High spectral and focal resolution RIXS with state-of-the-art flat crystal spectrometer

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Argonne NATIONAL LABORATORY

U.S. DEPARTMENT OF ENERGY

Acknowledgments

- **APS**

Sector 27 RIXS

D. Casa, T. Gog

Analyzer

A. Said

X-ray optics

X. Huang

High Pressure

Wenli Bi

- CEIT, CZECHIA

Jiri Chaloupka

- George Mason Univ.

N. J. Ghirmire

- POSTECH, KOREA

B. J. Kim

Jinkwang Kim

- IISERM, INDIA

Yogesh Singh

OUTLINE

- ✓ Post-Sample-Collimation flat crystal RIXS analyzer
- ✓ High spectral resolution

A. Ir L_3 (11.215 keV)

10 meV energy resolution with scattered polarization analysis

5.7 meV energy resolution

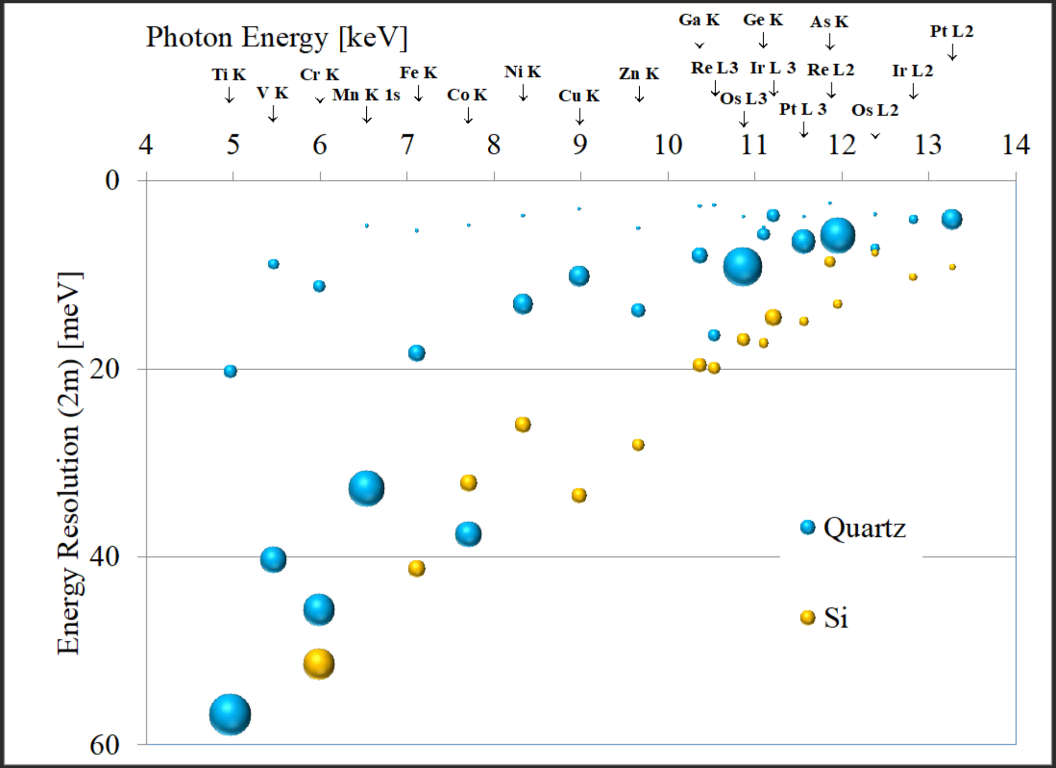
B. Ta L_2 (11.136 keV)

12.5 meV energy resolution

- ✓ Focal resolution

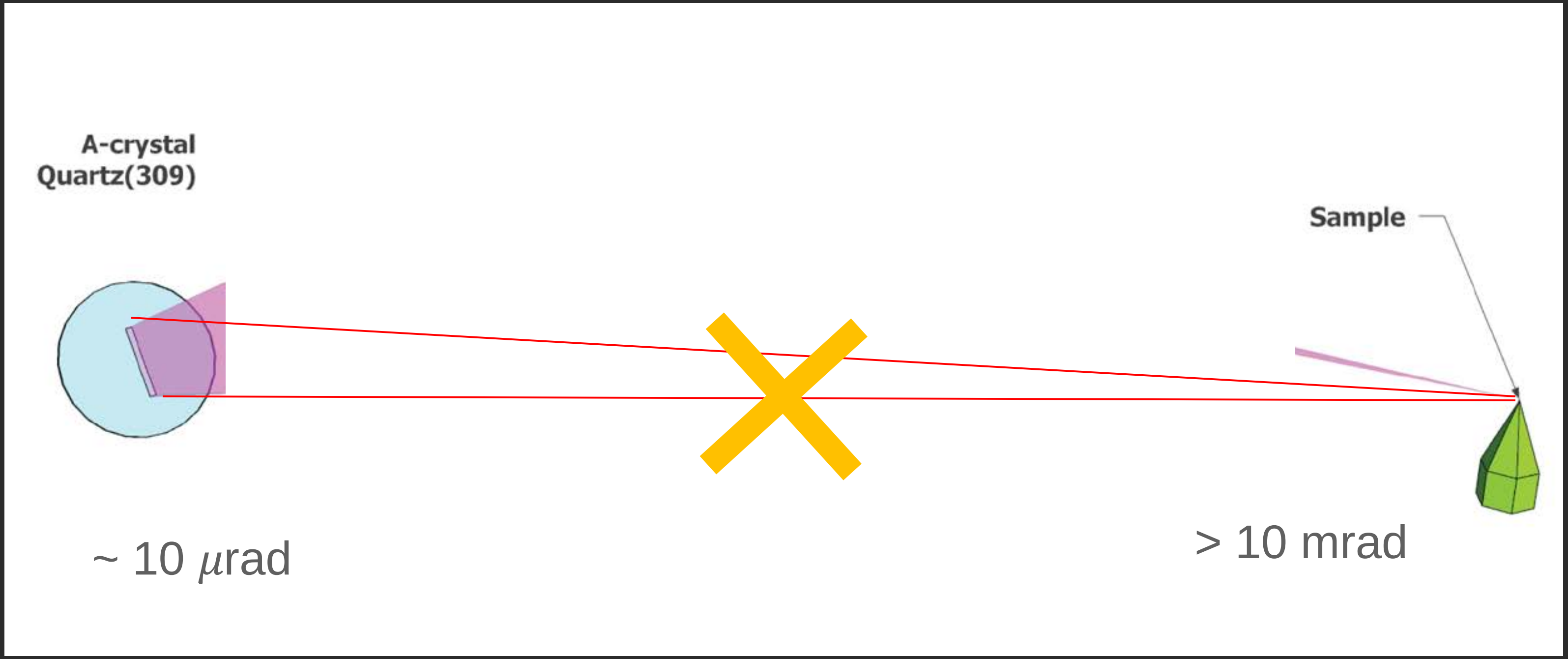
C. High pressure application using the confocal nature

Analyzer crystal choice for sub 10 meV : Quartz



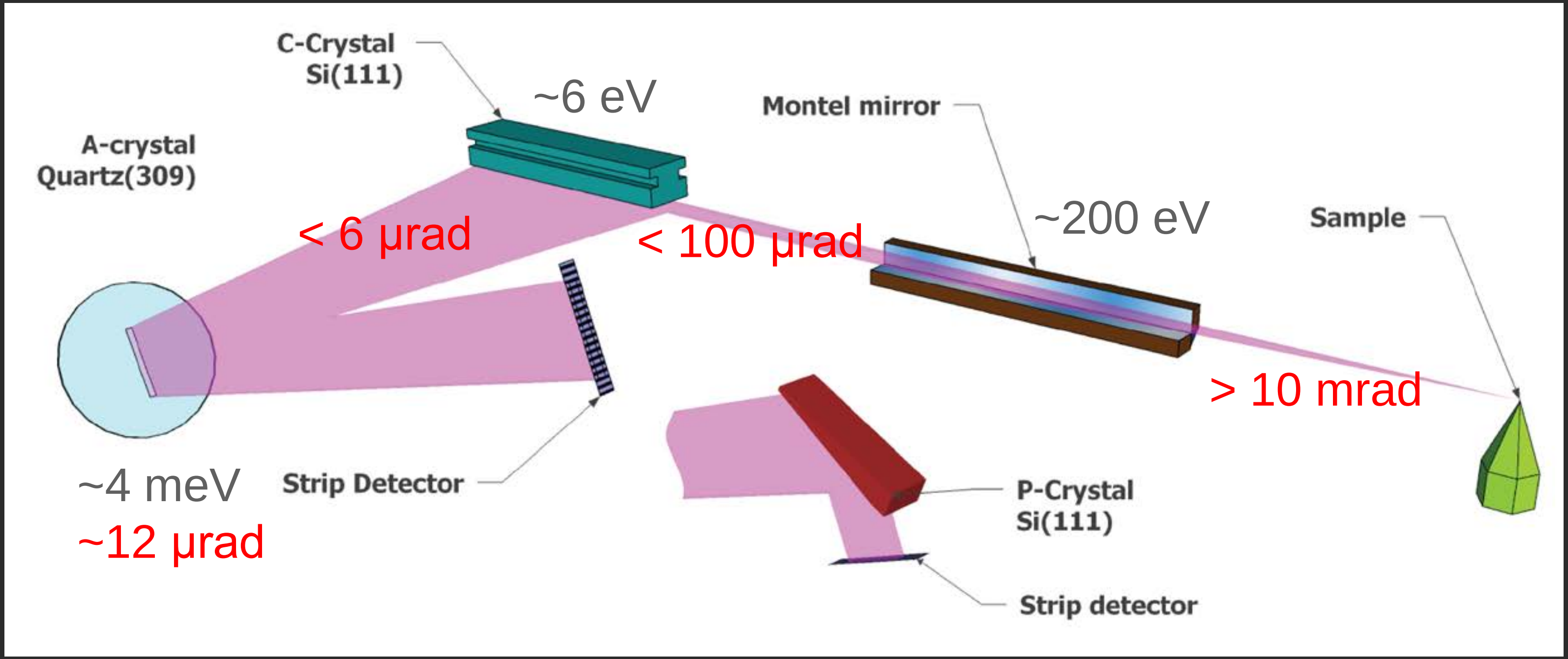
4 meV at 10 keV: J. P. Sutter et al., J. Synchrotron Rad. 13, 278-280 (2006)
1.14 meV at 14.4 keV (Mössbauer): Y. Imai et al., AIP Conference Proceedings 879, 930 (2007)
T. Gog et al., J. Synchrotron Rad. 20, 74-79 (2013)
X.-R. Huang et al., J. Appl. Cryst. 51, 140-147 (2018)

This is the simplest.



Post-sample-collimation

: Large enough collecting solid angle
: Smaller divergence than the analyzer acceptance angle.



