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The effect of dissipation on the torque and force experienced by nanoparticles in an AC field

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We discuss the force and torque acting on spherical particles in an ensemble in the presence of a uniform AC electric field. We show that for a torque causing particle rotation to appear the particle must be absorptive. Our proof includes all electromagnetic excitations, which in the case of two or more particles gives rise to one or more resonances in the spectrum of force and torque depending on interparticle distance. Several peaks are found in the force and torque between two spheres at small interparticle distances, which coalesce to just one as the separation grows beyond three particle radii. We also show that in the presence of dissipation the force on each particle is nonconservative and may not be derived from the classical interaction potential energy as has been done in the past.

Keywords: Nanoparticle dynamics; force and torque; electrorotation; nonconservative mechanical system; dissipation; polarization; multipole resonance.

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1. Introduction

The electromagnetic excitations and ensuing dynamics of nanoparticles, molecules and atoms in the presence of an electric field has been widely studied both theoretically and experimentally.^{1–5} The particles may initially be unpolarized, but due to the external field and their mutual interaction they may acquire induced dipole and higher electric moments. As a consequence electric forces and torques are produced, resulting in particle motion and the formation of equilibrium configurations. An important case is optical trapping and binding, which, if many particles are involved may lead to self-assembly of ordered structures.⁶ Structures are also formed in electro-rheological fluids, where a static or slowly varying field induces the formation of linear arrays and columns in a medium containing polarizable spheres in suspension.⁷ Examples where an understanding of forces and torques is also crucial are the dielectrophoresis and electrorotation effects, related to motion in a nonuniform field⁸ and a rotating AC field,⁹ respectively. Other applications include the control of agglomeration, and the separation of proteins or living cells in suspension.¹⁰ Nanorotors driven by a light force have also been studied.^{11,12}

Several methods have been used to obtain forces^{13–16} and torques^{17–20} in the past, some involving the use of an interaction potential energy whose gradient is taken to obtain the force.^{13,21–23} In this work we prove that if the particles are absorptive the system is nonconservative and the net force experienced by each member of the ensemble may not be derived from an interaction potential. In fact, we show explicitly that structure in the interaction energy arising from absorption resonances in a pair of gold nanospheres exhibits energy minima leading to unphysical equilibrium configurations that are not present if the force is calculated directly from Coulomb's law.^{23,24} Away from such resonances when absorption is negligible either method may be used leading to similar results.

In order to obtain explicit expressions for the force and torque we assume the particles to be spherical, thus allowing a multipolar analysis and a comparison with results obtained in the dipole approximation. For an AC external excitation we find the dipole approximation to give accurate results if the center to center separation between neighbors is not smaller than three particle radii, while at closer interparticle distance the inclusion of all multipoles gives rise to several resonances in the force and torque strength, shifted to lower frequencies owing to particle–particle couplings. This is in accordance with previous results on the electric excitation of dielectric particles arrays showing a similar distance dependent behavior.^{25–28} Location of such resonances in the frequency spectrum may be useful in applications when the force or torque strength becomes important. Within the same model we find that the appearance of a torque causing particle rotation requires that the particle be dissipative.

The paper is organized as follows. In Sec. 2 we present compact expressions for the time-averaged force and torque acting over a particle in an arbitrary array of nanoparticles in a uniform AC electric field. The very structure of the resulting expressions reveals the need for dissipation in order for a torque to arise. The cases of linear and circular polarizations are discussed. In order to assess the relevance of higher multipoles in both forces and torque, in Sec. 3 we apply our model to two gold nanospheres in an electric field parallel or perpendicular to the interparticle axis. In Sec. 4 we prove that the presence of dissipation makes the system nonconservative and in Sec. 5 we present our conclusions. Finally, two appendices are added to provide details of the calculations.

2. Forces and Torques on Interacting Particles in an AC Field

We consider a system of nanoparticles embedded in a non-absorptive dielectric medium, excited by an external AC electric field of angular frequency ω . The particles are uncharged and their material response to a local electric field may in general be characterized by a complex response function $\epsilon(\omega)$. The external field induces a dipole moment on each particle, which in turn excites multipoles on every other member of the ensemble owing to the nonuniformity of the electric field it produces at each particle site. For simplicity we shall assume in what follows that the particles are of spherical shape.

As known, for a dilute system with average center-to-center separation of the order of three times the particle radii or more, the accuracy of the dipole approximation is acceptable and the effect of higher multipoles may be neglected.²⁵ In such a case and if only two particles are present, the electric force between them may be simply obtained by direct application of the discrete form of Coulombs law, as described in Ref. 24. When separations less than three particle radii become involved however, the effect of higher multipoles must be included.^{26,27} The general form of Coulomb's law to be used is then,

$$\langle \mathbf{F}_i \rangle = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) \mathbf{E}(\mathbf{r}) d^3 \mathbf{r} \,, \tag{1}$$

where $\langle \mathbf{F}_i \rangle$ is the time-averaged force on particle *i*, $\rho_i^*(\mathbf{r})$ is its charge density and $\mathbf{E}(\mathbf{r})$ is the local electric field due to the external sources and other particles in the ensemble. A rather lengthy calculation then yields the force Cartesian components (see Appendix A),

$$\langle F_{ix} \rangle = \operatorname{Re} \sum_{l} C_{li} \operatorname{Re} T_{li} , \qquad (2)$$

$$\langle F_{iy} \rangle = \operatorname{Re} \sum_{l} C_{li} \operatorname{Im} T_{li} ,$$
 (3)

$$\langle F_{iz} \rangle = \operatorname{Re} \sum_{l} C_{li} \sum_{m=-l}^{l} \sqrt{(l-m)(l+m)} q_{lmi} q_{l-1,m,i}^{*},$$
 (4)

where the pole order index l here and in what follows covers the range of integers

