Time-averaged total force on a dipolar sphere in an electromagnetic field

P. C. Chaumet and M. Nieto-Vesperinas

Instituto de Ciencia de Materiales de Madrid, Consejo Superior de Investigaciones Cientificas, Campus de Cantoblanco, Madrid 28049, Spain

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We establish the time-averaged total force on a subwavelength-sized particle in a time-harmonic-varying field. Our analysis is not restricted to the spatial dependence of the incident field. We discuss the addition of the radiative reaction term to the polarizability to deal correctly with the scattering force. As an illustration, we assess the degree of accuracy of several previously established polarizability models. © 2000 Optical Society of America

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In the past few years there has been an increase of interest in the manipulation of small particles by means of the Lorentz force. For the subwavelength radius of a sphere the total force that is due to a light wave is usually split into two parts from the use of the dipole approximation (cf. Ref. 1): a gradient force $(\mathbf{p}\cdot\nabla)\mathbf{E}$, which is essentially due to interaction of the particle-induced dipole moment $\mathbf{p}$ with the electric field $\mathbf{E}$ and scattering and absorbing forces $^1/_c\, \dot{\mathbf{p}} \times \mathbf{B}$, where $\mathbf{B}$ is the magnetic vector, $\dot{\mathbf{p}} = \partial \mathbf{p}/\partial t$, and c is the speed of light in vacuum. It has been customary, after Ref. 1, to express the gradient force $\mathbf{F}_{\mathrm{grad}}$ as (see, e.g., Ref. 2)

$$\mathbf{F}_{\mathrm{grad}} = (1/2)\alpha_0 \nabla \mathbf{E}^2, \tag{1}$$

where α_0 is the particle polarizability that satisfies the Clausius–Mossotti equation

$$\alpha_0 = a^3 \frac{\epsilon - 1}{\epsilon + 2}, \tag{2}$$

where a is the particle radius and ϵ denotes the dielectric permittivity. On the other hand, the absorbing and scattering forces are written in the approximation of small spheres through the absorbing (C_{abs}) and scattering (C_{scat}) cross sections as

$$\mathbf{F} = \frac{|E|^2}{(8\pi)} (C_{\mathrm{abs}} + C_{\mathrm{scat}}) \frac{\mathbf{k}}{k}, \tag{3}$$

where $\mathbf{k}$ represents the light vector ($k = |\mathbf{k}|$). When one is using the expression of these cross sections in the dipole approximation, only the first term of their Taylor expansion versus the size parameter, $x = 2\pi a/\lambda$, is usually considered.[3]

At the optical frequencies involved in many experiments, however, only the time average of the electromagnetic force is observed. In this Letter we establish the form of the time-averaged total force on a particle without restriction on the spatial dependence of the electromagnetic field. Further, we discuss some of the consequences of this new relation. For time-harmonic electromagnetic waves,[4] we write $\mathbf{E}(\mathbf{r}, t) = \mathrm{Re}[\mathbf{E}_0 \exp(-i\omega t)]$, $\mathbf{B}(\mathbf{r}, t) = \mathrm{Re}[\mathbf{B}_0 \exp(-i\omega t)]$, and $\mathbf{p}(\mathbf{r}, t) = \mathrm{Re}[\mathbf{p}_0 \exp(-i\omega t)]$; $\mathbf{E}_0$, $\mathbf{B}_0$, and $\mathbf{p}_0$ are complex functions of position in space, and Re denotes the real part. Then the time average of the total force is

$$\langle \mathbf{F} \rangle = \frac{1}{4T} \int_{-T/2}^{T/2} \bigg[(\mathbf{p} + \mathbf{p}^*)\cdot\nabla(\mathbf{E} + \mathbf{E}^*) + \frac{1}{c} (\dot{\mathbf{p}} + \dot{\mathbf{p}}^*) \times (\mathbf{B} + \mathbf{B}^*) \bigg] \mathrm{d}t, \tag{4}$$

