APPLIED PHYSICS LETTERS **89**, 143508 (2006)

Fast ac electro-osmotic micropumps with nonplanar electrodes

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(Received 23 June 2006; accepted 15 August 2006; published online 3 October 2006)

This letter demonstrates dramatic improvements in flow rate and frequency range over conventional planar ac electro-osmotic (ACEO) pumps by exploiting three-dimensional (3D) stepped electrodes. A 3D ACEO pump was fabricated by electroplating steps on a symmetric electrode array and tested against a state-of-the-art asymmetric planar ACEO pump in a microfluidic loop. For all frequencies (0.1–100 kHz), the 3D pump had a faster flow rate, in some cases by an order of magnitude. Their experimental results suggest that, after some optimization, mm/s velocities will be attainable with alternating battery voltages, which presents an exciting opportunity for microfluidics. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2358823]

Microfluidics is a growing area of science and technology with important applications in biomedical devices and portable electronics. Traditional pressure-driven flows do not scale well with miniaturization, due to large viscous stresses, so other pumping techniques have been explored.[1] An attractive alternative is electro-osmosis, the effective slip of a liquid electrolyte past a solid surface in response to an applied electric field, since it does not involve any moving parts, is unaffected by (or even improves with) miniaturization and integrates well with standard microelectronics and fabrication methods.

Capillary electro-osmosis involves applying an electric field across the chip to drive plug flows through microchannels by acting on the equilibrium double-layer charge. Although widely used, this technique has some serious limitations. Since the effect is linear in the applied field, a direct current must maintained with Faradaic reactions, which can produce gas bubbles, unwanted reactions, electrode dissolution, and/or hydrodynamic instability. Moreover, a rather large voltage (e.g., 100 V across a 1 cm chip) is needed to obtain a relatively small velocity ($u \sim 100\ \mu\text{m/s}$), which exacerbates these problems and limits portability.

Since the late 1990s, several groups have begun to address these drawbacks by developing microfluidic pumps based on nonlinear electro-osmotic flow. In 1999, Ramos *et al.* reported the experimental observation of nonlinear electro-osmotic flow, varying as the square of the applied voltage, which they termed "ac electro-osmosis" (ACEO), over a pair of planar, parallel-stripe microelectrodes on a flat insulating surface,[2] as shown in Fig. 1(a). Ajdari then predicted that the same effect could be used to pump fluids over a microelectrode array by taking advantage of broken symmetry within each period, either by modifying the surface capacitance or by modulating the surface height,[3] but these suggestions have never been pursued. Instead, Brown *et al.* proposed breaking symmetry in the widths and spacings of each electrode pair in the array,[4] as shown in Fig. 1(b), and this planar design became the focus of experimental[5–7] and theoretical[8,9] studies of ACEO pumps. Some recent papers have considered pumping by traveling wave voltages in symmetric arrays, but still with planar electrodes.[10,11]

In this letter, it is demonstrated experimentally that much faster flows, with a wider frequency range, can be achieved with three-dimensional (3D) electrode arrays, consisting of asymmetrically placed steps electroplated on a symmetric planar array. Designs of this type were recently proposed by Bazant and Ben,[12] motivated by studies of induced-charge electro-osmosis around 3D metal structures.[13–15] Their simulations with the standard theory[2,3,8,9,15] (for low voltage and dilute solutions) predict faster flows in 3D versus planar ACEO pumps by more than an order of magnitude, at the same applied voltage and minimum feature size. Experiments on ACEO pumps, however, can differ markedly from the simple theory, especially at high voltage.[7] Here, we confirm the significant advantages of a 3D design, but also observe some deviations from the theory.

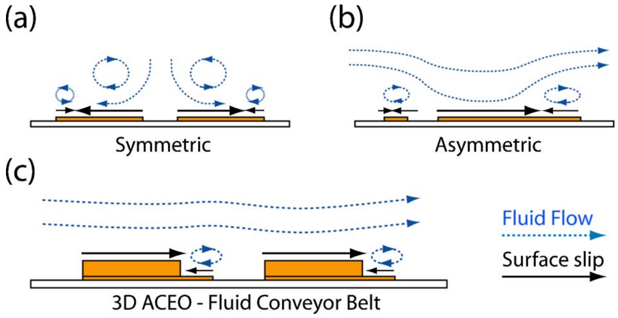

FIG. 1. (Color online) Schematic diagram of fluid flow which may be generated by an ac field applied between two electrodes on an insulating substrate. Surface slip at the electrodes, and resulting fluid streamlines are represented with solid and broken lines, respectively. (a) In the planar case, symmetrically sized electrodes will produce fluid vortices only, while (b) directional pumping may be generated using asymmetrically sized and spaced electrodes. Alternatively, symmetry can be broken by using partially raised electrodes (c).

