
**Purely bianisotropic scatterers**

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The polarization response of molecules or meta-atoms to external electric and magnetic fields, which defines the electromagnetic properties of materials, can either be direct (electric field induces electric moment and magnetic field induces magnetic moment) or indirect (magneto-electric coupling in bianisotropic scatterers). Earlier studies suggest that there is a fundamental bound on the indirect response of all passive scatterers: It is believed to be always weaker than the direct one. In this paper, we prove that there exist scatterers which overcome this bound substantially. Moreover, we show that the amplitudes of electric and magnetic polarizabilities can be negligibly small as compared to the magneto-electric coupling coefficients. However, we prove that if at least one of the direct-excitation coefficients vanishes, magneto-electric coupling effects in passive scatterers cannot exist. Our findings open a way to a new class of electromagnetic scatterers and composite materials.

The causality requirement leads to Kramers-Kronig relations for the polarizabilities and to sum rules [8].

Here we discuss fundamental limitations on the relative strength of direct and indirect coupling phenomena, measured by the absolute values of the polarizabilities and coupling coefficients. For simplicity of writing and in view of particular examples which we will study, we will mainly use the scalar versions of the linear relations (1), considering the polarization responses only along one direction. In particular, we consider relative magnitudes of the products \( \alpha_{ee}\alpha_{me} \) and \( \alpha_{mm}\alpha_{em} \). Note that the passivity limitation on the imaginary part of the six-dyadic \( \overline{\overline{\alpha}} = \overline{\overline{\alpha}^T} \) of lossy particles (it is positive definite) [8] for the case of scalar parameters leads to the condition \([7,9] Im\{\alpha_{em}\alpha_{me}\} < Im\{\alpha_{ee}\alpha_{mm}\}\), but the real parts or the absolute values of these products are not restricted by passivity.

In naturally occurring molecules and particles, the magneto-electric coupling effects are very small as compared with the direct polarization effects, suggesting that \(|\alpha_{em}\alpha_{me}| \ll |\alpha_{ee}\alpha_{mm}|\). However, by properly engineering meta-atoms it is possible to significantly enhance the coupling effects. This enables new unprecedented effects. Here, we focus on the question if the coupling effects can be even stronger than the direct polarization, that is, if it is possible to create a scatterer such that \(|\alpha_{em}\alpha_{me}| > |\alpha_{ee}\alpha_{mm}|\).

Based on known results, it is expected that the indirect coupling effects cannot be stronger than the direct ones [3,4,7,10,11]. In earlier studies, it was shown that the polarizabilities of metal spirals close to the fundamental resonance obey the relation [10,11]

\[
|\alpha_{em}\alpha_{me}| = |\alpha_{ee}\alpha_{mm}| \tag{3}
\]

(see also in Ref. [4]). Later, it was shown that this equality is not a general restriction and examples of resonant spirals for which \(|\alpha_{em}\alpha_{me}| < |\alpha_{ee}\alpha_{mm}|\) holds were shown [12]. However, to the
best of our knowledge, no results where the opposite inequality would hold are known. Moreover, in more recent studies [3], it is stated that in a small scatterer the magnetolectric coupling cannot exceed the direct polarization effects, so that for all linear passive scatterers

$$|\alpha_{em}^\dagger \alpha_{me}| \leq |\alpha_{ee}^\dagger \alpha_{mm}|. \quad (4)$$

However, this statement of Ref. [3] implies the presumption that the scatterer is modeled as a single-resonant RLC circuit. In such scatterers, both induced electric and magnetic dipole moments are formed by the same current distribution and have nearly the same frequency dispersion close to the fundamental resonance.

Here, we show that once we relax this presumption, we can largely overcome this fundamental bound. On the other hand, we establish a new constraint for a general linear passive dipole scatterer. We prove that indirect magnetolectric coupling coefficients (bianisotropy parameters) may be different from zero only if both of the direct coupling coefficients $\tilde{\alpha}_{ee}$ and $\tilde{\alpha}_{mm}$ are simultaneously nonzero.

