Collective electromagnetic response of discrete metamaterial systems

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We develop a general formalism to describe the interactions between a discrete set of plasmonic resonators mediated by the electromagnetic field. The resulting system of equations for closely-spaced meta-atoms represents a cooperative metamaterial response and we find that collective interactions between asymmetric split-ring resonators arranged in a 2D lattice explains the recent experimental observations of system-size dependent narrowing of the transmission resonance linewidth. We further show that this cooperative narrowing depends sensitively on the lattice spacing.

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Resonant multiple scattering plays an important role in mesoscopic wave phenomena that can also be reached with electromagnetic (EM) fields. In the strong scattering regime, interference of different scattering paths between discrete scatterers can result in, e.g., light localization—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, there has been a rapidly increasing interest in fabrication and theoretical modeling of such systems. The role of strong scattering and cooperative effects between discrete resonators in metamaterial arrays are largely unknown, as such phenomena cannot be easily captured by commonly employed theoretical techniques based on the homogenization approximation of the scattering medium or full solutions of Maxwell’s equations in small, computationally accessible, systems.

In this letter we develop a theoretical formalism of collective interactions between plasmonic resonators, or meta-atoms, mediated by EM field. In a computationally efficient model, which also provides physical insight into the interaction processes, 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 field then determine the collective interactions within the ensemble, resulting in collective resonance frequencies, linewidths, and line shifts. The resulting set of equations can represent a strong cooperative response in the case of sufficiently closely-spaced resonators, necessitating the inclusion of interference effects in multiple scattering between the meta-atoms. As a specific example, we study asymmetric split-ring (ASR) resonators, each 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. In a single ASR we derive eigenstates analogous to superradiant and subradiant states in a pair of atoms and in an ensemble of ASRs we demonstrate how strong collective interactions in a discrete metamaterial array are responsible for recent experimental observations of a dramatic narrowing of the transmission resonance linewidth (increasing quality factor) with the number of resonators. Our analysis indicates the necessity of accounting for the strong collective response of metamaterial systems in understanding the dynamics and design of novel meta-materials. 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 and disorder-related phenomena.

We consider an ensemble of N metamaterial unit elements, metamolecules, each formed by n discrete meta-atoms, such that the position of the meta-atom j is denoted by \( \mathbf{r}_j \) \((j = 1, \ldots, n * N)\). This ensemble is driven by an external beam with electric field \( \mathbf{E}_{\text{in}}(\mathbf{r}, t) \) and magnetic field \( \mathbf{B}_{\text{in}}(\mathbf{r}, t) \) whose frequency components have wavelengths much larger than the spatial extent of the meta-atoms. We assume that meta-atom j supports a single eigenmode of current oscillation whose state can be described by a single dynamic variable \( Q_j(t) \) with units of charge and whose spatial profile is described by time-independent functions \( \mathbf{p}_j(\mathbf{r}) \) and \( \mathbf{w}_j(\mathbf{r}) \). These mode functions are defined such that the polarization and magnetization densities associated with atom j are
\[
\mathbf{P}_j(\mathbf{r}, t) = Q_j(t)\mathbf{p}_j(\mathbf{r}) \quad \text{and} \quad \mathbf{M}_j(\mathbf{r}, t) = I_j(t)\mathbf{w}_j(\mathbf{r})
\]
respectively, where \( I_j(t) = dQ_j/dt \) is the current. We consider the Power-Zienau-Woolley Lagrangian in the Coulomb gauge for the coupled system of the EM field and matter (described by an arbitrary dynamic polarization and magnetization densities)

\[
\mathcal{L} = \mathcal{K} + \mathcal{L}_{\text{em}} + V_{\text{coul}} + \sum_j \left[ Q_j(t) \mathcal{E}_j + I_j(t) \Phi_j \right], \quad (1)
\]

where \( \mathcal{L}_{\text{em}} \) is the Lagrangian for the free EM field, \( V_{\text{coul}} \) is the instantaneous Coulomb interaction, \( \mathcal{E}_j(t) \equiv \int d^3r \mathbf{E}(\mathbf{r}, t) \cdot \mathbf{p}_j(\mathbf{r}) \) is an electromotive force on the circuit \( j \) and \( \Phi_j(t) \equiv \int d^3r \mathbf{B}(\mathbf{r}, t) \cdot \mathbf{w}_j(\mathbf{r}) \) is an effective field flux through circuit \( j \), \( \mathcal{K} = \sum_j U_j^2/2 \), and \( l \) is a phenomeno-
logical constant representing an inertial inductance which is present in the absence of any EM field interactions. This inertial inductance may result, e.g., from the effective mass of the charge carriers or surface plasmons as they move through the circuit.

