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We show that broadband optical magnetism can be achieved through incorporating multi-scaled 3D metallic meta-elements into Z-shaped nanohole arrays. The broadband effect arises from the excitation of multiple magnetic resonances in the meta-elements at different wavelengths. Moreover, the nanohole arrays exhibit a large transmission difference for left- and right-handed circularly polarized incident light due to the chiral arrangement of the meta-elements. More importantly, we have realized experimentally the broadband behavior for the optical range in Ag nanohole arrays fabricated by using a shadowing vapor deposition method. Our study opens up new opportunities for achieving broadband artificial magnetism at visible frequencies which allows possible applications in plasmonic bio-sensors or energy concentrators. Published by AIP Publishing.

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onto the nanohole array using a SVD technique in two directions at 45° and −45° from the Z-axis in the Y-Z plane as indicated by the orange arrows in the inset of Figs. 1(a) and 1(b) to fabricate the inverted U-shaped SRRs. Taking advantage of the SVD technique, the arm lengths of the inverted U-shaped SRRs can be controlled by adjusting the deposition angle. Three inverted U-shaped SRRs, labeled as black inverted letter “U,” are numbered in white in Fig. 1(a). Furthermore, two bottom nanostrips were also coated onto the ITO/glass substrate to form metal/dielectric nanostrip antennae shown as a top view of one unit cell in Fig. 1(c). The two nanostrips are connected to side walls of the inverted SRR 1, in effect creating more resonators. Figures 2(a) and 2(b) show Scanning Electron Microscopy (SEM) images of the fabricated Z- and inverse Z-shaped Ag nanohole arrays, respectively. The titled SEM images of the unit cells in the insets of Figs. 2(a) and 2(b) show clearly the coverage of Ag over the side walls which cannot be obtained by conventional normal deposition method. Note that the bottom Ag nanostrip antennae, ~18 nm thick, are also visible in the images.

We measured the normal transmittance of the Ag nanohole arrays by using a visible range, microscope-based optical setup with circularly polarized incident light from the substrate side.32 The transmittance of left-handed (LCP) and right-handed (RCP) circularly polarized incident light, $T_{\text{LCP}}$ and $T_{\text{RCP}}$, for the Z-shaped Ag nanohole array in Fig. 2(c) exhibits broad responses consisting of multiple plasmon resonances excited in the meta-elements of the array at different wavelengths. Furthermore, the prominent feature of large transmission difference, defined as $\Delta T = T_{\text{RCP}} - T_{\text{LCP}}$ and shown in Fig. 2(e), indicates that the Ag nanohole array has strong chiral behavior associated with the meta-elements of the array. The chiral behavior is further confirmed by the excellent anti-symmetry of the transmissions of circularly polarized light from the Z- and inverse Z-shaped Ag nanohole arrays as shown in Fig. 2.

To investigate the physical mechanisms for the resonances in the Ag nanohole arrays, we then performed numerical simulations using a commercial finite-integration time-domain algorithm (CST Microwave Studio) software to obtain the optical responses in the visible range. We used the dielectric constant of shadowing deposited Ag extracted following the procedures as reported earlier33 and also known values for the PMMA and ITO glass to calculate the optical responses (transmittance $T$, reflectance $R$, and absorption $A = 1 - T - R$) for the Ag nanohole arrays with parameters listed in Fig. 1 for circularly polarized light propagating along the Z direction from the substrate side.

