Supplemental Document

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Hybrid CO_2-Ti:sapphire laser with tunable pulse duration for mid-infrared-pump terahertz-probe spectroscopy: supplement

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A Hybrid CO_2-Ti:sapphire Laser with Tunable Pulse Duration for Mid-Infrared-Pump Terahertz-Probe Spectroscopy

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Supplementary Material

S1 – Optical parametric amplifier for injection seeding

S2 – Beam parameters of the optical setup

S3 – Pulse rise time dependence in semiconductor switching

S1 – Optical parametric amplifier for injection seeding

Optical parametric amplification of femtosecond pulses in combination with difference frequency generation was used in this work to convert near infrared pulses from 800 nm to 10.6 μm wavelength, which were subsequently used for injection seeding of a CO_2 laser (see setup schematic in Fig. S1).

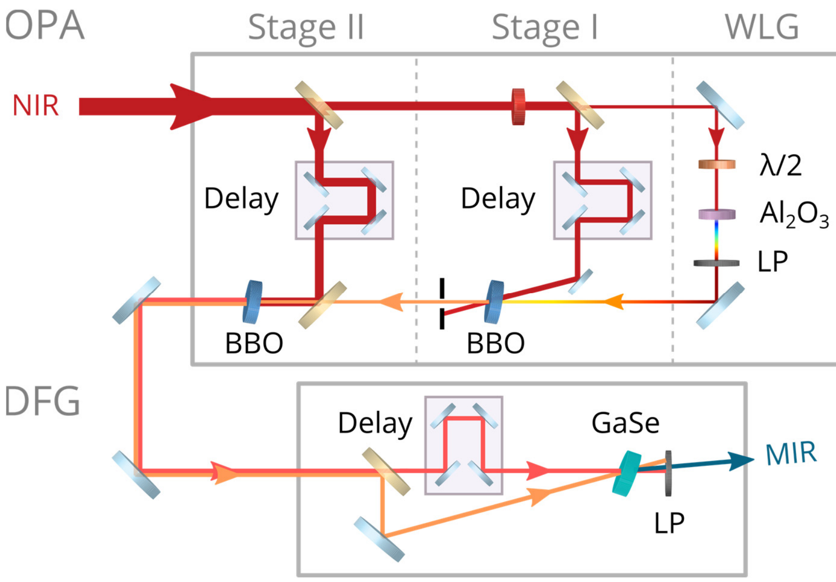

Figure S1: Starting from the 800 nm input pulses, broadband white light is generated in a sapphire crystal (WLG). Starting from this seed, the first OPA stage (based on a type II BBO crystal) generates a signal beam centered around 1.48 μm. This is then amplified further in a second BBO based OPA stage. The signal and idler beams from the last stage are then mixed in a GaSe crystal to obtain MIR pulses centered at 10.6 μm.

Here, the 800 nm input beam was generated from a conventional Ti:Al_2O_3 amplifier delivering an energy per pulse of about 3 mJ. After the pump light for the two OPA stages has been branched off by two beam splitters, a small portion of a few μJ remained for the generation of s-polarized white light in a 2 mm thick Al_2O_3 crystal. From its broadband spectrum, the visible part was removed by a low-pass filter (LP in Fig. S1) before it was focused on a 3 mm thick, type-II BBO crystal, cut at 28° for optimum phase matching. Here it was non-collinearly overlapped with the first 800 nm pump beam of about 200 μJ pulse energy resulting in the generation of about 5 μJ of λ_{sig} = 1.48 μm wavelength pulses. This is amplified in a second collinear OPA stage (3 mm thick, type-II BBO crystal, cut at 28°) pumped with about 2.7 mJ energy pulses centered at 800 nm and resulted in an amplified signal and an additional idler beam of λ_{idl} = 1.74 μm wavelength. The total output energy of the cross-polarized beams was about 800 μJ. The angles of the BBO crystals were tuned to give the above output wavelengths so that DFG of the signal and idler beams in a 1.5 mm thick GaSe crystal yielded up to 12 μJ of 10.6 μm wavelength radiation with a stability of 0.5 % and a pulse length of about 150 fs.

The wavelength of the DFG setup was verified by means of linear FTIR spectroscopy before the beam was used for injection seeding of the CO_2 laser.

