Theory of the dielectrophoretic behavior of clustered colloidal particles in two dimensions

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Abstract

Clustering behaviors in real colloidal suspensions can often be seen. In such clusters, the separation between the suspended particles is so close that we cannot neglect the multiple image effect on the dielectrophoresis of the particles. Since the exact multiple image method, so far, exists in two-dimensional cases, rather than in three-dimensional cases, as an initial model we have investigated the cluster colloidal suspension, in which many circular or cylindrical particles are randomly distributed in a sheet cluster. We have applied the multiple image method and the Maxwell–Garnett approximation (MGA) to study the dielectrophoretic (DEP) spectrum of the cluster. The dipole factor of a particle in the cluster has been exactly expressed in the spectral representation, thus simplifying the study. To one's interest, the multiple image method has been found to be in a good agreement with the MGA, even though they were founded in the light of different physical thoughts. To this end, it is found that the multiple image method predicts two characteristic frequencies, at which the dielectric dispersion occurs in the DEP spectrum. By choosing the appropriate material or geometrical parameters, the lower characteristic frequency can be achieved. © 2004 Elsevier B.V. All rights reserved.

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1. Introduction

The movement of a polarizable particle which is subjected to an applied ac electric field is called dielectrophoresis [1]. In fact, dielectrophoresis is caused by the interaction between the field-induced dipole moment inside the particle and the external electric field. By using dielectrophoresis and electrorotation, one may monitor the dielectric properties of dispersions of colloids or biological cells. It is important to study their frequency-dependent responses to ac electric fields, which yields valuable information on the structural (Maxwell–Wagner) polarization...
effects [2,3]. The polarization is characterized by a variety of characteristic frequency-dependent changes known as the dielectric dispersion.

In the last two decades, various experimental tools have been developed to analyze the polarization of biological cells, such as dielectric spectroscopy [4], dielectrophoresis [5] and electrorotation [6] techniques. Among these techniques, conventional dielectrophoresis and electrorotation are usually applied to analyze the frequency dependence of translations and rotations of individual cells in an inhomogeneous and rotating electric field, respectively, [5,6]. Furthermore, one is able to monitor the cell movements by using automated video analysis [7] as well as light scattering methods [3].

In the dilute limit, the dielectrophoresis of individual particle can be predicted by ignoring the mutual interaction between the particles (see, e.g., [8], and references therein). However, the clustering behavior can often be seen in nature. In this case, the particle concentration in the cluster is high, and the separation between particles are hence quite close. In such sheet cluster, the effect of multiple images from all the other particles will be very important. In this regard, the multiple image effect has to be considered when we investigate the dielectrophoretic (DEP) spectrum. As an initial model, we have recently studied the DEP spectrum of two approaching spherical particles in the presence of a nonuniform electric field [9]. We showed that when the two particles approach and finally touch, the mutual polarization interaction between the particles leads to a change in the dipole moment of individual particles and hence the DEP spectrum, as compared to that of isolated particles, via the multiple image method [10].

As an interesting extension of Ref. [9], in the present work we shall further discuss a cluster system, in an attempt to investigate the DEP spectrum of the cluster by taking into account the multiple image effect. Fortunately, ones examined the case of circular inclusions in a conducting sheet (namely, in two dimensions) [11]. Moreover, the two-inclusion problem was solved exactly by using a multiple image method [11]. In this Letter, we shall discuss a two-dimensional case, in which many circular or cylindrical particles are randomly distributed in a sheet cluster by applying the multiple image method. For comparison, we shall also apply a Maxwell–Garnett approximation (MGA) which takes into account the local field effect.

Regarding the spectral representation approach [12], it is a rigorous mathematical formalism of the effective dielectric constant of two-phase composite materials. Also, the approach was extended to deal with three-phase materials [13]. This approach offers the advantage of the separation of material parameters, i.e., the dielectric constant and conductivity, from the geometrical information, thus simplifying the study.

2. Formalism

Let us consider the circular or circular cylindrical particles of complex dielectric constant \( \tilde{\epsilon}_1 = \epsilon_1 + \sigma_1 / i \omega \) with concentration \( p \) in a two-dimensional sheet. The complex dielectric constant of the surrounding fluid is \( \tilde{\epsilon}_2 = \epsilon_2 + \sigma_2 / i \omega \), with \( \omega = 2 \pi f \) being the angular frequency of the applied nonuniform ac electric field, and \( i = \sqrt{-1} \). Here \( \epsilon \) and \( \sigma \) denote, respectively, the real dielectric constant and conductivity.

