(Resonant) Inelastic X-ray Scattering at the APS: The New RIXS Beamline 27-ID and The Improved HERIX Beamline 30-ID

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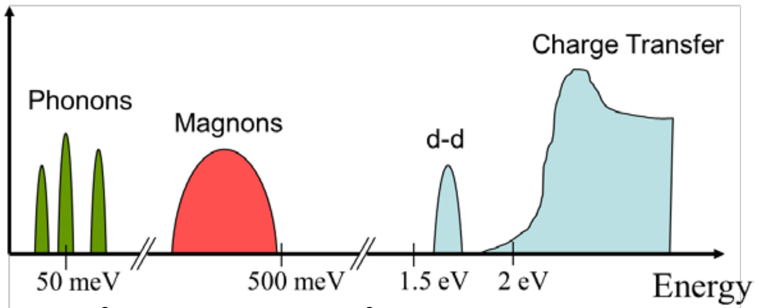
Why Inelastic X-ray Scattering ?



- Goal: Understanding Material Properties => Devices
- Need to understand both

Statics (crystal structure, magnetic structure, …)

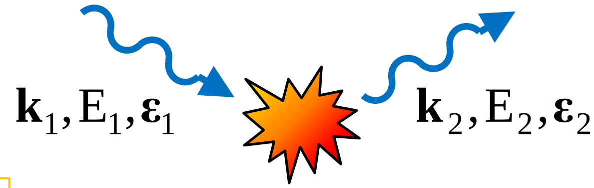
Dynamics (lattice vibrations, electronic excitations, magnetic interactions, …)

Examples: Giant Magneto-Resistance, Superconductivity

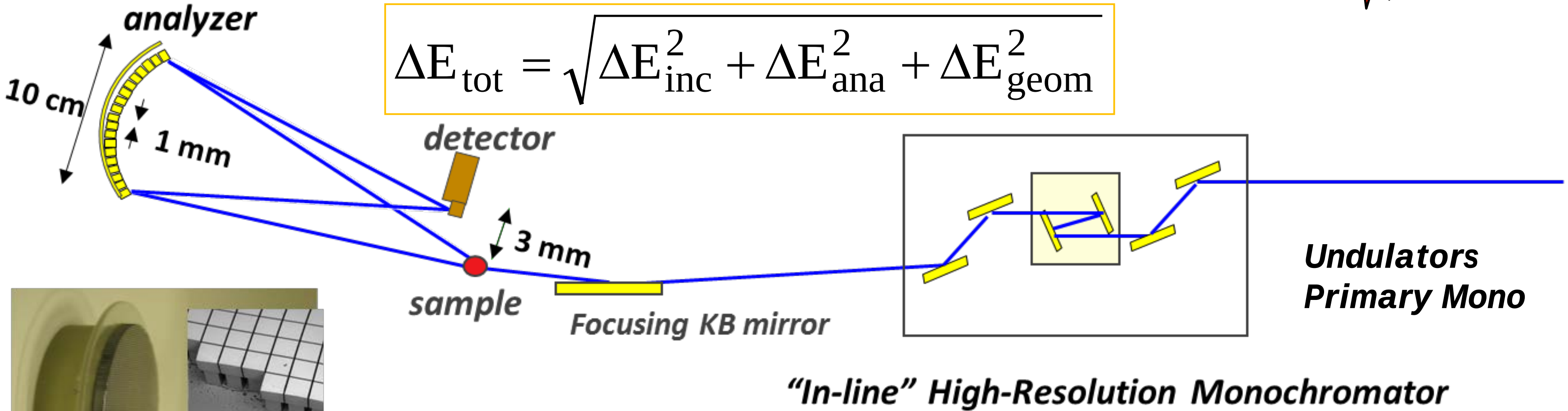
Inelastic X-ray Scattering (RIXS, IXS, HERIX, LERIX, …)

