New Scientific Possibilities
&
Directions

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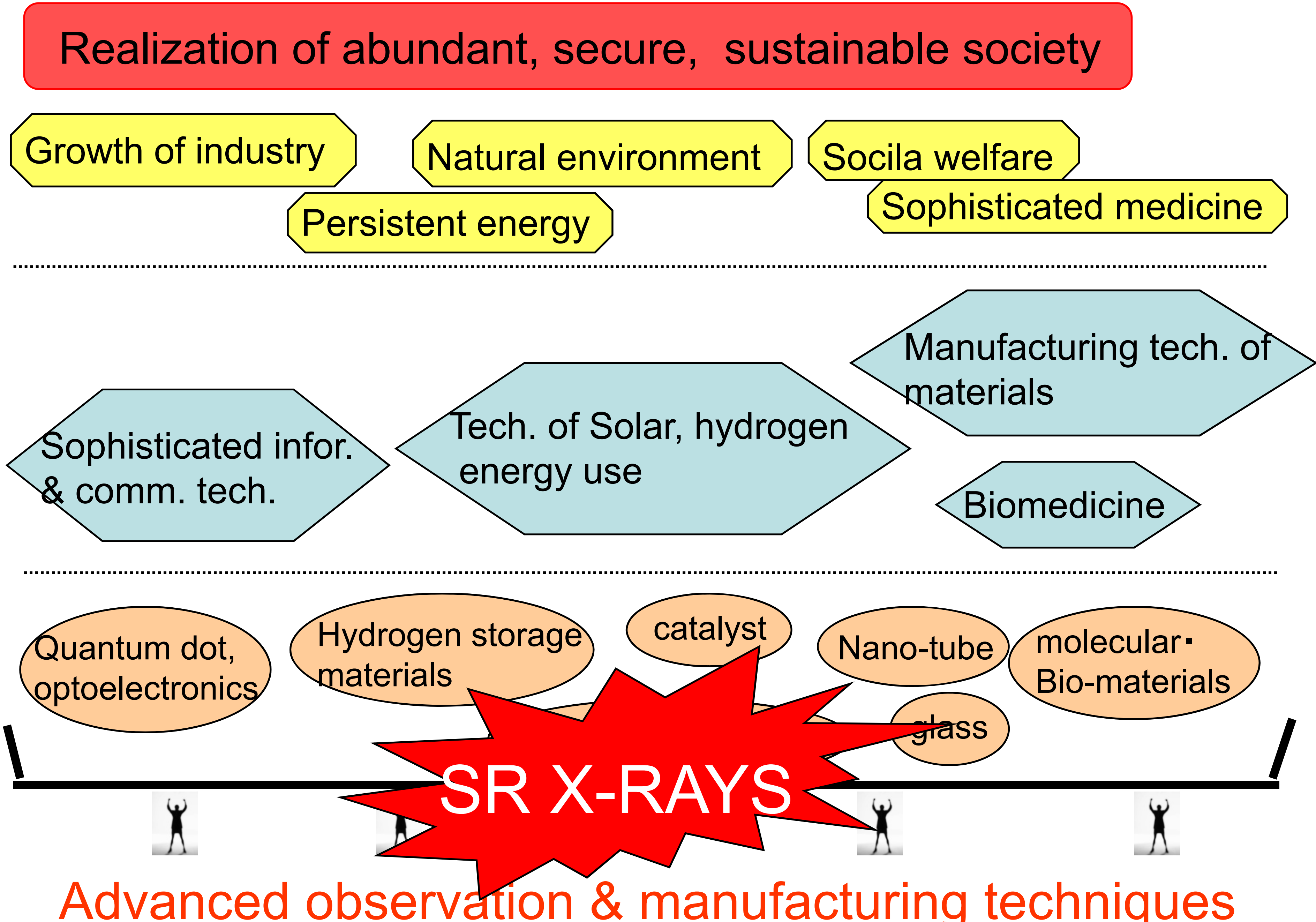
Key words:

- *In situ*
- Dynamics

Why SR?



Realization of abundant, secure, sustainable society

Growth of industry

Natural environment

Socila welfare

Persistent energy

Sophisticated medicine

Manufacturing tech. of materials

Sophisticated infor. & comm. tech.

Tech. of Solar, hydrogen energy use

Biomedicine

Quantum dot, optoelectronics

Hydrogen storage materials

catalyst

Nano-tube

molecular･Bio-materials

glass

SR X-RAYS

Advanced observation & manufacturing techniques

Energy tunability

↓

Resonance

Anomalous effect

Quantum Mechanical Description

Free particle:

$$H = \frac{P^2}{2m}$$

Interaction with an electromagnetic field:

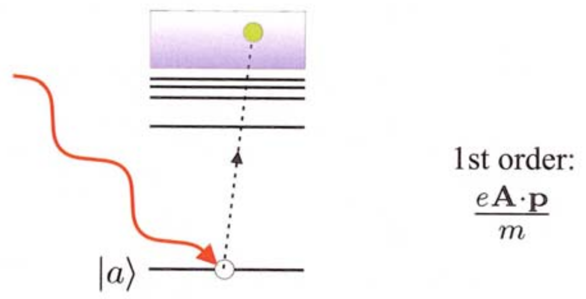
$$H = \frac{\left(\vec{P} - e\vec{A}\right)^2}{2m} = \frac{P^2}{2m} + \underbrace{\frac{e^2}{2m}A^2}_{\mathbf{H_1}} - \underbrace{\frac{e}{m}\vec{P} \bullet \vec{A}}_{\mathbf{H_2}}$$

Transition probability W:

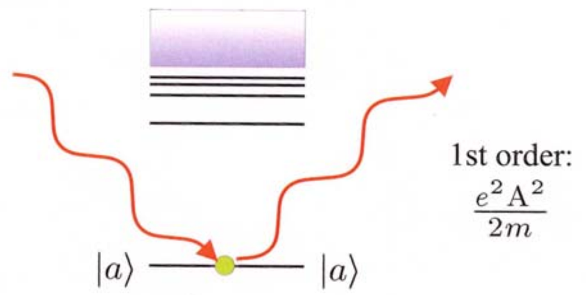
$$W = \frac{2\pi}{\hbar}\left| \underbrace{\langle f|H_1|i\rangle}_{f_0} + \underbrace{\sum_{n=1}^{\infty} \frac{\langle f|H_2|n\rangle\langle n|H_2|i\rangle}{E_i - E_n}}_{f_1 + if_2} \right|^2 \times \rho\left(\varepsilon_f\right), \qquad E_i = \hbar\omega + E_a$$

