Infrared and Terahertz Synchrotron Radiation

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$100\ \mu m = 100\ cm^{-1} = 12\ meV$
$= 3\ THz = 144\ K$

Outline

- Introduction to Infrared/terahertz spectroscopy and microscopy
  - Experimental methods and sources
  - Character of IR synchrotron radiation
  - Introduction of UVSOR-II
- Examples
  - IR micro-spectroscopy and imaging of correlated materials
    - Spatial imaging of metal-insulator transition of organic conductors under pressure
- Future IR/THz light sources
  - Coherent synchrotron radiation
- Summary

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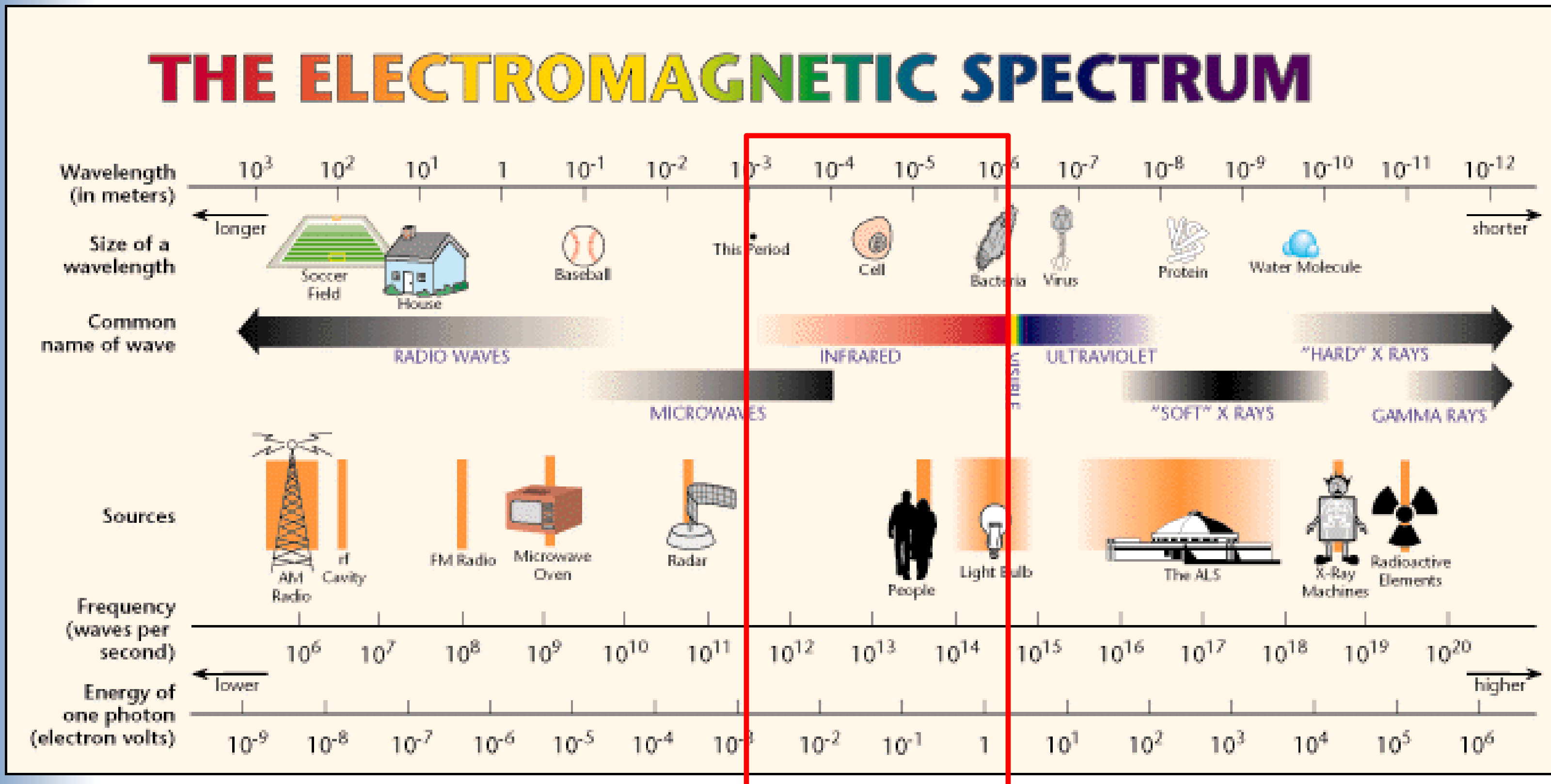
What are IR and THz?



IR/THz covers this region.

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

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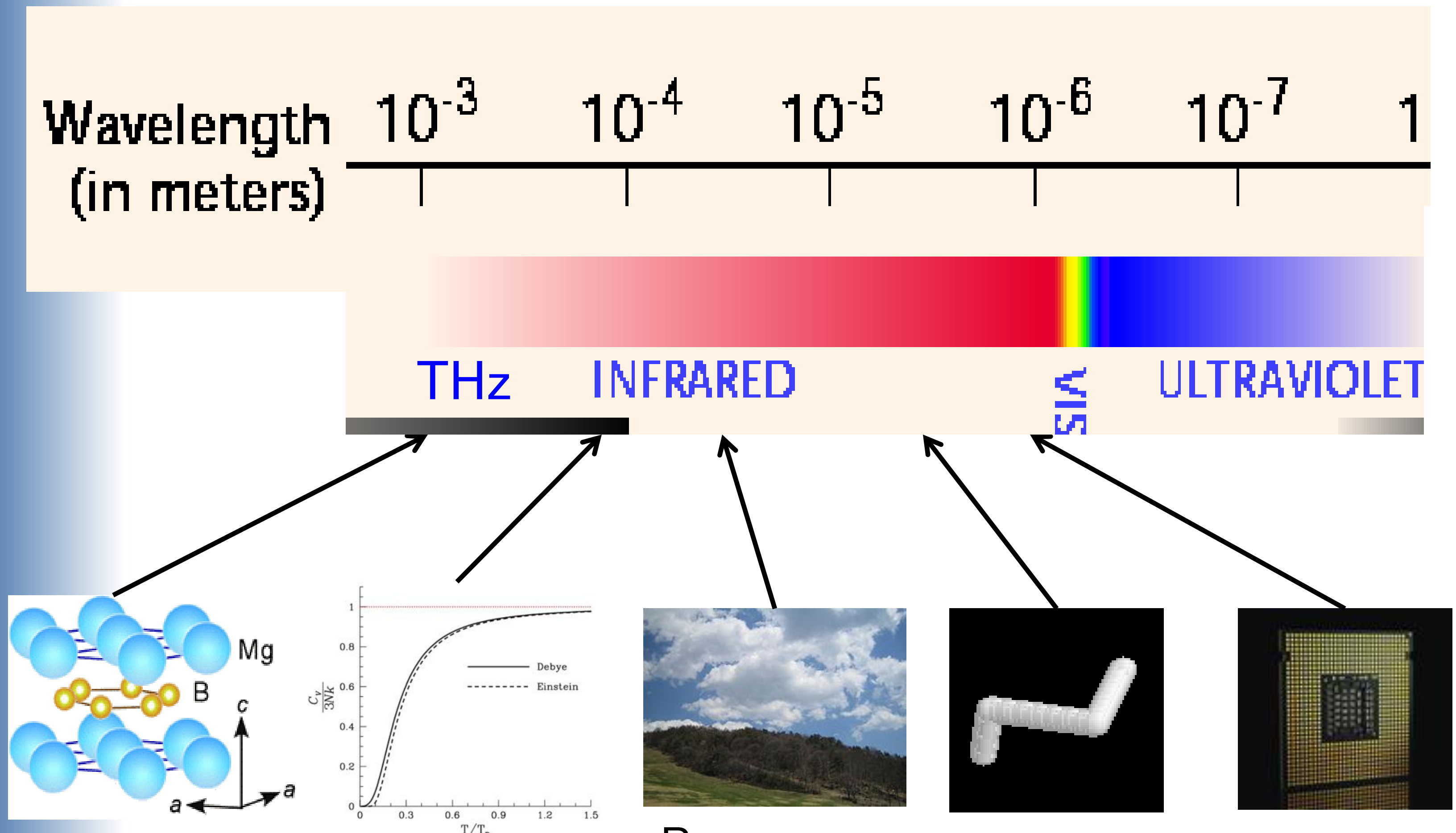
What can we see in IR+THz?



Superconducting gap
Quasiparticles

Debye
temperature

Room
temperature

Molecular
vibration

Energy gap of Si

··· So many elementary excitations

Fourier transform IR spectroscopy (Michelson interferometer)

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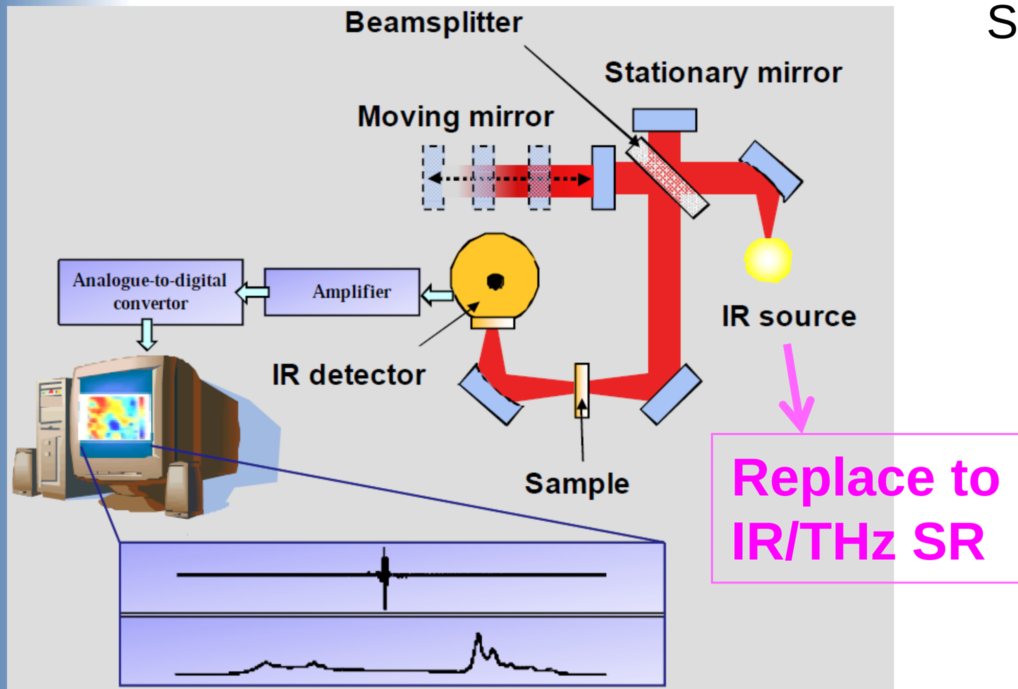
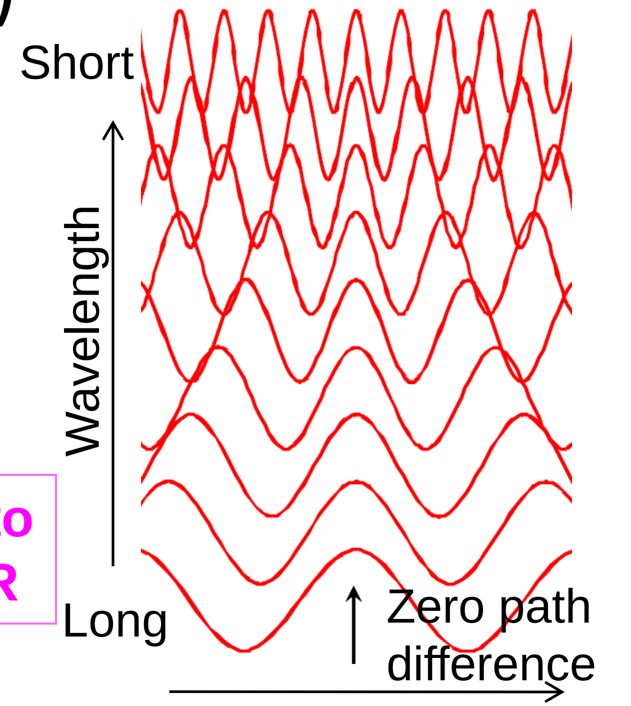
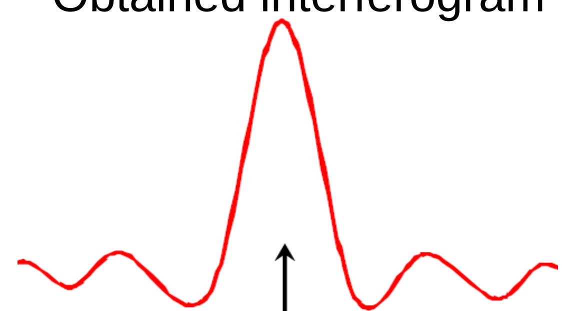


