Radiation torque on a sphere caused by a circularly-polarized electromagnetic wave

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The cross sections associated with absorption, scattering, extinction, and radiation pressure for homogeneous isotropic spheres illuminated by plane waves are well known. We derive a new fundamental cross section, namely, the one which gives the time-averaged torque caused by circularly-polarized illumination. Consider a z-directed wave with pure circular polarization corresponding to a positive value for the z projection of the photon spin. Formulation of the Maxwell stress dyad of the total (incident + scattered) field gives the following torque relative to the sphere's center, $\Gamma_z = I_L \pi a^2 Q_{\text{abs}}/\omega$. Here I_L and ω are the incident wave's irradiance and angular frequency and a and Q_{abs} are the sphere's radius and Mie-theoretic absorption efficiency. Consequently the effective cross section for torque is the same as that for energy absorption $\pi a^2 Q_{\text{abs}}$ as might be expected since the scattered radiation is shown to have the same ratio of z component of angular momentum to energy as the incident wave. This result is rigorous for stationary isotropic spheres *in vacuo*. It may be used to estimate the steady-state angular velocity ω_{sz} of a sphere in a gas which is achieved when Γ_z is balanced by the viscous-drag torque. A Rayleigh-scattering approximation for Q_{abs}, which should be useful for small spheres, gives $\omega_{sz} \simeq I_L M_g M' M'' / \eta c (M'^2 + 2)$ where the sphere's refractive index is $M' + iM''$ relative to that of the gas M_g, η is the viscosity of the gas, and c is the speed of light. The radiation torque caused by elliptically-polarized illumination and the torque on stratified spheres are also discussed.

I. INTRODUCTION

The purpose of this paper is to describe a novel *ab initio* calculation of the radiation torque on an isotropic sphere illuminated by a circularly-polarized electromagnetic wave. The transport of angular momentum by a circularly-polarized wave was suggested by Poynting[1] and by Sadowsky (whose work is noted in Ref. 2). Measurements[2] of the torque on a birefringent plate provided early macroscopic evidence that the angular momentum per photon in a pure circularly-polarized state is[3] $\hbar$. Subsequently, radiation torques were measured or approximately modeled for other *anisotropic* objects[4] (e.g., screens with unidirectional conductivity and thin wires). Recently, the rotation of small particles illuminated by circularly-polarized light was observed.[5] Rotation was apparently due to radiation torques; however, details of the experiment were not given.

There appears to have been no previous analysis of the radiation torque on spheres comparable to Debye's analysis of the radiation pressure.[6] Like Debye's analysis, our calculation of the torque is based on the exact classical description of scattering of a plane wave by a stationary-homogeneous-isotropic sphere[7–10] (presently known as "Mie theory"). From symmetry considerations the torque on such a sphere will vanish unless the incident wave is at least partially circularly polarized. The emphasis of this paper is on purely circularly-polarized illumination; however, we extend our results to elliptically-polarized illumination in Sec. VIII. In Sec. IX we note applications to stratified spheres.

There are classes of radiation torques which do not require the presence of a circularly-polarized component of the illumination. These are not germane to the problem under consideration. For example, there will be a torque (relative to the sphere's center) if the sphere is illuminated by a plane-polarized Gaussian beam and is displaced from the beam's axis of symmetry. Another example is the torque on an irregularly shaped particle due to the "windmill effect" which is thought to cause cosmic dust to spin.[11,12]

The sphere under consideration has a refractive index $M = M' + iM''$ and is surrounded by a vacuum. Its relative permeability is taken to be unity. The incident illumination has a wave vector $\vec{k}_{\text{inc}} = k\hat{z}$ (Fig. 1). The incident photons are in a pure circularly-polarized state of

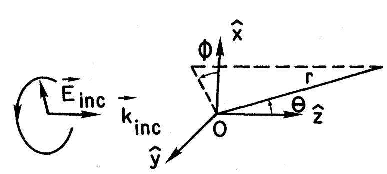

FIG. 1. The sphere (not shown) is illuminated by a circularly-polarized plane wave propagating in the direction of the z axis. The origin O of the coordinate system coincides with the center of the sphere. It is also the center about which the radiation torque is specified.

positive helicity so their z projection of spin angular momentum is positive. This incident wave's electric and magnetic fields may be written as

$$\vec{E}_{\rm inc}=E_0\,{\rm Re}(\vec{E}_1 e^{-i\omega t})\ , \tag{1}$$

$$\vec{B}_{\rm inc}=E_0\,{\rm Re}(\vec{B}_1 e^{-i\omega t})\ , \tag{2}$$

$$\vec{E}_1=(\hat{x}+i\hat{y})\exp(ikz)\ , \tag{3}$$

$$\vec{B}_1=-i\vec{E}_1\ , \tag{4}$$

and will be referred to as left circularly polarized.[13,14] This terminology and normalization, for which $\vec{E}_1$ and $\vec{B}_1$ are dimensionless, is chosen for ease of comparison with results in Jackson.[13] The time-averaged Poynting vector for this wave is $I_L\hat{z}$ where

$$I_L=E_0^2c/4\pi$$

and c is the speed of light *in vacuo*.

The scattered fields are defined such that the *total fields* (outside the sphere) are $\vec{E}_t=\vec{E}_{\rm inc}+\vec{E}_{\rm sc}$ and $\vec{B}_t=\vec{B}_{\rm inc}+\vec{B}_{\rm sc}$. It is convenient to introduce the dimensionless scattered fields $\vec{E}_2$ and $\vec{B}_2$ such that

$$\vec{E}_{\rm sc}=E_0\,{\rm Re}(\vec{E}_2 e^{-i\omega t})\ ,$$

$$\vec{B}_{\rm sc}=E_0\,{\rm Re}(\vec{B}_2 e^{-i\omega t})\ .$$

Mie-theoretic expressions for $\vec{E}_2$ and $\vec{B}_2$ are given in Sec. III for a sphere of radius a and arbitrary values of ka.

Our result for the torque, Eqs. (7) and (36), is proportional to the absorption cross section. We are unaware of any previous statement of this result, however, it appears to be consistent with the classical limit of the quantum theory of radiation according to the following argument. From the form of Eqs. (12)–(15), the expansions of both the incident and the scattered fields contain vector spherical harmonics $\vec{X}_{n,m}$ having only the azimuthal index $m=1$. Consequently, the quantization of the incident and scattered fields [following, e.g., the procedure given in Ref. 3(b)] will involve photons having only $\hbar$ units of z angular momentum. Therefore, it is reasonable that the classical limit for the z component of the torque, be proportional to the absorption. Our direct classical calculation of the flux of angular momentum into the sphere is merited because of subtleties of both classical and quantum theories of electromagnetic angular momentum (discussed, e.g., in Refs. 3, 13, and 14). Furthermore, our formulation provides a limiting case against which the solution of more complicated problems (e.g., the torque on a birefringent sphere) can be checked.