Energy selectivity → Use of anomalous dispersion

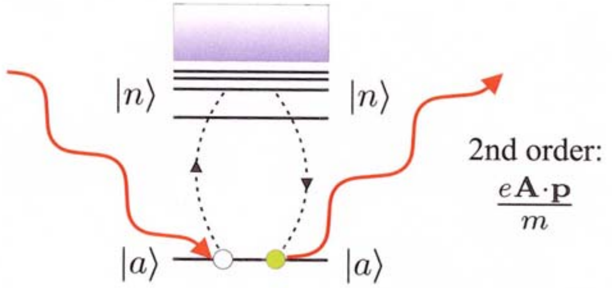
(a) Photoelectric absorption

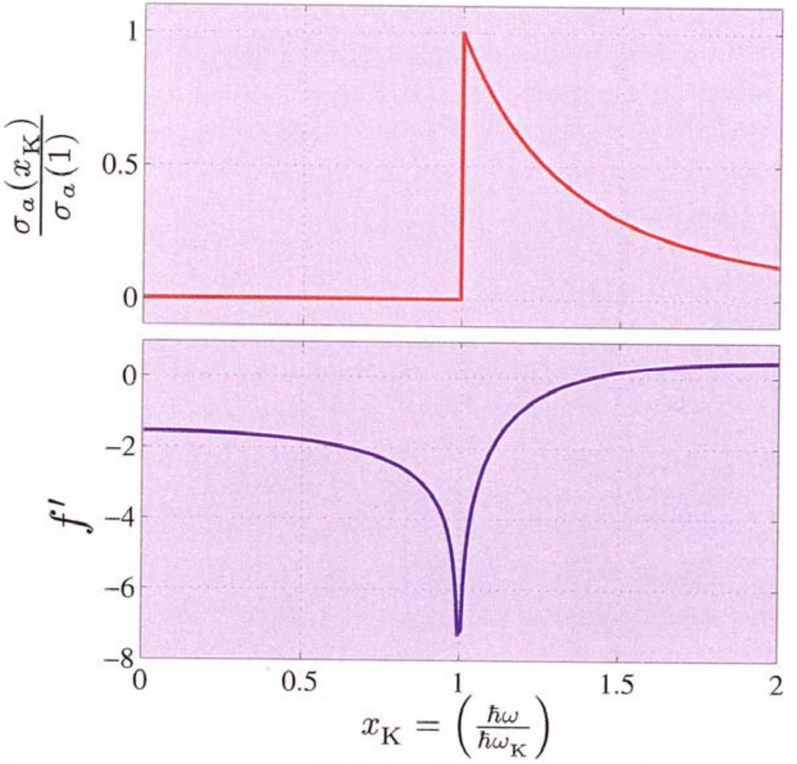


(b) Thomson scattering



(c) Resonant scattering



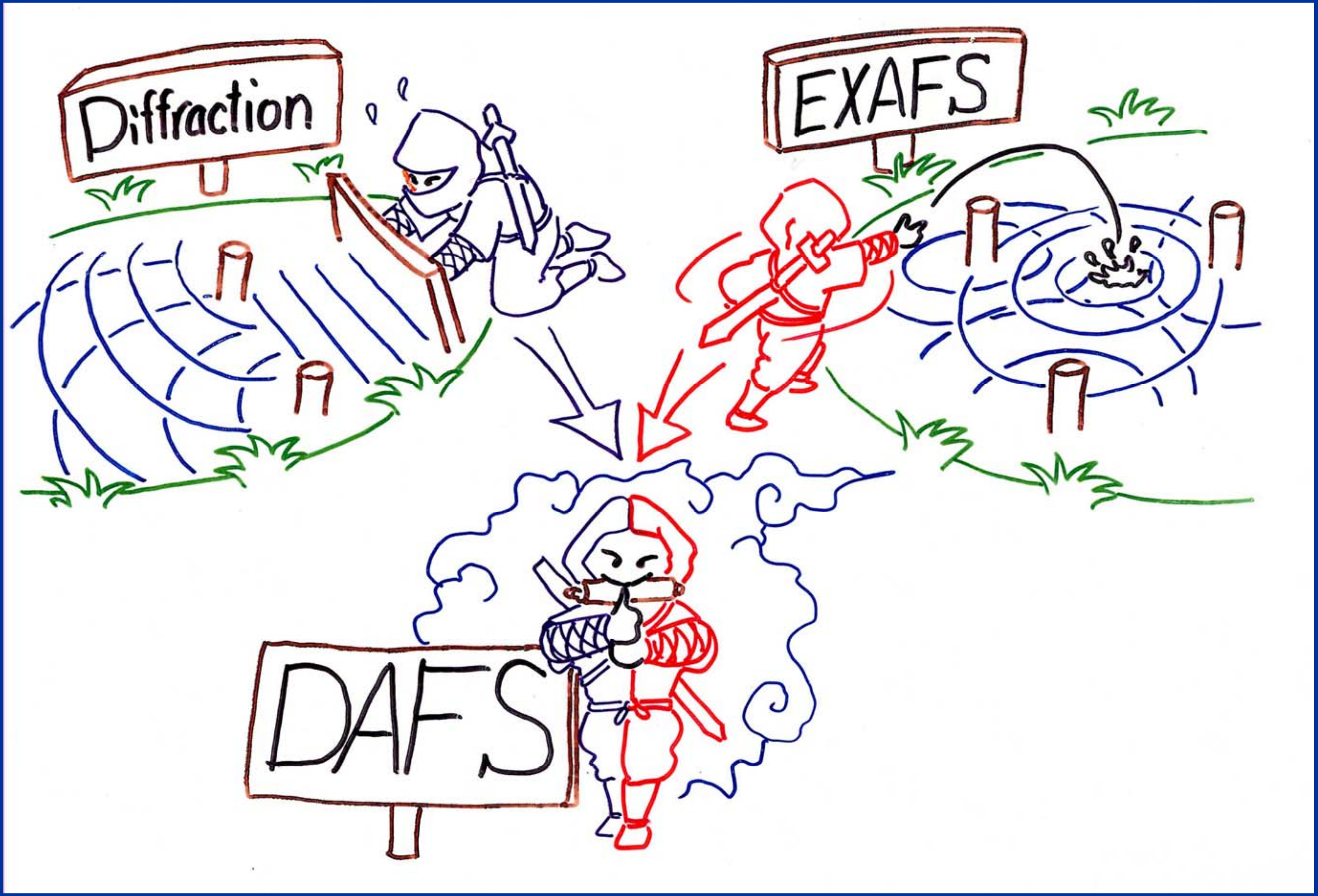


$$f(\mathbf{Q},\omega)=f^0(\mathbf{Q})+\underline{f'(\omega)+if''(\omega)}$$

K-K relation

From Elements of modern x-ray physics by J. Als-Nielsen and D. McMorrow

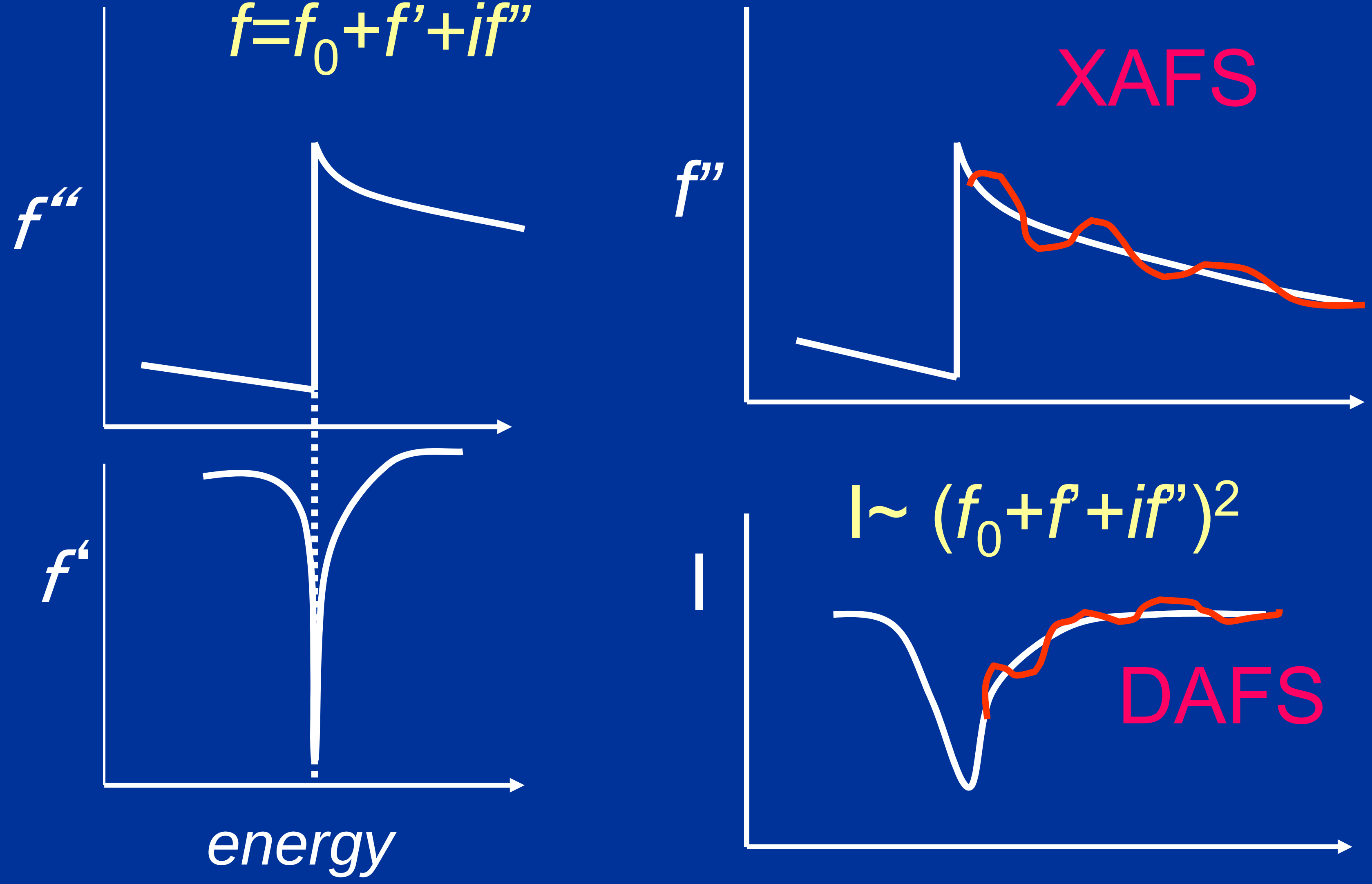
Marriage between XRD and XAFS



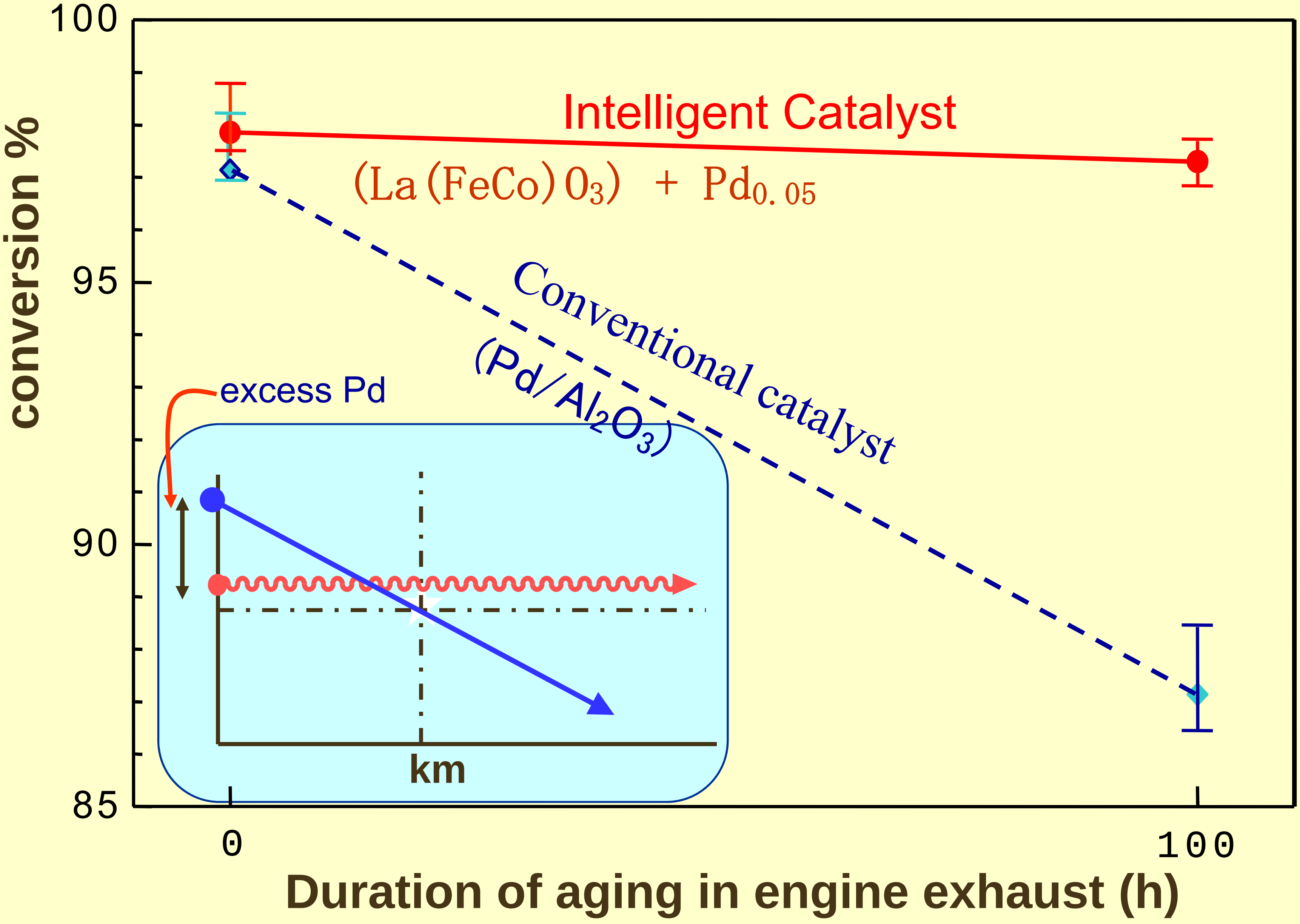
XAFS & DAFS

$$f=f_0+f'+if''$$

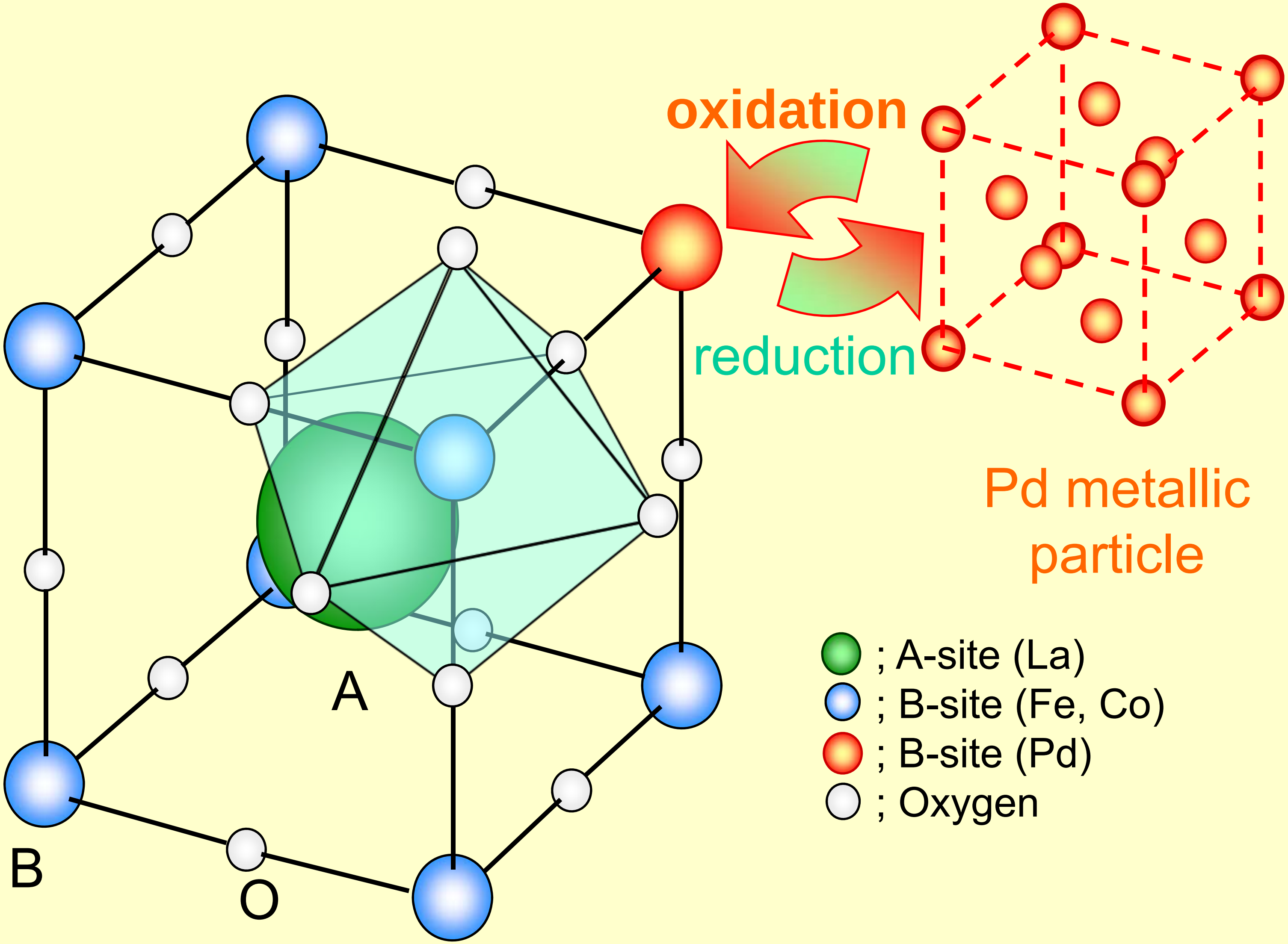
$$I\sim (f_0+f'+if'')^2$$



Life time of catalytic activity



Self-regeneration of Catalyst



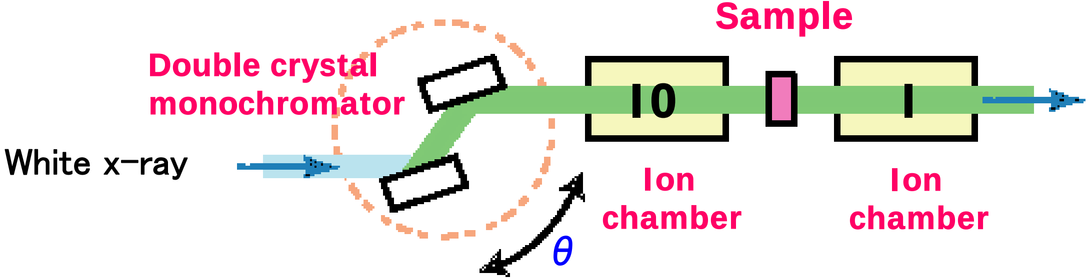
🟢 ; A-site (La)
🔵 ; B-site (Fe, Co)
🔴 ; B-site (Pd)
⚪ ; Oxygen

Y. Nishihata, J. Mizuki, et al.,Nature 418 ('02) 164

DXAFS (Energy Dispersive XAFS)

Standard XAFS

(Side-View)

