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**Coherence modulated third harmonic generation for interface vibrational spectroscopy** JESSE WILSON, DAVID KUPKA, OMID MASHZADEH, RANDY BARTELS, Colorado State University — Though third harmonic generation (THG) by tightly focused beams at an interface is generated in bulk regions of the two surrounding materials, we demonstrate measurement of surface-specific vibrational information by coherence-modulated THG (CM-THG). In CM-THG, a tightly focused ultrafast pulse in the presence of an impulsively-prepared vibrational coherence will undergo modulation by the coherence during the THG process. Measuring the CM-THG signal as a function of scanning the interface allows separation of bulk and interface phonon oscillation contributions to the CM-THG signal. Three distinct processes occur leading to modulation of the detected third harmonic signal by the coherence: 1) The third harmonic generation itself is modulated by the coherence through a transient nonlinear susceptibility, i.e. coherent second hyper-Raman scattering (CSHRS); 2) the fundamental is modulated by transient linear susceptibility cascading to the third harmonic; and 3) the index of refraction at the boundary modulates the fundamental prior to third harmonic generation in air after the sample interface. To our knowledge, this is the first measurement of CSHRS. Though hyper-Raman selection rules have been proposed, no experimental verification has yet been performed.

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