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Transition from Non-Markovian to Markovian Dynamics in a Dense Potassium Vapor VIRGINIA LORENZ, STEVEN CUNDIFF, JILA, University of Colorado and National Institute of Standards and Technology, Boulder, CO 80309 — We have observed non-Markovian dynamics in a dense potassium vapor, whose dephasing time is large compared to the collision duration τ_c, allowing clear separation of time scales, using transient four-wave mixing with pulses short compared to all time scales. This is in contrast to molecular systems limited by ultrafast dephasing due to solvent motion [1]. For pulse delays $\tau < \tau_c$, dynamics are dominated by fluctuations of the transition frequency as the atoms form transient dimers (non-Markovian limit). For $\tau \gg \tau_c$, atomic motions make the system homogeneously broadened (Markovian limit). The theoretical TFWM signal is obtained for a 3-level system in the delta-function pulse limit via a perturbation expansion of the density matrix equations of motion, including non-Markovian dynamics through an assumed correlation function [2], $M_{ij}(t) = \Delta_{ij} \exp(-\Lambda_{ij} t)$, where i and j are energy levels. Theory matches quite well with experimental results at both short (non-Markovian) and long (Markovian) time scales. [1] for a review see G. R. Fleming and M. Cho, "Chromophore- solvent dynamics," Annu. Rev. Phys. Chem. **47**, 109-134 (1996). [2] S. Mukamel, *Principles of Nonlinear Optical Spectroscopy*, Oxford Univ., NY (1995).

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