Ultrafast demagnetization of ferromagnetic films

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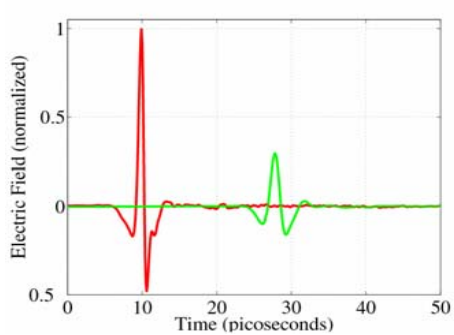
Outline

- Applications of Ultrafast THz
- THz emission mechanisms
  - Current Surge
  - Optical rectification
  - THz emission from Metals – Ultrafast Demagnetization
  - Towards higher electric field sources
- Ultrafast Demagnetization in Iron
  - History of ultrafast magnetization changes
  - Terahertz Emission Spectroscopy

THz Time Domain Spectroscopy

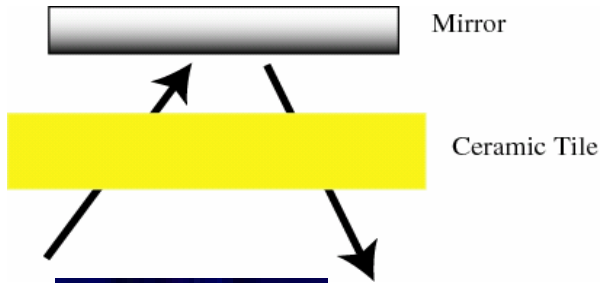
- Terahertz frequencies and THz -TDS

  (1 THz → 4 meV → 33 cm^{-1} → 300 μm)



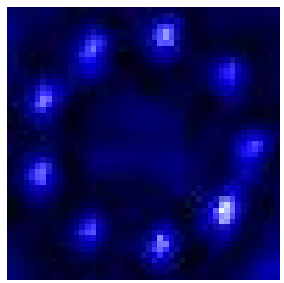
- Material characterization and bistatic ranging

  Feasibility studies (RCS characterization) for spaced-based broadband radar (FEM at 300 GHz with >30 GHz bandwidth)
  Increased range resolution, detection of embedded materials



- NDE of energetic materials

  Imaging of voids in plastic bonded explosives,
  THz spectroscopy of single crystal HMX

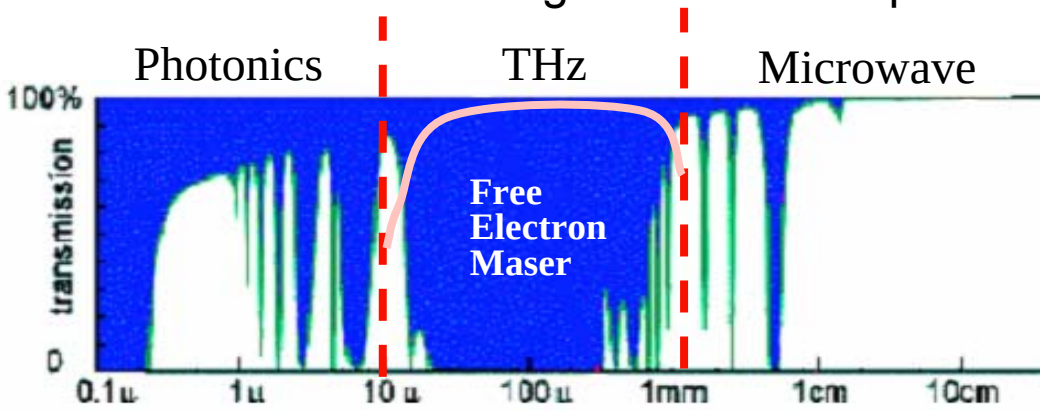


- Sensor Negation

  THz frequencies are only 1 to 2 orders of magnitude faster that the frequencies of electronics.

Terahertz time domain spectroscopy (THz-TDS)

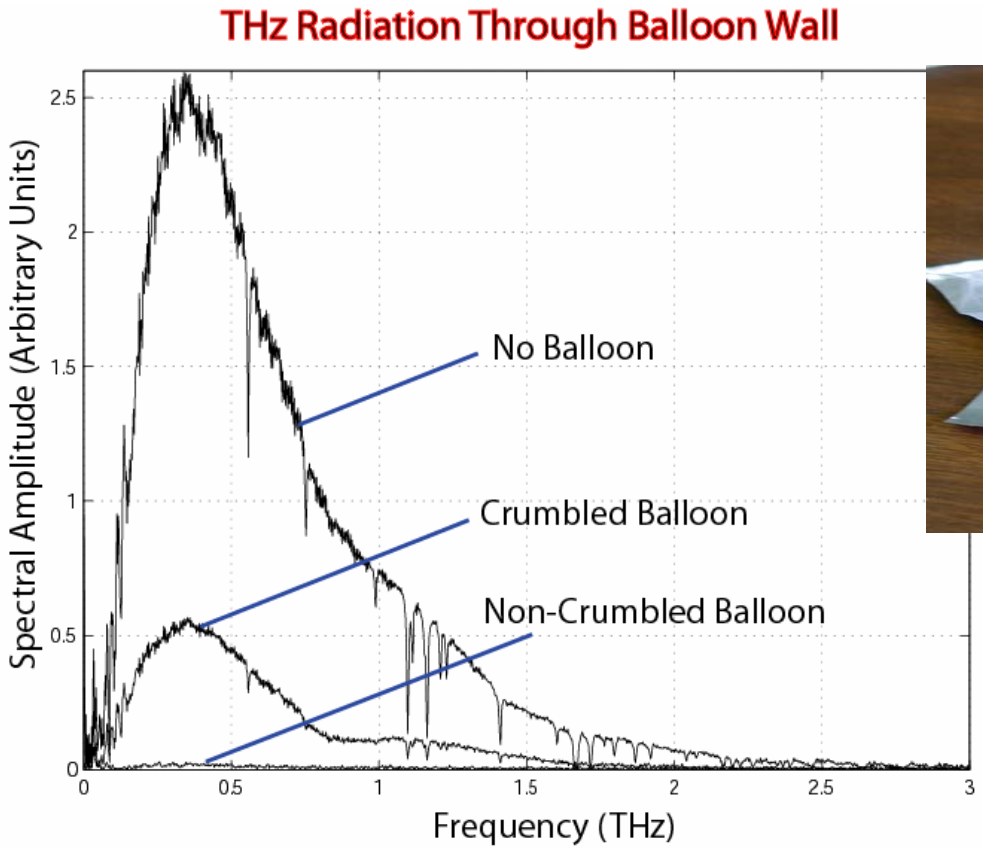
Ultrafast optical approach for generating pulses
in an underutilized region of the EM spectrum (“THz Gap”)

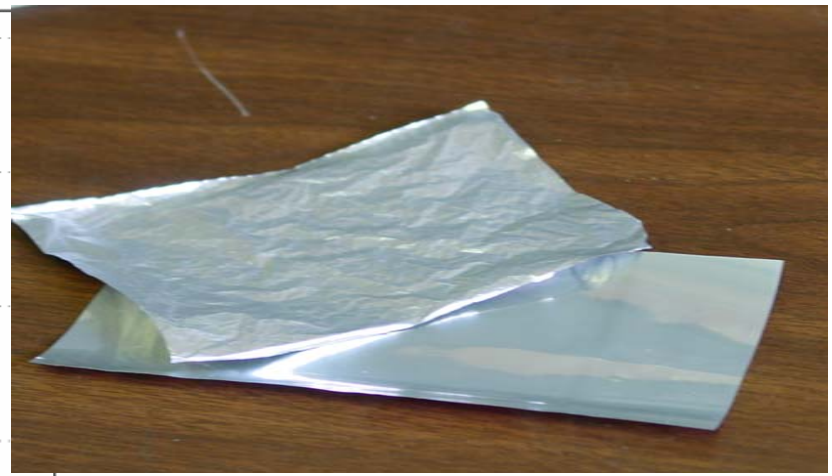


3 mm – 10 micron
100 GHz – 30 THz
3.33 cm^{-1} – 1000 cm^{-1}

- THz radiation (T-rays) consists of short pulse (< 1 ps), single-cycle, freely propagating FIR pulses generated via ultrafast optoelectronic techniques
- Directional, focusable, broadband (0.1 to 3 THz)
- The electric field (i.e. amplitude and phase) is directly obtained.
- Time-gated technique enables very high SNR.
- Easy access to the time and frequency response – both are useful in characterizing the THz response of materials.

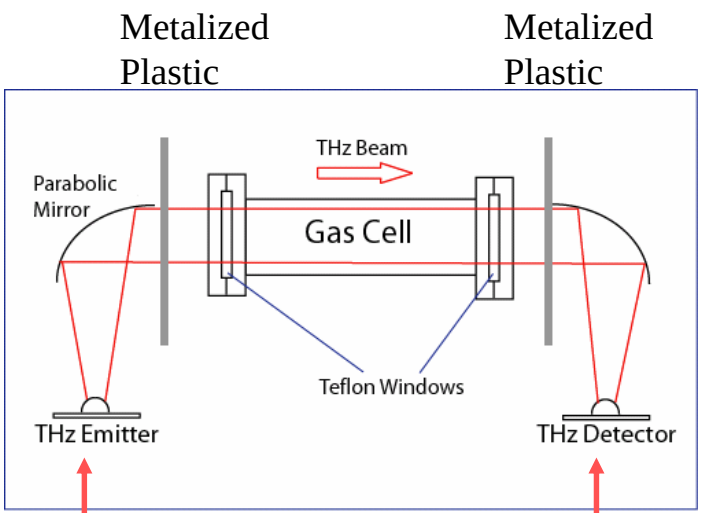
Metalized Plastics





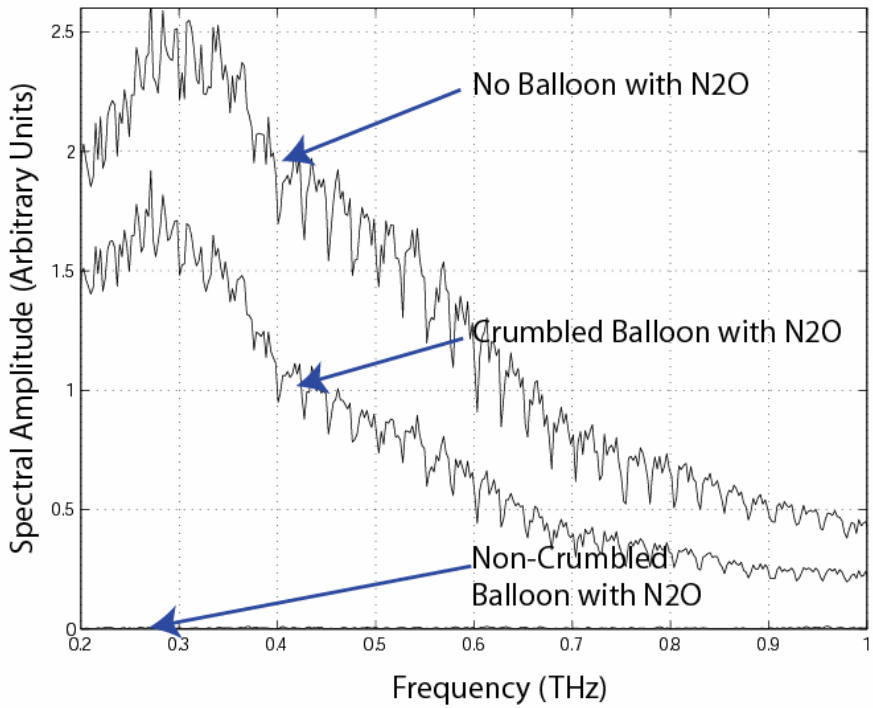
Transmission induced via disorder, not cracks in metal

N_2O absorption Spectra



Laser Excitation 1.5eV 100-fs Trigger Pulse

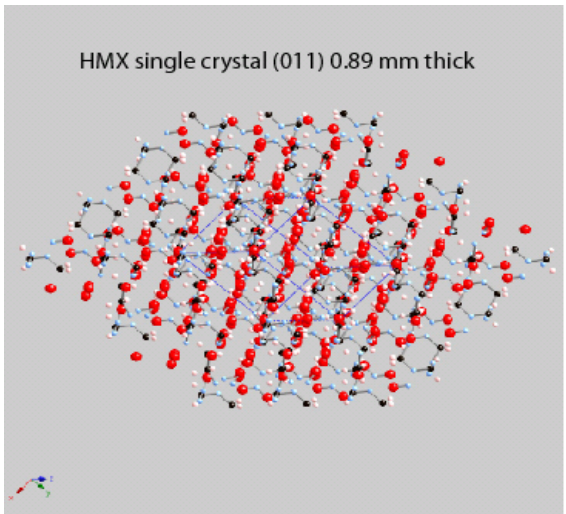
Laser Excitation 1.5eV 100-fs Gate Pulse

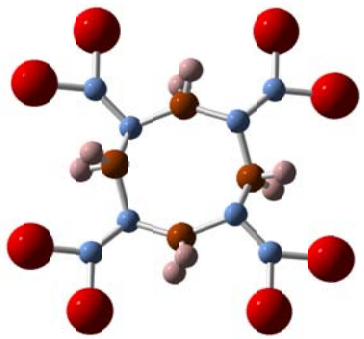


To simulate an inflated metalized plastic balloon, we inserted sheets of balloon material in the beam path on either side of the gas cell.

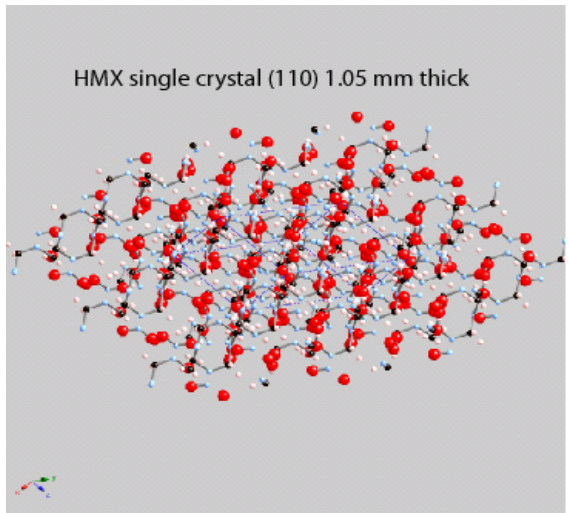
THz Spectroscopy of Explosives

Are there spectroscopic signatures of these molecules at THz frequencies?



