# Inelastic Studies of Complex Ground States: Spin, Charge, Lattice and Orbital Excitations in Novel Materials

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# Collaborators

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**$(La,Sr)_2NiO_4$**

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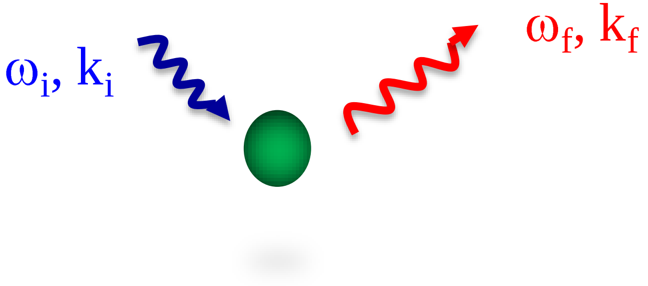
# Outline

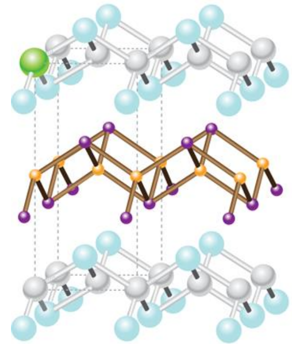
## 1. Introduction

- Excitations
- Inelastic x-ray scattering: Cross-section




## 2. Lattice Excitations

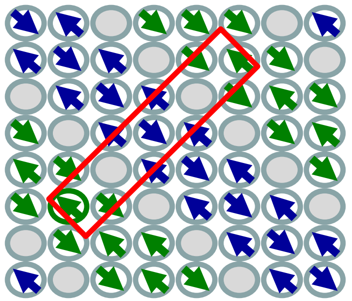
- Anomalous phonons in $SmFeAsO_{1-x}F_x$



## 3. Magnetic Excitations

## 4. Orbital and Charge Excitations

- Dispersive and non-dispersive excitations in $La_{2-x}Sr_x(Mn,Ni)O_4$



# Ground State and Excitations

A complete picture of a system requires knowledge of both its ground state and its excitations.

(*in high-energy physics parlance: Vacuum and particles*)

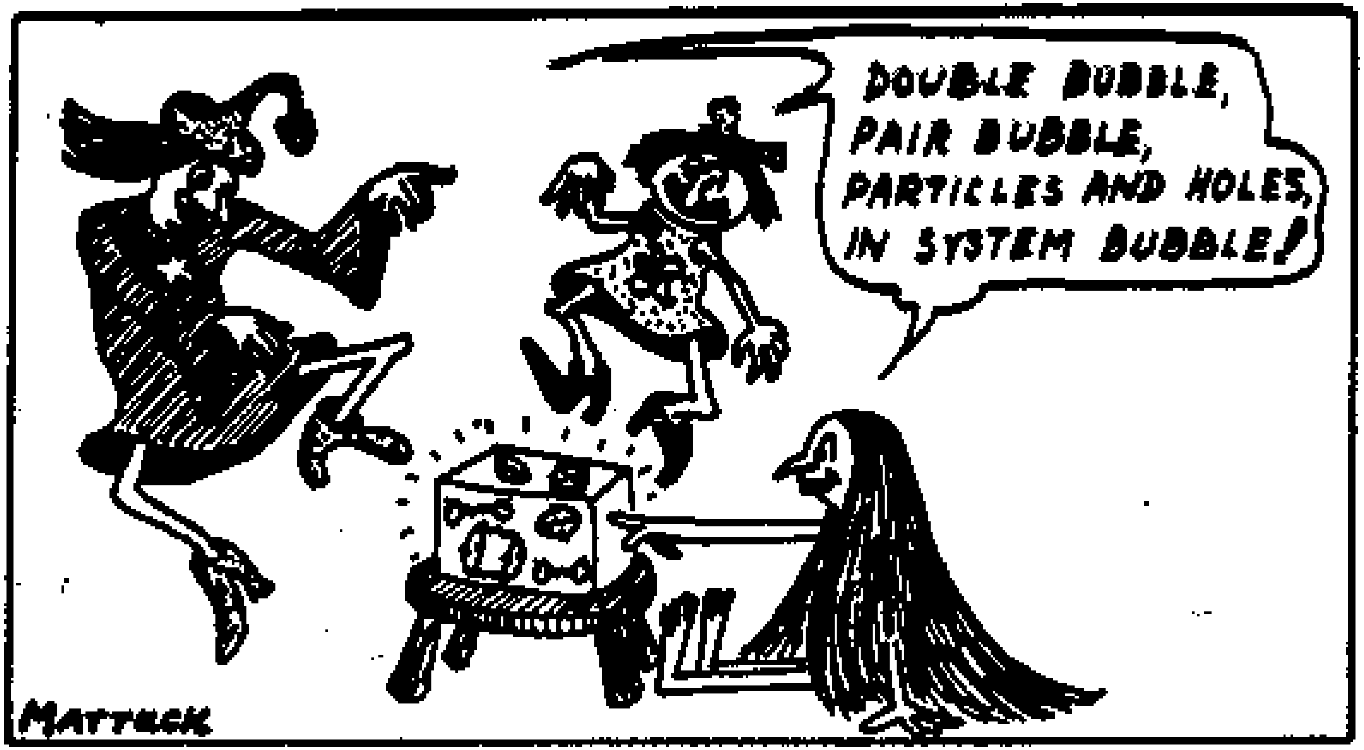
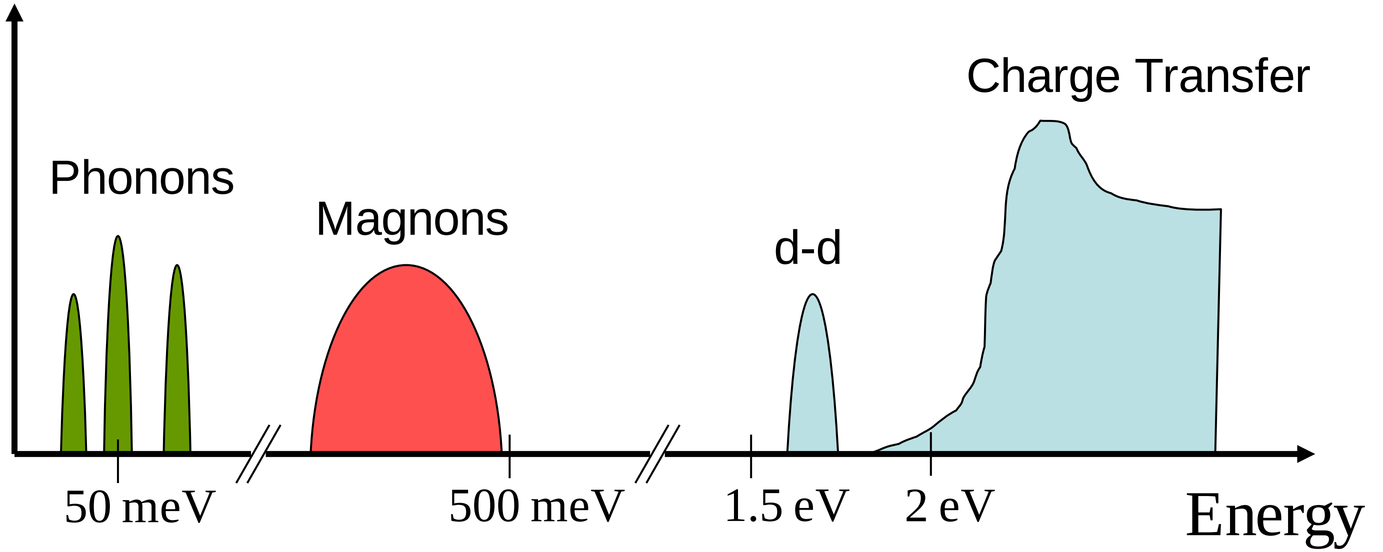



Fig. 1.5 *Modern View of a Many-body System in its Ground State*

# Excitations in Condensed Matter

1) Excitation spectrum determines the dynamic response of these materials.
2) Excitation spectra provide stringent test of theory.
3) "High-energy physics" of systems often controls their behavior:

$t \sim 1$ eV, $U \sim 8$ eV, $\Delta \sim 2$eV

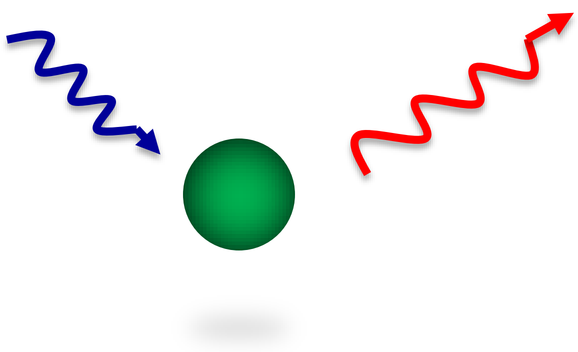



Need a momentum and energy resolved probe ⟹ IXS

# Inelastic X-ray Scattering

## Inelastic X-ray Scattering

$\omega_i$, $k_i$



$\omega_f$, $k_f$

$q = k_i - k_f$

$\omega = \omega_i - \omega_f$

Elastic scattering ($\omega$ = 0) gives static properties.

Inelastic scattering ($\omega \neq 0$) gives dynamic properties.

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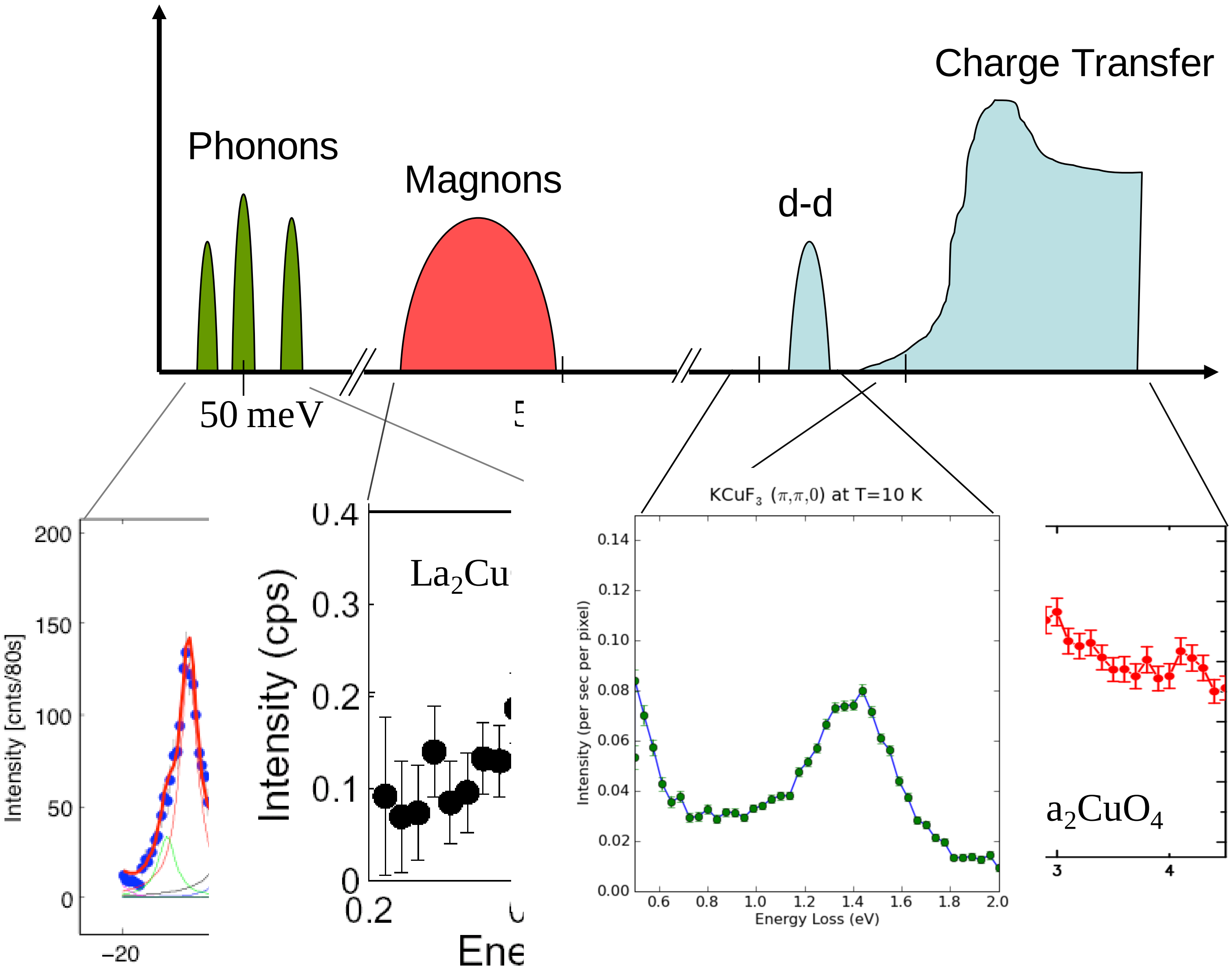
# Inelastic X-ray Scattering