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The basic principle of the 3D ACEO pump is illustrated in Fig. 1. In a planar electrode array, there is a competition between regions of opposing slip velocities, which perfectly cancel for a symmetric layout (a). Even in an asymmetric planar array (b), this competition remains, and the pumping velocity (proportional to the surface-averaged slip) is much smaller than the maximum slip velocity. By raising the portion of each planar electrode with slip in the desired direction, this competition is turned into cooperation, as the recessed portion with opposite slip drives a vortex, which recirculates at the level of the raised surfaces. Electric field singularities near the raised corners also enhance the flow. The net effect is to create a "fluid conveyor belt" (c), which is predicted to pump much faster than the planar design.[12]

To test this prediction in the first experimental study of 3D ACEO, several example geometries were microfabricated and systematically tested in comparison with a well characterized planar pump geometry[4,7] in a microfluidic loop.[7] The microfabrication of the 3D electrodes began by patterning sputtered metal interdigitated electrodes, with width 20 μm and spacings between electrodes of 5 μm, on transparent glass substrates. In contrast with traditional planar ACEO pumps, an electroplating process was then employed to create stepped 3D geometries, with step widths of 15 μm and heights of 2.7 μm. Heights in the range of 1–5 μm were typical in our experiments, which all showed a comparable improvement in pumping performance over planar pumps. The step height was controlled by adjusting the plating time. The step height was limited only by the thickness of photoresist used as a plating mask, although simulation suggests it is not necessary to create very tall steps to break symmetry.[12] The 3D ACEO pumps were then capped with polymer devices containing microchannels to study the fluid flow, following previous studies.[7] The interdigitated electrodes were aligned within the microfluidic loop containing a de-ionized water and fluorescent latex spheres were injected into the fluid loop opposite the electrodes to avoid electrophoresis of the spheres.[7,16] Velocities were extracted from tracer particle motion at a variety of applied ac voltages and frequencies. Details of the device fabrication and experimental procedures are available as supporting information. For comparison, the canonical planar ACEO geometry, with electrode widths of 4.2 and 25.7 μm and spacings between electrodes of 4.5 and 15.6 μm, was fabricated and tested using similar methods. The dimensions of the 3D ACEO pump studied in this letter were set to have the same 50 μm period between complementary electrode pairs as the planar pump, although a greater speedup is expected when comparing designs with the same minimum feature size.[12]

Figure 2 shows scanning electron micrographs of (a) the electroplated regions on planar substrates, and in (b) and (c) comparative images of the nonplanar and planar geometries, respectively. Further, (d) and (e) show photomicrographs of the planar and nonplanar geometries capped with polydimethylsiloxane (PDMS) loops. It should be explicitly noted that a planar version of the geometry in (b) would produce no global directional pumping due to a lack of broken symmetry.

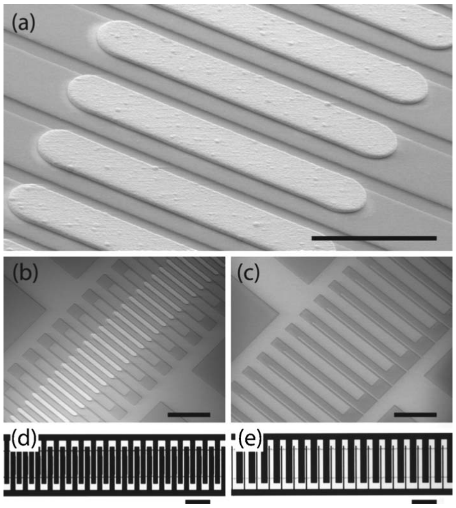

FIG. 2. (a) Detail of three dimensional electroplated steps on the interdigitated electrodes. The gold electrodes are patterned on a quartz substrate. Scale bar indicates 50 μm. The comparison between (b) nonplanar and (c) planar pump geometries clarifies a pumping lane. These ACEO designs are aligned within a PDMS microfluidic channel for characterization as pictured in (d) and (e). Scale bars in (b)–(e) indicate 100 μm.

