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Plasmonic nano-focused four-wave mixing for femtosecond nano-imaging VASILY KRAVTSOV, RONALD ULBRICHT, University of Colorado at Boulder, JOANNA ATKIN, University of North Carolina at Chapel Hill, MARKUS RASCHKE, University of Colorado at Boulder — We experimentally demonstrate efficient and broadband nonlinear optical four-wave mixing (FWM) in a deep sub-wavelength volume using adiabatic nanofocusing of surface plasmon polaritons (SPPs). We couple few-femtosecond laser pulses into SPPs that propagate and experience nanofocusing on a sharp Au tip, and detect blue-shifted intra-pulse FWM response of the nanofocused plasmons. Due to the asymptotic mode volume compression and resulting steep increase in SPP field enhancement when approaching the tip apex, the nonlinear signal is highly localized in a nanoscopic volume at the apex. The simultaneous enhanced generation and further FWM field compression into the tip apex provides for a highly sensitive nano-probe for ultrafast near-field microscopy and spectroscopy. We demonstrate the use of the nano-localized nonlinear frequency conversion to spatially resolve few-femtosecond dynamics in ultrafast coherent spatio-temporal nano-imaging of the localized plasmonic modes of an inhomogeneous Au layer edge. Our results are supported by simulations and provide a perspective for an all-optical implementation of the novel multidimensional nano-spectroscopies.

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