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ELECTROTHERMAL LIQUID MOTION IN MICROSYSTEMS SUBJECTED TO ALTERNATING AND ROTATING ELECTRIC FIELDS

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ABSTRACT

Electrothermal motion in an aqueous solution appears when an electric field is coupled with thermally-induced gradients of conductivity and permittivity in the fluid. The temperature field can be produced by external sources, such as strong illumination, or caused by the applied electric field through Joule heating. Electrothermal flow in microsystems is usually important at frequencies around 1 MHz and voltages around 10 V. In this work, we consider first the two-dimensional problem of an aqueous solution placed on top of two co-planar electrodes that are subjected to an ac potential difference when there is either a vertical or horizontal temperature gradient. Secondly, we study the three-dimensional problem of an aqueous solution lying on four co-planar electrodes which produce a rotating field. This electric field when combined with a vertical temperature gradient rotates the liquid. The resulting electric field and liquid motion in these problems are characterised using self-similar solutions. Finally, these analytical solutions are compared with numerical and experimental results.

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INTRODUCTION

Flows of electrothermal origin often appear in the manipulation of colloidal particles by means of ac electric fields in microsystems [1–4]. Electrothermal fluid flow is due to the coupling of an electric field with thermally-induced gradients of conductivity and permittivity in the fluid. The temperature field can be produced by external sources, such as strong illumination [3], or caused by the applied electric field through Joule heating. Observations show that the electrothermal effect is important in microsystems for frequencies of the order of 1 MHz and voltages of the order of 10 V.

In this work, we present some analytical solutions for the fluid flow induced by electric fields coupled with externally imposed gradients of temperature in microelectrode structures. We consider first the two-dimensional problem of an aqueous solution placed on top of two co-planar electrodes with parallel edges that are subjected to an ac potential difference when there is either a vertical or a horizontal gradient of temperature. The resulting electric field and liquid motion are characterised using self-similar solutions. These solutions are relevant to some fluid flows observed under strong illumination [4]. Secondly, we study the three-dimensional problem of an aqueous solution lying on four co-planar electrodes placed in each quadrant. When the ac voltages applied to adjacent electrodes differ by 90^o in phase, it results in a rotating field. The electrical problem can be solved using the superposition of previously studied 2D configurations. The electrical body force is then obtained and shown that gives rise to a liquid whirl. This system can be of interest in the design of rotatory micro-pumps and for local mixing in microfluidics. Finally, both 2D and 3D analytical self-similar solutions are compared with numerical and experimental results.

In our experiments, we have used simple electrode designs consisting of two or four co-planar electrodes mounted on a glass substrate. Aqueous solutions of KCl of different conductivities were placed on top of the electrodes. The fluid motion was observed using fluorescent latex particles as tracers.

ELECTROMECHANICAL EQUATIONS

Electrical and mechanical equations need to be solved simultaneously, since the electrical forces move the liquid, and this motion produces a charge density current that must be included in the electrical equations. Nevertheless, the low velocities involved [2] allow us to neglect this convection current compared with the ohmic current. In this case, the electrical problem can be solved independently and its solution inserted into the mechanical problem.

Electrical equations

The low frequencies involved and the insignificance of magnetic effects reduce the electromagnetic equations to the quasi-electrostatic limit [5]. In addition, the convection current can be neglected compared with the ohmic current for saline solutions. Assuming that the conductivity σ and the permittivity ε of the liquid are independent on time, the equations that govern the complex amplitude of the ac fields of frequency ω are

$$\nabla \times \mathbf{E} = 0 \qquad \nabla \cdot ((\sigma + i\omega\varepsilon)\mathbf{E}) = 0 \tag{1}$$

The electric field can be written as the gradient of an electric potential. The boundary conditions for this potential are given by the voltage applied to the electrodes, $V_s(x,y)$ at the plane $z=0$.

