Light-Gated Manipulation of Micro/Nanoparticles in Electric Fields

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The rapid development of micro/nanomanipulation technologies has opened unprecedented opportunities for the sorting, assembly, and actuation of biological and inorganic entities for applications ranging from live-cell separation, drug screening, biosensing to micro/nanomachines and nanorobots. To this end, remarkable progress has been made in the development of efficient, precise, and versatile nanomanipulation techniques based on individual or combined chemical and physical fields. Among them, techniques that fuse light stimuli with electric (E) fields, have achieved impressive performance in the versatility, reconfigurability, and throughput in the manipulation of both biological and inorganic micro/nanoscale objects compared to those of many other manipulation techniques, by leveraging the strong optoelectric coupling effect of semiconductor materials. This work provides a review of various types of light-gated electric manipulation systems – the working principles, experimental setups, limitations, applications, and future perspectives.

1. Introduction

It is of immense interest to manipulate micro/nanoscale objects in a highly versatile, precise, efficient, and biocompatible fashion due to the myriad potential applications as well as the rich fundamental science. However, the conventional mechanical tweezers even when miniaturized, e.g., into the atomic force microscope, can not meet such a demand.[1] Over the past decades, substantial research efforts have been devoted to the development of powerful nanomanipulation techniques, which have successfully realized facile actuation of both biological and man-made micro/nanoentities, including alignment, transport, assembly, rolling, spinning, and collective operation. With the capability in controlling the mechanical behaviors of particles, the exploitation of nanomanipulation techniques has enabled unprecedented applications ranging from live-cell separation,[2] biosensing and drug delivery with motorized nanoparticles (or nanomotors),[3,4] to the assembly and actuation of arrays of micro/nanomachines that operate synchronously.[5]

According to the used working mechanism, nanomanipulation techniques can be categorized into three types: physical fields based manipulation, such as optical,[4,6,7] electric,[5,8] magnetic,[9] and acoustic-field powered tweezers;[10] chemical-reaction propelled motorization, such as self-electrophoresis, diffusiophoresis and bubble propulsion;[11] and the hybrid manipulation based on at least two stimuli applied to a particle.[12]

All of these approaches have proven to be effective with merits and limitations depending on particular applications. For instance, the chemical-reaction-enabled self-electrophoresis and diffusiophoresis can generate autonomous locomotion due to the concentration-gradient of ions produced around a particle.[13] Optical tweezers dynamically trap and manipulate particles with high versatility and accuracy. They have been widely used to study fundamental biological problems, such as the unveiling of the folding transitions of single molecules.[14] Strong electrokinetic forces have demonstrated convenient and efficient separation and sorting of micro/nanoscale particles.[2,15] Magnetic and acoustic fields are compatible with complex suspension medium and remain effective when applied from a distance, which are desirable for in vivo drug delivery,[16] assembly of live cells in three dimensions,[17] and fundamental studies.[18]

Yet each of these manipulation approaches has its limitations. For instance, the weak nature of light forces requires high light power for optical manipulations. The strong electrokinetic forces are usually restricted to the regions defined by pre-patterned electrodes, which makes it difficult to generate reconfigurable manipulation. To address some of these challenges, techniques based on the combination of two or more
external stimuli have been keenly investigated, as they may inherit merits from their parent techniques and overcome the associated weaknesses.\cite{19,20,21} In particular, light-gated manipulations of micro/nanoparticles in electric fields have demonstrated impressive performance in the dynamic, individually controllable, and high-throughput manipulation of biological and inorganic micro/nanoparticles with photo-gated electric-induced forces.\cite{21,22,23} Also, various other hybrid techniques such as acoustic/chemical\cite{19} and light/chemical\cite{20} motorizations have been well summarized in the recent Review articles.\cite{19,24}

Herein, we present a focused review on the state-of-the-art techniques of light-gated manipulation in external E-fields. An overview of this work is shown in Figure 1. Four types of manipulation systems categorized according to their different working mechanisms are discussed. One of such systems is realized on photoresponsive substrates by generating light-defined localized electric fields to manipulate a variety of synthesized nanoparticles and biological cells via dielectrophoretic (DEP) forces or electroosmosis flows. Based on the used photoresponsive substrates, such a system can be further categorized into optoelectronic tweezers (OET) and photovoltaic optoelectronic tweezers (PVOET) and are introduced in Section 2 and 3, respectively. On the other side, the rapid electrokinetic patterning (REP) technique takes advantages of the light-induced temperature gradients and locally generated electrothermal (ET) vortices to assemble particles. The related progress is summarized in Section 4. Section 5 highlights another approach to realize light-reconfigurable electric manipulation of nanoparticles (LREMN) by using light to modulate the electrical properties of the particles themselves and thus altering the materials-E-field interactions in a given external E-field. The operational mechanisms, experimental setups, and potential applications of the four systems are discussed separately. Finally, we summarize this review with a discussion of challenges and opportunities of the aforementioned techniques toward future applications, including sorting, diagnosis, transportation, modification, and assembly of micro/nanoparticles.

2. Optoelectronic Tweezers

OETs can trap and manipulate micro/nanoparticles via locally generated DEP forces or electro-osmotic flows in an AC E-field by patterning light on semiconductor amorphous Si (a:Si) substrates. Due to these strong electrokinetic forces, the optical energy required by OET can be a few orders of magnitude lower compared with that of the conventional optical tweezers. Furthermore, the reconfigurable virtual electrodes directed by patterned light on a:Si bestow the OET with highly versatile, dynamic, and individually controllable manipulation capabilities. In this section, we will introduce the working principles of OET, diverse OET setups, and noteworthy applications.

