Second harmonic generation from complementary triangular Au metamaterials

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Abstract. Sub-wavelength square array of a triangular Au platelet and its complementary structure (i.e. that of a triangular hole in a square film of Au) are compared in terms of second harmonic generation efficiency for fundamental light in the near infrared and visible region of spectrum for normal incidence. Electric field strength around the convex corners of a triangular particle is at least 10 times larger than the one around the concave corners of triangular hole in the complementary structure. Nevertheless the SHG intensity at the respective resonant frequency is found to be comparable, which are numerically estimated by an overlap integral of nonlinear polarization and electric field at the SHG frequency in the nonlinear optical scattering theory originally proposed by Roke et al. (Phys. Rev. B 70, 115106 (2004)). The reason is due to the large electric field strength at the sides of the triangular hole at the resonance frequency, which compensates the suppressed electric field at the concave corners in the overlap integral.

1. Introduction

Metamaterials (MMs) are artificial periodic or non-periodic structure, whose electromagnetic properties are due to both the subwavelength cellular architecture and chemical composition, which are not readily found in nature. The emergence of MMs has changed the spirit of optics, such that today, researchers are not just restricted to variety of natural materials, but rather, they now have the freedom to nanostructure metallic surface with any geometry to tailor optical properties at will. By nanostructuring a metallic surface with geometries that breaks inversion symmetry, not only that new features such as strong local near field enhancement and surface plasmon resonance start to emerge but also second harmonic (SH) response is achievable from such surface[1].

In recent time, there has been growing interest in understanding the relationship between the strong near field enhancement in MMs and their Second Harmonic (SH) response. So far SH generation from MM are mostly from Split Ring Resonators (SRRs) of different geometries, which are usually characterized with mixed corners (i.e. convex and concave corners)[2]. It is a known fact that the local field enhancement around convex corners of metal can be very large compare to other areas with zero curvature, which is due to the affinity of electric field lines to confine around geometric corners[1]. Depending on whether the corners are convex or concave, the level of enhancement can be different, but since SRRs is characterized with mixed corners, it is difficult to separate the effect of local field enhancement around each corners on SH response.
In this study, we separate the effect of local field enhancement at the convex and concave corners, by designing a complementary Au MM with triangular resonators (i.e. Au MM with triangular particle and Au MM with triangular hole), where the Au MM with triangular particle is characterized with three convex corners while Au MM with triangular hole is characterized with three concave corners. In this design the triangular particle is simply replaced by air in metal to have a complementary structure. The aim for this design is to confirm how the difference in local field enhancement around convex/concave corners in complementary Au MM with triangular resonators translates into high SH emission intensity, in an attempt to understand the optical origin of nonlinearity. To achieve this objective, we will employ numerical simulation based on the nonlinear scattering theory, which is a numerical method that has proven to be efficient in predicting nonlinear emission from non-centrosymmetric medium using microscopic linear response[3].

2. Methods
The linear responses of our complementary Au MMs with isosceles triangular resonators were numerically calculated using the CST microwave electromagnetic solver. A plane wave (polarized either x- or y- direction) propagating along the z- direction is considered as an excitation and the symmetry axis of the triangle is fixed in x- direction throughout this computation. In order to estimate SHG from the artificial structure, the nonlinear optical scattering method originally proposed by Roke et al [4] and later applied for MMs by O’Brien et al [3], is used in this work. The SH electric field is evaluated in MATLAB by the following surface integral:

$$E_{ijk}^{SHG} \propto \iint E_{i,n}(2\omega)\chi_{nnn}^{(2)} E_{j,n}(\omega)E_{k,n}(\omega)dS$$

where $E_{ijk}^{SHG}$ is the second order non-linear emission, $\chi_{nnn}^{(2)}$ is the local nonlinear susceptibility at the surface, and $E_{j,n}(\omega)$ and $E_{k,n}(\omega)$ are the near field of the excitation by fundamental wave and $E_{i,n}(2\omega)$ is the mode at second harmonic frequency normal to the surface of the nanostructure. The notation i, j and k correspond to the direction of second harmonic emission and two fundamental excitations, respectively. In this work, we focus on the case where j = k. Then the integral vanishes if i = y when the second harmonic mode is odd upon y-inversion for isosceles triangles. In general, the integral can vanish depending on the symmetry, which gives the selection rule. If the symmetry allows, the SHG intensity is determined by the overlap of SH electric field and nonlinear polarization at the surface. In order to make our calculation simple, only the component $\chi_{nnn}^{(2)}$ is considered, where n indicates the normal component to the surface, which is justified by a recent experimental result showing the dominance of this component for a nano-structured metal [3]. The overlap integral estimates the efficiency of the mesoscopic nonlinear polarization distribution to emit SHG field at the far field. The SHG emission intensity is evaluated by using the relation

