Multipole light scattering by nonspherical nanoparticles in the discrete dipole approximation

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In the framework of the discrete dipole approximation we develop a theoretical approach that allows the analysis of the role of multipole modes in the extinction and scattering spectra of arbitrary shaped nanoparticles. The main attention is given to the first multipoles including magnetic dipole and electric quadrupole moments. The role of magnetic quadrupole and electric octupole modes is also discussed. The method is applied to nonspherical Si nanoparticles with resonant multipole responses in the visible optical range, allowing a decomposition of single extinction (scattering) peaks into their constituent multipole contributions. It is shown by numerical simulations that it is possible to design silicon particles for which the electric dipole and magnetic dipole resonances are located at the same wavelength under certain propagation directions of incident light, providing new possibilities in metamaterial developments.

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I. INTRODUCTION

The main optical feature of nanoparticles is their resonant response as a reaction on external optical fields. This makes them very attractive for different practical applications, such as real-time sensor technologies,[1–5] surface enhanced Raman spectroscopy (SERS) with sensitivity down to a single molecule,[6,7] light energy guiding along nanoparticle chains,[8,9] efficient micro-optical devices for surface plasmon-polaritons,[10] nanolensing,[11,12] creation of metamaterials with unique optical properties including negative refraction,[13–15] and electromagnetically induced transparency.[16–18] So the optical properties of nanoparticles attract considerable attention at the present time.

The Mie theory provides the unique analytical solution of Maxwell's equations for scattering of electromagnetic waves by single particles.[19] In this theory, the extinction σ_{ext} and scattering σ_{sct} cross sections are presented as a series of corresponding partial multipole cross sections,[20] which contain information about the resonant excitation of different multipole modes inside the particle.[21–23] Resonantly excited modes determine the distribution of electric and magnetic fields inside the particle as well as in the near- and far-field zones and as a consequence, the features of the particle behavior in electromagnetic fields.[24,25] However, the critical restriction of the Mie theory is that it can only be applied to spherical particles! In the general case of arbitrary shaped particles, numerical methods have to be used. Several approaches to solving this problem have been developed which give information about multipole scattering and are known as generalized multipole techniques.[26] These techniques are based on the presentations of the fields as a linear superposition of known basic functions with coefficients that are found numerically applying the field boundary conditions. Using the multipolar functions as the basic functions, one can get information about multipole structure of the fields in the considered systems. The most popular methods in this context are the T-matrix method[27–29] and the multiple multipole method.[30–32]

In this paper, we address our attention to another powerful numerical technique that is widely used for the computation of light scattering by arbitrary shaped particles and is called the discrete dipole approximation (DDA).[33–35] A comprehensive overview of this method can by found in Ref. 36. In contrast to the generalized multipole techniques, there are no multipole presentations of the fields in different domains of considered systems and ordinary calculated results include only total extinction (ECS), absorption (ACS), and scattering (SCS) cross sections. Up to now, there are no direct methods in the DDA which are capable of describing the role of different multipole modes to the ECSs, ACSs, and SCSs for arbitrary shaped particles.[28,37,38] Here, we develop such a direct method and demonstrate its applicability for analysis of multipole light scattering by nonspherical Si nanoparticles. Our approach is based on a simple straightforward idea. Because within DDA, nanoparticles are represented as an array of point dipoles in a local domain with dimensions smaller than the scattered wavelength, one can represent the scattered fields (radiated fields by the dipoles) approximately as a series of multipole contributions, just as it is demonstrated in classical textbooks[39,40] for localized systems of electric charges or electric currents. Moreover, using the connection between electric charge (electric current) density and matter polarization, we can adopt the expressions of the multipole moments of electric charge systems to obtain the multipole moments of electric dipole systems, as described in Sec. II and in the Appendix. In this paper, the main attention is given to the first multipoles including the electric dipole, magnetic dipole, and electric quadrupole. The role of magnetic quadrupole and electric octupole modes is also discussed.

The paper is organized as follows. In Sec. II, we show how the multipole presentations of the extinction cross sections and the scattering cross sections can be obtained in the framework of DDA. Section III demonstrates the capabilities of the developed approach, considering different examples of the light scattering by nonspherical Si nanoparticles. Concluding remarks concerning applicable perspectives of the developed method and the Si nanoparticles are presented in Sec. IV. Important details of the calculational procedure are presented in the Appendix.

II. MULTIPOLES AND DDA

The main idea of DDA is the replacement of a scattering object by a cubic lattice of electric point dipoles (Fig. 1)

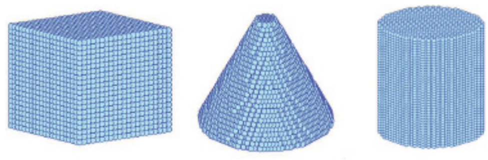

FIG. 1. (Color online) Several objects with different shapes are represented as a cubic lattice of spherical elements, each of which is characterized by an electric dipole polarizability.

with polarizability α_p.[36] Without loss of generality, α_p can be considered the same for all point dipoles. The dipole moment $\mathbf{p}_j$ induced in each lattice point j is determined by the local electric field, being the sum of the external electric field $\mathbf{E}_0(\mathbf{r}_j)$ at the position $\mathbf{r}_j$ of the dipole $\mathbf{p}_j$, and the electric fields at $\mathbf{r}_j$ generated by the point dipoles located at the positions $l \neq j$. For monochromatic fields the electric dipole moment of each point dipole is determined by the coupled-dipole equations

$$\mathbf{p}_j = \alpha_p \mathbf{E}_0(\mathbf{r}_j) + \alpha_p \frac{k_0^2}{\varepsilon_0} \sum_{l \neq j}^{N} \hat{G}(\mathbf{r}_j,\mathbf{r}_l)\mathbf{p}_l \,, \tag{1}$$

where k_0 is the vacuum wave number, ε_0 is the vacuum permittivity, $\hat{G}(\mathbf{r}_j,\mathbf{r}_l)$ is Green's tensor of the medium surrounding the scatterer,[41] and N is the total number of point dipoles in the system [time dependence $\exp(-i\omega t)$ is omitted, ω is the circular frequency].