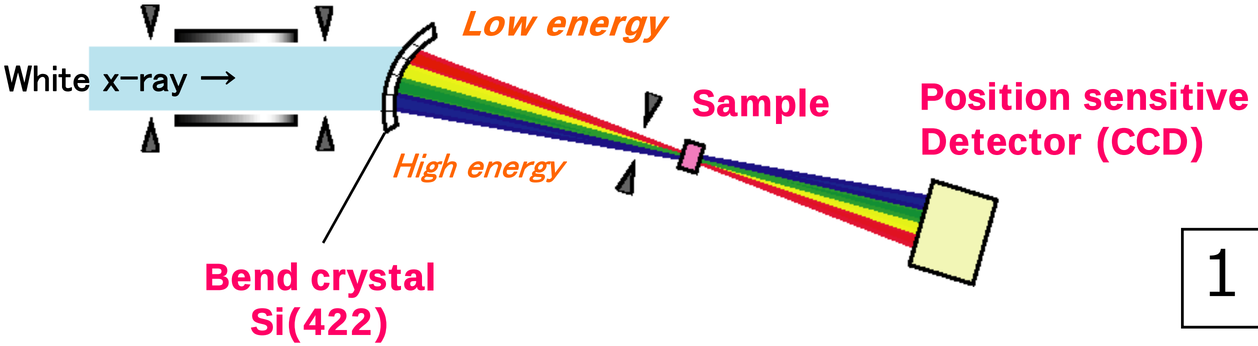


Data acquisition time

3 - 30 min

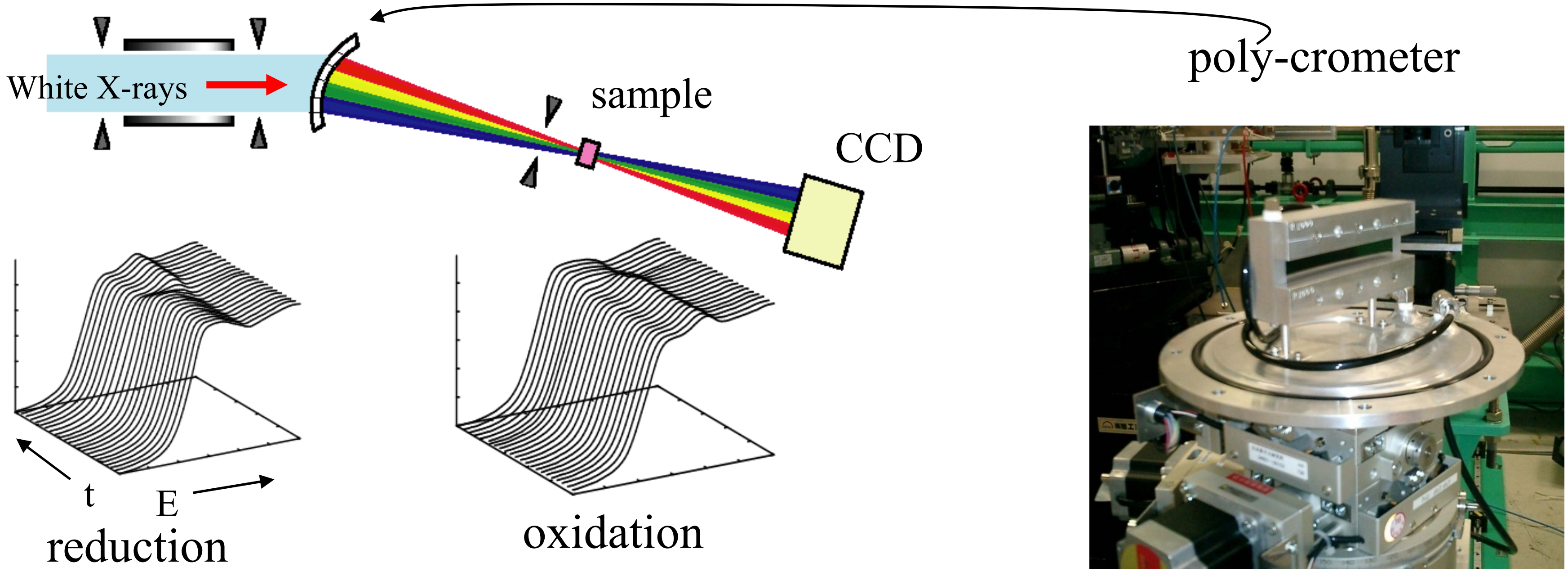
DXAFS

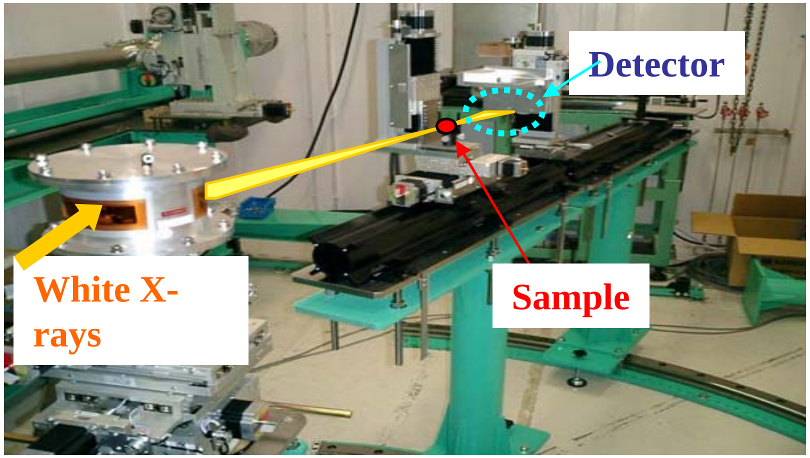
(Top-View)

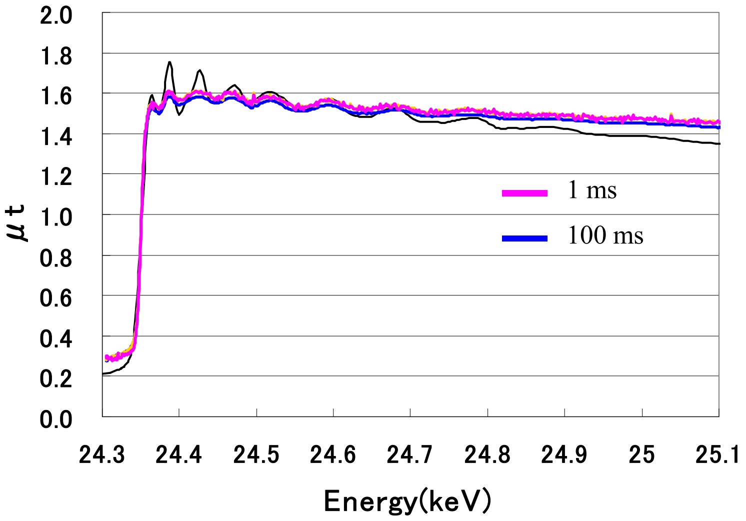


1 - 100msec

Dispersive XAFS (DXAFS) for studying dynamic





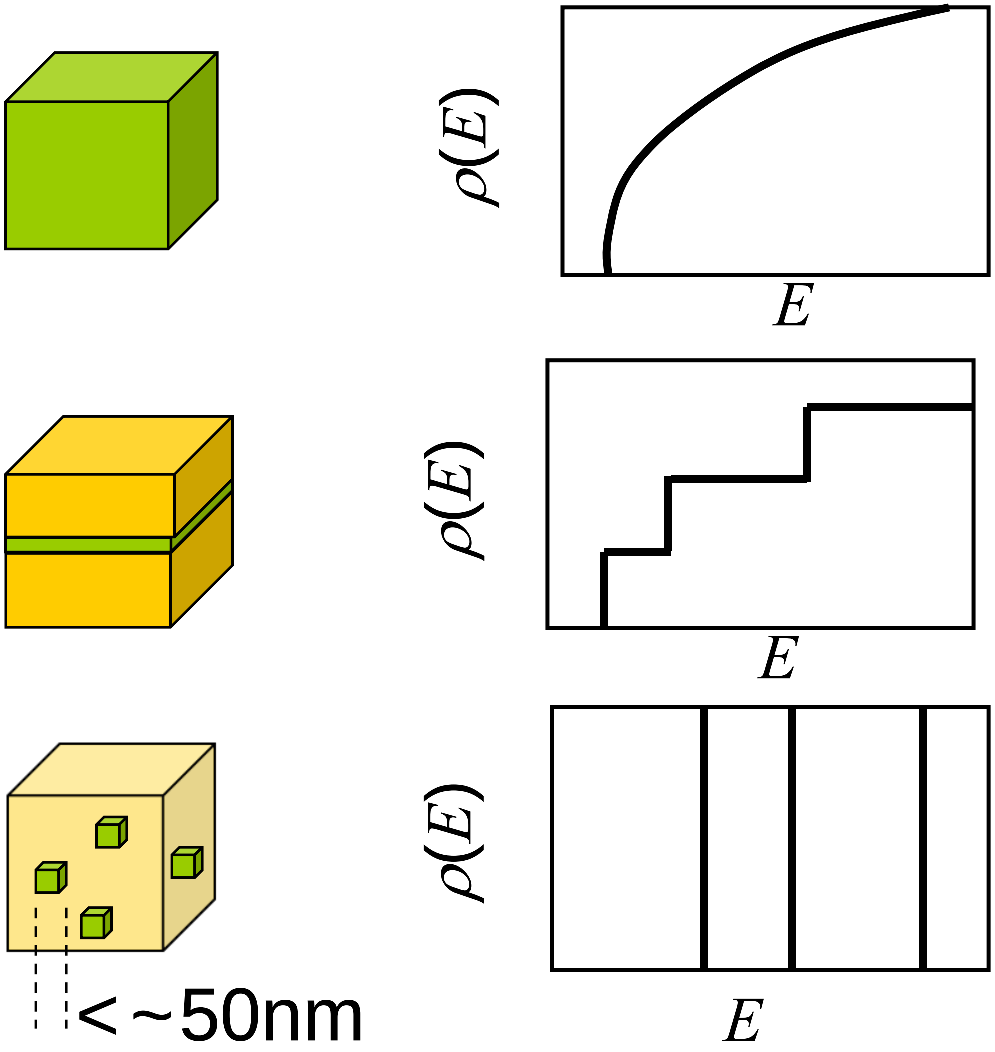


BL-14B1
Pd-K edge

Live in a convenient & sophisticated Society

- new information & communication tech -

Semiconductor nano-structure



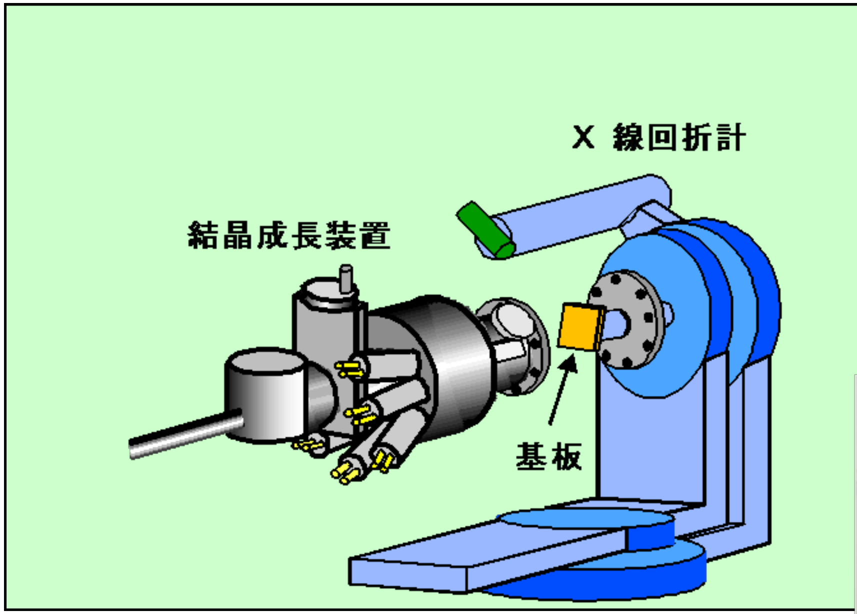
Bulk

Quantum well

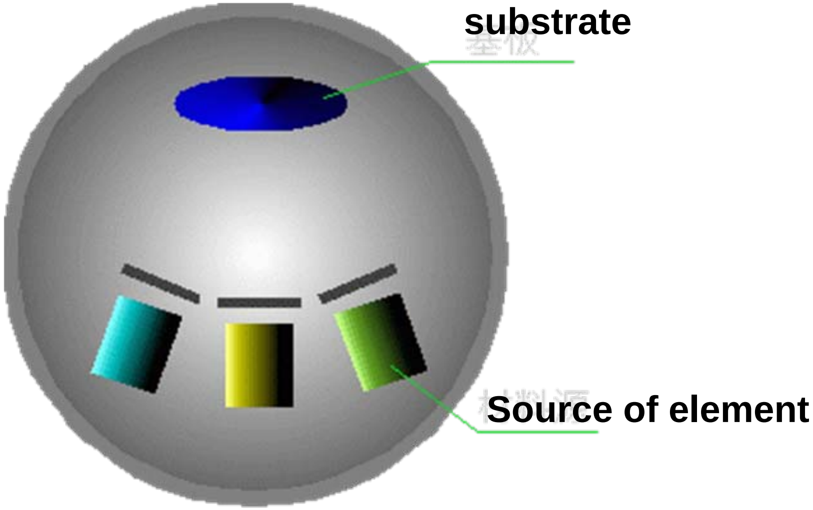
Quantum dot

Surface diffractometer + MBE

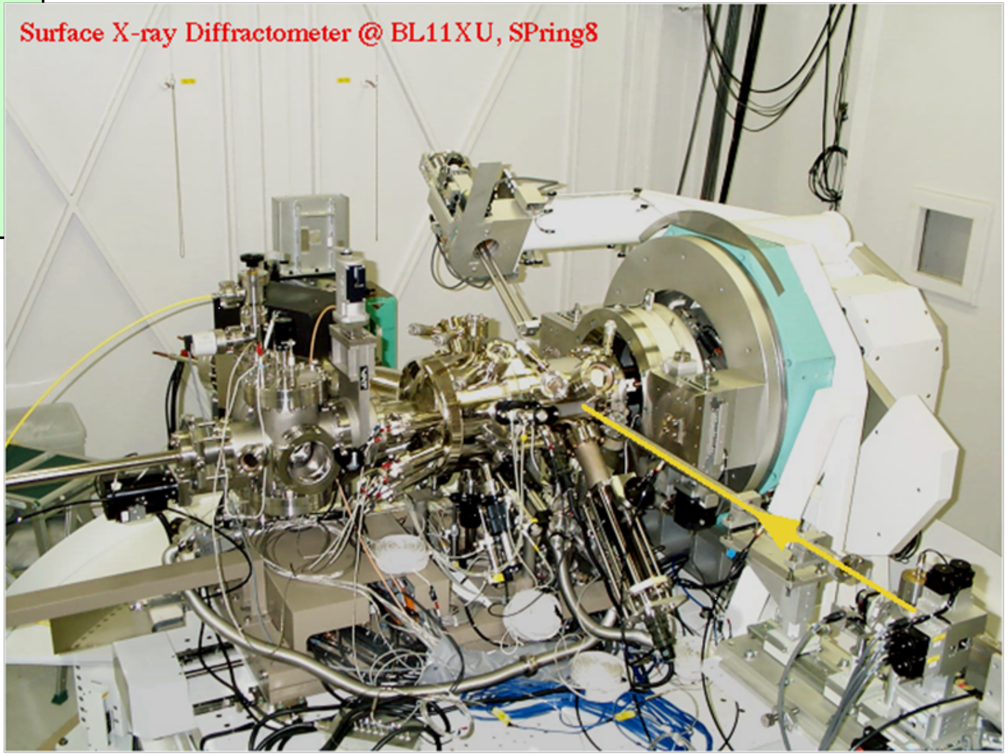
—study of reaction front—



Marriage between crystal growth and SR X-rays.



