Control of Femtosecond Surface Plasmon Coupled onto a Gold Tapered Tip and its Nonlinear Emission

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Abstract: Spatiotemporal nanofocusing of surface plasmon polariton excited by femtosecond laser pulses on a sharp conical Au tip with a tip edge radius of few tens of nanometers is deterministically controlled.

OCIS codes: (240.6680) Surface Plasmon, (250.5403) Plasmonics, (320.0320) Ultrafast optics

1. Introduction

Surface plasmon polaritons (SPPs) are transverse magnetic polarized optical surface waves that propagate along a metal-dielectric interface with fields that peak at the interface and decay away exponentially into both medium [1]. Recently, optical devices based on SPPs have attracted much attention because of their ability of confining and guiding electromagnetic waves in subwavelength scales, which allow for the manipulation and guidance of optical signals in very compact, fast and power-efficient devices. An efficient transformation of traveling delocalized SPPs into highly localized excitations is therefore of paramount importance to achieve bright illumination of confined volumes. Ropers *et al.* demonstrated efficient nonlocal optical excitation of the apex of a nanostructured metal taper by grating-coupled SPPs excited by femtosecond laser pulses [2]. These SPPs propagate over more than 10 μm along the tip shaft toward the apex of the tip, where they are reradiated into the far field. The tip emission is spectrally tunable by varying the angle of incidence of the illumination. The temporal characteristics of SPPs coupled on an Au metal taper have been experimentally analyzed by Berweger *et al*. with IFROG using second-order harmonics generated at the apex of the tip [3]. They demonstrated that the spectral phase of SPP pulses can be controlled by shaping the excitation laser pulses.

In this paper, we employed cross-correlation dark-field image microscope to analyze SPP pulses coupled on an Au metal taper. We demonstrate deterministic arbitrary plasmon pulse control by shaping excitation laser pulse. In addition, we control non-linear radiation at the apex of the tip.

2. Experimental setup

The schematic of cross-correlation dark-field image measurement is shown in Fig. 1. We used an Au tapered tip with a radius of curvature at the tip apex of ~20 nm and an opening angle of 15°. SPP can be excited in periodic structures on metallic surface via grating coupling. A linear grating with a 1730 nm period is fabricated by FIB onto the tip shaft at ~10 μm away from the tip apex. The linear grating consists of 8 grooves with a width of 860 nm and a depth of 200 nm.

The femtosecond laser source is Ti:sapphire oscillator (8 fs (FWHM), $\Delta\lambda$=600-1000 nm). One of laser pulse beams split at a beam-splitter is focused onto the grating with a spot size of ~10 μm using a microscope objective lens (x20, *NA*=0.35 and a working distance of 20 mm) at an angle of incidence of 29°. Scattered light at the apex of the tip is collected by a microscope objective lens (x40, *NA*=0.55 and a working distance of 7.5 mm) and imaged on a charge-coupled device (CCD). We employ a dark-field measurement scheme to prevent incident light from incoming directly to the objective lens. The other beam is used as a reference pulse, thus sent directly to the CCD. We measure fringe-resolved correlation images by varying an optical delay between two beams. The spatial resolution of this dark-field imaging is limited by the diffraction limit determined by the objective lens. However, this cross-correlation dark-field microscope will be a powerful tool to measure ultrafast SPP time histories both in the phase and the amplitude without any nonlinear optics. After the acquisition of a series of cross-correlation images, a cross-correlation waveform is obtained at the apex. Then, the spectral amplitude and phase of the SPP pulse are obtained by deconvolution using the reference pulse waveform which was fully characterized in advance. We obtain the response function of SPP associated with coupling and propagation, and control SPP pulse based on the response functions.

