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Infrared spectroscopies with synchrotron radiation and free electron lasers

Part II

THz studies with Storage Rings and Free Electron Laser radiation

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Outline

The THz spectral range

THz spectroscopy with Synchrotron Radiation

Accelerator-based Coherent sources of THz light

TeraFERMI – the THz beamline at FERMI

THz studies with FELs

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The Terahertz Gap

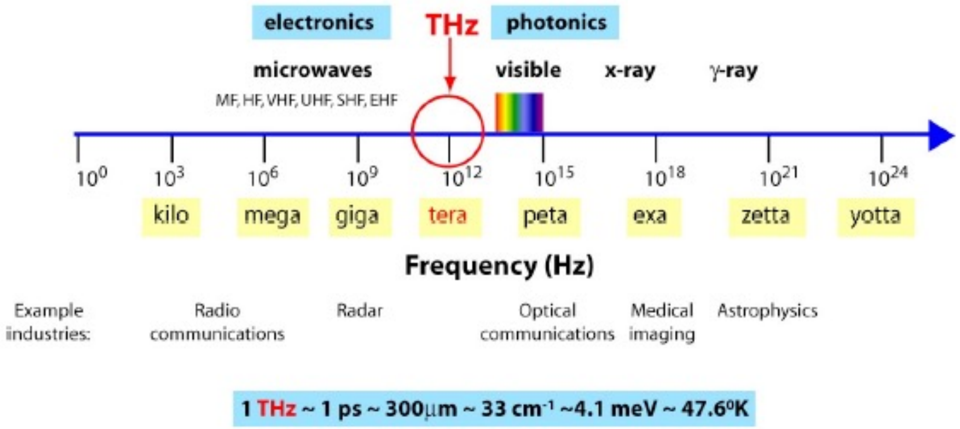
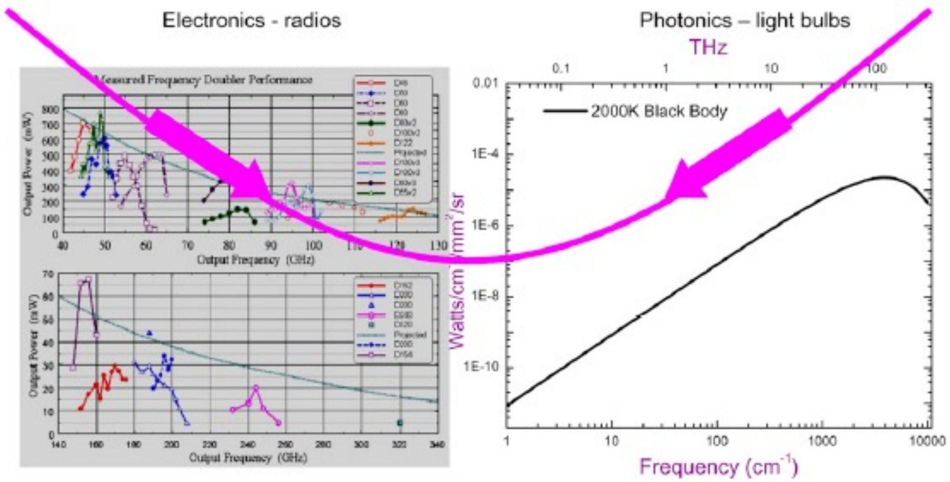


Figure 1. Schematic of the electromagnetic spectrum showing that THz light lies between electronics and photonics.



G. Williams, Rep Prog Phys (2006)

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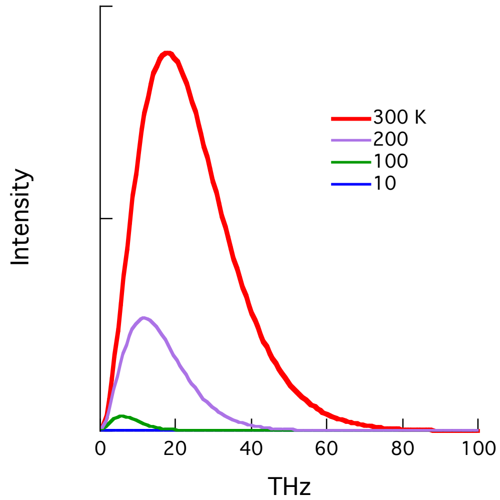
Properties of THz light

- **Non-ionizing**
safe use on living people/animals, non-destructive for biological samples
- **Highly penetrating**
sees through many materials, as packaging, clothing, walls
- **Chemical specificity**
distinguishes between different plastics, drugs, explosives
- **High contrast**
between strong (metals, water) and weak (plastics, tissues) absorbers
- **High-speed communications**
1000 times faster than GHz

…the main drawback is spatial resolution ~ mm

THz light and blackbody radiation



Planck's law

$$I(\nu,T) = \frac{2h\nu^3}{c^2}\frac{1}{e^{\frac{h\nu}{kT}}-1}$$

Wien's law

λ_{max}= b/T   b=2.897 10^{-3} K m

ν_{max}= 0.1035 T   THz K^{-1}

All objects at room temperature are THz radiation emitters

THz modes are normally populated at room T

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Technological applications

Pharmaceutical

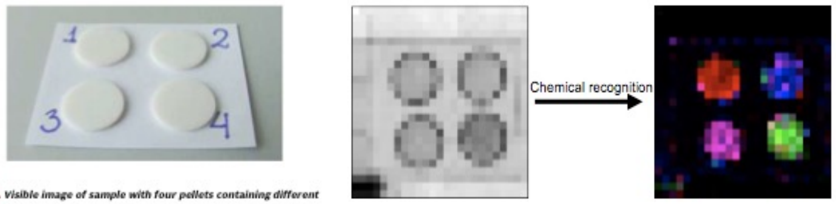
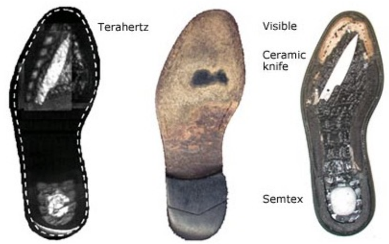


Fig. 8. *Visible image of sample with four pellets containing different chemicals: (1) lactose, (2) aspirin, (3) sucrose, and (4) tartaric acid.*

Security



Quality control

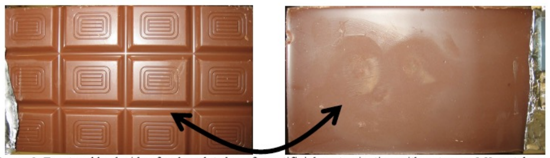
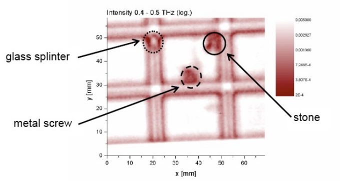


Figure 3. Front and back side of a chocolate bar after artificial contamination with a stone, a M2 metal screw and a glass splinter.



Medical imaging

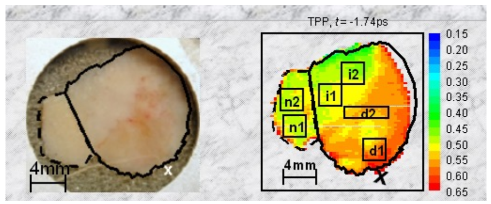


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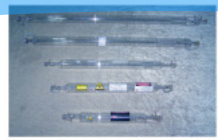
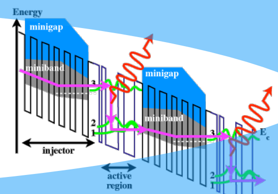
Table-top THz sources

Monochromatic Sources

Backward-Wave-Oscillators



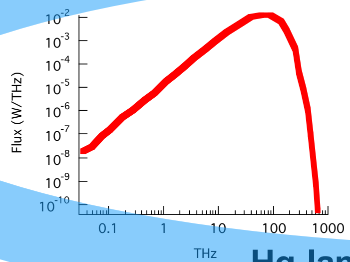
Quantum Cascade Lasers



Gas Lasers (CO_2 and CO_2-pumped)
Si/Ge Lasers

Broadband Sources

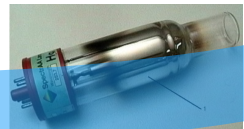
Globar (blackbody source)
silicon carbide rod electrically heated up to 1000 to 1650°C



Total Flux:
10 μW between 0-1 THz
5 mW between 0-10 THz
1,5 W between 0-100 THz

Because of poor collimation typically one has nW power at sample at THz frequencies

Hg-lamp
Performs slightly better than Globar, below 5 THz



Femtosecond THz sources

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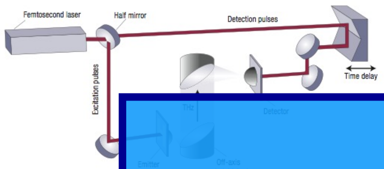
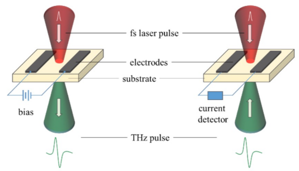
0.1 THz

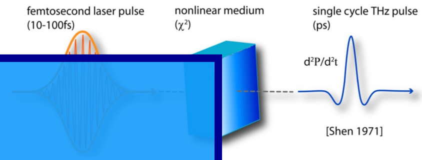
THz
Time Domain Spectroscopy



Photoconductive Antennas
GaAs, TiO_2, …



Optical Rectification
ZnTe, GaP, $LiNbO_3$, etc.



3 THz

$$\eta < 10^{-2}$$

Increasing THz power
---> Increasing Laser power
---> Dealing with radiation damage of the emitter
(losses in brightness/time-structure)

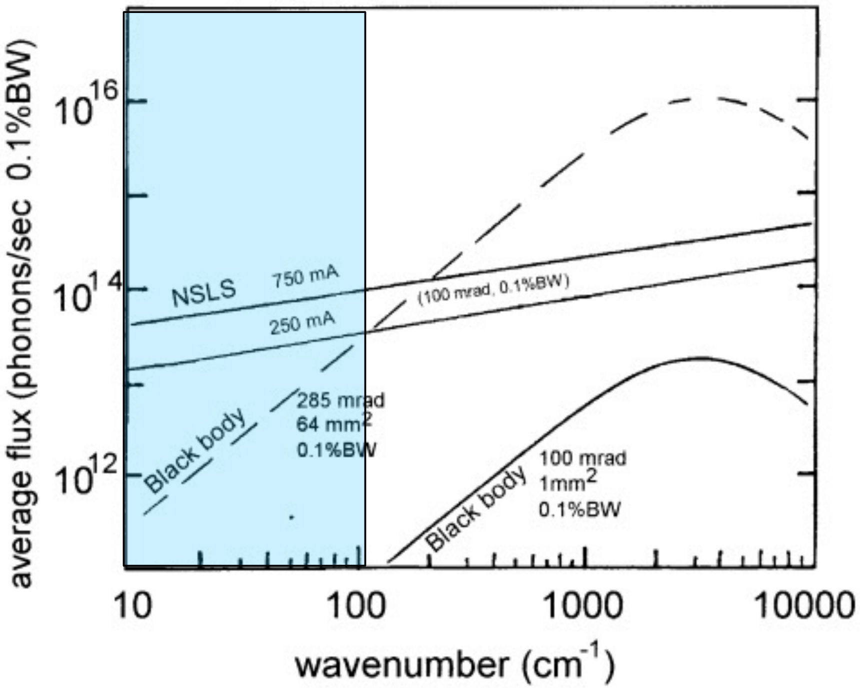
RESTSTRAHLEN BAND GAP
Optical Rectification in Organic Materials

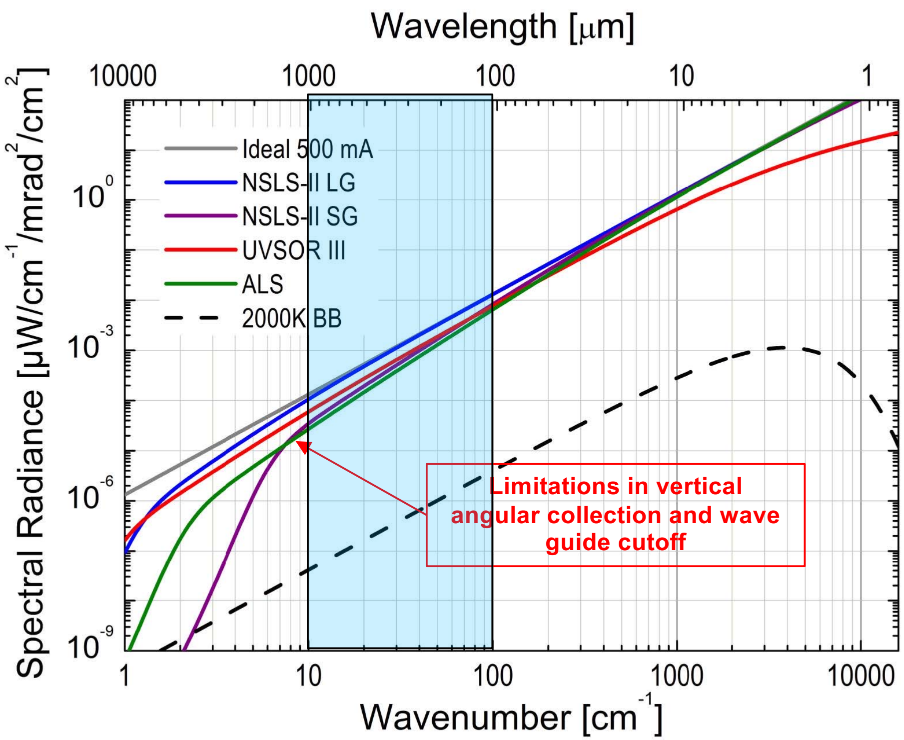
15 THz

Optical Parametric Amplifiers
Tunable, Narrow-Band

IRSR – The spectral radiance advantage

0.3-3 THz





P. Dumas, M.C. Martin, G.L. Carr **2020**

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TeraFERMI – the THz beamline at FERMI

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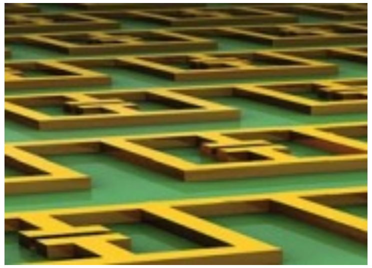
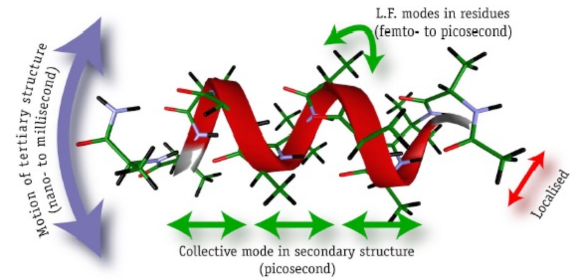
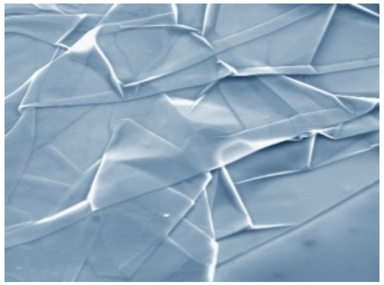
THz Spectroscopy

Superconductivity
Collective excitations
Multiferroics
Heterostructures
Metamaterials
Plasmonics

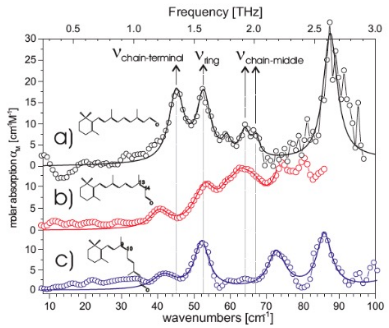




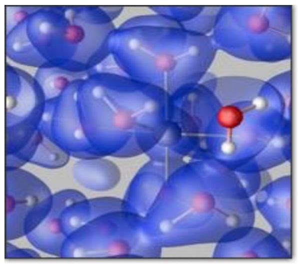
Protein Folding
Amyloid fibrils
Isomers



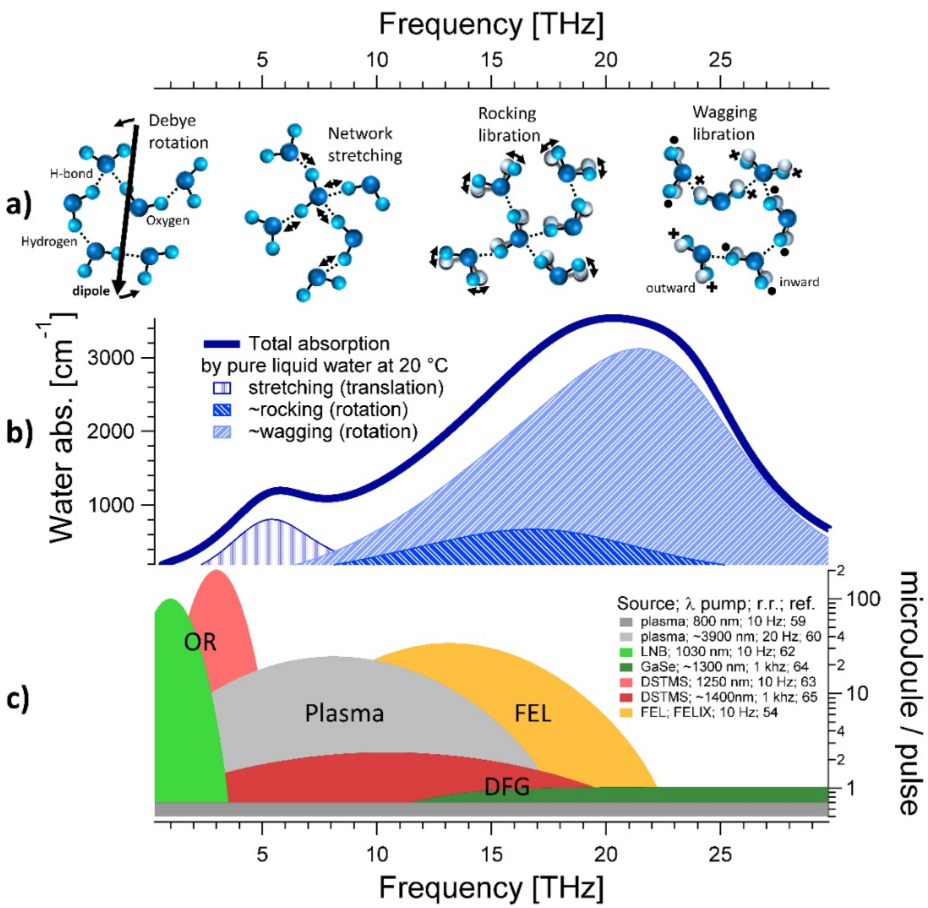
Graphene
2D chalcogenides
Black Phosphorus