A. Extinction

After solving the system of equations (1), the total ECSs can be found using the optical theorem:[33,37]

$$\sigma_{\text{ext}} = \frac{k_d}{\varepsilon_0 \varepsilon_d |\mathbf{E}_0|^2} \,\text{Im} \sum_{j=1}^{N} \mathbf{E}_0^*(\mathbf{r}_j) \cdot \mathbf{p}_j. \tag{2}$$

Here, ε_d is the permittivity of the environment, k_d is the wave number in the environment, and * denotes complex conjugation. For arbitrary shaped particles, the extinction or scattering spectra can have multiple resonances attributed to excitation of certain electromagnetic modes inside the scattering object. For multipole classification and interpretation of these resonances, knowledge of the field distributions inside and outside the object is usually needed.[38] Here, we show that information about the excited multipoles can be obtained directly from expression (2). Expanding the field in a Taylor series around a point $\mathbf{r}_0$, one obtains[42]

$$\begin{aligned}\mathbf{E}_0^*(\mathbf{r}_j) \cdot \mathbf{p}_j &= \mathbf{E}_0^*(\mathbf{r}_0) \cdot \mathbf{p}_j \\ &\quad + [\nabla \mathbf{E}_0^*(\mathbf{r}_0)] : [(\mathbf{r}_j - \mathbf{r}_0) \otimes \mathbf{p}_j] + \cdots \,,\end{aligned} \tag{3}$$

where $\nabla \mathbf{E}_0^*(\mathbf{r}_0)$ denotes the gradient of the electric field, the sign $\otimes$ denotes the dyadic product of vectors, the sign : denotes the double scalar product of dyads. In this article, we use the double scalar product of two dyads $\hat{A}$ and $\hat{B}$ in the form[43] $\hat{A} : \hat{B} = \sum_{nm} A_{nm} B_{nm}$. Note, that if the particle size satisfies the condition $k_d|\mathbf{r}_j - \mathbf{r}_0| < 1$ $(j = 1 \ldots N)$, the series converges fast and only the first terms need to be taken into account in Eq. (3), with the first two corresponding to the electric dipole (ED), the magnetic dipole (MD), and the electric quadrupole (EQ).

To evaluate Eq. (3), we split the gradient of the electric field $\nabla \mathbf{E}_0^*(\mathbf{r}_0)$ into its symmetric and antisymmetric parts:

$$\nabla \mathbf{E}_0^*(\mathbf{r}_0) \equiv \frac{\nabla \mathbf{E}_0^* + \mathbf{E}_0^* \nabla}{2} + \frac{\nabla \mathbf{E}_0^* - \mathbf{E}_0^* \nabla}{2} \,, \tag{4}$$

where we use the symbolic notation[44]

$$(\nabla \mathbf{E}^* \pm \mathbf{E}^* \nabla)_{\beta\gamma} = \frac{\partial E_\gamma^*}{\partial \beta} \pm \frac{\partial E_\beta^*}{\partial \gamma} \,, \tag{5}$$

and $\beta = x,y,z$ and $\gamma = x,y,z$.

By using the connection between electric and magnetic fields $\nabla \times \mathbf{E}_0^*(\mathbf{r}_0) = -i\omega\mu_0 \mathbf{H}_0^*(\mathbf{r}_0)$, the double scalar product between the antisymmetric part of Eq. (4) and the dyadic $[(\mathbf{r}_j - \mathbf{r}_0) \otimes \mathbf{p}_j]$ can be written as

$$\begin{aligned}&\frac{\nabla \mathbf{E}_0^* - \mathbf{E}_0^* \nabla}{2} : [(\mathbf{r}_j - \mathbf{r}_0) \otimes \mathbf{p}_j] \\ &\quad = \frac{\omega\mu_0}{2i} \mathbf{H}_0^*(\mathbf{r}_0) \cdot [(\mathbf{r}_j - \mathbf{r}_0) \times \mathbf{p}_j] \,,\end{aligned} \tag{6}$$

where $\times$ indicates the vector product and μ_0 is the vacuum permeability (we consider only nonmagnetic materials). Using the definition of the magnetic moment (see Appendix)

$$\mathbf{m}_j(\mathbf{r}_0) = \frac{\omega}{2i} [(\mathbf{r}_j - \mathbf{r}_0) \times \mathbf{p}_j] \tag{7}$$

located at $\mathbf{r}_0$ and associated with the electric dipole $\mathbf{p}_j$ located at $\mathbf{r}_j$, one can write

$$\frac{\nabla \mathbf{E}_0^* - \mathbf{E}_0^* \nabla}{2} : [(\mathbf{r}_j - \mathbf{r}_0) \otimes \mathbf{p}_j] = \mu_0 \mathbf{H}_0^*(\mathbf{r}_0) \cdot \mathbf{m}_j(\mathbf{r}_0) \,. \tag{8}$$

Similarly, the double scalar product between the symmetric part of Eq. (4) and the dyadic $[(\mathbf{r}_j - \mathbf{r}_0) \otimes \mathbf{p}_j]$ can be written in the form

$$\frac{\nabla \mathbf{E}_0^* + \mathbf{E}_0^* \nabla}{2} : [(\mathbf{r}_j - \mathbf{r}_0) \otimes \mathbf{p}_j] = \frac{\nabla \mathbf{E}_0^* + \mathbf{E}_0^* \nabla}{12} : \hat{Q}_j(\mathbf{r}_0) \,, \tag{9}$$

where $\hat{Q}_j(\mathbf{r}_0)$ is the traceless tensor of the EQ moment at point $\mathbf{r}_0$ associated with the ED $\mathbf{p}_j$. To obtain expression (9), we used the definition of the EQ tensor and took into account that the divergence of the external electric field is equal to zero. Note that in contrast to traditional considerations, one should use the EQ tensor expression for a system of electric dipoles instead of a system of charges. This expression can be obtained starting from the definition of the common EQ tensor of electric charges[39]

$$Q_{\beta\gamma} = \int \rho(\mathbf{r})(3\beta\gamma - r^2\delta_{\beta\gamma})\, d\mathbf{r}, \tag{10}$$

where $\beta = x,y,z$ and $\gamma = x,y,z$, $\rho(\mathbf{r})$ is the electric charge density at a point $\mathbf{r} = (x,y,z)$, and $\delta_{\beta\gamma}$ is the Kronecker delta. Using the relation between the polarization $\mathbf{P}$ and the associated bound charge density $\rho_b = -\nabla \cdot \mathbf{P}$ with $\mathbf{P}(\mathbf{r}) \equiv \mathbf{p}\delta(\mathbf{r} - \mathbf{r}_j)$ for a single dipole located at $\mathbf{r}_j$, and inserting the density ρ_b into Eq. (10), one obtains the EQ tensor $\hat{Q}$ located at the origin of a Cartesian coordinate system and associated with the ED $\mathbf{p}$ at $\mathbf{r}_p = (x_p,y_p,z_p)$