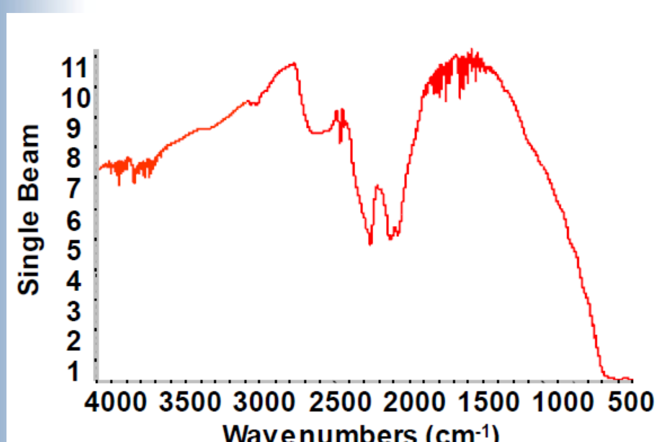


Summation

Obtained interferogram

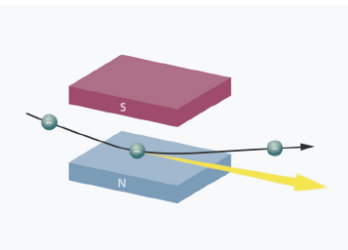


Fourier transform



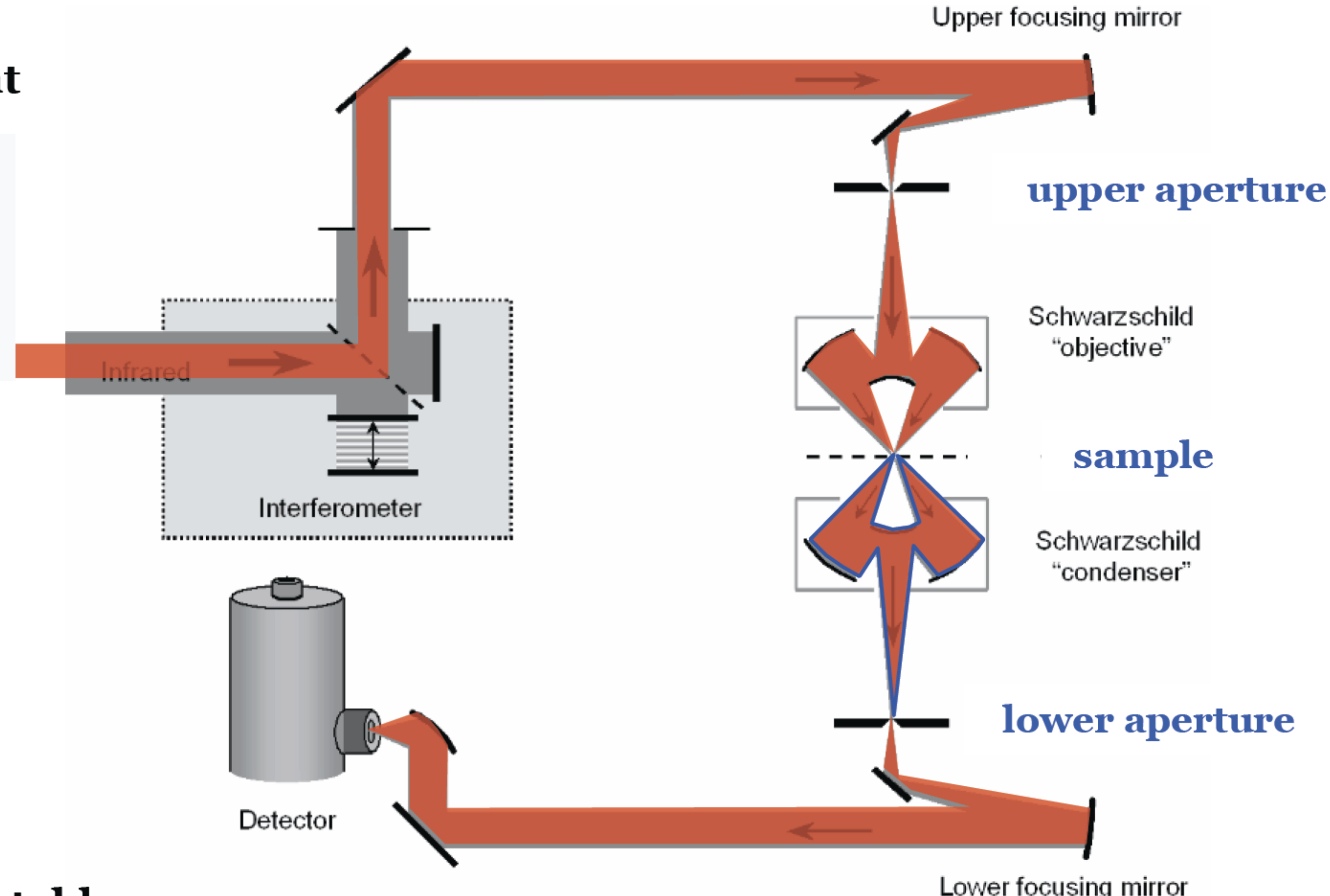
Infrared **microscopy** instrumentation

SR: very bright



or

thermal: very stable

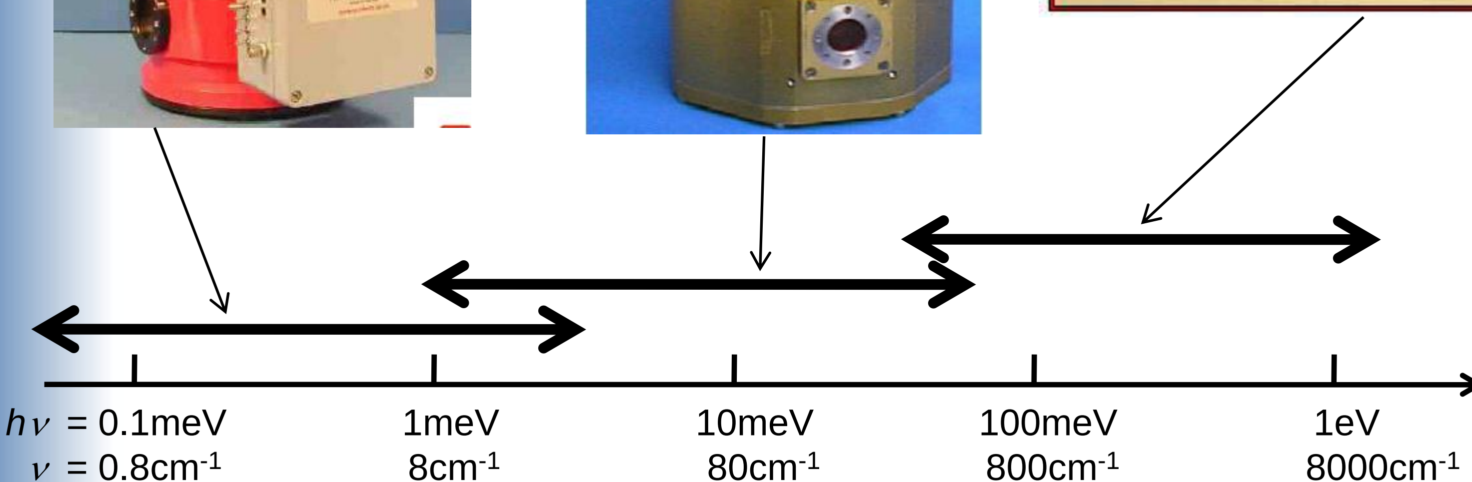


IR + THz detectors

LHe-cooled InSb hot-electron bolometer

LHe-cooled Si bolometer

LN_2-cooled HgCdTe (MCT)



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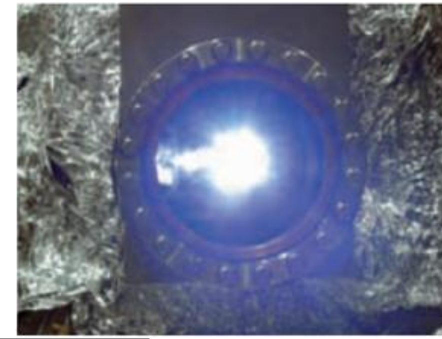
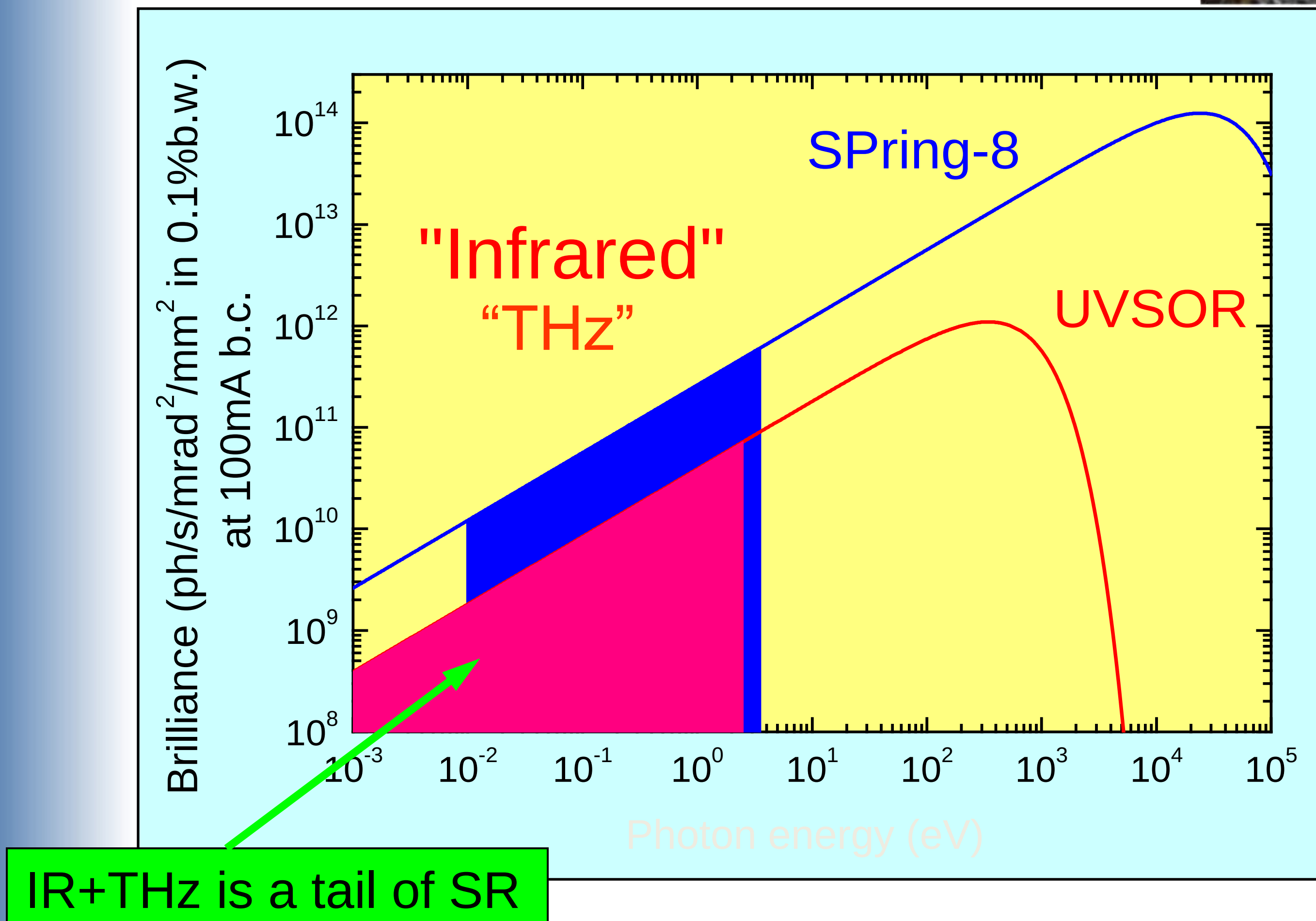
What's infrared/terahertz synchrotron radiation?





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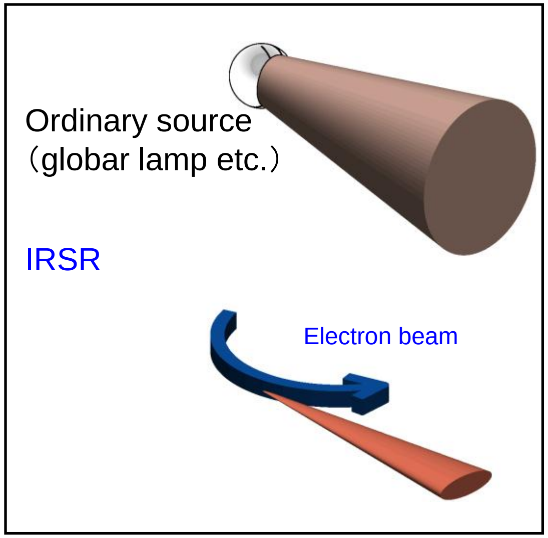
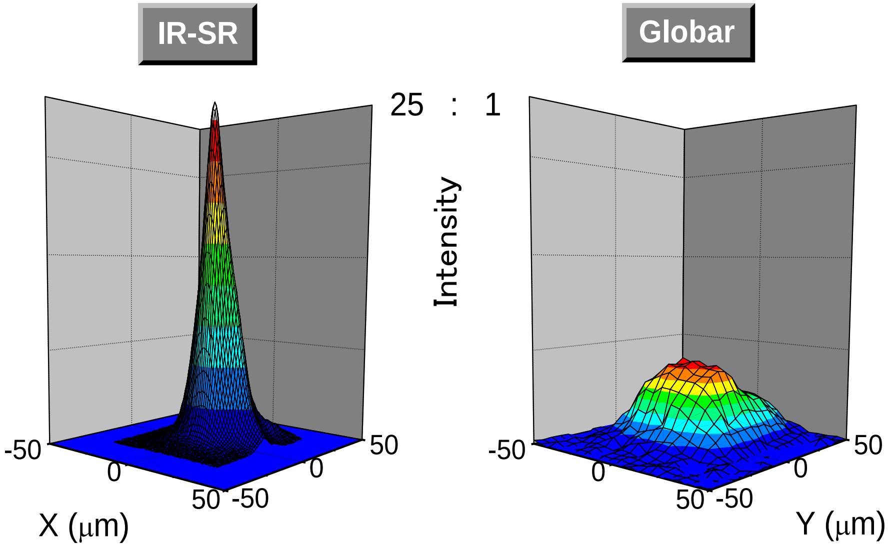
Why we use IR/THz SR?

What's the advantage compared with ordinary sources?

- High brilliance
  - Excellent for spectromicroscopy
- Broadband
  - Useful for spectroscopy
- Linear/circular polarization
  - Crystallic asymmetry, Molecular orbital, polar direction, MCD
- Pulse (sub-pico-second)
  - Time structure



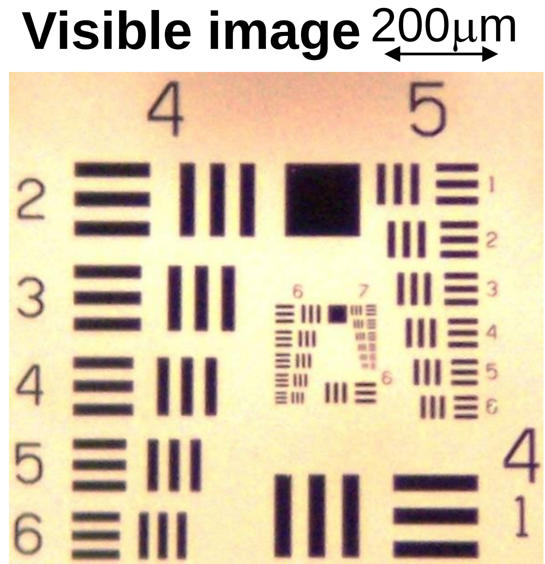


Spatial resolution test using IR-SR @ BL6B, UVSOR-II

(Ge/KBr + MCT, 500-8000cm^{-1})

IR-SR

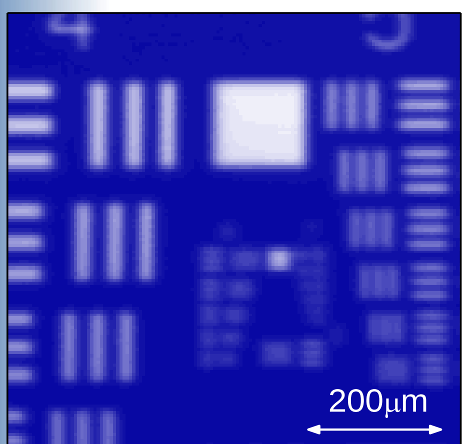
Visible image



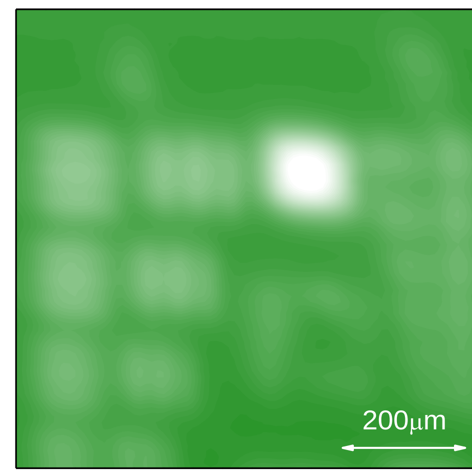
USAF test target

Globar lamp

(laboratorial source)



$\Delta r \sim 12\ \mu m$



$\Delta r \sim 50\ \mu m$

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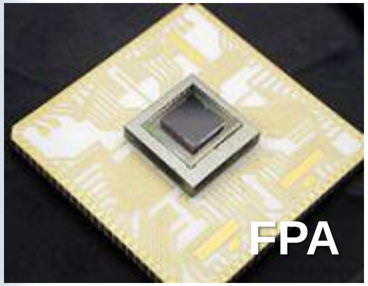
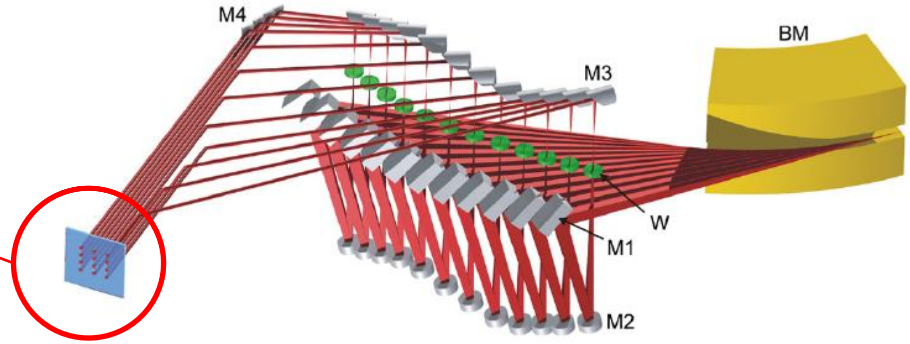
IR-SR + Focal Plane Array Detector

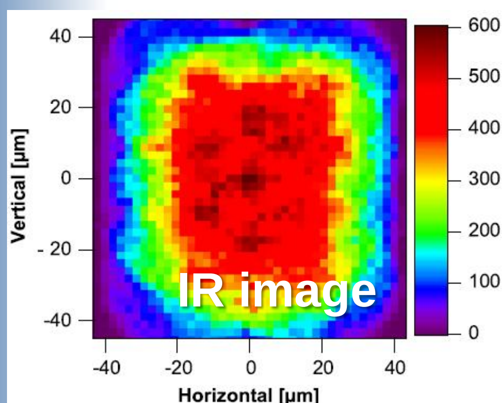
FPA: 2-dimensional MCT

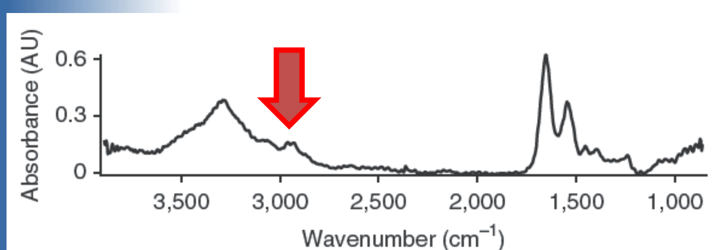


IRENI beamline at SRC, University of Wisconsin

[M. Nasse et al., *Nucl. Instrum. Meth. A* **582**, 107 (2007).]

