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Optical forces in time domain on arbitrary objects

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We develop a general theoretical and computational framework to describe, in time domain, the exchange of momentum between light and arbitrary three-dimensional objects. Our formulation can be used to study the time evolution of optical forces on any object with linear material response, including inhomogeneous, dispersive, and absorbing dielectrics and metals. We illustrate our approach by studying the behavior of the Abraham force on an object illuminated by a sequence of electromagnetic pulses.

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The mechanical coupling between light and matter is at the heart of many fundamental and applied research areas. Since the original formulation of the concept of optical tweezers by Ashkin [1], optical forces have led to numerous applications in a variety of disciplines, including atomic physics [2], biophysics [3], and nanophotonics [4–6]. In most cases matter-light interactions are studied in the time-harmonic regime where nonstationary contributions to the optical force vanish. However, when an object is illuminated by an arbitrary time-dependent electromagnetic wave, a full description of the exchange of momentum between light and the object requires a rigorous formulation of the electromagnetic forces that accounts for the nonstationary terms in the force. Recently, Hinds and Barnett presented a rigorous treatment of the interaction between an electromagnetic pulse and a two-level atom [7]. These authors showed that a nonstationary term involving the cross product of the electric dipole moment of the atom with the magnetic field, a term seldom considered in the treatment of optical forces, is not only important but also essential in understanding the fundamental difference between the Minkowsky and the Abraham formulations of optical momentum [8]. Hence, the work by Hinds and Barnett illustrates the necessity to use a rigorous treatment of optical forces when dealing with time-dependent electromagnetic fields. The question of the transfer of momentum between electromagnetic waves and matter has been studied analytically for special geometries [9] and numerically using the finite difference in time domain (FDTD) method for two-dimensional structures [10]. However, to the best of our knowledge, no general time-domain treatment of the optomechanical interaction between light and a three-dimensional object has been formulated that can handle arbitrary shapes, as well as dispersive and/or absorbing dielectrics and metals.

The aim of this Rapid Communication is first to provide a general treatment of optical forces on three-dimensional objects under arbitrary, time-dependent illumination. Our approach can handle a wide variety of scatterers, from a single atom in the two-level approximation to a large scatterer, several wavelengths in size. Second, we aim to illustrate how the Abraham force manifest itself for time-varying fields. This force which can be ignored in the time-harmonic case plays a subtle yet important role in the exchange of momemtum between the electromagnetic fields and the scatterer. We highlight the effect of the Abraham force by considering the interaction of a scatterer with a short pulse. Hence we hope that the general method presented in this article for the description of momentum exchange between light and three-dimensional structures will stimulate new experimental investigations, in particular regarding the Abraham force [11].

We start by describing the electromagnetic properties of our scattering object using the coupled dipole method (CDM, also called the discrete dipole approximation). This method was proposed by Purcell and Pennypacker in the 1970s [12,13] to study the scattering of light in time harmonic regime by interstellar grains with arbitrary shapes. Since then, the method has been used to study, in the frequency domain, a variety of electrodynamic problems, including optical forces and torques [14], near-field optical nanomanipulation [15], and optical trapping near a photonic crystal cavity [16]. In the CDM an arbitrary object is discretized as a collection of N dipolar subunits. One significant advantage of the CDM versus other computational electromagnetic techniques is that it possesses the versatility of a discrete formulation of Maxwell's equations (hence any geometry can be considered), while preserving a semianalytic description of the interaction between the subunits forming the object. Furthermore, space is discretized only over the scatterers thus allowing an analytical derivation of the electromagnetic fields outside the object, once the internal fields are known.

