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Enhanced nonlinear optical response of metal nanocomposite based photonic crystals

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We demonstrate enhanced nonlinear optical response from a one-dimensional metal nanocomposite based photonic crystal. A three-fold increase in the two photon absorption coefficient was observed for the photonic crystal structure when compared to a single layer of the metal nanocomposite having comparable metal content. The photonic crystal structure also shows a reduction in the optical limiting threshold by a factor of seven. The combination of metal nanoparticles with appropriately designed plasmon resonance in combination with photonic crystal structure provides an attractive approach for developing practical nonlinear optical devices with low thresholds and wide spectral bandwidth. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4751840]

The need for efficient nonlinear optical materials is driven by a wide array of applications, such as all-optical switches, optical memory, spectroscopy, imaging, ultrashort pulse generation, entangled photon generation, and quantum cryptography. Furthermore, the nonlinear optical effects are usually limited to the high optical intensity regime due to the weak nonlinear optical response of most naturally occurring materials. In this context, metals which possess large third-order nonlinearities and fast temporal optical response are highly attractive. However, due to their large linear optical absorption and therefore poor transparency, metals have not been exploited for developing practical nonlinear optical devices. Recently, however, embedding bulk metals in photonic crystals has enabled one to directly probe their large optical nonlinearities.1,2 By using a photonic crystal geometry, the precise location of the electromagnetic field can be controlled and thereby the overall absorption. While this approach did alleviate the issue of absorption to a reasonable extend, the overall transmission of these structures was still below 25%.1

Metal nanoparticles (NPs) have garnered much attention these days due to numerous application areas stemming from local field enhancement effects near the surface plasmon resonance frequency. High polarizability and ultrafast nonlinear optical response of metal NPs embedded in host matrices have made them attractive for nonlinear photonic applications.3,4 In this letter, we demonstrate enhancement in nonlinear absorption in a one-dimensional photonic crystal composed of alternating layers of a metal NP composite and a passive polymer layer. Open aperture z-scan measurements show enhancement of the nonlinear absorption coefficient of the one-dimensional metal-dielectric photonic crystal as compared to a single film of the same metal composite with similar combined thickness. In addition, optical limiting experiments carried out on the photonic crystal structure showed a seven-fold reduction in the threshold power when compared to a single nanocomposite film of comparable thickness.

As a first step towards the development of the metal-nanocomposite dielectric photonic crystal, we characterized the linear optical properties of the metal nanocomposite film. The silver nanocomposite film was synthesized through the reduction of silver nitrate by polyvinyl alcohol (PVA). 80 mg of silver nitrate dissolved in 0.5 ml of water is mixed with 60 mg of PVA dissolved in 5.2 ml of water to form a precursor solution. This solution is spin coated on a silicon or glass substrate and kept on a hot plate for 90 min at 95 °C where the silver NPs are formed.5 The film was characterized using a J.A. Woolam spectroscopic variable angle ellipsometer in reflection mode. The data were then fit to a Maxwell Garnett model to estimate the linear optical constants and fill factor of the composite film which was found to be 7% using the above process parameters.6

Following the linear optical characterization of the metal nanocomposite film, we fabricated the one-dimensional metal-dielectric photonic crystal as shown in Fig. 1(a) via spin coating. A transverse electron microscope (TEM) image of a dropcoated silver nanocomposite sample is also shown in Fig. 1(a) and shows a fairly uniform size distribution of the silver NPs approximately 5 nm in diameter. The structure consists of ten alternating layers of polymethylmethacrylate (PMMA) with a refractive index of 1.489 and the silver nanocomposite whose index is determined using ellipsometry.6 The PMMA solution is prepared by mixing 0.59 g of PMMA with 21 ml of toluene. The PMMA is spin coated for 50 s at 8000 rpm to attain a thickness of 120 nm and the silver composite is spin coated for 5 s at 500 rpm and then ramped up for 5 s at 1000 rpm to attain a thickness of approximately 160 nm. Spin coating can be used to realize the one dimensional photonic crystal structures because PMMA dissolves in a nonpolar solvent such as toluene and PVA dissolves in polar solvents such as water. This incompatibility of the solvents preserves the integrity of the individual layers while spin coating them on top of each other. A control sample consisting of the same fill fraction (7%) with a total thickness of 800 nm was also fabricated. The thickness of the control sample corresponds to the total

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thickness of silver nanocomposite films in the one-dimensional structure.

