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2D Fourier-transform Spectroscopy of Potassium Vapor XINGCAN DAI, ALAN D. BRISTOW, DENIS KARAISKAJ, STEVEN T. CUNDIFF, JILA, NIST and University of Colorado — 2D Fourier-transform (2DFT) spectroscopy is a time-domain technique that measures the coherent optical response in two spectral dimensions. This method has elucidated the structure and electronic dynamics in molecules and semiconductors [1], where many-body interactions are expected. Here we demonstrate how 2DFT spectroscopy isolates specific quantum excitation pathways in a simple quantum system, namely potassium vapor in a thin transmission cell [2]. A femtosecond pulse sequence excites both the D1 and D2 lines simultaneously to reveal coupling between the transitions as a result of quantum interference and Raman-like coherences. Observations agree well with numerical simulations based on the optical Bloch equations. The non-radiative Raman pathways have population-time dependence and are isolated by altering the 2DFT projection. Density-dependent measurements show distortion of the 2DFT spectral features due to pulse propagation effects. Unexpected two-quantum coherences are observed and attributed to interatomic interactions.

[1] Cundiff *et al*, Acc. Chem. Res. 42, 1423 (2009).
[2] Dai *et al*, arXiv:1001.1955v1

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