Optical theorem and multipole scattering of light by arbitrarily shaped nanoparticles

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The application of Cartesian multipoles in irreducible representations provides the possibility to get explicit contributions of the toroidal multipole terms in the extinction and scattering power without the introduction of special form factors. In the framework of the Cartesian multipoles, we obtained multipole decomposition (up to the third order) of the induced polarization (current) inside an arbitrarily shaped scatterer (nanoparticle). The third-order decomposition includes the toroidal dipole, magnetic quadrupole, electric octupole terms, and also nonradiating terms. The corresponding multipole decomposition of the scattering cross section, taking into account the electric octupole term, is derived and compared with the multipole decomposition of the extinction cross section obtained using the optical theorem. We show that the role of multipoles in the optical theorem (light extinction) and scattering by arbitrarily shaped nanoparticles can be different. This can result in seemingly paradoxical conclusions with respect to the appearance of multipole contributions in the scattering and extinction cross sections. This fact is especially important for absorptionless nanoparticles, for which the scattering cross section can be calculated using the optical theorem, because in this case extinction is solely determined by scattering. Demonstrative results concerning the role of third-order multipoles in the resonant optical response of high-refractive-index dielectric nanodisks, with and without a through hole at the center, are presented. It is shown that the optical theorem results in a negligible role of the third-order multipoles in the extinction cross sections, whereas these multipoles provide the main contribution in the scattering cross sections.

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I. INTRODUCTION

The well-known optical theorem relates the light extinction power, including absorption and scattering, to the imaginary part of the light scattering amplitude in the forward direction [1]. This theorem provides a simple algorithm for calculations of the extinction cross sections σ_{ext} by nanoparticles and nanostructures [2],

$$\sigma_{\text{ext}} = \frac{4\pi}{k_d|\mathbf{E}_0|^2}\text{Im}\{\mathbf{E}_0^* \cdot \mathbf{E}_0^{\text{sca}}(\mathbf{n}_{\text{inc}})\}, \quad (1)$$

where k_d is the wave number in the surrounding medium, $\mathbf{E}_0$ is the incident wave amplitude, $\mathbf{E}_0^{\text{sca}}(\mathbf{n}_{\text{inc}})$ is the scattering amplitude in the forward (incident) direction, $\mathbf{n}_{\text{inc}}$ is the unit vector directed along the incidence, and the asterisk denotes complex conjugation. For absorptionless scatterers the scattering cross section σ_{sca} also can be calculated using the optical theorem [3]. In general, a combination of σ_{ext} and the scattering cross section σ_{sca} with the multipole decomposition method, as it was first realized in the Mie theory, provides an extremely useful approach to analyze and understand light scattering processes [4]. This approach is especially important for investigations of resonant optical properties of nanoparticles, where their response (scattered field phase and directivity) is determined by certain multipole modes resonantly excited by incident light. In the Mie theory, derived for spherical scatterers, the multipole decompositions of σ_{ext} and σ_{sca} are obtained from the field expansion including only spherical electric a_E and magnetic a_M multipole coefficients which are associated with the scattered field structure. Another approach is based on the Cartesian multipole expansion of current induced in scatterers by incident light. In this case, depending on the definition of the multipoles, the multipole expansion can include contributions from the electric, magnetic, and toroidal terms [5]. A general approach taking into account all types of multipoles is based on the application of three families of electric, magnetic, and toroidal multipole form factors [6,7]. Recently, the electromagnetic excitation of dynamical toroidal multipoles (especially toroidal dipoles) has attracted significant attention due to their unusual electromagnetic properties [8]. In particular, the excitation of the toroidal dipole in a scatterer is a key condition for the realization of the anapole (nonradiating) mode, which can provide unique possibilities for the development of nonscattering (transparent) metamaterials [9,10]. The anapole mode can be viewed as a combination of electric and toroidal dipole moments, resulting in the destructive interference of radiation fields due to the similarity of their scattering patterns [9,11–14]. The scattered field patterns of other toroidal multipoles are also similar to corresponding electric and magnetic multipoles. This is the reason why in the Mie theory no explicit contributions of toroidal multipoles are present. Note that the toroidal dipole moment appears in the third-order multipole expansion and is associated with particular current configurations [5]. The multipole decomposition method involving the toroidal multipole terms provides important information about the influence of different current configurations on the spectral features in the scattering and extinction cross sections.

In this paper, we show how the optical theorem can be combined with multipole decomposition for arbitrarily shaped (nonspherical) nanoparticles and discuss the differences between multipole contributions in the extinction and scattering cross sections. For this aim, we perform multipole

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decompositions of $\sigma_{\rm sca}$ and $\sigma_{\rm ext}$ up to the third order using irreducible multipole representations in the Cartesian coordinate system [15]. It will be shown that the role of multipoles in the optical theorem (light extinction) and scattering by arbitrarily shaped nanoparticles can be different. This can result in seemingly paradoxical conclusions with respect to the appearance of multipole contributions in the scattering and extinction cross sections. This conclusion is especially important for absorptionless nanoparticles, for which the scattering cross section can be calculated by using the optical theorem, because in this case extinction is solely determined by scattering. Demonstrative results concerning the role of third-order multipoles in the resonant optical response of high-refractive-index dielectric nanodisks, with and without a through hole at the center, are presented.

II. MULTIPOLE APPROACH

We start from the multipole decomposition of polarization $\mathbf{P} = \varepsilon_0(\varepsilon_p - \varepsilon_d)\mathbf{E}$, induced by the incident light wave in an arbitrarily shaped nanoparticle, where ε_0, ε_p, and ε_d are the vacuum dielectric constant, relative dielectric permittivity of the nanoparticle, and relative dielectric permittivity of the surrounding medium, respectively, $\mathbf{E}$ is the total electric field inside the nanoparticle. We consider incident plane monochromatic waves with the time dependence defined by $\exp(-i\omega t)$, where ω is the angular frequency, and multipoles located at the origin of the Cartesian coordinate system coinciding with the nanoparticle center of mass.

