Dielectrophoresis of an Inhomogeneous Colloidal Particle under an Inhomogeneous Field: A First-Principles Approach

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In reality, colloidal particles often possess an arbitrary inhomogeneity profile. We present a first-principles approach to dielectrophoresis of such an inhomogeneous colloidal particle under an inhomogeneous field of an oscillating electric dipole moment. For numerical simulations, we treat the inhomogeneous particle as a specific graded one with a physically motivated dielectric model and conductivity profile. We find that both multipolar interactions and spatial fluctuations inside the particle can affect the dielectrophoretic spectrum of the particle significantly.

I. Introduction

The movement of colloidal particles or biological cells in an applied nonuniform AC (or DC) electric field is called dielectrophoresis,[1−3] which is actually the dielectric analogue of optical tweezers.[4] In dielectrophoresis, particles redistribute themselves around the system, and the dielectrophoretic force holds the particles in place depending on the local electric field and on the dielectric properties of the particles themselves and the suspending medium. This force comes from the interaction between the induced electric dipole moment inside the particle and the external electric field. So far, dielectrophoresis is typically used for micromanipulation and separation of biological cells, and it has recently been successfully applied to submicron size particles as well. Specific applications include diverse problems in medicine, colloidal science, and nanotechnology (e.g., separation of nanowires,[5] viruses,[6] latex sphere,[7,8] DNA,[9] and leukemic cells,[10] as well as lab-on-a-chip designs[11]). In contrast to electrophoresis[12] that functions with only charged particles, dielectrophoresis works for both neutral and charged particles. Similar to optical tweezers, dielectrophoresis must also dominate the thermal Brownian motion.

The dielectrophoretic force exerted on a particle can be either attractive or repulsive depending on the polarizability of the particle in comparison to the medium. For a nonuniform AC electric field, the magnitude and the direction of the dielectrophoretic force depends on the frequency, changes in surface charge density, and free charges in the vicinity of the particle. The frequency at which the DEP force changes its sign is called the crossover frequency (f_{CF}). Analysis of the crossover frequency as a function of the host medium conductivity can be used to characterize the dielectric properties of particles and is at present the principal method of dielectrophoretic analysis for submicrometer particles.[6,7,13]

For a colloidal particle like biological cells having dimensions of the order of more than 1 μm, located in a strong electric field gradient, higher-order multipolar moments must be taken into account.[14−17] In real situations, colloidal particles often possess an arbitrary inhomogeneity profile. For instance, biological cells are a kind of model colloidal particle with an arbitrary inhomogeneity profile of interest as their composition varies through the object. In general, inhomogeneous materials have quite different physical properties from the homogeneous materials.[18] However, traditional theories[19] like dipole approximation, multipole expansion,[20] and Maxwell stress tensor[14] cannot be used to deal with inhomogeneous particles directly. Jones[17] put forth the multipole moment method for homogeneous particles and multishell particles. In this paper, we shall present a first-principles approach to dielectrophoresis of such an inhomogeneous particle in the presence of an inhomogeneous electric field produced by an oscillating electric dipole moment.

The paper is organized as follows. In section II, based on a first-principles approach, we present the formalism for the dielectrophoretic force (as well as the crossover frequency) acting on an inhomogeneous particle under an inhomogeneous electric field. In section III, numerical results are given under different conditions. We end the paper with a discussion and conclusion in section IV.

II. Formalism

A. Polarization Due to an Oscillating Electric Dipole Moment. We shall investigate the force between a graded spherical particle and an oscillating electric dipole moment p, which is a function of time t, $p = p_0 \exp(2\pi i f t)$. Here p_0 is the amplitude of the dipole moment, $i = (-1)^{1/2}$, and f is the frequency of an external field. This dipole moment is located along the z axis but not at the origin, which can produce a multipole field, and thus, the particle located at the origin in this field will get a dielectrophoretic force. Suppose that the oscillating electric dipole moment p is a distance R from