Traditionally the CDM operates in the frequency domain (fields are assumed to be time-harmonic); however, recently we developed a time-domain formulation of the CDM to study light scattering by arbitrary objects [17]. Using our recent work as a starting point, we now develop a new time-domain method to study optical forces based on the CDM. The i-th component of the force on a small particle with electric dipole moment $\boldsymbol{\mathcal{P}}$, due to an electromagnetic field $\{\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{B}}\}$, is [18]:

$$\mathcal{F}_i(\mathbf{r},t)=\mathcal{P}_j(\mathbf{r},t)[\partial_i\mathcal{E}_j(\mathbf{r},t)]+\frac{\varepsilon_{ijk}}{c}\partial_t[\mathcal{P}_j(\mathbf{r},t)\mathcal{B}_k(\mathbf{r},t)], \quad (1)$$

where ε_{ijk} is the Levi-Civita tensor and i, j, and k stand for either x, y, or z. In the time harmonic regime the second term of Eq. (1) vanishes; however, in the general case, its contribution to the force cannot be neglected [7].

Let us now consider the case of an arbitrary object illuminated by a time-dependent electromagnetic wave with envelop $\mathcal{H}(t)$, and a spectrum centered at frequency f_0. Let $H(\omega)$ be the Fourier transform of $\mathcal{H}(t)$. The linearity of Maxwell's equations ensures that solving the time-harmonic scattering problem with incident field $H(\omega)\mathbf{E}_0(\mathbf{r},\omega)$, and computing the inverse Fourier transform of the resulting time harmonic scattered fields yields the correct time evolution for the scattered fields [17]. The self-consistent electric local field at subunit I is given by:

$$\mathbf{E}(\mathbf{r}_I,\omega) = \mathbf{E}_0(\mathbf{r}_I,\omega) + \sum_{J=1,J\neq I}^{N} \overleftrightarrow{T}(\mathbf{r}_I,\mathbf{r}_J,\omega)\,\overleftrightarrow{\alpha}(\mathbf{r}_J,\omega)\mathbf{E}(\mathbf{r}_J,\omega), \quad (2)$$

where $\overleftrightarrow{\alpha}(\mathbf{r}_J,\omega)$ is the polarizability of the dipolar subunits, $\overleftrightarrow{T}(\mathbf{r}_I,\mathbf{r}_J,\omega)$ is the field susceptibility tensor, and $\mathbf{p}(\mathbf{r}_I,\omega)=\overleftrightarrow{\alpha}(\mathbf{r}_J,\omega)\mathbf{E}(\mathbf{r}_J,\omega)$ the dipole moment associated to the subunit located at $\mathbf{r}_I$. In practice Eq. (2) is solved for a finite set of frequencies in accordance with the Nyquist-Shannon sampling theorem. See Ref. [17] for an efficient way to solve for the fields across many frequencies. The spatial derivative of the local electric field can also be obtained as [19]:

$$\boldsymbol{\partial}\mathbf{E}(\mathbf{r}_I,\omega) = \boldsymbol{\partial}\mathbf{E}_0(\mathbf{r}_I,\omega) + \sum_{J=1,J\neq I}^{N} \boldsymbol{\partial}\,\overleftrightarrow{T}(\mathbf{r}_I,\mathbf{r}_J,\omega)\,\overleftrightarrow{\alpha}(\mathbf{r}_J,\omega)\mathbf{E}(\mathbf{r}_J,\omega). \quad (3)$$

Notice that, unlike the two-level atom case considered in Refs. [7,20], here we account for retardation effects across the object and morphological (i.e., geometric) resonances. The first term in the expression of the optical force [Eq. (1)] on a dipole located at $\mathbf{r}_I$ can therefore be expressed as:

$$\mathcal{F}_i^h(\mathbf{r}_I,t) = \mathcal{G}^{-1}[H(\omega)p_j(\mathbf{r}_I,\omega)]\mathcal{G}^{-1}[H(\omega)\partial_i E_j(\mathbf{r}_I,\omega)], \quad (4)$$

where $\mathcal{G}$ denotes the temporal Fourier transform. The derivation of the contribution to the force from the second term in Eq. (1) is slightly more involved. First we note that we can use Maxwell's equations to express the electric field in terms of the magnetic field. We can also write the cross product of the dipole moment and the magnetic field, in time domain, as:

$$\mathcal{A}_i(\mathbf{r}_I,t) = \frac{\varepsilon_{ijk}}{c}\mathcal{G}^{-1}[H(\omega)p_j(\mathbf{r}_I,\omega)]\mathcal{G}^{-1}[H(\omega)B_k(\mathbf{r}_I,\omega)]. \quad (5)$$

