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Femtosecond optical control on the nanoscale

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Abstract. We demonstrate a generalized route to generate nanometer spatially confined ultrafast optical pulses with arbitrary deterministic femtosecond waveform control using surface plasmon polarition nanofocusing in 3D tapered noble metal tips.

1 Introduction

Plasmonic nanostructures as optical antennas have been proposed to overcome the size and mode mismatch between far-field propagating light and nanoscale quantum systems. Previous approaches to achieve nanometer control over ultrafast fields have relied on the use of appropriately shaped pulses to achieve spatial localization [1]. However, as the nanofocusing mechanism itself relies on the spectral phase and polarization characteristics, this fundamentally limits the control of the optical transient that can be obtained on the nanoscale.

Here we present an optical antenna concept capable not only of efficient nanoscale light concentration, but that also enables the simultaneous and independent control of the ultrafast optical transients via pulse shaping. This approach utilizes surface plasmon polarition (SPP) nanofocusing into the apex of 3D noble metal tips [2,3], which we have previously demonstrated to allow for nanometer spatial confinement of optical fields at the apex [4]. As shown in Figure 1a), this effect arises from the increase in index of refraction with decreasing tip radius experienced by propagating SPP's, which in combination with the associated decreasing group velocity and improved mode confinement leads to the continuous transformation of quasi-planar modes into a nanoconfined excitation at the tip apex [5,6].

We demonstrate the compatibility of SPP nanofocusing with ultrafast techniques enabled by the inherent broadband capability of the process [7]. As illustrated in Figure 1b), in combination with frequency domain pulse shaping we excert deterministic control over femtosecond pulses at the tip apex by generating transform-limited pulses as well as arbitrary waveforms. The resulting nanometer-femtosecond spatiotemporal control at the end of a scanning probe tip can be used to perform all-optical nanometer-resolved ultrafast and nonlinear near-field imaging.

2 Methods

SPP's are launched onto Au tips via grating coupling as illustrated in Figure 1b). The grating is fabricated via focused ion beam milling using a fan-shaped design to optimize broadband performance. The output of a Ti:Sa oscillator is shaped using a pulse shaper with a folded 4f geometry and focused onto the grating. The launched SPP pulses are nanofocused into the tip apex where in addition to the fundamental emission, the broken apex symmetry allows for second-harmonic generation (SHG) that is detected using an imaging spectrograph coupled to N_2(l) – cooled

CCD camera in a 90°-detection geometry. Access to the spectral phase provided by SHG is used for full tip-apex pulse characterization through interferometry frequency-resolved optical gating (IFROG) and pulse duration optimization using a multiphoton intrapulse interferometric phase scan (MIIPS) algorithm. Due to sensitive grating-coupling conditions pulse-pairs are generated using the pulse shaper to ensure collinearity and thus identical coupling.

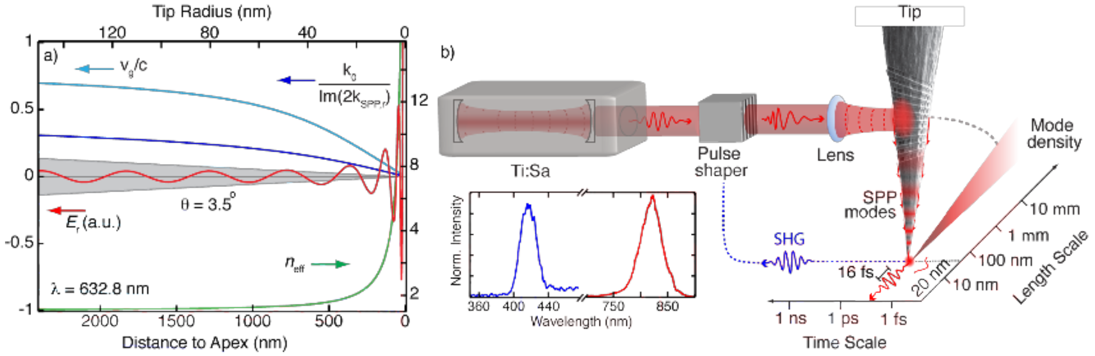

Fig. 1. a) SPP's propagating on a conical noble metal waveguide experience a radius-dependent effective index of refraction (green), associated decrease in group velocity (light blue), and improved mode confinement (dark blue), leading to a concentration of the SPP field into the tip apex (red) and nanometer spatial confinement. b) Illustration of the experiment. Pulses from a femtosecond Ti:Sa oscillator are focused onto a grating coupler to launch SPP pulses onto the tip shaft. The nanofocused pulses generate SHG at the tip apex that is collected and analyzed. Resulting access to the spectral phase allows for pulse characterization via FROG and optimization of the spectral phase via MIIPS.

3 Results

As previously shown, SPP nanofocusing can generate a nanoconfined excitation with 10's of nm spatial confinement where nanofocusing efficiencies as high as 9% enable efficient background-free imaging and spectroscopy [8,9]. Under sufficiently high illumination conditions SHG is generated locally at the tip apex that is used through a MIIPS algorithm to produce transform-limited pulses.

