Intense Coherent THz Pulses from the DUV-FEL Linac: Characteristics and Potential Applications

Henrik Loos, Brian Sheehy and G. Lawrence Carr

National Synchrotron Light Source

Brookhaven National Laboratory

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Why THz Pulses?

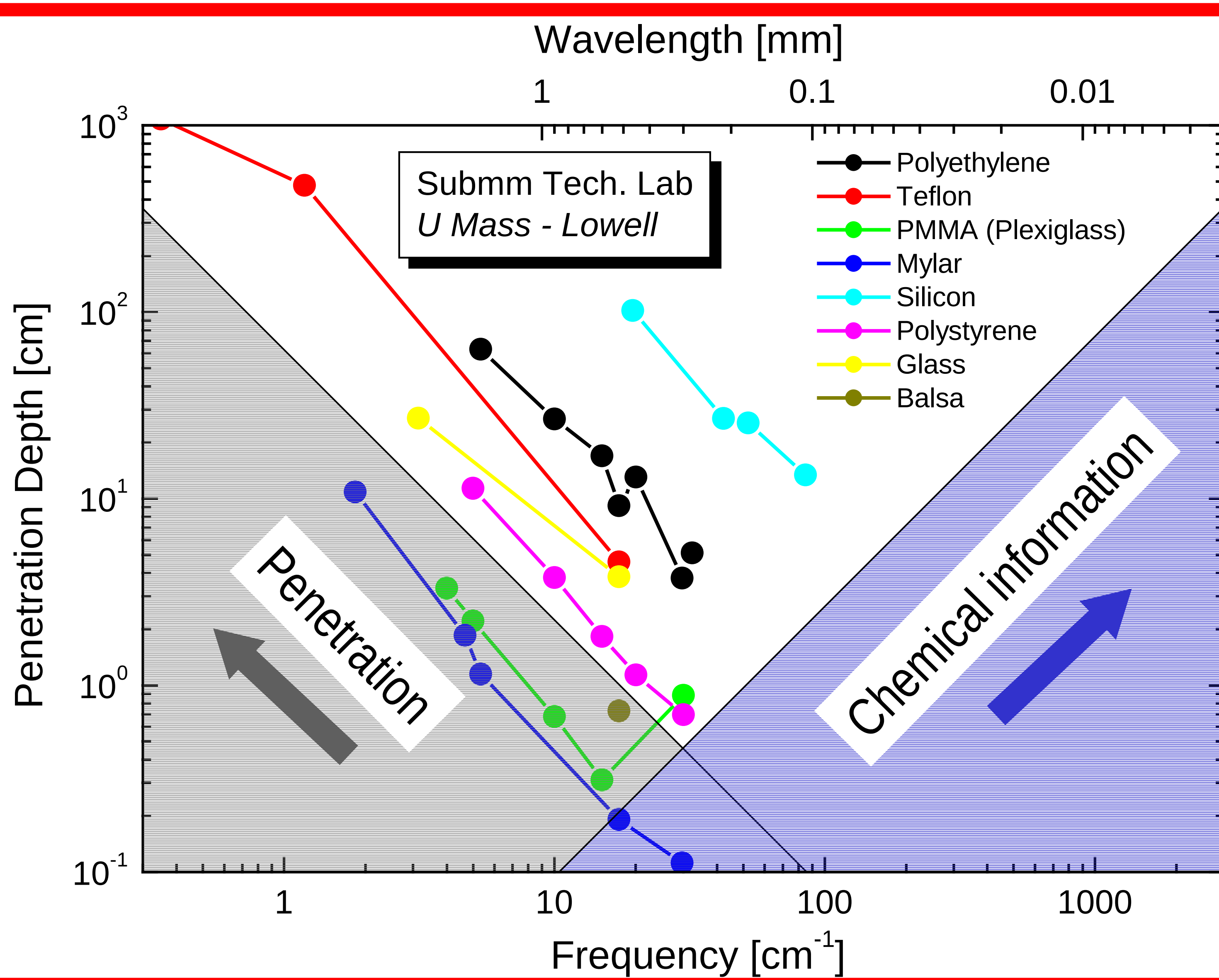
1 THz = 10^{12} Hz ; λ = 300 μm, ν = 33 cm^{-1}

- Ultra-fast dynamics (< 1 ps time scale)
  - electronic excitations
  - magnetic excitations
- Low frequency, non-linear properties of materials
  - vibrational modes (local phonon modes)
  - electronic modes (nanoparticles / quantum dots)
- Structural transitions
  - Large E-field to coherently “shove” atoms
- Imaging
  - non-ionizing
  - crossroads between depth of penetration, chemical info.

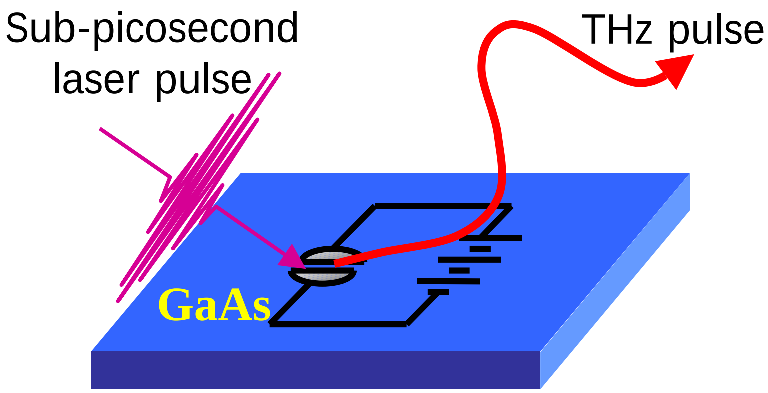
Pulsed THz Source Requirements

- Broad spectral content (up to at least 3 THz)
- Peak E-field approaching 1 MV/cm
- Tailored waveform
  - “Half-cycle’ pulse, other
- Two colors
  - a tunable, low energy pump pulse
  - similar or even lower energy spectroscopic probe pulse
- Sufficient average power for imaging large areas at high (video) rates.

THz Imaging: Absorption in Common Dielectric Materials



Energy/Power for "Table Top" THz Pulses



Radiation from acceleration of photocarriers.

$$P = \frac{2e^2a^2}{3c^3}\gamma^4 \quad (1\ e)$$

10^6 V/m => a ~ 10^{19} cm/s^2

Energy = 6x10^{-32} J per electron in 1 ps

or 10^{-32} J/cm^{-1}/electron

- Multiparticle (N^2) coherent enhancement (spotsize smalller than diff. limit).
- Typical energies per pulse are ~ 1 nJ from a conventional photoconductive switch and an amplified, 250 kHz rep rate drive laser.
- Optical rectification output is similar, but extends spectra to higher frequencies.
- Higher peak power lasers (at or below 1 kHz rate) and biased antenna structures have enabled **1 μJ**. (You et al, *Opt. Lett.* **18**, 290 (1993), Budiarto et al, *J. Quant. Elect.* **32**, 1839 (1996).
- Recently, Riemann et al [*Opt. Lett.* **28**, 471 (2003)] demonstrated multi-cycle mid-IR (~ 800 cm^{-1} = 24 THz) pulses with E-field of **1 MV/cm**. (mixing in GaSe)

Transition Radiation from Relativistic Electron

Transition radiation occurs when an electron crosses the boundary between two different media. For a relativistic electron ($\beta \equiv v/c \cong 1$) incident on a perfect conductor, the number of photons emitted per solid angle and wavelength range is:

$$\frac{dN}{d\lambda d\Omega} = \frac{\alpha}{\pi^2 \lambda} \frac{\beta^2 \sin^2\theta \cos^2\theta}{\left(1-\beta^2\cos^2\theta\right)}$$

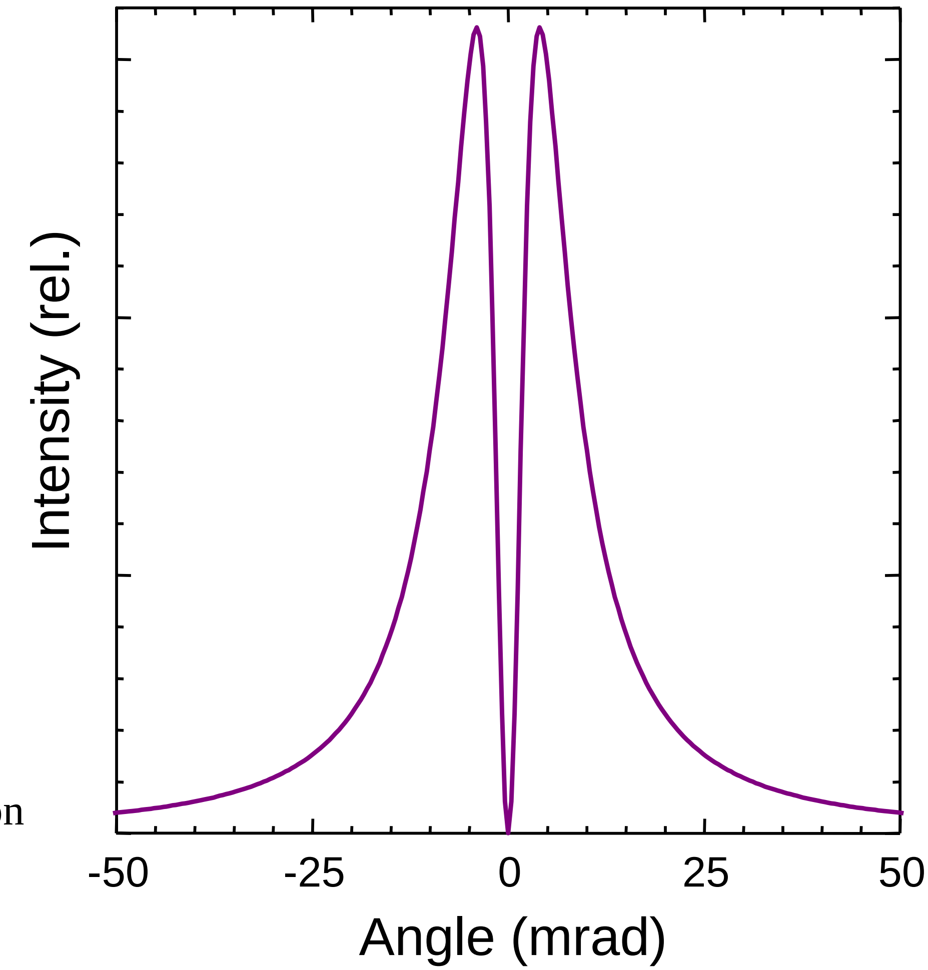
Intensity is 0 on axis, peaks at $\theta \sim 1/\gamma$.