Taking the temporal Fourier transform of $\mathcal{A}_i(\mathbf{r}_I,t)$ yields $\mathbf{A}(\mathbf{r}_I,\omega)$. The second term of the optical force in time domain can now be expressed as:

$$\mathcal{F}_i^p(\mathbf{r}_I,t) = \partial_t\mathcal{A}_i(\mathbf{r}_I,t) = \mathcal{G}^{-1}[-i\omega A_i(\mathbf{r}_I,\omega)]. \quad (6)$$

Finally, the time evolution of the total force on the arbitrary object is obtained by adding the partial forces acting on the subunits forming the object:

$$\mathcal{F}_i(t) = \mathcal{F}_i^h(t) + \mathcal{F}_i^p(t) = \sum_{I=1}^{N}\mathcal{F}_i^h(\mathbf{r}_I,t) + \sum_{I=1}^{N}\mathcal{F}_i^p(\mathbf{r}_I,t). \quad (7)$$

We have separated the force into two parts. The first part $\mathcal{F}_i^h(t)$ is the standard term that appears in the time-harmonic treatment of optical forces, while the second part $\mathcal{F}_i^p(t)$ is a term that vanishes in a time-harmonic picture and which has seldom been considered in the treatment of optical forces on complex objects. We can also define the momentum imparted to the object by the electromagnetic field as:

$$\mathcal{Q}_i(t) = \int_0^t \mathcal{F}_i(t)\mathrm{d}t = \mathcal{Q}_i^h(t) + \mathcal{Q}_i^p(t), \quad (8)$$

where we have assumed that the origin of time has been chosen such that the electromagnetic fields at the object are zero for $t<0$. We will now consider several examples that will help us illustrate the contribution of each term. Let us start by assuming that the incident field is a plane wave with a Gaussian envelop of the form:

$$\mathcal{H}(t) = \exp\left[-16\left(\frac{t-\tau}{\tau}\right)^2\right]\sin(2\pi f_0 t), \quad (9)$$

where $f_0=\omega_0/2\pi=c/\lambda_0$ is the central frequency of the pulse and $\tau=8/f_0$ is the duration of the pulse. The spectral and time profiles of the incident pulse are plotted in Fig. 1(a) and Fig. 1(b), respectively. Note that on the plots, the frequency is normalized to f_0 and the time is normalized to τ. The scattering object is a homogeneous sphere, initially at rest, with radius a and permittivity $\varepsilon=2.25$. We compute the time evolution of the force and the particle momentum for a dipolar

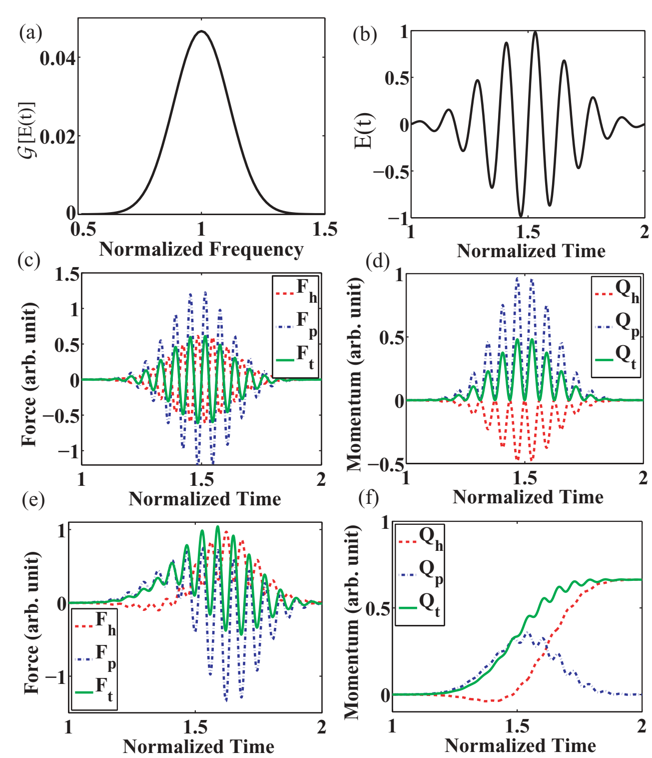

FIG. 1. (Color online) Spectral (a) and time (b) profiles of the incident field. (c) Force experienced by a dipolar sphere versus time. (d) Momentum transfer from the pulse to the dipolar sphere versus time. (e) and (f) same as (c) and (d) but with radius $a=\lambda_0/2$.

