REVEALING CHARGE (AND SPINS?) EXCITATIONS IN IRON-BASED SUPERCONDUCTORS USING RESONANT INELASTIC X-RAY SCATTERING

I. Jarrige[1,2]

Fe *L-edge / soft x-rays RIXS* :
Y. Harada[3], H. Niwa[3], C. Sakai[3]

Fe *K-edge / hard x-rays RIXS* :
K. Ishii[1,2], H. Gretarsson[4], Y.-J. Kim[4], J. Kim[5], M. Upton[5], D. Casa[5], T. Gog[5], J.P. Hill[6], X. Liu[6], T. Fukuda[1,2], M. Yoshida[1], N. Hiraoka[7], K.D. Tsuei[7], M. Ishikado[1,2], S. Shamoto[1,2]

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RIXS On The Fe Superconductors: A Slow Start…

- The iron-based superconductors are one of the top topics in recent science:
  - #1 most cited publication in 2008 (Y. Kamihara *et al.*, JACS 130, 3296)
  - #6 top topics of Thomson Reuters' 2009 compilation
- Still a lot of work ahead, superconductivity mechanism not fully understood yet.

- Only *three* RIXS studies published so far (metals are no good for RIXS!) :
  - W.L. Yang *et al.*, Phys. Rev. B 80, 014508 (2009) *No RIXS signal*
  - J.N. Hancock *et al.*, Phys. Rev. B 82, 020513(R) (2010) *No RIXS signal*
  - I. Jarrige *et al.*, Physica C 470, S377 (2010) *Very weak RIXS signal*

- Goal: Try with higher resolution, better statistics, and lot of patience.

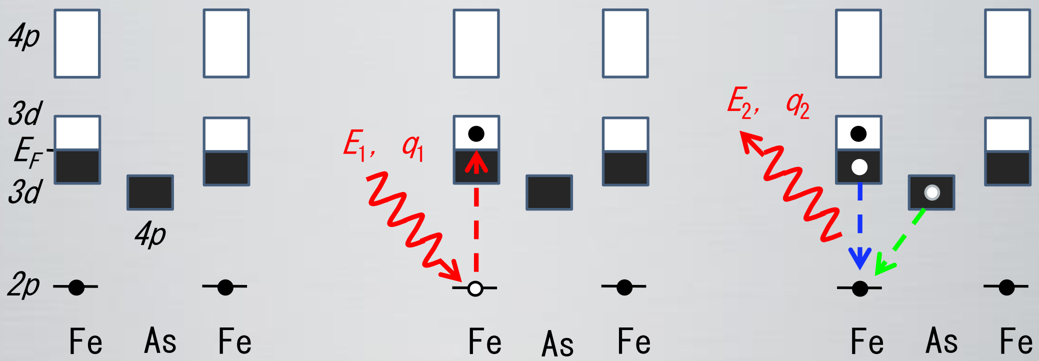
… But A Strong Finish?

What Is RIXS?

• *L*-edge – *direct* RIXS :

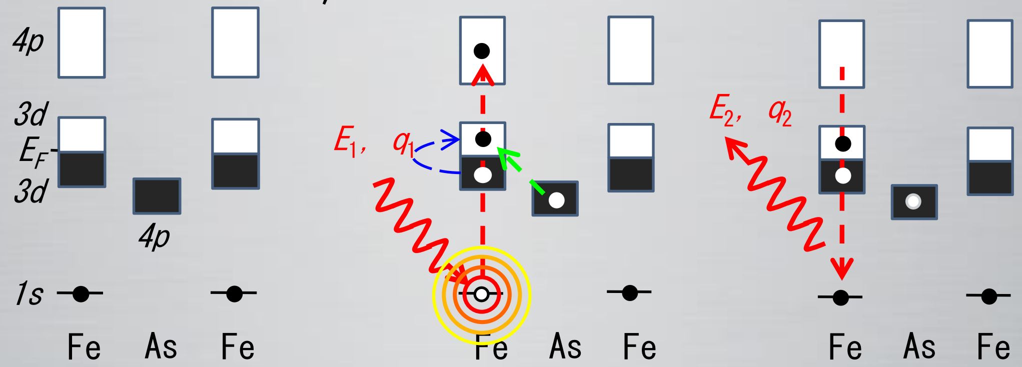
$$F^{direct}_{fg} = \langle f \,|\, D^{\dagger} G_0 D \,|\, g \rangle$$

|Initial State > |Intermediate State> |Final State>



• *K*-edge – *indirect* RIXS :

$$F^{indirect}_{fg} = \langle f \,|\, D^{\dagger} G_0 H_C G D \,|\, g \rangle$$



Excitation energy = $E_1 - E_2$

Why Two Edges For One Study?

K edge

(indirect)

