Terahertz Dynamics of Materials in Strong Fields

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T. Jefferson National Accelerator Facility
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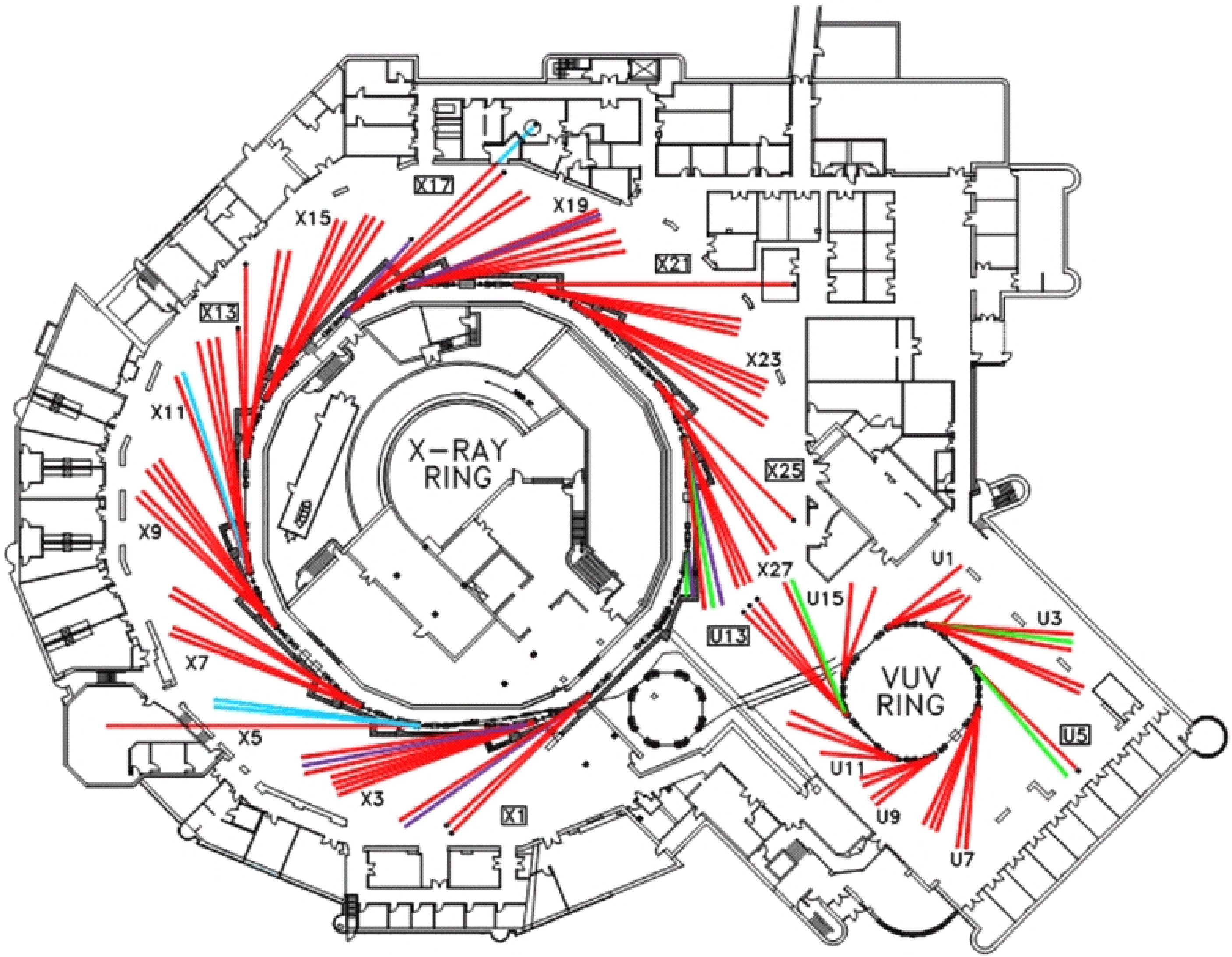
Outline

- Infrared / THz facilities & science at the NSLS VUV ring
  - *beamlines for incoherent synchrotron radiation, synchronized laser*
- THz spectroscopy as a probe of materials
  - *superconductivity*
    - magnetic fields (mixed state with vortices)
    - dynamics (laser pump, THz probe)
  - *mesoscopic carbon / graphene*
    - bandstructure, “Dirac” quasiparticles
    - Landau levels, magnetospectroscopy
    - graphene nanoplatelet films
    - non-linear optical (THz) material?
- THz pulses from Source Development Lab Linac
  - *sub-picosecond pulses, >1nC, strong EM fields (~ MV/cm)*
  - *dynamical EO effect*

NSLS Accelerator and Beamline Complex



VUV/IR Ring Operating Parameters

E = 800 MeV

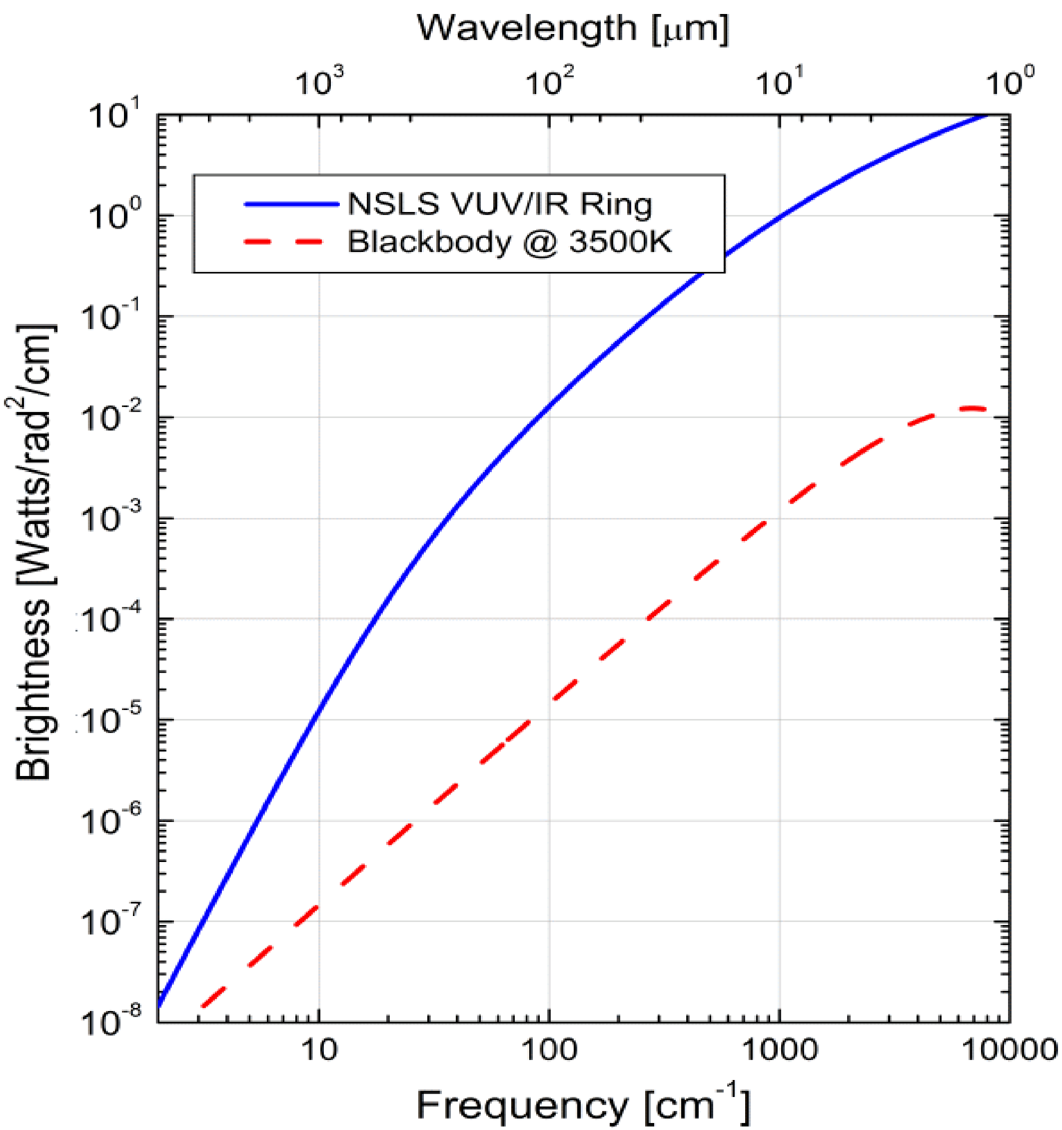
DBA w/ 4 super-periods, ρ = 1.91 m
4 straights (2 for IDs)

I (avg): 670 ma

RF @ 52.9 MHz

σ_{BL}: 850 ps down to 130 ps

Dipoles allow large aperture ports
(needed for very Far IR & THz)

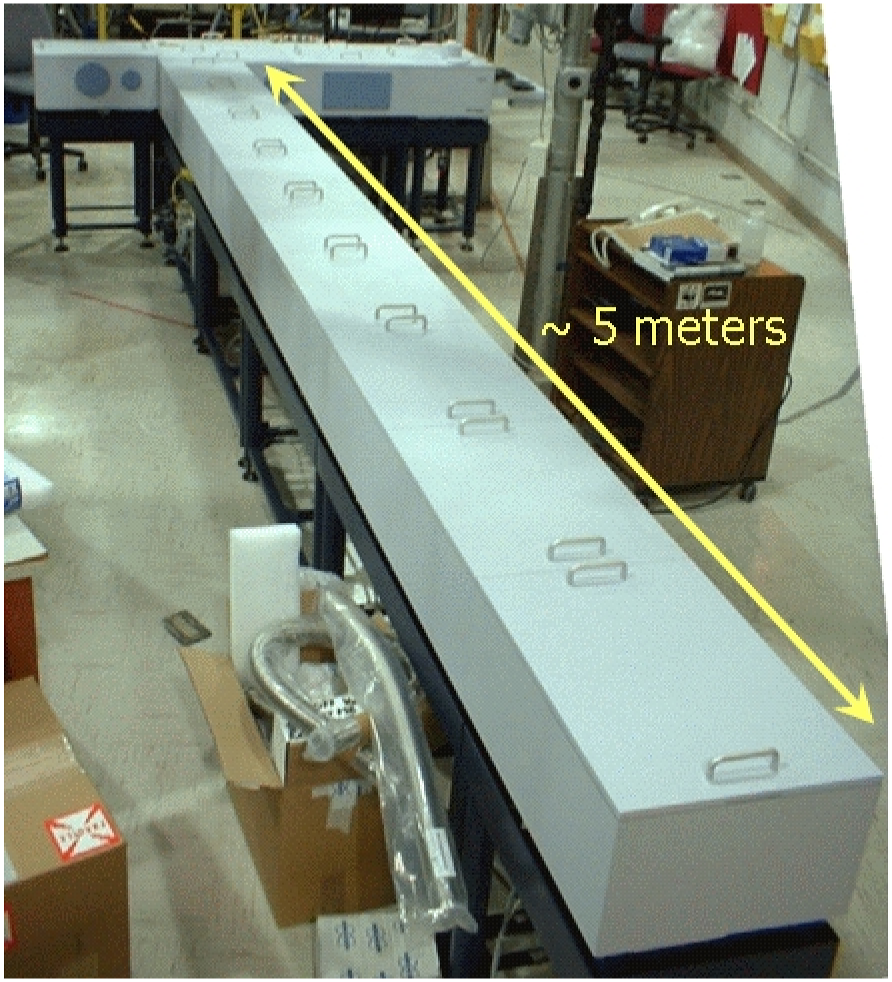


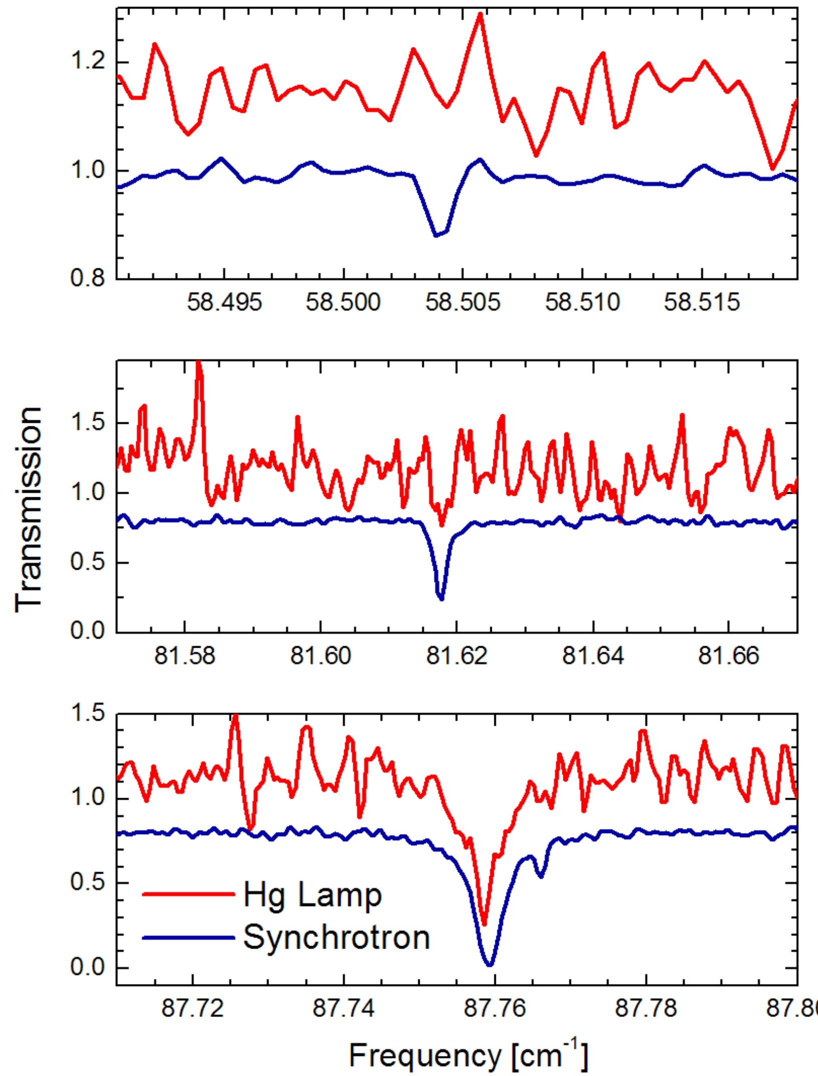
note: wavenumber is just 1/λ with λ in cm, 8066

U12IR: Extreme Resolution Spectroscopy

NSLS / Stony Brook Univ. / U. Florida

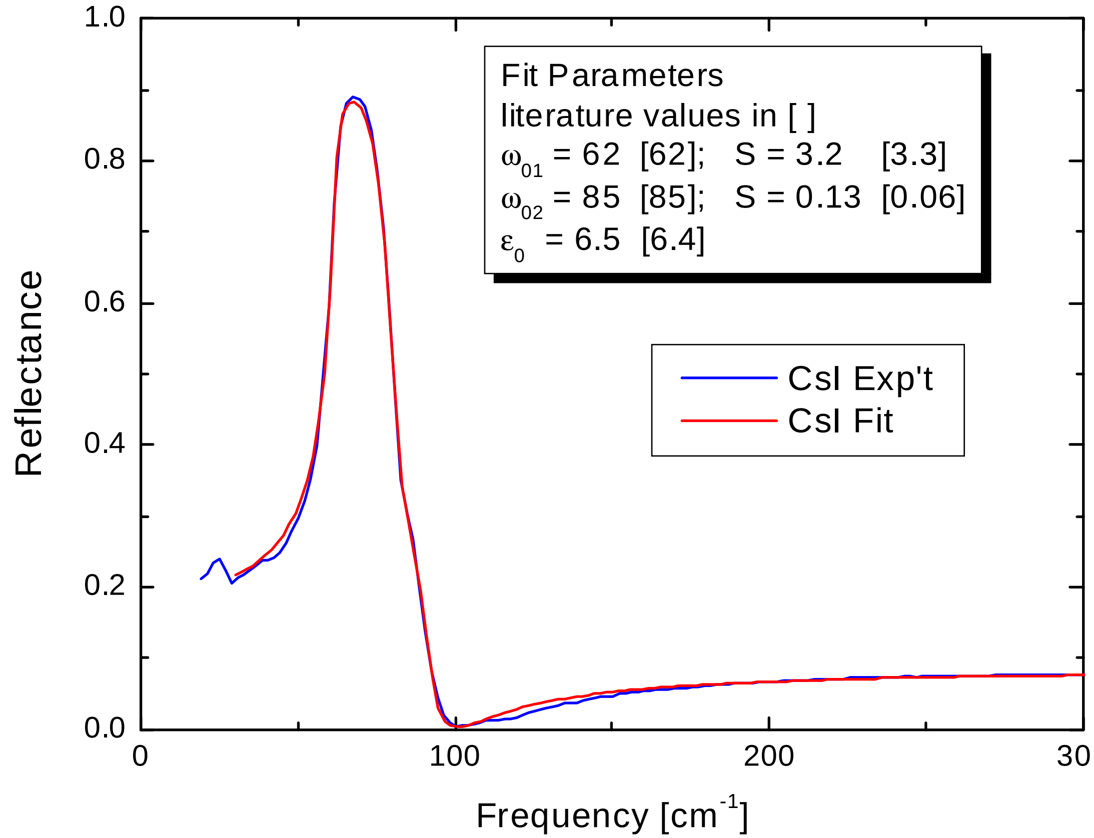
Bruker IFS 125HR, res'n 0.001 cm^{-1} (125 neV)





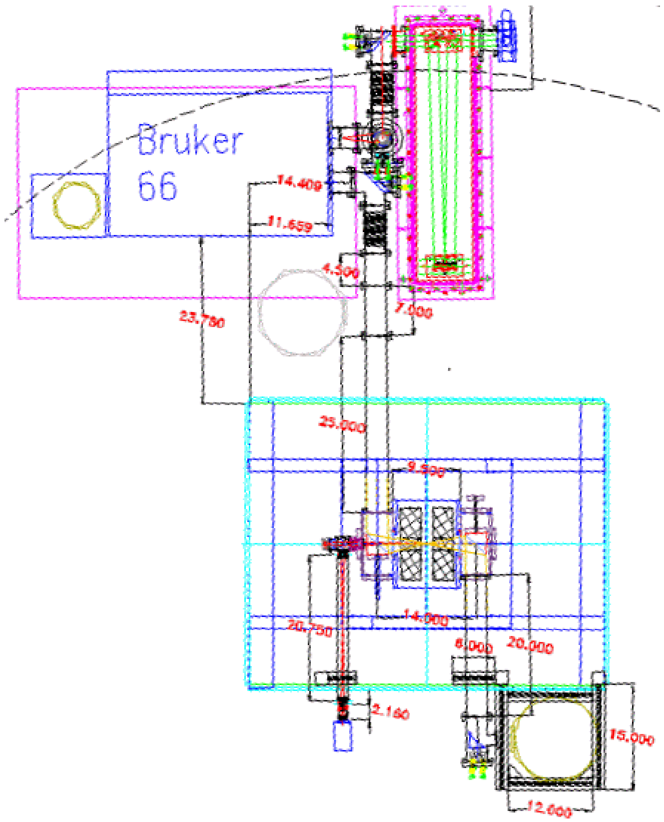
U10A: Far-IR Microspectroscopy

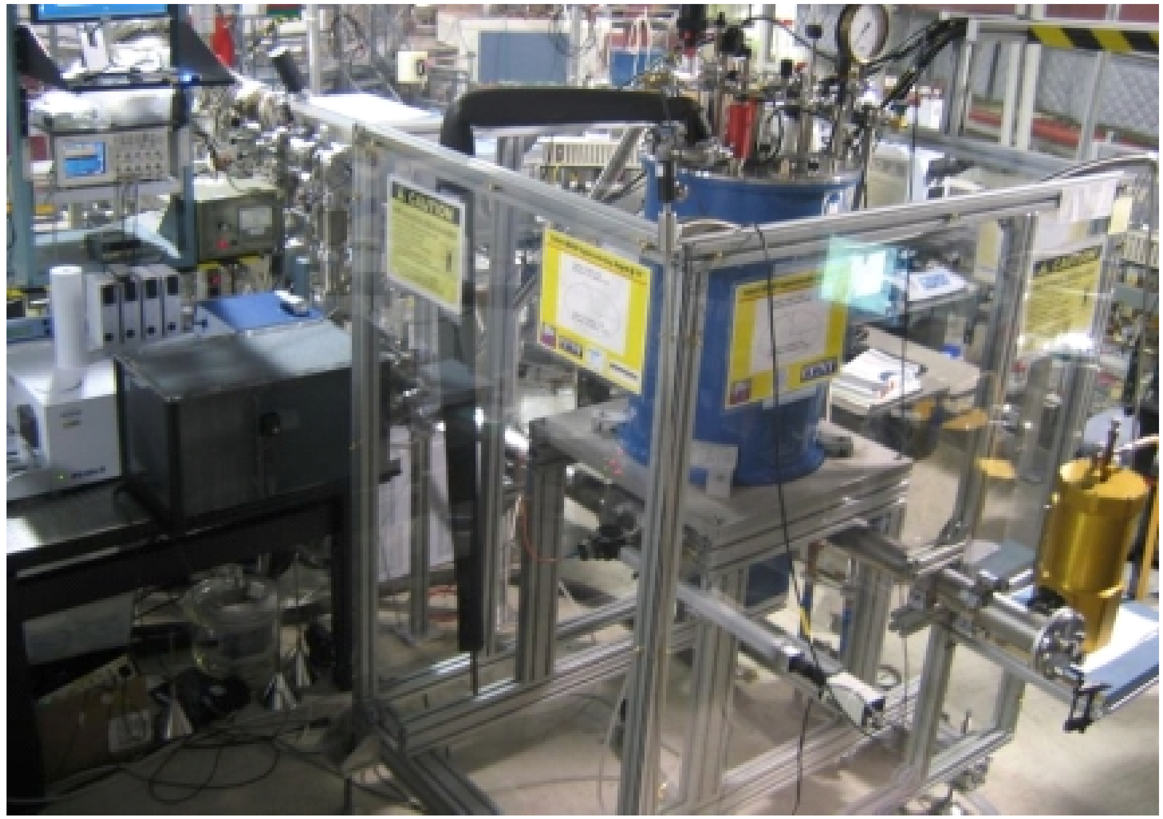
- Spectral coverage down to ~ 25 cm^{-1} (just below 1 THz)
- Used for NASA Stardust comet particle analysis.
- "Convenient" microscope operation, but larger aperture optics are needed for efficient and optimal performance down to 10 cm^{-1}.



