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Nanoscale sensing with surface-enhanced CARS spectroscopy DMITRI VORONINE, ALEXANDER SINYUKOV, XIA HUA, GUOWAN ZHANG, WENLONG YANG, KAI WANG, PANKAJ JHA, GEORGE WELCH, ALEXEI SOKOLOV, MARLAN SCULLY, Texas A&M University, College Station, TX 77840 — Time-resolved coherent anti-Stokes Raman scattering (CARS) is extended to the nanoscale regime where nano-molar amounts of molecules may be investigated. We show that spectral resolution of CARS generated by ultrashort laser pulses may be improved by collecting a sequence of time-resolved spectra. This procedure provides additional linewidth information and allows detecting two species of pyridine molecules in a vicinity of aggregated gold nanoparticles. Surface-enhanced CARS (SE-CARS) signals of the adsorbed pyridine monolayer are detected in the presence of a bulk pyridine background CARS signal. Time-resolved SE-CARS signals are stronger than the conventional CARS, and more sensitive to the surface environment which makes them suitable for nano-surface characterization with high molecular specificity. This technique allows measuring the vibrational dephasing time of molecules on nano-surfaces and characterizing the effects of the surface local environment on the ultrafast molecular dynamics. This technique may be applied to a variety of artificial and biological systems and complex molecular mixtures and has a potential for nanophotonic sensing applications.

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