II. RADIATION TORQUE AND THE FIELD'S ANGULAR MOMENTUM FLUX

The radiation torque may be computed from the average rate of transport of field angular momentum across a surface Σ which encloses the spherical particle of radius a. It is convenient to choose Σ to be the surface of a concentric sphere of radius $r \gg a$ though the average rate of transport must be independent of r provided $r>a$. Let $\vec{J}_p$ denote the total mechanical angular momentum of the particle and let $\vec{J}_f$ denote the total angular momentum of the electromagnetic fields within Σ (both internal and external to the particle). These angular momenta and the radiation torque on the particle $\vec{\Gamma}$ are relative to an origin at the center of the particle. The conservation law for the angular momentum of this system has the following well-known surface integral form (Ref. 13, p. 264; Ref. 14, p. 169; Ref. 15):

$$\frac{d}{dt}(\vec{J}_p+\vec{J}_f)=-\int_\Sigma \hat{r}\cdot\overleftrightarrow{M}\,dS\ , \tag{5}$$

where the angular-momentum flux-density (pseudo) tensor $\overleftrightarrow{M}=\overleftrightarrow{T}\times\vec{r}$ and $\overleftrightarrow{T}$ is the Maxwell stress tensor of the total field. Let $\langle\cdots\rangle$ denote time average over the period $2\pi/\omega$ of the incident wave. The radiation torque on the sphere is

$$\vec{\Gamma}=\left\langle\frac{d}{dt}\vec{J}_p\right\rangle=-\int_\Sigma \hat{r}\cdot\langle\overleftrightarrow{T}\rangle\times\vec{r}\,dS\ , \tag{6}$$

where we use the result that fields oscillating with a *steady-state* amplitude must have $\langle d\vec{J}_f/dt\rangle=0$. The right-hand side of (6) is the average flux of angular momentum into Σ due to the incident and scattered fields. For the isotropic sphere in the *unbounded* plane incident wave under consideration, symmetry considerations require[16] $\Gamma_x=\Gamma_y=0$.

The stress dyad $\overleftrightarrow{T}$ may be written[13–15] $\overleftrightarrow{T}=\overleftrightarrow{T}^{(1)}+\overleftrightarrow{T}^{(2)}$ with

$$\overleftrightarrow{T}^{(1)}=\frac{1}{4\pi}(\vec{E}_t\vec{E}_t+\vec{B}_t\vec{B}_t)$$

$$\overleftrightarrow{T}^{(2)}=\frac{-1}{8\pi}\overleftrightarrow{1}(E_t^2+B_t^2)\ ,$$

where $\overleftrightarrow{1}$ is the unit dyad. The vector $\hat{r}\cdot\overleftrightarrow{T}^{(2)}\times\vec{r}$ vanishes identically since $\hat{r}\cdot\overleftrightarrow{1}\times\vec{r}=0$. The time averages in the remaining contribution to (6) contain dyads of the form

$$\langle\vec{E}_t\vec{E}_t\rangle=\frac{E_0^2}{2}\,{\rm Re}(\vec{E}_1\vec{E}_1^*+\vec{E}_2\vec{E}_2^*+\vec{E}_1\vec{E}_2^*+\vec{E}_2\vec{E}_1^*)\ .$$

Consequently the torque may be written

$$\Gamma_z=I_L\pi a^2 Q_\Gamma/\omega\ , \tag{7}$$

$$Q_\Gamma=Q_{11}+Q_{12}+Q_{21}+Q_{22}\ , \tag{8}$$

$$Q_{ij}=-k(2\pi a^2)^{-1}G_{ij}\ , \tag{9}$$

$$G_{ij}=r^2\,{\rm Re}\int[\hat{r}\cdot\vec{E}_i(\vec{E}_j^*\times\vec{r}\cdot\hat{z}) +\hat{r}\cdot\vec{B}_i(\vec{B}_j^*\times\vec{r}\cdot\hat{z})]d\Omega\ , \tag{10}$$

and the integration is over a solid angle of 4π sr. Equation (8) partitions the *torque efficiency factor* Q_Γ into terms involving incident fields (Q_{11}), scattered fields (Q_{22}), and mixed terms ($Q_{12}+Q_{21}$). A similar partitioning is useful when computing the energy flow in the vicinity of a scatterer.[9,17]

Equations (3) and (4) facilitate the direct evaluation of the integrals in G_{11} with the result $Q_{11}=0$. Indeed, the average angular momentum transported across Σ by the incident wave *in the absence of a scatterer* must vanish. The angular momentum, which is transported inward across the hemisphere closest to the source, is transported outward across the opposing hemisphere. In Secs. IV and V the other Q_{ij} are evaluated in the limit $kr \to \infty$. We may neglect contributions to individual Q_{ij} which vanish as $kr \to \infty$ since the *sum* of these neglected terms must vanish in (8) for any surface having $r > a$.

III. MIE THEORY OF THE SCATTERED FIELDS

To evaluate G_{21} and G_{22}, the *radial* components of the scattered fields must be expressed. This motivates the vector spherical harmonic expansion of the fields given here. We find it convenient to use Jackson's notation[13] instead of the traditional notations of optical scattering theory.[8–10] Let $\vec{X}_{n,m}$ denote the vector spherical harmonic

$$\vec{X}_{n,m}(\theta,\phi)=[n(n+1)]^{-1/2}\vec{L}Y_{n,m}(\theta,\phi)\ , \tag{11}$$

where the operator $\vec{L}=i^{-1}(\vec{r}\times\vec{\nabla})$ and the $Y_{n,m}$ are spherical harmonics obeying the Condon-Shortley phase convention and the usual normalization.[13] The angles θ and ϕ are shown in Fig. 1. Orthogonality properties of the $\vec{X}_{n,m}$ facilitate the following expansions of the dimensionless incident fields,[13] (3) and (4):

$$\vec{E}_1=\sum_{n=1}^{\infty} i^n p_n\left[j_n\vec{X}_{n,1}+\frac{1}{k}\vec{\nabla}\times j_n\vec{X}_{n,1}\right]\ , \tag{12}$$

$$\vec{B}_1=\sum_{n=1}^{\infty} i^n p_n\left[\frac{-i}{k}\vec{\nabla}\times j_n\vec{X}_{n,1}-ij_n\vec{X}_{n,1}\right]\ , \tag{13}$$

where $p_n=[4\pi(2n+1)]^{1/2}$ and j_n denotes the spherical Bessel function $j_n(kr)$.

