Inelastic X-ray Scattering: Past, Present, and Future

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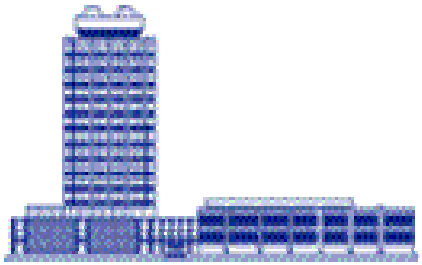
University of Toronto



Outline

- Past
  - Inelastic X-ray Scattering in 20th century
  - Many flavors of IXS
  - Some examples
- Present – mostly cuprates
  - Charge transfer excitation (> 2 eV)
  - Orbital selective d-d excitation (< 2eV)
  - Multi-magnon excitation (< 1eV)
- Future
  - Conclusions and Outlook

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THE

PHYSICAL REVIEW

A QUANTUM THEORY OF THE SCATTERING OF X-RAYS BY LIGHT ELEMENTS

By Arthur H. Compton

Abstract

A quantum theory of the scattering of X-rays and γ-rays by light elements.—The hypothesis is suggested that when an X-ray quantum is scattered it spends all of its energy and momentum upon some particular electron. This electron in turn scatters the ray in some definite direction. The change in momentum of the X-ray quantum due to the change in its direction of propagation results in a recoil of the scattering electron. The energy in the scattered quantum is thus less than the energy in the primary quantum by the kinetic energy of recoil of the scattering electron. The corresponding *increase in the wave-length of the scattered beam* is $\lambda_\theta - \lambda_0 = (2h/mc)\sin^2\frac{1}{2}\theta = 0.0484\sin^2\frac{1}{2}\theta$, where h is the Planck constant, m is the mass of the scattering electron, c is the velocity of light, and θ is the angle between the incident and the scattered ray. Hence the increase is independent of the wave-length. *The distribution of the scattered radiation* is found, by an indirect and not quite rigid method, to be concentrated in the forward direction according to a definite law (Eq. 27).

2008

the secondary. The zero point for the spectrum of both the primary and secondary X-rays was determined by finding the position of the first order lines on both sides of the zero point.

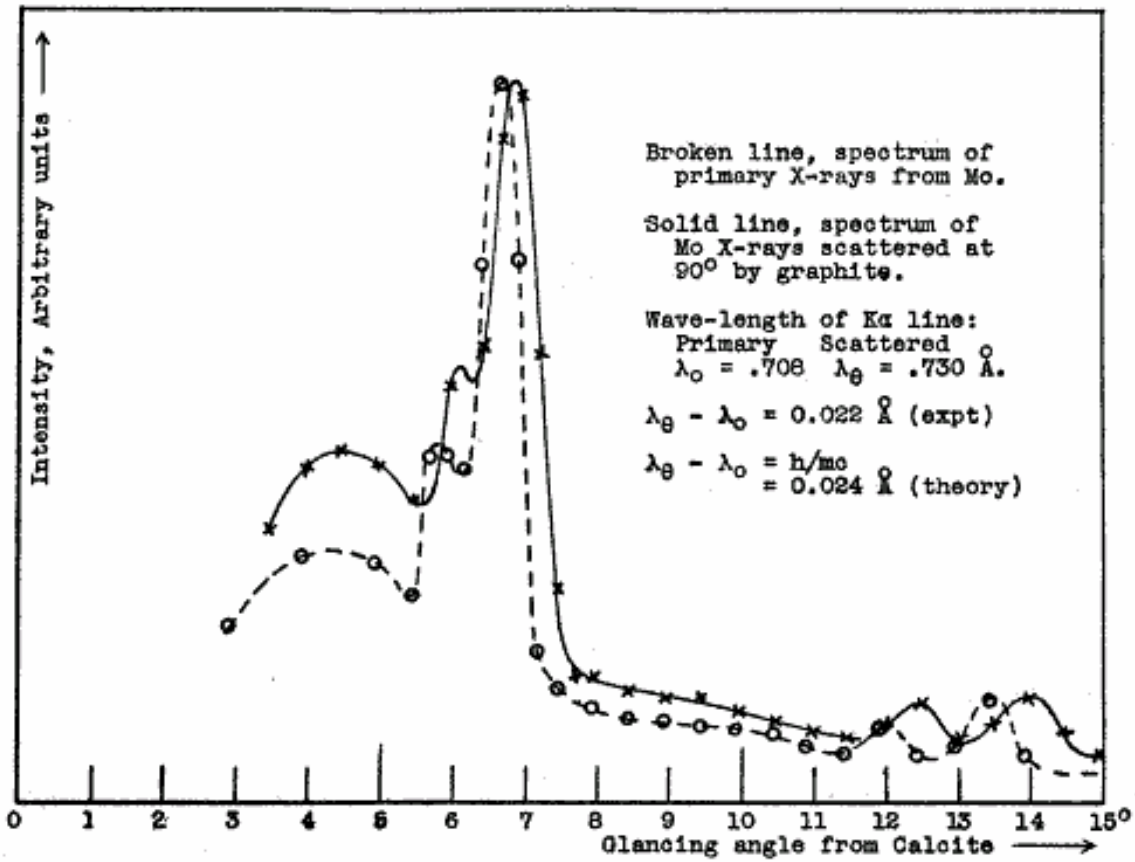
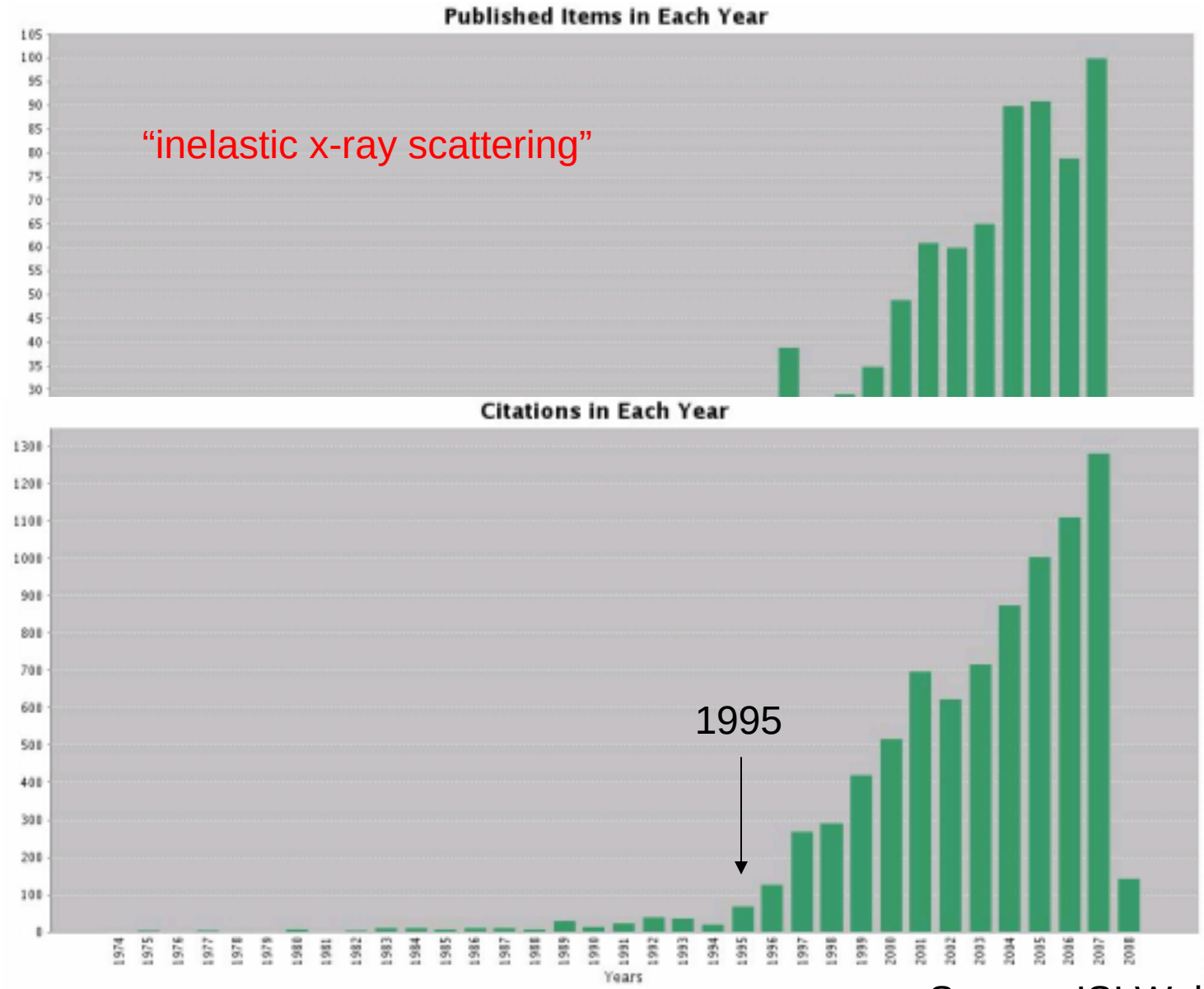


Fig. 4. Spectrum of molybdenum X-rays scattered by graphite, compared with the spectrum of the primary X-rays, showing an increase in wave-length on scattering.

