Higher-order dielectrophoresis of nonspherical particles

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Higher-order terms of dielectrophoretic (DEP) force are commonly ignored by invoking the simplifying dipole approximation. Concurrently, the trend towards micro- and nano-electrode structures in DEP design is bringing about an increasing number of instances where the approximation is expected to lose reliability. The case is severe for nonspherical particles (the shape of many biological particles) due to the shape-dependent nature of dielectric polarization. However, there is a lack of analytical means to determine multipole moments of nonspherical particles, numerical calculations of the same are regarded as unreliable, and there is a prevalence for higher-order force considerations to be ignored. As a result, the dipole approximation is used and/or nonspherical particles are approximated as spheres. This work proves the inefficacy of current qualitative criteria for the reliability of the dipole approximation and presents a quantitative substitute, with verified accuracy, that enables precise determination of the extent to which the dipole approximation would be reliable, and if found unreliable, corrects the approximation by adding second- and third-order terms of the DEP force. The effects of field nonuniformity, electrode design, and particle shape and aspect ratio on the significance of higher-order DEP forces is quantitatively analyzed. The results show that higher-order DEP forces are indeed of substantially increased significance for nonspherical particles; in the cases examined in this work, multipolar terms are seen to constitute more than 40% of the total force on ellipsoidal and cylindrical particles. It is further shown that approximating nonspherical particles as spheres of similar dimensions is subject to substantial error. Last, the substantial importance of the electrode design in influencing higher-order forces is shown.

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I. INTRODUCTION

Dielectrophoresis (DEP) describes the motion of dielectrics when subjected to nonuniform electric fields [1]. Electric fields polarize dielectrics, giving rise to the accumulation of charge at dielectric discontinuities. The assembly of polarization charge can be represented through an induced dipole and higher-order multipoles. In a nonuniform field, the charges experience unequal forces, hence the exertion of a net dielectrophoretic force. The magnitude and direction of the DEP force depend on the properties of the subject dielectric(s), and dielectric properties vary with changing electric field characteristics. As such, the dielectrophoretic force provides versatile access to the properties of the subject dielectric(s) and can be easily adjusted for the exertion of different forces on dielectrics of different morphological or internal properties. This feature and other advantages have led to DEP enabling a wide range of applications involving manipulation, separation, and characterization of dielectric particles [2–4].

As most biological particles and their suspending media exhibit dielectric properties, DEP has found particular usefulness in miniature devices known as laboratory-on-a-chip (LoC), aimed at providing point-of-care diagnostics with substantially reduced cost and analysis and reaction times. A typical LoC device houses a network of microchannels through which flow narrow streams of sample fluid, consisting of suspended micrometer and nanometer scale particles. Dielectrophoresis is one of the most common means of imparting particle motion in an LoC device, and has been used for the separation [5–8] (e.g., of healthy and cancerous cells [8]), characterization [5,9,10], and manipulation [11–13] of biological particles of different types.

It was initially believed that dielectrophoretic force would not be capable of handling particles of submicrometer dimensions due to the overpowering effect of randomizing forces [1]. Using electrode structures on the micrometer and nanometer scale fabricated using techniques borrowed from the electronics industry, DEP characterization, separation, and manipulation of submicron particles has been made possible [13–15], extending the applicability range of dielectrophoresis. With electrode gaps of smaller dimensions, dielectrophoretic forces of sufficient strength to dominate particle behavior can be generated by applying voltages of modest value, so that overheating of the fluid medium is avoided [16].

In all of the wide-ranging DEP applications, determining the dielectrophoretic force accurately is of crucial importance. The force can be determined from summing those on induced multipoles of ascending order, starting from the induced dipole. Yet such a calculation of the DEP force is subject to ambiguity due to the complex nature of the polarization charge comprising an unknown mixture of free and bound charges. The notion of effective multipoles circumvents this ambiguity by defining the effective multipoles as free-charge multipoles of ascending order, starting from the effective dipole, which replace the dielectric particle of interest in the electric field (and the suspending medium) and give the same electric potential as that arising from the particle itself. The DEP force is then calculated, by what is referred to as the effective moment (EM) method, from summing forces by the electric field on free-charge effective multipoles of ascending order [17].

The conventional theory for dielectrophoresis is based on a first-order (known as dipole) approximation that accounts

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only for the first-order terms of the DEP force. Higher-order terms that arise from higher-order effective moments and their interactions with increasing order field derivatives are assumed to be negligible. While competent in many circumstances, particularly the wide variety of applications that rely upon differences in DEP force sign rather than magnitude for particle separation, the dipole approximation is expected to become less reliable as particle dimensions become comparable to a characteristic length scale of electric field nonuniformity. Although the criterion is far from definitive, it suggests that with the current trend towards electrode structures of reduced dimensions, there will be an increasing number of instances where the dipole approximation is unreliable and higher-order forces need to be accounted for. For nonspherical particles, comprising the vast majority of biological particles, higher-order moments are expected to be of further significance due to the shape-dependent nature of polarization. It has been shown [18] that in uniform fields, while, as expected, the only nonzero moment is the dipole (*and all higher-order moments are zero*), nonspherical particles have nonzero values for higher-order moments. This indicates a fundamental difference in determining higher-order forces for spherical particles and nonspherical particles. The existing methods for determining higher-order forces have been defined only for spherical particles. As a result, in no DEP design involving nonspherical particles have higher-order terms of the dielectrophoretic force been accounted for.

In the very few DEP designs where higher-order forces have been accounted for, either only spherical particles are considered or nonspherical particles have been approximated as spheres of similar dimensions. Liang *et al.* [19] have determined the second-order DEP force on spherical particles (only) using an analytical method (which would inevitably be inapplicable to nonspherical particles) and identified positions within an interdigitated electrode structure where the higher-order term comprises 10% of the total DEP force. Dalir *et al.* [20] have also included higher-order terms for determining the dielectrophoretic force, but have again considered spherical particles only. For an electrode structure designed for cell fractionation and transport, Kua *et al.* [21] have analytically calculated the first-, second-, and third-order terms of DEP force, yet again (as the analytical nature of the method employed would imply) for spherical particles only. It was claimed in the paper that "these analytical expressions allow the evaluation on the importance of higher-order forces, which are not possible using conventional numerical techniques, such as those based on finite element or meshless methods." This paper will present quantitative evaluation (with verified accuracy) on the importance of higher-order DEP forces using a hybrid numerical-analytical technique implemented using the finite element method, an extension of previous work [18].

