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

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THz Spectroscopy using Coherent Synchrotron Radiation

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für Synchrotronstrahlung m.b.H.

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using
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Leibniz Gemeinschaft

THz Radiation from a Storage Ring:

Infrared Synchrotron Radiation: Source Parameters

Coherent Synchrotron Radiation: Mechanism, Properties

Instrumentation:

Infrared Beamline at BESSY, Detectors, Spectrometers

Application of the CSR:

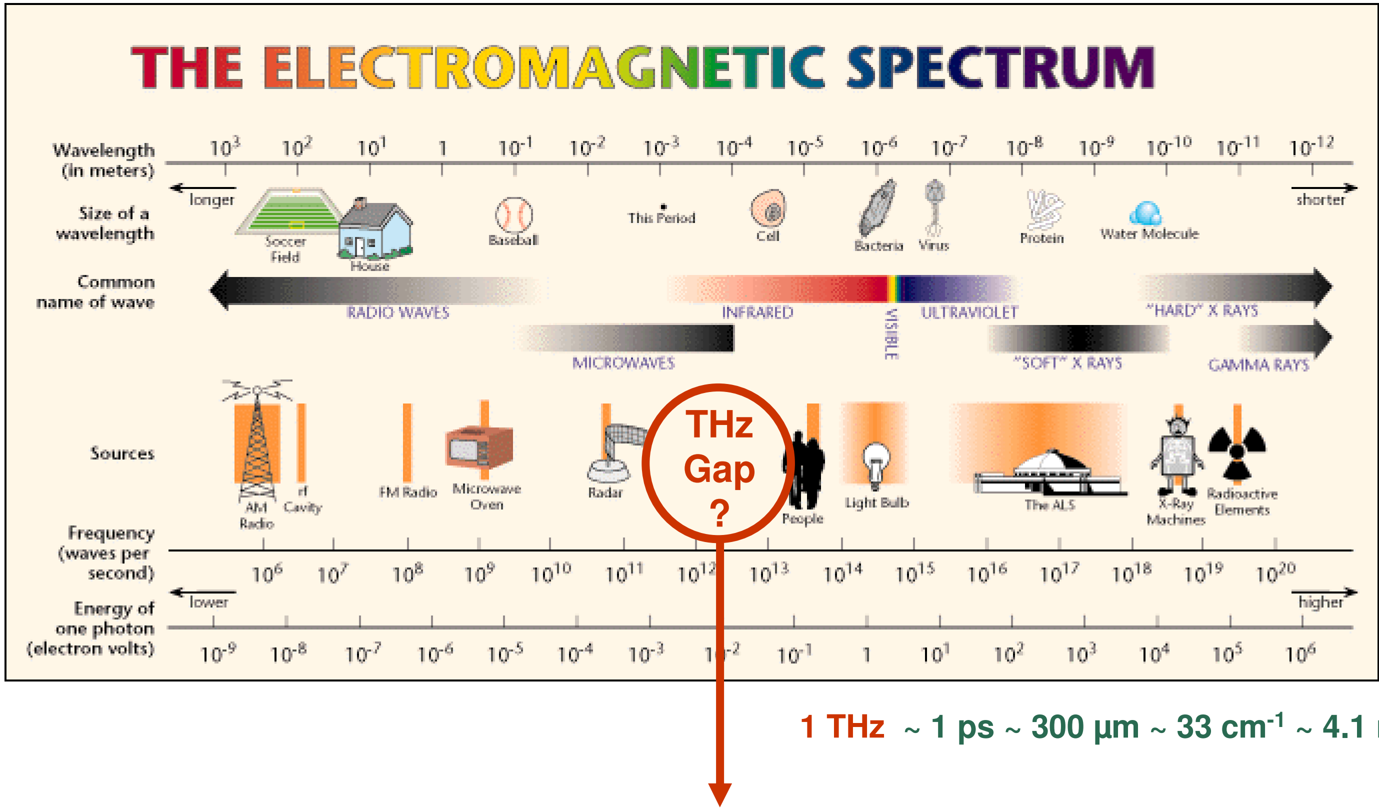
Superconductors

THz Near-field Spectroscopy

Conclusions:

Introduction:

http://www.lbl.gov/MicroWorlds/ALSTool/EMSpec/EMSpec2.html



1 THz ~ **1 ps** ~ **300 µm** ~ **33 cm^{-1}** ~ **4.1 meV** ~ **47.6 K**

Far-IR Broadband Techniques:

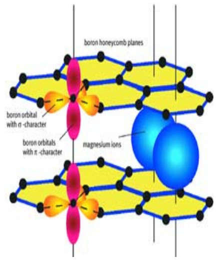
- Backward Wave Oscillators
- Time-domain Spectroscopy
- Coherent Synchrotron Radiation

Condensed Matter Physics

Superconductivity
- Energy gap
- Symmetry of the order parameter
- Strength of coupling

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

Strongly correlated electrons
- Kondo problem
- Heavy electrons



Life Sciences

Protein dynamic
- Secondary and tertiary structure

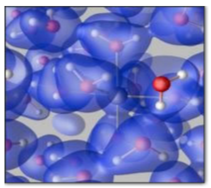
Metabolism
- Influence of nutrition, water
- Ion channels in cell membranes

Imaging
- 3D tomography of dry tissues
- Near-field

Physical and Analytical Chemistry

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

Solutions
- Interactions between solvated ions and solvent



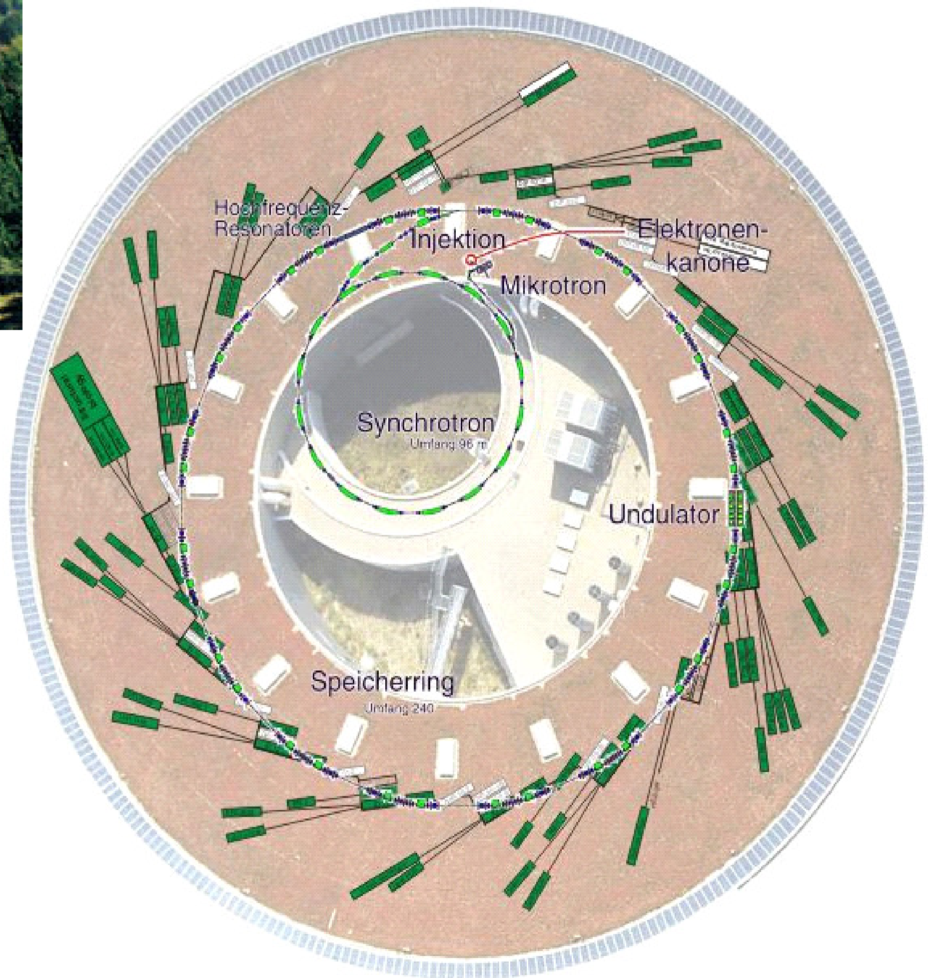
New Technologies

Medical diagnostic
- Early cancer detection

Industrial production
- Material inspection

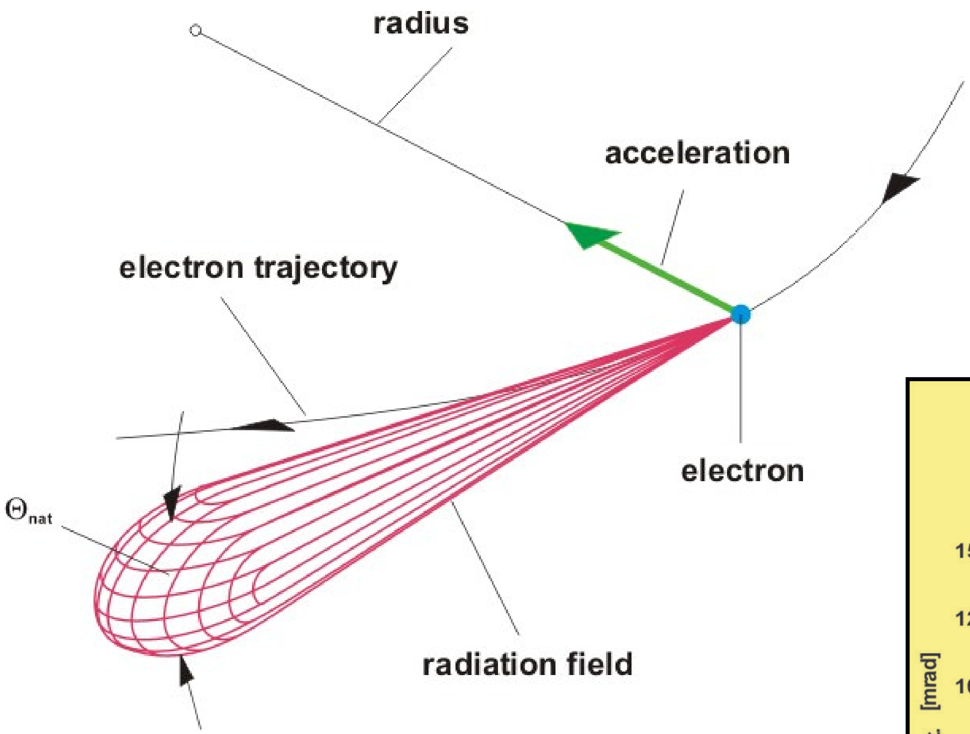
Defense industry/Homeland security
- Detection of explosives and biohazards

