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Cooperative resonance linewidth narrowing in a planar metamaterial

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Abstract. We theoretically analyse the experimental observations of a spectral line collapse in a metamaterial array of asymmetric split ring resonators (Fedotov *et al* 2010 *Phys. Rev. Lett.* **104** 223901). We show that the ensemble of closely-spaced resonators exhibits a cooperative response, explaining the observed system-size dependent narrowing of the transmission resonance linewidth. We further show that this cooperative narrowing depends sensitively on the lattice spacing and that significantly stronger narrowing could be achieved in media with suppressed ohmic losses.

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1. Introduction

Resonant multiple scattering plays an important role in mesoscopic wave phenomena. Such phenomena can be realized with electromagnetic (EM) fields. In the strong scattering regime, interference of different scattering paths between discrete scatterers can result in, e.g. light localization [1, 2]—an effect analogous to the Anderson localization of electrons in solids. Metamaterials comprise artificially structured media of plasmonic resonators interacting with EM fields. Due to several promising phenomena, such as the possibility for diffraction-free lenses resulting from negative refractive index [3], there has been a rapidly increasing interest in fabrication and theoretical modelling of such systems. Additionally, the discrete nature of closely-spaced resonators in typical metamaterial arrays raises the possibility to observe strong collective radiative effects in these systems.

In recent experiments Fedotov et al [4] observed a dramatic suppression of radiation losses in a two-dimensional (2D) planar metamaterial array. The transmission spectra through the metamolecular sheet was found to be strongly dependent upon the number of interacting metamolecules in the system. The transmission resonance quality factor increased as a function of the total number of active resonators, finally saturating at about 700 meta-molecules. The metamaterial unit cell in the experiment was formed by an asymmetric split ring (ASR) resonator, consisting of two circular arcs of slightly unequal lengths. The currents in these ASRs may be excited symmetrically (antisymmetrically), yielding a net oscillating electric (magnetic) dipole as shown in figure 1.

In this paper we theoretically analyse the collective metamaterial response, observed experimentally by Fedotov et al [4]. We find that strong interactions between a discrete set of resonators, mediated by the EM field, characterize the response of the ensemble and results in collective resonance linewidths and frequencies. We show how the cooperative response of sufficiently closely-spaced resonators is responsible for the observed narrowing of the transmission resonance linewidth (increasing quality factor) with the number of resonators [4]. In particular, the system exhibits a collective mode with an almost purely magnetic excitation, uniform phase profile, and strongly suppressed radiative properties with each ASR possessessing a nearly equal magnetic dipole moment. We show in detail how this mode can be excited by an incident plane wave propagating perpendicular to the array through an electric dipole coupling to an ASR, even when the magnetic dipole moments are oriented parallel to the propagation direction. We calculate the resonance linewidth of the phase-coherent collective magnetic mode that narrows as a function of the number of ASRs, providing an excellent agreement with experimental observations. At the resonance, and with appropriately chosen parameters, nearly all the excitation can be driven into this mode. Due to its suppressed decay rate, the transmission spectrum displays a narrow resonance. The linewidth is sensitive to the spacing of the unit-cell resonators, with the closely-spaced ASRs exhibiting cooperative response, due to enhanced dipole-dipole interactions. We find that the narrowing is limited by the ohmic losses of the ASR resonators and that a dramatically stronger narrowing could be achieved with a media exhibiting suppressed ohmic losses.

Our analysis demonstrates how essential features of the collective effects of the experiment in [4] can be captured by a simple, computationally efficient model, developed in [5], in which we treat each meta-atom as a discrete scatterer, exhibiting a single mode of current oscillation and possessing appropriate electric and magnetic dipole moments. Interactions with the EM

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Figure 1. ASR meta-molecules. (a) A schematic illustration of the two constituent meta-atoms of an ASR separated by distance u. Symmetric current oscillations produce parallel electric dipoles (blue arrows) but antiparallel magnetic dipoles (red arrows). (b) The symmetric mode with the currents in the two meta-atoms oscillating in-phase and (b) the antisymmetric mode with the current oscillating π out-of-phase. For the symmetric case the dominant contribution is a net electric dipole moment in the plane of the resonator and for the antisymmetric case a net magnetic dipole moment normal to the plane of the resonator. (d) An illustration of an incident EM field driving the uniform phase-coherent collective magnetic eigenmode in which all the meta-molecules exhibit a magnetic dipole normal to the metamaterial plane. The incident field has an electric polarization along the ASR electric dipoles, but a magnetic field perpendicular to the ASR magnetic dipoles.

field then determine the collective interactions within the ensemble. Moreover, our analysis indicates the necessity of accounting for the strong collective response of metamaterial systems and interference effects in multiple scattering between the resonators in understanding the dynamics and design of novel metamaterials. Strong interactions between resonators can find important applications in metamaterial systems, providing, e.g. precise control and manipulation of EM fields on a sub-wavelength scale [6–9], in developments of a lasing spaser [10], and disorder-related phenomena [11, 12]. Some features of interacting discrete resonators, such as the propagation of excitations through a one-dimensional chain of meta-molecules [13], could be modelled by introducing a phenomenological coupling between nearest neighbours into the Lagrangian describing the metamaterial [13, 14]. Capturing the emergence of superradiant and subradiant collective mode linewidths, however, requires one to consider the repeated emission and reabsorption of radiation between resonators as described in [5]. Radiative coupling between pairs of magnetoelectric scatterers has also been considered in [15].