Magnetospectroscopy at U4IR

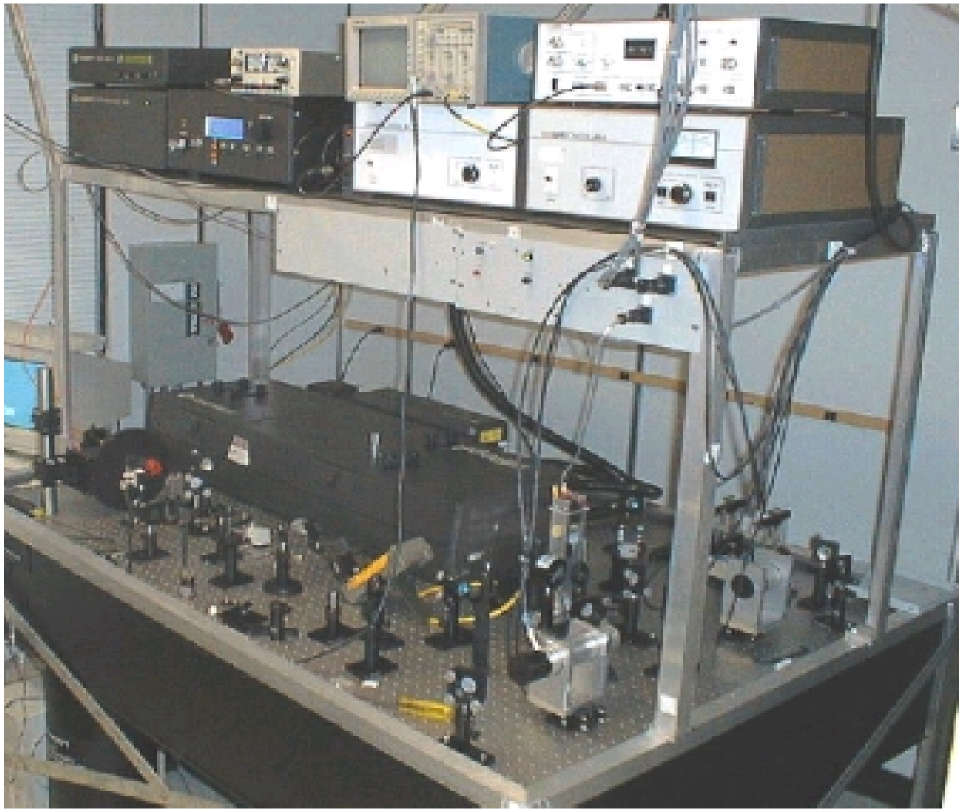
- Oxford *SpectroMag* 10T Split Coil installed in FY' 07
- Large NA access along bore, 8 cm^{-1} to 20,000 cm^{-1}
- Fiber optic feed for laser-based pump-probe.

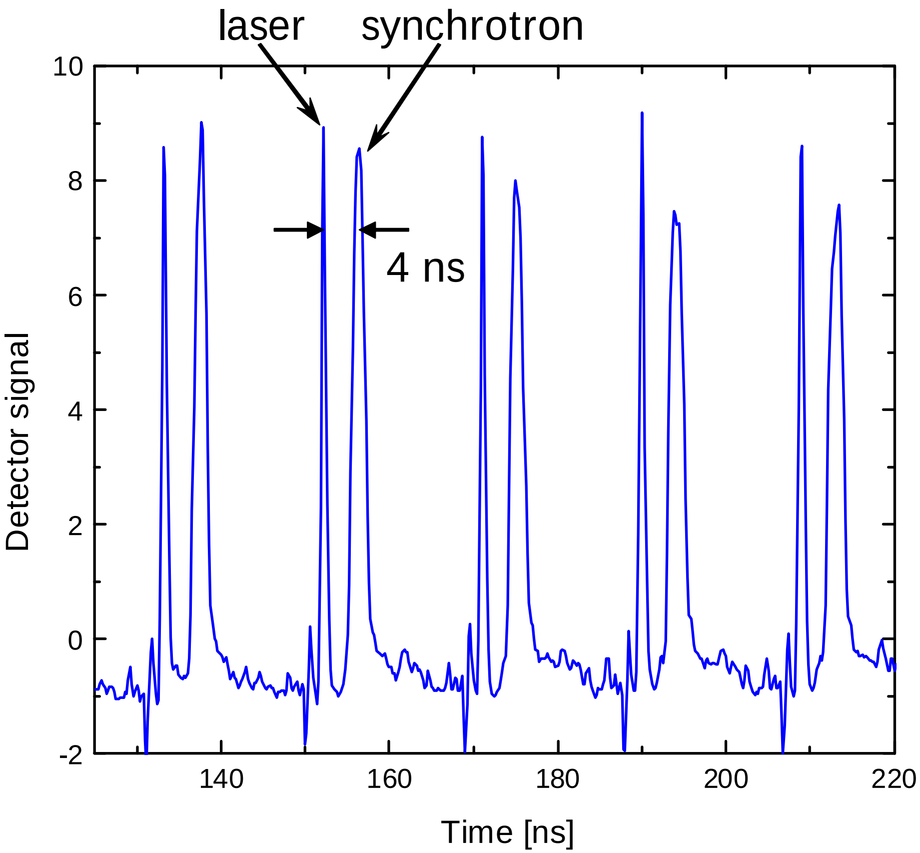




NSLS VUV Ring & Synchronized Ti:sapphire Laser

- VUV/IR Ring: 53 MHz RF, ~ 300 ps duration synchrotron pulses.
- Coherent Mira 900P (~ 2 ps pulses), synchronized storage ring RF
- Application: time-resolved (pump-probe) IR spectroscopy



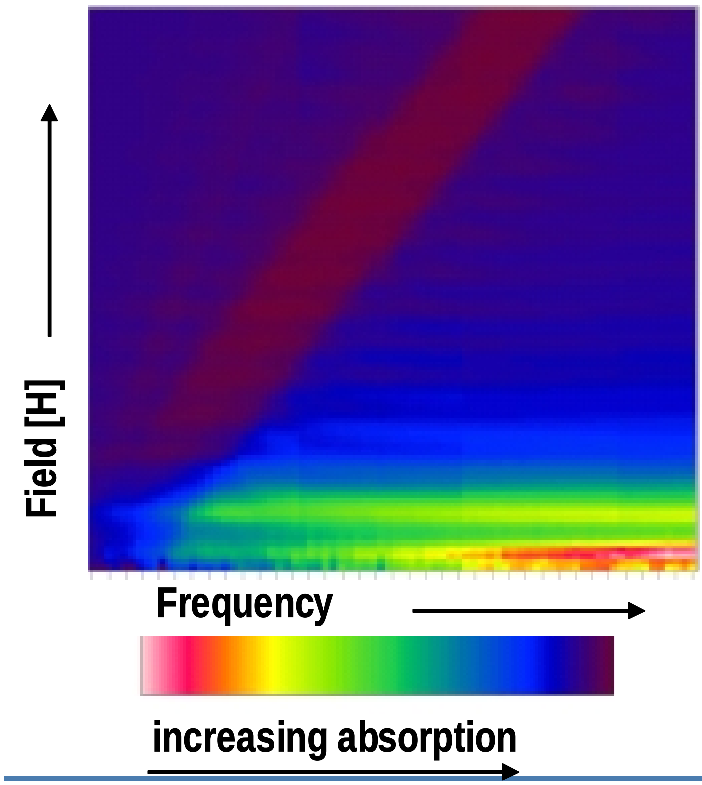


Science: Magnetospectroscopy

Magnetic-field induced M-I transition

- lightly doped InSb at low temperature

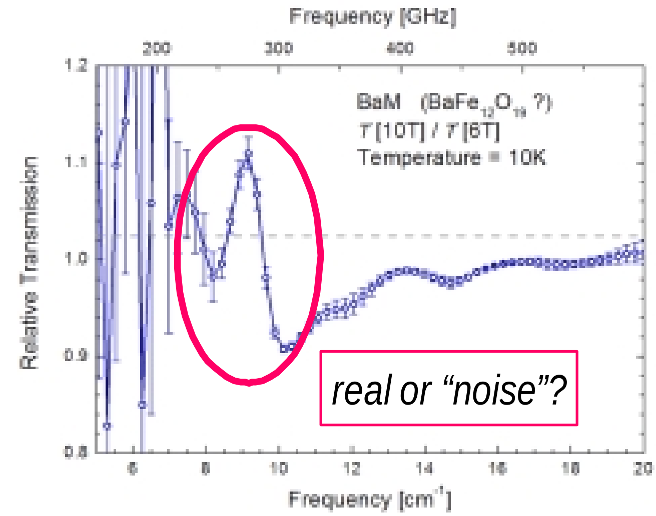
S. Cox, J. Singleton & S. Crooker (LANL)
M. Klopf & G.P Williams (JLab)



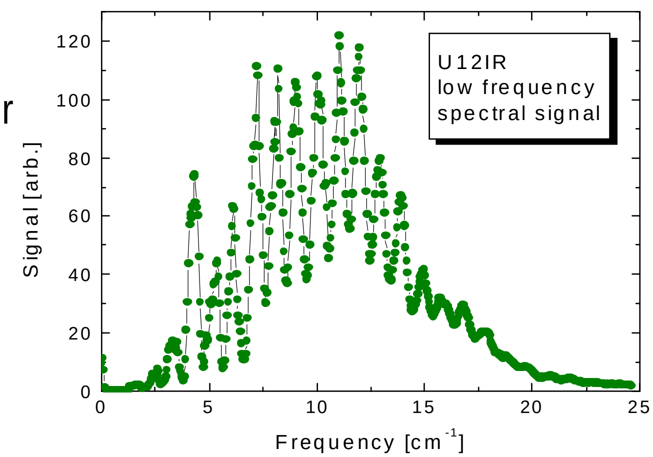
Ferro/ferrimagnetic resonance in Ba hexaferrite

- goal: resonance width to determine intrinsic speed of response.

B. Guralnick, V. Harris (Northeastern)
D. Arena, G.L. Carr (NSLS)



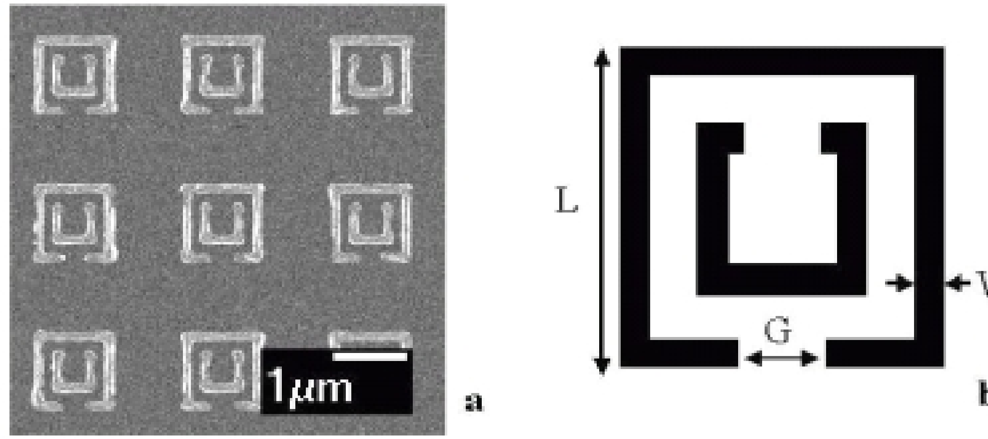
NOTE: lower frequencies can be achieved with proper optics & spectrometer, but NSLS dipole chamber design results in strong intensity fringes. Not static!



Science: Metamaterials

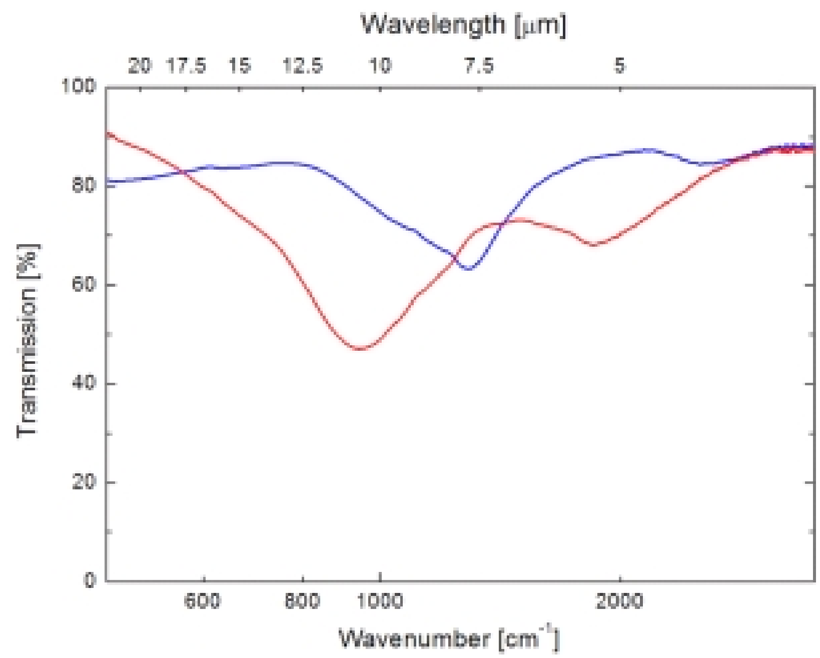
Arrays of split-ring resonators to create both negative dielectric function (ε) and magnetic permeability (μ) to achieve negative refractive index n. (so-called left-handed materials).

J. Wang, T. Timusk (McMaster Univ.)



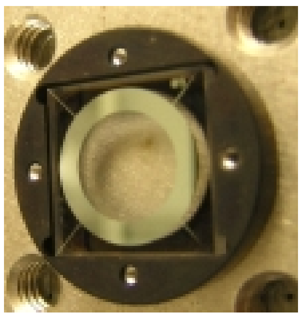
Measured transmission of a 100 μm by 100 μm SRR array using polarized infrared (orientation relative to the resonator gap showing shift in resonance frequency.

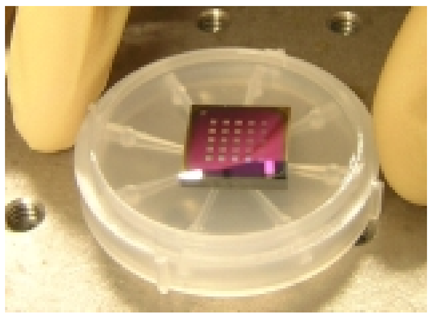
Beamline U10A.

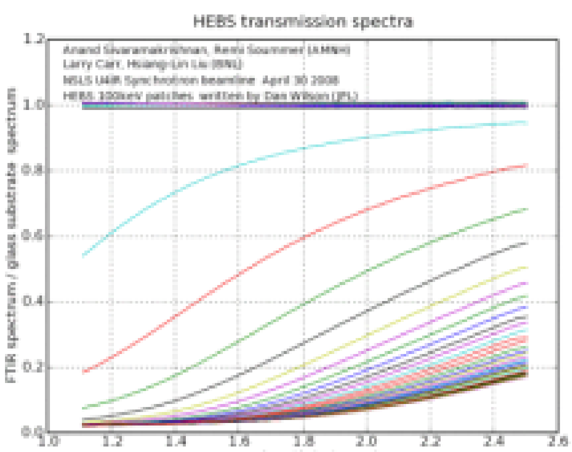


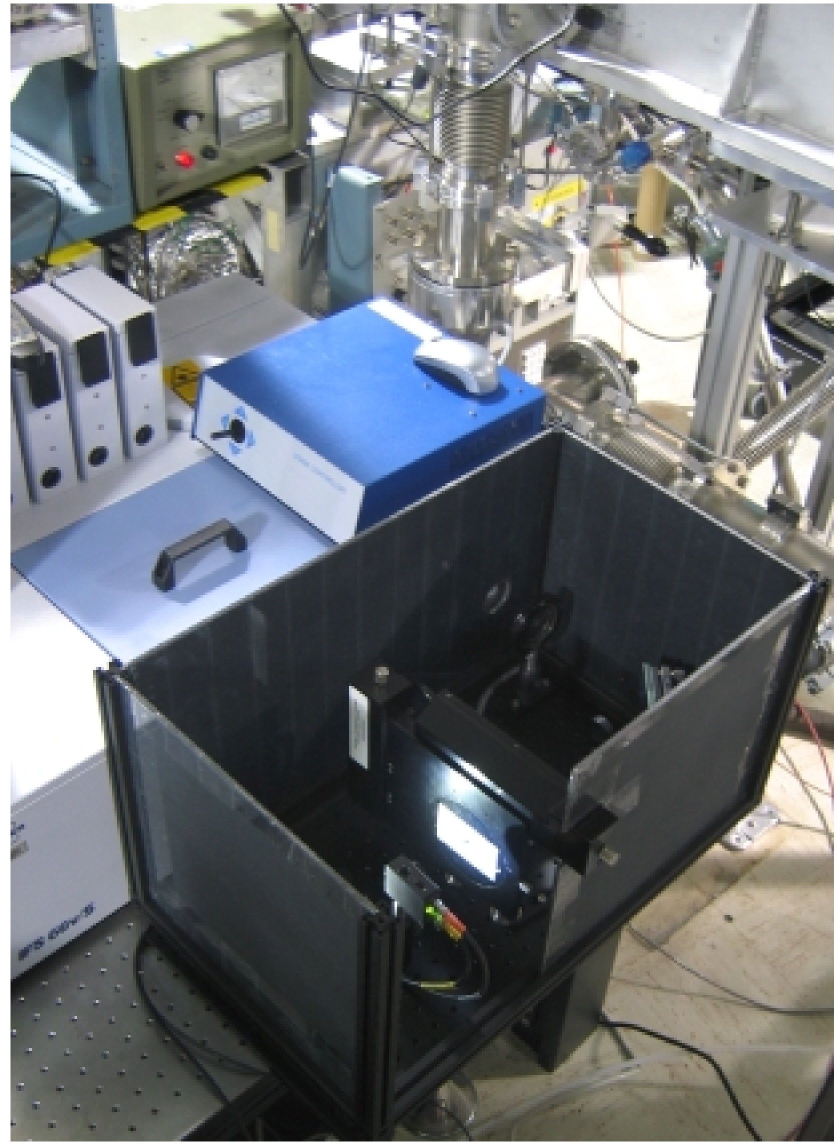
Apodization Filters for Extreme Adaptive Optics

- A. Sivaramkrishnan (AMNH & Stony Brook)
- Instrumentation for large telescope facilities involved in search for extra-solar planets.
- scanning near-IR spectroscopy at f/100 and 200 μm spotsize => near diffraction-limit









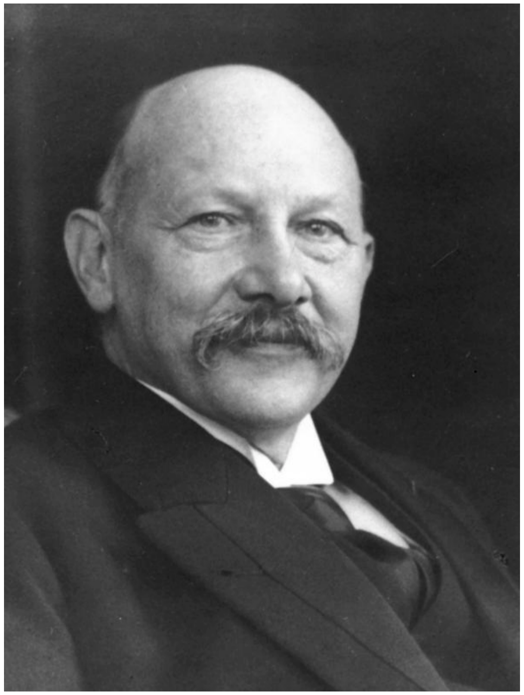
Superconductivity

- H. Kammerlingh Onnes (Leiden)
  - liquifies He in 1908 (this July is 100th anniversary).
  - discovers superconductivity in Hg in 1911.

