


Multipolar theory of bianisotropic response of meta-atoms

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The bianisotropy of meta-atoms is usually associated with their nonlocal response and mutual coupling between electric and magnetic dipole moments induced by an incident field. In this Letter, we generalize the theory of bianisotropy beyond the dipole response to cases of arbitrary high-order multipole resonances. We demonstrate that bianisotropy is fully connected to the geometrical structure of meta-atoms and caused exclusively by the absence of their inversion symmetry, while the strength of the bianisotropy response grows normally with the size of a meta-atom. Applying a group theory analysis, we reveal the explicit criteria when the dipole approximation is enough for the correct description of bianisotropy and when accounting for higher-order multipoles is required. We consider a triangular prism as an illustrative example of a bianisotropic particle and show how accounting for higher-order multipoles prevents the violation of Onsager-Casimir conditions for kinetic coefficients appearing in the dipole approximation. The developed theory is an essential step toward a deeper insight into the scattering properties of nanoantennas and meta-atoms.

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Metasurfaces, metamaterials, and nanoantennas are the main building blocks of nanophotonics which enable field enhancement and the precise control of light at the subwavelength scale [1]. Their optical properties are strongly governed by the response of *meta-atoms*—their elementary units. The accurate description of light interaction with individual meta-atoms, which can be also treated as *nanoresonators*, and their small clusters is of high interest to the nanophotonics community [2–4] as it opens prospects for the effective engineering of nanophotonic structures with predesigned optical properties [5]. A powerful tool for such an analysis is the *multipolar expansion* method [6–8]. This method describes the electromagnetic field outside [9] the scatterer of an arbitrary shape by a superposition of vector spherical harmonics (VSHs) forming a complete set of basis functions known as *multipoles* [10–12]. In the scattered field domain, meta-atoms can be considered as effective point scatterers with specific polarizabilities corresponding to their dominant multipole moments. Once meta-atoms become much smaller than a wavelength, the scattered field can be well described by only electric dipoles (EDs) and magnetic dipoles (MDs). This makes it extremely helpful to interpret and predict the optical properties of single meta-atoms and their ensembles [13]. In this way, many fascinating effects have already been demonstrated and explained, for example, the nonradiating anapole state [14–17], directional scattering [18,19], cloaking, and invisibility effects [20,21].

The electric and magnetic dipole moments (\mathbf{p} and \mathbf{m}) of meta-atoms are not always independent. They can be mutually coupled, i.e., both components of an incident electromagnetic field (\mathbf{E} and \mathbf{H}) simultaneously induce \mathbf{p} and \mathbf{m} . This effect

is usually called *bianisotropy* (BA) and can be formally described as follows [22–25],

$$\begin{aligned}\mathbf{p} &= \varepsilon_0 \hat{\alpha}^{EE} \mathbf{E} + \frac{1}{c} \hat{\alpha}^{EH} \mathbf{H}, \\ \mathbf{m} &= \hat{\alpha}^{HH} \mathbf{H} + \frac{1}{Z_0} \hat{\alpha}^{HE} \mathbf{E},\end{aligned}\quad (1)$$

where \mathbf{E} and \mathbf{H} are the electric and magnetic fields of the incident wave at the dipole coordinates, c is the speed of light, Z_0 is the wave impedance of free space, ε_0 is the vacuum permittivity, and $\hat{\alpha}^{EE}$, $\hat{\alpha}^{EH}$, $\hat{\alpha}^{HE}$, $\hat{\alpha}^{HH}$ are second-rank polarizability tensors.

In the recent decade, much attention has been paid to BA metamaterials and metasurfaces [26–31]. It has been shown that BA leads to the appearance of many fascinating effects such as polarization control, topological photonic states, broken reciprocity, unusual optical forces, etc. [32–40]. While the BA contribution is almost negligible for meta-atoms substantially smaller than a wavelength, however, it becomes more apparent in the structures with sizes comparable to a wavelength. Thus, higher-order resonances start to play a key role in particles with sizes comparable to a wavelength [41–44], allowing us to generalize many optical effects such as the Kerker effect [18,19], anapole effects [14,45], and increased the Q factors of resonant modes [46,47]. The selective excitation of higher-order resonances without coupling to the lower ones is possible using specific configurations of the incident optical beam [48–50]. Moreover, for some specific shapes of scatterers, the dipole model [Eqs. (1)] does not describe the electromagnetic response correctly and even results in a violation of Onsager-Casimir conditions for kinetic coefficients [51]. In this sense, the generalization of the BA effects for higher-order resonances is a natural step in the development of a multipole theory and all-dielectric metapotonics.