The (proven here to be false) claim by Kua *et al.* [21] regarding the inefficacy of numerical methods for determining higher-order DEP forces represents a presumptive obstacle in the correct determination of the dielectrophoretic force in most designs and devices, particularly those involving biological particles, the vast majority of which are nonspherical: Determining higher-order DEP forces requires determining the higher-order effective moments, and analytical expressions for higher-order moments are only available for spherical particles. The absence of analytical expressions for the higher-order moments of nonspherical particles has automatically led to neglecting higher-order forces, or if considering them, confining analysis to spherical particles, or else approximating nonspherical particles as spheres of similar dimensions. As an example, Rosales and Lim [22] have analyzed spherical and ellipsoidal particles in an octupolar DEP trap; for the spherical particle, the first-, second-, and third-order terms of the DEP force have been determined, while for the ellipsoidal particle, the authors have stopped at the first-order term. In another example, Zhu *et al.* [23] have analyzed a single-cell DEP trap for spherical and ellipsoidal particles, where they have determined the first- and second-order terms of the DEP force on the spherical, but only the first-order force on the ellipsoidal particle.

This paper will show that in the absence of analytical expressions for the higher-order moments of nonspherical particles, higher-order DEP forces on nonspherical particles can be determined using the presented alternative: a hybrid numerical-analytical method, based on previous work [18] that addresses concerns regarding the accuracy of numerical means of determining higher-order DEP force by verifying the calculations through comparison against total force calculations using the Maxwell stress tensor (MST) method. The MST method is well known as an unassailable method for determining electrical forces. By comparing the two sets of results (DEP force terms against the total force), as well as verifying the accuracy of the numerical calculations, contributions from each of the individual (first-, second-, and third-order) terms to the DEP force are accurately determined, and a quantitative analysis presented on the significance of higher-order dielectrophoretic forces. By considering two different electrode structures and three different particle shapes (spherical, ellipsoidal, and cylindrical), the large effect of field and electrode geometry and the particular significance of higher-order DEP forces for nonspherical particles will be analyzed in detail.

It is important to note that although the stress tensor method is an unassailable means of determining the total force, it is incapable of providing information as to the composition of the force from individual terms: first and higher order. The method presented in this work fills this void by presenting an accurate means of determining first- and higher-order force terms individually so that novel applications relying on circumstances where multipolar forces are significant and the dipolar force is not can be realized and extended beyond examples such as the levitation of dielectric particles under the influence of the quadrupolar force in a field null where the dipolar force is zero [24]. The MST method serves as a powerful verification tool guaranteeing the accuracy of force term calculations.

An important advantage of separate force term calculations using the method presented in this work over total force calculations using the MST method is in computational effort for the design or analysis of a DEP device. Using the multipole method, DEP force terms (the sum of which gives the total force) can be obtained for all positions in the device from a single simulation: a continuous solution. The MST method requires one simulation for every position within the device,

giving a discrete solution for the force at best. In summary, the computational effort to determine the multipole force terms at all positions within a device is the same as that required to determine the total force from the MST method for a single position, with a resulting computational overhead for the MST method orders of magnitude greater.

II. BACKGROUND AND THEORY

A. EM method determination of DEP force terms

The effective moment method presents an unambiguous formulation for determining first- and higher-order terms of the DEP force. The formulation relies on determining the effective moments $\mathbf{p}^{(n)}$ of the dielectric(s) subjected to the nonuniform electric field $\mathbf{E}$; the nth-order term of the DEP force is given by [25]

$$\mathbf{F}^{(n)} = \frac{1}{n!}\mathbf{p}^{(n)}[\cdot]^{(n)}\nabla^{(n)}\mathbf{E}, \tag{1}$$

where $[\cdot]^{(n)}$ is the generalized dot product and $\nabla^{(n)}$ denotes the nth-order derivative.

For spherical particles, analytical expressions are available for effective moments up to an arbitrary order, while no such expressions are available for the higher-order moments of nonspherical particles. The dipole approximation posits that higher-order moments, and hence higher-order terms of the DEP force, are negligible so that the DEP force can be simply determined from Eq. (1) for $n = 1$ [1] as follows:

$$\mathbf{F}^{(1)} = \mathbf{p}^{(1)} \cdot \nabla\mathbf{E}, \tag{2}$$

where $\mathbf{p}^{(1)}$ is the effective dipole moment. The dipole approximation is particularly simplifying in the case of nonspherical particles, given the unavailability of analytical means to determine higher-order effective moments. This is while shape-dependent polarization suggests that higher-order moments could be of added significance in the case of nonspherical particles.

In the absence of analytical expressions, the numerical-analytical method of Green and Jones [18] can be used to determine higher-order effective moments, up to an arbitrary order, of nonspherical particles in an axially symmetric setting. According to the method, the nth-order effective moment $p^{(n)}$ (reduced from the general tensor to a scalar in axial symmetry) of a dielectric particle suspended in a dielectric medium of permittivity ϵ_m is obtained from an integration of the electric potential $\phi_{R_{\text{int}}}$ due to the particle over a spherical enclosing surface of radius R_{int} [18]

$$p^{(n)} = 4\pi\epsilon_m R_{\text{int}}^{n+1}\frac{2n+1}{2}\int_0^{\pi}\phi_{R_{\text{int}}}P_n(\cos\theta)\sin\theta d\theta, \tag{3}$$

where $P_n(\cos\theta)$ are the Legendre polynomials, θ being the polar angle in spherical coordinates.

Higher-order effective moments of nonspherical particles can be obtained from Eq. (3) and the results incorporated into the effective moment method to obtain higher-order DEP forces. The first three terms of DEP force (which is as many as would fit in three-dimensional space) are determined in this work for ellipsoidal and cylindrical particles in two different electrode configurations to present a quantitative evaluation of the significance of higher-order DEP forces on nonspherical particles and the effects of variations in particle and electrode or field geometry on this significance. The quantitative measure for the significance of higher-order DEP forces will be individual and overall contributions from second- and third-order terms (determined from the EM method) to the total DEP force (determined from the MST method).

B. MST method determination of the total DEP force

The total dielectrophoretic force by an electric field $\mathbf{E}$ on a dielectric particle of permittivity ϵ_p suspended in a dielectric medium of permittivity ϵ_m can be obtained by integration of the Maxwell stress tensor over an enclosing surface [26]

$$\mathbf{F}_{\text{MST}} = \frac{1}{2}(\epsilon_p - \epsilon_m)\oint\left[E_{tm}^2 + E_{nm}^2\left(\frac{\epsilon_p}{\epsilon_m}\right)\right]\mathbf{n}dA, \tag{4}$$

where E_{tm} and E_{nm} are the tangential and normal components, respectively, of the electric field inside the suspending medium, and $\mathbf{n}$ is the unit vector normal to the integration surface A.

Through a comparison of force term calculations from the EM method and total force calculations using the MST method, contributions from first- and higher-order terms can be known and quantitatively compared. The MST method also importantly serves as a verification tool for DEP force term results.

In all of the instances studied in this paper (the different particle shapes, electrode configurations, and particle positions within electrode configurations), the sum of the first three DEP force terms determined from the EM method is seen to equal the total force determined from the MST method by an error margin never larger than 1%. This excellent match between the two sets of results approves numerical calculations of DEP force terms and contributions from first- and higher-order terms can be individually determined by dividing the respective term(s) over the MST method-calculated force. Such-obtained second- and third-order contributions provide an accurate quantitative measure for the significance of higher-order DEP forces and how they are affected by different factors including particle shape.

