Towards Single Molecule Manipulation with Dielectrophoresis Using Nanoelectrodes

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Abstract—We present measurements of the scaling of the dielectrophoresis force with electrode size and particle size, in order to determine the ultimate size limits for dielectrophoresis at the nanoscale. To demonstrate the feasibility of nano-manipulation we present studies on the dielectrophoretic manipulation of DNA with microfabricated electrodes using RF electric fields. We find that DNA undergoes positive dielectrophoresis (i.e. the DNA is attracted to high electric field regions) at frequencies between 500 kHz and 1 MHz. We present a nano-manipulation array platform which demonstrates the scalability of these concepts for massively parallel, economic manipulation at the molecular scale. Finally, based on these measurements, we present a concept for a nanomotor using nanotube electrodes. Applications in molecular electronics and molecular manufacturing are discussed.

I. INTRODUCTION

The electronic control of the position of individual molecules in aqueous solution is the goal of this work. Electronic rather than optical tweezers could allow for economical and massively parallel, heterogeneous interrogation and control of biological and electronic functions on one chip. A very promising technique to achieve this goal is to use dielectrophoresis forces generated by micro and nanofabricated electrodes.

Dielectrophoresis (hereafter DEP) is the electronic analog of optical tweezers[1], [2], using audio frequency, RF, and microwave electric fields generated from microfabricated electrodes on a chip. An ac electric field induces a dipole moment which, in the presence of a field gradient, experiences a force towards either the high-field intensity region (positive dielectrophoresis) or the low-field intensity region (negative dielectrophoresis). An advantage is that it can be scaled for massively parallel electronic manipulation of nanosized objects such as DNA, proteins, nanotubes, nanoparticles, and possibly even single molecules in aqueous solutions. (In contrast to conventional electrophoresis, dielectrophoresis functions with both neutral and charged particles.) Molecule by molecule assembly of matter is in principle possible. As with optical tweezers, for dielectrophoresis to be of use it must dominate the thermal Brownian motion. Thus scaling of dielectrophoresis and Brownian motion with particle size and electrode geometry is especially crucial if the technology is to be applied at the nanometer scale.





Fig. 1. The figure shows how the minimum size of trapped particles (latex beads in this experiment) varies with the applied bias. Solid squares and asterisks represent our measurements for 10 μ m and 100 μ m gap electrode geometries, respectively. Solid lines are our phenomological prediction.

II. SCALING LAWS

A. Scaling with particle size

It can be shown that the force acting on a spherical particle (the *dielectrophoresis* force) is given by[1], [3]

$$\vec{F}_{DEP} = 2\pi v \epsilon_m \alpha_r \vec{\nabla} (\vec{E}_{RMS}^2), \qquad (1)$$

where v is the volume of the particle, \vec{E}_{RMS} the RMS value of the electric field (assuming a sinusoidal time dependence), and α_r the real part of what is called the *Clausius-Mosotti* factor, which is related to the particle dielectric constant ϵ_p and medium dielectric constant ϵ_m by

$$\alpha_r \equiv Re\left(\frac{\epsilon_p^* - \epsilon_m^*}{\epsilon_p^* + 2\epsilon_m^*}\right). \tag{2}$$

Here the star (*) denotes that the dielectric constant is a complex quantity. For this force to be effective it must overcome the Brownian motion, which can be treated as a random force whose maximum value is given roughly by[1]:

$$F_{thermal} \approx k_B T / 2r_{particle},$$
 (3)



Fig. 2. Latex beads "trapped" between four electrodes under the influence of negative dielectrophoresis.

where k_B is the Boltzmann constant, T the temperature, and $r_{particle}$ the particle radius. Thus, the ratio of the DEP force to the thermal force is proportional to the fourth power of the particle radius.

B. Scaling with electrode size

It can be shown that, for a hemispherical electrode with radius of curvature r_1 , the gradient in the electric field squared is given by:

$$\vec{\nabla}(\vec{E}_{RMS}^2) \approx -\frac{2r_1^2 V^2}{r^5} \hat{r}.$$
 (4)

where \hat{r} is a unit vector in the radial direction, r the distance from the origin, and V the applied voltage.