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the center of the inhomogeneous spherical particle (located at the origin) with radius a and dielectric gradation profile $\tilde{\epsilon}_1(r)$, $r \leq a$, embedded in a host fluid of dielectric constant $\tilde{\epsilon}_2$. Here $\tilde{\epsilon} = \epsilon + \sigma/(2\pi i f)$, where ϵ denotes the real part of the dielectric constant, σ stands for conductivity, and $i = (-1)^{1/2}$. In other words, the oscillating dipole moment is placed on the z axis at $z = R$, so that the problem has azimuthal symmetry. Then, the Maxwell equations read

$$\nabla\cdot\mathbf{D} = 0 \tag{1}$$

$$\nabla \times \mathbf{E} = 0 \tag{2}$$

Moreover, from eq 2, $\mathbf{E}$ can be written as the gradient of a scalar potential Φ, $\mathbf{E} = -\nabla\Phi$, yielding a partial differential equation

$$\nabla\cdot[\tilde{\epsilon}(\mathbf{r})\nabla\Phi] = 0 \tag{3}$$

where $\tilde{\epsilon}(r) = \tilde{\epsilon}_1(r)/\tilde{\epsilon}_2$ in the particle and $\tilde{\epsilon}(r) = 1$ in the host. The derivation is a standard textbook problem.[19] In the spherical coordinates (r,θ,φ), Φ satisfies

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\tilde{\epsilon}(r)\frac{\partial\Phi}{\partial r}\right) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\,\tilde{\epsilon}(r)\frac{\partial\Phi}{\partial\theta}\right) + \frac{1}{r^2\sin^2\theta}\frac{\partial}{\partial\varphi}\left(\tilde{\epsilon}(r)\frac{\partial\Phi}{\partial\varphi}\right) = 0 \tag{4}$$

As the oscillating electric dipole moment is along the z axis, Φ is independent of the azimuthal angle φ. To separate the variables, we set $\Phi = f(r)\Theta(\theta)$. For the radial function $f(r)$, we obtain a radial equation

$$\frac{\mathrm{d}}{\mathrm{d}r}\left(r^2\tilde{\epsilon}(r)\frac{\mathrm{d}f(r)}{\mathrm{d}r}\right) - m(m+1)\tilde{\epsilon}(r)f(r) = 0 \tag{5}$$

where m is an integer. The radial equation [eq 5] yields two solutions $f_m^+(r)$ and $f_m^-(r)$ that are regular at the origin ($r = 0$) and infinity ($r \to \infty$), respectively. In addition, $\Theta(\theta)$ satisfies the Legendre equation (19). The potential inside and outside the graded sphere is given by respectively

$$\Phi_i(r,\theta) = \sum_{m=1}^{\infty} A_m f_m^+(r) P_m(\cos\theta) \tag{6}$$

$$\Phi_m(r,\theta) = \frac{p}{4\pi\tilde{\epsilon}_2}\sum_{m=1}^{\infty} -(m+1)\frac{r^m}{R^{m+2}}P_m(\cos\theta) + \sum_{m=1}^{\infty} B_m r^{-m-1} P_m(\cos\theta) \tag{7}$$

The boundary conditions are given by

$$\Phi_i(r,\theta)|_{r=a} = \Phi_m(r,\theta)|_{r=a} \tag{8}$$

$$\tilde{\epsilon}_i(r)\frac{\partial\Phi_i(r,\theta)}{\partial r}\Big|_{r=a} = \tilde{\epsilon}_2\frac{\partial\Phi_m(r,\theta)}{\partial r}\Big|_{r=a} \tag{9}$$

Solving the above equation, we can obtain the coefficients

$$A_m = \frac{p(m+1)}{f_m^+(a)}\frac{a^m}{R^{m+2}}(H_m - 1) \tag{10}$$

$$B_m = p(m+1)\frac{a^{2m+1}}{R^{m+2}}H_m \tag{11}$$

where

$$H_m = \frac{m(\alpha_m - 1)}{m(\alpha_m + 1) + 1} \tag{12}$$

is the multipole polarizability of a graded spherical particle[21] and

$$\alpha_m = \frac{\tilde{\epsilon}_i(a)}{\tilde{\epsilon}_2}\frac{f_m^{+\prime}(a)}{m f_m^+(a)} \tag{13}$$

is the equivalent dielectric constant of a graded spherical particle.[22]

Next, we will give the electric field in order to calculate the force between the spherical particle and the electric dipole moment

$$\mathbf{E} = -\nabla\Phi \tag{14}$$

We will focus on the induced polarization in the dipole moment, that is, $\mathbf{r} = R\hat{\mathbf{e}}_z$, and $\theta = 0$, i.e., $P_m(\cos 0) = 1$. Then

$$\Phi_{\mathrm{ind}}(R, 0) = \sum_{m=1}^{\infty} B_m R^{-m-1} \tag{15}$$

So far, the electric field due to the induced polarization may be expressed as

$$\mathbf{E}_{\mathrm{ind}} = \sum_{m=1}^{\infty} B_m(m+1)R^{-m-2}\hat{\mathbf{e}}_{\mathbf{z}} \tag{16}$$

