Describing meta-atoms using the exact higher-order polarizability tensors

Jungho Mun,1 Sunae So,2 Jaehyuck Jang,1 and Junsuk Rho1,2 ∗

1Department of Chemical Engineering, Pohang University of Science and Technology (POSTECH), 77 Chungam-ro, Republic of Korea
2Department of Mechanical Engineering, Pohang University of Science and Technology (POSTECH), 77 Chungam-ro, Republic of Korea
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In nanophotonics, multipole approach has become an indispensable theoretical framework for analyzing subwavelength meta-atoms and their radiation properties. Thus far, induced multipole moments have frequently used to illustrate the radiating properties of the meta-atoms, but they are excited at a specific illumination and do not fully represent anisotropic meta-atoms. On the other hand, dynamic polarizability (α) tensors contain complete scattering information of the meta-atoms, but have not often been considered due to complicated retrieval procedures. In this study, we suggest that exact higher-order α-tensor can be efficiently obtained from T-matrix using simple basis transformation. These higher-order α-tensors are necessary to describe recently reported coupled plasmonic and high-refractive-index particles, which we demonstrate from their retrieved α-tensors. Finally, we show that description of meta-atoms using α-tensors incorporated with multiple-scattering theory vastly extends the applicability of the multipole approach in nanophotonics, allowing accurate and efficient depiction of complicated, random, multi-scale systems.

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I. INTRODUCTION

Under the paradigm of metamaterial, its constituent meta-atoms and their configurations determine the material properties. The meta-atoms have been efficiently analyzed using the multipole decomposition technique [1–2], because a few low-order multipole moments efficiently reconstruct the electromagnetic radiation and the relevant physics from a subwavelength localized current charge source. Due to this feature, the multipole approach has become an useful and indispensable tool for nanophotonics [3]. Manipulation of light in the nanoscale has been facilitated by interference of multipole radiations, which provides the underlying principles behind many optical phenomena and relevant applications. Notably, the multipole approach has given insights on directional scattering [4], lattice Kerker effects [5], non-radiating anapoles [6, 7], lattice invisibility effects [8], Fano-like resonances [9, 10], optical anti-ferromagnetism [9], optical nonlinearity [11], radiative heat transfer, weak localization [12], photonic topological insulators [13], and bound states in the continuum [14].

Formulation of multipole radiation is a textbook problem [15], but given the importance of the multipole approach, expressions for multipole are still under research [2–10, 17] with possibility on toroidal multipoles as an extra multipole family [6, 18]. In general, the multipoles under discussion are excited at a specific illumination, but they do not provide complete information of highly anisotropic meta-atoms. On the other hand, dynamic polarizability α tensor (or transition T matrix) maps the induced multipole modes at arbitrary incident fields, and has been used to treat scattering objects in many different fields including optics, acoustics, and astrophysics [12, 19, 20]. In nanophotonics, analysis of meta-atoms based on their T-matrix started to become remarked rather recently [17, 21]. It has been pointed out that complicated coupled configurations involving multiple meta-atoms can be efficiently studied by describing the meta-atoms in terms of α-tensor (or T-matrix) and using the multiple-scattering theory (MST) [17]. Electromagnetically coupled discrete scattering objects can be self-consistently treated to describe for collective responses of multiple particles [22–24] and periodic particle arrays [25–31], and this approach significantly reduces the calculation loads for complicated, random [32, 33], or multi-scale systems [34, 36].

In the following work, we first discuss induced multipoles in different expressions: approximate Cartesian, exact Cartesian, and spherical multipoles, where exact Cartesian and spherical multipoles are essentially identical with different choice of basis [10]. In the next section, we present expression of local fields and field gradients in terms of spherical multipoles. This naturally leads us to obtain transformation between α-tensor and T-matrix. This basis transformation is used to analyze meta-atoms based on their α-tensors, whose properties can be intuitively interpreted due to the Cartesian basis. Finally, we show that the MST allows efficient and accurate description of electromagnetically coupled meta-atoms, and that analytic scattering objects can be implemented under the multipole approach.

II. EXACT CARTESIAN MULTipoles

In standard electrodynamic textbooks, the spherical multipoles appear from the multipole decomposition of
electromagnetic fields using the vector spherical wave functions (VSWFs) as the basis \[13\]. Because the VSWFs span the vector fields satisfying the transverse Helmholtz type equations, the electromagnetic fields in a homogeneous media can be exactly reconstructed, and the renowned Mie theory is also based on this expansion. Because of the difficulty in interpreting the spherical basis, the spherical multipoles are not directly analyzed per se, but their associated scattering power or radiation fields are.

Therefore, multipole approach in nanophotonics most frequently utilizes the approximate expressions for localized charge-current density multipoles in the Cartesian basis, which are sufficiently straightforward and resemble the expressions in electrostatics and magnetostatics. Despite their popularity, the approximate Cartesian multipoles cannot exactly reconstruct the electrodynamic radiation fields and the related scattering phenomena \[16\]. Scattering from subwavelength nanoparticles with moderate refractive-index generally shows good agreement, but the error grows for larger particles and high-refractive-index particles. This error has been corrected by toroidal multipoles, which appear from multipole decomposition of the localized current sources \[10\] \[37\] \[38\]. However, it has been pointed out that the radiation fields from toroidal multipoles do not have independent (orthogonal) basis to those from electric and magnetic multipoles \[16\]. Therefore, it is controversial whether to treat the toroidal multipoles as the third multipole family \[18\], or as a correction to the basic Cartesian multipoles.

Recently, exact expressions for the localized charge-current density multipoles in the Cartesian basis up to MQ were developed without relying on the toroidal multipoles \[16\] \[39\]. For completeness, we present expressions of the exact Cartesian multipoles up to MO as

\[
d^E_\alpha = -\frac{1}{i\omega} \int d^3r \left\{ \alpha r J_{\alpha\beta}(kr) + \frac{k^2}{2} \left[ 3(r \cdot J)r_\alpha - r^2 J_\alpha \right] \frac{j_2(kr)}{(kr)^2} \right\}
\]

\[
d^M_\alpha = \frac{3}{2} \int d^3r (r \times J)_\alpha \frac{j_1(kr)}{kr}
\]

\[
Q^E_{\alpha\beta} = -\frac{3}{i\omega} \int d^3r \left\{ \frac{2}{3} \delta_{\alpha\beta} - \frac{2}{3} \frac{1}{k^2} \left[ 5(r \cdot J)r_\alpha r_\beta - r^2 (r_\alpha J_\beta + r_\beta J_\alpha) - r^2 \delta_{\alpha\beta} (r \cdot J) \right] \frac{j_1(kr)}{kr^2} \right\}
\]

\[
Q^M_{\alpha\beta} = 5 \int d^3r \left[ r_\alpha (r \times J)_\beta + r_\beta (r \times J)_\alpha \right] \frac{j_2(kr)}{(kr)^2}
\]

where \(\alpha, \beta, \gamma = x, y, z\), and the multipoles are symmetrical and traceless \[10\]. The familiar approximate Cartesian multipoles and toroidal multipoles can be readily obtained by taking the long wavelength limit \[16\]. See Appendix A for more details on Cartesian multipoles.

