First-principles calculation of electrical forces among nanospheres in a uniform applied electric field

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ABSTRACT

We present a unified framework for a first-principles calculation of the electric force acting on dielectric or metallic nanospheres suspended in a dielectric host and subject to a uniform external electric field. This framework is based on the spectral representation of the local electric field in a composite medium. The quasi-static (or "surface-plasmon") eigenstates of a cluster of spheres are first calculated, numerically. Then those are used to calculate the force on any sphere as the gradient of the total electrostatic energy with respect to the position of that sphere. This approach is applicable even when the spheres are very closely spaced, and even when they are metallic: No infinities ever appear.

The forces are not limited to dipole-dipole forces. Moreover, the force acting on any sphere is not a simple sum of two-body forces: When the inter-sphere gaps are small, complicated many-body forces appear. This is due to the fact that, when a sphere center is displaced slightly, the electric polarization of all the other spheres is changed. Consequently, the total electrical energy is changed in a way that cannot be represented as a sum of two-body energy changes. Explicit calculations of these forces for a few selected sphere clusters are presented. The results are quite different from what is obtained in the dipole approximation.

Keywords: Electro-rheology, surface plasmon resonances, quasi-static eigenstates, many-body forces

1. INTRODUCTION

When an external electric field is applied to a suspension of particles in a host medium, each particle is usually subject to a nonzero force, as well as to a nonzero moment of force. These forces arise because, when the local electric permittivity of the medium $\varepsilon(\mathbf{r})$ varies in space, (i.e., the particles and the host medium are characterized by different values ε_p, ε_h of that permittivity) then the local electric field $\mathbf{E}(\mathbf{r})$ also becomes position dependent. These forces play a crucial role in electro-rheological fluids. The suspended particles in such a fluid have a higher permittivity than the host $\varepsilon_p > \varepsilon_h$, consequently the forces which act upon them when an external field is applied can bring about a clustering of those particles into long chains, dramatically increasing the viscosity of the suspension. As an extreme result, the system may thereby even change its macroscopic mechanical response from that of a fluid to that of a solid, i.e., a *qualitative, phase-transition-like, change of behavior*[1]! Other situations where such forces might play a crucial role is in the production of small clusters of closely spaced metal spheres for possible use in nano-optical devices. Those devices include nanolens[2] and SPASER[3] (acronym for "surface plasmon amplification by stimulated emission of radiation").

When an external uniform field $\mathbf{E}_0$ is applied to such a suspension, each particle becomes electrically polarized. This polarization is an additional source of electric field, causing the total local field $\mathbf{E}(\mathbf{r})$ to be position dependent. When the suspension is non-dilute, the particle polarization is not limited to electric dipole moments. Even if

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all the particles have spherical shape, electric multipole moments of all orders l, m will be induced when the particles are close to each other and the ratio $\varepsilon_p/\varepsilon_h$ is not close to 1.[4] Moreover, under those conditions the force on any particle cannot be approximated by a sum of two-body forces between pairs of particles. For these reasons, we have developed an alternative approach for calculating these forces without having to assume two-body interactions and without limiting the induced polarization of particles to electric dipole moments.[5]

In Section 2 we review the spectral representation for the local electric potential in a suspension of spherical particles when an external uniform electric field is applied and the quasi-static approximation is valid. Based on that representation, a method is developed in Section 3 for calculating the force acting on each of the suspended spheres. This method is then applied to compute the force in two cases—a two-sphere cluster and a three-sphere linear cluster of closely spaced metal spheres suspended in a conventional dielectric host.

2. THEORETICAL BACKGROUND

Given a two-constituent composite medium, a convenient way to express any solution of Maxwell's equations in the quasi-static regime is to expand the electric potential $\phi(\mathbf{r})$ in a series of the "quasi-static eigenstates", also known as "surface-plasmon resonances".[5,6] These eigenstates are solutions of the quasi-static-limit Maxwell equations for which $\phi(\mathbf{r})$ vanishes at the system boundaries.*. Such non-trivial solutions exist only for special values ("eigenvalues") of the electric permittivity ratio $\varepsilon_p/\varepsilon_h$. Using the following representation for the spatially varying local electric permittivity $\varepsilon(\mathbf{r})$

$$\varepsilon(\mathbf{r}) = \varepsilon_h \left(1 - \frac{\theta_p(\mathbf{r})}{s}\right), \tag{1}$$

where

$$s \equiv \frac{\varepsilon_h}{\varepsilon_h - \varepsilon_p} \tag{2}$$

and $\theta_p(\mathbf{r})$ is the characteristic step function of the p-constituent

