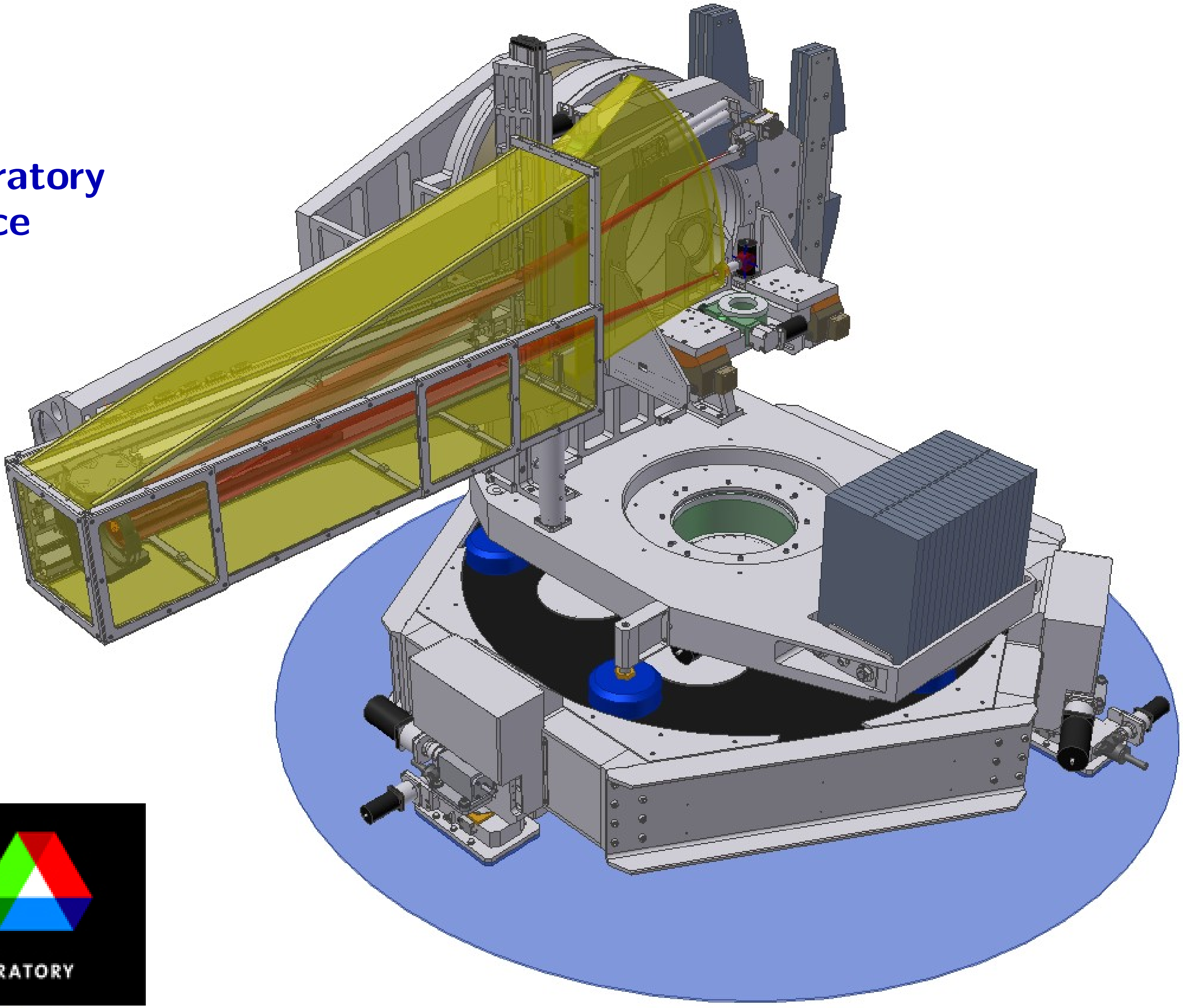
RIXS Spectroscopy with MERIX



Yuri Shvyd'ko

Argonne National Laboratory
Advanced Photon Source

Argonne NATIONAL LABORATORY

MERIX Team


John Hill
(BNL)


Scott Coburn
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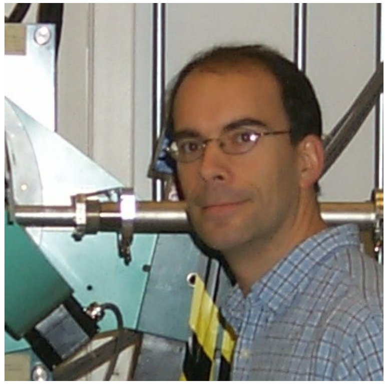
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Content

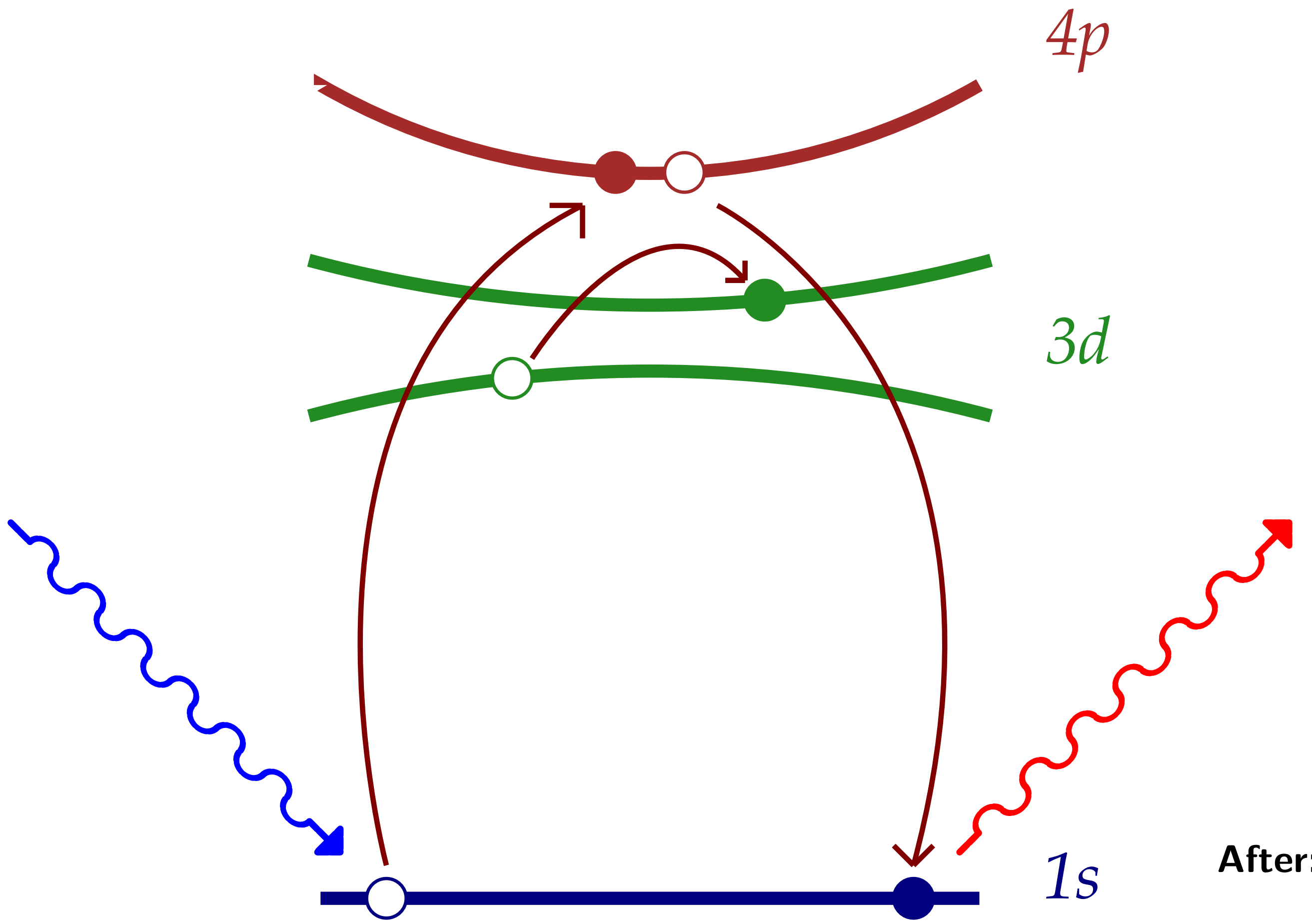
- Scientific Mission.
- Overview of the instrument.
- Next generation x-ray optics and detectos for K-edge RIXS:
  Better energy resolution.
  Count rates increased by a factor of > 10.
- Examples of K-edge RIXS Spectroscopy with Cu, Ni, and V.
- Outlook

Scientific Mission of MERIX

- **Purpose: study of electronic excitations.**
- **Technique: resonant IXS (RIXS) and non-resonant IXS.**
- **Incident photon energies 4 - 14 keV**
  (K-edges of 3d elements, L-edges of 4f elements, M-edges of U).
- **Energy resolution about 100 meV.**
- **Polarization dependence - scattering in horizontal and vertical plane.**
- **Increased flux and countrates, improved analyzer and detector design.**
- **Sample environments:**
  low and high temperatures, high pressure, magnetic field.

K-edge RIXS

RIXS: Photon-in Photon-out spectroscopy.
No charge particles enter or leave the sample.



Energy losses are due to a "shake-up" process of the valance $3d$ electrons, in between the creation and annihilation of the $1s-4p$ core exciton.

After: K. Tsutsui, T. Toyama, and S. Maekawa PRL 91, 117001 (2003)

RIXS vs. XAS, PES, ARPES, EELS

RIXS: captures the physics of charge dynamics by observation of momentum-dependent excitations across the gap.

- **Probes excited electronic states (charge-transfer, $d-d$, etc. excitations)**
- **Momentum-dependent information.**
- **Absence of the deep core hole effects (no lifetime broadening).**
- **Bulk sensitive.**
- **Element specific.**
- **Resonantly enhanced.**
- **Applicable to metals and insulators (photon-in, photon-out).**
- **Allows studies under extreme conditions (high pressure, magnetic fields, extreme temperatures, etc.).**

Scientific Mission of MERIX

- **Purpose: study of electronic excitations.**
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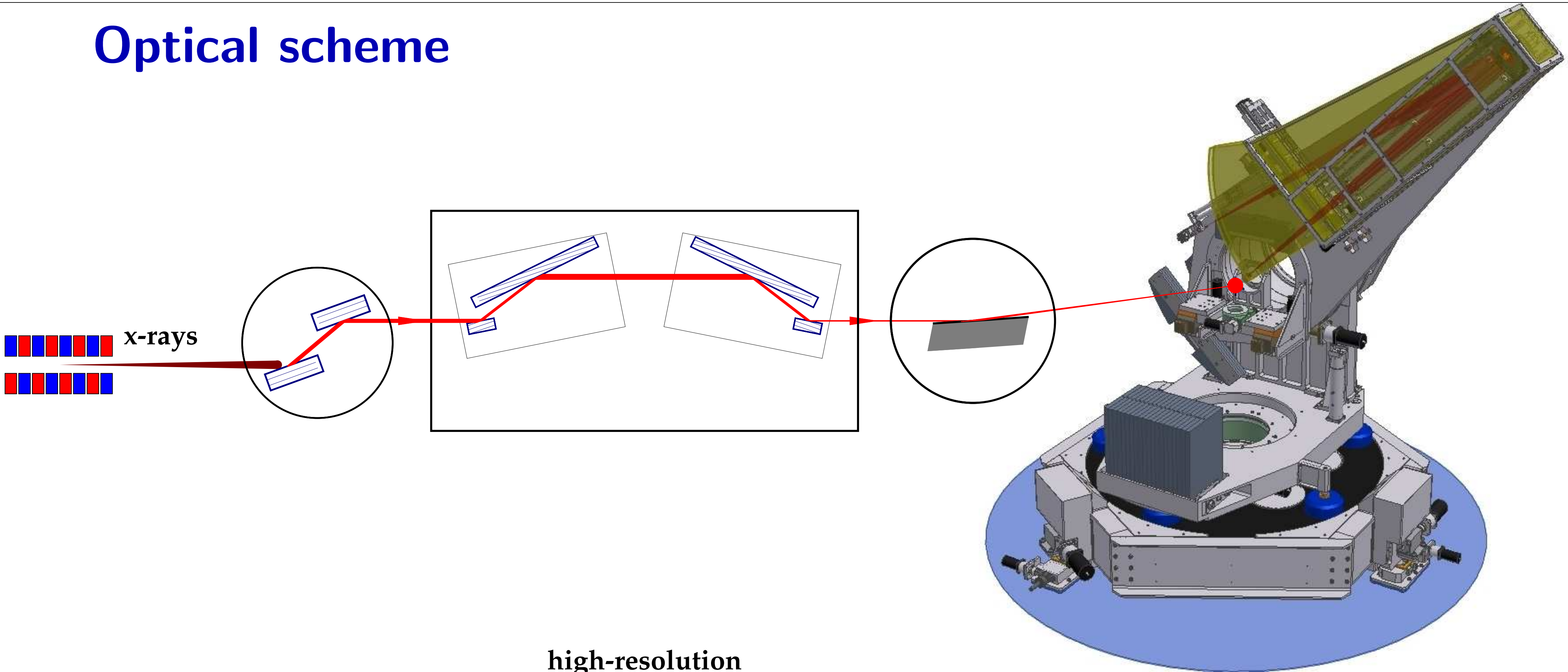
  low and high temperatures, high pressure, magnetic field.