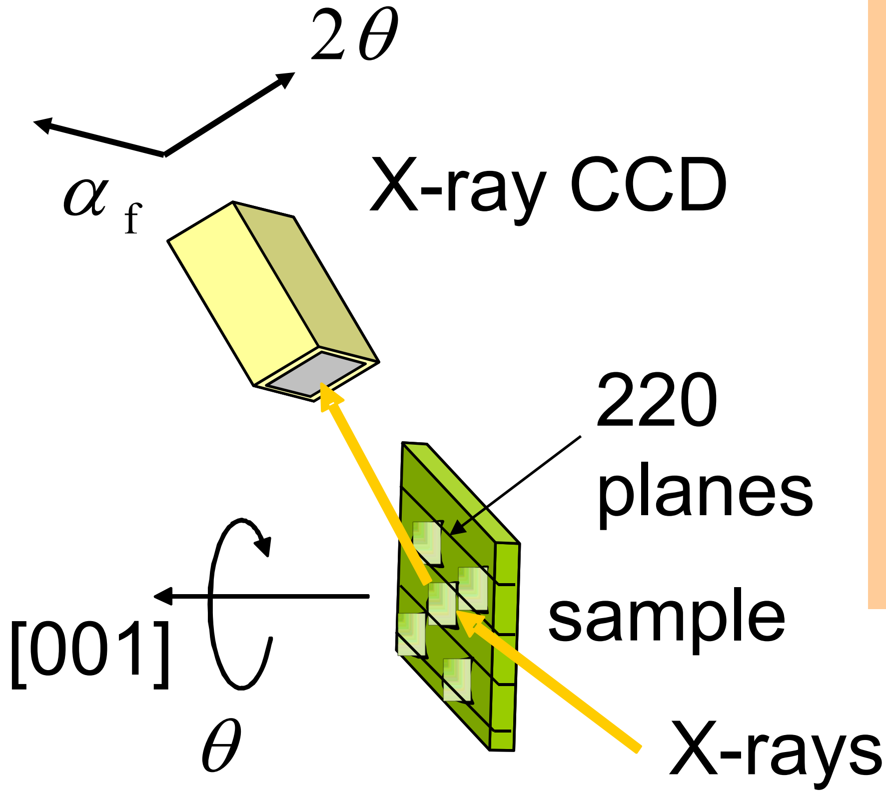
（from home page of Kishimoto Lab., Sophia Univ.）

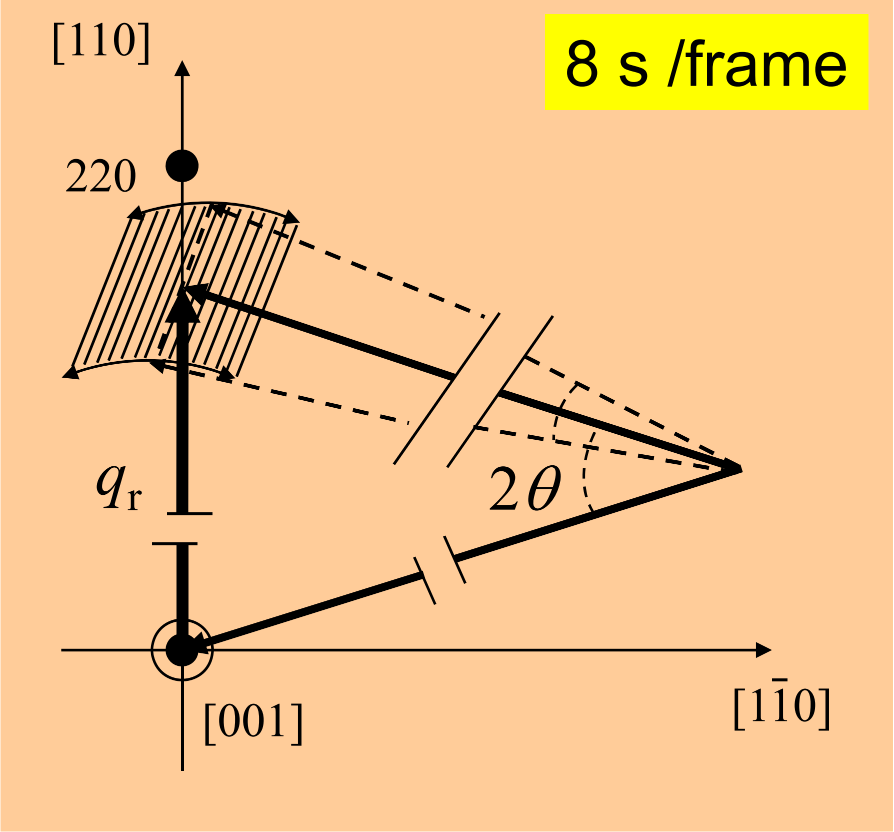


Surface X-ray Diffractometer @ BL11XU, SPring8

M. Takahasi J. M. et al., Jpn. J. Appl. Phys. 41 ('02) 6247

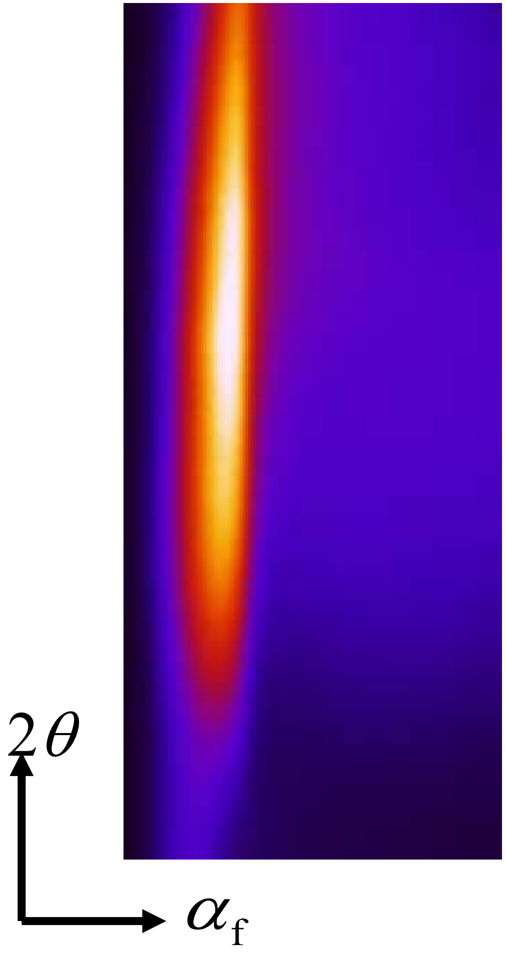
Experimental configuration





Interpretation of CCD images

4.9ML InAs
at 480℃



$$I(2\theta,\alpha_\mathrm{f}) \propto T(\alpha_\mathrm{i},z)T(\alpha_\mathrm{f},z)S(2\theta)$$

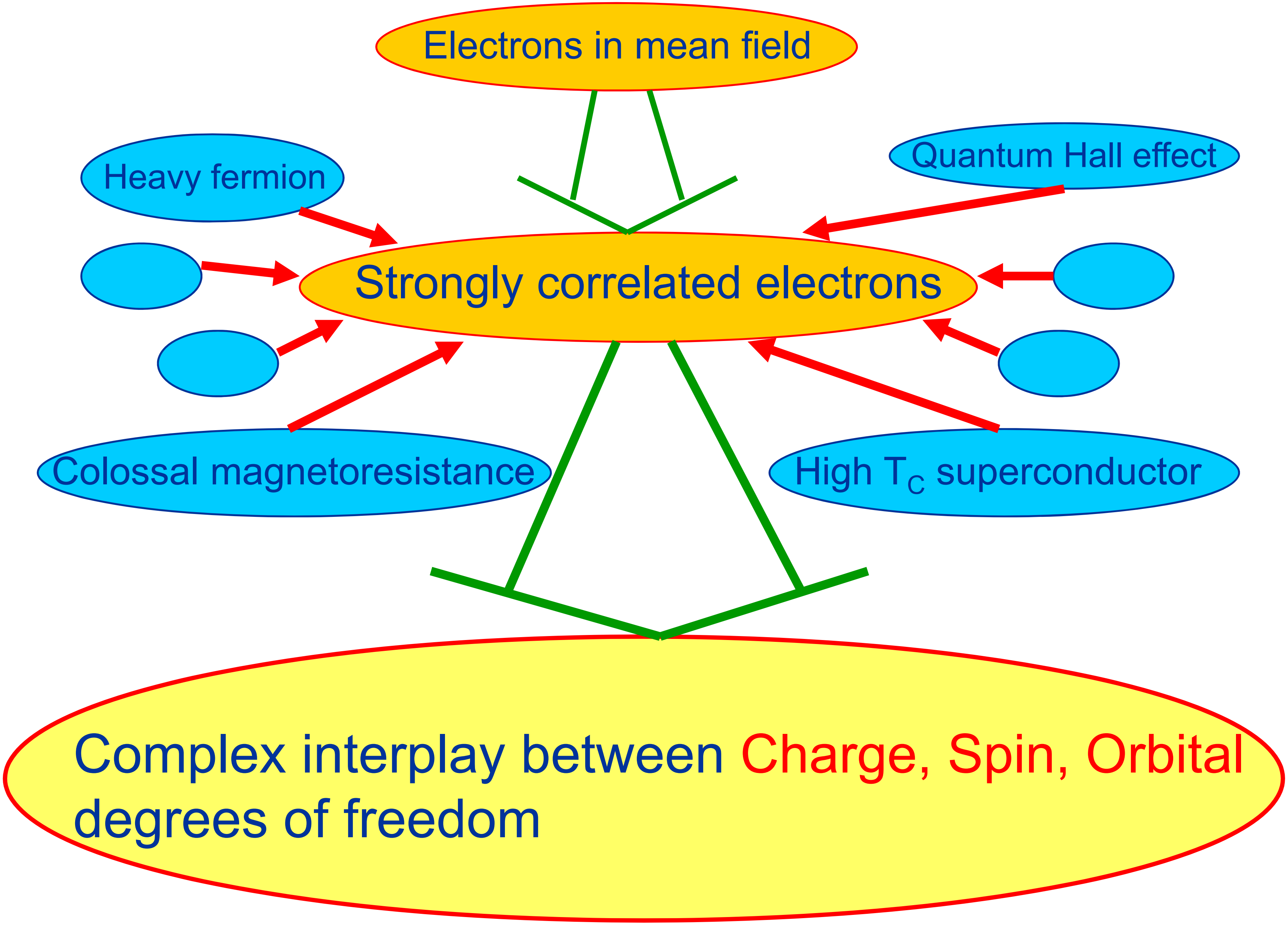
$S(2\theta)$: kinematical θ–2θ spectrum

→ strain (reciprocal space)

$T(\alpha_\mathrm{i,f},z)$: multiple-diffraction effect

→ height (real space)

Discover new scientific phenomena and invent new materials



Electrons in mean field

Heavy fermion

Quantum Hall effect

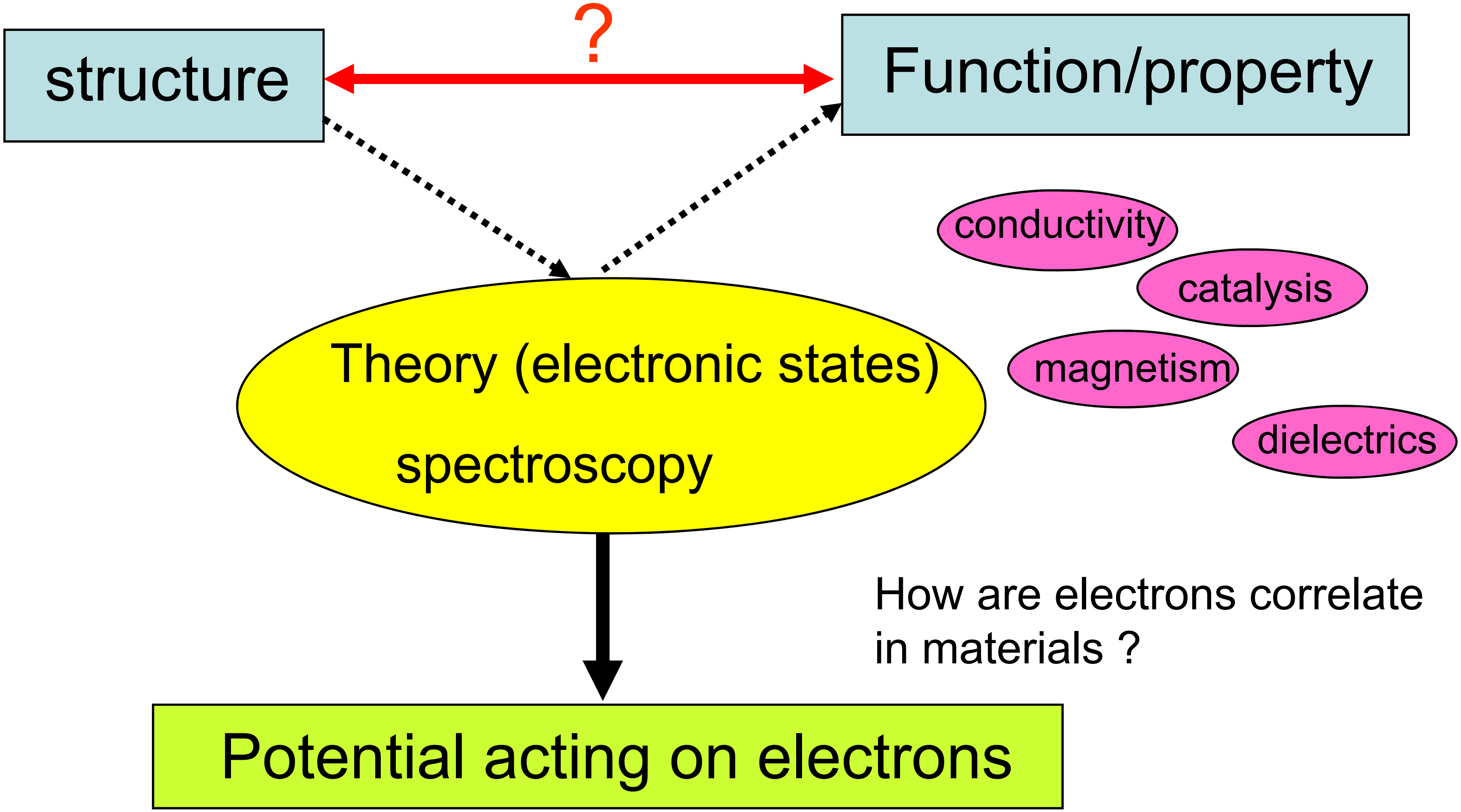
Strongly correlated electrons

Colossal magnetoresistance

High T_C superconductor

Complex interplay between Charge, Spin, Orbital degrees of freedom

Actor /actress in materials is the electron !



structure

?

Function/property

conductivity

catalysis

magnetism

dielectrics

Theory (electronic states)

spectroscopy

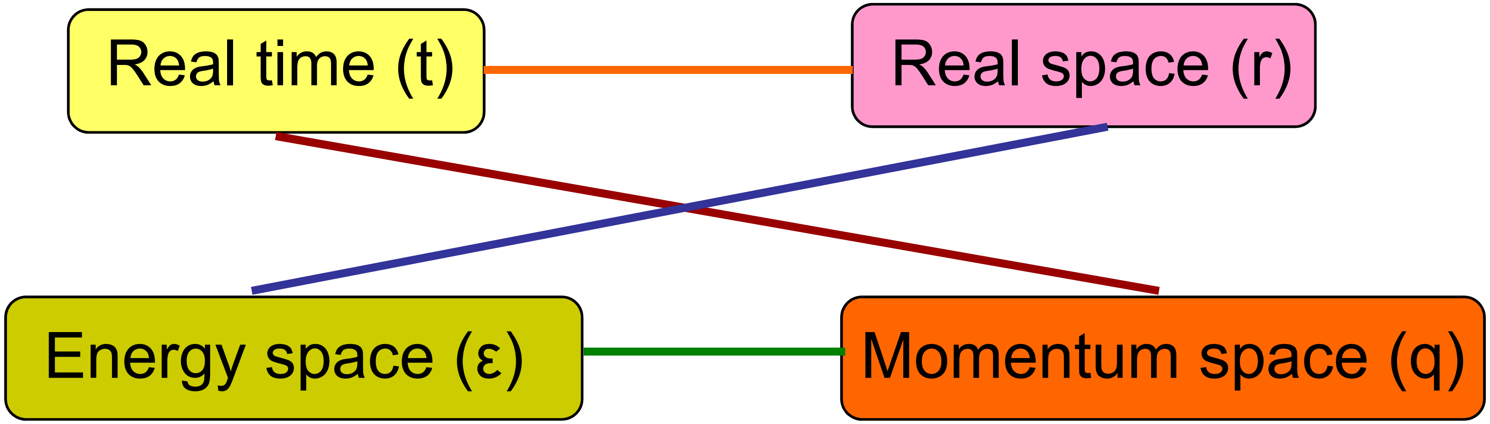
Potential acting on electrons

How are electrons correlate in materials ?

