortices were observed and illumination-dependent particle aggregation did not occur.

## Results and discussion

This compact, crystalline particle aggregation can be manipulated dynamically by modifying the illumination geometry,

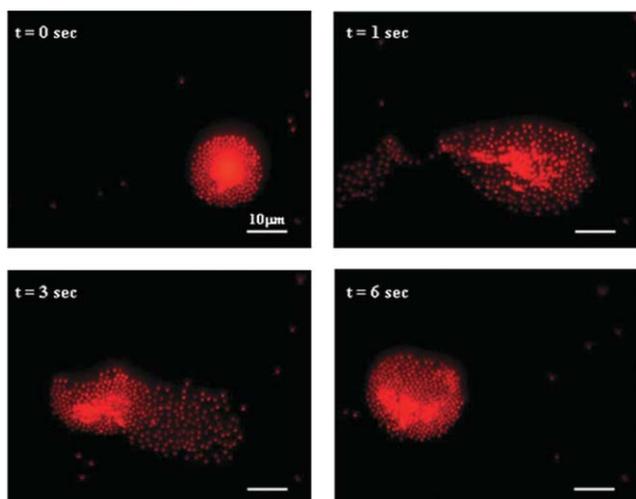
thus allowing translation, rotation, or shape alteration of the particle group. Fig. 2 shows an “L” shaped optical landscape (20 mW) and is used to capture and assemble polystyrene particles (690 nm) that resemble the illumination with an applied AC signal of 2.0 V<sub>pp</sub> and 1.6 kHz. It is important to note that this assembly phenomenon is not strictly electrokinetic or optical in nature. Various structures were assembled and manipulated using this technique, including accumulation of over 1200 polystyrene particles (Fig. S1†). The number of accumulated particles and the size of the trapping region increases with amplified illumination intensity and voltage.



**Fig. 2** (a) An optical landscape that resembles an “L” shape at 20 mW. (b) Polystyrene particles (690 nm) are captured resembling the illumination geometry with an applied AC signal of 2.0 V<sub>pp</sub> and 1.6 kHz.

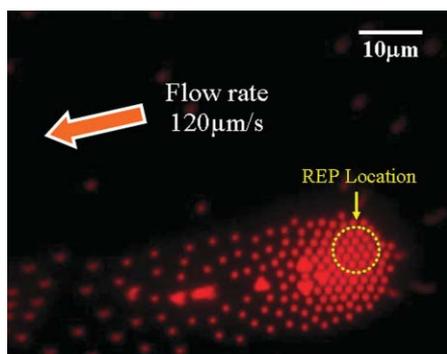
Further, particle accumulation is a function of applied AC frequency. From visual observation, an assembled group of polystyrene particles (1 µm) diminished as the frequency increased; at about 50 kHz all captured particles had dispersed. We attribute this behavior to low-frequency dispersion commonly referred to as the  $\alpha$ -dispersion in biophysics.<sup>14,31</sup> It is related to the polarization of the double layer around a particle whose relaxation time is dependent on surface charge. Utilization of this low-frequency behavior of colloids could lead to optimized particle assembly and selective sorting.

This technique can be used to pattern and translate hundreds of particles from one location to another by simply changing the location of the illumination (Fig. 3). Optical patterns were projected onto the surface of one of the parallel ITO electrodes using a 60× objective lens. In less than twenty seconds over 600 polystyrene particles (690 nm) accumulated in the vicinity of the optical pattern. Initially an illumination intensity of 20 mW was used with an applied AC signal of 2.0 V<sub>pp</sub> at 1.6 kHz (Fig. 3,  $t = 0$ ). The initial illumination is turned off (Fig. 3,  $t = 0$ ) and a new optical pattern is activated about 40 µm away with the same laser power. In less than six seconds the particle group disassembles, translates to this newly illuminated region and reassembles to form a new particle aggregation in a new location with a new shape (Fig. 3 and Video 1†). Instead of blinking the optical pattern, one can translate the particle aggregation by slowly translating the illumination relative to the electrode surface. Particle assembly experiments were repeated with polystyrene, latex, and silica particles ranging in size from 300 nm to 3 µm. REP is not restricted to these particle sizes or types; for example, previous low-frequency aggregation was shown with gold nanoparticles.<sup>17</sup>



**Fig. 3** Over 600 polystyrene particles (690 nm) move from one illumination location to another. The initial illumination is turned off ( $t = 0$ ) and another is activated about 40  $\mu\text{m}$  away. These particles disperse and then migrate to the newly illuminated pattern in under six seconds. The laser power is 20 mW and the AC signal is 2.0  $V_{\text{pp}}$  at 1.6 kHz.

REP can be used to enhance the performance of lab-on-a-chip sensors that are located at or near the surface of a microfabricated substrate. REP was used to attract particles within the microvortex and trap them on the illuminated surface. Next, pressure-driven bulk fluid motion of about  $120 \mu\text{m s}^{-1}$  was applied. The optically-driven vortex continuously concentrates latex particles (1  $\mu\text{m}$ ) on the ITO electrode surface (Fig. 4 and Video 2†). The illumination (40 mW) is a focused laser spot with the spot itself being diffraction limited in size ( $\sim 1 \mu\text{m}$ ). The dashed circle gives the approximate location of this spot. The applied AC signal was 10.5  $V_{\text{pp}}$  at 5.0 kHz. Laminar flow kept these concentrated particles near the wall, a benefit for subsequent downstream sensing processes.



**Fig. 4** Bulk fluid flow was applied to an active REP collection at 40 mW, 10.5  $V_{\text{pp}}$  and 5.0 kHz. Latex particles (1  $\mu\text{m}$ ) are continuously collected and concentrated in this near-wall region.

In addition to readily arranging and patterning particles, assemblies created with REP can be permanently adhered to the surface of the ITO electrode using an applied DC offset voltage of 2.5 V (Video 3†). This technique can therefore be used to assist in various micro- and nano-manufacturing applications.

## Conclusion

Here we describe a platform that offers a unique set of tools that have rarely been used simultaneously. The different techniques described here can be used in a variety of research applications. The fluidic vortex can be used for mixing or other microfluidic applications. Dynamic particle assembly can be used for the crystalline formation of colloids for artificial architecture research. Last, particle capture, translation, and concentration can be used towards the development of lab-on-a-chip technologies.

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