ARTICLE IN PRESS

Optics Communications ∎ (∎∎∎∎) ∎∎∎–∎∎∎

Contents lists available at SciVerse ScienceDirect

Optics Communications

journal homepage: www.elsevier.com/locate/optcom

Enhanced nonlinear optical response of a planar thick metal film combined with a truncated photonic crystal

Hai Lu [a,b], Chun-Hua Xue [a], Yong-Gang Wu [c], Shu-Qi Chen [d], Xiao-Liang Zhang [d], Hai-tao Jiang [a], Jian-Guo Tian [d], Hong Chen [a,*]

[a] Key Laboratory of Advanced Micro-structure Materials, Ministry of Education, Tongji University, Shanghai 200092, China
[b] College of Physics and Information Engineering, Henan Normal University, Xinxiang 453007, China
[c] Institute of Precise Optical Engineering and Technology, Tongji University, Shanghai 200092, China
[d] Key Laboratory of Weak Light Nonlinear Photonics, Ministry of Education, Teda Applied Physics School, Nankai University, Tianjin 300457, China

ARTICLE INFO

Article history:
Received 15 March 2011
Received in revised form
2 September 2011
Accepted 16 July 2012

Keywords:
Photonic crystals
Interface mode
Effective nonlinear susceptibility
Nonlinear absorption

ABSTRACT

Third-order nonlinear optical responses of a heterostructure composed of a planar thick silver film combined with a truncated all-dielectric photonic crystal are studied experimentally. Both open- and closed-aperture *Z*-scan measurements show a large nonlinear response within the metallic structure using a nanosecond laser. Compared with metal–dielectric photonic crystals with the same thickness of metal, the effective nonlinear optical response of heterostructure can be amplified nearly 3 times of the magnitude, due to the enhancement of fields in the metal under the tunneling mechanism. This configuration containing an isolated flat metal film has particular applications in reduced dimensions, such as surface sensor and microscopy.

1. Introduction

The exploration of surface plasmon polaritons (SPPs) was stimulated by various novel applications based on the metallic microstructure [1–3]. Because the propagation constant is greater than the wave vector in the dielectric, SPPs on metal's surface are commonly excited in near-field excitation configurations, such as prism coupling [4,5]. But these schemes inevitably lead to increased losses and scattering, which reduces the SPPs propagation length and increases background radiation [6]. Far-field excitation schemes, such as coupling at gratings and nanoscopic defects, can overcome these restrictions, but the influence between the metallic microstructure and introduced surface structure still cannot be eliminated. Based on very strong nonlinear response of metal (for example, the third-order nonlinear susceptibility of gold is over 3 orders of magnitude larger than that of lithium niobate) [7], a free-space excitation scheme was proposed for direct SPP excitation on flat surfaces of bulk metals [8]. However, for free-space excitation, the conversion efficiency is still low due to the high reflection of metals.

Recent theoretical investigations have shown that the nonlinear excitation of SPPs can be enhanced noticeably by utilizing tunneling modes [9]. Tunneling mode is a type of interface mode existing in the heterostructure with thick metallic film and truncated photonic crystal. In contrast to conventional SPPs, the tunneling modes have in-plane wave vectors less than the wave vector of light in vacuum, which allow far-field propagating radiation to couple into the metal [10,11]. Similar to SPP modes, the electromagnetic (EM) fields of tunneling modes are localized on the surface of the metal, which enhances the nonlinear effect greatly [12,13]. Furthermore, this planar geometry is particularly conducive to nonlinear excitation of SPP. Nevertheless, it is necessary to demonstrate experimentally the nonlinear properties of the tunneling modes in the heterostructure. In this paper we study experimentally the nonlinear optical response of the heterostructure using the *Z*-scan technique. To characterize the nonlinear response of the heterostructure, we compare it with a common metallic structure to be applied for nonlinearity of metals [14–16], i.e. a one-dimensional metal–dielectric photonic crystal (1D –MDPC). Our results show that nearly 3 times magnitude enhancement of nonlinear response could be observed.

2. Experiment and result

The heterostructure is modulated to $(CD)^6MS$, where metal M is chosen to be silver while dielectrics C, D and substrate S are selected to be SiO_2, TiO_2 and K_9 (a kind of glass), respectively. In the $(CD)^6MS$, the thicknesses of metal, SiO_2 and TiO_2 are 50 nm,

* Corresponding author. Tel.: +86 13701910215; fax: +86 2165981203.
E-mail address: hongchen@tongji.edu.cn (H. Chen).

