2139-32

School on Synchrotron and Free-Electron-Laser Sources and their Multidisciplinary Applications

26 April - 7 May, 2010

Terahertz Spectroscopy

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Terahertz Spectroscopy

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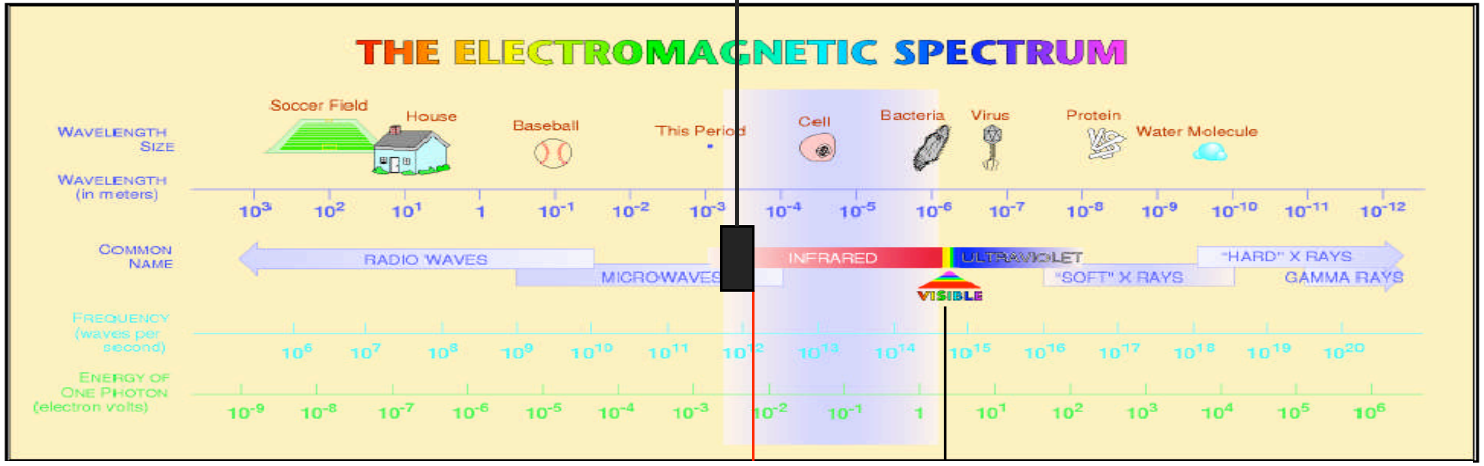
Outline

- THz gap
- Overview of existing and planned THz sources
- THz Science

Electromagnetic Spectrum

The “THzgap”, Collective Excitations in
Macromolecules and exotic electronic materials



IR Units: 200 cm^{-1}=300 K= 25 meV = 50 μm = 7 THz

FIR MIR NIR

Phonons;
Drude absorption;
Gaps in superconductors;
Molecular Rotations;

Molecular Vibrations
Fingerprints for
Chemistry, Biology,
And Geology

Molecular Overtones and
Combinations bands;
Excitons;
Gaps in semiconductors

The Terahertz gap

0.1 THz – 10 THz

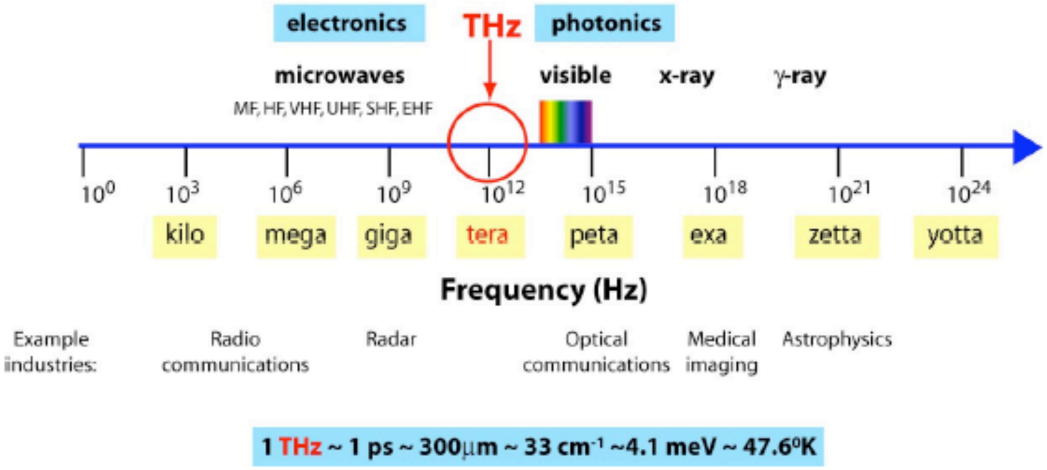
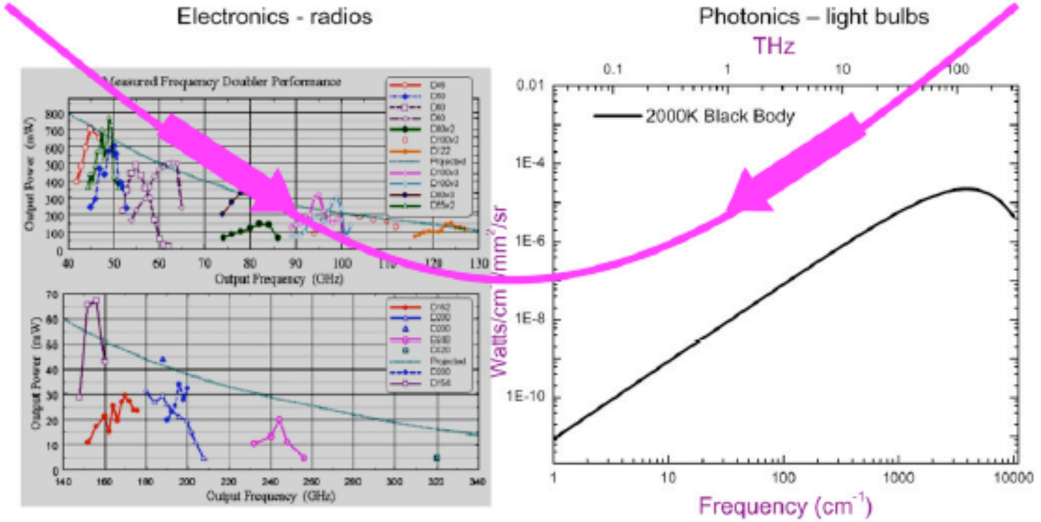


Figure 1. Schematic of the electromagnetic spectrum showing that THz light lies between electronics and photonics.


Electronics - radios

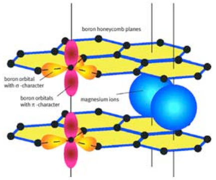
Photonics – light bulbs

No electronics, few microwaves generators

Vanishing thermal power, few tunable and pulsed lasers.

THz Science

Condensed Matter Physics



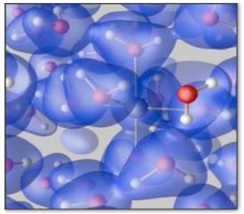
Superconductivity
- Energy gap
- Symmetry of the order parameter
- Direct determination of the superfluid density
- Dynamics of Cooper pairs

Low-dimensional effects
- Dimensionality crossover
- Non-Fermi liquid normal states
- Broken symmetry ground states

Strongly correlated electrons
- Kondo problem
- Heavy fermions

Magnetic Dynamics

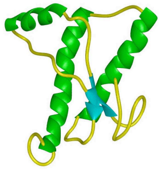
Physical and Analytical Chemistry



Polar liquids
- Hydrogen bond
- Van der Waals interactions
- Acoustic-Optic phonon mixing in water

Solutions
- Static and dynamic interactions between solvated ions and solvent

Life Sciences

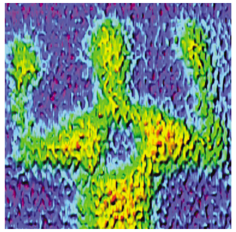


Macromolecules conformation
- Secondary and tertiary structure
- Coherent dynamic development

Metabolism
- Water distribution
- Ionic channel dynamics

Imaging
- 3D tomography of dry tissues
- Near-field sub-wavelenght spatial resolution

New Technologies



THz technologies
Array THz detectors
Metamaterials

Medical diagnostic
- Skin cancer detection

Industrial production
- Material inspection
- Production line monitoring

Defense industry/Homeland security
- Detection of explosives and biohazards

Available THz sources

Terahertz Lasers

- Quantum Cascade Lasers (>2 THz),
- Si- and Ge-lasers (>1.5 THz),
- Far-Infrared FELs: FELIX, FELBE,
- Gas-based lasers (only at some given frequency),

Sources based on electron acceleration

Narrow Band

- Stanford (>3 THz),
- ENEA compact FEL (<0.1 THz),
- Far-IR undulator @ FLASH (Hamburg),
- Backward-wave oscillators,

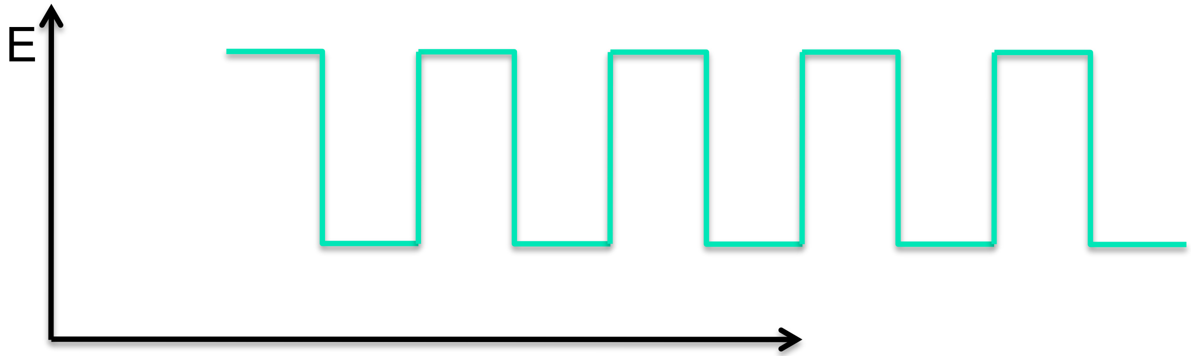
Broad Band

- Laser Amplifiers: plasma and non linear crystals,
- Coherent Synchrotron Radiation @ III Generation Machines,
- LINAC-based Coherent Radiation Sources,

Quantum Cascade Laser

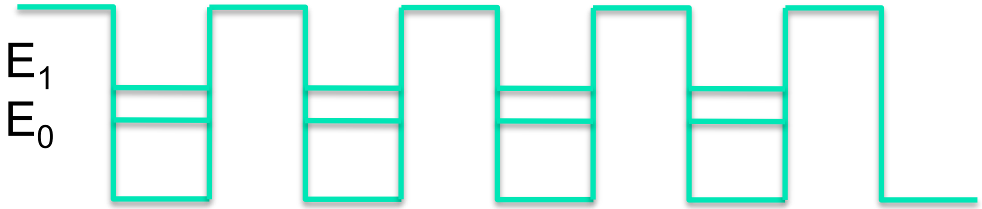
Federico Capasso, Bell Labs

Quantum Wells are periodic potential wells built by semiconductor multilayers

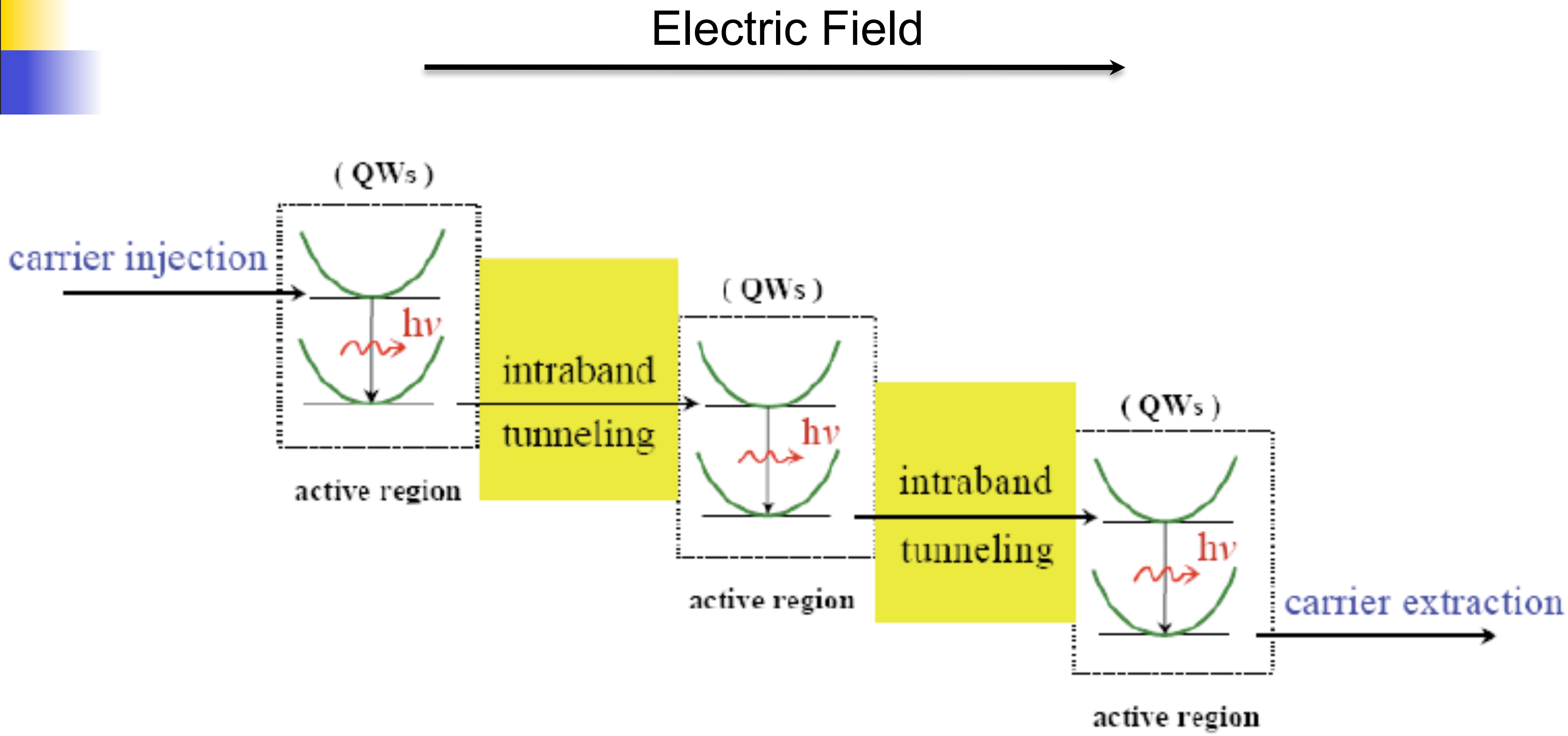


Grown direction of semiconductor multilayers

Through electrons or holes doping electronic states are created in the wells



The energy difference E_1-E_0 can be engineerized from the THz to the mid-IR region



