Symmetric and anti-symmetric magnetic resonances in double-triangle nanoparticle arrays fabricated via angle-resolved nanosphere lithography

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We report experimentally that for a particular high-symmetry planar periodic arrangement of metal double-triangle nanoparticle arrays fabricated via angle resolved nanosphere lithography, both anti-symmetric and symmetric magnetic resonances can be explicitly excited at off-normal incidence. Further, we demonstrate that the underlying mechanism for the formation of these two modes is a result of direct interactions with the incident electric and magnetic fields, respectively. As a consequence, with increasing the incident angle there is a relatively small blue-shift in the transmission for the electric-field induced anti-symmetric mode, while a remarkable red-shift is observed for the magnetic-field induced symmetric mode. Copyright 2011 Author(s). This article is distributed under a Creative Commons Attribution 3.0 Unported License. [doi:10.1063/1.3655439]

Metallic split-ring resonators (SRRs) first proposed by Pendry et al. have attracted intensive attention in the last decade mainly due to its unprecedented electromagnetic properties, such as a negative effective permeability near the resonant frequency. 1 By combining an array of such artificial atoms with a periodic array of metallic rods, a new class of optical materials known as metamaterials has been experimentally demonstrated to exhibit negative refractive index in the microwave regime. 2 After that, metallic SRRs have experienced tremendous progress in shifting the resonant response to higher frequencies, 3–7 and have been widely used as a building block for metamaterials. Recently, coupling between neighboring SRRs has been proven to play a critical role in the response of such metamaterials. 8 The importance of the coupling has been already demonstrated for different types of elements and relative orientation between them, for example, planar arrays of SRRs, 8, 9 SRR pairs with internal twisted angles 10, 11 and laterally stacked SRRs, 12, 13 and interesting coupling effects have been observed, like spectral shift or splitting of the single SRR magnetic resonance. Most of these researches focus only on the case of normal incidence, in which only the electric field component initially drives the magnetic response of the SRRs. 5–13 Since the magnetic resonance of SRRs is highly anisotropic, 14 it is important to investigate the optical response of SRRs under oblique incidence of light. Recently, a polarization and incident angle independent planar THz metamaterial with high Q-factors consisting of two concentric ring resonators with interdigitated fingers placed between the rings was experimentally demonstrated. 15 So far, however, still little attention has been paid to the case of oblique incidence. 4, 15–18

In this paper, we investigate magnetic resonances of double-triangle nanoparticle arrays at off-normal incidence when light polarization is oriented such that both its electric and magnetic fields simultaneously drive the constituent magnetic response. The SRR-like metal nanostructure arrays are fabricated using our recently reported method 18 that is based on angle-resolved nanosphere
FIG. 1. (color online) (a) Scanning electron microscope image of Au double-triangle arrays on a glass substrate. The whole structure can be reproduced by placing a unit cell consisting of two double-triangles with opposite openings (indicated by blue solid-line boxes) on a triangular lattice with two primitive vectors $a_1$ and $a_2$. Note that in our simulations the calculation domain (indicated by red dashed-line box) contains one complete and four quarter unit cells. (b) Schematic view of the incident light polarization configuration in the present studies with respect to the unit cell. (c) Experimental and simulated transmission spectra at normal incidence $\theta = 0^\circ$. (d) Current and magnetic field ($H_z$ component) distributions at the resonance of $\lambda = 1730$ nm.

We show that for such a polarization configuration both symmetric and anti-symmetric magnetic modes can be explicitly excited. The underlying mechanism for the formation of these two modes is a result of direct interactions with the incident electric and magnetic fields, respectively. As a consequence, with increasing the incident angle there is a relatively small blue-shift for the electric-field induced anti-symmetric mode, while a remarkable red-shift is observed for the magnetic-field induced symmetric mode.