0030-4018/$ - see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.optcom.2012.07.095

92 nm and 50 nm, respectively. The MDPC is modulated to $(CM)^2CS$, where the thicknesses of metal and SiO_2 are 25 nm and 128 nm, respectively. For comparison, the total thickness of metal layers within MDPC is the same as that of the heterostructure. All samples are prepared by the electronic beam evaporation technique and their scanning electron microscope images are shown in Fig. 1(a) and (b).

An ultraviolet–visible–near infrared spectrometer is used for spectral measurements of $(CD)^6MS$ and $(CM)^2CS$, which is shown in Fig. 1(c) and (d). The absorbance is calculated from $A=1–R–T$, in which R, T and A are the reflectance, the transmittance, and the absorbance, respectively. The numerical results are obtained by means of the transfer-matrix method [17]. The wavelength of tunneling mode is designed at 538.2 nm (denoted by λ_0) in $(CD)^6MS$ with $R=0.03\%$, $T=41.09\%$ and $A=58.88\%$. For the real structure, the wavelength of tunneling mode appears at 540 nm, and R, T, and A at the wavelength are 9%, 36%, and 55%, respectively. All spectra are scanned with the minimal oblique angle 5°, and such a small angle means that tunneling modes can be even excited with a negligible tangential wave vector. Meanwhile, $(CM)^2CS$ also exhibits considerable transparency at the 540 nm. The measured values of R, T, and A at 540 nm are 15%, 62%, and 23%, respectively.

The nonlinear response of these structures is characterized using a typical open/closed-aperture Z-scan system under identical experimental conditions. In the experiments, a Continuum Panther-EX optical parametric oscillator (OPO) was utilized, pumped by the third harmonic (355 nm) from a Continuum Surelite II Nd:YAG laser, producing a pulsed laser with wavelength of 540 nm, a repetition rate of 10 Hz and a pulse duration τ_p of ~5 ns. The beam waist ω_0 at focus is about 22 μm. The pulse energy ε_0 is 1.6 μJ, which is far from the damage threshold of silver. At every position, an average of 50 incident pulses that were within ±5% fluctuation was taken by a computer.

The open-aperture Z-scan data for two samples are shown in Fig. 2. The curves show the nonlinear transmittance as a function of the sample distance Z from the waist plane of a Gaussian beam, which is normalized by the linear transmittance measured far from the waist. In the open-aperture Z-scan, the curve comprises a normalized transmittance valley, indicating the presence of nonlinear response. The nonlinear response may be regarded as multiplication between the effective third-order nonlinear absorption coefficient $\beta_{eff}^{(3)}$ and the effective lengthL_{eff}, and can be evaluated from the open-aperture Z-scan normalized transmittance by the relation [18]

$$T(z)=\sum_{m=0}^{+\infty}\frac{\{-\beta_{eff}^{(3)}L_{eff}I_0/[1+(z/z_0)^2]\}^m}{(m+1)^{3/2}} \tag{1}$$

for $|\beta_{eff}^{(3)}L_{eff}I_0|<1$, where $L_{eff}=(1-e^{-\alpha L})/\alpha$ L is the sample length and α is the linear absorption coefficient. At 540 nm, the linear absorption coefficient α in $(CD)^6MS$ and $(CM)^2CS$ can be accessed from Fig. 1. In addition, $I_0=2\varepsilon_0/\pi^{3/2}\omega_0^2\tau_p$ is the laser intensity at the waist plane ($z=0$) and $z_0=\pi\omega_0^2/\lambda$ is the diffraction length of the beam. For both structures studied, the parameters $\beta_{eff}^{(3)}$ of the theoretical curve were fitted to the experimental data at $I_0=23.747$ MW/cm^2 and $m=1$. The nonlinear enhancement factor η is defined as the ratio of the change in nonlinear transmission of two structures δT ($\delta T\approx\beta_{eff}L_{eff}I_0$) at the waist plane, i.e. $\eta=\delta T_{(CD)^6MS}/\delta T_{(CM)^2CS}$. Thus, the deeper valley within the $(CD)^6MS$ reveals a larger positive nonlinear absorption response under the same total thickness of Ag (the nonlinearity of dielectric can be ignored). At this particular wavelength and power, one finds that the δT of the $(CD)^6MS$ is close to 26% whereas it reaches just 8% in the $(CM)^2CS$, corresponding to an enhancement factor of nearly 3.