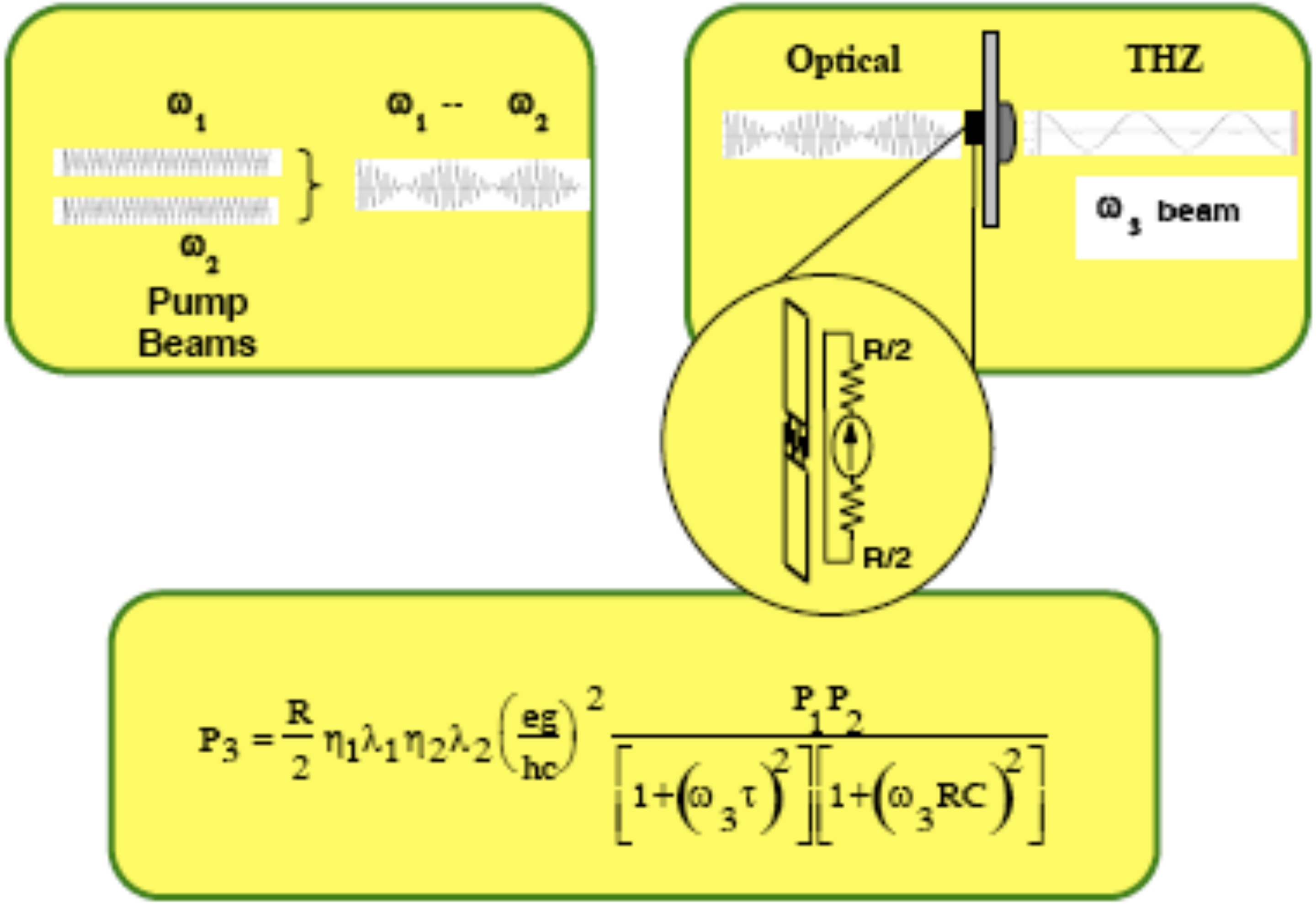
The relaxation from the excite state E_1 to the ground state E_0 produces in-phase photons which are amplified due to the optical cavity effect induces by the different semiconductors constituting the layers

What makes the QC-laser special?

- Wavelength agility
  - layer thicknesses determine emission wavelength
- Demonstrated applications in mid/far-IR gas sensing

- High optical power ~ 1W, room-T operation
  - cascading re-uses electrons
- Ultra-fast carrier dynamics
  - no relaxation oscillations
- Pure TM-polarization – efficient in-plane light coupling
  - Micro-lasers
- Small linewidth enhancement factor
- Intrinsic “design potential”

CW THz Generation by Photomixing or Heterodying

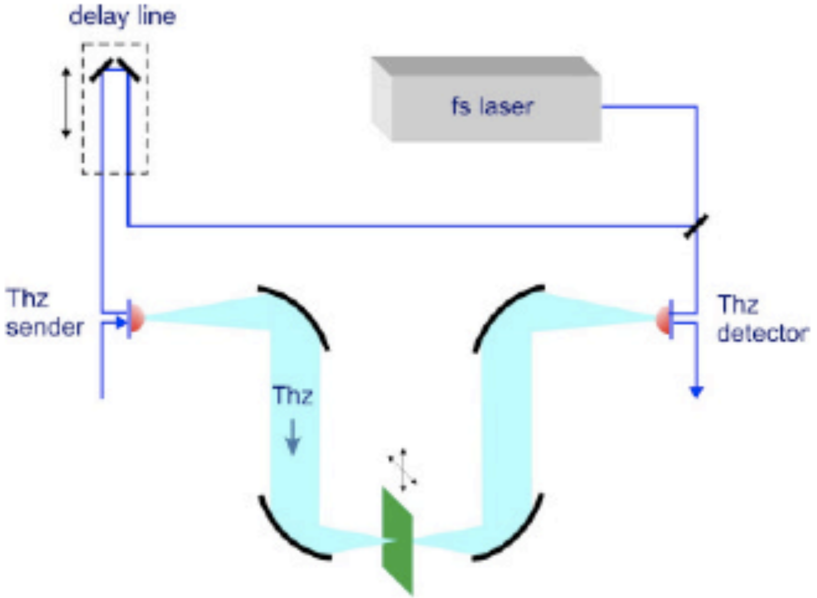


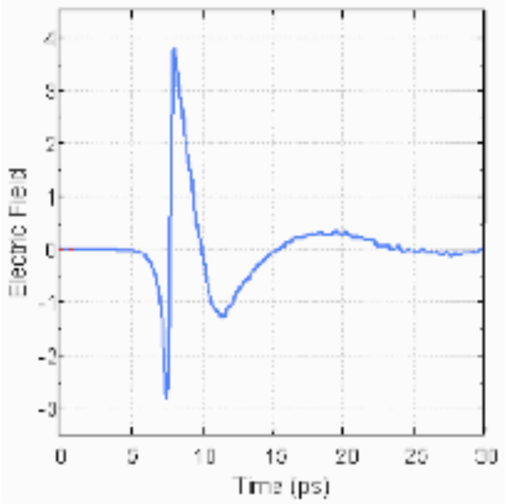
$$P_3 = \frac{R}{2}\eta_1\lambda_1\eta_2\lambda_2\left(\frac{eg}{hc}\right)^2 \frac{P_1P_2}{\left[1+(\omega_3\tau)^2\right]\left[1+(\omega_3 RC)^2\right]}$$

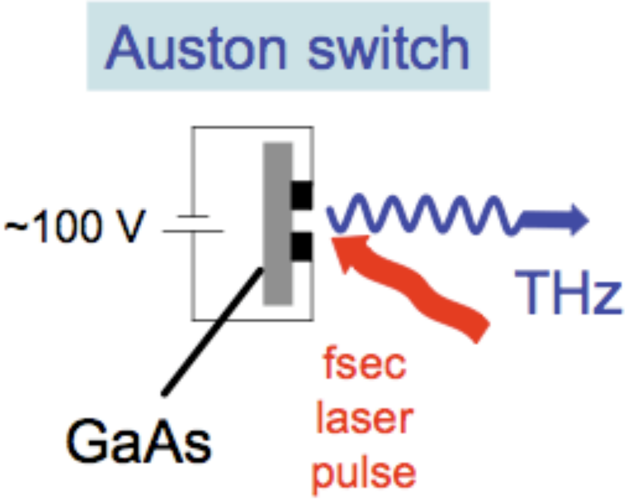
Table-Top Laser Amplifier

Time-domain and Scanning Techniques

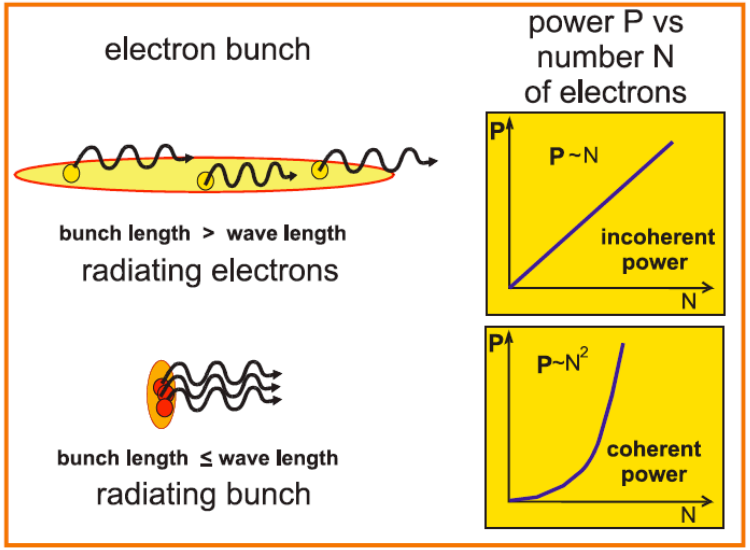
- Based on time domain spectroscopy (TDS)
- Uses recent advances in fs-laser technology.
- Measures intensity and phase of E-field vs time of a single cycle THz pulse
- Typically 0.2 – 2.5 THz, depending on fs laser