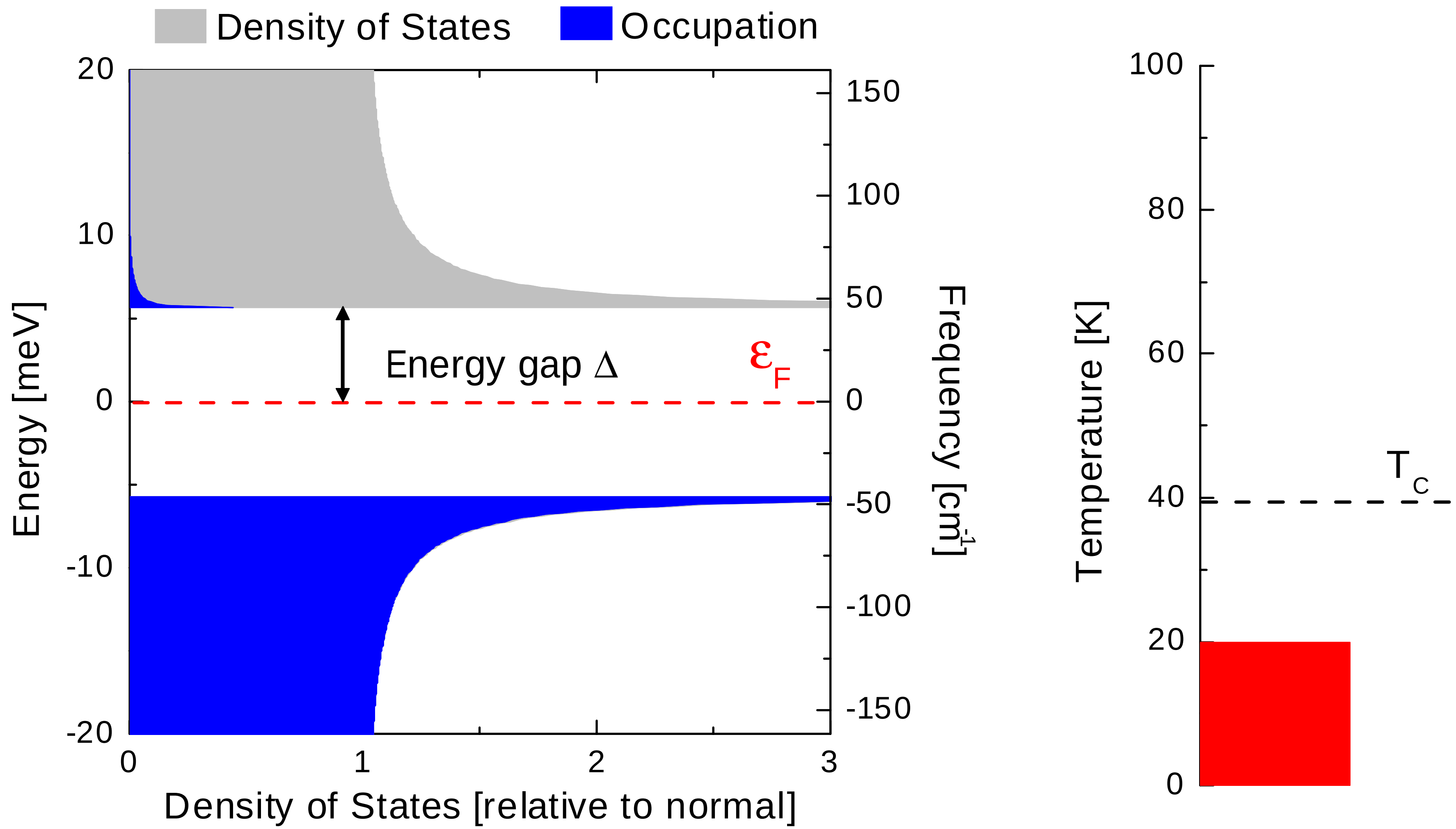
- Ordered/coherent quantum state of paired electrons.
  - pairing results from boson-mediated attractive interaction (e.g., exchange of phonons) … or it's not?

- Order parameter $\psi = n^{1/2} e^{i\varphi} \sim \Delta\, e^{i\varphi}$
  - energy gap appears in electronic density of states Δ for $T<T_c$.
    - gap indicates strength of superconducting state.
  - phase φ describes voltage and currents
    - $V \sim d\varphi/dt$ while $\boldsymbol{J} \sim \nabla\varphi$.

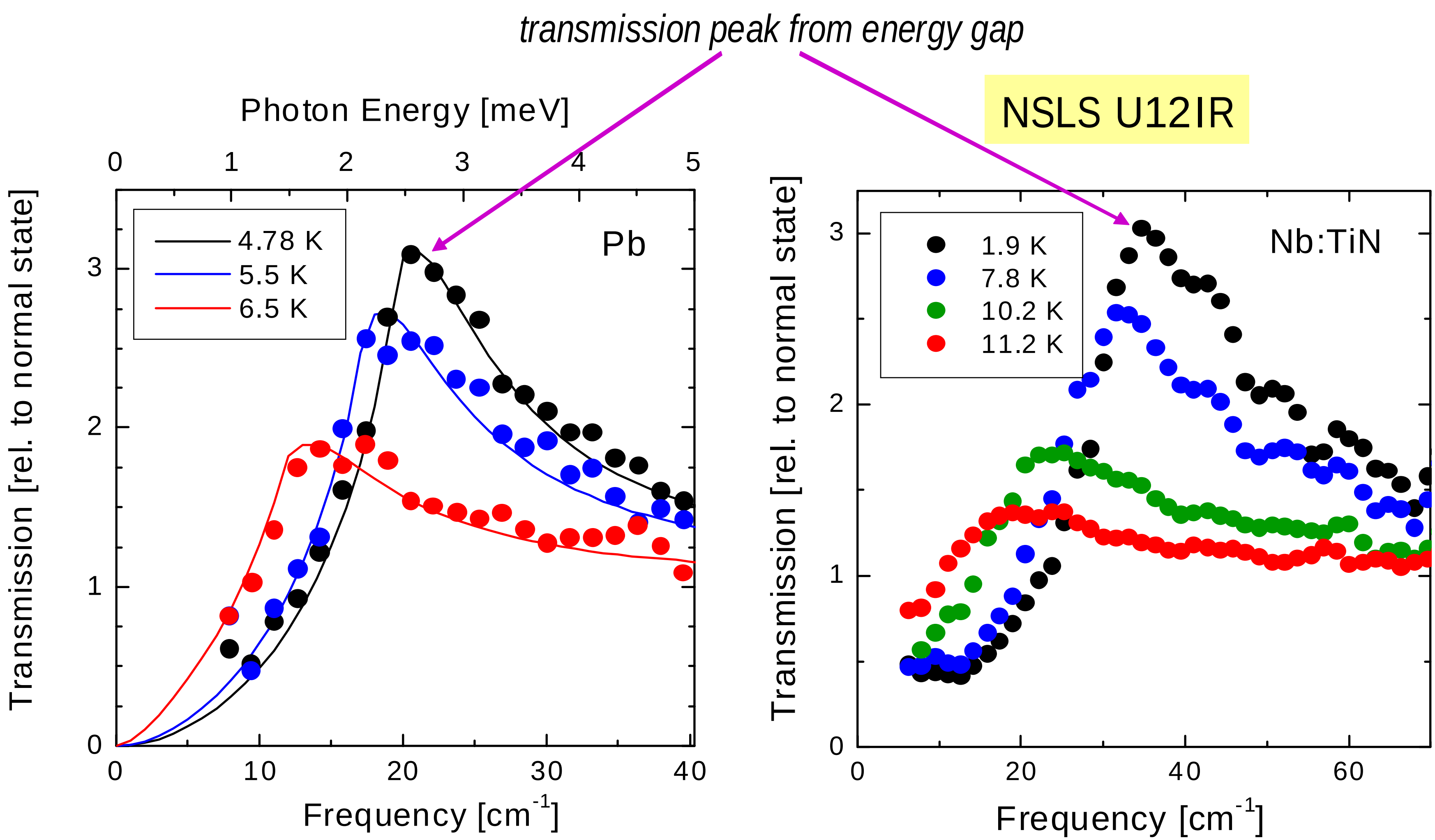
- Energy gap Δ for many materials ranges from <1meV to >10 meV
  - Far-infrared or "Terahertz" spectral range



BCS Superconductor



THz / Far-infrared & Superconductors



Superconductors (type II) in a Magnetic Field

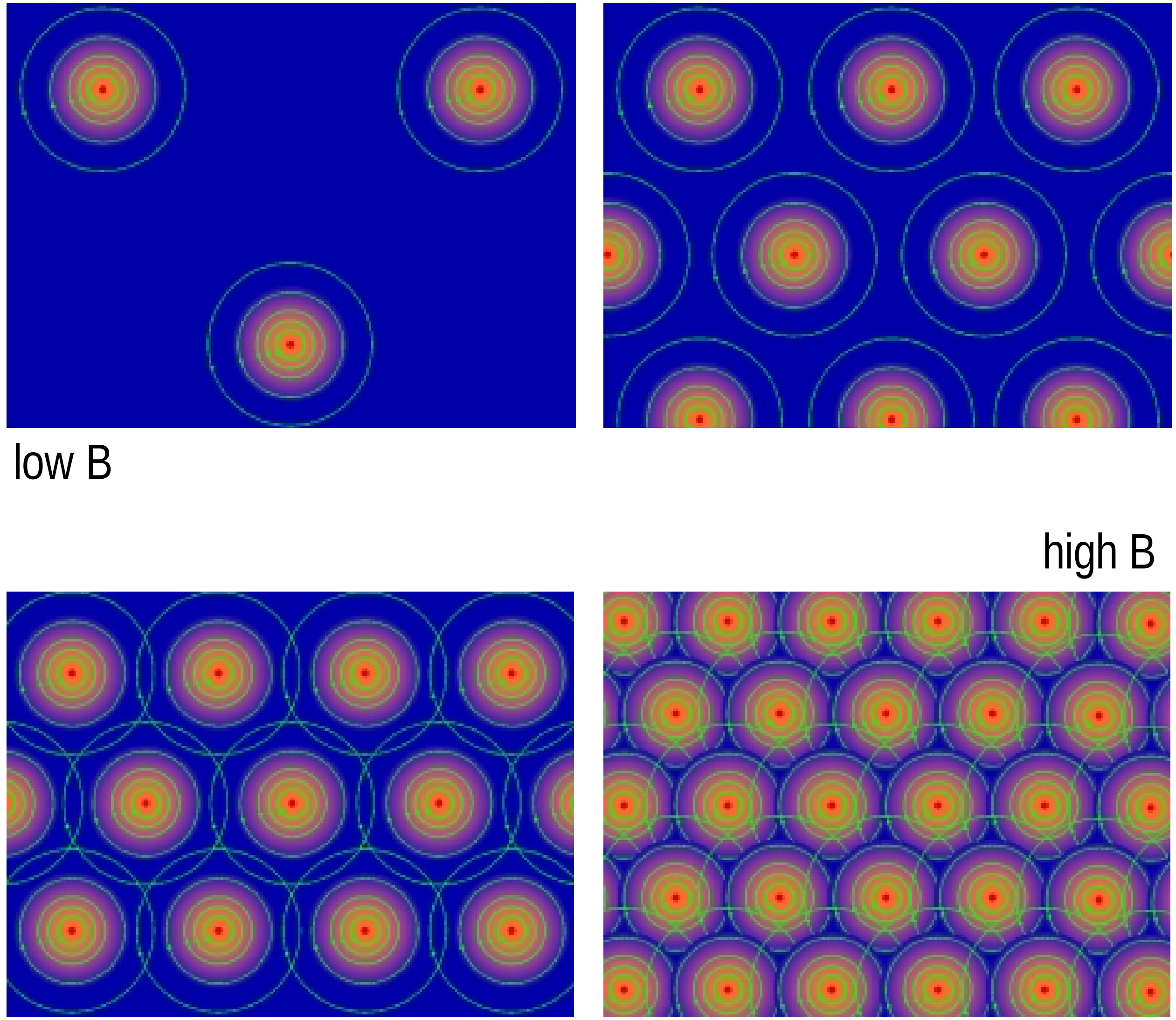
Field penetrates in form of an array of <u>fluxoids</u> (vortices). $\delta\varphi = 2\pi$

Each vortex has single flux quantum ($\Phi_0 = h/2e = 2.1\text{x}10^{-15}$ Wb)
=> vortex areal density increases linearly with applied field.

Vortices have a "normal" core of dimension ξ (coherence length). Upper critical field reached when vortex spacing ~ξ.

Magnetic field is screened over a scale λ_L (London penetration depth), often $\lambda_L > \xi$ (type II).

<u>Result:</u> Have mixed phase of normal metal & superconductor, with SC component "weakened" by penetrating magnetic field.

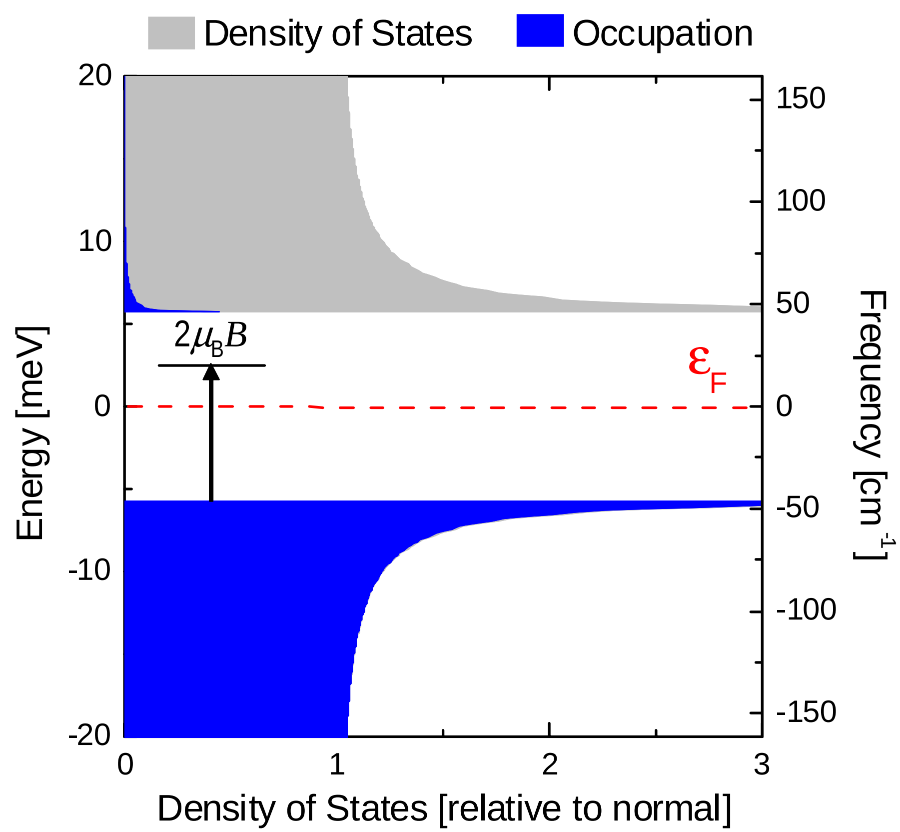


Photon absorption by pair-breaking in a Magnetic Field

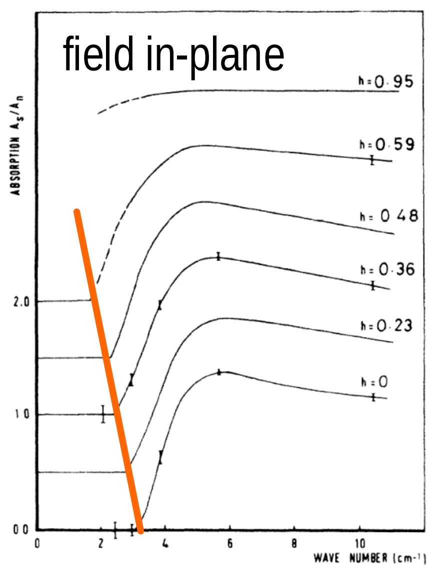
Cooper pairs are comprised of spin-up / spin down combination. ↑↓

Energy to flip electron spin from anti-aligned to aligned in field B is $2\mu_B B$ = 0.93 cm^{-1}/T

=> can break pair with $\hbar\omega < 2\Delta$ when anti-aligned spin flips. Effective gap edge shifts down by $2\mu_B B$.

