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"New Directions in Plasmonics: Pushing the Sensitivity, Space, and Time Limits"

During the last few years, there has been an explosion of interest and activity in the field of plasmonics. The goal of plasmonics is to control and manipulate light on the nanometer length scale using the properties of the collective electronic excitations in noble metal films or nanoparticles, known as surface plasmons. An improved understanding of the interactions between adsorbed molecules and plasmonic nanostructures (i.e., molecular plasmonics) is having a significant impact in a number of research areas. These include surface-enhanced Raman spectroscopy (SERS), localized surface plasmon resonance (LSPR) spectroscopy, sub-wavelength optical microscopy, and nanolithography.

This lecture will begin with some background material on the basic physical concepts underlying plasmonics with an emphasis on SERS and LSPR spectroscopy. Next, I will turn to the area of single molecule surface enhanced Raman spectroscopy (SMSERS) and a discussion of our efforts to provide a robust existence proof for SMSERS. Further, we will answer several fundamental questions such as: (1) what is the largest possible enhancement factor (EF) and (2) what nanostructure produces the largest EF. Our approach to answering these questions involved the development of new tools using SMSERS, single nanoparticle SERS and single nanoparticle LSPR spectroscopy spatially correlated with high resolution transmission electron microscopy (HRTEM).

The concluding section will focus on two very recent developments. For the first time, the revolutionary techniques of surface enhanced Raman spectroscopy and femtosecond stimulated Raman spectroscopy (FSRS) have been combined. Thus, plasmonically enhanced broadband Raman spectra using an ultrafast four wave mixing process, which can simultaneously achieve spectral and temporal resolution below the time-energy uncertainty limit, has been achieved! Further we report substantial progress in tip-enhanced Raman spectroscopy (TERS). The isotopologue proof of single molecule specificity in ambient TERS has been demonstrated. Further, a UHV-TERS instrument has been constructed with atomic resolution of the surface and sub-molecular resolution of the adsorbate. The capabilities of this machine are shown with the copper phthalocyanine (CuPc)/Ag(111) system. We can now forsee the day when it will be possible to combine UHV-TERS and surface enhanced FSRS to enable single-molecule spectroscopy with simultaneous nanometer spatial resolution and femtosecond time resolution.