Polar liquids
Hydrogen bonds
Van der Waals interactions
Solutions

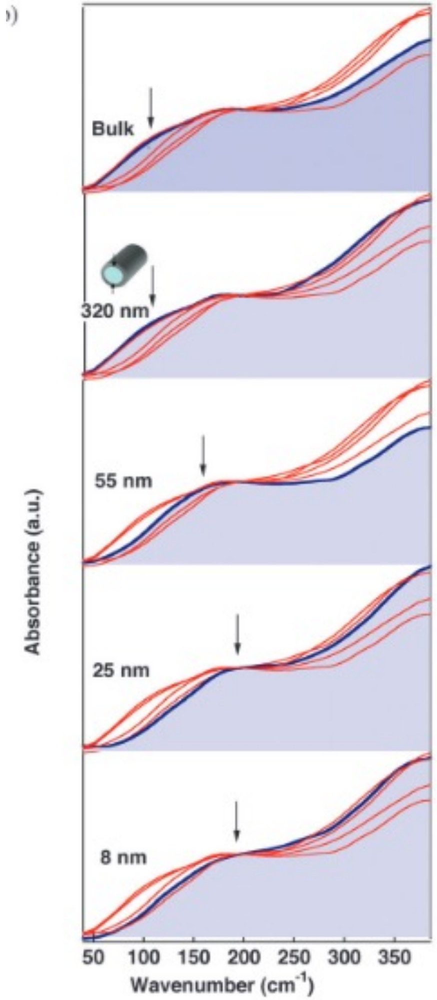


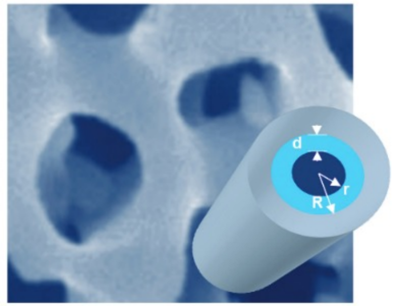
THz modes in water



F. Novelli et al., Materials **2020**

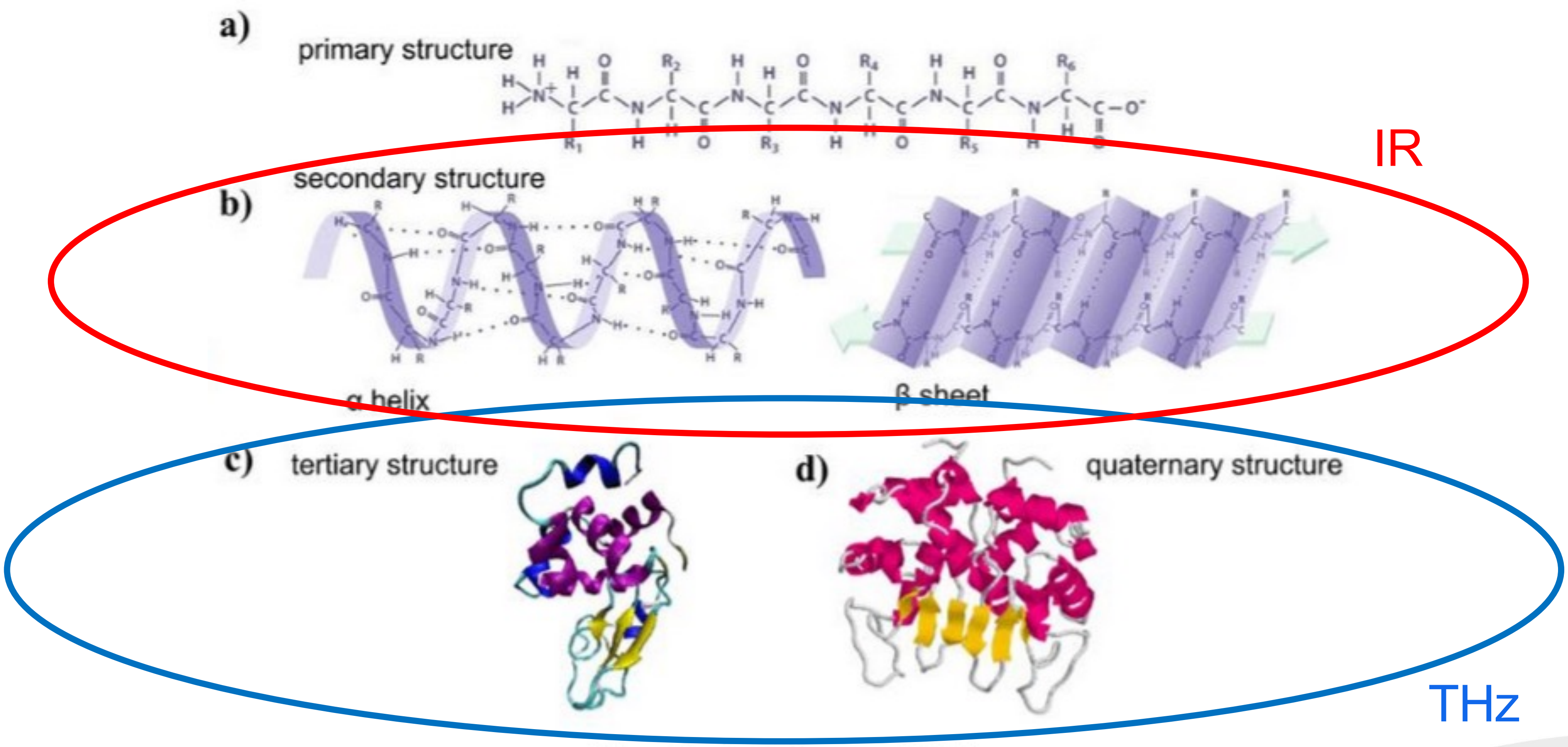
SOLEIL





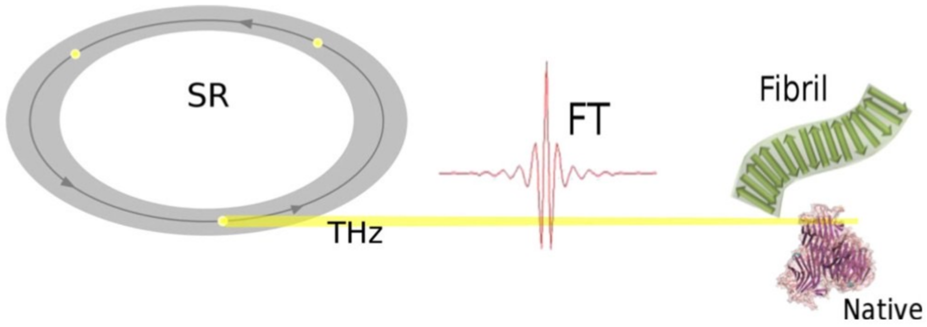
S. Le Caer et al., Phys Chem Chem Phys **2011**

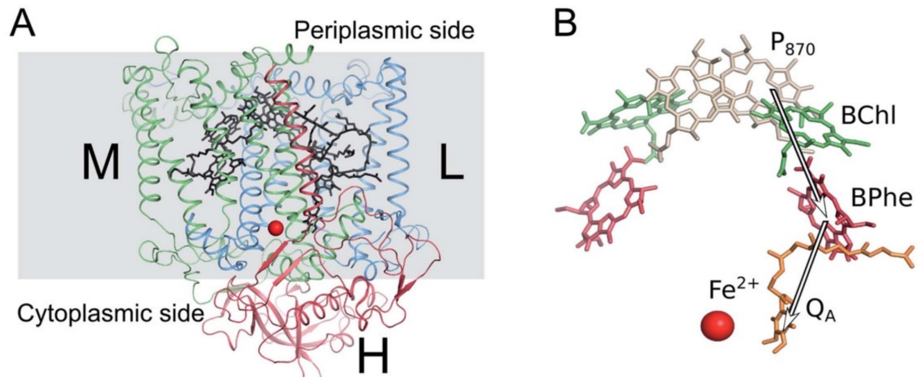
Hyerarchical structure of proteins



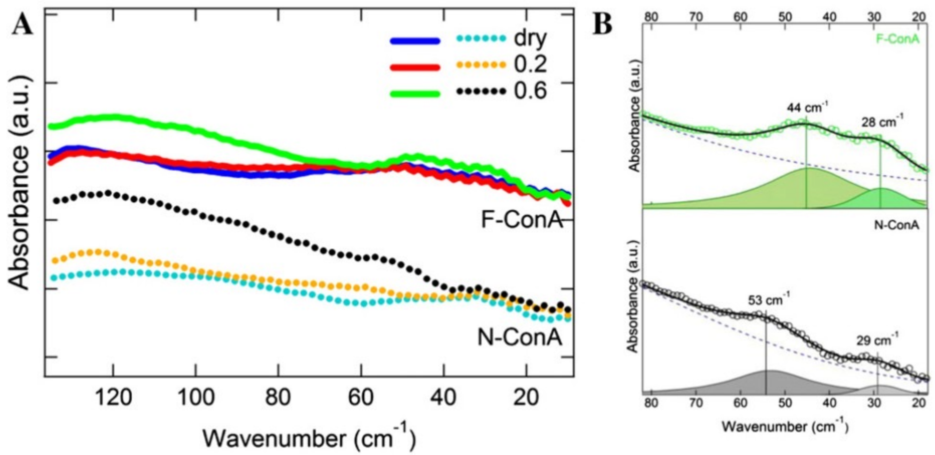
(Martin A Schroer 2011)

THz Spectroscopy of proteins

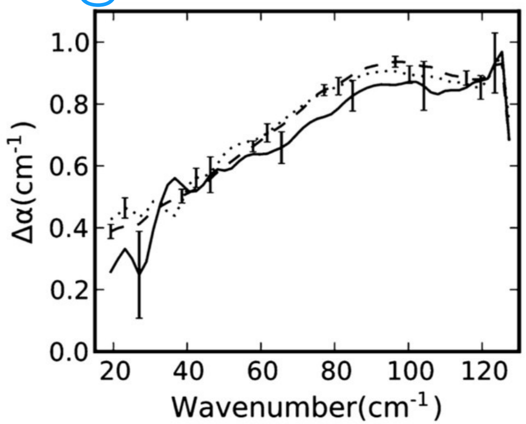




SISSI@Elettra



SISSI@Elettra



F. Piccirilli et al., Biophysical Chemistry **2015**

I. Lundholm et al., RSC Adv. **2014**

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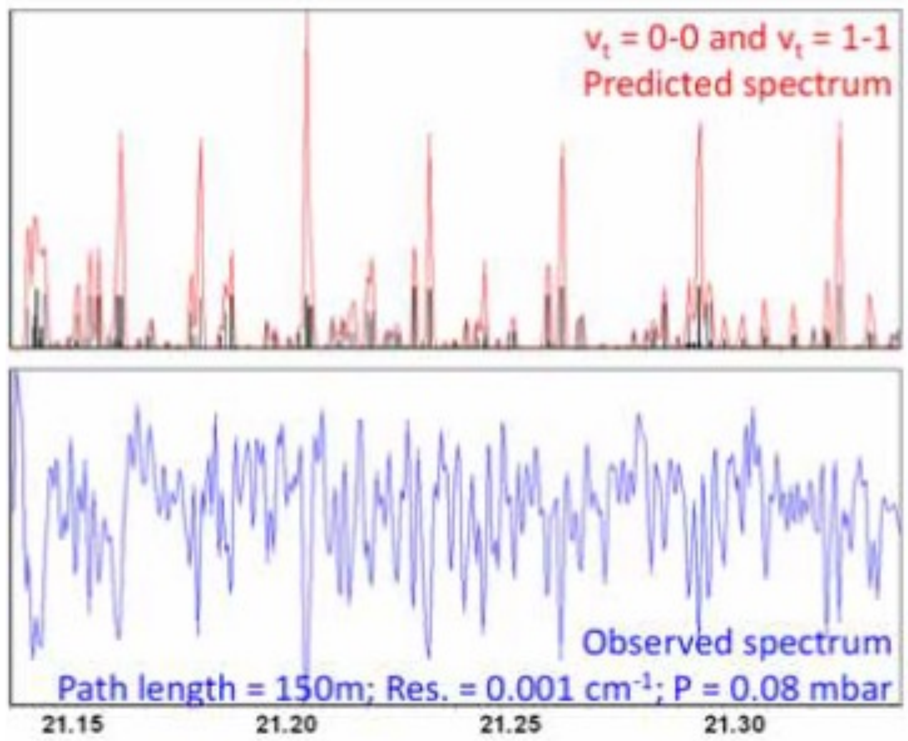
High Resolution Spectroscopy

Detecting molecular rotation and rovibration transitions in gas

SOLEIL



J.-B. Brubach et al., **2010**

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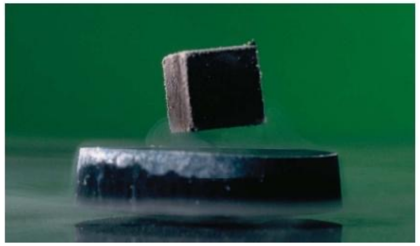
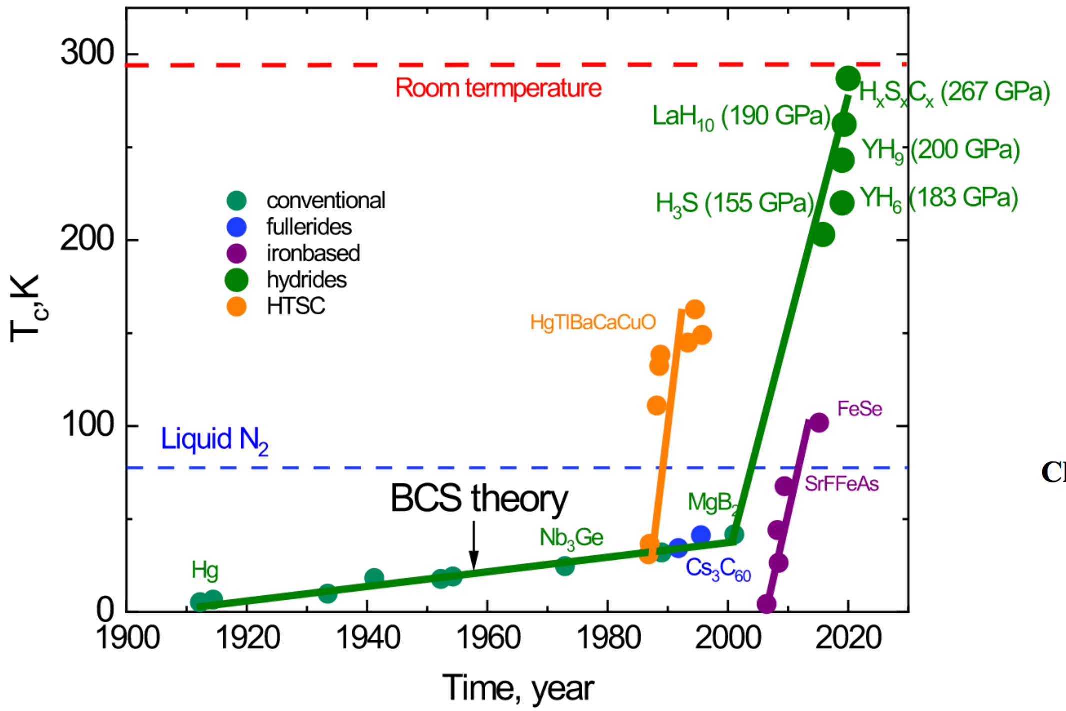
Superconductivity

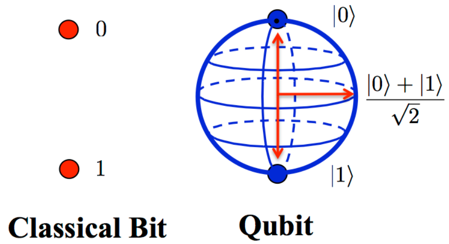


Boeri et al., 2022



Energy

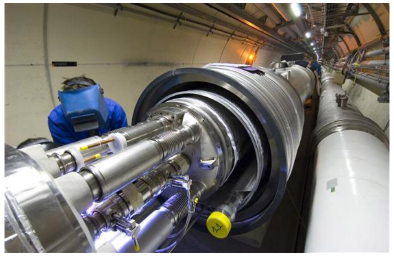
Quantum computing



Magnetic levitation trains (Maglev)