Start user operation:	1999
Circumference of the synchrotron:	96 m
Circumference of the storage ring:	240 m
Number of bending dipoles:	2 x 16
Number of possible insertion devices:	15
Number of beamlines commissioned:	~ 50
Commissioning of the IR-beamline IRIS:	2002

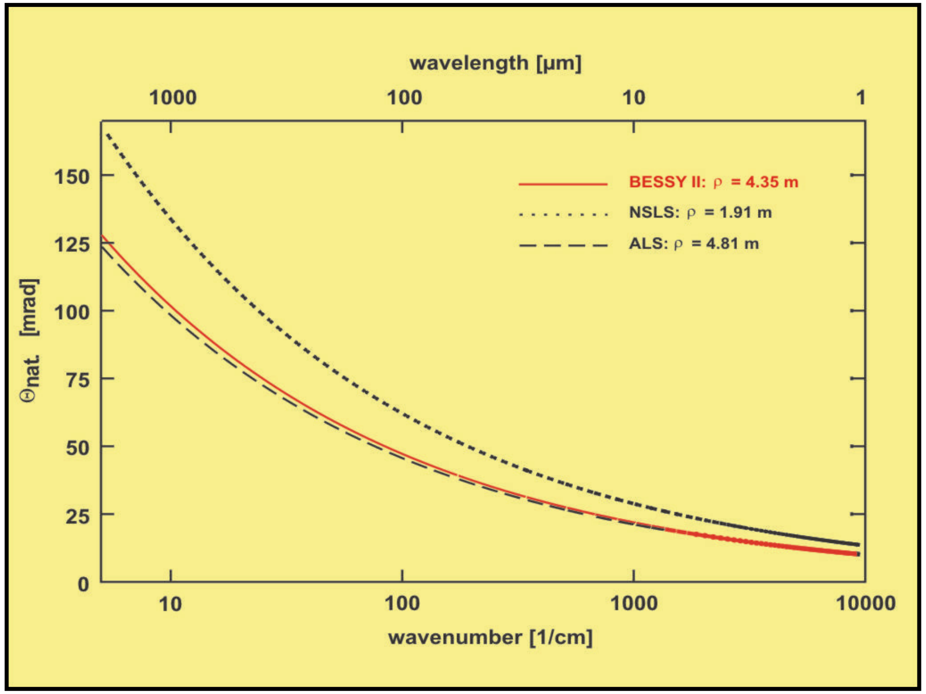


THz Radiation from a Storage Ring:

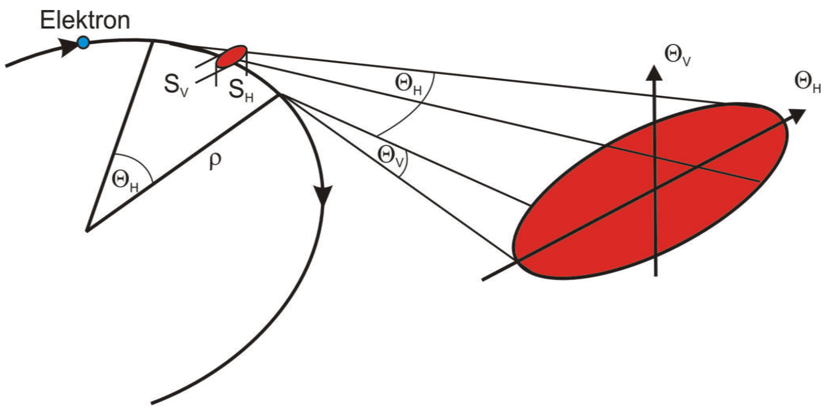
- Infrared Synchrotron Radiation: Source Parameters
- Coherent Synchrotron Radiation: Mechanism, Properties



Synchrotron radiation is generated from relativistic charged particles, e.g. electrons, subjected to a transverse acceleration, e.g. by a magnetic field and is emitted into an natural opening angle.



In the far-infrared (THz) region the natural opening angle of the synchrotron radiation drastically increases with decreasing photon energy.

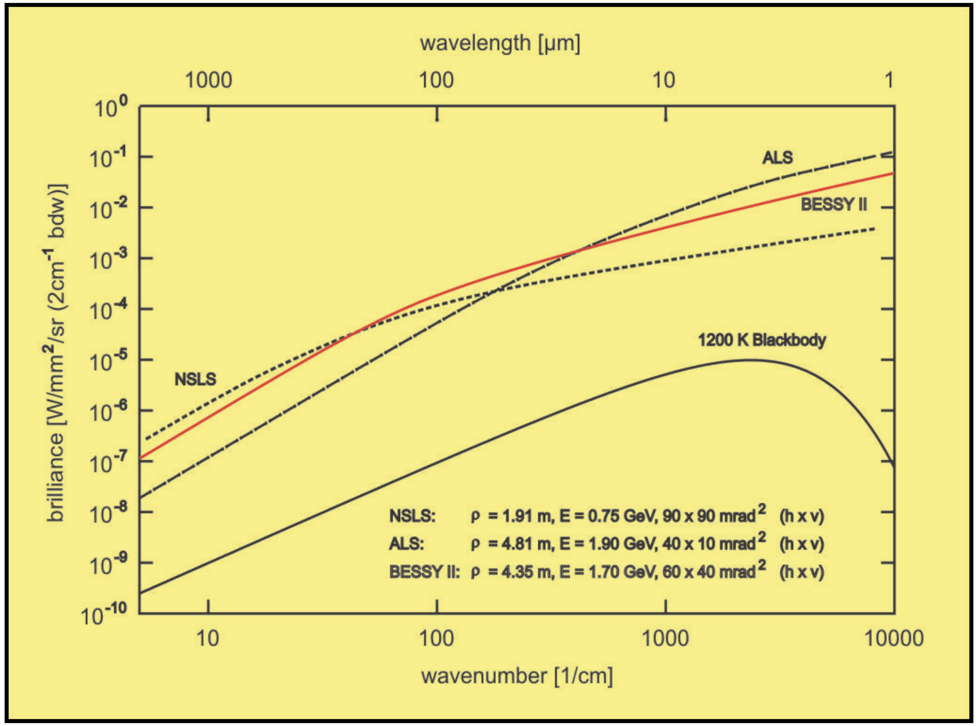


Source Dimensions:

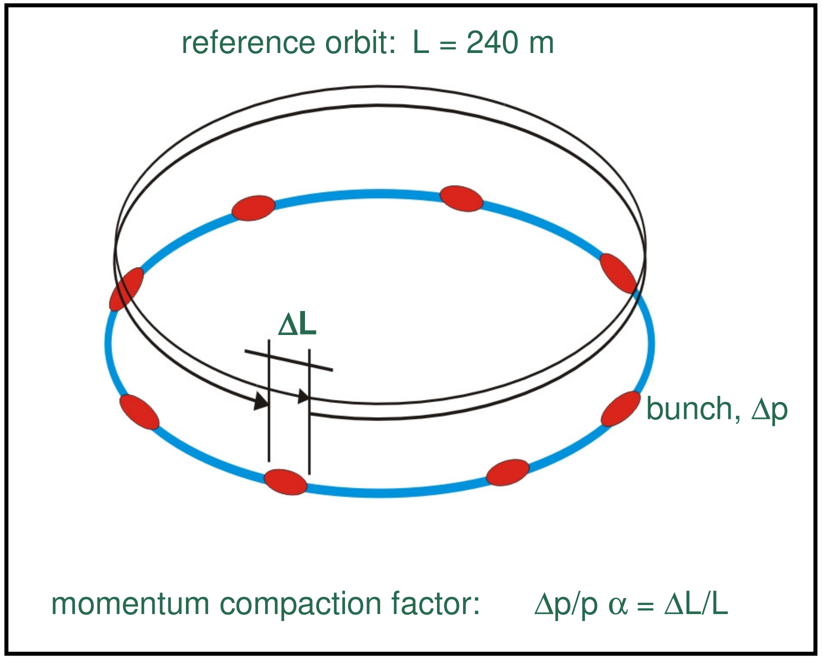
$$s = \sqrt{s_{electron}^2 + s_{projection}^2 + s_{diffraction}^2}$$

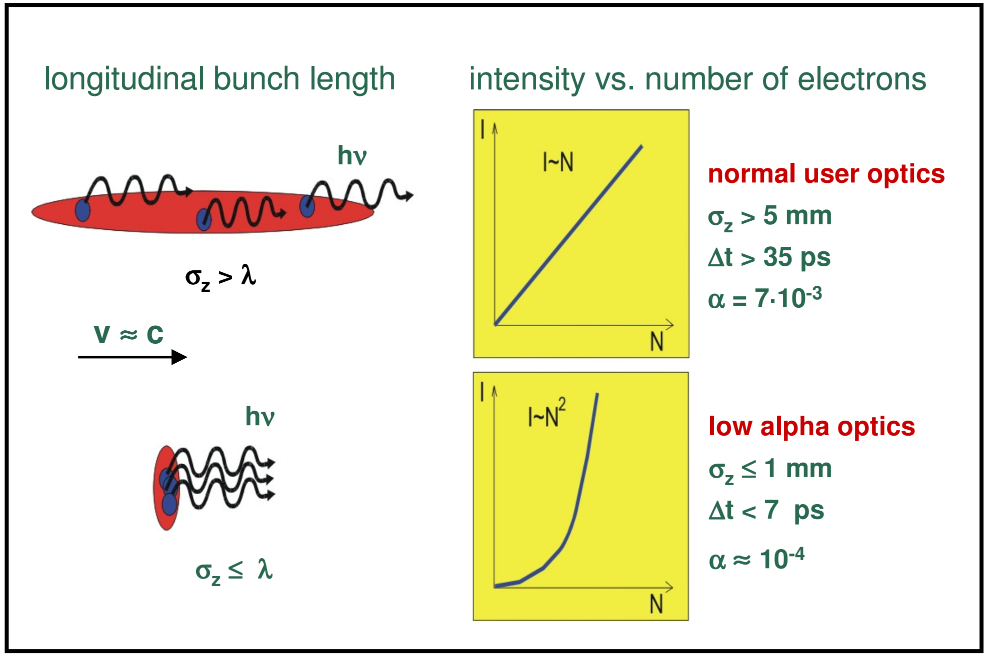
Source brilliance:

$$B = \frac{photon\ flux}{s_v \cdot s_h \cdot \Theta_v \cdot \Theta_h}$$



The high brilliance of the infrared synchrotron radiation source is of advantage to low-throughput experiments.