$$\theta_p(\mathbf{r}) = \begin{cases} 1 & \text{for } \mathbf{r} \text{ in the } p \text{ constituent,} \\ 0 & \text{otherwise,} \end{cases} \tag{3}$$

the eigenvalue problem can be written in terms of a (partial) differential equation with (zero) boundary conditions:

$$\begin{cases} s_n \nabla^2 \phi_n(\mathbf{r}) = \nabla \cdot [\theta_p(\mathbf{r}) \nabla \phi_n(\mathbf{r})], \\ \phi(\mathbf{r}) = 0 \ \text{ on the system boundaries.} \end{cases} \tag{4}$$

The eigenvalues s_n are special values of s, all of which lie on the semi-closed segment $[0, 1)$ of the real axis. Using Green's function $G(\mathbf{r}, \mathbf{r}') = G(\mathbf{r}', \mathbf{r})$ of this problem, which is the solution of

$$\begin{cases} \nabla^2 G(\mathbf{r}, \mathbf{r}') = -\delta^3(\mathbf{r} - \mathbf{r}'), \\ G(\mathbf{r}, \mathbf{r}') = 0 \ \text{ on the system boundaries,} \end{cases} \tag{5}$$

we can re-express the eigenvalue problem in terms of the following integro-differential operator:

$$\hat{\Gamma}\phi \equiv \int dV' \theta_p(\mathbf{r}') \nabla' G(\mathbf{r}, \mathbf{r}') \cdot \nabla' \phi(\mathbf{r}'). \tag{6}$$

This (linear) operator enables us to write the eigenvalue problem as

$$s_n \phi_n = \hat{\Gamma} \phi_n. \tag{7}$$

It also enables us to write the following integro-differential equation for $\phi_n(\mathbf{r})$ when an external uniform electric field of unit intensity $\mathbf{E}_0 = \mathbf{e}_z$ is applied in the z-direction:

$$\phi = z + \frac{1}{s} \hat{\Gamma} \phi. \tag{8}$$

*We note, in passing, that other types of zero boundary conditions may also be used, e.g., on the normal derivative of $\phi(\mathbf{r})$; also "mixed (Neumann-Dirichlet) boundary conditions". More details on this can be found in Refs. 6, 7, and 8

Defining a scalar product of two (potential) functions as

$$\langle\psi|\phi\rangle \equiv \int dV \theta_p(\mathbf{r}) \nabla\psi^*(\mathbf{r}) \cdot \nabla\phi(\mathbf{r}), \tag{9}$$

it is easy to show that $\hat{\Gamma}$ is a Hermitian operator. Consequently, inside the sub-volume of p-constituent, $\phi(\mathbf{r})$ can be expanded in terms of the complete set of eigenstates $\phi_n(\mathbf{r})$ of $\hat{\Gamma}$ as follows:

$$\phi = \frac{s}{s-\hat{\Gamma}} z = \sum_n \frac{s\langle\phi_n|z\rangle}{s-s_n}\phi_n. \tag{10}$$

An alternative representation for the integro-differential operator $\hat{\Gamma}$, as well as for $\phi(\mathbf{r})$, is obtained by using the complete set of eigenstates of the $\hat{\Gamma}_b$ operators of the *isolated inclusions* b:

$$\hat{\Gamma}_b\phi \equiv \int dV' \theta_b(\mathbf{r}') \nabla' G(\mathbf{r},\mathbf{r}') \cdot \nabla'\phi(\mathbf{r}'). \tag{11}$$

Here the step function $\theta_b(\mathbf{r})$ of the isolated inclusion b is similar to $\theta_p(\mathbf{r})$, except that $\theta_b(\mathbf{r}) \neq 0$ only when $\mathbf{r}$ is inside *that particular inclusion*. A complete set of states *in the p-constituent* is obtained by using the isolated inclusion eigenfunctions from *all the inclusions a*. In such a basis, $\phi(\mathbf{r})$, as well as $\phi_n(\mathbf{r})$, is represented by a vector of expansion coefficients, while the operator $\hat{\Gamma}$ is represented by a matrix.