Polarization is <u>radial</u>

$$\frac{dP}{d\bar{\nu}} \approx 4.61\times10^{-26} \underbrace{\left(\ln\frac{2}{1-\beta}-1\right)} \text{ J/cm}^{-1} \text{ per electron}$$

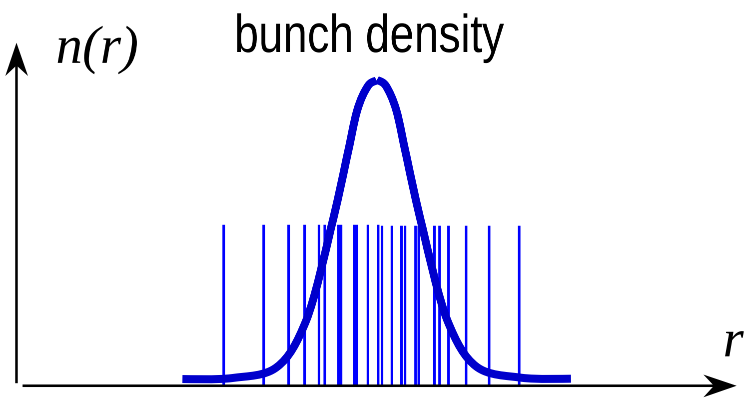
11.4 for 130 MeV
20 for 9 GeV

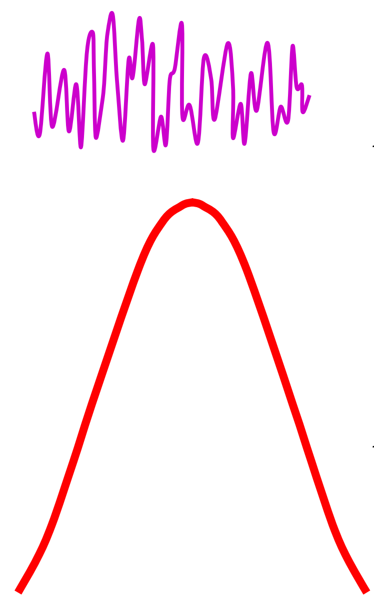
Far field distribution for γ = 200



Multi-particle Coherent Synchrotron Radiation (CSR)

- What are requirements for CSR from electrons in a bunch?
  - bunch (or some portion of it) has density variations on length scale comparable to wavelength.





$\lambda << l_b$
$E \sim N^{1/2}; I \sim N$

$\lambda >> l_b;$
$E \sim N; I \sim N^2$

$$\left.\frac{dI(\omega)}{d\omega}\right|_{multiparticle} = \left[N + N(N-1)f(\omega)\right]\frac{dI(\omega)}{d\omega}$$

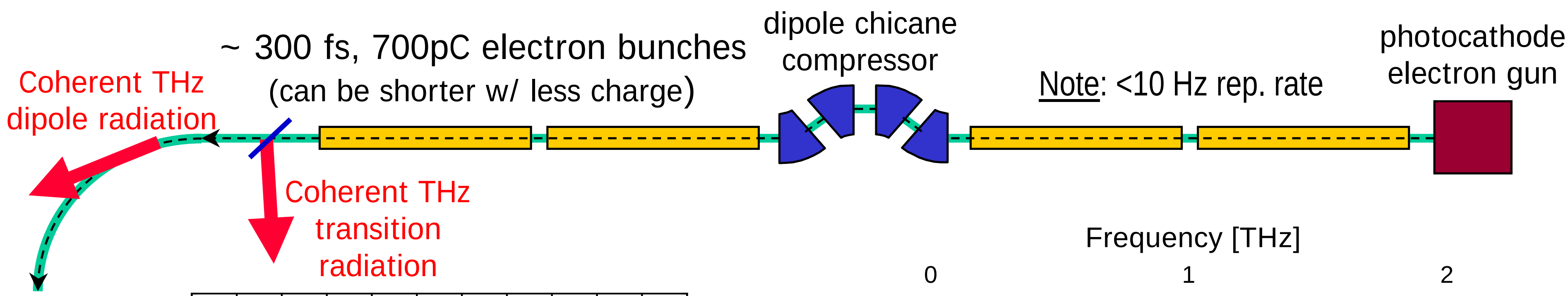
where $f(\omega) = \left| \int_{-\infty}^{\infty} e^{i\omega \hat{n}\cdot\vec{r}/c} S(r)dr \right|^2$ *(Nodvick & Saxon)*

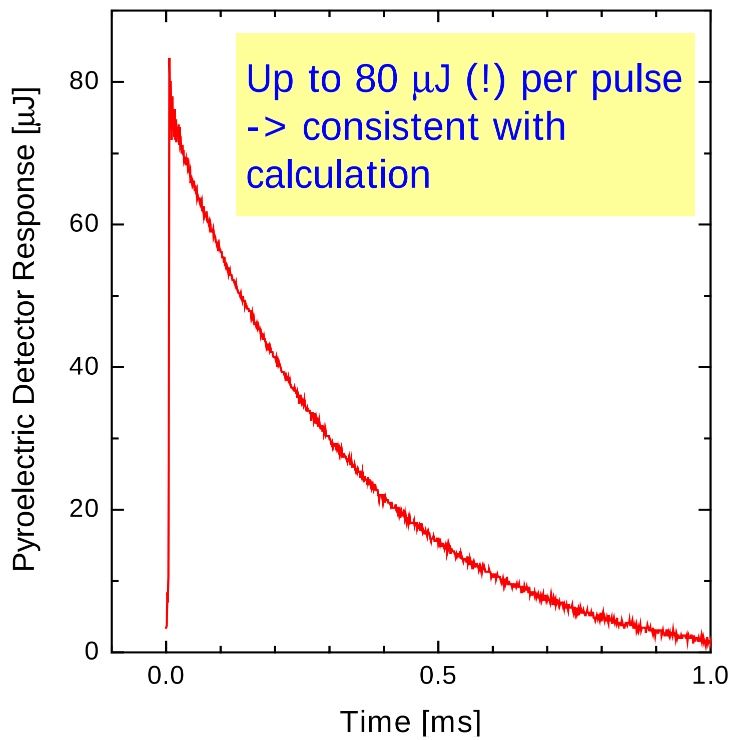
In some accelerators, bunch lengths are 100s of fs (=>THz), and N can be large e.g. ~ 10^{10}

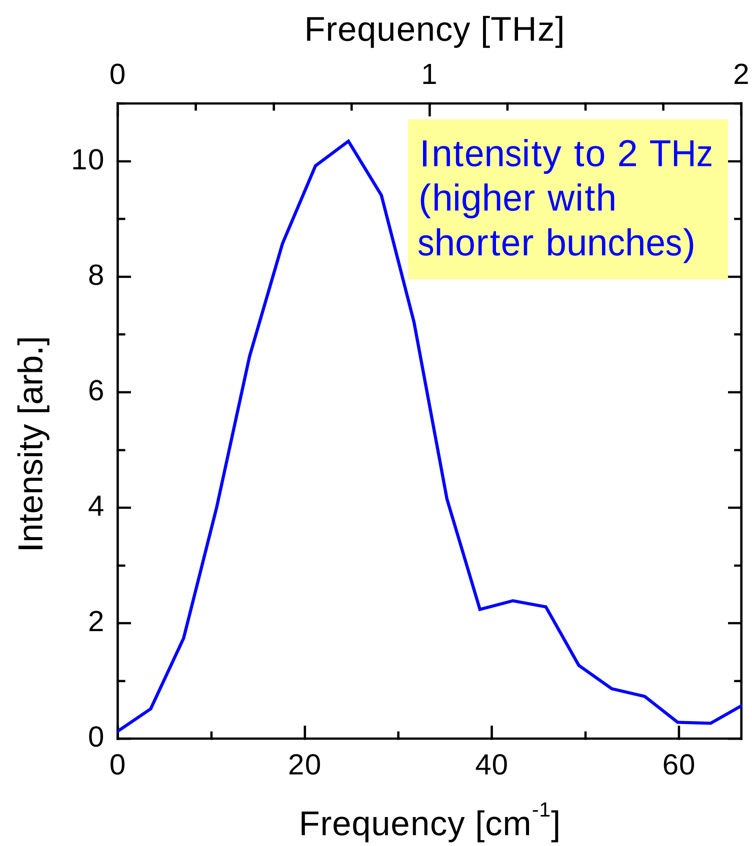
Coherent Synchrotron Radiation (CSR)

- 1st observations in linacs:
  - Nakazato et al (PRL ‘89), Happek et al (PRL ‘91)
- As a linac bunch diagnostic:
  - Shibata et al (PRE ‘94), Lai et al (PRE ‘94), Yan et al (PRL ‘00)
- As a THz source
  - Ishi et al (PRA ‘91), Takahashi et al (RSI ‘98), Carr et al., (Nature ‘02)
- CSR also from storage rings
  - Arpe et al, Carr et al, Anderson et al, Abo-Bakr et al. ...