- ⊙ Larger momentum range
- ⊙ Constrained environments

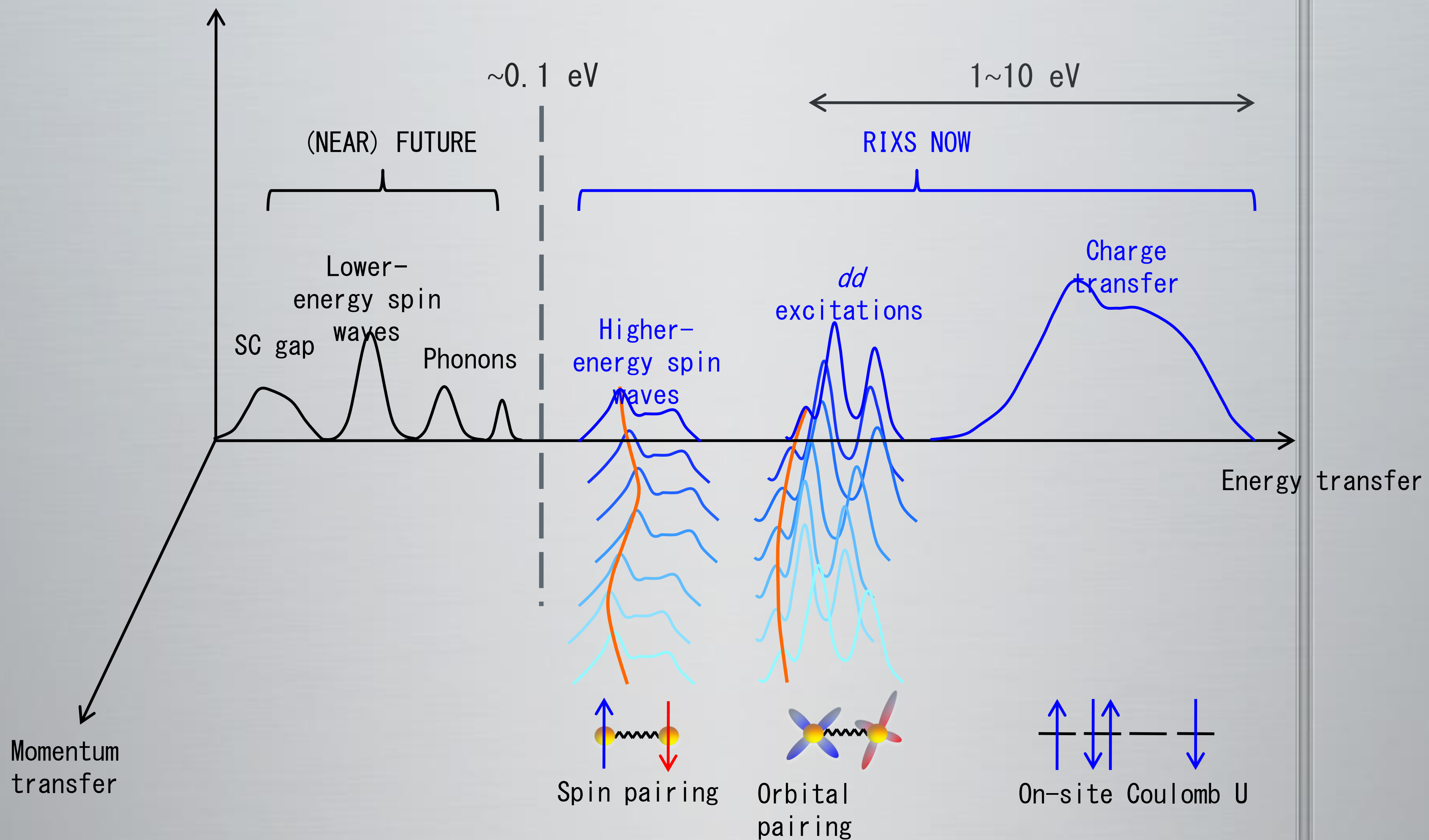
L edge

(direct)

- ⊙ Stronger inelastic signal
- ⊙ Weaker elastic line
- ⊙ Single spin-flip excitation
- ⊙ Higher E/ΔE than *K* edge for Fe

- Excellent complementarity
- The combined use of both edges enables an in-depth diagnosis of the electronic and magnetic properties

Why RIXS ?



- RIXS = spectroscopic + scattering techniques combined together.
- Information about the type of glue for superconducting electrons.
- Quantitative estimation of the on-site Coulomb interaction and Hund coupling.

EXPERIMENTAL: BEAMLINES

K edge

Fe-K : E=7120 eV, ΔE=230 meV

Beamline : 30-ID
Endstation : **MERIX**
(1-m Ge(620))

L edge

Fe-L_3 : E=710 eV, ΔE=160 meV

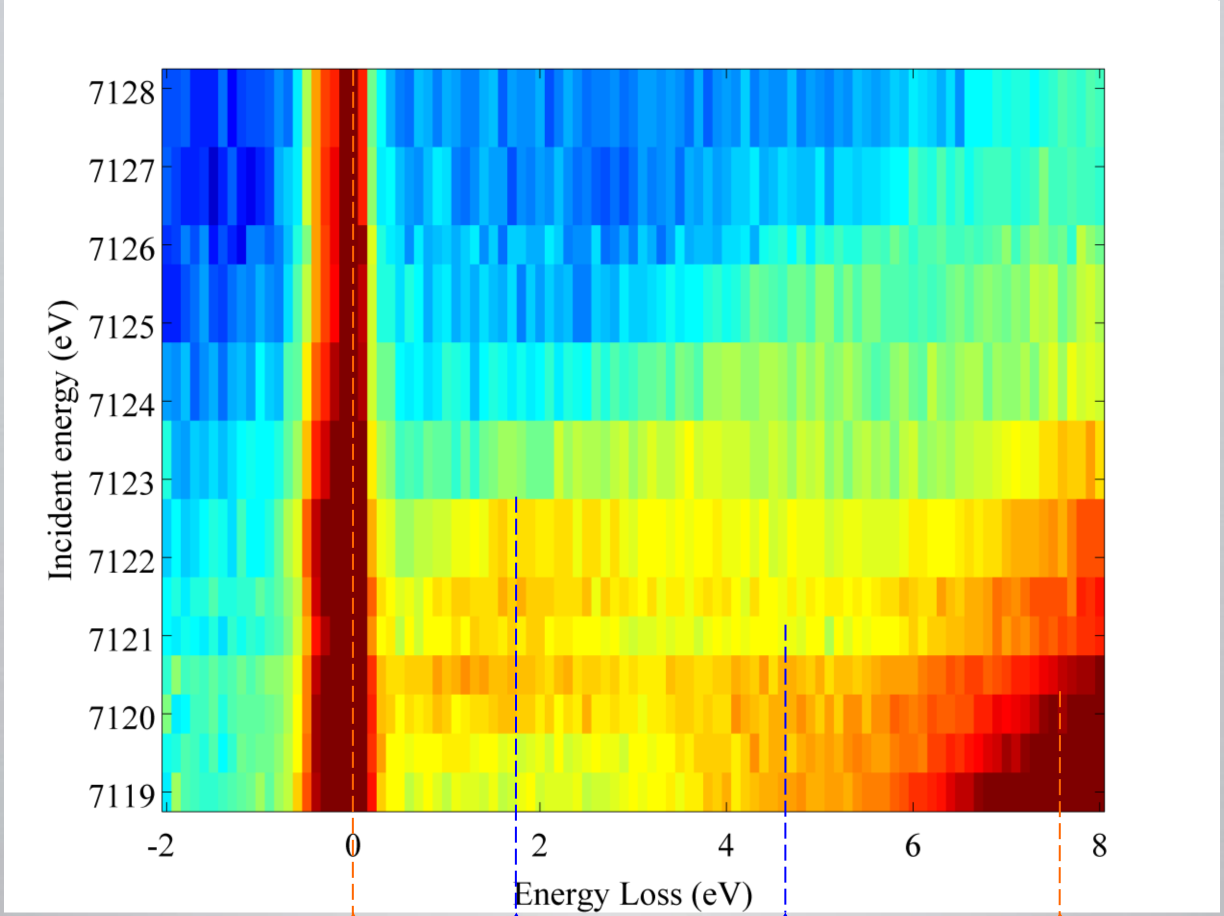
Beamline : BL07SU
Endstation : **HORNET**
(Sample - detector: 3m)

K edge

Incident Energy Dependence

PrFeAsO

- Log (Intensity)
- RIXS signal very weak!