Mechanical equations

The time-average value of the electrical volume force density acting on the fluid is given by [2]

$$\langle \mathbf{f} \rangle = \frac{1}{2}\mathrm{Re}\left(\rho\,\mathbf{E}^*\right) - \frac{1}{4}\mathbf{E}\cdot\mathbf{E}^*\nabla\varepsilon \tag{2}$$

where $*$ indicates complex conjugate and Re$(\ldots)$ the real part of $(\ldots)$. The electrical force also has an oscillating component of frequency 2ω. Since the observed fluid flow is steady, the analysis is focused on the time-average electrical force.

The time-average velocity field is governed by the Navier-Stokes equations, in the low Reynolds number regime

$$\nabla \cdot \mathbf{v} = 0 \qquad -\nabla p + \eta\nabla^2\mathbf{v} + \langle \mathbf{f} \rangle = 0 \tag{3}$$

The appropriate boundary conditions for the liquid velocity at the electrode plane are the no-slip and impenetrability conditions at a rigid boundary, $\mathbf{v} = 0$.

Weak temperature gradient

We assume that the permittivity and conductivity changes with temperature are small. In this way, we can expand ε and σ around a reference temperature as

$$\varepsilon = \varepsilon(1 + \alpha(T - T_0)) \qquad \sigma = \sigma(1 + \beta(T - T_0)) \tag{4}$$

where in the right hand side, σ, ε, α and β (the quantities and their relative derivatives at $T = T_0$) can be taken as constants. For aqueous solutions at $T_0 = 20°$C, $\alpha = (\partial\varepsilon/\partial T)/\varepsilon \approx -0.004\,\mathrm{K}^{-1}$ and $\beta = (\partial\sigma/\partial T)/\sigma \approx 0.02\,\mathrm{K}^{-1}$.

In the same approximation, the electric field can be written as $\mathbf{E} = \mathbf{E}_0 + \mathbf{E}_1$ with $|\mathbf{E}_1| \ll |\mathbf{E}_0|$. Substituting in the equations for the field, we have, at the lowest order,

$$\nabla \cdot \mathbf{E}_0 = 0 \qquad \nabla \times \mathbf{E}_0 = 0 \qquad \rho_0 = 0 \tag{5}$$

so that the electric potential at this order satisfies Laplace's equation, $\nabla^2\phi = 0$, with boundary conditions given by the applied voltage, V_s, at the plane $z = 0$.

Volume force

The first non-vanishing term in the expansion of the force requires the first order charge density ρ_1, whose complex amplitude can be obtained from the zero order field as

$$\rho_1 = \frac{\varepsilon(\alpha - \beta)}{1 + i\omega\tau}(\nabla T \cdot \mathbf{E}_0), \qquad \tau = \frac{\varepsilon}{\sigma} \tag{6}$$

Substituting into the time-average volume force (2), we obtain

$$\langle \mathbf{f} \rangle = \frac{1}{2}\mathrm{Re}\left(\frac{\varepsilon(\alpha - \beta)}{1 + i\omega\tau}(\nabla T \cdot \mathbf{E}_0)\mathbf{E}_0^*\right) - \frac{\varepsilon}{4}\alpha\,\mathbf{E}_0 \cdot \mathbf{E}_0^*\,\nabla T \tag{7}$$

where the first and second terms are the Coulomb and dielectric forces, respectively.

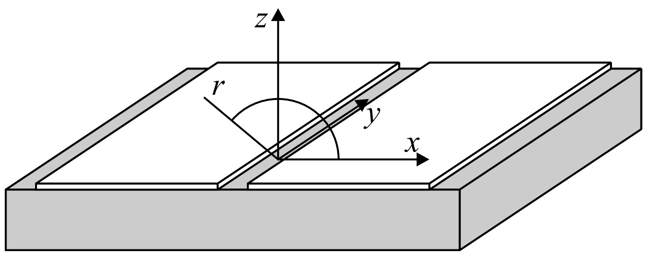

Figure 1. Schematic diagram of the two co-planar electrodes

TWO-DIMENSIONAL FLOWS

The first case we model is composed of two co-planar electrodes separated by a small gap (see Fig. 1), subjected to an ac potential difference of amplitude V_0 and frequency ω.

