ULTRAFAST EXCITED STATE DYNAMICS OF NON-LINEAR OPTICAL CHROMOPHORES PROBED BY FS/PS-CARS

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The development of a time-resolved coherent anti-Stokes Raman scattering (CARS) variant has been applied to the excited state vibrational dynamics of model non-linear optical chromophores in the condensed phase. This variant, termed fs/ps-CARS combines two initial femtosecond laser pulses (ω_1 and ω_2) generating vibrational population and a third picosecond narrowband probe to integrate the molecular response. The resulting signal contains reasonable spectral width (ω_3 bandwidth limited with a typical linewidth of ω_3 in the 6-15 cm^{-1}regime) while maintaining the femtosecond time resolution of the first two pulses. This technique proves amenable to a detailed study of the vibrational dynamics of excited state molecules by the addition of a femtosecond excitation pulse (ω_{ex}) with a total time resolution of about 150 fs. As a demonstration, the excited state dynamics, known to internally convert on sub-picosecond timescales, of two model chromophores (para-nitroaniline and N,N-dimethyl-para-nitroaniline) are followed.