A. Two multipole decompositions of induced polarization

To get multipole decomposition of the light-induced polarization,

$$\mathbf{P}(\mathbf{r}) = \int \mathbf{P}(\mathbf{r}')\delta(\mathbf{r}-\mathbf{r}')d\mathbf{r}', \tag{2}$$

the Dirac delta function $\delta(\mathbf{r}-\mathbf{r}')$ in (2) is expanded in a Taylor series [16] with respect to $\mathbf{r}'$ around the origin, and then, using the definitions of the corresponding multipole moments, one can write (see Ref. [17])

$$\mathbf{P}(\mathbf{r}) \simeq \mathbf{p}\delta(\mathbf{r}) - \frac{1}{6}\hat{Q}'\nabla\delta(\mathbf{r}) + \frac{i}{\omega}[\nabla\times\mathbf{m}\delta(\mathbf{r})] + \frac{1}{6}\hat{O}'[\nabla\nabla\delta(\mathbf{r})] - \frac{i}{2\omega}[\nabla\times\hat{M}'\nabla\delta(\mathbf{r})], \tag{3}$$

where

$$\mathbf{p} = \int \mathbf{P}(\mathbf{r}')d\mathbf{r}' \tag{4}$$

is the electric dipole moment, and

$$\hat{Q}' = 3\int[\mathbf{r}'\mathbf{P}(\mathbf{r}') + \mathbf{P}(\mathbf{r}')\mathbf{r}']d\mathbf{r}', \tag{5}$$

$$\mathbf{m} = -\frac{i\omega}{2}\int[\mathbf{r}'\times\mathbf{P}(\mathbf{r}')]d\mathbf{r}', \tag{6}$$

$$\hat{O}' = -\int \mathbf{r}'\mathbf{r}'\mathbf{r}'[\nabla\cdot\mathbf{P}(\mathbf{r}')]d\mathbf{r}' \equiv \int\{\mathbf{P}(\mathbf{r}')\mathbf{r}'\mathbf{r}' + \mathbf{r}'\mathbf{P}(\mathbf{r}')\mathbf{r}' + \mathbf{r}'\mathbf{r}'\mathbf{P}(\mathbf{r}')\}d\mathbf{r}', \tag{7}$$

$$\hat{M}' = -\frac{2i\omega}{3}\int[\mathbf{r}'\times\mathbf{P}(\mathbf{r}')]\mathbf{r}'d\mathbf{r}' \tag{8}$$

are the electric quadrupole tensor, the magnetic dipole moment, the tensor of the electric octupole moment, and the tensor of the magnetic quadrupole moment, respectively, at the origin of the coordinate system. Here $\nabla\nabla$, $\mathbf{r}'\mathbf{P}$, and $\mathbf{r}'\mathbf{r}'\mathbf{r}'$ represent the tensor products between corresponding vectors. Note that the electric quadrupole tensor $\hat{Q}'$ and electric octupole tensor $\hat{O}'$ are totally symmetric and not traceless, whereas the magnetic quadrupole tensor $\hat{M}'$ is traceless and not symmetric. Integration in the multipole definitions is performed over the scatterer volume.

Another approach is connected with using the multipole moments in the irreducible representations. It means that they have to satisfy both symmetric and traceless properties. Applying the traceless procedure to $\hat{Q}'$ and $\hat{O}'$, and the symmetrization procedure to $\hat{M}'$, as it has been described in Ref. [5], one can write the following:

(1)

$$\hat{Q}' = \hat{Q} + \hat{Q}'', \tag{9}$$

where

$$\hat{Q} = 3\int\left[\mathbf{r}'\mathbf{P}(\mathbf{r}') + \mathbf{P}(\mathbf{r}')\mathbf{r}' - \frac{2}{3}[\mathbf{r}'\cdot\mathbf{P}(\mathbf{r}')]\hat{U}\right]d\mathbf{r}' \tag{10}$$

is the irreducible tensor of the electric quadrupole moment ($\hat{U}$ is the 3×3 unit tensor), and

$$\hat{Q}'' = 2\int[\mathbf{r}'\cdot\mathbf{P}(\mathbf{r}')]\hat{U}d\mathbf{r}'. \tag{11}$$

(2)

$$\hat{O}' = \hat{O} + \hat{O}'', \tag{12}$$

where $\hat{O}$ is the irreducible tensor of the electric octupole moment with the components

$$O_{\beta\gamma\tau} = O'_{\beta\gamma\tau} - (\delta_{\beta\gamma}V_\tau + \delta_{\beta\tau}V_\gamma + \delta_{\gamma\tau}V_\beta), \tag{13}$$

and the vector $\mathbf{V}$ is determined by the expression

$$\mathbf{V} = \frac{1}{5}\int\{2[\mathbf{r}'\cdot\mathbf{P}(\mathbf{r}')]\mathbf{r}' + \mathbf{r}'^2\mathbf{P}(\mathbf{r}')\}d\mathbf{r}'. \tag{14}$$

Thus the components of the tensor $\hat{O}''$ are determined by

$$O''_{\beta\gamma\tau} = \delta_{\beta\gamma}V_\tau + \delta_{\beta\tau}V_\gamma + \delta_{\gamma\tau}V_\beta. \tag{15}$$

Here, $\beta = x,y,z$, $\gamma = x,y,z$, and $\tau = x,y,z$, $\delta_{\tau\beta}$ is the Kronecker delta.

