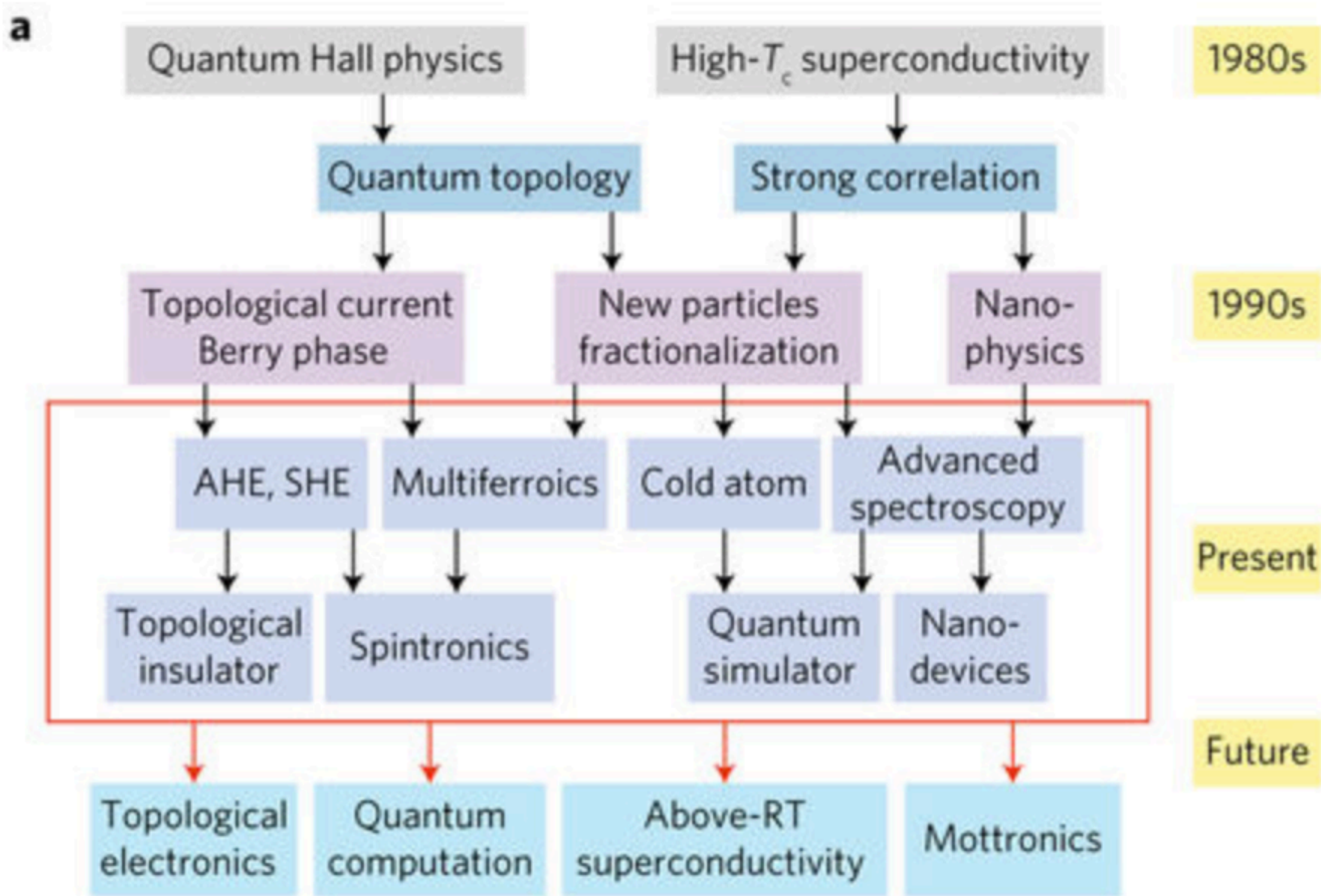
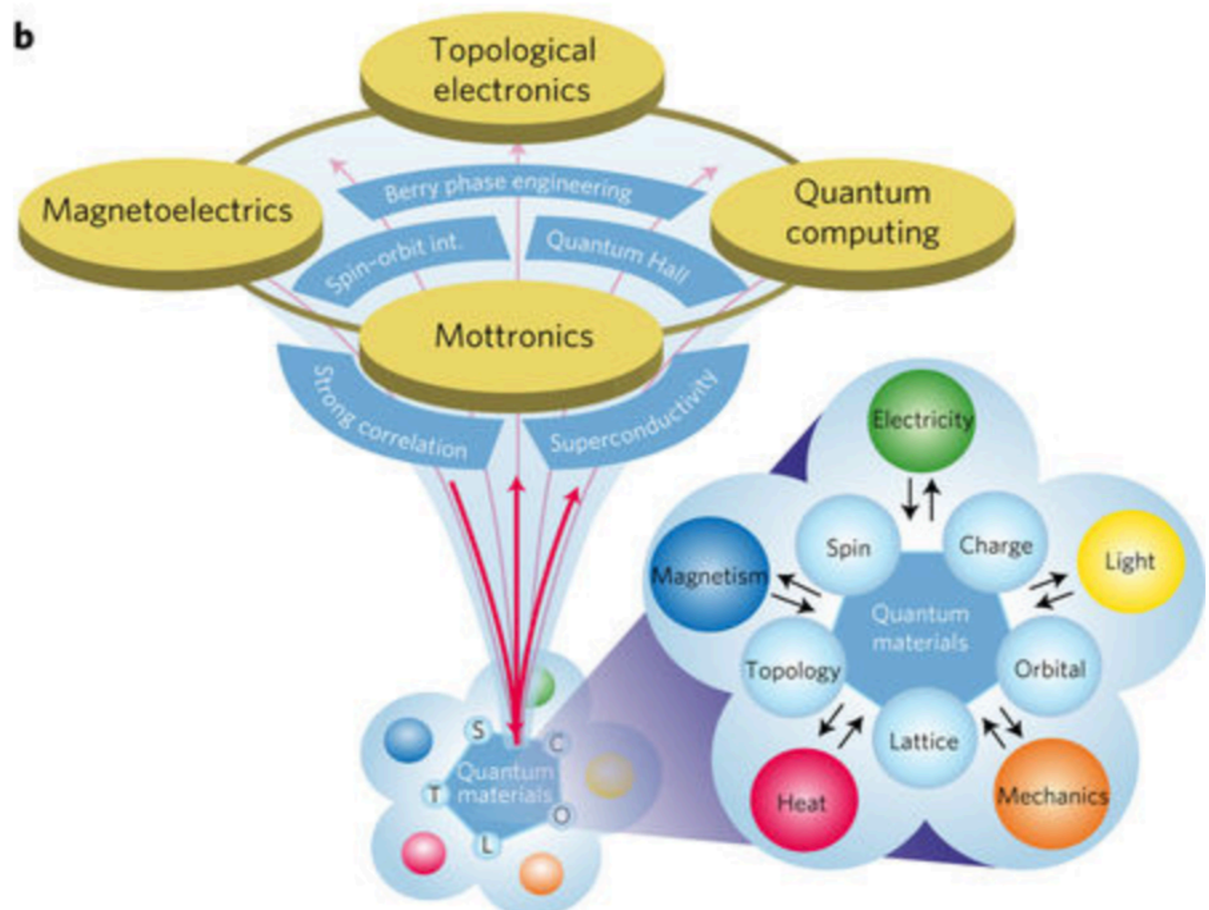
Emergent Materials and Spectroscopic Techniques

Brief history of quantum materials and functions



Yoshinori Tokura, Masashi Kawasaki & Naoto Nagaosa
Nature Physics **13**, 1056–1068 (2017)

Various degrees of freedom of strongly correlated electrons in solids



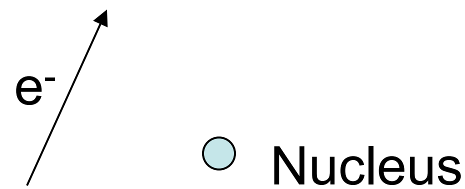
Yoshinori Tokura, Masashi Kawasaki & Naoto Nagaosa
Nature Physics **13**, 1056–1068 (2017)

Topological insulators

- “Topological”: topological properties of the band structure in the reciprocal space
- “Insulators”: well, not really. They have gap, but they are conducting (on edges)!
- Time-reversal-invariant topological insulators (Kane, Mele, Fu, Zhang, Qi, Bernevig, Molenkamp, Hasan and others, from 2006 and still on-going)

Spin-orbit coupling

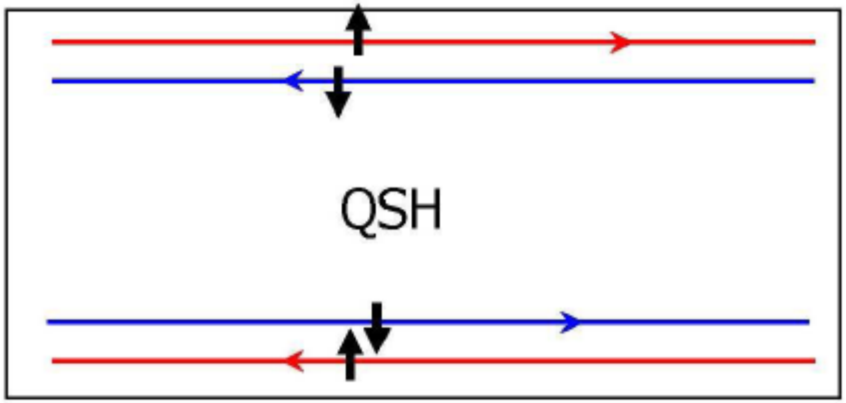
$$\vec{B} = -\frac{\vec{v} \times \vec{E}}{c^2}$$



$$H_{SO} = \frac{\mu_B}{\hbar m e c^2} \frac{1}{r} \frac{\partial U(r)}{\partial r} \vec{L} \cdot \vec{S}$$

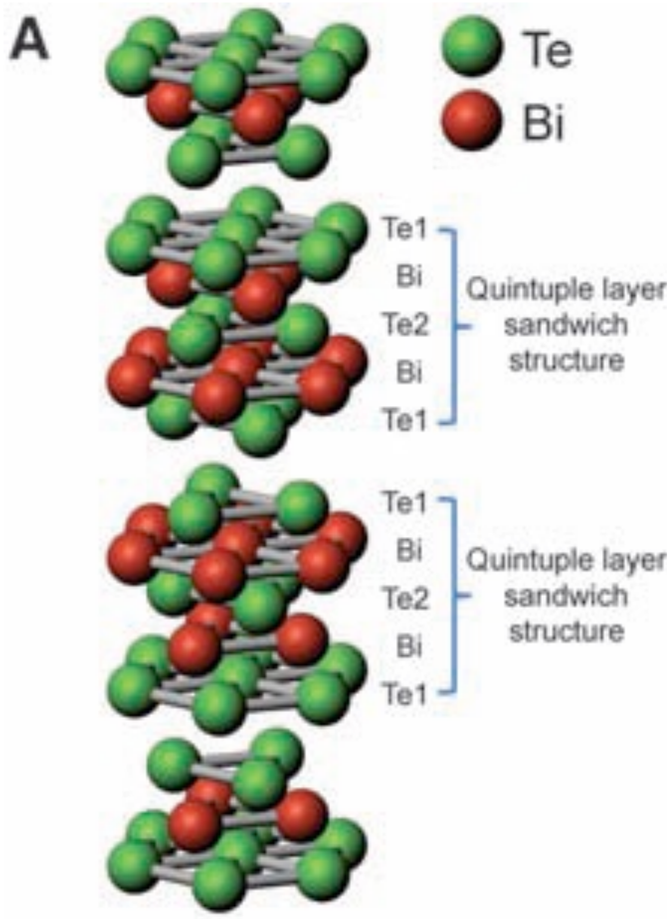
Stronger effect for heavy elements (Pb, Bi, etc.) from the bottom of the periodic system

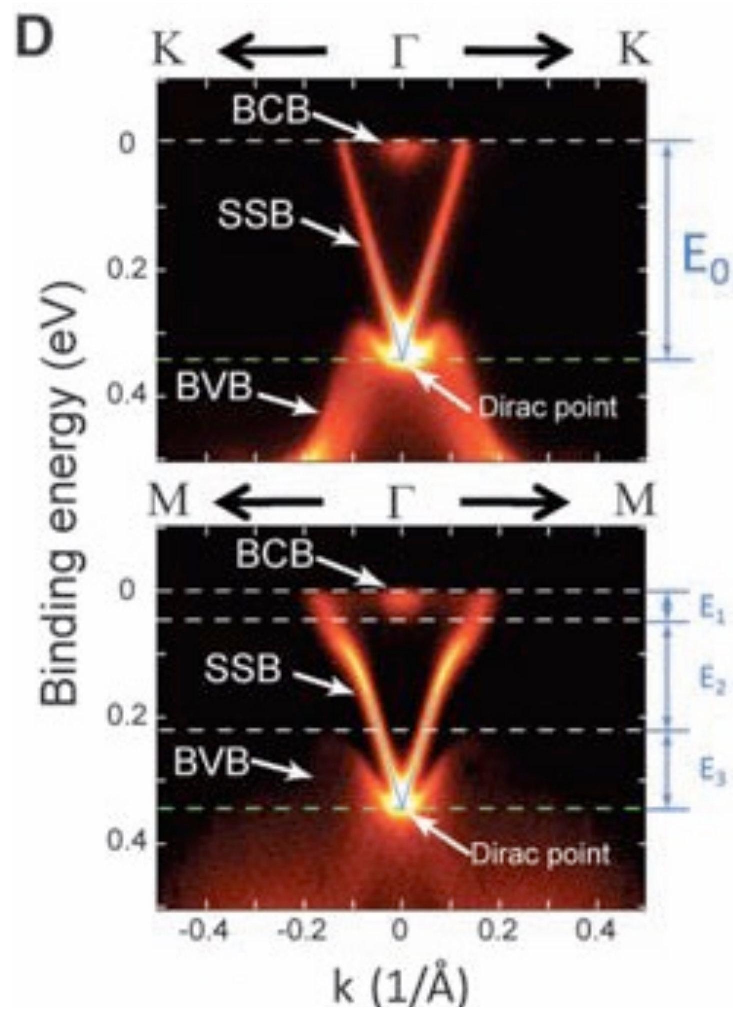
Edge states in 2D TIs



Helical modes: on each edge one pair of 1D modes related by the TR symmetry. Propagate in opposite directions for opposite spin.

Experimental detection in Bi_2Te_3

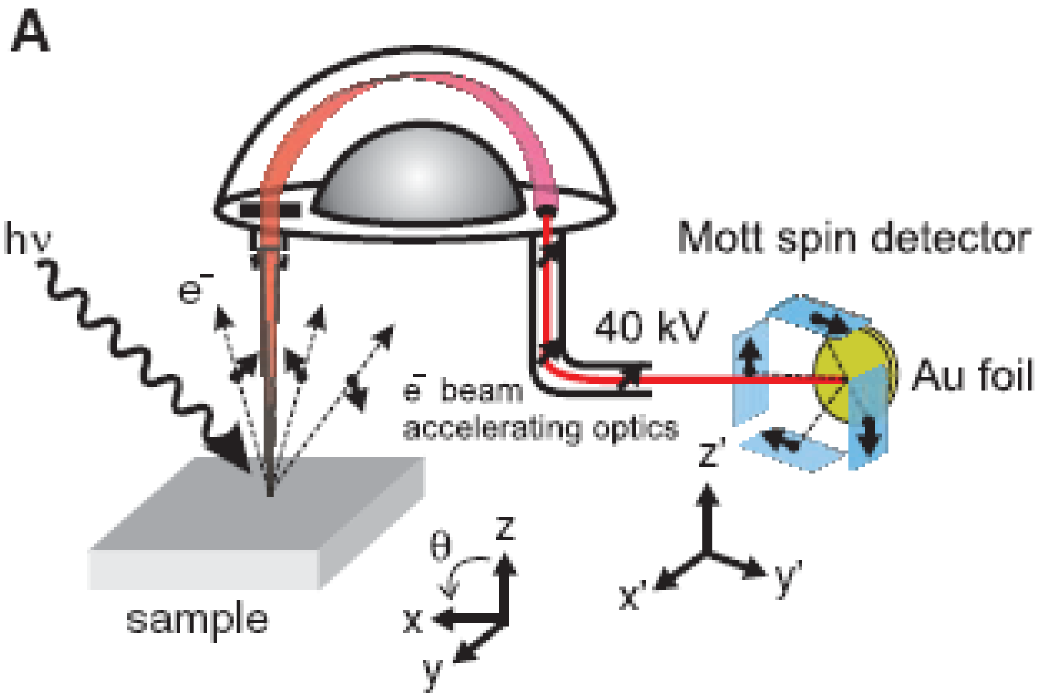


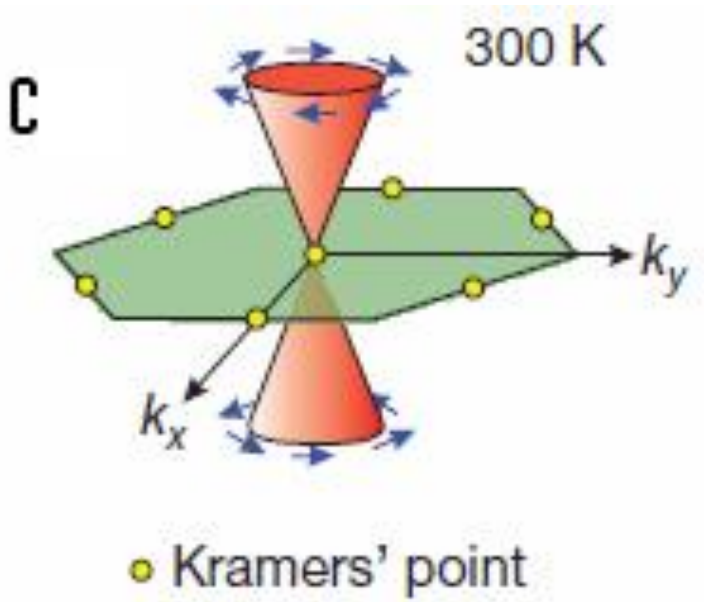


Chen et al. Science (2009)

Spin-momentum locking

$$\langle \mathbf{s}(-\mathbf{k})\rangle = -\langle \mathbf{s}(\mathbf{k})\rangle$$





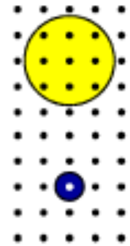
Spin-resolved ARPES

Hsieh, Science (2009)

Strongly correlated systems

d - f

open shells materials

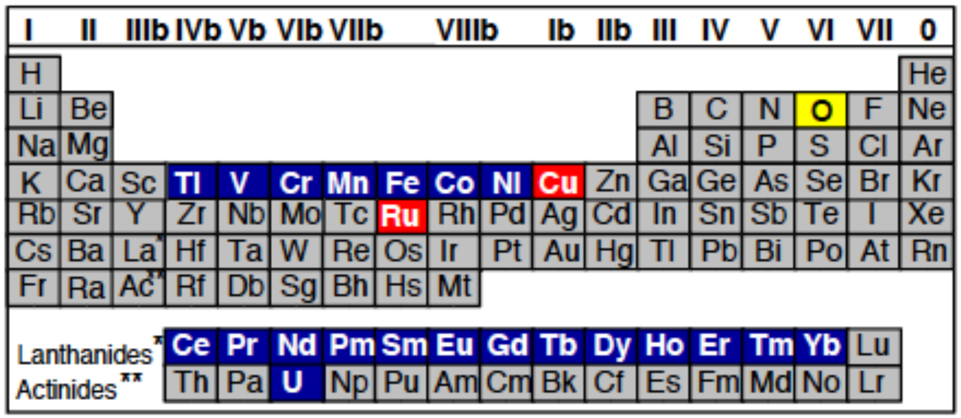


U<<W
Charge fluctuations

U>>W
Spin fluctuations

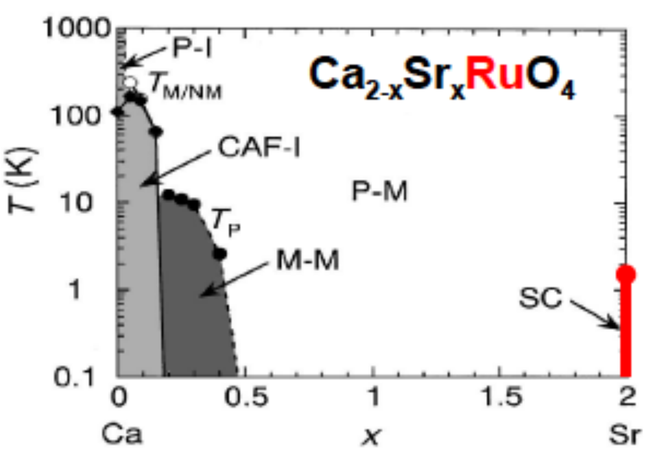
Control parameters

Bandwidth (U/W)
Band filling
Dimensionality

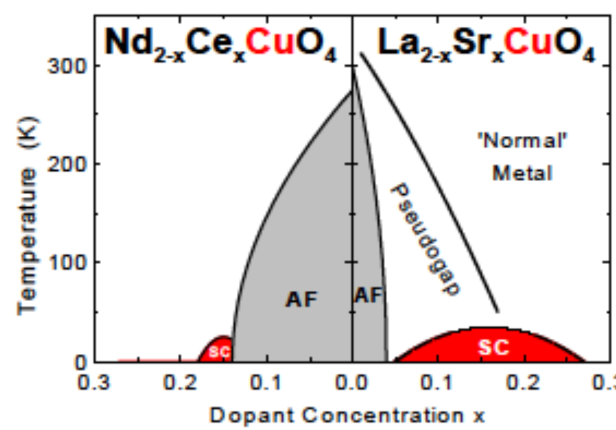


Degrees of freedom

Charge / Spin
Orbital
Lattice



- Kondo
- Mott-Hubbard
- Heavy Fermions
- Unconventional SC
- Spin-charge order
- Colossal MR