III. PHYSICAL PROBLEM SPECIFICATIONS

Figures 1(a) and 1(b) show the electrode structures studied in this work for assessing the effect of field or electrode geometry on the significance of higher-order DEP forces. By studying point-plane and disk-plane electrode structures with the same electrode separation (20 μm between the point or disk and plane electrodes) and with the same voltage applied across the electrodes, the importance of electrode design on the extent to which higher-order DEP forces gain significance is analyzed. It will be shown that slight variations in electrode shape could result in substantial difference in the significance of higher-order DEP forces on a given particle. To study the effect of electric field characteristics on the significance of higher-order DEP forces, a wide range of particle positions within each electrode structure is studied. The different particle positions correspond to electric fields with different degrees of nonuniformity, with the definition of nonuniformity extended to include second- and third-order as well as the first-order derivative of the electric field.

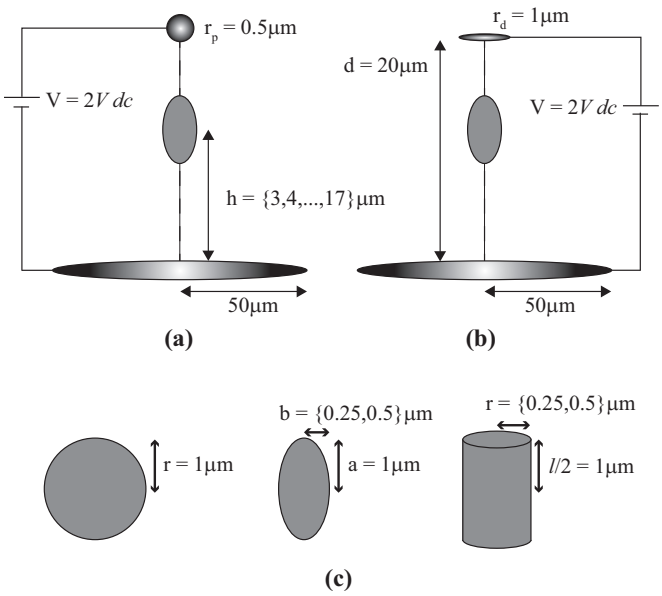

FIG. 1. Electrode structures and particle shapes and dimensions studied in this work for analysis of the effect of electrode or field and particle geometry on the significance of higher-order DEP forces: (a) the point-plane electrode structure, (b) the disk-plane electrode structure, (c) dimensions of spherical, ellipsoidal, and cylindrical particles.

The particle geometries (shapes and dimensions) studied in this work are shown in Fig. 1(c). It is the principal aim of this work to show, and quantify, the significance of higher-order DEP forces on particles of nonspherical shape (comprising the vast majority of biological particles). Ellipsoidal and cylindrical particles have been studied as example shapes, typically used as models for more highly nonspherical particles. To show the effect of nonspherical shape on the significance of higher-order DEP forces, results with ellipsoidal and cylindrical particles are compared against those for a most closely fitting spherical particle of the same dielectric properties. In addition to the dipole approximation, the comparison provides an assessment on the validity of another approximation commonly used, separately or in conjunction with the dipole approximation, in DEP design: that of nonspherical particles as a sphere of similar dimensions. It will be shown that the reliability (or otherwise) of the two approximations are related, in that modeling nonspherical particles as a sphere of equal volume is guaranteed to be error-free only if the dipole approximation is fully reliable, i.e., higher-order DEP forces are negligible. Two different aspect ratios for ellipsoidal and cylindrical particles are considered to assess the effect of particle thinness on the significance of second- and third-order DEP forces.

To focus attention on the effect of particle geometry (and electric field characteristics) on the significance of higher-order DEP forces, simple assumptions have been made regarding internal particle or medium properties: it has been assumed that the particles and their suspending media are lossless dielectrics with relative permittivities of 3 and 80 (pertaining to typical polymeric particles and water), respectively.

IV. RESULTS AND DISCUSSION

A. Derivatives of the electric field

Variations of the electric field magnitude and its first three derivatives with position h along the axis of field symmetry are shown in Fig. 2(a) for the point-plane electrode geometry, showing monotonically increasing field magnitude and derivatives towards the point electrode with a sharper rate of increase nearer the point electrode. Field magnitude and derivative profiles for the disk-plane geometry, shown in Fig. 2(b), are demonstrative of the pronounced effect of electrode design on field curvature. While the only difference between the two electrode structures is the change in shape of a 0.5-μm-radius point (spherical) electrode to a 1-μm-radius disk, field magnitude and derivative profiles are seen to differ substantially: in clear contrast to the point-plane geometry, the electric field and its derivatives in the disk-plane geometry remain within the same order of magnitude as position h along the axis of symmetry spans the full range; also, the second-order derivative attains negative values at half of the positions along the axis of field symmetry. In both electrode configurations, the electric field and its derivatives have been calculated analytically.

B. Effective moments

The first three effective moments of spherical ($r = 1\,\mu$m), ellipsoidal ($a = 1\,\mu$m,$b = \{0.5,0.25\}\,\mu$m), and cylindrical ($l/2 = 1\,\mu$m,$r = \{0.5,0.25\}\,\mu$m) particles at different positions within the point-plane electrode geometry are shown in Fig. 3(a). As with the field magnitude and derivatives, first- and higher-order effective moments of particles positioned within the point-plane geometry are positive-valued and monotonically increase with distance h from the plane electrode. Particle geometry (shape or size) is of no effect on the trends with which first- or higher-order moments vary with particle position with respect to the electrodes. However, the values of the effective moments at a given position h show strong dependency on particle geometry: With the exception of the spherical particle possessing third-order moments smaller than those of all (smaller in volume) nonspherical particles, the effective moments are found to be larger for particles of larger volume. It is noted, however, that only the dipole moment (and not higher-order moments) is directly proportional to particle volume. It has been common practice to model nonspherical particles as spheres of similar dimensions to simplify the calculation of DEP force. From the direct proportionality of only the effective dipole moment with particle volume it can be concluded that modeling nonspherical particles with spheres of equal volume will be error-free if and only if higher-order moments can be ignored, i.e., if and only if the dipole approximation is found to be reliable.