The DEP (dielectrophoretic) force \( \mathbf{F}_{\text{DEP}} \) acting on the particle in a cluster is given by [1]

\[
\mathbf{F}_{\text{DEP}} = 2 \pi \epsilon_2 a^2 \text{Re}[\text{dipole factor}] \nabla |E_{\text{rms}}|^2,
\]

where \( a \) stands for the radius of the particle, \( E_{\text{rms}} \) the root-mean-square magnitude of the applied field, and \( \text{Re}[\text{dipole factor}] \) the real part of the dipole factor. It is worth noting that Eq. (1) refers to a two-dimensional system. To investigate the DEP spectrum of a particle, we have to first determine the dipole factor of the particle.

2.1. Isolated particle

For an isolated circular or cylindrical particle, its dipole factor has the form

\[
b = \frac{\tilde{\epsilon}_1 - \tilde{\epsilon}_2}{\tilde{\epsilon}_1 + \tilde{\epsilon}_2},
\]
Next, we express $b$ in the spectral representation, to simplify the investigation. In order to introduce the spectral representation, one introduced two real material parameters like [14]

$$s = \left( 1 - \frac{\epsilon_1}{\epsilon_2} \right)^{-1}, \quad t = \left( 1 - \frac{\sigma_1}{\sigma_2} \right)^{-1}.$$  \hspace{1cm} (3)

They are both functions of the conductivity ratio and real dielectric constant ratio, respectively.

After some manipulation, we obtain exactly [8,9,15,16]

$$b = F_1 \frac{s - t}{s - s_1} - \frac{\delta \epsilon}{1 + i \omega/\omega_c}$$

with two geometrical parameters $F_1 = -1/2$ and $s_1 = 1/2$, where the dispersion strength $(\delta \epsilon)$ and the characteristic frequency $(\omega_c)$ are, respectively, given by

$$\delta \epsilon = F_1 \frac{s - t}{s - s_1} - \frac{\sigma_2}{\epsilon_2} \frac{s(t - s_1)}{t(s - s_1)}, \quad \omega_c = \frac{\sigma_2}{\epsilon_2} \frac{s(t - s_1)}{t(s - s_1)}.$$

2.2. Particle in a cluster: multiple image effect

The effective dielectric constant of the sample to the second order in the concentration $p$ is given by [11]

$$\tilde{\epsilon}_e \tilde{\epsilon} = 1 + 2bp + 2b^2 p^2 + b^2 p^2 F(b).$$  \hspace{1cm} (5)

Here $F(b)$ is an odd function of $b$, which contains the multiple image effect from the dipole moment of all the inclusions on the effective dielectric constant, and it is given by [11]

$$F(b) = 16 \int_0^\infty \int_0^\infty \sum_{n=1}^\infty b^{2n-1} \frac{\sinh^3 \gamma \cosh \gamma}{\sinh^2 (2n+1) \gamma} d\gamma,$$

with $\gamma$ satisfying the relation $\cosh \gamma = R/2a$, where $R$ represents the center-to-center separation between the particles. To discuss the dielectrophoresis of a special particle, we obtain the dipole factor in the presence of other particles as [15,17]

$$b^* = \frac{\tilde{\epsilon}_1 - \tilde{\epsilon} e}{\tilde{\epsilon}_1 + \tilde{\epsilon} e}.$$  \hspace{1cm} (7)

Thus, the multiple image effect on the dipole factor of a particle in the cluster is included in Eq. (7).

2.3. Particle in a cluster: local-field effect

On the other hand, the effective dielectric constant $\tilde{\epsilon}_{eMGA}$ for the system of interest can be determined by the well-known MGA (Maxwell–Garnett approximation) [18]

$$p \frac{\tilde{\epsilon}_1 - \tilde{\epsilon}_2}{\tilde{\epsilon}_1 + \tilde{\epsilon}_2} = \tilde{\epsilon}_{eMGA} - \tilde{\epsilon}_2$$

$$\frac{\tilde{\epsilon}_1 - \tilde{\epsilon}_2}{\tilde{\epsilon}_1 + \tilde{\epsilon}_2} = \tilde{\epsilon}_{eMGA} + \tilde{\epsilon}_2.$$  \hspace{1cm} (8)

The MGA is an effective mean field theory. In detail, each particle is assumed to be embedded in an effective medium of the complex dielectric constant $\tilde{\epsilon}_{eMGA}$. Then, the dipole factor $b_{MGA}$ determined by the MGA is further given by [15,17]

$$b_{MGA} = \frac{\tilde{\epsilon}_1 - \tilde{\epsilon}_{eMGA}}{\tilde{\epsilon}_1 + \tilde{\epsilon}_{eMGA}}.$$  \hspace{1cm} (9)
Therefore, Eq. (9) has included the local-field effect. Similar to Eq. (4), Eq. (9) can also be exactly expressed in the spectral representation for simplifying the study, such that

$$b_{\text{MGA}} = \frac{F^{(1)}}{s-s^{(1)}} + \frac{F^{(2)}}{s-s^{(2)}} + \frac{\Delta \epsilon^{(1)}}{1+i\omega/\omega^{(1)}} + \frac{\Delta \epsilon^{(2)}}{1+i\omega/\omega^{(2)}}$$