Figure 1(a) shows the scanning electron microscope image of a particular high-symmetry planar periodic arrangement of double triangles fabricated using our recently reported method\textsuperscript{18} that is based on angle-resolved nanosphere lithography.\textsuperscript{19} In brief, large area single-domain two-dimensional (2D) colloidal crystals are first assembled from monodisperse polystyrene colloids with a diameter of $D = 1.0$ $\mu m$ via a self-organization process confined in a wedge-shaped cell.\textsuperscript{20} Thin gold layers (20 nm) are deposited through such a colloidal crystal mask onto a glass substrate at two oblique deposition angles $\pm 7.5^\circ$, respectively. Then, the colloidal mask is peeled off and an array of gold double-triangles is prepared. As indicated by solid-line boxes in Fig. 1(a), each unit cell consists of two double-triangles with opposite openings and thus has two-fold rotational symmetry and two mirror planes ($D_{2h}$ point group), which is different from the usual arrangement of SRRs with the $C_{1v}$ point group (the unit cell contains a single SRR and has one-fold rotational symmetry and one mirror plane).\textsuperscript{4–7, 16} The whole structure can be reproduced by placing the unit cell on a triangular lattice with two primitive vectors $a_1 = D/2 + \sqrt{3}D/2$ and $a_2 = -D/2 + \sqrt{3}D/2$. Figure 1(b) schematically shows the incident light polarization configuration in the present studies with respect to the unit cell. Experimentally, all the transmission spectra are taken using a commercial Fourier-transform infrared spectrometer equipped with a polarizer. As shown in Fig. 1(c), a dip around $\sim 1780$ nm was observed in the measured transmission spectrum at normal incidence (incident angle $\theta = 0^\circ$).
FIG. 2. (Color online) (a) Experimental and simulated transmission spectra of the high-symmetry periodic arrangement of Au double-triangles at off-normal incidence (incident angle $\theta = 30^\circ$). (b) and (c) Current and magnetic field distributions at resonances $\lambda_1 = 1715$ nm and $\lambda_2 = 1850$ nm, respectively.

To understand the measured response, numerical simulations are carried out using a Finite Element Method (Comsol Multiphysics). The calculation domain, indicated as a dashed-line box in Fig. 1(a), constitutes one complete and four quarter unit cells. Periodic boundary conditions are applied to the four sides of the rectangular calculation domain. In the modeling, a uniform 20 nm thickness is assigned to the double-triangle including its overlapped region. The dielectric constant of the glass substrate is taken as 2.25. The permittivity of gold is described by a Drude model with plasma frequency $\omega_p = 2.175 \times 10^{15}$ Hz and collision frequency $\omega_c = 6.5 \times 10^{12}$ Hz. Our numerical simulation [Fig. 1(c)] has confirmed the presence of the transmission dip observed in the experiment. It is seen from the current and magnetic field ($H_z$ component) distributions [Fig. 1(d)] that the circular currents and thus magnetic dipoles are excited in each of the double-triangles. Therefore, the gold double-triangle can be viewed as an “artificial magnetic atom”.

When light impinges at off-normal incidence [Fig. 1(b)], the vertical component of its magnetic field ($H_z$ component) acquires the gold double-triangles and is ready to drive the magnetic response. As shown in the experimental transmission measured at the incident angle $\theta = 30^\circ$ [Fig. 2(a)], two transmission dips are observed at the wavelength of $\lambda_1 = 1715$ nm and $\lambda_2 = 1850$ nm, respectively. This result is somewhat surprising since in the previous reports only one magnetic resonance is observed for usual periodic SRR arrays under the same polarization configuration. The presence of these two dips is well reproduced in the numerically evaluated transmission spectrum [Fig. 2(a)], where the remaining discrepancies most likely arise from our simplified model for the nanostructure and the fabrication tolerances in the experiments, e.g. the resolution in controlling the deposition angle which could result in size difference of the triangles or misalignment along the triangle bases [see Fig. 1(a)].

To clarify the origin of these two resonances, current and magnetic field distributions at the corresponding resonances are calculated. As shown in Fig. 2(b), for the resonance wavelength $\lambda_1 = 1715$ nm the circular currents induced in each of the two double-triangles are oppositely wound, and consequently the two excited magnetic dipole moments are aligned anti-parallel (named as anti-symmetric mode). A quite different situation is found for the resonance wavelength $\lambda_2 = 1850$ nm [Fig. 2(c)], in which the circular currents and magnetic dipole moments excited in the two double-triangles oscillate in-phase (named as symmetric mode).

