HERTHA-SPONER-PREIS

Coupling games in metamaterials

How to design metamaterial structures for desired optical properties and resonant behavior.

Na Liu

Metamaterials have become one of the hottest fields of photonics since the pioneering work of John Pendry on negative refractive index, invisibility cloaking, and perfect lensing. For practical applications, three-dimensional metamaterials are required. Here, coupling effects between individual constituents play a dominant role for the optical and electronic properties. Metamaterials can offer both electric and magnetic responses at optical frequencies. Hence, electric as well as magnetic dipolar and higher-order multipolar coupling are the essential mechanisms. The intricate interplay between different coupling effects in a plasmon hybridization picture provides a hands-on tool to intuitively understand the evolution from molecule-like states to solid-state-like bands.

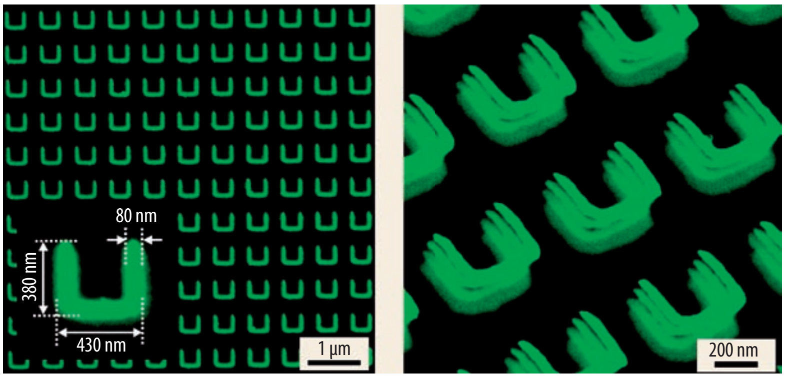

Fig. 1 Four-layer split-ring resonator (SRR) metamaterial. The individual nanostructures consist of 20 nm thick gold split rings with around 400 nm lateral size. The layers are stacked at a distance of 50 nm.

Metamaterials are artificial materials with designed electromagnetic functionality and sizes much smaller than the operating wavelength of light [1]. Metamaterials often consist of metallic nanostructures which allow tailoring of their optical properties [2]. Incident light can excite coherent oscillations of free conduction electrons which lead to localized particle plasmon resonances. The resonance frequencies depend on the size, on the shape, on the dielectric function of the metal, as well as on the dielectric function of the surrounding environment. The shape of the individual metamaterial constituents can vary substantially: from simple spheres or ellipsoids to wires, split-ring resonators (SRRs), meshes, or meanders. Metamaterials can offer new properties that natural materials do not have. For example, a negative magnetic permeability can be engineered, and together with negative electric permittivity, metamaterials can exhibit a negative refractive index [3]. These astounding properties are well suited for novel devices such as superlenses and hyperlenses which beat the diffraction limit, or optical cloaks that can render objects invisible.

For practical applications of metamaterials, often three-dimensional [4] or even bulk structures are needed. Due to the fact that the size of the metamaterial constituents and hence the unit cells are much smaller than the wavelength of light, the lateral as well as vertical finite spacing will inherently lead to strong interaction between the neighboring metamaterial elements. An example of such a complex metamaterial is a four-layer split-ring resonator (**Fig. 1**). Consequentially, the optical properties can be changed substantially compared to those of an individual metamaterial element. This is in analogy to solid state physics, where the electronic properties of solids can dramatically vary from those of individual atoms. Take for example carbon, where the optical spectra of individual atoms differ strongly from those of graphite or diamond. This also illustrates that the arrangement of the unit cells in the lattice of the solid is crucial for the resulting properties.

Theoretical Basics of Dipole-dipole Coupling

In a simple quasistatic picture, when coupling either electric or magnetic dipoles together, it is quite straightforward to derive the interaction energy. For simple geometries, it is sufficient to consider longitu-

IN BRIEF

- Gold nanowires act as very simple plasmonic structures. A dimer of such nanowires can act as an artificial magnetic "atom".
- A split-ring resonator (SRR) allows an extra degree of freedom, namely the inclusion of a magnetic response.
- In doing so, SRRs can be used to construct materials with negative permeability or even negative refractive index when combined with continuous wire media.
- Stacked systems have another degree of freedom. Thus, in these rather complex structures a number of coupling mechanisms are present – even higher-order multipoles – that can play a dominant role in the optical spectra and even lead to transparency.