NMR

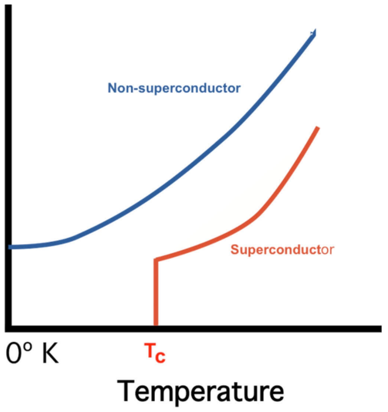
Accelerators

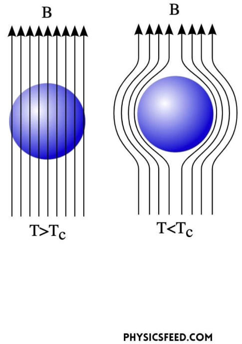


BCS Superconductivity

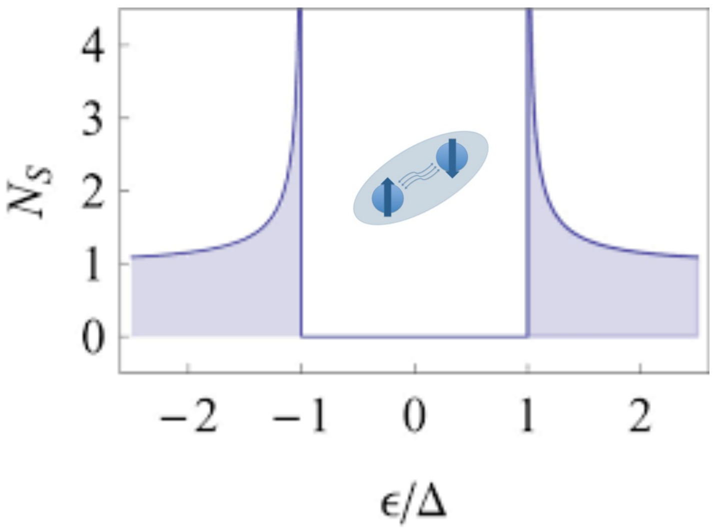
$$\Delta \approx \hbar\omega_D e^{-2/N(0)V}$$

$$2\Delta / k_B T_C = 3.52 \;\Rightarrow\; T_c \approx 10\text{ K} \rightarrow 2\Delta \approx 1\text{ THz}$$

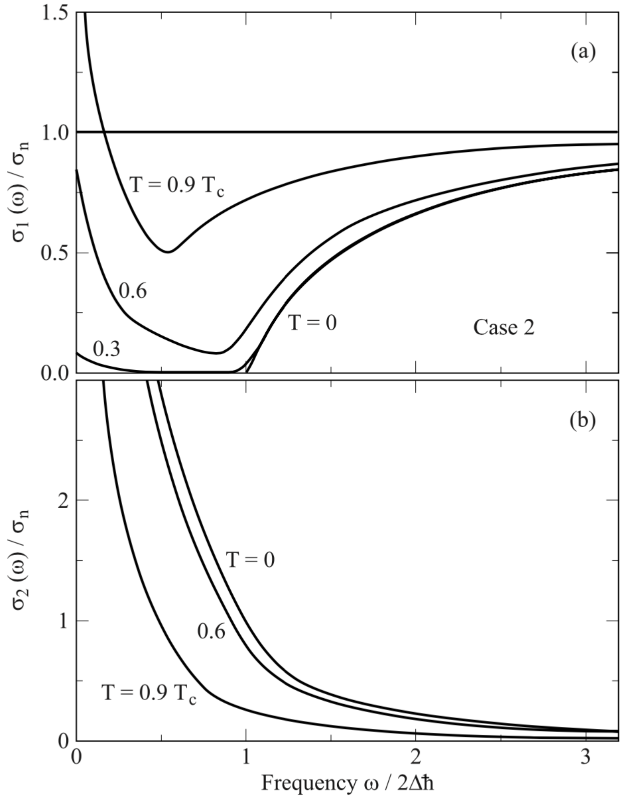


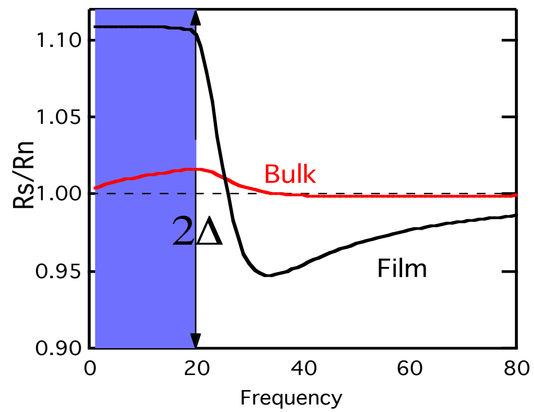


Mattis-Bardeen equations



$$g(E) = \frac{(E^2 + \Delta^2 + \hbar\omega E)}{(E^2 - \Delta^2)^{1/2}((E + \hbar\omega)^2 - \Delta^2)^{1/2}}$$





BCS Superconductors

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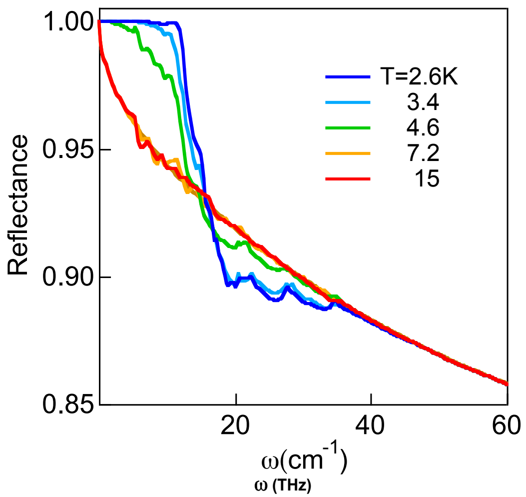
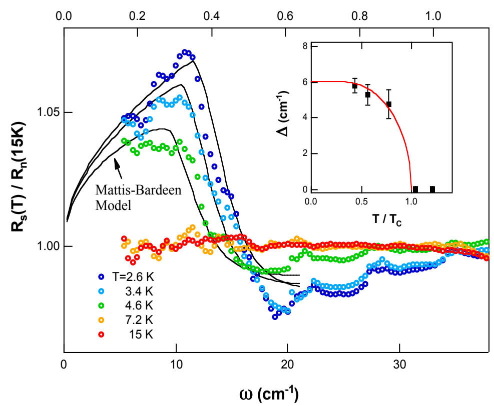
BESSY – low α

B-doped diamond

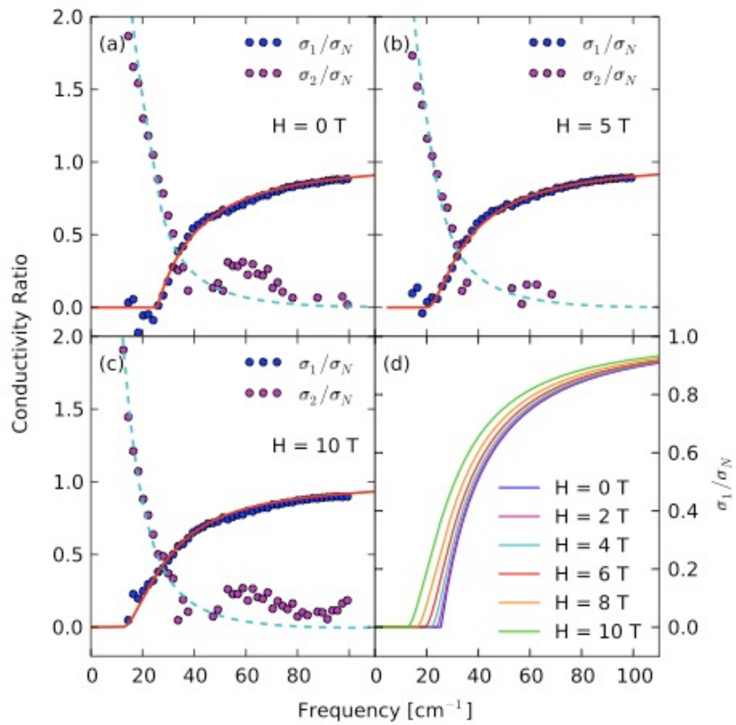




Ortolani et al., PRL (2006)

Brookhaven

$Nb_{0.5}Ti_{0.5}N$



Xi et al., PRL (2011)

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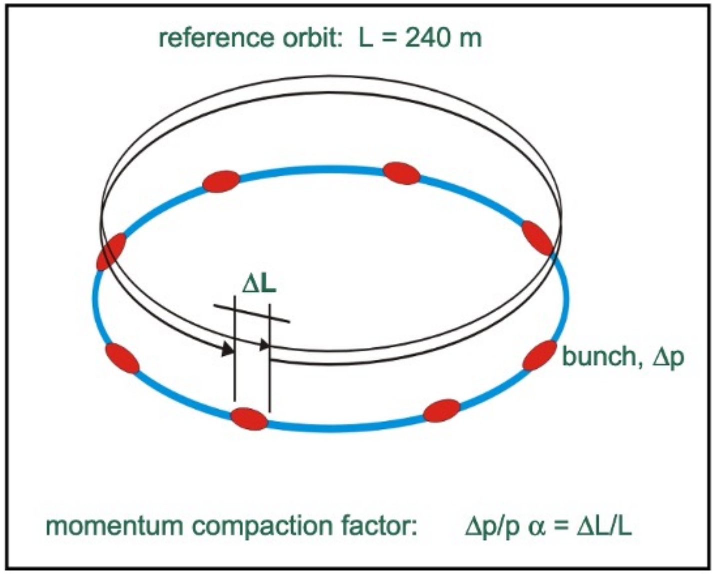
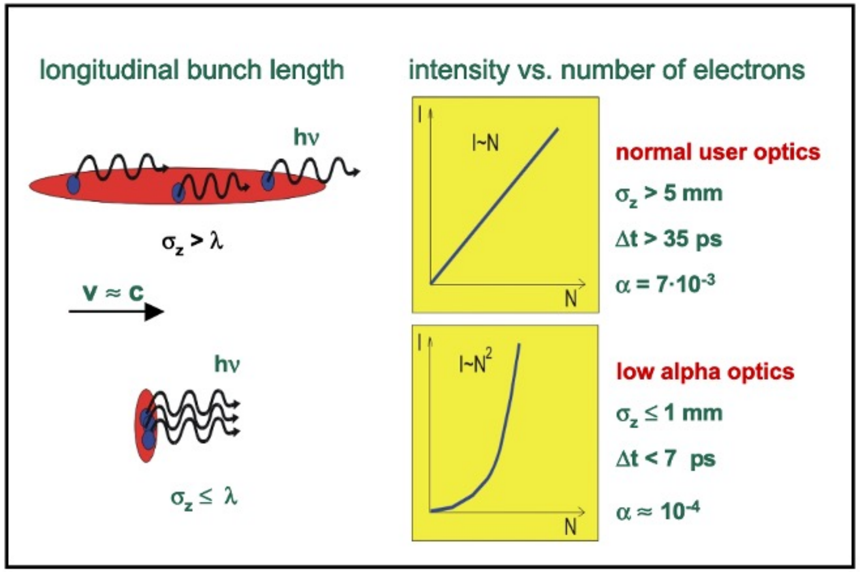
Coherent Synchrotron Radiation



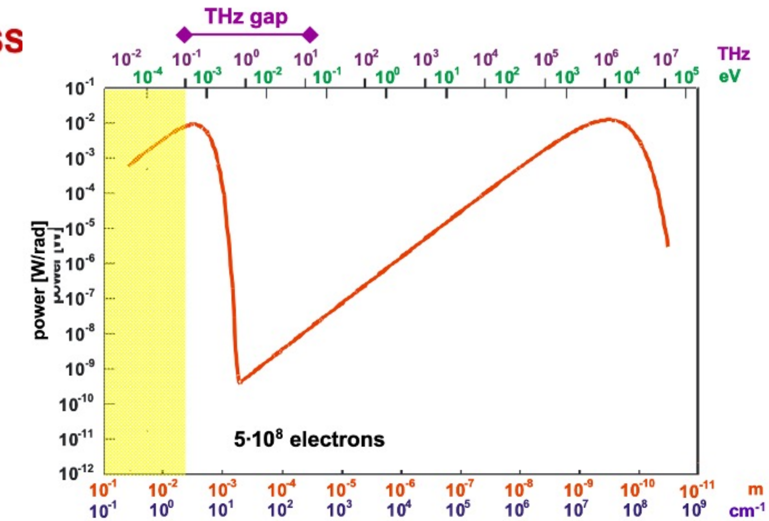


Dedicated Machine Mode: **"Low α" Optics at BESS**

- Bunch shortening down to and below the mm-range
- Emission in the FIR range is drastically enhanced:

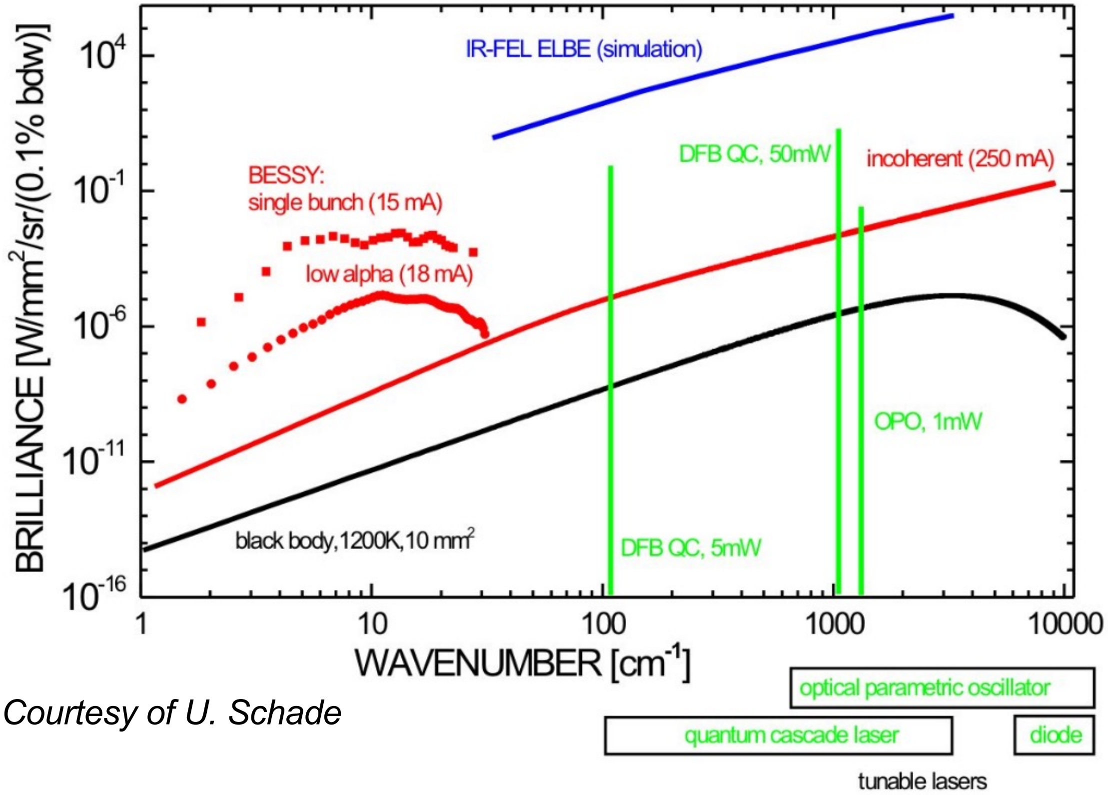
$$I = I_{incoh} + I_{coh} = Ni(1 + Nf_{\nu})$$

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



Courtesy of U. Schade

Coherent Synchrotron Radiation



Courtesy of U. Schade

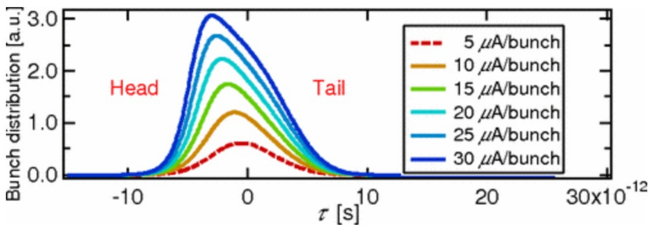
"bursting" mode

- high power CSR
- very noisy
- energy range: 2 - 50 cm^{-1}
- gain of ~10^8

"steady state" mode

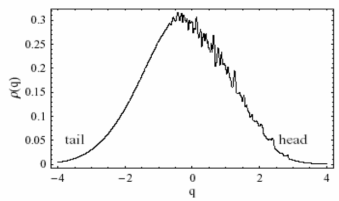
- low noise CSR
- energy range: 2 - 30 cm^{-1}
- gain of ~10^4

Steady State



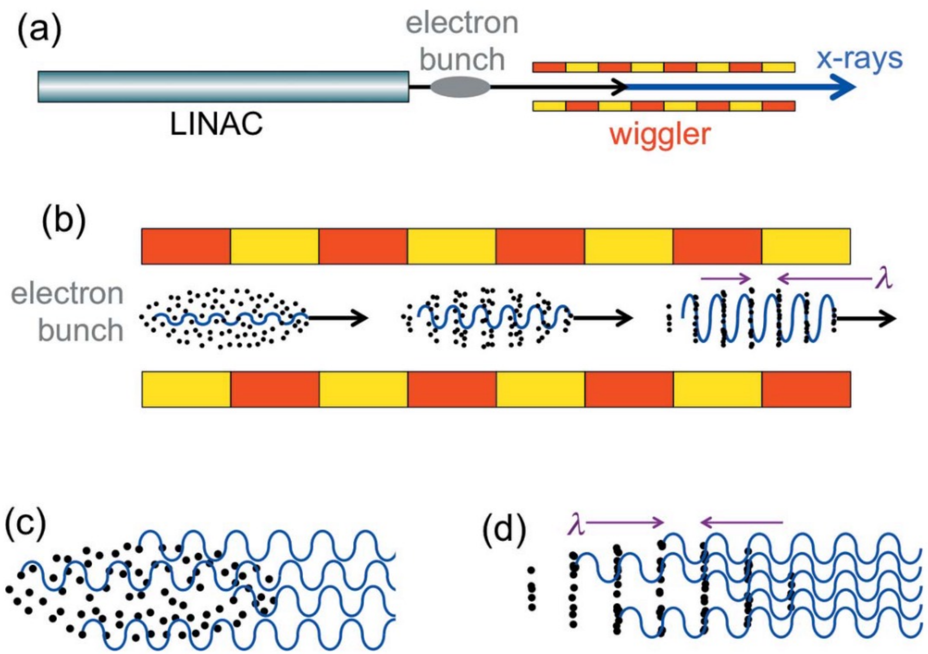
Sannibale et al., PRL 2004

Bursting



Simulated instability showing the microbunching.
Venturini, WarnocK SLAC

Microbunching and free-electron-lasers



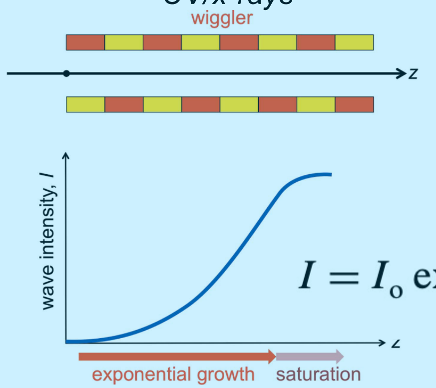
Ponderomotive force

$$f_p = eB_w v_T$$

Hwu and Margaritondo, JSR **2021**

High gain

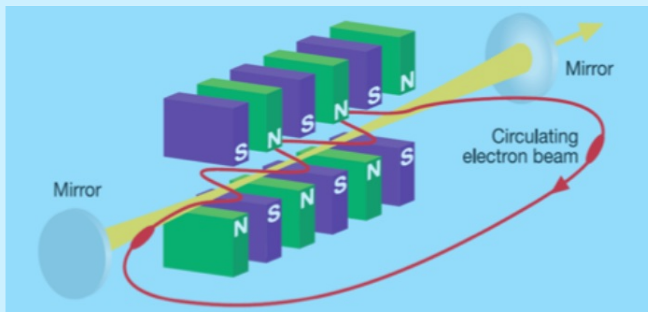
UV/x-rays



$$I = I_o \exp\left(\frac{z}{L_G}\right)$$

Low gain

IR/THz



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IR/THz low gain free-electron-lasers

