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Dynamical response of polarizable nanoparticles to a rotating electric field

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We discuss the transfer of angular momentum from light to classical nanoparticles. An optical torque is induced by a circularly polarized beam, causing the object to rotate. The effect depends on absorption and geometry in such a way that an isotropic dissipationless object is not affected by the external field. Under constant illumination an asymmetric object may rotate uniformly if the light intensity exceeds a minimum value, below which the object executes a rocking motion. These findings are applied to a bioparticle with spheroidal symmetry.
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I. INTRODUCTION

In undergraduate electromagnetic and introductory quantum mechanics courses, the interaction of light with matter is associated with the absorption and reflection of radiation, but seldom with changes in the motion of an object as a whole. A brief reference is made, if at all, to the linear and angular momentum carried by light and the acceleration that light can impart.

Such effects are negligible in ordinary macroscopic matter, but, if the size of the object is small, we need to consider the mechanical effects of the interaction to account for the object's mechanical behavior. In particular, efforts to manipulate minute objects by means of laser beams rely heavily on the transfer of linear and angular momentum from light to the objects. Because of the importance of miniaturization, much related research has been done in recent years,[1–7] and several applications are being considered, such as nanomotors and the use of laser radiation to control the motion of cells in suspension.[8]

Because light carries linear momentum, the conservation laws require that when absorption of light occurs, the lost momentum is transferred to the irradiated object. Similarly, if circularly polarized light is absorbed, the angular momentum of such a field must cause an angular acceleration of the object. In this paper we shall concentrate on the latter effect and discuss the rotation of small objects due to the torque caused by light. Because torque acts even when the object is homogeneous and spherical, the origin of the angular momentum transfer is more difficult to understand. In Sec. II we consider a uniform spherical dielectric object in a rotating electric field, and show how simple reasoning based on light as a collection of photons can be used to derive the torque. We also show that absorption is necessary for the torque to be present. In Sec. III we consider an asymmetric nanoparticle immersed in a fluid and derive the torque taking into account the different polarizabilities with respect to the major axes of the object. A torque appears due to the asymmetry, even if the object does not absorb light. As a simple application we apply our results in Sec. IV to a bioparticle exposed to a laser beam whose plane of polarization rotates using a half wave plate, and find that a minimum light intensity is required to induce a synchronous uniform rotation. Below this minimum the object is not able to reach a steady state and develops a rocking motion with twice the rotation frequency of the external field.

II. THE TORQUE ON A SPHERICAL OBJECT IN A CIRCULARLY POLARIZED BEAM

A polarizable isotropic sphere subject to a constant uniform electric field $\vec{E}$ acquires a dipole moment $\vec{p}$ aligned with the local field. The mechanical torque $\vec{\Gamma} = \vec{p} \times \vec{E}$ on the sphere is zero.[9] This statement holds if the field is time dependent, provided the polarization adjusts instantaneously to the new field direction as the latter changes. When internal friction is present, the polarization adjustment is not instantaneous and may lag the changing field and generate a torque. For example, because the free electrons in a metal adjust very quickly to the external field, their interaction with the lattice of comparatively heavy ions involves the inertia of the system as a whole and slows down the adjustment.

This effect is explained by standard classical electromagnetic theory.[10] An unbounded circularly polarized electromagnetic plane wave of angular frequency ω transfers angular momentum to an absorbing isotropic sphere of radius a at the rate,[11]

$$\Gamma_z = I_{\text{inc}} \frac{Q_{\text{abs}}}{\omega} \pi a^2, \tag{1}$$

where z is the direction of propagation of the laser beam, I_{inc} is the intensity of the incident wave, and Q_{abs} is the Mie absorption efficiency. For an idealized uniform lossless dielectric sphere $Q_{\text{abs}} = 0$, and no torque would be transferred by a circularly polarized electromagnetic wave.

