An Echelon-based Single Shot Optical and Terahertz Kerr Effect Spectrometer

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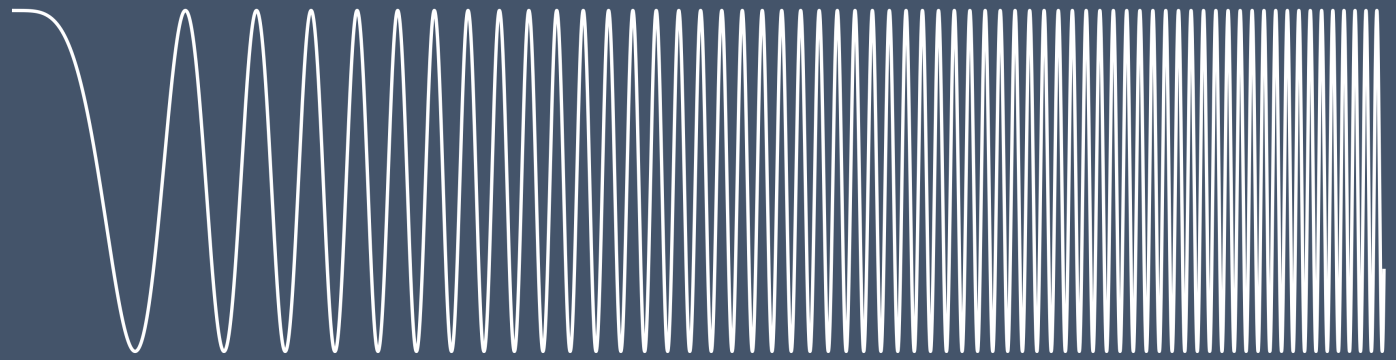
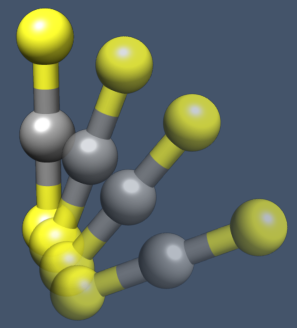
What are THz-domain phenomena?

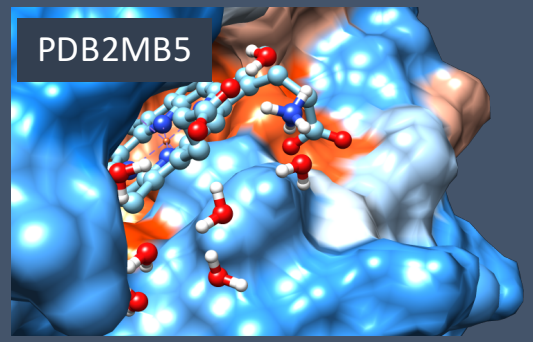


0.1 – 10 THz
0.4 – 40 meV
3.3 – 333 cm^{-1}

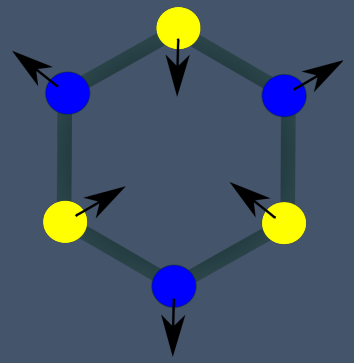
$k_BT \approx$ 6.2 THz at 298 K



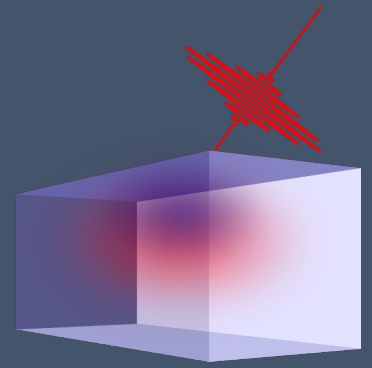
Liquid dynamics



Water of hydration



Phonon mode couplings



Charge carrier dynamics

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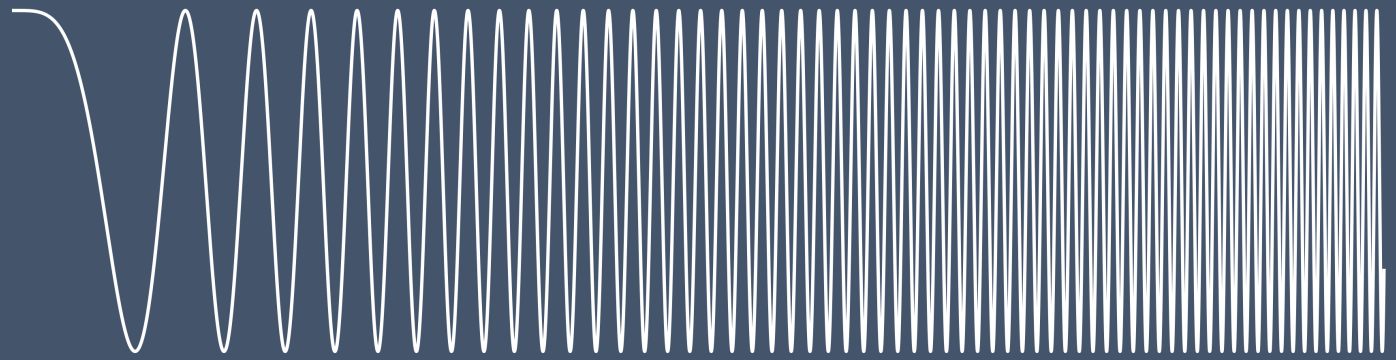

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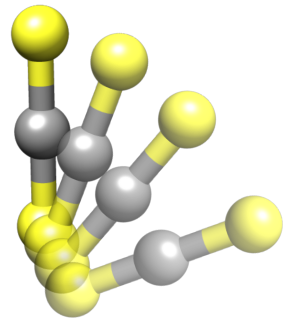

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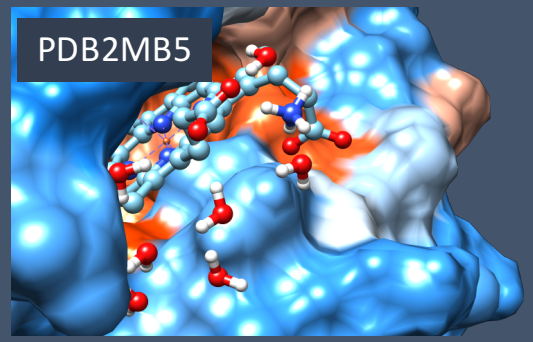