Coherent Transition Radiation Pulses from the SDL Linac

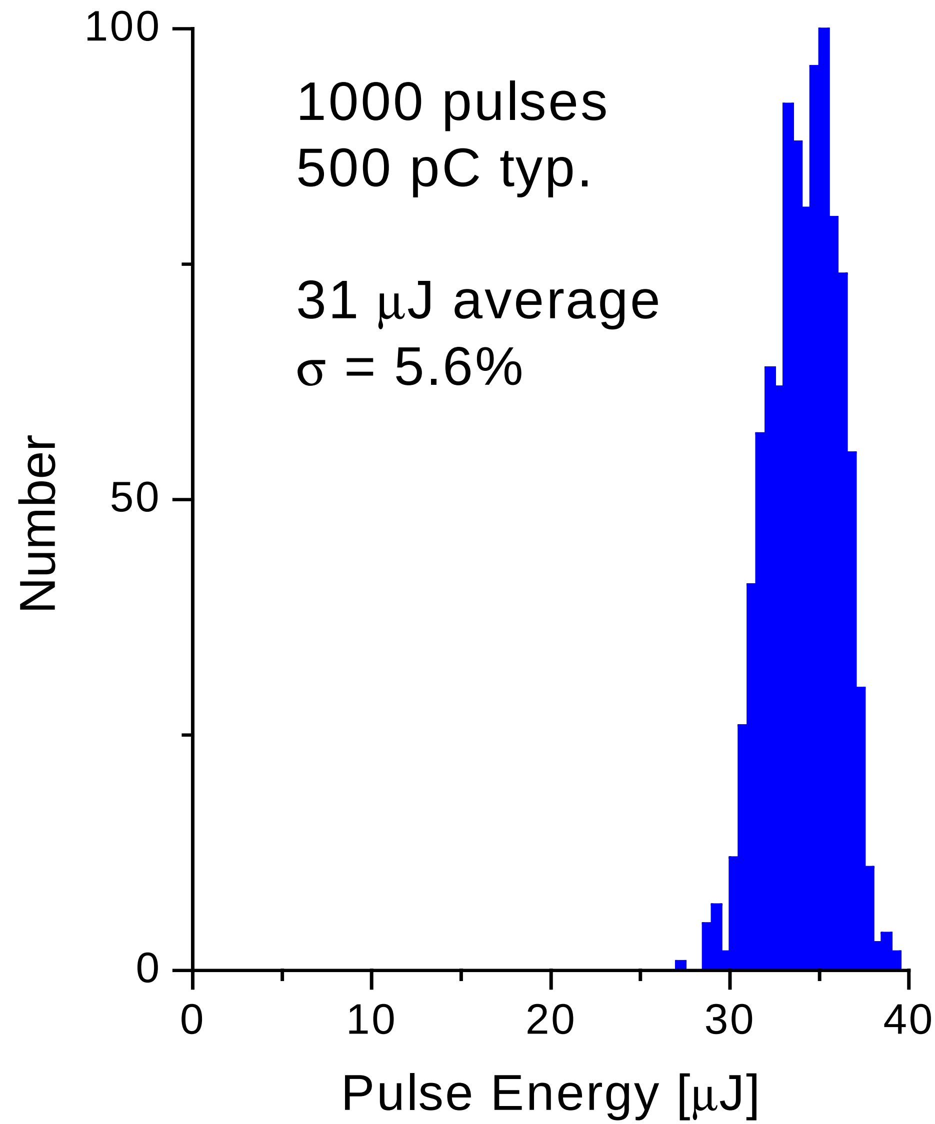




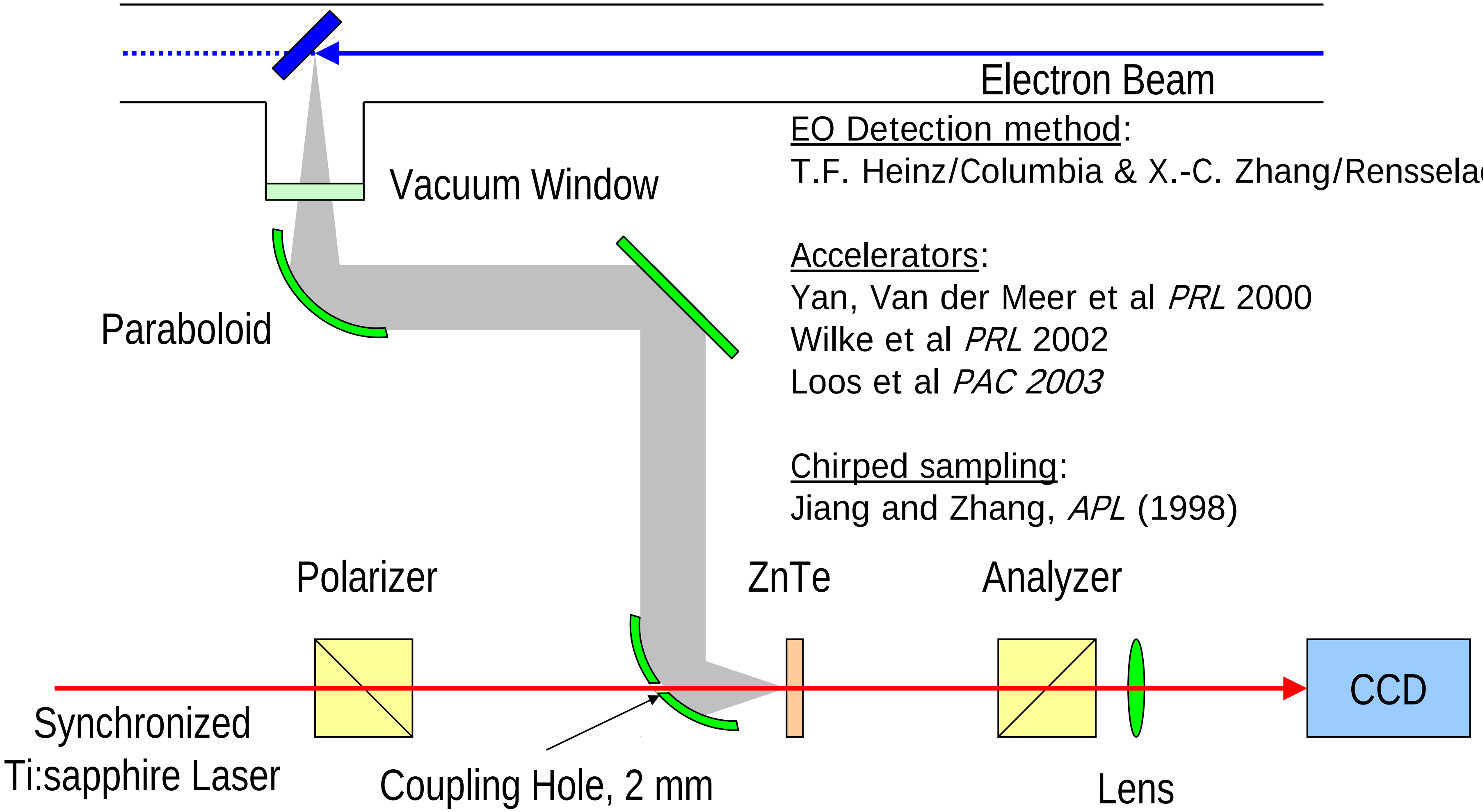


Statistics

- Shot-to-shot fluctuations affect pulse usefulness for some applications.
  - Interferometry
  - Pump-probe spectroscopy
- Typical fluctuations 4 to 6% RMS.
  - due mostly to variations in charge (particle number).
- High rep. rate (average) or single-shot capability needed.