Fe-Pnictide high temperature superconductors:

Binary componds of pnictogens. A pnictogen – an element from the nitrogen group N,P, As,Sb,Bi

RFeAsO (1111)
R = La, Nd, Sm, Pr, Gd
LaOFeP

AFe_2As_2 (122)
A = Ba, Sr, Ca

LiFeAs (111)

Fe(Se/Te) (11)

Fe-pnictides:

May 2006

J|A|C|S COMMUNICATIONS

Published on Web 07/15/2006

2006

Iron-Based Layered Superconductor: LaOFeP

Yoichi Kamihara,† Hidenori Hiramatsu,† Masahiro Hirano,†,‡ Ryuto Kawamura,§ Hiroshi Yanagi,§ Toshio Kamiya,†,§ and Hideo Hosono*,†,‡

ERATO-SORST, JST, Frontier Collaborative Research Center, Tokyo Institute of Technology, Mail Box S2-13, 4259 Nagatsuta, Midori-ku, Yokohama 226-8503, Japan, Frontier Collaborative Research Center, Tokyo Institute of Technology, Mail Box S2-13, 4259 Nagatsuta, Midori-ku, Yokohama 226-8503, Japan, and Materials and Structures Laboratory, Tokyo Institute of Technology, Mail Box R3-4, 4259 Nagatsuta, Yokohama 226-8503, Japan

Received May 15, 2006; E-mail: hosono@msl.titech.ac.jp

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ナノテクノロジーの専門情報

日経ナノビジネスについて

JSTと東工大、新しい酸化物超電導体LaOFePを発見

2006-07-18（黒川 卓）

印刷用ページ

科学技術振興機構（JST）は東京工業大学と協同で、層状構造の新しい酸化物超電導体を発見した。酸化物超電導体としては遷移金属の銅（Cu）を含む物質がよく知られ、すでに産業界で実用化が始まっている。今回発見されたのは、Cuの代わりに遷移金属の鉄（Fe）を含む酸化超電導体。超電導転移する温度（Tc）は4Kと今のところ低いが、今後、構成元素の種類と組成比を変えることによってTcをさらに高められる可能性がある。

今回発見された超電導体の化学組成はLaOFeP（ランタン・鉄・リン酸化物）。

2008

Superconductivity at 43 K in Samarium-arsenide Oxides $SmFeAsO_{1-x}F_x$

X. H. Chen* and T. Wu, G. Wu, R. H. Liu, H. Chen and D. F. Fang

Hefei National Laboratory for Physical Science at Microscale and Department of Physics, University of Science and Technology of China, Hefei, Anhui 230026, People's Republic of China

arXiv:0803.3603v1 [cond-mat.supr-con] 25 Mar 2008

(Dated: March 25, 2008)

$LaFeAsO_{1-x}F_x$, Tc = 26K

$SmFeAsO_{1-x}F_x$, Tc = 43K

nature International weekly journal of science

Letter

Nature 453, 761-762 (5 June 2008) |

Superconductivity at

X. H. Chen1 (#a1), T. Wu1 (#a1)

1. Hefei National Laboratory for Phys

Correspondence to: X. H. Chen1 (#a1)

Since the discovery of hig been devoted to exploring from Bardeen–Cooper–Sc copper oxide supercondu (ref. 2 (/nature/journal/v453 La–Nd, Sm and Gd) are n $LaO_{1-x}F_xFeAs$ (ref. 3 (/natu superconductivity in the r measurements reveal a tr high-temperature superc

nature International weekly journal of science

go | Advanced search

nature International weekly journal of science

Letter

Nature 459, 64-67 (7 May 2009) | doi:10.1038/nature07981; Received 4 November 2008; Accepted 13 March 2009

A large iron isotope effect in $SmFeAsO_{1-x}F_x$ and $Ba_{1-x}K_xFe_2As_2$

R. H. Liu1, T. Wu1, G. Wu1, H. Chen1, X. F. Wang1, Y. L. Xie1, J. J. Ying1, Y. J. Yan1, Q. J. Li1, B. C. Shi1, W. S. Chu2,3, Z. Y. Wu2,3 & X. H. Chen1

1. Hefei National Laboratory for Physical Sciences at Microscale and Department of Physics, University of Science and Technology of China, Hefei, Anhui 230026, China
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Accepted 5 May 2008; Published online 4 June 2008

;F0.15

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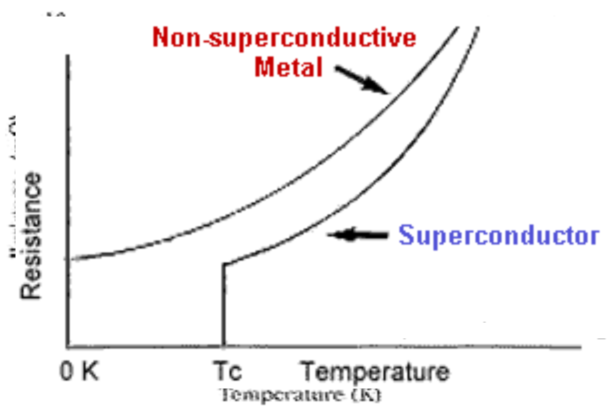
Superconductivity:

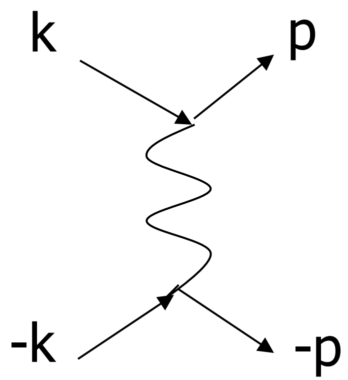
Zero-resistance state of interacting electrons



Electrons (fermions) attract each other and form bound states (bosons) . Bound states condence (a'la Bose-Einstein condensation) and move fully coherently under the electric field. One needs to destroy a bound state to stop the current.

BCS theory

• If there is an attractive interaction between fermions, they always form a bound state and condense below a certain Tc



• In conventional, low Tc superconductors, an attractive interaction is provided by exchanging phonons (lattice vibrations)

Superconductivity: High-T_c

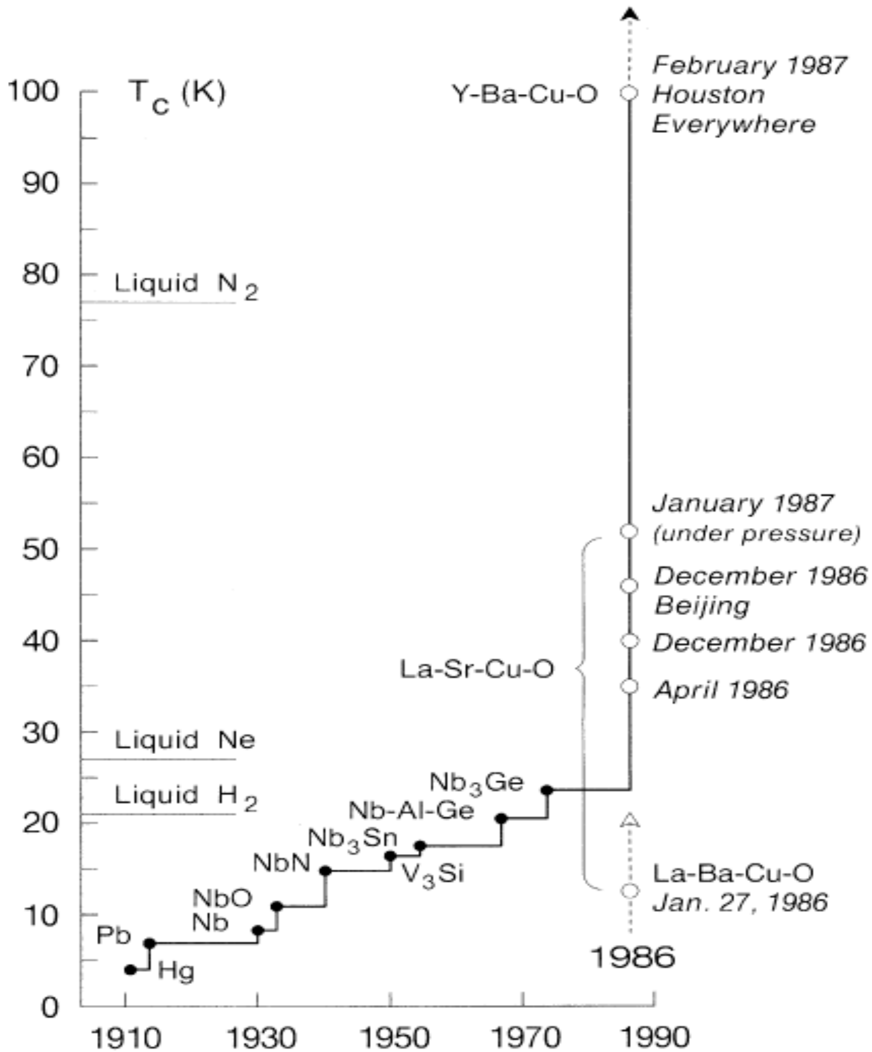
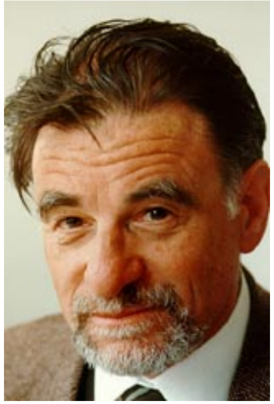


Fig. 1. Evolution of the superconductive transition temperature subsequent to the discovery of the phenomenon.



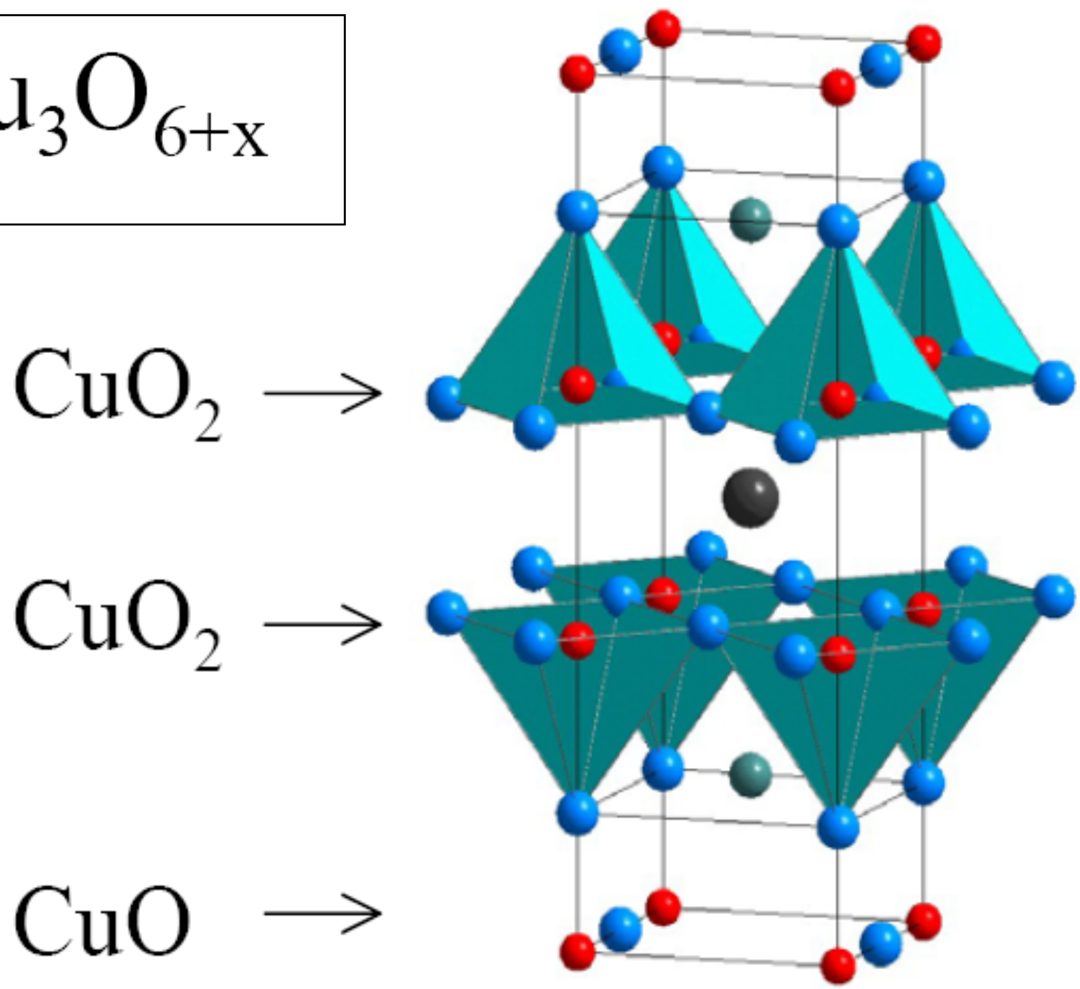
Alex Muller and Georg Bednortz

Nobel prize, 1987

10^5 publications

What is so exciting about high Tc superconductors?

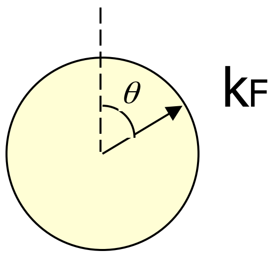
$YBa_2Cu_3O_{6+x}$



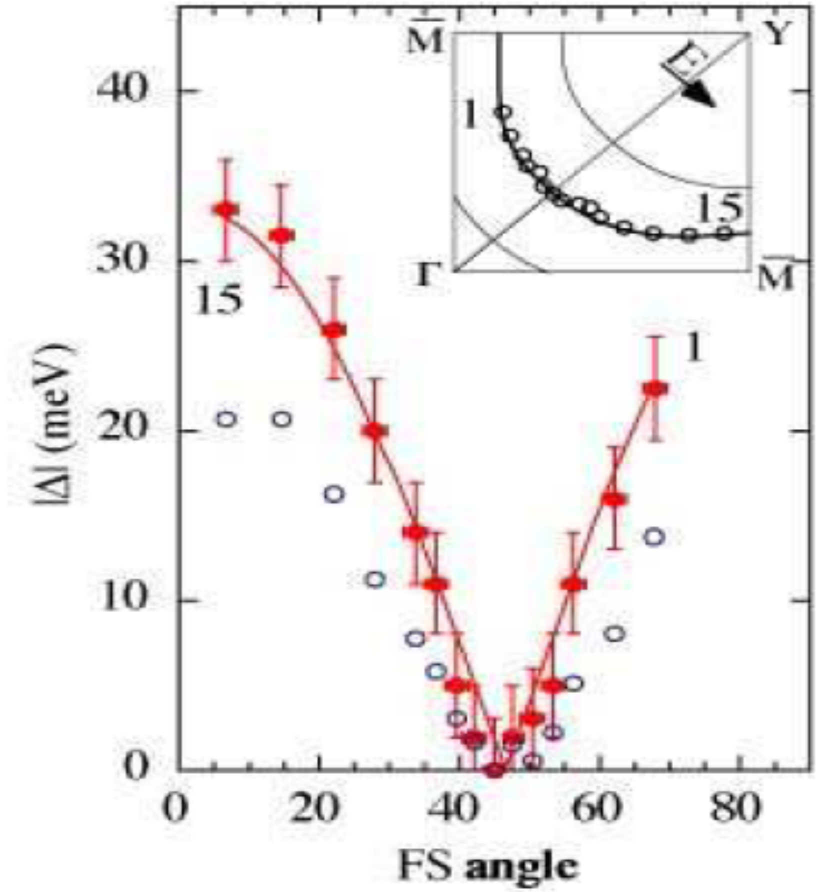
What is so exciting about high Tc superconductors?

d-wave symmetry of the superconducting gap

2. Most likely, electron-electron interaction
rather that electron-phonon interaction
is resp

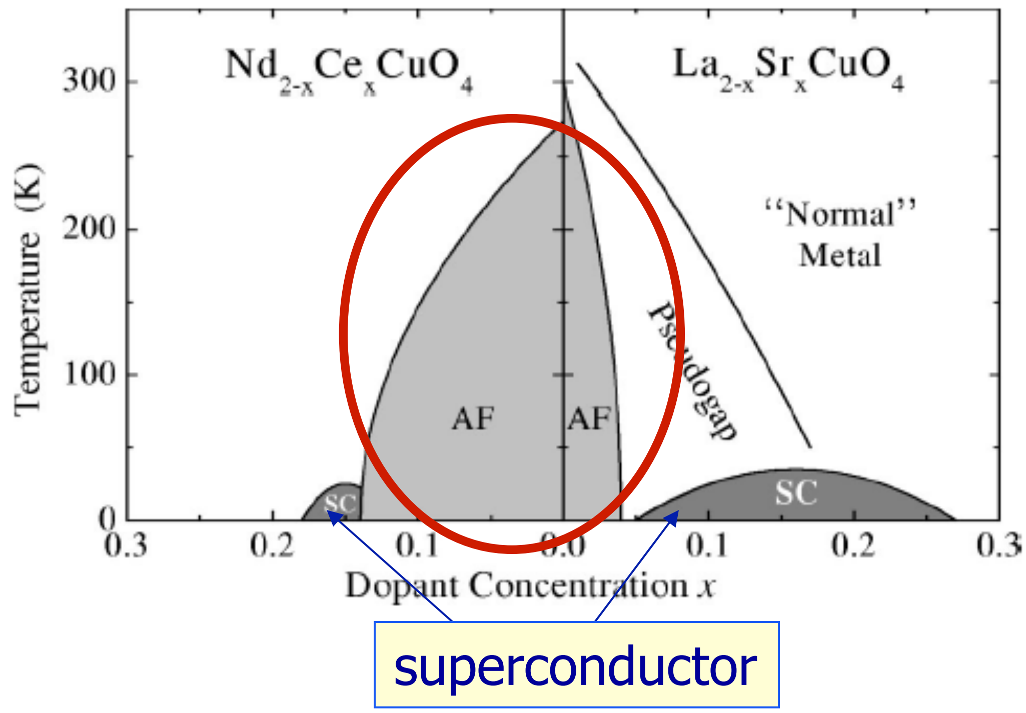


$$\Delta(\theta) = \Delta_0 \cos 2\theta$$



What is so exciting about high Tc superconductors?

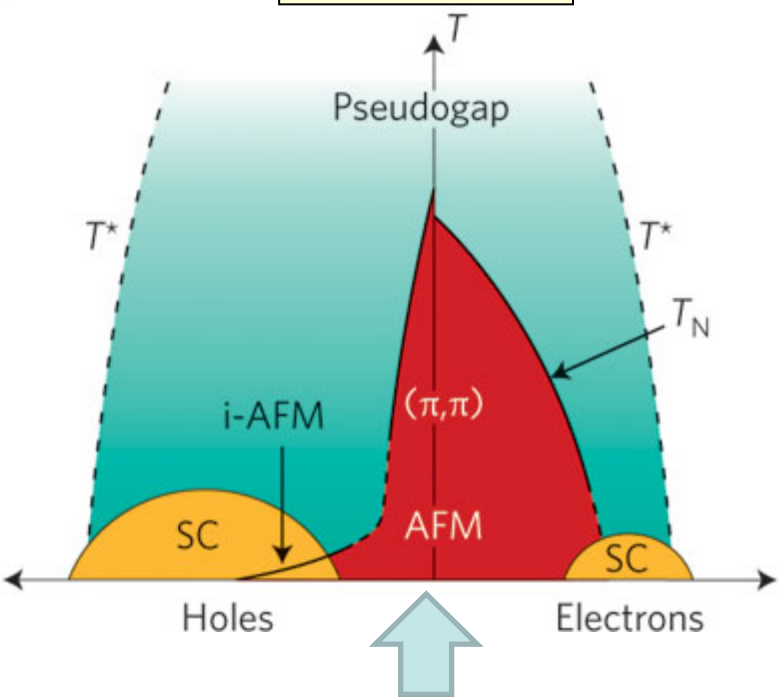
3. ... rs



Is antiferromagnetism related to superconductivity?