Approximating the electrodes as half-planes separated by an infinitesimal gap, the boundary conditions for the potential at $z = 0$ are

$$\phi(z=0) = V_s = \frac{V_0}{2}\,\mathrm{sgn}\,(x) \qquad \mathrm{sgn}\,(x) = \begin{cases} +1 & x>0 \\ -1 & x<0 \end{cases} \tag{8}$$

This problem can have a self-similar solution, independent of r, given in polar coordinates by.

$$\phi(r,\varphi) = \frac{V_0}{\pi}\left(\frac{\pi}{2}-\varphi\right) \qquad \mathbf{E}_0(r,\varphi) = \frac{V_0}{\pi r}\mathbf{u}_\varphi \tag{9}$$

The field lines describe half circles from one electrode to the other.

Vertical temperature gradient

Let us assume an imposed, uniform, vertical gradient of temperature, that can be written as $\nabla T = T'\mathbf{u}_z$. Substituting this expression in (7) we obtain the first order electrical body force

$$\langle \mathbf{f} \rangle = \frac{\varepsilon}{2}\left(\frac{V_0}{\pi r}\right)^2 T' \left[\left(\frac{\alpha-\beta}{1+(\omega\tau)^2} - \frac{\alpha}{2}\right)\cos\varphi\,\mathbf{u}_\varphi - \frac{\alpha}{2}\sin\varphi\,\mathbf{u}_r\right] \tag{10}$$

The r^{-2} radial dependence of $\langle \mathbf{f} \rangle$ permits us to look for self-similar solutions for the pressure and liquid velocity. The required functional forms are

$$p(r,\varphi) = \frac{P(\varphi)}{r} \qquad \mathbf{v} = v_r(\varphi)\mathbf{u}_r + v_\varphi(\varphi)\mathbf{u}_\varphi \tag{11}$$

The incompressibility equation provides a relation between the radial and azimuthal components

$$v_r = -\frac{dv_\varphi}{d\varphi} \tag{12}$$

Substituting these expressions in the Navier-Stokes equation we get a fourth-order equation for the azimuthal velocity

$$\frac{d^4 v_\varphi}{d\varphi^4} + 2\frac{d^2 v_\varphi}{d\varphi^2} + v_\varphi = -C\cos\varphi \tag{13}$$

where

$$C = \frac{\varepsilon V_0^2 T'}{2\pi^2\eta}\,\frac{\alpha(\omega\tau)^2+\beta}{1+(\omega\tau)^2} \tag{14}$$

Since $\alpha < 0$ and $\beta > 0$, the constant C changes sign at a characteristic frequency given by

$$\omega_c = \frac{1}{\tau}\sqrt{\frac{\beta}{-\alpha}} \tag{15}$$

For this particular solution, the fluid flow changes direction at this frequency. In the general case, there is a transition of fluid flow behaviour around ω_c, corresponding to fluid flow arrising from two different forces: the Coulomb term (for $\omega \ll \omega_c$) and the dielectric term (for $\omega \gg \omega_c$).

The solution for v_φ is given by

$$v_\varphi = -\frac{C}{8}\left((\pi\varphi-\varphi^2)\cos\varphi + (2\varphi-\pi)\sin\varphi\right) \tag{16}$$

And from this, we have the radial component

$$v_r = -\frac{dv_\varphi}{d\varphi} = \frac{C}{8}(2-\pi\varphi+\varphi^2)\sin\varphi \tag{17}$$

In this self-similar solution the velocity components do not depend on the distance from the origin r. Figure 2 shows a contour plot of a stream-function, defined by the relations

$$v_r = -\frac{1}{r}\frac{\partial\psi}{\partial\varphi} \qquad v_\varphi = \frac{\partial\psi}{\partial r} \tag{18}$$

The arrows indicate the direction of the fluid flow when the temperature decreases with height and the frequency is smaller than ω_c as defined by eq. (15). For $\omega > \omega_c$ the flow changes direction. The pattern of stream-lines is very close to that given in [4] for a similar numerical problem in the neighbourhood of $r = 0$ where the self-similar solution is expected to be valid.