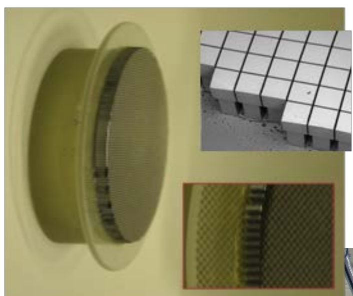
Elastic X-ray Scattering, Diffraction

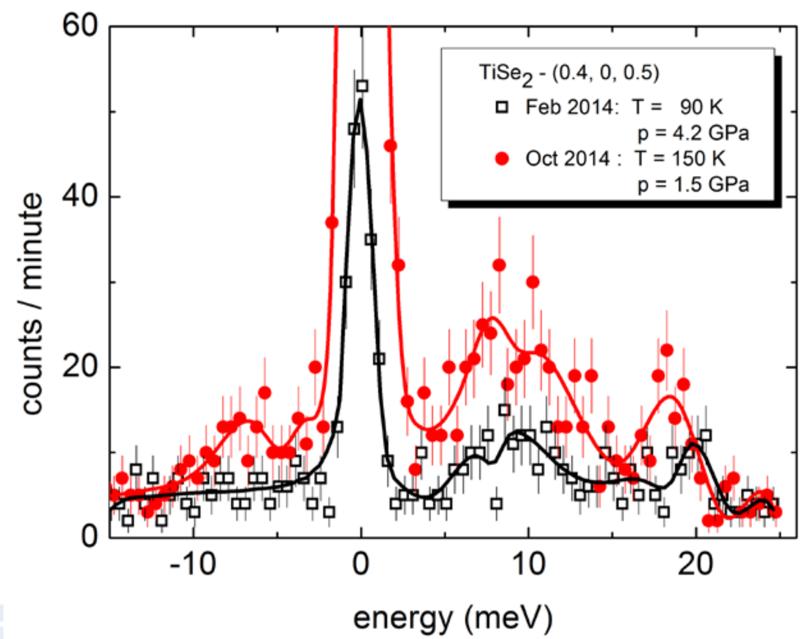
How is an IXS measurement done?
(HERIX 30-ID, lattice vibrations)



$$\Delta E_{tot} = \sqrt{\Delta E_{inc}^2 + \Delta E_{ana}^2 + \Delta E_{geom}^2}$$





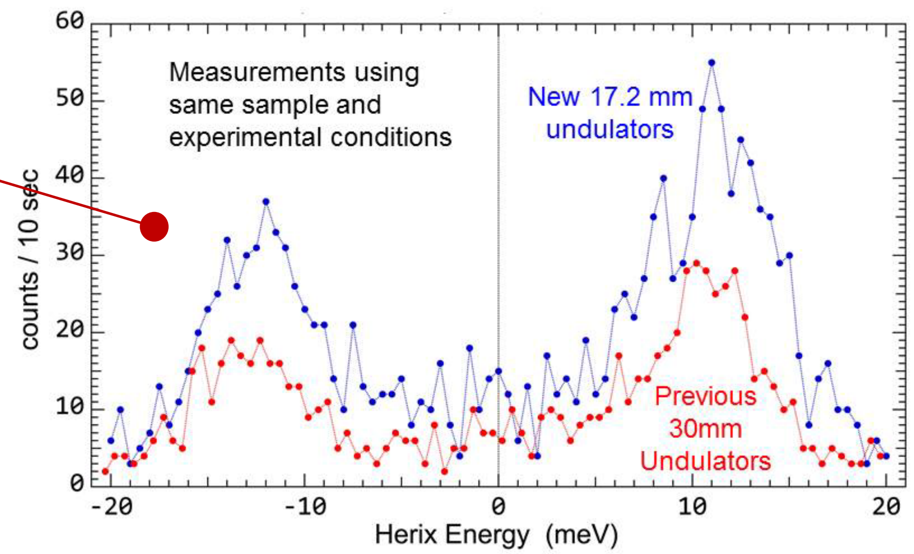


New Short-Period Undulators at 30-ID (Oct. 2014)

- High-resolution inelastic spectroscopy is extremely photon-hungry.
- Replacement of 30mm undulators by new, short-period 17.2mm devices **doubles the incident flux, substantially improves flux density, beam divergence and stability** for the HERIX instrument @ 23.7 keV
- **Immediate, significant impact on user operation** at 30-ID
- Doubling the incident flux enables users to obtain publishable data sets within as little as one visit to the APS, instead of multiple visits.

HERIX spectra featured in a recent publication (Budai et al., Nature 515, 535 (2014)) on the nature of the Metal-Insulator transition in Vanadium Dioxide (VO_2)
(**red curve**) before, (**blue curve**) after implementation of the new undulators.
John Budai: "Measurements needed for the Nature paper required two visits of 6 days each, that were separated by one year due to high demand for beamtime. These measurements can now be obtained in a single 6 day run."

HERIX – Inelastic x-ray scattering measurement of anharmonic phonons in VO_2



Why do we need R(esonant)IXS ?

Scattering Cross Section

for **lattice vibrations (phonons)**: small, but practical (HERIX)

for **electrons**: much, much smaller

(… and a lot of the interesting novel phenomena in materials are dominated by correlated electron systems …)

Why do we need RIXS ?

Scattering Cross Section

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

Non-Resonant (weak)

X-ray Optics:

Choose suitable parameters

- Energy resolution
- Throughput

↓

- Incident energy E_i

Resonant Enhancement (x 50…100)

X-ray Optics (RIXS):

Incident energy E_i predetermined (…and different for every material)

↓

Find "Best" optics (monochromator and analyzer) for every E_i of interest

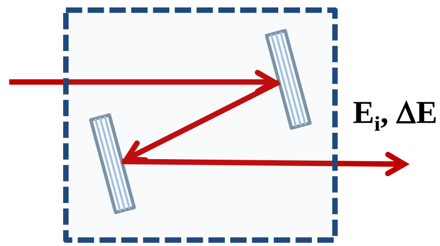
RIXS Science

Scattering Cross Section

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

Non-Resonant (weak)

Resonant Enhancement (x 50…100)



$$\frac{\text{dE}}{\text{E}} = \cot\Theta_B \Delta\Theta_B$$

$$\frac{\text{hc}}{\text{E}} = 2\text{d}_{\text{hkl}} \sin\Theta_B$$

d_{hkl} only discrete values !

