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Title: Ultrafast control and monitoring of material properties using terahertz pusles

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Ultrafast control and monitoring of material properties using terahertz pulses

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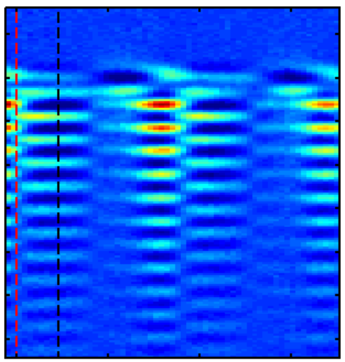

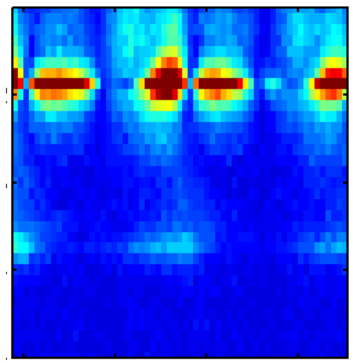

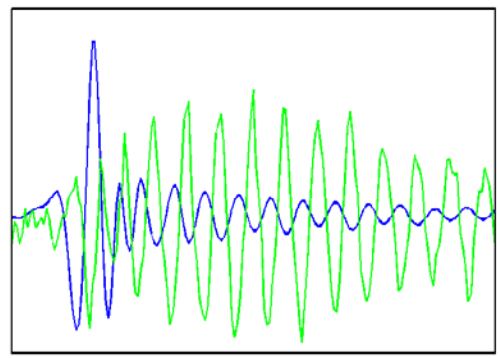

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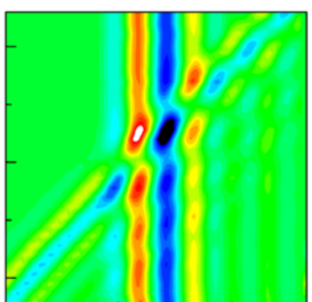

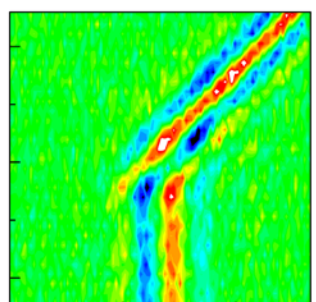

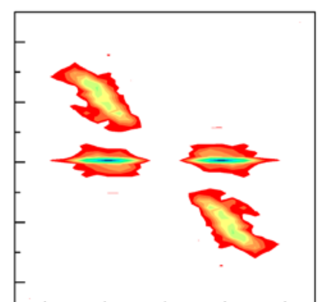

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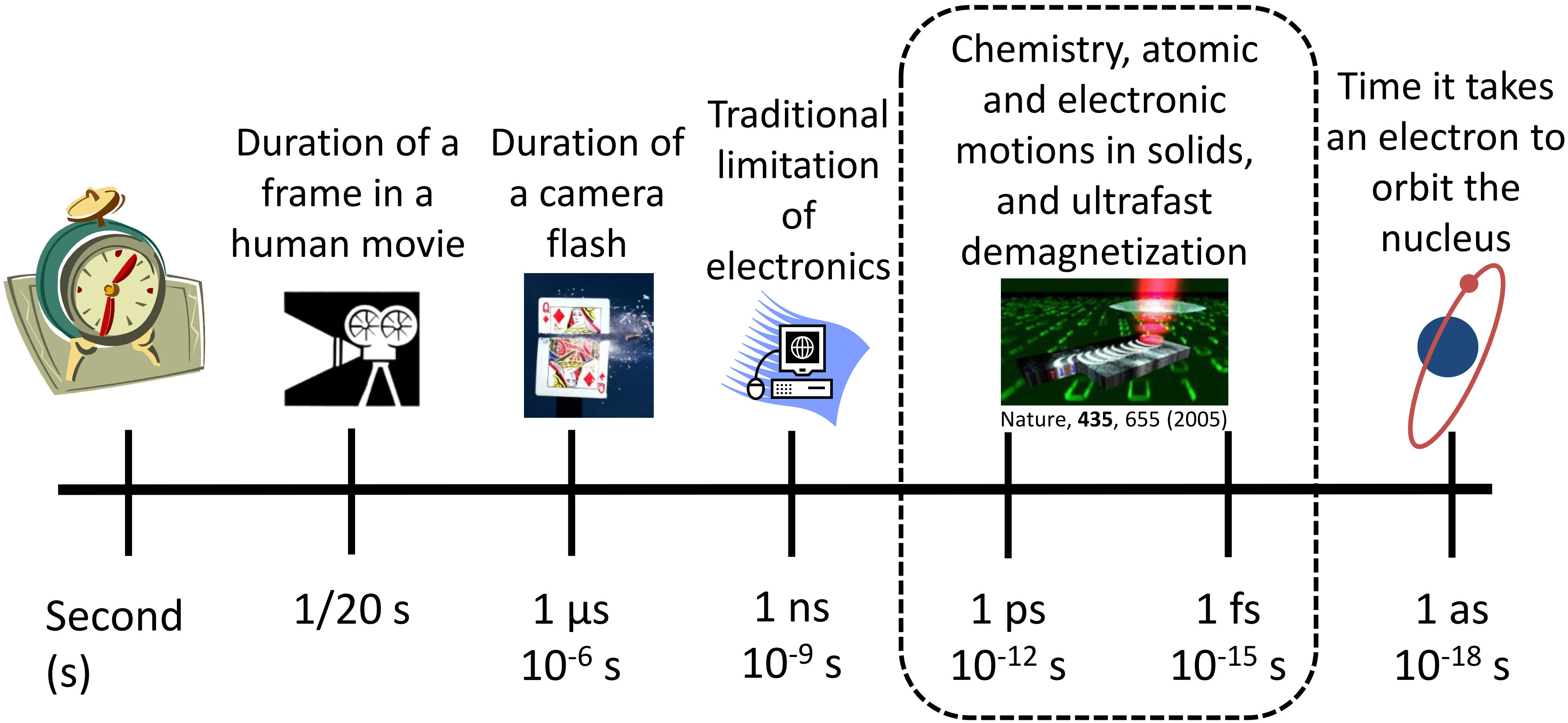
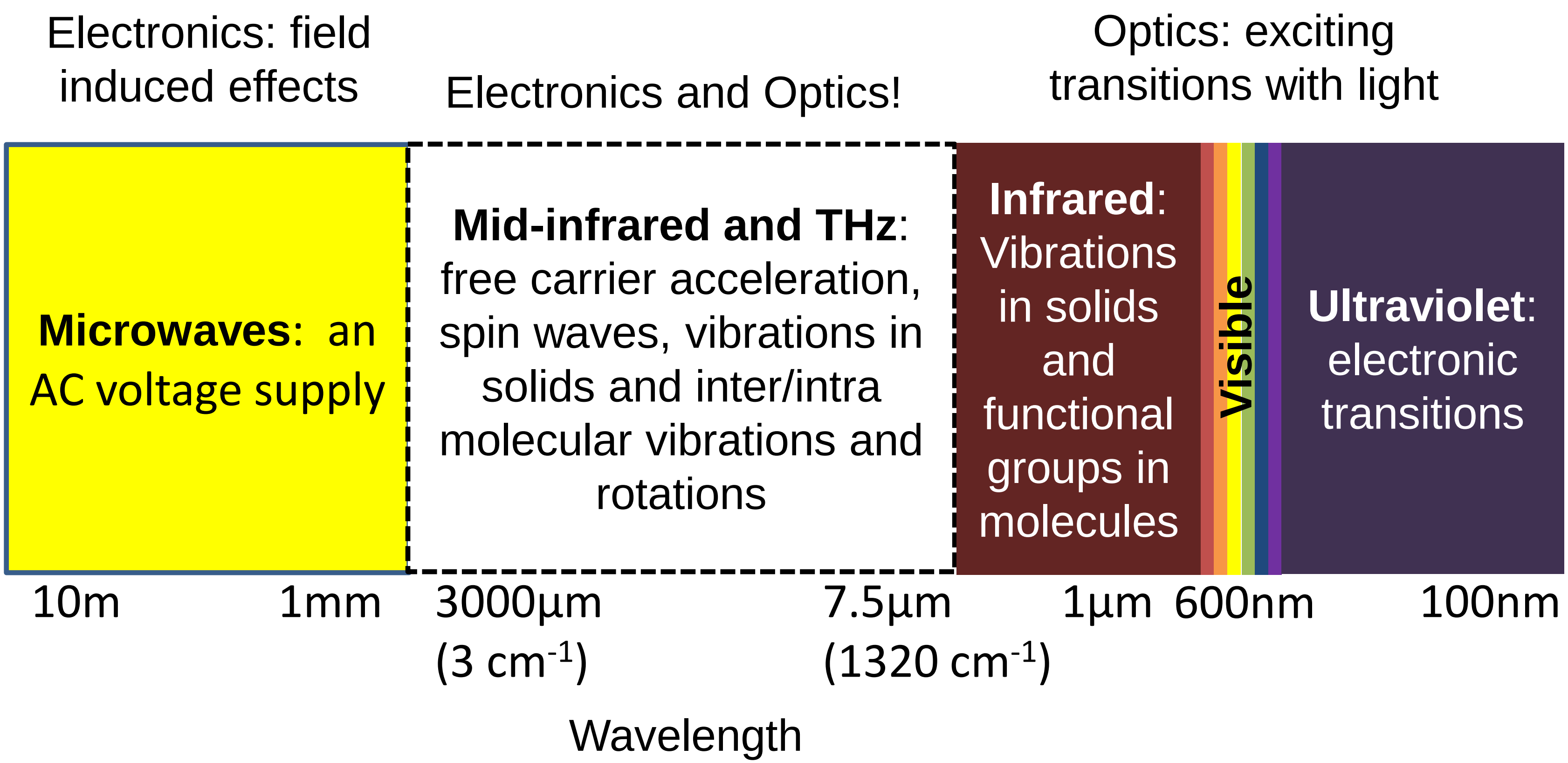
How fast is a femtosecond (fs)?



$$\frac{\text{1 second}}{\text{The age of the universe}} = \frac{\text{1 fs}}{\text{1 second}}$$

Different frequencies probe different properties of molecules or solids

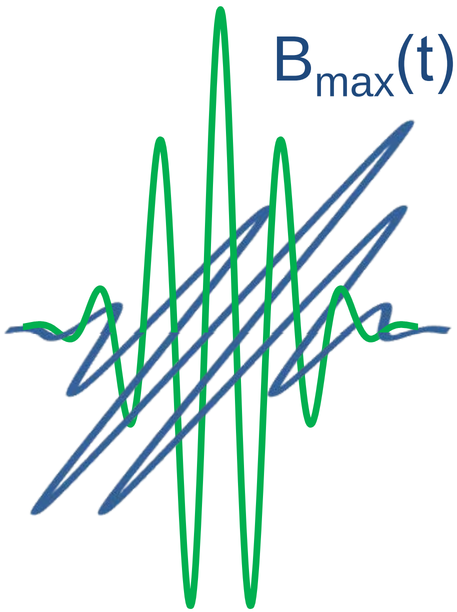


Field induced effects or optical transitions can be excited and probed with THz frequencies (0.1 – 40 THz).

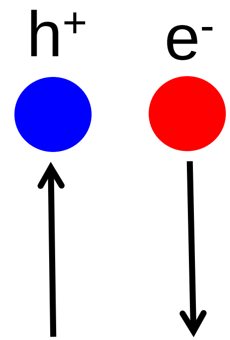
What can a THz pulse do to a material?

$E_{max}(t) \approx 200$ kV/cm

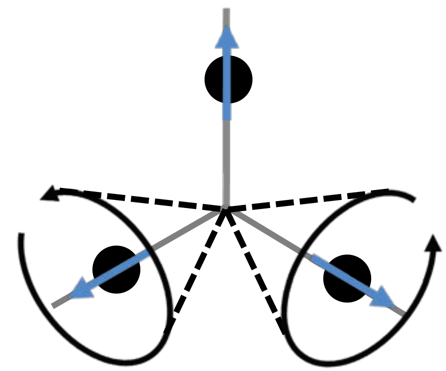
$B_{max}(t) \approx 66$ mT



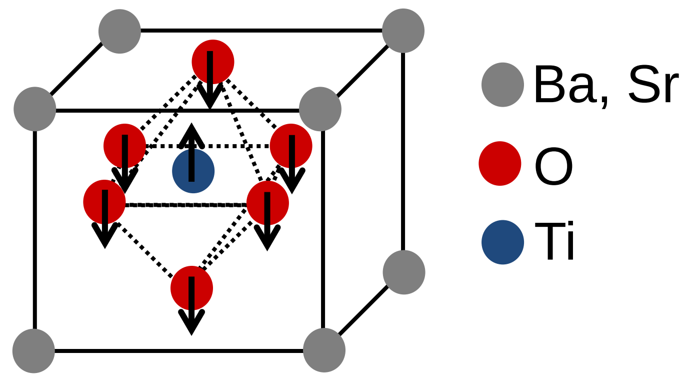
Accelerate charges



Change magnetic order



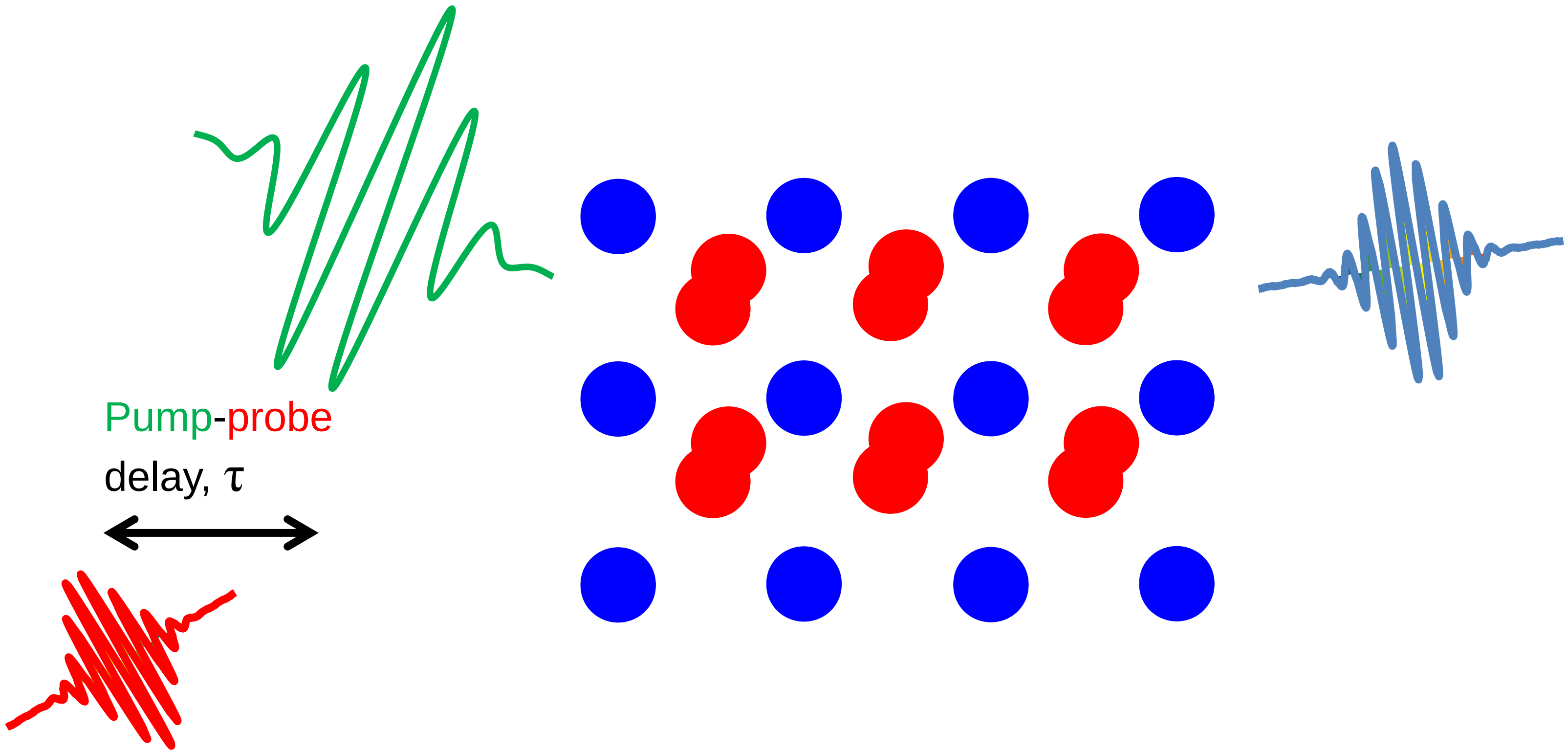
Excite vibrations (ferroelectric soft mode)



1 THz ↔ λ = 300 μm ↔ $1/\lambda$ = 33 cm^{-1} ↔ 4 meV ↔ T = 45 K

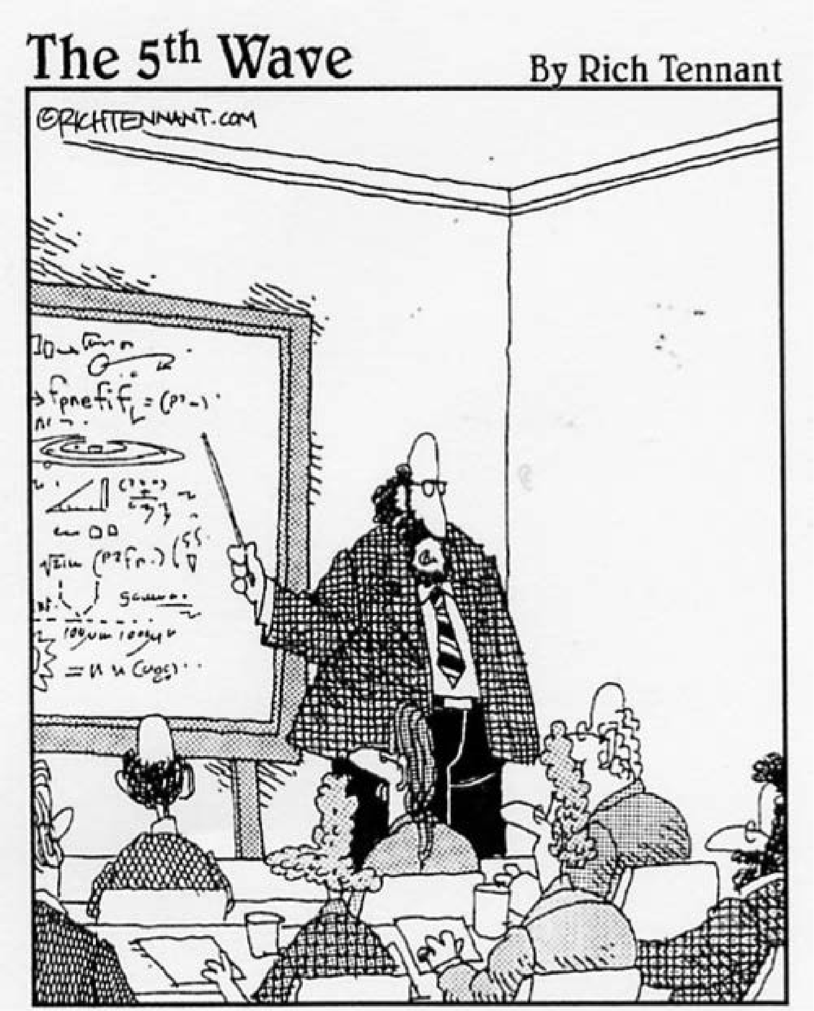
Review paper: T. Kampfrath et. al., Nature Photonics, **7**, 680 (2013)

Ultrafast spectroscopy



A pump pulse changes the material, and the probe pulse monitors these ultrafast changes.

Real world applications?



"After the discovery of 'antimatter' and 'dark matter', we have just confirmed the existence of 'doesn't matter', which does not have any influence on the Universe whatsoever."

http://quantumaniac.com/post/21079994364/dark-matter-antimatter-and-doesnt-matter

I will talk about…

1. Generating and measuring ultrashort THz pulses
2. Tracking ultrafast spin dynamics in antiferromagnets through spin wave resonances
3. Coherent two-dimensional THz spectroscopy
4. Probing vibrational dynamics at a surface

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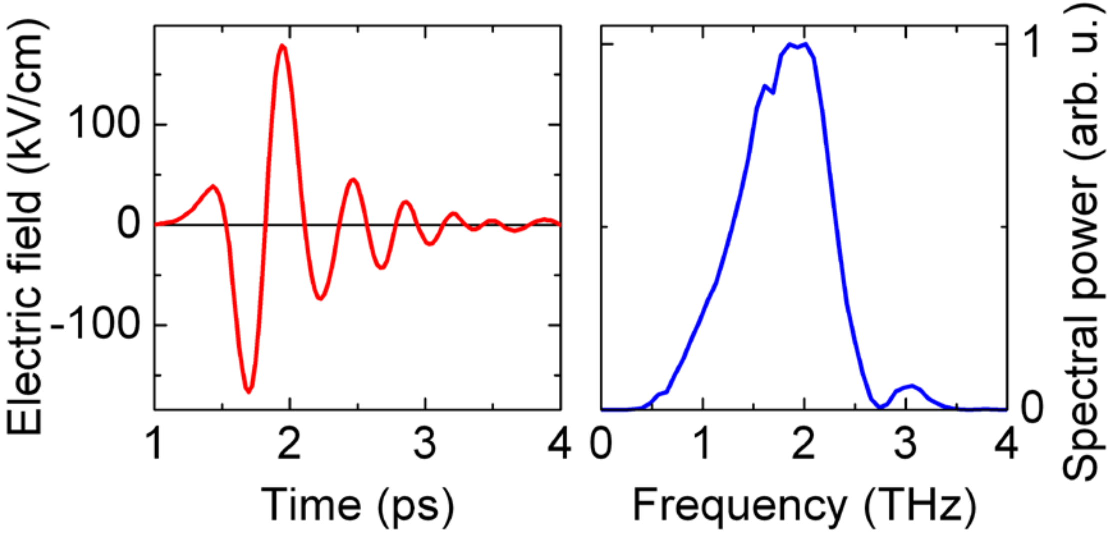
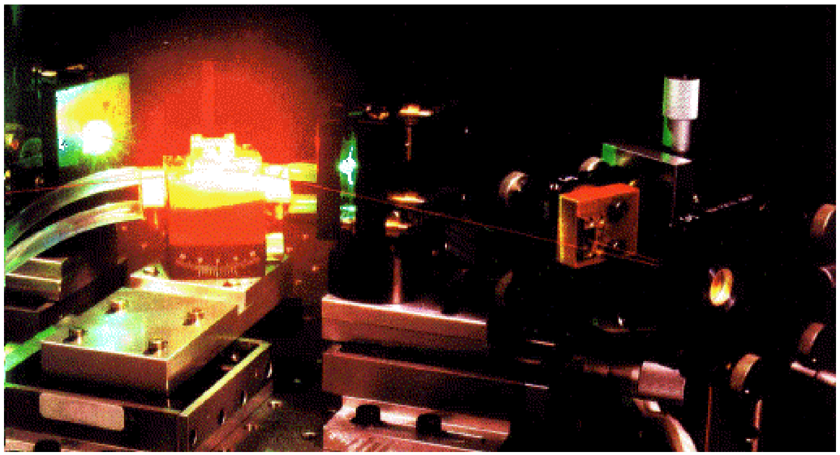


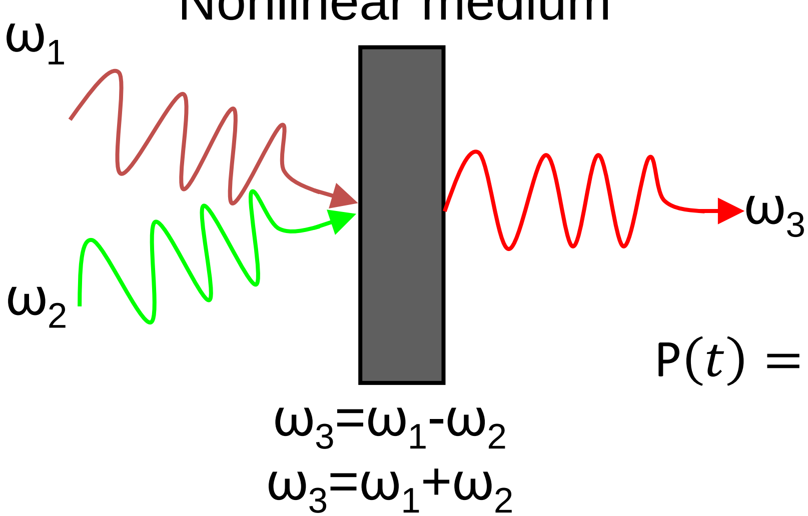
With Ti:Sa lasers and nonlinear optics, we can generate any frequency.

Ti:Sa femtosecond laser



$Ti:Al_2O_3$ (Ti:Sa) lasers/amplifiers are the most common source of intense pulses down to 4.5 fs.

The center wavelength is 700-1000 nm.