Electro-Optic THz Pulse Detection Setup

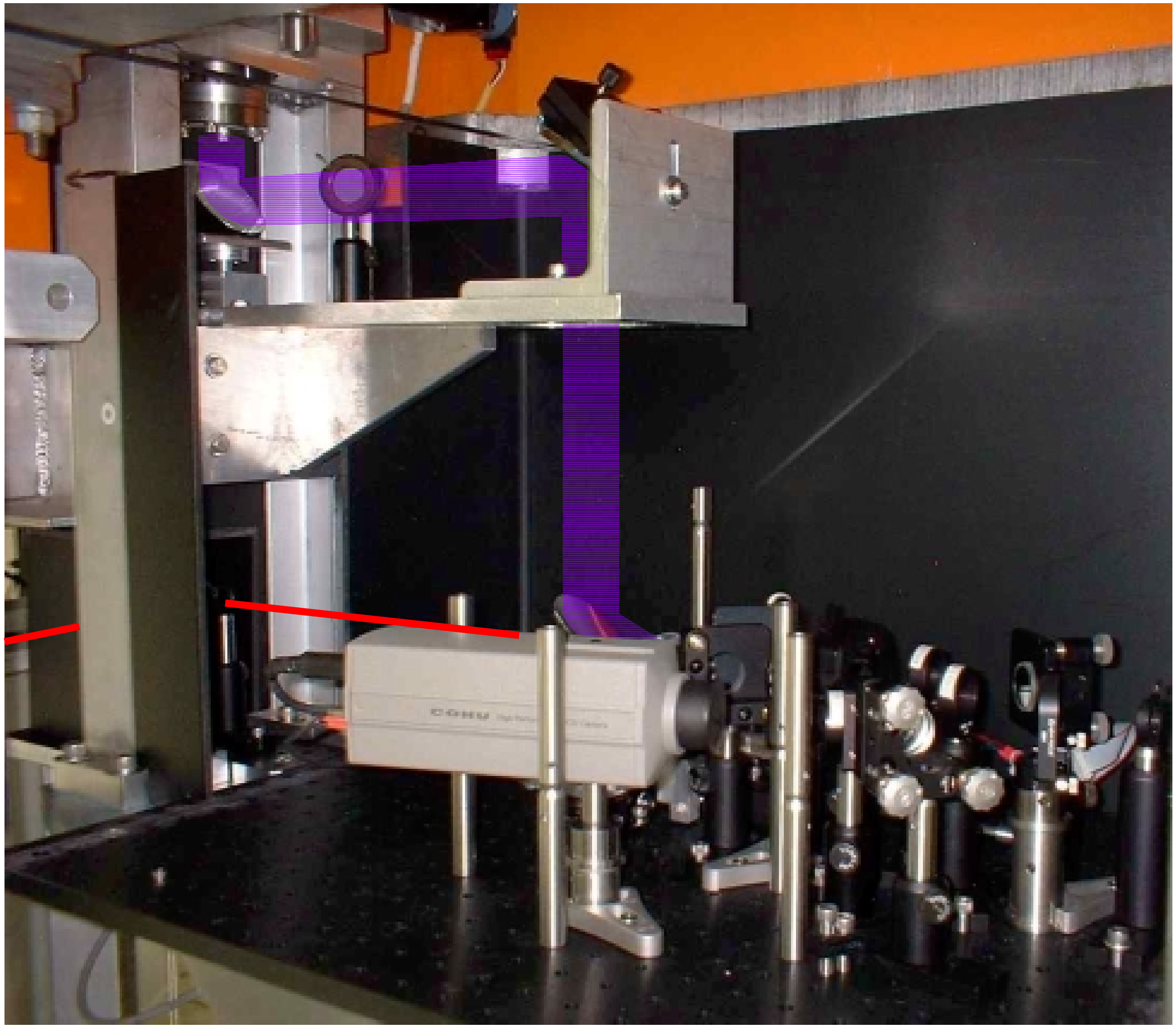


<u>EO Detection method</u>:
T.F. Heinz/Columbia & X.-C. Zhang/Rensselaer

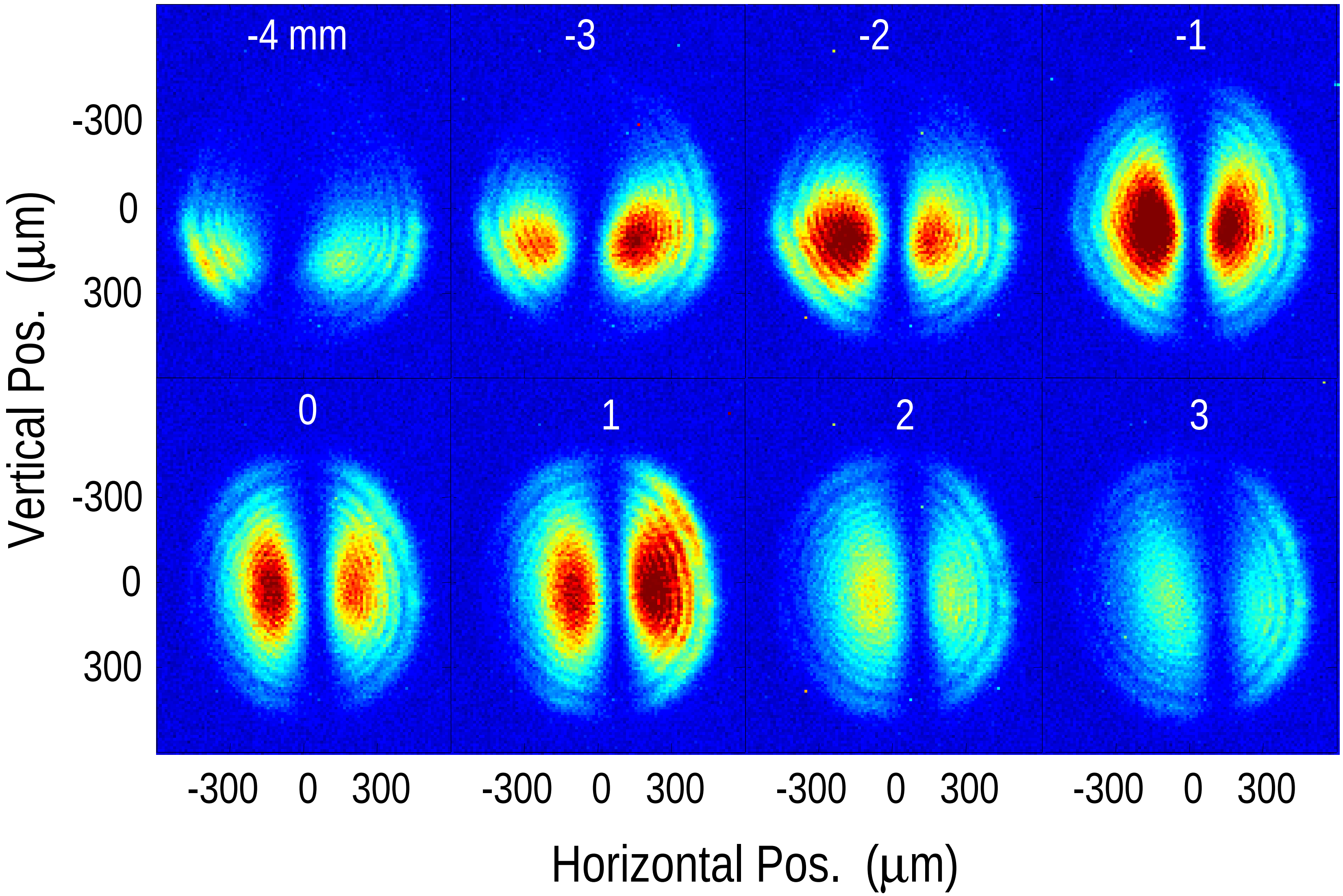
<u>Accelerators</u>:
Yan, Van der Meer et al *PRL* 2000
Wilke et al *PRL* 2002
Loos et al *PAC 2003*

<u>Chirped sampling</u>:
Jiang and Zhang, *APL* (1998)

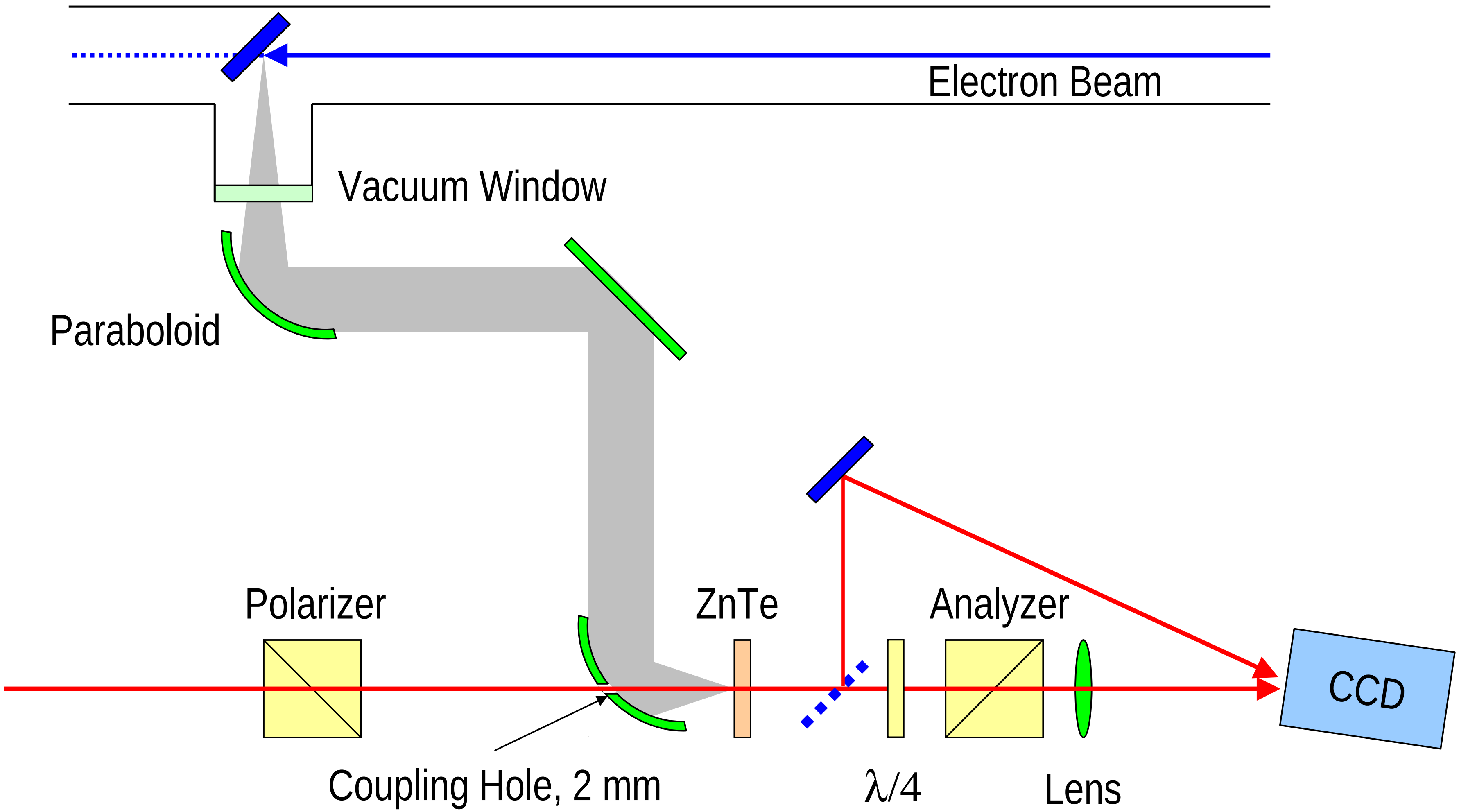
THz and Sampling Laser Beam Path



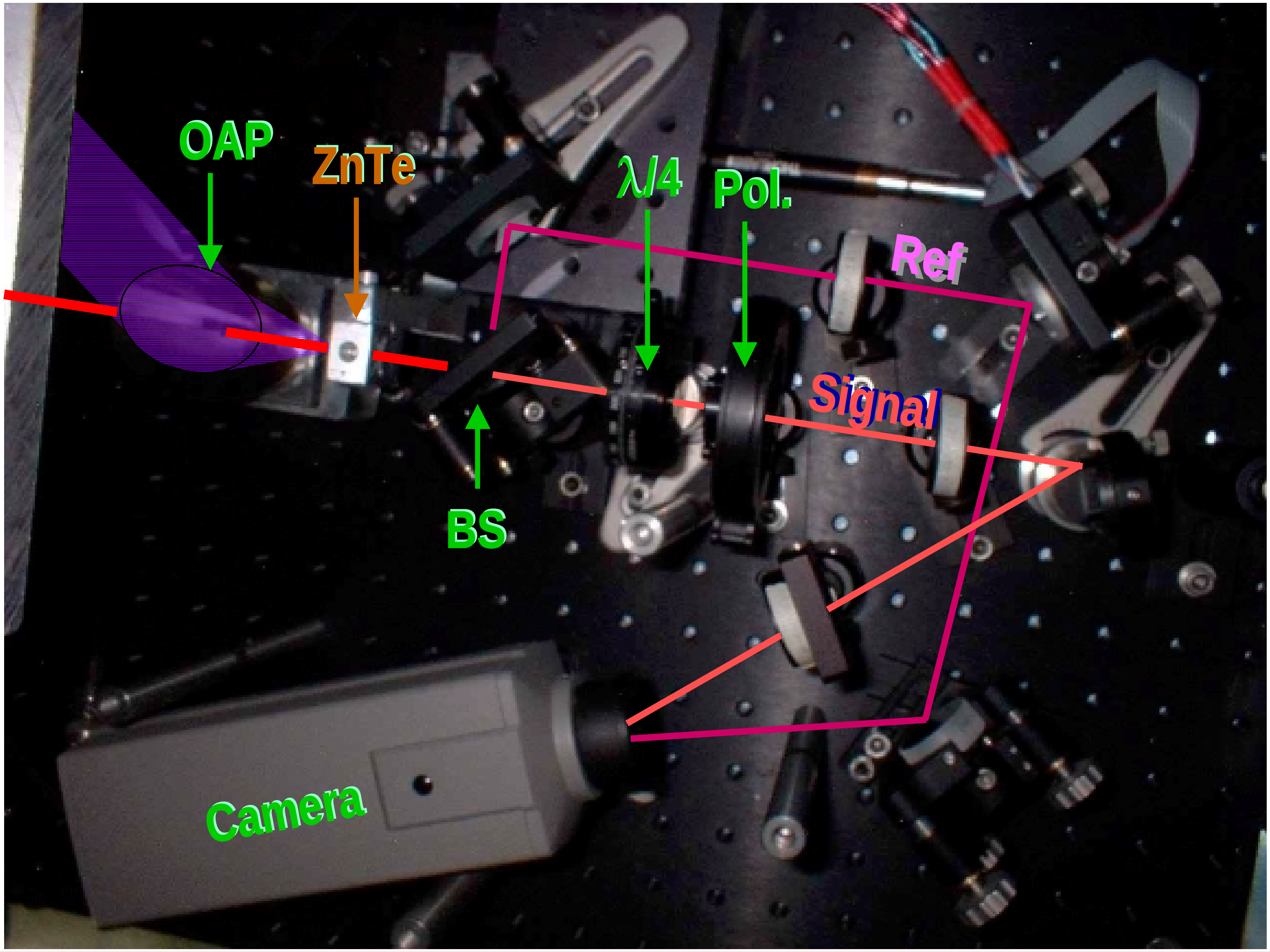
EO Images of Electric Field (magnitude) Through Focus



