Direct measurements of the frequency-dependent dielectrophoresis force

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Dielectrophoresis (DEP), the phenomenon of directed motion of electrically polarizable particles in a nonuniform electric field, is promising for applications in biochemical separation and filtration. For colloidal particles in suspension, the relaxation of the ionic species in the shear layer gives rise to a frequency-dependent, bidirectional DEP force in the radio frequency range. However, quantification methods of the DEP force on individual particles with the pico-Newton resolution required for the development of theories and design of device applications are lacking. We report the use of optical tweezers as a force sensor and a lock-in phase-sensitive technique for analysis of the particle motion in an amplitude modulated DEP force. The coherent detection and sensing scheme yielded not only unprecedented sensitivity for DEP force measurements, but also provided a selectivity that clearly distinguishes the pure DEP force from all the other forces in the system, including electrophoresis, electro-osmosis, heat-induced convection, and Brownian forces, all of which can hamper accurate measurements through other existing methods. Using optical tweezers-based force transducers already developed in our laboratory, we have results that quantify the frequency-dependent DEP force and the crossover frequency of individual particles with this new experimental method. © *2009 American Institute of Physics.* [DOI: 10.1063/1.3058569]

I. INTRODUCTION

Dielectrophoresis (DEP) was first described by Pohl in 1951 (Ref. 1) as the translational motion of neutral matter caused by polarization effects in a nonuniform electric field.[2] For colloidal particles in suspension, the force can produce directed motion of electrically polarizable particles in the convergent field direction. For particles in aqueous solution, the ionic shear layer surrounding the particle must be considered as part of the "composite particle"; the polarization of which as a whole is responsible for the frequency dependent DEP.[3] The DEP force as a function of frequency can be expressed as

$$F_{\mathrm{DEP}} = 2\pi r^3 \varepsilon_m K_{\mathrm{Re}}(\omega_{\mathrm{DEP}}) \nabla E^2, \tag{1}$$

where r is the particle radius, ε_m is the permittivity of the surrounding medium, E is the electric field, and $K_{\mathrm{Re}}(\omega)$ is the real part of the Claussius–Mossotti function

$$K(\omega_{\mathrm{DEP}}) = \frac{\tilde{\varepsilon}_p - \tilde{\varepsilon}_m}{\tilde{\varepsilon}_p + 2\tilde{\varepsilon}_m}, \tag{2}$$

where $\tilde{\varepsilon}_m$ and $\tilde{\varepsilon}_p$ are the complex permittivities of the medium and particle, respectively, and the complex permittivity is $\tilde{\varepsilon} = \varepsilon - i\sigma/\omega_{\mathrm{DEP}}$, where ε is the real part of the permittivity, σ is the conductivity, and ω_{DEP} is the angular frequency of the electric field.

In this paper we present measurements of the frequency-dependent DEP force using optical tweezers (OT) as a force transducer. In the past, the optical tweezers techniques have been used to quantify microparticle dynamics[4,5] and the microrheology of complex fluids.[6] The use of OT has the distinct advantage in that it will not interfere with the DEP effect since the polarization that

drives the OT occurs at optical frequencies, well out of the range of the radio frequencies (RFs) in DEP. We show results of an accurate determination of the frequency-dependent force for two different particle materials in a variety of particle sizes across several decades in frequency.

II. EXPERIMENTAL

A. Materials

The particles used in the experiment were polystyrene and silica microspheres obtained from Duke Scientific in diameters of 0.996, 1.5, 1.9, and 2.1 μm. The purchased particle suspensions were treated by sonication to prevent agglomeration; thereafter the solution was diluted with de-ionized water; the final solutions were measured to have a conductivity of (10^{-3} S/m). The prepared stock solution was further diluted so that only one sphere was visible in the field of view (60×100 μm) to ensure that the interactions between particles were negligible. The colloid solution was loaded onto a sample chamber composed of a microscope glass slide and a cover slip with patterned DEP electrodes.