(10)

with the poles $s^{(1)}$ and $s^{(2)}$, the residues $F^{(1)}$ and $F^{(2)}$, the dispersion strengths $\Delta \epsilon^{(1)}$ and $\Delta \epsilon^{(2)}$, and the characteristic frequencies $\omega^{(1)}$ and $\omega^{(2)}$. Obviously, two characteristic frequencies at which the dielectric dispersion appears in the DEP spectrum are predicted in Eq. (10). The above spectral parameters are, respectively, given by

$$s^{(1)} = \frac{1 - \sqrt{p}}{2}, \quad s^{(2)} = \frac{1 + \sqrt{p}}{2},$$

$$F^{(1)} = F^{(2)} = -\frac{1 - p}{4},$$

$$\Delta \epsilon^{(1)} = \frac{2(-1 + p)s(s-t)\sigma_2((1 + p - 2s)s\sigma_2 - (p + (1 - 2s)^2)i\omega^{(1)}\epsilon_2 + (1 - 2s)s\sigma_2))}{(p - (1 - 2s)^2)^2i\omega^{(1)}(\omega^{(1)} - \omega^{(2)})\epsilon_2^2},$$

$$\Delta \epsilon^{(2)} = \frac{2(-1 + p)s(s-t)\sigma_2(-(p + (1 - 2s)^2)i\omega^{(2)}\epsilon_2 + (1 + p - 2s)s\sigma_2 + 2s(-1 + 2s)i\tau)}{(p - (1 - 2s)^2)^2i\omega^{(2)}(-\omega^{(1)} + \omega^{(2)})\epsilon_2^2},$$

$$\omega^{(1)} = \frac{2s^2(1 - 2t)ie_2\sigma_2 + st(-1 + p + 2i)\epsilon_2\sigma_2 - 2\sqrt{ps^2(s-t)i^2\epsilon_2^2\sigma_2^2}}{(p - (1 - 2s)^2)^2i^2\epsilon_2^2},$$

$$\omega^{(2)} = \frac{2s^2(1 - 2t)ie_2\sigma_2 + st(-1 + p + 2i)\epsilon_2\sigma_2 + 2\sqrt{ps^2(s-t)i^2\epsilon_2^2\sigma_2^2}}{(p - (1 - 2s)^2)^2i^2\epsilon_2^2}.$$

3. Numerical results

Fig. 1 displays the spectral parameters $s^{(1)}$, $s^{(2)}$, $F^{(1)}$, $F^{(2)}$, $\omega^{(1)}$, $\omega^{(2)}$, $\Delta \epsilon^{(1)}$, and $\Delta \epsilon^{(2)}$, as a function of the concentration $p$. Note two curves in Fig. 1(b) are overlapped, which is quite different from the three-dimensional case discussed in Ref. [15]. Increasing the concentration $p$ causes $F^{(1)} = F^{(2)}$ to increase. As $p$ increases, $s^{(1)}$ increases (decreases) accordingly. For the isolated particle case, i.e., $p = 0$, it is evident to find both poles are equal to each other. In an other word, a single pole appears at $p = 0$, as predicted in Eq. (4). It is worth noting that the residues and poles satisfy the sum rules, namely $F^{(1)} + F^{(2)} = -(1 - p)/2$ and $s^{(1)} + s^{(2)} = 1$, as determined by the spectral representation. The characteristic frequency (Fig. 1(c)) and the dispersion strength (Fig. 1(d)) behaves the same as those in three dimensions which was studied in Ref. [15].

In Figs. 2 and 3, we investigate the multiple image effect on the DEP spectrum of a particle in the cluster. Obviously, there are two characteristic frequencies, as expected. In fact, the characteristic frequency for an isolated particle is quite different from the one predicted by the multiple image method (Re[$b_{\text{MGA}}$]) or local field effect (Re[$b^*$]) (see Fig. 3 below). In detail, both the multiple image effect and local field effect may strongly reduce the DEP force, when compared with the isolated particle.