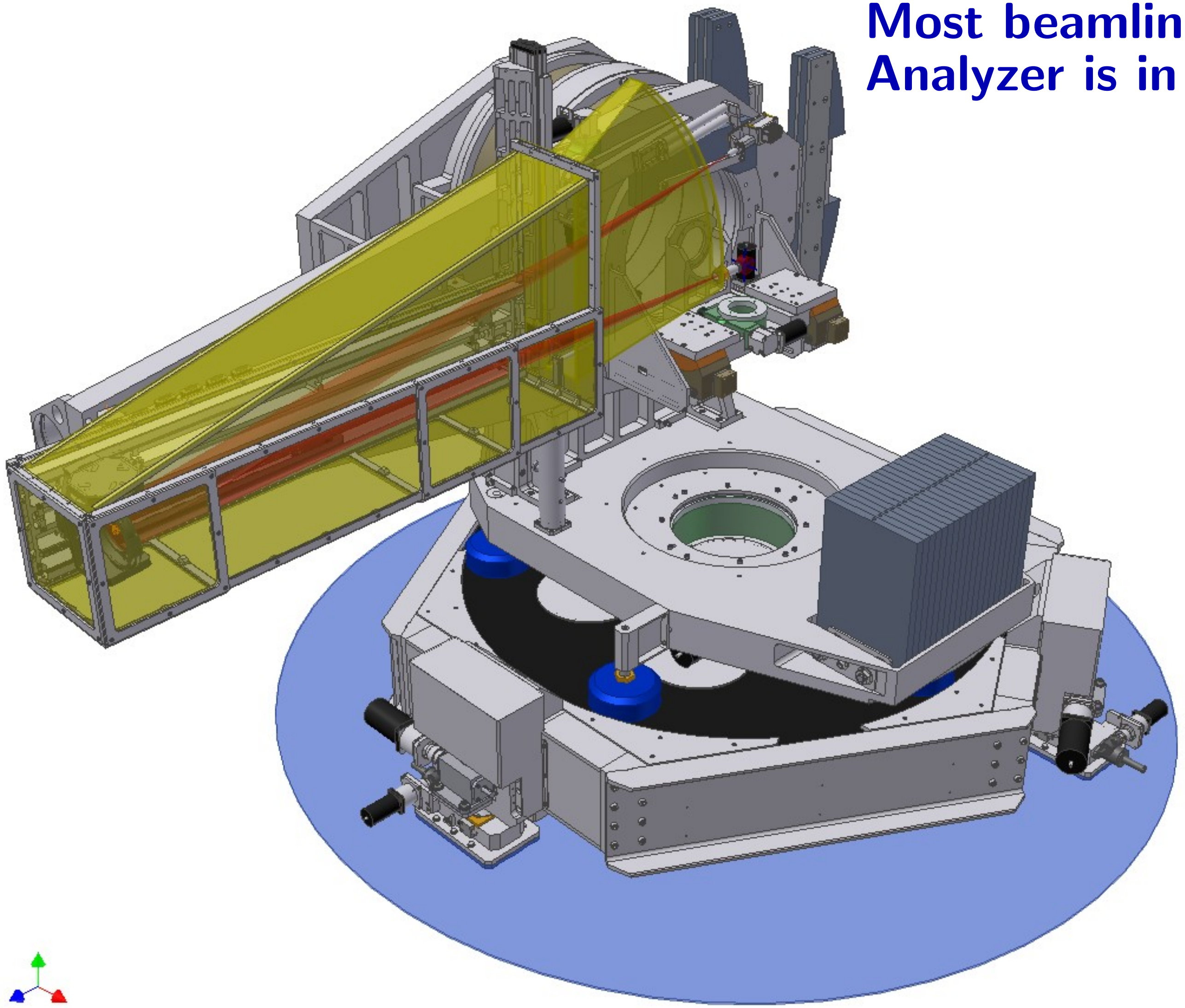
Layout of the Beamline 30-ID-XOR

Optical scheme



undulator	cooled monochromator	high-resolution monochromator 5 - 12 keV	KB focusing mirror	MERIX spectrometer & sample
bandwidth $\approx$100 eV	bandwidth $\approx$0.6 eV	bandwidth $\approx$ 72 or 120 meV	focus 5 μm $\times$45 μm	bandwidth $\approx$ 60 - 120 meV
	3×10^{13} Hz @ 9 keV		1.2×10^{12} Hz/72 meV @ 9 keV	

MERIX Spectrometer - Design



Most beamline components are in-vacuum
Analyzer is in helium tank

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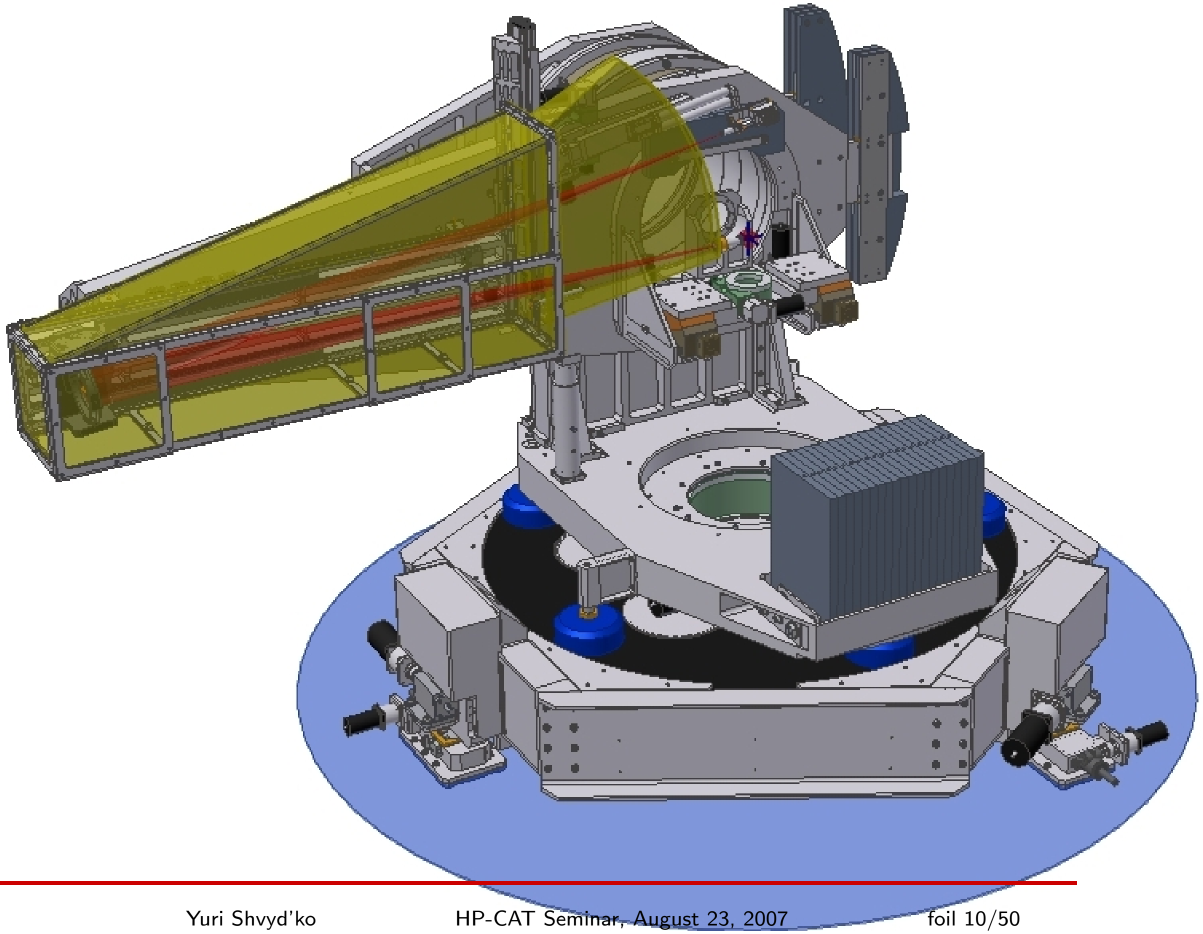
MERIX Design

Vertical Scattering Geometry



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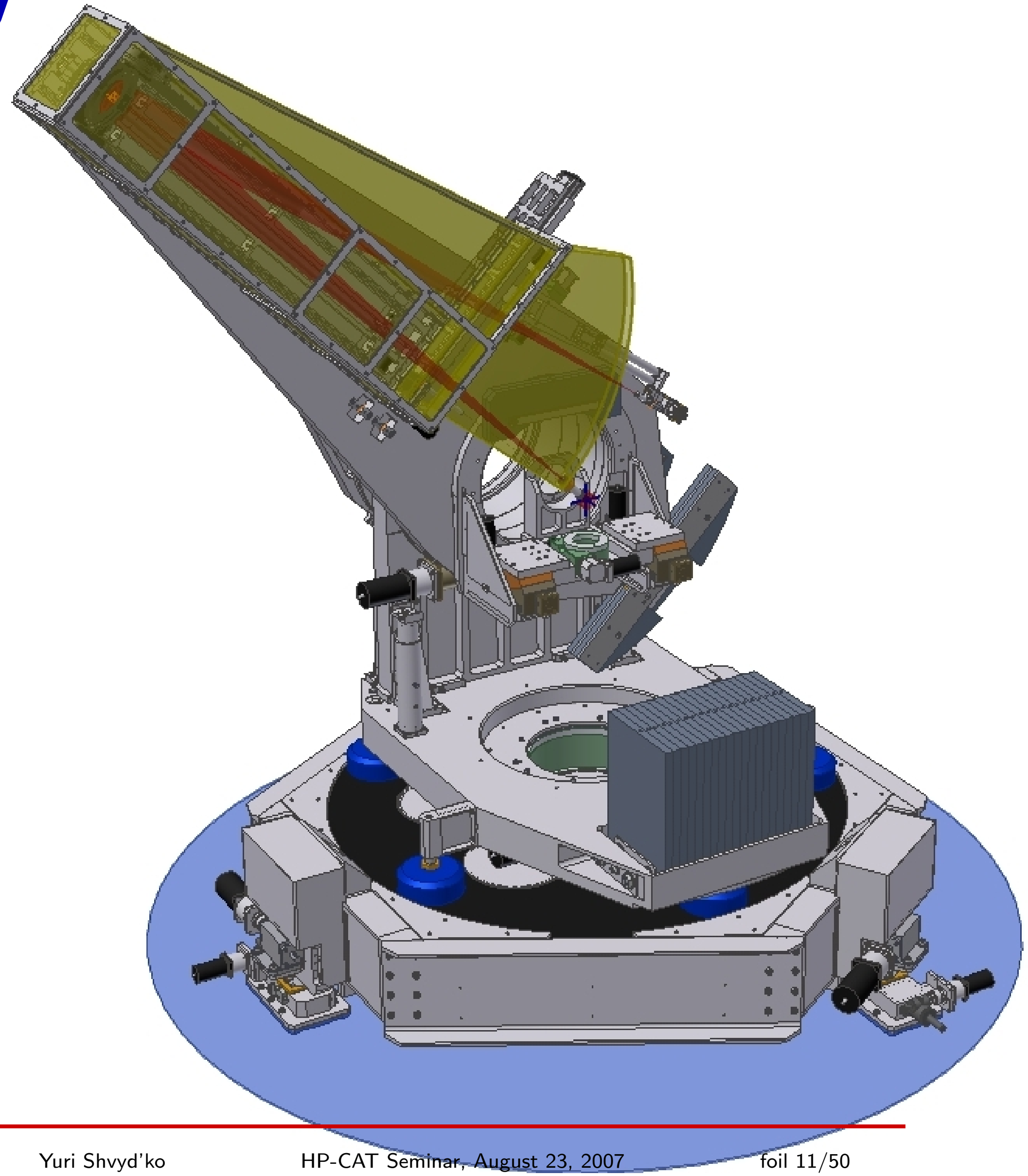

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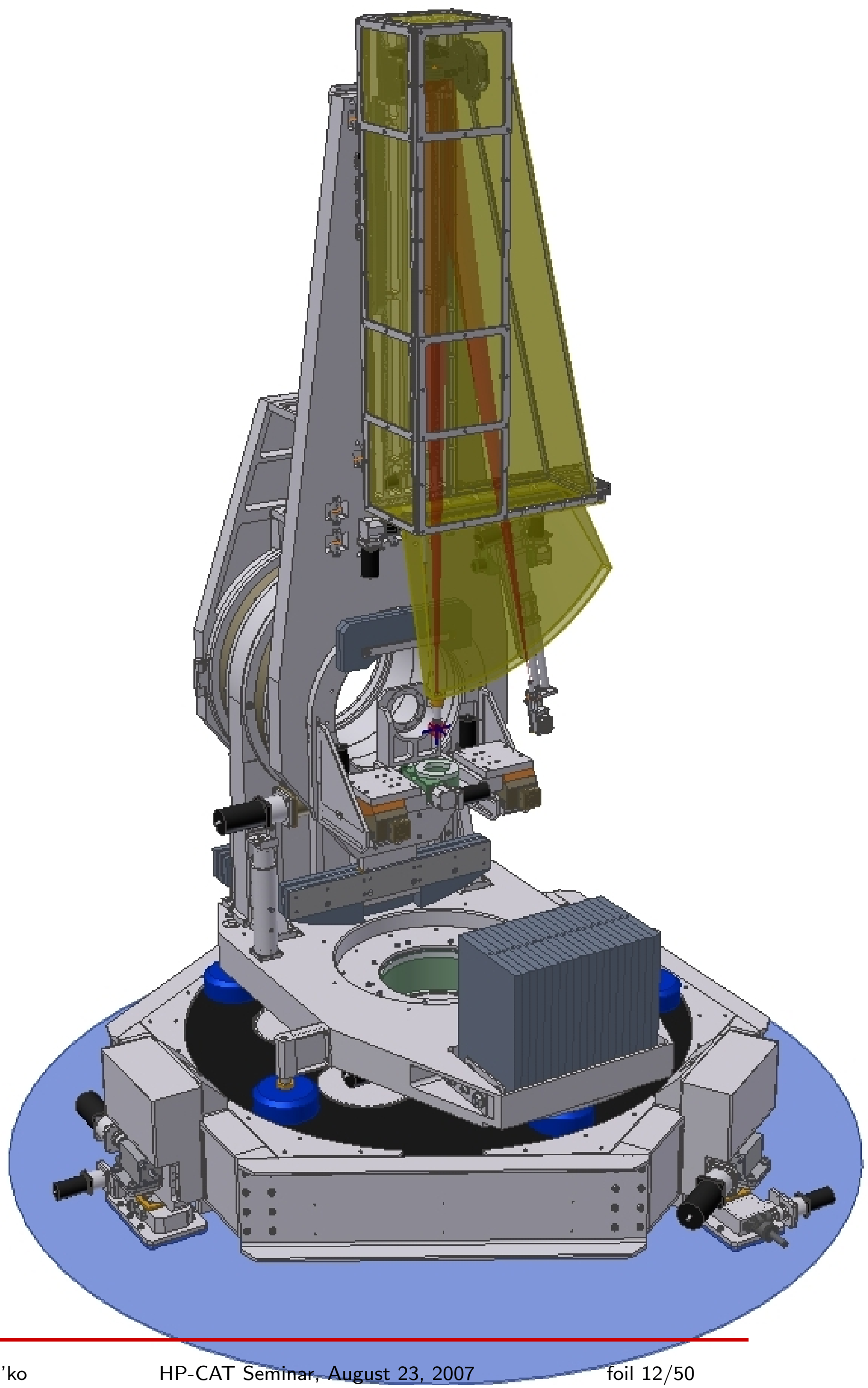
Vertical Scattering Geometry