B. Methods

1. Optical tweezer-based DEP force spectroscopy

To manipulate particles and measure the frequency-dependent DEP force, we used an optical tweezers as a force sensor while administering the DEP electric field. To facilitate coherent harmonic detection that significantly improved the signal-to-noise (S/N) ratio of force sensing, the DEP force was amplitude modulated (AM) at a frequency two orders of magnitude lower than the RF-range DEP frequencies. The use of amplitude modulation has a distinct advantage, since the motion of the particles in the optical tweezers will be at exactly twice the AM frequency due to the $\boldsymbol{E}^2$ dependence of the DEP force (see below). When using the standard phase lock-in detection technique to analyze the motion of the particle, the motion produced by the DEP force is therefore isolated from the effects of the other non-DEP forces.

2. The motion of the optically trapped particle in an AM DEP force field

An AM DEP field at position X can be written as $E(X,t)=E_0(X)\cos(\omega_m t)\cos(\omega_{\mathrm{DEP}}t)$, where ω_m is the amplitude modulation frequency and ω_{DEP} is the DEP frequency. According to Eq. (1) the DEP force on a particle in the direction of the gradient of this AM field $\hat{x}$ can be written as

$$F_{\mathrm{DEP}}(\hat{x}) = \tfrac{1}{4}AK_{\mathrm{Re}}(\omega_{\mathrm{DEP}})\nabla_x E_0^2[1+\cos(2\omega_m t)][1+\cos(2\omega_{\mathrm{DEP}}t)], \tag{3}$$

where A is the geometrical factor $2\pi r^3\varepsilon_m$.

A particle trapped by optical tweezers can be viewed as being situated in a Gaussian potential well. Under an external force the particle moves in the direction of that force while balanced by the restoring force of the tweezers. If the force is small such that the displacement from the center of the tweezers is only a small fraction of the width of the Gaussian potential well, the force balance can be written as $F_{\mathrm{ext}}=k_{\mathrm{OT}}\,x$, where F_{ext} is the external force, k_{OT} is the spring constant, and x is the displacement of the particle from the center of the optical tweezers.[4] Now if the particle held in the optical well is also acted on by the DEP force described by Eq. (3), we can write the equation of motion of the particle as

$$m\ddot{x} + 6\pi\eta\dot{x} + k_{\mathrm{OT}}x = \tfrac{1}{4}AK_{\mathrm{Re}}(\omega_{\mathrm{DEP}})\nabla_x E_0^2[1+\cos(2\omega_m t)][1+\cos(2\omega_{\mathrm{DEP}}t)]. \tag{4}$$

For micron size colloidal particles in water, inertial effects are small and since the particle is too sluggish to respond to the DEP frequency ($\omega_{\mathrm{DEP}}\geqslant 10^4$ rad/s), Eq. (4) can be simplified as follows:

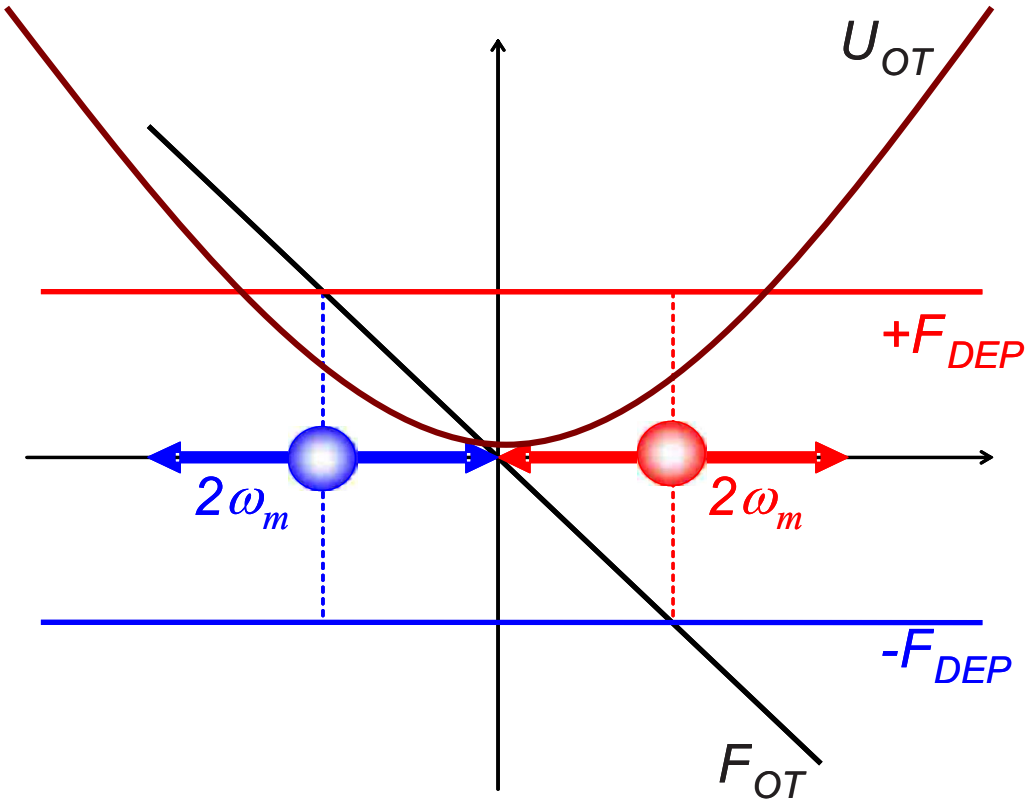

FIG. 1. A diagram depicting the potential well formed by OT. Under an AM DEP force the particle executes an oscillatory motion centered at the point off-set from the center of the optical trap.

$$6\pi\eta r\dot{x} + k_{\mathrm{OT}}x = \tfrac{1}{4}AK_{\mathrm{Re}}(\omega_{\mathrm{DEP}})\nabla_x E_0^2[1+\cos(2\omega_m t)]. \tag{5}$$

Note that "1" in the first bracket on the right-hand side of Eq. (5) produces a direct current offset of the particle position from the tweezer center. The particle oscillates around the off-set point and follows the steady state solution given by

$$x(\omega_m,\omega_{\mathrm{DEP}}) = \frac{AK_{\mathrm{Re}}(\omega_{\mathrm{DEP}})\nabla_x E_0^2}{4\sqrt{k_{\mathrm{OT}}^2+(12\pi\eta r\omega_m)^2}}\{1+\cos(2\omega_m t-\delta)\}, \tag{6a}$$

$$\delta = \tan^{-1}\left(\frac{12\pi\eta r\omega_m}{k_{\mathrm{OT}}}\right). \tag{6b}$$

The experimental conditions of our setup yielded a high enough k_{OT} and a low enough amplitude modulation frequency ($\omega_m \leqslant 200$ rad/s) such that $k_{\mathrm{OT}} > 12\pi\eta r\omega_m$, we have δ equal to a small constant at a constant ω_m.

From Fig. 1, we can see that the particle depicted as the red dot is under the influence of the potential well formed by the optical tweezers (U_{OT}). Under an AM DEP force the particle executes an oscillatory motion at the frequency $2\omega_m$ and is centered at the offset point slightly to the right (positive DEP) or to the left (negative DEP) of the OT center. According to Eq. (6), at the crossover frequency where $K(\omega_{\mathrm{DEP}})$ changes sign, the particle motion experiences a sharp change of phase shift by 180°, relative to the phase of the amplitude modulation frequency.

The motion of the particle can be tracked by a quadrant photodiode that detects the diffracted light from a probing laser beam aligned with the trapping laser. The displacement and the phase shift of the motion of the particle are analyzed by means of a lock-in amplifier operating at the