To obtain more details of the nonlinear optical response of the heterostructure, we exhibit the normalized closed-aperture Z-scan sample transmittance in Fig. 3. The transmission curves

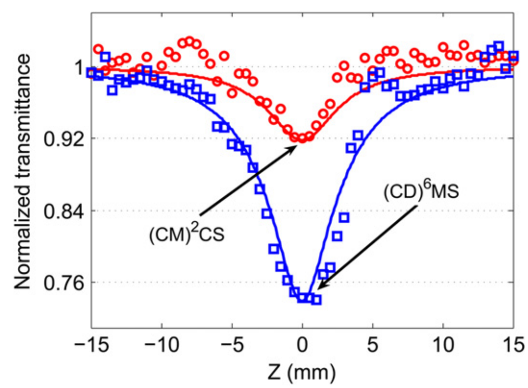

Fig. 2. Open-aperture Z-scan normalized transmittance illuminated by nanosecond pulses at 540 nm. The solid line is the curve fitted (red/blue) to the experimental data (red ○ and blue □) of the sample $(CM)^2CS$ and $(CD)^6MS$ using Eq. (1)., respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

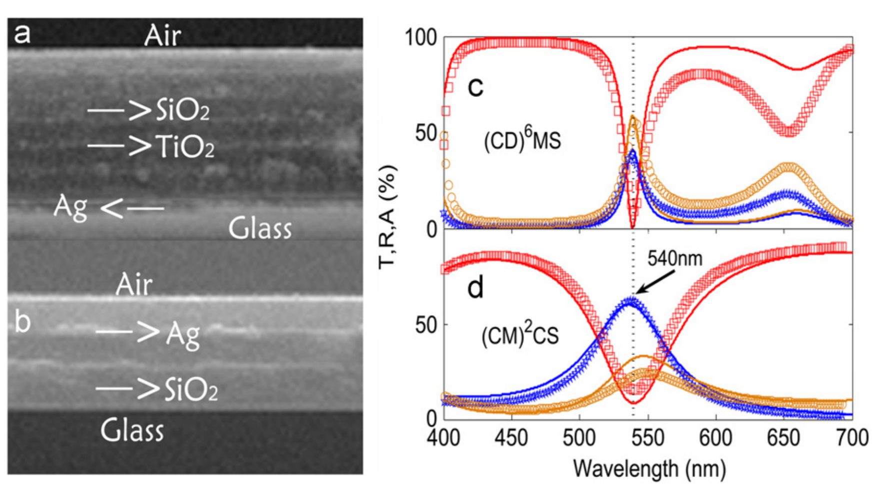

Fig. 1. (a) and (b) are SEM images of $(CD)^6MS$ and $(CM)^2CS$, respectively, (c) and (d) show reflectance, transmittance, and absorbance of $(CD)^6MS$ and $(CM)^2CS$: comparison between numerical values (solid lines) and measured values R (red □), T (blue ☆), A (yellow ○). The parameters are $n_C(SiO_2)=1.46$, $n_D(TiO_2)=2.16$ and $n_S(K_9$ glass$)=1.52$. The refractive indices of silver at wavelength 300 nm, 400 nm, 500 nm, 600 nm, and 700 nm are 1.51+i0.96, 0.17+i1.95, 0.13+i2.92, 0.12+i3.73, and 0.14+i4.52, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

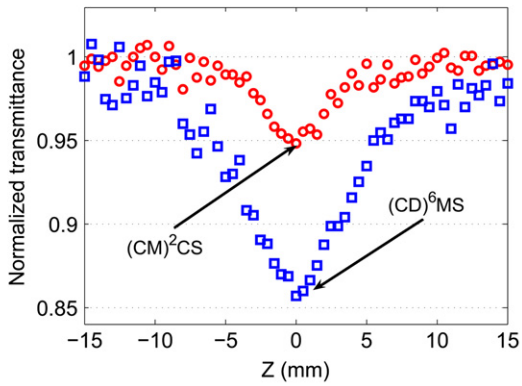

Fig. 3. Closed-aperture *Z*-scan normalized transmittance of the sample $(CM)^2CS$ and $(CD)^6MS$ (red ○ and blue □) by nanosecond pulses at 540 nm., respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

do not reveal the typical peak and valley profile which is usually observed in the case of weakly absorbing materials and photonic bandgap materials [18,19]. Even so, the distinct variation of closed-aperture transmission curve indeed indicates significant nonlinear response in the heterostructure. As known, at the beam waist, the change of nonlinear transmission in closed-aperture *Z*-scan is still attributed to the nonlinear absorption. In Fig. 3, at the waist plane, the δT of the $(CD)^6MS$ is close to 15% whereas in the $(CM)^2CS$ it reaches only 5%, corresponding to an enhancement factor of 3. In addition, a thermally driven transmission change would shift the whole transmission spectrum through thermal variations of the refractive index. However, at a low repetition rate (10 Hz) and low intensities (23.747 MW/cm^2) in our experiments, the contribution coming from thermally induced nonlinearities can be neglected.