A simple derivation of Eq. (1) is possible if we consider an isotropic spherical object centered at the origin in the presence of a uniform rotating electric field of amplitude E_0, as shown in Fig. 1. The electric field may be expressed as

$$\vec{E}(t) = E_0(\hat{x}\cos\omega t + \hat{y}\sin\omega t). \tag{2}$$

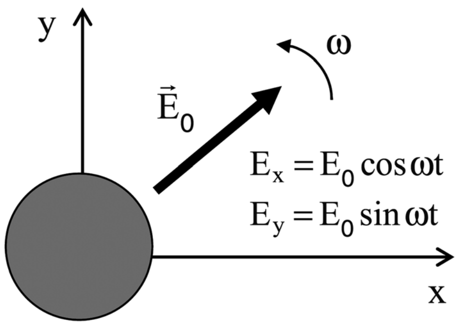

Fig. 1. Isotropic sphere in an electric field rotating counterclockwise with angular velocity ω.

Equation (2) is equivalent to the real part of $\vec{E}(t) = E_0(\hat{x} + i\hat{y})e^{-i\omega t}$. In the same notation an absorbing piece of matter has a complex polarizability $\alpha = |\alpha|e^{i\varphi}$. The instantaneous electric dipole moment is obtained by taking the real part of the complex dipole moment $\vec{p} = \alpha\vec{E}$, giving

$$p_x(t) = |\alpha| E_0 \cos(\omega t - \varphi), \tag{3a}$$

$$p_y(t) = |\alpha| E_0 \sin(\omega t - \varphi), \tag{3b}$$

where ϕ is the phase difference between the external field and the polarization of the object. The torque $\vec{\Gamma} = \vec{p} \times \vec{E}$ about the z-axis is given by

$$\Gamma_z = p_x E_y - p_y E_x = E_0^2 \mathrm{Im}(\alpha). \tag{4}$$

Equation (4) is obtained directly from Eqs. (2) and (3). The time-averaged power absorbed by the object from the external field is given by[12]

$$\bar{P}_{\mathrm{abs}} = \omega E_0^2 \, \mathrm{Im}(\alpha). \tag{5}$$

Equations (5) and (4) give the torque as

$$\Gamma_z = \frac{\bar{P}_{\mathrm{abs}}}{\omega}, \tag{6}$$

which is equivalent to Eq. (1).

We next show that Eq. (1) also follows from a semiclassical analysis in which light is considered to be a collection of photons carrying linear and angular momentum. Each photon in a right-/left-circularly polarized beam of light carries $\hbar$ units of angular momentum pointing along/against the direction of propagation, where $\hbar = h/2\pi$ and h is Plank's constant. Kiang and Young[13] calculated the angular momentum of a photon in a circularly polarized beam by evaluating the energy and angular momentum imparted by a classical plane wave to a point charge. Their result is valid for an absorbing particle and no explicit consideration is given to scattering and conservation of linear and angular momentum.

By using such a model, the generation of a torque due to the absorption of circularly polarized light may be explained as follows. By angular momentum conservation, the absorption of a photon of energy $\hbar\omega$ transfers angular momentum $\hbar$ to the particle. The torque generated is $\hbar$ times the number of photons absorbed per unit of time. To be specific, consider a uniform sphere immersed in a laser field of right-circularly polarized light. The sphere is treated classically and obeys Newton's laws. Let F_z be the rate of photons incident in the z-direction, each with energy $\hbar\omega$. If A is the photon absorption rate and G is the total rate of scattered photons in all directions, conservation of energy requires that

$$F_z \hbar\omega = A\hbar\omega + G\hbar\omega. \tag{7}$$

Because each incident photon carries a linear momentum in the z-direction, conservation of linear momentum requires that

$$F_z \frac{\hbar\omega}{c} = \frac{dP_z}{dt} + G_z \frac{\hbar\omega}{c}, \tag{8}$$