The first- and higher-order effective moments of the same particles positioned within the disk-plane electrode geometry are plotted against h in Fig. 3(b). The profile of each of the effective moments can be closely identified with that of the field derivative of the preceding order, in line with the nth-order effective moment representing the energy stored in the particle by the $(n-1)$-th-order field derivative (with $n = 0$

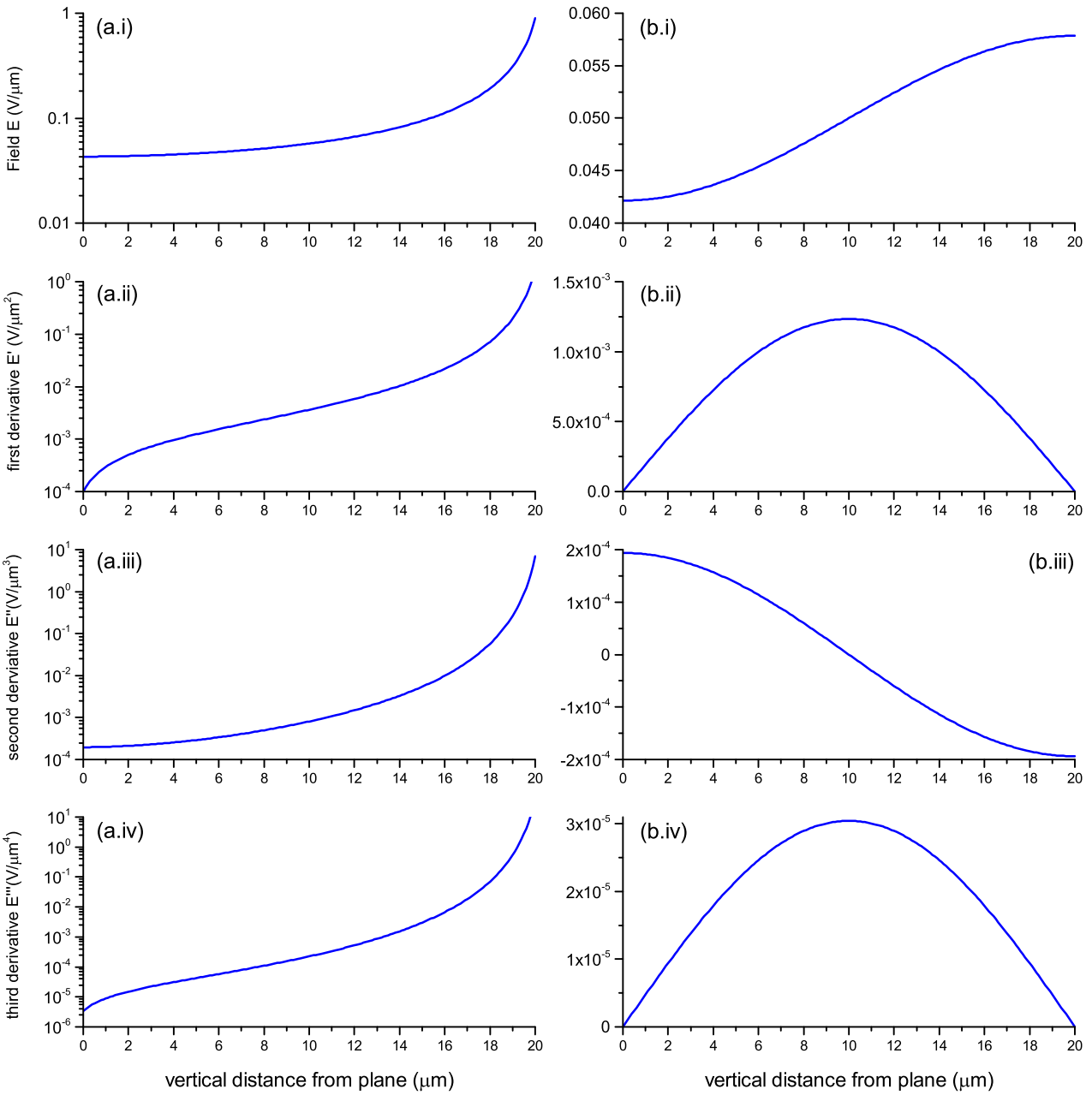

FIG. 2. (Color online) Field curvature profiles for point-plane and disk-plane electrode structures showing the pronounced effect of electrode design on electric field nonuniformity: (i) the electric field magnitude and its (ii) first-, (iii) second-, and (iv) third-order derivatives along the symmetry axes of (a) point-plane and (b) disk-plane electrode structures with the characteristics given in Fig. 1.

corresponding to the field magnitude). The same observations made with the point-plane configuration regarding the effect of particle geometry on the effective moments, and regarding the interdependency of the two common approximations in DEP design involving nonspherical particles (the dipole approximation and modeling the particles as spheres of similar dimensions) are made with the disk-plane configuration also. The importance of electrode design on the effective moments of spherical and nonspherical particles is clear from the substantial difference between trends and values of first- and higher-order moments in point-plane and disk-plane electrode structures, while the two electrode configurations differ only slightly in geometry.

C. Dielectrophoretic force terms

The first-, second-, and third-order terms of the DEP force on spherical, ellipsoidal, and cylindrical particles at different positions within the point-plane electrode configuration are plotted in Fig. 4(a). The DEP force terms are determined from combining the effective moment and field derivative results in accordance with the effective moment (EM) method. As expected from effective moment and field derivative results and the EM method formulation, first- and higher-order DEP forces on spherical and nonspherical particles increase monotonically as particles distance from the plane electrode and approach the point electrode. With the only exception of $F^{(3)}$ for the spherical particle, first- and higher-order DEP forces are found

