Efficient Generation of Second Harmonic from a Kind of Nonlinear Magnetic Metamaterial Composite

Shang Sun[1] **and Shumin Xiao**[2]

[1]Key Lab of Micro-Optics and Photonic Technology of Heilongjiang Province
Department of Physics, Harbin Institute of Technology, Harbin 158001, China
[2]Department of Material Science and Engineering, Shenzhen Graduate School
Harbin Institute of Technology, Shenzhen 518055, China

Abstract— We present a numerical investigation of efficient generation of second harmonic from a kind of nonlinear magnetic metamaterial composite. The analysis is focused on a structure consisting of periodic arrays of paired thin silver strips with second-order nonlinear material filled in the gap. The sandwich shaped periodic structure exhibits remarkable advantage in the generation of second harmonic signal. We, first, make an introduction to this sandwich structure, summarize the linear resonant properties and then investigate the wavelength dependence of the field enhancement in the gap. Next, we analyze about second-harmonic generation (SHG) from composite structure doping with isotropic nonlinear material by comparing with that from single layer nonlinear material and several other conditions. The results figure out that the magnetic resonance property of this nonlinear metamaterial composite makes a great contribution to the energy localization and gives rise to strong inhomogeneous field inside the gap. As a result, the strength of SHG is almost two orders larger than that of a pure nonlinear layer with comparable dimensions. Finally, we change the material property from isotropic to anisotropic, and investigate the impact combining with field enhancement, field profiles and linear and nonlinear polarization considerations, showing that most of the fundamental-harmonic electric field in the gap polarize along y direction, which is the power giving rise to the second-harmonic generation of the composite. This work leads to deeper understanding of singular nonlinear response in metamaterials.

1. INTRODUCTION

Metamaterials are artificially structured materials, whose general electromagnetic properties are different greatly from those expressed by their individual inclusions. The scale of these composite structures is much smaller than the wavelength of interest and that makes the whole material macroscopically uniform. The electromagnetic properties can be designed at will, other than just rely on the materials exist in the natural world. Benefit from that, we can get new materials exhibiting exotic properties, such as invisibility [1] and negative refraction index [2, 3]. The study about metamaterials mostly focused on how to get all kinds of linear optical materials with interesting properties. However, utilizing metamaterials to enhance nonlinear optical response is another exciting research field. It was first reported on split-ring resonators (SRRs) [4]. Since metamaterials are always designed with considerable magnetic activity, which implies there are strongly inhomogeneous fields in them. And that gives rise to large enhancement of local field which will naturally benefit the nonlinear effects. Nonlinear metamaterial composites have been studied widely in the microwave and near-infrared regions of the spectrum [5–7]. People mainly adopt a homogenization model to describe the material, and get the closed-form expressions of nonlinear susceptibilities. And then the design of the nonlinear metamaterials can be realized [8]. Among all kinds of nonlinear optical phenomenon, the optical process that two photons in fundamental frequency convert to one photon in double frequency, namely second-harmonic generation (SHG) is the most studied. Recently, the anomalously enhanced optical transmission (EOT) through arrays of sub-wavelength apertures in metal films has attracted significant attention. Lots of researches about structures with nonlinear materials like $LiNbO_3$ or GaAs filled in the apertures have been made by many groups [9–13]. Besides, increasing the field enhancement brought by nano-antennas arrays is another promising way to improve the conversion efficiency of SHG [14–16].

In this letter, we introduce a nonlinear magnetic metamaterial composite, which is a structure consisting of periodic arrays of paired thin silver strips with second-order nonlinear material filled in the gap and that makes the periodic unit's cross section shapes like a sandwich, exhibiting remarkable advantage in the generation of second harmonic.