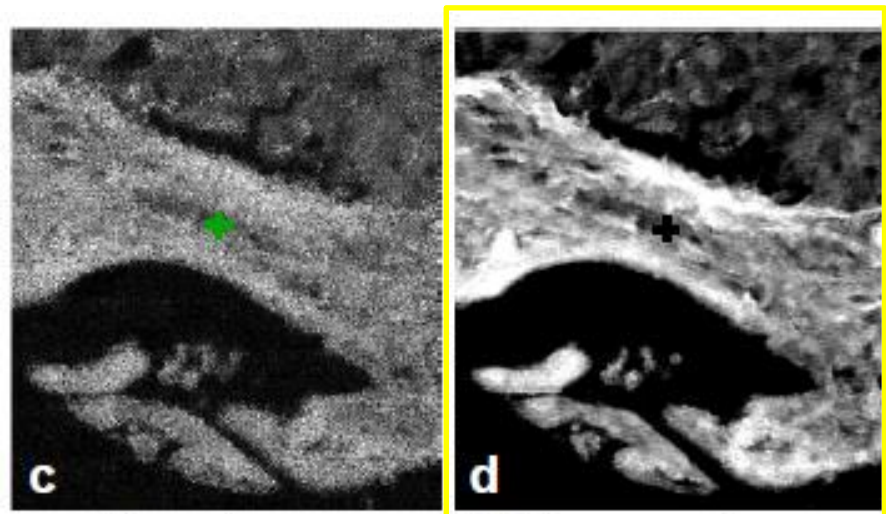






Images of CH stretch regions of living cell

[M. Nasse et al., *Nature Methods* **8**, 413 (2011).]



Thermal source
74× FPA (0.54 μm)

Multi-beam synchr. source
74× FPA (0.54 μm)

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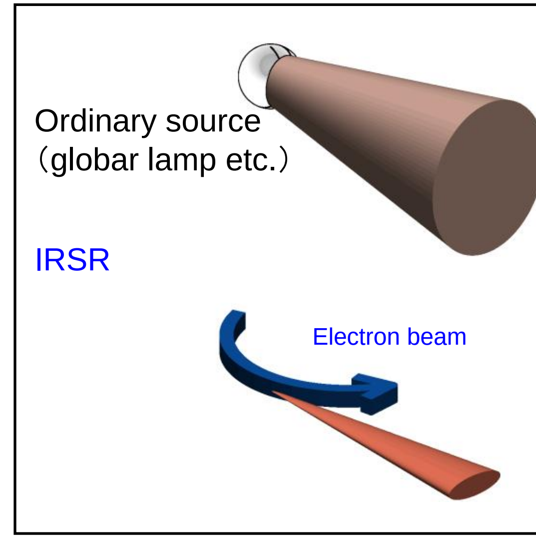

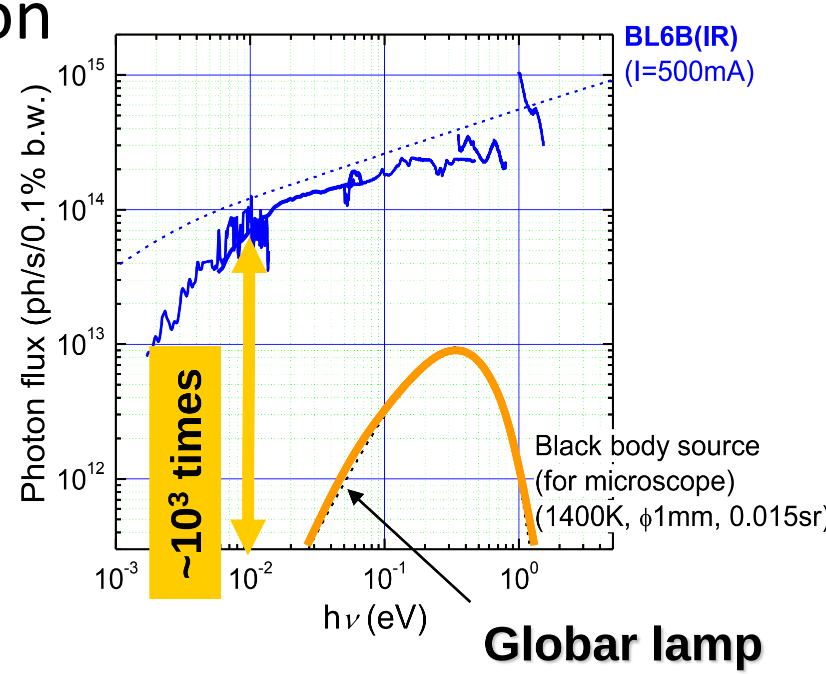

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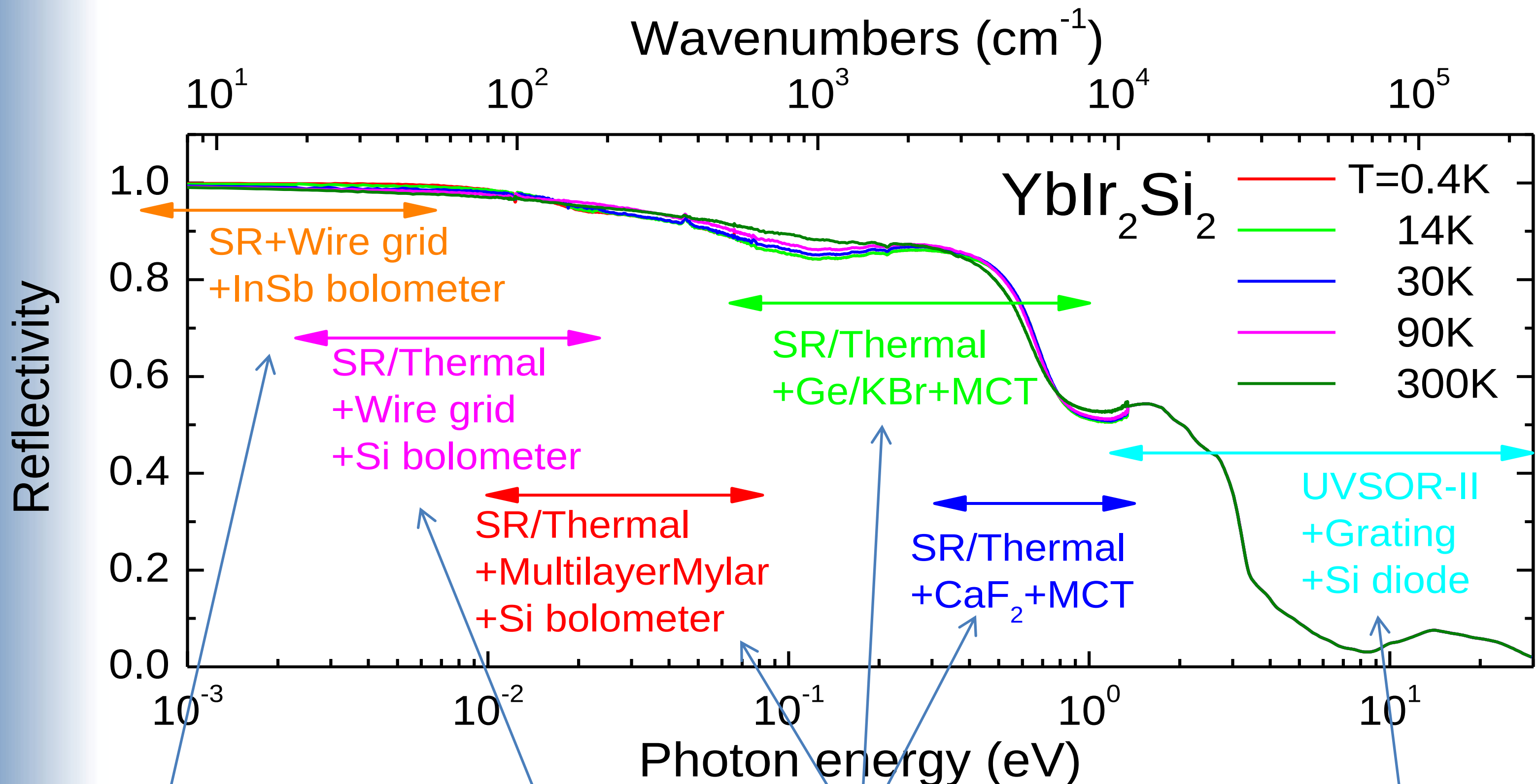
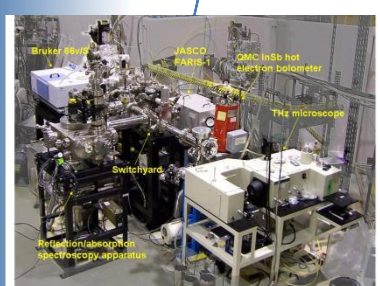
Reflectivity measurements in the very wide energy range of 1 meV to 30 eV





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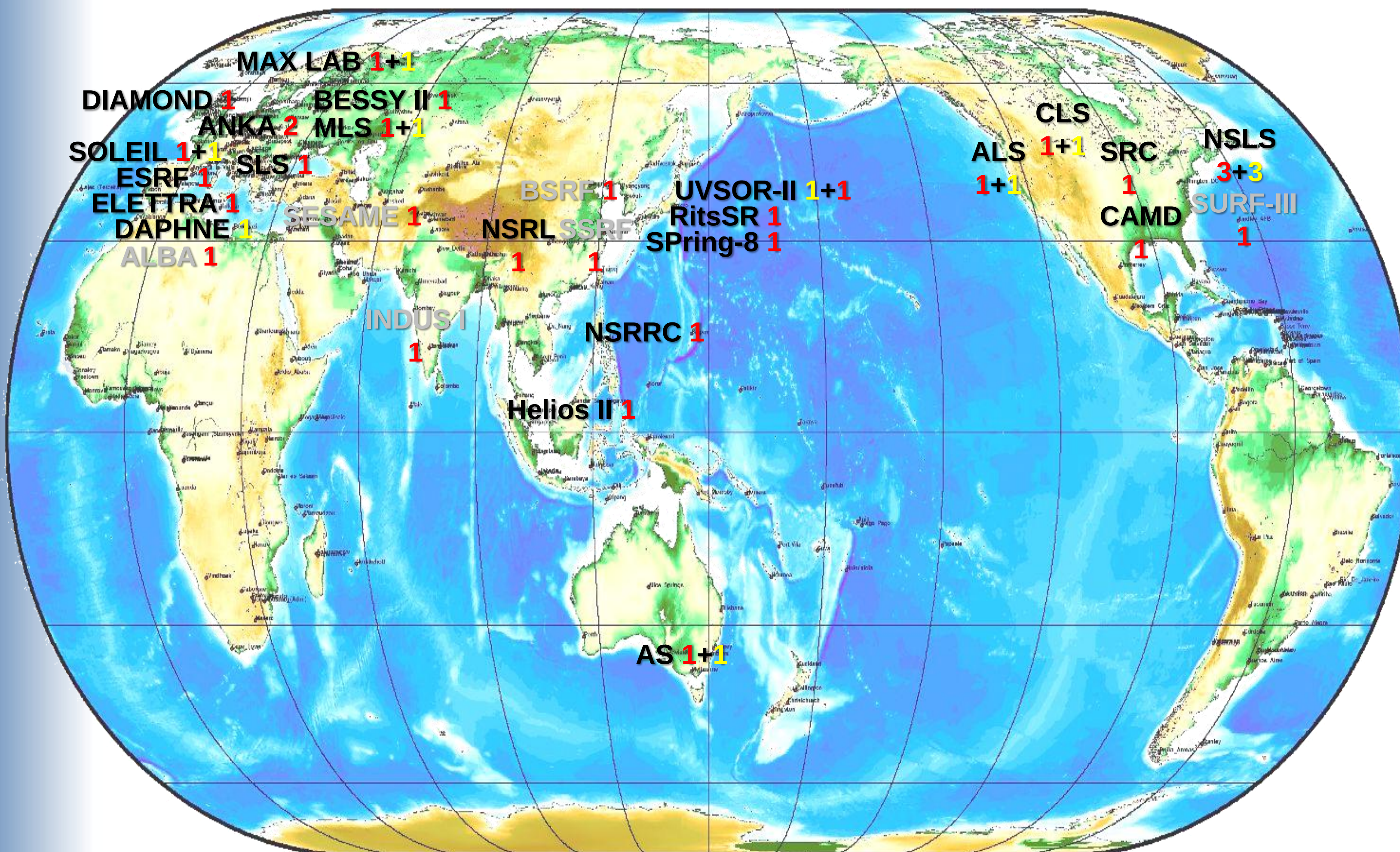
IRSR+ THzSR beamlines in the world



Operational + Commissioning: 23+9

Planned: 6+1

Total: 39 IR+THz beamlines in the world!

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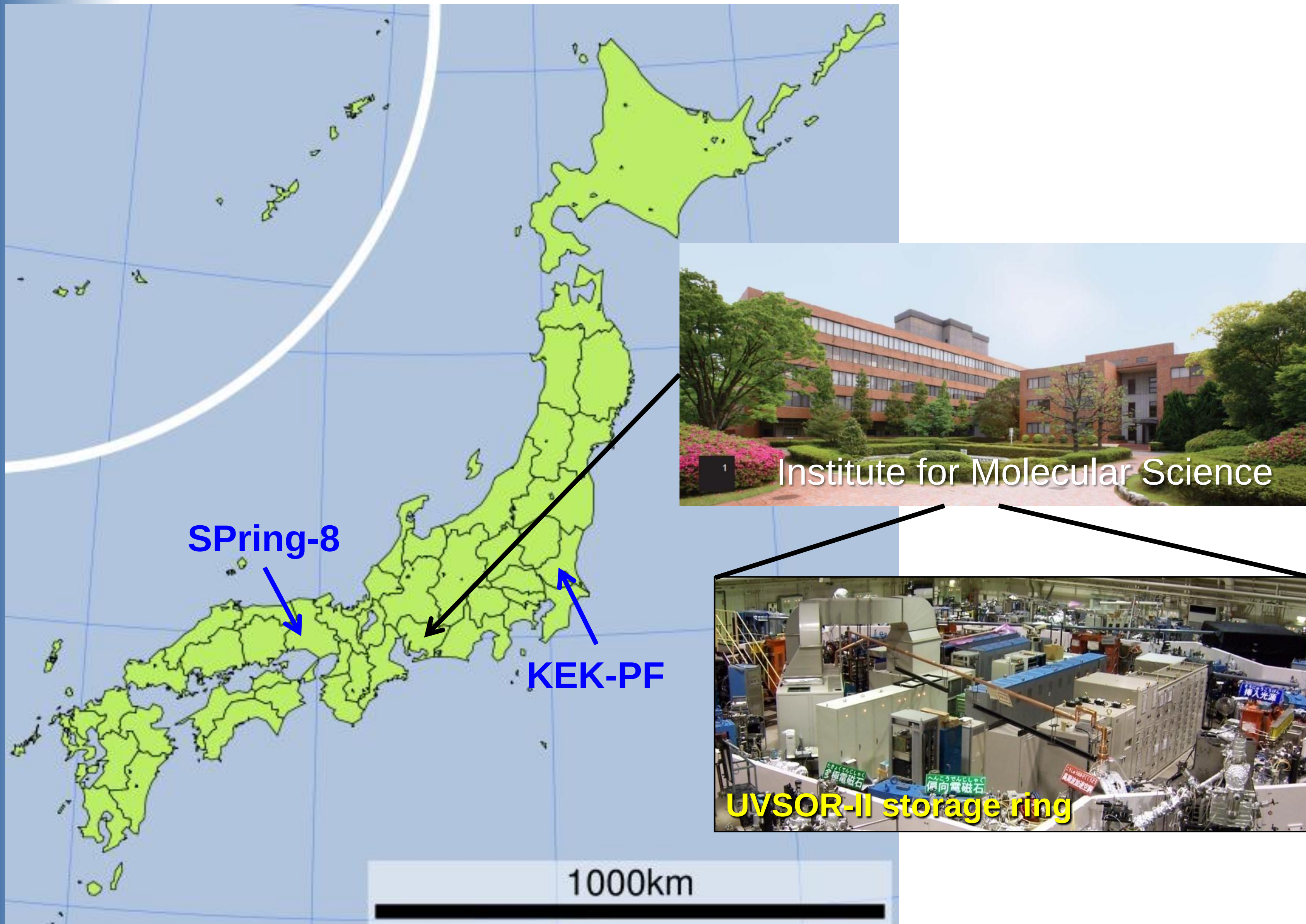
Where is UVSOR?



