

Dielectrophoresis of charged colloidal suspensions

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We present a theoretical study of dielectrophoretic (DEP) crossover spectrum of two polarizable particles under the action of a nonuniform ac electric field. For two approaching particles, the mutual polarization interaction yields a change in their respective dipole moments, and hence, in the DEP crossover spectrum. The induced polarization effects are captured by the multiple image method. Using spectral representation theory, an analytic expression for the DEP force is derived. We find that the mutual polarization effects can change the crossover frequency at which the DEP force changes sign. The results are found to be in agreement with recent experimental observation and as they go beyond the standard theory, they help to clarify the important question of the underlying polarization mechanisms.

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I. INTRODUCTION

When a polarizable particle is subjected to an applied electric field, a dipole moment is induced into it. The movement of colloidal particles in an applied ac electric field is called dielectrophoresis [1]. It is typically used for micromanipulation and separation of biological cellular size particles, 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 [2], viruses [3], latex spheres [4,5], DNA [6] and leukemic cells [7], as well as lab-on-a-chip designs [8].

The dielectrophoretic (DEP) 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 DEP 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 DEP analysis for submicrometer particles [3,4].

In the dilute limit, i.e., when a small volume fraction of charged particles are suspended in an aqueous electrolyte solution, one can focus on the DEP spectrum of an individual particle ignoring the mutual interactions between the particles. Although the current theory [1] captures some of the essential physics in the dilute case, it is not adequate [9–13]. This is due to the fact that even for a single colloidal particle in an electrolyte, it is not established that which mechanisms

control its dielectric properties. If the suspension is not dilute, the situation becomes even more complicated due to the mutual interactions between the particles. One should also note that particles may aggregate due to the presence of an external field, even when the suspension is at the dilute limit under zero field conditions. In this case, the mutual interactions have to be included in the description.

In this article, we present a theoretical study of the DEP spectrum of two spherical particles in the presence of a non-uniform ac electric field. We use the multiple image method [14], which is able to capture the mutual polarization effects. Using the spectral representation theory [15], we derive an analytic expression for the DEP force and determine the crossover frequency. Our theoretical analysis shows that the induced mutual polarization interactions plays an important role in DEP spectrum. In a more general framework, our results demonstrate the importance of correlation effects. This is analogous to the findings in charged systems, where phenomena such as overcharging, or charge inversion (see, e.g., Refs. [16,17]), provide spectacular demonstrations of correlation effects.

As our starting point, we consider a pair of interacting charged colloidal particles dispersed in an electrolyte solution. When the two particles approach each other, the mutual polarization interaction between them leads to changes in their respective dipole moments [18], and hence also in the DEP spectrum and crossover frequency. We analyze two cases: (1) longitudinal field (L), in which the field is parallel to the line joining the centers of particles, and (2) transverse field (T) in which the field is perpendicular. The former corresponds to positive dielectrophoresis, where a particle is attracted to regions of high field and the latter to the opposite case, referred to as negative dielectrophoresis.

This paper is organized as follows. In Sec. II we present the formalism and derive analytic expressions for the effective dipole factors in spectral representation. In Sec. III, we use the analytical results to numerically solve the crossover frequency, dispersion strength and DEP spectra under different conditions. This is followed by a discussion of the results in Sec. IV.

II. FORMALISM AND ANALYSIS

First, we consider a single charged spherical particle suspended in an electrolyte and subjected to a nonuniform ac electric field. The DEP force \mathbf{F}_{DEP} acting on the particle is then given by [19]

$$\mathbf{F}_{\text{DEP}} = \frac{1}{4} \pi \epsilon_2 D^3 \text{Re}[b] \nabla |\mathbf{E}|^2, \quad (1)$$

where D is particle diameter, ϵ_2 the real dielectric constant of host medium, \mathbf{E} the local RMS electric field, and $\text{Re}[b]$ the real part of the dipole factor (also called Clausius-Mossotti factor)

$$b = \frac{\tilde{\epsilon}_1 - \tilde{\epsilon}_2}{\tilde{\epsilon}_1 + 2\tilde{\epsilon}_2}. \quad (2)$$

Here, $\tilde{\epsilon}_1$ and $\tilde{\epsilon}_2$ are the complex dielectric constants of the particle and the host medium, respectively. In order for the two above equations to be valid in an ac field, the dielectric constant must include dependence on the frequency. The complex frequency dependent dielectric constant is defined as

$$\tilde{\epsilon} = \epsilon + \frac{\sigma}{i2\pi f},$$

where ϵ is the real dielectric constant, σ denotes conductivity, f the frequency of the external field, and $i \equiv \sqrt{-1}$.