Low-gain FELs with optical cavity

- UCSB (USA)
- Budker Institute (Russia)
- CLIO (France)
- FELIX / FLARE (Netherlands)
- FELBE (Germany)

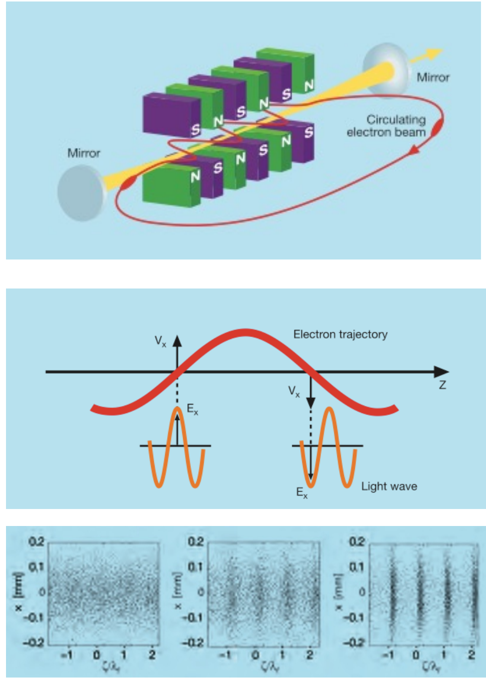
…

$$\lambda = \frac{\lambda_u}{2\gamma^2}\left(1 + \frac{K^2}{2}\right)$$

λ=100 μm
K~1

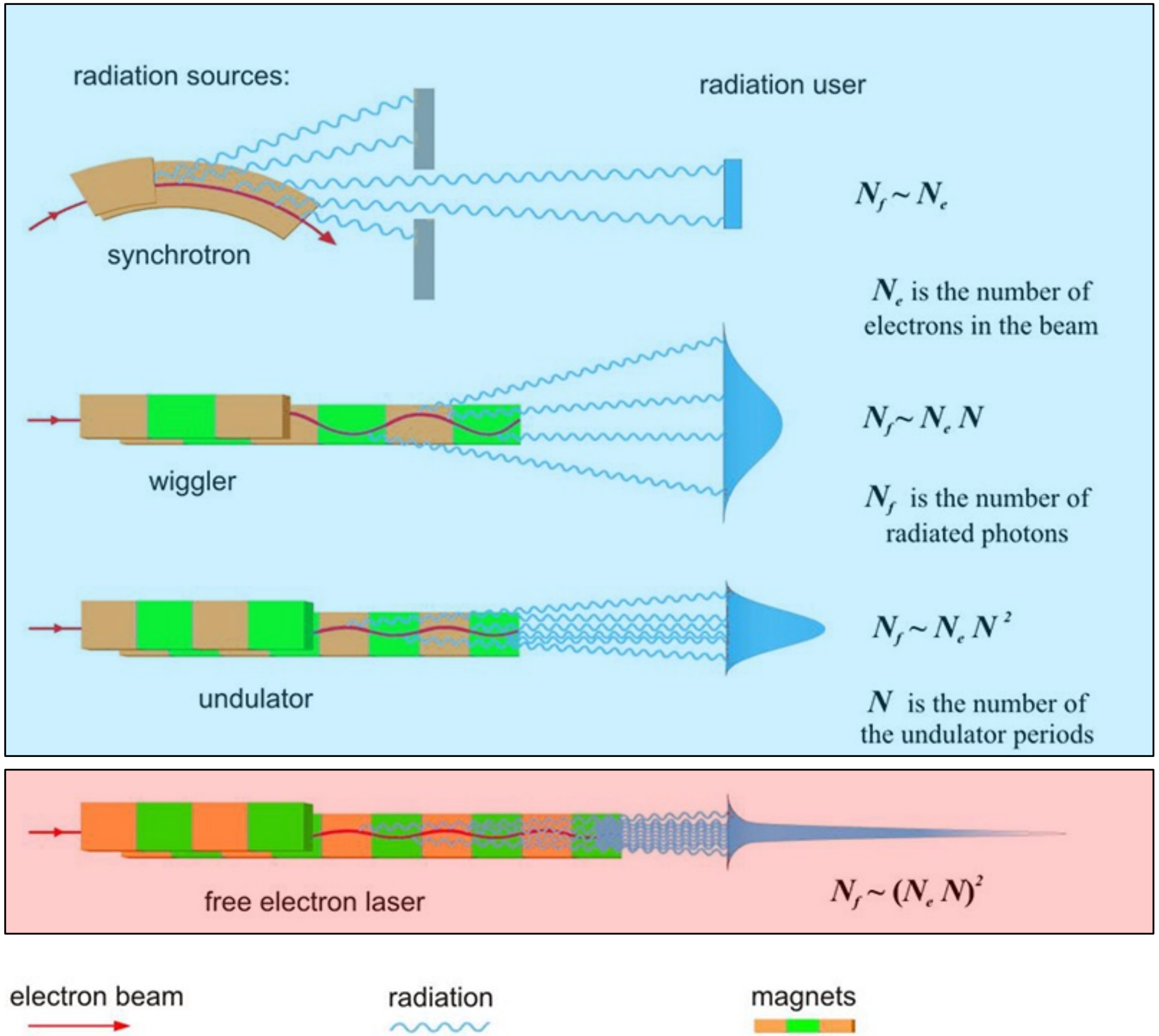
E ~10 MeV	**E ~ 1 GeV**
γ ~ 20	**γ ~ 2000**
λ_U ~ 50 mm	**λ_U ~ 500 m !!!**

Upon one passage in the undulator, radiation grows by a few percent
$\rightarrow$ Several passages are needed before reaching saturation



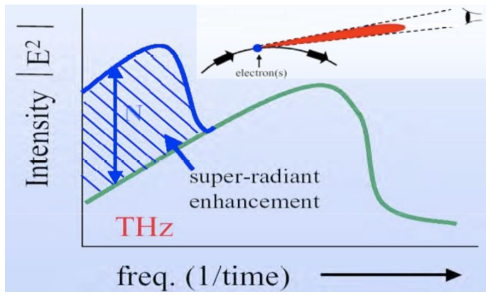
From DESY-FLASH brochure

... a recap



Incoherent

Can be made coherent through ***superradiance***



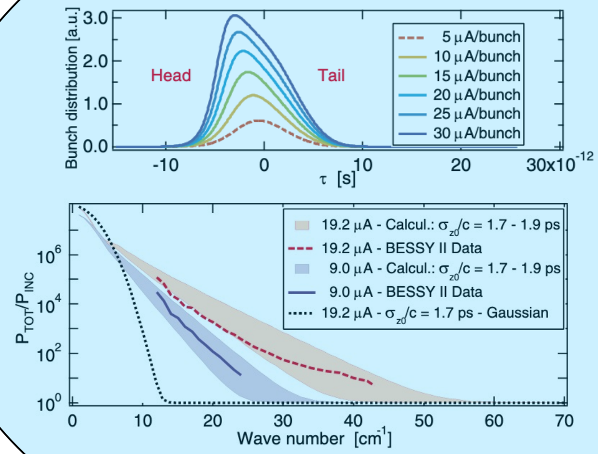
$$f_\nu = \left| \int n(z)\, e^{i\pi \cos(\theta) z}\, dz \right|^2$$

Coherent

Zhukowsky, **2015**

THz superradiant sources in SRs

Low-α mode

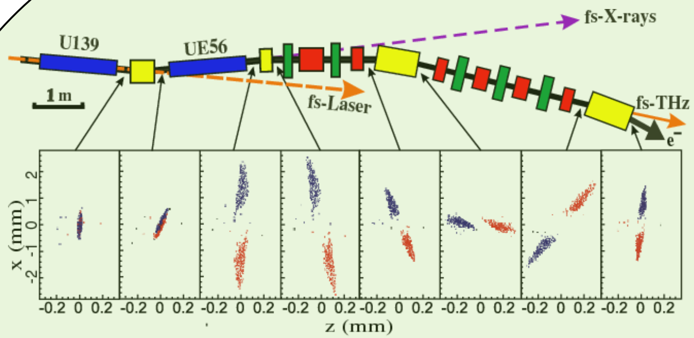


Sannibale, PRL **2004**

Beam shaping through Magnetic lattice

...but CSR wakefields play a role!

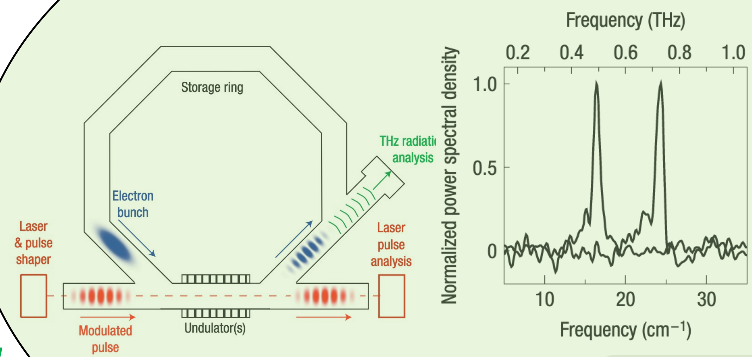
Femtoslicing



Holldack, PRL **2006**

Laser modulated

Narrow-band THz



Bielawski, Nature **2008**

THz superradiant sources in LINACs

$$I = I_{incoh} + I_{coh} = Ni(1 + Nf_\nu)$$

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

$$A_f = \frac{I_{coh}}{I_{incoh}} = Nf_\nu$$

1 C = 6,241 10^{18} e

For Storage Rings:
~1 pC/bunch → N~ 6 10^6 → E ~ 10^{-10} J/pulse

For linear accelerators:
~1 nC/bunch → N~ 6 10^9 → E ~10^{-4} J/pulse

Undulator
TELBE/FLASH

Transition Radiation
TeraFERMI

Bending magnet
J-Lab

Outline

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Elettra
Sincrotrone
Trieste

The FERMI seeded FEL

nature photonics ARTICLES
PUBLISHED ONLINE: 23 SEPTEMBER 2012 | DOI: 10.1038/NPHOTON.2012.233

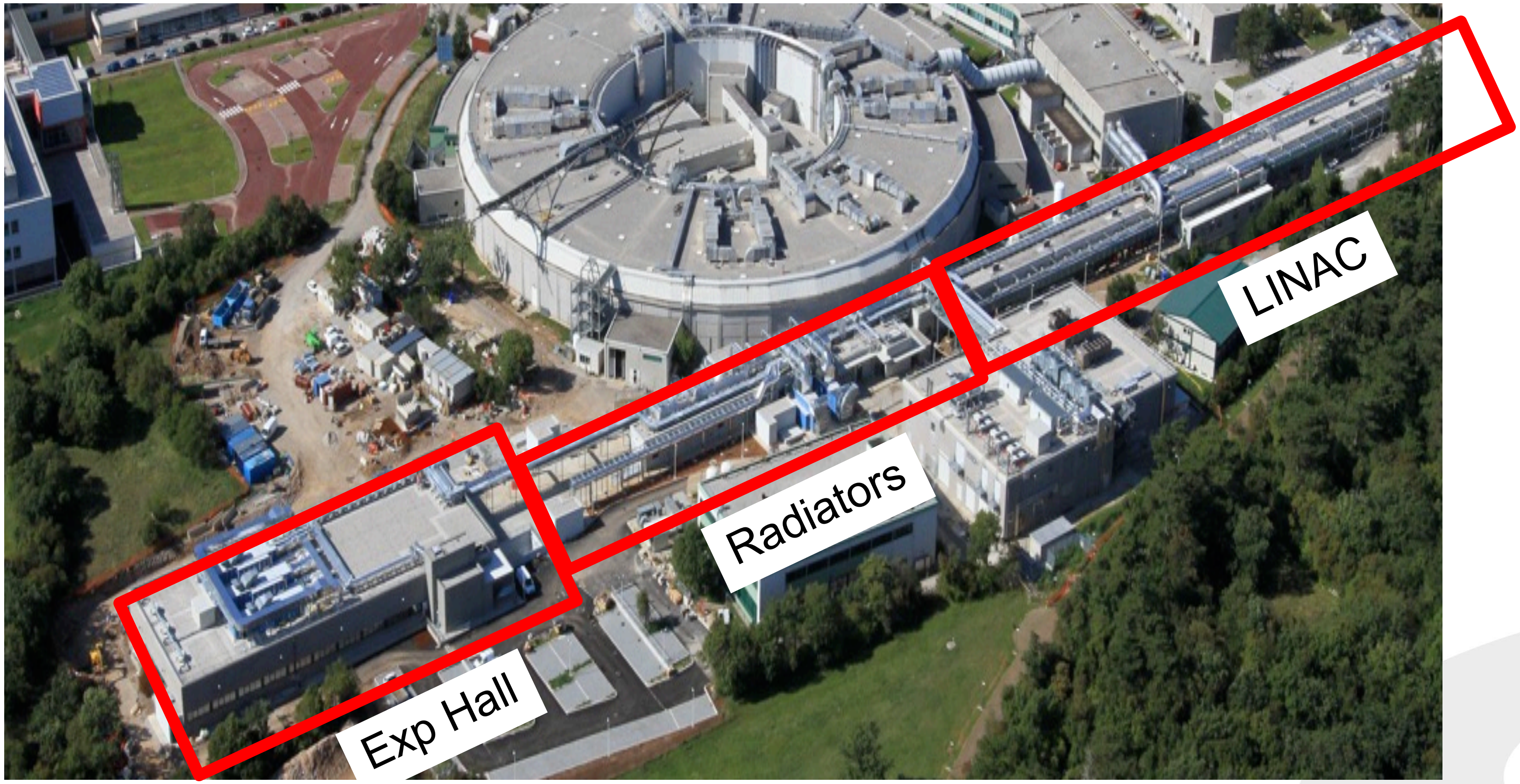
Highly coherent and stable pulses from the FERMI seeded free-electron laser in the extreme ultraviolet → **100-20 nm**

First user exp. Dec 2012

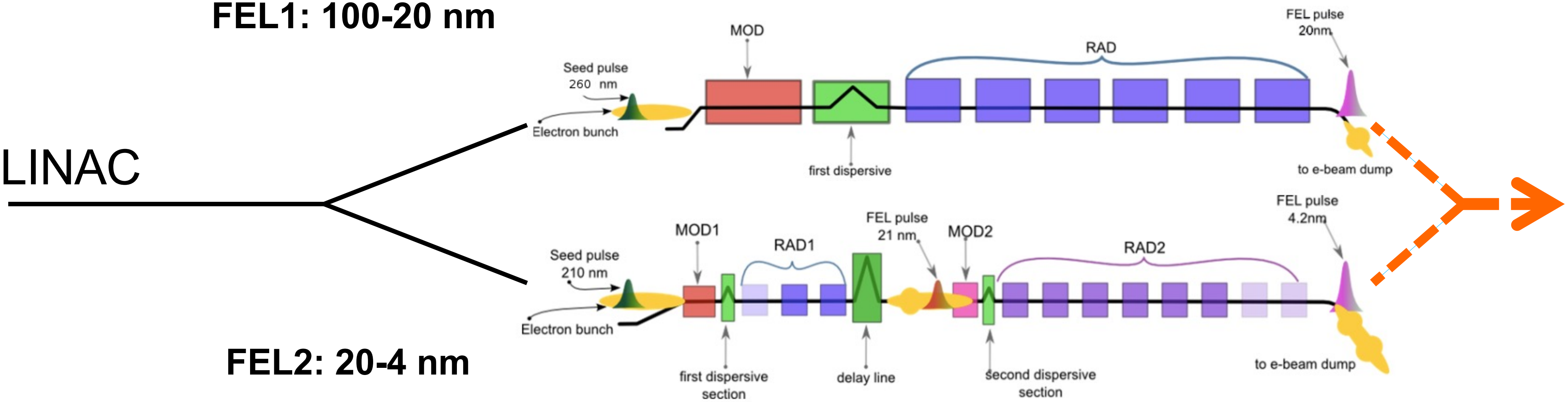
nature photonics ARTICLES
PUBLISHED ONLINE: 20 OCTOBER 2013 | DOI: 10.1038/NPHOTON.2013.277