It is straightforward to compute the force $\mathbf{F}_{\mathrm{p}}$ acting on the dipole moment, according to

$$\mathbf{F}_{\mathrm{p}} = (\mathbf{p}\cdot\nabla)\boldsymbol{E}_{\mathrm{ind}} \tag{17}$$

Then, we obtain

$$\mathbf{F}_{\mathrm{p}} = -\frac{\hat{\mathbf{e}}_z p}{4\pi\tilde{\epsilon}_2}\sum_{m=1}^{\infty} B_m(m+1)(m+2)R^{-m-3} \tag{18}$$

B. Dielectrophoretic Force Acting on the Graded Particle. According to Newton's third law (i.e., action−reaction principle), the force (which is the dielectrophoretic force) acting on the graded sphere is

$$\mathbf{F}_{\mathrm{M}} = \frac{\hat{\mathbf{e}}_z p}{4\pi\tilde{\epsilon}_2}\sum_{m=1}^{\infty} B_m(m+1)(m+2)R^{-m-3} \tag{19}$$

For the dipole moment with a function of time t [e.g., $p = p_0 \exp(2\pi i f t)$], we obtain the multipole time-averaged dielectrophoretic force acting on the graded sphere as

$$\mathbf{F}_{\mathrm{M}} = \frac{\hat{\mathbf{e}}_z {p_0}^2}{8\pi\epsilon_2}\sum_{m=1}^{\infty}\mathrm{Re}\left[(m+1)^2(m+2)\frac{a^{2m+1}}{R^{2m+5}}H_m\right] \tag{20}$$

In this equation, Re[$\cdots$] means the real part of [$\cdots$]. For $m = 1$, the dipole dielectrophoretic force $\mathbf{F}_{\mathrm{D}}$ is given by

$$\mathbf{F}_{\mathrm{D}} = \frac{\hat{\mathbf{e}}_z 3{p_0}^2}{2\pi\epsilon_2}\frac{a^3}{R^7}\mathrm{Re}\left[\frac{F_1 - 1}{F_1 + 2}\right] \tag{21}$$

C. Crossover Frequency. Crossover frequency f_{CF} is the frequency at which the dielectrophoretic force is zero. Thus, f_{CF} can be obtained by solving $F_M = 0$ and $F_D = 0$ for multipole and dipole contribution, respectively.

III. Numerical Results

For numerical calculations, take $\epsilon_2 = 80\epsilon_0$ (here ϵ_0 donates the dielectric constant of free space), $\sigma_2 = 2.8 \times 10^{-4}$ S/m. Similar to a biological cell case,[23] we take a physically motivated model gradation profile in an inhomogeneous particle (thus also called graded particles, namely, particles with spatial gradient in their structure)

$$\epsilon_1(r) = 75\epsilon_0 + A(r/a), \quad r \le a \tag{22}$$

$$\sigma_1(r) = 2.8 \times 10^{-2}(r/a)^k, \quad r \le a \tag{23}$$

where A and (dimensionless) k are parameters turning the gradation profile. Biomaterials such as cells and bones are typical examples as their composition varies through the object. Gradation can also be used to control and improve the strength and other properties; for a recent review, see ref 24. In the following calculations, for convenience, the particle's radius is set to be unity (e.g., the separation such as R is measured in unit of a).

From the results in Figures 1 and 3, it is evident that the multipole contribution to the dielectrophoretic force becomes significantly at small separation R, especially at the low frequency region. We estimate the importance of the multipole contribution, by plotting Figures 2 and 4 showing the ratio of a multipole to a dipole force. At small separations the ratio can deviate significantly from the unity, especially in the vicinity of crossover frequencies.