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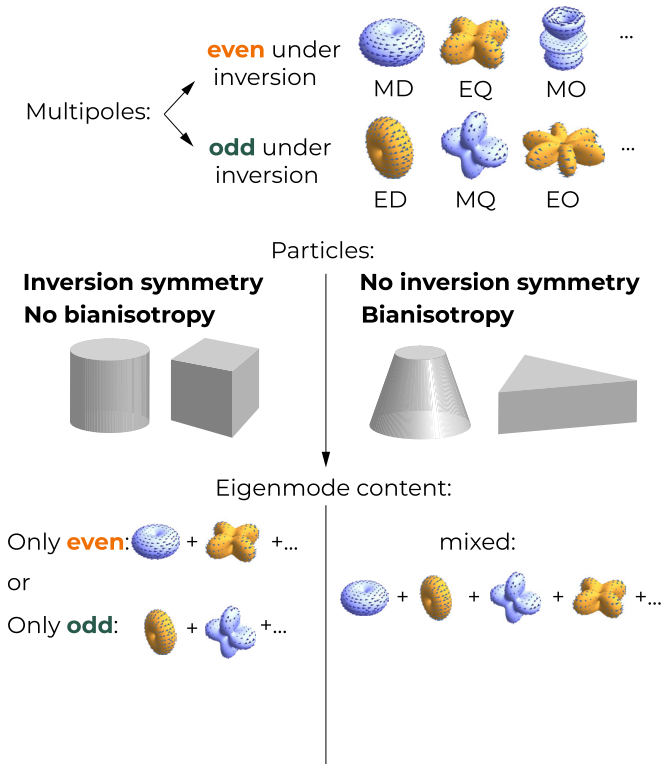


FIG. 1. General idea of multipolar bianisotropy. Vector spherical harmonics can be divided into even and odd under an inversion operation. The examples of even and odd multipoles are given at the top of the figure. If a particle is symmetric under inversion (left column), then it has no bianisotropic response and its eigenmodes consist of either only odd or even multipoles. If a particle does not possess inversion symmetry (right column), it has a bianisotropic response and its eigenmodes consist of multipoles of both parities.

In this Letter, we reconsider and generalize the concept of bianisotropy to meta-atoms of an arbitrary shape and size supporting high-order multipole resonances. We show that the origin of bianisotropy is fully defined by the symmetry of the particle rather than its size. The size of the particle, i.e., nonlocal response (spatial dispersion), is only responsible for the strength of BA but not for its presence. Applying the group theory formalism, we identify the selection rules enabling multipolar bianisotropy. With this knowledge, the predefined engineering of the bianisotropic response of arbitrarily shaped scatterers becomes possible and paves the way towards a great perspective in nanophotonics. The developed approach is consistent with all previous models of BA but brings a more solid and generalized view of the well-known electromagnetic BA effect.

Historically, BA was introduced for moving media [52,53]. In such systems, electric and magnetic displacements \mathbf{D} and \mathbf{H} are the functions of both electric and magnetic fields \mathbf{E} and \mathbf{B} . One can show that, in the general case, such a connection is possible only in media without inversion symmetry [23,54]. Therefore, being a structural element of the BA medium, a BA particle can be defined as one that has *no inversion symmetry*. Some of the well-known examples of BA meta-atoms are shown in Fig. 1. The absence of inversion symmetry immediately affects the multipolar content of the field scat-

tered by the particle. Indeed, the multipoles entering an eigenmode are transformed the same way as the eigenmode under all transformations of the particle's symmetry group. Thus, for particles with inversion symmetry, all the eigenmodes can be classified into even and odd ones with respect to the inversion operation. Consequently, each eigenmode consists of either all even or all odd multipoles. For BA particles, we may conclude that each eigenmode contains multipoles of *both parities*, which is illustrated in Fig. 1.