Simulated electric field distribution inside the photonic crystal as a function of wavelength is shown in Fig. 1(b). The calculations were carried out using a transfer matrix method incorporating experimentally determined refractive indices of the layers. The one-dimensional structure is designed such that the electromagnetic field at wavelength of 840 nm which corresponds to the two photon absorption (TPA) resonance of the silver NPs used in the present work is localized in the metal nanocomposite layers. This is done to ensure maximum nonlinear optical interaction between the exciting field and the silver NPs. It is this aspect of the design that enhances the overall nonlinear optical response as has been shown previously using bulk metal.1,2 The structures used in the present work have a large linear transmission (∼95%) at 840 nm as shown in the inset of Fig. 2 indicating minimal linear absorption losses.

The nonlinear optical properties of the metal nanocomposite film were characterized using open aperture Z-scan technique.3-9 The Z-scan set up consists of Ti:sapphire laser source with a 120 fs pulse width and a repetition rate of 1 KHz. The excitation wavelength used in this experiment was 840 nm which is twice the plasmon resonance frequency of the silver NPs (∼420 nm) so as to induce TPA. The Z-scan measurements were carried out on the one-dimensional metal nanocomposite-dielectric structure and compared to a single film having thickness and fill factor corresponding to the total thickness of the individual metal nanocomposite layers (5 × 160 nm = 800 nm). The results of the Z-scan measurements are shown in Fig. 2. It is observed that the one-dimensional photonic crystal embedded with alternating layers of the metal composite shows enhancement in the nonlinear absorption compared to the single metal composite layer consisting of the same amount of silver nanocomposite. Z-scan measurement on a pure PVA film with no silver NPs does not show nonlinear absorption indicating that the nonlinear response is entirely from the metal component. The inset in Fig. 2 shows the linear transmission spectra of the one-dimensional metal nanocomposite dielectric structure compared to the single nanocomposite film, obtained using a UV-VIS spectrophotometer. At resonance of the silver NPs (∼420 nm), the linear transmission is ∼15% while in the single layer having the same amount of silver the transmission is ∼60%. This is due to the greater reflectivity of the one-dimensional structure compared to the bulk sample. The one-dimensional photonic crystal structure considered here exhibits a band gap close to the plasmon resonance of the silver NPs. The interaction between the resonance of the one-dimensional photonic crystal and the plasmon frequency of the silver NPs results in this enhancement in reflection at resonance.6 However, at the two photon excitation wavelength of 840 nm (dotted line), the overall reflectivity and linear absorbance of the structure are low and therefore show greater than 90% linear transmission.

In contrast, the TPA is enhanced by a factor of three in the one-dimensional structure when compared to the single film of comparable thickness. This enhancement is quantified by calculating the nonlinear absorption coefficient of the structure by fitting the open aperture normalized transmittance data to

\[ T_{OP} = 1 - \frac{x_0}{2(1 + x^2)^{1/2}} \]

where \( x = \frac{z}{z_0} \) and \( z_0 = \frac{\pi w_0^2}{\lambda} \).