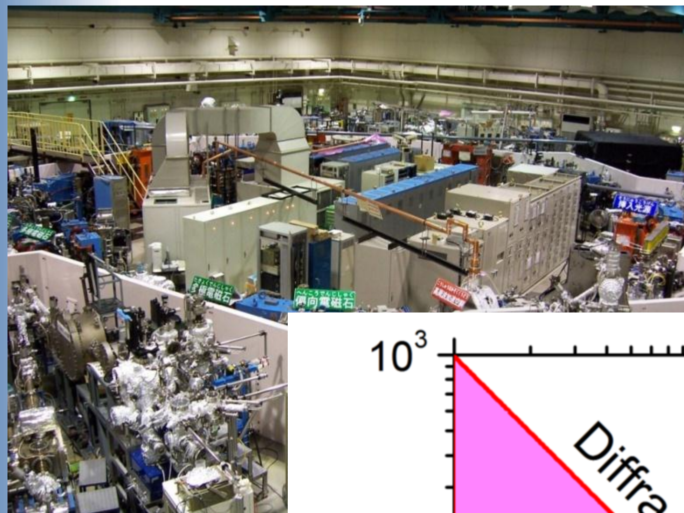
Light source of UVSOR-II

Electron Energy	750 MeV **(for VUV + SX)**
Circumstance	53.2 m **(small size)**
Emittance	27nm-rad
Straight Sections	4mx4+1.5mx4
Filling Beam Current	300 mA (multi-bunch)

Top-up operation is fully started.

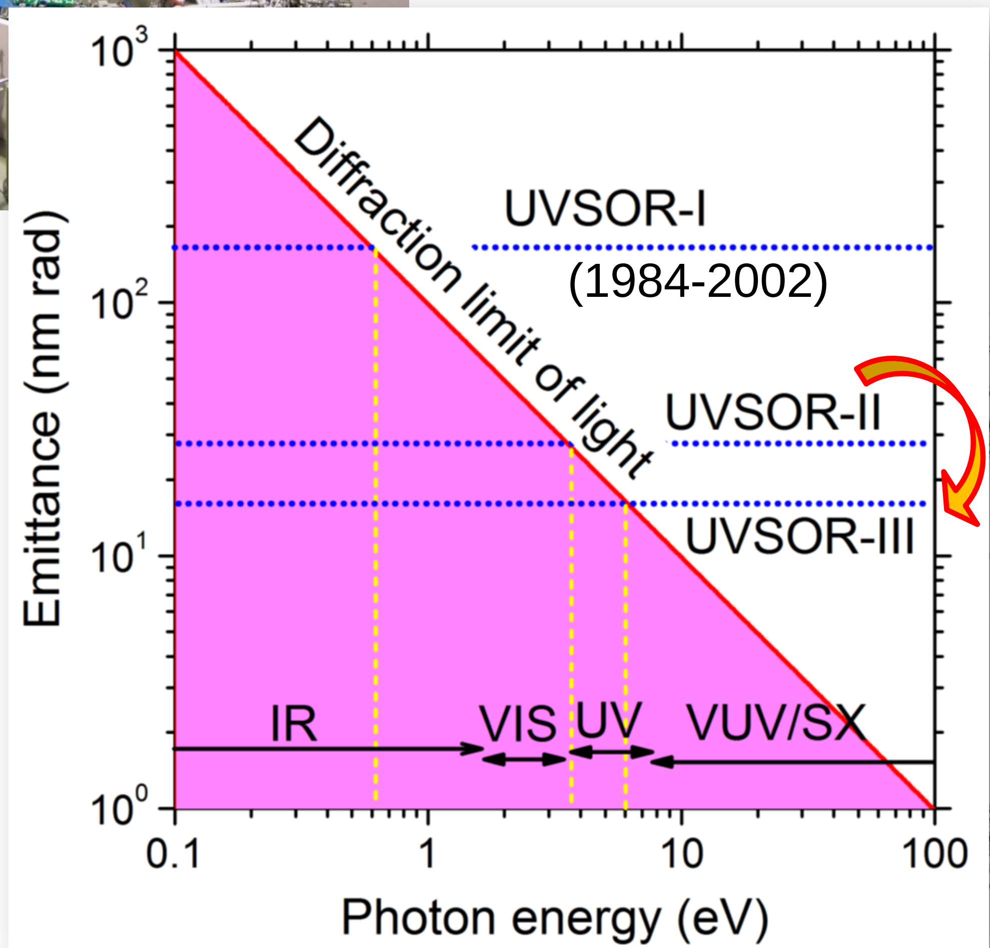
Light source of UVSOR-II

15 nm-rad since 2012



Electron Energy	750 MeV	**(for VUV + SX)**
Circumstance	53.2 m	**(small size)**
Emittance	27nm-rad	since 2003
Straight Sections	4mx4+1.5mx4	
Filling Beam Current	300 mA (multi-bunch)	

Top-up operation is fully started.



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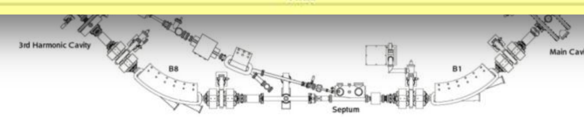
Light source of UVSOR-II

15 nm-rad since 2012

Electron Energy	750 MeV	**(for VUV + SX)**
Circumstance	53.2 m	**(small size)**
Emittance	27nm-rad	since 2003
Straight Sections	4mx4+1.5mx4	
Filling Beam Current	300 mA (multi-bunch)	

Top-up operation is fully started. since 2010



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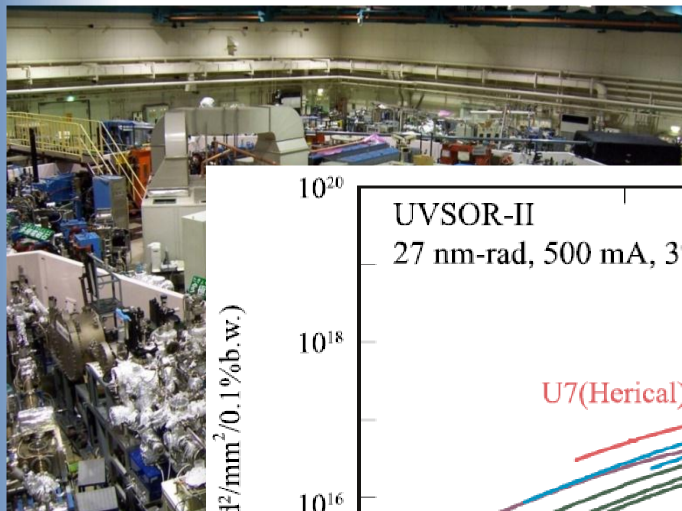
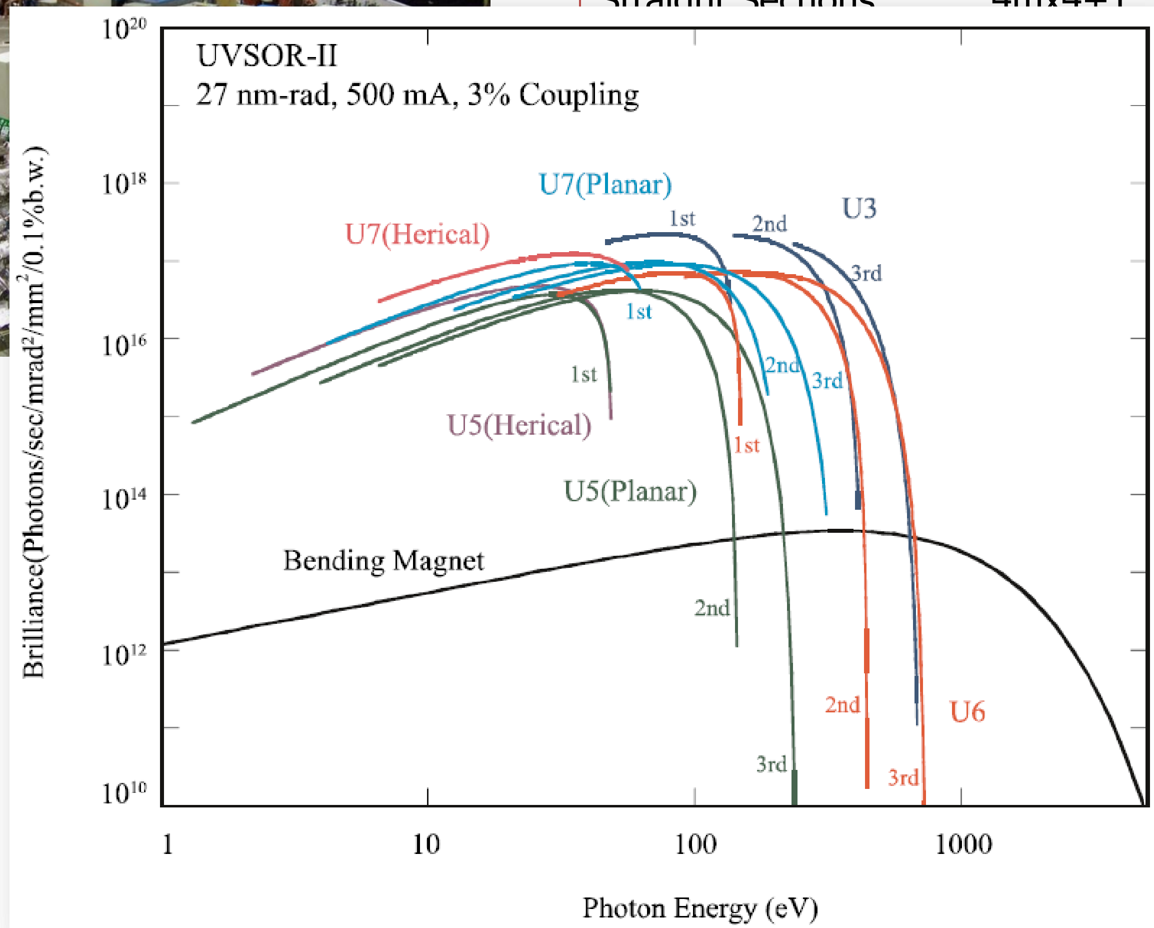
Light source of UVSOR-II

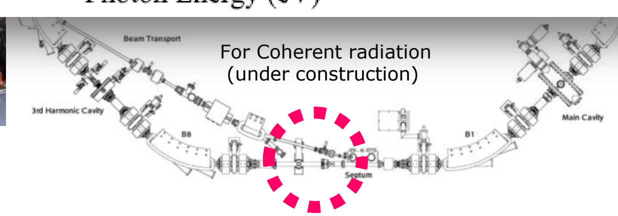


Electron Energy	750 MeV
Circumstance	53.2 m
Emittance	27nm-rad
Straight Sections	4mx4+1.5mx4
	ulti-bunch)
	l.



erved for
ure Undulator

2m In-vacuum Undulator



For Coherent radiation (under construction)

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14 Beamlines at UVSOR-II (Nov. 14, 2011)

ビームライン	分光器型式	エネルギー領域	実験
1U	自由電子レーザー・コヒーレント高調波		光源開発
1B	フーリエ変換テラヘルツ分光器	0.5meV 30meV	固体（反射・吸収）
2A	二結晶分光器	400eV 4keV	固体（吸収）
2B	18-m ドラゴン型分光器	24eV 205eV	気体（光イオン化，光脱離）
3U	不等刻線間隔平面回折格子分光器	60eV 800eV	気体・液体・固体 （光電子分光，発光）
3B	2.5-m off-plane Eagle型直入射分光器	2eV 30eV	固体（反射・吸収・発光）
4B	不等刻線間隔平面回折格子分光器	25eV 1keV	固体（吸収・MCD）
5U	SGM-TRAIN型分光器	5eV 200eV	固体（光電子分光）
5B	平面回折格子分光器	5eV 600eV	機器校正 固体（吸収）
6U	可変偏角斜入射分光器	30eV 500eV	気体（光イオン化，光脱離） 固体（光電子分光）
6B	フーリエ変換赤外分光器	3meV 1.5eV	固体（反射・吸収）
7U	10-m Wadsworth型直入射分光器	6eV 40eV	固体（光電子分光）
7B	3-m McPherson型直入射分光器	1.2eV 30eV	固体（反射・吸収・発光）
8B	平面回折格子分光器	1.9eV 150eV	固体（光電子分光）

IR+THz BL

Main energy range:
several eV～1 keV。

UVSOR-II light source is suitable for electronic structure investigation.