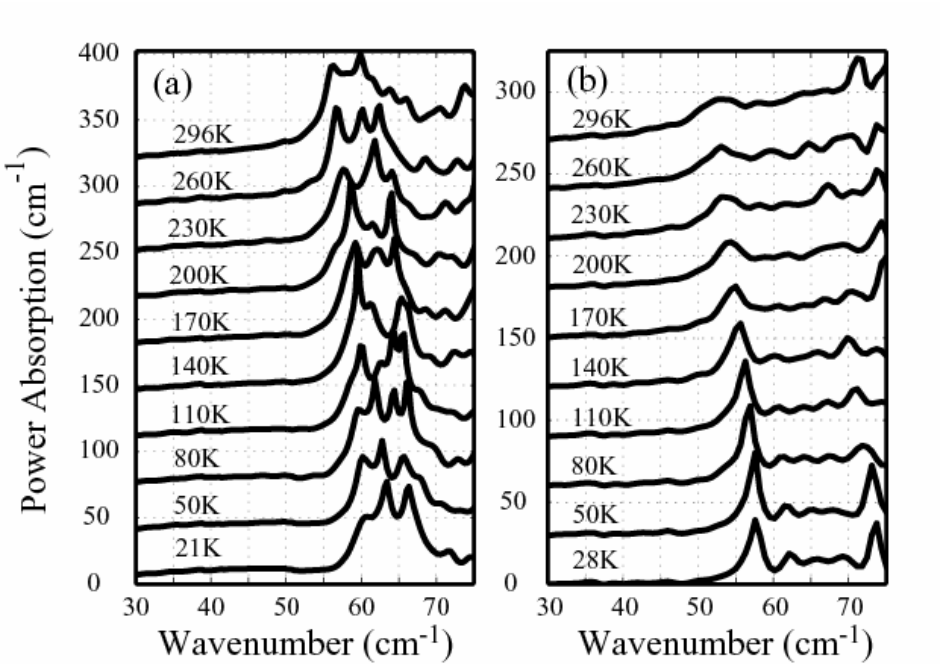


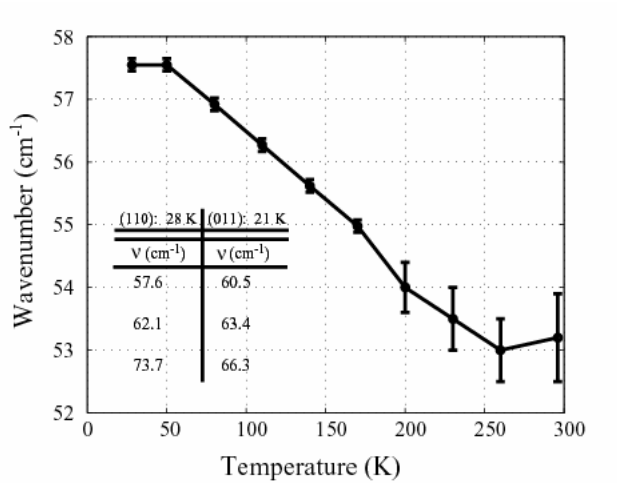
HMX



Done in collaboration with Dave Funk, Dan Hooks, and Jeff Barber, DX-2

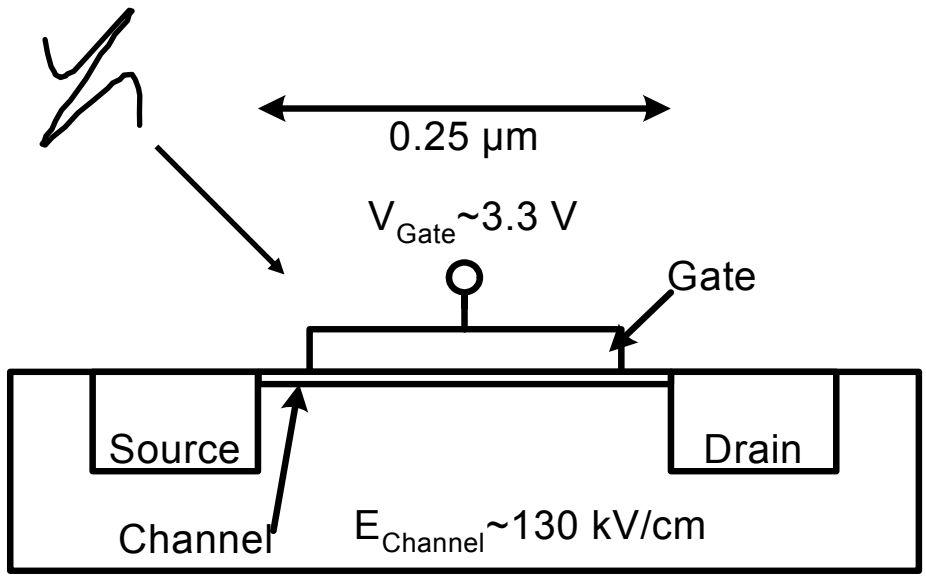
THz Spectroscopy of Explosives





Electrical Interactions

Introduced picosecond THz transient in working transistor to cause it to switch off.



Need E_{THz}~10^2 kV/cm *inside* channel to disrupt the transistor

Mode Locked Ti:Sapphire

- ~100,000 frequencies locked into phase coherently result in a fs pulse.
- Outputs 50 fs pulses at 80 MHz with a center wavelength of 800 nm (375 THz).
- Can be amplified using Ti:S based amplifier to produce 30 mJ/pulse.
- Higher pulse energies are possible.

THz emission via fs excitation

Mechanisms:

- Current Surge - FIR dipole radiation from acceleration of photo-injected carriers in a surface depletion field
- Pondermotive acceleration of electrons in a laser plasma
- Optical Rectification - Difference Frequency Mixing
  - Bulk electric-dipole: $\chi^{(2)}$
  - Bulk electric-quadrupole/magnetic dipole: $\chi^{(Q)}$
  - Field Induced (Surface) electric-dipole: $\chi^{(3)}$, E_d
  - Surface or bulk magnetization: $\chi^{(2)}(\boldsymbol{M})$
- Ultrafast demagnetization—FIR dipole radiation from rapid demagnetization following the creation of a nonthermal electron distribution with a fs optical pulse.

THz Generation Mechanism

Optical Rectification

$E_{THz} \sim \chi^{(2)} |E_{pump}|^2$

$$E_{THz} \propto \frac{d^2}{dt^2}\left[\chi^{(2)} E_{pump}{}^{*} E_{pump}\right]$$

Photoconductive Switch

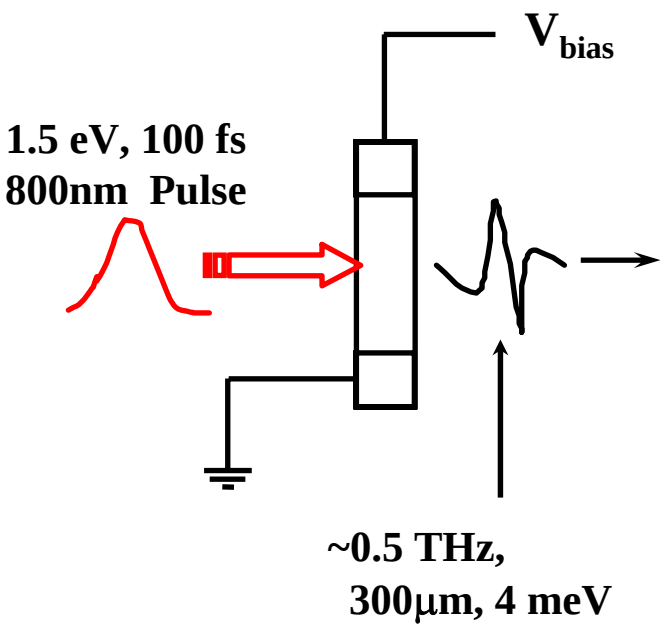
Pondermotive Acceleration

$$E_{THz} \propto \frac{dJ}{dt}$$

Ultrafast Demagnetization

$$E_{THz} \propto \frac{d^2 M}{dt^2}$$

Auston switches



- Photoconductive Emitters: Electrodes with a several kV/cm bias across a gap.
- A fast surface current transient is initiated by photo-injecting carriers with an ultrashort laser pulse. Ensuing THz radiation temporally tracks the time derivative of the total surface current.
- Peak output fields of ~ kV/cm
- Emitters: GaAs, LT-GaAs, InP
- Radiated THz field saturates with fluence, F: $E_{THz} \sim E_B F/F_o/(1 + F/F_o)$

THz detection

- Photoconductive receivers:
  - Based on the same principle of emitters.
  - THz E-field is used to bias a photo-gated detector, typically radiation damaged silicon-on-sapphire (SOS) or LT-GaAs.
  - Detector response time of SOS, $\tau_r < 0.5$ ps.
- Electro-optic sampling
  - Based on detection of polarization rotation Pockels effect in a $\chi^{(3)}$ material (ZnTe).
  - E-field from THz beam is used to rotate the polarization of an optical gate beam via electrooptic effect.
  - Detection bandwidth is limited by the group velocity mismatch between THz beam and optical beam.

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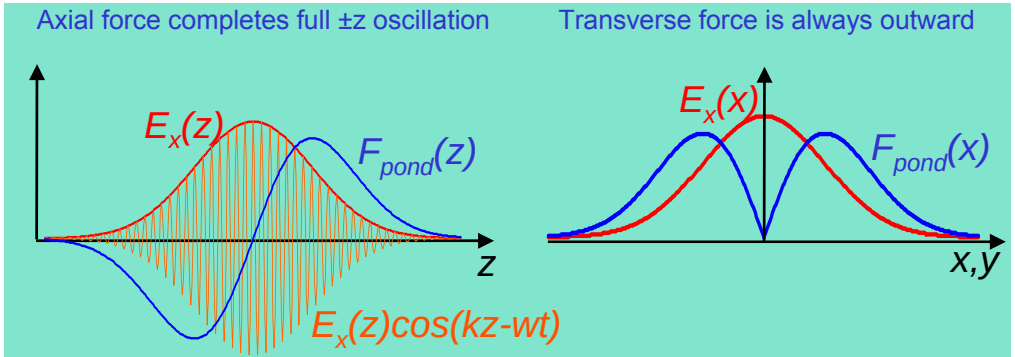
Ponderomotive Acceleration of electrons

- **PULSE code self-consistently propagates a pulse through ionizable media:**



- **THz radiation frequency linked to dominant laser gradient**

- **Laser E_x and $B_{y,z}$ fields accelerate free electrons:**

Force from laser E_x-field gradient in x-direction (seen by e^- motion in x)

Transverse force from $v_x xB_z$

Force from $v_x xB_y$ in axial direction

$$\vec{F}_{pond} \simeq \frac{-10^7 \pi e^2}{mc\omega^2} \vec{\nabla} I_{laser} \left(\mathrm{W/cm^2}\right) = \frac{-e^2}{4m\omega^2} \left[\hat{x} E_x \frac{dE_x}{dx} + \hat{y} E_x \frac{dE_x}{dy} + \hat{z} E_x \frac{dE_x}{dz} \right]$$

- **Radiated field from ponderomotively accelerated electrons:**

$$\vec{E}_{THz} \simeq \frac{e}{c} \left[\frac{\vec{n} \times (\vec{n} \times \dot{\vec{\beta}}_{pond})}{r} \right]_{ret} = \frac{e}{c} \left\{ \frac{\vec{n} \times \left[\vec{n} \times \left(\vec{F}_{pond} / m \right) \right]}{r} \right\}_{ret}$$

THz emission via fs excitation

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- Ultrafast demagnetization—FIR dipole radiation from rapid demagnetization following the creation of a nonthermal electron distribution with a fs optical pulse.

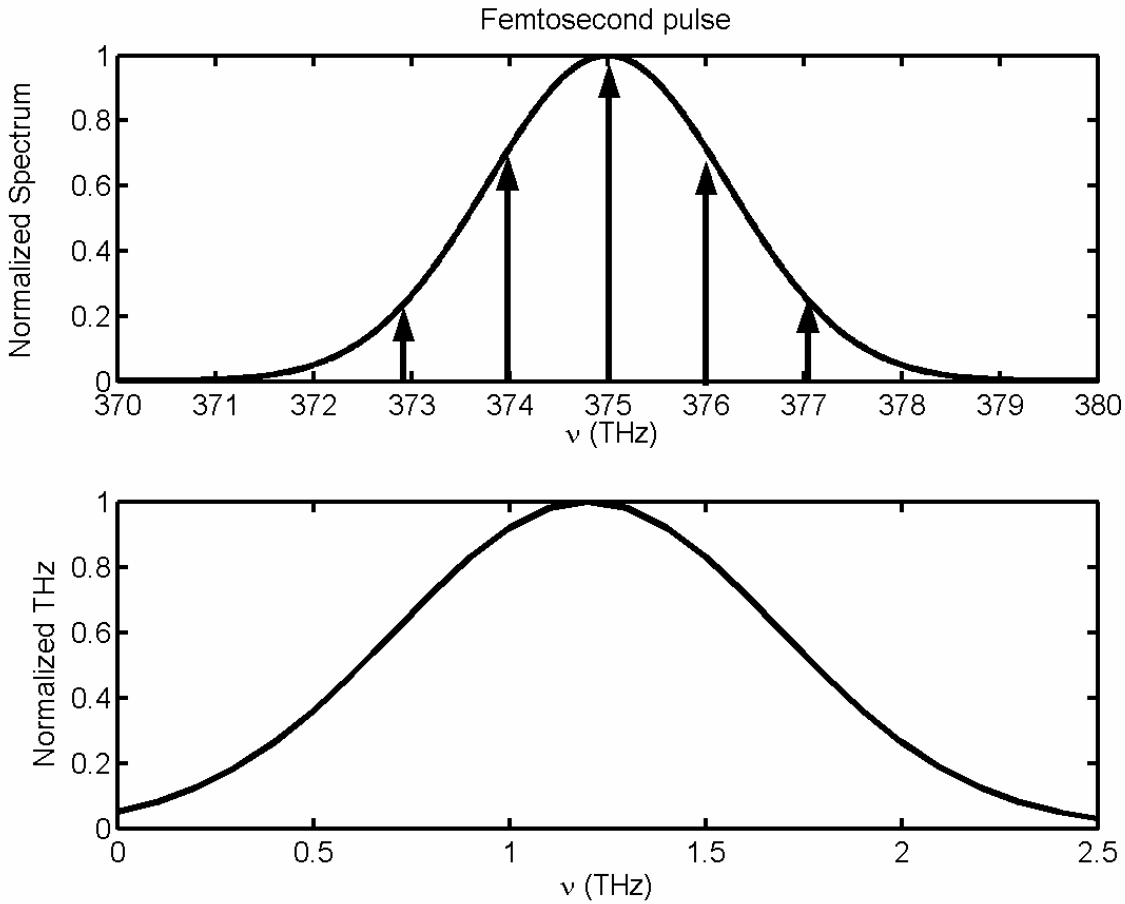
Difference Frequency Generation

- Uses a special class of materials with strong nonlinearities, $\chi^{(2)}$, which result in transfer of energy from one frequency to another by generating a far IR dipole in material.

$$E(\nu_3) = \frac{\partial^2}{\partial t^2}\left[\chi^{(2)} E(\nu_1) E^*(\nu_2)\right]$$

- Common materials: ZnTe, $LiNbO_3$, BBO, KTP, KDP, AgGaSe, AgGaS, *GaSe*, etc.

Optical Rectification

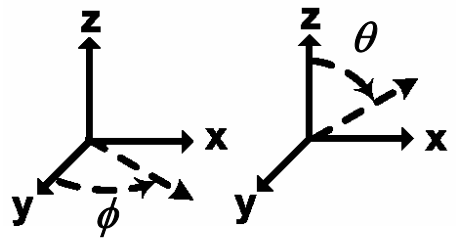


Special case of DFG using the bandwidth of the Ti:S pulse to generate the THz pulse.

THz bandwidth is limited by Ti:S pulse bandwidth.