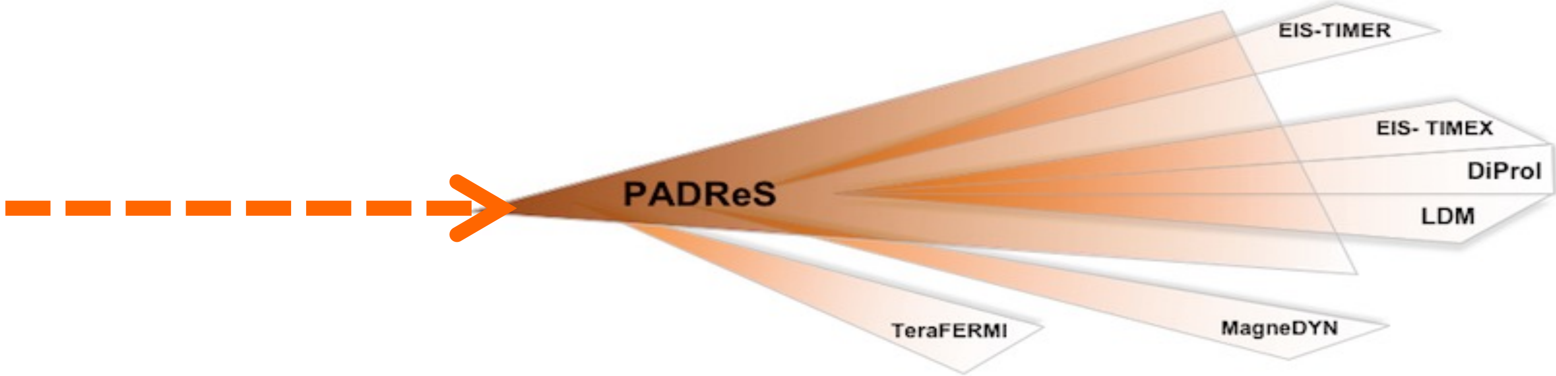
Two-stage seeded soft-X-ray free-electron laser → **20-4 nm**

First user exp. July 2016



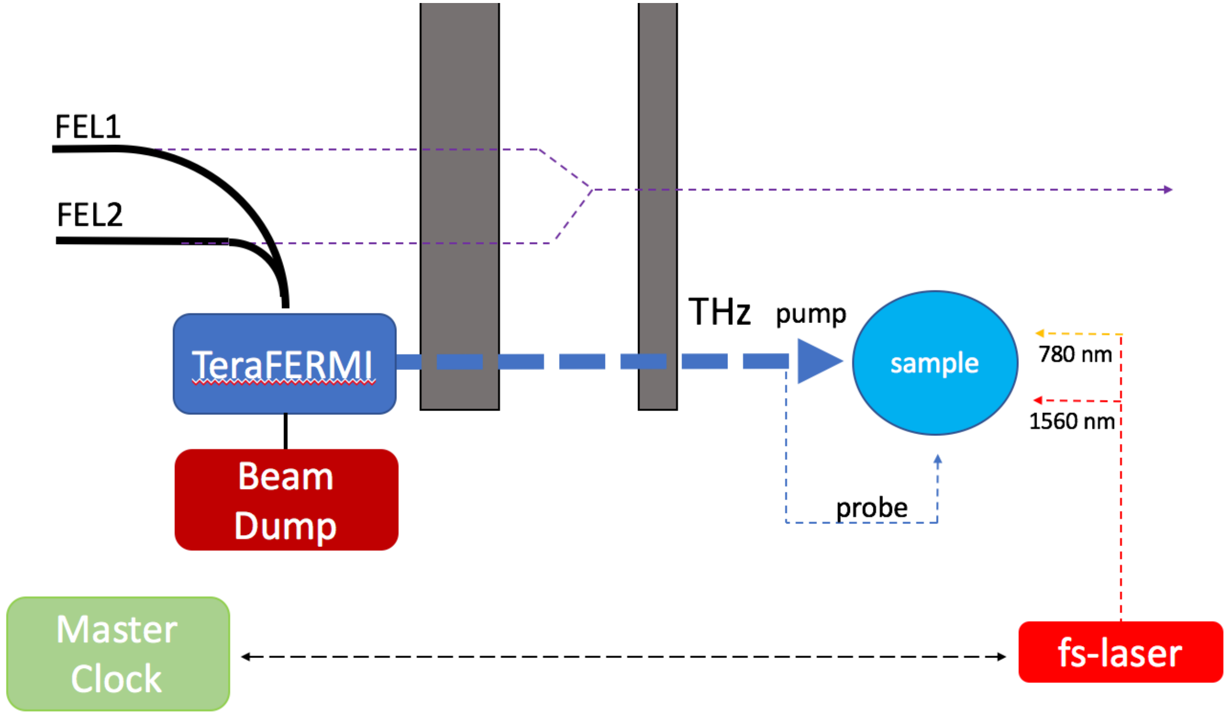
The FERMI seeded FEL

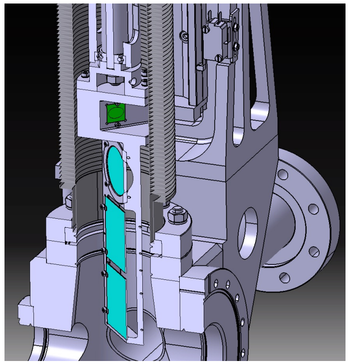




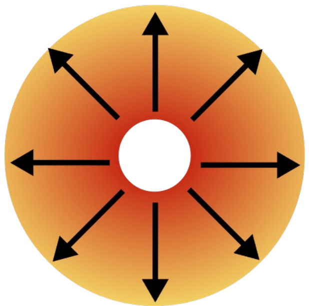
Elettra Sincrotrone Trieste

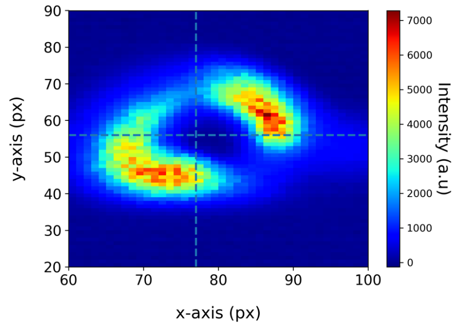
The TeraFERMI beamline





Coherent Transition Radiation

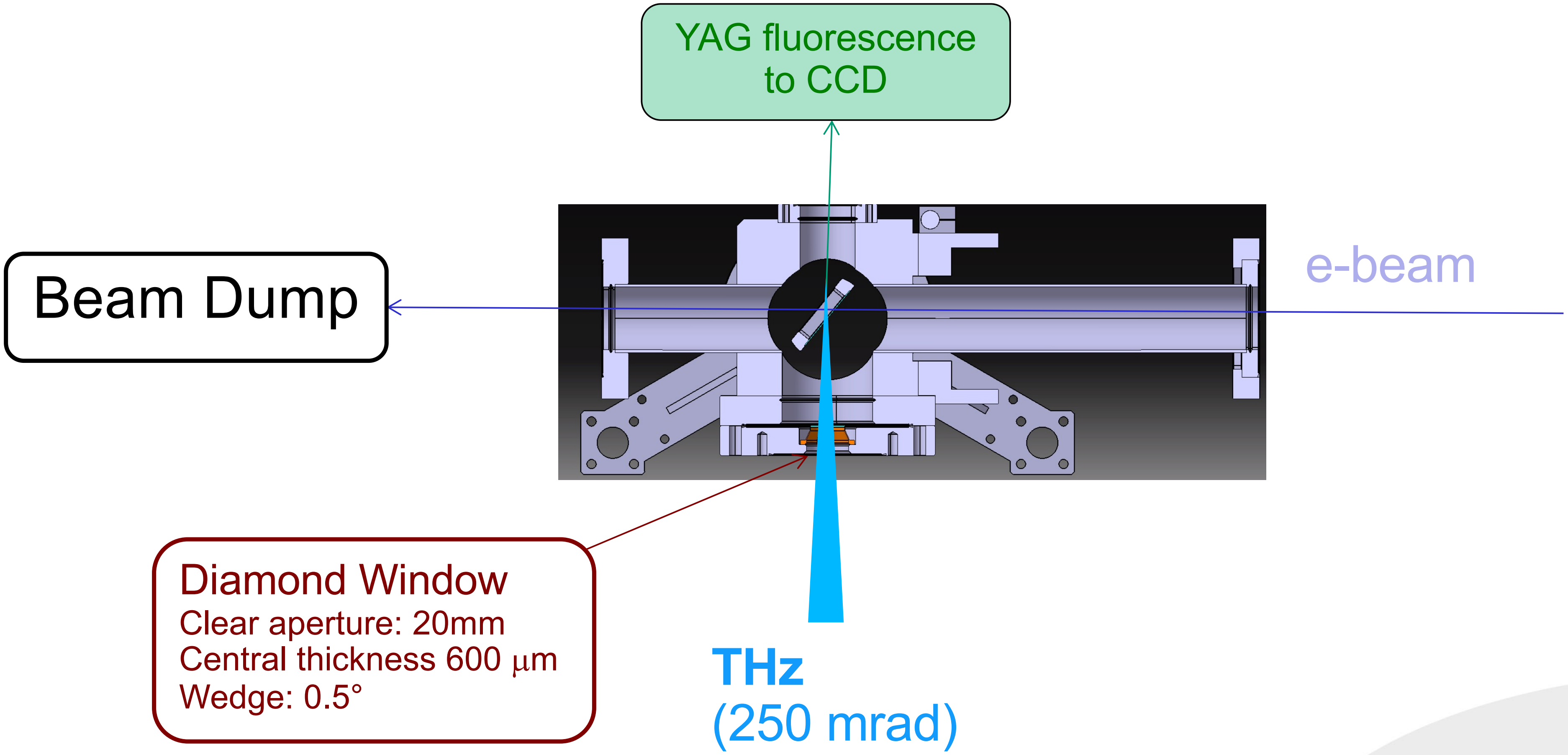




Cylindrical symmetry
Radial polarization

→ *Efficient generation of longitudinally polarized fields*

TeraFERMI Source

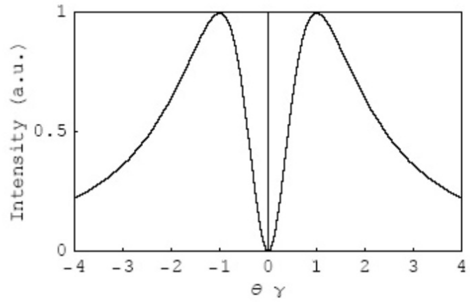


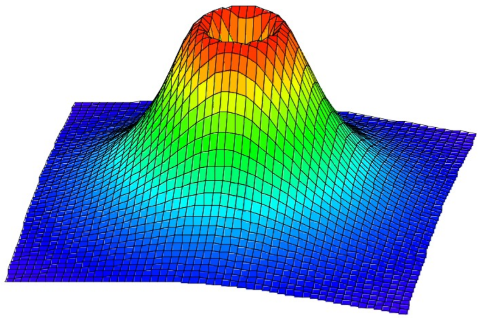
Transition Radiation

Transition Radiation occurs when relativistic electrons cross the boundary between two media of different dielectric constant

The Ginzburg-Frank equation:

$$\frac{d^2U}{d\omega d\Omega} = \frac{e^2}{4\pi^3\varepsilon_0 c}\frac{\beta^2 \sin^2\theta}{(1-\beta^2\cos^2\theta)^2}$$

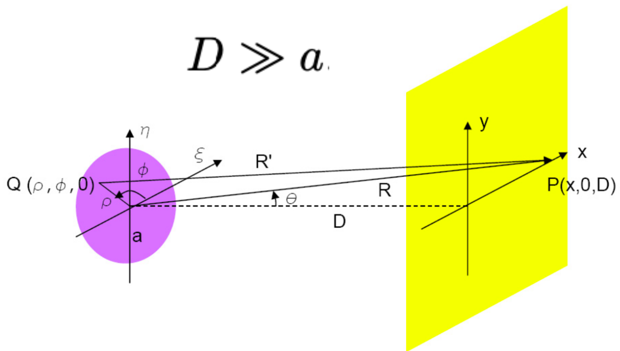




$$\theta_{max} = \arcsin\left(\frac{\sqrt{1-\beta^2}}{\beta}\right) = \arcsin\left(\frac{1}{\beta\gamma}\right) \simeq \frac{1}{\gamma} \quad \text{for} \quad \gamma \gg 1$$

Generalized Ginzburg-Frank equation (TR from finite screen, far-field)

$D \gg a$



Casalbuoni, TESLA Report 2005-15

$$\frac{d^2U}{d\omega d\Omega} = \frac{e^2}{4\pi^3\varepsilon_0\, c}\cdot\frac{\beta^2\sin^2\theta}{(1-\beta^2\cos^2\theta)^2}\left[1-T(\theta,\omega)\right]^2$$

with

$$T(\theta,\omega) = \frac{\omega a}{c\beta\gamma}J_0\left(\frac{\omega a\sin\theta}{c}\right)K_1\left(\frac{\omega a}{c\beta\gamma}\right) + \frac{\omega a}{c\beta^2\gamma^2\sin\theta}J_1\left(\frac{\omega a\sin\theta}{c}\right)K_0\left(\frac{\omega a}{c\beta\gamma}\right)$$

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Trieste

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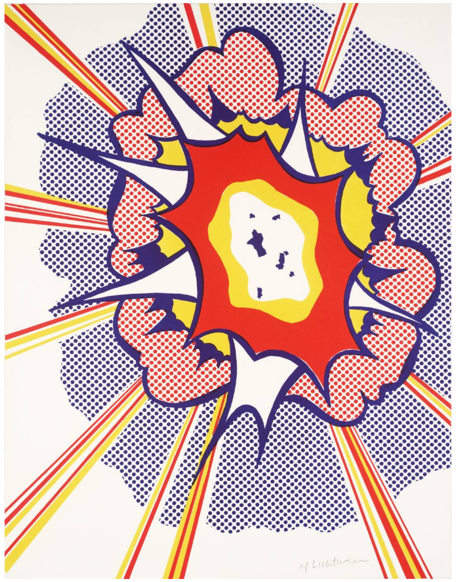
THz pumping

THz radiation is non-ionizing, highly penetrating, and provides high chemical specificity

THz light couples to electronic, vibrational and magnetic excitations

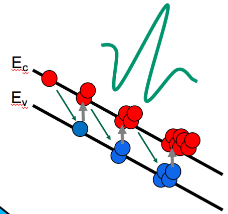


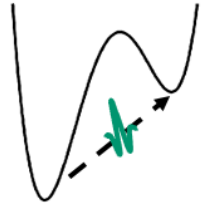
Optical pump

THz pump

Elettra Sincrotrone Trieste

THz control of matter



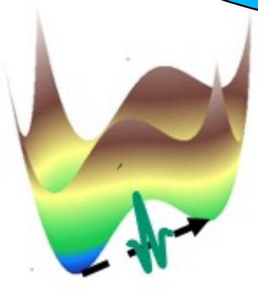


Nucleae/lattice

Nonlinear phononics
Macromolecules
Molecular alignment
Reaction pathways

Electrons

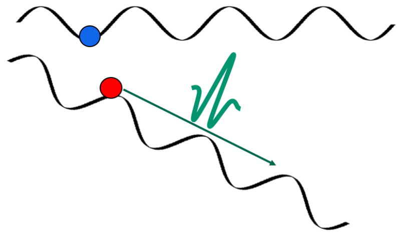
Impact ionization
Intervalley scattering
Zener tunneling
Franz-Keldysh

