

Spatiotemporal Ultrafast-Plasmon Control Based on Response Functions of Nanostructures Measured by Interferometric Cross-Correlation Microscopy

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Abstract. We demonstrate an electrical-field cross-correlation imaging technique to obtain a response function of localized plasmon generated by femtosecond laser pulses on gold nanostructures. Based on the measured response functions, we spatiotemporally control the plasmon by shaping the femtosecond excitation laser pulses.

1 Introduction

Study on plasmon in nanometer structure is in rapid development. Especially, the localized plasmon resonance at noble metal nano-structures has been devoting much attention to produce a novel interaction platform between light and matter. In addition, it has been reported that spatiotemporal control over localized plasmon can be achieved using polarization pulse shaping technique for femtosecond lasers in regularly-arranged nanostructures [1]. In order to make spatiotemporal control for localized plasmon or surface plasmon-polariton possible, the temporal characterization of femtosecond plasmon field is required. So far, temporal characterization of ultrafast plasmon using PEEM has been reported [1,2]. We hereby apply a new characterization method of ultrafast plasmon: electrical-field cross-correlation imaging using femtosecond laser dark-field microscopy. We set our research goal at spatiotemporal control of plasmon generated by ultra-broadband laser pulses, which are shaped based on the plasmon response function [3] measured with our novel method.

2 Electrical-field response function of localized plasmon

The electrical-field cross-correlation $\tilde{M}(\omega)$ is described using an electrical-field response function $R(r, \omega)$ of localized plasmon as follow:

$$\tilde{M}(\omega) = \tilde{E}(r, \omega) \tilde{E}_{ref}^*(\omega) = [\tilde{R}(r, \omega) \tilde{E}_{pump}(\omega)] \tilde{E}_{ref}^*(\omega), \quad (1)$$

where $\tilde{E}(r, \omega)$, $\tilde{E}_{ref}(\omega)$, and $\tilde{E}_{pump}(\omega)$ are the electrical field of a localized plasmon, a reference pulse and a pump pulse, respectively. According to Eq. (1), $R(r, \omega)$ is obtained from the cross-correlation and the reference spectrum amplitude when $E_{pump}(\omega) = E_{ref}(\omega)$.

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$$R(r, \omega) = \frac{\tilde{M}(\omega) \tilde{E}_{ref}^*(\omega)}{\tilde{E}_{pump}(\omega)} = \frac{\tilde{M}(\omega)}{|\tilde{E}_{ref}(\omega)|^2} \quad (2)$$

Therefore, the localized plasmon waveform $\tilde{E}_{shape}(r, \omega)$ can be shaped by a shaped laser pulse $\tilde{S}_{shape}(\omega)$ as follow:

$$\tilde{E}_{shape}(r, \omega) = \tilde{R}(r, \omega) \tilde{S}_{shape}(\omega) = \frac{\tilde{M}(\omega)}{|\tilde{E}_{ref}(\omega)|^2} \tilde{S}_{shape}(\omega) \quad (3)$$

3 Experimental setup and result

Experimental setup of cross-correlation measurement with femtosecond laser dark-field microscopy is shown in Fig.1. The ultra-broad band femtosecond laser (VENTEON, spectral width of 650~1100 nm) is used as an excitation laser source. The dark-field microscope consists of a parabola mirror ($f=1500$ mm), an objective lens ($\times 10$, N.A.=0.25) and a CCD camera. One beam irradiates a gold nanosystem and the scattered light is collected by the object lens, while the other laser beam directly reaches to the CCD camera with variable optical delay. We measured a series of image shots by varying the time delay and analyzed the fringe-resolved cross-correlation functions at several points in the dark-field image.

