AC Electro-osmotic Flow

Synonyms

Induced-charge electro-osmosis, AC pumping of liquids, traveling-wave electro-osmosis.

Definition

AC electro-osmosis (ACEO) is a nonlinear electrokinetic phenomenon of induced-charge electro-osmotic flow around electrodes applying an alternating voltage.

Overview

Classical electrokinetic phenomena, such as electro-osmotic flow and electrophoresis, are linear in the applied voltage and thus cannot produce any net flow under alternating current (AC) conditions. A variety of nonlinear electrokinetic phenomena, which persist in AC fields, have been known for decades in colloid science, but the focus has been on electrophoretic mobility and particle interactions. The advent of microfluidics has stimulated interest in the use of electric fields to drive fluid flows, without any moving parts. In this context, nonlinear electrokinetics offers some unique advantages, such as the reduction of unwanted electrochemical reactions (using AC voltages) and the ability to drive fast, programmable flows at low voltages (using closely spaced micro-electrodes).

In the late 1990s, Ramos et al. discovered steady electro-osmotic flow over a pair of micro-electrodes applying an AC voltage and dubbed the effect "AC electro-osmosis" (1). Around the same time, Ajdari predicted ACEO flow over periodic electrode arrays and showed how the effect could be used for long-range pumping (2). As the performance of ACEO pumps has advanced (3,4), ACEO has also been exploited, in conjunction with dielectrophoresis (DEP), in different geometries to manipulate particles and cells in microfluidic devices (5,6,7).

Basic Methodology

Local flow generation

ACEO is a phenomenon of induced-charge electro-osmosis (ICEO), where flow is generated by the action of an electric field on its own induced diffuse charge near a polarizable surface. The main difference with other examples of ICEO, such as flows around metal colloids, is that ACEO involves electrode surfaces, which supply both the electric field and the induced screening charge, in different regions at different times. For this reason, ACEO is inherently time-dependent (as the name implies) and tied to the dynamics of diffuse charge, as ions move to screen the electrodes.

Perhaps the easiest way to understand ACEO is to consider a pair of planar electrodes applying a sudden DC voltage (which is analogous to ICEO flow around a polarizable particle in an sudden electric field). As shown in Figure 1, charge relaxation can initially be described by an equivalent RC circuit, where the diffuse layers act as capacitors, connected to "current tube" resistors of varying length through the bulk solution. Since the resistance is smaller (and the field larger) near the gap, the inner portions of double layers on the electrodes charge more quickly than the outer portions. As shown in Figure 2, this causes ICEO flow to occur, directed outward from the gap, only when the electrodes are partially screened, as the tangential field from the unscreened outer portions acts on induced charge on the inner portions. Note that the

flow is independent of the sign of the applied voltage: If the polarity were reversed, then the field and induced charges would both change sign, resulting in the same ICEO flow.

Under AC forcing, the flow peaks when the oscillation period is comparable to the charging time (Fig 2b). ACEO flow decays at higher frequencies, since there is not enough time for charge relaxation (Fig 2a). It also decays at lower frequencies, since there is enough time to completely screen the bulk electric field (Fig 2c).

Mathematical models of ACEO follow other examples of ICEO, as described in the article on nonlinear electrokinetic phenomena. A major simplification in the case of small voltages is to assume sinusoidal response to sinusoidal AC forcing and solve only for the complex amplitudes of the potential and velocity components at a single frequency ω (Fourier mode) (7,4). In this regime, the basic scaling of time-averaged ACEO flow is

$$\langle u \rangle \propto \frac{\varepsilon V^2}{\eta(1+\delta)L\left[\frac{\omega}{\omega_c}+\frac{\omega_c}{\omega}\right]^2} \qquad [1]$$

where V is the applied voltage, ε and η are the permittivity and viscosity of the liquid (both assumed constant), L is electrode spacing (roughly center to center), δ is the ratio of the diffuse-layer to compact-layer capacitances (both assumed constant). The peak frequency is at the scale of the RC charging time

$$\omega_c \propto \frac{D(1+\delta)}{\lambda L} \qquad [2]$$

where λ is the Debye screening length and D is a characteristic ionic diffusivity.