Dedicated Machine Mode: "Low α" Optics at BESSY:

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

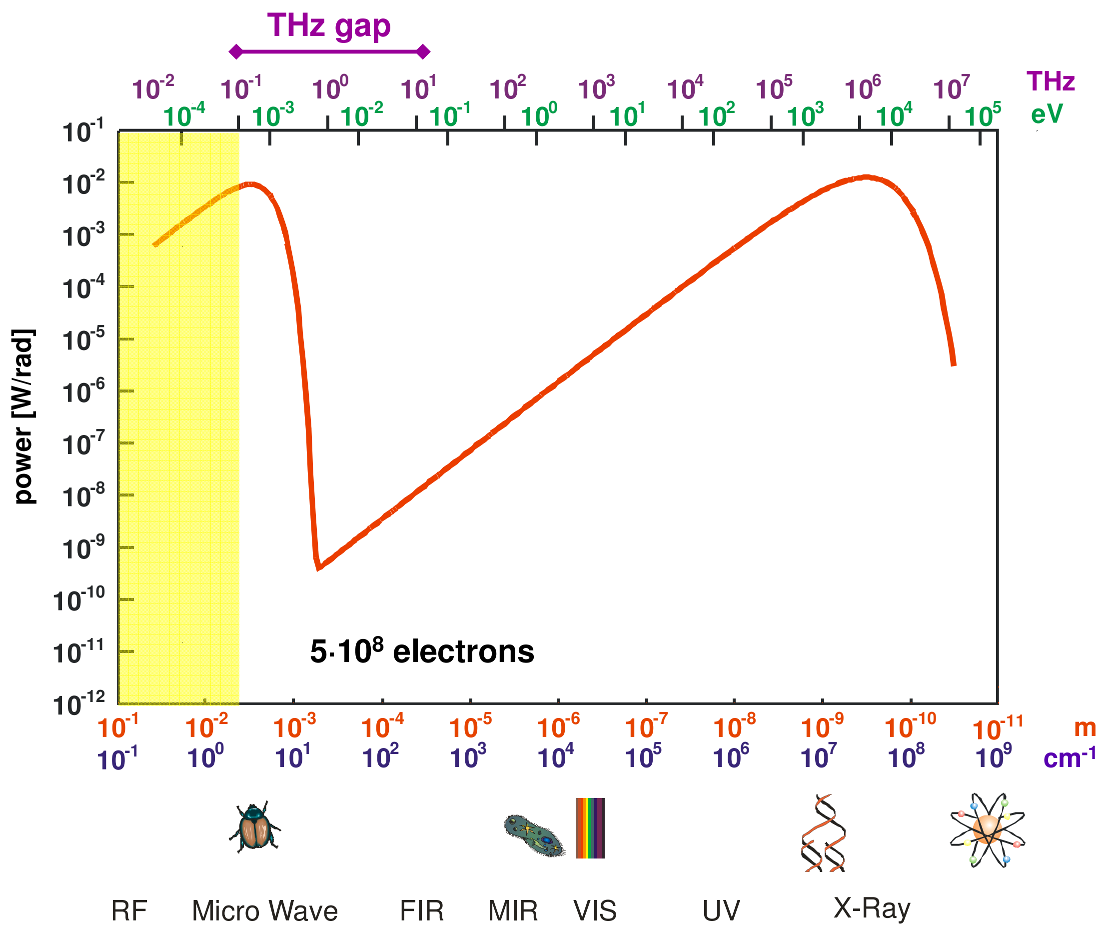
$$I = I_{incoh} + I_{coh} = Ni(1 + Nf_{\nu}) \,, \qquad A_f = \frac{I_{coh}}{I_{incoh}} = Nf_{\nu}$$

Coherent Synchrotron Radiation (CSR)

N-times higher intensity (Gaussian bunch assumed!).

Cut-off due to shielding effects.

Powerful source emitting in the THz and sub-THz range.



CSR at higher frequencies observed than for Gaussian bunches expected

With increasing current of the bunch:

- the CSR spectrum extends to higher photon energies.
- the low-frequency noise in the THz beam drastically inclines.

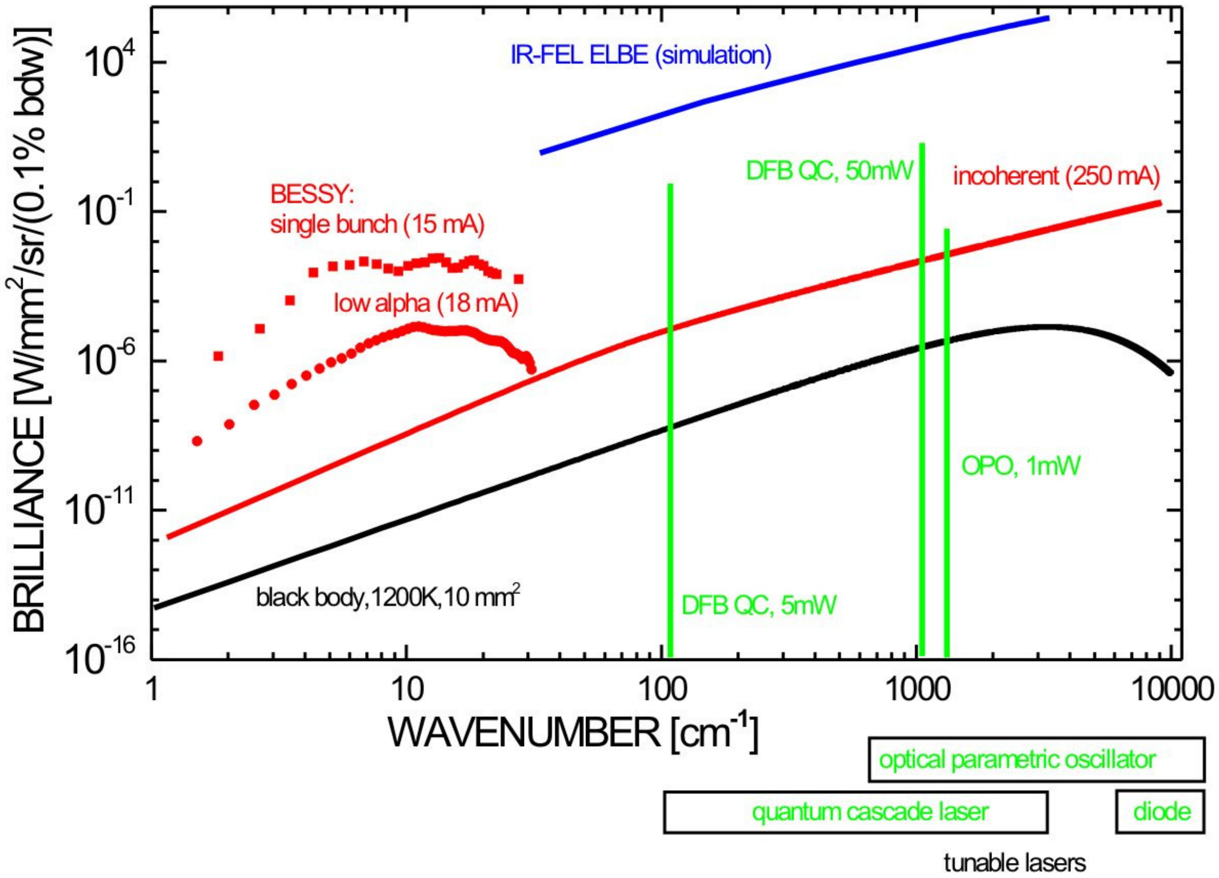
Present understanding:

Interaction of bunch with CSR-wakefield leads to:

- a static non-Gaussian deformation of the bunch (Bane, Krinsky and Murphy, 1996)
  $\rightarrow$ **steady-state CSR**

- bursting CSR emission above a current threshold (micro-bunching, Stupakow and Heifets, 2002)
  $\rightarrow$ **high power bursting CSR**

Steady-state vs. Bursting CRS



"bursting" mode

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

"low α" mode

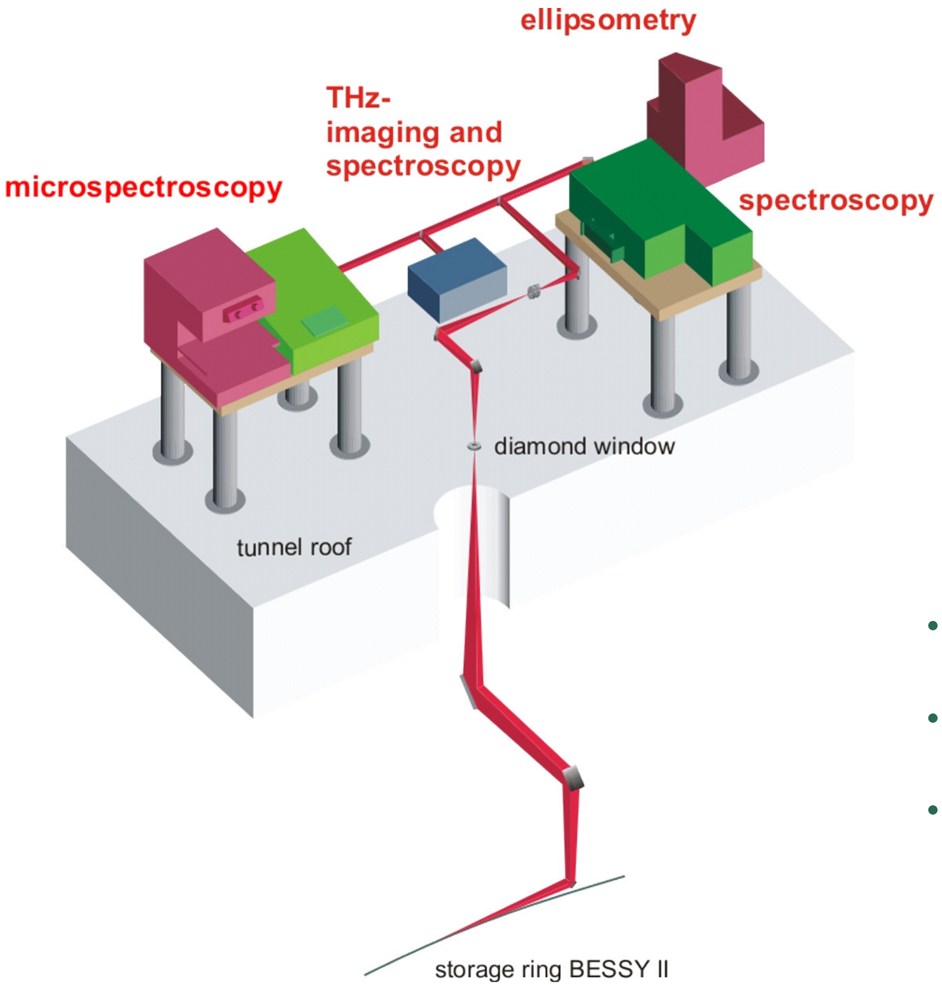
- steady-state CSR
- energy range: 2 - 30 cm^{-1}
- gain of ~10^4