II. NECESSARY CONDITIONS FOR THE PRESENCE OF INDIRECT COUPLINGS

Let us first apply the energy conservation law to a general lossless bianisotropic scatterer (linear response is assumed). Equating the power extracted by the scatterer from the incident fields and the power which it re-radiates back into surrounding space, we can write the following relation between incident fields and the power which it re-radiates back into surrounding space.

$$i \frac{k}{2} (\tilde{\alpha}^\dagger - \tilde{\alpha}) = \frac{k^3}{6\pi\varepsilon_0} \tilde{\alpha}^\dagger \tilde{\alpha}, \quad (5)$$

where $k$ is the wave number of the incident wave and $\varepsilon_0$ is the permittivity of the host medium that is assumed to be isotropic and dielectric only, $\dagger$ denotes the conjugate transpose operator, and the $6 \times 6$ polarizability dyadic is defined in (A2). The time-harmonic dependency in the form of $e^{-i\omega t}$ is assumed. From (A1) and (A2), the following expressions for the polarizability tensor of a lossless scatterer follow (see Appendix A):

$$\frac{\tilde{\alpha}_{ee}^\dagger - \tilde{\alpha}_{ee}}{2} = -i \frac{k^3}{6\pi\varepsilon_0} [\tilde{\alpha}_{ee}^\dagger \tilde{\alpha}_{ee} + \tilde{\alpha}_{me}^\dagger \tilde{\alpha}_{me}], \quad (6)$$

$$\frac{\tilde{\alpha}_{mm}^\dagger - \tilde{\alpha}_{mm}}{2} = -i \frac{k^3}{6\pi\varepsilon_0} [\tilde{\alpha}_{mm}^\dagger \tilde{\alpha}_{mm} + \tilde{\alpha}_{em}^\dagger \tilde{\alpha}_{em}]. \quad (7)$$

If we now assume that $\tilde{\alpha}_{ee}$ is exactly zero, then we see from (A3) that $\tilde{\alpha}_{me}^\dagger \tilde{\alpha}_{me} = 0$. From here it follows that $\tilde{\alpha}_{me} = \tilde{\alpha}_{em} = 0$ (see Appendix A). The same result can be obtained from (A5) if a vanishing magnetic tensor $\tilde{\alpha}_{mm}$ is assumed. Similar reasoning leads to the conclusion that the same result is valid also for lossy (dissipative) scatterers. It can be understood from observing that for lossy scatterers relations (A3) and (A5) contain additional positive terms in the right-hand side, which represent the dissipated power.

Therefore, we conclude that if at least one of the direct coupling tensors vanishes, the indirect coupling tensors must be zero accordingly. However, from the above considerations it does not follow that the indirect magnetolectric coupling coefficients cannot be stronger as compared to the direct ones, as long as both direct polarizabilities are not exactly zero. It is important to study if there are possibilities to design scatterers which overcome the inequality (4).

III. COUNTEREXAMPLES WHICH BREAK THE INEQUALITY $|\alpha_{em}\alpha_{me}| \leq |\alpha_{ee}\alpha_{mm}|$

A. Split-ring resonators

As a representative example, let us consider a scatterer that exhibits bianisotropic coupling of the so-called omega type [8]: a split-ring resonator (Fig. 1).

The resonator has a square shape with the outer edge equal to 110 nm and the square cross section with the side of 30 nm. The gap is 30 nm. The material is gold [14]. Based on the semianalytical approach developed in Ref. [15], we extract the main polarizability components of this scatterer excited by the illumination shown in Fig. 1. Figure 2(a) depicts magnitudes of the main components of the polarizability tensors and Fig. 2(b) shows the products of the direct and the indirect terms, respectively.

