# Rendering dark modes bright by using asymmetric split ring resonators

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**Abstract:** We have studied both theoretically and experimentally symmetric and asymmetric planar metallic Split Ring Resonators. We demonstrate that introducing structural asymmetry makes it possible to excite several higher order modes of both even (l = 2) and odd (l = 3, 5) order, which are otherwise inaccessible for a normally incident plane wave in symmetric structures. Experimentally we observe that the even mode resonances of asymmetric resonators have a quality factor 5.8 times higher than the higher order odd resonances.

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#### 1. Introduction

Split ring resonators (SRR) have attracted a lot of attention in recent years due to their ability to exhibit negative permeability and even negative refractive index at optical frequencies [1,2]. The unique properties of these artificially engineered materials are a consequence of their design rather than the constituent materials and have earned them the name *metamaterials*. SRR's utilize the constrained motion of free electrons in metals (plasmons) to achieve negative permeability. This constrained motion also gives rise to local field enhancement similar to a lightning rod effect but, unlike the later it occurs at resonant frequencies. Many works have exploited this effect for bio-sensing [3,4] nanolithography [5–7], lasing [8,9] and biomedical [10] applications.

Regretfully, the fundamental plasmon modes are generally broad, which limits the detection resolution in sensing applications. Recently, several groups have demonstrated that narrowing of plasmon resonances can also be obtained through diffraction coupling (Rayleigh anomaly) [11] and near field coupling (Fano resonances) through various shapes and forms

[12–15]. This approach becomes very difficult at optical frequencies, which demands very small dimensions. Another alternative route to bio-sensing consists of using the Raman signal of an analyte molecule enhanced by plasmonic nanoparticles [16–18]. Although this approach is generally superior in identifying the exact molecule compared to refractive index sensing, the complicated experimental setup requiring high power laser is a hurdle. One can also excite narrower resonances by breaking the symmetry of the nano structures and accessing the higher order resonances [12–15,19,20].

Recently we have witnessed a growing interest in identifying the role of symmetry breaking in SRR structures [19–24]. In particular, it has been demonstrated that, as a consequence of asymmetric components in the structures, higher order resonances with narrower line widths in the THz regime become accessible [20]. By gradually displacing the capacitive gap the authors of [20] Singh, et.al were able to excite higher order modes like l = 3, 5 modes besides the fundamental mode.

In this report we break the mirror symmetry of the SRR structures and explore these asymmetrical structures for the visible and Near Infra-Red (NIR) regime. We demonstrate that higher order odd modes l = 3, 5 can be excited in the structures and more strikingly, that even modes l = 2 becomes accessible for the asymmetric patterns. By designing the structures to have the fundamental resonances in the infrared we can have resonances with high Quality Factor (QF) in the near-infrared region close to the telecommunication wavelength.

## 2. Experimental details and simulation methods

A standard cover glass was coated with 40 nm thick Au and 3.5 nm Ag. A thin layer of negative resist - Hydrogen silsesquioxane (HSQ) was spin coated on top of the metal layers. The structures were written on HSQ using e-beam lithography. After developing the resist, the patterns in the metallic films were obtained by Xe ion beam etching. The fabrication process is similar to the one described in [14] by Verellen *et al.* Figure 1 shows the dimensions and the SEM images of the fabricated structures and the size of the whole array is 50 x 50  $\mu$ m.

Transmission spectra were measured by means of a Fourier Transform Infrared (FTIR) microscope (Bruker Vertex 80v + Hyperion). Two different sources were used to enable measurements in a broad spectral range; namely Tungsten source for wavelengths between 1  $\mu$ m and 2  $\mu$ m and Silicon Carbide rod (Globar<sup>TM</sup>) covering the range between 2  $\mu$ m and 6  $\mu$ m. The samples were illuminated by a plane polarized light at near normal angle (12°) by a 15x, N.A = 0.4 Cassegrain objective and collected in transmission by an identical objective. Liquid nitrogen cooled Mercury-Cadmium-Telluride (MCT) detector is used to detect the transmitted light.

The simulations were performed using a commercial Finite Difference Time Domain (FDTD) solver (Lumerical FDTD). The metal structures were modeled using an inbuilt multicoefficient model which uses an "extensive set of basis function to better fit the dispersion profiles that are not easily described by Drude, Debye or Lorentz models" [25]. The glass substrate is modeled with a constant refractive index of 1.4. For the sake of simplicity, losses of the substrate were not taken into account. This assumption is valid mostly for the wavelength range  $0.5 - 6 \mu m$ .



