Neutron and Photon Spectroscopy

Part II : Magnetism

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Outline

2.1 Magnetic Excitations

Crystal Field

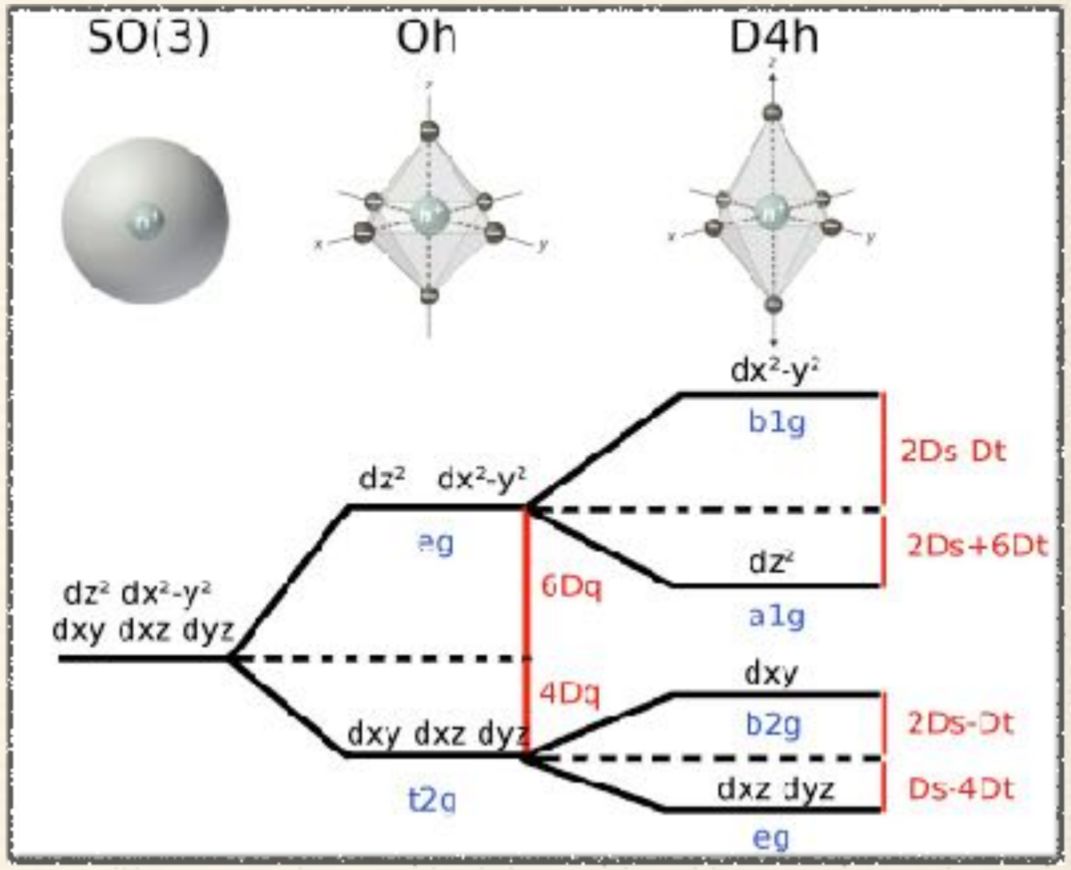
Non spherical environment:

$$\mathcal{H}_{CF} = \sum_{l,m} B_l^m O_l^m$$

O_l^m : Stevens operators (spherical harmonic decomposition of the local charge density)

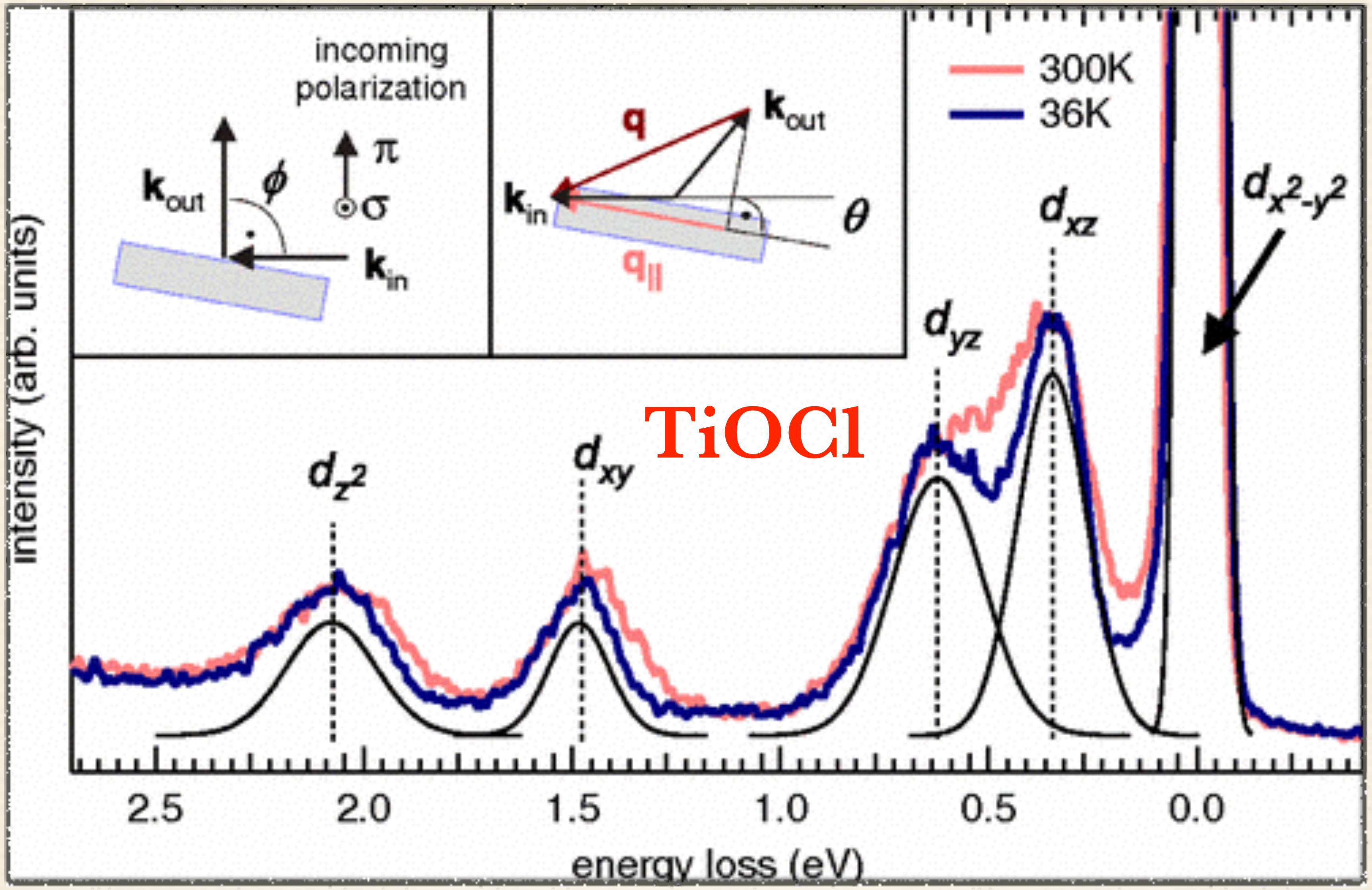
B_l^m : crystal field parameters

Local excitation : non dispersive (sometimes it may be trickier)



Resonant Inelastic X-ray Scattering

dd excitations



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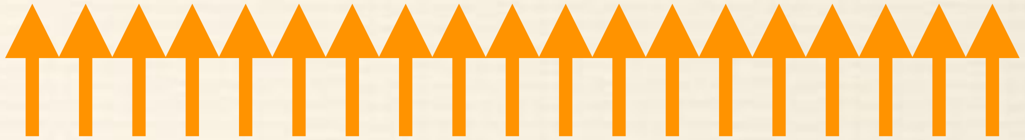
Crystal Field of Ti (3d[1]) L_3-edge (450eV)

Spin Wave - Magnon

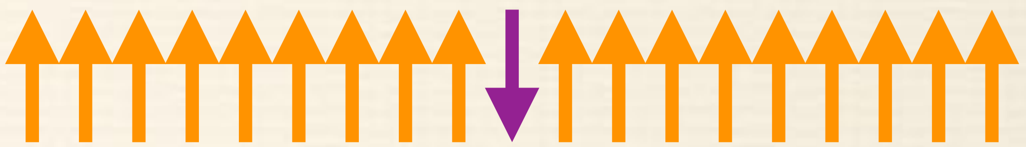
Deviation from ground state
Collective mode : S=1



Spin Wave - Magnon

Deviation from ground state

Collective mode : S=1



Spin Wave - Magnon

Deviation from ground state
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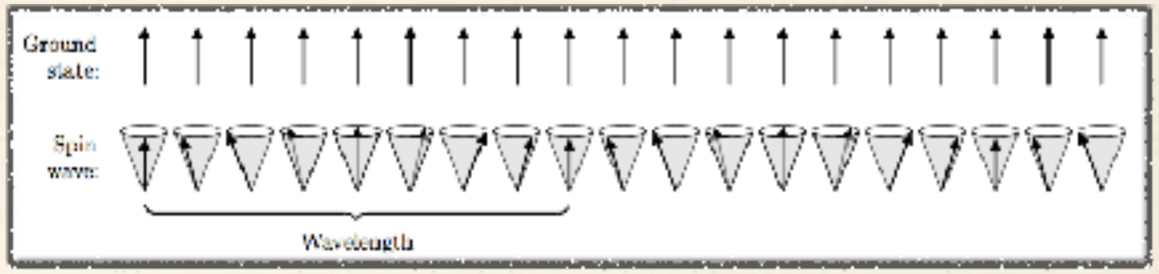
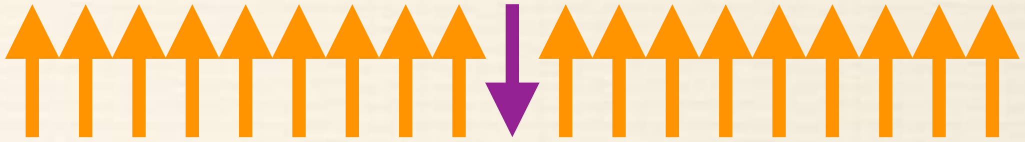
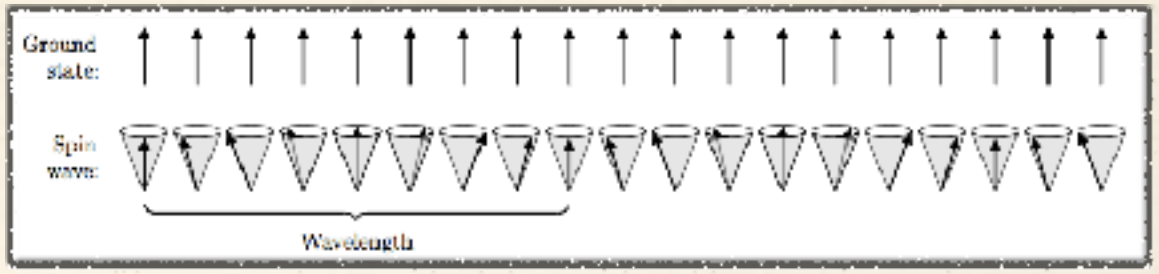
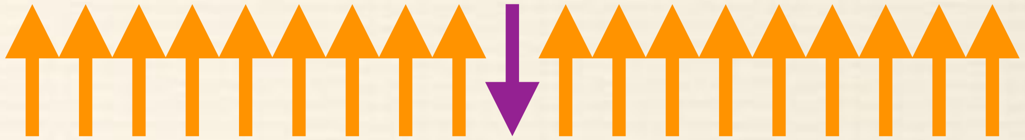


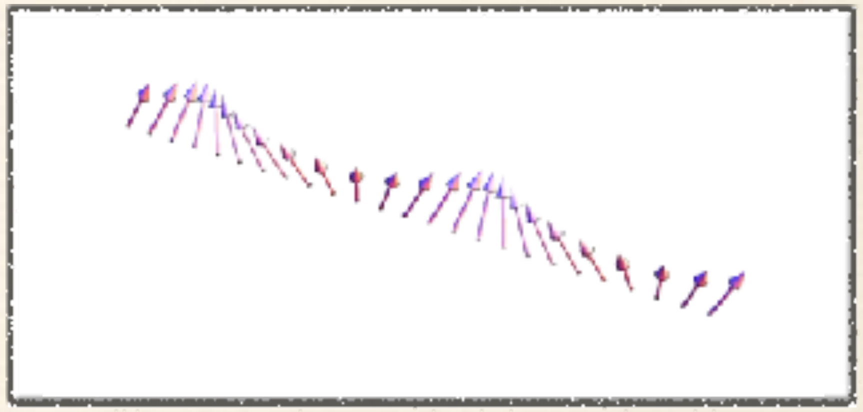


Spin Wave - Magnon

Deviation from ground state
Collective mode : S=1



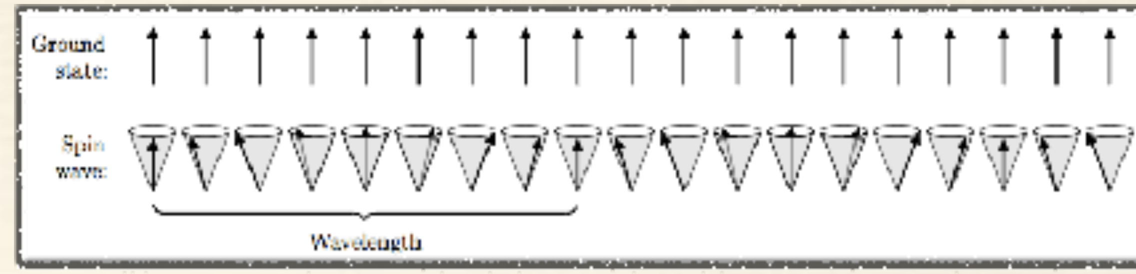


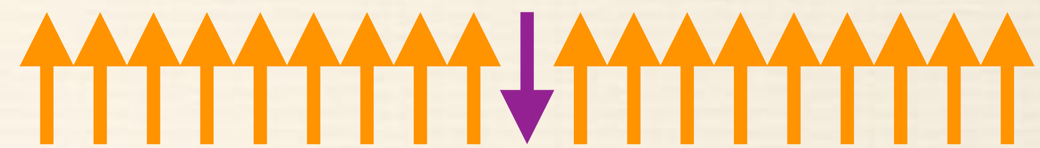


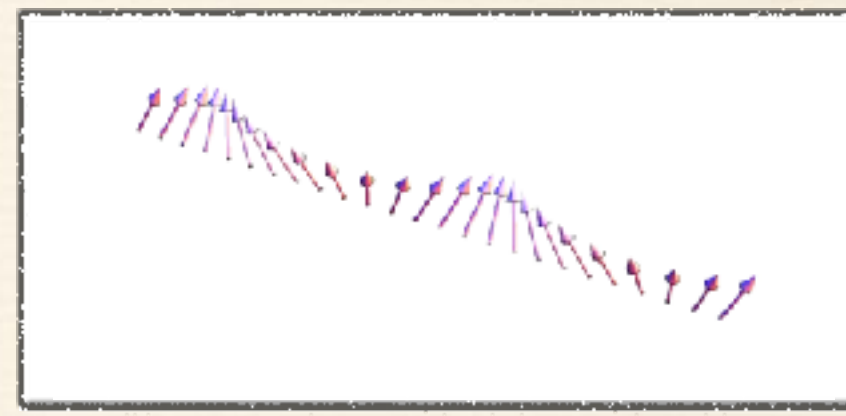
Spin Wave - Magnon

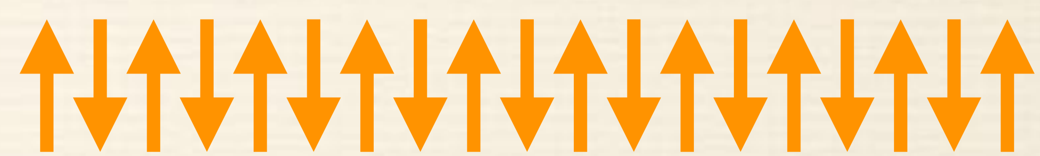
Deviation from ground state

Collective mode : S=1



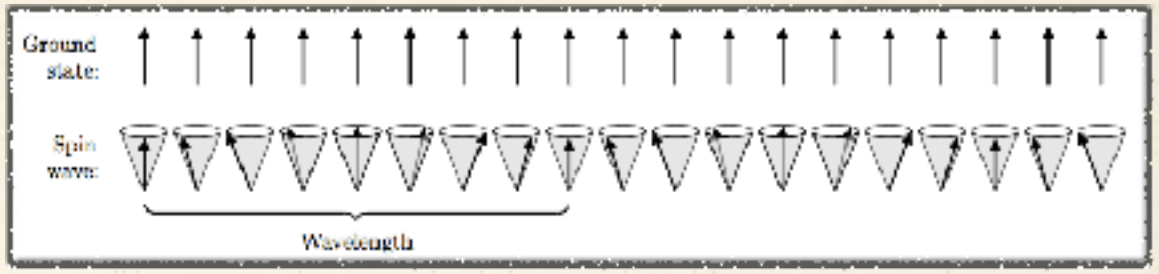


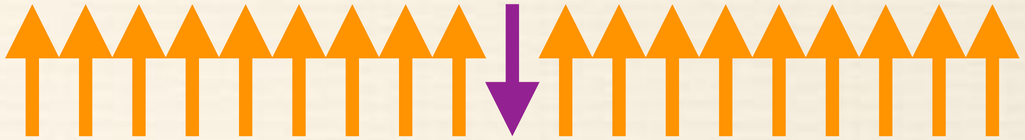


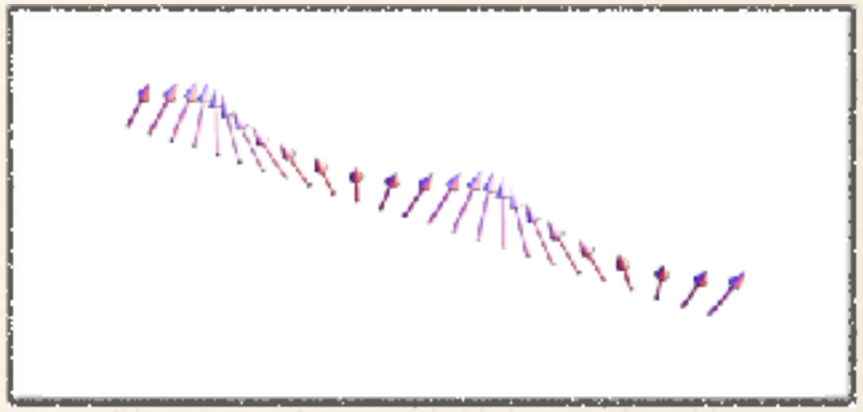


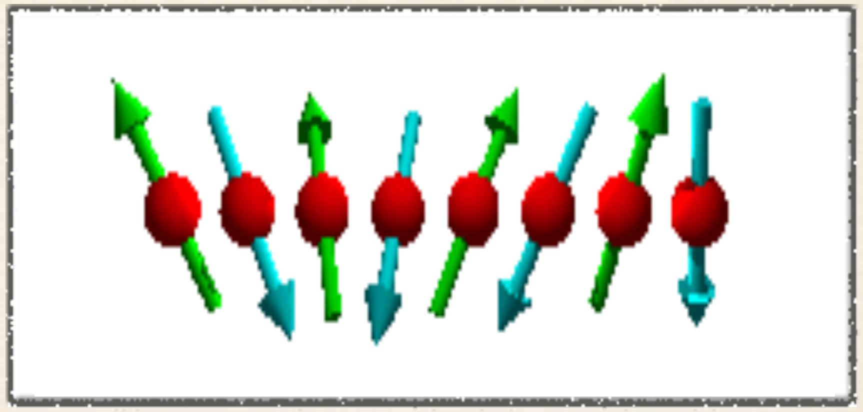
Spin Wave - Magnon

Deviation from ground state
Collective mode : S=1









Spin Wave - Magnon

Deviation from ground state

Collective mode : S=1

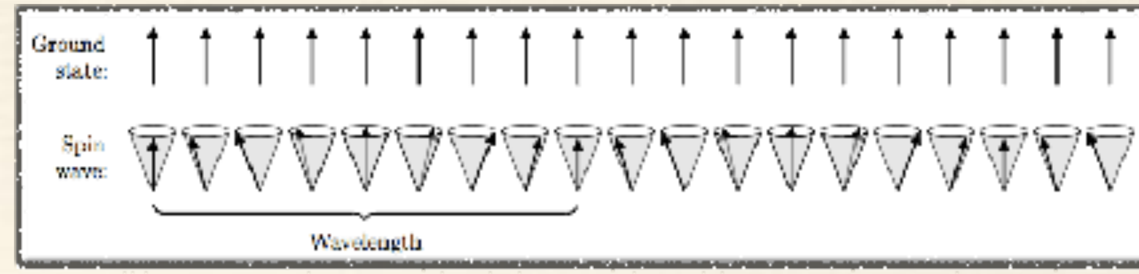
J-dependent dispersion

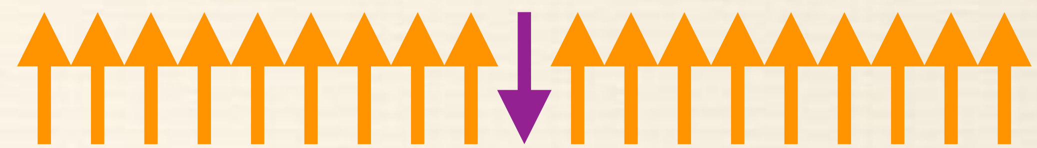
Ferromagnetic :

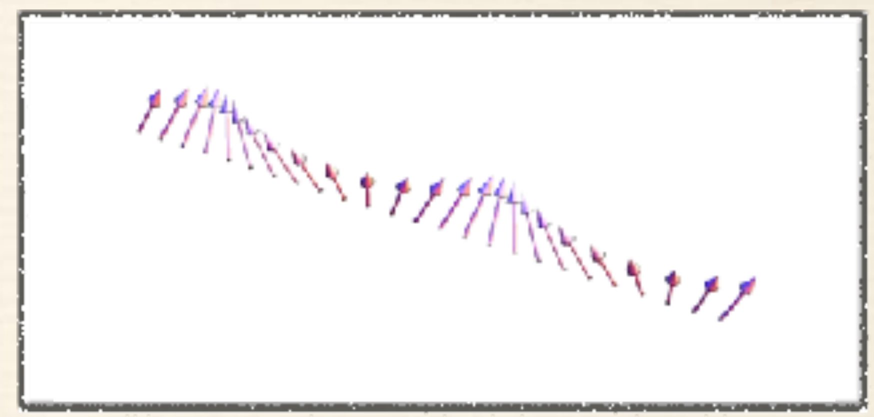
$$\hbar\omega = 2J\langle S\rangle(1 - cos(qa))$$

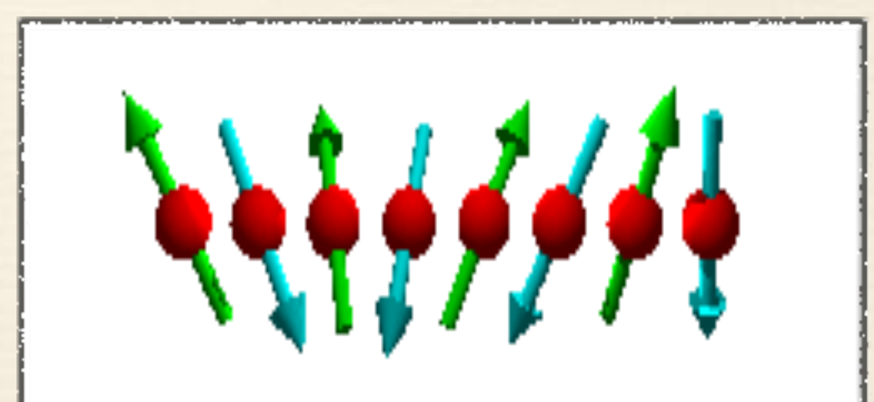
Antiferromagnetic :

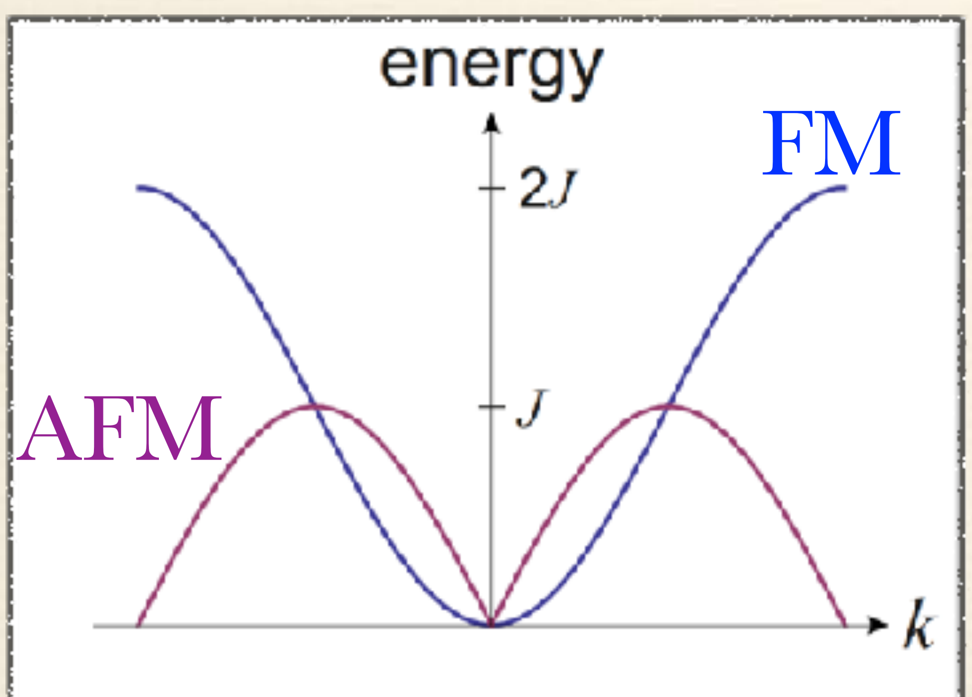
$$\hbar\omega = 2|J|\langle S\rangle|sin(qa)|$$





