Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

Identifying Single Molecule Dynamics in Real Time via Time-Resolved Coherent Anti-Stokes Raman Scattering[1] STEVEN YAMPOLSKY, DMITRY FISHMAN, SHIRSHENDU DEY, EERO HULKKO, MAYUKH BANIK, ERIC POTMA, V. ARA APKARIAN, Univ of California - Irvine — By enhancing the local response of a single molecule with a dipolar nano-antenna, the vibrational dynamics at the single molecule limit can be measured in real time, by Time-Resolved Coherent Anti-Stokes Raman Scattering (TR-CARS). Such measurements involve the preparation and subsequent probing of vibrational wavepackets on the ground electronic state. In contrast to ensemble measurements, the vibrational coherence of a single molecule is not subject to dephasing; it exhibits dynamic phase and amplitude noise due to the collapse of the wavepacket upon measurement. Continuous measurements of the amplitude noise distribution as a function of phase delay, allows the complete reconstruction of the state of the system. Under ambient conditions, repeated measurements of a single molecule coherence reduces to a statistical state of a system coupled to the thermal bath. The signature of the statistical state of a single molecule is characteristic, distinct from that of two, or three, or many; and this can be directly demonstrated through measurements.

[1]Thank You To NSF for funding the Chemistry at the Space Time Limit Center (CasTL) with (CHE-082913)

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Date submitted: 16 Nov 2013 Electronic form version 1.4