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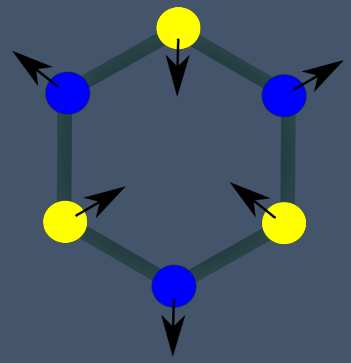

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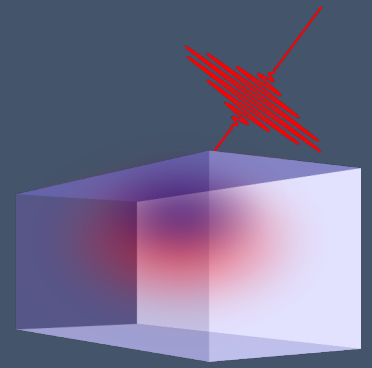

Charge carrier dynamics

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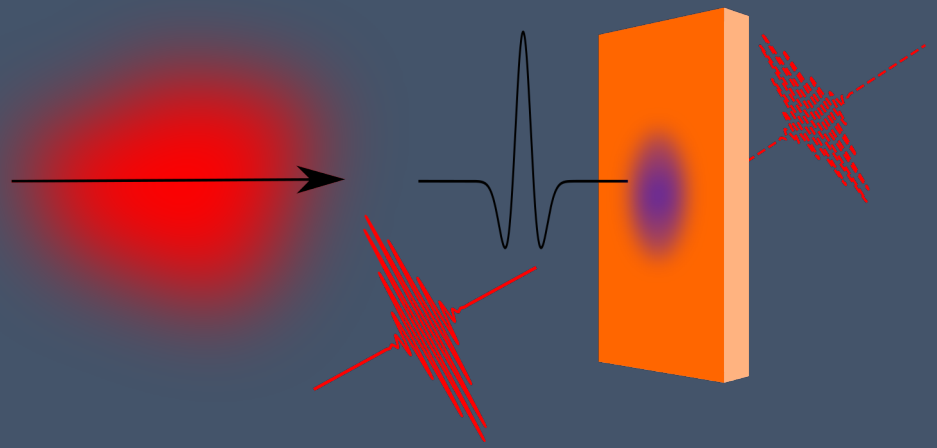
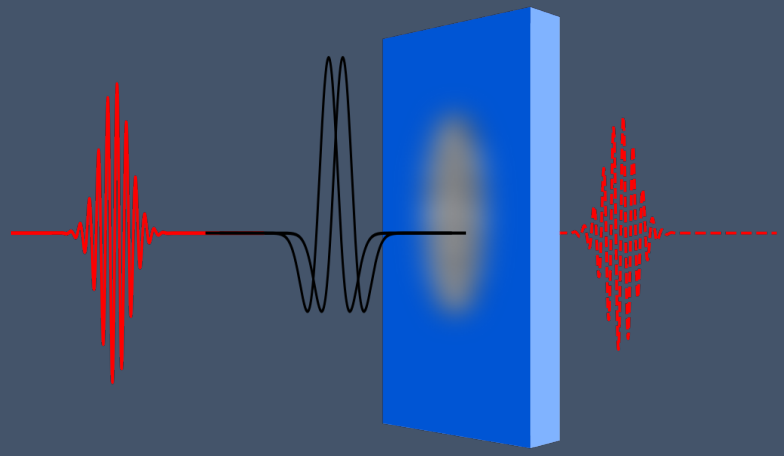
… and how can we study them?

Linear approaches
Electro-optically sampled THz spectroscopy



THz-TDS $\propto E_{THz}$

Nonlinear approaches
Kerr effect spectroscopies



Terahertz Kerr Effect (TKE) $\propto \mu \cdot E_{THz}^2$
Optical Kerr Effect (OKE) $\propto \alpha \cdot E_{Opt}^2$
Reports on ps timescale liquid dynamics

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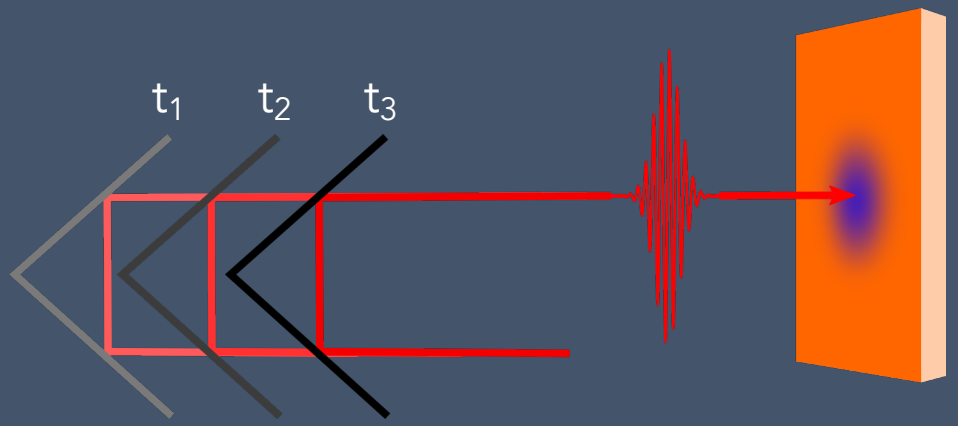
Time-domain measurements



The Dilemma

Cannot directly resolve THz signals with dispersive detectors. Instead, scanning mechanical delay stages are used.

Scanning stages can be time intensive, especially for multidimensional studies:

$$E_{Probe} \overset{t2}{\leftrightarrow} E_{THz} \overset{t1}{\leftrightarrow} E_{THz}$$

Can we make the same Kerr effect measurements without a motorized stage? If so, how much faster could data be collected?