(3)

$$\hat{M}' = \hat{M} + \hat{M}'', \tag{16}$$

where $\hat{M}$ is the irreducible tensor of the magnetic quadrupole moment

$$\hat{M} = \frac{\omega}{3i}\int\{[\mathbf{r}'\times\mathbf{P}(\mathbf{r}')]\mathbf{r}' + \mathbf{r}'[\mathbf{r}'\times\mathbf{P}(\mathbf{r}')]\}d\mathbf{r}', \tag{17}$$

and the components of the asymmetric tensor

$$\hat{M}'' = \frac{\omega}{3i}\int\{[\mathbf{r}'\times\mathbf{P}(\mathbf{r}')]\mathbf{r}' - \mathbf{r}'[\mathbf{r}'\times\mathbf{P}(\mathbf{r}')]\}d\mathbf{r}' \tag{18}$$

can be presented as

$$M''_{\beta\gamma} = \frac{1}{2}\sum_{\tau}\epsilon_{\beta\gamma\tau}W_{\tau}, \quad (19)$$

with the vector

$$\mathbf{W} = \frac{2\omega}{3i}\int\{\mathbf{r}'^2\mathbf{P}(\mathbf{r}') - [\mathbf{r}'\cdot\mathbf{P}(\mathbf{r}')]\mathbf{r}'\}d\mathbf{r}', \quad (20)$$

where $\hat{\epsilon}$ is the Levi-Civita tensor.

Finally, the induced polarization (or the induced current $\mathbf{j} = -i\omega\mathbf{P}$) can be written in the irreducible multipole representation as

$$\begin{aligned}\mathbf{P}(\mathbf{r}) \simeq\ & \mathbf{p}\delta(\mathbf{r}) - \frac{1}{6}\hat{Q}\nabla\delta(\mathbf{r}) + \frac{i}{\omega}[\nabla\times\mathbf{m}\delta(\mathbf{r})]\\ &+\frac{1}{6}\hat{O}[\nabla\nabla\delta(\mathbf{r})] - \frac{i}{2\omega}[\nabla\times\hat{M}\nabla\delta(\mathbf{r})]\\ &-\frac{i}{\omega}\mathbf{T}\Delta\delta(\mathbf{r}) - \frac{q}{6}\nabla\delta(\mathbf{r}) + [\nabla\nabla\delta(\mathbf{r})]\mathbf{L}, \end{aligned}\quad (21)$$

where $\Delta \equiv \nabla\cdot\nabla$ is the Laplace operator, and

$$\mathbf{T} = \frac{i\omega}{6}\mathbf{V} - \frac{1}{4}\mathbf{W} \equiv \frac{i\omega}{10}\int\{2\mathbf{r}'^2\mathbf{P}(\mathbf{r}') - [\mathbf{r}'\cdot\mathbf{P}(\mathbf{r}')]\mathbf{r}'\}d\mathbf{r}' \quad (22)$$

is the *toroidal* dipole moment [5,12]. The vector

$$\mathbf{L} = \frac{1}{3}\mathbf{V} - \frac{i}{4\omega}\mathbf{W} \equiv \frac{1}{10}\int\{3[\mathbf{r}'\cdot\mathbf{P}(\mathbf{r}')]\mathbf{r}' - \mathbf{r}'^2\mathbf{P}(\mathbf{r}')\}d\mathbf{r}' \quad (23)$$

and the scalar value

$$q = 2\int[\mathbf{r}'\cdot\mathbf{P}(\mathbf{r}')]d\mathbf{r}'.$$

Note that the structure of Eq. (21) differs from that of Eq. (3) due to the last three terms. The vectors $\mathbf{T}$ and $\mathbf{L}$ appear from the third-order multipoles, whereas the q term is the multipole of the second order.

Importantly, the contribution of the toroidal dipole term into the multipole decomposition of the induced polarization (current) and into the scattered field (see below) *naturally* appears in the framework of the irreducible representation for the Cartesian multipole moments [5] without the introduction of toroidal multipole form factors [7].

B. Electric fields generated by multipoles

The scattered electric field $\mathbf{E}_{\rm sca}$, generated in the far-wave zone by the induced polarization, can be presented as

$$\mathbf{E}_{\rm sca}(\mathbf{r}) = \frac{k_0^2}{\varepsilon_0}\int_{V_s}\hat{G}^{\rm FF}(\mathbf{r},\mathbf{r}')\mathbf{P}(\mathbf{r}')d\mathbf{r}', \quad (24)$$

where $\hat{G}^{\rm FF}(\mathbf{r},\mathbf{r}')$ is the far-field approximation of the Green's tensor for a system without a scatterer [17], and V_s is the scatterer volume where $\mathbf{P}(\mathbf{r}) \neq 0$. When $\hat{G}^{\rm FF}(\mathbf{r},\mathbf{r}')$ is replaced by the total Green's tensor $\hat{G}(\mathbf{r},\mathbf{r}')$, Eq. (24) determines the electric field in all wave zones around the scatterer. In this case, using the multipole decomposition (21), one can get electric fields generated by different multipoles. For example, the electric field $\mathbf{E}_{\rm TD}$, generated by the toroidal dipole $\mathbf{T}$ (21), is given by

$$\mathbf{E}_{\rm TD}(\mathbf{r}) = -\frac{ik_0^2}{\varepsilon_0\omega}\int_{V_s}\hat{G}(\mathbf{r},\mathbf{r}')\mathbf{T}\Delta\delta(\mathbf{r}')d\mathbf{r}'. \quad (25)$$

After integration, assuming that the toroidal dipole is located at $\mathbf{r}_T$, one obtains

$$\mathbf{E}_{\rm TD}(\mathbf{r}) = \frac{ik_0^2k_d^2}{\varepsilon_0\omega}\hat{G}(\mathbf{r},\mathbf{r}_T)\mathbf{T}, \quad (26)$$

where k_0 and k_d are the wave numbers in vacuum and in the surrounding medium;

$$\begin{aligned}\hat{G}(\mathbf{r},\mathbf{r}') = \Bigg[&\left(\frac{1}{R} + \frac{i}{k_dR^2} - \frac{1}{k_d^2R^3}\right)\hat{U}\\ &+\left(-\frac{1}{R} - \frac{3i}{k_dR^2} + \frac{3}{k_d^2R^3}\right)\mathbf{e}_R\mathbf{e}_R\Bigg]\frac{e^{ik_dR}}{4\pi}.\end{aligned}$$

Here, $R = |\mathbf{R}| = |\mathbf{r}-\mathbf{r}'|$, and $\mathbf{e}_R\mathbf{e}_R$ is the dyadic product of the unit vector $\mathbf{e}_R = \mathbf{R}/R$. As can be seen from (25), the toroidal dipole generates the electric field in all wave zones similar to the electric dipole.