Fig. 1. (a-c) Schematics of the fabricated structures and their dimensions, (d-f) scanning electron microscope images of the fabricated structures.

## 3. Modal analysis of individual structures

To investigate the influence of asymmetry on the optical response, we have analyzed three different structures: a symmetric SRR structure as shown in Figs. 1(a) and 1(d), and two asymmetric structures which we will refer to as SRR + [Figs. 1(b) and 1(e)] and SRR + + [Figs. 1(c) and 1(f)]. The mirror symmetry of the SRR structure was broken in the case of SRR + by a perpendicular extension to the lower arm of the structure as shown in Fig. 1(b) and 1(e). The motivation to study SRR + + is twofold. On one hand SRR + + has the same degree of asymmetry as SRR + structures, but the longer length of the structure (SRR + + is 580 nm longer than the SRR + structures) increases the total cavity length for the oscillating plasmon and accentuates retardation effects, providing an additional ingredient to access higher order resonances. On the other hand, SRR + + has a well defined chiral design whose nonlinear optical properties have been intensively studied, although an in-depth investigation of the linear response is still lacking [5,26–29].

We have studied the various resonant modes of the individual structures by numerical analysis and also investigated the effects of placing them in an array. By means of the electromagnetic solver software we determined both, (i) the extinction spectra to locate the resonances in a broad spectral range and (ii) the electric field distribution at the resonance point to identify the type of mode associated to the resonances. The fields are calculated 20 nm above the structure. For clarity and without loss of generality we show field intensity distribution of z-component of the electric field ( $|E_z|^2$ ). As already pointed out by Rockstuhl *et al.* [30] it is possible to identify the order of the modes directly from the  $|E_z|^2$  maps. We prove the validity of our models subsequently by comparing simulations of a periodic array with direct experimental results.

# 3.1. Individual Split Ring Resonators (SRR)

First we analyze the individual SRR structure. The calculated extinction spectra and the absorption spectra for all the considered structures are summarized in Fig. 2. The SRR was

illuminated by a plane wave at normal incidence with polarization parallel to the gap between the arms of the structure (i.e. as indicated by the arrow in Fig. 3(a)). This polarization is the most sensitive to the mirror symmetry of the SRR structures [30]. Extinction spectra [Fig. 2(a)] are plotted for the entire simulation range. Absorption spectra [Fig. 2(b)] are only plotted between  $0.5 - 4 \mu m$  to bring the higher order modes in contrast which when plotted for the full range will be completely overshadowed by the fundamental resonance. And these modes have very low absorption cross-section and therefore are noisy. One can also notice small differences between the absorption peak position and the extinction peak position as one can expect. This difference is not significant, and for the sake of clarity and continuity between experiment and simulation we will address the modes by their extinction peak position and not the absorption peak position.

From the spectra [Fig. 2(a)] we can clearly observe two strong modes at 7.0  $\mu$ m ( $\bullet$ ) and 2.8  $\mu$ m ( $\bullet$ ) for SRR structures. A closer inspection evidences the presence of two more modes that are weak and overlap with each other at 1.06  $\mu$ m ( $\star$ ) and 0.76  $\mu$ m ( $\bullet$ ). These points are not anomalies but represent distinct modes in the SRR. Modes for each of the resonances can be assigned by looking at the near field intensity distribution ( $|E_z|^2$  as shown in Fig. 3) and counting the number of nodes (i.e. minimum intensity) [30]. The near field mapping at resonances Figs. 3(a) and 3(b) indicate l = l at 7.0  $\mu$ m ( $\bullet$ ), and l = 3 at 2.8  $\mu$ m ( $\bullet$ ). We observe from the mode profiles that the whole structure acts as a single optical cavity (i.e. it is not possible to describe these modes as the superposition of modes of subparts of the structure) giving rise to these mode profiles.