The conductivity of a particle consists of three components: Its bulk conductivity (σ_{bulk}), surface effects due to the movement of charge in the diffuse double layer (conductance k_d), and the Stern layer conductance (k_s), i.e.,

$$\sigma_1 = \sigma_{\text{bulk}} + \frac{4k_d}{D} + \frac{4k_s}{D}. \quad (3)$$

The diffuse double layer conductance k_d can be given as [20]

$$k_d = \frac{4F_a^2 c z^2 \Xi (1 + 3\Lambda/z^2)}{R_0 T_0 \kappa} \left[\cosh\left(\frac{zF_a \zeta}{2R_0 T_0}\right) - 1 \right], \quad (4)$$

where Ξ is the ion diffusion coefficient, z the valency of counterions, F_a the Faraday constant, R_0 the molar gas constant, ζ the electrostatic potential at the boundary of the slip plane and T_0 the temperature. The reciprocal Debye length κ providing a measure for screening on the system is given by

$$\kappa = \sqrt{\frac{2czF_a^2}{\epsilon_2 R_0 T_0}}, \quad (5)$$

where c is the electrolyte concentration. Parameter Λ in Eq. (4) describes the electro-osmotic contribution to k_d , and it is given by

$$\Lambda = \left(\frac{R_0 T_0}{F_a}\right)^2 \frac{2\epsilon_2}{3\eta\Xi}, \quad (6)$$

where η is the viscosity of medium. In addition, the Stern layer conductance k_s has the form [11]

$$k_s = \frac{u\mu_r \Sigma}{2zF_a}, \quad (7)$$

where u is the surface charge density, Σ molar conductivity for a given electrolyte, and μ_r gives the ratio between the ion mobility in the Stern layer to that in the medium.

For a pair of particles at a separation R suspended in an electrolyte, we have to consider the multiple image effect. We consider two spheres in a medium, and apply an uniform electric field $\mathbf{E}_0 = E_0 \hat{z}$ to the suspension. This induces a dipole moment into each of the particles. The dipole moments of particles 1 and 2 are given by p_{10} and $p_{20} (\equiv p_{10} = \epsilon_2 E_0 D^3 b/8)$, respectively.

Next, we include the image effects. The dipole moment p_{10} induces an image dipole p_{11} into sphere 2, while p_{11} induces another image dipole in sphere 1. As a result, multiple images are formed. Similarly, p_{20} induces an image p_{21} into colloid 1. The formation of multiple images leads to an infinite series of image dipoles.

In the following, we obtain the sum of dipole moments inside each particle, and derive the desired expressions for dipole factors. We consider two basic cases: (1) longitudinal field (L), where the field is parallel to the line joining the centers of particles, and (2) transverse field (T), where the field is perpendicular to the line joining the centers of particles. Using the above notation, the effective dipole factors for a pair are given by [14]

$$b_L^* = b \sum_{n=0}^{\infty} (2b)^n \left[\frac{\sinh \alpha}{\sinh(n+1)\alpha} \right]^3, \quad (8)$$

$$b_T^* = b \sum_{n=0}^{\infty} (-b)^n \left[\frac{\sinh \alpha}{\sinh(n+1)\alpha} \right]^3,$$

where α is defined via $\cosh \alpha = R/D$. The summations in Eqs. (8) include the multiple image effects, the $n=0$ term giving the dipole factor of an isolated particle.

We have to derive the analytic expressions for $\text{Re}[b_L^*]$ and $\text{Re}[b_T^*]$ to resolve the DEP force in Eq. (1). To do that, we resort to spectral representation theory. It offers the advantage of being able to separate the material parameters (such as dielectric constant and conductivity) from structural information [15] in a natural way.