For each curve of Figures 2 and 4, the left peak (upward) actually corresponds to the crossover frequency at which $F_D = 0$ is predicted by the dipole contribution, and the right peak (downward) actually corresponds to the crossover frequency at which $F_M = 0$ is predicted by the multipole contribution. We also evaluate the dielectrophoretic crossover frequency in Figure 5. From Figures 2 and 4, it is evident to see that the multipole contribution can change the crossover frequency in comparison with that predicted by the dipole moment. That is, the dieletrophoretic crossover frequency also depends on the multipole contributions. On the other hand, the gradation parameter A can adjust the crossover frequency significantly (Figure 2), and the gradation parameter k can adjust the crossover frequency slightly (Figure 4). However, the existence of multipole contribution could increase the crossover frequency if the medium conductivities are given, see Figure 5. The difference between the multipole and dipole calculated frequencies becomes smaller and smaller as A approaches zero, see Figure 5. In detail, at low inhomogeneities in the particle properties, the effect of including multipolar interactions is small, but it can play a somewhat limited role. The effect of inhomogeneities themselves on the crossover frequency is much more pronounced. Increasing the degree of inhomogeneities in the particle properties (e.g., increasing A or k from low magnitudes to high magnitudes) can make the crossover frequency red-shifted (namely, be shifted to lower frequencies). Finally, we can conclude that the dielectrophoretic crossover spectrum is robust to any approximate theory used to calculate the force.

IV. Discussion and Conclusion

Here some comments are in order. Based on a first-principles approach, we have showed that the multipole contribution to the dielectrophoretic spectra and the dielectrophoretic crossover frequency is significant in comparison with the sole dipole contribution, especially for the effect of the gradation profile parameters k and A. Also, the importance of the multipole contribution is especially pronounced at small separations between the graded particle and the dipole moment. Some important applications do require such a small electrode/particle distance, for instance electrofusion of cells, as well as (electrostatic) near field scanning of cells. In these applications, the electrode/particle separation can be very small.

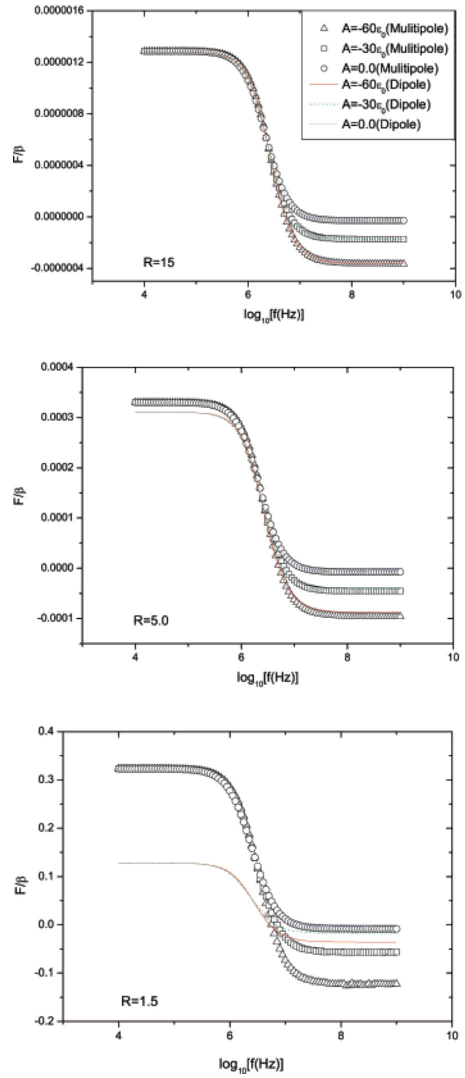

Figure 1. Dielectrophoresis spectra given by multipole and dipole forces, F_M [eq 20] and F_D [eq 21]. The forces are both denoted as F and normalized by the constant $\beta = p_0^2/(4\pi\epsilon_2)$ for different A and R. Parameter: $k = 0$.

A multi-shell approach[14] seems inadequate as the conductivity is not stratified or layered in cells. Although the dielectric profile is roughly constant in the interior of cells, the conductivity profile is rather continuous. Thus, we have modeled the conductivity profile as power-law dependence on radius, whereas the permittivity profile is linear. At the same time, the present continuous profiles admit an exact analytic solution, which seems to be superior to the multi-shell model where only numerical solutions exist.