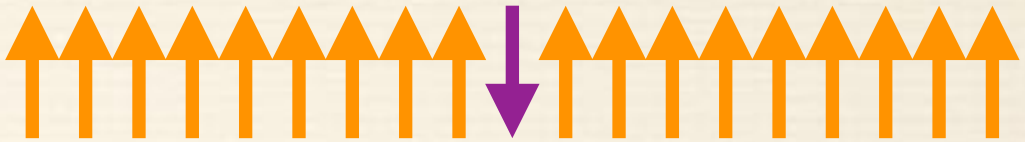






(one, two or bi)magnons

Single Magnon :
delocalized Spin-Flip

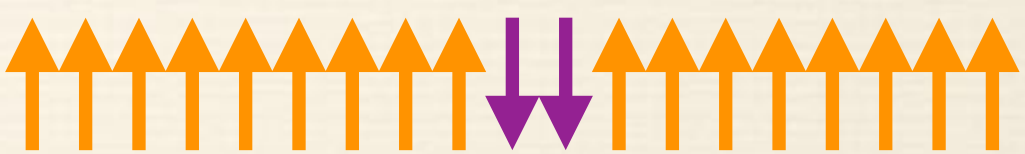


$\Delta E = 2J \qquad \Delta S = 1$

Two Magnons :
2 independent Spin-Flip delocalized

$\Delta E = 4J \qquad \Delta S = 2$

Bi-magnon :
2 neighbors Spin-Flip delocalized



$\Delta E = 2J \qquad \Delta S = 2$

Spinon

Deconfined half-magnon

Spinon

Deconfined half-magnon

Spinon

Deconfined half-magnon

Spinon

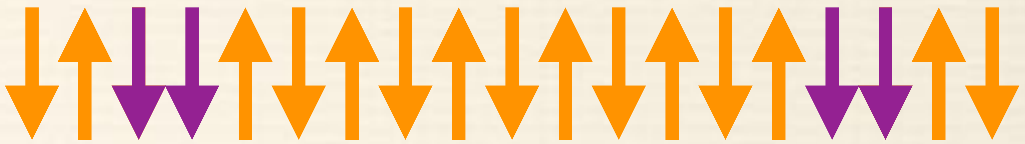
Deconfined half-magnon

Spinon

Deconfined half-magnon

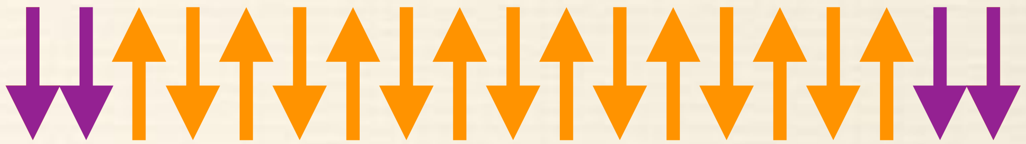
Spinon

Deconfined half-magnon



Spinon

Deconfined half-magnon

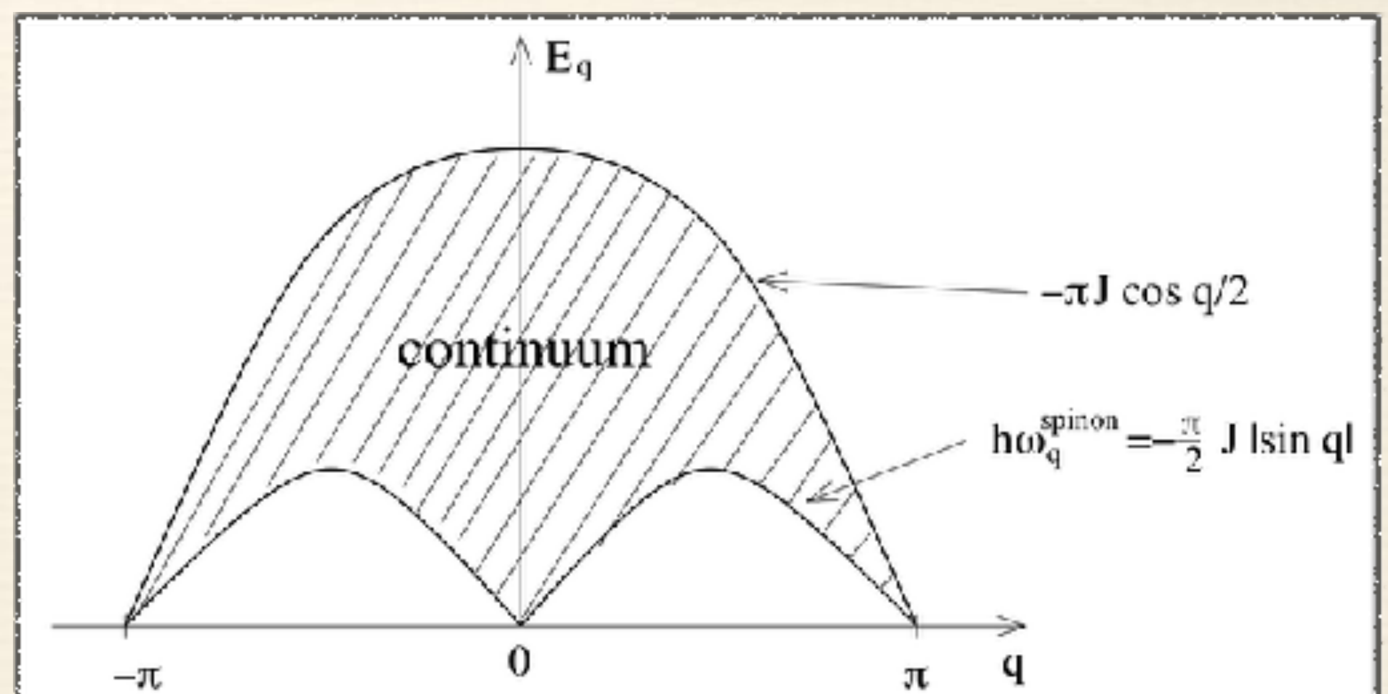


Spinon

Deconfined half-magnon
S=1/2
Collective mode
Only at 1d :
  d>1 : E~chain length

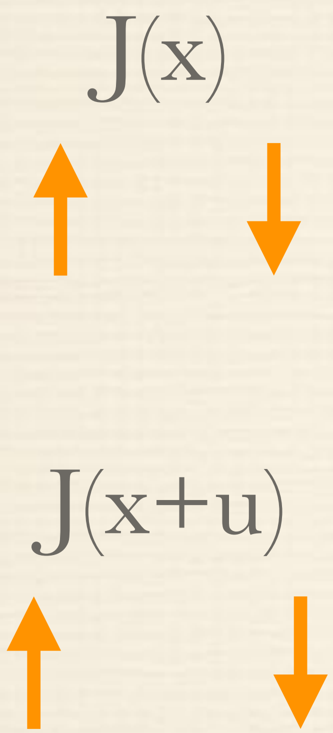
1 spinon dispersion:

$$\hbar\omega = -\frac{\pi}{2} J \, |sin(qa)|$$



Mixed excitations

Spin-phonon coupling



$$E_{magnon} = -\sum_{i,j} J_{0,ij} \langle \vec{S}_i \vec{S}_j \rangle$$

$$E_{phonon} = \frac{1}{2}\omega_0^2 u^2$$

$$J(u) = J_0 + \frac{1}{2}\frac{\partial^2 J_{ij}}{\partial^2 u}\langle \vec{S}_i \vec{S}_j \rangle u^2$$

$$\omega' = \omega_0 + \frac{\partial^2 J_{ij}}{\partial^2 u}\langle \vec{S}_i \vec{S}_j \rangle$$

Coupling parameter $\frac{\partial^2 J_{ij}}{\partial^2 u} \sim$ meV

Possibility to lift degeneracy of multiple mode (T —> E+A)

2.2 Inelastic Scattering

Inelastic Neutron Scattering

Interaction neutron-magnetic moment

Magnetic interaction

Spin contribution

$$\mathcal{V}(\vec{r}) = -\vec{\mu}_n . \frac{\mu_0}{4\pi}\left(r\vec{o}t\left(\frac{\vec{\mu_e} \wedge \vec{r}}{r^3}\right) - \frac{2\mu_B}{\hbar}\frac{\vec{p} \wedge \vec{r}}{r^3}\right)$$

Orbital contribution

$\vec{\mu_e} = 2\mu_B \vec{S}$: electron magnetic moment operator

$\vec{\mu_n} = -\gamma \mu_N \vec{\sigma}$: neutron magnetic moment operator

$$\mathcal{V}(\vec{r}) = -\vec{\mu}_n . r\vec{o}t(r\vec{o}t\left(\frac{\mu_e^S + \mu_e^L}{r}\right))$$

Inelastic Neutron Scattering

Interaction neutron-magnetic moment

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{c_1,c_2} e^{i\vec{Q}.(\vec{R}_{c_1}-\vec{R}_{c_2})} \sum_{a_1,a_2} f_{a_1}(\vec{Q}) f_{a_2}(\vec{Q}) e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}}$$

$$\times \int_{-\infty}^{+\infty} \langle \vec{S}_{\perp,c1,a1}(0).\vec{S}_{\perp,c2,a2}(t)\rangle e^{-i\omega t} dt$$

$$= \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G}\in RS} \delta(\vec{Q}-\vec{G}) \left| \sum_a r_e f_a(\vec{Q}) S_{\perp,a} e^{-W_a} e^{i\vec{Q}.\vec{r}_a} \right|^2 \delta(\omega)$$

$$+ \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G},\vec{q}} \delta(\vec{Q}-\vec{q}-\vec{G}) \sum_{\alpha,\beta} \left(\delta_{\alpha\beta} - \frac{Q^\alpha Q^\beta}{Q^2}\right)$$

$$\times \sum_{a_1,a_2} f_{a_1}(\vec{Q}) f_{a_2}(\vec{Q}) e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}} \int_{-\infty}^{+\infty} \langle S^\alpha_{a1}(0) S^\beta_{a2}(t)\rangle e^{-i\omega t} dt$$

Pair correlation function S(Q,ω)

Inelastic Neutron Scattering

Interaction neutron-magnetic moment

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{c_1,c_2} e^{i\vec{Q}.(\vec{R}_{c_1}-\vec{R}_{c_2})} \sum_{a_1,a_2} f_{a_1}(\vec{Q}) f_{a_2}(\vec{Q}) e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}}$$

$$\times \int_{-\infty}^{+\infty} \boxed{\langle \vec{S}_{\perp,c1,a1}(0).\vec{S}_{\perp,c2,a2}(t)\rangle} e^{-i\omega t} dt$$

$$= \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G}\in RS} \delta(\vec{Q}-\vec{G}) \left| \sum_a r_e f_a(\vec{Q}) S_{\perp,a} e^{-W_a} e^{i\vec{Q}.\vec{r}_a} \right|^2 \delta(\omega)$$

$$+ \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G},\vec{q}} \delta(\vec{Q}-\vec{q}-\vec{G}) \sum_{\alpha,\beta} \left(\delta_{\alpha\beta} - \frac{Q^\alpha Q^\beta}{Q^2} \right)$$

$$\times \sum_{a_1,a_2} f_{a_1}(\vec{Q}) f_{a_2}(\vec{Q}) e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}} \int_{-\infty}^{+\infty} \langle S^\alpha_{a1}(0) S^\beta_{a2}(t)\rangle e^{-i\omega t} dt$$

Contribution from components perpendicular to Q

Inelastic Neutron Scattering

Interaction neutron-magnetic moment

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{c_1,c_2} e^{i\vec{Q}.(\vec{R}_{c_1}-\vec{R}_{c_2})} \sum_{a_1,a_2} f_{a_1}(\vec{Q}) f_{a_2}(\vec{Q}) e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}}$$

$$\times \int_{-\infty}^{+\infty} \langle \vec{S}_{\perp,c1,a1}(0).\vec{S}_{\perp,c2,a2}(t)\rangle e^{-i\omega t} dt$$

$$= \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G}\in RS} \delta(\vec{Q}-\vec{G}) \left| \sum_a r_e f_a(\vec{Q}) S_{\perp,a} e^{-W_a} e^{i\vec{Q}.\vec{r}_a} \right|^2 \delta(\omega)$$

$$+ \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G},\vec{q}} \delta(\vec{Q}-\vec{q}-\vec{G}) \sum_{\alpha,\beta} \left(\delta_{\alpha\beta} - \frac{Q^\alpha Q^\beta}{Q^2} \right)$$

$$\times \sum_{a_1,a_2} f_{a_1}(\vec{Q}) f_{a_2}(\vec{Q}) e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}} \int_{-\infty}^{+\infty} \langle S^\alpha_{a1}(0) S^\beta_{a2}(t)\rangle e^{-i\omega t} dt$$

Elastic scattering

Inelastic Neutron Scattering

Interaction neutron-magnetic moment

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{c_1,c_2} e^{i\vec{Q}.(\vec{R}_{c_1}-\vec{R}_{c_2})} \sum_{a_1,a_2} f_{a_1}(\vec{Q}) f_{a_2}(\vec{Q}) e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}}$$

$$\times \int_{-\infty}^{+\infty} \langle \vec{S}_{\perp,c1,a1}(0).\vec{S}_{\perp,c2,a2}(t)\rangle e^{-i\omega t} dt$$

$$= \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G}\in RS} \delta(\vec{Q}-\vec{G}) \left| \sum_a r_e f_a(\vec{Q}) S_{\perp,a} e^{-W_a} e^{i\vec{Q}.\vec{r}_a} \right|^2 \delta(\omega)$$

$$+ \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G},\vec{q}} \delta(\vec{Q}-\vec{q}-\vec{G}) \sum_{\alpha,\beta} \left(\delta_{\alpha\beta} - \frac{Q^\alpha Q^\beta}{Q^2} \right)$$

$$\times \sum_{a_1,a_2} f_{a_1}(\vec{Q}) f_{a_2}(\vec{Q}) e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}} \int_{-\infty}^{+\infty} \langle S^\alpha_{a1}(0) S^\beta_{a2}(t)\rangle e^{-i\omega t} dt$$

Inelastic scattering

Inelastic Neutron Scattering

Interaction neutron-magnetic moment

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{c_1,c_2} e^{i\vec{Q}.(\vec{R}_{c_1}-\vec{R}_{c_2})} \sum_{a_1,a_2} \boxed{f_{a_1}(\vec{Q})f_{a_2}(\vec{Q})} e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}}$$

$$\times \int_{-\infty}^{+\infty} \langle \vec{S}_{\perp,c1,a1}(0).\vec{S}_{\perp,c2,a2}(t)\rangle e^{-i\omega t} dt$$

$$= \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G}\in RS} \delta(\vec{Q}-\vec{G}) \left| \sum_a r_e \boxed{f_a(\vec{Q})} S_{\perp,a} e^{-W_a} e^{i\vec{Q}.\vec{r}_a} \right|^2 \delta(\omega)$$

$$+ \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G},\vec{q}} \delta(\vec{Q}-\vec{q}-\vec{G}) \sum_{\alpha,\beta} \left(\delta_{\alpha\beta} - \frac{Q^\alpha Q^\beta}{Q^2}\right)$$

$$\times \sum_{a_1,a_2} \boxed{f_{a_1}(\vec{Q})f_{a_2}(\vec{Q})} e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}} \int_{-\infty}^{+\infty} \langle S^\alpha_{a1}(0) S^\beta_{a2}(t)\rangle e^{-i\omega t} dt$$

Magnetic form factor

Inelastic Neutron Scattering

Interaction neutron-magnetic moment

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{c_1,c_2} e^{i\vec{Q}.(\vec{R}_{c_1}-\vec{R}_{c_2})} \sum_{a_1,a_2} f_{a_1}(\vec{Q}) f_{a_2}(\vec{Q}) e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}}$$

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$$= \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G}\in RS} \delta(\vec{Q}-\vec{G}) \left| \sum_a r_e f_a(\vec{Q}) S_{\perp,a} e^{-W_a} e^{i\vec{Q}.\vec{r}_a} \right|^2 \delta(\omega)$$

$$+ \frac{k_f}{k_i}(\gamma r_e)^2 \sum_{\vec{G},\vec{q}} \delta(\vec{Q}-\vec{q}-\vec{G}) \sum_{\alpha,\beta} \left(\delta_{\alpha\beta} - \frac{Q^\alpha Q^\beta}{Q^2}\right)$$

$$\times \sum_{a_1,a_2} f_{a_1}(\vec{Q}) f_{a_2}(\vec{Q}) e^{i\vec{Q}.(\vec{r}_{a_1}-\vec{r}_{a_2})} e^{-W_{a1}-W_{a2}} \int_{-\infty}^{+\infty} \langle S^\alpha_{a1}(0) S^\beta_{a2}(t)\rangle e^{-i\omega t} dt$$

Geometric factor

Inelastic Neutron Scattering

Magnon in ferromagnet

$$\frac{\partial^2 \sigma}{\partial \Omega \partial \omega} = \frac{k_f}{k_i}(\gamma r_0)^2 S^2 \left(1 - \left(\frac{Q^z}{Q}\right)^2\right) \left| f(\vec{Q}) e^{-W} \right|^2 \sum_{\vec{G} \in RS} \delta(\vec{Q} - \vec{G}) \delta(\omega)$$

$$+ \frac{k_f}{k_i}(\gamma r_0)^2 S \left(1 + \left(\frac{Q^z}{Q}\right)^2\right) \left| f(\vec{Q}) e^{-W} \right|^2 \sum_{\vec{G}, \vec{q}} \delta(\vec{Q} - \vec{q} - \vec{G})$$

$$\times \left[(1 + n_B(\omega_p(\vec{q}), T)) \delta(\omega - \omega_p(\vec{q})) + n_B(\omega_p(\vec{q}), T) \delta(\omega + \omega_p(\vec{q}))\right]$$

Elastic intensity when Q perpendicular to S

Inelastic Neutron Scattering

Magnon in ferromagnet

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = \frac{k_f}{k_i}(\gamma r_0)^2 S^2 \left(1 - \left(\frac{Q^z}{Q}\right)^2\right) \left|f(\vec{Q})e^{-W}\right|^2 \sum_{\vec{G}\in RS} \delta(\vec{Q}-\vec{G})\delta(\omega)$$

$$+ \frac{k_f}{k_i}(\gamma r_0)^2 S \left(1 + \left(\frac{Q^z}{Q}\right)^2\right) \left|f(\vec{Q})e^{-W}\right|^2 \sum_{\vec{G},\vec{q}} \delta(\vec{Q}-\vec{q}-\vec{G})$$

$$\times \left[(1+n_B(\omega_p(\vec{q}),T))\delta(\omega-\omega_p(\vec{q})) + n_B(\omega_p(\vec{q}),T)\delta(\omega+\omega_p(\vec{q}))\right]$$

Linerarity in <S>

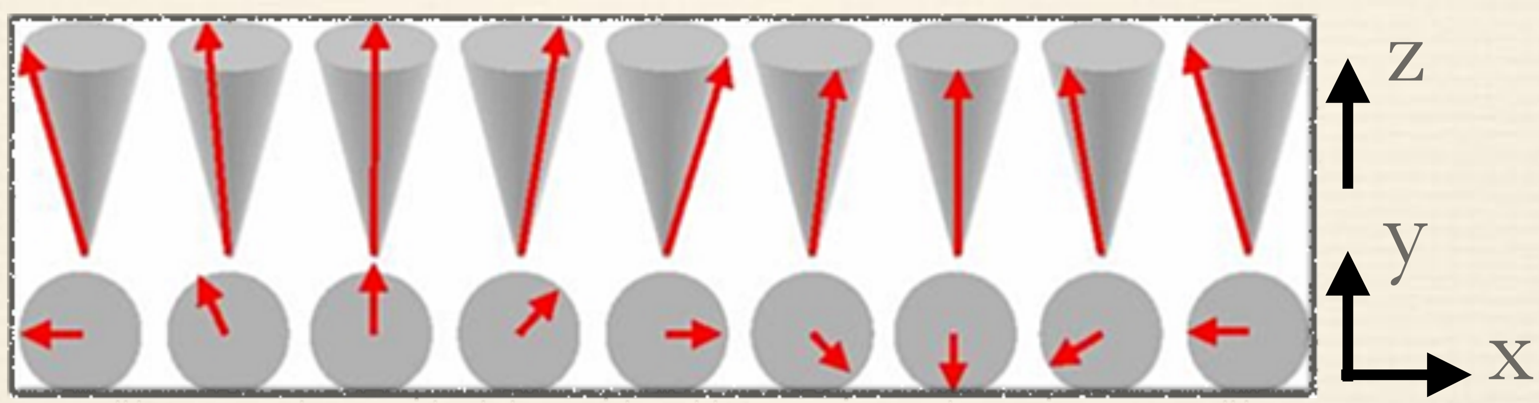
Inelastic Neutron Scattering

Magnon in ferromagnet

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = \frac{k_f}{k_i}(\gamma r_0)^2 S^2 \left(1 - \left(\frac{Q^z}{Q}\right)^2\right) \left|f(\vec{Q})e^{-W}\right|^2 \sum_{\vec{G}\in RS} \delta(\vec{Q}-\vec{G})\delta(\omega)$$

$$+ \frac{k_f}{k_i}(\gamma r_0)^2 S \left(1 + \left(\frac{Q^z}{Q}\right)^2\right) \left|f(\vec{Q})e^{-W}\right|^2 \sum_{\vec{G},\vec{q}} \delta(\vec{Q}-\vec{q}-\vec{G})$$

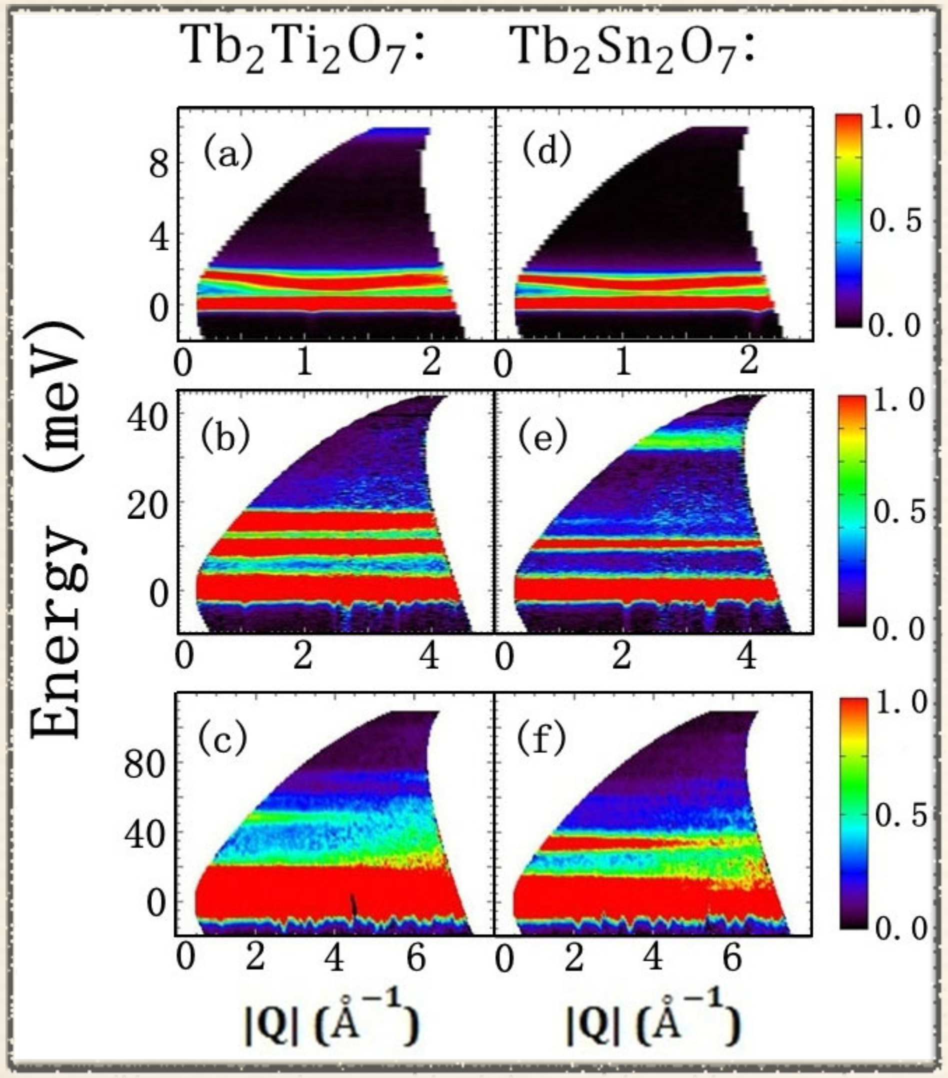
$$\times \left[(1+n_B(\omega_p(\vec{q}),T))\delta(\omega-\omega_p(\vec{q})) + n_B(\omega_p(\vec{q}),T)\delta(\omega+\omega_p(\vec{q}))\right]$$



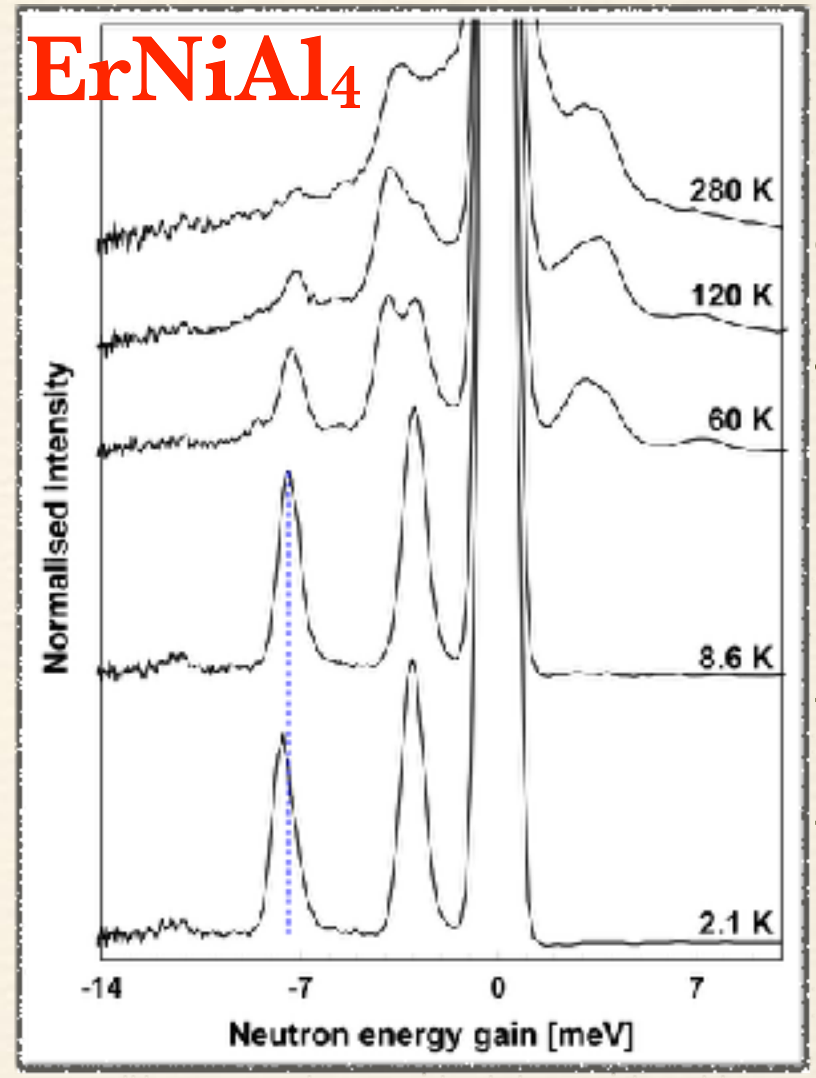
Inelastic intensity when Q parallel to S

Inelastic Neutron Scattering

Crystal field



PRB 89, 134410 (2014)



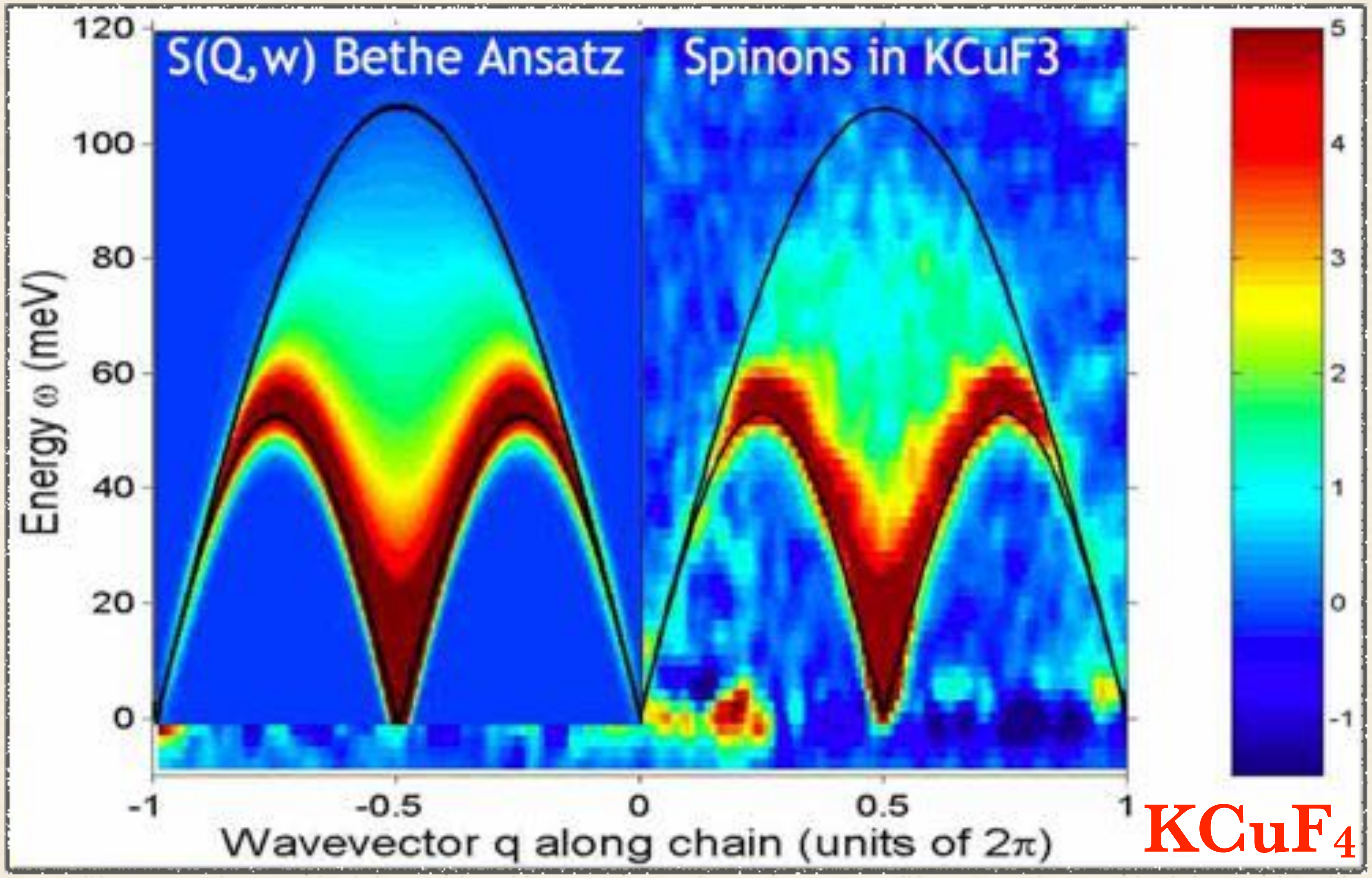
J. Phys. C 16, 841 (1983)