The remainder of this paper is organized as follows. Section 2 reviews the essential features of the theoretical model developed in [5] that are required to analyse the transmission resonance experiments [4]. We describe the metamaterial as an ensemble of discrete scatterers, or metaatoms, that dynamically respond to the EM field. In the context of this model, we describe the ASR meta-molecule that forms the unit-cell of our metamaterial in section 3. In section 4 we show how cooperative interactions lead to the formation of the uniform magnetic mode in which all ASR magnetic dipoles oscillate in phase. We demonstrate how the quality factor of this mode increases with the size of the system. We illustrate how to excite this mode and compare the results of our model to the experimental observations of Fedotov *et al*, demonstrating a remarkable agreement. Conclusions follow in section 5.

2. Theoretical model

In order to analyse the experimental observations of the transmission spectra [4], we employ the theoretical model developed in [5]. The model provides a computationally efficient approach for the studies of strong collective EM field mediated interactions between resonators in large metamaterial arrays. In this section, we provide a brief outline of the basic results of the general formalism of [5] that are needed to analyse the collective features of the EM response observed in experiments [4]. A more detailed description of the model is provided in [5]. In the following section we apply the theory specifically to an array of ASR metamolecules.

We consider an ensemble of N metamaterial unit elements, meta-molecules, each formed by n discrete meta-atoms, with the position of the meta-atom j denoted by \mathbf{r}_j ($j = 1, ..., n \times N$). An external beam with electric field $\mathbf{E}_{in}(\mathbf{r}, t)$ and magnetic field $\mathbf{H}_{in}(\mathbf{r}, t)$ with frequency Ω_0 drives the ensemble. We assume the extent of each meta-atom is much less than the wavelength $\lambda = 2\pi c / \Omega_0$ of the incident light so that we may treat meta-atoms as radiating dipoles and ignore the higher-order multipole-field interactions. Within each meta-atom, EM fields drive the motion of charge carriers resulting in oscillating charge and current distributions. For simplicity, we assume that each meta-atom j supports a single eigenmode of current oscillation governed by a dynamic variable $Q_j(t)$ with units of charge. Then the associated electric and magnetic dipole moment for the meta-atoms are

$$\boldsymbol{d}_j = \boldsymbol{Q}_j \boldsymbol{h}_j \boldsymbol{d}_j, \tag{1}$$

$$\boldsymbol{m}_j = I_j A_j \hat{\boldsymbol{m}}_j, \tag{2}$$

respectively, where \hat{d}_j and \hat{m}_j are unit vectors denoting the dipole orientations and $I_j(t) = dQ_j/dt$ is the current. Here h_j and A_j are the corresponding proportionality coefficients (with the units of length and area) which depend on the specific geometry of the resonators. In the dipole approximation the polarization and magnetization are given in terms of the density of electric and magnetic dipoles

$$\boldsymbol{P}(\boldsymbol{r}) = \sum_{j} \boldsymbol{P}_{j}(\boldsymbol{r}), \tag{3}$$

$$\boldsymbol{M}(\boldsymbol{r}) = \sum_{j} \boldsymbol{M}_{j}(\boldsymbol{r}), \qquad (4)$$

where the polarization and the magnetization of the resonator j are

$$\boldsymbol{P}_{j}(\boldsymbol{r},t) \approx \boldsymbol{d}_{j}\delta(\boldsymbol{r}-\boldsymbol{r}_{j}), \tag{5}$$

$$\boldsymbol{M}_{i}(\boldsymbol{r},t) \approx \boldsymbol{m}_{i}\delta(\boldsymbol{r}-\boldsymbol{r}_{i}), \qquad (6)$$

respectively.

The incident EM field drives the excitation of the current oscillations, generating an oscillating electric and magnetic dipole in each meta-atom. The resulting dipole radiation from the metamaterial array is the sum of the scattered electric and magnetic fields from all the

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meta-atoms

$$\boldsymbol{E}_{\mathrm{S}}(\boldsymbol{r},t) = \sum_{j} \boldsymbol{E}_{\mathrm{S},j}(\boldsymbol{r},t),\tag{7}$$

$$\boldsymbol{H}_{\mathrm{S}}(\boldsymbol{r},t) = \sum_{j} \boldsymbol{H}_{\mathrm{S},j}(\boldsymbol{r},t), \qquad (8)$$

where $E_{S,j}(r, t)$ and $H_{S,j}(r, t)$ denote the electric and magnetic field emitted by the metaatom *j*. The Fourier components of the scattered fields have the familiar expressions of electric and magnetic fields radiated by oscillating electric and magnetic dipoles [16],

$$\boldsymbol{E}_{\mathbf{S},j}^{+}(\boldsymbol{r},\,\Omega) = \frac{k^{3}}{4\pi\,\epsilon_{0}} \int \mathrm{d}^{3}\boldsymbol{r}' \left[\mathbf{G}(\boldsymbol{r}-\boldsymbol{r}',\,\Omega) \cdot \boldsymbol{P}_{j}^{+}(\boldsymbol{r}',\,\Omega) + \frac{1}{c} \mathbf{G}_{\times}(\boldsymbol{r}-\boldsymbol{r}',\,\Omega) \cdot \boldsymbol{M}_{j}^{+}(\boldsymbol{r}',\,\Omega) \right],\tag{9}$$

$$\boldsymbol{H}_{\mathbf{S},j}^{+}(\boldsymbol{r},\,\Omega) = \frac{k^{3}}{4\pi} \int \mathrm{d}^{3}\boldsymbol{r}' \left[\mathbf{G}(\boldsymbol{r}-\boldsymbol{r}',\,\Omega) \cdot \boldsymbol{M}_{j}^{+}(\boldsymbol{r}',\,\Omega) - c\mathbf{G}_{\times}(\boldsymbol{r}-\boldsymbol{r}',\,\Omega) \cdot \boldsymbol{P}_{j}^{+}(\boldsymbol{r}',\,\Omega) \right],\tag{10}$$

where we have defined the positive and negative frequency components of a time varying real quantity V(t) such that for a Fourier component of frequency Ω ($k \equiv \Omega/c$), $V^{\pm}(\Omega) \equiv \Theta(\pm \Omega)V(\Omega)$, and hence $V(t) = V^{+}(t) + V^{-}(t)$ with $V^{-}(t) = [V^{+}(t)]^{*}$. Here $\mathbf{G}(\mathbf{r} - \mathbf{r}', \Omega)$ denotes the radiation kernel representing the electric (magnetic) field observed at \mathbf{r} that is emitted from an electric (magnetic) dipole residing at \mathbf{r}' . The radiation kernel that represents the magnetic (electric) field at \mathbf{r} scattered from an electric (magnetic) dipole source residing at \mathbf{r}' is denoted by $\mathbf{G}_{\times}(\mathbf{r} - \mathbf{r}', \Omega)$. Explicit expressions for \mathbf{G} and \mathbf{G}_{\times} coincide with the standard formulae of electromagnetism describing dipole radiation [16].