The simulated transmittances of LCP and RCP in Figs. 3(a) and 3(e) for the Z- and inverse Z-shaped Ag nanohole arrays agree well for wavelengths below ~800 nm with the experimental results shown in Figs. 2(c) and 2(d) except that prominent resonances can now be identified clearly in the simulations. Note that there are differences between the simulations and experiments for wavelengths ≥800 nm in terms of bandwidths and amplitudes, due possibly to non-ideal dimensions/parameters used in the simulations because of the cell-to-cell variations of the fabricated Ag nanohole arrays, in addition to the variations of the deposition angles.
and the non-prefect circularly polarized incident light at long wavelengths. The reflectance and absorption spectra for the Z-shaped Ag nanohole array are shown in Figs. 3(b) and 3(c), respectively, in which multiple resonances are much more obvious. For example, two reflection dips are clearly observed at ~620 and 740 nm for RCP incidence (blue curve in Fig. 3(b)) whose spectral positions correspond well to the absorption peaks, indicated by blue arrows, as shown by the blue curve in Fig. 3(c). It is noteworthy that for any magnetic resonator, each magnetic dipolar mode manifests itself as a distinct peak in the absorption and a dip in the reflection. As a result, it is expected that these corresponding peaks and dips are originated from the pronounced magnetic dipolar resonances. For LCP incidence, similar spectral behavior of multiple resonances is observed with two dramatic absorption peaks at ~620 and 850 nm as indicated by red arrows in Fig. 3(c), corresponding well to the positions of two reflection dips of the red curve in Fig. 3(b), respectively. The absorption peaks reflect strong couplings between the meta-elements and the incident electromagnetic fields. Moreover, the absorption (~0.6–0.7) is much larger than the transmission and reflection (~0.2) suggesting that radiative scattering is greatly reduced by the spatially arrangement of the meta-elements in the nanohole array.

The multiple resonant properties of the Z-shaped Ag nanohole array can be further characterized by the transmittance and absorption difference spectra calculated as \( \Delta T = T_{RCP} - T_{LCP} \) and \( \Delta A = A_{RCP} - A_{LCP} \), respectively. The \( \Delta T \) spectrum, blue curve in Fig. 3(d), exhibits a broad prominent dip at ~690 nm, which agrees very well with the experimental dip at ~680 nm in Fig. 2(e) and another smaller dip at ~560 nm which, unfortunately, is hard to be identified in Fig. 2(e). The \( \Delta A \) spectrum, red curve in Fig. 3(d), exhibits three pronounced resonance peaks around ~550, 650, and 740 nm where the absorption for RCP incident light greatly surpasses that of the LCP. Opposite spectral behaviors are observed for the inverse Z-shaped Ag nanohole array as shown in the right column of Fig. 3, revealing that the inverted U-shaped SRRs when arranged with a handedness governed by Z- or inverse Z- symmetry exhibit strong polarization dependence, enabling potential applications in ultra-sensitive enantiomer sensing.

To understand further the magnetic resonant behavior of the plasmonic meta-elements in the Z-shaped Ag nanohole array, the magnitudes of magnetic fields in X and Y directions as a function of incident wavelength on selected cutting planes are shown in column 1–4 of Fig. 4. The snapshots of current distributions for some of the resonances are shown in columns 5 and 6. The positions of the relevant cutting planes are also included in column 6. It is clear that, at resonances, strong magnetic fields are highly concentrated in the dielectric material in regions surrounded by the displacement currents in the coated Ag.

For LCP incidence, columns 1 and 2 in Fig. 4, strong magnetic fields are induced at ~750–900 nm and ~550–700 nm. They correspond well to the absorption peaks at ~850 nm and ~620 nm in Fig. 3(c). The current distributions in Figs. 4(A) and 4(B) show clearly that the broad response for ~750–900 nm is resulted mainly from the magnetic resonances of SRRs 1 and 2. Note that in Fig. 4(A), there is an additional resonance, labeled as 1’, excited along the side walls below the SRR 1 because of the long arm length of the SRR. The currents of resonance 1’ are almost equal and anti-symmetric which generate a magnetic dipole opposite to that of SRR 1. More importantly, very weak or no net electric dipoles are excited. As a result, the two magnetic dipole moments are relatively pure, resulting in significant anti-phase coupling between each other. Such a coupling behavior between the SRR 1 and the resonator 1’ can serve as a building block for new magnetic metamaterials in low loss magnetic transportation.

The resonance for ~550–700 nm is related to the current flows at the bottom Ag strip antennae and will be discussed further in the bottom.

