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Proposed isotropic negative index in three-dimensional optical metamaterials

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A simple route toward achieving an isotropic optical negative index in three dimensions is theoretically proposed. We show that, in contrast with previous studies, the plasmonic ring resonators, symmetrically split with an odd number of gaps, have both degenerate electric and magnetic resonances and thus provide an isotropic negative index if randomly distributed in a host medium. For an even number of gaps, the electric and magnetic dipoles sufficiently overlap only by additional symmetry breaking, which is demonstrated to allow unique control of the relative contribution from higher-order multipolar moments.

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Optical metamaterials have seen tremendous progress in the past few years.[1–4] Applications such as perfect lenses,[5] rainbow trapping and light stopping devices,[6] directive antennas,[7] and cloaking[8,9] are the main motivations because of their potential in fields ranging from astrophysics, defense, medicine, or integrated nanophotonics. As one of the best designs for negative index materials at optical frequencies, the fishnet has attracted a considerable amount of attention, and possible low loss operation in this structure is being addressed using optical gain.[10] However, even a loss-free optical fishnet will not make a superlens because its index is unambiguously defined and negative only for normal incidence while a single light ray cannot form a subdiffractional image. Recent studies have also shown that imaging using the complex dispersion of fishnet metamaterial leads to degraded performance compared to conventional imaging systems due to the narrow acceptance angle of the fishnet.[11] While most applications, and especially the perfect lens, critically require a direction-independent negative refractive index, to the best of our knowledge, optical isotropy has not been realized for any of the structures reported so far. A theoretical proposal has been made for a structurally symmetric negative index, from microwave up to terahertz frequencies, but these systems suffer from issues limiting the possibility to push their operation to the optical domain. The structure reported in Ref. 12 has high spatial symmetry which is, however, not an equivalent of isotropy[13] and requires a lithographic resolution of 3 nm for operation at a 2-μm wavelength. Coupled coaxial waveguides[14] were theoretically proposed for a broad angle negative refractive index, but it is still anisotropic since its index varies with the angle. Designs based on coupling of waveguides are two dimensional in nature[15] and will hardly lead to a three-dimensional (3D) isotropic system. Physically, it is not possible to have the tensor response of a single nondegenerate atomic transition be isotropic (linear and equal in all directions).[16] The isotropy can either come from an averaging process of randomly oriented anisotropic particles if the transition is nondegenerate, or from the impossibility to separately excite the transitions if they are degenerate.

An array of four metal nanoparticles was proposed for a pure magnetic response.[17,18] A magnetic mode was also experimentally observed in a unit cell of core shell nanoclusters,[19] but this system also failed in providing an electric mode that is spectrally overlapping with the magnetic mode.

There have been two fundamental issues with the application of random particles that leads to negative index metamaterials. The first is that metamaterials with a negative index have employed long wires for negative permittivity, thus preventing the use of discrete particles. The second is that, even when discrete resonators are combined,[20] two subsets of particles have been used to separately control the permittivity and the permeability. In such dual-component systems, it is difficult achieve the required mixture. A single-particle approach that can exhibit strong electric and magnetic moments with sufficient overlap is thus crucial for the success of isotropic negative index. In this Rapid Communication, we identify a random monometatomic strategy to the negative index by using discrete metaparticles with overlapping electric and magnetic dipoles. We also show that it is possible to control the relative contribution of second-order degenerate multipolar radiation departing from the solely dipole moments reliably controlled so far. This would open unique avenues for the exploration of wave transport phenomena in random metamaterials as well as the design of more complex nanoantennas.

The meta-atoms under consideration are presented in Fig. 1. The first is a split-ring resonator with one gap (odd), and the second is a cut wire pair with its symmetry broken along the long side of the wire.[21] We argue that a random distribution (positions and orientations) of these unit cells can lead to an isotropic and polarization-independent negative index.