The maximum absolute value of v_r is for $\varphi = \pi/2$, where $v_r = -0.0584251C$. The maximum absolute value of v_φ is obtained for the angles $\varphi_1 = 0.887129$ and $\varphi_2 = \pi - \varphi_1$ where

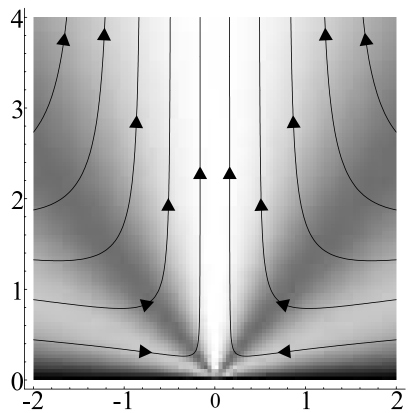

Figure 2. Streamlines and liquid speed for a vertically imposed temperature gradient. The gray levels indicate the magnitude of the liquid velocity, from zero (black) to maximum velocity (white)

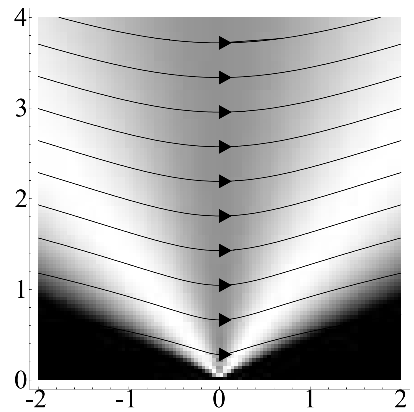

Figure 3. Streamlines and liquid speed for a horizontally imposed temperature gradient. The gray levels indicate the magnitude of the liquid velocity, from zero (black) to maximum velocity (white)

$v_\varphi(\varphi_1) = 0.025405C$ and $v_\varphi(\varphi_2) = -0.025405C$. This values of maximum velocities compare well with the numerical results presented in [4]. Using the values specified in this reference, i.e. $T' = 0.021\,\mathrm{K}\mu\mathrm{m}^{-1}$, $V_0 = 10$ V, a maximum radial velocity of 87.0 $\mu\mathrm{ms}^{-1}$ is obtained from the analytical solution, as compared to 80 $\mu\mathrm{ms}^{-1}$ from the numerical work.

Horizontal temperature gradient

Now let us suppose now an imposed horizontal gradient of temperature in the x direction, $\nabla T = T'\mathbf{u}_x$.

Assuming again that the permittivity and conductivity increments with temperature are small enough, we can write the electrical body force as:

$$\langle \mathbf{f} \rangle = -\frac{\varepsilon}{2}\left(\frac{V_0}{\pi r}\right)^2 T' \left[\left(\frac{\alpha-\beta}{1+(\omega\tau)^2} - \frac{\alpha}{2}\right)\sin\varphi\,\mathbf{u}_\varphi + \frac{\alpha}{2}\cos\varphi\,\mathbf{u}_r\right] \quad (19)$$

Since the body force varies again as r^{-2}, we can have self-similar solutions as in the previous section. The resulting equation for the azimuthal velocity is now

$$\frac{d^4 v_\varphi}{d\varphi^4} + 2\frac{d^2 v_\varphi}{d\varphi^2} + v_\varphi = -C\sin\varphi \quad (20)$$

where C is given by eq. (14).

