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Surface-conduction enhanced dielectrophoretic-like particle migration in electric-field driven fluid flow through a straight rectangular microchannel

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An electric field has been extensively used to manipulate fluids and particles via electrokinetic flow in microchannels and nanochannels for various lab-on-a-chip applications. Recent studies have demonstrated the action of a dielectrophoretic-like lift force on near-wall particles in an electrokinetic flow due to the particles' modifications of the field-line structure adjacent to a planar wall. This work presents a fundamental investigation of the lateral migration of dielectric particles in the electrokinetic flow of buffer solutions of varying molar concentrations through a straight rectangular microchannel. We find that the particle migration-induced electrokinetic centerline focusing is significantly enhanced with the decrease of the buffer concentration. This observed phenomenon may be attributed to the increased surface conduction effect in a lower-concentration buffer that yields a larger Dukhin number, *Du*. It seems qualitatively consistent with a recent theoretical study that predicts a greater wall-induced electrical lift with the increasing value of the Dukhin number for $Du \geq 1$. *Published by AIP Publishing.* https://doi.org/10.1063/1.4996191

I. INTRODUCTION

An electric field has been extensively used to manipulate fluids and various types of particles (from ionic species to macromolecules, viruses, beads, and cells) in microfluidic and nanofluidic devices for diverse chemical and biomedical applications.[1,2] It generates fluid electroosmosis and particle electrophoresis through the volumetric Coulomb force exerted on the net charge inside a spontaneously formed electric double layer (EDL) at the liquid-solid interface.[3,4] This often-called electrokinetic flow is essentially surface driven and has a nearly plug-like velocity profile in microchannels due to the typically thin EDL as compared to the characteristic channel dimension.[5,6] It thus experiences a much smaller flow resistance and causes a much smaller hydrodynamic dispersion than the traditional parabolic pressure-driven flow.[7,8] There have been numerous studies on particle electrophoresis in straight microchannels, which, however, have been focused primarily upon the wall effect on the axial particle translation.[9,10] Much less studies have been reported on the transverse particle migration in the electrokinetic flow.

Fluid electroosmosis and particle electrophoresis both remain unvaried along the flow direction in straight uniform microchannels. In the limit of thin EDLs (as compared to the particle size), many theoretical and numerical studies have been conducted to determine the electrophoretic velocity of spherical or cylindrical particles moving either next to a plane wall[11–13] or in a slit, cylindrical, or rectangular microchannel.[14–26] The predicted retardation of electrophoretic motion has been verified in two recent experiments.[27,28] However, when a particle moves in close proximity to a channel wall (i.e., with a very small particle-wall separation distance due to, for example, the gravity action), the presence of the wall is predicted to enhance the electrophoretic motion.[11,13,14,16,29,30] This prediction has been experimentally verified in two recent experiments on near-wall particle electrophoresis in both cylindrical[31] and rectangular[32] microchannels. The above described wall effects, either retardation or enhancement, on the electrophoretic motion become stronger at a larger EDL thickness,[33–35] which is also consistent with a recent experimental observation.[31]

Only until recently have a few studies considered the wall-induced lift effects on particle electrophoresis in microchannels. Yariv[36] conducted a theoretical analysis to demonstrate that a particle drifts away from a plane wall under the influence of a tangential electric field. This lateral migration, which is superimposed on the traditional electrophoretic motion parallel to the wall, is induced by an electrically originated dielectrophoretic-like lift force that is calculated by surface integration of the Maxwell stress tensor. Zhao and Bau[37] later investigated theoretically the forces on both polarizable and dielectric cylindrical particles adjacent to a planar insulating wall. They demonstrated that the wall-induced hydrodynamic force is negligible for dielectric particles as compared to the electrical lift. A similar electrical force was also studied by Young and Li[38] to counterbalance the gravity of a particle during its electrophoretic motion near a plane wall, such that the particle-wall separation distance can be theoretically determined. It was later considered by Kang *et al.*[39] via a

