Transmittance tuning by particle chain polarization in electrowetting-driven droplets

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A tiny droplet containing nano/microparticles commonly handled in digital microfluidic lab-on-a-chip is regarded as a micro-optical component with tunable transmittance at programmable positions for the application of micro-opto-fluidic-systems. Cross-scale electric manipulations of droplets on a millimeter scale as well as suspended particles on a micrometer scale are demonstrated by electrowetting-on-dielectric (EWOD) and particle chain polarization, respectively. By applying electric fields at proper frequency ranges, EWOD and polarization can be selectively achieved in designed and fabricated parallel plate devices. At low frequencies, the applied signal generates EWOD to pump suspension droplets. The evenly dispersed particles reflect and/or absorb the incident light to exhibit a reflective or dark droplet. When sufficiently high frequencies are used on to the nonsegmented parallel electrodes, a uniform electric field is established across the liquid to polarize the dispersed neutral particles. The induced dipole moments attract the particles each other to form particle chains and increase the transmittance of the suspension, demonstrating a transmissive or bright droplet. In addition, the reflectance of the droplet is measured at various frequencies with different amplitudes. © 2010 American Institute of Physics. [doi:10.1063/1.3516656]

I. INTRODUCTION

Optofluidics handles optics and fluidics on a single chip to deliver new functions and applications.¹⁻³ For the variety of the working fluids and the unique phenomena of microfluidics, e.g., laminar flow and smooth interface between immiscible fluids, fluids pumped in microchannels would provide optical functions, including optical waveguides,⁴⁻⁶ liquid lenses,⁷ and liquid grating.^{8,9} In addition to manipulating fluids or lights along continuous microchannels, discrete droplets employed in digital microfluidics have also been applied to optofluidics. Digital microfluidics,¹⁰ on the one hand, plays an important role in the fields of lab-on-a-chip (LOC), micrototal analysis systems (μ TAS), and other microelectromechanical systems (MEMS) applications because the droplets can be regarded as isolated microenvironments for chemical reactions¹¹ or sample carriers for biomedical detections.¹¹ On the other hand, when taking advantage of the optical properties of the droplets, the manipulated droplets become tunable optical microcomponents in micro-opto-fluidic-systems (MOFS). In all the applications, including chemical, biological, and optical ones, the ability to control individual droplets is essential. Among various droplet manipulating mechanisms,^{12,13} electrowetting-on-dielectric¹¹ (EWOD) is widely used because the device structure and fabrication are relatively simple. EWOD has been successfully demonstrated to create, move, cut, and merge droplets, which are fundamental for LOC and

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μ TAS applications.¹⁴ Moreover, by appropriately choosing the optical properties of the EWOD-driven droplets or liquids, electronic displays,^{15,16} liquid lens,^{17,18} prisms,¹⁹ and mirrors²⁰ have been demonstrated.

In this study, EWOD is the fundamental force to drive aqueous droplets traveling along patterned electrodes. Not only changing the position of the droplet, the transmittance can be altered by polarizing and arranging the particles suspended in the liquid. The presented results demonstrate cross-scale electric manipulations based on our study of digital microfluidic cell/particle concentrator.²¹ We previously reported cell/particle concentrated toward strong or weak electric field regions by dielectrophoresis (DEP)^{22,23} using nonuniform electric fields. Instead of using nonuniform electric fields, here we apply uniform electric fields to polarize and drive the particles. Although the regular polystyrene beads are investigated to show the tunable transmittance, other functionalized particles, e.g., bioconjugated beads, would hold special applications in the suspension droplets. Similar polarization mechanism was reported and adapted to displays.²⁴

II. PRINCIPLE

As described above, the droplets are driven electrically by EWOD in this study. EWOD is usually investigated by measuring the contact angle of a sessile drop at different applied voltages.²⁵ The applied voltage dropped across the dielectric layer changes the contact angle of a droplet placed on a dielectric-coated electrode. Similarly, EWOD occurs between parallel plates as shown in Fig. 1(a). The Lippmann–Young equation describes the droplet contact angle variation by EWOD,

$$\cos \theta_V = \cos \theta_0 + \frac{\epsilon_0 \epsilon}{2 \gamma_{LG} t} V^2, \quad (1)$$

where V is the voltage across the dielectric layer, θ_0 and θ_V are the contact angles before and after voltage application, respectively, ϵ_0 is the permittivity of vacuum, ϵ and t are the relative permittivity and the thickness of the dielectric layer, respectively, and γ_{LG} is the surface tension between the liquid and gas surrounding. As analyzed by the simplified equivalent circuit of the parallel plate device,^{21,26,27} in order to have efficient voltage drop across the dielectric layer, the frequency of the applied voltage should be limited below several kilohertz. The low frequency voltage essential to EWOD is demonstrated by V_{LF} in Fig. 1(a).