where * denotes the complex conjugate. On performing the integral and using $\mathbf{E}_0$, $\mathbf{B}_0$, and $\mathbf{p}_0$, we find that Eq. (4) yields, for each ith Cartesian component of the averaged total force,

$$\langle F^i \rangle = (1/2)\mathrm{Re}\bigg[p_{0j}\partial^j (E_0^i)^* + \frac{1}{c}\, \epsilon^{ijk} \dot{p}_{0j} (B_{0k})^* \bigg] \tag{5}$$

for $i = 1, 2, 3$, where ϵ_{ijk} is the Levi–Civita tensor. Using the relations $\mathbf{B}_0 = {}^c/_{i\omega}\, \nabla \times \mathbf{E}_0$, $\mathbf{p}_0 = \alpha \mathbf{E}_0$, and $\dot{\mathbf{p}}_0 = -i\omega \mathbf{p}_0$, one gets for Eq. (5)

$$\langle F^i \rangle = (1/2)\mathrm{Re}\{\alpha[E_{0j}\partial^j (E_0^i)^* + \epsilon^{ijk}\epsilon_{klm} E_{0j}\partial^l (E_0^m)^*]\}. \tag{6}$$

On taking into account that $\epsilon^{ijk}\epsilon_{klm} = \delta_l^i \delta_m^j - \delta_m^i \delta_l^j$ one can finally express $\langle F^i \rangle$ as

$$\langle F^i \rangle = (1/2)\mathrm{Re}[\alpha E_{0j}\partial^i (E_0^j)^*]. \tag{7}$$

Equation (7) is the main result of this Letter. It represents the total averaged force exerted by an arbitrary time-harmonic electromagnetic field on a small particle.

In this connection, Ref. 5 establishes the average force on an object represented by a set of dipoles when the electromagnetic field is a plane wave. We note that in this case Eq. (7) reduces to just Eq. (3), in agreement with the results reported in Ref. 5. However, as we illustrate next, Eq. (7) allows one to apply the coupled-dipole method (CDM) to more-complex configurations such as that of a small particle in front of a dielectric surface, under arbitrary illumination (see Ref. 6 for a discussion of the coupled-dipole method for large particles). Also, the

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absence of the magnetic field $\mathbf{B}_0$ in Eq. (7) eases the computations.

Conversely, when Eq. (2) for the polarizability is introduced into Eq. (7), one obtains for the ith component of the time-averaged optical force

$$\begin{aligned}\langle F^i\rangle &= (1/2)\alpha_0 \operatorname{Re}[E_{0j}\partial^i(E_0^j)^*] \\ &= (1/4)\alpha_0 \operatorname{Re}(\partial^i|\mathbf{E}_0|^2) = (1/4)\alpha_0(\partial^i|\mathbf{E}_0|^2),\end{aligned} \quad (8)$$

which is just the gradient force. Notice the factor (1/4) (see, e.g., Ref. 7) instead of (1/2), which often appears for nonaveraged fields in the literature (see, for example, Refs. 2, 8, and 9). In agreement with the remarks in Ref. 10, the scattering force, Eq. (3), vanishes, and thus $\langle\mathbf{F}\rangle$ reduces to the gradient force. Therefore, the static expression of α_0 from Eq. (2) must be replaced with an added damping term. This was done by Draine,[10] who, with the help of the optical theorem, obtained

$$\alpha = \alpha_0/[1 - (2/3)ik^3\alpha_0]. \quad (9)$$

The existence of the imaginary term for α in Eq. (9) is essential for deriving the correct value for the averaged total force that is due to a time-varying field.