An important result from Ref. \[16\] is that the exact Cartesian multipole can be expressed from the spherical multipoles. They have identical physical meaning, but are expressed in different basis, which can be systemati-
\[ \begin{bmatrix} O_{xx}^p, O_{xy}^p, O_{yx}^p, O_{yy}^p, O_{yz}^p, O_{zy}^p, O_{zz}^p, O_{g}^p, O_{gy}^p \end{bmatrix}^T, \quad b^p = \begin{bmatrix} b^n_{-n}, b^n_{-n+1}, \cdots, b^n_{-n-1}, b^n_{n+1} \end{bmatrix}^T \]

where superscript \( p \) = \( E \) or \( M \) denoting electric or magnetic multipoles, respectively. The basis transformation matrices are introduced as

\[
\tilde{O}_1 = \begin{bmatrix} 1 & 0 & -1 \\ -i & 0 & 0 \\ 0 & \sqrt{2} & 0 \end{bmatrix} \quad (3a)
\]

\[
\tilde{O}_2 = \begin{bmatrix} 1 & 0 & -\sqrt{2/3} & 0 & 1 \\ -i & 0 & 0 & 0 & i \\ 0 & 1 & 0 & -\sqrt{2/3} & 0 \\ -1 & 0 & 0 & -i & 0 \\ 0 & -i & 0 & 0 & i \end{bmatrix} \quad (3b)
\]

\[
\tilde{O}_3 = \begin{bmatrix} 1 & 0 & -\sqrt{3/5} & 0 & \sqrt{3/5} & 0 & -1 \\ -i & 0 & i\sqrt{1/5} & 0 & i\sqrt{1/5} & 0 & i \\ 0 & \sqrt{2/3} & 0 & -\sqrt{4/5} & 0 & \sqrt{2/3} & 0 \\ -1 & 0 & -i\sqrt{1/5} & 0 & i\sqrt{1/5} & 0 & i \\ 0 & i\sqrt{3/5} & 0 & 0 & i\sqrt{3/5} & 0 & i \\ -1 & 0 & -\sqrt{4/5} & 0 & \sqrt{2/3} & 0 & -1 \end{bmatrix} \quad (3c)
\]

The transformation relations for ED, MD, EQ cases (Eqns. [2a–c]) can be found in previous work [1, 2], although the expression for exact Cartesian multipoles has only been published recently [10, 39].

Finally, the farfield radiated fields from the multipoles are

\[
E = \frac{k^2}{4\pi \epsilon} e^{ikr} \left[ n \times (d^E \times n) - \frac{i}{2} k (\vec{Q}^E \times n) - \frac{k^2}{4} n \times (\vec{Q}^E \times n) \right]
\]

\[
+ \frac{\eta k^2}{4\pi} e^{ikr} \left[ d^M \times n - \frac{i}{2} k (\vec{Q}^M \times n) - \frac{k^2}{4} (\vec{Q}^M \times n) \right] \quad (4a)
\]

\[
H = -\frac{\omega k e^{ikr}}{4\pi} \left[ d^E \times n - \frac{i}{2} k (\vec{Q}^E \times n) - \frac{k^2}{4} (\vec{Q}^E \times n) \right]
\]

\[
+ \frac{k^2}{4\pi} e^{ikr} \left[ n \times (d^M \times n) - \frac{i}{2} k (\vec{Q}^M \times n) - \frac{k^2}{4} n \times (\vec{Q}^M \times n) \right] \quad (4b)
\]

where \( \vec{Q}^p = Q^p_{\alpha\beta\gamma} n_{\beta} \) and \( \vec{Q}^p = O^p_{\alpha\beta\gamma} n_{\beta} n_{\gamma} \). Total radiation power by multipoles is

\[
P = \frac{\omega k^3}{12\pi} \sum_{\alpha} |d^E_{\alpha}|^2 + \frac{\omega k^5}{160\pi} \sum_{\alpha\beta} |Q^E_{\alpha\beta\gamma}|^2 + \frac{\omega k^7}{1680\pi} \sum_{\alpha\beta\gamma} |O^E_{\alpha\beta\gamma}|^2
\]

\[
+ \frac{\eta k^4}{12\pi} \sum_{\alpha} |d^M_{\alpha}|^2 + \frac{\eta k^6}{160\pi} \sum_{\alpha\beta} |Q^M_{\alpha\beta\gamma}|^2 + \frac{\eta k^8}{1680\pi} \sum_{\alpha\beta\gamma} |O^M_{\alpha\beta\gamma}|^2 \quad (5)
\]

Note that the proportionality constants in Eqns. [4] and [5] are different from previous work [38], but the reconstructed scattered fields satisfying the Maxwell’s equations should be identical regardless of the choice of the notations. Also, the notations of the spherical multipoles and VSWFs differ by publications, and our expression is given in Appendix [5].

**III. EXTENDED POINT POLARIZABILITY AND T-MATRIX**

Many optical phenomena have been successfully resolved from induced multipoles, but they are given at a specific illumination. In general, the induced moments are not invariant under the interaction with other particles or under different external excitation fields. Considering that meta-atoms generally have strong anisotropy and can therefore exhibit different induced multipoles depending on its environment and excitation fields, the induced moments obtained from a specific situation do not consistently represent the inherent properties of the meta-atoms.

Hence, a different quantity is required to consistently describe an identical particle in an isolated state, nearby other particles, or in a lattice, and \( \alpha \)-tensor serves for this purpose. \( \alpha \)-tensor is defined as a response tensor linearly relating the local fields to the induced multipoles, so it is irrelevant of excitation conditions and allows us to calculate the induced moments at arbitrary incident fields. This feature allows us to calculate the collective responses of coupled particles using MST, which we discuss in later sections. However, \( \alpha \)-tensor is usually truncated at dipole order [40, 42], and higher-order \( \alpha \)-tensor, which includes higher-order multipole moments and field gradients [43], is rarely utilized due to the compensated retrieval process if not for spheres with isotropic responses [29].

These higher-order multipole transitions have been systematically treated using T-matrix, which linearly relates the spherical multipoles of incident field to those of scattered field and is expressed in matrix form as

\[
\begin{bmatrix} b^E \\ b^M \end{bmatrix} = \begin{bmatrix} T^E_{p} \\ T^M_{p} \end{bmatrix} \begin{bmatrix} a^E \\ a^M \end{bmatrix} \quad (6a)
\]

\[
T^p_{p'} = \begin{bmatrix} T^p_{p'1} & T^p_{p'2} & \cdots \end{bmatrix} \quad (6b)
\]

where \( T^p_{n'n'} \) is a \( (2n+1) \times (2n'+1) \) matrix corresponding to the transition from multipole order \( n \) of mode \( p \) to \( n' \) of \( p' \). \( \alpha \)- and \( \beta \)- are vectors containing spherical multipoles of incident and scattered fields, respectively [44].