Equations (9) and (10) provide the total electric and magnetic fields as a function of polarization and magnetization densities that are produced by current excitations in the metaatoms. In general, however, there is no simple way of solving for P(r) and M(r). The scattered fields from each meta-atom drive the dynamics of the other meta-atoms in the system, with the EM fields mediating interactions between the resonators. The radiated fields and the resonator excitations form a strongly coupled system when the separation between the resonators is of the order of the wavelength or less.

In order to solve the dynamics for the polarization and magnetization densities appearing in equations (9) and (10), we have derived a coupled set of equations for the EM fields and resonators [5]. In the metamaterial sample, current excitations in each meta-atom j exhibit behaviour similar to that of an LC circuit with resonance frequency

$$\omega_j \equiv \frac{1}{\sqrt{L_j C_j}},\tag{11}$$

where C_j and L_j denote effective self-capacitance and self-inductance, respectively. In this work we consider asymmetric meta-molecules consisting of two meta-atoms with different resonance frequencies ω_j , centred around the frequency ω_0 , with $|\omega_j - \omega_0| \ll \omega_0$. The oscillating electric and magnetic dipoles radiate energy from an isolated meta-atom at respective rates Γ_E and Γ_M [5] resulting in the scattered fields $E_{S,j}$ and $H_{S,j}$ (see equations (9) and (10)). The strengths of these radiative emission rates vary with the squares of the electric and magnetic dipole proportionality coefficients h_j and A_j , respectively [5]. We assume that the meta-atom resonance frequencies dominate the emission rates and that the resonance frequencies occupy

a narrow bandwidth around the dominant frequency of the incident field, i.e. $\Gamma_{E,j}$, $\Gamma_{M,j}$, $|\omega_0 - \Omega_0| \ll \Omega_0$. For simplicity, we also assume that the radiative electric and magnetic decay rates of each resonator Γ_E and Γ_M are independent of the resonator j.

The dynamics of current excitations in the meta-atom j may then be described by $Q_j(t)$ (introduced in equation (1)) and its conjugate momentum $\phi_j(t)$ (with units of magnetic flux) [5]. In the absence of radiative emission and interactions with external fields, the LC circuit, with resonance frequency ω_j , formed by the oscillating charge and current can be naturally described by the slowly varying normal variables

$$b_j(t) \equiv \frac{e^{i\Omega_0 t}}{\sqrt{2}} \left(\frac{Q_j(t)}{\sqrt{\omega_j C_j}} + i \frac{\phi_j(t)}{\sqrt{\omega_j L_j}} \right).$$
(12)

These normal variables are defined such that b_j undergoes a phase modulation with frequency $(\omega_j - \Omega_0)$, i.e. $b_j(t) = b_j(0) \exp[-i(\omega_j - \Omega_0)t]$ which is perturbed by nonzero radiative losses Γ_E , $\Gamma_M \ll \Omega_0$, and driving from the external fields.

The fields generated externally to each meta-atom j, composed of the incident field and fields scattered from all other meta-atoms in the system, drive the amplitude b_j of current oscillation within the meta-atom. The component of the external electric field oriented along the dipole direction \hat{d}_j provides a net external electromotive force (EMF), and the component of the external magnetic field along the magnetic dipole direction \hat{m}_j provides a net applied magnetic flux. The applied external EMF and oscillating magnetic flux induce current flow in the meta-atom. The oscillating current of that meta-atom, in turn, generates electric and magnetic dipoles which both radiate magnetic and electric fields, according to (9) and (10). These fields couple to dynamical variables of charge oscillations in other meta-atoms, producing more dipolar radiation. The result of this multiple scattering is that the EM fields mediate interactions between the meta-atom dynamic variables.

For the limits we consider in this paper, the metamaterial's response to the incident EM field is then governed by the set of coupled linear equations for the meta-atom variables b_i [5],

$$\dot{\mathbf{b}} = C\mathbf{b} + \mathbf{f}_{\rm in} , \qquad (13)$$

where we have introduced the notation for column vectors of normal variables b and the driving f_{in} caused by the incident field

$$\mathbf{b}(t) \equiv \begin{pmatrix} b_1(t) \\ b_2(t) \\ \vdots \\ b_{nN}(t) \end{pmatrix}, \qquad \mathbf{f}_{in}(t) \equiv \begin{pmatrix} f_{1,in}(t) \\ f_{2,in}(t) \\ \vdots \\ f_{nN,in}(t) \end{pmatrix}.$$
(14)

The applied incident fields induce an EMF and magnetic flux in each meta-atom j, producing the driving $f_{j,\text{in}}$ [5]. Under the experimental conditions we consider here, however, the meta-atom magnetic dipoles are aligned perpendicular to the incident magnetic field, and thus only the EMF contributes to the driving of each meta-atom, which is given by

$$e^{-i\Omega_0 t} f_{j,in}(t) = i \frac{h_j}{\sqrt{2\omega_j L_j}} \hat{d}_j \cdot E_{in}^+(\boldsymbol{r}_j, t) .$$
(15)