For the circularly-polarized incident fields (12) and (13), the scattered and internal fields are describable with vector spherical harmonics having only $m=1$. (It is necessary that the scatterer consist of a medium with *isotropic* constitutive relations connecting the electric displacement and current to the electric field. This is the case under consideration. We also exclude from consideration spheres consisting of *optically active* media.[10]) We have demonstrated this assertion by including terms having $m\neq 1$ in the expansions given below with the result that the coefficients of these terms vanish. It is convenient to expand the dimensionless scattered fields as

$$\vec{E}_2=-\sum_{n=1}^{\infty} i^n p_n\left[b_n h_n\vec{X}_{n,1}+\frac{1}{k}a_n\vec{\nabla}\times h_n\vec{X}_{n,1}\right]\ , \tag{14}$$

$$\vec{B}_2=\sum_{n=1}^{\infty} i^n p_n\left[\frac{i}{k}b_n\vec{\nabla}\times h_n\vec{X}_{n,1}+ia_n h_n\vec{X}_{n,1}\right]\ , \tag{15}$$

where h_n is the spherical Hankel function $h_n^{(1)}(kr)$ (which is appropriate for an outward propagating wave), and the unknown coefficients are determined below.

The fields *internal* to the particle may be written $\vec{E}_{\text{int}}=E_0\,\text{Re}(\vec{E}_3e^{-i\omega t})$ and $\vec{B}_{\text{int}}=E_0\,\text{Re}(\vec{B}_3e^{-i\omega t})$, where the dimensionless internal fields, $\vec{E}_3$ and $\vec{B}_3$, may be expanded as

$$\vec{E}_3=\sum_{n=1}^{\infty} i^n p_n\left[\beta_n j_n(k_3r)\vec{X}_{n,1}+\frac{i}{kM^2}\alpha_n\vec{\nabla}\times j_n(k_3r)\vec{X}_{n,1}\right]\ ,$$

$$\vec{B}_3=\sum_{n=1}^{\infty} i^n p_n\left[\frac{-i}{k}\beta_n\vec{\nabla}\times j_n(k_3r)\vec{X}_{n,1}+\alpha_n j_n(k_3r)\vec{X}_{n,1}\right]\ .$$

These fields may be shown to satisfy Maxwell's equations appropriate to a material with a complex refractive index M, wave number $k_3=Mk$, and a relative permeability of unity.

The unknown coefficients a_n, b_n, α_n, and β_n are determined by requiring that the following boundary conditions be met at the particle's surface at $r=a$: $\hat{r}\times(\vec{E}_1+\vec{E}_2-\vec{E}_3)=0$ and $\hat{r}\times(\vec{B}_1+\vec{B}_2-\vec{B}_3)=0$. Only the a_n and b_n are needed for the evaluation of (10) on a surface having $r\gg a$. The required coefficients are

$$a_n=\frac{M\psi(M\rho)\psi'(\rho)-\psi(\rho)\psi'(M\rho)}{M\psi(M\rho)\xi'(\rho)-\xi(\rho)\psi'(M\rho)}\ , \tag{16a}$$

$$b_n=\frac{\psi(M\rho)\psi'(\rho)-M\psi(\rho)\psi'(M\rho)}{\psi(M\rho)\xi'(\rho)-M\xi(\rho)\psi'(M\rho)}\ . \tag{16b}$$

Here $\rho=ka$, ψ and ξ are Riccati-Bessel functions, $\psi(\rho)=\rho j_n(\rho)$, $\xi(\rho)=\rho h_n^{(1)}(\rho)$, and primes denote differentiation with respect to the arguments indicated. These a_n and b_n are identical to the usual "scattering coefficients" of Mie theory and algorithms for their computation are well established.[10] This correspondence motivated the normalization used in (14) and (15). The series in (14) and (15) converge rapidly for n somewhat in excess of ka when $ka\gg 1$. Equations (14)–(16) are equivalent to results of Chew *et al.*[18] and to the far-zone predictions of standard Mie theory when adapted to circularly-polarized illumination.[10]

The scattered fields are attributable to oscillating electric (E) and magnetic (M) multipoles. The following subdivision of fields is convenient: $\vec{E}_2=\vec{E}^{(E)}+\vec{E}^{(M)}$, $\vec{B}_2=\vec{B}^{(E)}+\vec{B}^{(M)}$ where

$$\vec{E}^{(E)}=\frac{i}{k}\sum_n a_n^{(E)}\vec{\nabla}\times h_n\vec{X}_{n,1},\quad \vec{E}^{(M)}=\sum_n a_n^{(M)}h_n\vec{X}_{n,1}\ ,$$

$$\vec{B}^{(M)}=\frac{-i}{k}\sum_n a_n^{(M)}\vec{\nabla}\times h_n\vec{X}_{n,1},\quad \vec{B}^{(E)}=\sum_n a_n^{(E)}h_n\vec{X}_{n,1}\ ,$$

$$a_n^{(E)}=-p_na_ni^{n-1},\quad a_n^{(M)}=-p_nb_ni^n\ . \tag{17}$$

This subdivision gives $\hat{r}\cdot\vec{E}^{(M)}=\hat{r}\cdot\vec{B}^{(E)}=0$; $\hat{r}\cdot\vec{E}^{(E)}$ and $\hat{r}\cdot\vec{B}^{(M)}$ may be found from[13]

$$\hat{r}\cdot[\vec{\nabla}\times h_n^{(1)}(kr)\vec{X}_{n,m}]=i[n(n+1)]^{1/2}h_n^{(1)}(kr)Y_{n,m}r^{-1}\ , \tag{18}$$

which follows from (11) and the identity $\vec{\nabla}\times[\vec{r}\times\vec{g}(\vec{r})]=\vec{r}(\vec{\nabla}\cdot\vec{g})-r(2r^{-1}+\partial/\partial r)\vec{g}$. Since the G_{ij} are to be evaluated in the limit the radius r of Σ is such that $kr\to\infty$, the following approximations will be useful:[13]

$$\vec{E}_2\times\hat{r}\simeq-\vec{B}_2\;, \tag{19a}$$

$$\vec{B}_2\times\hat{r}\simeq\vec{E}_2\;, \tag{19b}$$

where the terms omitted from the right-hand sides are $O(1/kr)$ smaller than those retained.