Analogous to the exact Cartesian multipoles and the spherical multipoles, \( \alpha \)-tensor and T-matrix are identical but with different choice of basis. To transform between them, we additionally need to express the local fields and field gradients in terms of spherical multipoles, which we
present using the same transformation matrices as
\[
E = \frac{iE_0}{\sqrt{12} \pi} \tilde{O}_1 a_1^E \quad \quad \quad H = \frac{E_0}{\sqrt{12} \pi} \tilde{O}_1 a_1^H \quad (7a)
\]
\[
\frac{1}{k} \tilde{O} E = \frac{iE_0}{\sqrt{80} \pi} \tilde{O}_2 a_2^E \quad \quad \quad \frac{1}{k} \tilde{O} H = \frac{E_0}{\sqrt{80} \pi} \tilde{O}_2 a_2^H \quad (7b)
\]
\[
\frac{1}{k^2} \tilde{O}^2 E = \frac{iE_0}{\sqrt{120} \pi} \tilde{O}_3 a_3^E \quad \quad \quad \frac{1}{k^2} \tilde{O}^2 H = \frac{E_0}{\sqrt{120} \pi} \tilde{O}_3 a_3^H \quad (7c)
\]
where \( E = [E_x, E_y, E_z]^\top \), \( \tilde{O} E = (\partial_x E_x, (\partial_x E_y + \partial_y E_x)/2, (\partial_x E_z + \partial_z E_x)/2, \partial_y E_y, (\partial_y E_z + \partial_z E_y))/2 \)^\top, and
\[
\tilde{O}^2 E = \left[ \begin{array}{l}
\frac{1}{2}(\partial_x^2 E_x - \frac{1}{2}\nabla^2 E_x) \\
\frac{1}{2}(\partial_y^2 E_y + 2\partial_x \partial_y E_x) - \frac{1}{2}\nabla^2 E_y \\
\frac{1}{2}(\partial_z^2 E_z + 2\partial_x \partial_z E_x) - \frac{1}{2}\nabla^2 E_z \\
\frac{1}{2}(\partial_y^2 E_y + 2\partial_y \partial_z E_z + \partial_z \partial_y E_z) \\
\frac{1}{2}(\partial_z^2 E_z + \frac{1}{2}\nabla^2 E_z)
\end{array} \right]
\]
\[
\tilde{O}^2 H = \left[ \begin{array}{l}
\frac{1}{2}(\partial_x^2 E_x) \\
\frac{1}{2}(\partial_y^2 E_y + 2\partial_x \partial_y E_x) \\
\frac{1}{2}(\partial_z^2 E_z + 2\partial_x \partial_z E_x) \\
\frac{1}{2}(\partial_y^2 E_y + 2\partial_y \partial_z E_z + \partial_z \partial_y E_y) \\
\frac{1}{2}(\partial_z^2 E_z)
\end{array} \right]
\]

Now, \( \alpha \)-tensor and \( T \)-matrix can be transformed to each other. However, \( \alpha \)-tensor is expressed in SI units if Eqns. \([7] \) and \([2] \) are directly used. Because \( \alpha \)-tensor in SI units have different units per components and are difficult to be compared, we use \( \alpha \)-tensor in units of volume defined as
\[
\alpha_{\mu \nu}^{pn} = \frac{6\pi}{k^3} \tilde{O}_n \mathbf{T}_{p'n}^{pn} \tilde{O}_n^{-1}
\]
\[
(8)
\]
Note that similar expressions for isotropic (scalar) dipolar \([40] \) and quaquaropular objects \([29] \) can be found in literature. \( T \)-matrix is inversely obtained from \( \alpha \)-tensor as
\[
\mathbf{T}_{p'n}^{pn} = \frac{i k^3}{6\pi} \tilde{O}_n^{-1} \alpha_{\mu \nu}^{pn} \tilde{O}_n
\]
\[
(9)
\]
Eqns. \([8] \) and \([9] \) are the central results of this study, which we use to express exact higher-order \( \alpha \)-tensors in units of volume (see Appendix \([C] \) for more details).

\( \alpha \)-tensor or \( T \)-matrix contains information on particle symmetries and conservation laws, as well as complete information on scattering by a particle. Symmetry refers to the invariance upon certain operations, and includes rotational symmetry, mirror symmetry, parity and time-reversal symmetry, and reciprocity. \( N \)-fold rotational symmetry and mirror symmetry can be confirmed by checking if \( T \)-matrix or \( \alpha \)-tensor is invariant under the transformation. For Onsager reciprocal system, \( T \)-matrix elements satisfy the following relationship:
\[
(T_{p'm}^{pn})_{mn} = (-1)^{m+m'}(T_{p'n}^{pm})_{-m}^{-m}
\]
\[
(10)
\]
This expression has been used to check the accuracy of numerically calculated \( T \)-matrix in literature \([44] \). \( \alpha \)-tensor of reciprocal dipolar particles satisfy:
\[
\alpha_{\mu \nu}^{pn} = (\alpha_{\mu \nu}^{pn})^\top, \quad \alpha_{\mu \nu}^{m1} = (\alpha_{\mu \nu}^{m1})^\top, \quad \text{and} \quad \alpha_{\mu \nu}^{m1} = (\alpha_{\mu \nu}^{m1})^\top
\]
\[
(22)
\]
The parity operation is given as \( r \rightarrow -r \). Upon the parity operation, \( E \rightarrow -E, \quad H \rightarrow H, \quad \nabla \rightarrow -\nabla \). Also, electric multipoles \( (a_{nm}^E \text{ and } b_{nm}^E) \) have parity of \((-1)^n \), and magnetic multipoles \( (a_{nm}^M \text{ and } b_{nm}^M) \) have \((-1)^{n+1} \). From which, we see that \( T_{E_{2n}}^{M_n} \) and \( T_{M_{2n}}^{E_n} \) have parity of \((-1)^{n+n'} \), and \( T_{E_{2n}}^{M_n} \) and \( T_{M_{2n}}^{E_n} \) has \((-1)^{n+n'+1} \). The corresponding \( \alpha \)-tensor components have the same parity. Also, parity is closely related to the concept of true chirality, or reciprocal parity-odd \([46] \). Note that chirality of chiral molecules have been embedded in the magneto-electric molecules \([35] \), which attributes to the reciprocal parity-odd property. By breaking the reciprocity, it is possible to undergo false chirality, which is nonreciprocal, parity-odd \([11] \). A lossless particle has no intrinsic absorption, so its extinction equals to scattering. For a lossless particle, \( T^{\top} T = -\frac{1}{3}(T^1 + T) \), where the superscript \( \dagger \) denotes Hermitian conjugate \([48] \). This expression has also been used to check the accuracy of \( T \)-matrix for lossless particles in literature \([44] \). \( \alpha \)-tensor of lossless dipolar particles satisfy:
\[
\frac{k^2}{\pi} \alpha_{1}^\dagger \alpha = \frac{i}{\pi} (\alpha^\dagger - \alpha)
\]
\[
(15)
\]
which reduces to the optical theorem \( k^2/\pi |\alpha|^2 = \text{Im}(\alpha) \) for dipolar scalar \( \alpha \) \([45] \). Studies on properties of higher-order \( \alpha \)-tensor is currently lacking and remains future research.