Pros: no extrinsic contributions to the energy resolution other than analyzer crystal, high efficiency scattered x-ray polarization analysis

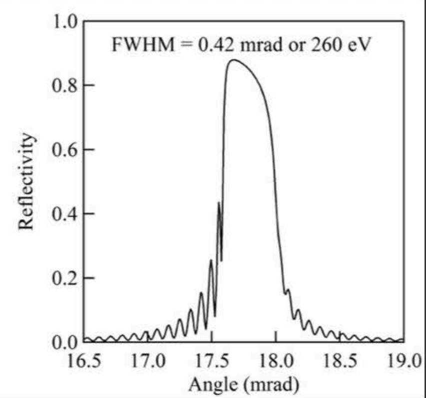
Cons: complicated, larger detector noise due to large beam

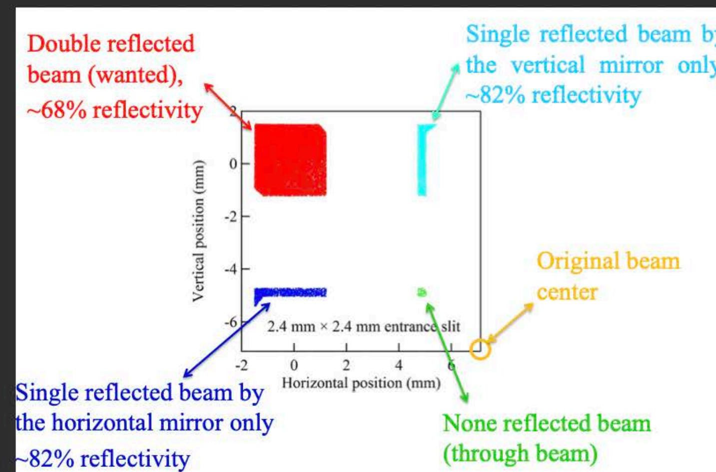
Sub-meV IXS:Y. Shvyd'ko et al., Nat. Communs. **5**, 4219 (2014)
NSLS-II IXS beamline: A. Suvorov et al., JSR, **21**, 473-478 (2014)

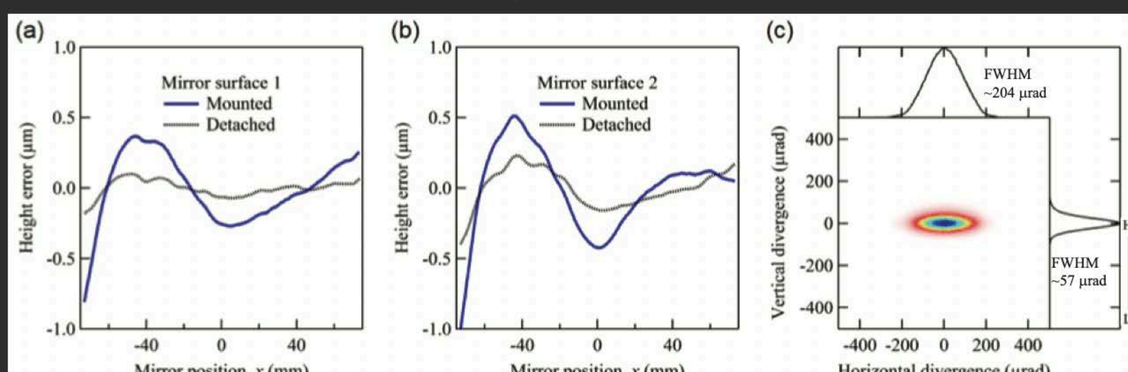
Montel mirror

J. Kim et al.,
J. Synchrotron Rad. (2016). 23, 880-886

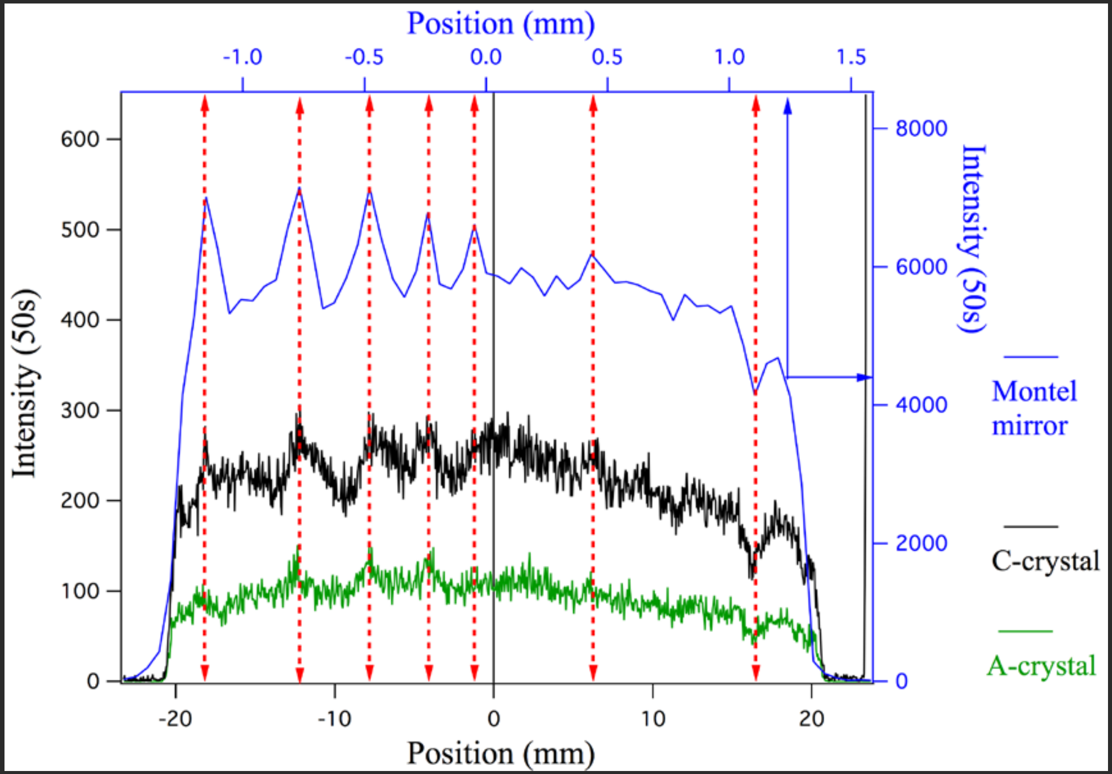
ML (parabolic) parameters	
sample-to-mirror *p* (mm)	200
center grazing angle, θ (mrad)	17.82
mirror length (mm)	150
energy (eV)	11215
parabolic parameter *a* (mm)	0.127
material (heavy)	Ru
material (light)	C
gamma ratio Γ	0.4
ML d spacing, Λ (nm)	3.20







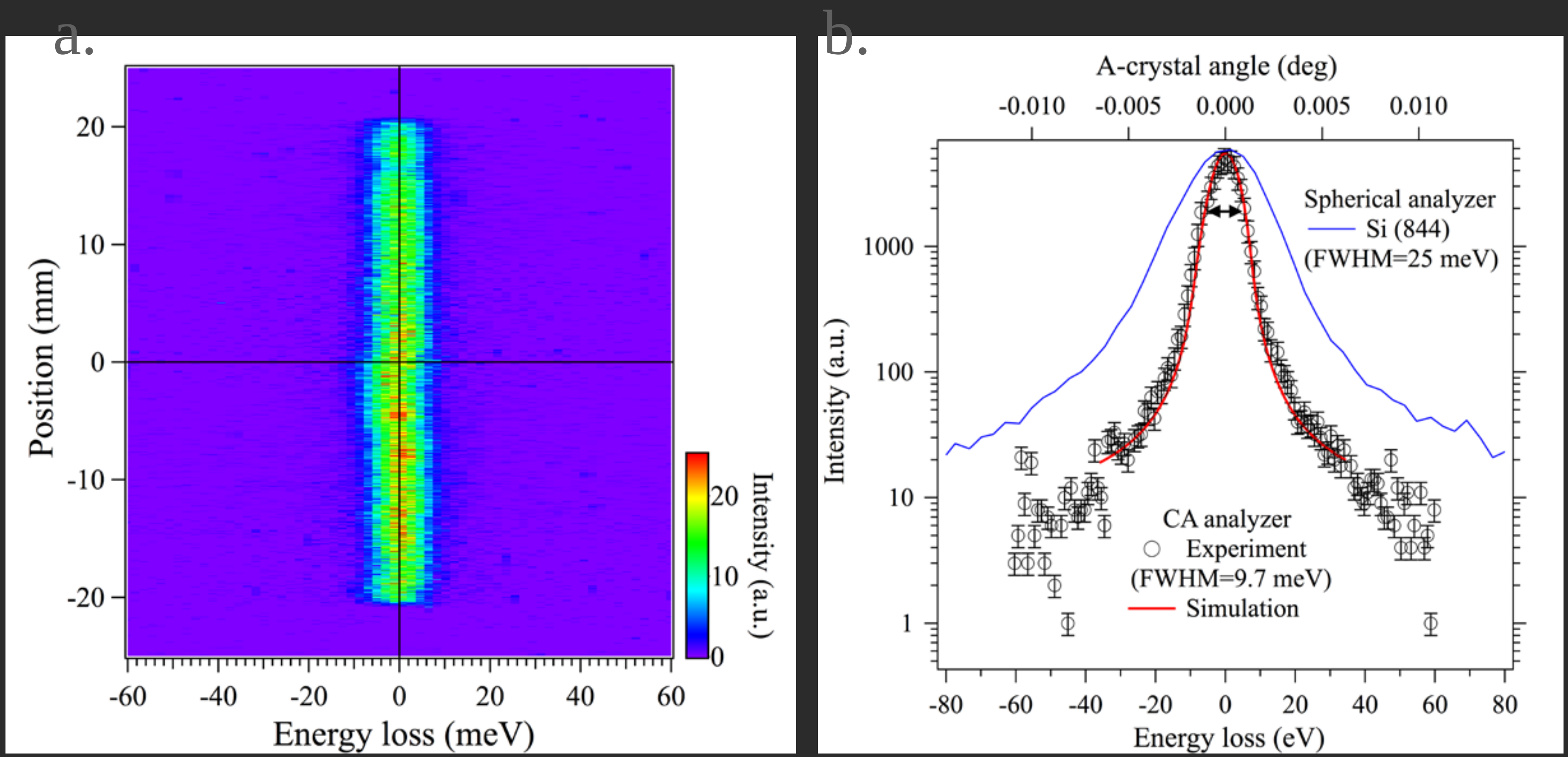
Collimated beam profile



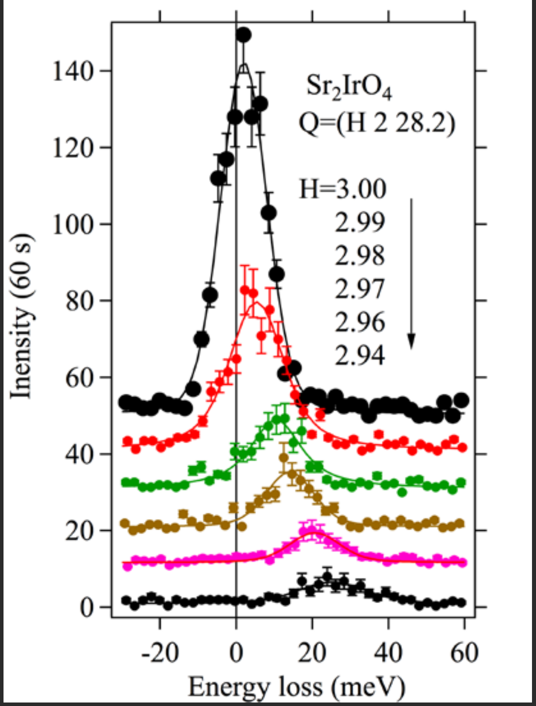
J. Kim et al., Scientific Reports 8, 1958 (2018)

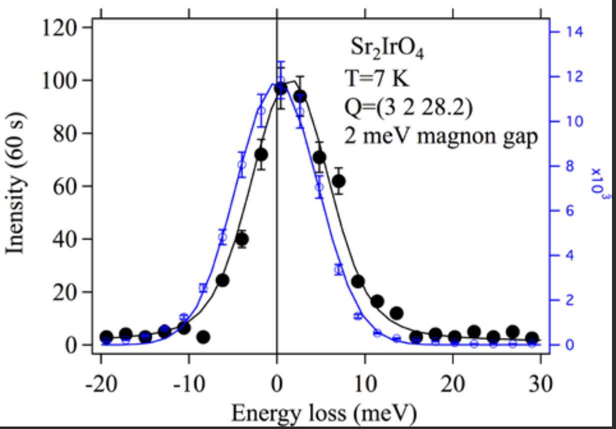
Resolution function :