Dispersive Crystal Field excitations : entanglement of crystal field wave functions

Temperature dependence : thermodynamic (detailed balance factor)

Inelastic Neutron Scattering

Spinon



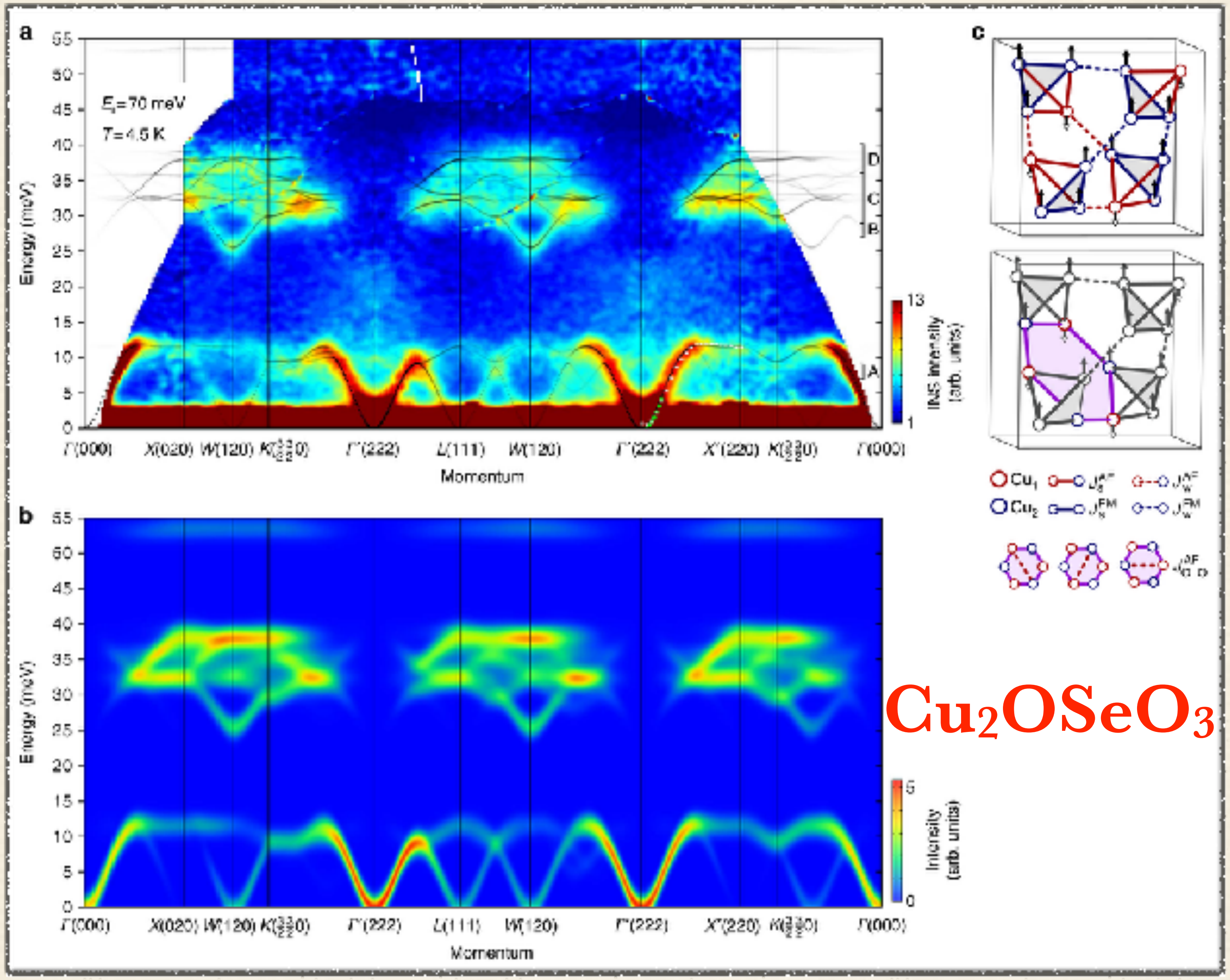
$KCuF_4$

PRL 70, 4003 (1993)

Unbound pair excitation of spinons

Inelastic Neutron Scattering

Magnon



Cu_2OSeO_3

Nature Com. 7, 10725 (2016)

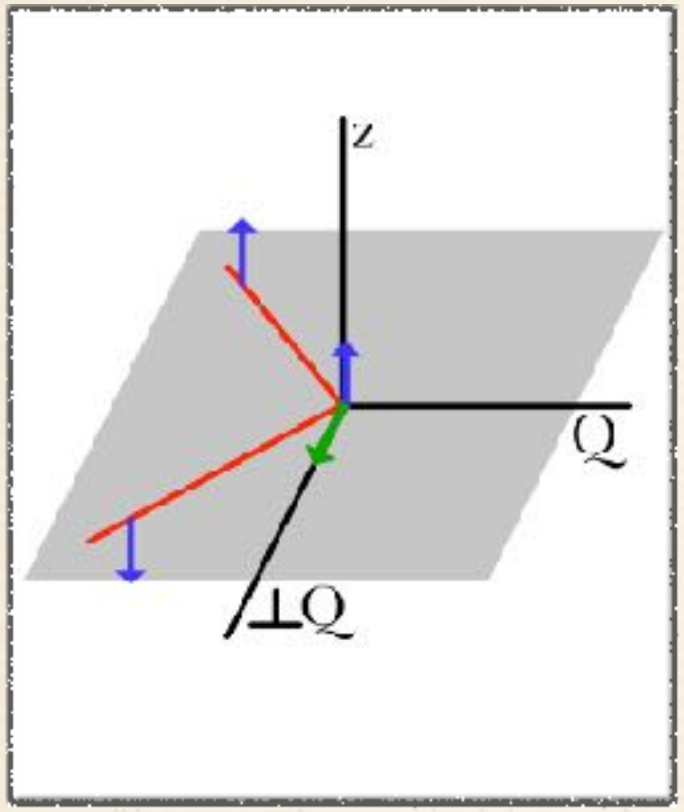
Inelastic Neutron Scattering

Difference phonon-magnon

Magnon	Phonon
Intensity decreases with **Q** (magnetic form factor)	Intensity proportional to $\mathbf{Q}^2$
Appears in ordered phase $T<T_C$	Always present

Inelastic Neutron Scattering

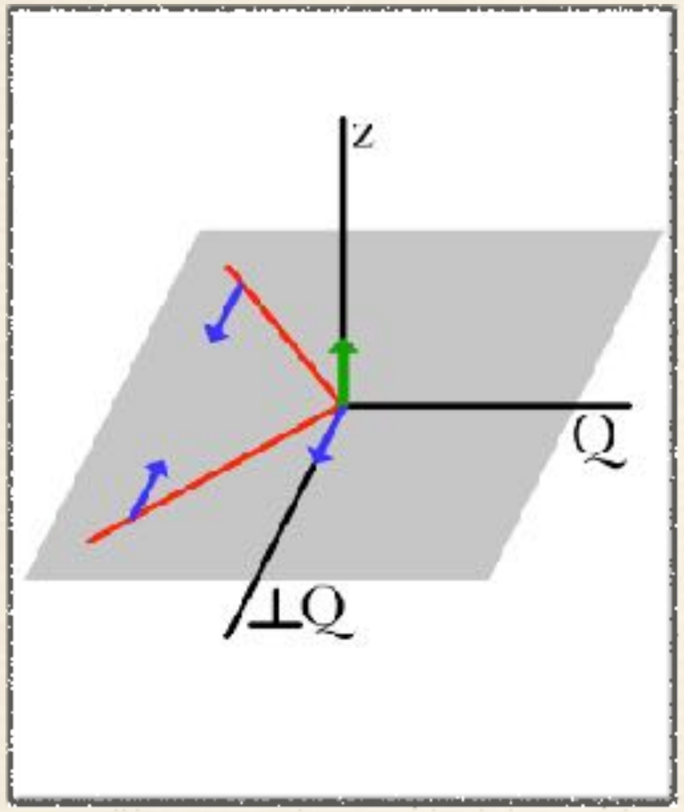
Polarized neutron diffraction



$$I_{SF}^{z} \propto \left|\vec{M}_y\right|^2$$

Inelastic Neutron Scattering

Polarized neutron diffraction



$$I_{SF}^{z} \propto \left| \vec{M}_y \right|^2$$

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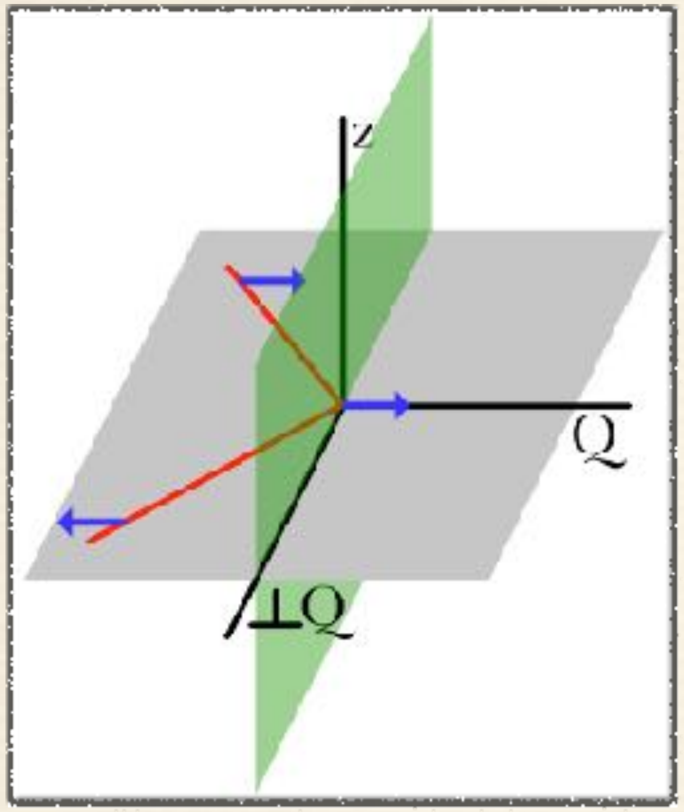
Inelastic Neutron Scattering

Polarized neutron diffraction



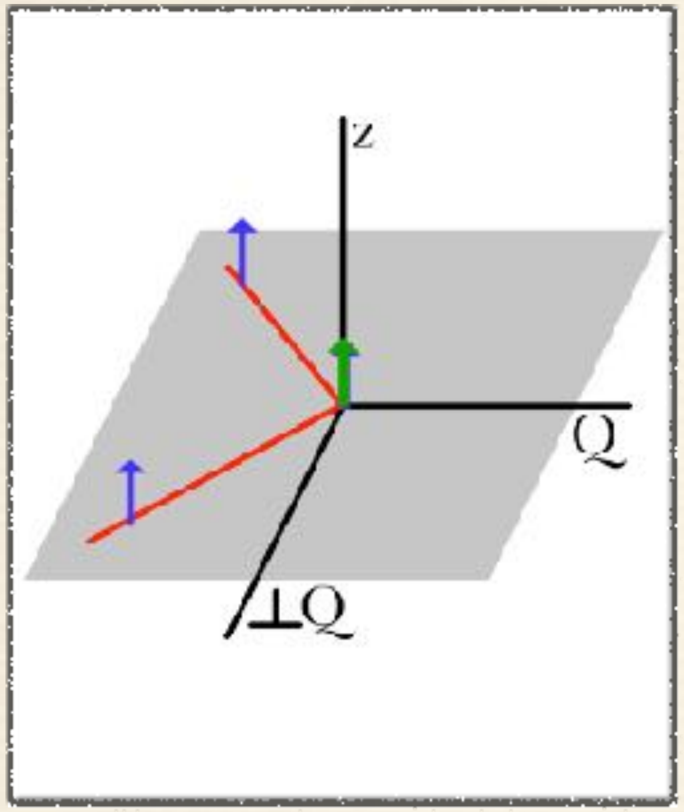
$$I_{SF}^{z} \propto \left|\vec{M}_y\right|^2$$

$$I_{SF}^{y} \propto \left|\vec{M}_z\right|^2$$

$$I_{SF}^{x} \propto \left|\vec{M}_y + \vec{M}_z\right|^2$$

Inelastic Neutron Scattering

Polarized neutron diffraction



$$I^z_{SF} \propto \left|\vec{M}_y\right|^2$$

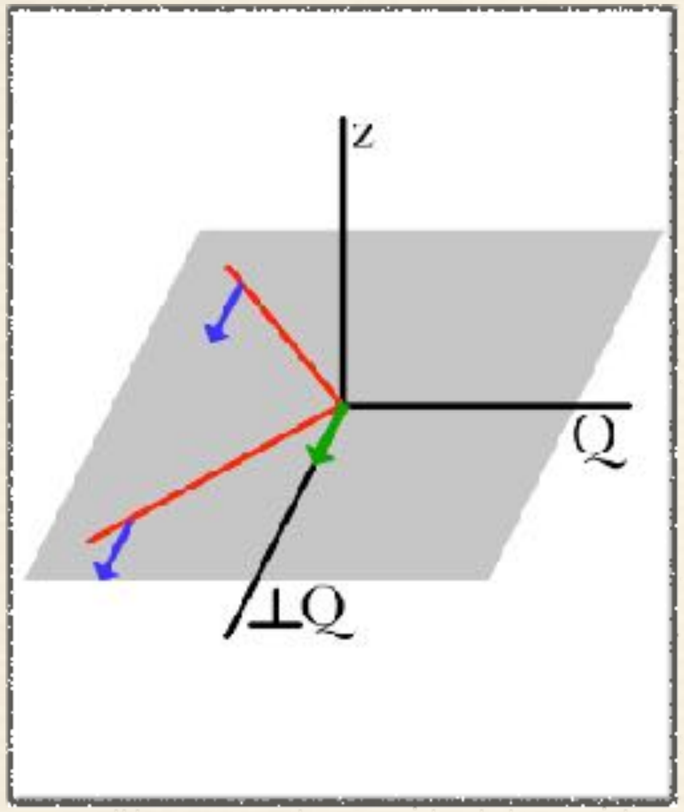
$$I^y_{SF} \propto \left|\vec{M}_z\right|^2$$

$$I^x_{SF} \propto \left|\vec{M}_y + \vec{M}_z\right|^2$$

$$I^z_{NSF} \propto N^2 + \left|\vec{M}_z\right|^2$$

Inelastic Neutron Scattering

Polarized neutron diffraction



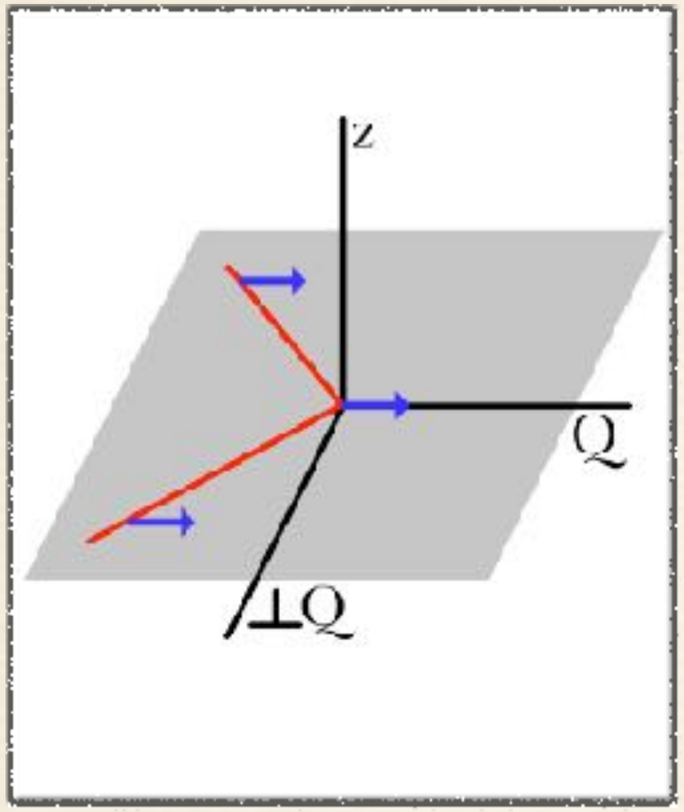
$$I_{SF}^{z} \propto \left|\vec{M}_y\right|^2 \qquad I_{NSF}^{z} \propto N^2 + \left|\vec{M}_z\right|^2$$

$$I_{SF}^{y} \propto \left|\vec{M}_z\right|^2 \qquad I_{NSF}^{y} \propto N^2 + \left|\vec{M}_y\right|^2$$

$$I_{SF}^{x} \propto \left|\vec{M}_y + \vec{M}_z\right|^2$$

Inelastic Neutron Scattering

Polarized neutron diffraction



$$I_{SF}^{z} \propto \left|\vec{M}_y\right|^2 \qquad I_{NSF}^{z} \propto N^2 + \left|\vec{M}_z\right|^2$$

$$I_{SF}^{y} \propto \left|\vec{M}_z\right|^2 \qquad I_{NSF}^{y} \propto N^2 + \left|\vec{M}_y\right|^2$$

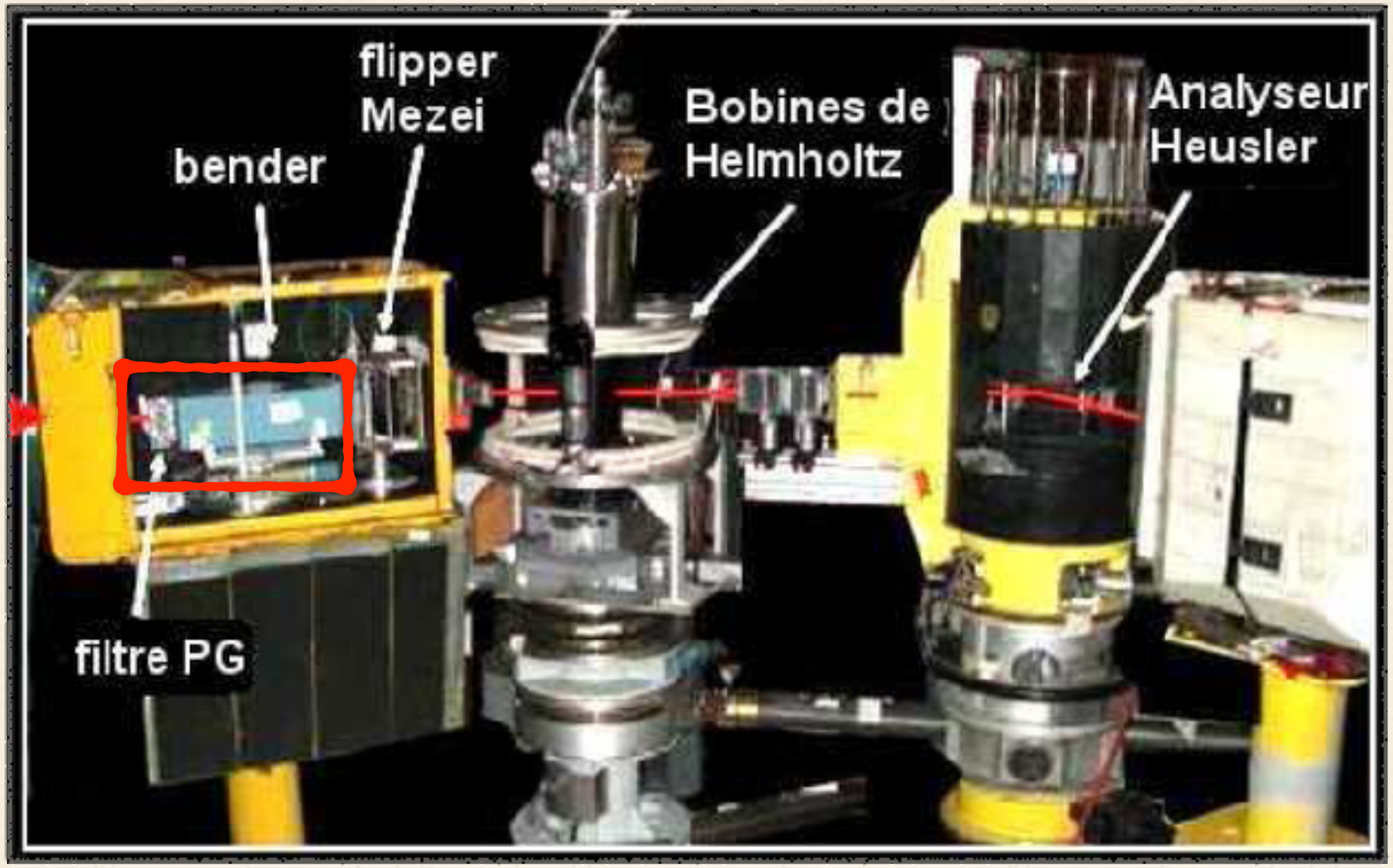
$$I_{SF}^{x} \propto \left|\vec{M}_y + \vec{M}_z\right|^2 \qquad I_{NSF}^{x} \propto N^2$$