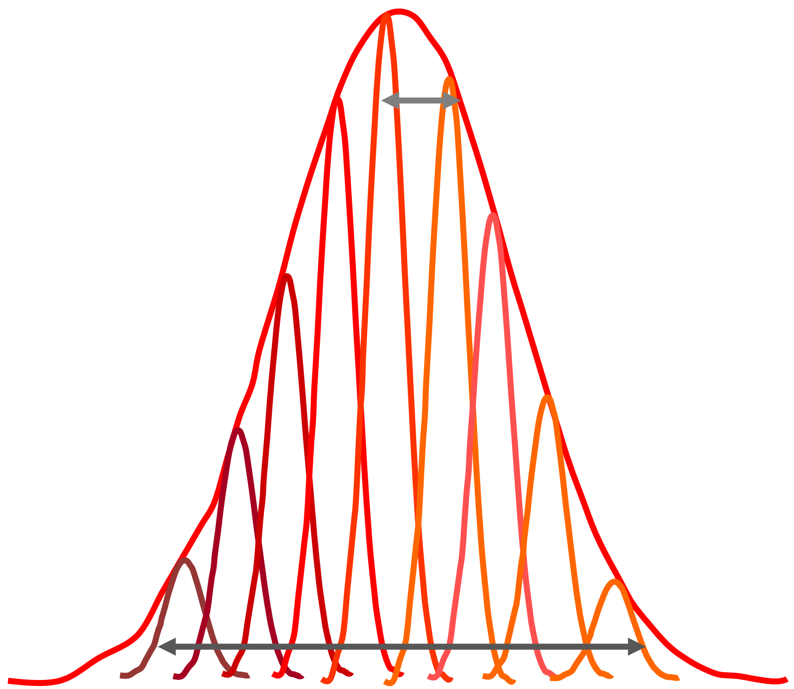


With nonlinear optics nearly any other frequency can be generated.

$$P(t) = \varepsilon_0\left(\chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + \cdots\right)$$

Generating THz pulses by optical rectification

$(\omega_1 - \omega_2)/\pi$ = (c/800-c/805) ≈ 2 THz



$(\omega_1 - \omega_2)/\pi$ = (c/780-c/820) ≈ 16 THz

Choose a nonlinear crystal:

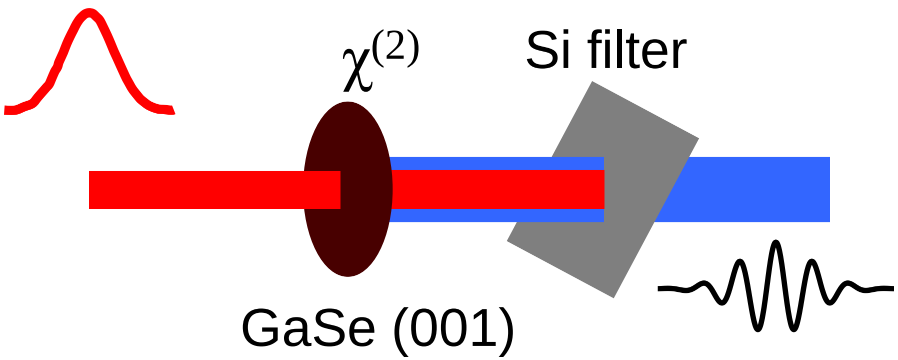
- ✓ $\chi^{(2)}$ must have the correct symmetry.
- ✓ It must transmit 800 nm and THz light.
- ✓ **Phase matching:**

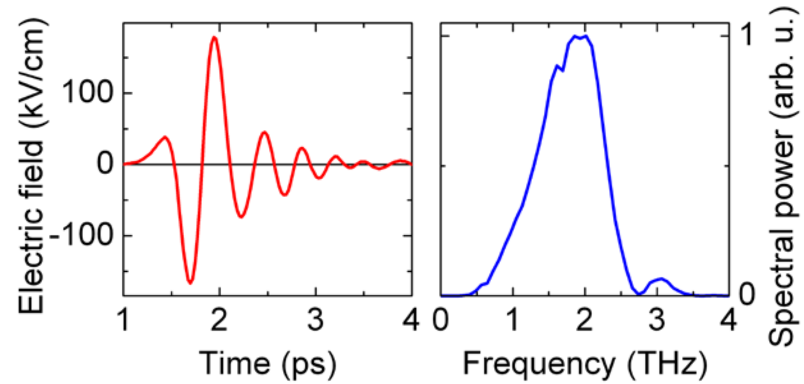
  v_{phase}(2THz)=v_g(800nm)

$$P^{(2)}_{Opt\ Rect} = \chi^{(2)}(\omega_{THz} = \omega_1 - \omega_2; \omega_1, -\omega_2)\tilde{E}(\omega_1)\tilde{E}^*(\omega_2)$$

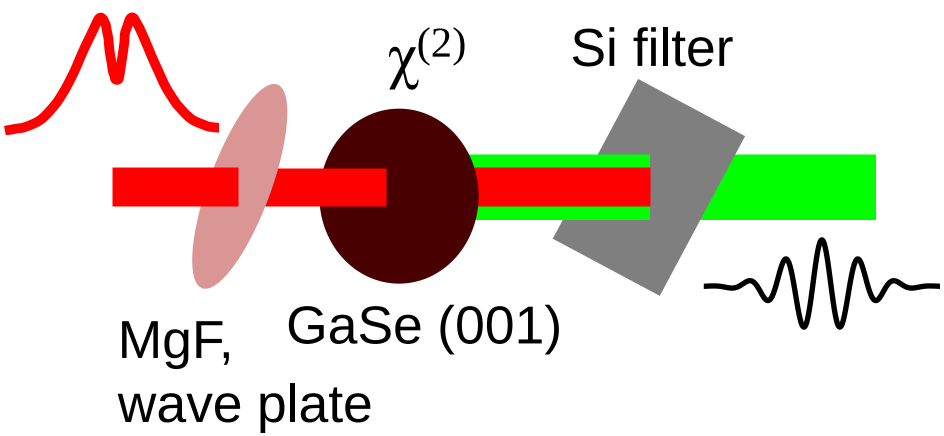
THz to mid-IR generation is GaSe

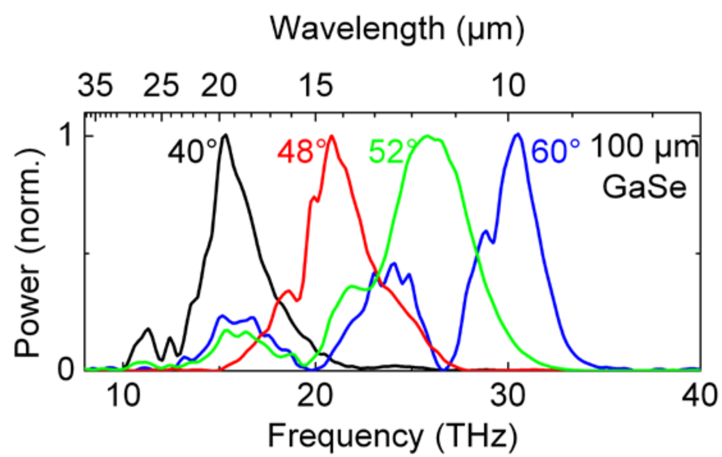
Normal incidence for
Generation of 0.1-3 THz





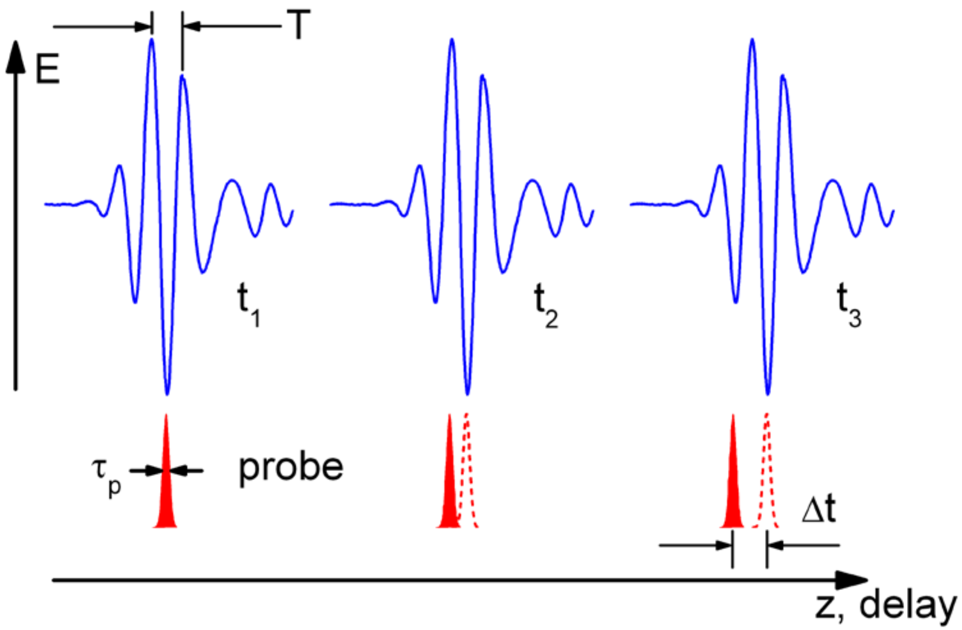
Angle tuning the crystal phase matches higher frequencies from 10 - 40 THz.

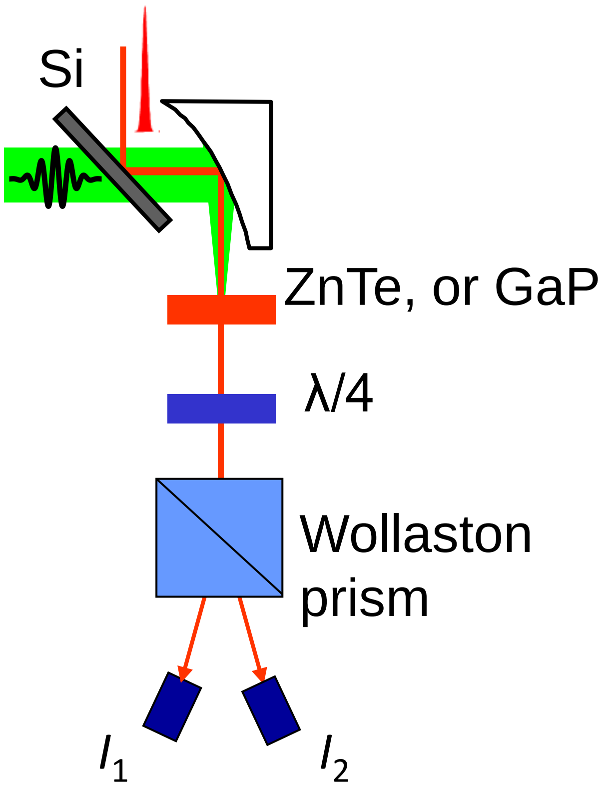




Reimann, et. al., Opt. Lett., **28**, 471 (2003)

Measuring THz E-fields with the linear electro-optic effect.





$$\Gamma = \frac{2\pi d}{\lambda} n^3(\lambda) r_{41} E_{THz} \qquad \frac{I_1 - I_2}{I_1 + I_2} = \sin(\Gamma) \approx \Gamma$$

The Induced birefringence, and polarization rotation are proportional to the THz electric field.

Wu, Zhang, APL **71**, 1285 (1997)

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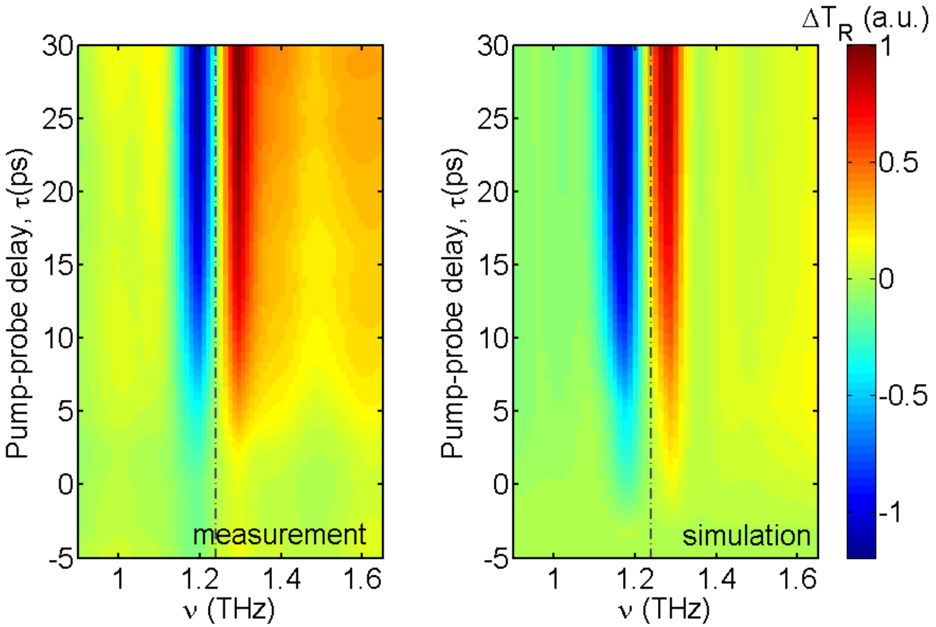
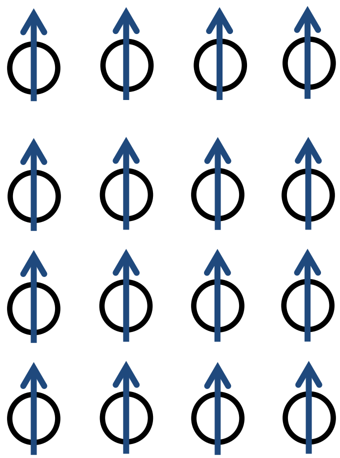


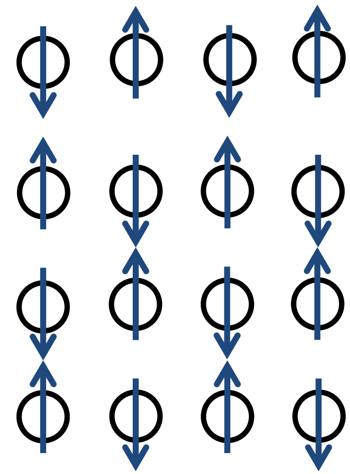
Optical control and monitoring of spins

Ferromagnet

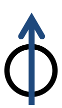


Net magnetic moment,
$M = m$ x number of atoms

Antiferromagnet



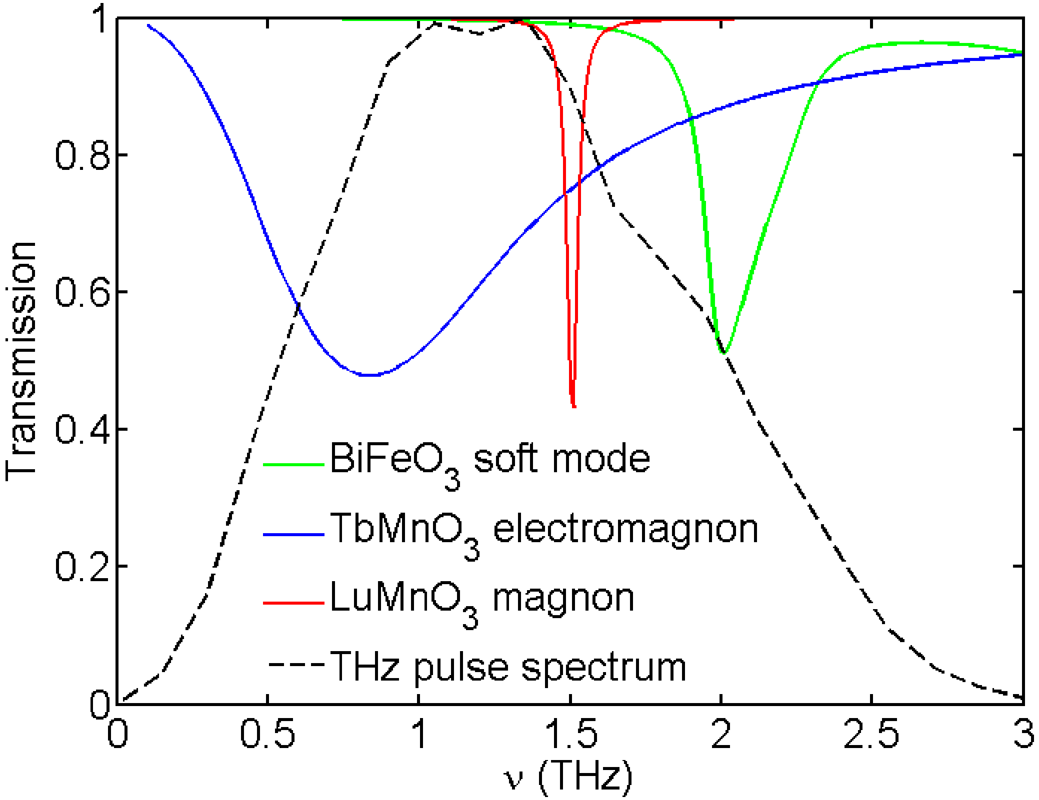
Net magnetic moment $M = 0$

 Magnetic atoms in a lattice, with magnetic moment m

$M = 0$ makes spin dynamics in antiferromagnets (AFMs) potentially faster, but how do you detect this?

Kirilyuk et. al., Rev. Mod. Phys. **82**, (2010).

Ultrafast probing of material properties through low energy modes

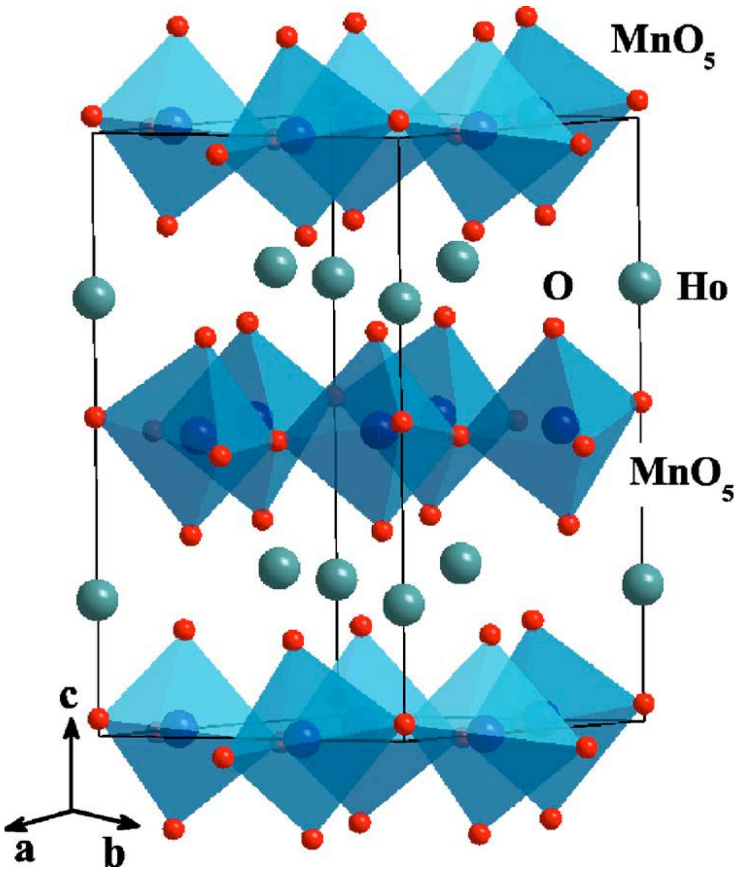


Antiferromagnetic order can be probed through spin waves.

Kamba et. al, PRB **75**, (2007), Standard et. al, PRB **85**, (2012), Sushkov et. al., PRL **98**, (2007)

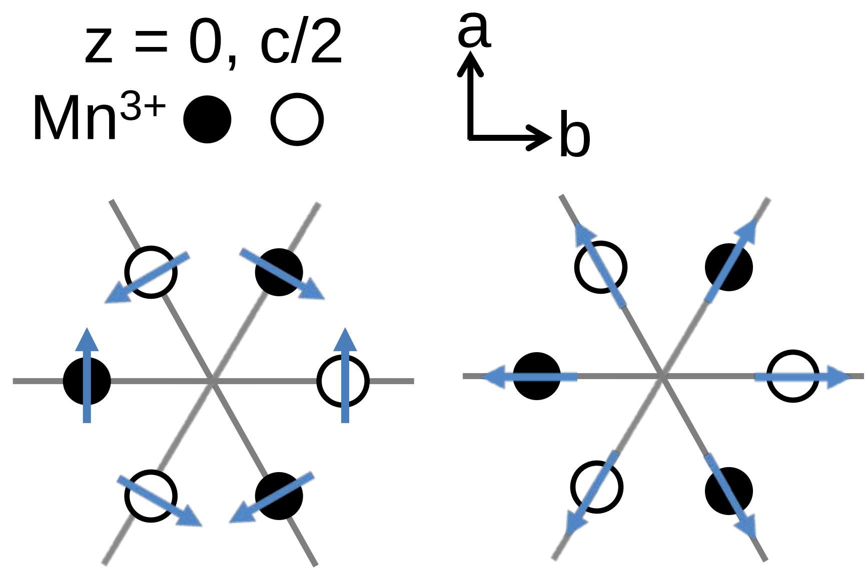
Probing antiferriomagnetic spin dynamics in $HoMnO_3$?

Hexagonal Lattice
$T \leq T_c$ (FE) = 875 K



Rai, et. al, PRB, **75** (2007).

Frustrated antiferromagnet below 78 K



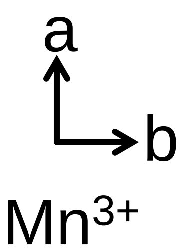
P$\underline{6}_3$c$\underline{m}$
$T < T_{SR} = 42$ K

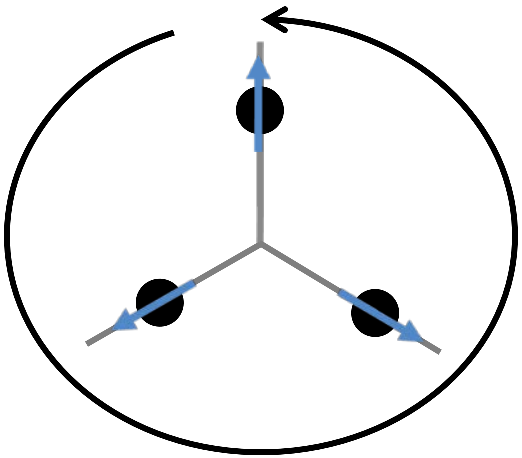
P$\underline{6}_3\underline{c}$m
$T < T_{Néel}$

Fiebig, et. al, JAP, **91** (2002).

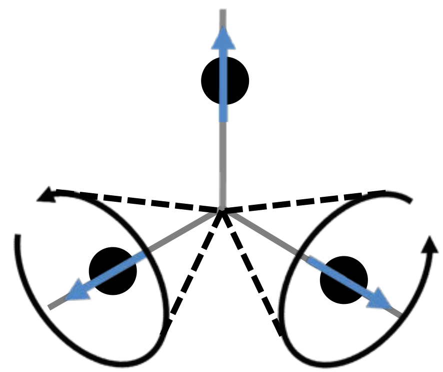
Magnons in $HoMnO_3$

Mode 1,
depends on K_2





Modes 2 and 3
(degenerate).
Depends on K_1 and λ



Uniaxial anisotropy

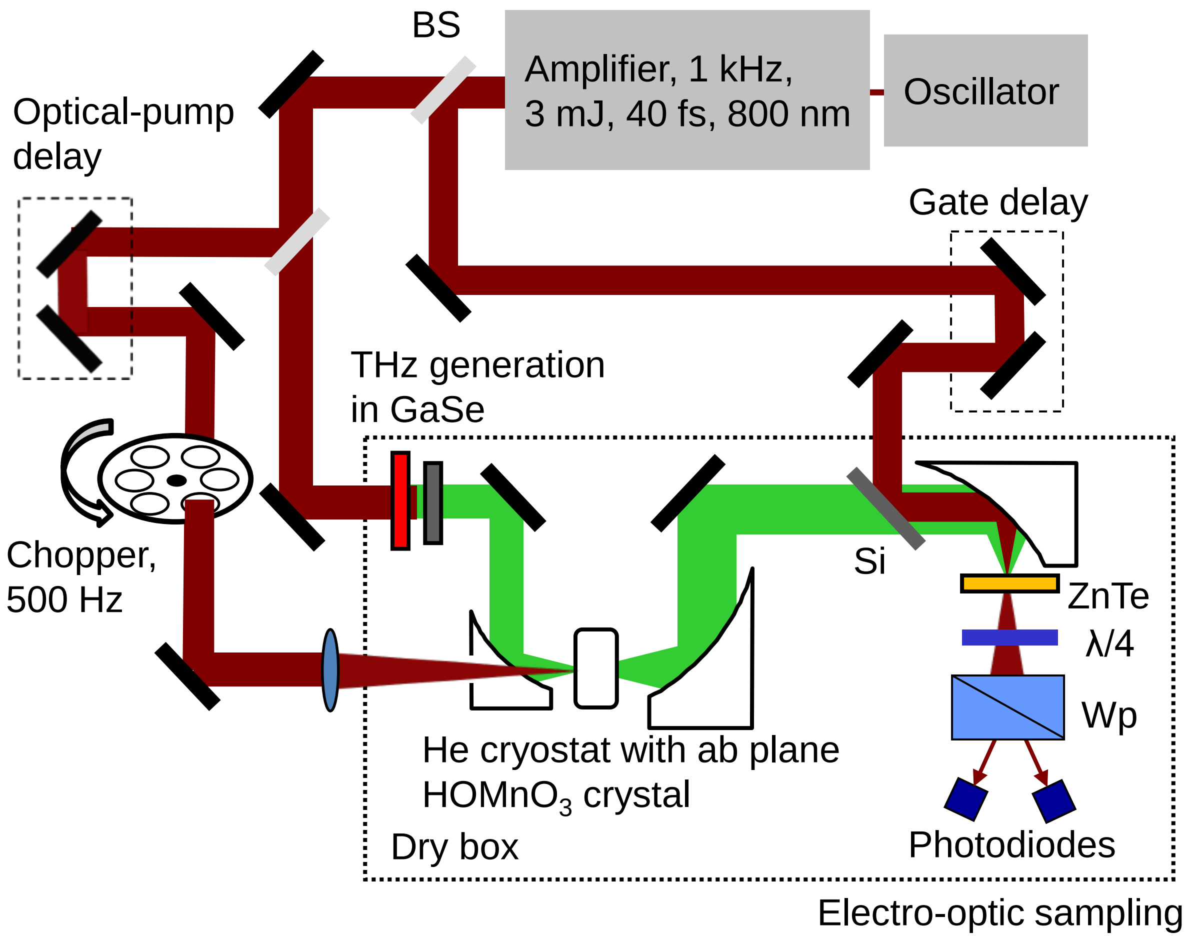
In-plane anisotropy

Exchange

$$E = -K_1 \sum_i cos^2 \theta_i - K_2 \sum_i cos^2 3\varphi_i + \lambda \sum_{i<j} M_i \cdot M_j$$