The relevant case of a large applied voltage, $V >> kT/e$ = 25 mV, is not yet fully understood. A variety of new effects could become important, such as steric effects of finite molecular sizes, viscoelectric effects, discrete electrostatic correlations, dissociation and de-solvation kinetics, and/or Faradaic electrochemical reactions passing current through the surface. In particular, "Faradaic charging" (as opposed to "capacitive charging", described above) has been proposed to explain experimentally observed flow reversal at high voltage (7), although this has not yet been borne out in a complete mathematical model of ACEO. Standard models of Faradaic reactions mainly predict suppression of the flow (by "short circuiting" of the double layer charging process) and flow reversal only in certain cases, at lower frequencies than in the experiments (2,8). Although reactions surely play a role in ACEO, high-frequency flow reversal may be attributable to other effects, such as ion crowding in the diffuse layer (9).

Fluid pumping

Regardless of the details of flow generation, there are some useful general principles to guide the development of microfluidic pumps. ACEO flow over a small set of electrodes can be used for local fluid mixing or particle trapping at stagnation points, but the flow decays quickly away from the electrode surfaces. A symmetric, periodic array of many inter-digitated electrodes (of alternating polarity at each moment in time) similarly produces an array of counter-rotating convection rolls, but no net pumping of the fluid in one direction. Instead, long-range pumping over an electrode array requires broken symmetry within each spatial period to rectify the AC forcing.

There are several ways to design ACEO pumps by breaking symmetry in a periodic electrode array. Ajdari originally suggested modulating either the electrode capacitance via a dielectric coating (Figure 3a) or the surface height (Figure 3b) with half the spatial period of the array, so that the one side of each electrode drives stronger ACEO flow compared to the other side and thus "wins" to produce net pumping over the array (2). In the first implementation of an ACEO pump, Brown, Smith and Rennie opted instead to break symmetry by using planar electrodes of different widths and gaps (10), and, until recently, this design was the only one studied experimentally (3) or theoretically (8). It has been shown to generate velocities over 100 microns/sec, although it also exhibits from poorly understood flow reversals (see below).

The performance of ACEO pumps can be greatly enhanced by designing appropriate non-planar electrode geometries. As recently predicted by Bazant and Ben (11), various "3D ACEO" designs exhibit dramatically increased flow rate without flow reversal, due to a special geometry in which the non-uniform slip profile on the electrodes all contributes to flow in the same direction. The basic idea is to create a "fluid conveyor belt" with electrodes each having steps of two different heights: On each electrode, the region of desired forward flow is raised up, while the region of reverse flow is recessed below, so as to recirculate in a vortex aiding the forward flow (rather than fighting it, as in planar designs). This can be accomplished with electrodes having electroplated metal steps, as shown in Figure 4, although other designs are possible, such as flat electrode steps deposited on a grooved surface (without the vertical metal surfaces). Simulations predict that 3D ACEO pumps are faster than planar pumps by more than an order of magnitude, at the same voltage and minimum feature size, and thus can achieve mm/sec velocities with only a few volts. This suggests using 3D ACEO pumps to drive flows in battery-powered, portable or implantable microfluidic devices.

Fluid pumping over electrode arrays can also be achieved by applying a traveling wave of voltage. At low frequency, a similar induced-charge electro-osmotic mechanism, which peaks at the RC frequency [2], is responsible for the flow (12). At high frequency (or with a thick dielectric coating on the electrodes), the classical Erlich-Melcher effect used to pump dielectric liquids, which peaks at the Debye frequency, $\sigma/\varepsilon = D/\lambda^2$, can also be observed (13). Although traveling-wave ACEO seems to produce slower flow than standing-wave ACEO with planar electrodes, the possibility of designing suitable non-planar electrodes has not yet been considered.

Key Research Findings

The groups of Ramos (1) and Green (15) have pioneered the study of ACEO by developing theoretical models and measuring flows around pairs of micro-electrodes in dilute KCl. Their work has ruled out electrothermal effects (below 5 Volts) and clarified the capacitive charging mechanism described above. The basic model successfully predicts the scaling of ACEO flow at low voltage and low salt concentration, although it tends to over-estimate velocities.

