Resonant coupling to a dipole absorber inside a metamaterial: Anticrossing of the negative index response

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The authors experimentally demonstrate a resonant hybridization between the magnetic dipole structural resonance in the permeability of a fishnet metamaterial and an electric dipole material resonance in the permittivity of the dielectric spacer layer. The hybrid resonances in the permeability and the negative index response exhibit an anticrossing behavior. A simple analytic model and numerical simulations using a rigorous coupled-wave analysis are in excellent qualitative agreement with the experiment. © 2010 American Vacuum Society. [DOI: 10.1116/1.3503898]

I. INTRODUCTION

Metamaterials, first discussed by Veselago in 1968 (Ref. 1), are a new class of nanostructured materials that offer novel optical properties such as a negative index of refraction. To date, much effort has been devoted to the fabrication of these materials, the characterization of their linear optical properties, and the extension of their operation wavelength range to the near-infrared and visible.2,3 Current theoretical and experimental work is directed toward exploring new properties and potential applications for negative index metamaterials. The infrared spectral region is of a particular interest because metal properties are intermediate between their very highly conductive properties in the RF and the lossy plasmonic characteristics in the visible. In the infrared wavelength region, most commonly used dielectrics have some absorption resonances. It is of interest to consider the coupling between the metamaterial resonance and the dipole resonance of the absorption. We present experimental and modeling results for a fishnet metamaterial structure where the dielectric spacer layer contains a simple Lorentzian-type electric dipole resonance and demonstrates a classic anticrossing behavior along with an exchange of oscillator strength. Possible applications for such behavior include the development of novel sensors and the use of absorption excitation to alter and switch the metamaterial optical properties.

II. SIMPLE PHYSICAL AND RIGOROUS NUMERICAL MODELS

The negative index of refraction of the fishnet structure comes from two parts: (1) a broadband negative electric permittivity ($\varepsilon$) that results from an array of thin metal wires parallel to the direction of electric field and (2) a resonant negative magnetic permeability ($\mu$) resulting from a pair of finite-width metal stripes separated by a dielectric layer along the direction of the incident magnetic field forming an $LC$ tank circuit. In order to investigate the physical effect of adding a dielectric with a dipole absorption resonance to a fishnet metamaterial, a simple analytical model for resonance coupling and more rigorous numerical models were constructed. The effective permeability of fishnet metamaterials can be associated with an effective tank ($LC$) circuit and is often described using a Lorentzian response.4,5 Following O’Brien et al.,6 we define the effective permeability of the fishnet structure as

$$\mu_{eff} = 1 - \frac{F\omega^2}{\omega^2 - \omega_0^2 + i\omega \gamma_0},$$

where the metamaterial resonance is characterized by a resonance frequency $\omega_0/2\pi$=33.0 THz (9.1 $\mu$m, 0.136 eV), a linewidth $\gamma_0/2\pi$=1.2 THz (0.33 $\mu$m, 0.005 eV), and a fill factor $F$~0.2. In terms of the equivalent circuit parameters,

$$\omega_0^2 = \frac{1}{LC} = \frac{1}{LC_0\varepsilon_{abs}}.$$

Here, $C_0$ is the capacitance without the absorber, and the absorber dielectric function has been explicitly separated out. Using a standard expression for the Lorentzian absorption,7

$$\varepsilon_{abs} = 1 - A \gamma_1 \left( \frac{1}{\omega + \omega_1 + i\gamma_1} + \frac{1}{\omega + \omega_1 - i\gamma_1} \right),$$

where $A$=0.3 and is related to the oscillator strength, $\omega_1/2\pi$=31.6 THz (9.5 $\mu$m, 0.130 eV) is the resonance frequency of the electric dipole transition, and $\gamma_1/2\pi$ =0.83 THz (0.25 $\mu$m, 0.0034 eV) is the inverse linewidth. This set of equations was used to evaluate the magnetic resonance response of a fishnet metamaterial.

Rigorous coupled-wave analysis (RCWA),8,9 a commonly used algorithm to calculate the transmission and reflection of periodic structures, was used for a detailed numerical modeling of the resonance anticrossing behavior. The effective parameters (refractive index $n$, impedance $\eta$, permittivity $\varepsilon$,

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and permeability $\mu$) can all be extracted from the complex transmission and reflection coefficients using methods described in Ref. 10. For all calculations, the incident light is normal to the surface and both the incident and outgoing media are air. For the simulation results, which will be compared to the experimental measurements, the outgoing media is an infinite slab of the material with a refractive index that matches the index of the substrate of the experimental sample. Only small changes in the peak value of the transmission and the effective refractive index were observed with no change in the general behavior with the addition of the substrate. In our simulations, we retained 13 diffraction orders in each direction; all of the simulation results converged. A common fishnet structure, as shown in Fig. 1, was used for the numerical modeling. The orthogonal pitches of the two-dimensional gratings ($a_x$, $a_y$) and the linewidths (critical dimensions: $d_x$, $d_y$) used to model a fishnet structure with the resonance at $\sim 9.2 \ \mu$m are listed in Table I.

