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## Frequency dependent rotation and translation of nanowires in liquid environment

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In this paper, an approach of aligning and handling silicon nanowires in liquid environment on the large scale is presented. Traveling dielectrophoresis was used to simultaneously pump a weakly ionic nanowire suspension and to rotate nanowires in a plane perpendicular to the electrodes. The pumping force on the solution was maximized by monitoring the cell impedance using impedance spectroscopy and by matching the frequency of the supply voltage to the impedance crossover. At frequencies above or below impedance crossover, trapping or rotation of nanowires was observed which is explained by means of a competition between stationary and drag forces. © 2010 American Institute of Physics. [doi:10.1063/1.3430738]

In recent years, microscale liquid and particle handling techniques suitable for assembly of randomly mass produced nanostructures and for lab-on-chip devices have attracted increased interest.<sup>1,2</sup> Dielectrophoresis (DEP) has been suggested as a means of particle handling by Pohl<sup>3</sup> in 1978. It is based on the complex permittivity difference between an object and a medium. By applying a multiphase signal to an electrode array, rotating and propagating fields can be generated, causing rotation and translation along the wave propagation vector rather than along a static gradient. The idea was exploited by Hagedorn<sup>4,5</sup> who linearized the electro-rotation effect and used it in an electrohydrodynamic pump to move liquid in a channel. Electrotation in early experiments was used to determine the Clausius–Mossotti factor of an object from its rotation speed<sup>6</sup> or to study bacteria behavior.<sup>7</sup> During past years this technique was also applied to metallic nanowires which can serve as polarization filters, microfluidic valves, or stirring devices.<sup>8</sup> Rotation frequencies upto  $f_{\text{rot}} = 1800$  rpm have been reached in the work of Fan *et al.*<sup>9</sup> for single nanowires and the synchronous rotation of hundreds of silicon nanowires (SiNWs) has been demonstrated by the Mayer group.<sup>8</sup> The aim of this work is the translation and rotation of SiNWs to control spacing and alignment. A DEP liquid pumping method based on resistive-capacitive (RC) dispersion between the insulating layer covering an electrode array and the bulk resistivity of the liquid was demonstrated previously.<sup>10</sup> Here, the recently developed pumping scheme is applied to a suspension of nanowires, and the viscous drag force on the nanowires in the moving liquid is set into competition with trapping forces in the static geometric field gradient above the electrode array. When the propagating field is applied, a pumping force applies to the liquid, whereas the direct electrostatic effect on the particles is dominated by trapping forces in the static field gradient and eventually a torque if a permanent or induced dipole moment is present. Experimentally, it has been observed that depending on the frequency, the nanowires are trapped between the electrodes, move together with the liq-

uid or rotate in a plane perpendicular to the electrode array. In previous works, frequency dependence of stationary forces was exploited,<sup>11</sup> whereas in this work, a competition is established between propagating and stationary forces. The resulting nanowire rotation occurs in the vertical plane, in contrast to electrorotation which is in the electrode plane.<sup>8,9</sup>

The design of the DEP chip involves four levels of photolithography. The chip intersection is shown in Fig. 1(a) and its layout in Fig. 1(b). On the  $\langle 100 \rangle$  n-type Si wafer a layer of SiO<sub>2</sub> was deposited, in which electrodes were buried. Next, the electrodes were covered with Si<sub>3</sub>N<sub>4</sub> and via holes were etched to provide connections to the subsequently deposited bus-lines. The bus-lines were covered with one more Si<sub>3</sub>N<sub>4</sub> layer, in order to prevent contact of the bus lines with the liquid. The interdigitated electrodes were provided with a four phase voltage to create the traveling field. The spacing and electrode width were 6  $\mu\text{m}$ , repeated 25 times, forming a 1.2 mm long channel. Channel sidewalls were made with Su-8 resist (not shown).

Slightly conductive liquid was prepared by adding 0.5  $\mu\text{l}$  of NaCl 0.9% solution (Gilbert, France) to 2 ml of

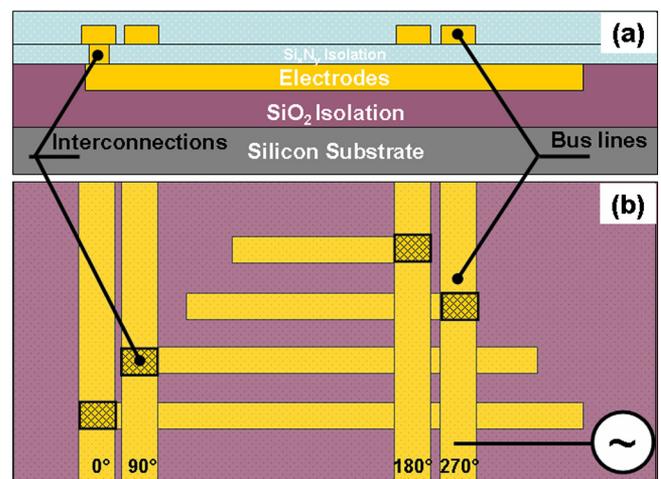


FIG. 1. (Color online) DEP chip design. Side view (a) and top view (b).