Close to the resonance, where the current around the ring is nearly uniform (the conduction current is continued in the gap as a displacement current with the same phase), its polarizabilities agree with the theory of Ref. [11] and obey the relation (3). Based on the earlier studies [3], it can be expected that when the frequency deviates from the resonant range, limitation (4) should continue to hold. However, at frequencies near 325 THz, the indirect coupling coefficient $\alpha_{em}$ clearly exceeds both direct ones (normalized by the free space impedance $\eta_0$) more than 2.5 times. In this frequency range, the current induced in the resonator is strongly nonuniform: The current in the middle part of the scatterer is directed oppositely to the displacement current in the gap and the external

![FIG. 1. (a),(b) A metal split-ring resonator in free space. At the main resonance, the induced electric and magnetic dipole moments are related to the same nearly uniform current distribution.](image)

![FIG. 2. Numerically obtained magnitudes of the main polarizability components of the split-ring resonator versus frequency.](image)
electric field excites predominantly a magnetic dipole moment. Likewise, the external magnetic field excites a strong electric dipole moment. We conclude that beyond the frequency range of the fundamental resonance, where the RLC model is not applicable, the limitation (4) does not hold.

### B. Dimers

Although the studied example scatterer overcomes the previously established limitation (4), the indirect coupling coefficients $\alpha_{em}$ and $\alpha_{me}$ are still of the same order as that of the direct ones $\eta_0/\alpha_{ee}$ and $\alpha_{mm}/\eta_0$. In order to further enhance the indirect coupling and suppress the direct polarization effects, we push the idea of multimode scatterers to the limit by designing a nanodimer whose two constituents have electric polarizabilities of the opposite signs. Despite the fact that dimer scatterers have been intensively studied (see, e.g., Refs. [5,16,17]), their potentials for enhancing bianisotropic coupling as compared to the direct ones appear not to be realized. Commonly, previous works (see, e.g., Ref. [5]) were devoted to designing dimers that have no excited electric dipole moment under illumination of an incident plane wave (e.g., to realize scattering cancellation cloaking [18]):

$$\mathbf{p} = \hat{\alpha}_{ee} \cdot \mathbf{E}_{inc} + \hat{\alpha}_{em} \cdot \mathbf{H}_{inc} \approx 0. \quad (8)$$

However, this condition is drastically different from our goal to minimize the direct coupling. Indeed, Eq. (8) implies that the indirect coupling should be of the same order as the direct one.

In this work, we aim to design a scatterer such that its electromagnetic response is almost solely defined by the indirect coupling (a “purely bianisotropic scatterer”). Neglecting the direct coupling coefficients, we can write the dipole moments induced in such a scatterer as follows:

$$\mathbf{p} \approx \hat{\alpha}_{em} \cdot \mathbf{H}_{inc}, \quad \mathbf{m} \approx \hat{\alpha}_{me} \cdot \mathbf{E}_{inc}. \quad (9)$$

To exactly meet the equality in (9) is impossible due to the constraint established above.

We design the nanodimer as a system of two closely spaced, very small (compared to the wavelength) dielectric spheres with the relative permittivity $\epsilon_1$ and $\epsilon_2$ and equal radii $r = 21.4$ nm (see Fig. 3).

The distance between the centers of the spheres is $3r = 64.2$ nm. Without loss of generality, we assume that the nanodimer is located in free space. We choose $\epsilon_1 = 0.4$ and $\epsilon_2 = 2$ at the operational frequency, so that the electric polarizabilities of the spheres in the quasistatic approximation [1] have equal amplitudes but opposite signs (see Appendix B). The magnetic polarizabilities of both spheres in the quasistatic approximation can be neglected.

In order to analyze the dimer polarizabilities, we excite it by a combination of two plane waves propagating along the $z$ axis in the opposite directions, forming a standing wave. When the dimer is positioned in the antinode of the external electric field $\mathbf{E}_{ext} = 2\mathbf{E}_{inc}$ [see Fig. 4(a)], where the external magnetic field is zero, the induced electric dipole moments in the two spheres $\mathbf{p}_1$ and $\mathbf{p}_2$ compensate each other, ensuring near-zero direct coupling coefficient $\alpha_{ee}$ of the total dimer system.

On the other hand, this configuration of the opposite electric dipoles forms an electric quadrupole moment and a magnetic dipole moment [which, according to (9), corresponds to the nonzero indirect coupling coefficient $\alpha_{me}$]. Figure 4(b) depicts the radiation pattern of the nanodimer under this excitation in terms of the scattering amplitude $f = \lim_{r \to \infty} E_{sc}(r) \cdot r$. The result corresponds to the typical pattern of combined electric quadrupole and magnetic dipole moments.