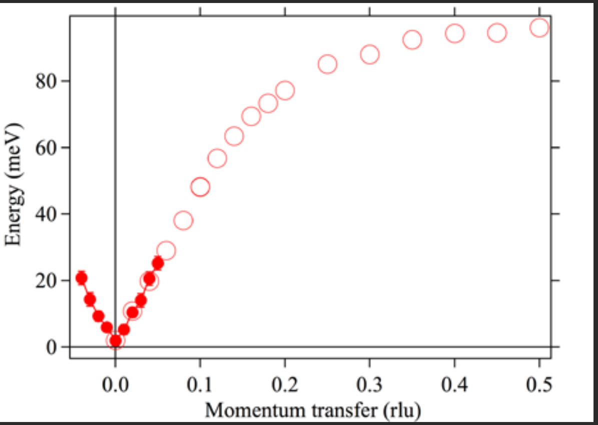
3.9 meV analyzer energy resolution + 8.9 meV incident bandpass → 9.7 meV total energy resolution



Example 1 : Sr_2IrO_4
: in-plane magnon gap at the magnetic zone center

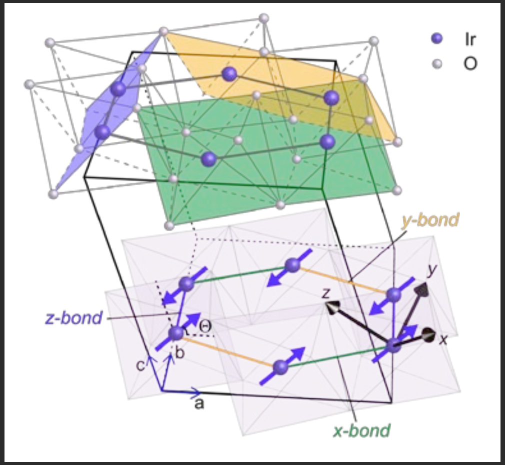






J. Kim et al., Scientific Reports 8, 1958 (2018)
J. Porras et al., Physical Review B 99, 085125 (2019)

Example 2 : Na_2IrO_3 2D honeycomb structure

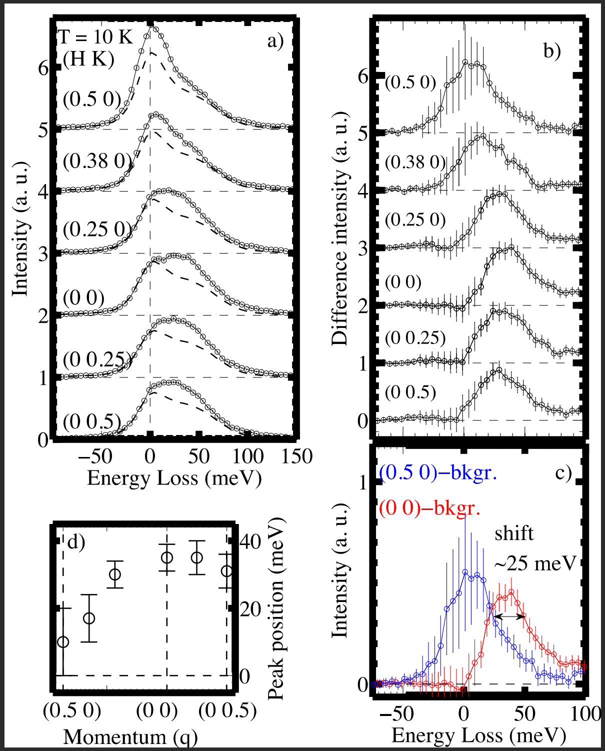


- Existence of the Kitaev exchange
- Sign and size of the Kitaev term
- Spin Hamiltonian
- Parameter values of various exchange terms (J_1,J_3, K, **Γ**, **Γ'**) in spin Hamiltonian
- Frustration and proximity to Kitaev quantum spin-liquid state

Magnetic excitation spectra

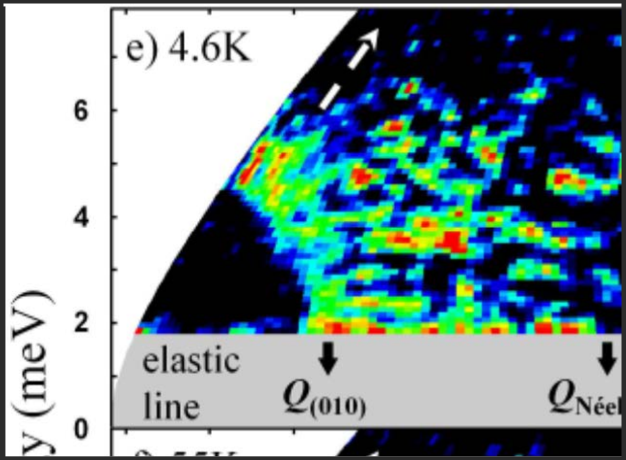
No detection of low energy magnetic excitation

RIXS



H. Gretarsson, et al., PRB **87**, 220407(R) (2013)

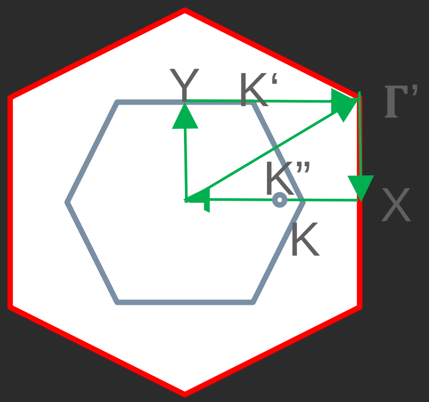
Inelastic neutron scattering of powder



S. K. Choi, et al., PRL **108**, 127204 (2012)

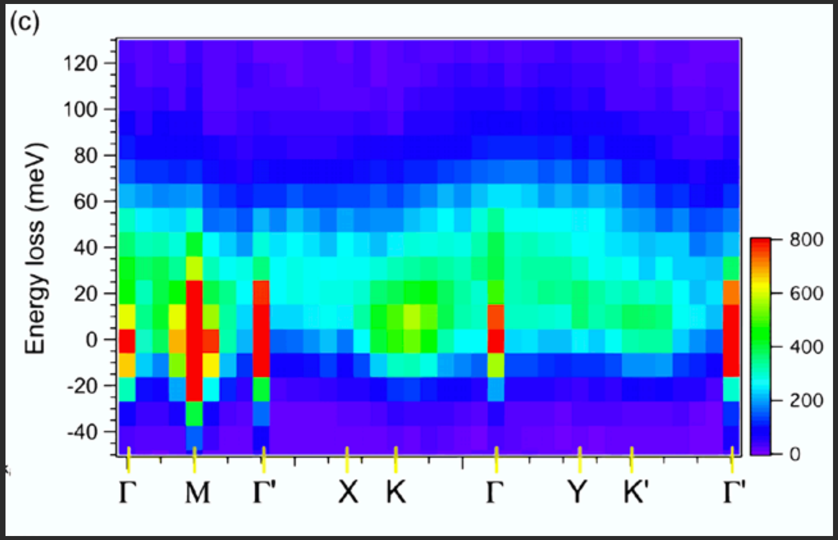
RIXS (ΔE=25 meV)

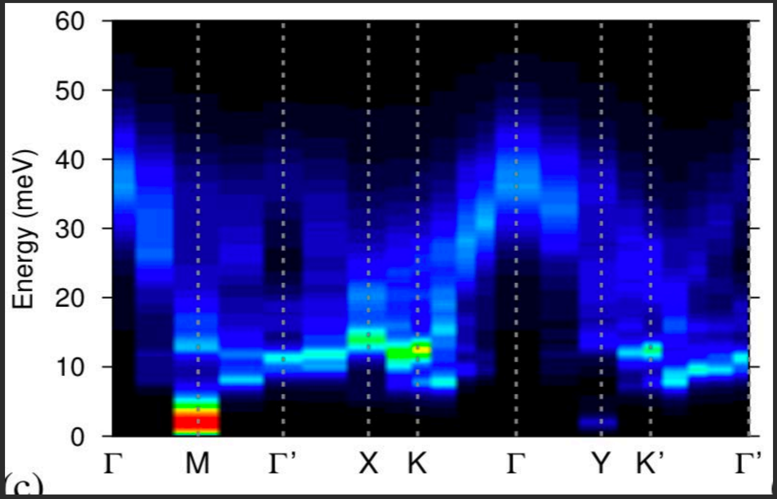
Magnetic excitation spectra along high symmetry Brillouin points



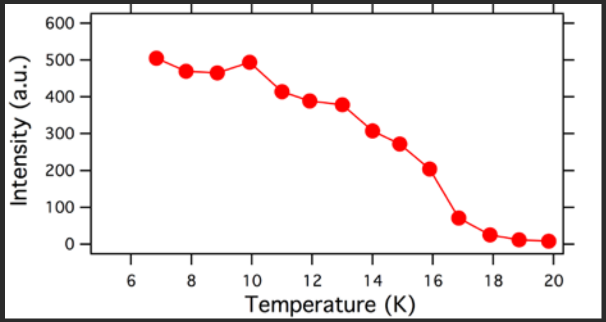
$$\mathcal{H}_{ij}^{(z)} = K\, S_i^z S_j^z + J\, \mathbf{S}_i \cdot \mathbf{S}_j + \Gamma\left(S_i^x S_j^y + S_i^y S_j^x\right) + \Gamma'\left(S_i^x S_j^z + S_i^z S_j^x + S_i^y S_j^z + S_i^z S_j^y\right).$$

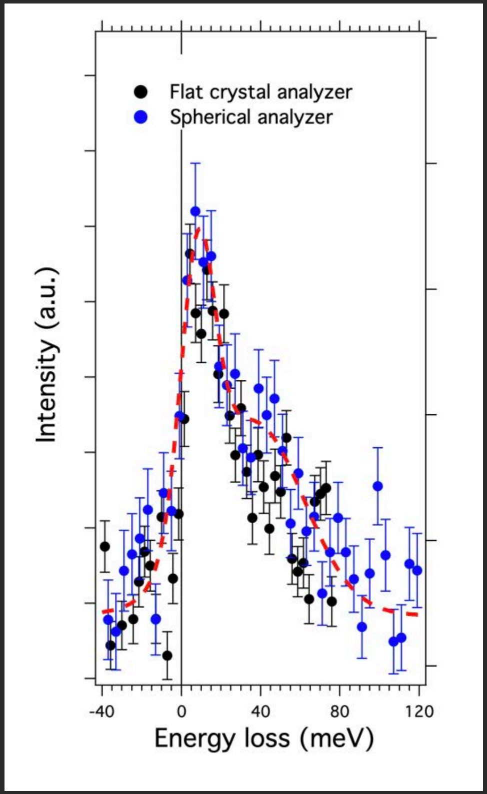
32 sites ED calculations
J=9 meV, K=12 meV, Γ=15 meV,
Γ'=2 meV, J_2= 1 meV, J_3= 2 meV



