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# Coherent and incoherent metamaterials and order-disorder transitions

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We present a concept of "coherent" and "incoherent" metamaterials, which show distinctly different resonant behavior upon disordering of their initially regular lattices. In the case of a coherent metamaterial a regular ensemble of metamolecules exhibits a collective narrow-band response that becomes broader and eventually disappears with increasing disorder, while in an incoherent metamaterial the disorder has little effect on the structure's resonant properties.

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Artificial electromagnetic metamaterials provide a uniquely fertile ground for achieving all kinds of unusual functionalities: they show a negative index of refraction required for the creation of diffraction-free superlenses,[1] exhibit strong optical magnetism,[2,3] and impose asymmetric transmission of light.[4] Metamaterial structures can be invisible,[5] act as electromagnetic cloaks,[6,7] show exceptionally high or zero refractive indices,[8] and even behave like optical frequency "superconductors" repelling the magnetic field of the optical wave, thus mimicking the Meissner effect.[9] The recent development of self-assembly techniques for fabrication of metamaterials that yield randomized arrays of metamolecules[10] and new ideas for using metamaterials in coherent sources of electromagnetic radiation[11] stimulated our interest in the effects of positional disorder on the electromagnetic properties of two-dimensional metamaterial arrays. In this work we have identified two distinctively different classes of artificial structures, the "coherent" and "incoherent" metamaterials with narrow-band resonant spectral response. The response of incoherent metamaterials is essentially determined by the properties of the individual metamolecules and is virtually insensitive to positional disorder. In "coherent" metamaterials, external electromagnetic excitation induces a magnetic response in individual metamolecules. Here the metamolecular disorder dramatically modifies the spectral response and the magnetization of the structure exhibits a collective, i.e., "coherent" nature, underpinned by short- and long-range interactions between the magnetic moments of the metamolecules.

Metallic ring metamolecules are probably the most popular building blocks of metamaterial structures. They are employed to provide a strong magnetic response with negative permeability, required for achieving a negative index of refraction. For this purpose, both the ring and the wave propagation direction lie in the same plane so that the circular current induced on the ring creates a magnetic moment *parallel* to the magnetic field of the incident wave. Here we investigate two types of metallic planar ring metamaterials supporting electromagnetic modes with high-quality factors,[12] where the magnetic moments associated with the induced currents are *perpendicular* to the plane of the array, while the incident wave is normal to this plane and, therefore, the interaction of the induced magnetic moments with the magnetic field of the incident wave is negligible.

Metamolecules of the first type were formed by pairs of concentric metal rings, while metamolecules of the second type had the form of asymmetrically split rings (Fig. 1). The metamaterial samples were manufactured from 1.6-mm-thick FR4 PCB laminates using photolithography techniques. The radius of the asymmetrically split ring structure was 6 mm, while the length of the arcs corresponded to angles of 160 and 140°. The radii of inner and outer rings forming pairs of concentric rings were 4.5 and 5.45 mm, respectively. The unit cells of the regular structures had the size of $15\times 15$ mm$^2$, which ensured no diffraction at frequencies below 20 GHz. Disorder was introduced by displacing the center of each unit cell according to a random uniform distribution defined in the square interval $x\epsilon(-\alpha/2,\alpha/2), y\epsilon(-\alpha/2,\alpha/2)$. The disorder parameter, $D$, was defined as the ratio of $\alpha$ over the unit-cell side and was varied between 0 and 1.33. All the experiments were performed in an anechoic chamber using linearly polarized broadband horn antennas with wave-front correction optics, and normal-incidence reflectance spectra were recorded with a vector network analyzer. In the case of asymmetrically split rings the polarization of the incident electromagnetic wave was set parallel to the arcs.

A special feature of the studied structures is that both support excitation of high-$Q$ antisymmetric current modes (trapped modes[12]) resulting in a sharp reflection minima (see

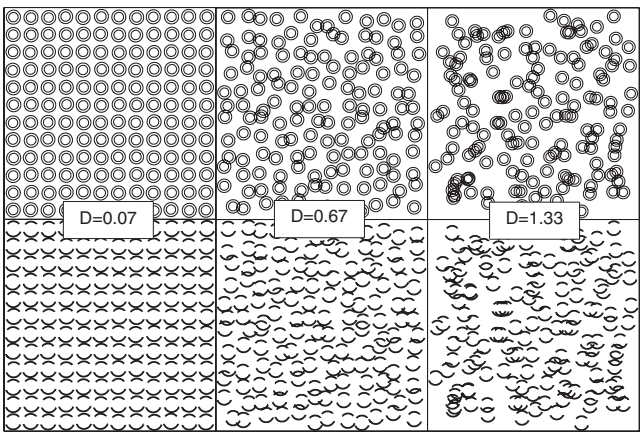


FIG. 1. Fragments of disordered metamaterial samples consisting of concentric rings (top row) and asymmetrically split rigs (bottom row). The disorder parameter, $D$, increases from left to right.