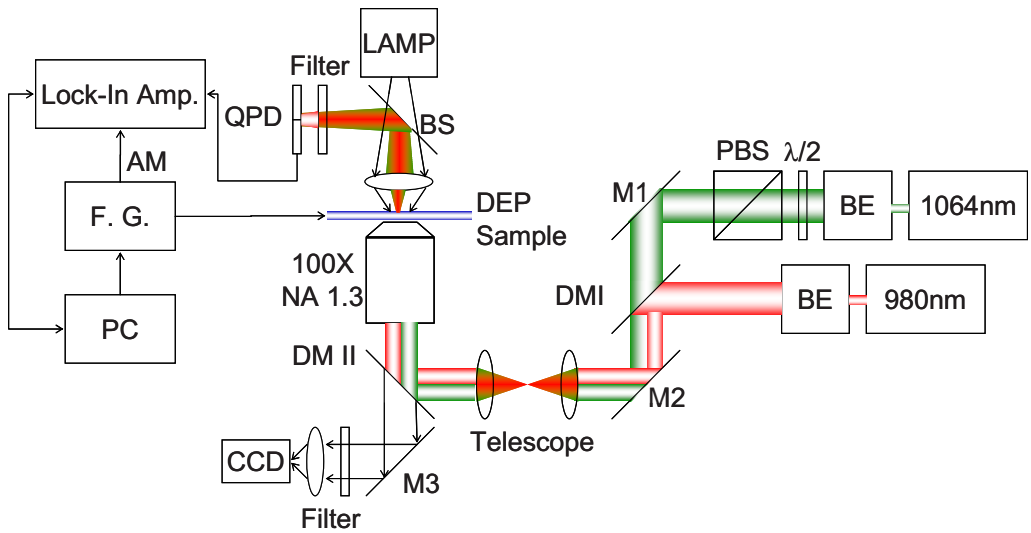

FIG. 2. Schematic of the DEP force spectroscopy setup.

second harmonic of the AM frequency ($2\omega_m$). The crossover frequency is obtained by monitoring where the phase of the particle motion relative to that of the AM field shifts by 180° while the magnitude of the particle displacement approaches zero.

The use of the phase lock-in technique significantly improves the S/N of the motion detection and allows us to measure the vanishingly small DEP forces near the crossover point. In addition, the $\boldsymbol{E}^2$ dependence of the DEP force in conjunction with the amplitude modulation scheme mitigates the problems usually associated with the intrinsic electrophoretic (EP) and electro-osmotic (EO) forces that can also cause particle motion. These problems are alleviated in our experiment since the particle motion corresponding to EP and EO occur at the frequency ω_m, and the AM DEP force-induced particle motion occurs at $2\omega_m$. Operating the lock-in amplifier with the reference frequency set to $2\omega_m$ easily rejects the signals caused by EP and EO. Furthermore, the thermally driven particle fluctuations will not affect the phase of the lock-in DEP force measurements since the thermal fluctuations were incoherent.

3. Instrumentation

The experimental setup is depicted in Fig. 2. Optical trapping is accomplished by a tightly focused 1064 nm infrared laser and particle tracking is made possible by the use of a 980 nm laser aligned to be collinear and parfocal to the same focal point as the 1064 nm laser. The 1064 nm laser is expanded and collimated to just overfill the back aperture of the objective lens, which ensures that a diffraction limited spot is created for particle trapping; as this is critical for isotropic trapping stiffness in the direction transverse to the beam propagation. To prevent contribution to the optical trapping effect, the 980 nm tracking beam is attenuated so that its power is two orders of magnitude lower than that of the 1064 nm laser. Movements of the particle were tracked by the 980 nm laser beam and detected by the quadrant photodiode (QPD). For accurate tracking, the voltage reading of the QPD was kept to be a linear function of the particle displacement from the trapping center. Our present particle tracking system provides spatial resolution in the subnanometer range. The calibration of the spring constant k_{OT} of the optical tweezers was done by oscillating a particle in water in the absence of DEP, and the phase of the displacement of the particle was used to determine the force constant of the trap.[4,7] Once the system is properly calibrated, we converted displacement readings in to absolute forces by Eq. (6a). We used a function generator (Stanford Research DS345) to administer the electric field to the DEP electrodes and we set the lock-in amplifier (Stanford Research SR830) to monitor the second harmonic of the DEP modulation frequency for the experiments.