Sum rule

$$I_{SF}^{x} = I_{SF}^{y} + I_{SF}^{z}$$

Inelastic Neutron Scattering

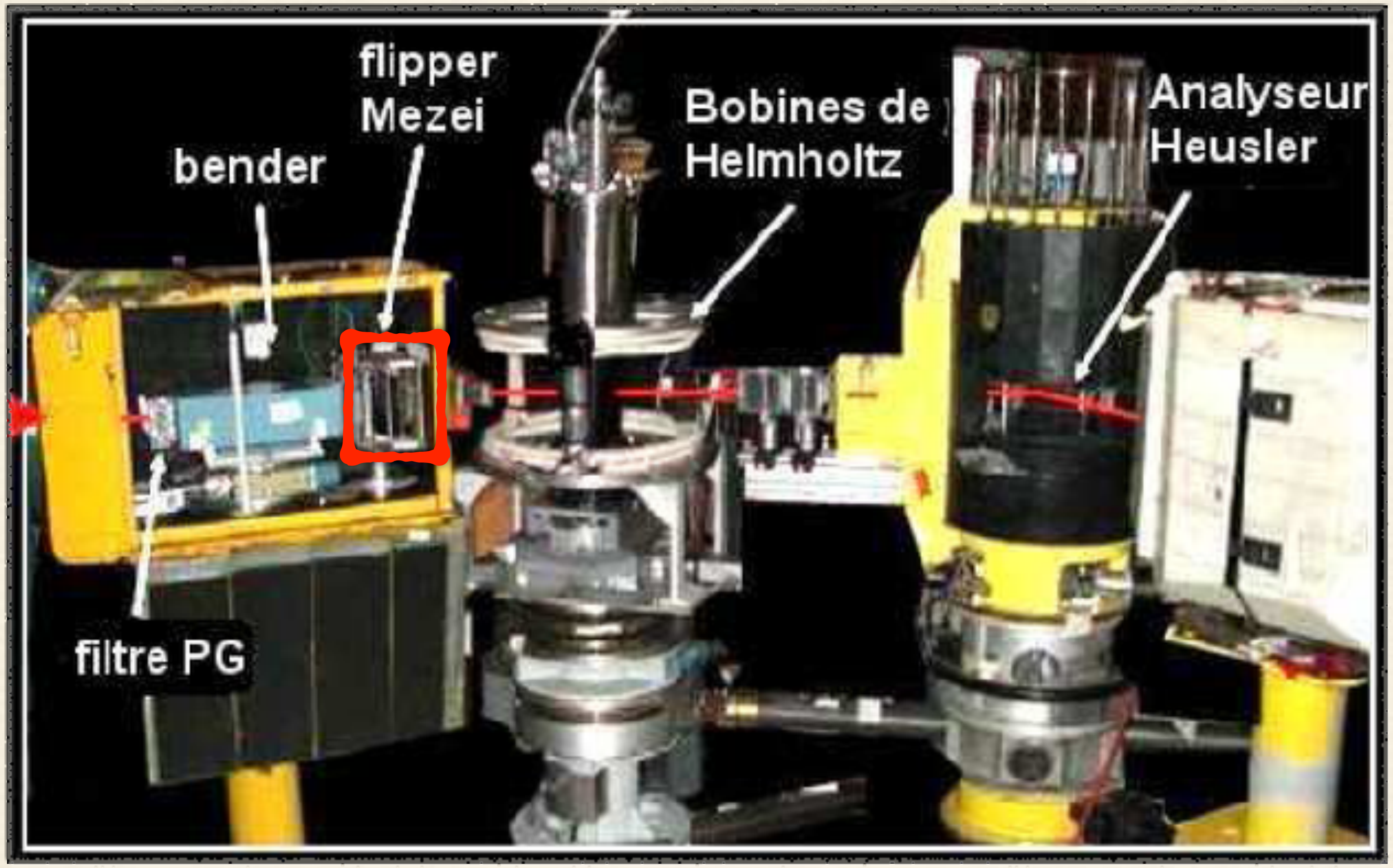
Polarized neutron experimental setup



Bender : polarize the beam up

Inelastic Neutron Scattering

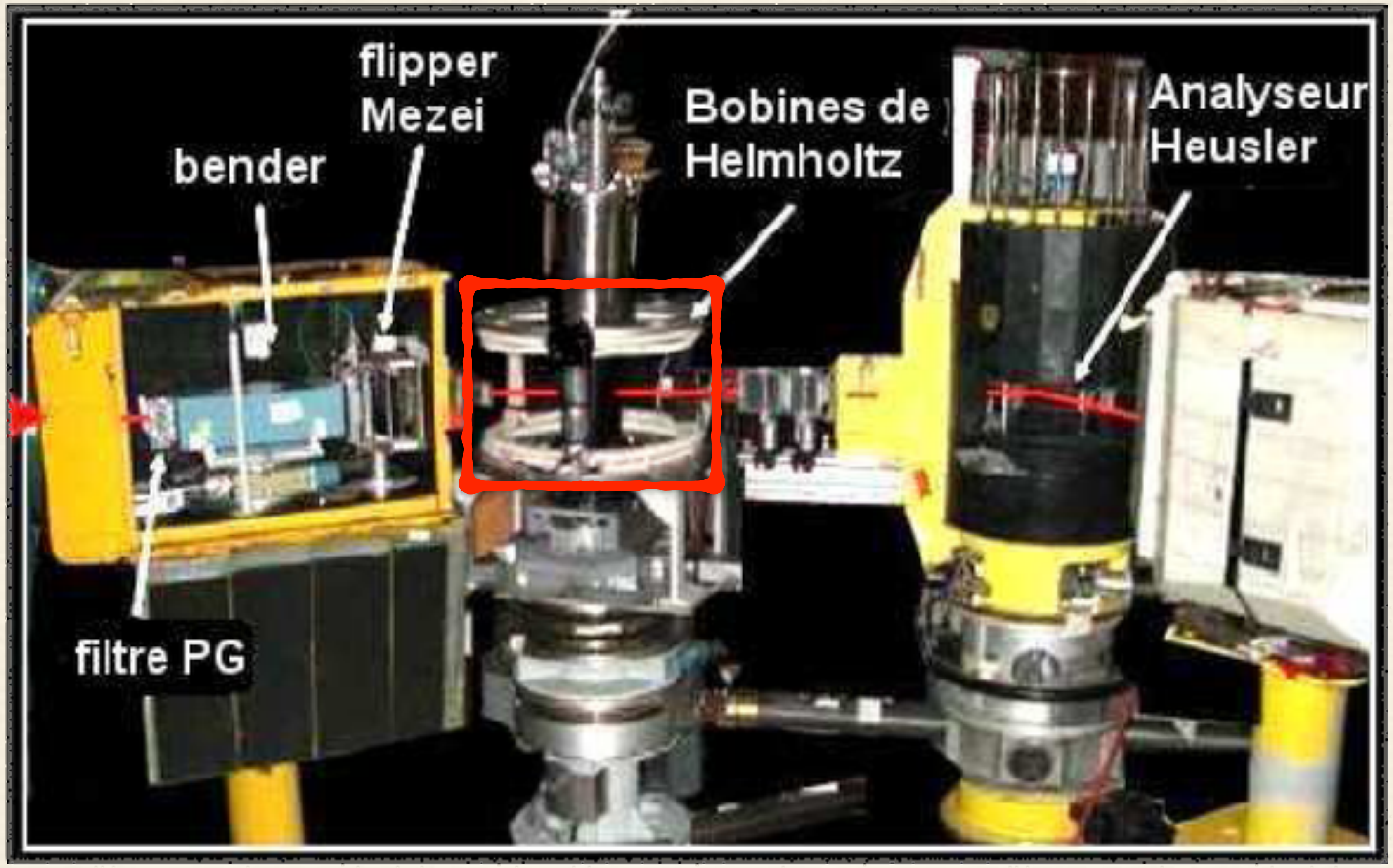
Polarized neutron experimental setup



Flipper : reverse the spin polarization

Inelastic Neutron Scattering

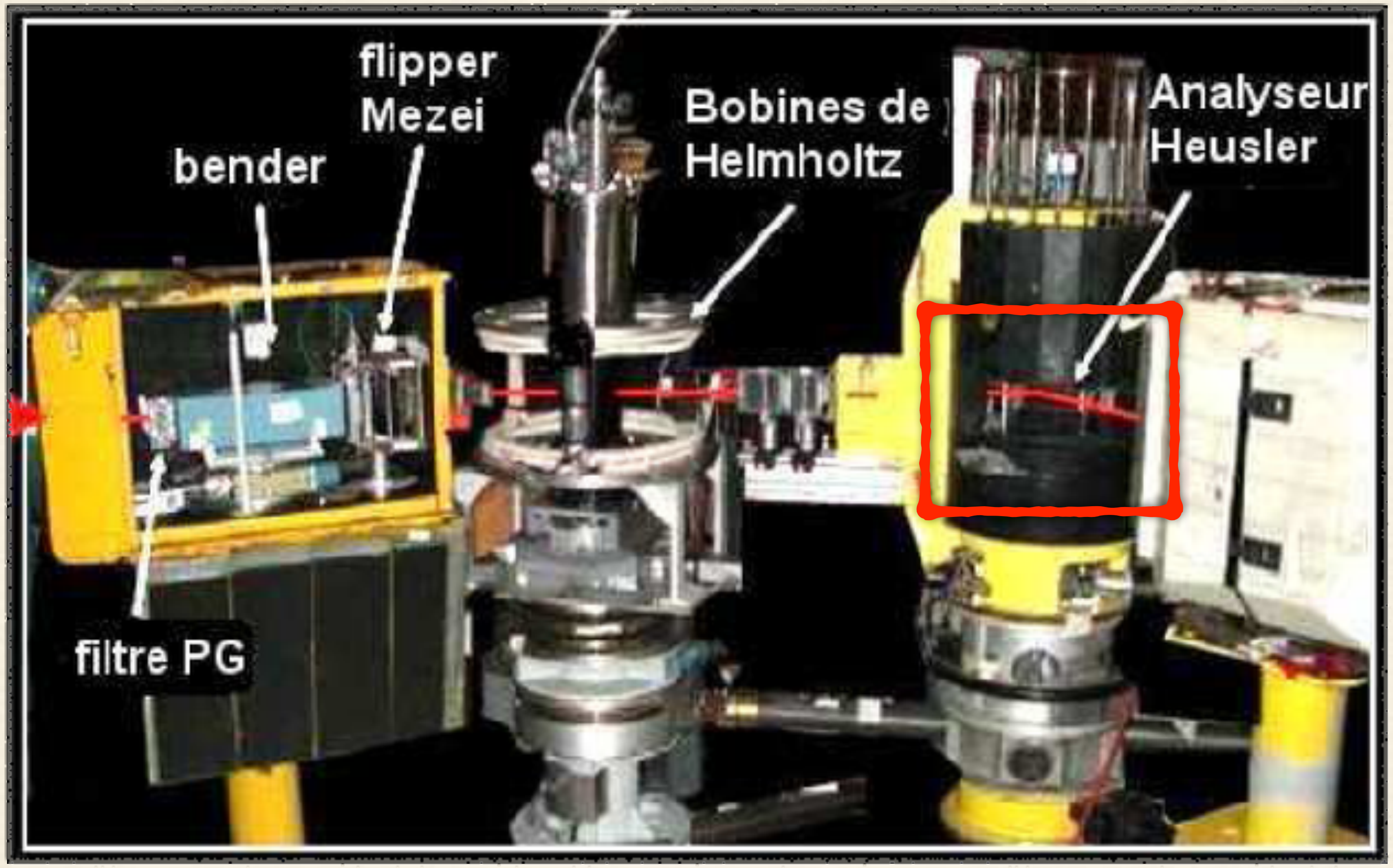
Polarized neutron experimental setup



Helmotz coils : manipulate spin orientation

Inelastic Neutron Scattering

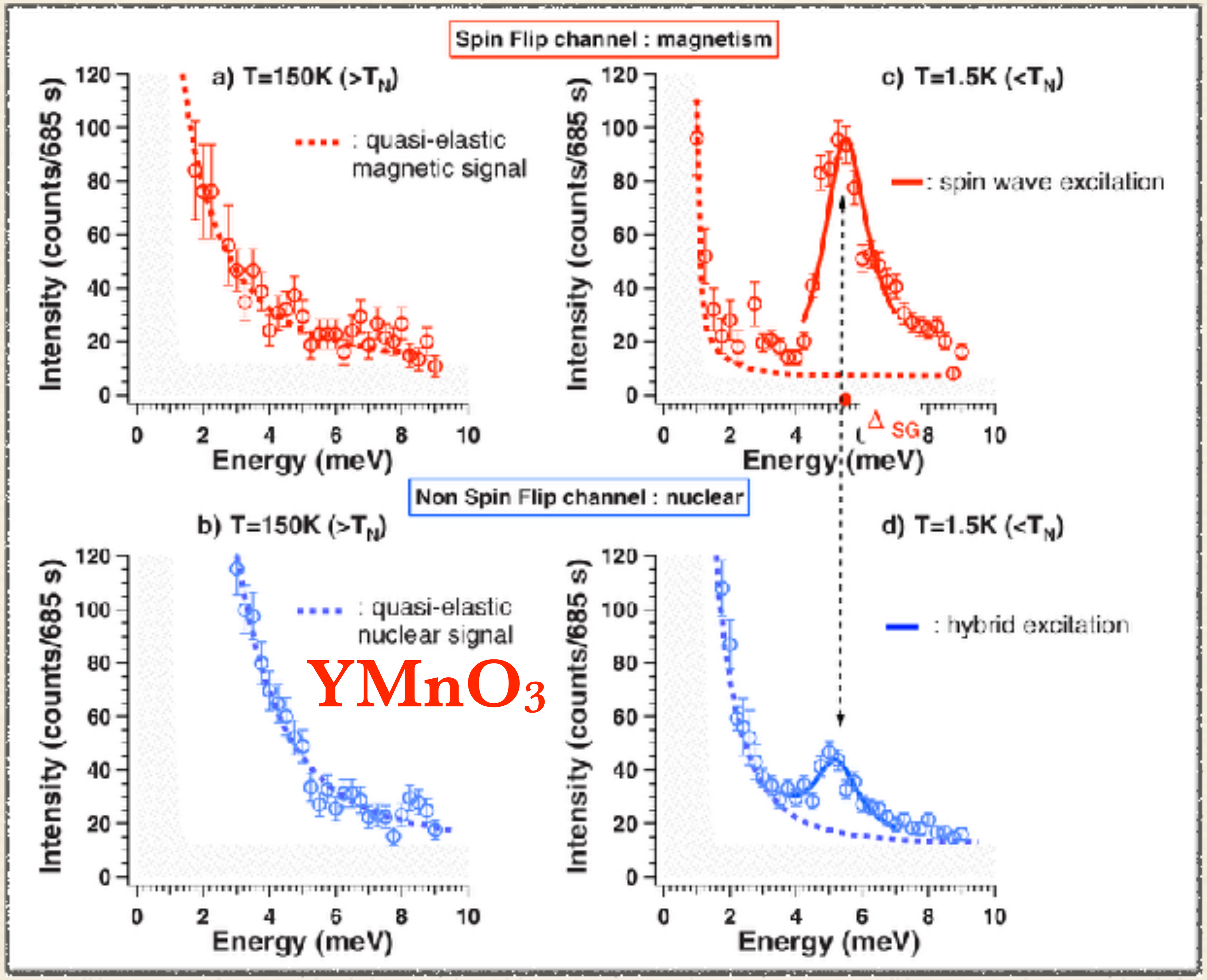
Polarized neutron experimental setup



Heusler analyzer : select energy and polarization

Inelastic Neutron Scattering

Electromagnon



YMnO$_3$

PRB 79, 134409 (2009)

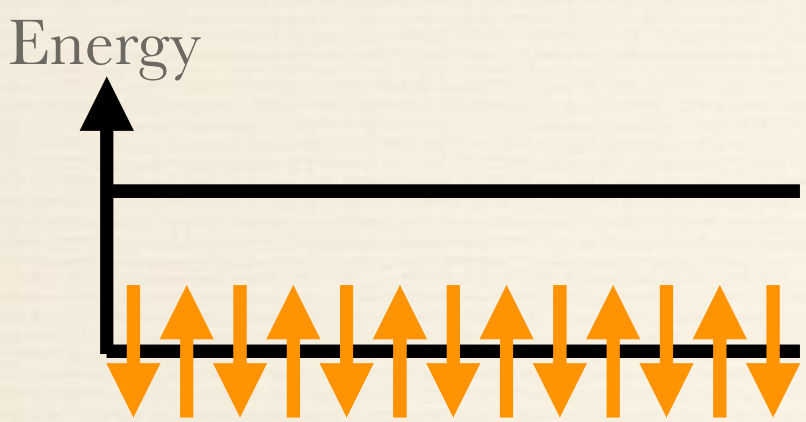
Inelastic Neutron Scattering

Summary

- Measure magnetic excitations dispersion (with restriction from kinematic limit)
- Intensity linearly proportional to the amplitude of the ordered moment (and not quadratic as elastic scattering))
- Polarized neutrons : separate phonons from magnons
- Mixes both orbital and spin contribution
- Requires relatively large samples (~mm^3)
- Give access to :
  Exchange magnetic coupling
  Magnetic anisotropy (gap…)
  Crystal field

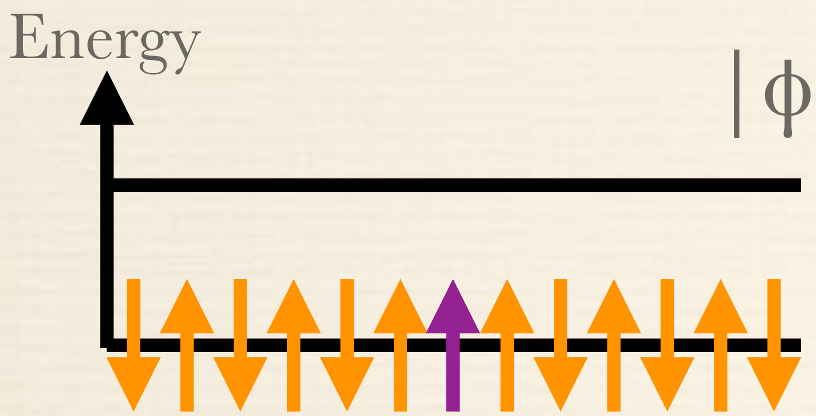
Raman Spectroscopy

Single magnon excitation



Raman Spectroscopy

Single magnon excitation



|ϕ>=$Y_{1,-1}$|↑> ± $Y_{1,1}$|↓>

Dipole transition : $\mathcal{V} = \vec{p}.\vec{\epsilon}$

$$\Delta L = 1$$
$$\Delta S = 0$$

Necessity of Spin-Orbit Coupling

Q=0

Raman Spectroscopy

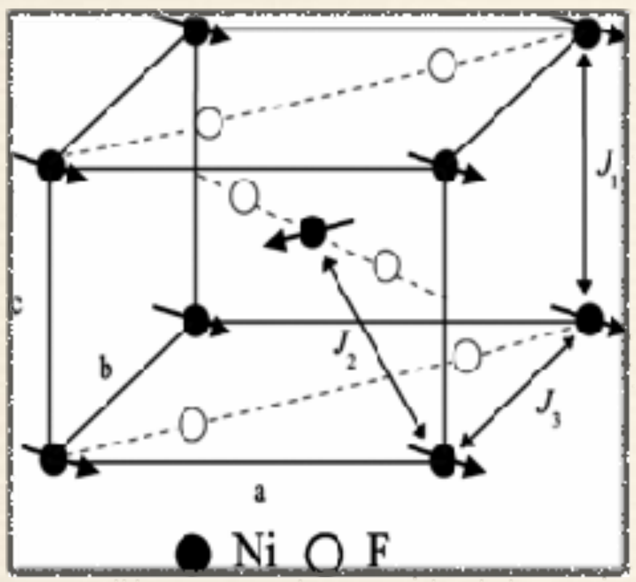
Two magnon excitation

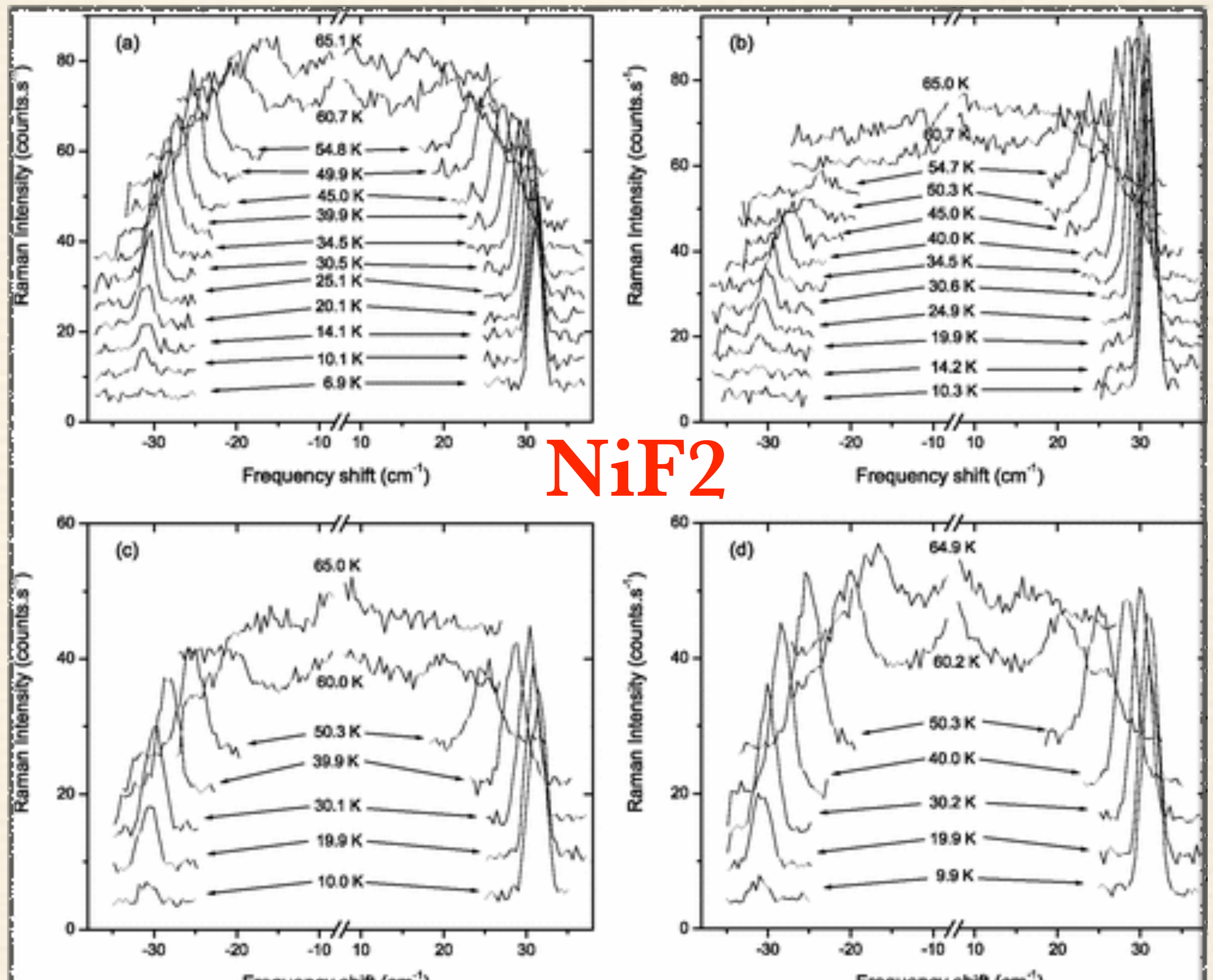
S=0

Q=0

Raman Spectroscopy

Single magnon excitation





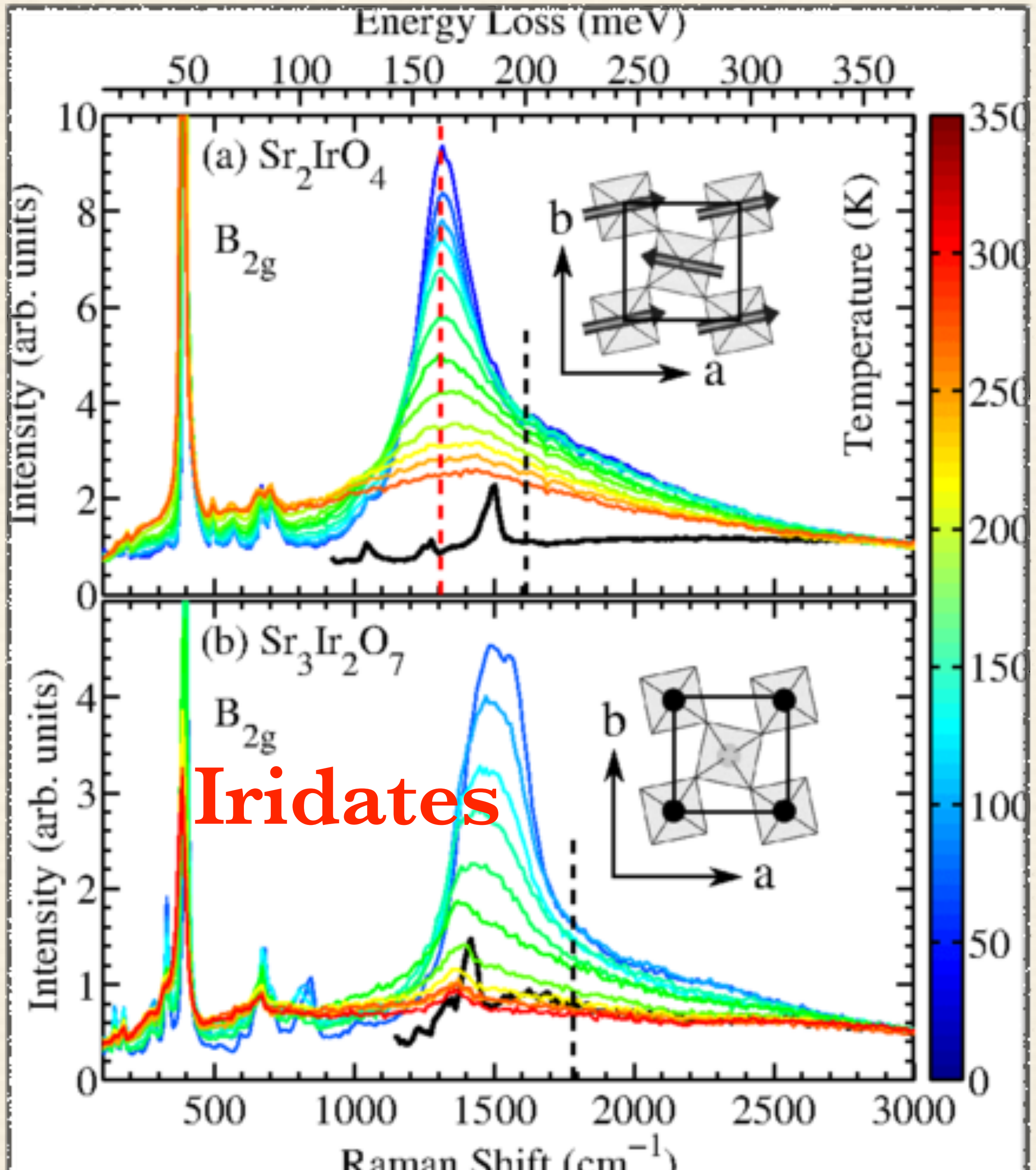
NiF2

PRB 81, 024426 (2010)

Magnetic anisotropy : gapped magnon at **Q**=0

Raman Spectroscopy

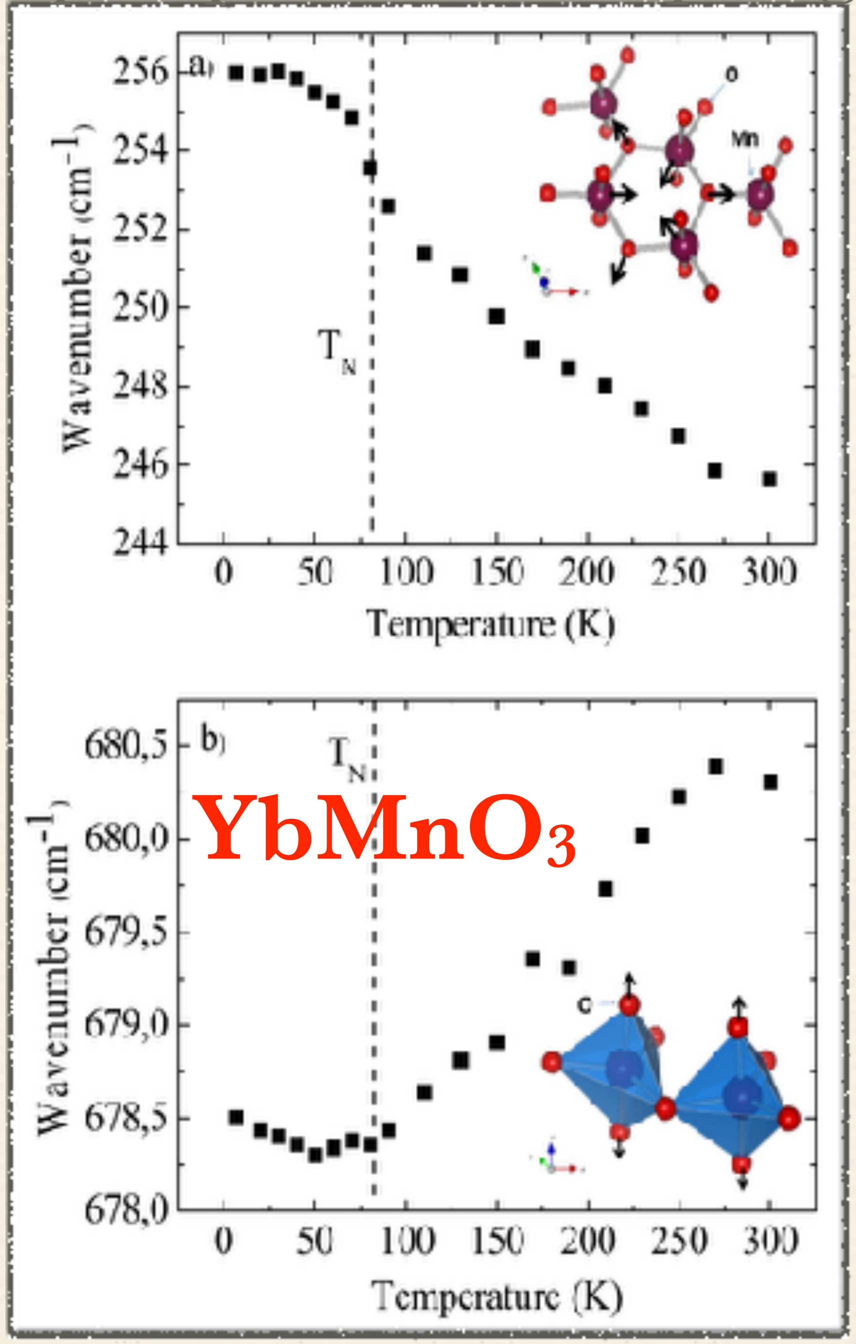
Two magnon excitation



PRL 116, 136401 (2016)

Raman Spectroscopy

Spin-phonon coupling



PRB 86, 184410 (2012)

Raman Spectroscopy

Summary

- Sensitive to magnon, bimagnon and Spin-Phonon coupling (and potential magnetic order signature)
- Requires relatively small samples (< 1 mm^2)
- Give access to
  - Single, bimagnon or two-magnons excitations at Q=0
  - Singulet or Triplet excitations (but selection rule not understood)
  - Magnetic anisotropy

2.2 Absorption and Emission

X-ray Magnetic Circular Dichroism

X-ray Magnetic Circular Dichroism

X-ray Circular Dichroism

Dipolar interaction : $\mathcal{V}(\vec{r}) = \frac{qE}{c}\vec{r}.\vec{\epsilon}$

Dipole Operator $\vec{r}.\vec{\epsilon}$ with Racah's tensor operators :

$P_{\pm1} = rC_{\pm1} = r\sqrt{\frac{4\pi}{3}}Y_{1,\pm1}$: circular polarization

$P_0 = rC_0 = r\sqrt{\frac{4\pi}{3}}Y_{1,0}$: linear polarization

Selection rules :

