The idea how to measure dynamical charge susceptibility combined with X-ray and Neutron inelastic scattering technique

- How to measure $\varepsilon(Q, \omega)$ -

Jun'ichiro Mizuki

Quantum Beam Science Directorate
Japan Atomic Energy Agency
(JAEA)

Inelastic X-ray scattering from a point of view of materials science

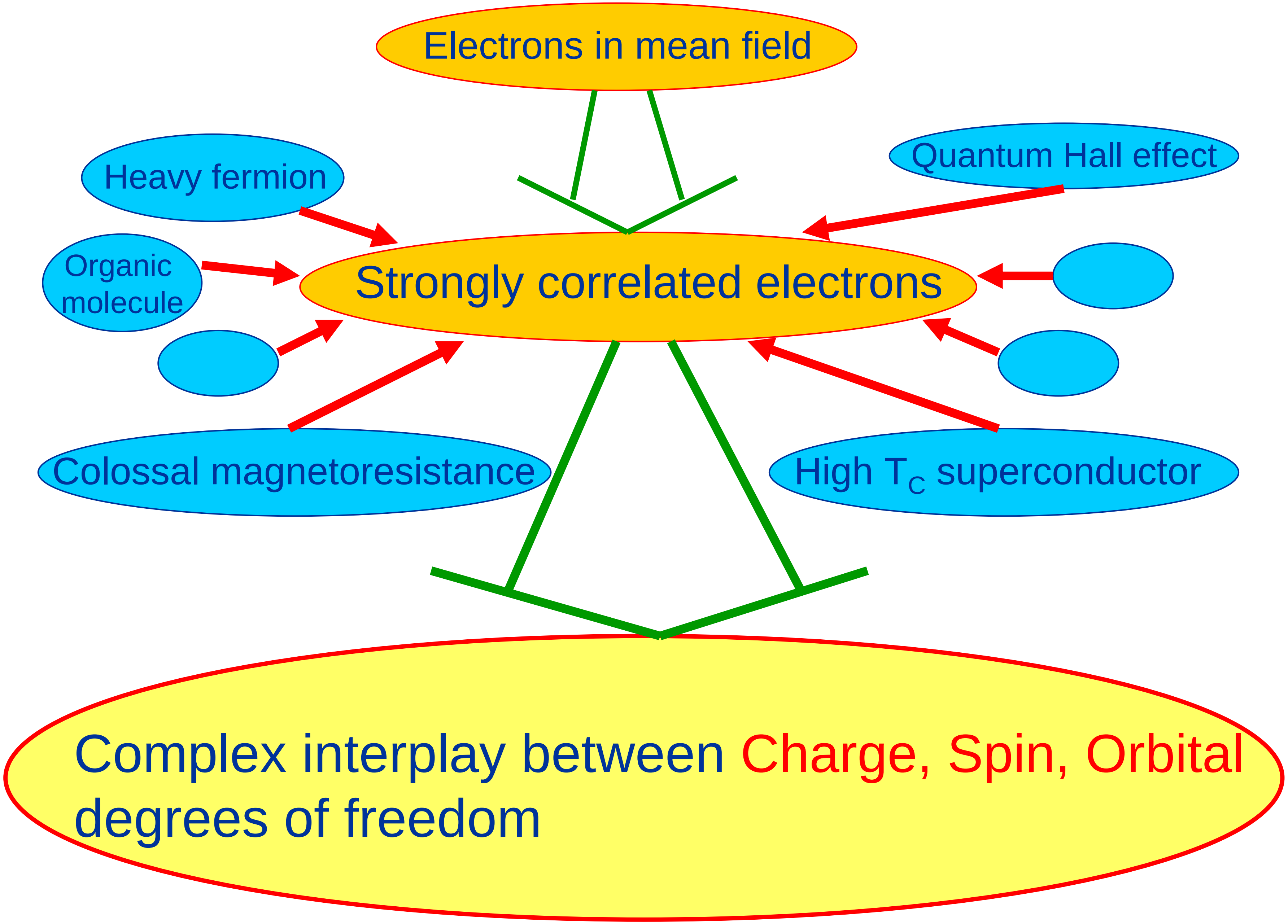
1. Why IXS ?

2. Current situations of IXS research focused on High T_C superconductors

   (a) Resonant IXS : charge excitation
   (b) non Resonant IXS (NIXS) : phonon

3. Prospective RIXS , NIXS)

   Idea to measure dynamical dielectric function in the room Temperature energy range.



Electrons in mean field

Heavy fermion

Quantum Hall effect

Organic molecule

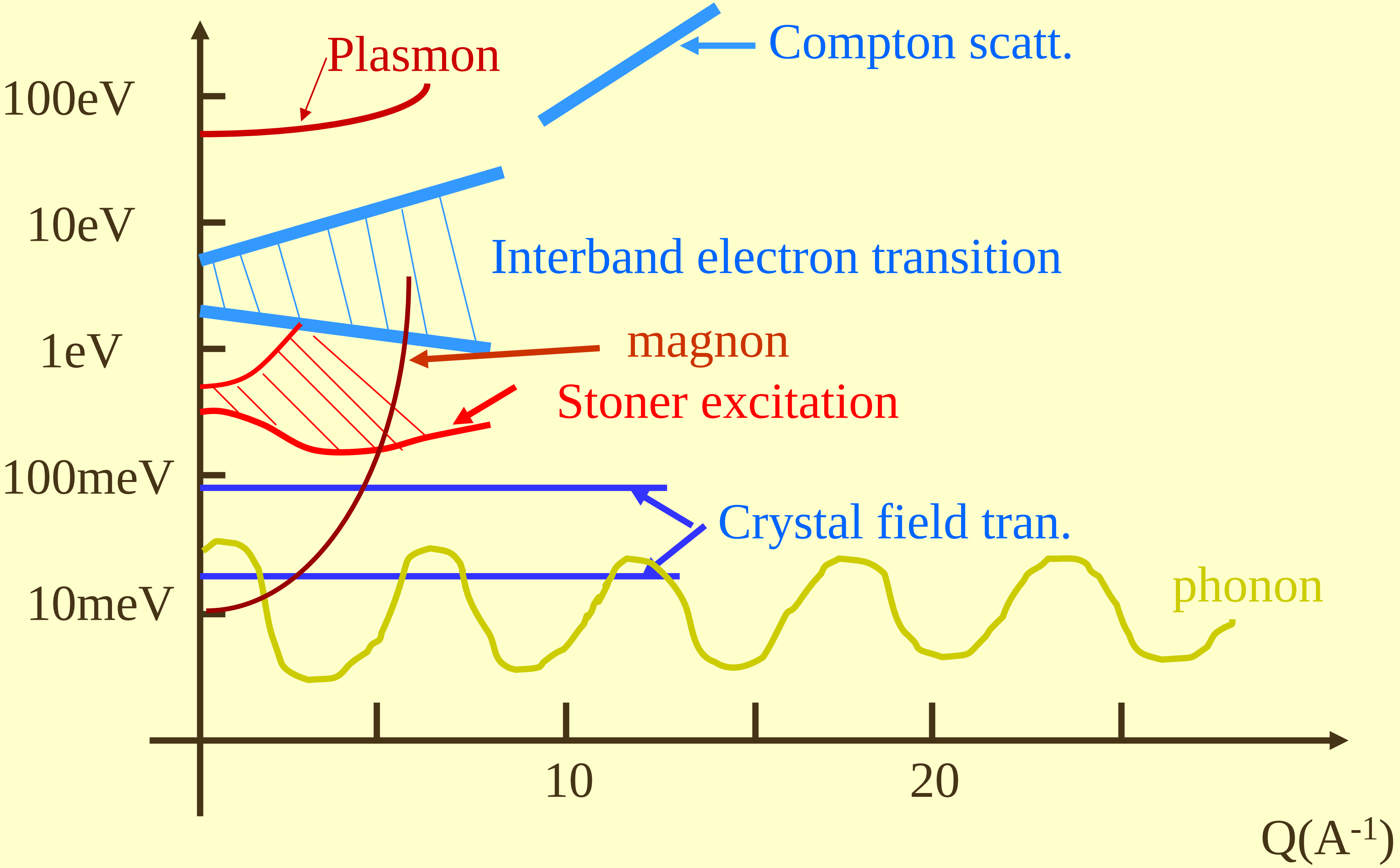
Strongly correlated electrons

Colossal magnetoresistance

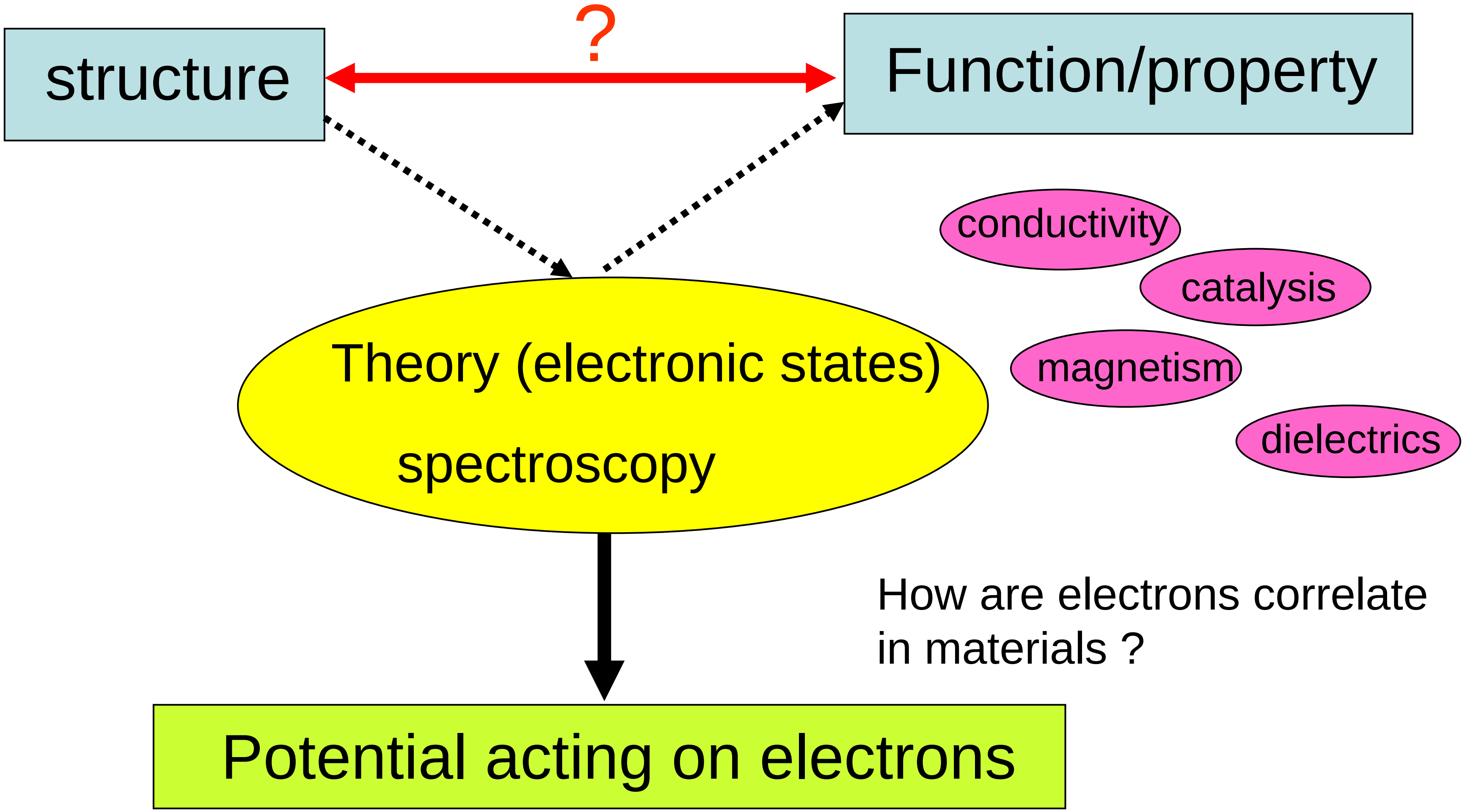
High T_C superconductor

Complex interplay between Charge, Spin, Orbital degrees of freedom

Elementary Excitations in Solids



Actor /actress in materials is the electron !



structure

?

Function/property

conductivity

catalysis

magnetism

dielectrics

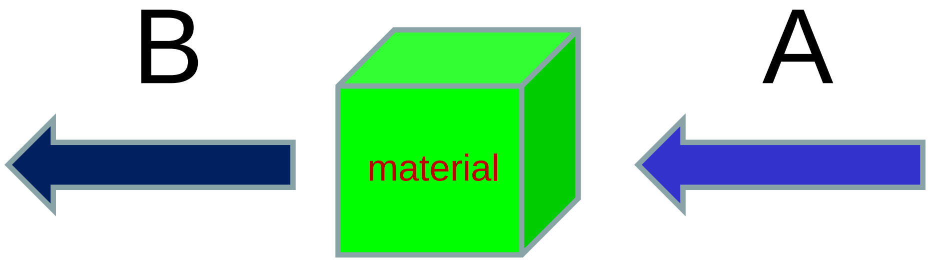
Theory (electronic states)

spectroscopy

How are electrons correlate
in materials ?