a

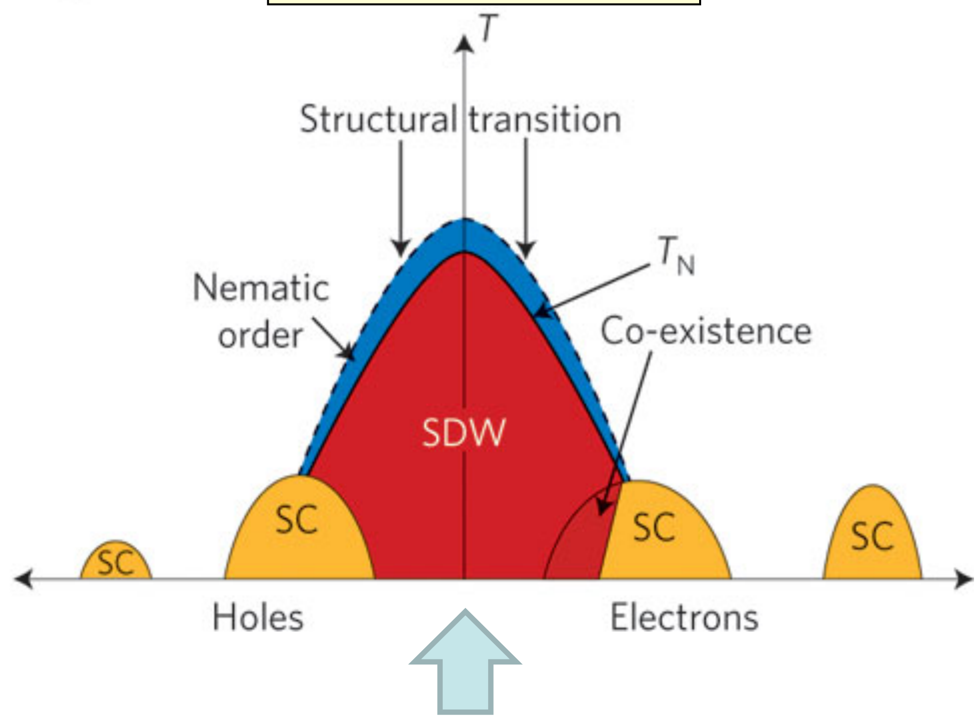
Cuprates



Parent compounds are insulators

b

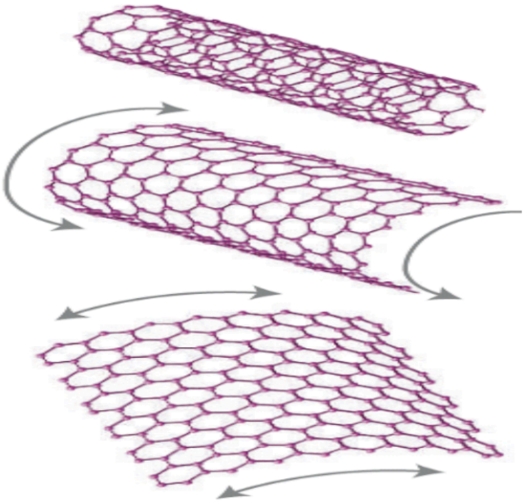
Pnictides

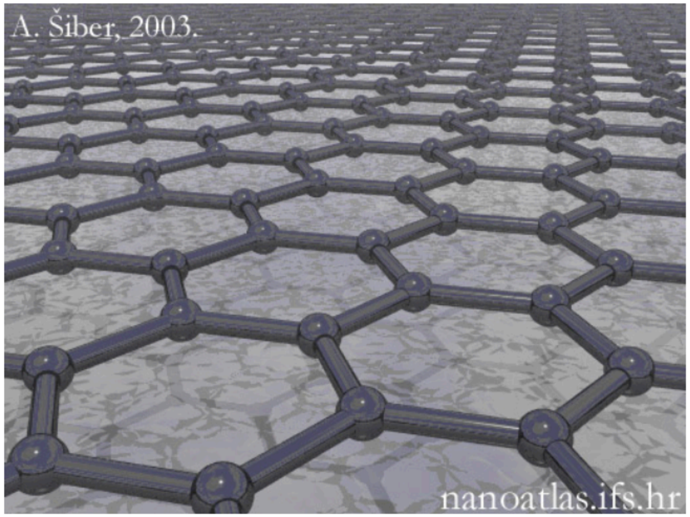


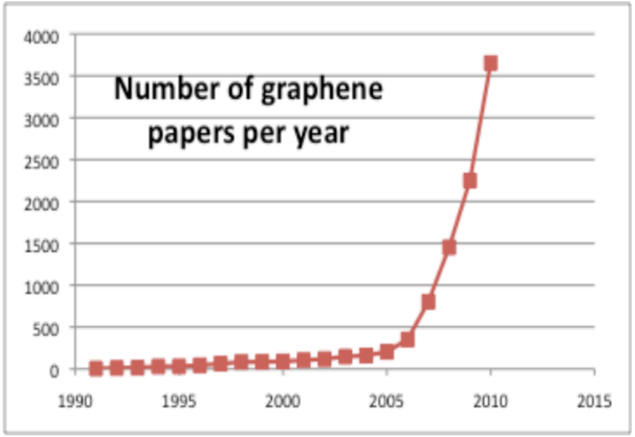
Parent compounds are metals

The first atom-thick material… graphene

Graphene is a two-dimensional layer of sp^2-bonded carbon atoms with amazing properties…



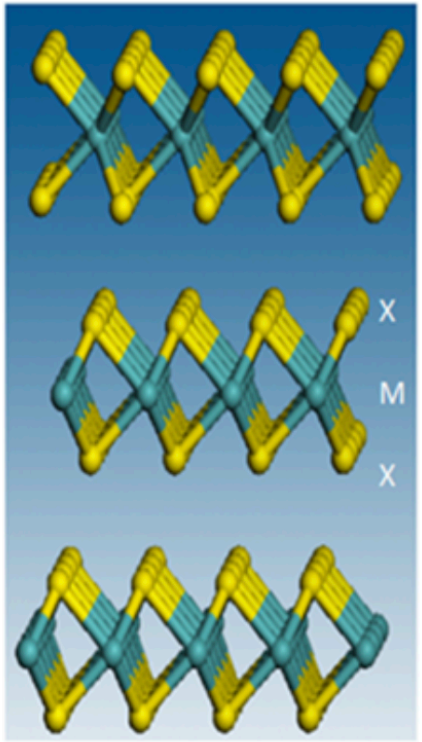




Important investments all-around the world

Graphene, a new substance coming from the world of atomic scale manipulation of matter, could be the wonder material of the 21st century. Discovering just how important this material will be for information and communication technologies (ICTs) is the main focus of the Seventh Framework Programme (FP7) Coordination Action GRAPHENE-CA, funded under the 'Ideas' Theme.

Graphene is not the only 2-D Material…



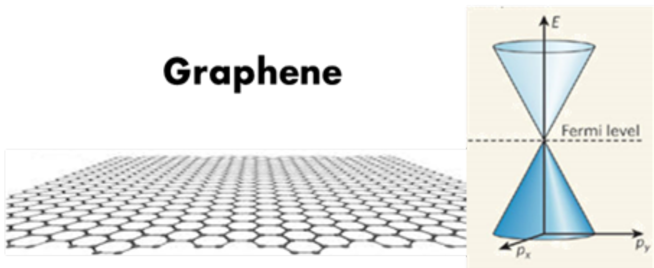
M	- S2	-Se2	-Te2
Ti	1.95(D),0.3(I)	1.55(D),0.15(I)	1.00(D),Semi-metal
Zr	1.68(D),2.10(I)	1.20(D), 1.61(I)	
Hf	2.7(D),1.93(I)	1.77(D),1.18(I)	Semi-metal(-0.4)
V	Metal	Metal	Semi-metal
Nb	Metal	Metal	Metal
Ta	Metal	Metal	Semi-metal
Mo	1.8(SL), 1.72(D),1.2(I)	1.49(SL), 1.38(D), 1.1(I)	1.13(SL), semi-metal
W	1.93(SL), 1.77(D),1.35(I)	1.60(D),1.1(I)	Semi-metal

D: direct bandgap, I:inderect bandgap, SL: single-layer bandgap

Diversity of 2D Atomic Layers

Semi-metal

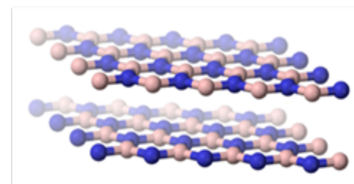
Graphene

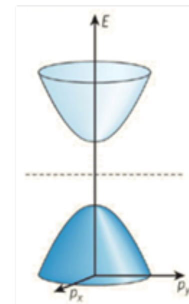


Insulator

Transition metal oxides

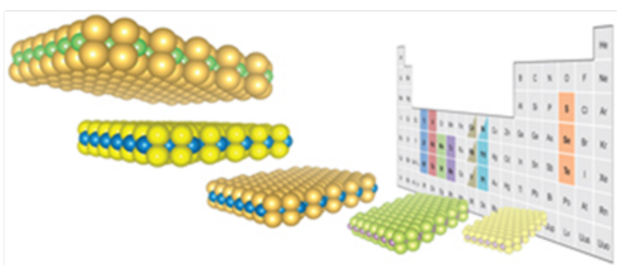
Boron nitride



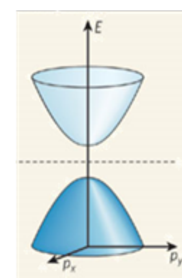


Semiconductor

Transition metal dichalcogenides (TMD)
e.g. MoS_2, $MoSe_2$, WS_2, WSe_2 ...

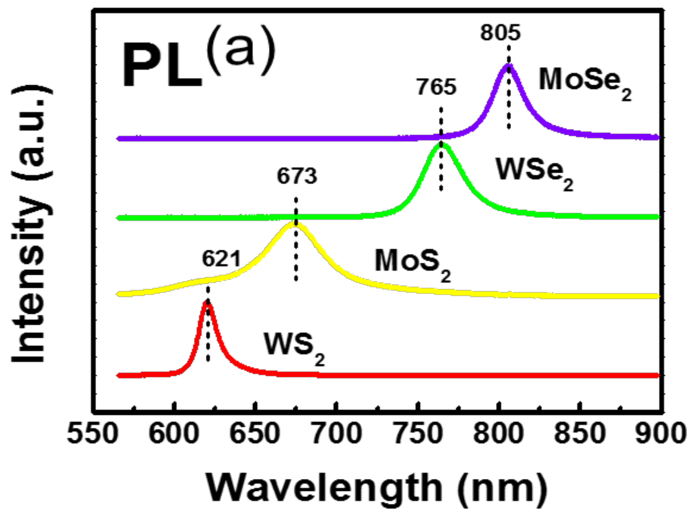


Black phosphorus



Ideal Bandgap

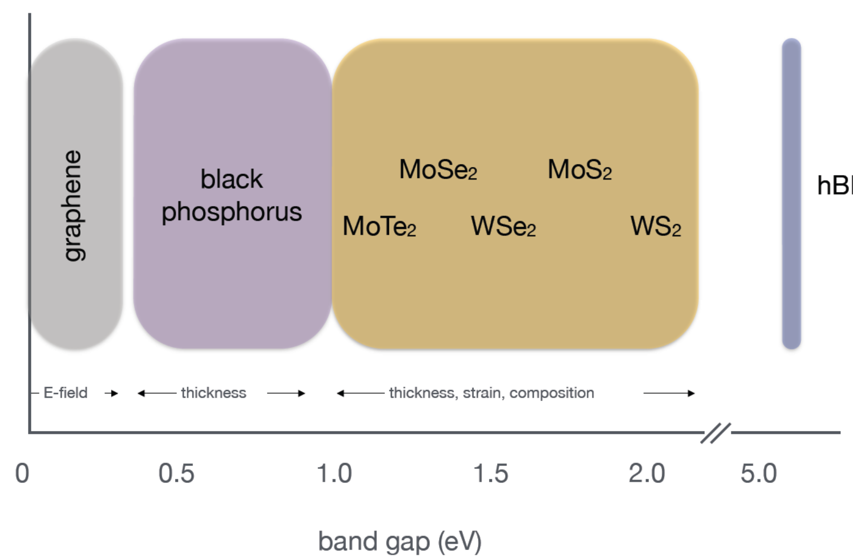
Direct Bandgap



Ref: Sam Yang, Y.H. Lee*, ***VLSI*** *(2016)*

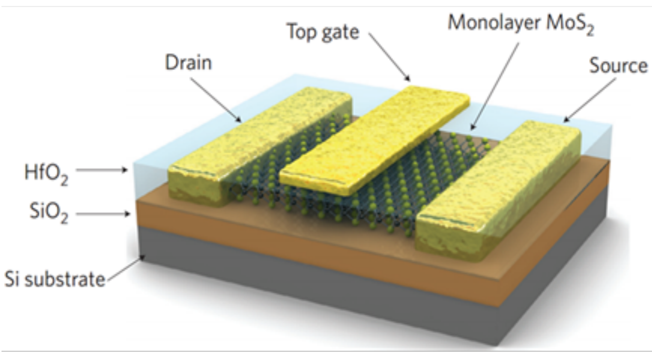
Bandgap Engineering

- Materials System
- Thickness
- Strain
- Composition



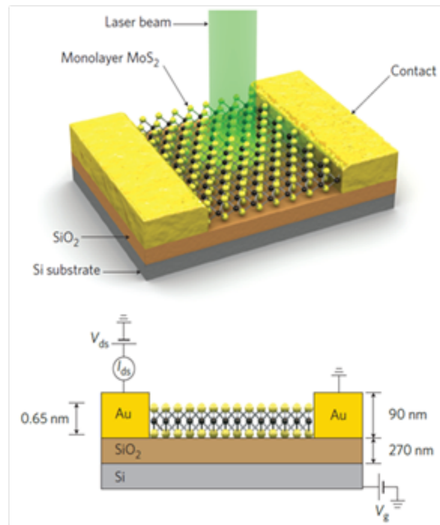
Prominent Properties

FET



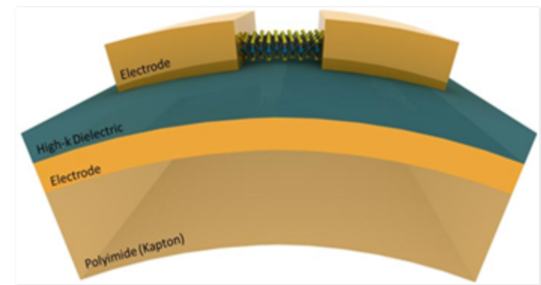
Nature Nanotech. **6**, 147–150 (2011).

Photodetector



Nature Nanotech. 8, 497–501 (2013).

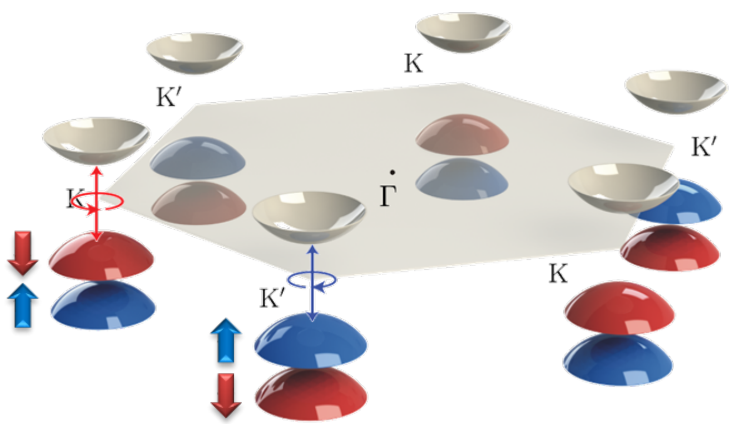
Flexible electronics device



ACS Nano. **2013**, 6, 5446-5452

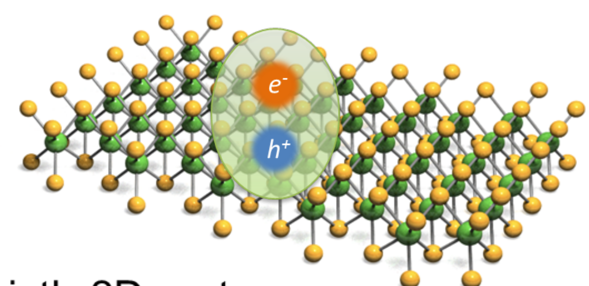
Novel Physics

Optically controlled spin-valley states

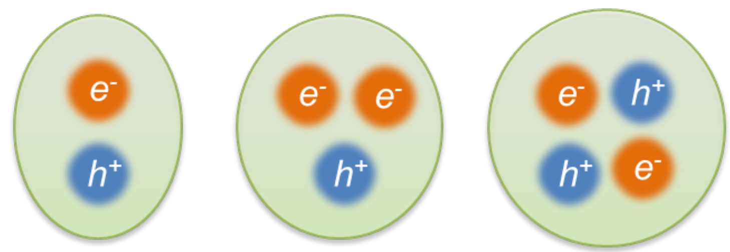


Spintronics and valleytronics

Many-body interactions



Strictly 2D system
→ strong Coulomb interactions



	exciton	trion	bi-exciton
E_b :	>500 meV (Bolotin)	>20 meV (Heinz)	>20 meV (Gedik)

Understand the

macroscopic electronic properties

and the role of

competing degrees of freedom

↓

Study the low-energy electronic excitations

↓

ARPES

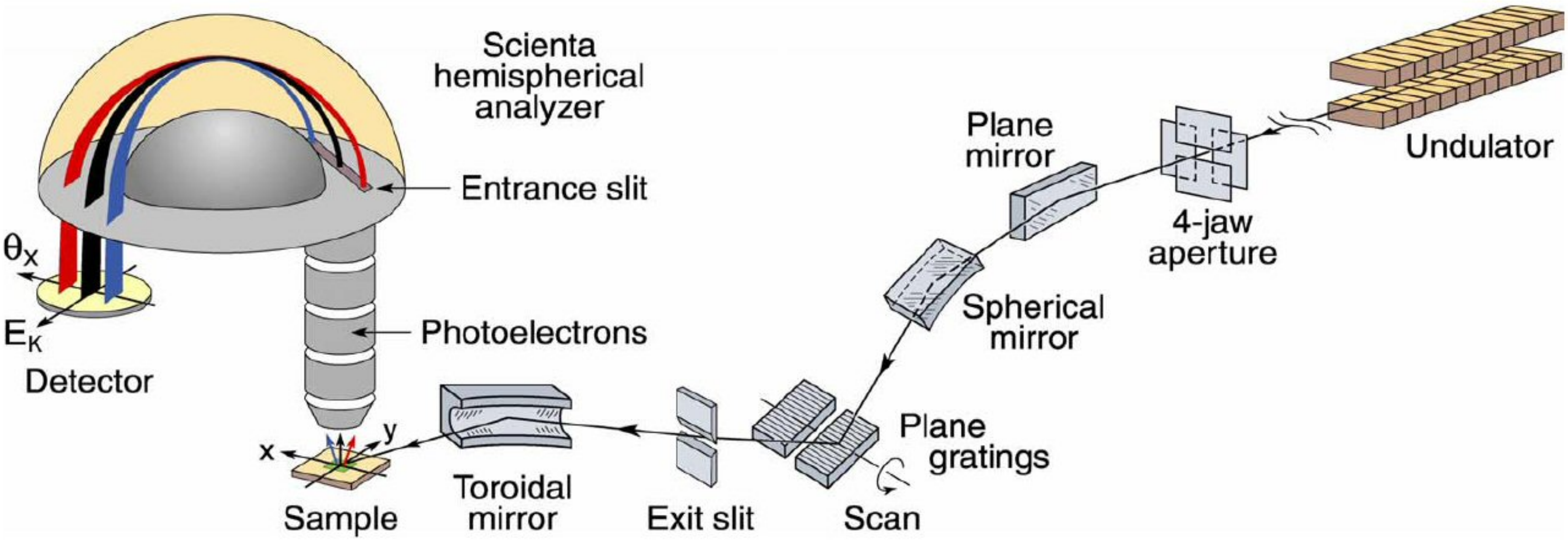
Velocity and direction of the electrons in the solid

Angle-Resolved
Photo-Emission Spectroscopy
(ARPES)

Working of ARPES

- An atomically flat sample is illuminated by a beam of monochromatic light.
- Due to the photoelectric effect, the sample emits electrons.
- The kinetic energy and direction of these electrons are measured by the rotatable spectrometer.
- The obtained data are used to map out the Fermi surface of the sample material.

ARPES setup



Parallel multi-angle recording

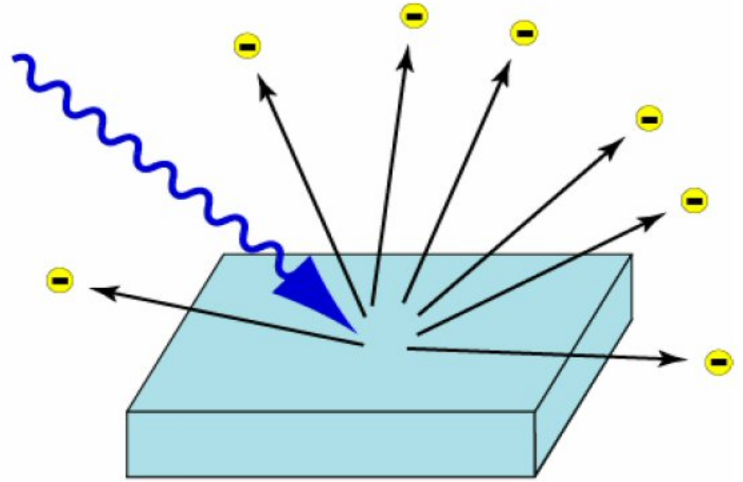
- Improved **energy resolution**
- Improved **momentum resolution**
- Improved **data-acquisition efficiency**

	ΔE (meV)	$\Delta\theta$
past	*20-40*	*2°*
now	*2-10*	*0.2°*

Photoelectric Effect

- Explained by Einstein (1905):

$$E_{k_{\max}} = hf - \phi$$



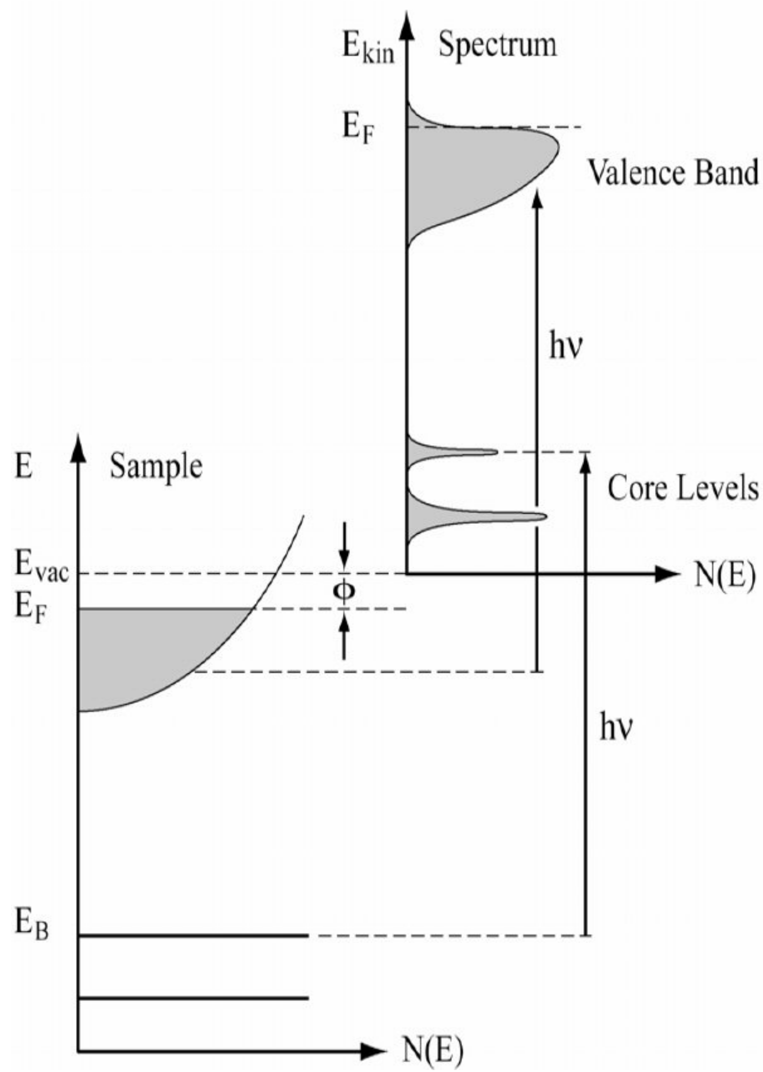
- More generally,

$$E_k = hf - \phi - |E_B|$$

where E_B is the binding energy of the electron.

