Optical multipolar torque in structured electromagnetic fields: On helicity gradient torque, quadrupolar torque, and spin of the field gradient

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Structured light mechanically interacts with matter via optical forces and torques. Optical torque is traditionally calculated via the flux of total angular momentum (AM) into a volume enclosing an object. In the work published in [Phys. Rev. A **92**, 043843 (2015)], a powerful method was suggested to calculate the optical torque separately from the flux of the spin and the orbital parts of optical AM, rather than the total, providing useful physical insight. However, the method predicted a new type of dipolar torque dependent on the gradient of the helicity density of the optical beam, inconsistent with prior torque calculations. In this work, we first intend to clarify this discrepancy and clear up the confusion. We rederive, from first principles and with detailed derivations, both the traditional dipolar torque using total AM flux, and the spin and orbital torque components based on the corresponding AM contributions, ensuring that their sum agrees with the total torque. We also test our derived analytical expressions against numerical integration, with the exact agreement. We find that "helicity gradient" torque terms indeed exist in the spin and orbital components separately, but we present corrected prefactors, such that upon adding them, they cancel out, and the helicity gradient term vanishes from the total dipolar torque, reconciling literature results. In the second part of the work, we derive the analytical expression of the quadrupolar torque, showing that it is proportional to the spin of the EM field *gradient*, rather than the local EM field spin, as sometimes wrongly assumed in the literature. We provide examples of counter-intuitive situations where the spin of the EM field gradient behaves very differently from the local EM spin. Naively using the local EM field spin leads to wrong predictions of the torque on large particles with strong contributions of quadrupole and higher-order multipoles, especially in a structured incident field.

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I. INTRODUCTION

The mechanical interaction between a structured optical beam and structured photonic matter is a very important subject to study, in both fundamental and applied research. Such interaction is often complex, and simple analytical models like a multipole theory of optical force and torque in a general (inhomogeneous) electromagnetic field can greatly help our understanding of the physics involved.

Since the realization of orbital angular momentum in a paraxial laser beam (related to a helical phase) in 1992, there has been much confusion and debate on the separation of angular momentum into its spin and orbital parts [1,2]. In the strict sense, the spin and orbital parts of the angular momentum are not separately meaningful physical quantities though both of them have the unit of angular momentum. However, it is still possible to separate the total angular momentum into spin and orbital parts in a laboratory frame of reference such that both satisfy the proper continuity relations and are separately conserved quantities [1–3]. Both being conserved quantities, their net flow into a volume can be associated with a torque, it is Ref. [4] that first proposed the interesting concept of deriving the optical torque from the separate spin and orbital parts of angular momentum. The analytical expression given in Ref. [5] of optical torque acting on a dipolar particle takes into account complex spatial structures of the incident electromagnetic field. Such a treatment, if done properly, should give more physical insights on the separation of spin and orbital angular momentum involved in the interaction of structured light and objects [6,7].

One of the most interesting results of Ref. [5] is the theoretical prediction of a torque dependent on the gradient of helicity density. Studying early literature on optical torque since Ashkin's invention of optical tweezers [8,9], a few important works actually studied the optical torque on spherical particles exerted by an inhomogenous electromagnetic field based on generalized Mie theory [10–16]. In some of these early works [11–13], the helicity density gradient is generally nonzero in the incident optical beam. However, the dipolar components of optical torque in these early results [11–13] do not seem to contain the 'gradient' torque term as predicted in Ref. [5], thus pointing to a contradiction.

In this work, we intend to clarify this discrepancy and clear up the confusion. We rederive, from first principles and with detailed derivations, both the traditional dipolar total torque using total AM flux, and the spin and orbital torque components based on the corresponding AM contributions, ensuring that their sum agrees with the total torque. We also test our derived analytical expressions against numerical integration, with exact agreement. We find that "gradient" torque terms

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indeed exist in the spin and orbital components separately, but we present corrected prefactors, such that upon adding them, they cancel out, and the gradient term vanishes from the total torque, reconciling literature results. The concept proposed in Ref. [5] is still a very powerful tool to study the separate SAM and OAM contributions of optical torque, providing many advantages in understanding optical manipulations in the interacting structured light and photonic nanostructures.

In the second part of the work, we derive the analytical expression of the optical torque acting on an isotropic electromagnetic quadrupole in a general (inhomogeneous) electromagnetic field. With the analytical result, we show that the quadrupolar torque on an isotropic Mie particle is still proportional to the absorption cross section, confirming the transfer of the angular momentum to mechanical action through absorption. However, the relevant physical property of the incident beam that must be used to indicate the orientation of the optical torque is not the spin of the electromagnetic field, but the spin of the EM field gradient. Using the two-wave interference as an example, we show that there are significant differences between the spin of the electromagnetic field and the spin of the electromagnetic field gradient. As a result, simply using the local EM field spin can give rise to wrong predictions on the orientation of optical torque on large particles with strong quadrupole resonance. We show that an extraordinary transverse spin of the magnetic field gradient appears in a field formed by purely TM-polarized two wave interference, even though the magnetic field spin is zero. Furthermore, some of the nonintuitive "negative" torque that arises in the two-wave interference is often due to an incorrect interpretation of the physical properties related to the optical torque acting on a quadrupole and other higher-order multipoles.

II. OPTICAL TORQUE AND ANGULAR MOMENTUM

It is well known that electromagnetic fields can carry linear and angular momentum, and that angular momentum is a conserved vector quantity. Consequently, if an electromagnetic field shows a net flow of electromagnetic angular momentum constantly flowing into a volume containing a material object, we can conclude that the "missing" angular momentum is being transferred to the object via a mechanical torque.

The flux density of the electromagnetic angular momentum at every point in space is given by a tensor denoted as $\langle \overleftrightarrow{\mathbf{M}} \rangle$. This is a tensor because it contains information about the flow of electromagnetic angular momentum (itself a vector quantity) along each spatial direction. The time-averaged mechanical torque vector $\mathbf{\Gamma}$ can therefore be calculated as the total flux integral of the time-averaged angular momentum flux density $\langle \overleftrightarrow{\mathbf{M}} \rangle$ over a closed surface surrounding the object, as

$$\mathbf{\Gamma} = \oiint \langle \overleftrightarrow{\mathbf{M}} \rangle \cdot d\mathbb{S}, \tag{1}$$

In turn, the angular momentum flux is calculated as $\langle \overleftrightarrow{\mathbf{M}} \rangle = \mathbf{r} \times \langle \overleftrightarrow{\mathbf{T}} \rangle$ where $\langle \overleftrightarrow{\mathbf{T}} \rangle$ represents the time-averaged flux density of electromagnetic linear momentum and is referred to as Maxwell's stress tensor $\langle \overleftrightarrow{\mathbf{T}} \rangle$ of the total field:

$$\langle \overleftrightarrow{\mathbf{T}} \rangle = \frac{1}{2}\Re\left\{\varepsilon_0 \mathbf{E}_{\rm tot} \otimes \mathbf{E}^*_{\rm tot} + \mu_0 \mathbf{H}_{\rm tot} \otimes \mathbf{H}^*_{\rm tot} - \frac{\varepsilon_0 |\mathbf{E}_{\rm tot}|^2 + \mu_0 |\mathbf{H}_{\rm tot}|^2}{2} \overleftrightarrow{\mathbf{I}}\right\}, \tag{2}$$

where $\mathbf{r} = (\mathbf{r}' - \mathbf{r}_0)$, $\mathbf{r}_0$ is the location of the object, $\mathbf{r}'$ denotes a point on the surface $\mathbb{S}$, and $\mathbf{E}_{\rm tot} = \mathbf{E}_{\rm inc} + \mathbf{E}_{\rm sca}$ and $\mathbf{H}_{\rm tot} = \mathbf{H}_{\rm inc} + \mathbf{H}_{\rm sca}$ are the total electromagnetic fields.

In this report, we study the interaction between tiny objects and a time harmonic general (inhomogeneous) incident electromagnetic field. A time dependence of $e^{-i\omega t}$ is assumed. Since the integration of the angular momentum flux is done relative to the center of the object, the mechanical action on the object related to the optical torque corresponds to a rotation about the object's own center.

The time-averaged spin angular momentum flux $\langle \overleftrightarrow{\mathbf{M}}^s \rangle$ and orbital angular momentum flux $\langle \overleftrightarrow{\mathbf{M}}^o \rangle$, which together make up the total angular momentum flux density $\langle \overleftrightarrow{\mathbf{M}} \rangle = \langle \overleftrightarrow{\mathbf{M}}^s \rangle + \langle \overleftrightarrow{\mathbf{M}}^o \rangle$, can be separately written as [2,5]

$$\langle \overleftrightarrow{\mathbf{M}}^s \rangle = \frac{1}{2\omega}\Im\{\mathbf{E}_{\rm tot} \otimes \mathbf{H}^*_{\rm tot} + \mathbf{H}^*_{\rm tot} \otimes \mathbf{E}_{\rm tot} - (\mathbf{E}_{\rm tot} \cdot \mathbf{H}^*_{\rm tot}) \overleftrightarrow{\mathbf{I}}\}, \tag{3}$$

$$\langle \overleftrightarrow{\mathbf{M}}^o \rangle = \frac{1}{4\omega}\Im\{\mathbf{E}^*_{\rm tot} \otimes \mathbf{H}_{\rm tot} + \mathbf{H}_{\rm tot} \otimes \mathbf{E}^*_{\rm tot} + [(\mathbf{r} \times \nabla) \otimes \mathbf{E}^*_{\rm tot}] \times \mathbf{H}_{\rm tot} + [(\mathbf{r} \times \nabla) \otimes \mathbf{H}_{\rm tot}] \times \mathbf{E}^*_{\rm tot}\}, \tag{4}$$

The optical torque $\mathbf{\Gamma}^s$ attributed to the spin angular momentum flux can be calculated by

$$\mathbf{\Gamma}^s = \oiint \langle \overleftrightarrow{\mathbf{M}}^s \rangle \cdot d\mathbb{S}, \tag{5}$$

while the optical torque $\mathbf{\Gamma}^o$ attributed to the orbital angular momentum flux can be calculated by

$$\mathbf{\Gamma}^o = \oiint \langle \overleftrightarrow{\mathbf{M}}^o \rangle \cdot d\mathbb{S}, \tag{6}$$

such that they represent two physically distinct parts of the total torque $\mathbf{\Gamma} = \mathbf{\Gamma}^s + \mathbf{\Gamma}^o$. Like the optical torque using the total angular momentum flux, the torque $\mathbf{\Gamma}^o$ arising from the orbital angular momentum flux depends on a certain reference point as $\langle \overleftrightarrow{\mathbf{M}}^o \rangle$ has components that are dependent on the position vector $\mathbf{r}$. As before, this reference point is often considered at the object's own center when one only considers the optical torque resulting in self-rotation of the object. However, the optical torque $\mathbf{\Gamma}^s$ attributed to the spin angular momentum flux does not have this dependence and therefore this torque can be calculated for any choice of coordinate origin.

III. PREDICTION OF A 'GRADIENT' TORQUE

We first follow the formulism in Ref. [17] and write down the analytical expression (in SI units) of optical dipolar torque $\mathbf{\Gamma}_{\rm N}$ derived from the total angular momentum flux,

$$\boldsymbol{\Gamma}_{\mathrm{N}}=\frac{1}{2}\Re\{\mathbf{p}^{*}\times\mathbf{E}_{\mathrm{inc}}\}+\frac{1}{2}\Re\{\mathbf{m}^{*}\times\mu_0\mathbf{H}_{\mathrm{inc}}\}-\frac{k^3}{12\pi\varepsilon_0}\Im\{\mathbf{p}^{*}\times\mathbf{p}\}-\frac{k^3\mu_0}{12\pi}\Im\{\mathbf{m}^{*}\times\mathbf{m}\}+\frac{3}{4\omega}\Im\left\{\frac{1}{\varepsilon_0}(\mathbf{p}\cdot\nabla)\mathbf{H}^{*}_{\mathrm{inc}}-(\mathbf{m}\cdot\nabla)\mathbf{E}^{*}_{\mathrm{inc}}\right\}. \tag{7}$$

We then follow the formulism in Ref. [5] and separate the total optical torque $\boldsymbol{\Gamma}_{\mathrm{N}}$ into a SAM related torque $\boldsymbol{\Gamma}^{s}_{\mathrm{N}}$ and OAM related torque $\boldsymbol{\Gamma}^{o}_{\mathrm{N}}$. $\boldsymbol{\Gamma}^{s}_{\mathrm{N}}$ and $\boldsymbol{\Gamma}^{o}_{\mathrm{N}}$ are derived following Eqs. (5) and (6), respectively, from the spin angular momentum flux and the orbital angular momentum flux that satisfy separate conservation laws,

$$\begin{aligned}\boldsymbol{\Gamma}^{s}_{\mathrm{N}}=&\frac{1}{2}\Re\{\mathbf{p}^{*}\times\mathbf{E}_{\mathrm{inc}}\}+\frac{1}{2}\Re\{\mathbf{m}^{*}\times\mu_0\mathbf{H}_{\mathrm{inc}}\}-\frac{k^3}{24\pi\varepsilon_0}\Im\{\mathbf{p}^{*}\times\mathbf{p}\}-\frac{k^3\mu_0}{24\pi}\Im\{\mathbf{m}^{*}\times\mathbf{m}\}\\&+\frac{1}{2\omega}\Im\left\{\frac{1}{\varepsilon_0}(\mathbf{p}\cdot\nabla)\mathbf{H}^{*}_{\mathrm{inc}}-(\mathbf{m}\cdot\nabla)\mathbf{E}^{*}_{\mathrm{inc}}\right\},\end{aligned} \tag{8}$$

$$\boldsymbol{\Gamma}^{o}_{\mathrm{N}}=-\frac{k^3}{24\pi\varepsilon_0}\Im\{\mathbf{p}^{*}\times\mathbf{p}\}-\frac{k^3\mu_0}{24\pi}\Im\{\mathbf{m}^{*}\times\mathbf{m}\}+\frac{1}{4\omega}\Im\left[\frac{1}{\varepsilon_0}(\mathbf{p}\cdot\nabla)\mathbf{H}^{*}_{\mathrm{inc}}-(\mathbf{m}\cdot\nabla)\mathbf{E}^{*}_{\mathrm{inc}}\right]. \tag{9}$$

For induced dipoles in an isotropic Mie particle as described in Appendix A, the total optical torque $\boldsymbol{\Gamma}_{\mathrm{N}}$ can further be separated into an intrinsic part $\boldsymbol{\Gamma}^{\mathrm{int}}_{\mathrm{N}}$ that is closely related to the local spin density and an extrinsic part $\boldsymbol{\Gamma}^{\mathrm{ext}}_{\mathrm{N}}$ closely related to the dipole moments and the local field gradient.

$$\begin{aligned}\boldsymbol{\Gamma}^{\mathrm{int}}_{\mathrm{N}}=&\frac{1}{2}\Re\{\mathbf{p}^{*}\times\mathbf{E}_{\mathrm{inc}}\}-\frac{k^3}{12\pi\varepsilon_0}\Im\{\mathbf{p}^{*}\times\mathbf{p}\}+\frac{1}{2}\Re\{\mathbf{m}^{*}\times\mu_0\mathbf{H}_{\mathrm{inc}}\}-\frac{k^3\mu_0}{12\pi}\Im\{\mathbf{m}^{*}\times\mathbf{m}\}\\=&\frac{6\pi}{k^3}[\Re(a_1)-|a_1|^2]\mathbf{s}^{\mathrm{e}}+\frac{6\pi}{k^3}[\Re(b_1)-|b_1|^2]\mathbf{s}^{\mathrm{m}},\\\mathbf{s}^{\mathrm{e}}=&\frac{1}{2}\varepsilon_0\Im\{\mathbf{E}^{*}_{\mathrm{inc}}\times\mathbf{E}_{\mathrm{inc}}\},\\\mathbf{s}^{\mathrm{m}}=&\frac{1}{2}\mu_0\Im\{\mathbf{H}^{*}_{\mathrm{inc}}\times\mathbf{H}_{\mathrm{inc}}\},\end{aligned} \tag{10}$$

where a_1 is the Mie coefficient for an induced isotropic electric dipole and b_1 is the Mie coefficient for an induced isotropic magnetic dipole, whose relations with the induced dipolar polarizabilities are given in Appendix A.

The extrinsic torque $\boldsymbol{\Gamma}^{\mathrm{ext}}_{\mathrm{N}}$, as given in Ref. [5], can be reformulated using vector calculus identities,

$$\begin{aligned}\boldsymbol{\Gamma}^{\mathrm{ext}}_{\mathrm{N}}=&\frac{3}{4\omega}\Im\left\{\frac{1}{\varepsilon_0}(\mathbf{p}\cdot\nabla)\mathbf{H}^{*}_{\mathrm{inc}}-(\mathbf{m}\cdot\nabla)\mathbf{E}^{*}_{\mathrm{inc}}\right\}\\=&\frac{9\pi}{2k^4c_0}\Re\{a_1(\mathbf{E}_{\mathrm{inc}}\cdot\nabla)\mathbf{H}^{*}_{\mathrm{inc}}-b_1(\mathbf{H}_{\mathrm{inc}}\cdot\nabla)\mathbf{E}^{*}_{\mathrm{inc}}\}\\=&-\frac{9\pi}{2k^4c_0}\frac{\Re\{a_1\}+\Re\{b_1\}}{2}\nabla\times\Re\{\mathbf{E}_{\mathrm{inc}}\times\mathbf{H}^{*}_{\mathrm{inc}}\}-\frac{9\pi}{2k^4c_0}\frac{\Im\{a_1\}+\Im\{b_1\}}{2}\nabla\Im\{\mathbf{E}_{\mathrm{inc}}\cdot\mathbf{H}^{*}_{\mathrm{inc}}\}\\&+\frac{9\pi}{2k^4c_0}\frac{\Re\{a_1\}-\Re\{b_1\}}{2}\{\nabla\Re\{\mathbf{E}_{\mathrm{inc}}\cdot\mathbf{H}^{*}_{\mathrm{inc}}\}+\mu_0\Im\{\mathbf{H}^{*}_{\mathrm{inc}}\times\mathbf{H}_{\mathrm{inc}}\}-\varepsilon_0\Im\{\mathbf{E}^{*}_{\mathrm{inc}}\times\mathbf{E}_{\mathrm{inc}}\}\}\\&+\frac{9\pi}{2k^4c_0}\frac{\Im\{a_1\}-\Im\{b_1\}}{2}\nabla\times\Im\{\mathbf{E}_{\mathrm{inc}}\times\mathbf{H}^{*}_{\mathrm{inc}}\}.\end{aligned} \tag{11}$$

The total dipolar optical torque discussed in Ref. [5] should correspond to the mechanical action of a particle rotating about its own center. However, the extrinsic part in the total dipolar torque expression implies that under certain conditions, i.e., $[\Im\{a_1\}+\Im\{b_1\}]\neq 0$, the torque is dependent on the gradient of the helicity density of the incident beam $\nabla\Im\{\mathbf{E}_{\mathrm{inc}}\cdot\mathbf{H}^{*}_{\mathrm{inc}}\}$. Based on this, Ref. [5] predicted the existence of the 'gradient' torque. The gradient torque is said to exist as long as the incident beam has a nonzero gradient of helicity density $\nabla\Im\{\mathbf{E}_{\mathrm{inc}}\cdot\mathbf{H}^{*}_{\mathrm{inc}}\}$, and the nanoparticle has a nonzero imaginary part of the dipolar Mie coefficients $\Im\{a_1\}$ and $\Im\{b_1\}$. However, unlike a spinning torque that introduces a rotation around the particle's own center, or a revolution torque that introduces a rotation around a fixed reference point, it is difficult to interpret what mechanical action the gradient torque introduces on the dipolar particle.