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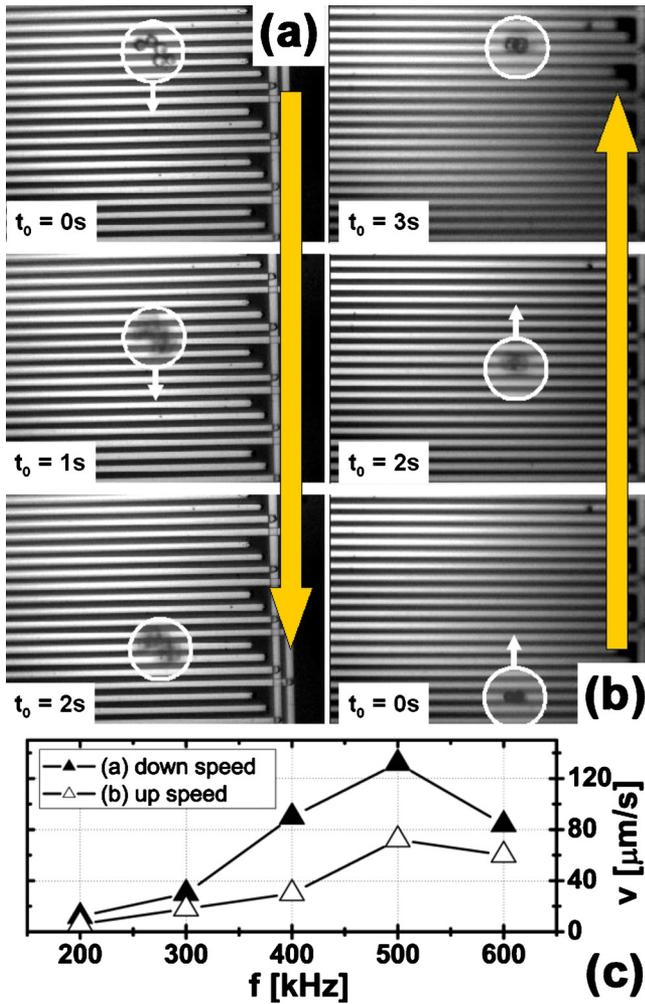


FIG. 2. (Color online) Liquid pumping. Bead ( $10 \mu\text{m}$  in diameter) transport with the liquid is visible in two directions—down (a) (left column) and after phase inversion—up (b) (right column). The pumping speed (c) is frequency dependent and its maximum is  $v=120 \mu\text{m/s}$  at  $f=500 \text{ kHz}$ .

deionized water. The resulting liquid conductivity was estimated to  $\sigma=0.001 \text{ S/m}$ . To visualize liquid pumping in the DEP device, fluorescent latex beads (Fluoresbrite™; diameter:  $10 \mu\text{m}$ ) are introduced in half of the solution. The other half is used to create a nanowire suspension. Nanowires were added together with  $4 \mu\text{l}$  of ethanol to prevent water repulsion from the hydrophobic nanowires.

SiNWs have been prepared by chemical vapor deposition via the vapor-liquid-solid (VLS) mechanism.<sup>12</sup> A Si  $\langle 111 \rangle$  wafer coated with a  $5 \text{ nm}$  layer of Au was used as a substrate to grow SiNWs. It was heated to  $500\text{--}600 \text{ }^\circ\text{C}$ , until the Au layer formed liquid alloy nanodroplets with the underlying Si substrate. A gas mixture of  $\text{SiH}_4$  diluted in  $10\% \text{ H}_2$  in Ar under gas flow dynamics was then introduced. The silicon preferentially dissolves in the Au–Si alloy nanodroplets. Once the liquid droplet is supersaturated with Si, precipitation of the solid SiNWs occurs. The vast majority of SiNWs grow out of the liquid droplet perfectly orthogonal to the substrate [inset of Fig. 3(d)]. For the DEP experiments, large diameter SiNWs ( $150\text{--}300 \text{ nm}$ ) have been selected (length  $5\text{--}10 \mu\text{m}$ ). This enables their real time observation by optical microscopy under changing DEP conditions.