$$Q_{\beta\gamma} = \int (r^2\delta_{\beta\gamma} - 3\beta\gamma)[\nabla \cdot \mathbf{p}\delta(\mathbf{r} - \mathbf{r}_p)]\, d\mathbf{r} \,, \tag{11}$$

where $\delta(\mathbf{r}-\mathbf{r}_p)$ is the Dirac delta function. After integration of Eq. (11) and shifting the coordinate system origin by the vector $-\mathbf{r}_0$, the elements of the EQ tensor $\hat{Q}$ located at the point $\mathbf{r}_0 = (x_0,y_0,z_0)$, associated with the ED $\mathbf{p}$ located at $\mathbf{r}_j = (x_j,y_j,z_j)$ in the new coordinate system, can be expressed by

$$Q_{\beta\beta} = 2[2(\beta_j-\beta_0)p_\beta - (\gamma_j-\gamma_0)p_\gamma - (\tau_j-\tau_0)p_\tau]$$
$$Q_{\beta\gamma} = 3[(\beta_j-\beta_0)p_\gamma + (\gamma_j-\gamma_0)p_\beta]\,, \quad (12)$$

where $\beta = x,y,z$, and $\gamma = x,y,z$, and $\tau = x,y,z$; moreover, $\beta \neq \gamma$, $\tau \neq \gamma$, and $\tau \neq \beta$.

Taking into account only the first two terms in Eq. (3), the ECS can be written as

$$\sigma_{\text{ext}} \approx \frac{k_d}{\varepsilon_0\varepsilon_d|\mathbf{E}_0|^2}\,\text{Im}\left(\mathbf{E}_0^*(\mathbf{r}_0)\cdot\mathbf{p} + \mu_0\mathbf{H}_0^*(\mathbf{r}_0)\cdot\mathbf{m} + \frac{\nabla\mathbf{E}_0^* + \mathbf{E}_0^*\nabla}{12} : \hat{Q}\right), \quad (13)$$

with

$$\mathbf{p} = \sum_{j=1}^{N}\mathbf{p}_j, \quad \mathbf{m} = \sum_{j=1}^{N}\mathbf{m}_j(\mathbf{r}_0), \quad \hat{Q} = \sum_{j=1}^{N}\hat{Q}_j(\mathbf{r}_0)\,.$$

Note that in general, the total magnetic $\mathbf{m}$ and the electric quadrupole $\hat{Q}$ moments depend on the choice of the origin of the coordinate system $\mathbf{r}_0$.[39] Only if the total electric dipole moment of the system is zero is the uncertainty removed and $\mathbf{m}$ and $\hat{Q}$ are independent of the origin of the coordinate system. In resonance, when only one multipole mode determines light scattering and absorption, the value of this multipole moment is still weakly dependent on the choice of origin of the coordinate system. For homogeneous particles, however, there exists a special point $\mathbf{r}_c = \sum_{j=1}^{N}\mathbf{r}_j/N$, coinciding with the center of mass, with respect to which the number of multipole terms in the expansion (3) is minimum. For arbitrary shaped particles, it is therefore convenient to choose this point $\mathbf{r}_c$ as the origin of the coordinate system.

To estimate the contribution of higher-order multipoles to the ECS, the next higher terms of the expansion (3) have to be taken into account. For example, the third term, including magnetic quadrupole (MQ) and electric octupole (EOC) multipoles, can be written as $\{(\mathbf{r}_j-\mathbf{r}_0)\otimes(\mathbf{r}_j-\mathbf{r}_0) : \nabla\otimes\nabla\}[\mathbf{E}_0^*(\mathbf{r}_0)\cdot\mathbf{p}_j]/2$. For an incident plane wave propagating in z and polarized along the x direction $[\mathbf{E}_0(\mathbf{r}) = E_{0x}\exp(ik_dz)]$, the third term reduces to $(-k_d^2)(z_j - z_0)^2 p_{jx} E_{0x}^* \exp(-ik_dz_0)/2$. To estimate the separate contributions of the EOC or MQ modes one should use the definition of the octupole or magnetic quadrupole moments. There are several definitions of these moments depending on their symmetrical and trace properties.[25,45] But our procedure is independent of the multipole definitions and can be apply in any case. However, interpretation of the obtained multipole expansions can be different. For example, if the octupole and magnetic quadrupole moments are both totally traceless and symmetric, the multipole expansion can include an additional contribution from the so-called toroid dipole moment.[25] For other definitions (see Appendix), the toroid dipole moment contribution disappears.[25,46] Note that for small particles, contributions of higher-order multipole modes give only small corrections. Therefore, in the numerical calculations presented here, we consider the MQ-EOC term without subdivision of the corresponding electric and magnetic contributions.

B. Scattering

To obtain information about the contributions of different multipole modes into the spatial distribution of the scattered fields, one can use the following equation

$$\mathbf{E}(\mathbf{r}) = \frac{k_0^2}{\varepsilon_0}\sum_{j=1}^{N}\hat{G}(\mathbf{r},\mathbf{r}_j)\mathbf{p}_j \quad (14)$$

describing the superposition of the electric fields generated by all dipoles of the system. If the observation point is far away from the scattering object, i.e., $r \gg r_j$ for all j, Green's tensor $\hat{G}(\mathbf{r},\mathbf{r}_j)$ can be expanded into Taylor series around the point $\mathbf{r}_c$ with respect to $\mathbf{r}_j$, taking into account only several first terms. To do this, it is convenient to present Green's tensor as a sum of three parts: a near-field part $\hat{G}_N(\mathbf{r},\mathbf{r}_j)$, an intermediate-field part $\hat{G}_I(\mathbf{r},\mathbf{r}_j)$, and a far-field part $\hat{G}_F(\mathbf{r},\mathbf{r}_j)$[47]

$$\hat{G}_N(\mathbf{r},\mathbf{r}_j) = (-\hat{U} + 3\mathbf{n}_j\otimes\mathbf{n}_j)\frac{e^{ik_dR_j}}{4\pi k_d^2R_j^3}\,, \quad (15)$$

$$\hat{G}_I(\mathbf{r},\mathbf{r}_j) = (\hat{U} - 3\mathbf{n}_j\otimes\mathbf{n}_j)\frac{i\,e^{ik_dR_j}}{4\pi k_dR_j^2}\,, \quad (16)$$

$$\hat{G}_F(\mathbf{r},\mathbf{r}_j) = (\hat{U} - \mathbf{n}_j\otimes\mathbf{n}_j)\frac{e^{ik_dR_j}}{4\pi R_j}\,, \quad (17)$$