In some of the early works [11–13] on optical torque, the helicity density gradient is generally nonzero in the incident optical beam. Therefore, according to the prediction of Ref. [5], there should exist a gradient torque in these early results. However, the dipolar components of the optical torque in [11–13] only show dependence on the absorption cross section of the dipole coefficients, i.e., $\Re\{a_1\} - |a_1|^2$ and $\Re\{b_1\} - |b_1|^2$. None of them has a dependence on $\Im\{a_1\}$ and $\Im\{b_1\}$ which are linked to the existence of a gradient torque. These early results seem to contradict the gradient torque prediction in Ref. [5]. Reference [18] also presented a multipolar theory of optical torque on an isotropic Mie particle in a general time-harmonic electromagnetic field. The analytical expression of dipolar torque in [18] is exactly the same as the intrinsic torque $\mathbf{\Gamma}_{\mathrm{N}}^{\mathrm{int}}$ in Ref. [5]. Just as the intrinsic torque $\mathbf{\Gamma}_{\mathrm{N}}^{\mathrm{int}}$, the dipolar torque derived in Ref. [18] depends only on the absorption cross section and the local spin density, agreeing with previous results [11–13]. However, the analytical expression in Ref. [18] does not include extrinsic torque $\mathbf{\Gamma}_{\mathrm{N}}^{\mathrm{ext}}$, and thus no torque that depends on the gradient of helicity density, again pointing to a contradiction.

In this work, we try to clarify this discrepancy and confusion on gradient torque. We rederive from first principles, with detailed calculations in Appendices, the analytical expressions of the dipolar torque both from the total angular momentum flux but also from the separate spin and orbital AM fluxes, ensuring that their sum exactly matches with the result obtained from the total AM flux. We also test our analytical expressions for the total torque against numerical integration of the angular flux density, finding exact agreement. Our results on total dipolar torque agree with Ref. [18], and contain only the intrinsic torque $\mathbf{\Gamma}_{\mathrm{N}}^{\mathrm{int}}$ but not the extrinsic torque $\mathbf{\Gamma}_{\mathrm{N}}^{\mathrm{ext}}$ presented in Ref. [5]. However, in our derivation of optical torques from the separate spin and orbital AM fluxes, we find that both the spin AM part and the orbital AM part of the dipolar torque contain components that are proportional to the extrinsic torque $\mathbf{\Gamma}_{\mathrm{N}}^{\mathrm{ext}}$. To be specific, we obtained the same analytical expression for SAM related torque as $\mathbf{\Gamma}_{\mathrm{N}}^{s}$ in Ref. [5]. However, we obtained a different coefficient for the extrinsic part of the OAM related torque compared to the expression $\mathbf{\Gamma}_{\mathrm{N}}^{o}$ given in Ref. [5]. In our derivation, the extrinsic parts in the SAM and OAM related torques exactly cancel each other out, and thus do not show in the total dipolar torque, which allows their sum to match the torque calculated via the more traditional conservation of total AM.

IV. ANALYTICAL EXPRESSION OF THE DIPOLAR OPTICAL TORQUE

In this section, we study the interaction between a time harmonic electromagnetic wave (described by its electric and magnetic fields $\mathbf{E}_{\mathrm{inc}}$ and $\mathbf{H}_{\mathrm{inc}}$) and a tiny particle that can be described by induced electromagnetic dipole moments. By definition, the torque can be calculated by integrating the total angular momentum flux $\langle \overleftrightarrow{\mathbf{M}} \rangle = \mathbf{r} \times \langle \overleftrightarrow{\mathbf{T}} \rangle$ over a closed surface centered at the origin,

$$\mathbf{\Gamma} = \oiint \langle \overleftrightarrow{\mathbf{M}} \rangle \cdot d\mathbb{S}. \tag{12}$$

Without loss of generality, we consider this enclosed surface to be spherical. The time-averaged torque on the Mie particle can then be expressed as

$$\begin{aligned} \mathbf{\Gamma} &= \oiint \langle \overleftrightarrow{\mathbf{M}} \rangle \cdot d\mathbb{S} \\ &= \int_0^{2\pi} \int_0^{\pi} \mathbf{r} \times (\langle \overleftrightarrow{\mathbf{T}} \rangle \cdot \hat{\mathbf{n}}) r^2 \sin\theta d\theta d\phi, \\ &= \Re \int_0^{2\pi} \int_0^{\pi} (r\hat{\mathbf{n}}) \times \left\{ \frac{\varepsilon_0}{2} \mathbf{E}_{\mathrm{tot}} (\mathbf{E}_{\mathrm{tot}}^* \cdot \hat{\mathbf{n}}) + \frac{\mu_0}{2} \mathbf{H}_{\mathrm{tot}} (\mathbf{H}_{\mathrm{tot}}^* \cdot \hat{\mathbf{n}}) \right\} \\ &\quad \times r^2 \sin\theta d\theta d\phi, \end{aligned} \tag{13}$$

where r is the radius of the spherical surface and $\hat{\mathbf{n}}$ is the outward radial unit vector normal to the surface.

Given the induced electric dipole moment $\mathbf{p}$, the corresponding radiation field $\mathbf{E}_{\mathrm{p}}$ and $\mathbf{H}_{\mathrm{p}}$ can be analytically calculated, as expressed in Appendix B. Knowing the total electromagnetic field $\mathbf{E}_{\mathrm{tot}} = \mathbf{E}_{\mathrm{inc}} + \mathbf{E}_{\mathrm{p}}$ and $\mathbf{H}_{\mathrm{tot}} = \mathbf{H}_{\mathrm{inc}} + \mathbf{H}_{\mathrm{p}}$, the analytical expression of optical torque on an induced dipole by a general time-hamonic electromagnetic field can be derived, as detailed in Appendix C based on the angular momentum flux of the total field.

The dipolar torque $\mathbf{\Gamma}_{\mathrm{p}}$, attributed to the interaction between induced electric dipole and incident field, can be decomposed into different parts,

$$\mathbf{\Gamma}_{\mathrm{p}} = \mathbf{\Gamma}_{\mathrm{inc}} + \mathbf{\Gamma}_{\mathrm{p,mix}} + \mathbf{\Gamma}_{\mathrm{p,recoil}}. \tag{14}$$

From the derivation in Appendix C, the torque component purely dependent on the incident field is

$$\mathbf{\Gamma}_{\mathrm{inc}} = 0, \tag{15}$$

the extinction torque component $\mathbf{\Gamma}_{\mathrm{p,mix}}$, dependent on the interference between incident and radiation fields, is analytically given

$$\mathbf{\Gamma}_{\mathrm{p,mix}} = \frac{1}{2} \Re\{\mathbf{p}^* \times \mathbf{E}_{\mathrm{inc}}\}, \tag{16}$$

and the recoil torque $\mathbf{\Gamma}_{\mathrm{p,recoil}}$, as a result of self-interaction of the induced electric dipole, is

$$\mathbf{\Gamma}_{\mathrm{p,recoil}} = -\frac{k^3}{12\pi\varepsilon_0} \Im\{\mathbf{p}^* \times \mathbf{p}\}. \tag{17}$$

The total optical torque for an induced electric dipole in a general optical field can therefore be written as

$$\mathbf{\Gamma}_{\mathrm{p}} = \frac{1}{2} \Re\{\mathbf{p}^* \times \mathbf{E}_{\mathrm{inc}}\} - \frac{k^3}{12\pi\varepsilon_0} \Im\{\mathbf{p}^* \times \mathbf{p}\}, \tag{18}$$

In the special case of an isotropic electric dipole as described in Appendix A, one can easily relate the induced electric dipole $\mathbf{p}$ to the incident electric field $\mathbf{E}_{\mathrm{inc}}$, via the Mie polarizabilities, and therefore the optical torque can be written as a function of polarizabilities and incident fields only, as

$$\begin{aligned} \mathbf{\Gamma}_{\mathrm{p}} &= \frac{6\pi}{k^3} [\Re(a_1) - |a_1|^2] \mathbf{s}^{\mathrm{e}}, \\ \mathbf{s}^{\mathrm{e}} &= \frac{\varepsilon_0}{2} \Im\{\mathbf{E}_{\mathrm{inc}}^* \times \mathbf{E}_{\mathrm{inc}}\}, \end{aligned} \tag{19}$$

which indicates that the optical torque for an isotropic electric dipole is proportional to the absorption cross section and the local spin density of the incident electric field.

The analytical expression of the optical torque for a magnetic dipole can be derived in a similar manner,

$$\mathbf{\Gamma}_{\rm m} = \frac{1}{2}\Re\{\mathbf{m}^* \times \mu_0\mathbf{H}_{\rm inc}\} - \frac{k^3\mu_0}{12\pi}\Im\{\mathbf{m}^* \times \mathbf{m}\}, \tag{20}$$

and in the case of an isotropic magnetic dipole,

$$\begin{aligned}\mathbf{\Gamma}_{\rm m} &=\frac{6\pi}{k^3}[\Re(b_1) - |b_1|^2]\mathbf{s}^{\rm m},\\ \mathbf{s}^{\rm m} &=\frac{\mu_0}{2}\Im\{\mathbf{H}^*_{\rm inc} \times \mathbf{H}_{\rm inc}\}.\end{aligned} \tag{21}$$

As we can see from the above expressions, the total optical torque on isotropic electric and magnetic dipoles derived from the conservation of total AM does not contain the extrinsic torque terms as in Refs. [5,17]. The transfer from spin to torque is purely through absorption, and the only relevant property of the incident field is its local spin angular momentum even in the case of a complex incident optical field.

V. DIPOLAR TORQUE AND CONSERVATION LAWS FOR SPIN AND ORBITAL PARTS OF THE ANGULAR MOMENTUM

In this section, we follow the optical torque calculation devised from the separate conservation laws for the spin and orbital parts of angular momentum, first outlined in Refs. [2,4], and we will see that after a careful derivation, their sum will equal the total torque derived in the previous section.

The dipolar optical torque attributed to the spin angular momentum flux $\mathbf{\Gamma}^s$ and the torque attributed to the orbital angular momentum flux $\mathbf{\Gamma}^o$ can be calculated by integrating the corresponding angular momentum flux as shown Eqs. (5) and (6). Based on this method, the spin and orbital torques of a general electromagnetic field acting on an induced electric dipole can be analytically derived as detailed in Appendix D,

$$\begin{aligned}\mathbf{\Gamma}^s_{\rm p,mix} &= \frac{1}{2}\Re\{\mathbf{p}^* \times \mathbf{E}_{\rm inc}\} + \frac{1}{2\omega\varepsilon_0}\Im[(\mathbf{p}\cdot\nabla)\mathbf{H}^*_{\rm inc}],\\ \mathbf{\Gamma}^o_{\rm p,mix} &= -\frac{1}{2\omega\varepsilon_0}\Im[(\mathbf{p}\cdot\nabla)\mathbf{H}^*_{\rm inc}],\\ \mathbf{\Gamma}^s_{\rm p,recoil} &= -\frac{k^3}{24\pi\varepsilon_0}\Im(\mathbf{p}^* \times \mathbf{p}),\\ \mathbf{\Gamma}^o_{\rm p,recoil} &= -\frac{k^3}{24\pi\varepsilon_0}\Im(\mathbf{p}^* \times \mathbf{p}).\end{aligned} \tag{22}$$

Similarly, the "spin" and "orbital" optical torque of a general electromagnetic field acting on an induced magnetic dipole can be analytically derived as,

$$\begin{aligned}\mathbf{\Gamma}^s_{\rm m,mix} &= \frac{\mu_0}{2}\Re\{\mathbf{m}^* \times \mathbf{H}_{\rm inc}\} - \frac{1}{2\omega}\Im\{(\mathbf{m}\cdot\nabla)\mathbf{E}^*_{\rm inc}\},\\ \mathbf{\Gamma}^o_{\rm m,mix} &= +\frac{1}{2\omega}\Im\{(\mathbf{m}\cdot\nabla)\mathbf{E}^*_{\rm inc}\},\\ \mathbf{\Gamma}^s_{\rm m,recoil} &= -\frac{k^3\mu_0}{24\pi}\Im\{\mathbf{m}^* \times \mathbf{m}\},\\ \mathbf{\Gamma}^o_{\rm m,recoil} &= -\frac{k^3\mu_0}{24\pi}\Im\{\mathbf{m}^* \times \mathbf{m}\}.\end{aligned} \tag{23}$$

By adding up both the spin and orbital angular momentum flux contributions, the optical torque on an electromagnetic dipole, described by an electric dipole moment $\mathbf{p}$ and a magnetic dipole moment $\mathbf{m}$, is given as

$$\begin{aligned}\mathbf{\Gamma}_{\rm p,mix} &= \mathbf{\Gamma}^s_{\rm p,mix} + \mathbf{\Gamma}^o_{\rm p,mix} = \frac{1}{2}\Re\{\mathbf{p}^* \times \mathbf{E}_{\rm inc}\},\\ \mathbf{\Gamma}_{\rm p,recoil} &= \mathbf{\Gamma}^s_{\rm p,recoil} + \mathbf{\Gamma}^o_{\rm p,recoil} = -\frac{k^3}{12\pi\varepsilon_0}\Im\{\mathbf{p}^* \times \mathbf{p}\},\\ \mathbf{\Gamma}_{\rm m,mix} &= \mathbf{\Gamma}^s_{\rm m,mix} + \mathbf{\Gamma}^o_{\rm m,mix} = \frac{\mu_0}{2}\Re\{\mathbf{m}^* \times \mathbf{H}_{\rm inc}\},\\ \mathbf{\Gamma}_{\rm m,recoil} &= \mathbf{\Gamma}^s_{\rm m,recoil} + \mathbf{\Gamma}^o_{\rm m,recoil} = -\frac{k^3\mu_0}{12\pi}\Im\{\mathbf{m}^* \times \mathbf{m}\}.\end{aligned} \tag{24}$$

The resulting total dipolar torque

$$\begin{aligned}\mathbf{\Gamma}_d =&\mathbf{\Gamma}_{\rm p,mix} + \mathbf{\Gamma}_{\rm p,recoil} + \mathbf{\Gamma}_{\rm m,mix} + \mathbf{\Gamma}_{\rm m,recoil}\\ =&\frac{1}{2}\Re\{\mathbf{p}^* \times \mathbf{E}_{\rm inc}\} - \frac{k^3}{12\pi\varepsilon_0}\Im\{\mathbf{p}^* \times \mathbf{p}\} + \frac{\mu_0}{2}\Re\{\mathbf{m}^* \times \mathbf{H}_{\rm inc}\}\\ &- \frac{k^3\mu_0}{12\pi}\Im\{\mathbf{m}^* \times \mathbf{m}\}\end{aligned} \tag{25}$$

agrees with the analytical expressions of dipolar torques devised from total angular momentum flux. The "orbital" optical torque on a source of combined electric and magnetic dipole has the same structure as the extrinsic torque $\mathbf{\Gamma}^{\rm ext}_{\rm N}$ in Ref. [5], apart from a different coefficient. However, as it can be seen, the difference means that this "extrinsic" type of torque, which includes the "gradient torque," is exactly canceled when adding the spin and orbital angular momentum contributions together.

VI. ON THE (NON) EXISTENCE OF "GRADIENT" TORQUE

As discussed previously, Ref. [5] predicts the existence of a gradient torque, i.e. an optical torque arises from a nonzero gradient of helicity density and is depedent on the imaginary part of the dipolar Mie coefficients $\Im(a_1)$ and $\Im(b_1)$. The existence of a gradient torque challenges our understanding of the nature of optical torque. Conventionally, we know that optical torque introduces a mechanical action on an object being either rotation around its own center or revolution around a fixed reference point. However, it is difficult to interpret what mechanical action the gradient torque introduces on the object. We try to examine this with an electromagnetic field designed such that there are no torques except for the gradient torque, if it exists at all.

We consider the special case of an electromagnetic field built up by the coherent interference of multiple N circularly polarized plane waves with constant radial wave vector $k_\rho = k$ and evenly distributed over the full 2π azimuthal directions in the $z = 0$ plane. Each plane wave has an electric field distribution $\mathbf{E}_v(\phi_v) = (\hat{\mathbf{e}}_p + i\hat{\mathbf{e}}_s)\frac{E_0}{N}\exp\{i(k\hat{\mathbf{e}}_v)\cdot(\rho\hat{\mathbf{e}}_\rho)\}$, where $\hat{\mathbf{e}}_s(\phi_v) = -\hat{\mathbf{e}}_{\phi_v} = \sin\phi_v\hat{\mathbf{e}}_x - \cos\phi_v\hat{\mathbf{e}}_y$ and $\hat{\mathbf{e}}_p(\phi_v) = -\hat{\mathbf{e}}_z$ are the unit vectors for transverse magnetic (TM or p-) and transverse electric (TE or s-) polarization, and $\hat{\mathbf{e}}_v = -\cos\phi_v\hat{\mathbf{e}}_x - \sin\phi_v\hat{\mathbf{e}}_y$. In the limit of infinitely many beams as $N \to \infty$, the electromagnetic field built up can be analytically calculated as

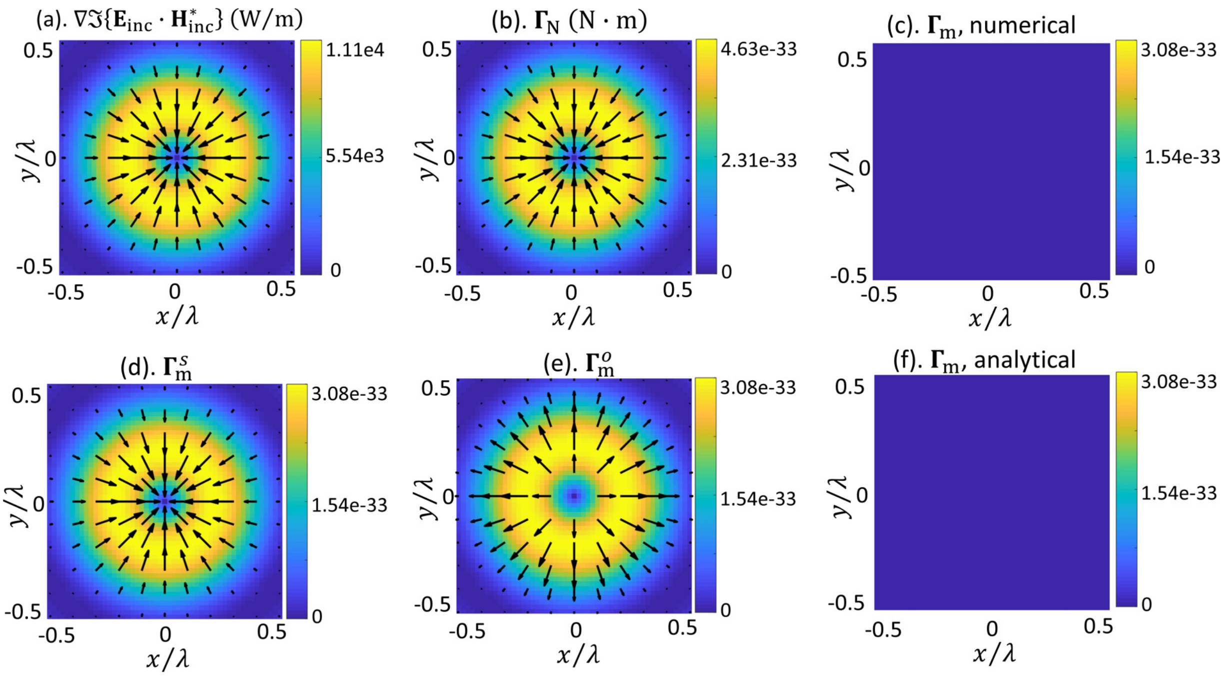

FIG. 1. (a) Gradient of helicity density of the designed optical field. (b) Total optical torque (unit N m) on a dipolar particle with Mie coefficient $b_1 = 0.969 - 0.173i$ in the designed optical field ($E_0 = 1$ V/m), based on the analytical results in Refs. [5,17]. (c) Optical torque on the same particle calculated based on the numerical integration of total angular momentum flux; Analytical results of the optical torque attributed to (d) the spin AM flux, (e) the orbital AM flux, and (f) the total AM flux.

$$\begin{aligned}\mathbf{E}_{\text{inc}}(\rho,\phi,z) &= \frac{1}{2\pi}\int_0^{2\pi}[\hat{\mathbf{e}}_p(\phi_v) + i\hat{\mathbf{e}}_s(\phi_v)]E_0\exp\{ik\rho(\hat{\mathbf{e}}_v\cdot\hat{\mathbf{e}}_\rho)\}d\phi_v\\ &= \hat{\mathbf{e}}_x E_0 J_1(k\rho)\sin\phi - \hat{\mathbf{e}}_y E_0 J_1(k\rho)\cos\phi - \hat{\mathbf{e}}_z E_0 J_0(k\rho),\\ \mathbf{H}_{\text{inc}}(\rho,\phi,z) &= -\hat{\mathbf{e}}_x i\frac{E_0}{Z_0}J_1(k\rho)\sin\phi + \hat{\mathbf{e}}_y i\frac{E_0}{Z_0}J_1(k\rho)\cos\phi + \hat{\mathbf{e}}_z i\frac{E_0}{Z_0}J_0(k\rho),\end{aligned} \tag{26}$$

where $Z_0 = \sqrt{\mu_0/\varepsilon_0}$ is the impedance of free space.