Such a representation is especially convenient when the inclusions b, which make up the p-constituent, all have spherical shape. In that case, we use a vector-index $\mathbf{b} \equiv (|\mathbf{b}|, \theta_\mathbf{b}, \varphi_\mathbf{b})$ to denote the center of such an inclusion, a to denote its radius. The normalized-to-one eigenfunctions and eigenvalues of such an inclusion are given by:

$$\phi_{\mathbf{b}lm} = \begin{cases} \frac{|\mathbf{r}-\mathbf{b}|^l}{l^{1/2}a^{l+1/2}} Y_{lm}(\Omega_{\mathbf{r}-\mathbf{b}}), & |\mathbf{r}-\mathbf{b}| < a, \\ \frac{a^{l+1/2}}{l^{1/2}|\mathbf{r}-\mathbf{b}|^{l+1}} Y_{lm}(\Omega_{\mathbf{r}-\mathbf{b}}), & |\mathbf{r}-\mathbf{b}| > a, \end{cases} \tag{12}$$

$$s_{\mathbf{b}lm} = s_l = \frac{l}{2l+1}, \tag{13}$$

while $\hat{\Gamma}$ is represented by the following matrix:

$$\Gamma_{\mathbf{b}lm,\mathbf{b}'l'm'} = (-1)^{l'+m'} \left(\frac{a}{|\mathbf{b}-\mathbf{b}'|}\right)^{l+1/2} \left(\frac{a'}{|\mathbf{b}-\mathbf{b}'|}\right)^{l'+1/2} \left(\frac{ll'}{(2l+1)(2l'+1)}\right)^{1/2} \times \frac{(l+l'+m-m')!}{[(l+m)!(l-m)!(l'+m')!(l'-m')!]^{1/2}} e^{i(m'-m)\varphi_{\mathbf{b}-\mathbf{b}'}} P_{l'+l}^{(m'-m)}[\cos(\theta_{\mathbf{b}-\mathbf{b}'})], \tag{14}$$

where $Y_{lm}(\Omega)$ is the normalized spherical harmonic and $P_l^{(m)}[\cos(\theta)]$ is the associated Legendre polynomial.

In this representation, the eigenstates are found by *numerical diagonalization* of the Γ-matrix, and the normalized-to-one eigenvector $U_{\mathbf{b}lm}^{(n)} = \langle\theta_\mathbf{b}\phi_{\mathbf{b}lm}|\phi_n\rangle \equiv \langle\mathbf{b}lm|\phi_n\rangle$ provides the expansion coefficients for the eigenfunction $\phi_n(\mathbf{r})$ in terms of the isolated particle eigenfunctions $\phi_{\mathbf{b}lm}(\mathbf{r})$:

$$\theta_p(\mathbf{r})\phi_n(\mathbf{r}) = \sum_{\mathbf{b}lm} U_{\mathbf{b}lm}^{(n)} \theta_\mathbf{b}(\mathbf{r})\phi_{\mathbf{b}lm}(\mathbf{r}). \tag{15}$$

The step function $\theta_p(\mathbf{r})$ appears in this expansion in order to signal that it is valid *inside the sub-volume of p-constituent*. The step functions $\theta_\mathbf{b}(\mathbf{r})$ appear in this expansion in order to ensure that a basis function $\phi_{\mathbf{b}lm}(\mathbf{r})$ is only used *inside the sub-volume of its particle* $\mathbf{b}$.

The scalar products which appear in the expansion for $\phi(\mathbf{r})$ [Eq. (10)] are calculated as follows, using the fact that the function $z = b_z + \sqrt{V_\mathbf{b}}\phi_{\mathbf{b}10}(\mathbf{r})$ for $\mathbf{r}$ inside the sphere centered at $\mathbf{b}$, which results in $\langle\mathbf{b}lm|z\rangle = \sqrt{V_\mathbf{b}}\,\delta_{lm,10}$ ($V_\mathbf{b} = 4\pi a^3/3$ is the volume of that sphere):

$$\langle\phi_n|z\rangle = \sum_{\mathbf{b}lm} \langle\phi_n|\mathbf{b}lm\rangle\langle\mathbf{b}lm|z\rangle = \sum_\mathbf{b} \sqrt{V_\mathbf{b}} \left(U_{\mathbf{b}10}^{(n)}\right)^* . \tag{16}$$

Substituting Eqs. (15) and (16) in Eq. (10), we get the following expansion for $\phi(\mathbf{r})$ in terms of $\phi_{\mathbf{b}lm}(\mathbf{r})$, which is valid inside the p-constituent:

$$\theta_p(\mathbf{r})\phi(\mathbf{r}) = \sum_{\mathbf{b}lm} A_{\mathbf{b}lm}(s)\theta_{\mathbf{b}}(\mathbf{r})\phi_{\mathbf{b}lm}(\mathbf{r}), \tag{17}$$

$$A_{\mathbf{b}lm}(s) = \sum_{n\mathbf{b}'} \frac{s}{s-s_n} U^{(n)}_{\mathbf{b}lm} \left(U^{(n)}_{\mathbf{b}'10}\right)^* \sqrt{V_{\mathbf{b}'}}. \tag{18}$$