Electro-Optic THz Detection with λ/4



Signal and Reference



EO Detection of SDL Linac Coherent THz Pulses

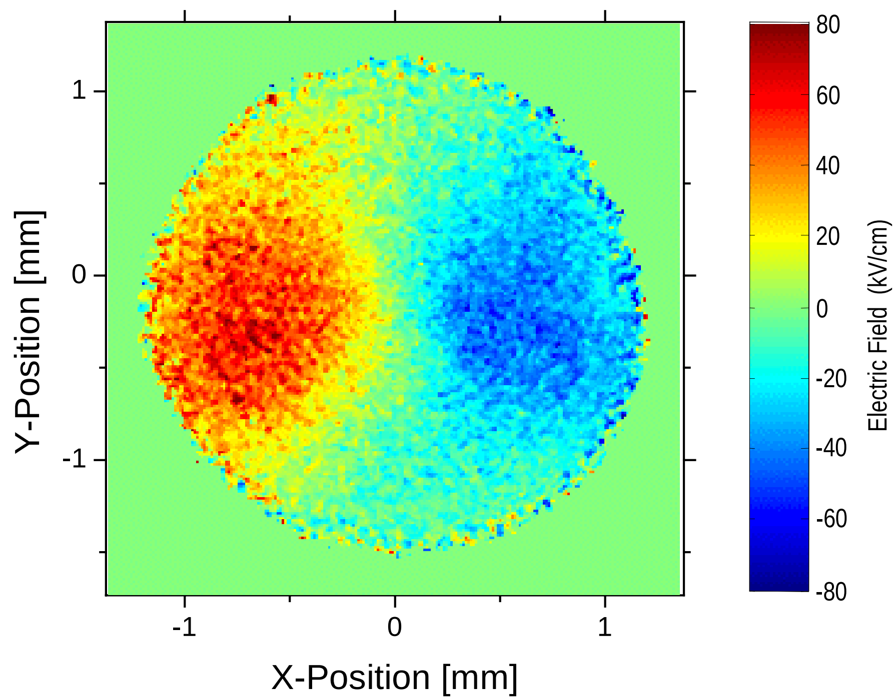
Focusing a 100 μJ pulse, 1 THz (nominal) pulse into a 1 mm^3 volume yields an energy density of $10^5 J/m^3$, so that $E = [2D_E/\varepsilon_0]^{1/2}$ **~ 10^8 V/m (~ 1 MV/cm)**.

At 1 MV/cm, the magnetic field (B=E/c) is **3 kG.**

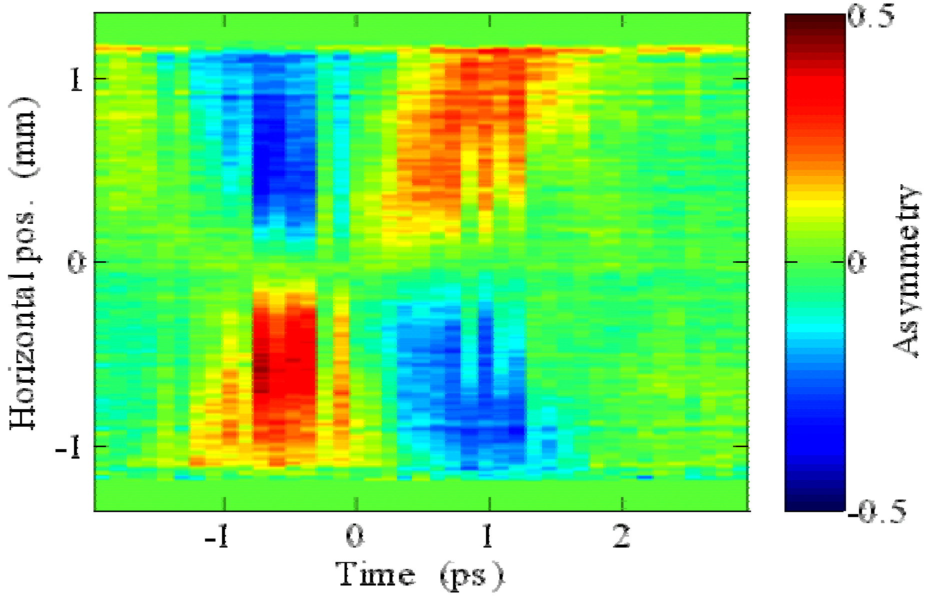
This E-field is <u>too large</u> for 500 μm ZnTe (E > 170 kV/cm yields > λ/4 phase shift)

=> Reduce compression, lower charge to get "on-scale"

single shot image of E_H



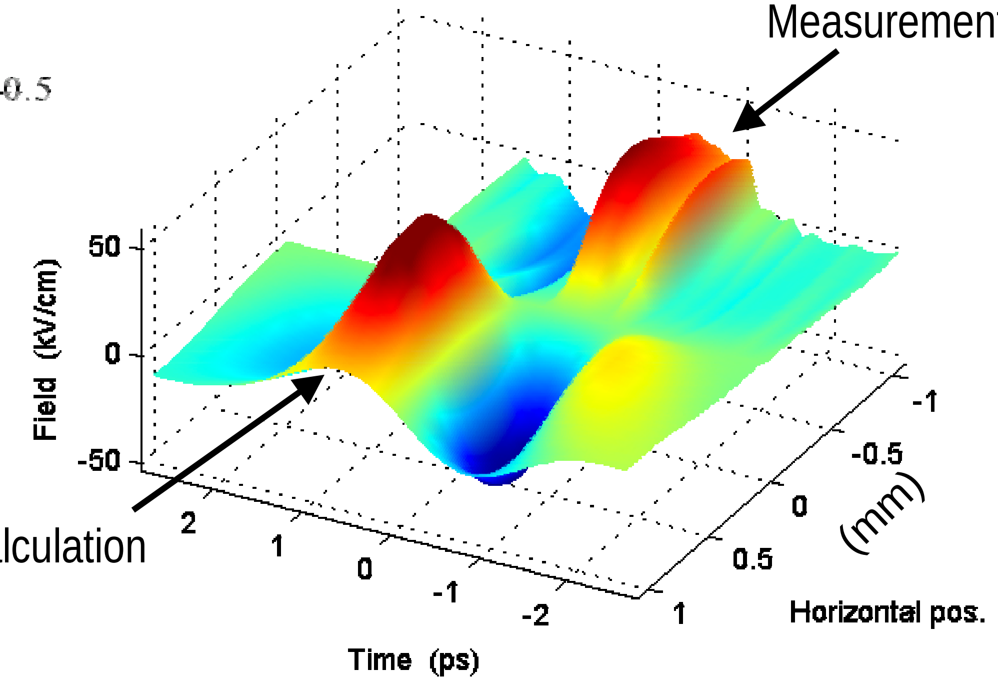
Temporal E-Field Cross Section at Focus



E-field along horizontal plane

Temporal-spatial E-field profile of coherent transition radiation pulse at ~ f/1.5 focus

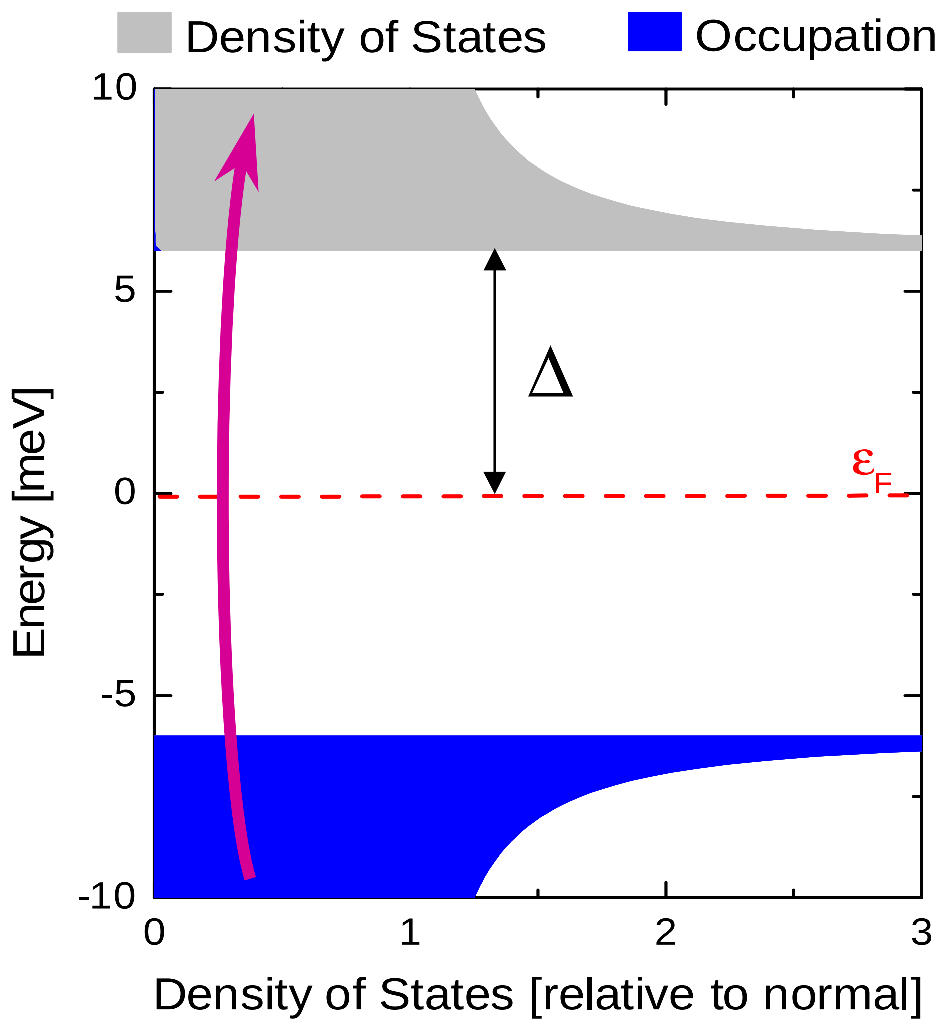
Note: opposite sides are asymmetric, as shown (radial polarization)



What Can We Do with these THz Pulses?

- ✶ Use THz pulses as the pump (excitation) source in “all THz” pump probe experiments.
- ✶ Study non-linear absorption is a variety of materials, including nanoparticles / quantum dots.
- ✶ Induce coherent current excitations
- ✶ Move atoms in their local potential wells
- ✶ Orient spins in magnetic systems.