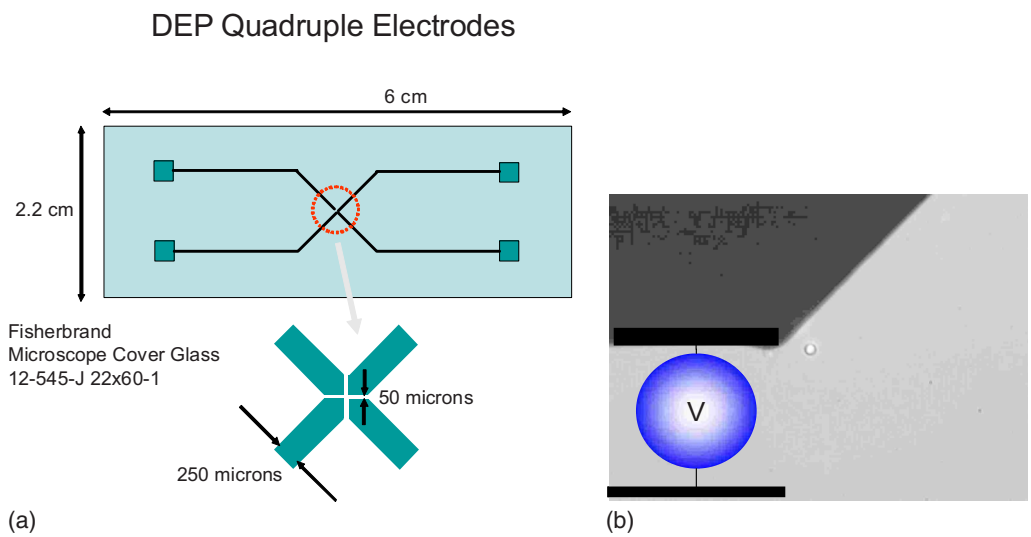

FIG. 3. (a) The schematic of a DEP quad-electrode and (b) a CCD image of a 1.5 μm optically trapped polystyrene particle near the edge of an electrode.

A standard microfabricated DEP quadruple electrode set used to conduct our experiments is shown in Fig. 3(a). Only the left pair of electrodes were used in this study. Figure 3(b) shows a charge coupled device (CCD) image of a 1.5 μm polystyrene particle trapped by an optical tweezer (not shown) located near an edge of one of the electrodes. The location was chosen so that it was not too close to the edge of the electrode so ∇E^2 is approximately a constant and the force was large enough to be reproducibly measurable.

III. EXPERIMENTAL RESULTS

To check the reliability of the optical tweezers force transducer, we first tested our setup to see if it yielded a DEP force that is a linear function of the square of the electric field.[8] The data in Fig. 4 shows that the readings of the displacement amplitude from the lock-in amplifier are linearly proportional to the square of the voltage across the DEP electrodes. The two sets of data shown in Fig. 4 were taken at two different DEP frequencies, 10 kHz and 1 MHz for a 1.5 μm polystyrene particle. Since the measurements were done at exactly the same location, a short distance from one of the electrodes, we expected the electric field strength to be proportional to the voltage across the electrodes. Because the QPD has a finite linear range, the displacement of the particle exceeded the linear range at high field strength, causing the apparent saturation of the DEP force for 10 kHz. Not shown in this figure is that the particle displacement went through a 180° phase shift when DEP frequency crossed 2×10^5 Hz (see Fig. 5). From the fact that the measured force occurred at twice the amplitude modulation frequency and that the magnitude of the force is linear with the square of the magnitude of the voltage across the electrodes, we concluded the measured forces were indeed pure DEP forces.

For measurements of the frequency-dependent DEP force, we held a particle in the calibrated optical tweezers and administered the DEP force with a 5 V (peak-to-peak) electric field across a 50 μm electrode gap while sweeping the DEP frequencies from 10 kHz to 10 MHz. Using the optical tweezers spring constant determined for each particle size, we were able to convert the lock-in output from the QPD to the absolute DEP force imparted on the optically held particles. Figure 5(a) shows the measured forces as a function of DEP frequency for four different sizes of polystyrene latex particles. Figure 5(b) shows the phase shift of the particle displacement as a function of frequency. A sharp 180° phase shift shown in the right panel was indeed observed at