IV. META-ATOMS AND METAPHOTONICS

In metamaterials and metaphotonics, manipulation of light at the nanoscale utilizes optically resonant subwavelength meta-atoms, whose properties have generally been analyzed by dipolar \( \alpha \)-tensor \([10] \). However, recently emerged high-refractive-index particles \([5] \) \([8] \) \([27] \) \([28] \) \([19] \) and coupled plasmonic systems \([17] \) \([43] \) often involve higher-order multipole transitions. It should be noted that \( T \)-matrix of meta-atoms can be systematically retrieved for arbitrary multipole order \([17] \), but detailed analysis on their properties from their \( T \)-matrix is difficult due to the spherical basis, while retrieval of higher-order \( \alpha \)-tensor can be cumbersome \([43] \). In this section, we analyze several meta-atoms from their \( \alpha \)-tensors, which are transformed from \( T \)-matrix using Eqn. \([8] \). We will show that higher-order \( \alpha \)-tensor are necessary to describe anisotropic meta-atoms, whose properties can be more intuitively analyzed due to the Cartesian basis. Refer to Fig. \([1] \) for the interpretation of \( \alpha \)-tensor. In addition, \( \alpha \)-tensor allows analysis on several particle properties including anisotropy, symmetries, spectral modal resonances, and origin of chirality and optical magnetism.

Hybridized plasmonic structures can exhibit higher-order multipole modes even with subwavelength feature sizes \([17] \) \([43] \). Among them, plasmonic double bars (PDB) has exhibited strong MD mode even at the visible regime \([20] \). Multipole-decomposed scattering cross-section exhibits broad ED resonance at 490 nm and sharp EQ and MD resonances at 530 nm (Fig. \([2] \)) and the origins of the multipolar modes can be analyzed from retrieved \( \alpha \)-tensor (Fig. \([2] \)) and spectra of its components (Fig. \([2] \)). The broad ED resonance is easily attributed to \( (\alpha_{x1}^E)^1_x \) and \( (\alpha_{x1}^M)^1_x \), and the sharp EQ and MD resonances are spectrally attributed to \( (\alpha_{m1}^E)^2_{xy} \), \( (\alpha_{e2}^E)^2_{xy} \), and \( (\alpha_{e2}^M)^2_{xy} \). Analysis from \( \alpha \)-tensor allows us to see that...
PDB has different origin of optical magnetism from splitting resonators \[40\] and high-refractive-index spheres (see Appendix \[F\] for more details). Importantly, only a few components are dominant in the retrieved \(\alpha\)-tensor, making the analysis easier (Fig. 2). This simplification partly comes from the particle symmetry. Notably, the parity symmetry removes the half of the components: \(\alpha_{x1}^{m1}, \alpha_{z1}^{m1}, \alpha_{x1}^{m2}, \alpha_{z1}^{m2}, \alpha_{m1}^{x1}\), and \(\alpha_{m1}^{x2}\). Strong anisotropy of the meta-atom further simplifies the \(\alpha\)-tensor, especially in the Cartesian basis. In addition, the reciprocity enforces \((\alpha_{x2}^{m2})_{xy} = (\alpha_{m1}^{x1})_{zy}\). Interestingly, \((\alpha_{m1}^{m1})_{zy}\), \((\alpha_{c2}^{m2})_{zy}\), and \((\alpha_{c2}^{c2})_{zy}\) have very similar spectral feature resembling Lorentzian resonances, indicating that some components may additionally be coupled together possibly using singular value decomposition technique \[10\] or modular analysis \[47\].

Plasmonic chiral particles have exhibited chiral responses far-exceeding those from natural materials. Among them, a twisted double-bars (TDB) has been widely used to generate exceptionally strong chiral responses \[51\], which we assess from circular dichroism (CD). Different from PDB, TDB is geometrically chiral, and therefore parity-odd, and \(\alpha_{x1}^{m1}, \alpha_{x2}^{m2}, \alpha_{c2}^{x2}\), and \(\alpha_{c2}^{c1}\) transition components are now allowed. An important property of TDB is strongly anisotropic CD, which is only visible for the light propagation parallel to the twist-axis \((\phi)\). Interestingly, this anisotropic CD cannot be explained by dipolar \(\alpha\)-tensor alone, because \((\alpha_{c2}^{c1})_{zy}\) and \((\alpha_{c2}^{c2})_{zy}\) contribute to chiral response for both \(k \parallel x\) and \(k \parallel y\). We confirmed that \((\alpha_{x2}^{m1})_{zy}\) and \((\alpha_{x2}^{c1})_{zy}\) are necessary for this anisotropic CD; they constructively contribute to CD for \(k \parallel y\) (red line, Fig. 3i), whereas destructively for \(k \parallel x\) and pretty much eliminating the total CD (black line, Fig. 3i). We do not explicitly show this, but we mention that their contributions can be quantified by directly calculating CD from their retrieved \(\alpha\)-tensor. Here, TDB clearly shows that higher-order multipole transition is necessary for describing plasmonic meta-atoms and reconstructing anisotropic chiral responses even in far-fields, as well as in near-fields \[52\].

FIG. 1. Structure of \(\alpha\)-tensor. \(\alpha_{c2}^{m2}\) corresponds to the transition from incident multipole order \(m\) of type \(p\) to induced multipole order \(n\) of type \(p'\), and the incident and induced Cartesian modes are denoted outside the parenthesis. For instance, \((\alpha_{c2}^{m1})_{zy}\) relates incident \(E_x\) to induced \(d_{xy}^{p}\); \((\alpha_{c2}^{m2})_{zy}\) relates incident \(E_z\) to induced \(\mathcal{G}_{x} = (\partial_y H_z + \partial_z H_y)/2\); \((\alpha_{c2}^{m1})_{zy}\) relates incident \(\mathcal{G}_{x} = (\partial_y H_z + \partial_z H_y)/2\) to induced \(d_{zy}^{p}\).

FIG. 2. Plasmonic double-bars. (a) Schematics. (b) Retrieved \(\alpha\)-tensor at \(\lambda = 530\) nm. (c) Multipole-decomposed scattering cross-section at x-polarized planewave incidence propagating in y-direction. (d) Spectra of \((\alpha_{x1}^{m1})_{xy}, (\alpha_{m1}^{m1})_{xy}, (\alpha_{c2}^{m2})_{xy},\) and \((\alpha_{c2}^{c2})_{xy}\). Geometrical parameters are: radius 20 nm, length 100 nm, and gap distance 20 nm.