The meta-atoms under consideration are bianisotropic and, as such, bianisotropic mixing rules should be applied to predict the properties of the composites. A split-ring resonator (SRR) is well recognized as a magnetic meta-atom.[22] However, the SRR is also an electric meta-atom, and this response was recently used to demonstrate a nonmagnetic cloak.[23] The decomposition of Fig. 1(a) shows that the electric and magnetic modes share the same structural resonance. This will be the case for all ring resonators symmetrically split with an odd number of gaps. In a periodic system of odd SRRs, it has not been possible to simultaneously exploit the electric and magnetic response because the dipole moment parallel to the wave vector does not contribute to the scattering. However, in a random distribution of odd split-ring resonators, the two dipole moments can be simultaneously excited while the resonances are naturally overlapping, resulting in an isotropic metamaterial. Odd-ring resonators can make a negative index only in random media (illustrated here with

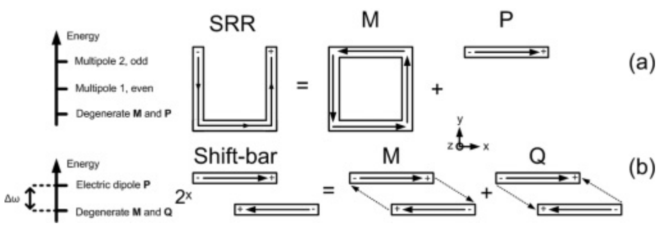

FIG. 1. Energy diagram of the degenerate transitions of symmetrically cut, odd- and even-ring resonators. (a) Decomposition of the fundamental resonance of a one-gap split-ring resonator as a superposition of magnetic (**M**) and electric (**P**) dipole resonances. (b) The antisymmetric mode of a broken symmetry bar pair (shift bar) as a superposition of magnetic (**M**) dipole and electric quadrupole (**Q**) modes. The dashed lines represent displacement currents. Cartesian coordinates with origins at the center of the particles are used.

one gap). A symmetrically cut ring resonator or bars with even number of gap have degenerate magnetic dipole and electric quadrupole moments,[24] but in general no overlapping electric and magnetic dipole moments in frequency if the bars are symmetrically aligned. However, breaking the symmetry of the double bars by shifting one of them in the direction of the wires [Fig. 1(b)] allows an overlap. A random distribution of such broken symmetry bar pairs also builds up an isotropic negative index composite. A nonisotropic negative index can be achieved in a periodic arrangement of broken symmetry even rings (see Ref. 25).

The most general way of describing an electromagnetic effective medium is given by

$$D = \bar{\varepsilon} E + \bar{\xi} H,$$
$$B = \bar{\zeta} E + \bar{\mu} H,$$

where ε, μ, ξ, and ζ are the permittivity, permeability, and magnetoelectric coupling terms, respectively. Adopting the 6×6 vector notation, the effective material parameter $\mathbf{M}_{\mathrm{eff}}$ gives the relation between the vacuum field $\mathbf{e}$ and the average flux densities $\langle \mathbf{d} \rangle$, $\langle \mathbf{d} \rangle = \mathbf{M}_0 \mathbf{e} + \langle \mathbf{P} \rangle = \mathbf{M}_{\mathrm{eff}} \mathbf{e}$, $\langle \mathbf{P} \rangle$ being the average polarization six vector. In order to predict the properties of the mixture, the polarizability of the elementary particle thus needs to be known.[26,27] A host dielectric with a refractive index of 1.33 (water) is considered in our calculations for the sake of possible realization. However, any isotropic nonmagnetic background will simply modify the resonant frequencies of the particles while the underlying physics remains the same. The general form of the polarizability tensor of a split-ring resonator has already been reported in numbers of previous papers (Ref. 28 and references therein). The multipole decomposition[29] of the field as well as the calculation of the dynamic electric and magnetic polarizabilities are carried out numerically using a three-dimensional finite-element Maxwell equation solver. The dimensions of the square metallic SRR resonator are $w = 60$ nm (width), $t = 30$ nm (thickness), and $L = 240$ nm (side length). Figure 2 confirms quantitatively that the electric and magnetic resonances indeed occur at the same frequency. For a split-ring resonator particle, the electric quadrupole resonance occurs at a higher frequency than the degenerate electric magnetic dipoles. Using the Maxwell-Garnett formalism the effective permittivity and permeability of a random distribution of such split-ring particles are presented on Fig. 2(c) as a function of the filling fraction at $\lambda = 2.4$ μm where the two polarizabilities are negative. The number of particles per unit volume n is simply related to the filling fraction f and the volume of the particle (cube enclosing the particle) by $f = n v_{\mathrm{particle}}$. We found that the permittivity and permeability are simultaneously negative for