Following Ajdari (2), there has been extensive experimental (3,4,10,12,13) and theoretical (8,9,11) work on ACEO flows driven by periodic inter-digitated electrode arrays. Velocities exceeding 100 μm/sec have been demonstrated in water (10) and dilute KCl (3) by applying several volts at 1-100 kHz frequencies using asymmetric arrays of planar electrodes with unequal widths and gaps, at the scale of several microns. Somewhat slower flows have also been achieved by applying traveling-wave voltages with inter-digitated planar electrodes (12,13), although the electrical connections are more complicated.

A puzzling feature of ACEO in many situations with planar electrodes is that the flow direction can reverse (compared to the theory) with increasing voltage, typically above 2 Volts. For example, high-voltage flow reversal has been observed in dilute KCl for AC forcing (3) and traveling-wave forcing (13) and in tap water for AC forcing with a DC offset (7). As shown in Figure 5(a), the reverse flow is eventually faster than the forward flow and seems to grow more quickly with voltage than the expected scaling $u \propto V^2$.

Another puzzling feature of ACEO is the strong decay of the flow with increasing salt concentration. For this reason, all experiments in the literature have used either very dilute solutions (mostly KCl) or water (de-ionized or from the tap). A few groups have studied the concentration dependence of ACEO in aqueous KCl solutions (3,15,16). These experiments and recent work on electrokinetic motion of heterogeneous particles suggest that flows due to induced-charge electro-osmosis exhibit a universal decay with concentration like $u \propto \log(c_{\max}/c)$ at large voltage (9). As shown in Figure 5(b), the flow effectively vanishes above a bulk salt concentration of only $c_{\max} \approx 10$ mM, which is an order of magnitude smaller than most biological solutions.

Nevertheless, advances have been made in applying ACEO flows to pumping, mixing, and trapping particles in microfluidic devices. Experiments on 3D ACEO pumps with non-planar stepped electrode arrays have demonstrated an order of magnitude increase in flow rate versus the fastest planar design, due to the robust fluid-conveyor-belt mechanism (4). For a wide range of near-optimal step heights, there is also no observed flow reversal (14). Therefore, it is now possible to drive fast, reliable mm/sec flows in water or dilute electrolytes with less than 3 Volts, which can be provided by a small lithium battery.

Progress has also been made in exploiting ACEO flows to manipulate colloidal particles and biological cells in microfluidic devices (5,6,7). The basic strategy is to use ACEO flow to draw particles to stagnation points on the electrodes, where they are trapped, presumably by DEP (although the classical theory does not seem to predict this effect). By increasing the voltage, the ACEO flow can be reversed, and particles are observed to move away from the stagnation lines, overcoming any remaining trapping force. In this way, it is possible to "write" and "erase" suspended particles, bacteria, or micro-algae on microelectrodes, as shown in figure 7(a). This effect can be enhanced by added a DC bias voltage to the low-frequency AC voltage (50-100 Hz) between adjacent electrodes (7). Particles are observed to collect only on the positively biased electrode, as shown in Figure 7(b). It has been suggested that opposing ACEO flows are produced by the competition between Faradaic charging on one electrode (positive bias) and capacitive charging on the other (negative bias), but a quantitative theory remains to be developed and confirmed systematically by experiments.

Future Directions for Research

ACEO is attractive to exploit in microfluidics since it offers precise local flow control without moving parts, using standard fabrication methods from the electronics industry. Device operation requires only the voltage (few Volts), power (mW), and current (mA) of a small battery, and Faradaic reactions are reduced or eliminated. These features make ACEO a promising route to portable or implantable microfluidic systems. In contrast, DC electro-osmotic pumps, such as those exploiting electroosmotic flow in porous media, typically require several 100 volts and produce bubbling at the electrodes due to electrolysis, which can require a catalytic gas management system. On the other hand, ACEO pumps have so far only

produced very small pressures (< 10 Pa) far below those of porous-plug DC pumps (> 100 kPa), but new designs could close this gap. In any case, the ability to tune flows and particle motion by ACEO at the micron scale by modulating relatively small AC voltages could have many uses, e.g. for mixing, flow fragmentation, or manipulation of colloids and cells.