Figure 1 shows a 2D schematic of the conventional coupled silver strips which has been discussed in [17]. Each sandwich unit consists of a pair of thin silver strips with thickness t separating by an

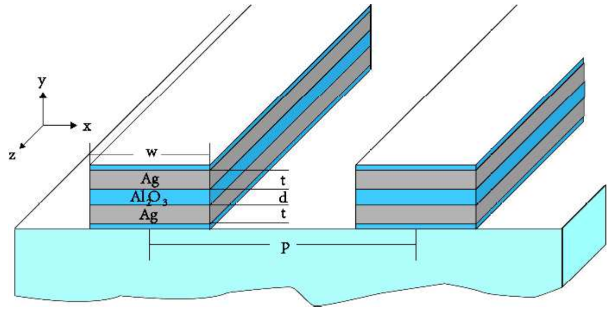

Figure 1: Structure of the coupled silver strips.

alumina layer with thickness d. The width of the sandwich stack is w, and the period is p. Two thin alumina layers of 10 nm are added to the top and bottom of sandwich stacks to make sure the fabrication is stable.

In our simulation, the analysis is performed using a FDTD model combined with analytical and numerical calculations, and the geometry parameters of the Ag-Al_2O_3-Ag structure are set to a specified size, with the thickness of each silver layer and alumina layer being $t = d = 30$ nm, the width of the stack being $w = 170$ nm, the period of the structure being $p = 400$ nm.

The schematic of the setup and coordinates in the model is presented in Figure 2(a). According to the study in Ref. [17], only under TM illumination, this structure could exhibit resonant properties. Therefore, we set the incident field in our model to be TM polarization with the magnetic field polarized along the strips and propagating toward the structure vertically. The experimental data of Johnson and Christy in the materials database is utilized to describe the optical properties of silver. Besides, the refractive index is 1.62 for alumina and 1.52 for the substrate.

The transmission, reflection and absorption spectra of our paired-strip structure simulated under TM polarization is plotted in Figure 2(b) with the electric resonance at $\lambda_e = 615$ nm and the magnetic resonance at $\lambda_m = 1040$ nm. Furthermore, as shown in Figures 2(c) and 2(d), the magnetic field distribution is calculated at these two characteristic wavelengths (λ_e and λ_m respectively) and the field magnitude is normalized to the one in the free space. The arrows in the figures represent the electric displacement. We can note that the electric displacement is mostly aligned along on one direction at the electric resonance. While around the magnetic resonance λ_m, the arrows present a loop shape and lead to an artificial magnetic moment, with a much stronger magnetic field inside

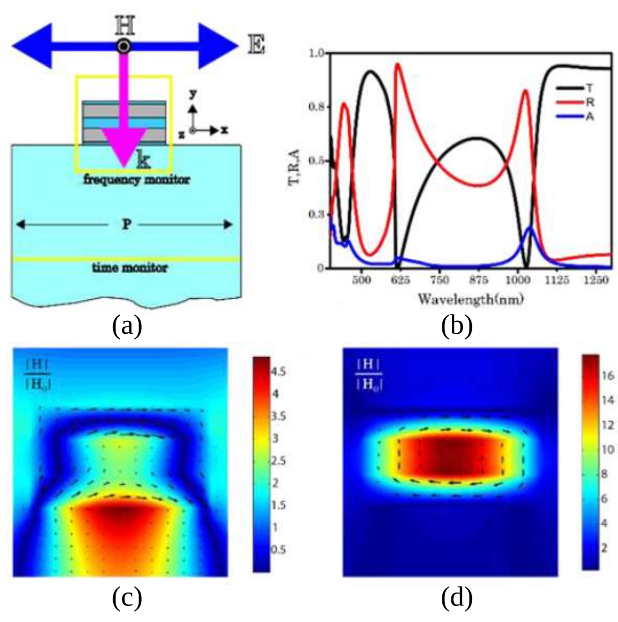

Figure 2: (a) Diagram of the model. (b) Simulated transmission (T) and reflection (R) and absorption (A) spectra of the structure for TM polarization. (c), (d) Magnetic field and the electric displacement at the electric resonance and the magnetic resonance.

the sandwich than the one at λ_e.