 $1, \infty$. In the above expressions the coefficient

$$C_{li} = \frac{2\pi}{\sqrt{(2l+1)(2l-1)}\alpha_{li}},$$
(5)

weights the strength with which the multipole of order l contributes, with α_{li} the corresponding particle polarizability, a complex quantity if absorption is present. Also

$$q_{lmi} = \int \rho_i(\mathbf{r}) r^l Y_{lm}^*(\theta, \phi) d^3 \mathbf{r}$$
(6)

is the induced multipole of indices l, m on particle i and

$$T_{li} = \sum_{m} \sqrt{(l-m)(l-m-1)} q_{lmi} q_{l-1,m+1,i}^* \,. \tag{7}$$

 Y_{lm} is the usual complex spherical harmonic function. Methods to obtain the multipoles q_{lmi} for arbitrary configurations are described in Refs. 26 and 27. Notice that since the force involves products of multipoles of different order, if there is a single spherical particle and the external field is uniform only the dipole moment is excited and the force is zero.

Spinning of coupled particles in an external field has been observed in the past.^{20,29} In order to capture this effect we consider next the time-averaged torque on sphere i due to the local field, as given by

$$\langle \boldsymbol{\tau}_i \rangle = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) \mathbf{r} \times \mathbf{E}(\mathbf{r}) d^3 \mathbf{r} ,$$
 (8)

where the origin is taken at the particle center. Work similar to that done above for the forces (see Appendix B) leads to the time-averaged torque Cartesian components

$$\langle \tau_{ix} \rangle = \operatorname{Im} \sum_{l=1}^{\infty} D_{li} \operatorname{Re} S_{li} ,$$
(9)

$$\langle \tau_{iy} \rangle = \operatorname{Im} \sum_{l=1}^{\infty} D_{li} \operatorname{Im} S_{li} ,$$
 (10)

$$\langle \tau_{iz} \rangle = \operatorname{Im} \sum_{l=1}^{\infty} D_{li} \sum_{m=-l}^{l} m |q_{lmi}|^2, \qquad (11)$$

where the coefficients

$$D_{li} = \frac{2\pi}{(2l+1)\alpha_{li}} \tag{12}$$

are complex if α_{li} is, and

$$S_{li} = \sum_{m=-l}^{l-1} \sqrt{(l-m)(l+m+1)} q_{lmi} q_{l,m+1,i}^* \,.$$
(13)

It is clear from Eqs. (9)–(13) that if the system has no dissipation, i.e., if α_{li} is real, the torque is zero. We conclude that in general a torque arises in such systems from dissipative electromagnetic interactions.

Even if there is dissipation however, the torque may be suppressed by special symmetries. Such is the case for a linear array subject to a uniform electric field parallel to the line joining their centers. By choosing the z-axis to be aligned with this line, only modes with m = 0 are excited leading to zero torque, as may be easily verified from the structure of the above equations. A similar situation occurs if the applied electric field lies on the xy plane since in this case only modes with $m = \pm 1$ are excited symmetrically and the torque is again zero. Nevertheless, it is worth noting that if the linear array is under a uniform electric field with components along the z-axis and the xy plane, modes with $m = \pm 1$ and m = 0 become excited. So, according to Eqs. (9)–(11) a torque is produced provided that electromagnetic dissipation is not negligible. A similar situation has been analyzed in Ref. 20 in the dipolar approach.

A torque does arise in such arrays also if they are subject to a rotating electric field on the xy plane. The field may be written as $\mathbf{E} = E_0(\pm \hat{x} - i\hat{y})e^{i\omega t}$ and the corresponding coefficients of expansion of the potential are either $V_{1,+1} = \sqrt{2\pi/3}E_0(1-i)$ or $V_{1,-1} = \sqrt{2\pi/3}E_0(-1-i)$ depending of the sense of rotation of the electric field vector given by the sign of the *x*-component.³⁰ Correspondingly the excited modes are either m = 1 or m = -1 and from Eq. (11) it follows that a torque may appear. In fact, from Eqs. (A.35) and (11) it can be shown that for this case the time-average of the *z*-component of the torque is given by

$$\langle \tau_{iz} \rangle = \frac{2\pi m}{a^{2l+1}} \sum_{l} \frac{l \operatorname{Im} \epsilon}{[l(\operatorname{Re} \epsilon - 1)]^2 + [l \operatorname{Im} \epsilon]^2} |q_{lmi}|^2.$$
(14)

For the special case of a single sphere in a rotating external field the torque is finite, in agreement with Refs. 31 and 32. The physical origin of such a torque is conservation of angular momentum. The rotating field carries angular momentum, which is transferred to the particles when absorption takes place causing them to experience a spinning torque. Also, as noted in Ref. 20 when a linearly polarized field is not aligned with a symmetry axis of a linear array such as a pair, the local field at each particle site has a rotating component, and the same argument applies.

3. Special Case: Two Particles

We shall apply our general results to the simplest case, that of two identical spheres of radii *a* subject to a uniform oscillating electric field, both parallel and perpendicular to a line joining the spheres centers, that we choose to be the *z*-axis. These conditions will be referred to as parallel and perpendicular excitation, respectively. In computing the force we found convenient to use Eq. (A.33) in Appendix A with the replacement $V_{lmi} = b_{lmi}$, since the uniform external field produces no direct

force. Using relation (A.7) then leads to,

$$\langle F_{iz} \rangle = -\frac{1}{2} \operatorname{Re} \sum_{lm} \sum_{l'm'} \sum_{j \neq i} (-1)^{l'} A_{lmi}^{l'm'j} \sqrt{\frac{(2l+1)}{(2l-1)}} (l-m)(l+m) q_{l'm'j} q_{l-1,m,i}^*,$$
(15)

where the coefficient $A_{lmi}^{l'm'j}$ that couples multipoles in different particles is given by Eq. (A.8) in Appendix A.