The channel was filled with the liquid containing the beads and the frequency of maximum pumping force was

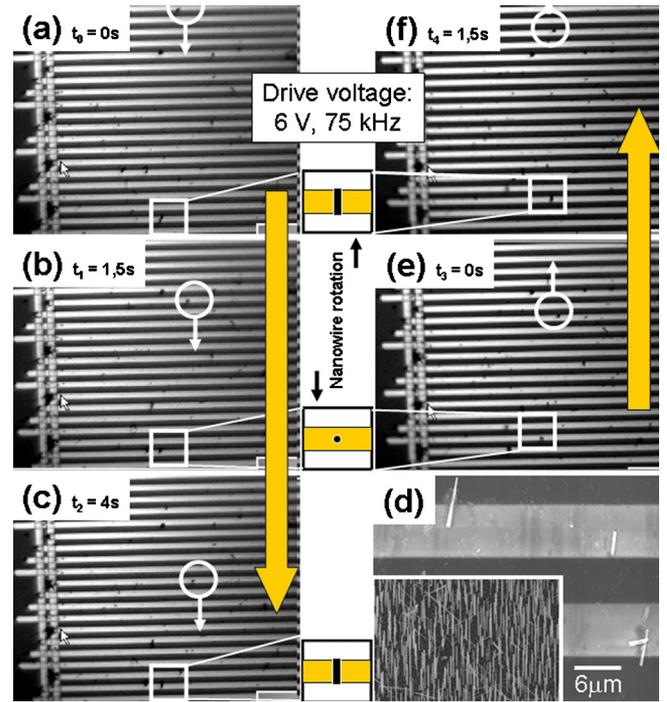


FIG. 3. (Color online) Nanowire propagation (circles) and rotation perpendicular to the electrodes (squares). Images (a)–(c) show a movie sequence with downward liquid motion; to prove the reproducibility of pumping, (e) and (f) show upward flow after inversion of the propagation direction. The maximum frequency of nanowire rotation was  $f_{\text{rot}}=200 \text{ rpm}$ . The SEM image (d) shows that SiNWs conserved their alignment after liquid evaporation. The inset presents a SEM image of the nanowires after VLS growth.

determined as described previously:<sup>10</sup> the voltage applied to a unit cell is divided between the liquid and the electrode insulator represented by a resistance  $R_{\text{liq}}$  and a capacitance  $C_{\text{ins}}$ , respectively. The polarization of the liquid  $P_{\text{liq}}$  is proportional to the fraction of the voltage drop across the liquid

$$P_{\text{liq}} \propto V_{\text{appl}} \exp(i\omega t) \frac{1}{1 + \frac{1}{i\omega R_{\text{liq}} C_{\text{ins}}}}, \quad (1)$$

where  $\omega=2\pi f$  is the field angular frequency. The maximum out-of-phase polarization of the liquid and consequently the highest pumping force occurs at  $f_{\text{max}}=1/(2\pi R_{\text{liq}} C_{\text{ins}})$ . This frequency coincides with the crossover frequency of the impedance of the RC series circuit and is found experimentally by impedance spectroscopy. At this frequency liquid pumping was observed, shown by the bead motion in the image sequence of Fig. 2(a). To exclude other causes of liquid motion, the pumping was reproduced in the opposite direction [Fig. 2(b)]. For both directions, speed versus frequency was registered in Fig. 2(c), showing a maximum at  $500 \text{ kHz}$  in good agreement with the predictions.

With these conditions the traveling wave dielectrophoresis (twDEP) propagation of SiNW suspensions was investigated. While applying  $f=545 \text{ kHz}$ , nanowires appeared to align between electrodes and to stick to the surface. When the frequency was reduced to  $f=200 \text{ kHz}$  translation of the nanowires along the channel was observed. At  $f=75 \text{ kHz}$ , nanowire translation and electrorotation has occurred. The movie sequence in Fig. 3 shows this behavior at  $75 \text{ kHz}$ : the nanowire surrounded by circles move in one direction [Figs. 3(a)–3(c)]. Translation in the opposite direction is observed