Let us begin by defining a complex material parameter $\tilde{s} = 1/(1 - \tilde{\epsilon}_1/\tilde{\epsilon}_2)$. Using this, the dipole factor for a pair takes the form

$$b^* = \sum_{n=1}^{\infty} \frac{F_n}{\tilde{s} - s_n}, \quad (9)$$

where n is a positive integer, and F_n and s_n are the n th microstructure parameters of the composite material [15]. As an example, the dipole factor of an isolated particle in spectral representation expression becomes $b = F_1/(\tilde{s} - s_1)$, where $F_1 = -1/3$ and $s_1 = 1/3$.

In order to obtain expressions for the dipole factors b_L^* and b_T^* in Eqs. (8), we introduce the following identity:

$$\frac{1}{\sinh^3 x} = \sum_{m=1}^{\infty} 4m(m+1) \exp[-(1+2m)x].$$

Its application into Eqs. (8) yields the exact transformations,

$$b_L^* = \sum_{m=1}^{\infty} \frac{F_m^{(L)}}{\tilde{s} - s_m^{(L)}}, \quad (10)$$

$$b_T^* = \sum_{m=1}^{\infty} \frac{F_m^{(T)}}{\tilde{s} - s_m^{(T)}},$$

where the m th components of the microstructure parameter of the composite material are given as

$$F_m^{(L)} \equiv F_m^{(T)} = -\frac{4}{3} m(m+1) \sinh^3 \alpha \exp[-(2m+1)\alpha],$$

$$s_m^{(L)} = \frac{1}{3} \{1 - 2 \exp[-(1+2m)\alpha]\},$$

$$s_m^{(T)} = \frac{1}{3} \{1 + \exp[-(1+2m)\alpha]\}.$$

To make this approach more tractable, we introduce dimensionless dielectric constant and conductivity [21], $s = 1/(1 - \epsilon_1/\epsilon_2)$ and $t = 1/(1 - \sigma_1/\sigma_2)$, respectively. Now, we are able to separate the real and imaginary parts of the arguments in the expressions for b_L^* and b_T^* in Eq. (10). The argument can be rewritten as

$$\frac{F_m}{\tilde{s} - s_m} = \left(\frac{F_m}{s - s_m} + \frac{\Delta \epsilon_m}{1 + f^2/f_{mc}^2} \right) - i \frac{\Delta \epsilon_m f/f_{mc}}{1 + f^2/f_{mc}^2}, \quad (11)$$

where

$$\Delta \epsilon_m = F_m \frac{s - t}{(t - s_m)(s - s_m)} \quad (12)$$

and

$$f_{mc} = \frac{1}{2\pi} \frac{\sigma_2 s (t - s_m)}{\epsilon_2 t (s - s_m)}. \quad (13)$$

The analytic expressions for $\text{Re}[b_L^*]$ and $\text{Re}[b_T^*]$ [Eq. (10)] become

$$\text{Re}[b_L^*] = \sum_{m=1}^{\infty} \left(\frac{F_m^{(L)}}{s - s_m^{(L)}} + \frac{\Delta \epsilon_m^{(L)}}{1 + f^2/f_{mc}^{(L)2}} \right), \quad (14)$$

$$\text{Re}[b_T^*] = \sum_{m=1}^{\infty} \left(\frac{F_m^{(T)}}{s - s_m^{(T)}} + \frac{\Delta \epsilon_m^{(T)}}{1 + f^2/f_{mc}^{(T)2}} \right).$$

Using these, we can obtain the DEP force \mathbf{F}_{DEP} which includes corrections due to the image effects. The DEP spectrum consists of a series of subdispersions with strength $\Delta \epsilon_m$ and characteristic frequency f_{mc} . In particular, the frequency which yields $F = 0$, namely, $\text{Re}[b^*] = 0$, is the desired crossover frequency f_{CF} .

III. NUMERICAL RESULTS

The above formalism enables us to study the effects due to multiple images under different physical conditions and to compare the theory to experimental results. In the following, we compare the crossover frequency of an isolated particle to that of two particles at different separations. We study the effects due to multiple images by varying medium conductivity, the ζ potential, medium viscosity, surface charge density, real dielectric constant of the particle and molar conductivity. Finally, we have computed the DEP spectrum and the dispersion strength.