Photoemission Spectra

- The work function is known/ measurable.
- The photon energy is known.
- We can calculate the energy of the electron in the solid!

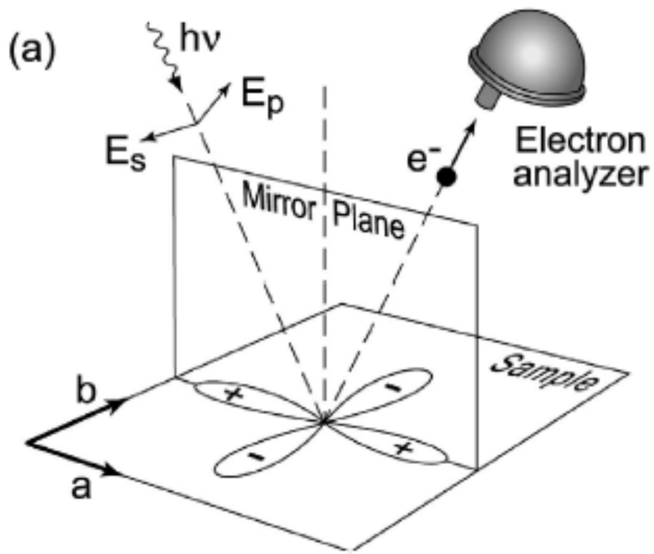


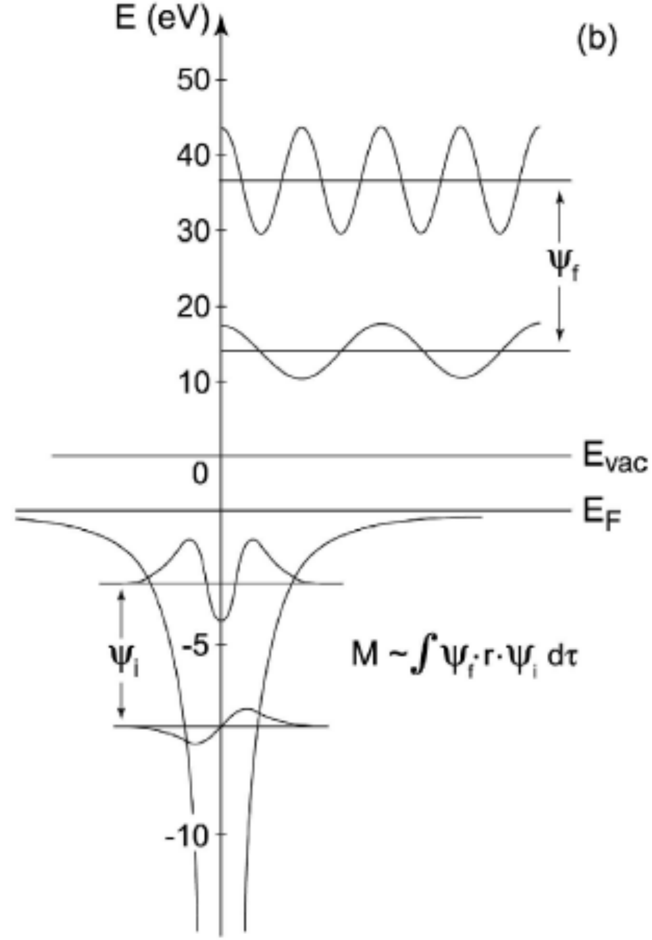
Basis of ARPES

ARPES is directly measuring the components of electron momentum that are parallel to the surface

- The flat surface of the sample has translational symmetry. Therefore, as electrons escape from the solid, linear momentum is conserved parallel to the surface.

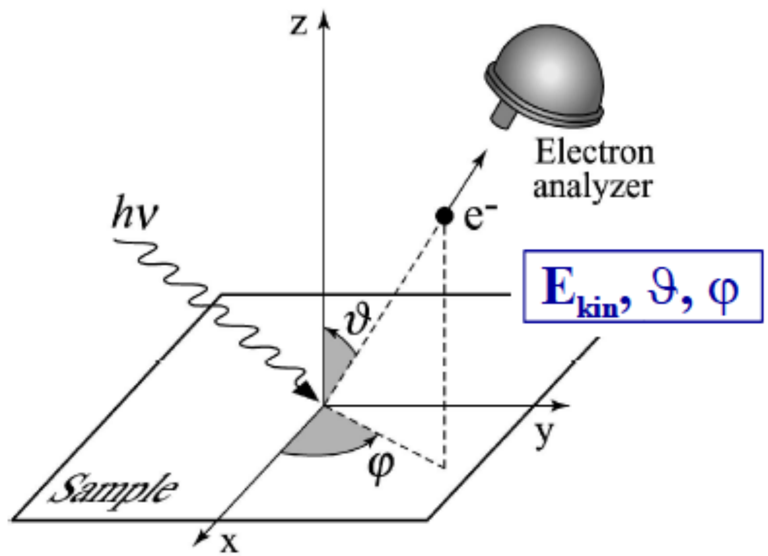
- The photon momentum is small and can be neglected.





$$w_{fi} = \frac{2\pi}{\hbar} \left| \langle \Psi_f^N | H_{int} | \Psi_i^N \rangle \right|^2 \delta(E_f^N - E_i^N - h\nu)$$

$$H_{int} = -\frac{e}{2mc}(\mathbf{A}\cdot\mathbf{p} + \mathbf{p}\cdot\mathbf{A}) = -\frac{e}{mc}\mathbf{A}\cdot\mathbf{p}$$

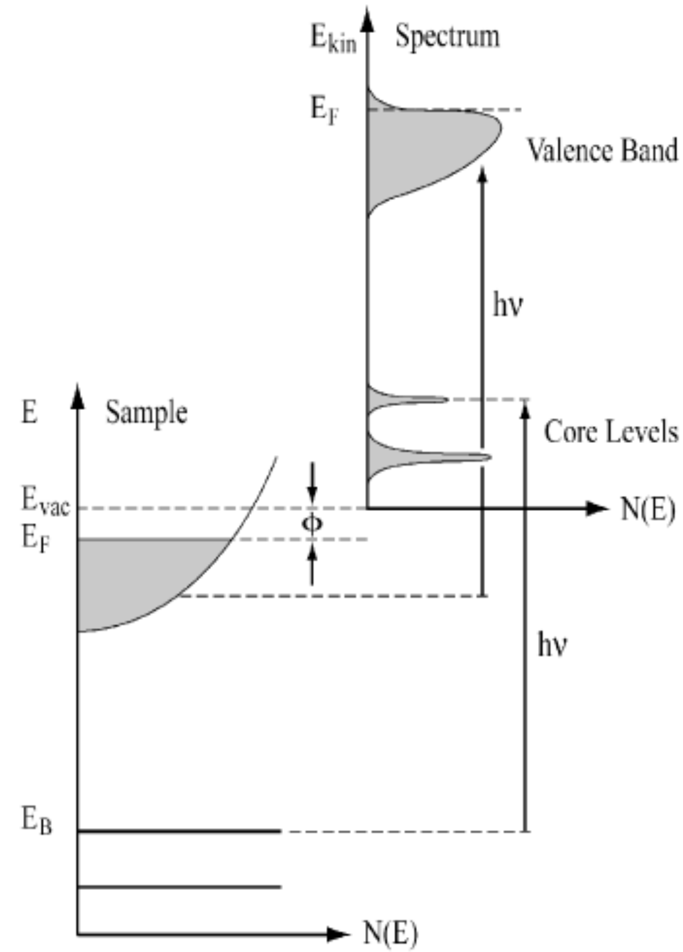


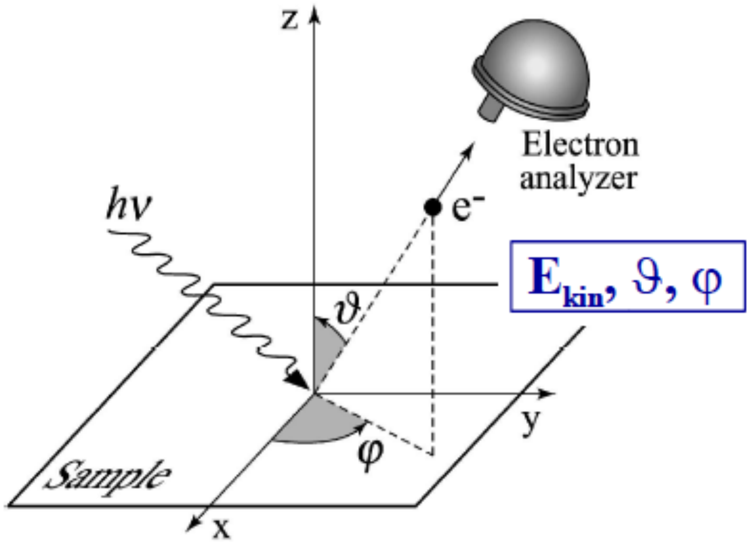
Energy Conservation

$$E_{kin} = h\nu - \phi - |E_B|$$

Momentum Conservation

$$p_{\parallel} = \hbar k_{\parallel} = \sqrt{2mE_{kin}} \cdot \sin\vartheta$$





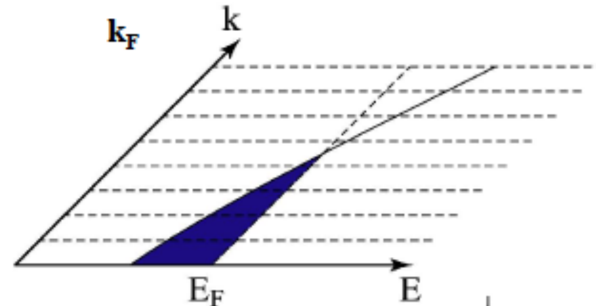
Energy Conservation

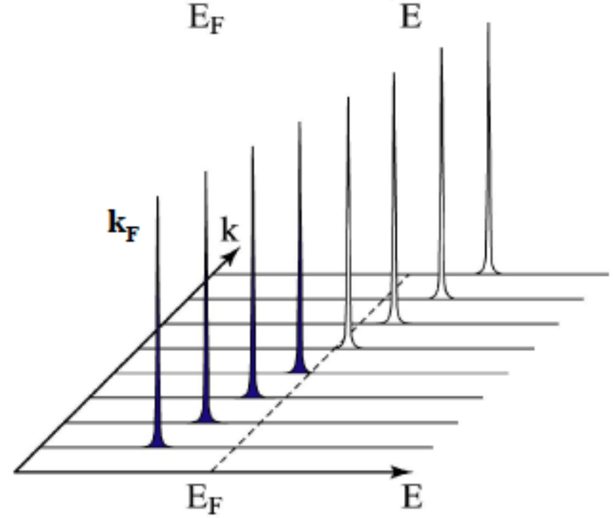
$$E_{kin} = h\nu - \phi - |E_B|$$

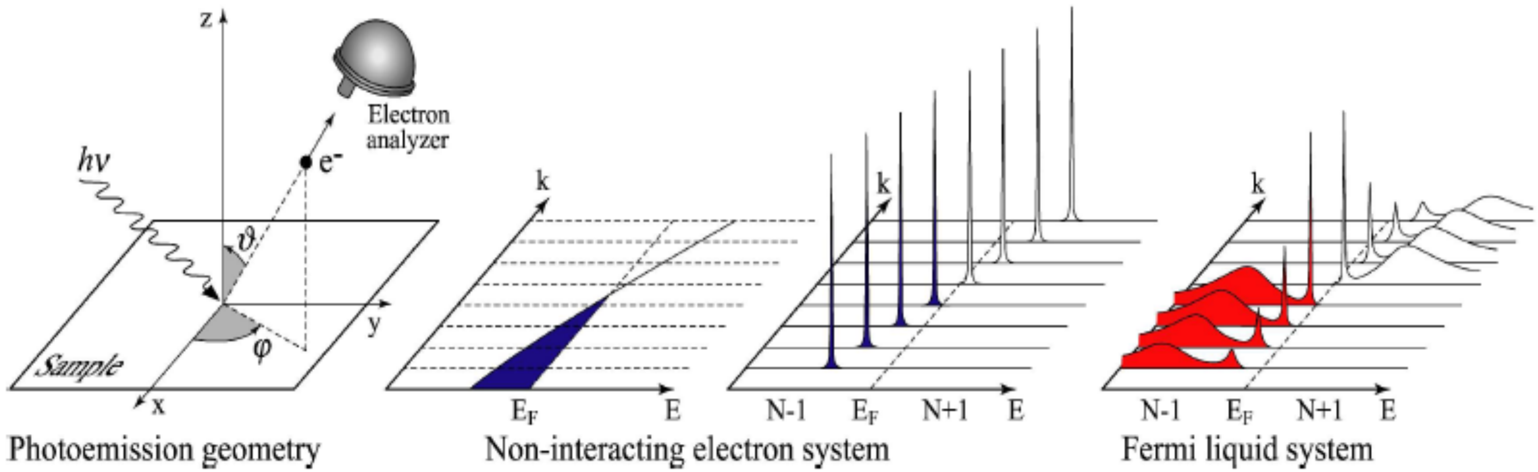
Momentum Conservation

$$\mathbf{p}_{\|} = \hbar \mathbf{k}_{\|} = \sqrt{2mE_{kin}} \cdot \sin\vartheta$$

Electrons in Reciprocal Space







Photoemission intensity: $I(k,\omega)=I_0\,|M(k,\omega)|^2 f(\omega)\,A(k,\omega)$

Single-particle spectral function

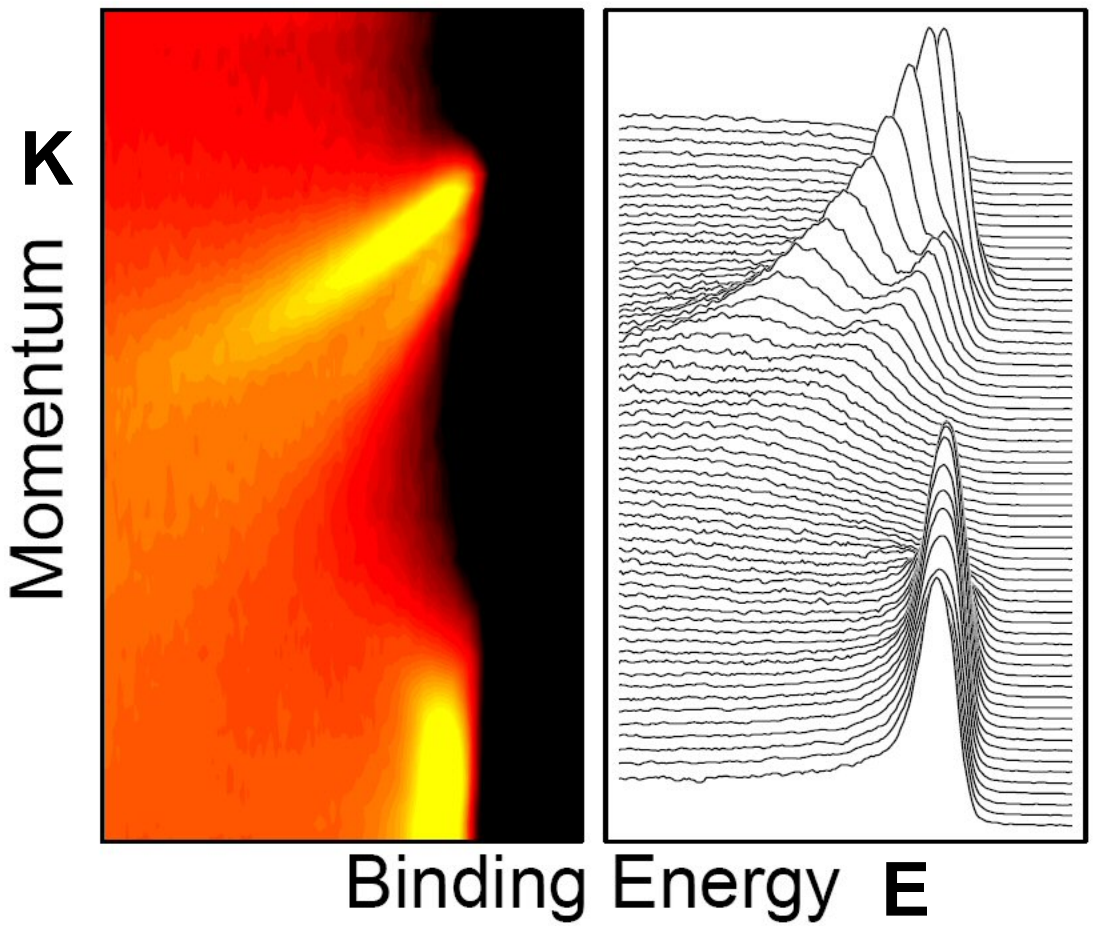
$$A(\mathbf{k},\omega) = -\frac{1}{\pi}\frac{\Sigma''(\mathbf{k},\omega)}{[\omega-\epsilon_{\mathbf{k}}-\Sigma'(\mathbf{k},\omega)]^2+[\Sigma''(\mathbf{k},\omega)]^2}$$

$\Sigma(k,\omega)$: the "self-energy" - captures the effects of interactions

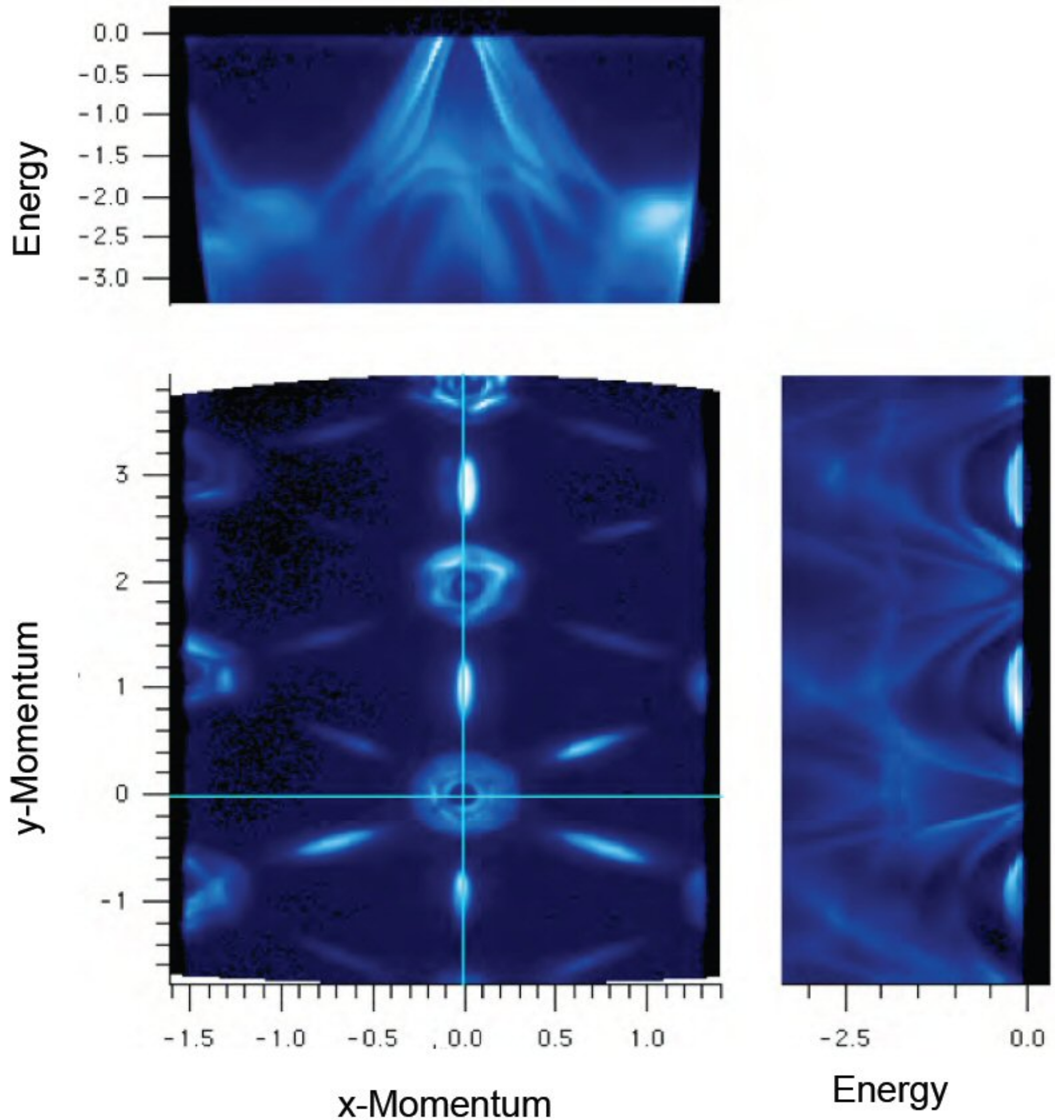
What is ARPES used for?