Note that the last two multipole terms containing q and $\mathbf{L}$ in expression (21) do not generate electromagnetic fields in all wave zones. This can be shown using a property of the Green's tensor $\hat{G}(\mathbf{r},\mathbf{r}')$:

$$\nabla'\cdot\mathbf{G}_i = \frac{\partial G_{i1}}{\partial x'} + \frac{\partial G_{i2}}{\partial y'} + \frac{\partial G_{i3}}{\partial z'} = 0, \quad \text{if } \mathbf{r}\neq\mathbf{r}'. \quad (27)$$

III. SCATTERING AND EXTINCTION

A. Scattering cross section

Applying the method described in Ref. [17] for the calculation of the scattered electric field $\mathbf{E}_{\rm sca}$ and using the multipole decomposition (21), one can obtain the following expression,

$$\mathbf{E}_{\rm sca}(\mathbf{r}) \simeq \frac{e^{ik_dr}}{r}\mathbf{E}_0^{\rm sca}(\mathbf{n}), \quad (28)$$

where

$$\begin{aligned}\mathbf{E}_0^{\rm sca}(\mathbf{n}) \simeq \frac{k_0^2}{4\pi\varepsilon_0}\Bigg([\mathbf{n}\times[\mathbf{D}\times\mathbf{n}]] + \frac{1}{v_d}[\mathbf{m}\times\mathbf{n}]\\ +\frac{ik_d}{6}[\mathbf{n}\times[\mathbf{n}\times\hat{Q}\mathbf{n}]] + \frac{ik_d}{2v_d}[\mathbf{n}\times(\hat{M}\mathbf{n})]\\ +\frac{k_d^2}{6}[\mathbf{n}\times[\mathbf{n}\times\hat{O}(\mathbf{nn})]]\Bigg)\end{aligned} \quad (29)$$

is the scattering amplitude into the direction $\mathbf{n}$ ($\mathbf{n}$ is the unit vector directed along $\mathbf{r}$). Here, we combined the electric dipole $\mathbf{p}$ and the toroidal dipole $\mathbf{T}$ as

$$\mathbf{D} = \mathbf{p} + \frac{ik_d}{v_d}\mathbf{T} \equiv \mathbf{p} + \frac{ik_0}{c}\varepsilon_d\mathbf{T}, \quad (30)$$

where $v_d = c/\sqrt{\varepsilon_d}$ is the speed of light in the surrounding medium. The vector $\mathbf{D}$ can be called the *total* electric dipole (TED) moment because the field propagators of $\mathbf{p}$ and $\mathbf{T}$ differ from each other by the only scalar factor ik_d/v_d. Since the contribution of $\mathbf{T}$ in the TED moment increases with growing dielectric permittivity ε_d, this can be used for the manipulation of the TED moment by changing the surrounding conditions.

The far-field scattered power $dP_{\rm sca}$ into the solid angle $d\Omega = \sin\theta\, d\varphi d\theta$ is determined by the time-averaged

Poynting vector [1] so that

$$dP_{\rm sca} = \frac{1}{2}\sqrt{\frac{\varepsilon_0\varepsilon_d}{\mu_0}}|\mathbf{E}_{\rm sca}|^2 r^2 d\Omega. \quad (31)$$

Inserting (29) in (31), after integration over the total solid angle [18], the total scattering power can be obtained, including the third-order multipoles: magnetic quadrupole (MQ), electric octupole (EOC), and toroidal dipole (TD),

$$P_{\rm sca} \simeq \frac{k_0^4}{12\pi\varepsilon_0^2 v_d\mu_0}\left|\mathbf{p}+\frac{ik_d}{v_d}\mathbf{T}\right|^2 + \frac{k_0^4\varepsilon_d}{12\pi\varepsilon_0 v_d}|\mathbf{m}|^2 + \frac{k_0^6\varepsilon_d}{1440\pi\varepsilon_0^2 v_d\mu_0}\sum_{\alpha\beta}|Q_{\alpha\beta}|^2 + \frac{k_0^6\varepsilon_d^2}{160\pi\varepsilon_0 v_d}\sum_{\alpha\beta}|M_{\alpha\beta}|^2 + \frac{k_0^8\varepsilon_d^2}{3780\pi\varepsilon_0^2 v_d\mu_0}\sum_{\alpha\beta\gamma}|O_{\alpha\beta\gamma}|^2. \quad (32)$$

The first term in (32) that is determined by the superposition of the electrical and toroidal dipoles includes the interference between them, which at certain conditions can result in the realization of a nonradiating anapole mode [9,12]. Due to the irreducible properties of the $\hat{Q}$, $\hat{M}$, and $\hat{O}$ tensors, there is no interference between them.