3.1. Parallel excitation

In this geometry $\mathbf{E} = E_0 e^{i\omega t} \hat{z}$ and modes with m = 0 become excited only, yielding a force along the z-axis. From Eqs. (2), (3) and (7) it is seen that the time-averaged value of the components x and y of the force is zero, as expected from symmetry considerations. Using Eq. (15) we find after some algebra the force component on sphere 1 centered at the origin

$$\langle F_{1z} \rangle = -2\pi \operatorname{Re} \sum_{ll'} (-1)^{l'} \frac{(l+l'+1)!}{l!l'! \sqrt{(2l+1)(2l'+1)}} q_{l,0,1}^* q_{l,0,1}^{l} q_{l',0,2}^{l'}, \qquad (16)$$

where the multipole moments may be obtained using the formalism of Ref. 27. Here R is the center to center distance between the two spheres. The dipole approximation applies keeping the first term in this series, (l = l' = 1) and the result agrees with that in Ref. 24 as it should.

3.2. Perpendicular excitation

In this case the external field is in the xy plane, and the external potential in Eq. (A.10) of Appendix A is expressed as $V^{\text{ext}} = V_{1,1,i}rY_{1,1}(\theta,\phi) + V_{1,-1,i}rY_{1,-1}(\theta,\phi)$ with $V_{1,\pm 1} = \sqrt{2\pi/3}(\pm E_x - iE_y)$. The coupling coefficients in Eq. (A.8) are null unless $m = m' = \pm 1$. From Eq. (15) we get this time,

$$\langle F_{1z} \rangle = 2\pi \operatorname{Re} \sum_{ll'} (-1)^{l'} \frac{(l+l'+1)!}{l!l'!R^{l+l'+2}} \\ \times \sqrt{\frac{ll'}{(2l+1)(2l'+1)(l+1)(l'+1)}} \left[q_{l,1,1}^* q_{l',1,2} + q_{l,-1,1}^* q_{l',-1,2} \right].$$
(17)

Keeping just the l = l' = 1 term in the series the dipole approximation is obtained, which agrees with the corresponding expression in Ref. 24.

3.3. Numerical results

We next show some numerical results for our test case of two particles. We use a Drude dielectric function with parameters $\epsilon_b = 9.9$, $\hbar\omega_p = 8.2$ eV, $\Gamma = 0.053$ eV, appropriate for gold nanospheres.³³ Figure 1 shows the average force for parallel



Fig. 1. Electric force between two identical gold nanospheres as a function of frequency of the applied field, with separation 2.005a between their centers. The solid (dashed) curve corresponds to parallel (perpendicular) excitation calculated including multipoles up to L = 40. The dash-dotted curve corresponds to the average force calculated for parallel excitation and separation 3a, with an amplification factor 1000.

(solid line) as well as perpendicular (dashed line) excitation. One particle is at the origin, while the other is at z = R. The separation is R = 2.005a and we have included multipoles up to order L = 40 in the computation, following the convergence criterion given in Ref. 26. The force acting on the particle at the origin is attractive (positive) in the parallel configuration and repulsive (negative) in the perpendicular geometry, as expected. Three multipolar resonances are clearly resolved at this separation, with force peaks greatly enhanced, about three orders of magnitude above the background value. As the separation between the particles is increased the resonances move to higher frequencies, decrease in size and fewer of them become resolved.²⁷ At a center to center separation of about three particle radii and larger, only one resonance is seen. This dipolar peak, at separation R = 3aand parallel excitation, has been included in the figure for comparison with an amplification factor of one thousand (dash-dotted curve).

In Figure 2 we show the z-component of the average torque acting on each nanoparticle as given by Eq. (14). Separations are R = 2.005a (solid curve) and R = 3a (dashed curve). The pair is subject to an electric field whose direction rotates in the plane xy. As for the force, several resonances are resolved at small separation, while beyond about separation R = 3a only one peak is observed. It can be seen that as the spheres become closer other resonances occur at frequencies below the single sphere dipole resonance value $\omega = \omega_p / \sqrt{\epsilon_b + 2}$. These additional resonance frequencies correspond to resonant modes associated with the multipole moments q_{lmi} .



Fig. 2. Time averaged torque acting on a particle for a system of two gold nanospheres subjected to a rotating electric field, as a function of frequency. Separation between their centers are 2.005a (solid curve) and 3a (dashed curve). Results were obtained including multipoles up to L = 40 and L = 10, respectively.

4. Limits in the Use of an Interaction Energy to Obtain the Force

The existence of dissipation makes a system nonconservative. To see this, recall that for ideal electromagnetic arrays where dissipation is absent, the force acting on a particle may be obtained as the gradient with respect to the particle coordinates of the configuration energy W. If the particle makes a virtual displacement $\delta\xi$ the corresponding electric force it is subject to is $F_e = \partial W_e / \partial \xi$, an expression obtained by the energy balance equation,

$$\delta W_{\text{source}} = F_e \delta \xi + \delta W_e \,, \tag{18}$$

where δW_{source} is the energy supplied by the sources to maintain the potentials of the electrodes fixed and δW_e is the variation in the energy stored in the field. It can be shown that for this case $\delta W_{\text{source}} = 2\delta W_e$ so that the expression $F_e = \partial W_e/\partial\xi$ is obtained.^{30,34} Nevertheless, for real systems dissipation effects must be taken into account and that is done adding a term δW_{loss} in the right side of Eq. (18). This term depends on the path followed during the virtual displacement since the polarization in the particle does and the energy loss is determined by its imaginary part. If the particle is brought from point A to point B, to the mechanical work done one must add the energy loss term $\int_0^{\tau} \bar{P}_{\text{abs}} dt$, where \bar{P}_{abs} is the time averaged power absorbed by the system and τ the time taken during the displacement. Both the integrand and the upper limit of this integral depend on the path making the mechanical system nonconservative.

