

Rectified elongational streaming due to asymmetric electro-osmosis induced by ac polarization

Jie-Tang Wu, Jiong-Rong Du, Yi-Je Juang, and Hsien-Hung Wei^{a)}

Department of Chemical Engineering, National Cheng Kung University, Tainan 701, Taiwan
and Center for Micro/Nano Science and Technology, National Cheng Kung University, Tainan 701, Taiwan

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We demonstrate a microelongational streaming generated by nonlinear electro-osmosis with high-frequency ac polarization. The phenomenon is attributed to the unique rectification mechanism that coordinates three-dimensional flow interactions between adjacent microvortices in an asymmetric quadrupole electric field. This streaming exhibits a stagnation-point structure with velocity of $300 \mu\text{m/s}$ at 100 Hz due to Faradaic polarization, but is reversed with slower velocity at 1 kHz by Ohmic charging. The measured extensional rate shows a quadratic dependence on the field in line with nonlinear Smoluchowski scale. The flow can be ready to control fluid transport and manipulate particles in microdevices. © 2007 American Institute of Physics.

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As the ability to control fluid motion and particle transport becomes a key to fulfilling a variety of functions in microfluidic devices, high-frequency ac electrokinetics provides a promising tactics for managing transport processes in small scales. It not only prevents from bubble generation commonly encountered in dc fields but also offers a precise manipulation capacity with relatively simple device designs. In this letter, we will utilize ac electrokinetics principles to demonstrate an example of engineering the flow in microscales.

Most of the electrokinetic flows are attributed to electro-osmosis (EO) that arises from the movement of mobile charge clouds driven by electric fields within the thin double layer of 10–100 nm. In classical electrokinetic theory, EO can be thought as a slip flow with a characteristic Smoluchowski velocity $U = -\epsilon\zeta E/\eta$, where ϵ is the permittivity of the fluid, η the viscosity, ζ the surface zeta potential, and E the applied field. However, as the irrotational nature does not permit vortices or closed streamlines, precise manipulation (e.g., mixing fluids or trapping particles) using *pure* EO (in the absence of pressure gradients) under dc fields will be impossible unless nonuniform charges are imposed on the surfaces¹ or other electrokinetic effects (e.g., electrophoresis due to charged particles) are included.² In addition, it could be susceptible to bubble formation or Faradaic reactions, especially if a device comprises microelectrodes that often produce large fields over small distance and hence set off Faradaic currents. All these shortcomings can be remedied by applying ac fields at frequencies much higher than the inverse Faradaic reaction time. At such high frequencies, unlike classical electrokinetics under dc fields, rapid charging and discharging make the double layer charges no longer at Poisson-Boltzmann equilibrium. Consequently, the external field will penetrate the double layer and hence produce a transient current, charging the double layer like a capacitor. As the charging varies with positions along the electrode, the induced double layer charges will become polarized with a nonuniform ζ potential of an order of the applied voltage

$V \sim EL$, with L being the electrode dimension or separation. While the field flips its polarity every half cycle and so does the induced polarization, the directions of the tangential Maxwell forces for driving the double layer charges remain unchanged. This in turn creates a time-average EO with velocity scale,³

$$U \sim \frac{\epsilon V^2}{\eta L}, \quad (1)$$

which is much faster than the dc Smoluchowski velocity by a factor of V/ζ . If the driving frequency ω is too high, the double layer will be hardly charged by the field and hence the flow will become sluggish. At very low ω , however, the external field (working at moderate voltages to avoid severe Faradaic currents) tends to be screened by the double layer, diminishing the tangential Maxwell forces for driving the flow. Therefore, the appropriate frequency for sustaining an ac electrokinetic flow at velocity scale [Eq. (1)] must be in a range near the inverse RC time,⁴ $D/(\lambda L)$, where D is the ionic diffusivity and λ the double layer thickness. In addition, there are two routes driving the flow: Ohmic charging and Faradaic charging,⁵ as illustrated by a pair of coplanar electrodes (Fig. 1).