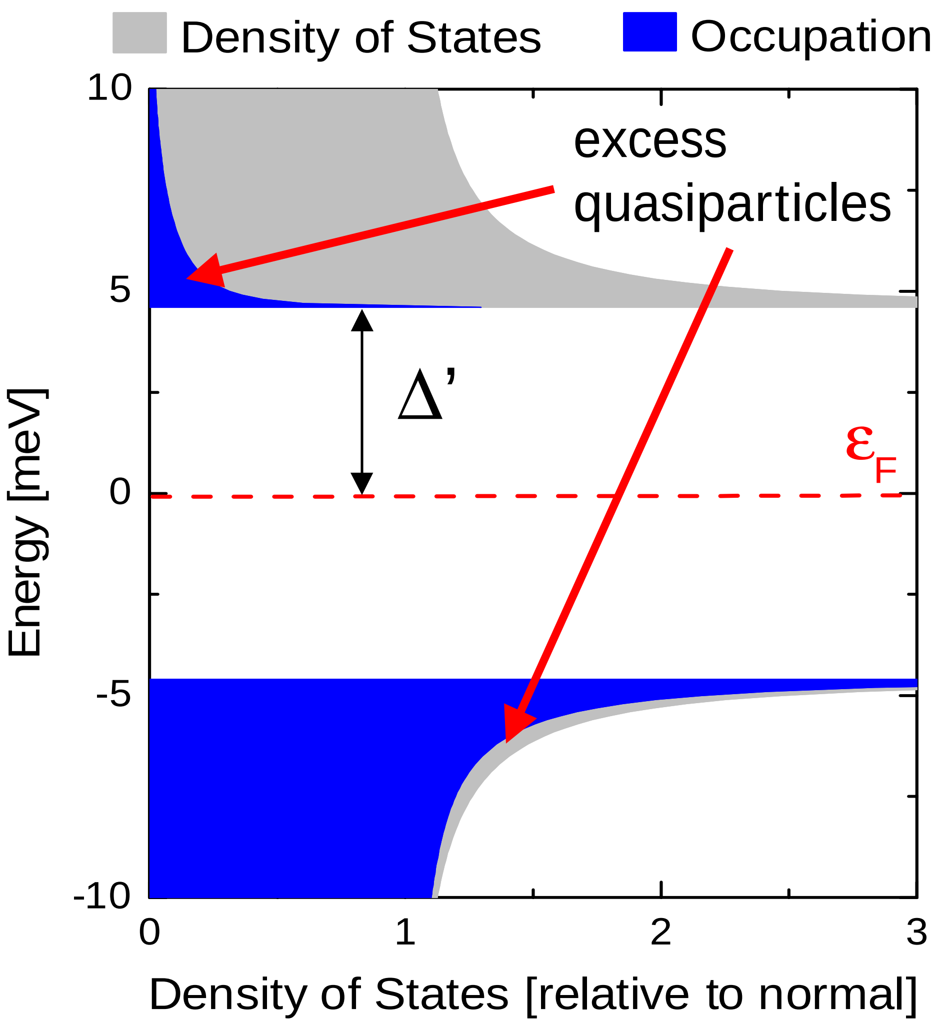
Photoexcitation in a BCS Superconductor



Step 1:
Photons (a pulse from near-IR laser) break pairs, creating high energy “quasiparticle” excitations.

Photon energy 1.5 eV.

Photoexcitation in a BCS Superconductor



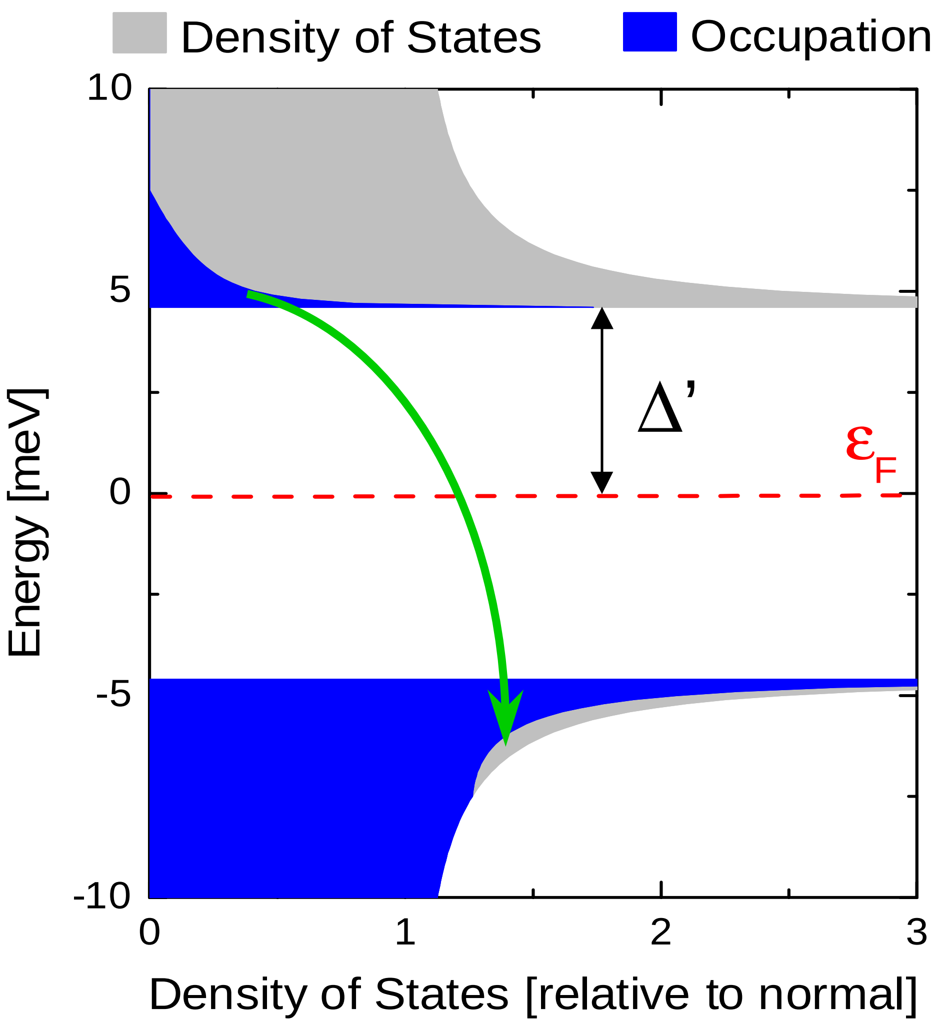
<u>Step 2:</u>

High energy quasiparticles scatter and relax toward gap edge. Many more pairs broken in this process (multiplication).

Weakened superconducting state appears as reduced gap; $\Delta' = \Delta - \delta$.

Note: Quasiparticle density is out of equilibrium, but energy distribution is approximately thermalized.

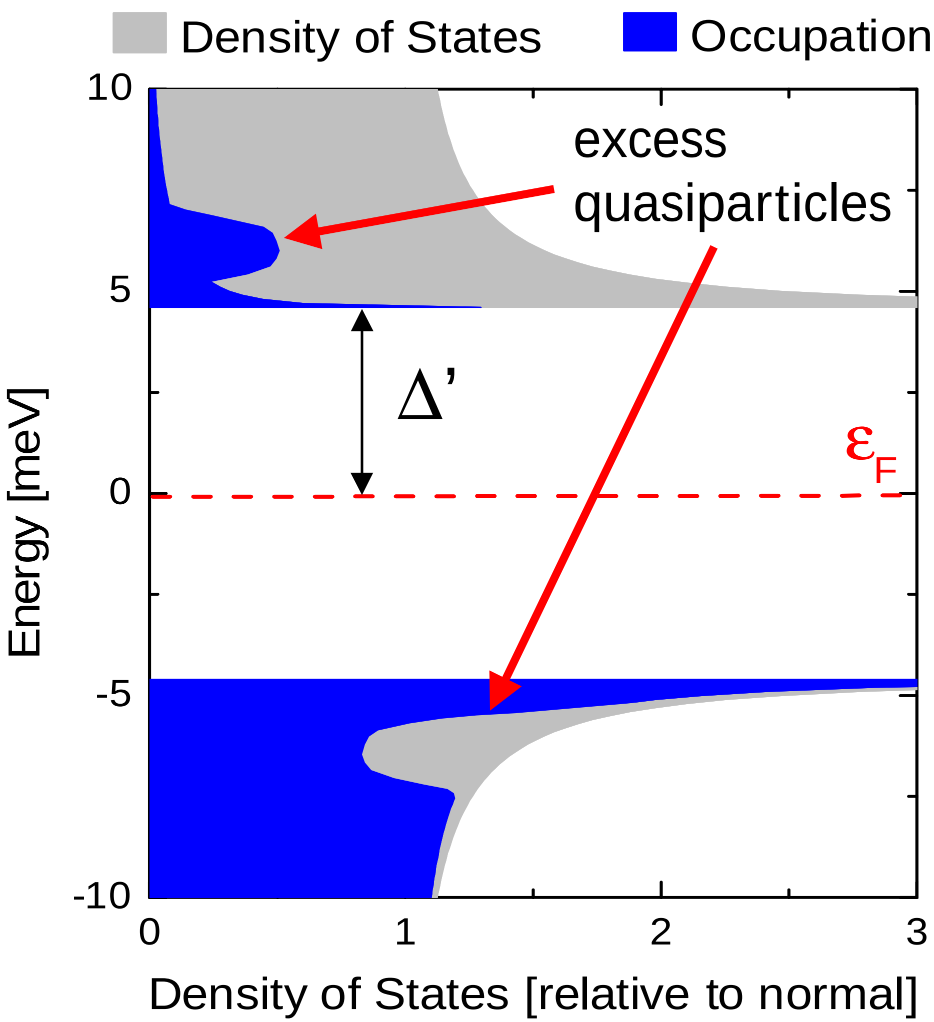
Photoexcitation in a BCS Superconductor



Step 3:
Excess quasiparticles recombine to form pairs and gap is restored to full value.

Relaxation time for thin films of BCS-type superconductors is ~ 1 ns.

Photoexcitation in a BCS Superconductor



New Experiment:

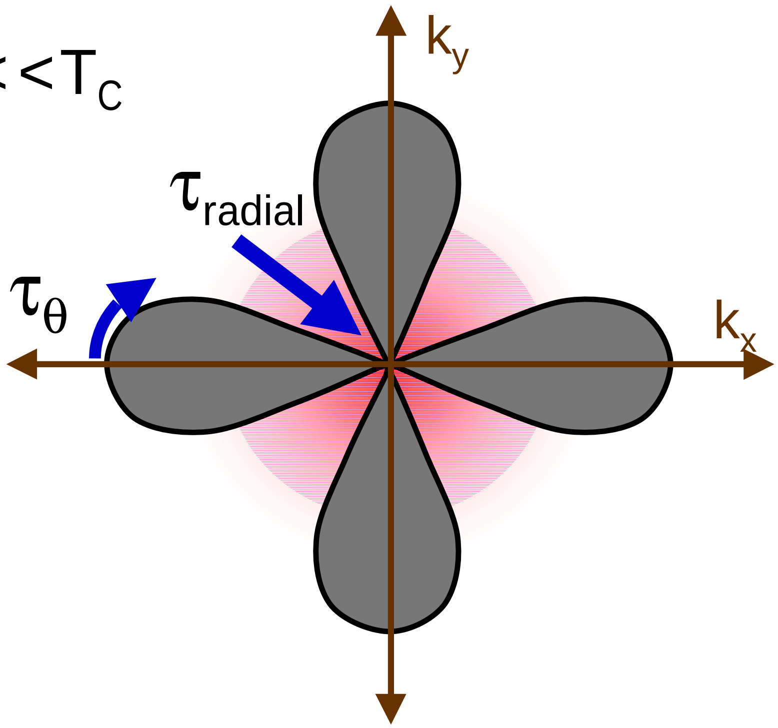
Use narrow-band, low energy pump to impose non-thermal initial distribution, probe with broadband pulse (spectroscopy).