Dynamics

Observation space

In what space should we measure physical properties?



Real time (t)

Real space (r)

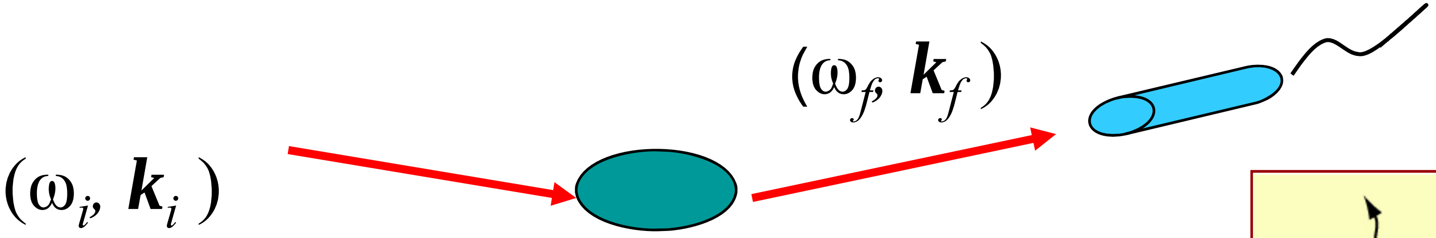
Energy space (ε)

Momentum space (q)

How do we know the potential for electrons?

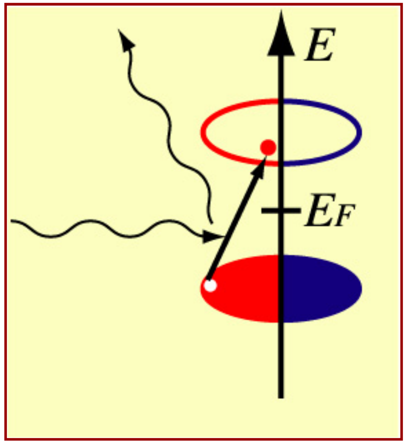
Dynamics!

Observation of dynamical properties of electrons and atoms



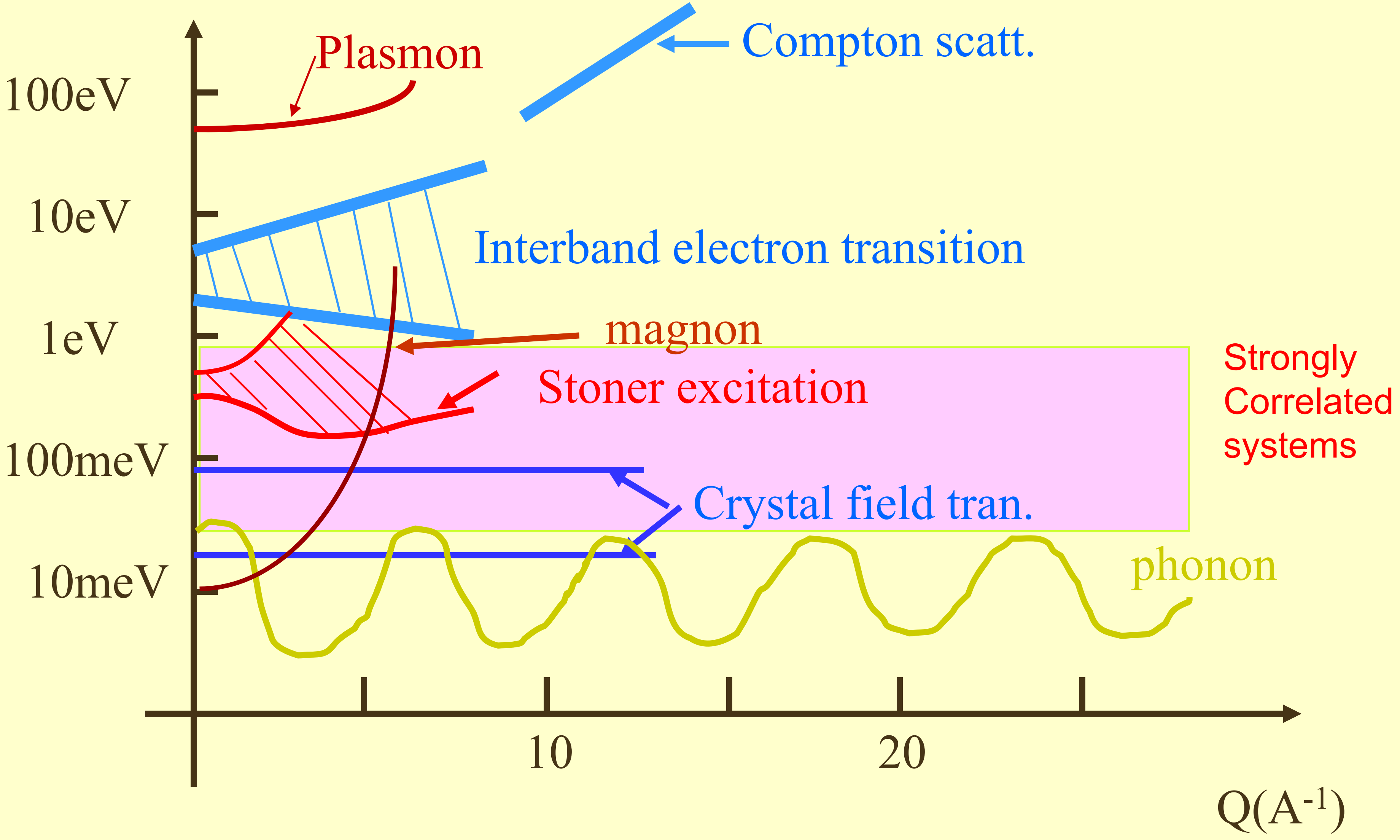
$$\mathbf{Q} = \boldsymbol{k}_i - \boldsymbol{k}_f$$

$$\Delta E = \omega_i - \omega_f \equiv \omega$$



inelastic scattering!

Elementary Excitations in Solids



Inelastic Scattering

$$I(Q,E) \sim \underline{[V(Q)]^2}\left[1-e^{-\beta E}\right]^{-1} \bullet \underline{\operatorname{Im}\chi(Q,E)}$$

Interaction of probe

Generalized susceptibility

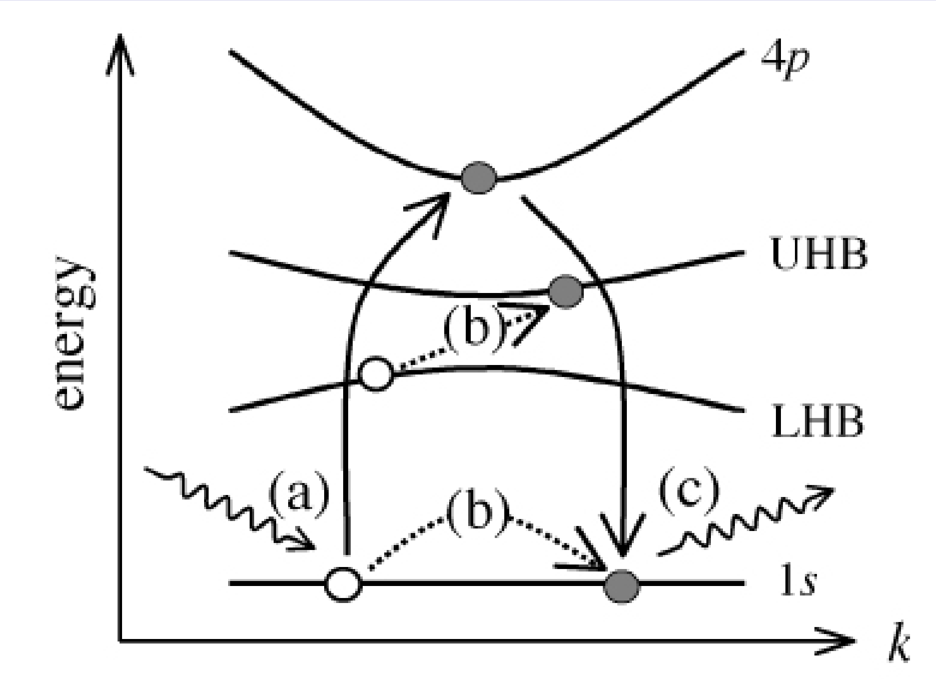
For neutrons → Spin susceptibility, Nuclear susceptibility

For X-rays, For electrons → Charge susceptibility

⇨ $$\chi(Q,E) = -(Q^2/4\pi^2 N)\, \underline{1/\varepsilon(Q,E)}$$

Dynamical dielectric function

RIXS of 3d transition elements



K-edge （1s→4p、several keV）

(a) absorption

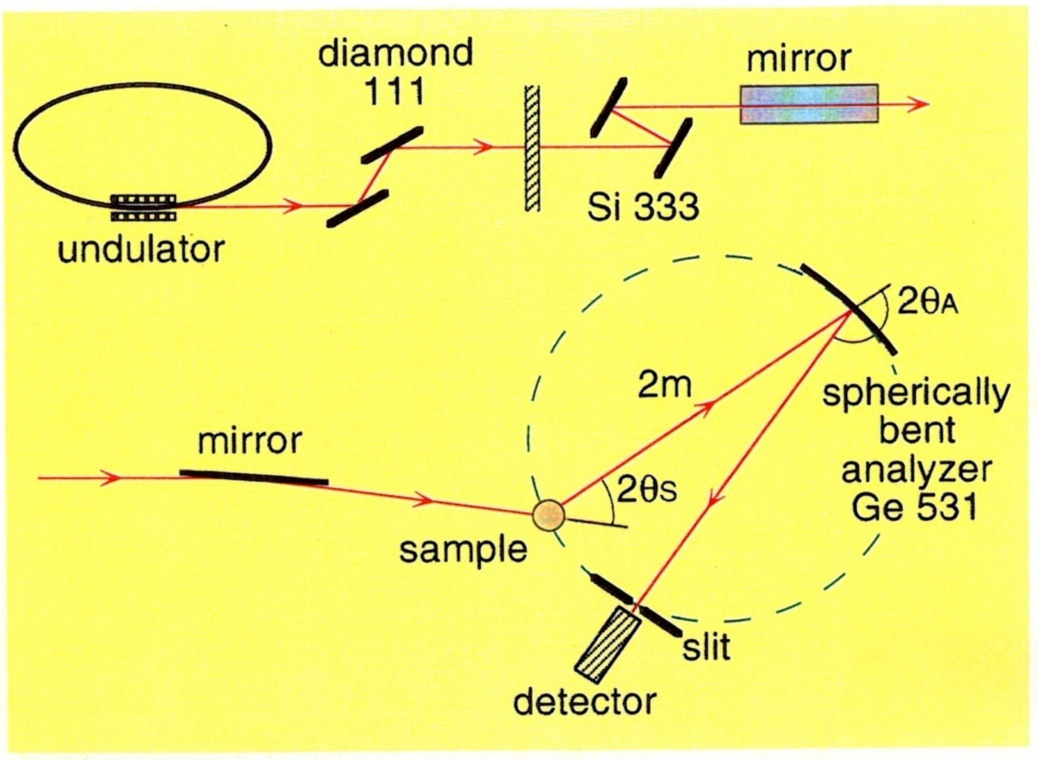
(b) interaction between

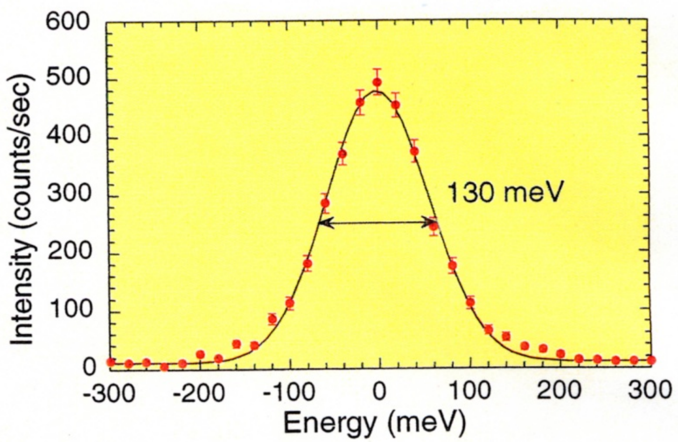
1s core-hole and 3d electron

(c) X-ray emission

$$I \propto \left| \sum_{m,n} \frac{\langle f | \mathbf{A}\cdot\mathbf{p} | n \rangle \langle n | V_{1s-3d} | m \rangle \langle m | \mathbf{A}\cdot\mathbf{p} | i \rangle}{(E_f - E_n + \hbar\varpi_f - i\Gamma)(E_i - E_m + \hbar\varpi_i - i\Gamma)} \right|^2$$