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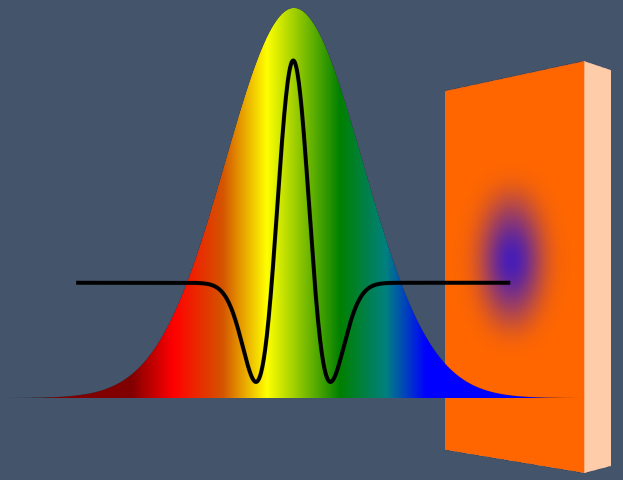
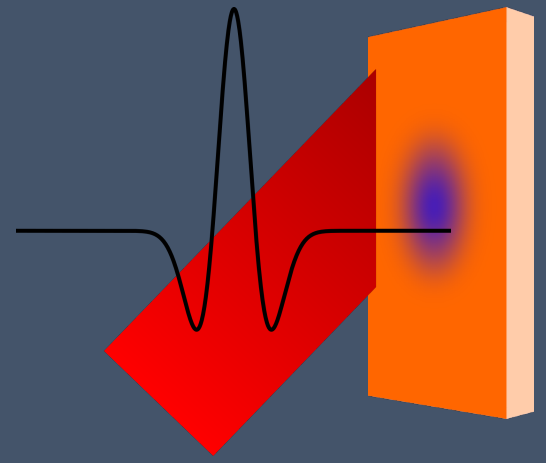
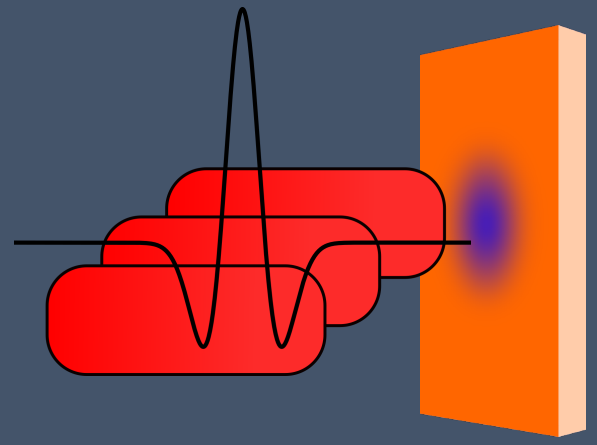
Single Shot THz Time-Domain Spectroscopy



1. Time-to-wavelength
Chirped probe pulse
Spectrometer detection



2. Time-to-angle
Non-colinear probe pulse
CCD detection



3. Time-to-space
Temporally dispersed probe pulse
CCD detection

Review of single-shot THz TDS techniques:
Teo et al., *RSI*, 2015, 86(5), 015301

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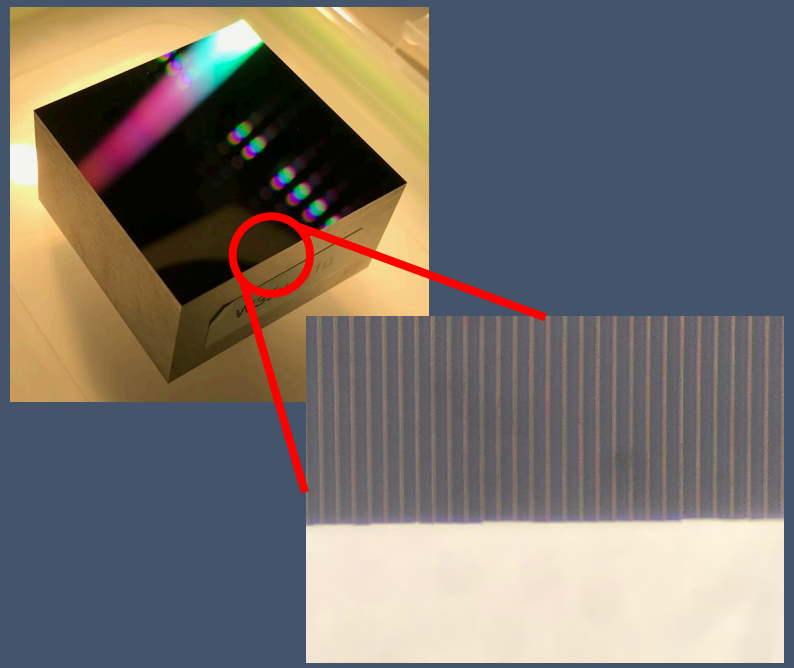
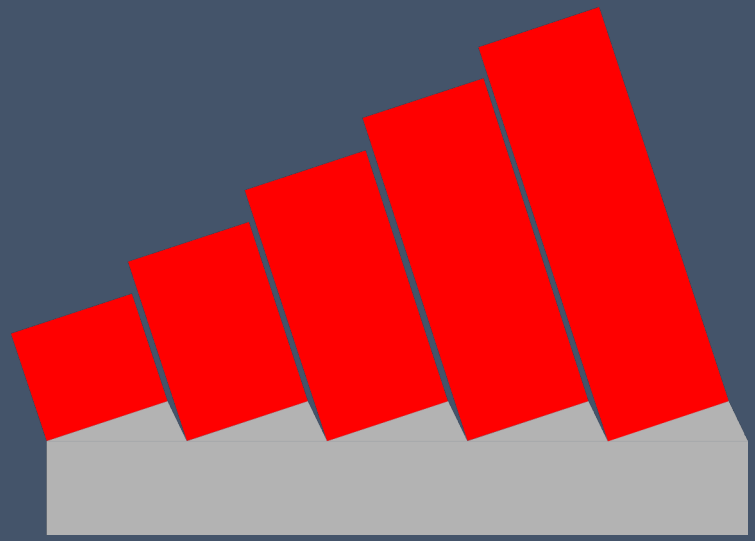
Time-to-space mapping with a reflective echelon



Reflective nickel echelon
1000 steps, CNC machined



Temporal duration and resolution are set by the echelon step width and height

Echelon-based THz-TDS spectroscopy
Noe et al., *Opt. Exp.*, 2016, 24(26), 30328

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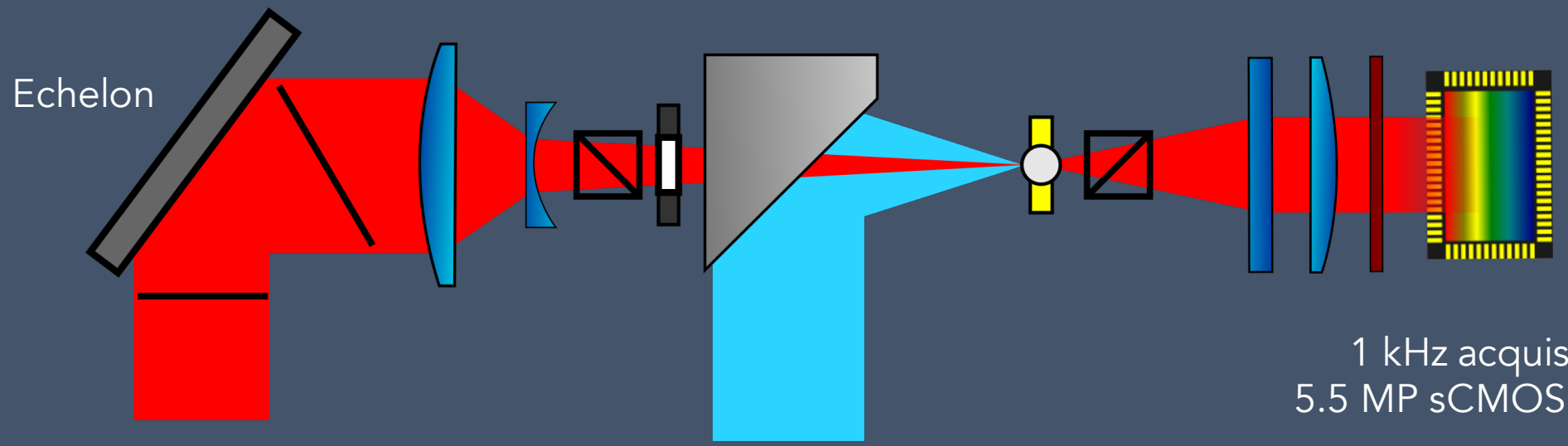
OKE/TKE Spectrometer Design



