## Ultrafast effects in 3D Metamaterials

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**Abstract:** The nonlinear optical properties of metamaterials have attracted lots of attention in the last decade, particularly because it provides novel ways of tailoring optical functions which will be useful in achieving higher speeds for optoelectronics and other photonics circuits. In most of these metamaterials, the nonlinearity of significance lies within it metallic constituents. In this paper we show how extraordinary optical characteristics and ultrafast response can be achieved with use of a chiral gold based metamaterial. These nonlinear functions is realized when plasmonic excitations are generated by electromagnetics interactions, which are strongly enhanced within the metamaterial leading to situation where effective nonlinearity can be engineered significantly by changes in the geometrical structure of the components of the metamaterial, and not only by transitions in the electronic band levels of the material [1]

**Keywords:** Metamaterial, Third order nonlinearity

#### 1. Introduction

The ultrafast modulation of light with light has been a long sought goal for researchers and remains a difficult task, especially in the IR and Terahertz frequency region. This is essentially so because the optical nonlinearities in conventional nonlinear materials are generally too weak to alter the intensity of light significantly on a subwavelength scale. [2] With the emergence of metamaterials research, which have significant metallic content in the early 2000s, a significant approach to tackle this problem is to design nanostructure which will have higher optical nonlinearities which will be provided by its metallic constituents. The typical nonlinearities in these case are usually higher than those of

common semiconductor such as Si and GaAs that are popular in the optoelectronics industry. At the interface of between metals and dielectrics it often possible at particular angles of incident of light to excite a surface wave which consist of electron oscillations, known as plasmons which could be very significant in enhancing nonlinear effects.

The great challenge in recent years has been to engineer new materials that will be able enhance short pulses of light generated by modern laser systems, without suffering significant loss, such that these new effects can be probed. These new materials with nonlinear optical characteristics, could be used for optical limiting and switching, sum-frequency generation, difference frequency generation, four-wave mixing, and cross-phase modulation, and will lead to faster and more efficient optical devices [3].

The focus of our research has been to examine designs of metamaterials composed of cylindrical hollow gold/silver nanorods within an alumina dielectric matrix. We have considered two kinds nanorods arrangement, for a unit cell structure; a rectangular lattice and a hexagonal lattice.

With the availability of high power lasers systems, one can actually investigate these metamaterials with well characterized laser beams. It is possible to measure the effective optical nonlinearity of these materials. Nevertheless before fabricating, measuring and characterizing should be done, it is very needful to carry on a holistic computational modeling of the problem which will guide both the fabrication and characterization processes, and this computational task has been the focus of my project thus far.

These Metamaterials provide a unique way to enhance the electric fields produced by these laser beams, since the enhancement is critically dependent on the geometry of metamaterial [4]. This is why literature is filled with many kinds of geometry which could enhance electric fields in a different ways. In this work we have extended our calculations beyond a two dimensional (2D) geometry less computationally intensive problem, to analyze a potentially realizable three dimensional (3D) geometry metamaterial (MM) with gold (Au) as the plasmonic metal of choice.

## 2. Use of COMSOL Multiphysics

To address this problem we typically solve Maxwell's equation in the frequency domain to calculate the spectral characteristics of the metamaterial, using equation (1) below. We also solve the problem in the transient time dependent case to analyze the ultrafast effects of the metamaterial, using equation (2) below.

$$\nabla \times \frac{1}{\mu_r} (\nabla \times \mathbf{E}) - k_0^2 (\varepsilon_r - \frac{j\sigma}{\omega \varepsilon_0}) \mathbf{E} = 0$$
 (1)

$$\nabla \times \frac{1}{\mu_{r}} (\nabla \times \mathbf{A}) + \mu_{0} \sigma \frac{\partial \mathbf{A}}{\partial t} + \mu_{0} \frac{\partial}{\partial t} (\varepsilon_{0} \frac{\partial \mathbf{A}}{\partial t} - \mathbf{P}) = 0 \quad (2)$$

The polarization term is given by  $\mathbf{P} = \varepsilon_0 \chi^{(1)} \mathbf{E} + \varepsilon_0 \chi^{(3)} |\mathbf{E}|^2 \mathbf{E}$ . We ignore the second order term because metals are centrosymmetric, therefore the second order term is small. This polarization term is very critical for the transient nonlinear case. We have also used Comsol to solve for heat generated in these materials, but this was not our primary goal.