THz generation via optical rectification

$$E_i^{rad}(\Omega) \propto \frac{\partial^2 P}{\partial t^2} = \frac{\partial^2}{\partial t^2}\chi_{ijk}^{(2)} E_j(\omega_1) E_k^*(\omega_2)$$



Azimuth Incidence

- **Bulk**: electric dipole

  $E^{THz} \sim \cos 2(\phi - \phi_o)$

  electric quadrupole/magnetic dipole $\chi^{(2)}$:

  $E^{THz} \sim \cos 4(\phi - \phi_o)\sin\theta$

- **Surface**: electric dipole nonlinearity:

  E^{THz}(p,p) and E^{THz}(s,p) ~ $\sin\theta$

  E^{THz}(p,s) and E^{THz}(s,s) ~ 0

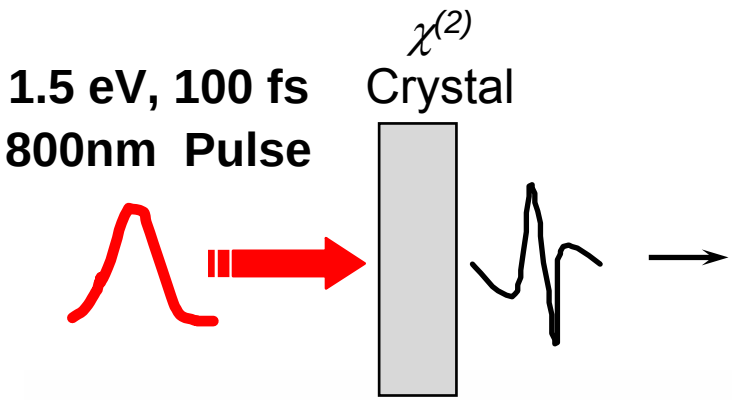
- **Magnetic nonlinearity**, $\chi^{(2)}(\boldsymbol{M})$:

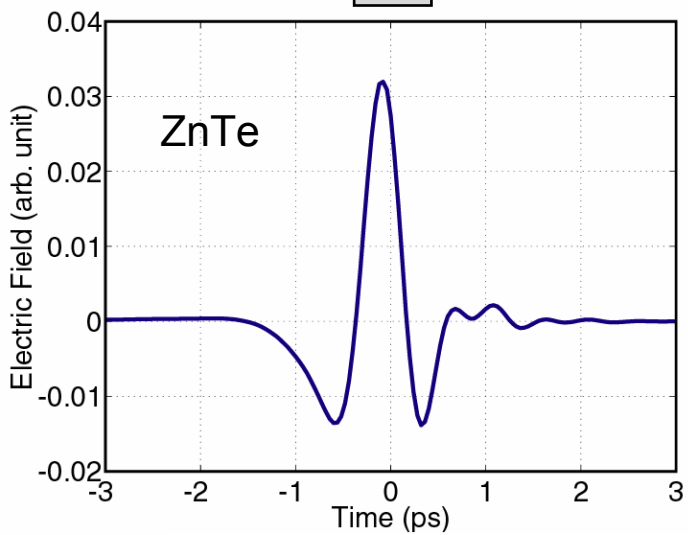
  $E^{THz} \sim \cos(\phi - \phi_o) + A\cos 3(\phi - \phi_1)$

  $|\omega^2\chi^{(2)}(\boldsymbol{M})| \sim 10^{-12}$ esu ref: *Phys. Rev.* B 48, 8607 (1993).

All of these are "instantaneous" processes and do not limit the emission bandwidth.

THz generation mechanism: Optical rectification

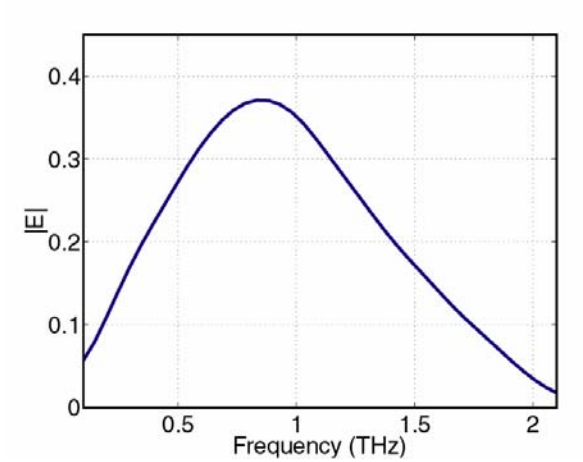




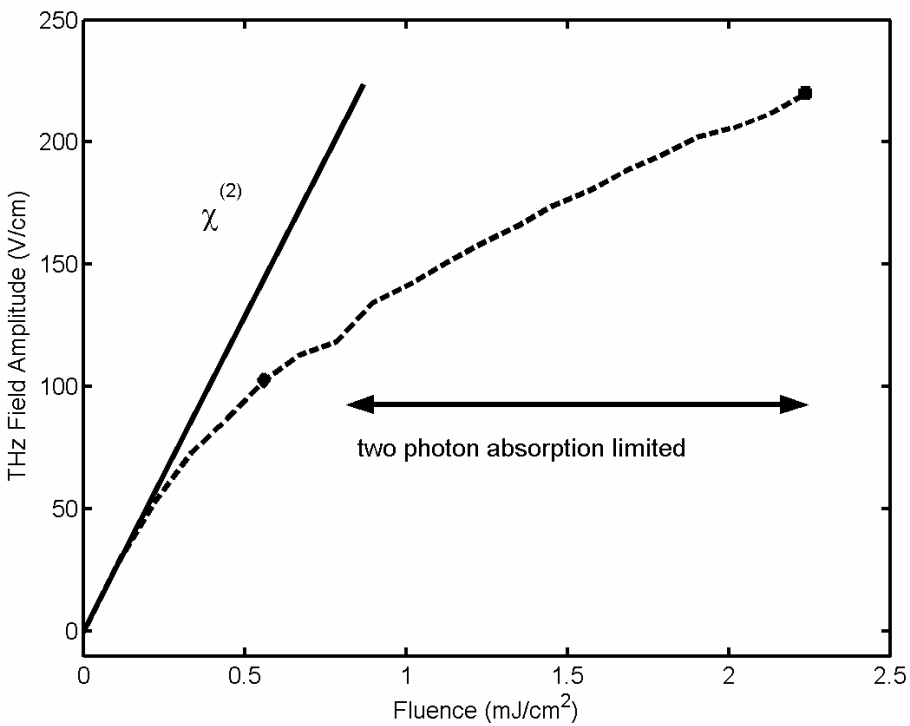
- Optical Rectification: Characterized by nonlinear optical difference frequency mixing :

$$E_i^{rad}(\Omega) \propto \frac{\partial^2 P}{\partial t^2} = \frac{\partial^2}{\partial t^2}\chi_{ijk}^{(2)} E_j(\omega_1) E_k^*(\omega_2)$$

- Peak field outputs of 10 - 100 V/cm
- Emitters: GaAs, InP, DAST, ZnTe, $LiNbO_3$, $LiTaO_3$, *GaSe*



Higher field emission from ZnTe



- Other nonlinear processes compete for the same pump photons.
- Instead of scaling the pump power, scale the area.
- Higher field source from ZnTe: too expensive! 1 cm x 1 cm x 1 mm ZnTe is about $3K.
- Either need new materials for higher fluences, or rely on photoconductive emitters.

THz emission via fs excitation

Mechanisms:

- Current Surge - FIR dipole radiation from acceleration of photo-injected carriers in a surface depletion field
- Pondermotive acceleration of electrons in a laser plasma
- Optical Rectification - Difference Frequency Mixing
  - Bulk electric-dipole: $\chi^{(2)}$
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  - Surface or bulk magnetization: $\chi^{(2)}(\boldsymbol{M})$
- Ultrafast demagnetization—FIR dipole radiation from rapid demagnetization following the creation of a nonthermal electron distribution with a fs optical pulse.

Ultrafast Demagentization

VOLUME 76, NUMBER 22 PHYSICAL REVIEW LETTERS 27 MAY 1996

Ultrafast Spin Dynamics in Ferromagnetic Nickel

E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot
Institut de Physique et Chimie des Matériaux de Strasbourg, Unité Mixte 380046 CNRS-ULP-EHICS, 23, rue du Loess, 67037 Strasbourg Cedex, France
(Received 17 October 1995)

The relaxation processes of electrons and spins systems following the absorption of femtosecond optical pulses in ferromagnetic nickel have been studied using optical and magneto-optical pump-probe techniques. The magnetization of the film drops rapidly during the first picosecond, but different electron and spin dynamics are observed for delays in the range 0–5 ps. The experimental results are adequately described by a model including three interacting reservoirs (electron, spin, and lattice). [S0031-9007(96)00167-6]

PACS numbers: 75.40.Gb, 75.70.–i, 78.20.Ls, 78.47.+p

First to report ultrafast changes to spins in a “basic” metal ferromagnet (~2 ps) . Before this, the fastest changes to *M* were thought to be hundreds of picoseconds to nanoseconds

Ref: E. Beaurepaire, J.-C. Merle, A. Daunois, and J. -Y. Bigot. Phys. Rev. Lett. **76**, 4250 (1996).

Ultrafast Demagnetization

VOLUME 85, NUMBER 4 PHYSICAL REVIEW LETTERS 24 JULY 2000

Ultrafast Magneto-Optics in Nickel: Magnetism or Optics?

B. Koopmans,* M. van Kampen, J. T. Kohlhepp, and W. J. M. de Jonge
Eindhoven University of Technology, Department of Applied Physics, COBRA Research Institute, P.O. Box 513, 5600 MB, Eindhoven, The Netherlands
(Received 22 February 2000)

Several magnetic and optical processes contribute to the magneto-optical response of nickel thin films after excitation by a femtosecond laser pulse. We achieved a first complete identification by explicitly measuring the time-resolved Kerr *ellipticity* and *rotation*, as well as its temperature and magnetic field dependence in epitaxially grown (111) and (001) oriented Cu/Ni/Cu wedges. The first hundreds of femtoseconds the response is dominated by state filling effects. The true demagnetization takes approximately 0.5–1 ps. At the longer (sub-ns) time scales the spins are found to precess in their anisotropy field. Simple and transparent models are introduced to substantiate our interpretation.

PACS numbers: 75.40.Gb, 75.70.–i, 76.50.+g, 78.47.+p

At least part of “ultrafast demagnetization” signal is due to state filling, not magnetization changes.

How fast are the magnetization changes in the sample?

Ref: B. Koopmans, M. van Kampen, J. T. Kohlhepp, W. J. M de Jonge, Phys. Rev. Lett. **85**, 844 (2000).

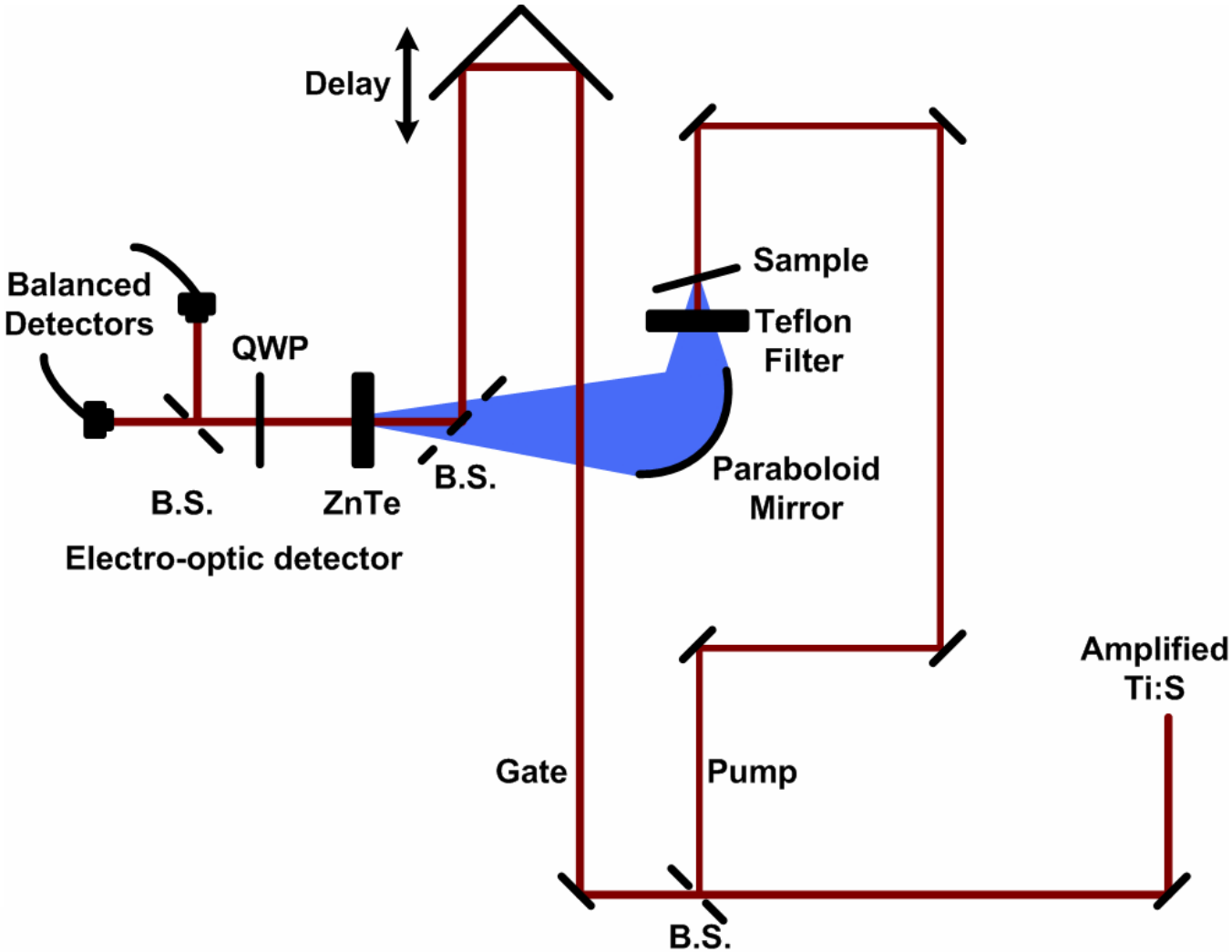
Terahertz Emission Spectroscopy

- Induce a time changing magnetic field (or current surge), which results in emission of a electromagnetic field (THz pulse).

$$E_{THz} \propto \frac{dJ}{dt} \quad \text{or} \quad E_{THz} \propto \frac{d^2M}{dt^2}$$

- Dynamics of magnetization changes (or current surge) limit the bandwidth of the THz pulse.
- No Kerr/Faraday rotation needed.
- Is this instead limited by induced changes to the material conductivity/transmission at the frequencies of emission?