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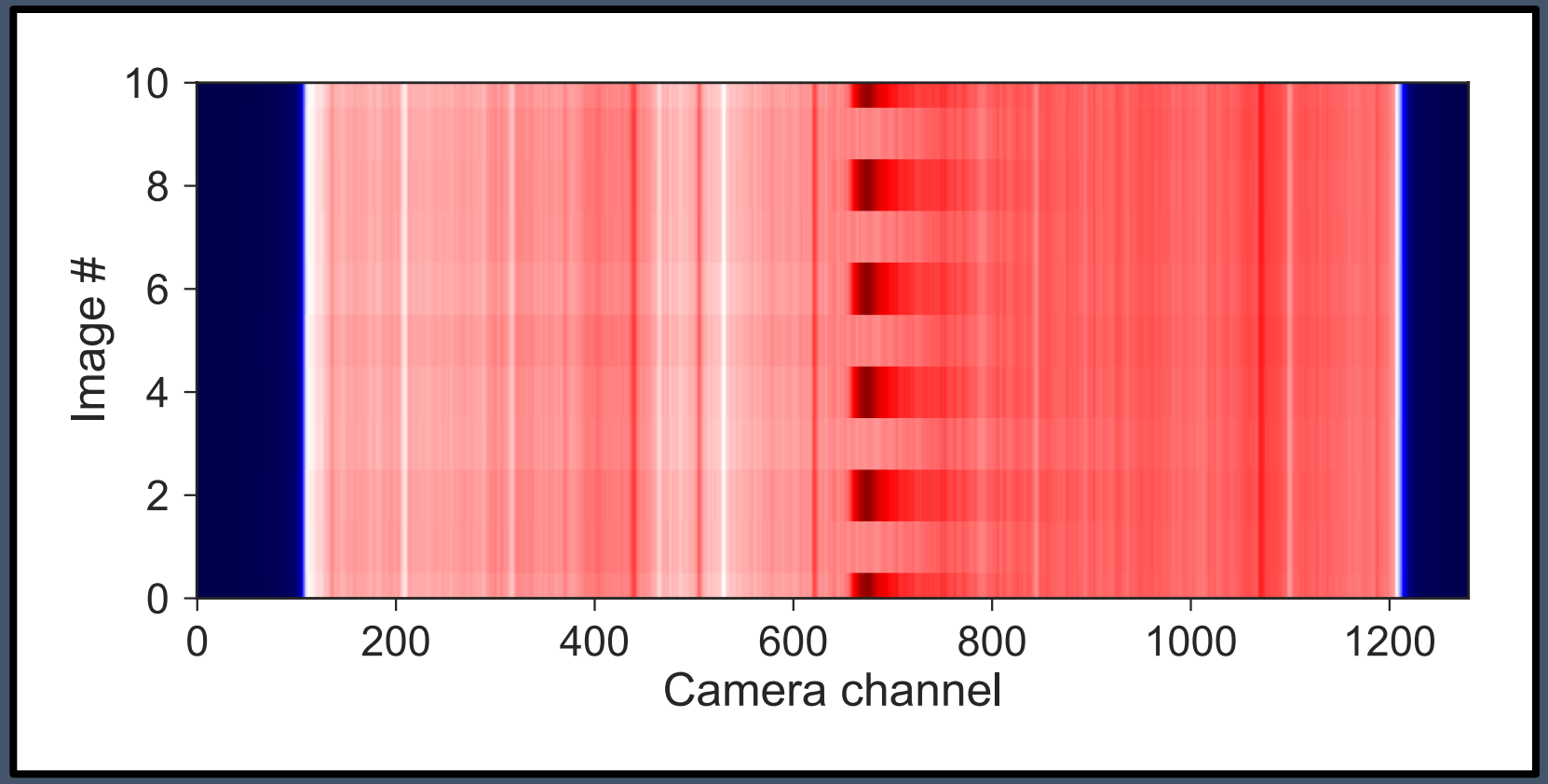
OKE/TKE data processing

On-Off Chopping
-Simple approach
-Easiest to implement at 500 Hz chopping frequency