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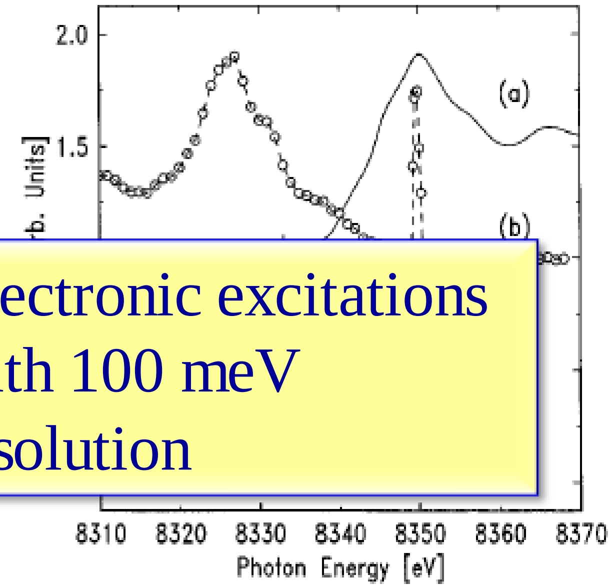
# Cross-Section

$$\frac{d^2\sigma}{d\Omega d\omega} \propto \left| \langle f \,|\, H_{\rm int} \,|\, i \rangle + \sum_{|n\rangle} \frac{\langle f \,|\, H_{\rm int} \,|\, n \rangle \langle n \,|\, H_{\rm int} \,|\, i \rangle}{E_i - E_n + i\Gamma} \right|^2$$

Non-resonant scatterin
(weak)

Phonons with few meV resolution




Kao et al, PRB 1996,

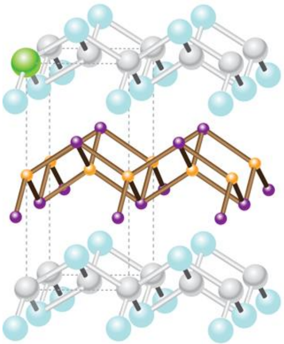
Electronic excitations with 100 meV resolution

Resonant IXS is > 100 x Non-Resonant IXS!

1. Introduction

2. Lattice Excitations

- Anomalous phonons in $SmFeAsO_{1-x}F_x$



3. Magnetic Excitations

4. Orbital and Charge Excitations

# "Iron Age" of Superconductivity

Discovery of a new class of iron-based superconductors

- Large variety of "families" of materials
  - "1111" LaFeAsO, CeFeAsO, SmFeAsO…
  - "122" $BaFe_2As_2$, $CoFe_2As_2$, ….
  - "111" LiFeAs, …
  - "11" FeSe, FeTe. …
  - + ….?
- Highest $T_c$=55 K in $SmFeAsO_{1-x}F_x$
- Metallic parent compounds, with SDW antiferromagnetism
- LDA seems to work reasonably well (small U)
- s-wave superconductors

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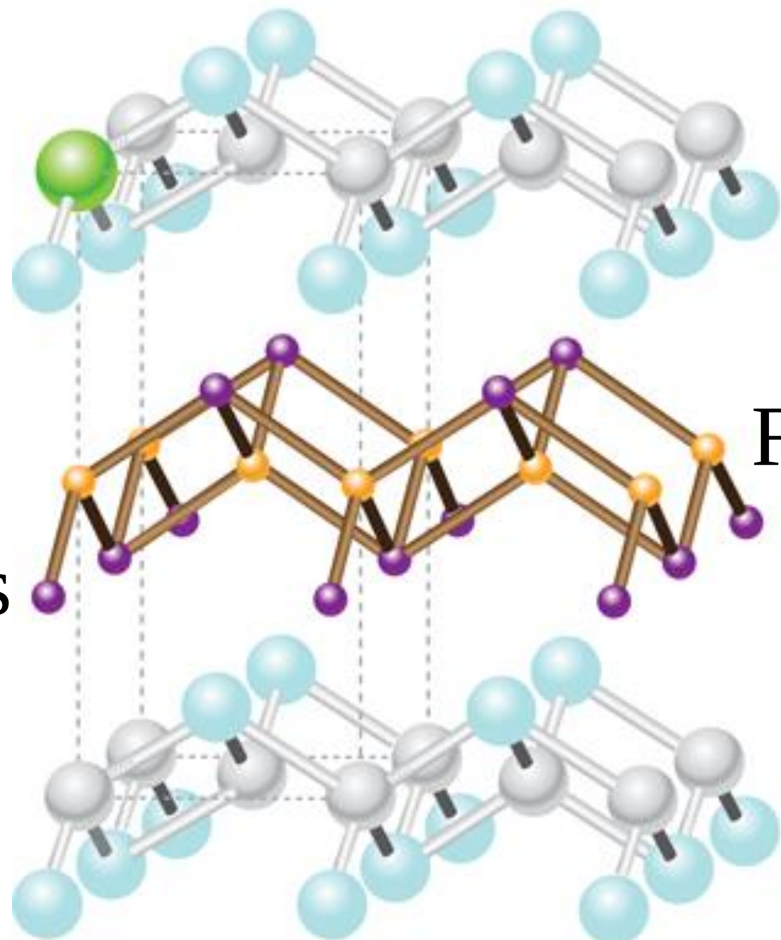
# $SmFeAsO_{1-x}F_x$

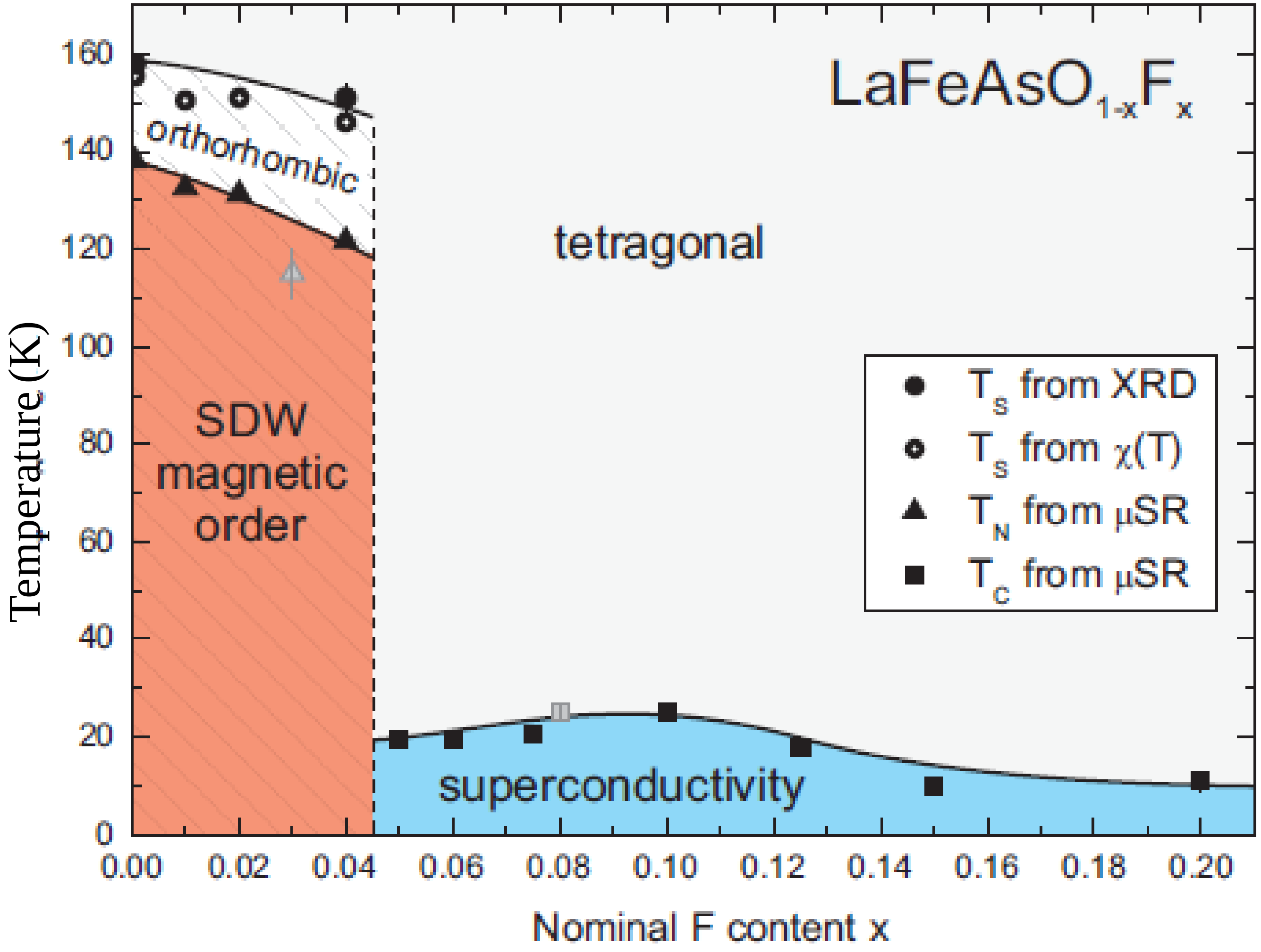
## Crystal Structure



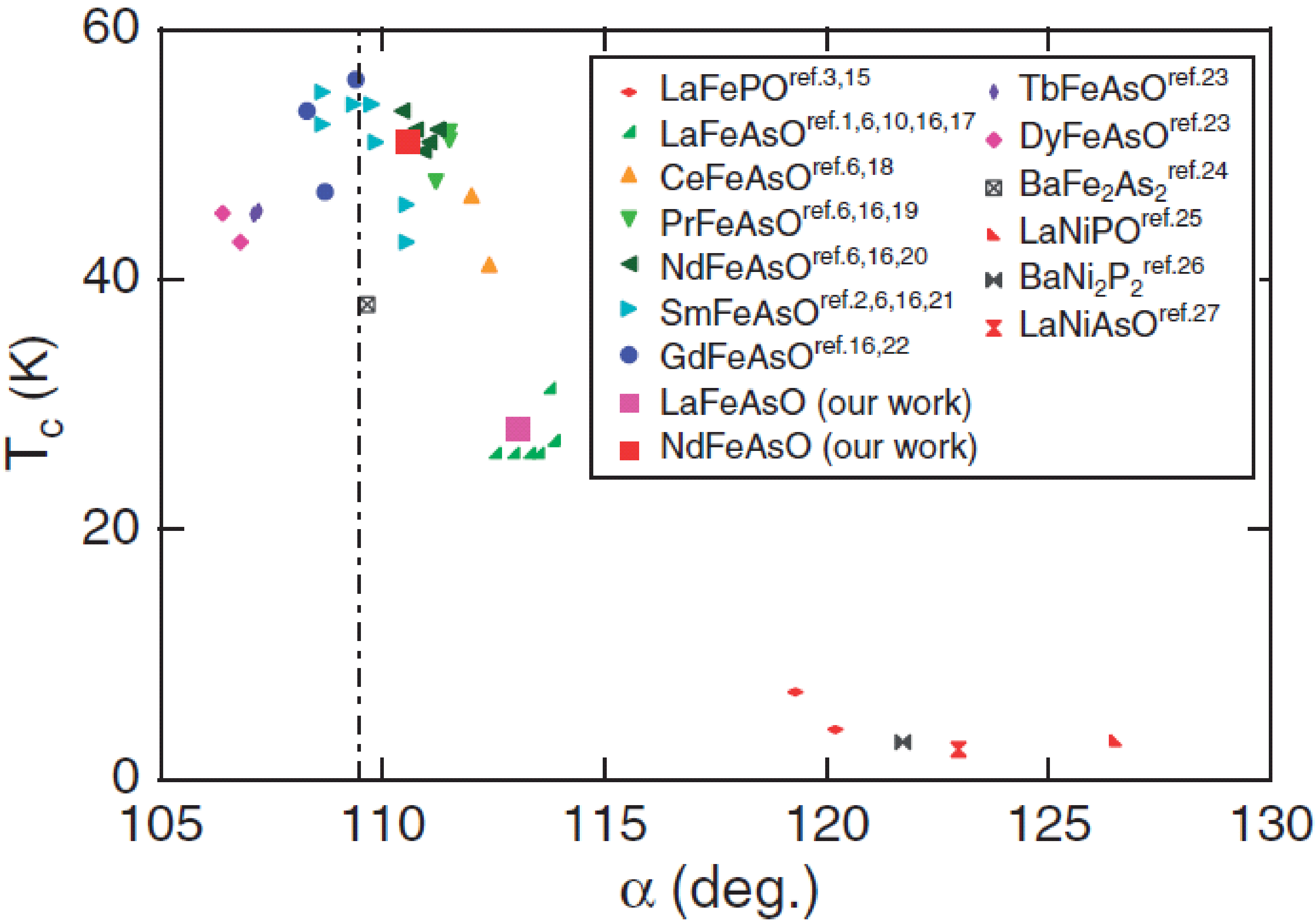

Carrier blocking layer

Carrier conducting layer

# Phase Diagram



# Pairing Mechanism




Lee et al. J. Phys. Soc Jpn (2008)