Integrating (31) we used the following useful relations,

$$\overline{n_\alpha n_\beta} = \tfrac{1}{3}\delta_{\alpha\beta}, \quad (33)$$

$$\overline{n_\alpha n_\beta n_\gamma n_\eta} = \tfrac{1}{15}(\delta_{\alpha\beta}\delta_{\gamma\eta}+\delta_{\alpha\gamma}\delta_{\beta\eta}+\delta_{\alpha\eta}\delta_{\beta\gamma}), \quad (34)$$

$$\overline{n_\alpha n_\beta n_\gamma n_\eta n_\tau n_\epsilon} = \tfrac{1}{105}(\delta_{\alpha\beta}\delta_{\gamma\eta}\delta_{\tau\epsilon}+\delta_{\alpha\beta}\delta_{\gamma\tau}\delta_{\eta\epsilon}+\delta_{\alpha\beta}\delta_{\gamma\epsilon}\delta_{\tau\eta} + \delta_{\alpha\gamma}\delta_{\beta\eta}\delta_{\tau\epsilon}+\delta_{\alpha\gamma}\delta_{\beta\tau}\delta_{\eta\epsilon}+\delta_{\alpha\gamma}\delta_{\beta\epsilon}\delta_{\tau\eta} + \delta_{\alpha\eta}\delta_{\beta\gamma}\delta_{\tau\epsilon}+\delta_{\alpha\eta}\delta_{\beta\tau}\delta_{\gamma\epsilon}+\delta_{\alpha\eta}\delta_{\beta\epsilon}\delta_{\tau\gamma} + \delta_{\alpha\tau}\delta_{\gamma\eta}\delta_{\beta\epsilon}+\delta_{\alpha\tau}\delta_{\beta\eta}\delta_{\epsilon\gamma}+\delta_{\alpha\tau}\delta_{\epsilon\eta}\delta_{\beta\gamma} + \delta_{\alpha\epsilon}\delta_{\gamma\eta}\delta_{\beta\tau}+\delta_{\alpha\epsilon}\delta_{\beta\eta}\delta_{\gamma\tau}+\delta_{\alpha\epsilon}\delta_{\tau\eta}\delta_{\beta\gamma}), \quad (35)$$

where averaging over all space directions of the unit vector **n** component product is performed (see Ref. [19]).

The scattering cross sections $\sigma_{\rm sca}$ are defined from $P_{\rm sca}$ by normalization to the energy flux of the incident wave $I_{\rm inc} = (\varepsilon_0\varepsilon_d/\mu_0)^{1/2}|\mathbf{E}_{\rm inc}|^2/2$:

$$\sigma_{\rm sca} \simeq \frac{k_0^4}{6\pi\varepsilon_0^2|\mathbf{E}_{\rm inc}|^2}\left|\mathbf{p}+\frac{ik_d}{v_d}\mathbf{T}\right|^2 + \frac{k_0^4\varepsilon_d\mu_0}{6\pi\varepsilon_0|\mathbf{E}_{\rm inc}|^2}|\mathbf{m}|^2 + \frac{k_0^6\varepsilon_d}{720\pi\varepsilon_0^2|\mathbf{E}_{\rm inc}|^2}\sum_{\alpha\beta}|Q_{\alpha\beta}|^2 + \frac{k_0^6\varepsilon_d^2\mu_0}{80\pi\varepsilon_0|\mathbf{E}_{\rm inc}|^2}\sum_{\alpha\beta}|M_{\alpha\beta}|^2 + \frac{k_0^8\varepsilon_d^2}{1890\pi\varepsilon_0^2|\mathbf{E}_{\rm inc}|^2}\sum_{\alpha\beta\gamma}|O_{\alpha\beta\gamma}|^2. \quad (36)$$

Note that in contrast to the previously developed expressions for the scattered power and scattering cross section in terms of electric dipole, magnetic dipole, and toroidal dipole terms [7,9], here we explicitly included the contribution from the electric octupole term. That should be done because the electric octupole, toroidal dipole, and the magnetic quadrupole moments are multipoles of the same order.

B. Extinction cross section

Now we turn to the consideration of the extinction power $P_{\rm ext}$ determined by the expression

$$P_{\rm ext} = \frac{\omega}{2}{\rm Im}\int \mathbf{E}^*_{\rm inc}(\mathbf{r})\cdot\mathbf{P}(\mathbf{r})d\mathbf{r}, \quad (37)$$

where $\mathbf{E}_{\rm inc}(\mathbf{r})$ is the electric field of the incident wave [17].

Inserting the multipole decomposition (21) and considering the incident plane wave $\mathbf{E}_{\rm inc}(\mathbf{r}) = \mathbf{E}_0\exp(i\mathbf{k}_d\mathbf{r})$, one can write

$$P_{\rm ext} \simeq \frac{\omega}{2}{\rm Im}\left\{\mathbf{E}_0^*\cdot\left(\mathbf{p}+\frac{ik_d}{v_d}\mathbf{T}+\frac{1}{v_d}[\mathbf{m}\times\mathbf{n}_{\rm inc}]-\frac{ik_d}{6}(\hat{Q}\mathbf{n}_{\rm inc}) + \frac{ik_d}{2v_d}[\mathbf{n}_{\rm inc}\times(\hat{M}\mathbf{n}_{\rm inc})]-\frac{k_d^2}{6}\hat{O}(\mathbf{n}_{\rm inc}\mathbf{n}_{\rm inc})\right)\right\}, \quad (38)$$

where $\mathbf{n}_{\rm inc} = \mathbf{k}_d/k_d$ is the unit vector along the incident direction. Equation (38) gives the multipole representation of the optical theorem. This follows from the comparison of (38) with the scattering amplitude (29) in the forward direction, when $\mathbf{n} = \mathbf{n}_{\rm inc}$. Similar to the scattering cross section, the extinction cross section is defined $P_{\rm ext}$ by normalization to the energy flux of the incident wave $I_{\rm inc} = (\varepsilon_0\varepsilon_d/\mu_0)^{1/2}|\mathbf{E}_{\rm inc}|^2/2$. The absorption cross section $\sigma_{\rm abs} = \sigma_{\rm ext} - \sigma_{\rm sca}$.