X-ray Optics (RIXS):

- High Energy Resolution:

Bragg (near-) Backscattering

↓

- **It is what it is !**

RIXS Science

Scattering Cross Section

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

Non-Resonant (weak)

Energy resolution:

1meV routine

< 1meV getting there

Resonant Enhancement (x 50…100)

RIXS Energy resolution:

~100 meV mid-2000

25 meV @ Ir-L3 in 2010

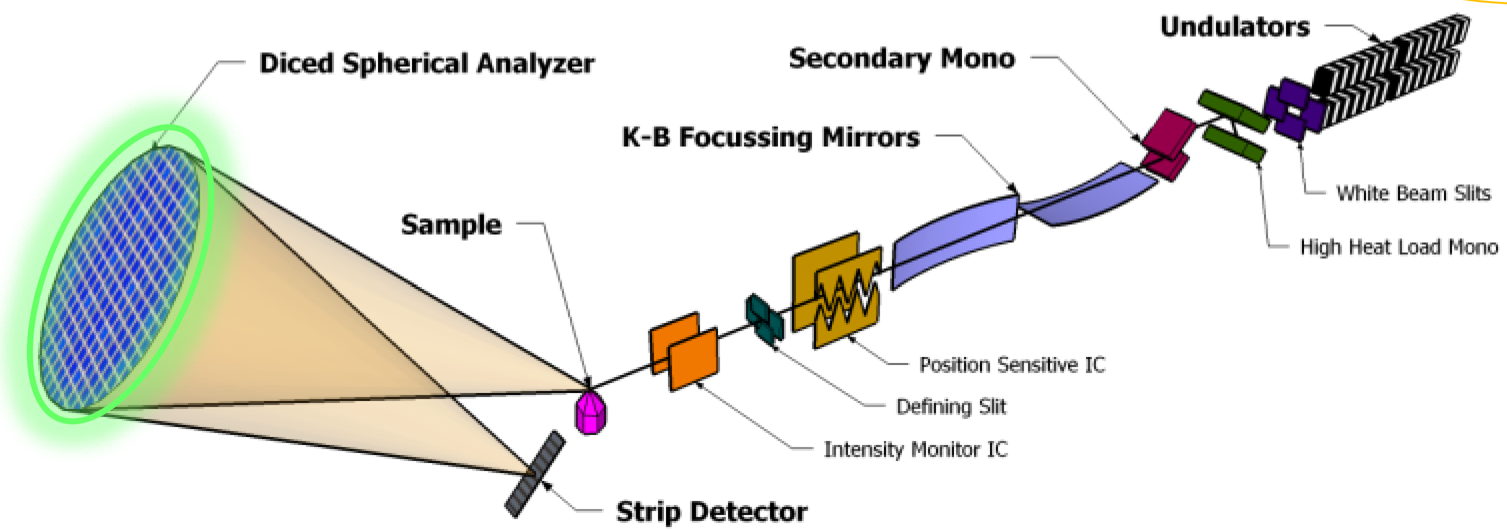
< 20 meV for many E_i new spherical analyzer materials