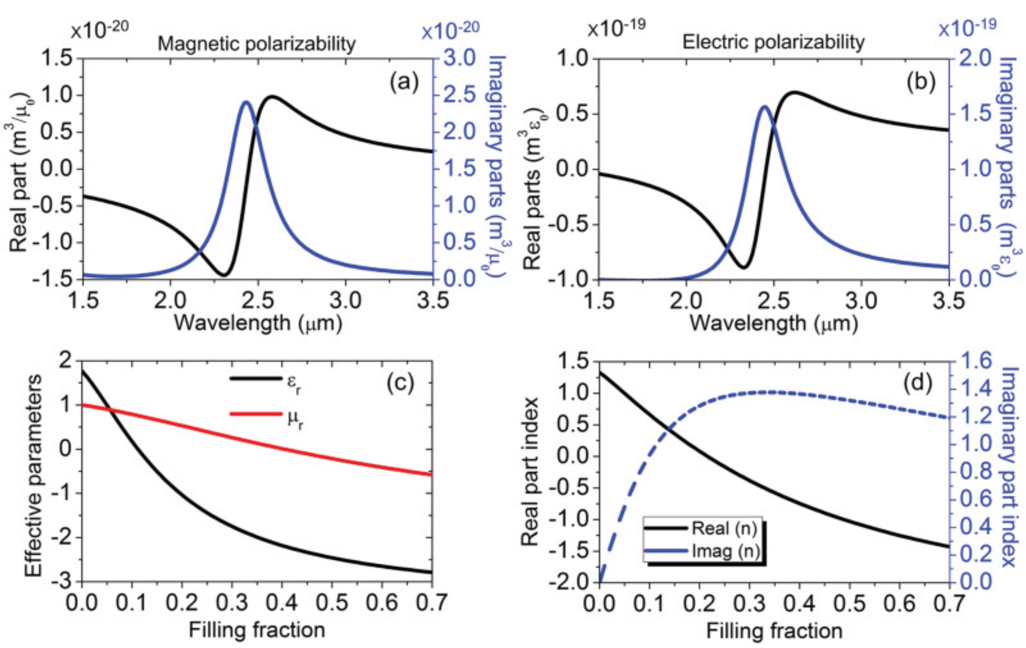

FIG. 2. (Color online) (a) Magnetic and (b) electric polarizabilities of the one-gap split-ring resonator. The black (continuous black) and blue (continuous dark gray) curves are real and imaginary parts, respectively. (c) Real parts of the relative permittivity (continuous black) and permeability (red, continuous dark grey) as a function of the filling fraction at $\lambda = 2.4$ μm and (d) the corresponding effective index (real and imaginary parts) for randomly distributed split-ring resonators.