Potential acting on electrons

Linear response theory



$$B = \chi A$$

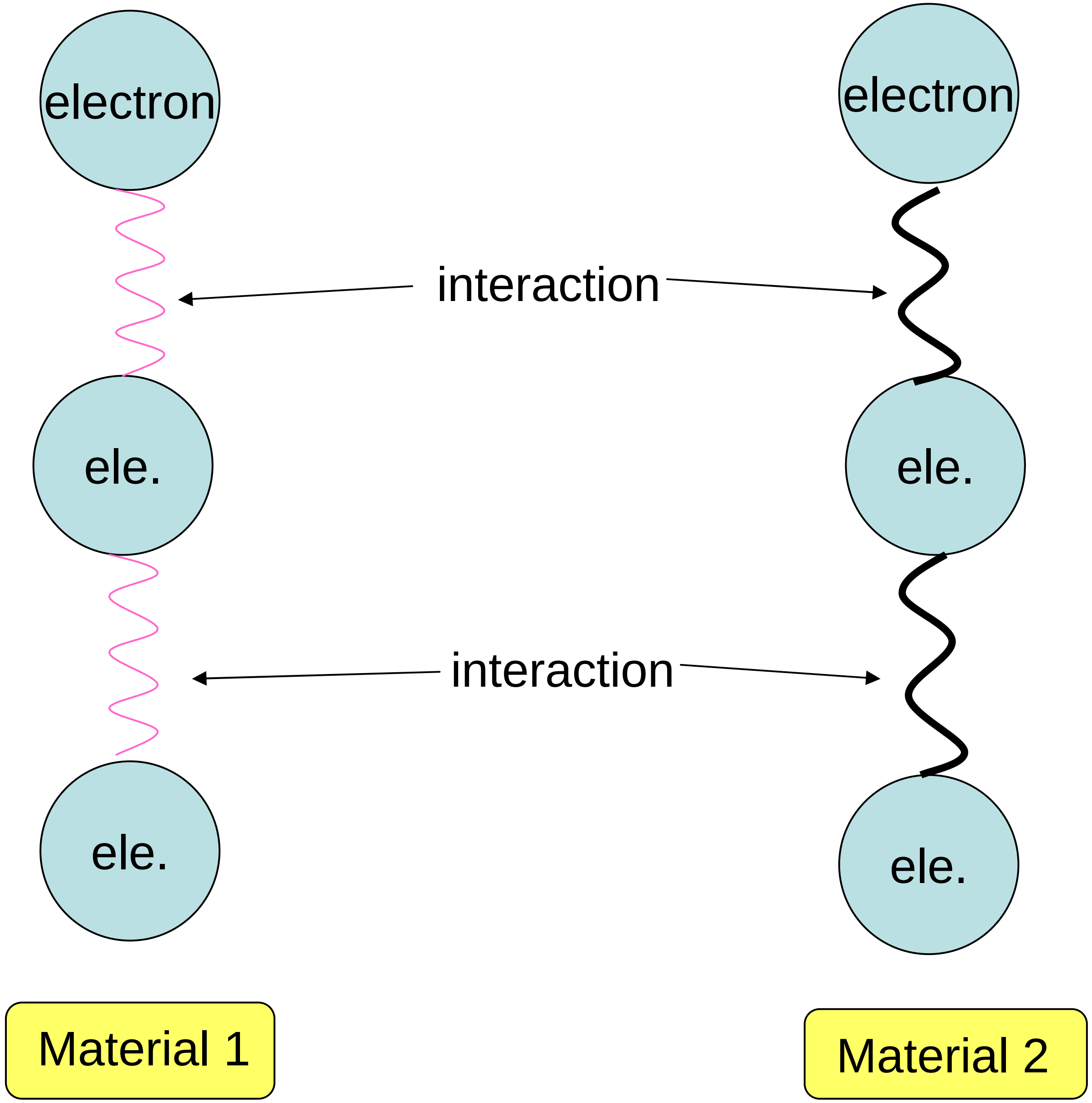
polarization ← Electric field

magnetism ← Magnetic field

strain ← stress

etc. etc.

For dynamics study → Use of fluctuation – dissipation theorem



electron

electron

interaction

ele.

ele.

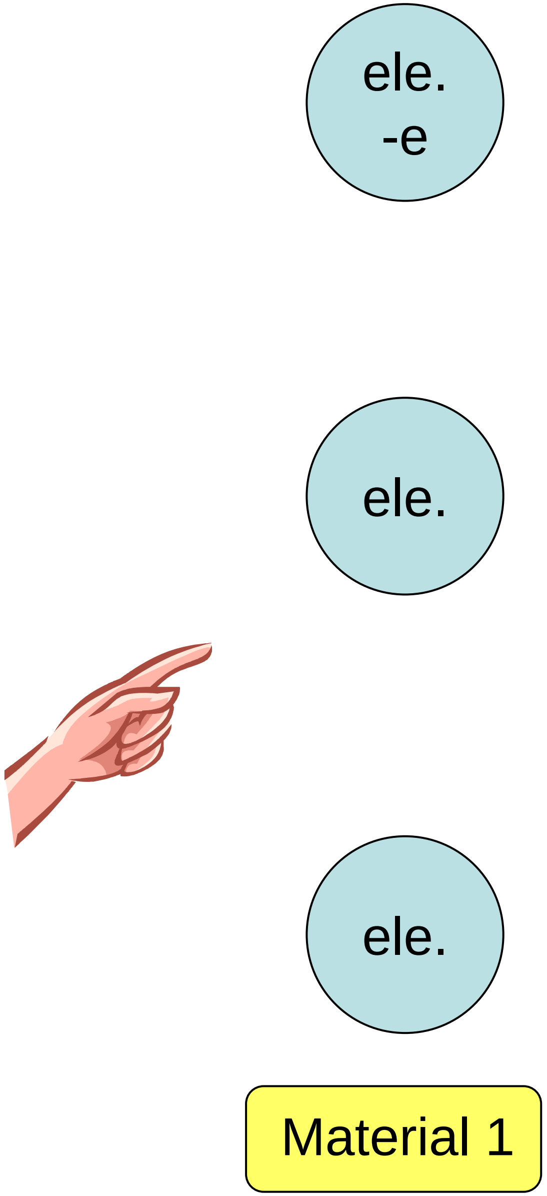
interaction

ele.

ele.

Material 1

Material 2

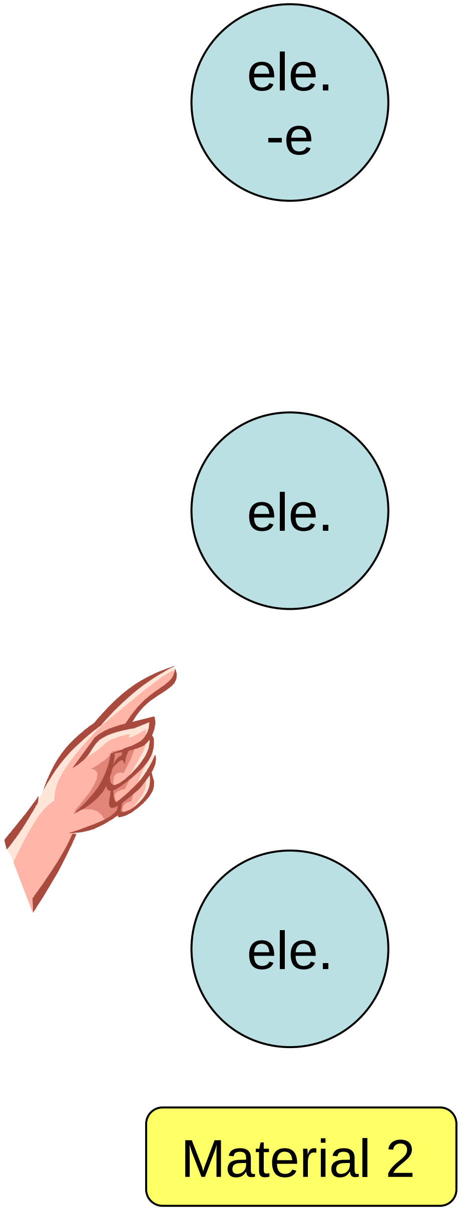


ele.
-e

ele.

ele.

Material 1



ele.
-e

ele.

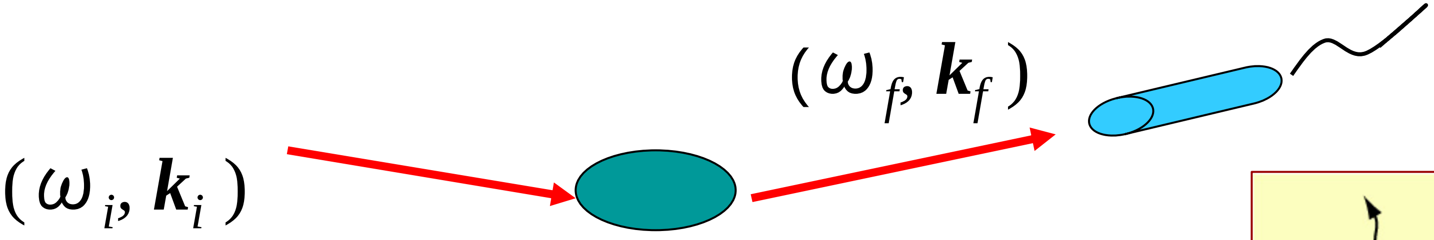
ele.

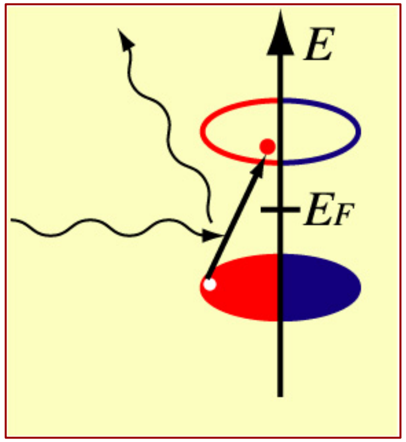
Material 2

How do we know the potential for electrons?

Dynamics!

Observation of dynamical properties of electrons and atoms





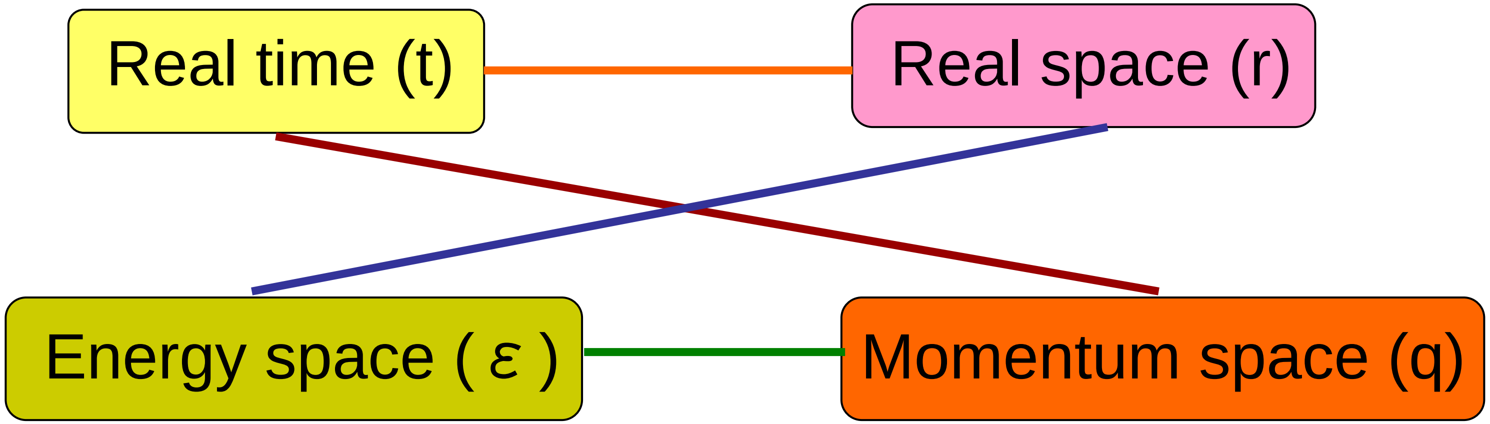
$$\mathbf{Q} = \boldsymbol{k}_i - \boldsymbol{k}_f$$

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

inelastic scattering!

Observation space

In what space should we measure physical properties?



Real time (t)

Real space (r)

Energy space (ε)

Momentum space (q)

Inelastic X-ray scattering

$$H = \sum_j \frac{1}{2m}\left(\mathbf{p}_j - \frac{e}{c}\mathbf{A}(\mathbf{r}_j)\right)^2 = \sum_j \left(\frac{\mathbf{p}_j^2}{2m} - \frac{e}{mc}\mathbf{A}(\mathbf{r}_j)\cdot\mathbf{p}_j + \frac{e^2}{2mc^2}\mathbf{A}(\mathbf{r}_j)^2\right)$$

$$\text{Fermi's golden rule}\quad I \propto \frac{2\pi}{\hbar}\left|\underline{\langle f|\mathbf{A}^2|i\rangle} + \underline{\frac{\langle f|\mathbf{A}\cdot\mathbf{p}|n\rangle\cdots\langle m|\mathbf{A}\cdot\mathbf{p}|i\rangle}{(E_i - E_m + \hbar\varpi - i\Gamma)}}\right|^2$$

- The first term: non-resonant inelastic scattering
  - All electrons（Ze）are contributed ⇒ phonon excitation

- The second term: resonant inelastic scattering (RIXS)
  - Electrons on the specific atom are contributed.
  - Resonance enhancement
  - Element specific ⇒ electronic excitation

