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Direct Comparison Between Multi-Dimensional Terahertz Vibrational Spectroscopies BRITTANY E. KNIGHTON, MEGAN F. NIELSON, R. TANNER HARDY, ALDAIR ALEJANDRO, LAUREN M. RAWLINGS, JEREMY A. JOHNSON, Brigham Young University — Multidimensional terahertz (THz) spectroscopy is a powerful tool for understanding nonlinear excitation, coherent energy flow, and coupling between collective degrees of freedom on ultrafast time scales. With extreme vibrational excitation using high-field THz light, 2D THz spectroscopy can reveal how anharmonic mode coupling results in coherent energy transfer and ascertain the excitation pathways behind nonlinear sample responses. We directly compare 2D THz-THz transmission measurement to 2D THz-THz-Raman measurements and explore evidence of anharmonic coupling between phonon modes in beta-barium borate (BBO). We find that 2D THz-THz-Raman spectroscopy produces a richer spectrum, better signal to noise, and is compatible with thicker samples than 2D THz-THz measurements. We model the phonon mode couplings in BBO to begin to untangle the complex 2D spectral features.

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