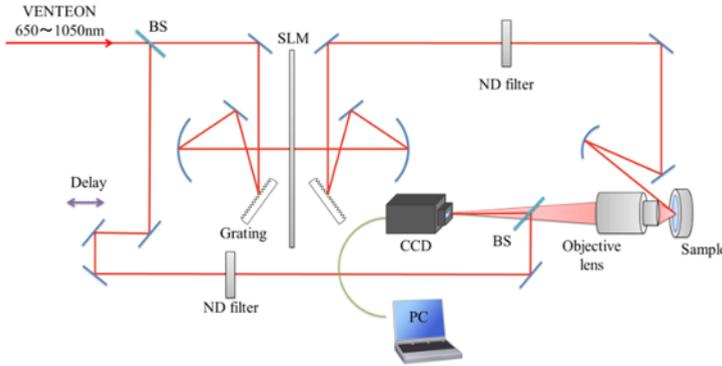


Fig. 1 Experimental setup of a cross-correlation dark-field microscope.

A typical example is shown in Fig. 2. These are the fringe-resolved cross-correlation functions measured for various shapes of Au nanocross. Using a similar setup but in autocorrelation measurement, we observed clear enhancement by plasmon resonance at the plasmon spectrum obtained by Fourier transform of the autocorrelation functions as well as the dependence on excitation pulse polarization. Therefore, although the spatial resolution of the dark-field microscopy is low, which is same as a conventional optical microscopy, we can observe the spectral response of nanostructures at multi-points simultaneously with this scheme. From these cross-correlation functions shown in Fig. 2, we obtained the response functions of the nanorods as shown in Fig. 3. The laser pulse can be shaped by a 4f-type pulse shaper consisting of a computer-controlled LC-SLM to generate desired plasmon pulses at a specific position in the nanosystem as described in the Eq.(3).

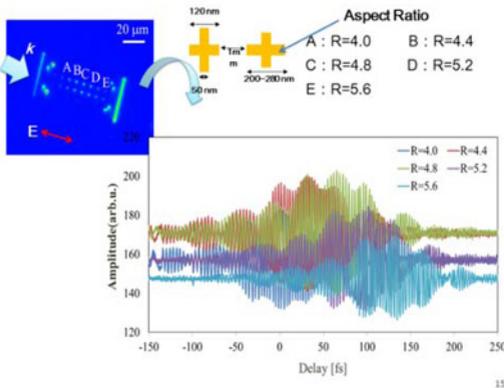


Fig. 2 Fringe-resolved cross-correlation functions measured for nanocrosses with various aspect ratio R (ratio between crossing rods).

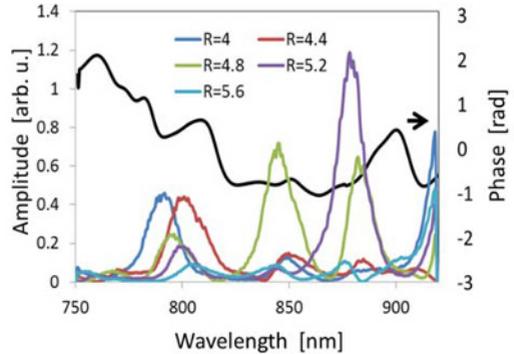


Fig. 3 Response functions measured for various Au nanocrosses. The spectral phase is for R=5.2.

We also developed a nearfield scanning optical microscopy combining a spectral interferometry to measure cross-correlation function with much higher spatial resolution of ~ 100 nm. Spectral interference is formed by plasmon field collected by a fiber probe and a reference laser pulse.

For well-defined nanostructures, the response function can be obtained by a FDTD numerical model. However, actual nanostructures tend to be out of a design and it is not easy to numerically model them. One can use a self-learning control algorithm to shape an incident laser pulse for generating a desired plasmon pulse. This adaptive control will be a powerful tool when employing vector shaped laser pulses instead of linearly polarized laser pulses[1], since it is not easy to measure a full description of plasmon response functions for orthogonal polarization components of incident laser pulses. Moreover, the response function itself must consist of orthogonal three vector components for each of the orthogonal incident laser polarization components..

When we combine spatially shaped laser pulses, such as in the form of the coherent superposition of high-order beams, with temporal pulse shaping, more comprehensive spatiotemporal plasmon control will be available.

4 Conclusion

We demonstrated an electrical-field cross-correlation imaging technique to obtain response function of localized plasmon generated by femtosecond laser pulses on gold nanostructures. Based on the measured response functions, we spatiotemporally shaped the plasmon on the nanorods by shaping the femtosecond excitation laser pulse.

References

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