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Controlling plasmon hybridization for negative refraction metamaterials

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The hybridization scheme of plasmon modes in cut-wire-based left-handed metamaterials is shown to critically depend on the coupling between paired cut wires. We show that an inverted hybridization scheme obtained with an asymmetric alignment of paired cut wires is the most appropriate to negative refraction. This is validated (numerically and experimentally) by the first demonstration of negative refraction in the microwave domain using only periodic ensembles of cut wires.

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I. INTRODUCTION

Metamaterials have attracted considerable interest since the pioneering work by Pendry *et al.*[1,2] and Smith *et al.*[3] While the association of split-ring resonators and continuous wires has been the main architecture of metamaterials in the microwave domain,[3] a periodic array of paired metallic cut wires was the first structure to exhibit negative refractive index in optics.[4] Surprisingly, attempts to reproduce negative refraction with this simple cut-wire structure in the microwave regime failed despite it being easier to fabricate at longer wavelengths.[5,6] In these experimental attempts, the spectral region of negative dielectric permittivity and that of negative magnetic permeability did not overlap. A broadband electric plasma (e.g., a periodic array of continuous wires) has thus been incorporated into the cut-wire structure to obtain negative refraction.[5,6] The resulting structure and its analogs, also called mesh or fishnet structures, are now recognized as universal structures for negative refraction in optics and microwaves.[5–11] Their great potential has been confirmed in recent research where negative refraction was extended for the first time to a three-dimensional optical metamaterial.[11] However, the large fraction of metal contained in fishnet structures is an important drawback because it can lead to high-level losses especially in the optical domain. In addition fishnet structures do not provide an independent control of the magnetic and electric responses of metamaterials as is necessary, for instance, in electromagnetic cloaking.[12,13]

Artificial magnetism in paired cut wires stems from the hybridization of the plasmon modes of each individual cut wire, thus forming two separate eigenmodes with opposite symmetry. The antisymmetric mode provides artificial magnetism, while the symmetric mode mainly contributes to the dielectric permittivity of the effective medium. Figures 1(a) and 1(b) show the hybridization scheme in the case where the degeneracy of cut-wire eigenmodes is lifted due to the strong coupling between vertically aligned cut wires. If the coupling strength between vertically aligned cut wires is very high (for very small spacing between them), the symmetric and antisymmetric bands are split further apart [Fig. 1(c)]. The design of metamaterials with a negative index of refraction consists of preserving a certain overlap between the symmetric and antisymmetric frequency bands.

In this work, we show that true negative index of refraction can be achieved by appropriately controlling the coupling strength between paired cut wires of adjacent layers. The coupling strength is itself controlled by adjusting either the spacing or the alignment of paired cut wires. Using an asymmetric alignment, an inverted hybridization scheme where the asymmetric mode is at higher frequency than the symmetric mode is predicted and thus more favorable for obtaining negative refraction. The first experimental demonstration of a negative refraction metamaterial exclusively based on paired cut wires is reported in the microwave range using such an inverted hybridization scheme. This scheme is also applicable to the optical domain.

