

**Supplementary Information for Excited State Electronic Landscape of mPlum
Revealed by Two-Dimensional Double Quantum Coherence Spectroscopy**

Patrick Konold,^{1,2} Ralph Jimenez^{1,2}

1. JILA, University of Colorado and NIST, Boulder CO 80309

2. Department of Chemistry & Biochemistry, University of Colorado, Boulder CO 80309

Multidimensional Data Processing Procedure

The raw spectral data for all time delays was compiled into a single matrix for data processing and normalized with the square root of the local oscillator intensity. Detected wavelength data was resampled to the frequency domain. Next, spectral filtering along the emission frequency dimension was conducted using Fourier analysis to remove extraneous signal components. Finally, Fourier transformation along the t_2 dimension was carried out to extract excitation frequency information. The excitation frequency vector was calculated in accordance with the Nyquist theorem assuming a temporal sampling interval of 0.89 fs.

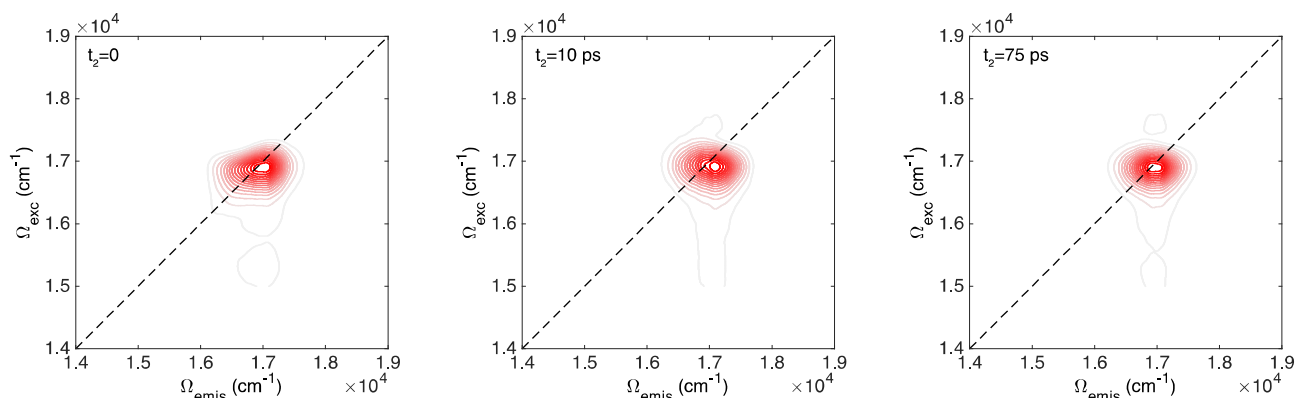


Figure S1. mPlum absolute value 2D1Q spectra at $t_2=0, 10$, and 75 ps.

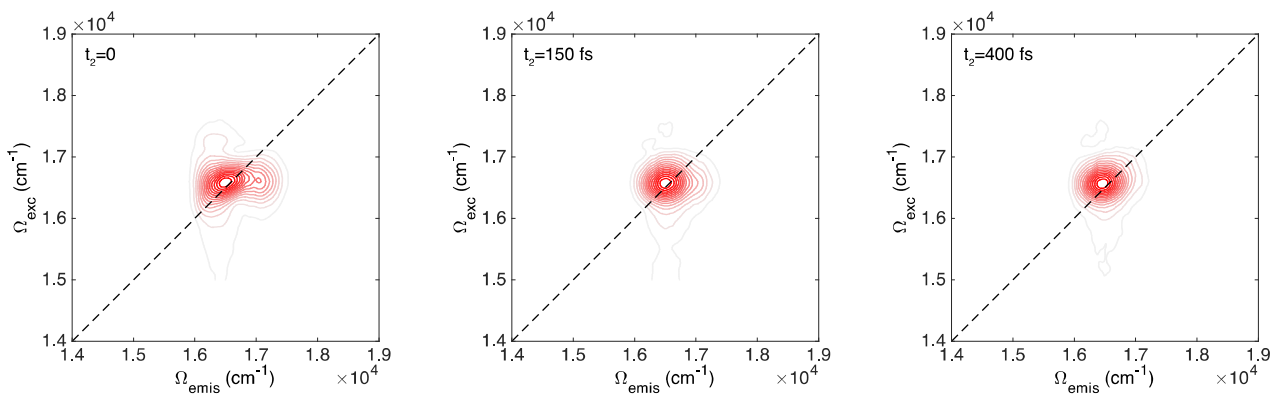


Figure S2. Cresyl violet absolute value 2D1Q spectra at $t_2=0, 150$, and 400 fs.