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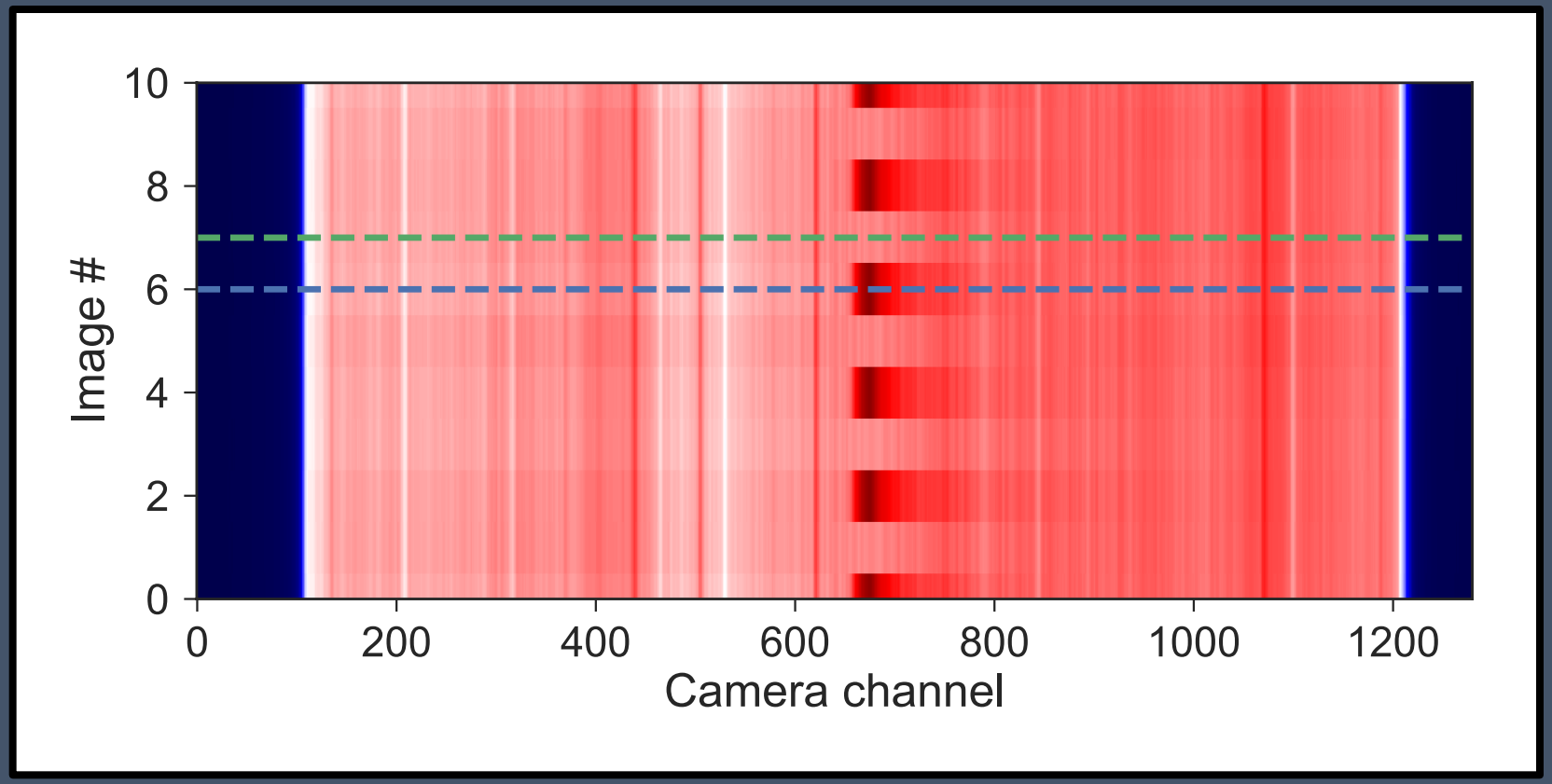

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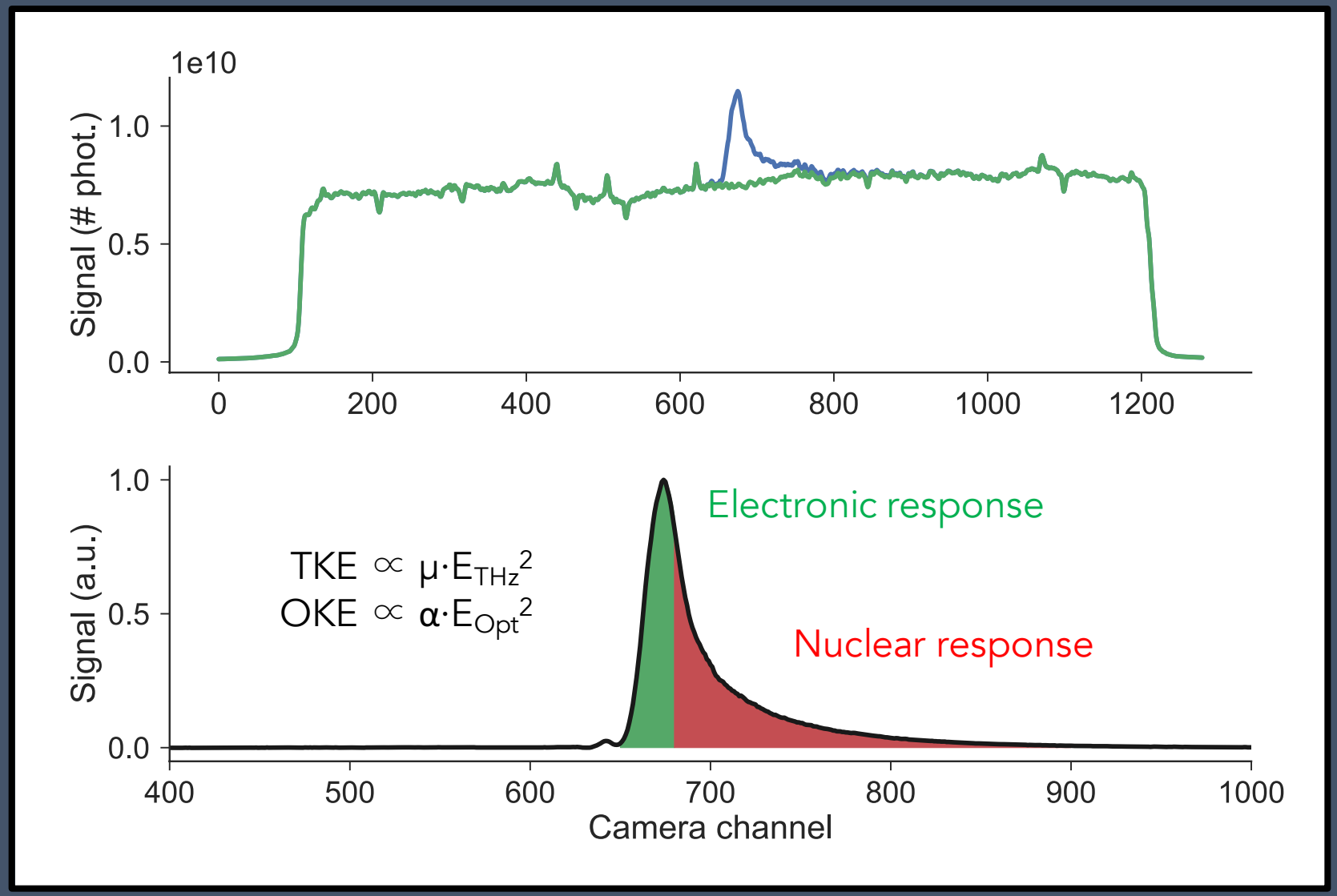