THz emission from Fe/MgO

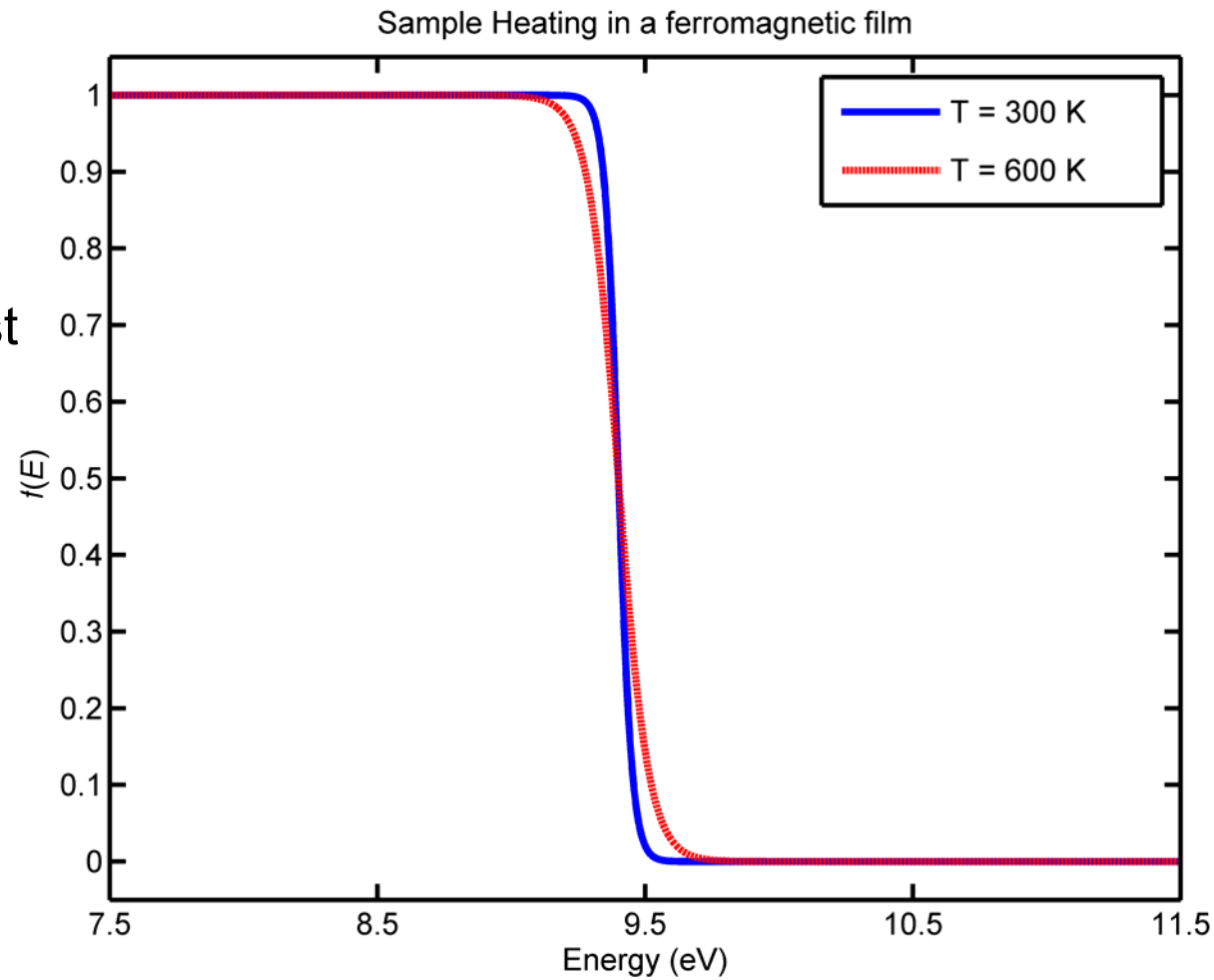


Sample heating

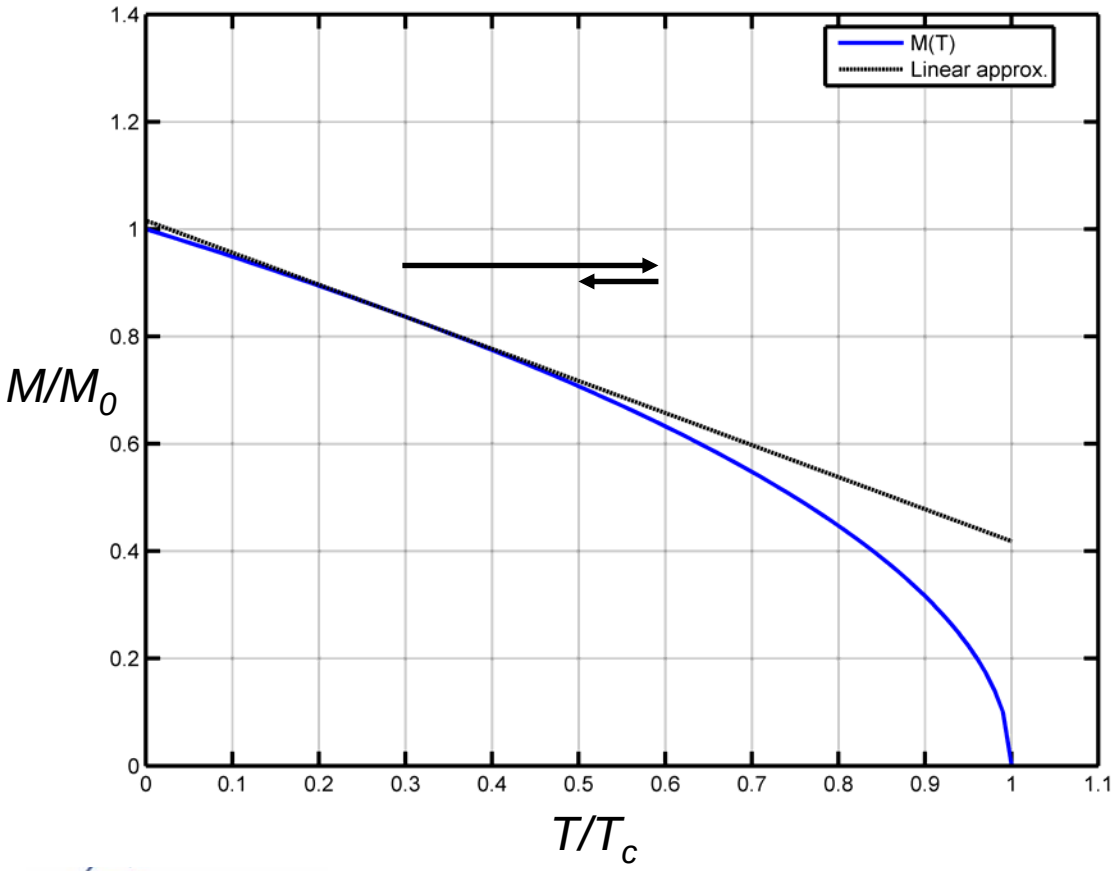
No resonant transitions available at 1.55 eV.

The femtosecond pulse is essentially an ultrafast heat source in the ferromagnetic film.

Electron thermalization time is typically <100 fs for metals



Temperature Dependent Magnetization



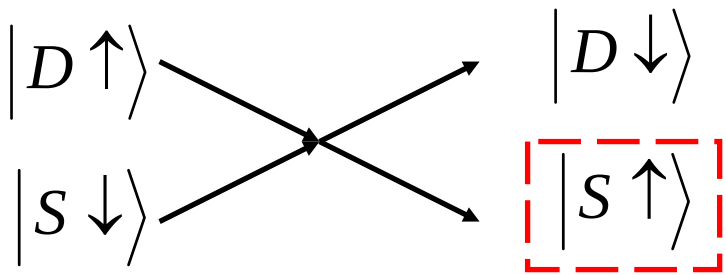
- Initial heating of sample, followed by relaxation to interim temperature.
- Magnetization does not necessarily "instantaneously" follow T_E.
- Can the emission help determine the time scale of the magnetization changes?

Stoner Model of Ferromagnetism

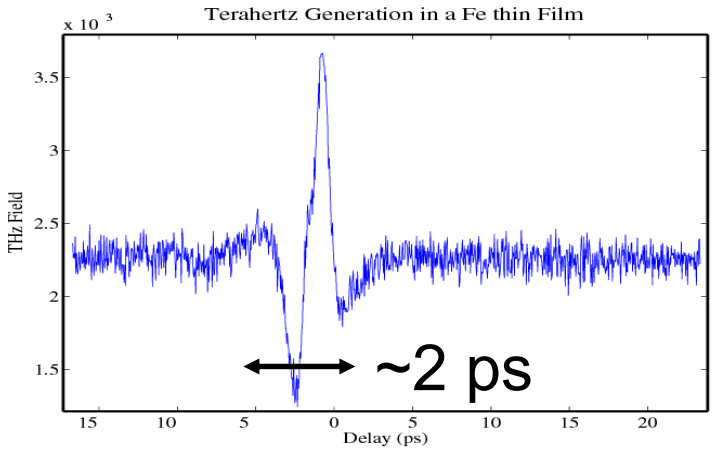
- Ferromagnetism in “simple” metals (Ni, Fe, Co, Gd) is characterized by a spin split *d-like* subband, resulting in more d electrons of the majority spin than the minority spin
- An increase in electron temperature results in an increase in the spin dependent scattering rate (spin-flip scattering).
- The spin population in the *s-like* subband rapidly looses it’s spin polarization due to the (lack of) spin orbit coupling.
- Spin is *consumed* by this process, leading to an overall reduction in the number of spin polarized d-electrons. Total spin is not conserved

$$N = n_{d\uparrow} - n_{d\downarrow}$$

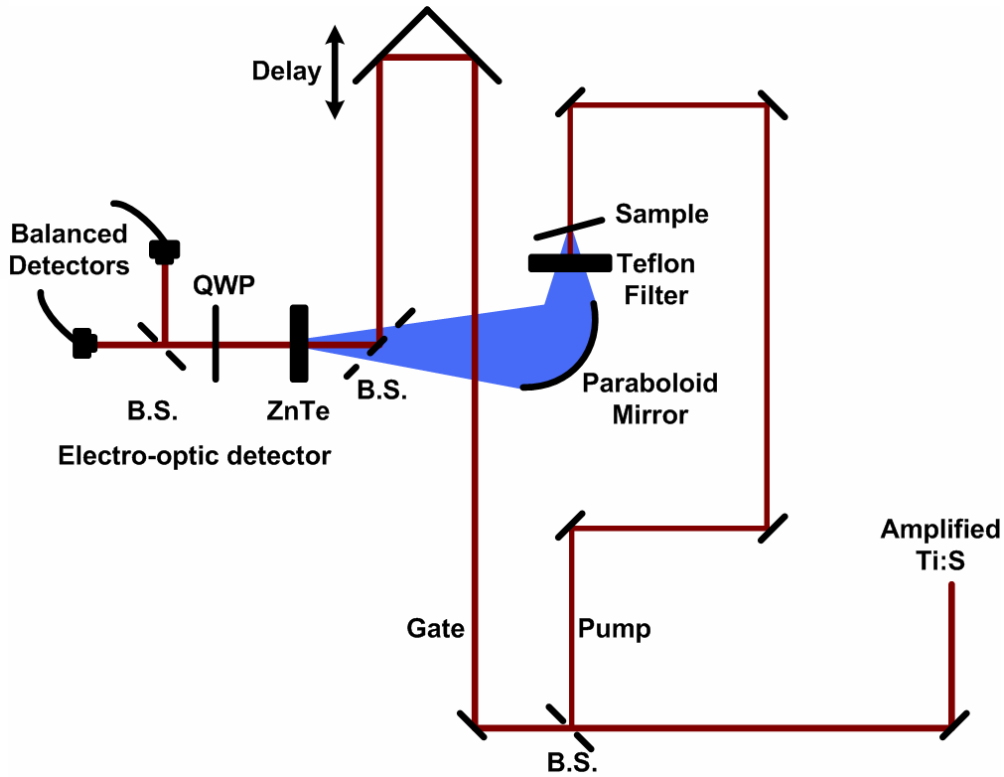
Spin Flip Scattering



THz emission from Fe/MgO



- Measured THz generated after transmission through iron film
- EO detection to measure field directly
- Can determine the amplitude and phase of THz emission
- Calculate spectrum from field

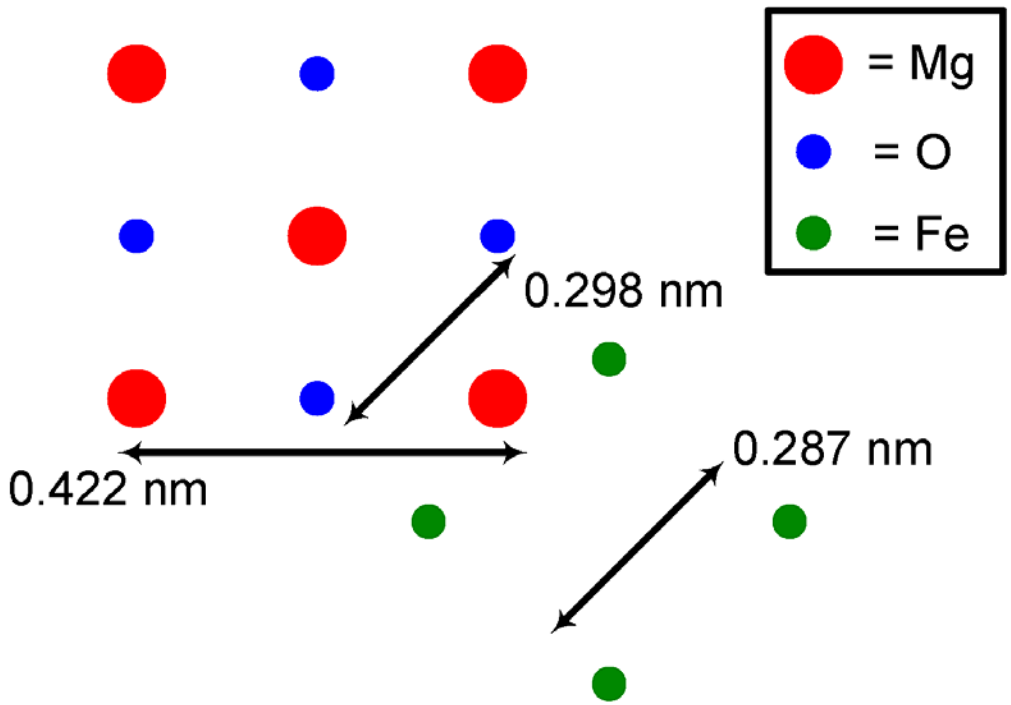


Hilton *et al.*, Opt. Lett. **29**, 1805 (2004)

MBE growth of Fe on MgO

Adapted from A. di Bona, *et al*. *Surf. Sci.* **498** (2002) 193-201.

- Lattice constant MgO is ~$\sqrt{2}$ x lattice constant Fe
- Fe is rotated by 45°
- ~4% mismatch
  - $d_{O\text{-}O}$ = 2.98Å
- Epitaxial Relationship:
  - Fe(001) || MgO(001)
  - Fe<100> || MgO<110>
  - Fe<110> || MgO<010>