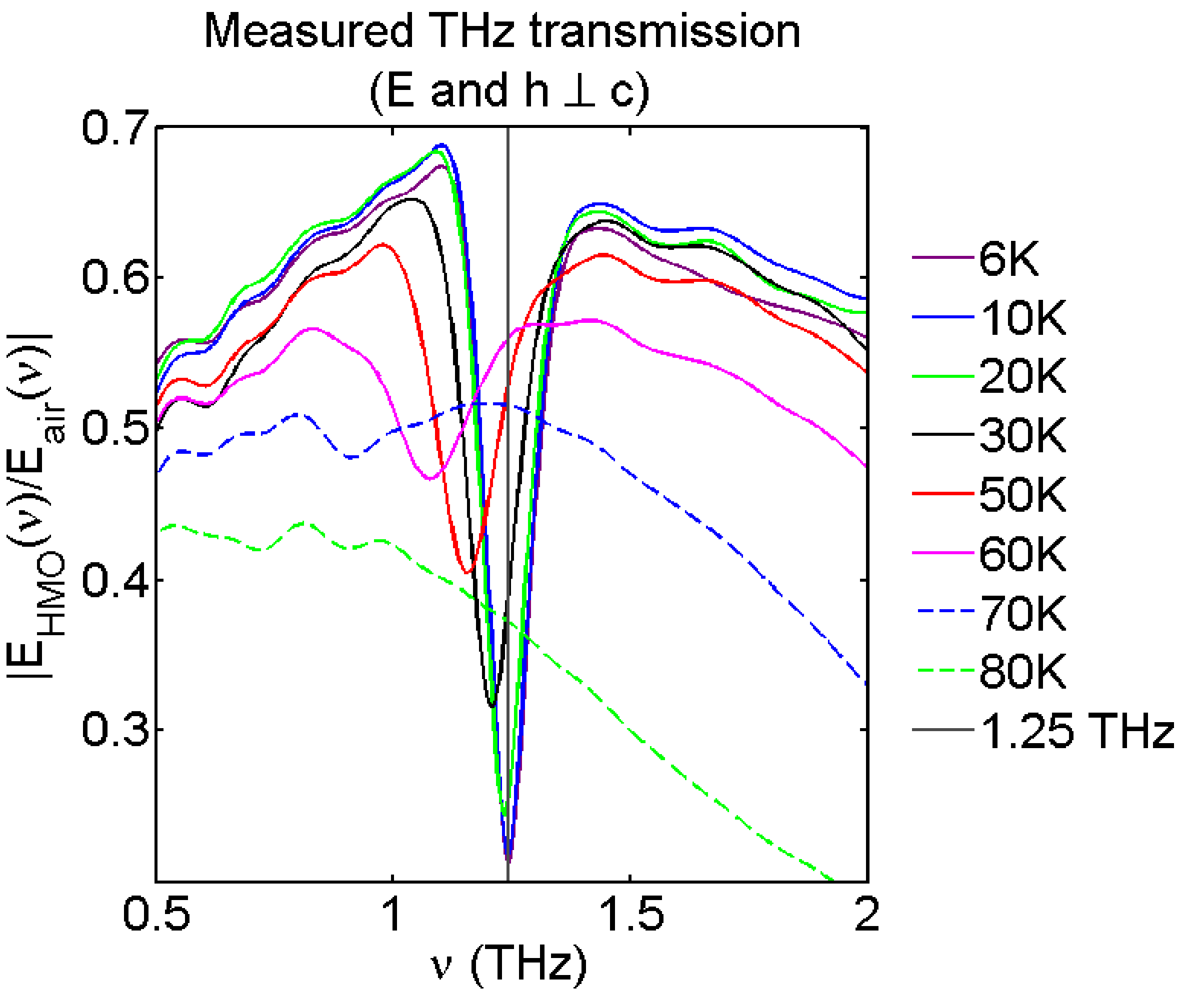
Penny et. al, JAP **40**, (1969), and Talbayev et. al., PRL **101**, (2008).

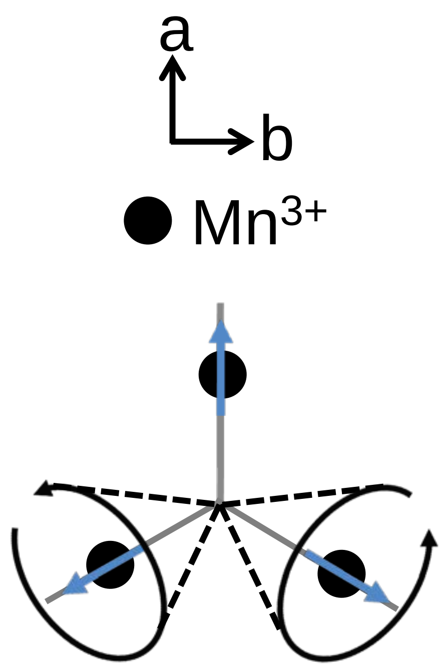
Optical-pump, THz-probe experimental setup



This time the optical pump excites electrons, and we use the THz pulse to probe how this couples to magnetic order.

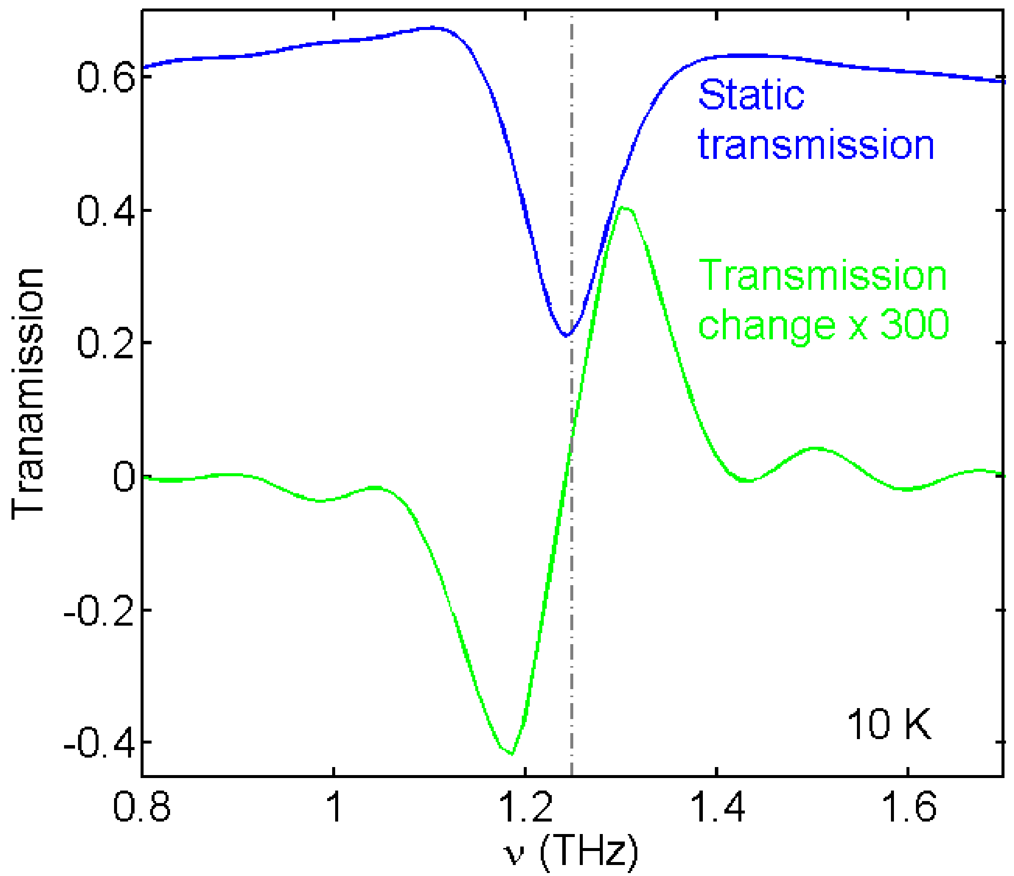
Steady state THz transmission in $HoMnO_3$

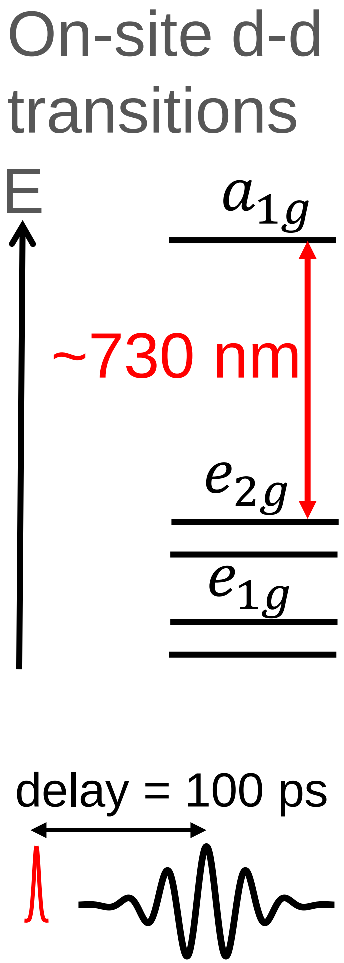




As the temperature increases, the restoring forces are weaker, so the mode broadens and lowers in frequency.

Photoexcitation changes the magnon line shape

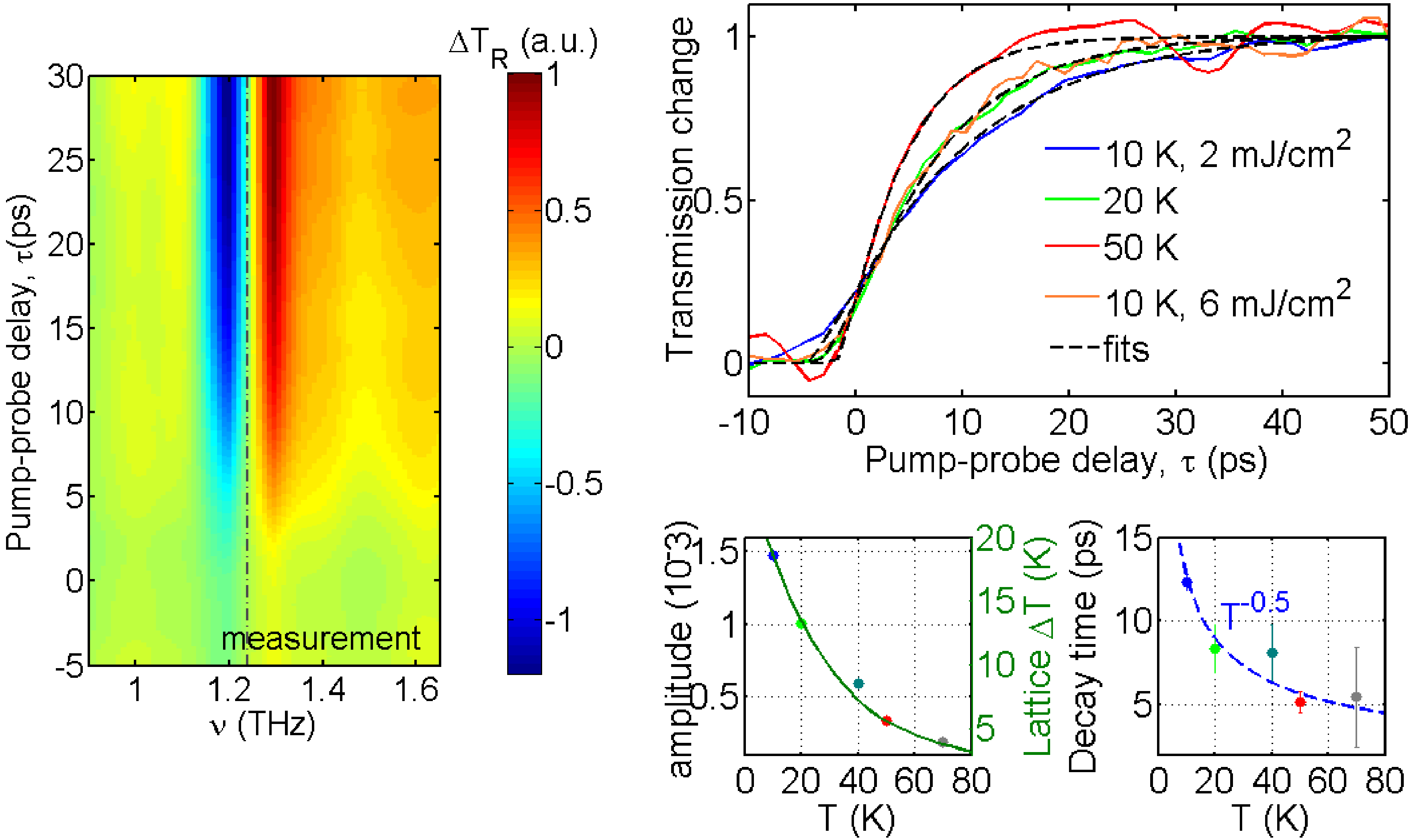




The THz pulse is sensitive to antiferromagnteic order!

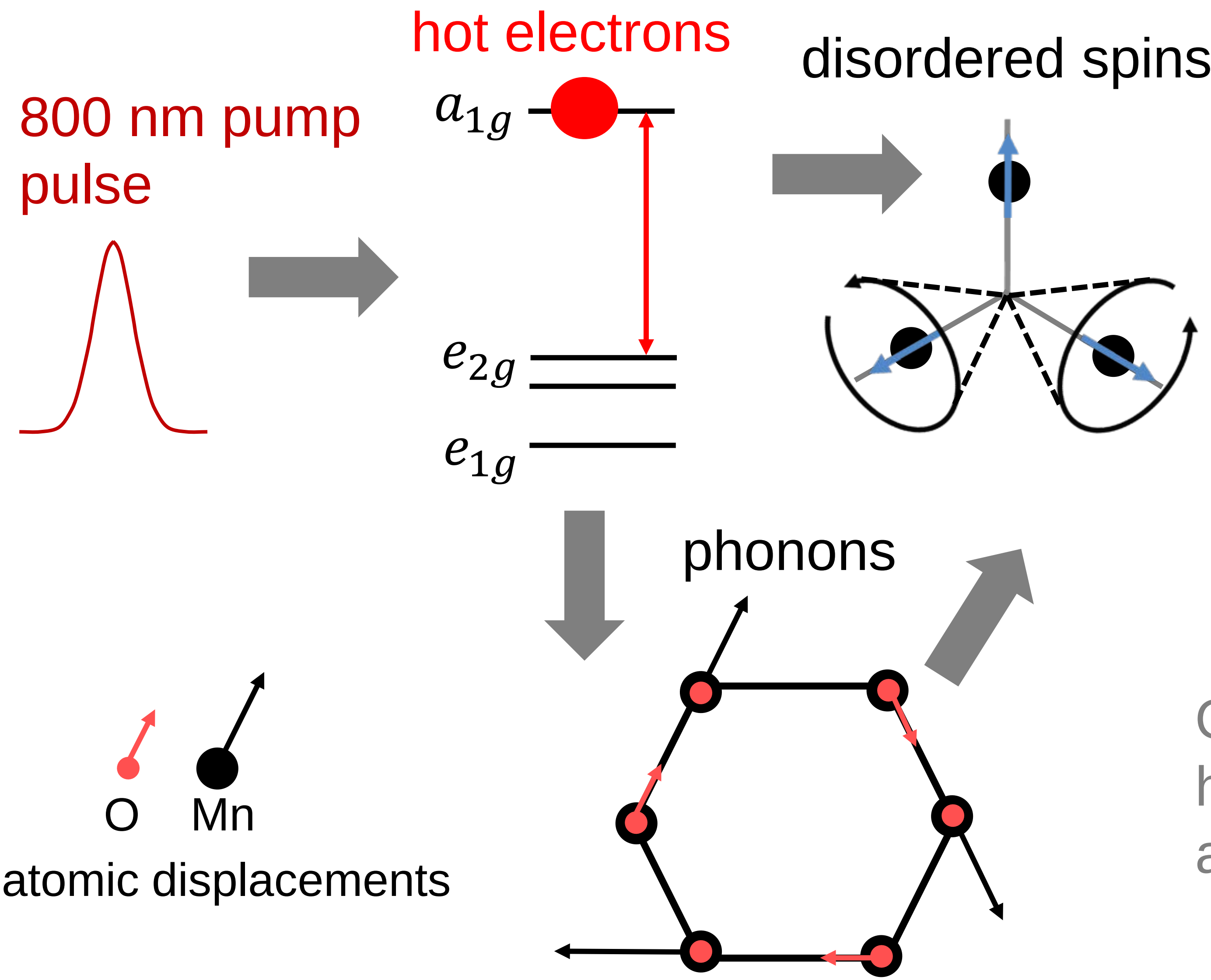
Optical absorption in HMO: Souchkov et. al. PRL **91** 2003 and Rai, et. al, PRB, **75** (2007).

Ultrafast magnon line shape changes



Lattice vibrations can directly heat spins in AFMs spins, but not in FMs.

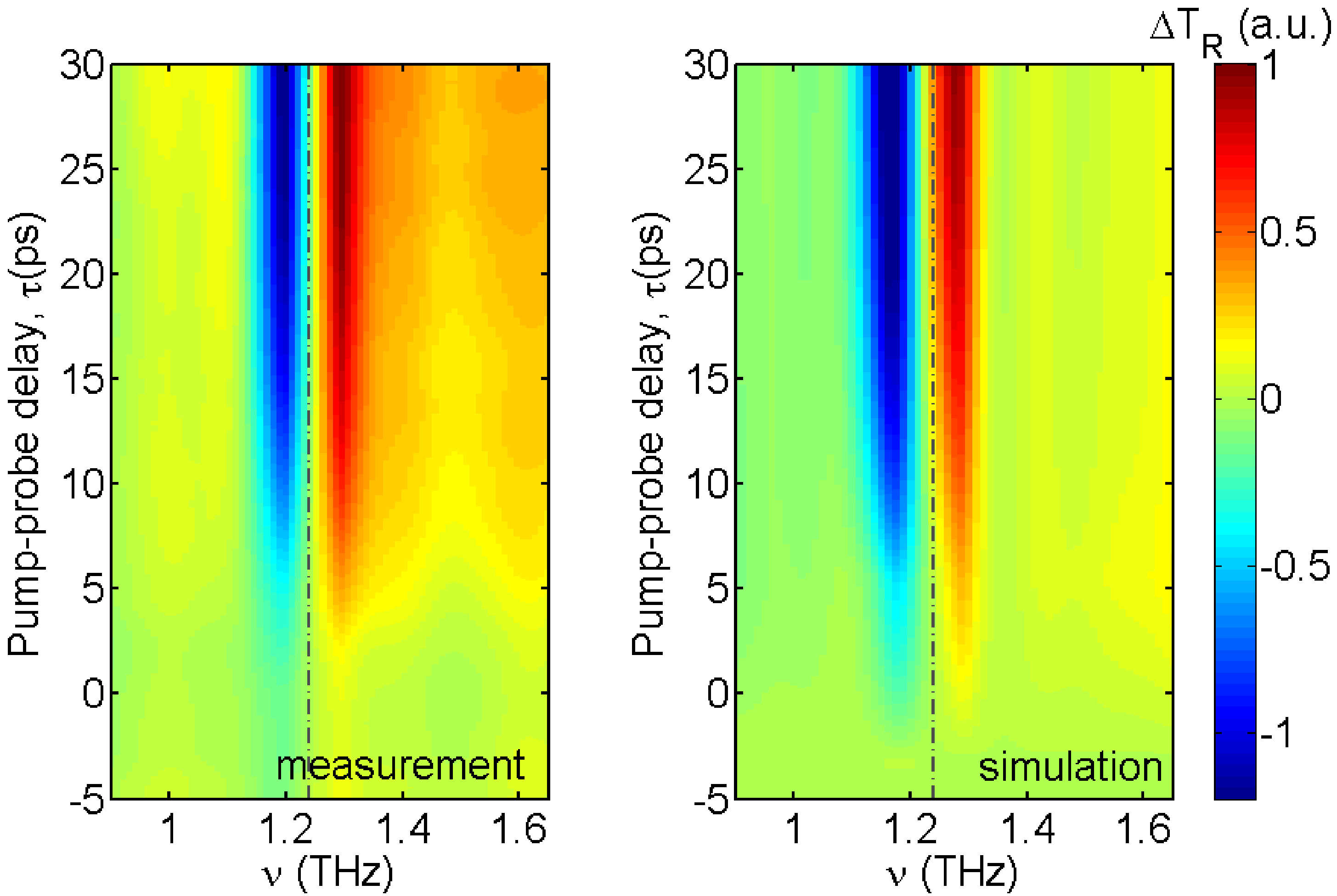
How does the pump pulse's energy get from electrons to spins?



Direct transfer of energy from electrons to spins?

Or is the spin heating phonon assisted?

Phonon assisted heating of spins

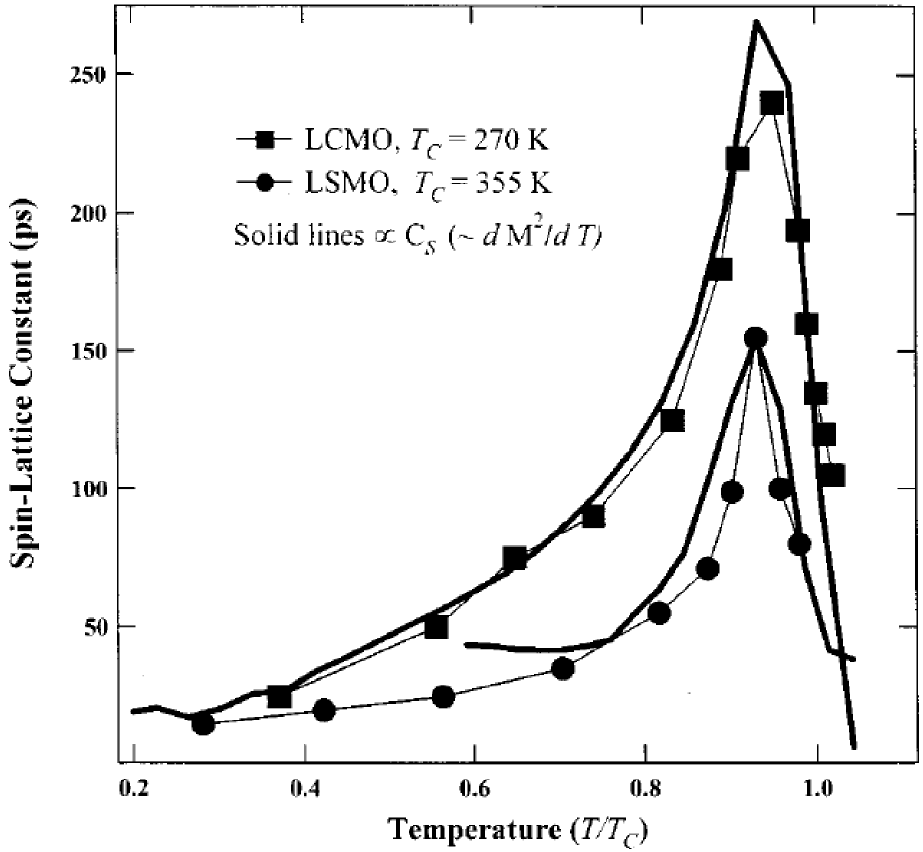


The 12 picosecond change on the magnon line shape is the time that it takes the lattice and the spins to come to thermal equilibrium.