For Ohmic charging [Fig. 1(a)] where there is no Faradaic current injection, the external field is partially screened by the double layer counterions, so the tangential field is in phase with the polarization. The resulting surface flow is outward and gives rise to a pair of microvortices above the electrodes. As for Faradaic charging [Fig. 1(b)] which occurs

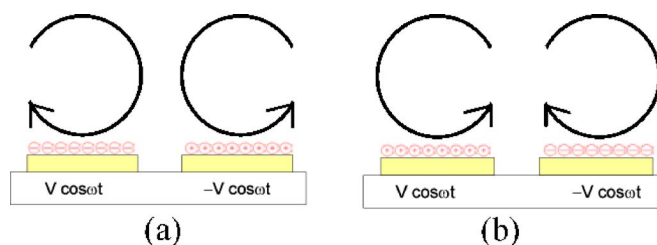


FIG. 1. (Color online) Routes driving an electrokinetic flow due to ac polarization. (a) Ohmic charging and (b) Faradaic charging.

^{a)} Author to whom correspondence should be addressed; electronic mail: hhwei@mail.ncku.edu.tw

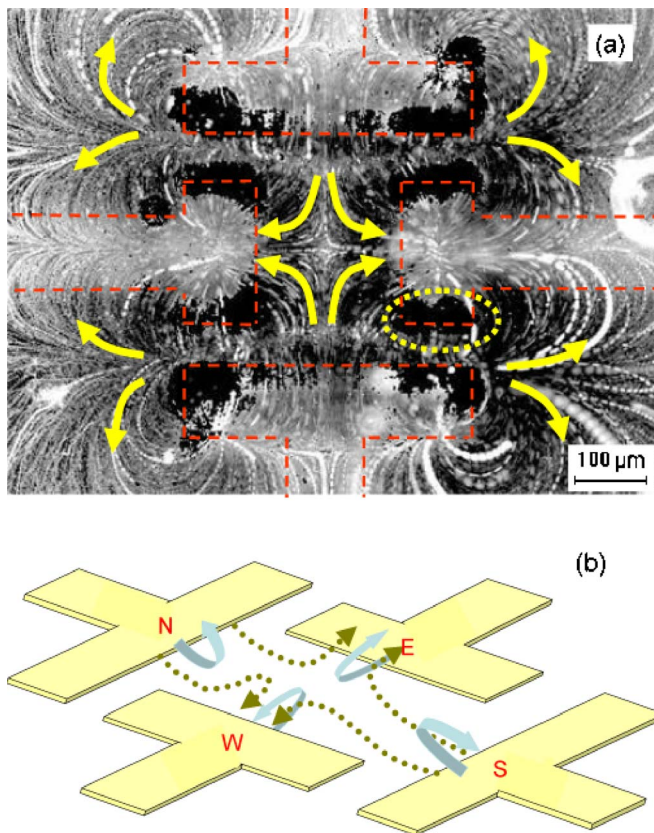


FIG. 2. (Color online) Flow pattern at 100 Hz under 20 $V_{p.p.}$. (a) Diverging stagnation-point flow in the middle with particle depletion zones as highlighted (in yellow) at the electrode corner. (b) Flow rectification mechanism.

at relatively low frequencies, since the coions tend to build up at the electrodes, the polarization will become the opposite of that in Ohmic charging and thereby reverse the circulating direction of the induced vortices. As the strong Faradaic charging will increase the zeta potential indefinitely because of the coion buildup and eventually generate bubbles,⁵ scaling Eq. (1) holds only when the electrodes are ideally polarized or Faradaic currents are small.

Here, we demonstrate an elongational streaming rectified by asymmetric ac electro-osmotic flow. Although a similar structure using ac fields has been shown previously,⁶ it was in part triggered by electrothermal effects using an additional laser source. Our flow system is driven purely by ac electrokinetic effects without moving parts. The main feature of our device consists of four T-shaped microelectrodes in a configuration for producing an asymmetric quadrupole field supplied by a function generator (Agilent 33220A). The gold electrodes have dimensions of 100 μm in width and 200 or 350 μm in length. The gap between the adjacent electrodes is 75 μm . The working electrolyte is a $10^{-3}M$ NaCl solution with the estimated double layer thickness of 30 nm. Adding the fluorescent latex particles of 0.92 μm in diameter as tracer particles in the solution, the flow patterns are observed at 50 μm above the electrode surface using a fluorescence microscope (Nikon TE2000-S) and recorded using a cooled intensified charge coupled device camera.⁷ In experiments, we choose the driving frequency ranging from 10^2 to 10^3 Hz around the inverse RC time $D/(\lambda L)$ and vary the electric field from 10 to 20 $V_{p.p.}$ (peak-to-peak voltage).