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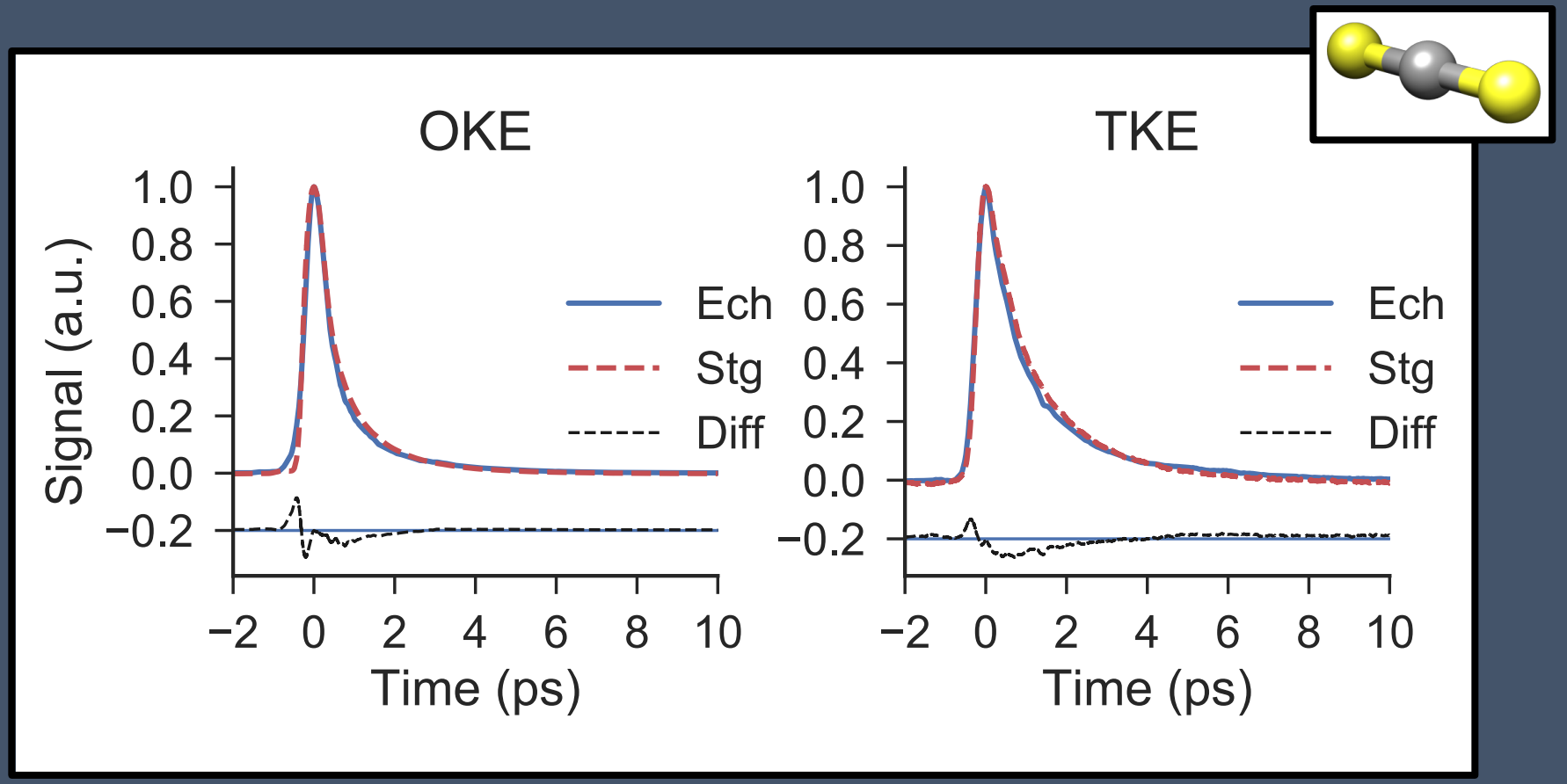
Results

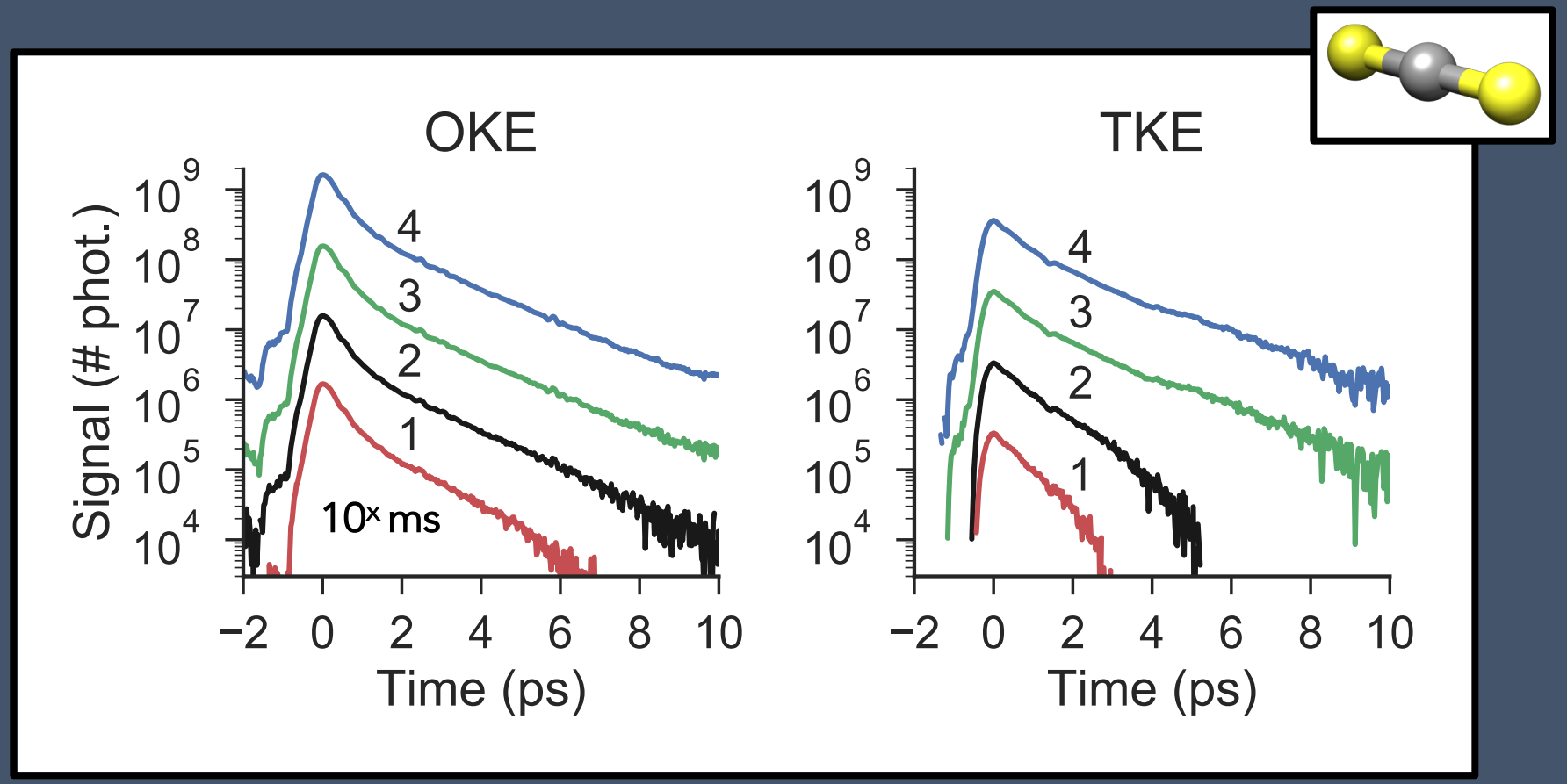
Carbon disulfide (CS_2)
- large $\chi^{(3)}$ constant
- easily detected Kerr response