Coherent Synchrotron Radiation (CSR)



electron bunch

bunch length > wave length

radiating electrons

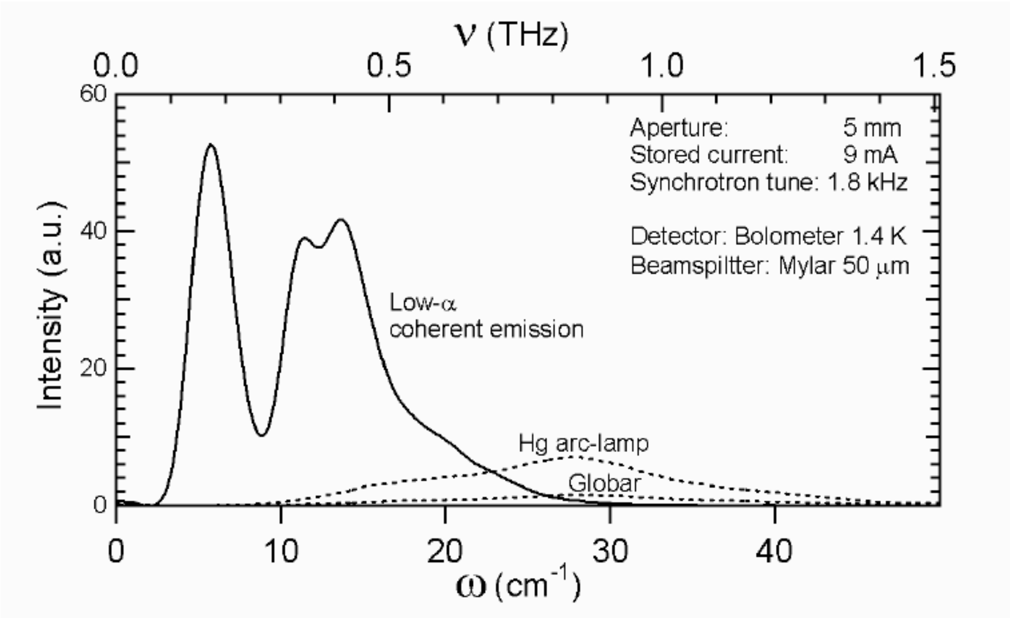
bunch length ≤ wave length

radiating bunch

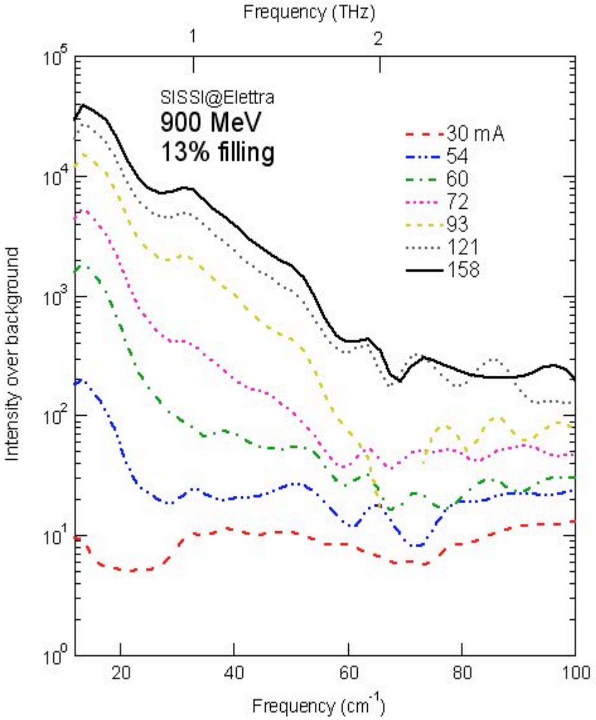
power P vs number N of electrons

$$I = I_{incoh} + I_{coh} = (N(1-f_{\nu}) + N^2 f_{\nu}) I_{incoh}$$

$$f_{\nu} = \left| \int n(z) e^{i\pi\cos(\theta)z} dz \right|^2$$



Low-α mode IRIS@Bessy-II: U. Schade et al, PRL 2003

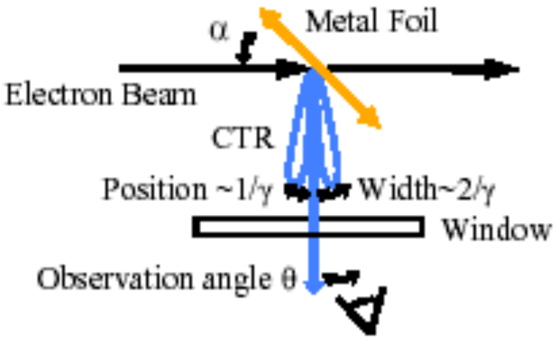


low-e beam energy

SISSI@Elettra: E. Karanzoulis, A. Perucchi, S.L et al, ,2007

Transition THz Radiation (CDR/CTR)

Transition Radiation occurs when an electron crosses the boundary between two different media



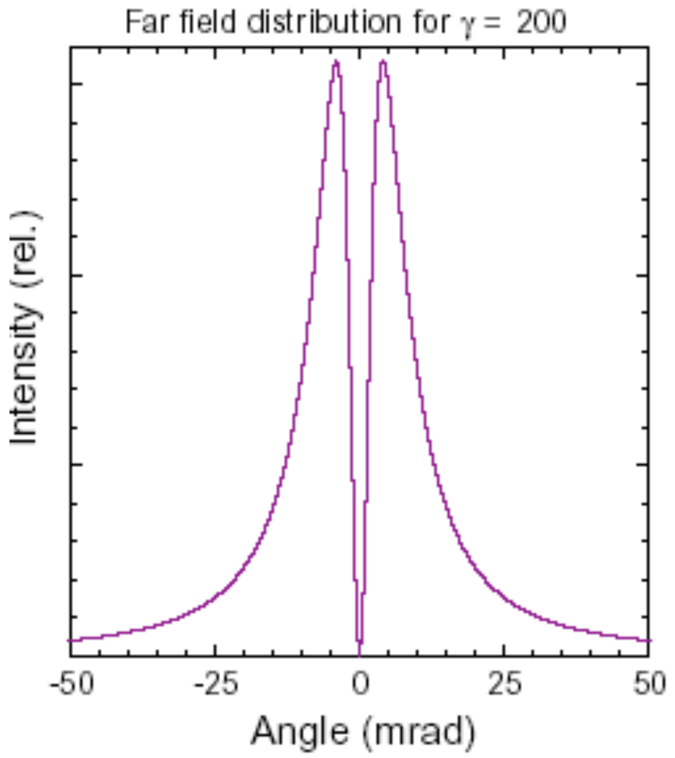
N$_e$ = number of electrons in single bunch
σ = bunch-length

- Coherent Transition Radiation

$$\frac{dI_{CTR}}{d\omega d\Omega}=\frac{e^2\beta^2}{\pi^2 c}\left(\frac{\sin\theta-\beta\cos\alpha}{(1-\beta\sin\theta\cos\alpha)^2-\beta^2\cos^2\theta}\right)^2 N_e\left[1+N_e\exp[-\frac{\sigma^2\omega^2}{c^2}]\right]$$

- Position, width of maxima
- Coherence

Intensity is 0 on axis and peaked at Θ~1/γ
Polarization is radial

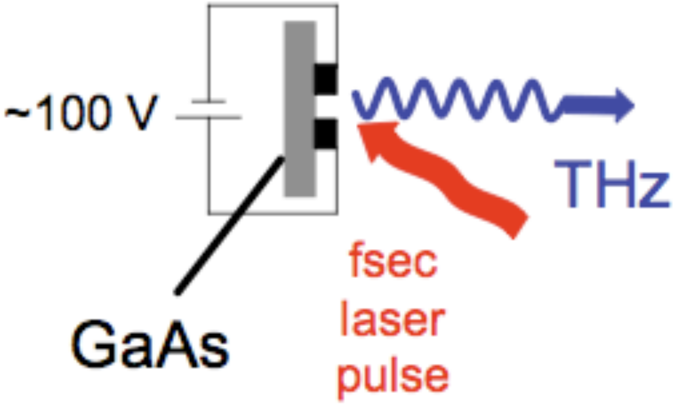


Comparing Coherent THz Synchrotron and Conventional THz Sources

$$\text{Larmor's Formula: Power} = \frac{3e^2a^2}{2c^3}?^4 \ \text{(cgs units)}$$

a=acceleration
c=vel. of light
γ=mass/rest mass

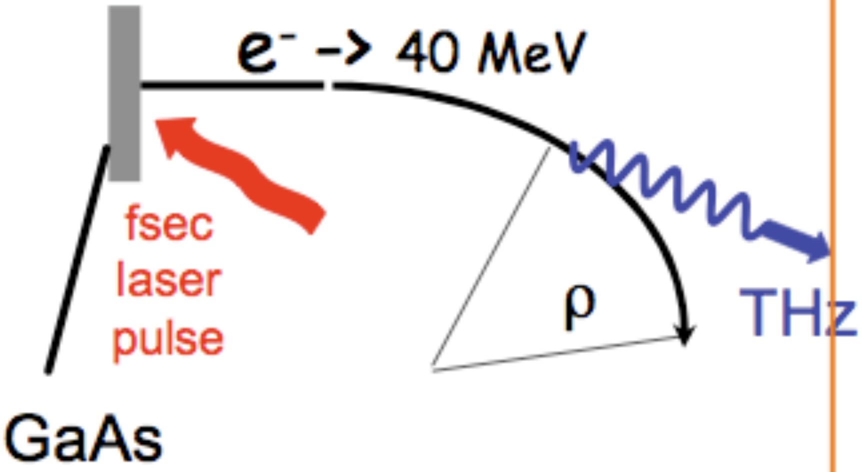
Auston switch



$$E = \frac{100V}{10^{-4}m} = 10^6 \, V/m$$

$$a = \frac{F}{m} = \frac{10^6 V \cdot e/m}{.5MeV/c^2} = \frac{10^6(3\times10^8)^2}{0.5\times10^6} \cong 10^{17} \, m/sec^2$$

Synchrotron radiation



$$a = \frac{c^2}{?} = \frac{(3\times10^8)^2}{1} \cong 10^{17} \, m/sec^2$$

$$\text{if } ? = 1 \ m$$

Comparing Coherent THz Synchrotron and Conventional THz Sources

$$\text{Larmor's Formula: Power} = \frac{3e^2a^2}{2c^3}?^4 \text{ (cgs units)}$$

$$\boxed{\text{Power} \propto \gamma^4}$$

Synchrotron

THz Antenna

$$\gamma = 1$$

To compare radiation in the THz region, ~40 MeV electrons will get the critical energy into the IR. So,

$$\gamma \approx 75$$

$$\gamma^4 \approx 10^7!$$

Relativistic electrons gain a huge factor in THz power.

Figures of Merit of THz Sources

Dedicated Broad Band Sources	Bandwidth	Pulse Width	Rep. Rate	Average Power	Pulse Energy	Peak E-Field
Linac (BNL)	2 THz	300 fs	10 Hz	mW	100 mJ	1 MV/cm
ER-Linac (Jeferson Lab.)	2 THz	300 fs	75 MHz	100 W	μJ	10 KV/cm
Storage Ring (BessyII, Elettra)	1 THz	1 ps	500 MHz	< W	<nJ	< 10V/cm
Table-top Laser	5 THz	100 fs	1 KHz	nW to μW	nJ to μJ	10 KV/cm

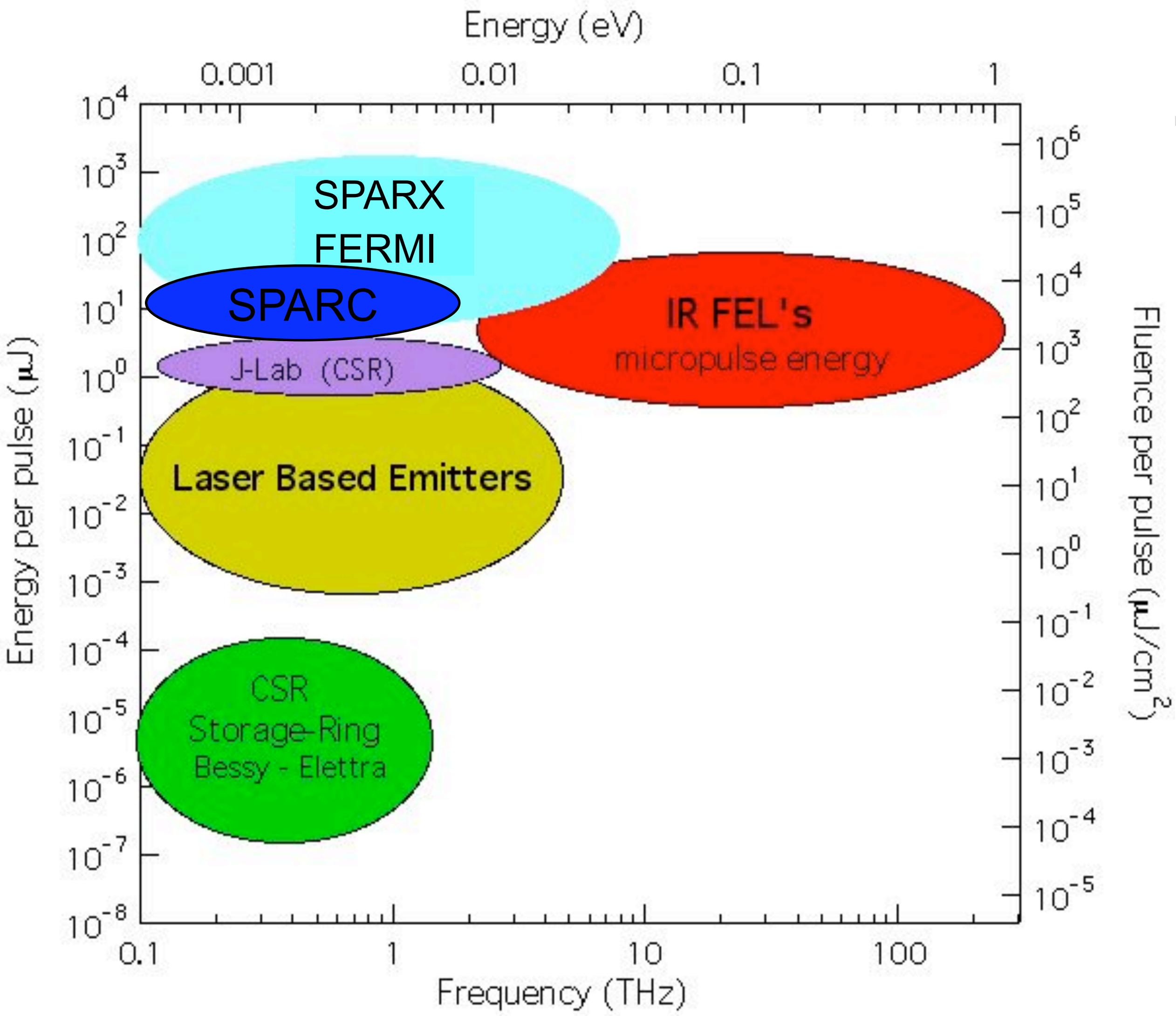
Narrow Band Sources	Op. Range	Pulse Width	Rep. Rate	Average Power	Pulse Energy	Peak E-Field
Stanford FEL	3.75 -20 THz	2-10 ps	11.8 MHz	<1 W	μJ	<250 kV/cm
UCSB FEL	0.88-4.8 THz	/	/	5-100 mW	/	<70 kV/cm
UCSB FEL	0.12-0.88 THz	/	/	5-100 mW	/	<20 kV/cm
FELIX	1.2-100 THz	6-100 cycles	1000/50/25 MHz	<1 W	1-50 μJ	<10 V/cm
ENEA FEL CATS	0.09-0.15 THz	50 ps	3 GHz	<1 W	0.5 μJ	<10 V/cm
FELBE	1.5.100 THz	1-10 ps	13 MHz	50 mW@2THz	0.01-2 μJ	10 kV/cm