2.1. Basic Working Principles of DEP Used in OET Manipulation

It is known that when a micro/nanoparticle is placed in a nonuniform E-field, it can experience DEP force due to the interaction between the electrically polarized particle and the E-field. The DEP force can be given by \cite{25}

\[
F_{\text{DEP}} = (p \cdot \nabla)E = \nabla_{\text{particle}} \varepsilon_{m} Re(K)(E \cdot \nabla E)
\]

\[
= \frac{1}{2} \nabla_{\text{particle}} \varepsilon_{m} Re(K) \nabla E^{2}
\]

(1)

where \( p = V_{\text{particle}} \varepsilon_{m} Re(K) E \) is the effective induced dipole moment of the particle in the E-field, \( V_{\text{particle}} \) is the volume of the particle, \( \varepsilon_{m} \) is the permittivity of the suspension medium, and \( Re(K) \) is the real part of the Clausius–Mossotti factor \( K \), which can be expressed by \cite{25}

\[
K = \frac{\varepsilon_{p}^{*} - \varepsilon_{m}^{*}}{\varepsilon_{m}^{*} + L(\varepsilon_{p}^{*} - \varepsilon_{m}^{*})}
\]

(2)
where \( \varepsilon_p = \varepsilon - i \sigma_p / \omega \) and \( \varepsilon_m = \varepsilon_m - i \sigma_m / \omega \) are the complex permittivity of the particle and medium, respectively. The value of them depends on the frequency of the applied AC \( E \)-field \( \omega \), as well as the permittivity \( \varepsilon_p \) and \( \varepsilon_m \) and electric conductivity \( \sigma_p \) and \( \sigma_m \) of the nanoentity and the medium. The dimension and orientation of the particle in an \( E \)-field also play important roles in determining the Clausius–Mossotti factor as given by the depolarization factor, \( L \), in the denominator.

From Equation (1), it can be readily seen that the creation of a nonuniform \( E \)-field is required for the generation of a net DEP force to propel a particle. The motion direction can be toward (positive DEP force) or opposite from (negative DEP force) the highest \( E \)-field region, as shown by the sign of the real part of the Clausius–Mossotti factor, which depends on the dielectric properties of both the particle and medium, as well as the frequency of the AC \( E \)-field.

In a typical OET setup for the DEP manipulation as shown in Figure 1, one transparent indium tin oxide (ITO) glass is used as the top electrode. Another ITO glass functionalized with a photosensitive layer is used as the bottom electrode. During the operation, an AC signal is applied between the two electrodes. As the electric conductivity of the bottom electrode will locally increase at the location that light illuminates, a strong nonuniform \( E \)-field will be generated in the vicinity of the light illuminated area. As a result, particles close to this region will experience positive/ negative DEP forces and respond with mechanical motions. By dynamically altering the light pattern, particles can be instantly manipulated by DEP forces directed by light on demand.

### 2.2. Basic Working Principles of Electro-osmosis Used in OET Manipulation

Another method to manipulate particles with an \( E \)-field is called AC electro-osmosis, which is a phenomenon related to the motion of ions that are loosely attracted near a charged surface (e.g., the ITO substrate surface) under an AC \( E \)-field. When an AC \( E \)-field is applied to microelectrodes, charges can be induced on the surface of the electrodes and then ions with opposite charges in the aqueous solution will be attracted near the surface, forming the so-called electrical double layer (EDL). As the migration of ions takes a finite amount of time, the building up of the EDL in an AC \( E \)-field depends on the frequency of the AC signal. If the frequency of the AC \( E \)-field is too high, the EDL cannot form effectively. If the frequency is too low, the built-up EDL will effectively screen the \( E \)-field from the bulk liquid. The two extreme situations will both result in a negligible slip velocity of the ions flow. Thus, an optimal frequency exists for the electro-osmosis under an AC \( E \)-field, which can be estimated as\(^{[26]}\)

\[
\int_{opt} = \frac{1}{2 \pi} \frac{\sigma \lambda_d}{\epsilon D} \tag{3}
\]

where \( \sigma \) and \( \epsilon \) are the conductivity and permittivity of the liquid medium, respectively, \( \lambda_d \) is the thickness of the double-layer, and \( D \) is the feature dimension of the liquid layer. Consequently, the electro-osmosis flow can drag away particles close to the electrode surface. In the OET systems operating based on electro-osmosis, the AC \( E \)-field between the electrodes can be turned on/off by light due to the light-controlled electrical conductivity of the bottom electrode. When an AC signal with optimal frequency for electro-osmosis generation is applied, electro-osmotic flows can carry particles to designated areas by light patterning.

### 2.3. OETs with Different Structures

As mentioned earlier and shown in Figure 1, a typical setup of an OET system consists of two transparent ITO electrodes separated by a spacer, the bottom one of which has a photosensitive layer on the surface. The commonly used photosensitive layers include amorphous hydrated silicon (a-Si:H), heterojunctions, titanium oxide phthalocyanine (TiOPc), and so on. The electric conductivity of the photosensitive electrode drastically increases in the area with light illumination, which generates localized nonuniform
E-field that can be turned on or off with light. As a result, particles can be manipulated in the electric field. The propulsion range of DEP forces to electro-osmotic flows depending on the AC frequency. Ever since the first OET setup developed by Wu and co-workers in 2003,[27] OET setups have been redesigned for various purposes. For instance, the homogenous photoconductive layer was replaced with phototransistors[28–30] to accommodate the high conductive medium. They are also patterned[31–34] to generate lateral E-field or “always on” devices, which will be discussed later.

2.3.1. OETs with Featureless Photoconductive Surface

The key concept of OET devices is to instantly generate localized E-fields according to designed patterns and features under light illumination. The first OET designed by Wu and co-workers utilized a sandwich structure as shown in Figure 2A.[22] A conductive and transparent ITO glass is chosen as the substrate. Next, a photoconductive surface made of n+ doped a-Si:H, undoped a-Si:H, and silicon nitride is deposited on the ITO glass. An electric voltage, e.g., ≈100 kHz, 10 \( V_{pp} \), is applied between the photosensitive functionalized ITO electrode and a bare ITO electrode. A spacer that contains particle suspensions is used to maintain the distance between these two electrodes. When there is no light, the voltage drop in the liquids between the electrodes is negligible due to the much higher resistance in the undoped a-Si:H layer compared with that of the liquid suspension. However, when there is light, the electric conductivity of the photosensitive a-Si:H surface can be increased by several orders of magnitude (Figure 3A(iii)), resulting in a major voltage drop across the liquid layer. For instance, ≈2 orders of magnitude increase in photoconductivity is obtained with 635 nm diode laser illumination at 1 W cm\(^{-2}\) compared with that at 0.01 W cm\(^{-2}\).[35] Typically, a laser intensity in the range of 0.01–10 W cm\(^{-2}\) was used to generate sufficiently different electric conductivity compared with the surrounding areas.[35] As a result, an E-field gradient can be readily built near the dark/bright region on the substrate. Particles close to the dark/bright region on the photoconductive electrode experience DEP forces and move accordingly. By utilizing a similar setup and choosing an optimized AC electrical frequency, AC electro-osmosis flow can also be induced.[26] As shown in Figure 2B, vortices can be generated at the light-illuminated regions with an AC frequency between 1 and 10 kHz. These vortices can successfully trap and carry particles to the illuminated area on the substrate. We noticed that most OET manipulations are based on DEP. Only a few demonstrated light-gated manipulation of microparticles based on electro-osmosis as discussed earlier. This suggests that more potential works could be achieved based on the use of light-gated electro-osmosis in the future, particularly in the manipulation of flows, including mixing and pumping.