How do these quasiparticle relax toward gap edge. How does this population affect the overall superconducting state and the energy gap?

Pump-Probe THz Spectroscopy of a d -wave Superconductor

Near-IR laser excitation
and fast initial relaxation

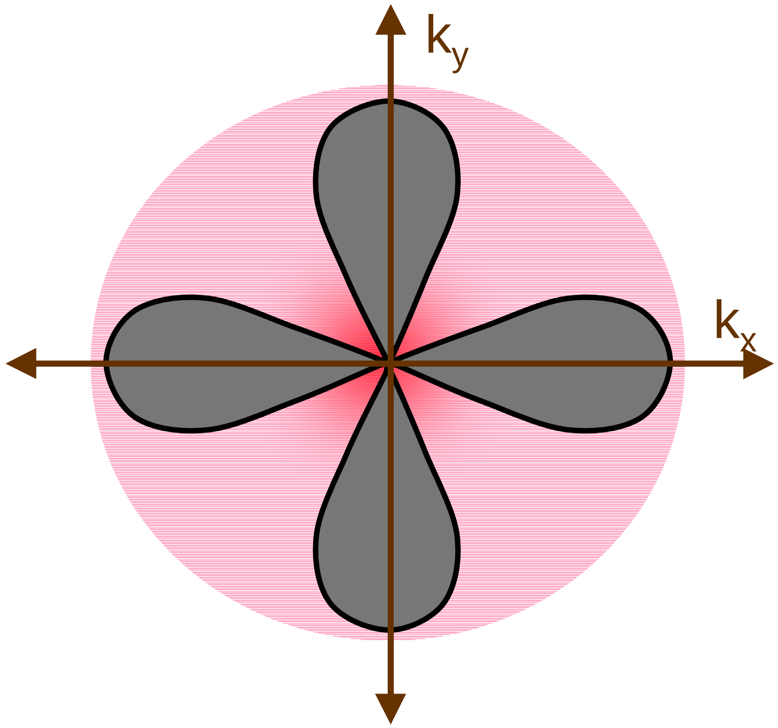
$T << T_C$



$\tau_\theta >> \tau_{radial}$

J. Orenstein et al, PRL UC Berkeley
P.J. Hirschfeld et al, PRB U. Florida

Narrow-band THz excitation
to limit starting population energy

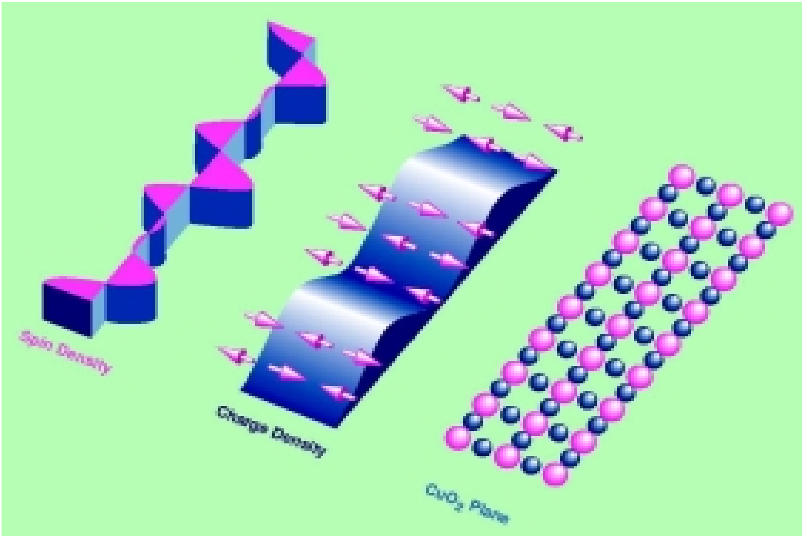


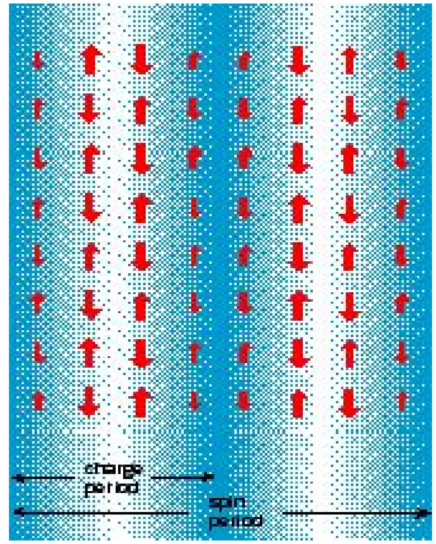
Goal: study τ_{relax} as a function of E_{pump} (pump photon energy)

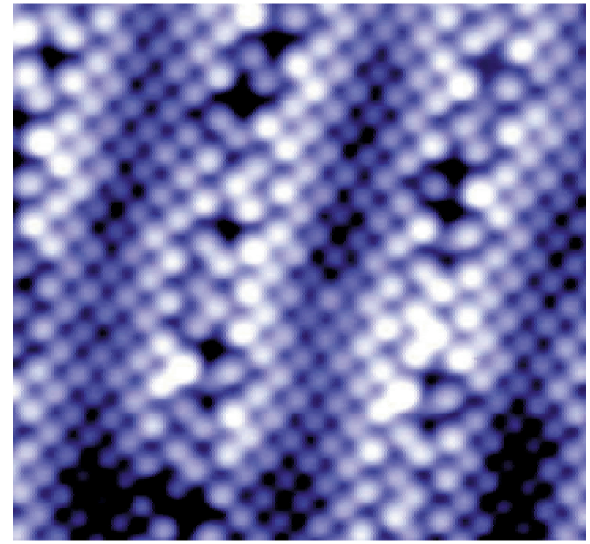
THz Pulse-driven Superfluid Excitations

- Can a THz pulse induce currents that exceed the critical current in a superconductor?
  - A new type of transient excitation (avoids complicated heating effects for a DC experiment).
  - What does this superconductor look like? Is there any evidence for an energy gap or other indication of superconductivity?
  - How does it recover/relax?
- Estimates for the necessary E-field:
  - $E = J/\sigma = 10^8$ A/cm^2 /1000 [Ω-cm]$^{-1}$ = 10^5 V/cm or 100 kV/cm.
- Experiment: Pump with high field THz pulse (low pass filter), probe with lower intensity, broader spectral coverage pulse.

Dynamics of Charge / Spin Stripes in Oxides







Tranquada *et al*., Nature (1995)

J E Hoffman et al. 2002 Science 295 466

Stripes not visible in many materials: dynamic fluctuations?

Can a high-field, half-cycle THz pulse temporarily induce a preferred orientation?

Goal: THz pump, polarized THz probe to sense induced anisotropy & relaxation.

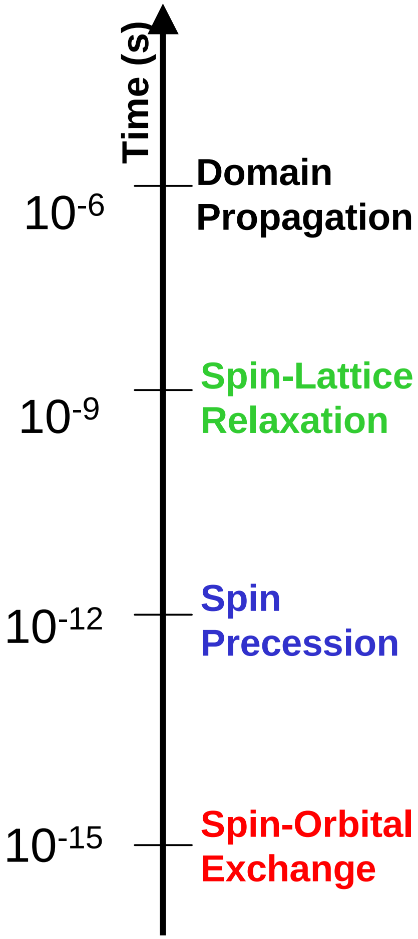
Requirements:

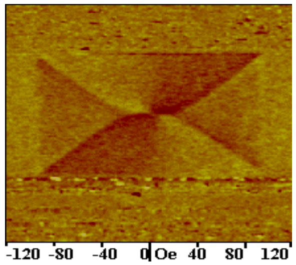
Intense source of half-cycle E-field THz pulses (how big?)

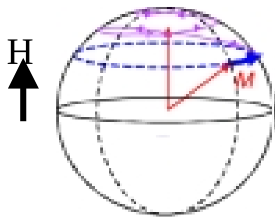
THz spectroscopy probe (polarized)

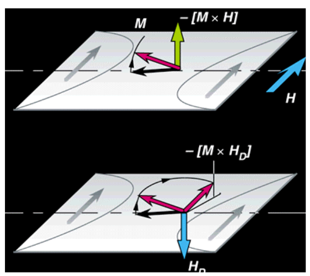
THz Driven Magnetic Dynamics

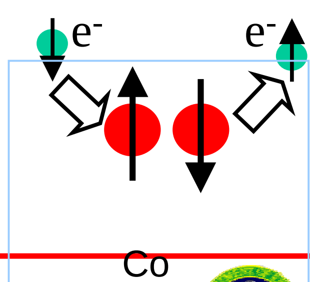
*Use ultra-short magnetic field pulses to induce spin excitations (**D. Arena / NSLS**)*





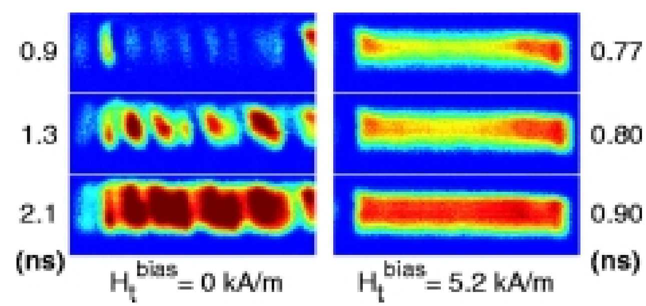






Excitation / Interaction	Timescale (sec)
Exchange interaction	10^{-15}
Stoner excitations	10^{-15} - 10^{-14}
Spin waves	10^{-12} (low q limit)
Spin – lattice relaxation	10^{-12} - 10^{-11} (in manganites)
Precessional motion	10^{-10} - 10^{-9}
Spin injection	TBD
Spin diffusion	TBD
Spin coherence	TBD