It is known that surface conductance effects on crossover frequency are relevant.[23] Nevertheless, a large conductivity at the particle surface (implied in our conductivity profile) captures the effects at least partially. Finally, it should be noted that eq

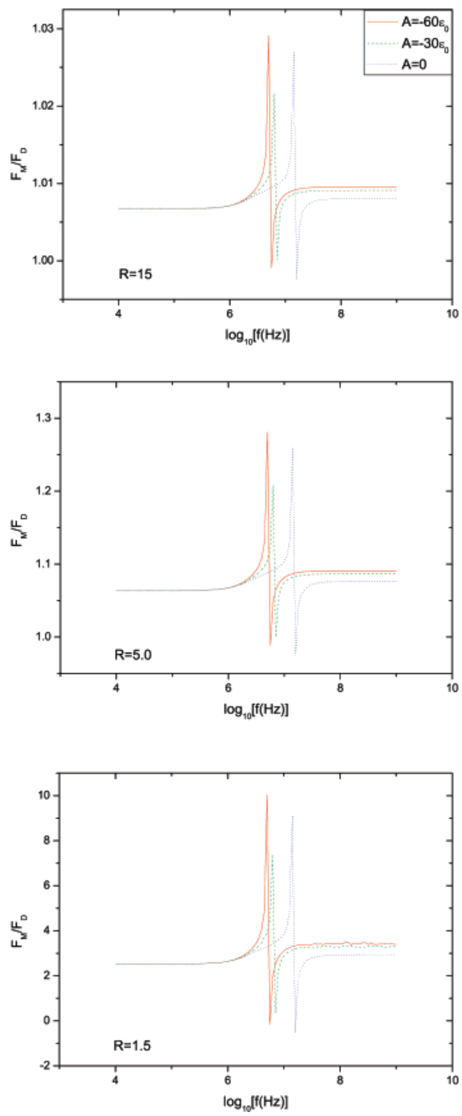

Figure 2. F_M/F_D as a function of the frequency of external electric fields for different A and R. Parameter: $k = 0$.

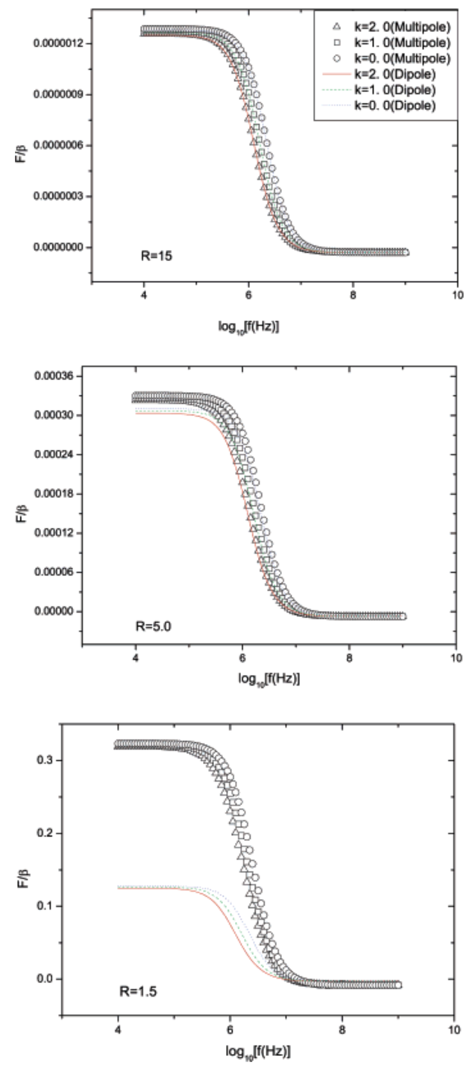

Figure 3. Dielectrophoretic spectra given by multipole and dipole forces, F_M [eq 20] and F_D [eq 21]. The forces are both denoted as F and normalized by the constant β, for different k and R. Parameter: $A = 0$.

20, which was obtained from the first-principles approach, is actually valid for the arbitrary gradation profile as long as the radial equation can be solved, at least numerically.

In summary, we have presented a first-principles approach to dielectrophoresis of an inhomogeneous particle under an inhomogeneous field of an oscillating electric dipole moment. We have shown that both the multipolar interaction and spatial fluctuations inside the particle can affect the dielectrophoretic spectrum of the particle significantly.

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Appendix: Dielectrophoresis of an Inhomogeneous Colloidal Particle under an Inhomogeneous Field of a Source Point Charge

Similarly, let us consider a graded spherical particle of radius with complex dielectric constant profile $\tilde{\epsilon}_1(r)$, embedded in a host fluid of dielectric constant $\tilde{\epsilon}_2$. Here $\tilde{\epsilon} = \epsilon + \sigma/(2\pi i f)$, where ϵ denotes real part of the dielectric constant, σ stands for conductivity, and f is the frequency of an external field, and $i = (-1)^{1/2}$.