It will be seen that the wave-length of the scattered rays is unquestionably greater than that of the primary rays which excite them

Historical perspective



Source: ISI Web of Science

1995

VOLUME 75, NUMBER 5 PHYSICAL REVIEW LETTERS 31 JULY 1995

Collective Dynamics in Water by High Energy Resolution Inelastic X-Ray Scattering

F. Sette,[1] G. Ruocco,[2] M. Krisch,[1] U. Bergmann,[1] C. Masciovecchio,[1,2] V. Mazzacurati,[2] G. Signorelli,[2] and R. Verbeni[1]

[1]*European Synchrotron Radiation Facility, B.P. 220 F-38043 Grenoble Cedex, France*
[2]*Universitá di L'Aquila and Istituto Nazionale di Fisica della Materia, I-67100, L'Aquila, Italy*
(Received 12 April 1995)

A propagating excitation with a velocity of sound of 3200 ± 100 m/s is measured in H_2O at 294 K between 4 and 14 nm^{-1}, using inelastic x-ray scattering with 5 meV energy resolution. The existence of *fast sound* is therefore demonstrated in an energy-momentum region much wider than that of previous neutron measurements on D_2O. The equivalence of the *fast sound* velocity in H_2O and in D_2O rules out models where this mode propagates only on the hydrogen network. These results show the ability of inelastic x-ray scattering to study the collective dynamics of liquids.

PHYSICAL REVIEW B VOLUME 54, NUMBER 23 15 DECEMBER 1996-I

X-ray resonant Raman scattering in NiO: Resonant enhancement of the charge-transfer excitations

C.-C. Kao, W. A. L. Caliebe, and J. B. Hastings
National Synchrotron Light Source, Brookhaven National Laboratory, Upton, New York 11973

J.-M. Gillet
Department of Material Science, Universite de Marne la Vallee, 93166 Noisy le Grand Cedex, France
(Received 13 September 1996)

An energy loss feature with energy transfer close to the anion-to-cation charge-transfer energy of NiO is observed in the inelastic-x-ray-scattering spectrum as the incident x-ray energy is tuned through the Ni *K* absorption edge of NiO. The inelastic-scattering cross section shows large resonant enhancement and strong incident energy dependence. These observations are interpreted using a configuration-interaction cluster model of NiO. [S0163-1829(96)04548-1]

Many flavors of IXS

$$H_{e-\gamma} \propto A^2 + A \bullet p$$

Non-resonant IXS (NIXS)

High-resolution IXS
- 1 meV resolution
- phonons + vibrational modes
- neutron scattering

Medium-resolution NIXS
- 200-1000 meV resolution
- Collective electronic excitations
- EELS

Resonant IXS (RIXS)

Core-electrons RIXS
- Typically 1 eV resolutions
- Started with soft x-ray
- XAS, XPS, XES, etc.

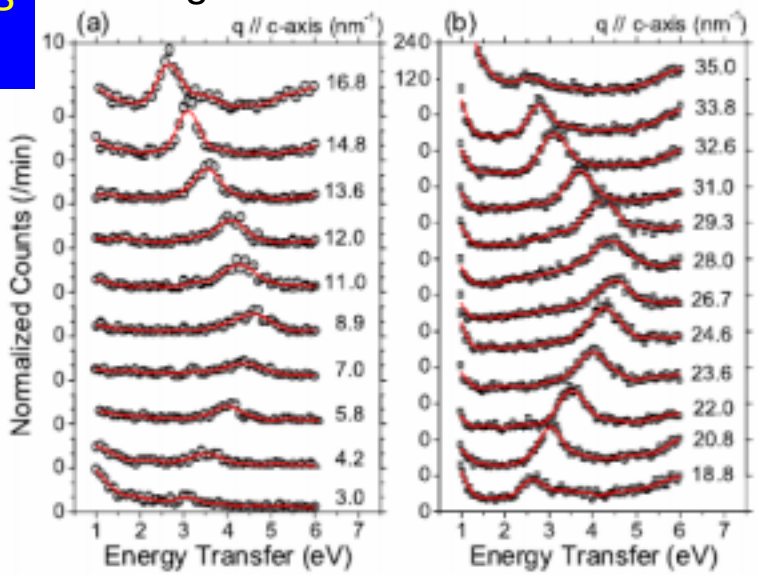
Core spectroscopy IXS
- Typically 1 eV resolution
- X-ray Raman
- e.g. low Z material in high pressu

Valence electrons RIXS
- 100-400 meV
- Charge gap, d-d, etc.
- IR, Raman, EELS

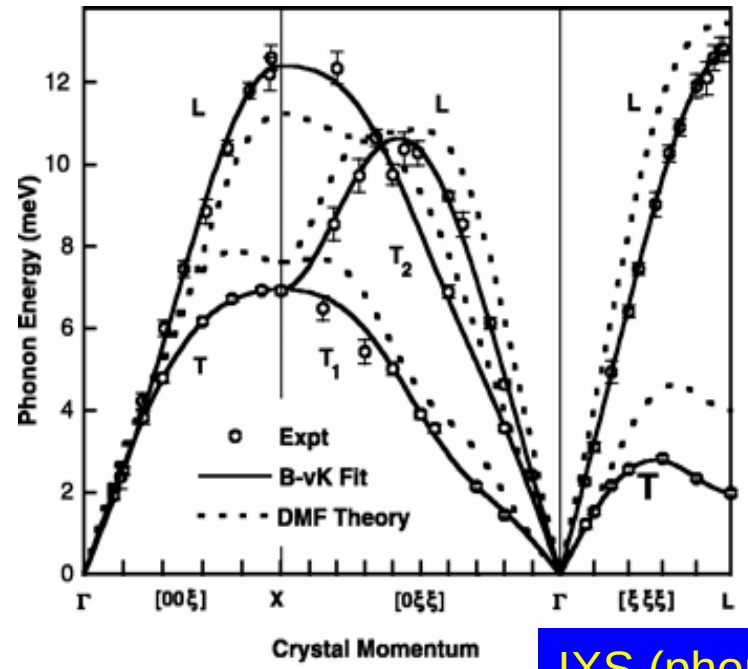
Examples

Medium Res NIXS

MgB2 Cai et al. PRL 2006

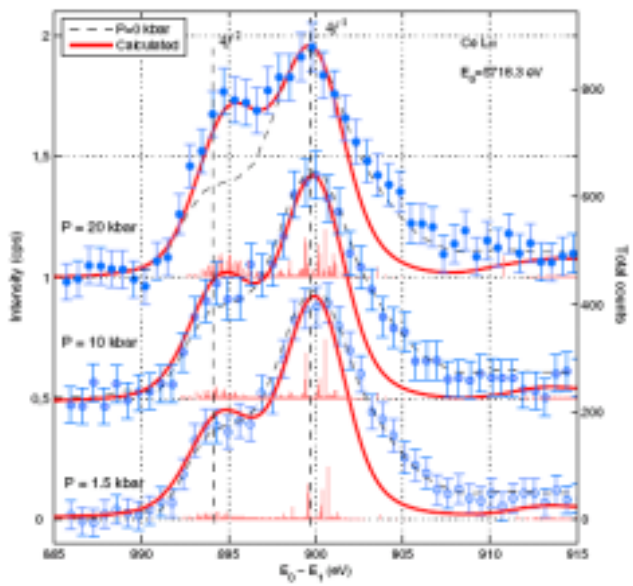


δ-Pu-Ga, Wong et al. Science 2003

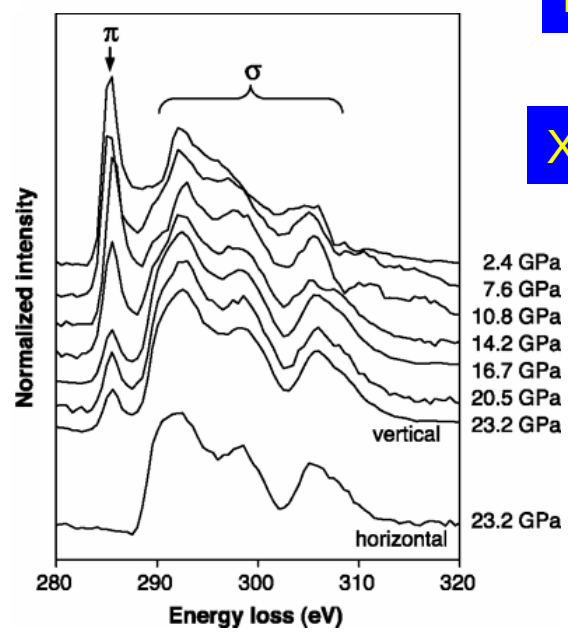


IXS (phonon)