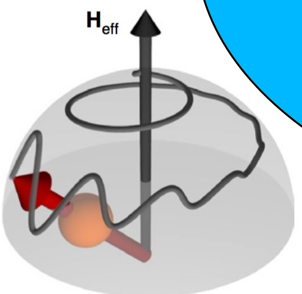


Spins

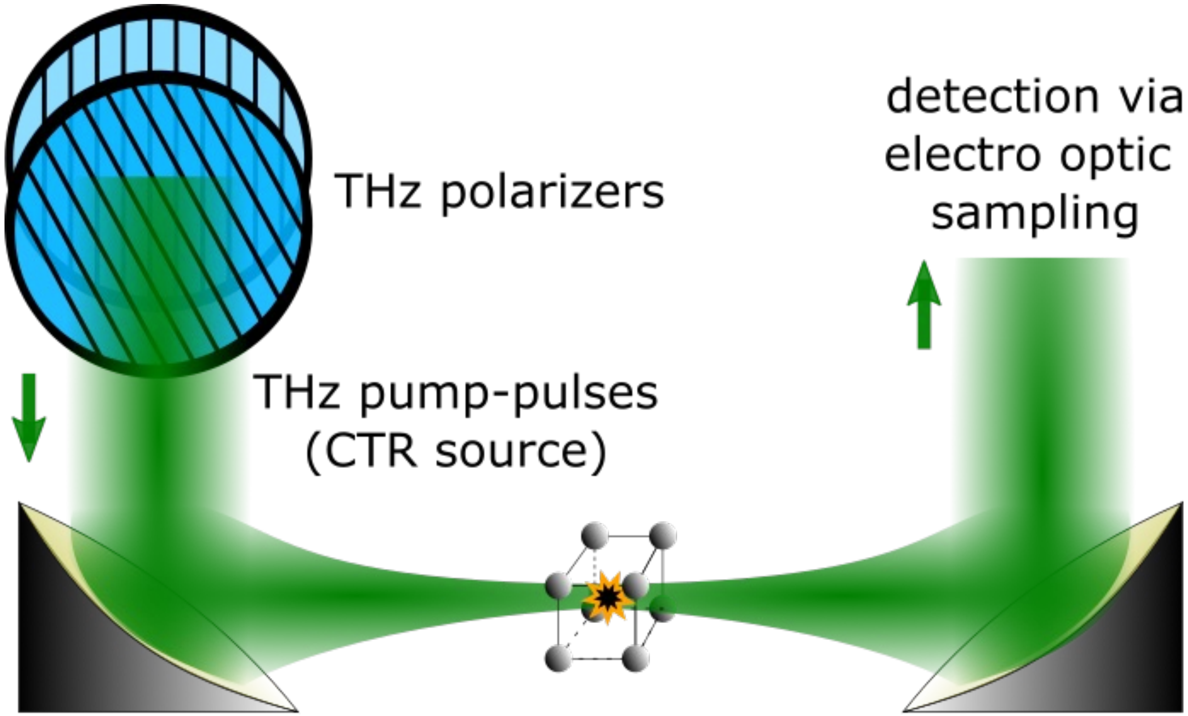
Ultrafast magnetic switching
Precession

1 MV/cm ~ 0.3 Tesla





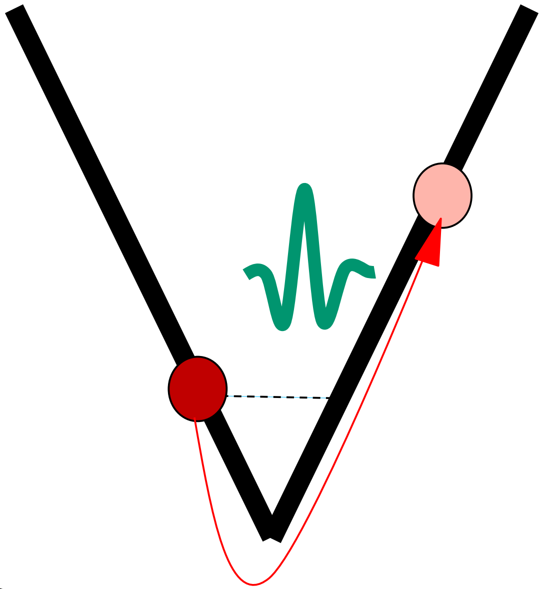
Fluence-dependent spectroscopy



Dirac materials

Graphene

Topological Insulators



TMDs

Black phosphorus

Saturable Absorption

Harmonics Generation

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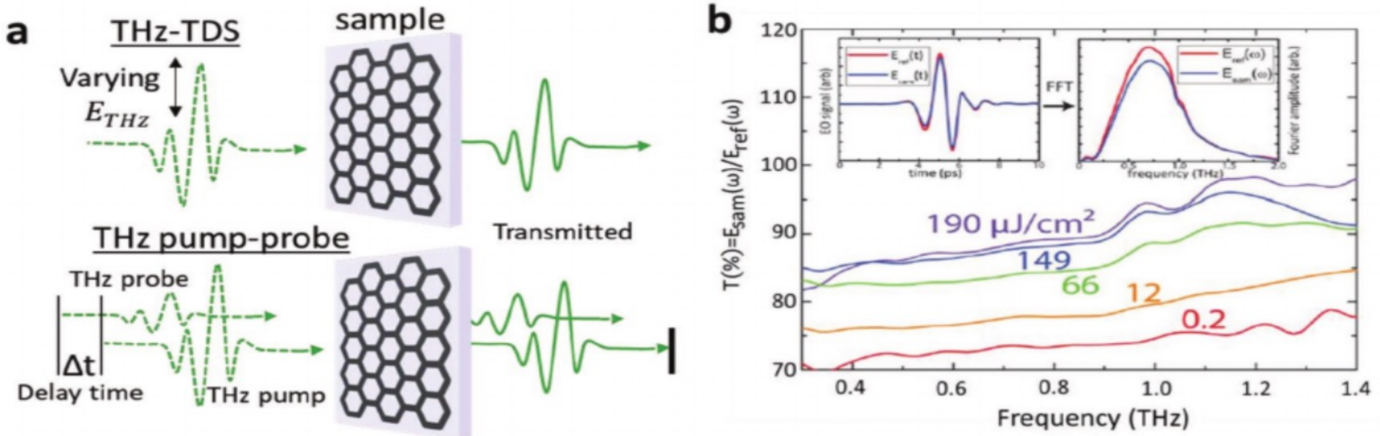
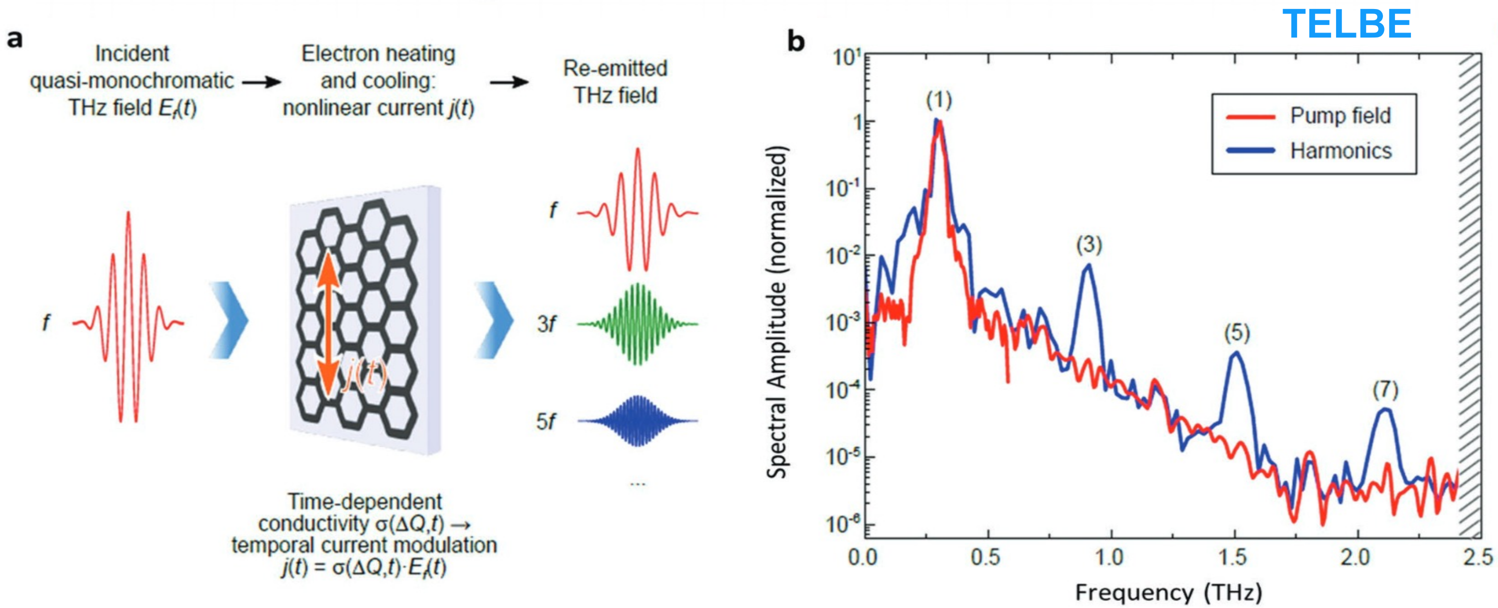
THz nonlinearities in graphene



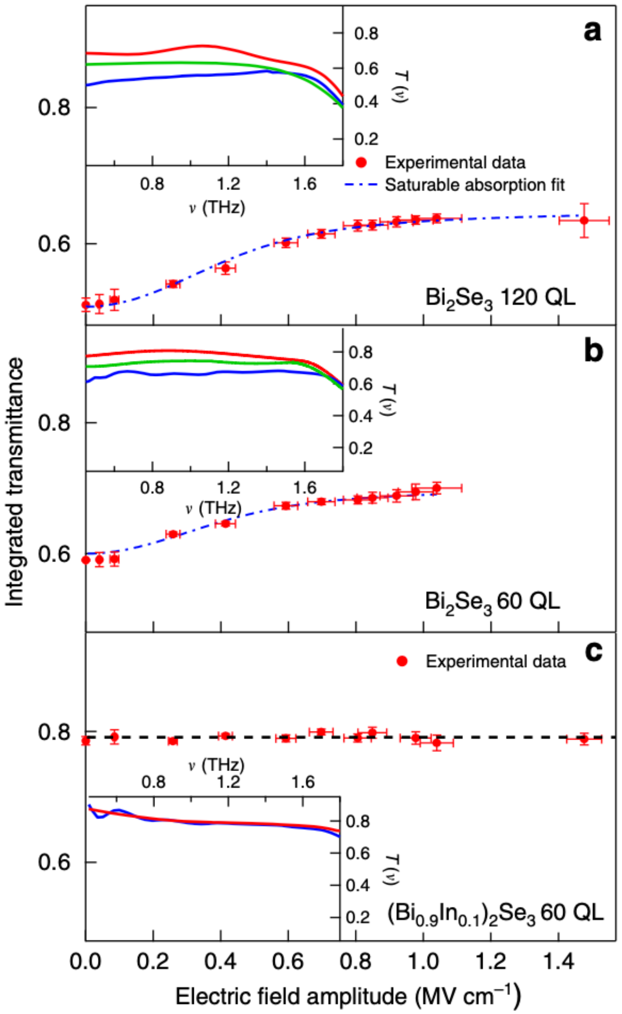


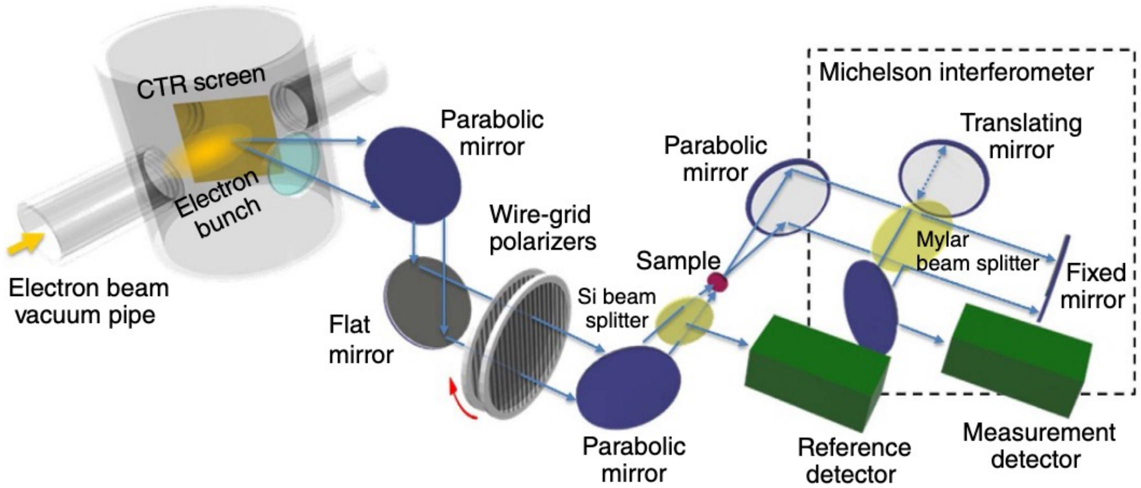
A. Hafez et al., Advanced Optical Materials (2022)

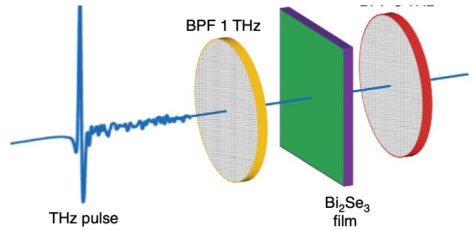
THz nonlinear properties of Bi_2Se_3

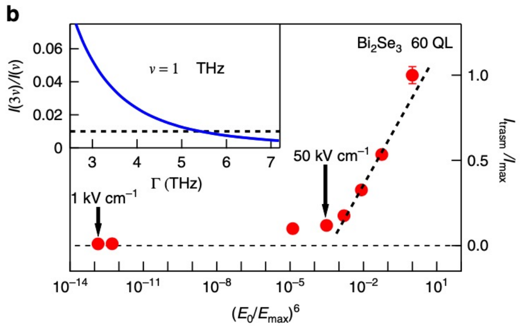
Elettra Sincrotrone Trieste

THz@SPARC-Lab



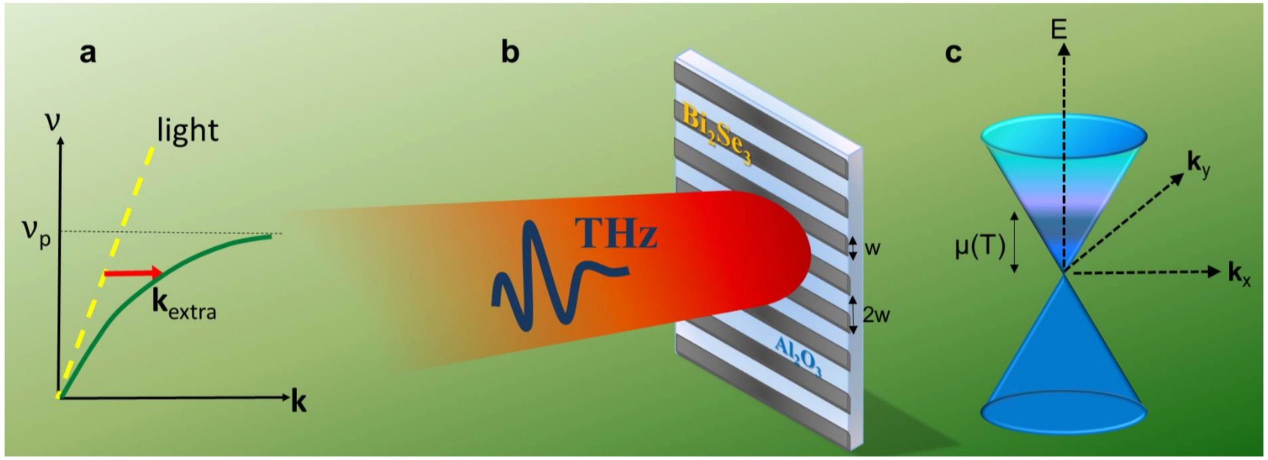


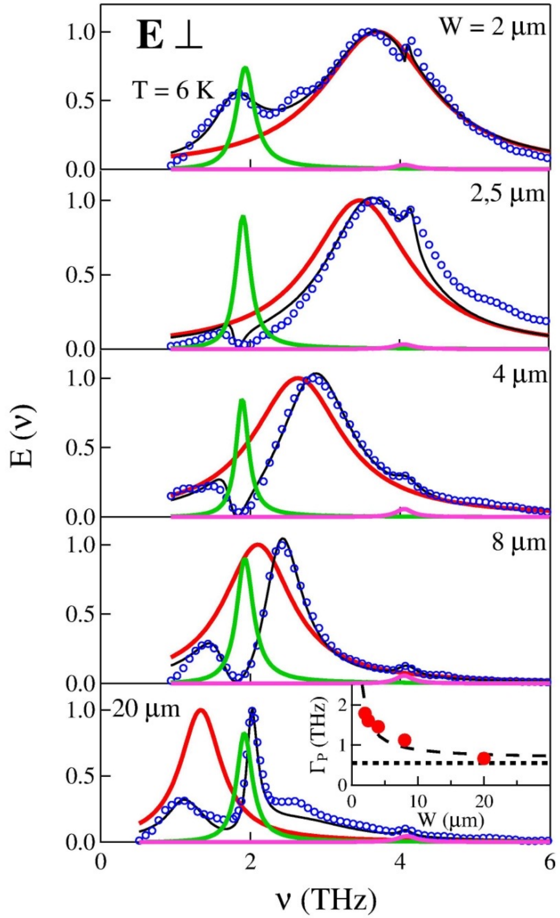




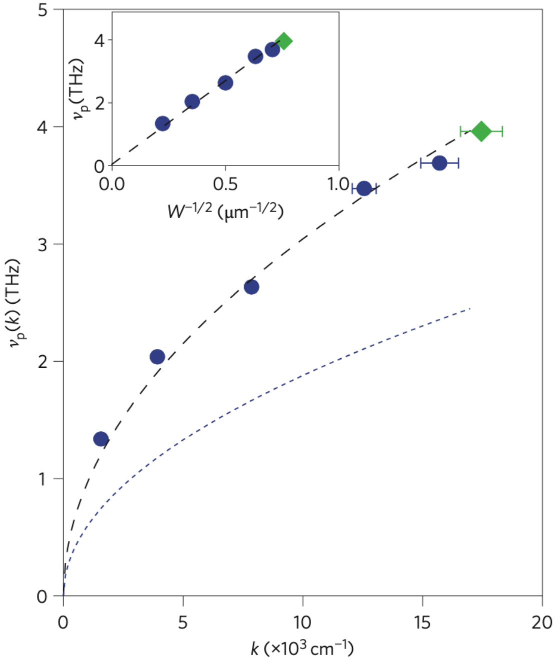
F. Giorgianni et al., Nature Communications (2016)