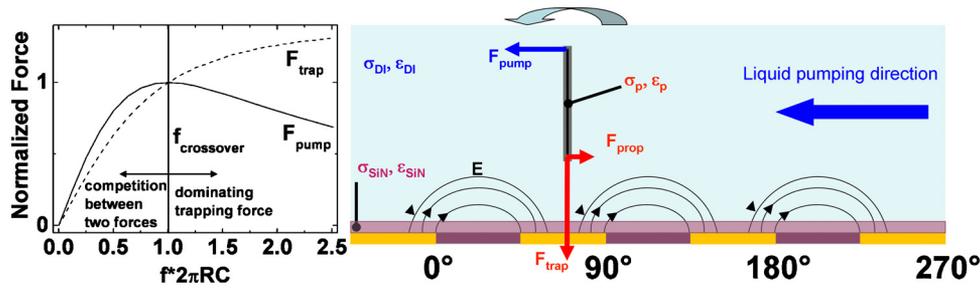


FIG. 4. (Color online) Competition between drag forces  $F_{\text{drag}}$  and trapping force  $F_{\text{trap}}$ . Forces are normalized to unity at the frequency of impedance crossover.

in Figs. 3(e) and 3(f) upon phase inversion. Figure 3 also shows two stages of electrorotation: the nanowire surrounded by a square is parallel to the surface in (a) and (c) and perpendicular to it in (b). The maximum observed rotation frequency was  $f_{\text{rot}}=200$  rpm. After applying the field until evaporation of the suspension, the nanowires stuck to the surface with a preferential alignment in the field propagation direction, see Fig. 3(d).

The observed translation and trapping of nanowires is explained by two simultaneous twDEP forces: (1) the pumping force  $F_{\text{pump}}$  on the slightly conductive liquid in the field gradient due to the propagation wave vector ( $i\vec{k}$ ); this is the only force experienced by the liquid since the trapping forces cancel within a unit cell; the liquid flow exerts a viscous drag force on the nanowires, due to their small Reynolds number<sup>9</sup> of  $10^{-15}$ ; (2) the trapping forces onto the nanowires: it is considered that the field gradient near the electrode edges is higher than the one due to  $\vec{k}$  and once the nanowire is between electrodes,  $F_{\text{trap}} \gg F_{\text{prop}}$  so that the direct propagating force on the nanowire can be neglected. Competition occurs therefore between the viscous drag force proportional to the liquid velocity and the direct trap force on the nanowires. The former is proportional to the imaginary part of Eq. (1),  $F_{\text{pump}} \propto \text{Im}[P_{\text{liq}}]$ . The latter is caused by the static field gradient due to the electrode geometry and is expected to depend only on the field strength in the liquid as  $F_{\text{trap}} \propto |P_{\text{liq}}|^2$ . The qualitative frequency dependence of both forces is shown in Fig. 4 (graph). Each curve is normalized to one at  $f_{\text{crossover}}$ , but different coefficients certainly do apply to  $F_{\text{pump}}$  and  $F_{\text{trap}}$ , shifting the crossing of the forces away from the impedance crossover ( $f_{\text{crossover}}$ ). At frequencies lower than the crossing, the viscous drag forces due to liquid pumping dominate. At frequencies above the crossing, the trapping force onto the nanowire dominates. Apparently in the present case, the coefficients of trapping and drag forces are such that the crossing occurs at 200 kHz. Trapping is always accompanied by alignment parallel to the field because the polarization along the long axis of a needlelike object is higher than perpendicular to it for geometrical reasons.

At low frequencies ( $f < 100$  kHz) the forces approach but it is not expected from Fig. 4 that trapping becomes dominant again. The explanation of the observed behavior, trapping and electrorotation at low frequency, remains specu-

lative. One possibility is that a dispersive effect does appear at low frequencies, e.g., the response of the Helmholtz layer around the SiNW might increase its polarizability and make the trapping forces dominant again. Concerning electrorotation it can be explained by a permanent dipole moment remaining on the nanowire. This can cause the nanowire to flip in the alternating field, favorably after one of its ends is lifted by the drag forces. The modeling of the frequency dependent dielectric behavior of the nanowire itself falls however beyond the scope of this paper.

We have presented a nanowire handling method based on the competition between viscous drag force and electrostatic trapping force in a previously developed twDEP micropump. Nanowire trapping, translation, or out-of-plane rotation was observed depending on the applied frequency. A qualitative explanation was derived from the proportionality of the trapping force to the modulus and the pumping force on the out-of-phase component of the liquid polarization, respectively. The switching between translation and trapping is sufficiently explained by the bulk properties. The interpretation of the electrorotation would require a deeper understanding of the frequency-dependent dielectric behavior of the nanowire and its surroundings on the microscopic scale.

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