

Abstract Submitted
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Time-Resolved Surface-Enhanced Coherent Sensing of Nanoscale Molecular Complexes GEORGE R. WELCH, DMITRI V. VORONINE, ALEXANDER M. SINYUKOV, XIA HUA, KAI WANG, PANKAJ K. JHA, ELANGO MUNUSAMY, STEVEN E. WHEELER, ALEXEI V. SOKOLOV, MARLAN O. SCULLY, Texas A&M University, College Station, TX 77843 — Nanoscale real-time molecular sensing requires large signal enhancement, small background, short detection time and high spectral resolution. We demonstrate a new vibrational spectroscopic technique which satisfies all of these conditions. This time-resolved surface-enhanced coherent anti-Stokes Raman scattering (tr-SECARS) spectroscopy is used to detect hydrogen-bonded molecular complexes of pyridine with water in the near field of gold nanoparticles with large signal enhancement and a fraction of a second collection time. Optimal spectral width and time delays of ultrashort laser pulses suppress the surface-enhanced non-resonant background. Time-resolved signals increase the spectral resolution which is limited by the width of the probe pulse and allow measuring nanoscale vibrational dephasing dynamics. This technique combined with quantum chemistry simulations may be used for the investigation of complex mixtures at the nanoscale and surface environment of artificial nanostructures and biological systems.

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