The strong magnetic activity at the magnetic resonance wavelength will naturally result in strong inhomogeneous fields in the stack, especially in the gap between the strips. The magnitude of the local field should be much larger than that in free space. To verify the large contribution from the magnetic resonance, we calculate the average strength of electric field and components in the gap illuminated by incident field of different wavelength, as shown in Figure 3.

In Figure 3, all the data are normalized to the magnitude of electric field in free space, which is presented as the green solid line for comparison. From the figure, we can see that curves show that all the ratios increase first and decrease after, with peaks existing around the magnetic resonance ($\lambda_m = 1040\,\text{nm}$). The average strength of electric field in the gap is more than ten times larger than that in free space, which is agreed to our expectation. Note that the two curves of total electric field and the y component almost coincide, whereas the x component stay very low, which means the polarization of the local field in the gap is mainly along y direction.

Since the optical nonlinearities are greatly dependent on the electromagnetic field, it's expecting to be strengthened by field enhancement. In our study, we predict doping the structure with nonlinear material in the gap would help get enhanced nonlinearity.

In order to make a better comparison with above work, our new nonlinear magnetic metamaterial composite keep all the settings same with before but the material in the gap between paired silver strips changed from alumina to an isotropic second-order nonlinear material with refractive index $n_d = 1.62$ and $\chi^{(2)} = 3 \times 10^{-10}\,\text{m/V}$ (an organic nonlinear optical crystal DAST can be qualified [18]). The amplitude of the incident source is set to be $1 \times 10^7\,\text{V/m}$.

First, we set the center wavelength of the fundamental harmonic (FH) source to be at the magnetic resonance 1040 nm to check if the second harmonic (SH) is generated. Figure 4(a) illustrates the SH (520 nm) electric and magnetic field distribution which are normalized to the FH ones in free space, and the arrows represent the electric displacement. It can be found that the SH electromagnetic energy is mostly concentrated in the gap with values cannot be ignored. We can also note that the second-harmonic generation result in two magnetic moments form between the paired

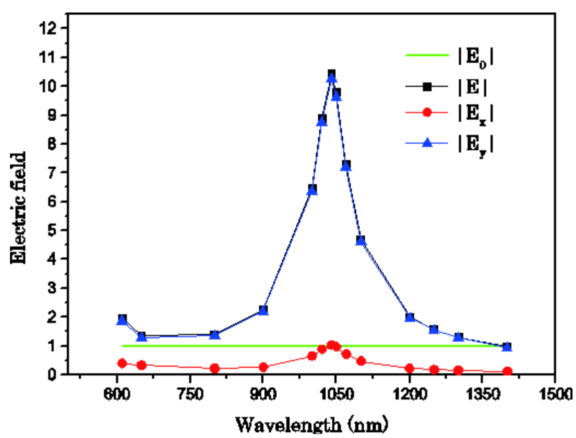

Figure 3: The dependence of the average magnitude of electric field and components on the wavelength of the incident field.

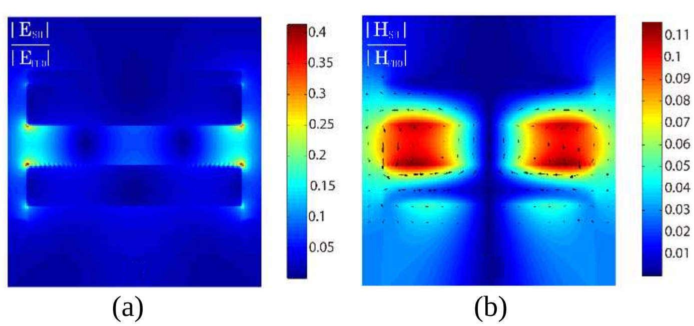

Figure 4: Simulated SH (520 nm) electric (a) and magnetic (b) field distribution with arrow representing the electric displacement.

