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Two-photon absorption measurement of Plasmon-active nanostructures using the open-aperture Z-scan technique

Aliasghar Ajami[1,*], Jan Svanda[2], Wolfgang Husinsky[3]

1 Faculty of Physics, Semnan University, P. O. Box 35195-363, Semnan, Iran
2 Department of Solid State Engineering, University of Chemistry and Technology, 16628 Prague, Czech Republic
3 Institute of Applied Physics, Vienna University of Technology, Wiedner Hauptstrasse. 8-10, 1060 Vienna, Austria

Abstract- In this work, the creation of ordered nanostructure arrays of metals (Ag, Au, Al, and Pd) and characterization of their nonlinear optical properties are described. Large scale ordered metal nanostructure arrays were created by the excimer laser patterning of a polymer surface and subsequent coating with Ag, Au, Al, and Pd. The successful creation of accepted structures was confirmed by the conductive AFM and FIB-SEM techniques. The open-aperture Z-scan technique with femtosecond laser pulses was performed to determine the two-photon absorption (2PA) coefficient of the prepared metal nanostructure arrays. It was found that the strong 2PA appears under the illumination at the wavelength of 800 nm with the polarization direction perpendicular to the long scale metal nano-wires whereas no 2PA was observed when irradiated with laser beam polarized parallel to the metal nano-wires. This reveals that the observed high 2PA activity is a plasmonic-based property due to the confinement effect. An extensive comparison with available literature data shows that the present structures exhibit markedly higher 2PA activity. Obtained results confirmed that the examined approach could be used for the preparation of plasmon-active nanostructures for many applications for a wide range of optical components and devices.

Keywords: Two-photon absorption; Z-scan; Plasmon; nanostructure

* ajami@semnan.ac.ir

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1. Introduction

The distinctive ability of plasmonic structures to concentrate light energy in sub-wavelength space is used to boost the nonlinear optics (NLO) effects without the materials damage [1]. This also provides an efficient pathway to decrease light intensity required for NLO phenomena induction.

Exposed to high-power laser illumination, metallic nanostructures can undergo a wide range of NLO responses including photo-thermal reshaping [2], second harmonic generation [3] and third order optical nonlinearities. In this work, the NLO properties of highly oriented metal (Ag, Au, Al, Pd) nanostructure arrays were investigated using the Z-scan technique [4]. The metal nano-resonators were created on the layer of highly resistant, optically transparent and cross-linked resins using excimer laser followed by tilted evaporation of the metals. The creation of proposed structures was confirmed by the conductive AFM and FIB-SEM techniques.

2. Materials and samples preparation

A solution of Su-8 epoxy-based photoresist was spin-coated onto glass substrates to form polymer films. The samples were dried at 50°C for 2 h and exposed to a UV-source for 30 min and then were dried at 60°C for 24 h.

The polymer surface was patterned by a KrF excimer laser, wavelength 248 nm, pulse duration 20−40 ns, repetition rate 10 Hz. Periodic structures (ripples) on the surface were produced by irradiation with 9000 pulses and fluence of 9 $mJ.cm^{-2}$. The patterned samples were coated with metals by vacuum evaporation. The deposition was performed under an angle of 50° with respect to the sample surface normal from both sides to create metallic nano-wires.

3. Measurement techniques

The Focus Ion Beam Scanning Electron Microscope (FIB-SEM) was used in order to characterize the shape of the metal nanostructures and visual representation of the samples (Figure. 1). The images were taken under the applied voltage of 10 kV. The surface morphology of the samples was examined using Atomic Force Microscope (AFM) working in Peak Force mode with 3 V bias voltage applied between the sample surface) and AFM probe.