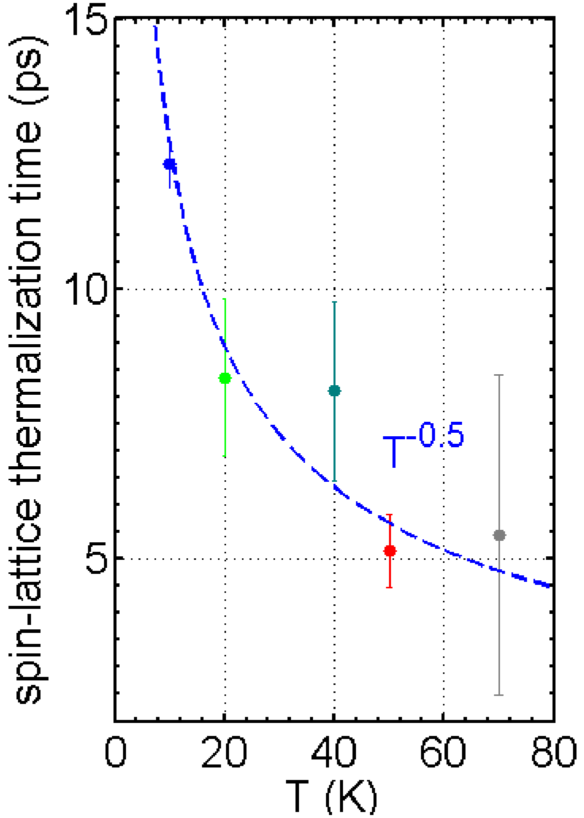
P. Bowlan, et. al., arXiv:1602.03872

Spin lattice heating in antiferromagnets versus ferromagnets

Ferromagnet $M > 0$



Antiferromagnet $M = 0$



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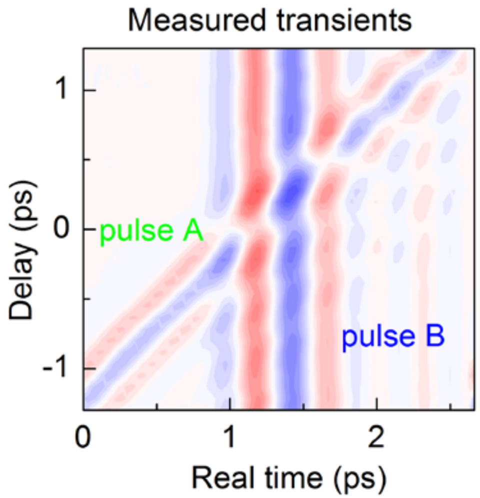
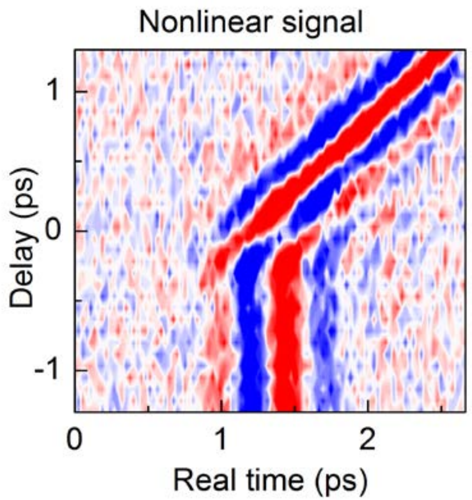
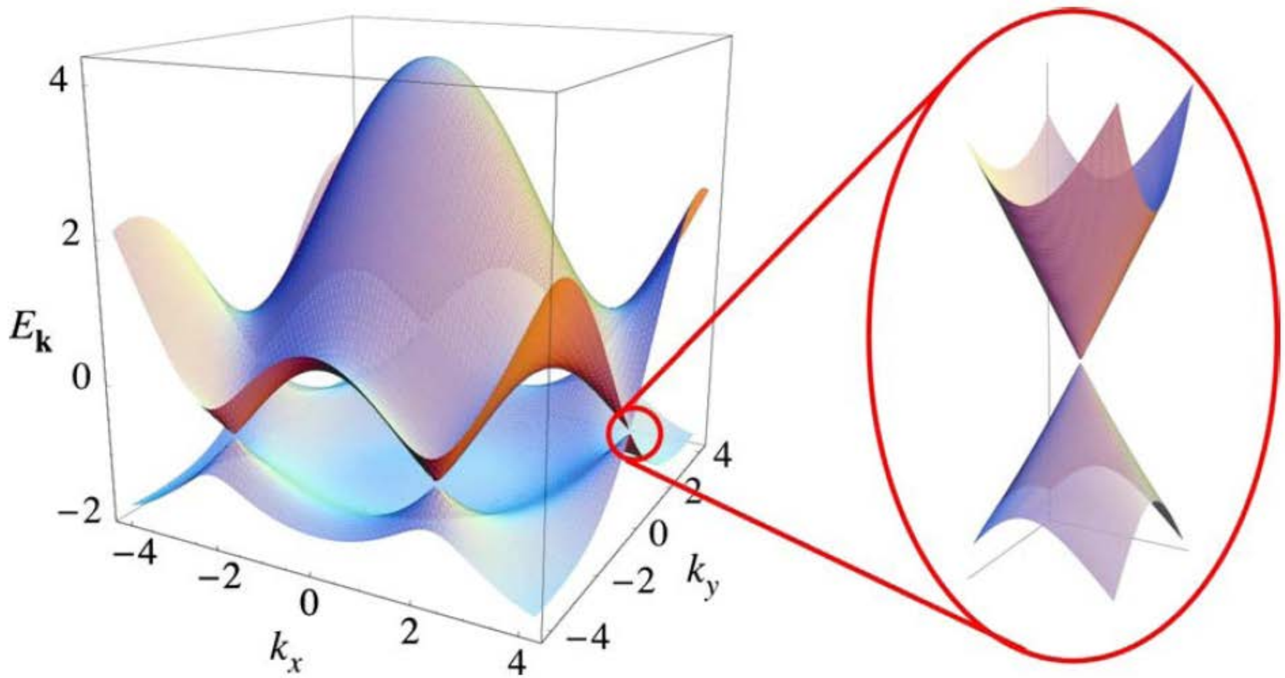




Graphene: Massless charge carriers

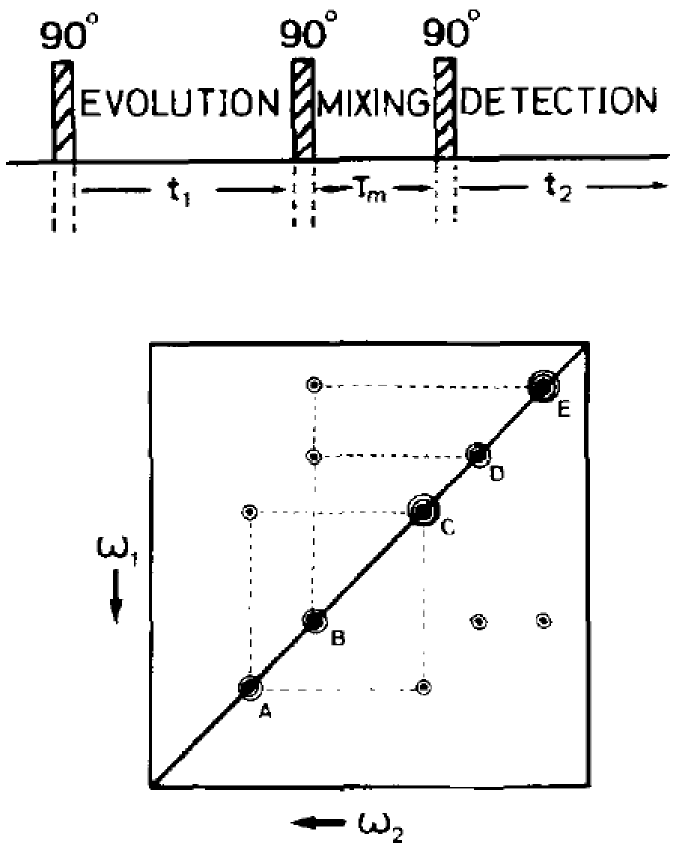


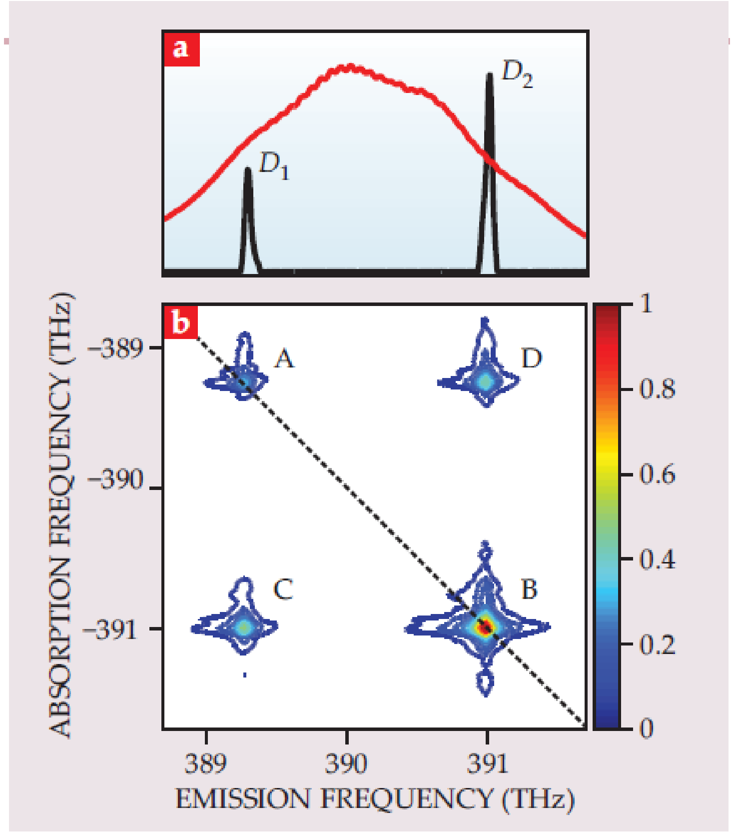
Quantum Absorption= $\pi\alpha$

What are the femtosecond carrier transport properties at low energies?

Castro Neto et. al., Rev. Mod. Phys., 81, (2009)

Coherent two-dimensional spectroscopy





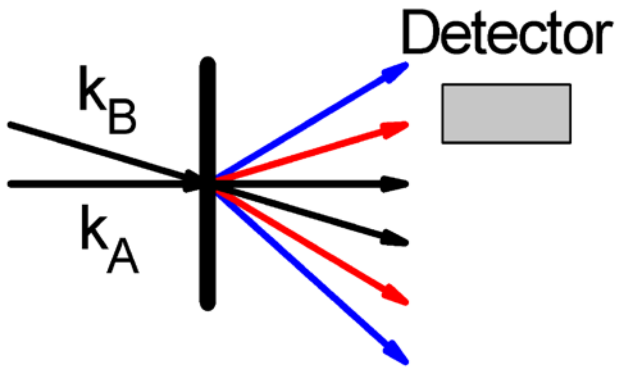
With radio frequencies for NMR: Biological macromolecule from R. R. Ernst et. al., Biochem and Biophys. Research Comm. **95** (1975)

In the visible: Potassium vapor in the review by, S. Cundiff and S. Mukamel, Physics Today, **66** (2013)

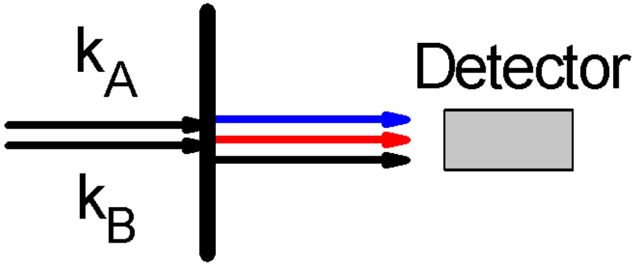
This gives information about coherence, and coupling between modes.

Extending Coherent two-dimensional spectroscopy to the THz

Noncollinear, in the visible:



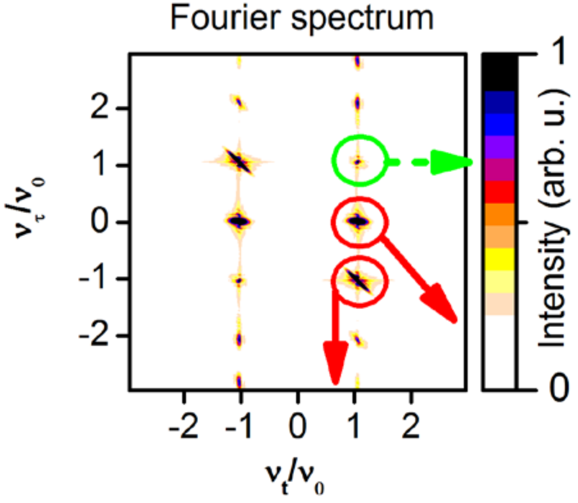
Collinear, in the THz:



Perturbative theory:

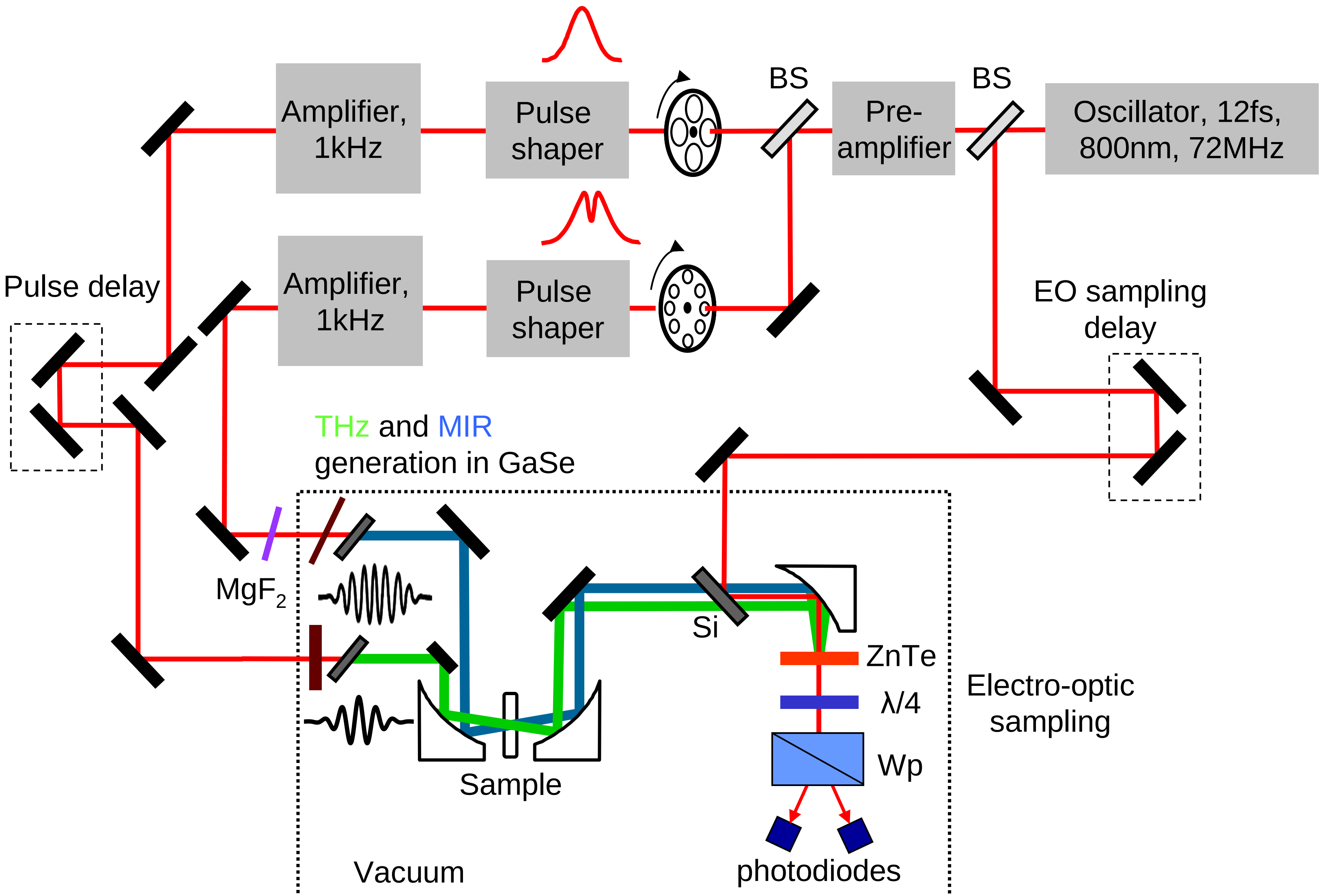
$$P = \varepsilon_0(\chi E + \chi^{(3)} E^3 + \chi^{(5)} E^5 ...)$$

W. Kuehn, et. al., J. Chem. Phys. 130, 164503 (2009).



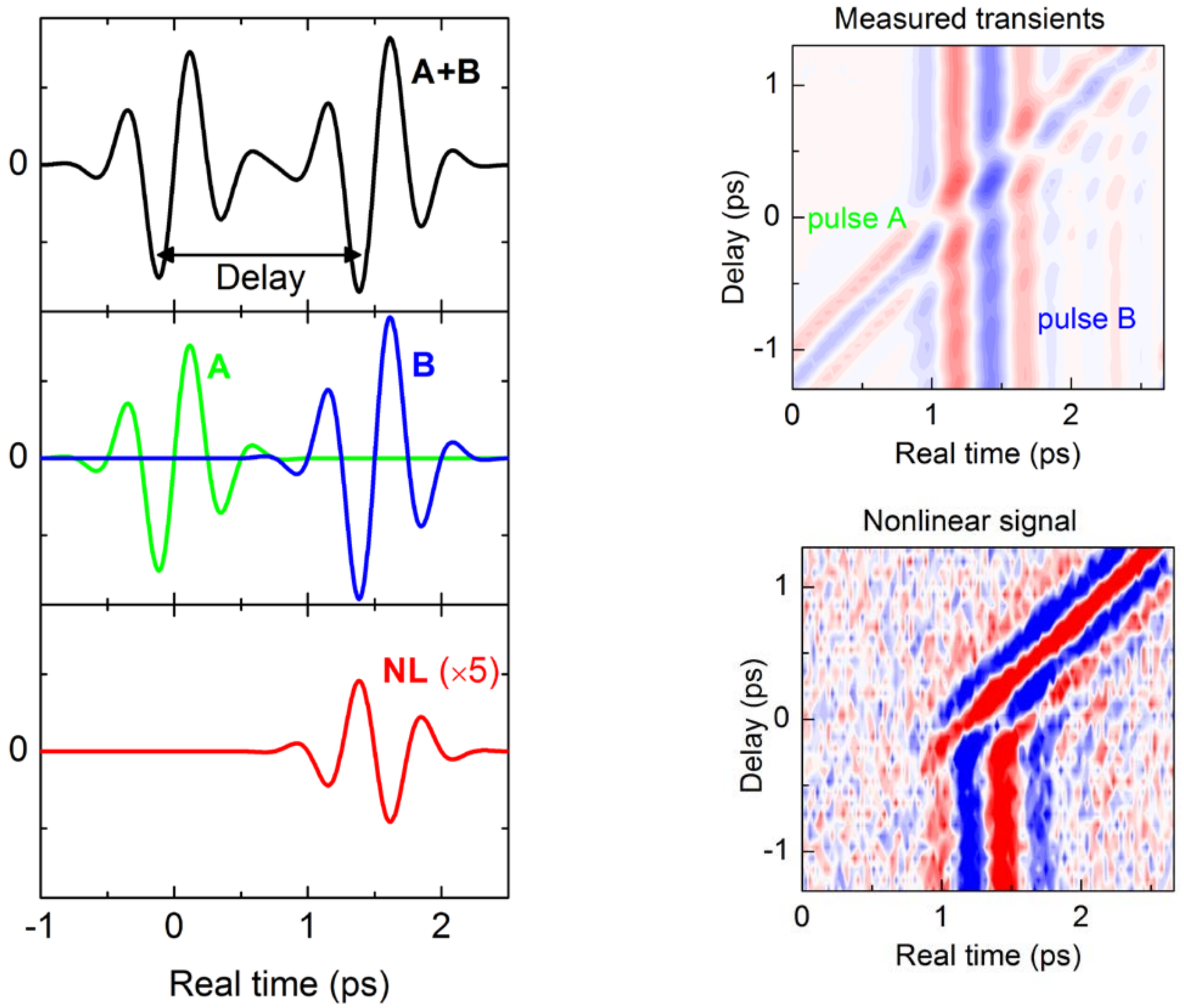
In the collinar case, we measure the entire nonlinear response and separate the signals in frequency.

Coherent two-dimensional THz spectroscopy



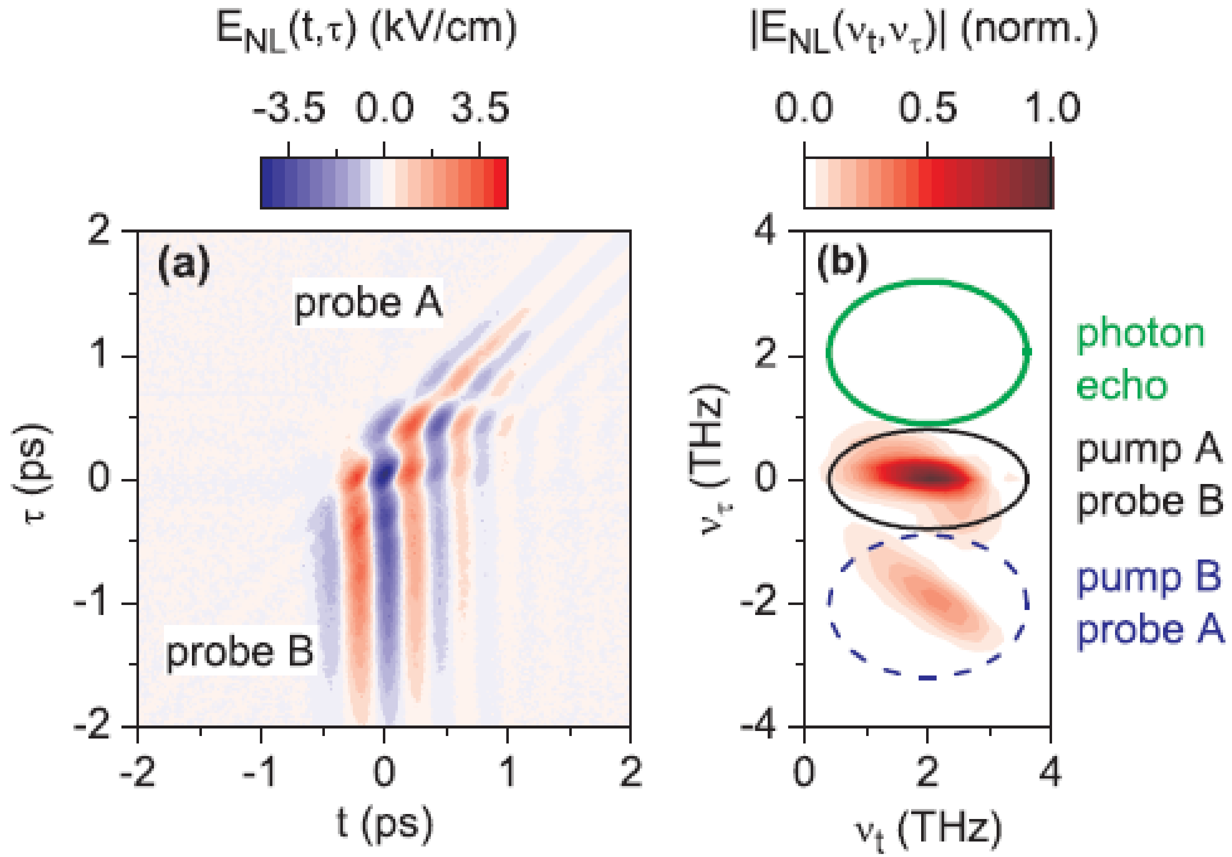
This apprach works in the 0.1 - 40 THz (3000-7.5 μm) spectral range.

Two-dimensional THz spectroscopy: experimental concept



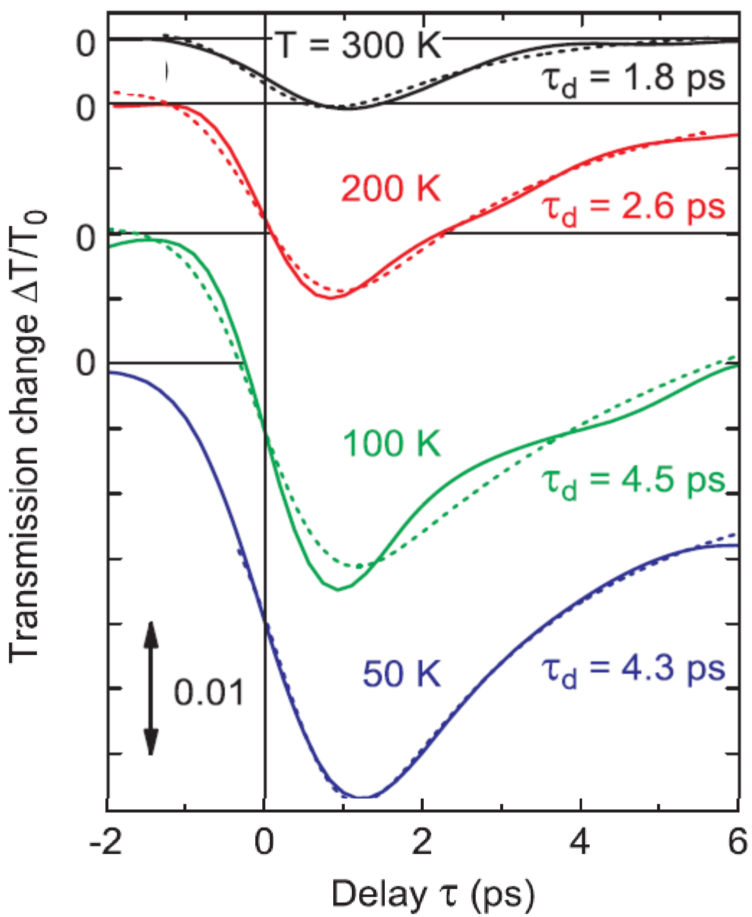
Our graphene sample is 40 layers thick, epitaxially grown on SiC?

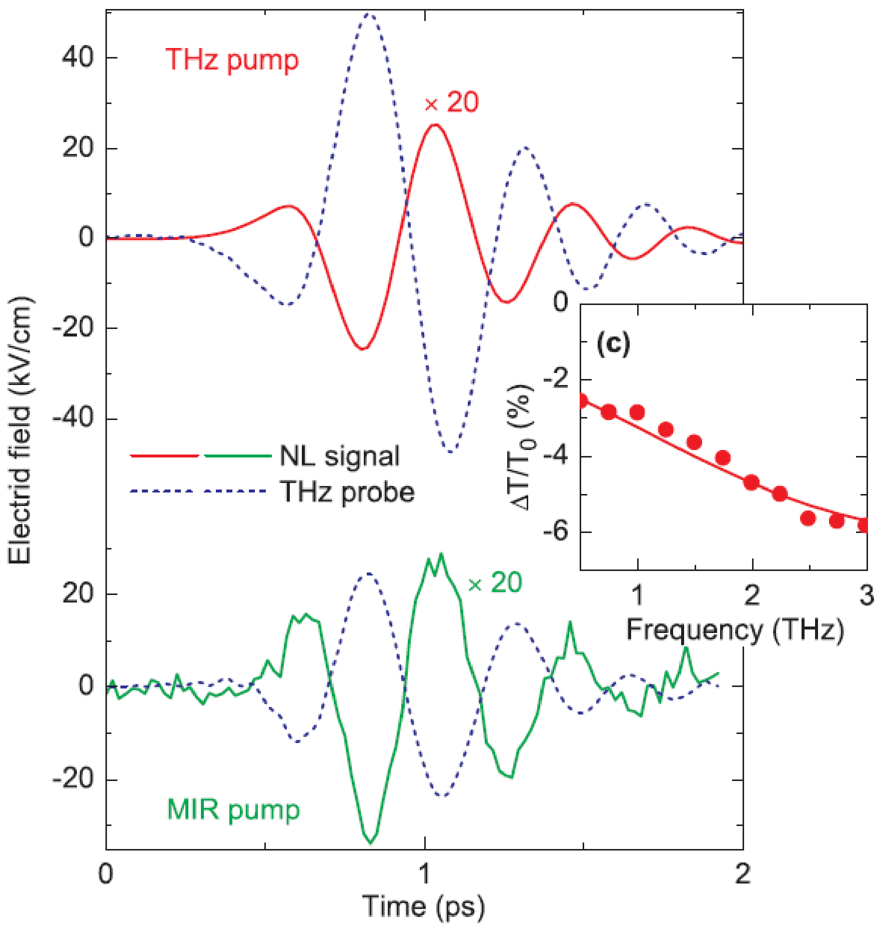
Two-dimensional THz spectroscopy: separating the nonlinear signals



Fourier transforming separates the nonlinear signals. For graphene photon echo is not present, indicating a very short coherence time.

