

Abstract Submitted  
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**Coherence-Modulated Third Harmonic Generation for Second Hyper-Raman Spectroscopy of Molecules at an Interface** KEVIN DILLMAN, Colorado State University, JESSE WILSON, Duke University, RANDY BARTELS, NANCY LEVINGER, Colorado State University — We have developed a method of probing the low-frequency (sub-1000  $\text{cm}^{-1}$ ) vibrational modes of molecules at interfaces using third-harmonic generation (THG). The THG process is enhanced at an interface due to the differences in the third-order nonlinear susceptibilities of the materials. We have used this method to collect low-frequency second hyper-Raman spectra from BGO,  $\text{BaF}_2$  and  $\text{CdWO}_4$  crystals. In addition, we have observed coherent second hyper-Raman scattering arising from  $\text{CCl}_4$  molecules at the liquid-glass interface. We are presently extending these techniques to observe resonant second hyper-Raman scattering from dye molecules adsorbed on gold nanoparticles in order to gain surface enhancement effects. We aim to use this method to characterize the environment at interfaces of reverse micelle systems. The development of this method is significant because we can sensitively probe the low-frequency vibrational modes of only those molecules at an interface.

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