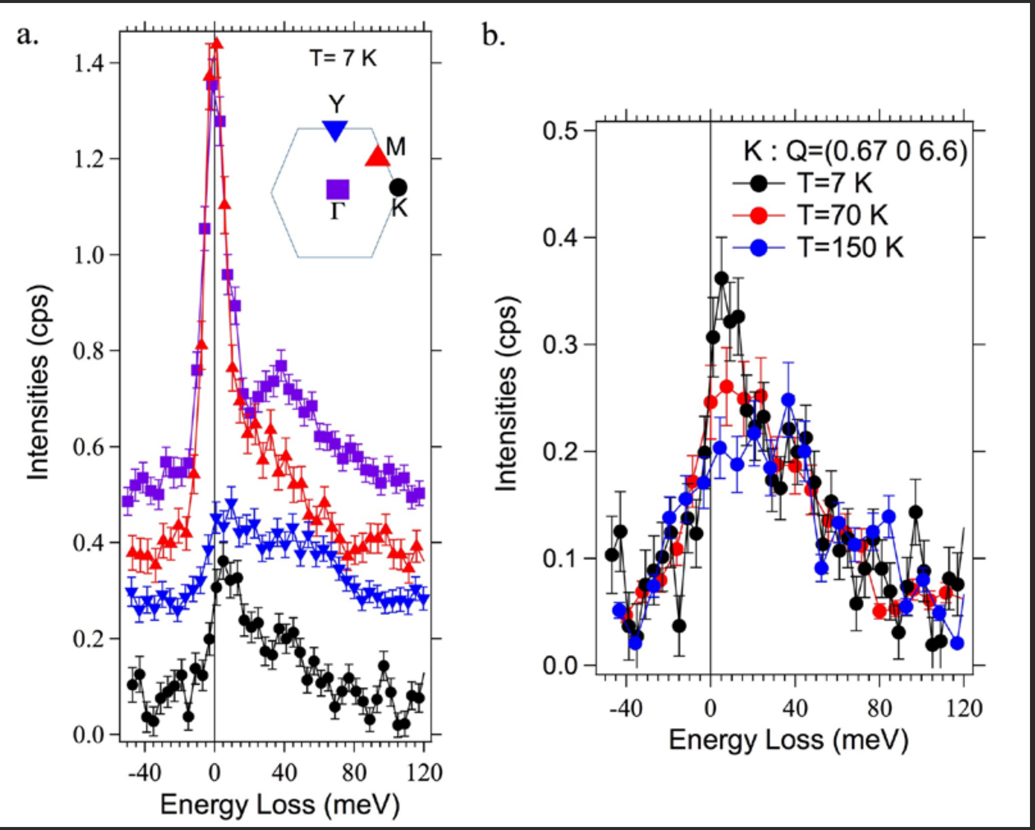


High resolution (ΔE=10 meV) RIXS spectra at K





Detection of coherent magnon and Identification of incoherent magnetic response in Na_2IrO_3



PRL **117,** 127203 (2016) PHYSICAL REVIEW LETTERS week ending 16 SEPTEMBER 2016

Resonant Inelastic X-Ray Scattering Response of the Kitaev Honeycomb Model

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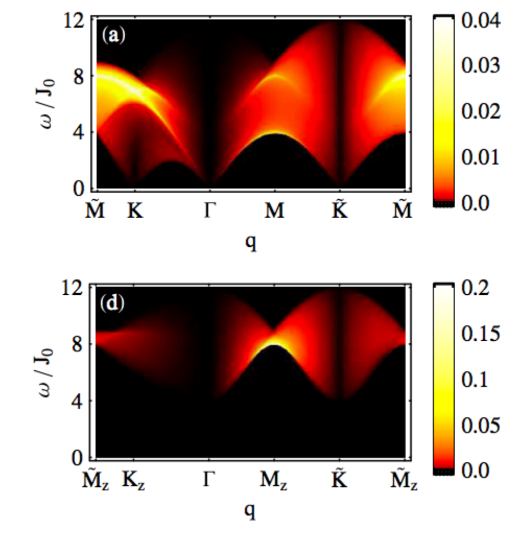
[2]*School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55116, USA*
[3]*IFW Dresden, Helmholtzstrasse 20, 01069 Dresden, Germany*
[4]*Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA*
(Received 10 May 2016; published 16 September 2016)

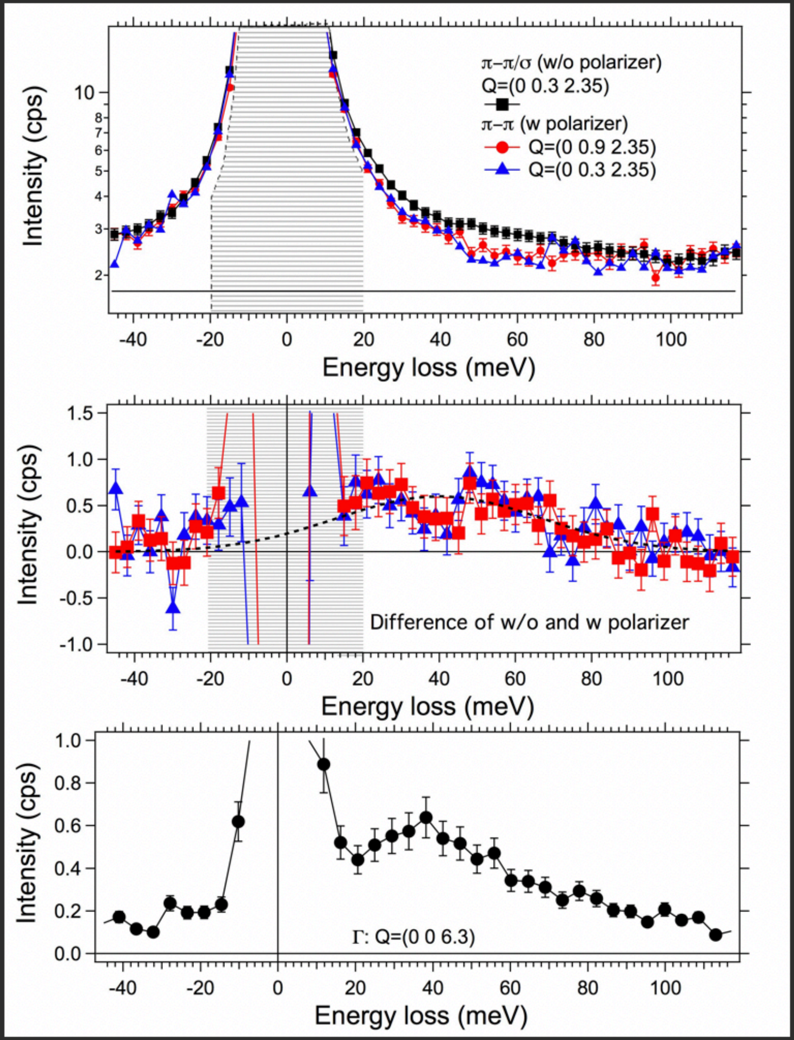
We calculate the resonant inelastic x-ray scattering (RIXS) response of the Kitaev honeycomb model, an exactly solvable quantum-spin-liquid model with fractionalized Majorana and flux excitations. We find that the fundamental RIXS channels, the spin-conserving (SC) and the non-spin-conserving (NSC) ones, do not interfere and give completely different responses. SC RIXS picks up exclusively the Majorana sector with a pronounced momentum dispersion, whereas NSC RIXS also creates immobile fluxes, thereby rendering the response only weakly momentum dependent, as in the spin structure factor measured by inelastic neutron scattering. RIXS can, therefore, pick up the fractionalized excitations of the Kitaev spin liquid separately, making it a sensitive probe to detect spin-liquid character in potential material incarnations of the Kitaev honeycomb model.

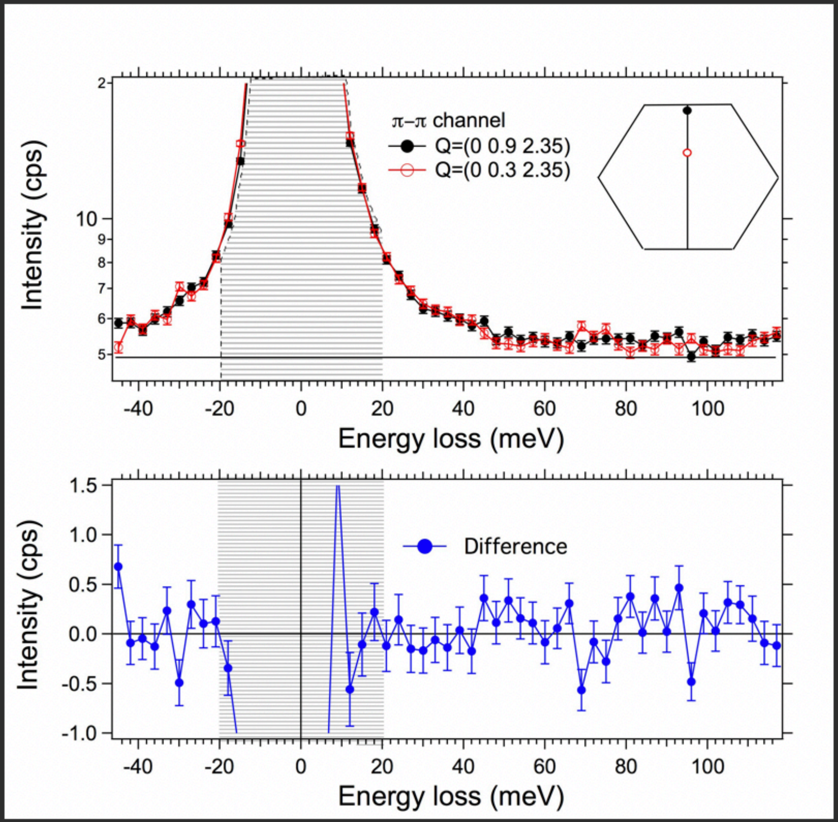
DOI: 10.1103/PhysRevLett.117.127203



Probing the elusive fractionalized Majorana excitations
→ Polarization analysis is required to distinguish the SC from the NSC (magnon).

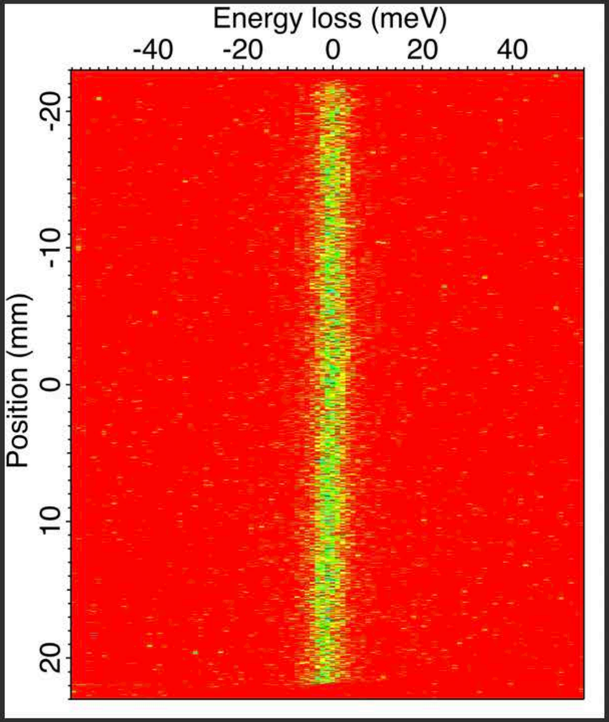
Scattered Polarization analysis

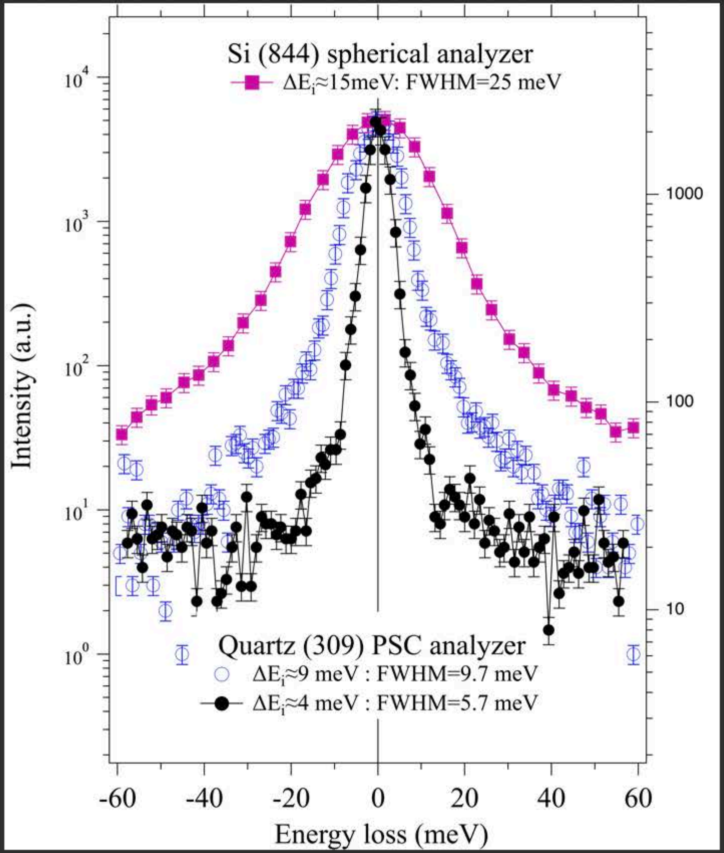




Scattering cross-section is too small ($\propto J/\Gamma^2$), whose detection is beyond the current detection noise-signal ratio.