Representative plots of centerline fluid velocity versus frequency at various peak to peak voltages (V) are shown in Fig. 3. The nonplanar pump of the present study is compared to the planar "base line" design. The peak velocity at 1 V was 10 μm/s for the base line device and was 55 μm/s for the nonplanar device. At 2 V, the base line device demonstrated a peak velocity of 75 μm/s vs 150 μm/s for the nonplanar device. Finally, at 3 V, the base line device showed a peak velocity of 150 μm/s vs 420 μm/s for the nonplanar device. In all cases, the peak performance of the nonplanar device was faster than the base line device, and at 1 V exceeded the base line device by a factor of 5. The latter is consistent with simulations using the standard low-voltage model,[12] although this particular design was not tested. At higher voltages, however, there are significant deviations from the theory evident in the frequency response.

The upper critical frequency for forward pumping in the nonplanar device increased substantially with voltage to allow a much broader band of operation. While the base line decayed quickly at frequencies above 1 kHz, the performance of the nonplanar designs demonstrated significant fluid velocity at 10 kHz for the higher voltages. Further, the performance of the nonplanar pumps improved as frequencies exceeded 4 kHz. At these higher frequencies, the difference in velocity between the base line and nonplanar devices

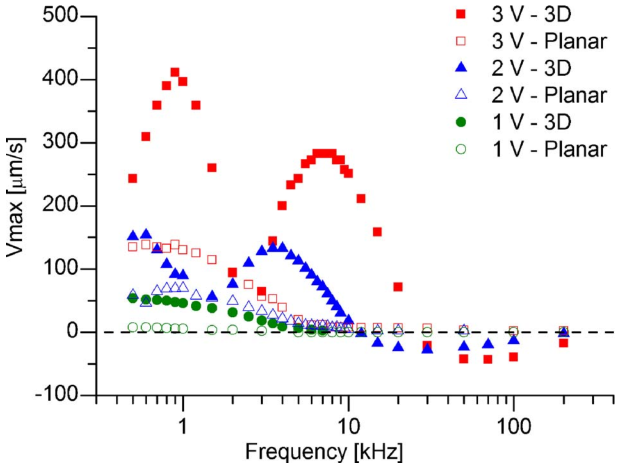

FIG. 3. (Color online) Centerline fluid velocity as a function of applied voltage (peak to peak) and frequency. The solid and open symbols represent results for nonplanar and base line devices, respectively.

becomes quite dramatic. For instance, at 2 V and 4 kHz, the base line exhibits velocity of 40 μm/s and the nonplanar device exhibits velocity of 140 μm/s. Further, at 3 V and 10 kHz, the base line device demonstrates a velocity of 10 μm/s, whereas the nonplanar device demonstrates a velocity of 275 μm/s, a factor of 27.5 difference.

At the higher voltages, there are several features not explained by the standard theory of ACEO. Curiously, the nonplanar device, at applied voltages of 2 and 3 V, exhibited a secondary peak in velocity at frequencies above 4 kHz. This double-peak behavior was not seen in the base line device at any of the operating voltages of the present study. Another characteristic only exhibited by the nonplanar devices is flow reversal: At high frequencies, both the study at 2 and 3 V tended towards negative velocities after their second peak. For the 2 V case, the crossover point was roughly 12 kHz, while the crossover was roughly 25 kHz for the 3 V case. Previous studies have observed flow reversal in base line devices,[7] though they occur at higher applied voltages and higher frequencies than those of the present study. The magnitude of velocity during flow reversal was observed to be lower than the peak velocities prior to flow reversal, though this could be in part due to the higher operating frequencies. Flow reversal at high voltage has also been observed in ACEO experiments with T-shaped planar electrodes and attributed to Faradaic reactions,[17] but a recent theoretical study has not been able to confirm this prediction.[9] In any case, the flow reversal and double-peak structure in our data pose interesting open questions for the theory of ACEO at large voltages.

In conclusion, in this letter we have experimentally shown that significant improvements in ac electrokinetic pumping may be achieved by regulating the height of the pumping electrodes. Symmetric planar electrode arrangements, which on their own do not generate directional flow, were modified using an electroplating step to break symmetry and become very fast pumps when compared to planar geometries of equal array periods. The nonplanar pumps also display a much larger frequency range, which could be useful in many applications. Theory suggests that further experiments to optimize the geometry of nonplanar pumps will lead to even more dramatic improvements in flow rate.[12] On the other hand, our observations of a double-peaked frequency response and flow reversal at high frequency highlight the need to further develop the theory of nonlinear electrokinetics at large voltages.

The authors thank Raymond Lam for assistance with data analysis. This research was supported by the U.S. Army through the Institute for Soldier Nanotechnologies, under Contract No. DAAD-19-02-0002 with the U.S. Army Research Office.

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