Next, we position the nanodimer in the antinode of the magnetic field of the standing wave $\mathbf{H}_{ext} = 2\mathbf{H}_{inc}$ as shown in Fig. 4(c). Importantly, in this configuration the induced

![FIG. 3. Illustration of a nanodimer.](image-url)
magnetic moment in the dimer corresponding to the direct coupling is negligibly small. There are two reasons for that. Firstly, the spheres have a small intrinsic magnetic response because we work far from their magnetic Mie resonances. Secondly, the external magnetic field due to the Faraday law creates circulation of the electric field around the center of the dimer. Due to the opposite electric polarizabilities of the nanodimer spheres, this circulating external electric field excites the nanodimer. Under the excitation shown in Fig. 6, an electric dipole moment \( \mathbf{p}_3 \) will be induced in the plasmonic particle along the \( y \) axis, while the induced electric dipole moments \( \mathbf{p}_1 \) and \( \mathbf{p}_2 \) in the dimer will form an effective magnetic moment along the \( y \) axis. Tuning the dimensions of the plasmonic nanoparticle and its loss factor, one can balance the induced effective magnetic moment with the electric moment \( \mathbf{p}_3 \) so that they have the same amplitude and phase. In this case, these two orthogonal dipole moments form a Huygens’ pair whose scattering pattern has a null along the \( +z \) or \( -z \) direction. Therefore, due to the conservation of linear momentum in the system, the nanocluster, illuminated by a wave along the \( y \) direction will experience a lateral force along the \( z \) direction. Such an effect of side optical forces can be interesting in micromanipulation (so-called optical tweezers) and fabrication.

V. CONCLUSION

In summary, we have proved that nonzero indirect coupling coefficients in a passive scatterer exist only if both direct coupling effects are present at least as very weak effects. Based on several examples, we have shown that the earlier published considerations do not impose any limit on the strength of the indirect coupling coefficients compared to the direct ones. Moreover, we have demonstrated that the indirect coupling can be largely enhanced in specifically designed dimer scatterers. Such dimers possess unique, almost purely bianisotropic response and exhibit unprecedented effects, for example, lateral optical forces.
This result confirms the nearly pure bianisotropic response of the nanodimer. The frequency shift from 417 THz to 350 THz can be explained by the mutual interaction of the spheres. Compared to the case of small spheres in the main paper, this nanodimer exhibits about three orders of magnitude stronger electromagnetic response.

APPENDIX A: PROOF THAT PASSIVE PURELY BIANISOTROPIC SCATTERERS DO NOT EXIST

In the main text, we state that if we assume that the electric and magnetic polarizabilities \( \tilde{\alpha}_{ee} \) and \( \tilde{\alpha}_{mm} \) of a dipolar scatterer are exactly zero, then the magnetoelectric polarizabilities \( \tilde{\alpha}_{em} \) and \( \tilde{\alpha}_{me} \) must also be zero. To prove that, we first employ the relations for the extracted and scattered powers by the scatterer in terms of the dipolar moments. Then, we consider the fact that these two must be equal when there is no absorption loss. From these considerations, we can write the relation between the polarizability dyadics of a lossless particle:

\[
\frac{i}{2} (\hat{\alpha}^\dagger - \hat{\alpha}) = \frac{k^3}{6\pi\varepsilon_0} [\hat{\alpha}^\dagger \cdot \hat{\alpha}_{ee} + \hat{\alpha}^\dagger \cdot \hat{\alpha}_{me} + \hat{\alpha}^\dagger \cdot \hat{\alpha}_{mm}], \tag{A1}
\]

where \( \hat{\alpha} \) is the conjugate transpose operator and

\[
\tilde{\alpha} = \left[ \begin{array}{c} \tilde{\alpha}_{ee} \\ \tilde{\alpha}_{em} \\ \tilde{\alpha}_{mm} \end{array} \right]. \tag{A2}
\]