The fabrication of high-quality a-Si on ITO glass, however, can be challenging. It requires high-temperature plasma-enhanced chemical vapor deposition with thorough experimental optimization. An OET device based on a facile approach was developed by utilizing photosensitive polymers. Lee and co-workers replaced the a-Si:H layer with a polymer film made of a mixture of regioregular poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl C61-butyric acid methyl ester (PCBM) film.[36] This bulk-heterojunction polymer layer can be spin-coated onto the surface of ITO at room temperature. When illuminated with visible light, the photogenerated electron–hole pairs separate at the interface between the P3HT and PCBM and alter the E-field distribution. Although the fabrication of the photoco
ductive P3HT/PCBM polymer layer is simple and inexpensive, the conjugated molecules inside the polymer layer are sensitive to moisture and oxygen, which results in a short lifetime of the device (≈100 min). Capping the polymer with a LiF layer can extend the lifetime of the device to 350 min, but the generated DEP force is also reduced by 15%.[36] In 2010,
Liu and co-workers developed an OET device with a similar setup. In contrast, they use a 200 nm TiOPc layer as the photoactive material.\cite{37}

The entire process only involves spin-coating and a post-baking process at 130 °C for 30 min. The device can last for months under standard operating conditions. These materials may provide promising alternatives for disposable OET devices.

Here, it was noticed that the aforementioned OETs encounter problems when being applied to systems with insulating mediums, such as two-phase droplet-based microfluidic systems involving oils. Due to the much higher impedance of the oil layer compared with that of the photoconductive layer, the applied voltage always drops across the oil, regardless of the light illumination. Thus, Chiou and co-workers developed a floating electrode optoelectronic tweezers (FEOET) as shown in Figure 2C.\cite{38}

In this device, two aluminum electrodes separated by a 1 cm gap were patterned on the photoconductive a-Si:H layer. Polydimethylsiloxane (PDMS) with an open chamber filled with electrically insulating oil medium was assembled on the top of the substrate. A DC bias was applied between the two aluminum electrodes. Without light illumination, the E-field between the electrodes is constant. When the projected light shined onto the photoconductive layer, the conductivity was locally changed, which altered the uniformly distributed E-field. As a result, strong E-field gradients can be created at the edges of the light spot, which can penetrate the insulating oil medium and induce the DEP force to manipulate aqueous droplets inside it. Although this strategy is applied for the manipulation of relatively large liquid droplets in oil, the same principle could be useful when manipulating much smaller objects, such as nanowires and live cells, in insulating solutions.

2.3.2. Phototransistor-Based OETs

OET is a promising tool for the in vitro study of living cells. However, if typical cell culture medium (conductivity about 1.4 S m\(^{-1}\)) is used in the conventional OET setup, most voltage drop would occur on the amorphous silicon (photoconductivity <0.01–0.1 S m\(^{-1}\)) due to the much lower electrical conductivity, making it difficult to manipulate objects in the cell culture medium. To circumvent this issue, phototransistor-based optoelectronic tweezers (Ph-OET) were investigated.\cite{28–30} Hsu et al. first designed a Ph-OET device using arrays of square NPN phototransistors with insulators in between, as shown in Figure 3A(i).\cite{28} The photoconductivity of the phototransistor and the a-Si:H layer as a function of light intensity is shown in Figure 3A(ii). The conductivity of the cell growth medium is greater than that of phototransistor in the dark state but lower than that under light illumination. Thus, cells can be manipulated in high conductive growth medium by Ph-OET when a light is on. For instance, a maximum force of 14.5 pN can be applied to HeLa and Jurkat cells in phosphate-buffered saline (PBS) and Dulbecco’s modified eagle’s medium (DMEM) with a light intensity around 1 W cm\(^{-2}\).\cite{28} Accordingly, cells can be transported, trapped, and separated by designed optical patterns. Chiou and co-workers further improved the design of Ph-OET to realize the self-locking of cells and a much larger field-of-view.\cite{30} The schematic of the setup is shown in Figure 3B(i). Here, an Al\(_2\)O\(_3\) dielectric layer was deposited on the arrays of ring-shaped phototransistors and played a vital role. An appropriate AC frequency was chosen to ensure a large voltage drop in the liquid layer above the dielectric surface without light illumination and negative DEP forces exerted on cells. Without light illumination,
the phototransistor was in the off state and the E-field was weaker in the center of the electrodes compared with other areas. Thus, negative DEP forces can trap the cells onto the phototransistors. When illuminated by light, phototransistors can be activated with a large E-field applied to the top, which repels the trapped cells back into the medium. Simulated E-field distribution and direction of DEP forces around a phototransistor with and without light illumination are shown in Figure 3B(ii). As the trapping of cells does not rely on light, the field-of-view in the system is no longer limited by the optical aperture. Rather, the manipulation area depends on the size of the wafer.

2.3.3. OETs with Patterned Photoconductive Layer

The earliest version of the OET system relies on vertical E-fields generated from and enclosed between two parallel in-plane electrode slides. As such, anisotropic micro/nanoparticles, such as nanowires, usually align in the vertical direction in an OET system. The spatially enclosed system also limits the integration of OET with other functionalized entities. To address these issues, a system incorporating lateral optoelectronic tweezers (LOET) with interdigitated electrodes patterned on a single ITO substrate was designed by Wu and co-workers. As shown in Figure 4A, the neighboring photosensitive electrodes are activated only when illuminated with light. The simulation of E-field lines between two activated microelectrodes, shown in Figure 4A(ii) indicates the generated E-field is primarily in the lateral direction. Therefore, anisotropic particles such as silicon or silver nanowires can be aligned parallel to the substrate. Arrays of nanowires can be patterned on the chip. With a similar setup, Wereley and co-workers realized an open optoelectrowetting device that can be utilized to manipulate liquid droplets.