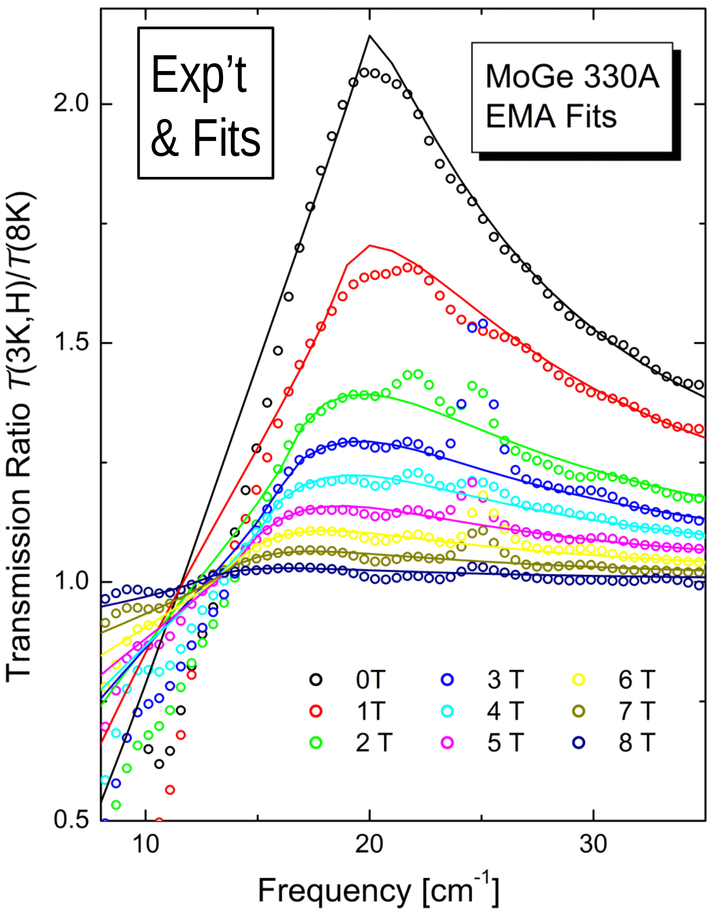


van Bentum & Wyder,
Phys. Rev. B **34**, 1582 ('86)



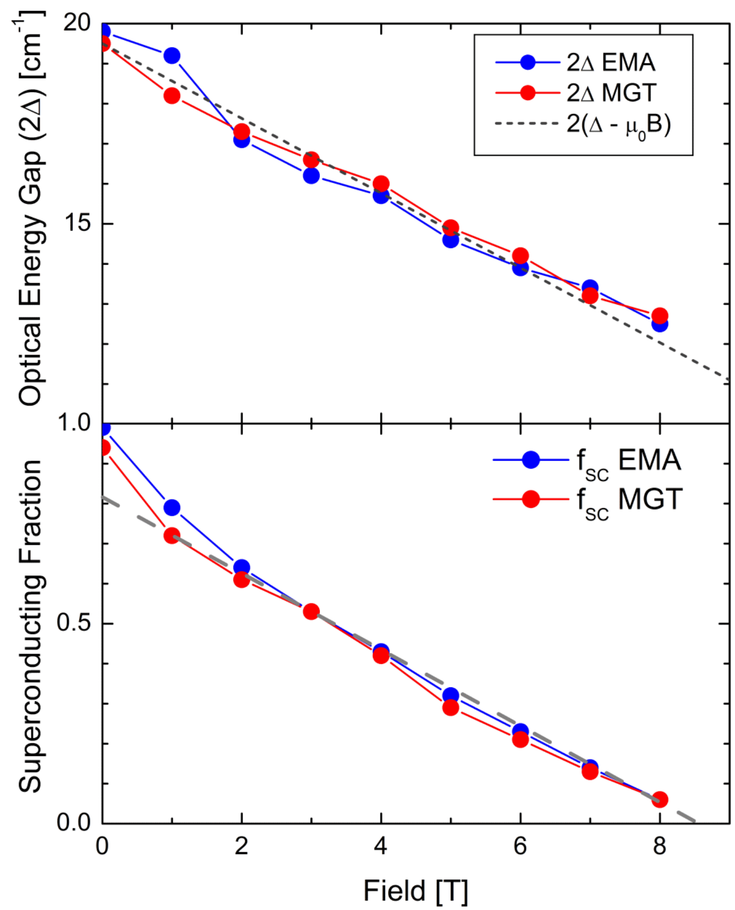
MoGe superconductor and Effective Medium Analysis

Effective medium theory for calculating electrodynamic reponse of a two-component system: superconductor and normal metal.



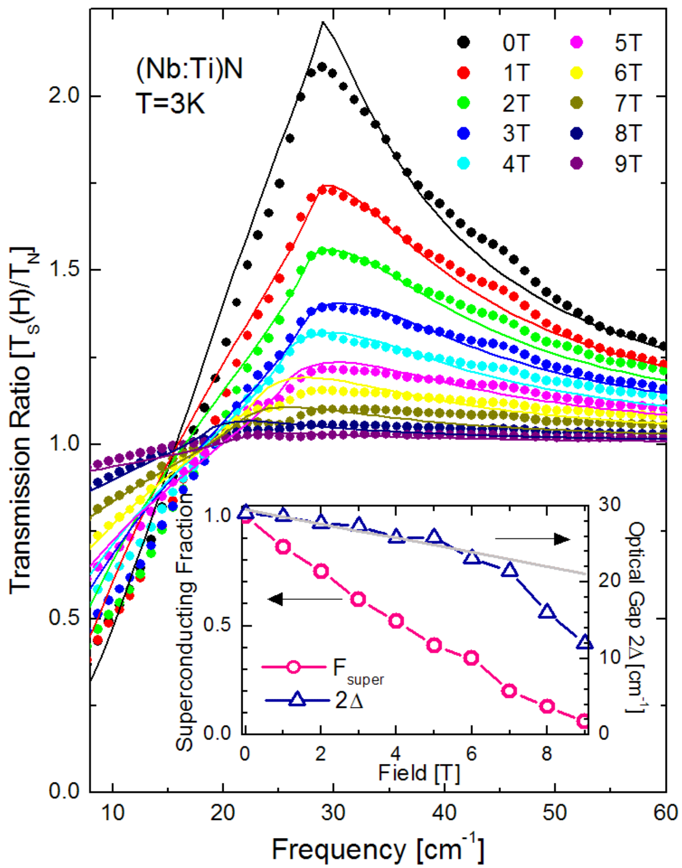
Gap shrinks due to field-assisted pair-breaking

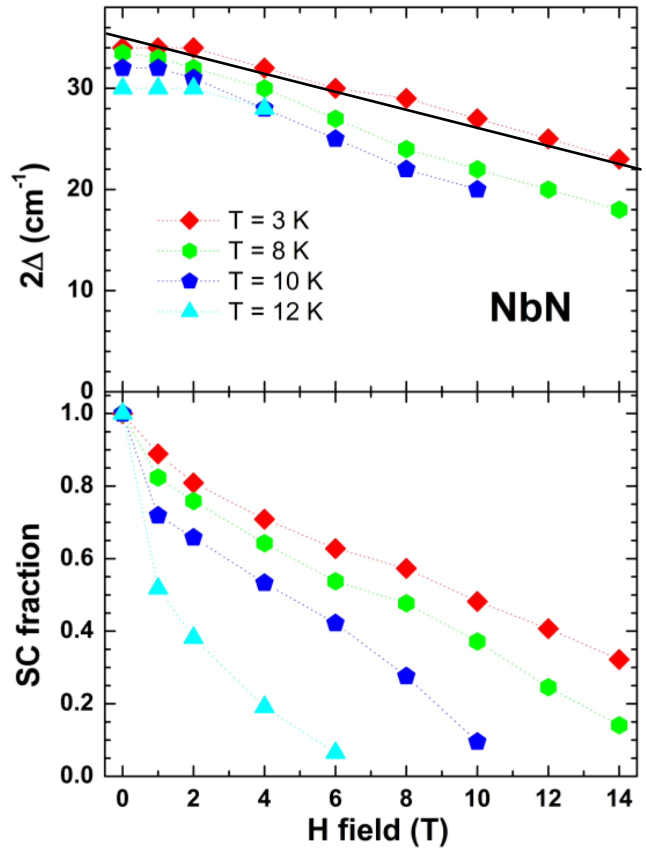
Superconducting fraction shrinks linearly with field



(Nb:Ti)N and NbN in Magnetic Field

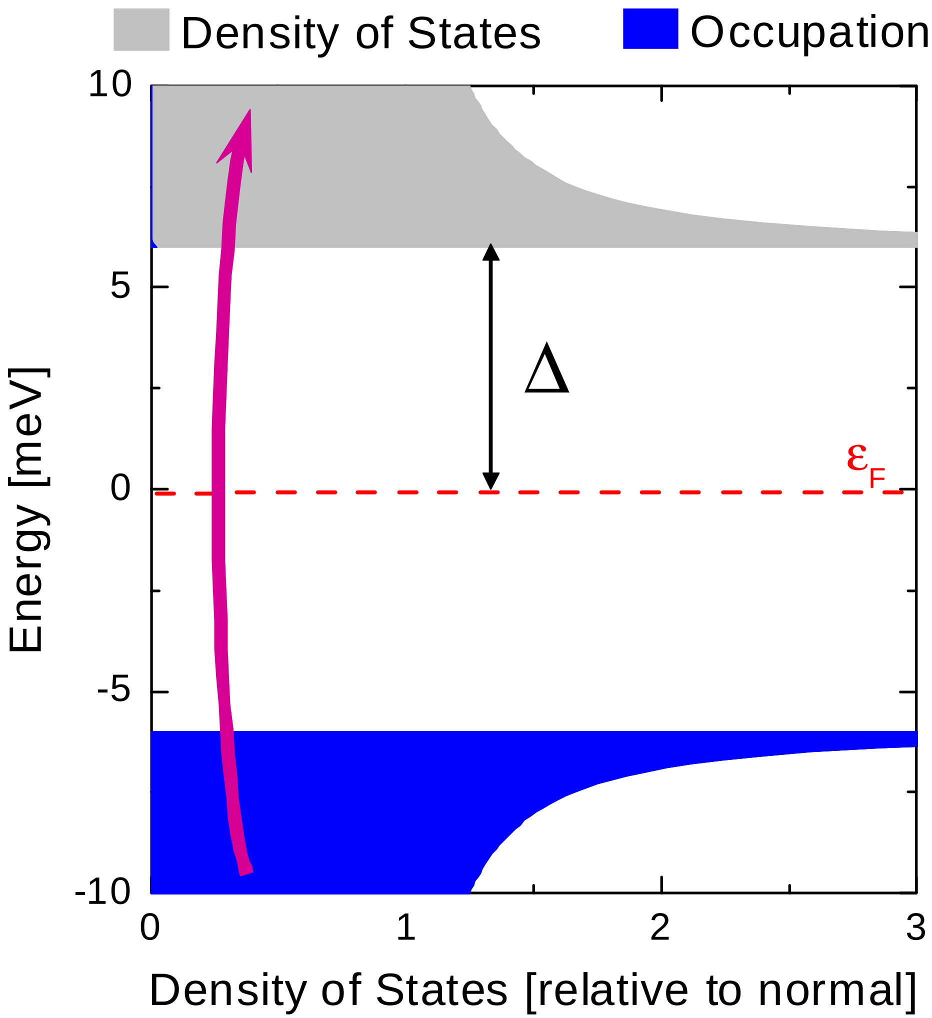
Similar results for other materials





Many applications of SCs invole magnetic fields (motors, energy storage).
How does a magnetic field affect non-equilibrium dynamics (i.e., recovery of system from a perturbation)?

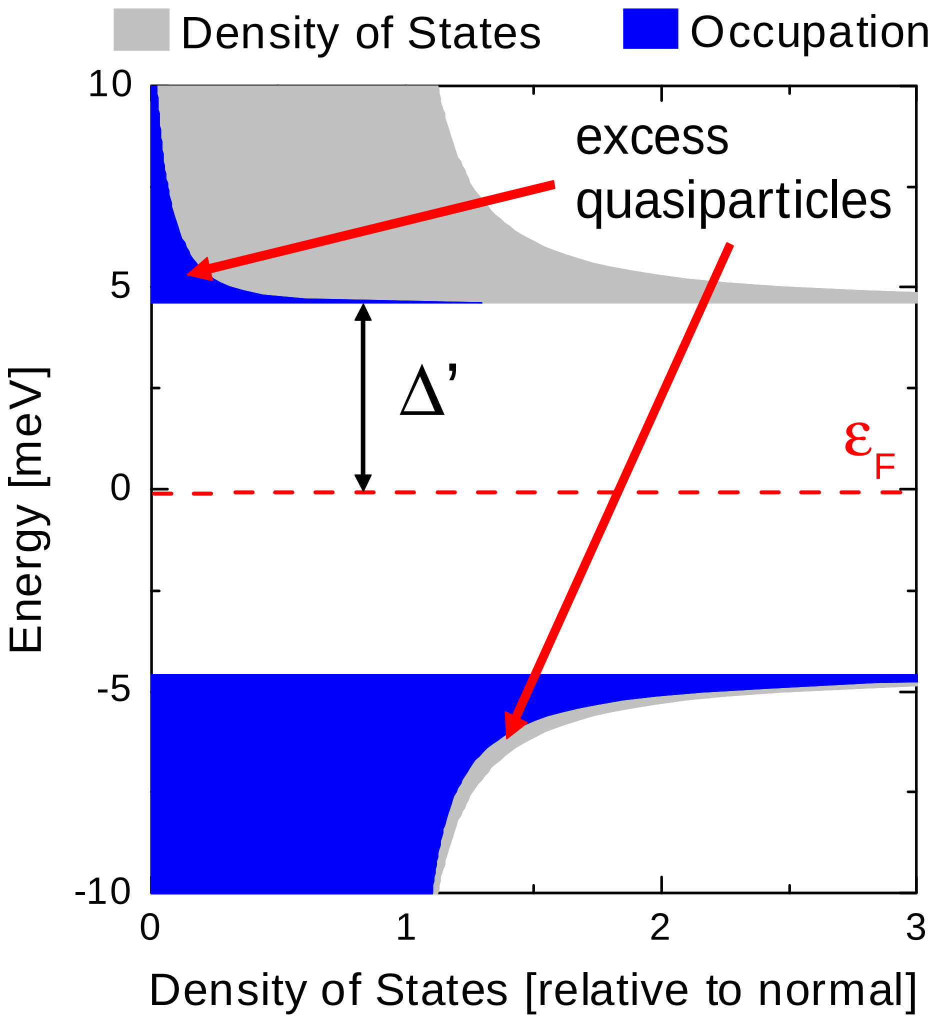
Photoexcitation in a BCS Superconductor



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

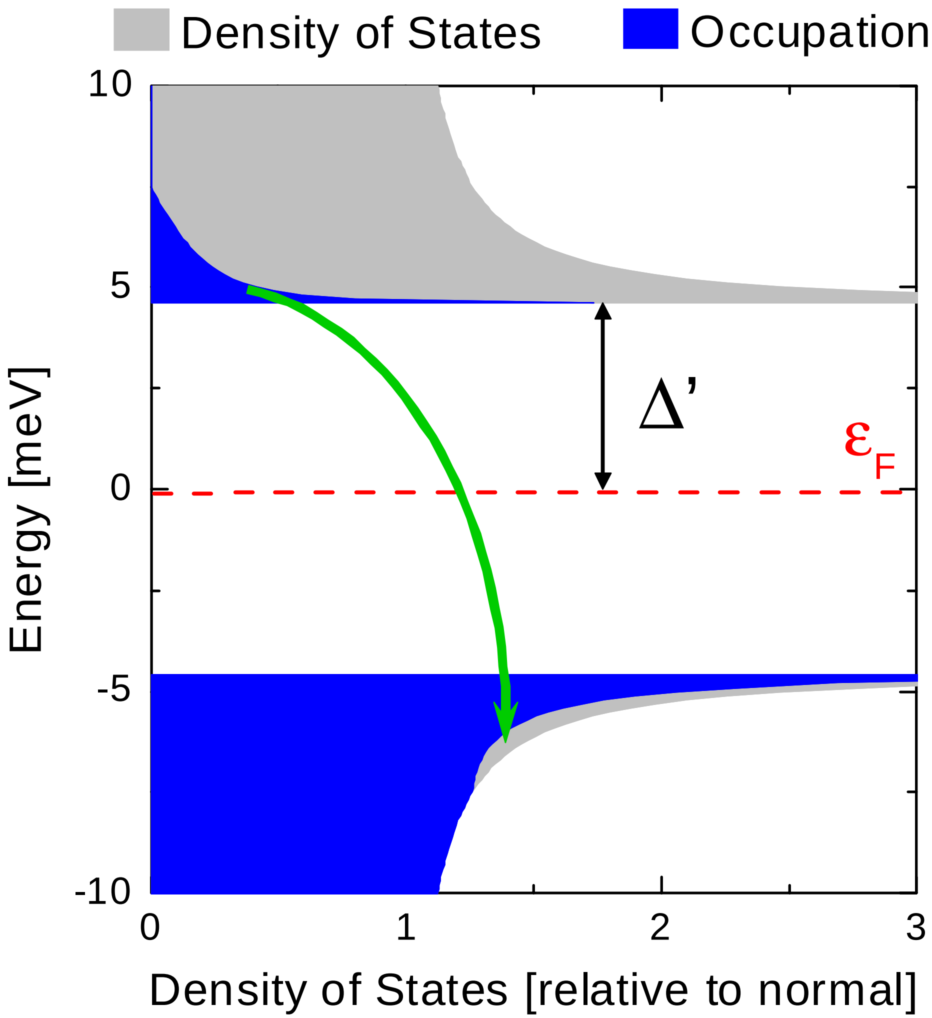


Step 2:
High energy quasiparticles scatter and relax toward gap edge. Many more pairs broken in this process (multiplication).

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

(note: the amplitude of the SC order parameter is affected, not the phase)

Photoexcitation in a BCS Superconductor



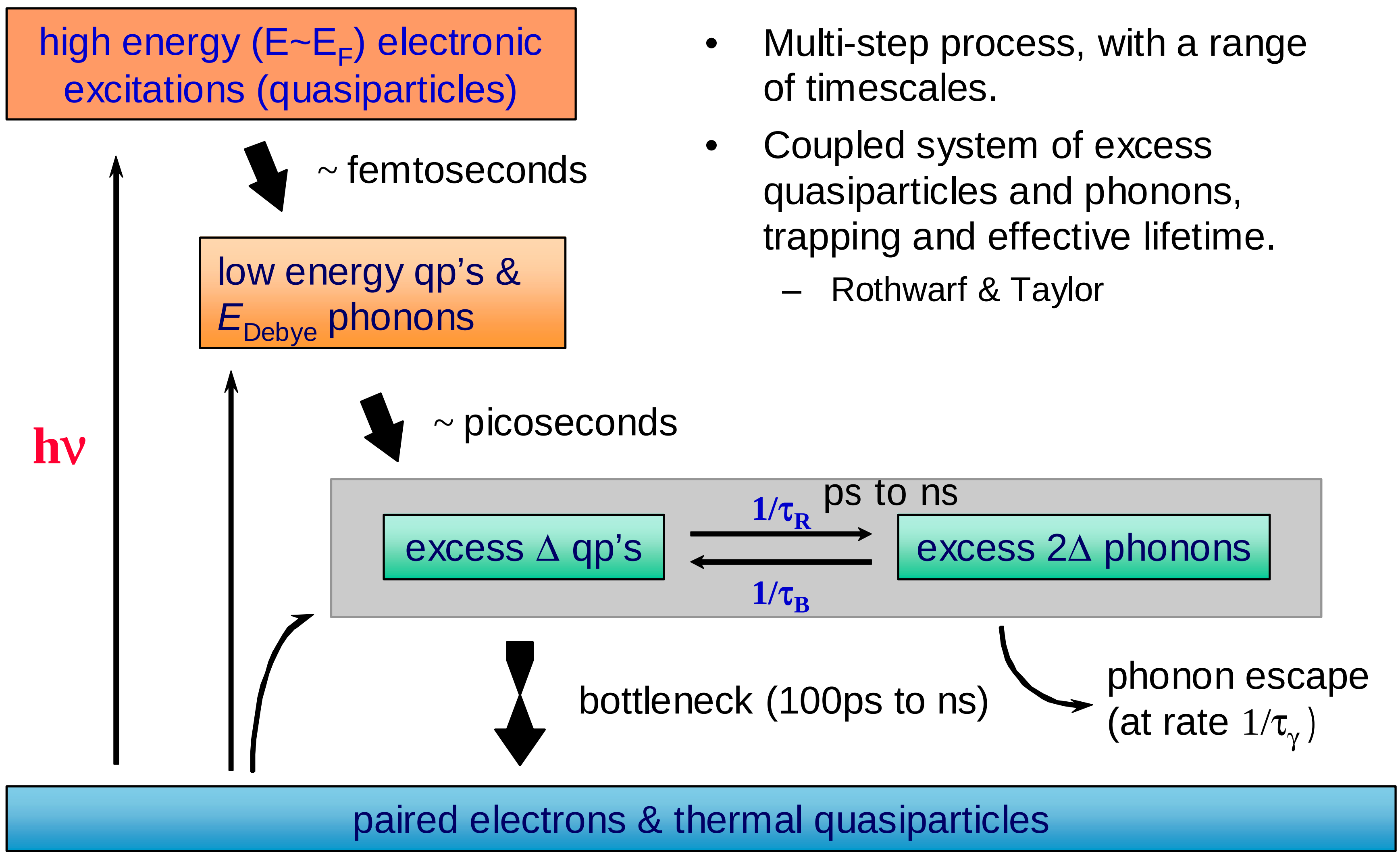
Step 3:

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.