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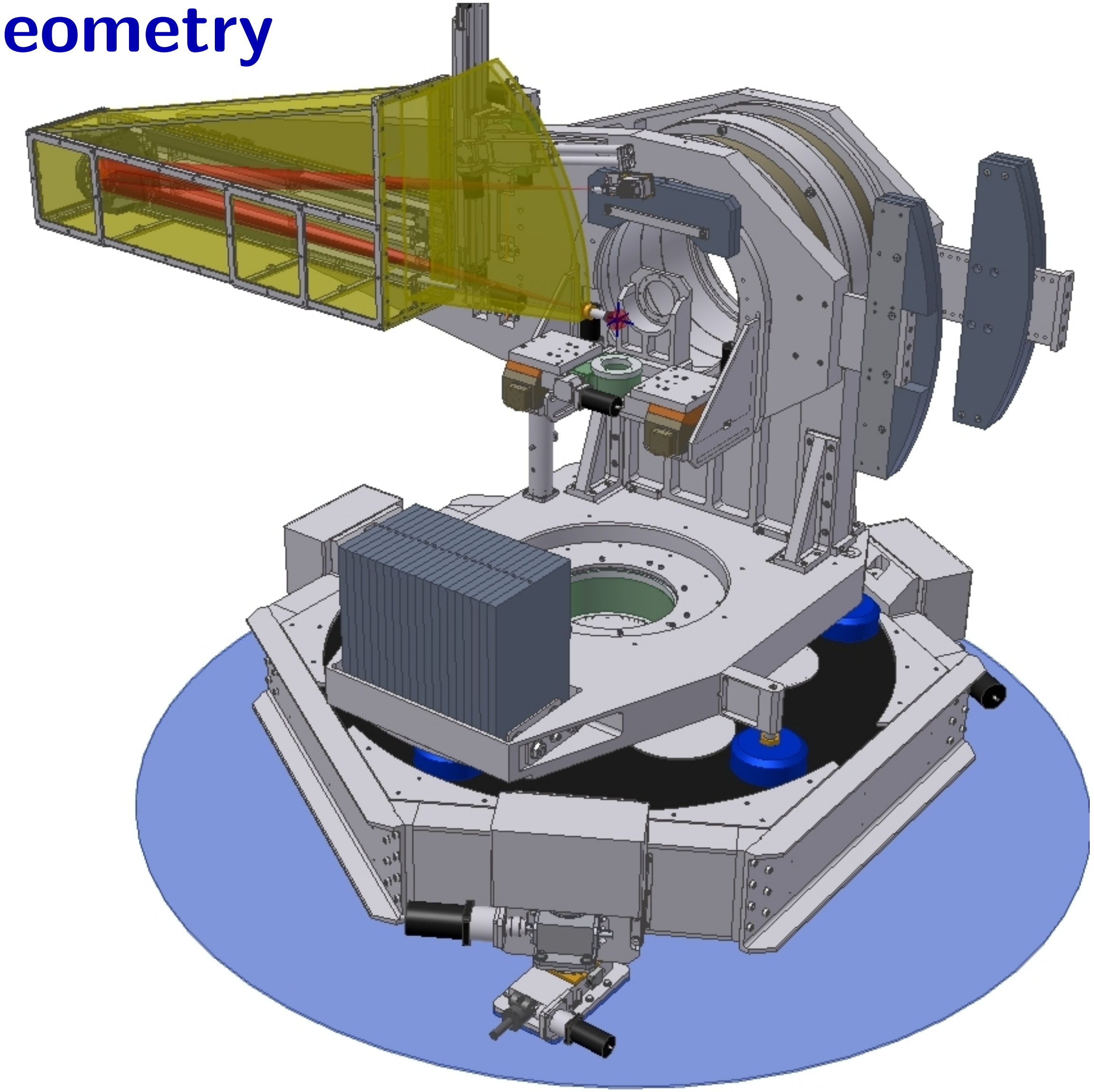
MERIX Design

Horizontal Scattering Geometry



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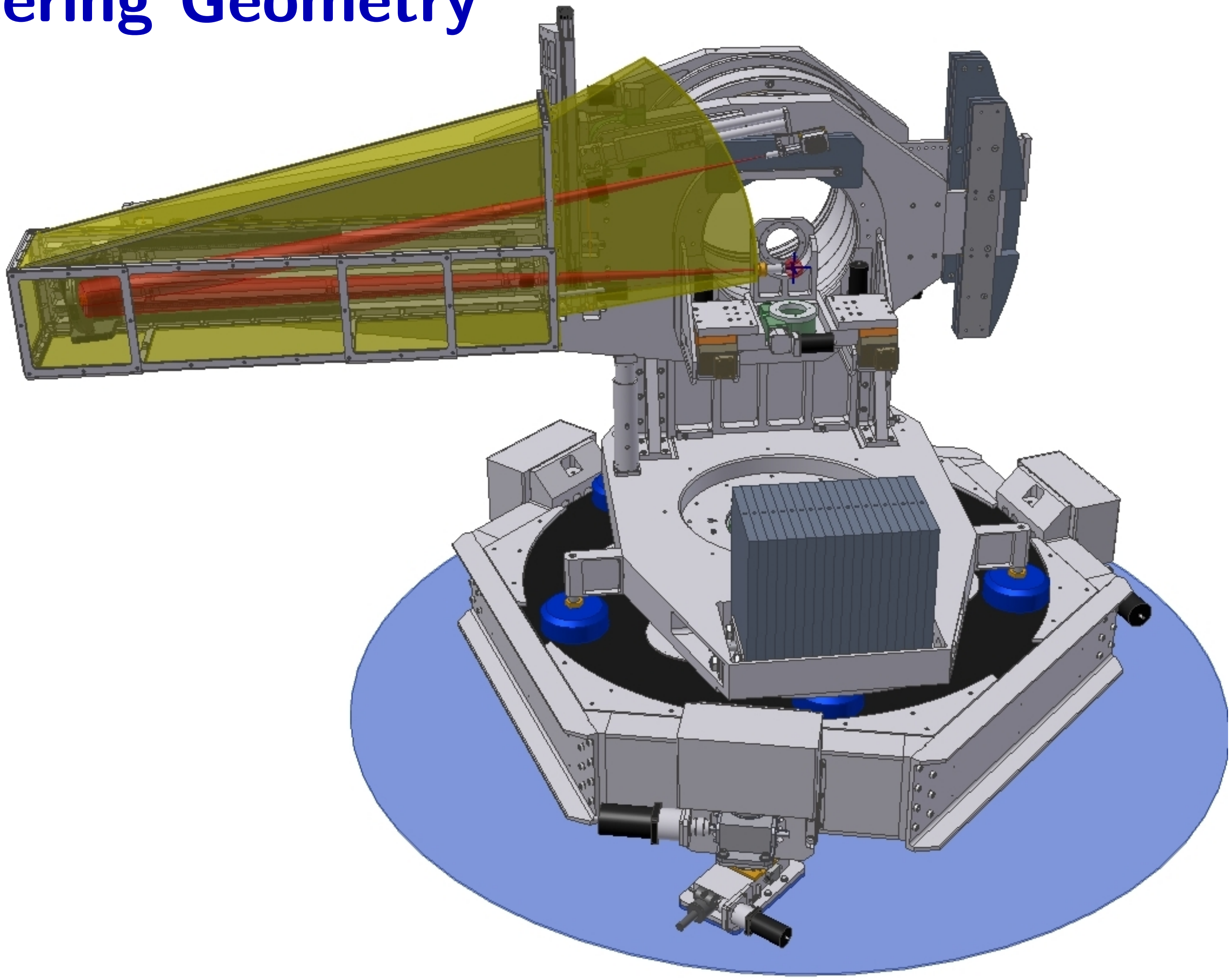
MERIX Design

Horizontal Scattering Geometry



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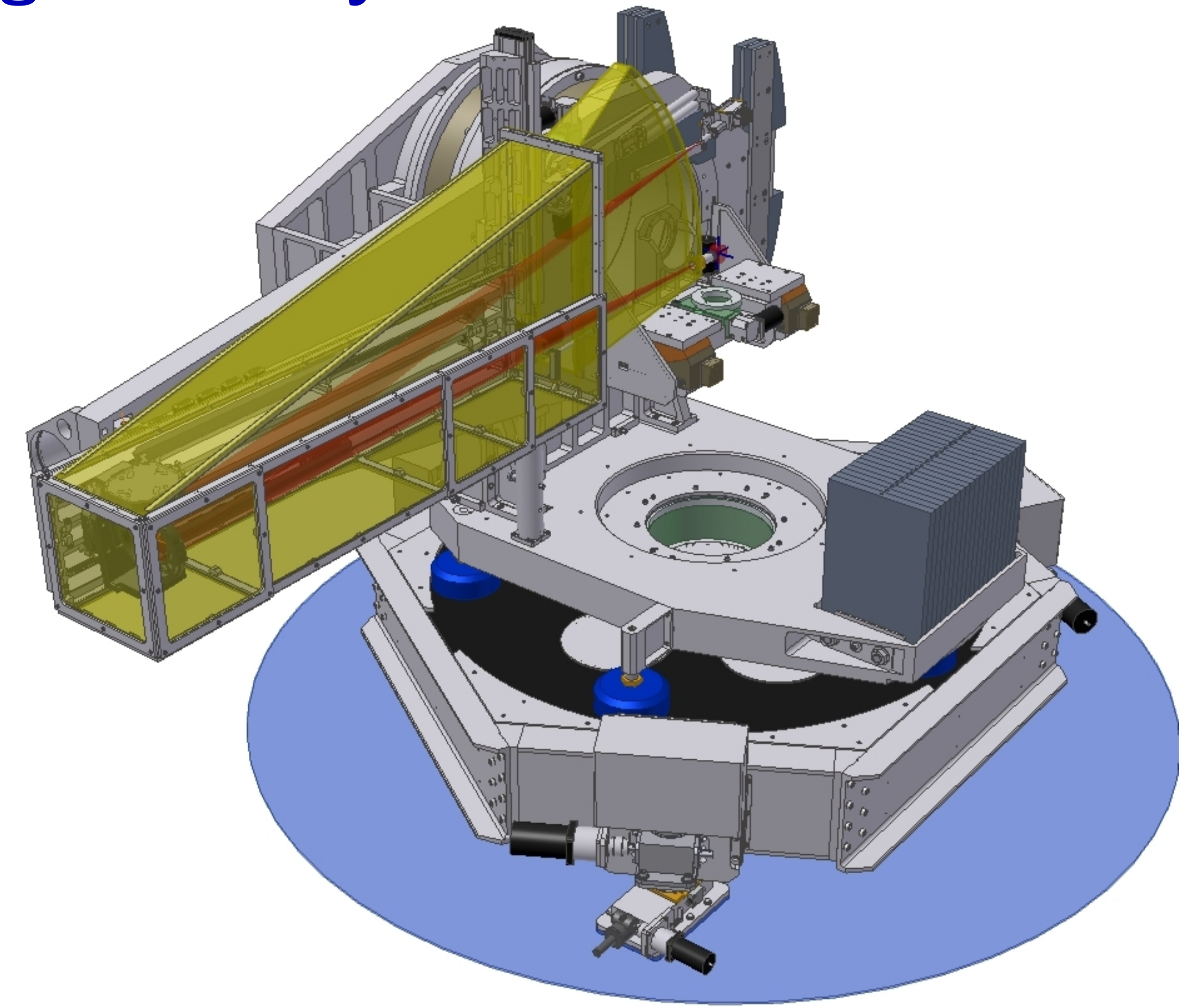
MERIX Design

Horizontal Scattering Geometry



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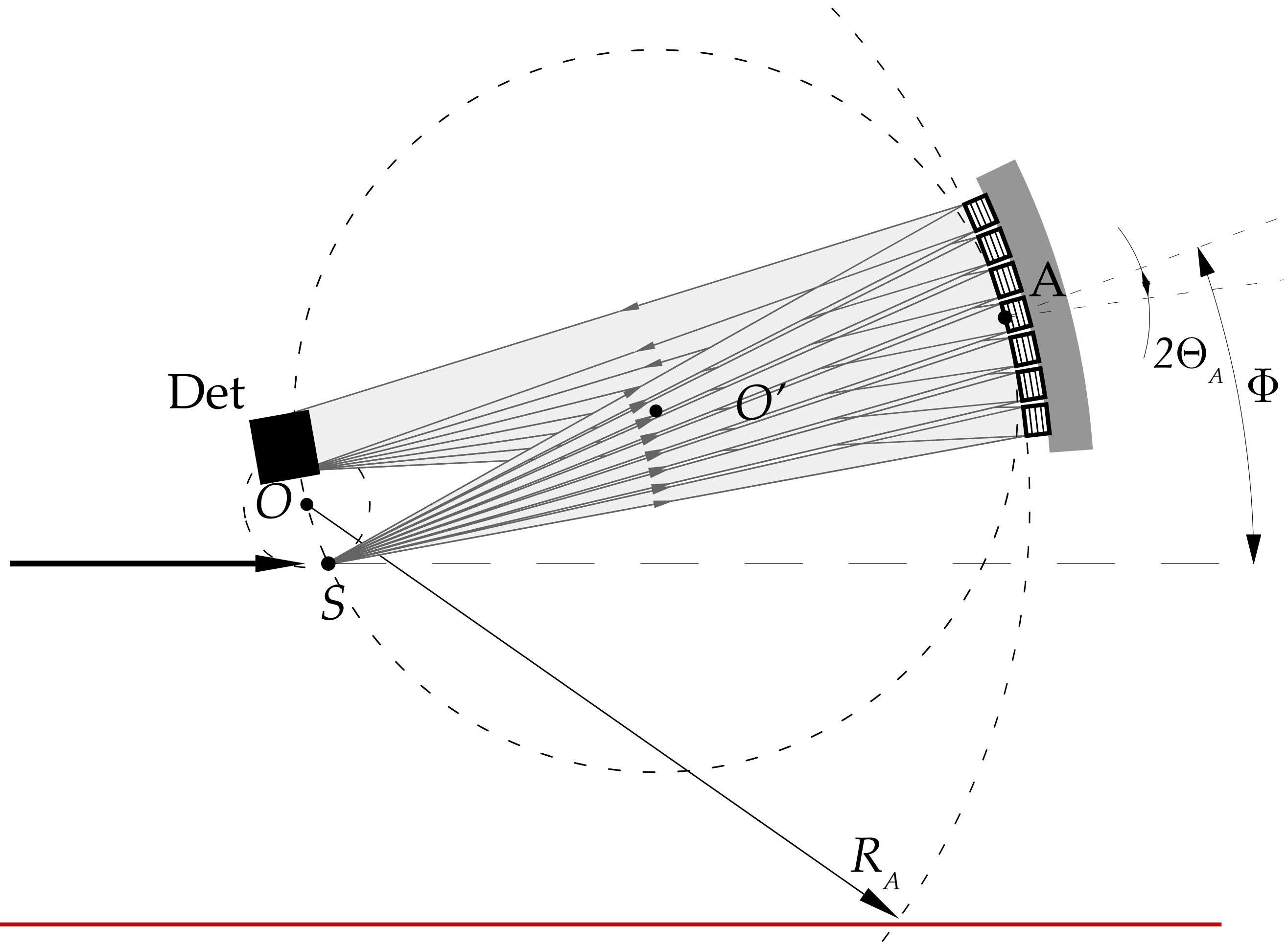
MERIX optical scheme

Scheme of gathering, focusing, and spectral analysis of x-rays by a segmented spherical crystal analyzer.



