A Combined Experimental and Computational Approach to Understanding Nonlinear THz Vibrational Excitation in Solids

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Intense terahertz (THz) light can nonlinearly drive vibrational modes in solids through processes such as electronic Raman scattering (ERS) and anharmonic phonon coupling; however, it is often impossible to distinguish between these nonlinear processes using one-dimensional spectroscopy. Two-dimensional (2D) spectroscopy can help unpack complicated excitations. 2D THz spectroscopy has been used recently to study electronic excitations [1], spin waves and coherent motion in gases [2,3], and molecular vibrations in liquids [4]. We use 2D THz spectroscopy to study nonlinear vibrational excitations in crystalline systems [5,6].

We have performed 2D measurements on cadmium tungstate ($CdWO_4$) using intense THz excitation pulses to nonlinearly excite vibrational modes between the frequencies of 1-6 THz. Modeling 2D experiments using the coupled equations of motion can help to untangle these complicated spectra. In $CdWO_4$, all points from the 2D spectra (Fig. 1a) have a quadratic dependence with respect to THz intensity, indicating that third-order nonlinear excitation pathways such as trilinear phonon coupling or electronic Raman scattering dominate. Since both possible nonlinear excitation pathways have the same field dependence, 2D modeling of the spectra is essential in beginning to unpack which signal arises from which nonlinear process. We create models that include both of these main processes and compare the modeled 2D spectra to the experimental data. Furthermore, we use first-principles calculations to determine THz Raman tensor elements and anharmonic coupling coefficients for an additional comparison to experiment (Fig. 1b).

This powerful combination of 2D THz spectroscopy and first-principles calculations provides a unique look at nonlinear vibrational excitation in solid materials.

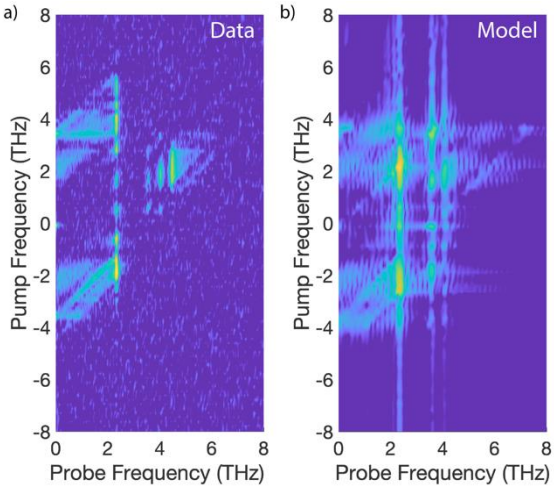

Figure 1 a) 2D THz measurements on $CdWO_4$. B) First-principles model of 2D THz spectra of $CdWO_4$.

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