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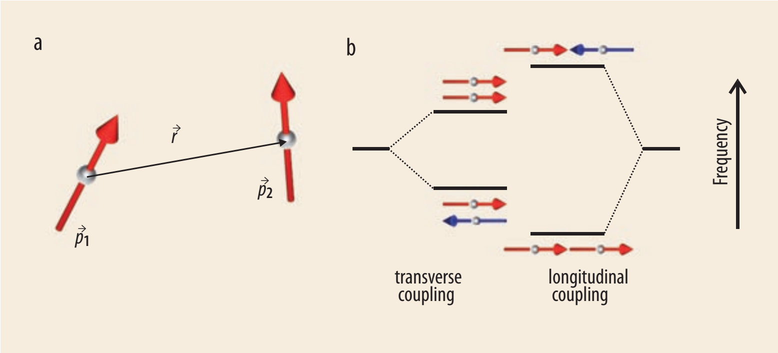

Fig. 2 The interaction between two dipoles $\vec{p}_1$ and $\vec{p}_2$ with $\vec{r}$ being the center-to-center vector (a) leads to the level schemes of two coupled dipoles (b). Left: transverse coupling. Right: longitudinal coupling.

dinal or transverse interaction. Here we will limit ourselves in a first approximation to only the dipole-dipole interaction, although in metamaterials higher-order multipoles can play a substantial role. If two dipoles with moments $\vec{p}_1$ and $\vec{p}_2$ (either electric or magnetic) interact at center-to-center distance r (**Fig. 2**), the quasi-static interaction energy V is given by

$$V = \frac{1}{4\pi\varepsilon_0}\left(\frac{\vec{p}_1\cdot\vec{p}_2}{r^3} - \frac{3(\vec{p}_1\cdot\vec{r})\,(\vec{p}_2\cdot\vec{r})}{r^5}\right)$$

$$= \frac{\vec{p}_1\cdot\vec{p}_2 - 3(\vec{p}_1\cdot\hat{r})(\vec{p}_2\cdot\hat{r})}{4\pi\varepsilon_0 r^3} \qquad (1)$$

with $\hat{r}$ as the unit vector from $\vec{p}_1$ and $\vec{p}_2$.

This reduces for purely transverse (side-by-side alignment) or longitudinal (end-to-end alignment) coupling of the two dipoles to:

$$V = \gamma\frac{p_1\cdot p_2}{\pi\varepsilon_0 r^3} \qquad (2)$$

with γ as the interaction index, which is +1 for transverse coupling and –2 for longitudinal coupling. p_1 and p_2 are the magnitudes of the dipole moments.

In the case of transverse coupling—for example two electric dipoles—laterally coupled dipoles in the antisymmetric mode attract each other, hence decreasing the restoring force and leading to a low-frequency resonance. In contrast, in the symmetric mode, the two dipoles are repulsive, giving rise to enhanced restoring force and leading to the higher frequency resonance (**Fig. 2b, left**). The opposite holds true for the longitudinally coupled dipoles: there, a symmetric arrangement leads to attraction of opposite charges and it therefore decreases the restoring force and constitutes the low-frequency mode. The high-frequency mode, on the other hand, is made up by the antisymmetric arrangement of the two dipoles, in which the restoring force is increased due to the repulsion of charges with the same sign. Similarly, for transverse (longitudinal) coupling of two magnetic dipoles, the repulsion and attraction of the two north and south poles will lead to an enhanced (reduced) magnetic interaction and therefore a higher (lower) resonance frequency (**Fig. 2b, right**).

Artificial Metamaterial "Atoms"

Let us start with the optical properties of a single metallic nanoparticle, for example a gold nanowire with a size of 500 × 150 × 20 nm^3. It resides on a glass substrate and is illuminated by light which is linearly polarized along the longer axis of the nanowire at normal incidence. There is a resonance at around 160 THz (~1870 nm or 5340 cm^{-1}) in the spectrum (**Fig. 3a**). Associated with it is the excitation of an electric dipole moment in the nanowire, which extends from the positive to the negative charges (**Fig. 3b**). All the simulated spectra and field calculations in this paper were performed by using the software package CST Microwave Studio.