This electromagnetic field has the following properties:

$$\begin{aligned}&\mathbf{E}_{\text{inc}}\times\mathbf{H}^*_{\text{inc}} = 0,\\ &\Im\{\mathbf{E}^*_{\text{inc}}\times\mathbf{E}_{\text{inc}}\} = 0,\ \ \Im\{\mathbf{H}^*_{\text{inc}}\times\mathbf{H}_{\text{inc}}\} = 0,\\ &\mathbf{E}_{\text{inc}}\cdot\mathbf{H}^*_{\text{inc}} = i\frac{E_0^2}{Z_0}\big[J_1^2(k\rho) + J_0^2(k\rho)\big].\end{aligned} \tag{27}$$

Based on the analytical results in Ref. [5] as given in Eq. (11), placing an isotropic magnetic dipole in the designed optical field would introduce an optical torque arising only from the gradient torque:

$$\begin{aligned}\mathbf{\Gamma}_{\text{N}} &= \mathbf{\Gamma}^s_{\text{N}} + \mathbf{\Gamma}^o_{\text{N}}\\ &= \mathbf{\Gamma}^{\text{ext}}_{\text{N}}\\ &= -\frac{3}{4\omega}\Im[(\mathbf{m}\cdot\nabla)\mathbf{E}^*_{\text{inc}}]\\ &= -\frac{9\pi}{4k^4c_0}\Im(b_1)\nabla\Im\{\mathbf{E}_{\text{inc}}\cdot\mathbf{H}^*_{\text{inc}}\}.\end{aligned} \tag{28}$$

Figure 1(b) shows the optical torque distribution based on this analytical result where a dipolar particle with $b_1 = 0.969 - 0.173i$ is placed in the $z = 0$ plane of the designed beam. Since $\Im\{\mathbf{H}^*_{\text{inc}}\times\mathbf{H}_{\text{inc}}\} = 0$, the instrinsic parts of both the mixed and recoil dipolar torque are zero. The optical torque shown in Fig. 1(b), calculated according to the expressions on Ref. [5], has only an extrinsic part and depends on $\Im(b_1)$ and on the gradient of helicity density $\Im\{\mathbf{E}_{\text{inc}}\cdot\mathbf{H}^*_{\text{inc}}\}$ as shown in Fig. 1(a). As a result, the analytical results of Ref. [5] give rise to a radially oriented torque across the incident beam in the $z = 0$ plane.

In order to test the existence of the gradient torque, we first calculate the optical torque numerically by integrating the total angular momentum flux $\langle \overleftrightarrow{\mathbf{M}} \rangle = \mathbf{r}\times\langle \overleftrightarrow{\mathbf{T}} \rangle$ over an enclosed surface surrounding the particle. The numerically calculated optical torque for the same dipole in the designed beam is shown in Fig. 1(c), whose values are close to zero (down to numerical errors) across the beam. On the other hand, Figs. 1(d)–1(f) show the torque distributions attributed to the spin, orbital and total AM fluxes based on our analytical results given in Secs. V and IV. The numerically calculated torque based on total angular momentum flux in Fig. 1(c) does not show the radially oriented gradient torque across the incident beam as predicted by by Ref. [5]. Instead, it agrees well with the intrinsic torque in Fig. 1(f) which is null and dependent on local spin density $\frac{\mu_0}{2}\Im\{\mathbf{H}^*\times\mathbf{H}\}=0$. This gradient torque structure does show up in the spin and orbital parts of the torque as in Figs. 1(d)–1(f). However, as discussed previously, the spin and orbital parts of the torque exactly cancel each other, leading to a null total optical torque.

VII. ANALYTICAL EXPRESSION OF THE ISOTROPIC QUADRUPOLAR OPTICAL TORQUE

Given the electric quadrupole moment $\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}}$, the corresponding radiation field $\mathbf{E}_{\mathrm{Qe}}$ and $\mathbf{H}_{\mathrm{Qe}}$ can be analytically calculated, as expressed in Appendix B. Knowing the total electromagnetic field $\mathbf{E}_{\mathrm{tot}}=\mathbf{E}_{\mathrm{inc}}+\mathbf{E}_{\mathrm{Qe}}$ and $\mathbf{H}_{\mathrm{tot}}=\mathbf{H}_{\mathrm{inc}}+\mathbf{H}_{\mathrm{Qe}}$, the analytical expression of optical torque on an induced quadrupole by a general time-hamonic electromagnetic field can be derived, as detailed in Appendix E based on the angular momentum flux of the total field.

The quadrupolar torque $\mathbf{\Gamma}_{\mathrm{Qe}}$ and $\mathbf{\Gamma}_{\mathrm{Qm}}$, attributed to the interaction between an induced electric and magnetic quadrupole in an isotropic Mie particle and a general electromagnetic field, can be analytically expressed as

$$\begin{aligned}\mathbf{\Gamma}_{\mathrm{Qe}}&=\frac{120\pi}{k^5}[\Re\{a_2\}-|a_2|^2]\mathbf{s}^{\mathrm{Qe}},\\ \mathbf{\Gamma}_{\mathrm{Qm}}&=\frac{120\pi}{k^5}[\Re\{b_2\}-|b_2|^2]\mathbf{s}^{\mathrm{Qm}},\end{aligned}\tag{29}$$

where

$$\begin{aligned}\mathbf{s}^{\mathrm{Qe}}&=\sum_{u=x,y,z}\frac{\varepsilon_0}{6}\Im\{\mathbf{D}_u^{\mathrm{e}*}\times\mathbf{D}_u^{\mathrm{e}}\},\\ \mathbf{s}^{\mathrm{Qm}}&=\sum_{u=x,y,z}\frac{\mu_0}{6}\Im\{\mathbf{D}_u^{m*}\times\mathbf{D}_u^{\mathrm{m}}\},\\ \mathbf{D}_u^{\mathrm{e}}&=\sum_{v=x,y,z}\hat{\mathbf{e}}_v[\overleftrightarrow{\mathcal{D}}^{\mathrm{e}}]_{uv},\quad \mathbf{D}_u^{\mathrm{m}}=\sum_{v=x,y,z}\hat{\mathbf{e}}_v[\overleftrightarrow{\mathcal{D}}^{\mathrm{m}}]_{uv},\\ [\overleftrightarrow{\mathcal{D}}^{\mathrm{e}}]_{uv}&=\frac{\partial_uE_v+\partial_vE_u}{2},\quad [\overleftrightarrow{\mathcal{D}}^{\mathrm{m}}]_{uv}=\frac{\partial_uH_v+\partial_vH_u}{2}.\end{aligned}\tag{30}$$

In previous literature, the local EM field spin densities $\mathbf{s}^{\mathrm{e}}$ and $\mathbf{s}^{\mathrm{m}}$ are often used to indicate the orientation of the optical torque [5,17,19,20]. Just like the dipolar case, the optical torque acting on a quadrupole is proportional to the corresponding quadrupolar absorption cross sections $\Re\{a_2\}-|a_2|^2$ and $\Re\{b_2\}-|b_2|^2$. However, the relevant physical properties of the incident beam is not the spin densities of the EM field $\mathbf{s}^{\mathrm{e}}$ and $\mathbf{s}^{\mathrm{m}}$, but rather the spin densities of the EM field gradient $\mathbf{s}^{\mathrm{Qe}}$ and $\mathbf{s}^{\mathrm{Qm}}$. As a result, simply using the local EM field spin densities can give rise to wrong predictions on the orientation and magnitude of optical torque.

The EM field spin vectors $\{\mathbf{s}^{\mathrm{e}},\mathbf{s}^{\mathrm{m}}\}$ and the EM field gradient spin vectors $\{\mathbf{s}^{\mathrm{Qe}},\mathbf{s}^{\mathrm{Qm}}\}$ can, perhaps unintuitively, show very different behaviours, including having opposite orientations. This can be shown through examples. In the remaining of this section, we will illustrate the differences in the very simple case of two-wave interference [21], but differences will appear in any general structured EM field.

The EM field, built up by two free-propagating plane waves along the wave vectors $\mathbf{k}_1=k_x\hat{\mathbf{e}}_x+k_z\hat{\mathbf{e}}_z$ and $\mathbf{k}_2=-k_x\hat{\mathbf{e}}_x+k_z\hat{\mathbf{e}}_z$, can be described in the transverse magnetic ($p-$) and transverse electric ($s-$) polarization basis,

$$\begin{aligned}\mathbf{E}_1&=\left(E_1^p\hat{\mathbf{e}}_1^p+E_1^s\hat{\mathbf{e}}_1^s\right)\exp(ik_xx+ik_zz),\\ \mathbf{E}_2&=\left(E_2^p\hat{\mathbf{e}}_2^p+E_2^s\hat{\mathbf{e}}_2^s\right)\exp(i\varphi_0-ik_xx+ik_zz),\\ \mathbf{H}_1&=\frac{\mathbf{k}_1}{Z_0k}\times\mathbf{E}_1,\quad \mathbf{H}_2=\frac{\mathbf{k}_2}{Z_0k}\times\mathbf{E}_2,\end{aligned}\tag{31}$$

where

$$\hat{\mathbf{e}}_1^s=\hat{\mathbf{e}}_y,\quad \hat{\mathbf{e}}_2^s=-\hat{\mathbf{e}}_y,\quad \hat{\mathbf{e}}_1^p=\hat{\mathbf{e}}_1^s\times\frac{\mathbf{k}_1}{k},\quad \hat{\mathbf{e}}_2^p=\hat{\mathbf{e}}_2^s\times\frac{\mathbf{k}_2}{k},$$

Here we focus on the field properties that are related to the optical torque in dipoles and quadrupoles. The field spin $\mathbf{s}^{\mathrm{m}}$ [as given in Eq. (21)] and the field gradient spin $\mathbf{s}^{\mathrm{Qm}}$ [as given in Eq. (30)] are evaluated on the $z=0$ plane:

$$\begin{aligned}s_x^{\mathrm{m}}&=\frac{k_x\varepsilon_0}{k}\Im\{E_1^{p*}E_1^s-E_2^{p*}E_2^s\}+\frac{k_x\varepsilon_0}{k}\Im\{\left(E_1^{p*}E_2^s+E_1^{s*}E_2^p\right)\exp(i\varphi_0-2ik_xx)\},\\ s_y^{\mathrm{m}}&=\frac{2k_xk_z\varepsilon_0}{k^2}\Im\{E_1^{s*}E_2^s\exp(i\varphi_0-2ik_xx)\},\\ s_z^{\mathrm{m}}&=\frac{k_z\varepsilon_0}{k}\Im\{E_1^{p*}E_1^s+E_2^{p*}E_2^s\}+\frac{k_z\varepsilon_0}{k}\Im\{\left(E_1^{s*}E_2^p-E_1^{p*}E_2^s\right)\exp(i\varphi_0-2ik_xx)\},\\ s_x^{\mathrm{Qm}}&=\frac{k_xk\varepsilon_0}{12}\Im\{E_1^{p*}E_1^s-E_2^{p*}E_2^s\}+\frac{k_x\left(3k_z^2-k_x^2\right)\varepsilon_0}{12k}\Im\{\left(E_1^{p*}E_2^s+E_1^{s*}E_2^p\right)\exp(i\varphi_0-2ik_xx)\},\\ s_y^{\mathrm{Qm}}&=\frac{2k_xk_z\left(k_z^2-k_x^2\right)\varepsilon_0}{3k^2}\Im\{E_1^{s*}E_2^s\exp(i\varphi_0-2ik_xx)\}+\frac{k_xk_z\varepsilon_0}{6}\Im\{E_1^{p*}E_2^p\exp(i\varphi_0-2ik_xx)\},\\ s_z^{\mathrm{Qm}}&=\frac{k_zk\varepsilon_0}{12}\Im\{E_1^{p*}E_1^s+E_2^{p*}E_2^s\}+\frac{k_z\left(k_z^2-3k_x^2\right)\varepsilon_0}{12k}\Im\{\left(E_1^{s*}E_2^p-E_1^{p*}E_2^s\right)\exp(i\varphi_0-2ik_xx)\}.\end{aligned}\tag{32}$$

For an induced magnetic dipole with Mie coefficient b_1 in such a field, an optical torque $\mathbf{\Gamma}_{\mathrm{m}}=\frac{6\pi}{k^3}[\Re(b_1)-|b_1|^2]\mathbf{s}^{\mathrm{m}}$ will be exerted on the Mie particle. For an induced magnetic quadrupole with Mie coefficient b_2 in such a field, an optical

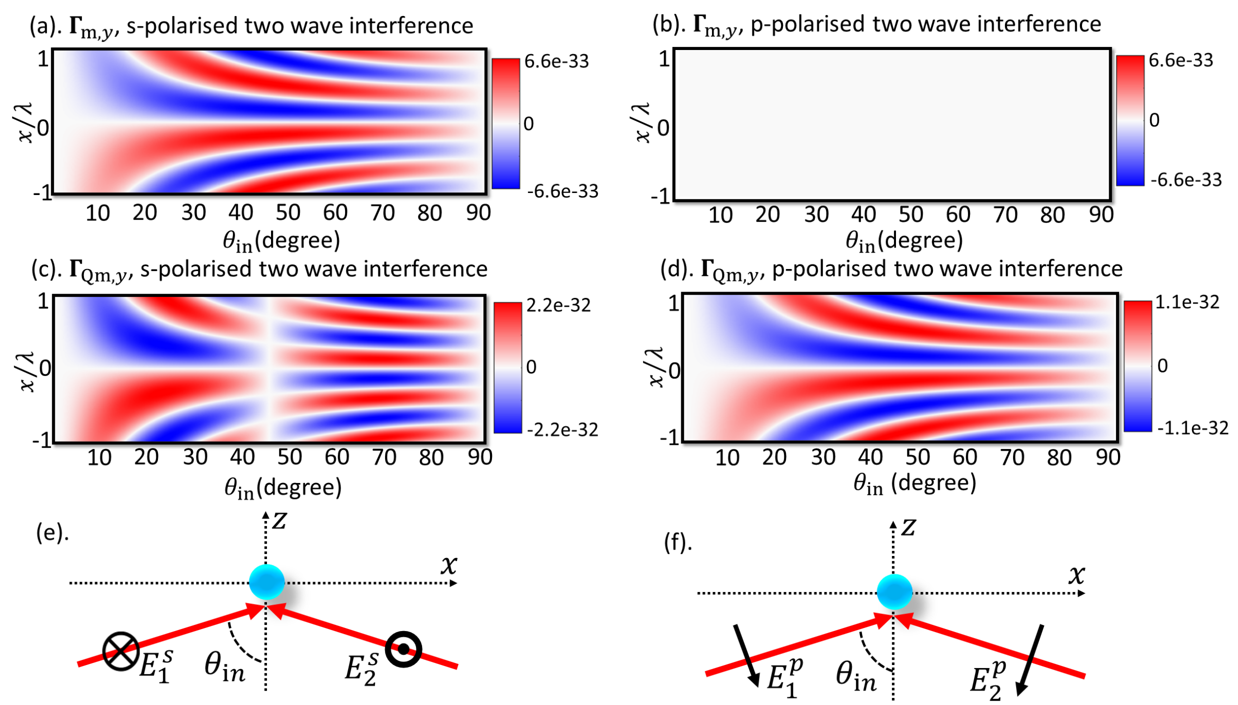

FIG. 2. Optical torques on an induced magnetic dipole and magnetic quadrupole (with equal Mie coeffcients $b_1 = b_2 = 0.802 - 0.047i$) in an electromagnetic field formed by two-wave interference, at varying x and incident angles θ_{in}. The y component of the optical torque $\Gamma_{\text{m},y}$ (N m) on a pure magnetic dipole in the EM field formed by (a) two s-polarized plane waves ($E_1^p = E_2^p = 1$ V/m) and (b) two p-polarized plane waves ($E_1^s = E_2^s = 1$ V/m); The y component of the optical torque on a pure magnetic quadrupole $\Gamma_{\text{Qm},y}$ in the EM field formed by (c) two s-polarized plane waves and (d) two p-polarized plane waves; Illustrations of two-wave interference by (e) purely s-polarized plane waves and (f) purely p-polarized plane waves.

torque $\mathbf{\Gamma}_{\text{Qm}} = \frac{120\pi}{k^5}[\Re\{b_2\} - |b_2|^2]\mathbf{s}^{\text{Qm}}$ is exerted on the Mie particle.

In Figs. 2 and 3, we show a magnetic dipole and a magnetic quadrupole with equal Mie coefficients $b_1 = b_2 = 0.802 - 0.047i$ in three types of EM field built up by two-wave interference: purely p-polarized plane waves ($E_1^p = E_2^p = 1$ V/m, corresponding to an intensity of 1.3×10^{-3} W/m^2 in a single beam), purely s-polarized plane waves ($E_1^s = E_2^s = 1$ V/m), and circularly polarized plane waves with the same helicity ($E_1^p = E_2^p = 1$ V/m, $E_1^s = E_2^s = i$ V/m).