where $\hat{U}$ is the 3×3 unit tensor and $R_j = |\mathbf{r}-\mathbf{r}_j|$, $\mathbf{n}_j = \mathbf{R}_j/R_j$ is the unit vector directed from the point $\mathbf{r}_j$ to the point $\mathbf{r}$. The dyadic $\mathbf{n}_j\otimes\mathbf{n}_j$ can be expanded with respect to $\mathbf{r}_j$ around the central point $\mathbf{r}_c$ of the scattering object

$$\mathbf{n}_j\otimes\mathbf{n}_j = \mathbf{n}_c\otimes\mathbf{n}_c + 2\frac{(\mathbf{n}_c\cdot\mathbf{R}_{jc})}{R_c}\mathbf{n}_c\otimes\mathbf{n}_c - \frac{1}{R_c}(\mathbf{n}_c\otimes\mathbf{R}_{jc} + \mathbf{R}_{jc}\otimes\mathbf{n}_c) + \cdots\,, \quad (18)$$

where $\mathbf{n}_c = \mathbf{R}_c/R_c$, $\mathbf{R}_c = (\mathbf{r}-\mathbf{r}_c)$, and $\mathbf{R}_{jc} = (\mathbf{r}_j-\mathbf{r}_c)$. The factors $\exp(ik_dR_j)$, $1/R_j^3$, $1/R_j^2$, and $1/R_j$ outside the brackets of Eqs. (15)–(17) also have to be expanded:

$$e^{ik_dR_j} = e^{ik_d|\mathbf{R}_c-\mathbf{R}_{jc}|} \approx e^{ik_dR_c - ik_d\mathbf{n}_c\cdot\mathbf{R}_{jc}},$$
$$R_j^{-l} \approx \frac{1}{R_c}(1 + l\mathbf{n}_c\cdot\mathbf{R}_{jc}/R_c),$$

where $l = 1,2,3$. Together with the condition $k_dR_{jc} < 1$ for any j and using the definitions of the MD and EQ moments, the scattered electric field can be represented as a superposition of the individual ED, MD, and EQ contributions (see Appendix)

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_p(\mathbf{r}) + \mathbf{E}_Q(\mathbf{r}) + \mathbf{E}_m(\mathbf{r}) + \cdots \quad (19)$$

Here, $\mathbf{E}_p(\mathbf{r})$ and $\mathbf{E}_m(\mathbf{r})$ are the electric fields generated by the total ED $\mathbf{p} = \sum_j\mathbf{p}_j$ and MD $\mathbf{m}(\mathbf{r}_c) = \sum_j\mathbf{m}_j(\mathbf{r}_c)$ located at $\mathbf{r}_c$, respectively. Corresponding expressions for these fields in all wave zones are

$$\mathbf{E}_p(\mathbf{r}) = \frac{k_0^2 e^{ik_d R_c}}{4\pi\varepsilon_0 R_c}\left\{\left(1+\frac{i}{k_d R_c}-\frac{1}{k_d^2 R_c^2}\right)\hat{U} + \left(-1-\frac{i3}{k_d R_c}+\frac{3}{k_d^2 R_c^2}\right)\mathbf{n}_c\otimes\mathbf{n}_c\right\}\mathbf{p}\,, \quad (20)$$

$$\mathbf{E}_m(\mathbf{r}) = \sqrt{\frac{\mu_0}{\varepsilon_0}}\frac{k_0 k_d e^{ik_d R_c}}{4\pi R_c}\left(1+\frac{i}{k_d R_c}\right)[\mathbf{m}\times\mathbf{n}_c]\,. \quad (21)$$

The electric field created by the total EQ tensor located at $\mathbf{r}_c$ is given by

$$\mathbf{E}_Q(\mathbf{r}) = \frac{k_0^2}{\varepsilon_0}\,\hat{G}^Q(\mathbf{r},\mathbf{r}_c)(\hat{Q}(\mathbf{r}_c)\mathbf{n}_c)\,, \quad (22)$$

where $\hat{Q}(\mathbf{r}_c) = \sum_j \hat{Q}_j(\mathbf{r}_c)$ and $\mathbf{n}_c$ is a unit vector directing from $\mathbf{r}_c$ to $\mathbf{r}$. The tensor $\hat{G}^Q(\mathbf{r},\mathbf{r}_c)$ describing the EQ generated electric field at the point $\mathbf{r}$ can be written as (see Appendix)

$$\hat{G}^Q(\mathbf{r},\mathbf{r}_c) = \left\{\left(-1-\frac{i3}{k_d R_c}+\frac{6}{k_d^2 R_c^2}+\frac{i6}{k_d^3 R_c^3}\right)\hat{U} + \left(1+\frac{i6}{k_d R_c}-\frac{15}{k_d^2 R_c^2}-\frac{i15}{k_d^3 R_c^3}\right)\mathbf{n}_c\otimes\mathbf{n}_c\right\} \times \frac{ik_d e^{ik_d R_c}}{24\pi R_c}\,. \quad (23)$$

Using the far-field approximation of the scattered electric fields allows calculating the distribution of radiated power in the far-field zone and the total radiated power.[39,42]

Choosing the Cartesian coordinate system with the origin at the central point, so that $\mathbf{R}_c$ becomes simply $\mathbf{r}$, and using the spherical coordinate presentation, the scattered electric field in the far-field zone from the scatterer is given by the expressions

(i) for the electric dipole $\mathbf{p} = (p_x, p_y, p_z)$

$$E_\varphi^p(r,\varphi,\theta) = \frac{k_d^2 e^{ik_d r}}{4\pi\varepsilon_0\varepsilon_d r}(p_y\cos\varphi - p_x\sin\varphi)\,,$$
$$E_\theta^p(r,\varphi,\theta) = \frac{k_d^2 e^{ik_d r}}{4\pi\varepsilon_0\varepsilon_d r}(p_x\cos\varphi\cos\theta + p_y\sin\varphi\cos\theta - p_z\sin\theta)\,; \quad (24)$$

(ii) for the electric quadrupole moment tensor $\hat{Q}$

$$E_\varphi^Q(r,\varphi,\theta) = \frac{ik_d^3 e^{ik_d r}}{24\pi\varepsilon_0\varepsilon_d r}\{(\hat{Q}\mathbf{n})_x\sin\varphi - (\hat{Q}\mathbf{n})_y\cos\varphi\}\,,$$
$$E_\theta^Q(r,\varphi,\theta) = \frac{ik_d^3 e^{ik_d r}}{24\pi\varepsilon_0\varepsilon_d r}\{-(\hat{Q}\mathbf{n})_x\cos\varphi\cos\theta - (\hat{Q}\mathbf{n})_y\sin\varphi\cos\theta + (\hat{Q}\mathbf{n})_z\sin\theta\}\,, \quad (25)$$

where one should take $\mathbf{n} = (\sin\theta\cos\varphi,\ \sin\theta\sin\varphi,\ \cos\theta)$;