 

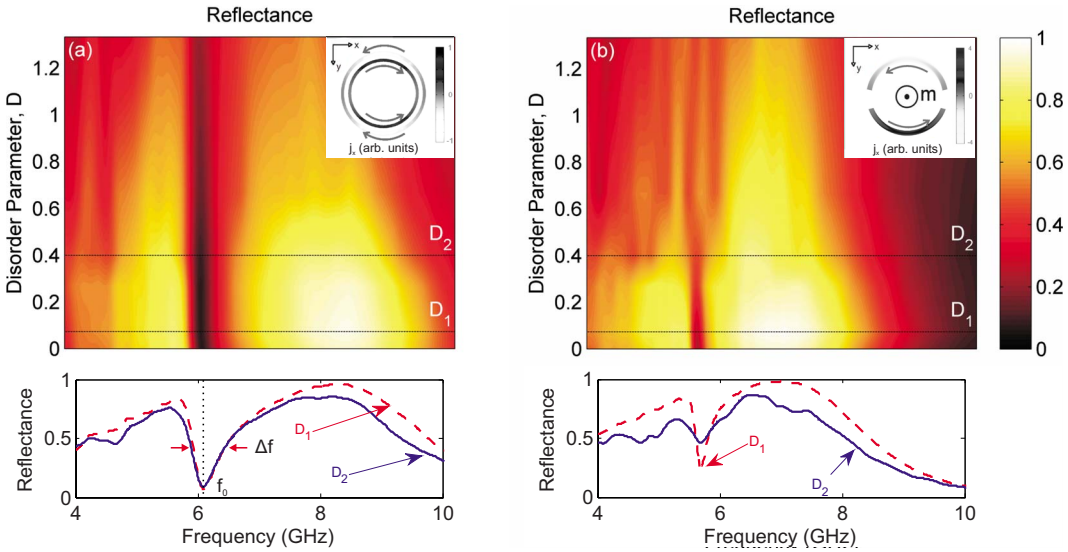


FIG. 2. (Color online) Evolution of the metamaterials' reflectance spectra with increasing disorder. Color maps (a) and (b) present the reflectance as a function of frequency and disorder parameter $D$ for the arrays of concentric and asymmetrically split rings, respectively. The two marked cross sections show reflectance profiles at $D_1=0.07$ (dashed red) and $D_2=0.40$ (solid blue), respectively. The antisymmetric current configuration is also shown for $D=0$ in the insets to the color maps.

Fig. 2). In an asymmetrically split ring, the arcs have closely spaced resonant frequencies due to the small difference in arc length. In between these frequencies, an antisymmetric mode is formed by standing current waves in the short and long arcs oscillating with opposite phases [see inset to Fig. 2(b)]. The waves radiated by such a current configuration interfere destructively leading to very weak dipole scattering in the far field. As a result an incident wave with frequency tuned at the trapped-mode resonance propagates through the metamaterial almost unaffected, while for the frequencies just below and above the resonance it is strongly reflected, making the band of suppressed reflection a narrow spectral feature. Similarly, in the case of a concentric-ring structure, counterpropagating currents are induced in neighboring sections of the inner and outer rings [see inset to Figs. 2(a)]. Weak far-field scattering by the antisymmetric current modes ensures low radiation losses, which results in efficient accumulation of energy in the ring resonators in the form of high amplitude current oscillations and naturally leads to high-quality factors of the resonances.[12]

Despite the similarity in the nature of the trapped-mode resonances the behavior of the two metamaterials becomes very different upon disordering their lattices. The color maps in Fig. 2 show the evolution of the reflectance spectra with increasing disorder. For the concentric-ring metamaterial, the resonant dip in the reflectivity robustly retains its magnitude and width even for very high levels of disorder, which may be seen as the persistence of the dark band centered at 6.2 GHz. For the asymmetrically split ring metamaterial, however, the similar resonant dip at 5.7 GHz degrades rapidly and completely vanishes for a moderate degree of disorder. This is further illustrated in Fig. 2 with two characteristic spectral plots of the reflectance corresponding to cases of weak ($D_1=0.07$) and moderate ($D_2=0.40$) disorder. The dramatic difference in the behavior of the two metamaterials becomes even more apparent, when the dependence of their resonance lifetime (estimated by fitting Fano profiles to the experimental curves) on the disorder is considered (see Fig. 3). In an ensemble of concentric rings, the resonant line is practically independent of disorder and the resonance retains its width and position even at very high degrees of disorder. On the contrary, in the case of asymmetrically split ring arrays, the linewidth appears to be extremely sensitive to disorder. Even for moderate degrees of disorder the resonance lifetime rapidly drops to zero, meaning that practically no resonant features exist in the spectrum.

