Vector Pulse Shaped Ultrafast Plasmon Based on Response Functions Measured for Orthogonally Polarized Excitation

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Abstract: For spatiotemporal vector pulse control of local plasmon at gold nanostructures, we measure plasmon response functions for orthogonally polarized ultrafast excitation by an electrical-field cross-correlation imaging method using a dark-field microscope. By shaping the vector pulse of the excitation laser, we arbitrary shape plasmon vector pulses.

OCIS codes: (240.6680) Surface plasmons; (320.5540) Pulse shaping; (320.7160) Ultrafast technology

1. Introduction

Localized plasmon resonance in noble metal nanostructures has been receiving much attention to prepare a sub-wavelength-scale interaction platform between light and matter. In addition, it was reported that spatiotemporal control over the localized plasmon or surface plasmon polariton in nanostructures can be achieved using a vector pulse shaping technique for femtosecond excitation laser pulses [1]. So far, a self-learning control algorithm was used to shape an incident femtosecond laser pulse for generating a desired plasmon pulse [2], where the plasmon response was treated as a black-box. In this study, we constructed an experimental setup combining dark field microscopy with cross-correlation interferometer to measure plasmon response functions of gold nanostructures excited by femtosecond laser pulses. The plasmon response functions measured at orthogonal polarizations can deterministically describe the localized plasmon pulse excited by an arbitrary vector-shaped femtosecond laser pulse.

2. Experimental setup

The experimental setup of our cross-correlation measurement with femtosecond laser dark-field microscope is shown in Fig. 1(a). We used an ultra-broadband femtosecond laser (VENTEON, 650~1050 nm, rep. rate of 150 MHz) as an excitation laser source. One beam irradiates the gold nanostructure, and the scattered light is collected by the objective mirror, while the other laser beam directly reaches the CCD camera with a variable optical delay. We measured a series of image shots by varying the time delay and analyzed the fringe-resolved cross-correlation functions at a specific point in the dark-field image. Nanostructures we measured are cross-shaped Au with various aspect ratio fabricated on SiO_2 substrate (Fig. 1(b)).

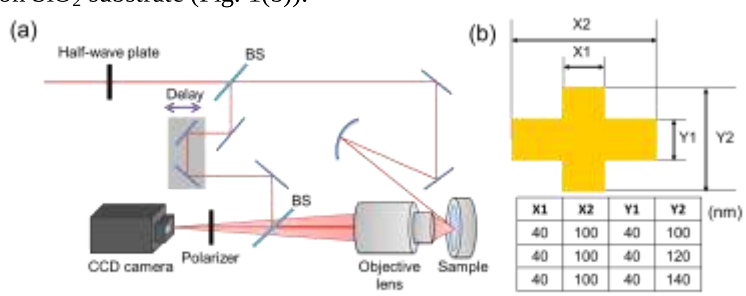

Fig. 1. (a) Experimental setup of dark-field microscopy. (b) Shape of Au cross-shaped nanostructure.

The spectral plasmon response functions are obtainable from the Fourier transform of a temporal cross-correlation function, when both the excitation and the reference pulses are known in their amplitude and phase [3].

3. Results and Discussion

First, the polarizations of the excitation and the measured light were set in parallel to the one of the nanocross arm. Figures 2(a) and (b) show the plasmon spectra and the plasmon response functions, respectively, obtained at an Au nanorod with R=2.5 and 3.5. We observed a clear plasmon resonance enhancement and a dependence on R. The spectral responses are in good agreement with those predicted by FDTD calculations.

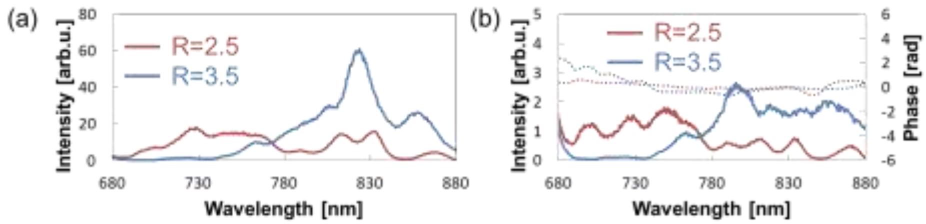

Fig. 2. (a) Plasmon spectra and (b) response functions of nanocross with R=2.5 and 3.5.

Next, the polarizations of the excitation and measured light were tilted at 45-degree to the Au nanocross arm. Figures 3(a) and (b) show the measured plasmon spectrum and the plasmon spectrum predicted from the orthogonal response functions in Fig. 2(b) for the nanocross consisting of the nanorods with R=2.5 and 3.5. The measured plasmon spectrum shows a strong resemblance to the calculated plasmon spectrum. Therefore, a linear description of the plasmon pulse based on the orthogonal plasmon response functions was validated. Based on this linear description, one can deterministically shape the plasmon vector pulse by shaping the pumping laser vector pulses.

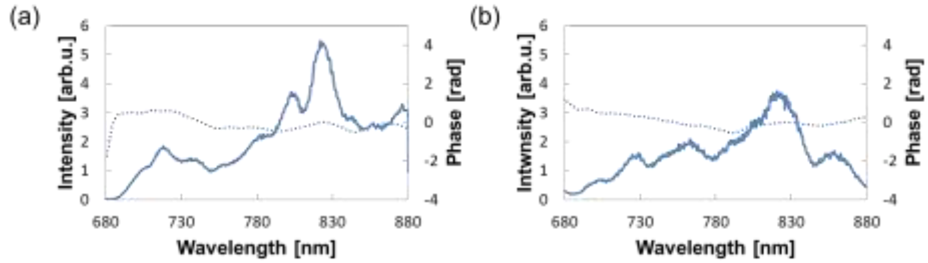

Fig. 3 (a) Measured plasmon spectrum when the excitation polarization was tilted at 45-degree from one of arms of nanocross consisting of nanorods of R=2.5 and 3.5. (b) Calculated plasmon spectrum with the orthogonal response functions shown in Fig. 2 (b).

Figure 4 shows examples of vector shaped plasmon pulses designed on the nanocross of R=2.5 and 3.5: a linearly polarized Fourier transform limited pulse at 45-degree (left) and a circularly polarized Fourier transform limited pulse (right). To generate the linearly polarized FTL pulse at 45-degree, we applied the inverse spectral phase of the response function for each the orthogonally polarized excitation pulses and shaped the spectral amplitude identical for the both.

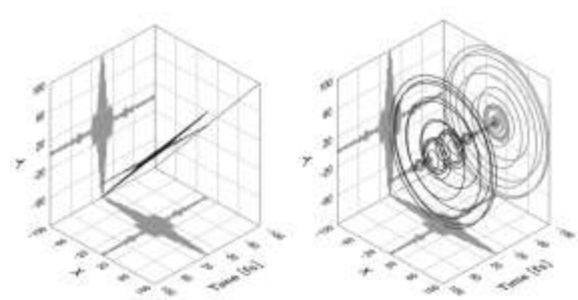

Fig. 4 Vector shaped plasmon pulse based on the orthogonal plasmon response finctions.

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