< 10 meV flat crystal optics

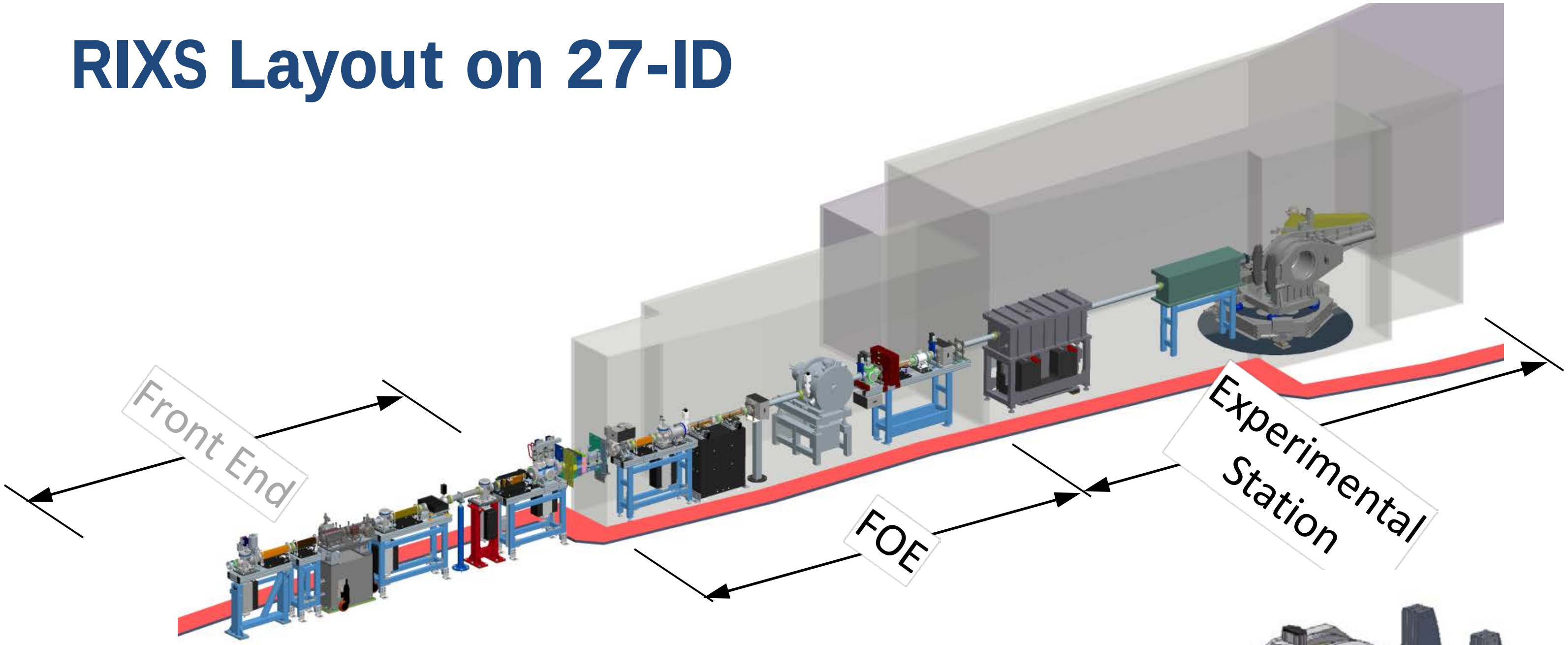
RIXS Analyzer R&D and Energy Resolution

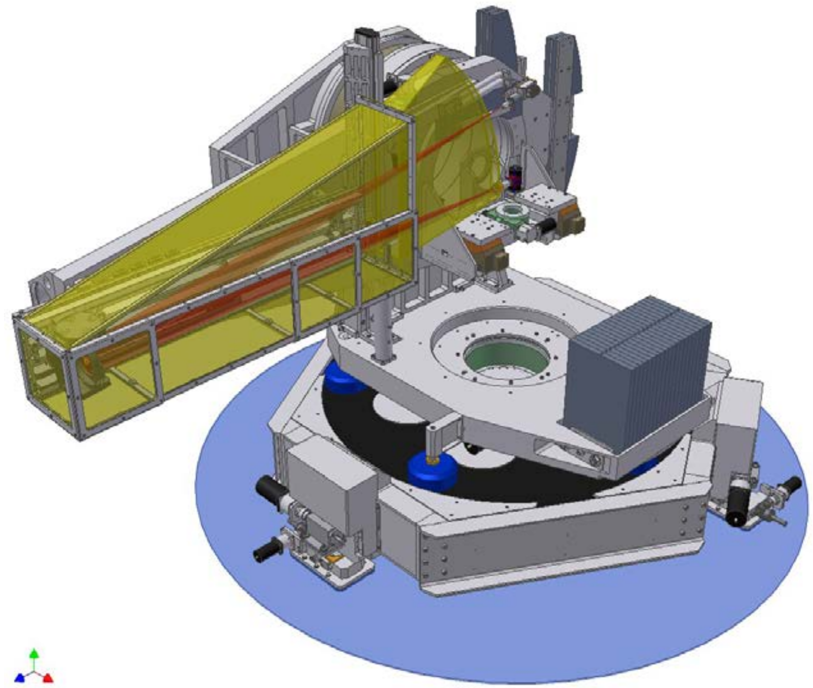
$$\Delta E_{tot} = \sqrt{\Delta E_{inc}^2 + \Delta E_{ana}^2 + \Delta E_{det}^2 + \Delta E_{spot}^2}$$