Elastic

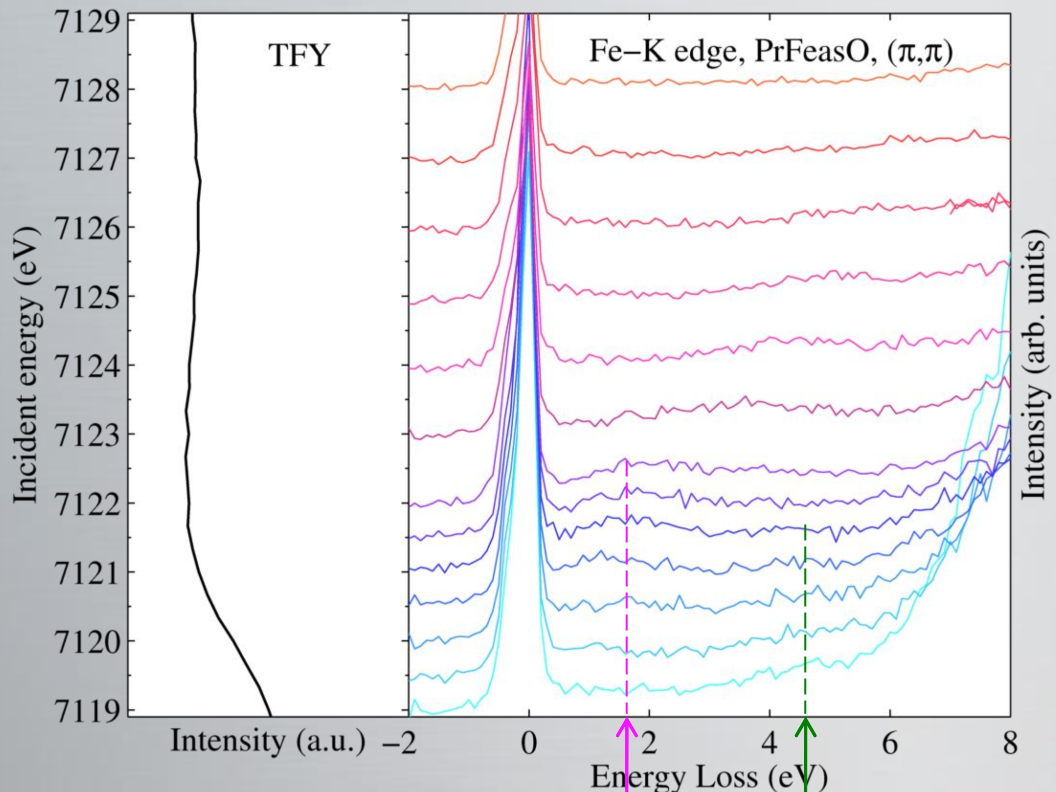
Inelastic ~ 0.1 cps
(nickelates, cuprates: up to 100 cps)

Fluorescence

K **edge**

PrFeAsO

Incident Energy Dependence



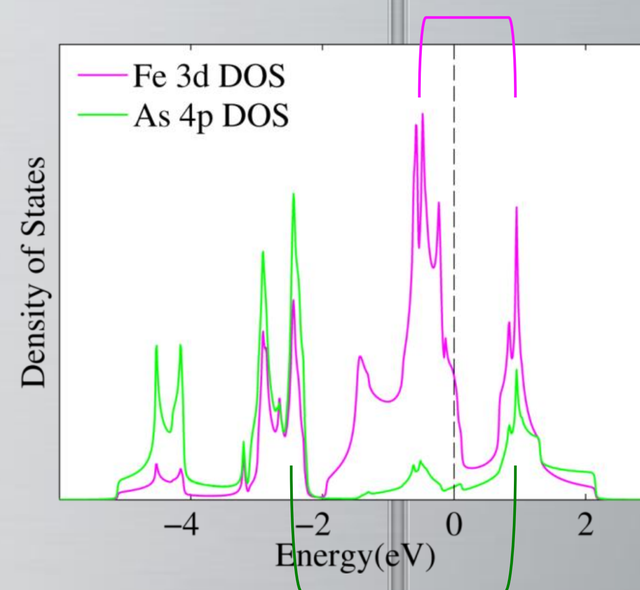
dd interband

Charge transfer (CT)

Density Of States

Calculated by Y. Yamakawa *et al.*

Fe*3d*–Fe*3d*



As*4p*–Fe*3d*

- ***dd* interband** : Fe *dd* interband excitation.
- **Charge tranfer**: Fe *3d*–As *4p* charge transfer.