Need measurements of the phonons

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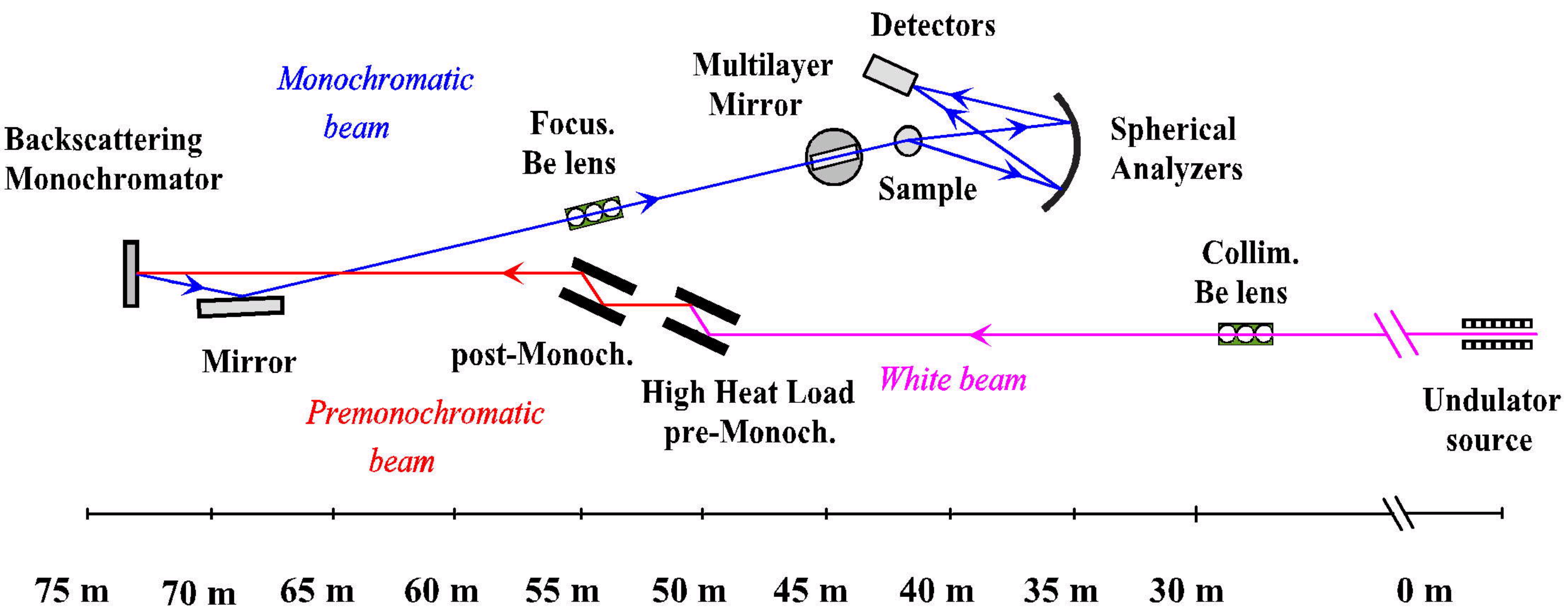
# High Resolution Inelastic Scattering




ID 28 at the ESRF

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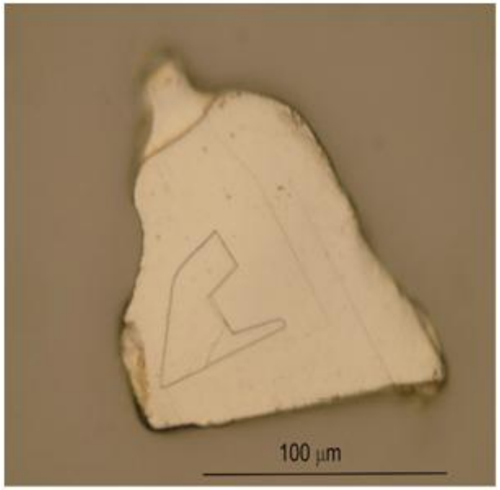
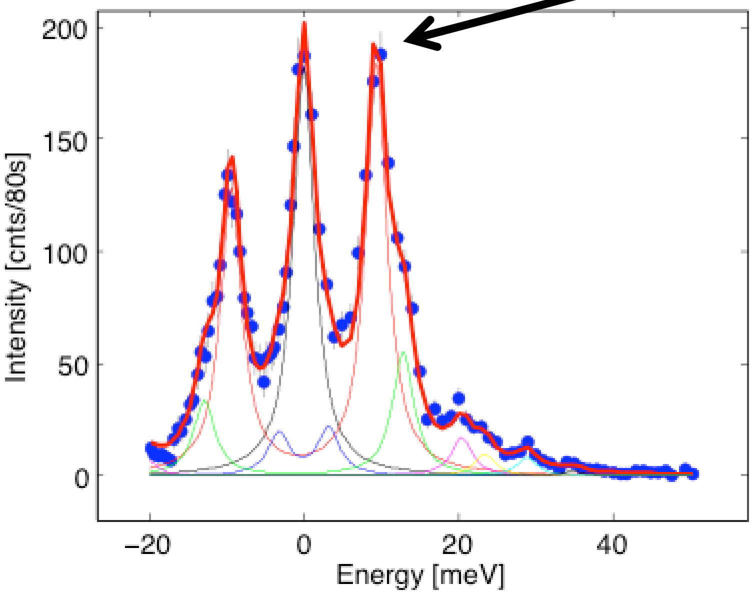
# Inelastic X-Ray Scattering

Single crystal $SmFeAsO_{0.85}F_{0.15}$ ($T_c$=52 K)

~ 3cps




Volume ~ 100 x 100 x 5 $\mu m^3$




ID28, ESRF, $\Delta E$ = 3 meV

LeTacon, *et al.*, PRB (2009)

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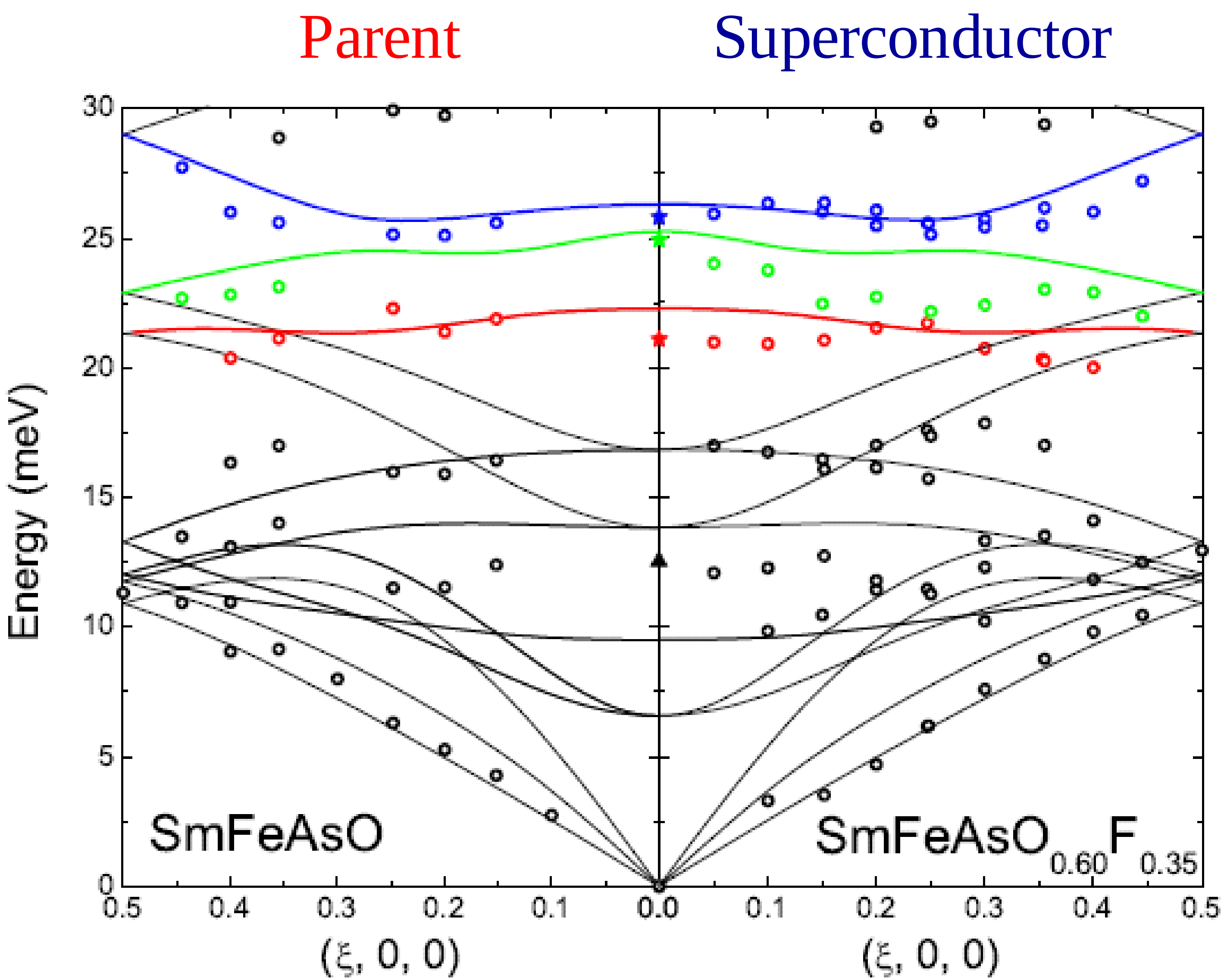
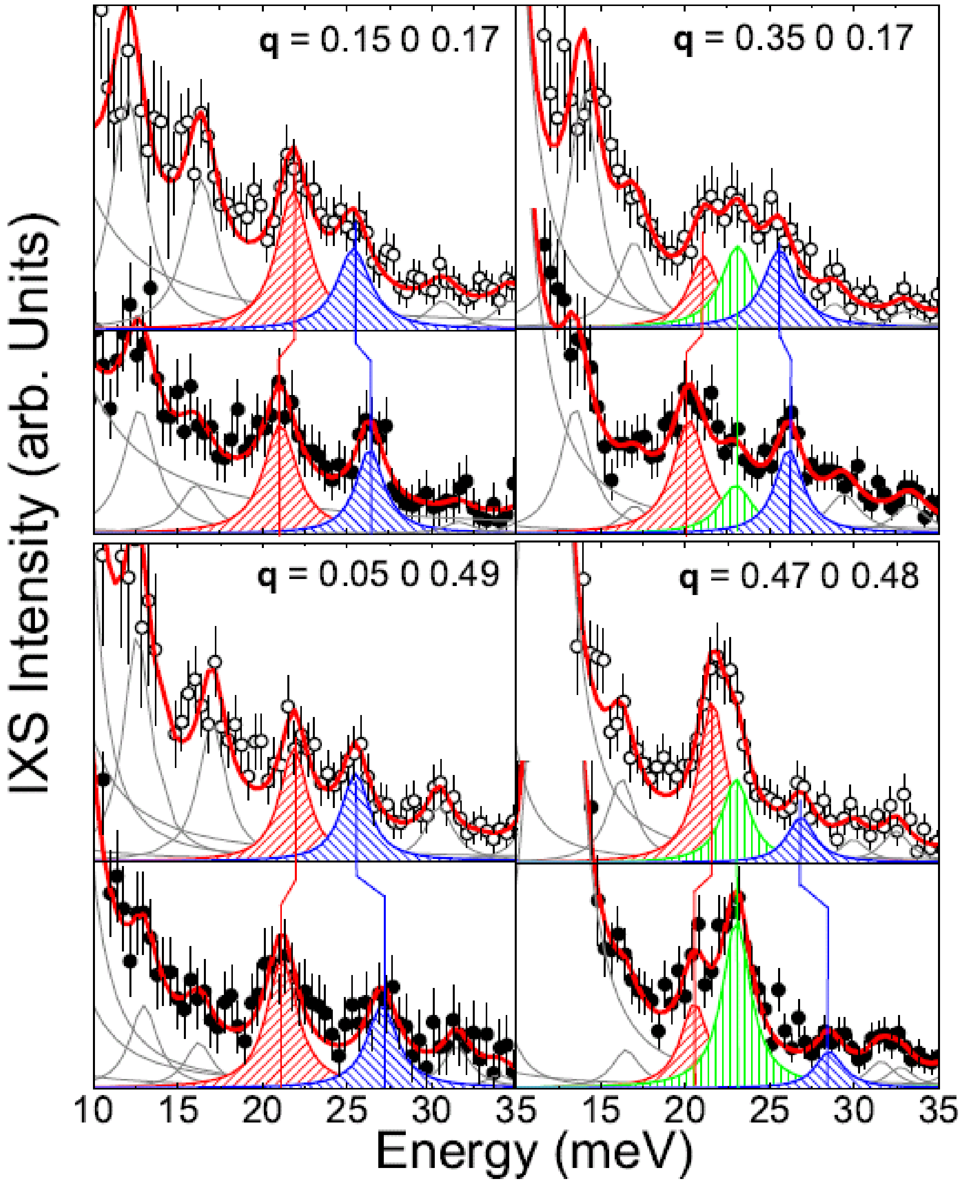
# Single Crystal Phonon Dispersions