where G_z is the net rate of photons scattered in the z-direction and P_z is the momentum acquired by the sphere in the z-direction. In addition, each incident photon carries an angular momentum $\hbar$ along the direction of propagation, and conservation of angular momentum requires that

$$F_z \hbar = \frac{dL_z}{dt} + G_z \hbar, \tag{9}$$

where L_z is the z component of the angular momentum acquired by the particle. From Eqs. (7)–(9) the torque acting on the particle becomes

$$\Gamma_z = \frac{dL_z}{dt} = \hbar[A + (G - G_z)]. \tag{10}$$

For an isotropic particle of radius comparable to the wavelength of the incident radiation or smaller, the angular distribution of the scattered radiation is highly peaked in the forward direction.[14] Therefore with $G_z = G$, Eq. (10) gives

$$\Gamma_z = A\hbar = \frac{\text{absorbed power}}{\omega}, \tag{11}$$

which is equivalent to Eqs. (1) and (6). The only mechanism for the appearance of a torque on an isotropic particle is thus the absorption of photons, a result in agreement with Ref. 11. If the particle is asymmetric and weakly absorbent, torque is produced by scattering of photons as we will show in the following sections.

III. THE CASE OF AN ASYMMETRIC PARTICLE

The transfer of angular momentum by the absorption of light is usually impractical due to energy dissipation that occurs in the absorption process. For that purpose it is better to use weakly absorbing particles subject to a torque due to an asymmetry in their dielectric response.[15] Experimental results obtained by the angular trapping of birefringent particles show the importance of asymmetry,[16] and as we shall see, predictions of a classical analysis of the interaction of a rotating electric field with an asymmetric particle yield a similar result.

Consider an ellipsoidal object characterized by two symmetry axes perpendicular to each other, labeled d and q in Fig. 2. Consider an external electric field with angular velocity ω in the x-y plane which makes an angle β with the d-axis. We write the field as

$$\vec{E}(t) = E_0(\hat{x} + i\hat{y})e^{-i\omega t}. \tag{12}$$

In response to the field the particle acquires a dipole moment with components

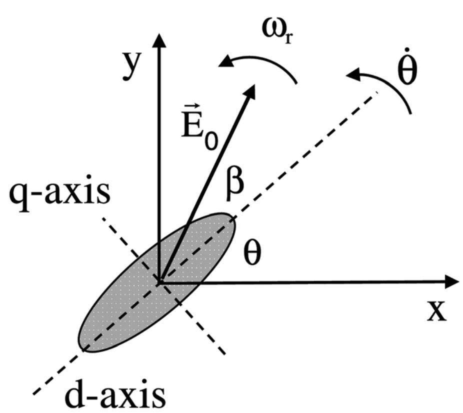

Fig. 2. Ellipsoidal particle with symmetry axes d and q in the presence of an electric field rotating with angular velocity ω_r.

$$p_d(t) = |\alpha_d||\vec{E}|\cos\beta e^{i(\omega t - \phi_d)} \tag{13a}$$

$$p_q(t) = |\alpha_q||\vec{E}|\sin\beta e^{i(\omega t - \phi_q)}, \tag{13b}$$

where $\alpha_d = |\alpha_d|e^{i\phi_d}$ and $\alpha_q = |\alpha_q|e^{i\phi_q}$ are the complex polarizabilities along d and q, respectively. The interaction between these electric dipoles and the electric field vector results in a torque acting on the object, whose time-averaged component along the electric field is given by

$$\Gamma_z = \frac{1}{2}\mathrm{Re}(p_d^* E_q) - \frac{1}{2}\mathrm{Re}(p_q^* E_d). \tag{14}$$

Because $|\vec{E}|^2 = \vec{E}\cdot\vec{E}^* = 2E_0^2$, we have

$$\Gamma_z = \frac{E_0^2}{2}\mathrm{Re}(\alpha_d - \alpha_q)\sin 2\beta. \tag{15}$$