We now derive a set of coupled equations governing the excitation of the current oscillations by a near resonant field. Following the procedure in Ref. [6], we find the system Hamiltonian in terms of the dynamic variables \(Q_j\) and their conjugate momenta \(\dot{q}_j = I_j + \Phi_j\) and arrive at the equations of motion \(\ddot{Q}_j(t) = (\dot{q}_j(t) - \Phi_j(t))/l\) and \(\dot{\Phi}_j(t) = E_j(t)\). In order to integrate the scattered fields we express the positive frequency components of the electric displacement and magnetic induction in terms of the normal modes \(a_q\), \(D^+(r) = \sum_q \hat{e}_q \xi_q e^{iqr} r\) and \(B^+(r) = \sqrt{\mu_0/\varepsilon_0} \sum_q \hat{k} \times \hat{e}_q \xi_q e^{iqr} r\), where \(D(r) = D^+(r) + D^-(r), (D^+)^* = D^-,\) and \(q\) refers to both wavevector \(q\) and the transverse polarization \(E_q\). We obtain the equations of motion for \(a_q\) from Eq. (1). The procedure resembles derivation for the expressions of the normal modes \(E, B\) and self-inductance \(L\) and their conjugate momenta \(\phi\) respectively, but due to the long-range interactions mediated by the circuit current mode. Calculation of \(C_j\) and \(L_j\) is analogous to the derivation of the radiative linewidth of an atom and the values of \(C_j\) and \(L_j\) depend on the geometric structure of the mode functions \(p_j\) and \(w_j\).

Consequently, an isolated meta-atom exhibits a resonance frequency \(\omega_j = 1/\sqrt{L_j C_j}\) and the charge oscillation results in radiative damping rates for electric and magnetic dipoles, \(\Gamma_{E,j}(k) = h^2 \omega_j C_j k^3/(6\pi\varepsilon_0)\) and \(\Gamma_{M,j}(k) = \mu_0 \omega_j A_j k^3/(6\pi L_j)\), respectively. We further simplify by neglecting the frequency dependence of \(\Gamma_{E/M,j}\) (the Markov approximation), the inertial inductance (taking the limit \(l/L_j \to 0\)), and assume \(\Gamma_{M,j} \ll \omega_j\), so that a single meta-atom behaves as a simple LC circuit with resonance frequency \(\omega_j\) and a radiative dissipation rate \(\Gamma_{E,j} + \Gamma_{M,j}\). In addition, meta-atoms exhibit nonradiative decay \(\Gamma_O\), e.g., due to ohmic losses.

By including the inter-circuit interactions mediated by the field we obtain the coupled equations of motion for the vector of dynamic variables \(Q = (Q_1, Q_2 , \ldots, Q_{n-N})^T\) and their conjugate momenta \(\phi = (\phi_1, \phi_2, \ldots, \phi_{n+n})^T\)

\[
-\Omega Q = C^T \left[ (\omega - i\Gamma_M) - 3\omega^2 \mathbf{G}_M \omega^{-2}/2 \right] \mathbf{L}^{-1} \phi
+ 3 \mathbf{G}_X \Omega^2 \mathbf{Q}^2 - \mathbf{L}^{-1} \mathbf{Q}_m
(\text{6a})
\]

\[
i\Omega \phi = L^T \left[ (\omega - i\Gamma_E) - 3\omega^2 \mathbf{G}_E \omega^{-2}/2 \right] \mathbf{C}^{-1} \mathbf{Q}
- 3 \mathbf{G}_X \phi^2 - \mathbf{E}_m
(\text{6b})
\]

