Molecule-surface interactions probed by optimized surface-enhanced coherent Raman spectroscopy

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— Nanoscale molecular sensing is carried out using a time-resolved coherent anti-Stokes Raman scattering (CARS) spectroscopy with optimized laser pulse configurations. This novel technique combines the advantages of an improved spectral resolution, suppressed non-resonant background and near-field surface enhancement of the Raman signal. We detect two species of pyridine in a vicinity of aggregated gold nanoparticles and measure their vibrational dephasing times which reveal the effects of surface environment and molecule-surface interactions 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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