Relaxation processes in superconductors

- Multi-step process, with a range of timescales.
- Coupled system of excess quasiparticles and phonons, trapping and effective lifetime.
  - Rothwarf & Taylor

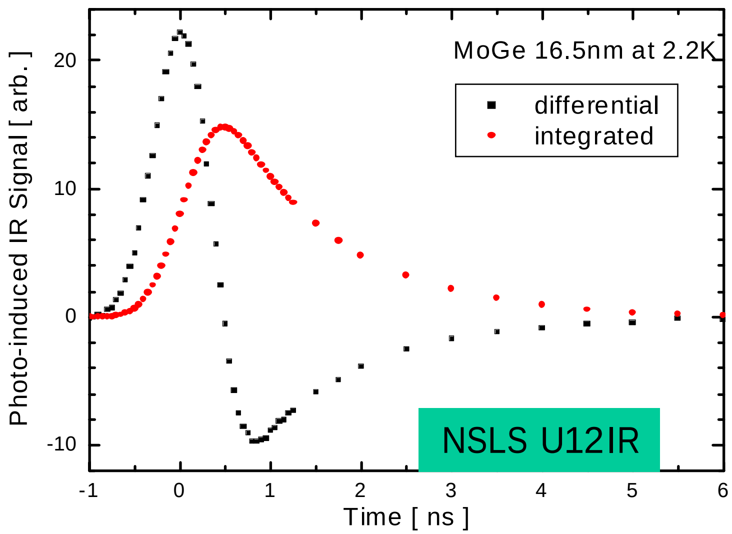


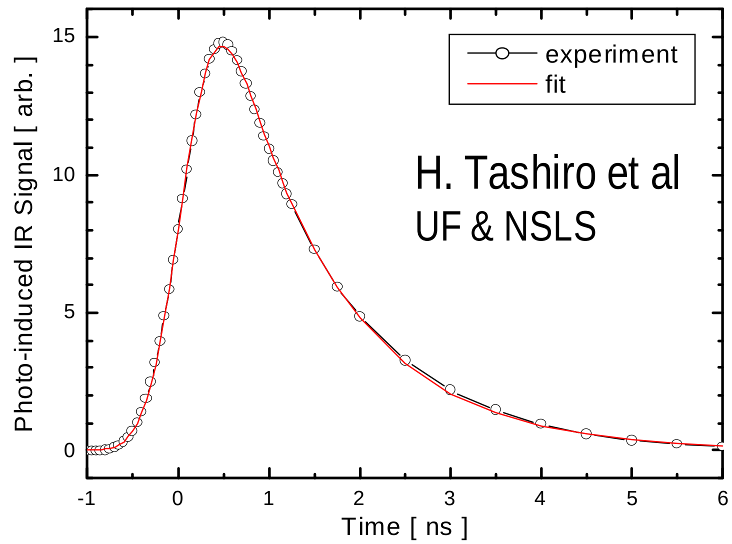
Pump-probe IR & THz spectroscopy

- Photo-induced changes (small) for thin SC films are linearly related to pair density.
- Differential Technique:
  - – pump-probe delay "dithered" at ~ 150Hz.
  - – lock-in detection yields time-derivative of decay. Integrate to recover decay.
- Pair Recombination Dynamics:
  - – recombination of two quasiparticles to remake a pair determined by 2 basic features:
    1. intrinsic electron-electron interactions (e.g., phonon mediated).

       => affects mostly the overall time-scale, not the temperature-dependence.
    2. available populations of quasiparticles (since each excess quasiparticle must find an appropriate "mate").

       => affects mostly the temperature-dependence.
  - – Vortex cores … lots of quasiparticles!

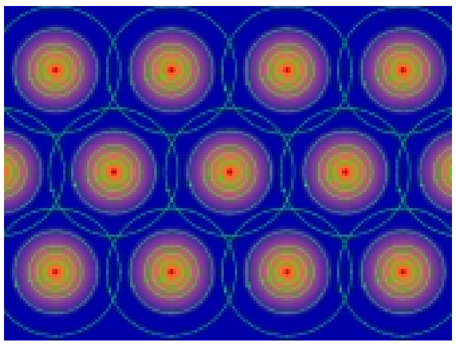


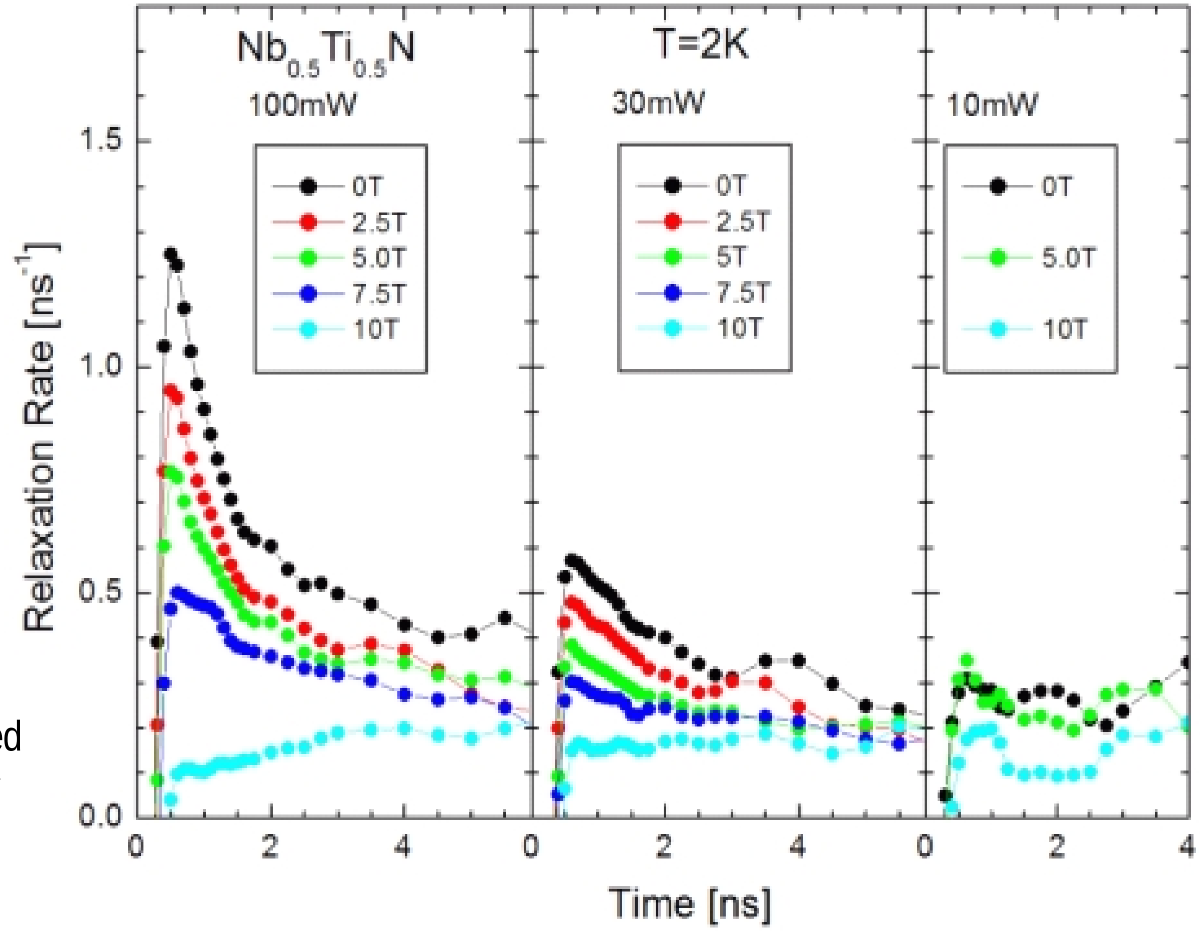


Excess Quasiparticle Relaxation (recombination) in a Magnetic Field

J. Hwang, H. Zhang, D.H. Reitze, D.B. Tanner (UF)
R.P.S.M. Lobo (CNRS-ESPCI)
G.L. Carr (NSLS-BNL)

field-induced vortex state





Result: Magnetic field leads to unexpected reduction in the relaxation rate, especially for high laser fluence (large excess quasiparticle population).

Spin polarization of quasiparticles in the field?

Possible method for measuring spin flip rates?

"Low" Energy Electrodynamics in a Superconductor

Measurements to-date: relaxation of excess quasiparticles involves order parameter amplitude.

What about phase excitations?
Phase <–> currents and voltage.

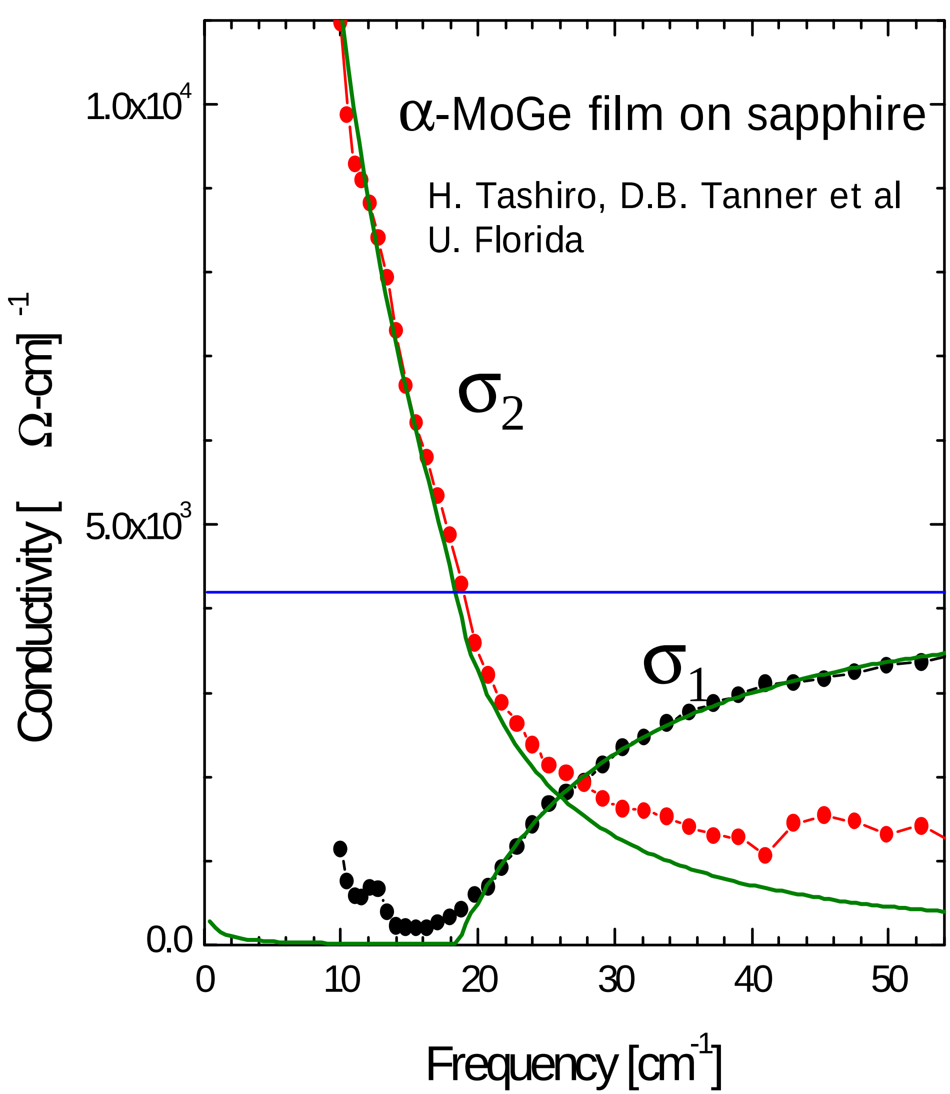
Answer: a probable "yes".
Consider a 100 kV/cm E-field transient with $\hbar\omega < 2\Delta$ (no pair breaking),

Low frequency response is dominated by imaginary part of conductivity:

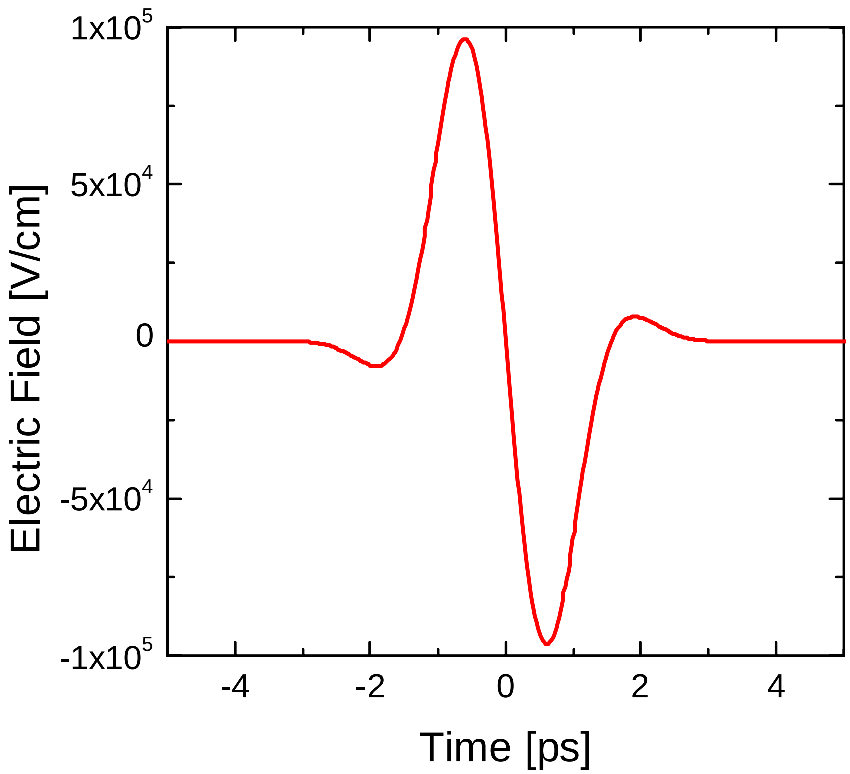
$\sigma_2 \cong A/\omega$; $A \cong \sigma_n \omega_g$ (purely inductive).

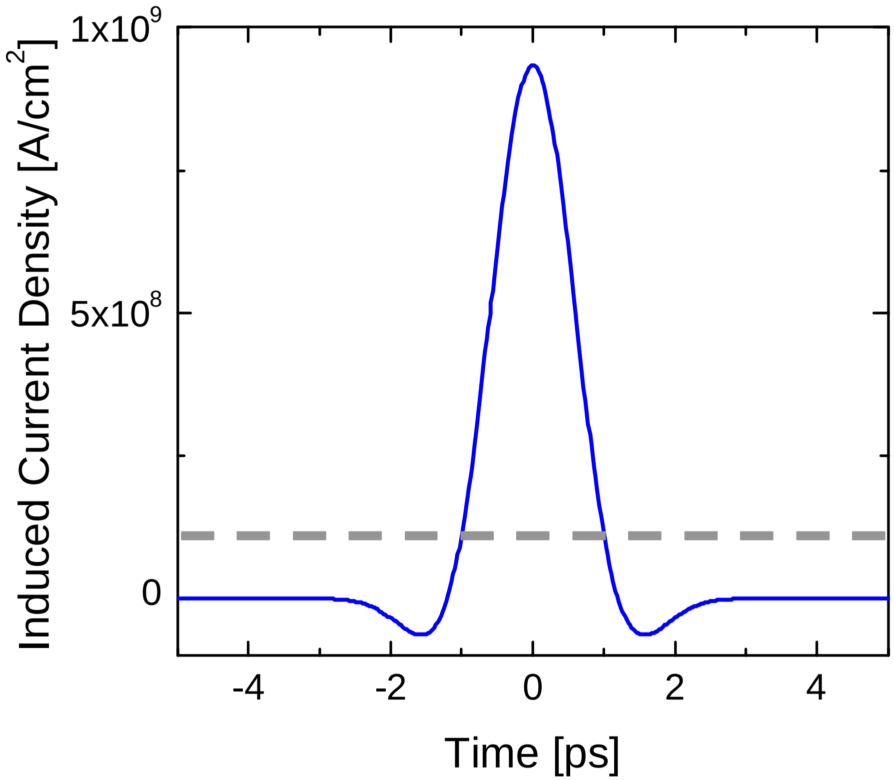
$$L\frac{dI}{dt} = V \qquad I(t) = \frac{1}{L}\int_{-\infty}^{t} V(t')dt'$$

$$J \cong \sigma_n \omega_g \int_{-\infty}^{t} E(t')dt'$$



Proposed Experiment: THz Pulse Driven Supercurrent





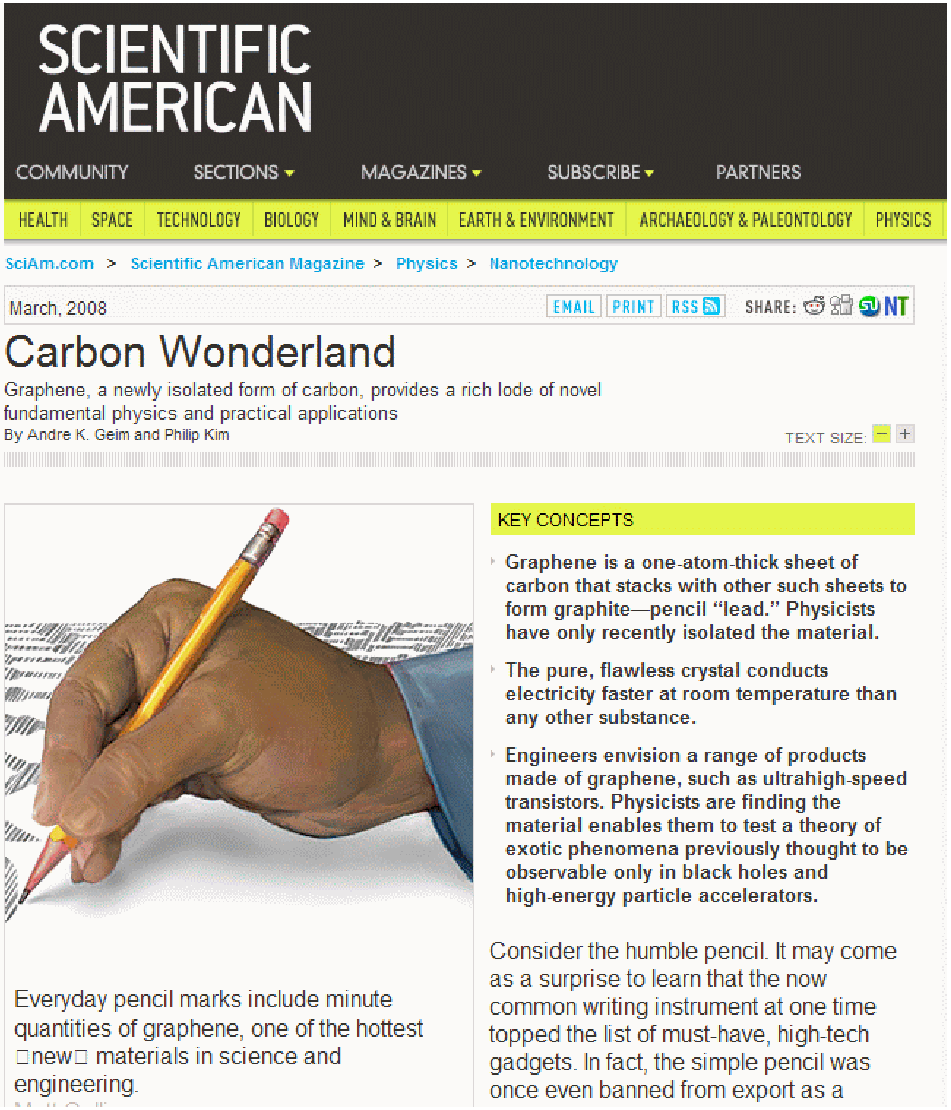
Typical superconductor $J_C \sim 10^8$ A/cm^2

=> “over twist” the local superconducting phase, expect to “spin off” vortices.
How quickly can a vortex be created? How does dissipation initially appear?