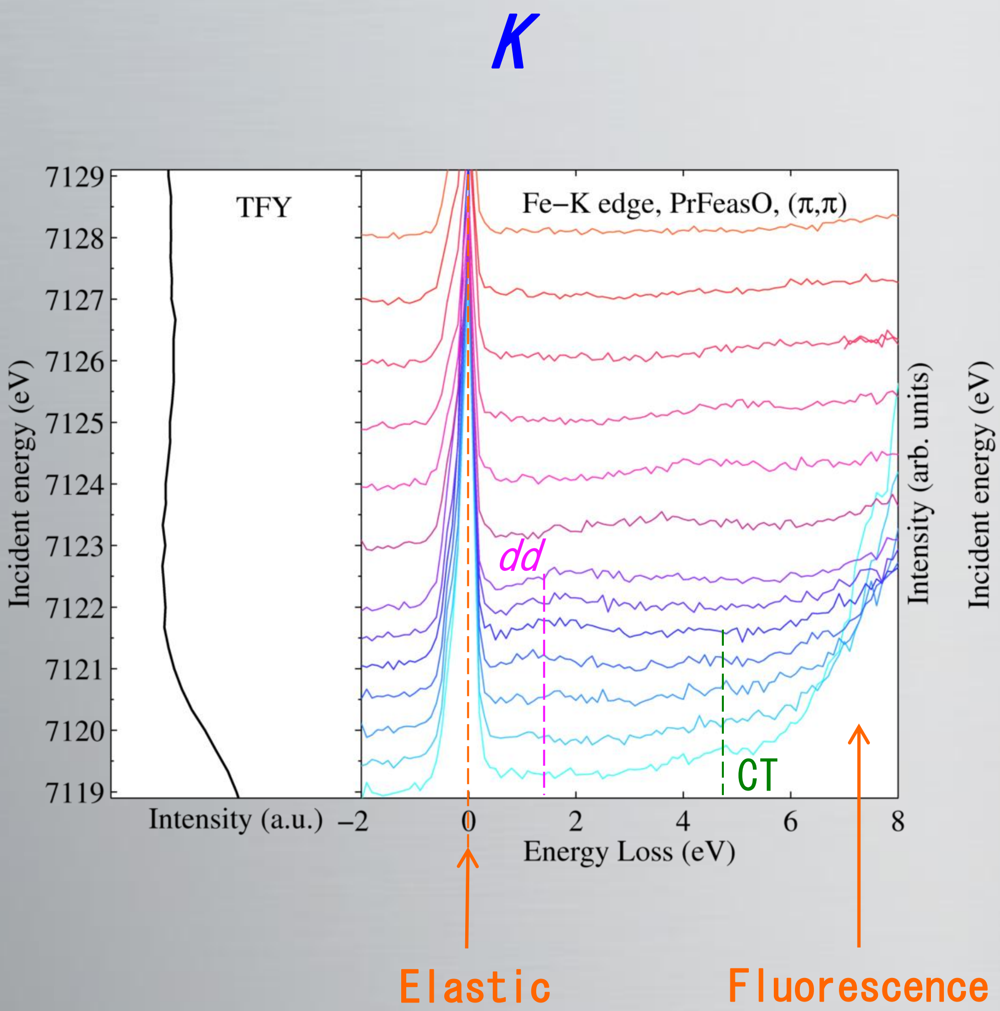
K edge

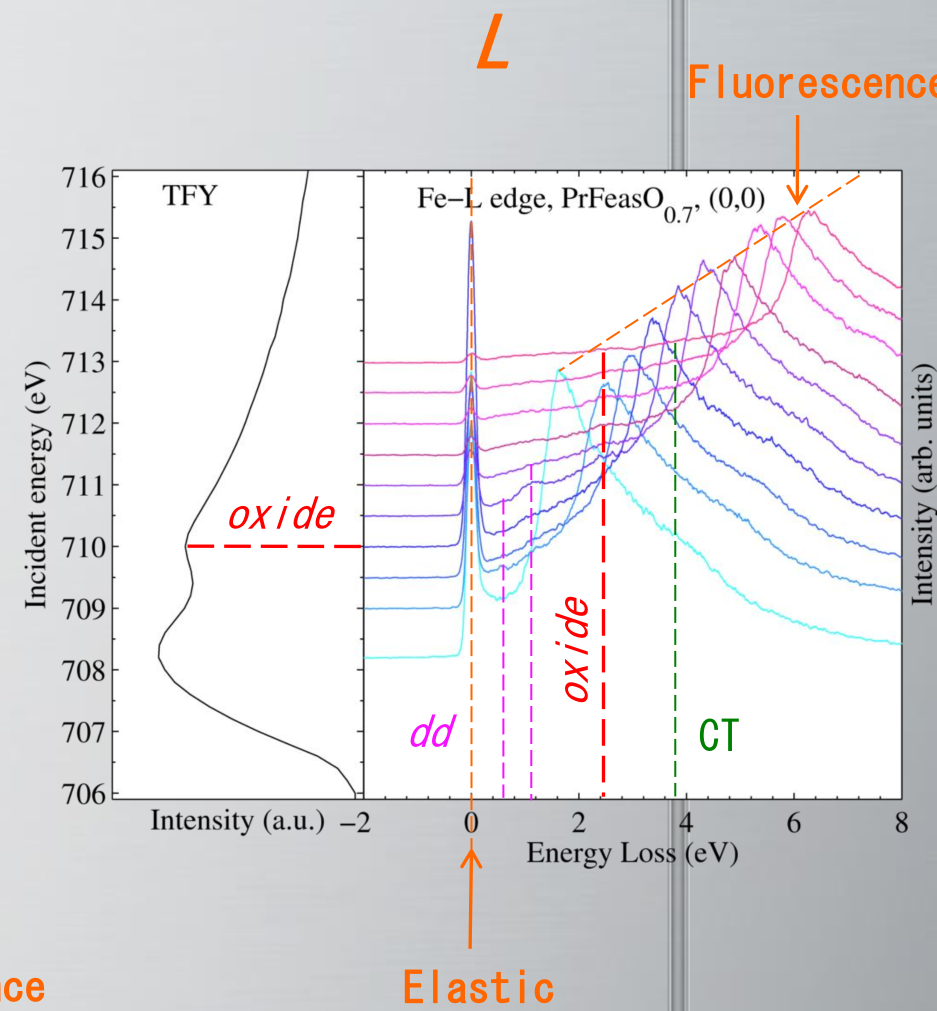
PrFeAsO

INCIDENT ENERGY DEPENDENCE

L edge

PrFeAsO$_{0.7}$





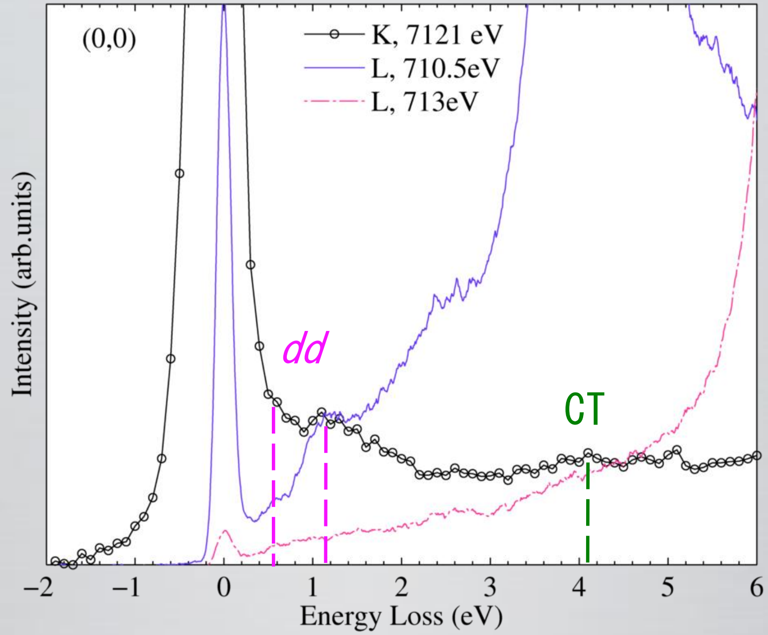
***K* edge**

PrFeAsO

INCIDENT ENERGY DEPENDENCE

***L* edge**

$PrFeAsO_{0.7}$



- Excitations appear at approximately the same energies for both *K*-edge and *L*-edge
- Better selectivity of the intermediate state at *L* edge as a function of Ei

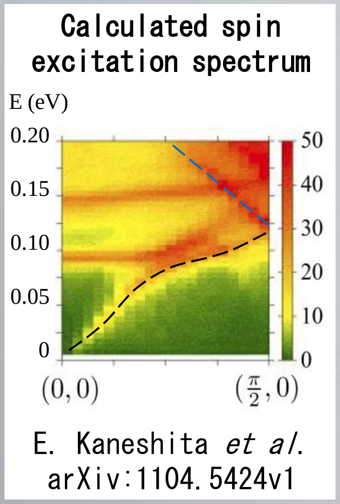
Edge	*K*	*L*
Acquisition time (h)	4	1
ΔE (meV)	230	160
Elastic intensity (a.u.)	7	1