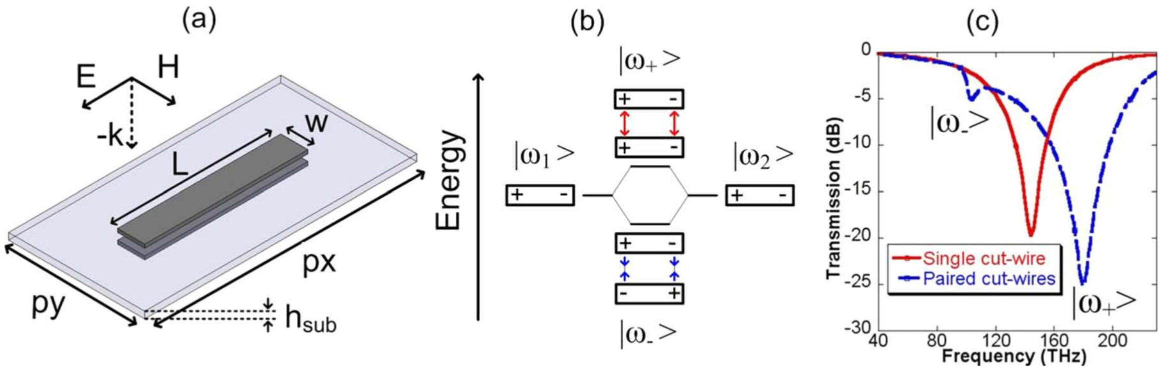

FIG. 1. (Color online) (a) Schematic of the symmetric cut-wire pair. (b) Hybridization scheme of the two coupled dipoles. (c) Transmission spectra calculated at normal incidence for a periodic array of cut wires [red (continuous dark gray)] and of paired cut wires [blue (dashed black)], respectively ($px=1.2$ μm, $py=200$ nm, $w=30$ nm, $L=600$ nm, and $h_{sub}=100$ nm). The 30–nm-thick gold cut wires are described using a Drude model whose parameters can be found in Ref. 13. The dielectric spacer (SiO_2) permittivity is $\varepsilon_{sub}=2.25$.

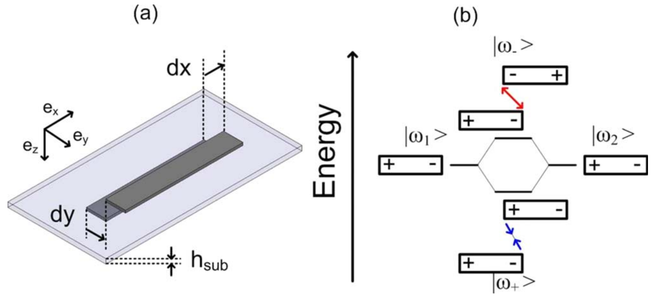

FIG. 2. (Color online) (a) Asymmetric cut-wire pair with the 3 degrees of freedom for the control of the coupling strength: h_{sub}, dx, and dy. (b) Inverted hybridization scheme.

II. MODE ENGINEERING AND COUPLING STRENGTH IN METAMATERIALS BASED ON PAIRED CUT WIRES

The plasmon hybridization scheme as recently introduced in Refs. 14 and 15 gives an intuitive electromagnetic analog of molecular orbital theory. Such a scheme has largely been used by the metamaterial community especially for simplifying metamaterial designs at optical wavelengths.[4,16] The coupling of two electric dipoles facing each other has thus been exploited to mimic magnetic atoms and alter the effective magnetic permeability of metamaterials in the optical range. While a magnetic activity was indeed obtained from metamaterials comprised of metallic dipoles,[4,16] negative refraction was reported only in the pioneering demonstration by Shalaev *et al.*[4] who used a periodic array of cut-wire pairs. In order to unambiguously achieve negative refraction, the magnetic activity must actually occur within a frequency band in which the dielectric permittivity is negative. For this purpose, one solution consists of associating magnetic "atoms" (coupled metallic dipoles) to a broadband "electric plasma" (continuous wires) in the same structure. Many authors have used this solution either in the microwave[5,6] or in the optical regime,[17,18] thereby contributing to the development of the so-called fishnet structure. We propose another solution based on the control of the coupling between metallic dipoles in such a way that the symmetric and antisymmetric bands have a sufficient overlap. The coupling strength is varied either by changing the distance between coupled dipoles or by breaking the symmetry of the structure.

