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In-Situ Optical Heterodyne in time Resolved Coherent Anti-Stokes Raman Scattering YEHIAM PRIOR, ANDREY SHALIT, YURI PASKOVER, Department of Chemical Physics, Weizmann Institute of Science, Rehovot, Israel 76100 — We show both theoretically and experimentally that pure vibrational modes cannot be observed in the power spectrum of time resolved degenerate Coherent Anti-Stokes Raman Scattering (CARS), unless the optical signal is linearized by an optical heterodyne detection scheme. A new heterodyne detection method is introduced, where the local oscillator is not provided externally, but is produced *in-situ* by the addition of small amounts of highly anisotropic molecules to the measured sample. The rotational anisotropy of the added molecules gives rise to a slowly evolving signal, which in turn serve as the local oscillator in heterodyned time resolved CARS. We have shown that the strength of the local oscillator can be controlled by amount of material added, in our experiments CS_2added to the measured chloroform or bromoform liquids. Small change of total anisotropy of the sample is expressed as dramatic changes in the intensity of the peaks corresponding to the fundamental vibrational frequencies. This method can be utilized for distinguishing of the fundamental frequencies from beats appearing in the signal due to quadratic (intensity) detection of the nonlinear signal.

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