sphere [$ka \ll 1$, Figs. 1(c) and 1(d)] and for a sphere with radius $a = \lambda_0/2$ [Figs. 1(e) and 1(f)].

It can be easily shown that for the dipolar sphere the oscillations of $\mathcal{F}_h$ are due to the gradient force and that the amplitude of these oscillations is half the amplitude of the oscillations of $\mathcal{F}_p$, which is consistent with the result of Hinds and Barnett on two-level atoms [7]. Since the term $\mathcal{F}_p$ corresponds to the time derivative of the Poynting vector, its contribution to the momentum of the object is only nonzero over the duration of the pulse. This is not the case for the other contribution to the optical force, which comprises the so-called radiation pressure. However, for a small particle in the dipole approximation, the strong oscillations of the gradient force result in a zero net momentum transfer to the particle (but a finite displacement). After the pulse has passed over the particle it will have imparted to the particle a small momentum increase corresponding to the radiation pressure. Figures 1(e) and 1(f) represent the time evolution of the force and the momentum for a sphere with a diameter equal to the central wavelength of the pulse. The sphere is discretized with $N = 4224$ subunits. In this case we find that during the interaction with the leading edge of the pulse the momentum imparted to the sphere is predominantly due to $\mathcal{F}_p$, whereas $\mathcal{F}_h$ remains very weak. Then, as the tail of the pulse passes over the object, $\mathcal{Q}_p$, the contribution of $\mathcal{F}_p$ to the momentum of the object tends toward zero. However, during the second half of the pulse $\mathcal{Q}_h$ increases and as a result the total momentum imparted to the object increases over the duration of the pulse, while exhibiting small oscillations. Because our approach is general we can investigate more complex geometries and/or material dispersions. In Fig. 2 we consider a cube in gold with a spectrum of the incident field in the visible range as shown in Fig. 2(a). The permittivity of gold is taken from experimental data [21]. The central frequency f_0 corresponds to the plasmon mode of a dipolar sphere, i.e., $\varepsilon(f_0) \approx -2$. For a cube of side $a = \lambda_0/10$, $\mathcal{F}_h$ oscillates but always remains positive. In fact, the optical force is mainly due to the Fourier component of the pulse at the resonance frequency f_0. At this frequency, the oscillations of the dipole moment induced in the small cube are in quadrature with the oscillations of the incident electric field, hence the induced dipole moment is in phase with the spatial derivative of the field, which results in a positive $\mathcal{F}_h$ term. For the larger cube [side of $a = \lambda_0/2$, discretized with $N = 125,000$ subunits, Figs. 2(e) and 2(f)] we observe a similar behavior with the net-momentum of the particle being mainly due to $\mathcal{F}_h$. Notice that with for an irradiance of 1 mW/μm^2 at the peak of the pulse, the force experienced by the larger (smaller) cube is 0.4 nN (20 f N). If we assume that the cube does not move significantly during its interaction with the pulse, from the momentum we get a velocity of 3.4 ms^{-1} (12.7 ms^{-1}) after the sequence of the pulse. Assuming the particle is in water and a low Reynolds number, this corresponds to an overall spatial displacement of about 450 nm (132 nm).