Summary of Superconductors

- Principal features of THz spectra for superconductors in a magnetic field are described by a two-component (S/N) model where the SC energy gap shrinks due to field-assisted pair-breaking.
- Magnetic field impedes relaxation of excess quasiparticles in SCs. Possible model based on spin-polarization (not yet quantitative).
- Storage-ring SR pulses (~100 ps) are excellent, stable source for studying the overall relaxation on a ~500 ps time scale. But dynamics can occur on 1 ps time scale … need shorter THz pulses.
- THz driven phase excitations will need strong THz pulses.

Graphene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SciAm.com > Scientific American Magazine > Physics > Nanotechnology

March, 2008 EMAIL PRINT RSS SHARE:

Carbon Wonderland

Graphene, a newly isolated form of carbon, provides a rich lode of novel fundamental physics and practical applications

By Andre K. Geim and Philip Kim

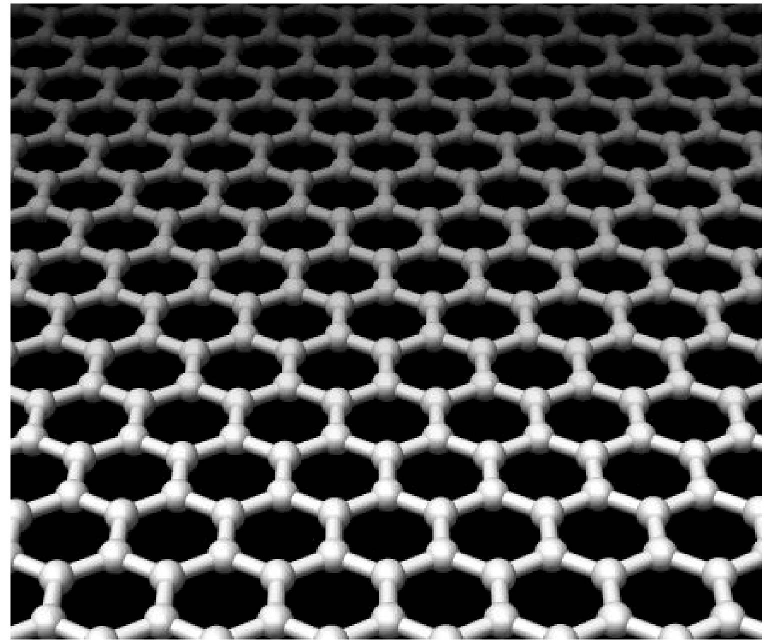
TEXT SIZE:

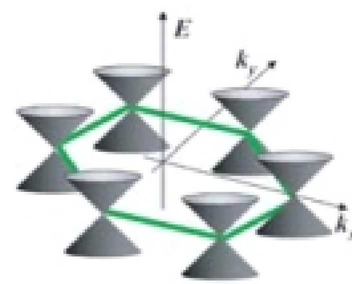
KEY CONCEPTS

- Graphene is a one-atom-thick sheet of carbon that stacks with other such sheets to form graphite—pencil "lead." Physicists have only recently isolated the material.
- The pure, flawless crystal conducts electricity faster at room temperature than any other substance.
- Engineers envision a range of products made of graphene, such as ultrahigh-speed transistors. Physicists are finding the material enables them to test a theory of exotic phenomena previously thought to be observable only in black holes and high-energy particle accelerators.

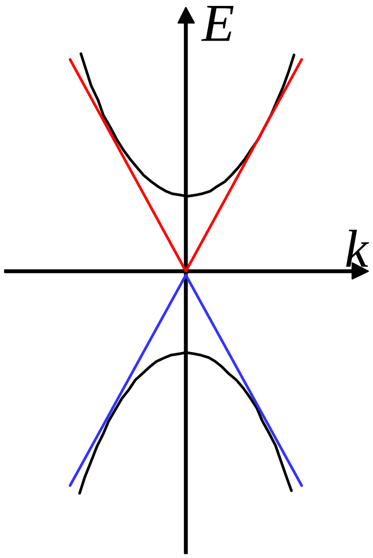
Everyday pencil marks include minute quantities of graphene, one of the hottest new materials in science and engineering.

Consider the humble pencil. It may come as a surprise to learn that the now common writing instrument at one time topped the list of must-have, high-tech gadgets. In fact, the simple pencil was once even banned from export as a


(from wikipedia)



Graphene: Electrons with Linear Dispersion



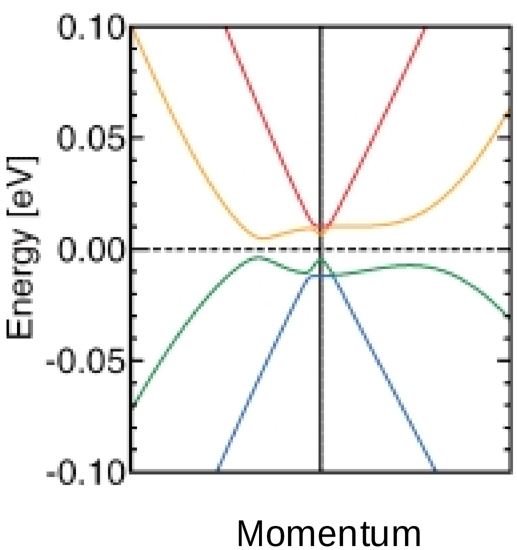
quasiparticles in a typical solid
=> nearly free electron behavior: $E = \hbar k_F^2/2m^* = p^2/2m^*$

but, for quasiparticles in graphene: $E = \hbar k\mathrm{v} = p\mathrm{v}$ (or $E^2 = p^2\mathrm{v}^2$)

compare to "relativistic" particles: $E^2 = (m_0\mathrm{v}^2 + p^2\mathrm{v}^2)$

Electrons (and holes) in graphene behave like nearly massless "Dirac particles" traveling at "relativistic" velocity v (10^6 m/s).

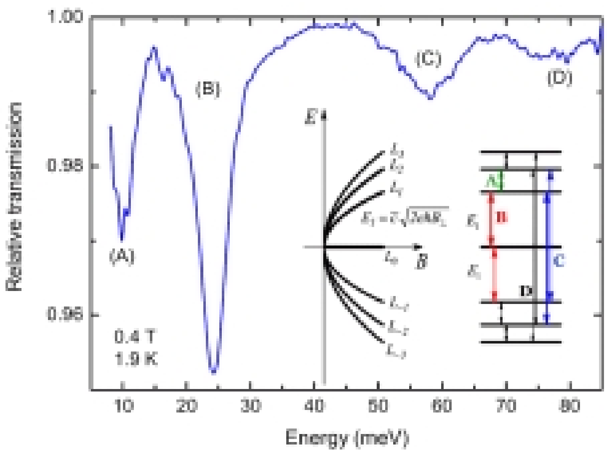
Result: Unusual transport & magnetic field effects: e.g. cyclotron resonance.
–> Landau level spacings scale as $B^{1/2}$ rather than B.

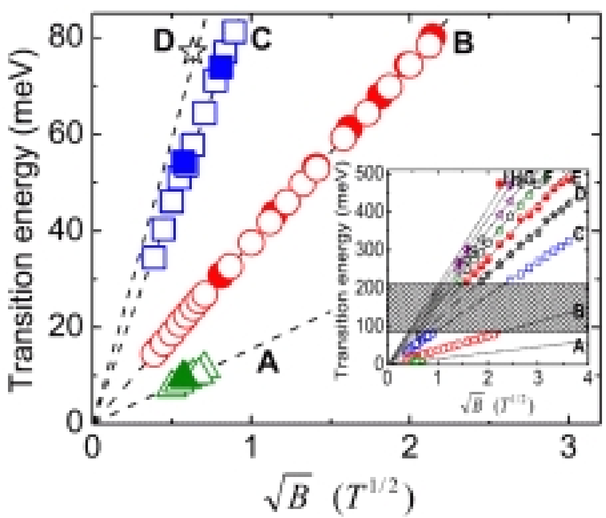


interactions in
4-layer graphene

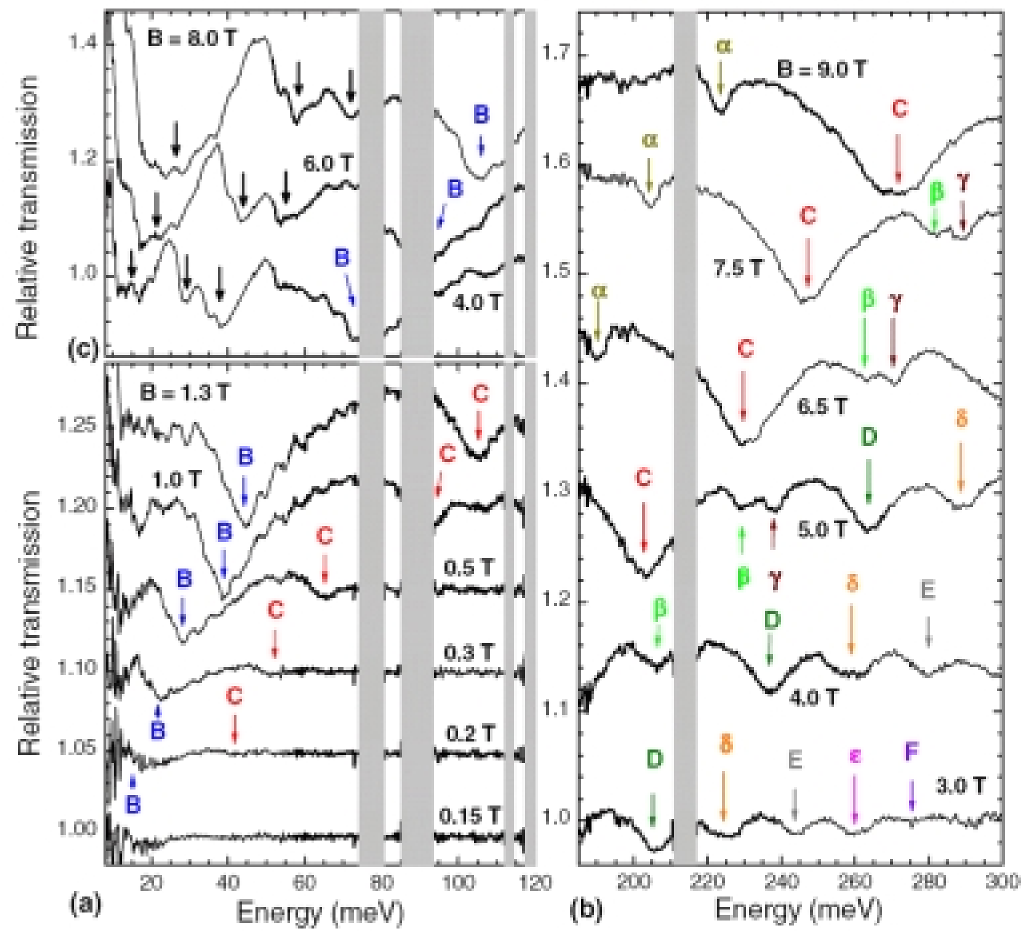
Landau Levels in Few Layer Graphene

Sadowski et al, *Phys. Rev. Lett.* **97**, 266405 ('06)
Li & Andrei, *Nat. Phys. Lett.* **3**, 623 ('07)





Graphite "flakes" show LLs with B & $B^{1/2}$ spacing
Orlita et al, *Phys. Rev. Lett.* **100**, 136403 ('08).



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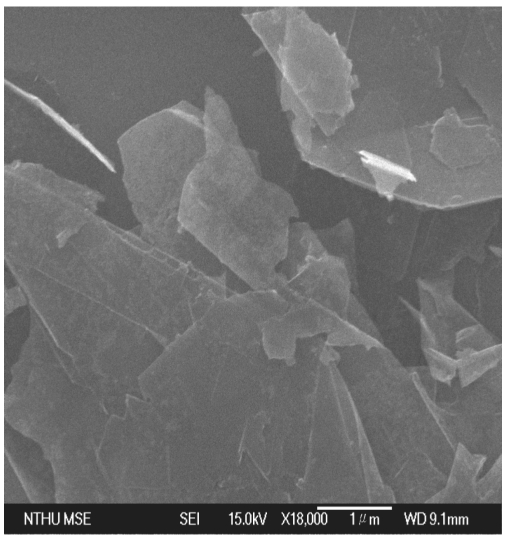
Graphene NanoPlatelet Films on Silicon

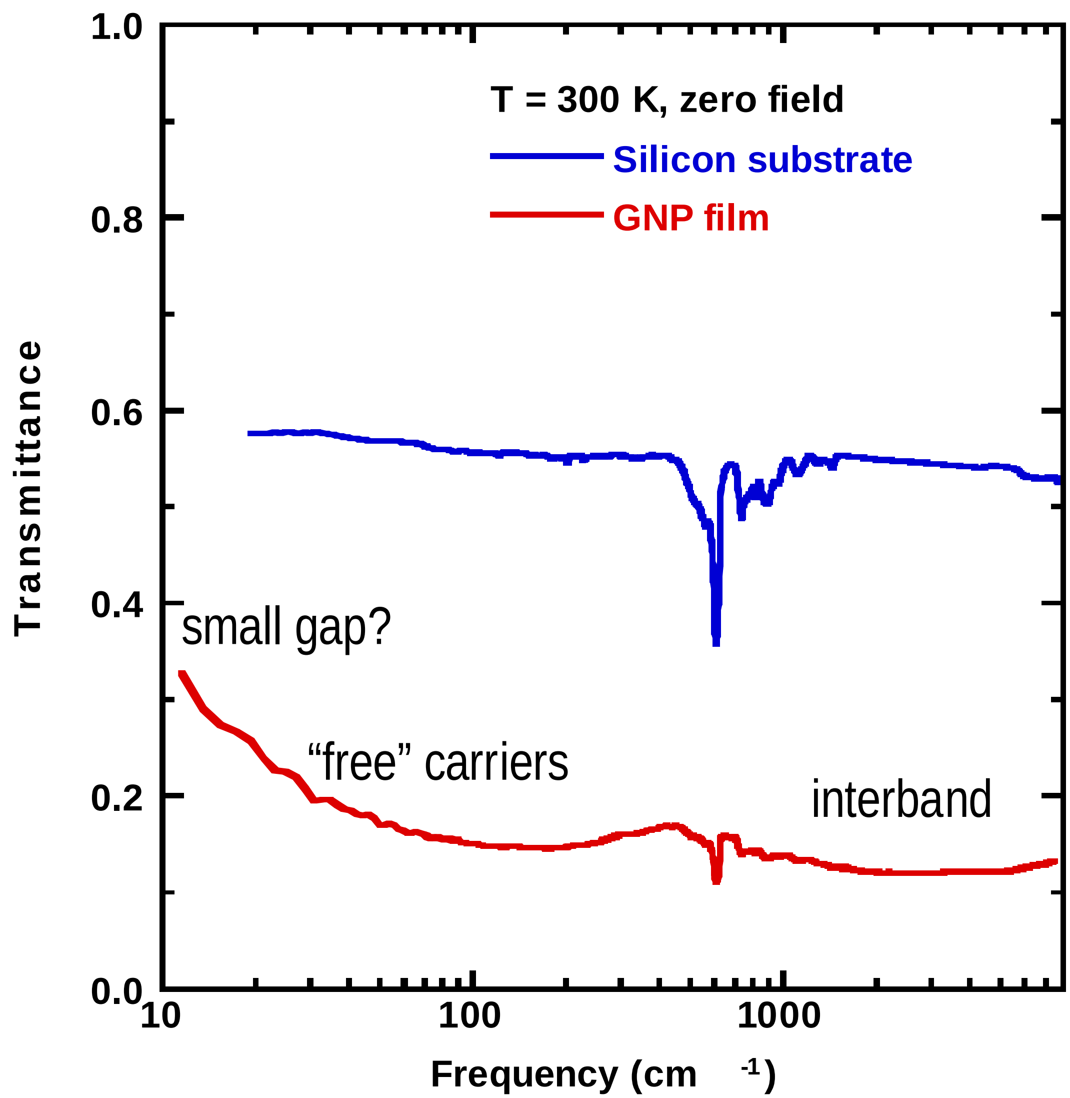
Vapor-grown graphite thin films.
Large area and thick.

IR and THz spectroscopy to extract carrier density, scattering rates, energy gaps).

Do they show any Dirac-particle behavior?

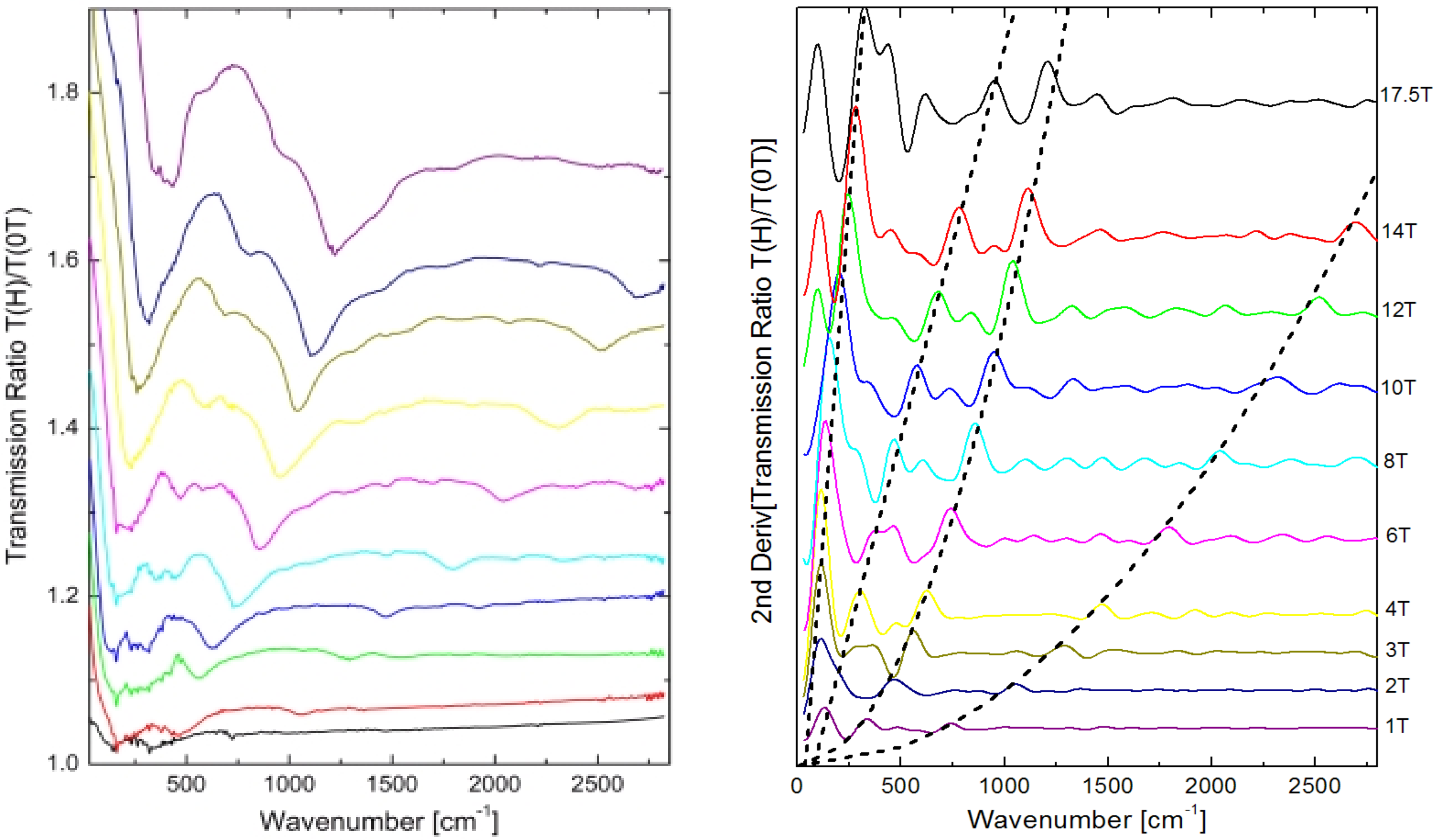


H-L Liu (NTNU), G.L. Carr (BNL)
A Caruso (N.Dakota St. Univ.)
K Worsley, R.C. Haddon (UC - Riverside)



NSLS: Landau Levels in GNP films: $B^{1/2}$ & B

H-L Liu (NTNU), Y-J. Wang (NHMFL) et al



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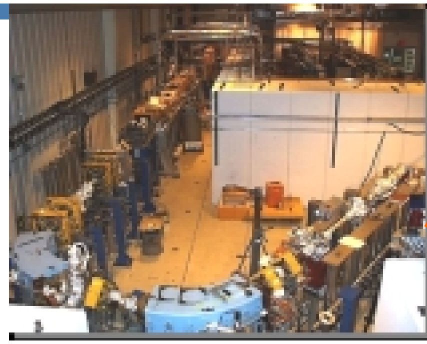
Summary (graphene)

- Graphene is difficult to produce and manage in form of single, non-interacting sheets.
- Interactions (such as in multi-layer graphene) leads to opening of gap near Fermi energy. But the gap is often quite small.
- Even with interactions, linear dispersion of quasiparticles is still observed in magnetospectroscopy. Near-massless Dirac particles present.
- Predicted strong non-linear response for Dirac particles
  - a good measurement for accelerator-based THz pulses.