Figure 1(a) shows the rectangular unit cell of the studied two-dimensional (2D) structure in the case where the coupled metallic dipoles (cut wires) are vertically aligned. This structure is henceforth referred to as the symmetric cut-wire structure to distinguish it from the asymmetric structure discussed later (see Fig. 2). Both structures consist of 2D periodic arrays of metallic cut wires separated by a dielectric spacer.[4] The electromagnetic wave should propagate normally to the layers with the electric field parallel to the longest side of dipolar elements. A normal incidence is thus considered. Such structures can be described in terms of effective index as long as the cut-wire width w and spacer thickness h_{sub} are much smaller than the wavelength.[4,7–11] Two series of calculations were carried out using a finite element simulation package (HFSS from Ansoft), one for symmetric structures and the other for asymmetric structures. The effective index was obtained from the calculated transmission and reflection coefficients.[19]

The first series of calculations were performed to compare the electromagnetic response of a symmetric cut-wire bilayer [blue curve (dashed black) in Fig. 1(c)] to that of a cut-wire monolayer [red curve (continuous dark gray) in Fig. 1(c)]. As is evident in the curve, only one resonance is observed for the cut-wire monolayer in the frequency range of interest. This resonance corresponds to the fundamental cut-wire dipolar mode which, in the optical regime, can also be interpreted in terms of a localized plasmon resonance.[20] Collective electronic excitations, also called surface plasmons, are indeed the main mechanism at short wavelengths. For the cut-wire bilayer, the coupling between paired cut wires lifts the degeneracy of the single cut-wire mode, which hybridizes into two plasmon modes. One mode is symmetric and corresponds to in-phase current oscillations, while the other is antisymmetric and corresponds to out-of-phase current oscillations. For a symmetric cut-wire pair with a vertical alignment of the two cut wires, the antisymmetric mode is the low-energy (low-frequency) mode since attractive forces are present in the system. Conversely, repellent forces are produced in the case of the symmetric mode that is therefore the high-frequency mode. The stronger the coupling (the smaller the spacing between the dipoles), the larger the frequency difference between the two modes. The evolution of the transmission spectra with the thickness of the dielectric spacer (or substrate) h_{sub} is illustrated in Fig. 3(a) in the case of a structure designed for operation in the microwave regime. Similar results were obtained for the structure in Fig. 1(c).

A second series of calculations were performed to analyze the influence of a vertical misalignment of metallic dipoles at a fixed spacer (or substrate) thickness. For this purpose, the cut-wire layers were shifted from each other in the horizontal XY plane [Fig. 2(a)] thus breaking the symmetry of the cut-wire structure. The relative displacements dx and dy in the X and Y directions, respectively, were used as parameters. The electromagnetic response of this type of asymmetric structure was studied both in the microwave and optical regimes. Similar evolutions of the hybridization scheme were found whatever the regime was. Results of Fig. 3 correspond to the microwave structure that was fabricated in this work. The substrate thickness was chosen to be equal to that of commercially available epoxy dielectric boards (1.2 mm). For this thickness and a vertical alignment of paired cut wires ($dx=dy=0$), the calculated transmission spectrum in Fig. 3(a) revealed a pronounced frequency separation between the symmetric (electric) and antisymmetric (magnetic) modes. Figures 3(b) and 3(c) show the evolution of the transmission spectrum for nonzero values of the longitudinal (dx) and lateral (dy) displacements, respectively. Quite surprisingly, as previously reported by Christ *et al.*[21] for the control of Fano resonances in a plasmonic lattice of continuous wires, symmetry breaking can invert the hybridization scheme due to modified Coulomb interactions [Fig. 2(b)] resulting in the symmetric resonance occurring at a lower frequency than the antisymmetric one. The Coulomb forces in our system result from the interaction of charges located at the cut-wire ends.