Inelastic Scattering

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

Interaction of probe

Generalized susceptibility

For neutrons → Spin susceptibility
Nuclear susceptibility

For X-rays
For electrons } → Charge susceptibility

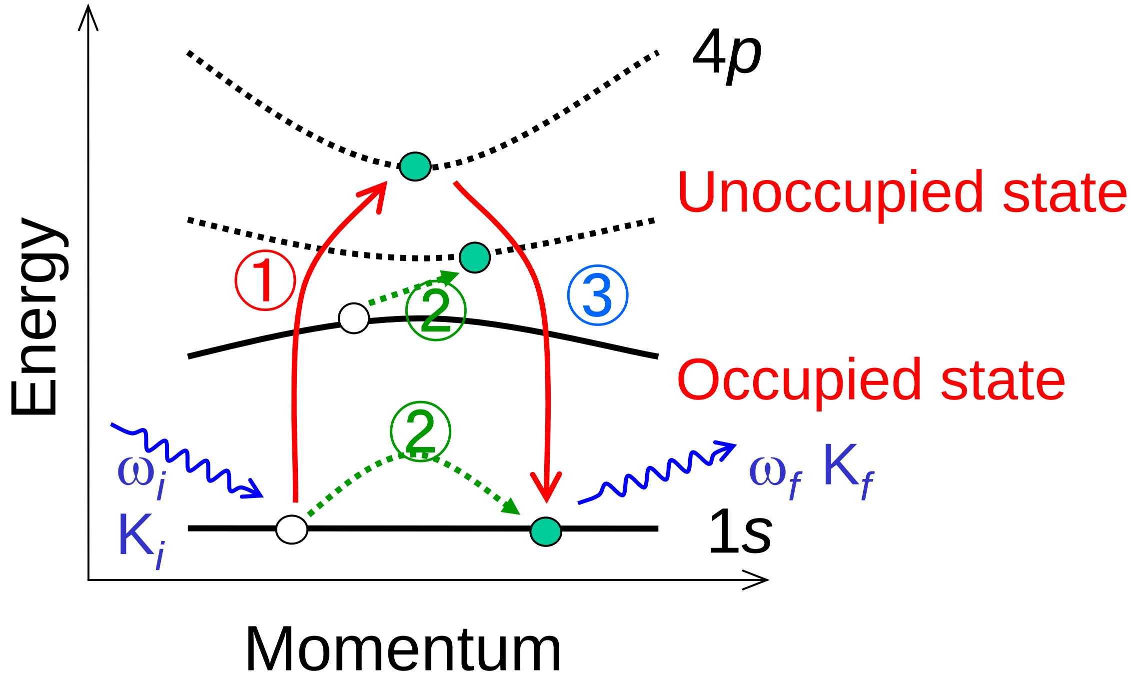
$$\Longrightarrow \quad \chi(Q,E) = -(Q^2/4\pi^2 N)\; 1/\underline{\varepsilon(Q,E)}$$

Dynamical dielectric function

Resonant Inelastic X-ray Scattering

In case of Cu-oxides:

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



① : absorption

② : 3d-electron excitation

③ : x-ray emission

Dynamical Dielectric Function (DDF) is important as they Determine the screening of electrostatic forces between extra charges in the lattice.

Dynamical Dielectric Function is a well-defined physical property: theoretical framework is well-defined.

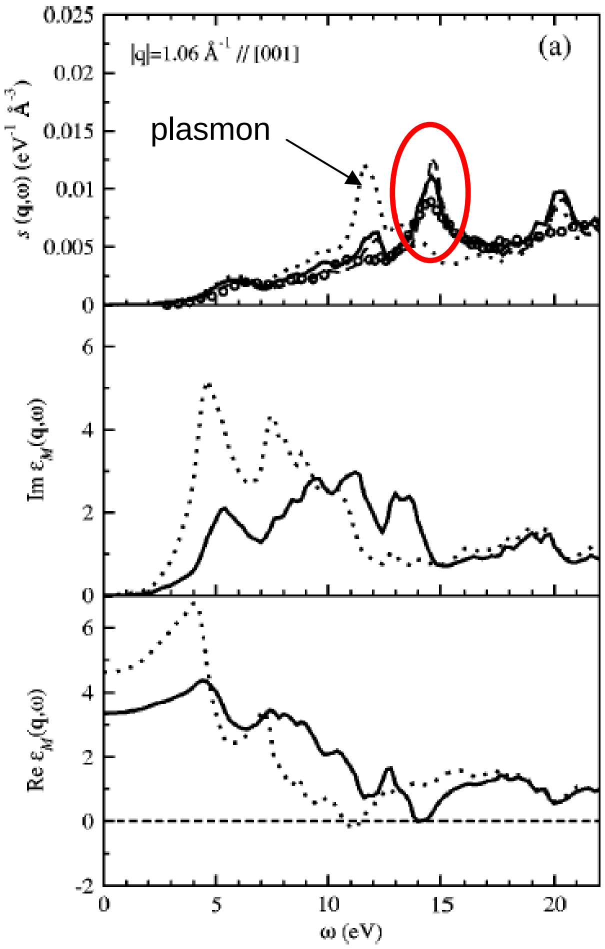
(We understand the important physics behind DDF)

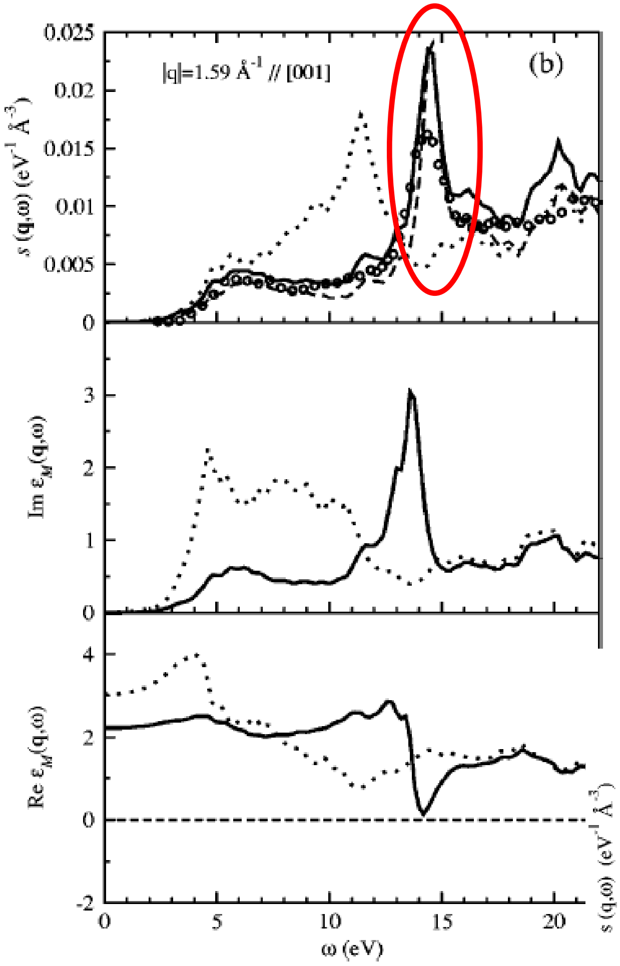
Direct comparison between experimental data and theoretical calculation can be done by

Inelastic X-ray Scattering (IXS)

Dynamical Structure Factor of TiO_2: Experiment & Theory

I. G. Gurtubay et. al., Phys. Rev. B70 ('04) 201201





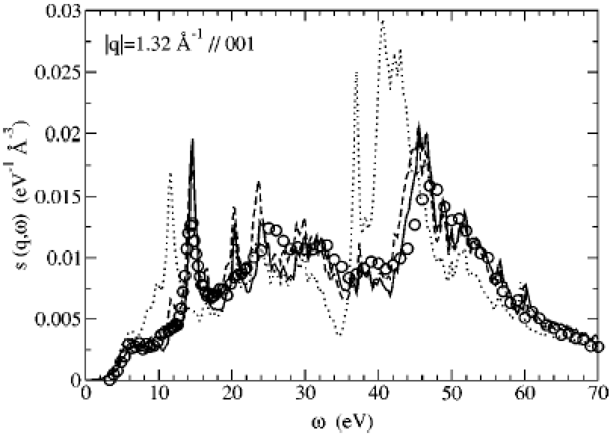
········ No crystal local-field effect

—— Crystal local-field effect

○ 14 eV peak:

- appeared by considering crystal local-field effect .
- persistent at large wave vector.
- nonmonotonic dependence on the wave vector.

Microscopic electric field acting on localized *d* -states



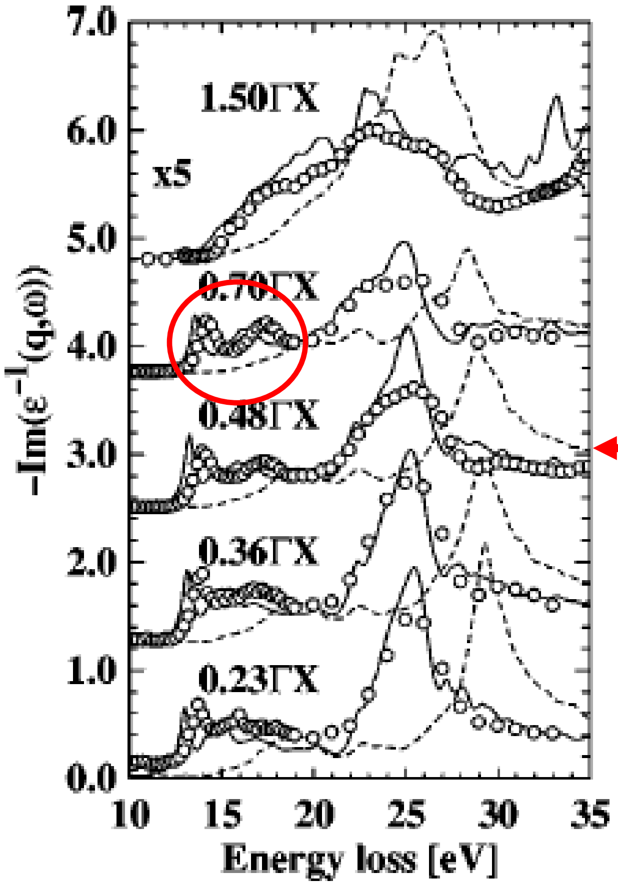
Effects of electron-hole interaction on the dynamic structure factor: Application to nonresonant inelastic x-ray scattering

J. A. Soininen
Department of Physics, POB 9, FIN-00014, University of Helsinki, .

Eric L. Shirley
Optical Technology Division, Physics Laboratory, National Institute of Standards and Technolc

(Received 24 January 2000)

P. R. B. 61 ('00) 16423



—— : including ele. –ho. inter.

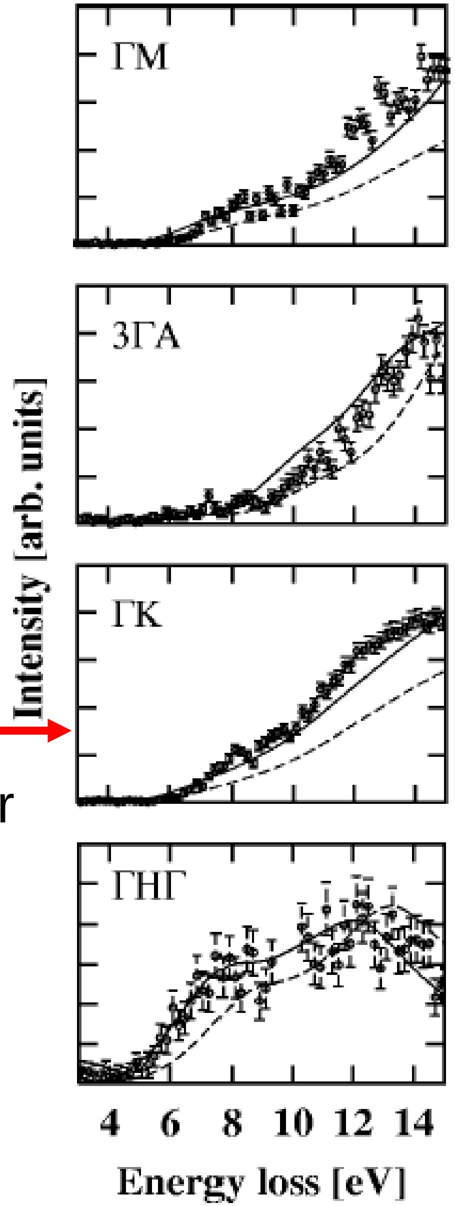
······· : not including ele.-ho. Inter.

LiF

insulator

GaN

semiconductor



More important energy region

$E < 1$ eV

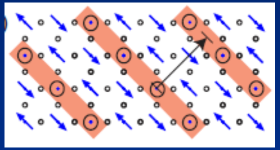
As an example: High T_C superconductors

Mechanism ?

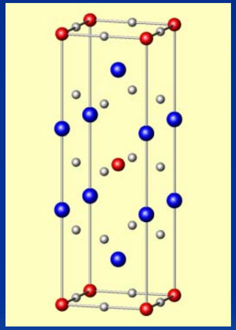
Couple to fluctuation of some order parameter

(Two electrons form a Cooper-pair by exchange with a boson.)

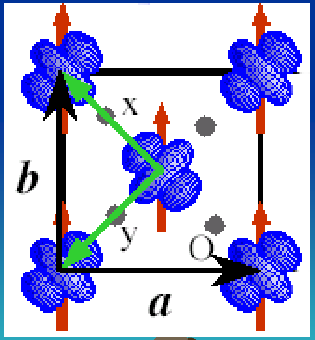
↓↑↓↑↓



Spin order?, Charge order?, Lattice order?,



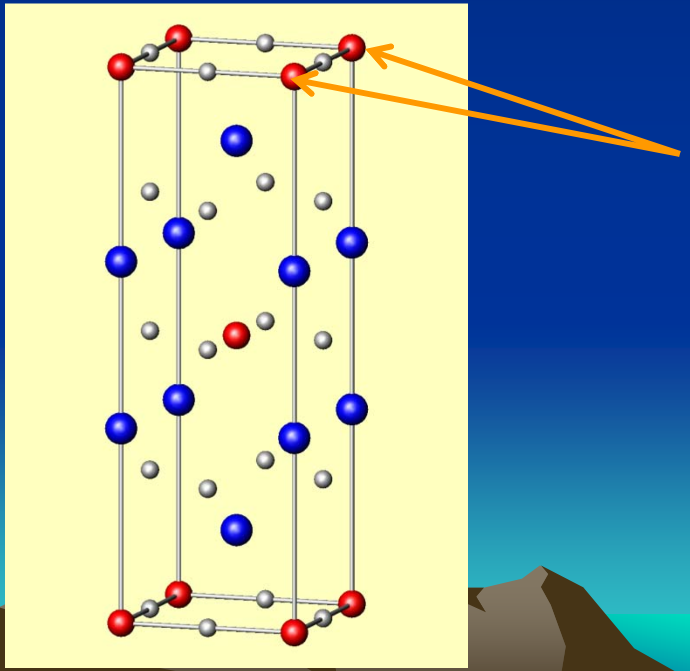
Orbital order?, or others?