Analyzing the pump-probe signal

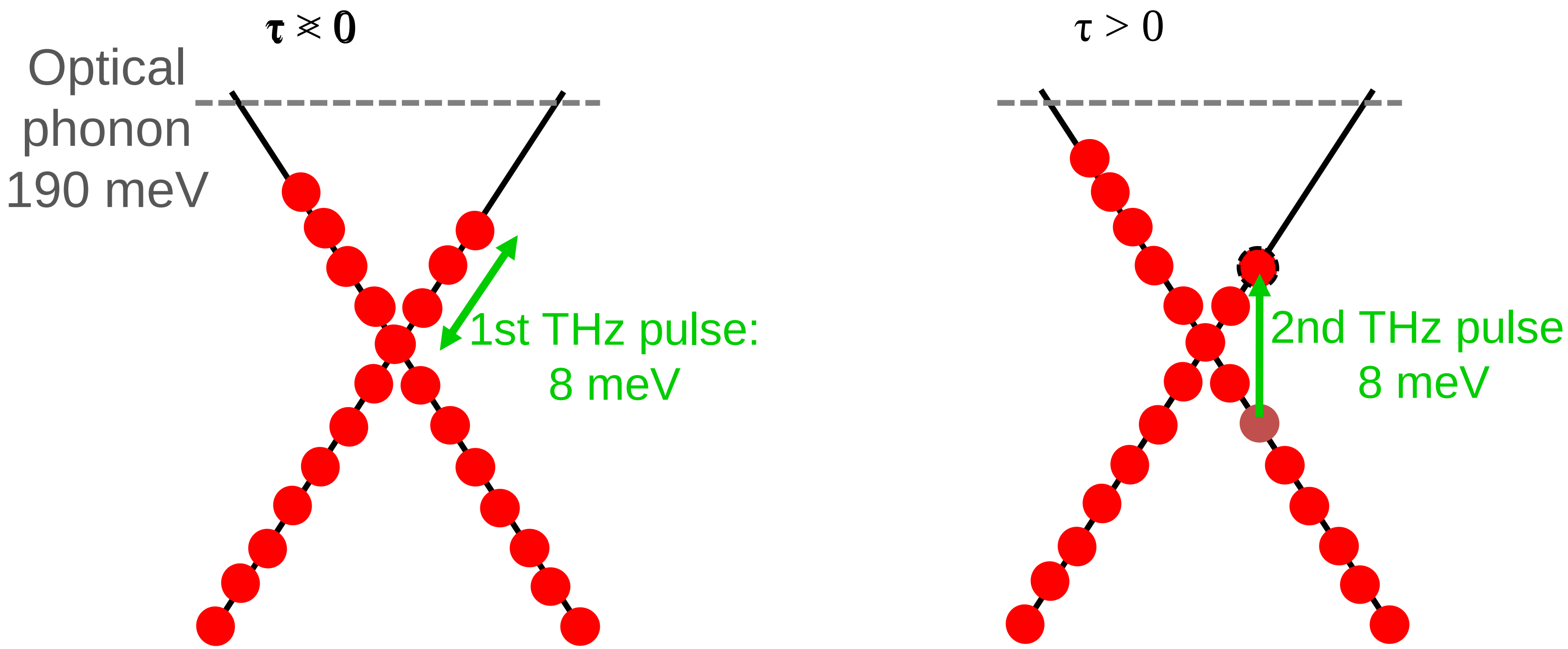




An induced absorption lasting 2-4 picosecond was observed.

What causes the induced absorption with a short life time?

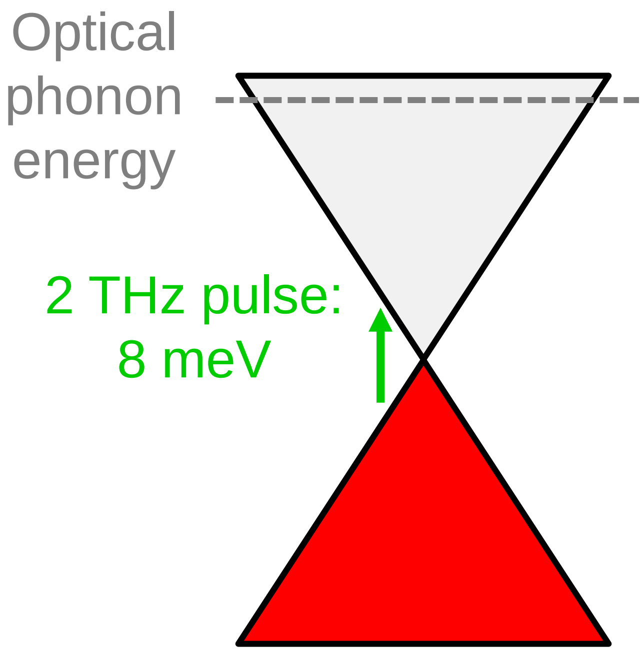
Usual semiconductor picture: Heating and cooling.



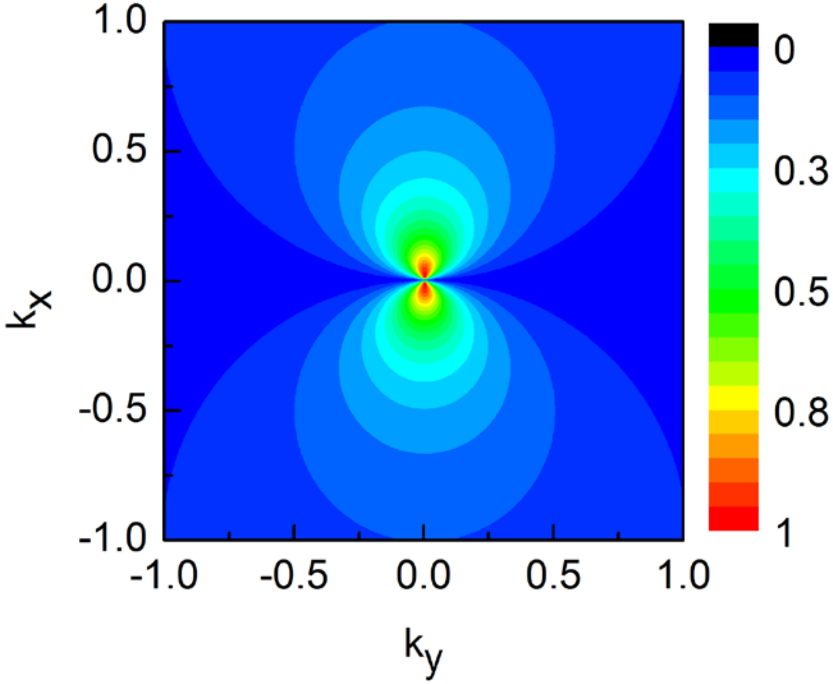
But cooling by acoustic phonons requires 100’s of picoseconds.
This picture works for higher energy excitations.

Winnerl et. al., Phys. Rev. Lett., 107, (2012) and Strait et. al. Nano lett., 11, (2011)

The light matter interaction is very strong



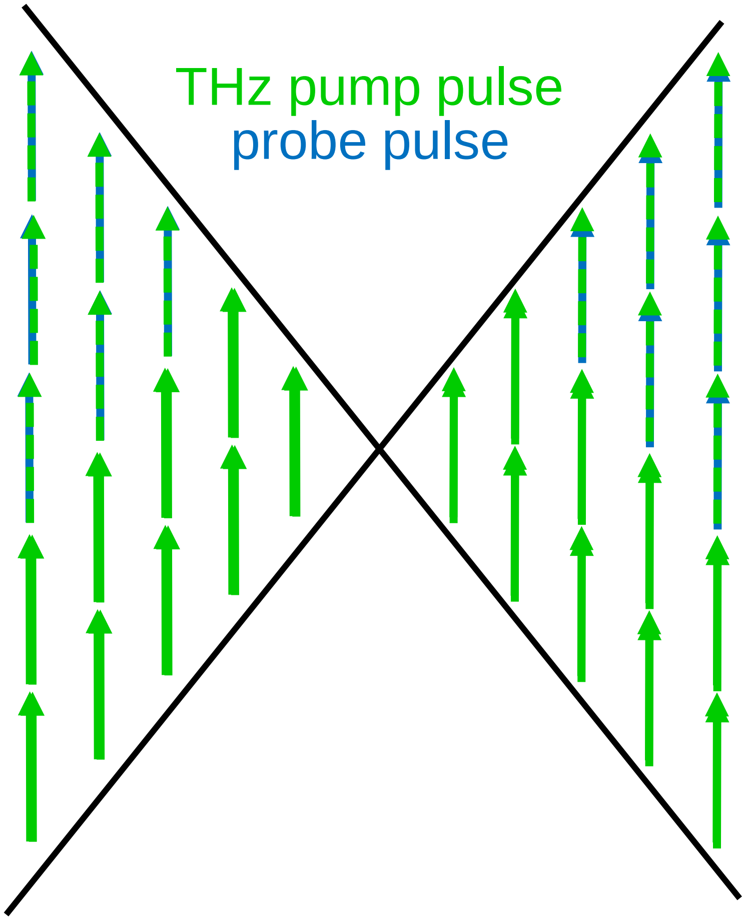
$$d_X = \frac{e_0 v_F}{\omega_{THz}} \cdot \frac{k_Y}{\sqrt{k_X^2 + k_Y^2}}$$



The transition dipole moment at 2 THz with 10 kV/cm is very large.
This is the **non-perturbative** regime.

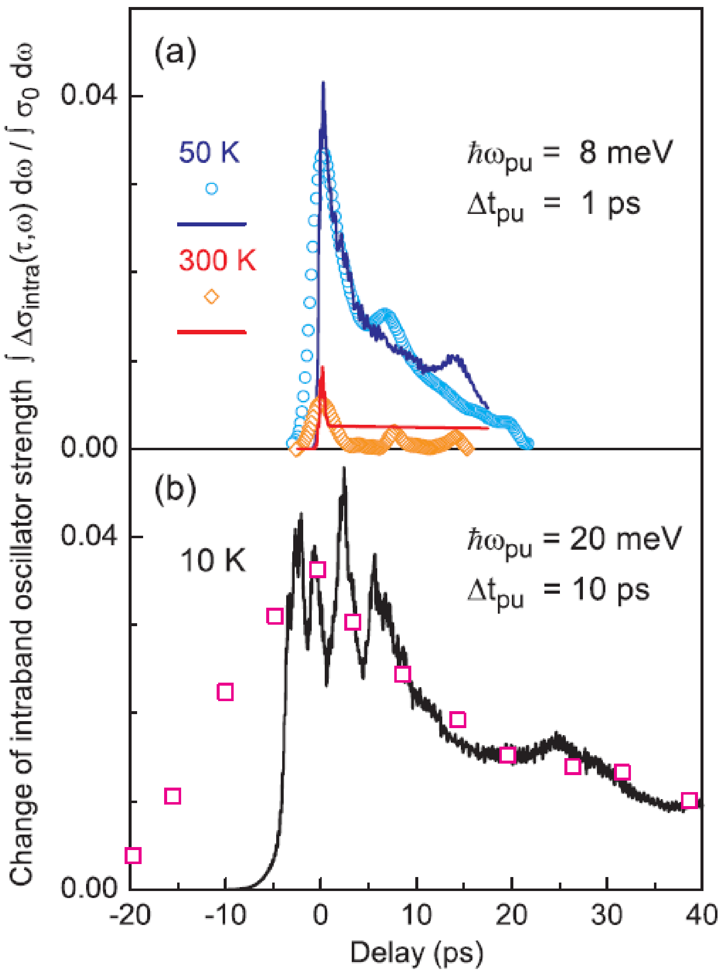
Bowlan et. al, PRB, **89**, 041408(R), 2014. and NJP, **16**,014017, 2014.

Radiative coupling and recombination in graphene



Graphene coherently stores energy from the pump pulse. This increases the absorption of the probe pulse until the stored energy is radiated away.

Model: radiative recombination and coupling

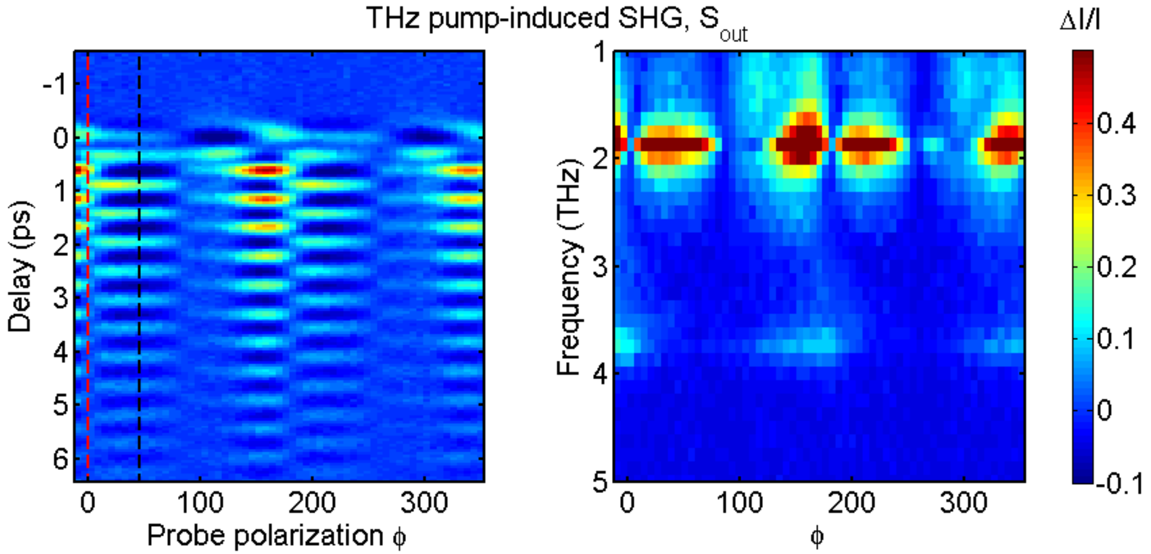


We solve the Schroedinger equation using the bandstructure, intra and inter-band transitions, radiative coupling, and **no scattering processes**.

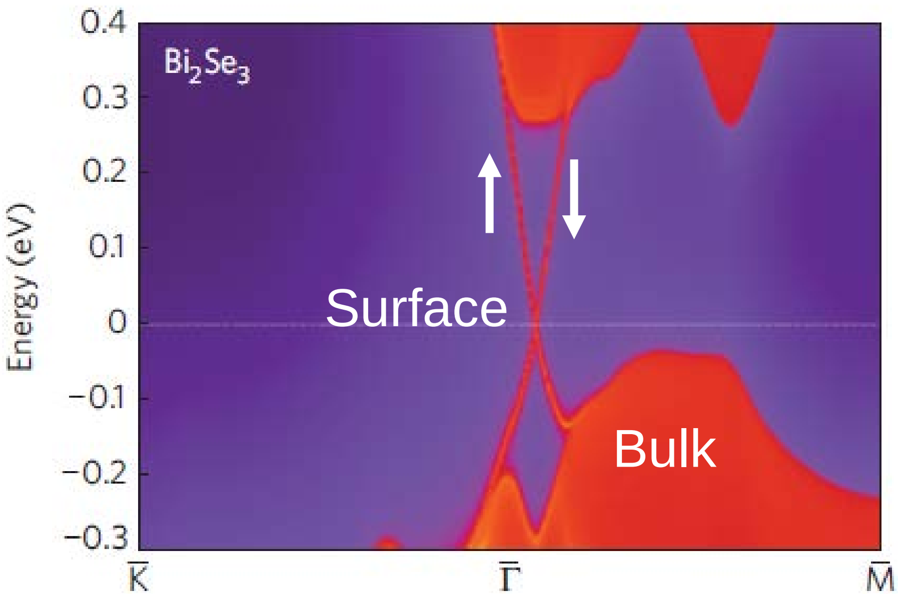
Bowlan et. al, PRB, **89**, 041408(R), 2014. and NJP, **16**,014017, 2014.

I will talk about…

1. Generating and measuring ultrashort THz pulses
2. Tracking ultrafast spin dynamics in antiferromagnets through spin wave resonances
3. Coherent two-dimensional THz spectroscopy
4. **Probing vibrational dynamics at a surface**

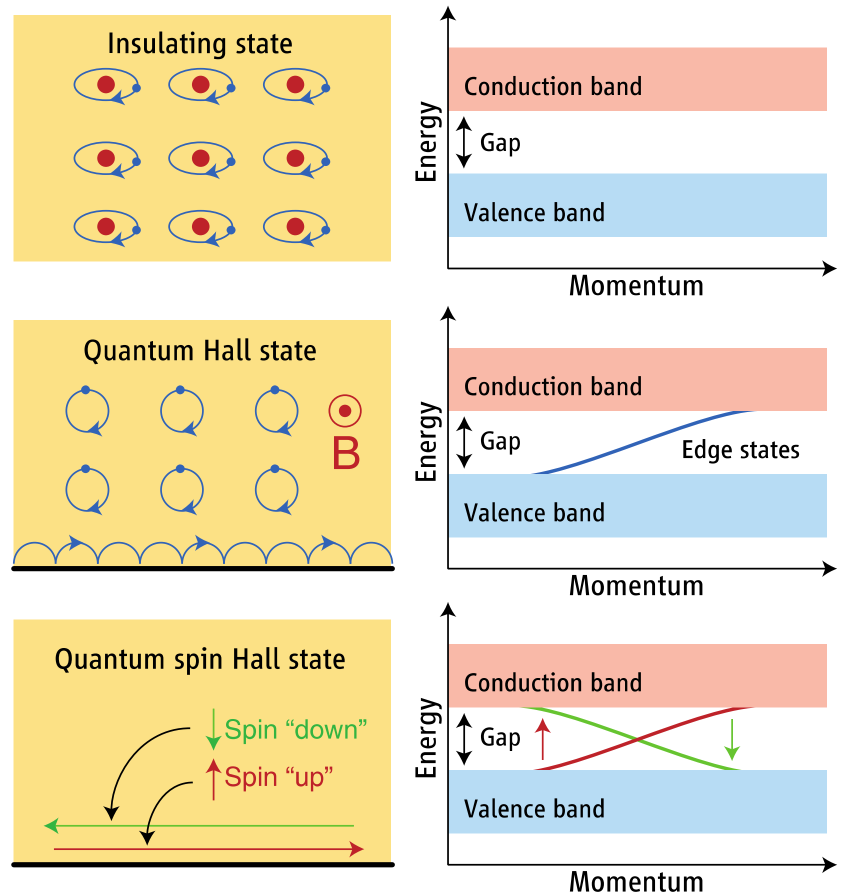


3D Topological Insulators are like graphene on the surface.



Haijun Zhang et. al., Nat. Phys., **5**, (2009)

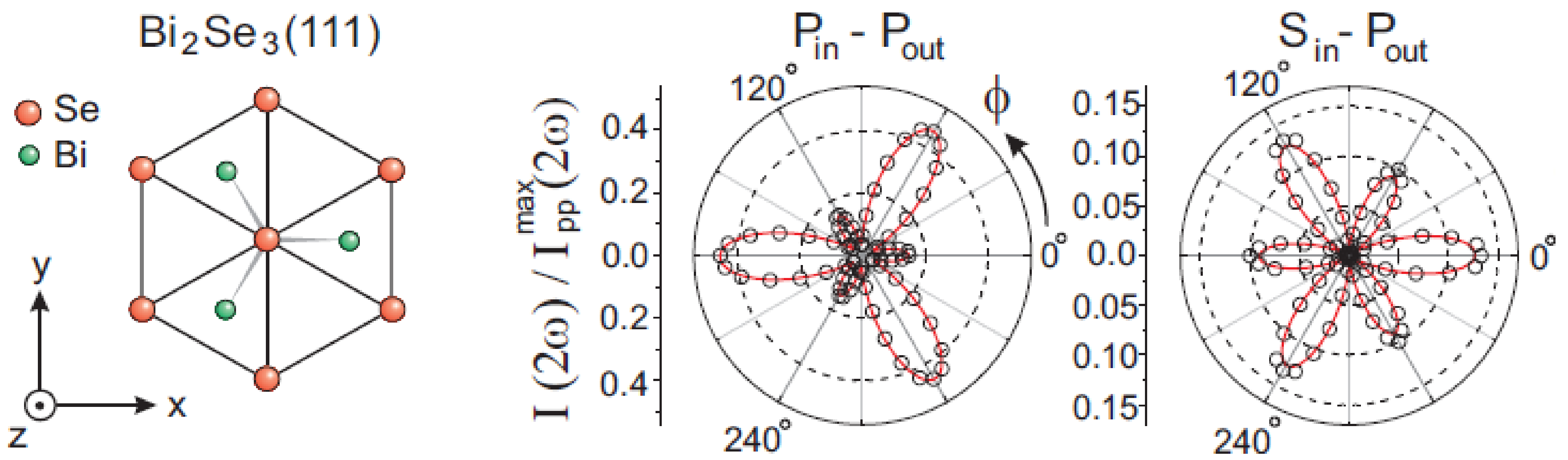
Symmetry, requires the presence a "topologically protected" conducting surface state.



C. Kane et. al., Science., **15**, (2006)

It is like a quantum Hall state without an applied B-field.

Probing material properties with optical second harmonic generation (SHG).



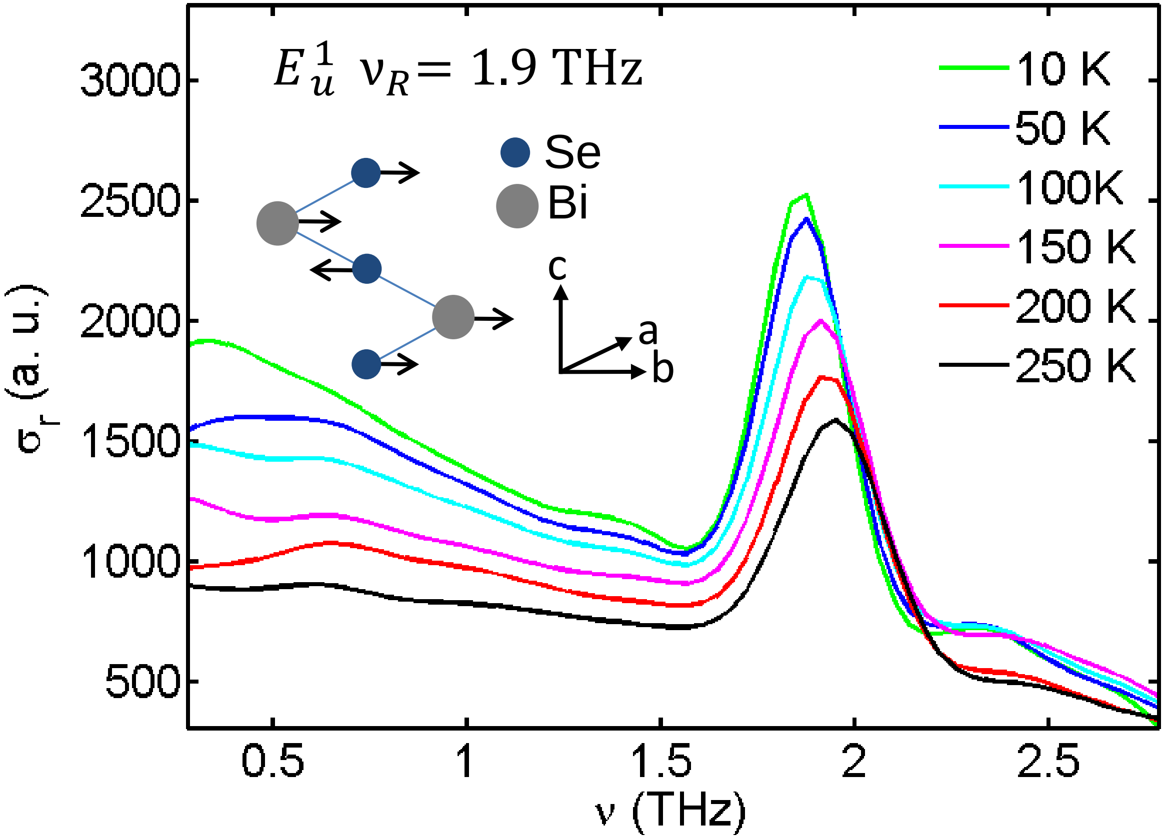
Hsieh et. al, PRL **106**, (2011)

$$E(2\omega) \propto P^{(2)}(2\omega) = \chi^{(2)} E^2(\omega)$$

SHG gives lattice symmetry information, and comes only from the surface when the bulk is inversion symmetric.