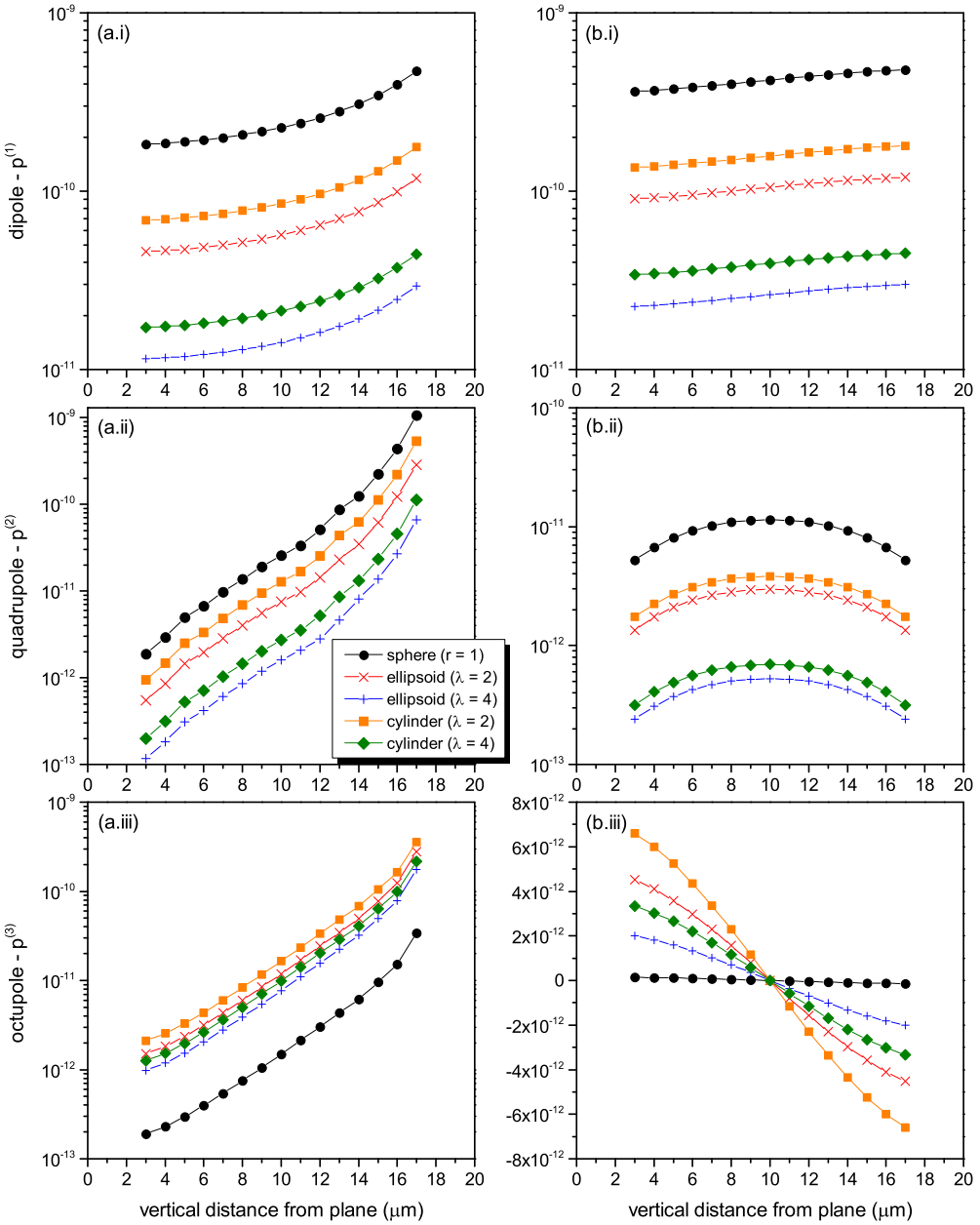

FIG. 3. (Color online) (i) First- ($p^{(1)}$), (ii) second- ($p^{(2)}$), and (iii) third-order ($p^{(3)}$) effective moments of spherical ($r = 1\ \mu$m), ellipsoidal ($\lambda = 2$, $\lambda = 4$), and cylindrical ($\lambda = 2$, $\lambda = 4$) particles at different positions within (a) point-plane and (b) disk-plane electrode structures.

to be larger for particles of larger volume, while only the DEP force obtained from the dipole approximation is directly proportional to the particle volume. The same observation is made with the first- and higher-order DEP forces on the same particles positioned within the disk-plane electrode structure [Fig. 4(b)], leading to the same conclusion as that made previously with the effective moments: in DEP designs involving nonspherical particles, modeling the particles with spheres of equal volume is error-free if and only if the dipole approximation is error-free.

As with the moments, the significant effect of electrode shape or design on the trends (i.e., variation patterns with h) and values of first- and higher-order DEP forces is clearly visible from the substantial difference between the plots for $F^{(n)}$ in Fig. 4 pertaining to point- and disk-plane electrodes. The substantial difference, which indeed arises from similar levels of difference presented previously for the field curvature and effective moment profiles between the two electrode configurations, signifies the importance of electrode design on first- and higher-order dielectrophoretic forces on spherical and nonspherical particles.

D. Verified significance of higher-order DEP forces

The reliability of the dipole approximation, and in other words the significance of higher-order DEP forces, has been determined on the basis of individual and overall contributions from second- and third-order terms to the total DEP force on particles, determined by dividing the respective terms (obtained using the effective moment method) over the total