Core RIXS



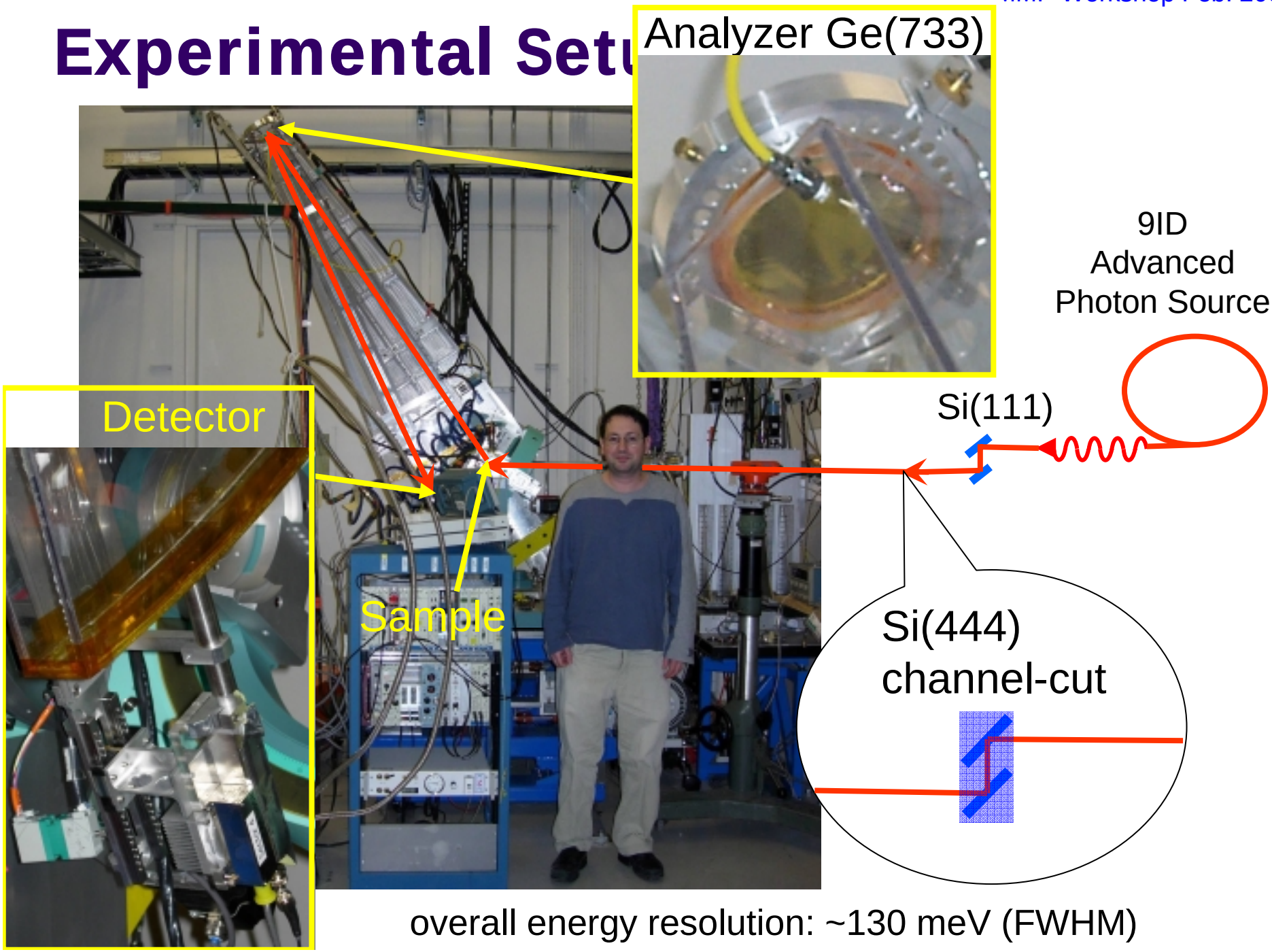
bulk Ce
Rueff et al.
PRL 2006



X-ray Raman

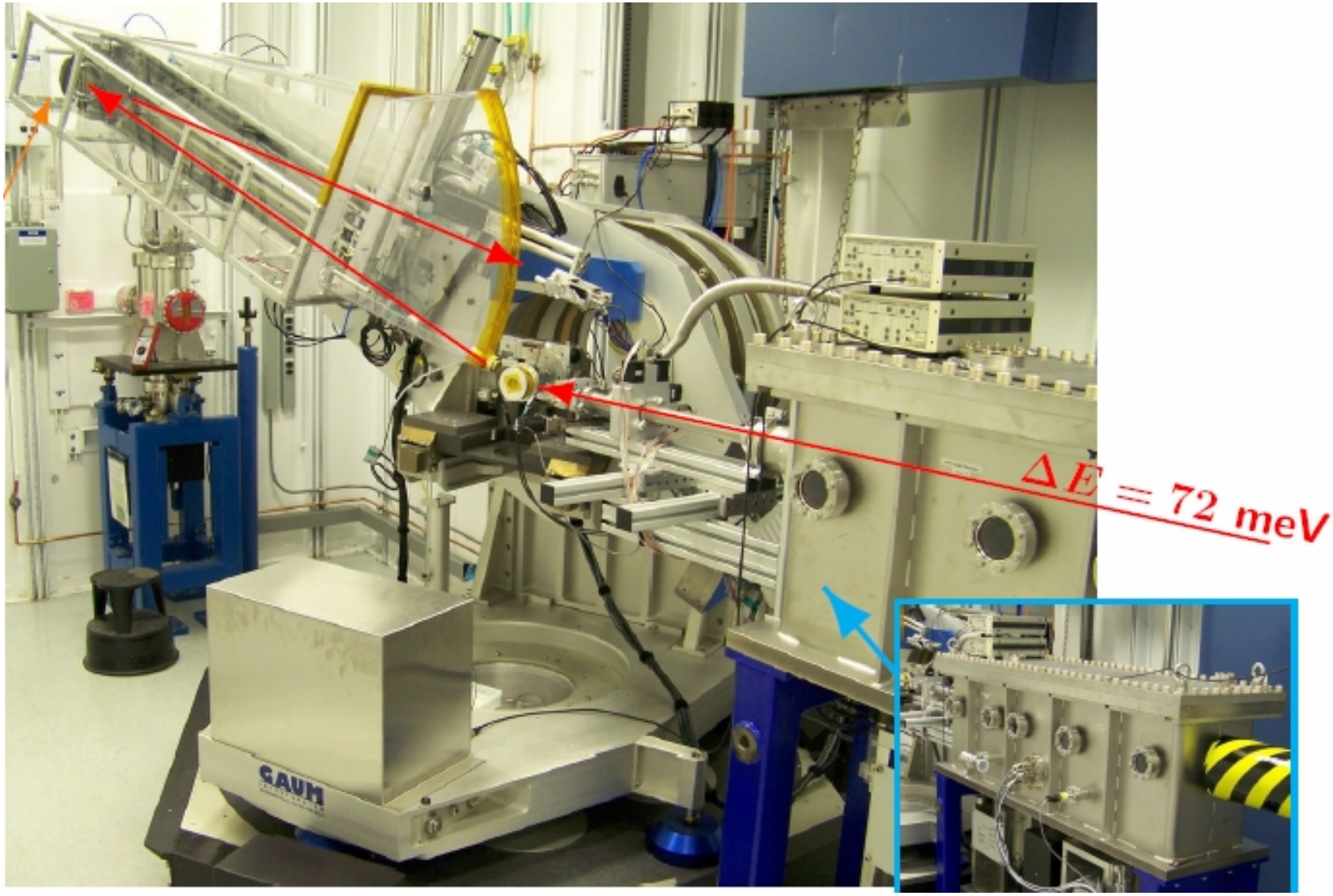
Graphite
Mao et al.
Science 2003

Experimental Setu

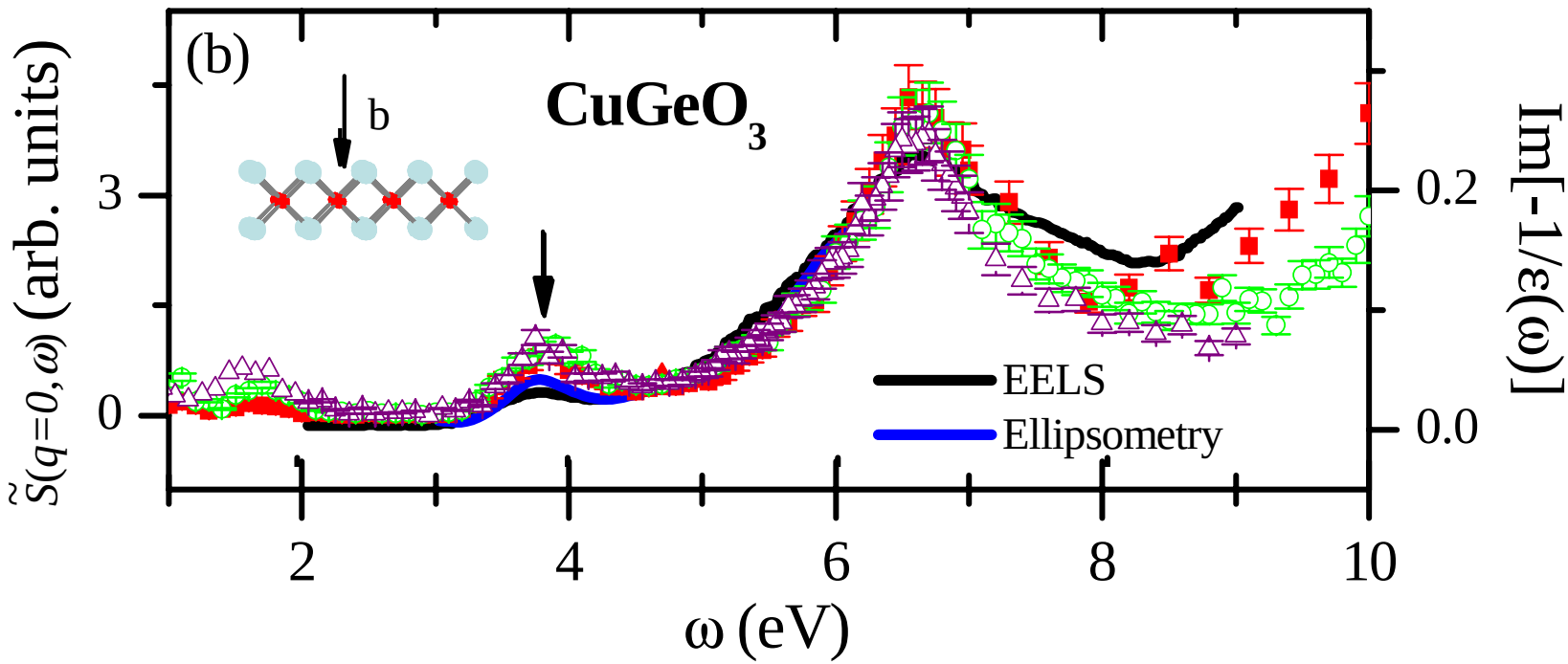


overall energy resolution: ~130 meV (FWHM)

MERIX spectrometer (APS 30ID)



E_i-independent spectrum



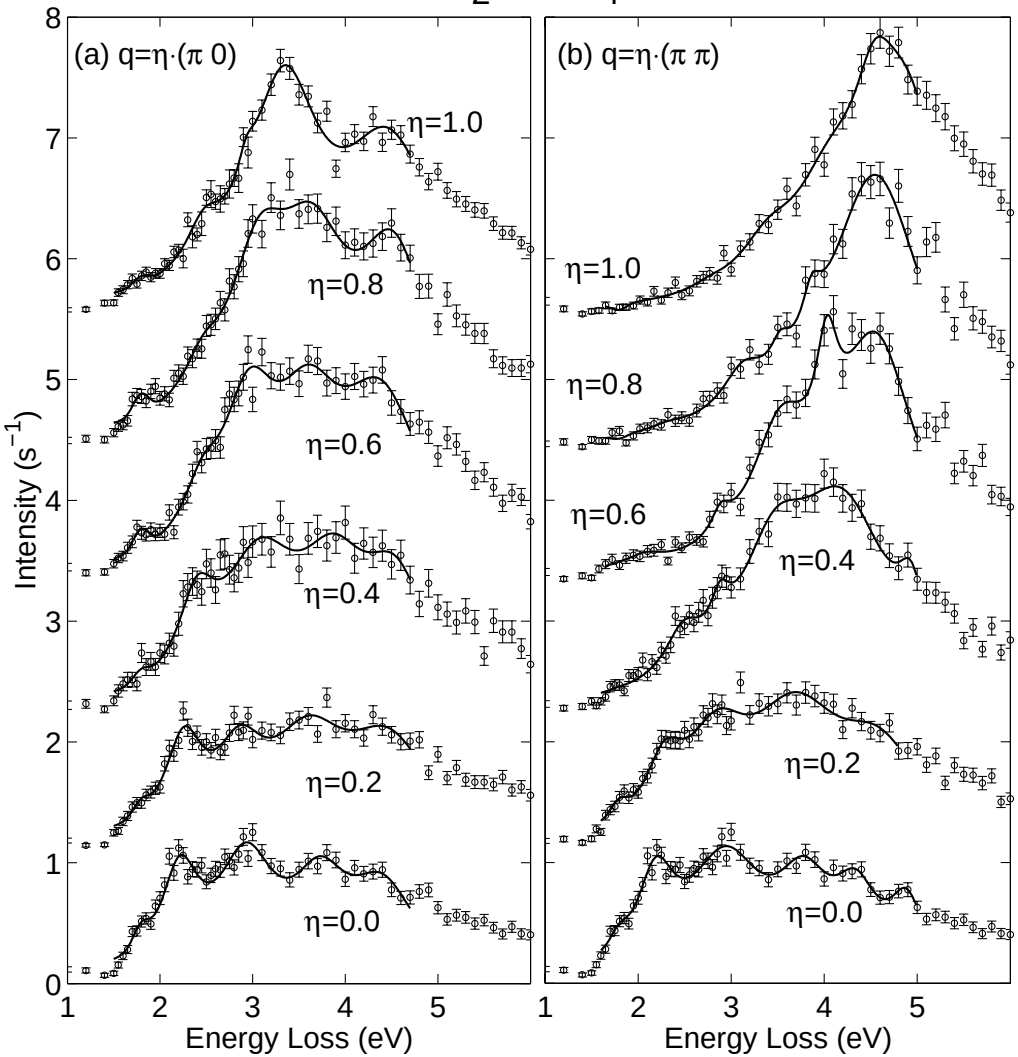
- Dielectric loss function from
  - ellipsometry
  - EELS