This difference can be understood on the basis of the re-

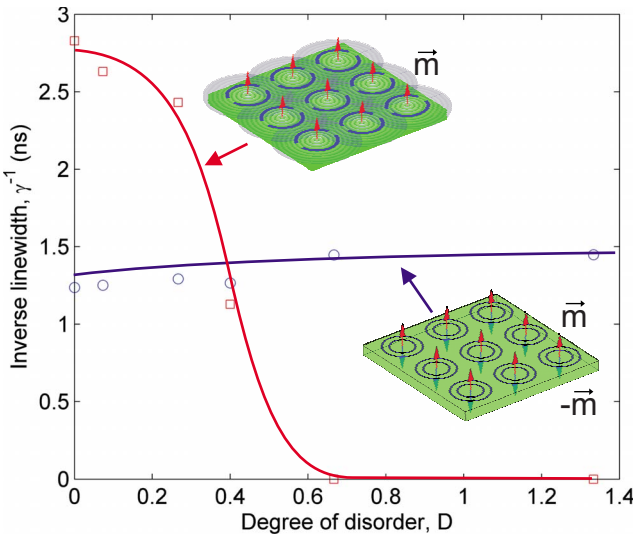


FIG. 3. (Color online) Inverse linewidth of the metamaterials as a function of the disorder parameter plotted for arrays of concentric (blue circles) and asymmetrically split (red squares) rings. Points correspond to experimentally measured values, while lines serve as guides for the reader. The insets illustrate the nature of interactions between the metamolecules.

spective resonant current configurations. In the case of the concentric-ring metamaterials, the antisymmetric currents of the inner and outer rings create magnetic moments pointing in opposite directions (see inset to Fig. 3), thus rendering the total magnetic response of each metamolecule extremely weak. Furthermore, by design, the resonant electric response of the concentric rings is also very weak. Therefore, mutual interactions between the metamolecules (both of electric or magnetic dipole type) are negligible, and the collective response of the arrays does not depend on the positions (or disorder) of the molecules being just a direct sum of the individual molecular contributions. On the contrary, the currents induced in the upper and lower arcs of the split rings oscillate in opposite phases and therefore create coherent magnetic-dipole moments oscillating in phase. These magnetic moments are oriented normal to the plane of the array. In a regular array, interference of waves reradiated by the oscillating magnetic dipoles results in the so-called magnetoinductive waves,[13] which are confined to the plane of the array mediating efficiently strong interactions between the metamolecules (see illustration in the inset to Fig. 3). The increase in disorder leads to intense scattering of these waves into free space on lattice irregularities, thus introducing losses that reduce the strength of the antisymmetric current mode. According to the alternative (but equivalent) approach, when disorder is introduced, the magnetic dipoles are allowed to radiate in directions outside the plane of the array, resulting in diffuse reflectance and increase in scattering losses. Consequently, the intermolecular interactions become weaker, resulting in a broader resonance and vanishing net magnetization.

The coherent and incoherent natures of the metamaterials can be further illustrated by considering the limiting case of single isolated metamolecules. Figure 4 shows a comparison between the scattering spectra of a self-standing periodic infinite array and a single unit cell for both asymmetrically split and concentric-ring resonators obtained through full three-dimensional finite element calculations. Since an infinite array can only scatter along directions normal to the array plane, the scattering properties of the array can be quantified by its reflectance (essentially scattering in the "backward" direction). One should note that scattering from a single concentric-ring resonator and reflection from an infinite periodic array of such resonators [Fig. 4(b)] are very similar: in both cases, the spectrum presents a sharp minimum as a signature of the trapped-mode resonance. The width at half maximum of the trapped-mode resonances is estimated as 1.219 and 1.005 GHz, approximately, for the single metamolecule and the metamaterial array, respectively. On the contrary, an array of asymmetrically split rings behaves completely different from a single asymmetrically split ring resonator. While the array exhibits a sharp, 228-MHz-wide (full width at half maximum) reflection dip corresponding to a minimum of scattering losses, an isolated resonator shows no scattering minimum [Fig. 4(a)] but a scattering maximum which is approximately three times broader (FWHM $\simeq$ 733 MHz) than the array resonance. Hence, in the case of asymmetrically split rings, suppression of scattering can only occur when an array is considered and, consequently, the observed resonant reflection dip is a collective property, which is absent in a single resonator.