(iii) for the magnetic dipole $\mathbf{m} = (m_x, m_y, m_z)$

$$E_\varphi^m(r,\varphi,\theta) = \sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon_d}}\frac{k_d^2 e^{ik_d r}}{4\pi r}(-m_x\cos\varphi\cos\theta - m_y\sin\varphi\cos\theta + m_z\sin\theta)\,,$$
$$E_\theta^m(r,\varphi,\theta) = \sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon_d}}\frac{k_d^2 e^{ik_d r}}{4\pi r}(m_y\cos\varphi - m_x\sin\varphi)\,. \quad (26)$$

Here φ and θ are the azimuthal and polar angle of the spherical coordinate system, respectively, $\mathbf{n} = \mathbf{r}/r$.

The far-field scattered (radiated) power dP into the solid angle $d\Omega = \sin\theta\, d\varphi\, d\theta$ is determined by the time-averaged Poynting vector so that

$$dP = \frac{1}{2}\sqrt{\frac{\varepsilon_0\varepsilon_d}{\mu_0}}\left(\left|E_\varphi^F\right|^2 + \left|E_\theta^F\right|^2\right)r^2 d\Omega\,, \quad (27)$$

where $E_\varphi^F = E_\varphi^p + E_\varphi^Q + E_\varphi^m + \cdots$ and $E_\theta^F = E_\theta^p + E_\theta^Q + E_\theta^m + \cdots$. The differential scattering cross section $\sigma(\varphi,\theta)$ is determined by the expression $\sigma(\varphi,\theta)d\Omega = dP/I$, where $I = \sqrt{(\varepsilon_0\varepsilon_d/\mu_0)}|\mathbf{E}_0|^2/2$ is the radiation flux of the incident (external) plane wave. The total scattered power P to the far-field zone is obtained from Eq. (27) by integrating over the total solid angle. Contributions of higher multipole modes to the scattered far fields can be estimated in a similar manner (see Appendix).

III. NUMERICAL DEMONSTRATION

We now demonstrate the capabilities of the developed approach, considering different examples of scattering objects. Recently, it has been shown that spherical silicon particles with radii between 50 and 100 nm have two Mie resonances (electric

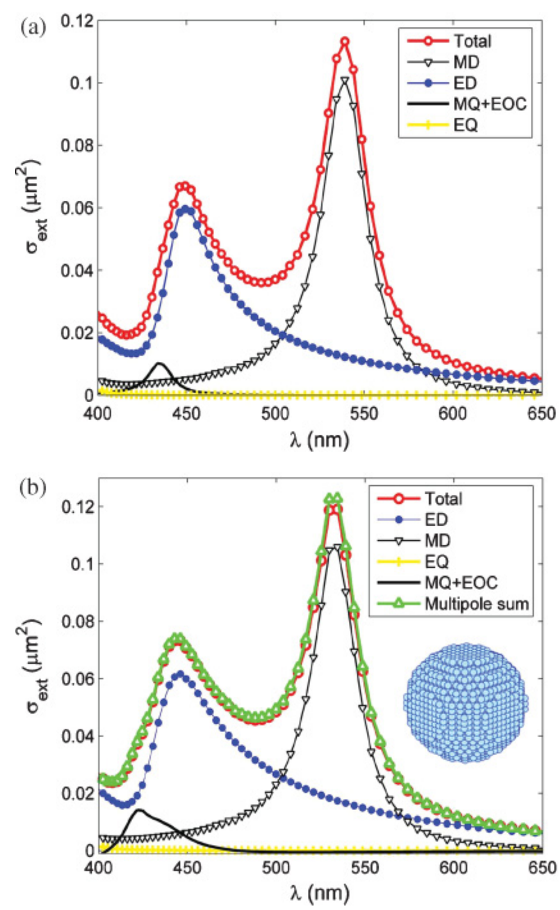

FIG. 2. (Color online) Extinction cross-section spectra of silicon spherical nanoparticles in air. (a) Mie theory; (b) DDA. The particle radius is $R_p = 63$ nm. The plane wave is linearly polarized. The plot shows different multipole contributions to the total ECS.

dipole and magnetic dipole) in the visible spectral range.[47] A test simulation of the total ECSs using the decomposed DDA method (DDDA) for spherical silicon particles shows very good agreement with results obtained by Mie theory, with respect to the contributions of electric and magnetic dipole modes, as shown in Fig. 2. The numerical calculations

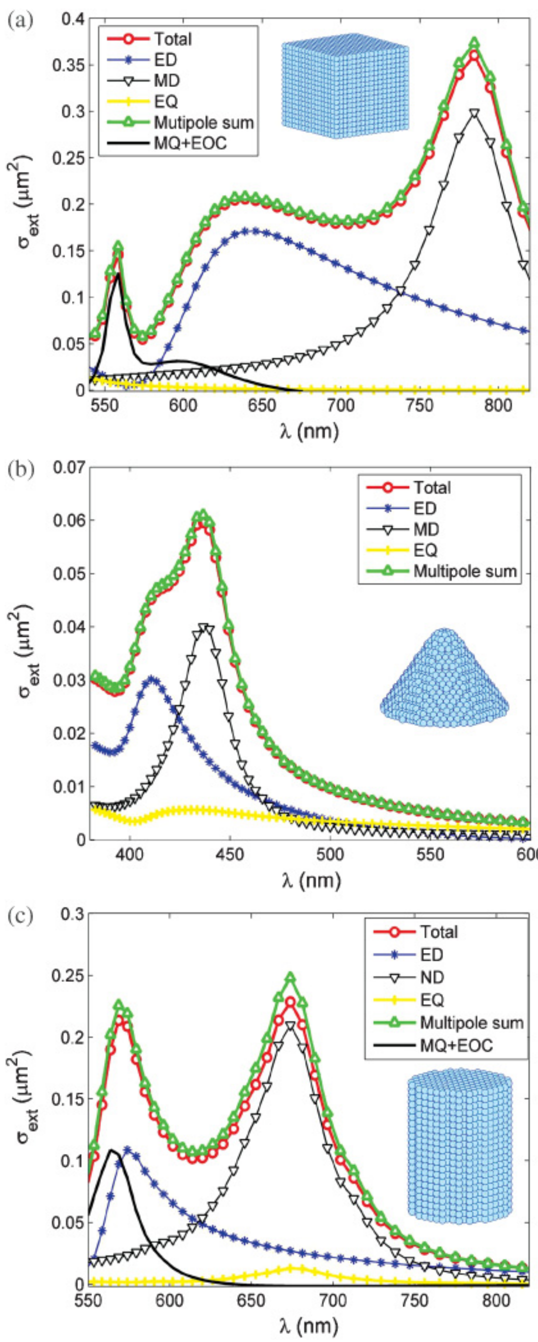

FIG. 3. (Color online) Extinction cross-section spectra of cubic (a), conical (b), and cylindrical(c) silicon nanoparticles in air. Cubic particle: side dimension 160 nm. Conical particle: height 100 nm, diameter 130 nm. Cylindrical particle: height 200 nm, diameter 130 nm. Incident plane wave is linearly polarized and propagates along the symmetrical axis of the particles. The graphs present contributions of different multipole modes excited in the particles.

