Relative Phase Change of Nearby Resonances in Temporally Delayed Sum Frequency Spectra

*Fadel Y. Shalhout, Sergey Malyk, and Alexander V. Benderskii**

Department of Chemistry, University of Southern California, Los Angeles, CA 90089

Supporting Information

Table 1. List of parameters obtained by fitting experimental PPP spectra at 0 fs and 300 fs IR-visible delay (shown in Figure 1 of the main text) to Eq. (2) of the main text.

IR-Vis Delay (fs)	A_{NR} (a.u.)	ω_g (cm^{-1})	σ_g (cm^{-1})	ϕ_{NR} (rad)	$B(r^+)$ (a.u.)	$B(r^-)$ (a.u.)	$\Gamma(r^+)$ (cm^{-1})	$\Gamma(r^-)$ (cm^{-1})	$\omega(r^+)$ (cm^{-1})	$\omega(r^-)$ (cm^{-1})
0	0.07	2902	296	-1.5	-1.0	-0.63	20.0	16.5	2915	2975
300	0.04	2940	300	0.39	0.38	-0.28	14.3	14.0	2907	2979

Table 2. List of parameters obtained by fitting simulated PPP spectra at 0 fs and 300 fs IR-visible delay (shown in Figure 3 of the main text) to Eq. (2) of the main text.

IR-Vis Delay (fs)	A_{NR} (a.u.)	ω_g (cm^{-1})	σ_g (cm^{-1})	ϕ_{NR} (rad)	$B(r^+)$ (a.u.)	$B(r^-)$ (a.u.)	$\Gamma(r^+)$ (cm^{-1})	$\Gamma(r^-)$ (cm^{-1})	$\omega(r^+)$ (cm^{-1})	$\omega(r^-)$ (cm^{-1})
0	0.09	2904	206	-1.7	-1.0	-0.63	18.6	16.3	2917	2974
300	0.05	2917	237	-2.6	0.39	-0.50	16.7	13.6	2922	2979

Table 3. Wavelength λ and pulse duration τ parameters used in Eqs. (9) and (10) to simulate the electric fields of the visible and IR pulses.

	λ (nm)	τ (fs)
IR	3440	80
Visible	796	50

Table 4. The amplitudes B, line widths Γ and central frequencies ω of the resonant response for the symmetric (r^+) and asymmetric (r^-) CH_3-stretch vibrational modes, as well as the amplitude and phase of the nonresonant background used in the simulations (Eq. (8) of the main text).

IR-Vis Delay (fs)	A_{NR} (a.u.)	ϕ_{NR} (deg)	$B(r^+)$ (a.u.)	$B(r^-)$ (a.u.)	$\Gamma(r^+)$ (cm^{-1})	$\Gamma(r^-)$ (cm^{-1})	$\omega(r^+)$ (cm^{-1})	$\omega(r^-)$ (cm^{-1})
0	130	245	-0.1	-0.07	12	10	2915	2964
300	$1.5\text{x}10^8$	190	0.1	0.07	12	8	2915	2975

Note: the nonresonant background amplitude A_{NR} for the 300 fs delayed case is large because it represents interaction with the (very weak) leading edge of the visible pulse (see Figure 2 of the main text). The amplitude of the visible pulse at -300 fs could not be quantified as it was below our detection limit, and thus the spectra for the 300 fs delay are fit using A_{NR} as an independent adjustable parameter.