The specific multipolar content of eigenmodes can be found using the Wigner theorem, which states that all the modes in a resonator are transformed by irreducible representations of the resonator's symmetry group [43,55–58]. The mode consists of multipoles, which transform by the same irreducible representation as the mode itself. We follow the notations of Ref. [59] to describe the multipoles by the VSHs \mathbf{M}_β (magnetic harmonic) and \mathbf{N}_β (electric harmonic). The index $\beta = \{e/o, m, \ell\}$ encodes the parity of the harmonic (even e or odd o), the total angular momentum $\ell = 1, 2, 3, \dots$, and its projection $m = 0, 1, \dots, \ell$. We should note that indices e and o are connected with the parity with respect to the change of sign of the azimuthal angle $\varphi \rightarrow -\varphi$ but not to the inversion [see Supplemental Material (SM) [60] for details].

The table in Fig. 2 presents the classification of eigenmodes and their multipole content for a truncated cone ($C_{\infty v}$ symmetry group) and a cylinder ($D_{\infty h}$ symmetry group), which are known as two distinct examples of BA and non-BA meta-atoms, respectively. The multipolar content for other shapes can be found in Ref. [43]. For cylindrical particles, it can be seen that the modes are classified by an azimuthal number m and parity with respect to the inversion. For $m = 0$, all the modes are additionally segregated into electric and magnetic ones. We use additional orange/green coloring to distinguish between the even/odd multipoles. The modes with $m \geq 1$ are doubly degenerated due to the cylindrical symmetry, and their linear combinations can be associated with clockwise and counterclockwise rotation directions.

The transformation of a cylinder into a truncated cone mixes the odd and even modes of the same azimuthal number m . Therefore, parity is no longer the quantum number. Let us consider the illumination of a cylinder by a plane wave propagating along the z direction and polarized along the y axis (see the multipolar decomposition of plane waves of different polarizations in Fig. S1 of the SM [60]). In this case, the wave excites the \mathbf{N}_{o11} harmonic from the E_{1u} and \mathbf{M}_{e11} harmonic from E_{1g} . They have different parities and therefore they are excited independently, i.e., the magnetic field of the plane wave excites only the modes of E_{1g} and the electric field excites only the modes of E_{1u} . However, when the cylinder is transformed into a truncated cone, both \mathbf{N}_{o11} and \mathbf{M}_{e11} enter the same mode, and their cross-excitation according to Eqs. (1) becomes possible, leading to the BA response. The strength of BA can be characterized by the corresponding T -matrix element as discussed below and in the SM [60].

Understanding the multipole content of resonators allows an immediate extension of BA to the domain of *higher-order multipoles* that becomes essential for resonators of larger sizes when the role of nonlocality (spatial dispersion) increases. Indeed, as seen from Fig. 2, electric and magnetic quadrupole multipoles also enter the same representation E_1 of the cone



























Azimuthal number	 $C_{\infty v}$	 $D_{\infty h}$	Spherical Multipoles: even, odd	
			Electric	Magnetic
$m=0$	$A_1(1)$	$A_{1u}(1)$	N_{e01}  N_{e03} 	— ...
		$A_{1g}(1)$	N_{e02}  N_{e04} 	— ...
	$A_2(1)$	$A_{2u}(1)$	—	M_{e02}  M_{e04}  ...
		$A_{2g}(1)$	—	M_{e01}  M_{e03}  ...
$m=1$	$E_1(2)$	$E_{1g}(2)$	N_{e12}  N_{e14}  M_{o11}  M_{o13}  ...	
		$E_{1u}(2)$	N_{o12}  N_{o14}  M_{e11}  M_{e31}  ...	
	$E_2(2)$	$E_{2g}(2)$	N_{e11}  N_{e13}  M_{o12}  M_{o14}  ...	
		$E_{2u}(2)$	N_{o11}  N_{o13}  M_{e12}  M_{e14}  ...	
$m=2$			

FIG. 2. Multipolar decomposition of eigenmodes of an object with the symmetry groups $C_{\infty v}$ and $D_{\infty h}$. In the left-hand column, there are azimuthal numbers m . In the column below the symmetry group identifier, there are irreducible representations of the group and their dimensions in parentheses. In the left-hand column of the table, there are irreducible representations of the group and their dimensions in parentheses. In the right-hand column, there are low-order electric (yellow) and magnetic (blue) multipoles (VSHs) of the eigenmodes associated with the corresponding irreducible representations. The figures show the field amplitude in the far-field zone, while the arrows show the polarization of the field. The orange and green letters correspond to even and odd inversion parity.

particles, thus enabling dipole-quadrupole and quadrupole-quadrupole BA observed in various photonic and plasmonic systems [61–63]. Notably, the common understanding that BA is a coupling between the electric and the magnetic resonances is partially true. As seen from Fig. 2, eigenmodes A_1 and A_2 of a truncated cone contain opposite parity multipoles with $m = 0$ of only one type, either magnetic or electric. These modes can be excited independently, for example, by an azimuthally or radially polarized Bessel beam [50,64,65]. Such a type of BA remains the subject of further research.