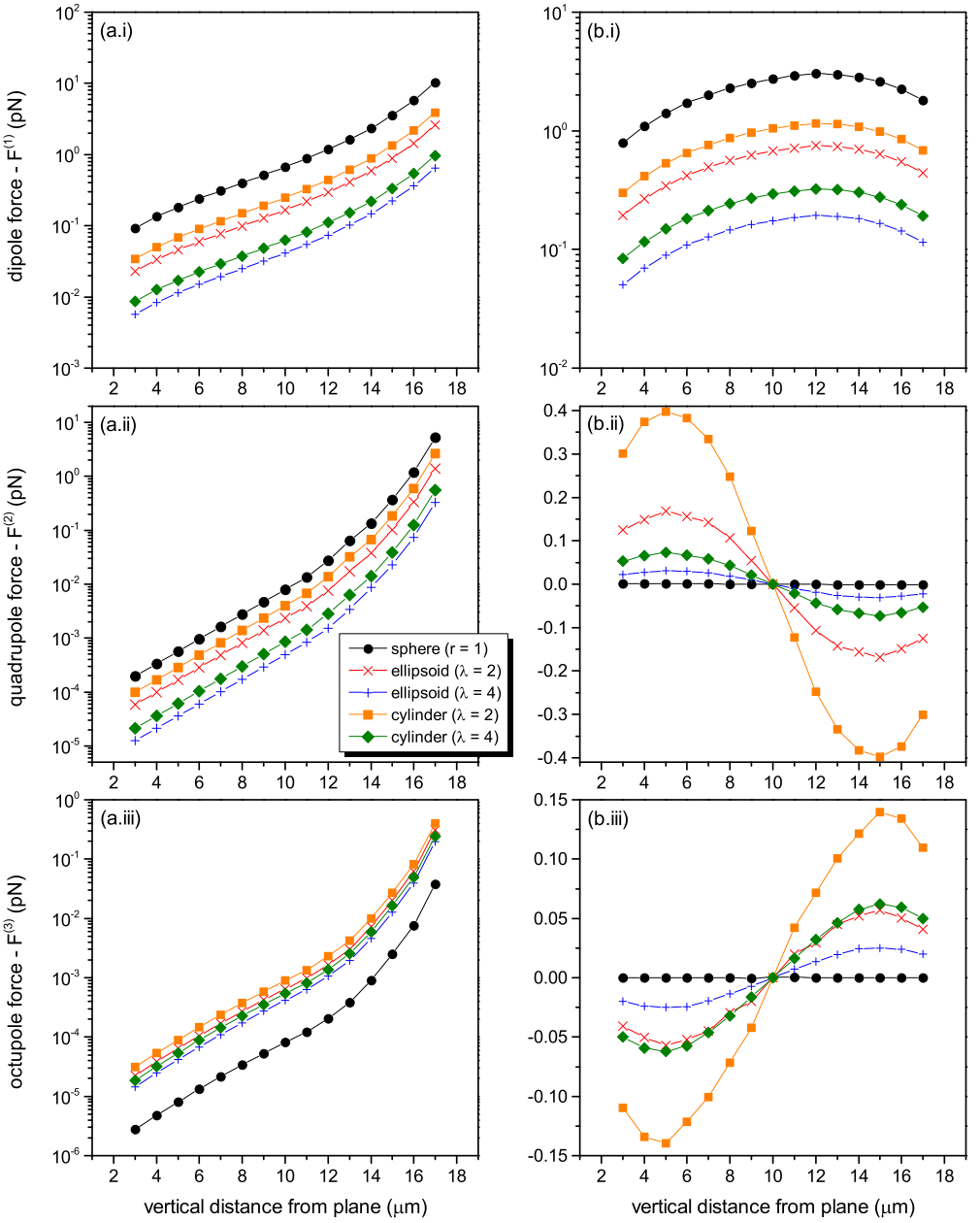

FIG. 4. (Color online) (i) First- ($F^{(1)}$), (ii) second- ($F^{(2)}$), and (iii) third-order ($F^{(3)}$) terms of DEP force on spherical ($r = 1\,\mu$m), ellipsoidal ($\lambda = 2$, $\lambda = 4$), and cylindrical ($\lambda = 2$, $\lambda = 4$) particles at different positions along the symmetry axes of (a) point-plane and (b) disk-plane electrode structures.

force (obtained using the Maxwell stress tensor method). Results for higher-order contributions to the DEP force on spherical, ellipsoidal, and cylindrical particles at different positions along the symmetry axes of point-plane and disk-plane electrode structures are shown in Fig. 5.

1. *Point-plane electrode structure*

In the case of the point-plane geometry, the plots only show the results for half of the examined range of particle positions along the symmetry axis; for points nearer the plane than the point electrode ($h < 10\,\mu$m), higher-order contributions to the DEP force are found to be below 2%, regardless of particle geometry. As particles approach the point electrode, the DEP force they experience is seen to become decreasingly dipolar: second- and third-order contributions to the DEP force are seen to rapidly increase as particles move closer to the point electrode.

All three plots for the point-plane geometry [Fig. 5(a)] show the pronounced effect of nonspherical particle shape on the significance of higher-order DEP forces: second- and third-order contributions to the DEP force are found to be notably larger for nonspherical particles than for the sphere of similar dimensions. For the $r = 1\,\mu$m spherical particle, the maximum contribution from the second-order term is nearly 10% at $h = 17\,\mu$m, while for the $a = 1\,\mu$m, $b = 0.5\,\mu$m ellipsoidal particle of the same dielectric properties at the same position within the electrode geometry, the second-order contribution

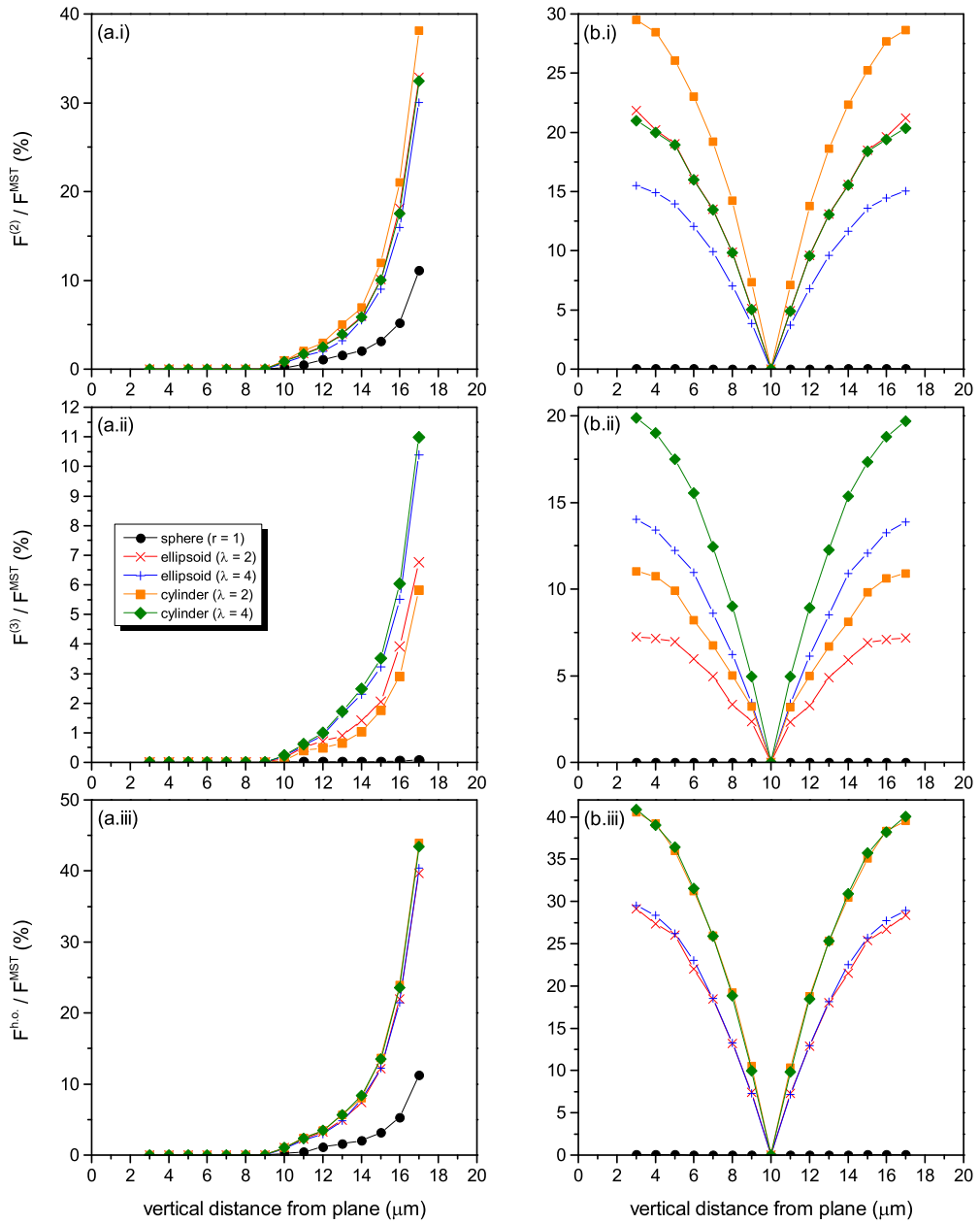

FIG. 5. (Color online) Higher-order contributions to the DEP force on spherical ($r = 1\,\mu$m), ellipsoidal ($\lambda = 2$, $\lambda = 4$), and cylindrical ($\lambda = 2$, $\lambda = 4$) particles in (a) point-plane and (b) disk-plane electrode configurations, determined by comparing force term calculations using the effective moment method against total force calculations using the MST method: Individual contributions from (i) second- and (ii) third-order terms, and (iii) the overall higher-order contribution, obtained by summing second- and third-order contributions.

is seen to be 34%, i.e., more than a third of the total DEP force. For a cylindrical particle ($\lambda = 2$) in the same position, the second-order contribution is seen to be even larger; nearly 38%. Third-order contributions to the DEP force are also seen to be largely different between nonspherical particles and a sphere of similar dimensions. While the third-order term is found to have negligible (<0.1%) contribution to the force on the spherical particle, up to >10% contributions from the third-order term are observed for ellipsoidal and cylindrical particles. A comparison of the plots in Figs. 5(a.i) and 5(a.ii) show that along the symmetry axis of the point-plane geometry, the second-order term dominates higher-order contributions to the DEP force on spherical and nonspherical particles.