MgO(001) → Fe(001) with a bcc crystal structure
~12 nm thick

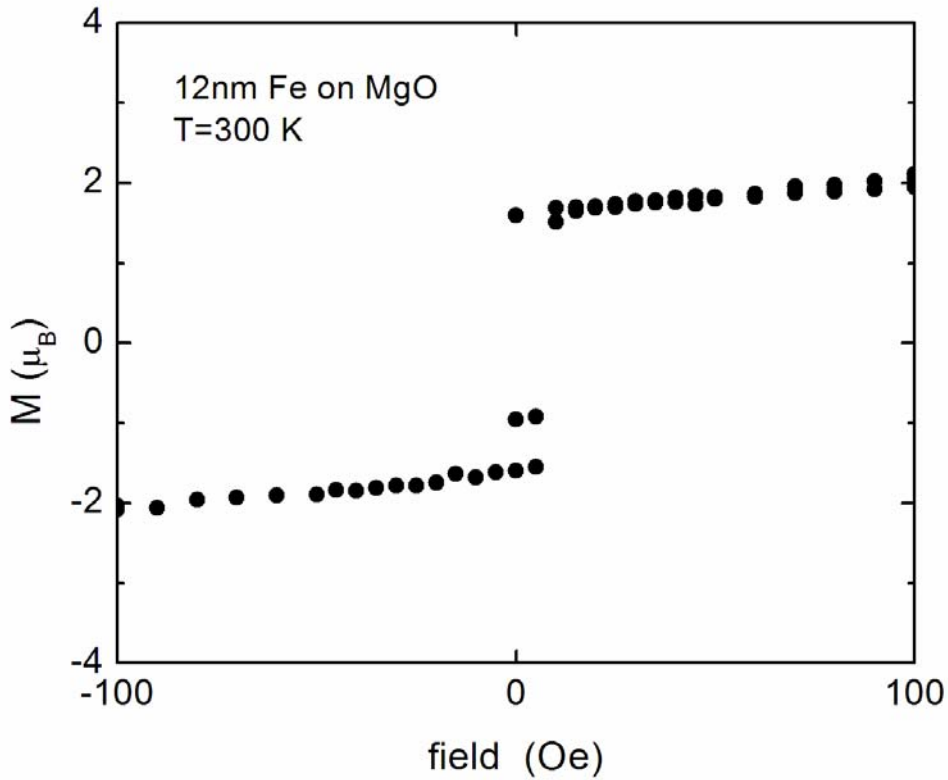
Magnetization of Fe film

Measured in a SQUID Magnetometer

Remanence =0.95 μ_B/atom

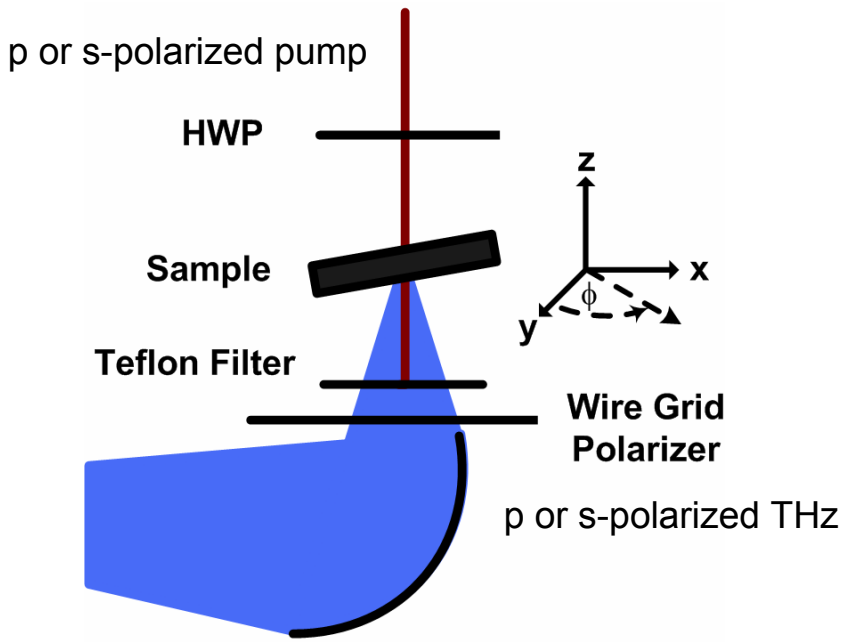
Saturation = 3.7 μ_B/atom

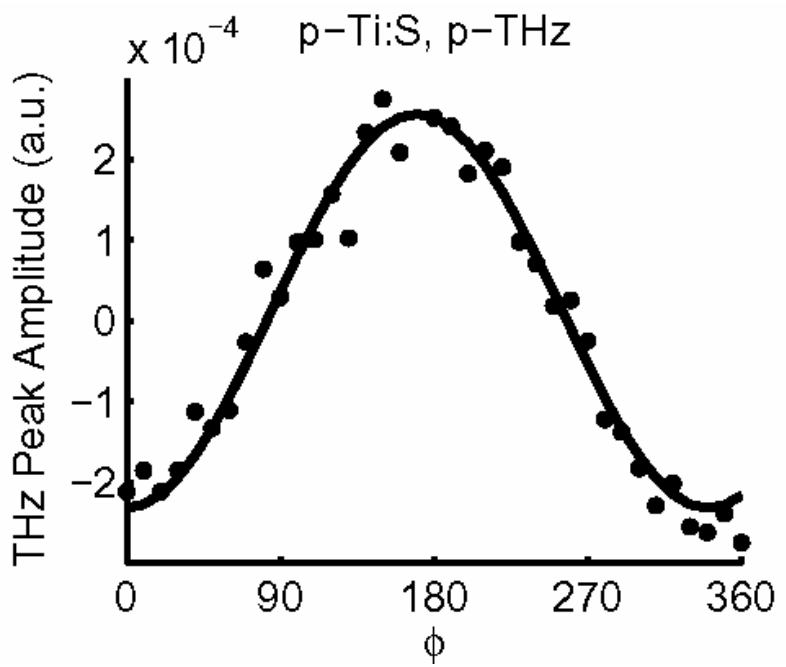
This as-grown film is multi-domain, but dominated by one magnetic direction over Ti:S excitation



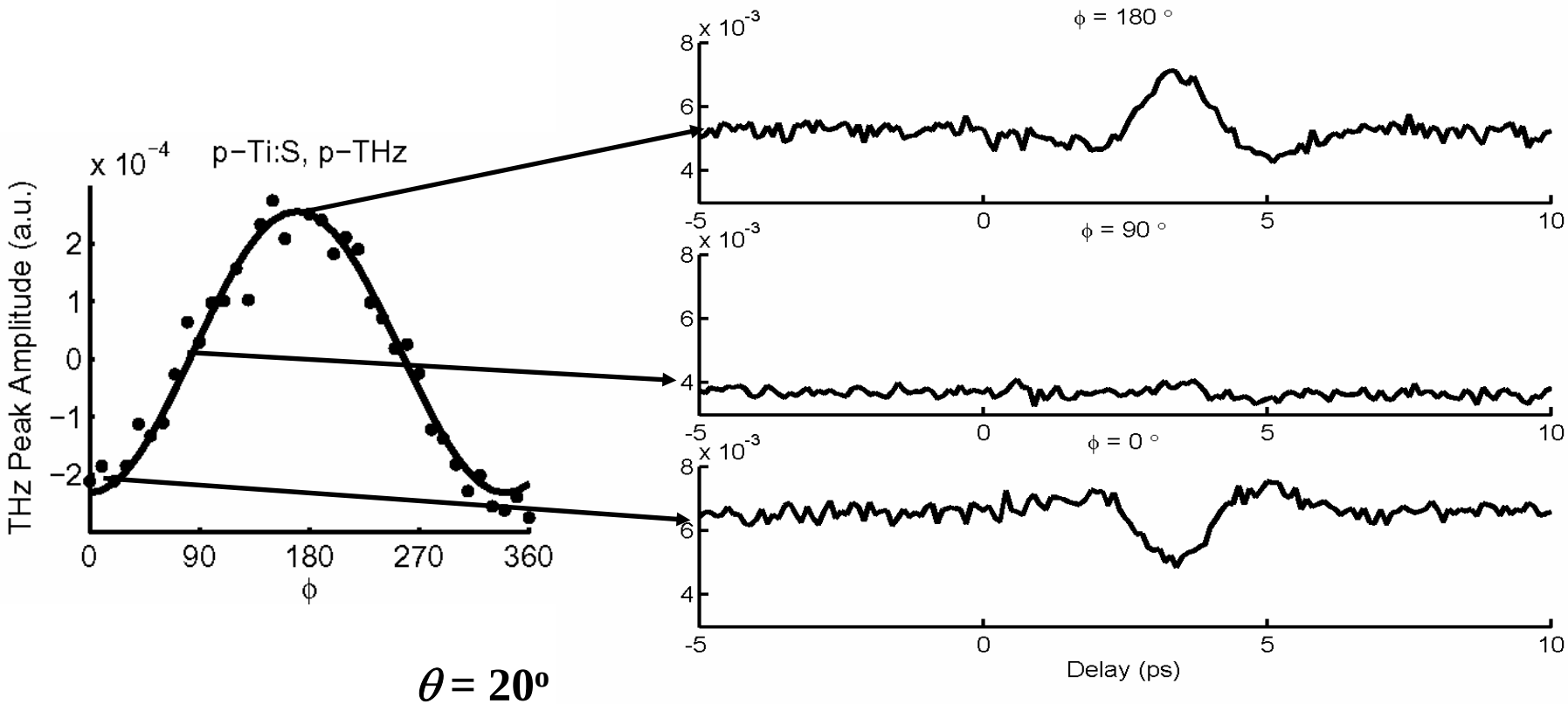
Azimuthal Dependence

Optical Rectification [$\chi^{(2)}$], ultrafast demagnetization, and Auston switches have known dependences on the angle between the pump field and the crystal/magnetization axes (ϕ).



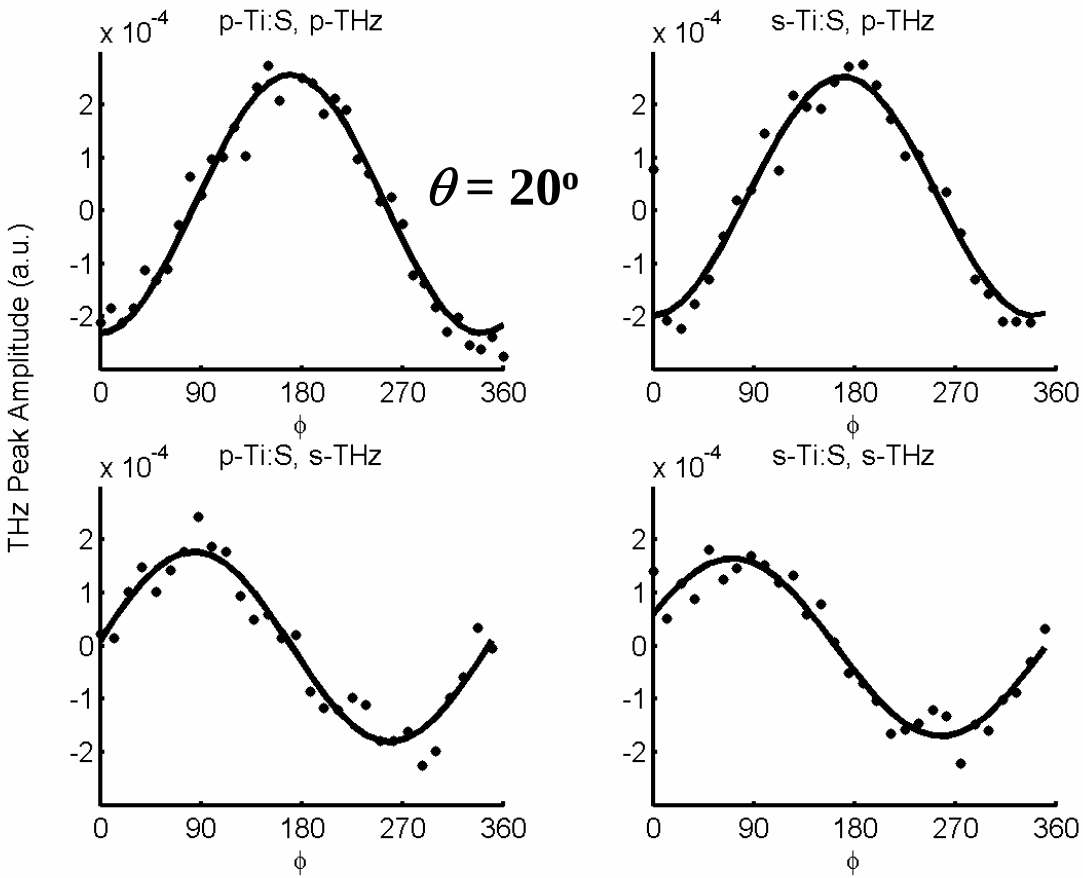


Azimuthal dependence (cont.)



$\theta = 20^{o}$

Azimuthal dependence (cont.)



$\chi^{(2)}_s$ $\chi^{(2)}(M)$

$E(p,p)=2.44\text{x}10^{-4}[0.13 - \cos(\phi + 0.7^o)]$

$E(s,p)=2.26\text{x}10^{-4}[0.12 - \cos(\phi + 0.5^o)]$

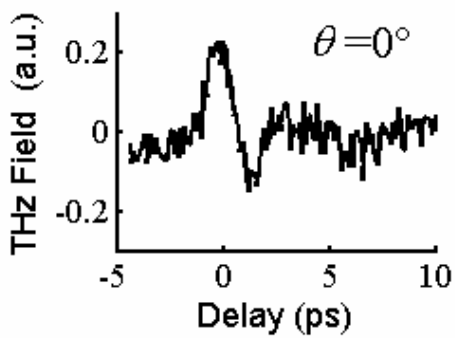
$E(p,s)=1.79\text{x}10^{-4}[-0.01 + \sin(\phi + 3.9^o)]$

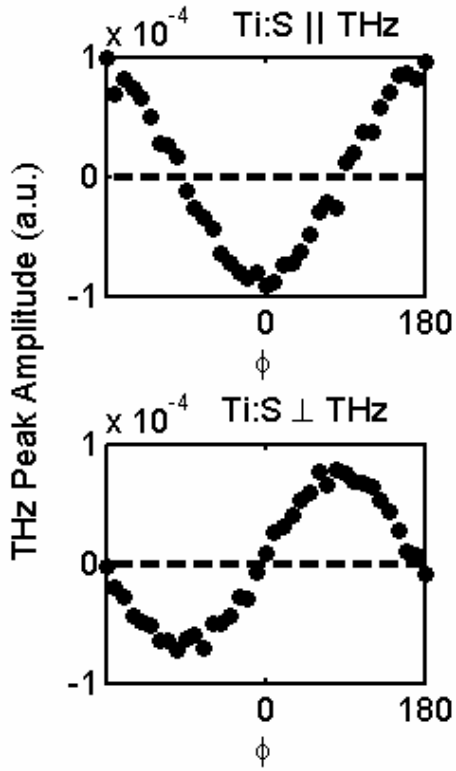
$E(s,s)=1.68\text{x}10^{-4}[-0.01 + \sin(\phi + 21.5^o)]$

The THz emission remains at normal incidence but the constant offset disappears,consistent with this offset resulting from a surface nonlinearity.

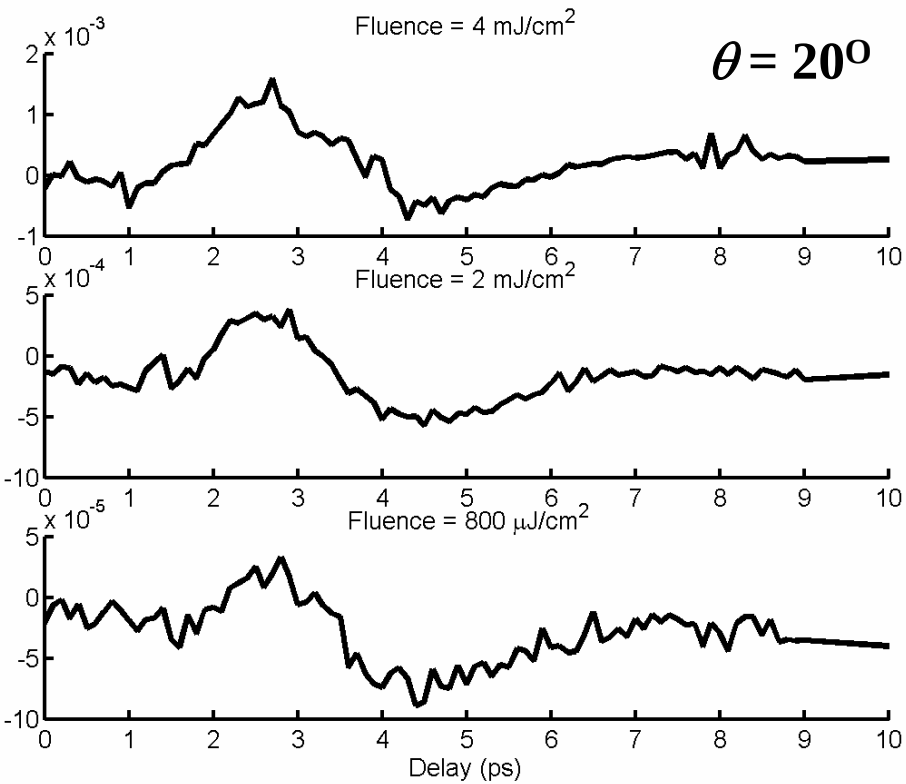
Surface Nonlinearity

- Azimuthally independent term.
- Also reported in amorphous gold and silver films. (Kadlec *et al.*, to be published in Optics Letters)



