PROBING PLASMON-INDUCED TEMPERATURES IN FLUOROPHORE-PLASMONIC SYSTEMS USING RAMAN THERMOMETRY.

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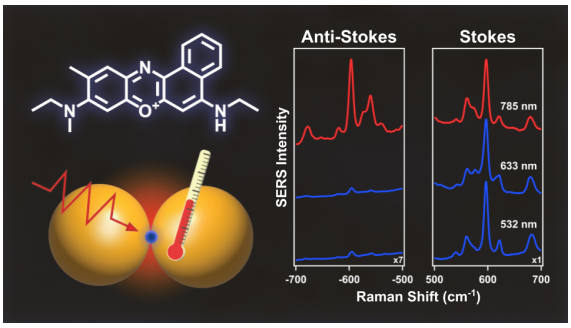
Plasmonic materials have increasingly grown in interest in chemical sensing, optoelectronics, and photocatalysis. Plasmonic media interact strongly with light, focusing and enhancing electromagnetic radiation to nanoscale volumes, not seen with typical propagation of electromagnetic radiation. Although plasmonic materials have countless desirable properties, we still struggle to form a fundamental understanding of energy and charge transfer at plasmonic interfaces.



We specifically desire to quantify energy transfer in plasmonic-molecular systems in this work. We utilize continuous wave, surface-enhanced anti-Stokes and Stokes Raman spectroscopy to probe the vibrational energy transfer. Further, we employ a Boltzmann distribution analysis to quantify our results, to correlate the anti-Stokes to Stokes scattering ratio of Raman-active vibrational modes to their corresponding temperatures. Specifically, we examine the temperatures of plasmonic-fluorophore systems, where molecules can undergo electronic transitions, which specifically follow an unforeseen mechanism. In comparison to room temperature population densities, we observe a 100K decrease in the temperature of various fluorophore molecules under resonant steady-state excitation. In contrast, under non-resonant excitation, we see an increase in temperature up to 200K. This resonant plasmonic cooling effect occurs regardless of vibrational mode selection and solvating environment. Our work provides new insight into plasmonic-molecular interactions and an initial investigation of this occurrence.