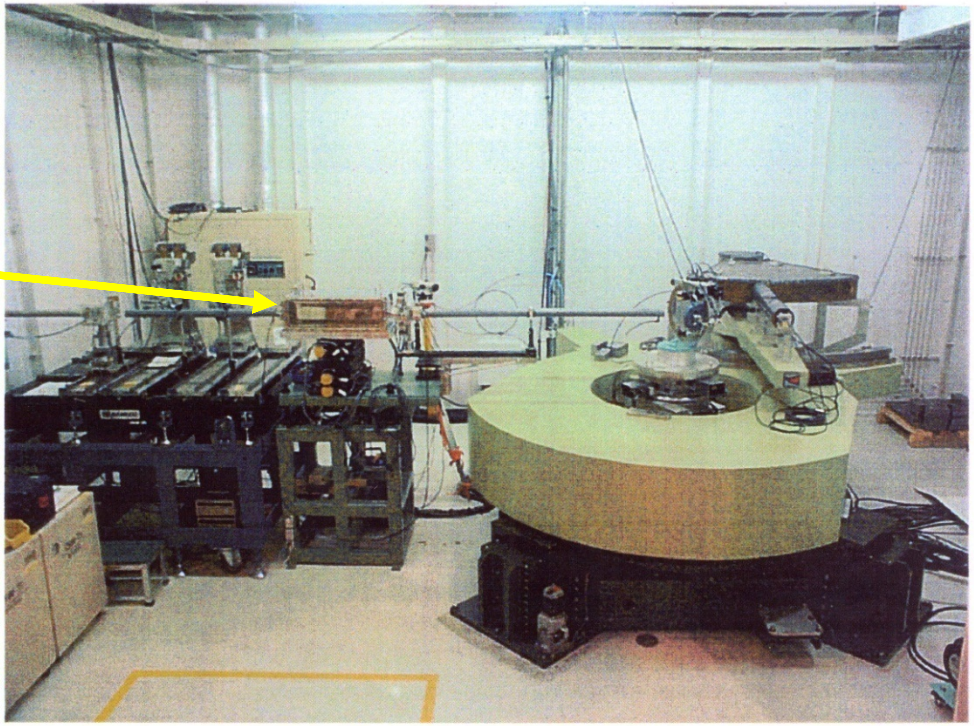
Set-up of Inelastic Scattering Spectrometer at BL-11XU



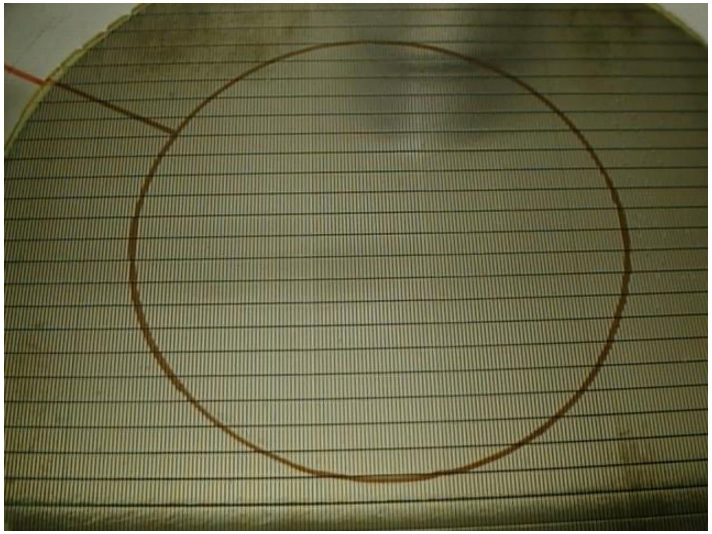


Observed energy Resolution at 6.5 keV

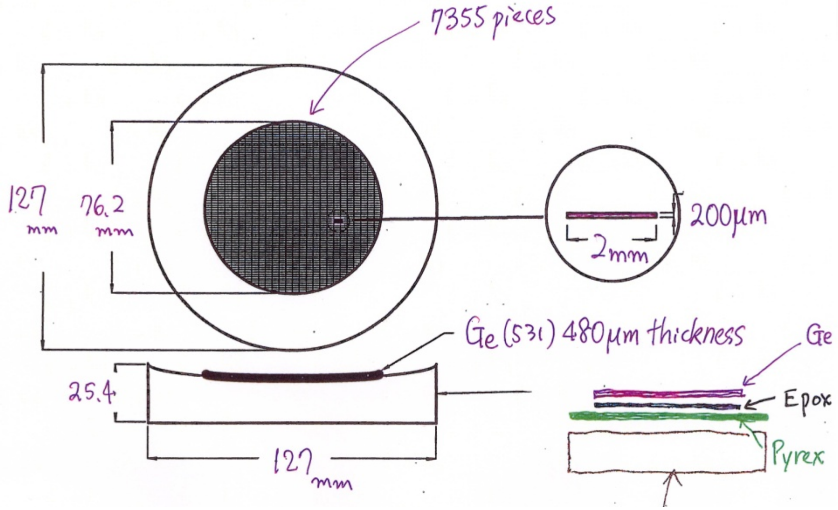
Picture of the spectrometer at BL-11 XU, SPring-8



Focusing mirror

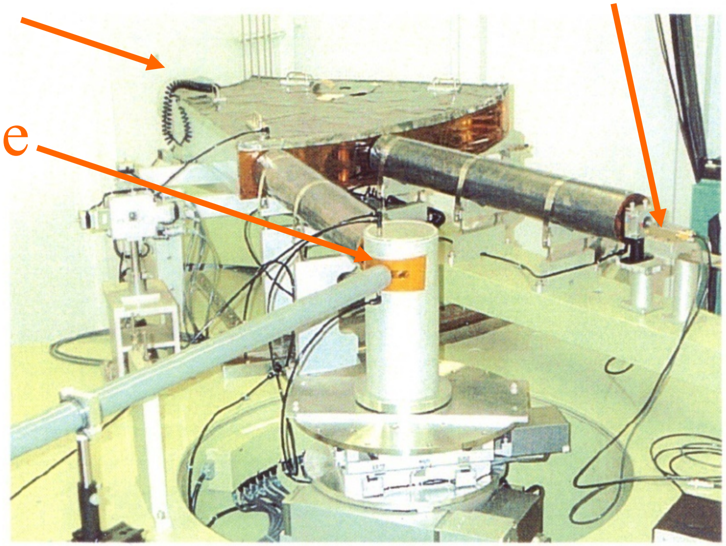


analyzer



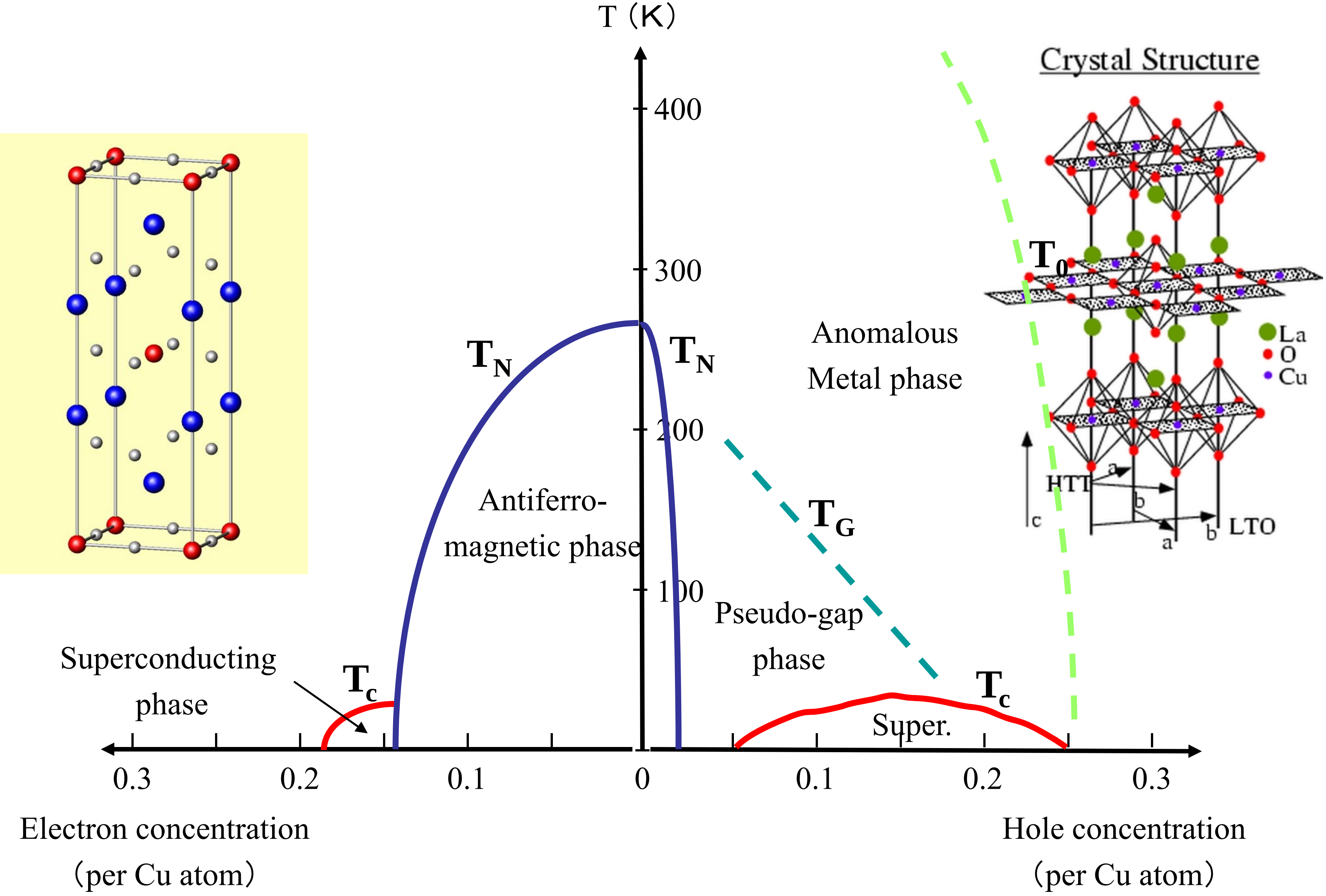
detector

sample

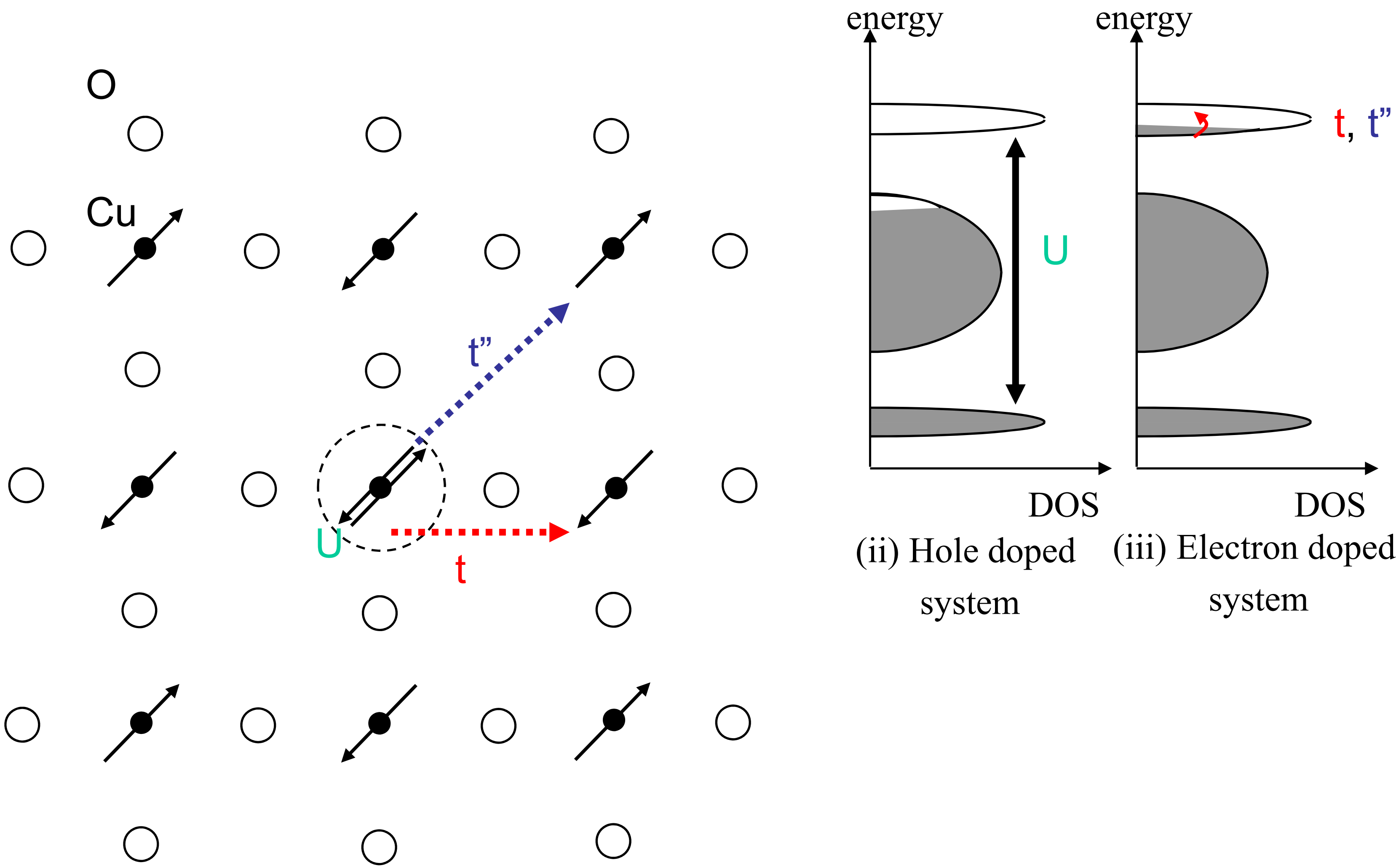


Mechanism of High T_C superconductor ?