E ~5-23 KeV



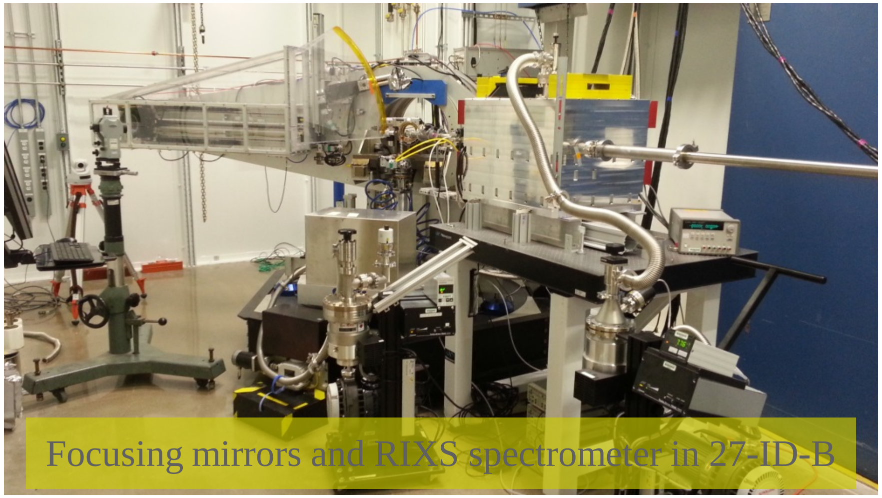
RIXS Layout on 27-ID



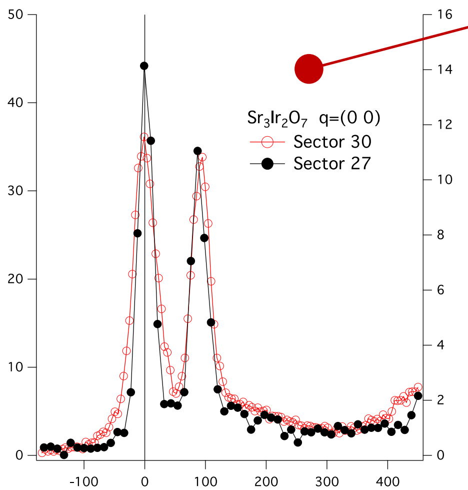


APS-U Construction Status of 27-ID RIXS

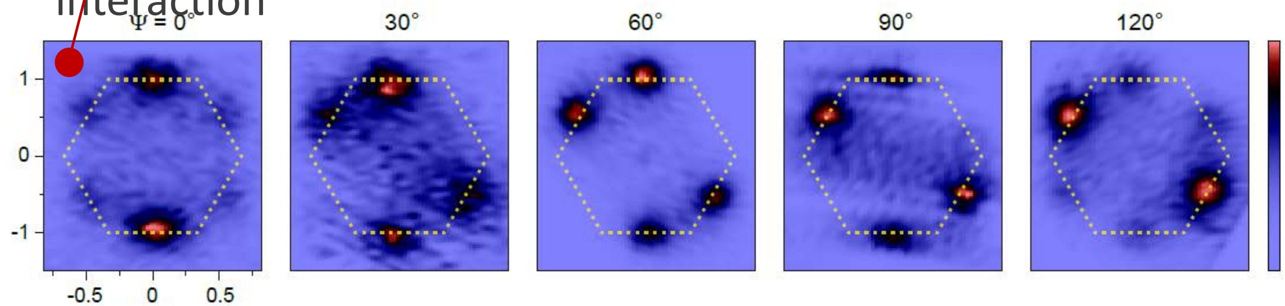
- Basic beamline construction was finished, on time and on budget by Dec 2014.
- All RIXS instrumentation and infra-structure operational by Dec 2014.
- Further commissioning during 2015-1 (high-P/low-T sample environment, new 25 μm strip detector, new spherical analyzers, …)
- Start of GU program in May 2015


Focusing mirrors and RIXS spectrometer in 27-ID-B

- First RIXS measurements on Iridium compounds demonstrate beamline performance



- Energy resolution measurements show improvement due to improved focusing
- Previously initiated diffuse magnetic scattering study on honey-comb structured Na_2IrO_3 completed at 27-ID, (submitted to Nature Physics), showing novel bond-directional magnetic interaction