FIG. 3. (a) The schematics of twisted double-bars. (b) Retrieved \(\alpha\)-tensor at \(\lambda = 520\) nm. (c) Extinction and (d) circular dichroism at planewave incidence propagating in x-, y-, and z-directions. (e) Spectra of \((\alpha_{x1}^{m1})_{xy}, (\alpha_{x1}^{m2})_{xy}, (\alpha_{x2}^{m2})_{xy}, (\alpha_{x2}^{c2})_{xy}, \) and \((\alpha_{c2}^{c1})_{xy}\). Geometrical parameters are: radius 20 nm, length 100 nm, gap distance 20 nm, and twist angle 45°.
Finally, we briefly discuss high-refractive-index nanoparticles, which are actively researched topic due to their higher-order multipolar modes. In general, sub-wavelength plasmonic nanoparticles exhibit dominant ED mode, but high-refractive-index nanoparticles often exhibit dominant higher-order modes. For instance, Si quadrumer exhibits strong MD mode exceeding ED near 520 nm (Fig. 4). In addition, broad MQ response was observed between 420 and 520 nm. We analyzed the origin of MQ response from the $\alpha$-tensor components that can contribute to the MQ mode (Fig. 4b,d). Although the MQ mode shows a broad response, it originates from the sum of many resonances (Fig. 4d). This example shows that deeply fundamental properties of meta-atoms may be analyzed from their higher-order $\alpha$-tensors.

V. MULTIPLE-SCATTERING THEORY AND ELECTROMAGNETICALLY COUPLED SYSTEMS

In the previous section, we have shown that higher-order $\alpha$-tensor is necessary for analyzing isolated meta-atoms and interpreting their interaction with light. Additionally, $\alpha$-tensor can be used to model interacting meta-atoms for further research. In literature, self-consistent coupled multipole equations have been formulated using the Green’s tensor to illustrate periodic 2D arrays of plasmonic \[55\] and dielectric \[27,29\] spheres, and finite \[55,56\] or random \[31\] systems. In this approach, electromagnetic interactions between scattering objects are taken into account without any approximation, while single scattering objects are described by $\alpha$-tensors approximated to low-order multipole orders. This approach is simplified version of superposition $T$-matrix method (STMM), which has been extensively studied for coupled spheres \[22,24\]. Earlier studies have usually incorporated small spheres, whose $\alpha$ can be easily obtained using the quasistatic approximation (see Appendix D for more details), to study their interaction with light and with nearby scattering objects or molecules \[35,36\]. However, meta-atoms with complicated multipolar transitions can also be modelled into $\alpha$-tensors, which are then inserted into the MST \[31\], potentially allowing studies on more complicated physics, e.g., Fano resonances \[9,10\] and hybridization of particle and lattice resonances in 2D \[8,20,57\] and 3D \[58,59\] arrays. In this section, we reconstruct several physical phenomena arising in electromagnetically coupled meta-atoms simply by implementing $\alpha$-tensors into the MST, and discuss the advantages of this method.

First, electromagnetically coupled two plasmonic nanorods are illustrated using MST (Fig. 5a and b), where only $(\alpha^2_{11})_x$ component is considered in their $\alpha$-
tensors. Such closely situated plasmonic particles are strongly coupled, resulting in strong spectral resonance shift. Spectral red-shift (Fig. 5a) and blue-shift (Fig. 5b) can be reconstructed depending on the configuration of the coupled nanorods. This phenomena can be intuitively interpreted by plasmon hybridization theory (PHT) [60]; the induced charge density configuration in Fig. 5a becomes stable by the hybridization redshifting the resonance, whereas the configuration in Fig. 5b becomes unstable blue-shifting the resonance. However, PHT is based on the quasistatic approximation, so quantitative agreement with the reference (dashed-line) cannot be achieved. Noticeably, this approach provides predictions for rigorously studying electromagnetic problems involving disordered, aperiodic structures with superior computational efficiency compared to the traditional numerical methods, such as finite-difference time-domain and finite-element methods. Importantly, the multipole approach has superior computational efficiency compared to the traditional numerical methods, such as finite-difference time-domain and finite-element methods. Noticeably, this approach has shown significant potentials for rigorously studying electromagnetic phenomena arising in complex, disordered media [30, 32, 33, 62], and multi-scale systems with a large number of particles (N ≳ 10,000) over a large volume [34]. Especially, aperiodic metasurfaces [30, 32, 33] and random media [62] could be accurately studied using this method, and optimization [63] and dataset construction for deep-learning neural networks [64] would significantly benefit from this approach. In addition, the multipole approach can implement localized shaped beams from simple Gaussian beams [65] to highly focused [66] and helical beams [35, 67], providing a versatile framework to rigorously study spin-orbit interactions. Therefore, the multipole approach could be used to rigorously study electromagnetic phenomena arising in complex, disordered media consisting of discrete scattering objects.

VI. TOY MODELS

In previous examples, α-tensors of realistic particles were considered, but it is also possible to consider α-tensors of arbitrary particles without information on their physical geometric parameters. Recently, Fernandez-Corbaton, et al. proposed the concept of dual particles, which are excited by light with one helicity, re-radiates light with the same helicity, and are completely transparent to light with the opposite helicity.
They also proposed that two oppositely dual particles are completely uncoupled to each other, and a media consisting of dual particles with a single handedness will be completely transparent to one helicity and opaque to the other helicity [65]. Demonstration of this concept is difficult, because purely dual particles have not been discovered, although approximately dual particles have been studied [17] [65]. Still, we can theoretically investigate the concepts above using \( \alpha \)-tensors and MST, because arbitrary \( \alpha \)-tensors without physical parameters can be devised, which cannot be done using the traditional numerical methods. Dual particles at dipole approximation have \( \alpha_{1}^{e1} = \alpha_{m1}^{e1} = \pm \alpha_{e1}^{m1} = \pm \alpha_{m1}^{e1} \), where \( \pm \) corresponds to left- (LDP) and right-dual particles (RDP), which interact with left- (LCP) and right-circularly-polarized lights (RCP), respectively.

In this section, we implemented dual particles using isotropic (scalar), dipolar \( \alpha \)-tensors with Lorentzian resonance. We confirmed that two oppositely dual particles are completely uncoupled with each other, and only LDP is excited at LCP incidence (Fig. 6a), and RDP is excited at RCP incidence (Fig. 6b). This is because LDP is excited at LCP incidence (Fig. 6a), and RDP is excited at RCP incidence (Fig. 6b). This is because LDP is excited by LCP and re-radiates LCP, which cannot excite RDP. In a mixture of LDPs and RDPs, RDPs are completely transparent upon LCP incidence, and LDPs are electromagnetically coupled.

This section shows \( \alpha \)-tensors as a powerful method to treat symmetries and conservation laws at the microscopic level (single scattering object) without dealing with the geometrical parameters. In fact, implementation of \( \alpha \)-tensors without their relevant geometrical structures has been performed in literature to treat realistic molecules at weak excitations [65], where the molecules were approximated as \( \alpha \)-tensors and implemented in the framework of MST to study plasmon-enhanced circular dichroism [35] and helical dichroism [36].