IV. EVALUATION OF THE MIXED TERMS Q_{12} AND Q_{21}

The integrand of G_{12}, call it I_{12}, may be simplified by using Eq. (19) and Eq. (4). This procedure gives $I_{12}\simeq(\vec{r}\cdot\vec{B}_1)(\hat{z}\cdot\vec{E}_2^*)-(\vec{r}\cdot\vec{E}_1)(\hat{z}\cdot\vec{B}_2^*)=-\vec{r}\cdot\vec{E}_1\hat{z}\cdot(\vec{B}_2^*+i\vec{E}_2^*)$. The error vanishes as $kr\rightarrow\infty$ as it does in the following approximations: $\vec{E}_2\simeq\vec{B}^{(E)}\times\hat{r}+\vec{E}^{(M)}$ and $\vec{B}_2\simeq-\vec{E}^{(M)}\times\hat{r}+\vec{B}^{(E)}$. Use of the latter approximations and the limiting form of $h^{(1)}(kr)$ give

$$\begin{aligned}(\vec{B}_2^*+i\vec{E}_2^*)&\\ &\simeq(\vec{B}^{(E)*}+i\vec{E}^{(M)*})\cdot(\vec{\vec{1}}+i\vec{\vec{1}}\times\hat{r})\\ &\simeq\sum_n i^{n+1}\frac{e^{-ikr}}{kr}(a_n^{(E)*}+ia_n^{(M)*})(\vec{X}_{n,1}^*+i\vec{X}_{n,1}^*\times\hat{r})\;.\end{aligned} \tag{20}$$

Equation (18) can also be used with $h^{(l)}(kr)$ replaced by $j_n(kr)$ so that $\vec{r}\cdot\vec{E}_1$ becomes

$$\vec{r}\cdot\vec{E}_1=\sum_n P_n k^{-1}j_n(kr)Y_{n,1}\;, \tag{21}$$

where $P_n=p_n[n(n+1)]^{1/2}i^{n+1}$. Insertion of (20) and (21) into I_{12} gives $G_{12}\rightarrow\mathrm{Re}(U^++U^-)$ as $kr\rightarrow\infty$ where we have defined

$$U^{\pm}(kr)=\frac{-1}{k^3}\sum_{n'}\sum_n P_{n'}p_n kr j_{n'}(kr)e^{-ikr}(a_n^*+b_n^*)F_{n,n'}^{\pm}$$

$$F_{n,n'}^{+}=\int\hat{z}\cdot\vec{X}_{n,1}^*Y_{n',1}d\Omega\;, \tag{22}$$

$$F_{n,n'}^{-}=i\int\hat{z}\cdot(\vec{X}_{n,1}^*\times\hat{r})Y_{n',1}d\Omega\;.$$

Evaluation of these integrals gives

$$\begin{aligned}F_{n,n'}^{+}&=[n(n+1)]^{-1/2}\int(L_zY_{n,1})^*Y_{n',1}d\Omega\\ &=\delta_{n,n'}[n(n+1)]^{-1/2}\;,\end{aligned} \tag{23}$$

$$F_{n,n'}^{-}=W_n^{+}\delta_{n+1,n'}-W_n^{-}\delta_{n-1,n'}\;, \tag{24}$$

$$W_n^{+}=\left[\frac{n^2(n+2)}{(2n+1)(n+1)(2n+3)}\right]^{1/2},\qquad W_n^{-}=\left[\frac{(n+1)^2(n-1)}{(2n+1)n(2n-1)}\right]^{1/2}. \tag{25}$$

The proof of (24) is summarized in Appendix A.

Since we need only evaluate $U^{\pm}(kr\rightarrow\infty)$, it is permissible to replace $krj_{n'}(kr)$ by $\sin(kr-n'\pi/2)$ in Eq. (22). The required summations may be condensed by using (23) and (24) and algebraic manipulations give

$$U^{\pm}(kr\rightarrow\infty)=-\frac{2\pi}{k^3}\sum_{n=1}^{\infty}(2n+1)(a_n^*+b_n^*)\times[1\mp(-1)^n e^{-i2kr}]\;.$$

The oscillating terms cancel in U^++U^- and consequently also in $G_{12}(kr\rightarrow\infty)$ with the result

$$Q_{12}=\frac{2}{(ka)^2}\sum_{n=1}^{\infty}(2n+1)\,\mathrm{Re}(a_n+b_n)\;.$$

The right-hand side is identical to the *extinction efficiency*[7–10] Q_{ext} of a sphere where πa^2Q_{ext} is the extinction cross section. We conclude that

$$Q_{12}=Q_{\mathrm{ext}}\;. \tag{26}$$

The integrand of G_{21}, call it I_{21}, may be simplified by using Eqs. (3) and (4) and the identities $(\vec{E}_1\times\vec{r})\cdot\hat{z}=(\hat{z}\times\vec{E}_1)\cdot\vec{r}$ and $(\vec{B}_1\times\vec{r})\cdot\hat{z}=(\hat{z}\times\vec{B}_1)\cdot\vec{r}$. The result may be written

$$I_{21}=(\hat{r}\cdot\vec{E}^{(E)})(\vec{r}\cdot\vec{B}_1^*)-(\hat{r}\cdot\vec{B}^{(M)})(\vec{r}\cdot\vec{E}_1^*)\;.$$

Application of Eqs. (18) and (21) shows that I_{21} is of order $k^{-2}r^{-3}$. Consequently, G_{21} is of order $k^{-2}r^{-1}$ and Q_{21} may be neglected from the sum in Eq. (8) for the reasons noted at the end of Sec. II. This conclusion has been confirmed by a detailed evaluation of G_{21} which need not be reproduced here.

V. EVALUATION OF THE SCATTERED-FIELD TERM Q_{22}

The integrand of G_{22}, call it I_{22}, may be simplified by using Eq. (19). This procedure gives

$$I_{22}\simeq-(\vec{r}\cdot\vec{E}_2)(\hat{z}\cdot\vec{B}_2^*)+(\vec{r}\cdot\vec{B}_2)(\hat{z}\cdot\vec{E}_2^*)\;.$$

The real part of the second term may be rewritten such that

$$\mathrm{Re}\,I_{22}\simeq\mathrm{Re}[-(\vec{r}\cdot\vec{E}_2)(\hat{z}\cdot\vec{B}_2^*)+(\vec{r}\cdot\vec{B}_2^*)(\hat{z}\cdot\vec{E}_2)]=\hat{z}\cdot\vec{j}_2$$

where

$$\vec{j}_i=\mathrm{Re}[\vec{r}\times(\vec{E}_i\times\vec{B}_i^*)],\quad i=1,2\;. \tag{27}$$