Based on the above argument we state that in a dissipative system it is incorrect to obtain the force as the gradient of a potential. To illustrate the difference between a direct application of Coulomb's law and the use of a potential we consider two polarizable spheres of radius a, a distance R apart in an electric field of frequency ω and amplitude E_0 which for simplicity we choose to be parallel to the line joining the centers. In the dipole approximation the interaction energy is of the form²³

$$W_{\rm int}(R) = U_0 - \frac{1}{2} \operatorname{Re}[\beta_1(R) - \beta)] a^3 E_0^2, \qquad (19)$$

where U_0 is the free-field interaction energy, $\beta = (\epsilon - 1)/(\epsilon + 2)$ with ϵ being the frequency dependent dielectric function of the spheres, $\beta_1(R) = \beta/(1 - \beta/4\sigma^3)$ and $\sigma = R/2a$. Differentiating the second term in Eq. (19) to get the force induced by the external field we obtain

$$F_w(R) = -\frac{a^2 E_0^2}{48\sigma^4} \operatorname{Re} \frac{1}{(n-u)^2},$$
(20)

where $n = (1 - 1/4\sigma^3)/3$ and the complex spectral variable $u = 1/(\epsilon - 1)$ has been used. By contrast, if the direct Coulomb's method is used one gets²⁴

$$F_c(R) = -\frac{a^2 E_0^2}{48\sigma^4} \frac{1}{|n-u|^2}.$$
(21)

The two forms (20) and (21) agree only when the dielectric function is real, and dissipation is absent. In Fig. 3 we compare the force obtained using these two expressions for a pair of gold nanospheres with a dielectric function as described in Sec. 3. As can be observed while the direct Coulomb's method gives an attractive force at all frequencies, the model based on the gradient of the interaction energy presents two peaks and an unphysical change of sign in the force.



Fig. 3. Force between two identical gold nanospheres in the parallel configuration as a function of frequency, with separation 3*a* between their centers. Solid and dashed curves correspond to the force calculated from Coulomb's law and using the derivative of an interaction potential, respectively.

5. Conclusions

We have shown that in an ensemble of polarizable spheres in an oscillating electric field, the presence of a rotation torque requires the particle material to be dissipative. We also show that energy loss due to dissipation makes the system nonconservative so that it is improper to use an interaction energy to derive the force, an approach that has been employed erroneously in the past.²¹ Our results are an extension of previous work done for the case of an isolated pair using the dipolar model.²⁴ When interparticle distances are shorter than three particle radii it is known that the dipole approximation is not adequate, and higher multipoles must be considered.^{25,27} Electromagnetic resonances associated with such multipoles are known to appear, that should have a mirror spectrum in the forces and torques as well. We have explicitly shown this to be the case in the simple case of a pair.

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Appendix A. Time-Averaged Force

We consider en ensemble of N spheres in the presence of an external electric field. Choosing a coordinate system with origin at the center of particle i, the electric potential at a point in the medium due to the polarized spheres is given by³⁰

$$V(\mathbf{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^{+l} \frac{4\pi}{2l+1} q_{lmi} \frac{Y_{lm}(\theta,\phi)}{r^{l+1}} + \sum_{l=1}^{\infty} \sum_{m=-l}^{+l} \sum_{j=1}^{N} \frac{4\pi}{2l+1} q_{lmj} \frac{Y_{lm}(\bar{\theta}_j,\bar{\phi}_j)}{\bar{R}_j^{l+1}}, \qquad (A.1)$$

where the multipole moment of order l, m in particle j has been defined in Eq. (6). The center of sphere j is at \mathbf{R}_j and $\mathbf{r} - \mathbf{R}_j = (\bar{R}_j, \bar{\theta}_j, \bar{\phi}_j)$ is the position vector of the observation point with respect to the center of sphere j. To uncouple vectors \mathbf{r} and \mathbf{R}_j we use the identities,³⁵

$$\frac{Y_{lm}(\bar{\theta}_j, \bar{\phi}_j)}{\bar{R}_j^{l+1}} = (-1)^{l+m} \left[\frac{2l+1}{4\pi (l+m)!(l-m)!} \right]^{1/2} \left[\frac{\partial}{\partial x} + i \frac{\partial}{\partial y} \right]^m \frac{\partial^{l-m}}{\partial z^{l-m}} \frac{1}{|\mathbf{r} - \mathbf{R}_j|},$$
(A.2)

$$\frac{1}{|\mathbf{r} - \mathbf{R}_j|} = \sum_{l=0}^{\infty} \frac{r_{<}^l}{r_{>}^{l+1}} \frac{4\pi}{2l+1} \sum_{m=-l}^{+l} (-1)^m Y_{lm}(\theta, \phi) Y_{l,-m}(\theta_j, \phi_j) , \qquad (A.3)$$