The solution for φ that satisfies the boundary conditions for zero velocity at $\varphi = 0$ and π is

$$v_\varphi = -\frac{C}{8}(\pi\varphi - \varphi^2)\sin\varphi \quad (21)$$

The expression for the radial component is now

$$v_r = \frac{C}{8}\left((\pi - 2\varphi)\sin\varphi + (\pi\varphi - \varphi^2)\cos\varphi\right) \quad (22)$$

Figure 3 shows a contour plot of the stream-function in this situation. The arrows indicate the direction of the fluid flow when the temperature decreases with height and $\omega < \omega_c$, with ω_c given by eq. (15). For $\omega > \omega_c$ the flow changes direction.

The maximum absolute value of v_φ is for $\varphi = \pi/2$, where $v_\varphi = 0.308425C$. The maximum absolute value of v_r is for the angles $\varphi_1 = 0.729827$ and $\varphi_2 = \pi - \varphi_1$ where $v_r(\varphi_1) = 0.304156C$ and $v_r(\varphi_2) = -0.304156C$.

THREE-DIMENSIONAL FLUID ROTATION

Extending the analysis to three dimensions, we consider a system of four electrodes placed in each quadrant, with small gaps in-between (see Fig. 4). An ac voltage is applied to each plate, with phase shifted 90° between consecutive electrodes, generating a rotating electric field.

Basic electric field

In this system, we consider the x and y axes to lie along the gaps between the four electrodes and the z axis normal to the electrodes plane.

In the lowest order, the electric potential verifies Laplace's equation with the boundary condition at the plane $z = 0$

$$\phi(z=0) = V_s = \begin{cases} V_0 & x>0, y>0 \\ V_0 i & x<0, y>0 \\ -V_0 & x<0, y<0 \\ -V_0 i & x>0, y<0 \end{cases} \quad (23)$$

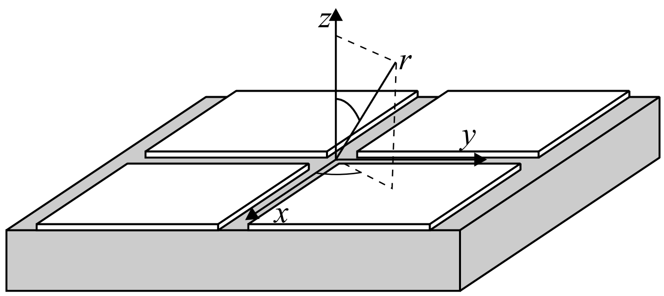

Figure 4. Schematic diagram of the four co-planar electrodes

The surface voltage can be written in a simpler form as

$$V_s = \frac{V_0(1-i)}{2}\operatorname{sgn}(x) + \frac{V_0(1+i)}{2}\operatorname{sgn}(y) \tag{24}$$

In this way, we can write the electric potential in the bulk as a superposition of two 2D solutions, as described in the previous sections. The solution for the potential is

$$\phi = \frac{V_0(1-i)}{\pi}\arctan\left(\frac{x}{z}\right) + \frac{V_0(1+i)}{\pi}\arctan\left(\frac{y}{z}\right) \tag{25}$$

The resulting complex amplitude for the electric field can also be expressed as a superposition

$$\mathbf{E}_0 = \frac{V_0(1-i)}{\pi}\left(\frac{-z\mathbf{u}_x + x\mathbf{u}_z}{x^2+z^2}\right) + \frac{V_0(1+i)}{\pi}\left(\frac{-z\mathbf{u}_y + y\mathbf{u}_z}{y^2+z^2}\right) \tag{26}$$

The direction of the electric field (in the time domain) rotates, although not uniformly, everywhere.

This field can be written equivalently as $\mathbf{E}_0 = (1-i)\mathbf{e}_a + (1+i)\mathbf{e}_b$, with $\mathbf{e}_a$ and $\mathbf{e}_b$ purely real.

Vertical temperature gradient

We now consider the combined action of the previous electric field with an imposed, uniform, vertical gradient, $\nabla T = T'\,\mathbf{u}_z$.