M. Abo-Bakr et al., Phys. Rev. Lett. 88 (2002) 254801 and Phys. Rev. Lett. 90 (2003) 094801.

Instrumentation:

- Infrared Beamline at BESSY, Detectors, Spectrometers

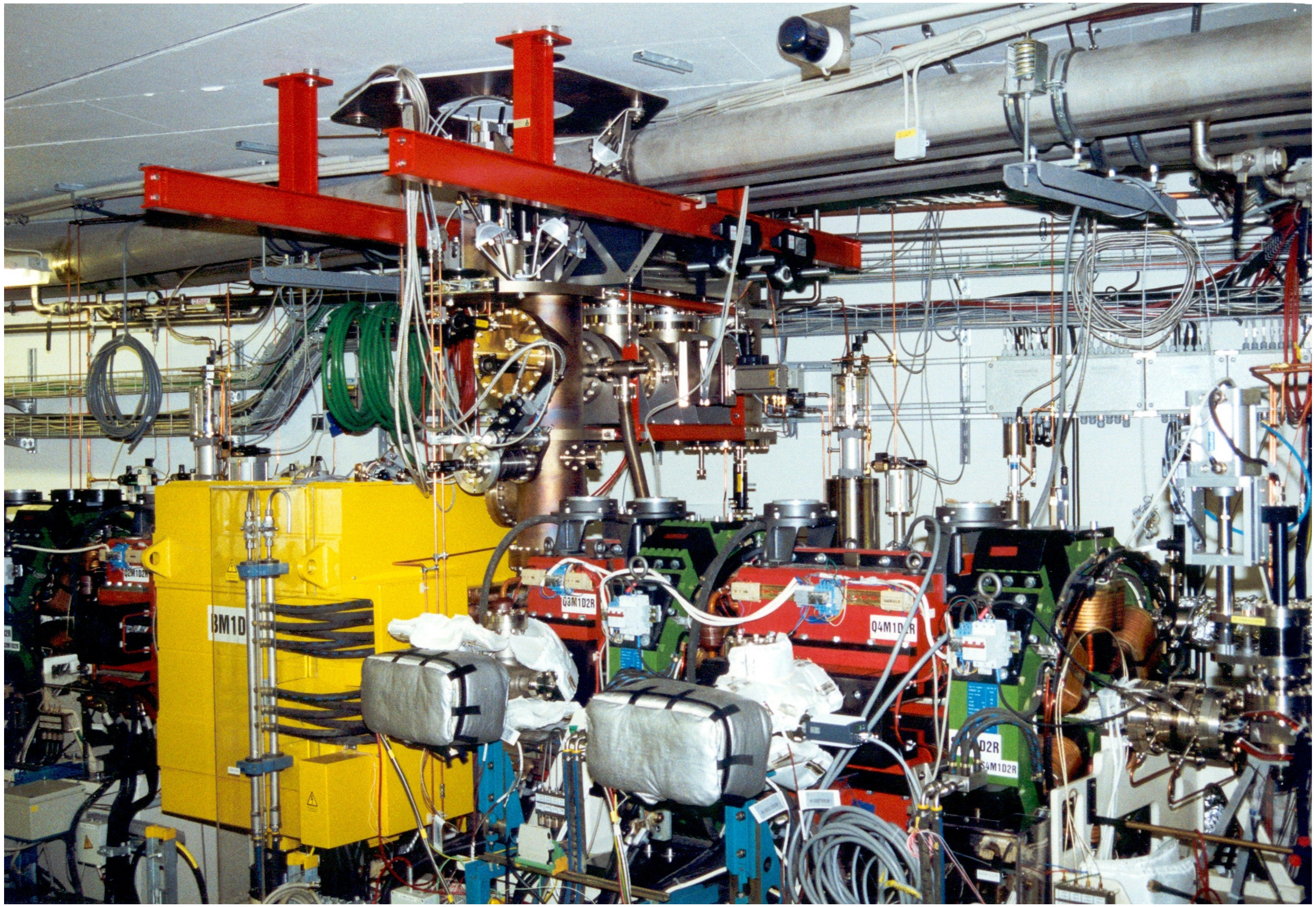
IRIS - Infrared Beamline at BESSY



- Dipole radiation from dipole 2.2
- NIR to FIR
- 60(h) x 40(v) $mrad^2$ acceptance

Schade et al., Rev. Sci. Instr. **73** 1568 (2002).

Infrared Beamline: Front End



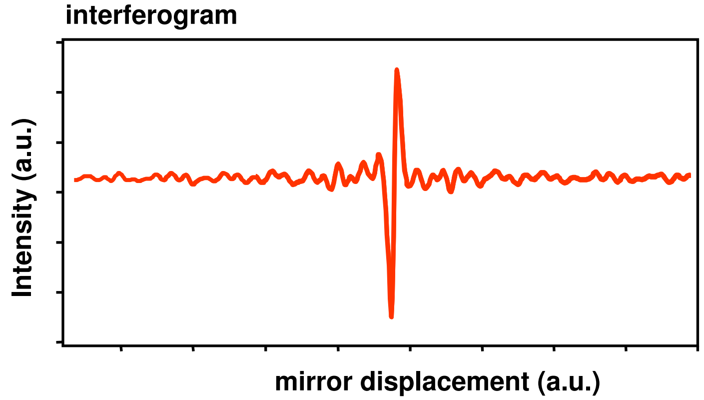
Spectrometer:

	Bruker 66/v	*Martin-Puplett (DLR)*
spectral range (cm^{-1})	2 – 600	2 - 100
beamsplitter	6 µm, 50 µm and 125 µm Mylar	free-standing wire grids

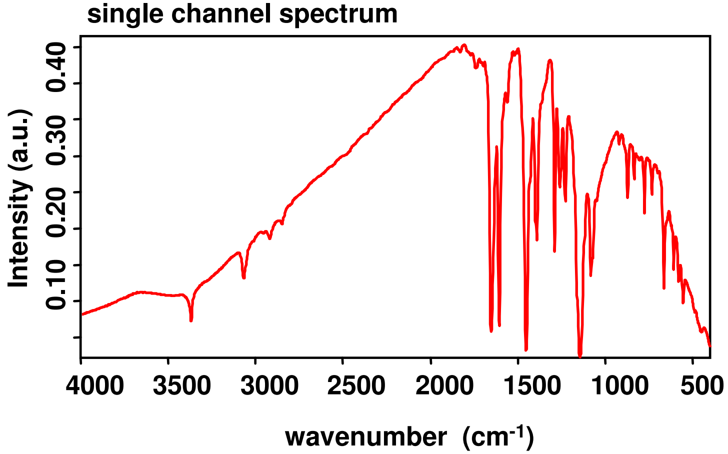
Detector:

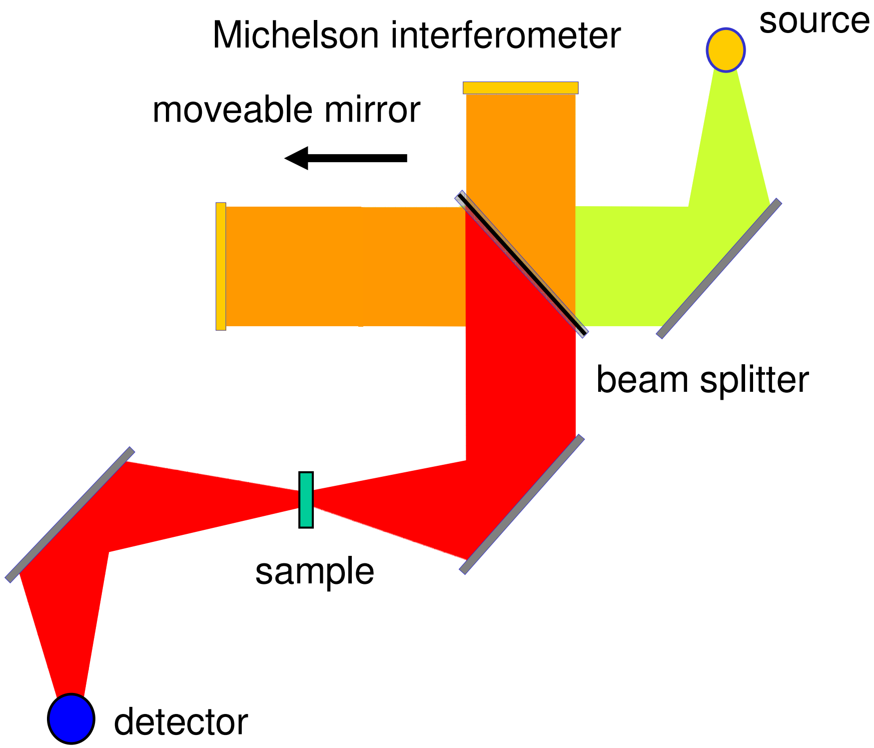
	DTGS	*Si-Bolo 4.2 K*	*Si-Bolo 1.2 K*	*InSb HEB*	*SC HEB*
spectral range (cm^{-1})	50 – 600	10 - 600	2 - 60	2 - 30	7 - 100
max. BW	1 kHz	1kHz	1kHz	1MHz	5 GHz
NEP ($W/\sqrt{Hz}$)	1e-9	1e-13	3e-15	1e-13	1e-12