The weaker resonances at 1.06  $\mu$ m ( $\star$ ) and 0.76  $\mu$ m ( $\blacklozenge$ ) (Fig. 3(c) and 3(d)) are a result of combination of different modes. The SRR structure can be regarded as a combination of a bar of 1  $\mu$ m length with their long axis perpendicular to the incident polarization and two bars of 1  $\mu$ m length with their long axis perpendicular to the incident polarization. As we can see from Fig. 3(e) and f the resonance at 1.06  $\mu$ m ( $\star$ ) is a combination of l = 3 resonance of the vertical bar and l = 1 resonance of the horizontal bar. Since the resonances of the individual subparts occur at wavelengths close to each other (for excitation with vertical polarization) they are excited at the same time and they combine to form this resonance. We will label such combination of resonances as C (3, 1) resonance; "C" to indicate that the underlying resonance is a combination of resonances. Similarly we can see from Figs. 3(g) and 3(h) that the C (5, 3) resonance at 0.76  $\mu$ m ( $\bigstar$ ) is a combination of l = 5 resonance of the vertical bar and l = 3 resonance of the horizontal bar. We would also like to highlight, that in Figs. 3(e) and 3(h), the resonances are of same order (l = 3), but the charge distribution as evidenced from their respective field plot is different.



Fig. 2. The calculated (a) extinction and (b) absorption spectra of individual structures; SRR (green), SRR + (red), SRR + + (blue). The symbols are used to indicate similar modes. The same convention is used throughout the text.

#### 3.2 Individual Split Ring Resonator plus (SRR + )

The calculated extinction spectra is shown in Fig. 2 and the field intensity patterns  $(|E_z|^2)$  for SRR + are shown in Fig. 4. The two strong resonances observed at 7.8 µm ( $\bullet$ ) and 2.8 µm ( $\bullet$ ) correspond to l = 1 and l = 3 modes of the structure, respectively. The red shift of l = 1 and 3 resonances in SRR + compared to SRR can be accounted for by the increase in cavity length (SRR + is 500 nm longer than SRR) of SRR + structure.

The resonance at 2.0  $\mu$ m ( $\blacktriangle$ ) [Fig. 4(c)] is of particular interest. Unlike the other modes observed, it is an even order mode (l = 2). For light polarized parallel to the gap; the internal fields in the two arms of SRR should have opposite flow direction. If the lengths of the arms were equal as in the case of SRR it would force the structure to have a phase difference of  $\pi$  between the arms resulting in only odd number modes [30]. In the case of SRR + the arm lengths are not the same, the symmetry is therefore broken and allows one to have an in phase oscillation between the arms and consequently an even mode. Another interesting feature of this even mode is that it appears at a higher energy than the l = 3 mode. On closer observation it is possible to see that the upper arm of the structure is dormant and the active part of the structure is this "L" shaped structure. When simulations were performed for the "L" structure we see a similar even mode resonance also appears at 2.2  $\mu$ m [Fig. 4(g)]. This result indicates that the light sees a different structure at this wavelength.

The resonance at 1.59  $\mu$ m ( $\Delta$ ) correspond to the l = 5 mode, whereas the resonances at 1.08  $\mu$ m ( $\bigstar$ ) and 0.71  $\mu$ m ( $\bigstar$ ) [Figs. 4(e) and 4(f)] correspond to a combination of modes, similar to the SRR structure. As evidenced from Fig. 4(h) – 4(l) the resonance at 1.08  $\mu$ m ( $\bigstar$ ) is a *C* (*3*, *1*). This mode is a combination of l = 3 resonance of the vertical bar of 1  $\mu$ m and l = l resonance of the horizontal bar of 1  $\mu$ m. The resonance at 0.71  $\mu$ m ( $\bigstar$ ) is a *C* (*5*, *3*, *3*) mode, and is a combination of l = 3 resonance of the vertical bar of 1  $\mu$ m and l = 3 resonance of the horizontal bar of 1  $\mu$ m and l = 3 resonance of a vertical bar of 500 nm.

## 3.3 Split Ring Resonators SRR + +

Figure 2 shows the extinction spectra and Fig. 5 shows the field distribution for SRR + +. The resonance at 8.4  $\mu$ m (•) (Fig. 5(a)) represents the l = 1 mode. Since the length of the structure is longer than the SRR and SRR + , the resonances are red shifted, with respect to these structures. The next strong resonance as seen in the extinction spectra appears at 3.1  $\mu$ m ( $\blacktriangle$ ) and has a shoulder at 3.7  $\mu$ m ( $\blacksquare$ ).