In case of an incident plane wave linear polarized along the x axis and propagated along the z-axis direction, the extinction cross section of an arbitrarily shaped scatterer can be written as

$$\sigma_{\rm ext} \simeq \frac{k_d}{\varepsilon_0\varepsilon_d|E_{0x}|^2}{\rm Im}\left\{E_{0x}^*\left(p_x+\frac{ik_d}{v_d}T_x-\frac{ik_d}{6}Q_{xz} + \frac{1}{v_d}m_y-\frac{ik_d}{2v_d}M_{yz}-\frac{k_d^2}{6}O_{xzz}\right)\right\}. \quad (39)$$

Another approach to multipole decomposition of the optical theorem (37) is the Taylor expansion of the exponential factor in the incident electric field $\mathbf{E}_{\rm inc}(\mathbf{r}) = E_{0x}\exp(ik_dz)$, so that one can write

$$\sigma_{\rm ext} \simeq \frac{k_d}{\varepsilon_0\varepsilon_d|E_{0x}|^2}{\rm Im}\left\{E_{0x}^*\left(\int P_x(\mathbf{r})d\mathbf{r} - ik_d\int zP_x(\mathbf{r})d\mathbf{r} - \frac{k_d^2}{2}\int z^2P_x(\mathbf{r})d\mathbf{r}\right)\right\}, \quad (40)$$

where the first integral is p_x, the second integral term combines the m_y and Q_{xz} contributions, and the third term is represented by the third-order multipoles T_x, M_{yz}, and O_{xzz}. For example, it can be directly verified that

$$\frac{ik_d}{2v_d}M_{yz}+\frac{k_d^2}{6}O_{xzz}-\frac{ik_d}{v_d}T_x = \frac{k_d^2}{2}\int z^2P_x(\mathbf{r})d\mathbf{r}. \quad (41)$$

From expression (40), one can see that the product $L_\parallel k_d$ of the scatterer length $L_\parallel$ in the z direction (chosen along the incident wave vector $\mathbf{k}_d$) and wave number k_d can be considered as the expansion parameter. If $L_\parallel k_d < 1$, the role of high-order multipoles in the extinction cross section will be negligibly weak even for scatterers with $L_\perp k_d \geqslant 1$, where $L_\perp$ is the scatterer size in the perpendicular direction with respect to the wave vector $\mathbf{k}_d$.

According to the optical theorem, the extinction cross section is determined by the imaginary part of the interference term between the incident and the only *forward* scattered

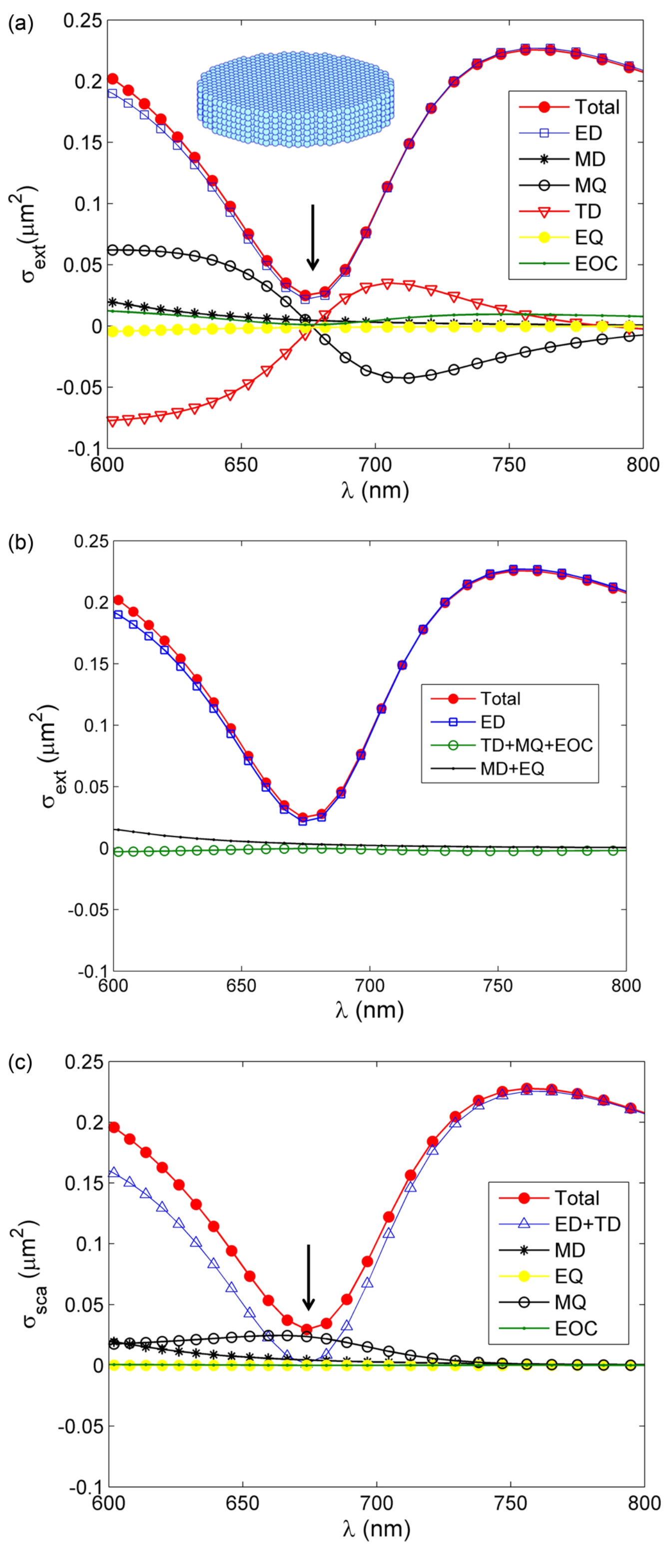

FIG. 1. (a), (b) Extinction cross-section spectra for a disk shaped dielectric nanoparticle, with a radius of 150 nm and height of 60 nm, located in air. (a) The multipole decomposition corresponds to expression (39). (b) The multipole decomposition corresponds to expression (40). (c) Scattering cross-section spectra for the same nanoparticle. The multipole decomposition corresponds to expression (36). Incident light is a linearly polarized plane wave propagating perpendicular to the disk surface. The graphs also show contributions from different multipoles.