At higher frequencies [V_{HF} in Fig. 1(b)], the voltage drop in the dielectric layer decreases and the fraction of voltage drop across the liquid increases. As a result, an ac electric field is established in the suspension to actuate the particles. Electrophoresis and polarization are the two major methods to manipulate the dispersed charged and neutral particles, respectively. In our case, the electrophoresis is negligible because the particles are neutral and the frequency of the AC electric field is high. The applied ac electric field causes the charge redistribution at the interface between the particles and the medium where a discontinuity of electric conductivity and/or permittivity exists. The particles in an electric field would hence be polarized due to the uneven charge distribution. The induced dipole moment of the suspended spherical particle can be expressed as²²

$$p = 4\pi a^3 \epsilon_m \left(\frac{\tilde{\epsilon}_p - \tilde{\epsilon}_m}{\tilde{\epsilon}_p + 2\tilde{\epsilon}_m} \right) E, \quad (2)$$

where p is the dipole moment, a is the radius of the particle, ϵ_m is the permittivity of the medium, E is the amplitude of the electric field, $\tilde{\epsilon}_m$ and $\tilde{\epsilon}_p$ are the complex permittivity of the medium and the particle, respectively. The complex permittivity $\tilde{\epsilon}$ is given by

$$\tilde{\epsilon} = \epsilon - i \frac{\sigma}{\omega}, \quad (3)$$

where ϵ and σ are the permittivity and conductivity, respectively, of the corresponding materials, and ω equaling to $2\pi f$ is the angular frequency of the electric field. The f is the frequency of the

FIG. 1. Electrowetting and polarization occurring in a parallel plate device to drive a droplet and dispersed particles on different scales by applying electric signals with different frequencies. (a) By applying low frequency signals (V_{LF}), the droplet would be deformed by EWOD. The solid lines show the large contact angle (θ_0) with no voltage applied. The electric signal with low frequencies decreases the contact angle (dashed line, θ_V). (b) The particle chains form when the applied frequency is high (V_{HF}).

electric field. The term in the bracket is called the Clausius–Mossotti (CM) factor, which is frequency related. According to Eq. (2), the dipole moment is proportional to the amplitude of the electric field, the cube of the radius, and the CM factor.

For a suspension droplet, when sufficient particles are evenly dispersed, the incident lights would be highly reflected or absorbed to display the color(s) of the particles straightforwardly. The particle manipulation technique introduced here is to polarize the particles by applying a uniform AC field between two parallel electrodes as shown in Fig. 1(b). As described above, the polarizability of the particles depends on the frequency of the electric field and the electric properties of both the particle and the medium. The dipole moments of each polarized particles attract one another to form particle chains along the electric field lines by the dipole-dipole interaction [Fig. 1(b)]. When particle chains are formed, most of the particles behind the top particles are shadowed and invisible from the top view. The particle chains decrease the projected particle density and hence reduce the reflection or absorption of the incident light. As a result, the transmittance of the suspension is increased by the ac electric field application. In comparison with other light regulators or displays based electrophoresis-addressed suspensions,²⁸ the moving distances for each

particle are much less because the particle chains are formed by local attraction of neighboring particles. It is noteworthy that the conformations, including density and length, of the particle chains are related to many parameters, for examples, the intensity of the external electric field, concentration, polarizability and size of particle.

As described in Eq. (2), the particle polarization as well as the particle chain transformation is not only related to the electric properties of materials but also to the amplitude and frequency of the electric field. The attractive force F between two induced dipoles is expressed by Eq. (4), where the dipole moment p described by Eq. (2) is incorporated. The electric field strength is also replaced with the voltage drop (V) over the thickness of the spacer/liquid (d),

$$F = \frac{6p^2}{4\pi\epsilon_m r^4} = \frac{24\pi a^6 \epsilon_m \left(\frac{\tilde{\epsilon}_p - \tilde{\epsilon}_m}{\tilde{\epsilon}_p + 2\tilde{\epsilon}_m} \right)^2 E^2}{r^4} = \frac{24\pi a^6 \epsilon_m \left(\frac{\tilde{\epsilon}_p - \tilde{\epsilon}_m}{\tilde{\epsilon}_p + 2\tilde{\epsilon}_m} \right)^2 V^2}{r^4 d^2}. \quad (4)$$