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numerically obtained empirical formula in order to match the experimentally obtained particle trajectories in the electrokinetic flow around an insulating hurdle. This dielectrophoresis-resembled (the translation of a particle in response to a non-uniform electric field that exists in the absence of the particle[40]) force[36,41,42] is different from and orders of magnitude stronger than the so-called electrokinetic or electro-viscous lift,[43,44] which arises from the non-uniform electric field of the induced streaming potential in the fluid between a moving charged particle and a nearby plane surface.[45,46]

The first experimental demonstration of the wall-induced electrical lift was reported by Liang *et al.*,[47] who observed the centerline focusing of polystyrene microparticles in electrokinetic flow through a straight rectangular microchannel.[48] The authors also used the formula proposed by Yariv[36] to simulate the particle migration under varying electric fields. The predicted particle trajectories agree reasonably well with the experimental observation.[47] The existence of the electrical lift was further confirmed by Yoda and colleague[49,50] through the use of evanescent wave-based particle-tracking velocimetry to study the near-wall particle distribution in the electrokinetic flow. This wall-induced electrical lift has been used to tune the gap distance of non-neutral particles from an insulated wall for a fundamental study of the wall effects on both electrophoresis in a straight rectangular microchannel[32] and particle dielectrophoresis in a diverging microchannel.[51] A similar idea was later demonstrated to separate particles by size in electrokinetic flow through a single-spiral microchannel via the particle-size dependent equilibrium position.[52] The measured particle-wall gap agrees well with the prediction of Yariv's formula for particles of different sizes. The wall-induced electrical lift has also been recently exploited to separate sheath-focused polystyrene particles[53] and oil droplets[54] by size in straight rectangular microchannels. Moreover, the same approach works for the continuous electrokinetic separation of polystyrene particles and yeast cells based on the difference in surface charges.[55] Later, Li *et al.*[56] demonstrated the use of the wall-induced electrical lift to achieve a sheath-free particle separation in a bifurcating microchannel. They also developed a theoretical model based on Yariv's formula[36] to understand and simulate the particle focusing and separation processes.

In a recent article, Yariv[57] revisited the particle-wall repulsion force under a uniform electric field[36] by considering the balance between bulk conduction and surface conduction in an electrolyte solution. He obtained a revised formula of the wall-induced electrical lift for remote particle-wall interactions, which predicts a non-monotonic dependence on the particle Dukhin number (the relative magnitude of the surface conduction of the particle to the bulk conduction of the suspending fluid[58]), Du. Specifically, the electrical lift first decreases rapidly ($0 \le Du \le 1$) and then gradually increases ($Du \ge 1$) with the Dukhin number. The objective of this work is to experimentally study the surface conduction effect on the wall-induced electrical lift force and in turn the particle migration in a straight rectangular microchannel. Using a similar approach to that in our earlier papers,[47,48] we investigate the electrokinetic focusing of particles of various sizes in the electrokinetic flow of buffer solutions of varying molecular concentrations. A substantially enhanced particle migration is observed when the buffer concentration is decreased or equivalently the particle Dukhin number is increased due to the increasing role of surface conduction.

II. EXPERIMENT

A straight rectangular microchannel was used in our experiment. It was fabricated with polydimethylsiloxane (PDMS) using the standard soft lithography technique. The channel was measured 65 μm wide, 25 μm deep, and 2 cm long. It has an expansion region at each end where an array of posts are designed for the purpose of blocking out any debris. Spherical polystyrene particles of 3 μm, 5 μm, and 10 μm diameters (Sigma Aldrich, USA) were re-suspended in phosphate buffer solutions of 7 molar concentrations (ranging from 0.01 to 0.1, 0.2, 0.5, 1, 5, and 10 mM) and as well in de-ionized (DI) water (viewed as a 0 mM buffer solution). The electric conductivities of 0.01, 0.1, and 1 mM buffer solutions were measured to be 5, 25, and 210 μS/cm, respectively. These values were used to obtain a linear trendline between the fluid conductivity and the molar concentration as shown in Fig. 1. The particle Dukhin number, Du, was employed to quantify the effect of surface conduction,[59] which is defined as follows:

$$Du = \frac{\sigma_s}{\sigma a}, \tag{1}$$

where σ_s is the surface conductance of particles, σ is the bulk electric conductivity of the suspending buffer, and a is the particle radius. The values of Du for 5 μm diameter dielectric particles in buffers of varying molar concentrations were calculated from Eq. (1) by the use of the suggested value, σ_s = 1 nS, for polystyrene particles.[59] As seen from Fig. 1, Du increases from nearly zero in 10 mM buffer to over 10 in 0.001 mM buffer for the constant σ_s value we used here, indicating an increasing influence of surface conduction when the buffer concentration is decreased.

Tween 20 (Fisher Scientific, Pittsburg, PA) was added into each particle suspension at a volume ratio of 0.5% to avoid particle adhesions and aggregations. The electrokinetic motion of particles was driven by DC electric fields generated by a high-voltage power supply (Glassman High Voltage, Inc., High

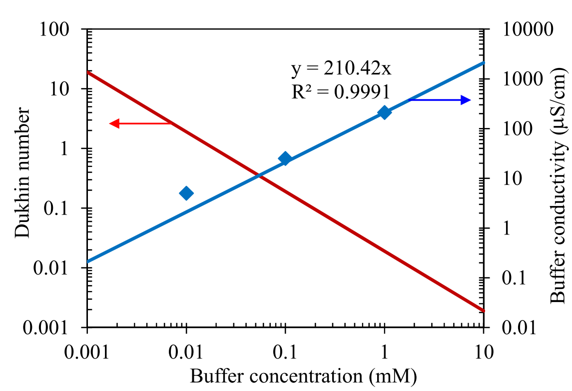

FIG. 1. Variation of the Dukhin number, Du in Eq. (1), for 5 μm diameter polystyrene particles and the electric conductivity with the molar concentration of buffer solutions. The symbols represent the measured buffer conductivities, from which a linear trendline (with the equation and the R-squared value displayed on the chart) was obtained in Microsoft Excel®.

Bridge). The pressure-driven particle movement was minimized by carefully balancing the liquid heights in the two end-reservoirs prior to every test. Particle motion was visualized at the channel inlet and outlet through an inverted microscope (Nikon Eclipse TE2000U, Nikon Instruments, Lewisville, TX) equipped with a CCD camera (Nikon DS-Qi1MC). Digital videos were recorded at a rate of around 15 frames/s with an exposure time of 1 ms. Superimposed images were obtained by stacking the frames in a 30 s video using the Nikon imaging software (NIS-Elements AR 3.22). The electrokinetic velocity of particles was determined by tracking the particle position with time in the main-body of the microchannel. The effects of Joule heating were estimated weak because the measured electric current shows an insignificant increase during the course of any experiment.[60]

III. RESULTS AND DISCUSSION

Figure 2 shows the experimentally obtained top-view images of 5 μm diameter particles in the electrokinetic flow of buffer solutions of varying molar concentrations under a 300 V/cm DC electric field. At the inlet of the microchannel, particles are randomly and uniformly distributed over the channel width. They are directed toward the channel centerline by the wall-induced electrical lift, leading to a tighter particle stream at the channel outlet. Such a sheathless electrokinetic particle focusing gets apparently enhanced when the molar concentration of the suspending buffer solution is decreased (or accordingly the Dukhin number is increased as seen from Fig. 1), especially within the range from 10 mM to 0.1 mM. This trend with the increase of the Dukhin number seems to be qualitatively similar to the prediction of Yariv's theoretical model[57] for $Du \geq 1$, which can be easily fulfilled for highly charged dielectric particles (e.g., when the zeta potential of particles becomes comparable with several thermal voltages[57,58]). In this regard, we will conduct in the future further experiments on dielectric particles that carry varying surface charges (e.g., coated with different functional groups) in order to understand their effects on the dielectrophoretic-like electrical lift.