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$$\frac{\partial^{n}}{\partial z^{n}} Y_{lm}(\theta, \phi) r^{l} = \left[\frac{2l+1}{(2l-2n+1)} \frac{(l+m)!}{(l+m-n)!} \frac{(l-m)!}{(l-m-n)!} \right] Y_{l-n,m}(\theta, \phi) r^{l-n} ,$$
(A.4)

$$\left[\frac{\partial}{\partial x} + i\frac{\partial}{\partial y}\right]^p Y_{lm}(\theta,\phi)r^l = \left[\frac{2l+1}{(2l-2p+1)}\frac{(l-m)!}{(l-m-2p)!}\right]Y_{l-p,m+p}(\theta,\phi)r^{l-p}.$$
(A.5)

In Eq. (A.3) $r_{<}(r_{>})$ is the lower (higher) value between $r = |\mathbf{r}|$ and $R_j = |\mathbf{R}_j|$; Eq. (A.4) is valid for $l \ge n$ and $|m| \le l - n$ while Eq. (A.5) is valid for $l \ge p$ and $-l \le m \le l - 2p$. From Eqs. (A.2)–(A.5) and adding the potential V^{ext} due to the external field, Eq. (A.1) becomes

$$V(\mathbf{r}) = \sum_{l,m} \frac{4\pi}{2l+1} q_{lmi} \frac{Y_{lm}(\theta,\phi)}{r^{l+1}} + \sum_{l,m} b_{lmi} Y_{lm}(\theta,\phi) r^l + V^{\text{ext}}, \qquad (A.6)$$

where

$$b_{lmi} = \sum_{l'm'} \sum_{j \neq i} A_{lmi}^{l'm'j} q_{l'm'j} \,. \tag{A.7}$$

Here $A_{lmi}^{l'm'j}$ is the coupling coefficient between q_{lmi} and $q_{l'm'j}$ (with $i \neq j$)²⁷

$$A_{lmi}^{l'm'j} = (-1)^{m'} \frac{Y_{l+l',m-m'}^*(\theta_{ij},\phi_{ij})}{|R_{ij}|^{l+l'+1}} \\ \times \left[\frac{(4\pi)^3(l+l'+m-m')!(l+l'-m+m')!}{(2l+1)(2l'+1)(2l+2l'+1)(l+m)!(l-m)!(l'+m')!(l'-m')!} \right]^{1/2},$$
(A.8)

and $\mathbf{R}_i - \mathbf{R}_j = (R_{ij}, \theta_{ij}, \phi_{ij})$. Equations (A.6)–(A.8) are general and valid for any array of spherical particles and arbitrary direction of the applied electric field. All expressions here and below are given in Gaussian units.

In order to obtain the average force we use Eq. (1) making the replacement $E(\mathbf{r}) = -\nabla V_i(\mathbf{r})$ for the local electric field due to the polarized system. Here

$$V_i(\mathbf{r}) = \sum_{lm} b_{lmi} r^l Y_{lm}(\theta, \phi) + V^{\text{ext}}(\mathbf{r}) \,. \tag{A.9}$$

If we expand the external potential as

$$V^{\text{ext}}(\mathbf{r}) = \sum_{lm} V_{lmi}^{\text{ext}} r^l Y_{lm}(\theta, \phi) , \qquad (A.10)$$

the above equation may be written in the form

$$V_i(\mathbf{r}) = \sum_{lm} V_{lmi} r^l Y_{lm}(\theta, \phi) , \qquad (A.11)$$

where $V_{lmi} = V_{lmi}^{\text{ext}} + b_{lmi}$.

In order to obtain explicit expressions for the components of the force we first write the spherical harmonics in the above equation in terms of Legendre functions using the relation

$$Y_{lm}(\theta,\phi) = \sqrt{\frac{(2l+1)}{4\pi} \frac{(l-m)!}{(l+m)!}} P_l^m(\cos\theta) e^{im\phi} .$$
(A.12)

Then Eq. (A.11) may be recast as

$$V_i(\mathbf{r}) = \sum_{lm} D_{lmi} r^l P_l^m(\cos\theta) e^{im\phi} , \qquad (A.13)$$

where

$$D_{lmi} = V_{lmi} \sqrt{\frac{(2l+1)}{4\pi} \frac{(l-m)!}{(l+m)!}}.$$
(A.14)

Therefore the spherical components of the electric field are

$$E_r = -\frac{\partial V_i(\mathbf{r})}{\partial r} = -\sum_{lm} D_{lmi} lr^{l-1} P_l^m(\xi) e^{im\phi} , \qquad (A.15)$$

$$E_{\theta} = -\frac{1}{r} \frac{\partial V_{i}(\mathbf{r})}{\partial \theta} = -\sum_{lm} D_{lmi} r^{l-1} \frac{\partial}{\partial \theta} P_{l}^{m}(\xi) e^{im\phi}$$
$$= \sum_{lm} D_{lmi} r^{l-1} \sqrt{1 - \xi^{2}} \frac{\partial}{\partial \xi} P_{l}^{m}(\xi) e^{im\phi} , \qquad (A.16)$$

$$E_{\phi} = -\frac{1}{r\sin\theta} \frac{\partial V_i(\mathbf{r})}{\partial\phi} = -\sum_{lm} D_{lmi} r^{l-1} \frac{1}{\sqrt{1-\xi^2}} P_l^m(\xi) ime^{im\phi} \,. \tag{A.17}$$