The concentration of the particle solution determines the tints of the droplets and the mean distance of particles (r). The thickness (d) of the spacer/liquid relates to electric field strength and tints. While the relationship is built, this phenomenon could be further applied in digital microfluidic system or color electronic display by tuning these parameters for different color particles. According to the previous research and the equations described above, particle chain transformation is frequency-related and material-related. This technique also gives a great opportunity to drive different kinds of particles at different frequency ranges in various applications.

III. EXPERIMENT AND RESULT

The commercially available polystyrene suspension was used to eliminate the uncertainties of the CM factor due to different material combinations. In addition, dyed polystyrene particles can be purchased in different colors with precise sizes suspended in water. As shown in Fig. 1, we first tested particle chain formation in a droplet placed between parallel plates. Two pieces of indium-tin-oxide (ITO) coated glass plates were used to build a uniform electric field in the droplet to polarize the particles and form particle chains rather than concentrating the particles toward strong or weak electric field regions by DEP using nonuniform electric fields.^{21,22} A transparent dielectric layer was applied onto one of the plates to prevent electric short when using conductive media of the suspensions. More importantly, the dielectric layer is necessary for EWOD actuations.

The parallel plates were prepared by the following processes. The ITO-coated glass plates were rinsed by acetone and DI water to remove contaminants before the 120 °C dehydration bake. The 5- μm -thick dielectric layer SU-8 (negative photoresist, MicroChem) was spun on the bottom plate. Finally, the hydrophobic surface was prepared by spinning Teflon (AF 1600, DuPont) on both plates with a thickness of 50 nm. The thicknesses of the dielectric and hydrophobic layers are not critical to the polarization phenomena but would affect the amplitude of the required driving voltage. The suspension droplet (1.25 μl) containing the polystyrene particles (Polysciences) was sandwiched between the top and bottom plates. The stainless spacer or double-side tape determined the distance between plates and the height of the droplet was 100 μm . Figure 2 (enhanced) shows the top views of the device before and after the electric field application. The concentration of the tested 5 μm polystyrenes in the droplet was 1.4×10^8 particles/ml. At the beginning, the particles were evenly dispersed in the droplet, displaying a reflective droplet in Fig. 2(a). With an ac electric field, the particles were polarized and formed particle chains along the electric field lines to display a more transmissive droplet as shown in Fig. 2(b). The particles in the droplet were driven by an electric field with the strength of 280 kV/m at 200 kHz. Some small satellite droplets caused by electrostatic pressure²⁹ were found at the contour line of the droplet, which can be prevented by replacing the air environment with an immiscible liquid or a solid packaging. In Fig. 2(c), the dispersed particles observed under a microscope displayed a reflective or dark state. When applying an electric field, the projected density of particles decreased from the top view

FIG. 2. Transmittance change of a $1.25\ \mu\text{l}$ droplet sandwiched between parallel ITO electrodes. (a) No voltage applied. (b) Voltage applied; the electric field strength was $280\ \text{kV/m}$ at $200\ \text{kHz}$. (c) Evenly dispersed particles observed under a microscope with no voltage applied. (d) The projected density of the particle decreased when voltage was applied. (enhanced online). [URL: <http://dx.doi.org/10.1063/1.3516656.1>]

because of the formation of the particle chains as shown in Fig. 2(d). The droplet was changed from reflective to transmissive. The process of the particle chain formation can be clearly seen in supplementary video 1.