A. Polarization Due to a Point Charge. Suppose that a point charge $q(t)$ [which is a function of time t, $q(t) = q_0 \sin(2\pi f t)$] is a distance R from the center of the inhomogeneous spherical particle with radius a and dielectric gradation profile $\tilde{\epsilon}_1(r)$, $r \leq a$. Let us place the point charge on the z axis at $z = R$, so that the problem has azimuthal symmetry. The Maxwell equations reads

$$\nabla\cdot\mathbf{D} = \rho \tag{24}$$

$$\nabla \times \mathbf{E} = 0 \tag{25}$$

In eq 24, the free charge density ρ is due to electrical conduction: $\mathbf{J} = \sigma\mathbf{E}$, where $\mathbf{J}$ is the free current density. By invoking the equation of continuity $\nabla\cdot\mathbf{J} + \partial\rho/\partial t = 0$ or $\nabla\cdot\mathbf{J} =$

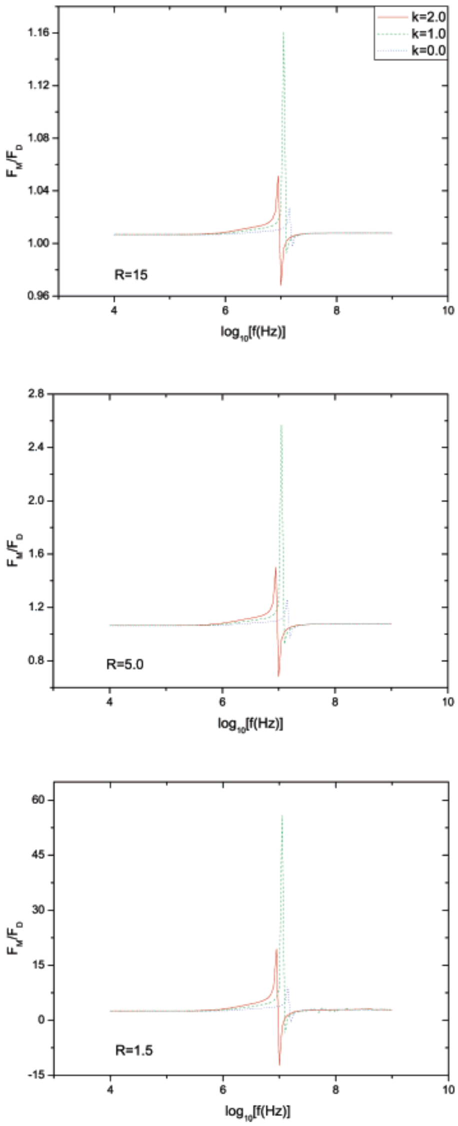

Figure 4. F_M/F_D as a function of the frequency of external electric fields for different k and R. Parameter: $A = 0$.

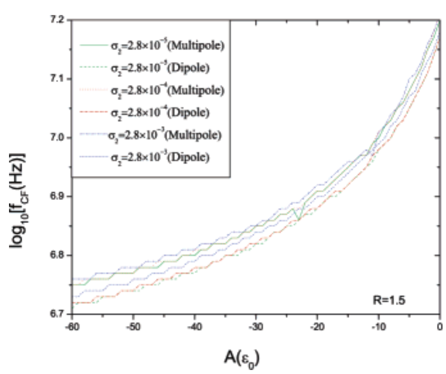

Figure 5. Crossover frequency f_{CF} versus A for various medium conductivities σ_2. Parameters: $k = 0$ and $R = 1.5$.

$-2\pi i f\rho$ due to the sinusoidal variation of the physical quantities, we arrive at

$$\nabla\cdot\left(\frac{\sigma(r)}{-2\pi i f}\mathbf{E}\right) = \rho = \nabla\cdot\epsilon(r)\mathbf{E} \qquad (26)$$

Moreover, from eq 25, $\mathbf{E}$ can be written as the gradient of a scalar potential Φ, $\mathbf{E} = -\nabla\Phi$, yielding a partial differential equation

$$\nabla\cdot[\tilde{\epsilon}(r)\nabla\Phi] = 0 \qquad (27)$$

where $\tilde{\epsilon}(r) = \tilde{\epsilon}_1(r)/\tilde{\epsilon}_2$ in the particle and $\tilde{\epsilon}(r) = 1$ in the host.