3. CALCULATING THE FORCES IN A COLLECTION OF SPHERES

We recall that the total electric energy of the system is given by

$$W = \frac{1}{8\pi}\int dV \varepsilon(\mathbf{r})|\nabla\phi(\mathbf{r})|^2. \tag{19}$$

From this it follows that the force exerted on the sphere centered at $\mathbf{b}$ is given by the following partial derivative

$$\mathbf{F}_{\mathbf{b}} = +\frac{\partial W}{\partial \mathbf{b}}, \tag{20}$$

where all the other micro-structural parameters (i.e., all the other $\mathbf{b}'$) are kept fixed, as well as the externally applied field $\mathbf{E}_0 = \mathbf{e}_z$. Keeping the externally applied field fixed is equivalent to keeping fixed the potential difference between two large capacitor plates, where the entire system is situated. In that case, the force on a particle is given by the appropriate gradient of W with a *plus sign* rather than a *minus sign*.[†]

A word of caution is in order here: Although it is well known that Eq. (19) is stationary with respect to small variations in the potential field $\phi(\mathbf{r})$, we cannot use this property to calculate the derivative of Eq. (20) by considering only the change in $\varepsilon(\mathbf{r})$ which appears explicitly in the integrand of Eq. (19). This is due to the fact that the changes in $\phi(\mathbf{r})$ are not small in the neighborhood of the interface between the two constituents. I.e., at the surface of a particle $\phi(\mathbf{r})$ is not a differentiable function of the position $\mathbf{b}$ of that particle.

Keeping this cautionary note in mind, one can still calculate the derivative of Eq. (20) in terms of the unperturbed electric field on the two sides of that interface. In order to do this, we first calculate the change δW in W, which results from a small change $\delta\mathbf{b}$ in the position of the particle $\mathbf{b}$. The resulting small change δW is expressed in terms of the local changes $\delta\varepsilon(\mathbf{r})$, $\delta\mathbf{E}(\mathbf{r})$, which are not necessarily small near the surface of the particle $\mathbf{b}$—see Fig. 1:

$$8\pi\delta W = \int dV(\varepsilon+\delta\varepsilon)(\mathbf{E}+\delta\mathbf{E})^2 - \int dV\varepsilon\mathbf{E}^2 = 2\int dV\varepsilon(\mathbf{E}\cdot\delta\mathbf{E}) + \int dV\varepsilon(\delta\mathbf{E})^2 + \int dV\delta\varepsilon(\mathbf{E}+\delta\mathbf{E})^2. \tag{21}$$

The first integral on the right hand side (rhs) is easily shown to vanish:

$$\int dV\varepsilon(\mathbf{E}\cdot\delta\mathbf{E}) = \int dV(\mathbf{D}\cdot\nabla\delta\phi) = \int dV\nabla\cdot(\mathbf{D}\delta\phi) - \int dV\delta\phi(\nabla\cdot\mathbf{D}). \tag{22}$$

On the rhs, the second integral vanishes because $\nabla\cdot\mathbf{D} = 0$ everywhere, while the first integral vanishes after transformation to a surface integral, where $\delta\phi = 0$ everywhere. The second integral on the rhs of Eq. (21) is evaluated to lowest order in $\delta\mathbf{b}$ by noting that $\delta\mathbf{E}(\mathbf{r})$ is small everywhere except in the two small volumes V_1, V_2, which represent the changed position of the spherical particle $\mathbf{b}$—see Fig. 1:

$$\delta\mathbf{E}(\mathbf{r}) = \begin{cases} [E_r(in) - E_r(out)]\,\mathbf{e}_r, & \text{in } V_1 \\ [E_r(out) - E_r(in)]\,\mathbf{e}_r, & \text{in } V_2 \\ \text{small}, & \text{elsewhere.} \end{cases} \tag{23}$$

[†]Specifically, when the spheres are moved at fixed potential on the boundaries, the energy W of the system changes. However, the batteries which hold the potential fixed supply twice as much energy to the system as the change in W. For a discussion of differences between force calculations at fixed potential and fixed charge, see, e.g., J. D. Jackson, *Classical Electrodynamics*, 3rd ed. (Wiley, New York, 1999), pp. 167–169