VII. CONCLUSIONS

In summary, we have introduced the systematic transformation between \( \alpha \)-tensor in the Cartesian basis and \( T \)-matrix in the spherical basis using the basis transformation between the exact Cartesian and the spherical multipoles (Eqn. 2) and between the local fields and field gradients and the spherical multipoles (Eqn. 7). In general, characterization of meta-atoms using \( \alpha \)-tensor has been limited to dipolar regime, but recent advances in nanophotonics and metamaterials utilize higher-order multipole transitions coming from coupled plasmonic and high-index dielectric nanoantennas. These scattering systems with higher-order multipole transitions can be interpreted at a fundamental level using higher-order \( \alpha \)-tensor, which can be easily obtained from \( T \)-matrix using the facile basis transformation. Incorporated with the MST, the multipole approach can be a versatile theoretical framework in nanophotonics to rigorously investigate optical phenomena arising in coherently coupled multi-body systems [12]. Here, the well-defined symmetries and conservation laws can be treated at the microscopic level (single scattering object) using \( \alpha \)-tensors, and the electromagnetic interaction between them are treated using the Green’s tensors. It is worthwhile to note that the potential applicability of \( \alpha \)-tensor (or \( T \)-matrix) into the MST has been mentioned in many previous papers, but only spherical particles have been generally considered. By simply taking nonspherical structured meta-atoms into account, the multipole approach can be extended into many different applications.

The multipole approach is especially advantageous for complicated, random, multi-scale problems due to computational efficiency, and analytic scattering objects, such as realistic molecules and dual particles, can also be implemented. We hope this study may serve as a fundamental reference for the multipole approach in nanophotonics. Moreover, the uniqueness of the multipole approach allows this work applicable to other fields including acoustics, astronomy, and remote sensing.

Appendix A: Notes on approximate Cartesian multipoles

The exact Cartesian multipoles exactly reconstruct multipole radiations of the localized current sources with arbitrary sizes, but their expressions (Eqn. 1) are rather unfamiliar. The expressions of the familiar approximate Cartesian multipoles can be obtained by taking the long-wavelength limit [16] as:

\[
\begin{align*}
\alpha_{1}^{e1} &= \frac{1}{i\omega} \int d^3r \, J_\alpha \\
\alpha_{m1}^{e1} &= \frac{1}{2} \int d^3r \, (r \times J)_\alpha \\
Q_{\alpha\beta}^{E} &= \frac{1}{i\omega} \int d^3r \, \left[ r_\alpha J_\beta + r_\beta J_\alpha - \frac{2}{3} \delta_{\alpha\beta} (r \cdot J) \right] \\
Q_{\alpha\beta}^{M} &= \int d^3r \, \left[ r_\alpha (r \times J)_\beta + r_\beta (r \times J)_\alpha \right] \\
O_{\alpha\beta\gamma}^{E} &= \frac{2}{i\omega} \int d^3r \, \left[ r_\alpha r_\beta J_\gamma + r_\beta r_\gamma J_\alpha + r_\gamma r_\alpha J_\beta \\
&- \frac{1}{5} \delta_{\alpha\beta} (r^2 J_\gamma + 2 (r \cdot J) r_\gamma) - \frac{1}{5} \delta_{\beta\gamma} (r^2 J_\alpha + 2 (r \cdot J) r_\alpha) \\
&- \frac{1}{5} \delta_{\alpha\gamma} (r^2 J_\beta + 2 (r \cdot J) r_\beta) \right] \\
O_{\alpha\beta\gamma}^{M} &= \frac{1}{6} \int d^3r \, \left[ r_\alpha r_\beta (r \times J)_\gamma + r_\beta r_\gamma (r \times J)_\alpha + r_\gamma r_\alpha (r \times J)_\beta \\
&- \frac{1}{5} \delta_{\alpha\beta} r^2 (r \times J)_\gamma - \frac{1}{5} \delta_{\beta\gamma} r^2 (r \times J)_\alpha - \frac{1}{5} \delta_{\gamma\alpha} r^2 (r \times J)_\beta \right]
\end{align*}
\]

Interestingly, toroidal multipoles can also be recovered from the higher-order terms [16]. In addition, we numerically confirmed the transformation between the exact Cartesian multipoles and the spherical multipoles. The exact Cartesian multipoles (solid lines) and the approximate Cartesian multipoles (dashed lines) were calculated numerically using FEM and were compared with the exact Cartesian multipoles.
transferred from the spherical multipoles that were calculated analytically using the Mie theory. Two cases: a smaller sphere with lower refractive-index (Fig. 7a) and a larger sphere with higher refractive-index (Fig. 7b) were compared. The exact Cartesian multipoles that were numerically calculated (dashed lines) and those transformed from the analytically calculated spherical multipoles (solid lines) agree very well for both smaller and larger spheres. For the larger sphere, the approximate Cartesian multipoles deviate strongly from the exact Cartesian multipoles.

FIG. 7. Induced multipoles of (a) a small dielectric sphere with \( R = 50 \) nm and \( n = 2 \) and (b) a large sphere with \( R = 300 \) nm and \( n = 3 \). Dashed line: the numerically calculated exact Cartesian multipoles; dot-dashed line: the numerically calculated approximate Cartesian multipoles; solid line: the exact Cartesian multipoles transformed from the analytically calculated spherical multipoles; black: real; red: imaginary.

Appendix B: Notes on the proportionality constants

It should be noted that the proportionality constants for multipoles are rather arbitrary, but the reconstructed multipole radiations satisfying the Maxwell’s equations should be identical, for which the proportionality constants are compensated by the expressions for the multipole radiations (Eqn. 4).

Due to the same reason, the proportionality constants for the spherical multipoles are also arbitrary. In general, the spherical multipoles are not expressed explicitly, but the vector spherical wave functions (VSWFs) are given, from which the spherical multipoles are obtained using the orthogonality of the VSWFs. Note that expressions for VSWFs differ by publications, and our expression is

\[
M_{nm}^{(i)}(kr) = i\gamma_{nm} z_{nm}^{(i)}(kr)(i\pi_{nm}\hat{e}_\theta - \tau_{nm}\hat{e}_\phi)e^{im\phi} \quad \text{(B1a)}
\]

\[
N_{nm}^{(i)}(kr) = i\gamma_{nm} [n(n+1)\frac{z_{nm}^{(i)}(kr)}{k_r}P_n^m(\cos \theta)\hat{e}_r + \frac{1}{k_r}\frac{d[krz_{nm}^{(i)}(kr)]}{d(kr)}(\tau_{nm}\hat{e}_\theta + i\pi_{nm}\hat{e}_\phi)]e^{im\phi} \quad \text{(B1b)}
\]

\[
\tau_{nm}(\theta) = \frac{d}{d\theta} P_n^m(\cos \theta) \quad \text{(B1c)}
\]

\[
\pi_{nm}(\theta) = \frac{m}{\sin \theta} P_n^m(\cos \theta) \quad \text{(B1d)}
\]

\[
\gamma_{nm} = \sqrt{(2n+1)(n-m)!/4\pi n(n+1)(n+m)!} \quad \text{(B1e)}
\]

where superscripts (1) and (+) refer to the regular and singular spherical waves, respectively; \( z_{nm}^{(i)}(kr) = h_{nm}^{(i)}(kr) \) and \( z_{nm}^{(1)}(kr) = j_{nm}(kr) \). The spherical multipoles can also be obtained from the localized current sources \[2\]

\[15\]. The incident and scattered fields are reconstructed as

\[
E_{\text{sc}} = E_0 \sum_{n=1}^{\infty} \sum_{m=-n}^{n} [b_{nm}^E N_{nm}^{(i)}(kr) + b_{nm}^M M_{nm}^{(i)}(kr)]
\]

\[
E_{\text{inc}} = E_0 \sum_{n=1}^{\infty} \sum_{m=-n}^{n} [a_{nm}^E N_{nm}^{(1)}(kr) + a_{nm}^M M_{nm}^{(1)}(kr)]
\]