$\Delta L = \pm1 \quad \Delta S = 0$

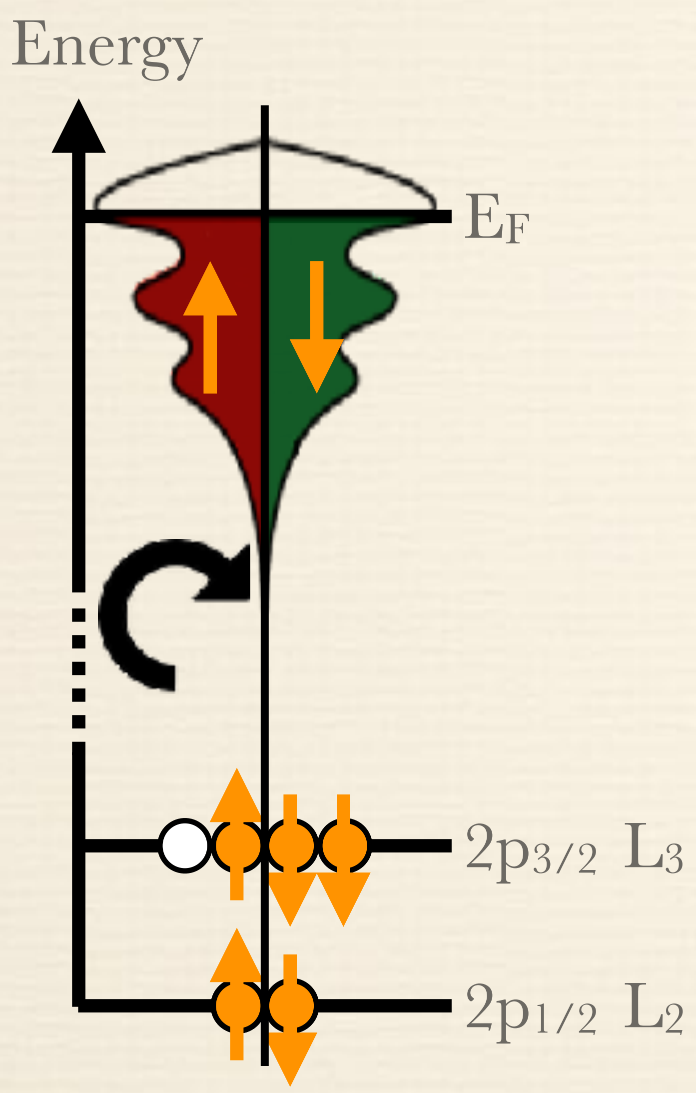
$\Delta m_l = +1$: Right Circular

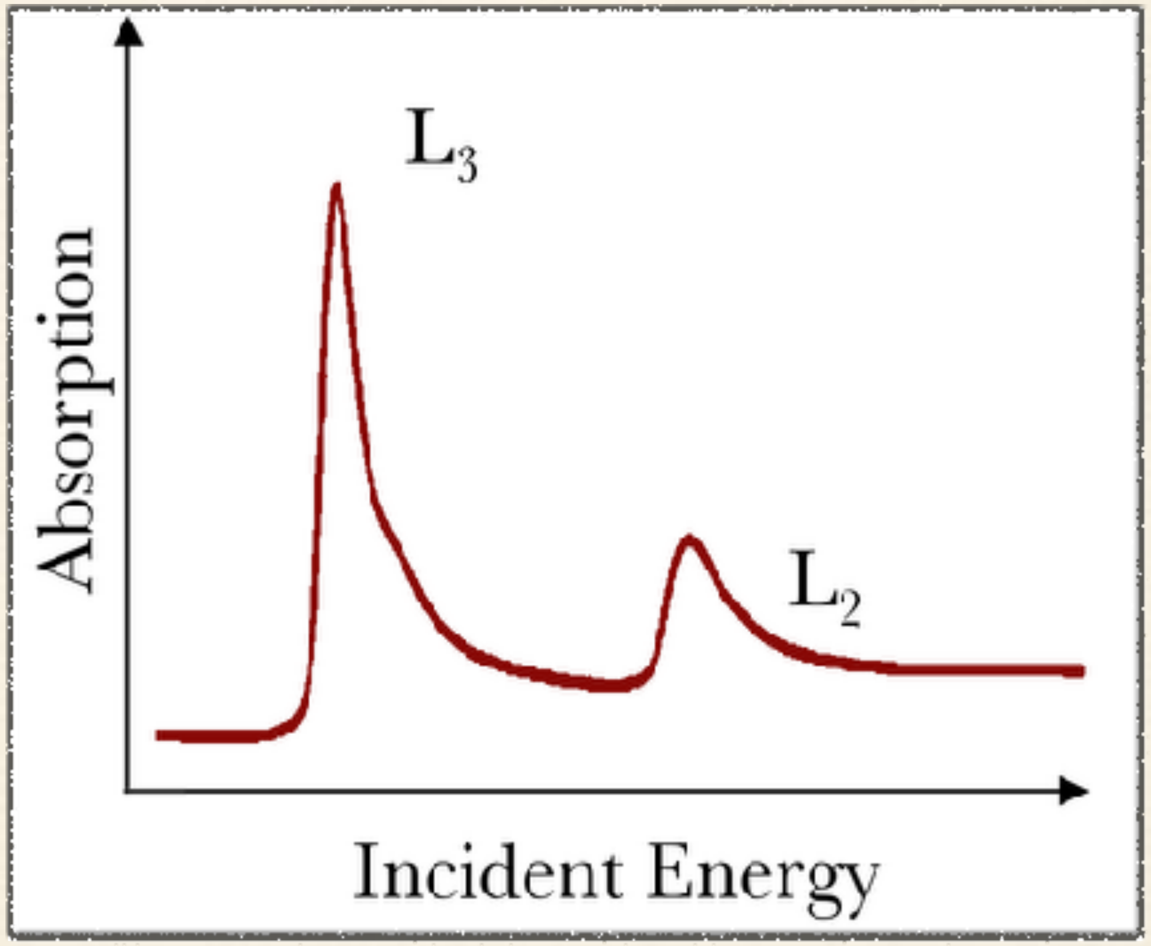
$\Delta m_l = -1$: Left Circular

X-ray Magnetic Circular Dichroism

X-ray Magnetic Circular Dichroism

L_2 L_3-edge M=0

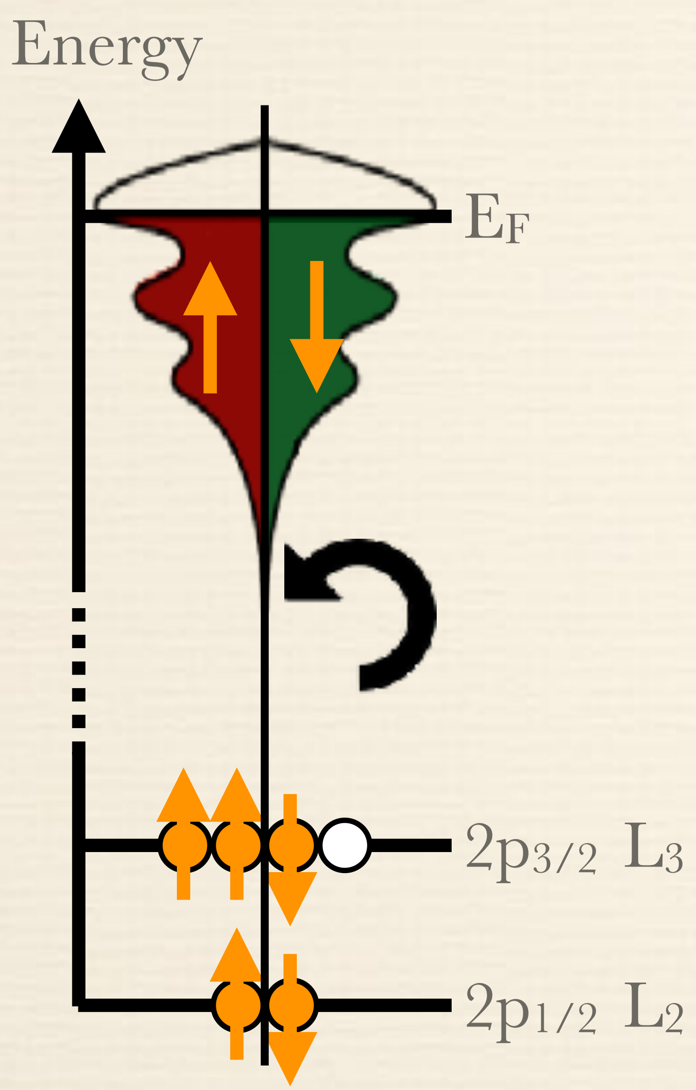


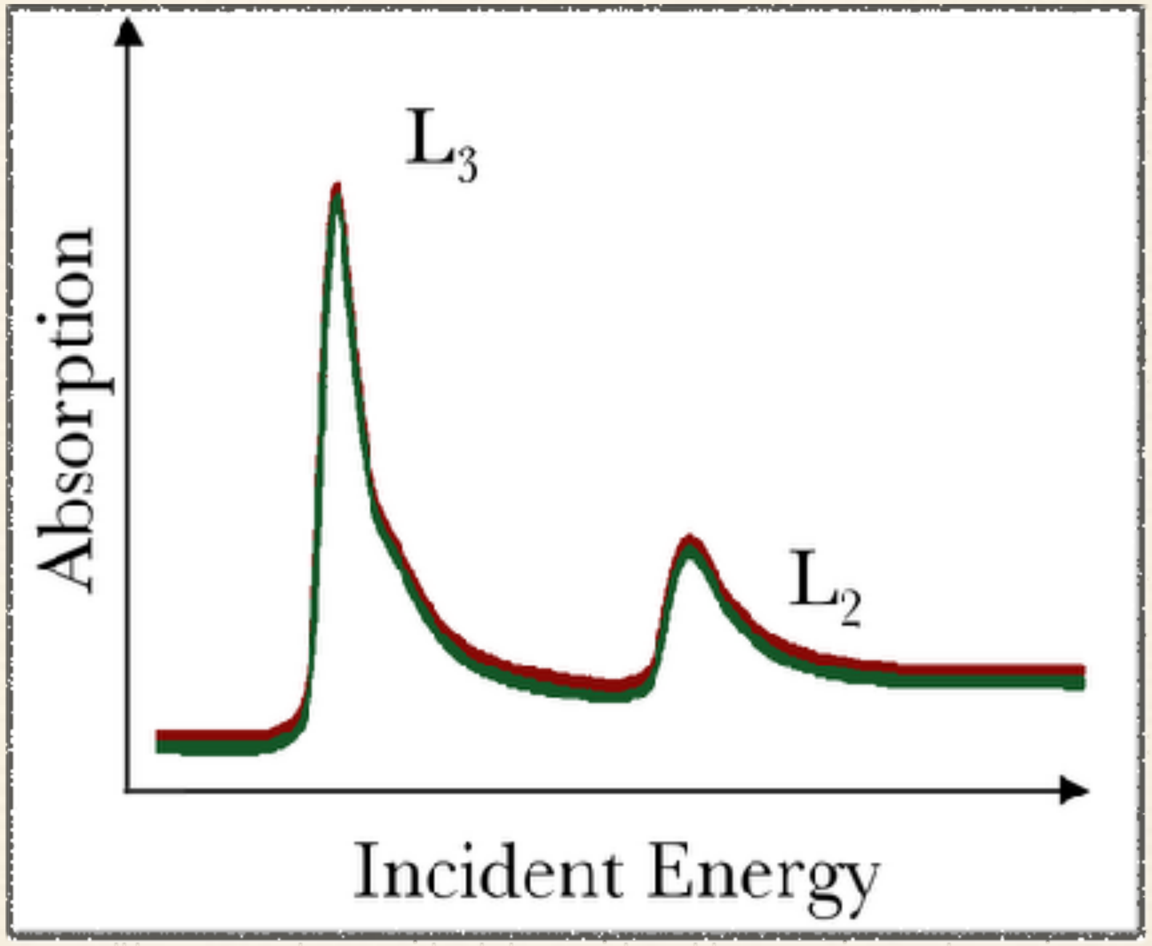


X-ray Magnetic Circular Dichroism

X-ray Magnetic Circular Dichroism

L_2 L_3-edge M=0

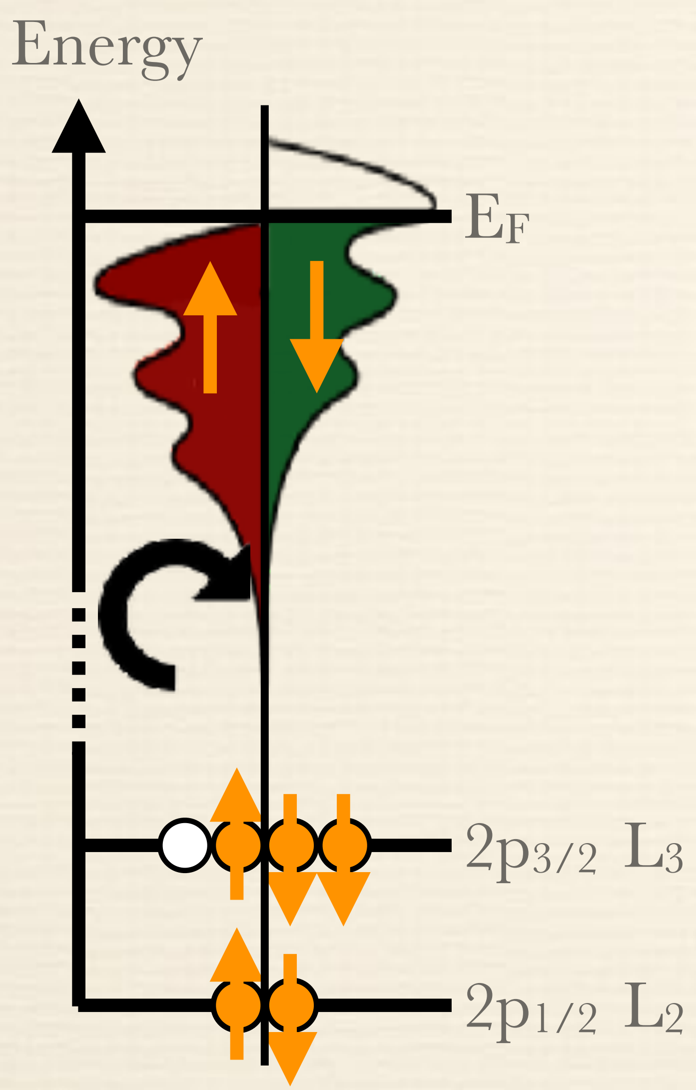


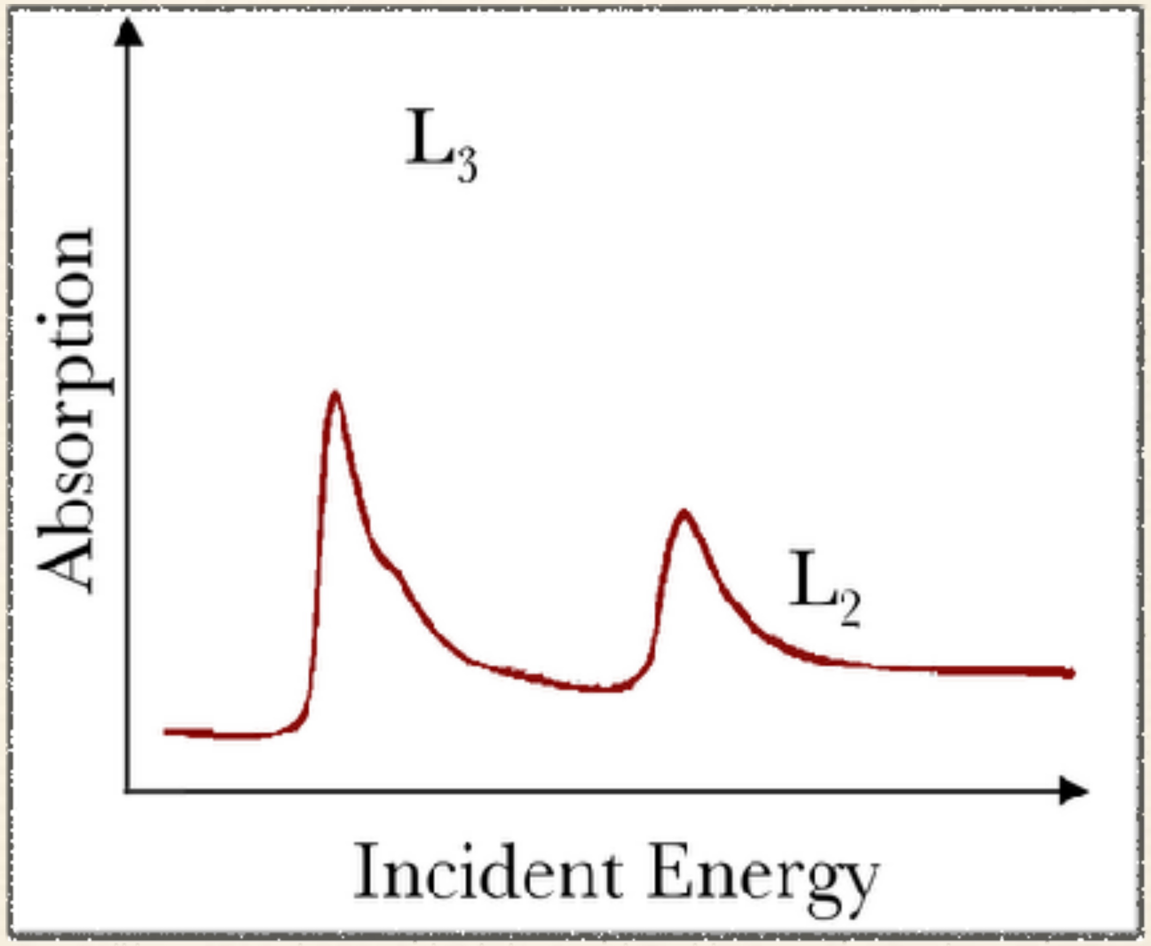


X-ray Magnetic Circular Dichroism

X-ray Magnetic Circular Dichroism

L_2 L_3-edge $M \neq 0$

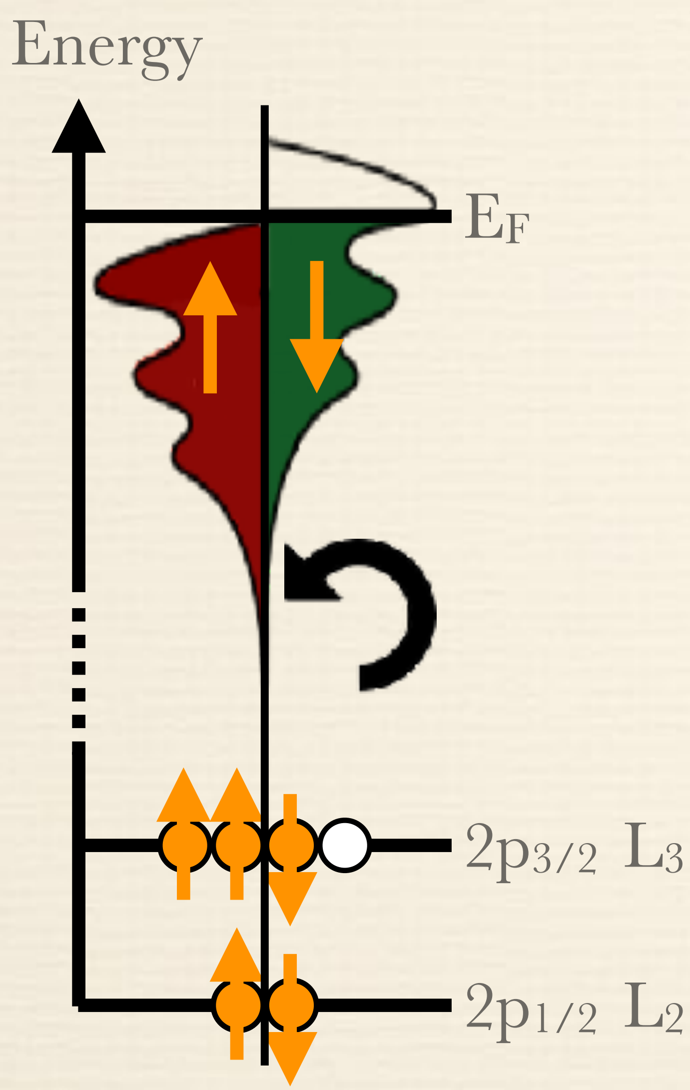


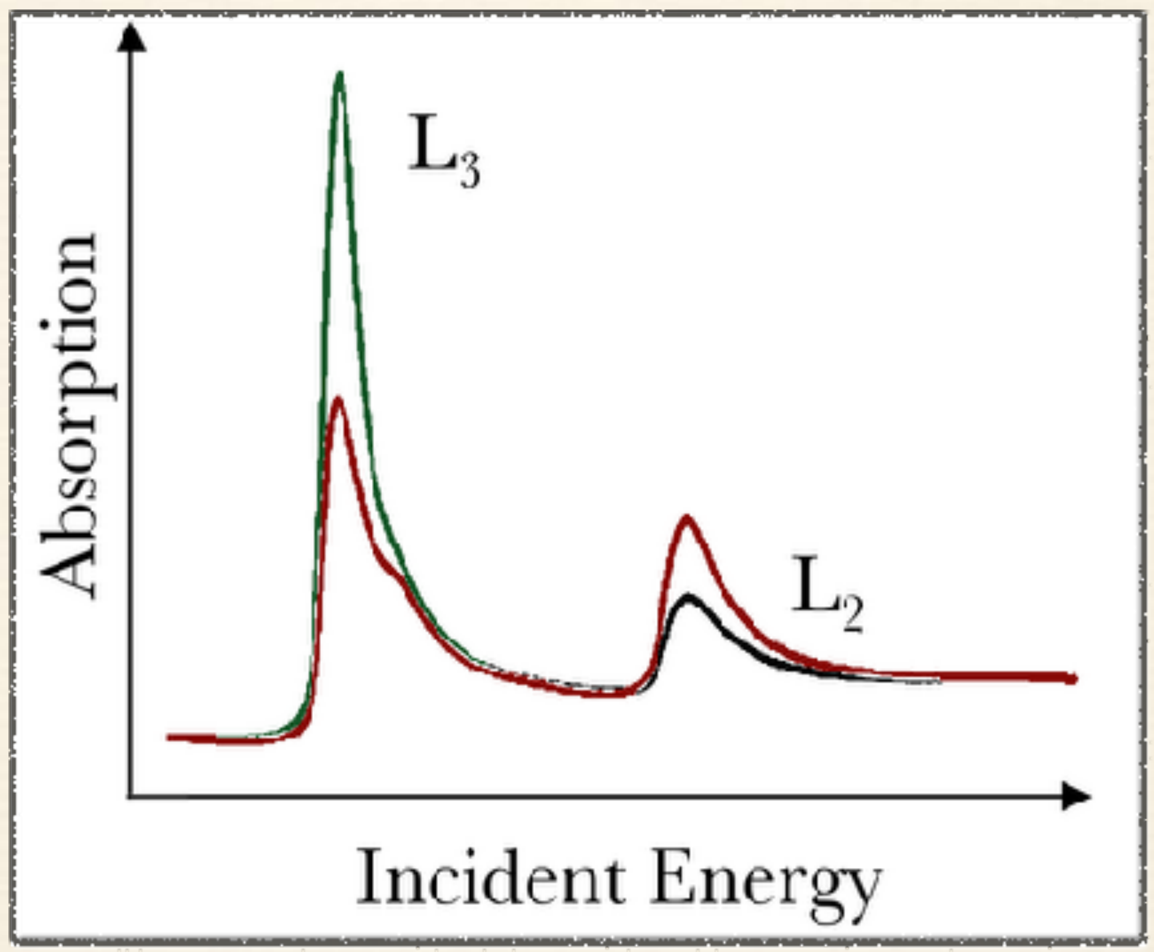


X-ray Magnetic Circular Dichroism

X-ray Magnetic Circular Dichroism

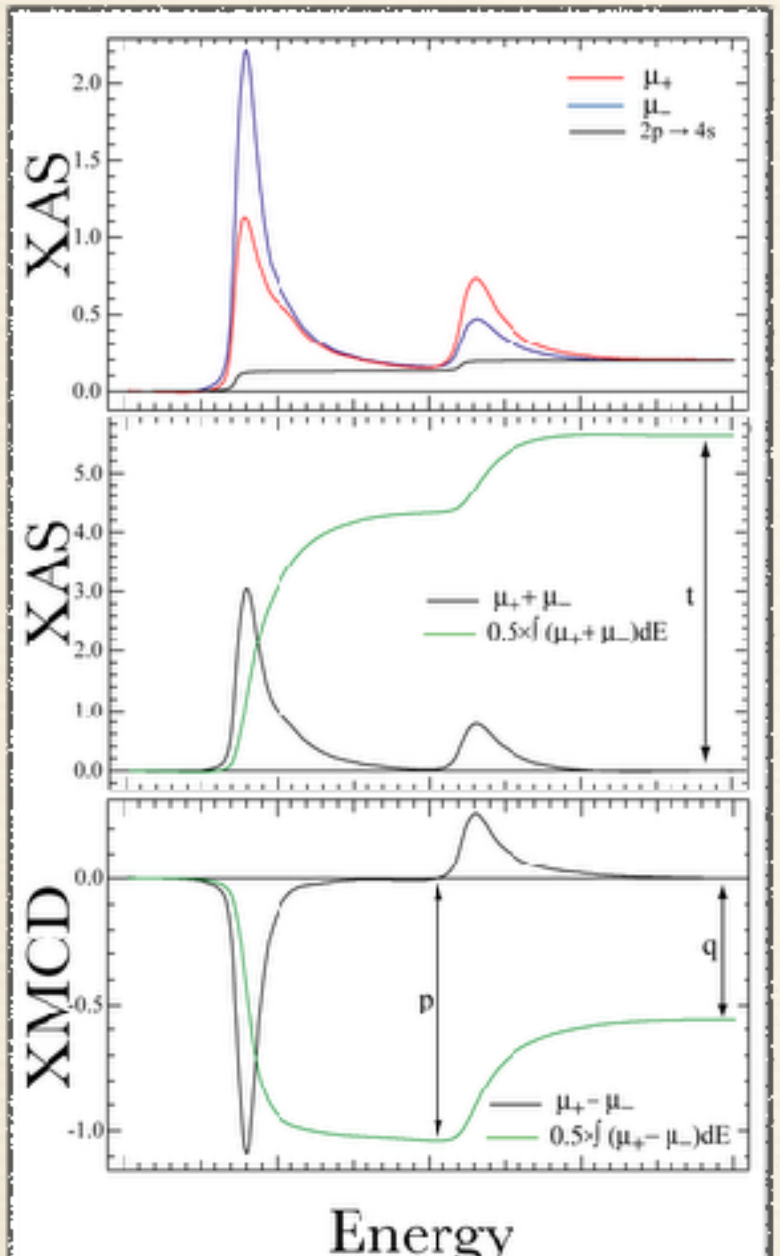
L_2 L_3-edge $M \neq 0$





X-ray Magnetic Circular Dichroism

X-ray Magnetic Circular Dichroism



$$L = -\,n_h \frac{4}{3} \frac{\int_{L_2,L_3} (\mu_+ - \mu_-) dE}{\int_{L_2,L_3} (\mu_+ + \mu_-) dE}$$

$$= -\frac{4}{3} n_h \frac{q}{t}$$

$$S = -n_h \frac{6 \int_{L_3} (\mu_+ - \mu_-) dE - 4 \int_{L_2,L_3} (\mu_+ - \mu_-) dE}{\int_{L_2,L_3} (\mu_+ + \mu_-) dE}$$

$$= -n_h \frac{6p - 4q}{t}$$

XMCD possible if no overlap $2p_{3/2}$ $2p_{1/2}$

Tricky data analysis : normalization…

X-ray Magnetic Circular Dichroism

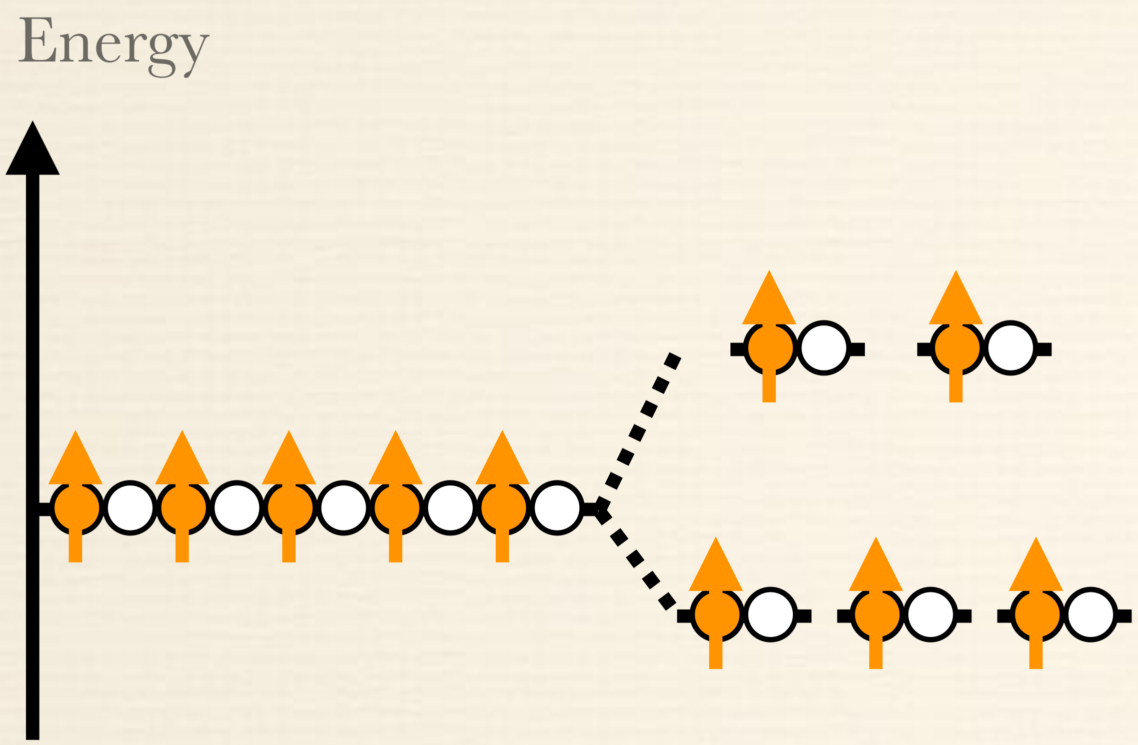
Summary

- Chemical/Element selectivity
- Sum rule : distinguish orbital and spin contribution
- Signal proportional to the magnetic moment along **k** of the absorbing atom (ferro/ferri-magnetism)
- For anti-ferromagnetism, analog of XMCD : XNLD