Here $E_r(in)$, $E_r(out)$ denote the radial component of $\mathbf{E}(\mathbf{r})$ just inside and just outside the surface of the undisplaced sphere. Of course, the two values differ by a non-small amount, even at very nearby points, due to the discontinuity in the radial component of $\mathbf{E}(\mathbf{r})$. As explained above, only the small volumes V_1, V_2 make a non-negligible contribution (i.e., first order in the small quantity $|\delta\mathbf{b}|$) to the second integral on the rhs of Eq. (21). Assuming that $\delta\mathbf{b} \parallel z$, this contribution is easily found to be given by

$$\begin{aligned}\frac{1}{a_{\mathbf{b}}^2|\delta\mathbf{b}|}\int dV\varepsilon(\delta\mathbf{E})^2 \quad &\cong \quad \int_{0<\theta_{\mathbf{r}-\mathbf{b}}<\pi/2} d\Omega_{\mathbf{r}-\mathbf{b}}\;\varepsilon_p\;[E_r(in)-E_r(out)]^2\cos\theta_{\mathbf{r}-\mathbf{b}} \\ &\quad - \int_{\pi/2<\theta_{\mathbf{r}-\mathbf{b}}<\pi} d\Omega_{\mathbf{r}-\mathbf{b}}\;\varepsilon_h\;[E_r(in)-E_r(out)]^2\cos\theta_{\mathbf{r}-\mathbf{b}}. \end{aligned} \tag{24}$$

Here and later, $\theta_{\mathbf{r}-\mathbf{b}}$, $\varphi_{\mathbf{r}-\mathbf{b}}$ denote polar angles with respect to the center of the sphere at $\mathbf{b}$. Due to the appearance of $\delta\varepsilon(\mathbf{r})$ in the integrand of the last integral on the rhs of Eq. (21), that integration is also limited to the two small volumes V_1, V_2. Thus we get

$$\frac{1}{a_{\mathbf{b}}^2|\delta\mathbf{b}|}\int dV\delta\varepsilon(\mathbf{E}+\delta\mathbf{E})^2 \cong \int_{0<\theta_{\mathbf{r}-\mathbf{b}}<\pi/2} d\Omega_{\mathbf{r}-\mathbf{b}}(\varepsilon_p-\varepsilon_h)\mathbf{E}^2(in) + \int_{\pi/2<\theta_{\mathbf{r}-\mathbf{b}}<\pi} d\Omega_{\mathbf{r}-\mathbf{b}}(\varepsilon_h-\varepsilon_p)\mathbf{E}^2(out). \tag{25}$$

Since the two radial field components at the sphere surface are connected by $\varepsilon_p E_r(in) = \varepsilon_h E_r(out)$, we can easily write all the above integrals in terms of just the *in* field, which is more convenient because we can compute it directly from the expansion for $\phi(\mathbf{r})$ of Eq. (17). Some algebra finally leads to the following expression for the z-component of the force acting on the sphere centered at $\mathbf{b}$:

$$F_z = \frac{a_{\mathbf{b}}^2}{8\pi}\int d\Omega_{\mathbf{r}-\mathbf{b}}\left[(\varepsilon_p-\varepsilon_h)\mathbf{E}^2(in) + \frac{(\varepsilon_p-\varepsilon_h)^2}{\varepsilon_h}E_r^2(in)\right]\cos\theta_{\mathbf{r}-\mathbf{b}}. \tag{26}$$

Note that the angular integration is now over the entire solid angle. A similar procedure, starting with displacements $\delta\mathbf{b} \parallel x$ or $\delta\mathbf{b} \parallel y$ leads to the other two force components

$$F_x = \frac{a_{\mathbf{b}}^2}{8\pi}\int d\Omega_{\mathbf{r}-\mathbf{b}}\left[(\varepsilon_p-\varepsilon_h)\mathbf{E}^2(in) + \frac{(\varepsilon_p-\varepsilon_h)^2}{\varepsilon_h}E_r^2(in)\right]\sin\theta_{\mathbf{r}-\mathbf{b}}\cos\varphi_{\mathbf{r}-\mathbf{b}}, \tag{27}$$

$$F_y = \frac{a_{\mathbf{b}}^2}{8\pi}\int d\Omega_{\mathbf{r}-\mathbf{b}}\left[(\varepsilon_p-\varepsilon_h)\mathbf{E}^2(in) + \frac{(\varepsilon_p-\varepsilon_h)^2}{\varepsilon_h}E_r^2(in)\right]\sin\theta_{\mathbf{r}-\mathbf{b}}\sin\varphi_{\mathbf{r}-\mathbf{b}}. \tag{28}$$

