Luis Velarde

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University at Buffalo, NY

WHEN Wednesday, Sept. 19, 2018

WHERE Widtsoe Hall 330

TIME 4:00-5:00 pm

Refreshments Served 3:45 pm

Host: Yi Rao

Nonlinear Optics and the Interplay between Electronic Structure and Molecular Vibrations at Interfaces

Understanding the delicate interplay between nuclear motion and electronic structure at interfaces is a challenging task of high relevance to many scientific areas such as photocatalysis, photovoltaics, and nanophotonics. We demonstrate that Doubly-Resonance Sum Frequency Generation (DR-SFG) spectroscopy can be used as a surface and species-selective probe in order to characterize how specific vibrational modes of interfacial species are coupled to excited surface electronic states and influence functional properties. We show how second-order nonlinear probes for vibronic coupling can shed light on surface aggregates in photosensitized catalysts, structural conformers of substituted azobenzene molecules on surfaces and phonon-assisted transitions in nanostructured carbon films. These studies allow us to gain insights into how specific nuclear motions may drive electronic processes at surfaces. In addition, we show how sum frequency generation vibrational spectroscopy can be used to examine the binding, orientation, and ordering of organic ligands in solution-processed nanomaterials. We present how the surface order of ionic surfactants on semiconducting single-walled carbon nanotubes is influenced by the conditions of the surrounding aqueous environment, providing new physical insights for solution-processing of carbon nanotubes in nanoelectronics. Furthermore, we present the direct relationship between surface orientation and structural motifs in thiophene- and selenophene-substituted chalcogenopyrylium (CP) dyes on gold nanoparticles as a new class of ultra-bright, NIR-absorbing Raman reporters. These results advance our understanding of the role of molecular orientation in SERS probes. The results of this work provide an opportunity for the development of controlled chemistries for the design of nanosurfaces with tailored properties.

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