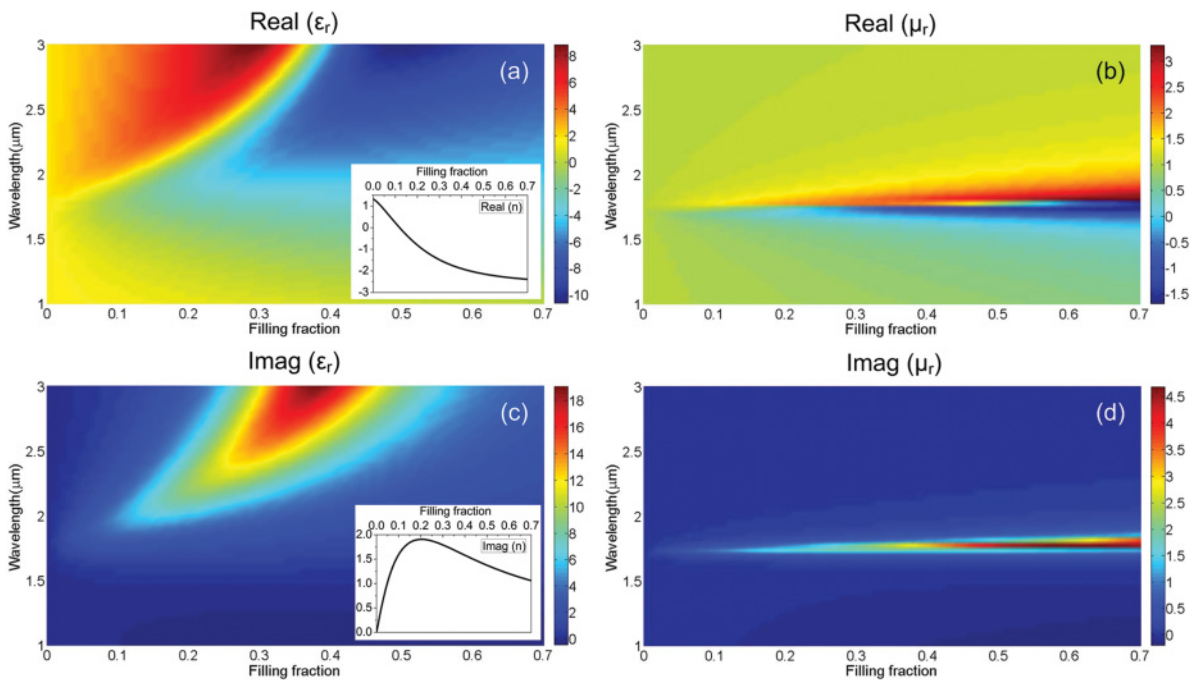

FIG. 3. (Color online) Real and imaginary parts of the relative permittivity (a), (c) and permeability (b), (d) as a function of the wavelength and the filling fraction for a randomly distributed shift bar in water with shift= 120 nm. The dimensions of the particle are $L = 340$ nm (length), $w = 40$ nm (width), $t = 30$ nm (thickness), and $s = 70$ nm (separation). The insets in (a) and (c) are the real and imaginary parts of the effective refractive index as a function of the filling fraction at 1.7 μm.

a filling fraction of ∼40%. This filling fraction can be reduced by working at lower frequencies where the polarizabilities will be more important due to a lower Drude loss from the metal. We underline here that a comparable filling fraction has been recently reported experimentally for the study of metallic nanoparticles.[30] The high confinement of fields in SRRs at the fundamental resonance also prevents interparticle coupling. The negative index obtained for a lower filling fraction at ∼21% [Fig. 2(d)] corresponds to a single negative medium. Our further calculation revealed that the effective parameters are not negative for a random split-ring resonator with one gap in the visible region. The first reason is the increasing contribution of the kinetic energy of electrons at higher frequencies,[31] and the second reason is the fact that, contrary to the case where the resonators are aligned, only one third of the polarizability of a split-ring resonator now works to cancel the incident field for a given direction. The saturation of the magnetic response thus comes earlier in frequency (infrared) in the random system compared to a fully aligned configuration where it occurs in the visible. However, the maximum frequency can be increased by using split rings with an increasing odd number of gaps.[31] The ring geometry reported in Ref. 32 for visible wavelengths can indeed be regarded as a multigap SRR, where conduction currents are progressively replaced by displacement currents.[32] However, this geometry does not exhibit a degeneracy of the electric and/or magnetic resonance, irrespective of the parity of the number of particles, making it unsuitable for isotropy. The effective bianisotropic parameter is proportional to the effective chirality in the 3D random medium. Since SRRs have a plane of symmetry, we can conclude that the effective magnetoelectric coupling should be zero in the random system.

