Cascaded Plasmon Resonances for Enhanced Ultrafast Nonlinear Optical Switching

Pieter G. Kik[1,2]
[1]CREOL , The College of Optics and Photonics, University of Central Florida, 4000 Central Florida Blvd, Orlando, FL 32816
[2] Department of Physics, University of Central Florida, 4000 Central Florida Blvd, Orlando, FL 32816
Corresponding author email: kik@creol.ucf.edu phone: (407) 823-6800, fax: (407) 823-6880

Abstract: Surface plasmon resonances on metallic nanostructures produce strongly enhanced third order nonlinear susceptibilities, while adding linear loss. We show that the use of cascaded resonances enables improved figures of merit for nonlinear switching.

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1. Introduction

Ultrafast optical switching based on Kerr-type nonlinearities typically requires large irradiance and long interaction lengths in order to achieve sufficient nonlinearly induced changes in optical path length or extinction. Currently, a large amount of work focuses on the use of surface plasmon resonances in nanostructured metamaterials in order to increase the nonlinear response. While effective media containing isolated plasmon resonant nanoparticles have demonstrated significantly enhanced $\chi^{(3)}$ response near the plasmon resonance wavelength, the introduction of linear loss by the plasmon resonance poses a challenge to the use of this enhanced response in applications. Consequently, the search is on for materials that can produce enhanced nonlinear susceptibility while maintaining acceptable linear loss. Early work focused on the use of regular arrangements of silver nanoparticles of a single size, and looked at the balance of the nonlinear response as a function of in-plane nanoparticle arrangement.[1] The study demonstrated that particle arrangements with reduced inter-particle spacing produce to stronger field enhancement and confinement. Despite the fact that the field enhancement occurred in a reduced volume, a larger nonlinear optical response relative to the linear absorption was predicted, indicating improved performance. An additional method for improving the performance of nonlinear refractive and absorptive materials is the utilization of cascaded plasmon resonances. An early investigation of a cascaded plasmonic nanoantenna or 'nanolens' simulated the field enhancement around a triplet of silver nanospheres with stepwise reduced volume.[2] It was demonstrated that a dramatically enhanced field could be obtained on the smallest of the particles through stepwise enhancement of the fields in a cascading process. Subsequent work investigated the use of a similar structure for second harmonic generation.[3] Given the fact that cascaded resonant structures provide enhanced field concentration and amplification, cascaded metamaterials that contain cascading elements could prove an effective design for nonlinear optical switching composites.

2. Methods

The investigation of the balance between linear and nonlinear optical response and the resulting figures of merit (FOM) for nonlinear optical switching is carried out through a combination of numerical modeling in order to obtain linear electromagnetic field distributions in a metamaterial of choice, followed by the determination of effective medium properties through an integral method.[1] In the present work metal-dielectric composites are considered consisting of regular arrangements of silver nanospheres in a host material with a refractive index of 1.5. The system is excited through plane wave illumination at normal incidence at frequencies near plasmon resonances of the composite. Three-dimensional frequency dependent electric field distributions are obtained using a numerical solver and subsequently converted to a linear absorption spectrum of each structure. A similar analysis is carried out in order to estimate the third order nonlinear susceptibility enhancement values $g^{(3)}$ of the structure based on the same electric field distributions, as described in detail in Ref. 1. The latter approach yields a prediction of the nonlinear response of the composite $\chi_c^{(3)}$ through the relation $\chi_c^{(3)} = \Sigma\, f_i\, g_i^{(3)}\, \chi_i^{(3)}$ where f_i is the fill fraction of constituent material i, $g_i^{(3)}$ is the calculated third order susceptibility enhancement factor of material i, and $\chi_i^{(3)}$ is the presumed Kerr-type third order nonlinear susceptibility of material i. Literature values are assumed for the optical properties of the constituent materials.

