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Optimizing coherent Raman scattering with plasmonic nanoparticles DMITRI VORONINE, XIA HUA, ALEXANDER SINYUKOV, CHARLES BALLMANN, ALEXEI SOKOLOV, MARLAN SCULLY, Texas A&M University — Two commonly used techniques that provide species-specific spectroscopic signals in the form of vibrational fingerprints are surface-enhanced Raman scattering (SERS) and coherent anti-Stokes Raman scattering (CARS) spectroscopies. In order to enhance the signal, SERS takes advantage of the electromagnetic near-field enhancement while CARS employs molecular coherence. We have combined these two techniques to achieve best-of-both-worlds maximum signal enhancement by using optimized laser pulse shaping and time-resolved detection. We applied this new time-resolved surface-enhanced coherent anti-Stokes Raman scattering (tr-SECARS) technique to investigate various molecular complexes in a vicinity of gold nanoparticles. While large signal enhancement has previously been achieved in SERS, surfaced-enhanced coherent signals have shown lower values. We investigate the mechanisms of these effects by analyzing the spatial dependence of the coherent Raman spectra for different hot spots in aggregated plasmonic nanoparticles. Understanding coherence effects in surface-enhanced Raman scattering may lead to improved nanoscale sensors.

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