where we defined diagonal matrices \(C_{jj,j'} = C_j \delta_{jj',} L_{jj,j'} = L_j \delta_{jj'}, \omega_{jj,j'} = \omega_j \delta_{jj'}, \Gamma_{E,j,j'} = \Gamma_{E,j}(k) \delta_{jj'},\) and \(\Gamma_{M,j,j'} = \Gamma_{M,j}(k) \delta_{jj'}\). The matrix \(L = L^+ [1 + i\Omega \omega - (3/2) \omega^2 \mathbf{G}_M \omega^{-2}/2] \mathbf{L}^+\) appearing in Eq. (6a) is the inductance matrix. The driving terms are given in terms of the incident fields as \(\mathbf{E}_{i,m} = h_j \mathbf{E}_{in}(r_j, \Omega) \cdot \hat{d}_j\) and \(\mathbf{F}_{i,m} = A_j \mathbf{B}_{in}(r_j, \Omega) \cdot \hat{m}_j\). The radiative interactions between the scatterers are described by \(\mathbf{G}_{E,M,X}\). These have vanishing diagonal elements and off-diagonal elements \((j \neq j')\) given by

\[
[\mathbf{G}_{E}]_{jj,j'} = \hat{d}_j \cdot \sqrt{\mathbf{G}}(r_j - r_{j'}, k) \sqrt{\mathbf{E}} \cdot \hat{d}_{j'}
(\text{7})
\]

\[
[\mathbf{G}_{M}]_{jj,j'} = \hat{m}_j \cdot \sqrt{\mathbf{G}}(r_j - r_{j'}, k) \sqrt{\mathbf{M}} \cdot \hat{m}_{j'}
(\text{8})
\]

\[
[\mathbf{G}_X]_{jj,j'} = \hat{d}_j \cdot \sqrt{\mathbf{G}_X}(r_j - r_{j'}, k) \sqrt{\mathbf{M}} \cdot \hat{m}_{j'}
(\text{9})
\]

The first lines of Eqs. (6a,b) are analogous to frequency dependent inductance and capacitance matrices, respectively, but due to the long-range interactions mediated
by the radiation field, these can substantially differ from the quasi-static expressions for which \( G_s \) is also absent.

We consider an incident EM field with a dominant frequency \( \Omega \) and bandwidth \( \delta \Omega \), and introduce the normal variables for the meta-atoms whose resonance frequencies are centered around \( \omega_0 \)

\[
b_j(t) = \frac{e^{i\omega t}}{2\omega_j} \left( \frac{Q_j}{\sqrt{C_j}} + i \frac{\phi_j}{\sqrt{L_j}} \right).
\]  

(10)

When the EM field and the meta-atoms themselves have frequency components in a narrow bandwidth about \( \omega_0 \), such that \( \delta \Omega, \Gamma_E/M/\delta_j, |\omega_j - \omega_0| \ll \omega_0 \), we may further simplify by substituting \( \omega_0/c \) for \( k \) in the expressions for \( \Gamma_E/M \) and \( G_{E/M} \). Then, from Eqs. (6) and (10), we arrive at coupled equations for \( b_j \)’s. We make the rotating wave approximation, in which we neglect terms oscillating at high frequencies \( \sim 2\omega_0 \), and obtain the following closed system for the current oscillation dynamics

\[
\dot{b}(t) = Cb(t) + F(t),
\]  

(11)

where the coupling matrix and the driving are given by

\[
C = -i\delta - \frac{\Gamma}{2} + \frac{3}{4} \left( iG_E + iG_M + G_x + G_x^T \right),
\]  

(12)

\[
F = i\frac{e^{i\omega t}}{\sqrt{2\omega}} \left( \frac{1}{\sqrt{L}}\epsilon_{in}(t) + i \frac{1}{\sqrt{C}} e^{-i\Phi_{in}(t)} \right).
\]  

(13)

Here the detuning \( \delta \equiv \omega - \omega_0 \) and \( \Gamma \equiv \Gamma_E + \Gamma_M + \Gamma_O \), where we phenomenologically account for ohmic losses through \( \Gamma_O \). In the limits we’ve considered, there exist as many distinct collective eigenmodes of oscillation as there are meta-atoms in the system. Each collective mode corresponds to an eigenvector of the matrix \( C \) and has a distinct resonance frequency and decay rate, determined by the imaginary and real parts of the corresponding eigenvalue. Eqs. (11, 13) resemble the set of equations describing a cooperative response of atomic gases to resonant light in which case the coupling is due to electric dipole radiation alone [9, 10].