5.7 meV energy resolution:
Quartz (309) monochromator and analyzer 3.9 meV analyzer





~10 meV analyzer for 5d TM

Examples

Edge	E_i (keV)	Collimator (Asymmetric Si 111)				Analyzer (Symmetric Quartz)			
		Θ_{Bragg}* (deg)	Θ_{Asym}*. (deg)	$\Omega_{acceptance}$ (μrad)	$\Omega_{emission}$ (μrad)	Miller index	Θ_{Bragg} (deg)	$\Omega_{acceptance}$ (μrad)	ΔE_{ana} (meV)
Ir L_3	11.215	10.154	8.95	94.7	6.1	**3 0 9**	88.358	11.5	3.7
Os L_3	10.871	10.478	9.28	99.6	6.2	**0 4 8**	89.421	83.7	9.18
Re L_3	10.535	10.816	9.62	104.7	6.3	**0 6 5**	82.9459	6.8	8.8
W L_3	10.207	11.168	9.97	110	6.4	**8 -3 0**	87.717	18.7	7.6
Ta L_3	9.881	11.542	10.34	115.8	6.5	**6 0 4**	87.661	22.9	9.2

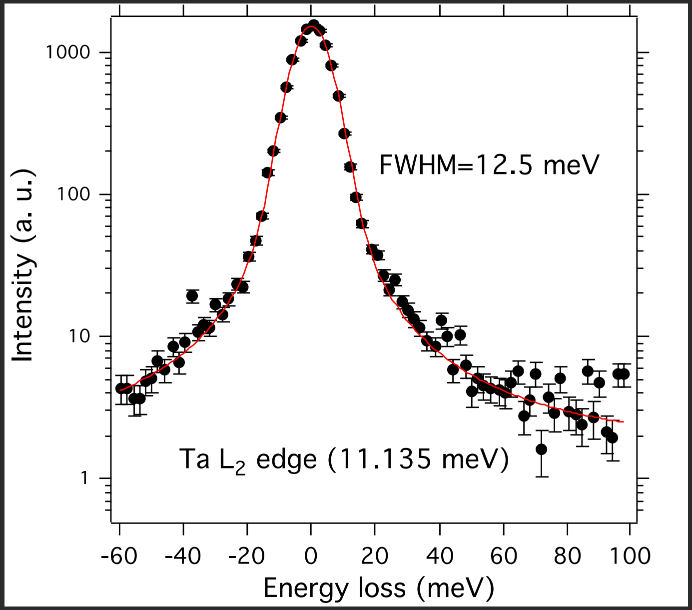
by T. Gog

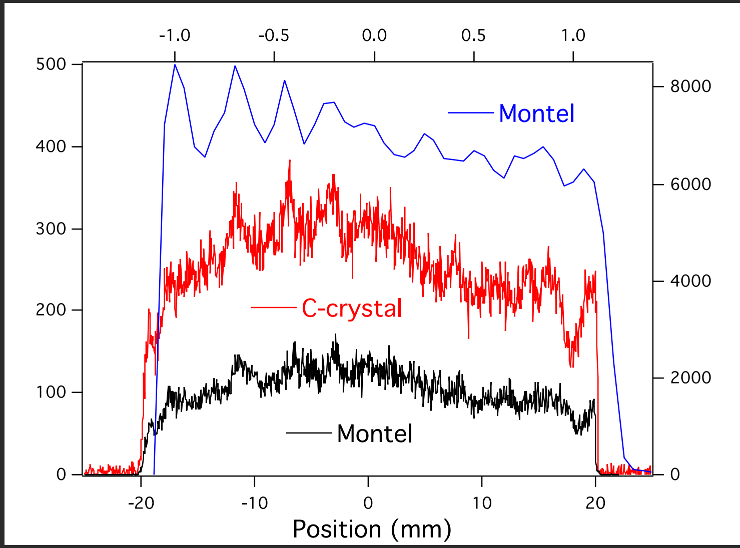
Resolution function for Ta L_2 edge (11.135 keV):

Quartz (2 4 -7) 6.3 meV analyzer energy resolution + 10.4 meV incident bandpass

→

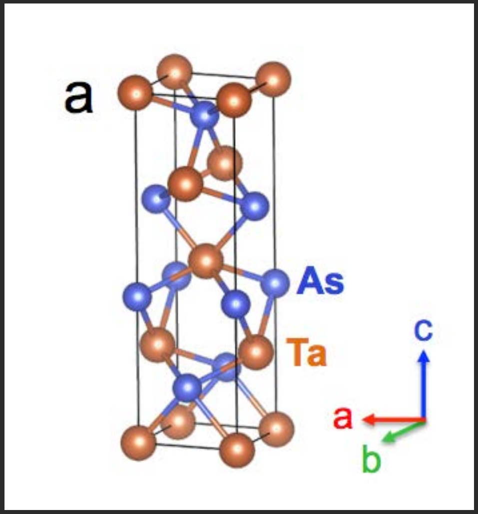
12.2 meV theoretical energy resolution
12.5 meV measured energy resolution

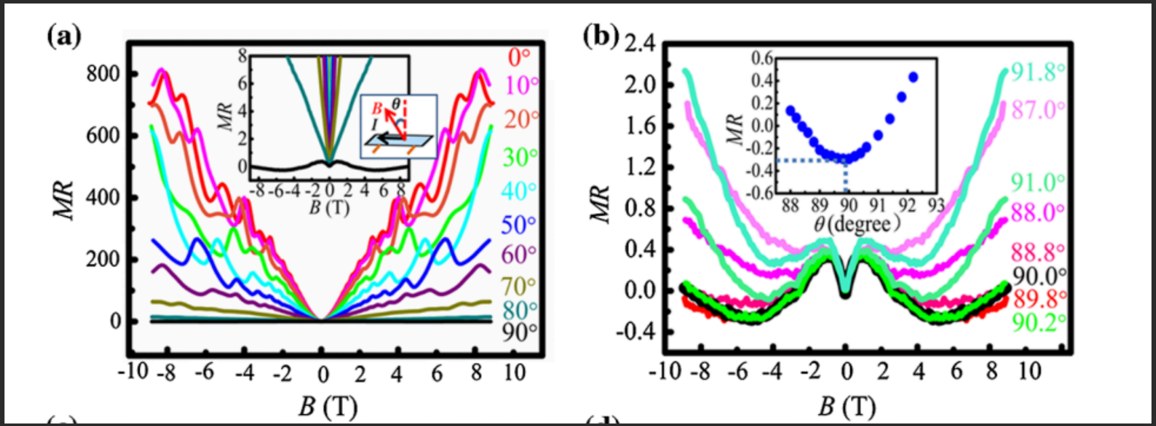




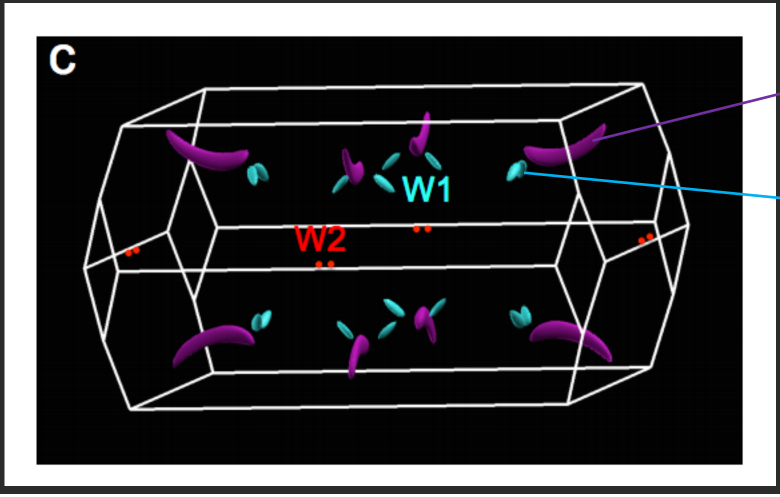
TaAs: Weyl semimetal : 3D analog of graphene