As an illustration, let the field that illuminates the particle be the beam whose electric vector is

$$E_x = \exp(-x^2/2)\exp[i(kz - \omega t)], \qquad E_y = 0, E_z = 0. \quad (10)$$

Using Eqs. (2) and (10) in Eq. (7), we find

$$\langle F_x\rangle = -(\alpha_0/2)x \exp(-x^2), \quad (11a)$$

$$\langle F_z\rangle = 0. \quad (11b)$$

On the other hand, if the correct polarizability, Eq. (9), is introduced with Eqs. (10) into Eq. (7), the total force is then expressed as

$$\begin{aligned}\langle F_x\rangle &= (1/2)\operatorname{Re}[-\alpha x \exp(-x^2)] \\ &= \frac{-(\alpha_0/2)x \exp(-x^2)}{1 + (4/9)k^6\alpha_0^2},\end{aligned} \quad (12a)$$

$$\begin{aligned}\langle F_z\rangle &= (1/2)k \exp(-x^2)\operatorname{Re}(-i\alpha) \\ &= \frac{\exp(-x^2)k^4\alpha_0^2/3}{1 + (4/9)k^6\alpha_0^2}.\end{aligned} \quad (12b)$$

For a particle with a radius $a \ll \lambda$, e.g., $a = 10$ nm, at wavelength $\lambda = 632.8$ nm and $\epsilon = 2.25$, the factor $[1 + (4/9)k^6\alpha_0^2]$ is very close to 1 (notice in passing that the expression used for α in Ref. 11 makes this factor unity). Thus we can see that, in contrast with Eqs. (11), the correct form for the polarizability, Eq. (9), leads to a total force given by Eqs. (12a) and (12b), which can be associated with the gradient and scattering components, namely, with the time average of Eq. (1) and Eq. (3) with $C_{\text{abs}} = 0$, respectively.

In the case of an absorbing sphere, the dielectric constant becomes complex, and so is α_0. Then, Eqs. (12) with $a \ll \lambda$ become

$$\langle F_x\rangle = -(1/2)\operatorname{Re}(\alpha_0)x \exp(-x^2), \quad (13a)$$

$$\langle F_z\rangle = \frac{\exp(-x^2)k^4|\alpha_0|^2}{3} + \frac{k \exp(-x^2)}{2}\operatorname{Im}(\alpha_0). \quad (13b)$$

The imaginary part of α_0 does not contribute to the component $\langle F_x\rangle$, that is, to the gradient force, Eq. (13a). On the other hand, the absorbing and scattering force, Eq. (13b), exactly coincides with the expression obtained from Eq. (3).

We next illustrate the above arguments with numerical calculations that permit us to assess the degree of accuracy of several previously established polarizability models. We first compare the relative difference between the force obtained from the exact Mie calculation and the most-typical polarizability models, namely, those of Lakhtakia[12] (LAK) and Dungey and Bohren[13] (DB) and the Clausius–Mossotti relation with the radiative reaction term[10] (CM-RR), versus the radius a of a sphere illuminated by a propagating plane wave in free space (Fig. 1). Next, when this sphere is illuminated by an evanescent wave created by total internal reflection on a dielectric surface, the component of the force perpendicular to the incident wave vector (Fig. 2) is compared with the result derived from the CDM.[6] All curves are represented up to $a = \lambda/10$. The percent relative difference in Fig. 2 is defined as $100 \times (F_{\text{ref}} - F_{\text{pol}})/F_{\text{ref}}$, where pol denotes the force obtained from the corresponding method used for the polarizability (LAK, DB, or CM-RR) and ref stands for

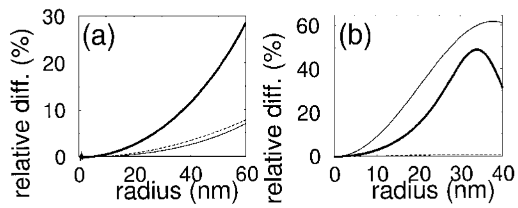

Fig. 1. (a) Relative difference between the force computed by the exact Mie calculation and by the dipole approximation: thin curve, CM-RR; thick curve, LAK; dashed curve, DB. The sphere is glass ($\epsilon = 2.25$) illuminated by an incident propagating plane wave ($\lambda = 600$ nm). (b) Same as (a) but for a silver sphere ($\lambda = 400$ nm, $\epsilon = -4 + i0.7$).