Understanding flow reversal in ACEO is an important open question, from both fundamental and practical points of view. As noted above, it has been attributed to Faradaic reactions, using theoretical arguments suggesting a scaling $u \propto e^V$ (7). Simulations of ACEO pumps using standard Butler-Volmer reaction kinetics have failed to predict any flow reversal, except at low frequencies below 100 Hz (2,8), although this may be consistent with experiments showing low-frequency flow reversal with a DC bias (7). In contrast, high-voltage flow reversal with unbiased AC forcing in electrode-array pumps tends to occur at high frequency (>10 kHz), as shown in Figure 5. An alternative hypothesis, which predicts high-frequency, high-voltage flow reversal due to capacitive charging alone, is based on the reduction of double-layer capacitance due to ion crowding effects (9). The question is far from settled, however, and more theoretical and experimental work is needed.

Another important open question concerns the dependence of ACEO flow on the solution chemistry (concentration, ionic species, solvent characteristics, etc.). The suppression of ACEO above 10 mM bulk salt concentration limits its use in biological applications and points to possible new physics of the double layer at large voltages, such as a dramatic increase in the diffuse-layer viscosity upon ion crowding (9). Very few electrolytes have been studied experimentally, but current work suggests that varying the ionic species can also substantially alter the flow. An interesting direction for future research would be to develop surface coatings, electrode materials, and buffer solutions to mitigate or manipulate these complex interfacial phenomena.

Cross-references

Dielectrophoresis
Dielectrophoretic Motion of Particles and Cells
Electrical Double Layers
Electrokinetic Motion of Polarizable Particles
Electrokinetic Motion of Heterogeneous Particles
Electroosmotic Flow
Electro-osmotic Flow in Porous Media
Induced-Charge Electro-osmosis
Nonlinear Electrokinetic Phenomena

Further Reading

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Figure Legends

Figure 1

Equivalent RC circuit model for double-layer charging over a pair of electrodes. The inner edges of the electrodes encounter less bulk resistance (due to shorter "current tubes") and thus charge more quickly than the outer edges. (Reproduced from Ramos et al 1999.)

Figure 2

The basic mechanism of AC electro-osmosis: Electrochemical relaxation (top) and induced-charge electro-osmotic flow (bottom) in response to a suddenly applied voltage across an electrode pair. (a) At first the electric field has no tangential component on the electrodes, since they are equipotential surfaces, and thus there is no electro-osmotic flow. (b) Capacitive double-layer charging begins near the gap where the initial normal current is strongest and causes the unscreened field lines dip down and provide tangential components over the induced charge; the result is ICEO flow directed away from the electrode gap. (c) After the charging time passes, the electrodes are fully screened, leaving no electric field and thus no flow. An AC voltage can drive a steady time-averaged flow, similar to (b), if its period is comparable to the charging time.

Figure 3

Sketches of local broken symmetries in a periodic electrode array which lead to global time-averaged ACEO pumping: (a) non-uniform surface coatings; (b) non-uniform surface height. (Reproduced from Ajdari 2000.)

Figure 4

Top: Simulations of ACEO microfluidic pumps, showing the time-averaged flow over a pair of micro-electrodes (dark regions) in one spatial period of an interdigitated-electrode array. (a) A nearly optimal planar design with different electrode sizes and gaps; the smaller electrode has the largest local slip velocity, but the larger electrode "wins" in overall pumping from left to right. (b) A much faster 3D ACEO design with stepped electrodes having a symmetric footprint and the same minimum feature size; the reverse slip now recirculates in a vortex to create a "fluid conveyor belt" for the raised pumping flow from left to right. (Reproduced from Bazant and Ben 2006). Bottom: Scanning electron microscopy images of each design fabricated in gold on glass with minimum feature size (gap) of 5 microns (courtesy of JP Urbanski and JA Levitan, using methods of ref 4).

Figure 5

Experimental data for ACEO pumping of KCl around a microfluidic loop, one-fifth covered by the asymmetric planar electrode array of Fig. 4(a). (a) Contour plot of mean velocity versus AC frequency and RMS voltage at a bulk salt concentration of 0.1 mM (reproduced from Studer et al 2004, Ref 3), where "electrode damaging" refers to bubble formation and inability to observe consistent flows. (b) Velocity versus AC frequency at 2 Volts peak-to-peak for different concentrations (courtesy of JP Urbanski). Both show high-voltage, high-frequency flow reversal, and the latter shows the strong decay with salt concentration.

