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Adiabatic Plasmon Nanofocusing for Ultrashort Pulses and Spectroscopy JOANNA ATKIN, SAMUEL BERWEGER, ROBERT OLMON, XIAOJI XU, MARKUS RASCHKE, University of Colorado at Boulder — The simultaneous control of optical fields on both nanometer spatial and femtosecond time scales would enable direct spectroscopic access to the elementary electronic and vibrational excitations in matter. Here, we utilize adiabatic surface plasmon polariton (SPP) nanofocusing on free-standing 3D tapered metal tips in order to generate nanometer confined field localization at the tip apex. Using the second harmonic generation (SHG) at the tip apex we perform MIIPS pulse optimization and frequency-resolved optical gating (FROG) characterization of the nanofocused pulses. With the combination of high bandwidth coupling using a chirped grating, pulse-shaping, and low-dispersion nanofocusing, we can achieve full optical control on the nanoscale, from $<$ 16 fs pulse duration to arbitrary optical waveforms. This technique enables linear and non-linear plasmon-enhanced spectroscopy, with the simultaneous temporal control over ultrashort pulses opening the possibility for true time-resolved scanning-probe imaging. We demonstrate this capability for background-free probing of individual molecular and nanocrystalline systems.

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