In contrast, to trap or manipulate micro/nanoparticles by the conventional OET, the light illumination needs to be “on” all the time, which may damage live cells. Light-induced heating effects may also accelerate the evaporation of liquid suspension, leading to unexpected complications in the system. Recently, Wheeler and co-workers developed patterned optoelectronic tweezers (p-OET) to alleviate these issues. As shown in Figure 4B(i), the photoconductive layer deposited on the ITO substrate was patterned by design. Due to the high conductivity of ITO, an E-field gradient near the exposed ITO film on the substrate is always present, regardless of the light being on or off. As a result, particles close to the ITO/a-Si:H boundaries always experience strong DEP force. The simulation of electric-field intensity distribution on the patterned electrode is shown in Figure 4B(ii). Meanwhile, light illumination on the a-Si:H can select and manipulate particles along designed paths. Therefore, once particles have been directed into the patterned trap by dynamic light patterning, particles can be continuously trapped even when the light is turned off. Figure 4B(iii–vi) shows the process of manipulation and trapping of two cells by p-OET. This design may pave the way for making long-term cell-trapping devices for fundamental studies in single-cell biology.

2.4. Applications of OETs

Since the report of the first OET device, the OET technique has demonstrated its versatility in the trapping and manipulation of both biological and inorganic particles at all scales, including biomolecules, nanowires, microspheres, macroscopic capacitors, and water droplets. Benefiting from the low threshold requirement for optical energy and the flexibility of the reconfigurable virtual microelectrodes, OET devices are particularly useful for in vitro cellular studies. They can benignly assemble live cells and massively manipulate cells in parallel, sort, distinguish, and electroporate cells.

In another aspect, early research on electrostatics indicates that a droplet-on-substrate system reaches the lowest energy when the droplet–substrate capacitance or droplet–substrate interface is maximized. As such, electric fields can be utilized

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**Figure 4.** OET with patterned photoconductive layer. A) The lateral OET devices with interdigitated electrode arrays. (i) The schematic experimental setup. (ii) Calculated electric field profile (streamlines) between two adjacent electrodes in the lateral OET. Reproduced with permission. Copyright 2007, IEEE. B) Patterned OET (p-OET) for particle/cell trapping. (i) The illustration of an OET trap by patterning the a-Si:H layer on ITO glass and (ii) the simulated electric potential distribution for a device at 7 Vpp (25 kHz). (iii–vi) Bright-field microscope images showing the manipulation and trapping of a 15 μm diameter polystyrene bead by a p-OET device. Reproduced with permission. Copyright 2018, Wiley-VCH Verlag GmbH & Co.
to alter the wettability of a substrate, the so-called electrowetting effect. It can be readily known that OET setups can be utilized to generate and dynamically pattern optoelectrowetting.

Wereley and co-workers took advantage of the optoelectronic wetting phenomenon and further demonstrated the transport and merging of droplets on a hydrophobic substrate. Shah et al. integrated the OET device with an electrowetting-on-dielectric (EWOD) chip and successfully extracted live HeLa cells from a big droplet into a new tiny droplet. These works have been well summarized in Wereley et al. and Park and Hwang’s reports. In this Review, we focus on some of the most recent applications on micro/nanoscale particles realized by OETs as follows.

Recently, Liu and co-workers used the OET device to measure the specific membrane capacitance (SMC) of benign human urothelial cells (SV-HUV-1) and malignant human bladder cancer cells (RT4). As established, cells will experience positive or negative DEP forces depending on whether the applied frequency is above or below the crossover frequency. The crossover frequency depends on the dielectric properties and thus the capacitance of the cell membrane. By applying AC signals with sweeping frequency, one can obtain the crossover frequency of particles using OET devices. Accordingly, the SMC of cells can be related to their sizes and their crossover frequencies. Figure 5A(i) shows the time sequence motions of a cell under AC fields with sweeping frequencies, indicating its crossover frequency is around 20 kHz. More motion trajectories of 13 cells are shown in Figure 5A(ii). The calculated quantified SMC values of RT4 cells (54 counts) and SV-HUC-1 cells (48 counts) are shown in Figure 5A(iii), indicating the SMC values of RT4 cells are much higher than that of SV-HUC-1 cells. Wheeler and co-workers recently explored a new application of OET by combining the OET manipulation and freeze-drying technique. Microparticles such as polystyrene beads can be well patterned using OET under light illumination. However, the patterned microbeads disperse quickly when the light is off. Combined with the freeze-drying process, the group successfully fixed the beads pattern on the substrate. Furthermore, the topographical micropatterns (TMPs) can be easily replicated to other substrates, such as PDMS stamps. They also demonstrate the potential application of topographical micropatterning for making security/identification markers of authenticity by transferring the patterns to paper currencies, nylon ribbons and coins, as shown in Figure 5B. Moreover, to overcome the limitation of field-of-view of microscope objectives used in OET devices, researchers have integrated OET with lens-free holographic microscopy and demonstrated real-time manipulation of cells and microparticles on a substrate of an area of 240 mm². Setups of OET are also able to power microrobots in the shapes of cogwheel, box, and spaceship to “scoop up”, transport, and...
separate microtissues from a mixture. Overall, researchers are integrating the OET systems with other advanced techniques to enable new applications in biology research, reconfigurable micromachines, and heterogeneous nanodevices.

3. Photovoltaic OETs

As previously mentioned, the OET technique leverages the reconfigurability of optical patterning and the high throughput of electrokinetic manipulation via optically patterned virtual electrodes based on the semiconductor a:Si. The replacement of traditional prepatterned thin-film electrodes by virtual electrodes from light-pattern projection greatly enhances individual controllability of micro-/nano-objects manipulated by electrokinetics. Thus, researching highly photosensitive materials applicable for virtual electrodes is essential for further development of OET technology. In addition to the photoconductive substrates, other materials with bulk photovoltaic effect (PVE) emerged in the recent decade and have been successfully demonstrated in particle manipulations.

In contrast to previously mentioned OET devices, which all require an external voltage supply, the PVE-based manipulation system does not require an external E-field. Instead, internal charge separation can be induced by nonuniform light illumination on the PVE crystal, resulting in E-field generation on the crystal surface. Due to the nonuniform light-induced excitation of carriers inside the noncentrosymmetric crystal lattice, a charge separation along the polar axis of the crystal is generated even without any applied voltage, as shown in Figure 6A.