TaAs : inversion symmetry breaking Weyl semimetal





Negative longitudinal magnetoresistance

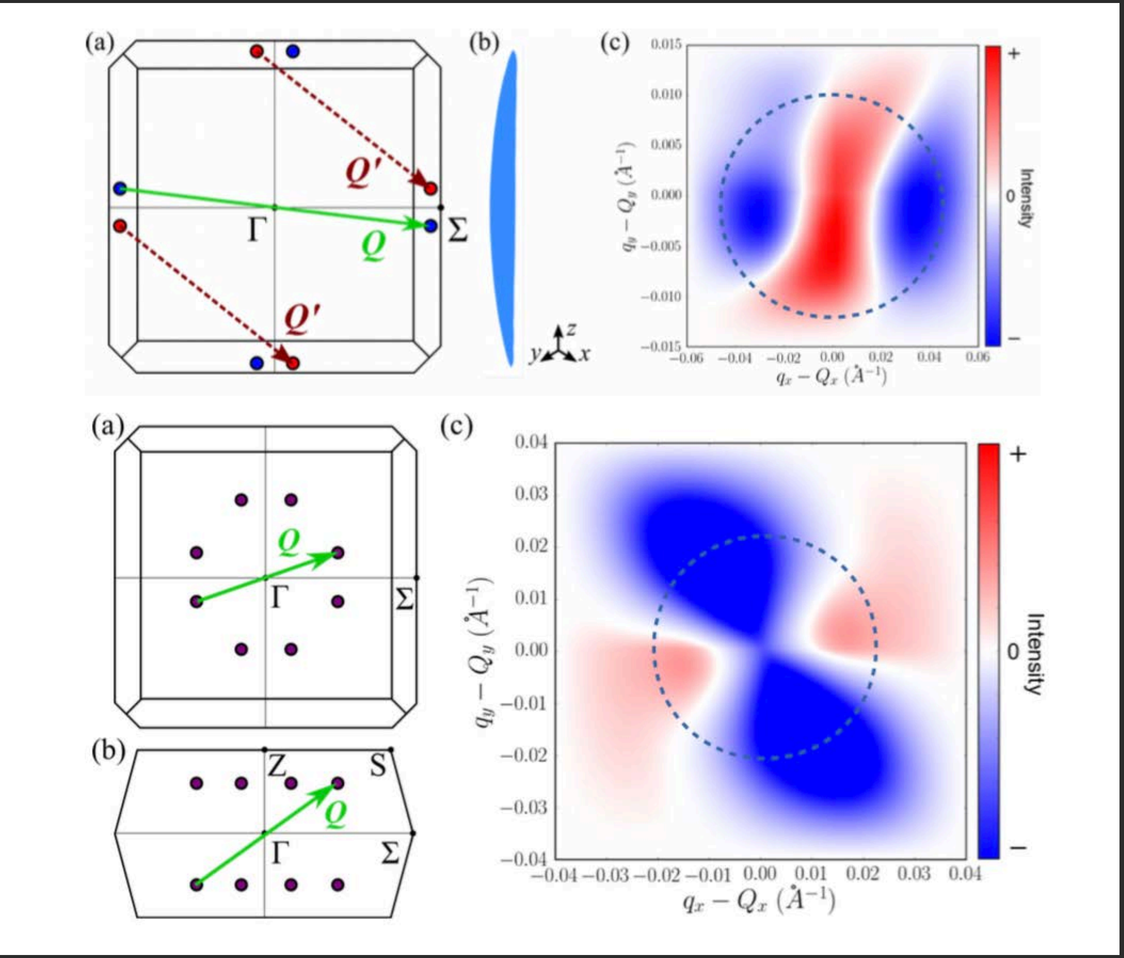


Topologically trivial Fermi surface

Weyl nodes 21 meV below E_F

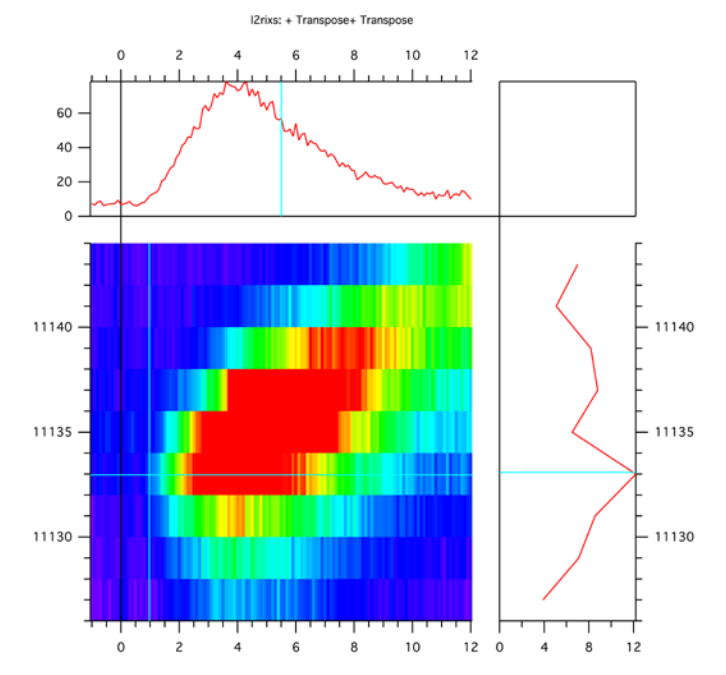
B. Q. Lv et al., Nat. Phys. **11**, 724–727 (2015)
X. Huang, et al., PRX **5**, 031023 (2015)

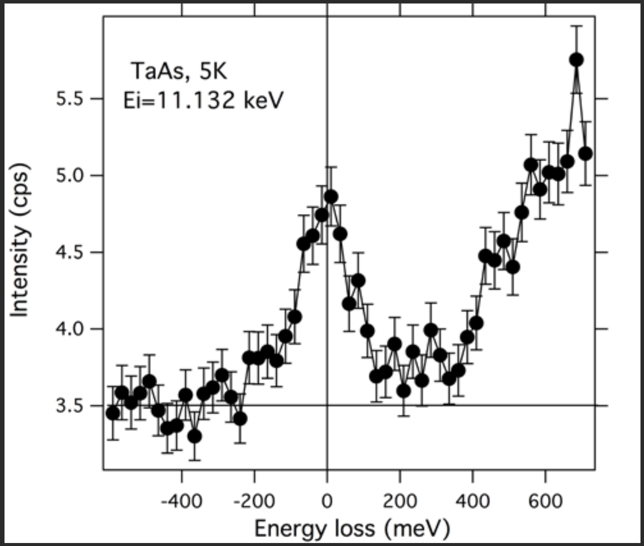
Inter-node resonant inelastic x-ray scattering



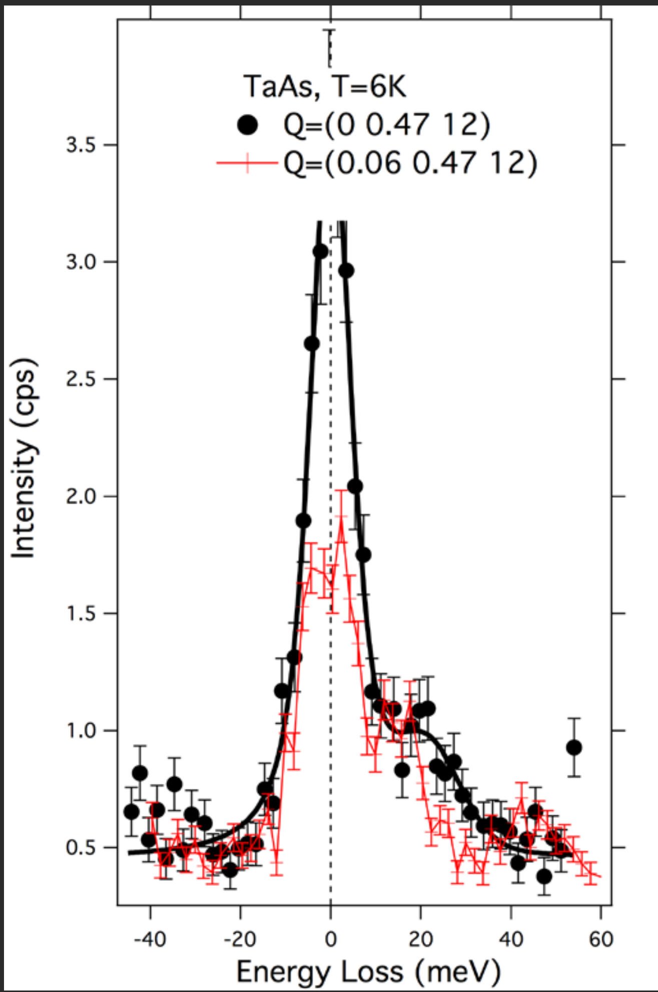
Kourtis, Phys. Rev. B **94**, 125132 (2016)