S2 – Beam parameters of the optical setup

The beam parameters used in the optical setup depicted in Fig. 1 in the main text are summarized in the following table:

Pos.	**Description**	**Diameter $1/e^2$**	**Pulse energy**	**Pulse duration**	**Central wavelength**	**Rep. rate**
1	Ti:Sa oscillator Coherent Vitara S	2.0 mm	6 nJ	< 40 fs	800 nm	80 MHz
2a	Ti:Sa amplifier Coherent Legend Elite	11.0 mm	3.2 mJ	~ 100 fs	800 nm	900 Hz
2b	Ti:Sa amplifier Coherent Legend Elite	11.0 mm	4.5 mJ	~ 100 fs	800 nm	900 Hz
3	OPA first amplification stage	4.5 mm	5 µJ	~ 150 fs	1490 nm	900 Hz
	OPA second amplification stage	5.0 mm	800 uJ	~ 150 fs	1490+1730 nm	900 Hz
4	After difference frequency gen.	5.0 mm	10 uJ	~ 150 fs	10.6 µm	900 Hz
	Seed radiation in front of cavity	5.0 mm	60 nJ	~ 150 fs	10.6 µm	900 Hz
5	Before pockels cell (pulse train)	5.0 mm	40 mJ	~ 1 us	10.6 µm	18 Hz
	After pockels cell (single pulse)	5.0 mm	0.5 mJ	~ 1.3 ns	10.6 µm	18 Hz
6	After 10-pass amplifier	5.0 mm	10 mJ	~ 1.3 ns	10.6 µm	18 Hz
7	Before first slicing wafer	3.0 mm	10 mJ	~ 1.3 ns	10.6 µm	18 Hz
	Before second slicing wafer	3.0 mm	1 mJ	~ 1.3 ns	10.6 µm	18 Hz
8	After second slicing	3.0 mm	55 uJ	100 ps	10.6 µm	18 Hz

S3 – Pulse rise time dependence in semiconductor switching

Although the reflectivity rise time in a plasma mirror should mainly depend on the excitation pulse length, which for our laser is about 100 fs, the minimum rise time in our setup was on the order of 3 ps. This was caused by geometrical constraints of the optical setup. Figure S2a displays the geometry of the sliced MIR beam and the NIR beam used for plasma generation. For a finite angle α the distances a and b are not the same. If the two pulses are perfectly overlapped in time at the left-most contact point with the semiconductor, this results in an overlap mismatch at the right-most contact point that corresponds to a temporal delay of $\Delta c = a - b \;\; c$, with c being the speed of light. Such a delay effectively limits the slicing response time when integrating the intensity spatially across the beam. Figure B.2 b shows this delay for different angles and beam diameters. The black circle indicates the situation that was realized in our setup, which explains why the measured cross-correlations of sliced pulses were always limited to rise-times of several picoseconds.

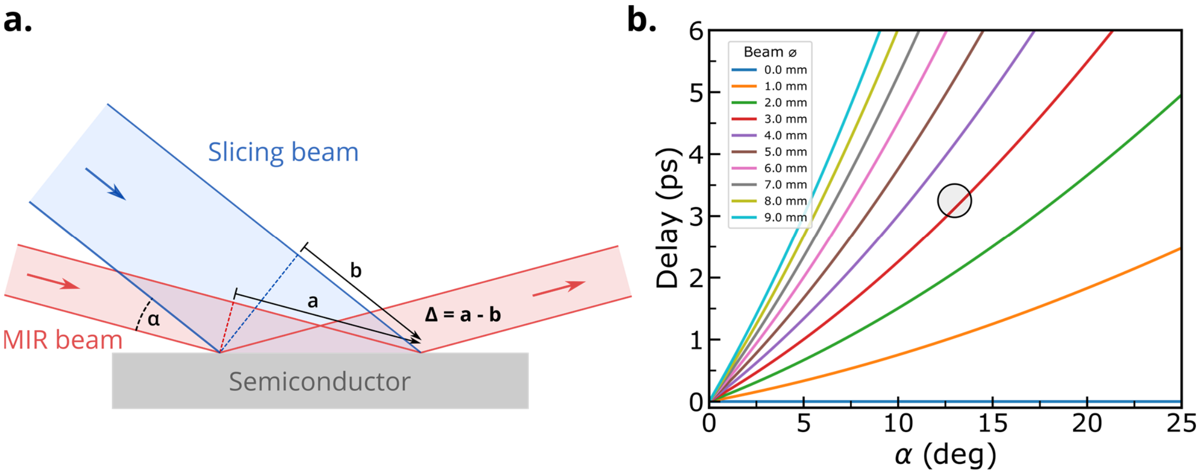

Figure S2: a. Schematic of the slicing geometry. Δ is the maximum path length difference. **b.** Calculation of the resulting time delay for different angles α and beam diameters. The black circle indicates the region of operation in our setup.