Note that Eq. (15) can also be obtained from the derivative–$\partial W_E/\partial\beta$, where $W_E = -p_d E_d - p_q E_q$ is the electrostatic energy of an electric dipole subjected to an electric field.[17]

The instantaneous value of the absorbed power is

$$P_{\mathrm{abs}}(t) = \mathrm{Re}(E_d)\mathrm{Re}\left(\frac{dp_d}{dt}\right) + \mathrm{Re}(E_q)\mathrm{Re}\left(\frac{dp_q}{dt}\right) \tag{16a}$$

$$= |\vec{E}|\cos\beta\cos\omega t\frac{d}{dt}\left[|\alpha_d||\vec{E}|\cos\beta\cos(\omega t - \varphi_d)\right] + |\vec{E}|\sin\beta\cos\omega t\frac{d}{dt}\left[|\alpha_q||\vec{E}|\sin\beta\cos(\omega t - \varphi_q)\right] \tag{16b}$$

$$= -\omega|\alpha_d||\vec{E}|^2\cos^2\beta\sin(\omega t - \varphi_d)\cos\omega t - \omega|\alpha_q||\vec{E}|^2\sin^2\beta\sin(\omega t - \varphi_q)\cos\omega t. \tag{16c}$$

If we use simple trigonometric identities, Eq. (16c) may be rewritten as

$$P_{\mathrm{abs}}(t) = -\frac{\omega}{2}|\alpha_d||\vec{E}|^2\cos^2\beta(\sin(2\omega t - \varphi_d) - \sin\varphi_d) - \frac{\omega}{2}|\alpha_q||\vec{E}|^2\sin^2\beta(\sin(2\omega t - \varphi_q) - \sin\varphi_q). \tag{17}$$

We take the time average over the period $2\pi/\omega$ so that the oscillating terms with frequency 2ω vanish, and we obtain

$$\bar{P}_{\mathrm{abs}} = \frac{\omega}{2}|\alpha_d|\sin\varphi_d|\vec{E}|^2\cos^2\beta + \frac{\omega}{2}|\alpha_q|\sin\varphi_q|\vec{E}|^2\sin^2\beta \tag{18a}$$

$$= \frac{\omega E_0^2}{2}(\mathrm{Im}(\alpha_d + \alpha_q) + \mathrm{Im}(\alpha_d - \alpha_q)\cos 2\beta) \tag{18b}$$

We have assumed that the angle β reaches a constant value after some initial transients.

The total torque on a nanoparticle is obtained by adding the contributions of the alignment torque, Eq. (15), and the torque due to absorption, Eq. (18), leading to

$$\bar{\Gamma} = \frac{P_{\mathrm{abs}}}{\omega} + \frac{E_0^2}{2}\mathrm{Re}(\alpha_d - \alpha_q)\sin 2\beta \tag{19a}$$

$$= \frac{E_0^2}{2}(\mathrm{Im}(\alpha_d + \alpha_q) + \mathrm{Im}(\alpha_d - \alpha_q)\cos 2\beta + \mathrm{Re}(\alpha_d - \alpha_q)\sin 2\beta). \tag{19b}$$

Thus, the average torque induced by a rotating field on an ellipsoidal object includes three terms, two of which are due to the different polarizabilities along the symmetry axes. The last term in Eq. (19b) depends only on the real part of this difference and is therefore present even in the absence of absorption. This result may be extended to objects of other shapes.