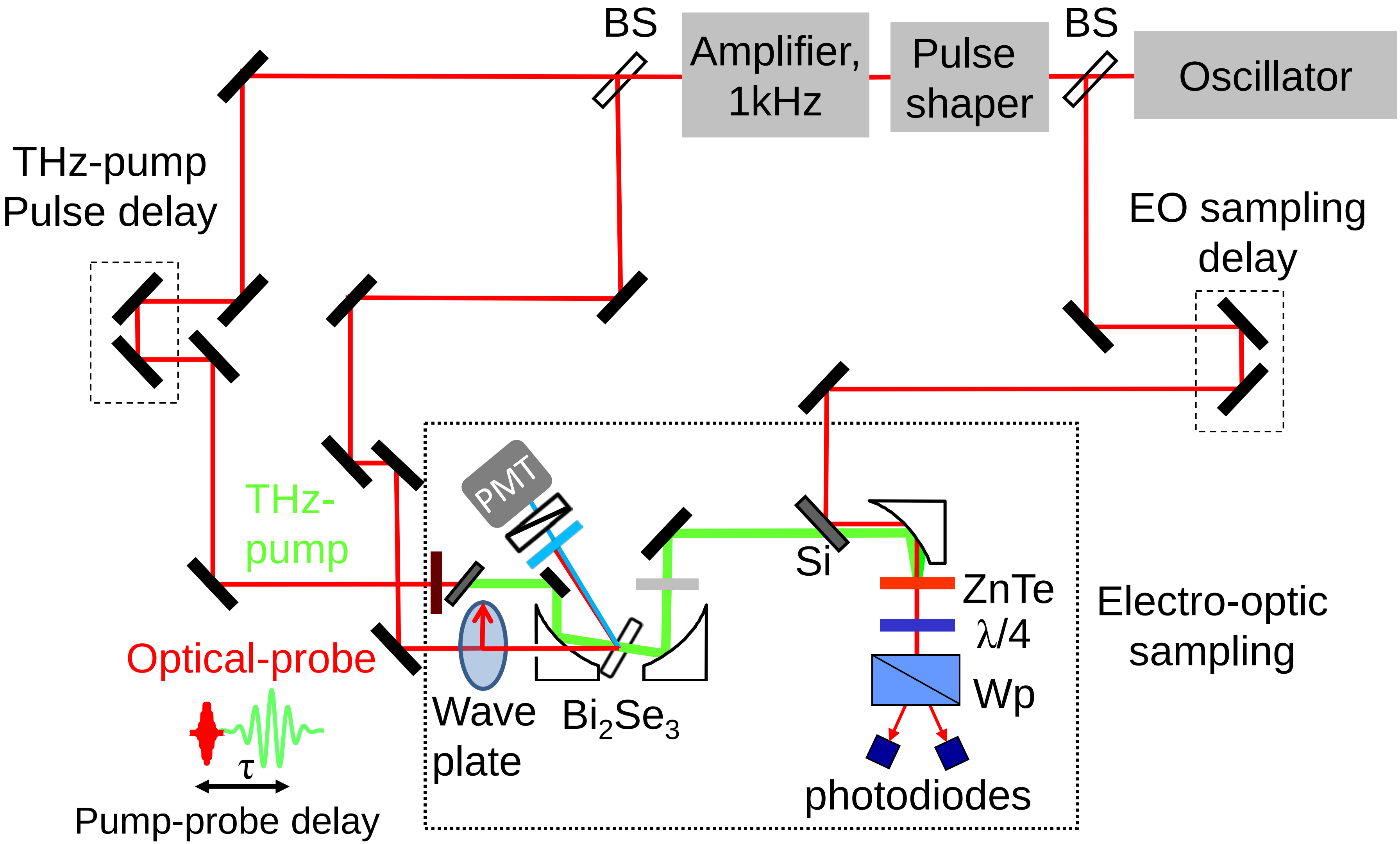
Studying topological insulators requires a surface sensitive probe.

Conductivity of our 25 nm thick Bi_2Se_3 film



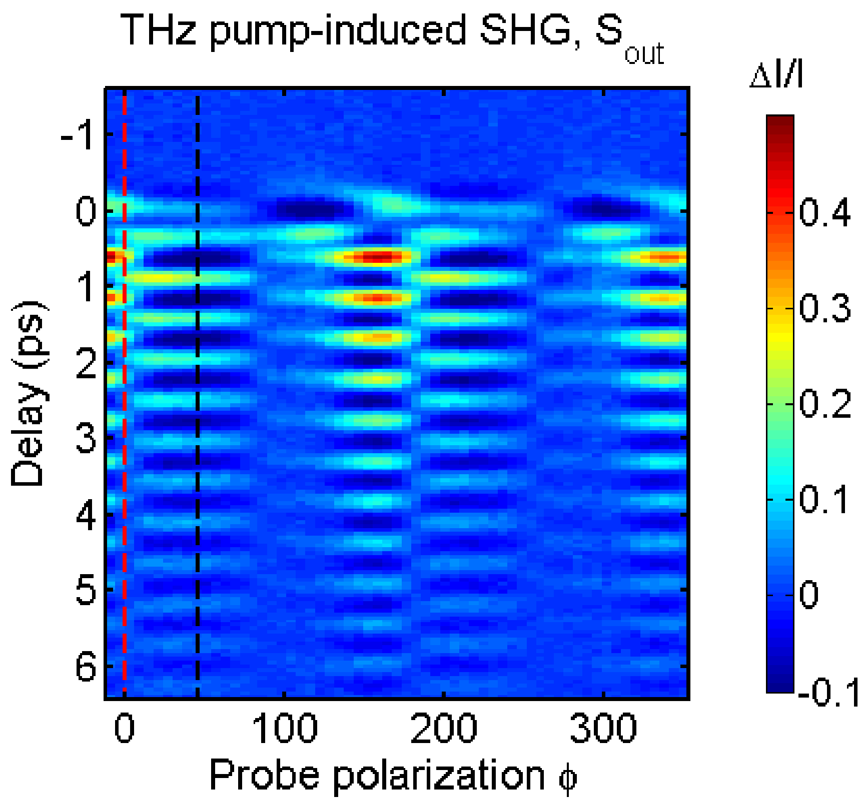
Besides the Drude response, there is also an IR active phonon at ~2 THz.

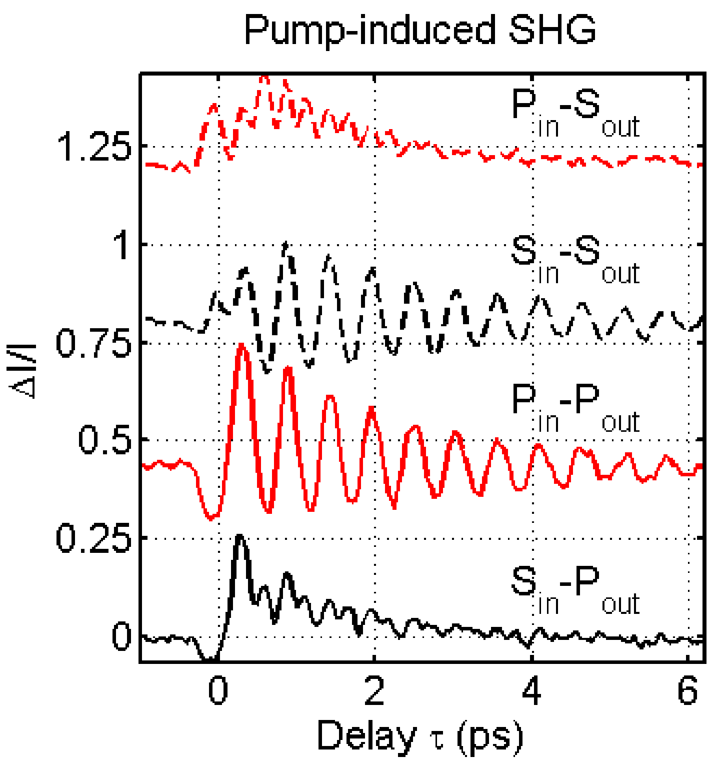
Measuring THz-induced symmetry changes



We study Bi_2Se_3 /Al_2O_3 25 nm thick films at a sample temperature of 10 K.

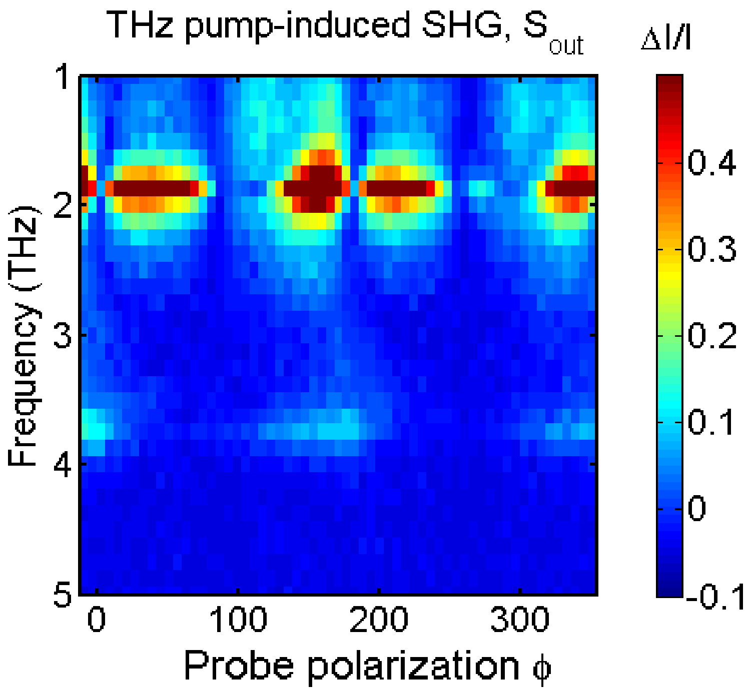
Nonlinear phonon dynamics in Bi_2Se_3

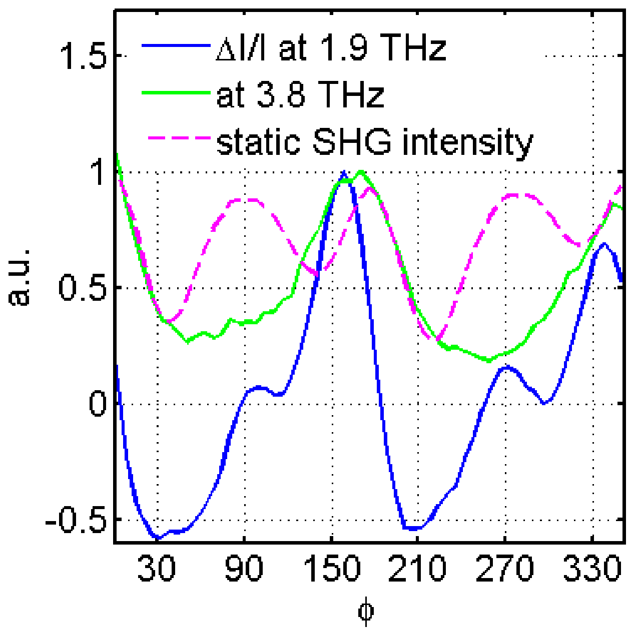




THz excitation results in osillations of the SHG which follow the phonon frequency and also its second harmonic.

The nonlinear and linear responses have different symmetries and field dependences.

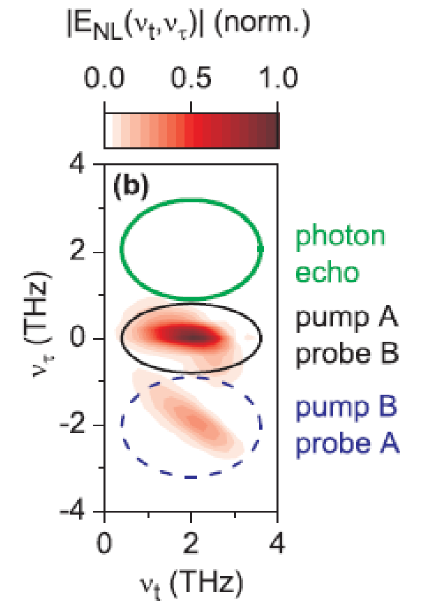




$$P_i^{2\omega}(\tau) \approx E_j^{\omega} E_k^{\omega} \chi^{(2)}(x(\tau))$$

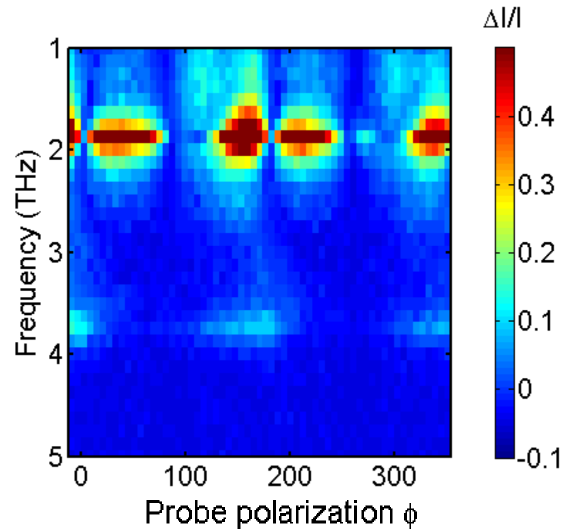
THz excitation of the phonon along the direction x modulates the nonlinear susceptibility so that $x(\tau) \propto \mathrm{A}(\tau)\cos(\omega_R \tau)$.

Conclusions



Coherent two-dimensinal THz spectroscopy: a poweful approach for studying coherence and dynamics of low energy resonances. Applying this to graphene we investigated the very strong THz light mater interaction which dominates over scattering. Useful for studying coupled excitations in multiferroics and monitoring chemical reactions.

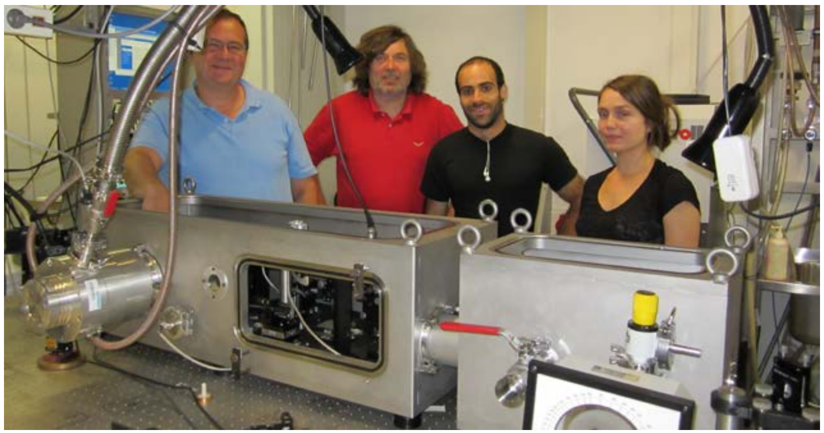
THz-pump, SHG-probe spectoscopy: an ultrafast, surface sensitive probe of atomic-scale symmetry changes and nonlinear phonon dymanics. We are using this in Bi_2Se_3 to investiage the nonlinear surface phonon dynamics. This is potentailly very useful for studying catalysis.



MBI LDRD CINT Center for Integrated Nanotechnologies Sandia Los Alamos LUMOS Laboratory for Ultrafast Materials and Optical Science

Acknowledgments

Max-Born institute for nonlinear optics and ultrafast spectroscopy, Berlin, Germany:



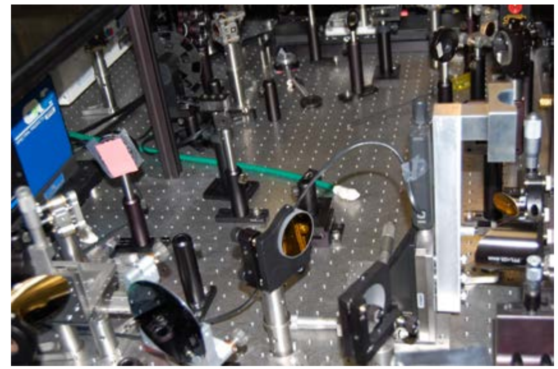
W. Kuehn, E. Martinez-Moreno, K. Reimann, M. Woerner, T. Elsaesser

Collaborators:

R. Hey, Paul-Drude-Institut, Berlin (GaAs samples)

C. Flytzanis, Laboratoire Pierre Aigrain, Paris

Laboratory for Ultrafast Materials and Optical Science:



R. Prasankumar and D. Yarotski (mentors), J. Bowlan, Y. Dai, B. McFarland, T. Taylor.

Collaborators:

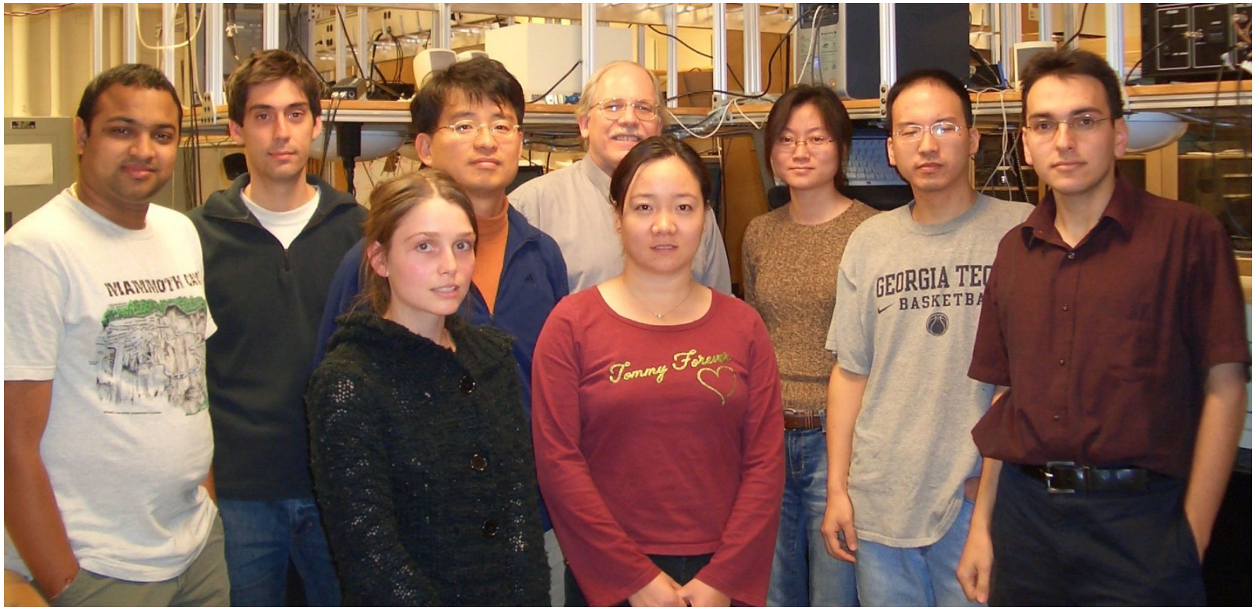
S. Trugman, CINT (theory)

J. Furdyna and X. Liu at Notre Dame (Bi_2Se_3 thin films)

R. Valdes Agular, The Ohio State University

N. J. Hur, Inha University, Icheon, Korea and S-W. Cheong Rutgers University ($RMnO_3$) crystals

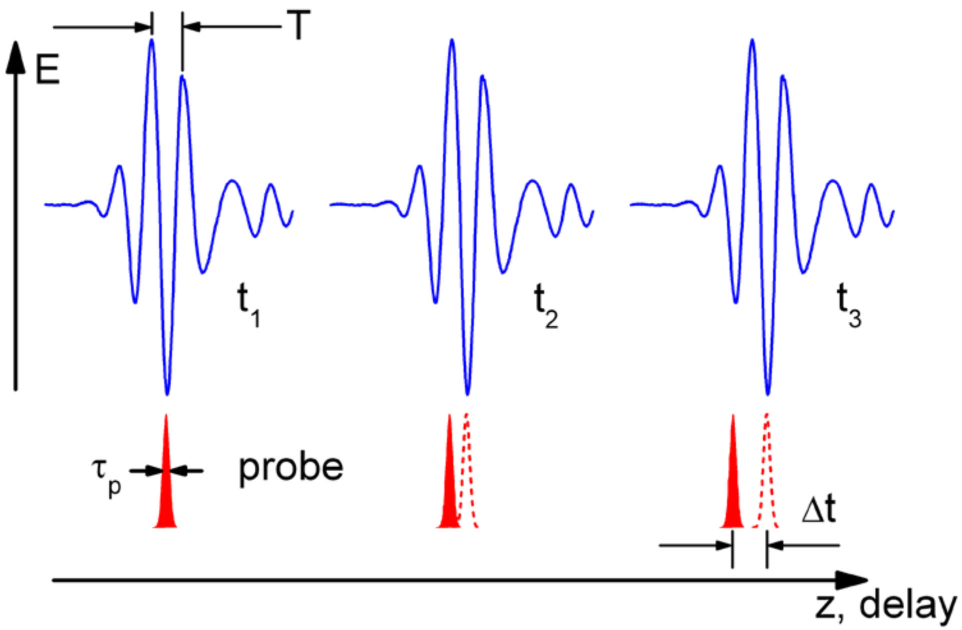
My thesis work: Rick Trebino's ultrashort pulse characterization group at Georgia Tech

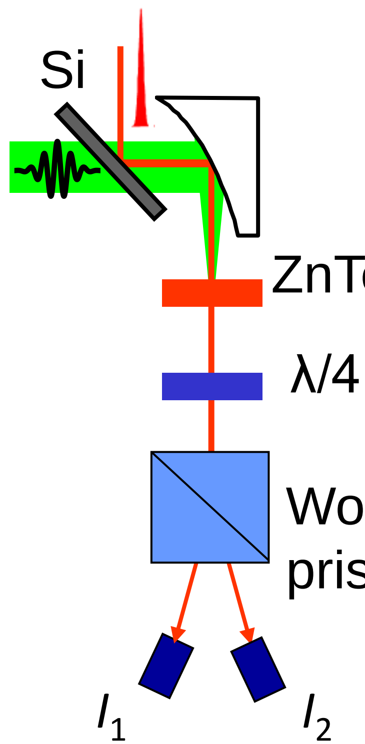


Collaborators:
Peeter Saari, university of Tartu in Estonia.
Ulrike Fuchs, Fraunhofer Institute in Jena, Germany
Robert Levis, from Temple University

This work was partially funded by NSF fellowship IGERT-0221600.

Measuring THz E-fields with the linear electro-optic effect.



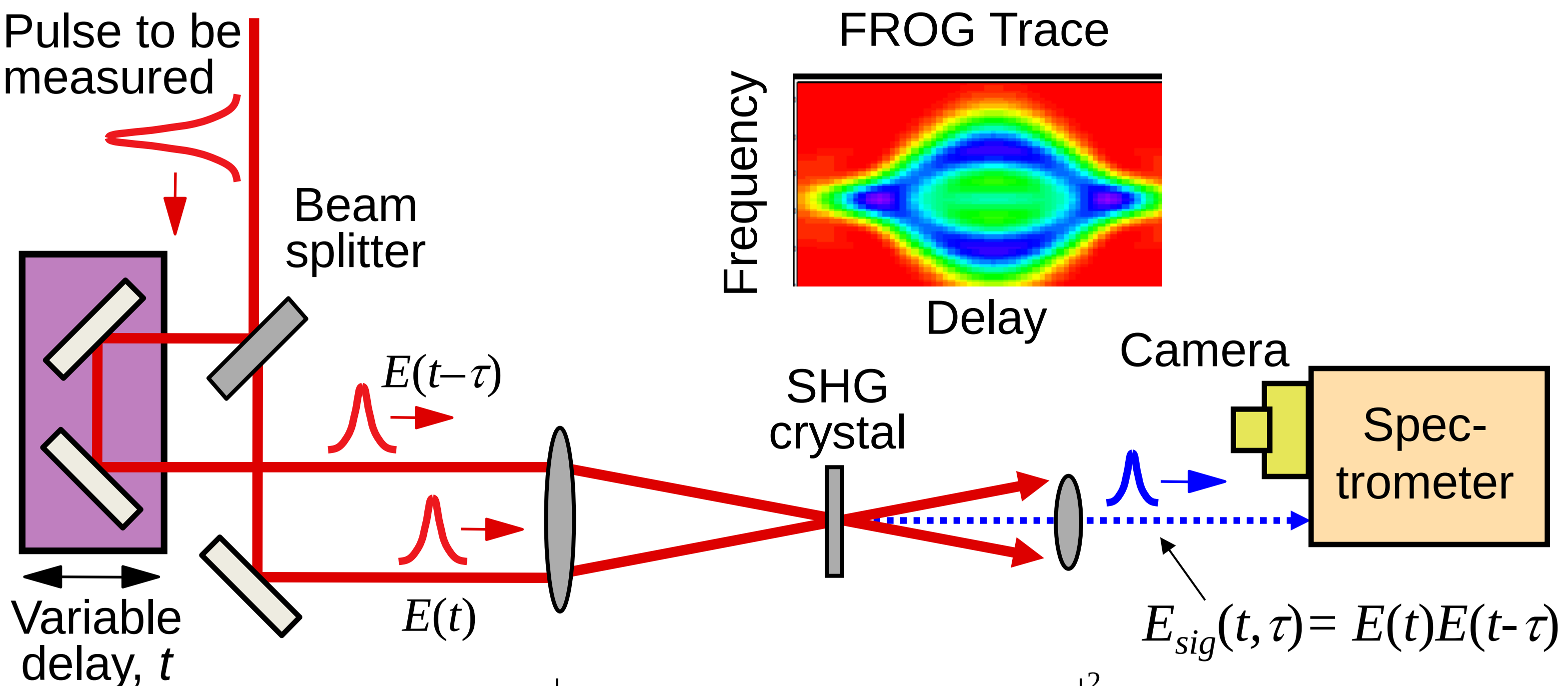


$$\Gamma = \frac{2\pi d}{\lambda} n^3(\lambda) r_{41} E_{THz} \qquad \frac{I_1 - I_2}{I_1 + I_2} = \sin(\Gamma) \approx \Gamma$$

The Induced birefringence, and polarization rotation are proportional to the THz electric field.

Wu, Zhang, APL **71**, 1285 (1997)

Measuring ultrashort pulses with Frequency resolved Optical Gating (FROG).

