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Anharmonic Coupling in Beta Barium Borate Using Two-dimensional Spectroscopy. MEGAN NIELSON, BRITTANY KNIGHTON, TANNER HARDY, LAUREN RAWLINGS, ALDAIR ALEJANDRO, JEREMY JOHNSON, Brigham Young University — High-field, ultrafast pulses of terahertz (THz) light allow the extreme excitation of atomic motion that allow us to probe regions of the underlying potential energy surface (PES). The PES, which dictates the material properties of a sample, has been studied with one-dimensional (1D, single pump pulse) measurements. At times, 1D measurements can be challenging to interpret. More information is available using 2D Terahertz spectroscopy to investigate the specific nonlinear couplings between excitations in solid materials. Here we use 2D Terahertz spectroscopy to gain insight into how phonon modes interact at large oscillation amplitudes to study β-barium borate (BBO), a commonly used nonlinear optical crystal. BBO has 16 phonon modes that are both Raman and IR active within our pump excitation bandwidth (1-6 THz), and the interaction between these modes leads to a feature rich 2D spectrum. We study the BBO sample responses, as a function of THz polarization with respect to the sample, to learn about the modes that nonlinearly interact to contribute to the 2D spectrum. Modeling the anharmonic coupling of the vibrational modes allows us to disentangle the 2D spectrum and gain insights into the complex underlying PES.

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