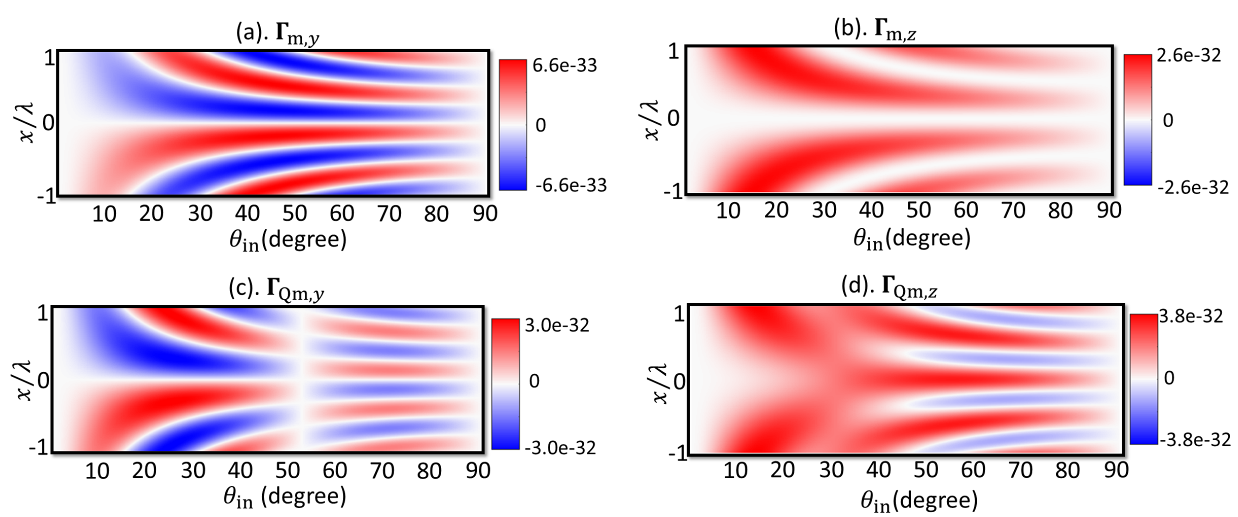

FIG. 3. Nonzero components of optical torque on an induced magnetic dipole and magnetic quadrupole (with equal Mie coeffcients $b_1 = b_2 = 0.802 - 0.047i$), at varying x and incident angles θ_{in}, in an electromagnetic field formed by two circularly polarized plane waves ($E_1^p = E_2^p = 1$ V/m, $E_1^s = E_2^s = i$ V/m). (a) y component of the optical torque $\Gamma_{\text{m},y}$ on a pure magnetic dipole; (b) z component of the optical torque $\Gamma_{\text{m},z}$ on a pure magnetic dipole; (c) y component of the optical torque $\Gamma_{\text{Qm},y}$ on a pure magnetic quadrupole; (d) z component of the optical torque $\Gamma_{\text{Qm},z}$ on a pure magnetic quadrupole.

The differences between the field spin vector $\mathbf{s}^{\mathrm{m}}$ and the field gradient spin vector $\mathbf{s}^{\mathrm{Qm}}$ are shown in Eq. (32) and clearly illustrated in Fig. 2. From Eq. (32), we can clearly see that the electromagnetic field, built up by two purely TE (s-) polarized plane waves, introduces both a transverse spin in the magnetic field $\mathbf{s}_y^{\mathrm{m}}$ and a transverse spin in the magnetic field gradient $\mathbf{s}_y^{\mathrm{Qm}}$. As can be seen from Eq. (32), both the transverse spins of the magnetic field $\mathbf{s}_y^{\mathrm{m}}$ and magnetic field gradient $\mathbf{s}_y^{\mathrm{Qm}}$ are generated via the interference effect $\Im\{E_1^{s*}E_2^s\exp(i\varphi_0 - 2ik_x x)\}$. However, the transverse spins of the magnetic field and field gradient are strongly dependent on how the interfering electromagnetic field is structured. In the two-wave interference cases, the structure of the resulting electromagnetic field is dependent on the incidence angle θ_{in} (following the relations that $k_x = k\sin\theta_{\mathrm{in}}$ and $k_z = k\cos\theta_{\mathrm{in}}$). We can see from Eq. (32) that the transverse spin of the magnetic field has a dependence of $k_x k_z$, while the transverse spin of the magnetic field gradient has a dependence of $k_x k_z(k_z^2 - k_x^2)$. $k_x k_z$ does not change sign with incidence angle θ_{in} varying from 0° to 90°, but $k_x k_z(k_z^2 - k_x^2)$ will change sign around $\theta_{\mathrm{in}} = 45°$. Exactly at 45° when $(k_z^2 - k_x^2) = 0$, the transverse spin of the magnetic field gradient $\mathbf{s}_y^{\mathrm{Qm}}$ is zero everywhere, regardless of the interference effect. As a result, the interfering electromagnetic field will exert a transverse optical torque (proportional to the field spin $\mathbf{s}_y^{\mathrm{m}}$) on an isotropic magnetic dipole, as well as a transverse torque (proportional to the field gradient spin $\mathbf{s}_y^{\mathrm{Qm}}$) on an isotropic magnetic quadrupole. However, as shown in Figs. 2(a) and 2(c), the optical torque acting on a magnetic quadrupole $\Gamma_{\mathrm{Qm},y}$ has a different dependence on incidence angle θ_{in} from its dipolar counterpart $\Gamma_{\mathrm{m},y}$. The different torques can be expressed analytically, together with their ratio, as

$$\Gamma_{\mathrm{m},y} = \frac{6\pi}{k^3}[\Re(b_1) - |b_1|^2]\frac{2k_x k_z \varepsilon_0}{k^2}\Im \times \left\{E_1^{s*}E_2^s\exp(i\varphi_0 - 2ik_x x)\right\},$$

$$\Gamma_{\mathrm{Qm},y} = \frac{120\pi}{k^5}[\Re\{b_2\} - |b_2|^2]\frac{2k_x k_z\left(k_z^2 - k_x^2\right)\varepsilon_0}{3k^2}\Im \times \left\{E_1^{s*}E_2^s\exp(i\varphi_0 - 2ik_x x)\right\},$$

$$\frac{\Gamma_{\mathrm{Qm},y}}{\Gamma_{m,y}} = \frac{20}{3}\frac{[\Re(b_2) - |b_2|^2]}{[\Re(b_1) - |b_1|^2]}\frac{\left(k_z^2 - k_x^2\right)}{k^2}. \tag{33}$$

When θ_{in} is below 45°, the torque on a magnetic quadrupole $\Gamma_{\mathrm{Qm},y}$ and the dipolar torque $\Gamma_{\mathrm{m},y}$ are in phase, as $(k_z^2 - k_x^2)/(k^2) > 0$. Above 45°, the torque on a magnetic quadrupole $\Gamma_{\mathrm{Qm},y}$ and the dipolar torque $\Gamma_{\mathrm{m},y}$ are out of phase, as $(k_z^2 - k_x^2)/(k^2) < 0$. In other words, the optical torque acting on a magnetic quadrupole points along the opposite orientation of the local magnetic field spin vector $\mathbf{s}^{\mathrm{m}}$. If one uses the local field spin vector $\mathbf{s}^{\mathrm{m}}$ to indicate the orientation of the optical torque, this result might seem counter-intuitive and be interpreted as a "negative" torque [5,17,19,20]. However, as we demonstrated, this counter-intuitive impression is due to the fact that the field spin vector should not be used to predict the orientation of a quadrupolar torque in the first place. Instead, the quadrupolar torque is always aligned with the field gradient spin vector as the absorption cross section is always a positive value for an absorbing isotropic Mie particle, and thus a "positive" torque.

The difference between dipolar and quadrupolar torque is even starker in the case of an electromagnetic field built up by two purely TM (p-) polarized plane waves. This type of electromagnetic field does not have any magnetic field spin as indicated in Eq. (32). As a result, it does not introduce optical torque on an isotropic magnetic dipole as shown in Fig. 2(b). However, a transverse spin of magnetic field gradient exists in the same electromagnetic field, and can introduce a strong optical torque to a magnetic quadrupole as shown in Fig. 2(d). Again this result argues against the use of local field spin vector to indicate optical torque on an object beyond the dipolar approximation, especially in a general structured electromagnetic field. It is also very interesting to notice from Figs. 2(a) and 2(d) the remarkable similarity between the dipolar torque $\Gamma_{\mathrm{m},y}$ for s-polarized two-wave interference and the quadrupolar torque $\Gamma_{\mathrm{Qm},y}$ for p-polarized two-wave interference, as they have the same incidence angle dependence of $(k_x k_z)$.

In Fig. 3, we study an electromagnetic field set up by the interference of two circularly polarized plane waves ($E_1^p = E_2^p = 1$ V/m, $E_1^s = E_2^s = i$ V/m). For this specific interfering electromagnetic field, the spin properties can also be described from Eq. (32). We can see that the x components of both the field and field gradient spin are zero. The transverse spin of the magnetic field is only dependent on the s-polarization components of each wave, while the transverse spin of the magnetic field gradient is dependent on both polarization components. As discussed earlier, the transverse spin of the magnetic field gradient in p polarized two-wave interference has the incidence angle dependence of $(k_x k_z)$, while the transverse spin of the magnetic field gradient in s polarized two-wave interference has a dependence of $k_x k_z(k_z^2 - k_x^2)$. Adding these two together will lead to an overall weaker transverse spin $\mathbf{s}_y^{\mathrm{Qm}}$ for incidence angles above 45° and a shift of zero $\mathbf{s}_y^{\mathrm{Qm}}$ angle to around 52° in this specific case, as can been seen in Fig. 3(c). The two types of EM field based on purely p-polarized or purely s-polarized two-wave interference don't exhibit net spin densities when integrating the spin densities in either $x = 0$, $y = 0$ or $z = 0$ plane. This can be easily derived from Eq. (32) for these two cases and clearly shown in Fig. 2. However, for the circularly polarized two wave interference, there is a net nonzero spin density along the z direction when integrating the spin of the EM field or field gradient in the $z = 0$ plane. The z component of the optical torque on a magnetic dipole $\Gamma_{\mathrm{m},z}$ shows the interference fringe pattern along x, but none of its values is negative. Only positive values of torque are seen, aligned with the net magnetic field spin density of the incident waves along the z direction, as shown in Fig. 3(b). Meanwhile, the z component of the optical torque $\Gamma_{\mathrm{Qm},z}$ on a magnetic quadrupole shows locally negative values compared to the net magnetic field spin density along z direction as shown in Fig. 3(d). They might be rightly interpreted as 'negative' torques, with reference to the net spin densities of the magnetic field gradient integrated across the entire $z = 0$ plane. Yet when referred to the magnetic field gradient spin vector, they are still positive torques for any absorbing isotropic Mie particle.

The results in Figs. 2 and 3 have also been verified using numerical integration of the total angular momentum flux, and agree with the results of our analytical expressions.

VIII. CONCLUSIONS

In this report, we study the optical torque acting on an induced multipole in an isotropic Mie particle and a time harmonic general (inhomogeneous) electromagnetic field.

We first try to address the confusion on the 'extrinsic' part of the dipolar torque present in Refs. [4,5], including the prediction of a torque that is dependent on the gradient of helicity density. With detailed calculations in the appendices, we rederive the analytical expressions of the dipolar torque in a general electromagnetic field by exploring the separate conservation laws of the total, spin and orbital angular momenta, a method proposed by Refs. [4,5]. We prove that the 'gradient' torque does not exist in an isotropic dipole. Though the "extrinsic" type of terms exist in the analytical expressions of dipolar torque attributed separately to the spin and orbital parts of the angular momentum, they cancel exactly with each other in the total dipolar torque.

In the second part of the work, we derive the analytical expression of the optical torque acting on an isotropic electromagnetic quadrupole in a general (inhomogeneous) electromagnetic field. With the analytical result, we show that the quadrupolar torque on an isotropic Mie particle is still proportional to the absorption cross section, confirming the transfer of the angular momentum to mechanical action through absorption. However, the relevant physical property of the incident beam that must be used to indicate the orientation of the optical torque is not the spin of the electromagnetic field, but the spin of the EM field gradient. Using the two-wave interference as an example, we show that there are significant differences between the spin of the electromagnetic field and the spin of the electromagnetic field gradient. As a result, simply using the local EM field spin can give rise to wrong predictions on the orientation of optical torque on large particles with strong quadrupole resonance. We show that an extraordinary transverse spin of the magnetic field gradient appears in a field formed by purely TM-polarized two wave interference, even though the magnetic field spin is zero. Furthermore, some of the nonintuitive negative torque that arises in the two-wave interference is often due to an incorrect interpretation of the physical properties related to the optical torque acting on a quadrupole and other higher-order multipoles.

Considering the growing interest from the nanophotonics community on studying and understanding the mechanical interaction between structured light and structured materials, we felt the need to address some discrepancies in the literature. By using consistency checks, double-checking our results using different methods, and scrutinizing our expressions via examples, we have produced what we feel are trusty analytical expressions, which we hope serve the community as much as it served us to clear up some confusions and improve our understanding of the role of angular momentum in the mechanical interaction of light with matter.

ACKNOWLEDGMENTS

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APPENDIX A: INDUCED DIPOLE AND QUADRUPOLE MOMENTS IN ISOTROPIC MIE PARTICLES

For an isotropic Mie particle, the polarizabilities for electric dipole α_e, magnetic dipole α_m, electric quadrupole α_{Qe} and magnetic quadrupole α_{Qm} are defined by the corresponding Mie coefficients as follows:

$$\alpha_e = i\frac{6\pi}{k^3}\varepsilon_0 a_1, \quad \alpha_m = i\frac{6\pi}{k^3} b_1,$$
$$\alpha_{Qe} = i\frac{120\pi}{k^5}\varepsilon_0 a_2, \quad \alpha_{Qm} = i\frac{120\pi}{k^5} b_2.$$

The induced electric and magnetic dipole moments are vectors determined by the EM dipole polarizabilities and the incident EM field:

$$\mathbf{p} = \alpha_e \mathbf{E}_{inc}, \quad \mathbf{m} = \alpha_m \mathbf{H}_{inc}. \tag{A1}$$

The induced electric and magnetic quadrupole moments are tensors determined by the EM quadrupole polarizabilities and the incident EM field and the field gradients [22–24]:

$$\overleftrightarrow{\mathbf{Q}}^{e} = \alpha_{Qe} \overleftrightarrow{\mathcal{D}}^{e}, \quad \overleftrightarrow{\mathbf{Q}}^{m} = \alpha_{Qm} \overleftrightarrow{\mathcal{D}}^{m}, \tag{A2}$$

where

$$\overleftrightarrow{\mathcal{D}}^{e} = \frac{\nabla \otimes \mathbf{E}_{inc} + \mathbf{E}_{inc} \otimes \nabla}{2},$$
$$\overleftrightarrow{\mathcal{D}}^{m} = \frac{\nabla \otimes \mathbf{H}_{inc} + \mathbf{H}_{inc} \otimes \nabla}{2}. \tag{A3}$$

APPENDIX B: RADIATION FIELDS OF ELECTROMAGNETIC DIPOLE AND QUADRUPOLE

Knowing the electromagnetic dipole and quadrupole moments, the corresponding radiation fields can be analytically expressed [25]. The radiation fields of the dipoles are expressed as

$$\mathbf{E}_p = \frac{k^2}{\varepsilon_0}(\overleftrightarrow{\mathbf{G}}^{d} \cdot \mathbf{p}),$$
$$\mathbf{H}_p = -ikc_0 \nabla \times (\overleftrightarrow{\mathbf{G}}^{d} \cdot \mathbf{p}) = -ikc_0(\mathbf{g}^d \times \mathbf{p}),$$
$$\mathbf{E}_m = \frac{ik}{c_0\varepsilon_0}\nabla \times (\overleftrightarrow{\mathbf{G}}^{d} \cdot \mathbf{m}) = \frac{ik}{c_0\varepsilon_0}(\mathbf{g}^d \times \mathbf{m}),$$
$$\mathbf{H}_m = k^2(\overleftrightarrow{\mathbf{G}}^{d} \cdot \mathbf{m}), \tag{B1}$$

where

$$\overleftrightarrow{\mathbf{G}}^{d} = \left\{\left(1+\frac{i}{kr}-\frac{1}{(kr)^2}\right)\overleftrightarrow{\mathbf{I}} - \left(1+\frac{3i}{kr}-\frac{3}{(kr)^2}\right)\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}\right\}\frac{e^{ikr}}{4\pi r},$$
$$\mathbf{g}^{d} = \hat{\mathbf{r}}\left(ik-\frac{1}{r}\right)\frac{e^{ikr}}{4\pi r}, \tag{B2}$$

with $\overleftrightarrow{\mathbf{I}}$ being the 3-by-3 identity tensor.