The volume force, eq. (7), has a Coulomb term and a dielectric one. Both can be decomposed into four terms, according to the components, $\mathbf{e}_a$ and $\mathbf{e}_b$, of the electric field involved

$$\mathbf{f} = \mathbf{f}_{aa} + \mathbf{f}_{ab} + \mathbf{f}_{ba} + \mathbf{f}_{bb} \tag{27}$$

The first and last terms produce purely two-dimensional motions, although at perpendicular planes, as described in the previous section. In what follows, we will be mostly concerned with the terms $\mathbf{f}_{ab}$ and $\mathbf{f}_{ba}$, which are responsible for the rotatory motion.

The total fluid flow is a superposition of the flow driven by $\mathbf{f}_{ab}$ and $\mathbf{f}_{ba}$, plus the two-dimensional motions that have components in the xz plane (due to $\mathbf{f}_{aa}$) and the yz plane (due to $\mathbf{f}_{bb}$). The resulting stream-lines can be quite complicated for the total motion.

For the dielectric force, the cross terms cancel each other. The only new terms appear in the Coulomb force.

$$\mathbf{f}_{ab} + \mathbf{f}_{ba} = \frac{\varepsilon(\alpha-\beta)\omega\tau}{(1+\omega^2\tau^2)}\left((\nabla T\cdot\mathbf{e}_a)\mathbf{e}_b - (\nabla T\cdot\mathbf{e}_b)\mathbf{e}_a\right) \tag{28}$$

Before calculating the spatial dependence of the coupled volume force density, we can establish that the dependence on the frequency is given by the function

$$g(\omega\tau) = \frac{\omega\tau}{1+\omega^2\tau^2} \tag{29}$$

This implies that $\mathbf{f}_{ab} + \mathbf{f}_{ba}$ tends to zero for low and high frequencies, with a maximum at the charge relaxation frequency $\omega = 1/\tau = \sigma/\varepsilon$. Unlike the previous 2D fluid flows, this 3D flow does not change direction with frequency.

The spatial dependence of the coupled force density is

$$\mathbf{f}_c = \frac{Az(-y\mathbf{u}_x + x\mathbf{u}_y)}{(x^2+z^2)(y^2+z^2)} \qquad A = -\frac{V_0^2 T'(\alpha-\beta)\varepsilon\omega\tau}{\pi^2(1+\omega^2\tau^2)} \tag{30}$$

or, using spherical coordinates,

$$\mathbf{f}_c = \frac{A\cos\theta\sin\theta\mathbf{u}_\varphi}{r^2(\cos^2\theta + \sin^2\theta\cos^2\varphi)(\cos^2\theta + \sin^2\theta\sin^2\varphi)} \tag{31}$$

This force is directed along the azimuthal direction, causing rotation.

The dependence on the angular coordinate φ is periodic with a period of length $\pi/2$, repeating each quadrant. This can be written as a Fourier series. Only the zero order term produces net rotation. The component of this mode is obtained by averaging over a period

$$\langle f_\varphi \rangle = \frac{1}{2\pi}\int_0^{2\pi} f_\varphi\, d\varphi = \frac{2A\sin\theta}{r^2(1+\cos^2\theta)} \tag{32}$$

To calculate the resulting velocity we have to solve the Navier-Stokes equations. Multiplying by $\mathbf{u}_\varphi$ and averaging, we obtain an equation for the mean azimuthal velocity

$$\frac{1}{r}\frac{\partial^2}{\partial r^2}\left(r\langle v_\varphi\rangle\right) + \frac{1}{r^2}\frac{\partial}{\partial\theta}\left(\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\,\langle v_\varphi\rangle\right)\right) = -\frac{\langle f_\varphi\rangle}{\eta} \tag{33}$$

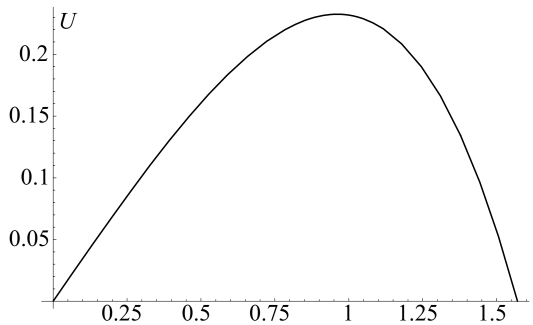

Figure 5. Dependence of the function U with the polar angle θ.