The difficulty to scale SRR to higher frequencies calls for a different design. We propose the shift-bar system of Fig. 1(b), which is, to the best of our knowledge, the only discrete particle reported so far that can be potentially scaled to the visible and exhibit strong overlapping magnetic and electric dipole moments. In this system, $\Delta\omega$, the frequency difference between electric and magnetic dipoles, can be continuously altered from positive to negative values by the level of symmetry breaking, i.e., the shift between two bars. Positive $\Delta\omega$ corresponds to the usual situation with the magnetic mode at a lower energy compared to the electric mode, while negative $\Delta\omega$ corresponds to the opposite situation. The structure can be described by the electric, magnetic, and magnetoelectric polarizabilities tensors of the form

$$\overline{\overline{\alpha_{ee}}} = \begin{pmatrix} \alpha_{eexx} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \overline{\overline{\alpha_{mm}}} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \alpha_{mmzz} \end{pmatrix},$$

$$\overline{\overline{\alpha_{em}}} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ \alpha_{emxz} & 0 & 0 \end{pmatrix}.$$

We shall ensure that, for the description with the tensors above, the matrix coefficients are nonzero. Apart from a brute force calculation of these coefficients, a simple gedanken experiment can be performed. It is obvious that α_{eexx} and α_{mmzz} can be taken to be nonzero. Let us now consider the plane-wave excitation at a normal incidence of the particle of Fig. 1(b) with

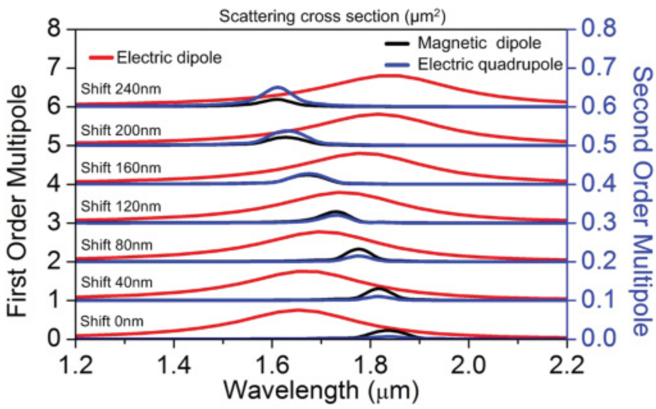

FIG. 4. (Color online) Scattering cross sections (μm^2) of the electric dipole (left axis) as well as the electric quadrupole and magnetic dipole (right axis) of the shift-bar particle in water for different shifts.

$\mathbf{E} = E_0\,\mathbf{e_x}$ and $\mathbf{k} = k_0\,\mathbf{e_z}$. It is not possible to excite the magnetic dipole because the two cut wires sit in an equiphase plane of the exciting field. We can thus conclude that $\alpha_{emxz} = 0$. The electric and magnetic polarizabilities are numerically calculated as a function of the relative shift of the bars. While the dipoles modes are spectrally separated in the initial configuration (shift $= 0$ symmetric case), they largely overlap and cross each other for a sufficient shift of the bars (Fig. 4). The coupling strength κ between the two bars in the shift-bar particle can be expressed as $\Delta\omega = \omega_+ - \omega_- = \kappa\omega_0$, where ω_+ and ω_- are frequencies of the symmetric and antisymmetric modes, respectively, and ω_0 is the resonance frequency of uncoupled bars. The crossing of the eigenmodes of the structure can thus simply be interpreted as a negative coupling between the particles, and a random collection of such negatively coupled bars builds an isotropic negative index metamaterial. To illustrate the possibility of achieving a negative index in a random distribution of such particles, we calculated the effective parameters as a function of the filling fraction (see Fig. 3) for the shift-bar particle with the bars also embedded in water. It appears that a filling fraction of $\sim$23% at $\sim$1.7 μm generates a negative index medium. At that wavelength, the medium is double negative and the effective index is $n \sim -1$. The real and imaginary parts of the index are presented in the insets of Fig. 3(a) and 3(c), respectively. It can also be noticed in Fig. 3 that resonance wavelength of the permittivity moves to a longer wavelength with the filling fraction. This can be understood simply as a continuous transition of the electric response toward a Drude model.