The radiation fields of the quadrupoles are expressed as

$$\mathbf{E}_{\mathrm{Qe}} = \frac{k^2}{\varepsilon_0}[\overleftrightarrow{\mathbf{G}}^{\mathrm{Q}}\cdot(\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}}\cdot\hat{\mathbf{r}})],$$
$$\mathbf{H}_{\mathrm{Qe}} = -ikc_0\nabla\times[\overleftrightarrow{\mathbf{G}}^{\mathrm{Q}}\cdot(\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}}\cdot\hat{\mathbf{r}})] = -ikc_0[\mathbf{g}^{\mathrm{Q}}\times(\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}}\cdot\hat{\mathbf{r}})],$$
$$\mathbf{E}_{\mathrm{Qm}} = \frac{ik}{c_0\varepsilon_0}\nabla\times[\overleftrightarrow{\mathbf{G}}^{\mathrm{Q}}\cdot(\overleftrightarrow{\mathbf{Q}}^{\mathrm{m}}\cdot\hat{\mathbf{r}})] = \frac{ik}{c_0\varepsilon_0}[\mathbf{g}^{\mathrm{Q}}\times(\overleftrightarrow{\mathbf{Q}}^{\mathrm{m}}\cdot\hat{\mathbf{r}})],$$
$$\mathbf{H}_{\mathrm{Qm}} = k^2[\overleftrightarrow{\mathbf{G}}^{\mathrm{Q}}\cdot(\overleftrightarrow{\mathbf{Q}}^{\mathrm{m}}\cdot\hat{\mathbf{r}})], \tag{B3}$$

where

$$\overleftrightarrow{\mathbf{G}}^{\mathrm{Q}} = \left\{\left(-1-\frac{3i}{kr}+\frac{6}{(kr)^2}+\frac{6i}{(kr)^3}\right)\overleftrightarrow{\mathbf{I}} + \left(1+\frac{6i}{kr}-\frac{15}{(kr)^2}-\frac{15i}{(kr)^3}\right)\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}\right\}\frac{ike^{ikr}}{24\pi r},$$
$$\mathbf{g}^{\mathrm{Q}} = \hat{\mathbf{r}}\left[1+\frac{3i}{kr}-\frac{3}{(kr)^2}\right]\frac{k^2e^{ikr}}{24\pi r}. \tag{B4}$$

APPENDIX C: DERIVATION OF THE ANALYTICAL DIPOLAR TORQUE

In this section, we will outline the main derivation steps for the analytical expressions of optical torque $\boldsymbol{\Gamma}_{\mathrm{p}}$ on an electric dipole in a general electromagnetic field. Assume an isotropic Mie particle is placed at the origin $\mathbf{r}_0$. As shown in Eq. (13), the total optical torque be calculated by integerating the total angular momentum over an enclosed spherical surface surrounding the object,

$$\boldsymbol{\Gamma} = \int_0^{2\pi}\int_0^{\pi}\mathbf{r}\times(\langle\overleftrightarrow{\mathbf{T}}\rangle\cdot\hat{\mathbf{n}})r^2\sin\theta d\theta d\phi, = \Re\int_0^{2\pi}\int_0^{\pi}(r\hat{\mathbf{n}})\times\left\{\frac{\varepsilon_0}{2}\mathbf{E}_{\mathrm{tot}}(\mathbf{E}^*_{\mathrm{tot}}\cdot\hat{\mathbf{n}}) + \frac{\mu_0}{2}\mathbf{H}_{\mathrm{tot}}(\mathbf{H}^*_{\mathrm{tot}}\cdot\hat{\mathbf{n}})\right\}r^2\sin\theta d\theta d\phi, \tag{C1}$$

where $\mathbf{r} = \mathbf{r}' - \mathbf{r}_0 = r\hat{\mathbf{n}}$, $\mathbf{r}'$ denotes a point on the spherical surface, r is the radius of the spherical surface and $\hat{\mathbf{n}}$ is the outward unit vector normal to the surface, $\hat{\mathbf{n}} = \hat{\mathbf{r}} = n_x\hat{\mathbf{e}}_x + n_y\hat{\mathbf{e}}_y + n_z\hat{\mathbf{e}}_z$ ($n_x = \sin\theta\cos\phi$, $n_y = \sin\theta\sin\phi$ and $n_z = \cos\theta$). Here we only consider the torque contributed by the induced electric dipole and the incident field so that $\mathbf{E}_{\mathrm{tot}}(\mathbf{r}') = \mathbf{E}_{\mathrm{inc}}(\mathbf{r}') + \mathbf{E}_{\mathrm{p}}(\mathbf{r}')$ and $\mathbf{H}_{\mathrm{tot}}(\mathbf{r}') = \mathbf{H}_{\mathrm{inc}}(\mathbf{r}') + \mathbf{H}_{\mathrm{p}}(\mathbf{r}')$. The analytical expressions of optical torque on an electric dipole can be derived based on the knowledge that the radiation field of an induced electric dipole can be analytically expression as in Appendixes A and B.

The optical torque $\boldsymbol{\Gamma}_{\mathrm{p}}$, attributed to the interaction between the induced electric dipole and incident EM field, can be decomposed into different parts as shown in Eq. (13) and Eq. (14), namely the torque component purely dependent on incident EM field,

$$\boldsymbol{\Gamma}_{\mathrm{inc}} = \frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\mathrm{inc}})(\mathbf{E}^*_{\mathrm{inc}}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi + \frac{\mu_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{H}_{\mathrm{inc}})(\mathbf{H}^*_{\mathrm{inc}}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi, \tag{C2}$$

the extinction torque $\boldsymbol{\Gamma}_{\mathrm{p,mix}}$ dependent on the interference between incident and radiation fields

$$\boldsymbol{\Gamma}_{\mathrm{p,mix}} = \frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\mathrm{inc}})(\mathbf{E}^*_{\mathrm{p}}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi + \frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\mathrm{p}})(\mathbf{E}^*_{\mathrm{inc}}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi$$
$$+ \frac{\mu_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{H}_{\mathrm{inc}})(\mathbf{H}^*_{\mathrm{p}}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi + \frac{\mu_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{H}_{\mathrm{p}})(\mathbf{H}^*_{\mathrm{inc}}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi, \tag{C3}$$

and the recoil torque $\boldsymbol{\Gamma}_{\mathrm{p,recoil}}$ as a result of self-interaction of the induced electric dipole

$$\boldsymbol{\Gamma}_{\mathrm{p,recoil}} = \frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\mathrm{p}})(\mathbf{E}^*_{\mathrm{p}}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi + \frac{\mu_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{H}_{\mathrm{p}})(\mathbf{H}^*_{\mathrm{p}}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi. \tag{C4}$$

Due to the fact that the angular momenta of the incident, radiation and total fields are separately conserved quantities, the corresponding contributions to the dipolar torque can be calculated independent of the radius of the enclosed surface,

$$\begin{aligned}\boldsymbol{\Gamma}_{\mathrm{p}} &= \lim_{r\to\infty}\boldsymbol{\Gamma}_{\mathrm{p}}(r) = \lim_{r\to 0}\boldsymbol{\Gamma}_{\mathrm{p}}(r),\\ \boldsymbol{\Gamma}_{\mathrm{inc}} &= \lim_{r\to\infty}\boldsymbol{\Gamma}_{\mathrm{inc}}(r) = \lim_{r\to 0}\boldsymbol{\Gamma}_{\mathrm{inc}}(r),\\ \boldsymbol{\Gamma}_{\mathrm{p,recoil}} &= \lim_{r\to\infty}\boldsymbol{\Gamma}_{\mathrm{p,recoil}}(r) = \lim_{r\to 0}\boldsymbol{\Gamma}_{\mathrm{p,recoil}}(r),\\ \boldsymbol{\Gamma}_{\mathrm{p,mix}} &= \lim_{r\to\infty}\boldsymbol{\Gamma}_{\mathrm{p,mix}}(r) = \lim_{r\to 0}\boldsymbol{\Gamma}_{\mathrm{p,mix}}(r),\end{aligned} \tag{C5}$$

It follows from Eqs. (B1) and (B2) that

$$\mathbf{H}_{\mathrm{p}}^{*}\cdot\hat{\mathbf{n}} = 0. \tag{C6}$$

It is thus easy to prove that

$$\begin{aligned}&\frac{\mu_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{H}_{\mathrm{inc}})(\mathbf{H}_{\mathrm{p}}^{*}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi = 0,\\ &\frac{\mu_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{H}_{\mathrm{p}})(\mathbf{H}_{\mathrm{p}}^{*}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi = 0.\end{aligned} \tag{C7}$$

In the small r limit that $r\to 0$, we can approximate the incident electric field on the integration spherical surface, to the first order as

$$\begin{aligned}\mathbf{E}_{\mathrm{inc}}(\mathbf{r}') &\approx \mathbf{E}_0 + r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0,\\ \mathbf{H}_{\mathrm{inc}}(\mathbf{r}') &\approx \mathbf{H}_0 + r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0,\end{aligned} \tag{C8}$$

where $\mathbf{E}_0 = \mathbf{E}_{\mathrm{inc}}(\mathbf{r}_0)$ and $\mathbf{H}_0 = \mathbf{H}_{\mathrm{inc}}(\mathbf{r}_0)$.

The optical torque component $\boldsymbol{\Gamma}_{\mathrm{inc}}$ attributed purely to the incident EM field can be proven to be zero as follows:

$$\begin{aligned}\boldsymbol{\Gamma}_{\mathrm{inc}} &= \lim_{r\to 0}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\mathrm{inc}})(\mathbf{E}_{\mathrm{inc}}^{*}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi + \lim_{r\to 0}\frac{\mu_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{H}_{\mathrm{inc}})(\mathbf{H}_{\mathrm{inc}}^{*}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= \lim_{r\to 0}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_0)(\mathbf{E}_0^{*}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi + \lim_{r\to 0}\frac{\mu_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{H}_0)(\mathbf{H}_0^{*}\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= 0.\end{aligned} \tag{C9}$$

We then calculate the remaining nonzero terms of $\boldsymbol{\Gamma}_{\mathrm{p}}$. The following properties related to $\hat{\mathbf{n}}$ have been used in the derivations:

$$\begin{aligned}&\int_0^{2\pi}\int_0^{\pi} n_l\sin\theta d\theta d\phi = 0,\\ &\int_0^{2\pi}\int_0^{\pi} n_u n_v\sin\theta d\theta d\phi = \frac{4\pi}{3}\delta_{uv},\\ &\int_0^{2\pi}\int_0^{\pi} n_l n_u n_v\sin\theta d\theta d\phi = 0,\end{aligned} \tag{C10}$$

and

$$\begin{aligned}&\int_0^{2\pi}\int_0^{\pi} n_l n_u n_v n_m\sin\theta d\theta d\phi = 0, \quad \text{except}\\ &\int_0^{2\pi}\int_0^{\pi} (n_u)^4\sin\theta d\theta d\phi = \frac{4\pi}{5},\\ &\int_0^{2\pi}\int_0^{\pi} (n_u)^2(n_v)^2\sin\theta d\theta d\phi = \frac{4\pi}{15},\ \ u\neq v.\end{aligned} \tag{C11}$$

It follows from Eqs. (B1) and (B2) that

$$\mathbf{E}_{\mathrm{p}}^{*}\cdot\hat{\mathbf{n}} = \frac{k^2 e^{-ikr}}{4\pi\varepsilon_0 r}\left[\frac{2i}{kr} + \frac{2}{(kr)^2}\right](\mathbf{p}^{*}\cdot\hat{\mathbf{n}}), \tag{C12}$$

and using aforementioned properties of $\hat{\mathbf{n}}$, we can derive that

$$\begin{aligned}\lim_{r\to 0}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\rm inc})(\mathbf{E}_{\rm p}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi &= \lim_{r\to 0}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\rm inc})\frac{k^2 e^{-ikr}}{4\pi\varepsilon_0 r}\left[\frac{2i}{kr}+\frac{2}{(kr)^2}\right](\mathbf{p}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= \lim_{r\to 0}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\rm inc})\frac{e^{-ikr}}{4\pi}[ikr+1](\mathbf{p}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= \frac{1}{4\pi}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_0)(\mathbf{p}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= \frac{1}{3}\Re\{\mathbf{p}^*\times\mathbf{E}_0\}.\end{aligned} \tag{C13}$$

It follows from Eqs. (B1) and (B2) that

$$\hat{\mathbf{n}}\times\mathbf{E}_{\rm p} = \frac{k^2 e^{ikr}}{4\pi\varepsilon_0 r}\left[1+\frac{i}{kr}-\frac{1}{(kr)^2}\right](\hat{\mathbf{n}}\times\mathbf{p}), \tag{C14}$$

and using aforementioned properties of $\hat{\mathbf{n}}$, we can derive that

$$\begin{aligned}&\lim_{r\to 0}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\rm p})(\mathbf{E}_{\rm inc}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= \lim_{r\to 0}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}\frac{k^2 e^{ikr}}{4\pi\varepsilon_0 r}\left[1+\frac{i}{kr}-\frac{1}{(kr)^2}\right](\hat{\mathbf{n}}\times\mathbf{p})(\mathbf{E}_{\rm inc}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= \lim_{r\to 0}\Re\int_0^{2\pi}\int_0^{\pi}\frac{e^{ikr}}{8\pi}(k^2r^2+ikr-1)(\hat{\mathbf{n}}\times\mathbf{p})(\mathbf{E}_{\rm inc}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= -\frac{1}{8\pi}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{p})(\mathbf{E}_0^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= \frac{1}{6}\Re\{\mathbf{p}^*\times\mathbf{E}_0\}.\end{aligned} \tag{C15}$$

It follows from Eqs. (B1) and (B2) that

$$\hat{\mathbf{n}}\times\mathbf{H}_{\rm p} = \frac{-ikc_0 e^{ikr}}{4\pi r}\left[ik-\frac{1}{r}\right][\hat{\mathbf{n}}\times(\hat{\mathbf{n}}\times\mathbf{p})], \tag{C16}$$

it is easy to prove that

$$\begin{aligned}&\lim_{r\to 0}\frac{\mu_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{H}_{\rm p})(\mathbf{H}_{\rm inc}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= \lim_{r\to 0}\frac{\mu_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}\frac{-ikc_0 e^{ikr}}{4\pi r}\left[ik-\frac{1}{r}\right][\hat{\mathbf{n}}\times(\hat{\mathbf{n}}\times\mathbf{p})](\mathbf{H}_{\rm inc}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= \lim_{r\to 0}\Re\int_0^{2\pi}\int_0^{\pi}\frac{\mu_0 c_0 e^{ikr}}{8\pi}(k^2r^2+ikr)[\hat{\mathbf{n}}\times(\hat{\mathbf{n}}\times\mathbf{p})](\mathbf{H}_{\rm inc}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\ &= 0.\end{aligned} \tag{C17}$$

The last nonzero term contributing to the electric dipolar torque is the so called ‘recoil’ torque. Using the relations of $(\hat{\mathbf{n}} \times \mathbf{E}_\mathrm{p})$ and $(\mathbf{E}_\mathrm{p}^* \cdot \hat{\mathbf{n}})$ just derived, we can arrive at the analytical expression of $\mathbf{\Gamma}_\mathrm{p,recoil}$:

$$\begin{aligned}
\mathbf{\Gamma}_\mathrm{p,recoil} &= \lim_{r\to\infty} \frac{\varepsilon_0 r^3}{2} \Re \int_0^{2\pi} \int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{E}_\mathrm{p})(\mathbf{E}_\mathrm{p}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi \\
&= \lim_{r\to\infty} \frac{\varepsilon_0 r^3}{2} \Re \int_0^{2\pi} \int_0^{\pi} \frac{k^2 e^{ikr}}{4\pi\varepsilon_0 r}\left[1 + \frac{i}{kr} - \frac{1}{(kr)^2}\right](\hat{\mathbf{n}} \times \mathbf{p}) \frac{k^2 e^{-ikr}}{4\pi\varepsilon_0 r}\left[\frac{2i}{kr} + \frac{2}{(kr)^2}\right](\mathbf{p}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi \\
&= \Re \int_0^{2\pi} \int_0^{\pi} \frac{ik^3}{16\pi^2\varepsilon_0}[\hat{\mathbf{n}} \times \mathbf{p}](\mathbf{p}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi \\
&= \Re \frac{ik^3}{16\pi^2\varepsilon_0} \frac{4\pi}{3}\{\mathbf{p}^* \times \mathbf{p}\} \\
&= -\frac{k^3}{12\pi\varepsilon_0}\Im\{\mathbf{p}^* \times \mathbf{p}\}.
\end{aligned} \tag{C18}$$

The analytical expression of optical torque corresponding to an induced electric dipole of a particle positioned in a general electromagnetic field is thus derived from the total angular momentum flux method. The analytical expression of a magnetic dipole torque can be easily derived in a similar manner. Both analytical expressions can be given as

$$\begin{aligned}
\mathbf{\Gamma}_\mathrm{p} &= \frac{1}{2}\Re\{\mathbf{p}^* \times \mathbf{E}_0\} - \frac{k^3}{12\pi\varepsilon_0}\Im\{\mathbf{p}^* \times \mathbf{p}\} \\
&= \frac{1}{2}\Re\{\mathbf{p}^* \times \mathbf{E}_\mathrm{inc}\} - \frac{k^3}{12\pi\varepsilon_0}\Im\{\mathbf{p}^* \times \mathbf{p}\}, \\
\mathbf{\Gamma}_\mathrm{m} &= \frac{1}{2}\Re\{\mathbf{m}^* \times \mu_0\mathbf{H}_0\} - \frac{k^3\mu_0}{12\pi}\Im\{\mathbf{m}^* \times \mathbf{m}\} \\
&= \frac{1}{2}\Re\{\mathbf{m}^* \times \mu_0\mathbf{H}_\mathrm{inc}\} - \frac{k^3\mu_0}{12\pi}\Im\{\mathbf{m}^* \times \mathbf{m}\}.
\end{aligned} \tag{C19}$$

For a combined electric and magnetic dipole, additional terms due to the interference of the electric and magnetic dipole could arise. However, these are shown to be zero, following Eq. (B1), Eq. (B2), and the aforementioned properties of $\hat{\mathbf{n}}$, it is easy to prove that

$$\begin{aligned}
\mathbf{\Gamma}_\mathrm{pm,recoil} &= \frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{E}_\mathrm{p})(\mathbf{E}_\mathrm{m}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi + \frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{E}_\mathrm{m})(\mathbf{E}_\mathrm{p}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi \\
&\quad + \frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_\mathrm{p})(\mathbf{H}_\mathrm{m}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi + \frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_\mathrm{m})(\mathbf{H}_\mathrm{p}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi \\
&= \lim_{r\to\infty}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{E}_\mathrm{m})(\mathbf{E}_\mathrm{p}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi + \lim_{r\to\infty}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_\mathrm{p})(\mathbf{H}_\mathrm{m}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi \\
&= 0.
\end{aligned} \tag{C20}$$

To summarize, the dipolar optical torque in a general electromagnetic field can be expressed as

$$\mathbf{\Gamma}_d = \mathbf{\Gamma}_\mathrm{p} + \mathbf{\Gamma}_\mathrm{m}. \tag{C21}$$

APPENDIX D: DERIVATION OF THE ANALYTICAL “SPIN” AND “ORBITAL” DIPOLAR TORQUE

In this section, a brief derivation of the optical torque acting on an electric dipole using the spin and orbital angular momentum flux method is shown, and the optical torque acting on a magnetic dipole can be derived in a similar way. The optical torque attributed to the spin angular momentum flux $\langle \overleftrightarrow{\mathbf{M}}^s \rangle$ can be derived by integrating the SAM flux over a spherical surface with the object at its center as defined in previous section,

$$\begin{aligned}
\mathbf{\Gamma}_\mathrm{p}^s &= \int_0^{2\pi}\int_0^{\pi} (\langle \overleftrightarrow{\mathbf{M}}^s \rangle \cdot \hat{\mathbf{n}}) r^2 \sin\theta d\theta d\phi, \\
&= \int_0^{2\pi}\int_0^{\pi} \frac{1}{2\omega}\Im\{\mathbf{E}_\mathrm{tot}(\mathbf{H}_\mathrm{tot}^* \cdot \hat{\mathbf{n}}) + \mathbf{H}_\mathrm{tot}^*(\mathbf{E}_\mathrm{tot} \cdot \hat{\mathbf{n}}) - (\mathbf{E}_\mathrm{tot} \cdot \mathbf{H}_\mathrm{tot}^*)\hat{\mathbf{n}}\} r^2 \sin\theta d\theta d\phi,
\end{aligned} \tag{D1}$$

where $\mathbf{E}_\mathrm{tot}(\mathbf{r}') = \mathbf{E}_\mathrm{inc}(\mathbf{r}') + \mathbf{E}_\mathrm{p}(\mathbf{r}')$ and $\mathbf{H}_\mathrm{tot}(\mathbf{r}') = \mathbf{H}_\mathrm{inc}(\mathbf{r}') + \mathbf{H}_\mathrm{p}(\mathbf{r}')$.