Deviations are seen from DFT calculations

# Doping Dependence




Parent

Super-
conductor

Parent

Super-
conductor

Certain phonons around 23 meV show unusual shifts on doping

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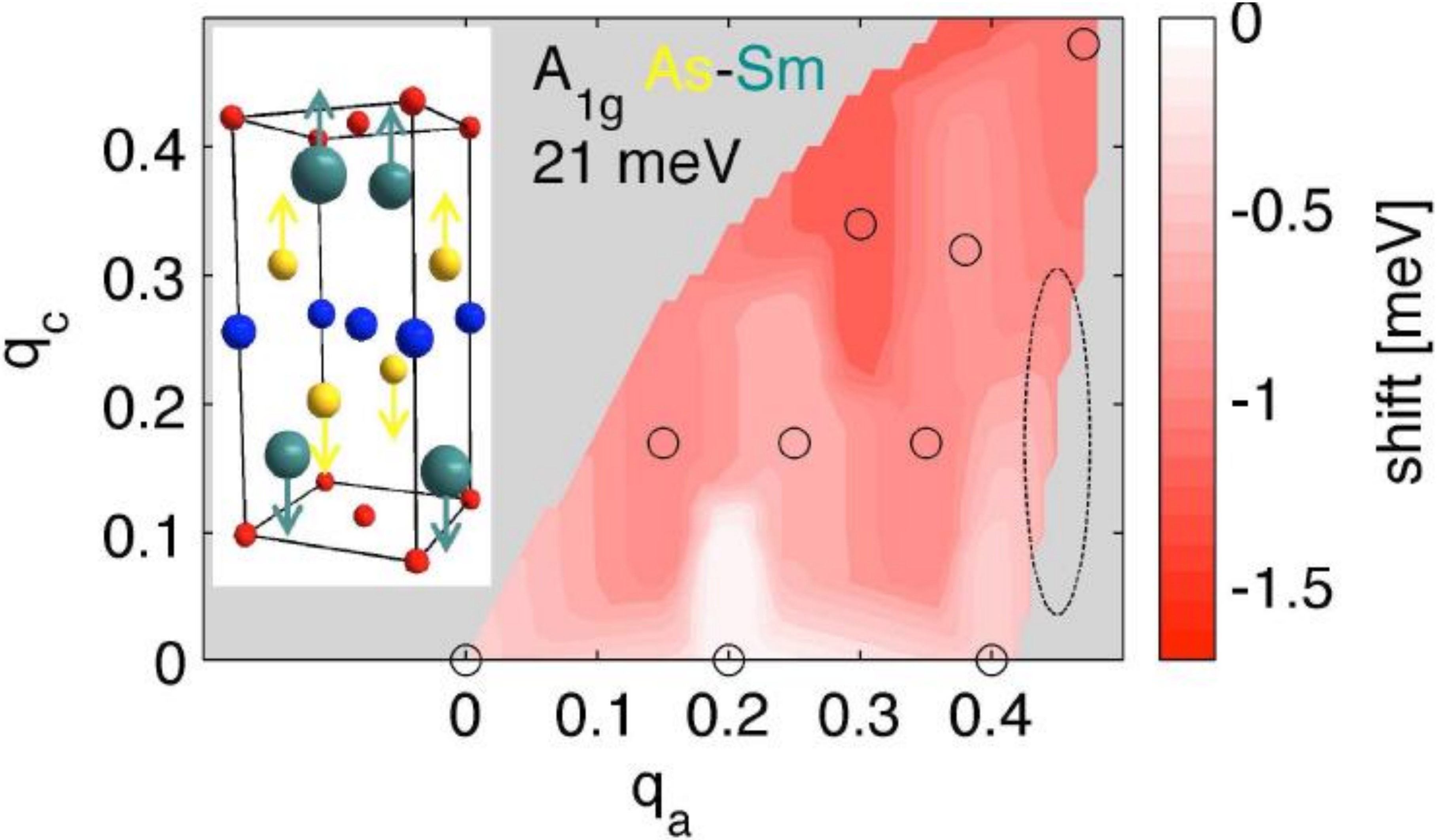
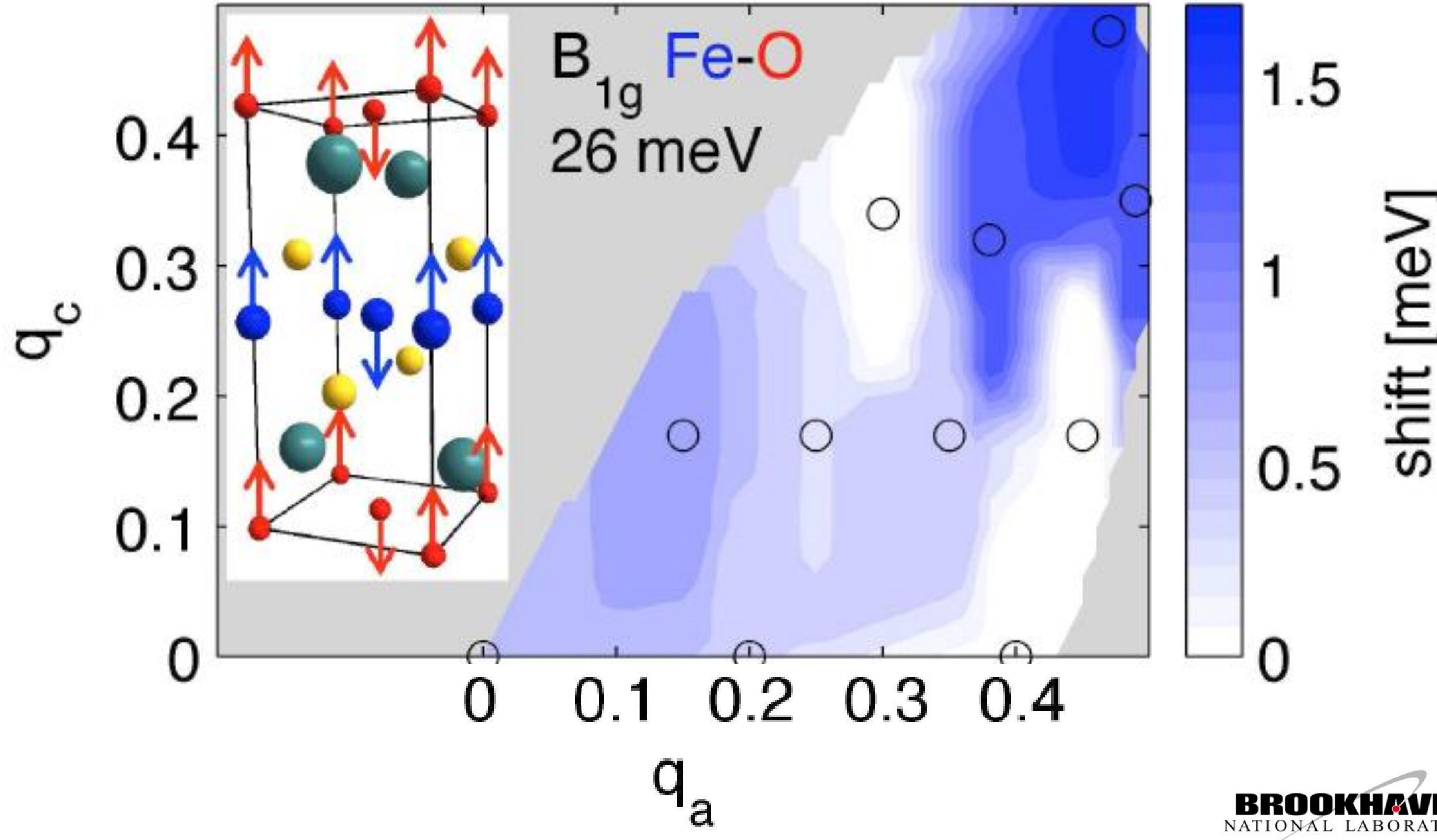
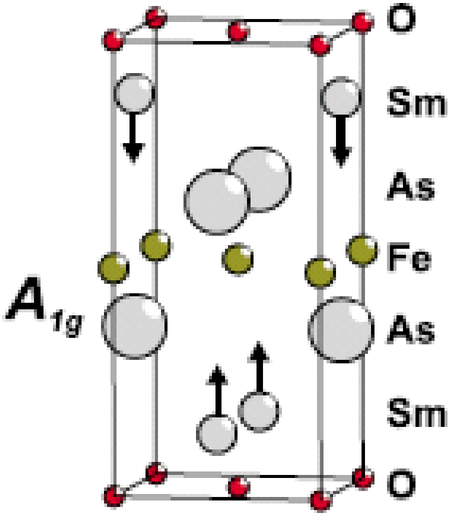
# 21 meV Mode



# 26 meV Mode

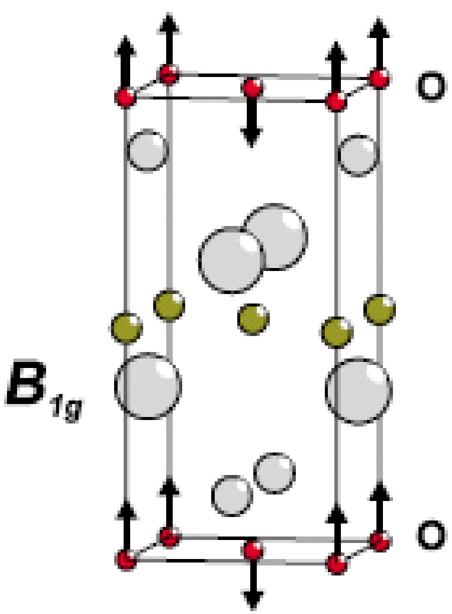


# Other Nearby Phonon Modes




Sm + As mode ~ 25 meV

To first order, these don't couple to the FeAs tetrahedra – and therefore do not couple to the Fe electronic degrees of freedom