If one assumes that the particle is as close to the electrode as its radius then it is straightforward based on the above equations to show that the minimum voltage required for the DEP force to exceed the thermal force scales as the inverse of the electrode radius, and the 1.5th power of the particle size, i.e.

$$V_{min} \sim r_{particle}^{1.5} / r_{electrode} \tag{5}$$

The goal of our work is to test this scaling law quantitatively. If this scaling law is correct, then by using nano-electrodes, very small particles and possibly single molecules can be manipulated.

C. Quadrupole electrode traps

Huang and Pethig[4] have shown that four electrodes can form a DEP "trap" if the particles are undergoing negative DEP. We have lithographically fabricated a set of fourelectrode traps using Ti (10 nm)/Au(100 nm) on glass microscope cover slips. The geometry considered in the above section for the derivation of the scaling law (hemispherical electrodes) is different than the experimental quadrupole geometry, and so the predicted scaling law can only serve as a qualitative guide. Thus our conclusions shown in figure 1 should be considered an empirically determined scaling law, motivated by general arguments based on similar geometries.

Fig. 3. DNA trapped at the high field region of gold electrodes. Individual DNA molecules cannot be resolved in this image.

D. Experimental tests of scaling laws

In order to test the scaling of DEP with particle size and electrode geometry, we have used fluorescently labeled latex beads (Molecular Probes, Eugene, Oregon) of various sizes, ranging from 20 nm to 15 μ m. The beads were dispersed in DI water and imaged using an inverted microscope.

Since the dielectric constant of the latex beads is frequency dependent, the beads will undergo negative dielectrophoresis at only certain frequencies [2]. In our experiments we varied the frequency between 10 kHz and 30 MHz. Generally above some critical frequency a transition from positive to negative dielectrophoresis occurs. Above this critical frequency, particles near the plane of the electrodes are trapped in the central region. The dependence of this crossover frequency on medium conductivity has been studied in some detail[2], [5].

A typical image of a trapped latex bead from our experiments is shown in figure 2. We have measured the minimum voltage required to trap the beads for a variety of bead sizes and several electrode geometries. Above the critical crossover frequency, we find that the minimum voltage to trap a given particle size using a given electrode geometry depends only weakly on frequency. Using the data from electrodes with a 10 μ m and 100 μ m gap, we plot in figure 1 the minimum voltage required to trap a bead as a function of its size for beads of diameter 100 nm, 200 nm, 500 nm, 1 μ m, and 2 μ m. Our results clearly indicate that smaller electrode geometries allow the trapping of smaller particles for a given applied voltage. Since the two different electrode sizes have slightly different geometries, the tests at this point should be considered semi-quantitative.

III. MANIPULATION OF DNA

In order to demonstrate the feasibility of nano-manipulation with DEP we have performed experiments with fluorescently labeled λ phage DNA in DI water. The DNA is labeled with SYBR Green, an intercalating fluorescent dye, and imaged using an inverted microscope. The quadrupole electrode geometry was used, based on our (incorrect) expectation that the DNA would undergo negative DEP. The electrodes were fabricated on glass cover slips. A drop of solution containing the DNA is placed onto the cover slip, which is then covered by a second slip. This configuration was necessary for the inverted microscope configuration used.

We show in figure 3 DNA which has been trapped in the high-field region between microfabricated gold electrodes with a 10 μ m gap. We clearly observe positive dielectrophoresis (i.e. the DNA is attracted to high electric field intensity regions) over a range of frequencies between 500 kHz and 1 MHz.

We have repeated this experiment dozens of times with electrodes with central gaps of 5 μ m, 10 μ m and 20 μ m. With four electrode geometries with 50 μ m gaps, we were unable to see any effect of the electric fields on the DNA for applied voltages of up to 10 V. This is consistent with the scaling arguments presented above: large electrodes are less effective in trapping nano-sized objects than smaller electrodes. We have also been able to trap DNA using interdigitated, castellated electrodes[6].

Our work is consistent with the work of Washizu[7] but inconsistent with that of other published work on the DEP manipulation of DNA[8]. We speculate that this discrepancy may be related to the different solvents used. Tsukahara[8] used a buffer solution with conductivity of 4×10^{-3} S/m, while our work and that of Washizu[7] were done with DI water as the solvent with conductivity less than 2×10^{-4} S/m.

The electronic manipulation of DNA using DEP could have numerous applications, including electronic sequencing and point-of-care diagnostics. A further possible application is in the integration of top-down (microfabrication) with bottomup (chemically programmed) methods of manufacturing. Electronically manipulated DNA is a natural candidate for this project.