The SAM related torque on an electric dipole $\mathbf{\Gamma}^s_{\mathrm{p}}$ can also be separated into three parts,

$$\mathbf{\Gamma}^s_{\mathrm{p}} = \mathbf{\Gamma}^s_{\mathrm{inc}} + \mathbf{\Gamma}^s_{\mathrm{p,mix}} + \mathbf{\Gamma}^s_{\mathrm{p,recoil}}, \tag{D2}$$

where $\mathbf{\Gamma}^s_{\mathrm{inc}}$ depends purely on the incident field,

$$\mathbf{\Gamma}^s_{\mathrm{inc}} = \int_0^{2\pi}\int_0^{\pi} \frac{1}{2\omega}\Im\{\mathbf{E}_{\mathrm{inc}}(\mathbf{H}^*_{\mathrm{inc}}\cdot\hat{\mathbf{n}}) + \mathbf{H}^*_{\mathrm{inc}}(\mathbf{E}_{\mathrm{inc}}\cdot\hat{\mathbf{n}}) - (\mathbf{E}_{\mathrm{inc}}\cdot\mathbf{H}^*_{\mathrm{inc}})\hat{\mathbf{n}}\}r^2\sin\theta d\theta d\phi. \tag{D3}$$

$\mathbf{\Gamma}^s_{\mathrm{p,mix}}$ relies on the interference between the incident and radiation field,

$$\mathbf{\Gamma}^s_{\mathrm{p,mix}} = \int_0^{2\pi}\int_0^{\pi} \frac{1}{2\omega}\Im\{\mathbf{E}_{\mathrm{inc}}(\mathbf{H}^*_{\mathrm{p}}\cdot\hat{\mathbf{n}}) + \mathbf{E}_{\mathrm{p}}(\mathbf{H}^*_{\mathrm{inc}}\cdot\hat{\mathbf{n}}) + \mathbf{H}^*_{\mathrm{inc}}(\mathbf{E}_{\mathrm{p}}\cdot\hat{\mathbf{n}}) + \mathbf{H}^*_{\mathrm{p}}(\mathbf{E}_{\mathrm{inc}}\cdot\hat{\mathbf{n}}) - (\mathbf{E}_{\mathrm{inc}}\cdot\mathbf{H}^*_{\mathrm{p}} + \mathbf{E}_{\mathrm{p}}\cdot\mathbf{H}^*_{\mathrm{inc}})\hat{\mathbf{n}}\}r^2\sin\theta d\theta d\phi, \tag{D4}$$

while $\mathbf{\Gamma}^s_{\mathrm{p,recoil}}$ is attributed to the radiation field only,

$$\mathbf{\Gamma}^s_{\mathrm{p,recoil}} = \int_0^{2\pi}\int_0^{\pi} \frac{1}{2\omega}\Im\{\mathbf{E}_{\mathrm{p}}(\mathbf{H}^*_{\mathrm{p}}\cdot\hat{\mathbf{n}}) + \mathbf{H}^*_{\mathrm{p}}(\mathbf{E}_{\mathrm{p}}\cdot\hat{\mathbf{n}}) - (\mathbf{E}_{\mathrm{p}}\cdot\mathbf{H}^*_{\mathrm{p}})\hat{\mathbf{n}}\}r^2\sin\theta d\theta d\phi. \tag{D5}$$

Due to the fact that the spin angular momenta of the incident, radiation and total fields are separately conserved quantities, the corresponding contributions to the dipolar torque can be calculated independent of the radius of the enclosed surface,

$$\begin{aligned}
\mathbf{\Gamma}^s_{\mathrm{p}} &= \lim_{r\to\infty}\mathbf{\Gamma}^s_{\mathrm{p}}(r) = \lim_{r\to 0}\mathbf{\Gamma}^s_{\mathrm{p}}(r),\\
\mathbf{\Gamma}^s_{\mathrm{inc}} &= \lim_{r\to\infty}\mathbf{\Gamma}^s_{\mathrm{inc}}(r) = \lim_{r\to 0}\mathbf{\Gamma}^s_{\mathrm{inc}}(r),\\
\mathbf{\Gamma}^s_{\mathrm{p,recoil}} &= \lim_{r\to\infty}\mathbf{\Gamma}^s_{\mathrm{p,recoil}}(r) = \lim_{r\to 0}\mathbf{\Gamma}^s_{\mathrm{p,recoil}}(r),\\
\mathbf{\Gamma}^s_{\mathrm{p,mix}} &= \lim_{r\to\infty}\mathbf{\Gamma}^s_{\mathrm{p,mix}}(r) = \lim_{r\to 0}\mathbf{\Gamma}^s_{\mathrm{p,mix}}(r).
\end{aligned} \tag{D6}$$

In the small r limit that $r \to 0$, using the approximation that $\mathbf{E}_{\mathrm{inc}}(\mathbf{r}') \approx \mathbf{E}_0 + r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0$ and $\mathbf{H}_{\mathrm{inc}}(\mathbf{r}') \approx \mathbf{H}_0 + r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0$, it is easy to prove that the optical torque component $\mathbf{\Gamma}^s_{\mathrm{inc}}$ attributed purely to the incident field is zero:

$$\begin{aligned}
\mathbf{\Gamma}^s_{\mathrm{inc}} &= \lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi} \frac{1}{2\omega}\Im\{\mathbf{E}_{\mathrm{inc}}(\mathbf{H}^*_{\mathrm{inc}}\cdot\hat{\mathbf{n}}) + \mathbf{H}^*_{\mathrm{inc}}(\mathbf{E}_{\mathrm{inc}}\cdot\hat{\mathbf{n}}) - (\mathbf{E}_{\mathrm{inc}}\cdot\mathbf{H}^*_{\mathrm{inc}})\hat{\mathbf{n}}\}r^2\sin\theta d\theta d\phi\\
&= \lim_{r\to 0} r^2\int_0^{2\pi}\int_0^{\pi} \frac{1}{2\omega}\Im\{\mathbf{E}_0(\mathbf{H}^*_0\cdot\hat{\mathbf{n}}) + \mathbf{H}^*_0(\mathbf{E}_0\cdot\hat{\mathbf{n}}) - (\mathbf{E}_0\cdot\mathbf{H}^*_0)\hat{\mathbf{n}}\}\sin\theta d\theta d\phi\\
&= 0.
\end{aligned} \tag{D7}$$

The various components of $\mathbf{\Gamma}^s_{\mathrm{p,mix}}$ can be derived following a similar method as in the previous section, using the dipolar radiation field properties in Eqs. (B1) and (B2), the aforementioned integration properties of the unit vector $\hat{\mathbf{n}}$, and the approximation of the incident field in the small r limit $\mathbf{E}_{\mathrm{inc}}(\mathbf{r}') \approx \mathbf{E}_0 + r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0$ and $\mathbf{H}_{\mathrm{inc}}(\mathbf{r}') \approx \mathbf{H}_0 + r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0$. Their analytical results are listed below:

$$\int_0^{2\pi}\int_0^{\pi} \frac{1}{2\omega}\Im\{\mathbf{E}_{\mathrm{inc}}(\mathbf{H}^*_{\mathrm{p}}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi = 0, \tag{D8}$$

$$\begin{aligned}
\lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi} \frac{1}{2\omega}\Im\{\mathbf{H}^*_{\mathrm{p}}(\mathbf{E}_{\mathrm{inc}}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi &= \Im\int_0^{2\pi}\int_0^{\pi}\left\{\frac{-i}{8\pi}(\hat{\mathbf{n}}\times\mathbf{p}^*)(\mathbf{E}_0\cdot\hat{\mathbf{n}})\right\}\sin\theta d\theta d\phi\\
&= \frac{1}{6}\Re\{\mathbf{p}^*\times\mathbf{E}_0\},
\end{aligned} \tag{D9}$$

$$\begin{aligned}
\lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi} \frac{1}{2\omega}\Im\{-(\mathbf{E}_{\mathrm{inc}}\cdot\mathbf{H}^*_{\mathrm{p}})\hat{\mathbf{n}}\}r^2\sin\theta d\theta d\phi &= \Im\int_0^{2\pi}\int_0^{\pi}\left\{\frac{i}{8\pi}[(\hat{\mathbf{n}}\times\mathbf{p}^*)\cdot\mathbf{E}_0]\hat{\mathbf{n}}\right\}\sin\theta d\theta d\phi\\
&= \frac{1}{6}\Re\{\mathbf{p}^*\times\mathbf{E}_0\},
\end{aligned} \tag{D10}$$

$$\begin{aligned}
\lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi} \frac{1}{2\omega}\Im\{\mathbf{E}_{\mathrm{p}}(\mathbf{H}^*_{\mathrm{inc}}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi &= \Im\int_0^{2\pi}\int_0^{\pi}\frac{-1}{8\pi\omega\varepsilon_0}\{[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}^*_0]\cdot\hat{\mathbf{n}}\}\mathbf{p}\sin\theta d\theta d\phi\\
&\quad + \Im\int_0^{2\pi}\int_0^{\pi}\frac{3}{8\pi\omega\varepsilon_0}\{[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}^*_0]\cdot\hat{\mathbf{n}}\}(\hat{\mathbf{n}}\cdot\mathbf{p})\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&= \Im\int_0^{2\pi}\int_0^{\pi}\frac{3}{8\pi\omega\varepsilon_0}\{[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}^*_0]\cdot\hat{\mathbf{n}}\}(\hat{\mathbf{n}}\cdot\mathbf{p})\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&= \frac{1}{10}\Re\{\mathbf{p}^*\times\mathbf{E}_0\} + \frac{1}{5\omega\varepsilon_0}\Im\{(\mathbf{p}\cdot\nabla)\mathbf{H}^*_0\},
\end{aligned} \tag{D11}$$

$$\begin{aligned}\lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi}\frac{1}{2\omega}\Im\{\mathbf{H}_{\mathrm{inc}}^*(\mathbf{E}_{\mathrm{p}}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi &= \Im\int_0^{2\pi}\int_0^{\pi}\left\{\frac{1}{4\pi\omega\varepsilon_0}(\hat{\mathbf{n}}\cdot\mathbf{p})[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0^*]\right\}\sin\theta d\theta d\phi\\ &= \frac{1}{3\omega\varepsilon_0}\Im\{(\mathbf{p}\cdot\nabla)\mathbf{H}_0^*\},\end{aligned} \tag{D12}$$

$$\begin{aligned}\lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi}\frac{1}{2\omega}\Im\{-(\mathbf{E}_{\mathrm{p}}\cdot\mathbf{H}_{\mathrm{inc}}^*)\hat{\mathbf{n}}\}r^2\sin\theta d\theta d\phi &= \Im\int_0^{2\pi}\int_0^{\pi}\left\{\frac{1}{8\pi\omega\varepsilon_0}\{[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0^*]\cdot\mathbf{p}\}\hat{\mathbf{n}}\right\}\sin\theta d\theta d\phi\\ &\quad+\Im\int_0^{2\pi}\int_0^{\pi}\frac{-3}{8\pi\omega\varepsilon_0}\{[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0^*]\cdot\hat{\mathbf{n}}\}(\hat{\mathbf{n}}\cdot\mathbf{p})\hat{\mathbf{n}}\sin\theta d\theta d\phi\\ &= \frac{1}{6}\Re(\mathbf{p}^*\times\mathbf{E}_0)+\frac{1}{6\omega\varepsilon_0}\Im[(\mathbf{p}\cdot\nabla)\mathbf{H}_0^*]\\ &\quad-\left\{\frac{1}{10}\Re(\mathbf{p}^*\times\mathbf{E}_0)+\frac{1}{5\omega\varepsilon_0}\Im[(\mathbf{p}\cdot\nabla)\mathbf{H}_0^*]\right\}.\end{aligned} \tag{D13}$$

From the above results, we can get the analytical expression for $\mathbf{\Gamma}^s_{\mathrm{p,mix}}$

$$\mathbf{\Gamma}^s_{\mathrm{p,mix}} = \frac{1}{2}\Re\{\mathbf{p}^*\times\mathbf{E}_{\mathrm{inc}}\}+\frac{1}{2\omega\varepsilon_0}\Im\{(\mathbf{p}\cdot\nabla)\mathbf{H}_{\mathrm{inc}}^*\}. \tag{D14}$$

Similarly, the various components of $\mathbf{\Gamma}^s_{\mathrm{p,recoil}}$ can be derived using the dipolar radiation field properties in Eq. (B1) and Eq. (B2) and the aforementioned integration properties of the unit vector $\hat{\mathbf{n}}$ in the large r limit as $r\to\infty$,

$$\begin{aligned}&\lim_{r\to\infty}\int_0^{2\pi}\int_0^{\pi}\frac{1}{2\omega}\Im\{\mathbf{H}_{\mathrm{p}}^*(\mathbf{E}_{\mathrm{p}}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi = 0,\\ &\int_0^{2\pi}\int_0^{\pi}\frac{1}{2\omega}\Im\{\mathbf{E}_{\mathrm{p}}(\mathbf{H}_{\mathrm{p}}^*\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi = 0,\end{aligned} \tag{D15}$$

$$\begin{aligned}\lim_{r\to\infty}\int_0^{2\pi}\int_0^{\pi}\frac{1}{2\omega}\Im\{-(\mathbf{E}_{\mathrm{p}}\cdot\mathbf{H}_{\mathrm{p}}^*)\hat{\mathbf{n}}\}r^2\sin\theta d\theta d\phi &= \Im\int_0^{2\pi}\int_0^{\pi}\left\{-\frac{k^3}{32\pi^2\varepsilon_0}[(\hat{\mathbf{n}}\times\mathbf{p}^*)\cdot\mathbf{p}]\hat{\mathbf{n}}\right\}\sin\theta d\theta d\phi\\ &= -\frac{k^3}{24\pi^2\varepsilon_0}\Im\{\mathbf{p}^*\times\mathbf{p}\},\end{aligned} \tag{D16}$$

and we can get the analytical expression for $\mathbf{\Gamma}^s_{\mathrm{p,recoil}}$ as

$$\mathbf{\Gamma}^s_{\mathrm{p,recoil}} = -\frac{k^3}{24\pi^2\varepsilon_0}\Im\{\mathbf{p}^*\times\mathbf{p}\}. \tag{D17}$$

The optical torque on an electric dipole attributed to the orbital angular momentum flux can be calculated from

$$\begin{aligned}\mathbf{\Gamma}^o &= \int_0^{2\pi}\int_0^{\pi}(\langle\overleftrightarrow{\mathbf{M}}^o\rangle\cdot\hat{\mathbf{n}})r^2\sin\theta d\theta d\phi,\\ &= \int_0^{2\pi}\int_0^{\pi}\frac{-1}{4\omega}\Im\{\mathbf{E}_{\mathrm{tot}}(\mathbf{H}_{\mathrm{tot}}^*\cdot\hat{\mathbf{n}})+\mathbf{H}_{\mathrm{tot}}^*(\mathbf{E}_{\mathrm{tot}}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi\\ &\quad+\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{E}_{\mathrm{tot}}^*]\times\mathbf{H}_{\mathrm{tot}}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi\\ &\quad+\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_{\mathrm{tot}}]\times\mathbf{E}_{\mathrm{tot}}^*\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi.\end{aligned} \tag{D18}$$

In the following derivation, both expressions of ∇ in the Cartesian and spherical coordinates are applied such that

$$\begin{aligned}r\hat{\mathbf{n}}\times\nabla &= r(\hat{\mathbf{e}}_x n_x+\hat{\mathbf{e}}_y n_y+\hat{\mathbf{e}}_z n_z)\times\left(\hat{\mathbf{e}}_x\frac{\partial}{\partial_x}+\hat{\mathbf{e}}_y\frac{\partial}{\partial_y}+\hat{\mathbf{e}}_z\frac{\partial}{\partial_z}\right)\\ &= r\hat{\mathbf{r}}\times\nabla\\ &= \hat{\theta}\frac{\partial}{\partial_\theta}+\hat{\phi}\frac{1}{\sin\theta}\frac{\partial}{\partial_\phi}.\end{aligned} \tag{D19}$$

The OAM related torque on an electric dipole $\mathbf{\Gamma}^o_{\mathrm{p}}$ can be separated into three parts,

$$\mathbf{\Gamma}^o_{\mathrm{p}} = \mathbf{\Gamma}^o_{\mathrm{inc}}+\mathbf{\Gamma}^o_{\mathrm{p,mix}}+\mathbf{\Gamma}^o_{\mathrm{p,recoil}}, \tag{D20}$$

where $\boldsymbol{\Gamma}^o_{\rm inc}$ depends purely on the incident field,

$$\begin{aligned}\boldsymbol{\Gamma}^o_{\rm inc} &= \int_0^{2\pi}\int_0^{\pi}\frac{-1}{4\omega}\Im\{\mathbf{E}_{\rm inc}(\mathbf{H}^*_{\rm inc}\cdot\hat{\mathbf{n}})+\mathbf{H}^*_{\rm inc}(\mathbf{E}_{\rm inc}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi\\&+\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{E}^*_{\rm inc}]\times\mathbf{H}_{\rm inc}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi\\&+\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_{\rm inc}]\times\mathbf{E}^*_{\rm inc}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi,\end{aligned}\tag{D21}$$

$\boldsymbol{\Gamma}^o_{\rm p,mix}$ relies on the interference between the incident and radiation field,

$$\begin{aligned}\boldsymbol{\Gamma}^o_{\rm p,mix} &= \int_0^{2\pi}\int_0^{\pi}\frac{-1}{4\omega}\Im\{\mathbf{E}_{\rm inc}(\mathbf{H}^*_{\rm p}\cdot\hat{\mathbf{n}})+\mathbf{H}^*_{\rm p}(\mathbf{E}_{\rm inc}\cdot\hat{\mathbf{n}})+\mathbf{E}_{\rm p}(\mathbf{H}^*_{\rm inc}\cdot\hat{\mathbf{n}})+\mathbf{H}^*_{\rm inc}(\mathbf{E}_{\rm p}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi\\&+\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{E}^*_{\rm inc}]\times\mathbf{H}_{\rm p}+[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{E}^*_{\rm p}]\times\mathbf{H}_{\rm inc}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi\\&+\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_{\rm p}]\times\mathbf{E}^*_{\rm inc}+[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_{\rm inc}]\times\mathbf{E}^*_{\rm p}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi,\end{aligned}\tag{D22}$$

while $\boldsymbol{\Gamma}^o_{\rm p,recoil}$ is attributed to the radiation field only,

$$\begin{aligned}\boldsymbol{\Gamma}^o_{\rm p,recoil} &= \int_0^{2\pi}\int_0^{\pi}\frac{-1}{4\omega}\Im\{\mathbf{E}_{\rm p}(\mathbf{H}^*_{\rm p}\cdot\hat{\mathbf{n}})+\mathbf{H}^*_{\rm p}(\mathbf{E}_{\rm p}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi+\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{E}^*_{\rm p}]\times\mathbf{H}_{\rm p}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi\\&+\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_{\rm p}]\times\mathbf{E}^*_{\rm p}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi.\end{aligned}\tag{D23}$$

Due to the fact that the orbital angular momenta of the incident, radiation and total fields are separately conserved quantities, the corresponding contributions to the dipolar torque can be calculated independent of the radius of the enclosed surface,

$$\begin{aligned}\boldsymbol{\Gamma}^o_{\rm p} &= \lim_{r\to\infty}\boldsymbol{\Gamma}^o_{\rm p}(r)=\lim_{r\to 0}\boldsymbol{\Gamma}^o_{\rm p}(r),\\\boldsymbol{\Gamma}^o_{\rm inc} &= \lim_{r\to\infty}\boldsymbol{\Gamma}^o_{\rm inc}(r)=\lim_{r\to 0}\boldsymbol{\Gamma}^o_{\rm inc}(r),\\\boldsymbol{\Gamma}^o_{\rm p,recoil} &= \lim_{r\to\infty}\boldsymbol{\Gamma}^o_{\rm p,recoil}(r)=\lim_{r\to 0}\boldsymbol{\Gamma}^o_{\rm p,recoil}(r),\\\boldsymbol{\Gamma}^o_{\rm p,mix} &= \lim_{r\to\infty}\boldsymbol{\Gamma}^o_{\rm p,mix}(r)=\lim_{r\to 0}\boldsymbol{\Gamma}^o_{\rm p,mix}(r).\end{aligned}\tag{D24}$$