Although the observed spectral characteristic is similar to those observed in stereo-SRR dimers or planar 90°-rotated SRR pairs in which an inductive coupling between two SRR elements is responsible to the splitting of magnetic resonance, we argue that in our case the anti-symmetric and symmetric magnetic modes are excited by the incident electric and magnetic fields, respectively. To understand this mechanism, it is instructive to separate the contribution of the incident electric field from that of the magnetic field. Let us first consider the contribution of the electric field. It
FIG. 3. (Color online) Experimental (solid symbols) and simulated (lines with open symbols) resonance wavelengths taken from transmission spectra as a function of the incident angle. Experimental data are measured from $\theta = 0^\circ$ to $\theta = 50^\circ$ with a $10^\circ$ step. Note that at incident angle $\theta = 10^\circ$ the symmetric mode is too weak to be extracted from the measured transmission spectrum.

is known that when the incident electric field is parallel to the SRRs base, it can couple to the capacitance of the SRRs and induce ring currents in the SRRs. The induced ring currents along the SRR base should have the same vector direction as the incident electric field. For the same reason, ring currents can be induced in double-triangles by the incident electric field in our case. Because the two double-triangles in a unit cell have opposite openings [Fig. 1(b)], the induced ring currents in these two double-triangles must circulate oppositely, which results in two anti-parallel magnetic dipoles oriented perpendicular to the sample plane. This is exactly consistent with the simulated resonant current and magnetic field distributions shown in Fig. 2(b).

On the other hand, when the incident magnetic field has a component normal to the SRRs plane, it can couple to the inductance of the SRRs and also excite the magnetic resonance. According to the Faraday’s law, the induced current loops in the SRRs should create magnetic dipoles against the buildup of the magnetic field. Because the buildups of the magnetic fields in our case are same, two magnetic dipoles in two double-triangles in the same unit should be aligned parallel. Again, due to the opposite openings of the two double-triangles in a unit cell, the circular currents in these two double-triangles must wound in the same direction, which obviously agrees with the simulated result shown in Fig. 2(c). From the above analysis, we can see that the formation of these two magnetic modes is a result of direct interactions with the incident electric and magnetic fields, respectively.

To further strengthen the above interpretation of the formation of the two magnetic resonances and to determine their dependencies upon the incident angle, we measured and simulated transmission spectra for other incident angles. The resonant transmission wavelengths are collected and plotted in Fig. 3 as functions of the incident angle. With increasing the incident angle from $\theta = 0^\circ$ to $\theta = 50^\circ$ the resonance wavelength of the anti-symmetric mode almost does not change, while the symmetric mode shows a remarkable red-shift. Such different angle-dependencies for the two magnetic resonances are indeed expected as a direct deduction of our above analysis. Similar to the intuitive dipole-dipole interaction model used for planar SRR arrays, we substitute each double-triangle by two dipoles: an electric dipole oriented parallel to the incident electric field and a magnetic dipole oriented perpendicular to the double-triangle. In the present polarization configuration [Fig. 1(b)], since the incident electric field is always parallel to the double-triangle base, its effective strength (defined as a component that can drive the magnetic resonance) does not change with the variation of the incident angle. This indicates that not only the strength of the induced electric and magnetic dipoles but also the coupling strength between them are constant at any incident angles. Hence, the electric-field induced anti-symmetric mode is anticipated not to undergo any changes with varying the incident angle. The observed slight blue-shift is possibly caused by the phase difference between the two double-triangles, which we have neglected in the dipole-dipole interaction model. In
contrast, with increasing the incident angle the effective component of the magnetic field becomes stronger, which correspondingly enlarges the strength of the electric and magnetic dipoles, and the coupling strength between them as well. Note that both the electric and magnetic dipoles in the same unit cell are only transversely coupled. It is also worth noting that for the symmetric magnetic mode two electric dipoles in the two double-triangles in a unit are anti-parallel, while two excited magnetic dipoles are aligned parallel [Fig. 2(c)]. The interaction of the anti-parallel oriented electric dipoles decreases the resonance frequency while the interaction of the parallel oriented magnetic dipoles tends to counteract this effect.\textsuperscript{8, 9} Since the interaction between the two double-triangles with opposite openings is dominated by dipole-dipole coupling,\textsuperscript{9} the magnetic-field induced symmetric mode is expected to occur red-shift with increasing the incident angle.

In conclusion, we report that symmetric and anti-symmetric magnetic resonances for a particular high-symmetry planar periodic arrangement of metallic double-triangles can be explicitly excited without the need of inductive coupling at off-normal incidence. We have argued that the anti-symmetric and symmetric modes stem from the incident electric and magnetic fields, respectively. Furthermore, we observe a remarkable red-shift for the magnetic-field induced symmetric mode and a relatively small blue-shift for electric-field induced anti-symmetric mode with increasing the incident angle which, in turn, verifies our proposed interpretations to the formation of two modes.

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