$$S(q=0,\omega) \propto \mathrm{Im}\left[-\frac{1}{\varepsilon(\omega)}\right]$$

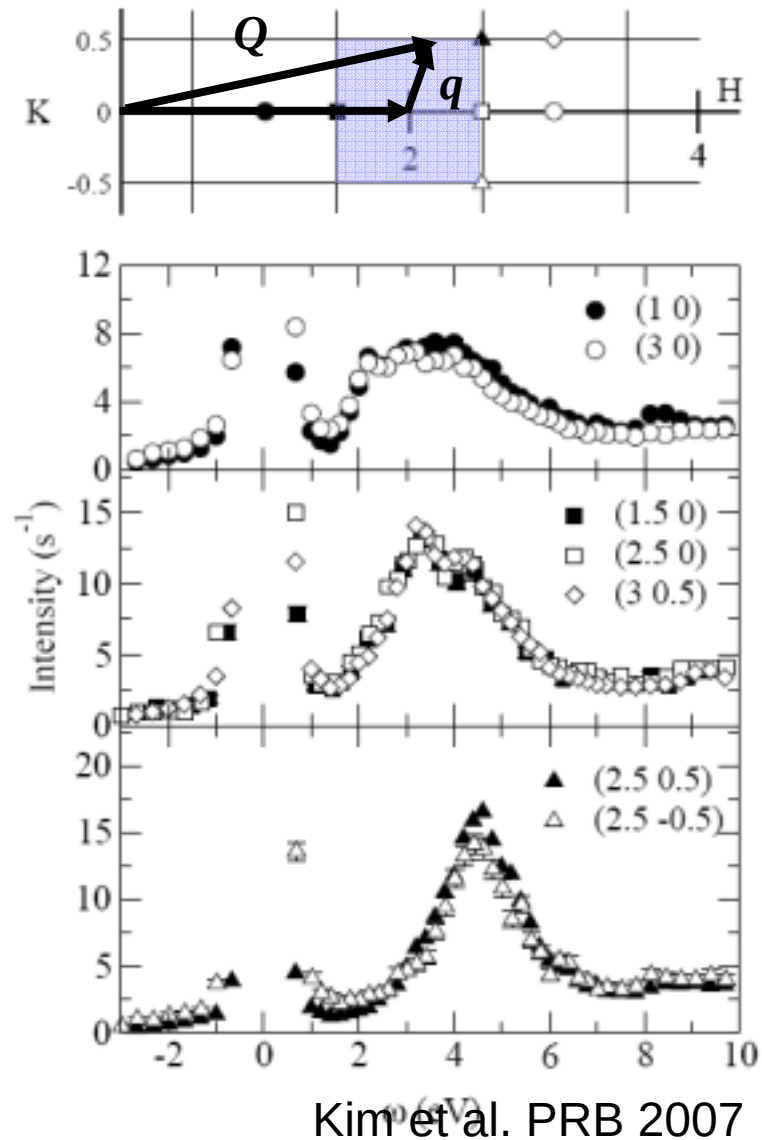
Momentum dependence

$$\vec{Q} = \vec{q} + \vec{G}$$

La_2CuO_4



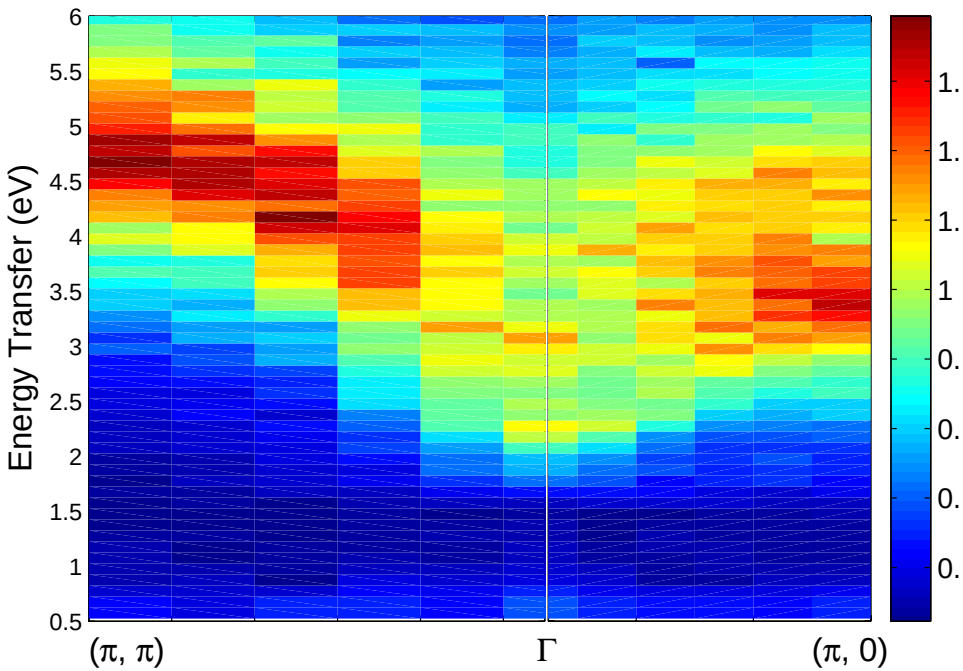
Ellis et al. PRB in press



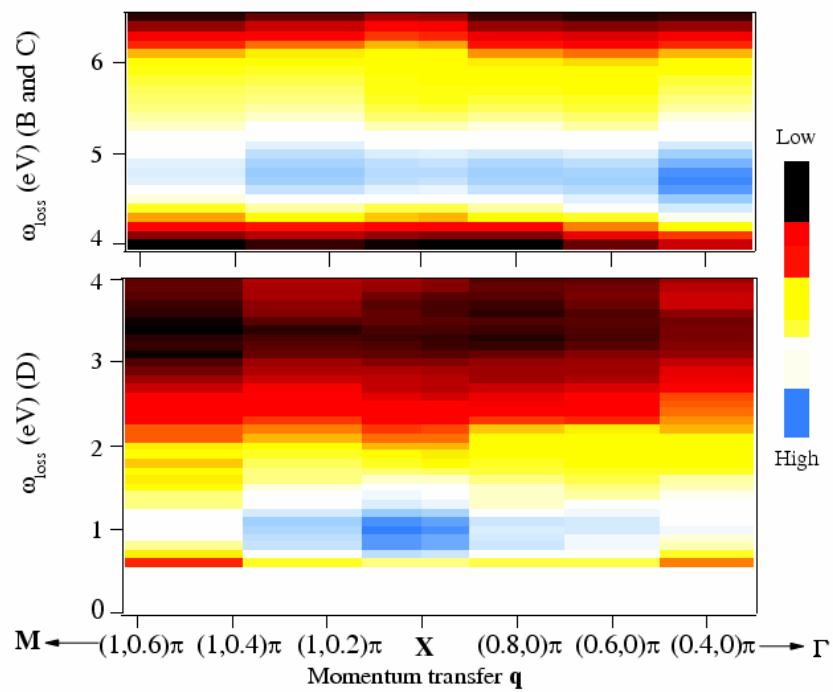
Kim et al. PRB 2007

Is this unusual ?

La_2CuO_4



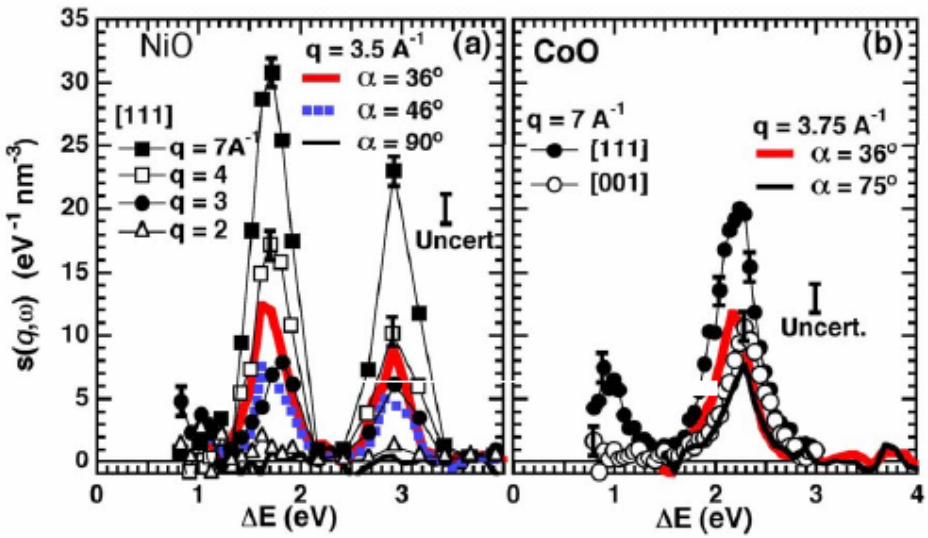
$La_{2-x}Sr_xNiO_4$ (x=0.15)