IV. SOME NUMERICAL RESULTS

As an illustration of our results we now discuss the motion of a spheroid in the presence of a slowly rotating electric field. Such a field can be produced by rotating the plane of polarization of a trapping linearly polarized beam using a halfwave plate. In this form Friese and co-workers were able to rotate trapped particles so that their alignment could follow the rotation of the plane of polarization, achieving rotation frequencies of about 10 rev/s.[18] Spheroids are a special class of ellipsoids, with two of their major axes of equal length. The polarizability of an ellipsoid in a field parallel to one of its principal axes is given by[19]

$$\alpha_j = 4\pi abc\frac{\varepsilon_p - \varepsilon_m}{3\varepsilon_m + 3L_j(\varepsilon_p - \varepsilon_m)}, \tag{20}$$

where $j = x$, y, and z, are the principal axes, L_j is a geometrical factor such that $L_x + L_y + L_z = 1$, $a > b \geq c$ are the principal dimensions of the ellipsoid, and ε_p and ε_m are the dielectric permittivities of the particle and the medium where it is immersed, respectively. For prolate spheroids for which $b = c$ the geometrical factors are

$$L_y = L_z, \tag{21a}$$

$$L_x = \frac{1 - e^2}{e^2}\left[-1 + \frac{1}{2e}\ln\frac{1+e}{1-e}\right], \tag{21b}$$

where e is the eccentricity given by $e = 1 - b^2/a^2$. We assume frequency dependent permittivities for both the particle and medium of the form,

$$\varepsilon_{p,m} = K_{p,m} - i\frac{\sigma_{p,m}}{\omega\varepsilon_0}. \tag{22}$$

Here $K_{p,m}$ is the high frequency limit dielectric constant, $\sigma_{p,m}$ is the conductivity of the particle (medium), and ω is the angular frequency of the electromagnetic field.

Asymmetric particles are common in living organisms, and their dynamic control is of much interest. Examples are cells in suspension, cellular rotors, and other bioparticles present in a liquid medium.[20] As an example we consider a prolate bioparticle of length $2a = 50$ nm and radius $b = 9$ nm immersed in an aqueous solution, with $K_p = 55$, $K_m = 78.5$, $\sigma_p = 0.085\ (\Omega\text{m})^{-1}$, and $\sigma_m = 0.001\ (\Omega\text{m})^{-1}$.[21] The particle's major axis is in the x-y plane. It is illuminated by a laser beam of wavelength 1050 nm propagating in the z-direction, whose plane of polarization rotates with angular velocity ω_r (see Fig. 2).

At time t the principal axis and the electric field vector are at angles $\theta(t)$ and $\Omega = \omega_r t$ with respect to the x-axis, respectively, so that the relative angle between them is $\beta = \omega_r t - \theta(t)$. Although small, the dynamics of the particle is assumed to be well described by classical mechanics and by a bulk dielectric function. For a weakly absorbing sample, dielectric losses may be neglected, and the equation of motion can be written as[22]

$$\frac{E_0^2}{2}(\alpha_d - \alpha_q)\sin 2\beta = \frac{E_0^2}{2}(\alpha_d - \alpha_q)\sin 2(\omega_r t - \theta) = J\ddot{\theta} + \gamma\dot{\theta}. \tag{23}$$

We used Eq. (19b), assuming that α_d and α_q are given by Eq. (20), and used only the real part of the dielectric function (22). J is the moment of inertia of the particle with respect to the z-axis. The angular drag coefficient γ is obtained from Stokes' law for rotation in a viscous medium[23]

$$\gamma = \frac{\pi\eta a^3}{3[\ln(a/2b) - 0.66]}, \tag{24}$$

where η is the viscosity of the medium, $2a$ is the length of the bioparticle, and b is its radius.

After the electric field is applied and transients are dissipated, it is expected that the system reaches a steady state of rotation.[24] As can be verified from Eq. (23), if we set $\ddot{\theta} = 0$, the angular velocity $\dot{\theta}$ becomes equal to ω_r and β acquires a constant value given by

$$\sin 2\beta = \frac{2\gamma\omega_r}{E_0^2(\alpha_d - \alpha_q)}. \tag{25}$$

Note that the left-hand side of Eq. (25) cannot be greater than one, a condition that requires that the external field have a minimum threshold value for a steady solution to exist. In terms of the intensity of the beam incident on the particle the condition is

$$I_{L,\min} = \frac{\gamma c \omega_r \sqrt{K_m}}{|\alpha_d - \alpha_q|}, \tag{26}$$

where c is the speed of light in vacuum and K_m is the high frequency limit dielectric constant of the medium in which the particle is immersed. Note that the minimum laser intensity increases linearly with the rotation frequency of the external field.

