

**Title:**

Vibrational Specificity and Resolution Control in Nonlinear Microscopy

**Abstract:**

Imaging is a valuable tool for many areas of science. One form of microscopy used coherent nonlinear signals as a contrast mechanism for image formation. In this work, we make use of coherent third harmonic generation (THG) and phase-sensitive impulsive stimulated Raman scattering (ISRS) for imaging applications. We have developed a new method of phase-sensitive coherent ultrafast vibrational Raman spectroscopy. These techniques are particularly adept at resolving low-frequency vibrational modes. We are extending the sensitivity of this technique for specific tailored vibrational spectra with a combination of pulse shaping and phase-sensitive detection. In THG imaging, we have developed a technique to programmably control the polarization state of a microscope focal field. Using basic properties of nonlinear optics, we apply polarization control of the focal field to control over the resolution and point spread function of the microscopy system.