3. Discussion

When the nonlinear contribution of the dielectric is negligible compared to that of the metal, the effective third-order nonlinear susceptibility is given by [20]

$$\chi_{eff}^{(3)}(\omega) = \chi_m^{(3)}(\omega)\frac{(1/V)\int_m |E_{loc}|^2 E_{loc}^2\, dV}{|E_0|^2 E_0^2} \quad (2)$$

It can be found that the effective third-order susceptibility $\chi_{eff}^{(3)}(\omega)$ results essentially from the susceptibility $\chi_m^{(3)}(\omega)$ of the metal, and that it may be strongly enhanced through the amplification of the linear local electric field E_{loc} within the metallic component for the metal–dielectric composite material. As shown in Fig. 4, the electromagnetic energy is mostly localized around the interface between the PC and the silver film in the $(CD)^6MS$, and the integral $\int_m |E_{loc}|^2 E_{loc}^2\, dV$ is 3581, where E_0 is the applied electric field and is supposed to be 1. By comparison, $\int_m | E_{loc}|^2 E_{loc}^2 dV$ in $(CM)^2CS$ is only 851 since the nodes of the intensity are located at each thin silver layer, which limits the enhancement of the nonlinear optical effect. The enhancement factor could be predicted as 4.2.

These results show that light can be efficiently coupled into the metal and thereby the nonlinear response can be significantly increased through embedding metallic layer(s) in dielectric matrices properly. For the tunneling mode in heterostructure studied in this work, the key ability is to couple the far-field propagating light into the metal without any microstructure and retain strong local fields near the surface of the metal. A peculiarity of the MDPC structure is that it can exhibit large nonlinearities with reasonable transmittance because of the nodes of the electrical field located at each metal layer. As seen in Fig. 1, the transmittance for the $(CM)^2CS$ is higher than $(CD)^6MS$ at the incident wavelength of 540 nm. In general, increase of field strength in the metal means decrease of transmission because more energy passes through the metallic material. Thus for an optimally tuned structure some trade-off exists between transmission and effective nonlinear response. In other words, if the distribution of EM fields in the MDPC structure is changed at the expense of a part of transparency, MDPC may have a larger nonlinear response, larger than even that of a heterostructure. However, the heterostructure is particularly simple because there is only an isolated flat metal film and thus the momentum conservation in nonlinear multiphoton process will not be influenced by the translational variance. More importantly, the interface local fields similar to SPP in heterostructure can boost the nonlinear conversion efficiency between the tunneling mode and SPP mode.

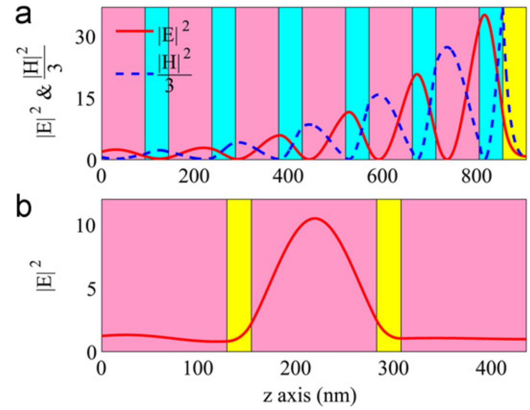

Fig. 4. (a) and (b) are distributions of intensities of EM in the sample $(CD)^6MS$ and $(CM)^2CS$ at the wavelength of 540 nm with the parameters in Fig. 1 when the silver's loss is not taken into account, respectively. The materials in the structure are shown as follows: SiO_2 (pink), TiO_2 (green) and Ag (yellow). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Conclusion

In conclusion, by using the *Z*-scan technique, we have experimentally demonstrated the effective third-order nonlinear optical response of heterostructure composed of a planar thick metal film and truncated all-dielectric PC at the tunneling wavelength 540 nm of nanosecond pulses. Our experimental studies revealed that for the particular heterostructure discussed in this letter one could achieve nonlinear response as much as 3 times larger than that of MDPC consisting of the same thickness of metal. The finding that freely propagating incident radiation can access the optical nonlinearity of flat metals will promote development of surface nonlinear optics and extend the nonlinear applications of an isolated metal film, such as surface-enhanced nonlinear four-wave mixing.