We now consider an inhomogeneous sphere with radius $a = 0.8\lambda_0$, and permittivity $\varepsilon(r) = 2.25 + 2\sin^2(\pi r^2/a^2)$, where r is the distance from the center of the sphere. To account for the spatial variation of the index we discretize the sphere into $N = 33,552$ subunits. Figures 3(a) and 3(b) show the time evolution of the optical force and momentum with the

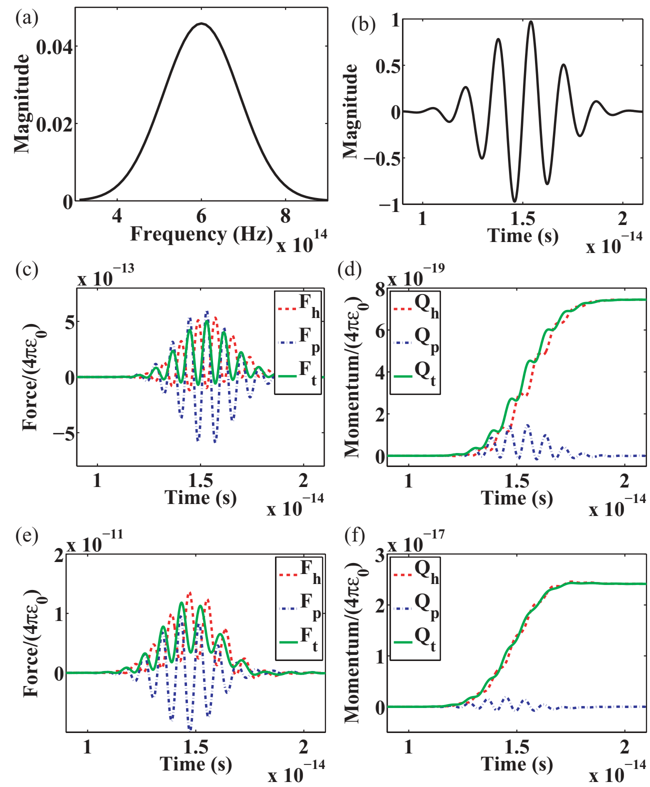

FIG. 2. (Color online) Spectral (a) and time (b) profiles of the incident field. (c) Force experienced by a cube with side $a = \lambda_0/10$ versus time. (d) Momentum transfer from the pulse to the cube versus time. (e) and (f) same as (c) and (d) but with $a = \lambda_0/2$.

pulse defined as in Fig. 1. We observe a strong contribution from $\mathcal{Q}_p$ to the momentum when the pulse reaches the object. At the same time a negative value of $\mathcal{Q}_h$ is observed. This is similar to the case of a homogeneous dielectric sphere [Fig. 1(f)]. However, by contrast with the homogeneous case, we also observe that the force remains nonzero long after the incident pulse has passed over the object. This is a consequence of the excitation, by the incident pulse, of creeping waves at the surface of the sphere. Accordingly, the force, and to a lesser extent the momentum, exhibit weak oscillations at the optical frequency $2f_0$ and stronger oscillations at a frequency corresponding to the reciprocal of the period of the creeping wave (time required for the wave to travel around the sphere).

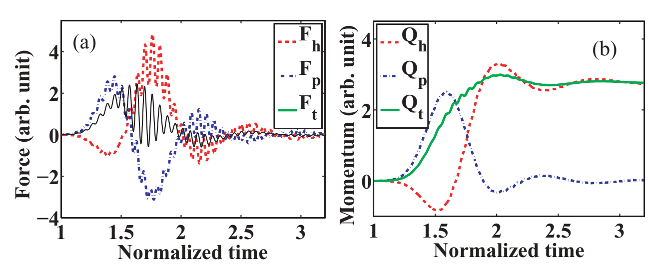

FIG. 3. (Color online) (a) Force experiences by a sphere in the presence of a creeping wave. (b) Momentum imparted by the pulse to the object.