The time-averaged classical angular momentum density of the scattered field[13,14,19] (considered alone such that interference with the incident field is neglected) is manifestly proportional to $\vec{j}_2$. It is convenient to use the subdivisions $\vec{E}_2=\vec{E}^{(E)}+\vec{E}^{(M)}$ and $\vec{B}_2=\vec{B}^{(E)}+\vec{B}^{(M)}$ (which were introduced in Sec. III) and to obtain the following expression: $\vec{j}_2=\mathrm{Re}(\vec{r}\cdot\vec{B}^{(M)*}\vec{E}_2-\vec{r}\cdot\vec{E}^{(E)}\vec{B}_2^*)$. The terms $\vec{E}^{(E)}$ and $\vec{B}^{(M)*}$ may be replaced by using the relations

$$\vec{E}^{(E)}=\frac{i}{k}\vec{\nabla}\times\vec{B}^{(E)},\quad\vec{B}^{(M)}=\frac{-i}{k}\vec{\nabla}\times\vec{E}^{(M)}$$

which follow from the definitions of the multipole fields. This procedure and a vector identity give

$$\vec{j}_2=k^{-1}\mathrm{Re}[-\vec{E}_2\vec{L}\cdot(\vec{E}^{(M)*})+\vec{B}_2^*(\vec{L}\cdot\vec{B}^{(E)})]\;,$$

where $\vec{L}$ is defined as below Eq. (11). The $\vec{E}^{(E)}$ and $\vec{B}^{(M)}$ parts of $\vec{E}_2$ and $\vec{B}_2$ may similarly be eliminated to obtain the following expression:

$$\vec{j}_2 = k^{-1}\operatorname{Re}\{\vec{B}^{(E)*}(\vec{L}\cdot\vec{B}^{(E)}) + \vec{E}^{(M)}(\vec{L}\cdot\vec{E}^{(M)})^* + ik^{-1}[(\vec{\nabla}\times\vec{E}^{(M)*})(\vec{L}\cdot\vec{B}^{(E)}) + (\vec{\nabla}\times\vec{B}^{(E)})(\vec{L}\cdot\vec{E}^{(M)})^*]\} . \quad (28)$$

For the case under consideration, Eq. (10) becomes

$$G_{22}(kr\to\infty) = r^2\hat{z}\cdot\int \vec{j}_2 d\Omega . \quad (29)$$

Evaluation of this integral proceeds from (28) and the multipole expansions of the fields in terms of the coefficients given by (17). The expansions are evaluated in the limit $kr\to\infty$ with the approximations $h^{(1)}(kr) \simeq (-i)^{n+1}(kr)^{-1}\exp(ikr)$ and

$$\vec{\nabla}\times[h^{(1)}(kr)\vec{X}_{n,m}] \simeq (-i)^n r^{-1}e^{ikr}\hat{r} \times\vec{X}_{n,m} + O(kr^{-2}) .$$

The proof of the latter is similar to the one described for Eq. (18). In the limit $kr\to\infty$, we find[20]

$$r^2\int \vec{j}_2 d\Omega = \frac{1}{k^3}\operatorname{Re}\sum_{n'=1}^{\infty}\sum_{n=1}^{\infty}(i)^{n'}(-i)^n[(a_{n'}^{(E)*}a_n^{(E)} + a_{n'}^{(M)*}a_n^{(M)})\vec{K}^{+}_{n',n} + (a_{n'}^{(E)*}a_n^{(M)} - a_{n'}^{(M)*}a_n^{(E)})\vec{K}^{-}_{n',n}] , \quad (30)$$

$$\vec{K}^{+}_{n',n} = \int (\vec{L}\cdot\vec{X}_{n',1})^*\vec{X}_{n,1}d\Omega , \quad (31)$$

$$\vec{K}^{-}_{n',n} = \int (\vec{L}\cdot\vec{X}_{n',1})^*\hat{r}\times\vec{X}_{n,1}d\Omega . \quad (32)$$

It is convenient to partition Q_{22} as $Q_{22} = Q_{22}^{+} + Q_{22}^{-}$ where from Eqs. (9), (17), (29), and (30) we find

$$Q_{22}^{+} = \frac{-1}{2\pi(ka)^2}\operatorname{Re}\sum_{n'}\sum_{n} p_{n'}p_n(a_{n'}^*a_n + b_{n'}^*b_n)\hat{z}\cdot\vec{K}^{+}_{n',n} ,$$

$$Q_{22}^{-} = \frac{-1}{2\pi(ka)^2}\operatorname{Re} i\sum_{n'}\sum_{n} p_{n'}p_nN_{n',n}\hat{z}\cdot\vec{K}^{-}_{n',n} , \quad (33)$$

where $N_{n',n} = a_{n'}^*b_n + b_{n'}^*a_n$. Equation (11) and the relations $L^2Y_{n,m} = n(n+1)Y_{n,m}$ and $L_zY_{n,m} = mY_{n,m}$ give $\hat{z}\cdot\vec{K}^{+}_{n',n} = \delta_{n',n}$ and, consequently,

$$Q_{22}^{+} = \frac{-2}{(ka)^2}\sum_{n=1}^{\infty}(2n+1)(|a_n|^2 + |b_n|^2) . \quad (34)$$

Using Eq. (A3) we find that

$$\hat{z}\cdot\vec{K}^{-}_{n',n} = [n'(n'+1)]^{1/2}\int Y_{n',1}^*\hat{z}\cdot(\hat{r}\times\vec{X}_{n,1})d\Omega = i[n'(n'+1)]^{1/2}(W_n^-\delta_{n',n-1} - W_n^+\delta_{n',n+1}) ,$$

where $W_n^{\pm}$ is defined in Eq. (25). The required summations for Q_{22}^{-} reduce to

$$Q_{22}^{-} = \frac{2}{(ka)^2}\sum_{n=1}^{\infty} n(n+2)\operatorname{Re}(N_{n,n+1} - N_{n+1,n}) .$$

Since $\operatorname{Re}N_{n,n+1} = \operatorname{Re}N_{n+1,n}$, we find that $Q_{22}^{-}=0$.

Inspection of Eq. (34) gives $Q_{22}^{+} = -Q_{sc}$ where Q_{sc} is the usual *scattering efficiency* according to Mie theory[7–10] and πa^2Q_{sc} is the scattering cross section. We conclude that

$$Q_{22} = -Q_{sc} . \quad (35)$$

VI. DISCUSSION OF THE Q_{ij}, Q_Γ, AND THE RADIATION TORQUE

The efficiency Q_{22} accounts for the angular momentum radiated by electric and magnetic multipoles oscillating with the strengths given by Eq. (17). These are the multipole strengths appropriate for an incident wave of positive helicity. Equation (35) has the following quantum interpretation: the wave radiated by these multipoles, when considered separate from the incident wave, radiates $\hbar$ units of z angular momentum per $\hbar\omega$ units of energy radiated. Our proof of Eq. (35) made use of the result $Q_{22}^{-}=0$ which has the following interpretation: the interference between the electric and magnetic multipoles does not contribute to the radiation of z angular momentum. (For a general collection of multipoles, interference affects the radiated angular momentum.[21]) The results $Q_{11}=Q_{21}=0$ together with Eq. (26) may be interpreted as follows: the extinction of $\hbar\omega$ units of energy from the incident wave makes available $\hbar$ units of z angular momentum for reradiation by the scattered wave and for producing torque on the sphere.