Figure 5(a.iii) presents a quantitative evaluation of the reliability of the dipole approximation for a range of particle and field geometries. It can be seen that while the first-order approximation is a highly reliable means of predicting DEP force on spherical and nonspherical particles positioned nearer the plane electrode ($h < 10\,\mu$m), it becomes increasingly erroneous as particles approach the point electrode. At $h = 17\,\mu$m, the dipole approximation will underestimate the DEP force on ellipsoidal and cylindrical particles by ~40% and ~44%, respectively. For a most closely fitting spherical particle, the error is ~10%. The plot shows the strong dependency of the reliability of the dipole approximation on particle and electric field geometry: the approximation can

be highly accurate or highly erroneous for the same particle, depending on the position within the electrode geometry, i.e., field curvature. The reliability of the dipole approximation is also found to vary significantly for particles of different shapes: A significant increase in the error incurred upon invoking the dipole approximation is observed as soon as the particle deviates from spherical shape.

Figure 5(a.iii) shows another characteristic of higher-order DEP forces: The specificity of the error incurred upon invoking the dipole approximation to particle shape and independence from particle aspect ratio for a given nonspherical shape. Overall higher-order contribution to the DEP force is found to be identical for $a = 1\,\mu$m ellipsoidal particles of different aspect ratios, and for $l/2 = 1\,\mu$m cylindrical particles of different aspect ratios. It is found that for a given (nonspherical) particle shape, increasing aspect ratio (i.e., thinning the particle) gives rise to smaller quadrupolar and larger octupolar contributions to the DEP force experienced by the particle, in a manner that the sum of second- and third-order contributions remains independent of the aspect ratio and specific to the particle shape. A similar observation can be made in the case of the disk-plane geometry.

2. Disk-plane electrode structure

The plots in Fig. 5(b) show percentage higher-order contributions to the DEP force on spherical, ellipsoidal, and cylindrical particles at different positions along the symmetry axis of the disk-plane electrode geometry. It can be seen that for the spherical particle, second- and third-order terms constitute a negligible (<1%) portion of the dielectrophoretic force, while for ellipsoidal and cylindrical particles, multipolar contributions are seen to reach maximum values of ∼30% and ∼40%, respectively. The positions along the axis of field symmetry where higher-order forces are more significant differ notably from those for the point-plane geometry; showing the importance of the choice of electrode geometry on the reliability of the dipole approximation. In the disk-plane geometry, higher-order forces are found to contribute in largest proportion to the total DEP force when particles are positioned near the disk or plane electrode. Multipolar contributions are seen to almost symmetrically drop towards nearly zero at the midpoint $h = 10\,\mu$m. Compared to the case of the point-plane geometry, third-order contributions to the DEP force are seen to be of added significance. For the $\lambda = 4$ cylindrical particle positioned at $h = 3\,\mu$m or $h = 17\,\mu$m, nearly 20% of the DEP force is constituted by the third-order term alone. For the same particle at the same positions, another ∼20% of the total force is found to be constituted by the second-order term, making the DEP force predicted by the dipole approximation account for only ∼60% of the actual force.

Figure 5(b.iii) shows that the error incurred upon invoking the dipole approximation is in excess of 10% for all of the nonspherical particles at almost all positions along the symmetry axis of the disk-plane geometry. Figure 5(b.iii) also shows that as with the point-plane geometry, the overall contribution from higher-order terms to the dielectrophoretic force on a nonspherical particle of a given shape is specific to the shape and independent of aspect ratio, i.e., particle thinness. As with the point-plane electrode geometry, an increasing aspect ratio is seen to give rise to larger third-order and smaller second-order contributions to the DEP force experienced by the particle, with the increase and decrease occurring in a manner that the overall higher-order contribution remains independent of the aspect ratio.

V. CONCLUSION

A. Summary of key findings

The commonly and indiscriminately invoked dipole approximation can be highly erroneous in predicting the dielectrophoretic force. Circumstances where the dipole approximation would be unreliable are currently identified based on general and qualitative criteria that, at best, (claim to) provide a yes or no answer. It has been shown in this work that the criteria are not at all general and can provide a wrong yes or no answer. This work has presented a quantitative alternative that can accurately determine the extent to which the dipole approximation is reliable and, if found unreliable, correct the approximation by adding second- and third-order forces. The method has been applied to spherical, ellipsoidal, and cylindrical particles at different positions within two electrode structures to quantitatively analyze the significance of higher-order DEP forces for a range of particle and field geometries, with particular attention to the case of nonspherical particles that comprise the vast majority of biological particles.

A major reason for the inattention to higher-order DEP forces, particularly on nonspherical particles, has been the lack of availability of analytic means to determine the higher-order moments of nonspherical particles. In the absence of analytic means, numerical methods are deemed inaccurate and higher-order forces are deemed almost always negligible. As a result, higher-order DEP forces are partially or (most often) totally ignored or nonspherical particles are approximated as spheres of similar dimensions. The accuracy of the numerical method employed in this work has been verified and the results have shown that higher-order terms can constitute nearly half of the total DEP force. It has also been shown that higher-order DEP forces are notably more significant for nonspherical particles and that approximating nonspherical particles with spheres of similar dimensions is subject to substantial error. Finally, it has been shown that electrode structure or design has a pronounced effect on the significance of higher-order DEP forces.

B. Significance of results

Figure 6 is a summary of the results of this work, showing the proportional contributions of first-, second-, and third-order DEP forces across the domains of the electrode geometries. Higher-order terms have been shown to constitute up to ∼45% and ∼40% of the total DEP force on the examined cylindrical and ellipsoidal particles, respectively. This highlights the error that can be incurred upon invoking the dipole approximation for determining the DEP force on nonspherical particles. However, it has been noted that higher-order terms do not always contribute in such significant proportion to the DEP force on nonspherical particles. There are instances where nonspherical particles are subject to higher-order forces of minimal significance; e.g., at $h = 10\,\mu$m in the disk-plane [Fig. 6(b)] and positions not close to the point electrode in the