When this approach is applied to a collection of spherical particles, the electric potential field $\phi(\mathbf{r})$ inside any sphere is expanded in the (spherical harmonic) eigenstates of Eq. (12), with expansion coefficients $A_{\mathbf{b}lm}(s)$ given by Eq. (18). Using that expansion, the electric field $\mathbf{E}(in)$ can also be written in terms of spherical harmonics inside that sphere. Consequently, the integrals which appear in Eqs. (26)–(28) can be evaluated, term by term in the expansion, in closed form, leading to the following expressions for the force components:

$$8\pi a_{\mathbf{b}}F_x = (\varepsilon_p-\varepsilon_h)\sum_{lm}(-1)^m\alpha_{lm}\left(\frac{2l+3}{l+1}+\frac{\varepsilon_p-\varepsilon_h}{\varepsilon_h}\right)\left(A_{\mathbf{b}lm}A_{\mathbf{b}\,l+1\,m-1} - A_{\mathbf{b}\,l\,-m}A_{\mathbf{b}\,l+1\,m+1}\right), \tag{29}$$

$$8\pi a_{\mathbf{b}}F_y = (\varepsilon_p-\varepsilon_h)\sum_{lm}(-1)^m\alpha_{lm}\left(\frac{2l+3}{l+1}+\frac{\varepsilon_p-\varepsilon_h}{\varepsilon_h}\right)\left(A_{\mathbf{b}lm}A_{\mathbf{b}\,l+1\,m-1} + A_{\mathbf{b}\,l\,-m}A_{\mathbf{b}\,l+1\,m+1}\right), \tag{30}$$

$$8\pi a_{\mathbf{b}}F_z = (\varepsilon_p-\varepsilon_h)\sum_{lm}(-1)^m\beta_{lm}\left(\frac{2l+3}{l+1}+\frac{\varepsilon_p-\varepsilon_h}{\varepsilon_h}\right)\left(A_{\mathbf{b}lm}A_{\mathbf{b}\,l+1\,-m} + A_{\mathbf{b}\,l\,-m}A_{\mathbf{b}\,l+1\,m}\right), \tag{31}$$

where

$$\alpha_{lm} \equiv \left(\frac{l(l+1)(l+m+1)(l+m+2)}{(2l+1)(2l+3)}\right)^{1/2}, \quad \beta_{lm} \equiv \left(\frac{l(l+1)(l+m+1)(l-m+1)}{(2l+1)(2l+3)}\right)^{1/2}. \tag{32}$$

To illustrate the usefulness of this approach, we considered a linear array of three metal spheres with monotonically increasing radii $a_1 = 1\,\mu$m, a_2, a_3, where $a_3/a_2 = a_2/a_1$, and smallest separations between neighboring

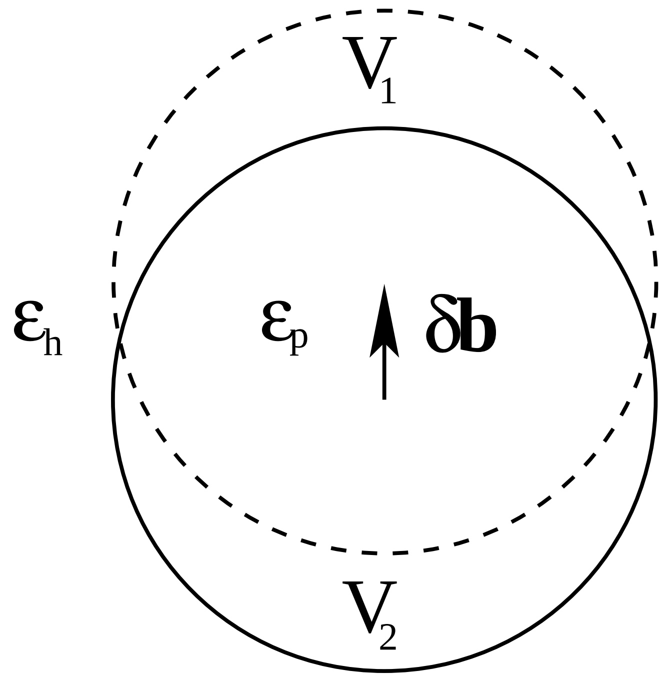

Figure 1. Spherical particle centered at $\mathbf{b}$ (solid line) and displaced by small amount $\delta\mathbf{b}$ (dashed line). In the small volume V_1, the change in permittivity is $\varepsilon_p - \varepsilon_h$, while in the small volume V_2, the change in permittivity is $\varepsilon_h - \varepsilon_p$. Everywhere else the permittivity is unchanged. If $\delta\mathbf{b}$ is small, then so are the changes in the local electric field $\mathbf{E}(\mathbf{r})$ everywhere, except for the two small volumes V_1, V_2—see Eq. (23).