As in the previous sections, the r^{-2} dependence of the force dictates the functional form for the self-similar velocity solution. We assume a functional form independent of r as $\langle v_\varphi \rangle = U(\theta)A/\eta$, which reduces the problem to an ordinary differential equation. The solution of this equation, which gives a vanishing velocity at $\theta = 0$ and at $\theta = \pi/2$ is

$$U = \cot\theta \left(\frac{\pi}{2} - 2\arctan(\cos\theta) + \ln(2) \right) - \frac{\ln(1+\cos^2\theta)}{\sin\theta} \quad (34)$$

This function is always positive, and goes from zero at the axis to zero at the electrodes reaching the maximum speed for $\theta = 0.961$ with a value $U_{\max} = 0.232$ (see Fig. 5).

COMPARISON WITH EXPERIMENTS

Observations of fluid flows under conditions similar to the 2D problems previously analyzed have been reported in [3]. In these experiments, a simple electrode design consisting of two co-planar plates (2mm long, 0.5mm wide) with parallel edges separated by 25μm was used. The aqueous solutions were placed on top of the electrodes. Fluid flow was observed when the electrodes were subjected to voltages around 10 V and frequencies around 1 MHz under strong illumination. The observed fluid flow pattern agrees well with the prediction from the electrothermal theory if there is a vertical gradient of temperature, generated by the light [4]. The experimental fluid flow changed direction at a certain frequency and the fluid velocity amplitude varied with frequency as predicted. The 2D analytical solutions of the present work compare well with the numerical and experimental results.

Preliminary experiments demonstrate that the liquid rotates when subjected to a rotating electric field under strong illumination. The aqueous solutions were placed on top of four hyperbolic shaped planar electrodes mounted on glass. The rotating field was created by applying a four-phase signal to the electrodes. The voltage on consecutive electrodes is phase shifted by 90°. Figure 6 is a superimposition of several images showing particle tracks at a plane 40 μm up from the plane of the electrodes. The particle tracks show that the liquid is rotating. Maximum velocities around 100 μm/s were recorded at an applied voltage of 10 V and a frequency of 1 MHz. The rotatory fluid motion is predicted by the electrothermal theory if there is a vertical gradient of temperature. The observed fluid velocity was proportional to the light intensity, an indication that the temperature gradient in the liquid is induced by the light. As predicted by the 3D analytical solution, the rotating fluid flow was significant over a certain frequency range and did not change direction with frequency. Furthermore, the expected velocity values are in accordance with observations if they are calculated for a temperature gradient of 0.021 K μm^{-1}. This was the estimated temperature gradient induced by light in previous experiments [3, 4].

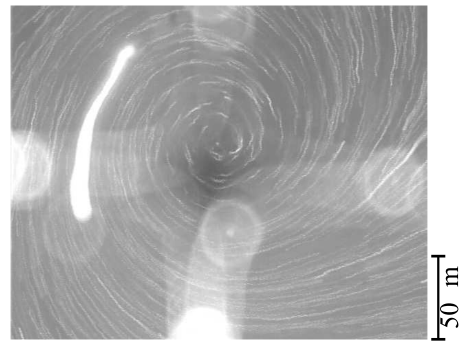

Figure 6. Top view of a rotating aqueous solution placed on top of four coplanar electrodes. Image of particle tracks obtained by superimposing successive video frames

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