Figure 2(a) shows a stagnation-point flow in the middle region at 100 Hz under 20 $V_{p.p.}$. The flow enters from the

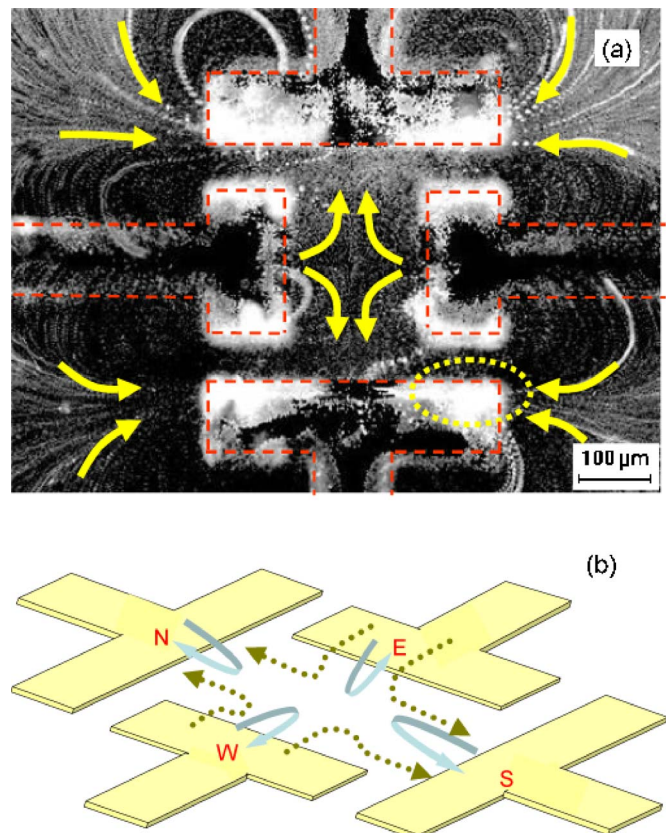


FIG. 3. (Color online) Flow pattern at 1 kHz under 20 $V_{p.p.}$. (a) Converging stagnation-point flow in the middle with particle trapping zones as highlighted (in yellow) at the electrode corner. (b) Flow rectification mechanism.

north and south electrodes, impinges in the middle, and then leaves toward the east and west electrodes. This flow is further surrounded by a number of superfast microvortices of 400 $\mu\text{m}/\text{s}$ near the sharp electrode edges. The highest velocity in the middle region is approximately 300 $\mu\text{m}/\text{s}$ for the fluid approaching the edges of the east and west electrodes. We identify that this two-dimensional elongational streaming is a result of asymmetric three-dimensional flow rectification, as illustrated in Fig. 2(b). Near the electrode, a strong flow ejects from the inner edges of the electrode, moves upward toward the center in the middle region, and recirculates back to the electrode surface. This flow behavior is mainly driven by Faradaic charging [Fig. 1(b)], as also indicated by the highlighted region in Fig. 2(a) that there are spots devoid of particles around the electrode corners where inward flow between the interelectrode space tends to deplete particles out of the electrode surfaces. In the middle region, since the local electric fields created by east and west electrodes are higher than those by the north and south ones due to shorter electrode length in the former, the induced vortices are much stronger in the east-west direction. This creates an asymmetric vortex pair between adjacent electrodes and thus rectifies the flow when these vortices come across each other in the middle region. As the flow (represented by tracer particles) ejected from the north and south electrodes arrives near the center in the middle region, instead of returning back to the electrodes to form a recirculating flow, it is redirected by stronger streams from the east and west electrodes and moves toward them. As a result, the flow behavior in a two-dimensional view portrays like a stagnation-point flow diverging along the horizontal axis.⁸