Element selective charge excitation

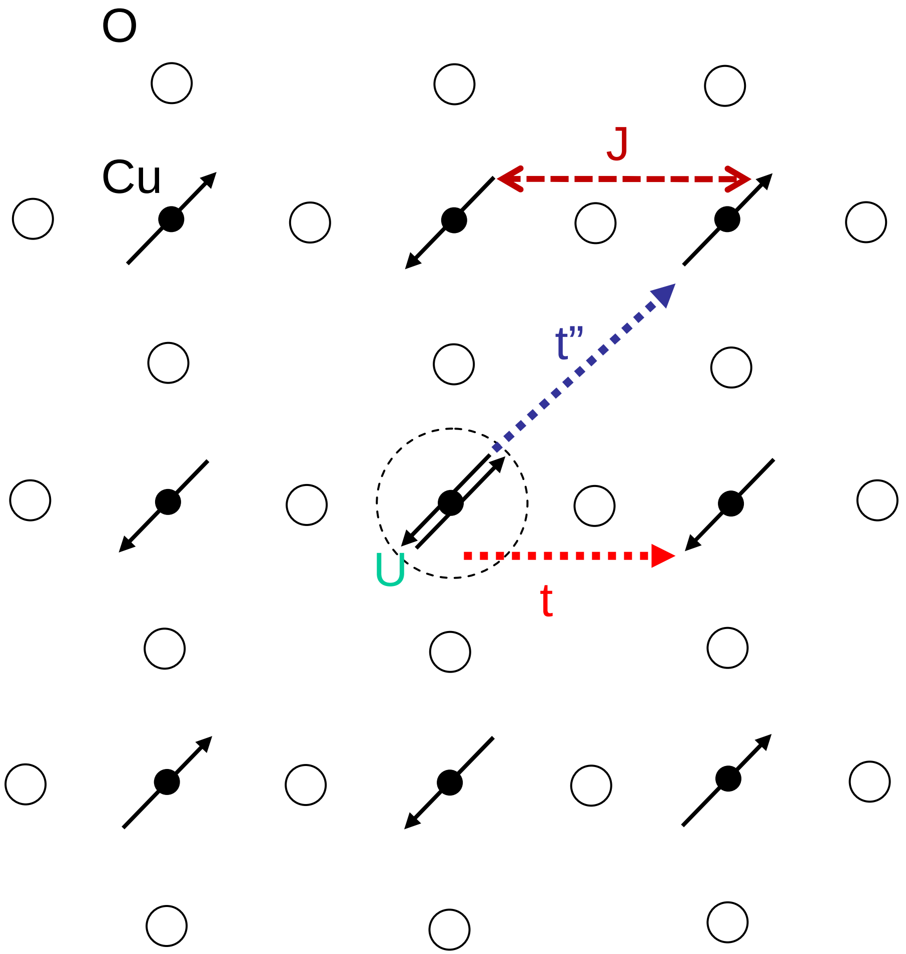
⇩

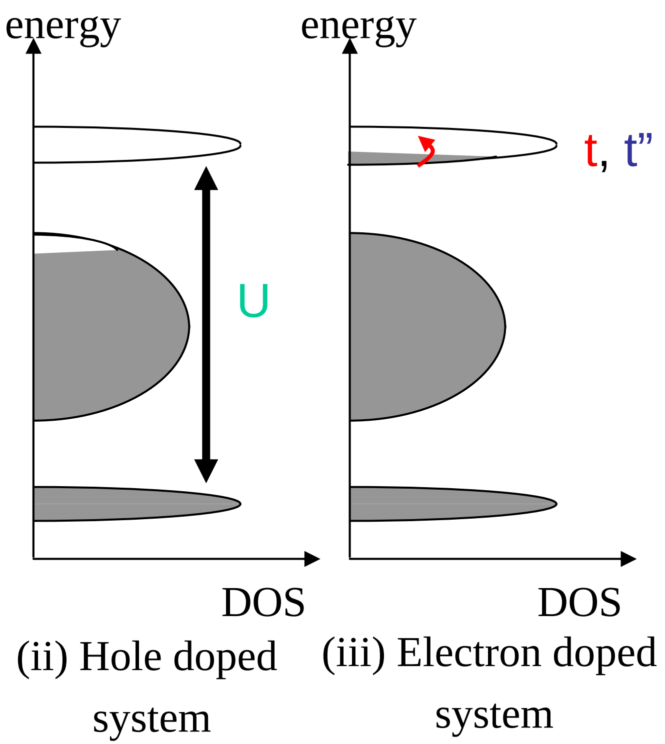
Use of Resonance



focus

Schematic diagram of electronic states





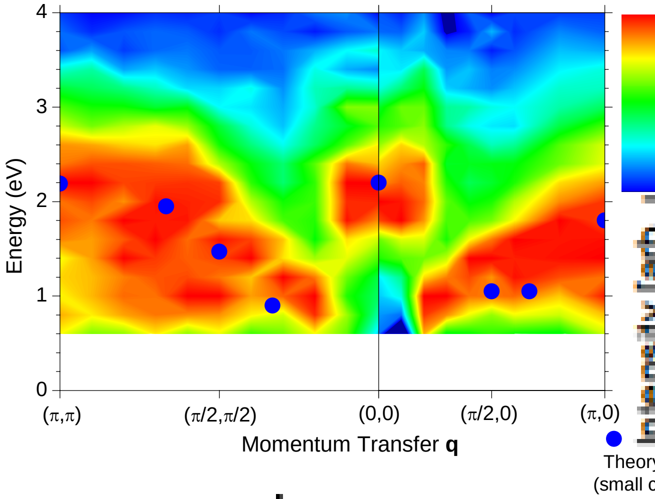
(ii) Hole doped system

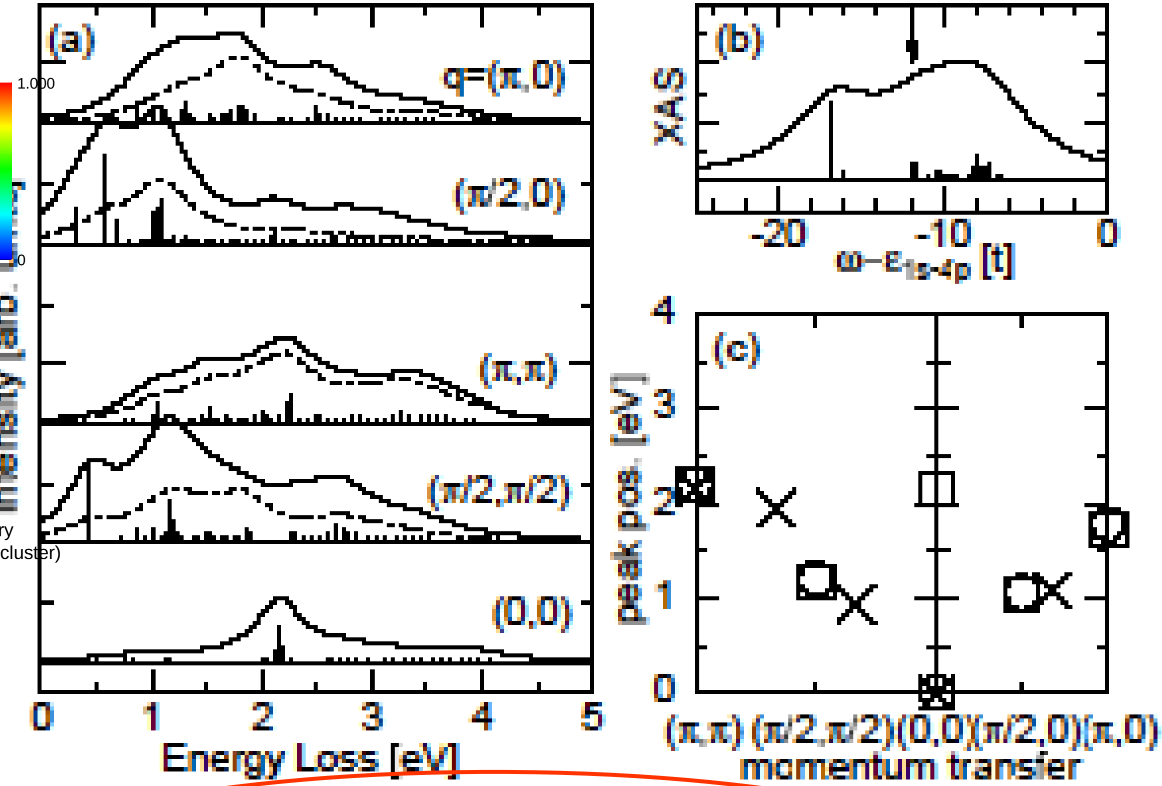
(iii) Electron doped system

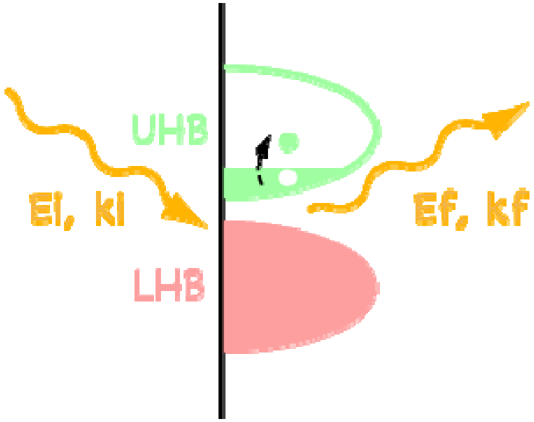
Electron doping

X=0.15

Calculated by K. Tsutsui







— RIXS spectra

······ Dynamical density response function

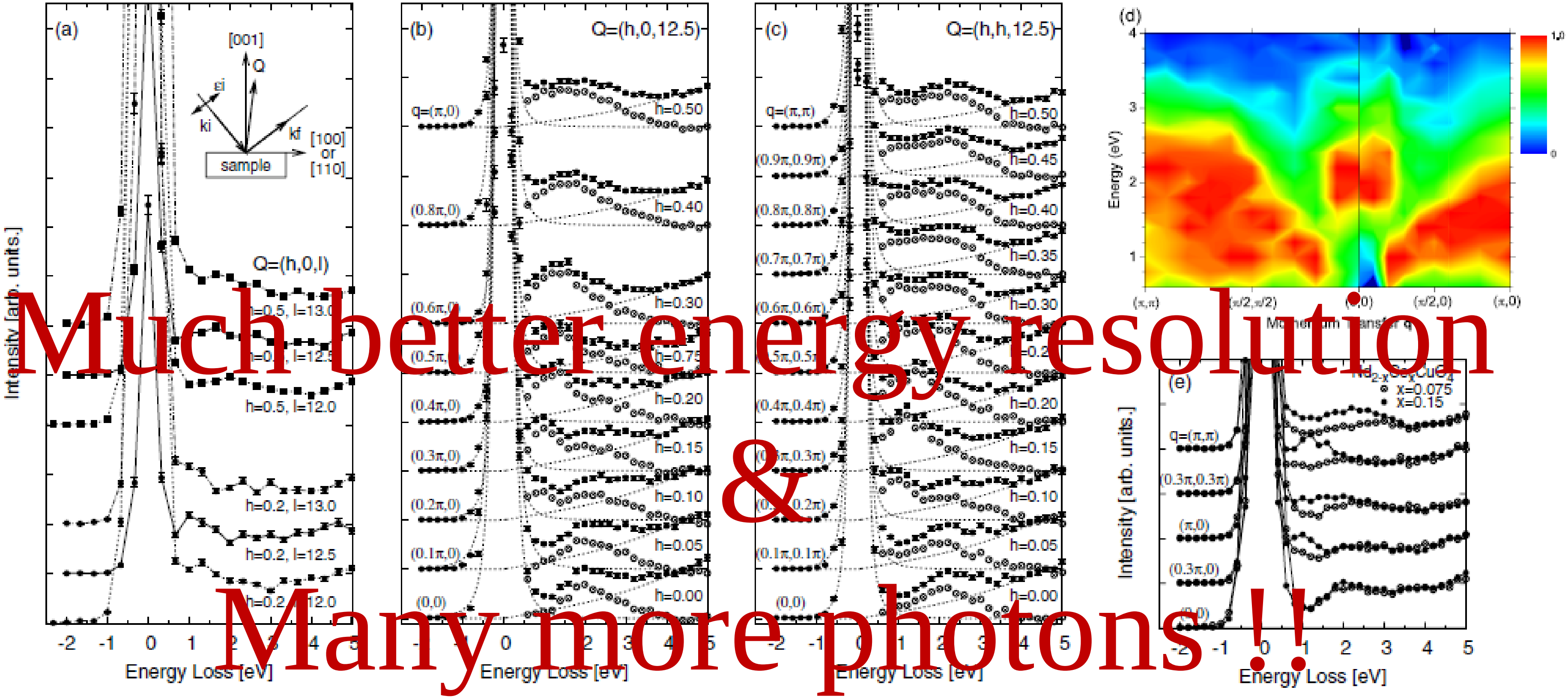
The electron involved in dynamical density response function can be selected by RIXS !

K. Ishii, J. M. et al.,P.R.L. 94 ('05)207003

Raw data of RIXS on $Nd_{2-X}Ce_XCuO_4$

Less than 1 c / sec.