Among several materials with the bulk PVE, LiNbO$_3$ shows a remarkable ability of internal field generation reaching up to several kilovolts per mm, making it the major focus in this area. Additional iron doping with optimized concentration can further enhance the response in the visible light range. Depending on whether the polar axis ($c$-axis) is parallel ($x$- or $y$-cut, as shown in Figure 6A(ii)) or perpendicular ($z$-cut, as shown in Figure 6A(i)) to the surface of the crystal, E-fields with different configurations can be built, and various particles can be assembled and trapped into desired patterns as shown in Figure 6B. Taking advantage of the reconfigurable virtual electrodes in photovoltaic OET, Clark and co-workers realized physically rewritable circuitry by manipulating micro-solder beads. In this setup, LiNbO$_3$:Fe was used to provide the light-induced DEP force and thus trapped and manipulated solder beads to complete the circuits. As shown in Figure 6D, strings of solder beads can be formed in sequence to bridge the base electrodes and turn on the green, yellow, and red LED, correspondingly. The building up of a solder string takes around 5 mins and it can be easily destructed by pipette aspiration.

Despite the advantage of spontaneous E-field generation without external power supply, bulk-photovoltaic-material-based manipulation systems usually suffer from low temporal resolution originating from the relatively long time needed to build up the internal charge separation. Materials with more rapid response time are needed to achieve real-time particle manipulation based on the bulk PVE.

4. Photo-Induced Thermal Electrokinetics

As great endeavors were made to overcome the limitations associated with each type of noncontact manipulation/assembly/

![Figure 6. Photovoltaic OET. A) Schematic images for the generated electric field of (i) $y$-cut and (ii) $z$-cut LiNbO$_3$ crystals. In the case of a $y$-cut crystal, strong positive (dark) and negative (white) alternating electric fields along the surface provide high field gradients for DEP trapping; in a $z$-cut geometry, a field normal to the surface is induced only for the illuminated regions, the resulting electric field gradients are substantially smaller. Reproduced with permission. Copyright 2013, AIP Publishing LLC. B) Photographic images of a diffractive patterned made of 70 nm spherical Al particles by illuminating a LiNbO$_3$:Fe crystal surface ($X$-cut) with a sinusoidal light-intensity profile. Reproduced with permission. Copyright 2016, The Optical Society. C) Microscope image of a 2D pattern formed by pollen after illumination with a mosaic of squares (200 $\mu$m) on z-cut LiNbO$_3$: Fe crystal. Reproduced with permission. Copyright 2016, AIP Publishing LLC. D) Demonstration of a rewritable three-way switch by photovoltaic OET. One-by-one assembly of solder beads with z-cut LiNbO$_3$: Fe crystal was realized by patterned light illumination. The solder beads bridged the upper and bottom electrodes to illuminate the green, yellow, and red LED, correspondingly. A pipette was used to destroy the formed solder beads. Reproduced with permission. Copyright 2017, Springer Nature.](image-url)
A promising example of such combinatorial technique (i.e., ones based on individually optical, electrokinetic, or thermal effect), researchers started to explore the synergistic effects of combining light, heat, and E-field into one platform.\cite{68} A promising example of such combinatorial technique, REP, will be discussed as follows.

In an REP system, the primary operating mechanism is an optically initiated ET vortex.\cite{69} When a near-infrared laser light is focused on an ITO electrode surface, local temperature increases on the electrode as well as in the nearby fluid. The generated temperature gradient leads to a change in local permittivity and conductivity, which creates spatial gradients of electric permittivity and conductivity in the fluid. When those gradients undergo an applied AC E-field, the aforementioned-ET flow is generated. Ramos and co-workers\cite{69} determined the equation for a time-averaged body force produced from ET flow as

$$
\langle f_i \rangle = \frac{1}{2} \text{Re}\left[ \left( \sigma \nabla E - \varepsilon \nabla \sigma \right) \cdot \frac{E}{\sigma + i \omega} \right] E^* - \frac{1}{2} |E|^2 \nabla
$$

(4)

where $E$ is the applied E-field, its complex conjugate is $E^*$, $\varepsilon$ and $\sigma$ are electrical permittivity and conductivity of the medium, respectively, and $\omega$ is the angular frequency of the applied AC E-field. As the ET flow acts on a particle through hydrodynamic drag forces, pushing it closer to the electrode surface, the electrode–particle interaction within the focused laser spot traps the particle. This trapping is a result of multiple forces balancing out each other. These forces were assessed by Wereley and co-workers in the body force diagram referenced in Figure 7A.\cite{70}

They include: a) a repulsive force between dipole and dipole ($F_{\text{particle-particle}}$), b) an attractive force between particle and electrode ($F_{\text{particle-electrode}}$), c) an electrohydrodynamic (EHD) attractive force between particles ($F_{\text{Lateral}_{\text{EHD}}}$, $F_{\text{Vertical}_{\text{EHD}}}$), and lastly d) an ET flow induced drag force ($F_{\text{Lateral}_{\text{ET}}}$, $F_{\text{Vertical}_{\text{ET}}}$). The attractive EHD force between particles stems from local flow generated around polarized particles due to the interaction between the $E$-field and EDL on the electrode and particles.\cite{70}

The same group also developed the first device based on REP as shown in Figure 7B.\cite{68} Particles suspended in deionized (DI) water are introduced between a pair of conductive and transparent ITO-coated glass slides, which are separated with a 50 $\mu$m spacer and biased with a low frequency AC signal ($< 100$ kHz). A 1064 nm laser source is integrated with a spatial light modulator (SLM) and used to generate computer-designed optical landscapes. When the setup is operational, the laser beam generates a temperature gradient in the fluid, which produces a microfluidic vortex by local-heating-induced EHD flows. The vortex ushers suspended particles toward the center and closer to the AC-biased electrode surface, which traps the particles through electrostatic interaction.\cite{68}

The impact of selected AC frequency on the force balance in REP systems was investigated (referring to Figure 7A).\cite{71} If the frequency is too low, attractive forces dominate and the optical effect vanishes, whereas if the frequency is too high (at or above an experimentally critical frequency, $f_c$), particles cannot be stably accumulated and end up swept into the bulk. The unstable accumulation of particles at frequencies above $f_c$ was attributed to the much stronger ET vortex. This $f_c$ is a consequence of the AC frequency-dependent ET-vortex-based vertical particle-electrode-based forces. $f_c$ is linearly proportional to the $E$-field strength, and inversely proportional to particle diameter, making it a consequent parameter for designing particle-sorting applications.

