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Recent Progress in the Study of Single Molecule Chemistry at the Nanometer Length Scale and Picosecond Time Scale

RICHARD VAN DUYNE, Departments of Chemistry, Biomedical Engineering, and The Applied Physics Program, Northwestern University

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 including electrochemistry, surface science, catalysis for energy conversion and storage, the materials science of nanoparticles, biomedical diagnostics, art conservation science, and nanolithography. In the first part of this lecture, I will provide some background material on the basic physical concepts underlying molecular plasmonics with an emphasis on surface-enhanced Raman spectroscopy (SERS), localized surface plasmon resonance (LSPR) spectroscopy, and tip-enhanced Raman spectroscopy (TERS). In the second part of this lecture, I will focus in on three recent advances in TERS which illustrate the power of this nanoscale vibrational spectroscopy. First, new insights into the nature of the relative intensity fluctuations in single molecule tip-enhanced Raman spectroscopy (SMTERS) will be discussed. Second, our current understanding of the adsorbate surface interactions involved in the low temperature (LT), ultrahigh vacuum (UHV) TERS of the Ag tip/Rhodamine 6G (R6G) /Ag(111) system will be described. Finally, an update on our new results in coupling ultrafast lasers with TERS. This last topic illuminates a path forward toward the goal of understanding chemistry at the space-time limit.