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RAPID COMMUNICATION

Electric field induced fluid flow on microelectrodes: the effect of illumination

Nicolas G Green†§, Antonio Ramos†, Antonio González†, Antonio Castellanos† and Hywel Morgan‡

† Departamento Electronica y Electromagnetismo, Facultad de Fisica, Universidad de Sevilla, Reina Mercedes s/n, 41012 Sevilla, España
‡ Bioelectronics Research Centre, Department of Electronics and Electrical Engineering, University of Glasgow, Rankine Building, Oakfield Avenue, Glasgow G12 8LT, UK

E-mail: ngreen@cica.es

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Abstract. The electrokinetic manipulation of particles suspended in a fluid medium is accomplished using microelectrodes that generate non-uniform fields of significant strength from low applied potentials. The high strength fields produce not only forces on the particles but also on the fluid medium used for suspension. This paper presents qualitative and semi-quantitative observations of the movement of the fluid at applied field frequencies of the order of 1MHz and higher. The importance of the illumination in generating the fluid flow is described, the flow depending on both the intensity of illumination and the applied electric field. The theory of electrothermally induced fluid flow is briefly described and compared with the experimental observations. Reasonable agreement is found between the experiments and the theory, with the light generating temperature gradients, and therefore gradients in fluid permittivity and conductivity, and the electric field responsible for the motive force.

(Some figures in this article appear in colour in the electronic version; see www.iop.org)

Microelectrode structures, such as those used for dielectrophoresis (DEP) [1], generate high strength, non-uniform ac electric fields. Microelectrodes have been used for the dielectrophoretic manipulation of particles in solution over a range of sizes, from cells (∼10 μm diameter) down to viruses (∼100 nm diameter) [2–5]. However, the strong electric fields can also interact with the suspending fluid medium, to produce forces on the fluid and hence flow. The study of this interaction is referred to as electrohydrodynamics (EHD) [6–8].

There are a number of mechanisms through which an electric field can interact with a fluid to produce a force. Recently, we reported a new type of fluid flow occurring in microelectrodes at frequencies below 100 kHz [9–11] arising from the interaction of the non-uniform field and the induced charges in the electrical double layer at the electrode-solution interface. This flow, referred to as ac electro-osmosis, is not fully understood and has been characterized and discussed in other publications [9–11].

This communication is concerned with fluid flow patterns observed in microelectrodes at frequencies around 1 MHz and above [5], where electrode polarization and ac electro-osmosis are negligible. The mechanism has been postulated to be electrothermal [7], where the electric field acts on gradients in permittivity and conductivity produced by non-uniform heating of the fluid [6]. For small changes in temperature, where the relative increments in permittivity $\Delta\varepsilon/\varepsilon$ and conductivity $\Delta\sigma/\sigma$ are much smaller than 1, the force on the fluid per unit volume is [7]:

$$\langle \boldsymbol{f}_E \rangle = \frac{1}{2}\mathrm{Re}\left[\left(\frac{(\sigma\nabla\varepsilon - \varepsilon\nabla\sigma)\cdot\boldsymbol{E}}{\sigma + \mathrm{i}\omega\varepsilon}\right)\boldsymbol{E}^* - \frac{1}{2}|\boldsymbol{E}|^2\nabla\varepsilon\right] \quad (1)$$

where $\boldsymbol{E}$ is the applied electric field and $\mathrm{i} = \sqrt{-1}$. The first term on the right-hand side represents the Coulomb force and the second the dielectric force. The gradients of the permittivity and conductivity are related to the temperature gradient by the expressions, $\nabla\varepsilon = (\partial\varepsilon/\partial T)\nabla T$ and $\nabla\sigma = (\partial\sigma/\partial T)\nabla T$. The volume force is dominated by the dielectric force at high frequencies ($\omega \gg \sigma/\varepsilon$). At low frequencies ($\omega \ll \sigma/\varepsilon$), the Coulomb force dominates since, for an aqueous solution, the relative change in the conductivity is greater than that for the permittivity.

In this paper, experimental observations and preliminary measurements of fluid flow on microelectrodes at frequencies around the charge relaxation frequency of the medium ($\omega \sim \sigma/\varepsilon$) are presented and discussed. The previously unreported importance of the intensity of the illumination

§ Corresponding author.