Here \( \beta \) is the nonlinear absorption coefficient given in terms of the fitting parameter.
The nonlinear optical response of the metal NP embedded photonic crystals was further explored by studying its optical limiting properties. Optical power limiting devices are designed to control the transmission intensity of a system. At low powers, the device displays linear transmission until a set threshold is reached at which point the transmitted intensity becomes constant. Such devices have attracted much interest due to applications in providing protection from laser damage to eyes and optical sensors. In addition, efficient control of optical limiting devices can lead to applications requiring pulse generation, shaping and stabilization. However, to exploit the nonlinear behavior of metals for use in optical limiters, efficient control over transmission properties as well as absorption losses is needed. This is achieved here by the combination of the metal-polymer composite and the one-dimensional photonic crystal geometry. The metal embedded polymer film results in a composite utilizing the nonlinear response of the metal particles without severely affecting the overall transmission properties. Additionally, the photonic crystal is designed to maximize the interaction of electromagnetic field with the metallic layers allowing a substantial increase in the nonlinear absorption and thereby improving the optical limiting performance of the entire structure.

The nonlinear response as a function of intensity of the single-layered nanocomposite and the one-dimensional metal-nanocomposite structure are studied at the second harmonic (532 nm) of a Nd:YAG laser with a repetition rate of 10 Hz and a pulse width of 6 ns. Figs. 3(a) and 3(b) show the optical limiting response of the single layer and one-dimensional structure, respectively. For both samples, we see that with increasing input fluence the output eventually begins to clamp. The output fluence at which the one-dimensional structure starts to clamp is ~4 mJ/cm² which is seven times smaller than the bulk composite film whose output clamps at ~28 mJ/cm². The output intensity is fit using the equation

\[ I_o = \frac{I e^{-2L}}{1 + (1 - e^{-2L})\beta l/2\sqrt{2\pi} \lambda} \]

where the two-photon absorption coefficient \( \beta \) is used as a fitting parameter and is found to be \( \beta_{1D} = 3.41 \times 10^{-7} \text{ cm/W} \) and \( \beta_{\text{single}} = 1.11 \times 10^{-7} \text{ cm/W} \) which is similar to values estimated previously using Z-scan measurements. This confirms our estimates of the two photon absorption coefficients and the enhancement observed in the one-dimensional structure is indeed due to the field manipulation.

In summary, we have demonstrated the possibility to enhance the nonlinear optical response of metal nanocomposites by embedding them into photonic crystal structures. The metal nanocomposite allows for the control of the metallicity of the layers through fill fraction while the photonic crystal design helps in precise control of the electromagnetic field distribution. Furthermore, unlike bulk metals, the use of metal NPs allows for scaling such a structure for different wavelengths by changing the particle size and host dielectric constants. A three-fold enhancement in the nonlinear absorption coefficient was shown for the one-dimensional structure as compared to a single nanocomposite film. We also show enhanced optical limiting properties of the metal nanocomposite based photonic crystal structure when compared to a single layer of comparable thickness. Thus, through careful design of structures that combine the advantages of metal NPs and photonic crystals, photonic structures with engineered linear and nonlinear optical properties can be realized.

\[ q_0 = \beta L_{\text{eff}} \quad \text{where} \quad L_{\text{eff}} = \frac{(1 - e^{-2L})}{\lambda}, \]

where \( I \) is the on-axis peak intensity, \( \lambda \) is the linear absorption coefficient, and \( w_0 \) is the beam waist. The intensities used in the open aperture experiment were 21 GW for the one-dimensional sample and 27 GW for the single layer. The fitting parameters for the one-dimensional structure are found to be \( z_0 = 52.8 \text{ mm} \) and \( q_0 = 0.984 \) while those of the single nanocomposite film are \( z_0 = 36.04 \text{ mm} \) and \( q_0 = 0.32687 \). Using the linear absorption to calculate effective length of the samples, the nonlinear absorption coefficients are calculated to be \( \beta_{1D} = 3.67 \times 10^{-7} \text{ cm/W} \) and \( \beta_{\text{single}} = 1.01 \times 10^{-7} \text{ cm/W} \). The enhancement factor which is given by the ratio of \( \beta_{1D}/\beta_{\text{single}} \) is \( \sim 3.11 \).
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