## 3 Results and Discussion

Based on some previous studies, the main task of in this current work is to examine properties in the linear regime, of proposed designs of Metamaterials (MM) which would guarantee high field enhancements needed for the excitation of nonlinear optical activity [5]. A novelty in our study this time was to introduce chirality in our design. A chiral structure is a structure whose mirror image cannot be superimposed on it. It has been reported in literature that chirality enhances nonlinear optical activity, and particularly polarization rotation which is well known, and these kind of nonlinear activity is strongly linked to the Kerr

effect and its corresponding third order susceptibility[4]. To make the structure chiral we introduced a gold hemispherical hump within another alumina matrix, normal to the axis of propagation which is the z-axis.

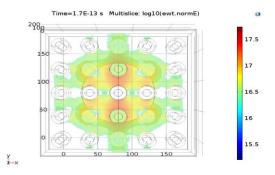


Figure 1: Shows the basic structure of the chiral metamaterial. We also notice very strong electric field couplings around the central nanorods. This result was obtained after a propagation time of 170[fs].

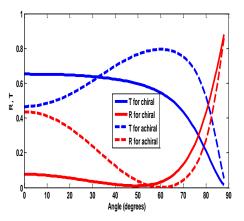


Figure 2: This figure shows the Transmission and Reflection versus angle of incidence for both achiral rectangular MM and the chiral rectangular MM at a wavelength of 800nm

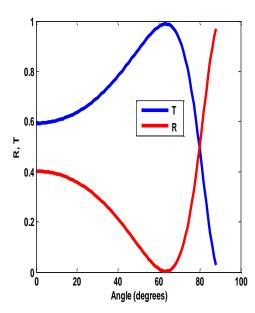


Figure 3: This figure shows the Transmission and reflection versus angle of incidence for the chiral MM at a wavelength of 2000nm.

From the figures above, we notice that there is a similarity in the profile for the achiral case at 800nm and the chiral case at 2000nm, as in both cases there is a maximum in Transmission and a minimum in Reflection at around 63 degrees. But the value of the Transmission at 2000nm in the chiral case rises to almost a value of 1. It is of interest to study these materials at longer wavelengths since there is a critical shortage of modulating devices in the infrared and terahertz regions.

For the achiral MM, when we varied the inner air hole radius from 4nm to 10nm, while leaving the outer air hole radius at a constant value of 16nm we obtained the result shown below for Transmission and Reflection.

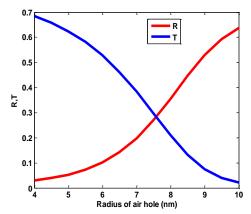


Figure 4: Shows the Transmission and Reflection versus the Radius of air, when the incident angle is 0, degree at an operating wavelength of 800nm, for the achiral MM.

The above results, gives us the necessary ideas we need, so as to optimize our designs for the applications of interest such as switching, and optical limiting. A study of the chiral structure, at zero degree incident angle, for various wavelengths of interest shows that the size of the hemisphere does not critically impact the Transmission, Reflection and Absorption. On the other hand if we look at the spectrum as seen in Figure 5, for the case where the radius of the hemisphere is 50nm we notice a maximum in absorption at about 759nm. The absorption later falls quickly to zero for longer wavelength. This result provides us with a good idea of how to further investigate the system.