Continous wave Sources	Op. Range	Average Power
Backward Wave Oscillator	0.1-1.2 THz	1 mW@1.2 THz
Gas Lasers (CO2-pumped)	not cont. tunable	1 W@2.5 THz
Quantum Cascade Lasers	2-10 THz	10 mW@3.5 THz

IV UV-X FEL	Bandwidth	Pulse Width	Rep. Rate	Average Power	Pulse Energy	Peak E-Field
SPARX-FERMI-SPARC (long bunch)	1 THz	1 ps	0-50 Hz	1-5 mW	100 μJ	MV/cm
SPARX-FERMI (medium bunch)	2 THz	500 fs	0-50 Hz	2-7 mW	150 μJ	MV/cm
SPARX-FERMI (short bunch)	10 THz	50 fs	0-50 Hz	>10 mW	>mJ	10 MV/cm

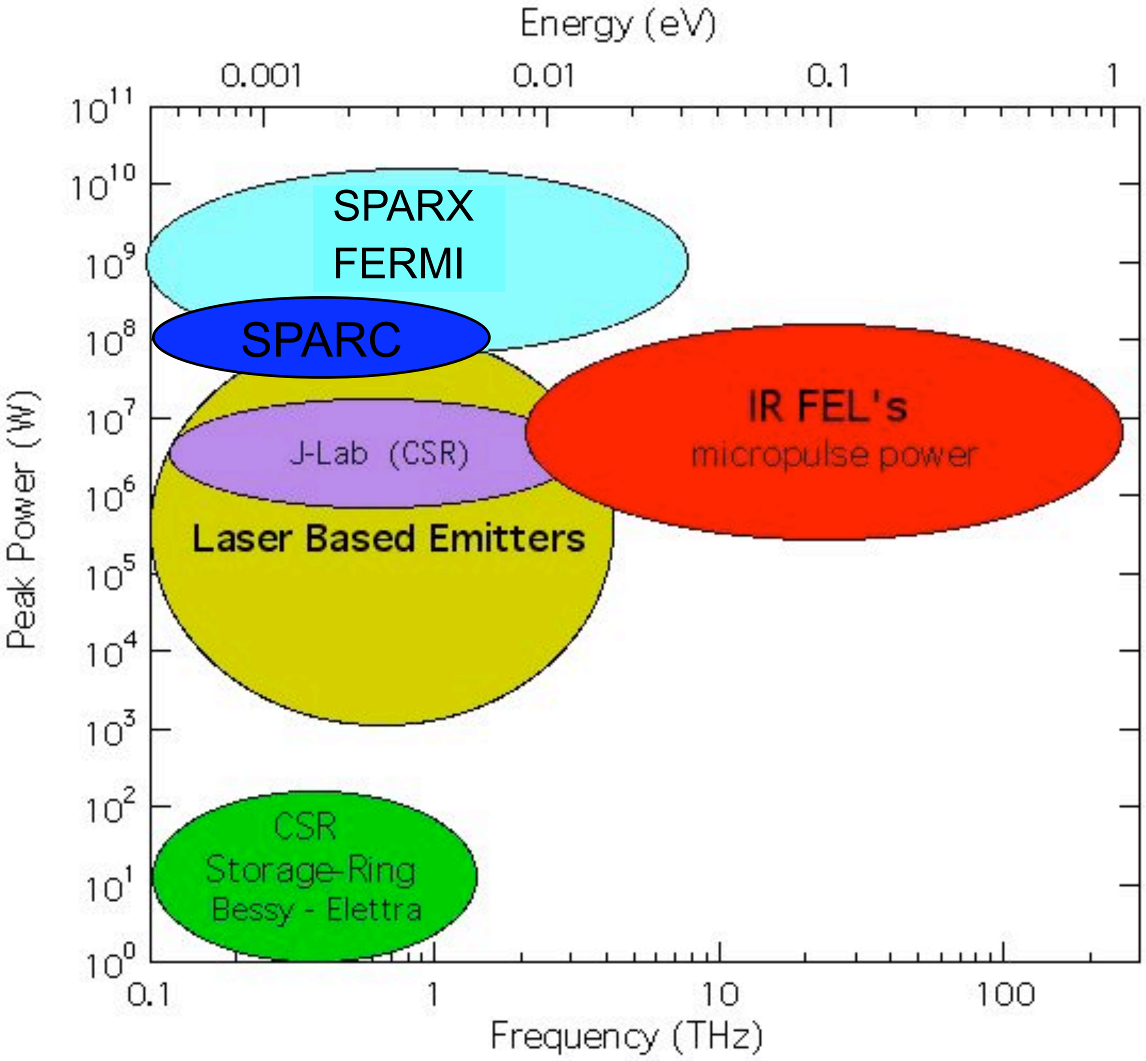
Data from: Biedron et al 2007; Riemann 2007; A. Perucchi et al 2007

Figures of Merit of THz sources: energy/pulse



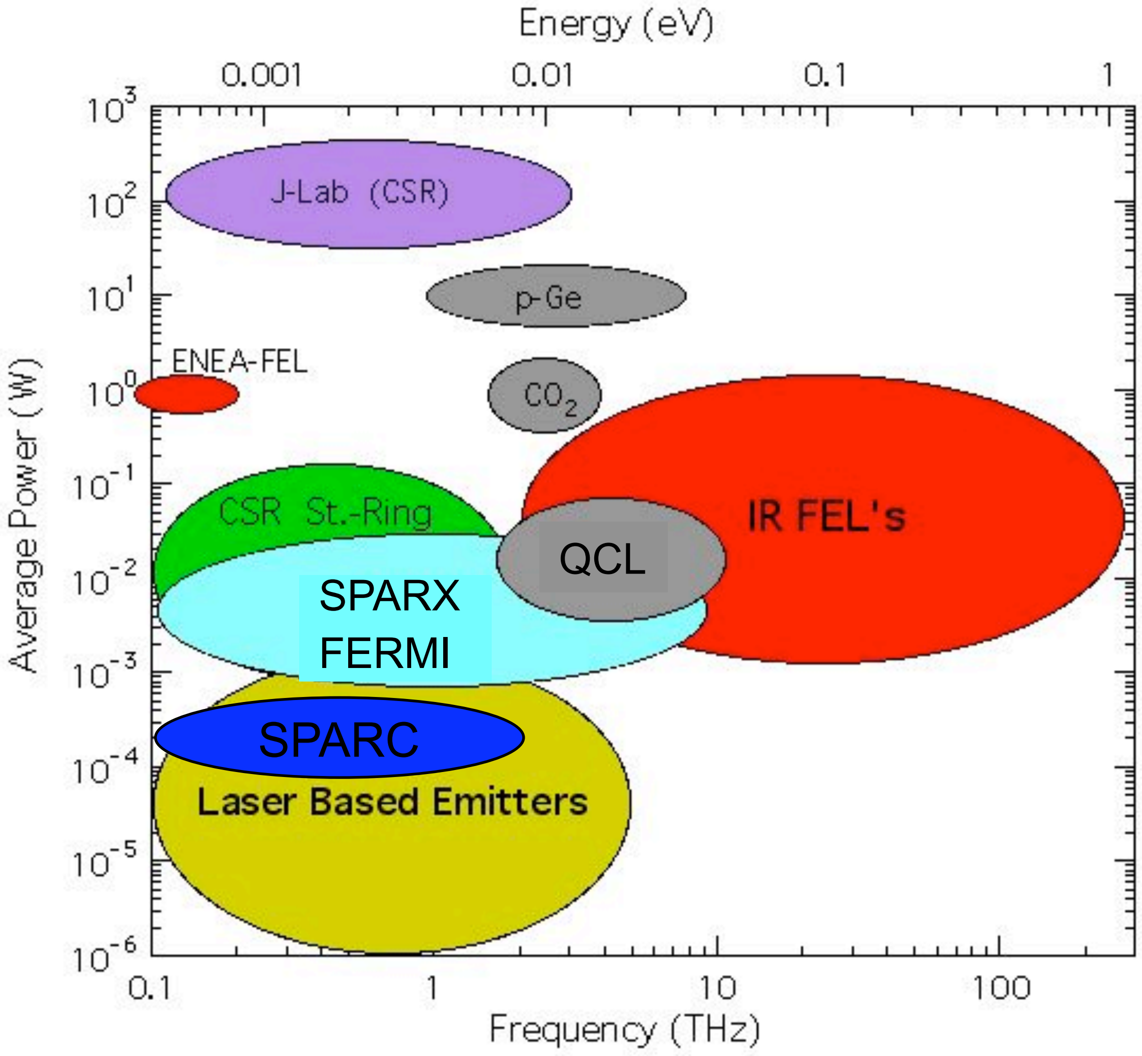
A. Perucchi et al, 2008

Figures of Merit of THz sources: peak power



A. Perucchi et al, 2008

Figures of Merit of THz sources: average power



A. Perucchi et al, 2008

THz Science

Linear Spectroscopy

Superconductivity today:

1. Looking for new materials
   - 1986: High-T_c cuprates
   - 1990: C_{60} fullerene
   - 2000: MgB_2
   - 2003: $Na_xCoO_2\cdot 3H_2O$
   - 2004: B-doped diamond
2. Understanding new materials

↓

THz spectroscopy plays a role...

... because

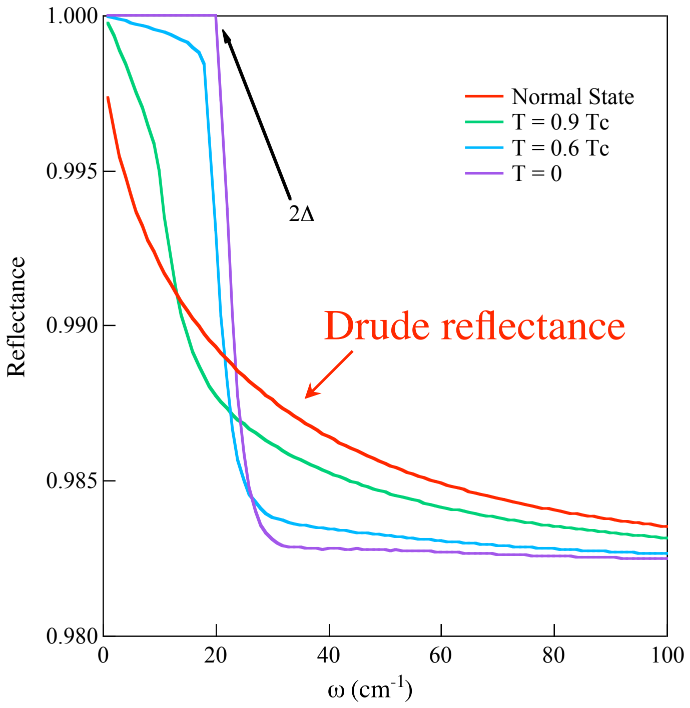
Superconductivity is ruled by low-energy electrodynamics:

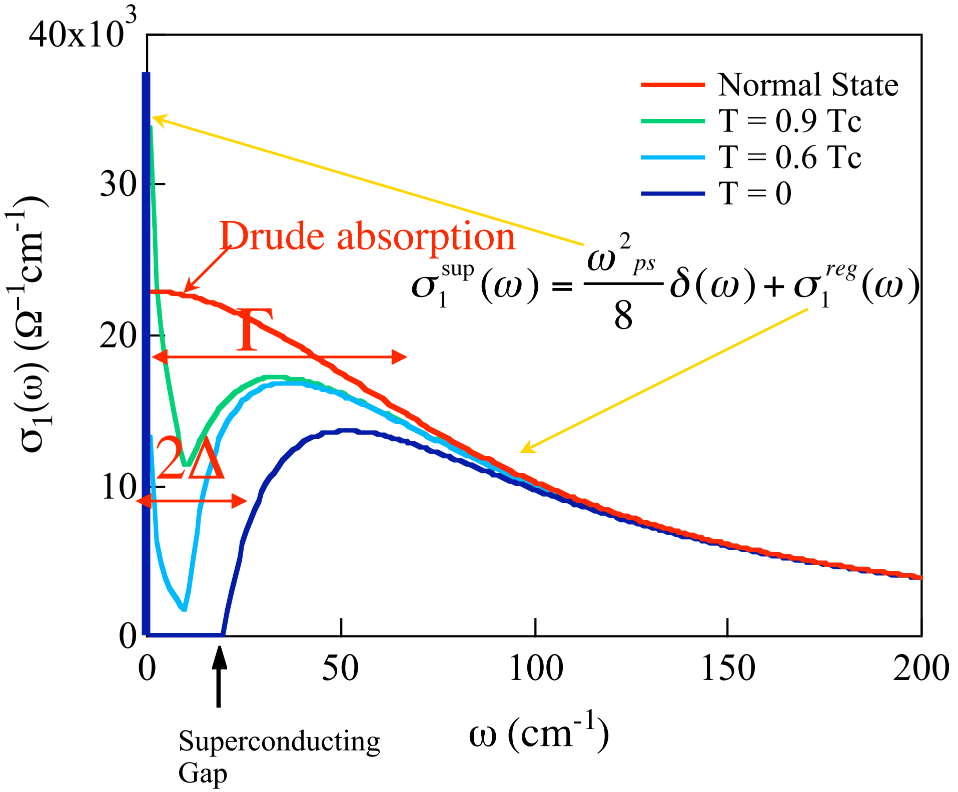
- Superconducting gap : THz-range
- Spectral weight of condensate and penetration depth:THz-range
- Mediators of pairing (phonons, etc.): Far-infrared
- Free-carrier conductivity above T_c: Infrared

Basic optics of Superconductors

Minimum excitation energy:
Cooper-pair breaking 2Δ

Superconducting gap observed if:
- sample in the dirty-limit ($2\Delta < \Gamma$)
- Cooper pairs in s-wave symmetry





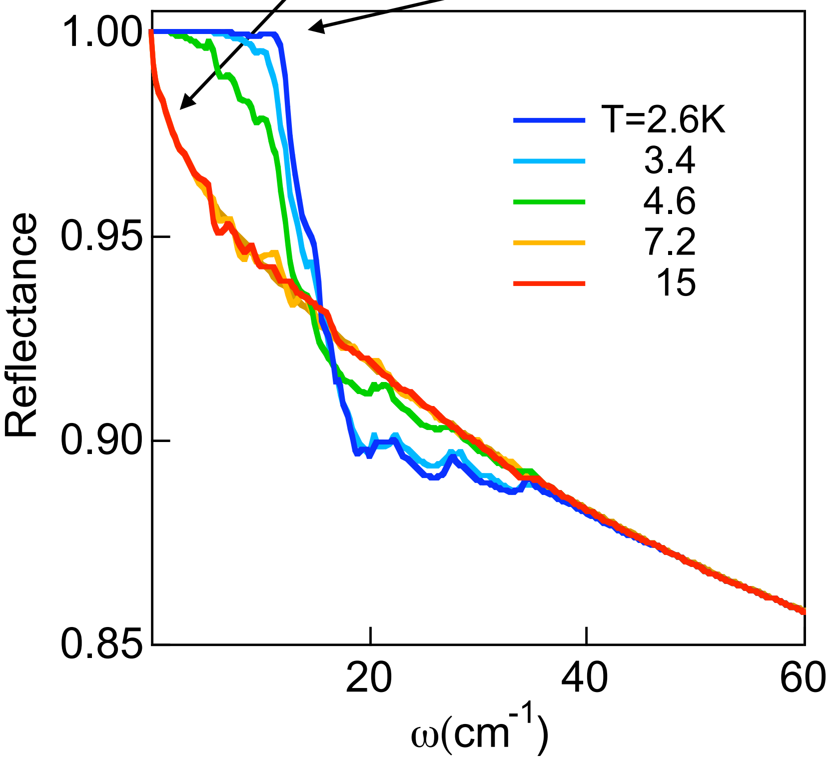
$$\int[\sigma_1(\omega, T>Tc) - \sigma_1(\omega, T<Tc)]\, d\omega = \omega^2_{ps}/8 = n_s e^2/m^* \dashrightarrow \lambda = c/\omega_{ps}$$

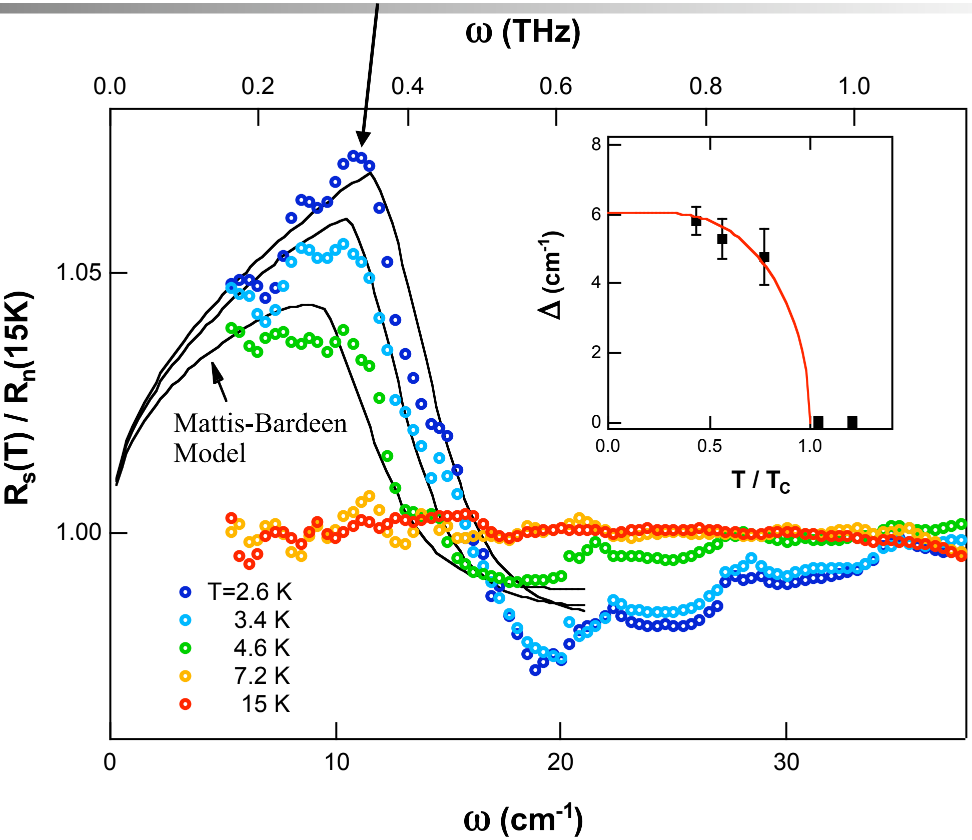
Ferrel-Glover-Tinkham Rule

Superconducting Diamond

$\omega \leq \Gamma(T)$: $R_n(\omega) = 1 - [8\omega\Gamma(T)/\ \omega_p^2]^{1/2}$ $\omega \leq 2\Delta(T)$: $R_s(\omega) = 1$

Peak at 2Δ in Rs/Rn





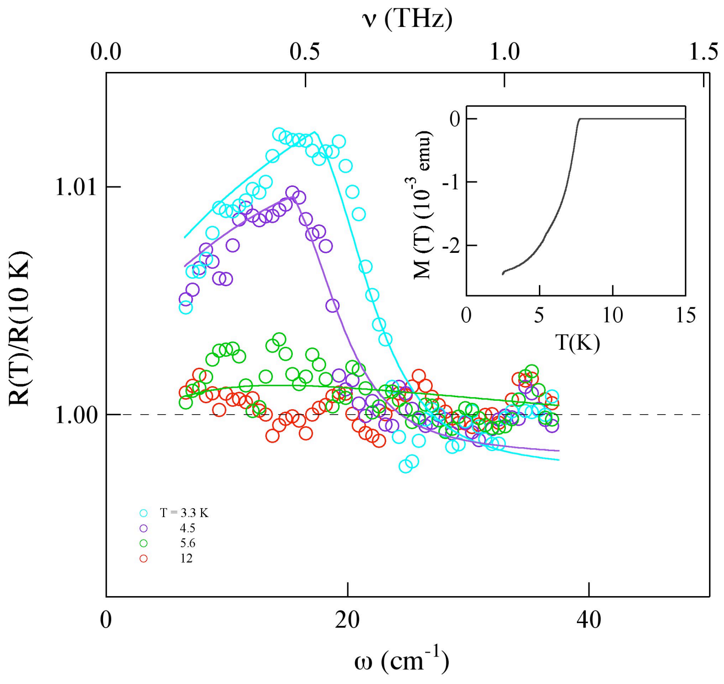
s-wave Dirty-Limit Regime; $2\Delta(2.6\ K)=12\pm1\ cm^{-1}$----> $2\Delta/k_BT_C=3.2 \pm 0.5$

M. Ortolani, S. L. et al, PRL 97, 097002 (2006)

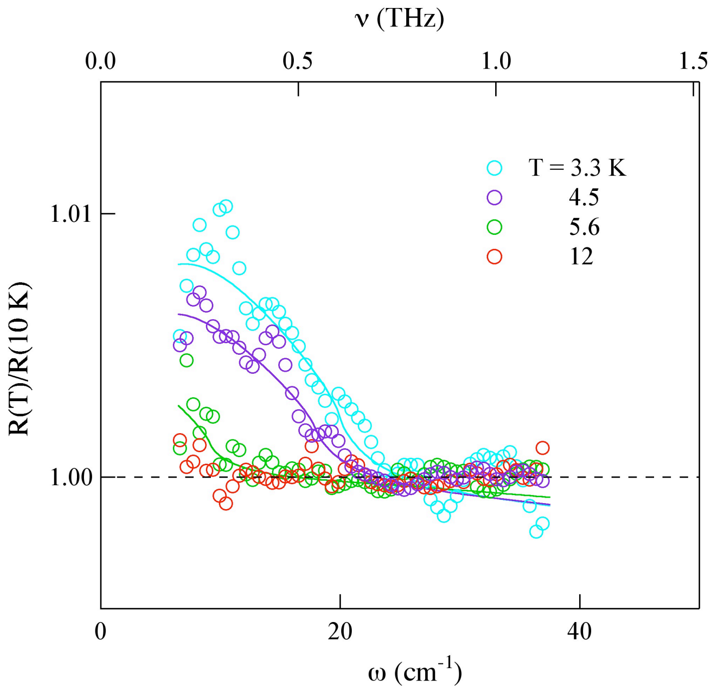
CaAlSi (anisotropic superconductor)

polarized gap measurements

exagonal plane



c-axis

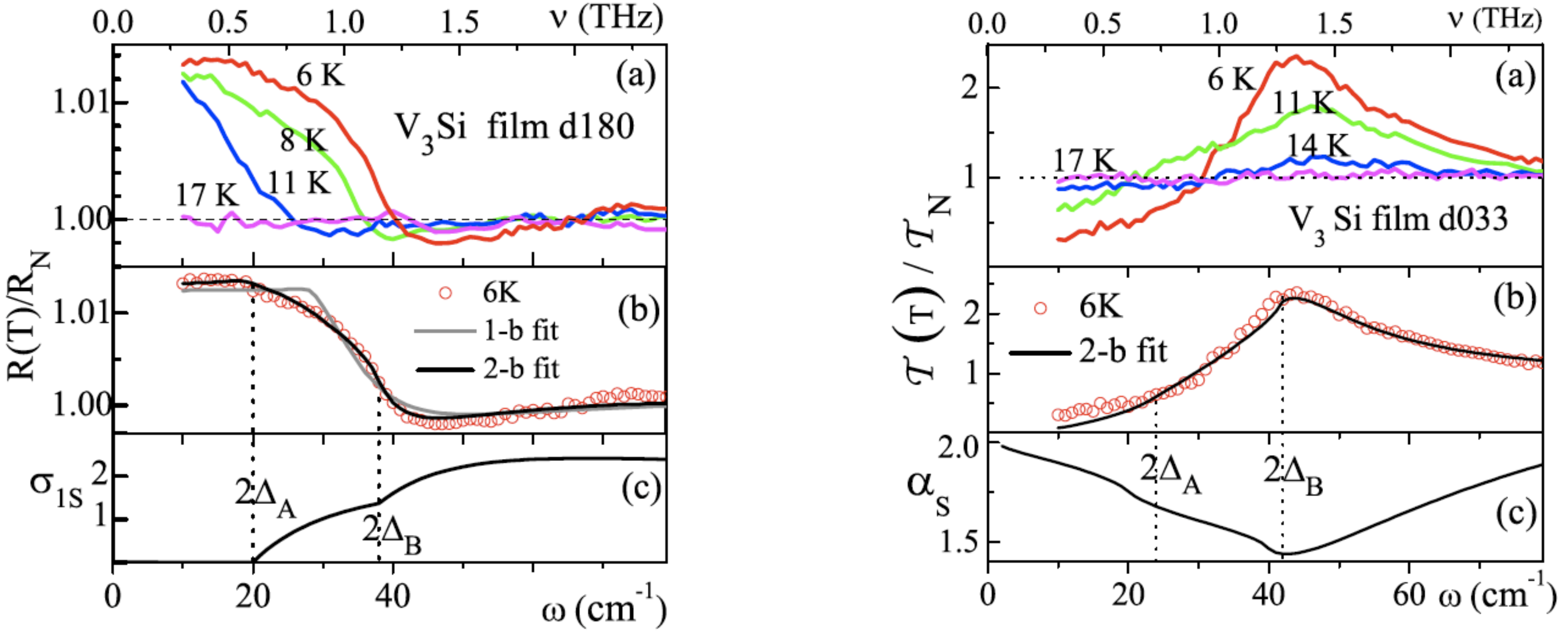


s-wave Dirty-Limit Regime both along the exagonal plane and along the c-axis:

