Femtosecond Time-Resolved Infrared-Resonant Third-Order Sum-Frequency Spectroscopy

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S1- 2D Spectrum Simulation

We simulate the 2D spectrum of the film by considering the strong near-resonant absorption and pre-pulse effects. ITS spectroscopy is sensitive to the IR active modes. For the sample we use, they are at 2853 cm-1 and 2926 cm-1 separately according to the FTIR spectrum. The full width at half maximum (FWHM) of the 2853 cm^{-1} and 2926 cm^{-1} absorption lines are set as 40 cm^{-1} and 95 cm^{-1}, respectively. In this simulation, we assume that the probe NIR pulse contained a pre-pulse, which is π phase difference from the main pulse. The pre pulse is due to the edge bandpass filter, which results in a sinc function for the pulse. The simulated results are shown in fig. S1.

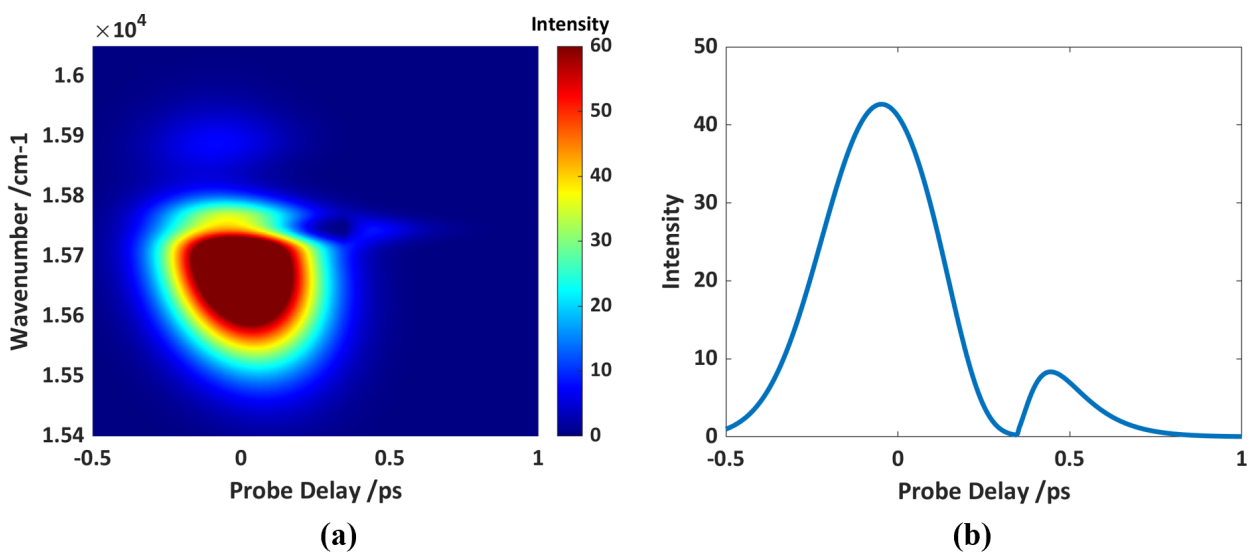

Figure S1. (a) Simulated 2D spectrum of the time-resolved ITS signal generated from the film. The X-axis is the delay time between the NIR and MIR pulses. The zero probe delay is the time

when the two pulses overlaps. Y-axis is the wavenumber of the visible signal. (b) Signal intensity changes with the time delay at 15740 cm^{-1}. X-axis is the time delay. Y-axis is the intensity.