DETECTION AND CHEMICAL IDENTIFICATION OF SINGLE MOLECULES AT THE SURFACE OF NOBLE METAL NANOSTRUCTURES

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The enhancements of absorption, fluorescence emission, and Raman scattering from analytes positioned near the surface of appropriate enhancing noble metallic nanoparticles are widely known.[1,2] Surface-enhanced Raman scattering (SERS) and surface-enhanced resonance Raman scattering (SERRS), as the most significant of these phenomena, permit single molecule detection at the surfaces of Ag and Au nanoparticles.[3,4] The detection and vibrational spectroscopy of single molecules employing surface-enhanced Raman scattering, although still largely in its infancy, is quickly contributing to changing the fundamental way in which molecules are viewed. It is proving to be a very powerful analytical technique with potential application towards a wide variety of problems. Not only do SERS/SERRS provide the high cross sectional values necessary for the detection of single, isolated molecules, as fluorescences does[5], they also offer the high information content that is characteristic of molecular vibrational techniques. This last fact is particularly important in studying the unique features of individual molecules when ensemble averaging is removed.

In this study, the unique characteristics of single molecules, and the exceptional optical properties of noble metallic nanoparticles, are explored through a series of surface-enhanced resonance Raman scattering experiments carried out on Langmuir-Blodgett monolayers deposited on metal island films. This technique towards single molecule SERRS is unique, and has been pioneered in our group.[6,7] It involves spatial resolution with a Raman microscope, of single target PTCD molecules isolated and embedded in easily transferred, and non-interfering, fatty acid monolayer matrices, deposited on enhancing metallic substrates **(Fig. 1)**. Here, single molecule results are presented from a variety of experiments representing combinations of different possible molecular and surface plasmon resonance conditions. As well, several metallic nanostructured substrates have been employed **(Fig. 2)** with a variety of analytes, utilizing single-point, mapping **(Fig. 3)**, and global imaging techniques. Detailed results are presented for several interesting analytes, and, more generally, insight is provided into the nature of the local electromagnetic "hot spots" that provide enhancements of Raman signals up to 10^{10}, and are essential for single molecule SERS/SERRS detection.

References:

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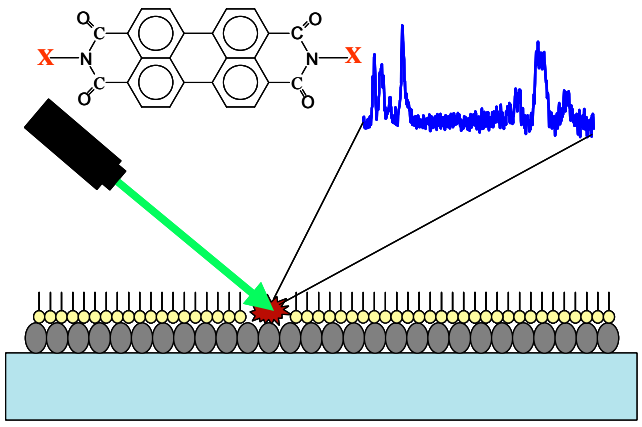

Figure 1. Schematic representation of single molecule LB-SERRS measurement. Also shown is structure of PTCD derivatives.

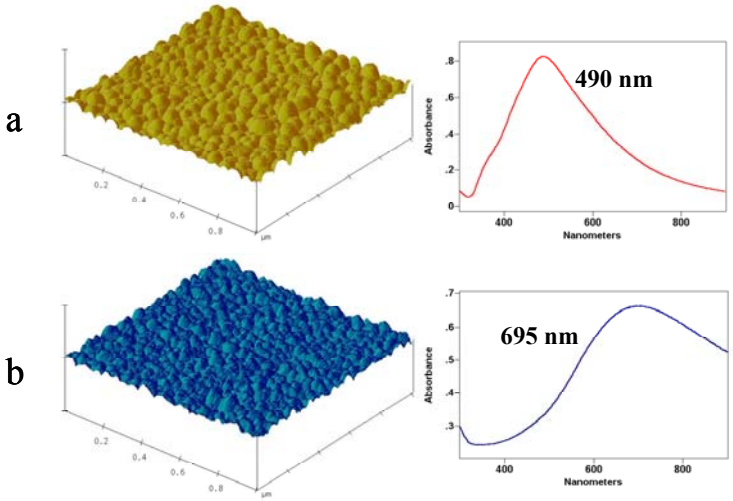

Figure 2. AFM images, and surface plasmon absorption spectra for **(a)** Ag, and **(b)** Ag/Au mixed island films.

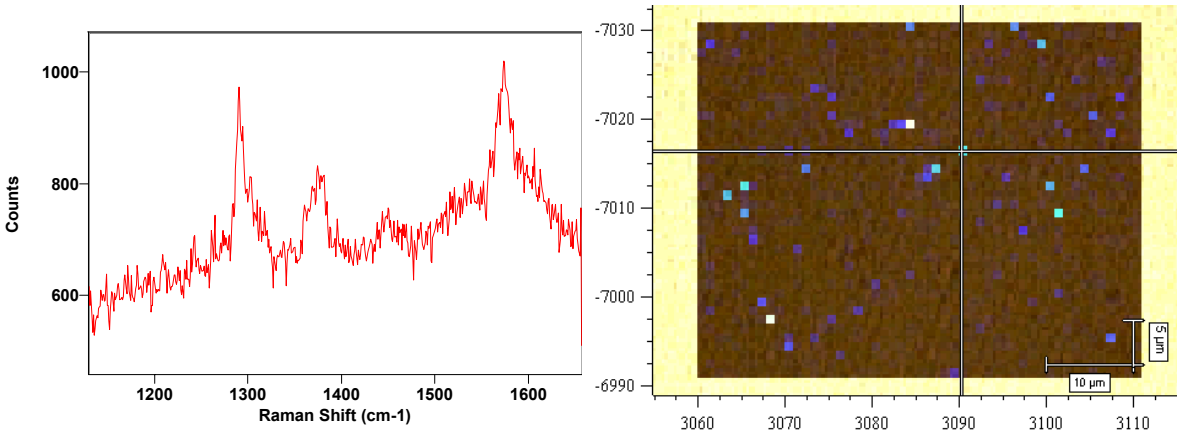

Figure 3. LB-SERRS mapping of isolated single PTCD molecules on a Ag island film.