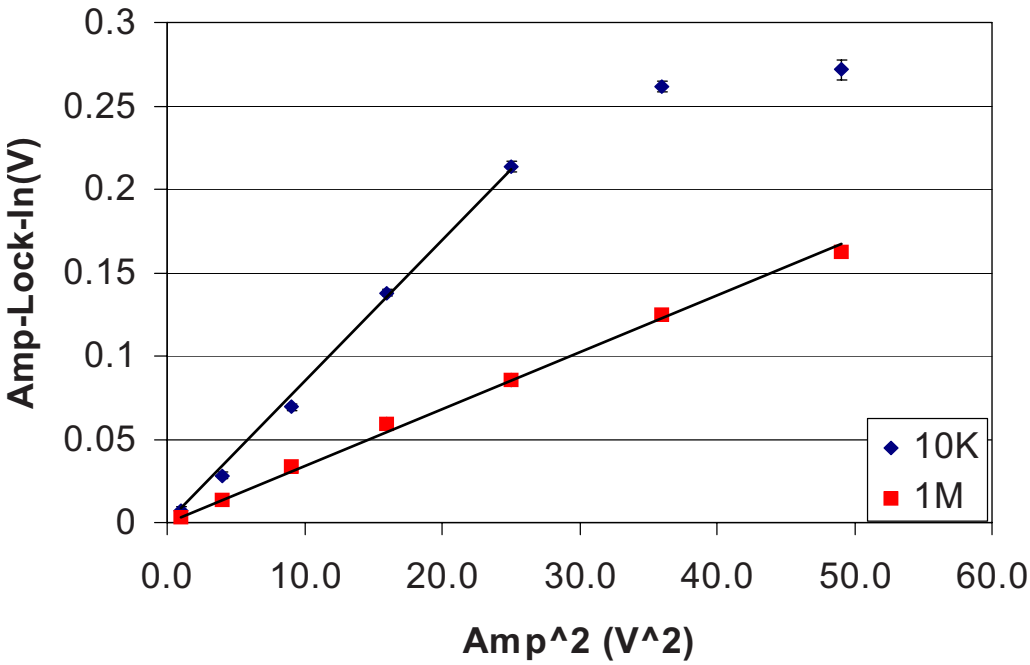

FIG. 4. Data depicting the linearity of the DEP force as a function of electric field amplitude squared.

a frequency corresponding to the exact same frequency where the DEP force was zero shown in the left panel. This phase shift data provided a much more clear definition of the crossover frequency.

Figure 6 shows the crossover frequency as a function of particle size for eight different latex particle sizes. The data for the four smaller size particles (polystyrene latex particles 557, 282, 216, and 93 nm in diameter, open squares) are taken from the data presented in a paper by Green and Morgan.[8] Data from our study are represented in the figure (crosses) for polystyrene particles with diameters of 2.1, 1.9, 1.5, and 0.996 μm in a buffer solution with low (10^{-3} S/m) electrolyte conductivity. The power law dependence of the crossover frequency on the particle size appears to be weaker than the expected −1 for submicron particles, however, the exponent appears to be stronger than −4 for larger particles. It is not clear to us why there exists such dramatic change in the power law exponents over this particle size range.

Figure 7 shows the frequency dependent force and phase shift data for two identically sized particles of different dielectric properties. Both the polystyrene and silica particles were used under the same solution conductivity and electrode conditions. The higher overall magnitude of

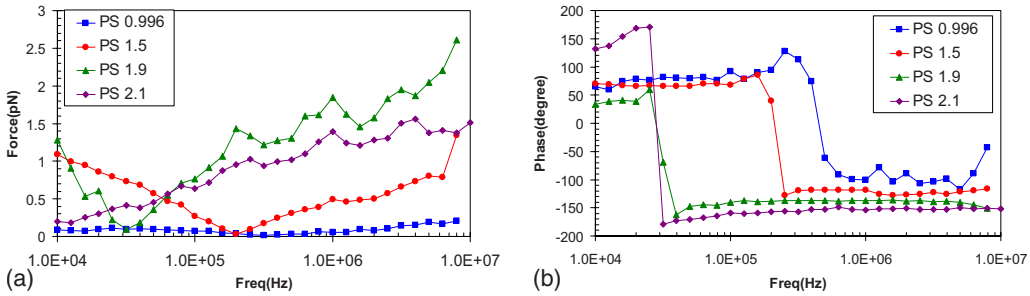

FIG. 5. The DEP force (left) and displacement phase (right) as a function of frequency for four different sizes of polystyrene particles with diameters ranging from 0.998 to 2.1 μm.