Comparing with the SRR + structures we expect they represent the l = 3 and l = 2 modes. We can also see an l = 2 mode at 3.2 µm for the "LG" structure as shown in Fig. 5(g). However the clear field distribution of l = 3 and l = 2 mode as seen in the case of SRR + is disturbed due to the strong spectral overlap between the modes [Figs. 5(c) and 5(d)] and only the dominant l = 3 mode is visible in the field intensity plots. The resonance at 1.53 µm ( $\Delta$ ) [Figs. 5(d), 5(g)-5(h)] is a combination of l = 5 mode of SRR + structure and an l = 4 mode arising solely in the lower part of the SRR + + structure. The resonances at 1.06 µm ( $\bigstar$ ) and 0.71 µm ( $\blacklozenge$ ) [Figs. 5(e) and 5(f)] correspond to combination of modes similar to the SRR structure. As evidenced from Fig. 5(i) – 5(o) the resonance at 1.06 µm ( $\bigstar$ ) is a C (3, 1, 1) mode is a combination of l = 3 resonance of the vertical bar 1 µm, l = 1 resonance at 0.71 µm ( $\blacklozenge$ ) is C (5, 3, 3, 3) mode is a combination of l = 5 resonance of the vertical bar of 1 µm and l = 3 resonance of the horizontal bar of 1 µm, l = 3 resonance of a vertical bar of 500 nm and l = 3 resonance of a horizontal bar of 1 µm, l = 3 resonance of a vertical bar of 580 nm and l = 3 resonance of a horizontal bar of 500 nm.



Fig. 3. Field intensity distribution of the z component of Electric field ( $|Ez|^2$ ) of SRR at resonant positions (a) 7.0 µm, (b) 2.8 µm, (c) 1.06 µm and (d) 0.76 µm. (e - h) show the underlying modes which couple to produce the modes shown in c and d. The vertical arrow in Fig. 3(a) indicate the polarization of the incident plane wave.

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Fig. 4. Field intensity distribution of the z component of Electric field (|Ez|2) of SRR + at resonant positions (a) 7.8  $\mu$ m, (b) 2.8  $\mu$ m, (c) 2.0  $\mu$ m, (d) 1.59  $\mu$ m, (e) 1.08  $\mu$ m and (f) 0.71  $\mu$ m. (g – l) show the underlying modes which couple to produce the modes shown in c, e and f. The vertical arrow in Fig. 4(a) indicate the polarization of the incident plane wave.

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Fig. 5. Field intensity distribution of the z component of Electric field (|Ez|2) of SRR + + at resonant positions (a) 8.4 µm, (b) 3.7 µm, (c) 3.1 µm, (d) 1.53 µm, (e) 1.06 µm and (f) 0.71 µm. (g – p) show the underlying modes which couple to produce the modes shown in c –f. The vertical arrow in Fig. 5(a) indicate the polarization of the incident plane wave.

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#### 4. Periodic arrays and experimental validation

Bearing the characteristics of the surface plasmon modes of individual structures in mind, in this section, we will (i) distinguish the lattice modes from the intrinsic modes of the nano structures under study and (ii) study the influence of periodic array on the resonances of the individual nanostructure.

We studied SRR, SRR + and SRR + + structures in square arrays with a periodicity of 1.2  $\mu$ m (i.e. 200 nm inter-particle separations). Theoretical and experimental extinction spectra for SRR, SRR + and SRR + + structures are presented in Fig. 6. One can immediately observe very good agreement between the theoretical and experimental spectra for all the structures. The small shift in the peak positions between the experimental and theoretical data can be attributed to (i) Unavoidable variations of the fabricated sample not considered in simulation. This could also explain why certain peaks are shifted blue while some others are shifted red. (ii) Differences in the permittivity values used for calculations from the actual values. The difference in peak position between the resonances of individual structures and the periodic array can be attributed to the dynamics of the array [31].

Figure 6 shows the extinction spectra of the array for wavelength range between  $1 - 6 \mu m$ . Due to the heavy losses in the substrate for wavelengths above 6  $\mu m$  we were unable to measure the extinction in our experiments. However, we can simulate the wavelength region above 6  $\mu m$ , as the substrate losses were ignored during simulation. For the sake of clarity we have only shown in Fig. 6 only the wavelengths range  $1 - 6 \mu m$  for the calculated spectra as well.