- ARPES is an almost ideal tool for imaging the Fermi surface of 1-D and 2-D solids.
- Since many of the high temperature superconductors are essentially 2-D materials, much of the work in this field is done using ARPES.

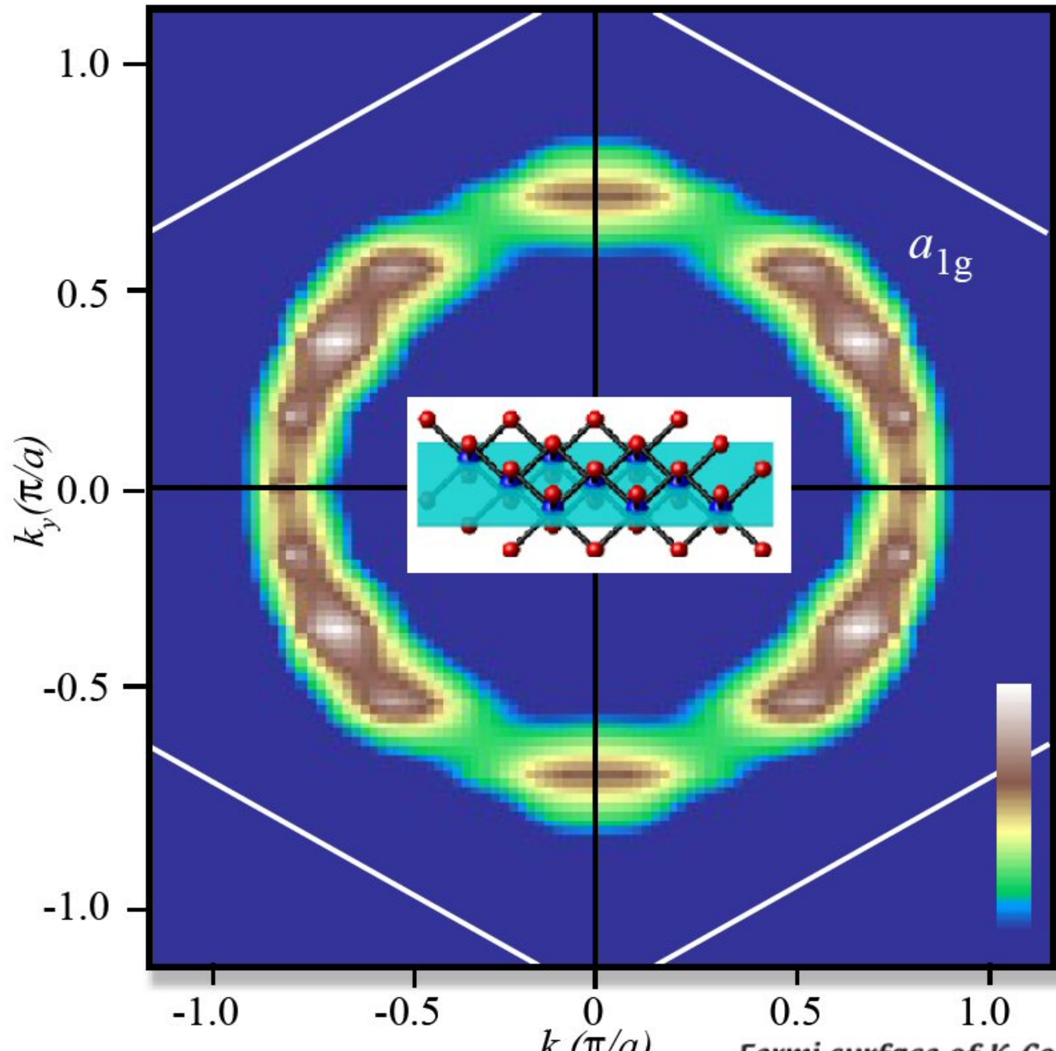
Momentum and Binding Energy



Direct *k* Space Imaging

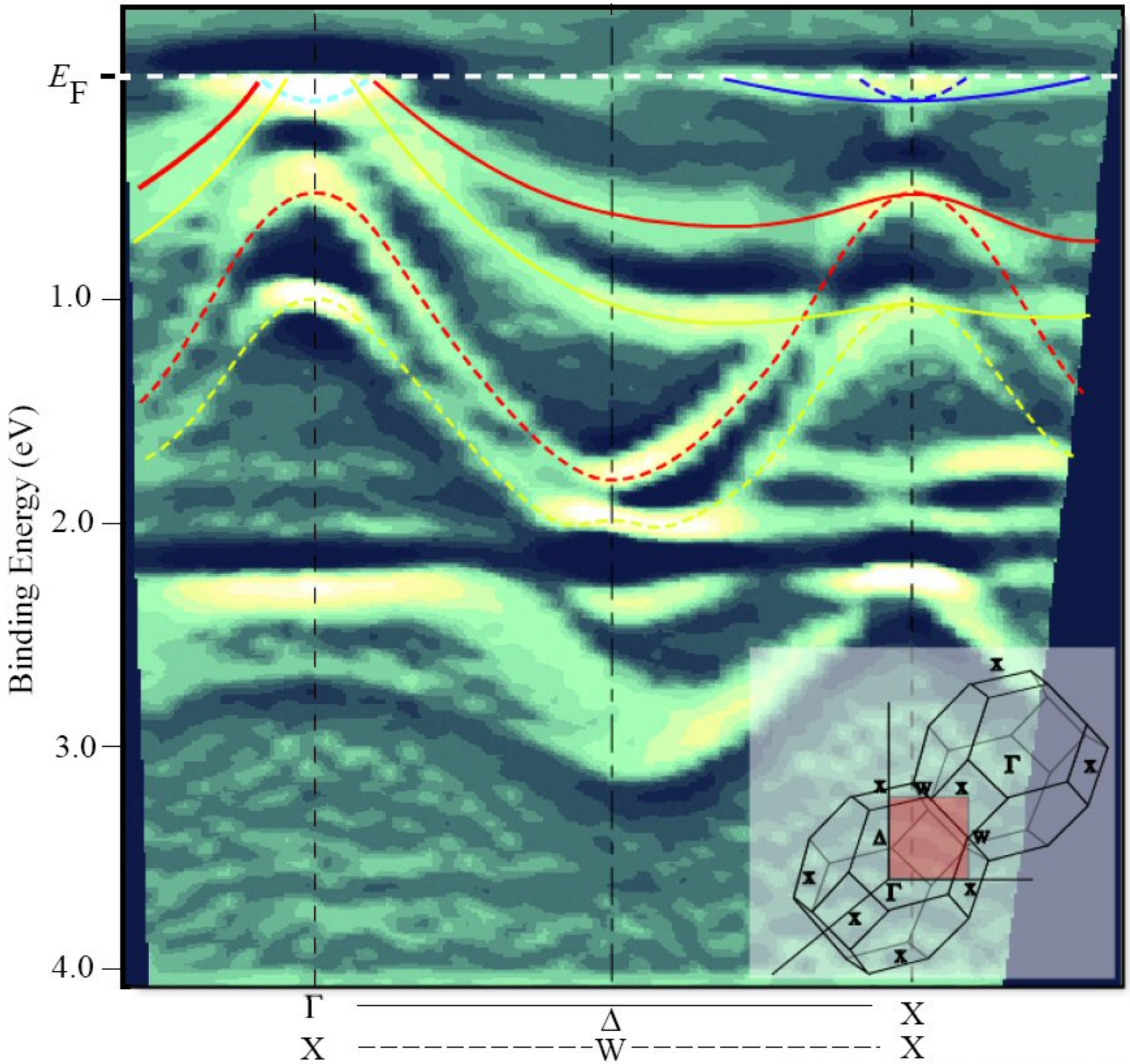


Fermi Surface Images



Fermi surface of K_xCoO_2

Band Structure Images

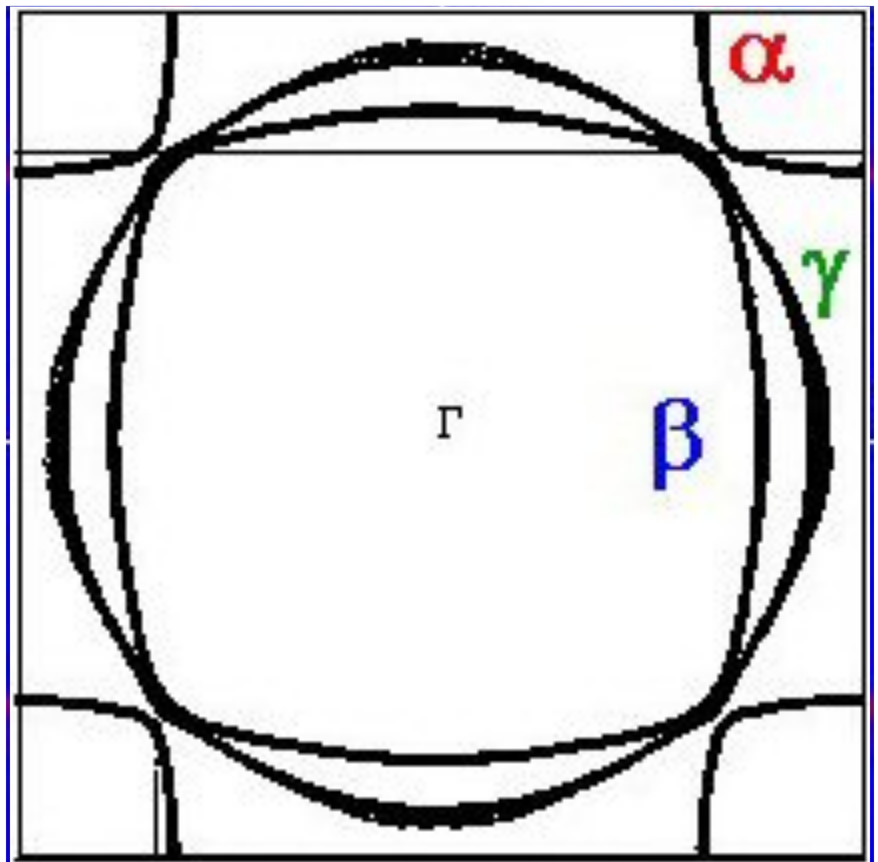


Band structure of CeSb

Validation of Predictions

Sr_2RuO_4 : ARPES Measurement

Sr_2RuO_4 : Theoretical Calculation



Advantages

• **Direct information about electronic states!**

• Straightforward comparison with theory - little or no modelling.

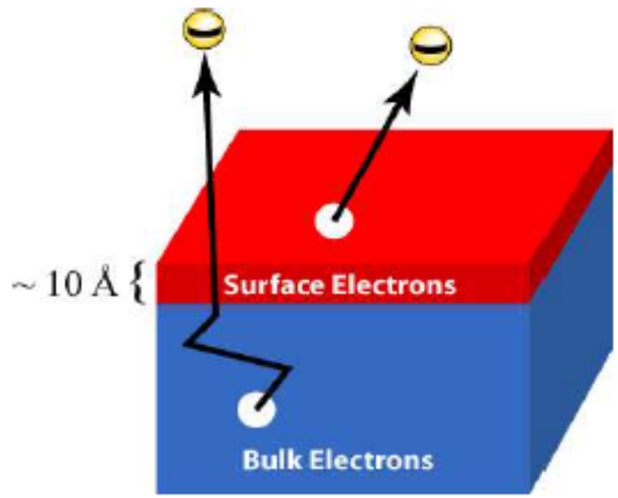
• High-resolution information about **BOTH energy** and **momentum**

• **Surface-sensitive probe**

• Sensitive to "many-body" effects

• Can be applied to small samples (100 μm x 100 μm x 10 nm)

Limitations



• **Not bulk sensitive**

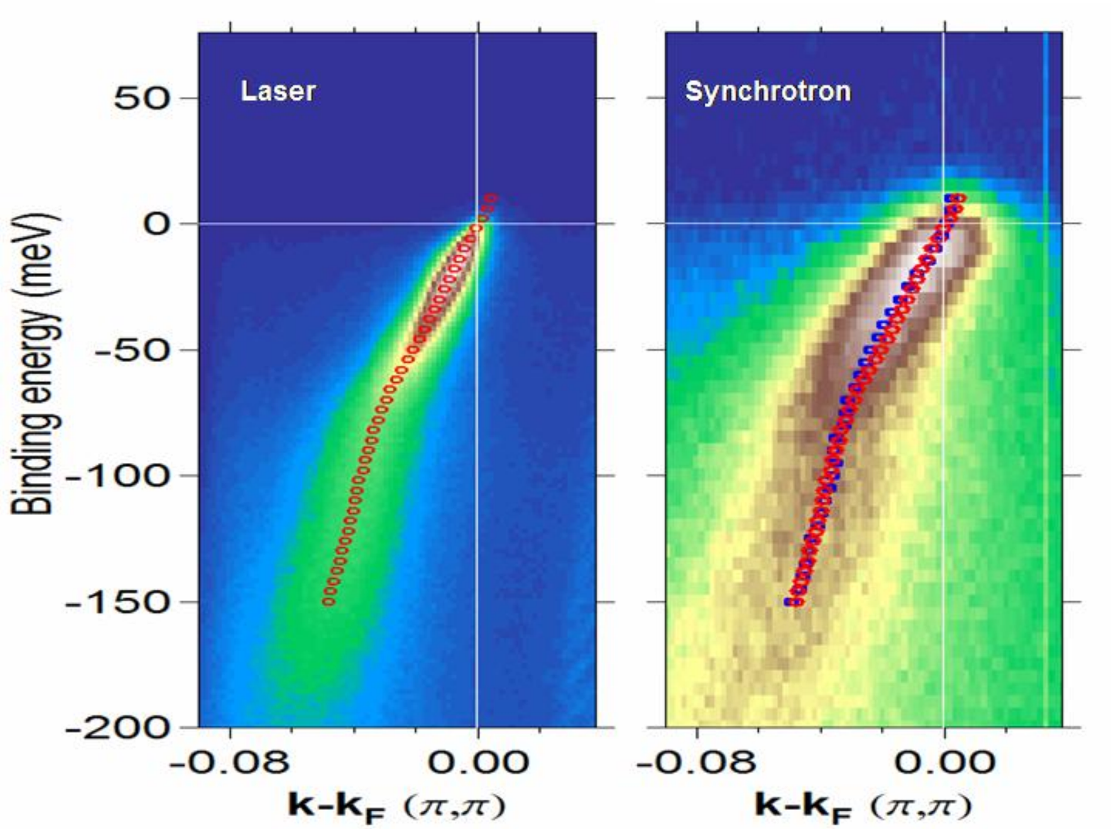
• Requires clean, atomically flat surfaces in **ultra-high vacuum**

• Cannot be studied as a function of pressure or magnetic field

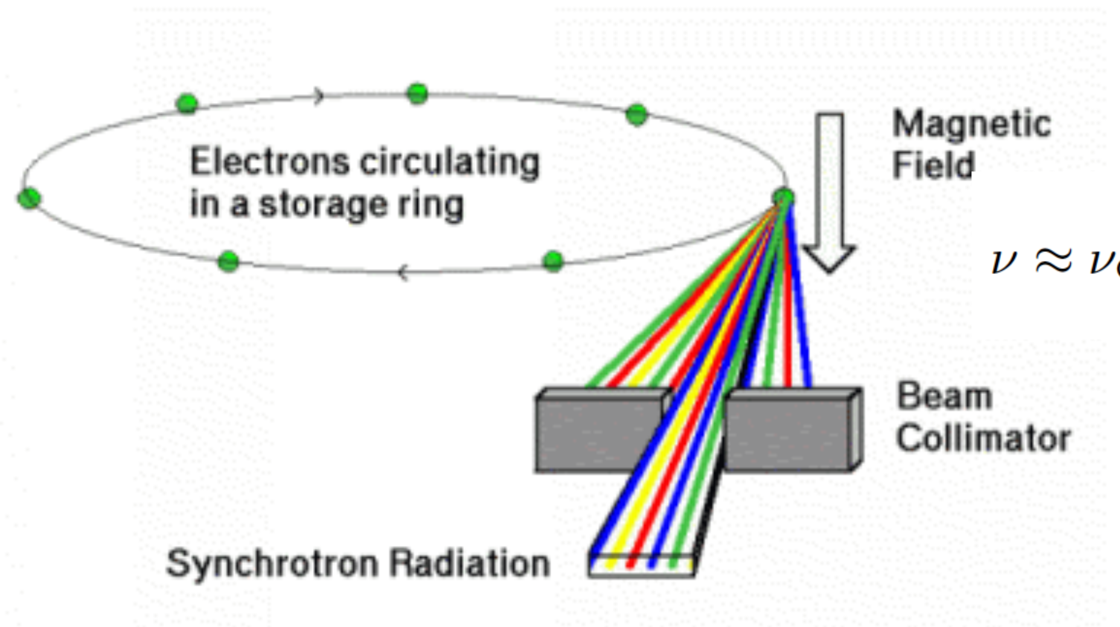
Further Advances

- Laser ARPES: lower energy means sharper pictures

(image of $Bi_2Sr_2CaCu_2O_8$ in "nodal" direction)



Why we need synchrotron radiation



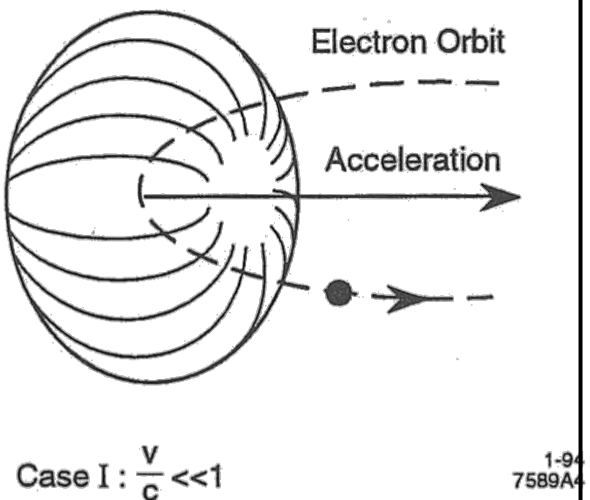
$$\nu \approx \nu_{\mathrm{c}} \approx \gamma^2 \nu_{\mathrm{g}} = \left(\frac{E}{m_{\mathrm{e}} c^2}\right)^2 \nu_{\mathrm{g}};$$

$$\nu_{\mathrm{g}} = \frac{eB}{2\pi m_{\mathrm{e}}}.$$

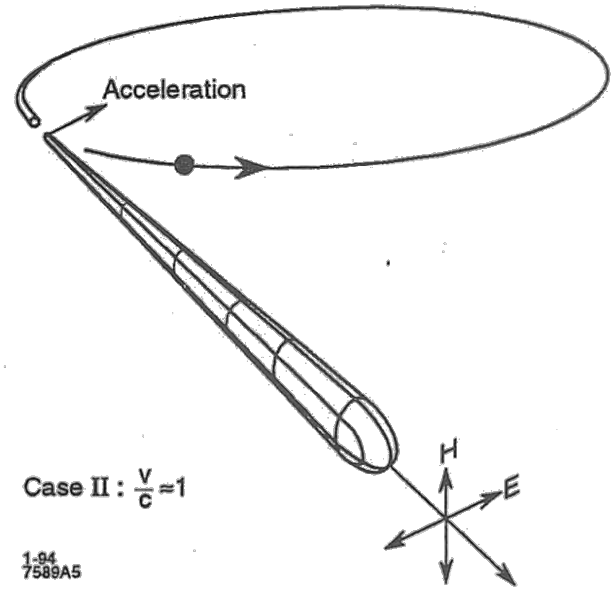
Synchrotron radiation is electromagnetic radiation emitted when charged particles are radially accelerated (move on a curved path).