MERIX optical scheme

Scheme of gathering, focusing, and spectral analysis of x-rays

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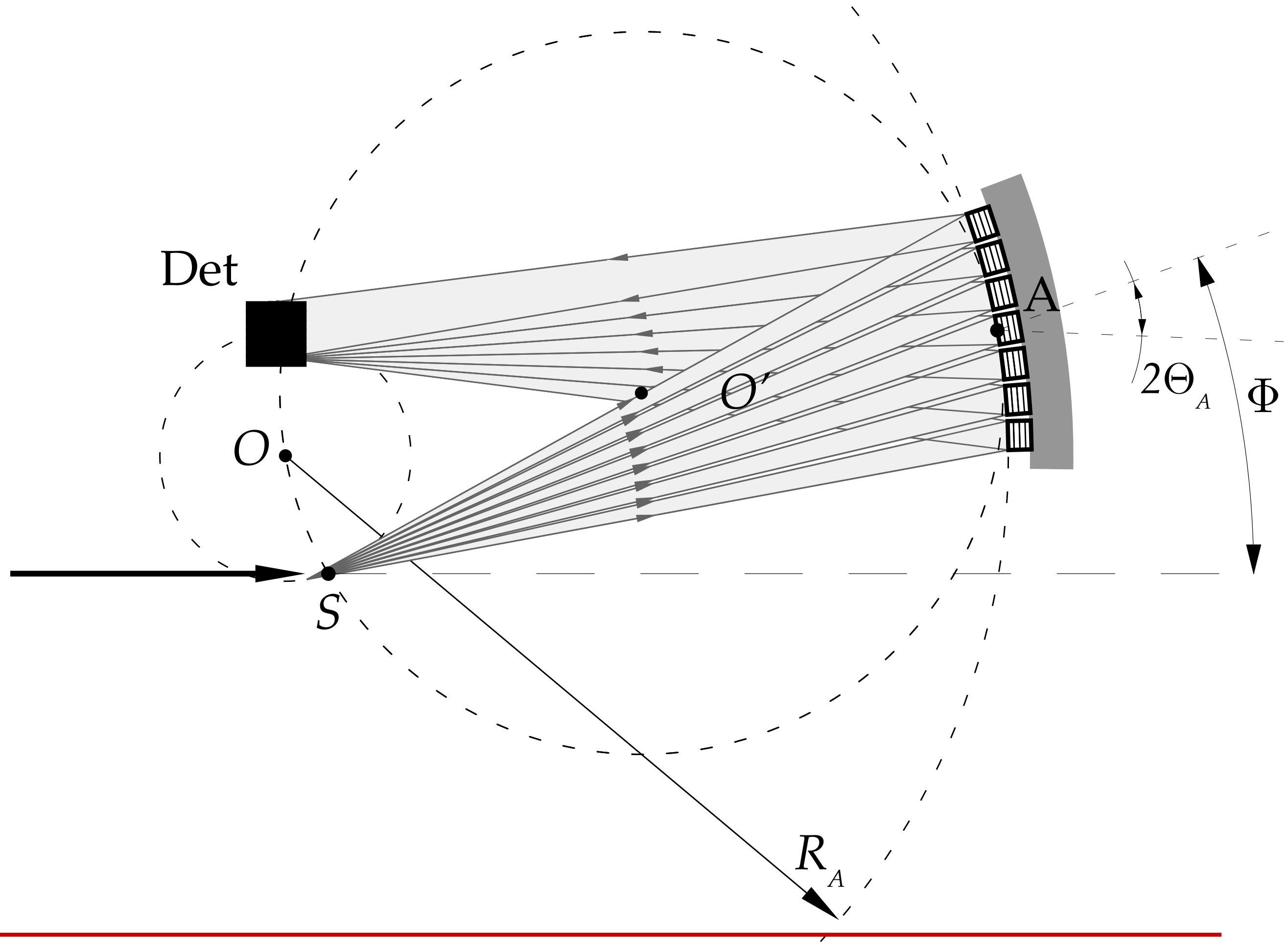


Analyzer Resolution & Geometrical Broadening

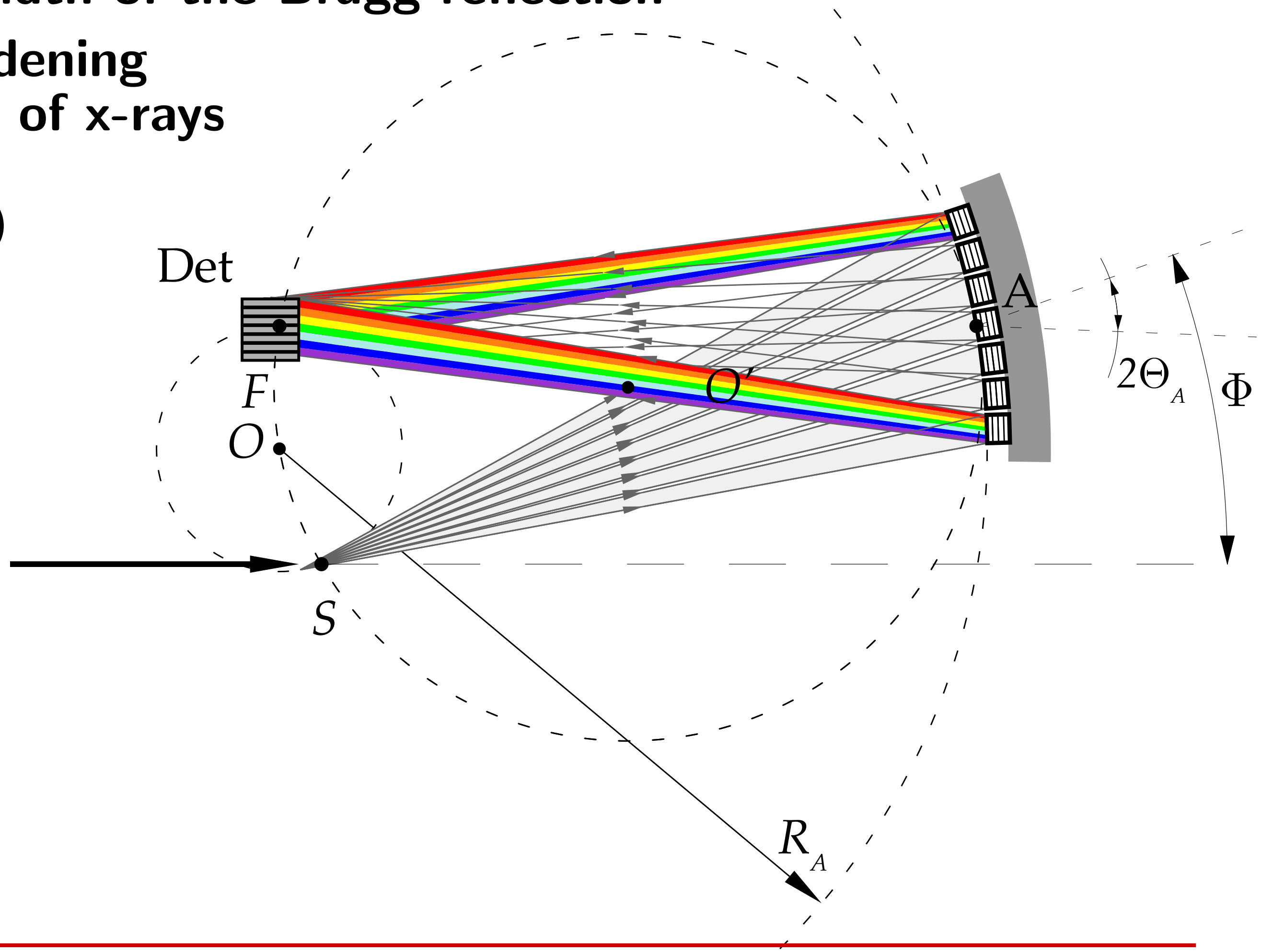
$$\Delta E_{\mathrm{tot}} = \sqrt{\Delta E_{\mathrm{i}}^2 + \Delta E_{\mathrm{g}}^2}$$

$\Delta E_{\mathrm{i}} =$ **intrinsic (Darwin) width of the Bragg reflection**

$\Delta E_{\mathrm{g}} =$ **“geometrical” broadening due to angular spread of x-rays**

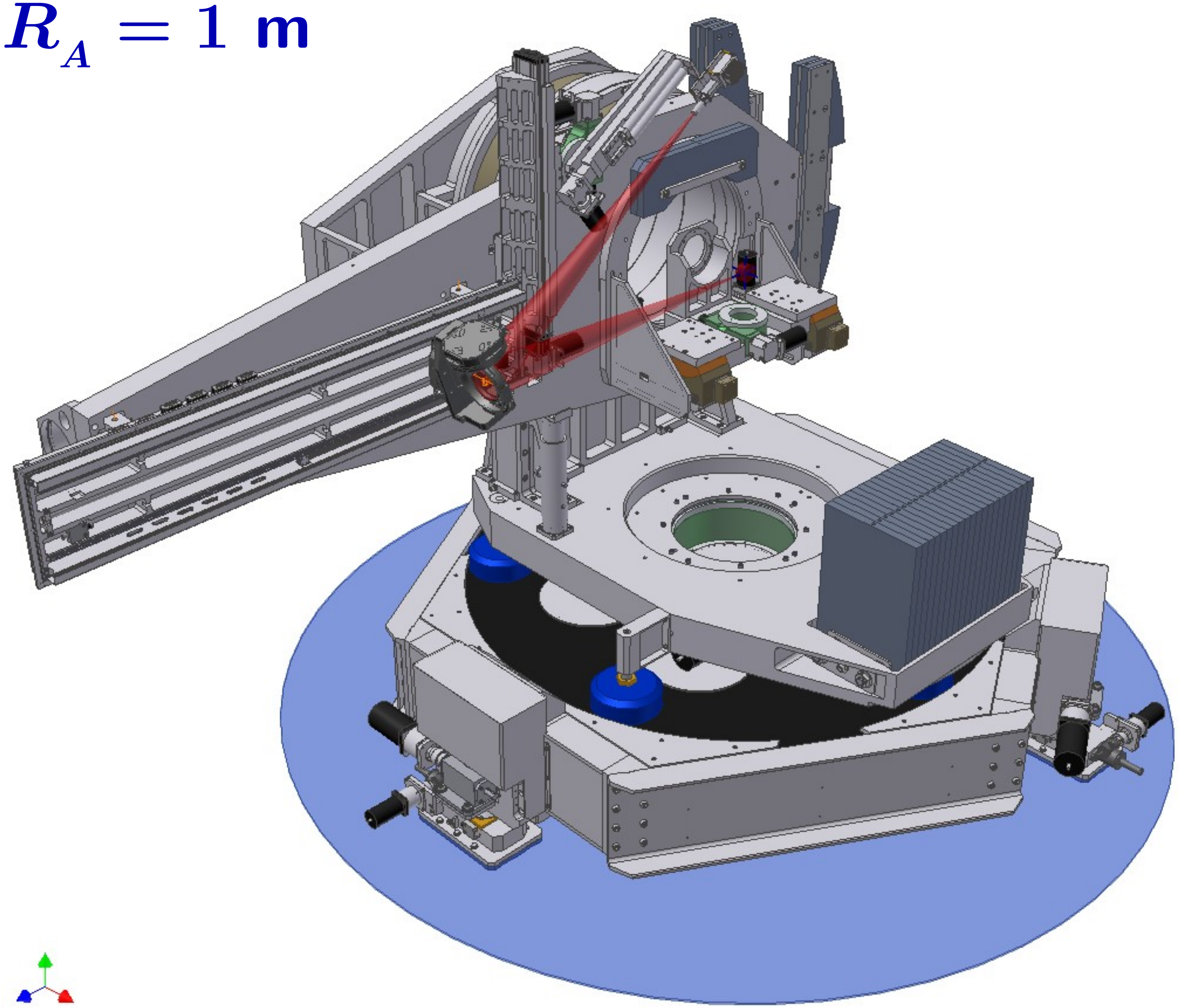
$$\Delta E_{\mathrm{g}} = E \cos\theta_{\mathrm{B}}\, \Delta d/(2R_{\mathrm{A}})$$