From Fig. 2, it is evident that the conductivity (i.e., $i$ and $\sigma_2$) plays an important role in the DEP spectrum. In this case, the red-shift of characteristic frequencies (namely, the characteristic frequency appears at a lower frequency) may be achieved by choosing the appropriate conductivities. However, the role of dielectric constants (i.e., $s$ and $\epsilon_2$) is minor even though the red-shift behavior is shown as well.

The concentration effect is discussed in Fig. 3. We find that the high concentration predicts two characteristic frequencies, whereas the low predicts one only. That is, in the case of the high concentration, the multiple image
The spectral parameters \( s^{(1)} \), \( s^{(2)} \), \( F^{(1)} \), \( F^{(2)} \), \( \omega_c^{(1)} \), \( \omega_c^{(2)} \), \( \Delta \varepsilon^{(1)} \), and \( \Delta \varepsilon^{(2)} \), as a function of the concentration \( p \). Parameters for (c) and (d): \( s = 1.1, t = -1/90, \epsilon = 80, \) and \( \sigma_2 = 2.8 \times 10^{-4} \text{ S/m} \).

The effect does play a significant role. Note that \( p = 0.01 \) here can be approximated as the case for an isolated particle too, for which only one characteristic frequency is predicted.

Fig. 4 shows three cases:

(a) isolated cylinder (Re[\( b^* \)]);
(b) many cylinders randomly dispersed in a cluster by considering the multiple image effect (Re[\( b^* \)]);
(c) same as (b), but based upon the MGA, namely the local field effect is considered [15] (Re[\( b_{\text{MGA}} \)]).

Firstly, we compare (b) with (c). We find that the MGA predicts two characteristic frequencies as well [15,17]. Above of all, both methods are in good agreement. When we use the MGA to discuss the characteristic frequency of electrorotation in a recent work [15], two peaks are actually predicted as well, but they are located close enough to be overlapped due to the low concentration. In contrast to the present phenomena of two separated dispersions predicted by the MGA, it is further shown that the local-field effect will be more strong for the increasing concentration. However, we believe the present multiple image method predicts a more exact result. Secondly, we compare (a) with (b). The characteristic frequency obtained from the multiple image effect may be red-shifted or blue-shifted (namely, located at a higher frequency) when compared with the isolated particle case. Finally, for more information regarding the comparison between (a) and (c), we refer the reader to Ref. [15].
Fig. 2. DEP spectrum denoted by $\text{Re}[b^*]$, for $p = 0.6$. Parameters: (a) $s = 1.1, \epsilon_2 = 80$ and $\sigma_2 = 2.8 \times 10^{-4}$ S/m; (b) $t = -1/90, \epsilon_2 = 80$ and $\sigma_2 = 2.8 \times 10^{-4}$ S/m; (c) $t = -1/90, s = 1.1$ and $\sigma_2 = 2.8 \times 10^{-4}$ S/m; (d) $s = 1.1, t = -1/90$ and $\epsilon_2 = 80$.

Fig. 3. DEP spectrum for different concentrations $p$. Parameters: $\epsilon_2 = 80, \sigma_2 = 2.8 \times 10^{-4}$ S/m, $s = 1.1$, and $t = -1/90$.

Fig. 4. DEP spectrum for $\epsilon_2 = 80, \sigma_2 = 2.8 \times 10^{-4}$ S/m, $s = 1.1, t = -1/90$, and $p = 0.6$ for three cases: (a) isolated cylinder ($\text{Re}[b]$); (b) many cylinders randomly distributed in a cluster by considering the multiple image effect ($\text{Re}[b^*]$); (c) same as (b), but considering the local-field effect ($\text{Re}[b_{\text{MGA}}]$).
4. Discussion and conclusion

Here a few comments on our results are in order. We have considered the DEP spectrum of the circular or cylindrical particles in a cluster which is subjected to an applied nonuniform ac electric field. The multiple image arising from all the other particles has been taken into account by using the multiple image method. Moreover, we have compared the results obtained from the MGA and the multiple image method, and good agreement between them is shown. We believe the exact multiple image method predicts a more exact result than the MGA.

In the present Letter, we have studied a two-dimensional case since the existing multiple image method is exact in two dimensions, but approximate in three dimensions [10]. Due to the mathematical similarity in theory, this work might, to some extent, help to understand the possible exact multiple image method of three-dimensional cases. In addition, a similar treatment in three dimensions, albeit approximate, is relevant to realistic situation as well.

In summary, we have investigated the DEP spectrum of two-dimensional cluster colloidal suspensions, by taking into account the multiple polarization interaction or local-field effect. In doing so, we have applied the multiple image method and the MGA to study the DEP spectrum of the cluster, respectively. It has been found that the DEP spectrum can be modified due to the multiple images between the particles. Also, the multiple image method has been shown to be in good agreement with the MGA.

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