Synchrotron Radiation

- Radiated power increases at higher velocities
- Radiation becomes more focused at higher velocities



At low electron velocity (non -relativistic case) the radiation is emitted in a non-directional pattern



When the electron velocity approaches the velocity of light, the emission pattern is folded sharply forward. Also the radiated power goes up dramatically

How a storage ring light source works

Parts of Synchrotron

(1) Electron gun

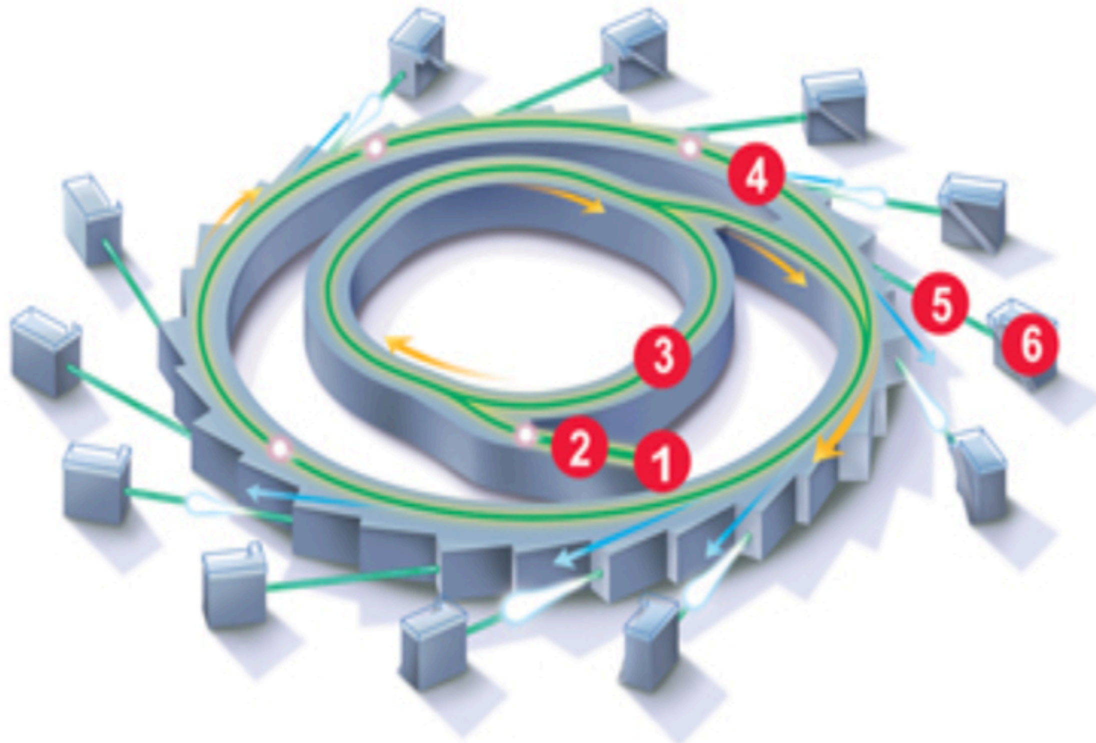
(2) LINAC

(3) Booster ring

(4) Storage ring

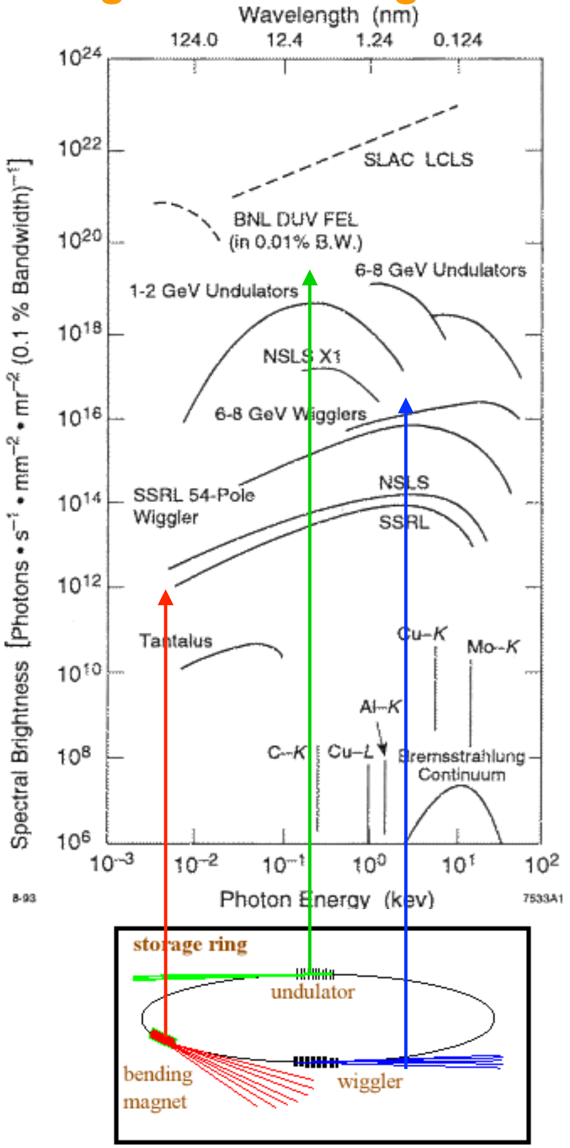
(5) Beamline

(6) End station

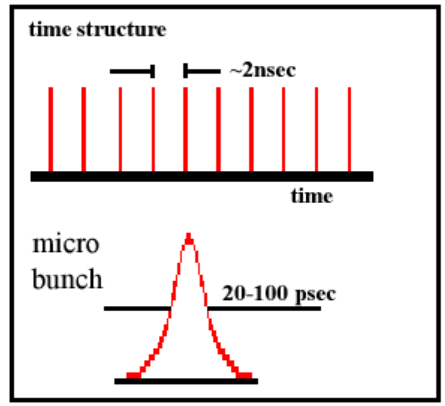


Synchrotron Radiation - Basic Properties

High flux and brightness



Pulsed time structure



Broad spectral range

Polarized (linear, elliptical, circular)

Small source size

Partial coherence

High stability

Flux = $\dfrac{\text{\# of photons in given } \Delta\lambda/\lambda}{\text{sec}}$

Brightness = $\dfrac{\text{\# of photons in given } \Delta\lambda/\lambda}{\text{sec, mrad } \theta\text{, mrad } \varphi\text{, mm}^2}$

(a measure of concentration of the radiation)

Basic Properties of Synchrotron Radiation

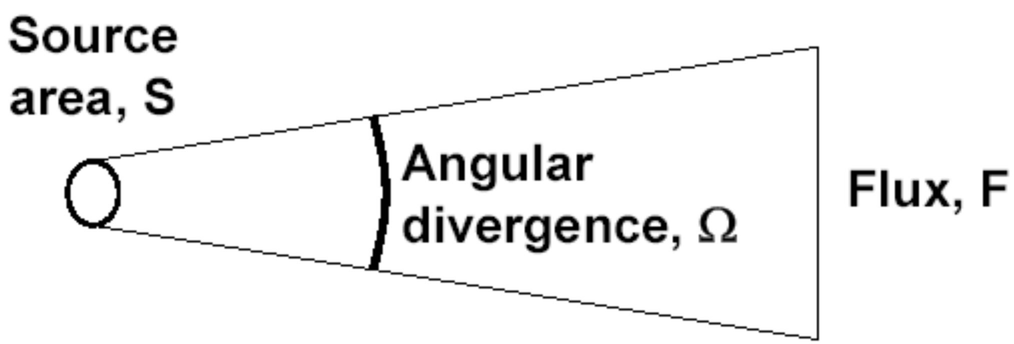
1. **HIGH FLUX, BRIGHTNESS, STABILITY**
2. **BROAD SPECTRAL RANGE - Tunability**
3. **POLARIZATION (linear, elliptical, circular)**
4. **PULSED TIME STRUCTURE (0.01 - 1 nsec)**
5. **SMALL SOURCE SIZE ($\leq$ mm)**
6. **PARTIAL COHERENCE**

The brightness of a light source

$$\textbf{Flux} = \frac{\text{\# of photons in given } \Delta\lambda/\lambda}{\text{sec}}$$

$$\textbf{Brightness} = \frac{\text{\# of photons in given } \Delta\lambda/\lambda}{\text{sec, mrad } \theta\text{, mrad } \varphi\text{, mm}^2}$$

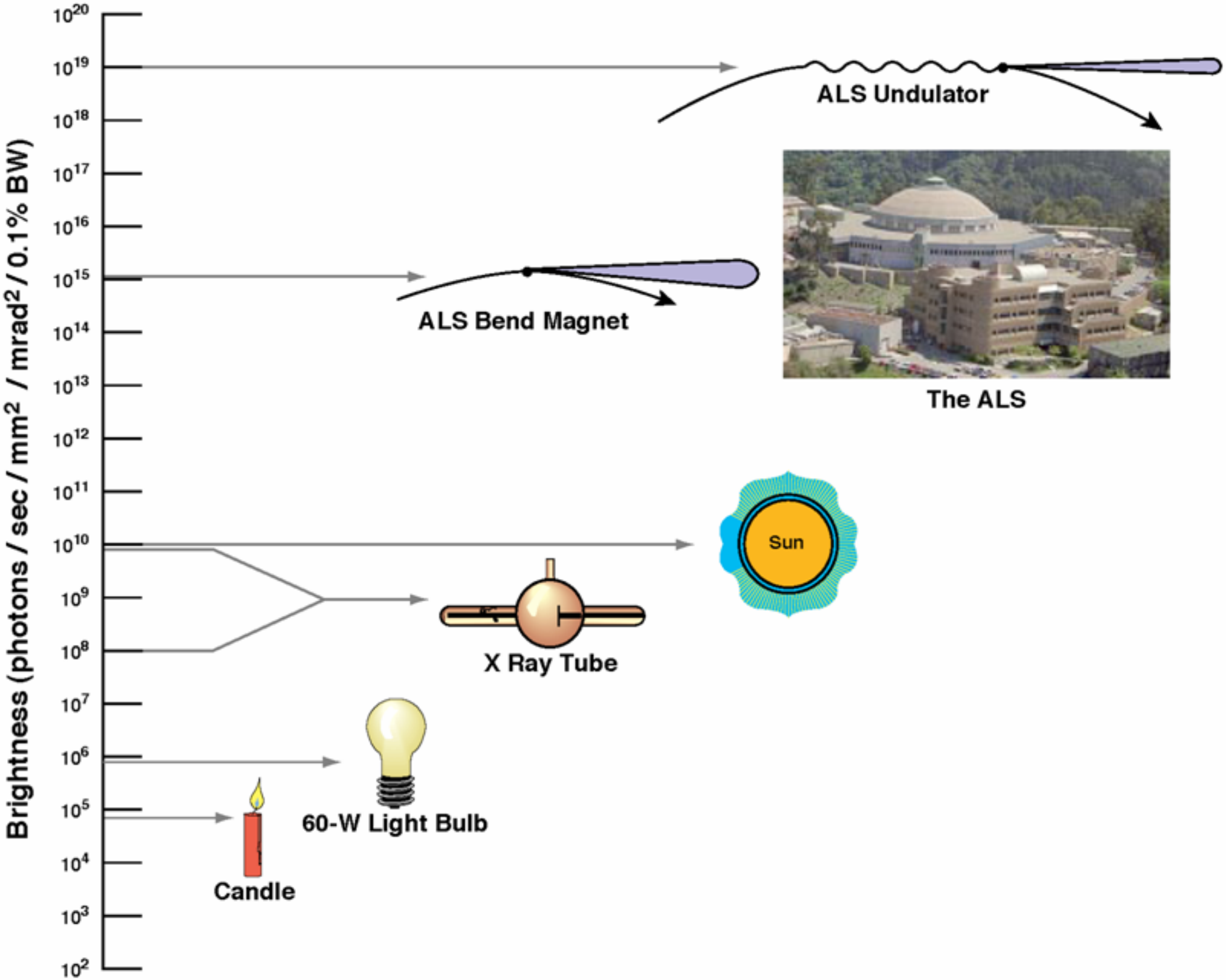
(a measure of concentration of the radiation)



$$\textbf{Brightness} = \text{constant} \times \frac{F}{S \times \Omega}$$

How Bright Is the Advanced Light Source?

ALS



Synchrotron Radiation Facilities Around the World

- **54 in operation in 19 countries used by more than 20,000 scientists**

  (Brazil, China, India, Korea, Taiwan, Thailand)

- **8 in construction**

  Armenia, Australia, China, France, Jordan, Russia, Spain, UK

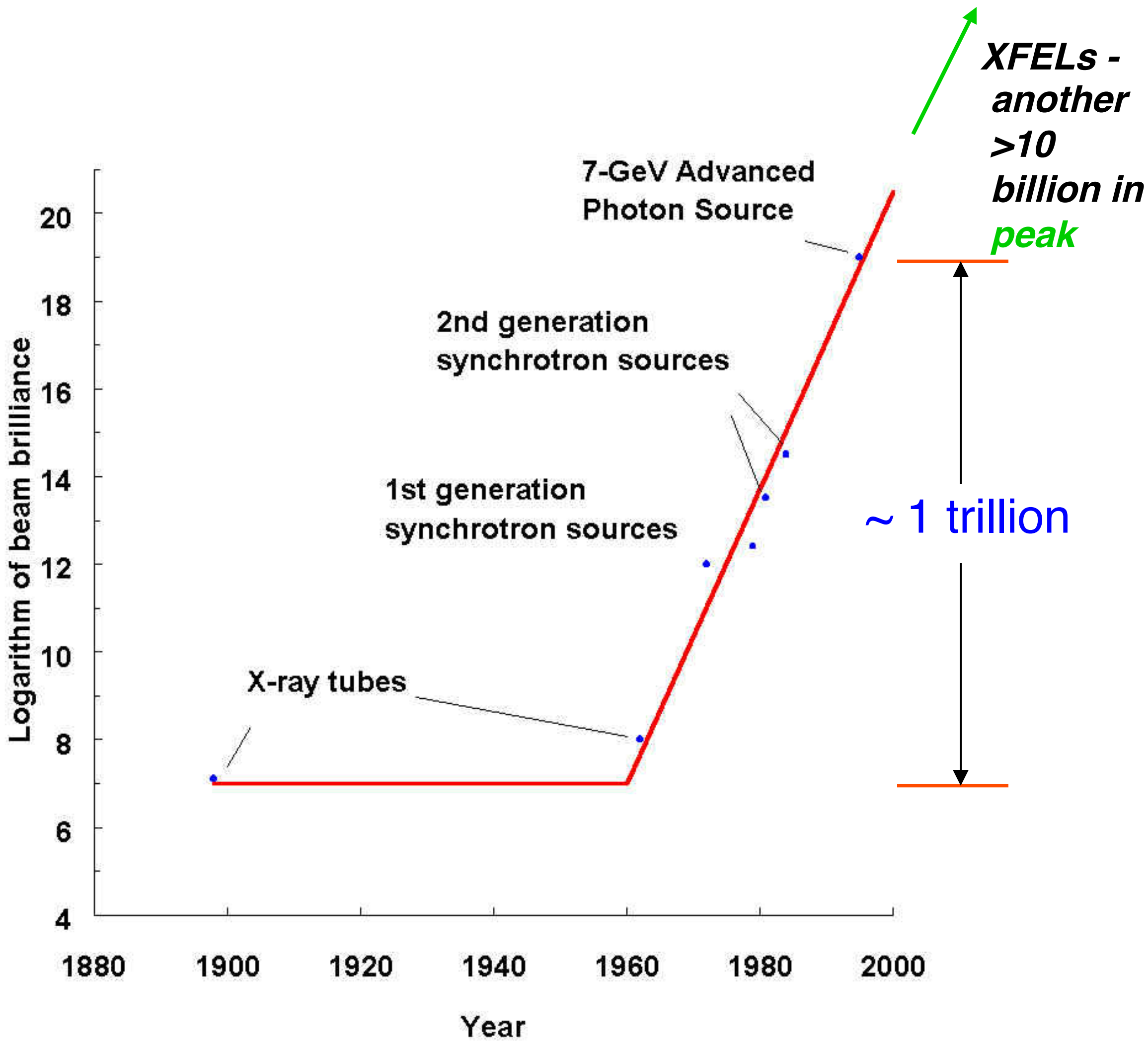
- **11 in design/planning**

For a list of SR facilities around the world see

http://ssrl.slac.stanford.edu/SR_SOURCES.HTML

www.sesame.org.jo

Steep growth in brightness

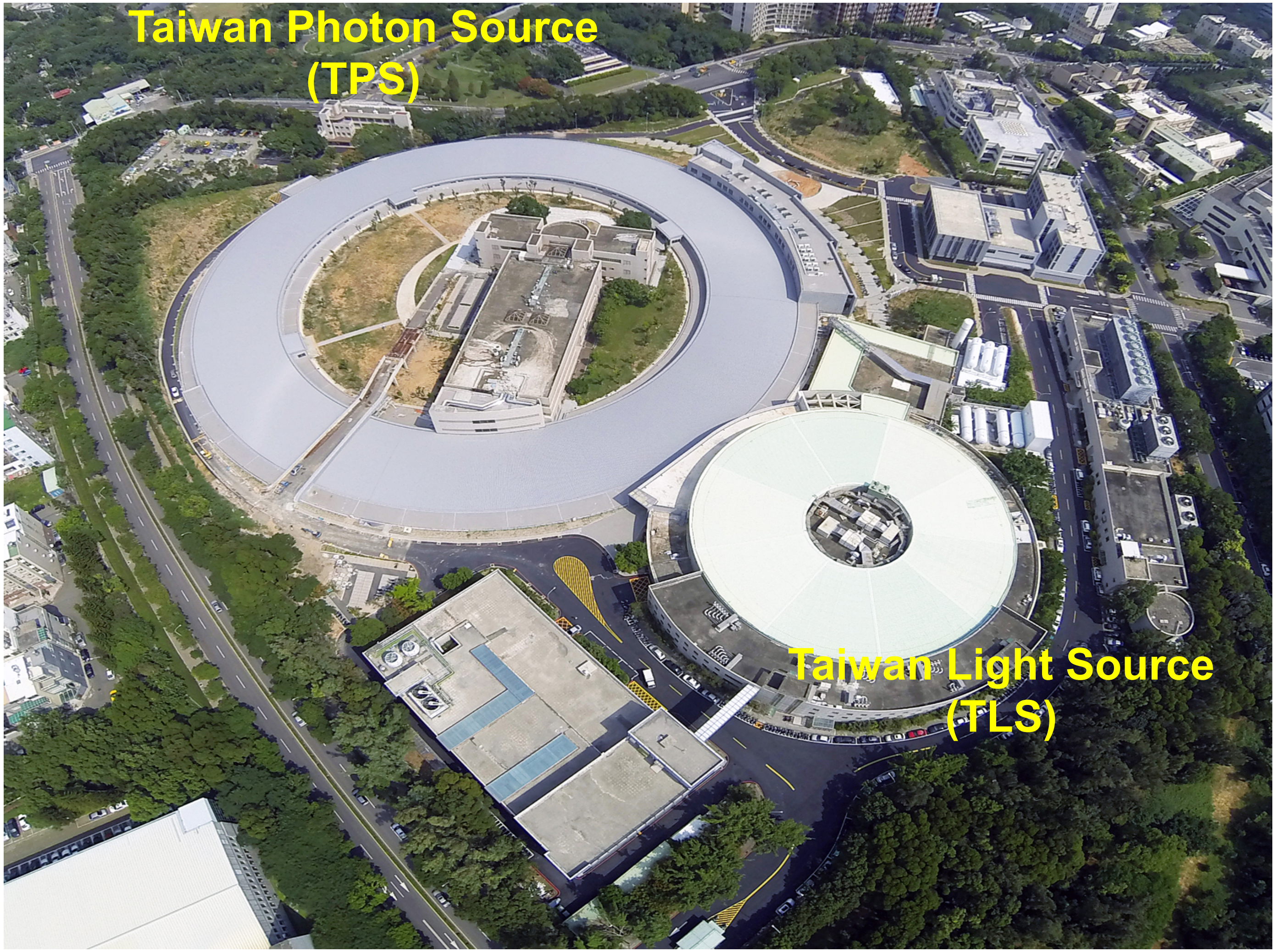


Future of Synchrotron Radiation

- Higher Brightness
  - Free Electron Lasers
- Shorter Pulse Lengths
  - Femto (10^{-12}) and Attosecond (10^{-15})
- Terahertz (T-rays)
  - Coherent Synchrotron Radiation

Taiwan Photon Source (TPS) – Hsinchu, Taiwan

http://www.nsrrc.org.tw



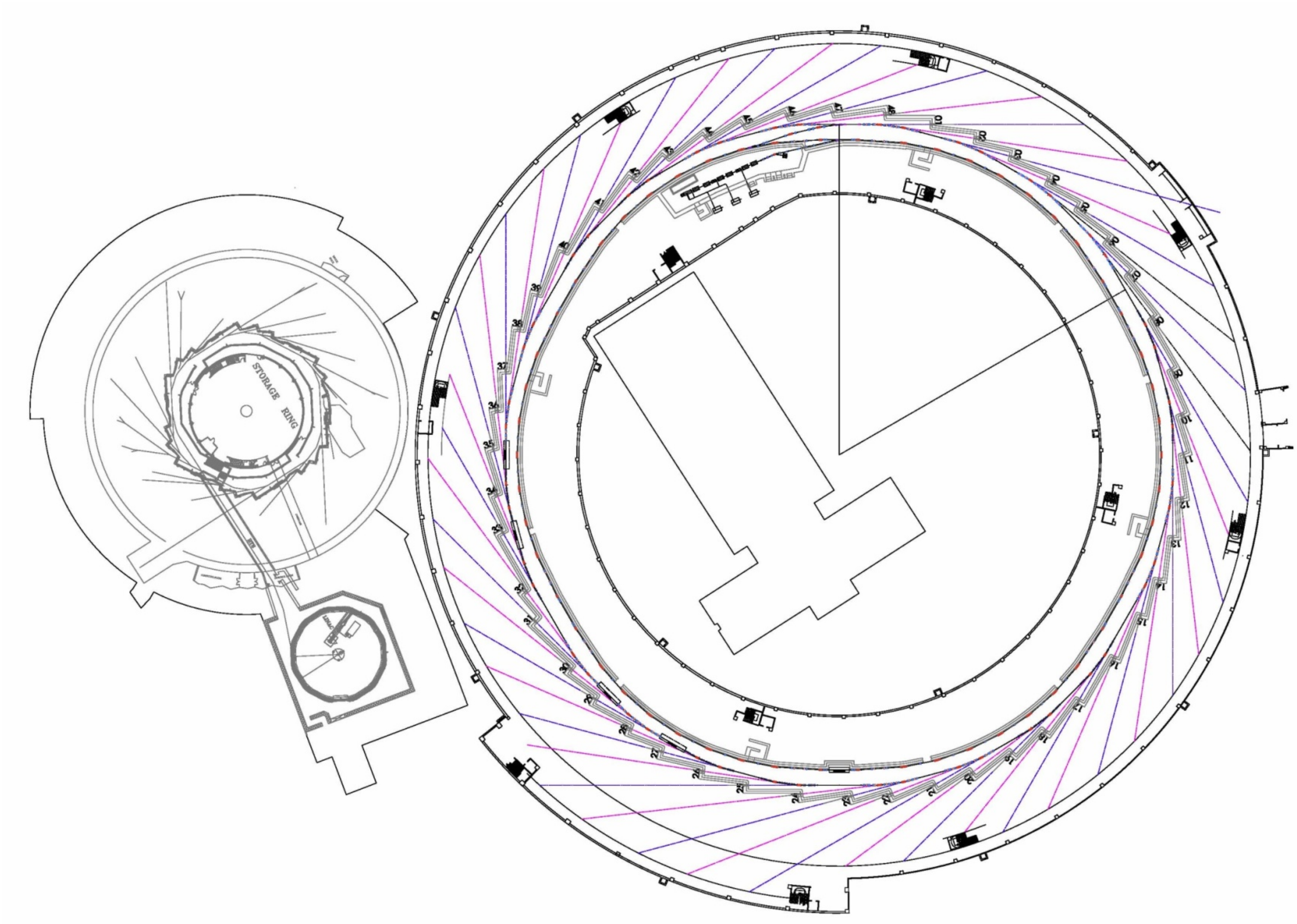
Taiwan Photon Source (TPS) – Hsinchu, Taiwan (continued)