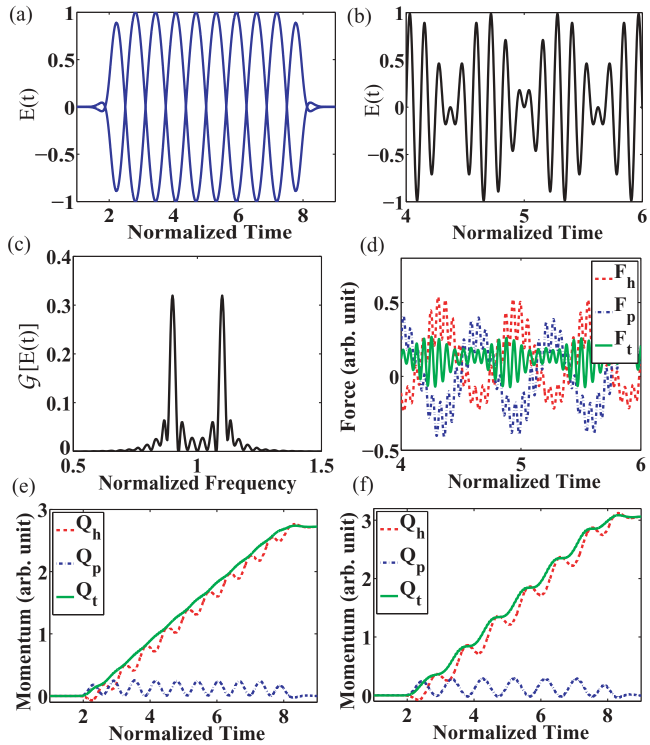

FIG. 4. (Color online) (a) Envelop of the incident pulse. (b) Zoom on the incident pulse. (c) Spectrum of the incident pulse. (d) Optical force experienced by the object. (e) Momentum imparted by the pulse to the object for $w_1 = w_0/10$. (f) Same as in (e) but with $w_1 = w_0/15$.

The strong initial contribution to the momentum of the object from $\mathcal{F}_p$ in the presence of creeping waves may give us an insight on how to observe experimentally the influence of $\mathcal{F}_p$. The idea is to achieve a coherent control of the interaction between the incident wave and the creeping wave. To see that let us construct a new incident field from a sequence of pulses such that the envelop of the incident wave is now:

$$\mathcal{H}(t) = \left[\frac{1}{1+e^{10(-t+2\tau)/\tau}} - \frac{1}{1+e^{10(-t+8\tau)/\tau}}\right] \times \sin(\omega_0 t)\sin(\omega_1 t), \quad (10)$$

where ω_0 is the frequency defined previously. We choose $\omega_1 = \omega_0/10$ such that the time between two consecutive pulses matches the period of the creeping wave. The time profile of the envelop of the incident wave is shown in Fig. 4(a), and a portion of the time profile of the amplitude of the electric field is shown in Fig. 4(b). The spectrum of the incident field in given in Fig. 4(c). The optical force experienced by the sphere is plotted in Fig. 4(d). We observe the usual oscillations at frequency $2f_0$ but we also have strong oscillations at the frequency $f_0/5$, with $\mathcal{F}_p$ and $\mathcal{F}_h$ oscillating 180° out of phase. As a result, the total force exhibits oscillations at frequency $2f_0$ around a positive average value. As shown in Fig. 4(e) this leads to a linear increase of $\mathcal{Q}_t$ with time despite the fact that the incident wave consists of a series of pulses. This surprising result is due to the contribution from the $\mathcal{F}_p$ term of the force as $\mathcal{Q}_h$ alone would exhibit oscillations at the frequency $2f_1 = f_0/5$. Note that if $\omega_1 = \omega_0/15$ then the time between two consecutive pulses is equal to 1.5 times the period of the creeping wave. In this case, we clearly see in Fig. 4(f) that $\mathcal{Q}_t$ oscillates at frequency $2f_1$.

In conclusion we developed a general approach to optical forces in the time domain. We illustrated our approach on several objects, both at and beyond the dipole approximation. In particular, we showed that if a particle supports a surface state, by tailoring the time profile of the incident wave such that it enhances or suppresses the contribution of the surface state to the momentum exchange, the influence of the Abraham term in the force can be highlighted. Therefore it would be interesting to create the type of oscillations of the momentum described in this letter at a slow-enough frequency to be observed experimentally. This might be achievable by exciting high-Q modes such as whispering gallery modes in spherical particles.

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