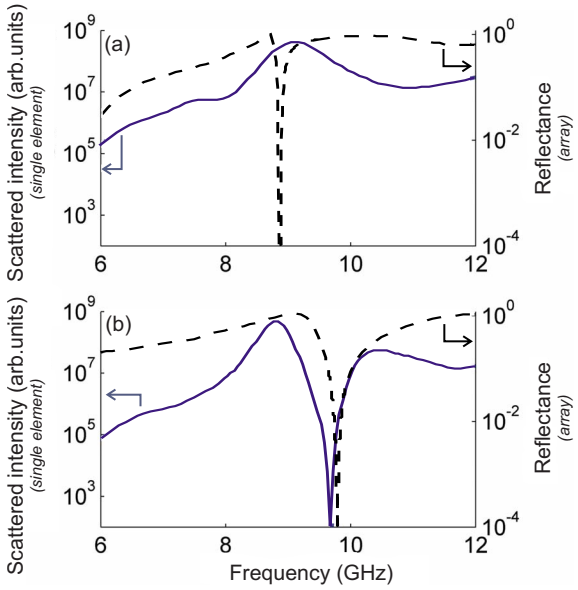


FIG. 4. (Color online) Electromagnetic scattering from self-standing single metamolecules and metamaterial arrays of asymmetrically split rings (a) and concentric rings (b). Blue solid lines represent scattered intensity for single unit-cell resonators, while black dashed lines correspond to the reflectance of a double-periodic infinite array.

Thus, the disorder-induced broadening/weakening of the metamaterial collective response cannot be explained by splitting or shifting of the high-$Q$ resonances of the individual metamolecules that form clusters of closely spaced (or even overlapping) split rings in the disordered arrays as the resonance simply does not exist for an individual metamolecule. This is in contrast to ordinary metamaterials, where the behavior of a single metamolecule and a metamaterial array is very similar[14,15] and renders the case presented here fundamentally different from the recently investigated effects of disorder in photonics crystals[16] and metamaterials,[17–19] where random changes in the geometry of the individual metamolecules actions affected the line shape (spectral shift and split) leading to inhomogeneous broadening. This is a trivial case that is not relevant to the "coherent" metamaterial behavior considered here. In "coherent" metamaterials, narrow lines are *not seen* in individual metamolecules and appear only in ordered arrays. Here, the sharp spectral feature is a collective resonance of the entire ordered structure and randomization leads to radiation damping of the coherent response of the array.

We would like to point out a phenomenological relation of the coherent asymmetrically split ring metamaterial to a well-known phenomenon in many-body physics, namely, the Mössbauer effect,[20] where transitions of atomic nuclei with extremely narrow linewidths can only be observed when the atoms form a crystal lattice, while in gas phase the emission line is strongly broadened by the recoil during the emission of the high-energy gamma quanta. In a similar way, a single metamolecule of the coherent metamaterial or a strongly disordered array does not exhibit the narrow resonance associated with the antisymmetric current mode (also known as

trapped-mode resonance) as a result of scattering losses, while in a regular array no scattering or diffraction losses are possible for wavelengths longer than the array pitch and thus a low-frequency high-quality mode is formed.

The two planar metamaterial structures considered above are examples of artificial media with strong and weak intermetamolecular interactions representing two characteristic and antipode classes of what we call "coherent" and "incoherent" metamaterials. The fundamental difference in the nature of their narrow resonances, collective in the first case and individual in the latter, determines the potential applications of these structures, most notably the lasing spaser.[11] The lasing spaser, a metamaterial analog of spaser,[21] is a planar narrow-diversion coherent source of electromagnetic radiation that is fuelled by plasmonic oscillations of a two-dimensional resonator array. Here the coherency of the optical source is ensured by the synchronous oscillations of the plasmonic currents in the array. In a "coherent" metamaterial formed by asymmetrically split rings, the regular array gives the highest value of quality factor compared to disordered arrays. We argue that, similarly, when both phased (coherent) and uncorrelated (incoherent) current oscillations are present in an array of metamolecules of this type, the uncorrelated component will decay more rapidly. Therefore in the presence of gain the phased coherent component of amplified spontaneous current fluctuations will win over incoherent fluctuations providing for a self-starting regime of the lasing spaser. On the contrary, "incoherent" metamaterials with weak intermolecular interactions, such as concentric rings, do not possess a mechanism of synchronization of current oscillation in individual metamolecules and are not suitable for lasing spaser applications. However, the resonant properties of "incoherent" metamaterials are more tolerant to disorder, making them more suitable for manufacturing using methods prone to imperfections such as self-assembly.[10]

Finally, another intriguing and unique property of the "coherent" metamaterials will be the dependence of their electromagnetic response on the size of the sample array since narrow-band resonance is only featured in the spectrum when a large number of metamolecules are involved in the formation of the response. Complete characterization of the transition from single resonator behavior to array's collective response remains the subject of our current investigations and will be reported elsewhere.

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