Major Parameters of Taiwan Photon Source

Energy	3 GeV (maximum 3.3 GeV)
Current	500 mA at 3 GeV (Top-up injection)
SR circumference	518.4 m (h = 864 = $2^5 \cdot 3^3$, dia.= 165.0 m)
BR circumference	496.8 m (h = 828 = $2^2 \cdot 3^2 \cdot 23$, dia.= 158.1 m)
Lattice	24-cell DBA
Straight sections	12 m x 6 (σ_v = 12 μm, σ_h = 160 μm) 7 m x 18 (σ_v = 5 μm, σ_h = 120 μm)
Bending magnets	48
Emittance	1.6 nm·rad at 3 GeV (Distributed dispersion)
Coupling	1 %
RF frequency	500 MHz
RF gap voltage	2.8~3.5 MV (3 SRF cavities)
RF power	750 kW (3 SRF cavities)
Location	No. 101, Hsin-Ann Road, Hsinchu, Taiwan
Building	Outer diameter 210 m ; Inner diameter 129 m

Taiwan Photon Source (TPS) – Hsinchu, Taiwan (continued)

TPS & TLS Lattice Diagram



Taiwan Photon Source (TPS) – Hsinchu, Taiwan (continued)

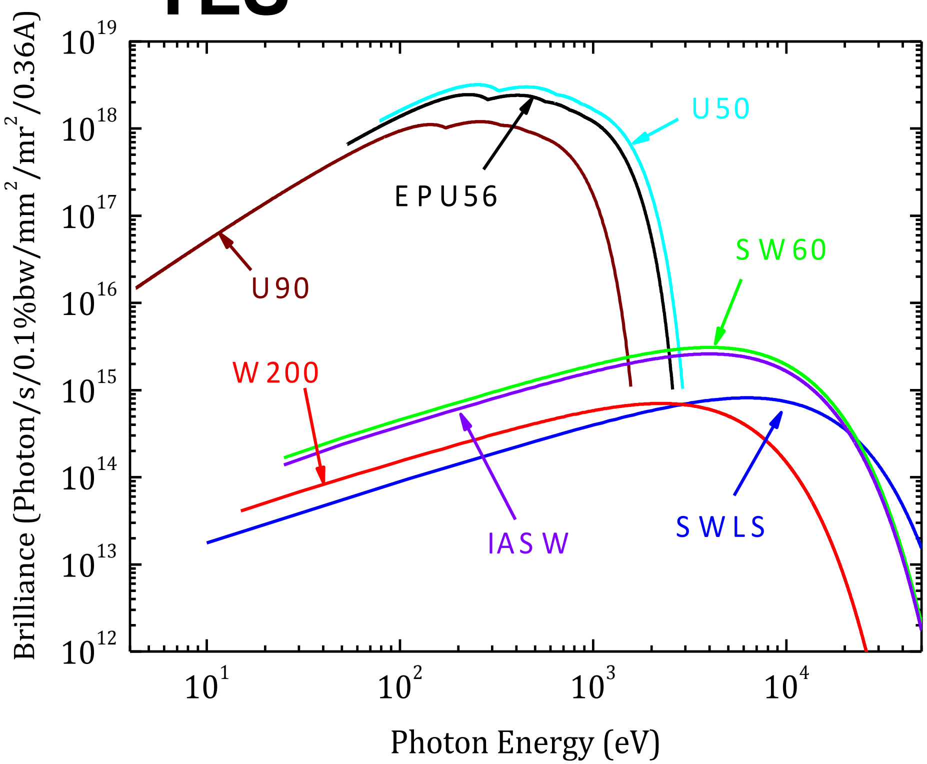
Brightness Comparison of TLS and TPS

The X-ray spectrum (photon energy 8 keV~70 keV)：

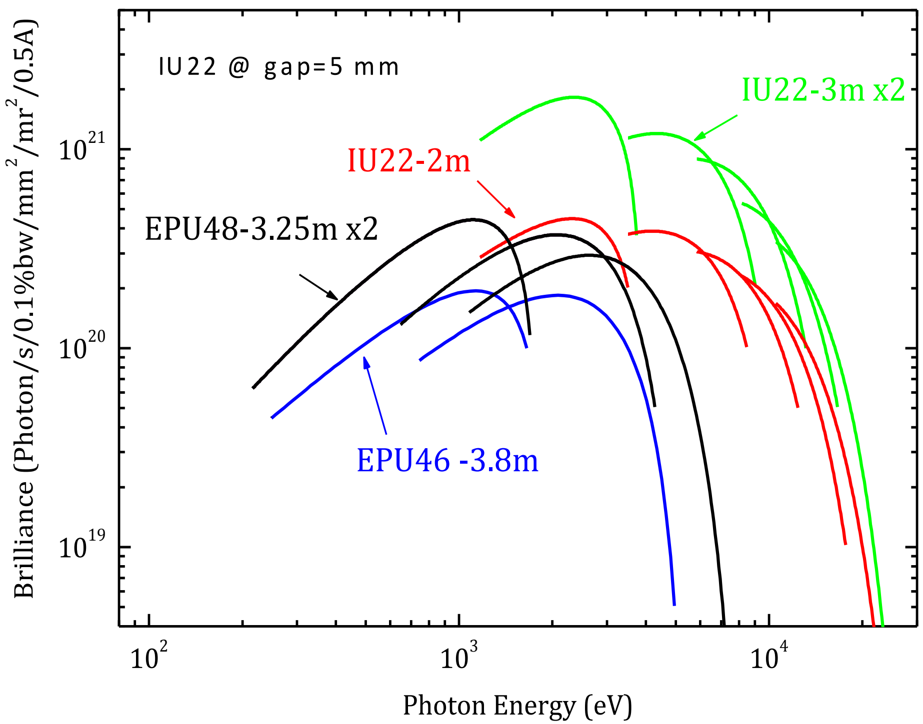
the brilliance of bending magnet increases by $>10^2$.

the brilliance of bending IDs increases by 4~6 orders of mag.

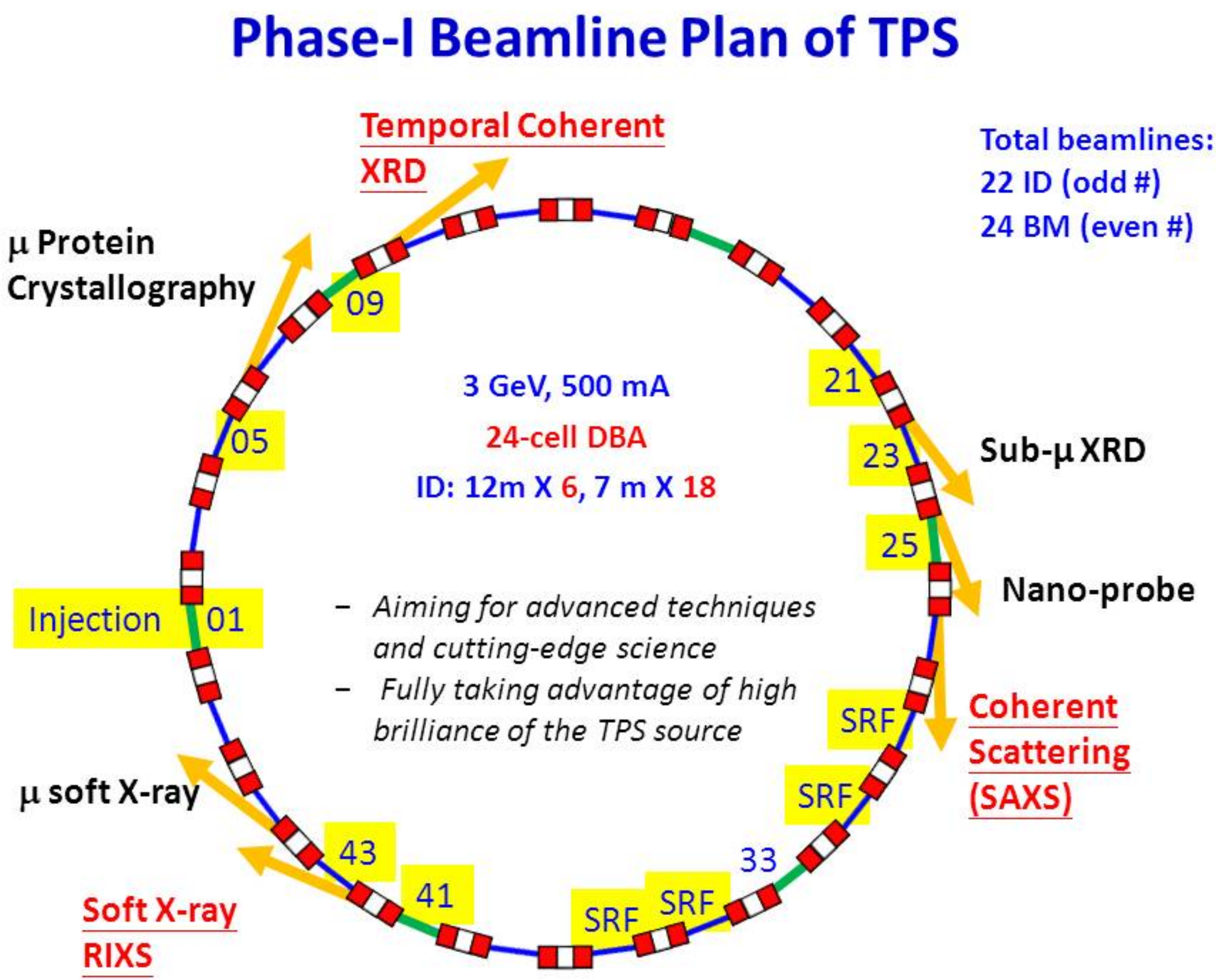
TLS



TPS



Taiwan Photon Source (TPS) – Hsinchu, Taiwan (continued)



Neutron Scattering

Neutrons have No Charge!

- Highly penetrating
- Nondestructive
- Can be used in extremes

Neutrons have a Magnetic Moment!

- Magnetic structure
- Fluctuations
- Magnetic materials

Neutrons have Spin!

- Polarized beams
- Atomic orientation
- Coherent and incoherent scattering

The Energies of neutrons are similar to the energies of elementary excitations!

- Molecular Vibrations and Lattice modes
- Magnetic excitations

The Wavelengths of neutrons are similar to atomic spacing!

- Sensitive to structure
- Gathers information from 10^{-10} to 10^{-7} m
- Crystal structures and atomic spacings

Neutrons probe Nuclei!

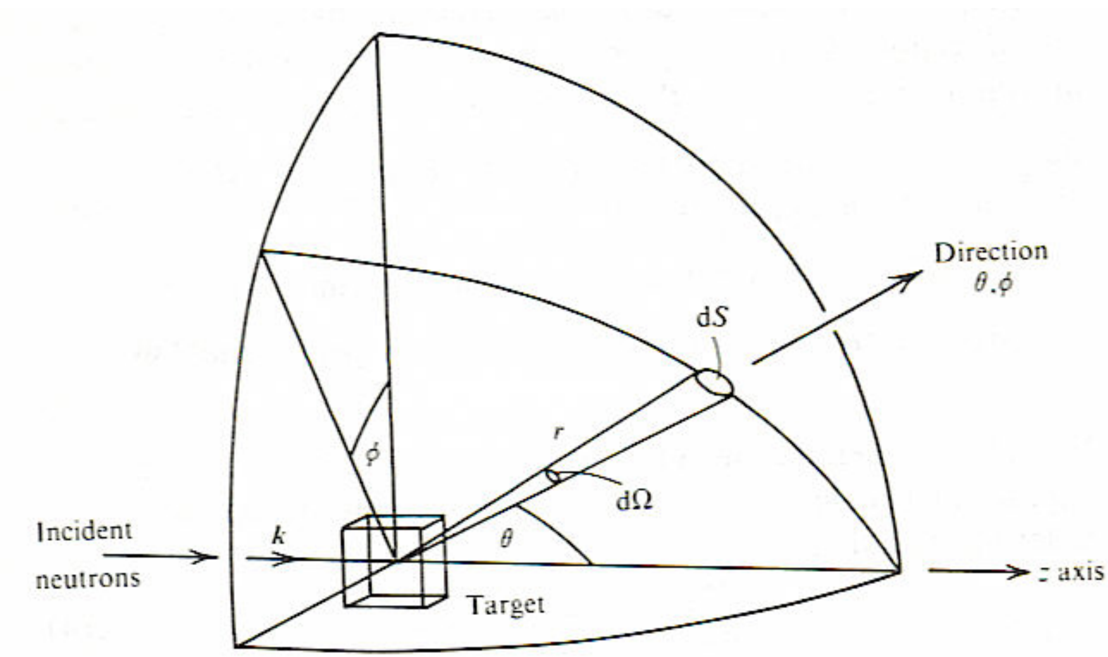
- Light atom sensitive
- Sensitive to isotopic substitution

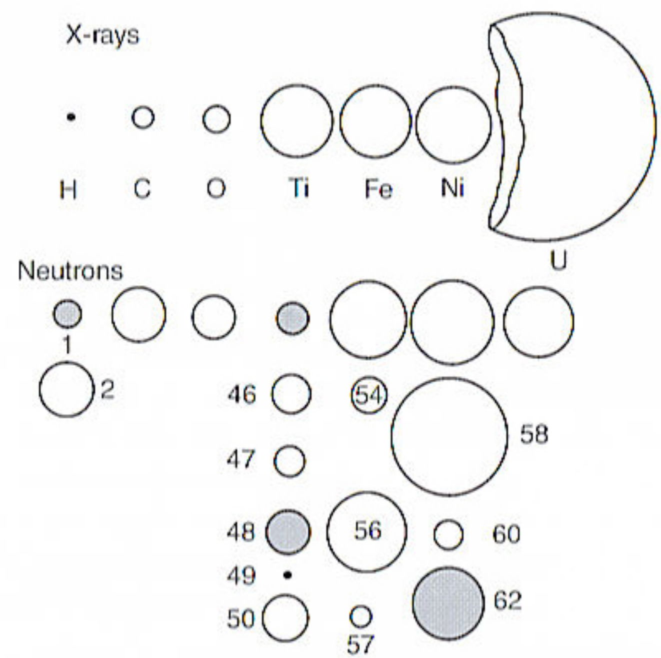
de Broglie Wavelength

$$\lambda = \frac{h}{p} = \frac{h}{mv} = \frac{h}{\sqrt{2mE}}$$

$E = 81.6\ meV$	$E = 1\ meV$
$v = 3950\ m/s$	$v = 437\ m/s$
$\lambda = 1 \times 10^{-10}\ m$	$\lambda = 9 \times 10^{-10}\ m$

Neutrons vs. X-rays

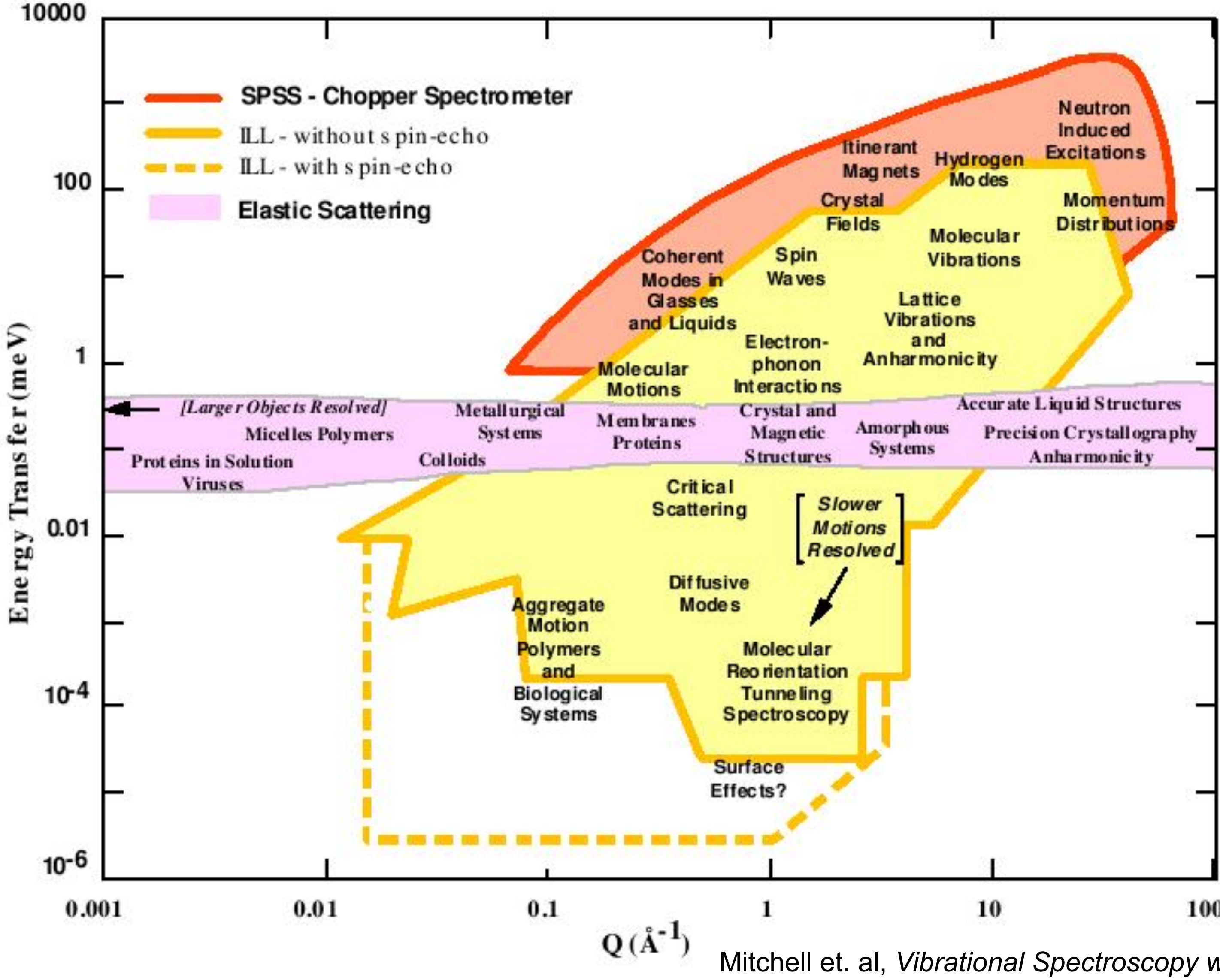




Chatterji, *Neutron Scattering from Magnetic Materials* (2006)

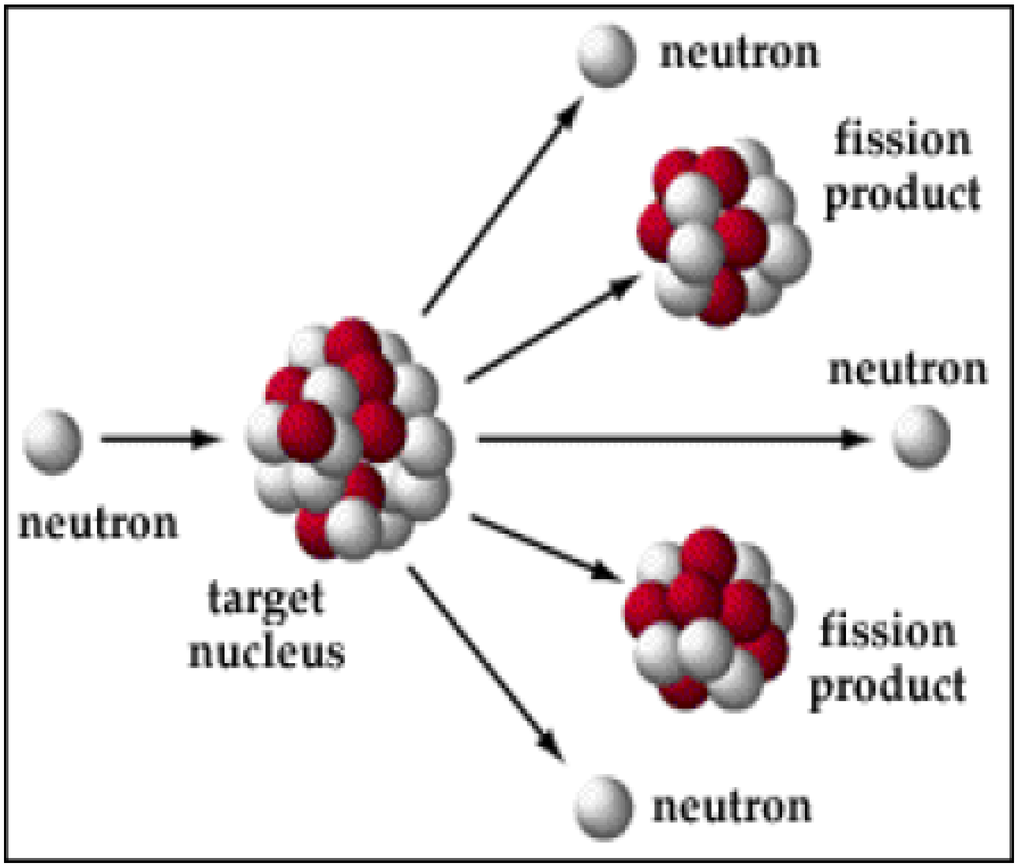
Neutrons allow easy access to atoms that are usually unseen in X-ray Scattering

How are neutrons useful?