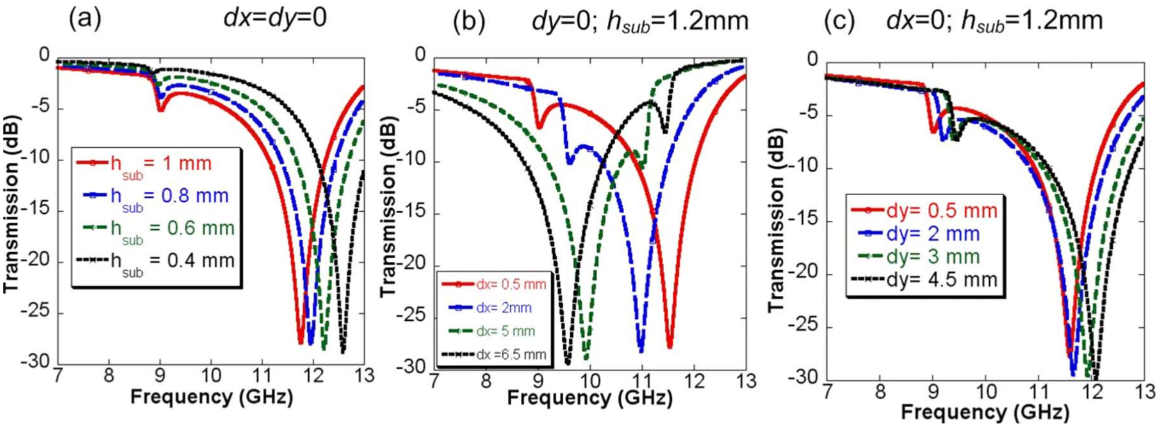

FIG. 3. (Color online) Influence of the coupling strength on the transmission spectra of a bilayer structure ($px=19$ mm, $py=9.5$ mm, $w=0.3$ mm, $L=9.5$ mm, and $h_{\mathrm{sub}}=1.2$ mm). The substrate permittivity is $\varepsilon_{\mathrm{sub}}=3.9$. (a) Variation in the dielectric spacer (or substrate) thickness h_{sub}. (b) Variation in the longitudinal shift dx. (c) Variation in the lateral shift dy.

When the longitudinal shift (dx) is progressively increased, the signs of the charges in close interaction change. As a result the repulsive force becomes attractive and vice versa. Correspondingly, the symmetric mode becomes the low-energy mode while the asymmetric mode is shifted to higher frequencies. It is evident that this inversion process is impossible in the case of a lateral dy displacement of the dipoles [Fig. 3(c)]. Let us notice that another structural asymmetry has recently been reported in Ref. 22 leading to an increase in the antiphase resonant mode response, but no inverted hybridization scheme was evidenced in that case.

Controlling the coupling between metallic dipoles thus allows the two plasmons resonances to be engineered. When the magnetic and electric modes are very close together, a negative refraction material can be obtained. More generally, the design of true negative index metamaterials can be achieved by appropriate design of the three degrees of freedom h_{sub}, dx, and dy.

III. NEGATIVE INDEX OF REFRACTION IN ASYMMETRIC CUT-WIRE STRUCTURES

Results in Fig. 3(a) clearly show that strong coupling and small spacing between paired dipoles do not favor negative refraction in cut-wire bilayers. In the optical regime, the thickness of dielectric spacers is in general easily controlled using standard layer deposition techniques. However, breaking the symmetry of a multilayer stack (Fig. 2) can be of interest when the use of additional techniques, such as planarization, modifies the thickness of each deposited layer.[16] Moreover, the fact that a strict vertical alignment of cut wires is not required simplifies the fabrication process. In the microwave regime, the thickness of the dielectric spacer or substrate is usually that of commercially available dielectric boards. Figure 3(a) most likely explains why negative refraction was not obtained with standard substrates whose thickness was of the order of $\lambda/25$. Breaking the symmetry of the structure is in this case an attractive solution to provide negative index. Our experiments were carried out to demonstrate the possibility of negative refraction metamaterial in the microwave spectral regime using an asymmetric cut-wire structure.

