Abstract Submitted
for the MAR12 Meeting of
The American Physical Society

Experimental determination of Hamiltonian via 3D Fourier-transform spectroscopy[1] HEBIN LI, ALAN BRISTOW, MARK SIEMENS, GALAN MOODY, STEVEN CUNDIFF, JILA, University of Colorado and National Institute of Standards and Technology, Boulder, CO 80309 — Prediction and control of quantum mechanical processes requires knowledge about the system Hamiltonian. For coherent control, information about interfering quantum pathways or the underlying Hamiltonian is essential for achieving deterministic control. Even in cases of closed-loop control, a priori knowledge about the system Hamiltonian provides guidance for designing an efficient learning algorithm and a good initial guess for faster convergence. The complete Hamiltonian of a complicated system, especially the effects of inter-particle interactions and coupling to the environments, can only be determined experimentally. Here we demonstrate an experimental determination of the Hamiltonian of an atomic vapor, achieved by using 3-dimensional Fourier transform (3DFT) spectroscopy. The 3DFT spectra provide complete information about the third-order coherent response of the vapor. The contributions from different quantum pathways are unambiguously isolated such that the components of the Hamiltonian, including energy levels, dipole moments and relaxation rates, can be determined. The 3DFT spectroscopy opens an avenue towards identifying the Hamiltonian of complex molecular systems, which can be useful for designing coherent control strategies and for studying the molecular dynamics.

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[1]The work was supported by the NSF Physics Frontier Center program through the JILA AMO Physics Center.

Date submitted: 27 Nov 2011 Electronic form version 1.4