⊿E～400 meV



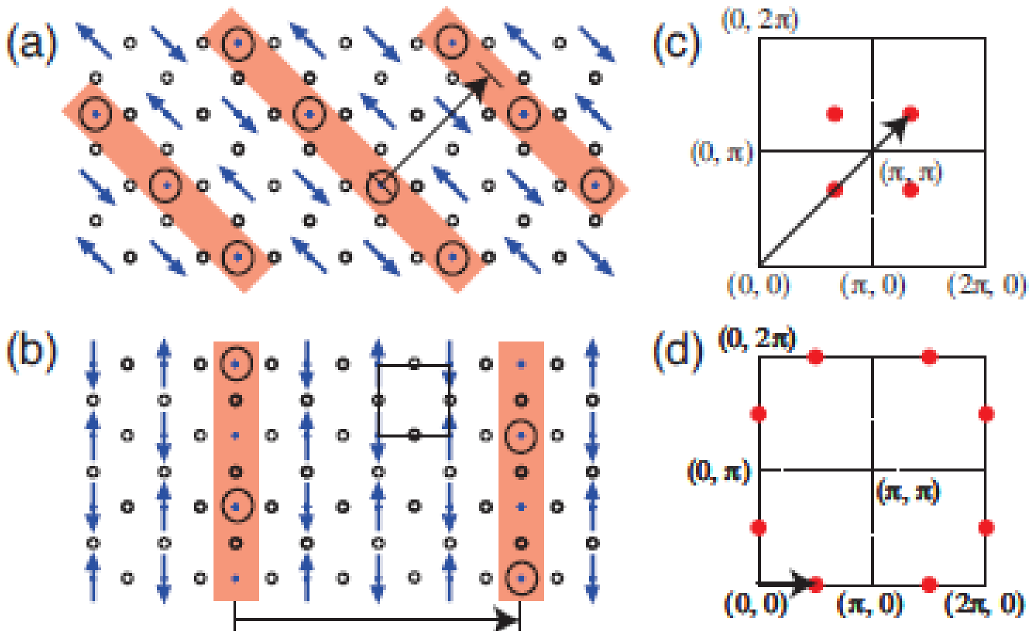
Much better energy resolution

&

Many more photons !!

K. Ishii, J. M. et al., P. R. L. 94 ('05) 207003

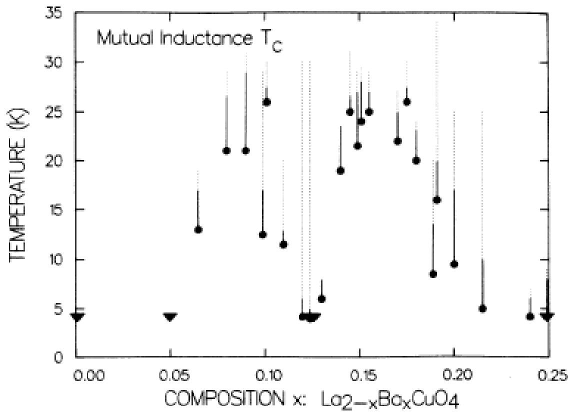
Relation between Striped-order and Superconductivity!?



$La_{2-X}Sr_XNiO_4$, X=1/3

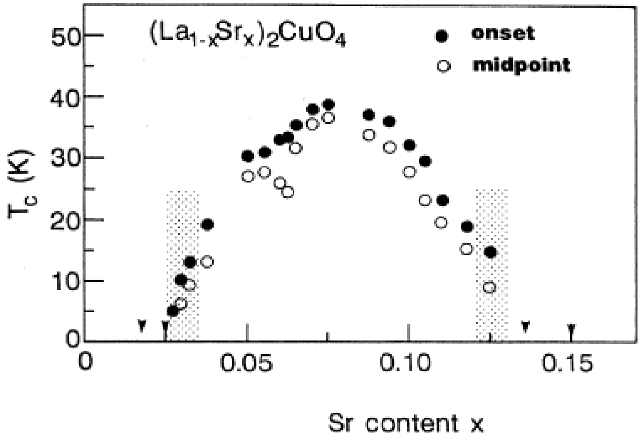
$La_{2-X}Ba_XCuO_4$, X=1/8

$La_{1.88}Sr_{0.12}CuO_4$



LBCO

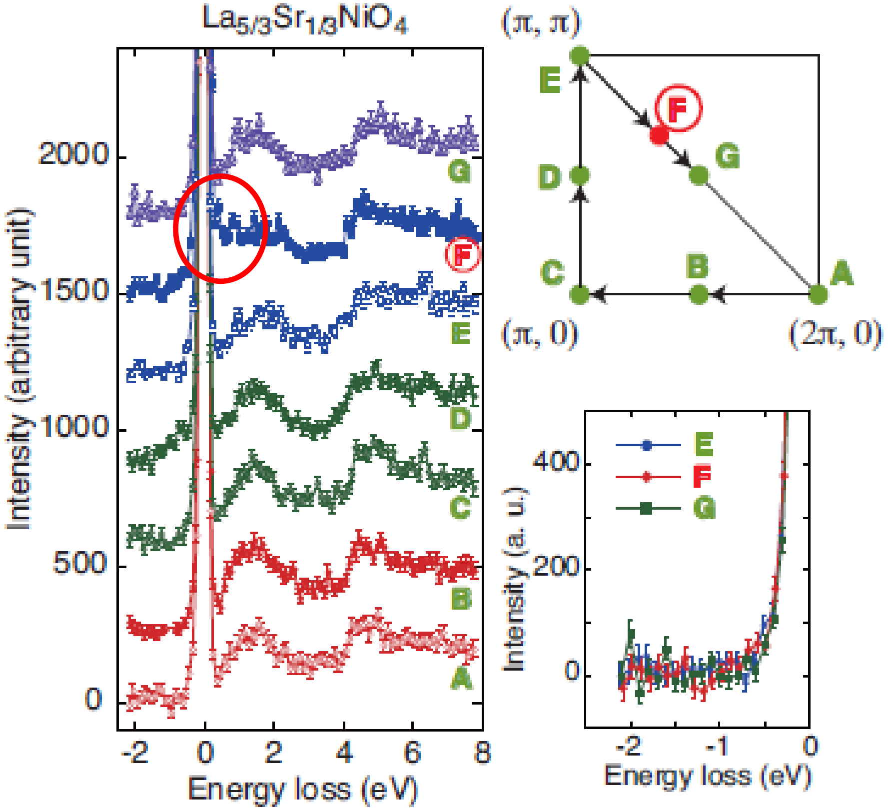
A. R. Moodenbaugh, et al., Phys. Rev. B 38 (‘88) 4596

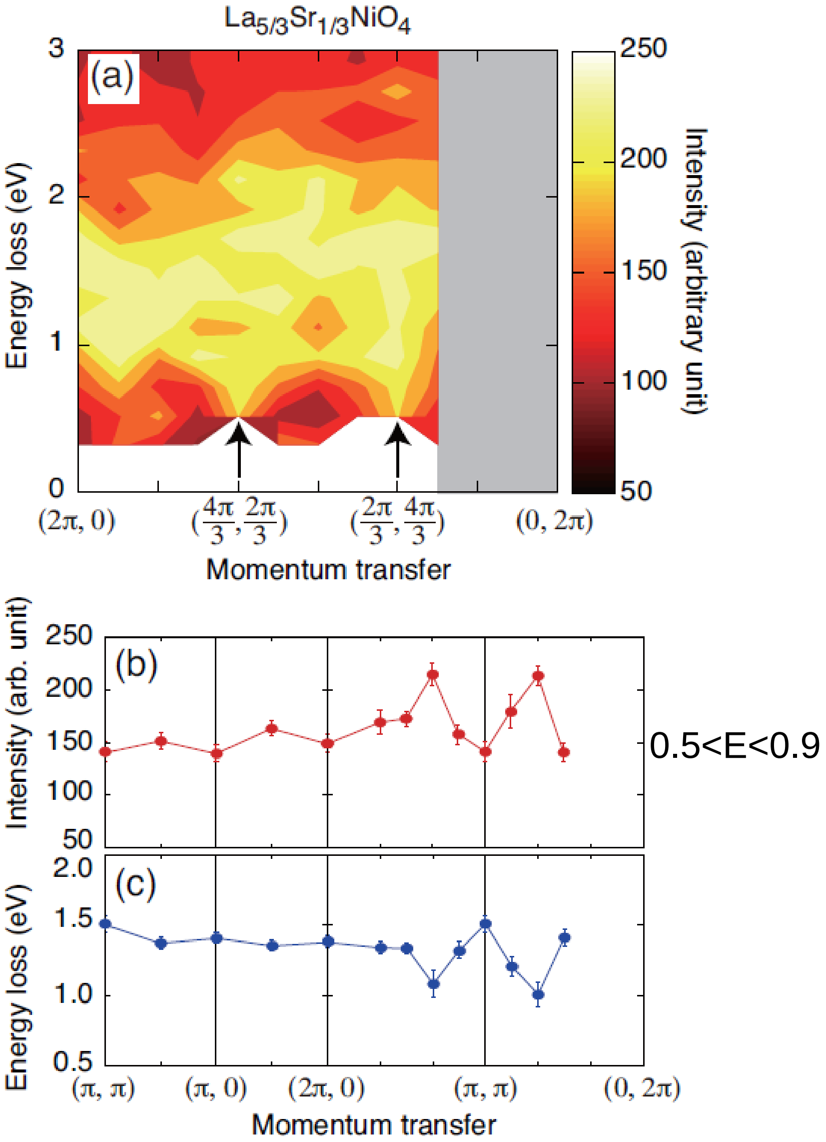


LSCO

H. Takagi, et al., Phys. Rev. B 40 (‘89) 2245

First Observation of charge excitation from charge stripe!?