The crucial component of the strong cooperative response of closely-spaced scatterers are recurrent scattering events [9, 11]—in which a wave is scattered more than once by the same dipole. Such processes cannot generally be modeled 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 Ref. [12] where the translational symmetry of an infinite lattice simplifies the response.

We next apply the formalism to an array of ASRs. A single ASR (metamolecule) consists of two separate coplanar circular arcs (meta-atoms), labeled by \( j \in \{l, r \} \) and separated by \( a \equiv r - r_l \). The current oscillations in each meta-atom produce electric dipoles with orientation \( \hat{d} (\hat{d} \perp \hat{a}) \) and magnetic dipoles with opposite orientations \( \hat{m}_r = -\hat{m}_l (\hat{m}_r \perp \hat{a}, \hat{d}) \). Each element of the ASR possesses decay rates \( \Gamma_{E/M/O,j} = \Gamma_{E/M/O} \). An asymmetry between the rings, e.g., resulting from a difference in arc length, manifests itself as a difference in resonance frequencies with \( \omega_r = \omega_0 + \delta \omega \) and \( \omega_l = \omega_0 - \delta \omega \). The interaction matrix \( C \) for a single ASR is thus given by

\[
\begin{pmatrix}
-\frac{i\delta \omega - \Gamma/2}{2i\Gamma G/4} & 3i\Gamma G/4 - 3\Gamma S/4 & \ldots \\
\frac{3i\Gamma G/4 - 3\Gamma S/4}{2i\Gamma G/4} & -\frac{i\delta \omega - \Gamma/2}{2i\Gamma G/4} & \ldots \\
& & \ddots
\end{pmatrix}
\]  

(14)

where \( \Gamma \equiv \sqrt{\Gamma_E M/O}, \Gamma_E \equiv \Gamma_E - \Gamma_M, G \equiv \hat{d} \cdot G(a,k) \cdot \hat{d} \), and \( S \equiv \hat{d} \cdot G_x(a,k) \cdot \hat{m} \) (which is here assumed to be real). To analyze the collective modes of the ASR, we consider the dynamics of symmetric \( c_+ \) and antisymmetric \( c_- \) modes of oscillation defined by \( c_\pm \equiv (b_r \pm b_l)/\sqrt{2} \) that represent the eigemodes of the ASR in the absence of asymmetry \( \delta \omega = 0 \). From Eqs. (12) and (13), one finds \( \dot{c}_\pm = \gamma_\pm c_\pm - i\omega c_\pm + (\Gamma_s \pm F_0) c_\pm \), where the decay rates \( \gamma_{\pm} = \Gamma_M [1 \pm 3 \text{Im}(G)/2] + \Gamma_M [1 \mp 3 \text{Im}(G)/2] + \Gamma \text{Re}(S) + \Gamma_O \), and the frequency shift \( \Delta = -3 \text{Re}(G) \Gamma/2 - 3 \text{Im}(S)/4 \). 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 \( \delta \omega \neq 0 \) introduces an EM coupling between these modes: the incident field with \( E_{in} || \hat{d} \) and \( B_{in} \perp \hat{m} \) only excites the electric mode when \( \delta \omega = 0 \), but for \( \delta \omega \neq 0 \) it can resonantly pump the anti-symmetric magnetic mode.