Mitchell et. al, *Vibrational Spectroscopy with Neutrons* (2005)

Neutrons from Reactor Sources

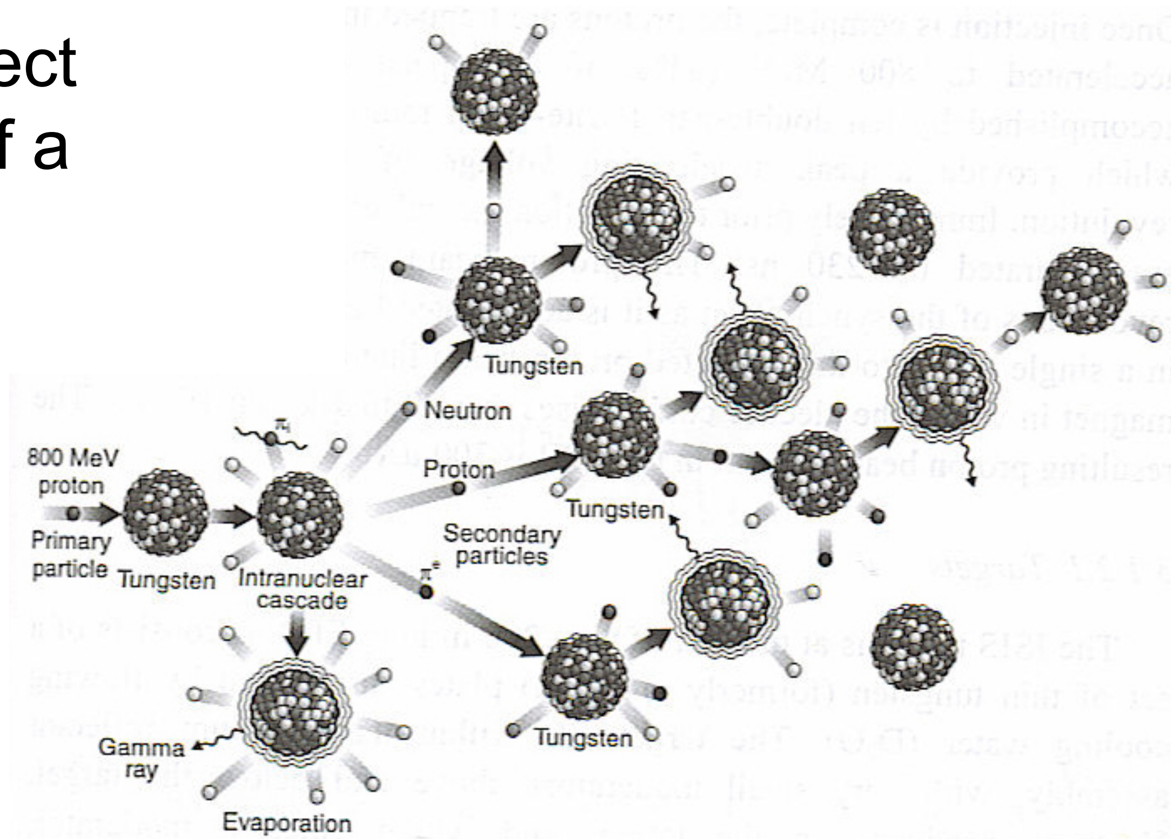


- Uses nuclear fission to create neutrons
- Continuous neutron flux
- Flux is dependent on fission rate
- Limited by heat flow in from the reaction
- Creates radioactive nuclear waste

Pynn, *Neutron Scattering: A Primer* (1989)

Neutrons from Spallation Sources

- Uses a cascade effect from the collision of a proton on a heavy metal.
- Pulsed Source
- High Intensity
- Heat production is relatively low



Mitchell et. al, *Vibrational Spectroscopy with Neutrons* (2005)

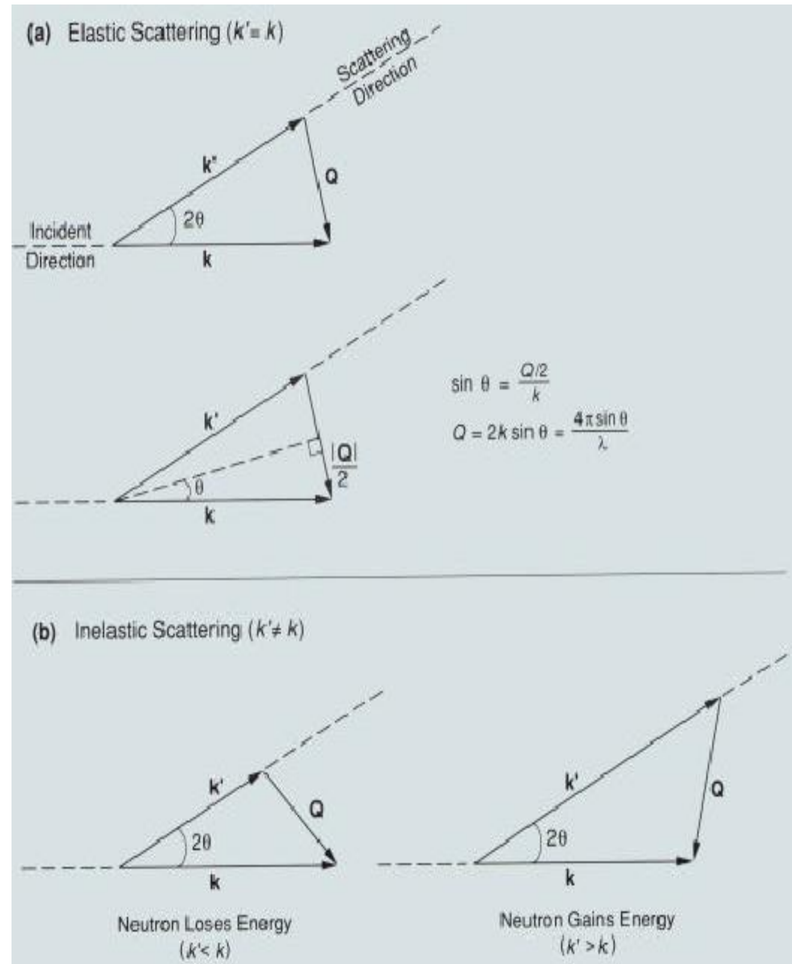
Neutron scattering

Elastic Neutron Scattering

- No loss of energy
- Examines the change in momentum or angle of the neutrons.

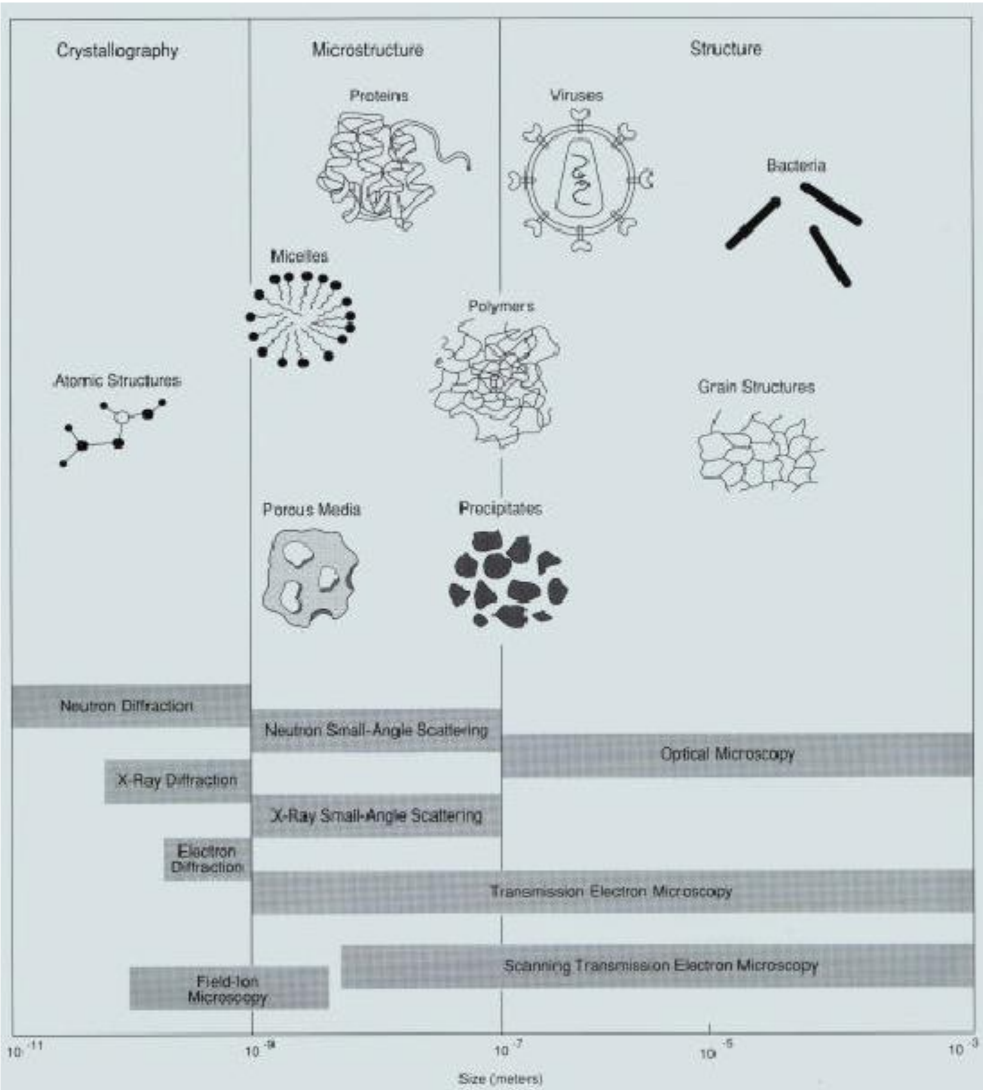
Inelastic Neutron Scattering

- Examines both momentum and energy dependencies.

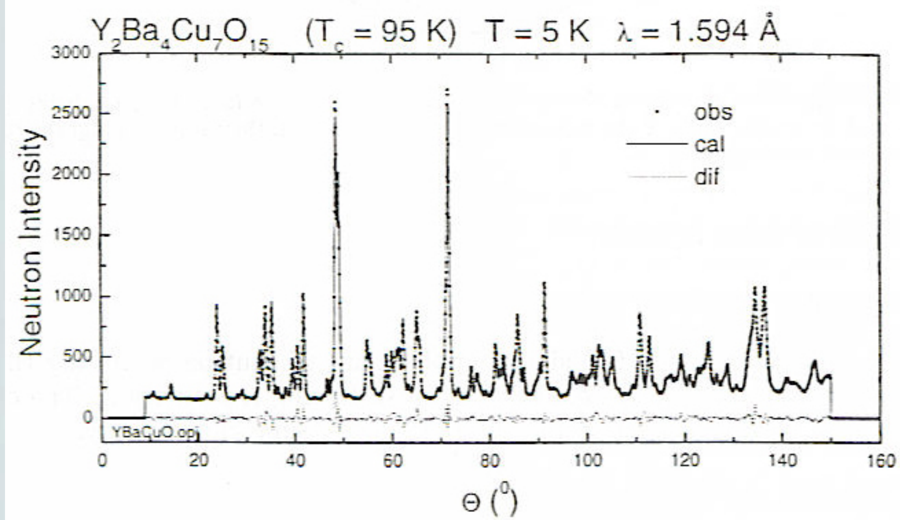


Pynn, *Neutron Scattering: A Primer* (1989)

Elastic Neutron Scattering



Pynn, *Neutron Scattering: A Primer* (1989)

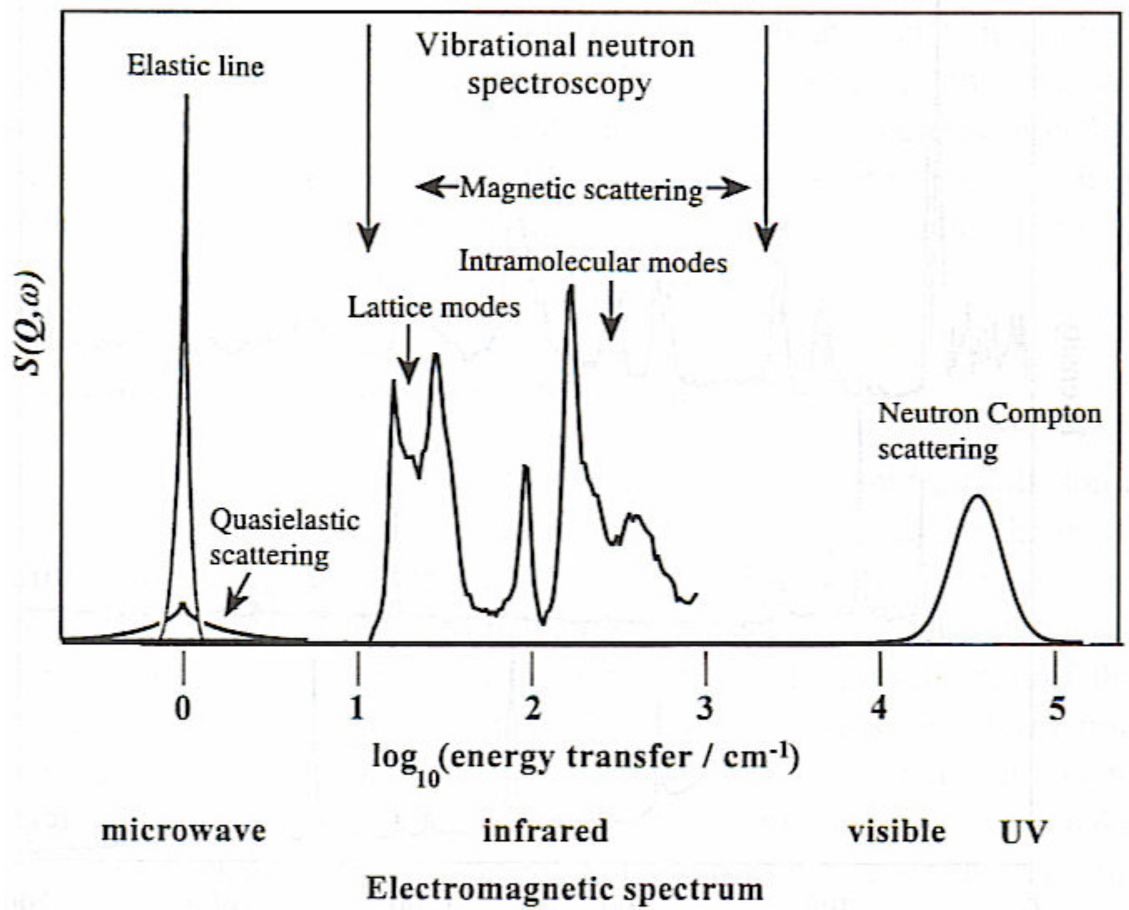


Mitchell et. al, *Vibrational Spectroscopy with Neutrons* (2005)

- Determine length scales and differentiate between nano-, micro-, and macro-systems.
- Utilizes position and momentum correlation.

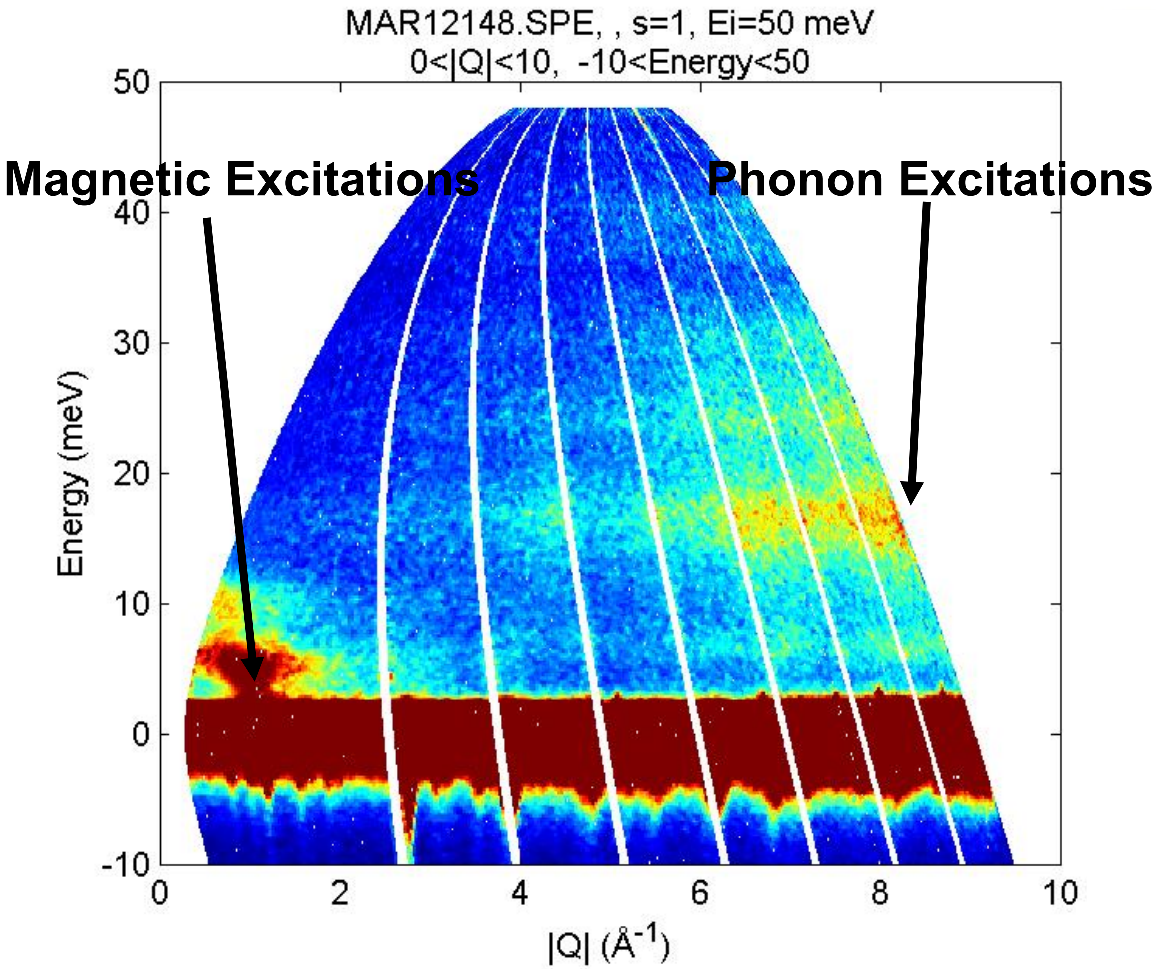
Inelastic Neutron Scattering

Uses both change in momentum and energy to characterize a systems vibrational, magnetic, and lattice excitations.



Mitchell et. al, *Vibrational Spectroscopy with Neutrons* (2005)

Vibrational and Magnetic Excitations



- Vibrational excitations are broad, large excitations.

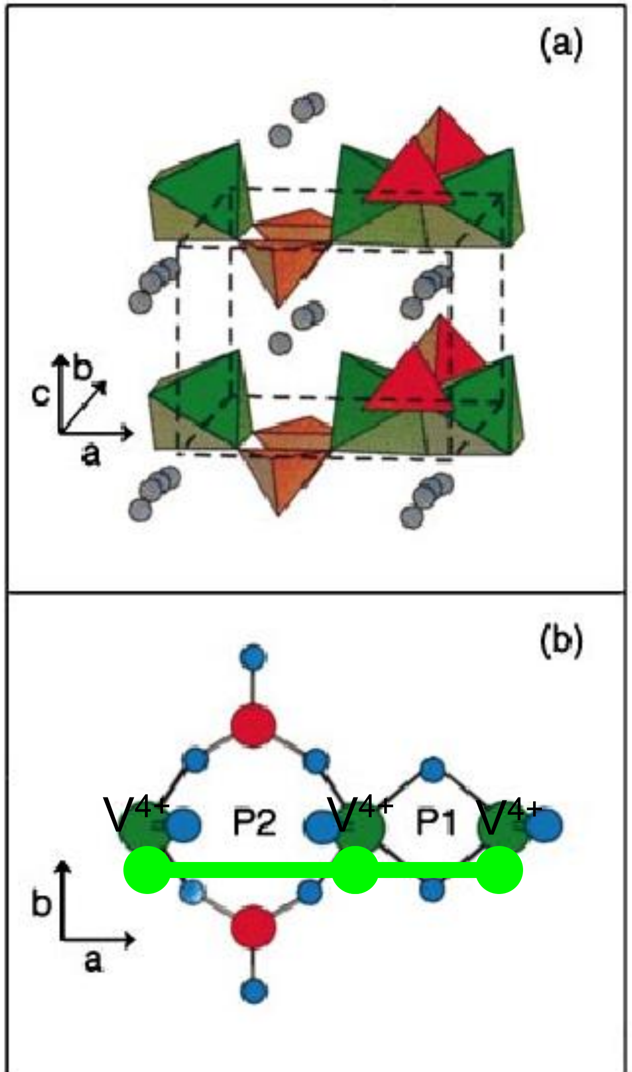
Neutrons observe all phonon and vibrational excitations. The intensity is determined by the phonons polarization vectors.

- Magnetic excitations are detailed by spin transitions of $\Delta S = 0$ and ± 1.

Q-dependence of magnetic excitations help determine the magnetic structure within the material.

M. B. Stone et al. Unpublished (2007)

Inelastic Neutron Scattering from magnetic sample



Tennant et. al, PRL (1997)

The use of neutron scattering on the material of $VODPO_4 \bullet ½\ D_2O$ clarified the magnetic structure of the material.