dd excitation using hard X-ray

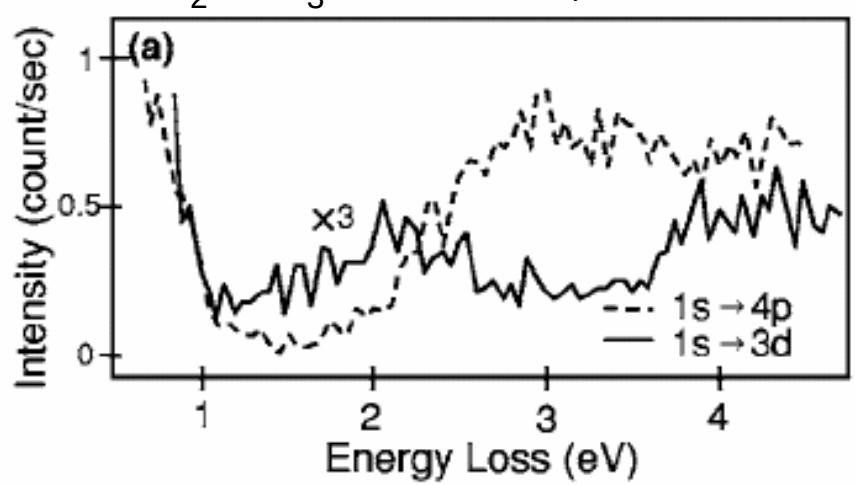
- Going to very large Q

Larson et al., PRL 2007

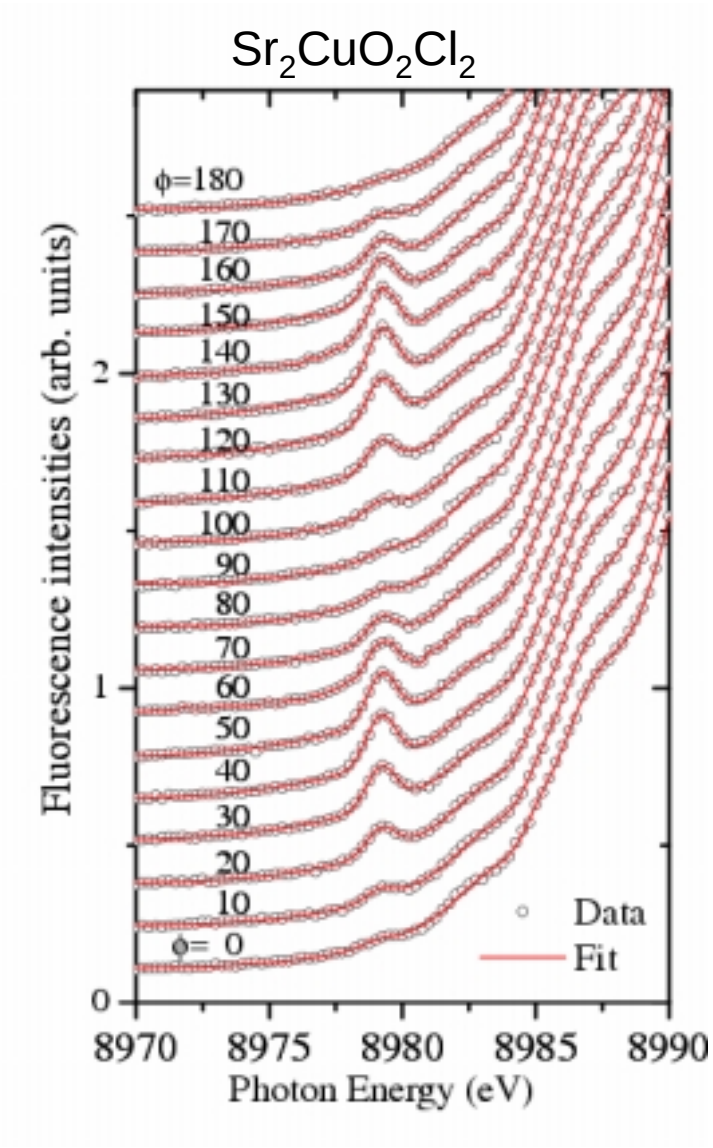


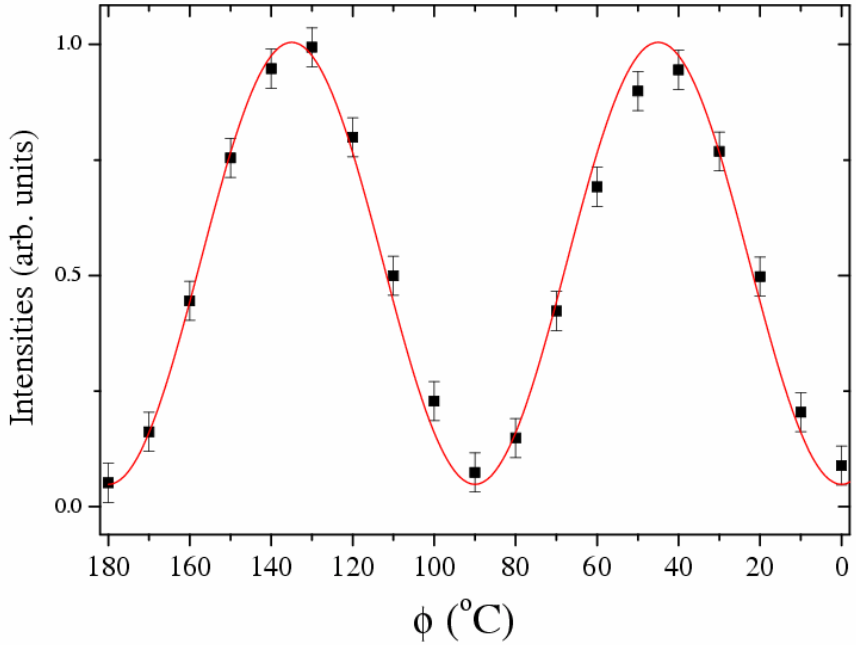
- Using quadrupole transition as an intermediate state