$\Delta d =$ **crystal segment**



MERIX Spectrometer - Design

$R_A = 1$ **m**



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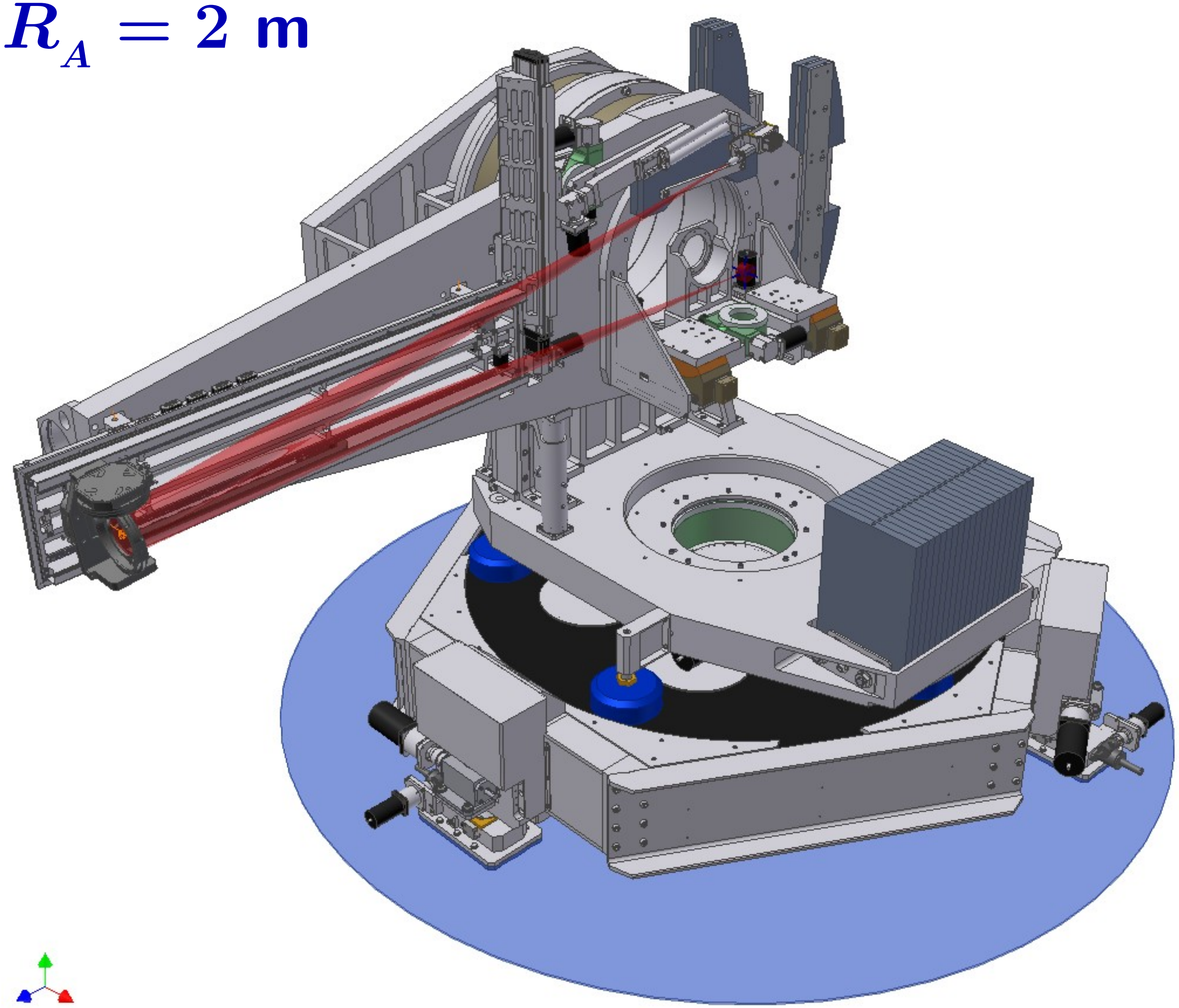
MERIX Spectrometer - Design

$R_A = 2$ m



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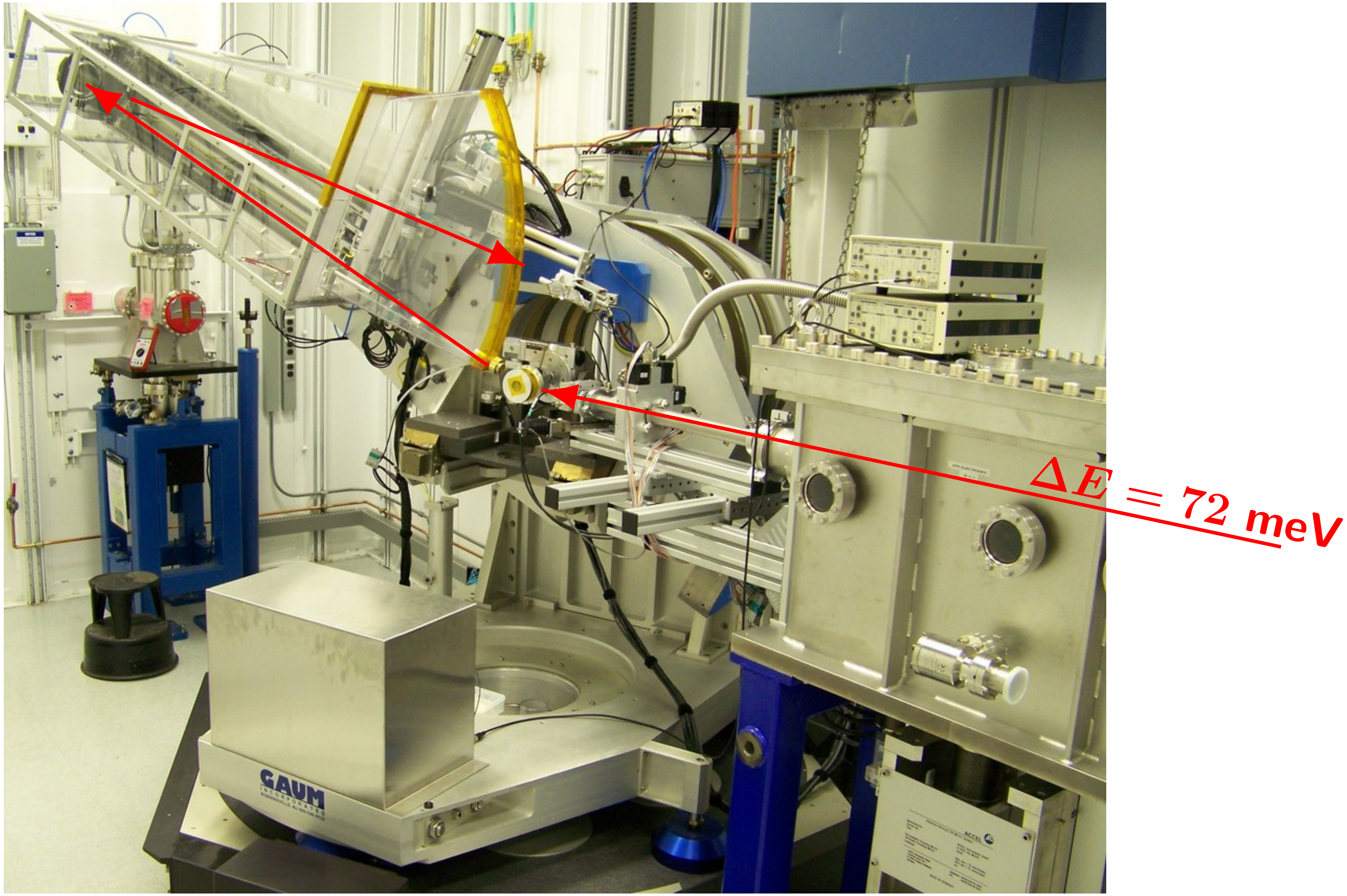
Possible RIXS Analyzers for MERIX

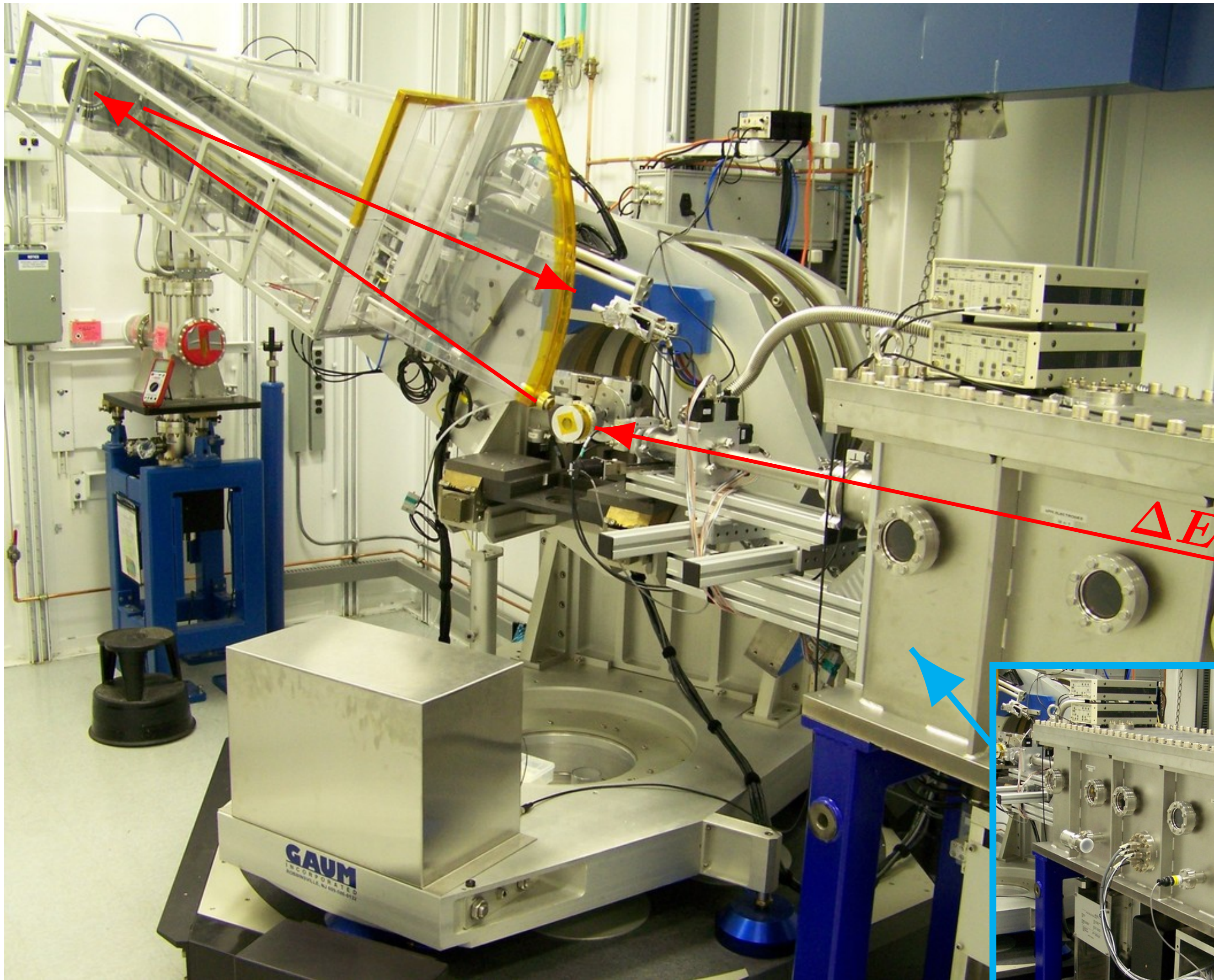
K-edge L-edge

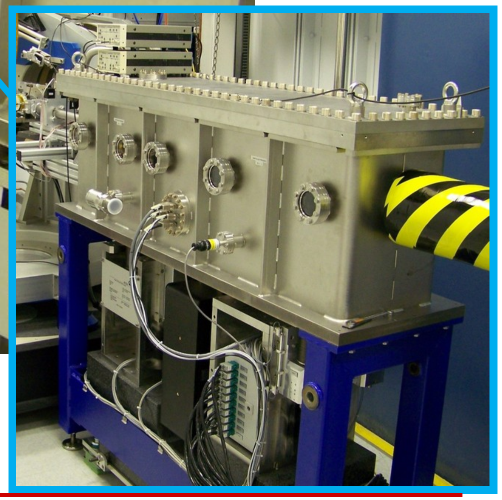
Element	E [keV]	Crystal	Reflection	θ_B [deg]	ΔE_i intr. [meV]
V(O)	5.480	$LiNbO_3$	$(0\ 0\ \bar{0}\ 12)$	82.5	109
Cr(O)	6.009	Si	(5 1 1)	80.7	52.2
Mn(O)	6.555	Si	(0 4 4)	80.0	62
Fe(O)	7.130	Ge	(6 2 0)	76.3	115
Co(O)	7.720	$LiNbO_3$	$(3\ 3\ \bar{6}\ 6)$	86.5	49
Ni(O)	8.345	$LiNbO_3$	$(0\ 6\ \bar{6}\ 0)$	87.9	50
		Ge	(2 4 6)	79.2	76
Cu(O)	8.990	Ge	(3 3 7)	85.8	42
Eu	6.977	Ge	(6 2 0)	83.3	112
Yb	8.944	Ge	(0 0 8)	77.5	64

MERIX Spectrometer@30-ID.APS, October 2006



MERIX Spectrometer@30-ID.APS, October 2006

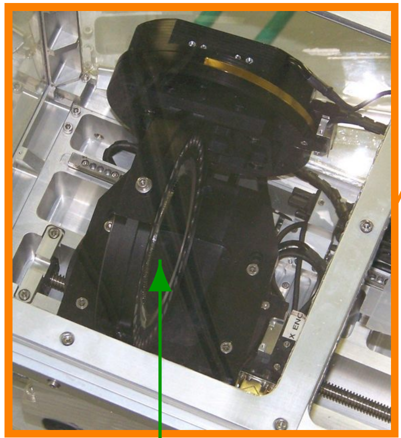


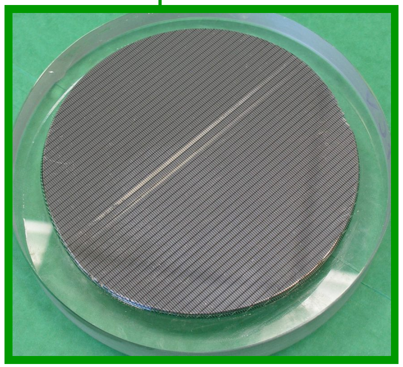


MERIX mirror.
Focus: $5\ \mu$m (V) $\times 40\mu$m (H)