In Eqs. (A.15)–(A.17) we have defined $\xi = \cos \theta$. The corresponding Cartesian components of the electric field are given by

$$E_x = E_r \sin \theta \cos \phi + E_\theta \cos \theta \sin \phi - E_\phi \sin \phi , \qquad (A.18)$$

$$E_y = E_r \sin \theta \sin \phi + E_\theta \cos \theta \cos \phi + E_\phi \cos \phi , \qquad (A.19)$$

$$E_z = E_r \cos \theta - E_\theta \sin \theta \,. \tag{A.20}$$

It is useful to calculate linear combinations of E_x and E_y defined as

$$E_+ = E_x + iE_y \,, \tag{A.21}$$

$$E_{-} = E_x - iE_y \,. \tag{A.22}$$

Introducing relations (A.15)–(A.19) into Eq. (A.21) one obtains

$$E_{+} = -\sum_{lm} D_{lmi} r^{l-1} \times \left[l \sqrt{1 - \xi^2} P_l^m(\xi) - \xi \sqrt{1 - \xi^2} \frac{\partial}{\partial \xi} P_l^m(\xi) - \frac{m}{\sqrt{1 - \xi^2}} P_l^m(\xi) \right] e^{i(m+1)\phi}.$$
(A.23)

The effect of dissipation on the torque and force experienced by nanoparticles

The relations

$$(1-\xi^2)\frac{\partial P_l^m(\xi)}{\partial \xi} = (l+m)P_{l-1}^m(\xi) - l\xi P_l^m(\xi), \qquad (A.24)$$

$$(l-m)P_l^m(\xi) - \xi(l+m)P_{l-1}^m(\xi) = \sqrt{1-\xi^2} P_{l-1}^{m+1}(\xi), \qquad (A.25)$$

then lead to

$$E_{+} = -\sum_{lm} D_{lmi} r^{l-1} P_{l-1}^{m+1}(\xi) e^{i(m+1)\phi} .$$
 (A.26)

Using Eq. (A.14) and (A.12) one obtains

$$E_{+} = -\sum_{lm} V_{lmi} r^{l-1} \sqrt{\frac{2l+1}{2l-1}(l-m)(l-m-1)} Y_{l-1,m+1}(\theta,\phi) \,. \tag{A.27}$$

Similarly, from Eqs. (A.22) and (A.15)–(A.19) follows

$$E_{-} = -\sum_{lm} D_{lmi} r^{l-1} \times \left[l \sqrt{1 - \xi^2} P_l^m(\xi) - \xi \sqrt{1 - \xi^2} \frac{\partial}{\partial \xi} P_l^m(\xi) + \frac{m}{\sqrt{1 - \xi^2}} P_l^m(\xi) \right] e^{i(m+1)\phi} .$$
(A.28)

The recurrence relation $\xi P_{l-1}^{m}(\xi) - P_{l}^{m}(\xi) = (l + m - 1)\sqrt{1 - \xi^2} P_{l-1}^{m-1}(\xi)$ and Eq. (A.24) can be used to find that

$$E_{-} = \sum_{lm} V_{lmi} r^{l-1} \sqrt{\frac{2l+1}{2l-1}(l-m)(l+m-1)} Y_{l-1,m-1}(\theta,\phi) \,. \tag{A.29}$$

To obtain E_z we use the relation $(1 - \xi^2)(\partial P_l^m(\xi)/\partial \xi) = (l+m)P_{l-1}^m(\xi) - l\xi P_l^m(\xi)$, and Eqs. (A.15), (A.16) and (A.20) to give

$$E_z = -\sum_{lm} V_{lmi} r^{l-1} \sqrt{\frac{2l+1}{2l-1}(l-m)(l+m)} Y_{l-1,m}(\theta,\phi) \,. \tag{A.30}$$

The Cartesian components of the time-averaged force acting upon sphere i are then given by

$$\langle F_{ix} \rangle = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) E_x d^3 \mathbf{r} = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) \frac{1}{2} (E_+ + E_-) d^3 \mathbf{r}$$

$$= -\frac{1}{4} \operatorname{Re} \int \rho_i^*(\mathbf{r}) \sum_{lm} V_{lmi} r^{l-1} \sqrt{\frac{2l+1}{2l-1}} \times \left[\sqrt{(l-m)(l-m-1)} Y_{l-1,m+1}(\theta,\phi) - \sqrt{(l+m)(l+m-1)} Y_{l-1,m-1}(\theta,\phi) \right] d^3 \mathbf{r}$$

$$= -\frac{1}{4} \operatorname{Re} \sum_{lm} V_{lmi} \sqrt{\frac{2l+1}{2l-1}} \times \left[\sqrt{(l-m)(l-m-1)} q_{l-1,m+1,i}^* - \sqrt{(l+m)(l+m-1)} q_{l-1,m-1,i}^* \right],$$
(A.31)

$$\langle F_{iy} \rangle = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) E_y d^3 \mathbf{r} = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) \frac{i}{2} (-E_+ + E_-) d^3 \mathbf{r}$$

$$= \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) \frac{i}{2} \sum_{lm} V_{lmi} r^{l-1} \sqrt{\frac{2l+1}{2l-1}}$$

$$\times \left[\sqrt{(l-m)(l-m-1)} Y_{l-1,m+1}(\theta,\phi) + \sqrt{(l+m)(l+m-1)} Y_{l-1,m-1}(\theta,\phi) \right] d^3 \mathbf{r}$$

$$= \frac{1}{4} \operatorname{Re} i \sum_{lm} V_{lmi} \sqrt{\frac{2l+1}{2l-1}} \times \left[\sqrt{(l-m)(l-m-1)} q_{l-1,m+1,i}^* + \sqrt{(l+m)(l+m-1)} q_{l-1,m-1,i}^* \right],$$
(A.32)

$$\langle F_{iz} \rangle = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) E_z d^3 \mathbf{r}$$

= $-\frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) \sum_{lm} V_{lmi} r^{l-1} \sqrt{\frac{2l+1}{2l-1}(l-m)(l+m)} Y_{l-1,m}(\theta,\phi) d^3 \mathbf{r}$
= $-\frac{1}{2} \operatorname{Re} \sum_{lm} V_{lmi} \sqrt{\frac{2l+1}{2l-1}(l-m)(l+m)} q_{l-1,m,i}^*$. (A.33)