Oxygen mode ~ 43 meV

From Hadjiev *et al.* PRB (2008)

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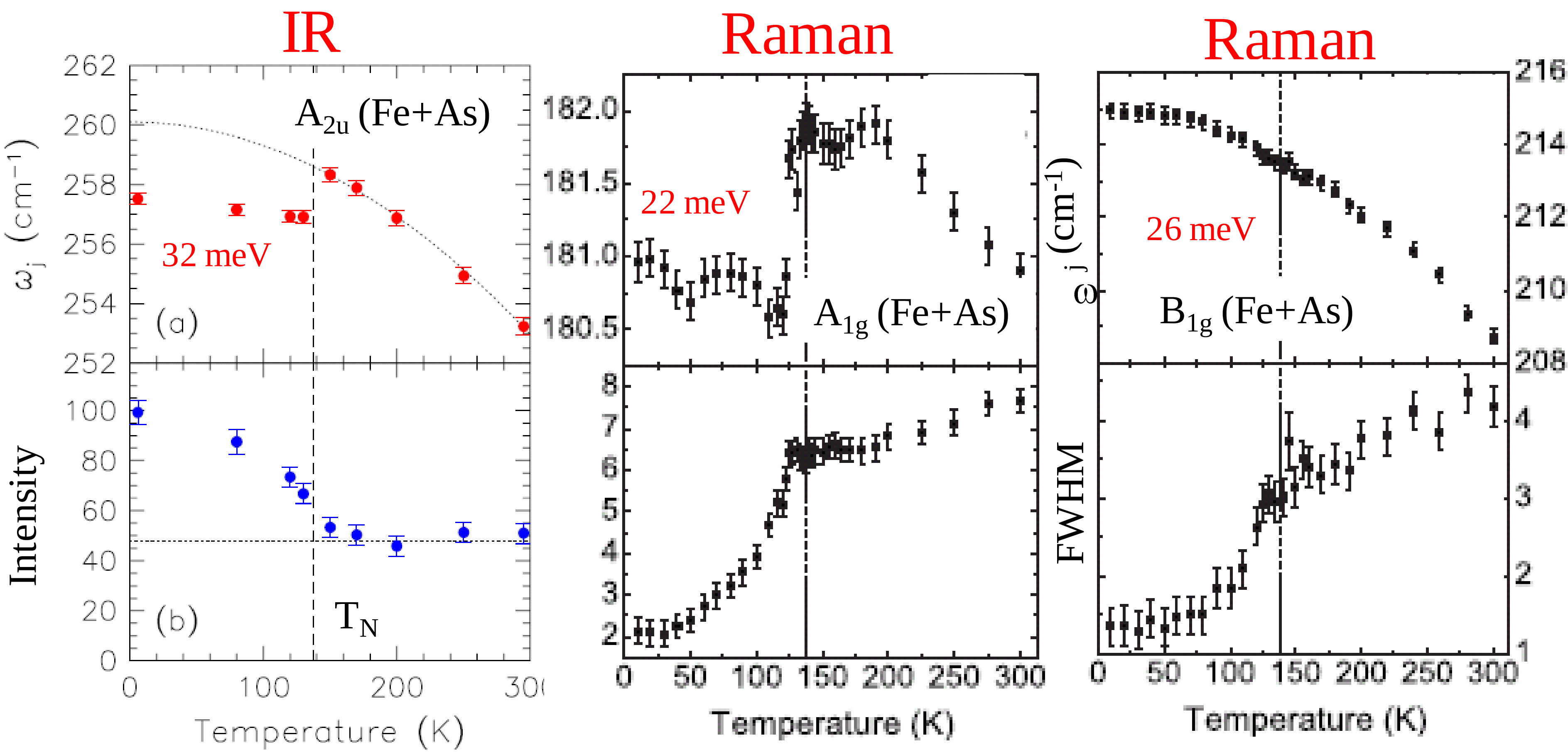
# Spin/Orbital-phonon Coupling

$BaFe_2As_2$



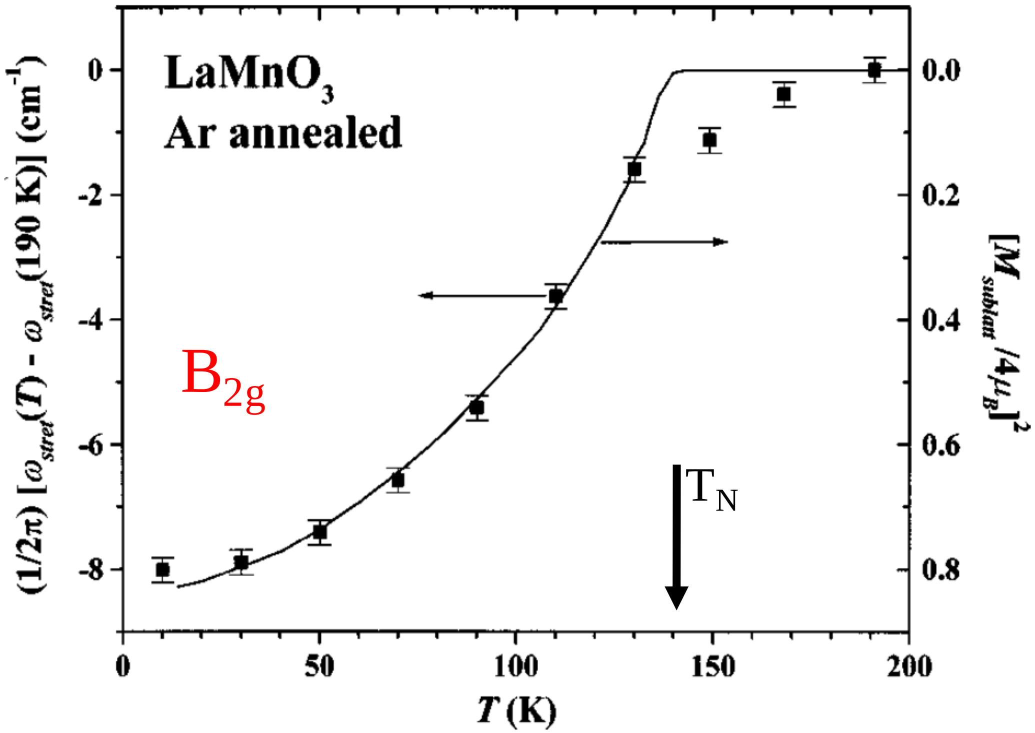

Akrap *et al.* PRB (2009)

Rhalenbeck *et al.* PRB (2009)

# Other Systems

## $LaMnO_3$

Raman




Grandon *et al.* PRB (1999)

Classic example of spin-phonon coupling. See softening of phonon proportional to magnetic order parameter.

This is not seen in the pnictides.

# Iron-pnictides

- It is possible to carry out high-quality phonon measurements on small volume single crystals with IXS. Limitation is number of instruments!
- Certain c-axis modes are anomalously renormalized on doping and with temperature. These shifts have unusual momentum dependence – signature of electron-phonon coupling.
- The affected modes are magnetically active. Suggests spin or orbital fluctuations may be important.

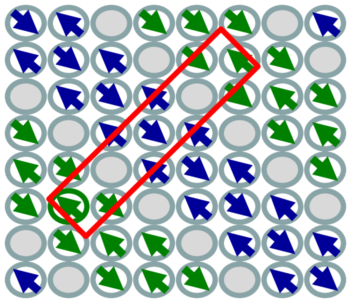
1. Introduction

2. Lattice Excitations

3. Magnetic Excitations

4. Orbital and Charge Excitations

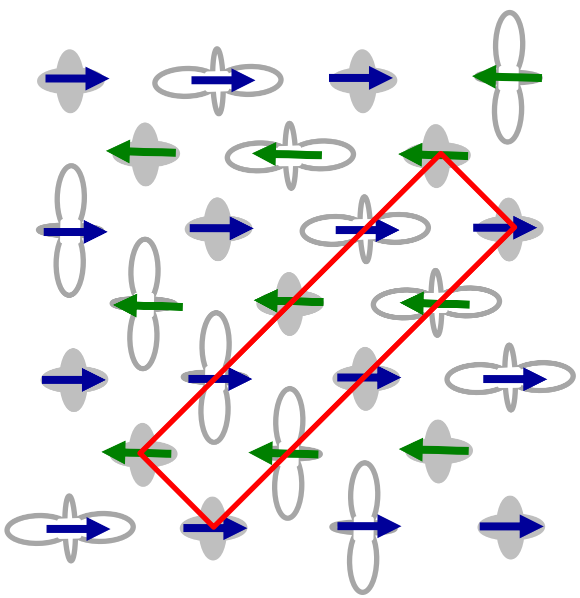
- Dispersive and non-dispersive excitations in $La_{2-x}Sr_x(Mn,Ni)O_4$



# Electronic Ground States

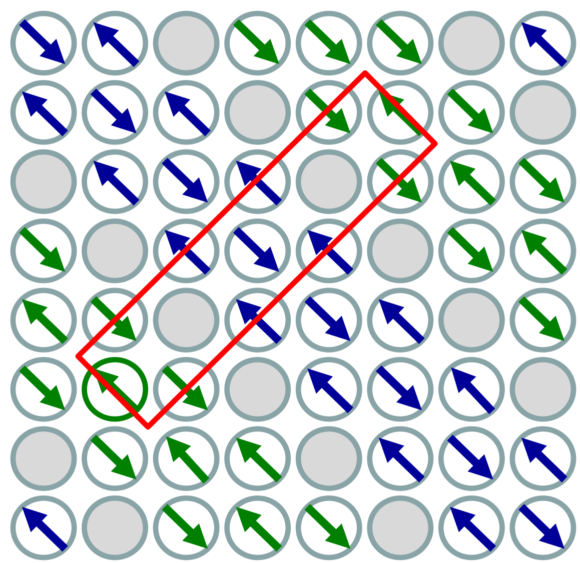
Transition metal oxides exhibit a range of charge, spin and orbitally ordered ground states (new *vacua*). What are the excitations (new *particles*) associated with these states?