are carried out with a discretization step of 6.5 nm. The small deviations between Mie theory and DDDA in the short-wavelength range result from the discrete approximation of the spherical shape. ECSs of nonspherical silicon particles are presented in Fig. 3. The particles have several resonances depending on their sizes and shapes. Decomposition of the ECS into different multipole contributions allows classifying the resonances and evaluating their relative efficiencies. With this new method it becomes obvious that the cubic particle [Fig. 3(a)] has an isolated resonance at ≃560 nm, which can basically be attributed to the MQ-EOC term of the multipole expansion, whereas for the cylindrical particle, the ED and MQ-EOC resonances overlap [Fig. 3(c)]. For the conical particle, the ED and MD resonances are very close [Fig. 3(b)], resulting in a single peak in the total ECS. Taking into account that for spherical Si particles the resonance of the MQ-EOC term [see the black solid curve in Fig. 2(a)] corresponds to the MQ mode, which follows from Mie theory, we assume that the MQ-EOC resonances in Fig. 3 are also determined by the excitation of the magnetic type mode. Small deviations between the total cross sections and the multipole sums in Fig. 3 can be attributed to the fact that these sums include only three first terms of the Taylor series (3). Indeed, the maximum deviations are observed for the cylindrical particle

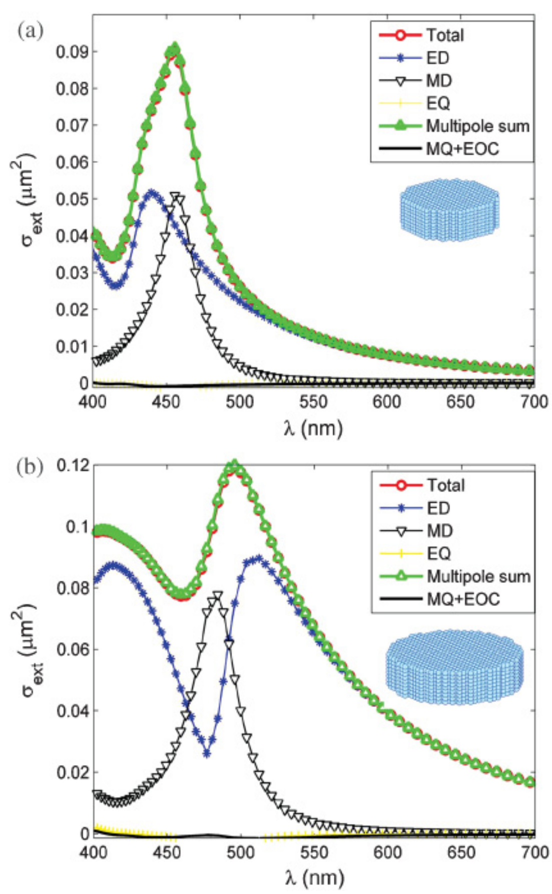

FIG. 4. (Color online) Extinction cross-section spectra of silicon cylindrical nanoparticles in air. Cylindrical particles: (a) height 60 nm, diameter 130 nm; (b) height 60 nm, diameter 170 nm. The incident plane wave has linear polarization and propagates along the symmetrical axis of the particles.

[Fig. 3(c)] elongated parallel to the incident wave. In this case, the expansion parameter, particle size divided by optical wavelength, is relatively large which requires high-order multipole corrections. If the height of the cylindrical particles decreases, the influence of the high-order multipole terms also decreases, as can be seen in Fig. 4, where the total cross sections and the multiple sums coincide.

IV. CONCLUDING REMARKS

The proposed DDDA method obtains insight into the multipole composition of resonant peaks. It can be used to control the positions of different multipole resonances as a function of particle size and shape. For example, Fig. 4 demonstrates that by changing parameters of cylindrical Si nanoparticles, one can obtain nanoparticle dimensions for which the ED and MD resonances are located at the same wavelength or at different wavelengths with well-controlled spectral separation. This unique property of Si nanoparticles opens new ways for the construction of novel nano-optical elements and can be particularly important for solving the problem of low-loss negative index metamaterials. The reported results demonstrate high potential of the developed DDDA method for studies of scattering properties of arbitrary shaped nanoparticles. This method can easily be generalized to many nanoparticle systems and clusters located in different environments.

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APPENDIX: QUADRUPOLE PROPAGATOR AND SCATTERED FIELDS

Let us consider the electric field **E** at a point **r** in homogeneous medium with the dielectric constant ε_d created by an electric dipole located at a point $\mathbf{r}'$ (Fig. 5). This field is determined by the expression[41]

$$\mathbf{E}(\mathbf{r}) = \frac{k_0^2}{\varepsilon_0}\,\hat{G}(\mathbf{r},\mathbf{r}')\mathbf{p}, \tag{A1}$$

where Green's tensor $\hat{G}(\mathbf{r},\mathbf{r}') = \hat{G}_N(\mathbf{r},\mathbf{r}') + \hat{G}_I(\mathbf{r},\mathbf{r}') + \hat{G}_F(\mathbf{r},\mathbf{r}')$ is determined by Eqs. (15)–(17). We consider that

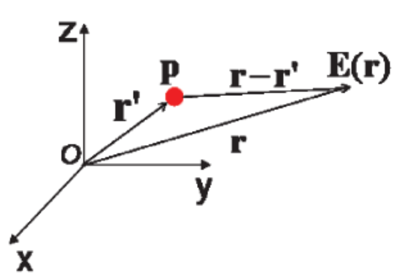

FIG. 5. (Color online) Position of the dipole **p** in the chosen coordinate system.