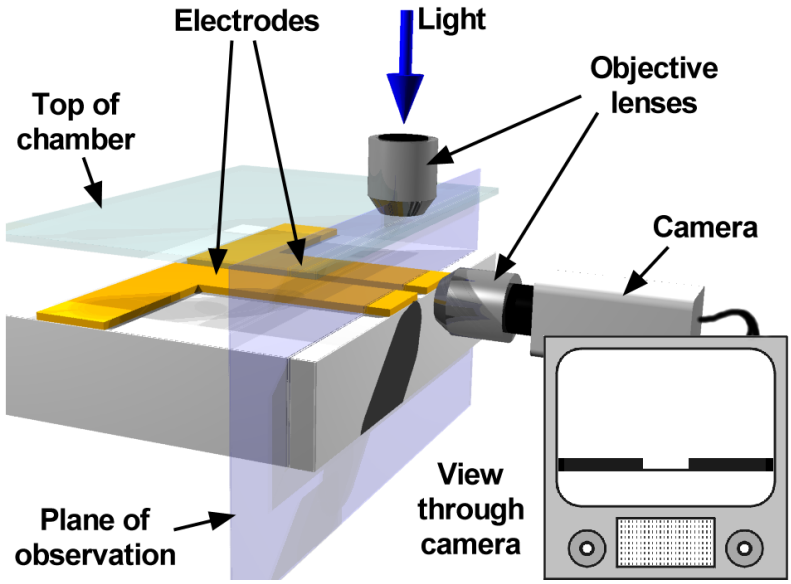

Figure 1. A schematic diagram of the experimental set-up, with the two plate electrodes (1×0.5 mm) separated by a parallel gap of 25 μm. The electrodes are enclosed in a chamber constructed from thin glass cover slips and observed both from above and the end. The sample is illuminated from above by a fluorescence microscope and the plane of observation for the horizontally placed objective is shown.

in determining the fluid flow profile and magnitude is highlighted.

Mizuno *et al* [12] have reported a flow pattern created around an infrared laser focal point in an ac electric field. The fluid would absorb the infrared light and temperature gradients would appear. It seems likely that the electrothermal mechanism is also responsible for the fluid flow but electrode polarization effects may be important below 1 MHz in this case [10].

Carboxylate modified latex spheres (Molecular Probes, Oregon, USA) 557 nm in diameter were used as tracer particles to observe the movement of the fluid. These particles, typical of those used in DEP experiments [13, 14], are strongly fluorescent and small sizes can be clearly observed, even at low light intensities. The suspending medium was potassium chloride with a measured conductivity of 2.1×10^{-3} Sm^{-1}. A simple electrode design was used for the experiments, consisting of two plates, 1 mm long and 0.5 mm wide, with parallel edges separated by 25 μm, similar to electrodes used previously for fluid flow measurements [9–11]. The electrodes were fabricated on planar glass substrates using photolithography and consisted of three layers: 10 nm titanium, 100 nm gold and 20 nm titanium. A chamber was constructed around this microelectrode using glass cover slips. The end of the chamber was closed with a cover slip as shown in figure 1, so that the particles could be observed from the end. The behaviour of the fluid could therefore be observed in two planes: horizontal and a vertical cross-section of the electrodes. The electrodes were illuminated and observed using epi-fluorescence microscopy (Nikon Microphot). A second microscope and objective were aligned horizontally with the electrodes and used to observe behaviour in the vertical direction. Figure 1 shows a schematic diagram of the experimental set-up.

A sample consisting of different sizes of spheres (557 nm, 216 nm and 93 nm diameter) was prepared and pipetted into the chamber. The ac signals generated by a digital synthesizer, with frequencies in the range 100 Hz to 20 MHz and voltages up to 20 volts peak to peak, were then applied to the two electrodes.