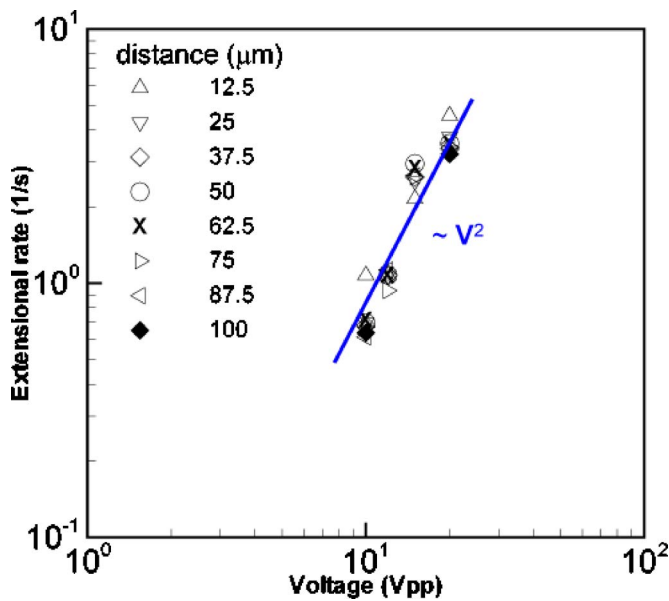


FIG. 4. (Color online) Dependence of the extensional rate on the applied voltage at 100 Hz. The data are measured by tracking the movements of the tracer particles in a range of 30–60 μm above the bottom surface. Different symbols are the data measured at various horizontal distances away from the stagnation point.

However, at 1 kHz, a similar elongational flow pattern is observed, but with the opposite flow direction (Fig. 3). The reverse of the flow is attributed to a shift of the charging mechanism from Faradaic charging at low frequencies to Ohmic charging at high frequencies. The highest velocity in the middle region is approximately 50 $\mu\text{m}/\text{s}$, which is smaller than that via Faradaic charging at 100 Hz since the double layer screening is stronger for Ohmic charging. Also, because such a sluggish flow is less effective to coordinate rectifying effects between the electrodes, the elongational flow can only be sustained for no more than 10 s. In addition, we find considerable particle trapping/accumulation at the electrode corners, as the flow induced by Ohmic charging tends to bring particles from the bulk toward the electrode gap [Fig. 1(a)].

Figure 4 is the measured extensional rate $\dot{\epsilon}$ at 100 Hz (Faradaic charging) as a function of the applied voltage V . Note that there is no apparent elongational streaming observed at voltage below 10 $V_{\text{p.p.}}$. The result shows that $\dot{\epsilon}$ follows a quadratic scale in V in accord with scaling Eq. (1). This suggests that net Faradaic currents on the electrodes could be small over a cycle. As the coion ejection likely occurs only locally on the electrodes, the incomplete double layer screening not only makes tangential polarization at work but also allows the induced EO to be enhanced by small Faradaic leakage. In addition, the largest extensional

rate is $\sim 4 \text{ s}^{-1}$ and comparable to that created purely by hydrodynamic extensional flow in microchannels.⁹

In conclusion, we demonstrate an elongational flow purely driven by ac electrokinetics. There are two underlying principles for this strategy. First, the electrode configuration must be asymmetric to permit a net flow generated from a pair of electrodes,¹⁰ a rectified streaming would be impossible if the electrode pair is perfectly symmetric. Second, the flow must be strong enough to render a sufficient penetration depth in which flow rectification between neighboring electrodes can sustain the streaming; otherwise, the flow will be simply recirculating upward or downward near the electrode edges and attenuated very rapidly away from the electrodes. In addition, as the mobility of this flow grows linearly with the applied field, it can reach as high as about $10^5 \mu\text{m}^2/(\text{V s})$ which is at least an order of magnitude faster than conventional EO under dc fields. As such, this elongational streaming, in concert with charge polarization regulated by ac fields, is robust and advantageous for precise control and manipulation of fluids or particles in small scales.

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⁴Here the RC time $\tau = R_b C_D$ can be understood in a sense that the double layer with capacitance $C_D \sim \epsilon/\lambda$ is charged by an external field through the bulk resistor $R_b \sim L/\sigma$ with the conductivity σ .

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⁷Having the electrode system housed in a slide-thick polydimethylsiloxane microchannel (of length of 5.5 cm, width of 0.2 cm, and depth of 120 μm), we turn the entire device upside down on the platform for observing the flow using an inverted microscope without being interfered by the electrodes.

⁸We also observe the flow pattern near the bottom surface and find that the extensional flow structure is virtually identical to that (at 50 μm) above the surface. In addition, the measured extension rate is 3 s^{-1} under 20 $V_{\text{p.p.}}$ at 100 Hz, which is close to 4 s^{-1} above the surface under the same condition. All the evidence above suggests that the rectified extensional flow is indeed nearly two-dimensional despite its three-dimensional origin.

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