Fourier Transform-Infrared (FT-IR) Spectroscopy



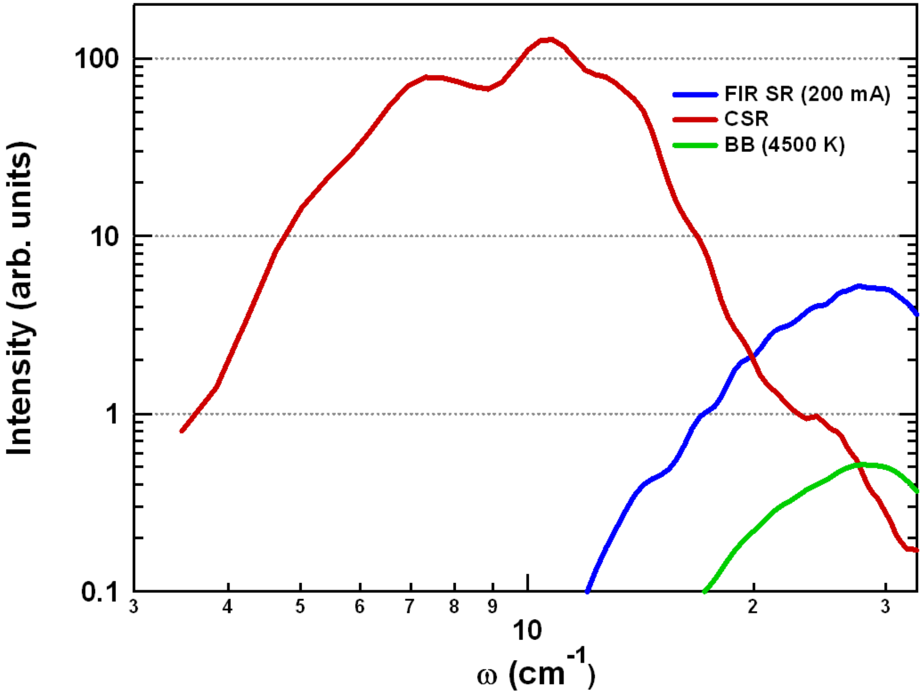
Fourier-Transformation

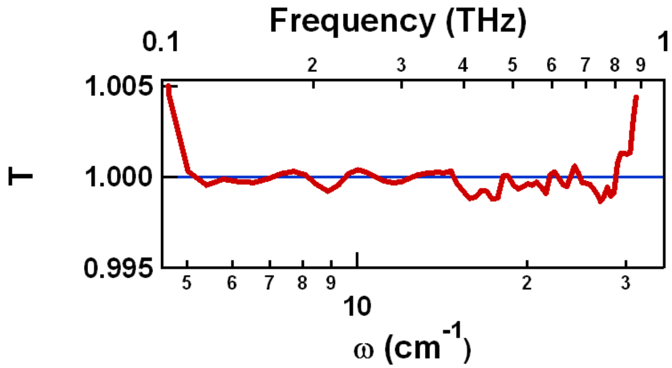




Advantages:

Jacquinot: high throughput
Fellget: multiplex
Connes: high resolution, broadband





"100%-line" for CSR

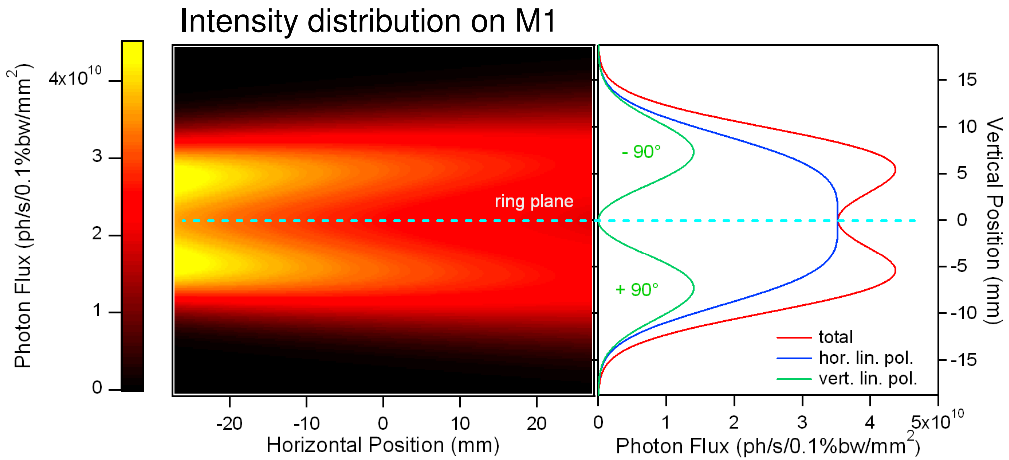
Source Comparison in the Spectrometer

256 scans, $\Delta\omega = 0.5$ cm^{-1}, 1.4 K Bolometer, 5 mm aperture diameter

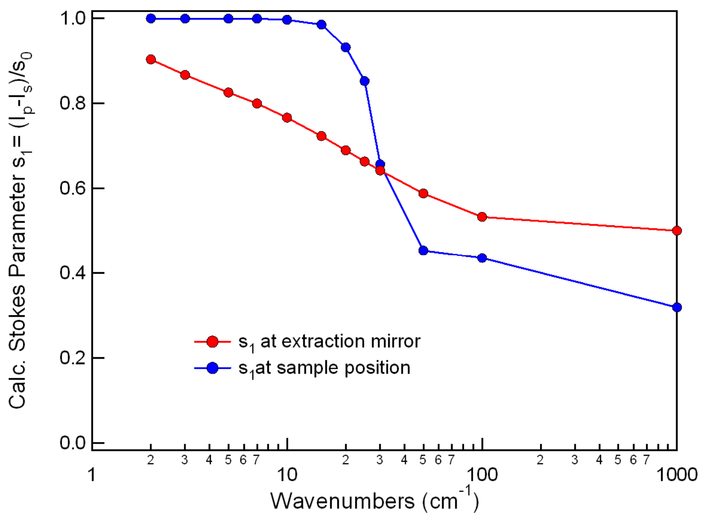
- long life time of the beam (>20 h)
- gain of 10^3 below 10 cm^{-1} (0.3 THz)
- highly reproducible

M. Ortolani *et al.,* Phys. Rev. B **73**, 184508 (2006).

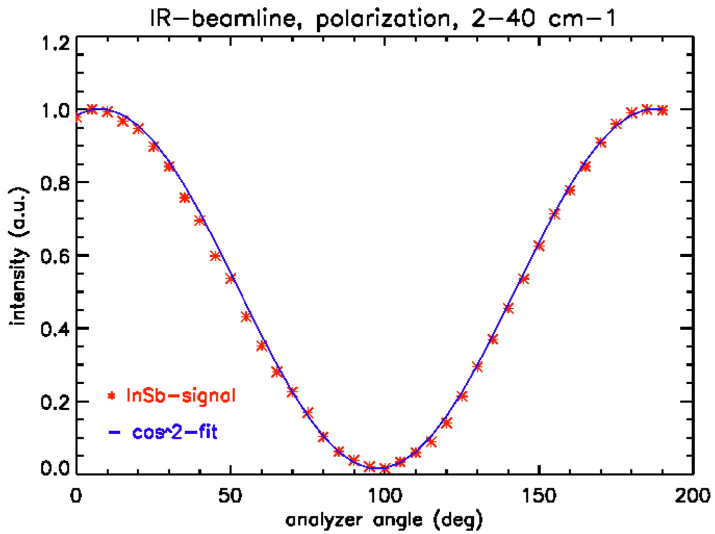
Polarization Characteristics of CSR



Polarization properties of synchrotron radiation from a bending magnet source at 500 cm^{-1}.



Calculated s1 for the entrance and for the end focus of the beamline.

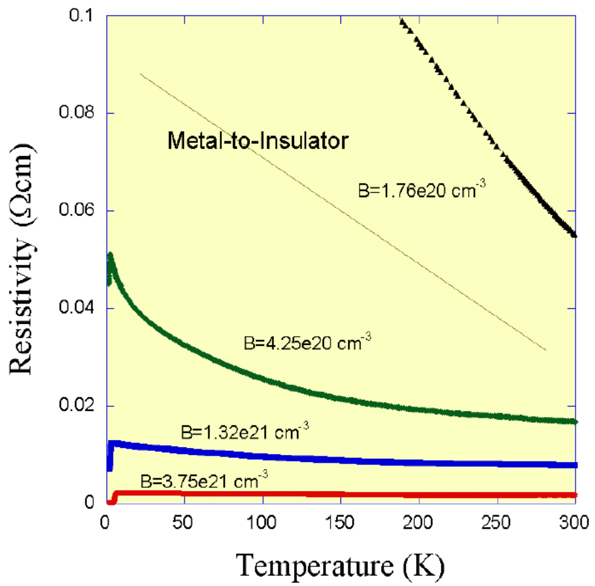


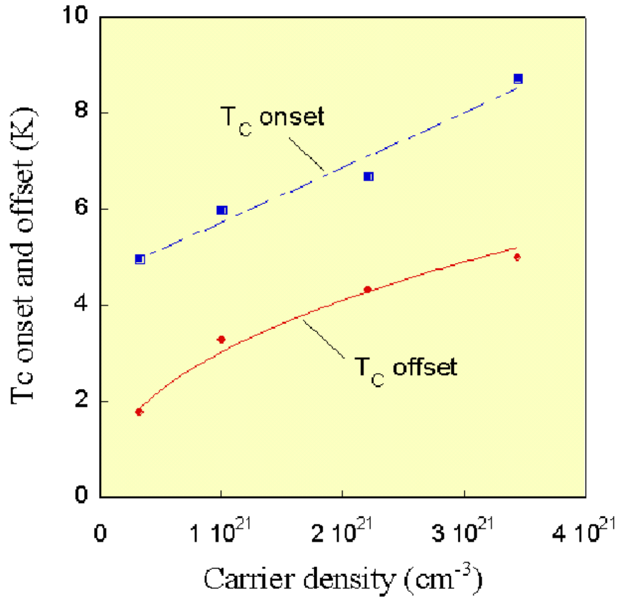
Normalized CSR intensity at F3 as a function of the azimuth angle of the analyzer.

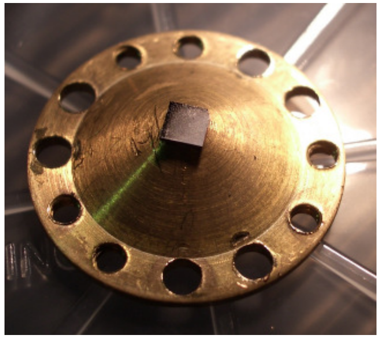
Application of the CSR:

- Superconductors
- THz Near-field Spectroscopy

- Recently discovered superconductor: E.A. Ekimov, Nature **428**, 542 (Nov. 2004).
- Superconductivity appears at high B-doping beyond the Metal-to-Insulator transition.
- T_c increases to 8 K with increasing Boron concentration.