The current oscillations excited by the incident electric field then simultaneously produce electric and magnetic dipoles which scatter fields to other meta-atoms, which then rescatter

the fields. This multiple scattering between the meta-atoms results in the linear coupling matrix

$$\mathcal{C} = -i\Delta - \frac{\Gamma}{2}I + \frac{1}{2}\left[i\Gamma_{\rm E}\mathcal{G}_{\rm E} + i\Gamma_{\rm M}\mathcal{G}_{\rm M} + \bar{\Gamma}\left(\mathcal{G}_{\times} + \mathcal{G}_{\times}^{\rm T}\right)\right],\tag{16}$$

where *I* represents the identity matrix, and $\overline{\Gamma} \equiv \sqrt{\Gamma_{\rm E}\Gamma_{\rm M}}$ is the geometric mean of the electric and magnetic dipole emission rates. Here the detunings of the incident field from the meta-atom resonances are contained in the diagonal matrix Δ with elements

$$\Delta_{j,j'} \equiv \delta_{j,j'} \left(\omega_j - \Omega_0 \right) \,, \tag{17}$$

and the energy carried away from individual meta-atoms by the scattered fields manifests itself in the decay rate [5]

$$\Gamma \equiv \Gamma_{\rm E} + \Gamma_{\rm M} + \Gamma_{\rm O} \tag{18}$$

appearing in the diagonal elements of C. In the limits we consider here, a meta-atom's magnetic and electric dipoles oscillate $\pi/2$ out of phase [5] with one another. The fields radiated from a single meta-atom's electric and magnetic dipoles therefore neither constructively nor destructively interfere with each other, and an isolated meta-atom's radiative emission rate is the sum of Γ_E and Γ_M [5]. In addition to the radiative losses, we have included a phenomenological rate Γ_0 to account for nonradiative, e.g. ohmic losses. The inter-metaatom interactions produced by the scattered fields result in electric and magnetic dipoledipole interactions, accounted for by matrices \mathcal{G}_E and \mathcal{G}_M , respectively, that depend on the relative positions and orientations of the meta-atom dipoles (the precise form is given in [5]). Additionally, the *electric* field emitted by the *magnetic* dipole of one meta-atom will drive the electric dipole of another. The resulting interaction reveals itself in the cross coupling matrix \mathcal{G}_{\times} . Similarly, the magnetic field emitted by the electric dipole of one meta-atom interacts with the magnetic dipoles of others. These interactions manifest themselves as \mathcal{G}_{\times}^{T} , the transpose of the cross-coupling matrix. The interaction processes between the different resonators, mediated by dipole radiation, are analogous to frequency dependent mutual inductance and capacitance, but due to the radiative long-range interactions, these can substantially differ from the quasi-static expressions for which C_{\times} is also absent.

In order to calculate the EM response of the system, we solve the coupled set of equations (13) involving all the resonators and the fields. A system of $n \times N$ single-mode resonators then possesses $n \times N$ collective modes of current oscillation. Each collective mode exhibits a distinct collective linewidth (decay rate) and resonance frequency, determined by the imaginary and real parts of the corresponding eigenvalue [5]. The resulting dynamics resemble a cooperative response of atomic gases to resonant light in which case the EM coupling between different atoms is due to electric dipole radiation alone [17–23]. The crucial component of the strong cooperative response of closely-spaced scatterers are *recurrent* scattering events [18–21, 24, 25]—in which a wave is scattered more than once by the same dipole. Such processes cannot generally be modelled by the continuous medium electrodynamics, necessitating the meta-atoms to be treated as discrete scatterers. An approximate calculation of local field corrections in a magnetodielectric medium of discrete scatterers was performed in [26] where the translational symmetry of an infinite lattice simplifies the response.

3. The asymmetric split ring meta-molecule

In this paper, we provide a theoretical analysis of the experimental findings by Fedotov *et al* [4] and explain the observed linewidth narrowing of the transmission spectrum for a 2D metamaterial array of ASRs. We will show that the observed transmission resonance [27] and its enhanced quality factor as a function of the size of the system result from the formation of a collective mode whose decay rate becomes more suppressed for increased array sizes. Within our model, a single ASR, consisting of two meta-atoms, has two modes of oscillation, each of which decays at a rate comparable to the single, isolated meta-atom decay rate Γ . It is only when the ASRs act in concert that the transmission resonance due to linewidth narrowing can be observed. In order to understand the collective dynamics of the metamaterial, we first show how the EM mediated interactions between meta-atoms determine the behaviour of its constituent meta-molecule. To that end, it is instructive to first apply our theoretical model to describe the behaviour of a single, isolated ASR.

A single ASR consists of two separate concentric circular arcs (meta-atoms), labelled by $j \in \{1, r\}$ (for 'left' and 'right') as illustrated in figure 1. The ASR is an example of a split ring resonator, variations of which are instrumental in the production of metamaterials with exotic properties such as negative indices of refraction [28, 29]. To illustrate the qualitative physical behaviour of the ASR, we approximate the meta-atoms as two point sources located at points \mathbf{r}_r and \mathbf{r}_1 separated by $\mathbf{u} \equiv \mathbf{r}_r - \mathbf{r}_1$. The current oscillations in each meta-atom produce electric dipoles with orientation $\hat{d}_r = \hat{d}_1 \equiv \hat{d}$, where $\hat{d} \perp \hat{u}$. Owing to the curvature of each meta-atom, current oscillations produce magnetic dipoles with opposite orientations $\hat{m}_r = -\hat{m}_1 \equiv \hat{m}$, where $\hat{m} \perp \mathbf{u}$ and $\hat{m} \perp \hat{d}$. An asymmetry between the rings, in this case resulting from a difference in arc length, manifests itself as a difference in resonance frequencies with $\omega_r = \omega_0 + \delta \omega$ and $\omega_1 = \omega_0 - \delta \omega$.