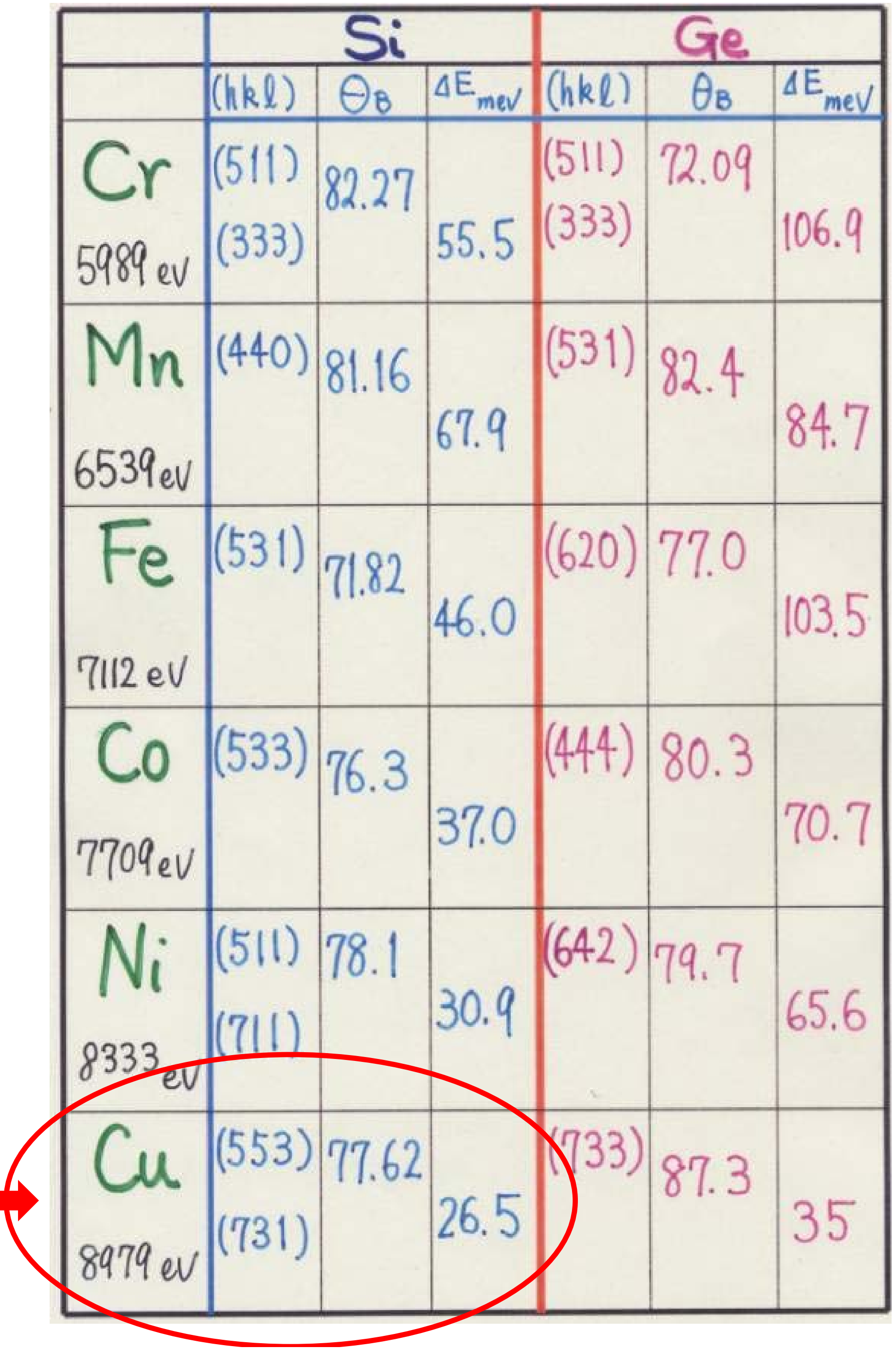


T_S ~180 K

~1.5 eV: valence → in-gap band
~4.5 eV: across the CT gap

S. Wakimoto, J. M. et al., P. R. L. 102 ('09)157001

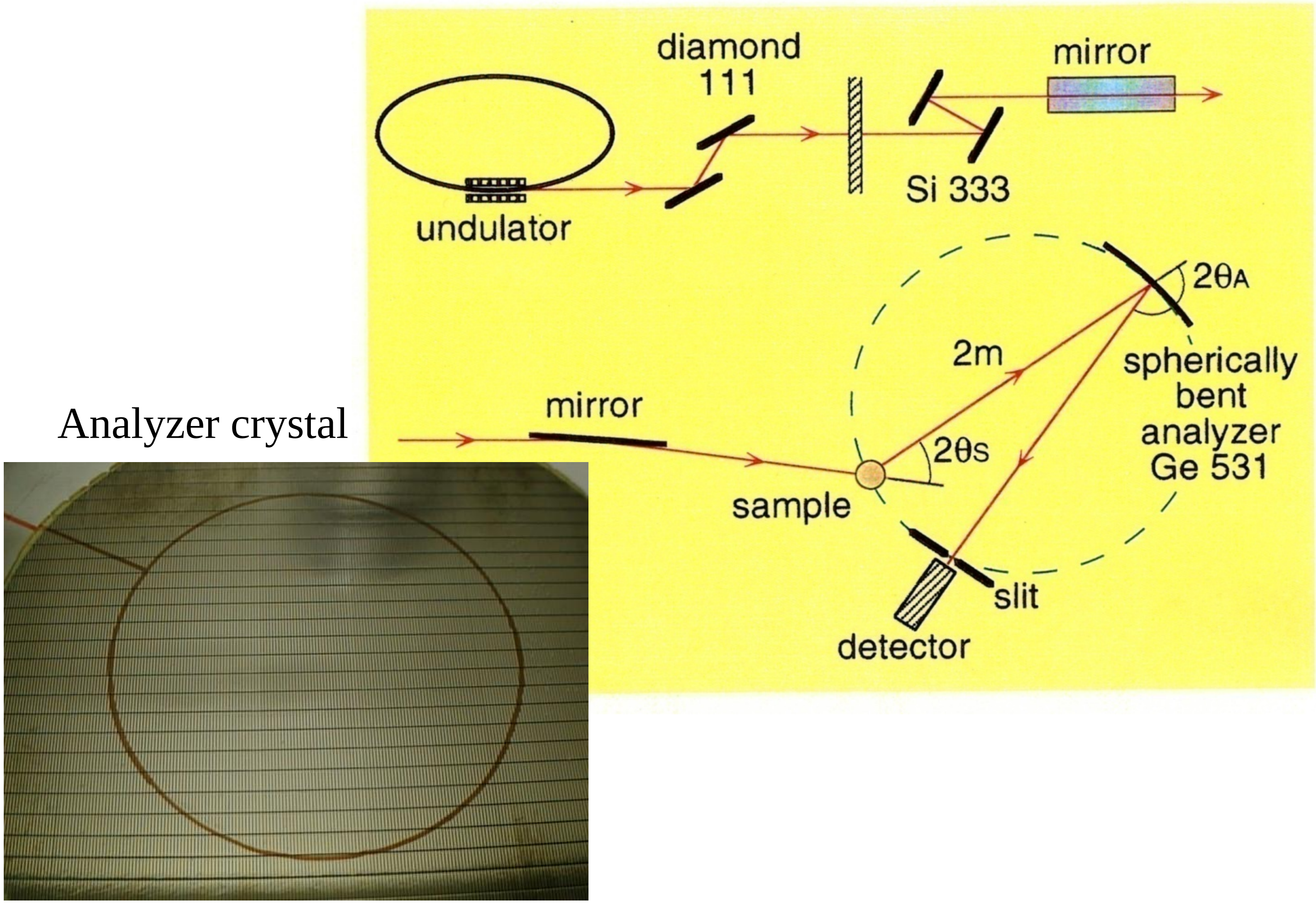
Candidates of the analyzer crystals an their plains for transition metals



	Si			Ge		
	(hkl)	Θ_B	ΔE_{meV}	(hkl)	θ_B	ΔE_{meV}
Cr 5989 eV	(511) (333)	82.27	55.5	(511) (333)	72.09	106.9
Mn 6539 eV	(440)	81.16	67.9	(531)	82.4	84.7
Fe 7112 eV	(531)	71.82	46.0	(620)	77.0	103.5
Co 7709 eV	(533)	76.3	37.0	(444)	80.3	70.7
Ni 8333 eV	(511) (711)	78.1	30.9	(642)	79.7	65.6
Cu 8979 eV	(553) (731)	77.62	26.5	(733)	87.3	35

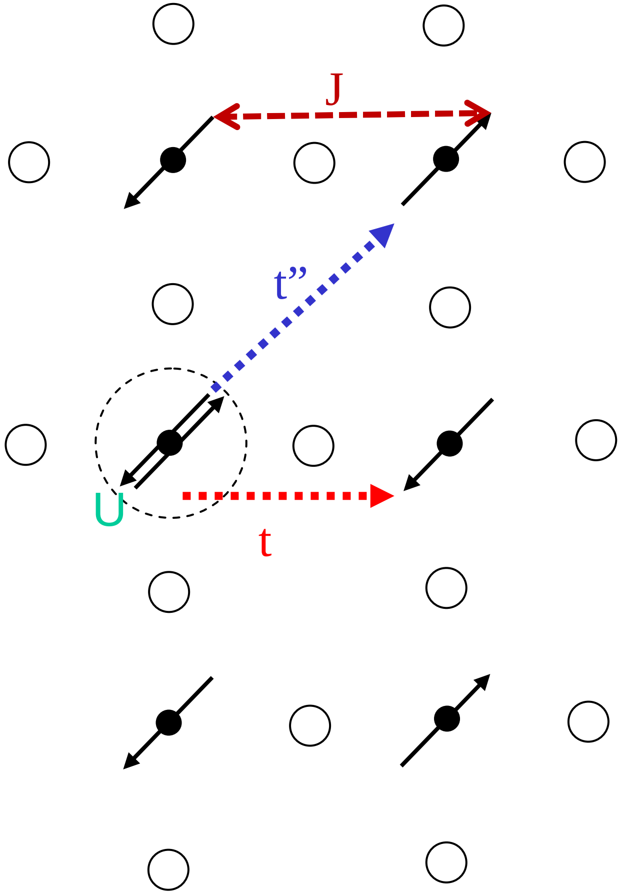
Intrinsic energy resolution

Bottle neck for getting high resolution


Analyzer crystal

Future direction

Charge dynamics in strongly
Correlated electron systems



-Excition across Mott/charge transfer gap

U: ΔE~0.5eV

-Excitation within bands across the Fermi level

t: ΔE~0.1eV

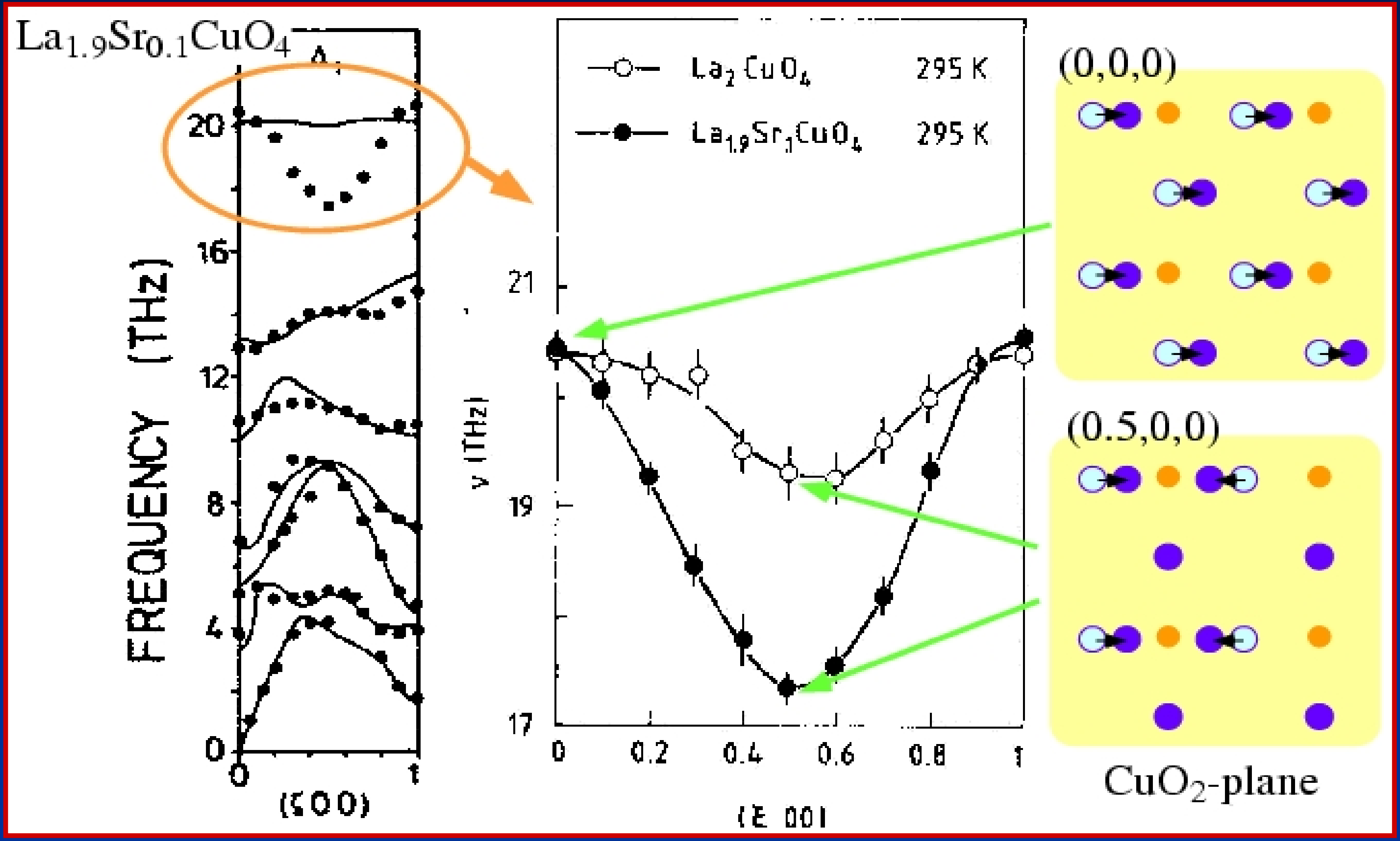
-Excitations related to the Spin degree of freedom

J: ΔE~0.05eV

What does phonon play a role on superconducting?

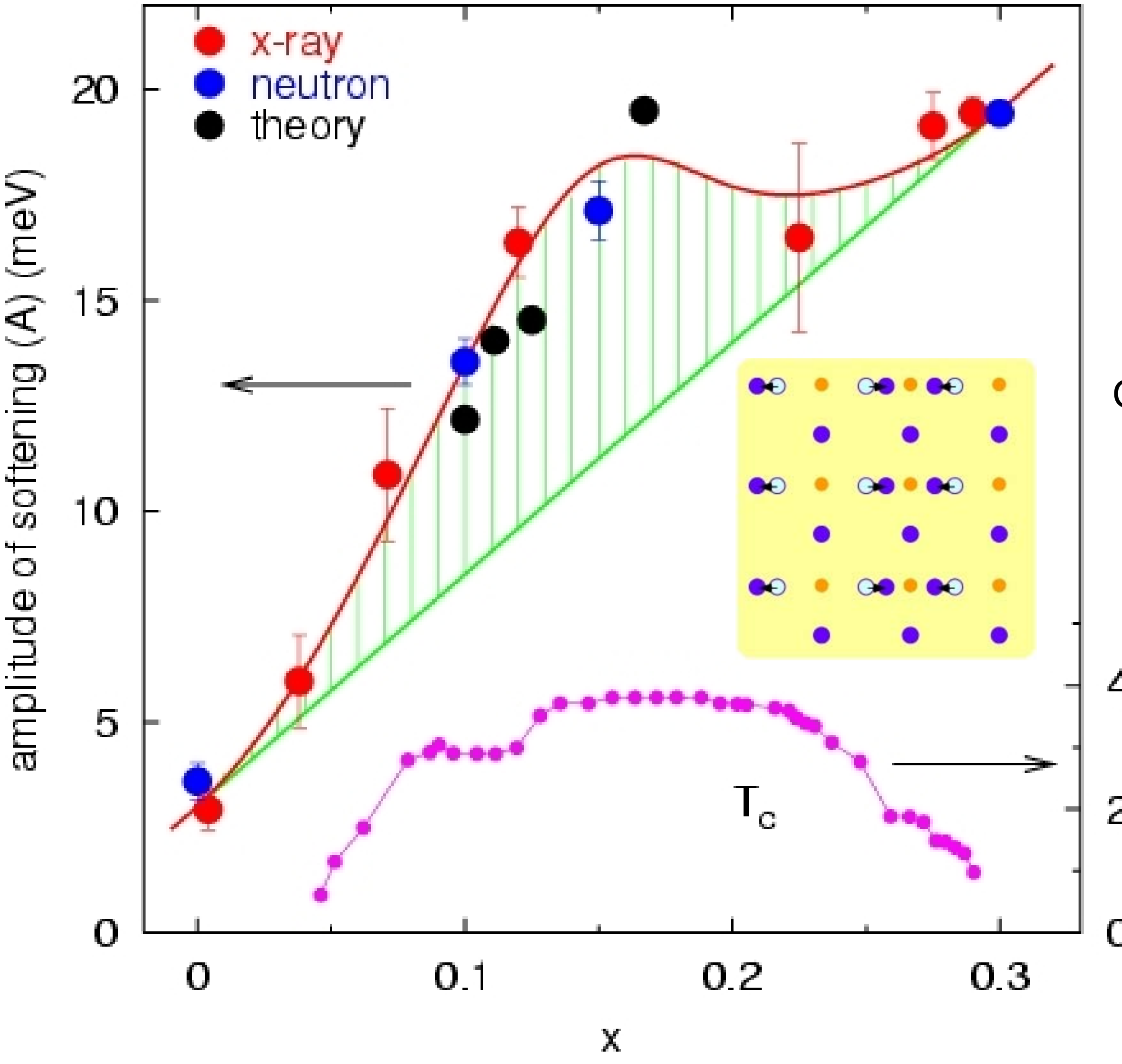
Anomalous dispersion of LO Cu-O bond stretching phonon modes observed by Neutron inelastic scattering