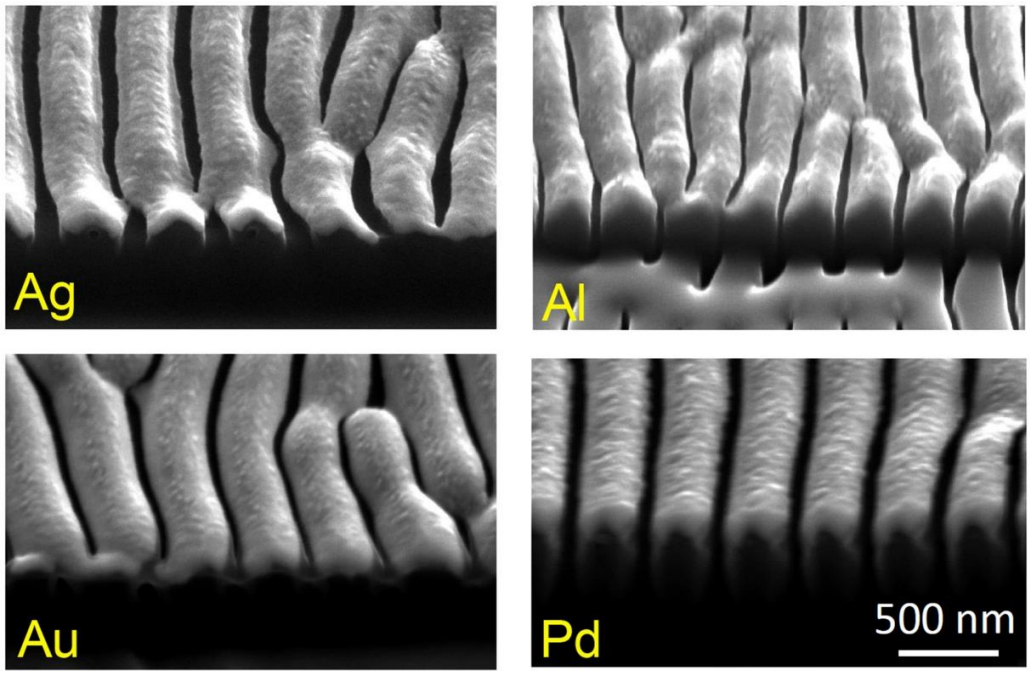


Figure 1. SEM images of the ordered metal nanostructure arrays.

The evaluation of the 2PA coefficient was accomplished using the open-aperture Z-scan. In this method, the normalized transmittance as a function of the sample position is measured in the course of the sample moving through the focal point of

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a focused Gaussian laser beam of high intensity. Then, the 2PA coefficient of the examined sample can be extracted by fitting the measured normalized transmittance to equation 1.

$$T(z) = \sum_{n=0}^{\infty} \frac{\left(\alpha_2 L I_0\right)^n}{(n+1)^{3/2}\left(1+\left(z/z_0\right)^2\right)^n}$$

where I_0 is the peak intensity, L is the sample thickness, z_0 is the Rayleigh length and α_2 is the 2PA coefficient.

In this work, a femtolaser amplifier was employed to perform the Z-scan technique. This system produces 30 fs laser pulses at the central wavelength of 800 nm with a repetition rate of 1 kHz. The laser beam was focused using a 250 mm focal length plano-convex lens. The samples were moved using a motorized translation stage. The transmitted intensity was detected by a Si photodiode. All processes of controlling, measuring and extracting data were accomplished automatically via a LabVIEW program.

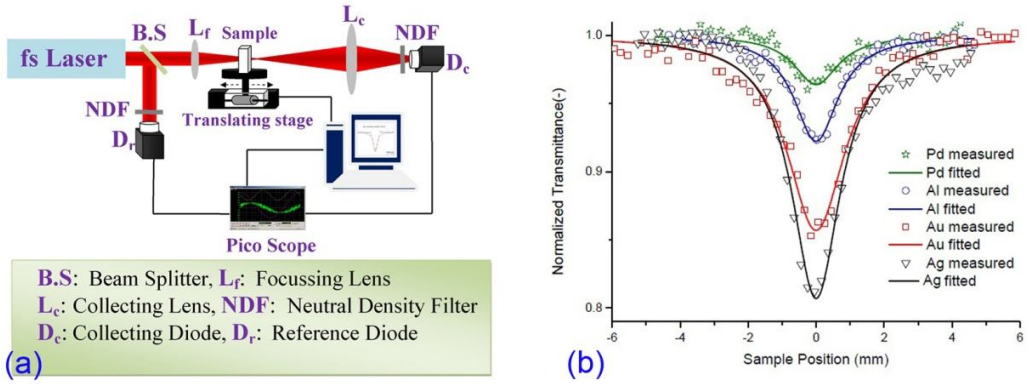


Figure 2. Open-aperture Z-scan setup (a) and Z-scan data for different samples (b).

Two probing polarizations were used to measure the nonlinear optical response of the ordered metal nanostructure arrays. In the first one electric vector was oriented along the main axes of the metal nanostructures. Since the length of each metal structure is too long in comparison with light wavelength used in Z-scan, the given structure can be considered as having no spatial potential which affects the periodical light-induced oscillation of the free electron gas in metallic nanostructures. In other words, the structure did not show any plasmon behavior and its response to the high light intensity was governed by the intrinsic metal properties. As can be expected, no optical nonlinearity was measured under this polarization. Quite different results were obtained using the light polarization with electric field vector orientation perpendicular to the metal nanostructures main axes. In this case, the pronounced effect of transmitted light intensity on the material transparency was detected (figure 2b). It must be additionally noted that in this case, the effective plasmon excitation took place.

Table 1. Summarized 2PA coefficient of examined samples determined by open-aperture Z-scan technique

Metal	Thickness (nm)	Peak intensity ($GW.cm^{-2}$)	2PA coefficient (cm/GW)
Au	21	2270	1080
Ag	25	2640	1160
Al	23	3770	288
Pd	33	4530	71

Conclusion

The large scale ordered metal (Ag, Au, Al and Pd) nanostructure arrays were produced via the excimer laser patterning of polymer surface followed by the tilted metals deposition. Examination of the structural response under the high light intensity indicated the high value of the 2PA coefficient which is strongly related to plasmonic properties of nanostructures.

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