**Figure 7.** REP system overview. A) A free body diagram indicating forces acting upon a suspended colloidal particle near an electrode surface with an applied laser beam, which include vertical forces arising from ET, EHD, and electrostatic (particle–electrode) origins, and lateral forces from ET, EHD, and electrostatic (particle–particle) origins. Reproduced with permission.\cite{70} Copyright 2012, Springer. B) A typical REP platform illustration, where the components include two ITO-coated glass slides separated by a spacer, a source of AC signal, and a source of 1064 nm laser illumination work in conjunction to generate a microfluidic vortex arising from temperature gradients in the fluid after laser beam application, which drives a group of colloidal particles (suspended in DI water) to assemble at an optically induced electrokinetic trap. C) A panel displaying experimental result of REP application for particle patterning, in (i) the optical landscape generated from laser light (at 20 mW) modulated by an SLM is shown, whereas in (ii) the assembly of 690 nm PS spheres on the REP trap (at 1.6 kHz and 2 $V_{pp}$) is displayed, both scale bars are 10 $\mu$m. Reproduced with permission.\cite{69} Copyright 2008, Royal Society of Chemistry. D) Typical geometry of REP trap, (i) a 1 $\mu$m circular optical landscape generated from a 1064 nm laser, and (ii) collection of 1 $\mu$m red-fluorescent PS spheres on the REP trap, both scale bars are 5 $\mu$m. Reproduced with permission.\cite{71} Copyright 2010, American Chemical Society.
To show the robustness of the REP technique, the Wereley and co-workers deployed the setup shown in Figure 7B to create an optical landscape of 20 mW intensity in the shape of the letter “L”, and successfully patterned 690 nm PS spheres using an AC signal of 1.6 kHz at 2 Vpp, as seen in Figure 7C. In addition, they reported fast translation of a cluster of PS spheres from one concentration spot to another spot 40 μm away in 6 s, just by moving the optical landscape. Moreover, they observed that adding a DC offset of 2.5 V can permanently attach the patterned particles to the ITO substrate, which is deemed impactful for printing-based applications. Additional research revealed that changing the materials of the electrodes or underlying substrate in REP systems allows the use of alternative wavelengths in addition to infrared. For example, a 532 nm laser can be used in a REP setup with a sputtered gold electrode, 300 nm laser for a REP with silver electrodes, and 600–1000 nm for a REP with silicon substrate.

In general, REP devices provide a complement for light-stimulated electric manipulation systems with a simpler platform (no need for photoconductive or photovoltaic layer). However, it is difficult for REP to realize single particle/cell operation. As shown in Figure 7D, a cluster of particles with a size several times larger than the size of the optical landscape is a typical result of REP trapping, which may be advantageous for particle/cell concentration applications.

5. Light-Gated Reconfigurable Electric Manipulation of Nanoparticles

To realize future micro/nanomachines and robots, a key breakthrough should be made so that an individual robot in a swarm and subcomponents within the body of a robot can be reconfigurably manipulated with a high degree of freedom to carry out meaningful tasks on the micro/nanoscale. Among electrokinetic phenomena, there are several types of manipulations that can compel the motions of particles in E-fields with controlled forces and torques, i.e., DEP, electro-alignment, and electro-rotation. All of these manipulations are governed by the interaction between the dipole moment of the particle and the E-field. One obvious approach to control the motion of particles is to reconfigure the E-field in either strength or distribution. For instance, in most OET-related techniques, light patterns projected on a photoconductive substrate locally change E-field distribution and realize particle manipulation and trapping by DEP forces in the light-stimulated areas. Interestingly, an alternative approach could be the usage of light as a stimulus to instantly change the electrical properties of particles themselves, which results in the modulated mechanical motions of the particles due to the altered materials-E-field interactions in the same external E-fields.

5.1. Working Principle of Optically Tunable Polarization

Recently, Fan and co-workers demonstrated an innovative mechanism that can reconfigurably manipulate semiconductor Si nanowires in AC E-fields with plain visible light stimuli. Zeng and co-workers also demonstrated that a similar working mechanism can be applied to perovskite nanorods of CsPb(Br/I)₃. The working principle is based on light modulation of electric conductivity of light-responsive nanowires in an electric field, which effectively changes the particle-E-field-interaction. To understand the working mechanism, the optical effects on the electrical polarization, electro-alignment, and electro-rotation of silicon nanowires was studied. One can consider a silicon nanowire to be aligned by an AC E-field $\mathbf{E} = \text{Re}[E_0 e^{i \omega t}]$. The nanowire receives an electric torque of $\tau_\text{ele} = -\frac{1}{2} E_0^2 \text{Re}(\alpha_\parallel - \alpha_\perp) \sin \theta \cos \phi$, where $\alpha_\parallel$, $\alpha_\perp$ are the electric polarizability of the nanowire along the longitudinal and transverse directions, respectively, and $\theta$ is the angle between the long direction of the nanowire and the E-field. An alignment rate (A) can be defined as $A = \frac{\pi}{2} \text{Re}(\alpha_\parallel - \alpha_\perp)$, where $\gamma$ is the drag coefficient defined by $\tau_\text{drag} = -\gamma \mathbf{v}$. For electro-rotation, a circularly polarized rotating AC E-field, expressed as $\mathbf{E}(t) = E_0 \text{Re}(\hat{\mathbf{E}}(\theta)e^{i \omega t})$, is applied. The rotational torque is calculated as $\tau_\text{rotation} = -\frac{1}{2} E_0^2 \text{Im}(\alpha_\parallel + \alpha_\perp) \hat{\mathbf{E}}$. Therefore, it can be known that the electro-alignment and electro-rotation are governed by the real-part and imaginary-part of electric polarizability of a nanowire, respectively.