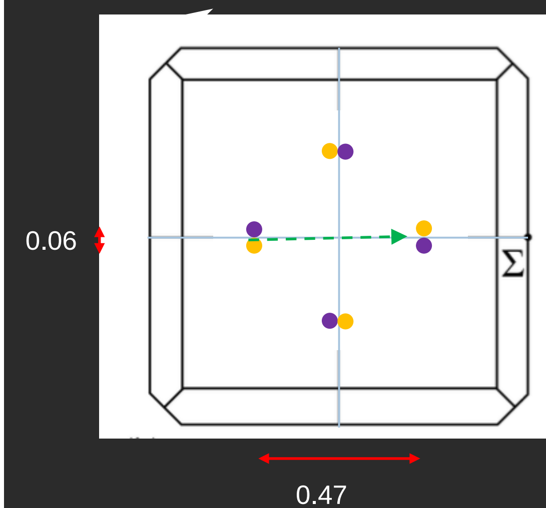
RIXS at the L2 edge

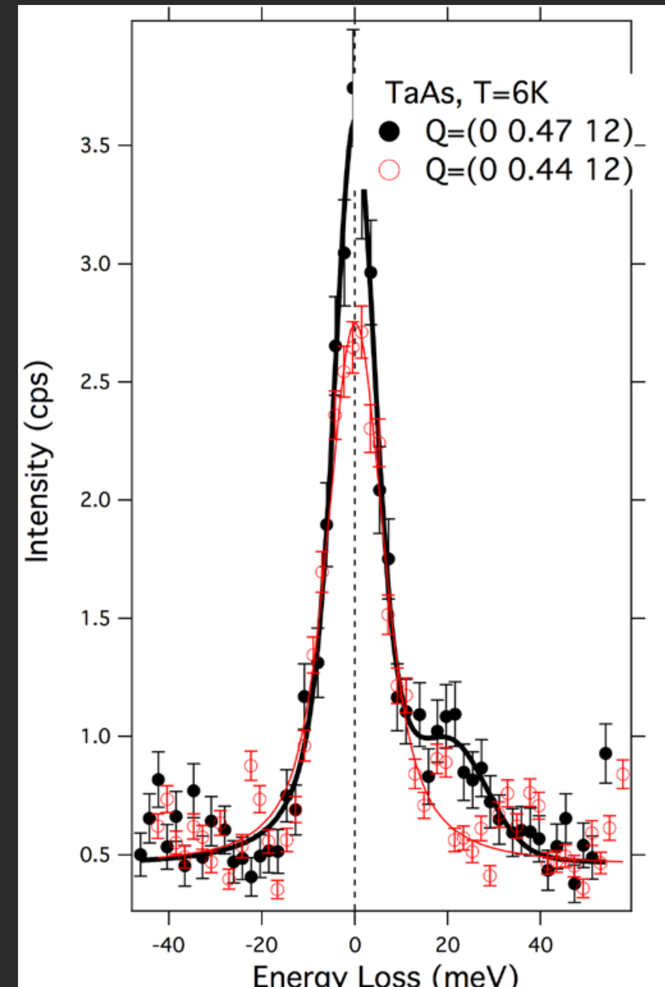




Low energy excitation at ~ 20 meV

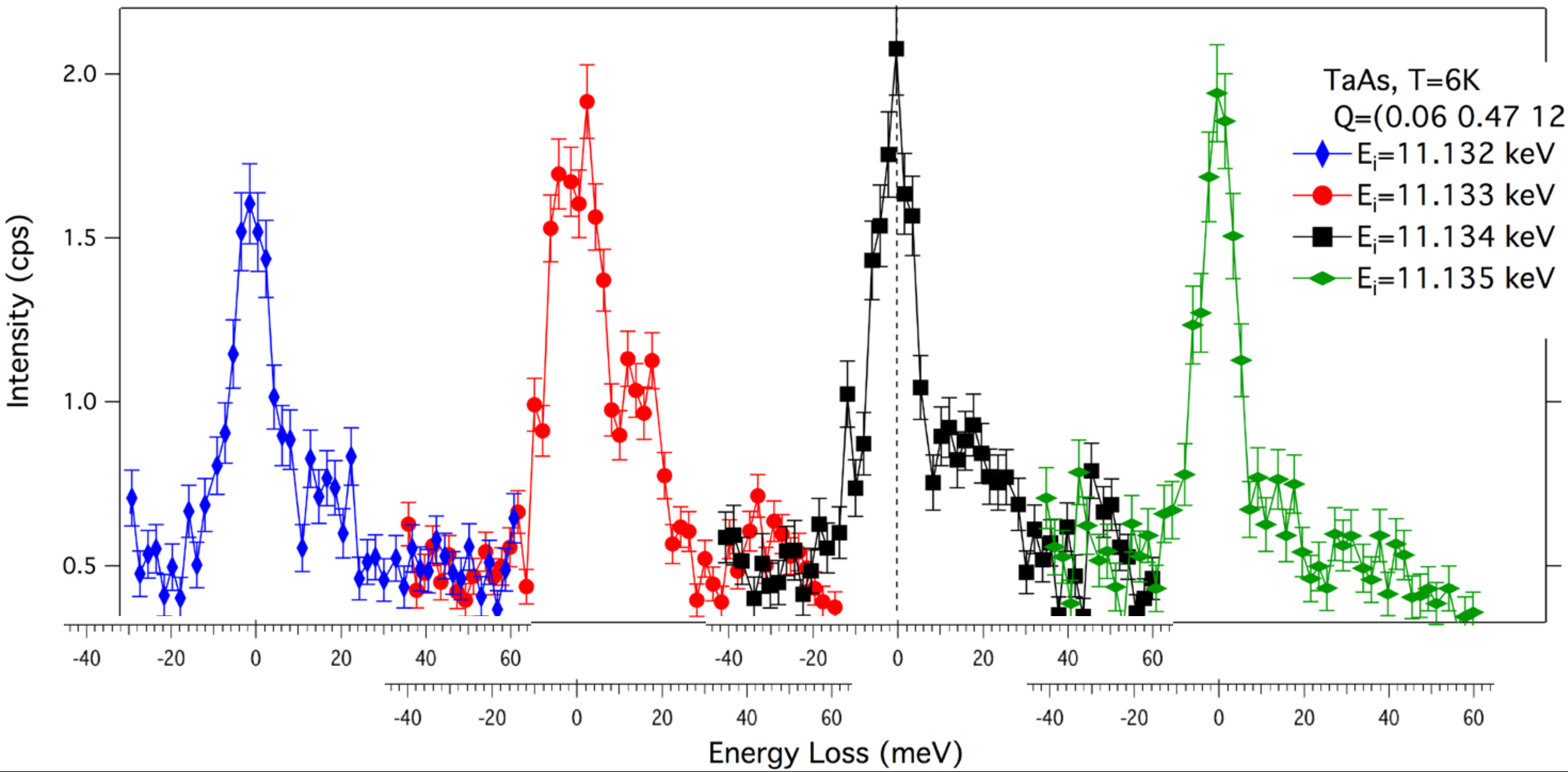


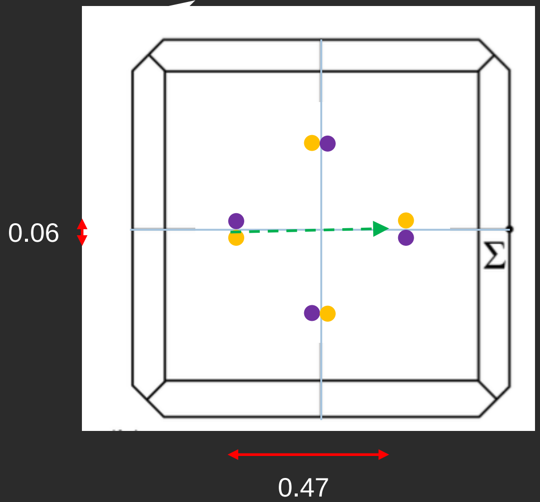


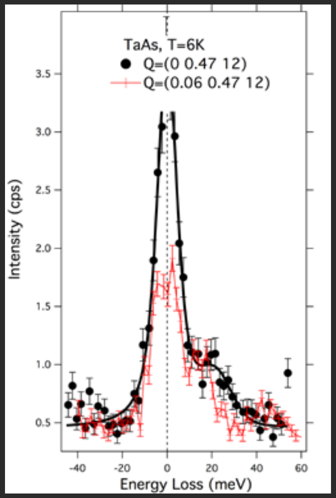


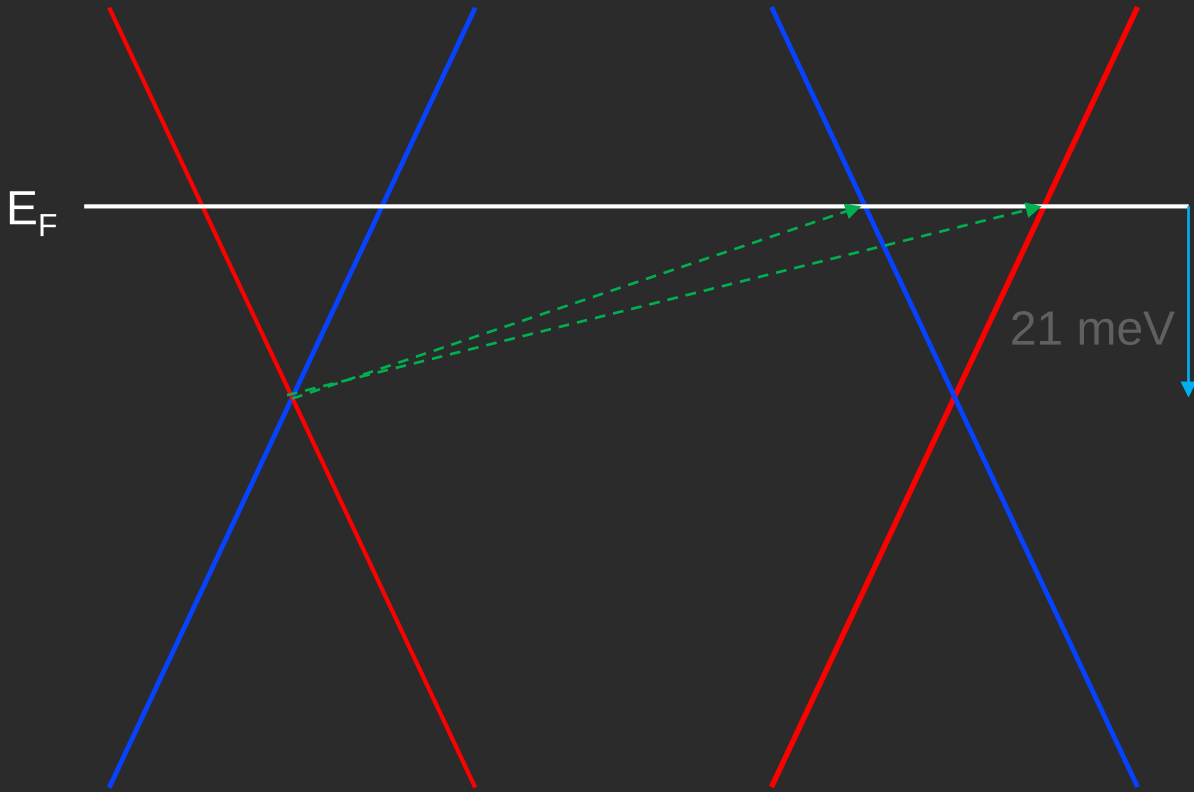
Momentum resolution : (ΔH,ΔK,ΔL)~(0.004,0.009,0.06) rlu

Resonance profile



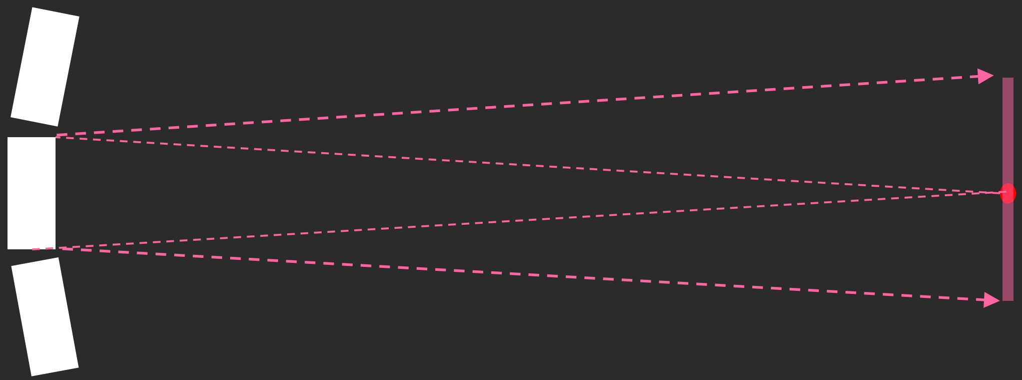

0.06
Σ
0.47


TaAs, T=6K
Q=(0 0.47 12)
Q=(0.06 0.47 12)
Intensity (cps)
Energy Loss (meV)


E_F
21 meV

Spherical analyzer
for a sample in the diamond anvil-Be gasket environment

Diced

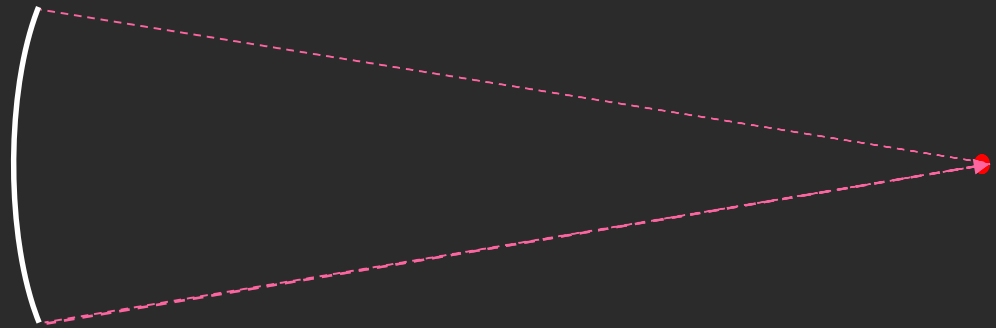


- High energy resolution
- Large image on detector

: Scatterings from nearby scattering source largely overlaps.

