Supplemental Document

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Amplification of mid-IR continuum for broadband 2D IR spectroscopy: supplement

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Supplemental information for

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S1. Spatial profile measurement

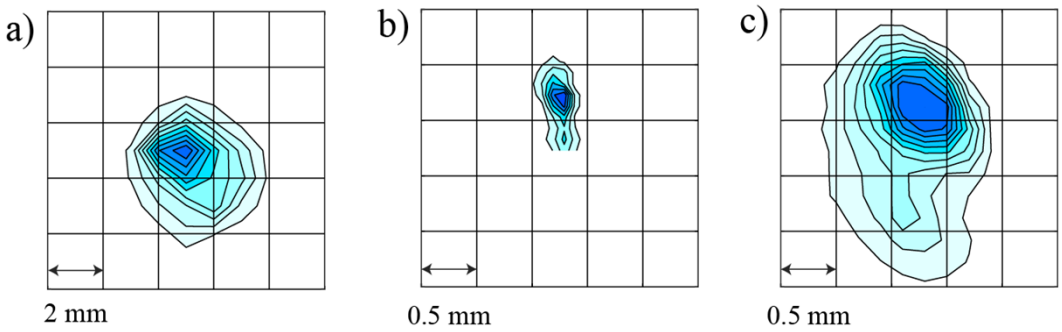

Fig. S1. Spatial profile measurements of (a) the collimated seed after spatial filtering, (b) seed at the position of the GaSe crystal, and (c) pump at the position of the GaSe crystal.

Fig. S1 shows the spatial profile characterization of seed and pump beams. Measurements were made with the methodology described in the main text. A room temperature InSb detector was mounted on a biaxial stage behind a pinhole. The pinhole was placed at the measurement location and scanned in the two transverse directions to map out the beam spatial profile. The collimated seed profile (a) was measured using a 400 μm diameter pinhole, after the seed spatial filter. The beam diameter is approximately 5 mm $1/e^2$. The seed (b) and pump (c) were both measured at the location of the GaSe front face by removing the crystal and placing the pinhole at the same location, which is approximately 25 mm in front of the focus. A 100 μm diameter pinhole was used for these measurements. The measured beam diameters at the crystal position are approximately 0.5 mm and 1.6 mm for seed and pump, respectively. Notably, the pump diameter is larger than the seed at the crystal. This is by design, as this configuration amplifies most of the seed using the more intense portions of the pump.

S2. Beam diagram for FROG characterization

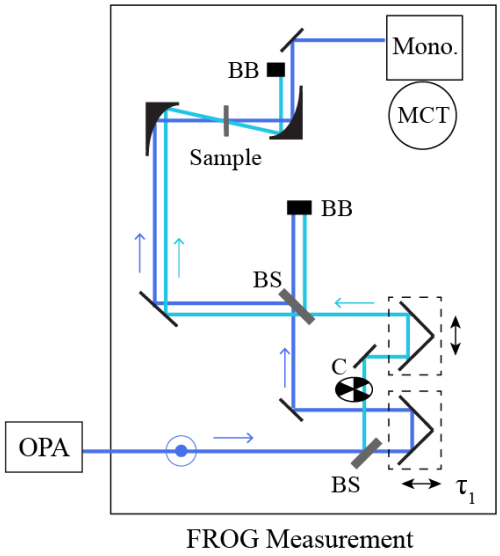

Fig. S2. Beam diagram for FROG characterization. Legend: BB, beam block; BS, beamsplitter; C, optical choppter; MCT, HgCdTe detector; Mono., monochromator. Color scheme: dark blue, input pulse and interferometer moving arm; light blue, linterferometer stationary arm.

For FROG measurements, the interferometer and spectrometer in Fig. 1 are modified as displayed in Fig. S2. The CaF_2 wedge is removed and the stationary stage in the interferometer is displaced horizontally such that the output of the interferometer is two parallel, noncollinear beams. The two copies of the pulse are crossed at the focus in a 0.5-mm Si window (for signal) or 1-mm Ge window (for idler). The stationary arm is chopped at 500 Hz and blocked after the sample. Changes in the intensity of the moving arm are measured as a function of wavelength and interferometer delay τ_1 with the monochromator and MCT detector. Pulse intensity was attenuated with an iris before the first BS to prevent two-photon absorption in the semiconductor at the sample position.