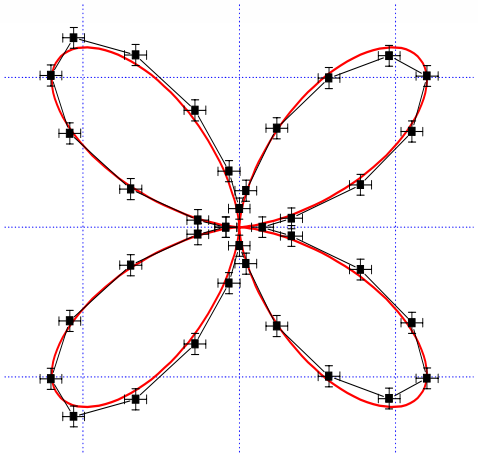
Sr_2CuO_3 - Seo et al., PRB 2006



K-edge quadrupole transition

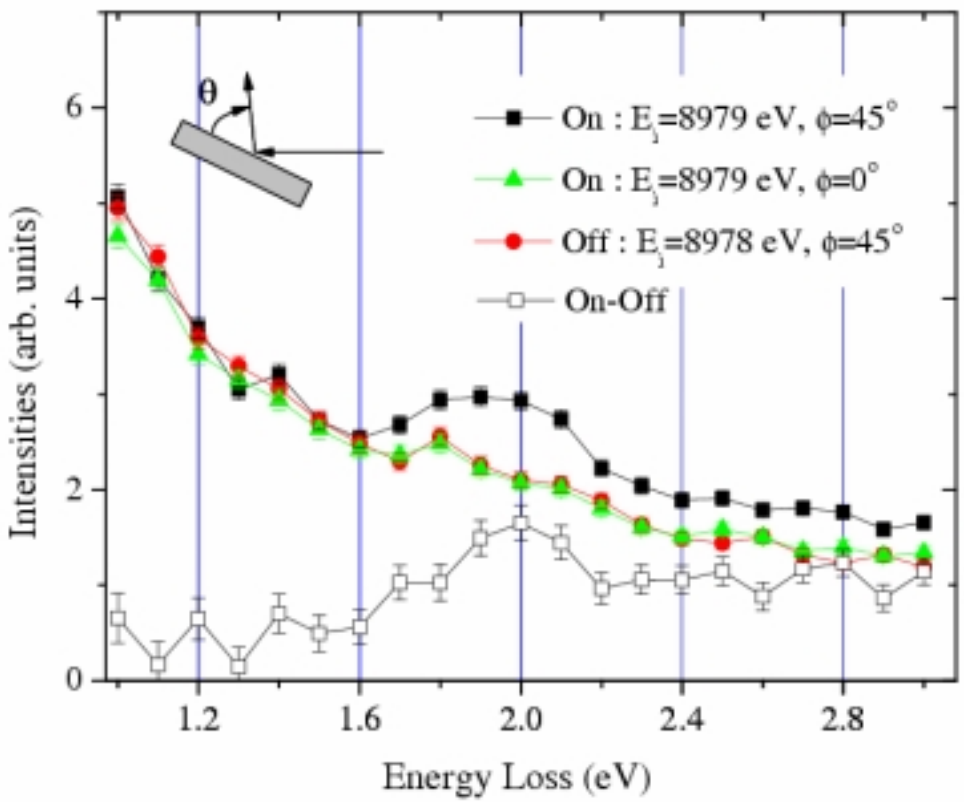


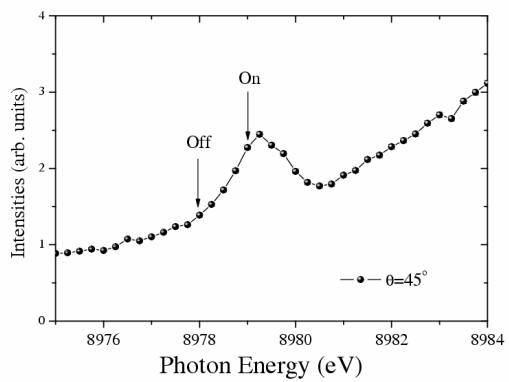




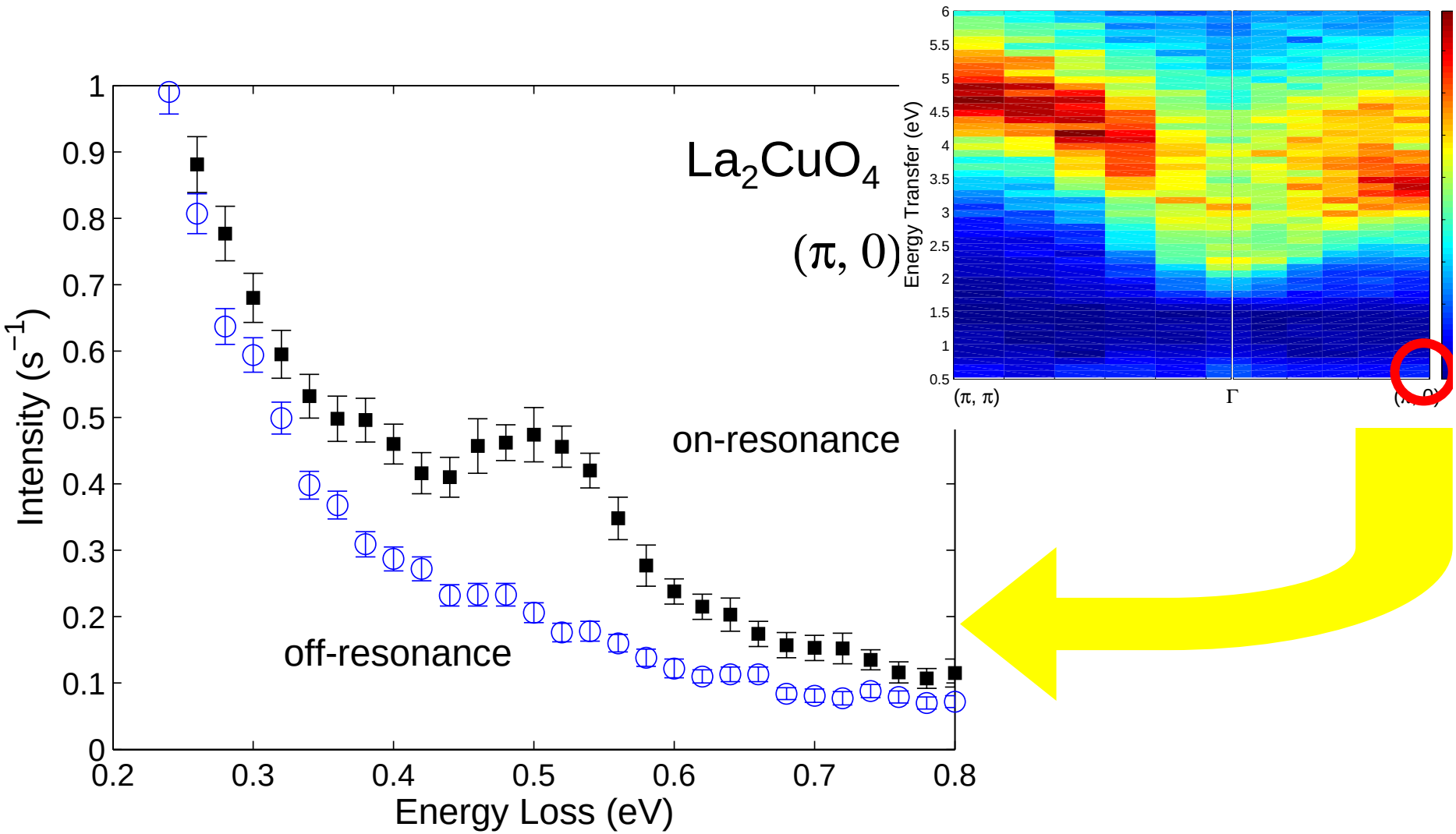
d-d excitation in $Sr_2CuO_2Cl_2$