From (A1) and (A2) we derive the following expressions for the polarizability dyadics of a lossless particle:

\[
\frac{\tilde{\alpha}_{ee}^\dagger - \tilde{\alpha}_{ee}}{2} = -i \frac{k^3}{6\pi\varepsilon_0} [\hat{\alpha}^\dagger \cdot \hat{\alpha}_{ee}], \tag{A3}
\]

\[
\frac{\tilde{\alpha}_{em}^\dagger - \tilde{\alpha}_{em}}{2} = -i \frac{k^3}{6\pi\varepsilon_0} [\hat{\alpha}^\dagger \cdot \hat{\alpha}_{em}], \tag{A4}
\]

\[
\frac{\tilde{\alpha}_{mm}^\dagger - \tilde{\alpha}_{mm}}{2} = -i \frac{k^3}{6\pi\varepsilon_0} [\hat{\alpha}^\dagger \cdot \hat{\alpha}_{mm}], \tag{A5}
\]

\[
\frac{\tilde{\alpha}_{em}^\dagger - \tilde{\alpha}_{em}}{2} = -i \frac{k^3}{6\pi\varepsilon_0} [\hat{\alpha}^\dagger \cdot \hat{\alpha}_{mm}], \tag{A6}
\]

Next, let us assume that both electric and magnetic polarizabilities of the scatterer are zero, i.e., \( \tilde{\alpha}_{ee} = \tilde{\alpha}_{mm} = 0 \). Then, from (A3) and (A5) it follows that

\[
\tilde{\alpha}_{me}^\dagger \cdot \tilde{\alpha}_{me} = 0, \tag{A7}
\]

\[
\tilde{\alpha}_{em}^\dagger \cdot \tilde{\alpha}_{em} = 0. \tag{A8}
\]

If we consider \( \tilde{\alpha}_{em} \) and \( \tilde{\alpha}_{me} \) as three-dimensional matrices:

\[
\tilde{\alpha}_{em} = \begin{bmatrix} \alpha_{xx}^{em} & \alpha_{xy}^{em} & \alpha_{xz}^{em} \\ \alpha_{yx}^{em} & \alpha_{yy}^{em} & \alpha_{yz}^{em} \\ \alpha_{zx}^{em} & \alpha_{zy}^{em} & \alpha_{zz}^{em} \end{bmatrix}, \tag{A9}
\]

and

\[
\tilde{\alpha}_{me} = \begin{bmatrix} \alpha_{xx}^{me} & \alpha_{xy}^{me} & \alpha_{xz}^{me} \\ \alpha_{yx}^{me} & \alpha_{yy}^{me} & \alpha_{yz}^{me} \\ \alpha_{zx}^{me} & \alpha_{zy}^{me} & \alpha_{zz}^{me} \end{bmatrix}, \tag{A10}
\]

then from (A7) and (A8) we obtain the following two sets of equations:

\[
\alpha_{xx}^{em}^2 + \alpha_{xy}^{em^2} + \alpha_{xz}^{em^2} = 0, \tag{A11}
\]

\[
\alpha_{xy}^{em^2} + \alpha_{yy}^{em^2} + \alpha_{zx}^{em^2} = 0, \tag{A12}
\]

\[
\alpha_{xz}^{em^2} + \alpha_{yz}^{em^2} + \alpha_{zz}^{em^2} = 0 \tag{A13}
\]

FIG. 7. Analytical polarizabilities of spheres with \( \varepsilon_1 = 0.6 \) and \( \varepsilon_2 = 10 \) at frequencies near the Mie resonances. At 417 THz, the real parts of the electric polarizabilities have equal amplitudes but opposite signs, while their imaginary parts are negligible.

FIG. 8. Distribution of the electric field scattered by the nanodimer at 350 THz. The dimer is excited by external (a),(b) electric field and (c),(d) magnetic field. (b),(d) The corresponding patterns of the scattering amplitude.
and
\[ |\alpha_{ae}^{3x}|^2 + |\alpha_{ae}^{3y}|^2 + |\alpha_{ae}^{3z}|^2 = 0, \]
\[ |\alpha_{mn}^{3x}|^2 + |\alpha_{mn}^{3y}|^2 + |\alpha_{mn}^{3z}|^2 = 0, \]
\[ |\alpha_{me}^{3x}|^2 + |\alpha_{me}^{3y}|^2 + |\alpha_{me}^{3z}|^2 = 0. \]

Now from (A11) and (A12) one can conclude that if \( \alpha_{ae} = \alpha_{mn} = 0 \), then there is no possibility to have nonzero \( \alpha_{em} \) and \( \alpha_{me} \), i.e.,
\[ \text{if} \ {\alpha_{ae} = \alpha_{mn} = 0} \implies \{\alpha_{em} = \alpha_{me} = 0\}. \] (A13)

This conclusion is applied to any dipolar scatterer.