The torque is proportional to the rate of absorption of the incident wave's energy. The results $Q_{11}=Q_{21}=0$ together with Eqs. (8), (26), and (35) give the following torque efficiency factor:

$$Q_\Gamma = Q_{ext} - Q_{sc} = Q_{abs} , \quad (36)$$

where $Q_{abs} = Q_{ext} - Q_{sc}$ is the usual definition of the absorption efficiency[8–10] and πa^2Q_{abs} is the cross section for absorption. Evidently Q_Γ vanishes for an idealized lossless dielectric sphere. Real materials have $M''>0$ if we neglect the possibility of stimulated emission so that $Q_{abs}>0$.

The Mie series for Q_{ext} and Q_{sc} (and hence for Q_{12}, Q_{22}, Q_{abs}, and Q_Γ) may be evaluated with computer algorithms.[10,22] Computationally efficient approximations for Q_{abs} have also been derived.[10,23,24] We consider Q_{abs} for a few cases.

(a) *Large nearly-black spheres.* Consider a sphere having $ka\gg1$, $M'\simeq1$, $M''\ll1$, and $kaM''\gg1$. Such a sphere should be weakly reflecting and yet highly absorbing. It is to be expected that $Q_{ext} \simeq 2$ and (due to forward diffraction) $Q_{sc} \simeq 1$; consequently we expect $Q_{abs} \simeq 1$. This was confirmed for the case $M = 1 + i0.01$ by using Wiscombe's Mie scattering algorithm[22] to compute Q_{abs} for ka of 500, 1000, and 2000; we found Q_{abs} of 0.9966, 0.9993, and 0.9987, respectively.

(b) *Spheres with weak or moderate absorption.* Bohren and co-workers have compared geometrical-optics approximations for Q_{abs} with Mie computations for weakly and moderately absorbing spheres.[10,24] The geometrical-optics approximations predict Q_{abs} which increase with radius; the exact Q_{abs} are similar to the approximate ones except for a superposed ripple structure. The geometrical-optics contribution to Q_{abs} can be substantial for a range of radii and wavelengths $\lambda = 2\pi/k$ for cases of practical interest calculated in Ref. 24. For example, water droplets at $\lambda = 2.0$ μm have[24] $M = 1.304 + i0.001\,082$ and Q_{abs} increases smoothly from $\simeq 0.43$ to $\simeq 0.66$ as a increases from 50 to 100 μm.

(c) *Small spheres with strong surface modes.* Q_{abs} can be greatly enhanced for small metallic particles at ultraviolet frequencies because of surface plasmons. Enhanced absorption is also possible for small insulating particles at infrared frequencies because of surface phonons (Chap. 12 of Ref. 10 and Ref. 17). An example of the former enhancement is the prediction[10,17] that an aluminum sphere having $ka \simeq 0.3$ has $Q_{abs} \simeq 18$ when $\hbar\omega \simeq 8.8$ eV. It should be remembered that our derivation of $Q_\Gamma = Q_{abs}$ required the scatterer to have isotropic constitutive relations and that this is not necessarily the case for these particles.

(d) *Rayleigh scattering.* If the Mie series for Q_{ext} and Q_{sc} are dominated by terms proportional to $\mathrm{Re}a_1$ and $|a_1|^2$, respectively, then the scattering is usually referred to as[8,9] Rayleigh scattering. If ka is sufficiently small and $|M|ka \ll 1$ the resulting dominance of a_1 leads to the following approximation[8–10]

$$Q_{abs} \simeq 4ka \,\mathrm{Im}[(M^2-1)/(M^2+2)] \ . \qquad (37)$$

For this approximation to be accurate, it is necessary that the right-hand side be $\ll 3(ka)^{-2}$. The scattered field is attributable to an electric dipole spinning with an angular velocity ω about the z axis. For such a dipole, the evaluation of Eq. (29) is relatively simple[14] and the multipole interference term $Q_{\overline{22}}$ need not be considered. The result is that $\hbar$ units of z angular momentum are radiated per $\hbar\omega$ units of energy radiated. This confirms Eq. (35) for the special case of Rayleigh scattering.

Our prediction for the radiation torque, Eqs. (7) and (36), is the exact classical result which should be applicable to radiation fields containing many photons. (For a discussion of the relevant limit of quantum mechanics, see e.g., Sec. 16.3 of Ref. 13.) A single incident plane wave of infinite extent is an idealized case. The predicted torque should be applicable to spheres which are centered in a circularly polarized Gaussian beam whose width greatly exceeds the sphere's diameter. Tam and Corriveau[25] have calculated the extinction and scattering by a sphere in a Gaussian beam and have explicitly demonstrated that the Q_{ext} and Q_{sc} reduce to the Mie theoretic results in the limit of a beam of infinite extent.

There has been considerable interest in the classical theory of the angular momentum transport of circularly-polarized plane waves and bounded beams.[3(a),14,26–28] The time-averaged classical z component of the angular momentum density of the incident wave, which is proportional to $\hat{z}\cdot\vec{j}_1$ of Eq. (27), vanishes for a plane wave. Simmons and Guttmann[26] give a pedagogical discussion of the classical perspective of the radiation torque on an absorbing disk illuminated by a wave which is well approximated by Eqs. (1)–(4). When the disk is large in comparison with the wavelength, the scattering is nearly z directed. The z projection of the angular momentum density of the *total* (incident + scattered) field is predominantly negative. Consequently the torque Γ_z is positive even though $\hat{z}\cdot\vec{j}_1$ vanishes at the position of the disk. A similar argument applies to the large nearly-black sphere which was previously described as case (a). The classical field angular momentum may be separated into an orbital and a spin part.[3,14,28] This separation was not required in our analysis. Instead, our calculation computed the torque directly from the Maxwell stress tensor by way of Eq. (6).