In this perspective, a nontrivial example of an equilateral triangular prism is of special interest since the standard dipole model described by Eqs. (1) is not applicable, resulting in a violation of the Onsager-Casimir conditions for kinetic coefficients [51,60]. As a noncentrally symmetric particle, the prism should formally possess the BA response, however, electric and magnetic dipole components are never transformed by the same representation: The x - and y -oriented EDs (N_{e11} and N_{o11}) enter E' , while x - and y -oriented MDs (M_{e11} and M_{o11}) enter E'' . The z -oriented magnetic and electric dipoles also enter different representations, A_2' and A_2'' , respectively. Thus, formally, *dipole-dipole BA is forbidden, while a dipole-*






























 D_{3h} irrep	Spherical Multipoles: even, odd	
	Electric	Magnetic
$A_1'(1)$	N_{e02}  N_{e33} 	M_{o34}  ...
$A_1''(1)$	N_{o34} 	M_{e02}  M_{e33}  ...
$A_2'(1)$	N_{o33} 	M_{e01}  M_{e03}  ...
$A_2''(1)$	N_{e01}  N_{e03} 	M_{o33}  ...
$E'(2)$	N_{o11}  N_{o22} 	M_{e12}  M_{e23}  ...
	N_{e11}  N_{e22} 	M_{o12}  M_{o23}  ...
$E''(2)$	N_{e12}  N_{e23} 	M_{o11}  M_{o22}  ...
	N_{o12}  N_{o23} 	M_{e11}  M_{e22}  ...

FIG. 3. Multipolar decomposition of eigenmodes of an object with the symmetry group D_{3h} , corresponding to prism particles. In the left-hand column of the table there are irreducible representations of the group and their dimensions in parentheses. The figure shapes and the coloring of the letters are defined the same way as in Fig. 2.

quadrupole and higher-order BA is allowed [66]. Indeed, the electric quadrupole N_{o22} and electric dipole N_{o11} are both present at the same mode, corresponding to the E' representation (see Fig. 3). Hence, if the incident field contains N_{o22} , then the mode from E' is excited and the scattered field contains the electric dipole N_{o11} with opposite parity. This manifestation of higher-order BA is shown schematically in Fig. 4. To demonstrate this effect numerically, we consider an isosceles triangular prism in the air made of a material with dielectric permittivity $\epsilon_p = 12.67$ that is close to silicon in the near infrared spectral range [67]. The geometric parameters are listed in the caption. The prism is excited by the spherical wave, whose electric field contains only the harmonic N_{o22} . Figure 4(b) shows the spectra of the total and partial scattering powers corresponding to N_{o11} , N_{o22} , and M_{e12} calculated in COMSOL MULTIPHYSICS. The prism scatters the incident field proportional to the even harmonic N_{o22} into the channels corresponding to odd harmonics N_{o11} and M_{e12} manifesting higher-order BA.

The more general case is presented in the table in Fig. 5, which contains all possible couplings between multipoles up to second order in the symmetry groups C_{nv} and D_{nh} [43]. Note that all cells of the table strongly depend on the choice of the main rotation axis (here, it is the z axis). From the table, it is clear that in groups D_{nh} (for any natural number n), dipoles cannot belong to the same representation, i.e., coupling between them is prohibited. It is crucial to note that the coupling between multipoles with the empty cell at the