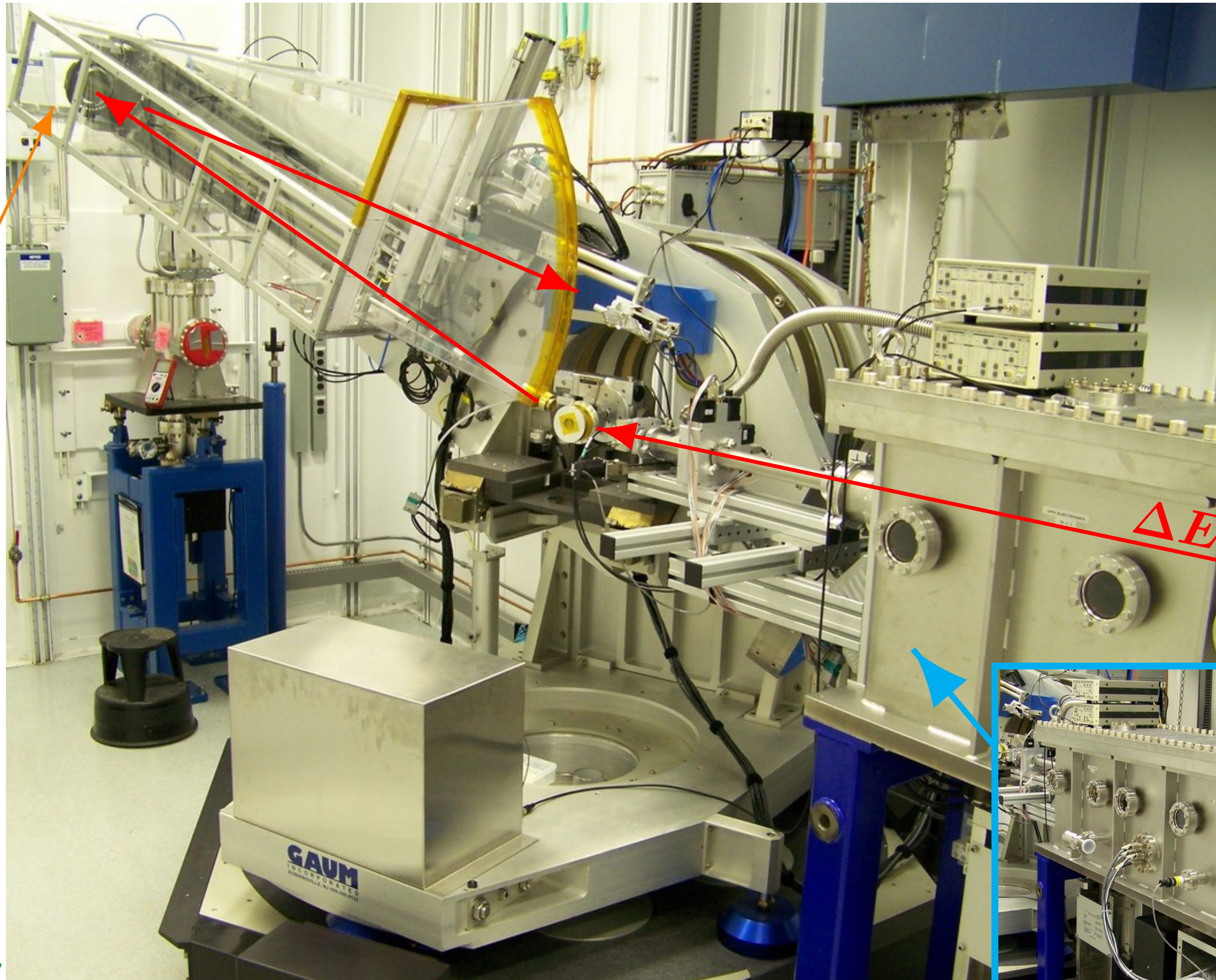
MERIX Spectrometer@30-ID.APS

Analyzer gimbal

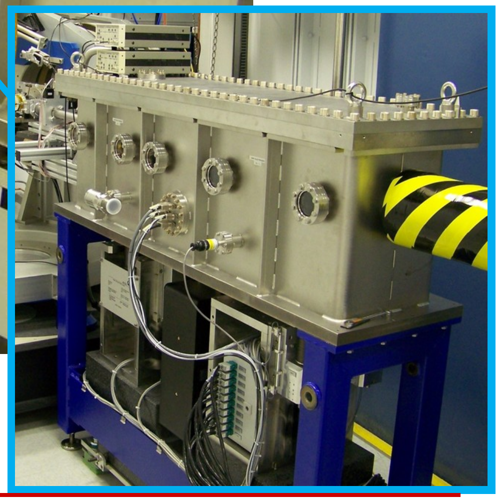




Ge(337)
diced analyzer:
$\Delta E = 120$ meV



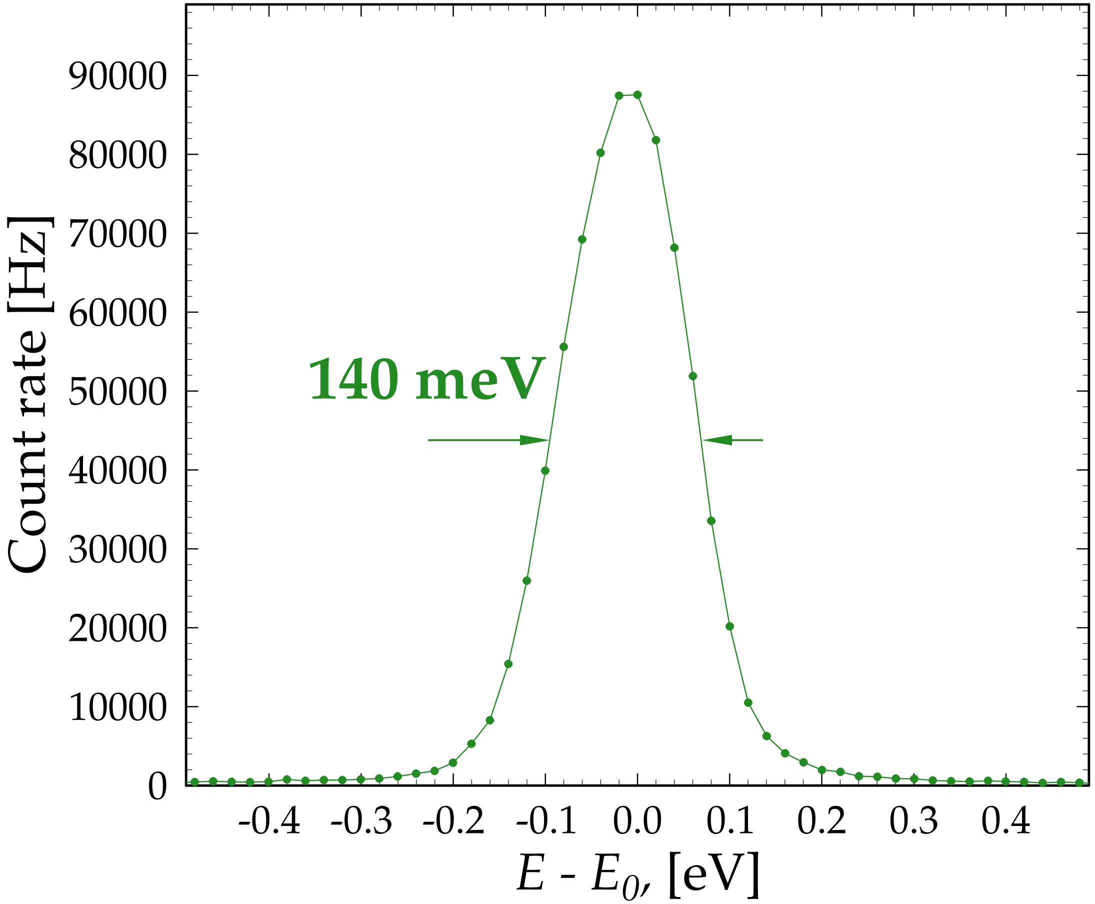
$\Delta E = 72$ meV

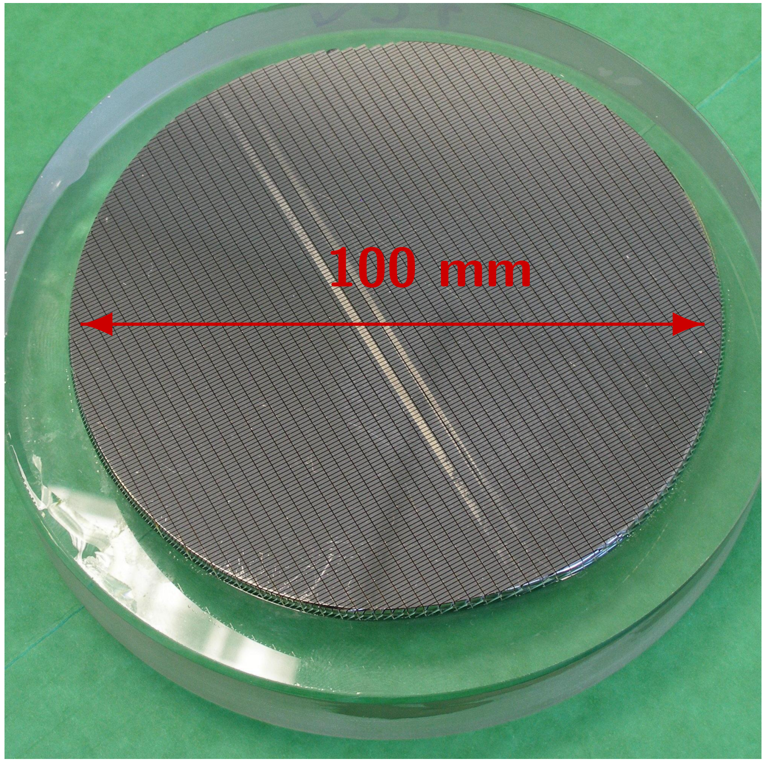


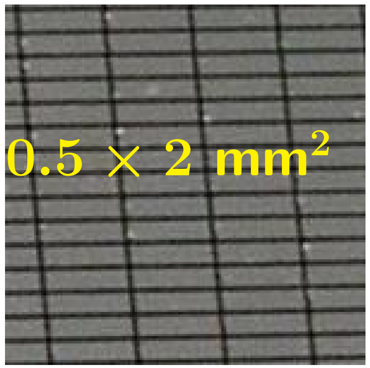
MERIX mirror.
Focus: $5\ \mu$m (V) $\times 40 \mu$m (H)