In the absence of a dielectric dipole resonance, a conventional single resonance peak is observed in both the simple physical and RCWA models, resulting from the coupling of the broadband negative permittivity with the structurally resonant negative permeability associated with the LC circuit between the two metal plates. Figures 2(a) and 2(b) show the calculated effective permeability and transmission response of the fishnet metamaterial. However, the addition of a dipole absorber in the dielectric changes the response of the fishnet structure such that it exhibits doubly resonant behavior. This behavior is observed in the form of two resonance peaks in Figs. 2(c) and 2(d), showing the response of the fishnet structure when the dipole absorption is centered at $9.5 \ \mu$m. It is interesting to note that the presence of an electric dipole resonance in the dielectric of the fishnet structure manifests itself in a modification of the magnetic permeability, which in turn modifies the negative index behavior.

The anticrossing behavior of the two resonances is clearly evident in both the simple physical model and the RCWA numerical simulation, with the presence of the electric dipole absorber in the dielectric media, as the bare metamaterial structural resonance is tuned through the center of the dipole transition. There is good qualitative agreement between the two models, as shown in Fig. 3.

### III. EXPERIMENTAL PROCESS

For the experimental demonstration, a fishnet structure consisting of Al and bisbenzocyclobutene (BCB) was fabricated on a BaF$_2$ substrate (refractive index of $\sim 1.42$). The layers of the structure were Al (thickness of 100 nm) on top of BCB (thickness of 800 nm) on top of Al (thickness of 100 nm). Thicknesses within the functional layer were selected to optimize and maximize the transmission of the structure. Figure 4 shows measured $n$ and $k$ data profiles for the BCB dielectric material (measurement courtesy of G. Boreman). There are two absorption peaks in the long-wave infrared, at $7.9 \ \mu$m (0.157 eV) and at $9.5 \ \mu$m (0.130 eV). The additional material resonances at wavelengths longer than $10 \ \mu$m are outside the wavelength range explored in this experiment. The resonances of the fishnet structure were structurally tuned over the range from 7.4 to 10.3 $\mu$m (from 0.16 to 0.120 eV).

A set of seven samples was fabricated on a single BaF$_2$ substrate using standard optical lithography and cleanroom processing. An e-beam evaporated Al film was deposited on the top of a 2 in. BaF$_2$ substrate, followed by the spin-on coating of the BCB film and a second Al e-beam evaporation. A layer of SiO$_2$ was deposited by plasma-enhanced chemical vapor deposition. This silicon oxide layer will act as a hard mask for the etching of previously deposited structural films. A spin-deposited photoresist layer is patterned using conventional optical lithography and the resulting profile is transferred into the underlying SiO$_2$ layer using reactive ion etching (RIE) with the remaining photoresist as an etch mask. After stripping the resist mask, a three-step etch process was used to transfer the pattern from the hard mask to the structural films. A Cl$_2$ based inductively coupled plasma-RIE was used to etch through the aluminum layers, while a CF$_4$/O$_2$ RIE was used to transfer the pattern through the BCB film. A thin layer of the silicon oxide was left on the top of the aluminum film to protect it from oxidation. Figure 5 shows optical microscope images of two of the structures with the indicated geometrical parameters. Due to the variations of the lithographic and etch processes, final values for the critical dimensions (CDs) are slightly off of the design parameters, but the pitch was matched accurately. For example, in Fig. 5, left, the final $d$’s are 2.54 and 1.41 $\mu$m, while target values were 2.64 and 1.32 $\mu$m, respectively. There is an $\sim 10\%$ deviation; it is necessary to use the actual values in the model analysis.

<table>
<thead>
<tr>
<th>Pitch ($a_x$, $a_y$)</th>
<th>CD ($d_x$)</th>
<th>CD ($d_y$)</th>
<th>$n_{\text{dielectric}}$</th>
<th>Al thickness</th>
<th>Dielectric thickness</th>
</tr>
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<tbody>
<tr>
<td>5.2 $\mu$m</td>
<td>3.12 $\mu$m</td>
<td>1.56 $\mu$m</td>
<td>1.5</td>
<td>100 nm</td>
<td>800 nm</td>
</tr>
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**Table I.** Geometrical parameters of a fishnet structure with a structural resonance wavelength of $\sim 9.2 \ \mu$m.

**Fig. 1.** (Color online) Side view schematic of the metal-dielectric-metal fishnet structure.
IV. EXPERIMENTAL RESULTS

Fourier transform infrared spectroscopy (FTIR) measurements were carried out to obtain transmission spectra for each individual sample, normalized to a bare BaF$_2$ substrate. The effective refractive index has not been extracted because only transmission spectra have been obtained. Once reflection and phase information are obtained, the behavior of effective $n$ can be evaluated.

