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Molecular monolayers under high pressure: a study using surface enhanced Raman scattering and vibrational sum frequency generation spectroscopy¹ YUANXI FU, DANA DLOTT, School of Chemical Sciences, University of Illinois at Urbana-Champaign — Vibrational spectra of molecular monolayers in a diamond anvil cell (DAC) reveal the conformation and packing state of the monolayers undergoing large-amplitude deformations. Measuring monolayer spectra under high pressure can be difficult due to the small number of molecules. We used surface enhanced Raman scattering (SERS) and vibrational sum frequency generation (VSFG) spectroscopy in a DAC to address the challenge. Localized surface plasmon resonance generated on curved metal surfaces enhances the adsorbates' Raman scattering cross-sections by factors of 10^6 , allowing SERS spectra of monolayers formed by organic thiols on silver coated polystyrene nanospheres and dyes on silver colloidal nanoparticles to be studied up to several GPa. To better understand the role of curvature, monolayers on planar surfaces were studied using a new diamond anvil cell for VSFG spectroscopy. VSFG is a nonlinear coherent vibrational spectroscopy that uses converging IR and visible femtosecond laser pulses. The VSFG spectra of long-chain alkane monolayers were studied up to several GPa.

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> Yuanxi Fu School of Chemical Sciences, University of Illinois at Urbana-Champaign

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