Appendix C: Units of polarizability tensors

The \( \alpha \)-tensor linearly relates the fields and field gradients at the origin to the induced multipoles as:

\[
\begin{bmatrix}
\mathbf{d}^E \\
\mathbf{Q}^E \\
\mathbf{d}^M \\
\mathbf{Q}^M
\end{bmatrix} =
\begin{bmatrix}
\mathbf{E} \\
\mathbf{H} \\
\mathbf{E} \\
\mathbf{H}
\end{bmatrix} =
\begin{bmatrix}
\alpha_{E}^E & \alpha_{E}^H & \alpha_{H}^E & \alpha_{H}^H \\
\alpha_{E}^{E} & \alpha_{E}^{H} & \alpha_{H}^{E} & \alpha_{H}^{H} \\
\alpha_{E}^{M} & \alpha_{E}^{H} & \alpha_{H}^{M} & \alpha_{H}^{M} \\
\alpha_{E}^{M} & \alpha_{E}^{H} & \alpha_{H}^{M} & \alpha_{H}^{M}
\end{bmatrix}
\begin{bmatrix}
\mathbf{E} \\
\mathbf{H} \\
\mathbf{E} \\
\mathbf{H}
\end{bmatrix}
\]

\( \text{(C1)} \)

This definition, however, is expressed in SI units. The \( \alpha \)-tensor in SI units has different units per components as can be seen in the dipolar \( \alpha \)-tensor:

\[
\begin{bmatrix}
\mathbf{d}^E \\
\mathbf{d}^M
\end{bmatrix} =
\begin{bmatrix}
\alpha_{E}^E & \alpha_{E}^{H} \\
\alpha_{E}^{M} & \alpha_{E}^{H}
\end{bmatrix}
\begin{bmatrix}
\mathbf{E} \\
\mathbf{H}
\end{bmatrix}
\]

\( \text{(C2)} \)

The components of \( \alpha \)-tensor in SI unit can differ by several orders, so they cannot be compared directly. Therefore, the normalized \( \alpha \)-tensor in units of volume is used in this work, which at dipole regime is expressed as:

\[
\begin{bmatrix}
\mathbf{d}^E/\epsilon \\
\gamma \mathbf{d}^M
\end{bmatrix} =
\begin{bmatrix}
\alpha_{E}^{E1} & \alpha_{E}^{H1} \\
-i\alpha_{M1} & \alpha_{M1}
\end{bmatrix}
\begin{bmatrix}
\mathbf{E} \\
\mathbf{H}
\end{bmatrix}
\]

\( \text{(C3)} \)
where $a^0_{+1} = \alpha^0_{d_1}/\epsilon$, $ia^m_{+1} = \alpha^m_{H_1}/(\epsilon \eta)$, $-ia^m_{+1} = \eta a^m_{d_1}$, and $\alpha^m_{-1} = \alpha^m_{d_1}$. The $\alpha$-tensor components in SI units can be easily obtained from Eqn. 6 and 7. For instance, ED-ED transition term can be expressed as $E_{d_1} = (\sqrt{\frac{k^2}{2m^2\omega^2}O_1}) T_{E1} E_{d_1} (\sqrt{\frac{1}{2k^2m^2\omega^2}O_1})^{-1} = \frac{6e}{2\pi \Omega_1} T_{E1} \Omega_1^{-1} = \epsilon^2 a^m_{+1}$. The other transition components in SI units can also be obtained in a similar manner.

**Appendix D: Quasistatic polarizabilities**

The multipole approach has been successful in providing simple analytic form for describing small nanoparticles. This allowed modelling small dielectric, plasmonic, or chiral nanospheres for many different phenomena including plasmon-enhanced scattering, optical trapping, and chiral optical forces, to name a few. In this section, we compare quasistatic polarizability expressions with the exact polarizability. The quasistatic expressions can be applied to very small nanoparticles and do not describe higher-order multipole contributions that arise in near-field interactions or larger particles or clusters.

Notably, subwavelength nanospheres have often been expressed by the quasistatic polarizability

$$\alpha = 4\pi R^3 \frac{\epsilon_f - 1}{\epsilon_f + 2},$$

(D1)

where $\epsilon_f$ is the relative permittivity. Note that the quasistatic expression normalized by the sphere volume is independent of the radius. For a small Ag sphere with $R = 10$ nm, the quasistatic limit (solid line) shows good agreement with the exact polarizability (dashed line), but for a larger Ag sphere with $R = 30$ nm, the exact polarizability shows red-shifted and broadened resonance due to the larger radiative damping, which the quasistatic model cannot incorporate (Fig. 8).

The quasistatic expression for polarizability of subwavelength nanorods was only recently reported as simple analytic form [69, 70]

$$\alpha_x = 4\pi a^2 c \frac{\epsilon_f - 1}{3 + 3L_z (\epsilon_f - 1)}$$

(D2a)

$$\alpha_z = 4\pi a^2 c \frac{\epsilon_f - 1}{3 + 3L_z (\epsilon_f - 1)}$$

(D2b)

$$L_z = \frac{1 - e^2}{e^2}\left[\frac{1}{2e} \ln \frac{1 + e}{1 - e} - 1\right]$$

(D2c)

$$L_x = \frac{(1 - L_z)}{2}$$

(D2d)

where the focal length $f = \sqrt{c^2 - a^2}$ and the eccentricity $e = f/c$. Note that the quasistatic expression for nanorod normalized by its volume also is independent of the size, but only depends on the eccentricity. For the short-axis mode, the quasistatic and exact polarizabilities agree well (Fig. 9a). For the long-axis mode, quasistatic polarizability (solid line) agrees well with the exact polarizability of the small nanorod (dashed line), but the exact polarizability of the larger nanorod shows red-shifted and broadened response due to the larger radiative damping (Fig. 9b).

**FIG. 9.** Polarizabilities (a) $\alpha_{xx}$ and (b) $\alpha_{zz}$ of an Ag nanorod. Solid line: quasistatic; dashed line: exact, small ($a = 1$ nm and $c = 5$ nm); dot-dashed line: exact, large ($a = 10$ nm and $c = 50$ nm); black: real; red: imaginary.

