

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

BENJAMIN D. PRINCE, MIKHAIL N. SLIPCHENKO, BETH M. PRINCE, ALEX J. BLOM, and HANS U. STAUFFER, *Department of Chemistry, Iowa State University, Ames, Iowa, 50011-3111.*

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 ( $\omega_1$  and  $\omega_2$ ) generating vibrational population and a third picosecond narrowband probe to integrate the molecular response. The resulting signal contains reasonable spectral width ( $\omega_3$  bandwidth limited with a typical linewidth of  $\omega_3$  in the 6-15  $\text{cm}^{-1}$  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 ( $\omega_{ex}$ ) 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.