By comparing with the extinction spectra of the individual structures the l = l modes can be undoubtedly assigned to the peaks in simulation (not shown in Fig. 6) at 6.6  $\mu$ m ( $\bullet$ ) for SRR structure, 7.3  $\mu$ m (•) for SRR + structure and 8.5  $\mu$ m (•) for SRR + +. Similarly, the *l* = 3 modes can be assigned for SRR structure at 2.8  $\mu$ m ( $\blacksquare$ ) and SRR + structure at 2.8  $\mu$ m ( $\blacksquare$ ). The line shape of SRR + + structure has a peak around 2.7 µm ( $\blacktriangle$ ) and a shoulder at 3.0  $\mu$ m ( $\blacksquare$ ). Surface plasmon mode corresponding to the shoulder at 3.0  $\mu$ m ( $\blacksquare$ ) is the l = 3mode. We also see the even mode for the SRR + structure at 1.8  $\mu$ m ( $\blacktriangle$ ) and for the SRR + + structure at 2.7  $\mu$ m ( $\blacktriangle$ ). It can be seen that this otherwise forbidden mode has a sharper line width compared to the odd (l = 1, 3) modes. In fact the even mode resonances have a quality factor (as extracted from the experimental extinction spectra by using a multi-peak Lorentzian fit) of 8.6 in SRR + and 7.4 in SRR + + structure and are 23% (SRR + ) and 580% (SRR + + ) sharper than their respective l = 3 mode. We could also see C (5, 4) mode in the SRR + + appearing at 1.9  $\mu$ m ( $\Delta$ ). To highlight the quality of the even mode resonance for refractive sensing we also simulated SRR + structures in a periodic array covered by materials of four different indices, starting from 1.33 (typical refractive index of water) and by increasing in steps of 0.02. And it resulted in a figure of merit  $(\delta \lambda \delta n)/fwhm$  [32] of 5.05 for the l = 2mode and 1.72 for the l = 3 mode, which again highlights the quality of the even mode resonance.

The sharp resonances seen in the calculated spectra (Fig. 6) for all the structures are known as Rayleigh Anomaly (RA) and appear around 1.2  $\mu$ m and 1.6  $\mu$ m due to the air-metal and the substrate-metal interface, respectively. A quick analytical calculation based on Kravets [11]

$$\lambda_{R_m}^{air} = \frac{a}{m} \left[ 1 \pm \sin(\theta) \right] \tag{1}$$

$$\lambda_{R_m}^{sub} = \frac{a}{m} [n_s \pm \sin(\theta)]$$
<sup>(2)</sup>

Where *a* denotes the periodicity, *m* is an integer,  $\theta$  the angle of incidence and *n<sub>s</sub>* denote the refractive index of the substrate.  $\lambda^{air}_{R_m}$  and  $\lambda^{sub}_{R_m}$  denote the two types of RA: air diffraction mode and substrate diffraction mode. Substituting the values for periodicity *a* =

1.2  $\mu$ m and  $\theta = 0^{\circ}$  we find that the air diffraction mode appears at 1.2  $\mu$ m and substrate diffraction mode appears at 1.68  $\mu$ m. In our simulations we observe them at 1.12 and 1.56  $\mu$ m for SRR arrays, 1.12 and 1.54  $\mu$ m for SRR + arrays and 1.13 and 1.55  $\mu$ m for SRR + + arrays. They correspond very well with the asymmetric peak seen at 1.06 and 1.66  $\mu$ m for SRR arrays, 1.07 and 1.63  $\mu$ m for SRR + and 1.06 and 1.65  $\mu$ m for SRR + + arrays seen in the experiment. These strong RA modes also overlap with the weak l = 5 modes observed in the individual structures around 1.6  $\mu$ m and the combination modes seen around 1  $\mu$ m, and could also be the reason for the small shifts observed in the experimental and simulated RA modes from the analytical calculations.



Fig. 6. Experimental and theoretical extinction spectra of periodic array of SRR, SRR + and SRR + + nano structures. SRR spectra are denoted in green (a,b); SRR + spectra are denoted in red (c,d); SRR + + spectra are denoted in blue (e,f). The orange squares denote the l = 3 mode, the blue triangles denote the l = 2 mode and the brown trapezoid denotes the C(5,4) mode. The two RA arising due to the air-metal interface and the substrate-metal interface are denoted as RA<sub>air</sub> and RA<sub>sub</sub>.

# 5. Conclusion

We have studied symmetric and asymmetric split ring resonator plasmonic structures experimentally and theoretically. We have analyzed the underlying modes and shown that it is possible to excite higher order modes (l = 3, 5). We have also shown that by breaking the symmetry we are able to access forbidden even mode (l = 2) in the SRR + and SRR + + structures both experimentally and theoretically. The quality factors of the odd higher order mode (l = 3) is higher than the fundamental mode (l = 1). In addition, the even mode (l = 2) in the asymmetric structures has an even higher quality factor than the higher order odd mode (l = 3). In SRR + + structures we report a 5.8 times increase in the quality factor of l = 2 mode compared to l = 3 mode. Using asymmetric split ring resonators thus provides an alternative way to get resonances with high quality factor.

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