Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

Resonant Pump-dump Quantum Control of Solvated Dye Molecules with Phase Jumps ARKAPRABHA KONAR, VADIM LOZOVOY, MARCOS DANTUS, Michigan State University — Quantum coherent control of two photon and multiphoton excitation processes in atomic and condensed phase systems employing phase jumps has been well studied and understood. Here we demonstrate coherent quantum control of a two photon resonant pump-dump process in a complex solvated dye molecule. Phase jump in the frequency domain via a pulse shaper is employed to coherently enhance the stimulated emission by an order of magnitude when compared to transform limited pulses. Red shifted stimulated emission from successive low energy Stokes shifted excited states leading to narrow-band emission are observed upon scanning the pi step across the excitation spectrum. A binary search space routine was also employed to investigate the effects of other types of phase jumps on stimulated emission and to determine the optimum phase that maximizes the emission. Understanding the underlying mechanism of this kind of enhancement will guide us in designing pulse shapes for enhancing stimulated emission, which can be further applied in the field of imaging.

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Date submitted: 11 Nov 2013 Electronic form version 1.4