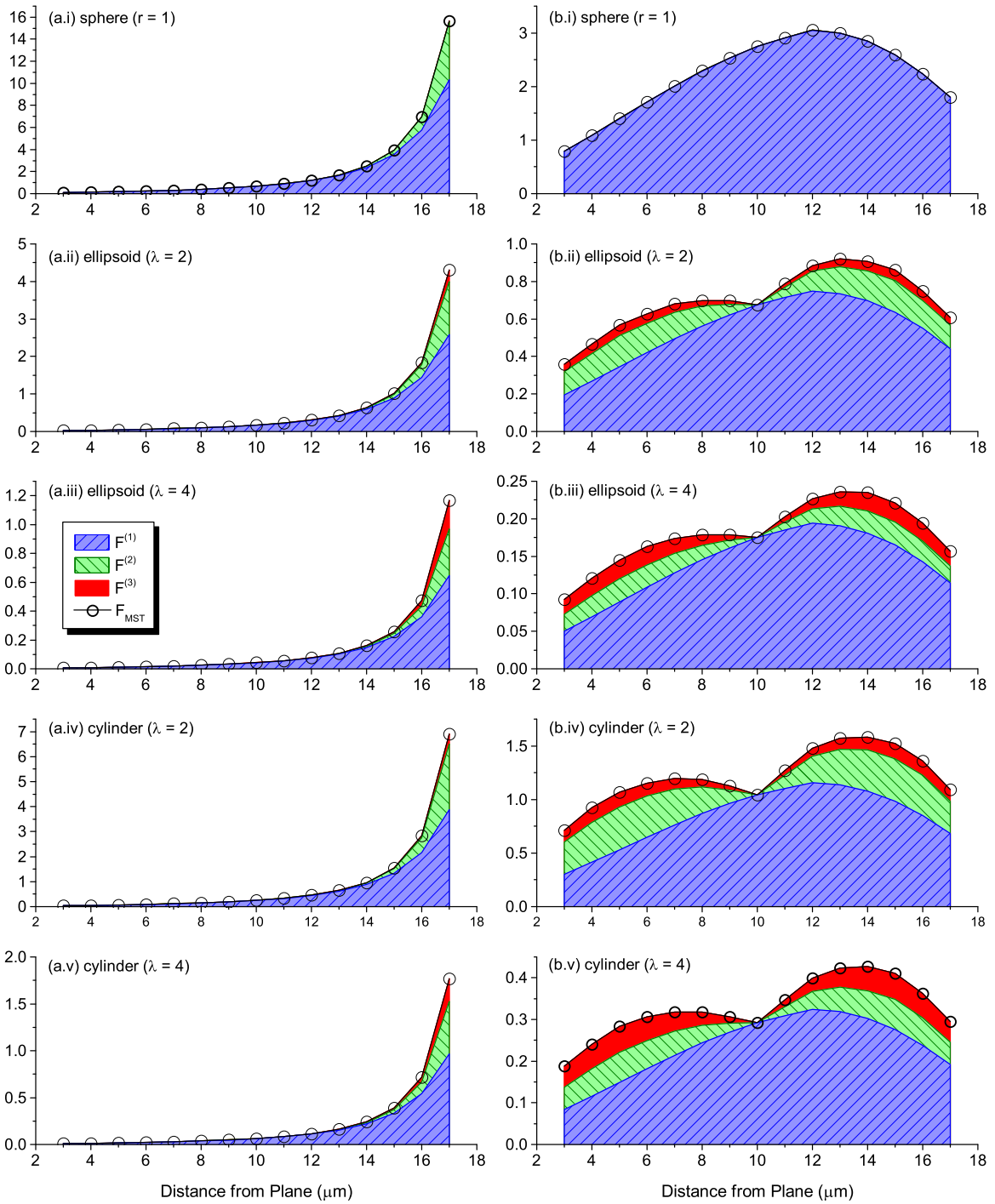

FIG. 6. (Color online) Stacked area plots showing the first three terms of DEP force ($F^{(1)}$, $F^{(2)}$, and $F^{(3)}$) and the total DEP force (F_{MST}), determined numerically, on (i) spherical ($r = 1\,\mu$m), ellipsoidal [(ii) $\lambda = 2$, (iii) $\lambda = 4$], and cylindrical [(iv) $\lambda = 2$, (v) $\lambda = 4$] particles in (a) point-plane and (b) disk-plane electrode configurations.

point-plane configuration [Fig. 6(a)]. The instances, however, importantly do not correspond to the commonly stated criterion for the limit of the reliability of the dipole approximation, namely that higher-order forces gain increased significance when the electric field varies more notably across particle dimensions. A clear example counter to the criterion has been shown to be the case of negligible higher-order forces at $h = 10\,\mu$m in the disk-plane configuration [Fig. 6(b)] where, based on field curvature profiles [Fig. 2(b)], the field varies most notably across particle dimensions. Addressing the fallacy of such criteria, this work has provided a quantitative evaluation approach, with verified accuracy, to substitute general criteria and "by-inspection" predictions (proven to be subject to large error in this work) on the significance of higher-order DEP forces.

Figure 6 also demonstrates the verification of the numerical results presented in this work of higher-order DEP forces on nonspherical particles. Above all the stacked area plots lies

a F_{MST} curve exactly fitting the sum of $F^{(1)}$, $F^{(2)}$, and $F^{(3)}$. Given the unassailability of the MST method in encompassing all interactions between an electric field and the subject dielectric(s) that generate force, the excellent match between the two sets of results provides verification for the numerical second- and third-order DEP force calculations presented in this work.

The clear contrast between the plots in Fig. 6 for point- and disk-plane electrode geometries emphasizes the considerable effect of electrode shape or design on the significance of higher-order DEP forces. It has been shown in this work that a simple change of shape of one electrode (with all other geometrical parameters and the applied voltage unchanged) from a 0.5-μm sphere to a 1-μm disk will result in substantial change in the proportions by which higher-order terms contribute to the total DEP force on particles, as well as positions within the electrode geometries where these proportions are more significant. A clear example is positions near the plane electrode in the two electrode structures: negligible ($<1\%$) higher-order forces in the point-plane and significant ($>30\%$) higher-order forces in the disk-plane geometry. The notable differences show that only after accurate derivation of higher-order DEP force terms can judgements be ruled on whether or not (and in either case to what extent) the dipole approximation is reliable and, importantly, the results will only hold valid for the given electrode design and not at all generalizable to "similar" designs.

A major roadblock for including higher-order forces in DEP designs involving nonspherical particles has been the lack of availability of analytic expressions for the higher-order effective moments of nonspherical particles. In the absence of analytic means, numerical calculations of higher-order forces are perceived as inaccurate and subject to differentiation error, and the computational effort to circumvent the error is seen as unworthy of the deemed negligence of higher-order DEP forces. In this work (a) it has been shown that higher-order DEP forces on nonspherical particles are far from negligible and can comprise nearly half of the total DEP force, (b) a hybrid numerical-analytical method has been used to determine higher-order DEP forces, and the accuracy of the method has been verified by comparing results against total force calculations using the unassailable Maxwell stress tensor method, (c) the computational power required for the calculations has been very modest, and (d) it has been shown that a quantitative evaluation of the significance of higher-order forces is essential in DEP designs involving nonspherical particles, of which shape are most biological particles. Approximation with spheres, as is commonly done, has been shown to cause substantial error.

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