IV. NANO-MANIPULATION ARRAY PLATFORM

In order to demonstrate the scalability of these concepts for massively parallel, economic manipulation at the molecular scale, we have developed a nano-manipulation platform based on a trilayer process for arrays of DEP traps. Our current generation contains almost 300 traps on one chip, but this easily can be increased to thousands of traps. The platform is shown in figure 4. Fabricating an nxn array of quadrupole traps requires the crossing of the interconnects, hence a lithographically fabricated insulating layer needs to be deposited. Our arrays currently have quadrupoles of many different geometries with the goal to do massively parallel testing of the scaling of DEP with electrode geometry. The platform also clearly demonstrates the scalability of the use of DEP with lithographically fabricated electrodes, which is more difficult with other nano-manipulation processes such as STM and optical tweezers.

Nano-manipulation array platform



Fig. 4. We have developed a trilayer process for massively parallel nanomanipulation array platform.

V. NANOELECTRODES

Based on our phenomenological scaling data presented above, we predict that nanoparticles and maybe even single molecules can be trapped to the center of the electrodes with a 10 nm gap under the bias of about 1 volt. Carbon nanotubes present a natural candidate as nanoelectrodes for this project, as indicated in figure 5. This concept could provide a route to single molecule electronic devices which are actively fabricated. Much work to date on single molecule devices involves passive diffusion of molecules to small electrodes followed by passive covalent bonding to a Au electrode[9]. An advantage of our proposed scheme is that this assembly process could be actively, electronically controlled. This technique contains the rudimentaries of single molecule transistor fabrication with an essentially nano-electrochemistry approach[10]. In principle, by adjusting the frequency and amplitude of the applied voltages to the nanotube nanoelectrodes, chemical selectivity and reversibility can be achieved. A second advantage of the proposed scheme for organic molecular electronics is that the leads themselves would be organic, i.e. carbon. We speculate that this may allow new ways to control the alignment between the "source" and "drain" Fermi levels and the HOMO-LUMO separation in the single molecule, a crucial technological and scientific issue in the achievement of electrical contacts to individual molecules.

As a step in this direction, we have fabricated single walled carbon nanotubes between gold electrodes using a CVD process[11]. A typical device is shown in figure 6. In principle, the nanotube does not need to be contacted electrically at dc in order for DEP forces (which are based on ac electric fields) to apply. Its mere presence even as a floating electrode will modify the boundary conditions for the (ac) electric field,



Fig. 5. Schematic indication of the strongly divergent electric fields expected at the tip of a nanotube. These field gradients should exert strong influence which can overcome Brownian motion even at the molecular scale.



Fig. 6. Scanning electron micrograph image of a single walled carbon nanotube. This nanoelectrode could allow the electronic manipulation of individual molecules in solution using dielectrophoresis.

causing strong DEP forces in its vicinity. This considerably simplifies the fabrication requirements for such nanoelectrode systems.

VI. NANOMOTOR CONCEPT

Based on the above concepts and experiments, we present a concept for a dielectric nanomotor in figure 7. The electrodes are fabricated out of carbon nanotubes, and the DEP forces are used to rotate a nano-rotor in solution. By properly phasing the applied ac voltage to four concentric nanoelectrodes (the stators), a very small particle suspended in solution (the rotor) could be rotated at an RPM determined electronically. (This concept was briefly discussed in reference [2]). While such a nanomotor is technologically achievable in the not-to-distant future, the applications of such a device have yet to be discovered.

VII. CONCLUSIONS

We have presented a phenomenological scaling argument for the applicability of dielectrophoresis at the nanoscale based on measurements on latex beads between 20 nm and 10 μ m. We demonstrated the feasibility of nano-manipulation

Concept for a nanomotor



Fig. 7. Concept for a dielectric nanomotor. The electrodes are the stator, the nanoparticle in the center the rotor.

using DNA, and demonstrated the scalability by developing a nano-manipulation array platform. Based on our work we predict that nanoelectrodes should be useful for manipulating nanoparticles and as the components of a nanomotor. These are small steps towards an economical, massively parallel nanomanipulation platform of both neutral and charged objects at the nanoscale on a chip for programmable, rational, electronically controlled molecular manufacturing.

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