In the spherical coordinates (r,θ,φ), Φ satisfies

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\tilde{\epsilon}(r)\frac{\partial\Phi}{\partial r}\right) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\,\tilde{\epsilon}(r)\frac{\partial\Phi}{\partial r}\right) + \frac{1}{r^2\sin^2\theta}\frac{\partial}{\partial\varphi}\left(\tilde{\epsilon}(r)\frac{\partial\Phi}{\partial\varphi}\right) = 0 \qquad (28)$$

As the external field is applied along the z axis, Φ is independent of the azimuthal angle φ. To separate the variables, we set $\Phi = f(r)\,\Theta(\theta)$. For the radial function $f(r)$, we obtain a radial equation

$$\frac{\mathrm{d}}{\mathrm{d}r}\left(r^2\tilde{\epsilon}(r)\frac{\mathrm{d}f(r)}{\mathrm{d}r}\right) - l(l+1)\tilde{\epsilon}(r)f(r) = 0 \qquad (29)$$

where l is an integer. The radial equation [eq 29] yields two solutions $f_l^+(r)$ and $f_l^-(r)$ that are regular at the origin and infinity, respectively. In addition, $\Theta(\theta)$ satisfies the Legendre equation (19).

By virtue of the regularity of the solution at $r = 0$ and $r \to \infty$, the potentials are given by

$$\Phi_1(r,\theta) = \sum_{l=0}^{\infty} A_l f_l^+(r)P_l(\cos\theta), \quad r \le a \qquad (30)$$

$$\Phi_2(r,\theta) = \frac{q(t)}{4\pi\tilde{\epsilon}_2|\mathbf{r} - R\hat{\mathbf{z}}|} + \sum_{l=0}^{\infty} B_l r^{-(l+1)}P_l(\cos\theta), \quad r \ge a \qquad (31)$$

To apply the boundary conditions, we expand the Coulomb potential [in eq 31] due to the point charge $q(t)$ using[19]

$$\frac{q(t)}{4\pi\tilde{\epsilon}_2|\mathbf{r} - R\hat{z}|} = \frac{q(t)}{4\pi\tilde{\epsilon}_2}\sum_{l=0}^{\infty}\frac{r^l}{R^{l+1}}P_l(\cos\theta) \qquad (32)$$

Next, in view of the boundary conditions

$$\Phi_1|_{r=a} = \Phi_2|_{r=a} \qquad (33)$$

and

$$\tilde{\epsilon}_1(a)\left(\frac{\partial\Phi_1}{\partial r}\right)_{r=a} = \tilde{\epsilon}_2\left(\frac{\partial\Phi_2}{\partial r}\right)_{r=a} \qquad (34)$$

we have a set of simultaneous linear equations for the coefficients A_l and B_l

$$A_l f_l^+(a) = \frac{q(t)}{4\pi\tilde{\epsilon}_2}\frac{a^l}{R^{l+1}} + \frac{B_l}{a^{l+1}} \qquad (35)$$

$$A_l\frac{\tilde{\epsilon}_1(a)}{\tilde{\epsilon}_2}\left[\frac{\mathrm{d}f_l^+(r)}{\mathrm{d}r}\right]_{r=a} = \frac{q(t)}{4\pi\tilde{\epsilon}_2}\frac{la^{l-1}}{R^{l+1}} - B_l(l+1)a^{-(l+2)} \qquad (36)$$

Solving for A_l and B_l, we obtain

$$A_l = \frac{q(t)}{4\pi\tilde{\epsilon}_2}\frac{a^l}{R^{l+1}}\frac{2l+1}{f_l^+(a)[l(F_l+1)+1]} \qquad (37)$$

$$B_l = -\frac{q(t)}{4\pi\tilde{\epsilon}_2}\frac{a^{2l+1}}{R^{l+1}}\frac{l(F_l-1)}{l(F_l+1)+1} \tag{38}$$

where the lth order equivalent complex dielectric constant F_l of the graded particle is given by

$$F_l = \frac{\tilde{\epsilon}_1(a)}{\tilde{\epsilon}_2}\frac{a}{l}\frac{1}{f_l^+(a)}\left[\frac{\mathrm{d}}{\mathrm{d}r}f_l^+(r)\right]_{r=a} \tag{39}$$