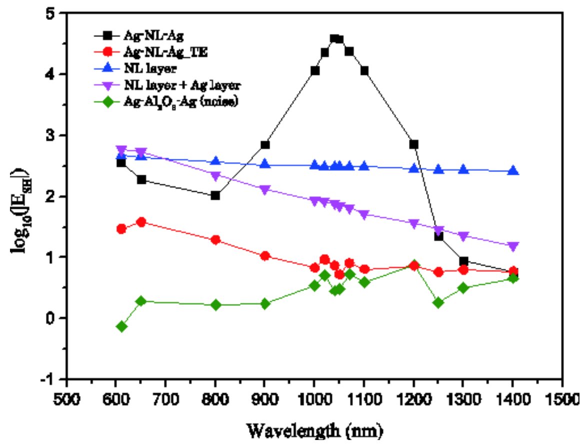

Figure 5: The average strength ($\log_{10}$) of SHG at the exit end of the model for different conditions.

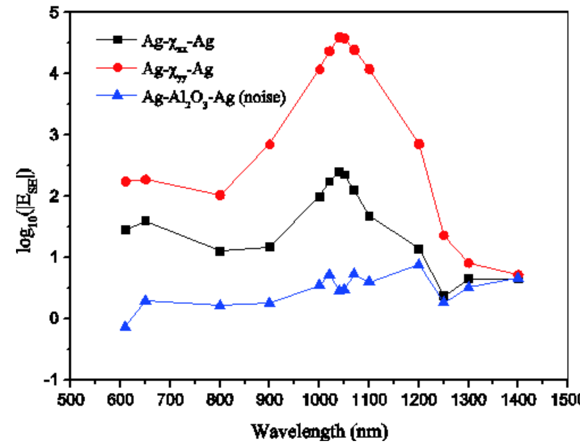

Figure 6: The average strength ($\log_{10}$) of SHG at the exit end of the model for two different anisotropic nonlinear composit.

silver strips.

For the purpose of verifying the advantage of our composite in the enhancement of the generation of SH, we make a comparison about the strength of SHG at exit end between different conditions, as shown in Figure 5. The figure is plotted by the wavelength of the incident FH source on the horizontal axis and the average SH electric field intensity ($\log_{10}$) at the exit end on the vertical.

All the data were calculated for TM polarization except the red ones. For the Ag-Al_2O_3-Ag structure, there are also SH signals existing in the numerical calculation, which are very weak and treated as noise in our analyses.

As expected, our composite (Ag-NL-Ag) shows great advantage for SHG around the wavelength of magnetic resonance (1040 nm). The SH electric field is almost two orders of magnitude larger than that from one single layer of nonlinear materials with thickness of 30 nm (NL layer) at exactly 1040 nm. The result for composite under TE polarization (Ag-NL-Ag_TE) is also presented in the figure, which is weak and display a non-resonant wavelength dependence over the spectrum range we study. Besides, the structure of one layer of nonlinear material covering one layer of silver (NL layer + Ag layer), both thickness of 30 nm, has been studied, which shows no significant benefits and gets even lower in SHG towards the longer wavelength.

For a better understanding of the generation of second-harmonic in this composite, we have made a simulation study about the structures filling with two different anisotropic nonlinear materials, assuming the second-order nonlinear susceptibility has only non-zero components of $\chi_{xx}^{(2)} = 3 \times 10^{-10}$ m/V and $\chi_{yy}^{(2)} = 3 \times 10^{-10}$ m/V respectively, as shown in Figure 6.

Thus it can be seen that it is the y-polarized FH electric field in the gap makes a large contribution to the SHG and give rise to a much better effect for structure Ag-χ_{yy}-Ag than Ag-χ_{xx}-Ag.

In summary, we have investigated the wavelength dependence of the field enhancement in the gap of paired silver strips and the SHG conversion efficiency from the structures composited with nonlinear material. We found that, the magnetic resonance property of the nonlinear metamaterial composite makes a great contribution to the energy localization and SHG, with the strength almost two orders of magnitude larger than that of a single layer of nonlinear material with comparable dimensions. We also show that most of the FH electric field in the gap polarized along y direction, which is the power giving rise to the second-harmonic generation of the composite.