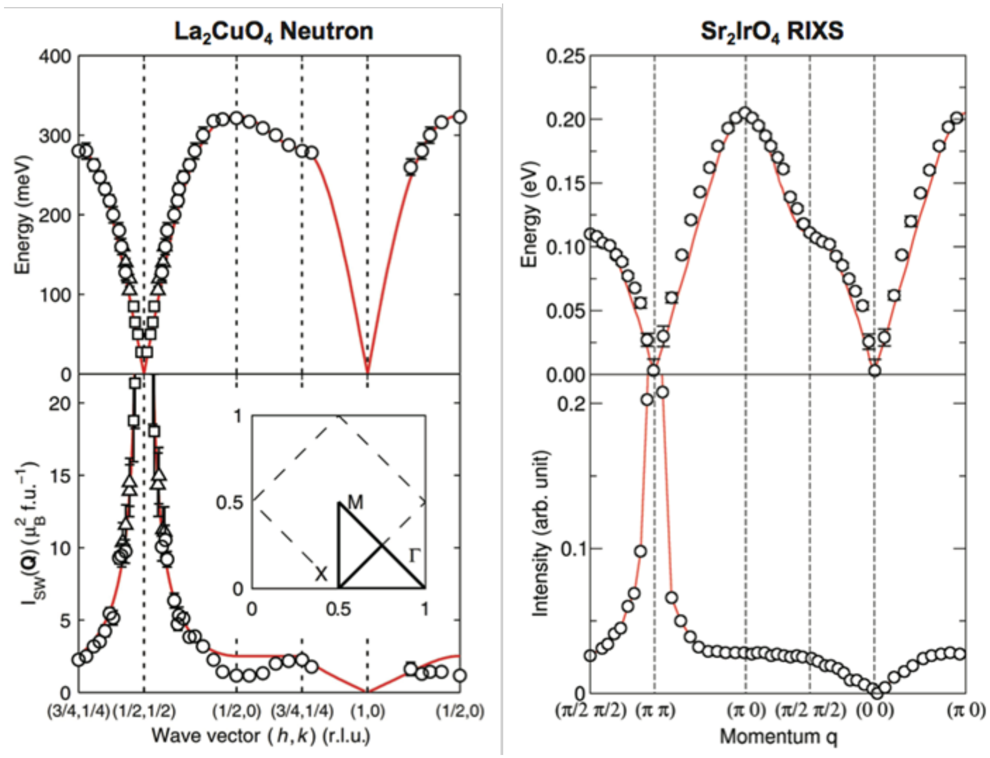
RIXS Science

5d-Transition Metal Oxides and High T_c - Superconductivity

- High T_c: discovered mid-1980s in cuprates (~140K)
- Mechanism ? Room-temperature Superconductor ?
- Strategy: Find material similar to cuprates
- Identified Sr_2IrO_4 as candidate material, **but**:
- Contains heavy element with large spin-orbit coupling =>
- Magnetic Properties vastly different ?
- Typically: Use Inelastic Neutron Scattering (INS) to check, **but** :
- No large enough Crystals available, Ir good neutron absorber

RIXS Science Example (cont.)

- RIXS: until recently not enough **Energy Resolution** (>~100 meV)
- **Breakthrough**: ~30 meV RIXS instrumentation for Iridates



Dispersion (top)
Intensity (bottom)
of Spin Wave in
La_2CuO_4 (INS)
Sr_2IrO_4 (RIXS)

N.S. Headings, et al.
Phys. Rev. Lett. **105**, 247001 (2010)
Jungho Kim, et al.
Phys. Rev. Lett. **108**, 177003 (2012)

- Spin Wave behavior virtually identical =>
- Sr_2IrO_4 might superconduct when doped with carriers

RIXS Science Example (cont.)

- If Sr_2IrO_4 does not superconduct

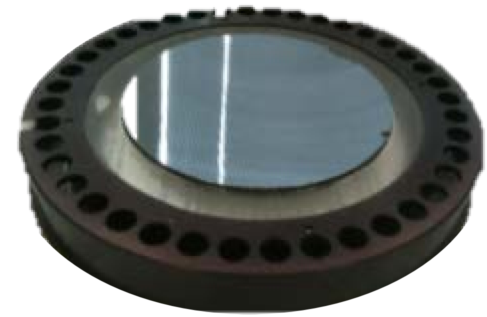
  => Refinement of High T_c Model
- **Next Steps**:

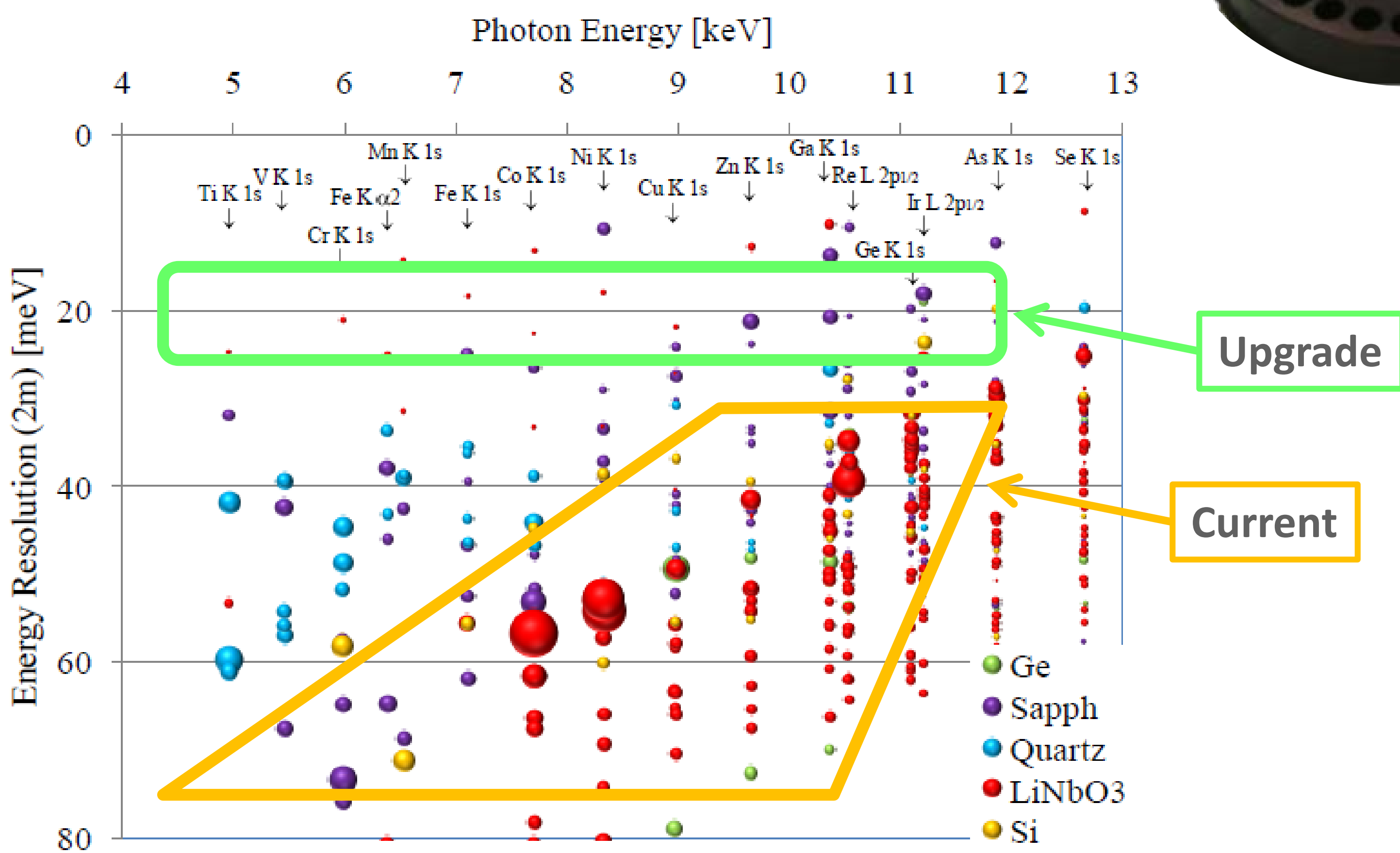
  study doped samples when available,

  with even better energy resolution

RIXS Analyzer R&D and Energy Resolution

- Complete in-house fabrication and expertise
- Si, Ge highly symmetric → restricted choices of reflections
- New materials: Sapphire, $LiNbO_3$, Quartz