Although this simplified model does not account for higher order multipole contributions of an individual meta-atom, oscillations of the ASR meta-molecule consisting of two metaatoms do exhibit nonvanishing quadrupole moments. While this quadrupole contribution can be inaccurately represented in the dipole approximation, in the case of ASR modes, the electric quadrupole moment is notably suppressed in most experimental situations when compared to the corresponding dipolar field [30]. The dipole approximation also provides an advantage in computational efficiency and in maintaining the tractability of the calculation. Despite the dipole approximation implemented in the numerics we find in section 4 that the model is able to reproduce the experimental findings of the enhanced quality factor of the transmission resonance observed by Fedotov *et al* [4].

The dynamics of a single, isolated ASR are described by the two normal variables b_r and b_l for the right and left meta-atoms, respectively. These two meta-atoms interact via electric and magnetic dipole–dipole interactions as well as interactions due to the electric (magnetic) fields emitted by the other meta-atom's magnetic (electric) dipole. The dynamics of the ASR are given by (see equation (13))

$$\frac{\mathrm{d}}{\mathrm{d}t} \begin{pmatrix} b_{\mathrm{r}}(t) \\ b_{\mathrm{l}}(t) \end{pmatrix} = \mathcal{C}^{(\mathrm{ASR})} \begin{pmatrix} b_{\mathrm{r}}(t) \\ b_{\mathrm{l}}(t) \end{pmatrix} + \begin{pmatrix} f_{\mathrm{r,in}}(t) \\ f_{\mathrm{l,in}}(t) \end{pmatrix} .$$
(19)



Figure 2. A diagram illustrating the symmetric and antisymmetric modes of an ASR driven by an electric field resonant on the antisymmetric mode. The resonance frequency of the symmetric (antisymmetric) mode is shifted up (down) from the central frequency ω_0 by δ . Radiative decay is illustrated by the decay rates γ_{\pm} of the mode variables c_{\pm} . The asymmetry $\delta \omega$ manifests itself as a coupling between the symmetric and antisymmetric modes. If the incident magnetic field is perpendicular to the magnetic dipole, it cannot excite the asymmetric mode in the absence of the asymmetry in the two halves of the split ring.

The driving terms $f_{r,in}$ and $f_{l,in}$ are given in (15), while the interaction matrix

$$\mathcal{C}^{(ASR)} = \begin{pmatrix} -i\left(\omega_0 + \delta\omega - \Omega_0\right) - \Gamma/2 & i\left(\Gamma_E - \Gamma_M\right)G - \bar{\Gamma}S\\ i\left(\Gamma_E - \Gamma_M\right)G - \bar{\Gamma}S & -i\left(\omega_0 - \delta\omega - \Omega_0\right) - \Gamma/2 \end{pmatrix}.$$
 (20)

The off-diagonal matrix elements are identical and account for the EM interactions between the two meta-atoms. Because the meta-atoms oscillating in phase produce parallel electric dipoles, but antiparallel magnetic dipoles, the magnetic dipole–dipole interaction (proportional to Γ_M) differs in sign from the electric dipole–dipole interactions (proportional to Γ_E) and have a strength related to the meta-atom separation by the factor

$$G \equiv \frac{3}{4}\hat{d} \cdot \mathbf{G}(\boldsymbol{u}, \Omega_0) \cdot \hat{d} = \frac{3}{4}\hat{\boldsymbol{m}} \cdot \mathbf{G}(\boldsymbol{u}, \Omega_0) \cdot \hat{\boldsymbol{m}}.$$
(21)

The interaction between the electric (magnetic) dipole of one meta-atom and the magnetic (electric) dipole of the other is proportional to $\overline{\Gamma}$, and is associated with the geometrical factor

$$S \equiv \frac{3}{4} \hat{\boldsymbol{d}} \cdot \boldsymbol{G}_{\times}(\boldsymbol{u}, \Omega_0) \cdot \hat{\boldsymbol{m}} .$$
⁽²²⁾

In an isolated ASR the radiative interactions between the two resonators result in eigenstates analogous to superradiant and subradiant states in a pair of atoms. In order to analyse these eigenstates, we consider the dynamics of symmetric c_+ and antisymmetric c_- modes of current oscillation (figure 2) that represent the exact eigenmodes of the ASR in the absence of

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asymmetry $\delta \omega = 0$ [5]. In terms of the individual meta-atom normal variables the symmetric and antisymmetric modes are given by

$$c_{\pm}(t) \equiv \frac{1}{\sqrt{2}} \left(b_{\rm r}(t) \pm b_{\rm l}(t) \right) \,. \tag{23}$$

Excitations of these modes possess respective net electric and magnetic dipoles and will thus be referred to electric and magnetic dipole excitations. The split ring asymmetry $\delta \omega \neq 0$, however, introduces an effective coupling between these modes in a single ASR, so that

$$\frac{\mathrm{d}c_{\pm}}{\mathrm{d}t} = \left[-\gamma_{\pm}/2 - \mathrm{i}\left(\omega_0 \pm \delta - \Omega_0\right)\right]c_{\pm} - \mathrm{i}\delta\omega c_{\mp} + F_{\pm}\,,\tag{24}$$

where γ_{\pm} and δ denote the decay rates and a frequency shift, respectively, and F_{\pm} represents the driving by the incident field. The decay rates and frequency shifts of the ASR modes, which arise from the inter-meta-atom interactions, are independent of the meta-atom asymmetry, and their exact form is given in [5].