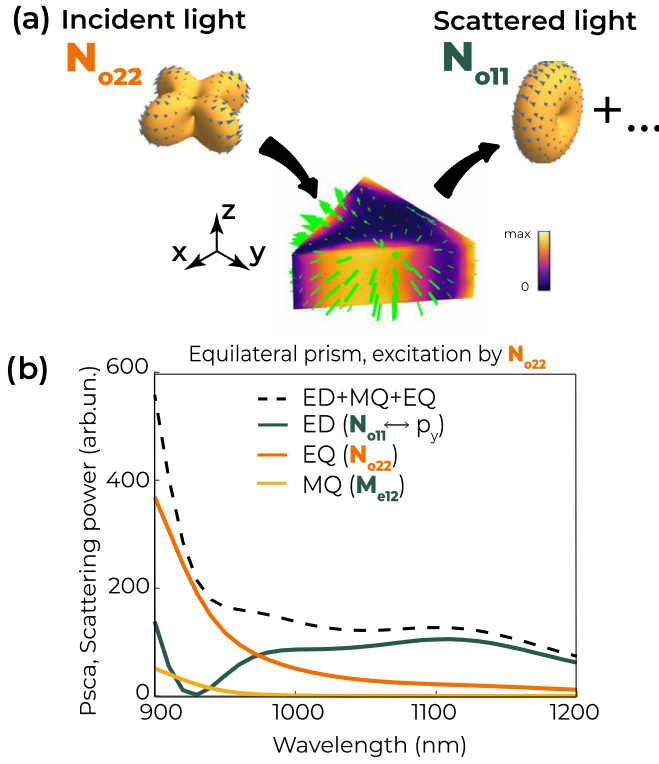


FIG. 4. (a) Schematic depiction of the dipole-quadrupole BA. Green arrows on the prism denote the direction and amplitude of the electric field excited by the field, whose multipole decomposition consists of an electric quadrupole N_{o22} . The color represents the distribution of the module of the electric field. (b) The spectrum of the total and partial scattering powers at wavelength $\lambda = 1050$ nm under the illumination of electric quadrupole N_{o22} . The height of the prism in the simulation was 188 nm, and the side was 520 nm.

intersection can be possible in some lower-symmetry groups. For instance, dipoles p_z and m_z belong to the same representation in the group C_2 . One can formulate the general selection rules for the symmetry groups, in which the bianisotropic coupling of the dipoles is possible, assuming the most symmetric orientation of the particle along the coordinate axes:

(1) m_i, p_i , where $i = x, y, z$, can couple to each other if there are no mirror reflections and inversion in the group since these dipoles transform in the same way under rotations and the opposite way under reflections;

(2) m_i, p_j can couple to each other, if there is no reflection in the ij plane and no rotations around the i or j axis are allowed (the scalar spherical harmonic corresponding to p_k belongs to the invariant representation), where (i, j, k) is any combination of (x, y, z) , and none of the indices (i, j, k) take the same value.

The standard BA model [Eq. (1)] can be directly extended beyond the dipole terms by including the spatial derivatives of the electric and magnetic fields, which is equivalent to accounting for the higher-order multipoles in the incident field [68,69],

$$p_i = \varepsilon_0 \alpha_{ij}^{EE} E_j + \frac{1}{c} \alpha_{ij}^{EH} H_j + \frac{\varepsilon_0}{2k_0} A_{ijk}^{EE} (\nabla_k E_j) + \dots, \\ m_i = \alpha_{ij}^{HH} H_j + \frac{1}{Z_0} \alpha_{ij}^{HE} E_j + \frac{1}{2Z_0 k_0} A_{ijk}^{HH} (\nabla_k H_j) + \dots \quad (2)$$

Legend:

- dipole-dipole «classical» bianisotropy (purple)
- higher-order bianisotropy (blue)
- non-bianisotropic coupling (white)

Coordinate system: z main rotation axis, x, y axes.

	ED		MD			EQ				MQ						
	$e11$ p_x	$o11$ p_y	$e01$ p_z	$e11$ m_x	$o11$ m_y	$e01$ m_z	$e02$	$e12$	$e22$	$o12$	$o22$	$e02$	$e12$	$e22$	$o12$	$o22$
ED	all	C_{IV}	C_{nv}	C_{nv}	C_{IV}	C_{IV}	C_{nv}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}
MD	C_{nv}	C_{IV}	all	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}
EQ	C_{IV}	C_{nv}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}
MQ	C_{IV}	C_{nv}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}	C_{IV}

FIG. 5. Selection rules of intramode multipolar coupling up to quadrupole terms for symmetry groups C_{nv}, D_{nh} . The main rotation axis is z . The names of the multipoles are given at the top and the side of the table. Cells at the intersection of the columns and rows contain the symmetry groups in which the multipoles belong to the same representation, i.e., they are coupled. The orange and green letters correspond to even and odd inversion parities, respectively. The violet cells indicate that the coupling is identified as bianisotropic in the classical meaning (dipole-dipole) [Eq. (1)]. The blue cells indicate higher-order bianisotropic coupling. The white cells indicate nonbianisotropic coupling. Empty cells mean that there is no such coupling in C_{nv}, D_{nh} .