Manganites



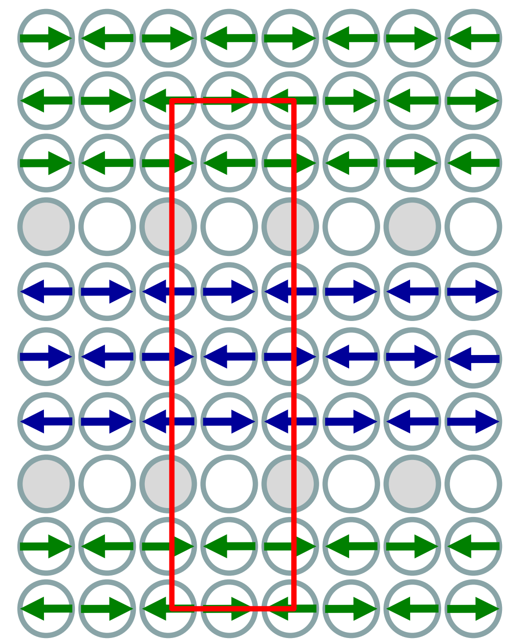
$MnO_2$ plane, $n_h$=0.5

Nickelates



$NiO_2$ plane, $n_h$=0.25

Cuprates



$CuO_2$ plane, $n_h$=0.125

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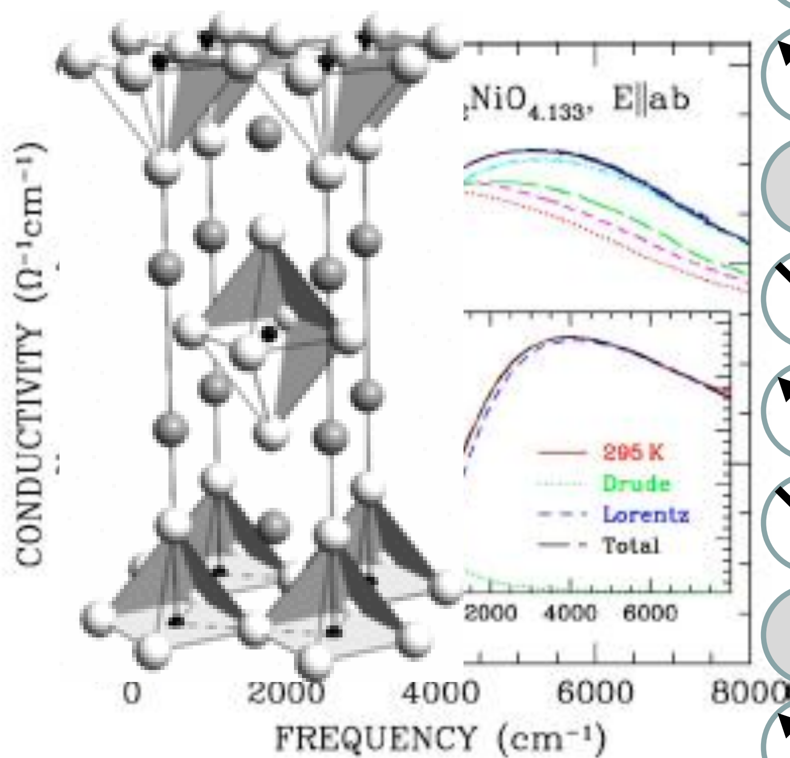
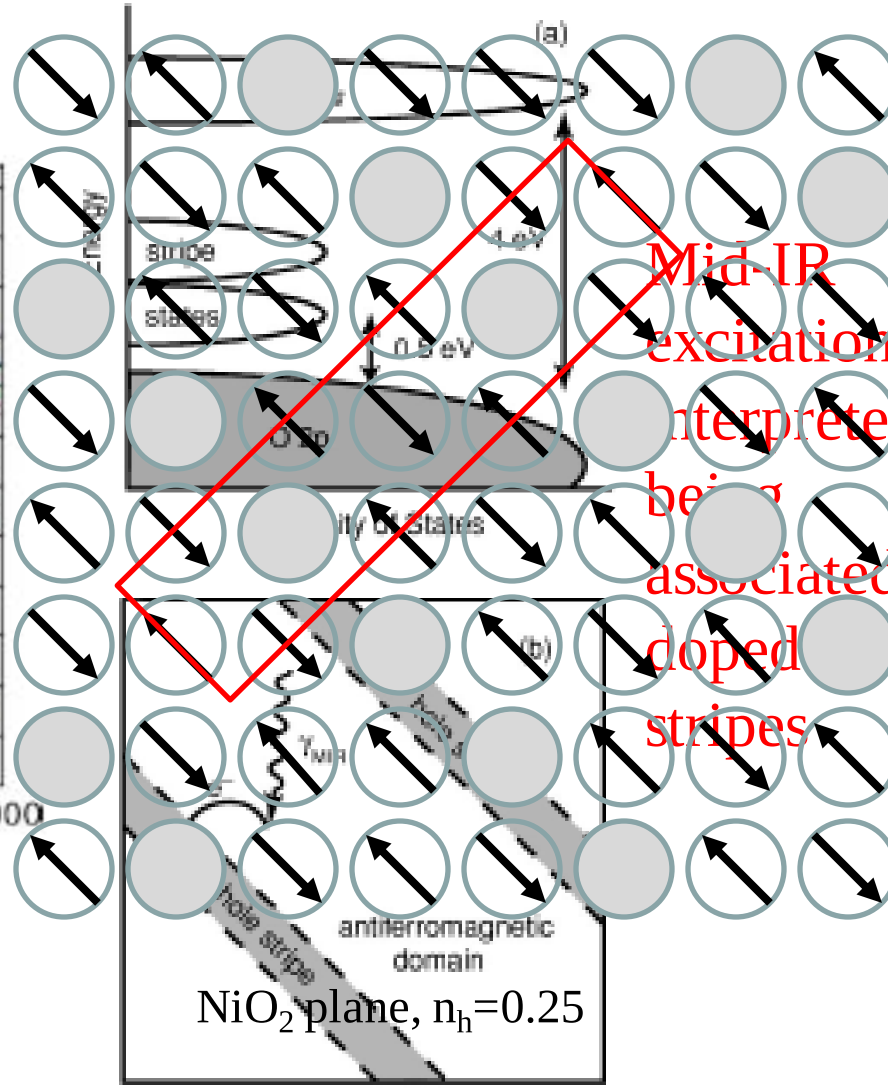
# $La_{2-x}Sr_xNiO_4$: A Stripe Ordered System

## Optical Conductivity




Mid-IR excitations interpreted as being associated with doped holes in stripes

Homes et al. PRB (2003)

Tranquada et al. Nature (1995)

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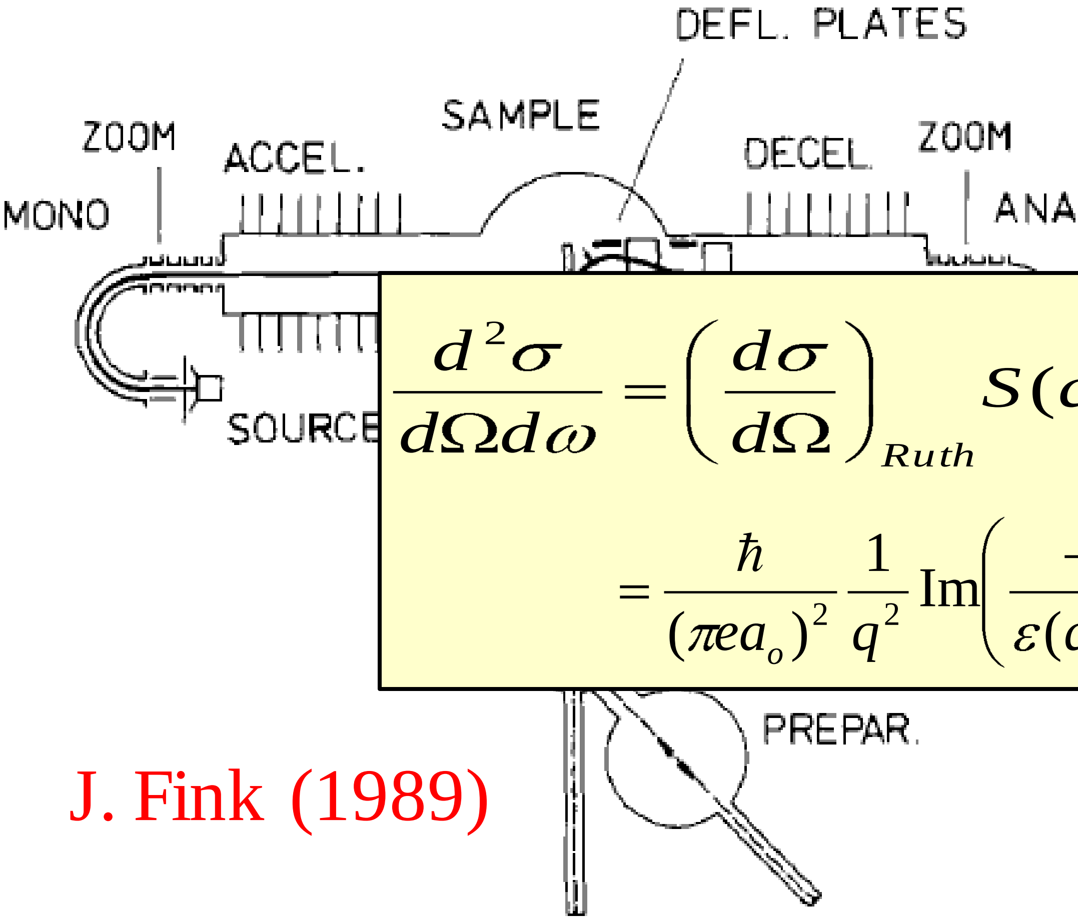
# Electron Energy Loss Spectroscopy




$$\frac{d^2\sigma}{d\Omega d\omega} = \left(\frac{d\sigma}{d\Omega}\right)_{Ruth} S(q,\omega)$$

$$= \frac{\hbar}{(\pi e a_o)^2} \frac{1}{q^2} \mathrm{Im}\left(\frac{-1}{\varepsilon(q,\omega)}\right)$$

$E_o = 170$ keV
$\Delta E = 70$ meV
$\Delta q = 0.04$ $A^{-1}$

J. Fink (1989)

(Instrument now at IFW-Dresden: M. Knupfer)

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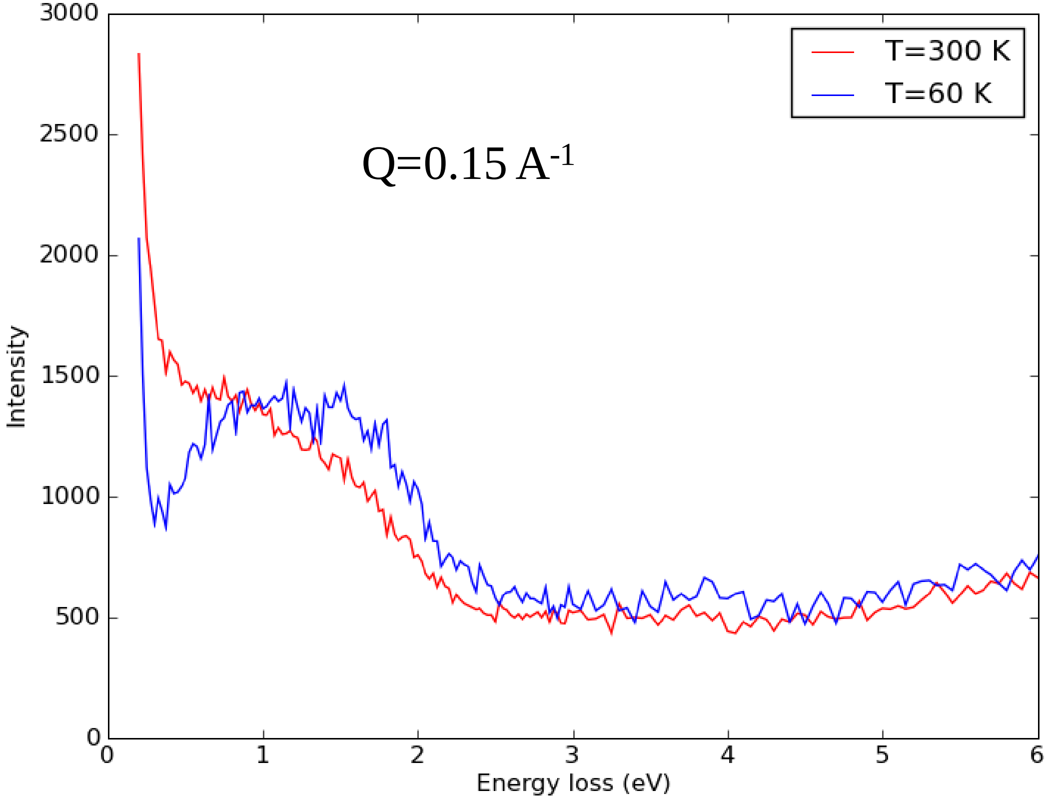
# EELS Data

$La_{1.67}Sr_{0.33}NiO_4$




At small momentum transfers, EELS reproduces optical data – observe opening of gap in stripe phase