We now consider two gold nanowires with a finite separation. Due to close proximity, the two are strongly coupled. The interaction can lift the degeneracy of the bare plasmonic mode of the individual nanowires and leads to two new modes due to plasmon hybridization – one with a symmetric alignment of the two electric dipoles and one with an antisymmetric alignment. This plasmon hybridization picture, which was introduced by Peter Nordlander, demonstrates a compelling analogy between plasmon resonances of metallic nanoparticles and wavefunctions of simple atoms and molecules

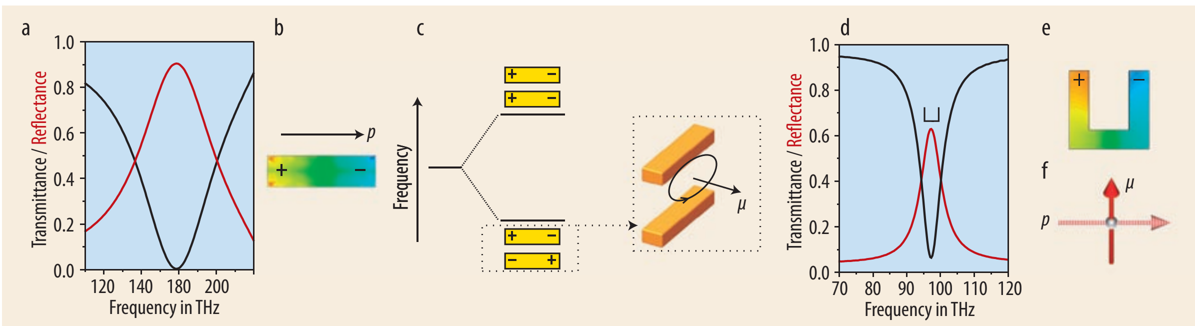

Fig. 3 (a) Simulated transmittance and reflectance spectra of a gold nanowire structure. The normally incident light is polarized along the longer axis of the nanowire. (b) The electric field distribution at the resonance. Positive (red) and negative (blue) charges are excited at the ends of the nanowire, which corresponds to the excitation of an electric dipole moment p. (c) Transverse coupling of two coupled metallic nanowires. A magnetic dipole moment μ can be excited in the antisymmetric mode, acting as a magnetic "atom". (d) Simulated spectra of a gold SRR on a glass substrate. The normally incident light is polarized along the SRR's gap-bearing side. The symbol represents the currents in the SRR at the resonance. (e) The electric field distribution at the resonance. Electric dipole-like plasmons are excited along the entire SRR, giving rise to a magnetic dipole moment perpendicular to the SRR plane as shown in (f).

[5]. The assignment as to which alignment represents the lower-frequency resonance and which one represents the higher-frequency resonance depends crucially on how the two nanowires are arranged with respect to one another as already discussed.

In particular, the antisymmetric mode of the two nanowires in the transverse configuration is also termed "magnetic resonance". The reason for this terminology is as follows: the antisymmetric currents in the two wires together with the displacement currents between the two wires can lead to a resonant excitation of the magnetic dipole moment (**Fig. 3c**), giving rise to a magnetic response in the system. Therefore, the structure of two nanowires in a stacked fashion can act as a magnetic "atom", a fundamental building block of metamaterials. As we will show later, stacked metamaterials can be constructed by combining several layers of these "atoms" and take their coupling into account [6].

The split-ring resonator structure (SRR) is another fundamental building block of metamaterials. It has been widely utilized for constructing materials with negative permeability or even negative refractive index when combined with continuous wire media. When linearly polarized light is incident along the gap-bearing side of an SRR at normal incidence, electric dipole-like plasmons can be excited in the entire structure (**Fig. 3d**), giving rise to a magnetic dipole moment perpendicular to its plane (**Fig. 3e**). When arranging two such "atoms" in different configurations, for example next to or above each other, similar coupling rules as in the case of the electric dipoles apply to the magnetic dipoles. However, the coupling behavior is more complex than the case of two coupled metallic nanoparticles due to the fact that both the electric as well as the magnetic coupling should be taken into account. Also, it is not clear ab-initio which coupling mechanism is dominant. As we will show below, it is possible to reduce or even switch off the electric dipolar coupling and retain only the magnetic coupling, which in turn simplifies the understanding of the coupling mechanisms of rather complex metamaterial systems.

Lateral split-ring resonator coupling

In this section, we investigate planar SRR dimers, which consist of laterally coupled SRR pairs with a certain rotation angle. Let us first consider a side-by-side configuration with 0° angle between the two (**Fig. 4a**). The polarization of the normally incident light is along the gap-bearing side of the SRRs. In this case, the light excites circulating currents, which correspond to the excitation of two electric dipoles oriented along the direction of the gaps. The two resulting magnetic dipoles are perpendicular to the plane of the SRRs. As a result, the two electric dipoles are longitudinally coupled whereas the two magnetic dipoles are transversely coupled (**Fig. 4c**). As there is no phase retardation between the two SRRs, they are excited symmetrically. Hence, the spectrum of the 0° rotated pair shows only a single resonance (**Fig. 4b**). At resonance, the two electric dipoles are aligned parallel and so are the two magnetic dipoles. Tilted incidence would cause symmetry breaking and introduce a certain lateral phase shift between the two SRRs, making the antisymmetric mode weakly observable.