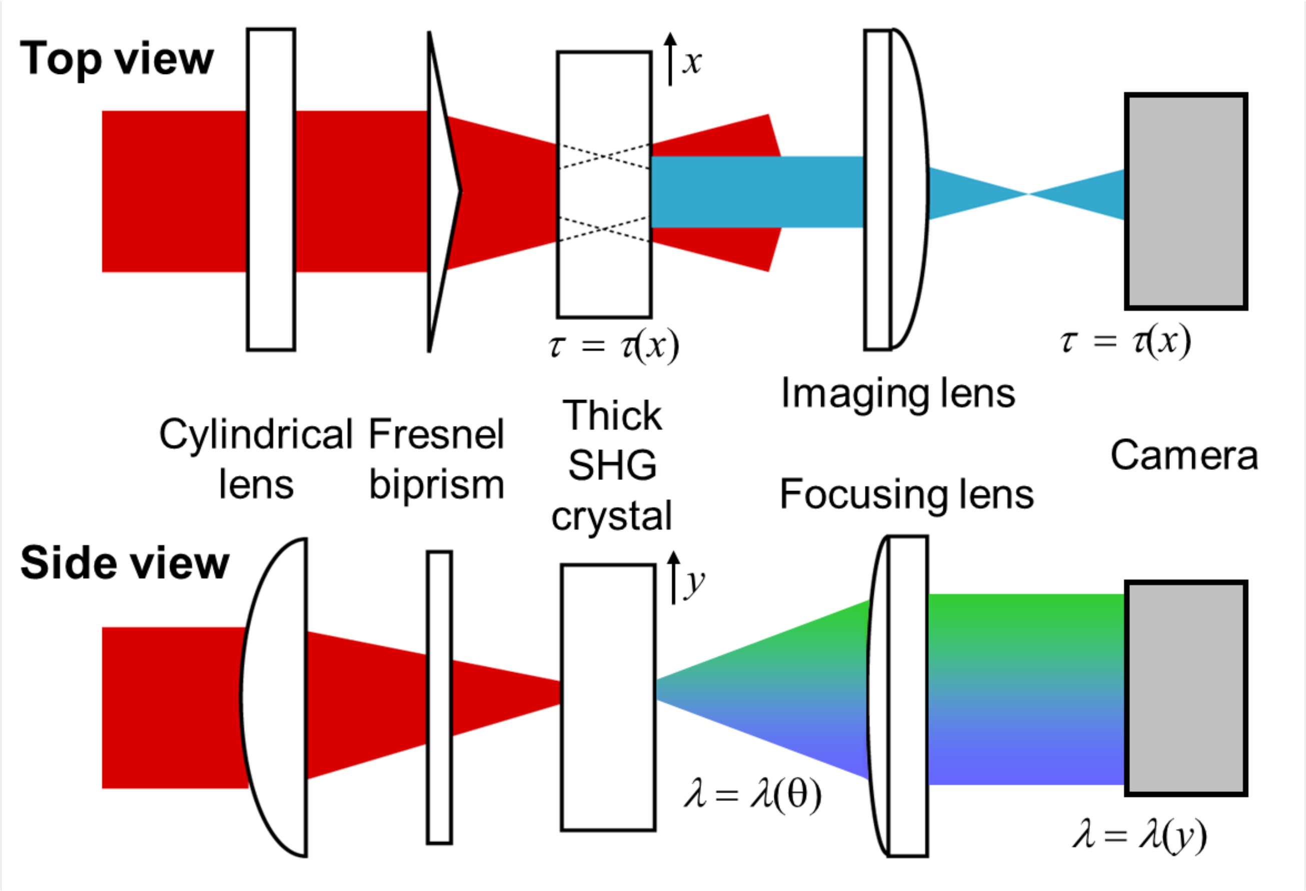


$$I_{FROG}(\omega,\tau) = \left| \int_{-\infty}^{\infty} E_{sig}(t,\tau)\exp(-i\omega t)\,dt \right|^2$$

Measuring the spectrum of the autocorrelation allows one to determine amplitude and phase of the optical pulse.

Trebino et. al., Revs. Sci. Instrum. **68**, 3277 (1997)

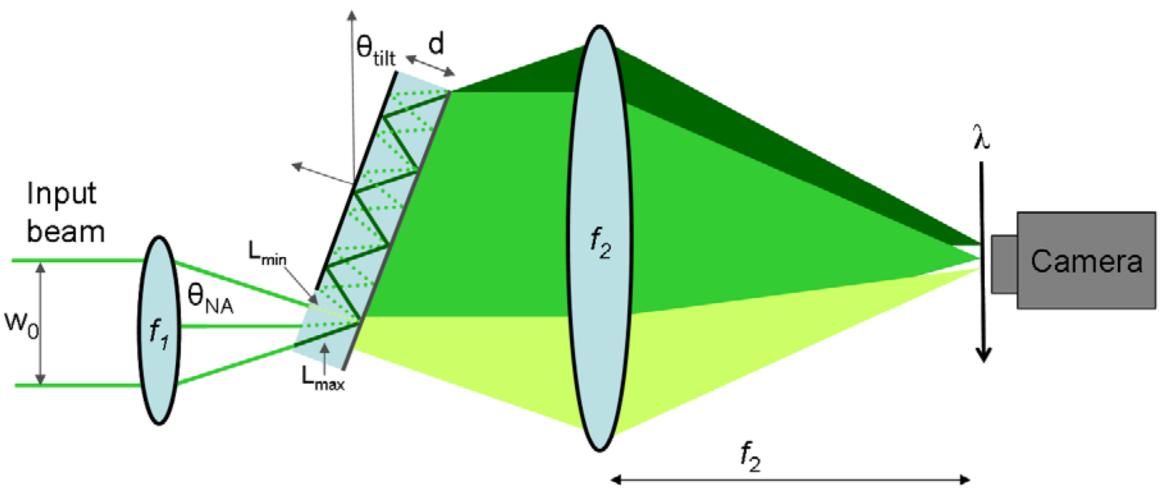
Single-shot FROG: GRENOUILLE



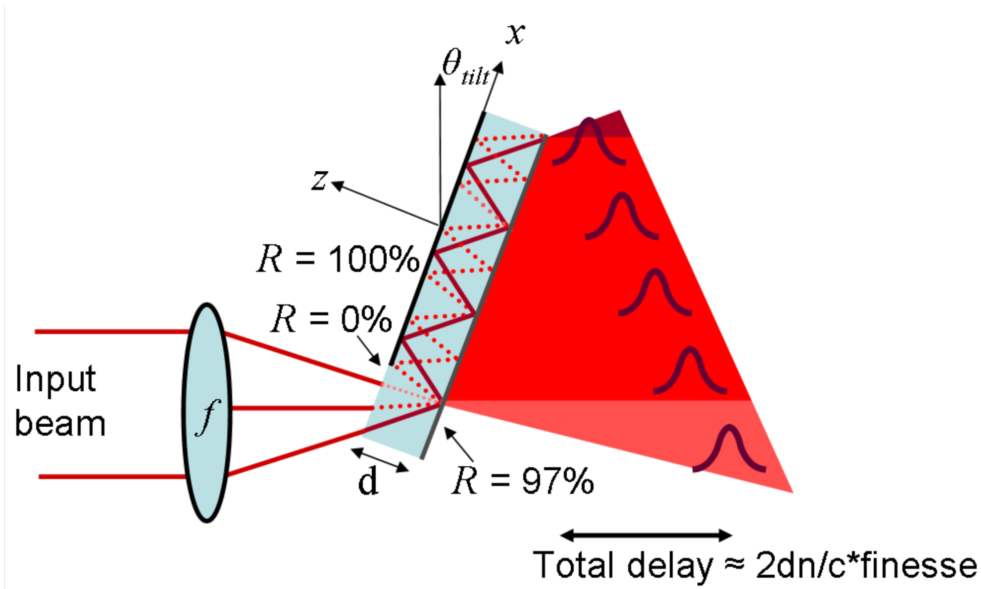
GRENOUILLE is a complete single-shot FROG, but is more sensitive.
It uses the standard SHG FROG algorithm.

P. O'Shea, et. al., Opt. Lett. **26**, 932 (2001)

A single-shot nanosecond FROG



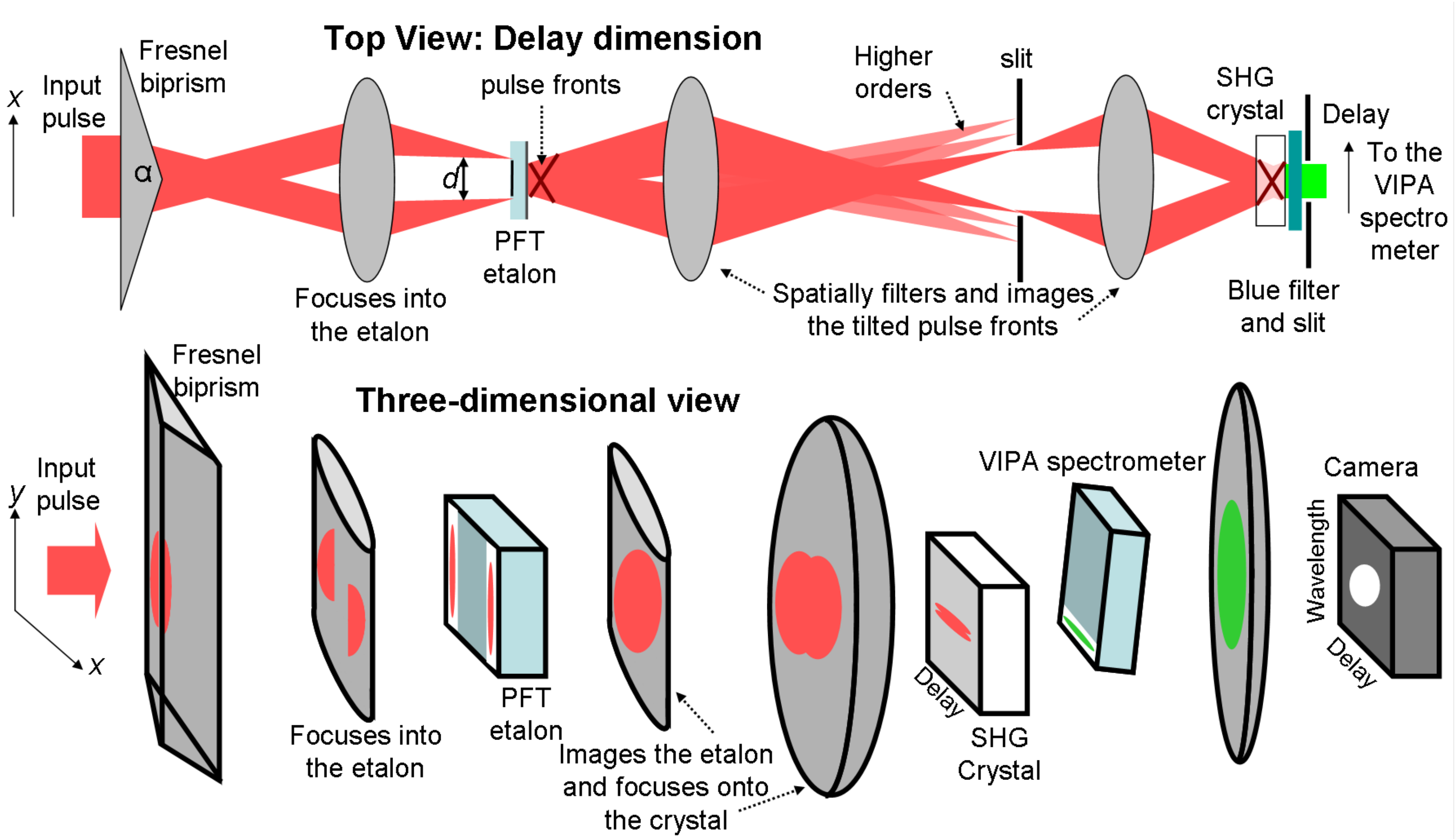
An Etalon spectrometer gives us 0.1 picometer spectral resolution.



Using the ~98º pulse front tilt from the etalon gives a 10 ns delay range.

P. Bowlan et. al., Opt. Express, **19**, 1367 (2011) and P. Bowlan et. al. JOSA B, **27**, 2322, (2010).

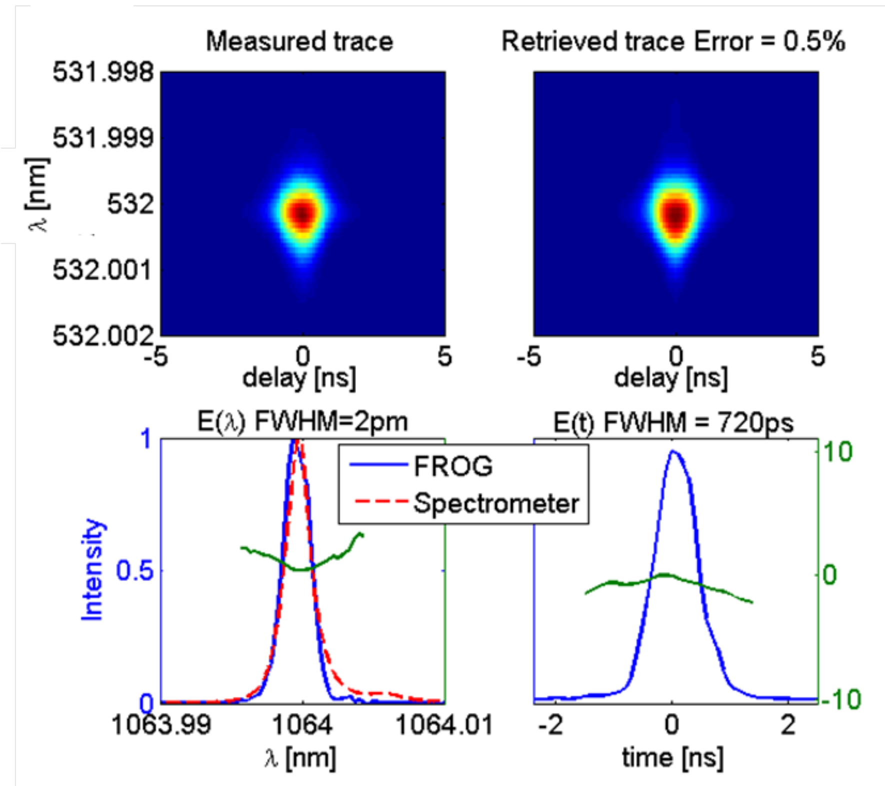
Single-shot nanosecond FROG



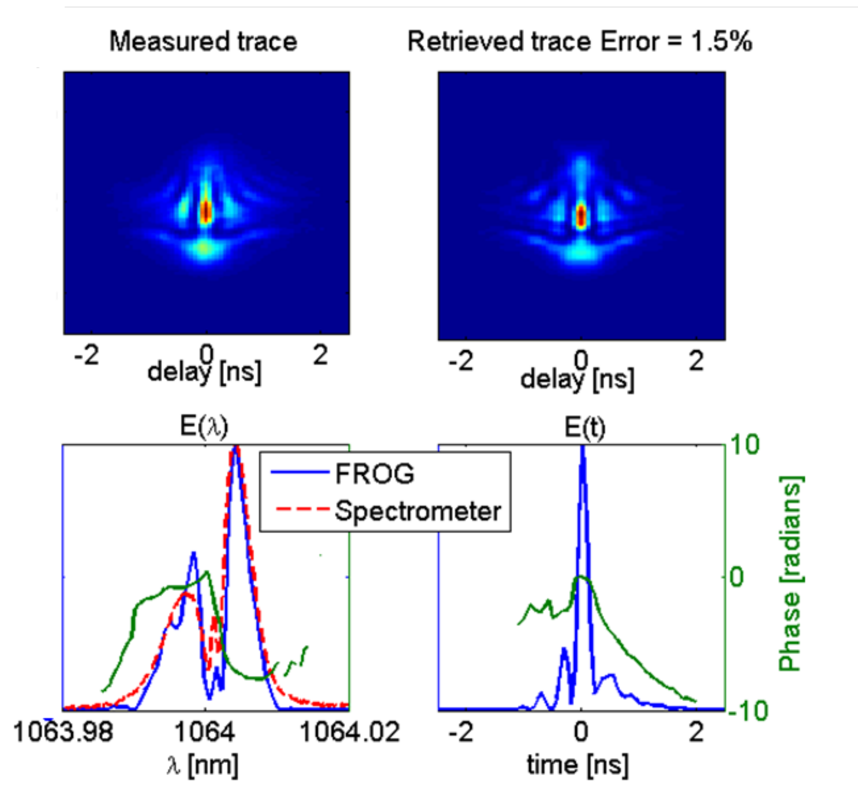
This results in a single-shot spectrally resolved autocorrelation at the camera with ns temporal and pm spectral resolutions.

P. Bowlan et. al., Opt. Express, **19**, 1367 (2011), supported by DARPA .

Single-shot nanosecond pulse measurements



Pulse from a thin-disk laser

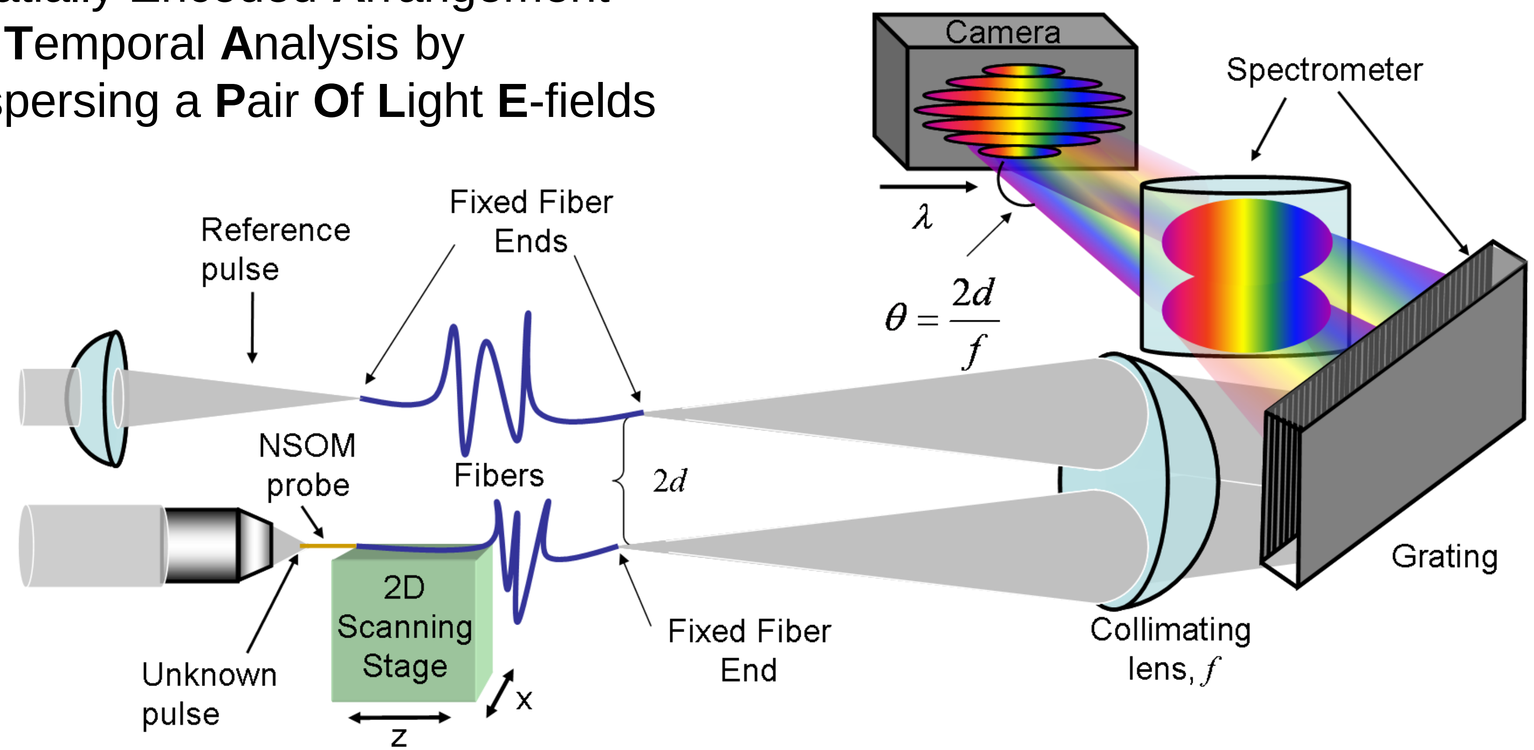


After amplification in a Yb-doped fiber

P. Bowlan et. al., Opt. Express, **19**, 1367 (2011).

Adding dimensions to optical laser pulse diagnostics: spatiotemporal characterization

Spatially **E**ncoded **A**rrangement for **T**emporal **A**nalysis by **D**ispersing a **P**air **O**f **L**ight **E**-fields

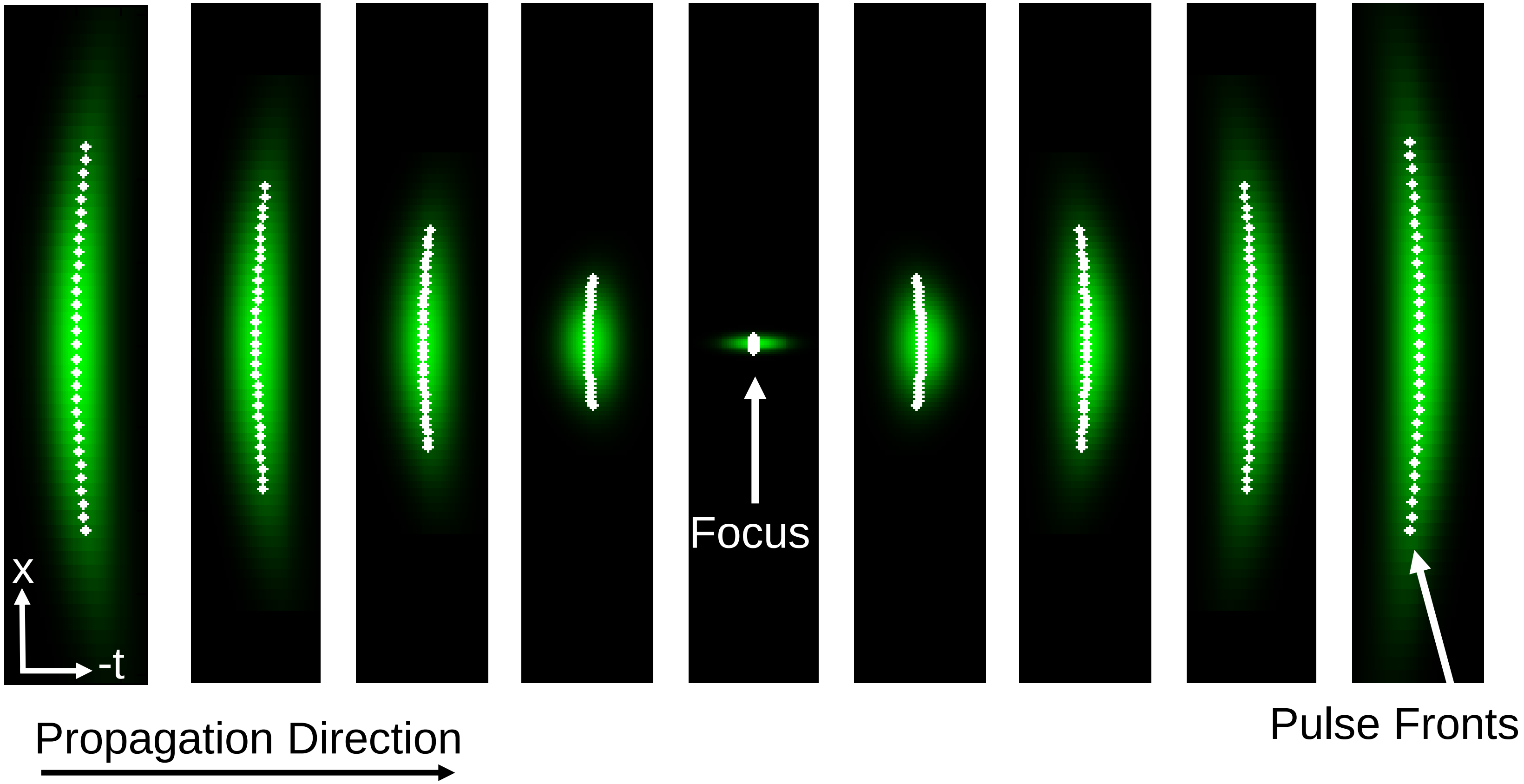


Combining spectral interferometery with Near Field Scanning Microscopy, achieves nanometer and femtosecond spatiotemporal resolutions.

P. Bowlan et. al., Opt. Express, **16**, 13663 (2008).
For a single-shot technique see: P. Gabolde et. al., Opt. Express, **14**, 11460 (2006).