The common parameters used in all numerical computations are the following: Temperature $T_0 = 293$ K, dielectric constant of host medium $\epsilon_2 = 78\epsilon_0$, bulk conductivity of the colloidal particle $\sigma_{1\text{bulk}} = 2.8 \times 10^{-4}$ S/m, ion diffusion coefficient $\Xi = 2.5 \times 10^{-9}$ m²/s, the ratio between the ion mobility in the Stern layer to that in the medium $\mu_r = 0.35$, particle diameter $D = 2.16 \times 10^{-7}$ m, counterion valency $z = 1$. The dielectric constant of vacuum is denoted by ϵ_0 .

Figure 1 shows the DEP crossover frequency as a function of medium conductivity for an isolated particle and for two particles at different separations. In agreement with recent experiments [22], we find that a peak in the crossover frequency appears at a certain medium conductivity. The appearance of a peak is preceded by an increase of f_{CF} upon increasing medium conductivity and followed by an abrupt drop [11,22]. Compared to an isolated particle, the multiple image effect leads to a redshift (blue shift) in f_{CF} in the longitudinal (transverse) field case. Furthermore, for longitudinal (transverse) field, the stronger the polarization interaction, the lower (higher) the crossover frequency. In addition, it is worth noting that the effect of the multiple images is the opposite in the longitudinal and transverse cases. As the ratio R/D grows, the predicted crossover spectrum approaches to that of an isolated particle, i.e., at large separations the multiple image interaction becomes negligible.

Motivated by a recent experiment [13], we analyzed the effect of particle size on the crossover frequency by keeping the ratio R/D fixed and varying the particle diameter. In agreement with the experiments, we find that the location of the peak is shifted to higher frequencies and higher conductivities when the diameter of the particle is reduced, see Fig. 2.

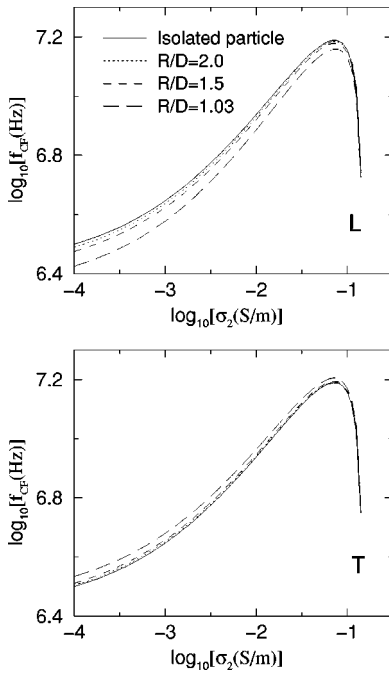


FIG. 1. DEP crossover frequency vs medium conductivity for an isolated particle (solid line) and two particles at different separations. *L* denotes longitudinal field case and *T* transverse field case. Parameters: $\zeta = 0.12$ V, $\eta = 1.0 \times 10^{-3}$ Kg/(ms), $u = 0.033$ C/m², $\Sigma = 0.014$ Sm²/mol, $\epsilon_1 = 2.25\epsilon_0$.

Figure 3 displays the effect of the ζ potential. It has been experimentally observed by Hughes and Green [11] that decreasing the ζ potential may redshift the DEP crossover frequency. The system used by them contained many latex beads suspended in a solution, and hence the multipolar in-

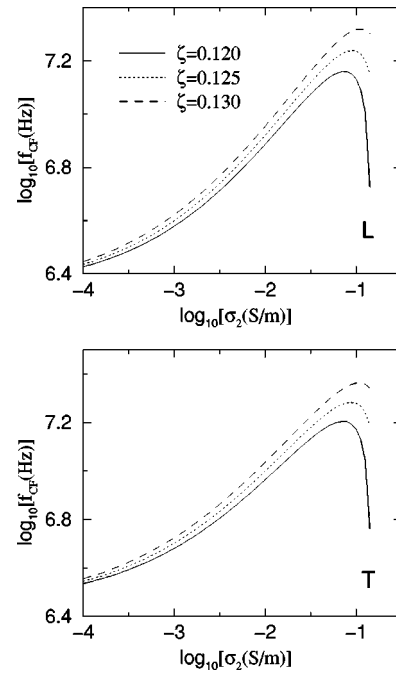


FIG. 3. DEP crossover frequency vs medium conductivity for different ζ potentials. Parameters as in Fig. 1.

teraction is expected to play a role. Our results are in qualitative agreement with the above experimental findings. Furthermore, an increase in the ζ potential leads to higher f_{CF} in both the longitudinal and transverse field cases. Similarly, increasing the real part of the dielectric constant leads to an increase in f_{CF} , as displayed in Fig. 4. Increasing the viscosity of the medium (figure not shown here), however, has exactly the opposite effect for both the longitudinal and transverse field cases.