Similarly, for a 180° rotated SRR pair (**Fig. 4d**), due to the lack of phase retardation, there is also only a single resonance observable in the spectrum (**Fig. 4e**). At resonance, the two electric dipoles are aligned parallel whereas the magnetic dipoles are aligned antiparallel resulting from the 180° rotation of the right SRR (**Fig. 4f**).

The situation becomes intriguing for a 90° rotated pair (**Fig. 4g**). Because of its orientation with respect to the incident polarization, circulating currents in the right SRR cannot be directly excited by the external light. This introduces phase retardation between the two. The external light couples to the left one. On resonance, its excitation is transferred to the right one by inductive coupling owing to the mutual inductance between the two elements [7, 8]. Due to the fact that the excited electric dipoles are perpendicular to each other, the electric dipole-dipole interaction is zero in a first approximation. As a result, the coupling between the two magnetic dipoles plays a key role and it leads to the spectral splitting (**Fig. 4h**). In analogy to the states of two simple atoms hybridized into molecular orbitals, we term the resulting coupled system "SRR molecule",

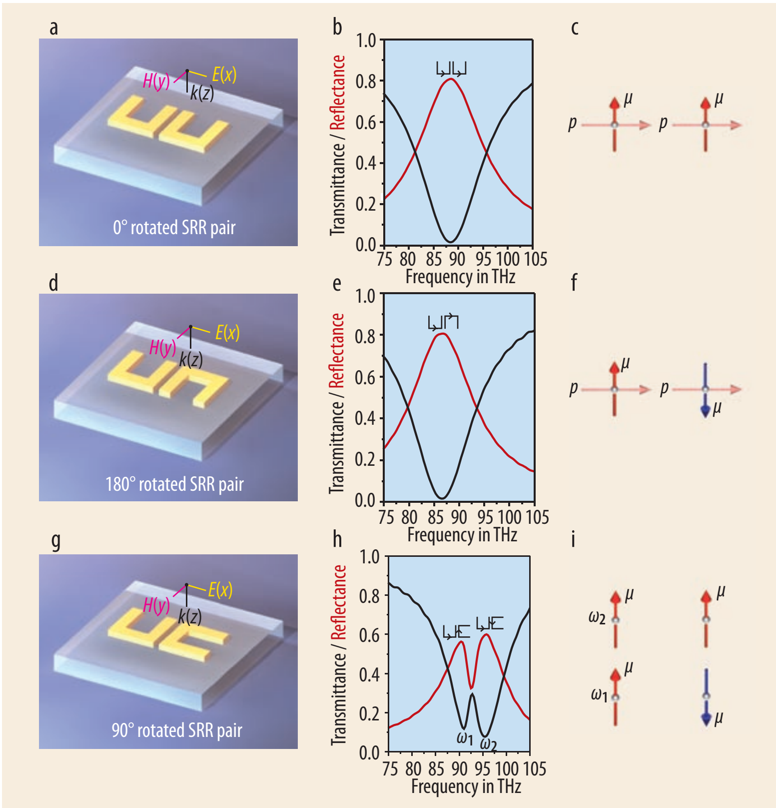

Fig. 4 Schematic of the differently rotated SRR pairs on a glass substrate. The size of each is like that in **Fig. 1**. The separation between them is 50 nm. The normally incident light is polarized along the x-direction (a, d, g). Simulated transmittance and reflectance spectra of the respective SRR pairs. The symbols represent the currents in the SRRs at the resonance (b, e, h). Schematic diagram of the alignment of the magnetic dipoles in the two SRRs at the respective resonances. Solid and dashed arrows represent magnetic and electric dipole moments (c, f, i).

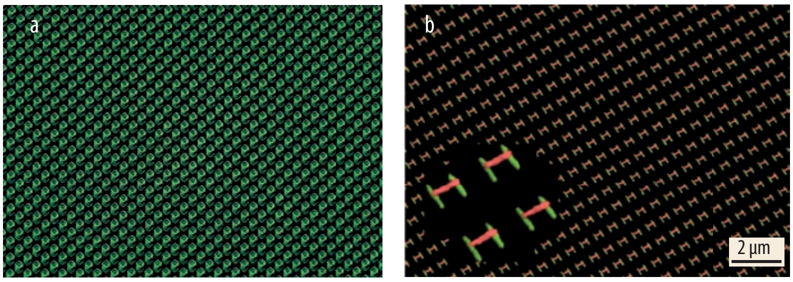

Fig. 5 A 4-layer stereometamaterial, consisting of U-type plasmonic resonators (a), and a stacked two-layer metamaterial which can display the plasmonic analog of EIT due to dipole-quadrupole coupling (b).