Cu K-edge RIXS Analyzer

Ge(3 3 7) analyzer
R_A = 2 m & conventional detector

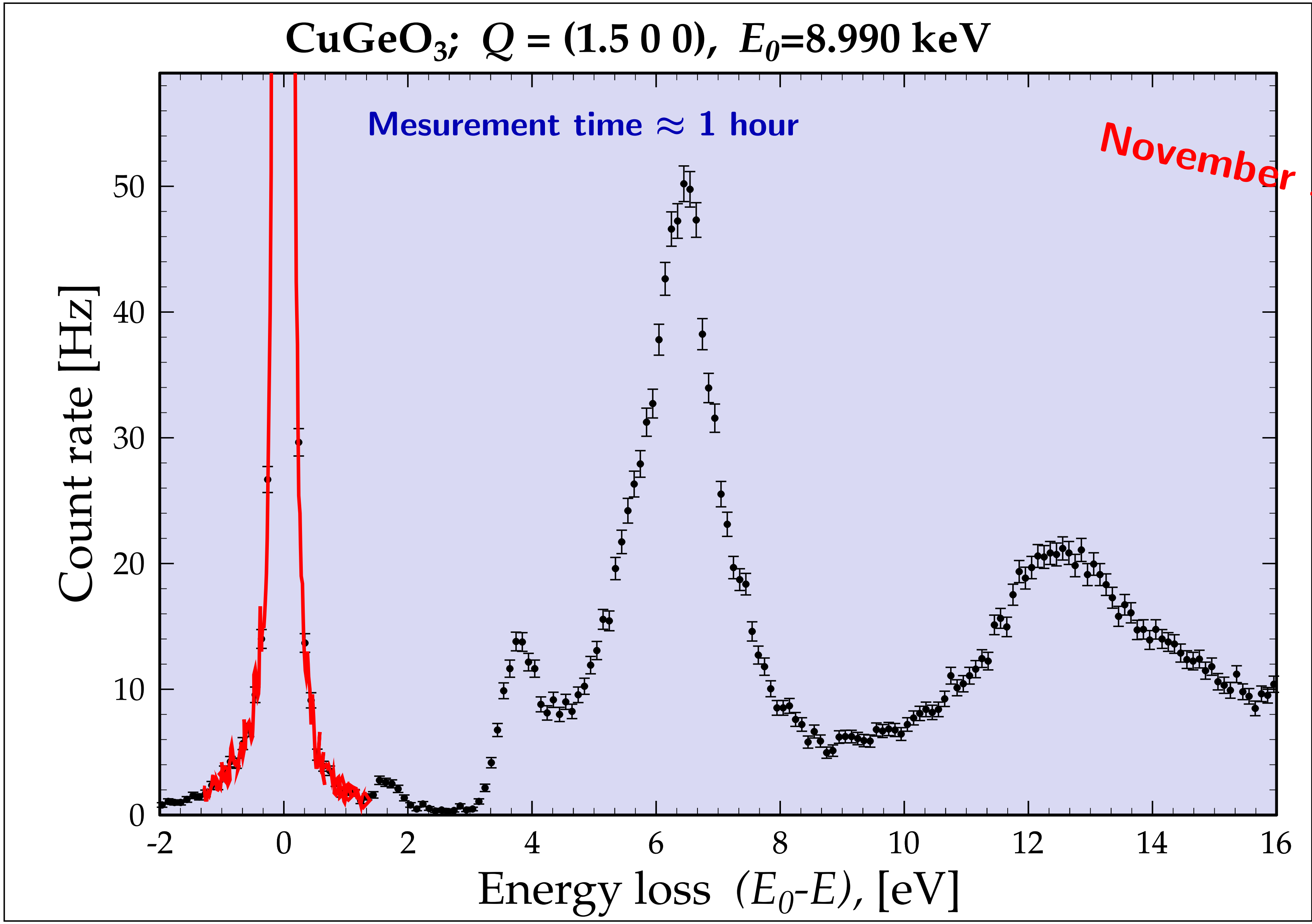






Expected: 115 eV
Deconvoluted width : 120 meV

First RIXS Spectrum Measured with MERIX



Analyzer Resolution & Geometrical Broadening

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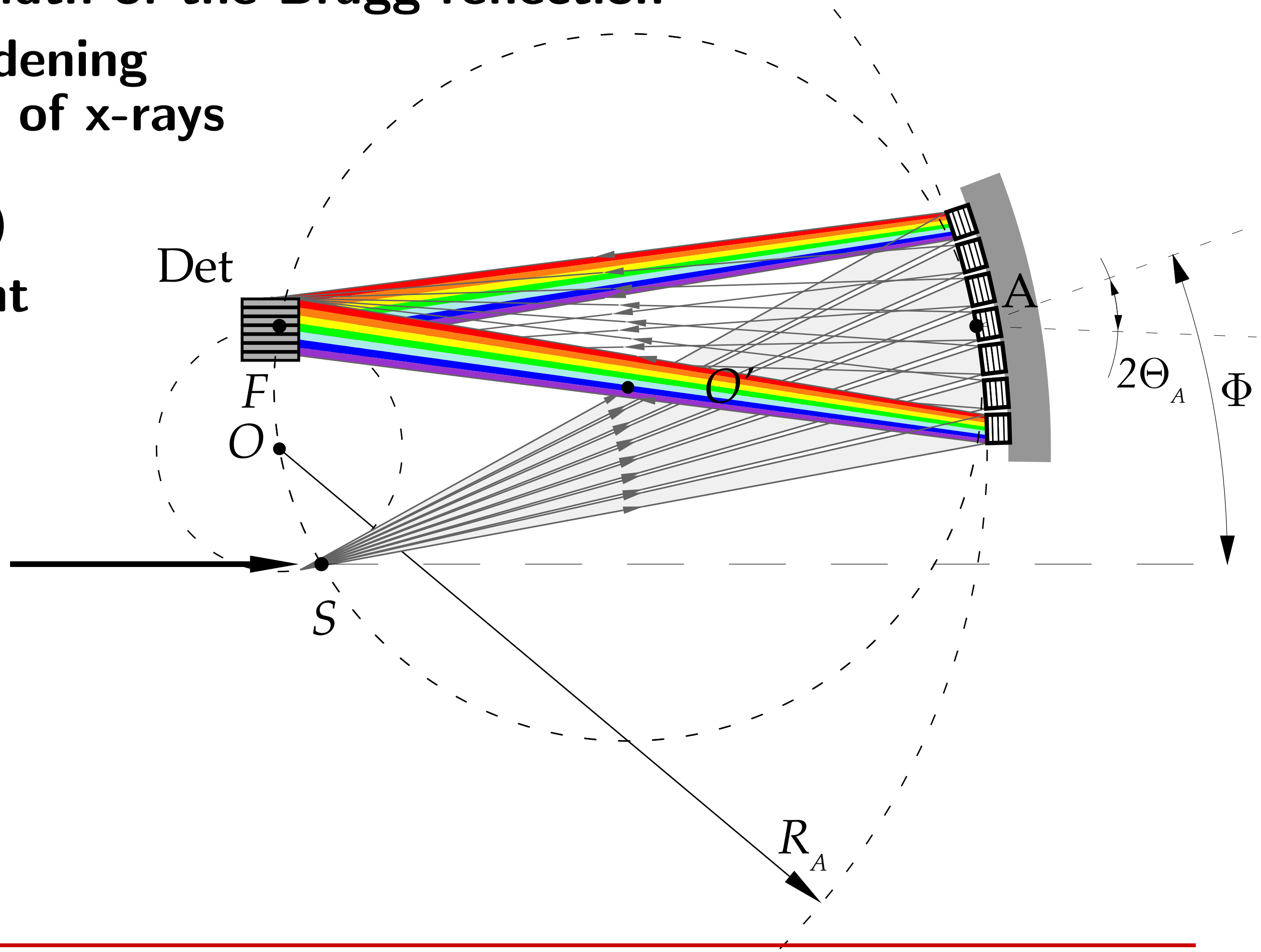
$\Delta E_{\mathrm{i}} =$ **intrinsic (Darwin) width of the Bragg reflection**

$\Delta E_{\mathrm{g}} =$ **"geometrical" broadening due to angular spread of x-rays**

$$\Delta E_{\mathrm{g}} = E \cos\theta_{\mathrm{B}}\, \Delta d/(2R_{\mathrm{A}})$$

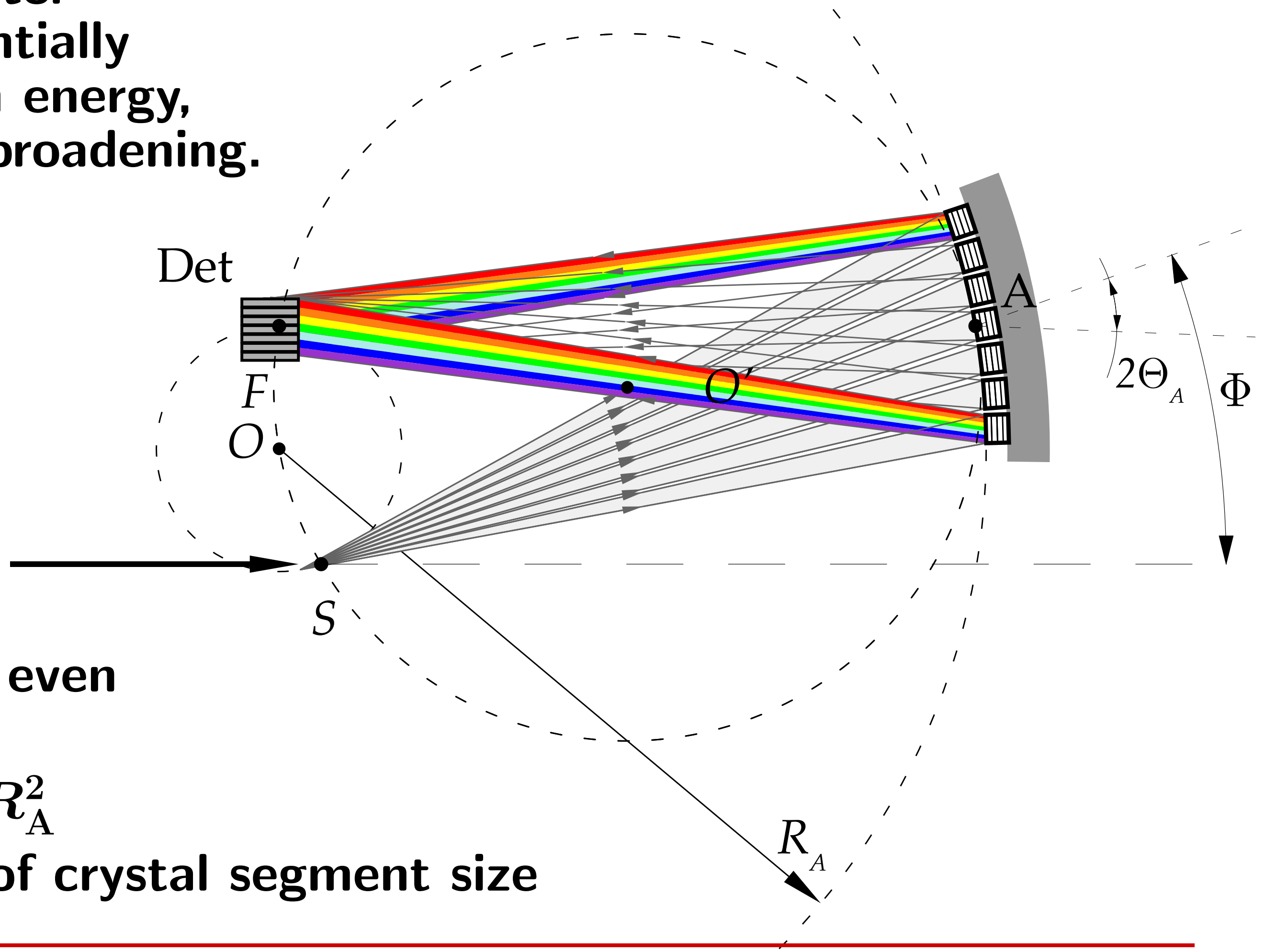
$\Delta d =$ **either crystal segment or detector size**



Next generation x-ray optics and detectors for IXS

S. Huotari, Gy. Vanko, F. Albergamo, C. Ponchut, H. Graafsma,
C. Henriquet, R. Verbeni, and G. Monaco, Synchrotron Rad. (2005) 12, 467 :

Use position sensitive detector to detect IXS signal differentially in space and thus in photon energy, and overcome geometrical broadening.



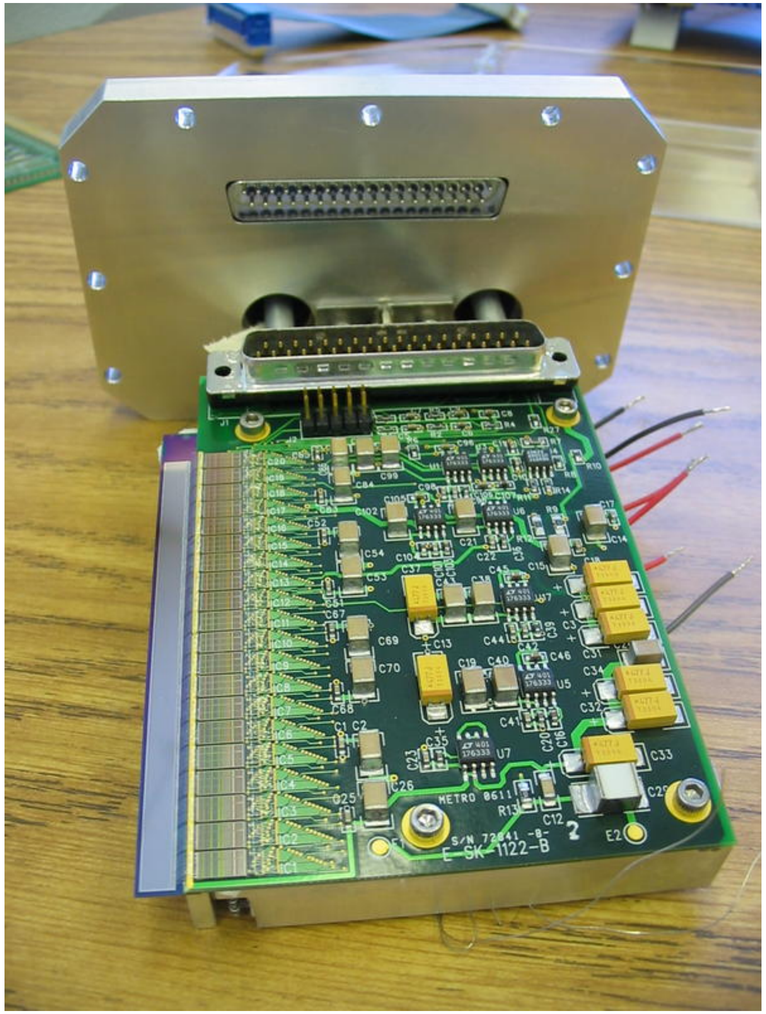
Implications for MERIX:

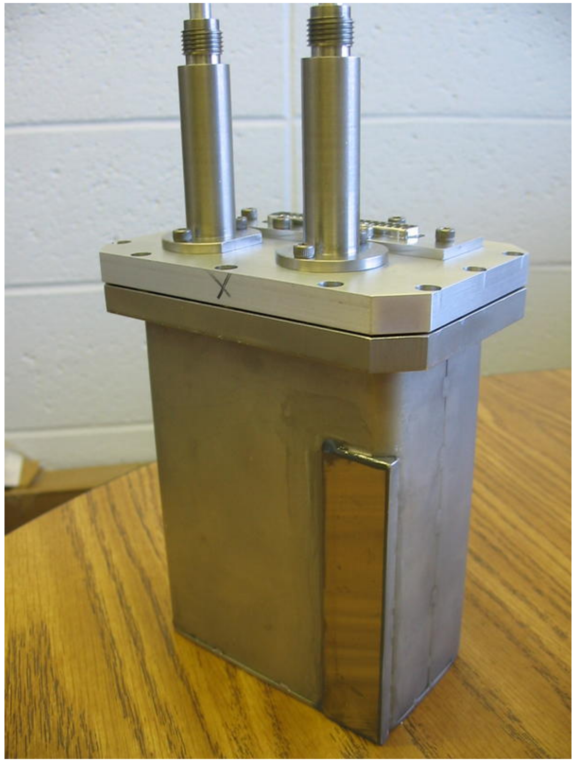
1. Better energy resolution even with smaller R_{A}
2. Higher countrates $\propto 1/R_{\mathrm{A}}^2$
3. Resolution independent of crystal segment size