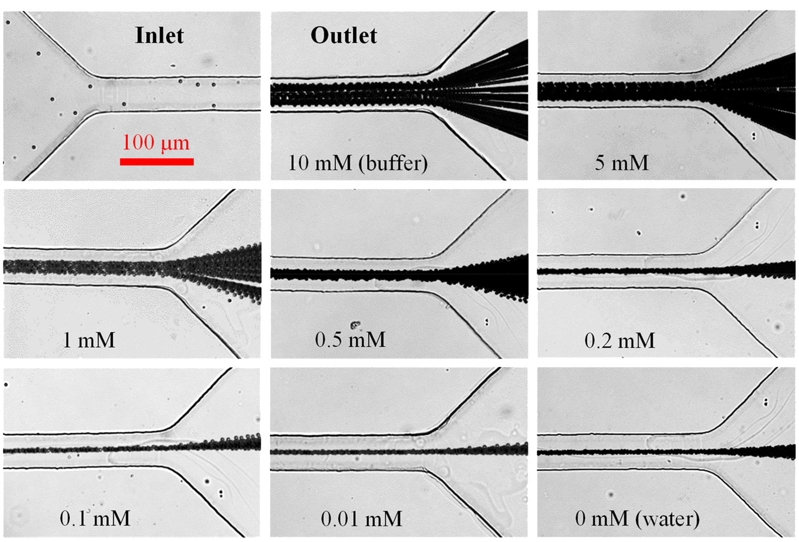

FIG. 2. Top-view images at the channel inlet (top left, snapshot) and outlet (all others, superimposed) illustrating the surface-conduction enhanced migration (and hence centerline focusing) of 5 μm diameter polystyrene particles in the electrokinetic flow of buffer solutions of varying molar concentrations (as labeled on the images). The average DC electric field across the channel length is 300 V/cm in all cases. Particles travel from left to right in all images.

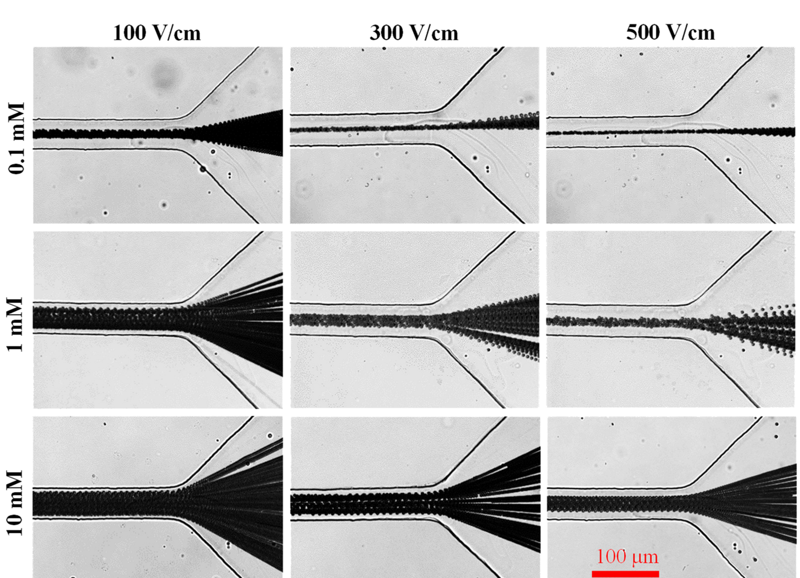

FIG. 3. Superimposed images at the channel outlet illustrating the migration-induced centerline focusing of 5 μm diameter polystyrene particles in the electrokinetic flow of buffer solutions of varying molar concentrations under different DC electric fields. Particles travel from left to right in all images.