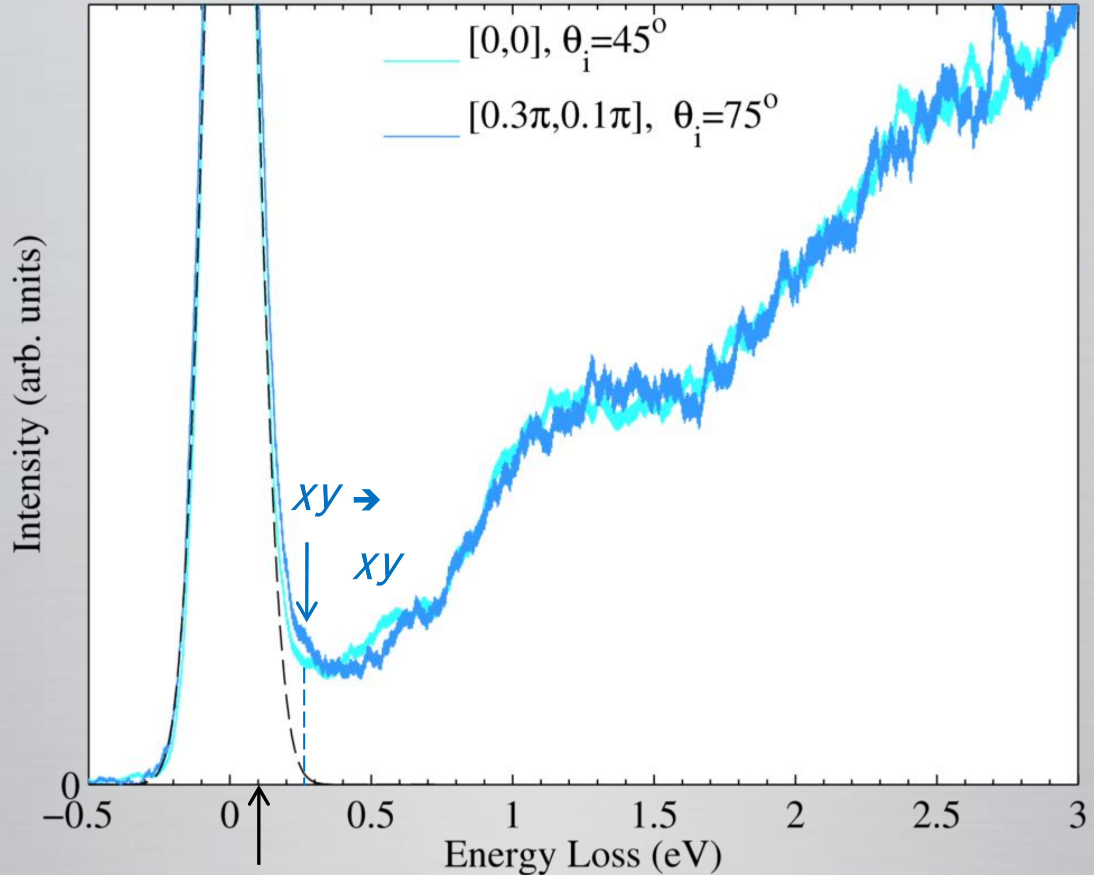
∴ ANY SPIN EXCITATION?

L edge

PrFeAsO$_{0.7}$

Spin Excitations ?





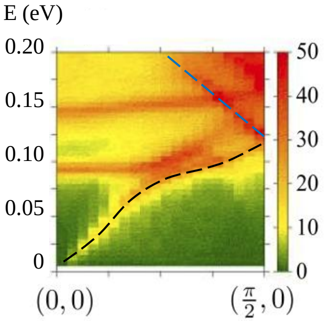
- Low-energy (~0.25 eV) excitation enhanced with in-plane momentum contribution: assigned to *xy* → *xy* excitation.
- **Spin excitation** predicted to reach 0.1 eV at (0.5π, 0)

L edge

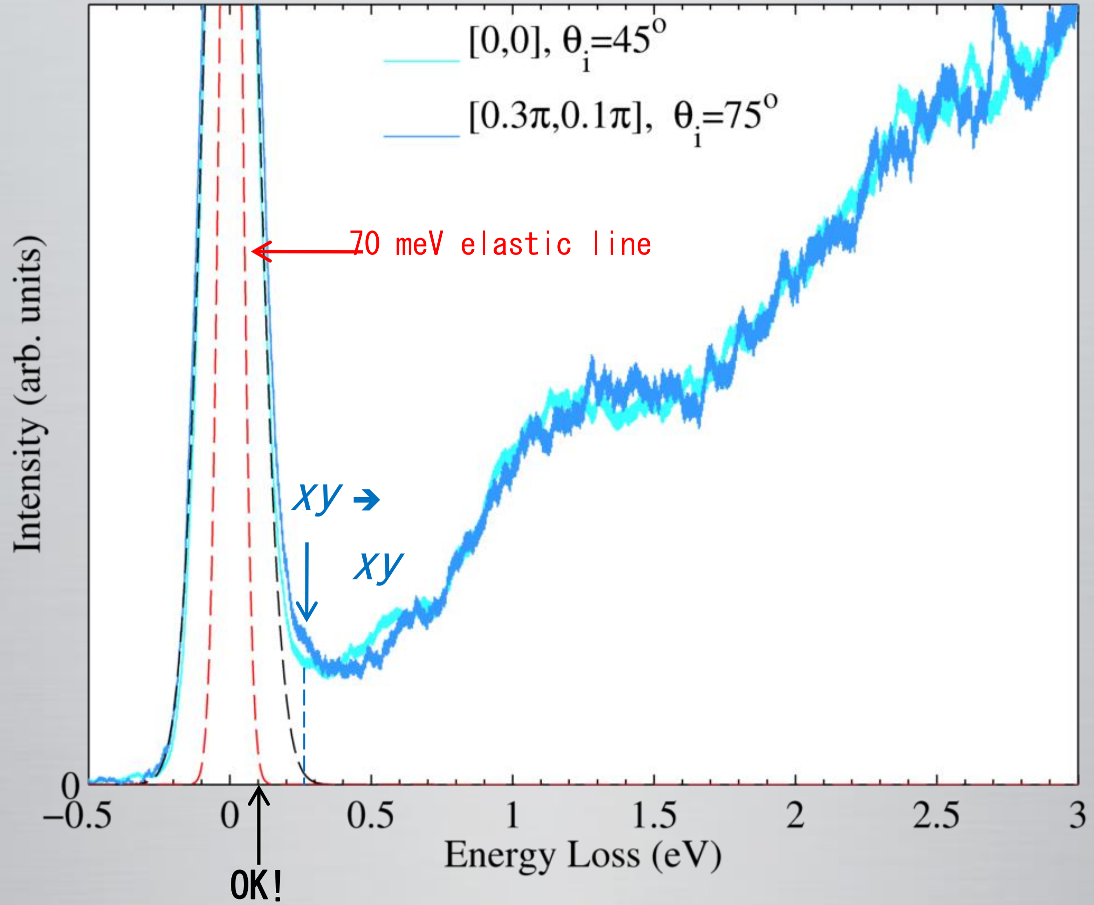
$PrFeAsO_{0.7}$

Spin excitations ?