When the spacing between meta-atoms is much less than a wavelength, the symmetric mode decays entirely due to electric dipole radiation, while the antisymmetric mode suffers decay from magnetic dipole radiation resulting in the ASR mode decay rates

$$\gamma_{+} \approx 2\Gamma_{\rm E} + \Gamma_{\rm O} \,, \tag{25}$$

$$\gamma_{-} \approx 2\Gamma_{\rm M} + \Gamma_{\rm O} \,. \tag{26}$$

Furthermore, the symmetric and antisymmetric modes are driven purely by the external electric and magnetic fields respectively, with $F_+ \propto \hat{d} \cdot E_{in}(R, t)$ and $F_- \propto \hat{m} \cdot H_{in}(R, t)$ where R is the centre of mass of the ASR.

The asymmetry $\delta \omega$ results in a coupling between the symmetric and antisymmetric modes. Figure 2 illustrates these modes as being analogous to molecular excitations [31, 32] with an unexcited ASR represented by the ground state. Consider an incident EM field whose magnetic field is perpendicular to \hat{m} so that it only drives the meta-molecule electric dipoles. In the absence of asymmetry, this incident field could only drive the symmetric mode. The coupling induced by the asymmetry, on the other hand, can allow such an incident field to additionally excite the antisymmetric mode. For example, figure 2 illustrates an incident field resonant on the antisymmetric mode (with $\Omega_0 = \omega_0 - \delta$), but which exclusively drives the symmetric mode. The asymmetry, permits a resonant excitation of the antisymmetric mode in a process analogous to a two photon atomic transition.

In the calculations of the properties of a single, isolated ASR the precise forms of the geometrical factors G and S are a result of treating the meta-atoms as point emitters. More exact expressions for these factors, which influence the coupling between meta-atoms, could be obtained by accounting for the finite spatial extent of the circuit elements [5]. Approximating each meta-atom as a point emitter has its greatest effect on the frequency shift δ between the symmetric and antisymmetric oscillation of individual meta-molecules. The frequency shift δ depends on dipole–dipole interaction energies and is therefore very sensitive ($\sim 1/u^3$) to the inter-meta-atom spacing. On the other hand, the decay rates γ_{\pm} are insensitive to the meta-atom spacing and would be unaltered if one did not approximate the meta-atoms as point emitters. As only the numerical coefficients would be affected, however, the physical behaviour described by equation (24) is unchanged in the dipole approximation.

4. Theoretical analysis of the experimentally observed transmission resonance linewidth narrowing

In practice, one does not observe excitation of a single ASR's magnetic dipole by an incident field which solely drives the electric dipole. This is because the radiative decay rate of a magnetic dipole excitation in an isolated ASR is approximately as fast as the decay rate of an electric dipole excitation. Any energy in the antisymmetric mode is therefore radiated away before it can be appreciably excited. We find that this changes dramatically when many ASRs respond cooperatively.

An incident field driving only electric dipoles can excite a high quality magnetic mode in which all ASR magnetic dipoles oscillate in phase. Excitation of this magnetic mode at the expense of other modes is responsible for the transmission resonance observed by Fedotov *et al* [4, 27]. We find that the collective decay rate of this mode decreases (the quality factor increases) with an increasing number of ASRs participating in the metamaterial. Allowing for ohmic losses, our model provides excellent agreement with the enhanced transmission resonance quality factor for increased system size observed by Fedotov *et al* [4].

To model the experimentally observed collective response [4] we study an ensemble of identical ASRs (with $u = u\hat{e}_x$ and $d = d\hat{e}_y$; figure 1) arranged in a 2D square lattice within a circle of radius r_c , with lattice spacing a, and lattice vectors $(a\hat{e}_x, a\hat{e}_y)$. The sample is illuminated by a monochromatic plane wave

$$\boldsymbol{E}_{in}^{+}(\boldsymbol{r}) = \frac{1}{2} \mathcal{E} \hat{\boldsymbol{e}}_{y} e^{i\boldsymbol{k}\cdot\boldsymbol{r}}, \qquad (27)$$

with $\mathbf{k} = k_0 \hat{\mathbf{e}}_z$, coupling to the electric dipole moments of the ASRs. In the experimentally measured transmission resonance through such a sheet [4] the number of active ASRs, N, was controlled by decoupling the ASRs with $r > r_c$ from the rest of the system with approximately circular-shaped metal masks with varying radii r_c . The resonance quality factor increased with the total number of active ASRs, saturating at about N = 700.

The electric and magnetic fields scattered from each ASR impinge on other ASRs in the metamaterial which, in turn, rescatter the fields. Multiple scattering processes result in an interaction between all the meta-atoms in the array, manifesting themselves in the dynamic coupling matrix C (16) between the normal variables. Collective modes of the metamaterial are represented by eigenvectors of C, with the *i*th eigenvector denoted by v_i. The corresponding eigenvalue λ_i gives the collective mode decay rate and resonance frequency shift

$$\gamma_i = -2\operatorname{Re}\lambda_i,\tag{28}$$

$$\delta_i = -\operatorname{Im} \lambda_i - (\omega_0 - \Omega_0), \qquad (29)$$

respectively.

We find that an incident plane-wave drives all meta-molecules uniformly and is phasematched to collective modes in which the electric and/or magnetic dipoles oscillate in phase. In the absence of a split-ring asymmetry, only modes involving oscillating electric dipoles can be driven. These modes strongly emit perpendicular to the array (into the $\pm \hat{e}_z$ directions) enhancing incident wave reflection. The magnetic dipoles, however, dominantly radiate into EM field modes within the ASR plane. This radiation may become trapped through recurrent scattering processes in the array, representing modes with suppressed emission rates and reflectance, and resulting in a transmission resonance. In order to quantify the effect, we study the radiation