$r' \ll r$ and $r' \ll \lambda'$ (where λ' is the wavelength, in the medium with ε_d, of the radiated waves by the dipole) and expand Green's tensor in a Taylor series around the point $r' = 0$ up to the linear terms with respect to the small parameters r'/r and r'/λ' (the ratio r/λ' can be arbitrary).

For the near-field part of Green's tensor, one can write

$$\begin{aligned}\hat{G}_N(\mathbf{r},\mathbf{r}')\mathbf{p} &\approx \left(-\hat{U} + 3\mathbf{n}\otimes\mathbf{n} + 6\frac{(\mathbf{n}\cdot\mathbf{r}')}{r}\mathbf{n}\otimes\mathbf{n} - \frac{3}{r}(\mathbf{n}\otimes\mathbf{r}' + \mathbf{r}'\otimes\mathbf{n})\right)\frac{e^{ik_d r}}{4\pi k_d^2 r^3} \\ &\quad\times\left(1 - ik_d\mathbf{n}\cdot\mathbf{r}' + 3\frac{\mathbf{n}\cdot\mathbf{r}'}{r}\right)\mathbf{p} \\ &\approx (-\hat{U} + 3\mathbf{n}\otimes\mathbf{n})\frac{e^{ik_d r}}{4\pi k_d^2 r^3} \\ &\quad\times\left(\mathbf{p} - ik_d(\mathbf{n}\cdot\mathbf{r}')\mathbf{p} + 3\frac{(\mathbf{n}\cdot\mathbf{r}')\mathbf{p}}{r}\right) \\ &\quad+ \frac{e^{ik_d r}}{4\pi k_d^2 r^3}\left(6\frac{(\mathbf{n}\cdot\mathbf{r}')}{r}(\mathbf{n}\otimes\mathbf{n})\mathbf{p} - \frac{3}{r}[(\mathbf{n}\otimes\mathbf{r}')\mathbf{p} + (\mathbf{r}'\otimes\mathbf{n})\mathbf{p}]\right),\end{aligned} \tag{A2}$$

where $\mathbf{n} = \mathbf{r}/r$. In the following calculations, we use the identity

$$(\mathbf{a}\otimes\mathbf{b})\mathbf{c} \equiv \mathbf{a}(\mathbf{b}\cdot\mathbf{c}), \tag{A3}$$

where **a**, **b**, and **c** are arbitrary vectors, and the definitions of the electric quadrupole tensor and magnetic dipole vector (see below). Using the vector identity $[\mathbf{a}\times[\mathbf{b}\times\mathbf{c}]] = \mathbf{b}(\mathbf{a}\cdot\mathbf{c}) - \mathbf{c}(\mathbf{a}\cdot\mathbf{b})$ one can write

$$(\mathbf{n}\cdot\mathbf{r}')\mathbf{p} = \frac{1}{2}[\mathbf{r}'(\mathbf{n}\cdot\mathbf{p}) + (\mathbf{n}\cdot\mathbf{r}')\mathbf{p}] + \frac{1}{ik_d v_d}[\mathbf{n}\times\mathbf{m}], \tag{A4}$$

where $v_d = 1/\sqrt{\varepsilon_0\varepsilon_d\mu_0}$ is the light speed in homogeneous medium with ε_d, and

$$\mathbf{m} = -\frac{i\omega}{2}[\mathbf{r}'\times\mathbf{p}] \tag{A5}$$

is the magnetic moment associated with the dipole **p** [this expression is obtained using the expression[39] for the magnetic moment of a current density $\mathbf{J}(\mathbf{r}) = -i\omega\mathbf{p}\delta(\mathbf{r}-\mathbf{r}')$]. By direct verification one can prove the equality

$$3[\mathbf{r}'(\mathbf{n}\cdot\mathbf{p}) + (\mathbf{n}\cdot\mathbf{r}')\mathbf{p}] = \hat{Q}\mathbf{n} + 2\mathbf{n}(\mathbf{r}'\cdot\mathbf{p}), \tag{A6}$$

where the elements of the traceless quadrupole moment tensor $\hat{Q}$ associated with the dipole **p** are presented by

$$\begin{aligned}Q_{\beta\beta} &= 2(2\beta' p_\beta - \gamma' p_\gamma - \tau' p_\tau), \\ Q_{\beta\gamma} &= 3(\beta' p_\gamma + \gamma' p_\beta),\end{aligned} \tag{A7}$$

where $\beta = x,y,z$, and $\gamma = x,y,z$, and $\tau = x,y,z$; moreover, $\beta \neq \gamma$, $\beta \neq \tau$, and $\gamma \neq \tau$. After some tedious but straightforward transformations, using the expression for the magnetic dipole and electric quadrupole moments, one gets for the near-field part

$$\hat{G}_N(\mathbf{r},\mathbf{r}')\mathbf{p} \approx (-\hat{U} + 3\mathbf{n}\otimes\mathbf{n})\frac{e^{ik_d r}}{4\pi k_d^2 r^3}\,\mathbf{p} + \frac{e^{ik_d r}}{4\pi k_d^2 r^3}\left\{\frac{1}{v_d}[\mathbf{n}\times\mathbf{m}] - ik_d\frac{2\mathbf{n}(\mathbf{r}'\cdot\mathbf{p})}{3} + \left[\left(ik_d - \frac{6}{r}\right)\hat{U} + \left(\frac{15}{r} - 3ik_d\right)\mathbf{n}\otimes\mathbf{n}\right]\frac{\hat{Q}\mathbf{n}}{6}\right\}. \tag{A8}$$

Applying this approach to the intermediate-field and far-field parts of the expression (A1), one obtains

$$\hat{G}_I(\mathbf{r},\mathbf{r}')\mathbf{p} \approx (\hat{U} - 3\mathbf{n}\otimes\mathbf{n})\frac{ie^{ik_d r}}{4\pi k_d r^2}\,\mathbf{p} + \frac{ie^{ik_d r}}{4\pi k_d r^2} \times \left\{\left[-\frac{1}{v_d} - \frac{1}{ik_d v_d r}\right][\mathbf{n}\times\mathbf{m}] + \left[ik_d + \frac{1}{r}\right]\frac{2\mathbf{n}(\mathbf{r}'\cdot\mathbf{p})}{3} + \left[\left(-ik_d + \frac{5}{r}\right)\hat{U} + \left(-\frac{12}{r} + 3ik_d\right)\mathbf{n}\otimes\mathbf{n}\right]\frac{\hat{Q}\mathbf{n}}{6}\right\}, \tag{A9}$$

and

$$\hat{G}_F(\mathbf{r},\mathbf{r}')\mathbf{p} \approx (\hat{U} - \mathbf{n}\otimes\mathbf{n})\frac{e^{ik_d r}}{4\pi r}\,\mathbf{p} + \frac{e^{ik_d r}}{4\pi r}\left\{-\frac{1}{v_d}[\mathbf{n}\times\mathbf{m}] + \frac{2\mathbf{n}(\mathbf{r}'\cdot\mathbf{p})}{3r} + \left[\left(-ik_d + \frac{2}{r}\right)\hat{U} + \left(-\frac{3}{r} + ik_d\right)\mathbf{n}\otimes\mathbf{n}\right]\frac{\hat{Q}\mathbf{n}}{6}\right\}. \tag{A10}$$