Symmetry of doping-phase diagram



Schematic diagram of electronic states



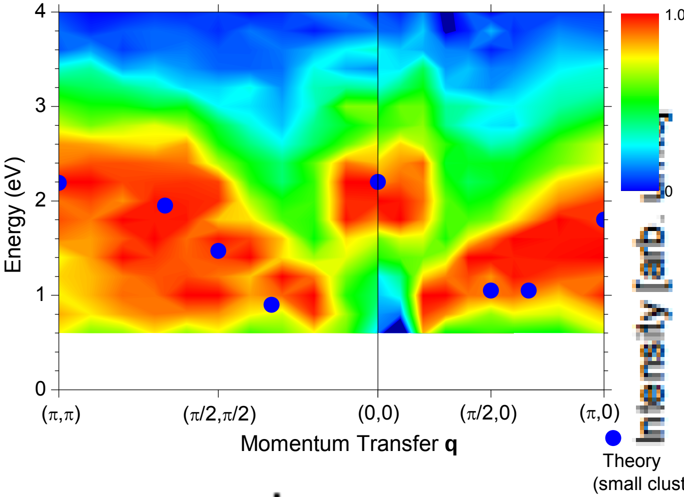
(ii) Hole doped system

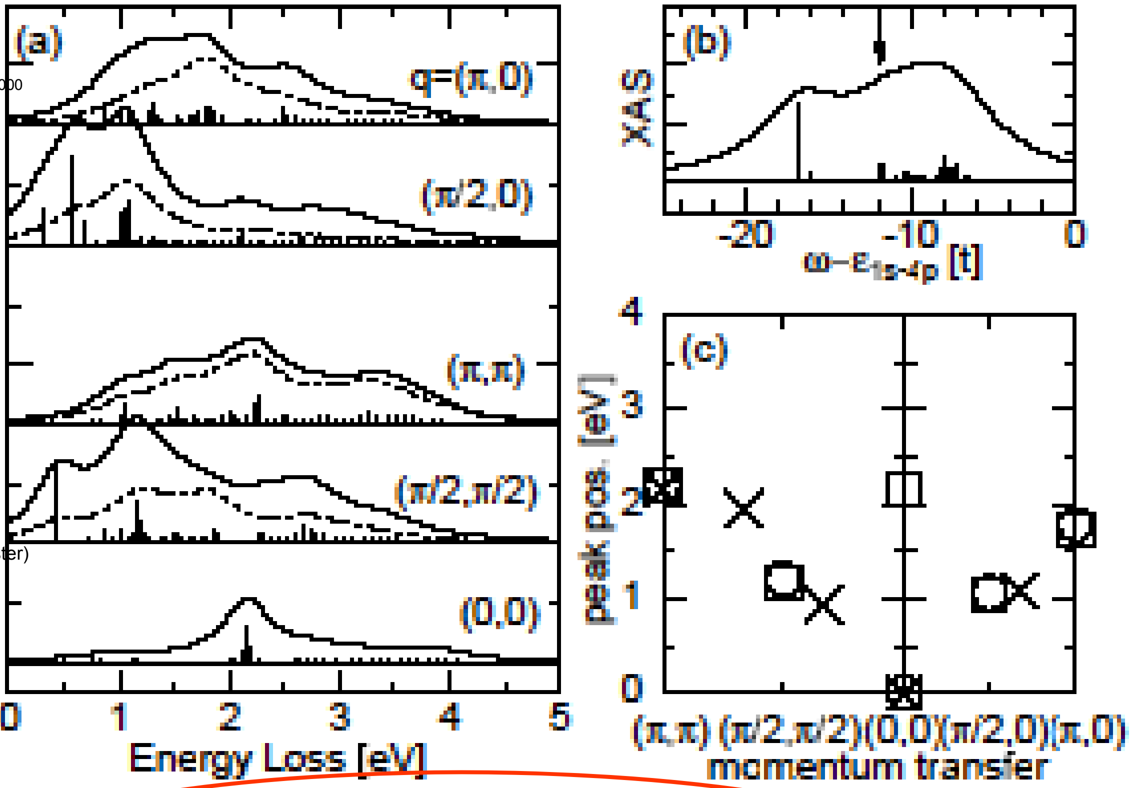
(iii) Electron doped system

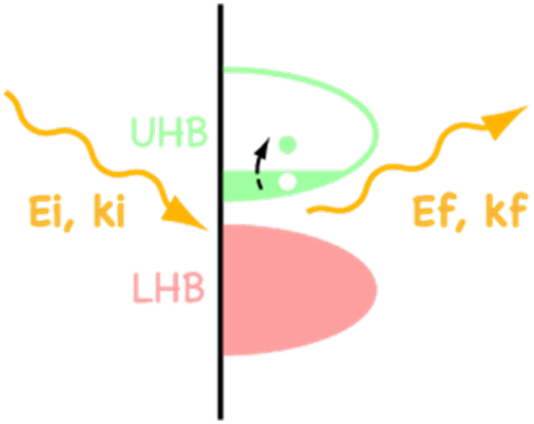
Electron doping

X=0.15

Calculated by K. Tsutsui







—— RIXS spectra

······· Dynamical density response function

The electron involved in dynamical density response function can be selected by RIXS !

K. Ishii, J. M. et al.,PRL. 94 ('05)207003

toward future

Required energy resolution

- Inter-band excitation: "U"~2eV

  Excitations across the Mott / charge-transfer gap

  ΔE~0.5eV

- Intra-band excitation: "*t*"~0.4eV

  Excitations within bands across the Fermi level

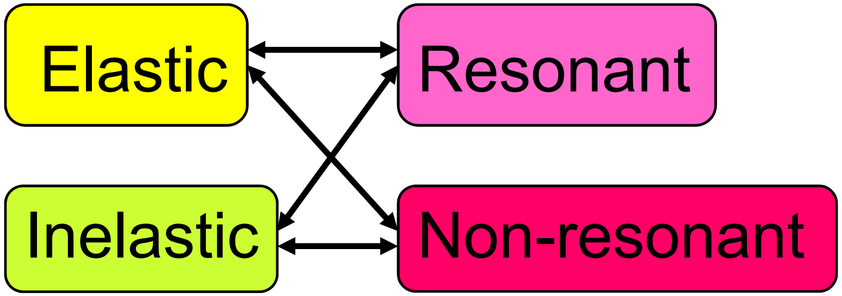
  ΔE~0.1eV

- Low-energy excitation: "*J*"~0.1eV

  Excitations related to the spin degree of freedom

  ΔE~0.05eV

Importance of the collaboration between Experiment and theory



Elastic

Resonant

Inelastic

Non-resonant

Actinide compounds

Transition metal compounds

Dynamical structure factor of IXS in the phonon energy region

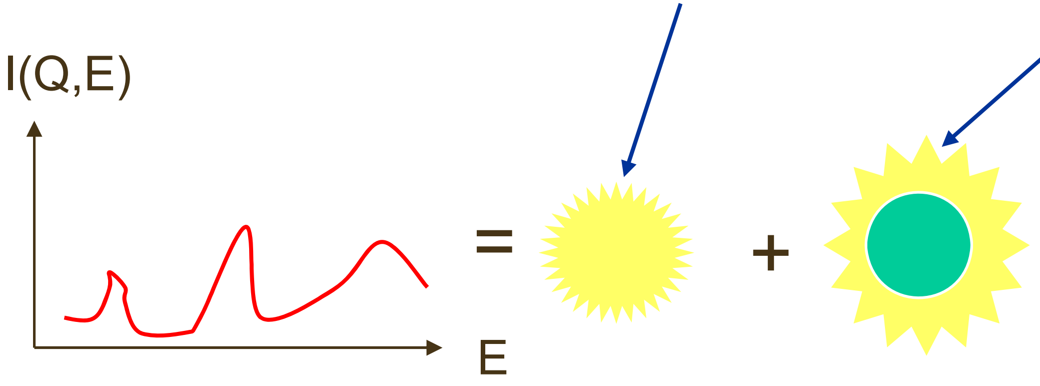
$$\varepsilon(Q,\omega)=\varepsilon_{el}(Q,\omega)+\varepsilon_{ion}(Q,\omega)-1$$

$$\downarrow$$

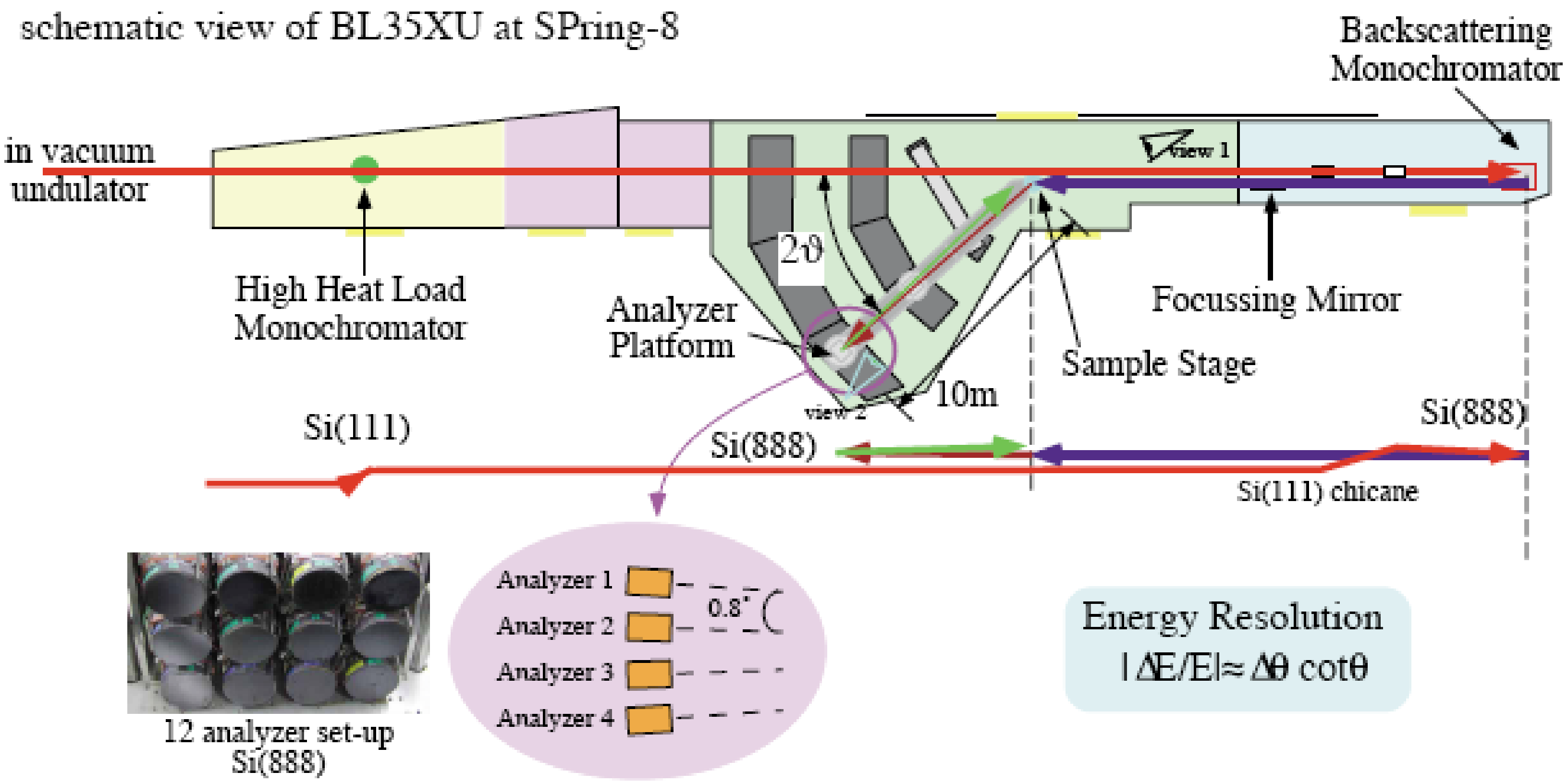
$$\chi(Q,\omega)= -(Q^2/4\pi^2N)\ 1/\varepsilon(Q,\omega)$$

$$\downarrow$$

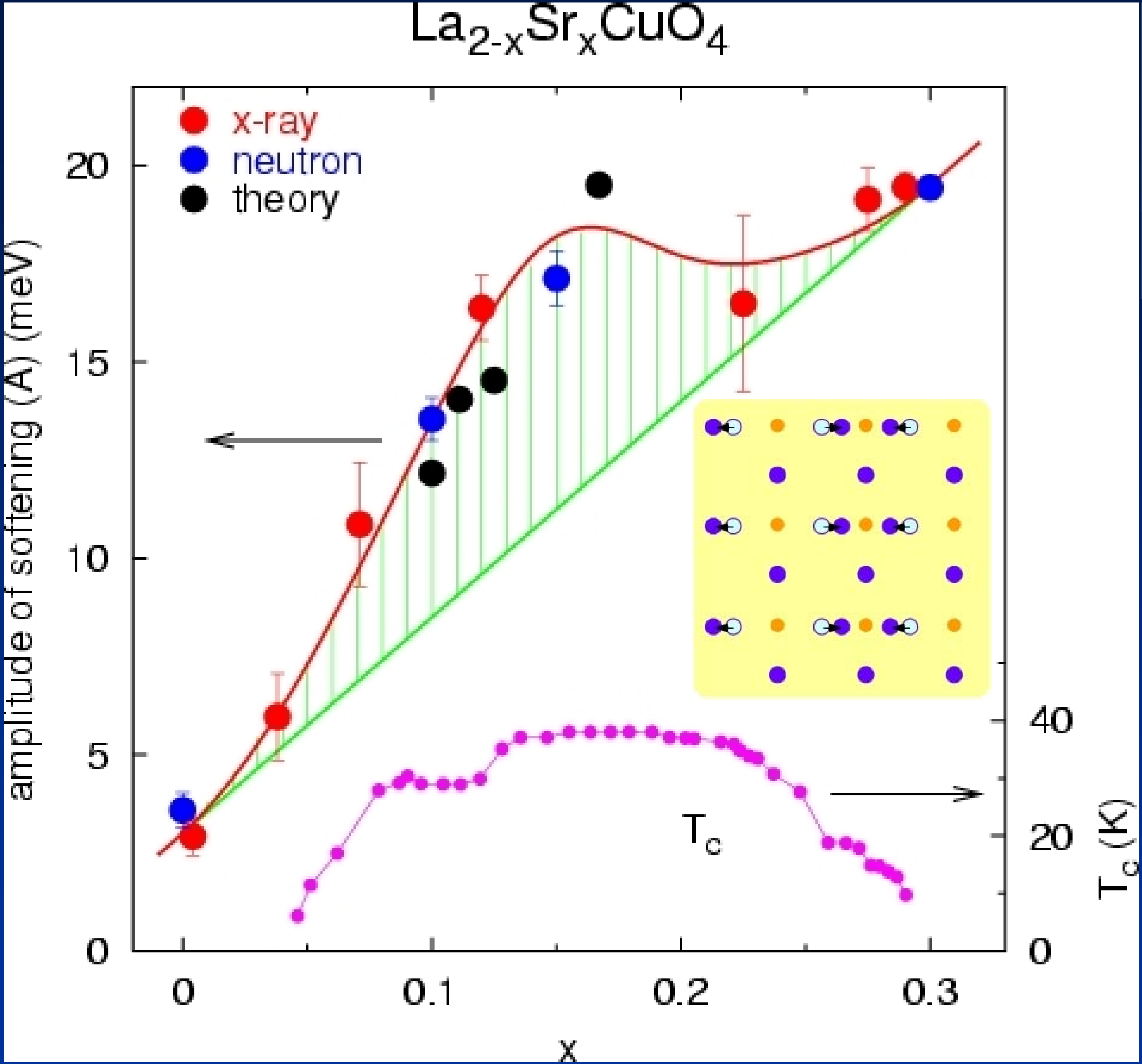
$$I(Q,\omega)= F(\varepsilon_{el})+G(\varepsilon_{el})\cdot H(\varepsilon_{ion})$$



What does the phonon play a role on superconducting?