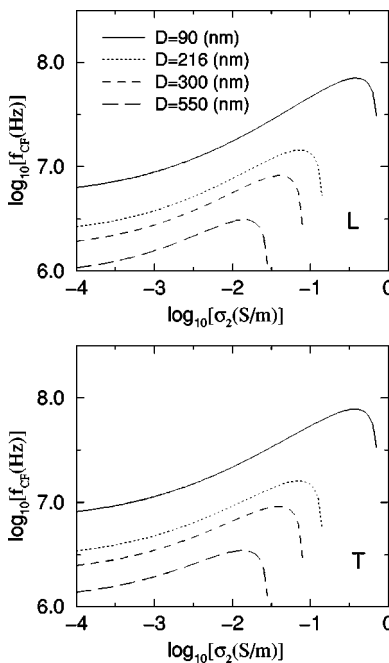


FIG. 2. DEP crossover frequency vs medium conductivity when the particle size is varied. Parameters as in Fig. 1.

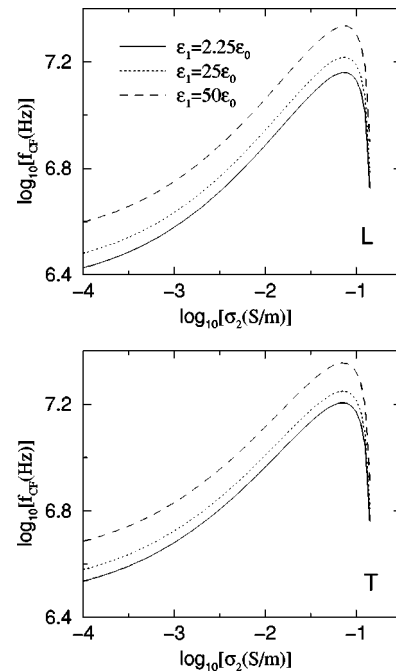


FIG. 4. DEP crossover frequency vs medium conductivity. The real part of the dielectric constant is varied. Parameters as in Fig. 1.

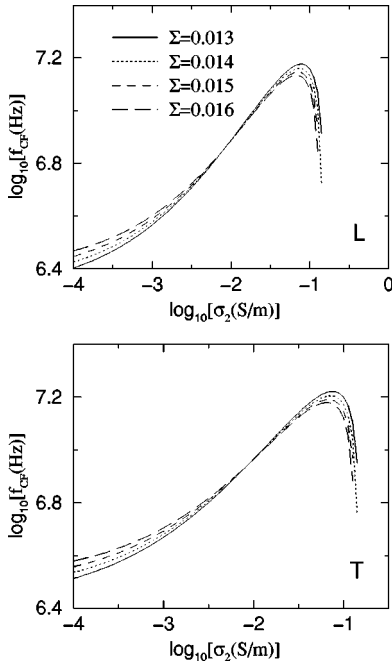


FIG. 5. The effect of molar conductivity on the DEP crossover frequency. Parameters as in Fig. 1.

Figure 5 shows the effect of molar conductivity Σ on crossover frequency. For small medium conductivities (here, $\sigma_2 < 10^{-2}$ S/m), increasing Σ leads to an increase in the crossover frequency. However, there is a crossover after which lower values of Σ yield higher f_{CF} . Similar behavior for the low surface conductivity regime has been observed in experiments [11], but the authors are not aware of any systematic study of the molar conductivity on f_{CF} . As Fig. 5 shows, the effect is similar for both longitudinal and transverse fields.

Figure 6 shows the effect of varying the surface charge density on the crossover frequency. Variations in the surface charge density lead to more pronounced effects in the low frequency region, but close to the peak the variations differences are very small. In addition, the location of the peak is only weakly dependent on surface charge density. These results are in agreement with the experimental observations of Green and Morgan [13].