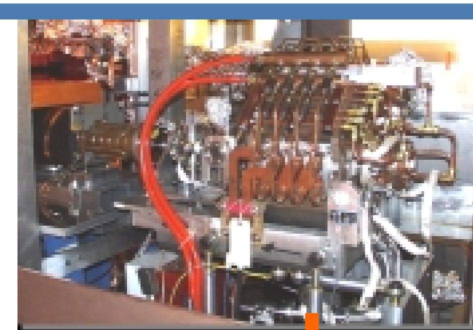
next: NSLS source development lab linac source of THz radiation

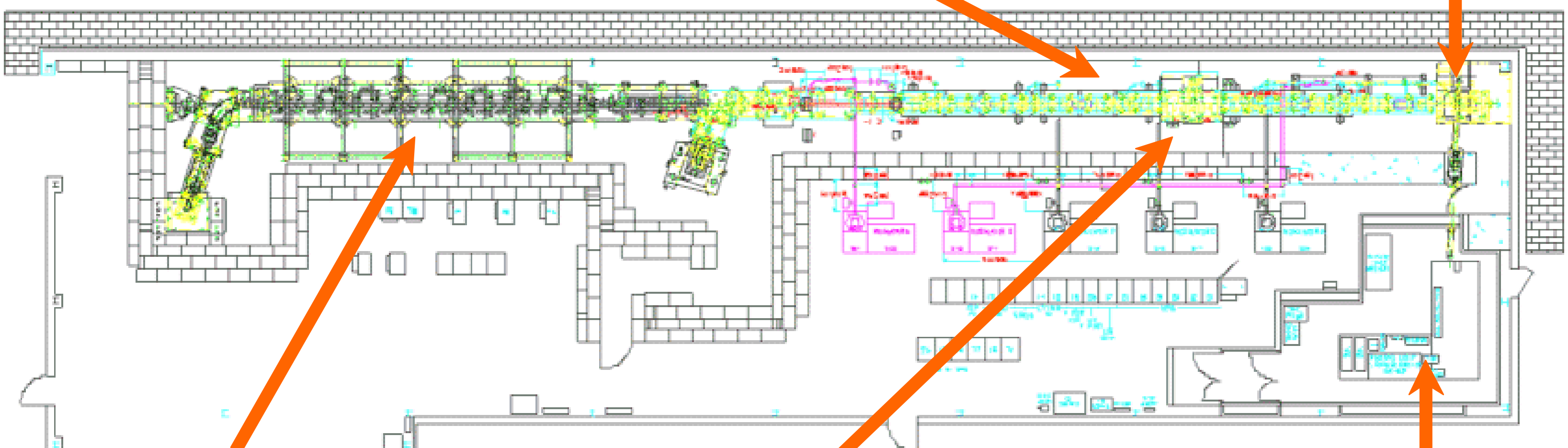
Short Bunch Source: The NSLS Source Development Lab Photo-injected Linac

300 MeV
S-Band Linac
(DARPA)

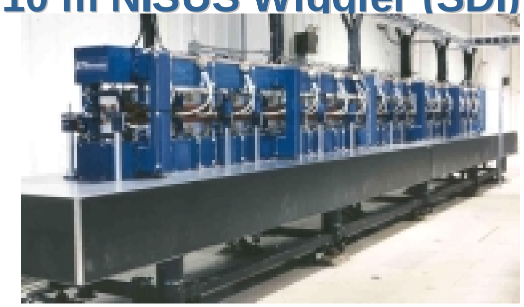


BNL
Photo-injector IV





10 m NISUS Wiggler (SDI)



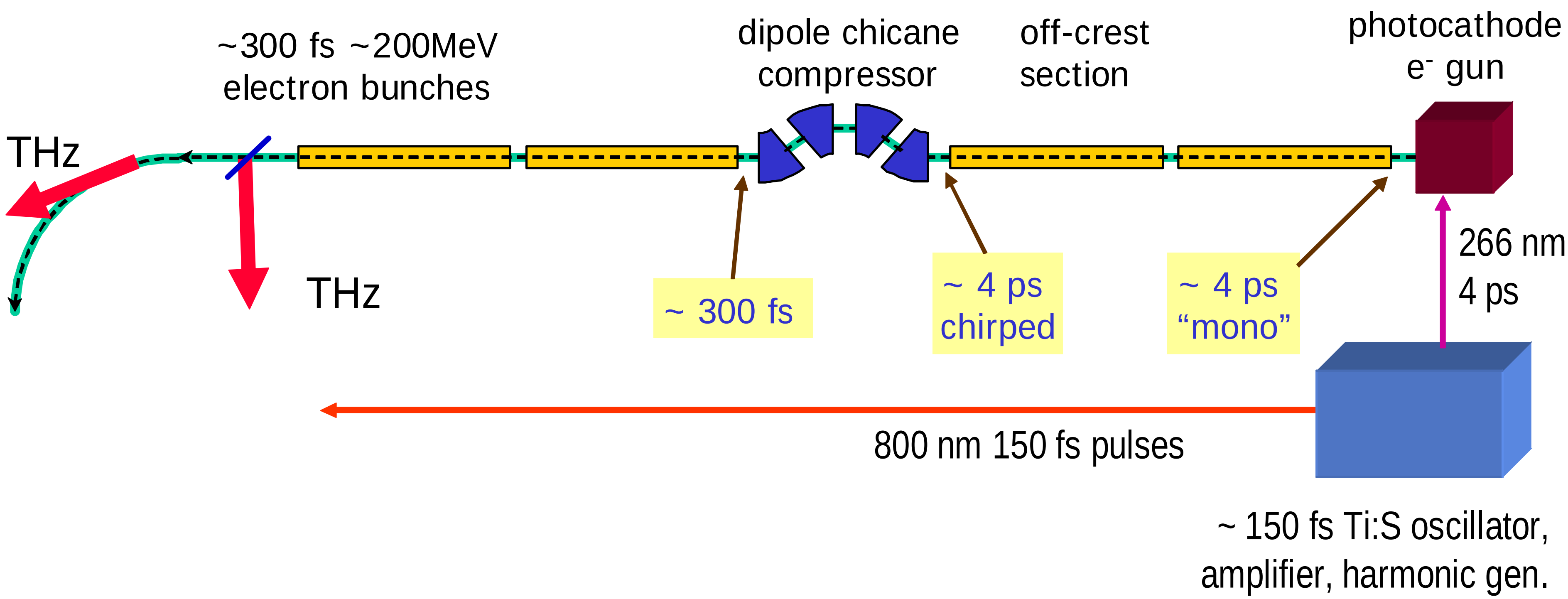
Chicane Bunch Compressor

Ti:Sapphire Laser

The NSLS Source Development Lab Linac

X.-J. Wang et al

- Photocathode gun produces ~ 0.84nC ($5x10^9$ electrons) per "shot"



- Coherent output to over 1 THz. Potential for shorter bunches with less charge.
- Low rep. rate (1 to 10 Hz)

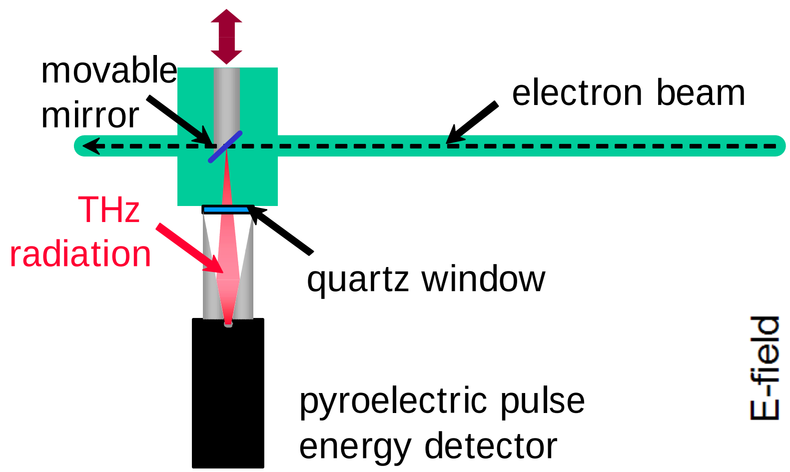
Coherent THz Pulses

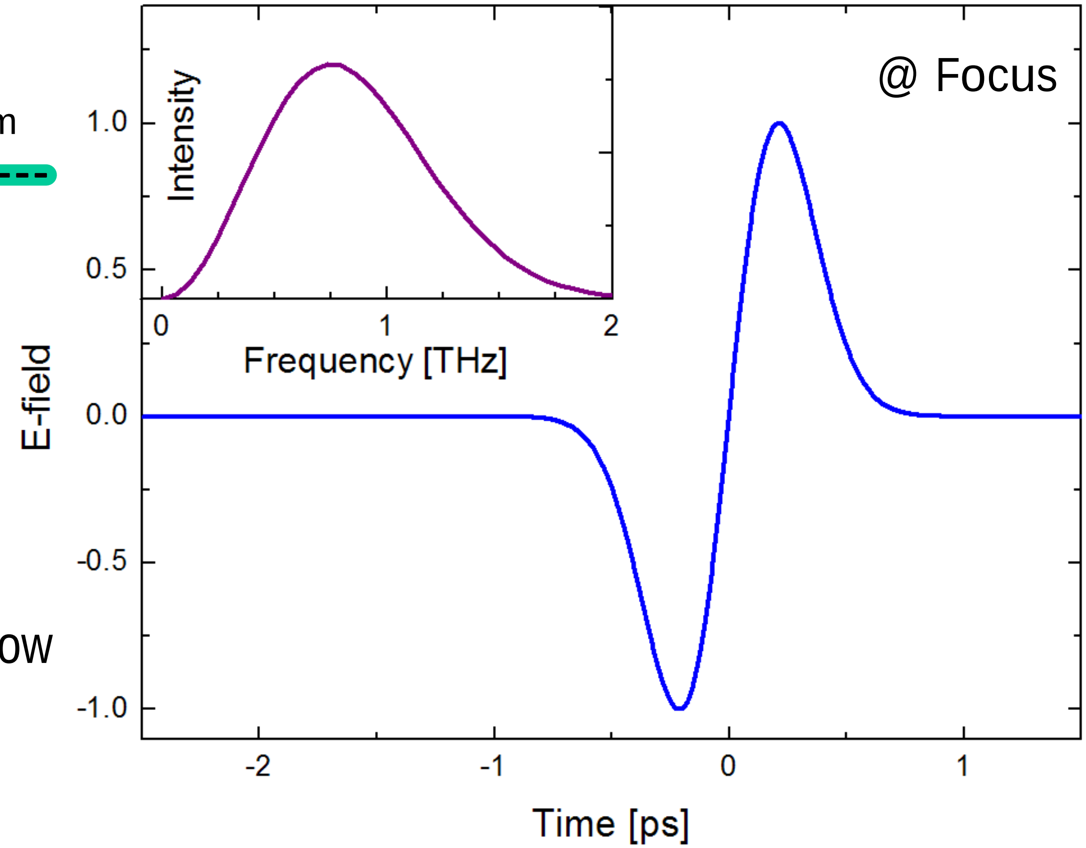
Transition Radiation: Energy per electron per ω ?

$$E = \frac{e^2}{\pi c}\left[\ln\left(\frac{2}{1-\beta}\right) - 1\right]$$

(Happek et al, PRL)

10^{10} electrons, 116 MeV coherent to 1 THz
=> pulse energy of 400 μJ





Finite source and aperture, quartz window reflection => ~ 35% efficient or 140 μJ.
We have measured 100 μJ per pulse.

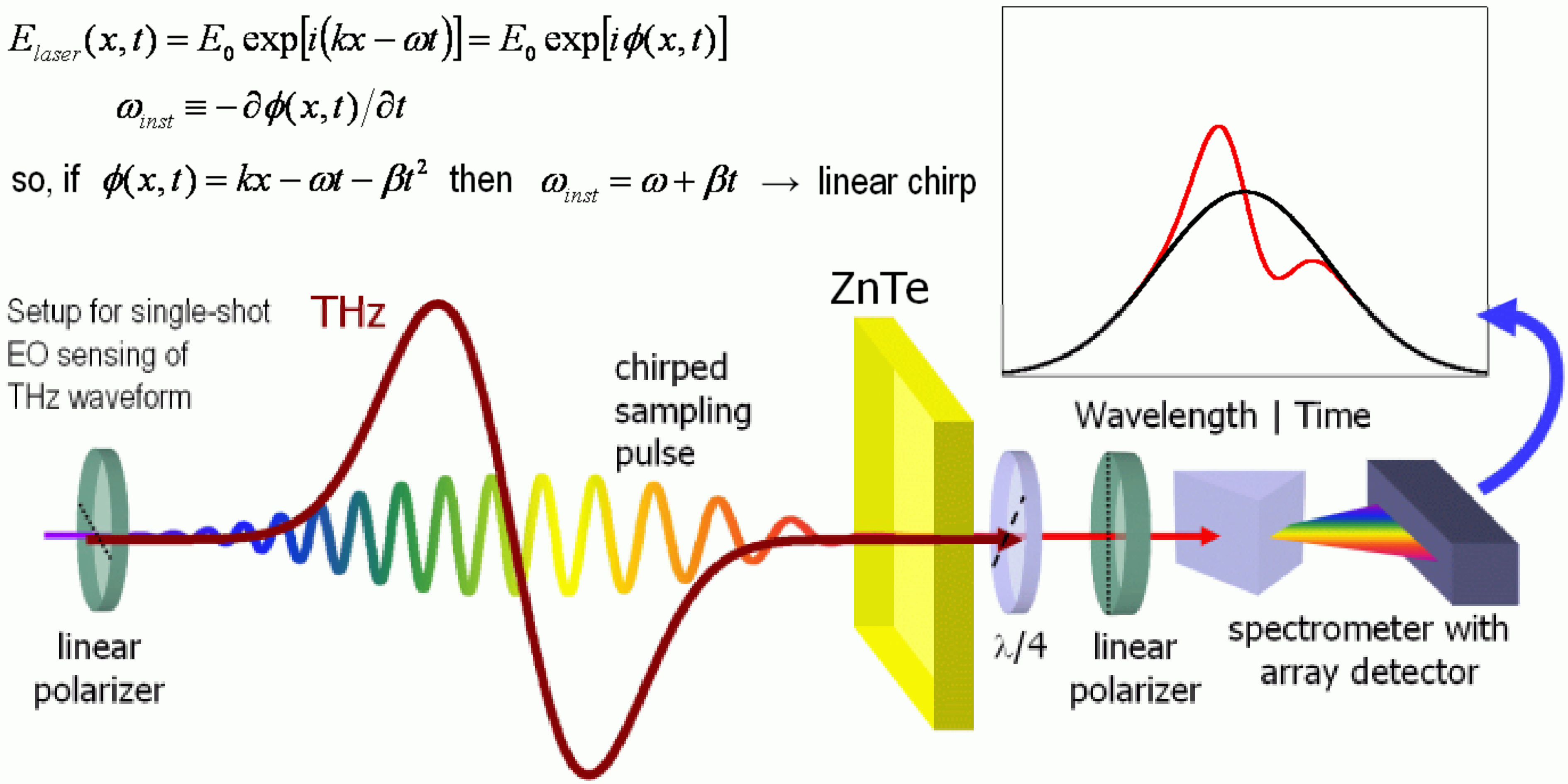
Single-Shot Electro-Optic Method

Use chirped sampling laser to encode waveform's entire time-dependence onto different wavelengths of laser in a single pulse. Avoids need for multiple sampling.
[Jiang and Zhang, *Appl. Phys. Lett.* **72**, 1945 (1998)].

$$E_{laser}(x,t) = E_0 \exp[i(kx - \omega t)] = E_0 \exp[i\phi(x,t)]$$

$$\omega_{inst} \equiv -\partial\phi(x,t)/\partial t$$

so, if $\phi(x,t) = kx - \omega t - \beta t^2$ then $\omega_{inst} = \omega + \beta t$ $\rightarrow$ linear chirp



Time-Dependent THz E-field and Phase Modulation Effects

- Pockels electro-optic effect in terms of the induced phase $\phi[E_{THz}(t)]$ for the sampling laser:

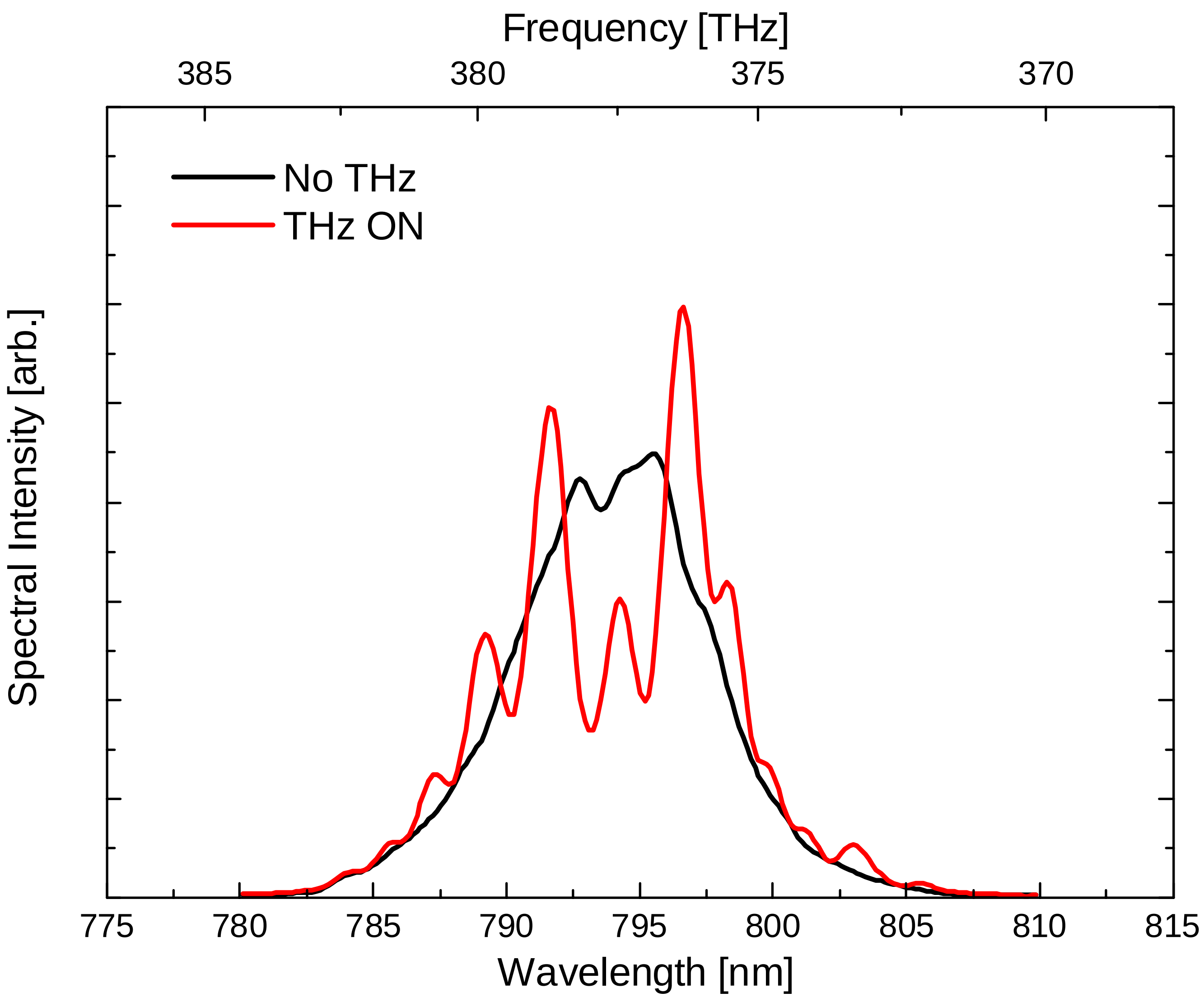
$$E_{laser} \sim \cos\left[\phi_0 + \phi(t)\right]; \qquad \phi(t) = \frac{2\pi L}{\lambda_0} n\left[E_{THz}(t)\right] - \omega t$$

- Taylor series expansion of laser phase:

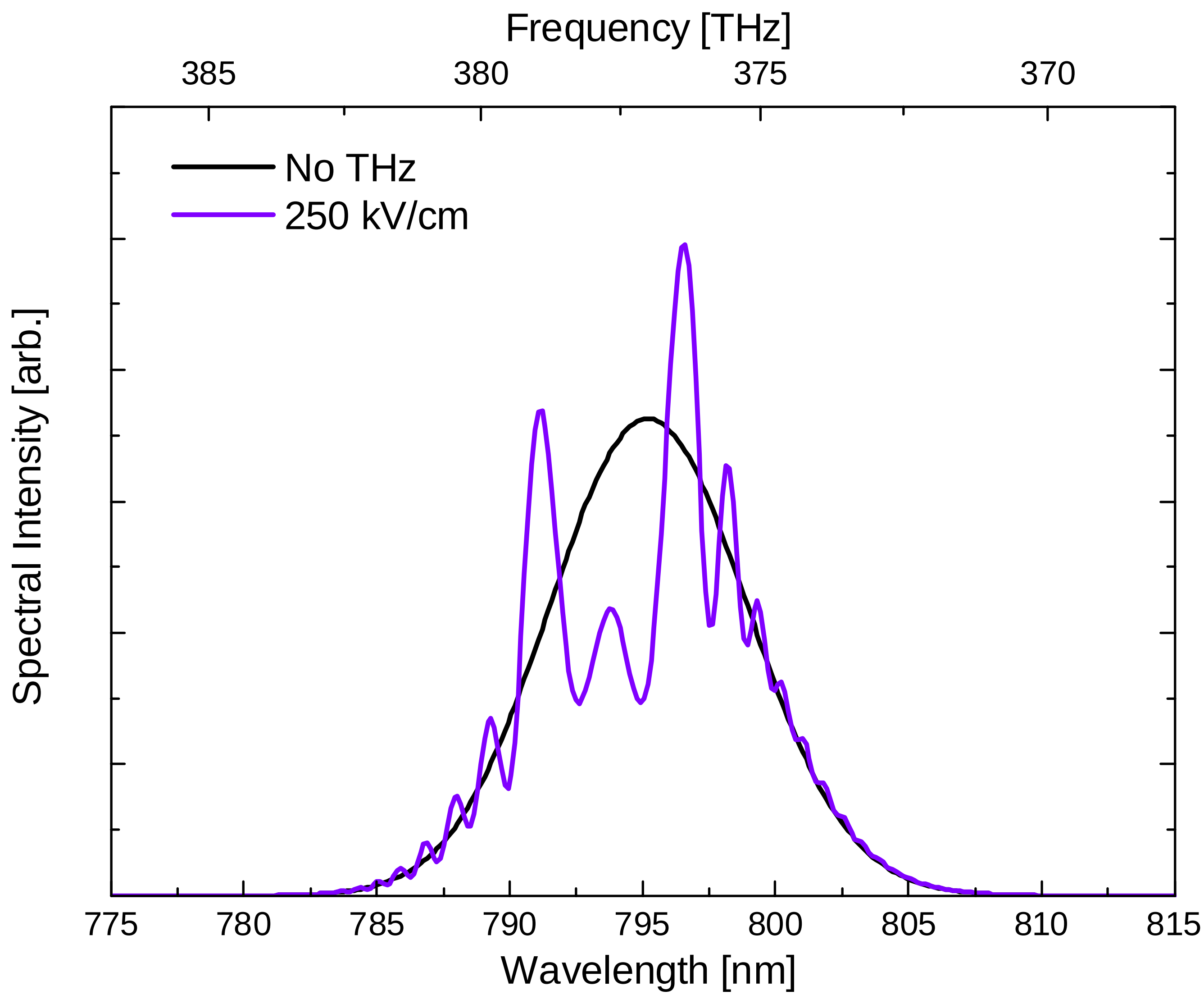
$$\phi(t) = \underbrace{\eta E(0)}_{\substack{\text{conventional}\\ \text{EO effect}}} + \left[\underbrace{\eta\left(dE_{THz}/dt\right)}_{\substack{\text{freq.}\\ \text{shift}}} - \omega\right] t + \left[\underbrace{\eta\left(d^2E_{THz}/dt^2\right)}_{\substack{\text{linear}\\ \text{chirp}}}\right] t^2 + \ldots \quad \text{where} \quad \eta = \frac{2}{1+\sqrt{\varepsilon}} \frac{2\pi L}{\lambda_0} n_0^3 r_{41}$$

- Different terms in phase correspond to simple phase shifts, spectral shifts and even spectral chirping.
- Result: When THz is sufficiently strong, it modifies the spectral content of the Ti:S laser.
- Note: for 1% wavelength shift at λ=800nm with 0.5mm ZnTe, need dE/dt = 1.3 MV/cm/ps
- Application: THz *control* of ultra-fast laser pulses (tuning, chirp+compression, lensing, …)
- Effects simplified using an unchirped laser (to sample just a small segment of THz waveform).

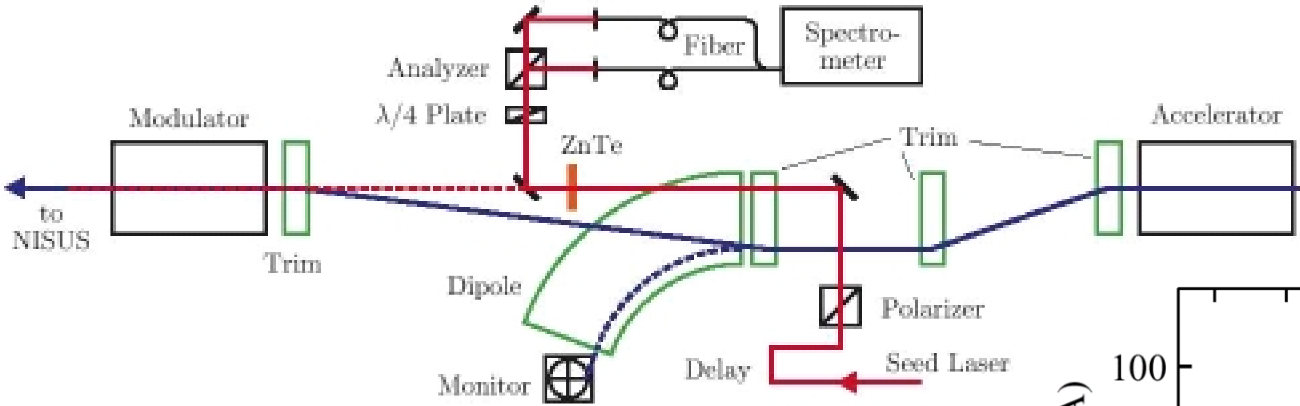
Single-Shot EO Sampling of SDL THz Pulse: Higher intensity



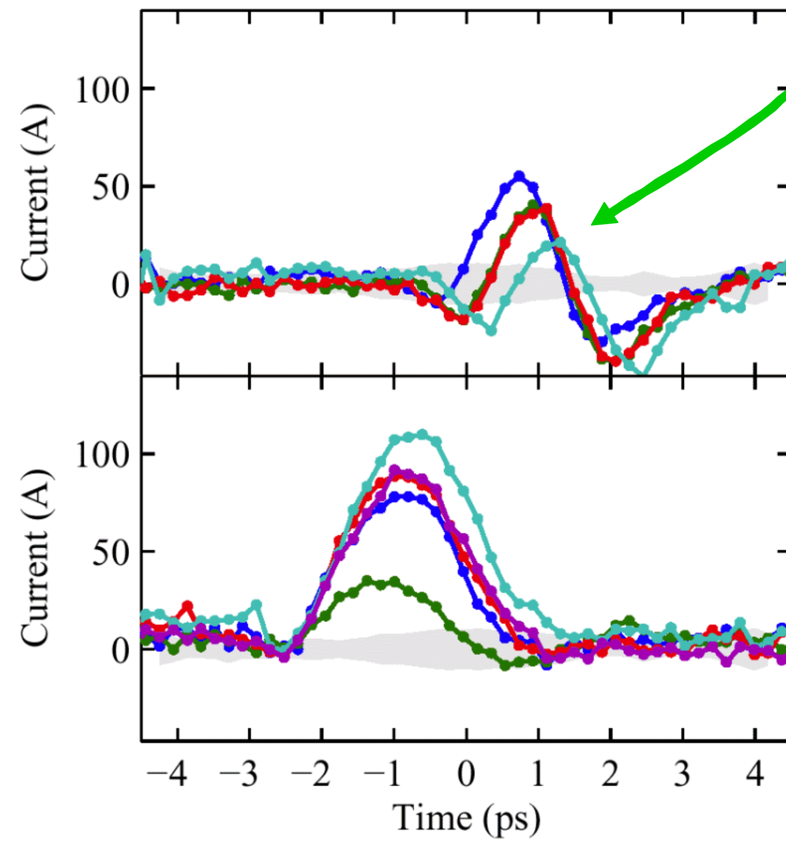
Full Electro-Optic Calculation

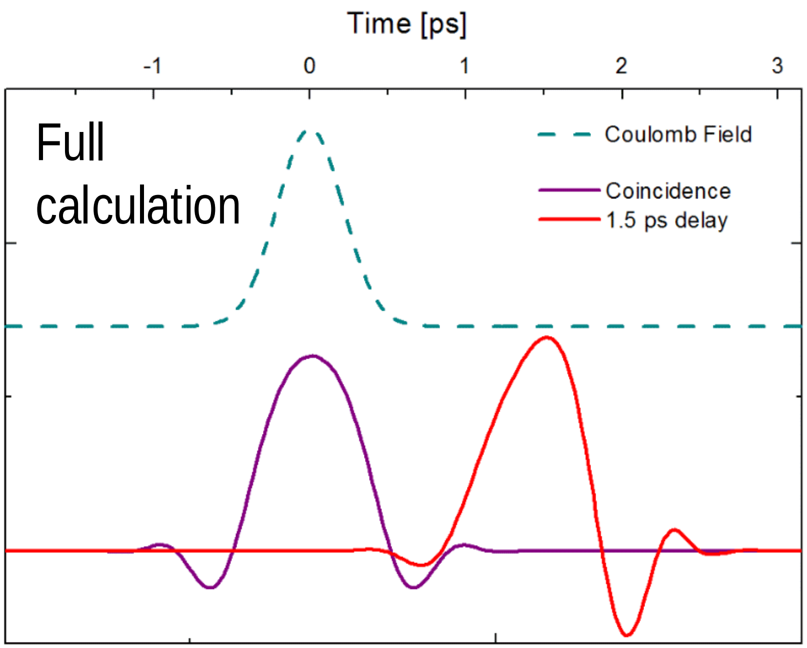


EO Detection of Bunch Coulomb Field (inside linac)



X. Yan et al (PRL '00)
I. Wilke et al (PRL '02)
H. Loos et al (PAC '03)



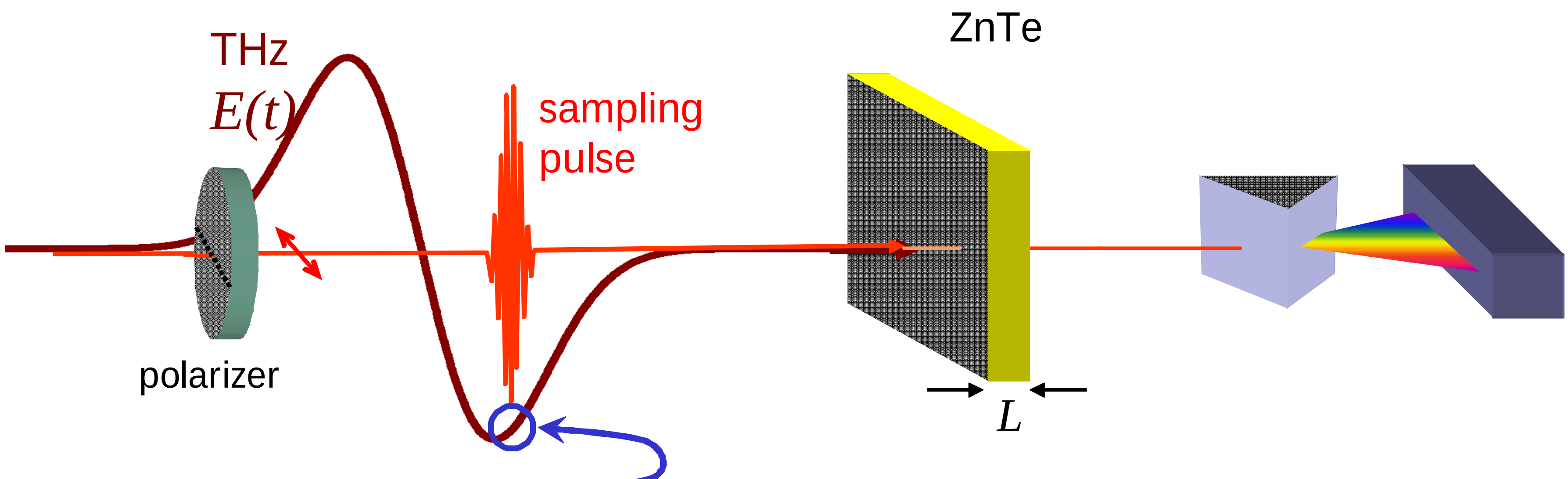


THz Control of Ultrafast Laser

"Simple" EO setup to observe time-dependent phase modulation

$$E_{laser} \sim \cos\left[kz + \Delta\phi_E(t) - \omega t\right] \quad \text{where} \quad \Delta\phi_E(t) = \left(2\pi\, L/\lambda_0\right)\Delta n\left[E_{THz}(t)\right]$$

Electro-optic material (ZnTe) acts as cross phase modulator

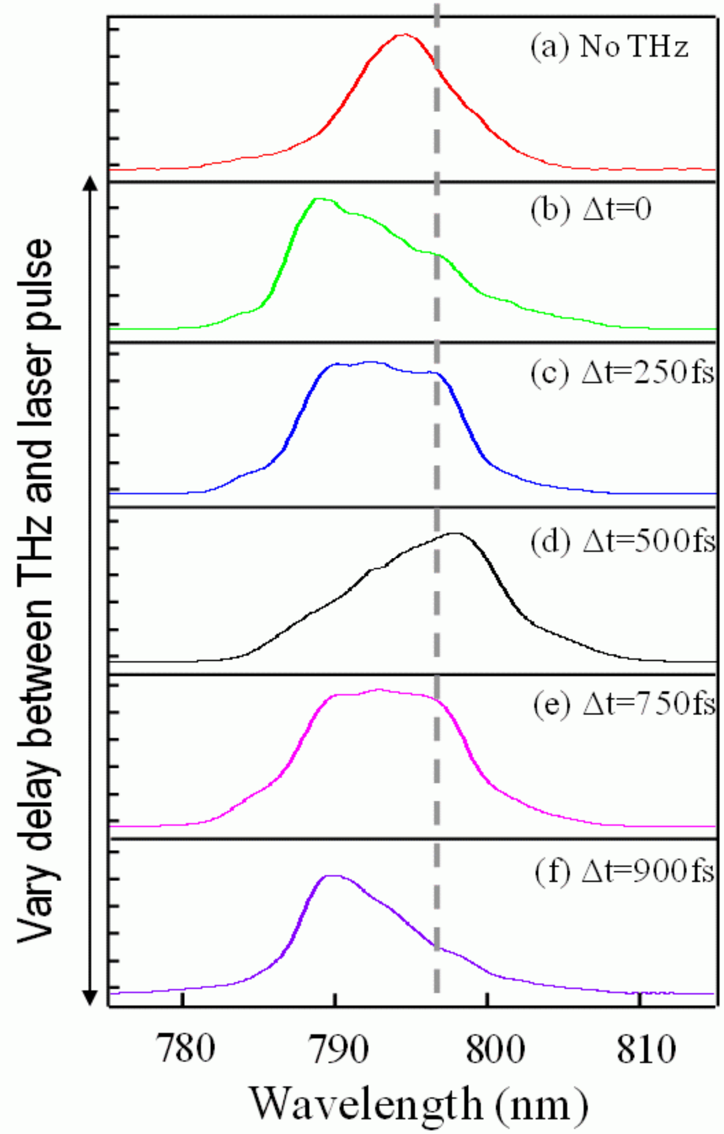


$$E(t) = E(t_0) + \frac{dE}{dt}(t - t_o) + \frac{1}{2}\frac{d^2E}{dt^2}(t - t_o)^2 + \ldots$$

Measured Phase Modulation with SDL Linac Coherent THz

- Electro-optic measurements of SDL THz pulses.
  - 35 μJ pulses, 2mm focus, 0.5mm ZnTe.
- ~ 130 fs (FWHM) unchirped laser sampling pulse, no polarization analysis.
- Probably still a mixture of effects
  - optical alignment and waveform distortion
  - walk-off (velocity mis-match)
  - phase modulation (2nd and 3rd order NLO)
  - dynamic lensing that affects coupling into spectrometer's optical fiber.

Y. Shen, T. Watanabe, D. Arena, T. Tsang, C.-C. Kao, J.B. Murphy, X.-J. Wang, G.L. Carr, *Phys. Rev. Lett.* **99**, 043901 (2007)



Summary / Conclusions

- New science can be explored with THz
  - novel superconductors
  - ferroelectrics, multiferroics
  - mesoscopic carbon
- Need shorter pulses for better temporal resolution:
  - time scale of 1 ps rather than 100 ps
- Strong pulses to induce new phenomena:
  - phase excitations in superconductors
  - non-linear properties of materials
  - strong-field effects: dielectric breakdown