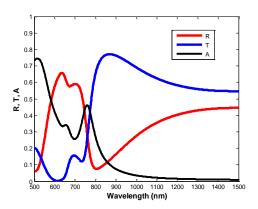


Figure 5: Transmission, Reflection and Absorption versus wavelength for a chiral MM, with a hemisphere radius of 50nm at zero degree incident.

The true nonlinearities can only be measured experimentally, with a pump probe experiment or a Z-scan experiment. Yet in order for us to do that we need to figure out with these initial calculations ways to increase these nonlinear ultrafast effects. It will be very needful for these devices to function at low intensities. Typically it takes intensities in the 1-100GW/cm² range to excite significant nonlinearities in MM. There several experiments lately that show significant excitations with intensities of the order 1MW/cm² [1, 6]. It will be good to be able to realize these ultrafast effects at lower incident light intensities

In our initial transient calculations we incident a beam of pulse width 20[fs] which has an intensity at focus of about 100MW/cm^2. Then we look at exit power leaving the MM it case where no nonlinearities are excited and in the case where nonlinearities are excited. In literature there are some many different values of these nonlinearities. Typically for Au it is the order of  $10^{-18}$  m<sup>2</sup>/V<sup>2</sup>, yet there are conditions where these values can be increased, because of chirality and order nano-mechanical forces to values of about  $10^{-12}$  m<sup>2</sup>/V<sup>2</sup> [4, 6,7]. We obtained results of the exit power as function of time as shown in Figure 6 below.

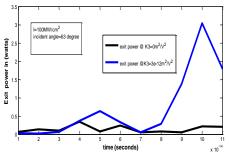


Figure 6: Shows the exit powers as function of time. The blue line represents the case when nonlinear effects are present, and the black line represents the case when, no nonlinearities are exited.

#### 4. Conclusion

We have shown how, nonlinear effects could be designed by changing several physical parameters such as cylinder, radius, and designing for chirality. We also notice, that our studies provides a significant way to approach the problem of

design nonlinear devices which will function in the deep into the IR.

Experimental results are expedient at this point to guide the theoretical frame work we have developed and to give us the actual range of values of nonlinearities we can observe with the materials we have chosen. This is necessary because we can chose to explore other materials such as carbon nanotubes and graphene which have been shown to enhanced plasmonic nonlinearities [2].

#### 5. References

[1] Neira, A.D. et al. Eliminating material constraints for nonlinearity with plasmonic metamaterials. Nat. Commun. 6:7757 doi: 10.1038/ncomms8757 (2015).

[2] A. Nikolaenko, N. Papasimakis, A. Chipouline, F. Angelis, E. Fabrizio, and N. Zheludev, "THz bandwidth optical switching with carbon nanotube metamaterial". OPTICS EXPRESS 6068, Vol 20 No 6 March 2012. [3] Martti Kauranen, and Anatoly V. Zayats S. "Nonlinear Plasmonics", Nature Photonics 6, 737-748 November 2012 [4] Ren, M. et al. Giant nonlinear optical activity in a plasmonic metamaterial. Nat. Commun. 3:833 doi: 10.1038/ncomms1805 (2012). [5] P. Evans and N. Katte "Numerical study of Chemically Synthesized 3D Nonlinear Metamaterials", Proposal Submitted to Department of Energy office of Science. January 2014.

[6] Jun-Yu Ou, E. Plum, J. Zhang, and N. I. Zheludev 2015 "Modulating light with light via giant nano-opto-mechanical nonlinearity of plasmonic metamaterial" arxiv.org/pdf/1506.05852

[7] Zhu, Y., Hu, X.Y., Fu, Y.L., Yang, H. & Gong, Q.H. Ultralow-power and ultrafast alloptical tunable plasmon-induced transparency in metamaterials at optical communication range. Sci. Rep. 3, 2338; DOI: 101038/srep02338 (2013).

# 6. Acknowledgements

This work was supported in part by the U.S. Department of Energy, Office of Science, Office of Workforce Development for Teachers and Scientists (WDTS) under the Visiting Faculty Program (VFP).