In Fig. 7, we investigate the real part of the dipole factor, and thus the DEP force. The figure shows that the effect due to multiple image plays an important role at low frequency region when the particles separation is not large, whereas its effect is smaller in the high frequency region. In the low frequency region, the DEP force is to be enhanced (reduced) due to the presence of multiple images for longitudinal (transverse) field case. As the particle separation grows, the multiple image effect becomes negligible as expected. We also studied the effect of particle size on the real part of the dipole factor and the effect of multiple images increases as the particle size decreases, and the effect is stronger in the longitudinal field case.

Finally, in Fig. 8, we plot the dispersion strengths ($\Delta\epsilon_m^{(L)}$ and $\Delta\epsilon_m^{(T)}$) as a function of the characteristic frequencies

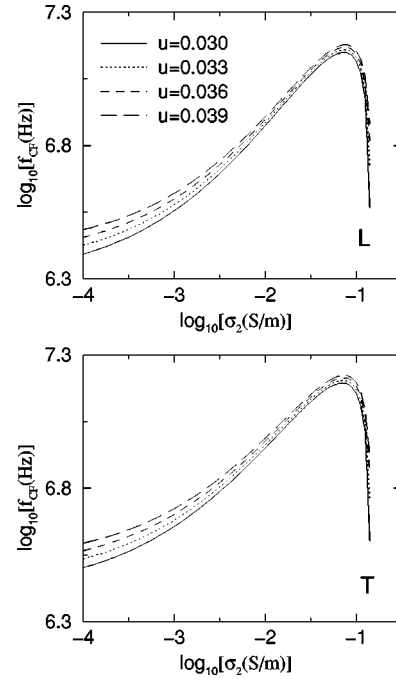


FIG. 6. The effect of surface charge density on the DEP crossover frequency. Parameters as in Fig. 1.

($f_{mc}^{(L)}$ and $f_{mc}^{(T)}$), for $m = 1$ to 100 with different medium conductivities σ_2 . Here, m is a positive integer, and F_m and s_m are the microstructure parameters of the composite material, see Eqs. (11)–(13). Hence, $\Delta\epsilon_m$ and f_{mc} are the m th dispersion strength and characteristic frequency due to the presence of multiple images as discussed in Sec. II.

The advantage of using the spectral representation theory is shown in Fig. 8. Based on Fig. 7, it may appear that only

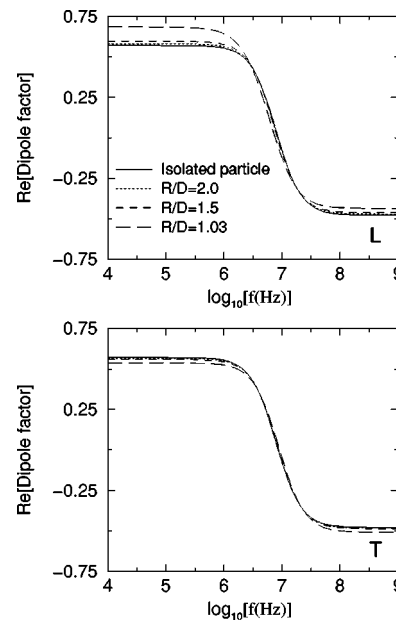


FIG. 7. DEP spectrum (the real part of the dipole factor). Parameters as in Fig. 1.