Incident E(keV)	Resolution(meV)	Spot size(μm^2)
21.747	1.6	60 x 80
15.816	6.0	120 x 90

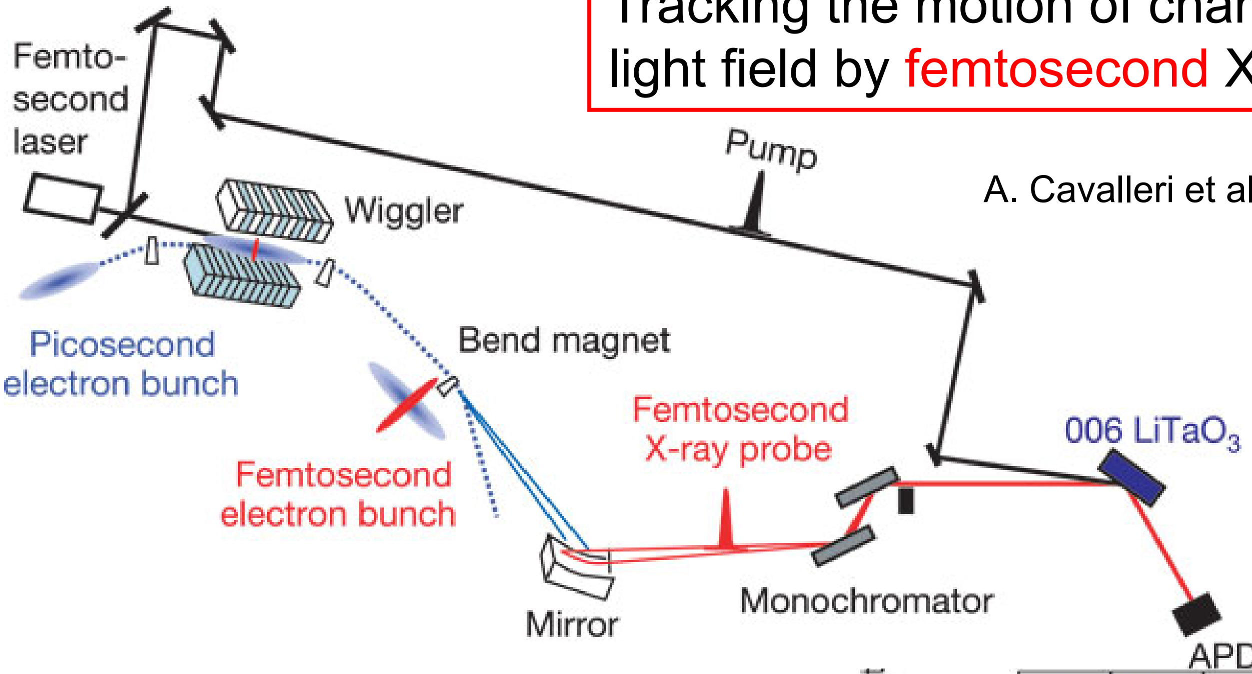


T. Fukuda, J. Mizuki,
et al., P. R. B. in press

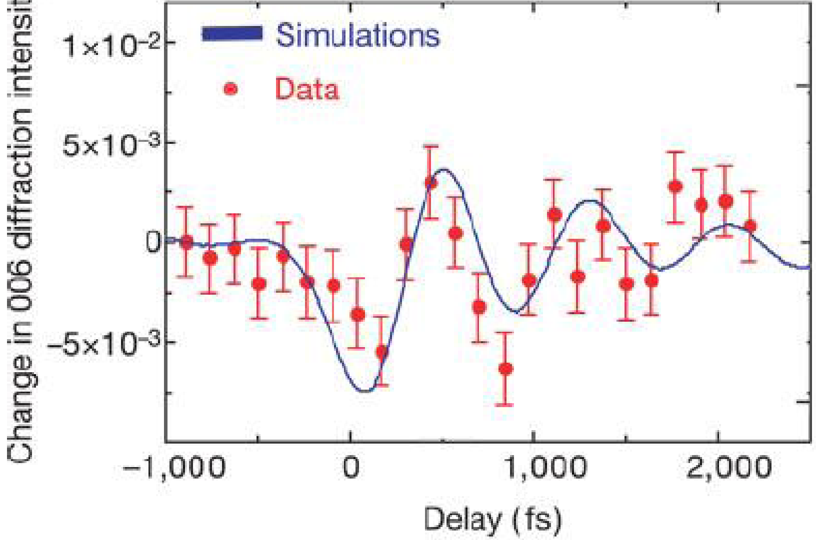
Dynamics in time domain

Tracking the motion of charges in a terahertz light field by femtosecond X-ray diffraction

A. Cavalleri et al., Nature 442 (‘06) 664

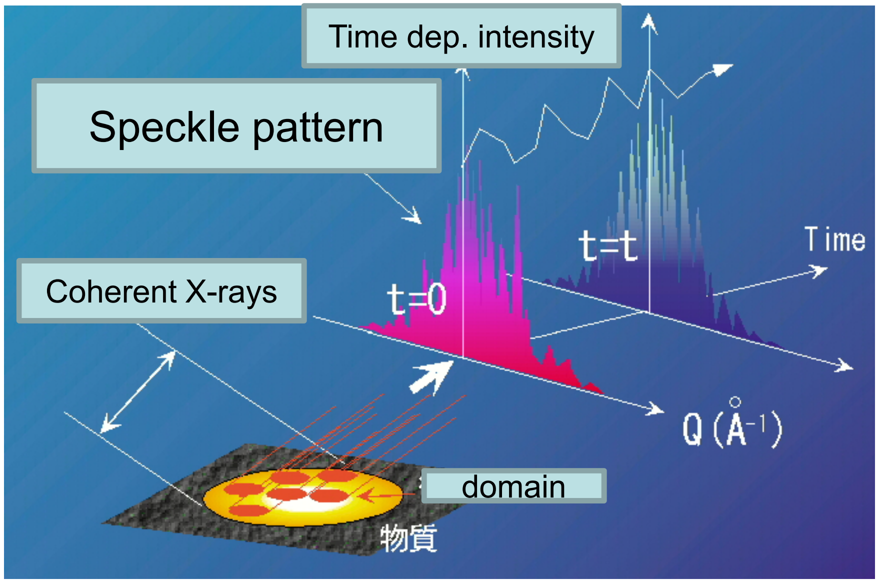


Time-dependent
Structure factor

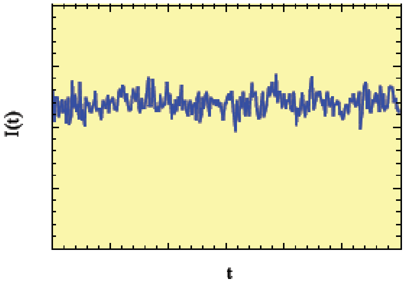


Excitation of phonon polaritons in LiTaO3

Domain Dynamics by Coherent X-ray Diffraction

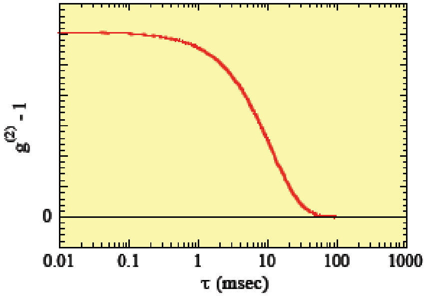


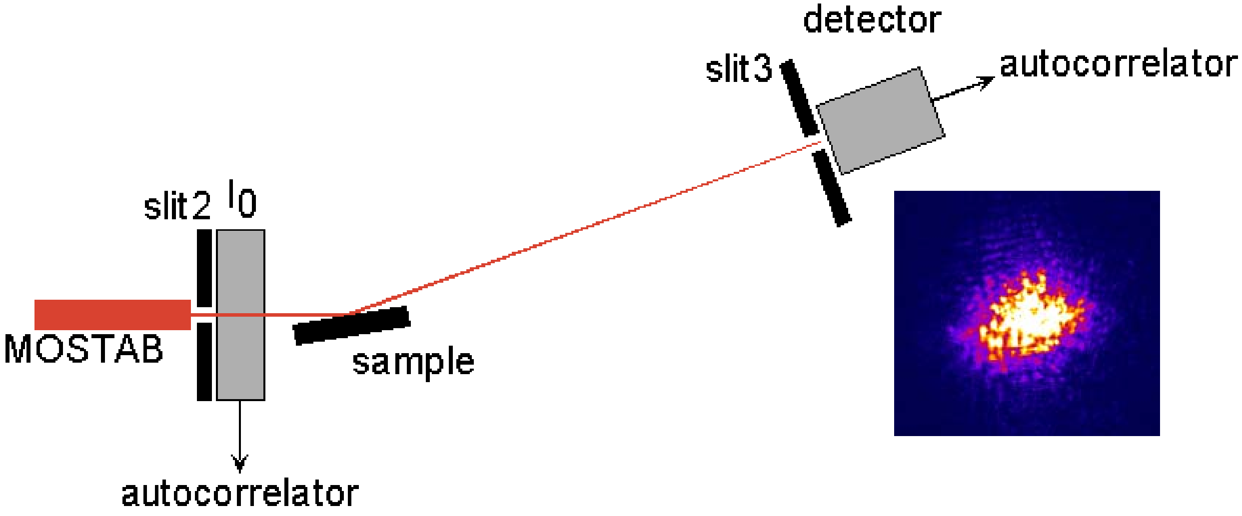
Intensity fluctuation by time



Self-correlation

$$g^{(2)}(\tau)=\frac{\langle I(t+\tau)I(t)\rangle}{\langle I(t)\rangle^2}$$





Future direction for SR performance:

more lower emittance → better spatial coherence

more shorter pulse duration → better time resolution

↓

XFEL

Coherence：Bose condensation

Photon number in a coherent volume

Longitudinal (temporal) coherence: $\sigma_t \sim 2h/\Delta\omega$

Transverse (spatial) coherence: $\sigma_{x,y} \sim \lambda \cdot L/2\pi S_{X,Y}$

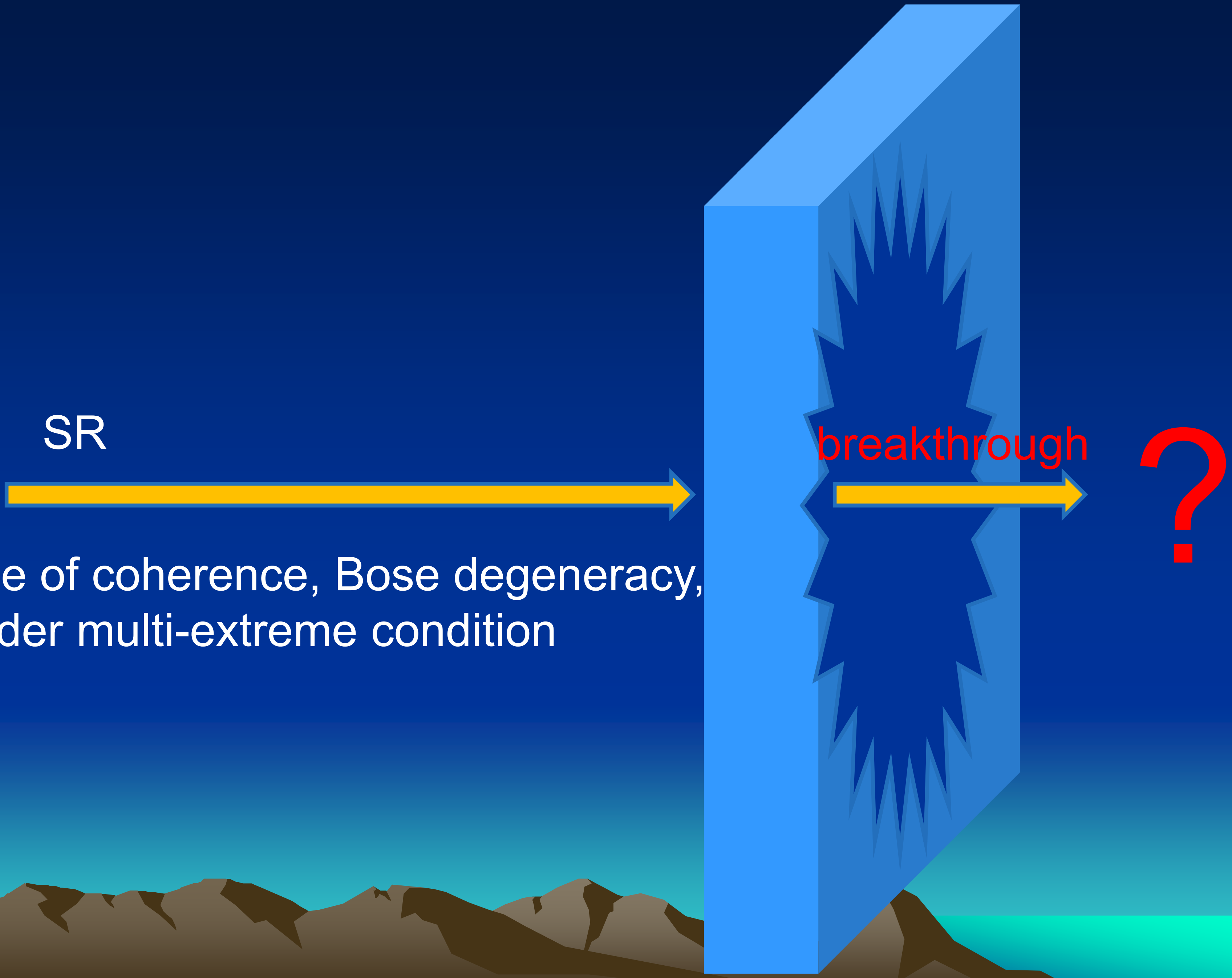
Coherent volume: $V_C = \sigma_X \cdot \sigma_Y \cdot \sigma_t = \lambda^2 \cdot L^2 \cdot 2h/(4\pi^2 \cdot S^2 \cdot \Delta\omega)$

Beam Volume: $V = \Delta X \cdot \Delta Y \cdot \Delta t$

Bose degeneracy: $\delta = n \cdot V_C/V \sim 0.1$ for SPring-8

for XFEL(SASE-mode): $\delta \sim 10^{10}$!!

non-linear optical phenomena



SR

Use of coherence, Bose degeneracy, under multi-extreme condition

breakthrough

?