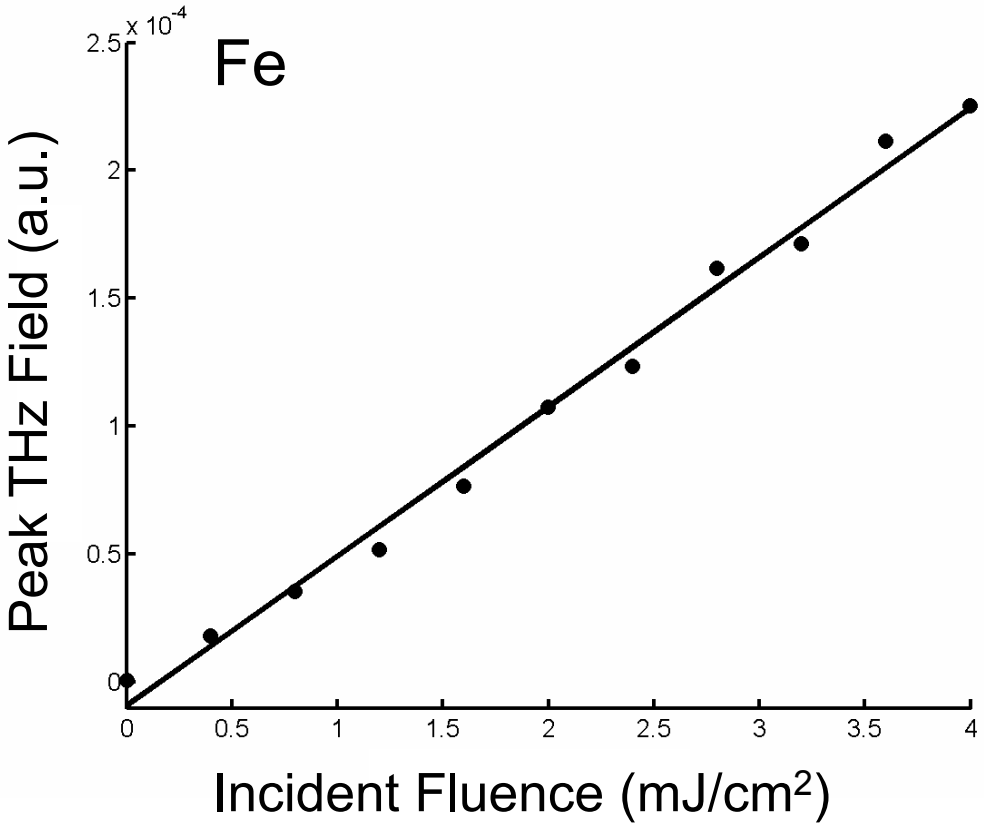


Fluence dependence of THz bandwidth



The bandwidth of the emission is constant at each fluence, ruling out any induced absorption mechanism to explain narrow bandwidth

THz field scaling- DFG



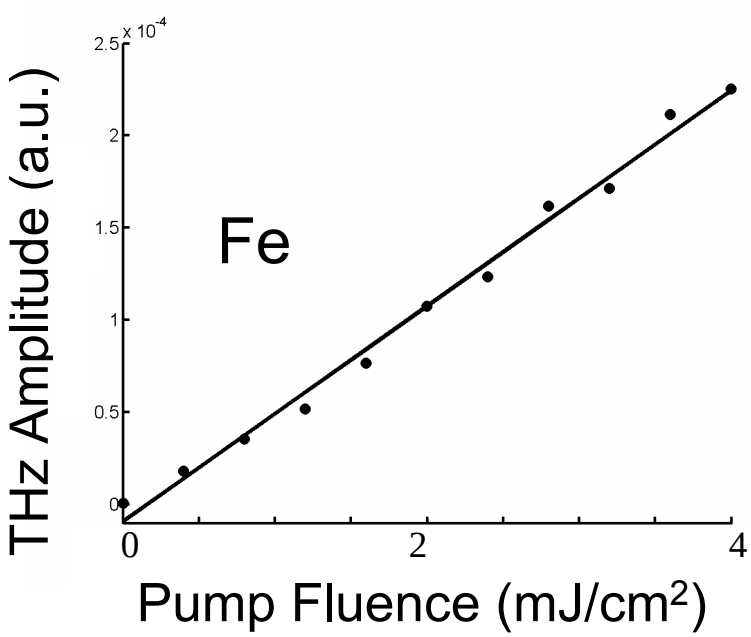
Advantages

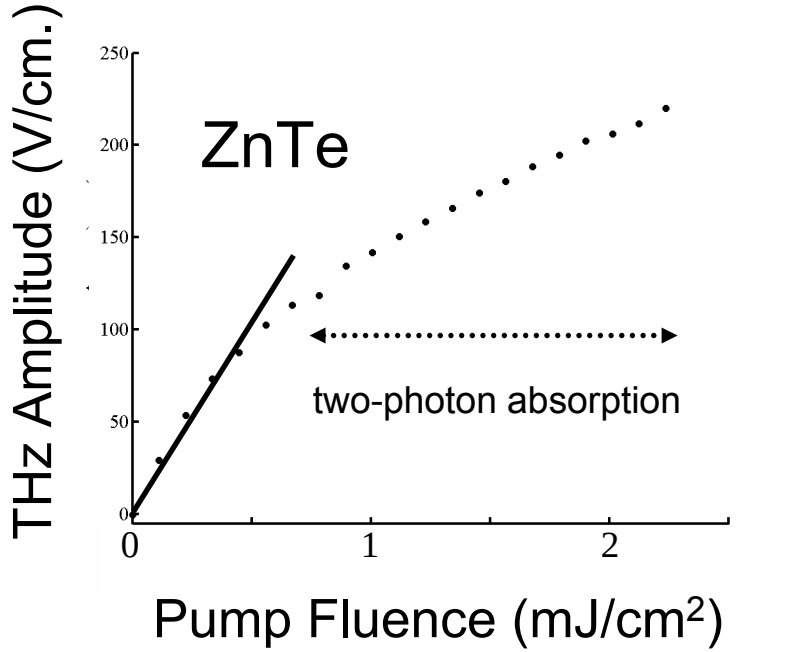
- cost
- remote THz generation

Disadvantages

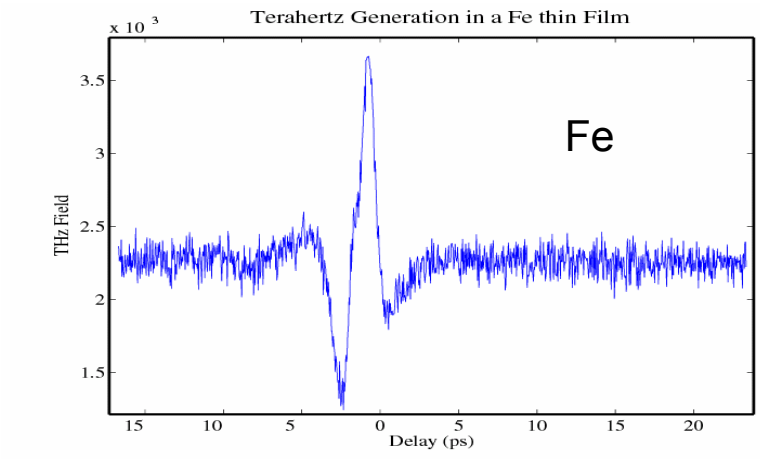
- lower conversion efficiency
- larger area
- limited bandwidth?

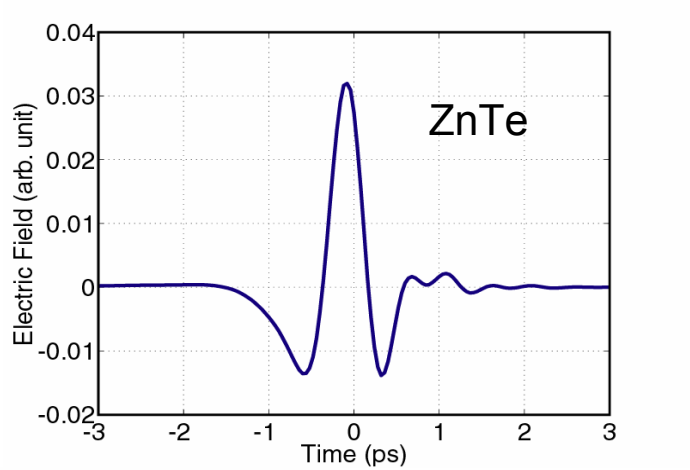
Fluence dependence of THz amplitude



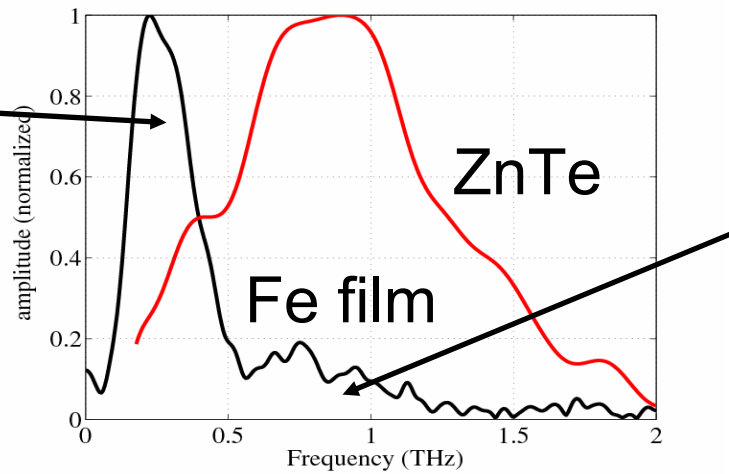


THz emission at θ=20°





Ultrafast Demagnetization



Surface nonlinearity

Second ΔM lifetime

???

Drude Conductivity

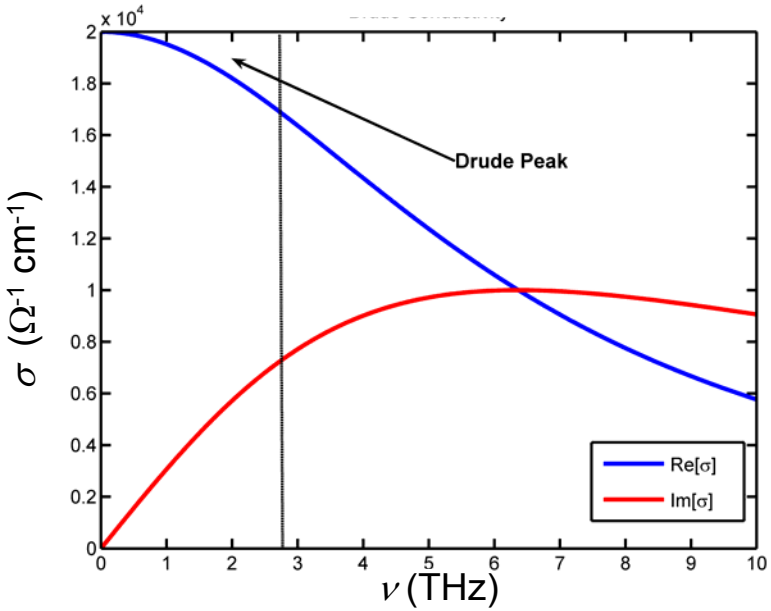
Electrons are “free” to move in metal with an effective mass, *m**.

$$\sigma(\omega) = \frac{\sigma_0}{1 - i\omega\tau} = \frac{\sigma_0}{1 - \omega^2\tau^2}(1 + i\omega\tau)$$

τ = transport scattering time

σ_0 = dc conductivity = ne^2/m^*

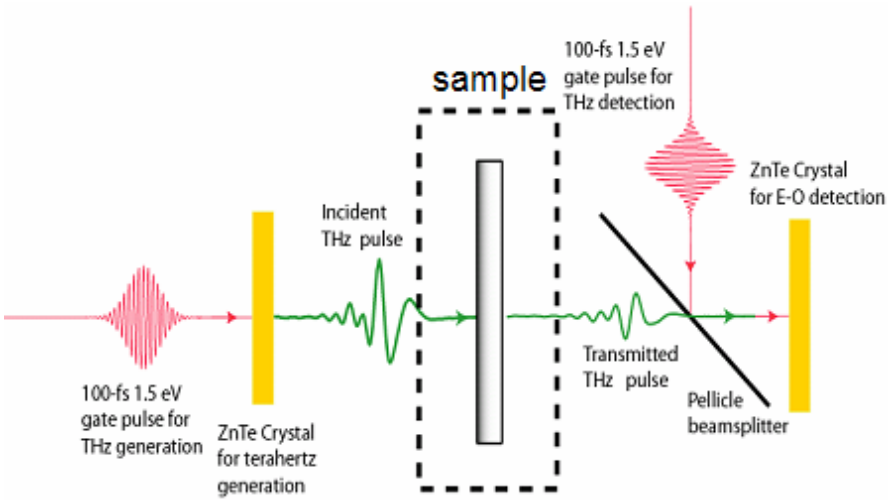
~ carrier density, n

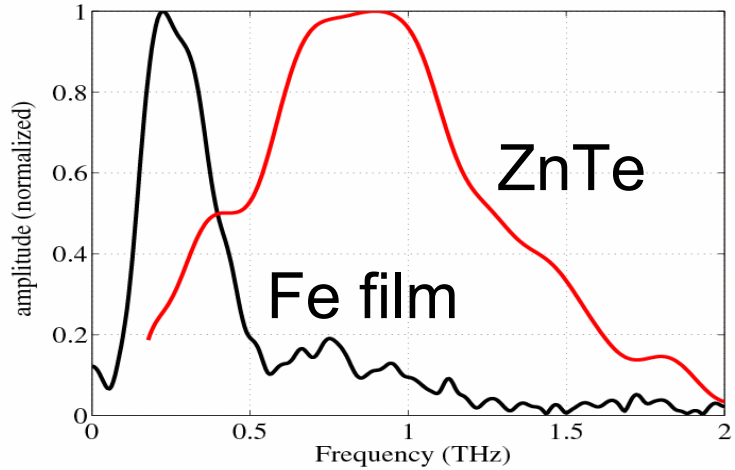


$\Delta\tau \rightarrow$ phase shift

$\Delta n \rightarrow$ amplitude

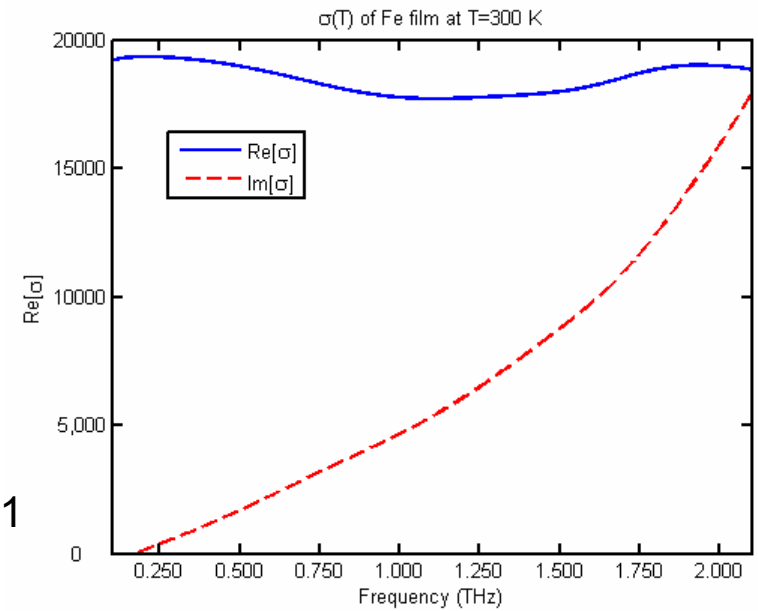
Optical Conductivity of Fe





Absorption in iron does *not* significantly narrow the bandwidth of the emission, but does result in a shift in phase of the emitted pulse.

$\tau = 70$ fs, $\sigma_o = 20{,}000\ (\Omega\text{–cm})^{-1}$



Ultrafast Demagnetization

- Ultrashort pulse excites the Fe sample, first creating a non-thermal electron distribution, and then a thermal electron distribution, but at an elevated temperature.
- An increased T_e-dependent spin scattering rate results in fast (~2 ps) reduction in the magnetization.
- The rapid demagnetization results in emission of a THz pulse, with a 1/(~2 ps) ~ 500 GHz bandwidth.