The coefficients V_{lmi} and q_{lmi} are related by²⁶

$$q_{lmi} = -\frac{2l+1}{4\pi} \alpha_{li} V_{lmi} , \qquad (A.34)$$

where α_{li} is the multipole polarizability of the sphere *i* given by²⁷

$$\alpha_{li} = \frac{l(\epsilon - 1)}{l(\epsilon + 1) + 1} a_i^{2l+1} \,. \tag{A.35}$$

We next use relation (A.34) in Eqs. (A.31), (A.32) and (A.33) to get the force components as a sum, bilinear in the induced multipole moments. Using the property $q_{l,-m}^* = (-1)^m q_{lm}$ that arises from definition (6) and the properties of spherical harmonics, one then gets

$$\langle F_{ix} \rangle = \operatorname{Re} \sum_{l} C_{li} \operatorname{Re} T_{li} , \qquad (A.36)$$

$$\langle F_{iy} \rangle = \operatorname{Re} \sum C_{li} \operatorname{Im} T_{li} ,$$
 (A.37)

$$\langle F_{iz} \rangle = \operatorname{Re} \sum_{l} C_{li} \sum_{m} \sqrt{(l-m)(l+m)} q_{lmi} q_{l-1,m,i}^{*},$$
 (A.38)

where

$$C_{li} = \frac{2\pi}{\sqrt{(2l+1)(2l-1)}\alpha_{li}}$$
(A.39)

is in general a complex quantity involving the polarizability α_{li} , and

$$T_{li} = \sum_{m} \sqrt{(l-m)(l-m-1)} q_{lmi} q_{l-1,m+1,i}^* \,. \tag{A.40}$$

The force components are thus given in compact form, convenient for numerical computation.

Appendix B. Time-Averaged Torque

In this Appendix we derive general expressions for the time-averaged components of the torque acting upon particle i in a set of N polarizable spherical nanoparticles of radii a in the presence of a uniform AC electric field. The time averaged torque over particle in the ensemble is given by

$$\langle \boldsymbol{\tau}_i \rangle = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) \mathbf{r} \times \mathbf{E}(\mathbf{r}) d^3 \mathbf{r} \,.$$
 (B.1)

The corresponding Cartesian components are

$$\langle \tau_{ix} \rangle = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) (y E_z - z E_y) d^3 \mathbf{r} ,$$
 (B.2)

$$\langle \tau_{iy} \rangle = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) (zE_x - xE_z) d^3 \mathbf{r} ,$$
 (B.3)

$$\langle \tau_{iz} \rangle = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) (x E_y - y E_x) d^3 \mathbf{r} \,.$$
 (B.4)

Using complex field and distance variables defined as $E_{\pm} = E_x \pm iE_y$ and $r_{\pm} = x \pm iy$, the x and y components of the torque can be expressed as

$$\langle \tau_{ix} \rangle = \frac{1}{4} \operatorname{Re} \int \rho_i^*(\mathbf{r}) i (W_+ - W_-) d^3 \mathbf{r} \,, \tag{B.5}$$

$$\langle \tau_{iy} \rangle = \frac{1}{4} \operatorname{Re} \int \rho_i^*(\mathbf{r}) (W_+ + W_-) d^3 \mathbf{r} , \qquad (B.6)$$

where $W_+ = zE_+ - r_+E_z$ and $W_- = zE_- - r_-E_z$. From Eqs. (A.26) and (A.30) for E_+ and E_z , respectively, and introducing relation (A.12) we have

$$zE_{+} = -r\xi \sum_{lm} D_{lmi} r^{l-1} P_{l-1}^{m+1}(\xi) e^{i(m+1)\phi} , \qquad (B.7)$$

$$r_{+}E_{z} = -r\sqrt{1-\xi^{2}}e^{i\phi}\sum_{lm}D_{lmi}r^{l-1}P_{l-1}^{m}(\xi)e^{i(m)\phi}.$$
 (B.8)

Equations (B.7), (B.8) and the identity $-\xi P_{l-1}^{m+1}(\xi) + (l+m)\sqrt{1-\xi^2}P_{l-1}^m(\xi) = -P_l^{m+1}(\xi)$ lead to

$$W_{+} = \sum_{lm} D_{lmi} r^{l} \Big[-\xi P_{l-1}^{m+1}(\xi) + (l+m)\sqrt{1-\xi^{2}} P_{l-1}^{m}(\xi) \Big] e^{i(m+1)\phi}$$

$$= \sum_{lm} V_{lmi} \sqrt{\frac{(2l+1)}{4\pi} \frac{(l-m)!}{(l+m)!}} r^l [-P_l^{m+1}(\xi)] e^{i(m+1)\phi}$$
$$= -\sum_{lm} V_{lmi} r^l \sqrt{(l-m)(l+m+1)} Y_{l,m+1}(\theta,\phi) .$$
(B.9)

Using Eqs. (A.29) and (A.30) for E_{-} and E_{z} , respectively, and introducing relation (A.12) we have

$$zE_{-} = r\xi \sum_{lm} D_{lmi} r^{l-1} (l+m)(l+m-1) P_{l-1}^{m-1}(\xi) e^{i(m-1)\phi}, \qquad (B.10)$$

$$r_{-}E_{z} = -r\sqrt{1-\xi^{2}}e^{-i\phi}\sum_{lm}D_{lmi}r^{l-1}(l+m)P_{l-1}^{m}(\xi)e^{im\phi}.$$
 (B.11)