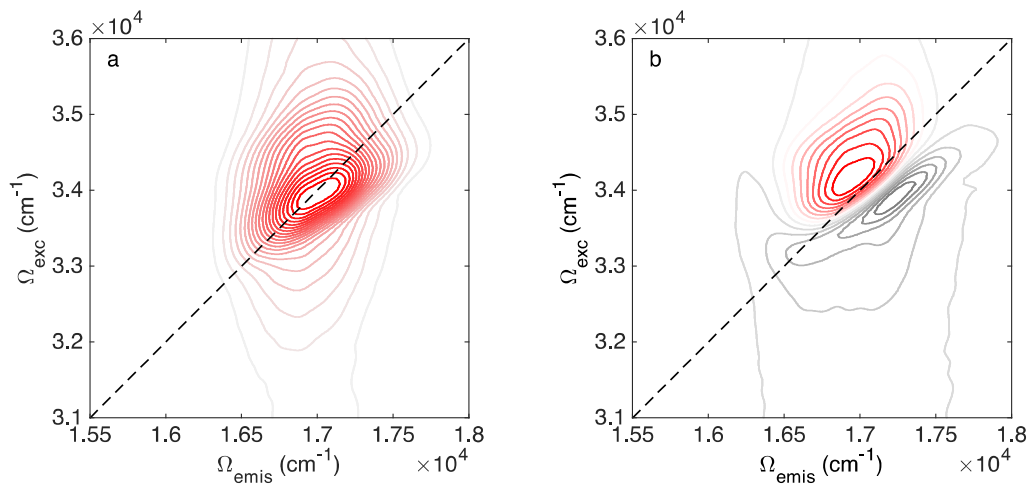


Figure S3. (a) Absolute value and (b) real 2D2Q spectra of cresyl violet in methanol.

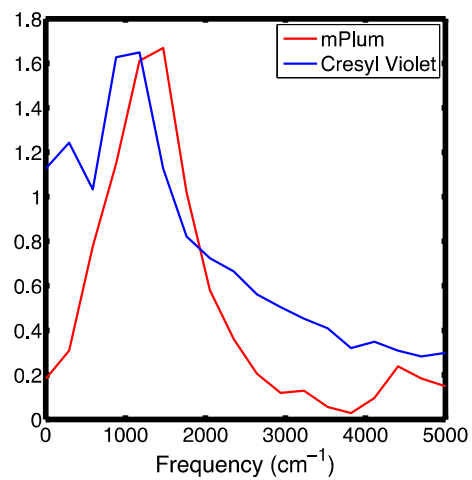


Figure S4. Frequency analysis of integrated FWM signal for cresyl violet and mPlum.

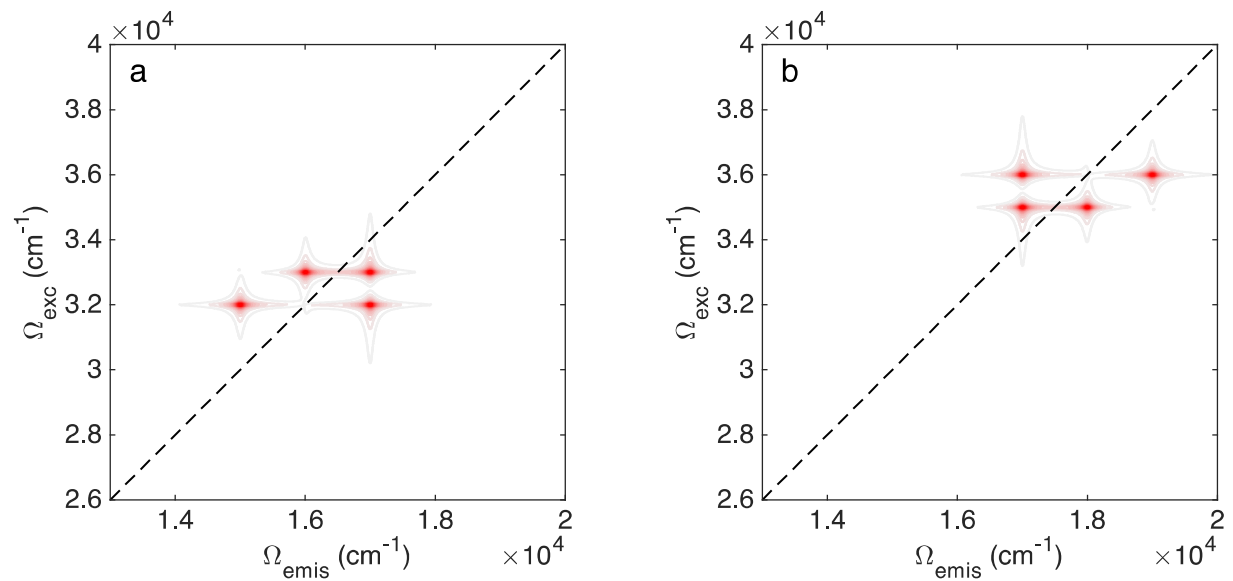


Figure S5. Simulated absolute 2D2Q spectra of two 3LS with (a) negative and (b) positive anharmonicity.