in which the two SRR "atoms" are inductively coupled, taking the advantage of structural aysmmetry. The two magnetic dipoles are aligned antiparallel and parallel at the lower and higher resonances, respectively (**Fig. 4i**). This complies with the hybridization picture of two transversely coupled dipoles. In the antisymmetric mode, the north and south poles of the two neighboring magnetic dipoles attract each other, therefore leading to the lower frequency resonance. In the symmetric mode, the poles with same sign are repulsive, leading to the higher frequency resonance.

More complex three-dimensional systems

The same principles that we have described here, namely longitudinal and transverse coupling of electric and magnetic dipole moments can be applied to three-dimensional systems as well [9]. In such stacked systems, we have another degree of freedom. This can lead to rather complex structures such as stacked cut-wire metamaterials [6], stacked fishnet metamaterials [10], stereometamaterials (**Fig. 5a**) [8], which can possess chiral optical properties and a complex twisting dispersion, where even higher-order multipoles can play a dominant role in the optical spectra [11]. It is quite intriguing to couple dipole and quadrupole resonances together, which can lead to the plasmonic analog of electromagnetically induced transparency (EIT) (**Fig. 5b**) [12]. In this case, the single wires constitute the dipole resonances, which can couple to the light and are therefore bright, whereas the double wires in the adjacent layer are associated with quadrupole resonances, which are dark. Tuning the resonance frequencies as well as the coupling strength suitably will lead to Fano-type lineshapes, which are only limited by the intrinsic losses of the metal. Such effects will exhibit pronounced and narrow resonances in the usually broad particle plasmon spectrum and are well suited for novel sensors with very high sensitivity and attoliter sensing volumes [13]. In three dimensions, there is such a plethora of possible combinations that many effects from solid state physics can be found as possible analogs, for example classical ferro- and antiferromagnetism [14]. Plasmonic oligomers with pronounced Fano resonances is another playground that requires our coupling schemes [15].

In the future, we envision that three-dimensional plasmonic nanostructures will be combined with quantum emitters to form hybrid nanostructures, which can act as optical nanoantennas and lead to efficient emission and detection of light in nanoscopic volumes.

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References

[1] *S. Linden* and *M. Wegener*, Physik Journal, December 2006, p. 29
[2] *G. Dolling, M. Wegener,* and *S. Linden*, Physik in unserer Zeit **38**, 24 (2006)
[3] *C. M. Soukoulis, S. Linden,* and *M. Wegener*, Science **315**, 47 (2007)
[4] *N. Liu, H. C. Guo, L. W. Fu, S. Kaiser, H. Schweizer,* and *H. Giessen*, Nature Materials **7**, 31 (2008)
[5] *E. Prodan, C. Radloff, N. J. Halas,* and *P. Nordlander*, Science **302**, 419 (2003)
[6] *N. Liu, H. C. Guo, L. W. Fu, S. Kaiser, H. Schweizer,* and *H. Giessen*, Adv. Mater. **19**, 3628 (2007)
[7] *N. Liu, S. Kaiser,* and *H. Giessen*, Adv. Mater. **20**, 4521 (2008)
[8] *N. Liu, H. Liu, S. Zhu,* and *H. Giessen*, Nature Photonics 3, 157 (2009)
[9] *N. Liu* and *H. Giessen*, Angew. Chemie, in press (2010)
[10] *N. Liu, L. W. Fu, S. Kaiser, H. Schweizer,* and *H. Giessen*, Adv. Mater. **20**, 3859 (2008).
[11] *H. Liu* et al., Phys. Rev. B **81**, 241403 (2010)
[12] *N. Liu, L. Langguth, T. Weiss, J. Kästel, M. Fleischhauer, T. Pfau,* and *H. Giessen*, Nature Materials **8**, 758 (2009)
[13] *N. Liu, T. Weiss, M. Mesch, L. Langguth, U. Eigenthaler, M. Hirscher, C. Sönnichsen,* and *H. Giessen*, Nano Lett. **10**, 1103 (2010)
[14] *N. Liu* and *H. Giessen*, Opt. Express **16**, 21233 (2008)
[15] *M. Hentschel, M. Saliba, H. Giessen, A. P. Alivisatos,* and *N. Liu*, Nano Lett. **10**, 2721 (2010)

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