light amplitudes. As a result, the extinction cross section is determined by the polarization component parallel to the incident electric field amplitude $\mathbf{E}_0$ (40). Equations (39) and (40) are equivalent, but one should be careful with their physical interpretation. In Eq. (39), only certain parts of the multipole components are presented in explicit form. As it will be shown below, the contribution of multipole terms in Eq. (39) can be both positive and negative. Therefore, a physical interpretation of the extinction cross section Eq. (39) in terms of multipoles can be problematic. However, terms in Eq. (40), providing information about total contributions of the multipoles of a certain order, are justified.

IV. NUMERICAL RESULTS

Expressions (32)–(40) are applicable for arbitrarily shaped nanoparticles. In addition to the well-known textbooks [19–21], they present the multipole decomposition of the scattering and extinction powers (cross sections) including the contribution of the third-order multipole moments: electric octupole, magnetic quadrupole, and toroidal dipole. These expressions can be used for investigations of the role of different multipole moments in light scattering. It could be expected that multipole decompositions of σ_{ext} and σ_{sca} will provide the same information about multipole contributions in light extinction and scattering. However, this statement is not generally valid for arbitrarily shaped scatterers. In contrast to extinction calculated from the optical theorem, the scattering power is determined by the integrated scattered light intensity. For arbitrarily shaped scatterers, their multipoles depend on the particle geometry and irradiation conditions and their contributions to (36) and (39) can be significantly different. Even for scatterers with small sizes in the light incident direction, the total scattering can be determined by high-order multipoles. Thus the multipole decomposition obtained from the optical theorem cannot be in general used for estimation of which multipoles give the main contributions in the scattered fields. Note that the main difference between the calculations of the extinction and scattering powers is in the different information about the scattering. The optical theorem considers only scattering in the forward direction, whereas the total scattering is determined by integrated scattered light in all spatial directions. As a result, the forward scattering can be basically determined by the electric dipole moment, because of weak contributions from high-order multipoles (40). In contrast, the total scattering cross section can contain strong contributions from side scattering generated by high multipole moments appearing without interference with other multipoles (36). For an illustration of the above discussion, we consider below several numerical examples. In our calculations, the discrete dipole approximation with the multipole decomposition procedure described in Ref. [17] is used.

Figure 1 demonstrates the multipole contributions into the extinction and scattering cross sections for a dielectric nanodisk with the dielectric constant $\varepsilon_p = 16$. Note that in this (absorptionless) case, $\sigma_{\text{ext}} = \sigma_{\text{sca}}$. From the optical theorem (39) it follows that the extinction is determined solely by the electric dipole (ED) contribution, whereas the contributions from the magnetic quadrupole (MQ) and toroidal dipole (TD) terms nearly compensate each other (see Fig. 1(a)].

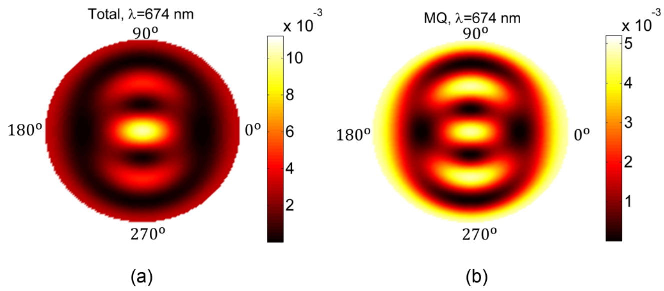

FIG. 2. Scattering directivity (arbitrary units) for the same disk nanoparticle and irradiation conditions (see Fig. 1) at 674 nm wavelength in the spherical coordinates. The diagram center ($\theta = 0$) corresponds to forward scattering; the diagram edges ($\theta = \pi$) correspond to backward scattering. (a) Total scattering and (b) MQ scattering diagrams.

Moreover, at the extinction minimum [black arrow in Fig. 1(a)] the separate contributions of the MQ and TD terms are close to zero. The multipole decomposition in Fig. 1(b) calculated using (40) confirms the negligible role of the second-, third-, and higher-order multipoles in the extinction.

For the scattering cross section shown in Fig. 1(c) the multipole contributions are different. At the cross-section minimum shown by the black arrow, the main contribution is provided by the MQ term and contributions of other multipoles are close to zero, including the magnetic dipole (MD), electric quadrupole (EQ), and electric octupole (EOC) terms. The corresponding scattering diagrams, calculated at $\lambda = 674$ nm wavelength and presented in Fig. 2, confirm the main role of the MQ moment at the minimum of the scattering cross section presented in Fig. 1(c). Note that the destructive interference between the electric and toroidal dipoles suppresses the total electric dipole (ED+TD) contribution in the scattering cross section at its minimum [Fig. 1(c)]. This effect, which is not visible in the extinction cross section, corresponds to the excitation of the anapole mode [12].