(Pintschovius et al., Physica B 174 (1991) 323)



Observation of the LO phonon anomaly

$La_{2-x}Sr_xCuO_4$



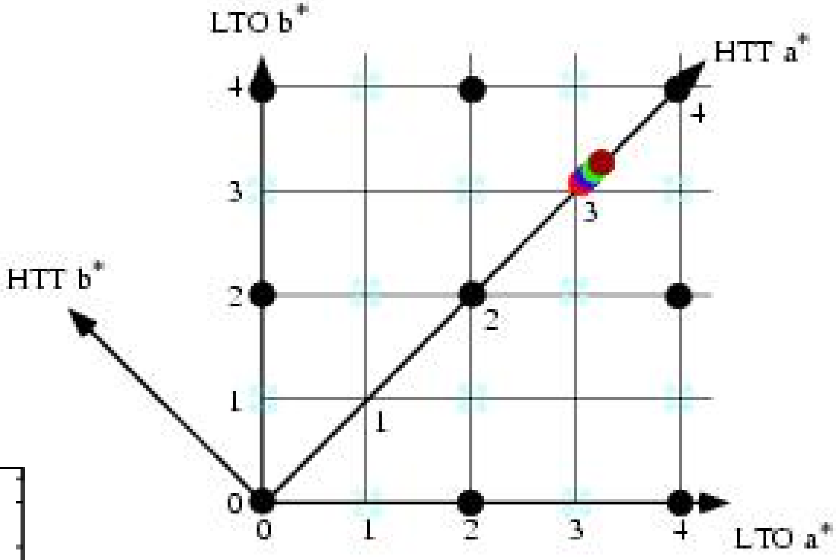
Grown by K. Ikeuchi

T. Fukuda, J. M,et al.,
P. R. B. 71 (‘05) 06050(R)

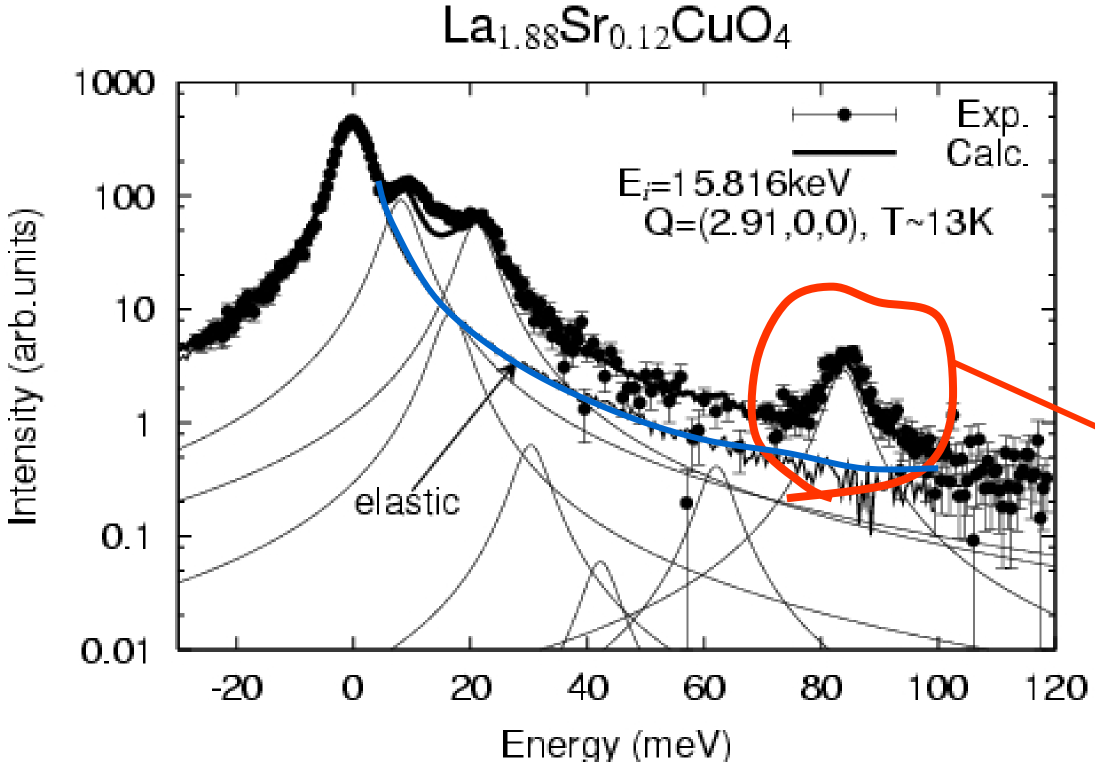
Phonon observed by inelastic x-ray scattering is not only phonon, but also elementary excitation of electrons

Raw data taken at BL-35XU, SPring-8

3 X 10^{10} photons/sec in ΔE=4 meV at 15.816 keV
(∼100 μm spot size)

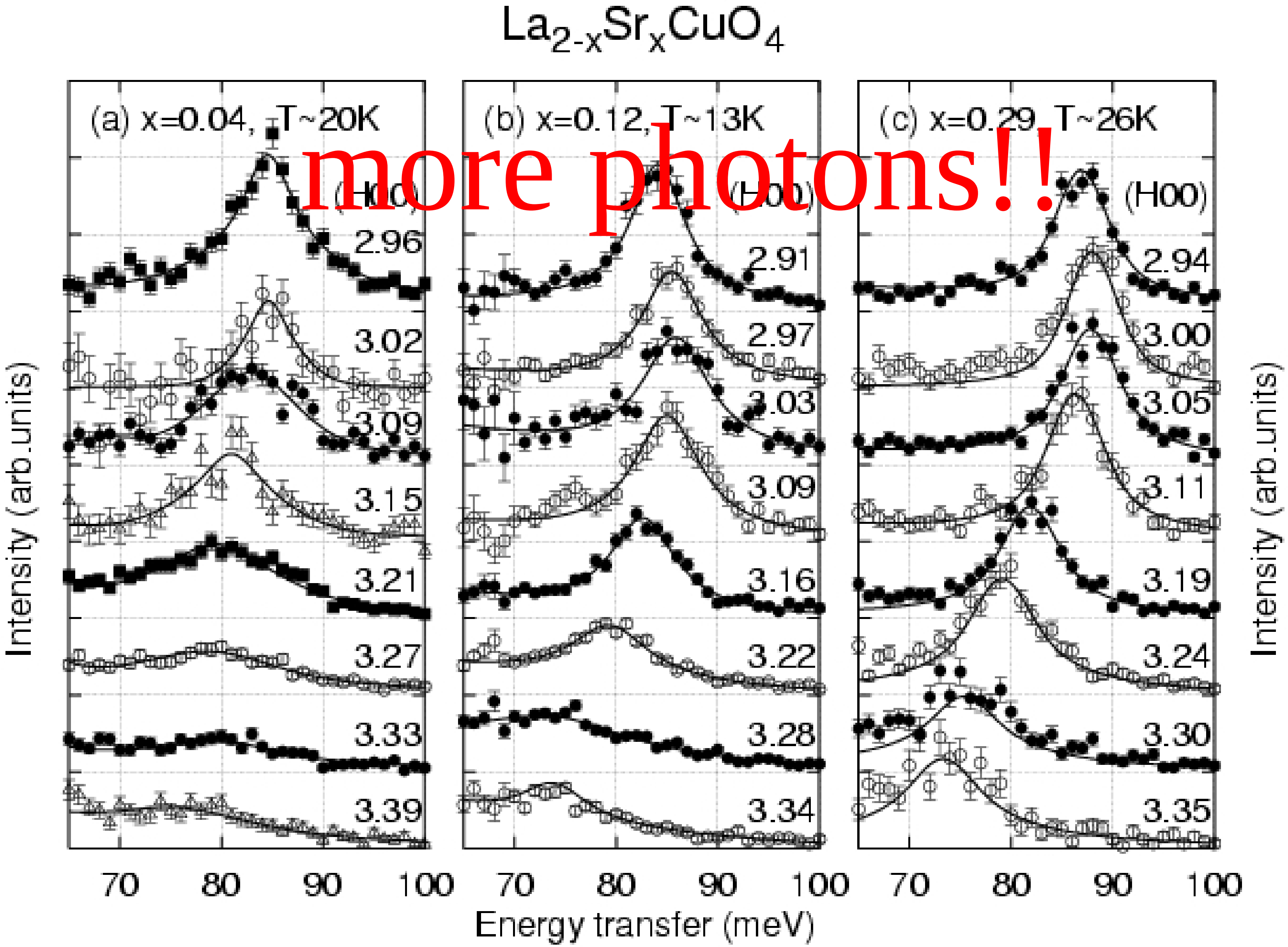


Const. Q energy scan


$La_{1.88}Sr_{0.12}CuO_4$

Cu-O LO bond-stretching phonon

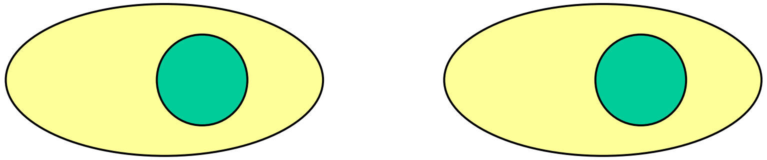
Comparison between X=0.04 and X=0.29



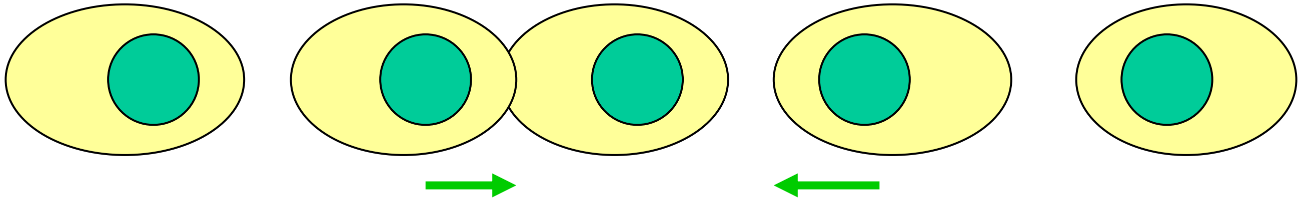
more photons!!

Dynamical Dielectric Function $\varepsilon\,(Q,\omega)$ consists of two parts:

1. Polarization properties of electrons in a field of rigid lattice.



2. Contribution of oscillating ions. (phonon)



O. V. Dolgov et. al., Rev. Mod. Phys. 53 ('81) 81

Dynamical structure factor of IXS in the phonon energy region

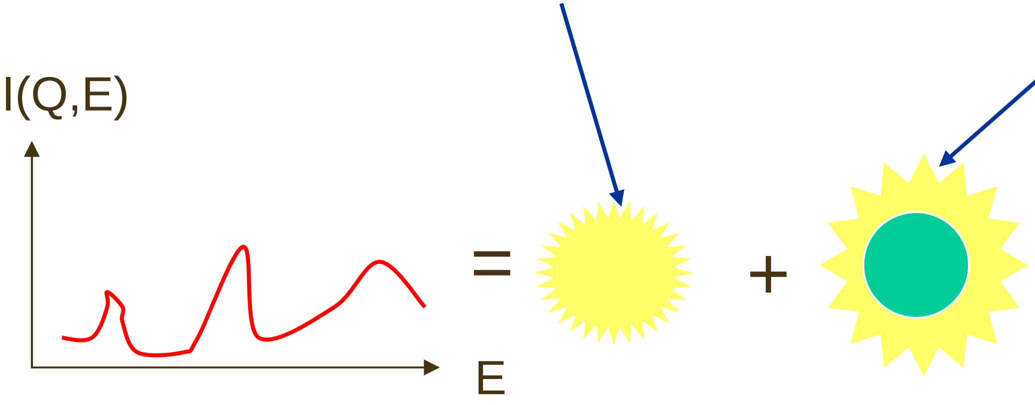
$$\varepsilon(Q,\omega)=\varepsilon_{el}(Q,\omega)+\varepsilon_{ion}(Q,\omega)-1$$

$$\downarrow$$

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

$$\downarrow$$

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



PHYSICAL REVIEW B 67, 174506 (2003)

Vibronic mechanism of high-T_c superconductivity

M. Tachiki **et. al.,**

$$\boxed{\epsilon(\mathbf{q},\omega)=\epsilon_{el}(\mathbf{q},\omega)+\epsilon_{ion}(\mathbf{q},\omega)-1.}$$

$$\epsilon_{ion}(\mathbf{q},\omega)=\frac{\omega^2-\omega_{LO}^2}{\omega^2-\omega_{TO}^2},$$

ω_{LO}: LO ph. in insulating state
ω_{TO}:TO ph. in insulating state

$$I(\mathbf{q},\omega)=-\frac{1}{\pi}\mathrm{Im}\left[\frac{1}{\epsilon(\mathbf{q},\omega)}\right]$$