Calculated spin excitation spectrum



E. Kaneshita *et al.* arXiv:1104.5424v1



- Low-energy (~0.25 eV) excitation enhanced with in-plane momentum contribution: assigned to *xy* ➔ *xy* excitation.
- **Spin excitation** predicted to reach 0.1 eV at (0.5π,0), should be observable with ΔE≈70 meV (E/ΔE=10000).

L edge

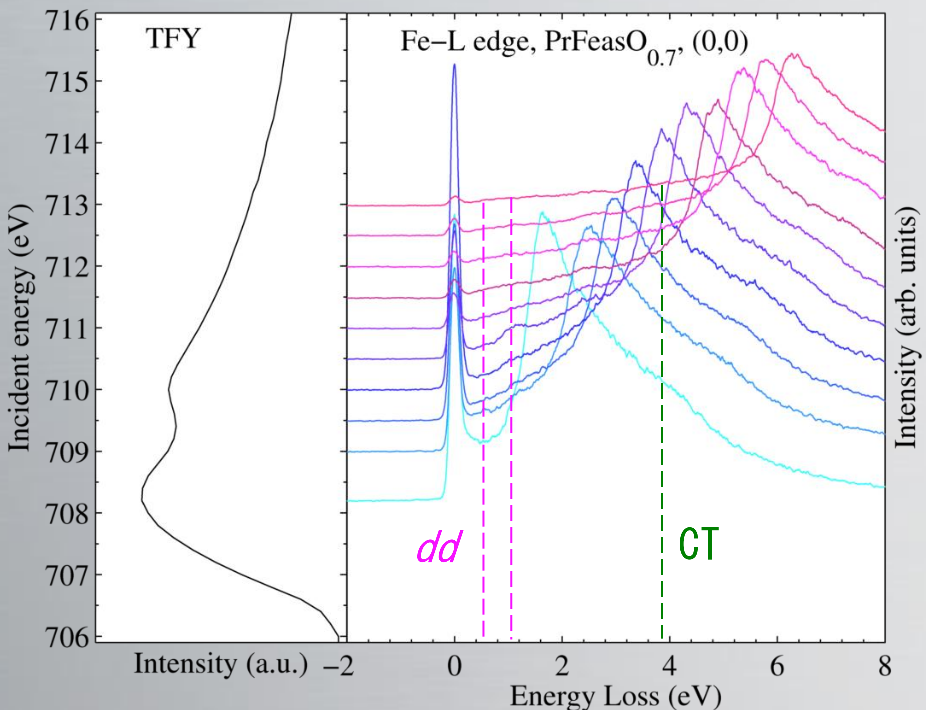
PrFeAsO$_{0.7}$

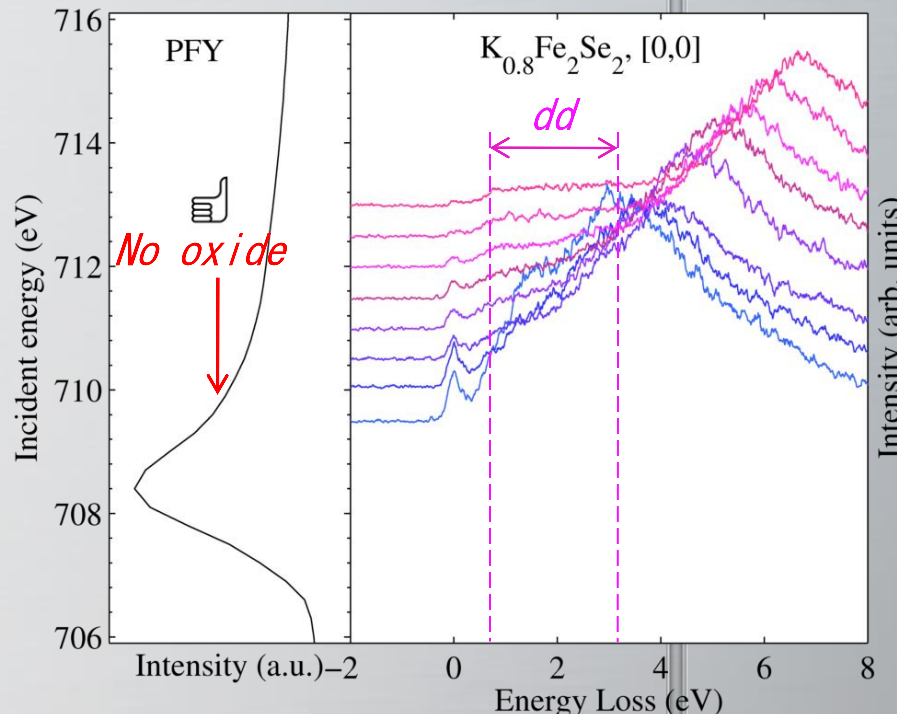
INCIDENT ENERGY DEPENDENCE

L edge

KFe$_{1.8}$Se$_2$

-Flat surface > **Weaker elastic line**
- Large magnetic moment > **High-energy magnons**