Another example is devoted to the optical properties of a silicon nanodisk having a through hole at the center [see the inset in Fig. 3(a)]. In this case again, due to the small imaginary part of dielectric permittivity of crystalline silicon in the visible range [22,23], the total extinction is basically determined by total scattering (Fig. 3). Multipole decompositions (39) and (40), presented in Figs. 3(a) and 3(b), respectively, show that the contribution of the third-order multipoles is negligibly small and the extinction cross section is mainly determined by the contribution of the ED term. The contributions from MQ and TD terms nearly compensate each other [see Fig. 3(a)]. For the scattering cross section shown in Fig. 3(c) the multipole contributions are different. Around the cross-section maximum, the main contribution is provided by the multipoles of all considered orders, including MQ resonance. Note that in this case the anapolelike mode can be excited resonantly. This happens when the resonantly excited ED moment is partially compensated by the resonantly excited TD moment. The multipole decomposition presented in Fig. 3(c) shows that the maximum of the ED contribution into the scattering cross section at $\lambda \approx 500$ nm coincides with the maximum of the TD contribution [see the black arrow

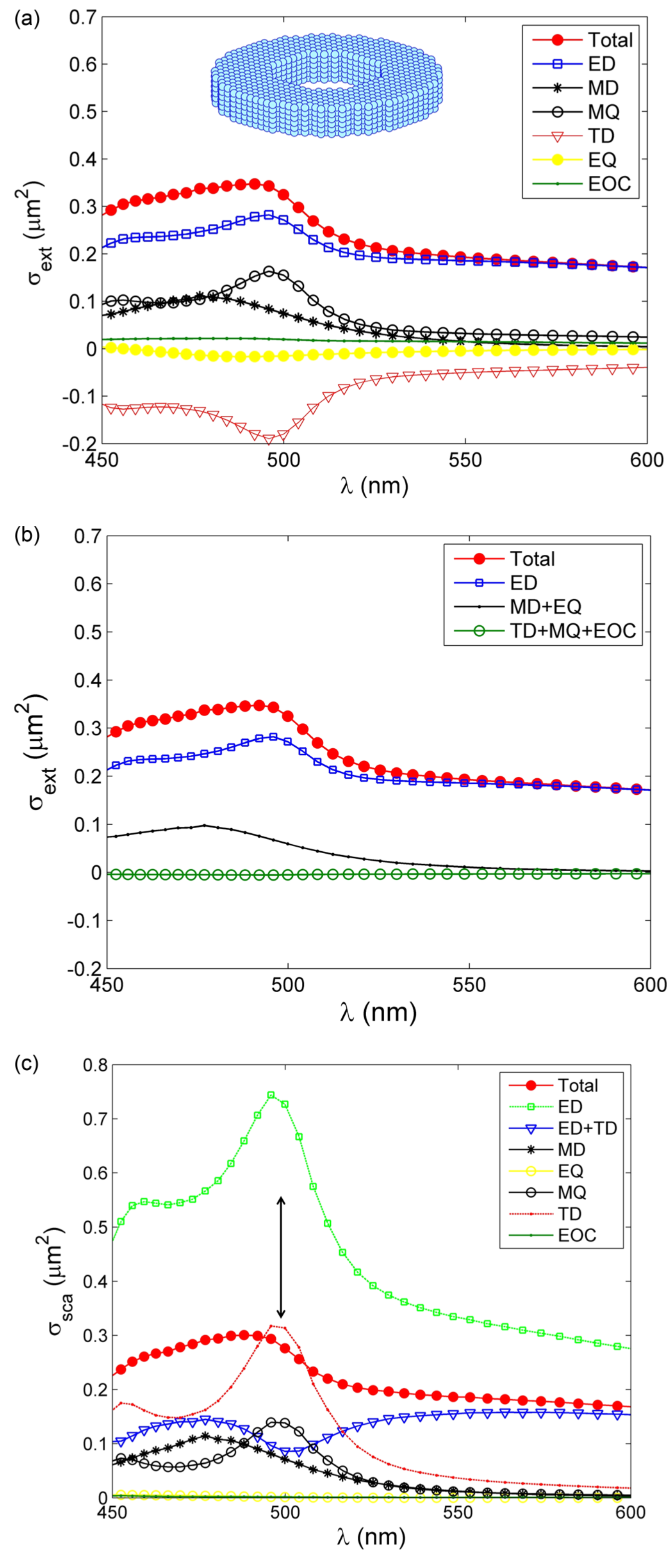

FIG. 3. (a) Extinction cross-section spectra for the disk shaped silicon [22] nanoparticle, with a radius of 150 nm and height of 60 nm having a through hole at the center with a radius of 75 nm, located in air. (a) The multipole decomposition corresponds to expression (39). (b) The multipole decomposition corresponds to expression (40). (c) Scattering cross-section spectra for the same nanoparticle. Irradiation conditions are the same as in Fig. 1. The graphs also show contributions from different multipoles.

in Fig. 3(c)]. However, the interference between the ED and TD terms provides the minimum of the ED+TD contribution into the scattering cross section at this spectral range [blue curve in Fig. 3(c)]. Note that only the total electric dipole **D** (30), which is a vector combination of the electric and toroidal dipole moments ED+TD, explicitly appears in the multipole decomposition of the scattering cross section (36) and is presented by a blue-triangle curve in Fig. 3(c). Separate contributions of the electric dipole ED and toroidal dipole TD without interference between them [Fig. 3(c)] are shown only for demonstrative purposes.

V. CONCLUSION

In conclusion, Cartesian multipoles in irreducible representations have been used for the multipole decomposition of the induced polarization (current) up to the third order inside an arbitrarily shaped scatterer (nanoparticle). It has been demonstrated that the third order of multipole decomposition includes the toroidal dipole, magnetic quadrupole, electric octupole terms, and also nonradiating terms. It has been demonstrated that, in the developed approach, for arbitrarily shaped scatterers, the multipole decompositions of σ_{ext} and σ_{sca} provide different information about multipole contributions. In contrast to σ_{sca}, where the total values of all multipole components are present, the multipole decomposition of σ_{ext} includes only certain parts of the multipole components. Therefore, for absorptionless nanoparticles, a physical interpretation of the scattering process in terms of multipole decomposition of the extinction cross section (derived from the optical theorem) can be problematic and cannot be justified. The obtained results are of principal importance for a correct analysis of light scattering by nanoparticles and nanoparticle structures.

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