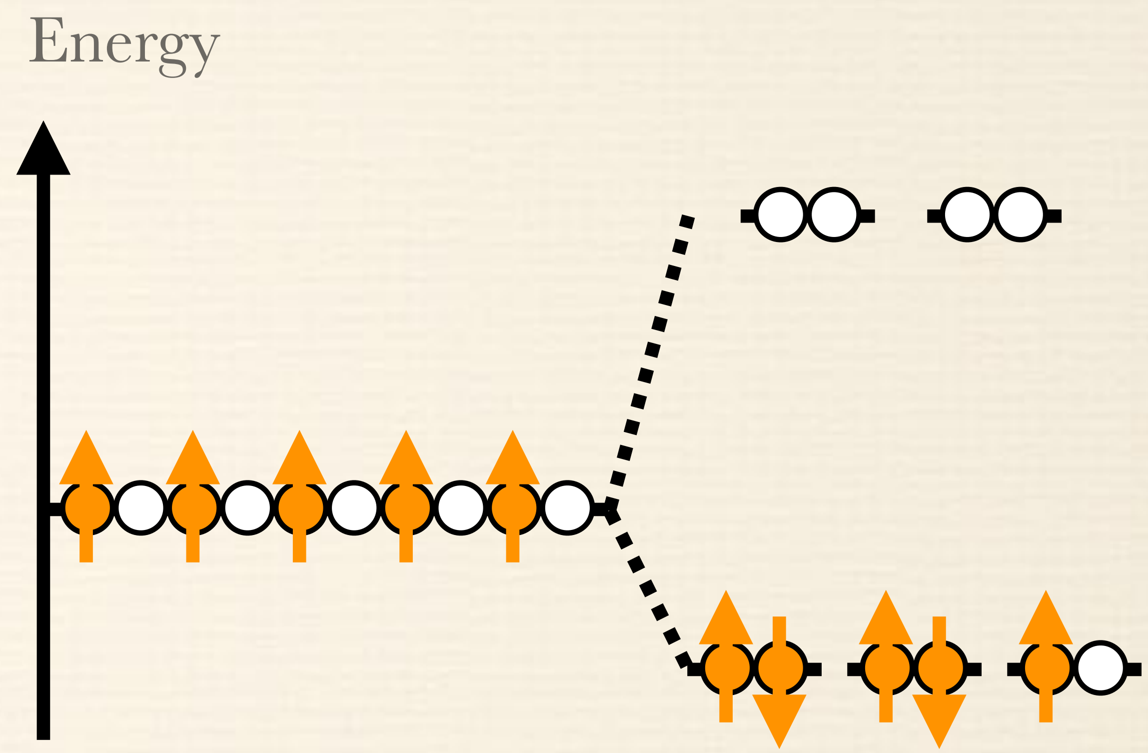
X-ray Emission Spectroscopy

X-ray Emission Spectroscopy

High and Low Spin states



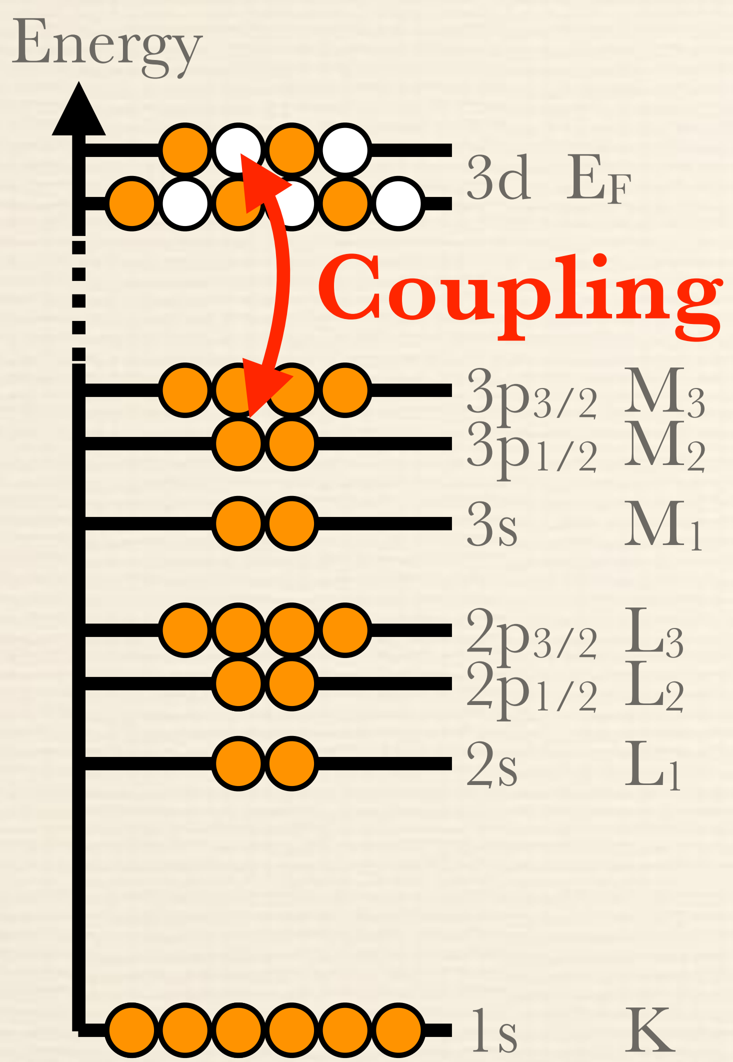
High Spin state



Low Spin state

X-ray Emission Spectroscopy

High and Low Spin states



$K_{\beta 1}$: $3p_{3/2}$ —> 1s
$K_{\beta 3}$: $3p_{1/2}$ —> 1s

$K_{\beta 1,3}$: 3p —> 1s

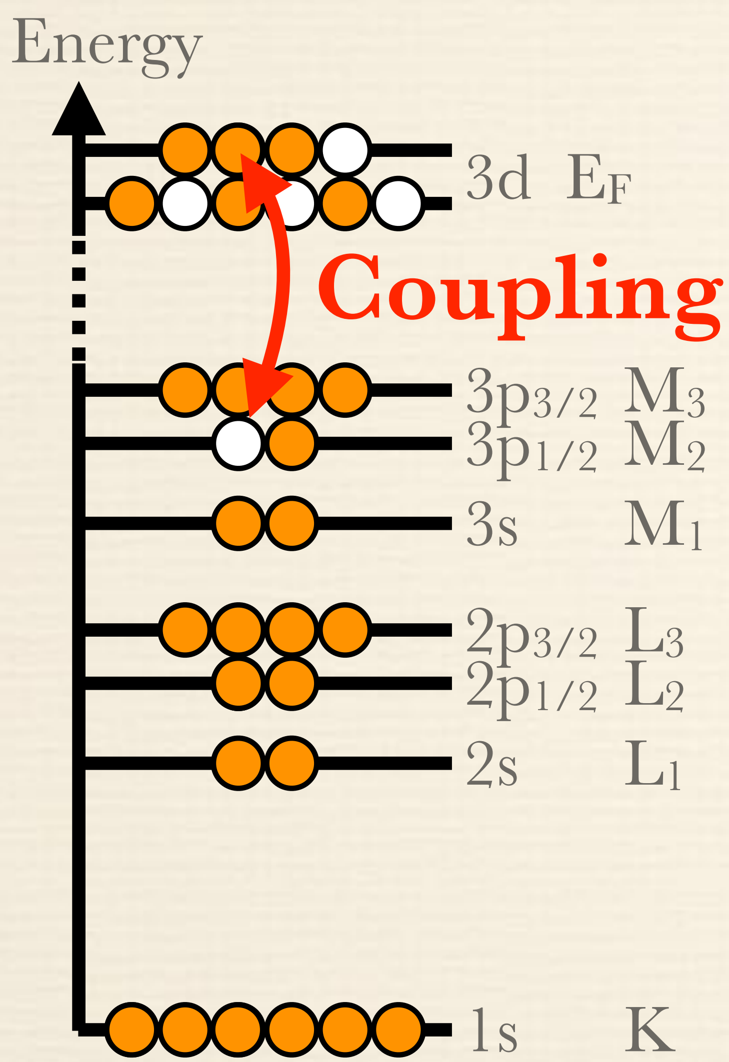
Coupling between unpaired *3d* and *3p* states

Lift of degeneracy : two final states for K_{β}

Sensitive to spin state : ratio $K_{\beta 1,3}/K_{\beta'}$

X-ray Emission Spectroscopy

High and Low Spin states



$K_{\beta1}$: $3p_{3/2}$ —> 1s
$K_{\beta3}$: $3p_{1/2}$ —> 1s

$K_{\beta1,3}$: 3p —> 1s

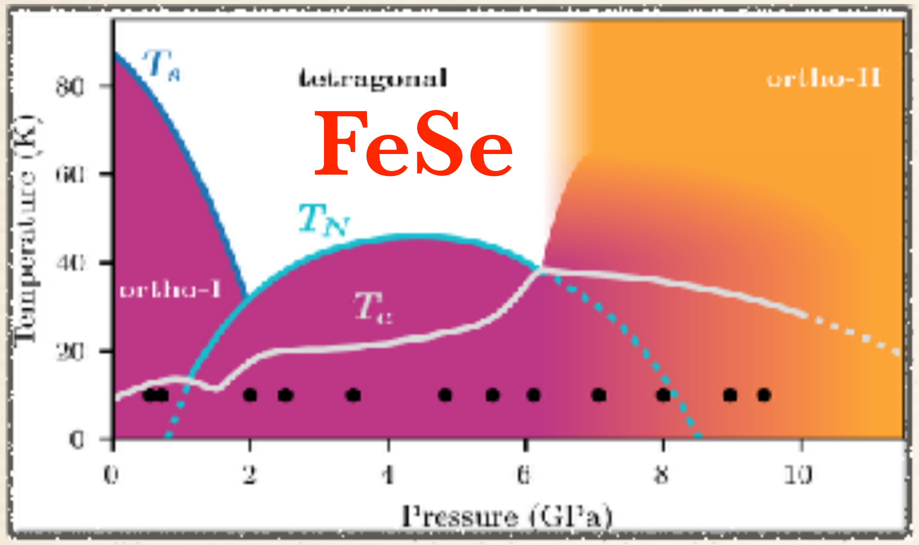
Coupling between unpaired *3d* and *3p* states

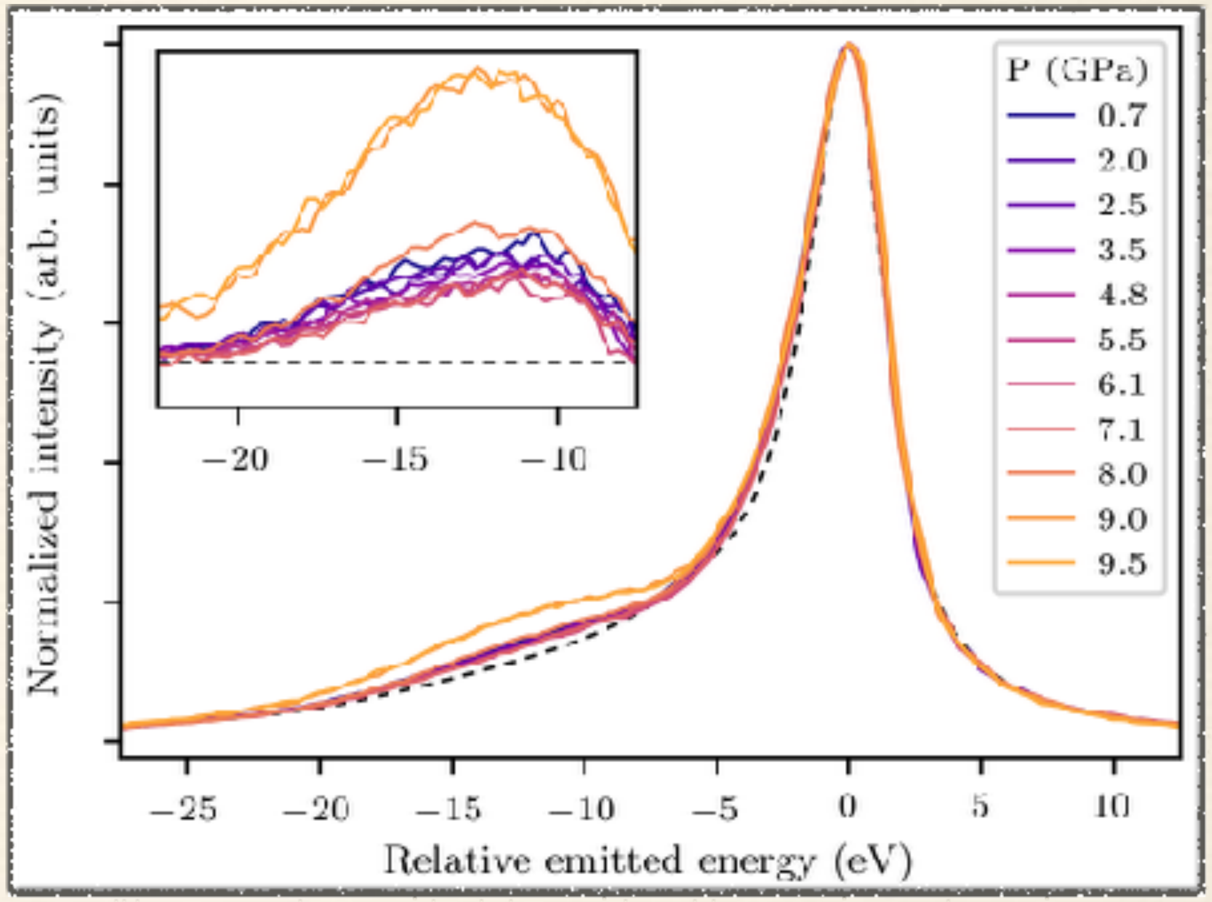
Lift of degeneracy : two final states for K_β

Sensitive to spin state : ratio $K_{\beta1,3}/K_{\beta'}$

X-ray Emission Spectroscopy

High-Spin — Low-Spin transition





X-ray Emission Spectroscopy

Summary

- Chemical/Element selectivity
- Orbital selectivity (selection of emission line)
- Signal proportional to the absolute value of local magnetic moment of absorbing atom (unpaired electrons)
- Local consideration : independent of magnetic ordering

RIXS

Can RIXS be sensitive to (one, two, bi)magnon ?

Matrix elements **Final state** **Initial state**

$$\sum_n \langle d_{x^2-y^2,\uparrow}|\mathcal{D}|n\rangle\langle n|\mathcal{D}|d_{x^2-y^2,\downarrow}\rangle$$

$\mathcal{D}$: Dipolar operator : $\Delta L = 1$ and $\Delta S = 0$

RIXS

Can RIXS be sensitive to (one, two, bi)magnon ?

Matrix elements **Final state** **Initial state**

$$\sum_n \langle d_{x^2-y^2,\uparrow}|\mathcal{D}|n\rangle\langle n|\mathcal{D}|d_{x^2-y^2,\downarrow}\rangle$$

$\mathcal{D}$: Dipolar operator : $\Delta L = 1$ and $\Delta S = 0$

For K-edge : 1s hole **L**=0, **L**.**S**=0, $|n\rangle = |\uparrow\rangle$ *or* $|\downarrow\rangle$

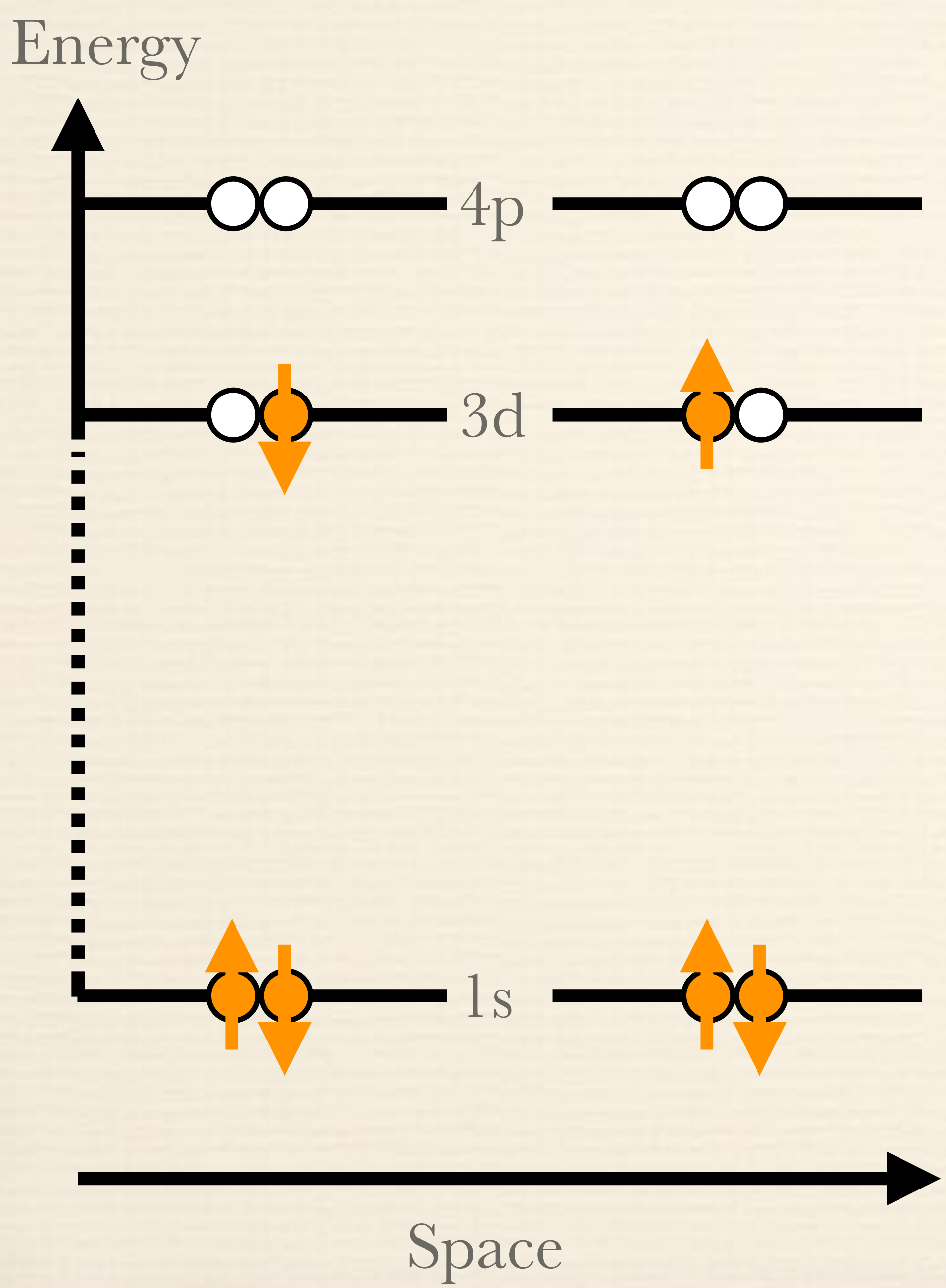
$$\langle d_{x^2-y^2,\uparrow}|\mathcal{D}|\uparrow\rangle\langle\uparrow|\mathcal{D}|d_{x^2-y^2,\downarrow}\rangle + \langle d_{x^2-y^2,\uparrow}|\mathcal{D}|\downarrow\rangle\langle\downarrow|\mathcal{D}|d_{x^2-y^2,\downarrow}\rangle$$