With uniform illumination from the fluorescent microscope, flow was observed as shown in figure 2. This figure is a superimposition of successive video frames, clearly showing the particle trajectories above the electrode due to the movement of the fluid. The pattern of the movement consisted of two rolls, one over either electrode, symmetrical around the vertical plane running along the centre of the gap. From 0.5–1.5 MHz, the flow was observed to follow the arrows shown, moving from the region above the electrodes, towards the gap between the electrodes and then vertically upwards. As the flow reached the top of the chamber, the fluid circulated, forming a convection pattern. At 2 MHz, no fluid flow was observed and the particles remained almost stationary. At 3 MHz and above, the fluid flow pattern was the same as at 1.5 MHz but in the opposite direction, with the fluid moving from the region above the gap between the electrodes downwards and out across the electrode surface. The fluid flow velocity increased as the applied potential increased. The velocity of the particles was measured in a region at a height of $\sim$40 μm, far enough away from the electrodes so that the effects of the dielectrophoretic force were negligible. For a peak to peak potential of 20 volts with a frequency of 1 MHz, the measured velocity was approximately 80 μm s^{-1} upward. At 5 MHz the velocity was 15 μm s^{-1} downward.

For a two-dimensional parallel plate geometry, it is expected that Joule heating would produce fluid movement similar to the observations. However, the importance of the intensity of the light used to illuminate the device was not expected: as the intensity of the light was reduced, the velocity of the fluid flow decreased. At the lowest intensity at

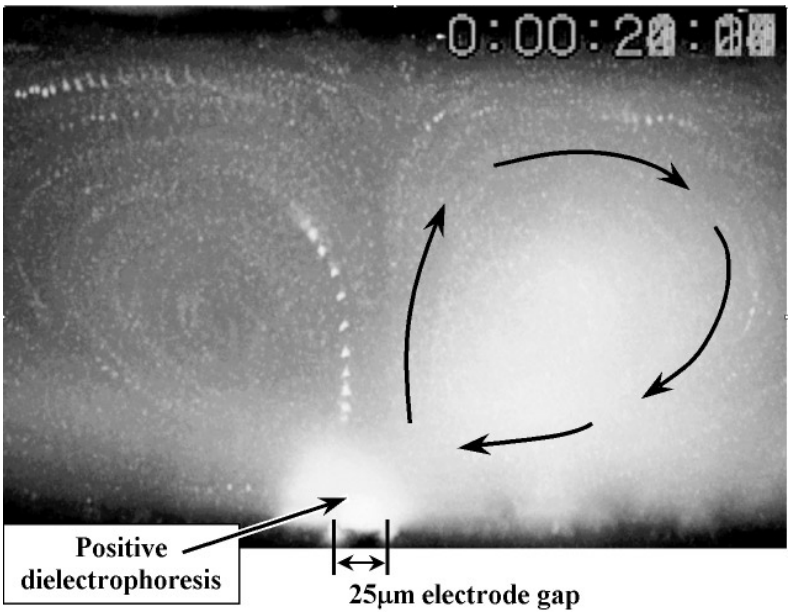

Figure 2. Diagram of the fluid flow observed above the planar microelectrodes in the plane of observation as shown in figure 1 for uniform illumination. This diagram is a superimposition of several images over a period of four seconds showing individual particle tracks. The arrows show the direction of the movement, which was symmetrical about a vertical line through the centre of the 25 μm gap. The fluid moved along the direction indicated by the arrows for applied field frequencies up to 1.5 MHz. For a frequency of 3 MHz and greater, the fluid moved along the same lines but in the opposite direction.