A photograph of the fabricated cut-wire bilayer is shown in Fig. 4(a). The structure consists of cut-wire pairs with $L=9.5$ mm, $w=0.3$ mm, $px=19$ mm, $py=9.5$ mm, $h_{\mathrm{sub}}=1.2$ mm, $dx=9.5$ mm, and $dy=0$. Measurements were performed in free space with an Agilent 8722ES vectorial network analyzer and two X-band horn antennas. Figure 4(b) shows the measured and calculated transmission/reflection spectra. There is an excellent agreement between experiments and theory. The dips in the transmission spectra near 9.5 and 11.5 GHz correspond to the symmetric and antisymmetric modes, respectively. The real and imaginary parts of effective index, n, retrieved from both experiments and

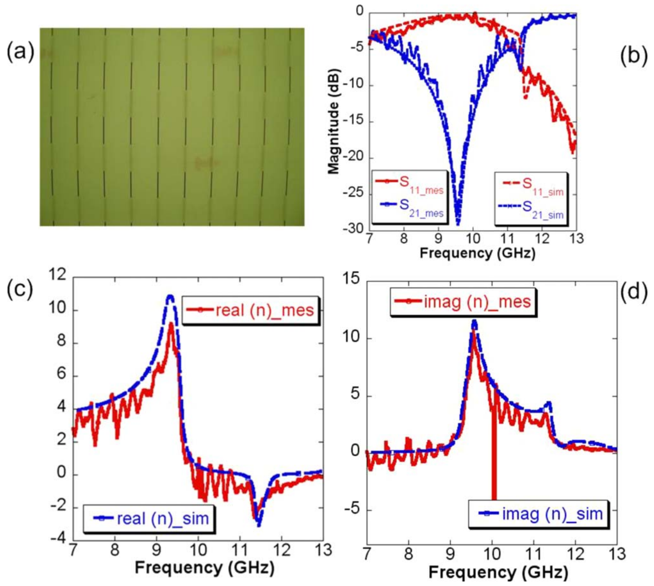

FIG. 4. (Color online) (a) Top photograph of the microwave prototype. Cut wires on the bottom face of the semitransparent substrate appear in light gray. (b) Measured and simulated reflection (S_{11}) and transmission (S_{21}) coefficients. (c) and (d) Retrieved effective index: real and imaginary parts of the relative refractive index.

theory are reported in Figs. 4(c) and 4(d). A negative index of refraction is unambiguously observed around 11.5 GHz. The figure of merit (ratio between the real and imaginary parts of n) at this frequency is found to be as high as 42, which is the highest value reported so far for this kind of metamaterial. We have thus demonstrated a negative refraction metamaterial in the microwave regime using only cut wires as the metallic constituents. Direct measurements of the refractive index using a prism configuration will be published elsewhere. We may notice that the cut-wire length is ~2.75 times smaller than the negative index wavelength, which itself is ~22 times larger than the metamaterial thickness. The propagation of light at normal incidence and within some angular range is mainly dominated by the material thickness, not by the in-plane periodicity.[11] In contrast, because of in-plane dimensions comparable to half the wavelength, the structure should exhibit a "photonic-crystal-like" behavior for plane waves with large in-plane wave-vector component. As for most of the negative-refraction metamaterials published in the literature, our structure suffers from the fact that negative refraction is obtained over a limited range of incident angles. In turn, the present concept of inverted hybridization for negative refraction can be applied to more "compact" structures so as to better satisfy the homogenization rules at all angles of incidence.[23]

IV. CONCLUSION

Design rules have been established for the control of the coupling strength between paired metallic dipoles in cut-wire bilayers, thereby allowing the engineering of the symmetric and antisymmetric modes. An inversion of the hybridization scheme has been shown when an asymmetric alignment of the paired cut wires is used. More generally, the coupling strength can be controlled by adjusting either the spacing or the alignment of the paired cut wires. Negative refraction has been demonstrated in an asymmetric cut-wire structure in the microwave domain. These results open up solutions for the design of metamaterials both in the microwave and optical spectral regimes. Clearly, experimental demonstration in the microwave regime cannot be automatically transferred to the optical regime. However, numerical calculations accounting for finite conductivity and losses of metal at optical frequencies indicate that the inverted hybridization scheme still works for obtaining negative refraction metamaterials in optics.