As an example of a collective response of a metamaterial array, we study an ensemble of identical ASRs (metamolecules) with \( a = \alpha \hat{e}_x \) and \( d = d \hat{e}_y \) arranged in a 2D square lattice within a circle of radius \( r_c \) and with lattice spacing \( u \) and lattice vectors \( \mathbf{u}_1 = u \hat{e}_x \) and \( \mathbf{u}_2 = u \hat{e}_y \). The sample is illuminated by a cw plane wave \( E_{in}^x = \frac{1}{2} C \hat{e}_y \hat{e}^{ik\cdot r} \) with \( k = \beta \hat{e}_z \), coupling to the electric dipole moments of the ASRs. A transmission resonance through such a sheet was experimentally measured in Ref. [3] where the number of active ASRs \( N \) was controlled by decoupling the ASRs with \( r \gtrsim r_c \) 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 incident field drives all metamolecules uniformly, and therefore is phase-matched 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 into the \( \pm \hat{e}_z \) directions enhancing incident wave reflection. The magnetic dipoles, on the other hand, 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, resulting in a transmission resonance. Consequently, we study the radiation properties of the magnetic eigenmode \( \nu_m \) of \( C \) which maximizes the
overlap $O_m(b_A) \equiv |v_m^T b_A|^2 / \sum_i |v_i^T b_A|^2$ with the pure magnetic excitation $b_A = (1, -1, \ldots, 1, -1)^T / \sqrt{2N}$. We then show that the introduction of an asymmetry allows the excitation of $v_m$ by the incident field. This mode closely resembles that responsible for the experimentally observed transmission resonance $[3, 13]$.

Figure 1(b) shows dependence of the resonance linewidth $\gamma$ of $v_m$ on the number of metamolecules $N$ for different lattice spacings $u$. In the absence of ohmic losses and for sufficiently small $u$ and $\delta \omega$, $\gamma \propto 1/N$ for large $N$. The split ring asymmetry only weakly affects $v_m$. For $\delta \omega = 0.01 \Gamma$, the curves representing $\gamma$ are indistinguishable from those for $\delta \omega = 0$. For the relatively large $\delta \omega = 0.1 \Gamma$, however, $\gamma$ is increased for $N \gtrsim 200$. The cooperative response is very sensitive to $u$, resulting in different linewidth narrowing. For a larger $u = 3/2\lambda$, however, $\gamma$ is insensitive to $N$ indicating the limit of independent scattering of isolated metamolecules and a diminished role of cooperative effects.

As with an isolated ASR, an asymmetry $\delta \omega \neq 0$ generates an effective two-photon coupling between collective electric and magnetic modes. This is analogous to electromagnetically induced transparency: one leg of the transition is provided by the coupling of the incident field to collective electric dipole excitations, while the other is provided by the effective coupling introduced by the asymmetry. We illustrate this in Fig. 1(b), showing the relative population $O_m(b_f)$ of the collective magnetic mode $v_m$ produced by a resonant field as a function of $\gamma$ and $\delta \omega$, where $b_f$ is the state induced by the uniform driving resonant on $v_m$. One sees that for $\gamma < \Gamma$ and $\delta \omega \gtrsim \gamma$, one can excite a state in which more than 98% of the energy is in the target mode $v_m$. For $\delta \omega < \gamma$, any excitation that ends up in $v_m$ is radiated away before it can accumulate; the array behaves as a collection of symmetric metamolecules. For larger $\delta \omega$, the population of $v_m$ decreases since the increased strength of the effective two-photon transition begins to excite other modes with nearby resonance frequencies. Although the density of modes which may be excited increases linearly with $N$, the corresponding reduction of $\gamma$ 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 excited. The narrowing in $\gamma$ 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 enhanced as in Ref. $[3]$. The observed saturation $[3]$, however, may result from a combination of a fixed $\delta \omega$, which leads to the population of other modes for larger arrays, and ohmic losses in the resonators which put a floor on the collective decay rate $\gamma$. Fig. 1(b), shows that an ohmic loss rate of $\Gamma/100$ results in the expected saturation of quality factor with $N$.

In conclusion, we developed a computationally efficient formalism describing the collective interactions between discrete resonators. In principle, one can calculate the EM response also by having knowledge of the material comprising the circuit elements and numerically solving Maxwell’s equations with a numerical mesh small enough to resolve the features of each meta-atom. This, however, becomes computationally intractable when the system contains more than a few resonators. In ASRs we showed how an asymmetry leads to excitation of collective magnetic modes by a field which does not couple directly to ASR magnetic moments and results in cooperative response exhibiting a dramatic resonance linewidth narrowing, consistently with experimental findings $[3]$.

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