The transmittance variation was interpreted and presented by means of the reflectance. The reflectance of the sandwiched suspension droplet ($2\ \mu\text{l}$) was measured using a custom-made apparatus containing a $50\ \text{W}$ halogen light source. The incident light was first evenly diffused through an integrating sphere to create an almost uniform incident light to the parallel plate device. The light reflected from the device with a reflective background was then focused by a lens, into a pinhole, and through a waveguide to be collected and analyzed by a spectrometer connected to a computer. Figure 3(a) shows the measured reflectance from 400 to $750\ \text{nm}$ plotted against varied applied frequency at the same electric field strength. We applied $25\ \text{V}$ to the device with parallel plates separated by $100\ \mu\text{m}$ high spacers. At low frequencies, most of the voltage dropped across the dielectric layer to generate EWOD. Therefore, the electric field strength in the liquid droplet was not sufficient to polarize the particles and change the reflectance. As can be seen from the curves, the reflectance did not change at $1\ \text{kHz}$ but dramatically increased at several hundred kilohertz. The curve of $1\ \text{kHz}$ shows no difference from that of no voltage applied. For the frequencies of 100 – $600\ \text{kHz}$, the reflectance decreased as the frequency increased. It was mainly caused by the frequency-dependent dipole moment, CM factor, and particle chain formation. The contrast ratio of the reflectance is shown in Fig. 3(b). When applying the tunable transmittance or reflectance based on the particle chain polarization to optical devices requiring a higher contrast ratio, e.g., displays, a suspension with smaller particles at a higher concentration to increase the opacity at the reflective or dark state is necessary.

In addition to manipulating particles on the micrometer scale, we further tested EWOD to drive the suspension droplet at the millimeter scale. The pumping of a droplet with tunable transmittance was examined in a parallel plate device as shown in Fig. 4. When V_{LF} is applied on a driving electrode [shown the middle one in Fig. 4(a)], EWOD moves the droplet on the powered driving electrode as shown in Figs. 4(a)–4(c). V_{HF} would further polarize the suspended particles and change the transmittance of the droplet as shown in Fig. 4(d). The fabrication process of the device was similar to the previous one except that the ITO was patterned to form EWOD driving electrodes on the bottom plate. After cleaning, positive photoresist, FH-6400L (Fujifilm), was spun, exposed, and developed on the ITO layer of the bottom plate to be a $1.5\ \mu\text{m}$ thick ITO

FIG. 3. The reflectance and contrast ratio of the droplet at different driving frequencies. (a) The reflectance from 400 to 750 nm. (b) The contrast ratio calculated from (a).

etching mask layer. After hard bake at 120 °C, the uncovered and exposed ITO was etched when immersing the bottom plate into aqua regia with predetermined parameters. The photoresist was then removed with acetone and ALEG 310 (photoresist stripper, Baker) after the etching process. The bottom plate was subsequently coated with the dielectric and hydrophobic layers.

Figure 5 (enhanced) shows that a 0.2 μl droplet containing polarizable particles was manipulated by EWOD to travel on the driving electrodes (indicated by dashed lines) from one side to the other between a 100 μm high gap. As shown in Figs. 5(a)–5(c), 50 V and 1 kHz were applied to drive the droplet by EWOD. The droplet first traveled by EWOD was then polarized by a high frequency electric field (500 kV/m, 200 kHz) on the center driving electrode. By applying appropriate electric signals, the droplet and/or the embedded particles would be selectively driven. EWOD requires the voltage drop across the dielectric layer, while the particle polarization needs the electric field inside the liquid. Therefore, the frequency of the electric signal determines which phenomenon occurring on the device. At low frequencies, most of the applied voltage drops across

FIG. 4. Cross-scale suspension droplet manipulations at different driving frequencies in a parallel plate device. (a) No voltage applied. The particles are randomly dispersed in the droplets. [(b) and (c)] The droplet travels on the electrode when a voltage at a low frequency is applied to decrease the contact angle on the energized electrode. (d) The particles in the droplet are polarized to form particle chains when the driving frequency is high.

the dielectric layer to generate EWOD exerting on the droplet. While at high frequencies, most voltage drop establishes the electric field in the liquid to manipulate particles. Certainly, the proposed transmittance change would be potentially applied to LOC for analysis purposes when other bioparticles, i.e., cells, proteins, and DNA, of interest are investigated.

IV. CONCLUSION

An EWOD-driven droplet with tunable transmittance was achieved in a parallel plate device to demonstrate a potential optical component in future MOFS applications. Two essential electric manipulations, EWOD and particle chain polarization, were investigated for droplet and particle actuations on difference scales, respectively. The particle chains altered the transmittance of the suspension and exhibited reflective and transmissive droplets whose positions were addressable by EWOD. The polarization was formulated and investigated by reflectance measurements.

FIG. 5. Positioning and transmittance altering of a suspension droplet by EWOD and polarization, respectively. [(a)–(c)] Droplet traveling on the patterned electrodes with a 1 kHz applied signal. (d) Polarization of the particles in the droplet at 200 kHz (enhanced online). [URL: <http://dx.doi.org/10.1063/1.3516656.2>]

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