To study the optical effect on the manipulation of photosensitive particles in an AC E-field, single-crystal intrinsic silicon nanowires were synthesized with metal-assisted chemical etching. A diode-pumped solid-state (DPSS) laser of 532 nm was used to illuminate Si. The schematic of the experimental setup is shown in Figure 8A. The light effect on both the electro-alignment and electro-rotation of silicon nanowires was studied, as shown in Figure 8. Dramatic changes in both alignment rate and rotation speed were observed and they depend on laser intensity from 0 to 255 mW cm⁻². Here, a positive value of alignment rate $A$ indicates that Re($\alpha_\parallel - \alpha_\perp$) > 0, where the electric torque drives a nanowire to align in the same direction with the E-field. A negative $A$, where Re($\alpha_\parallel - \alpha_\perp$) < 0, indicates the nanowire aligns in the perpendicular direction with the E-field, which was observed at around 750 kHz without laser illumination. For electro-rotation, a positive and negative value of rotation speed indicates that the rotation direction is the same and opposite direction of the E-field, respectively. Upon light exposure, both the alignment rate and electro-rotation speed can significantly change in a wide range of frequencies with various features. The laser beam can increase, decrease, and even reverse the sign of the electric polarization of a Si nanowire at suitable conditions of E-field frequency and laser intensity. Moreover, this effect can be found in nanowires as small as sub-100 nm in diameter (Figure 8D). The very rich phenomena from the optically tunable electrical polarization offer great potential in applications of reconfigurable manipulation of micro/nanomachines and will be discussed in the following section.

5.2. Applications of Optically Reconfigurable Manipulation

Reproducible switching of the alignment of Si nanowires controlled by light can be achieved at 750 kHz. Without a laser, a silicon nanowire aligned perpendicular to the E-field. Once the laser is turned on, the nanowire instantly switched to the parallel direction with the E-field. Such light-controlled switching between two directions is rapid, reversible, and highly repeatable.
As shown in Figure 9A, when the laser is periodically toggled on and off, the nanowire consistently switched for hundreds of cycles.

At an AC frequency of $\approx 100$ kHz, the alignment rate can be greatly enhanced by the laser exposure for multiple folds. Based on this light-stimulated enhancement of the electric torque, an optically controlled microscale stepper motor, mimicking the macroscopic counterparts, can be successfully obtained. A micro-stepper-motor rotated in-phase with an $E$-field and turned to specific angles on demand. The operation of the stepper motor was realized by an $E$-field made of a hybrid AC signal, which consisted of an AC signal at a relatively high frequency $f_1 = 100$ kHz that continuously rotated at a low frequency $f_2 = 1.5$ Hz. The high-frequency AC component generated an alignment torque that propelled a nanowire to align toward the $E$-field. Meanwhile, the $E$-field was rotating, so that the nanowire continuously rotated to follow the $E$-field. When the alignment torque was sufficiently high enough to overcome the drag torque, the nanowire rotated synchronously with the $E$-field, as shown in Figure 9B(i). Conversely, when the alignment torque was insufficient to overcome the drag torque, the nanowire could not follow the $E$-field, and only exhibited a periodic oscillation (Figure 9B(ii)). As a result, by tuning the alignment torque, one can readily switch the motor between the in-phase and out-of-phase operation modes. With a digital light-processing system, the microscale stepper motors can be controlled independently by light patterns. Two motors under the same electric field can be independently controlled by two light spots projected as shown in Figure 9C, which offers a potential approach for individual manipulation of arrays of motors in the future.

Different from the aforementioned demonstration based on the real part of electropolarization of nanowires, light also impacts electro-rotation of silicon nanowires, which is governed by the imaginary part of electropolarization. By finely tuning light intensity, electro-rotation of a nanowire can be precisely controlled to accelerate, decelerate, stop, and even reverse the direction. A commercial projector was used to project light patterns onto nanowires, and the light intensity was controlled by tuning the grayscale of the input image, as shown in Figure 9D. At 0.5 and 0.01 MHz, the rotation speed of a nanowire increased with light intensity. At 0.05 MHz, the rotation speed first decelerated and then accelerated in the opposite direction with the increase in light intensity.

Leveraging the unique optically tunable electric polarization of semiconductor Si nanowires, reconfiguration in electromechanical manipulation has been demonstrated for the first time. The effect could be further generalized to other photoconductive...
materials,[74] including both soft semiconductor polymers, various semiconductor composites, and even 2D materials, as long as there is a prominent change in electrical conductivity under proper lighting conditions. Modulating the electrical properties on particles themselves rather than changing the electric field offers a new paradigm in reconfigurable nanomanipulation, which can inspire the development of novel micro/nanorobotic devices with versatile mechanical motions and functionalities.

6. Discussion

Both electrokinetic and optical tweezers are powerful tools for wireless manipulation of micro/nanoscale objects. The advantage of electrokinetic manipulation arises from the strong electrokinetic force, low-cost setup, and high throughput. However, electrokinetic forces are largely restricted by prepatterned electrodes and are often confined to a small region. It is difficult to generate reversible and reconfigurable motions of objects with simple electric fields. Only a few works have achieved versatile and/or high-precision manipulation based on strategically designed electric fields,[75] combined with additional propulsion forces,[74] or computer-vision-assisted control.[77] Optical tweezers, in contrast, can dynamically manipulate micro/nanoparticles with high force and spatial resolution. This technique has become a standard tool for understanding basic biological interaction and biomachinery, and already fostered profound impact.[6,78,79] However, optical tweezers require
high laser intensity and high-numerical-aperture focusing lens. Standing apart from the simple electrokinetic and optical tweezers, the various optoelectronic nanomanipulation systems developed in the past two decades provide a desired solution to realize flexible, dynamic, and high-throughput manipulation of micro/nanoparticles at low cost via leveraging the integrations of reconfigurable light stimuli with the strong electrokinesis forces. The essential materials that enable the desired effects are semiconductors with high photoelectric conductivities.

We summarize the specifics of the different manipulation methods discussed in this Review in Table 1. The methods include OET, PVOET, REP, and LREMN. The performance capabilities of typical optical tweezers are also listed for comparison. It can be found that the optoelectronic techniques require substantially lowered optical power in addition to the merits of high versatility and throughput compared to that of the optical tweezers. Also listed in the table are the used active materials, structural and material design, and the demonstrated applications.