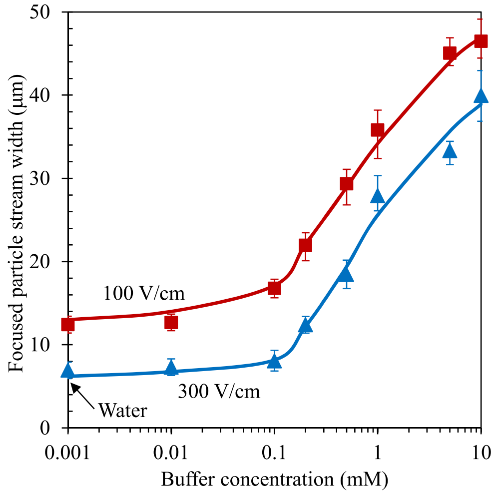

FIG. 4. Experimentally measured values (symbols with error bars, measured directly from the superimposed images in Fig. 3) of the focused stream width for 5 μm diameter polystyrene particles in the electrokinetic flow of buffer solutions of varying molar concentrations under two different DC electric fields. The solid lines represent a manual fit to the experimental data points and are used to guide the eyes only. Note that the data points at 0.001 mM are actually obtained in DI water.

Figure 3 shows the experimental images of 5 μm particles at the channel outlet in buffer solutions of three different molar concentrations (0.1, 1, and 10 mM) under the electric fields of 100, 300, and 500 V/cm, respectively. The particle migration-induced electrokinetic focusing gets better with increasing electric field in every buffer solution. This is consistent with the theoretical predictions of Yariv's theoretical models with[36] and without[57] consideration of the surface conduction effect, respectively. It is also consistent with our previous experimental studies on the same particles in 1 mM buffer solution.[47,48] The electrokinetic focusing of 5 μm particles is also enhanced with the decrease of the buffer concentration at every single electric field due to the increased surface conduction effects (or equivalently the increasing value of *Du*) as noted above. Figure 4 presents a quantitative comparison of the measured 5 μm particle stream width at the channel outlet, which reduces by nearly 2 folds (from 46.5 μm to 16.8 μm) and 4 folds (from 40.0 μm to 8.0 μm) in 100 V/cm and 300 V/cm, respectively, when the buffer concentration is decreased from 10 mM to 0.1 mM. For even lower buffer concentrations, the electrokinetic particle focusing increases less significantly in 0.01 mM buffer and eventually levels off in 0 mM buffer (i.e., water).

To understand the relative roles of the axial electrokinetic motion (the larger the weaker particle focusing) and the lateral dielectrophoretic-like migration (the larger the stronger particle focusing), we used experimental measurements to determine the electrokinetic velocity and mobility of 5 μm particles. Figure 5(a) shows the measured electrokinetic particle velocities (the observed absolute particle velocity with respect to the stationary microchannel walls) in buffer solutions of varying concentrations under the application of different DC electric fields. As expected from the classical electrokinetics,[4,8,61] the electrokinetic velocity is a linear function of the applied electric field in all buffer solutions in Fig. 5(a). The linear trendlines (through the origin of the coordinates) of the experimental data points are also included in the same plot. The slopes of these straight lines give the values for the electrokinetic mobility (defined as the electrokinetic velocity per unit electric field) of 5 μm particles in varying buffer solutions, which are displayed in Fig. 5(b). Consistent with the theory and experimental data in the review articles from Kirby and Hasselbrink,[62,63] these measured electrokinetic mobility values (proportional to the zeta potential difference between the wall and the particle[7]) increase approximately logarithmically [see the trendline in Fig. 5(b)] with the decrease in the molar concentration of the buffer solution. As such, our observed phenomenon of enhanced electrokinetic particle focusing in buffers of reduced concentrations (see Figs. 3 and 4) should result from the substantially increased electrical lift.