Finally, the electric field (A1) can be presented approximately as a superposition of the electric fields created by the electric dipole $\mathbf{p}$, magnetic dipole $\mathbf{m}$, and the electric quadrupole $\hat{Q}$ located at the origin of the coordinate system (Fig. 5):

$$\mathbf{E}(\mathbf{r}) = \frac{k_0^2}{\varepsilon_0}[\hat{G}_N(\mathbf{r},\mathbf{r}') + \hat{G}_I(\mathbf{r},\mathbf{r}') + \hat{G}_F(\mathbf{r},\mathbf{r}')]\mathbf{p} \approx \mathbf{E}_p(\mathbf{r}) + \mathbf{E}_m(\mathbf{r}) + \mathbf{E}_Q(\mathbf{r})\,, \tag{A11}$$

where

$$\mathbf{E}_p(\mathbf{r}) = \frac{k_0^2 e^{ik_d r}}{4\pi\varepsilon_0 r}\left\{\left(1 + \frac{i}{k_d r} - \frac{1}{k_d^2 r^2}\right)\hat{U} + \left(-1 - \frac{i3}{k_d r} + \frac{3}{k_d^2 r^2}\right)\mathbf{n}\otimes\mathbf{n}\right\}\mathbf{p}\,, \tag{A12}$$

$$\mathbf{E}_m(\mathbf{r}) = \sqrt{\frac{\mu_0}{\varepsilon_0}}\frac{k_0 k_d e^{ik_d r}}{4\pi r}\left(1 + \frac{i}{k_d r}\right)[\mathbf{m}\times\mathbf{n}]\,, \tag{A13}$$

$$\mathbf{E}_Q(\mathbf{r}) = \frac{k_0^2}{\varepsilon_0}\left\{\left(-1 - \frac{i3}{k_d r} + \frac{6}{k_d^2 r} + \frac{i6}{k_d^3 r^3}\right)\hat{U} + \left(1 + \frac{i6}{k_d r} - \frac{15}{k_d^2 r^2} - \frac{i15}{k_d^3 r^3}\right)\mathbf{n}\otimes\mathbf{n}\right\}\frac{ik_d e^{ik_d r}}{24\pi r}\hat{Q}\mathbf{n}\,. \tag{A14}$$

Formally, Eqs. (A12)–(A14) determine electric fields in all wave zones created by point dipole and quadrupole sources located in the origin of the coordinate system. If we assume that these sources are located at the point $\mathbf{r}_c$, we get expressions (20)–(22) with the quadrupole propagator determined by Eq. (23).

The problem is significantly simplified if we consider electric fields only in the far-field zone (in the radiation zone $r \gg \lambda \gg r'$). In this case, we should take only zero-order terms with respect to r'/r in the Taylor series of the far-field part of Green's tensor. The electric field at the point $\mathbf{r}$ radiated by the electric dipole $\mathbf{p}$ located at the point $\mathbf{r}'$ is given by

$$\mathbf{E}^F(\mathbf{r}) = (\hat{U} - \mathbf{n}\otimes\mathbf{n})\frac{k_0^2 e^{ik_d r}}{4\pi\varepsilon_0 r}e^{-ik_d(\mathbf{n}\cdot\mathbf{r}')}\mathbf{p}. \tag{A15}$$

To obtain the multipole presentation one should expand the factor $\exp[-ik_d(\mathbf{n}\cdot\mathbf{r}')]$ with respect to the small parameter r'/λ'. The zero-order and the first terms correspond to the electric dipole contribution and both magnetic dipole and electric quadrupole contributions, respectively. Second-order term corresponds to the magnetic quadrupole and electric octupole contributions and so on. Including the MQ and EOC terms, the electric field can be presented in the following form:[25,46]

$$\mathbf{E}^F(\mathbf{r}) = \frac{k_0^2 e^{ik_d r}}{4\pi\varepsilon_0 r}\Big([\mathbf{n}\times[\mathbf{p}\times\mathbf{n}]] + \frac{ik_d}{6}[\mathbf{n}\times[\mathbf{n}\times\hat{Q}\mathbf{n}]] + \frac{1}{v_d}[\mathbf{m}\times\mathbf{n}] + \frac{ik_d}{2v_d}[\mathbf{n}\times(\hat{M}\mathbf{n})] + \frac{k_d^2}{6}[\mathbf{n}\times[\mathbf{n}\times((\hat{O}\mathbf{n})\mathbf{n})]]\Big), \tag{A16}$$

where

$$\hat{M} = \frac{-2i\omega}{3}[\mathbf{r}'\times\mathbf{p}]\otimes\mathbf{r}' \equiv \frac{4}{3}\mathbf{m}\otimes\mathbf{r}'$$

is the magnetic quadrupole moment at the origin of the coordinate system associated with the dipole $\mathbf{p}$ located at the point $\mathbf{r}'$. The symmetrical tensor $\hat{O}$ is the electric octupole moment with components

$$O_{\beta\beta\beta} = 3p_\beta\beta'^2\,, \quad O_{\beta\gamma\tau} = p_\beta\gamma'\tau' + p_\gamma\beta'\tau' + p_\tau\beta'\gamma',$$
$$O_{\beta\beta\gamma} = O_{\beta\gamma\beta} = O_{\gamma\beta\beta} = 2p_\beta\beta'\gamma' + p_\gamma\beta'^2,$$

where $\beta = x,y,z$, and $\gamma = x,y,z$, and $\tau = x,y,z$; moreover, $\beta \neq \gamma$, $\beta \neq \tau$, and $\gamma \neq \tau$. Here $\hat{M}$ is traceless, but not symmetric; while $\hat{O}$ is totally symmetric, but not traceless.[25] Note that in our considerations we use the definition of the traceless electric quadrupole moment $\hat{Q}$ from Ref. 39 that differs from the quadrupole moment considered in Ref. 25 by a factor of 3.

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