REFERENCES

1. Schurig, D., J. J. Mock, B. J. Justice, et al., "Metamaterial electromagnetic cloak at microwave frequencies," *Science*, Vol. 314, No. 5801, 977–980, 2006.
2. Smith, D. R. and N. Kroll, "Negative refractive index in left-handed materials," *Physical Review Letters*, Vol. 85, No. 14, 2933, 2000.
3. Shalaev, V. M., "Optical negative-index metamaterials," *Nature Photonics*, Vol. 1, No. 1, 41–48, 2007.
4. Pendry, J. B., A. J. Holden, D. J. Robbins, et al., "Magnetism from conductors and enhanced

nonlinear phenomena," *IEEE Transactions on Microwave Theory and Techniques*, Vol. 47, No. 11, 2075–2084, 1999.

5. Shadrivov, I. V., A. B. Kozyrev, D. W. van der Weide, et al., "Nonlinear magnetic metamaterials," *Optics Express*, Vol. 16, No. 25, 20266–20271, 2008.
6. Zharov, A. A., I. V. Shadrivov, and Y. S. Kivshar, "Nonlinear properties of left-handed metamaterials," *Physical Review Letters*, Vol. 91, No. 3, 037401, 2003.
7. O'brien, S., D. McPeake, S. A. Ramakrishna, et al., "Near-infrared photonic band gaps and nonlinear effects in negative magnetic metamaterials," *Physical Review B*, Vol. 69, No. 24, 241101, 2004.
8. Poutrina, E., D. Huang, and D. R. Smith, "Analysis of nonlinear electromagnetic metamaterials," *New Journal of Physics*, Vol. 12, No. 9, 093010, 2010.
9. Fan, W., S. Zhang, K. J. Malloy, et al., "Second harmonic generation from patterned GaAs inside a subwavelength metallic hole array," *Optics Express*, Vol. 14, No. 21, 9570-9575, 2006.
10. Fan, W., S. Zhang, N. C. Panoiu, et al., "Second harmonic generation from a nanopatterned isotropic nonlinear material," *Nano Letters*, Vol. 6, No. 5, 1027–1030, 2006.
11. Vincenti, M. A., D. De Ceglia, V. Roppo, et al., "Harmonic generation in metallic, GaAs-filled nanocavities in the enhanced transmission regime at visible and UV wavelengths," *Optics Express*, Vol. 19, No. 3, 2064–2078, 2011.
12. Barakat, E. H., M. P. Bernal, and F. I. Baida, "Second harmonic generation enhancement by use of annular aperture arrays embedded into silver and filled by lithium niobate," *Optics Express*, Vol. 18, No. 7, 6530–6536, 2010.
13. Scalora, M., M. A. Vincenti, D. De Ceglia, et al., "Second- and third-harmonic generation in metal-based structures," *Physical Review A*, Vol. 82, No. 4, 043828, 2010.
14. Thyagarajan, K., S. Rivier, A. Lovera, et al., "Enhanced second-harmonic generation from double resonant plasmonic antennae," *Optics Express*, Vol. 20, No. 12, 12860–12865, 2012.
15. Park, S., J. W. Hahn, and J. Y. Lee, "Doubly resonant metallic nanostructure for high conversion efficiency of second harmonic generation," *Optics Express*, Vol. 20, No. 5, 4856–4870, 2012.
16. Bar-Lev, D. and J. Scheuer, "Efficient second harmonic generation using nonlinear substrates patterned by nano-antenna arrays," *Optics Express*, Vol. 21, No. 24, 29165–29178, 2013.
17. Cai, W., U. K. Chettiar, H. K. Yuan, et al., "Metamagnetics with rainbow colors," *Optics Express*, Vol. 15, No. 6, 3333–3341, 2007.
18. Marder, S. R., J. W. Perry, and C. P. Yakymyshyn, "Organic salts with large second-order optical nonlinearities," *Chemistry of Materials*, Vol. 6, No. 8, 1137–1147, 1994.