Figure 3. The numerically calculated uniform magnetic mode for an ensemble of 335 ASRs in which all magnetic dipoles oscillate in phase with minimal contribution from ASR electric dipole excitations. (a) The electric dipole excitation $|c_+|^2$ and (b) the magnetic dipole excitations $|c_-|^2$ of the ASRs in the uniform magnetic mode v_m . The phase of the electric (c_+) and magnetic (c_-) dipole excitations are indicated by the colour of the surfaces in (a) and (b) respectively. The black dots indicate the positions of the ASRs in the array. This mode was calculated for a lattice spacing of $a \simeq 0.28\lambda$ and an ASR asymmetry of $\delta \omega = 0.3\Gamma$. The spacing between constituent meta-atoms in an ASR is $u = 0.125\lambda$.

properties (for $\Gamma_E = \Gamma_M$) of the numerically calculated collective magnetic eigenmode v_m of the system (figure 3) which maximizes the overlap

$$O_{\rm m}(\mathbf{b}_{\rm A}) \equiv \frac{\left|\mathbf{v}_{\rm m}^{\rm T}\mathbf{b}_{\rm A}\right|^2}{\sum_{i} \left|\mathbf{v}_{i}^{\rm T}\mathbf{b}_{\rm A}\right|^2} \tag{30}$$

with the pure magnetic excitation in which all meta-atom magnetic dipoles oscillate in phase

$$b_{A} = \sqrt{\frac{1}{2N}} \begin{pmatrix} +1 \\ -1 \\ \vdots \\ +1 \\ -1 \end{pmatrix}.$$
 (31)

The alternating signs between elements of b_A indicate that the current oscillations of the meta-atoms in an ASR oscillate antisymmetrically. We then show that the introduction of an asymmetry $\delta \omega$ in the resonances allows the excitation of v_m by the incident field. This mode closely resembles that responsible for the experimentally observed transmission resonance [4, 27].

In an infinite array, each ASR in the magnetic mode would be excited uniformly, perfectly matching the pure magnetic excitation b_A in the absence of asymmetry. This changes in a finite system where boundary effects alter the distribution of the mode v_m . This is illustrated in figure 3 which shows the numerically calculated uniform magnetic mode v_m in an ensemble of 335 ASRs with a lattice spacing of $a \simeq 0.28\lambda$ as in the experiment of Fedotov *et al* [4]. To characterize this mode, we examine the electric (symmetric) and magnetic (antisymmetric) excitations of each ASR. The state of ASR ℓ ($\ell = 1, 2, ..., N$) is described by the excitations of its constituent



Figure 4. Collective resonance narrowing. The resonance linewidth γ of the collective magnetic mode v_m in the units of an isolated meta-atom linewidth Γ as a function of the number of meta-molecules *N*; the lattice spacings $a = 1/4\lambda$ (solid line), $3/8\lambda$ (dashed line), $1/2\lambda$ (dot-dashed line), and $3/2\lambda$ (dotted line). The magenta (intermediate) dot-dashed line corresponds to an asymmetry $\delta\omega = 0.1\Gamma$, while $\delta\omega = 0$ for all other curves. The orange (lower) dot-dashed line incorporates nonradiative loss $\Gamma_0 = 0.01\Gamma$ with all other curves assuming $\Gamma_0 = 0$.

meta-atoms, $b_{2\ell-1}$ and $b_{2\ell}$. Therefore, as with a single ASR (23), the electric excitation $c_{\ell,+}$ and the magnetic excitation $c_{\ell,-}$ of ASR ℓ are given by the respective symmetric and antisymmetric combinations

$$c_{\ell,\pm} = \frac{1}{\sqrt{2}} \left(b_{2\ell-1} \pm b_{2\ell} \right) \,. \tag{32}$$

The magnetic mode excitation consists largely of the ASR magnetic dipoles oscillating in phase. In the absence of asymmetry the uniform magnetic mode in a regular array of ASRs is almost exclusively magnetic in nature [5]. Figure 3 shows that, on the other hand, an asymmetry of $\delta \omega = 0.3\Gamma$ provides a small electric dipole excitation to the magnetic mode, allowing it to be addressed by the incident field.

Figure 4 shows the dependence of the resonance linewidth γ of the collective magnetic mode v_m on the number of meta-molecules N for different lattice spacings a (λ denotes the wavelength of a resonant incident field). In the absence of ohmic losses and for sufficiently small $\delta\omega$, $\gamma \propto 1/N$ for large N when the lattice spacing $a \leq \lambda$. The split ring asymmetry only weakly affects v_m . For $\delta\omega = 0.01\Gamma$, the curves representing γ are indistinguishable from those for $\delta\omega = 0$. For the relatively large $\delta\omega = 0.1\Gamma$, however, γ is increased for N > 200. This reduction in quality factor for larger N results from the mixing of electric dipoles into the magnetic dipole mode (see figure 3), allowing the collective mode to emit in the forward and backward, $\pm \hat{e}_z$, directions. The cooperative response and linewidth narrowing sensitively depends on the lattice spacing a. For larger a (e.g. $a = 3/2\lambda$), γ becomes insensitive to N, indicating the limit of independent scattering of isolated meta-molecules and a diminished role of cooperative effects.

The collective behaviour can be understood because the ASR magnetic dipoles emit largely into the plane of the metamaterial and the bulk of the magnetic mode excitation lies in its interior.





Figure 5. The overlap $O_m(\mathbf{b}_f)$ of \mathbf{v}_m with the state \mathbf{b}_f excited by an incident field resonant on the mode \mathbf{v}_m (for $u = 1/2\lambda$, and $\Gamma_0 = 0$). For $\gamma \ll \Gamma$ there is a range of asymmetries $\delta \omega$ for which the incident field almost exclusively excites the mode \mathbf{v}_m .