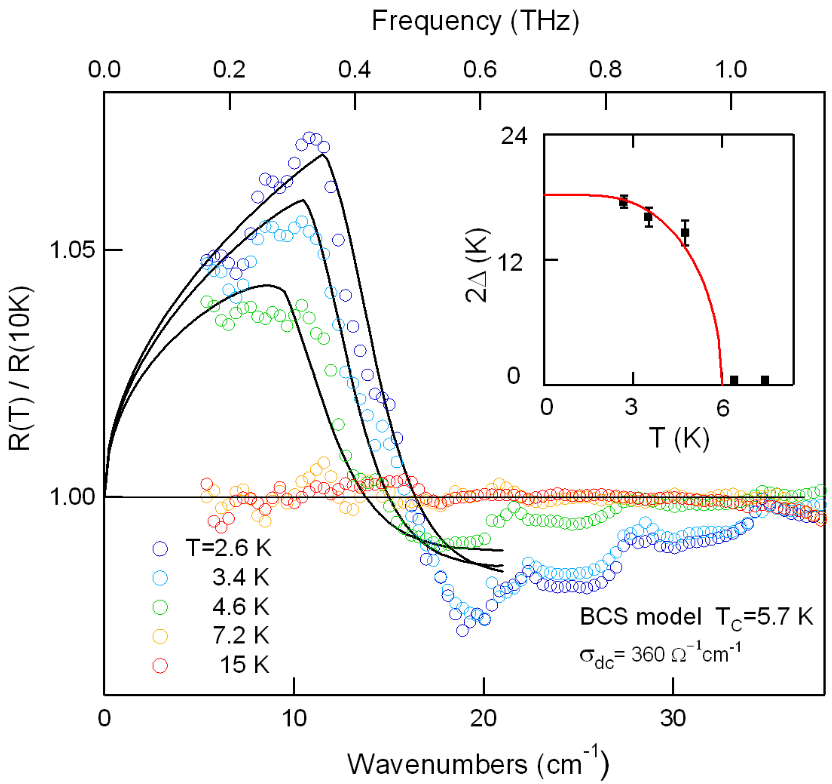


Size: 3 mm x 3 mm

Y. Takano et al., Diamond & Related Mat. **14**, 1936 (2005) and Nature **438**, 647 (2005).

Boron–doped Diamond

La Sapienza
Università degli Studi di Roma
NIMS
BESSY



Increase of the normal-incidence reflectivity below T_c for $\omega < 2\Delta$ (total screening) observed.

The peak in the R_S/R_N ratio indicates the energy of the optical gap.

As a result of the BCS theory for weak electron-phonon coupling:

→ $2\Delta_0 = 3.53\ T_c$

Our sample: $\omega = 2\Delta = 12\ cm^{-1} = 17\ K$

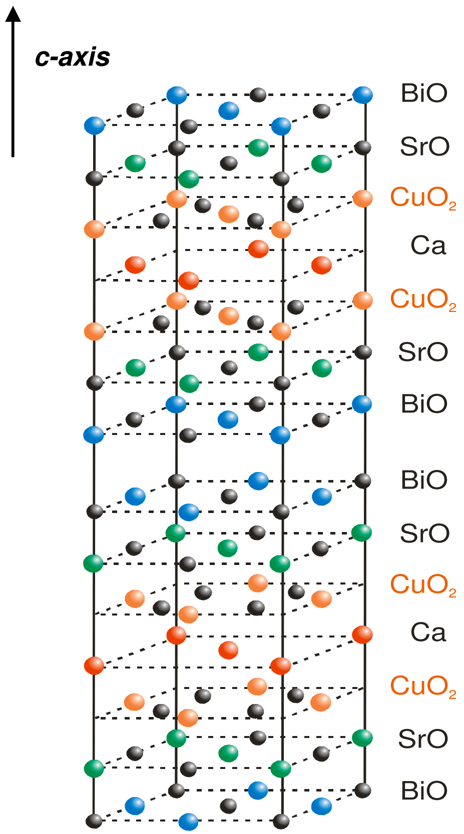
→ $T_c = 5\ K$

M. Ortolani *et al.*, Phys. Rev. Lett. **97**, 097002 (2006).

High–T_C “Cuprate” Superconductor

Project proposed by M. Martin et al.

UCSD · Berkeley Lab · BESSY



c-axis reflectance of optimally doped BSCCO 2212

- structural anisotropy
- high T_c (90 K) but low “gap energy”

$T>T_C$

- Charge transport is blocked by insulating layers.
- Behaves like an insulator with R <1.

$T<T_C$

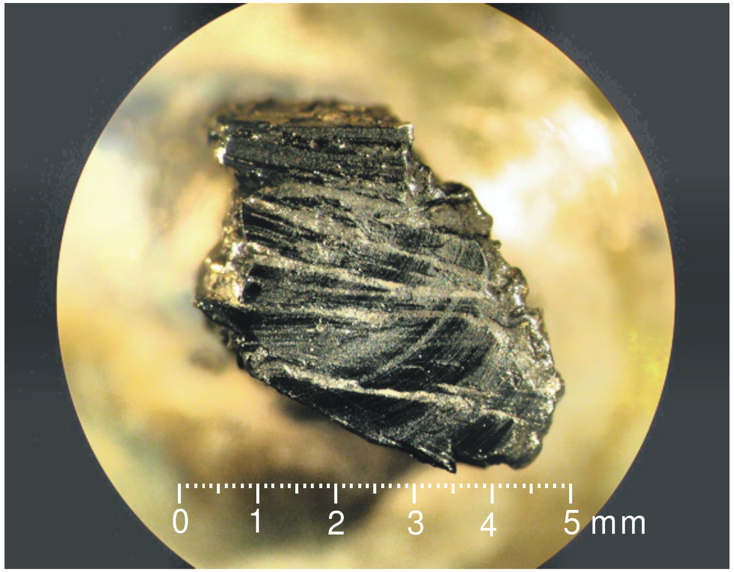
- Cooper pairs tunnel through insulating layers, R ~ 1.
- Josephson Plasma Resonance (JPR) below 10 cm^{-1}

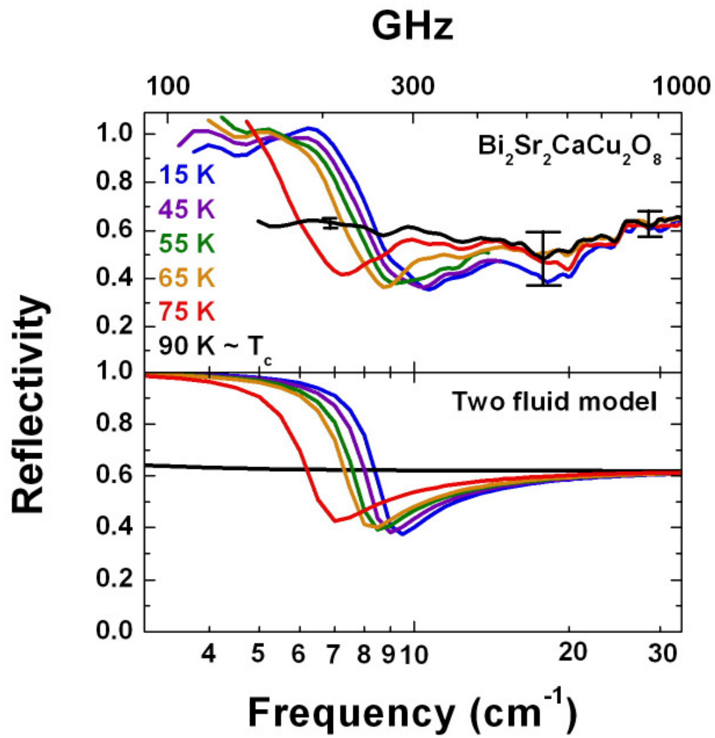
$$\omega_{JPR}^2 = \frac{4\pi n e^2}{m^*}$$

$Bi_2Sr_2CaCu_2O_8$: - extreme structural anisotropy

- highly insulating

Optimally doped $Bi_2Sr_2CaCu_2O_8$



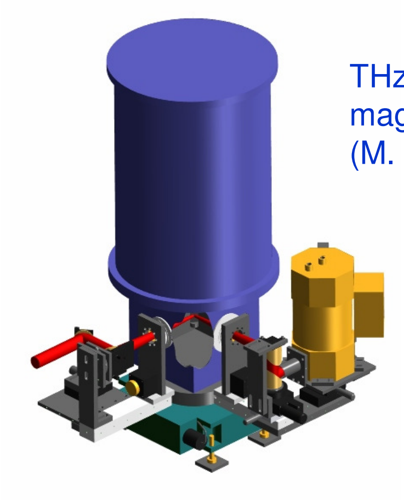


- First scientific experiment using coherent synchrotron radiation as a spectroscopic source.
- Absolute measurements of reflectivity with high photometric accuracy at low temperatures.
- Direct measurement of JPR in optimally doped $Bi_2Sr_2CaCu_2O_8$.
- Bridge between microwave magnetoabsorption and conventional far-IR spectroscopy.