Beyond the ED approximation, the quasistatic polarizabilities of a chiral sphere is given as [71]

$$\alpha_x = 4\pi R^3 \frac{(\epsilon_f - 1)(\mu_f + 2) - \kappa^2}{(\epsilon_f + 2)(\mu_f + 2) - \kappa^2}$$

(D3a)

$$\alpha_m = 4\pi R^3 \frac{(\mu_f - 1)(\epsilon_f + 2) - \kappa^2}{(\mu_f + 2)(\epsilon_f + 2) - \kappa^2}$$

(D3b)

$$\alpha_c = 4\pi R^3 \frac{3\kappa}{(\epsilon_f + 2)(\mu_f + 2) - \kappa^2}$$

(D3c)

where $\mu_f$ and $\kappa$ are relative relative permeability and chirality parameter of the chiral sphere, respectively. The quasistatic and exact polarizabilities of a small nonmagnetic chiral sphere agree very well (Fig. 10).

**Appendix E: Appendix: T-matrix retrieval**

Up to arbitrary multipole orders, T-matrix of several particle systems can be analytically calculated including...
notably spheres from Mie coefficients, chiral spheres [72], homogeneous anisotropic spheres [73], and even nonlocal spheres and cores [74], and T-matrix of a system of multiple particles can also be defined [20]. In general, T-matrix of nonspherical particles should be calculated numerically using Extended Boundary Condition Method [75]. Discrete-Sources Null-Field Method [76], or FEM [17]. In this work, we used FEM to extract T-matrix of meta-atoms for its convenient implementation. However, it should be noted that FEM is very costly for calculating T-matrix compared to surface-integral methods [75, 76], because they require only particle surfaces to be discretized, whereas FEM requires larger simulation domains including the PML and the spacer between the particle surface and the PML.

The retrieved T-matrix is then used to obtain exact higher-order α-tensor. The retrieved T-matrix (or α-tensor) can be analytically treated to efficiently calculate orientation-averaged optical responses, or inserted in the multiple-scattering theory to calculate coherently coupled optical responses between discrete scattering objects.

Appendix F: T-matrix and polarizability-tensors of meta-atoms

1. Meta-atoms in the main text

Here, we present T-matrices of meta-atoms, whose α-tensors are presented in the main text. Compared to the α-tensors with only a few components are visible (Fig. 2b and 3b), T-matrices show more complicated structures, which are difficult to interpret (Fig. 11b) because of the spherical basis. On the other hand, meta-atoms with rotational symmetries about z-axis show similar level of complexity in their T-matrices and α-tensors (Fig. 11c). In later parts, we demonstrate analysis of meta-atoms from their retrieved α-tensors.

2. Plasmonic nanorod

First, we analyze a plasmonic nanorod with strongly anisotropic response, which is excited when the incident electric field is parallel to the nanorod axis. This long-axis mode is also known to be strongly redshifted compared to the short-axis mode. The retrieved α-tensor of a Ag nanorod clearly demonstrates this feature (Fig. 12a,b). ED response in nanorod-axis (x) direction is red-shifted and stronger as can be seen from (α_{e_1}^{e_1})_x^x than in short-axis direction shown in (α_{e_1}^{e_1})_z^z, which is blue-shifted and far weaker than (α_{e_1}^{e_1})_x^x. In addition, a plasmonic nanorod has only one dominant α-tensor component (note that the colorbar is in logarithmic scale). This strongly anisotropic response of a plasmonic nanorod al-
lows it to be safely approximated as a point polarizable anisotropic element with \((\alpha_{e1}^1)^x_z\) present as in Fig. 5 in the main text.

3. High-index dielectric spheres

Recently, high-refractive-index dielectric nanoparticles have been noted for their low loss and higher-order multipole modes coming from Mie-like resonances [47]. A small Si sphere with \(R = 70\) nm by planewave incidence shows strong MD radiation at 580 nm (Fig. 13a). The ED and MD resonances originates from \(\alpha_{e1}^1\) and \(\alpha_{m1}^1\), respectively. Due to the isotropic response coming from the spherical symmetry, only diagonal terms appear in \(\alpha\)-tensor (Fig. 13b).

4. Split-ring resonators

A split-ring resonator (SRR) is one of the most widely studied elements for achieving optically magnetic responses. Interestingly, the origin of this magnetic dipole mode can be explained from the retrieved \(\alpha\)-tensor (Fig. 14a). Incident \(x\)-polarized electric field on SRR generates current loop in \(xy\)-plane, which corresponds to magnetic dipole moment oriented in \(z\)-direction. This transition is visible in the \((\alpha_{m1}^1)^x_z\) component, which shows transition from \(E_z\) into \(d_m^m\). However, it should be noted that the magnetic response of SRR is rather weak in visible regime due to large Ohmic losses [50], as can be seen from the weak scattering cross-section intensities (Fig. 14a).

Another interesting property from SRR is extrinsic chirality, or helicity dependent chiral response from a geometrically achiral structure at obliquely incident field [77]. It is counter-intuitive that an achiral structure can undergo chiral interaction. Extrinsic chirality occurs because incident field has a defined wavevector, which is not included in the mirror plane of the system. At oblique incidence, two resonances at 480 nm and 680 nm are observed (Fig. 14b), which corresponds to dominantly \((\alpha_{e1}^1)^x_z\) and \((\alpha_{e1}^1)^y_y\), respectively. However, the two modes cannot explain extrinsic chirality at the oblique incidence (Fig. 14c). From the retrieved \(\alpha\)-tensor, this extrinsic chirality comes from magneto-electric coupling term \((\alpha_{m1}^1)^x_z\) [77, 78]. It should be noted that this magneto-electric coupling term disappears for asymmetric SRR [47] due to even-parity.

5. Helicoids

Finally, we present \(\alpha\)-tensor of an interesting system with 4-fold rotational symmetry without inversion symmetry. Such system was recently demonstrated in Au helicoids synthesized in solution-phase [79]. Due to the 4-fold rotational symmetry in 3D space, only diagonal terms appear in dipole order with degenerate \(xx\), \(yy\), and \(zz\) components. Notably, chiral response is preserved even with this high symmetry. Due to the small size of this particle, dipolar order is sufficient to describe both achiral and chiral responses (Fig. 15c) from the imaginary parts of \((\alpha_{e1}^1)^x_z\) and \((\alpha_{m1}^1)^x_z\), respectively (Fig. 15d). Finally, EQ–EQ transition terms also appear and may become nonnegligible at larger sizes [79].
FIG. 15. (a) The schematics of Au helicoid with side length 50 nm. (b) The retrieved $\alpha$-tensor at 580 nm. (c) Extinction and circular dichroism. (d) Spectra of $(\alpha_e^1 e_1)^{xx}$, $(\alpha_m^{1e})^{xx}$, $(\alpha_e^2 e_2)^{xx}$, and $(\alpha_e^2 e_2)^{xy}$.

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DISCLOSURES

The authors declare no conflicts of interest.

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