The observed increasing dielectrophoretic-like lateral migration of 5 μm particles in Figs. 3 and 4 also holds true for polystyrene particles of 3 μm and 10 μm in diameter. As illustrated in Fig. 6, the particle migration-induced electrokinetic focusing under a fixed 100 V/cm DC electric field increases with the decrease in buffer concentration for all three types of particles. Moreover, it increases with the increasing particle size in both 0.1 mM and 1 mM buffer solutions. The electrokinetic mobility values of 3 μm and 10 μm particles are found to be nearly identical to that of 5 μm ones in every tested buffer solution. This is consistent with our previous studies on electrokinetic motions of polystyrene particles of varying sizes from the same company.[47,48,53,56] Figure 7 shows the quantitative comparison of the normalized particle stream width (by

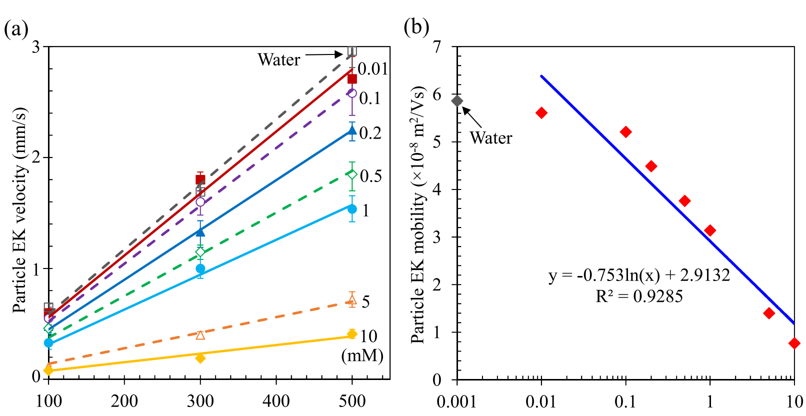

FIG. 5. The electrokinetic velocity (a) and mobility (b) of 5 μm diameter polystyrene particles in buffer solutions of varying molar concentrations. The symbols with error bars in (a) are from the experimental measurement, while the straight lines are the linear trendlines (through the origin of the coordinates, the equation and the R-squared value are displayed on the chart) of these experimental data that were obtained in Microsoft Excel. The symbols in (b) represent the data points directly determined from the slopes of the linear trendlines in (a). The straight line in (b) is the logarithmic trendline (with the equation and the R-squared value displayed on the chart) of the symbolic data points (excluding the one for water) that was obtained in Microsoft Excel.

