Optical magnetic response in three-dimensional metamaterial of upright plasmonic meta-molecules

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Abstract: We report the first three-dimensional photonic metamaterial, an array of erected U-shape plasmonic gold meta-molecules, that exhibits a profound response to the magnetic field of light incident normal to the array. The metamaterial was fabricated using a double exposure e-beam lithographic process. It was investigated by optical measurements and finite-element simulations, and showed that the magnetic field solely depends on the plasmonic resonance mode showing either enhanced in the centre of the erected U-shape meta-molecule (16 times enhancement) or enhanced around two prongs of erected U-shape meta-molecule (4 times enhancement).

OCIS codes: (160.3918) Metamaterials; (240.6680) Surface plasmon; (140.4780) Optical resonators; (250.5403) Plasmonics.

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1. Introduction

Metamaterials are created as an array of artificial sub-wavelength structures, often exhibit unique optical properties which are not found in nature [1–5]. Metamaterials composited with sub-wavelength split ring resonators (SRRs) have attracted a wide attention because of a number of extraordinary properties, such as artificial magnetism [6–8], optical chirality [9,10], negative refraction index [11,12] and optical spectrum manipulation [13–15]. Electromagnetic (EM) properties of SRR were first studied in microwave region in 2000 [16]. Since 2004, interest in the EM properties of SRRs expanded to the optical frequency region [17,18]. In particular, U-shape SRRs were shown to exhibit a series of resonant modes as predicted by the *LC* circuit model [19,20], in which the capacitance was contributed by the charges accumulating near the ends while the inductance was contributed by the current flowing inside the U-shape SRR which can be viewed as a magnetic coil [21]. U-shape SRRs can be excited by an incident light with electric field perpendicular to two prongs of U-shape SRR (capacitance response, $\vec{E}\ //\ \hat{x}$, see Fig. 1(a)), or an incident light with magnetic field oscillating through the gap of U-shape SRR (inductance response, $\vec{H}\ //\ \hat{y}$, see Fig. 1(a)) [22], both showing pronounced dips in the transmission spectra. By increasing the dimension of the U-shape SRR, higher order resonance modes can also be excited [23–26]. However, due to the challenges in fabrications, so far most U-shape SRRs were fabricated as planar or multi-layered structures, some of those are three dimensional structures but in micrometer scale [27–30]. To excite the magnetic resonances of such planar SRRs, one must apply an off-normal incident light which possesses the required H field component. Such a restriction significantly lowers the coupling efficiency between the magnetic field of incident light and the resonant SRR.

In this paper, we successfully fabricated photonic metamaterial, an array of *vertical (off-plane)* U-shape SRRs in nanometer scale, and studied their optical properties by both experiments and numerical simulations. A unique characteristic of our structure is that its magnetic resonance can be excited by normally incident light, overcoming the above-mentioned coupling issues faced by planar SRRs. We found such erected SRRs to exhibit rich plasmonic resonances, showing unique magnetism responses related to the magnetic field of incident light.