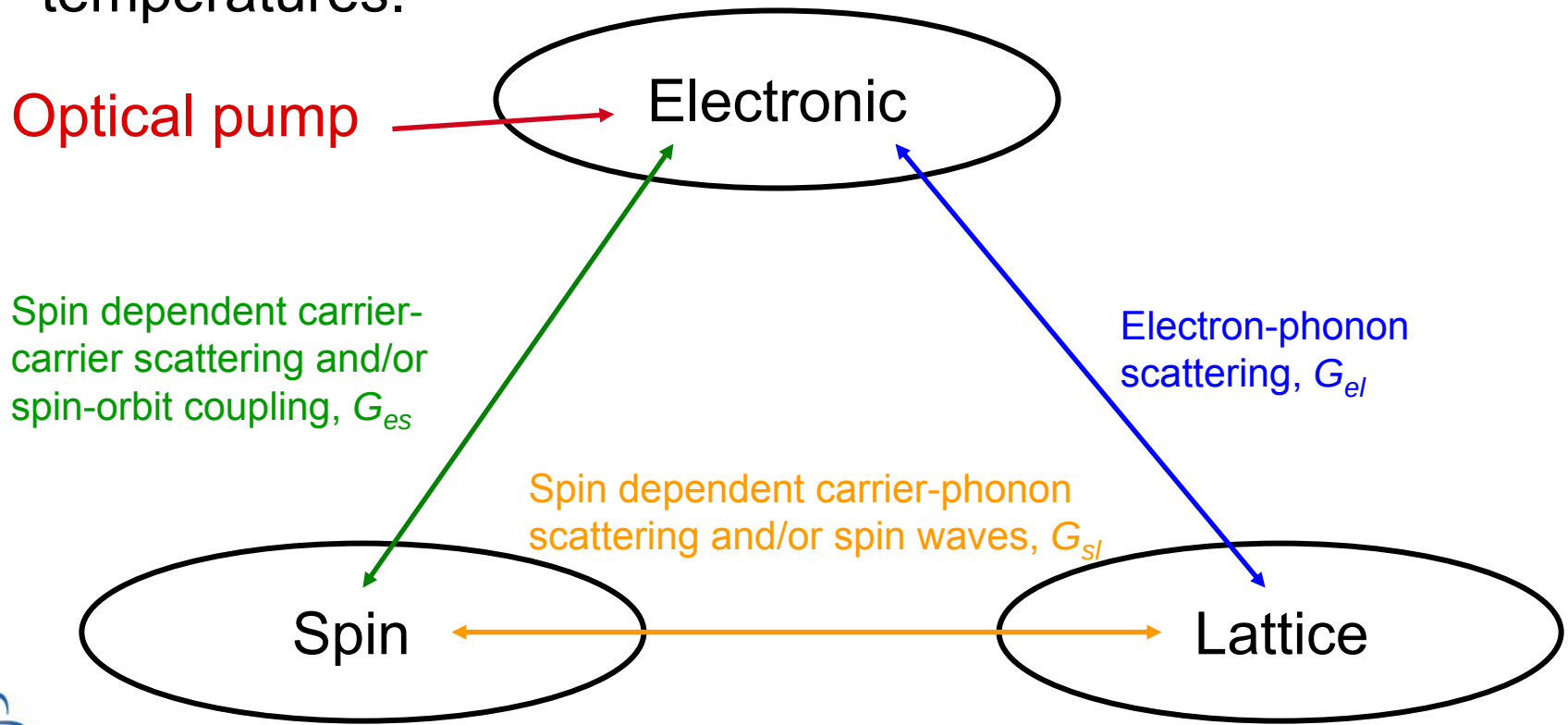
$$E_{THz} \sim \frac{\partial^2 M}{\partial t^2}$$

- Emission is a direct measure of the changing magnetization (versus pump-probe measurements which probe electronic processes)
- Polarization dependence of emission consistent with this picture

Beaupaire *et al.*, Appl. Phys. Lett. **85,** 3025 (2004).

Three Temperature Model

Model the metal as interacting electron, lattice, and spin subsystems that are described by three coupled temperatures.



Ultrafast Demagnetization

Coupled electron, lattice and spin systems result in 3-temperature model for their temperatures:

$$C_E(T_E)\frac{dT_E}{dt} = -G_{EL}(T_E - T_L) - G_{ES}(T_E - T_S) + P(t)$$

$$C_L(T_L)\frac{dT_L}{dt} = +G_{EL}(T_E - T_L) - G_{SL}(T_S - T_S)$$

$$C_S(T_S)\frac{dT_S}{dt} = +G_{ES}(T_E - T_S) - G_{SL}(T_S - T_S)$$

C_i = Specific Heat

G_{ij} = Coupling Constant

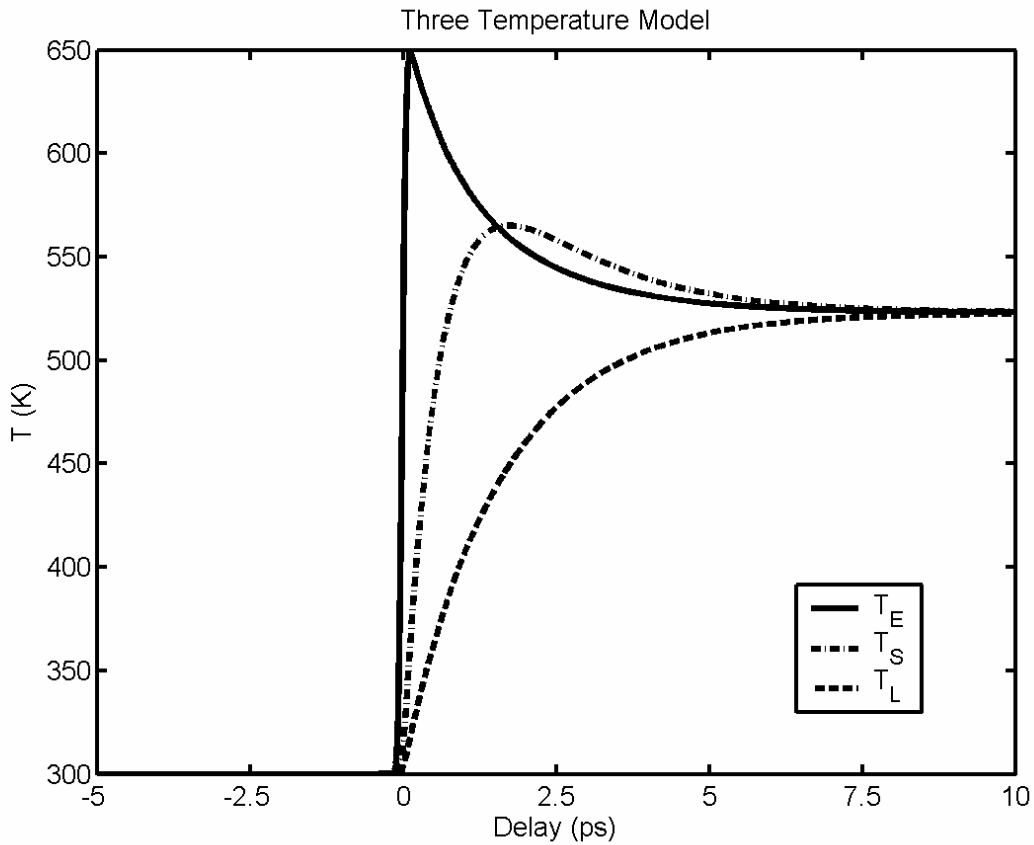
$P(t)$ = Pump Fluence

$$M = M(T_E)$$

Heating the sample results in an increase in the spin dependent scattering rate

$$E_{THz} \sim \frac{\partial^2 M}{\partial t^2}$$

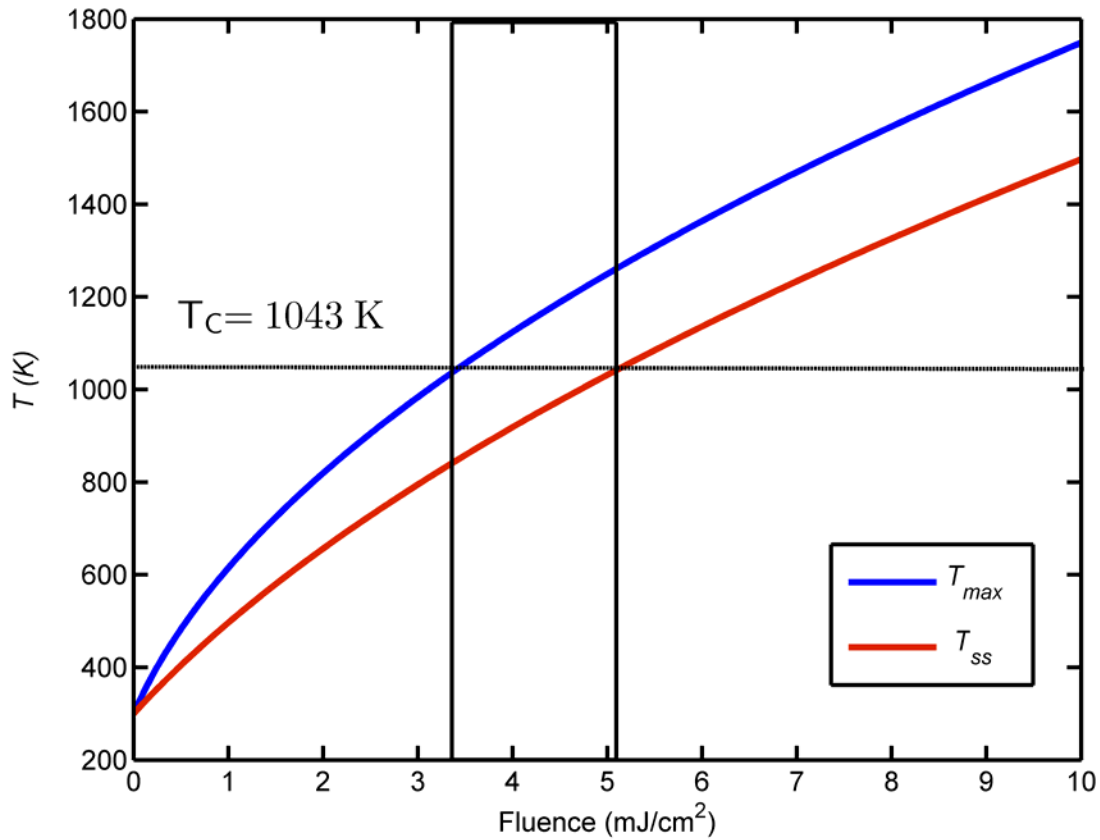
Three Temperature Model



Fast Electron Temperature relaxation time of a few ps.

Emission bandwidth limit ~500 GHz.

High Fluence Emission



Optical Pump, THz probe

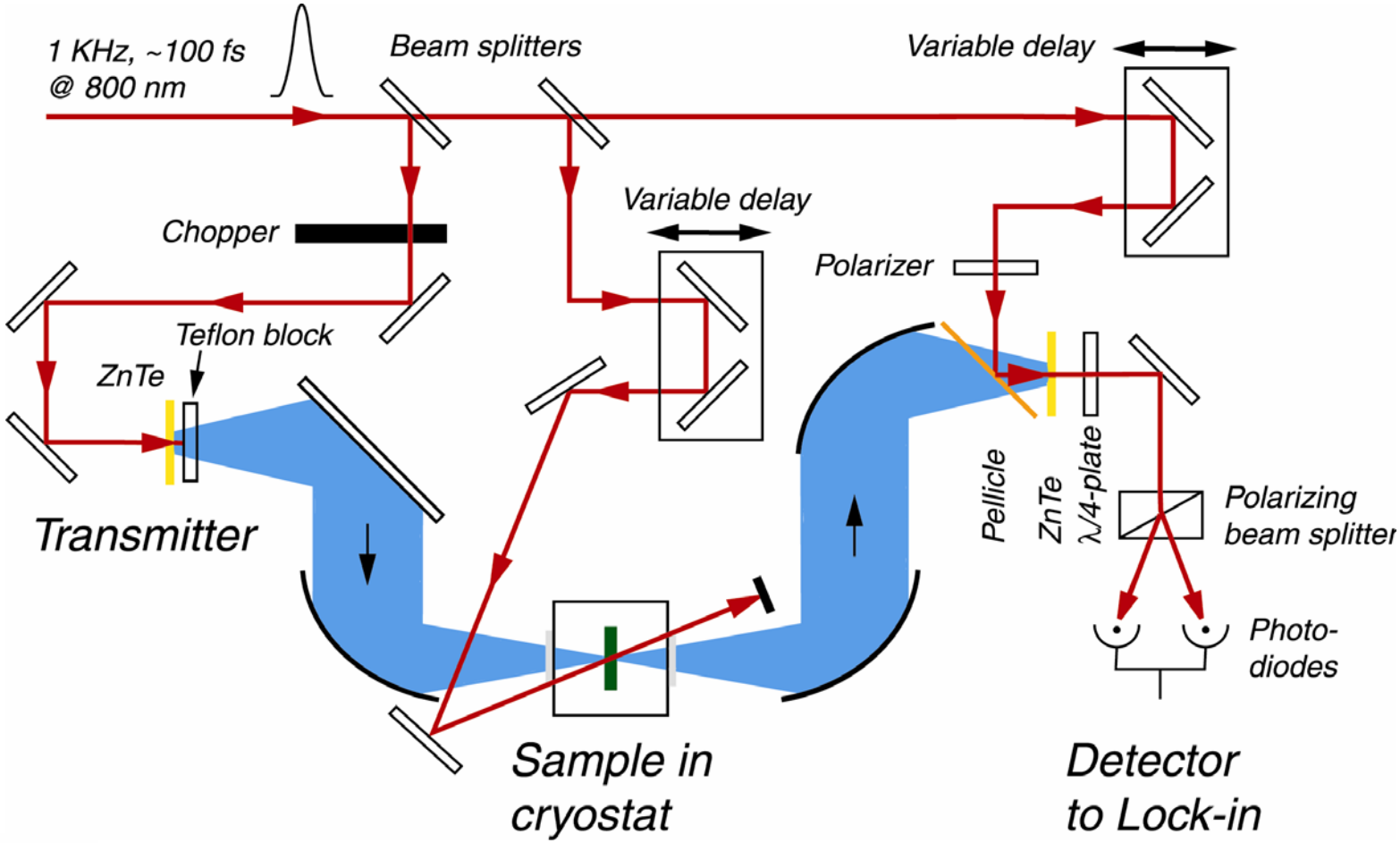
If the magnetization changes are caused by a change to the scattering rate ($\tau_0 \rightarrow \tau_0 + \Delta\tau$), then a THz pulse transmitted through an optically excited sample should see a time-dependent phase shift.

$$\sigma(\omega) + \Delta\sigma(\omega) \approx \frac{\sigma_0}{1-\omega^2\tau^2}\left(1 + i\omega\tau + i\omega\Delta\tau\right)$$

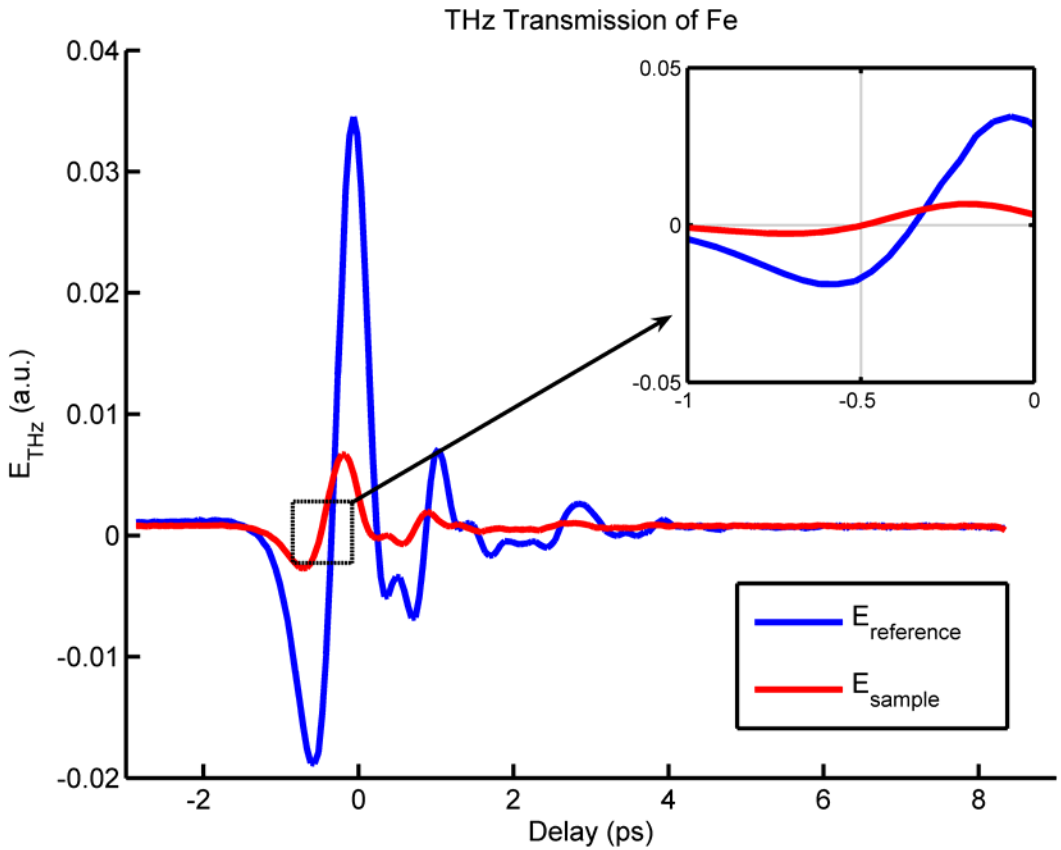
$$\Delta\sigma(\omega) \approx i\left[\frac{\omega\sigma_0}{1-\omega^2\tau^2}\right]\Delta\tau$$

TRTS can see the change to the full scattering time, not just the *spin-dependent* contribution.