$2\Delta_{ab}(0\ K)=19\pm1\ cm^{-1}$ → **$2\Delta_{ab}/k_BT_C=3.9\pm0.2$**
$2\Delta_{c}(0\ K)=23\pm1\ cm^{-1}$ **$2\Delta_{c}/k_BT_C=4.5\pm0.2$**

S. Lupi et al, PRB 2007

Multigap Superconductivity: V3Si



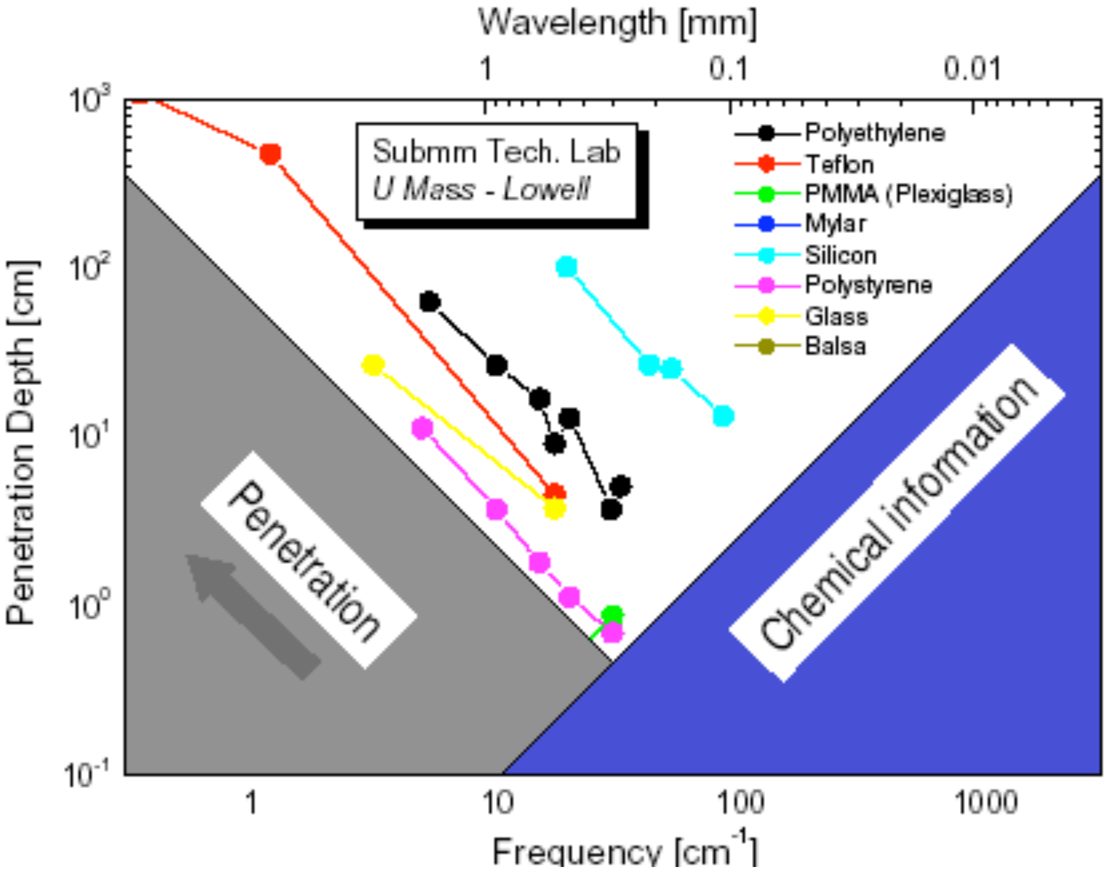
THz transmittance and reflectance demonstrate the presence of two BCS gaps $2\Delta_1$=0.71 THz, $2\Delta_2$=1.27 THz

A. Perucchi et al, PRB 2010

THz Science

Imaging

Imaging vs Penetration



L. Carr et al, 2006

Chemical Pharmaceutic recognition

Fig. 8. *Visible image of sample with four pellets containing different chemicals: (1) lactose, (2) aspirin, (3) sucrose, and (4) tartaric acid.*

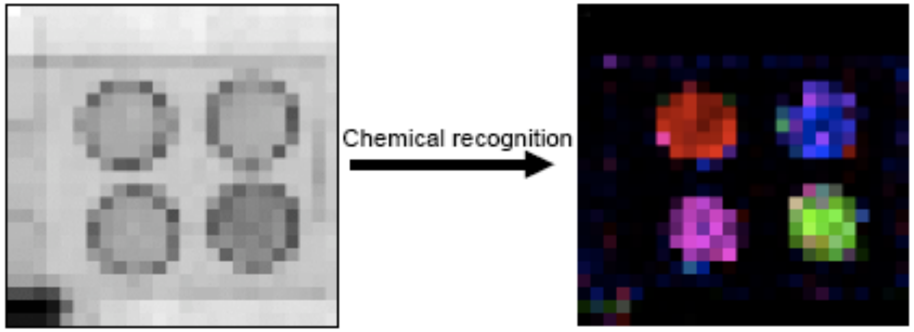
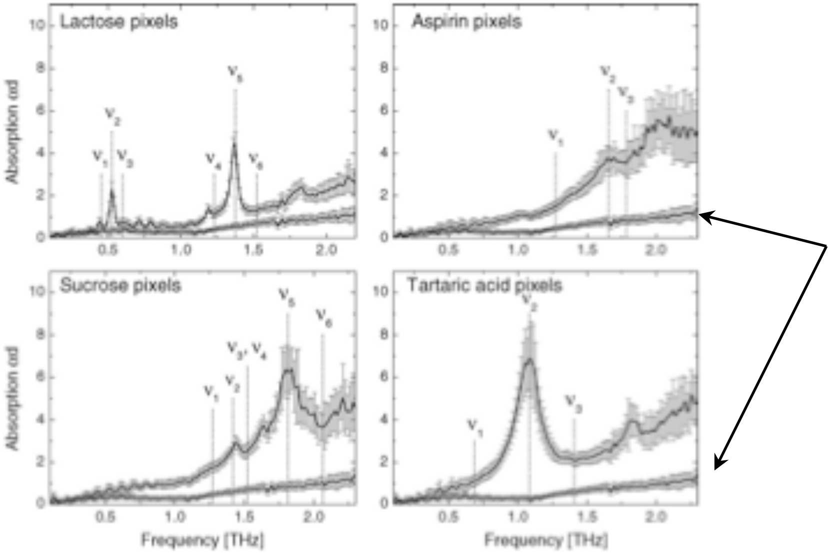


Figure 9.2 The left panel shows a traditional THz transmission image of four pills containing different chemicals (lactose, aspirin, sucrose, and tartaric acid). The right image has color-coded according to the spectral information contained in the full THz trace recorded at each pixel, and is known as "functional imaging". Figure courtesy of Y. Watanabe et al., Yamagata University.



Background absorption

Fig. 10. *Solid lines show the average absorption of lactose (top, left), aspirin (top, right), sucrose (down, left), and tartaric acid (down, right) in the sample. The lower curve in each panel shows the absorption of the packaging material. The error bars represent one standard deviation from the mean of typically 20–30 measurements. The indicated frequencies are used for chemical recognition. After [20].*

Imaging of Bio-materials, molecular in-vivo imaging of pathogenesis

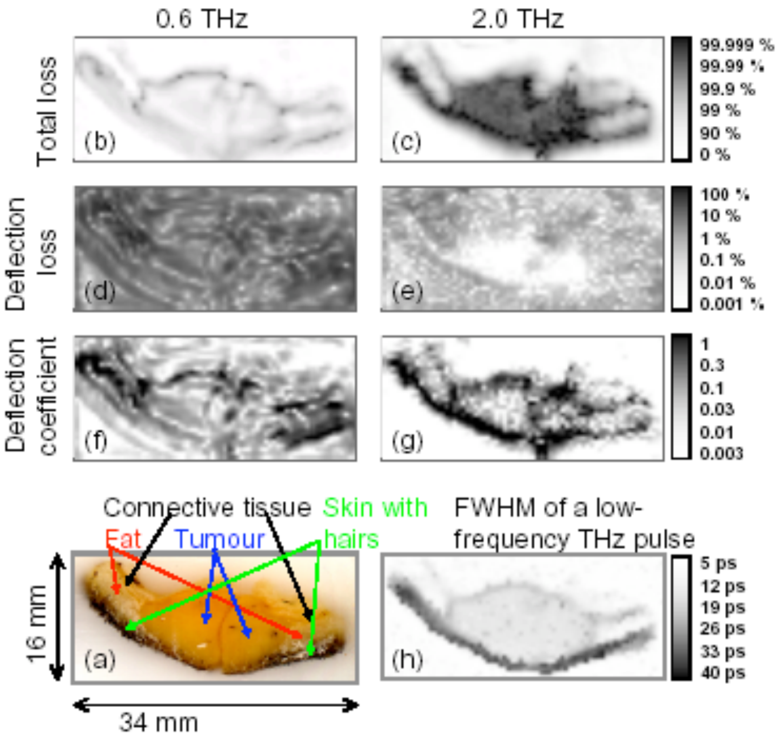


Fig. 3. (a) Optical image of the sample; (b) and (c): Total loss in transmission, (d) and (e): Loss induced by deflection; (f) and (g): Deflection coefficient. (h) Pulse duration (FWHM) of a low-frequency THz pulse. Click on Fig. 3(b,d,f) to see the data as a function of the frequency. (426 kB QuickTime movie)

T. Lffler et al, Optics Express 9, 616 (2001)

Ferguson et al, Nature Materials1, 26 (2002)

X.-C Zhang Phys. Med. Biol. 47, 3667 (2002)

THz do not subject a biological tissue to harmful radiation and may provide both imaging and spectroscopic information on biological materials.

Needs of:

1. High S/N ratio
2. High acquisition rate and resolution
3. THz database for biological tissues
4. High power to increase sensing and penetration
5. Near-field imaging to increase spatial resolution (up to now 1 microns resolution has been obtained)