S3. Signal FROG characterization

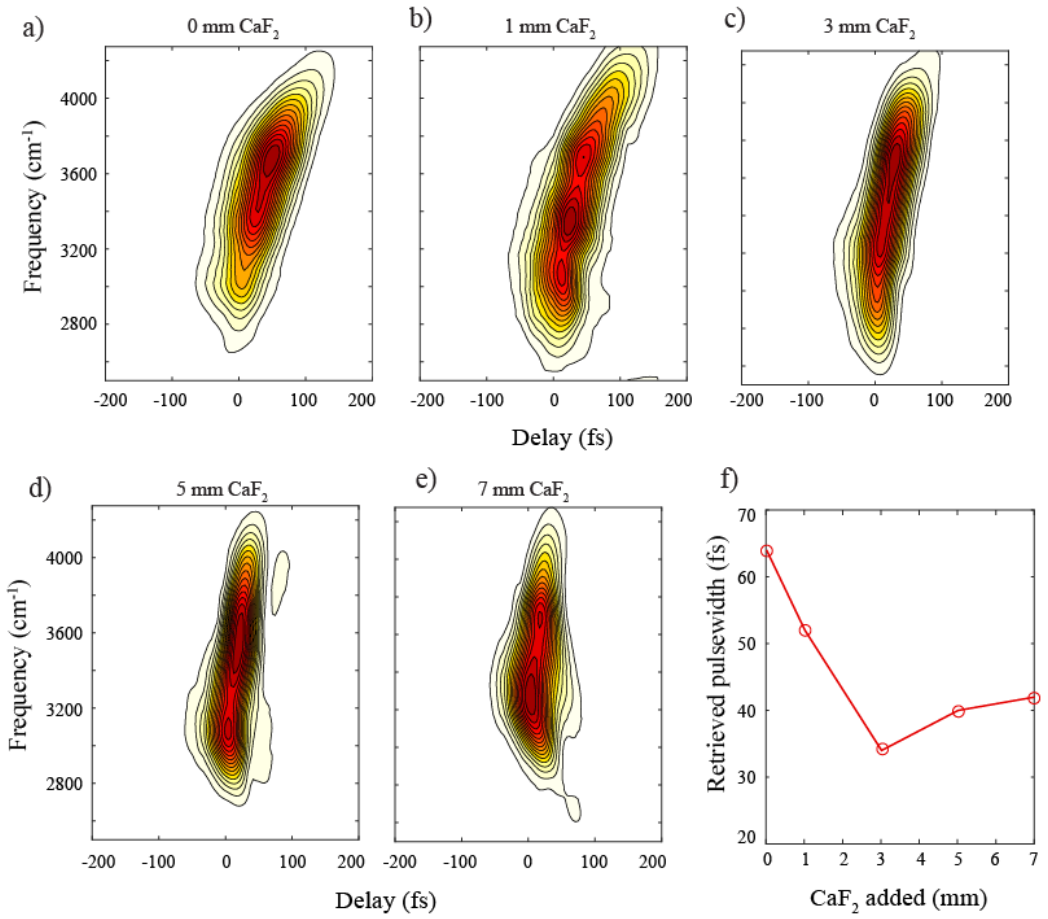

Fig. S3. FROG characterization of the signal pulse as a function of CaF_2 bulk material added before the interferometer. (a-e) Experimental FROG traces using the nonresonant response in 0.5 mm Si. (f) Retrieved pulsewidth as a function of CaF_2 added.

Signal compression was measured as a function of CaF_2 bulk material added to the beam before the interferometer. FROG traces were collected using nonresonant response in 0.5 mm Si as described in the main text and section S2. Fig. S3 shows the measured FROG spectrogram (a-e) with increasing CaF_2 from 0 to 7 mm. The retrieved pulse width is minimized at 34 fs when 3 mm CaF_2 is added.

S4. Instrument response function

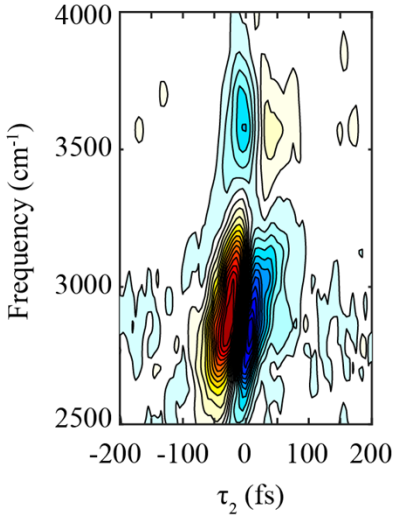

Fig. S4. Transient absorption of a 1 mm CaF_2 window using the compressed signal for excitation and detection.

The instrument response function of the 2D IR instrument described in Fig. 1 in the main text is determined by the nonresonant response of the 1-mm CaF_2 window before the sample. Fig. S4 displays the transient absorption of a 1-mm CaF_2 window using the signal for both excitation and detection. To reproduce the conditions of the 2D IR measurements reported in Fig. 4 in the main text, both excitation and detection pulses pass through a total of 2 mm CaF_2 before reaching this window in the focus. The window nonresonant response has decayed by 100 fs waiting time (τ_2) or earlier.