$$I_{ijk}^{SHG} \propto |E_{ijk}^{SHG}|^2$$

3. Results and Discussion
The structure of considered complementary Au MMs with triangular resonators is defined by the base (b) and height (h) of the triangle and a unit cell size (L) as in Figure 1. In all cases, the thickness of the complementary Au MMs is 25 nm. In this work we consider the cases for $b = h = K; K = 150, 200, 250, 300$ nm and $L = 400$ nm. To mimic the experimentally fabricated samples we blend the vertex of a triangle with radius of 30 nm in all cases. In the following we call a MM with triangular platelets a “particle” and its complementary structure with triangular holes a “hole” for simplicity. Figure 1 shows transmission spectra of the complementary structures for different resonator sizes (K). The curves are shifted vertically for better visibility. The resonant wavelength of linear response is identified by evaluating the transmission as a function wavelength,
the resonant wavelength for different sizes of complementary particle and hole upon $x$-polarization excitation is as shown in Figure (1a and b), where we have the resonant wavelength of particle and hole shifting to longer wavelength with increase in size of the particle and hole. The same trend was observed upon $y$-polarization excitation (not shown). For larger particle size with $K = 250 \text{ nm}$ and $300 \text{ nm}$, two resonance dips (i.e. narrow and wide) emerged, which might be due to multipolar effect coming from the increment in size of the particle. More interestingly at every wavelength for all $K$ where we have wide resonant dip in particle, we always have a corresponding wide peak rather than a dip in hole at approximate wavelength, which should be due to the complementary nature of the two structures. Based on the identified resonance wavelength from the linear response of complementary particle and hole as shown in Figure 1, the optimum SH emission intensity was evaluated for $x$- and $y$-directions upon $x$ and $y$ polarization excitation at normal incidence using nonlinear scattering theory as defined in Equation (1).

**Figure 1.** Transmission spectra as a function of wavelength for different sizes of (a) Particle (b) Hole.

**Figure 2.** SH intensity as a function of SH wavelength with $K = 300 \text{ nm}$ for (a) Particle (b) Hole. Local field enhancement at optimum SH resonant wavelength as a function of position along the curve surrounding the surface upon $x$-polarization excitation for (c) particle (d) hole.

Figure 2 (a and b) shows the SH intensity as a function of SH wavelength for particle and hole respectively with the dimension as specified in the figure. Due to the inversion symmetry that is broken along $x$-direction of the particle and hole, SH intensity is only observed in the $x$-direction upon $x$ and $y$ polarization excitation as indicated by term $I_{xxx}$ and $I_{yyy}$ in Fig. 2 (a and b), with optimum SH wavelength for particle at approximately 467 nm for $x$ and $y$ polarization while the optimum SH wavelength for hole upon $x$ and $y$ polarization is 455 nm. In order to investigate local field enhancement, we plot the field amplitude along the surface of the particle and hole 5 nm away from the surface in Figure 2 (c and d). As is seen in the figure, the local field enhancement around the
convex corners are at least ten times more than that of concave corners. The same trend was observed for other sizes of the complementary particle and hole.

**Figure 3.** Optimum SH intensity for different sizes of resonators (a) in the x-direction upon x-polarization excitation. (b) in the x-direction upon y-polarization excitation. Optimum SH wavelength for different sizes of resonators (c) upon x-polarization excitation (d) upon y-polarization excitation.

Despite the large difference in enhancement around convex corners of a particle as compared to concave corners hole, the optimum SH intensity for particle and hole are found to be comparable for x and y polarization excitation at normal incidence as depicted in Figure 2 (a and b). A comparable optimum SH intensity is also observed for the other sizes of complementary particle and hole for x and y polarization excitation at normal incidence as depicted in Figure 3 (a and b) respectively. The optimum SH wavelength at which the comparable optimum SH intensity were recorded for the different sizes of particle and hole upon x and y polarization excitation is as shown in Figure 3(c and d) respectively. By comparing the optimum SH resonant wavelength for different sizes of particle and hole, we can obviously notice how close they are for both x and y polarization excitation. This closeness of optimum SH resonant wavelength can be related to complementary nature of the particle and hole structures.

**4. Conclusion**

In summary, we compared SHG from a Au MM with triangular particle and its complementary Au MM with triangular hole, with each having a convex and concave corners respectively. Evaluation of the local field enhancement at optimum SH resonant wavelength for the complementary structure confirms that local field enhancement around convex corners are far larger than the enhancement around concave corners. Despite the large difference in local field enhancement around corners of the complementary structure, their respective SH intensity are comparable upon x and y polarization excitation at normal incidence, which implies that near field enhancement at the corners are not always the major determinant of SH emission intensity from metamaterial, the major contributor to SH efficiency can be other part of the geometrical structure in concave cornered nanostructures.

**References**

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