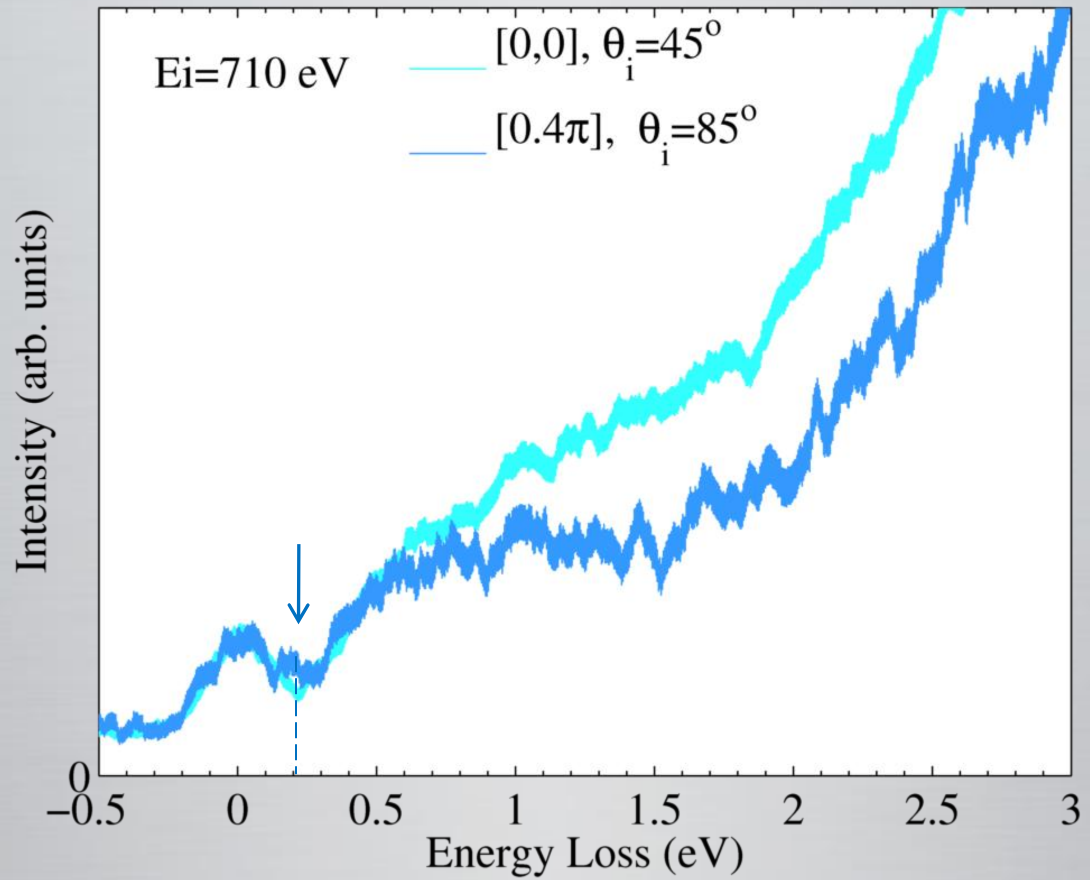


- 1h per scan for PrFeAsO$_{0.7}$, 30 min per scan and weaker flux for K$_{0.8}$Fe$_2$Se$_2$
- Weaker elastic line and stronger Raman spectral weight for K$_{0.8}$Fe$_2$Se$_2$, but Raman features not as sharp

Spin Excitations?

L edge

$KFe_{1.8}Se_2$



- Low-energy (~0.2 eV) excitation enhanced with in-plane momentum contribution: Spin excitation? ➔ Perhaps, but need to measure other momenta + increase statistics (spectrum shown above is 3h measurement time)

Conclusion

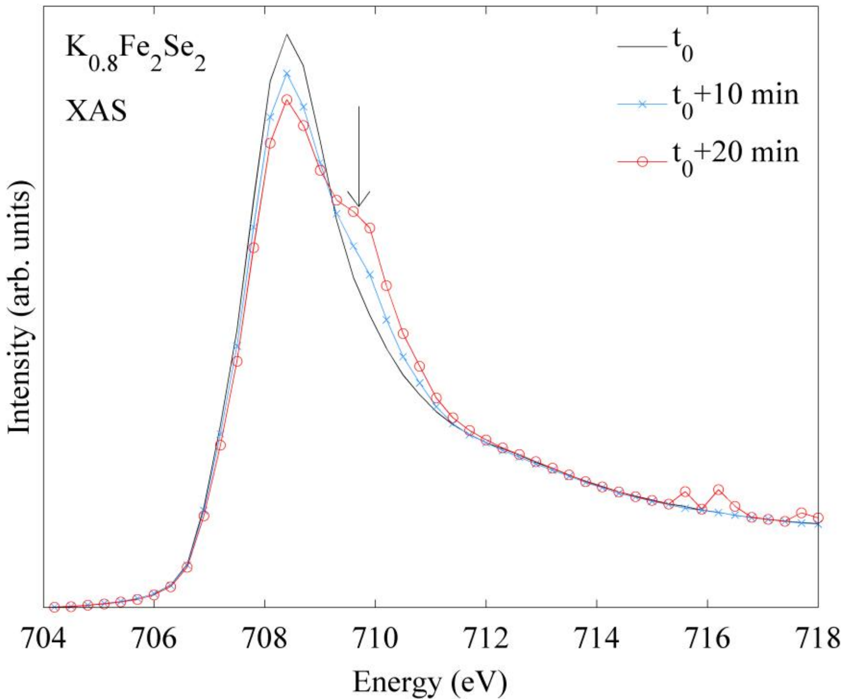
- First observation of charge excitations using RIXS in an iron-based superconductor

- *K*-egde: Comparison with theory yields U=2.4 eV for PrFeAsO

- *L*-egde: Higher energy resolution, stronger Raman signal, and weaker elastic line compared with *K*-edge, but needs to be cautious with surface oxidation. Spin excitations can be observed, although with more flux or longer acquisition time.

- Wish list for HORNET:
  - In-situ manipulator for sample cleaving
  - Low-temperature
  - More flux… or more beamtime?

Time Dependence



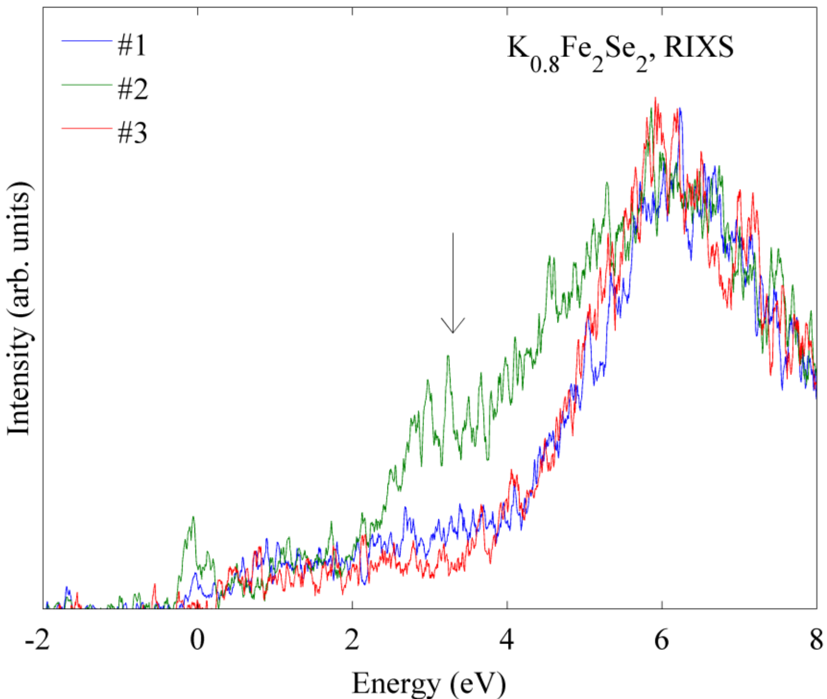


Figure 3: Time dependence of the Fe-*L* XAS spectrum measured on $K_{0.8}Fe_2Se_2$ in the fluorescence yield mode. The sample position is fixed.