Figure 6

(a) Comparison of ACEO pumping of water around a microfluidic loop by planar and (non-optimal) 3D electrode arrays with similar 5 μm minimum feature size (reproduced from Urbanski et al 2007, ref 4). (b) Experimental optimization of the step height for 3D ACEO pumping of dilute KCl (reproduced from ref 15).

Figure 7

(a) Collection of *E. Coli* bacteria in tap water along the stagnation lines of ACEO flow on Au microelectrodes at low frequency (100 Hz) and moderate voltage (1 V). (b) Preferential particle trapping by asymmetric polarization ACEO on electrodes with positive DC bias at higher voltage (> 3 V). (Reproduced from Wu 2006, Ref 7)

Figures

Figure 1

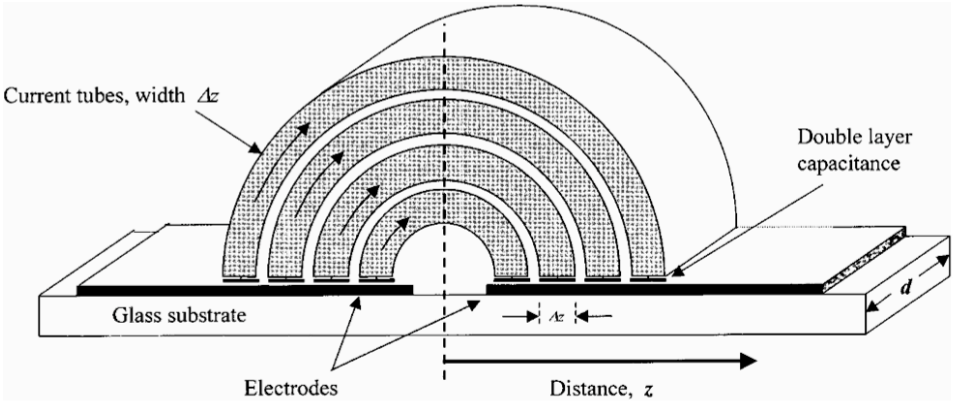


Figure 2

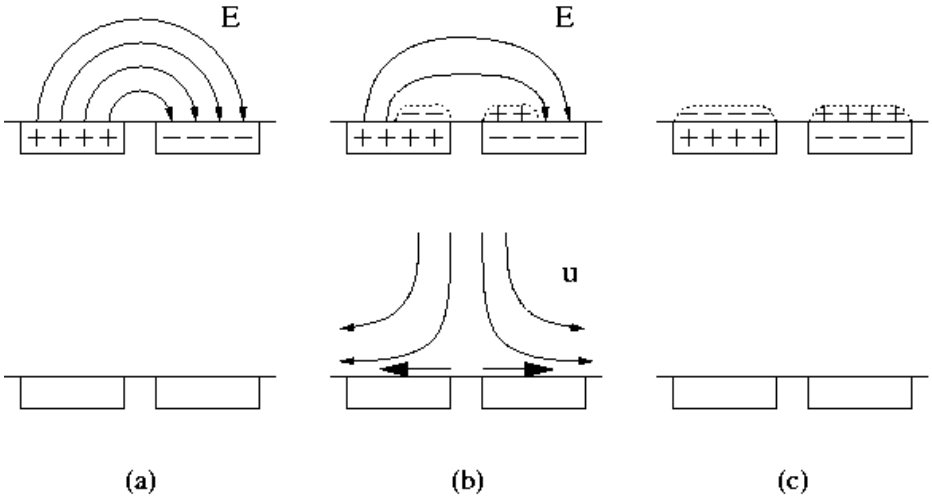


Figure 3

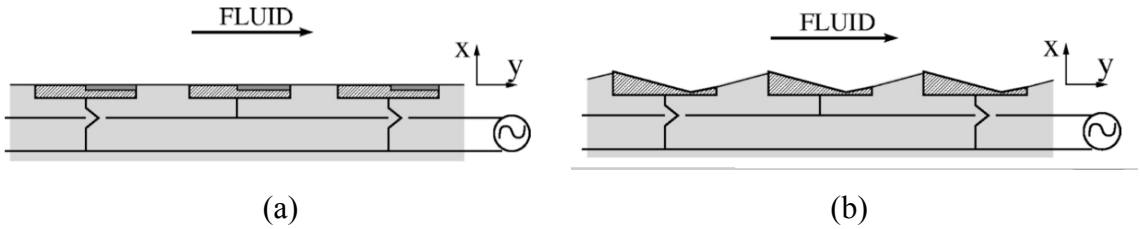
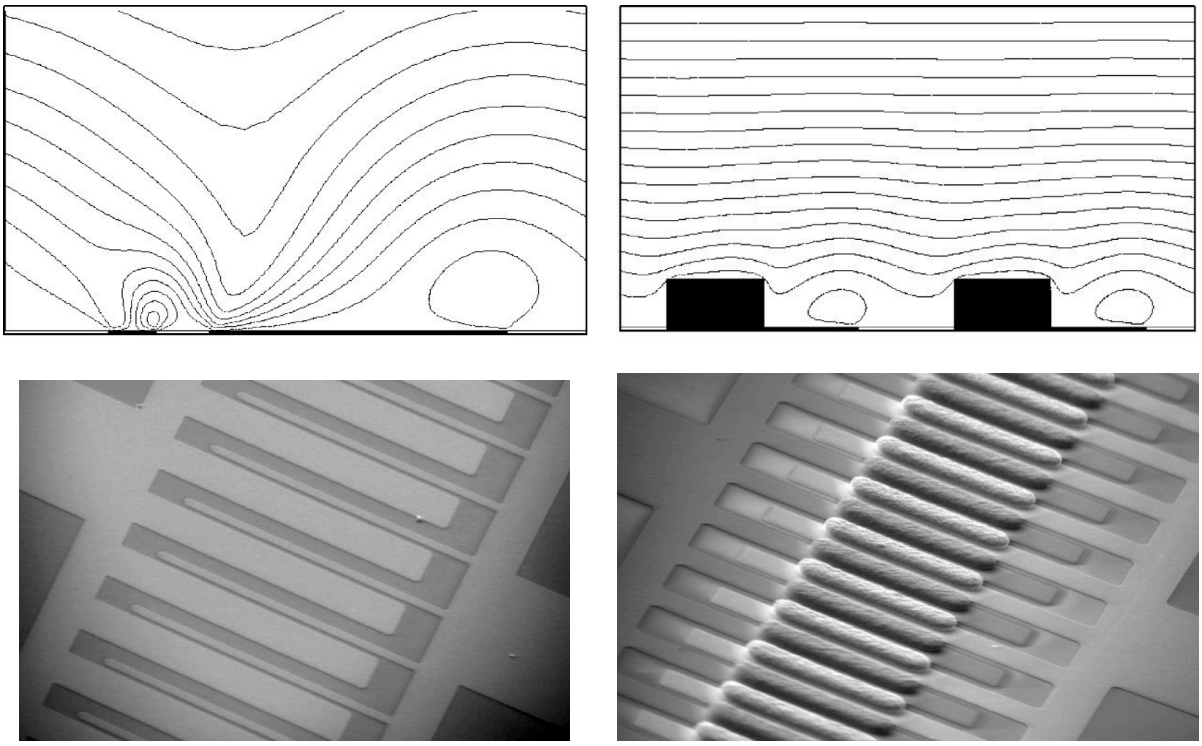
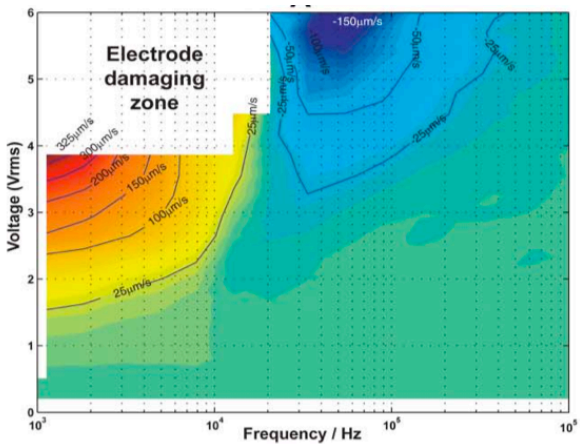