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[1] J. B. Pendry, A. J. Holden, D. J. Robbins, and W. J. Stewart, J. Phys.: Condens. Matter **10**, 4785 (1998).

[2] J. B. Pendry, A. J. Holden, D. J. Robbins, and W. J. Stewart, IEEE Trans. Microwave Theory Tech. **47**, 2075 (1999).

[3] D. R. Smith, W. J. Padilla, D. C. Vier, S. C. Nemat-Nasser, and S. Schultz, Phys. Rev. Lett. **84**, 4184 (2000).

[4] V. M. Shalaev, W. Cai, U. K. Chettiar, H. K. Yuan, A. K. Sarychev, V. P. Drachev, and A. V. Kildishev, Opt. Lett. **30**, 3356 (2005).

[5] J. Zhou, L. Zhang, G. Tuttle, T. Koschny, and C. M. Soukoulis, Phys. Rev. B **73**, 041101(R) (2006).

[6] K. Guven, M. D. Kaliskan, and E. Ozbay, Opt. Express **14**, 8685 (2006).

[7] M. Kafesaki, I. Tsiapa, N. Katsarakis, T. Koschny, C. M. Soukoulis, and E. N. Economou, Phys. Rev. B **75**, 235114 (2007).

[8] S. Zhang, W. Fan, N. C. Panoiu, K. J. Malloy, R. M. Osgood, and S. R. J. Brueck, Phys. Rev. Lett. **95**, 137404 (2005).

[9] G. Dolling, M. Wegener, C. M. Soukoulis, and S. Linden, Opt. Lett. **32**, 53 (2007).

[10] G. Dolling, C. Enkrich, M. Wegener, C. M. Soukoulis, and S. Linden, Science **312**, 892 (2006).

[11] J. Valentine, S. Zhang, T. Zentgraf, E. U. Avila, D. A. Genov, G. Bartal, and X. Zhang, Nature (London) **455**, 376 (2008).

[12] D. Schurig, J. J. Mock, B. J. Justice, S. A. Cummer, J. B. Pendry, A. F. Starr, and D. R. Smith, Science **314**, 977 (2006).

[13] B. Kanté, A. de Lustrac, J.-M. Lourtioz, and S. N. Burokur, Opt. Express **16**, 9191 (2008).

[14] U. Kreibig, A. Althoff, and H. Pressmann, Surf. Sci. **106**, 308 (1981).

[15] E. Prodan, C. Radloff, N. J. Halas, and P. Nordlander, Science **302**, 419 (2003).

[16] N. Liu, H. Guo, L. Fu, S. Kaiser, H. Schweizer, and H. Giessen, Nature Mater. **7**, 31 (2008).

[17] E. Pshenay-Severin, U. Hübner, J. Petschulat, C. Rockstuhl, T. Pertsch, F. Lederer, and A. Tünnermann, Proceedings of META'08, Marrakech, Morocco, 2008 (unpublished).

[18] T. F. Gundogdu, N. Katsarakis, M. Kafesaki, R. S. Penciu, G. Konstantinidis, A. Kostopoulos, E. N. Economou, and C. M. Soukoulis, Opt. Express **16**, 9173 (2008).

[19] D. R. Smith, S. Schultz, P. Markos, and C. M. Soukoulis, Phys. Rev. B **65**, 195104 (2002).

[20] C. Rockstuhl, F. Lederer, C. Etrich, T. Zentgraf, J. Kuhl, and H. Giessen, Opt. Express **14**, 8827 (2006).

[21] A. Christ, Y. Ekinci, H. H. Solak, N. A. Gippius, S. G. Tikhodeev, and O. J. F. Martin, Phys. Rev. B **76**, 201405(R) (2007).

[22] T. Pakizeh, A. Dmitriev, M. S. Abrishamian, N. Granpayeh, and M. Käll, J. Opt. Soc. Am. B **25**, 659 (2008).

[23] Constantin R. Simovski and S. A. Tretyakov, Phys. Rev. B **75**, 195111 (2007).