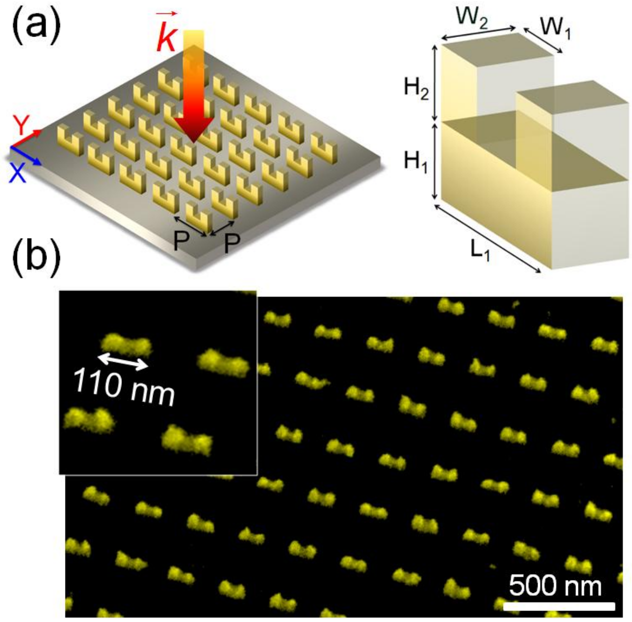

Fig. 1. (a) Schematic diagram showing the feature size of erected U-shape three-dimensional resonance ring, L_1 = 110 nm, H_1 = 30 nm, H_2 = 30 nm, W_2 = 40 nm, W_1 = 40 nm, P = 200 nm, respectively. The optical properties of the fabricated sample are studied in the case of x-polarized illumination as well as y-polarized illumination. In the case of x-polarized illumination, the electric field of incident light oscillate parallel to the resonant ring. In the case of y-polarized illumination, the electric field of incident light pass through the resonant ring. (b) SEM image of a small region from the fabricated sample. The inset shows a magnified view of four erected U-shape three-dimensional resonance rings with bottom length L_1 110 nm.

2. Fabrication

Using electron beam lithography with a double exposure process, we fabricated 375 × 375 erected U-shape three-dimensional (3D) gold resonance rings on fused silica substrate, covering a total area of ~75 × 75 μm^2. For the precision alignment of e-beam double exposure process, two golden cross alignment marks (size 9 × 150 μm) with a thickness 150 nm are fabricated and used. For the first exposure process, the bottoms of resonance rings are defined in positive resist (495k PMMA) after first e-beam exposure and lift-off process. Subsequently, the two prongs of resonance ring are made by second e-beam exposure and lift-off process. The Espacer (Kokusai Eisei Co., Showa Denso Group, Japan)/poly methyl methacrylate (PMMA-495K) bi-layered resist are spin-coated on a cleaned substrate. A 200 nm-thick PMMA layer is spin-coated on the fused silica wafer, then backed on a hot plate for 3 minutes at 180°C. The Espacer, a resist to eliminate the static charge problem during e-beam exposure, is spin-coated at 1500 rpm over the PMMA layer. An e-beam lithography system (Elionix ELS-7000) at the acceleration voltage of 100 kev with 30 pico-ampere of current is used.

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It is worth mentioning that we prepare our samples without sputtering any adhesion layer, such as Ti or Cr, between the gold film and the glass substrate. This fabrication method is especially suitable for our purpose, as the Ti or Cr layer usually changes the resonances of the metallic nanostructures, and it is too thin to be accurately modeled with our simulation software. Figure 1(a) shows the schematic diagram of the 3D resonant nano-rings. The size of each unit-cell is 110 nm length and 60 nm height, and the periodicity is 200 nm. The resonant rings are illuminated at normal incidence using x- and y-polarized light. Figure 1(b) shows an SEM micrograph of the fabricated pattern. The inset of Fig. 1(b) shows a magnified view of four erected U-shape three-dimensional resonance rings with the bottom length (L_1) 110 nm.

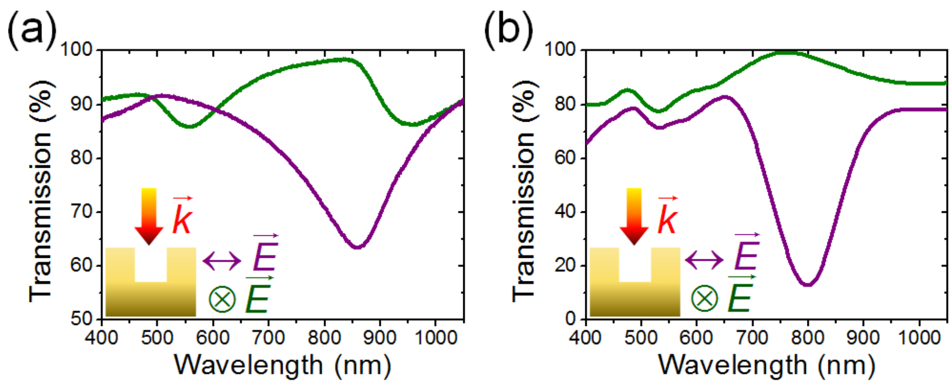

Fig. 2. (a) Experimental transmission spectra for x-polarized illumination (the purple curve) and y-polarized illumination (the green curve). (b) Finite-element simulation transmission spectra for x-polarized illumination (the purple curve) and y-polarized illumination (the green curve). The inset in both frames (a) and (b) show the propagating direction of incident light as well as the polarization direction of incident light related to the resonance ring.