Results

Photon counting provides a quantitative measure of signal intensity

>10 ps of dynamics collected in only 10-100 ms



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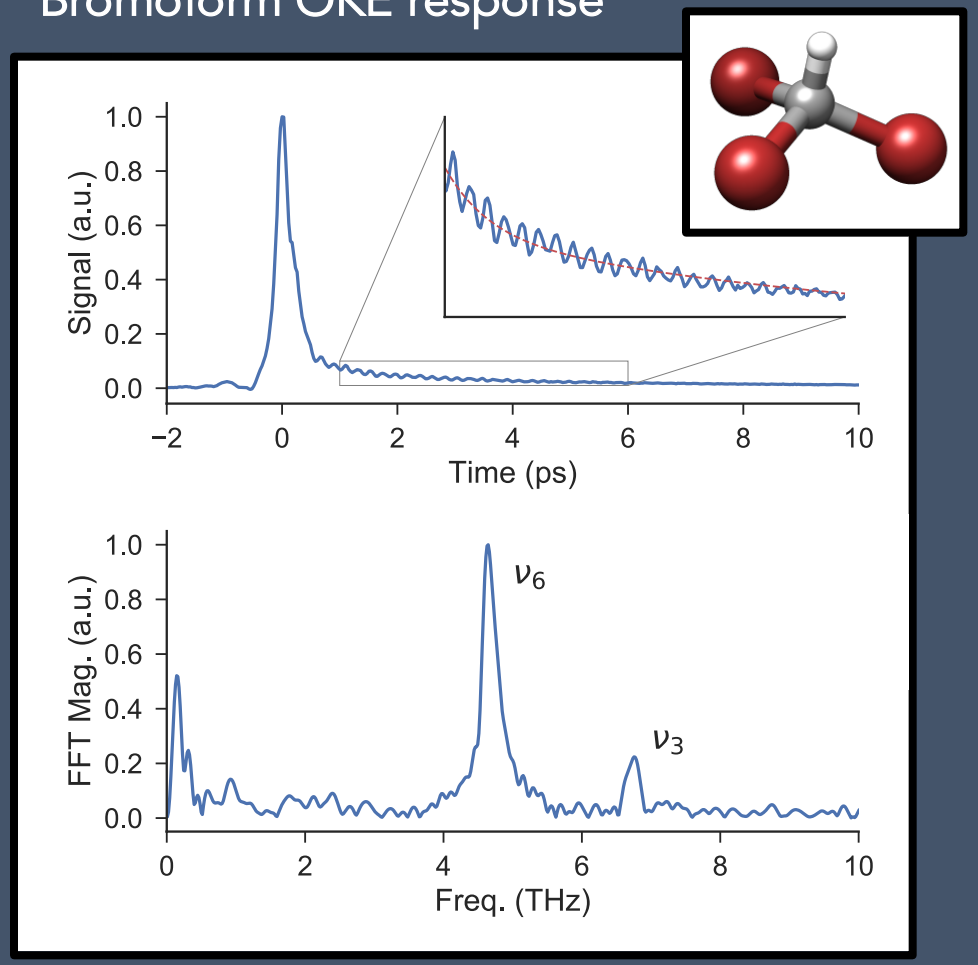
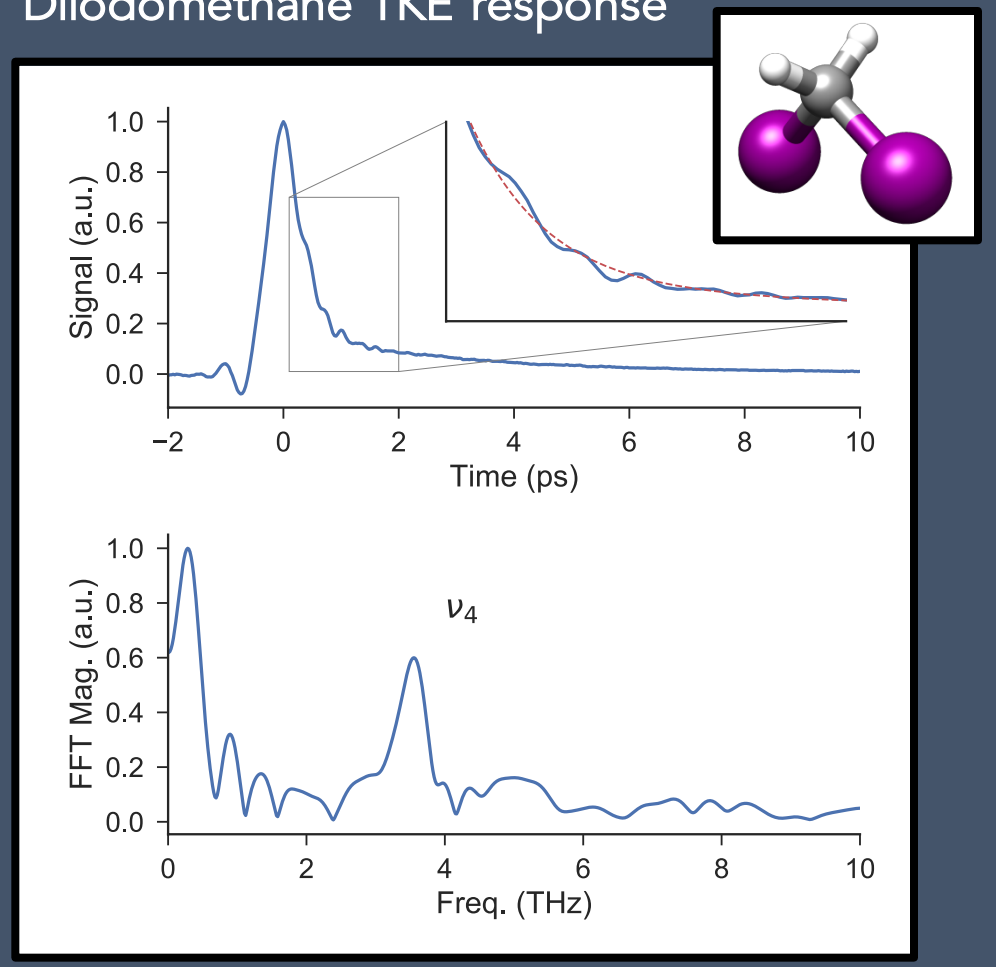
Detection of intramolecular vibrational coherences