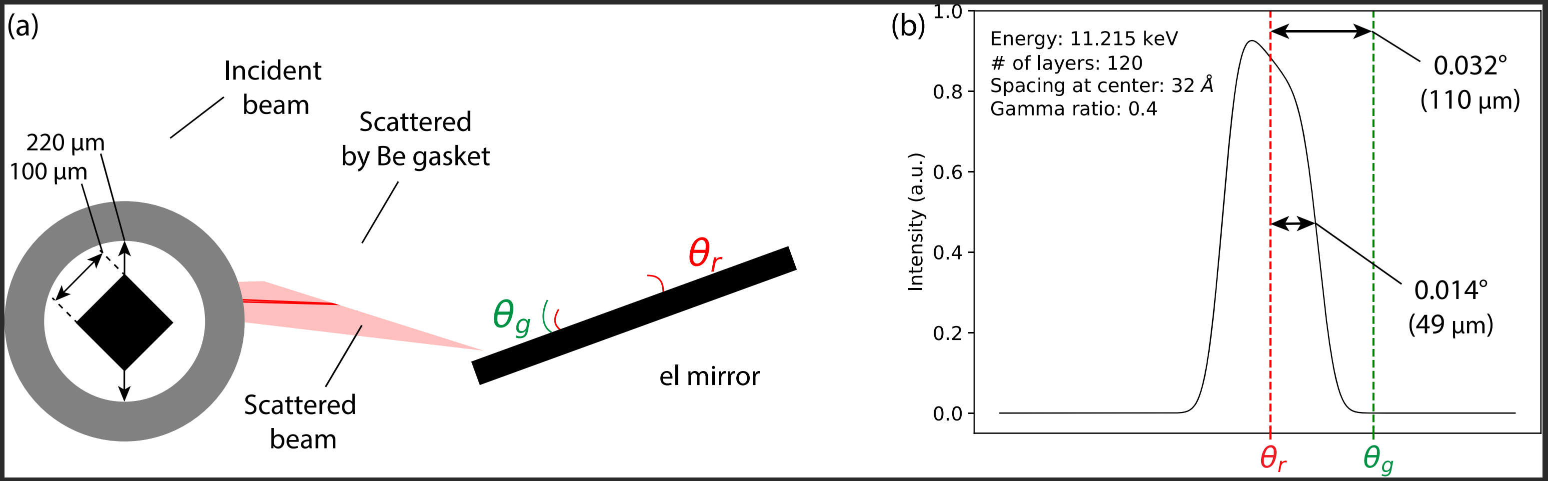
- Big sample at low pressure

Bent

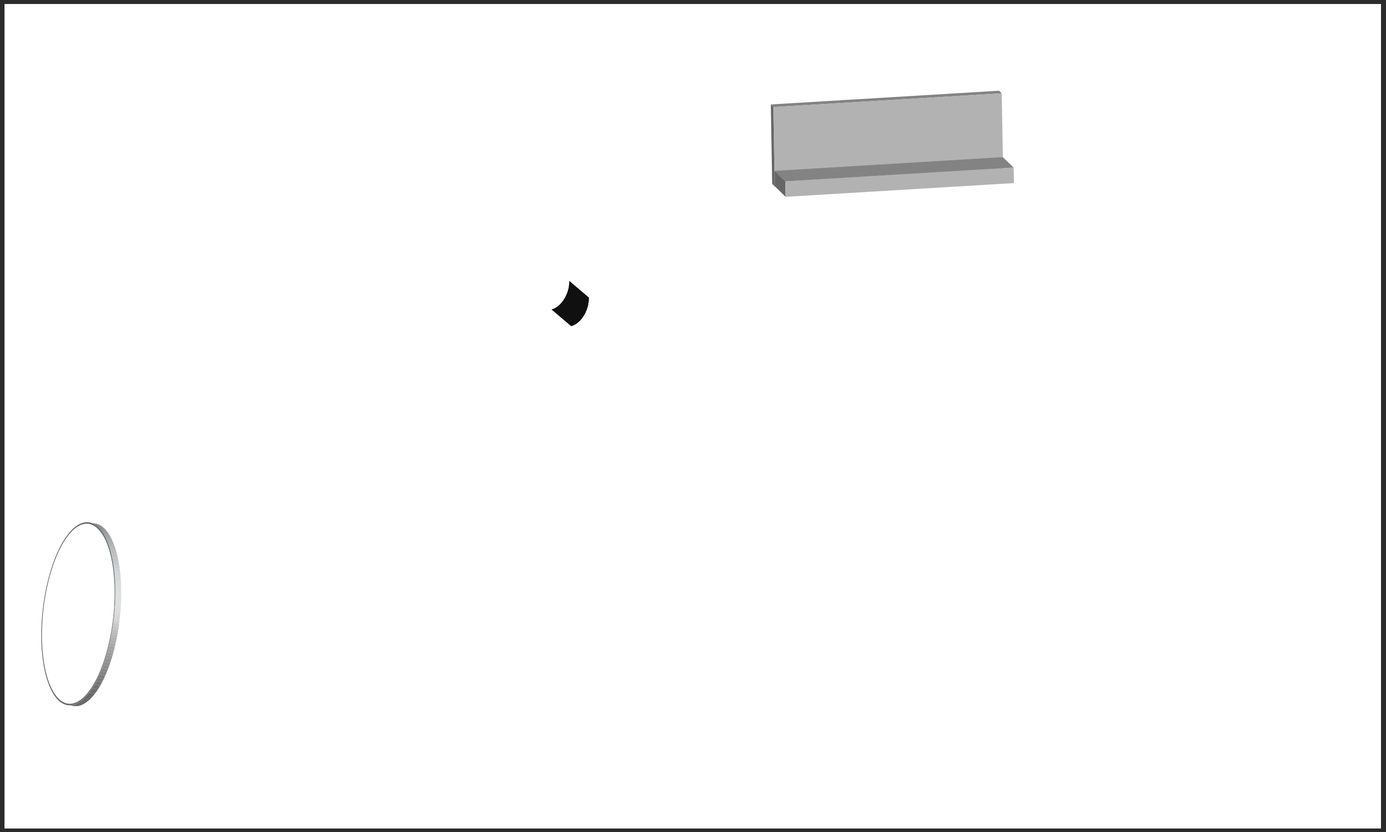


- Good focal resolution
- Poor energy resolution due to intrinsic strain and figure errors

Montel mirror
for a sample in the diamond anvil-Be gasket environment



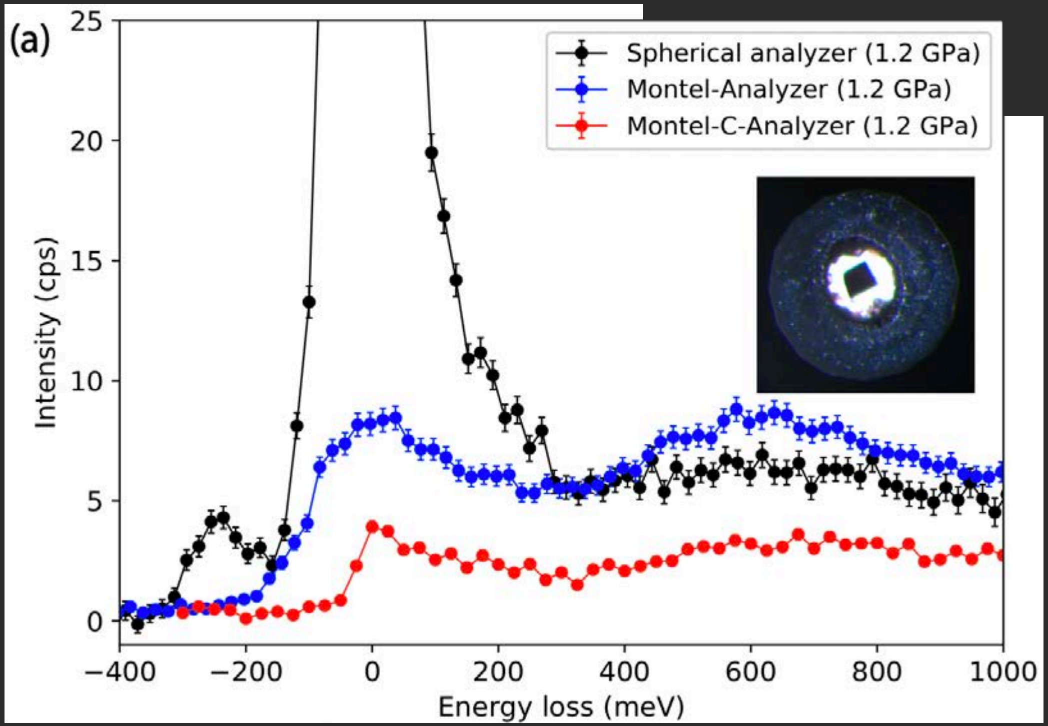
Two configurations for medium energy resolution



MCA

MA

Sr_2IrO_4



- **Spherical analyzer measurement :same sample and same solid angle**
- **Scatterings from nearby scattering source is largely reduced in the PSC analyzer measurements.**

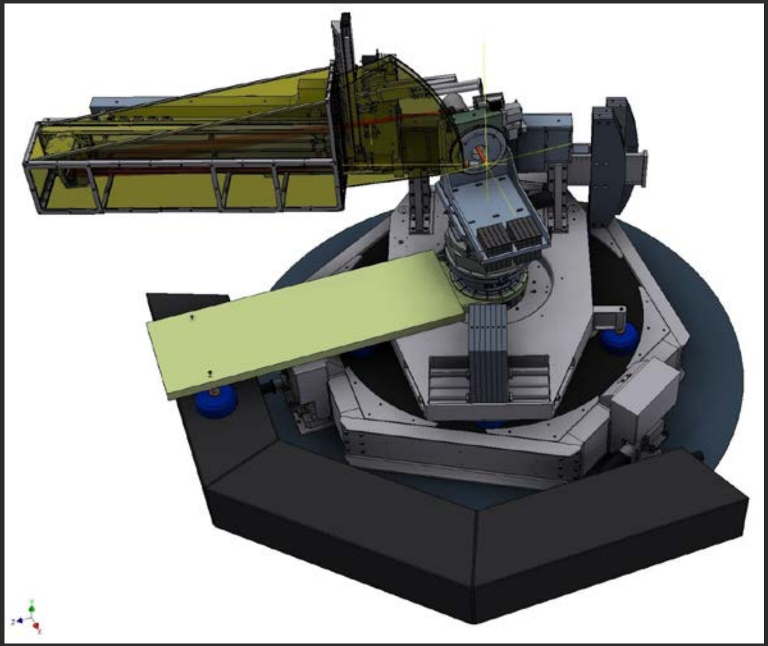
Summary

- High spectral resolution of post-sample-collimation flat crystal RIXS analyzer
  : For 5d TM elements, 5~10 meV is achievable.
  : 10 meV resolution spectra of iridium oxides.
  : Scattered polarization analysis
  : Near theoretical energy resolution of 5.7 meV

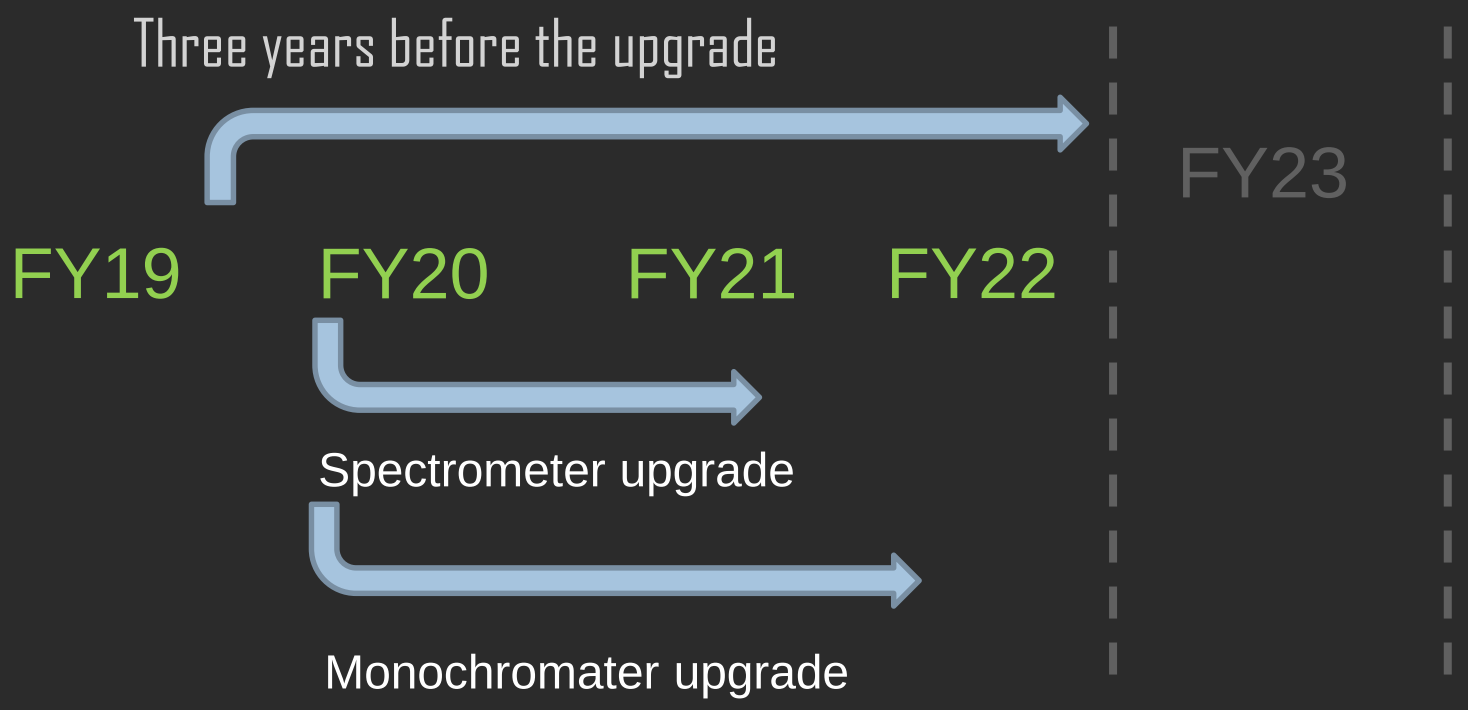
Facility upgrades will make such high resolution more practical and promise an exciting journey to explore otherwise impossible quantum phenomena.

- High focal resolution of collimating optics (Montel and C-crystal)
  : RIXS analyzer system for a sample in the diamond anvil/Be gasket

➢ Spectrometer and monochromater upgrades



Energy [keV]	Energy Resolution [meV]			
	4-bounce Si(220) Path #1	4-bounce Si(440) Path #1	4-bounce Si(660) Path #1	6-bounce Si(220)SI(hkl)Si(220) Path #2
6	84			
7	80	30		
8	70	30		
9	59	30		
10	49	30	12	5
11	39	30	11	5
12	30	30	11	5


Three years before the upgrade
FY23
FY19
FY20
FY21
FY22
Spectrometer upgrade
Monochromater upgrade