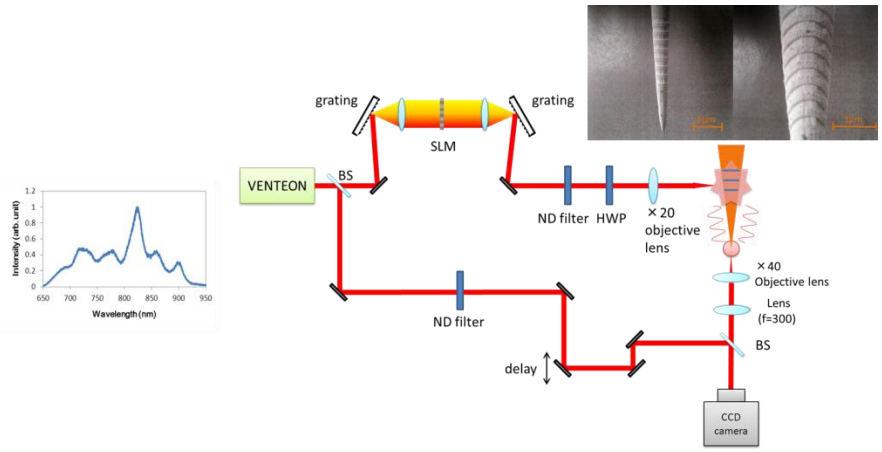

Fig. 1 Experimental setup of cross-correlation dark-field image measurement. Insets are excitation laser spectrum and a SEM pictures of Au tapered tip.

3. Results and Discussion

Figure 2 shows results obtained from the analysis of correlation waveforms measured at the apex. Figure 2(a) shows measured response function of SPP. The excitation of SPP onto the tip is achieved by grating coupling to overcome the momentum mismatch between the incident wave vector and the in-plane SPP momentum [2, 3]. In our experiment, spectral component around 800 nm efficiently couples to the grating and propagates. Once the response function is obtained, temporal plasmon characteristics can be deterministically designed. We shape the excitation laser pulse by a conventional pulse shaper (Fig. 1) so that the dispersion of the response function is fully compensated and Fourier transform limited (FTL) plasmon pulse is generated at the apex. As a result, the SPP pulse width is reduced from 36.0 fs to 12.3 fs as shown in Fig. 2(b).

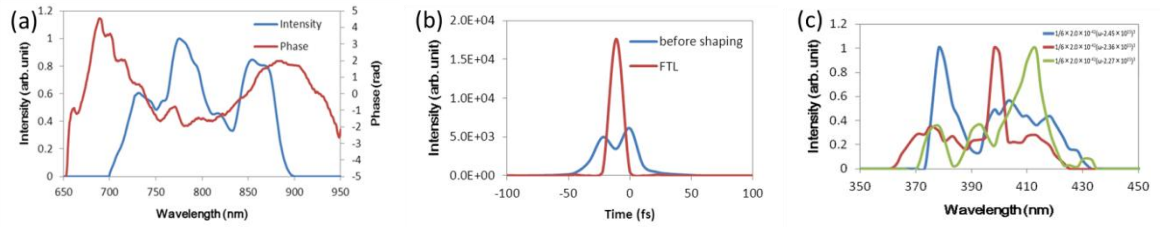

Fig. 2 (a) Measured plasmon response function, (b) measured plasmon pulses excited by FTL laser pulse and shaped laser pulse to generate a FTL plasmon pulse, and (c) control of SH spectrum at the apex by 3rd-order dispersion.

In addition to the fundamental wavelength of SPP pulses, the broken axial symmetry at the tip apex allows local generation of optical second harmonic (SH). Since SH emission is extremely weak, we employ a photon counting method to detect SH light. We shape the spectral phase of the femtosecond excitation pulse based on the response function and examine the pulse shaping scheme by the SH spectrum change. When third-order dispersions are designed for the SPP pulse at the apex, the peak of SH spectrum peak is exactly shifted in 370-420 nm to the point of inflection of the each third-order dispersion curve as shown in Fig. 2 (c). Therefore, we can selectively perform two-photon excitation at the apex with this control scheme. Similar control with spectral phase shaping can be applied for difference frequency mixing and four-wave mixing at the apex. Thus, nonlocal optical excitation of the apex of a nanostructured metal taper will be a versatile nano nonlinear light source.

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