Acknowledgments

This research was funded by the National Basic Research Program of China (Grant 2011CB922001), from the National Natural Science Foundation of China (NSFC) (Grants 10634050 and 11074187), and from the Program for Key Basic Research of the Shanghai Science and Technology Committee (Grant 08dj1400301).

Please cite this article as: H. Lu, et al., Enhanced nonlinear optical response of a planar thick metal film combined with a truncated photonic crystal, Optics Communications (2012), http://dx.doi.org/10.1016/j.optcom.2012.07.095

References

[1] V.M. Shalaev, S. Kawata, Nanophotonics With Surface Plasmons, Elsevier, Kidlington, Oxford, UK, 2007.
[2] S.A. Maier, Plasmonics: Fundamentals and Applications, Springer, New York, NY, USA, 2007.
[3] S.I. Bozhevolnyi, Plasmonic Nanoguides and Circuits, Pan Stanford, Danvers, MA, USA, 2009.
[4] E. Kretschmann, H. Raether, Zeitschrift fuer Naturforschung, Teil A 23 (1968) 2135.
[5] A. Otto, Zeitschrift fuer Physik 216 (1968) 398.
[6] A. Drezet, A. Hohenau, D. Koller, A. Stepanov, H. Ditlbacher, B. Steinberger, F.R. Aussenegg, A. Leitner, J.R. Krenn, Materials Science and Engineering B 149 (2008) 220.
[7] P. Bharadwaj, B. Deutsch, L. Novotny, Advances in Optics and Photonics 1 (2009) 438.
[8] J. Renger, R. Quidant, N. van Hulst, L. Novotny, Physical Review Letters 104 (2010) 046803.
[9] C.H. Xue, H.T. Jiang, H. Chen, Optical Letters 36 (2011) 855.
[10] J.A. Gaspar-Armenta, F. Villa, Journal of the Optical Society of America B 20 (2003) 2349.
[11] Aldo S. Ramírez-Duverger, Jorge Gaspar-Armenta, Raúl García-Llamas, Journal of the Optical Society of America B 25 (2008) 1016.
[12] M. Kaliteevski, I. Iorsh, S. Brand, R.A. Abram, J.M. Chamberlain, A.V. Kavokin, I.A. Shelykh, Physical Review B 76 (2007) 165415.
[13] M.E. Sasin, R.P. Seisyan, M.A. Kalitteevski, S. Brand, R.A. Abram, J.M. Chamberlain, A. Yu., A.P. Egorov, V.S. Vasil'ev, Mikhrin, A.V. Kavokin, Applied Physics Letters 92 (2008) 251112.
[14] N.N. Lepeshkin, A. Schweinsberg, G. Piredda, R.S. Bennink, R.W. Boyd, Physical Review Letters 93 (2004) 123902.
[15] M. Scalora, N. Mattiucci, G. D'Aguanno, M. Larciprete, M.J. Bloemer, Physical Review E 73 (2006) 016603.
[16] M.C. Larciprete, A. Belardini, M.G. Cappeddu, D. de Ceglia, M. Centini, E. Fazio, C. Sibilia, M.J. Bloemer, M. Scalora, Physical Review A 77 (2008) 013809.
[17] A. Yariv, P. Yeh, Optical Waves in Crystals, Wiley, 1984.
[18] M. Sheik-Bahae, A.A. Said, T.H. Wei, D.J. Hagan, E.W. Van Stryland, IEEE Journal of Quantum Electronics 26 (1990) 760.
[19] S.Q. Chen, W.P. Zang, A. Schülzgen, X. Liu, J.G. Tian, J.V. Moloney, N. Peyghambarian, Optical Letters 34 (2009) 3665.
[20] H.R. Ma, R.F. Xiao, P. Sheng, Journal of the Optical Society of America B 15 (1998) 1022.

Please cite this article as: H. Lu, et al., Enhanced nonlinear optical response of a planar thick metal film combined with a truncated photonic crystal, Optics Communications (2012), http://dx.doi.org/10.1016/j.optcom.2012.07.095