E. J. Singley et al., Phys. Rev. B. 69, 092512 (2004).

THz Problems Near-field Approaches Could Solve

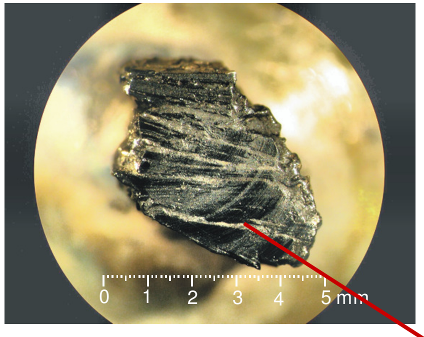
Small-Throughput Experiments in the THz, too!
- complicated optical path (cryostat, magnets, etc.)
- large F#



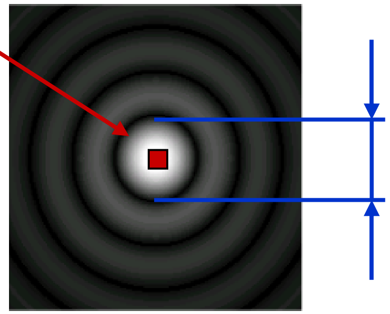
THz ellipsometer for magneto-optic investigations (M. Schubert, U. of Leipzig)

Small Sample Geometry
- new and rare materials
- spatial resolution

$Bi_2Sr_2CaCu_2O_8$



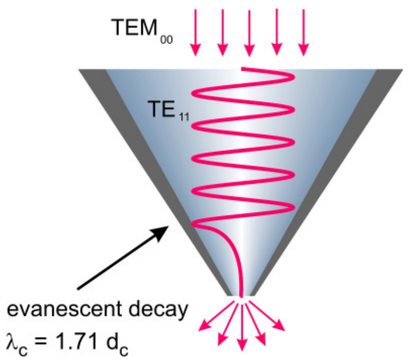
Large THz Focal Spot
- Frauenhofer diffraction (1. disk: 84 % intensity)

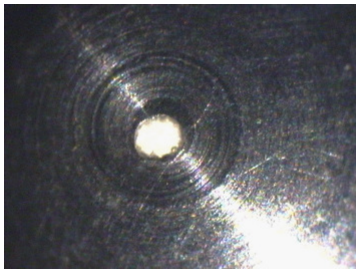


D = 10 mm
($F/4$, 10 cm^{-1})

D = 25 mm
($F/4$, 4 cm^{-1})

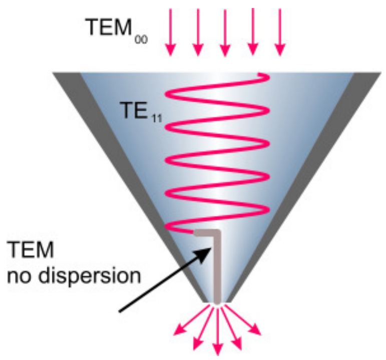
Aperture Cone



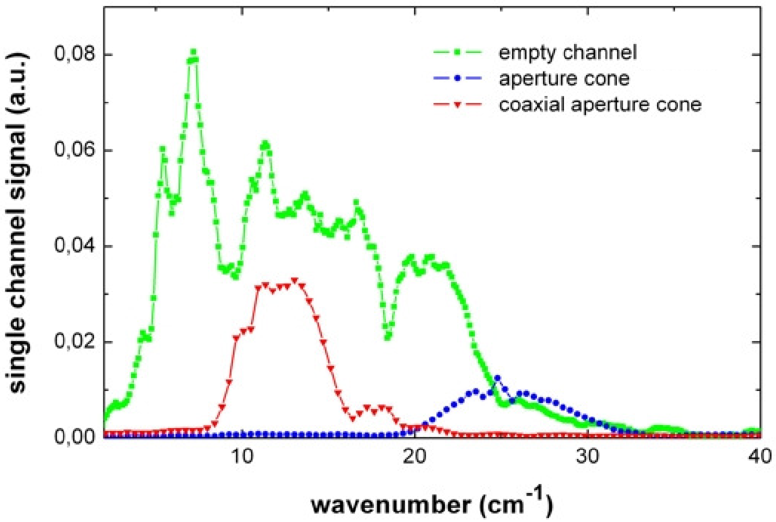


200 µm diameter aperture

Coaxial Aperture cone



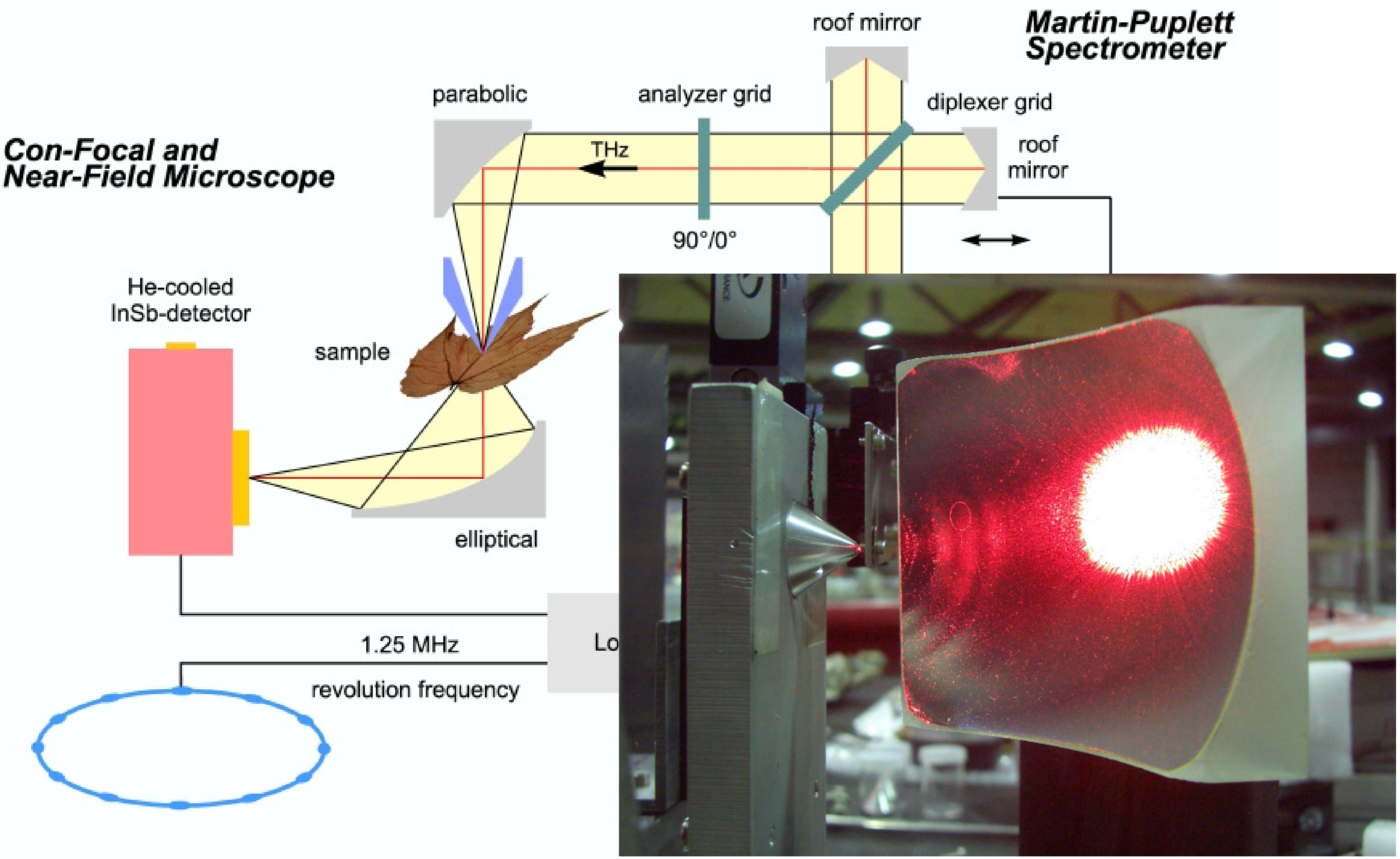
200 µm diameter aperture, 80 µm wire diameter



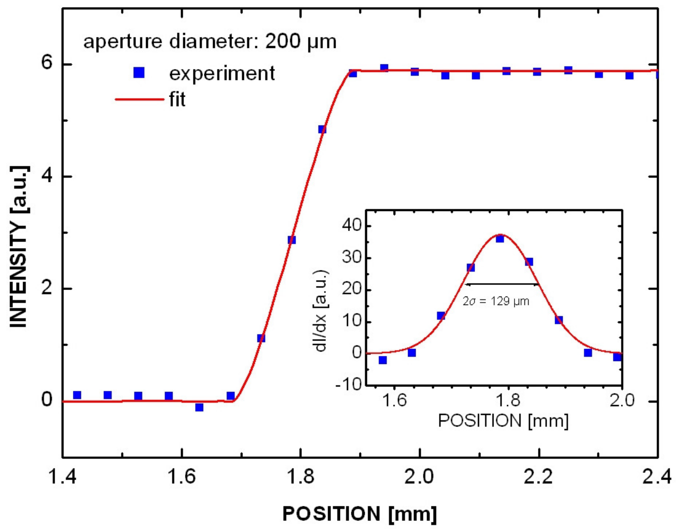
Spectra of the empty spectrometer (to be multiplied by 100), of the aperture cone and of the coaxial aperture cone.

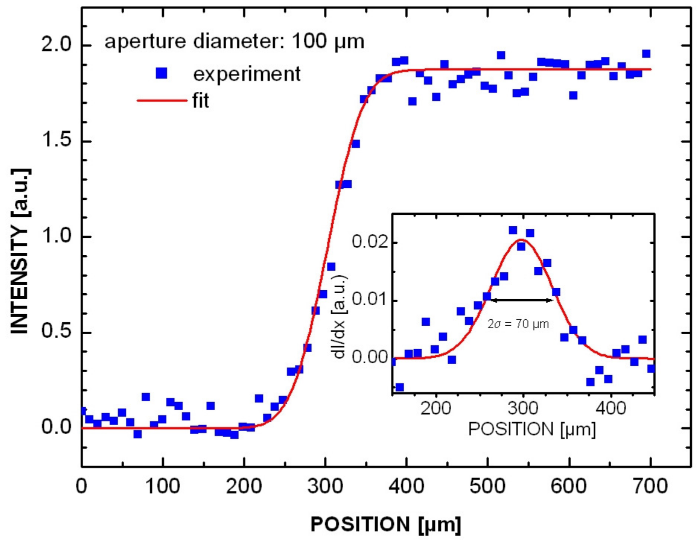
probe design according to: F. Keilmann, *Infrared Phys. & Technol.* **36** 217 (1995).