Figure 4: Time dependence of the Fe RIXS spectrum measured on $K_{0.8}Fe_2Se_2$ for E_i=712.5 eV. The sample position was continually scanned for scans #1 and #3, and fixed for the first 10 minutes of scan #2 (one scan measurement time = 30 minutes).

K edge

PrFeAsO

ELECTRON CORRELATION EFFECTS

Quantitative estimation of U!

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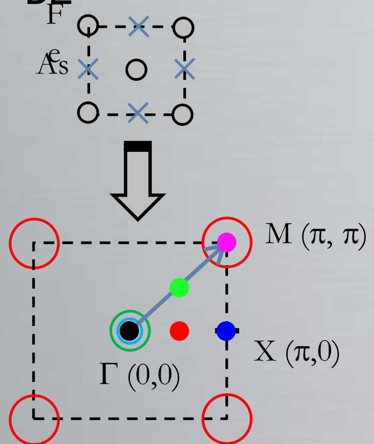

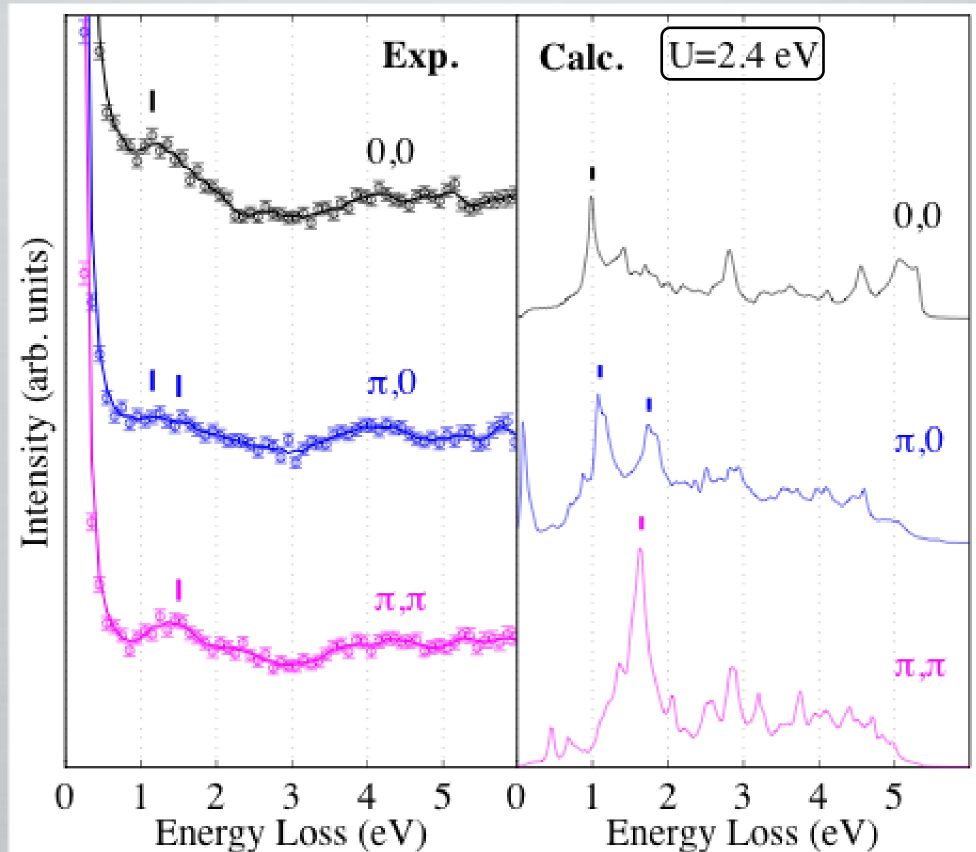

- Comparison with calculations based on a **16 band** ***dp*** **model** for the **AF ground state**
- **Good agreement** with experiment for **U=2.4 eV**

K edge

PrFeAsO

MOMENTUM DEPENDENCE

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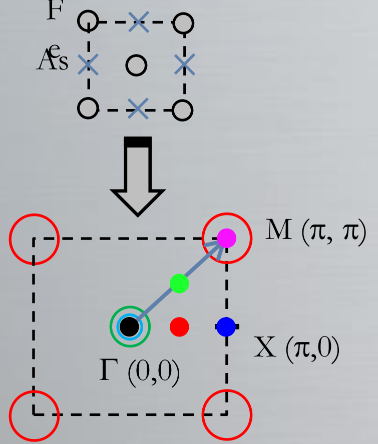

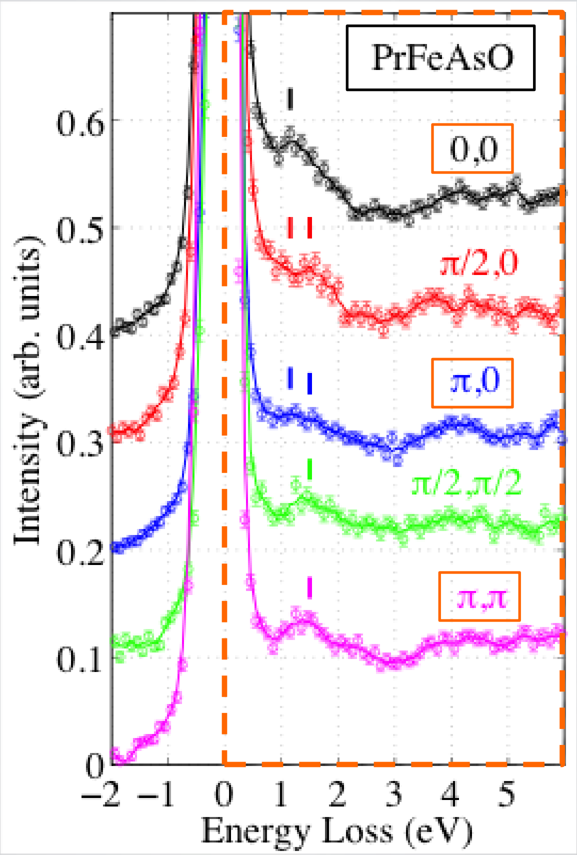

- Weak, but sizeable **momentum** dependence