3. Results and Discussion

Figure 1(a) shows the simulated field enhancement (thin black line) in the center of a silver nanoparticle that is embedded in a regular array of silver nanoparticles with a 20 nm diameter, separated by 30nm center-to-center along the x-direction, and 140nm along the y-direction, with an assumed fill fraction of 1%, excited using an x-polarized plane wave. A clear plasmon resonance is observed at a wavelength of 445 nm with a moderate field enhancement. The thick blue line in Fig. 1(a) indicates the corresponding result if the particle size is alternated along the x-direction, with adjacent particles having a diameter of 8nm and 25nm respectively. The added periodicity in the structure is seen to produce an additional resonance peak, and the maximum field enhancement increases significantly, indicative of a cascaded resonance effect. In order to evaluate whether the cascaded enhancement can produce an improved figure of merit for nonlinear absorption or refraction, the linear absorption spectra were calculated and the third order susceptibility enhancement factor $g^{(3)}$ was determined (not shown). Based on these results, a ‘figure of merit for nonlinear susceptibility enhancement’, defined as $|g^{(3)}|/\alpha$, was calculated. The results for the two structures are shown in Fig. 1(b), with each structure represented by two curves: one curve shows the enhancement of the nonlinear response of the silver (solid line), while the other shows the enhancement of any third order nonlinear susceptibility of the host material. The non-cascaded structure is seen to exhibit a large enhancement FOM near the plasmon resonance frequency, both for the nanoparticles (NP) and the host. The cascaded structure on the other hand shows two main maxima, and frequency regions where the FOM exceeds that of the structure composed of isolated nanoparticles. These results demonstrate that designing cascaded resonant metamaterials can lead not only to increase electric field enhancement, but can also improve the nonlinear optical response relative to the linear response, which would enable improved performance in device applications.

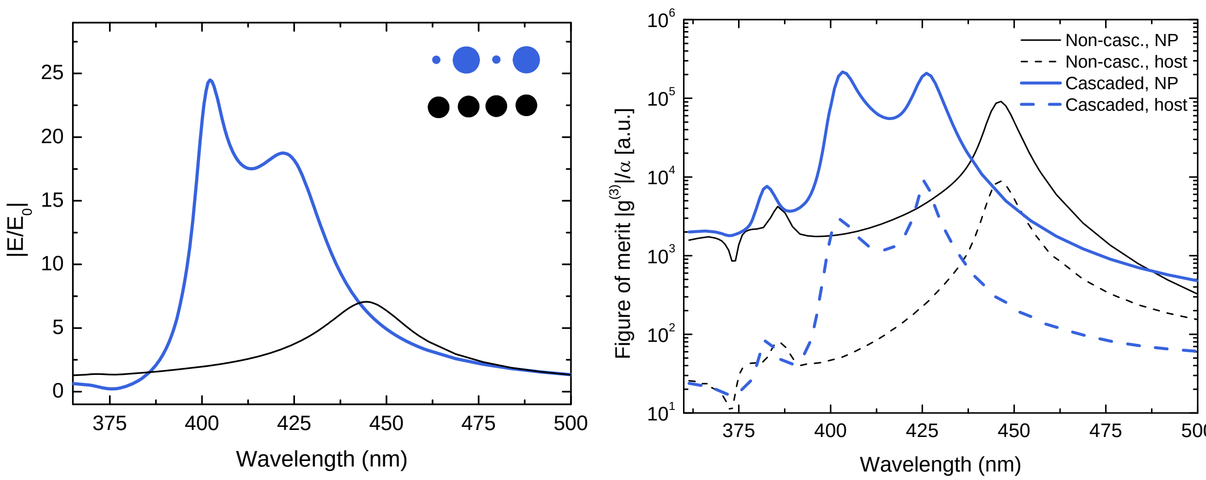

Fig. 1: (a) Simulated field enhancement in the center of the smallest Ag nanoparticle in a metamaterial containing Ag nanoparticles with alternating diameters of 8nm and 25nm, as well as in a nanoparticle embedded in an array with a single particle diameter of 20nm. (b) Calculated Figure of Merit for the enhancement of the third order nonlinear refractive response of silver nanoparticles (NP) and of the surrounding host material, shown for the composite with a single size (‘non-cascaded’), and for the cascaded configuration.

4. Conclusions

It was demonstrated that the use of cascaded plasmon resonances in a nonlinear metamaterial can lead to enhanced performance, as indicated by the observed improvement in the peak figure of merit for the third order nonlinear optical susceptibility enhancement. The presented approach could be extended to multi-level cascading, and cascaded resonances in metamaterials of non-spherical metal inclusions. This material is based upon work supported in part by the U. S. Army Research Office under contract/grant number 50372-CH-MUR.

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