Figure 6 shows overlapped experimental (FTIR) and modeled (RCWA) transmission curves, as well as the RCWA simulated effective refractive index curve for the sample with a metamaterial structural resonance (MSR) at 7.67 $\mu$m. Three resonance dips are apparent in the model; however, only two dips are clearly visible in the FTIR data. The third dip is very shallow and is hard to locate. Dips $n'$, $n_1$, and $n_2$ are due to the hybridization between BCB absorption peak at 7.9 $\mu$m (0.157 eV), the structural resonance at 7.67 $\mu$m (0.162 eV), and the second absorption BCB peak at 9.5 $\mu$m (0.13 eV). Overall, the experimental data show a good qualitative agreement with the RCWA model.

As the metamaterial structural resonance is scanned through the BCB absorption peaks to longer wavelengths, only two hybridized resonance dips are observed, as shown

Fig. 2. (Color online) (a) Simple physical model calculations for the effective permeability of the fishnet structure without the dielectric absorber; MSR is at 9.1 $\mu$m (0.136 eV) (top left). (b) RCWA calculated transmission with a dielectric material ($n=1.5$) without an absorber; MSR is at 9.2 $\mu$m (0.135 eV) (top right). A single MSR is observed for both models; (c) simple physical model calculations for the effective permeability with the dipole absorber (low left), where MSR is at 9.1 $\mu$m (0.136 eV) and absorption resonance is at 9.5 $\mu$m (0.130 eV); (d) RCWA calculated transmission (low right) of the fishnet structure with the presence of the dipole absorber in the dielectric; MSR is at 9.2 $\mu$m (0.135 eV) and absorption resonance (abs) is at 9.5 $\mu$m (0.130 eV). The hybridization of the resonances is clearly visible for both models.

Fig. 3. (Color online) Anticrossing behavior of the structural and absorption resonances as the electric dipole resonance tuned through the bare fishnet resonance is clearly observed. A bare fishnet resonance at 9.2 $\mu$m (0.135 eV) and an absorber resonance at 9.5 $\mu$m (0.130 eV) are assumed. Good qualitative agreement between detailed RCWA calculation (red, short lines) and simple physical model (black, long lines) is obtained.
in Fig. 7 for the samples with MSRs at 9.82 μm (0.126 eV) and 10.3 μm (0.12 eV). The change in the hybridization behavior can be explained by the fact that the MSR of the fishnet structure is now located close to the much stronger BCB absorption resonance at 9.5 μm (0.13 eV), therefore only two hybridized resonances \( n_1 \) and \( n_2 \) are observed along with a small dip at 7.9 μm (0.13 eV). Both experimental samples show a good qualitative agreement between the model and the experiment.

Fig. 4. (Color online) Measured \( n \) and \( k \) data profiles for the BCB dielectric material used in the designed fishnet structure. There are absorption peaks at 8 μm (0.155 eV) and at 9.5 μm (0.130 eV) in the wavelength range of interest (area between the vertical dashed lines).

Fig. 5. (Color online) Optical microscope images of the two extreme fishnet samples. Geometrical parameters for the critical dimensions are indicated on the images.

Fig. 6. (Color online) Overlapped experimental (FTIR) and modeled (RCWA) transmission curves (left) and RCWA model for the corresponding effective refractive index (right) curve for the sample with MSR at 7.67 μm (0.162 eV). Hybridized resonances \( n' \), \( n_1 \), and \( n_2 \) are indicated.

Fig. 7. (Color online) FTIR experimental and RCWA modeled overlapped transmission curves for the samples with MSRs at 9.82 μm (0.126 eV) on the left and 10.3 μm (0.12 eV) on the right. Hybridized resonances \( n_1 \) and \( n_2 \) are indicated on both graphs.
RCWA model results for the effective refractive index reveal the sharing of the oscillator strength, shown in Fig. 8, as the bare fishnet metamaterial resonance is tuned through the absorber resonances.

Figure 9 shows the anticrossing behavior of resonances. The experimental and numerical model data show a very good qualitative agreement. However, there are several issues that require further analysis. Several points on the highest and lowest energy hybridization branches cross the bare resonance curves. In particular, the lowest energy branch shows significant deviation from the model. This behavior differs from a classical anticrossing response that asymptotically approaches, but does not cross, the bare resonance frequencies. These results suggest a more complex behavior probably due to the presence of additional resonance and absorption peaks that are not yet included in our model. A more comprehensive analysis of the anticrossing behavior of the resonances is being developed.

V. CONCLUSIONS

We have experimentally and numerically demonstrated a novel resonance hybridization effect between metamaterial and dielectric absorption resonances. A simple physical model, detailed RCWA calculations, and experimental data all clearly show a coupling between the resonances with hybridization behavior and the sharing of oscillator strength. There is good qualitative agreement between the RCWA model calculations and experimental data. This effect is of interest for sensors and other optical applications at infrared frequencies and can be extended to different wavelengths.

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