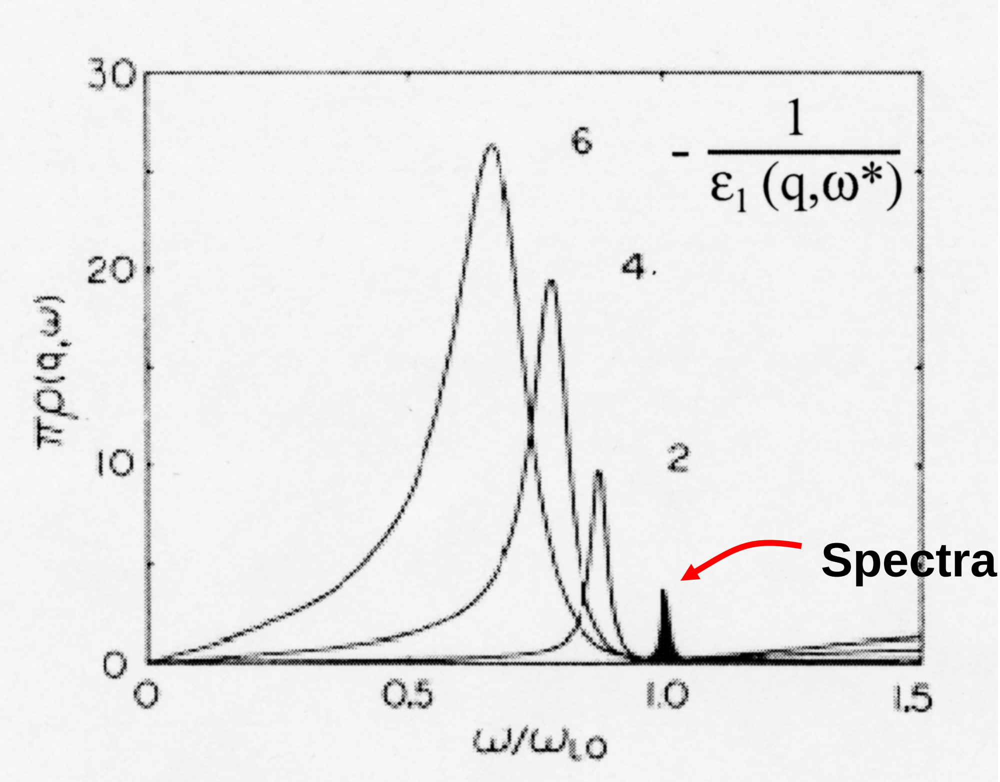
ω^*_{LO}:LO ph. softened by mixing with charge oscillations

$$=-\frac{1}{\pi}\mathrm{Im}\left[\frac{1}{\epsilon_{el}(\mathbf{q},\omega)}\right]+\frac{\omega_{LO}^2(\mathbf{q})-\omega_{TO}^2}{\epsilon_{el}(\mathbf{q},\omega^*_{LO})^2}\,\delta(\omega^2-\omega^{*2}_{LO}),$$

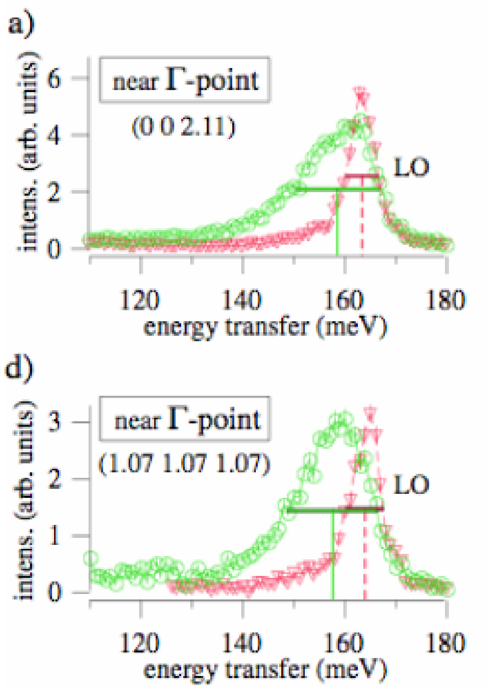
electric part

(electric + phonon) part

Spectral intensities of the charge-transfer oscillations associated with the LO phonon as functions of the normalized ω for several values of $1/\varepsilon_1(q,\omega^*)$



Spectral intensity of the bare LO ph.



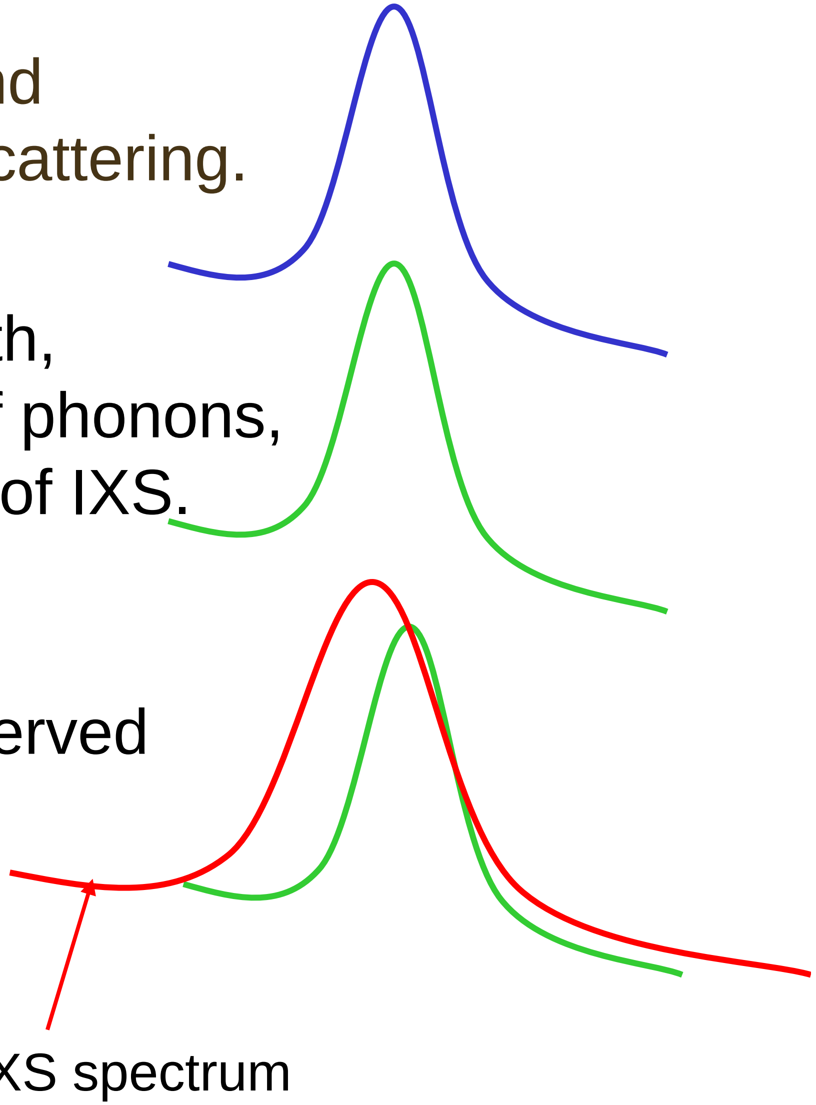
M. Tachiki and S. Takahashi, P. R. B. 38 ('88) 218.

How to get the information on electric dynamical function

1. Measure the phonon spectrum and dispersion by Neutron Inelastic Scattering.

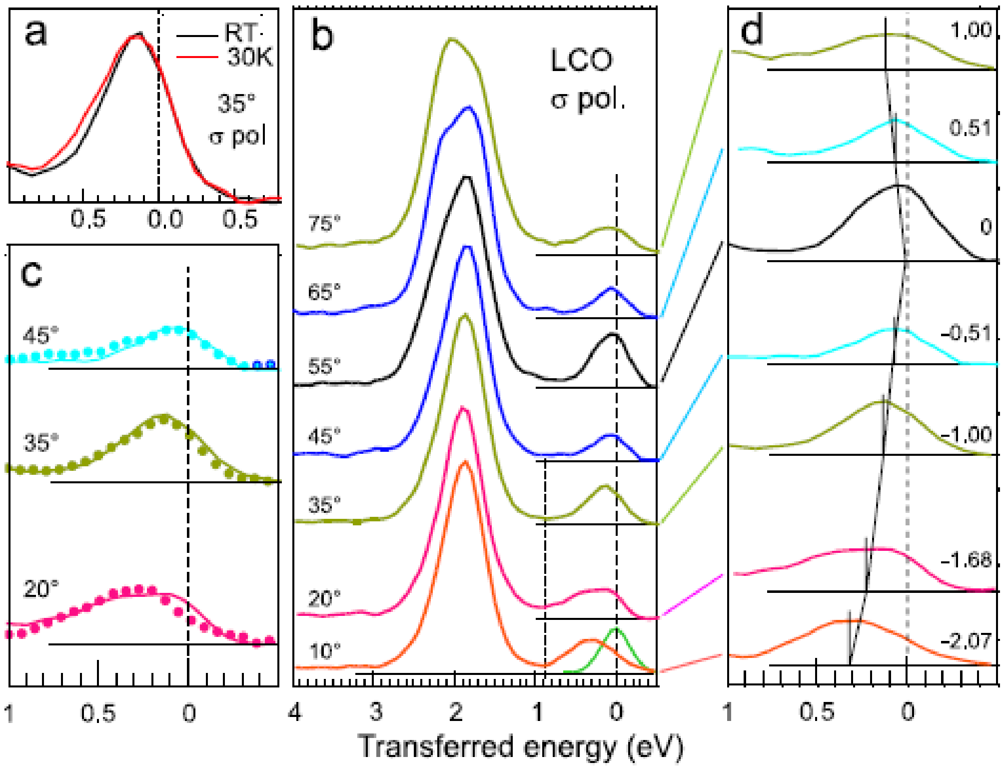
2. Derive the information on the width, eigenvector and structure factor of phonons, and calculate the phonon intensity of IXS. (C-IXS)

3. Subtract the C-IXS from the observed IXS spectrum.

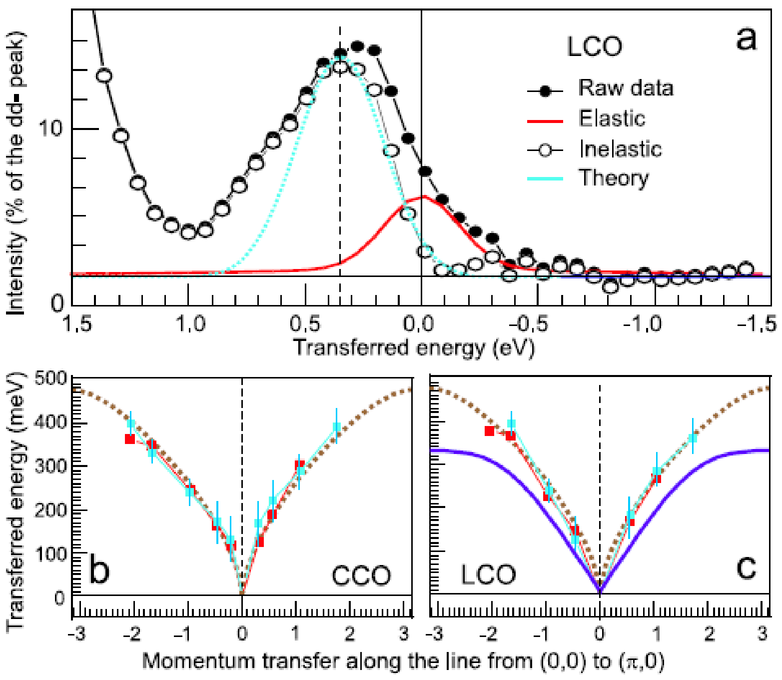


Magnetic excitations observed by RIXS

La_2CuO_4

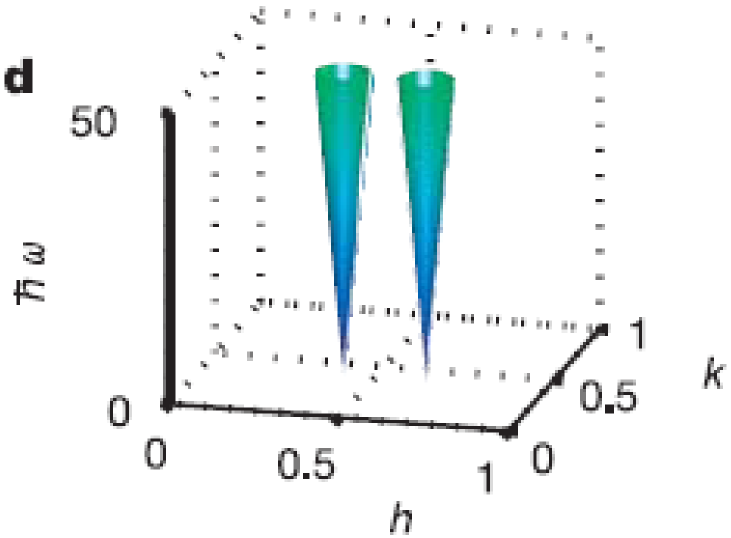


Magnon Dispersion



L. Braicovichi et al., PRL 102 ('09) 167401

Neutron experiment



J. M. Tranquada, et al., Nature 429(‘04)534

Theory

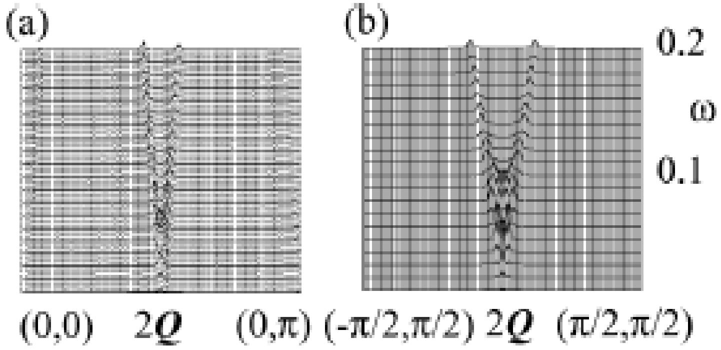


FIG. 1. Dispersion curves of the charge collective mode around 2**Q**. We plot Im$\chi_{nn}(\mathbf{q},\omega)$ for two paths along 2**Q** (a) and perpendicular to 2**Q** (b).

E. Kaneshita, et al., P. R. L. 88(’02) 115501