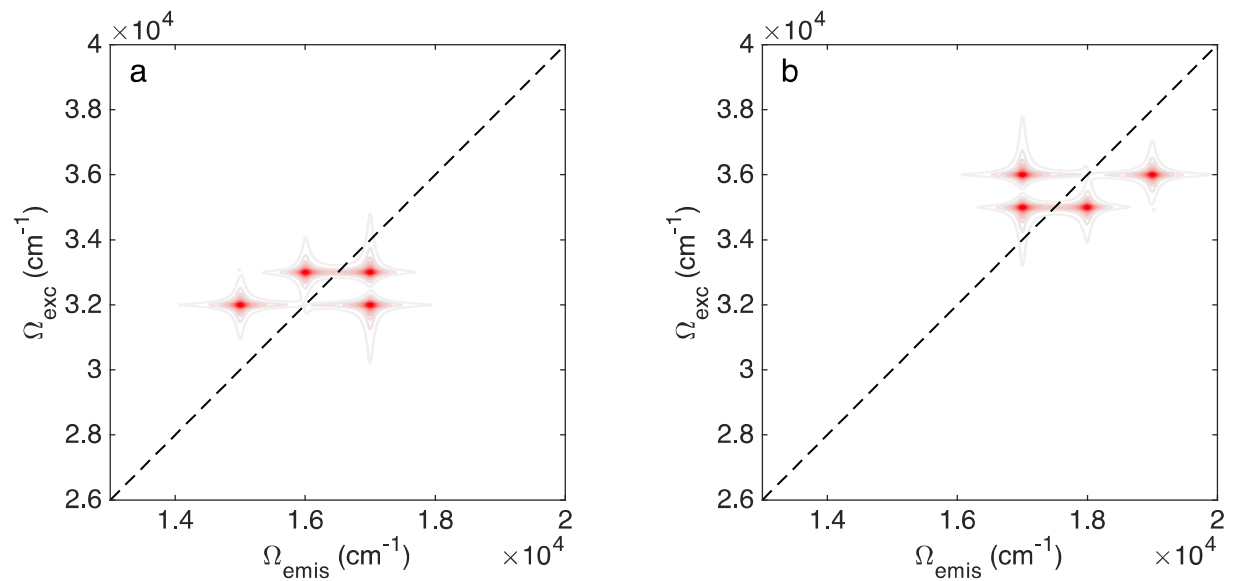


Figure S6. Simulated 2D2Q spectra of the 1-1-2 level scheme with (a) negative and (b) positive anharmonicity.

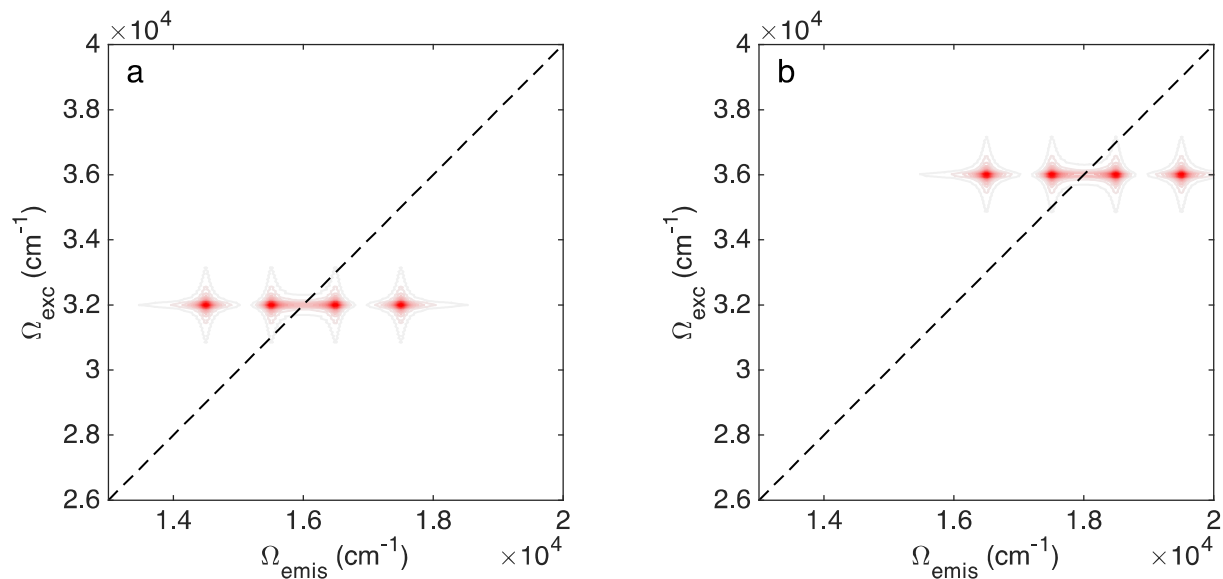


Figure S7. Simulated absolute value 2D2Q spectra of the 1-2-1 level scheme with (a) negative and (b) positive anharmonicity.

Simulation Details

Electronic energy level positions in cm^{-1} used to model double quantum coherence responses of potential level structures.

Two 3LS

I (+-) Equal linear absorption peaks

0

17000

33000

0

17000

35000

II (+-) different LA

0

16500

32000

0

17500

36000

III (++)

0

17000

35000

0

17000

36000

IV (- -)

0

17000

32000

0

17000

33000

1-1-2

I (+-)

0

17000

33000

35000

II (++)

0

17000

35000

36000

III (- -)

0

17000

32000

33000

1-2-1

I (+)

0

16500

17500

36000

II (-)

0

16500

17500

32000