R. Kraus, J. Geck, M. Knupfer, B. Buchner and JPH, unpublished data

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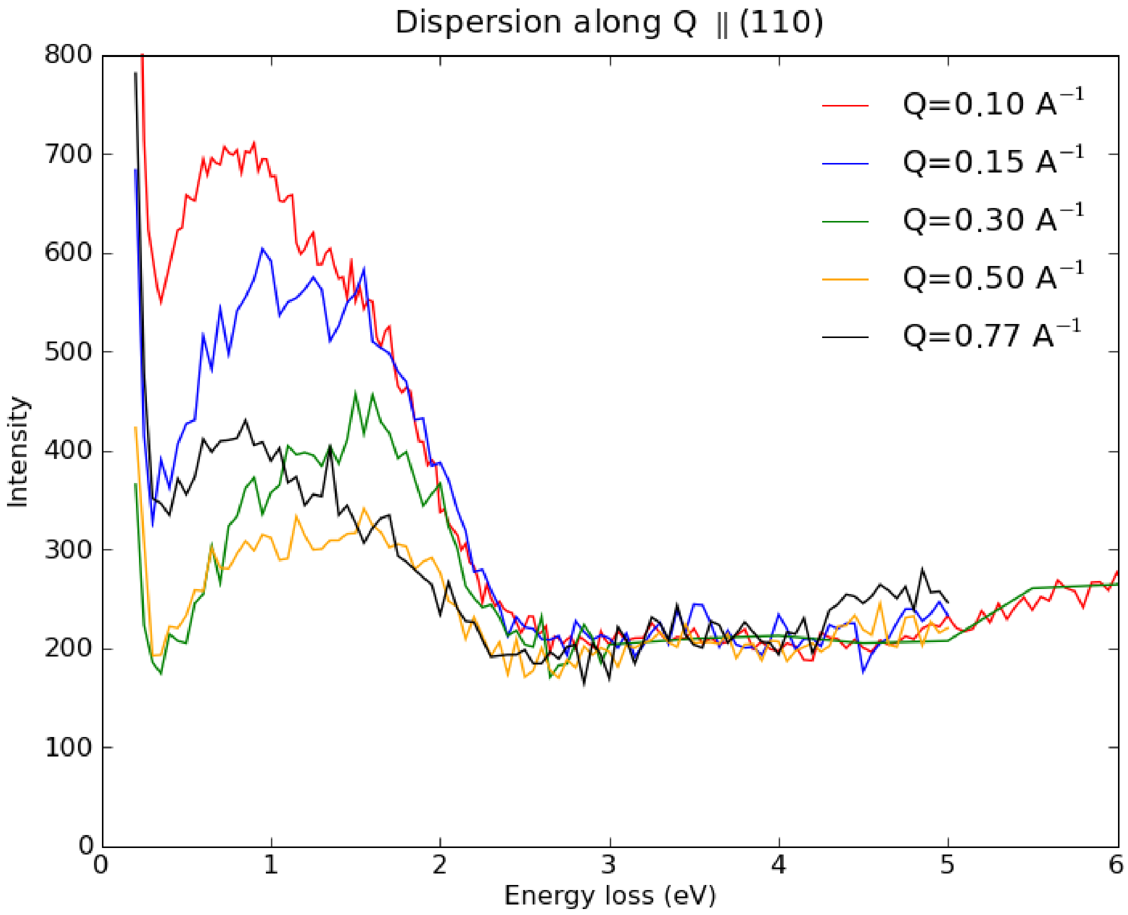
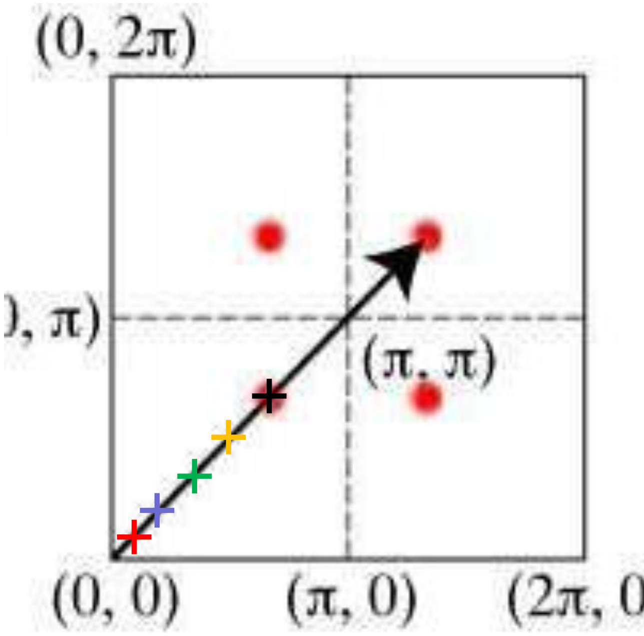
# Momentum Dependence

$La_{1.67}Sr_{0.33}NiO_4$




R. Kraus, J. Geck, M. Knupfer, B. Buchner and JPH, unpublished

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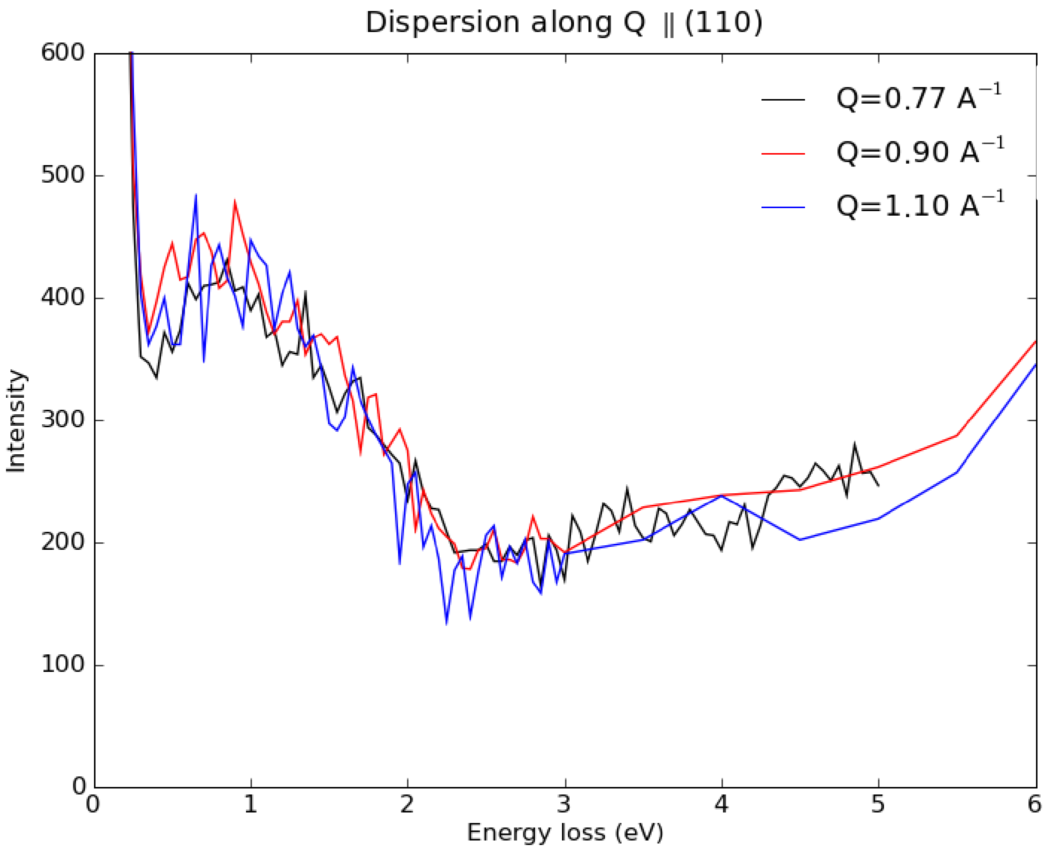
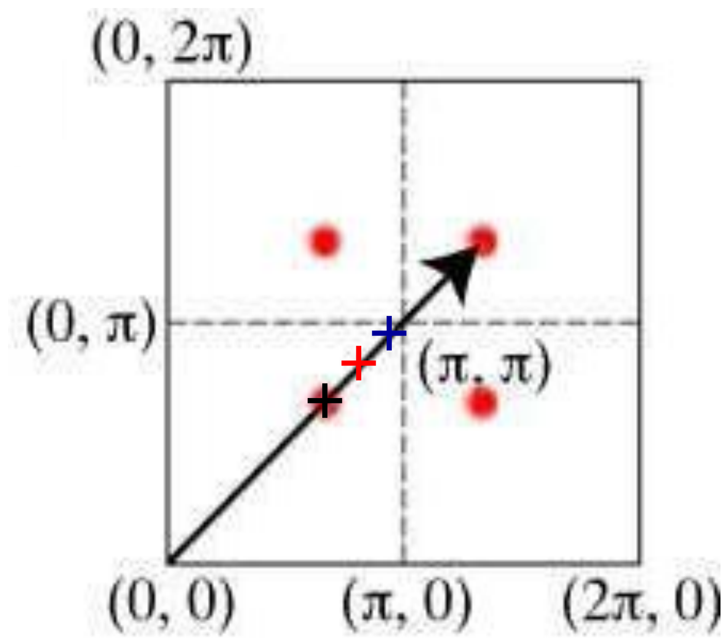
# Momentum Dependence

$La_{1.67}Sr_{0.33}NiO_4$

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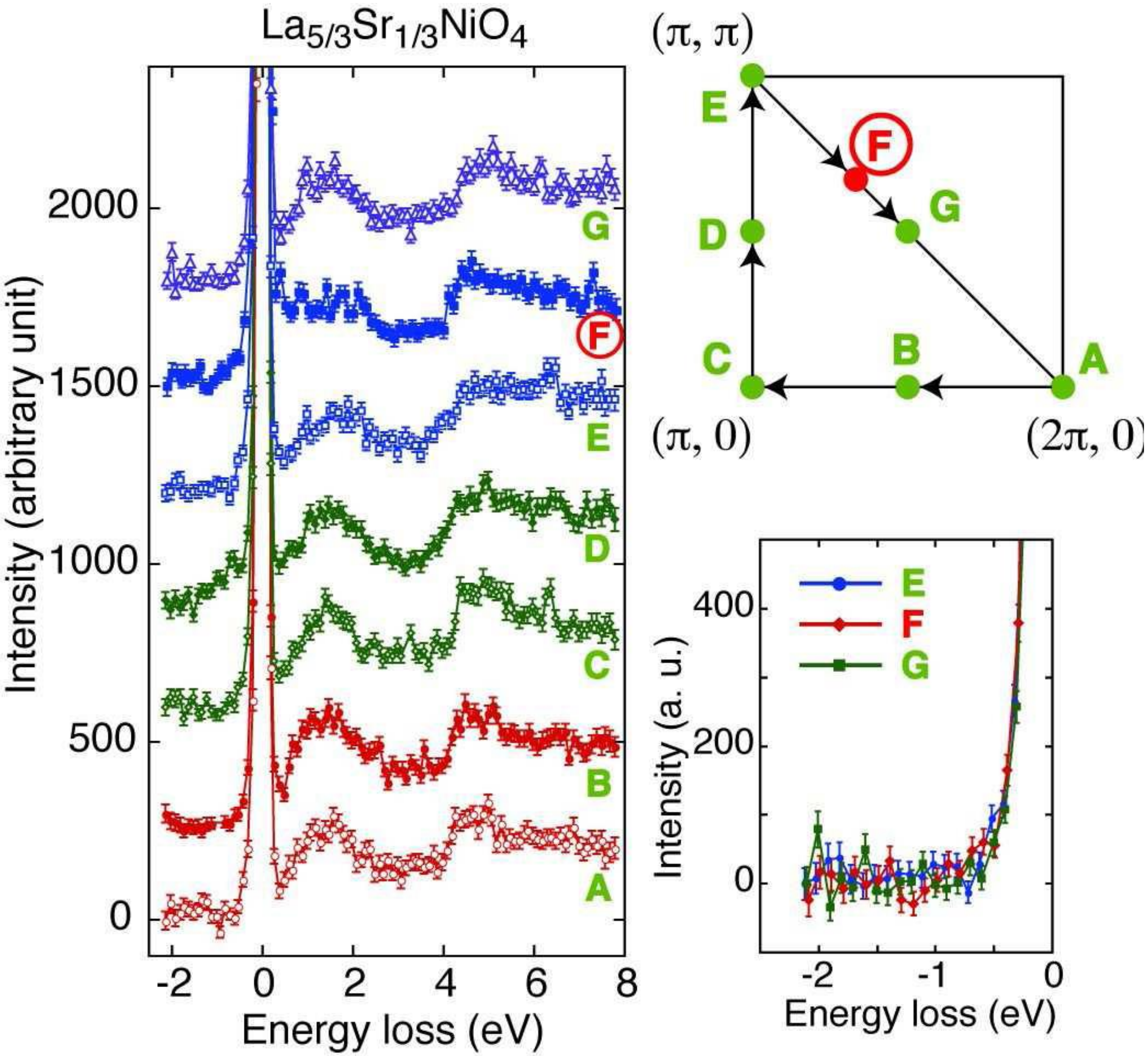
# RIXS data




Wakimoto *et al*, PRL (2009)

$\Delta E$ = 150 meV

MERIX, APS
Ge(642)