PROTOTYPE DESIGN OF T-RAY SCIENCE'S COMPACT SKIN CANCER DETECTION SYSTEM

Quality Assurance of Chocolate Products with Terahertz Imaging

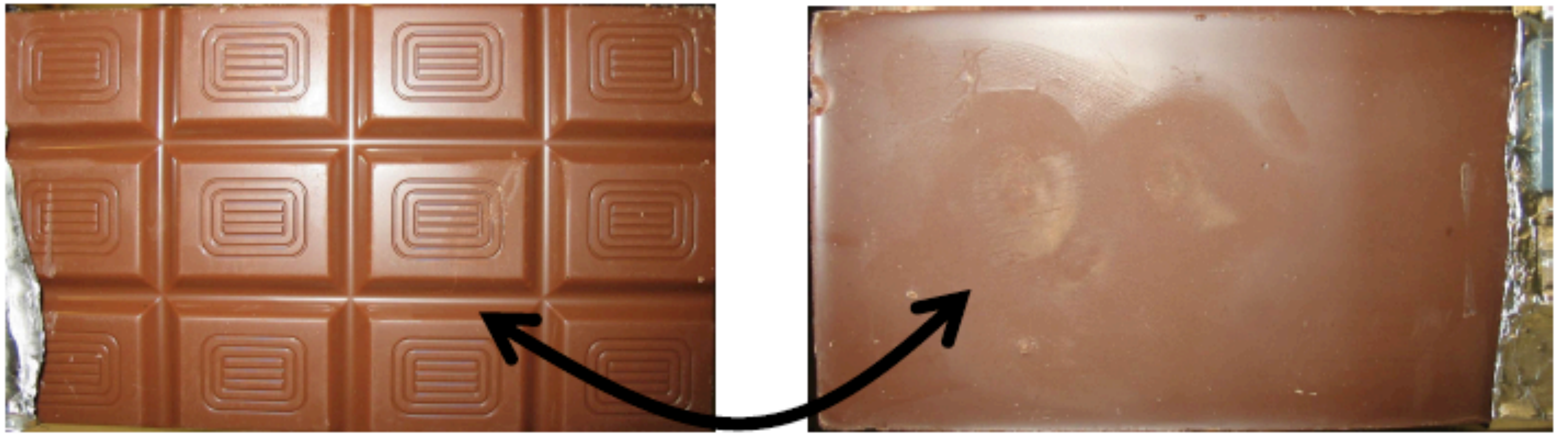
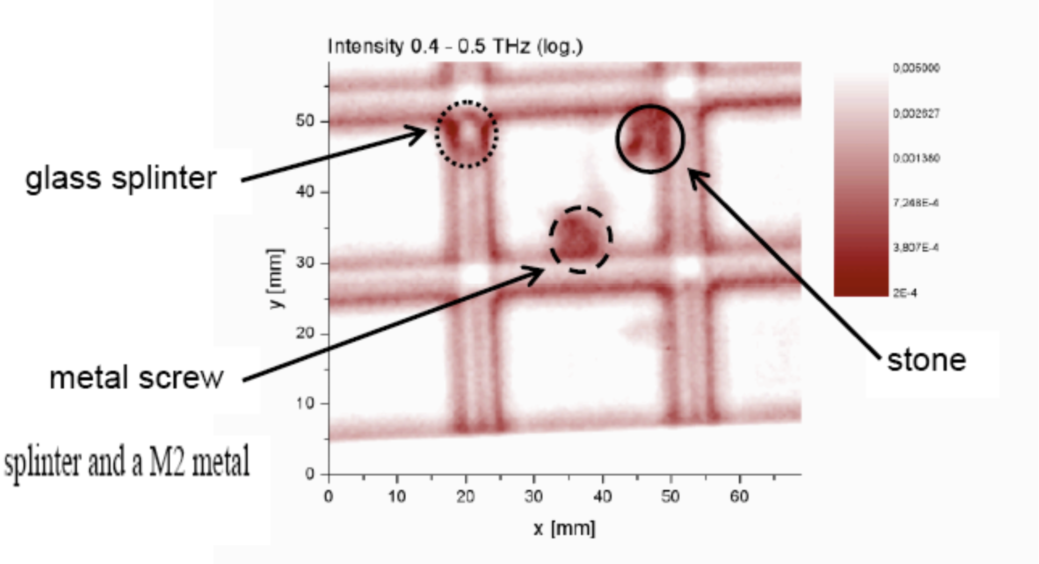


Figure 3. Front and back side of a chocolate bar after artificial contamination with a stone, a M2 metal screw and a glass splinter.

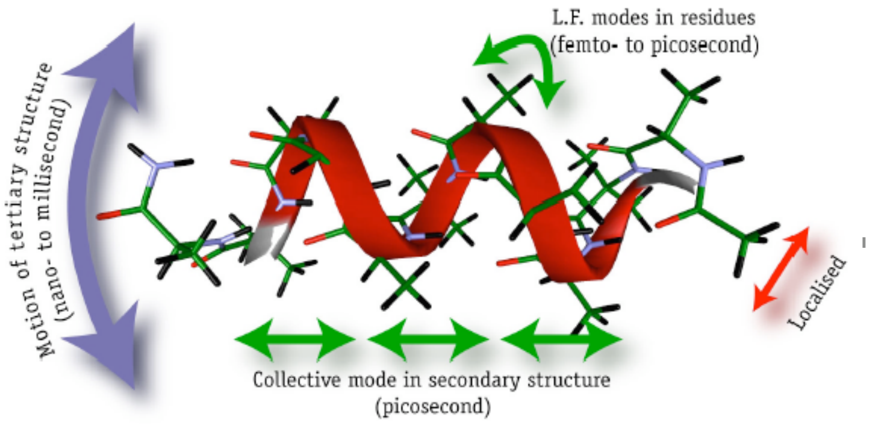
Figure 4. Photograph of different kind of contaminations like a small stone, a glass splinter and a M2 metal screw. The M2 screw may help for size reference.

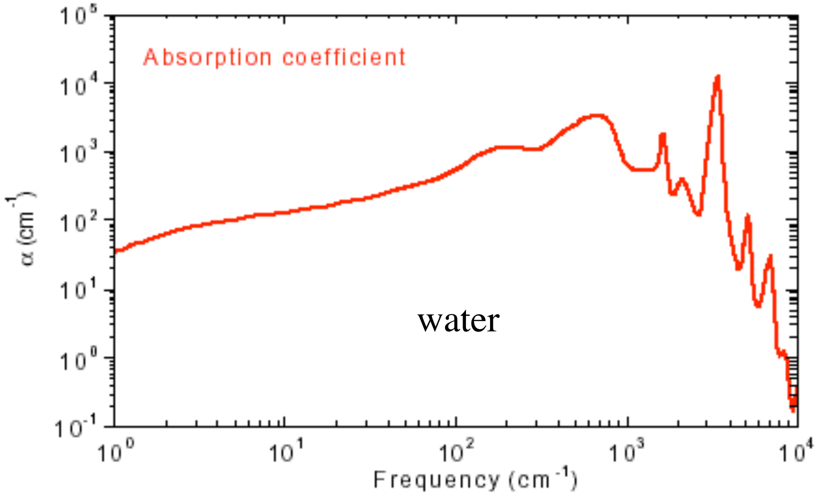


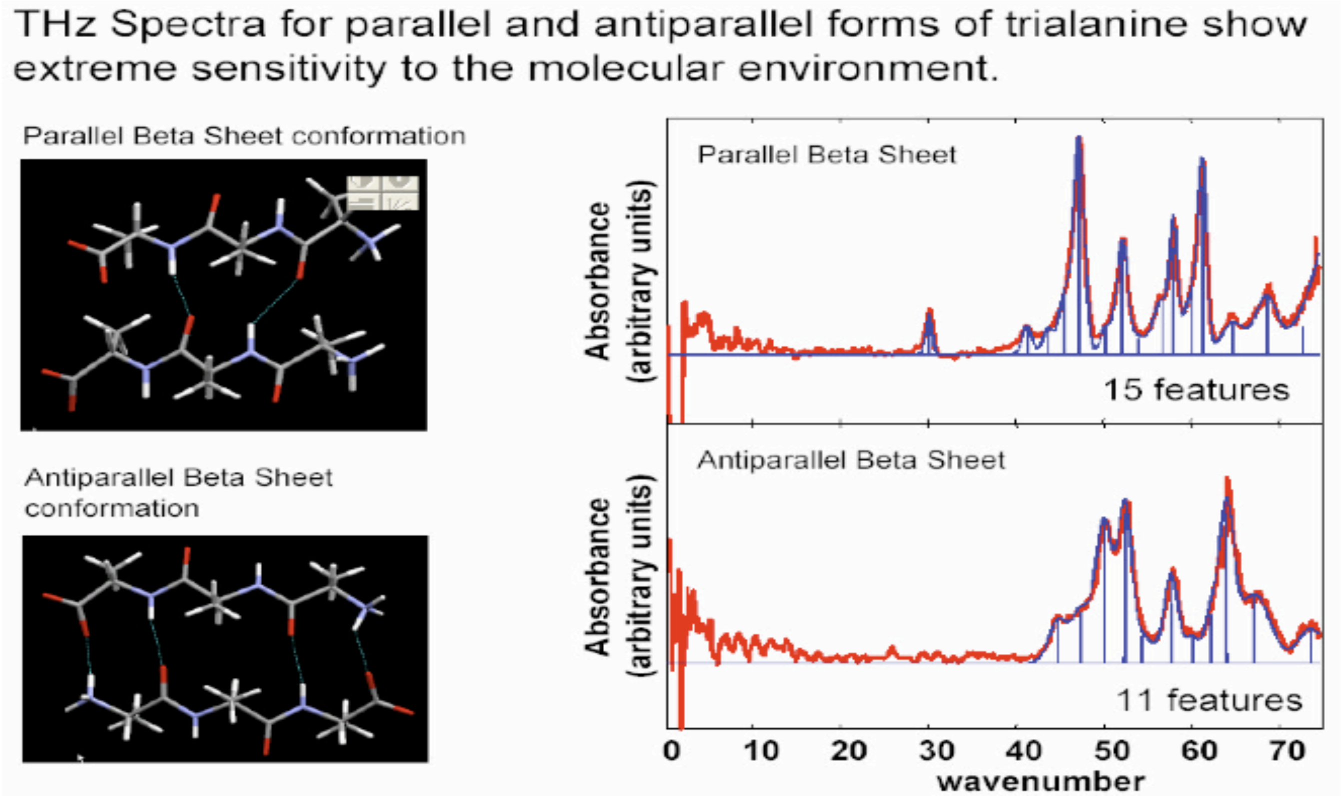
THz Science

Linear Spectroscopy in Life Science

Conformational Collective modes of macromolecules







THz Spectra for parallel and antiparallel forms of trialanine show extreme sensitivity to the molecular environment.

Femto-Chemistry

THz is sensitive to interaction time between a molecule and the surrounding solvent on sub-ps scale

Standard high-frequency time-resolved experiments:
excite in the UV-VIS a soluted molecule -----> probe in the THz the dynamical effects on solvent

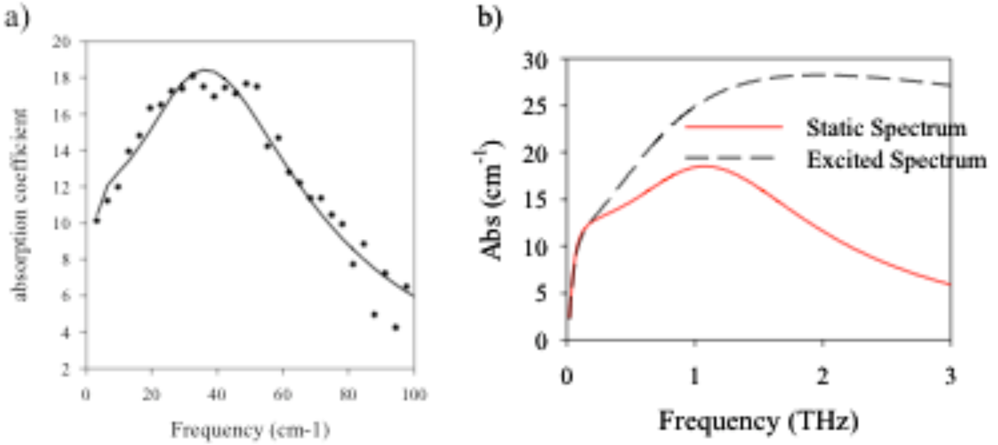


Figure 6.2. Static spectrum of chloroform (part a). Absorption spectrum of collective solvent modes near photoexcited dye molecule (part b). Image courtesy of Charles Schmuttenmaer, Yale University.

THz Science

Pump-Probe Time Resolved Spectroscopy

Pump-Probe Spectroscopy

1) **Optical Pump-Optical Probe Spectroscopy** Pump and Probe pulses (often at a single frequency) fall in the visible (near-IR)

- High energy excitation;
- Strong scattering effects;
- High energy dynamics;

Extrinsic dynamics

2) **Optical Pump-THz Probe Spectroscopy** Pump (single frequency) in the visible (near-IR) Probe in the far-IR and THz range;

- Similar inelastic effects in the Quasi-Particle decay like in (1) but investigation of the low-energy dynamics;

3) **THz Pump-THz Probe Spectroscopy** Accordable Pump pulses falling in the far-IR and THz;

•Possibility to resonate and/or selectionate several fundamental excitations;

•Intrinsic dynamics

4) **THz Pump- IR+VIS SR Probe Spectroscopy**

•Accordable Pump pulses falling in the far-IR and THz;

•Possibility to resonate and/or select several fundamental excitations;

•Intrinsic strongly-coupled different energy scale dynamics

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Exploring the Dynamics of Superconductors by Time-Resolved Far-Infrared Spectroscopy

G. L. Carr*
R. P. S. M. Lobo, J. LaVeigne, D. H. Reitze, and D. B. Tanner

High Energy (Visible) Pump
THz Probe

Many Cooper pairs are broken

→ Extrinsic Dynamics

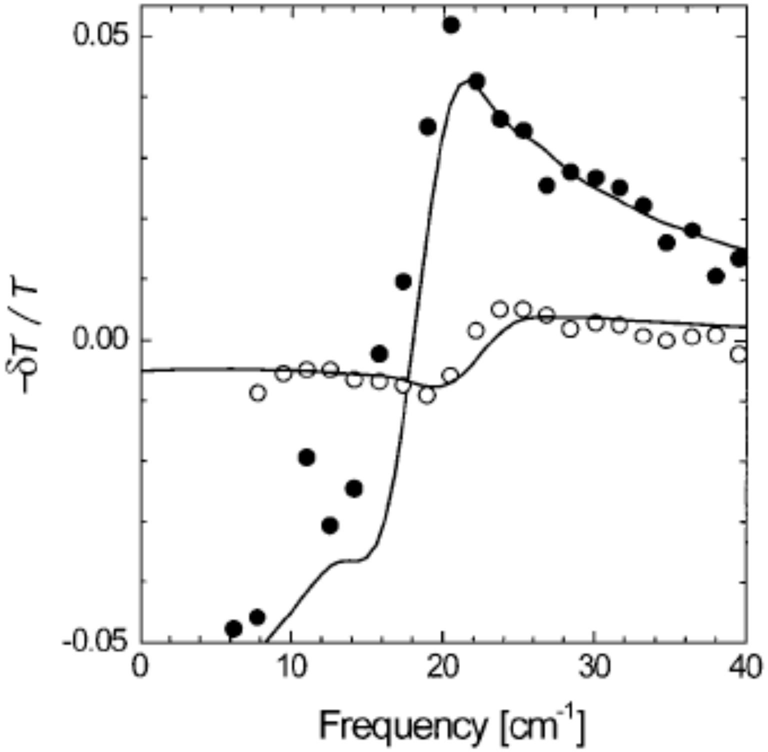

FIG. 3. Photoinduced spectral changes for two samples. The laser pulse energies were 1.8 nJ (solid circles) and 0.4 nJ (open circles). Also shown are fits (solid curves) assuming an ~3% and 0.6% reduction in the energy gap, respectively. All other parameters were held fixed at the values determined from the temperature-dependent T_S/T_N fits.