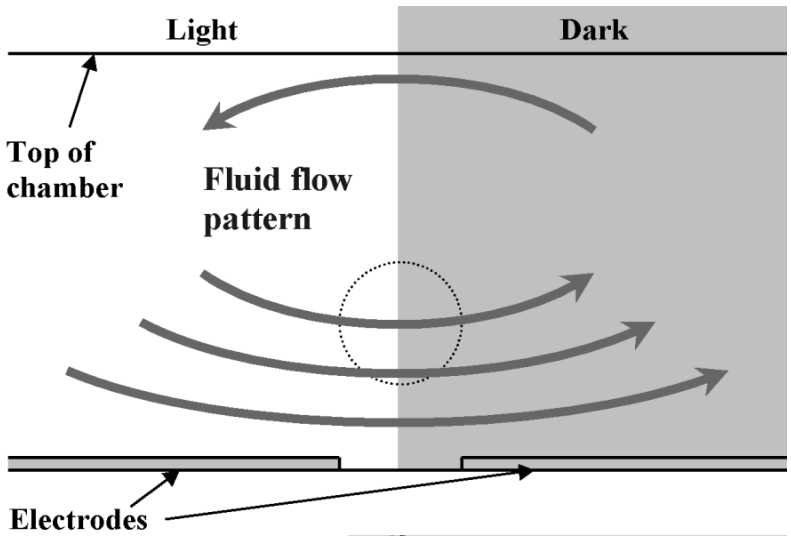

Figure 3. Schematic diagram of the fluid flow observed above the planar microelectrodes in the plane of observation as shown in figure 1, for the case of only one electrode illuminated. The fluid moved in the direction indicated by the arrows at a frequency of 1 MHz and in the opposite direction at 5 MHz.

which the particles could still be seen there was no observable fluid motion. Under these conditions and at 1 MHz, the particles behaved as expected from simple positive DEP, moving to and collecting at the electrode edges. At 5 MHz, the particles were repelled from the electrode edges by negative DEP (also as expected). Increasing the intensity of the light resulted in the onset of fluid flow following the pattern shown in figure 2.

In order to demonstrate the importance of the light in the generation of fluid flow, the objective lens of the microscope was moved to illuminate only one of the two electrodes. This produced the flow patterns shown in figure 3. At 1 MHz, the fluid followed the arrows shown, moving from the region above the lit electrode, toward the gap between the electrodes and then outwards over the dark electrode. At the top of the chamber, there was a flow in the opposite direction producing a circulation. The velocity was measured in the region indicated by the circle, approximately 40 μm above the surface, and was found to be approximately 80 μm s^{-1}. At 5 MHz, the direction of the flow was opposite to the arrows shown in the figure and the measured velocity was $\sim$10 μm s^{-1}.

The mechanism responsible for this fluid flow is likely to be electrothermal. However, as discussed in [7], the source

of the temperature rise and the gradients in the medium cannot be Joule heating (from the electric field), as this would produce fluid flows much smaller than those observed experimentally. This is confirmed by the fact that at very low light levels the flow vanishes. Since the fluid flow depends both on the electric field and the intensity of illumination (without either, there is no observable flow), a plausible explanation is that the mechanism producing the force on the fluid is electrothermal and the source of the temperature gradient the illumination. Removing either the field or the light results in cessation of the fluid flow.

As discussed by Ramos *et al* [7], in electrodes similar in design to those used in this paper, the electrothermal force given by equation (1) is frequency dependent with a characteristic frequency:

$$f_c = \frac{\sigma}{2\pi\varepsilon}\left(2\left|\frac{1}{\sigma}\frac{\partial\sigma}{\partial T}\right| \Big/ \left|\frac{1}{\sigma}\frac{\partial\sigma}{\partial T}\right|\right)^{\frac{1}{2}}. \qquad (2)$$

For the electrolyte used in this work and with textbook values for $(1/\sigma)(\partial\sigma/\partial T)$ of +2% per degree and $(1/\varepsilon)(\partial\varepsilon/\partial T)$ of -0.4% per degree [15], f_c can be calculated to be 1.52 MHz. This frequency is close to the experimentally observed frequency of transition in fluid flow direction (approximately 2 MHz). Furthermore, the expected difference in magnitude between the Coulomb force at low frequencies and the dielectric force at high frequencies can be calculated to be 6.7 [7]. This also agrees with the experimentally observed ratio between the fluid flow frequencies on either side of the transition, approximately 5.5. It should be noted that the geometry used by Ramos *et al* [7] is different from the experimental geometry and may account for the differences.

There are several mechanisms through which the light could produce heat in the system. If there were an infrared component to the illumination, this would directly heat parts of the system. However, experiments were performed using an infrared filter that removed everything above ~720 nm in wavelength and no measurable change in the fluid flow was observed.

Also, the fluorescent particles gain energy through the absorption/re-emission process and could generate heat, but since the particles are essentially uniformly distributed, this would not generate global temperature gradients. It could however, result in fluid flow if there were a high concentration of particles, for example at electrode edges during positive DEP collection.