In the small r limit that $r\to 0$, using the approximation that $\mathbf{E}_{\rm inc}(\mathbf{r}')\approx\mathbf{E}_0+r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0$ and $\mathbf{H}_{\rm inc}(\mathbf{r}')\approx\mathbf{H}_0+r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0$, it is easy to prove that the optical torque component $\boldsymbol{\Gamma}^s_{\rm inc}$ attributed purely to the incident field is zero:

$$\begin{aligned}\boldsymbol{\Gamma}^o_{\rm inc} &= \lim_{r\to 0}r^2\int_0^{2\pi}\int_0^{\pi}\frac{-1}{4\omega}\Im\{\mathbf{E}_0(\mathbf{H}^*_0\cdot\hat{\mathbf{n}})+\mathbf{H}^*_0(\mathbf{E}_0\cdot\hat{\mathbf{n}})\}\sin\theta d\theta d\phi\\&+\lim_{r\to 0}r^2\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{E}^*_0]\times\mathbf{H}_0\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\&+\lim_{r\to 0}r^2\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_0]\times\mathbf{E}^*_0\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi,\\&=0.\end{aligned}\tag{D25}$$

The various components of $\boldsymbol{\Gamma}^o_{\rm p,mix}$ can be derived as follows. First of all, using the analytical expressions of integrals developed when deriving $\boldsymbol{\Gamma}^s_{\rm p,mix}$, one can easily arrive at the result that

$$\begin{aligned}&\int_0^{2\pi}\int_0^{\pi}\frac{-1}{4\omega}\Im\{\mathbf{E}_{\rm inc}(\mathbf{H}^*_{\rm p}\cdot\hat{\mathbf{n}})+\mathbf{H}^*_{\rm p}(\mathbf{E}_{\rm inc}\cdot\hat{\mathbf{n}})+\mathbf{E}_{\rm p}(\mathbf{H}^*_{\rm inc}\cdot\hat{\mathbf{n}})+\mathbf{H}^*_{\rm inc}(\mathbf{E}_{\rm p}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi\\&=\lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi}\frac{-1}{4\omega}\Im\{\mathbf{H}^*_{\rm p}(\mathbf{E}_{\rm inc}\cdot\hat{\mathbf{n}})+\mathbf{E}_{\rm p}(\mathbf{H}^*_{\rm inc}\cdot\hat{\mathbf{n}})+\mathbf{H}^*_{\rm inc}(\mathbf{E}_{\rm p}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi\\&=-\frac{2}{15}\Re\{\mathbf{p}\times\mathbf{E}^*_0\}-\frac{4}{15\omega\varepsilon_0}\Im\{(\mathbf{p}\cdot\nabla)\mathbf{H}^*_0\},\end{aligned}\tag{D26}$$

Using the dipolar radiation field properties in Eqs. (B1) and (B2) and the aforementioned integration properties of the unit vector $\hat{\mathbf{n}}$, one can arrive at the following result:

$$\begin{aligned}
&\lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{E}^*_{\mathrm{inc}}]\times\mathbf{H}_{\mathrm{p}}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi\\
&\quad=\lim_{r\to 0}\Im\int_0^{2\pi}\int_0^{\pi}\frac{i}{16\pi}\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{E}^*_{\mathrm{inc}}]\times(\hat{\mathbf{n}}\times\mathbf{p})\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&\quad=0.
\end{aligned}\tag{D27}$$

Using the dipolar radiation field properties in Eqs. (B1) and (B2), the aforementioned integration properties of the unit vector $\hat{\mathbf{n}}$, and the approximation of the incident field in the small r limit $\mathbf{E}_{\mathrm{inc}}(\mathbf{r}')\approx\mathbf{E}_0+r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0$ and $\mathbf{H}_{\mathrm{inc}}(\mathbf{r}')\approx\mathbf{H}_0+r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0$, one can derive that

$$\begin{aligned}
&\lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{E}^*_{\mathrm{p}}]\times\mathbf{H}_{\mathrm{inc}}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi\\
&=\Im\int_0^{2\pi}\int_0^{\pi}\frac{i3k}{16\pi\omega\varepsilon_0}\{\{(r\hat{\mathbf{n}}\times\nabla)\otimes[(\mathbf{p}^*\cdot\hat{\mathbf{n}})\hat{\mathbf{n}}]\}\times\mathbf{H}_0\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&\quad+\Im\int_0^{2\pi}\int_0^{\pi}\frac{3}{16\pi\omega\varepsilon_0}\{\{(r\hat{\mathbf{n}}\times\nabla)\otimes[(\mathbf{p}^*\cdot\hat{\mathbf{n}})\hat{\mathbf{n}}]\}\times[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0]\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&=\Im\int_0^{2\pi}\int_0^{\pi}\frac{3}{16\pi\omega\varepsilon_0}\{\{(r\hat{\mathbf{n}}\times\nabla)\otimes[(\mathbf{p}^*\cdot\hat{\mathbf{n}})\hat{\mathbf{n}}]\}\times[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0]\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&=\frac{1}{20}\Re\{\mathbf{p}^*\times\mathbf{E}_0\}-\frac{3}{20\omega\varepsilon_0}\Im\{(\mathbf{p}\cdot\nabla)\mathbf{H}^*_0\}.
\end{aligned}\tag{D28}$$

In the derivation of Eq. (D28), the following identity relations are applied:

$$\begin{aligned}
&(r\hat{\mathbf{n}}\times\nabla)\otimes[(\mathbf{p}^*\cdot\hat{\mathbf{n}})\hat{\mathbf{n}}]\\
&=\begin{pmatrix}
-p_y^*n_xn_z+p_z^*n_xn_y & -p_x^*n_xn_z-2p_y^*n_yn_z-p_z^*(n_z^2-n_y^2) & p_x^*n_xn_y+p_y^*(n_y^2-n_z^2)+2p_z^*n_yn_z\\
2p_x^*n_xn_z+p_y^*n_yn_z+p_z^*(n_z^2-n_x^2) & p_x^*n_yn_z-p_z^*n_xn_y & -p_x^*(n_x^2-n_z^2)-p_y^*n_xn_y-2p_z^*n_xn_z\\
-2p_x^*n_xn_y-p_y^*(n_y^2-n_x^2)-p_z^*n_yn_z & p_x^*(n_x^2-n_y^2)+2p_y^*n_xn_y+p_z^*n_xn_z & -p_x^*n_yn_z+p_y^*n_xn_z
\end{pmatrix}\\
&=\begin{pmatrix}
n_x[\hat{\mathbf{e}}_x\cdot(\hat{\mathbf{n}}\times\mathbf{p}^*)] & -n_z(\mathbf{p}^*\cdot\hat{\mathbf{n}})+n_y\big[\hat{\mathbf{e}}_x\cdot(\hat{\mathbf{n}}\times\mathbf{p}^*)\big] & n_y(\mathbf{p}^*\cdot\hat{\mathbf{n}})+n_z[\hat{\mathbf{e}}_x\cdot(\hat{\mathbf{n}}\times\mathbf{p}^*)]\\
n_z(\mathbf{p}^*\cdot\hat{\mathbf{n}})+n_x[\hat{\mathbf{e}}_y\cdot(\hat{\mathbf{n}}\times\mathbf{p}^*)] & n_y[\hat{\mathbf{e}}_y\cdot(\hat{\mathbf{n}}\times\mathbf{p}^*)] & -n_x(\mathbf{p}^*\cdot\hat{\mathbf{n}})+n_z[\hat{\mathbf{e}}_y\cdot(\hat{\mathbf{n}}\times\mathbf{p}^*)]\\
-n_y(\mathbf{p}^*\cdot\hat{\mathbf{n}})+n_x[\hat{\mathbf{e}}_z\cdot(\hat{\mathbf{n}}\times\mathbf{p}^*)] & n_x(\mathbf{p}^*\cdot\hat{\mathbf{n}})+n_y[\hat{\mathbf{e}}_z\cdot(\hat{\mathbf{n}}\times\mathbf{p}^*)] & n_z[\hat{\mathbf{e}}_z\cdot(\hat{\mathbf{n}}\times\mathbf{p}^*)]
\end{pmatrix}\\
&=(\mathbf{p}^*\cdot\hat{\mathbf{n}})[(r\hat{\mathbf{n}}\times\nabla)\otimes\hat{\mathbf{n}}]+(\hat{\mathbf{n}}\times\mathbf{p}^*)\otimes\hat{\mathbf{n}},
\end{aligned}\tag{D29}$$

and

$$[\overleftrightarrow{\mathbf{A}}\times\mathbf{a}]_{il}=\sum_{jk}\epsilon_{jkl}[\overleftrightarrow{\mathbf{A}}]_{ij}[\mathbf{a}]_k,\tag{D30}$$

where ϵ_{jkl} is the Levi-Civita symbol.

Using the dipolar radiation field properties in Eqs. (B1) and (B2), the aforementioned integration properties of the unit vector $\hat{\mathbf{n}}$, and the approximation of the incident field in the small r limit $\mathbf{E}_{\mathrm{inc}}(\mathbf{r}')\approx\mathbf{E}_0+r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0$ and $\mathbf{H}_{\mathrm{inc}}(\mathbf{r}')\approx\mathbf{H}_0+r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0$, one can derive that

$$\begin{aligned}
&\lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_{\mathrm{inc}}]\times\mathbf{E}^*_{\mathrm{p}}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi\\
&=\Im\int_0^{2\pi}\int_0^{\pi}\frac{-1}{16\pi\omega\varepsilon_0}\{[(\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_0]\times\mathbf{p}^*\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&\quad+\Im\int_0^{2\pi}\int_0^{\pi}\frac{3}{16\pi\omega\varepsilon_0}\{[(\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_0]\times[(\mathbf{p}^*\cdot\hat{\mathbf{n}})\hat{\mathbf{n}}]\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&=\Im\int_0^{2\pi}\int_0^{\pi}\frac{-1}{16\pi\omega\varepsilon_0}\{[(\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_0]\times\mathbf{p}^*\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&=-\frac{1}{12\omega\varepsilon_0}\Im\{(\mathbf{p}\cdot\nabla)\mathbf{H}^*_0\}.
\end{aligned}\tag{D31}$$

Using the dipolar radiation field properties in Eqs. (B1) and (B2), the aforementioned integration properties of the unit vector $\hat{\mathbf{n}}$, and the approximation of the incident field in the small r limit $\mathbf{E}_{\rm inc}(\mathbf{r}') \approx \mathbf{E}_0 + r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0$ and $\mathbf{H}_{\rm inc}(\mathbf{r}') \approx \mathbf{H}_0 + r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{H}_0$, one can derive that

$$\begin{aligned}
&\lim_{r\to 0}\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_{\rm p}]\times\mathbf{E}^*_{\rm inc}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi\\
&=\Im\int_0^{2\pi}\int_0^{\pi}\frac{i}{16\pi}\{[(r\hat{\mathbf{n}}\times\nabla)\otimes(\hat{\mathbf{n}}\times\mathbf{p})]\times\mathbf{E}_0^*\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&=\Im\int_0^{2\pi}\int_0^{\pi}\frac{i}{16\pi}\{\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\hat{\mathbf{n}}]\times\mathbf{p}\}\times\mathbf{E}_0^*\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&=\frac{1}{12}\Re\{\mathbf{p}^*\times\mathbf{E}_0\},
\end{aligned} \tag{D32}$$

where the following identity relation is applied in the derivation:

$$(r\hat{\mathbf{n}}\times\nabla)\otimes\hat{\mathbf{n}} = \begin{pmatrix} 0 & -n_z & n_y\\ n_z & 0 & -n_x\\ -n_y & n_x & 0\end{pmatrix}. \tag{D33}$$

From above results, we can get the analytical expression for $\boldsymbol{\Gamma}^o_{\rm p,mix}$

$$\boldsymbol{\Gamma}^o_{\rm p,mix} = -\frac{1}{2\omega\varepsilon_0}\Im[(\mathbf{p}\cdot\nabla)\mathbf{H}^*_{\rm inc}], \tag{D34}$$

The components of $\boldsymbol{\Gamma}^o_{\rm p,recoil}$ can be derived using the dipolar radiation field properties in Eqs. (B1) and (B2) and the aforementioned integration properties of the unit vector $\hat{\mathbf{n}}$ in the large r limit as $r\to\infty$,

$$\begin{aligned}
\int_0^{2\pi}\int_0^{\pi}\frac{-1}{4\omega}\Im\{\mathbf{E}_{\rm p}(\mathbf{H}^*_{\rm p}\cdot\hat{\mathbf{n}})+\mathbf{H}^*_{\rm p}(\mathbf{E}_{\rm p}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi &= \lim_{r\to\infty}\int_0^{2\pi}\int_0^{\pi}\frac{-1}{4\omega}\Im\{\mathbf{H}^*_{\rm p}(\mathbf{E}_{\rm p}\cdot\hat{\mathbf{n}})\}r^2\sin\theta d\theta d\phi\\
&=0,
\end{aligned} \tag{D35}$$

$$\begin{aligned}
&\lim_{r\to\infty}\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{E}^*_{\rm p}]\times\mathbf{H}_{\rm p}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi\\
&=\Im\int_0^{2\pi}\int_0^{\pi}\frac{k^3}{64\pi^2\varepsilon_0}\{\{(r\hat{\mathbf{n}}\times\nabla)\otimes[\mathbf{p}^*-(\mathbf{p}^*\cdot\hat{\mathbf{n}})\hat{\mathbf{n}}]\}\times(\hat{\mathbf{n}}\times\mathbf{p})\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&=\Im\int_0^{2\pi}\int_0^{\pi}\frac{k^3}{64\pi^2\varepsilon_0}\{\{(r\hat{\mathbf{n}}\times\nabla)\otimes[-(\mathbf{p}^*\cdot\hat{\mathbf{n}})\hat{\mathbf{n}}]\}\times(\hat{\mathbf{n}}\times\mathbf{p})\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&=-\frac{k^3}{48\pi^2\varepsilon_0}\Im(\mathbf{p}^*\times\mathbf{p}),
\end{aligned} \tag{D36}$$

$$\begin{aligned}
&\lim_{r\to\infty}\int_0^{2\pi}\int_0^{\pi}\frac{1}{4\omega}\Im\{[(r\hat{\mathbf{n}}\times\nabla)\otimes\mathbf{H}_{\rm p}]\times\mathbf{E}^*_{\rm p}\}\cdot\hat{\mathbf{n}}r^2\sin\theta d\theta d\phi\\
&=\Im\int_0^{2\pi}\int_0^{\pi}\frac{k^3}{64\pi^2\varepsilon_0}\{\{(r\hat{\mathbf{n}}\times\nabla)\otimes(\hat{\mathbf{n}}\times\mathbf{p})\}\times[\mathbf{p}^*-(\mathbf{p}^*\cdot\hat{\mathbf{n}})\hat{\mathbf{n}}]\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&=\Im\int_0^{2\pi}\int_0^{\pi}\frac{k^3}{64\pi^2\varepsilon_0}\{\{(r\hat{\mathbf{n}}\times\nabla)\otimes(\hat{\mathbf{n}}\times\mathbf{p})\}\times\mathbf{p}^*\}\cdot\hat{\mathbf{n}}\sin\theta d\theta d\phi\\
&=-\frac{k^3}{48\pi^2\varepsilon_0}\Im(\mathbf{p}^*\times\mathbf{p}),
\end{aligned} \tag{D37}$$

and we can get the analytical expression for $\boldsymbol{\Gamma}^o_{\rm p,recoil}$ as

$$\boldsymbol{\Gamma}^o_{\rm p,recoil} = -\frac{k^3}{24\pi^2\varepsilon_0}\Im\{\mathbf{p}^*\times\mathbf{p}\}. \tag{D38}$$

APPENDIX E: DERIVATION OF THE ANALYTICAL EXPRESSION OF THE OPTICAL TORQUE ON AN INDUCED ELECTRIC QUADRUPOLE

In this section, a detailed derivation for the analytical expression of the optical quadrupolar torque $\boldsymbol{\Gamma}_{\rm Qe}$ is provided. $\boldsymbol{\Gamma}_{\rm Qe}$ is derived in the same way as the dipolar torques using the total angular momentum flux,

$$\boldsymbol{\Gamma} = \Re \int_0^{2\pi} \int_0^{\pi} (r\hat{\mathbf{n}}) \times \left\{ \frac{\varepsilon_0}{2} \mathbf{E}_{\rm tot}(\mathbf{E}^*_{\rm tot} \cdot \hat{\mathbf{n}}) + \frac{\mu_0}{2} \mathbf{H}_{\rm tot}(\mathbf{H}^*_{\rm tot} \cdot \hat{\mathbf{n}}) \right\} r^2 \sin\theta d\theta d\phi, \tag{E1}$$

where we only consider the torque contributed by the induced electric quadrupole and the incident field so that $\mathbf{E}_{\rm tot}(\mathbf{r}') = \mathbf{E}_{\rm inc}(\mathbf{r}') + \mathbf{E}_{\rm Qe}(\mathbf{r}')$ and $\mathbf{H}_{\rm tot}(\mathbf{r}') = \mathbf{H}_{\rm inc}(\mathbf{r}') + \mathbf{H}_{\rm Qe}(\mathbf{r}')$.