We can gain insight into the solution of the nonlinear differential equation (23) for fields below the threshold by taking the limit of a very weak field. To first order, the left-hand side may then be neglected. The solution is an exponential decay $\theta_1 = Ae^{-t/\tau} + B$, with A and B constants and $\tau = J/\gamma$. In the next order of approximation we replace θ_1 on the left-hand side of Eq. (23) and realize that for times greater than τ, Eq. (23) has a solution that oscillates with angular frequency twice the external angular frequency ω_r.[25]

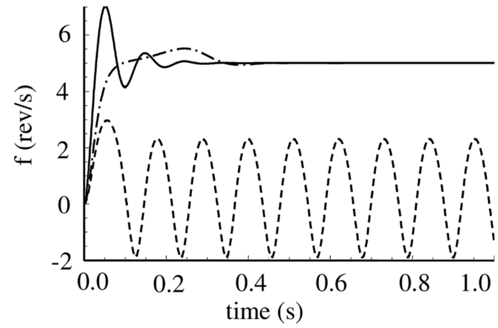

Fig. 3. The solution to Eq. (23) for the rotation frequency $f = \dot{\theta}/2\pi$ of a bioparticle that starts from rest with initial position $\theta = 0$. The viscosity is $\eta = 10^{-4}$ Ns/m^2. The laser intensities are $1.5I_0$ (continuous line), I_0 (dashed-dotted line), and $0.9I_0$ (dashed line), where $I_0 = 4.7 \times 10^9$ W/m^2 is the threshold laser intensity.

We verified these results by solving Eq. (23) numerically. Figure 3 shows the evolution of the speed of rotation starting from rest for $\eta = 10^{-4}$ Ns/m^2 and the laser intensity equal to I_0, $I_1 = 1.5I_0$, and $I_2 = 0.9I_0$, where $I_0 = 4.7 \times 10^9$ W/m^2 is the minimum intensity required to reach a steady state rotation frequency of 5 rev/s. A value of 10^{-27} kg m^2 was assumed for the moment of inertia J of the particle with respect to the z-axis, which together with the value of γ obtained from the parameters given previously and Eq. (24) give $\tau = 0.2$ s. As shown by the lower curve in Fig. 3, for a value below the minimum intensity the angular velocity oscillates with a frequency of about 10 Hz, whereas for intensity values above threshold a steady state condition is reached after a time of ≈ 0.4 s, consistent with the calculated value of τ.

Note that the rotation alternates from clockwise to counterclockwise for intensities below the threshold value I_0. Because the relative angle β no longer can reach a constant value and increases with time, the torque given by Eq. (15) changes sign causing this oscillatory effect. The transition from uniform rotation to this rocking behavior has been observed experimentally.[22]

V. CONCLUSIONS

The transfer of angular momentum from light to a material object occurs only when the electromagnetic radiation is absorbed and/or the object is asymmetric. If light is circularly polarized or the plane of polarization is rotated, a torque appears and a steady rotation is reached after some transients. When the object is asymmetric, such a steady state is achieved if the intensity of the light beam exceeds a threshold value. When this condition is not satisfied, the angular frequency of rotation of the object oscillates with twice the rotation frequency of the electric field, changing sign as time evolves as in a rocking motion. Because this effect is due to the response of the object to the oscillating electric field, it is not necessary that a laser field be used.

ACKNOWLEDGMENTS

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[25] A similar frequency doubling effect was observed by Richard Feynman in the rotation and wobbling motion of a flying disc, a finding that eventually led him to develop quantum electrodynamics. See R. P. Feynman, *Surely You're Joking, Mr. Feynman!* (Norton, New York, 1985), p. 174.