Another possible explanation is that the light is heating the electrodes, since a percentage is absorbed by the metal during reflection [16]. Since the electrodes are thin, and therefore have a higher thermal resistance than water, the heat will radiate away through the electrolyte creating temperature, conductivity and permittivity gradients. The electric field then interacts with these gradients producing fluid flow. This is illustrated by figure 4 for the two experimental cases. Where the illumination is uniform, the temperature gradient would be directed towards the electrodes as shown in figure 4(a). At low frequencies, assuming a simple circular electric field, the force can be expressed as:

$$\langle \boldsymbol{f}_E \rangle \approx \frac{1}{2}\frac{\varepsilon}{\sigma}\frac{\mathrm{d}\sigma}{\mathrm{d}T}\,|\nabla T|\,|\boldsymbol{E}|^2\cos\varphi\hat{\boldsymbol{u}}_\varphi \qquad (3)$$

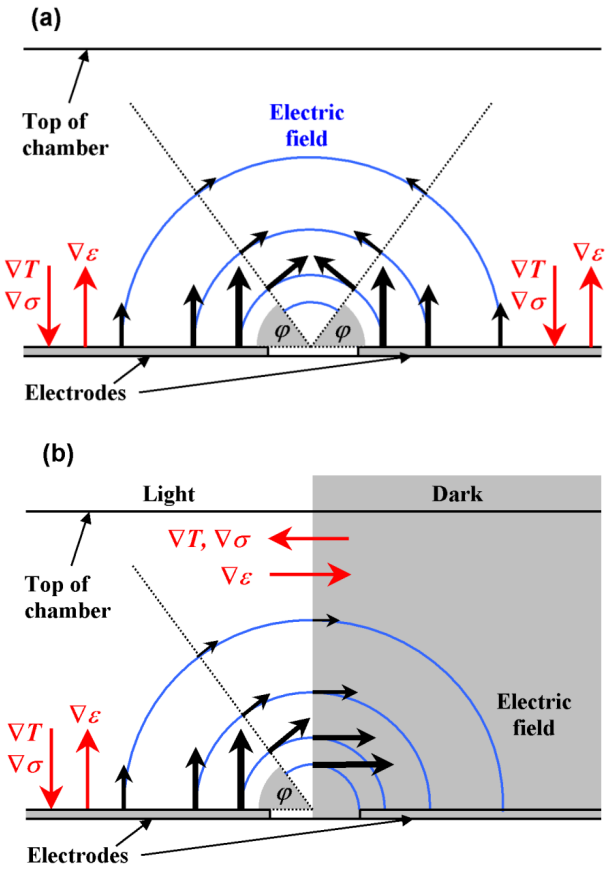

Figure 4. Schematic diagram of the temperature gradient ∇T, the permittivity gradient $\nabla\varepsilon$ and the conductivity gradient $\nabla\sigma$, produced if electrode heating is present in the system. The solid lines indicate the electric field lines and the unmarked arrows indicate direction and approximate strength of the force on the fluid produced by the interaction of the field and the gradients in the system. The case of uniform illumination is shown in (a) and the case of one electrode illuminated is shown in (b).

where φ is the angle shown in the figure and $\hat{\boldsymbol{u}}_\varphi$ is the angular unit vector in circular polar co-ordinates. The approximate volume force on the fluid at different positions is also shown in the figure. The cumulative effect would be to produce circulating patterns similar to that observed in figure 2. At frequencies above f_c it can be calculated that the curl of the force is in the opposite direction, creating a pattern similar to observations at higher frequencies. The case where only one electrode is illuminated is shown in figure 4(b). In this case, the same force profile would be generated but only over one electrode. In addition, there would be a thermal gradient from the illuminated region to the dark region and this would produce a horizontal force component close to the electrode surface where the field is strongest. At low frequencies, the direction of this force is from hotter to colder regions and at high frequencies in the opposite direction. The force profile is shown in figure 4(b) for the case of low frequencies. Again, this would produce a flow pattern similar to the experimental observations (figure 3).

In conclusion, it has been demonstrated that an electric field can drive fluid flow where the temperature gradients are generated by the illumination source. The fluid flow pattern

and frequency dependence are in accordance with the theory of electrothermal body forces. A qualitative explanation based on a simple electric field geometry and a temperature gradient created through heating of the electrodes by the light adequately describes the experimental observations. These findings may be of interest to researchers in the field of electrokinetics since this type of fluid flow could affect observations and measurements.

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