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**Differentiating Between Different Nonlinear Excitation Terahertz Pathways Through the Use of Two-Dimensional Spectroscopy** ALDAIR ALEJANDRO , BRITTANY E. KNIGHTON, R. TANNER HARDY, LAUREN M. RAWLINGS, MEGAN F. NIELSON , JEREMY A. JOHNSON, Brigham Young University — Intense, short pulses of terahertz (THz) radiation has made it possible to study and manipulate the properties of materials on ultrafast (<1 ps) time scales. As with any region of the electromagnetic spectrum, intense terahertz pulses can induce nonlinear excitation pathways. In order to understand how THz pulses can be used to manipulate structure in solids, it is of great interest to understand the process or processes that cause this type of nonlinear behavior. In this work, Raman-active modes in a $CdWO_4$ crystal are nonlinearly excited. With single-pulse (one-dimensional) THz measurements, we cannot distinguish between nonlinear photonic (two-photon absorption) and nonlinear phononic (anharmonic coupling between vibrational modes) processes. We show that two-dimensional (2D) spectroscopy allows us to identify the main nonlinear excitation pathway (two-photon absorption), and by making slight (but significant) changes to the experimental setup, we can isolate the secondary excitation pathway (anharmonic coupling). The nonlinear phononic (anharmonic coupling) pathway has been posited with theoretical work, but never before experimentally confirmed.

Aldair Alejandro
Brigham Young University

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