An Ideal focused ultrashort pulse

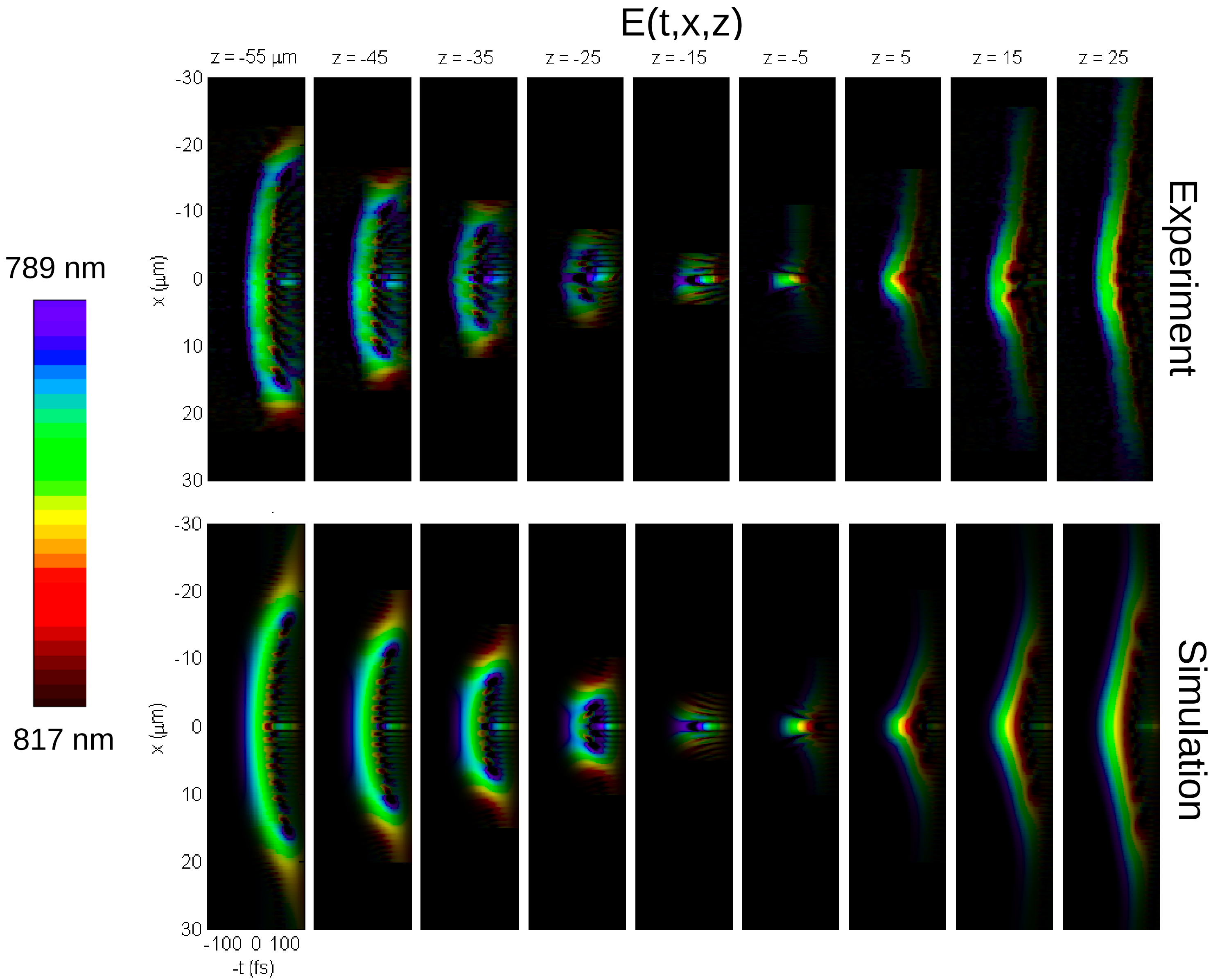
Spatiotemporal snap snots in flight of a focusing pulse (E(x,t))



When no distortions are present the pulse's color is the same everywhere in space and time, so that the Intensity at the focus is maximal

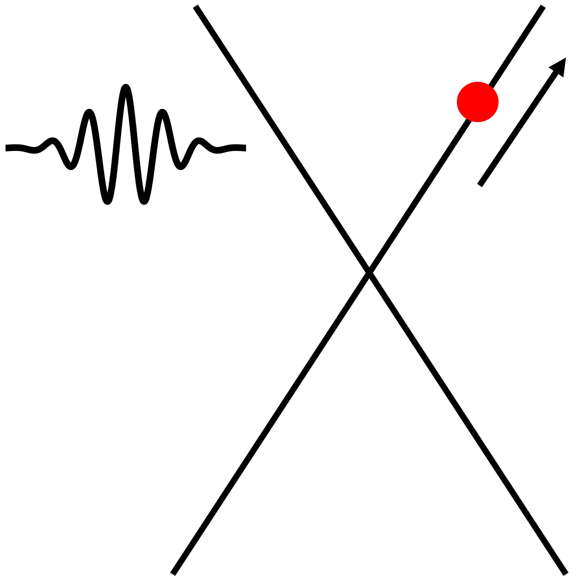
For simulations: M. Kempe, et. al. , Phys. Rev. A **48**, 4721-4729 (1993).
Ulrike Fuchs, et. al., Opt. Express **13**, 3852-3861 (2005).

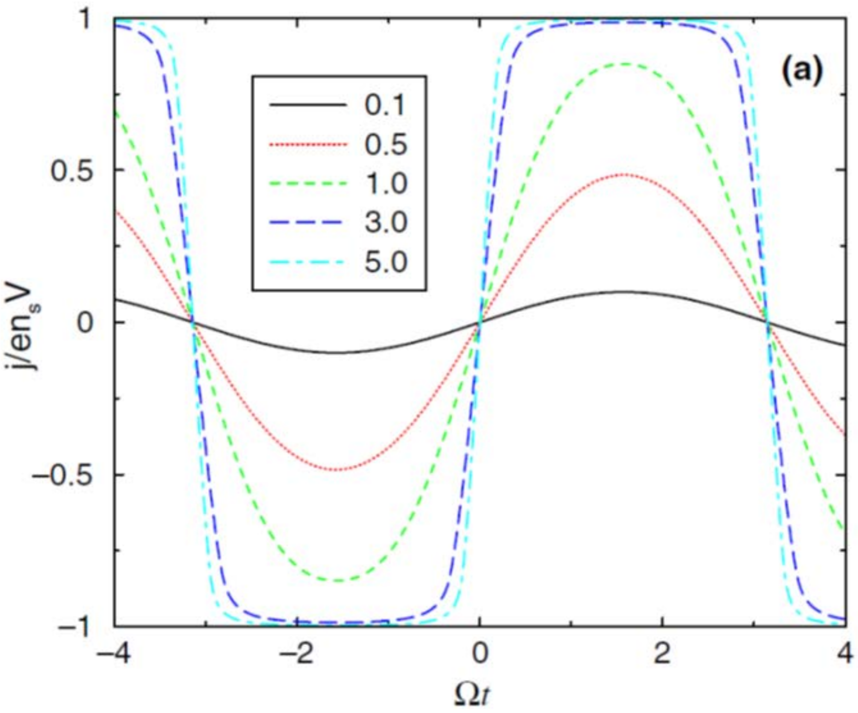
Measured Pulse focused with an aspheric lens (NA = .4)



The combination of overfilling the lens, and chromatic aberrations result in an additional pulse that is ahead of the main pulse

THz transport in graphene

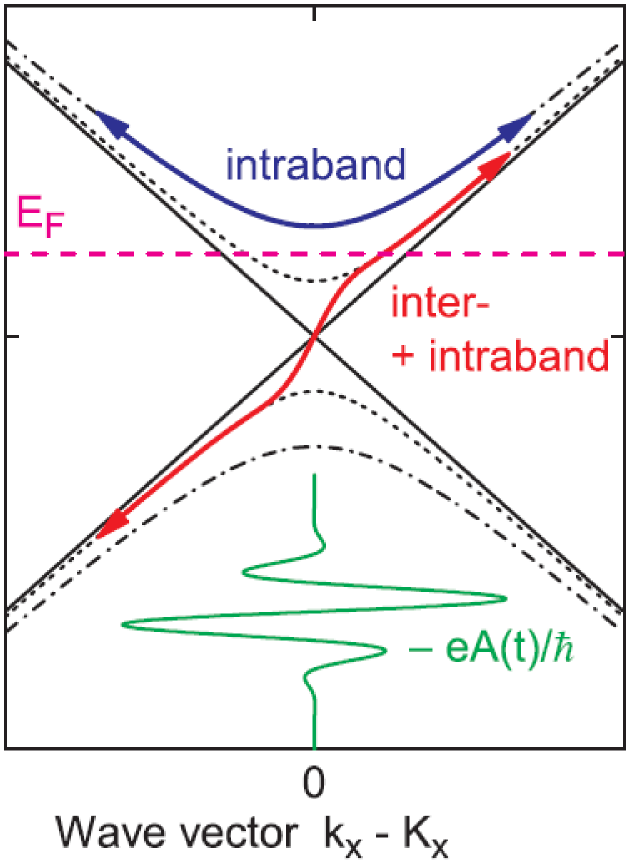


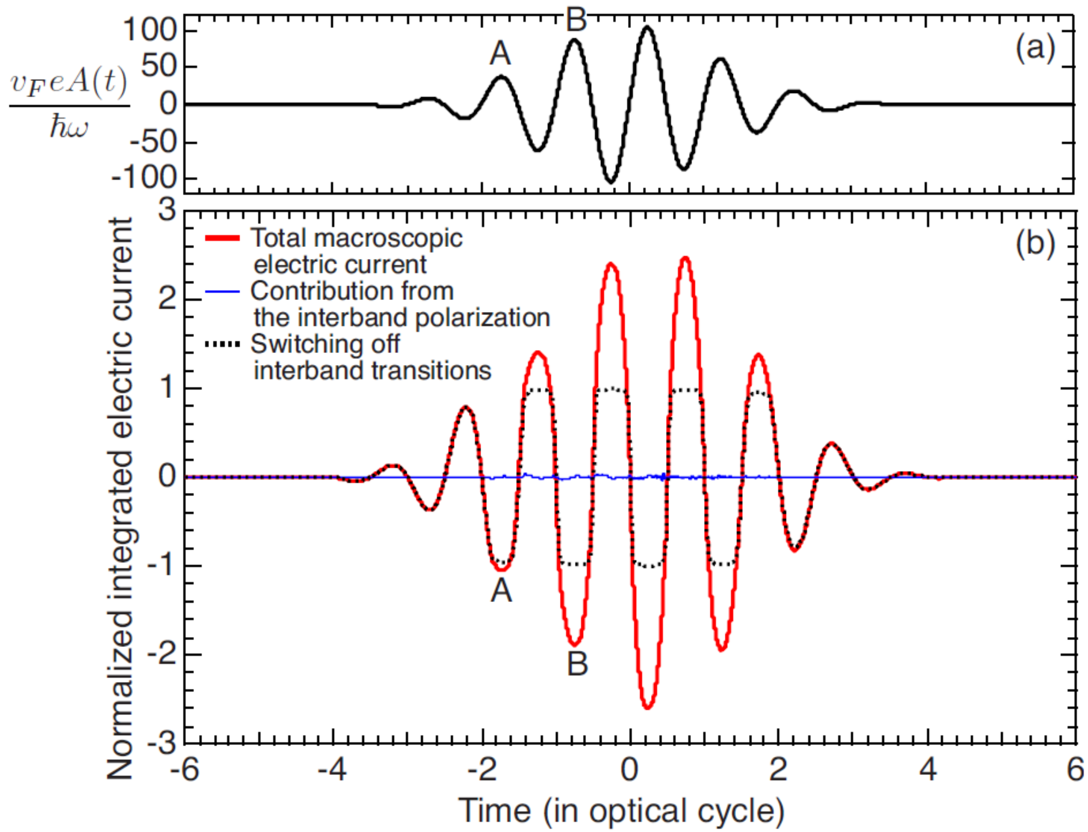


Very efficient THz harmonic generation was predicted in graphene if only intra-band current is considered.

S. A. Mikhailov 2007 *EPL* **79** 27002

Strong THz pulses result in simultaneous electron-hole pair generation and transport.

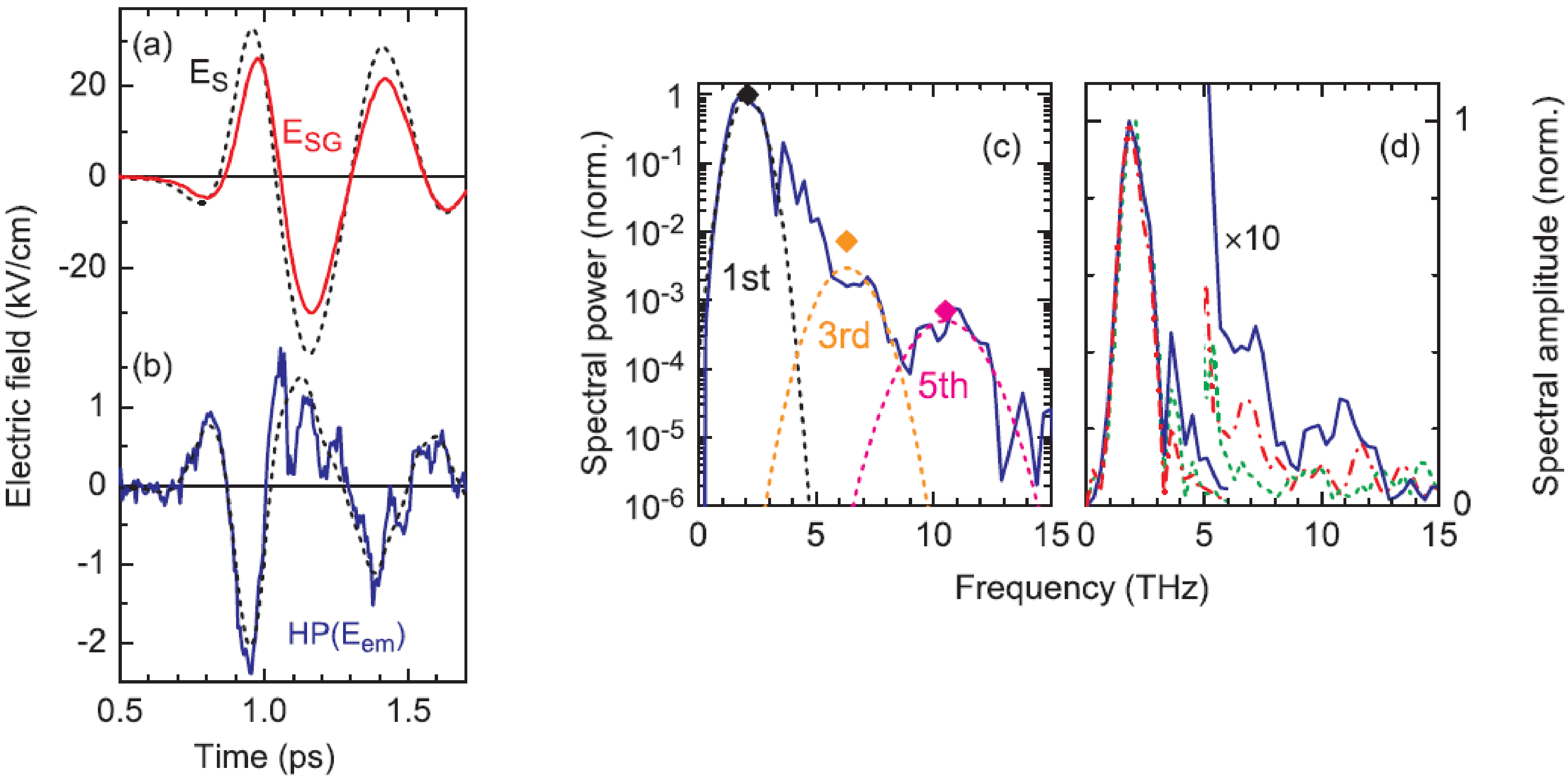




K. L. Ishikawa, PRB **82**, 201402(R) (2010)

Including the interband current partially cancels out the harmonics.

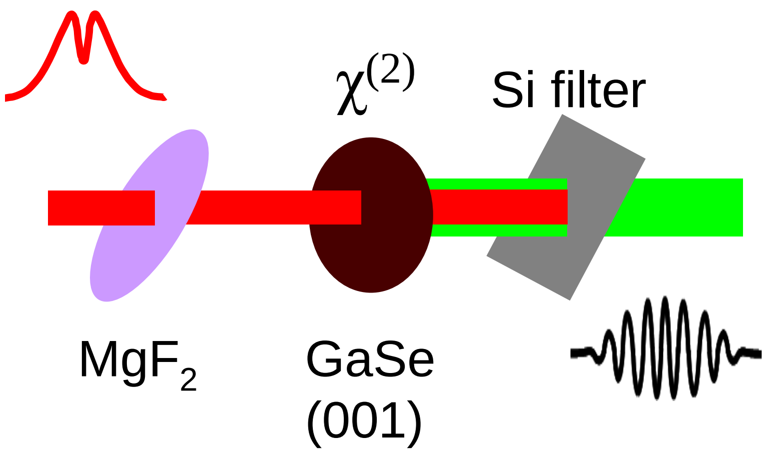
In transmission we see weak harmonics

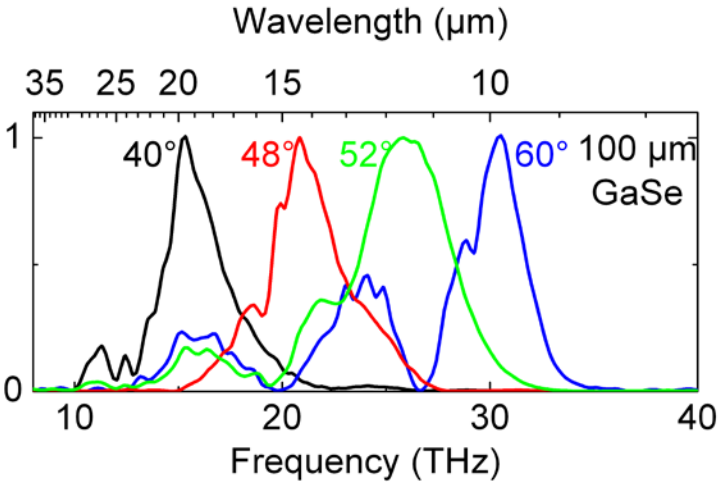


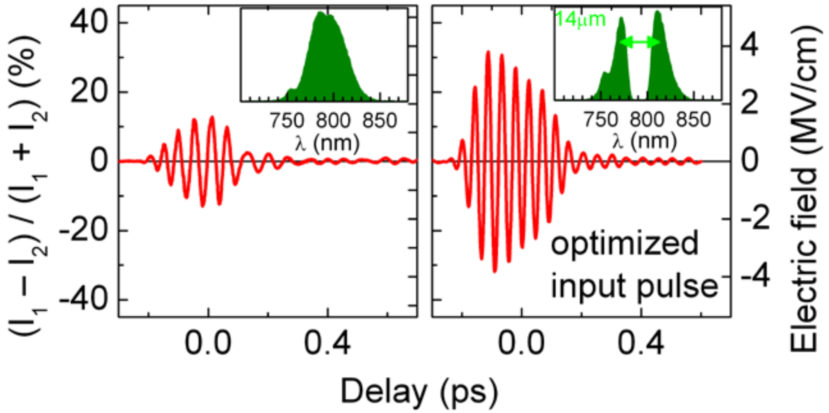
This is consistent with our radiative recombination picture. Without intraband absorption, the model does not reproduce our results.

Bowlan et. al, PRB, **89**, 041408(R), 2014.

Mid-infrared frequencies can be generated in GaSe too.

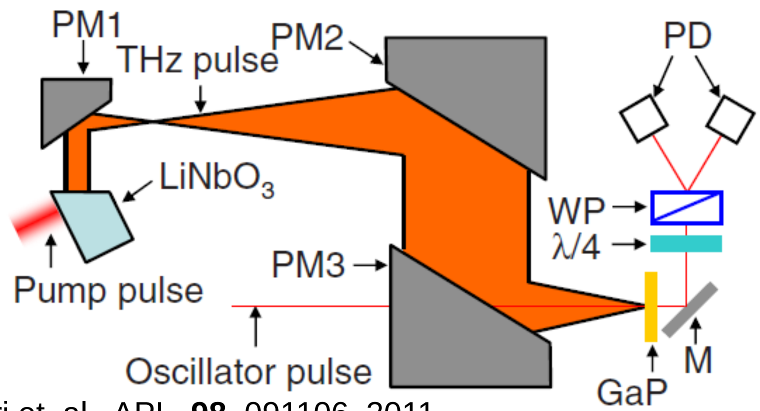






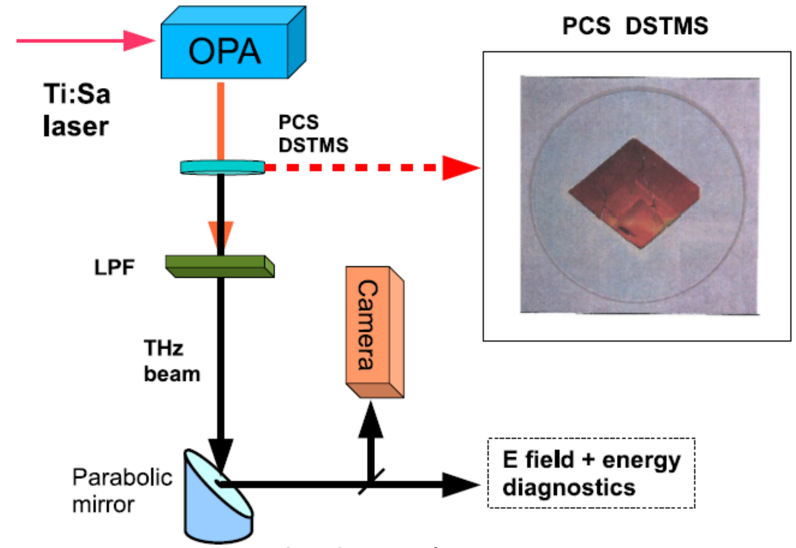
Methods of intense THz pulse generation

Tilted Pulse front method in $LiNbO_3$



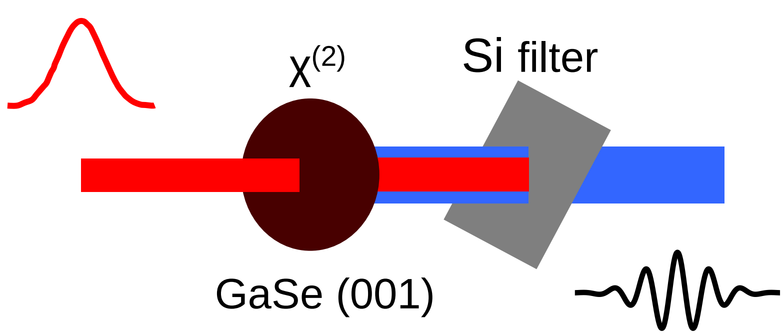
Hirori et. al., APL, **98**, 091106**,** 2011.

Organic crystals

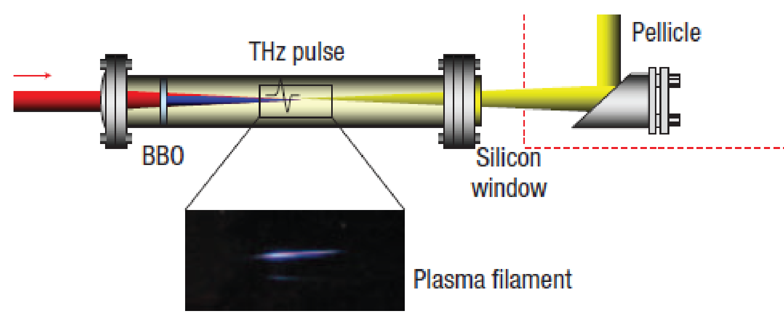


Vicario et. al., PRL, **112**, 213901, 2014.

Semi-conductor crystals

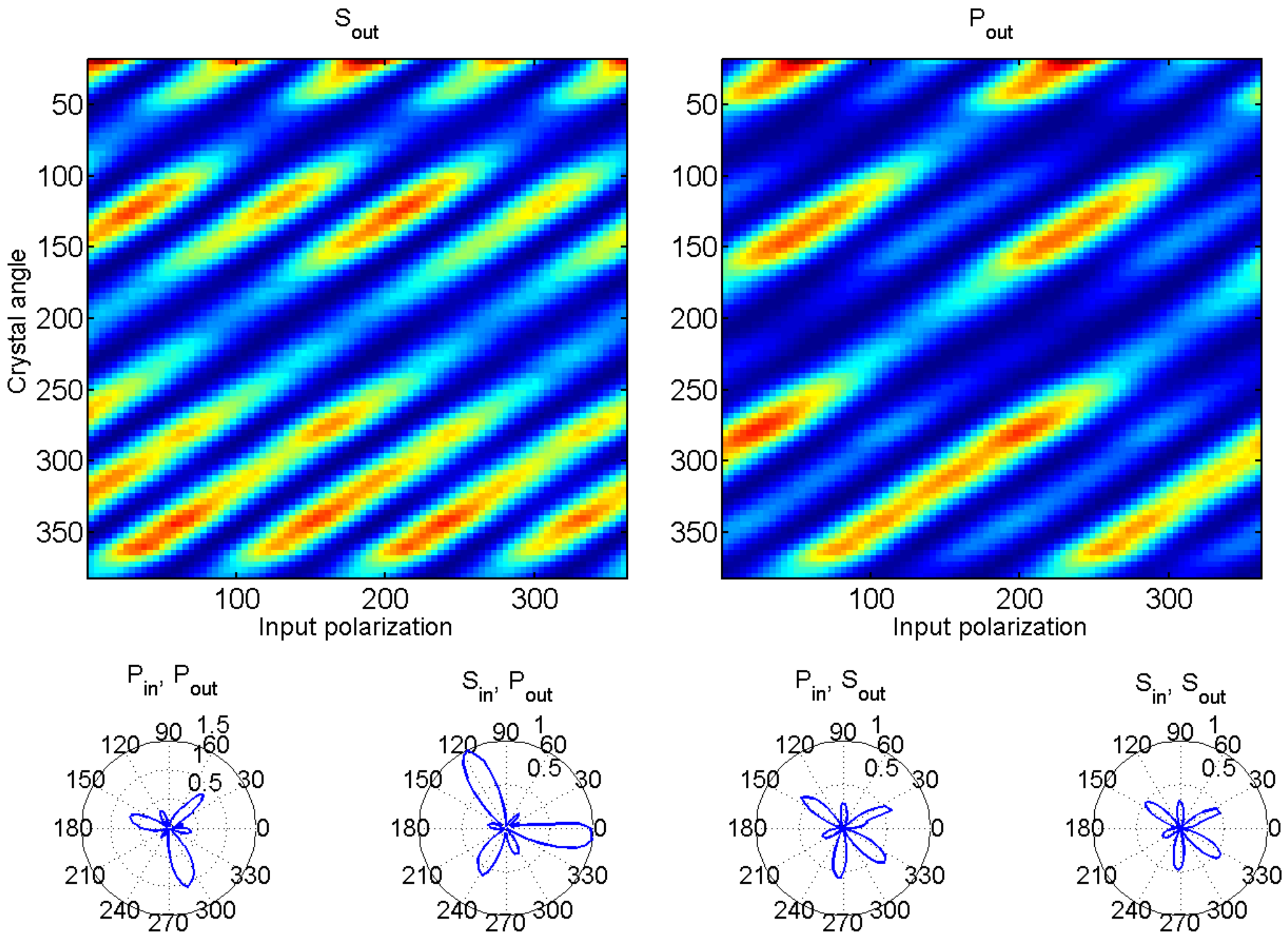


In a Plasma



Kim et. al., Nature Photonics, **2,** 605**,** 2008.

10 nm thick Bi2Se3 film



Pout is 3.6 times larger and Sout is 2.2 times larger, compared to the thicker film.

25 nm thick Bi2Se3 film