For the shift-bar particles, the magnetic dipole and electric quadrupole are degenerate. They are contribution from, respectively, the antisymmetric and symmetric parts of the current in the symmetrical bar pair [Fig. 1(b)]. It is thus not possible to suppress preferentially one of them in this system. However, the relative contribution of these second-order multipolar radiations surprisingly can be controlled by symmetry breaking (Fig. 4). A larger relative shift of the bars decreases the magnetic dipole while it enhances the electric quadrupole. For the non-shift-bar meta-atom (shift $= 0$), the scattering cross section of the magnetic dipole is $\sim$3.5 times that of the electric quadrupole. In contrast, for the meta-atom with a shift of 240 nm, the electric quadrupole contribution becomes $\sim$2.75 times more than the magnetic dipole. This can be qualitatively explained by the progressive elimination of the loop needed to obtain a magnetic moment. We have thus identified a method to control the higher-order multipolar moment in metamaterials. This unique ability as well as the recent identification of the toroidal moment in metamaterials[33] is in sharp contrast with works reported so far where the control was limited to the lower-order electric and magnetic dipoles, and will open an avenue in the control of more elaborated metastructures as well as the design of complex nanoantennas. The resonant modes (electric, magnetic dipoles, and electric quadrupole) of our meta-atom can, for example, be made to spectrally overlap (shift $\sim$120 nm) so that we can create a metamaterial superscatterer.[34]

The extent to which mixing theories actually predicts the behavior of the complex mixture has been a long-standing question.[35] However, we have validated our calculations in periodic systems, more stringently in terms of interparticle coupling (see Ref. 25). A more elaborated calculation such as Monte Carlo simulations could be used to better estimate the coherent coupling between the particles.[36,37] Such calculations are, however, beyond the scope of this Rapid Communication. Nevertheless, our work revealed a class of particles that can achieve an isotropic optical negative index, but also calls for experiments in those systems. Recent studies revealed that broader loss should be expected in real experiments,[30] especially in the presence of multipolar radiation. The principal contribution of this Rapid Communication lies in the classification of ring resonators and the identification of discrete particles with overlapping electric and magnetic responses at optical frequencies. The particles proposed here can be made by bottom-up self-assembly methods, or top-down fabrications. The meta-atoms could then be dispersed into any liquid or gel media, followed by condensation to fabricate the 3D isotropic negative index medium. Such explorations will be crucial for further developments in the field of plasmonic metamaterials.

In conclusion, we have demonstrated a random monometatomic route to a 3D isotropic negative index based on ring-resonator symmetry and/or parity. We showed that symmetrically cut ring resonators with an odd number of gaps and broken symmetry ring resonators with an even number of gaps lead to an isotropic negative index if randomly distributed in a host medium. The proposal conceptually differs from strategies reported so far and is based on discrete particles, each with overlapping and sufficiently strong electric and magnetic dipole moments. We have also identified a mechanism toward the control of second-order multipoles of a metamaterial. This work opens a door for electromagnetic field control beyond the effective-medium approach as well as mixing using active and nonlinear metamaterials.

We would also like to note that we recently became aware of another paper[38] on a similar topic.

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