3. Measurement and simulation

Figure 2 shows the measured and simulated transmission spectra through the sample as described in Fig. 1, illuminated by normal-incidence lights polarized along x and y directions. The transmission spectra from λ = 400 nm to λ = 1100 nm are measured using a B&W Tek Inc. BRC642E spectrometer combined with Carl Zeiss Axio microscope (10 × objective, numerical aperture NA = 0.3, 100W halogen light source and visible to near-infrared polarizer). The transmission spectra are normalized by the transmissivity of an un-patterned region of the fused silica wafer. The simulation spectra are obtained by solving three-dimensional Maxwell equation with the finite-element method (commercial software, Comsol Multiphysics), in which the refractive index of the fused silica glass substrate was taken as 1.4584 while the permittivity of gold was described by the Drude-Lorentz model with a damping constant 0.14 ev and a plasma frequency 8.997 ev [31,32].

We note that overall the experimental results (Fig. 2(a)) are in good agreement with the simulation results (Fig. 2(b)). In particular, both experiments and simulations show that a pronounced resonance dip exist at λ~800 nm in the spectra for $\vec{E} // \hat{x}$ while the structure is nearly transparent for light with the polarization $\vec{E} // \hat{y}$. The slight differences between experimental and simulation results are possibly due to the surface roughness in the real sample (ignored in the simulations) and the inaccuracies of the Drude model for the dielectric constant of Au adopted in our simulations.

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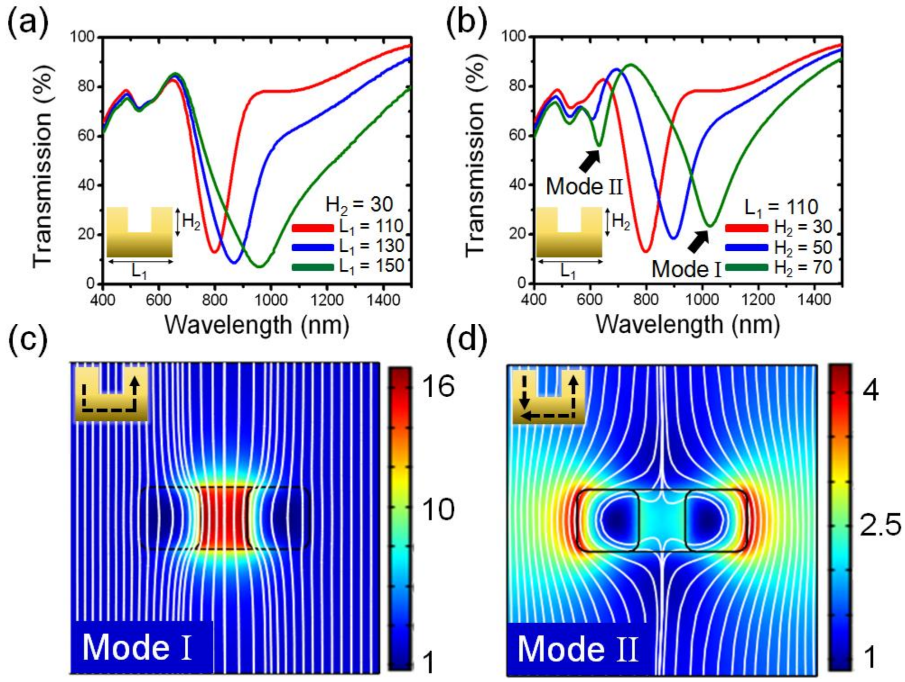

Fig. 3. Evolution of the finite-element simulation transmission spectra for x-polarized illumination upon varying the bottom length L_1 (a) and the prong length H_2 (b) of erected U-shape three-dimensional gold ring. The inset in frames (a) and (b) show the dimensions (in nm) of the prong length H_2 and the bottom length L_1 of erected U-shape gold ring. Simulation magnetic field in arbitrary units (color-coded) and field line of magnetic field (white line) for mode I (c) and mode II (d) of Fig. 3(b). The simulation results of a top view shown are at the root plane of the prongs of the erected U-shape nano gold ring. The inset of Figs. 3(c) and 3(d) show the direction of flowing surface current (black arrows) at a side view.