New paradigm:
THz pump→THz probe

- 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.

High-power half-cycle THz pulses

A 100 μJ, half-cycle THz pulse, focused into a volume of 1 mm^3 or less.

- **E-field** = $[2D_E/\varepsilon_0]^{1/2}$ **~ 10^8 V/m (~ 1 MV/cm)**.
- => Use large electric field to displace atoms in polar solids (structural phase transitions, soft modes, ferroelectricity, …)
- **H-field = E/c ~ 0.3 T**
- => Use transient magnetic field to create magnetic/spin excitations and follow dynamics on ps time scale (e.g., time-resolved MOKE).

Or, some other shape pulse?

$$\left.\frac{dI(\omega)}{d\omega}\right|_{\text{multiparticle}} = [N + N(N-1)f(\omega)]\frac{dI(\omega)}{d\omega} \qquad f(\omega) = \left|\int_{-\infty}^{\infty} e^{i\omega\hat{n}\cdot\vec{r}/c} S(r)\,dr\right|^2$$

=> shape electron bunch profile to control E-field shape (coll. W/J. Neuman, U. Md.)

Coherent Control of Electronic Properties of Manganites

Matteo Rini[1], Ra'anan Tobey[2], Nicky Dean[2], Jiro Itatani[1,3], Yasuhide Tomioka[4], Yoshinori Tokura[4,5], Robert W. Schoenlein[1] & Andrea Cavalleri[2,6]

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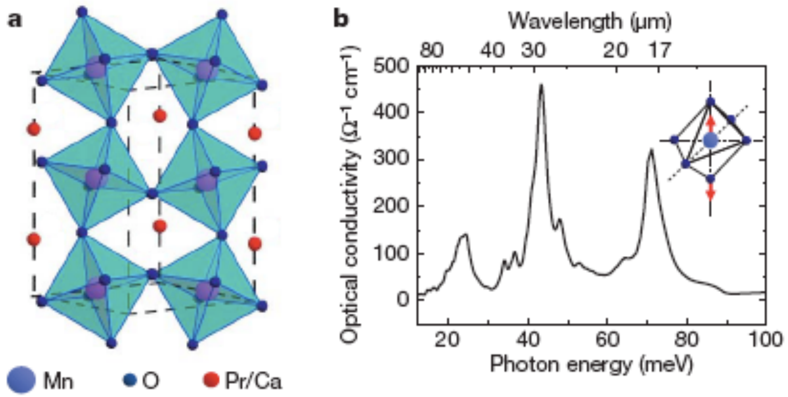
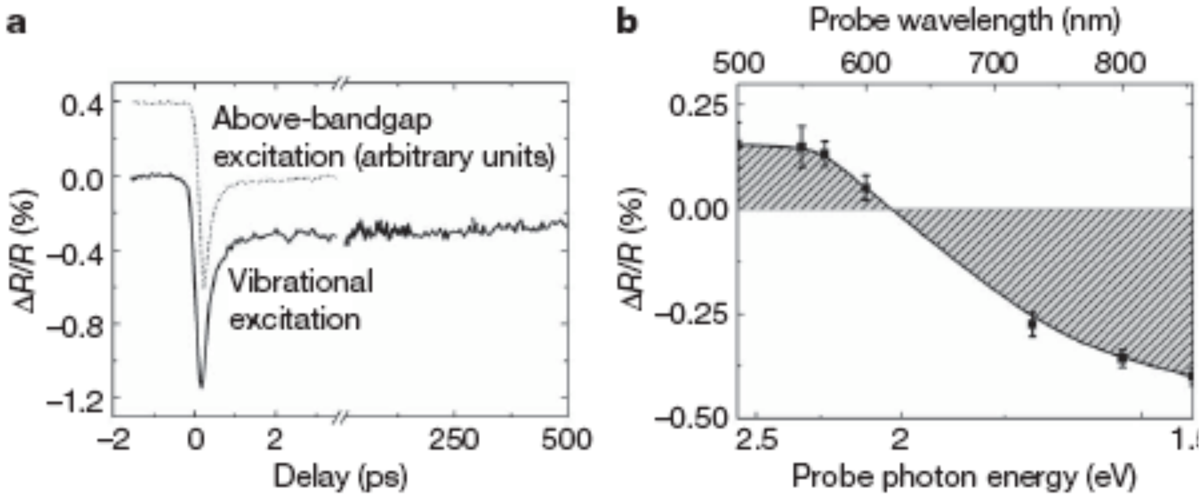
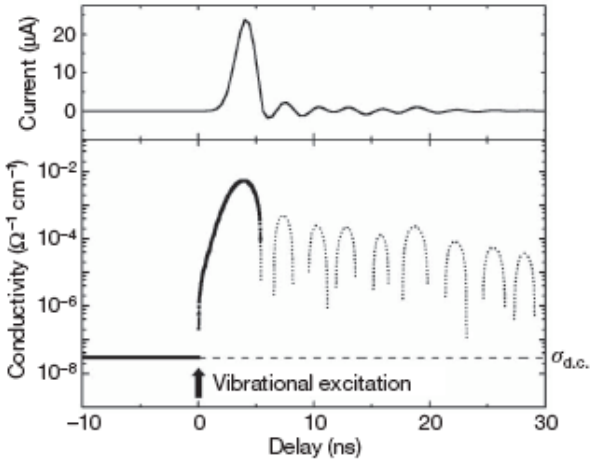


Several exotic materials show a strong interaction between low and high energy degrees of freedom

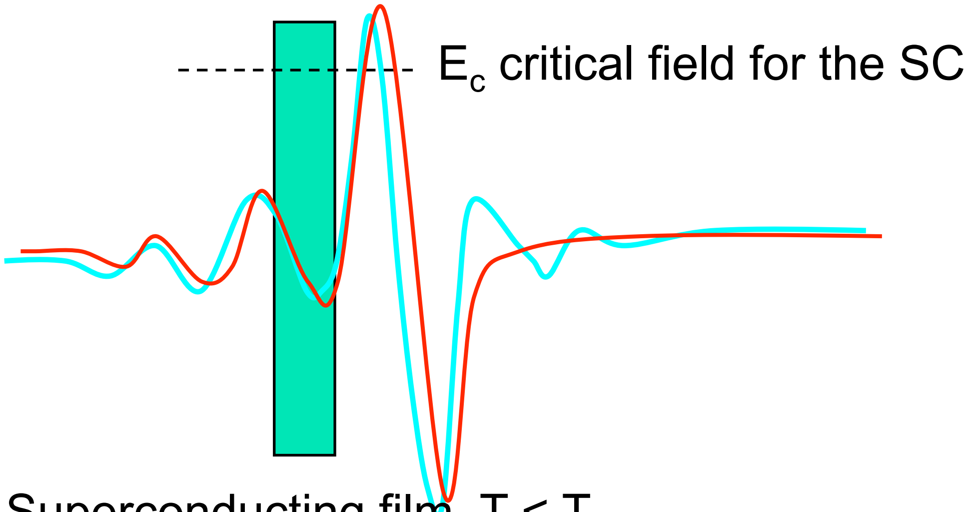
→Through the pump of low energy states one can coherently modify the high energy states



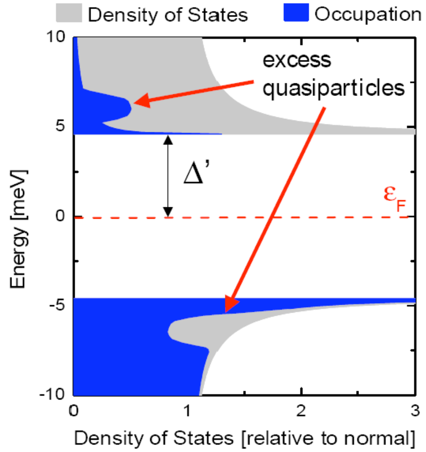


New Experiment in Superconductivity

Breaking the superconducting state through an electric field exceeding the critical electric field Ec



Superconducting film, T < T_c

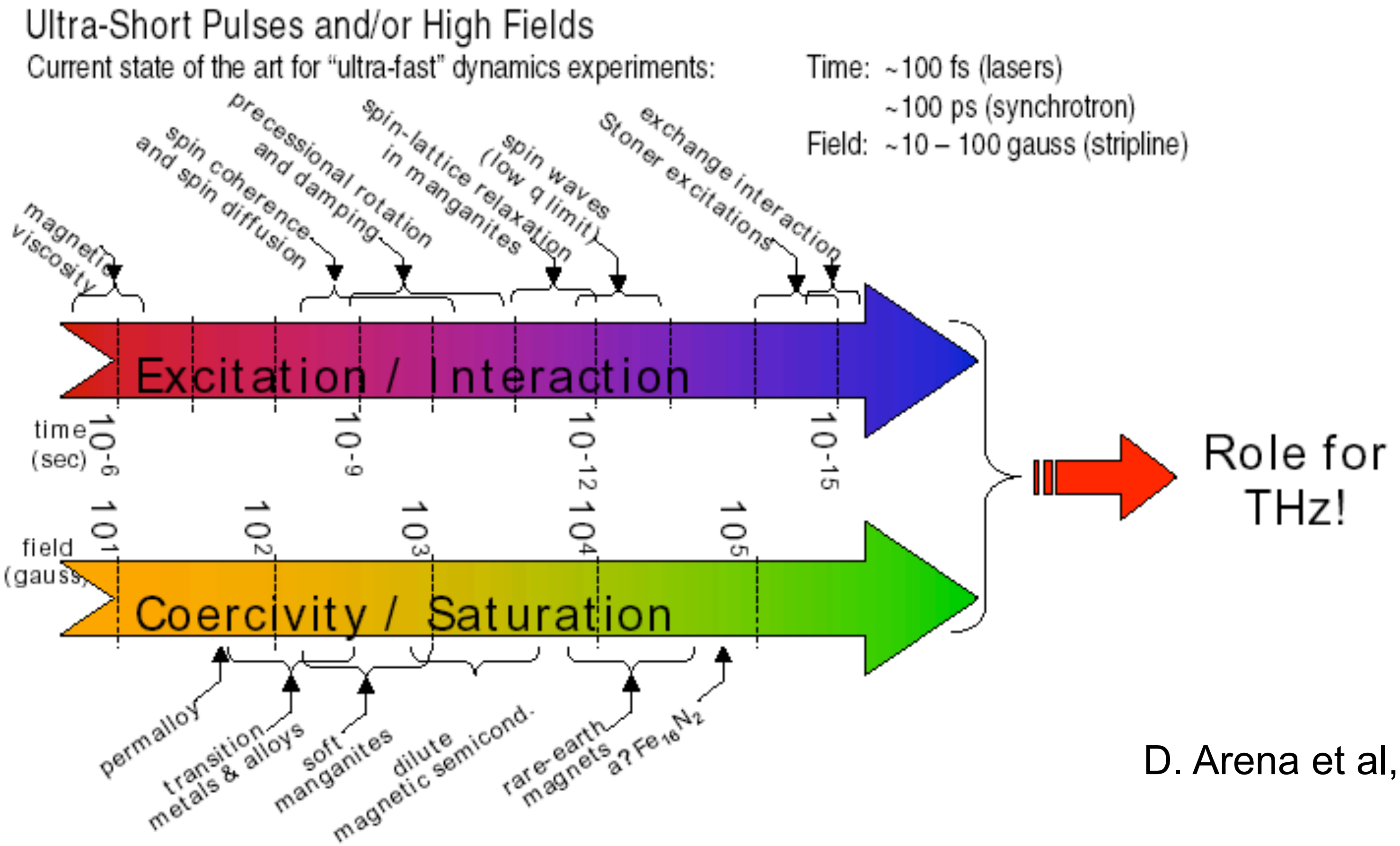


Ultrashort, intense (E>1 MV/cm)THz pulses needed!

Magnetic Dynamics with THz pulses

A sub-ps THz pulse associate to an E field of 1 MV/cm a B field of 0.5 T

THz induced magnetic transitions



Ultra-Short Pulses and/or High Fields

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

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

D. Arena et al, 2006

Conclusion

- Relevant scientific cases for Linear and Pump-ProbeTHz science;
- Relevant technological applications for THz radiation;
- Lack of broad-band ultra-fast sources in the THz range;
- Most of those experiments could be performed through THz radiation produced in LINAC sources;

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