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IR/THz beamline, BL6B, UVSOR-II

(replaced in 2004, old BL was constructed in 1985)

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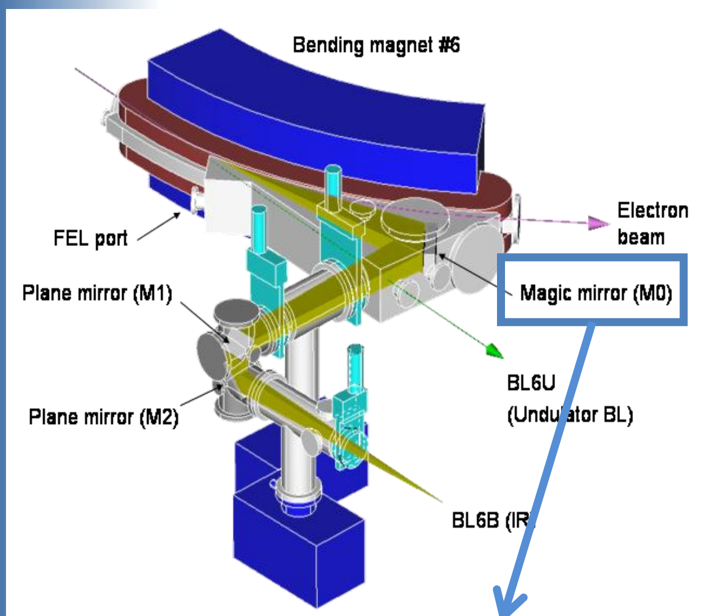
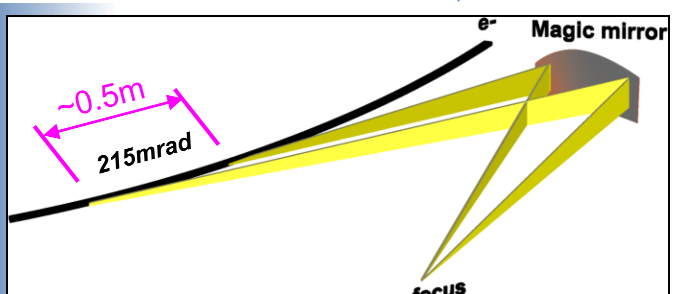
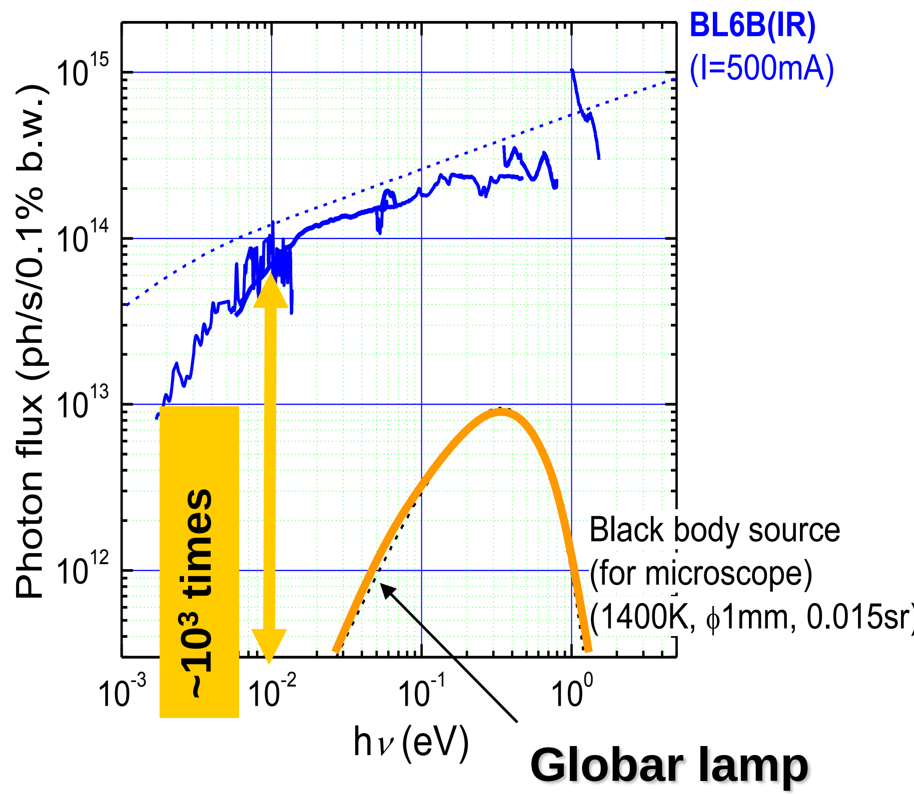
Acceptance angle of SR:

215(H) x 80(V) mrad^2

(largest acceptance angle in the world)







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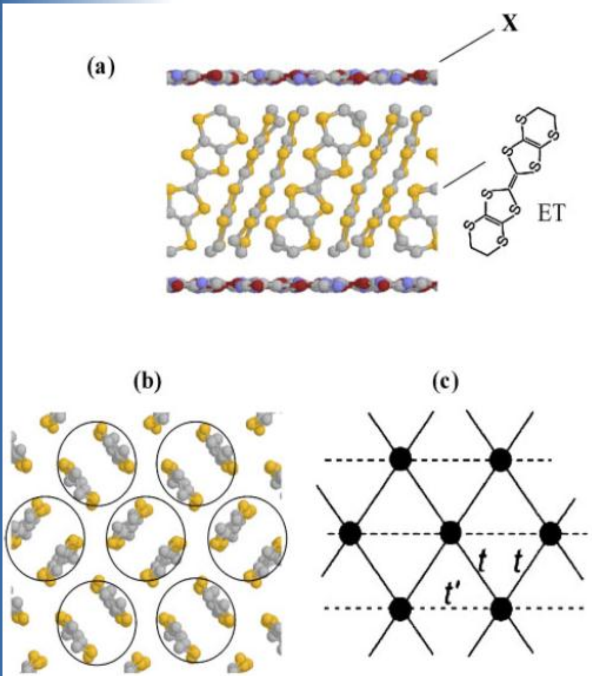
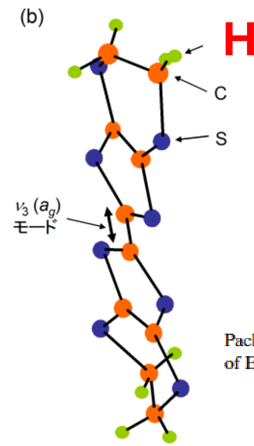
Physical properties of κ-$(ET)_2Cu[N(CN)_2]Br$

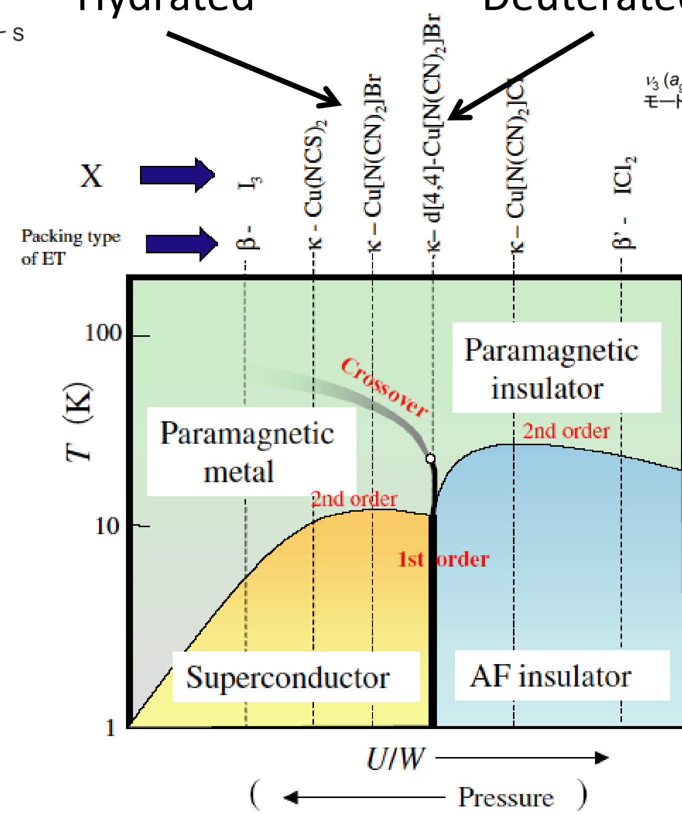
Crystal structure

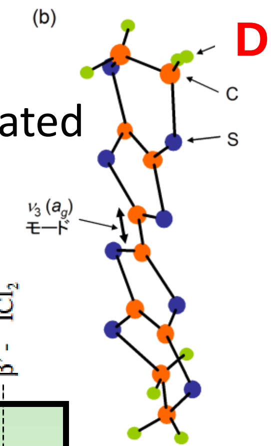




Hydrated

Deuterated





The parameter can be controlled by deuteration.

Carrier (hole) number = 1 / (ET x 2)

Quarter filling of conduction band

Electron Correlation

Mott insulator

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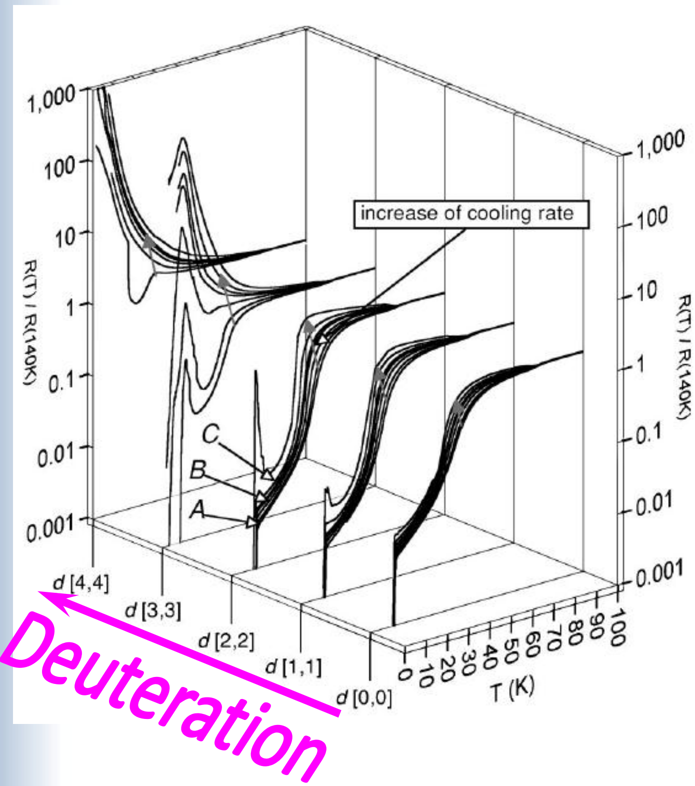
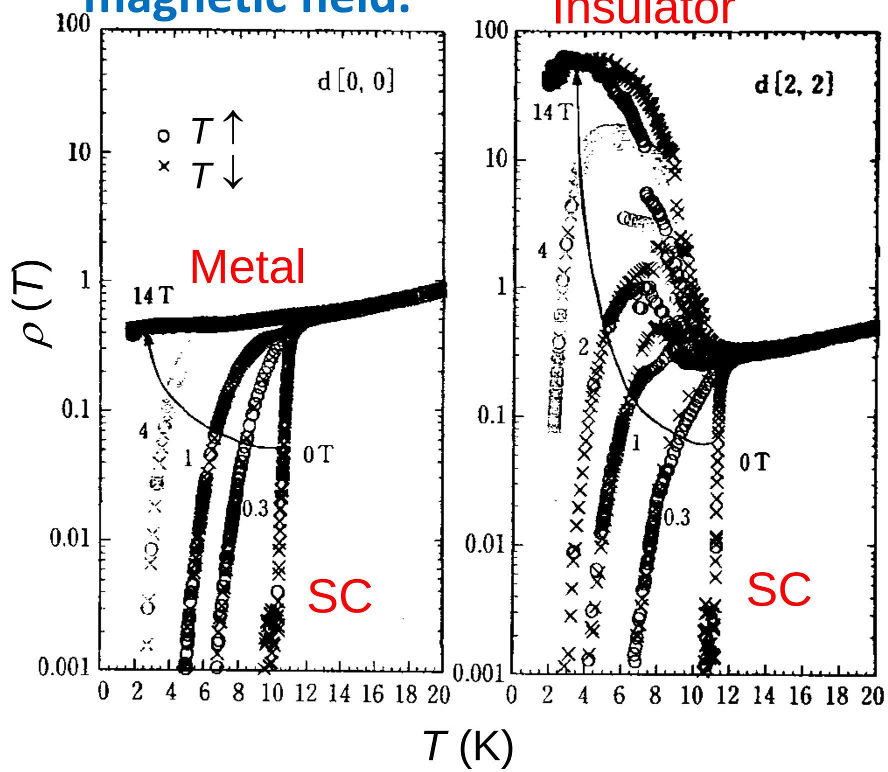
Electric resistivity of κ-(d[n,n]-ET)$_2$Cu[N(CN)$_2$]Br

Deuteration and cooling rate dependences of ρ(T)



ρ (T) of d[0,0] and d[2,2] under magnetic field.



[H. Taniguchi et al., PRB **67**, 014510 (2003).]

Deuteration	⇒ SC → AFI	⬅	Mott transition
Cooling rate（d[3,3]）	⇒ SC → AFI	⬅	???
Magnetic field（d[2,2]）	⇒ SC → AFI	⬅	???

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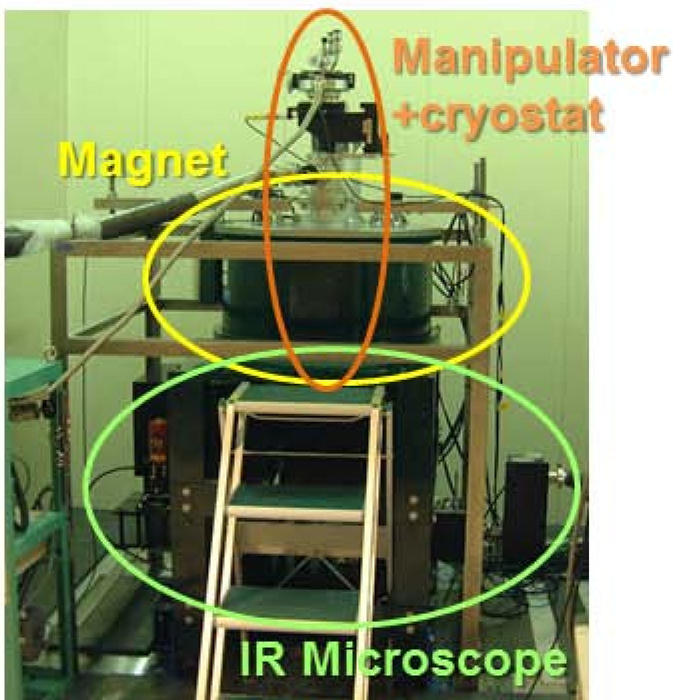
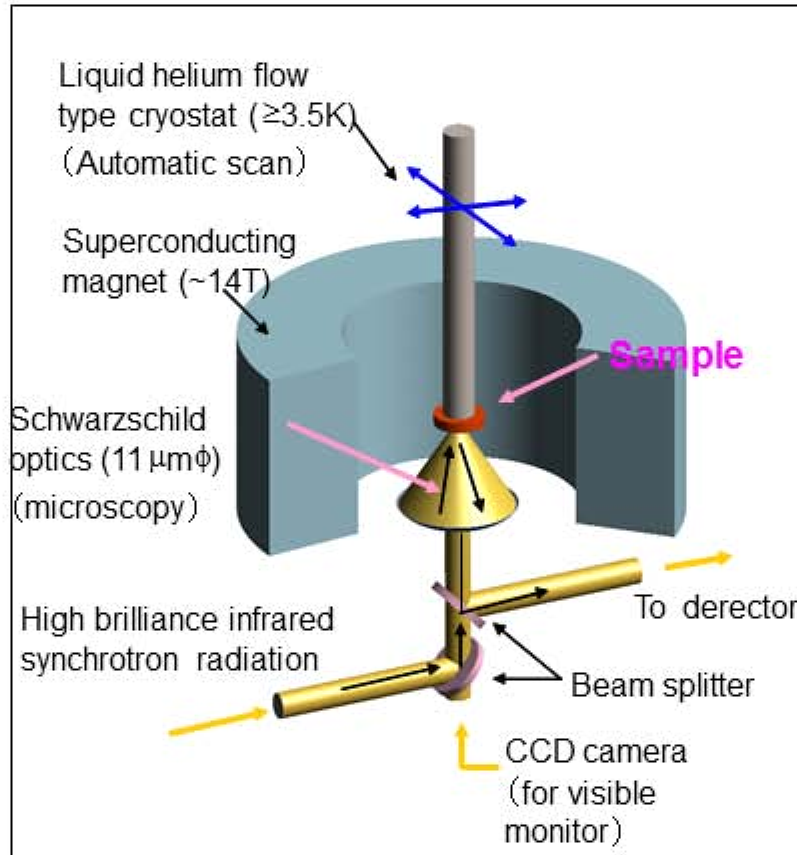
IR magneto-optical imaging station

[SK et al., Nucl. Instrum. Meth. A **467-468**, 437 (2001);
Nucl. Instrum. Meth. A **467-468**, 893 (2001);
Physica B **329-333**, 1625 (2003).]

