Predicting 2D THz Spectra Due to Nonlinear Phononics with First-Principles Calculations

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Terahertz (THz) electric field pulses with high peak-field strengths and relatively low photon energies have enabled the observation of fascinating physics in a number of materials [1]. Dynamic control over a crystal lattice using strong THz pulses to influence material properties, study the potential energy surface, and track coherent energy flow in a system are promising and still relatively unexplored areas of study.

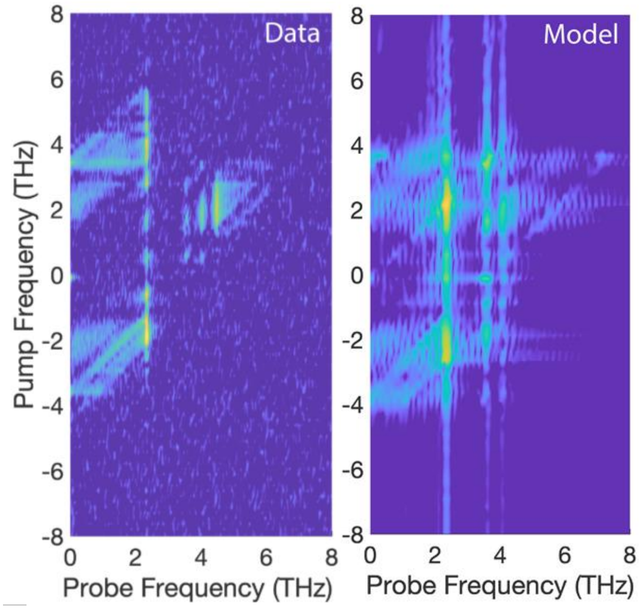

Fig. 1 Experimental (left) and modeled (right) 2D THz spectra.

When intense electromagnetic pulses are used in any kind of pump-probe spectroscopy, several nonlinear excitation pathways can result. Two-dimensional (2D) THz spectroscopy has recently been employed to understand and distinguish competing nonlinear excitation pathways, and it provides a powerful tool to extract microscopic details of the potential energy surface [2,3]. However, 2D spectra can be challenging to interpret. Our presentation will demonstrate that first-principles calculations of anharmonic coupling between phonon modes enable us to predict 2D spectra to further understand nonlinear phononic pathways.

We are able to recreate 2D THz spectra from first-principles, allowing us to better understand the complex couplings that occur between vibrational modes in $CdWO_4$ and other solids. By determining coupling constants from first-principles calculations and modeling 2D THz spectra, we can quantitively analyze experimental spectra. Comparing the modeled and experimental 2D THz spectra determines which mode couplings most influence the potential energy surface. Understanding the microscopic details of the potential energy surface contributes to the budding field of nonlinear phononics and can eventually lead to coherent control of interesting materials.

[1] T. Kampfrath, K. Tanaka, and K. A. Nelson, Nat Photon 7, 680 (2013).

[2] C. L. Johnson, B. E. Knighton, and J. A. Johnson, Physical Review Letters 122, 073901 (2019).

[3] B. E. Knighton, R. Tanner Hardy, C. L. Johnson, L. M. Rawlings, J. T. Woolley, C. Calderon, A. Urrea, and J. A. Johnson, Journal of Applied Physics 125, 144101 (2019).