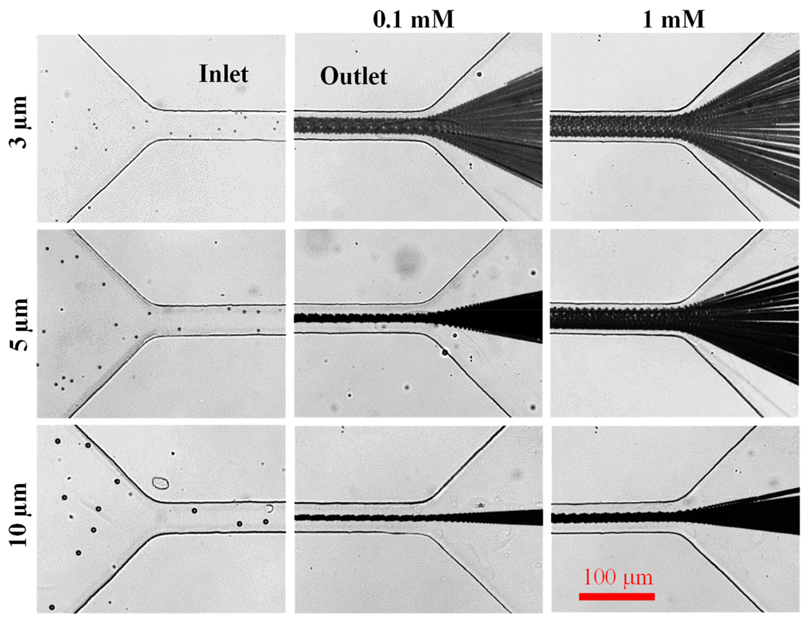

FIG. 6. Top-view images at the channel inlet (left column, snapshot) and outlet (middle and right columns, superimposed) illustrating the surface-conduction enhanced migration and focusing of polystyrene particles of varying sizes in the electrokinetic flow of 0.1 mM (middle column) and 1 mM (right column) buffer solutions. The average DC electric field across the channel length is 100 V/cm in all cases. Particles travel from left to right in all images.

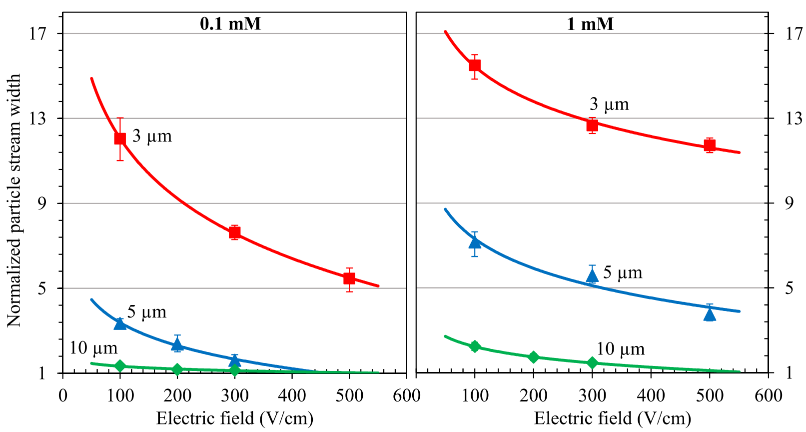

FIG. 7. Comparison of the experimentally measured (symbols with error bars) stream widths (normalized by the particle diameter) of polystyrene particles of varying diameters (as labeled on the charts) in 0.1 mM (left chart) and 1 mM buffer solutions under the application of varying DC electric fields. The solid lines represent a logarithmic fit to the corresponding experimental data points and are used to guide the eyes only.

the particle diameter) among the three types of particles under varying DC electric fields in 0.1 mM and 1 mM buffer solutions, respectively. It is apparent that the particle migration-induced electrokinetic particle focusing gets enhanced for larger particles or in a buffer solution of lower concentration or under the application of a higher electric field. This trend is again qualitatively consistent with the prediction of Yariv's model[57] if the involving Dukhin number is no less than 1 for each type of particles as noted above.

IV. CONCLUSIONS

We have performed a comprehensive experimental study of the lateral dielectrophoretic-like particle migration in the electrokinetic flow of buffer solutions of varying molecular concentrations through a straight rectangular microchannel. It is found that the electrokinetic focusing of particles toward the channel centerline is significantly enhanced when the buffer concentration is decreased. This trend remains valid for all the types of polystyrene particles under test in every single electric field. As the particle's electrokinetic velocity increases with the logarithm of the buffer concentration, we attribute the observed phenomenon to the surface-conduction enhanced particle migration in lower-concentration buffers. This trend seems to be qualitatively consistent with the prediction of Yariv's theoretical model[57] for $Du \geq 1$, which may be true for highly charged dielectric particles. For future work, we will study the effect of particle's surface charge on electrokinetic focusing of dielectric particles in straight microchannels. We will also study the lateral dielectrophoretic-like migration of conducting particles in the electrokinetic flow[64] and if/how the addition of polymers into the buffer solution[65,66] affects the wall-induced electrical lift on both dielectric and conducting particles in straight microchannels.

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