As we know, nothing interesting is ever completely one-sided. In the following, we will discuss the issues related to each individual type of optoelectronic manipulation method. The search for new photoconductive materials other than a-Si:H is highly desirable for OET devices. It is known that the exploited OET devices rely on the fabrication of a photoconductive layer. In particular, a-Si:H has been most widely used. However, the fabrication of a-Si:H is a laborious and expensive process. Although other photoconductive materials such as P3HT/PCBM polymer or TiOpc film have been developed to replace the a-Si:H, their relatively short lifetime presents additional problems. Moreover, limited by the photoconductivity of a-Si:H, the conductivity of suspension medium used in OET is relatively low. Replacing the a-Si:H layer with patterned phototransistors can address this problem. However, the fabrication of phototransistors that involves doping and photolithography remains a technical challenge for researchers. Thus, new photoconductive materials that can overcome these challenges without degradation in performance could broaden the applications of OET.

The highly interesting photovoltaic OET devices also encounter major obstacles as the PVEs rely on the use of single-crystal substrates (LiNbO3:Fe for instance). The building up of the space charge separation may not be instant depending on light intensity (a few seconds at light intensity <1 W cm−2)\(^{[59]}\). Therefore, it is necessary to balance the temporal resolution with the light intensity. The medium used in this system is usually oil to circumvent the screening effect of aqueous suspension to the E-field, which limits its biomedical applications. The manipulations achievable by the x- or y-cut crystal is limited due to the rigid space charge separation parallel to the surface. Z-cut substrates can offer more flexible manipulations, as it usually demands the particle to be surface charged. These are barriers preventing the large-scale application of photovoltaic OETs.

Table 1. Comparison of OET, REP, and conventional optical tweezers.

| Structure         | Active material          | Particle                          | Power/Power density          | Voltage/Frequency/Gap distance (Typical) | Ref. |
|-------------------|--------------------------|-----------------------------------|------------------------------|-----------------------------------------|------|
| OET (dielectrophoresis) | a-Si:H                   | 25 μm latex particles             | 0.1–10 W cm\(^{-2}\) (635 nm) | 10 V/100 kHz/100 μm                   | [36] |
| OET (electro-osmosis)   | a-Si:H                   | PS sphere, DNA molecule, Quantum dots | 25 nW/pixel (625 nm LED)     | 4 V\(_{pp}\)/1 kHz/100 μm             | [26] |
| OET P3HT/PCBM        | PS sphere                | 20 μm PS sphere                   | 7.8 W cm\(^{-2}\) (white light) | 24 V\(_{pp}\)/100 kHz/50 μm           | [37] |
| OET TiOpc            | HepG2 cells              |                                   | –                            | 7 V/20 kHz/–                          | [38] |
| Ph-OET a-Si:H        | HeLa and Jurkat Cells    |                                   | –                            | 20 V\(_{pp}\)/9 MHz/100 μm           | [28] |
| LOET a-Si:H          | droplets                 |                                   | 15 mW cm\(^{-2}\)          | 42 V\(_{pp}\)/100–800 Hz/250 μm      | [32] |
| LOET a-Si:H          | Si NW                    | 10 W cm\(^{-2}\) (650 nm diode laser) | 5 V\(_{pp}\)/50 kHz/–      | –                                       | [31] |
| p-OET a-Si:H         | PS sphere, MCF-7 cells   |                                   | –                            | 7–25 V\(_{pp}\)/25 kHz/150 μm        | [34] |
| PVOET z-cut LiNbO\(_3\): Fe | glassy carbon (2–12 μm) |                                   | 8.9 mW cm\(^{-2}\) (532 nm laser) | –                                       | [65] |
| PVOET x- or y-cut LiNbO\(_3\): Fe | 100 nm silicon carbide particles |                                   | 600 mW cm\(^{-2}\)       | –                                       | [58] |
| REP ITO only         | 300 nm–3 um silica, PS, Latex spheres |                                   | 20 mW (1064 nm)       | 0–20 V\(_{pp}\)/1–100 kHz/50 μm      | [68] |
| REP ITO & gold sputtered | 50 nm PS spheres        |                                   | 180 mW (1064 nm)       | 14 V\(_{pp}\)/10 kHz/50 μm           | [80] |
| REP ITO & gold or silver & SiO\(_2\) substrate | 1 um PS spheres |                                   | 18 mW (1064 nm, also 532 nm for gold only, 300 nm for silver only, 600–1000 nm for silica substrate) | 13.5 V\(_{pp}\)/100 kHz/50 μm | [72] |
| LREMN Standard microelectrodes | Si NW | 8 mW cm\(^{-2}\) (532 nm) (not the limit) | 5–20 V\(_{pp}\)/5 kHz–4 MHz/500 μm (lateral) | [23] |
| Optical Tweezer      | 10–20 μm glass sphere   | 100 mW (514.5 nm laser)           | –                            | [79] |
| Optical Tweezer      | E. coli                 | 80 mW (1.06 μm laser)             | –                            | [81] |
| Plasmon Optical Tweezer | Patterned 4.99 μm PS beads | 250 mW (250 W cm\(^{-2}\))       | –                            | [82] |
However, the advantages of photovoltaic OET lie in the simple setup without external circuit wiring for E-field input. It complements other OETs as a wireless method.

Compared with OET devices, the REP setup does not require a photoconductive or photovoltaic layer. Instead, the commercially available ITO glass is sufficient for the REP manipulation. However, the flow-based manipulation is also a double-edged sword, which renders it inclusive for various kinds of particles, but gives it a poor spatial resolution for single particle/cell operation. The optically gated polarization-induced reconfiguration system is another scenario. As the light stimuli are used to change the electrical properties of particles themselves, the manipulation of this system offers much higher degrees of freedom and manipulation precision for individual particles and swarms of particles. However, the working mechanism is limited to photoresponsive particles. It will be interesting to investigate various efficient and biocompatible photoresponsive particles and their assembly with interesting micro-/nano-objects, which can functionalize nonphotoresponsive objects with versatile reconfigurability.

Overall, we discussed the working principles, experimental setups, and applications of the recently developed optoelectronic manipulation systems. We compared the advantages and limitations. Both challenges and exciting opportunities are present in these methods. Compared with many other approaches, optoelectronic manipulation systems can offer truly versatile and low-cost manipulation for the assembly and propulsion of future micro/nanorobots.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

electric fields, nanomanipulations, nanomotors, nanorobotics, optically reconfigurable manipulation

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