- pre-edge (quadrupole) intermediate state



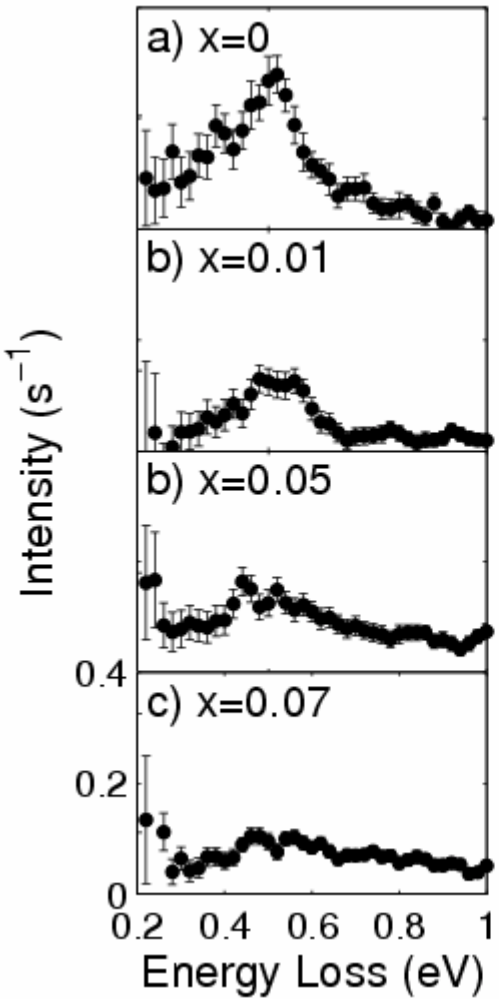


Magnetic Excitation?

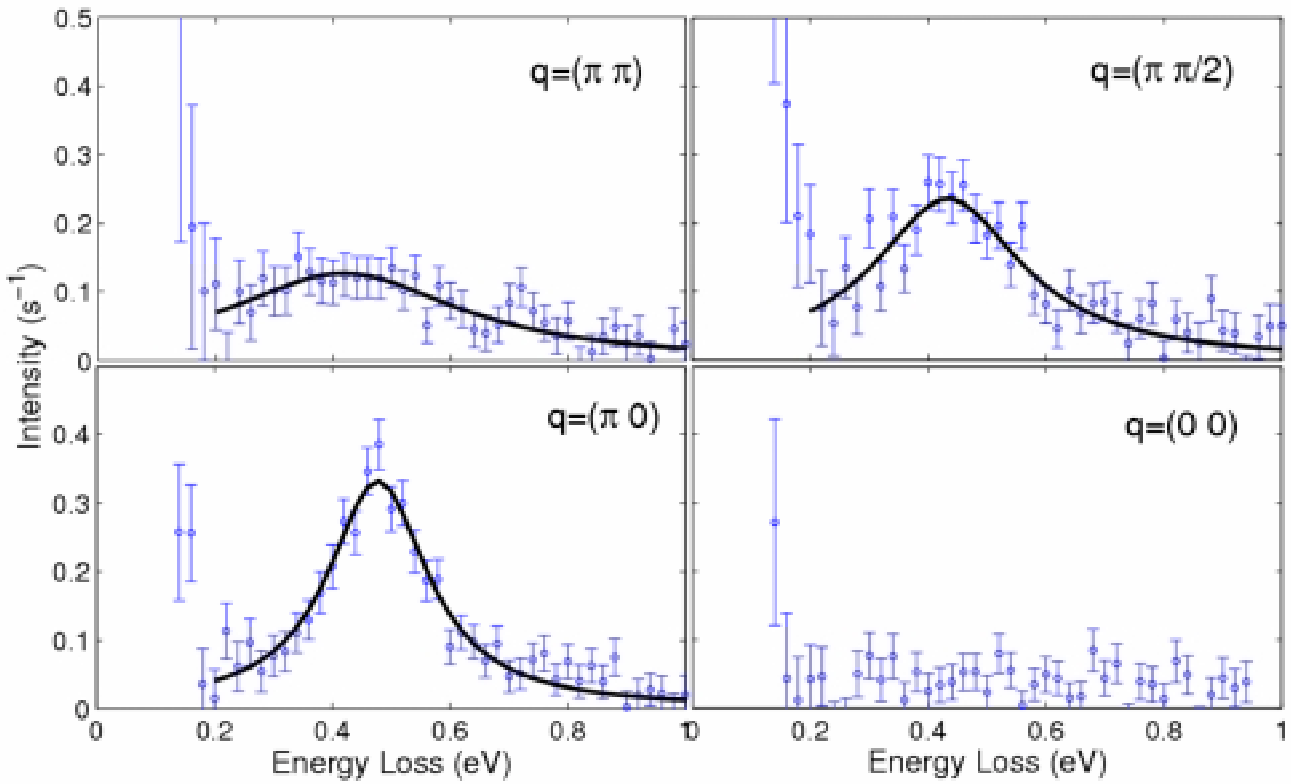


Hill et al. PRL in press

Doping

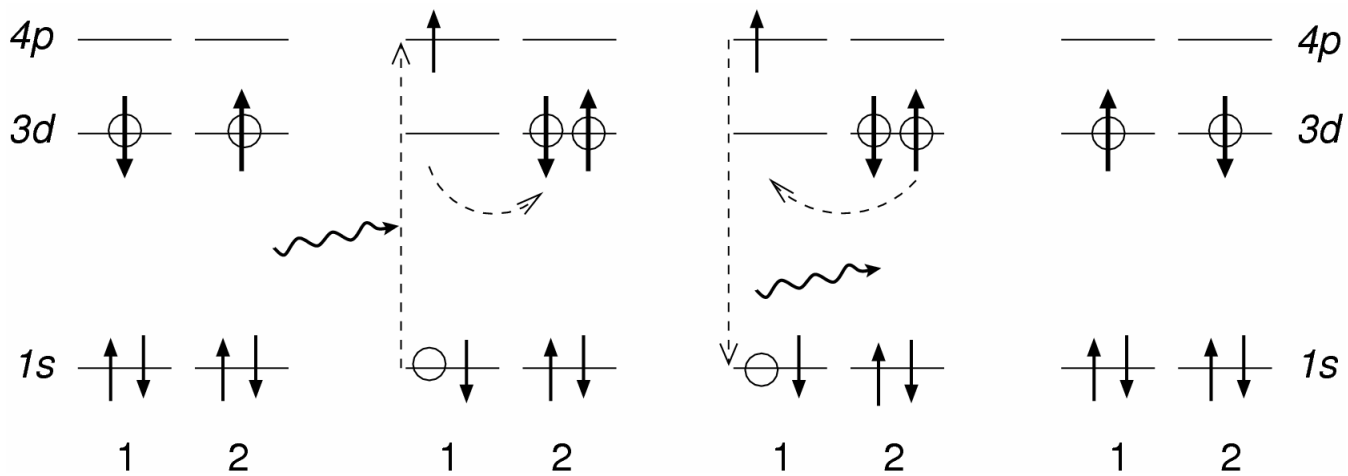


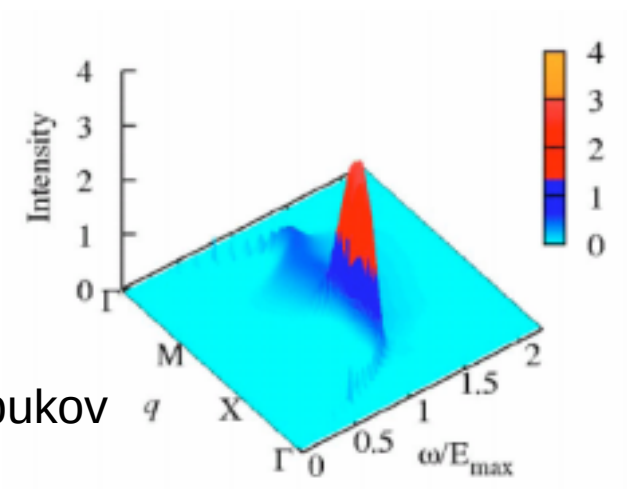
Momentum



Possibilities?