Nul matrix elements

No single magnon possible, only bimagnons

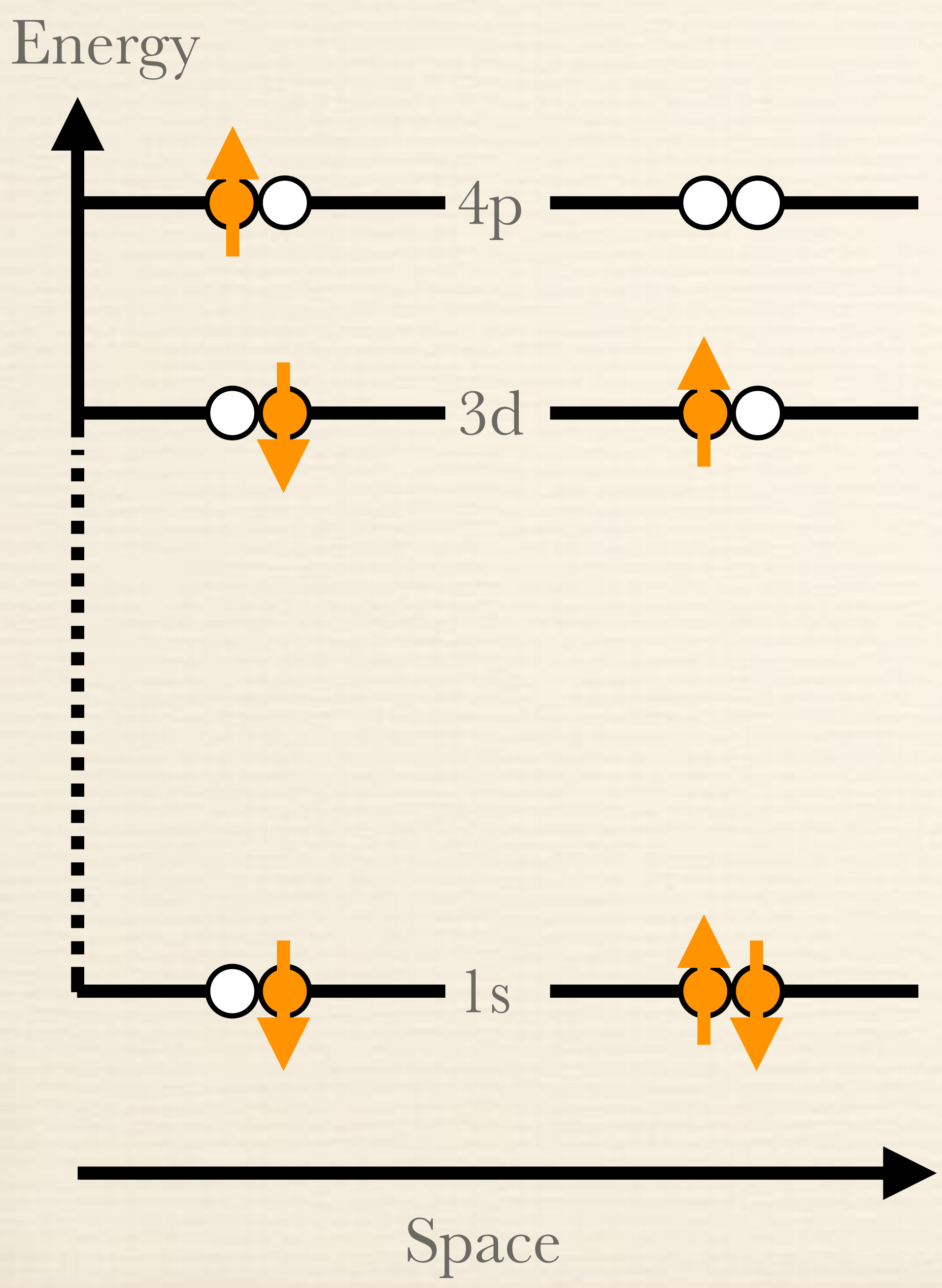
RIXS

K-edge bimagnon in cuprate



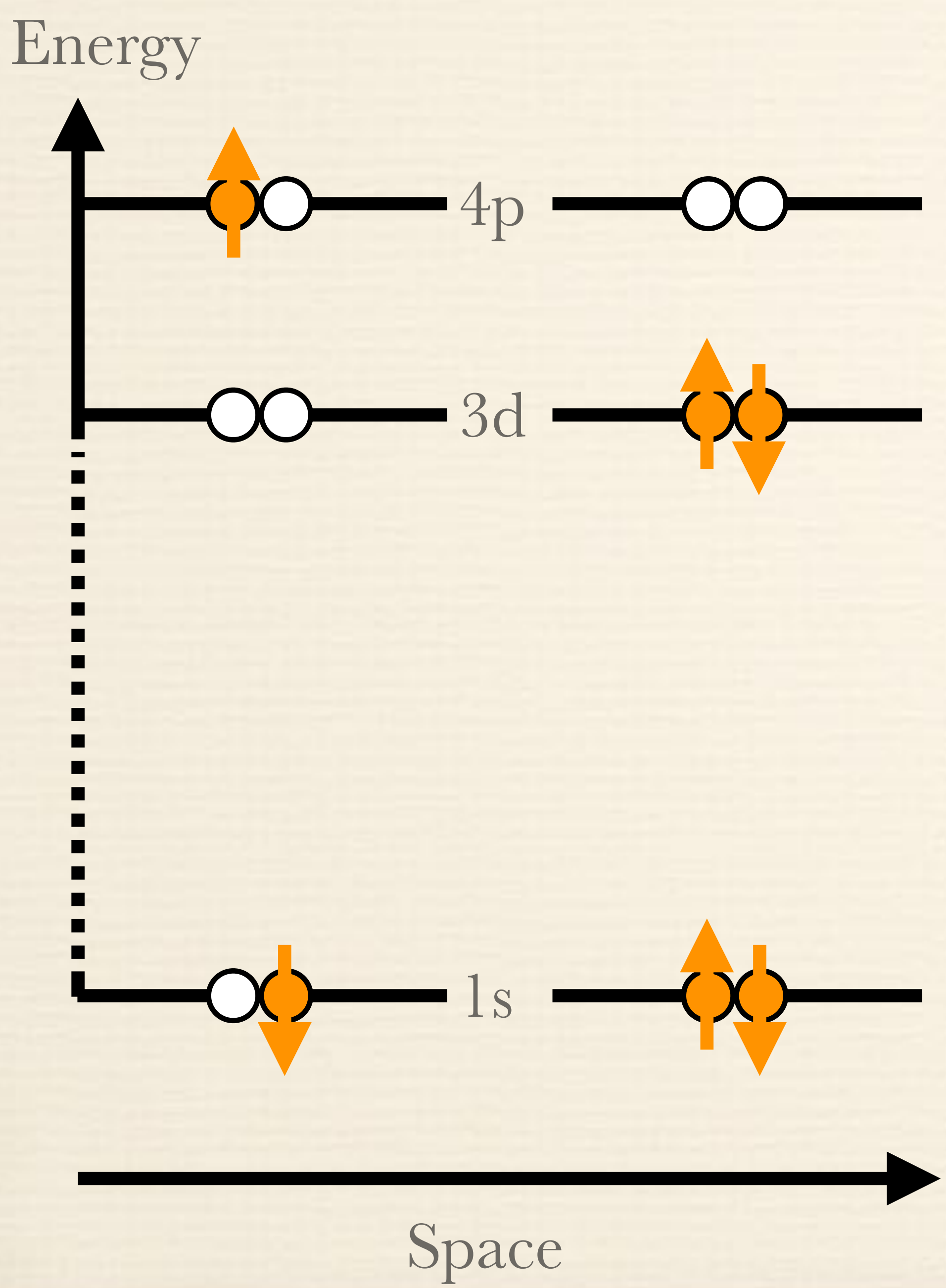
RIXS

K-edge bimagnon in cuprate



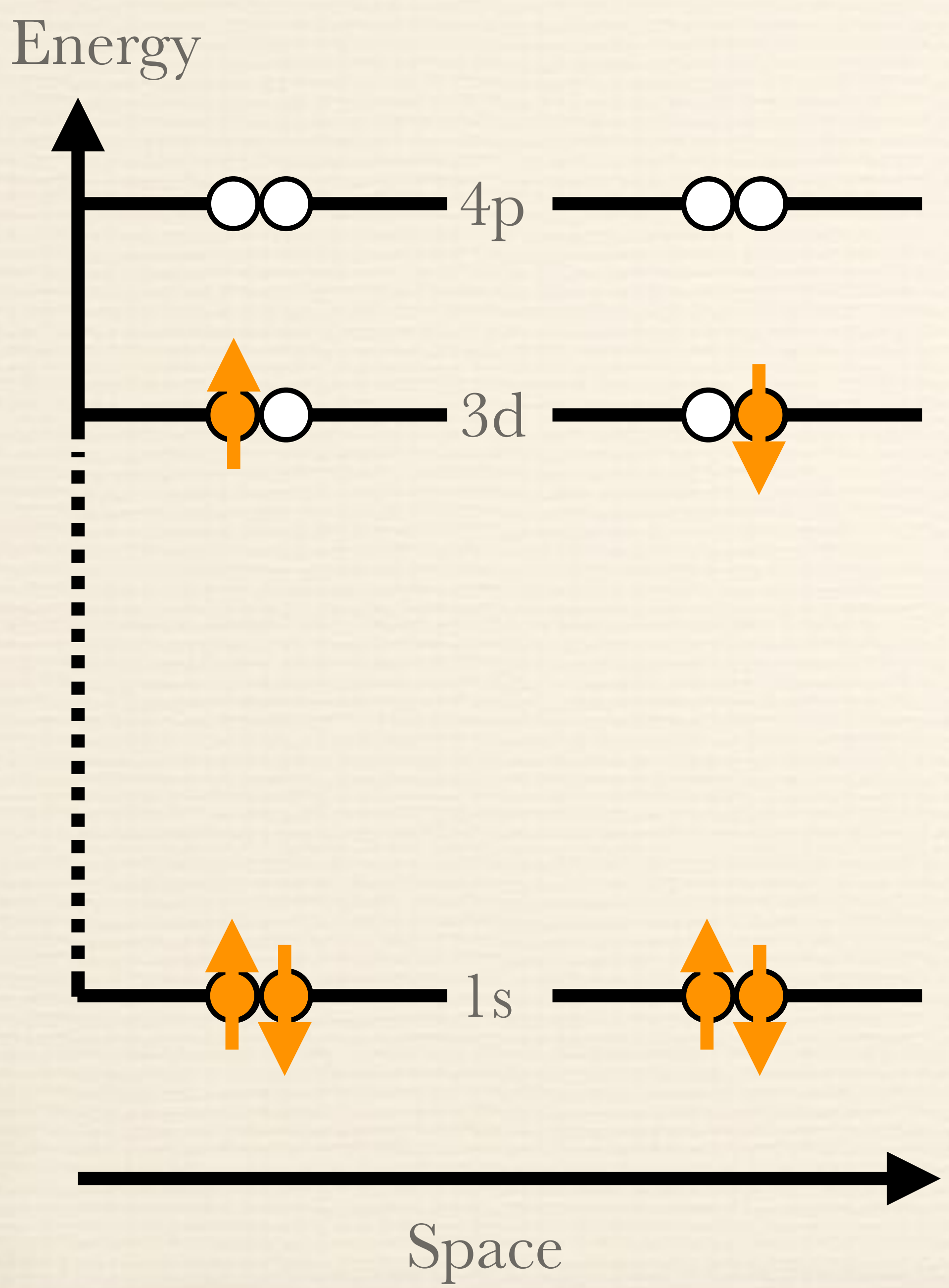
RIXS

K-edge bimagnon in cuprate



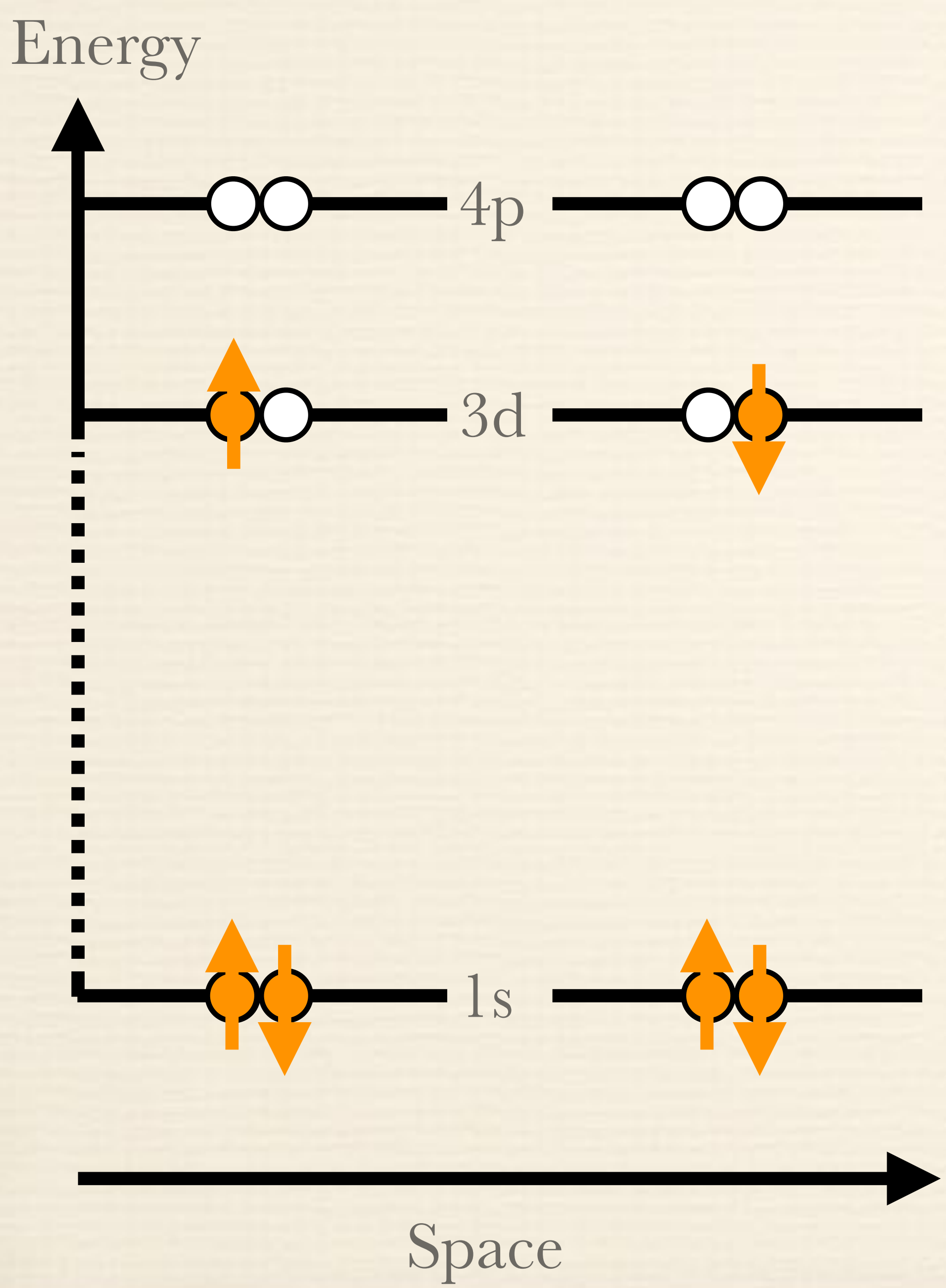
RIXS

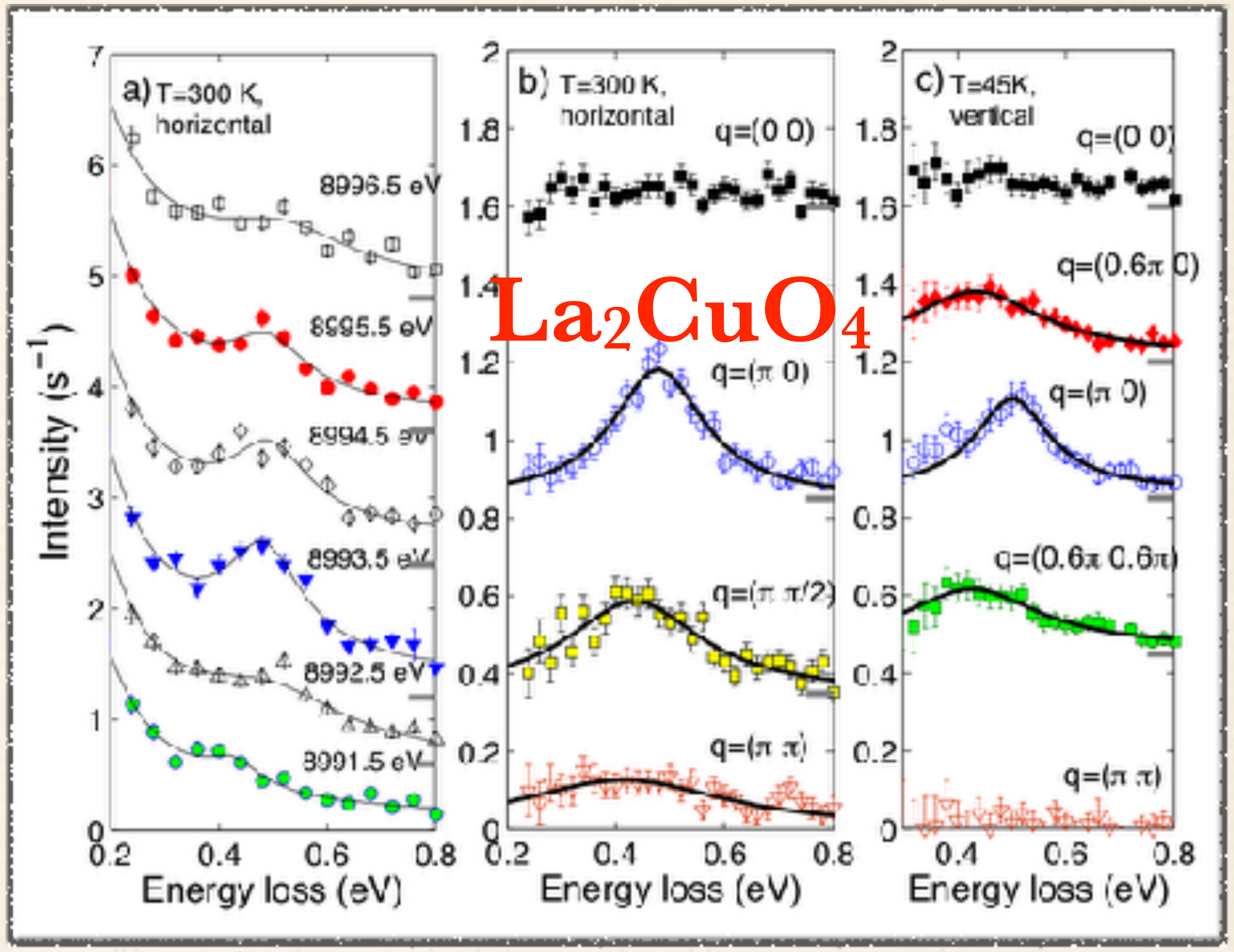
K-edge bimagnon in cuprate



RIXS

K-edge bimagnon in cuprate



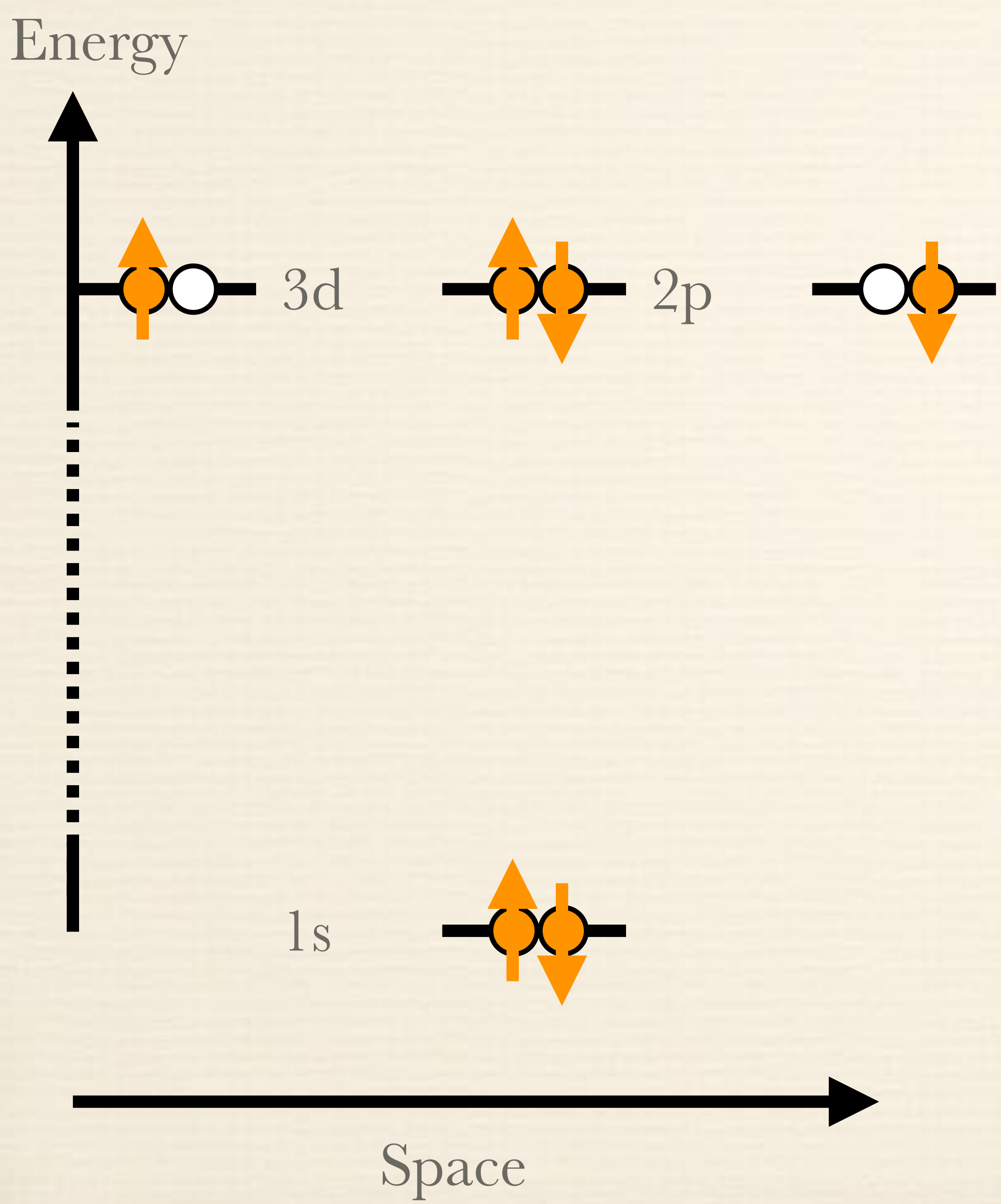


Cu K-edge bimagnon

PRL 100, 097001 (2008)

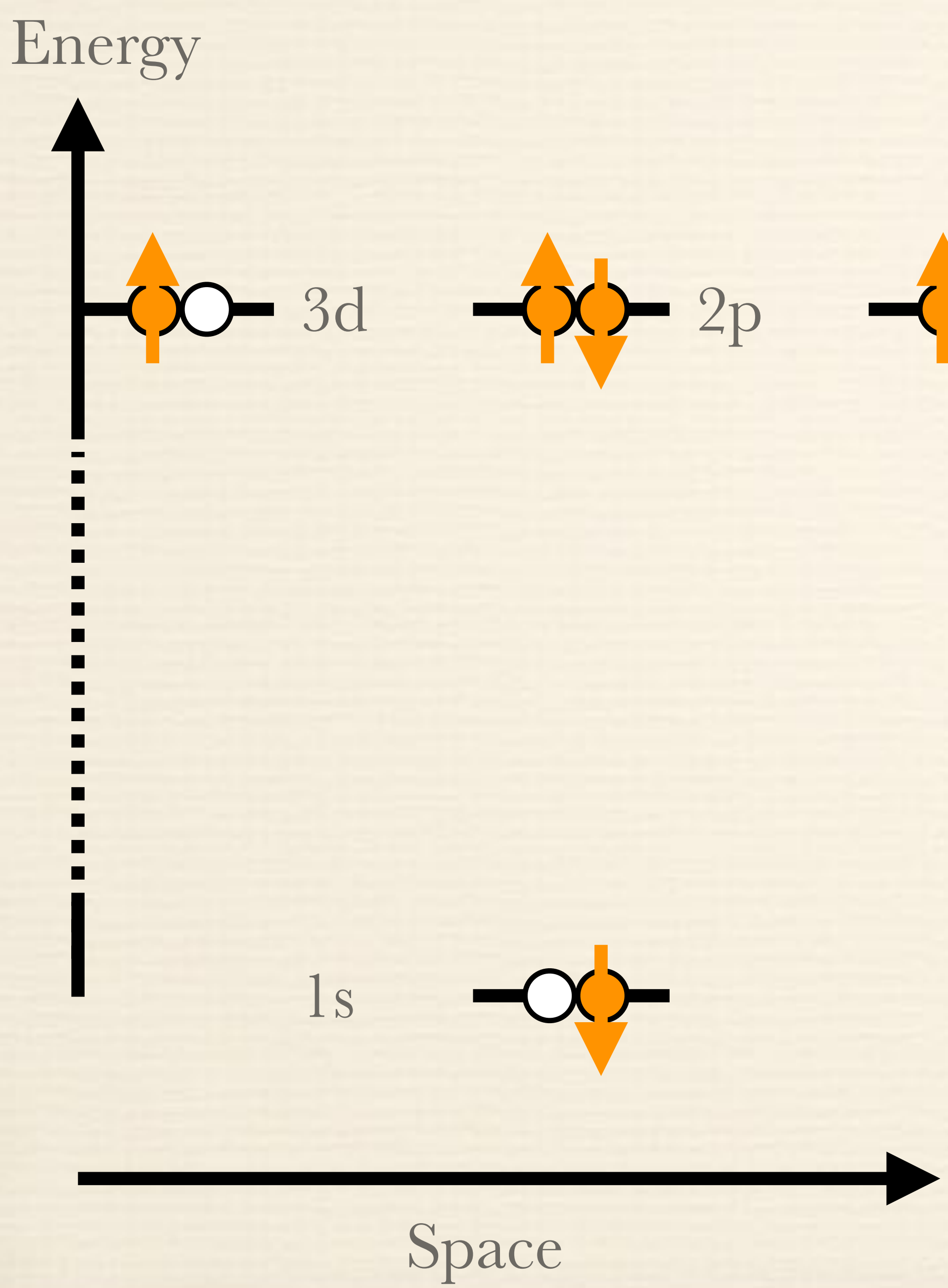
RIXS

K-edge bimagnon in cuprate



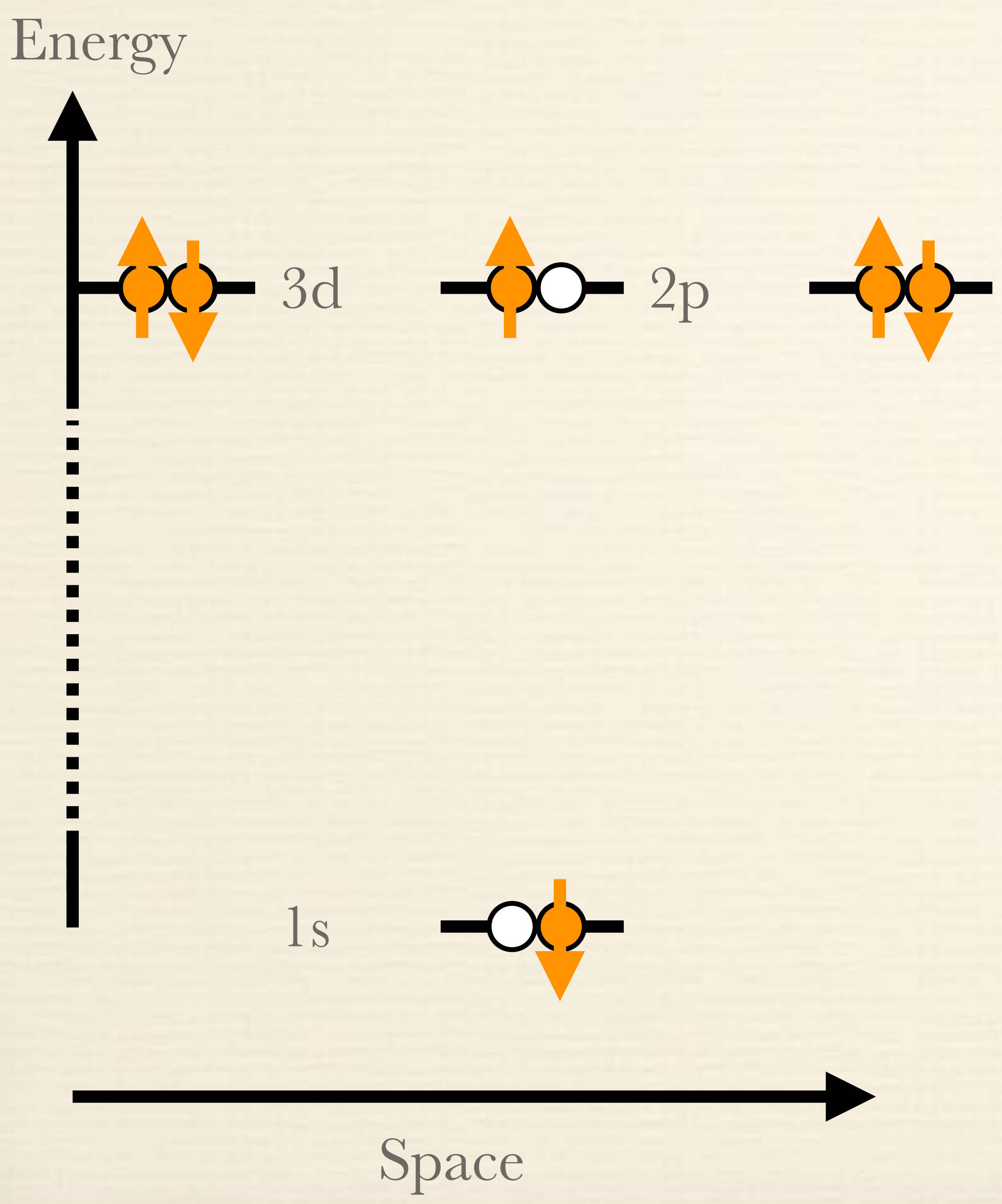
RIXS

K-edge bimagnon in cuprate



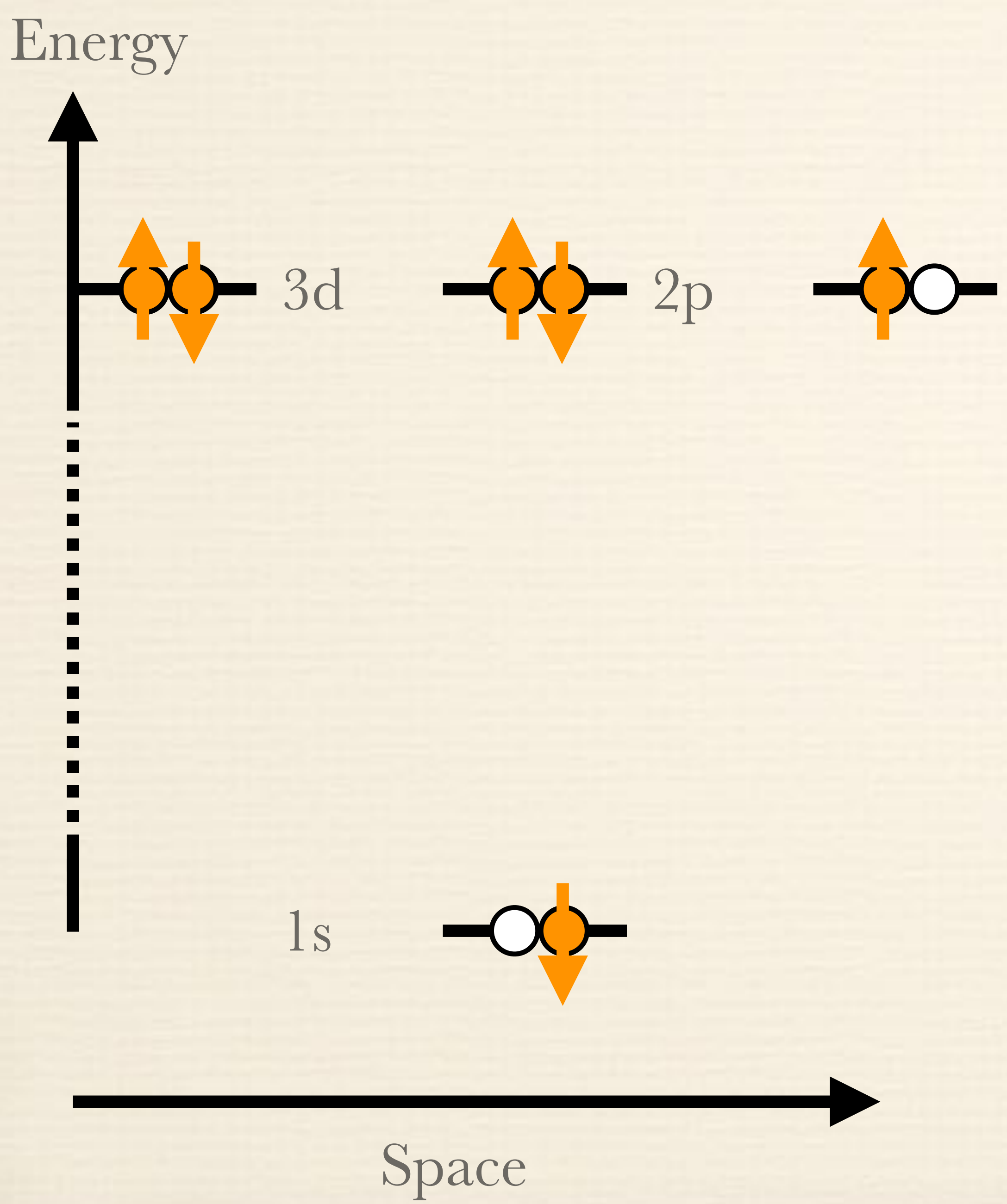
RIXS

K-edge bimagnon in cuprate



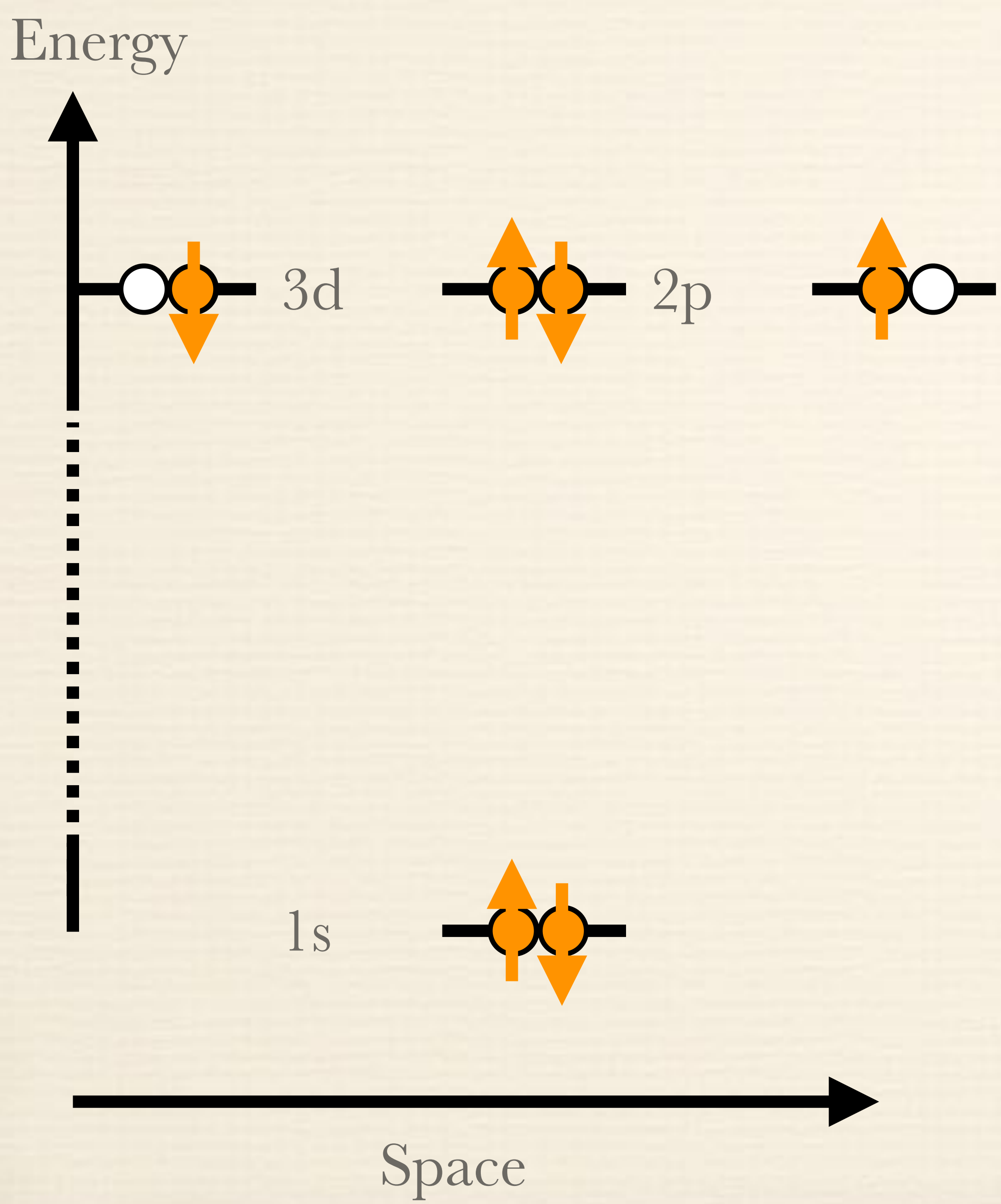
RIXS

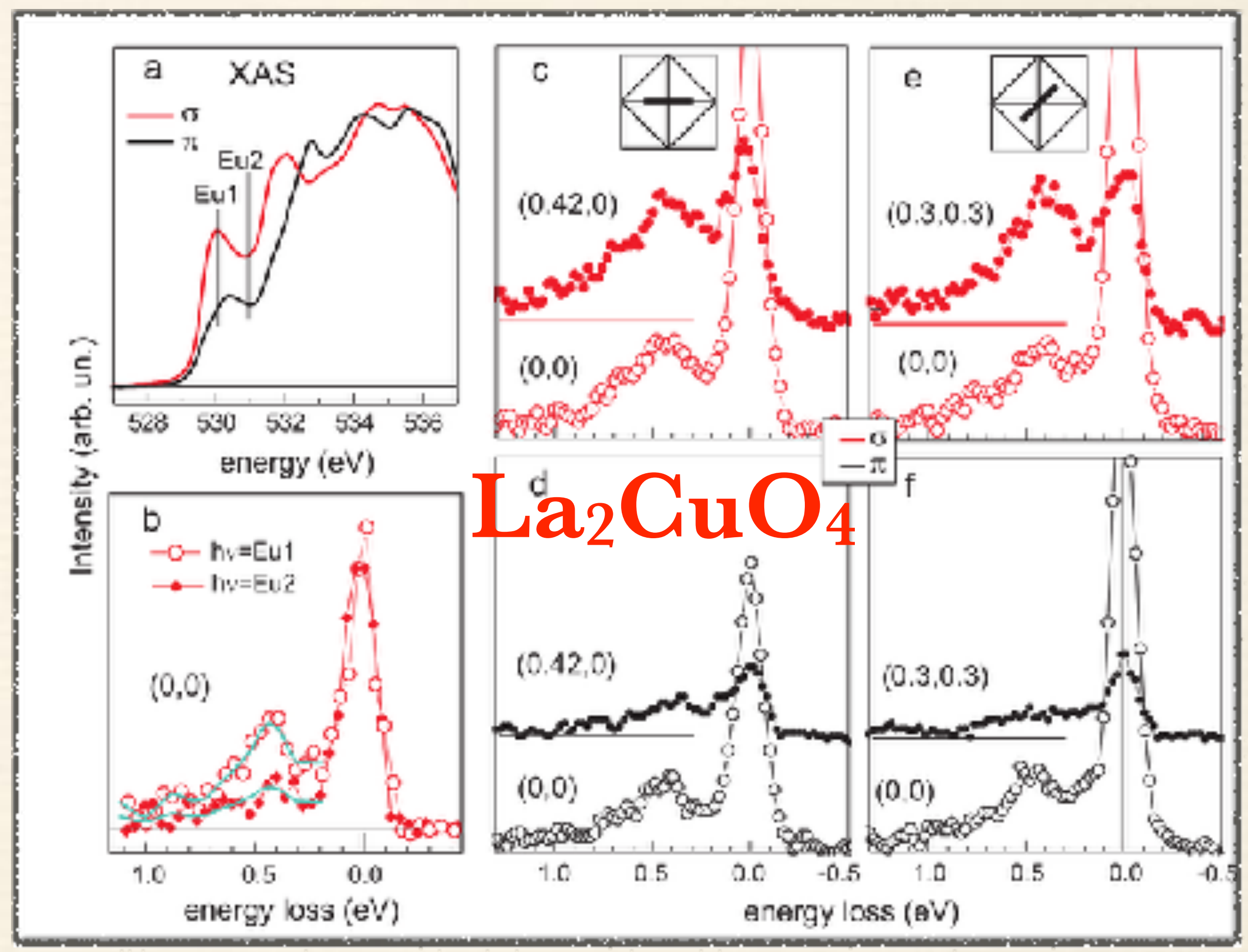
K-edge bimagnon in cuprate



RIXS

K-edge bimagnon in cuprate





PRB 85, 214527 (2008)

O K-edge bimagnon

RIXS

Can RIXS be sensitive to (one, two, bi)magnon ?