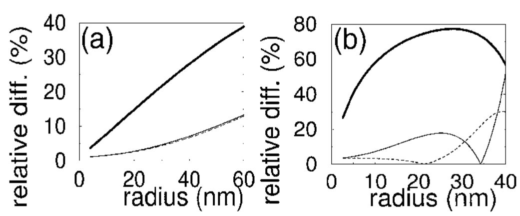

Fig. 2. (a) Relative difference between the component of the force perpendicular to the incident wave vector obtained by the CDM and by the dipole approximation: thin curve, CM-RR; thick curve, LAK; dashed curve, DB. The sphere is glass ($\epsilon = 2.25$) illuminated by an incident evanescent wave ($\lambda = 600$ nm). (b) Same as (a) but for a silver sphere ($\lambda = 400$ nm, $\epsilon = -4 + i0.7$).

the force derived from the Mie calculation when the incident wave is propagating and from the CDM when the incident wave is evanescent.

We first consider a dielectric sphere (glass, $\epsilon = 2.25$) illuminated at $\lambda = 600\,\mathrm{nm}$ [Figs. 1(a) and 2(a)]. We observe that, for an incident propagating wave [Fig. 1(a)], the result from the CM-RR relation is better than that of DB, and this, in turn, is better than the result from LAK. The force over a dielectric particle given by the exact Mie calculation is $F = C_{\mathrm{scat}}(1 - \overline{\cos\theta})|E|^2/(8\pi)$, and that obtained from the dipole approximation is $F = (1/2)|E|^2\,\mathrm{Re}(-i\alpha)$. When the DB model is used, $\alpha = (3/2)ia_1/k^3$, where a_1 is the first Mie coefficient, and hence, $4\pi\,\mathrm{Re}(-i\alpha)$ is the scattering cross section for an electric dipole. However, when Eq. (9) for the CM-RR is employed, $4\pi\,\mathrm{Re}(-i\alpha)$ constitutes only the first term of the Taylor expansion of the scattering cross section versus the size parameter x. This is why C_{scat} is underestimated when it is calculated from the CM-RR model. Therefore the DB model should be better. However, in both cases the factor $\overline{\cos\theta}$ has not been taken into account in the dipole approximation, and thus both results overestimate the force. Hence, this factor $\overline{\cos\theta}$ produces a balance, making the CM-RR result closer to the Mie solution. In the case of an incident evanescent wave [Fig. 2(a)], the DB and CM-RR results are very close together; this is due to the fact that the real parts of both polarizabilities are very close to each other. One can see that the LAK result, as with a propagating wave, is far from the correct solution.

As a second example, we consider a metallic sphere (silver) illuminated at $\lambda = 400\,\mathrm{nm}$ ($\epsilon = -4 + i0.7$). We now observe that for an incident propagating wave [Fig. 1(b)] the DB model yields the best result. The force can be exactly written as $F = (C_{\mathrm{ext}} + C_{\mathrm{scat}}\overline{\cos\theta})|E|^2/(8\pi)$. Notice that now $C_{\mathrm{scat}}\overline{\cos\theta}$ is of the sixth order in x in comparison with C_{ext}. Since $C_{\mathrm{ext}} \propto \mathrm{Re}(a_1)$ in the electric dipole limit, the DB formulation appears to be the best. Also, for incident evanescent waves [Fig. 2(b)], the DB formulation gives the most accurate solution. However, for a metallic sphere, the relative permittivity greatly depends on the wavelength used. Hence, it is difficult to establish a generalization of these results. We checked and found that, for a gold or silver sphere in free space in the visible, the DB formulation is often the best.

In summary, we have established the average total force on a small particle in a time-harmonic-varying field of arbitrary form and thus clarified the use of this finding in the interpretation of experiments as well as of previous theoretical works. For instance, we showed that Eq. (7) is not just the gradient force as stated previously (see, e.g., Ref. 14). Also, this general expression shows the importance of the radiative reaction term in the polarizability of the sphere as put forward by other authors. In the derivation of Eq. (7) we make no assumptions about the surrounding environment. It is necessary only to know both the electric field and its derivative at the position of the sphere, and thus Eq. (7) permits easy handling of illuminating evanescent fields. An immediate important consequence is that it allows one to assess the adequacy of several polarizability models.

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