Equations (B.10), (B.11) and the identity $\xi(l+m-1)P_{l-1}^{m-1}(\xi) + \sqrt{1-\xi^2}P_{l-1}^{m+1}(\xi) = (l-m+1)P_l^{m-1}(\xi)$ lead to

$$W_{-} = \sum_{lm} D_{lmi} r^{l} (l+m) \Big[\xi (l+m-1) P_{l-1}^{m-1}(\xi) + \sqrt{1-\xi^{2}} P_{l-1}^{m}(\xi) \Big] e^{i(m-1)\phi} ,$$

$$= \sum_{lm} V_{lmi} \sqrt{\frac{(2l+1)}{4\pi} \frac{(l-m)!}{(l+m)!}} r^{l} (l+m) (l-m+1) P_{l}^{m-1}(\xi) e^{i(m-1)\phi} ,$$

$$= \sum_{lm} V_{lmi} r^{l} \sqrt{(l+m)(l-m+1)} Y_{l,m-1}(\theta,\phi) .$$
(B.12)

Using Eqs. (B.9), (B.12) and the definition $q_{lmi} = \int \rho_i(\mathbf{r}) r^l Y_{lm}^*(\theta, \phi) d^3 \mathbf{r}$ we get

$$\langle \tau_{ix} \rangle = \frac{1}{4} \operatorname{Re} \int \rho_i^*(\mathbf{r}) i(W_+ - W_-) d^3 \mathbf{r} ,$$

$$= -\frac{1}{4} \operatorname{Re} \sum_{lm} i V_{lmi} \left[\sqrt{(l-m)(l+m+1)} q_{l,m+1,i}^* + \sqrt{(l+m)(l-m+1)} q_{l,m-1,i}^* \right] .$$
 (B.13)

From the relation $q_{lmi} = -(2l+1)/(4\pi)\alpha_{lmi}V_{lmi}$ between the multipole moment lm induced in particle *i* and the corresponding expansion coefficient V_{lmi} we obtain

$$\langle \tau_{ix} \rangle = \operatorname{Re} \sum_{l} \frac{i\pi}{(2l+1)\alpha_{li}} [S_{li} + S_{li}^*], \qquad (B.14)$$

where

$$S_{li} = \sum_{m=-l}^{l-1} \sqrt{(l-m)(l+m+1)} \, q_{lmi} q_{l,m+1,i}^* \,. \tag{B.15}$$

A similar development for the y-component of the torque gives

$$\langle \tau_{iy} \rangle = \operatorname{Re} \sum_{l} \frac{\pi}{(2l+1)\alpha_{li}} [S_{li} - S_{li}^*] \,. \tag{B.16}$$

The z-component of the torque may be rewritten as

$$\langle \tau_{iz} \rangle = \frac{1}{4i} \operatorname{Re} \int \rho_i^*(\mathbf{r}) [r_- E_+ - r_+ E_-] d^3 \mathbf{r} \,. \tag{B.17}$$

Using definitions of r_+ and r_- and relations (A.27) and (A.29) for E_+ and E_- we get

$$r_{-}E_{+} - r_{+}E_{-} = -\sum_{lm} D_{lmi}r^{l} \Big[\sqrt{1-\xi^{2}}P_{l-1}^{m+1}(\xi) + (l+m)(l+m-1)\sqrt{1-\xi^{2}}P_{l-1}^{m-1}(\xi)\Big]e^{im\phi}.$$
 (B.18)

Using $(l + m - 1)\sqrt{1 - \xi^2} P_{l-1}^{m-1}(\xi) = \xi P_{l-1}^m(\xi) - P_l^m(\xi)$, the expression between square brackets in Eq. (B.18), which we denote by C becomes

$$C = \sqrt{1 - \xi^2} P_{l-1}^{m+1}(\xi) + (l+m)[\xi P_{l-1}^m(\xi) - P_l^m(\xi)],$$

= $(l+m)\xi P_{l-1}^m(\xi) + \sqrt{1 - \xi^2} P_{l-1}^{m+1}(\xi) - (l+m)P_l^m(\xi).$ (B.19)

Since $(l+m)\xi P_{l-1}^m(\xi) + \sqrt{1-\xi^2} P_{l-1}^{m+1}(\xi) = (l-m)P_l^m(\xi)$ we get $C = (l-m)P_l^m(\xi) - (l+m)P_l^m(\xi)$, $= -2mP_l^m(\xi)$. (B.20)

Using Eqs. (B.17), (B.18) and (B.20) we find

$$\langle \tau_{iz} \rangle = \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) \frac{1}{2i} (r_- E_+ - r_+ E_-) d^3 \mathbf{r}$$
$$= \frac{1}{2} \operatorname{Re} \int \rho_i^*(\mathbf{r}) \frac{1}{2i} \sum_{lm} V_{lmi} r^l 2m Y_{lm}(\theta, \phi) d^3 \mathbf{r} \,. \tag{B.21}$$

With the definition $q_{lmi} = \int \rho_i(\mathbf{r}) r^l Y_{lm}^*(\theta, \phi) d^3 \mathbf{r}$ and the relation $q_{lmi} = -(2l+1)/(4\pi)\alpha_{lmi}V_{lmi}$ for eliminating V_{lmi} we obtain our final result for the z-component of the torque

$$\langle \tau_{iz} \rangle = \frac{1}{2} \operatorname{Re} \frac{1}{i} \sum_{lm} V_{lmi} m q_{lmi}^*$$
$$= \operatorname{Re} \sum_{lm} \frac{2\pi i}{2l+1} \frac{m}{\alpha_{li}} q_{lmi} q_{lmi}^*.$$
(B.22)

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