- Photon energy: 2 – 0.1 eV
- Magnetic field: 0 - 14 T
- Temperature: 4.0 – 300 K
- Spatial resolution: 5 – 20 μm





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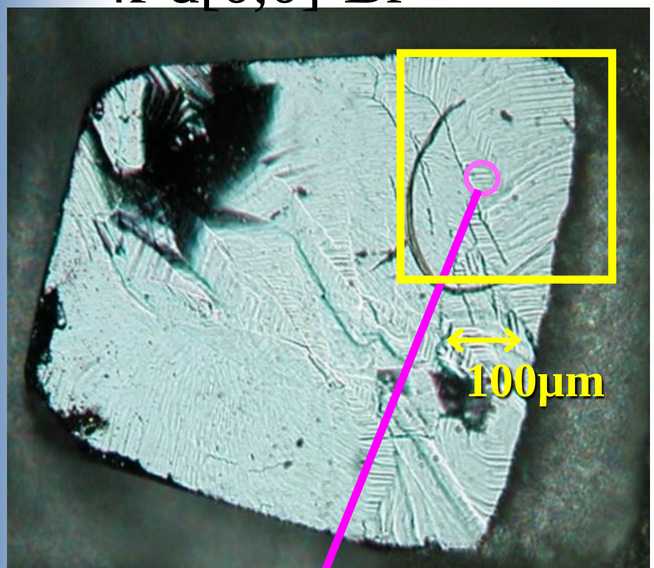
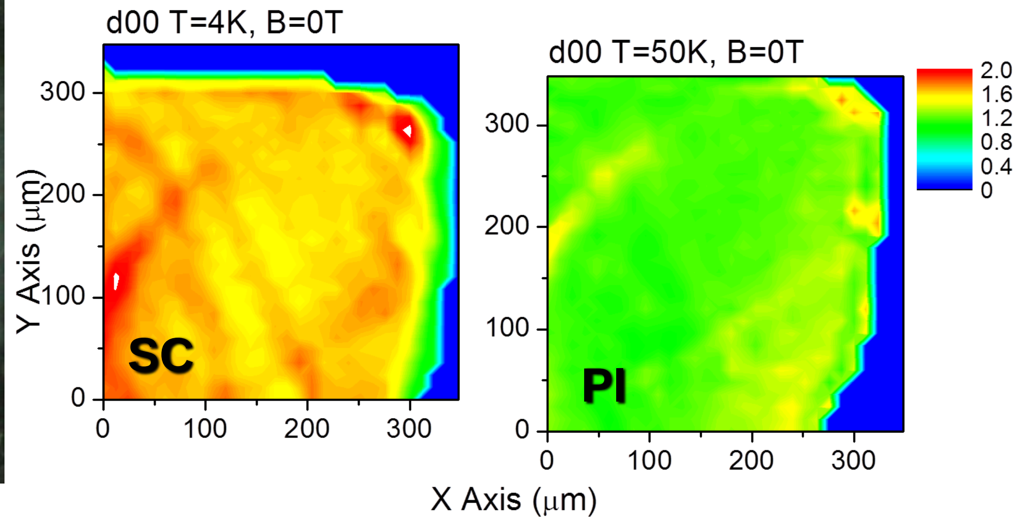
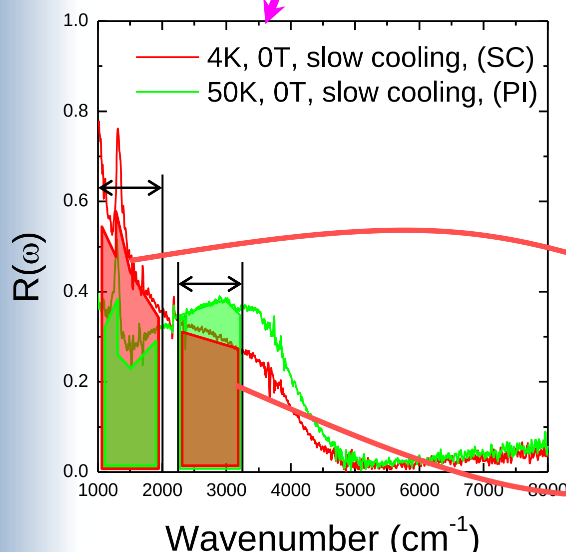
Phase imaging of κ-(d[0,0]-ET)$_2$Cu[N(CN)$_2$]Br

[T. Nishi, SK et al., Phys. Rev. B **75**, 014525 (2007).]

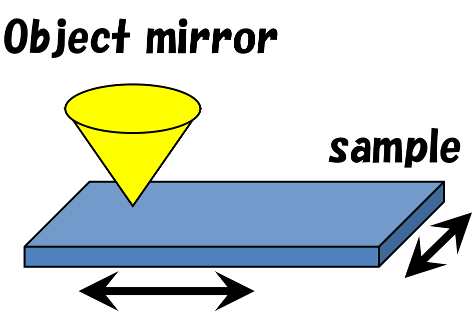
κ-d[0,0]-Br





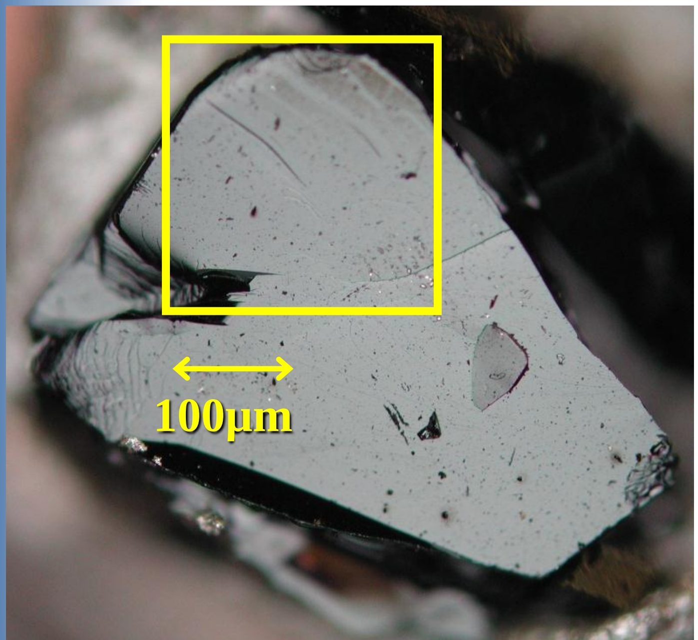


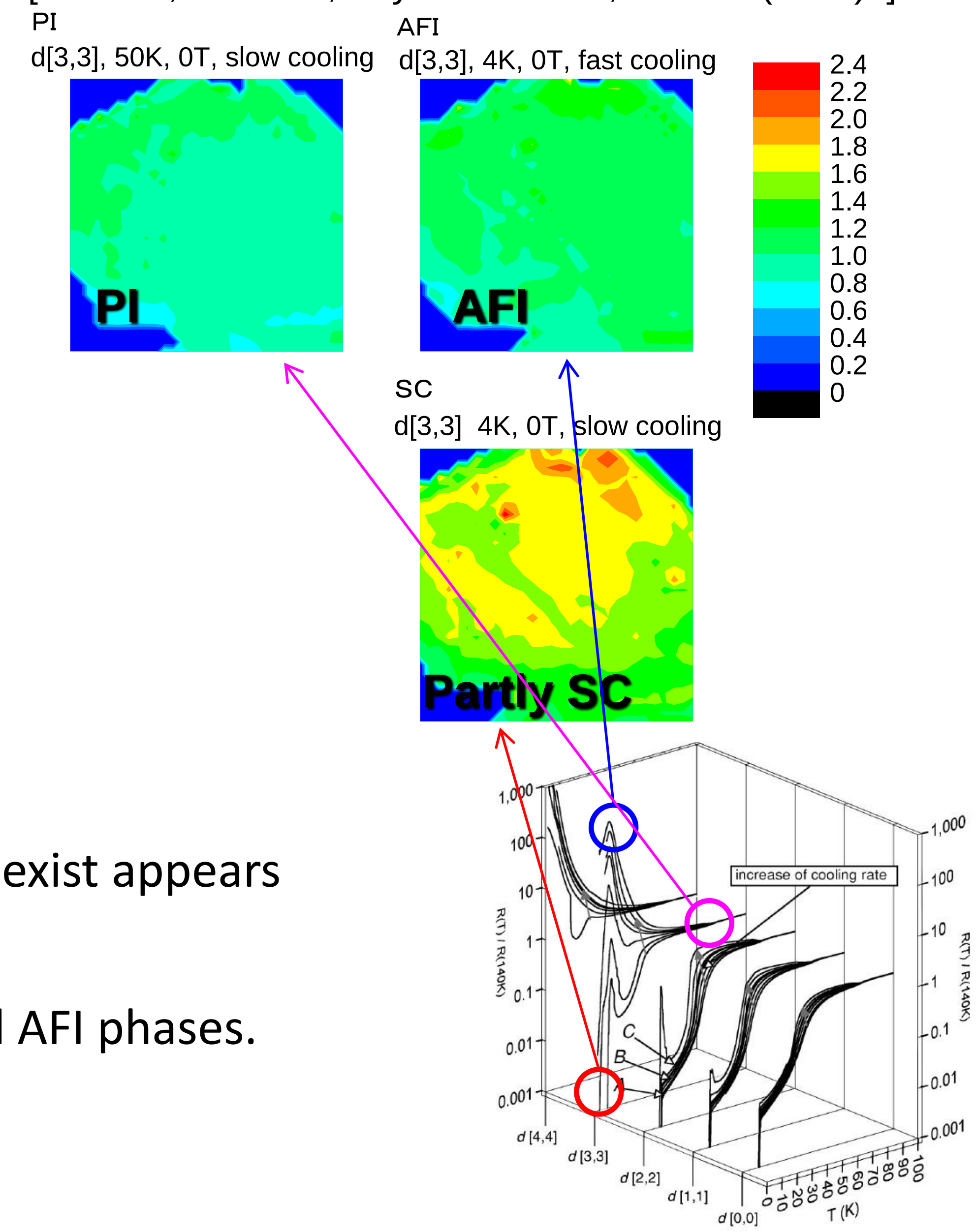
$$A = \frac{\int_{1000cm^{-1}}^{2000cm^{-1}} R(\omega)d\omega}{\int_{2350cm^{-1}}^{3350cm^{-1}} R(\omega)d\omega}$$



Metal-insulator phase imaging of κ-d[3,3]-Br

[T. Nishi, SK et al., Phys. Rev. B **75**, 014525 (2007).]

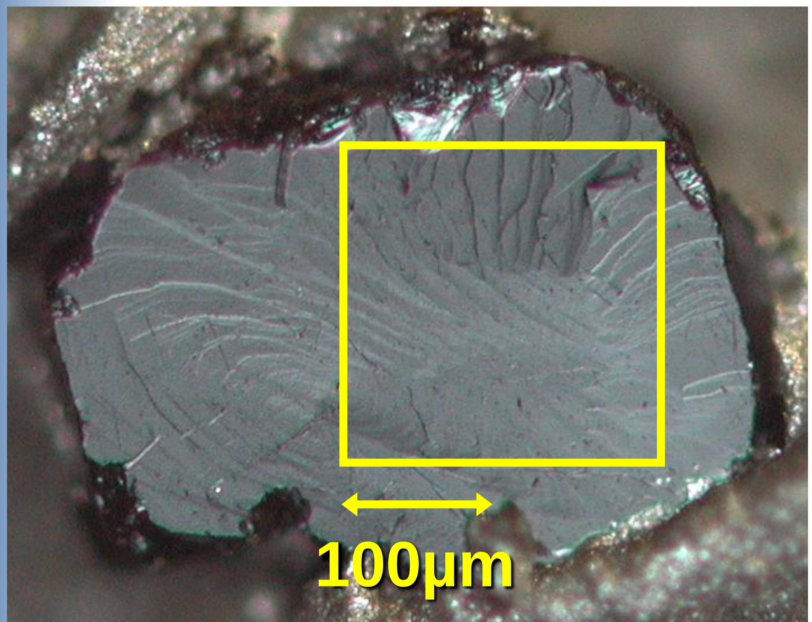




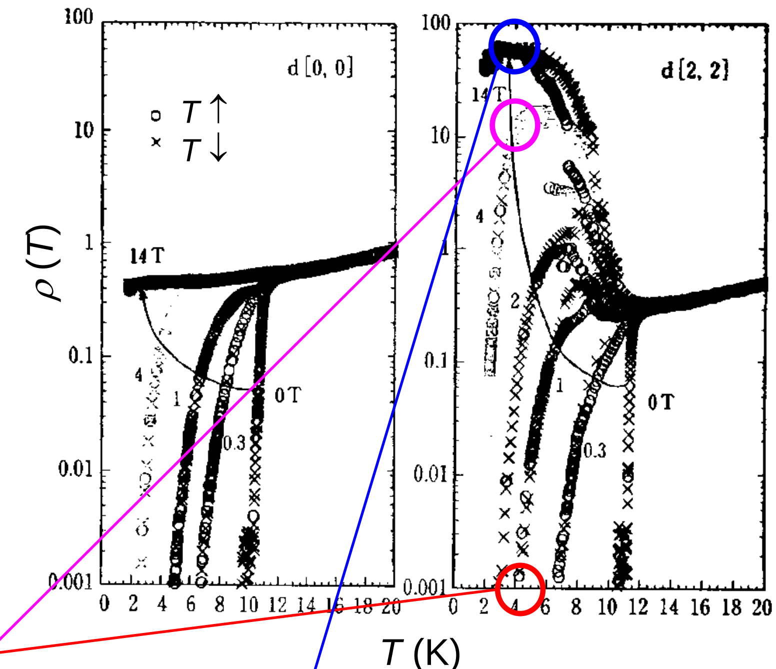
- Phase separation or phase co-exist appears in SC phase.
- Single phase appears in PI and AFI phases.

Metal-insulator phase imaging of κ-d[2,2]-Br

[T. Nishi, SK et al., Phys. Rev. B **75**, 014525 (2007).]



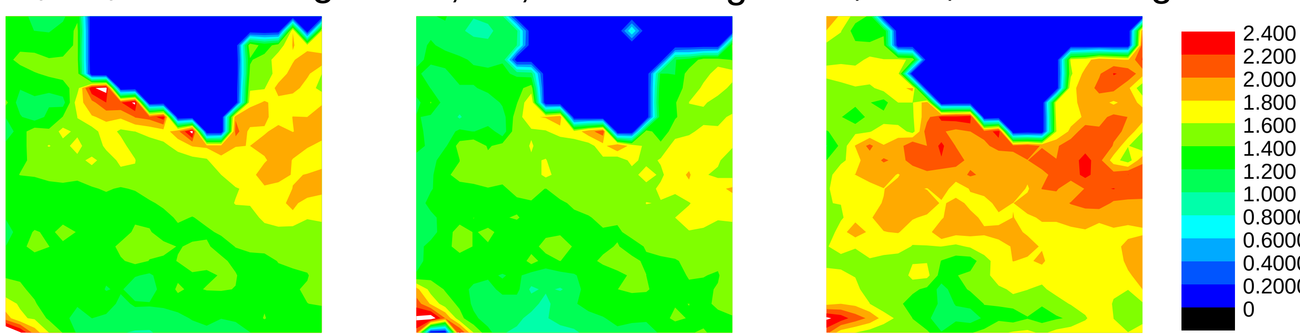
ρ(T) under *H* of d[0,0] and d[2,2]



SC
4K, 0T, fast cooling

AFI
4K, 5T, fast cooling

PM
4K, 10T, fast cooling



Outline

- Introduction to Infrared/terahertz spectroscopy and microscopy
  - Experimental methods and sources
  - Character of IR synchrotron radiation
  - Introduction of UVSOR-II
- Examples
  - IR micro-spectroscopy and imaging of correlated materials
    - Spatial imaging of metal-insulator transition of organic conductors under pressure
- Future IR/THz light sources
  - Coherent synchrotron radiation
- Summary