Here, \hat{A}^{EE} and \hat{A}^{HH} are the nonlocal polarizability tensors of the third rank, and k_0 is the vacuum wave number. Applying model (2) to the case of scattering of a plane wave propagating along the y axis with x polarization, for which $\alpha_{yx}^{EE} = 0$ and $\alpha_{yz}^{EH} = 0$, one can write

$$p_y = a_{yxy} \left[\frac{\partial}{\partial y} (E_x e^{ik_y y}) \right]_{r=0} + \dots = a_{yxy} i k_y E_x + \dots, \quad (3)$$

where $a_{yxy} = \varepsilon_0 A_{yxy}^{EE} / 2k_0$. The field component on the right-hand side of Eq. (3) corresponds to an electrical quadrupole N_{o22} contained both in the incident and scattered fields. However, the model [Eqs. (2)] is not complete within the quadrupole approximation as it accounts for inducing the ED by the electric quadrupole (EQ), but it does not account for the reciprocal effect. A more natural formalism to account for the cross-coupling between the different multipoles is by the use of the T matrix. Its elements $T_{\beta_{out}, \beta_{in}}^{W_{out}, W_{in}}$ show the connections between the complex amplitudes of the incident and scattered fields. Here, the index $W_{in/out} = \{N, M\}$ encodes the type of harmonic (electric or magnetic) and index $\beta_{in/out} = \{o/e, m, n\}$. For the case considered in Fig. 4(a), one can find

the following relation [see Eq. (S44) in the SM [60], and Refs. [70,71] therein],

$$a_{\text{xyy}} = -\frac{5i\pi}{2\sqrt{3}k_0\omega} T_{\sigma 1, \sigma 2}^{NN}, \quad (4)$$

which is valid for small particles. The general relation between the T -matrix elements and multipolar polarizability coefficients can be found in Ref. [72].

Finally, we need to stress that the group analysis allows finding the selection rules for the cross-coupling between the multipoles—*intramode multipolar coupling*—but it does not predict the strength of the BA response. The strength of the BA is described by the off-diagonal elements of the corresponding T -matrix elements. They can be found, for instance, using the resonant state expansion method [60,73]

$$T_{\beta_{\text{out}}, \beta_{\text{in}}}^{W_{\text{out}}, W_{\text{in}}} = \sum_{j_\sigma} \frac{\omega \tilde{\alpha}_{W_{\text{out}}, \beta_{\text{out}}}^{j_\sigma}}{2(\omega_{j_\sigma} - \omega)} \int d\mathbf{r}' \mathbf{E}_{j_\sigma}^\sigma \mathbf{W}_{\beta_{\text{in}}} \Delta \varepsilon(\mathbf{r}'). \quad (5)$$

Here, σ encodes a particular irreducible representation and j_σ denotes the number of the eigenmode within this representation, $\tilde{\alpha}_{W_{\text{out}}, \beta_{\text{out}}}^{j_\sigma}$ is the multipolar expansion amplitude of eigenmode $\mathbf{E}_{j_\sigma}^\sigma$, $\Delta \varepsilon(\mathbf{r}') = \varepsilon(\mathbf{r}') - 1$, where $\varepsilon(\mathbf{r})$ is the permittivity function of the resonator, and ω_{j_σ} is the complex eigenfrequency. Here, we assume that the surrounding medium is the air. One can see that all the modes from the

same irreducible representation are excited simultaneously but with a different efficiency defined, in particular, by the proximity of ω to ω_{j_σ} .

To conclude, we have generalized the concept of bianisotropy beyond the dipole approximation, making it applicable to meta-atoms of arbitrary shapes and sizes. We show that the origin of bianisotropy is the absence of inversion symmetry that results in the coupling between multipoles of different parity within a single mode. The selection rules for multipolar coupling are revealed from a group symmetry analysis. Thus, we found the explicit criteria when the dipole approximation is enough for the correct description of bianisotropy and when it requires accounting for higher-order multipoles. The exact analytical expressions defining the strength of the bianisotropic response are derived based on the resonant state expansion and T -matrix formalism. The developed theory describes well the optical response even for complicated shapes of particles when classical dipole-dipole bianisotropy is violated. It also predicts a different bianisotropy when either magnetic or electric multipoles are coupled. We believe that the results obtained are an essential step towards a deeper insight into the scattering properties of nanoantennas and meta-atoms and their smart engineering.

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