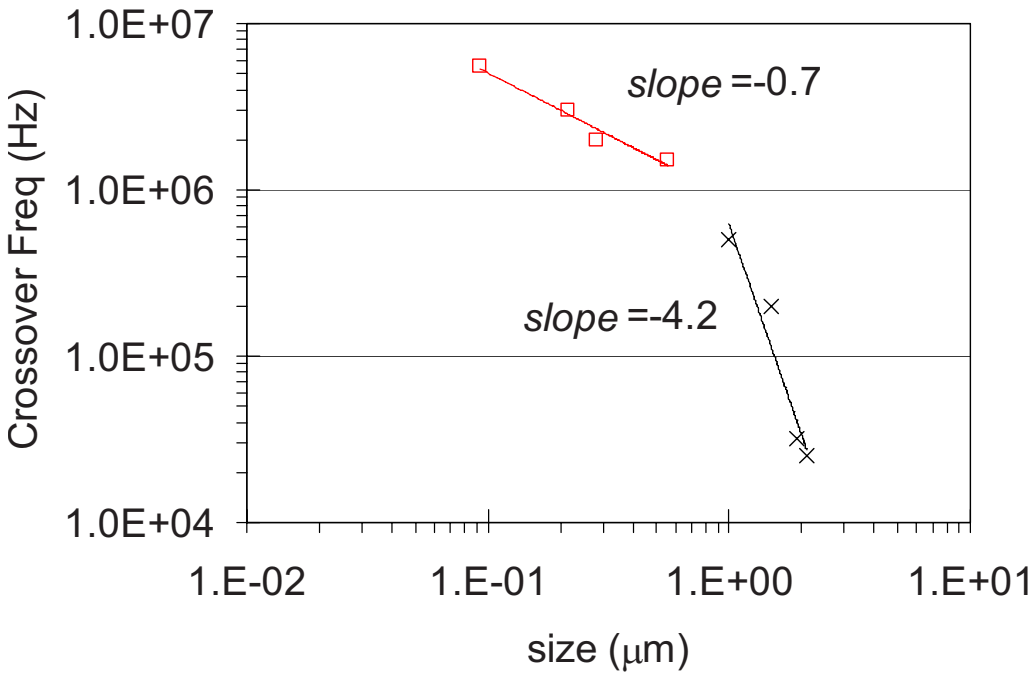

FIG. 6. Crossover frequencies as a function of particle size [data shown in squares are from the paper by Green and Morgan (Ref. 8)].

the DEP force and the higher crossover frequency of the polystyrene than that of the silica particles are consistent with the higher surface charge density usually associated with polystyrene particles relative to that of the silica particles.

IV. CONCLUSIONS

We have shown in this paper a novel optical tweezer-based DEP force spectroscopy that is not only reliable in determining the crossover point, but also in measuring the frequency-dependent DEP forces with unprecedented accuracy across a large range of RFs. Polystyrene microspheres with four different particle sizes and comparable size silica microspheres were tested using this new method. Through modification of electrolyte conditions and particles surfaces coupled with judicious design of electrode geometries, the force method reported here can be used to further the understanding of the dielectric response of materials through a close comparison between experiments and the newly advanced DEP theories.

In addition to scientific interests, the DEP force invites inventive tuning for applications, and it is widely considered to be advantageous for concentration and sorting of micron scale objects

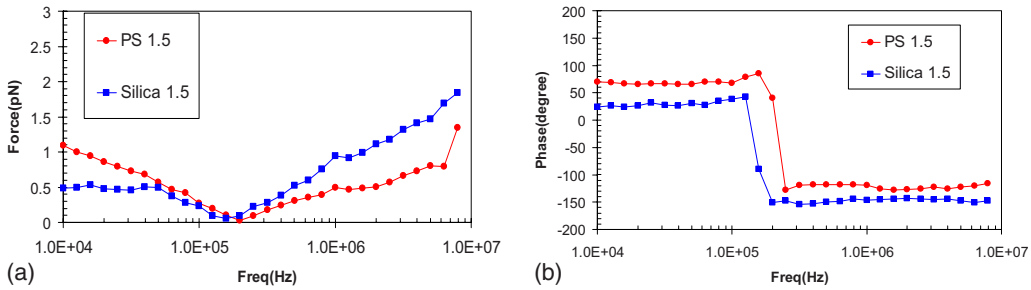

FIG. 7. (a) The magnitude and (b) phase shift of the particle displacement due to the DEP force for 1.5 μm silica and polystyrene spheres.

with high selectivity.[9–12] The DEP method potentially possesses the unique capability of isolating and analyzing a wide range of particle types (cells,[13,14] bacteria,[15] viruses,[16,17] DNA and proteins[18]). To date, the use of DEP to perform these sorting tasks has been limited by the qualitative nature of the force characterization methods. The advent of the accurate OT-based DEP force spectroscopy is critical to the design and optimization of device applications that utilize DEP. By designing a DEP system with well-characterized forces, it is feasible that it could be used for high throughput particle sorting as well as a multiscale approach to electrically based sorting.

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