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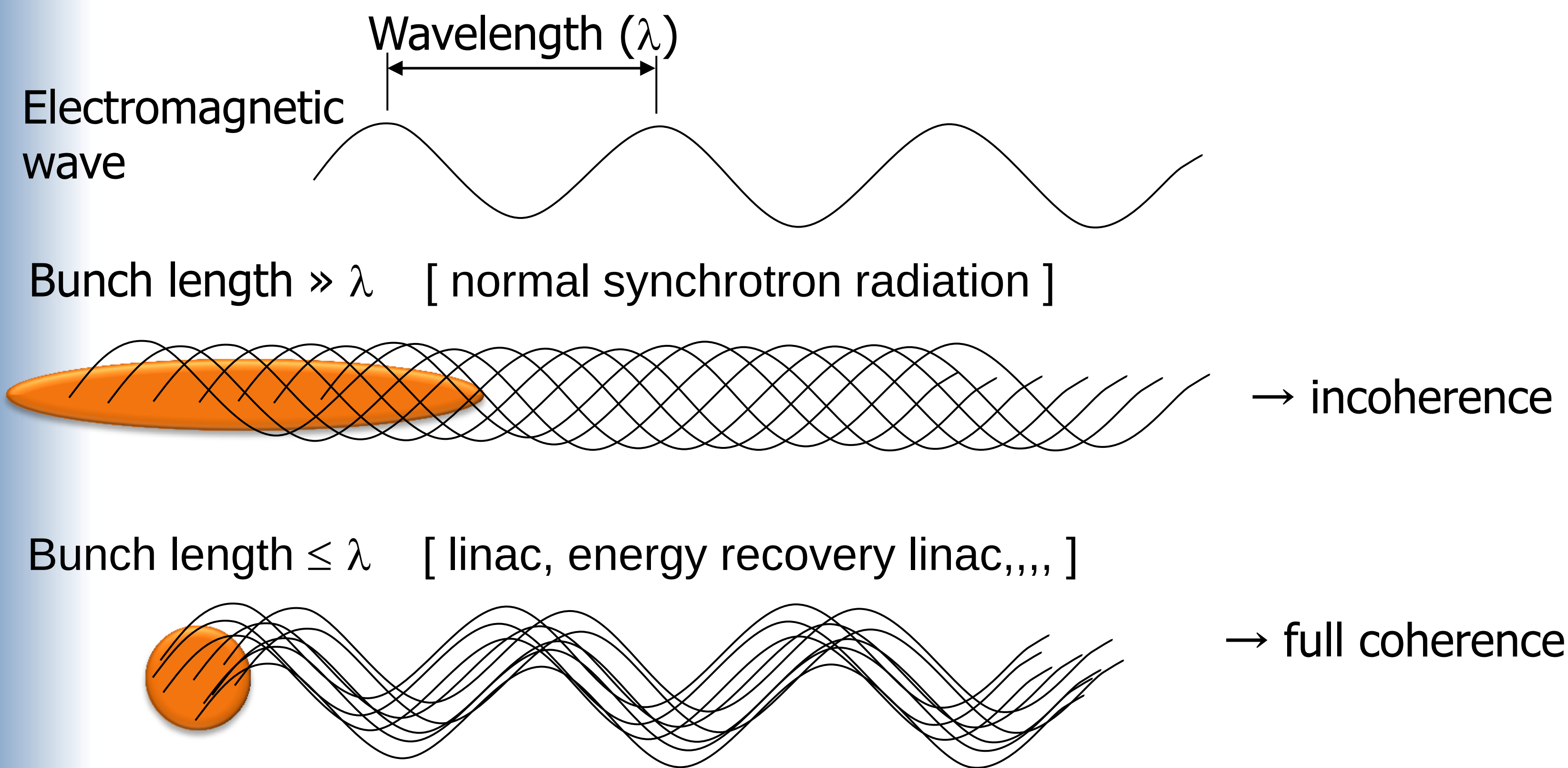
What's coherent synchrotron radiation (CSR) ?



Coherent Radiation

SR Power emitted by an electron bunch

Normal (Incoherent) SR

Coherent SR

$N_e \sim 10^{10}$

$$P = P_0 (N_e + N_e^2 F_e)$$

$$F_e = \left(\int \cos(2\pi z / \lambda) S(z) dz \right)^2$$

P_0; SR power from a single electron
N_e; Number of electrons in a bunch
F_e; Form factor of electron bunch
S(z); Longitudinal density distribution of electron bunch

First observation of THz coherent synchrotron/transition radiation

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Coherent synchrotron radiation (CSR)

VOLUME 63, NUMBER 12 PHYSICAL REVIEW LETTERS 18 SEPTEMBER 1989

Observation of Coherent Synchrotron Radiation

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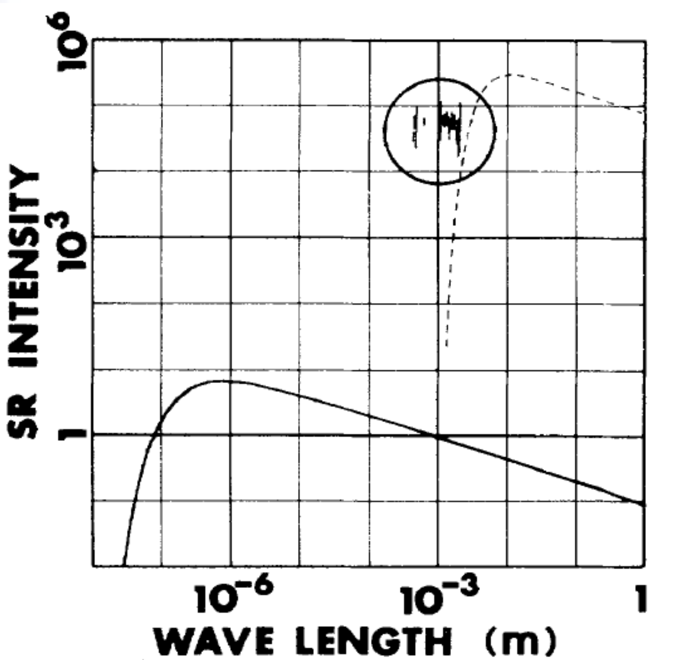
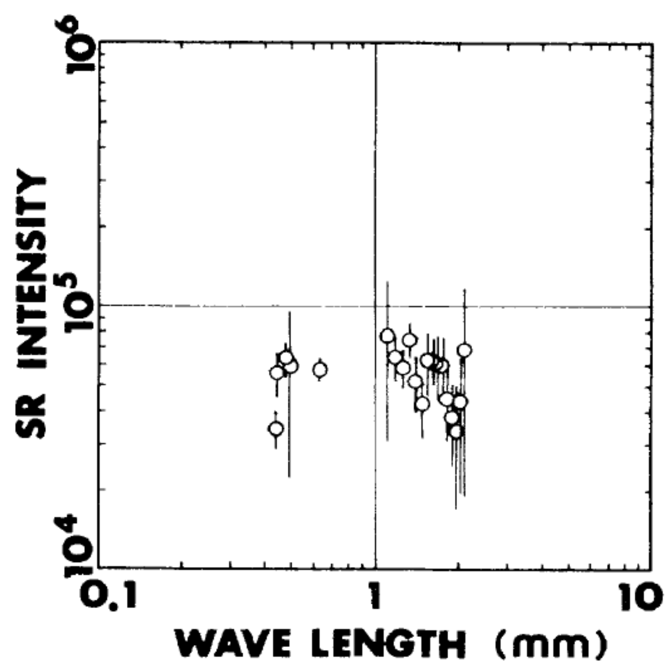
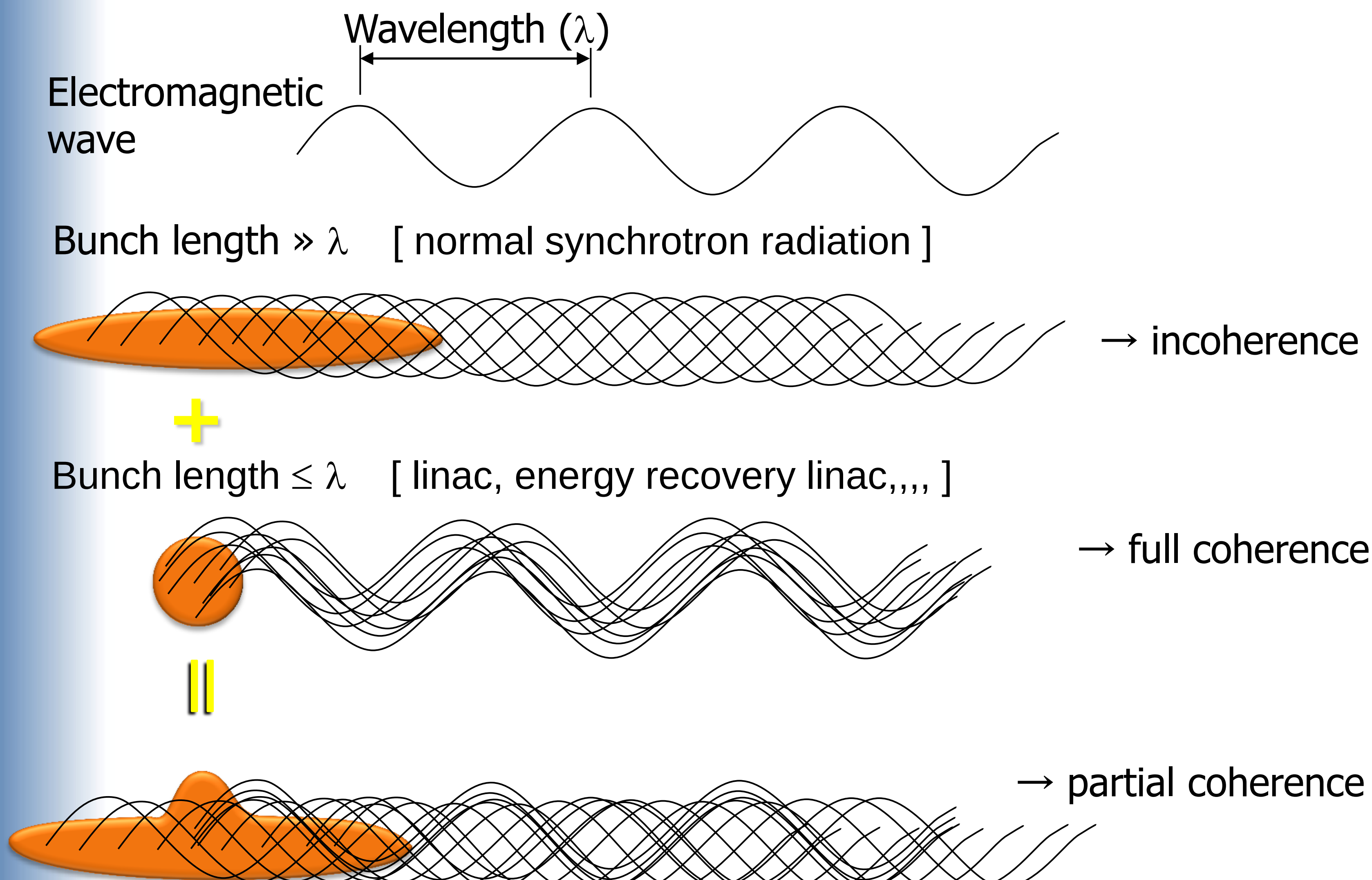
Y. Shibata, K. Ishi, T. Ohsaka, and M. Ikezawa
Research Institute for Scientific Measurements, Tohoku University, Katahira Sendai 980, Japan





What’s coherent synchrotron radiation (CSR) ?



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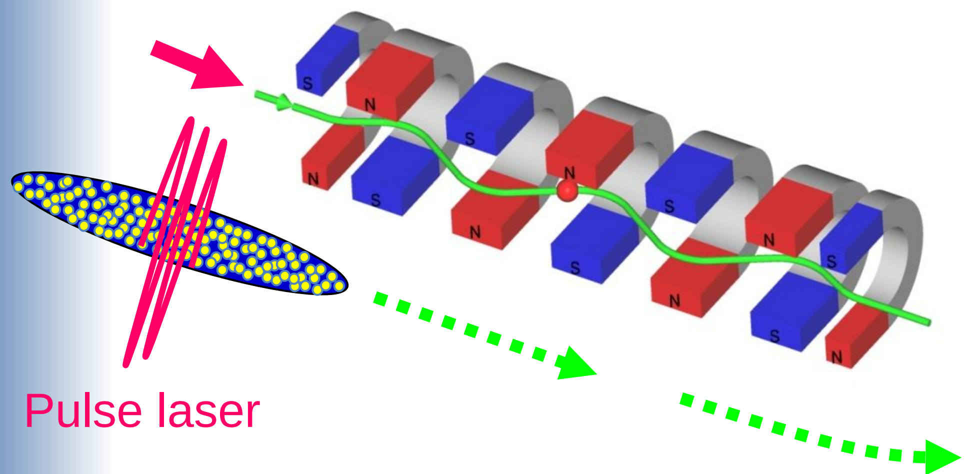
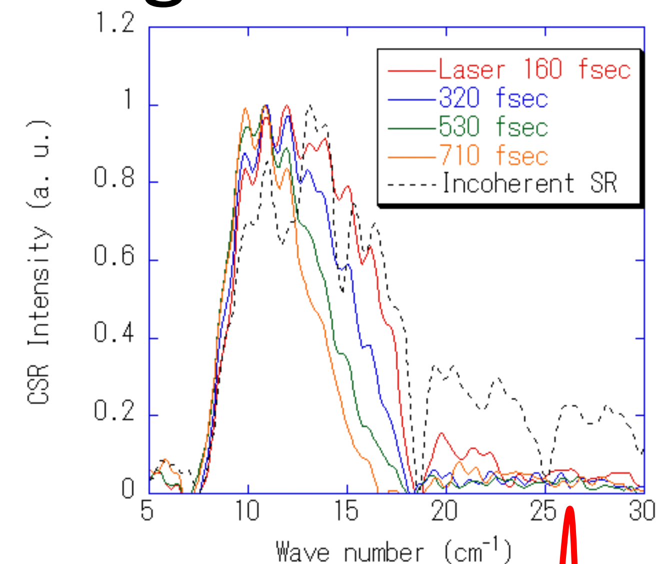
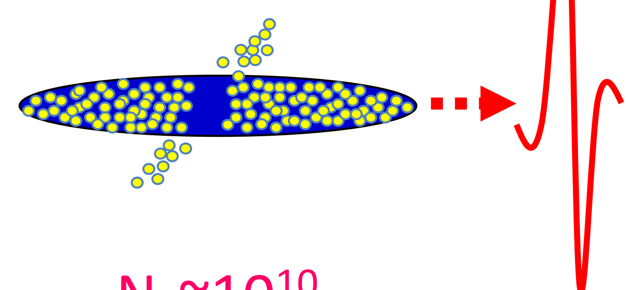
THz CSR via Laser Bunch Slicing at UVSOR-II

[M. Shimada, SK *et al.*, Jpn. J. Appl. Phys. **46**, 7939 (2007).]