So far, the electric field at the point $\mathbf{r} = R\hat{\mathbf{z}}$ due to the induced polarization may be expressed as

$$E_{\mathbf{r}=R\hat{\mathbf{z}}} = -\left(\frac{\partial\Phi_2}{\partial r}\right)_{\mathbf{r}=R\hat{\mathbf{z}}} = \sum_{l=0}^{\infty}B_l\frac{l+1}{R^{l+2}} \tag{40}$$

It is straightforward to obtain the time-dependent force $\mathbf{F}_q(t)$ acting on the charge, according to $\mathbf{F}_q(t) = \hat{\mathbf{z}}q(t)E_{\mathbf{r}=R\hat{\mathbf{z}}}$

$$\mathbf{F}_q(t) = -\hat{\mathbf{z}}\frac{q(t)^2}{4\pi\tilde{\epsilon}_2}\sum_{l=0}^{\infty}\frac{a^{2l+1}}{R^{2l+3}}\frac{(F_l-1)l(l+1)}{(F_l+1)l+1} \tag{41}$$

For large R, the force $\mathbf{F}_q(t)$ falls off as R^{-5}. For a homogeneous dielectric particle, namely $\tilde{\epsilon}_1(r) = \tilde{\epsilon}_1$ is a constant, solving eq 29 yields $f_l^+(a) = Ca^l$ (C = a constant), and hence we obtain $f_l^{+'}(a)/f_l^+(a) = l/a$. The substitution of this into eq 39 yields $F_L = \tilde{\epsilon}_1/\tilde{\epsilon}_2$. Thus, in this case, eq 41 reduces to the known expression for the force F_0 acting on a charge q due to a homogeneous particle

$$\mathbf{F}_0 = -\hat{\mathbf{z}}\frac{q^2}{4\pi\tilde{\epsilon}_2}\sum_{l=0}^{\infty}\frac{a^{2l+1}}{R^{2l+3}}\frac{(\tilde{\epsilon}_1/\tilde{\epsilon}_2-1)l(l+1)}{(\tilde{\epsilon}_1/\tilde{\epsilon}_2+1)l+1} \tag{42}$$

As $F_q(t)$ can be expressed in terms of F_l, which involves the product of the ratio $f_l^{+'}(a)/f_l^+(a)$ (here $f_l^{+'}(a) \equiv \mathrm{d}[f_l^+(r)]/\mathrm{d}r|_{r=a}$) and $\tilde{\epsilon}_1(a)$ at the spherical surface only (see eq 39). We may solve the radial equation [eq 29] numerically with some boundary conditions at $r = 0$. Since only the ratio $f_l^{+'}(a)/f_l^+(a)$ is needed, the actual value of $f_l^{+'}(0)$ is unimportant. One can assign any constant value without affecting the solution. Also, it is worth noting that there is $f_l^+(0) = 0$ always for the interior solution.

B. Dielectrophoretic Force Acting on the Graded Particle. Based on eq 41, we take one step forward to express the multipole time-average dielectrophoretic force $\mathbf{F}_M$ acting on the graded particle as

$$\boldsymbol{F}_M = \frac{\hat{\mathbf{z}}q_0^2}{8\pi\epsilon_2}\sum_{l=0}^{\infty}\mathrm{Re}\left[\frac{a^{2l+1}}{R^{2l+3}}\frac{(F_l-1)l(l+1)}{(F_l+1)l+1}\right] \tag{43}$$

Here Re[$\cdots$] means the real part of $\cdots$. Note, according to Newton's third law (i.e., action−reaction principle), the dielectrophoretic force acting on the particle should be in the direction opposite to $\mathbf{F}_q(t)$. In the first term of the right-hand side of eq 43, we have to include ϵ_2 (the real part of $\tilde{\epsilon}_2$) rather than $\tilde{\epsilon}_2$. The proof has been given by Jones[14] by considering the integral of the Maxwell stress tensor over the particle surface. In addition, we have used the time average $\langle q(t)^2\rangle = q_0^2/2$.

As $l = 1$, we obtain the dipole dielectrophoretic force $\boldsymbol{F}_D$

$$\mathbf{F}_D = \frac{\hat{\mathbf{z}}q_0^2}{4\pi\epsilon_2}\frac{a^3}{R^5}\mathrm{Re}\left[\frac{F_1-1}{F_1+2}\right] \tag{44}$$

C. Crossover Frequencies. Crossover frequencies f_{CF} is the frequency at which the dielectrophoretic force is zero. Thus, f_{CF} can also be obtained by solving $F_M = 0$ and $F_D = 0$, for multipole and dipole contribution, respectively.

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