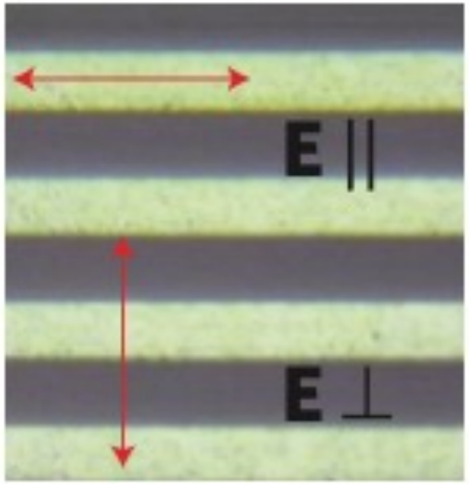
THz Plasmonics on Bi_2Se_3





Fano fitting

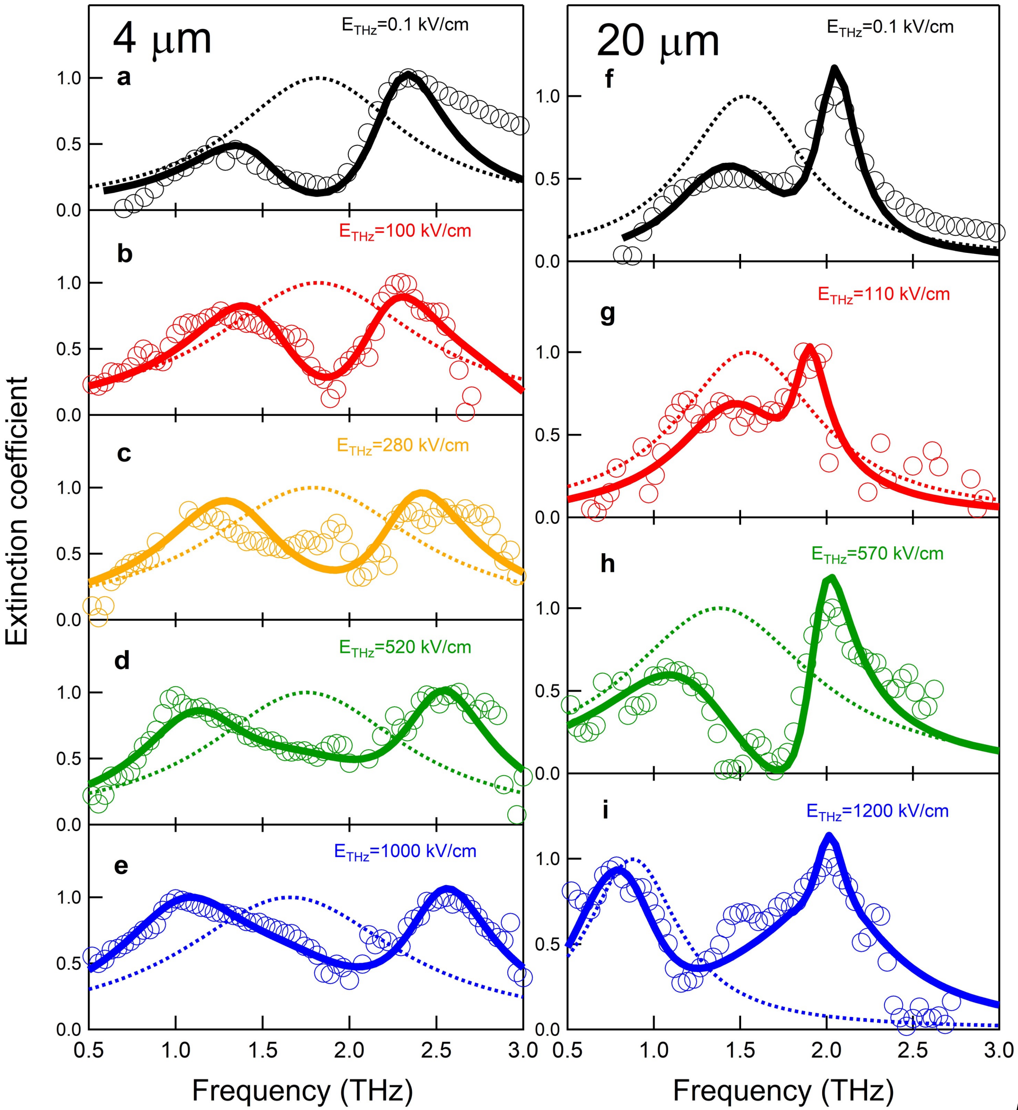


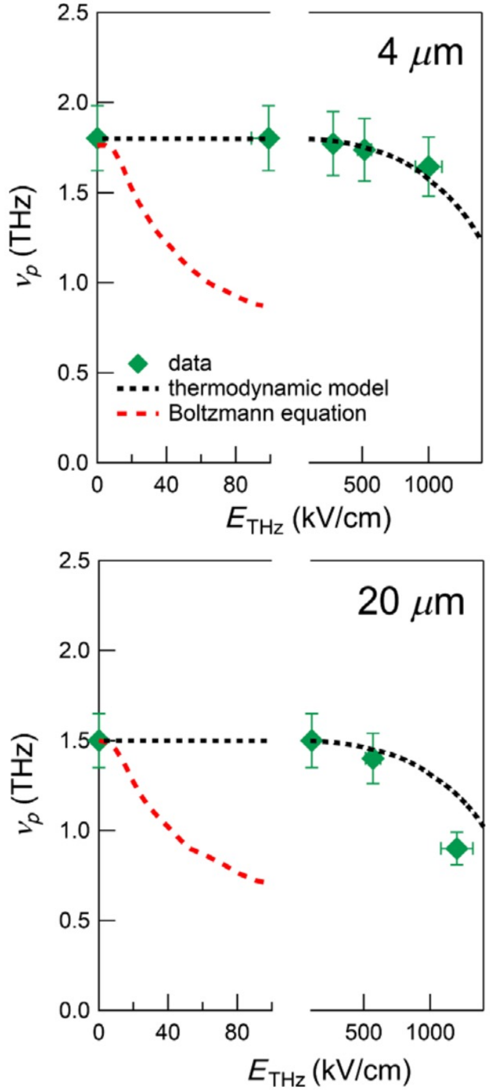


P. Di Pietro et al., Nature Nano. 2013

Fluence-dependent THz properties

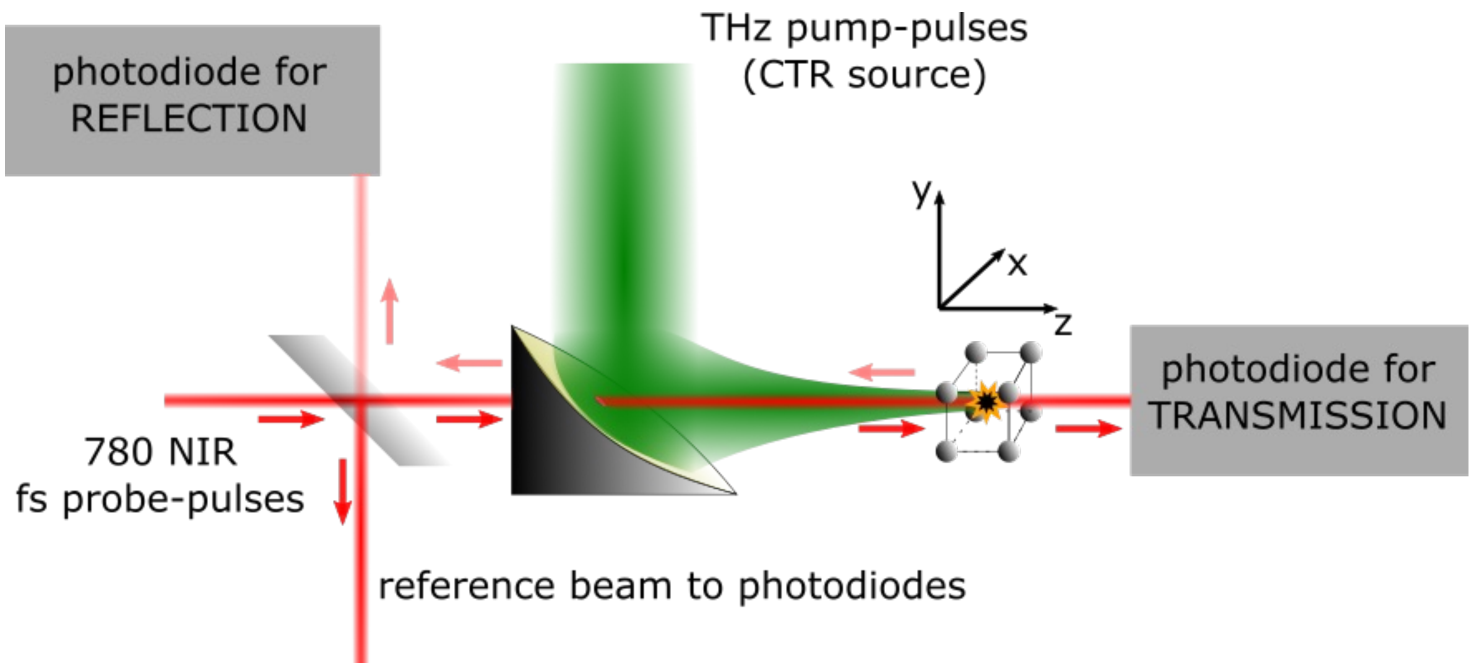
TeraFERMI





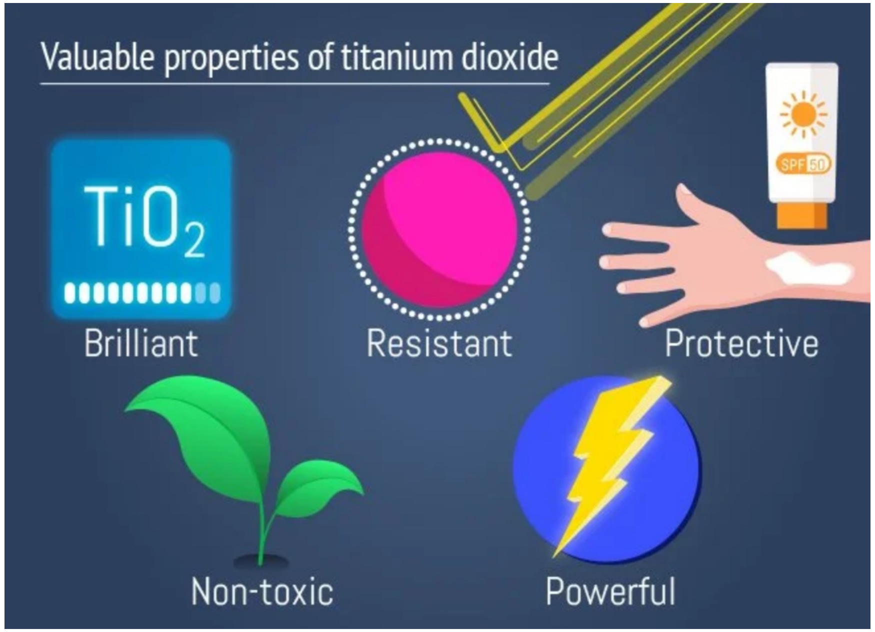
P. Di Pietro et al., PRL 2020

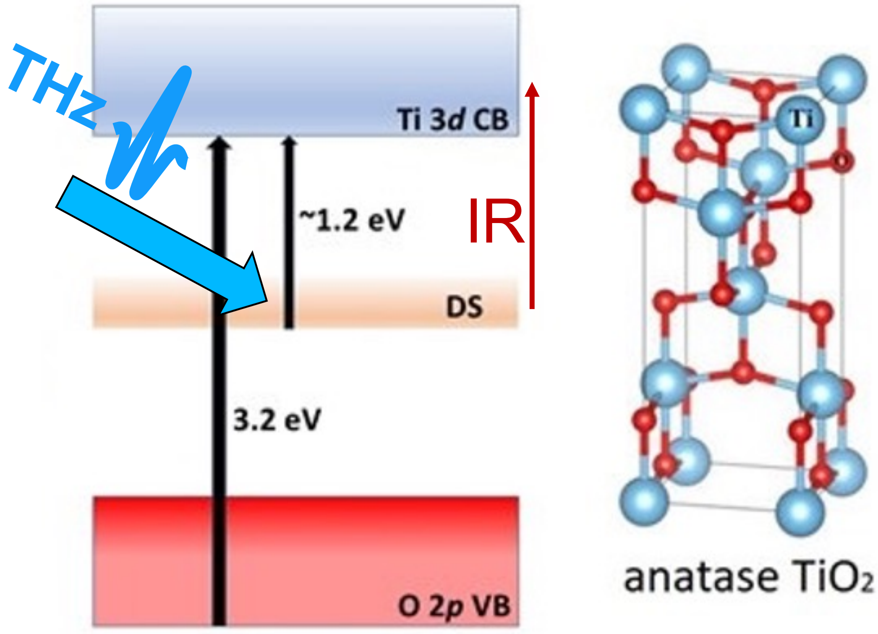
THz-pump/IR-probe



- Normal incidence geometry
- Simultaneous measurement of reflectance and transmittance
- Terahertz Kerr configuration also available

THz control of the O_2 defect state in TiO_2



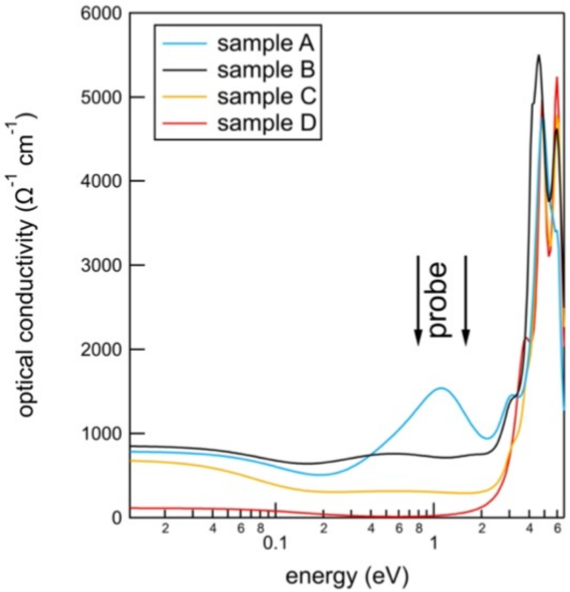


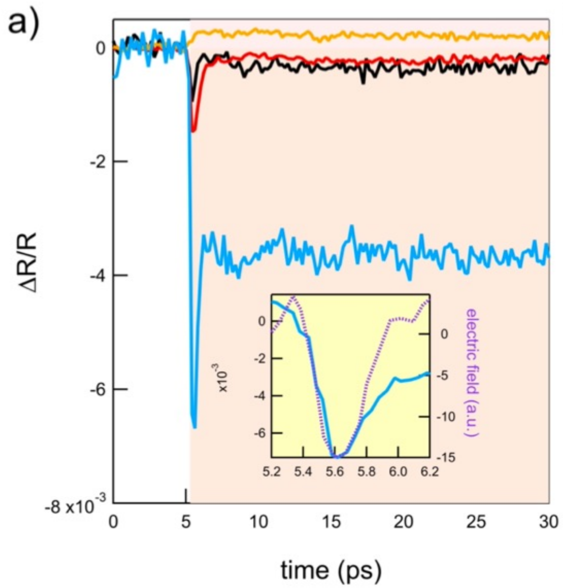
Chemical doping arising from oxygen vacancies induces the creation of in-gap defect states (DS)
ARPES measurements confirm the existence of both localized (IG) and delocalized (2DEG) electronic states with Ti^{3+} and Ti^{4+} character, respectively.

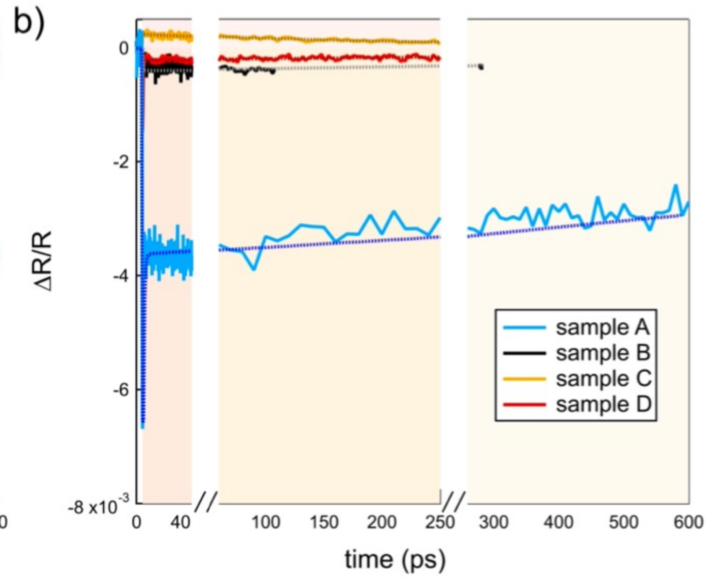
Using THz to control and modulate on ultrafast timescales the photoexcitation of the Defect State

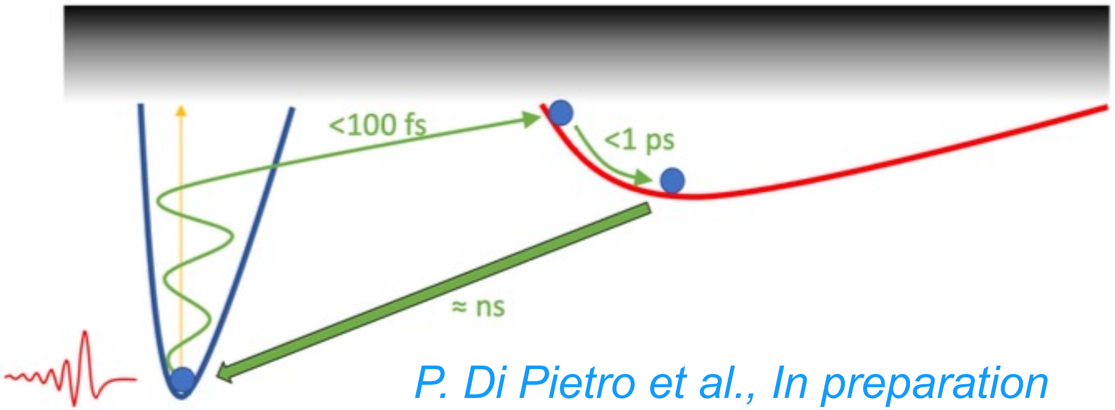
THz control of the O_2 defect state in TiO_2