**APPENDIX B: DIELECTRIC SPHERES CLOSE TO THE MIE RESONANCES**

The electric polarizabilities of the spheres within the quasistatic approximation read
\[ \alpha_{el1.2} = \frac{4}{3} \pi r^3 \varepsilon_0 \frac{3(\varepsilon_{1.2} - 1)}{\varepsilon_{1.2} + 2} \] (B1)
and their magnetic polarizabilities can be neglected. In the main text, we presented a scatterer system of two dielectric spheres whose magnetoelectric polarizability is stronger than both electric and magnetic polarizabilities. The individual electric \( \alpha_{ee} \) and magnetic \( \alpha_{mm} \) dipole polarizabilities of a dielectric sphere can be determined by [19]:
\[ \alpha_{ee} = \frac{6\pi i \varepsilon}{k^3} a_1, \quad \alpha_{mm} = \frac{6\pi i \mu}{k^3} b_1, \] (B2)
where \( k = \omega \sqrt{\varepsilon \mu} \) is the host medium wave number, \( \mu \) and \( \varepsilon \) are the permeability and permittivity of the host medium, \( a_1 \) and \( b_1 \) are the dipolar terms of the scattering Mie coefficients for a sphere of an arbitrary size in a uniform host medium. The scattering Mie coefficients of all the multipoles excited in the sphere are denoted by [20]:
\[ a_n = \frac{\mu \mu_0^2 j_n(mx)(xj_o(x))' - \mu_0 j_n(x)(mxj_o(mx))'}{\mu \mu_0^2 j_n(mx)(xh_n^{(1)}(x))' - \mu_0 h_n^{(1)}(x)(mxj_o(mx))'}. \] (B3)

FIG. 9. Numerically obtained magnitudes of the main polarizability components of the purely bianisotropic nanodimer operating close to the Mie resonances of its inclusions.

and
\[ b_n = \frac{\mu \mu_0 j_n(mx)(xj_o(x))' - \mu_0 j_n(x)(mxj_o(mx))'}{\mu \mu_0 j_n(mx)(xh_n^{(1)}(x))' - \mu_0 h_n^{(1)}(x)(mxj_o(mx))'}. \] (B4)

Here, \( x = \omega \sqrt{\varepsilon \mu} D/2 \), \( m = \sqrt{\varepsilon \mu} \), \( \mu_p \) and \( \varepsilon_p \) are the permeability and permittivity of the sphere particle, respectively. In these relations \( D \) is the sphere diameter, \( j_n \) and \( h_n^{(1)} \) are the spherical Bessel functions of the first and third kind, respectively, and sign “′” denotes the derivative with respect to the argument.

We choose the radii of the spheres \( r = 65 \) nm and the distance between their centers \( 3r = 195 \) nm, and calculate the polarizabilities using the full-wave Mie theory. To prevent strong magnetic response from the spheres, we choose the operational frequency far enough from the nearest magnetic resonance of the spheres occurring at 696 THz. Next, we choose the permittivities of the spheres \( \varepsilon_1 = -0.6 \) and \( \varepsilon_2 = 10 \) such that at the frequency 417 THz their electric polarizabilities have equal amplitudes but opposite signs \( \text{Re}(\alpha_{ee1}) = -\text{Re}(\alpha_{ee2}) \). Figure 7 depicts the analytically calculated polarizabilities of such spheres.

Due to this property, the external electric/magnetic field induces magnetic/electric dipole moment in the nanodimer as shown in Figs. 8(a) and 8(c). The scattering patterns for these two cases are presented in Figs. 8(b) and 8(d). Close to 350 THz, the direct coupling coefficients are suppressed, while the indirect coupling coefficient is strong [see Figs. 9(a) and 9(b)].