A comparison of the form of Eqs. (7) and (36) with the form of the net force $\vec{f}$ on the sphere due to radiation pressure is in order. Debye's theory[6,8–10] gives $f_x = f_y = 0$ and $f_z = I_L \pi a^2 Q_{pr}/c$ where the *efficiency for radiation pressure* $Q_{pr} = Q_{ext} - Q_{sc}g$. Here g is commonly known as the asymmetry parameter. It is the average of $\cos\theta$ over 4π sr with the scattered irradiance as a weighting function; $Q_{sc}g$ may be expressed in terms of a series containing the scattering coefficients a_n and b_n. Unlike Γ_z, f_z does not vanish for a lossless isotropic dielectric sphere and it contains no intrinsic ω^{-1} factor.

In Beth's experiment,[2] the illuminated object was a half-wave plate (in the form of a crystalline-quartz disk) for which the helicity of the forward transmitted wave was reversed from that of the incident wave. The existence of a radiation torque along the propagation axis did not rely on the absorption of energy. It is to be anticipated that circularly-polarized illumination of a sphere consisting of an *anisotropic* dielectric media will generally produce nonvanishing Γ_x, Γ_y, and Γ_z even if absorption is negligible. Furthermore, linearly polarized illumination of such spheres will produce nonvanishing $\vec{\Gamma}$ except for specific orientations.

In the remainder of this paper we consider some applications and extensions of our calculation of the radiation torque. Consideration is limited to spheres having isotropic constitutive relations.

VII. ROTATION DRIVEN BY THE RADIATION TORQUE

In response to the radiation torque Γ_z, the sphere will undergo an angular acceleration. If the sphere is surrounded by a fluid, there will be an additional torque Γ_{Dz} due to viscous drag. If the fluid's refractive index M_g is close to unity, as is the case for air, we assume that Eq. (7) and $Q_\Gamma = Q_{abs}$ are good approximations where M, used in the calculation of Q_{abs}, is replaced by the refractive index of the sphere relative to that of the fluid and k becomes the wave number in the outer dielectric $M_g\omega/c$. (The resulting Γ_z may be exact; however, we have not considered in detail the problem of electromagnetic momentum transport *within* dielectric media as reviewed by Brevik.[27]) The z-angular velocity of the sphere, ω_{sz} will reach a steady value when $\Gamma_z+\Gamma_{Dz}=0$. The drag torque is[29] $\Gamma_{Dz}=-8\pi\eta a^3\omega_{sz}$, where η is the viscosity of the surrounding fluid. It is assumed that the sphere rotates as a rigid body and that the flow is Stokes flow with the fluid at infinity at rest. The steady-state angular velocity becomes

$$\omega_{sz}=I_L Q_{\text{abs}}/8\omega\eta a \ . \tag{38}$$

When the sphere is sufficiently small for Eq. (37) to be applicable, the steady-state angular velocity becomes

$$\omega_{sz}\simeq\frac{I_L M_g}{2\eta c}\,\text{Im}\left[\frac{M^2-1}{M^2+2}\right] \tag{39}$$

which is independent of a and depends on ω by way of the relative refractive index M. It is assumed that the sphere is sufficiently large that classical continuum theories apply both in the electrodynamics (which was implicit in our use of constitutive relations appropriate for bulk material[10]) and to the mechanics (as in the derivation[29] of Γ_{Dz}). When $M=M'+iM''$ is such that $M''\ll M'$, Eq. (39) simplifies to

$$\omega_{sz}\simeq I_L M_g M'M''/\eta c(M'^2+2) \ .$$

Consider the case of a spherical drop of water surrounded by air for the conditions noted in case (b) of Sec. VI, namely $\lambda=2.0$ μm and $M\simeq1.304+i0.001\,082$. The viscosity of air at 20 °C is 0.018 cP and we take $I_L=10^8$ W/m^2. The drop's rotation should approximate that of a rigid body. For a drop with a radius of 50 μm, $Q_{\text{abs}}\simeq0.43$ and Eq. (38) gives $\omega_{sz}\simeq6.3$ sec^{-1}. For small drops ($a\lesssim0.2$ μm), Eq. (39) is applicable; it gives $\omega_{sz}\simeq7.1$ sec^{-1}. The effect of radiation torques, though not large, may be observable for drops or other spheres illuminated with circularly-polarized laser beams. Since the effects of viscous drag appear to be an important limitation, it may be preferable to observe the response to radiation torques on spheres optically levitated in *a vacuum* as noted in Ref. 5.

Some caveats concerning our analysis of the rotation of spheres should be noted.

(a) Molecular collisions with the sphere's surface give rise to random rotations commonly known as the Brownian rotation.[30] The mean-square angular displacement in a time interval t was estimated by Einstein[31] to be $KTt/4\pi\eta a^3$ rad^2, where K is Boltzmann's constant and T is the temperature. When compared with the response to radiation torque, these angular displacements may be significant, especially when the sphere's radius a is small.

(b) The result $Q_\Gamma=Q_{\text{abs}}$ indicates that torques are accompanied by the absorption of energy. The microscopic loss mechanisms incorporated into M'' may be classified as "nonradiative" or as "radiative." In the nonradiative case, the absorption gives rise to heating of the sphere. This heating leads to thermal reradiation of energy and heat conduction away from the sphere. It results in photophoretic forces on the sphere;[32] however, the torques due to these forces should be negligible if both the thermal and the electromagnetic constitutive relations are isotropic. In the relatively unusual case of radiative loss mechanisms, some of the energy is radiated at frequencies other than ω because of fluorescence. For example, the sphere may be embedded with fluorescent molecules.[18] Torques due to fluorescence should be negligible if this embedding has radial symmetry and if the radiating state is buffered from the polarization of the exciting fields within the sphere $\vec{E}_{\text{int}}$ and $\vec{B}_{\text{int}}$. Such buffering could result from intermediate nonradiative transitions. Torques due to fluorescence and to heating were not included in our analysis.

(c) The field scattered from a spinning scatterer may contain frequency components which differ from the frequency ω of the incident wave.[33] Furthermore, the irradiance distribution of the scattered field for spheres may be affected by the angular velocity.[34] The resulting errors in our calculation of the torque should be small when $\omega_{sz}\ll\omega$ and $\omega_{sz}a\ll c$.

(d) The nonsphericity of liquid drops will not, necessarily, significantly affect the radiation torque relative to the drop's centroid. Drops deform into an oblate spheroid if spun at sufficiently low ω_{sz} in a gas. Let a denote the radius of the equivalent sphere which is defined to have the same volume as the drop. If the parameter $\tilde{\omega}=\omega_{sz}^2a^3(\rho_{\text{liq}}-\rho_{\text{gas}})/8\gamma$ is $\ll1$, the ratio of the minor to major axis lengths of the spheroid is[35] $\simeq(1-\tilde{\omega})$. Here ρ denotes the indicated mass density and γ is the surface tension. For example, a drop of water having $a=50$ μm and $\omega_{sz}=6.3$ s^{-1} has $\tilde{\omega}\simeq10^{-8}$. Drops will also deform in response to the surface distribution of radial radiation stresses. For weakly absorbing spherical drops the geometrical-optics contribution to πa^2Q_{abs} is roughly proportional to a^3; consequently, if the ripple structure is also negligible, absorption is seen to be a *volume* effect.[10] The absorption cross section should change only slightly with small deviations from sphericity. The $\vec{\Gamma}$ computed for the equivalent sphere should well approximate that of the nearly spherical object whose surface is one of revolution about the z axis.