spheres (i.e., polar gaps) $\delta_{12} = a_1/100$, $\delta_{23} = a_2/100$. For such particles, $\varepsilon_p \to \infty$, therefore $s \to 0$. This does not affect the numerical computation of the eigenvectors $U^{(n)}_{\mathbf{b}lm}$, which do not depend on the material parameters of the constituents ε_p, ε_h, or on s. The expression for $A_{\mathbf{b}lm}(s)$ actually simplifies in this limit, as do the expressions for the force components. In Fig. 2 we plot the force F_z acting upon the smallest sphere, as function of the radii ratio $a_3/a_2 = a_2/a_1$, when an external electric field of magnitude 1000 V/cm is applied along the array axis. For comparison we also plot that force when a dipole approximation is used to describe the polarization of each sphere. We also plot the force obtained as a sum of two body forces exerted on the smallest sphere by each of the other spheres *when the third sphere is absent*. Clearly, the last two calculations give very bad results. Thus, not only is the dipole approximation bad, but the forces cannot be considered as sums of two-body forces. Many body forces are crucially important when the spheres are close to each other. That is a consequence of the fact that, when a particle is displaced, the polarization of all the particles changes, changing the electric field felt by that particle in a complicated fashion which depends on the detailed micro-geometry.

We also considered a pair of different sized metal spheres, with radii $a_1 = 1\,\mu$m, $a_2 = 2a_1$, and polar separation $\delta = a_1/10$, centered in the xy-plane and subject to an external uniform field $\mathbf{E}_0 = 1000\,$V/cm along y. In Fig. 3 we show the radial and azimuthal force components acting on the smaller sphere, as function of the azimuthal angle θ between the relative position vector $\mathbf{b}_1 - \mathbf{b}_2$ and the y-axis. We also plot, for comparison, the results for those forces in the dipole approximation.

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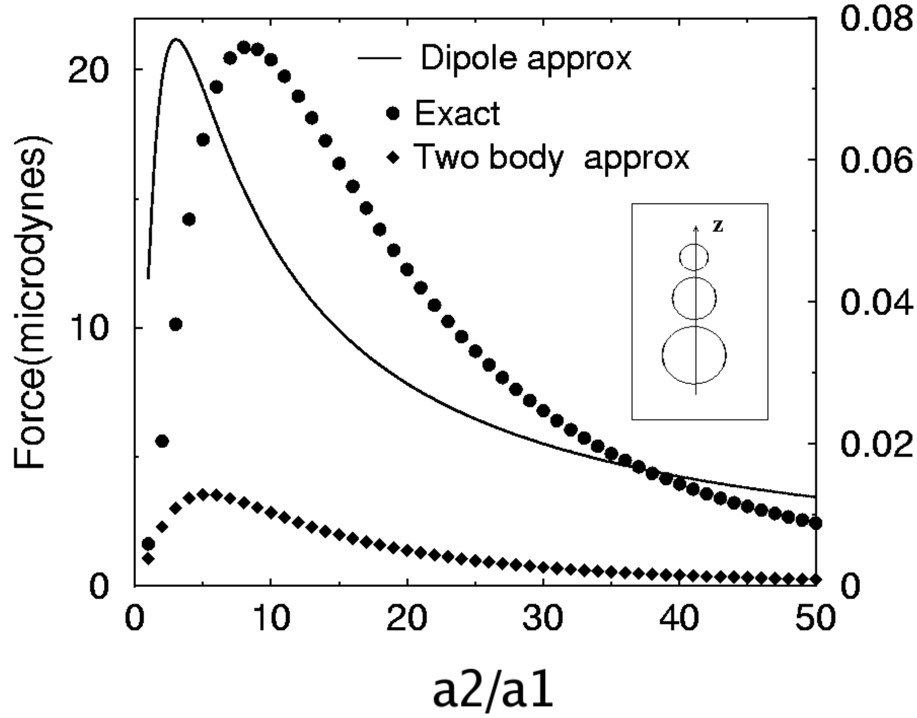