For RCP incidence, columns 3 and 4 in Fig. 4, strong magnetic fields are induced at ~650–850 nm and ~600–750 nm, respectively, corresponding well to the absorption peaks at ~740 nm and ~620 nm in Fig. 3(c). Figure 4(C) shows that the hot spots in column 3 arise from the magnetic resonance of SRR 3, which is responsible to an ultra-broad range ~650–850 nm. The ultra-broad response of the magnetic resonance may come from first the inverted U-shaped SRR 3 has extended thickness ~100 nm along the x-direction (see Fig. 1(c), and similar for SRRs 1 and 2) supporting nanocavity-like resonance and second the highly-compact meta-elements of the nanohole array. Note that due to symmetry of the Z-shaped Ag nanohole array, the SRRs respond.

FIG. 3. Simulated transmittance, reflectance, absorption, transmission and absorption difference of Z- (left column) and Inverse Z- (right column) shaped Ag nanohole arrays for circularly polarized light under normal incidence.
differently for circularly polarized incident light: SRRs 1 and 2 by LCP and SRR 3 by RCP and thus produce the strong chiral behavior as observed in the experiment.

The inverted U-shaped SRRs explain mostly the prominent magnetic responses observed. However, these resonances alone cannot explain the high and broad absorptions of the Z-shaped Ag nanohole array as shown in Fig. 3(c). It is clear that strong magnetic fields are also induced at the Ag strip/ITO glass interface for both polarization incidence, labeled as resonator 4, as seen in columns 2 and 4 of Fig. 4. We attribute this strong magnetic field to the electromagnetic field enhancement of localized surface plasmon resonance at the metal/dielectric interface.38,39

Moreover, it is worth mentioning that magnetic hot spots are observed in dielectric regions surrounded by sharp corners/turns of the Ag nanohole array as indicated, for illustrations, by the current loops shown in Figs. 4(D) and 4(E). Even though the strengths of these resonances are weaker than those of the inverted SRRs and the bottom Ag nanostraps, the number of these hot spots is significant, leading to the additional broadening of the whole spectrum. These by-products demonstrate well the advantages of our shadowing vapor deposition method, which allows depositions onto the side walls of the Z-shaped hole, creating corners/turns joining orthogonally oriented metallic components which cannot be easily achieved in other methods.

It is noteworthy that another advantage of the Ag nanohole array is that far field radiation, usually very strong for electric resonances in planar meta-elements, is greatly reduced by the specific arrangement of the meta-elements at different levels of the array. First, dipole radiations from the bottom Ag nanostraps can be re-absorbed by the SRRs at the top. Furthermore, the displacement currents along the arms of the SRRs are mostly along the Z direction and thus do not couple to the incident electric field. Lastly, the displacement currents on the top Ag layer are associated with magnetic resonances in the SRRs and can only be excited in the inner surfaces of the metal layer, leading to only small electrical scattering. Hence, our design provides a new approach for energy concentration which may find applications in photovoltaics and plasmon lasers/emitters at subwavelength scales.

Finally, as mentioned above, the SRRs in the Z-shaped Ag nanohole array respond distinctively to LCP and RCP. The reason can be explained as follows: just like the helical path in a 3D helix, the displacement current induced by the incident light will be forced to move along the path dictated by the handedness of the helix.40,41 Thus, different regions have different responses to LCP and RCP.42 In our Ag nanohole array, SRR 3 is the most critical meta-element in generating a strong chiral response in the visible range due to its strong sensitivity to RCP than LCP from 600 to 800 nm. After 800 nm, the polarization sensitivity will be dominated by SRRs 1, 2 and resonance 1 as shown in column 1 of Fig. 4 for LCP incidence. In short, the dramatic polarization selectivity of a particular meta-element allows ultrasensitive enantiomer sensing at the single molecule level.

In conclusion, ultra-broadband optical magnetism is realized by incorporating different plasmonic inverted U-shaped meta-elements in Z- or inverse Z-shaped Ag nanohole arrays by using the shadowing vapor deposition method. The broadband spectral behavior arises from excitations of multiple magnetic resonances in the meta-elements of the arrays. The meta-elements exhibit great sensitivity to LCP and RCP incident light due to the chiral configuration of the nanohole array. Our Z- and inverse Z-shaped Ag nanohole arrays are instructive for understanding the broadband nature of artificial magnetism in the visible range and have potential applications as the broadband metamaterial absorber and biosensor.
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