VIII. ELLIPTICALLY-POLARIZED ILLUMINATION OF SPHERES

Consider the case of right circularly-polarized illumination with an irradiance I_R. The incident field is given by Eqs. (1) and (2) with Eqs. (3) and (4) replaced by $\vec{E}_1=(\hat{x}-i\hat{y})\exp(ikz)$ and $\vec{B}_1=i\vec{E}_1$. This corresponds to incident photons in a pure circularly-polarized state of negative helicity.[13,14] Expansions of the incident and scattered fields[13] are similar in form to Eqs. (12)–(15) except for some reversals in signs and the replacement of all $\vec{X}_{n,1}$ by $\vec{X}_{n,-1}$. Symmetry considerations show that Eqs. (7) and (36) are replaced by $\Gamma_z=-I_R\pi a^2Q_{\text{abs}}/\omega$, where πa^2Q_{abs} is again the absorption cross section of the sphere.

Consider now the case of elliptically-polarized illumination. The incident wave's electric field may be written

$$\vec{E}_{\text{inc}}=\text{Re}\{[E_L(\hat{x}+i\hat{y})+E_R(\hat{x}-i\hat{y})]e^{ikz-i\omega t}\} \ ,$$

where E_L and E_R are complex constants which determine the ellipsometric parameters of the plane wave.[10] The expansions of the incident and scattered fields now contain both $\vec{X}_{n,1}$ and $\vec{X}_{n,-1}$. An important result is that the *cross terms* between fields having $m=1$ and those having $m=-1$ do not contribute to the G_{ij} because of the integration over the azimuthal angle. This result follows from the forms of Eqs. (11), (A1), and (A2). As a consequence of this result and of our previous results for pure circular polarization, the torque is predicted to be

$$\Gamma_z=(I_L-I_R)\pi a^2 Q_{\text{abs}}/\omega \,, \tag{40}$$

where $I_L=|E_L|^2c/4\pi$ and $I_R=|E_R|^2c/4\pi$ are the irradiances of the left- and right-handed components considered separately. Consideration of the integration over azimuthal angle in Eq. (6) gives the result $\Gamma_x=\Gamma_y=0$ where use is made of vector spherical harmonic expansions of the fields. In the case of a linearly polarized incident wave, $I_L=I_R$ and we find $\vec{\Gamma}=0$.

IX. THE RADIATION TORQUE ON STRATIFIED SPHERES

In this section we consider the generalization of our previous results to a sphere whose optical properties are stratified such that the complex refractive index may vary *radially* from the center to the outer surface. For definiteness we first consider the problem of a homogeneous sphere coated with a homogeneous layer of uniform thickness illuminated by the wave of Eqs. (1)—(4). The problem of scattering from such a sphere was first solved by Aden and Kerker.[36] For the purpose of our present discussion we need only note that the vector spherical harmonic expansion of the scattered field is given by Eqs. (14) and (15) *provided* the scattering coefficients a_n and b_n are suitably chosen. Concise equations for the suitable a_n and b_n, which replace those of Eqs. (16), are well known and need not be reproduced here, see, e.g., Eq. (8.2) of Ref. 10. With this replacement, the calculations of the Q_{ij} are identical to those given in Secs. IV and V. The torque on the coated sphere is given by Eqs. (7) and (36) where $\pi a^2 Q_{\text{abs}}$ is the absorption cross section of the coated sphere and Q_{ext} and Q_{sc} are computed using the revised a_n and b_n.

The general procedure for selecting the scattering coefficients a_n and b_n for a sphere consisting of many concentric shells is described by Kerker.[37] For the incident wave of Eqs. (1)—(4), the scattered field is generally given by (14) and (15) even though the refractive index varies *continuously* provided there is radial symmetry. The generalization of Eqs. (7) and (36) follows; however, the exact formulation of Q_{abs} may be impractical for the case of continuously varying refractive index. For elliptically-polarized illumination, Eq. (40) is again applicable and $\Gamma_x=\Gamma_y=0$.

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APPENDIX

Introduce the basis vectors $\hat{e}_0=\hat{z}$ and $\hat{e}_{\pm 1}=\mp 2^{-1/2}(\hat{x}\pm i\hat{y})$ and the vector spherical harmonic[14]

$$\vec{Y}^{\,l}_{n,m}(\theta,\phi)=\sum_{\mu=0,\pm 1}\langle l,1,m-\mu,\mu\,|\,l,1,n,m\rangle \hat{e}_\mu Y_{l,m-\mu}(\theta,\phi)\,. \tag{A1}$$

In the notation of Ref. 14, the Clebsch-Gordan coefficient[38] is $c_\mu(lnm)$ and $\vec{Y}^{\,l}_{n,m}$ is $\vec{T}^{\,l}_{nm}$; notice that $\vec{Y}^{\,n}_{n,m}=\vec{X}_{n,m}$. The following relation is given by identities involving the $\vec{Y}^{\,l}_{n,m}$ (Ref. 14, p. 270; Ref. 18; and Ref. 39):

$$\hat{r}\times\vec{X}_{n,m}=i(2n+1)^{-1/2}[n^{1/2}\vec{Y}^{\,n+1}_{n,m}+(n+1)^{1/2}\vec{Y}^{\,n-1}_{n,m}]\,. \tag{A2}$$

Tabulated Clebsch-Gordan coefficients[14,38] give the following results:

$$\hat{z}\cdot(\hat{r}\times\vec{X}_{n,1})=i(W_n^- Y_{n-1,1}-W_n^+ Y_{n+1,1})\,, \tag{A3}$$

where $W_n^\pm$ is defined in (25). Insertion of (A3) into $F^-_{n,n'}$ gives (24).

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$$\mathrm{Re}[\vec{\mathrm{B}}^{(E)*}(\vec{\mathrm{L}}\cdot\vec{\mathrm{B}}^{(E)})]=\mathrm{Re}[\vec{\mathrm{B}}^{(E)}(\vec{\mathrm{L}}\cdot\vec{\mathrm{B}}^{(E)})^*] \ .$$

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