Like the dipolar torque, $\boldsymbol{\Gamma}_{\rm Qe}$ attributed to the interaction between the induced electric quadrupole and incident field can be decomposed into different parts,

$$\boldsymbol{\Gamma}_{\rm Qe} = \boldsymbol{\Gamma}_{\rm inc} + \boldsymbol{\Gamma}_{\rm Qe,mix} + \boldsymbol{\Gamma}_{\rm Qe,recoil}. \tag{E2}$$

As known in previous sections, the torque component purely dependent on the incident field does not contribute to the total quadrupolar torque,

$$\boldsymbol{\Gamma}_{\rm inc} = 0, \tag{E3}$$

while the extinction torque $\boldsymbol{\Gamma}_{\rm Qe,mix}$, as a result of the interference between incident and radiation fields, can be separated into different components as

$$\begin{aligned}\boldsymbol{\Gamma}_{\rm Qe,mix} = {}& \frac{\varepsilon_0 r^3}{2}\Re \int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{E}_{\rm inc})(\mathbf{E}^*_{\rm Qe} \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi + \frac{\varepsilon_0 r^3}{2}\Re \int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{E}_{\rm Qe})(\mathbf{E}^*_{\rm inc} \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi \\ &+ \frac{\mu_0 r^3}{2}\Re \int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_{\rm inc})(\mathbf{H}^*_{\rm Qe} \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi + \frac{\mu_0 r^3}{2}\Re \int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_{\rm Qe})(\mathbf{H}^*_{\rm inc} \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi\end{aligned} \tag{E4}$$

and the recoil torque $\boldsymbol{\Gamma}_{\rm Qe,recoil}$ as a result of self-interaction of the induced electric quadrupole is expressed as

$$\boldsymbol{\Gamma}_{\rm Qe,recoil} = \frac{\varepsilon_0 r^3}{2}\Re \int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{E}_{\rm Qe})(\mathbf{E}^*_{\rm Qe} \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi + \frac{\mu_0 r^3}{2}\Re \int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_{\rm Qe})(\mathbf{H}^*_{\rm Qe} \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi. \tag{E5}$$

Due to the fact that the angular momenta of the incident, radiation, and total fields are separately conserved quantities, the corresponding contributions to the dipolar torque can be calculated independent of the radius of the enclose surface,

$$\begin{aligned}\boldsymbol{\Gamma}_{\rm Qe} &= \lim_{r\to\infty} \boldsymbol{\Gamma}_{\rm Qe}(r) = \lim_{r\to 0} \boldsymbol{\Gamma}_{\rm Qe}(r),\\ \boldsymbol{\Gamma}_{\rm Qe,recoil} &= \lim_{r\to\infty} \boldsymbol{\Gamma}_{\rm Qe,recoil}(r) = \lim_{r\to 0} \boldsymbol{\Gamma}_{\rm Qe,recoil}(r),\\ \boldsymbol{\Gamma}_{\rm Qe,mix} &= \lim_{r\to\infty} \boldsymbol{\Gamma}_{\rm Qe,mix}(r) = \lim_{r\to 0} \boldsymbol{\Gamma}_{\rm Qe,mix}(r).\end{aligned} \tag{E6}$$

It follows from Eqs. (B3) and (B4) that

$$\mathbf{H}^*_{\rm Qe} \cdot \hat{\mathbf{n}} = 0. \tag{E7}$$

It is thus easy to prove that

$$\begin{aligned}\frac{\mu_0 r^3}{2}\Re \int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_{\rm inc})(\mathbf{H}^*_{\rm Qe} \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi = 0,\\ \frac{\mu_0 r^3}{2}\Re \int_0^{2\pi}\int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_{\rm Qe})(\mathbf{H}^*_{\rm Qe} \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi = 0.\end{aligned} \tag{E8}$$

The analytical expressions of the remaining nonzero terms of $\boldsymbol{\Gamma}_{\rm Qe}$ can be derived as follows.

It follows from Eqs. (B3) and (B4) that

$$\mathbf{E}^*_{\rm Qe} \cdot \hat{\mathbf{n}} = \frac{-ik^3 e^{-ikr}}{24\pi\varepsilon_0 r}\left[\frac{-3i}{kr} + \frac{9}{(kr)^2} + \frac{9i}{(kr)^3}\right]\left[(\overleftrightarrow{\mathbf{Q}}^{\rm e*} \cdot \hat{\mathbf{n}}) \cdot \hat{\mathbf{n}}\right]. \tag{E9}$$

Using the aforementioned properties of $\hat{\mathbf{n}}$ and the approximation that $\mathbf{E}_{\rm inc}(\mathbf{r}) \approx \mathbf{E}_0 + r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0$, we can derive that

$$\begin{aligned}
&\lim_{r\to 0}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\rm inc})(\mathbf{E}_{\rm Qe}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\
&=\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_0)\times\frac{-ik^3}{24\pi\varepsilon_0 r}\left[\frac{9}{(kr)^2}\right]\left[(\overleftrightarrow{\mathbf{Q}}^{\rm e*}\cdot\hat{\mathbf{n}})\cdot\hat{\mathbf{n}}\right]\sin\theta d\theta d\phi\\
&\quad+\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}\{\hat{\mathbf{n}}\times[(r\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0]\}\frac{-ik^3}{24\pi\varepsilon_0 r}\left[\frac{9i}{(kr)^3}\right]\left[(\overleftrightarrow{\mathbf{Q}}^{\rm e*}\cdot\hat{\mathbf{n}})\cdot\hat{\mathbf{n}}\right]\sin\theta d\theta d\phi\\
&=\Re\frac{-3ik}{16\pi}\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_0)[(\overleftrightarrow{\mathbf{Q}}^{\rm e*}\cdot\hat{\mathbf{n}})\cdot\hat{\mathbf{n}}]\sin\theta d\theta d\phi+\Re\frac{3}{16\pi}\int_0^{2\pi}\int_0^{\pi}\{\hat{\mathbf{n}}\times[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0]\}[(\overleftrightarrow{\mathbf{Q}}^{\rm e*}\cdot\hat{\mathbf{n}})\cdot\hat{\mathbf{n}}]\sin\theta d\theta d\phi\\
&=\Re\frac{3}{16\pi}\int_0^{2\pi}\int_0^{\pi}\{\hat{\mathbf{n}}\times[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0]\}[(\overleftrightarrow{\mathbf{Q}}^{\rm e*}\cdot\hat{\mathbf{n}})\cdot\hat{\mathbf{n}}]\sin\theta d\theta d\phi\\
&=\Re\left\{\frac{ikZ_0}{10}\mathbf{H}_0\left(\sum_{u=x,y,z}Q_{uu}^{e*}\right)\right\}-\Re\left\{\frac{ikZ_0}{20}(\overleftrightarrow{\mathbf{Q}}^{\rm e*}\cdot\mathbf{H}_0)\right\}+\frac{1}{10}\Re\left\{\sum_{u=x,y,z}\mathbf{Q}_u^{\rm e*}\times\mathbf{D}_u^{\rm e}\right\},
\end{aligned}\tag{E10}$$

where we introduce the notation that $\mathbf{D}_u^e=\sum_{v=x,y,z}\hat{\mathbf{e}}_v[\overleftrightarrow{\mathcal{D}^e}]_{uv}$ and $\mathbf{Q}_u^e=\sum_{v=x,y,z}\hat{\mathbf{e}}_v[\overleftrightarrow{\mathbf{Q}^e}]_{uv}$

For an induced electric quadrupole moment in an isotropic Mie particle in which $\mathbf{Q}_u^e=\alpha_{\rm Qe}\mathbf{D}_u^e$, using the relations in Eqs. (A2) and (A3) so that $\sum_{u=x,y,z}Q_{uu}^{e*}=0$ and the fact that $\mathbf{D}_u^{\rm e*}\times\mathbf{D}_u^{\rm e}$ are purely imaginary, Eq. (E10) is reduced to

$$\lim_{r\to 0}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\rm inc})(\mathbf{E}_{\rm Qe}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi=\Re\left\{\frac{-ikZ_0}{20}(\overleftrightarrow{\mathbf{Q}}^{\rm e*}\cdot\mathbf{H}_0)\right\}+\frac{\Im\{\alpha_{\rm Qe}\}}{10}\sum_{u=x,y,z}\Im\left\{\mathbf{D}_u^{\rm e*}\times\mathbf{D}_u^{\rm e}\right\}.\tag{E11}$$

It follows from Eqs. (B3) and (B4) that

$$\hat{\mathbf{n}}\times\mathbf{E}_{\rm Qe}=\frac{ik^3e^{ikr}}{24\pi\varepsilon_0 r}\left[-1-\frac{3i}{kr}+\frac{6}{(kr)^2}+\frac{6i}{(kr)^3}\right][\hat{\mathbf{n}}\times(\overleftrightarrow{\mathbf{Q}}^{\rm e}\cdot\hat{\mathbf{n}})],\tag{E12}$$

and using the aforementioned properties of $\hat{\mathbf{n}}$ and the approximation that $\mathbf{E}_{\rm inc}(\mathbf{r}) \approx \mathbf{E}_0 + r(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0$, we can derive that

$$\begin{aligned}
&\lim_{r\to 0}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\rm Qe})(\mathbf{E}_{\rm inc}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\
&=\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}\frac{ik^3}{24\pi\varepsilon_0 r}\left[\frac{6}{(kr)^2}\right][\hat{\mathbf{n}}\times(\overleftrightarrow{\mathbf{Q}}^{\rm e}\cdot\hat{\mathbf{n}})](\mathbf{E}_0^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi\\
&\quad+\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}\frac{ik^3}{24\pi\varepsilon_0 r}\left[\frac{6i}{(kr)^3}\right][\hat{\mathbf{n}}\times(\overleftrightarrow{\mathbf{Q}}^{\rm e}\cdot\hat{\mathbf{n}})]\{[(r\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0^*]\cdot\hat{\mathbf{n}}\}\sin\theta d\theta d\phi\\
&=\Re\frac{-1}{8\pi}\int_0^{2\pi}\int_0^{\pi}[\hat{\mathbf{n}}\times(\overleftrightarrow{\mathbf{Q}}^{\rm e}\cdot\hat{\mathbf{n}})]\{[(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E}_0^*]\cdot\hat{\mathbf{n}}\}\sin\theta d\theta d\phi\\
&=\frac{1}{15}\Re\left\{\sum_{u=x,y,z}(\overleftrightarrow{\mathbf{Q}}^{\rm e*}\cdot\hat{\mathbf{e}}_u)\times(\overleftrightarrow{\mathbf{D}}^{\rm e}\cdot\hat{\mathbf{e}}_u)\right\}\\
&=\frac{1}{15}\Re\left\{\sum_{u=x,y,z}\mathbf{Q}_u^{\rm e*}\times\mathbf{D}_u^{\rm e}\right\}.
\end{aligned}\tag{E13}$$

For an induced electric quadrupole moment in an isotropic Mie particle, this is equivalent to

$$\begin{aligned}
\lim_{r\to 0}\frac{\varepsilon_0 r^3}{2}\Re\int_0^{2\pi}\int_0^{\pi}(\hat{\mathbf{n}}\times\mathbf{E}_{\rm Qe})(\mathbf{E}_{\rm inc}^*\cdot\hat{\mathbf{n}})\sin\theta d\theta d\phi&=\frac{1}{15}\Re\left\{\sum_{u=x,y,z}\mathbf{Q}_u^{\rm e*}\times\mathbf{D}_u^{\rm e}\right\}\\
&=\frac{\Im\{\alpha_{\rm Qe}\}}{15}\sum_{u=x,y,z}\Im\left\{\mathbf{D}_u^{\rm e*}\times\mathbf{D}_u^{\rm e}\right\}\\
&=\frac{\Im\{\alpha_{\rm Qe}\}}{15}\Im\left\{\sum_j\hat{\mathbf{e}}_j\sum_l\sum_u\sum_v\epsilon_{jlv}[\overleftrightarrow{\mathcal{D}^{\rm e}}]_{lu}^*[\overleftrightarrow{\mathcal{D}^{\rm e}}]_{uv}\right\}.
\end{aligned}\tag{E14}$$

It follows from Eqs. (B3) and (B4) that

$$\hat{\mathbf{n}} \times \mathbf{H}_{\mathrm{Qe}} = \frac{-ik^3 c_0 e^{ikr}}{24\pi r}\left[1 + \frac{3i}{kr} - \frac{3}{(kr)^2}\right]\{\hat{\mathbf{n}} \times [\hat{\mathbf{n}} \times (\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}} \cdot \hat{\mathbf{n}})]\}, \tag{E15}$$

using the aforementioned properties of $\hat{\mathbf{n}}$ and the approximation that $\mathbf{H}_{\mathrm{inc}}^*(\mathbf{r}) \approx \mathbf{H}_0^* + r(\hat{\mathbf{n}} \cdot \nabla)\mathbf{H}_0^*$, we can derive that in the small r limit,

$$\lim_{r\to 0} \frac{\mu_0 r^4}{2} \Re \int_0^{2\pi} \int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_{\mathrm{Qe}})\{[(\hat{\mathbf{n}} \cdot \nabla)\mathbf{H}_0^*] \cdot \hat{\mathbf{n}}\} \sin\theta d\theta d\phi = 0, \tag{E16}$$

$$\begin{aligned}
\lim_{r\to 0} \frac{\mu_0 r^3}{2} \Re \int_0^{2\pi} \int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_{\mathrm{Qe}})(\mathbf{H}_{\mathrm{inc}}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi &= \Re \frac{ikc_0\mu_0}{16\pi} \int_0^{2\pi} \int_0^{\pi} \{\hat{\mathbf{n}} \times [\hat{\mathbf{n}} \times (\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}} \cdot \hat{\mathbf{n}})]\}(\mathbf{H}_0^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi \\
&= \Re\left\{\frac{ikZ_0}{60}\mathbf{H}_0^*\left(\sum_{u=x,y,z} Q_{uu}^{\mathrm{e}}\right)\right\} - \Re\left\{\frac{ikZ_0}{20}(\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}} \cdot \mathbf{H}_0^*)\right\} \\
&= \Re\left\{\frac{-ikZ_0}{60}\mathbf{H}_0\left(\sum_{u=x,y,z} Q_{uu}^{\mathrm{e}*}\right)\right\} + \Re\left\{\frac{ikZ_0}{20}(\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}*} \cdot \mathbf{H}_0)\right\}.
\end{aligned} \tag{E17}$$

For an induced electric quadrupole moment in an isotropic Mie particle, this reduces to

$$\lim_{r\to 0} \frac{\mu_0 r^3}{2} \Re \int_0^{2\pi} \int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_{\mathrm{Qe}})(\mathbf{H}_{\mathrm{inc}}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi = \Re\left\{\frac{ikZ_0}{20}(\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}*} \cdot \mathbf{H}_0)\right\}. \tag{E18}$$

To summarize the above results, the analytical expression for $\mathbf{\Gamma}_{\mathrm{Qe,mix}}$ is

$$\begin{aligned}
\mathbf{\Gamma}_{\mathrm{Qe,mix}} &= \lim_{r\to 0} \frac{\varepsilon_0 r^3}{2} \Re \int_0^{2\pi} \int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{E}_{\mathrm{inc}})(\mathbf{E}_{\mathrm{Qe}}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi + \lim_{r\to 0} \frac{\varepsilon_0 r^3}{2} \Re \int_0^{2\pi} \int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{E}_{\mathrm{Qe}})(\mathbf{E}_{\mathrm{inc}}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi \\
&\quad + \lim_{r\to 0} \frac{\mu_0 r^3}{2} \Re \int_0^{2\pi} \int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{H}_{\mathrm{Qe}})(\mathbf{H}_{\mathrm{inc}}^* \cdot \hat{\mathbf{n}}) \sin\theta d\theta d\phi \\
&= \Re\left\{\frac{ikZ_0}{12}\mathbf{H}_0\left(\sum_{u=x,y,z} Q_{uu}^{\mathrm{e}*}\right)\right\} + \frac{1}{6}\Re\left\{\sum_{u=x,y,z} \mathbf{Q}_u^{\mathrm{e}*} \times \mathbf{D}_u^{\mathrm{e}}\right\}.
\end{aligned} \tag{E19}$$

For an induced electric quadrupole moment in an isotropic Mie particle, this is equivalent to

$$\begin{aligned}
\mathbf{\Gamma}_{\mathrm{Qe,mix}} &= \frac{\Im\{\alpha_{\mathrm{Qe}}\}}{6} \Im\left\{\sum_{u=x,y,z} \mathbf{D}_u^{\mathrm{e}*} \times \mathbf{D}_u^{\mathrm{e}}\right\} = \frac{120\pi\varepsilon_0}{k^5}\Re(a_2)\mathbf{s}^{\mathrm{Qe}}, \\
\mathbf{s}^{\mathrm{Qe}} &= \frac{\varepsilon_0}{6} \Im\left\{\sum_{u=x,y,z} \mathbf{D}_u^{\mathrm{e}*} \times \mathbf{D}_u^{\mathrm{e}}\right\}.
\end{aligned} \tag{E20}$$

The last nonzero term contributing to the electric quadrupolar torque is the "recoil" torque $\mathbf{\Gamma}_{\mathrm{p,recoil}}$, which can be derived using the dipolar radiation field properties in Eq. (B3), Eq. (B4), Eq. (A3), and the aforementioned integration properties of the unit vector $\hat{\mathbf{n}}$ in the large r limit as $r \to \infty$,

$$\begin{aligned}
\mathbf{\Gamma}_{\mathrm{Qe,recoil}} &= \lim_{r\to\infty} \frac{\varepsilon_0 r^3}{2} \Re \int_0^{2\pi} \int_0^{\pi} (\hat{\mathbf{n}} \times \mathbf{E}_{\mathrm{Qe}})\big(\mathbf{E}_{\mathrm{Qe}}^* \cdot \hat{\mathbf{n}}\big) \sin\theta d\theta d\phi \\
&= \lim_{r\to\infty} \Re \frac{3ik^5}{2\varepsilon_0 \cdot (24\pi)^2}\left[1 + \frac{3i}{(kr)^3} + \frac{18i}{(kr)^5}\right] \int_0^{2\pi} \int_0^{\pi} [\hat{\mathbf{n}} \times (\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}} \cdot \hat{\mathbf{n}})][(\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}*} \cdot \hat{\mathbf{n}}) \cdot \hat{\mathbf{n}}] \sin\theta d\theta d\phi \\
&= \Re \frac{3ik^5}{2\varepsilon_0 \cdot (24\pi)^2}[1] \int_0^{2\pi} \int_0^{\pi} [\hat{\mathbf{n}} \times (\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}} \cdot \hat{\mathbf{n}})][(\overleftrightarrow{\mathbf{Q}}^{\mathrm{e}*} \cdot \hat{\mathbf{n}}) \cdot \hat{\mathbf{n}}] \sin\theta d\theta d\phi \\
&= \frac{3ik^5}{2\varepsilon_0 \cdot (24\pi)^2}\left[\frac{-i8\pi}{15}\right] \sum_j \hat{\mathbf{e}}_j \sum_l \sum_u \sum_v \Im\{\epsilon_{lvj} Q_{lu}^{\mathrm{e}} Q_{uv}^{\mathrm{e}*}\} \\
&= -\frac{k^5}{720\pi\varepsilon_0} \Im\left\{\sum_{u=x,y,z} \mathbf{Q}_u^{\mathrm{e}*} \times \mathbf{Q}_u^{\mathrm{e}}\right\}.
\end{aligned} \tag{E21}$$

For an induced electric quadrupole moment in an isotropic Mie particle, this is equivalent to

$$\begin{aligned}\boldsymbol{\Gamma}_{\text{Qe,recoil}} &= = -\frac{k^5}{720\pi\varepsilon_0}\Im\left\{\sum_{u=x,y,z}\mathbf{Q}_u^{e*}\times\mathbf{Q}_u^{e}\right\}\\ &= -\frac{k^5}{720\pi\varepsilon_0}\Im\left\{|\alpha_{\text{Qe}}|^2\sum_{u=x,y,z}\mathbf{D}_u^{e*}\times\mathbf{D}_u^{e}\right\}\\ &= -\frac{120\pi\varepsilon_0}{k^5}|a_2|^2\mathbf{s}^{\text{Qe}}.\end{aligned} \tag{E22}$$

To summarize above results, the torque acting on the isotropic Mie particle that is attributed to the induced electric quadrupole can be written analytically as

$$\boldsymbol{\Gamma}_{\text{Qe}} = \boldsymbol{\Gamma}_{\text{Qe,mix}} + \boldsymbol{\Gamma}_{\text{Qe,recoil}} = \frac{120\pi\varepsilon_0}{k^5}[\Re(a_2) - |a_2|^2]\mathbf{s}^{\text{Qe}}. \tag{E23}$$

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