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INVITED

Surface-enhanced coherent anti-Stokes Raman scattering

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Abstract

Although the Raman effect is a weak light-matter interaction, strong signals can nonetheless be generated with the help of the enhanced optical properties of nano-structured metals. The concentration of the excitation field and the enhancement of radiative rates mediated by the metal's plasmonic modes can raise the otherwise imperceptible Raman signal from single molecules to detectable levels. The amplifying qualities of metallic nano-structures are ideally suited for detecting and identifying molecular species at low concentration, spurring the development of chemical sensors based on surface-enhanced Raman scattering (SERS) into a burgeoning field. Translation of these principles to nonlinear Raman techniques, however, has not been trivial. The different heating kinetics under pulsed illumination, combined with nonlinear optical radiation from the metallic antenna itself, has complicated the realization of clean and reproducible nonlinear Raman experiments. Nonetheless, various efforts have shown potential [1]-[3]. Given the tangible benefits that nonlinear Raman techniques offer, including stronger enhancement of the signal and the ability to time-resolve the molecular response, obtaining a better understanding of nonlinear Raman processes in the vicinity of plasmonic amplifiers is a meaningful endeavor.

In this contribution, we discuss various realizations of surface-enhanced coherent anti-Stokes Raman scattering (SE-CARS) experiments. We start with SE-CARS on flat gold surfaces, followed by experiments on nano-antennas in the single molecule limit. We study the relevant mechanisms at play and discuss the implications for tip-based CARS experiments.

Reference:

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