Bromoform OKE response



Diiodomethane TKE response



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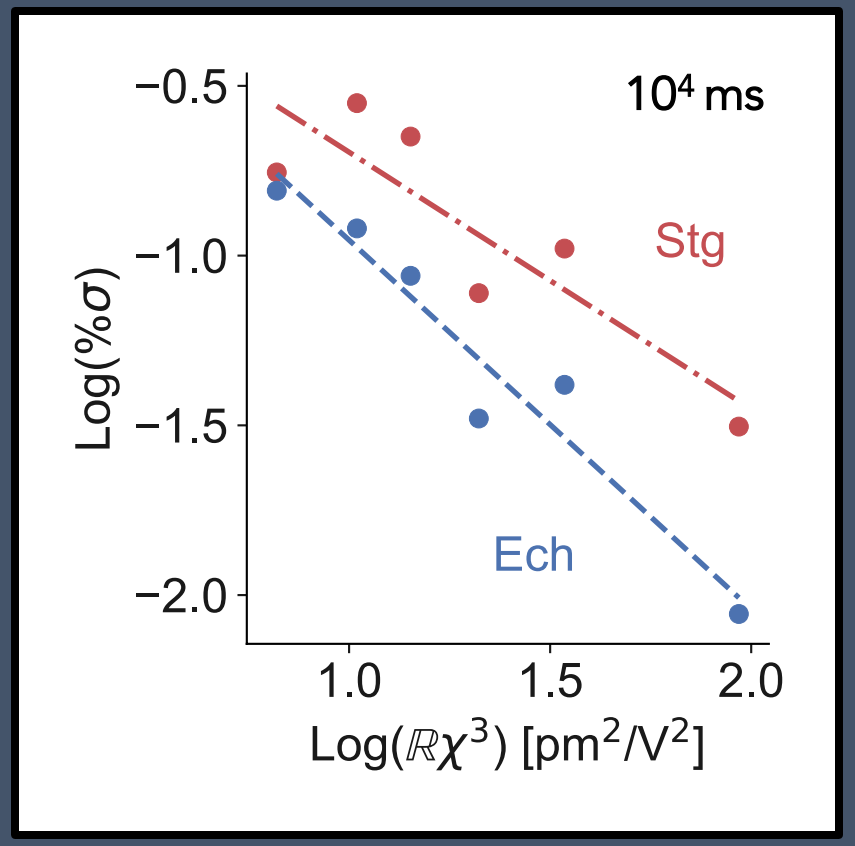
Sensitivity and SNR

Molecule	Re$\chi^{(3)}$ [pm^2/V^2]*
Carbon disulfide	93.17
Benzene	34.34
Nitrobenzene	21.02
Dimethyl sulfoxide	14.22
Acetone	10.46
Acetonitrile	6.61



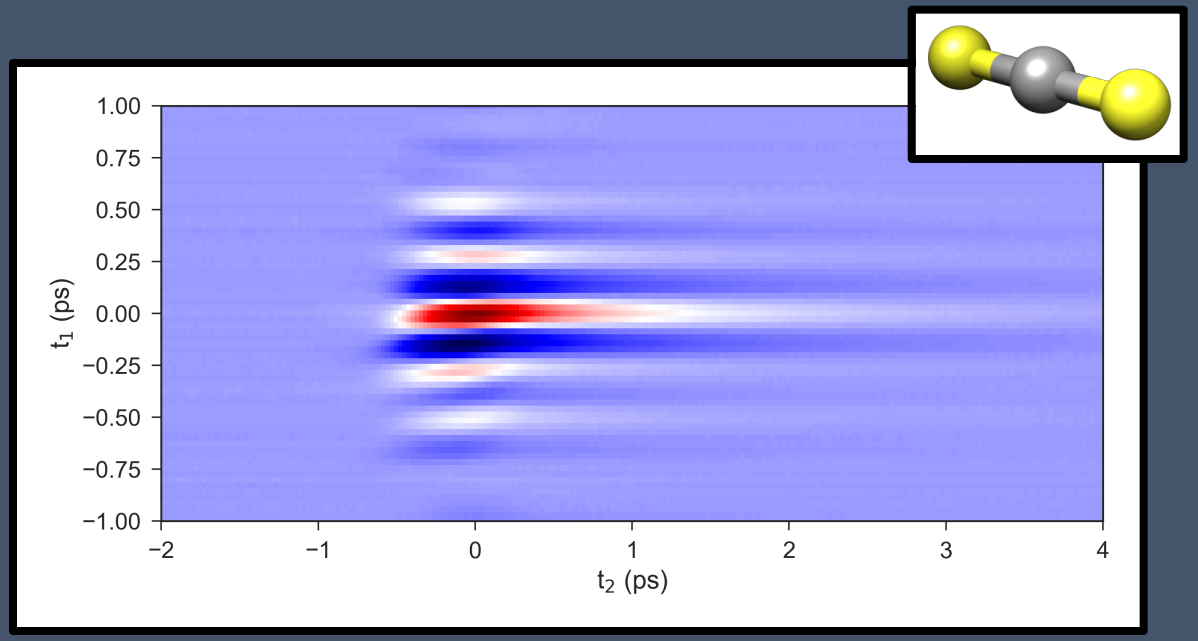
*Iliopoulos *et al.*, *Opt. Exp.*, 2015, 23(19), 24171

Summary

1. The single shot technique can measure nonlinear Kerr effect signals in liquids

2. Data spanning 10s of picoseconds can be acquired in as few as 10 ms

3. Further instrumental development will yield improved performance and speed



Going forward: single shot 2D THz results on CS_2 are promising

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Acknowledgements

Experimental Collaboration:
Ikufumi Katayama (Yokohama National University)
Jun Takeda (Yokohama National University)

Theory Collaboration:
P3955: Simulating the THz-THz-Raman spectrum of molecules: application to bromoform
Ioan-Bogdan Magdău, Thomas F. Miller III
Chemical and Life Sciences B102
Today at 5 PM

Publication on the single shot technique:
Mead *et al.*, *RSI*, 2019, *90*(5), 053107

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