Figure 2. Downwards force exerted on smallest metal sphere in a three-sphere nano-lens, plotted versus the common radii ratio $a_2/a_1 = a_3/a_2$. The external electric field $\mathbf{E}_0$ lies along the line through the three sphere centers, its magnitude is $1000\,\mathrm{V/cm}$, the radius of the smallest sphere is $a_1 = 1\mu\mathrm{m}$, and the inter-sphere gaps are $\delta_{12} = 0.01a_1$, $\delta_{23} = 0.01a_2$. The force scale for the dipole approximation results only is on the right, while for the other two curves it is on the left (after Ref. 5).

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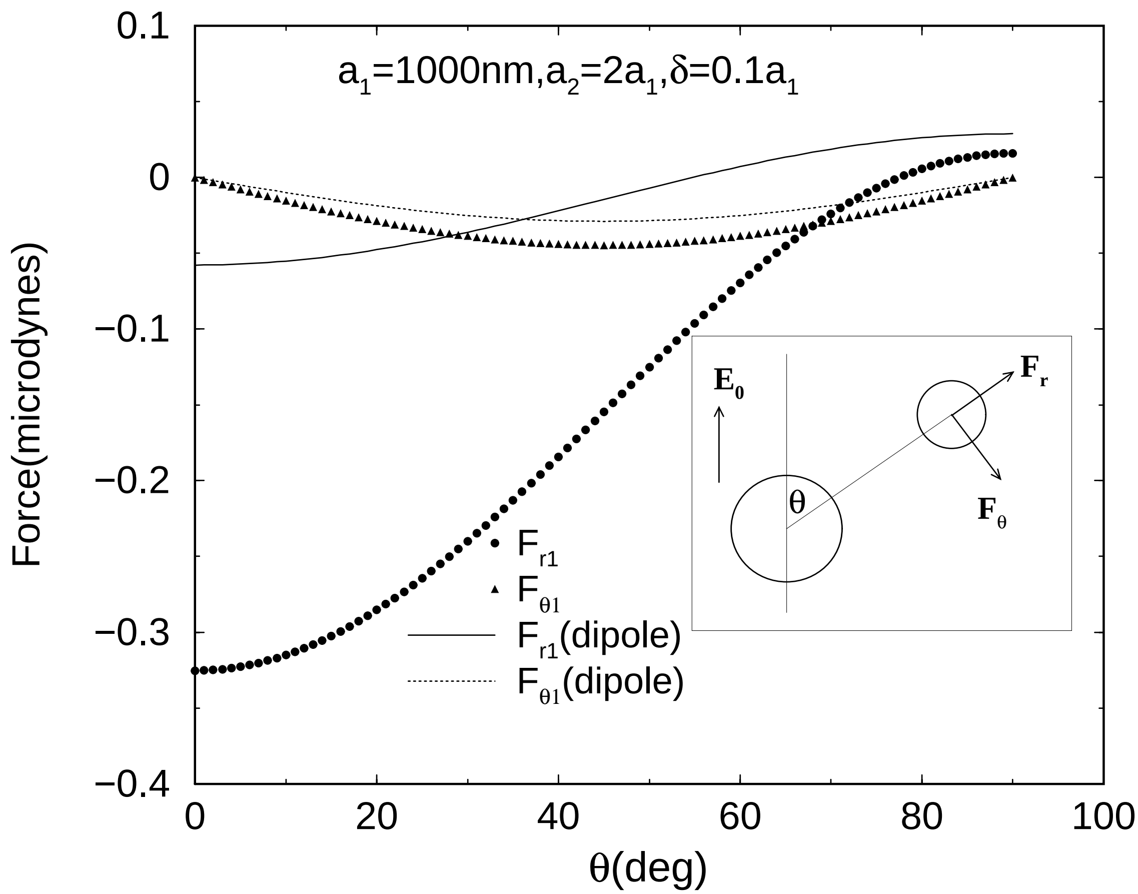

Figure 3. Force components F_{r1}, $F_{\theta 1}$ exerted on sphere No. 1 (the one on the upper right in the inset) when two spheres are placed in a uniform external field $\mathbf{E}_0$, directed at an angle θ with respect to the line through the sphere centers. The field magnitude is $1000\,\mathrm{V/cm}$, and the radius of the smaller sphere is $a_1 = 1\mu\mathrm{m}$. The dipole approximation results for this configuration are also plotted for comparison (after Ref. 5).