Demonstrated at ALS, BESSY-II, UVSOR-II, SLS…







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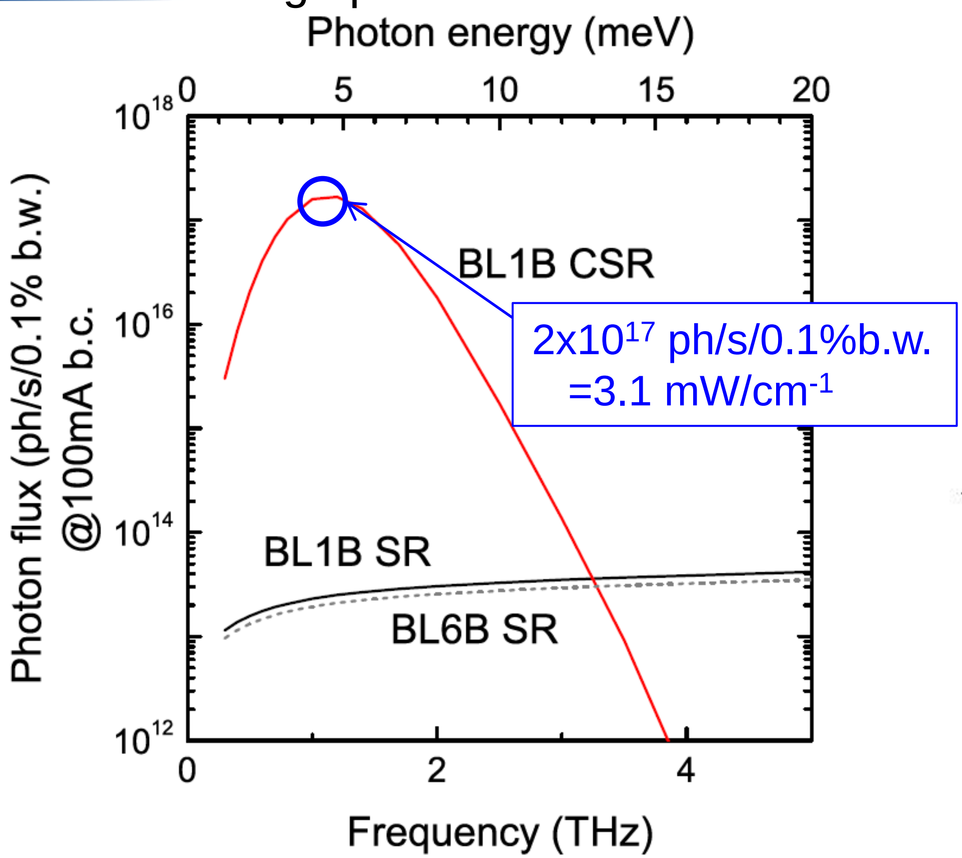
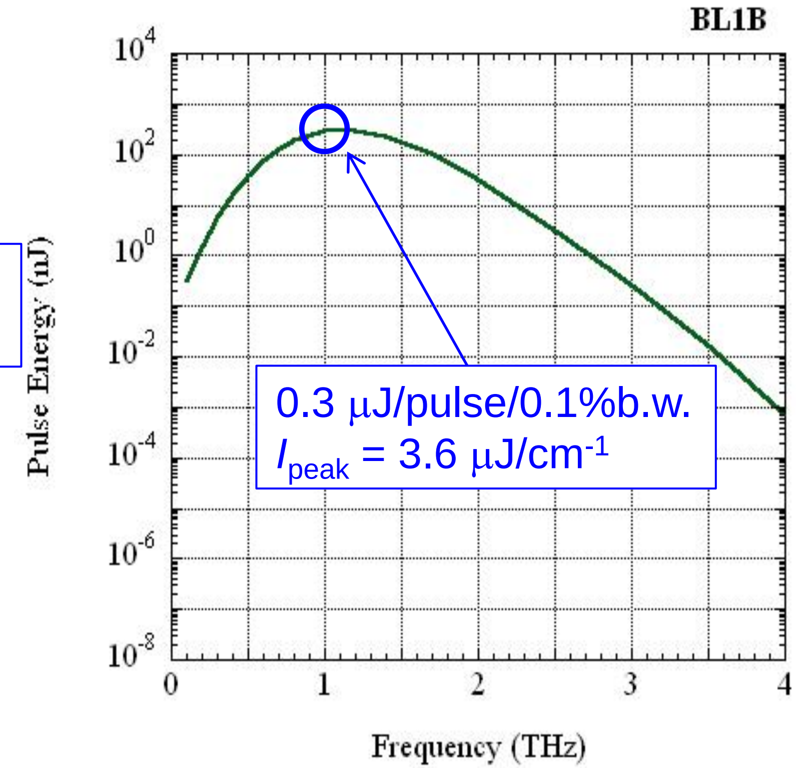
Photon flux and peak power of THz-CSR

(Expected by calculation)

Average photon flux



Peak power

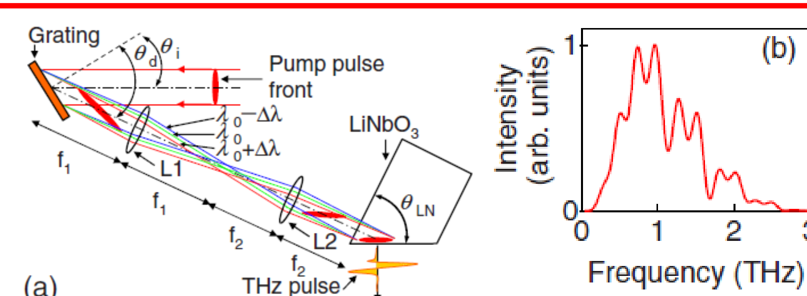


cf.) Peak power of laser THz source：

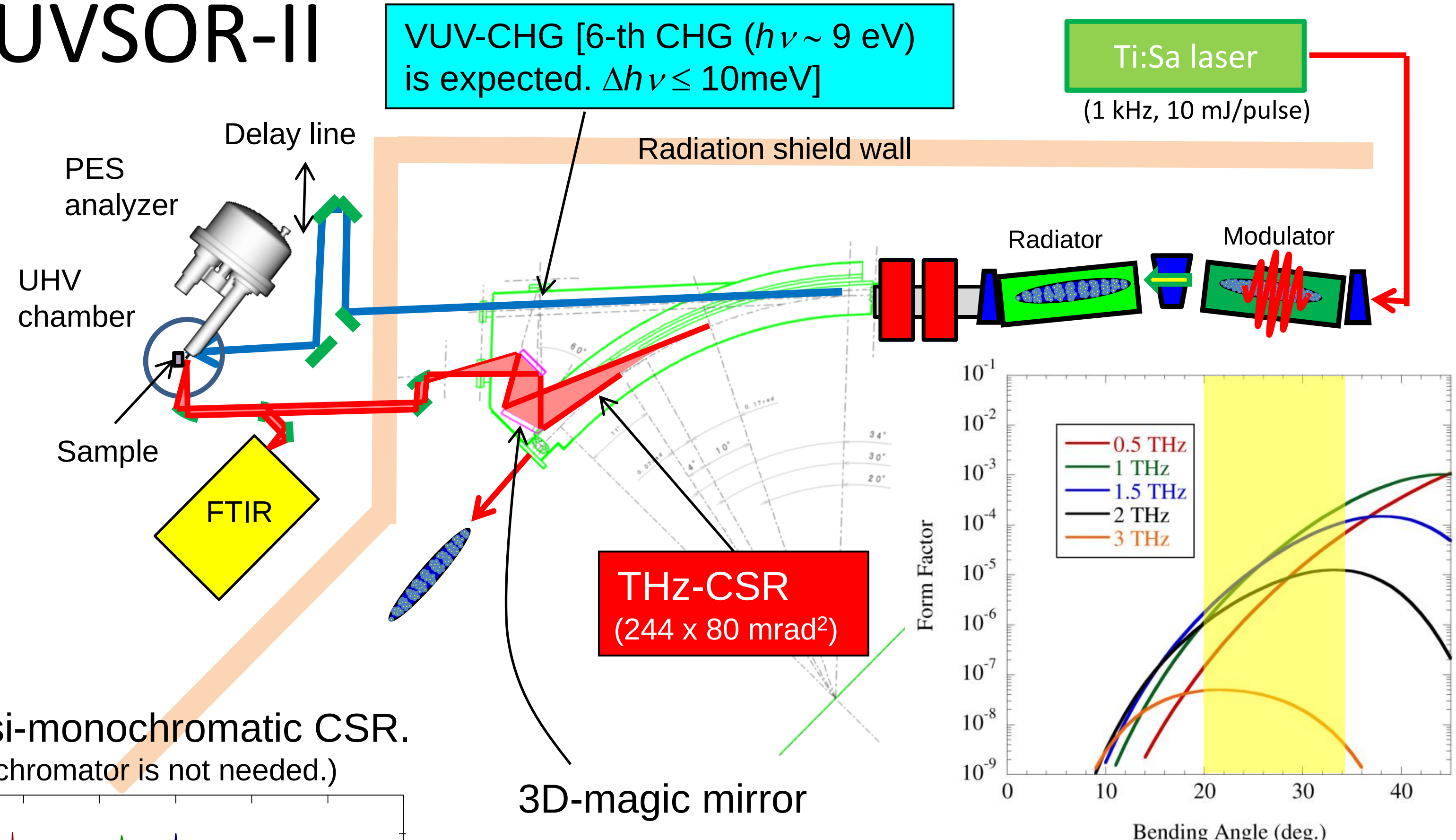
2 μJ/pulse

[Hiroi et al., APL **98**, 091106 (2011).]

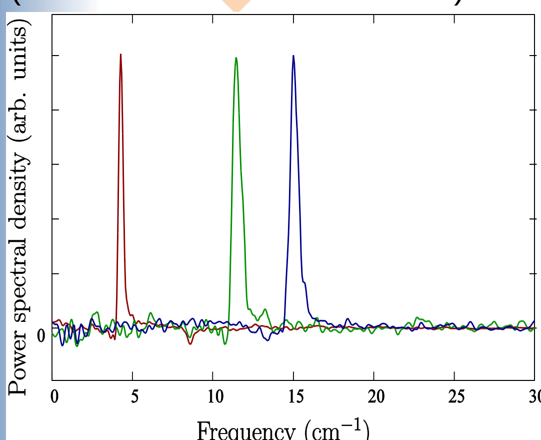
~ 2 nJ/pulse/0.1%b.w.

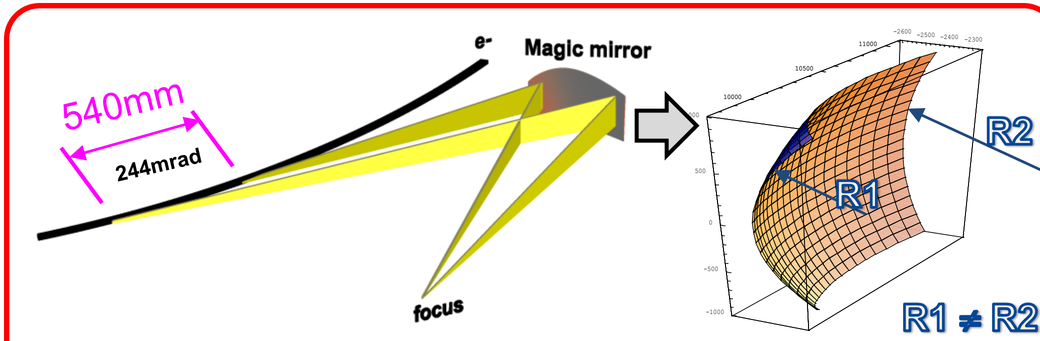


THz pump – PES probe beamline at BL1 of UVSOR-II



Quasi-monochromatic CSR.
(Monochromator is not needed.)





[S. Kimura et al., NIMA **467-468**, 437 (2001).]

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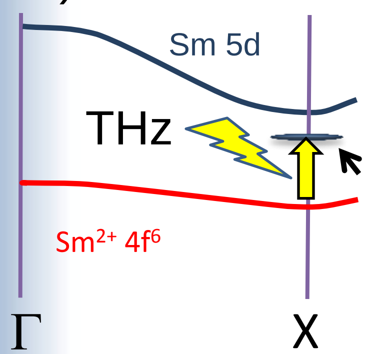
Target of THz pump – PES probe spectroscopy

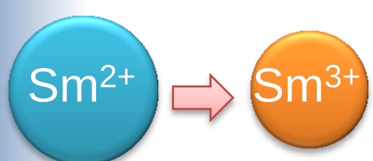
➢To clarify the origin of the functionalities and to find new physics.

✓Local relaxation of mixed valent system

ex.) SmS

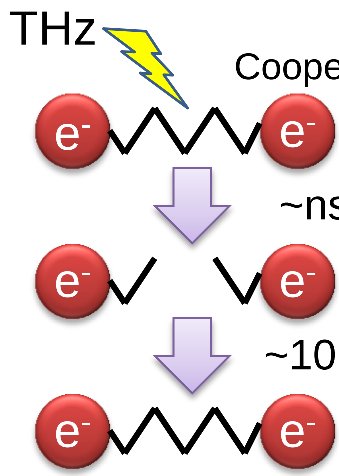


Excitonic instability.

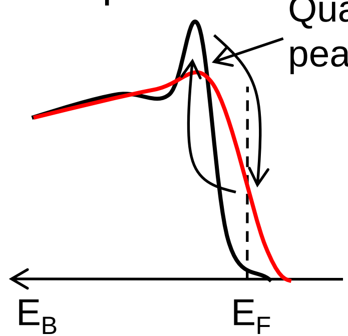


Lattice contraction→Change of electronic structure. (~psec - msec?)

✓Collapse and recombination of Cooper pairing.

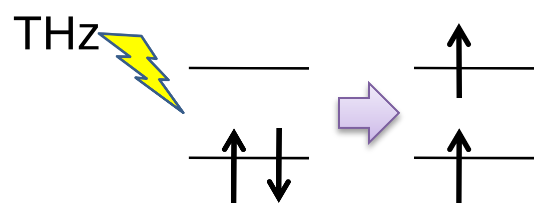


PES spectrum

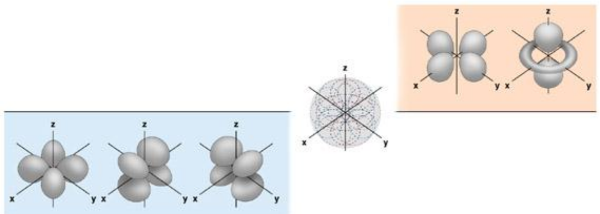


✓Relaxation process of spin excitation and lattice reformation due to Jahn-Teller effect.

Ex.) V^{3+}, Cr^{4+} system



Change of electric field due to the disposition of electrons in ligands.



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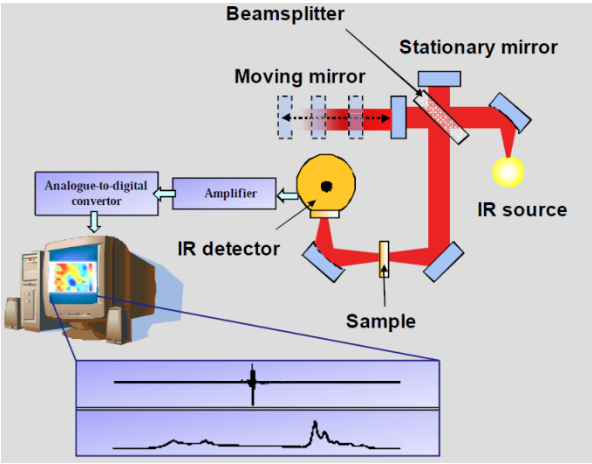

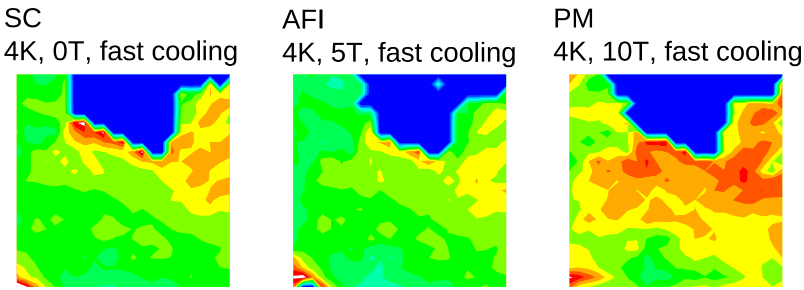

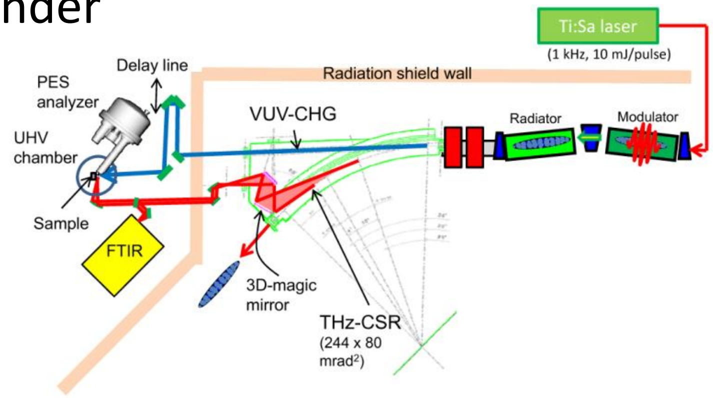

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