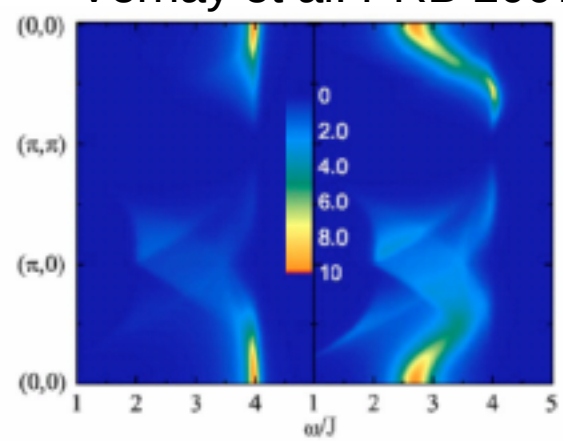
- Two magnon excitation





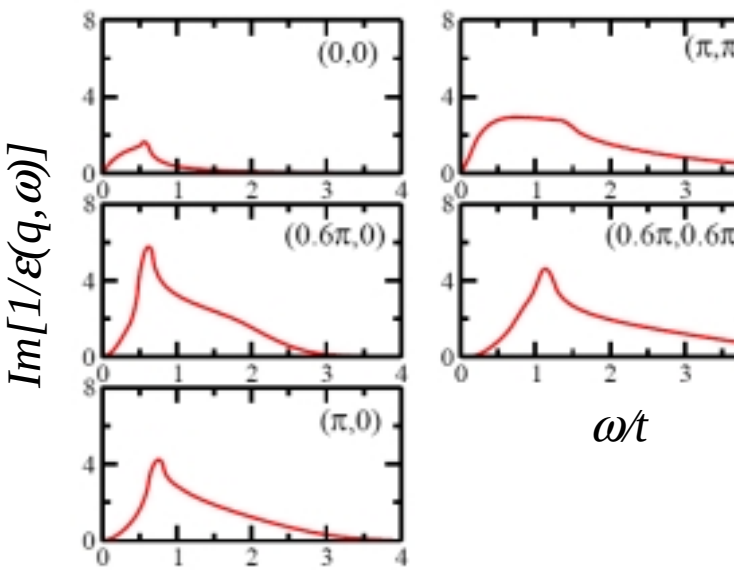
Donkov+Chubukov
PRB 2007

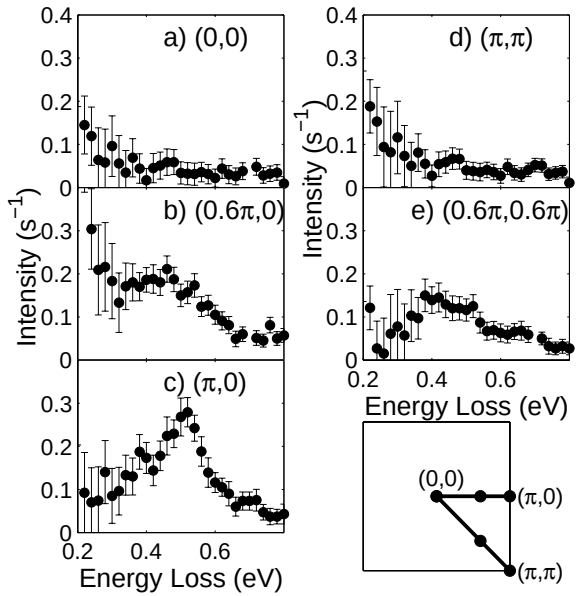
Vernay et al. PRB 2007



More possibilities?

- Charge 2e boson (Choy, Leigh, and P. Phillips)

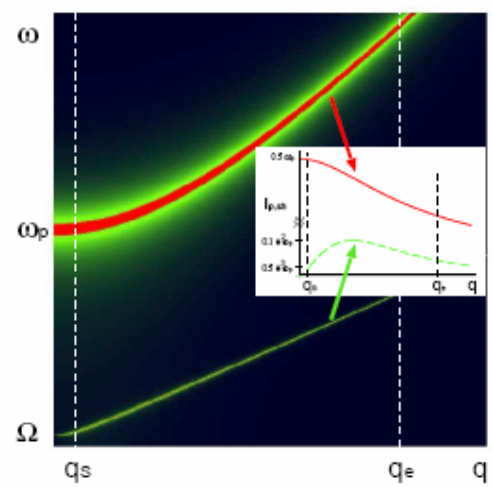




- What about d-d excitation?
  - $d(3z^2-r^2)$ to $d(x^2-y^2)$ is not experimentally identified yet in cuprates
  - But…

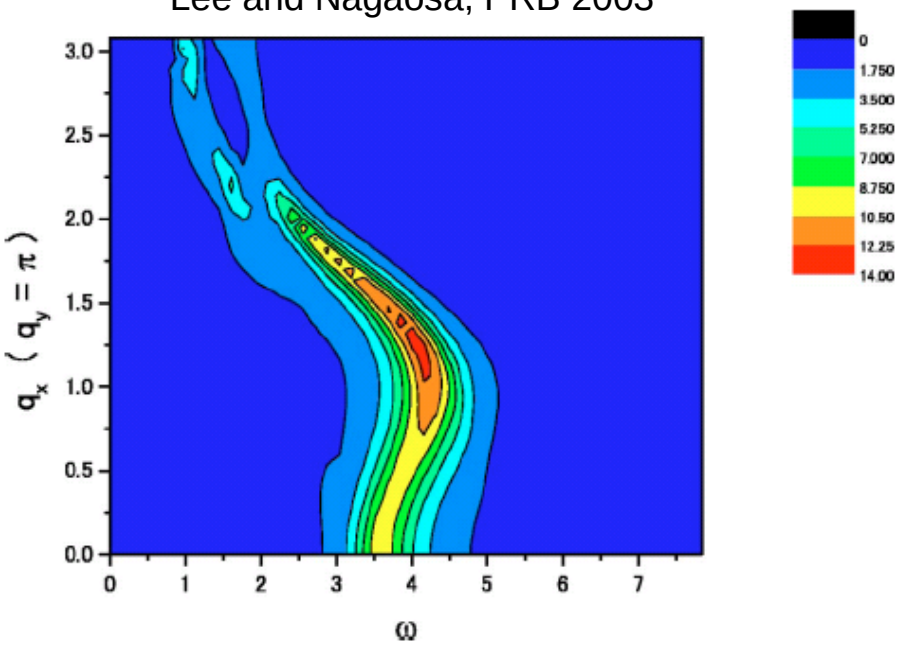
Perhaps in the future?

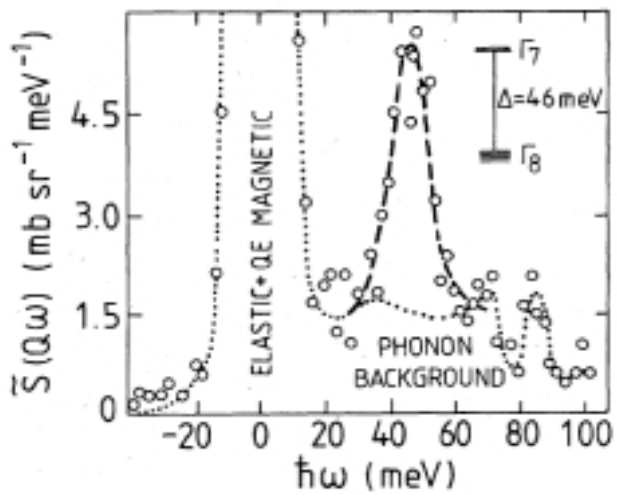
Cvetkovic et al., cond-mat/0607402



COLLECTIVE MODES IN THE SUPERCONDUCTING . . .

Lee and Nagaosa, PRB 2003





CeB6, Zirngiebl et al. PRB 1984

NSLS-II wish list

- Improved energy resolution
  - 10-20 meV resolution will allow us to study excitations in the 50-100 meV range
  - Pseudo gap in high Tc superconductors
  - Magnetic excitation
  - Crystal field in Rare-earths
  - Magnon-phonon-electron interaction
- Smaller beam size (20 nm?)
  - Scanning IXS?
  - Probing electronic structure in inhomogeneous sample

Thanks to

- Toronto
  - Jung Ho Kim

  - David Ellis

- John P. Hill (BNL)
- Girsh Blumberg (Lucent)
- APS XOR
  - T. Gog
  - D. Casa
  - Y. Shvydko
- Samples
  - N. Motoyama, K. M. Kojima, and S. Uchida (U. Tokyo)
  - F. C. Chou (MIT)
  - S. Wakimoto (JAEA)
  - G. Gu, M. Huecker (BNL)
  - S. Komiya, Y. Ando (CRIEPI)
  - H. Zhang (Toronto)

Advanced Photon Source

NSERC CRSNG

Canada Foundation for Innovation

Fondation canadienne pour l'innovation

Conclusion and Outlook

- Resonant Inelastic X-ray Scattering can probe — For La_2CuO_4
  - d-d orbital excitations — ~1.8 eV
  - Charge-transfer gap — ~2.2 eV
  - Dispersion bandwidth — ~0.3 eV
  - Perhaps magnetic excitation — ~0.5 eV
    - Needs more work
    - Mid-IR excitation at (π, 0) In $La_{2-x}Sr_xCuO_4$

    ➔ two magnon?
- But more is in the store if we can improve energy resolution
  - Goal: study < 100 meV physics