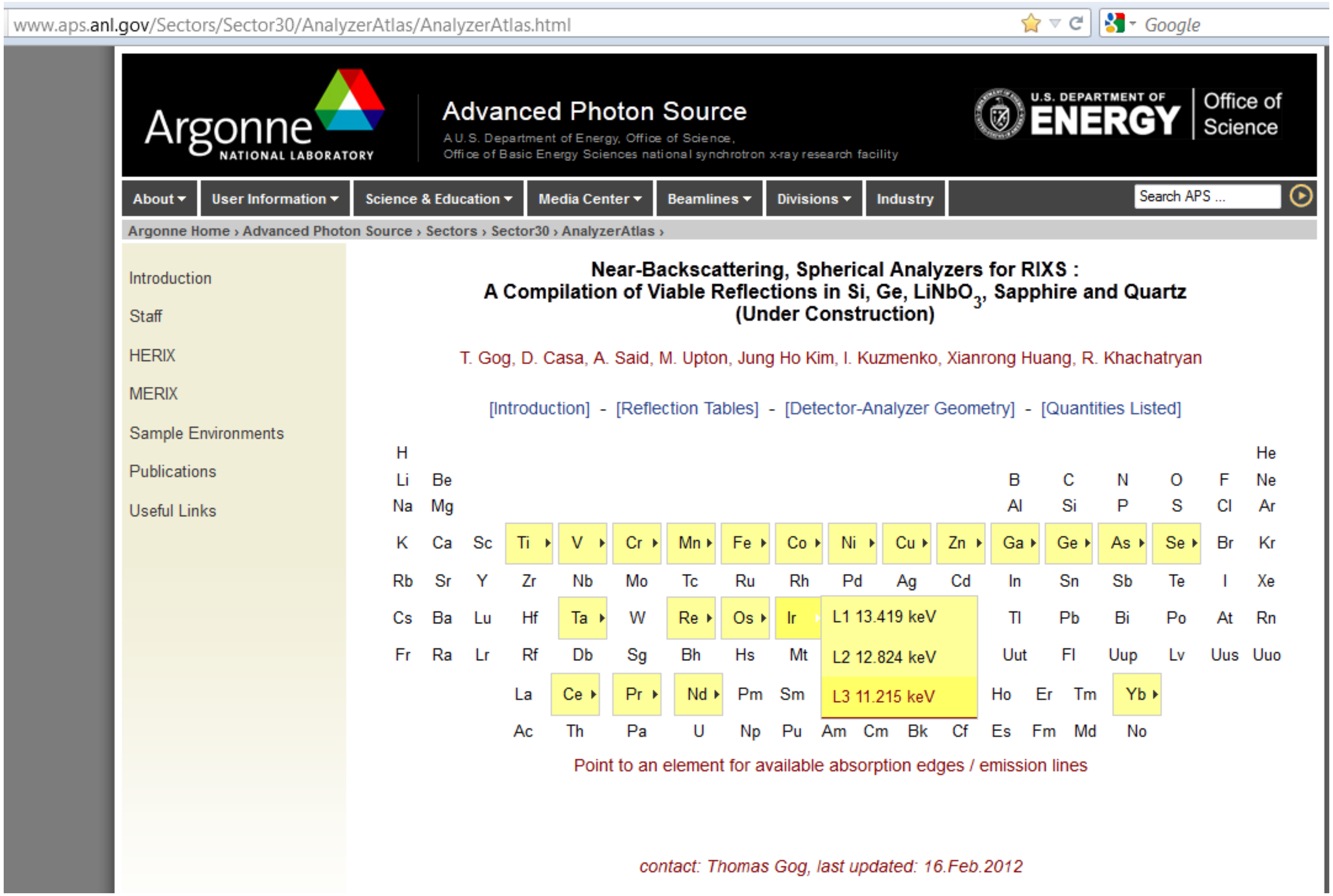




RIXS Upgrade: Energy Resolution: Reflections

Compilation of viable Reflections in Si, Ge, Sapphire, $LiNbO_3$, Quartz

- www.aps.anl.gov/Sectors/Sector30/AnalyzerAtlas/Analyzer Atlas.html



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Ei =	8.9805	keV						ΔEg	ΔEt
Cryst	Refl (h,k,l)	EB [keV]	ΘB [°]	∫IR dΘ [μrad]	Width [μrad]	Ei cotΘ [meV/μrad]	ΔE [meV]	2m, 50μm [meV]	2m, 50μm [meV]
Ge	(3,3,7)	8.969	87.14	80.2	81.5	0.448	36.51	5.6	36.94
Ge	(0,0,8)	8.766	77.46	30.3	28.2	1.998	56.34	24.97	61.62
Si	(2,4,6)	8.542	72.02	13.1	11.5	2.915	33.48	36.44	49.48
Si	(1,3,7)	8.768	77.5	11.8	10.8	1.991	21.56	24.89	32.93
	Equiv. Refl.: (3,5,5)								
LiNbO3	(1,5,-10)	8.941	84.6	58.1	55.8	0.85	47.45	10.62	48.62
	Equiv. Refl.: (5,-6,-10), (6,-5,-10), (-1,6,-10), (-5,-1,-10), (-6,1,-10)								
LiNbO3	(1,-6,10)	8.941	84.6	56.4	55.8	0.85	47.45	10.62	48.62
	Equiv. Refl.: (5,1,10), (6,-1,10), (-1,-5,10), (-5,6,10), (-6,5,10)								
Quartz	(-4,6,4)	8.972	87.44	37.5	34.4	0.401	13.78	5.01	14.66
	Equiv. Refl.: (-4,-2,-4)								
Quartz	(6,-2,4)	8.972	87.44	37.4	34.4	0.401	13.77	5.01	14.65
	Equiv. Refl.: (6,-4,-4), (-2,6,-4), (-2,-4,4)								
Quartz	(4,-6,-4)	8.972	87.44	36.3	34.4	0.401	13.78	5.01	14.66
Quartz	(2,4,-4)	8.972	87.44	36.2	34.4	0.401	13.77	5.01	14.66
	Equiv. Refl.: (2,-6,4), (4,2,4), (-6,2,-4), (-6,4,4)								
Quartz	(6,-2,-4)	8.972	87.44	28.5	26.9	0.401	10.79	5.01	11.89

Summary

- HERIX: Doubling of flux -> immediate positive impact on user operation and science
- RIXS: Vibrant Science Program, currently focused on 5-d TMOs, Iridates
- Consolidation of RIXS on one optimized, dedicated ID beamline
- Improvement of Energy Resolution to 10 meV … 20 meV, and better **CRITICAL !**
  - Spherical Analyzers
  - Flat Crystal Optics
- Critical Enabling Technology: Sapphire , $LiNbO_3$, Quartz Spherical Analyzers
- In-situ Sample Environments

MBA Perspectives for IXS

Improved brilliance of MBA machine will allow nano-scale focusing

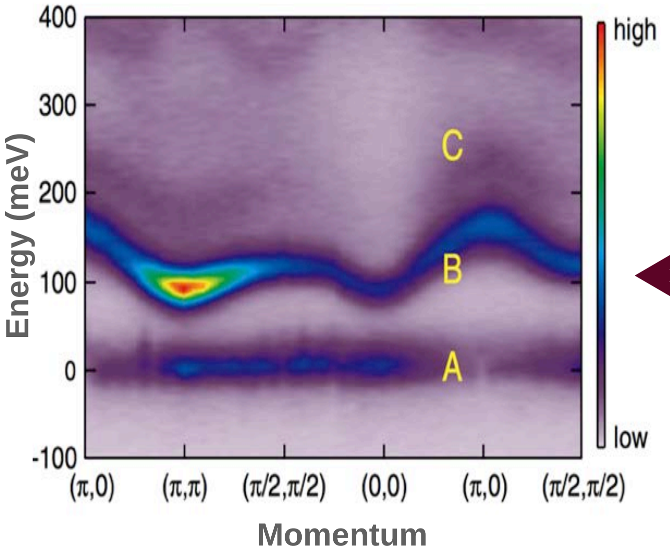
- RIXS: Improvement of energy resolution
- IXS: Enables study of thin-films, samples under high-pressure
- Very small samples (femtomole science)
- Imaging of inelastic texture

Coherence ?

Nano Hard-Xray Resonant Inelastic Scattering (RIXS)

- Electronic and magnetic excitations in transition metal oxides and other novel materials can now be probed with RIXS with unprecedented energy resolution
- DLSR sources, providing nanoscale beams, will further enable RIXS measurements mapping the intrinsic electronic and magnetic texture in a sample.

RIXS spectra of Magnons in Sr_2IrO_4



Superconducting Gap in $Bi_2Sr_2CaCu_2O_{8+\delta}$