TeraFERMI





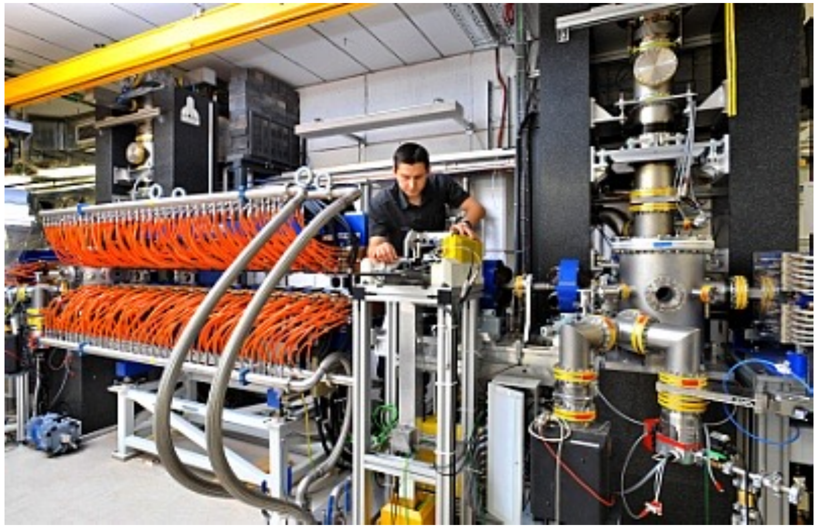




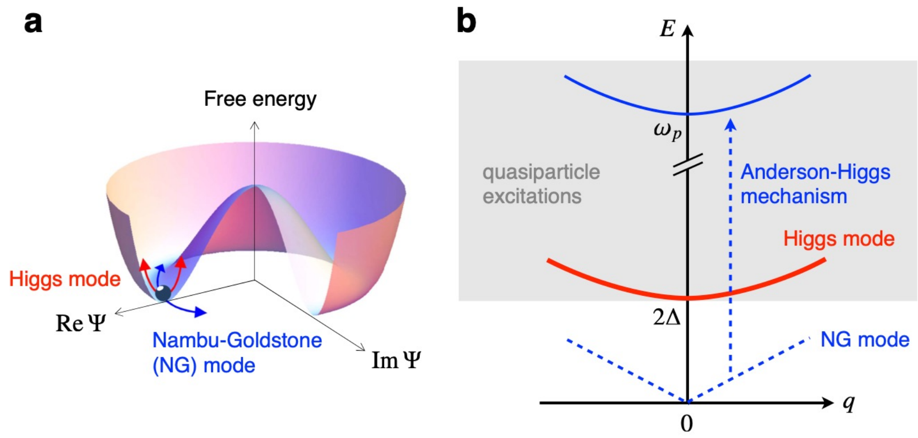
P. Di Pietro et al., In preparation

Narrowband THz

Superradiant undulator
TELBE



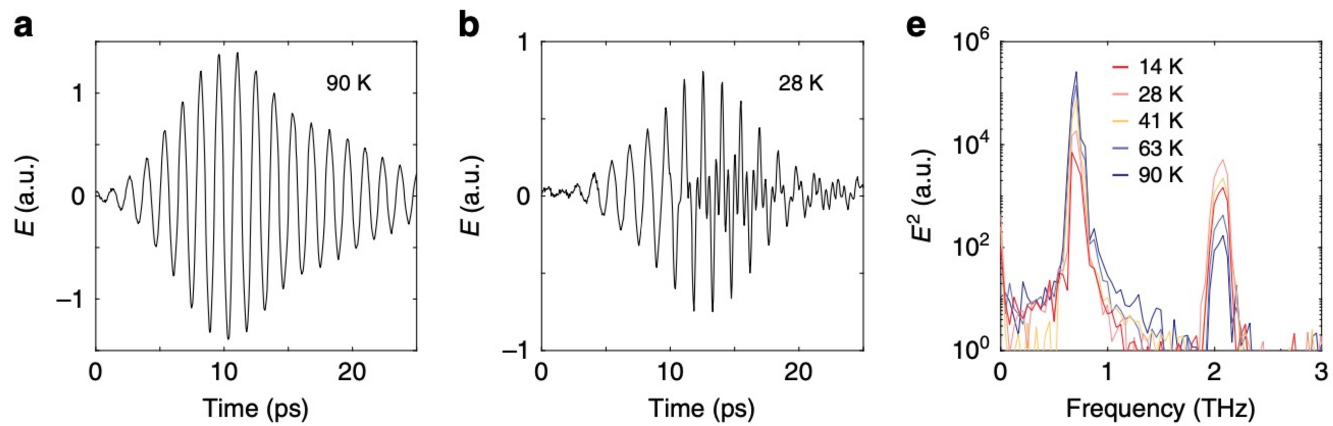
THz Higgs modes in superconductors



The Higgs mode does not have any electric charge, electric dipole, magnetic moment, and other quantum numbers. Therefore ***it does not couple*** *to external probes such as electromagnetic fields in the* ***linear****-response regime"*

R. Shimano & N. Tsuji, Ann. Rev. Cond. Mat. Phys. **2020**

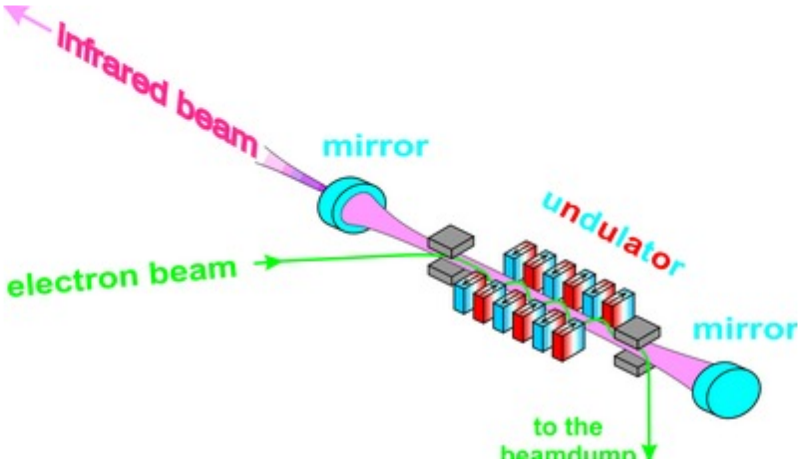
TELBE



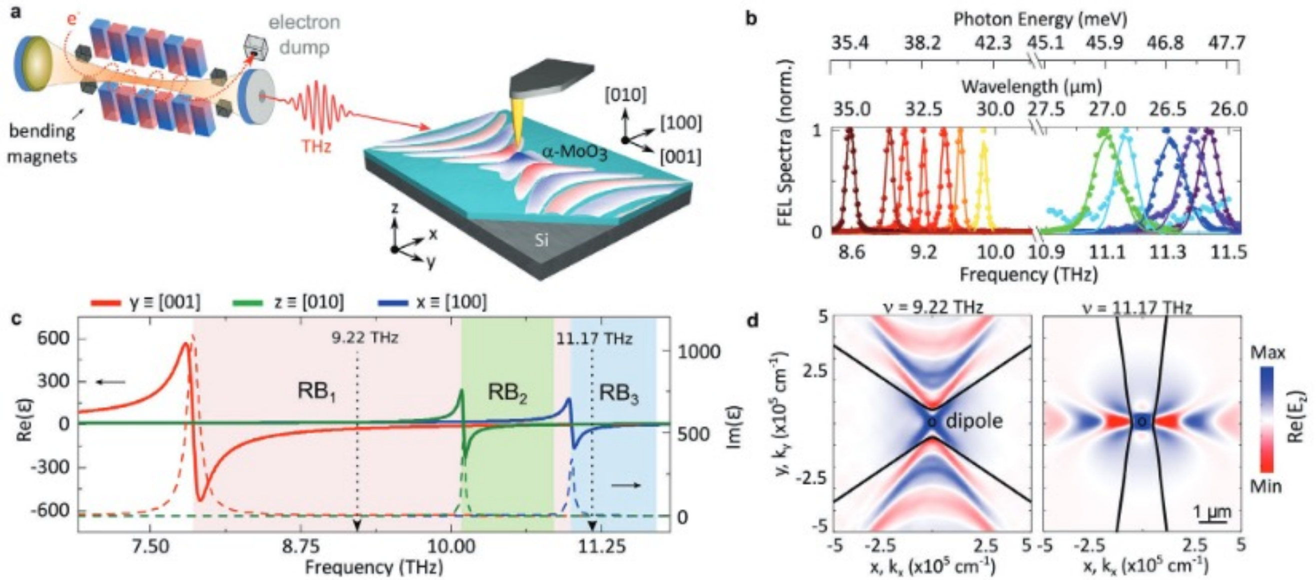
H. Chu et al., Nat. Comm **2020**

Narrowband THz

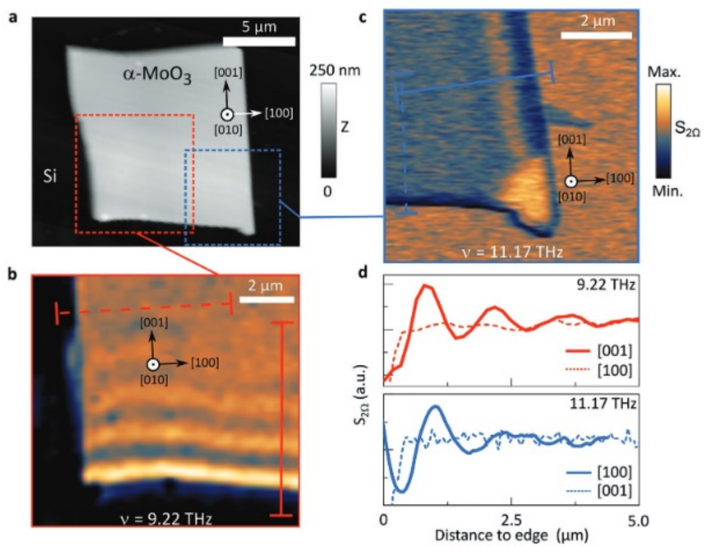
Cavity FEL
FELBE



Nanoscale THz phonon polaritons



FELBE

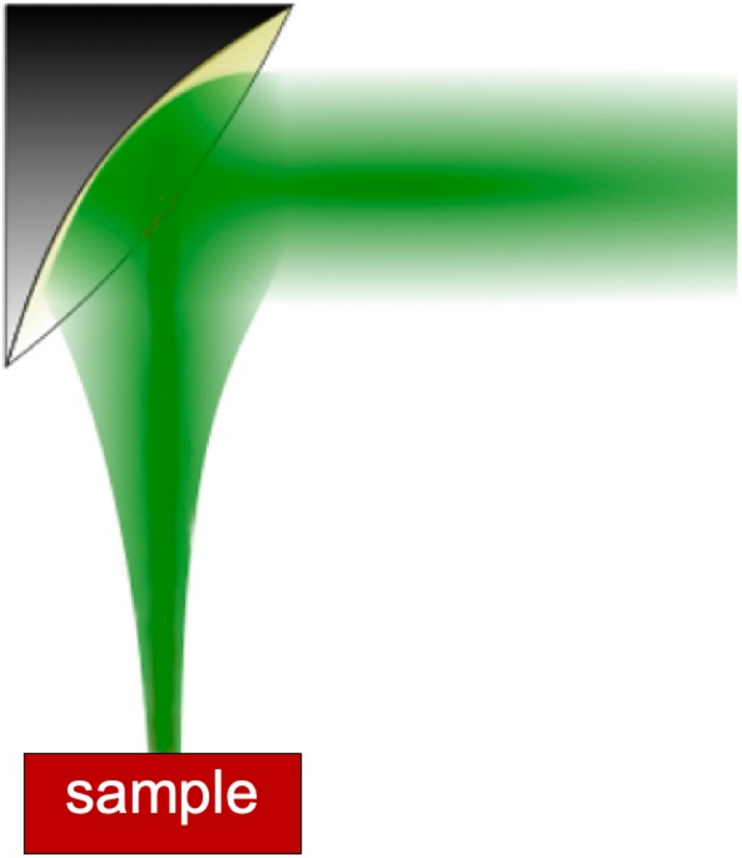


Near-field images show periodic signals (fringes) parallel to specific flake edges. This reveals PhPs propagating with strongly in-plane anisotropic character.

At 9.22 THz the polariton wavelength is 2.82 µm << λo = 32.5 µm

At 11.7 THz the polariton wavelength is 3.9 µm << λo = 26.8 µm

Terahertz Irradiation



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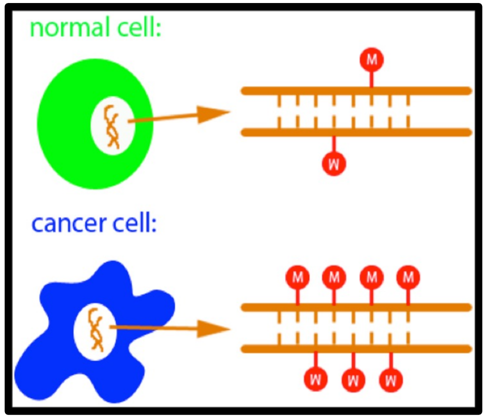
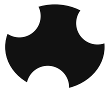
THz irradiation

THz-induced demethylation



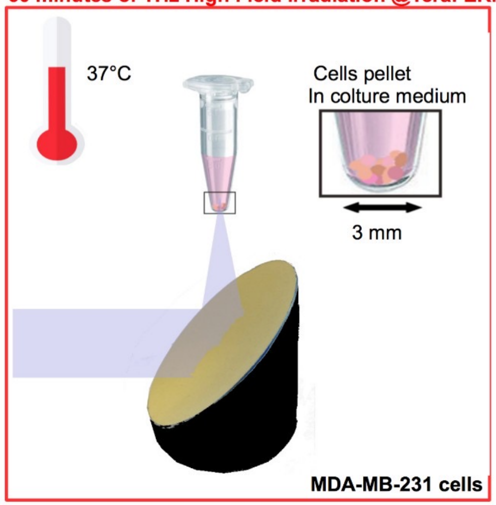
Cancer cells can show enhanced
DNA Methylation

Methyl groups



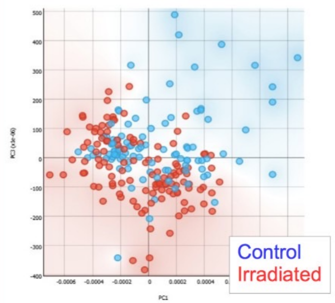
Treatment

60 minutes of THz High Field Irradiation @TeraFERMI



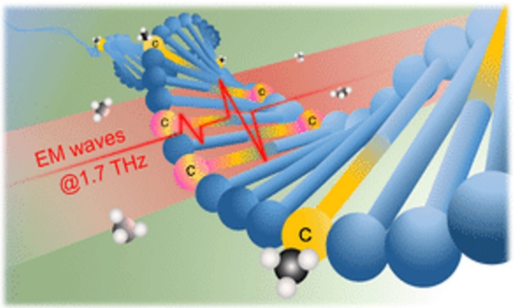
Probe

SR-IR @SISSI on single cells



F. Piccirilli, M. Pachetti et al. (2021)

TeraFERMI



Intense resonant THz radiation induces DNA demethylation

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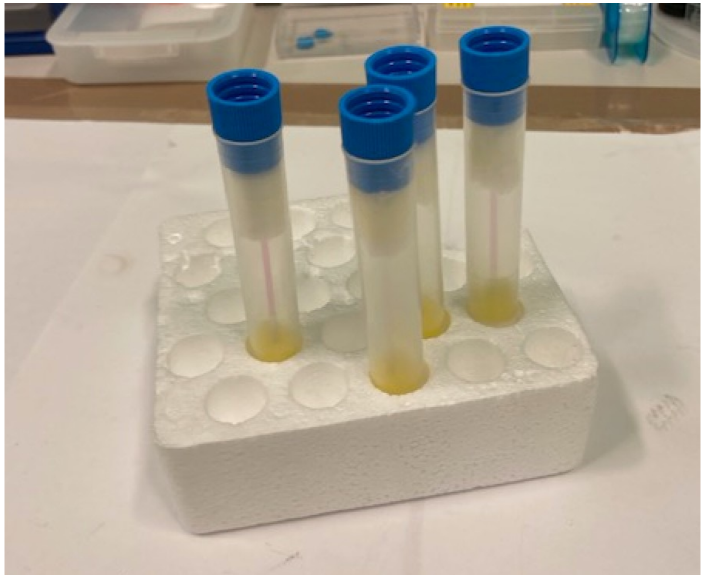
THz irradiation

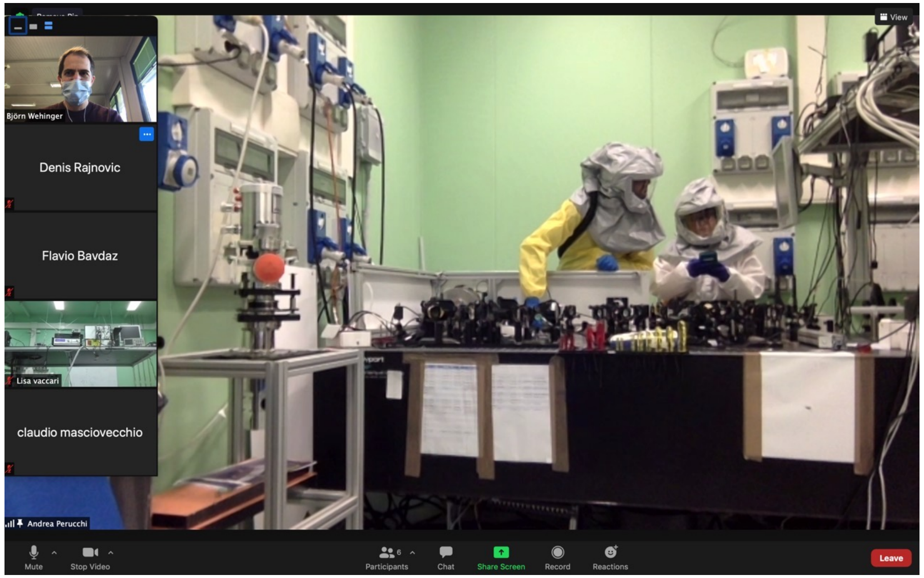
Virus inactivation through THz irradiation

RNA modes resonate at THz frequencies

➔ Inducing RNA damage while keeping the capsid intact

➔ vaccines





Most plastics are transparent to THz radiation
Unbreakable sample holders can be used
Virus is sealed within 2 plastic containers

Elettra
Sincrotrone
Trieste

Acknowledgments

TeraFERMI

P. Di Pietro

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SISSI-Bio

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G. Birarda
F. Piccirilli
D. Bedolla
M.C. Stani

SISSI-MAT

S. Lupi
M. Zacchigna
V. Stopponi

iom
Istituto Officina dei Materiali

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