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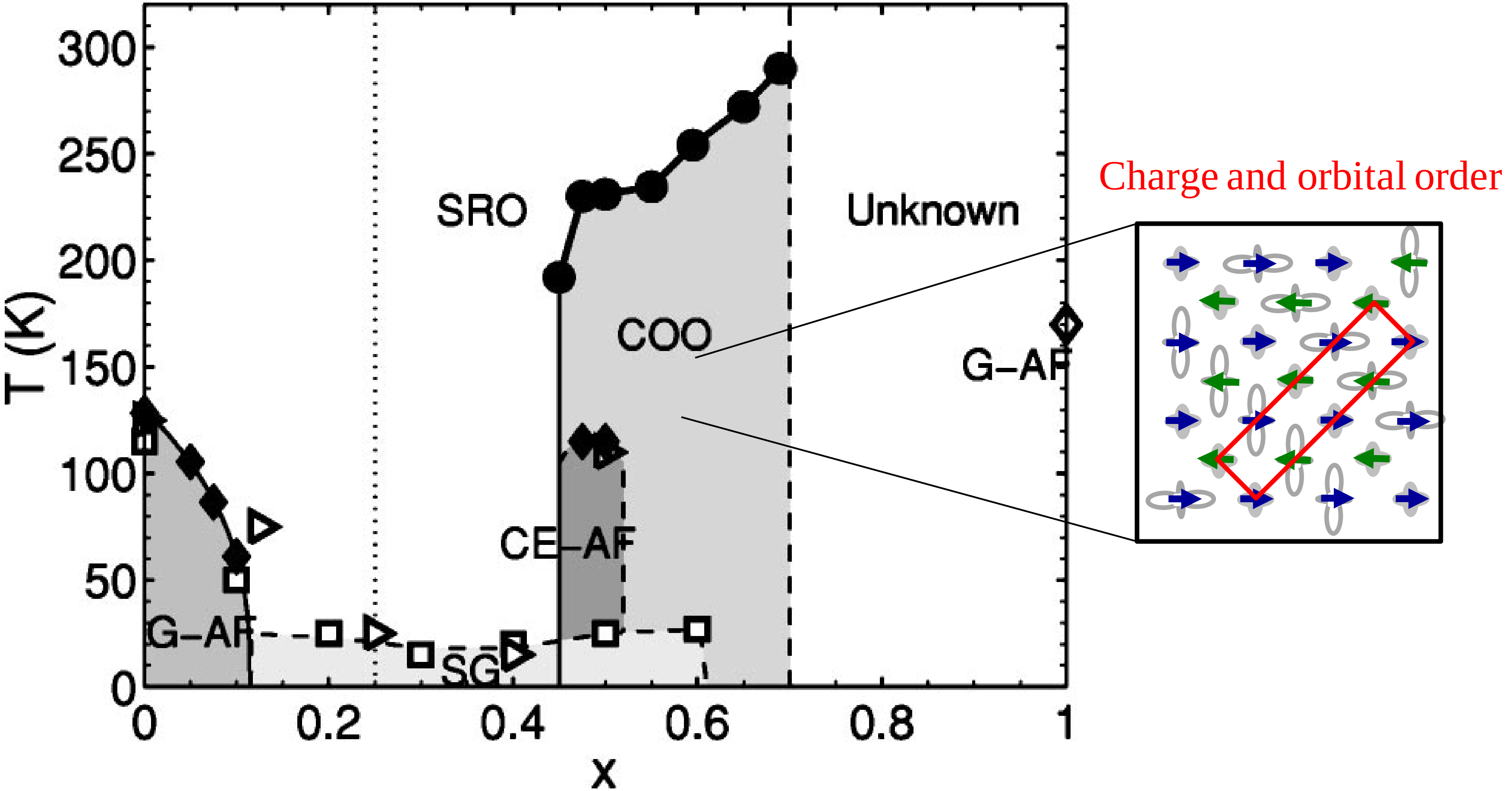
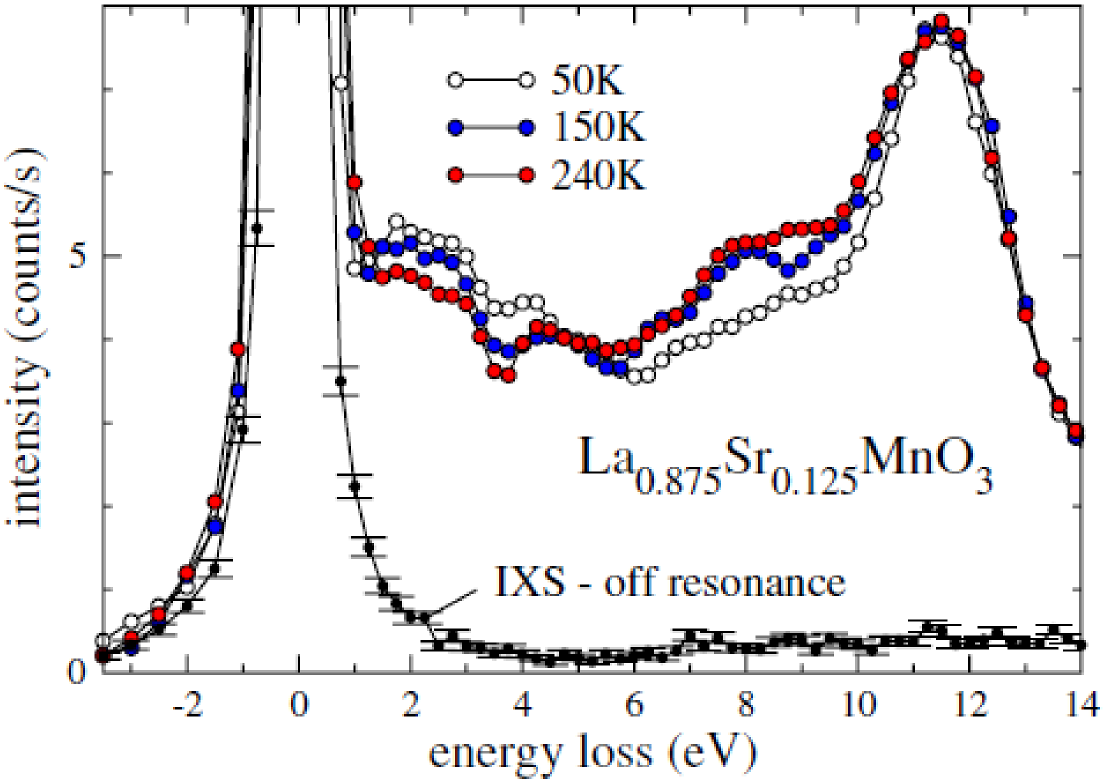
# $La_{1-x}Sr_xMnO_4$




Larochelle *et al.* PRB (2005)

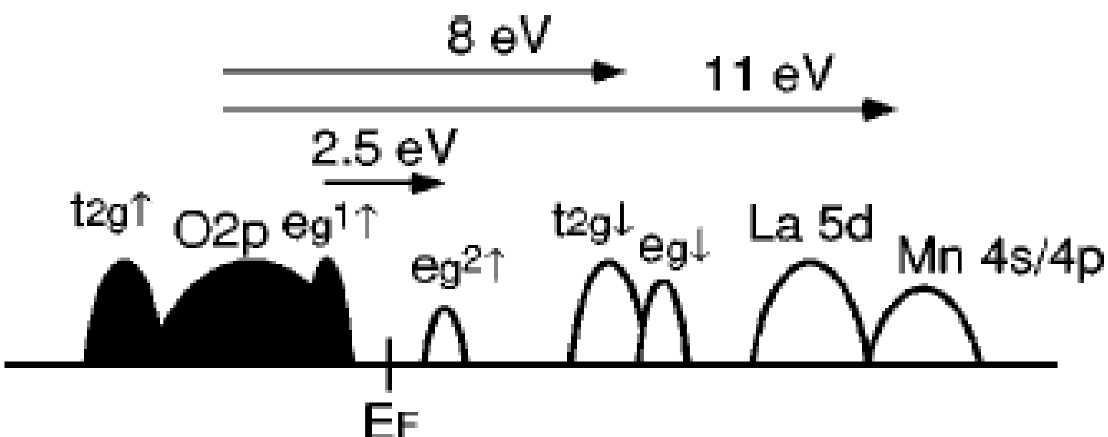
# Manganite K-edge RIXS



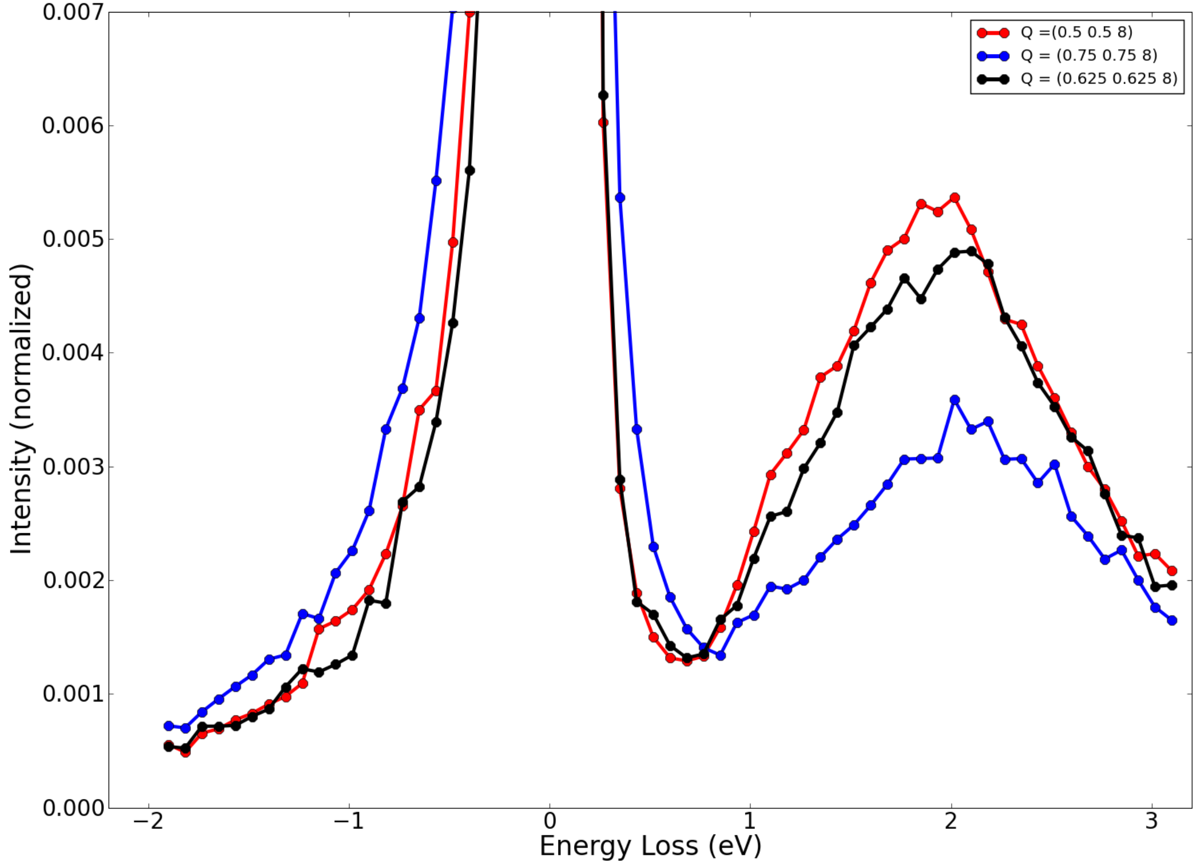

9ID ΔE=300 meV

Grenier *et al.* PRL (2005)

Density of states



# Charge and Orbital Order




● Q=(0.625,0.625,8)

● Q=(0.5,0.5,8) CO

● Q=(0.75,0.75,8) OO

ΔE=200 meV

30ID, APS

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# Summary

Inelastic x-ray scattering can probe many of the relevant excitations in condensed matter over the important (q,ω).

- Anomalous renormalization seen in magnetically active phonons in SmFeAs(O,F).

- In stripe-ordered phase in $La_{1.67}Sr_{0.33}NiO_4$, a dispersive charge excitation is observed with EELS at small momentum transfers. Nothing new is seen at the stripe wave-vector (caveat: multiple scattering effects) – in contrast to RIXS results.

- In charge-ordered $La_{1.5}Sr_{0.5}MnO_{4,.}$ A non-dispersive orbital excitation is observed at 2 eV. No new excitation is observed with RIXS at the charge or orbital order wave-vectors.

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