Matrix elements

$$\sum_n \langle d_{x^2-y^2,\uparrow}|\mathcal{D}|n\rangle\langle n|\mathcal{D}|d_{x^2-y^2,\downarrow}\rangle$$

$\mathcal{D}$: Dipolar operator : $\Delta L = 1$ and $\Delta S = 0$

For L_3-edge : $2p_{3/2}$ hole **L**=1, **L**.**S**≠0, $|n\rangle = |\uparrow\rangle$

$$|\uparrow\rangle + |\downarrow\rangle$$
$$|\uparrow\rangle - |\downarrow\rangle$$
$$|\downarrow\rangle$$

$$\langle d_{x^2-y^2,\uparrow}|\mathcal{D}|\uparrow\rangle\langle\uparrow|\mathcal{D}|d_{x^2-y^2,\downarrow}\rangle + \langle d_{x^2-y^2,\uparrow}|\mathcal{D}|\uparrow+\downarrow\rangle\langle\uparrow+\downarrow|\mathcal{D}|d_{x^2-y^2,\downarrow}\rangle$$
$$+\langle d_{x^2-y^2,\uparrow}|\mathcal{D}|\uparrow-\downarrow\rangle\langle\uparrow-\downarrow|\mathcal{D}|d_{x^2-y^2,\downarrow}\rangle + \langle d_{x^2-y^2,\uparrow}|\mathcal{D}|\downarrow\rangle\langle\downarrow|\mathcal{D}|d_{x^2-y^2,\downarrow}\rangle$$

Single magnon possible

RIXS

L-edge magnon in Cuprates

Flip the spin if in xy plane

$$\mathcal{D} = \sum_n r S_n^z e^{i\vec{k}_f.\vec{R}_n} d_n^\dagger p_n + r S_n^z e^{i\vec{k}_i.\vec{R}_n} p_n^\dagger d_n$$

Emission **Absorption**

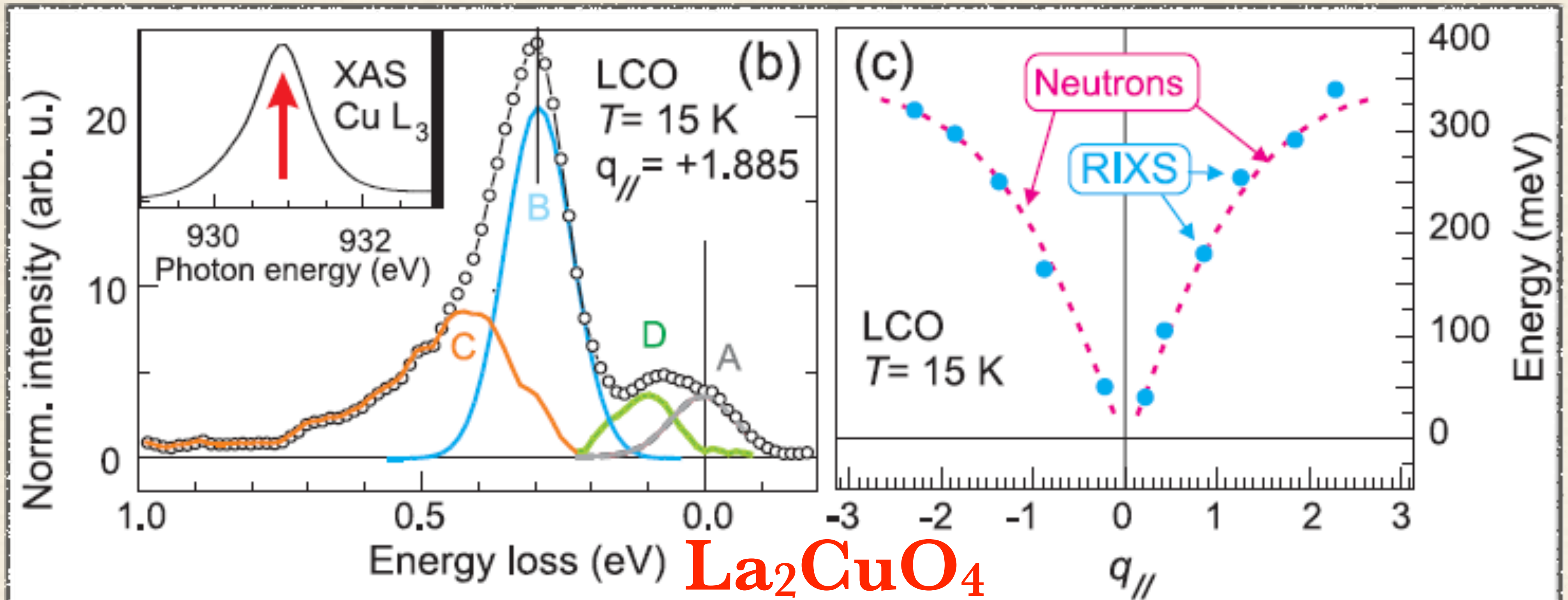
Direct Spin-Flip impossible for Spin S^z for $d_{x^2-y^2}$

Fondamental reason : $\Delta_L^z = 2$ (dipole forbidden)

Polarization is important to determine if matrix elements are zero or not (but hidden here)

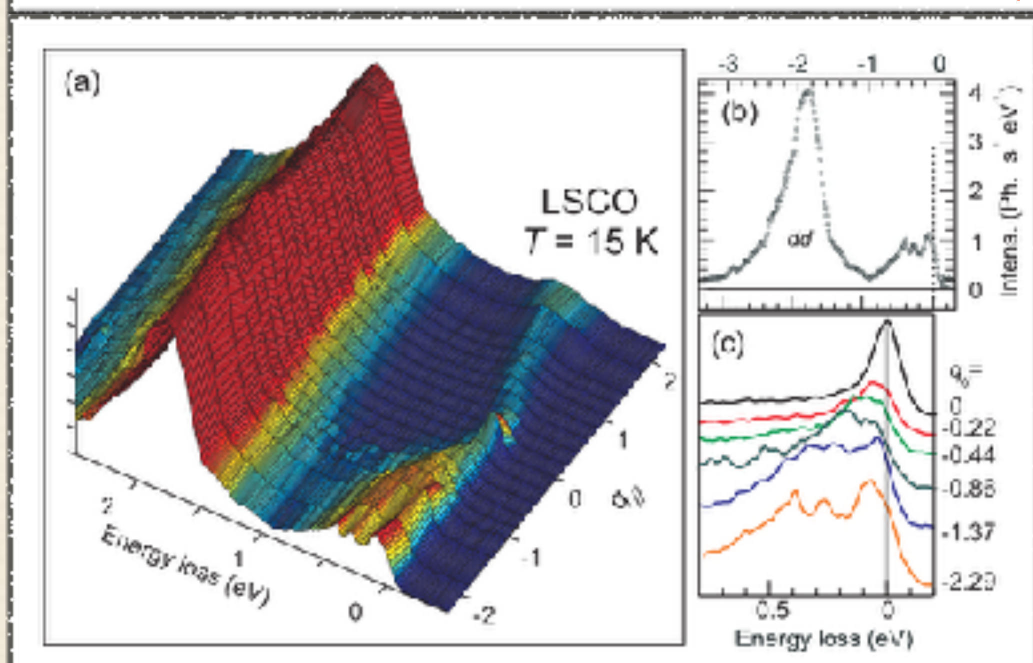
RIXS

L-edge magnon



La_2CuO_4

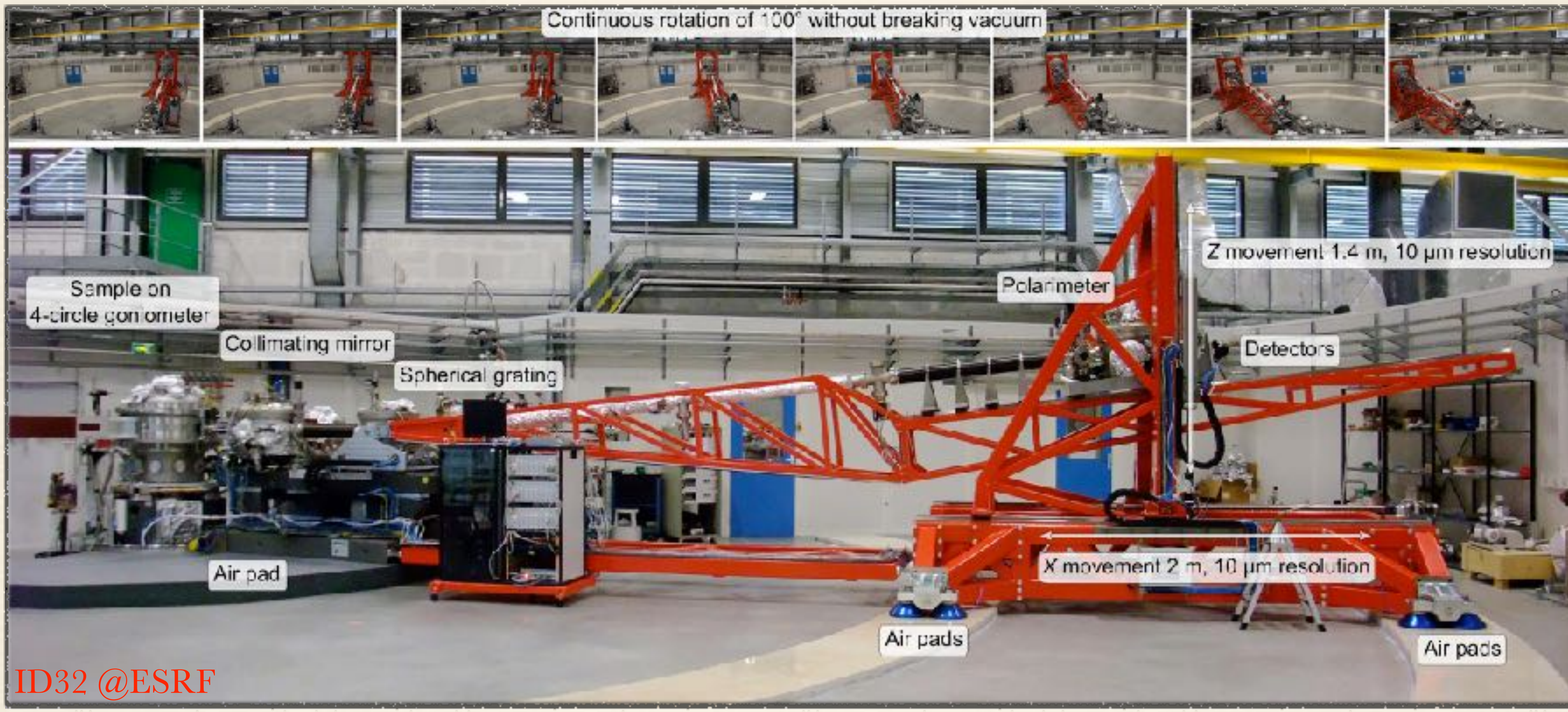
PRL 104, 077002 (2010)



Cu L-edge magnon

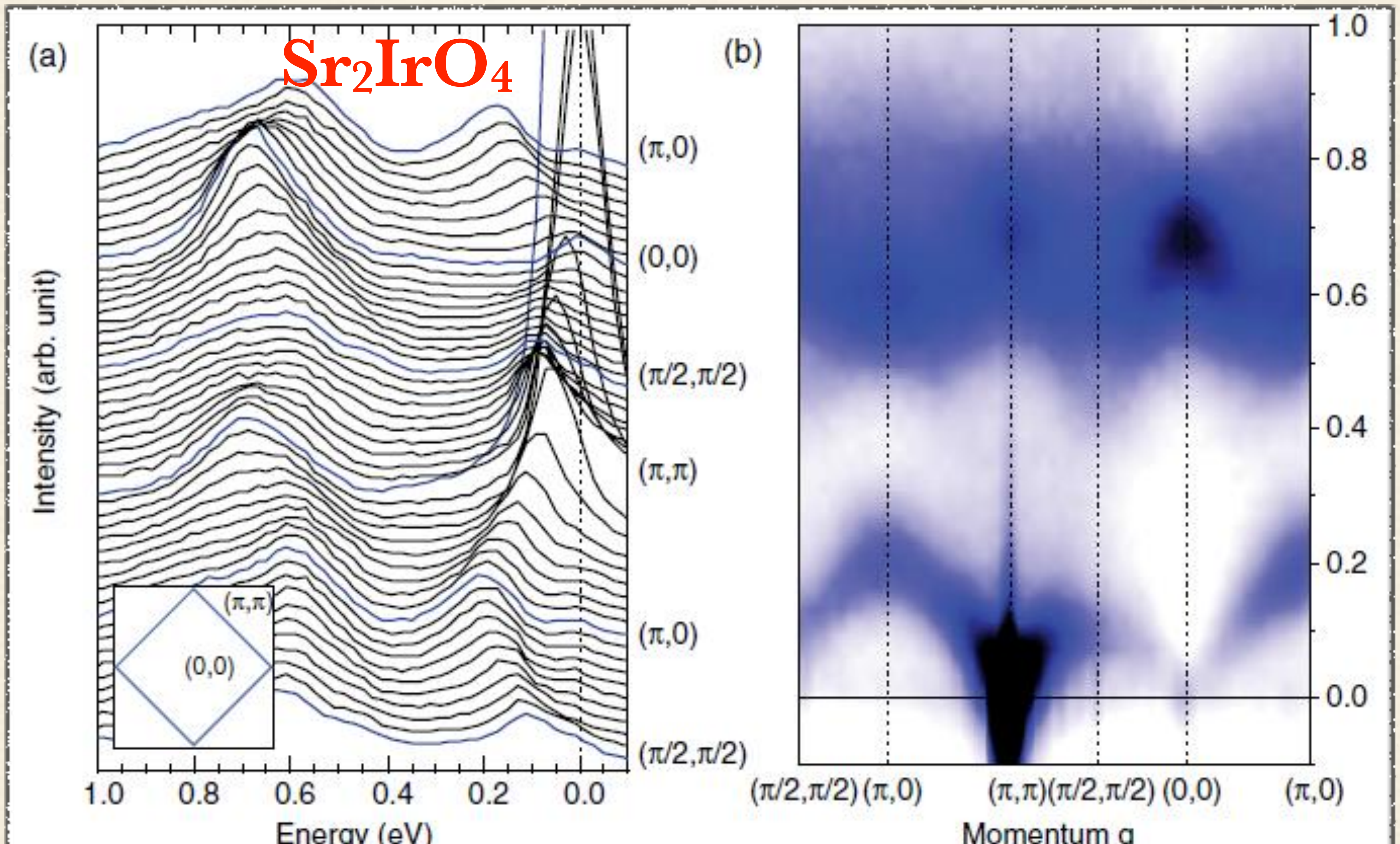
RIXS

Experimental Setup



RIXS

L-edge magnons and bimagnons

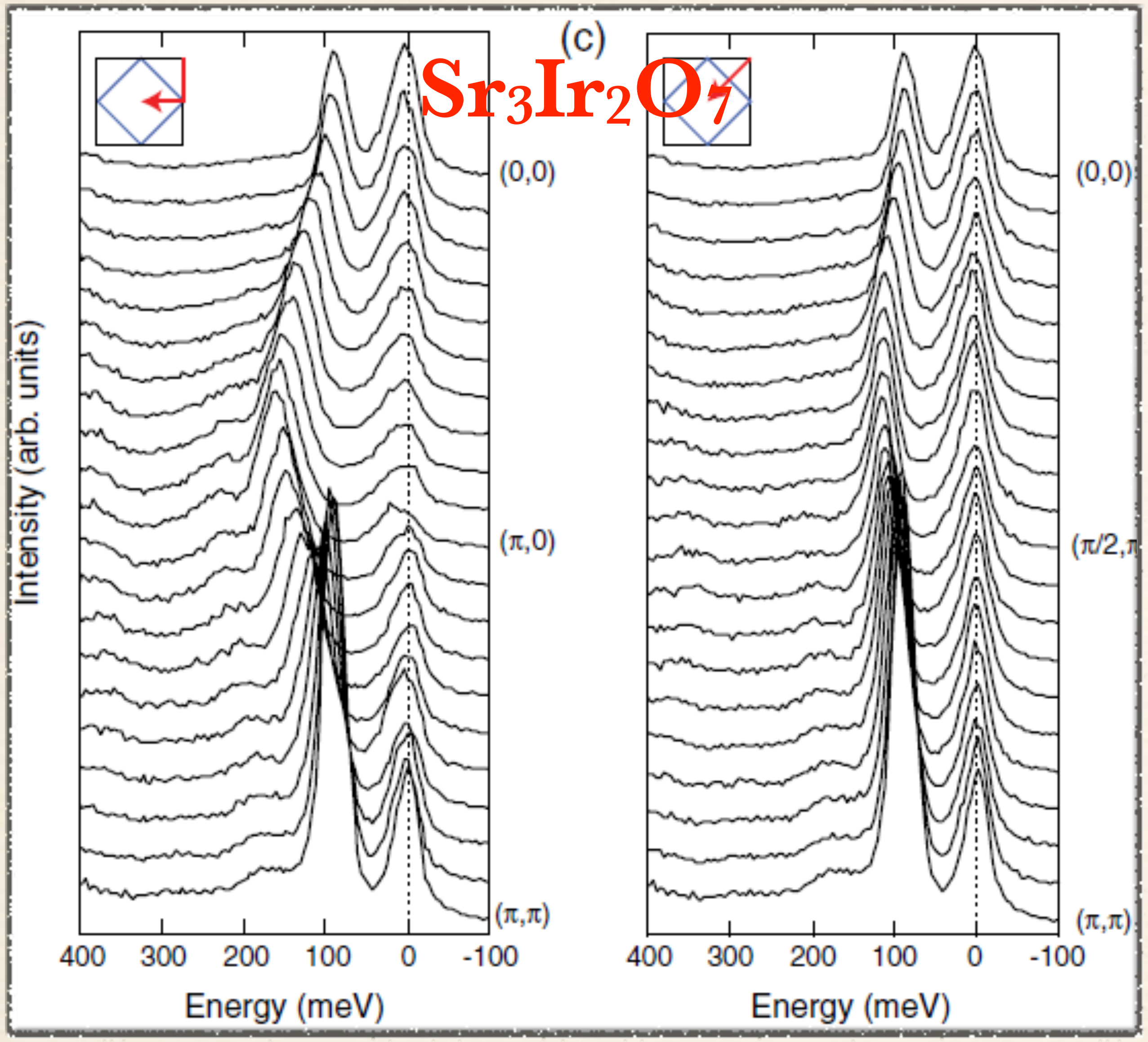


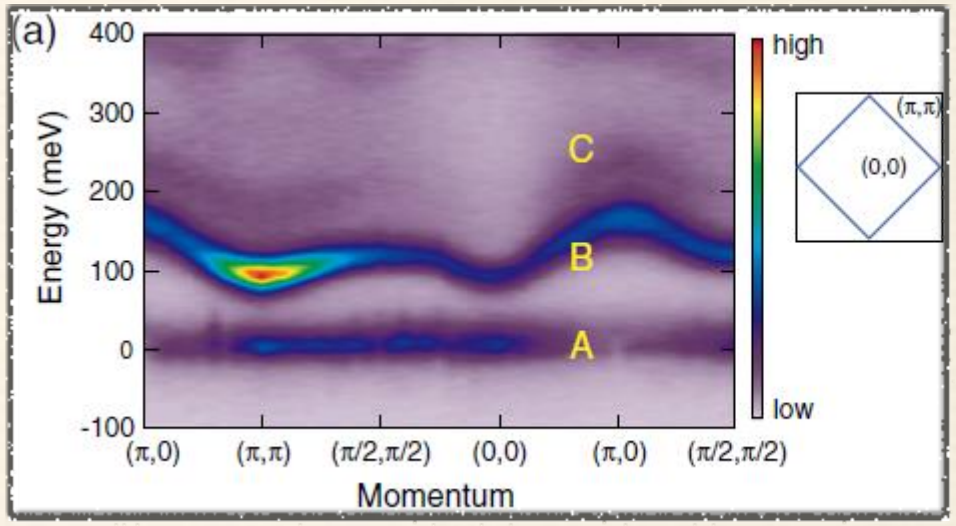
PRB 108, 177003 (2012)

L_3-edge Ir $5d^5$

RIXS

L-edge magnons



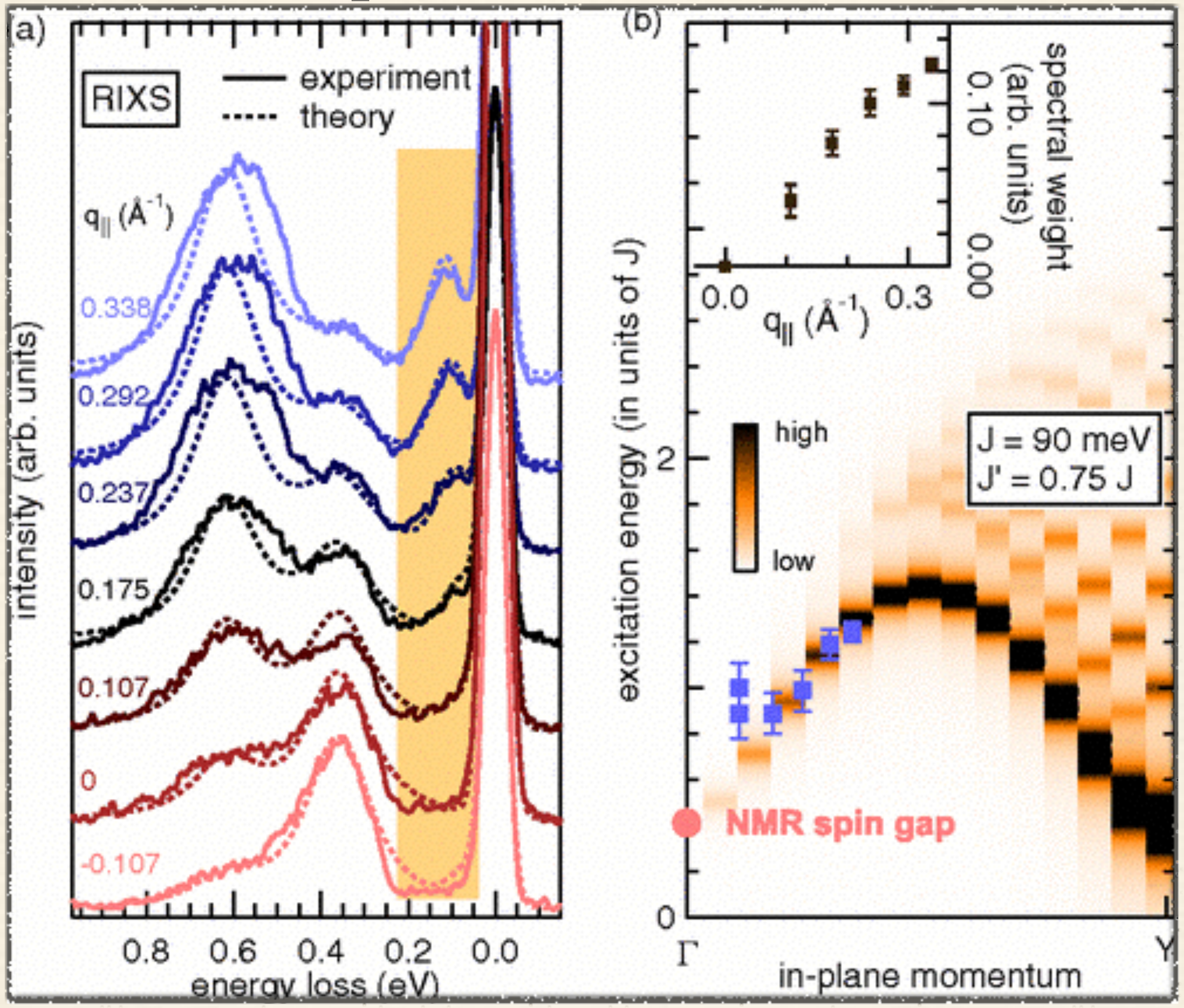


PRB 109, 157402 (2012)

L3-edge Ir

Resonant Inelastic X-ray Scattering

2 spinons excitation



PRL 107, 107402 (2017)

Spin-Peierls : determination of intra and inter-dimer J, J'

Ti L_3-edge (450eV)

RIXS

Summary

- Chemical/Element selectivity
- Resolution limited (~35meV)
- Half Brillouin Zone accessible (for low energy)
- Give access to :
  - two or bi - magnon if no spin-orbit coupled intermediate state (K-edge)
  - (large) spin-gap
  - single magnon (+multiple magnon) if spin-orbit coupling in the intermediate state (L, M-edge)
- Beyond this lecture : polarization, geometrical dependance (disentangle *dd* excitation and magnon…)

End of part II

(And there's nothing more…)

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End of part II

(And there's nothing more…)

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