Soft Ferromagnet Dynamics Time-resolved MOKE on permalloy strip. B.C. Choi *et al.*,PRL **86**, 728, (2001)



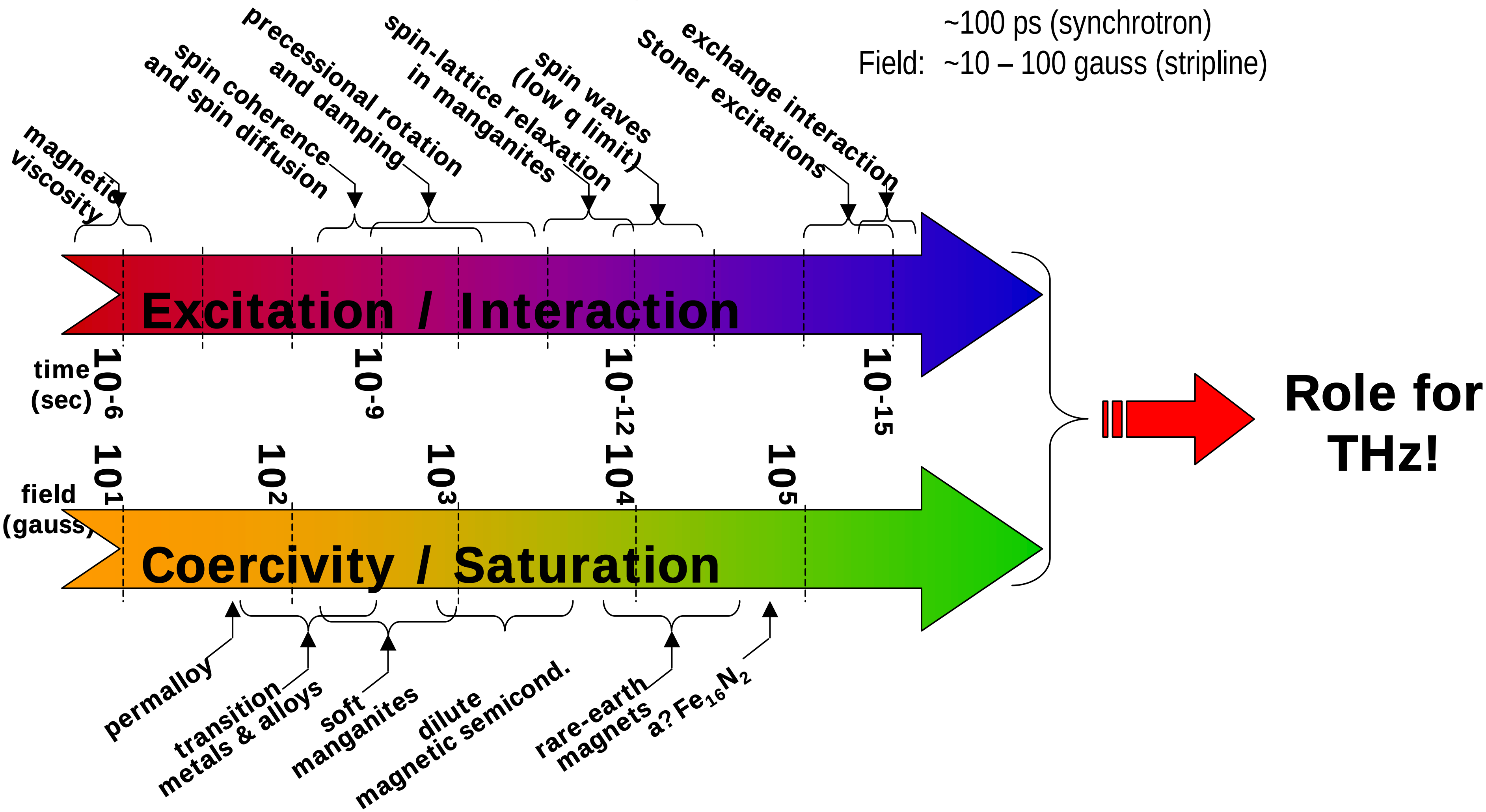
Other systems of interest: Dilute Mag. Semiconductors, Manganites.

Opportunities in Magnetism with THz Pulses

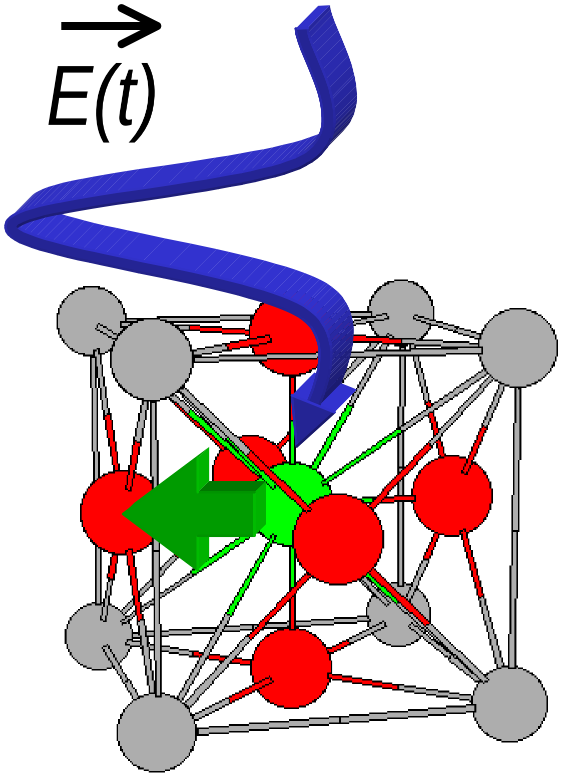
Ultra-Short Pulses and/or High Fields -- D. Arena / NSLS

Current state of the art for “ultra-fast” dynamics experiments:

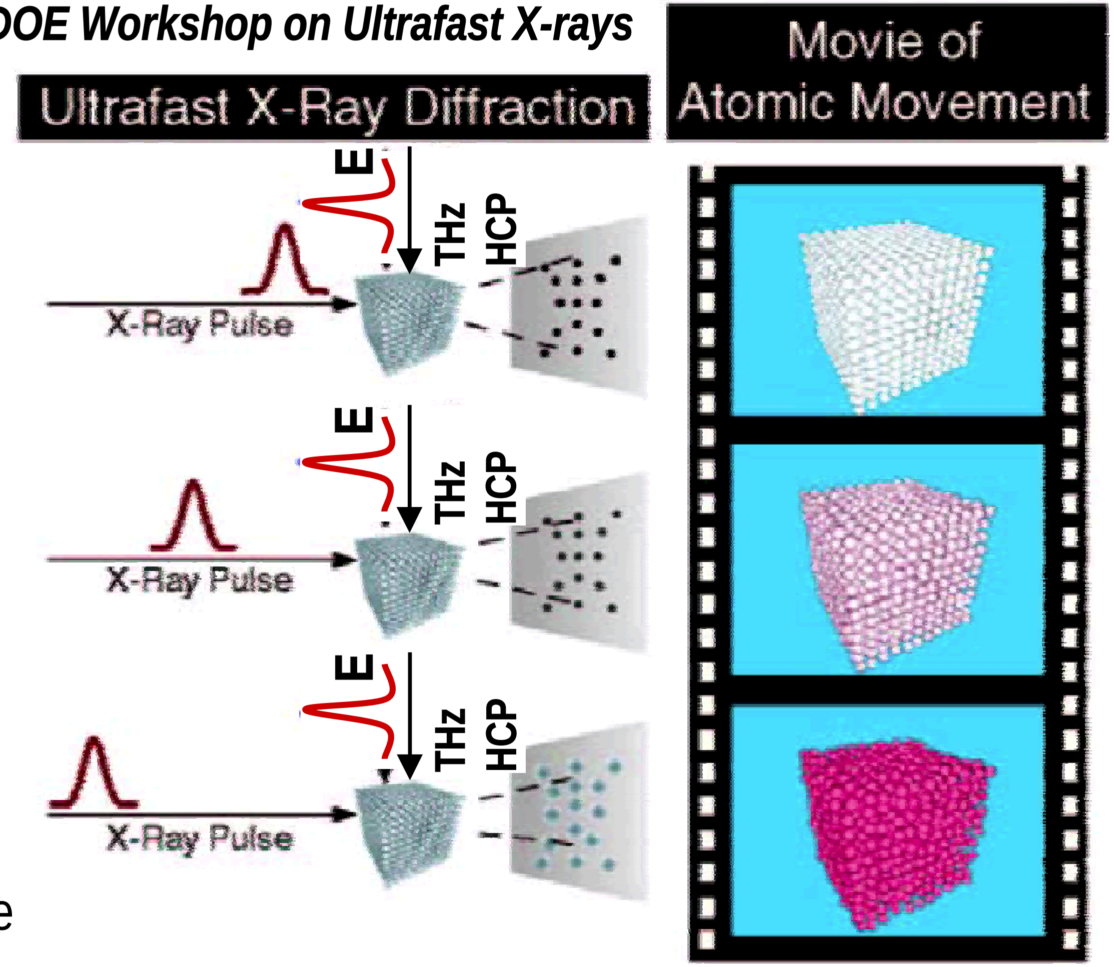
Time: ~100 fs (lasers)
~100 ps (synchrotron)
Field: ~10 – 100 gauss (stripline)



Soft Modes in Ferroelectrics & Perovskites ($PbTiO_3$)



DOE Workshop on Ultrafast X-rays



Use half-cycle pulse to coherently drive atoms, probe motion as a function of time (needs diffraction probe).

Observe "shift" in diffraction spot(s)

Summary

- Per particle, relativistic electrons offer a significant intensity/energy advantage over conventional pulsed THz sources.
  - *80 μJ per pulse, E-field approaching 1 MV/cm demonstrated at DUV-FEL/SDL.*
  - *High pulse energy should be sufficient to create novel coherent excitations in solids.*
  - *Radial polarization of transition radiation may be useful for coupling into coaxial guides.*
- Characteristics compatible with EO detection methods.
  - *Necessary for coherent imaging technique*
  - *Single-shot spectroscopy (chirped sampling pulse) desirable.*
- Spectral content determined by electron beam density.
  - *Can we make two colors? What shapes can be achieved?*
- Existing accelerators producing coherent THz pulses were not designed for this purpose.
  - *Opportunity to optimize parameters for greatest flexibility versus cost/size.*
  - *Shortest achievable bunch length versus charge, versus energy?*