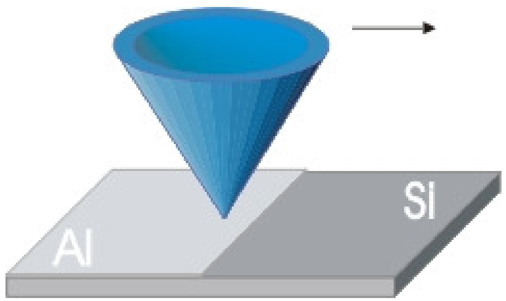
THz Near-field Imaging with CSR



U. Schade et al., *APL* **84** 1422 (2004)



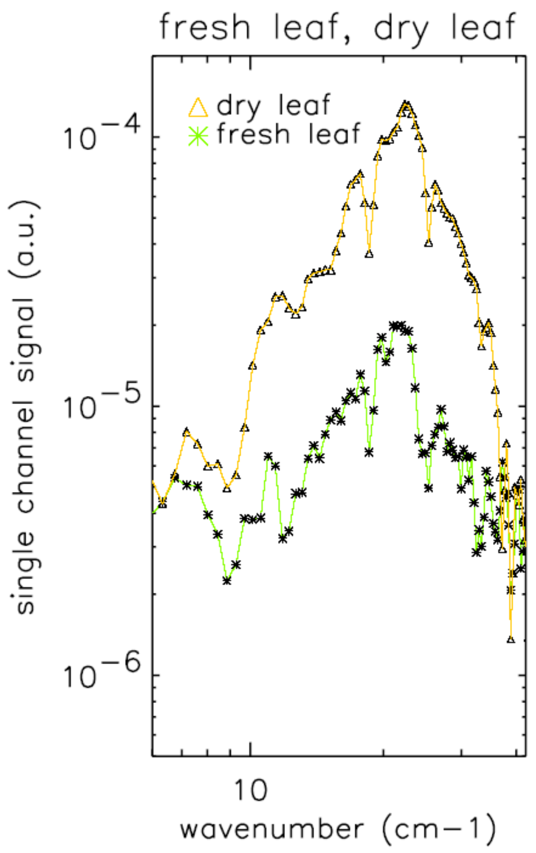




- "knife edge test" on Al-film on Si-substrate
- spatial resolution @ 1 mm wavelength (0.33 THz):

100 µm aperture:	70 µm ≈ 1/14 λ
200 µm aperture:	130 µm ≈ 1/8 λ
(@ 5 mm wavelength (0,066 THz):	≈ 1/38 λ)

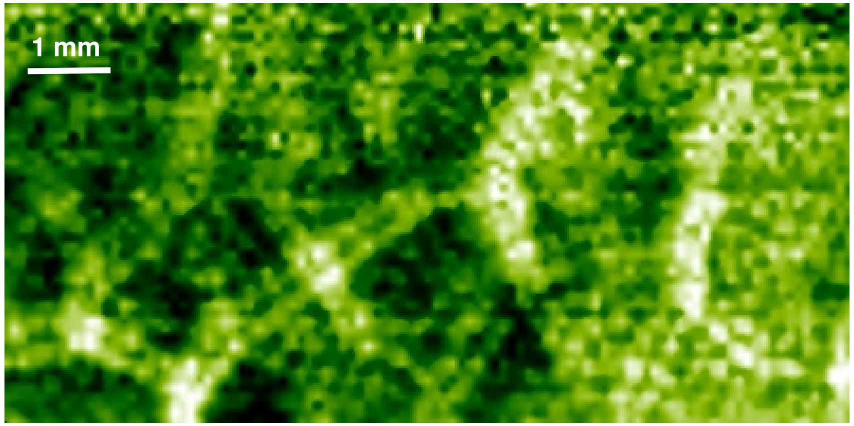
THz Near-field Imaging with CSR



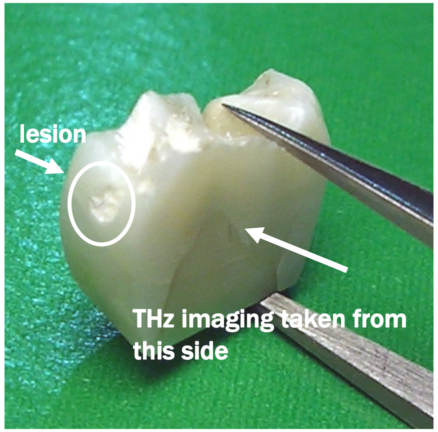
Single channel transmittance spectra of a fresh cut leaf and the same leaf but dried.

VIS- image

Near-field image



- VIS and THz near-field image of a laurel leaf
- spatial resolution: 130 µm
- spectral weight @ 0.5 mm wavelength



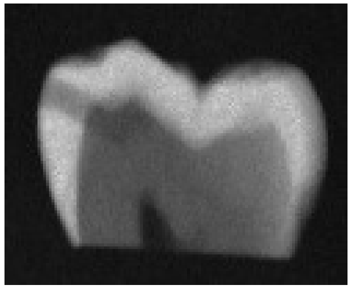
Tooth decay diagnostics:

X-ray: little material contrast due to demineralization

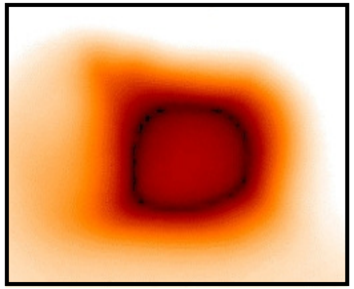
NIR: good for enamel but dentin almost opaque

THz: ?

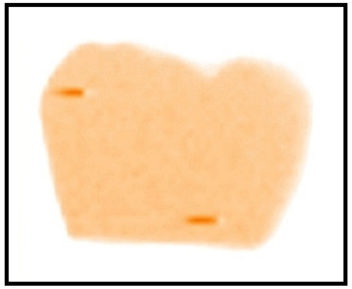
Simulated caries lesion (tooth decay) composed of hydroxyapatite powder.



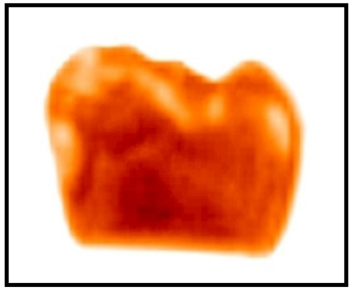
- shadow image
- x-ray



- far-field @ 1 mm (0.3 THz)
- con-focal geometry
- bursting mode



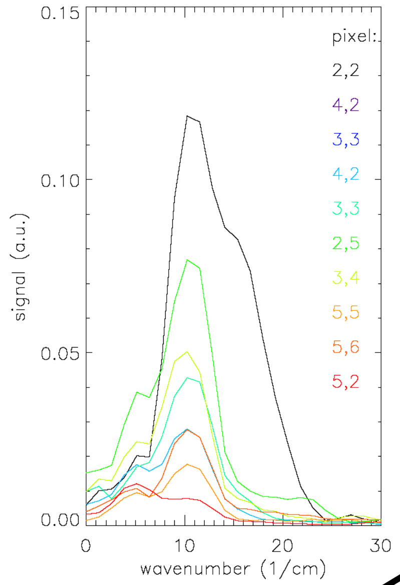
- near-field @ 1 mm
- 200 µm aperture
- bursting mode

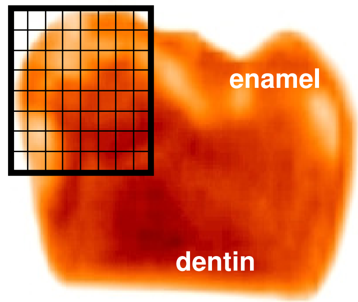


- near-field @ 1 mm
- 200 µm wire cone
- low alpha mode

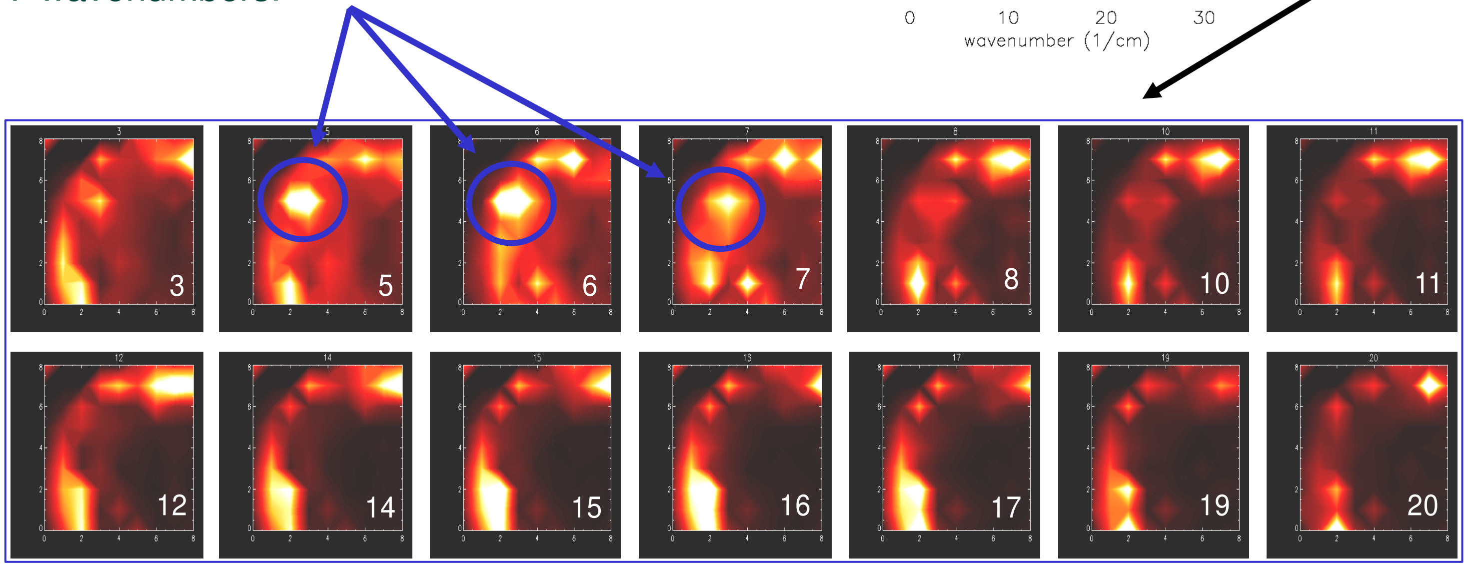
Spectral near-field images of the lesion region between 3 and 20 cm^{-1} (between 0.5 and 3 mm wavelength).

The corresponding wavenumber is indicated on top of each frame. Note that the simulated caries lesion is indicated by a lower absorption between 5 and 7 wavenumbers.





Integral THz Image



U. Schade et al., *Proc. SPIE Vol.* **5725** 46 (2005).

Coherent Synchrotron Radiation

Low-noise, broadband, steady-state, high power, diffraction limited, polarized, pulsed (fs)

New science opportunities

- superconducting gap
- hybridization energy in heavy fermion systems
- intra-molecular vibrations
- phonons, plasmons, cyclotron resonances …
- electron energy levels in confined systems
- …

Triggering new technologies

- THz near-field optics
- THz Martin-Puplett-ellipsometer
- remote sensing for homeland security applications
- fs-slicing diagnostics
- …

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