4. Results and discussions

As expected, when illuminated by a *y*-polarized light, the magnetic resonance of the 3D nano-rings cannot be excited so that the transmission spectra (the green curves in Figs. 2(a) and 2(b)) are nearly flat in both experiment and simulation. On the contrary, our structures show significant optical response to an *x*-polarized incident light, resulting in a pronounced dip in the transmission spectrum at the wavelength (λ) of 850 nm (the purple curves in Figs. 2(a) and 2(b)). This resonant mode results from a ring-like current flowing on the surface of the 3D nano-ring which has been demonstrated by FDTD simulation, and is a magnetic resonance which can be excited by external lights with a y component of magnetic field.

To gain a deeper understanding on the resonance mode, we varied the structural details of the nano-rings and then performed simulations to study the transmission spectra. Figure 3(a) shows the simulated spectra for nano-rings with H_2 (length of prong) kept as a constant 30 nm and L_1 (length of the bottom bar) changed from 110 nm to 150 nm. Obviously, the fundamental resonance mode shifts to longer wavelength as L_1 increases, which is understandable noting that the current flows on the longer path. Figure 3(b) shows the evolution of transmission spectra as H_2 increases. Again, we found a red shift of the resonance mode similar to Fig. 3(a). Intriguingly, we found another resonance at a higher frequency being excited by the *x*-polarized normal-incident light as H_2 increases. Magnetic field pattern and field line are shown in Figs. 3(c) and Fig. 3(d) to illustrate the resonance response of the erected U-shape nano gold ring. The plane is chosen as the root plane of its two prongs. Simulation also tell us the currents distribution in the nano gold rings. We can find that, the low order resonance mode, so called mode I, shows strong response to external magnetic field

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of incidence light in the gap of standing U-shape ring. Contrarily, the higher order mode, so called mode II, shows to push its magnetic field line outside its two standing prongs. In the case of mode I, the enhanced magnetic field happened between the prongs of the nano U-shape ring resulted from clockwise and anticlockwise surface current oscillation on the surface of gold ring structures (shown at the inset of Fig. 3(c)). The mode I exhibits parallel-like response to the magnetic field of incident light. In the case of mode II, a pair of anti-phase current along two prongs of the U-shape ring (shown at the inset of Fig. 3(d)) and the flowing current along the bottom of the U-shape resonance ring results in destructive (constructive) magnetic response at the inside (outer) wings of the resonance U-shape gold nano ring. The magnetic response at mode II shows anti-parallel-like response to the magnetic field of incident light. The currents distributions at the surface of rings (shown in the inset of Fig. 3(c) and Fig. 3(d)) can perfectly explain the magnetic field pattern illustrated here.

Due to the bi-anisotropy of the SRR, both electric (along x direction) and magnetic (along y direction) responses can be excited under the present normal-incident excitation. To identify the nature of discovered resonance modes, we calculated the transmission spectra under p-polarized oblique-incident excitation keeping $\vec{H} \,//\, \hat{y}$. We found that modes I and II can still be excited at essentially the same wavelengths under such oblique-incident excitations, demonstrating that these modes are predominantly magnetic resonances coupled to external H_y field. However, in order to gain a complete picture of these modes, one needs to study both the magnetic susceptibility and the bi-anisotropic magneto-electric susceptibility of the present structure, which is an interesting subject for further study.

5. Summary

Array of erected U-shape three-dimensional nano gold rings are fabricated vertically by a double e-beam lithography process. Plasmonic resonance modes of this U-shape three-dimensional nano gold rings are found from the experimental transmission spectra and finite-element simulations. Results show the low order resonance mode shifts to longer wavelength as the bottom of the vertical U-shape gold ring increased. When the length of prongs of the erected U-shape nano gold ring grows, a higher resonance mode can be excited by x-polarized illumination at normal incidence. The low order resonance mode is related to the U-shape surface current of resonance gold ring, and the higher order resonance mode is associated to a pair of anti-phase current along two prongs of the U-shape gold nano ring. Most interesting things are that the low and high order resonance modes show very distinct electromagnetic response to the magnetic field of incident light. The distribution of the magnetic field is either squeezed and enhanced between two prongs, or compelled and enhanced around two prongs of erected U-shape gold ring for low order and high order plasmonic resonance mode, respectively.

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