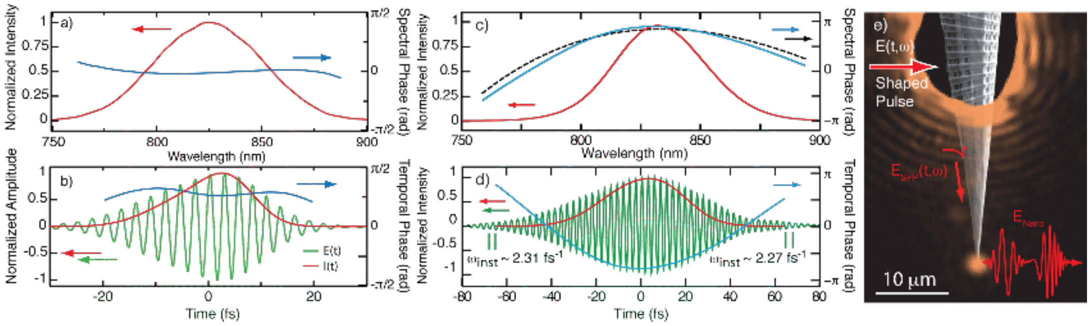

Fig. 2. Pulses reconstructed from tip-apex IFROG acquired with MIIPS-optimized pulses generated using the pulse shaper. a) Spectral profile of a reconstructed pulse with a spectral width of ~60 nm and flat spectral phase, b) temporally corresponding to a transform-limited duration of 16 fs with the electric field transient shown in green. The generation of arbitrary optical transients is also possible. c) reconstructed spectrum (red) and phase (blue) along with the applied phase of GDD = 200 fs^2 (black dashed) along with the d) corresponding temporal profile. The electric field transient of the engineered chirped pulse (green) shows the expected time varying instantaneous frequency. e) In combination with pulse shaping the new antenna concept enables full nanometer-femtosecond spatiotemporal control for quantum coherent control as well as ultrafast and nonlinear near-field imaging and spectroscopy.

Measurement of pulses is performed via tip-apex IFROG generated using nanofocused pulses after MIIPS optimization with the reconstructed temporal and spectral profile in Figure 2a) and b), respectively. The spectral profile with a flat spectral phase and spectral FWHM = 60 nm corresponds to a transform-limited pulse with a temporal duration as short as 16 fs [7].

The spectral characteristics of an arbitrarily engineered pulse are shown in Figure 2c) with the spectrum (red) and phase (blue) reconstructed from an XFROG measurement, showing good agreement between the applied phase of 200 fs^2 (black dashed) and the reconstructed phase. The temporal characteristics in panel d) show the correspondingly chirped pulse with a broadened profile and a varying instantaneous frequency as seen from the electric field transient (green) [7]. The high degree of waveform engineering achievable arises from the inherently broadband nanofocusing process with no dependence on the spectral phase and only a weak wavelength dependence.

These results show the compatibility of this antenna concept with ultrafast techniques, allowing not only for the optimization of pulse duration, but also for the arbitrary control and generation of optical waveform transients as illustrated in Fig. 2e). The resulting qualitatively new control of the light-matter interaction on simultaneous nanometer – femtosecond scales control will open up applications ranging from quantum coherent control to high harmonic generation. Furthermore with spatial localization occurring at the apex of a scanning probe tip, this spatiotemporal control will open up new avenues to study the nature of the fundamental electronic and vibrational excitations of matter on their characteristic length and time scales simultaneously.

References

1. M. Aeschlimann *et al.*, "Adaptive subwavelength control of nano-optical fields," Nature **446**, 301 (2007)
2. M.I. Stockman, "Nanofocusing of optical energy in tapered plasmonic waveguides," Phys. Rev. Lett. **93**, 137404 (2004)
3. C. Ropers, C.C. Neacsu, T. Elsaesser, M. Albrecht, M.B. Raschke, and C. Lienau, "Grating-Coupling of Surface Plasmons onto Metallic Tips: A Nanoconfined Light Source," Nano Lett. **7**, 2784 (2007)
4. C.C. Neacsu, S. Berweger, R.L. Olmon, L.V. Saraf, C. Ropers, and M.B. Raschke, "Near-field localization in plasmonic superfocusing: A nanoemitter on a tip," Nano Lett. **10**, 592 (2010)
5. N. Issa and R. Guckenberger, "Optical Nanofocusing on Tapered Metallic Waveguides," Plasmonics **2**, 31 (2007)
6. S. Berweger, J.M. Atkin, R.L. Olmon and M.B. Raschke, "Light on the Tip of a Needle: Plasmonic Nanofocusing for Spectroscopy on the Nanoscale," J. Phys. Chem. Lett. *In Press* (2012)
7. S. Berweger, J.M. Atkin, R.L. Olmon and M.B. Raschke, "Femtosecond nano-focusing with full optical waveform control," Nano Lett., **11**, 4309 (2011)
8. D. Sadiq, J. Shirdel, J.S. Lee, E. Selishcheva, N. Park, and C. Lienau, "Adiabatic Nanofocusing Scattering-Type Optical Nanoscopy of Individual Gold Nanoparticles," Nano Lett. **11**, 1609 (2011)
9. S. Berweger, J.M. Atkin, R.L. Olmon and M.B. Raschke, "Adiabatic tip-plasmon focusing for nano-Raman spectroscopy," J. Phys. Chem. Lett., **1**, 3427 (2010)