Optical Pump/Terahertz Time-Domain Spectroscopy—Ultrafast Ohm-Meter



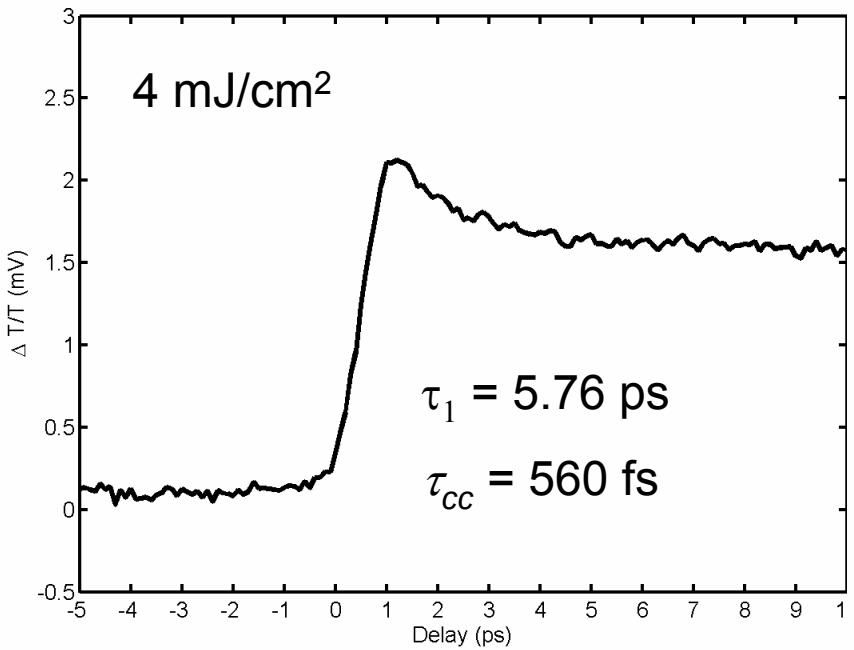
Waveform Phase Shift



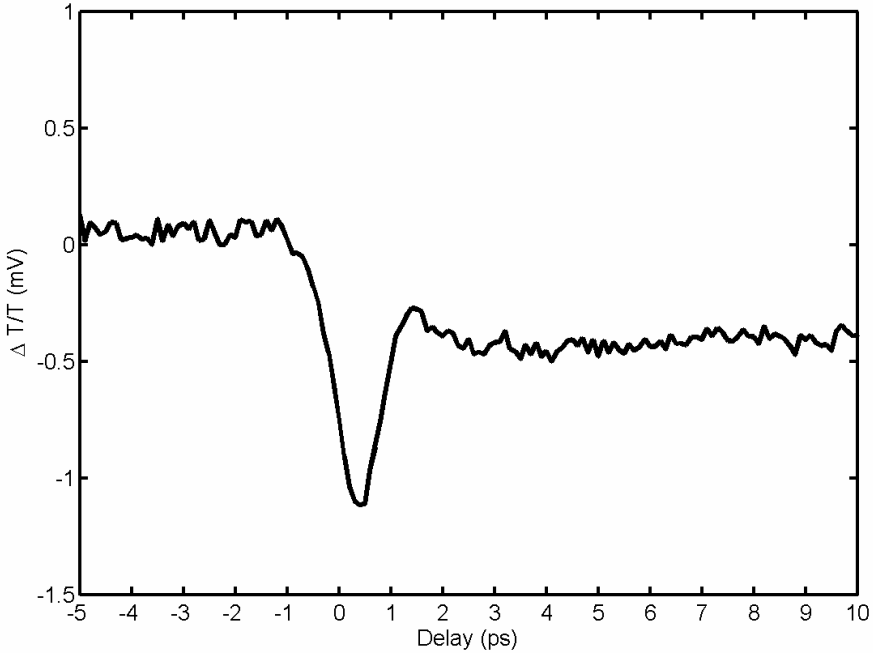
A phase shift results in a change in the position of the zero.

Optical Pump, THz probe

Amplitude Change



Phase Change

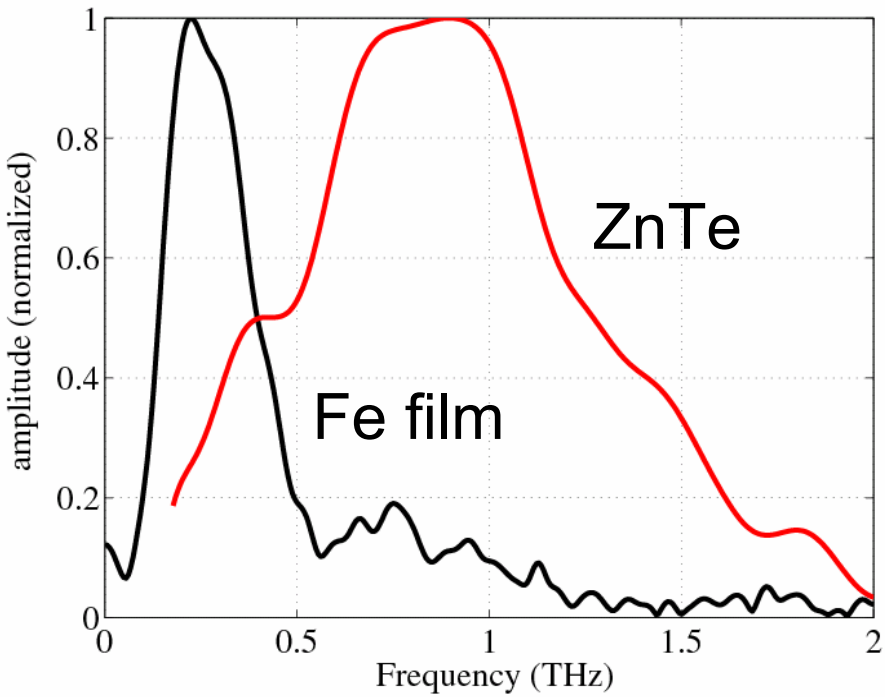


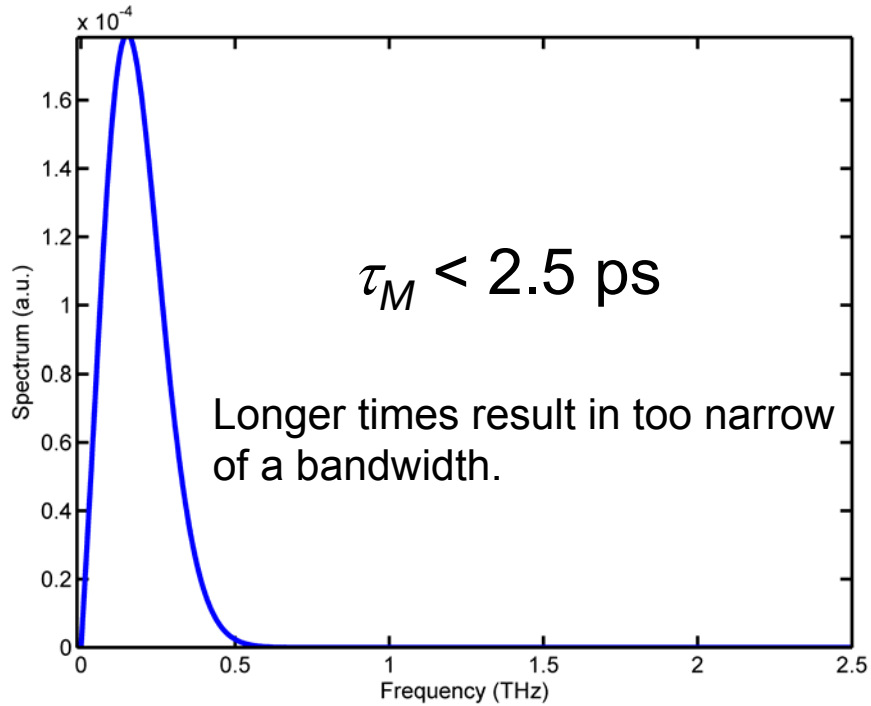
This measures changes to the *electronic* subsystem

Estimate the Emission Bandwidth

Using the electronic parameters, simulate *M(T(t))* and find *E(t)* using Three Temperature Model.

τ_E = 5.76 ps and τ_{cc} = 560 fs





τ_M < 2.5 ps

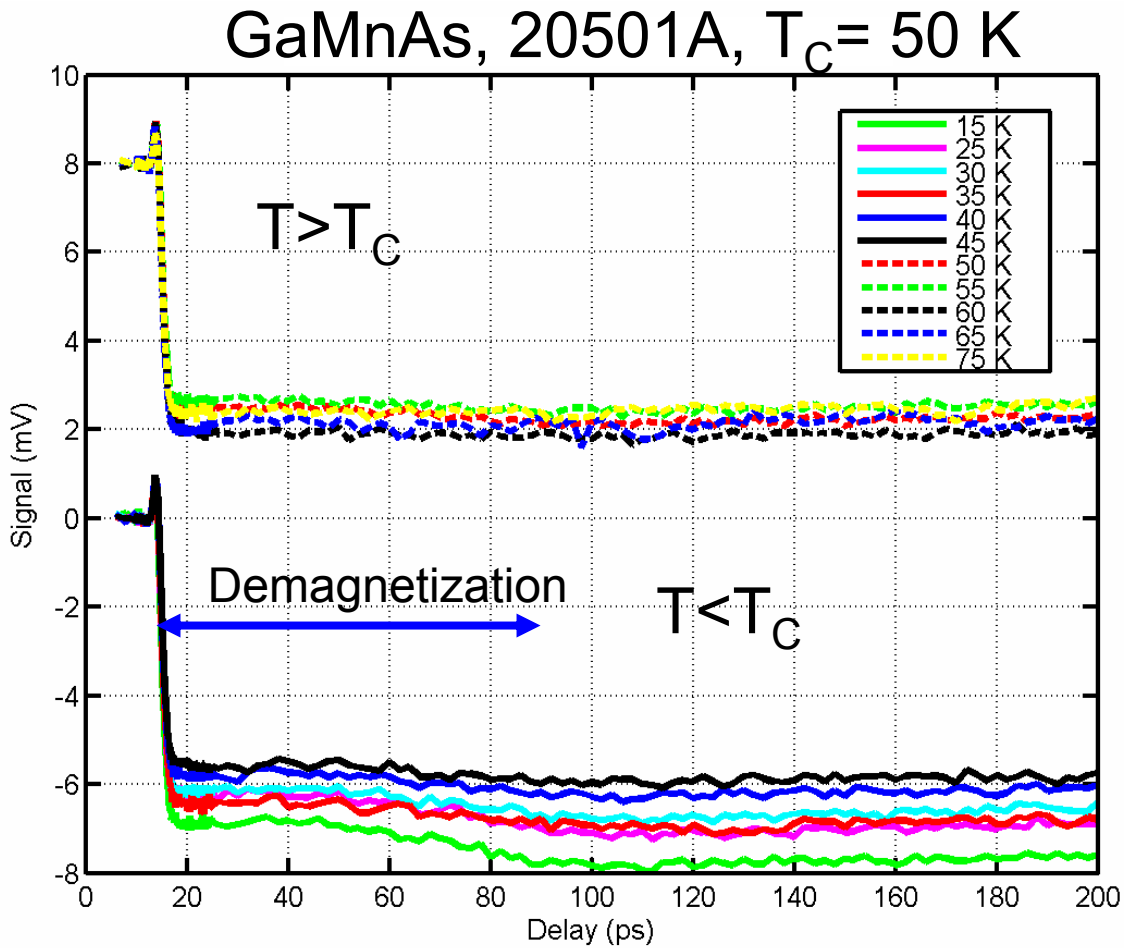
Longer times result in too narrow of a bandwidth.

Ferromagnetic Semiconductors

Carrier mediated ferromagnetism (magnetic polarons)

Demagnetization appears to proceed on a ~100 ps timescale (10 GHz).

More to come…



With J. Furdyna Group, Notre Dame

THz emission via fs excitation

Mechanisms:

- Current Surge - FIR dipole radiation from acceleration of photo-injected carriers in a surface depletion field
- Coherent Phonon Generation
- Pondermotive acceleration of electrons in a laser plasma
- Optical Rectification - Difference Frequency Mixing
  - Bulk electric-dipole: $\chi^{(2)}$
  - Bulk electric-quadrupole/magnetic dipole: $\chi^{(Q)}$
  - Field Induced (Surface) electric-dipole: $\chi^{(3)}$, E_d
  - Surface or bulk magnetization: $\chi^{(2)}(\boldsymbol{M})$
- Ultrafast demagnetization—FIR dipole radiation from rapid demagnetization following the creation of a nonthermal electron distribution with a fs optical pulse.

Summary

Conclusions:

- **THz emission following ultrashort pulse excitation of an Fe film was observed after transmission through the film.**
- **THz field amplitude scales linearly with optical excitation fluence up to 4 mJ/cm^2 of incident fluence.**
- ***E*(ϕ, θ) dependence of THz radiation reveals that the generation mechanism is dominated by a magnetic mechanism with a contribution from a surface electric dipole nonlinearity.**
- **Mechanism of ultrafast demagnetization results in narrowed THz bandwidth.**
- **Preliminary optical pump, THz probe data show an increase in scattering rate immediately after fs pulse excitation… is this spin dependent scattering?**

Future Directions:

- **Measurement of the THz emission in reflection geometry.**
- **Measure emission at fluence's that result in complete demagnetization.**
- **Measurement of THz emission from non-magnetic metals.**
  - **Recent work has demonstrated emission from amorphous gold and silver films (Kadlec, *et al*. scheduled for publication in Optics Letters).**