Any energy radiated from magnetic dipoles would preferentially come from the ASRs near the edge of the metamaterial since radiation emitted from an ASR on the interior would more likely be re-scattered by other ASRs. On the other hand, the fraction of ASRs at the boundary varies inversely with N, leaving an ever larger proportion of the magnetic mode v_m in the interior of the array for large N. In the limit of an infinite array $(N, r_c \rightarrow \infty)$, γ would be zero in the absence of asymmetry $\delta \omega$ and ohmic losses, for $a \leq \lambda$. For a sub-wavelength lattice spacing, the only Bragg diffraction peak that the array could emit into corresponds to the forward and backward scattered fields since all other Bragg peaks would be evanescent; but both forward and backward emission from the magnetic dipoles are forbidden owing to their orientation.

An asymmetry $\delta \omega \neq 0$ generates an effective coupling between the electric and magnetic dipoles of individual ASRs in an array (see (24)). This coupling produces a slight mixing of electric dipoles into the phase matched magnetic mode v_m of the array as illustrated in figure 3. We show in figure 5 that the slight mixing of electric dipoles into v_m permits its excitation by a uniform resonant driving field propagating perpendicular to the plane of the array. We represent the steady state excitation of the array induced by this field as b_f . Figure 5 shows the relative population $O_m(b_f)$ (see (30)) of the phase-matched magnetic mode v_m . This population represents the fraction of b_f that resides in the mode v_m . We find that for $\gamma \ll \Gamma$ and $\delta \omega \gtrsim \gamma$, one can induce a state in which more than 98% of the energy is in the target mode v_m . For $\delta \omega \ll \gamma$, any excitation that ends up in v_m is radiated away before it can accumulate; the array behaves as a collection of radiating electric dipoles. For larger $\delta \omega$, the population of v_m decreases since the increased strength of the coupling between ASR electric and magnetic dipoles begins to excite other modes with nearby resonance frequencies. Although the density of modes which may be driven increases linearly with N, the corresponding reduction of γ means that a smaller $\delta \omega$ is needed to excite the target mode, and there is a range of asymmetries for which v_m is populated.

The narrowing in γ combined with the near exclusive excitation of this mode implies that for larger arrays the radiation from the sheet is suppressed and hence the transmission is enhanced as seen by Fedotov *et al* [4]. In figure 6 we compare



Figure 6. Comparison between experimentally measured transmission resonance quality factors Q/Q_0 (stars) from [4], where $Q_0 \simeq 4.5$ denotes the single meta-atom quality factor (for $\lambda \simeq 2.7$ cm), and numerically calculated resonance linewidth γ of the collective magnetic mode v_m with $\Gamma_0 \simeq 0.14\Gamma$ (solid line) and $\Gamma_0 = 0$ (dashed line). Here $\delta \omega \simeq 0.3\Gamma$, and $a \simeq 0.28\lambda$.

the experimentally observed transmission resonance [4] to our numerics. We use the experimental spacing $a \simeq 0.28\lambda$. The numerical values for the asymmetry $\delta \omega \simeq 0.3\Gamma$ and the ratio between electric and magnetic spontaneous emission rates $\Gamma_E/\Gamma_M \simeq$ 1 were estimated from the relative sizes of the ASR meta-atoms [27] and their relationship to the resonant wavelength in the experiments by Fedotov et al [4]. The spacing between the meta-atoms $u \simeq 0.125\lambda$ was chosen so that the resonance frequencies of the mode v_m and the collective mode in which all electric dipoles oscillate in phase are shifted by less than Γ with respect to one another so as to be consistent with experimental observations in [4]. For a given nonzero Γ_0 , one can obtain the collective decay rate of the magnetic mode by adding Γ_0 to the decay rate calculated in the absence of ohmic losses. We therefore fit the shape of the numerically calculated curve of the resonance linewidth as a function of the number of resonators to the experimental observations of Fedotov et al [4] using Γ_0 as a fitting parameter. The ohmic loss rate $\Gamma_0 \simeq 0.14\Gamma$ produces the expected saturation of quality factor for large N. The vertical shift of the experimental data set is determined by the single meta-atom quality factor $Q_0 \equiv \omega_0 / \Gamma$, and the best value $Q_0 \simeq 4.5$ is roughly consistent with full numerical solutions to Maxwell's equations for scattering from a single ASR presented by Papasimakis et al [11]. The excellent agreement of our simplified model that only includes dipole radiation contributions from each meta-atom can be understood by a notably weaker quadrupole than dipolar radiation field from an ASR [30].

The result also confirms the importance of the uniform magnetic mode v_m on the observed transmission resonance. The observed saturation is due to a combination of a fixed $\delta \omega$, which in larger arrays leads to the population of several other modes in addition to v_m , and ohmic losses in the resonators which set an ultimate limit to the narrowing of γ . If ohmic losses were to be reduced, the quality factor of the resonance would saturate at a correspondingly higher value, as shown in figure 6. In the displayed case the resonance linewidth narrowing is limited by the relatively large asymmetry $\delta \omega \simeq 0.3\Gamma$. For larger arrays, figure 4 indicates that one could further

enhance the quality factor by reducing the asymmetry $\delta\omega$. So long as $\delta\omega$ sufficiently exceeds the magnetic mode's decay rate γ , a large, and potentially greater, fraction of the metamaterial excitation would reside in the magnetic mode v_m as shown in figure 5. The reduced excitation of other collective modes could then further enhance the quality of the observed transmission resonance. A reduction or elimination of ohmic losses would therefore dramatically increase the resonance's quality factor in larger arrays.

5. Conclusion

In conclusion, we analysed the recent observations of transmission spectra in a metamaterial array of ASRs. We showed that the system can exhibit a strong cooperative response in the case of sufficiently closely-spaced resonators. Moreover, we demonstrated how an asymmetry in the split rings leads to excitation of collective magnetic modes by a field which does not couple directly to ASR magnetic moments. The excitation of this uniform phase-coherent mode results in a cooperative response exhibiting a dramatic resonance linewidth narrowing, explaining the experimental findings [4].

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