Figure 4



(a) (b)

Figure 5



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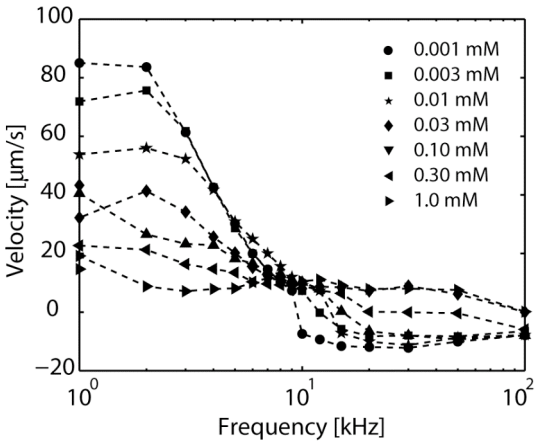
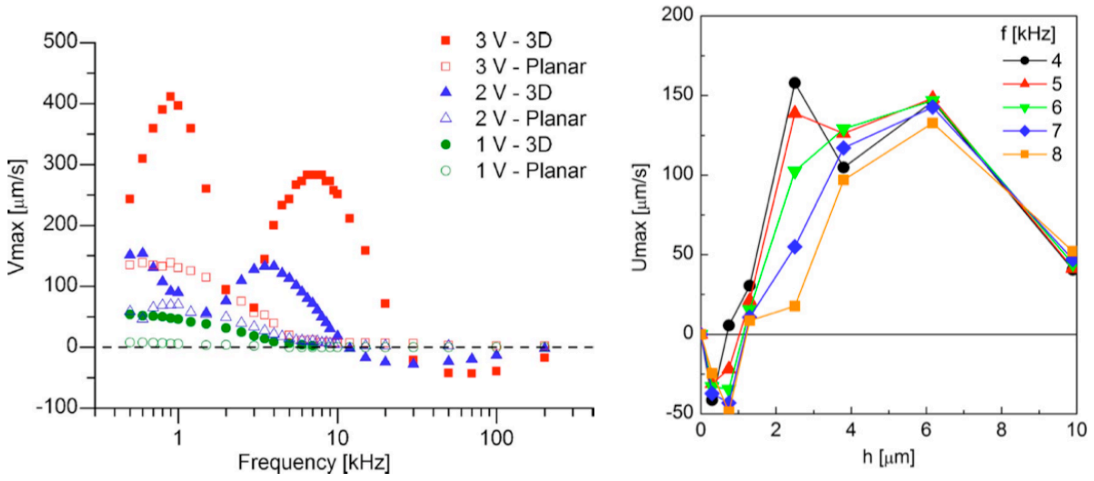


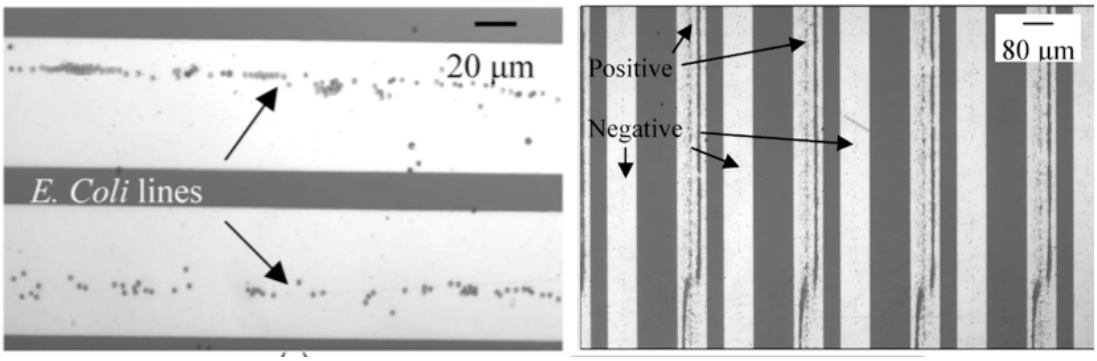
(b)

Figure 6



(a) (b)

Figure 7



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