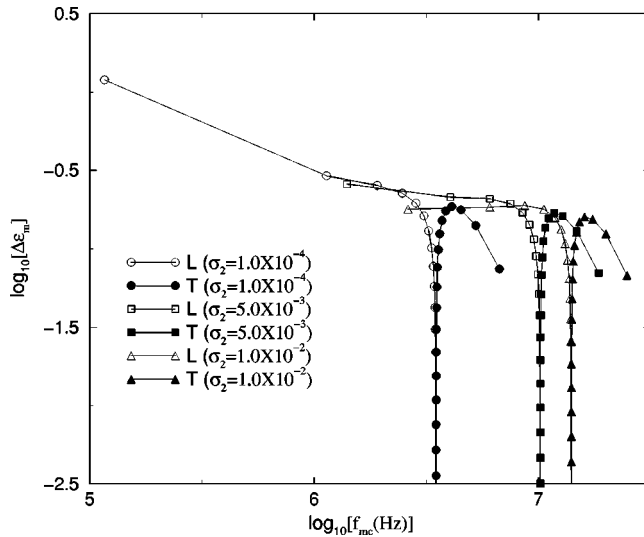


FIG. 8. Dispersion strength vs the characteristic frequency for different medium conductivities. Parameters: $\zeta=0.12\text{V}$, $\eta=1.0 \times 10^{-3} \text{ Kg}/(\text{ms})$, $R/D=1.03$, $\epsilon_1=2.25\epsilon_0$, $u=0.033 \text{ C}/\text{m}^2$, $\Sigma=0.014 \text{ Sm}^2/\text{mol}$. The lines are drawn as a guide to the eye.

one dispersion exist. Figure 8 shows, however, that subdispersions with strength $\Delta\epsilon_m$ and characteristic frequency f_{mc} coexist, and most of them lie close to the main dispersion. Thus, the spectral representation theory helps us to gain more detailed information about the system and it provides a detailed comparison between the longitudinal and transverse field cases.

At a given σ_2 , for the longitudinal (transverse) field case, increasing m leads to corresponding subdispersions in the characteristic frequency due to the presence of multiple images. The crossover frequencies f_c are $3.49 \times 10^6 \text{ Hz}$, $1.0 \times 10^7 \text{ Hz}$, and $1.4 \times 10^7 \text{ Hz}$ (Fig. 8). From Fig. 8 we find that at a lower medium conductivity (say, $\sigma_2=1.0 \times 10^{-4} \text{ S}/\text{m}$), multiple images have a stronger effect on the DEP spectrum for the longitudinal field case than for the transverse field. This is also apparent in Fig. 7 as well. Moreover, for longitudinal field case the multiple images play a role in the low frequency range (i.e., smaller than f_c). For the transverse field the situation is the opposite. At a larger σ_2 (say, $\sigma_2=5.0 \times 10^{-3} \text{ S}/\text{m}$ or $\sigma_2=1.0 \times 10^{-2} \text{ S}/\text{m}$), the subdispersion strengths for the two cases have only a minor difference. These observations may partly explain the results of Green and Morgan [13] whose data suggests that there exists a dispersion below the frequencies predicted by the current theory. The importance of these observation lies in the fact that they help to clarify the interesting question of which polarization mechanisms are present.

IV. DISCUSSION AND CONCLUSION

In this study, we have investigated the crossover spectrum of two approaching polarizable particles in the presence of a nonuniform ac electric field. When the two particles approach, the mutual polarization interaction between the particles leads to changes in the dipole moments of each of the individual particles, and hence in the DEP crossover spectrum. This can be interpreted as a correlation effect analogous to those seen in charged systems [16].

For charged particles, there is a coexistence of an electrophoretic and a dielectrophoretic force in the presence of a nonuniform ac electric field. The DEP force always points towards the region of high field gradient. It does not oscillate with the change of direction of the field. In contrast, the electrophoretic force points along the direction of field, and hence is oscillatory under the same conditions. How to separate the DEP force from the electrophoretic force is a question of interest in many experimental setups [6,23]. In different frequency ranges, either the electrophoretic force or the DEP force dominates, and the transition from one to the other occurs at a frequency f_{tr} , which has been approximately determined [4]. Here, we have chosen a frequency region where electrophoretic effects are negligible and the DEP force dominates. In addition, although we are at finite temperature, Brownian motion is not included in our analysis. In experiments Brownian motion is always present and has posed difficulties in dielectrophoresis of submicrometer particles. However, with current techniques it is possible to access also this range [4,5].

One of the interesting questions is what happens, when the volume fraction of the suspension becomes large. It turns out that it is possible to extend our approach by taking into account local field effects which may modify the DEP crossover spectrum. Work is in progress to address these questions. In addition to dielectrophoresis, the extension of the present approach is also of interest from the point of view of electrorotation.

To summarize, using the multiple image method, we have been able to capture mutual polarization effects of two approaching particles in an electrolyte. Using spectral representation theory, we derived an analytic expression for the DEP force, and using that the crossover frequency was determined. From the theoretical analysis, we find that the mutual polarization effects can change the crossover frequency substantially.

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