Photon-Counting Si Microstrip Detector

Built by D. Peter Siddons, Brookhaven

640 strips
1 strip = 125 μm
1 strip = 40 meV
640 strip = 25 eV

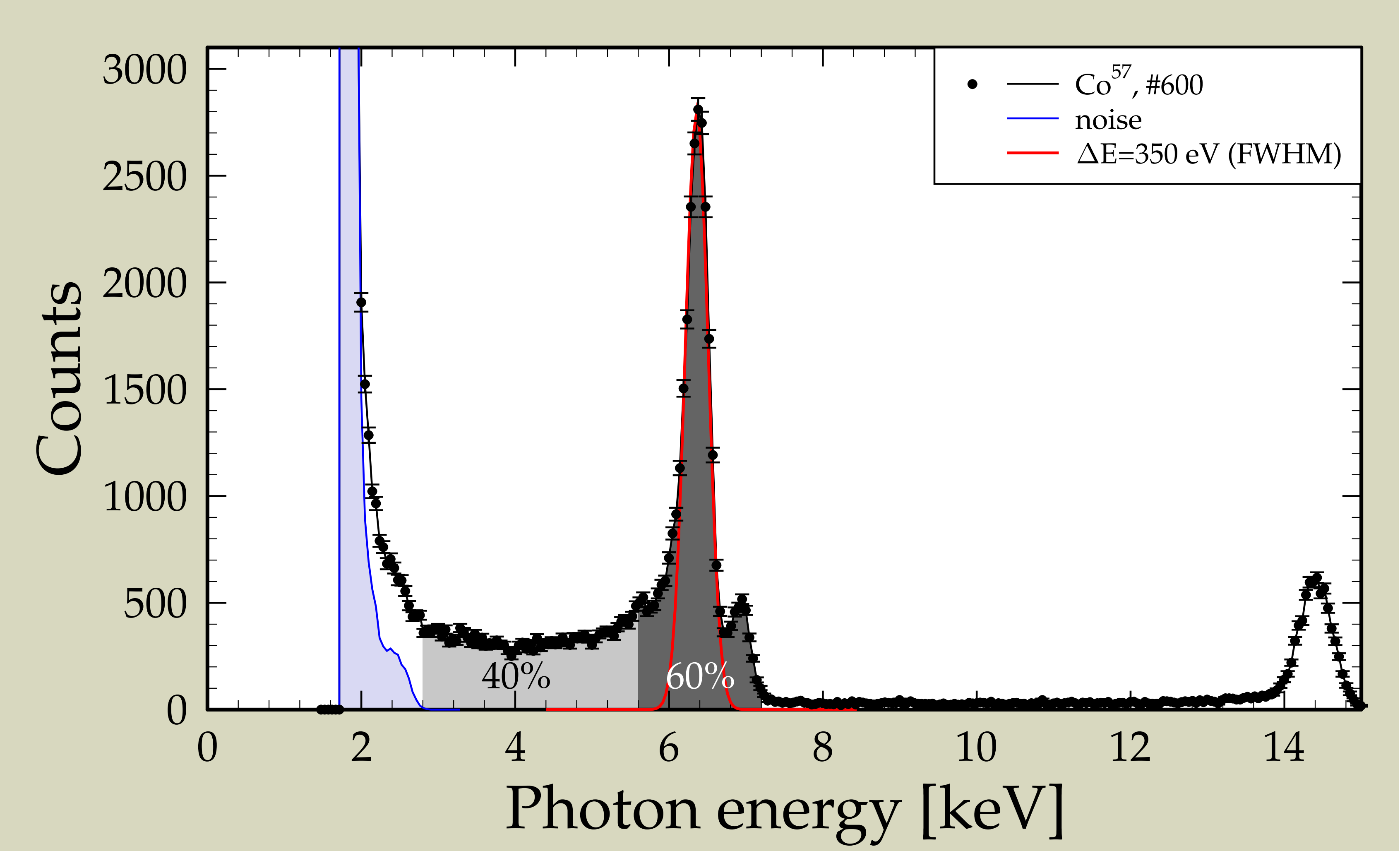




Donated to IXS-CDT by Steve Cramer, UC Davis

Photon-Counting Si Microstrip Detector

Pulse height spectrum taken with ^{57}Co radioactive source

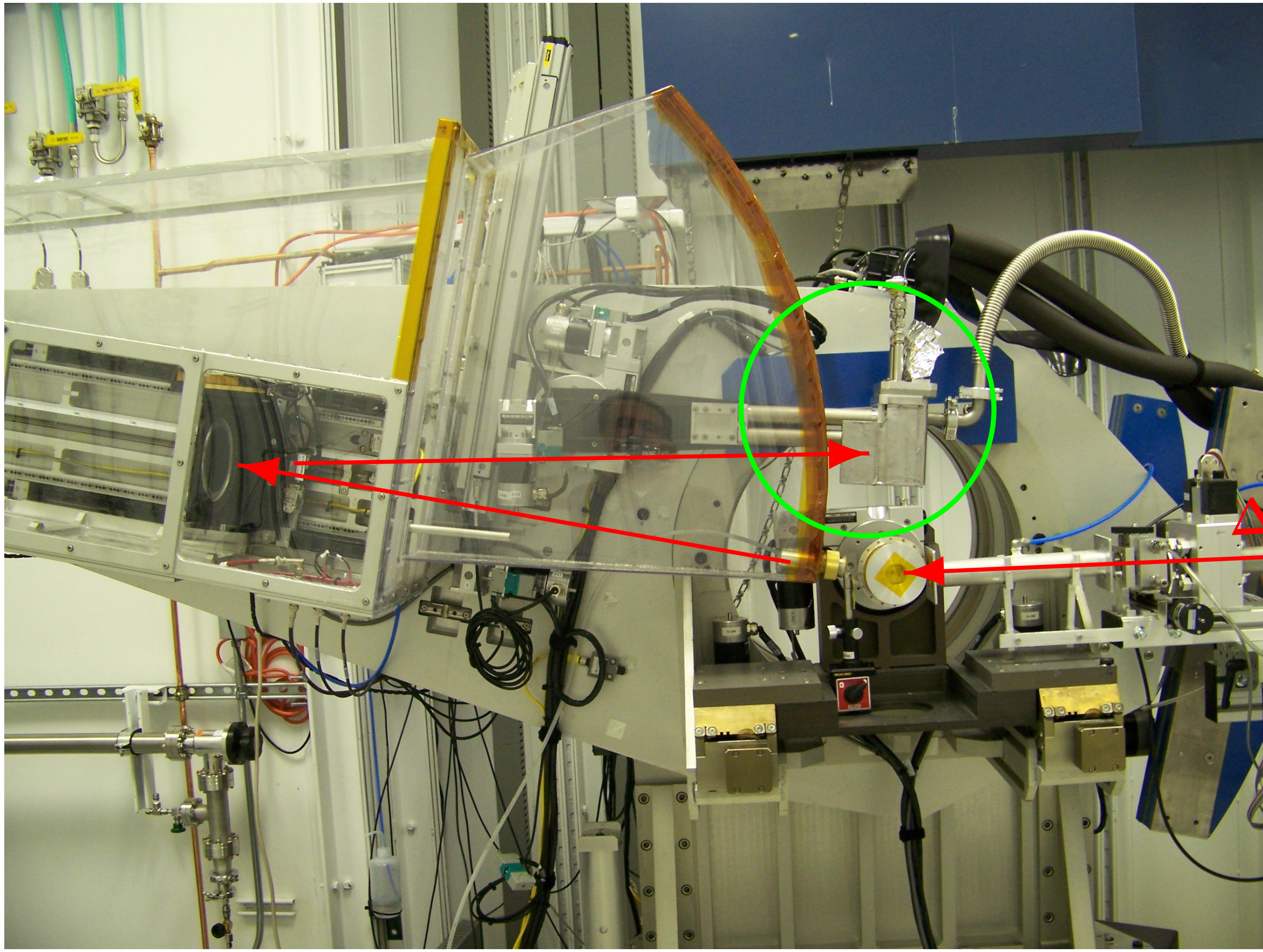


Background: $< 10^{-3}$ Hz/channel.
Effective noise: $\simeq 3$ keV.

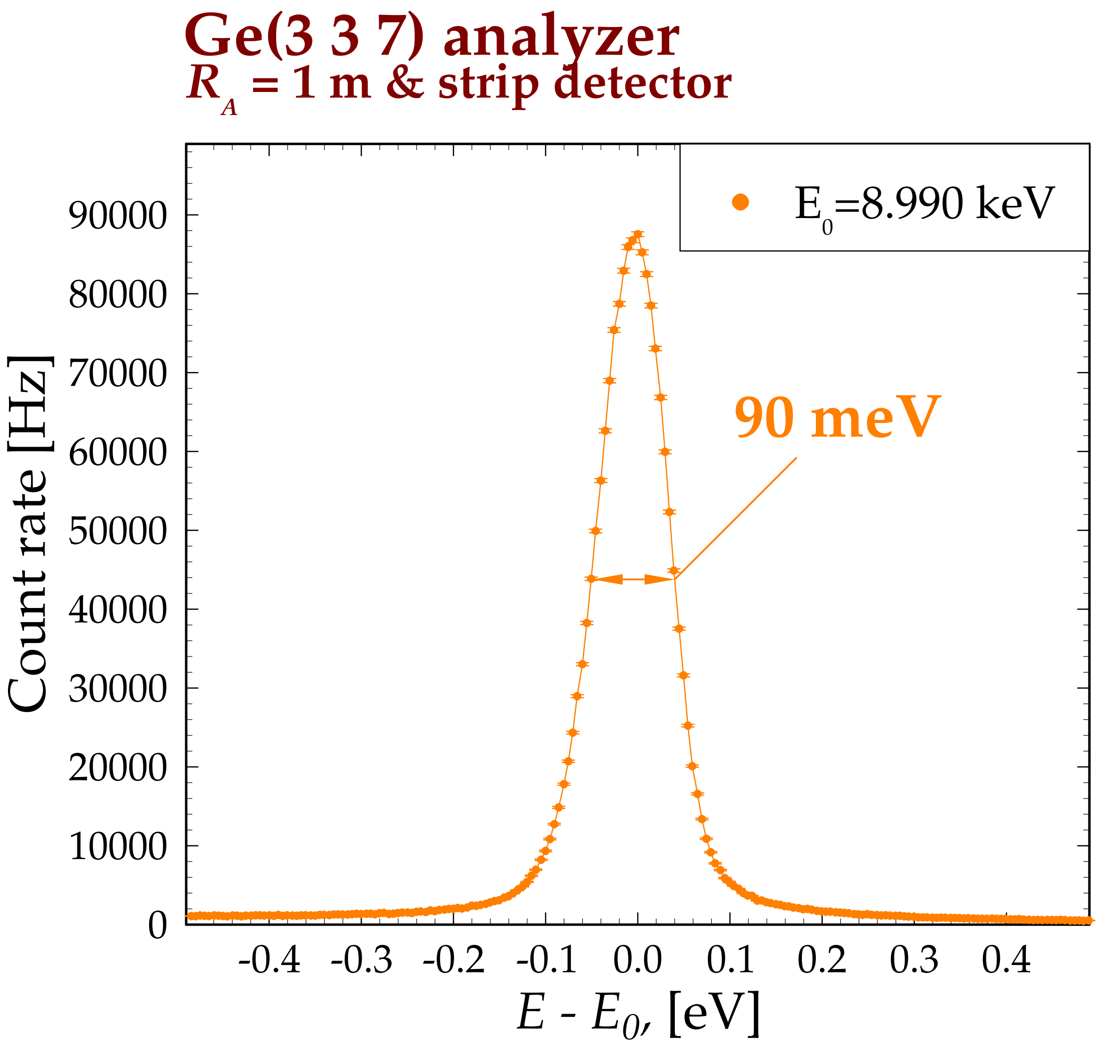
Measurements with $E \geq 4$ keV are possible!

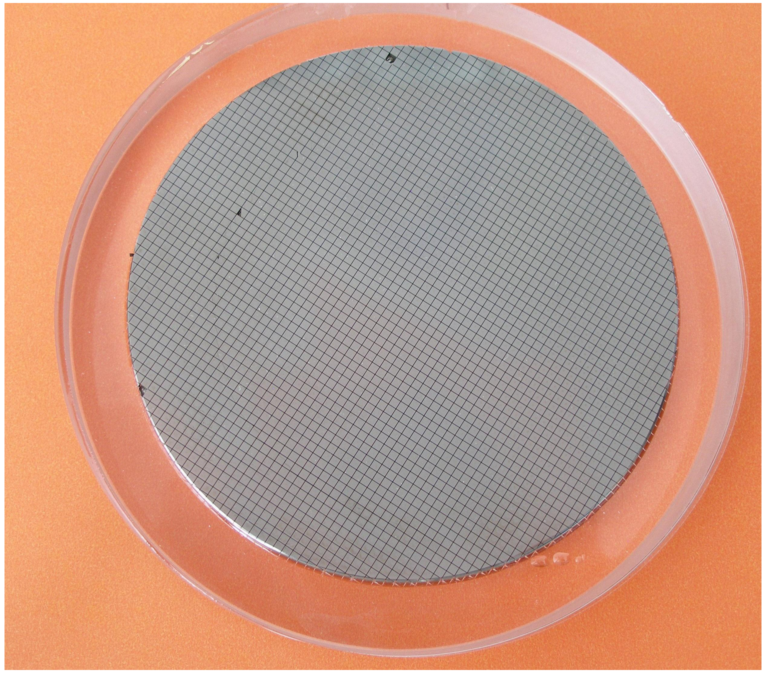
MERIX Spectrometer with Strip Detector

March 2007

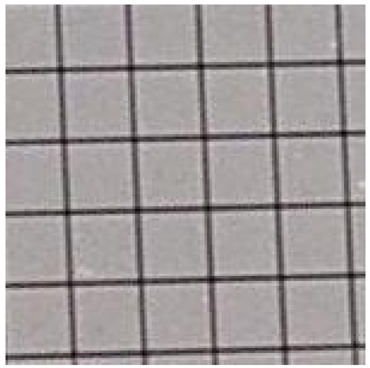


Cu K-edge RIXS Analyzer: Improved Resolution





Crystal segments:
1.5×1.5 mm^2

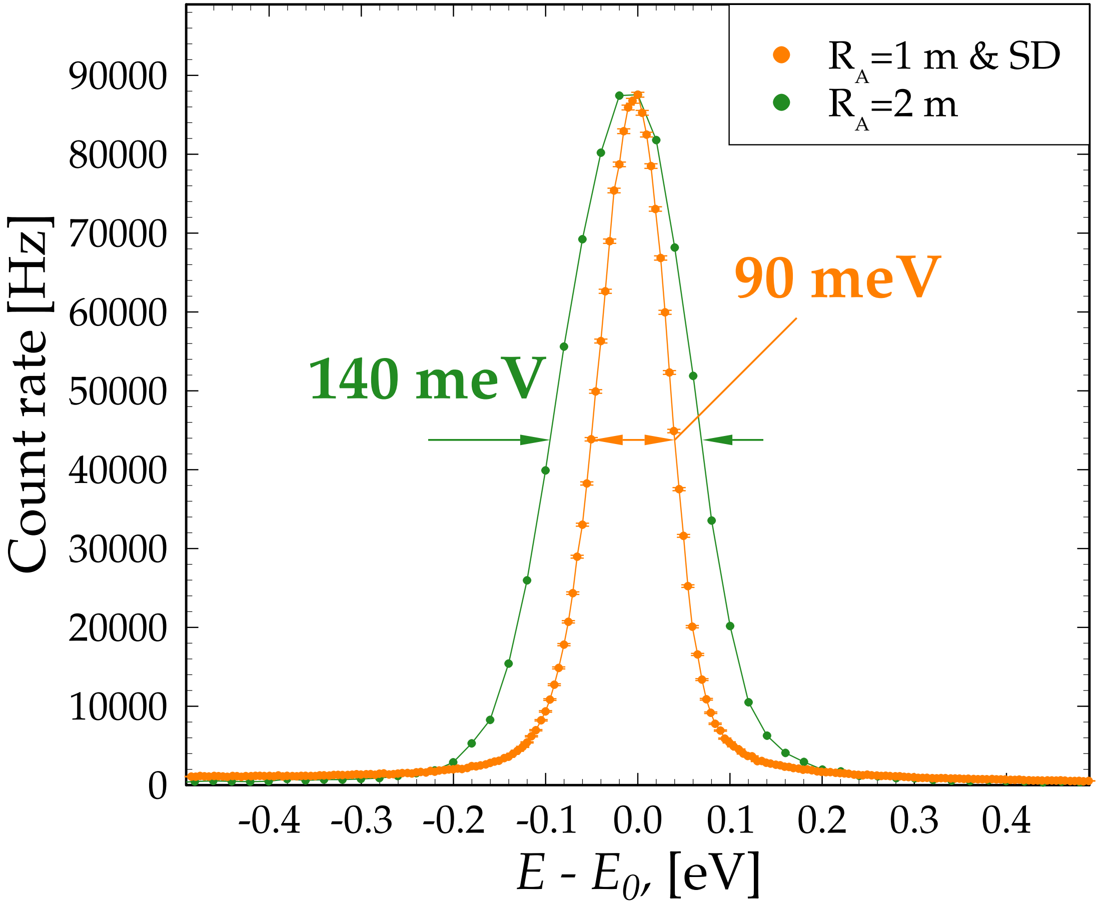


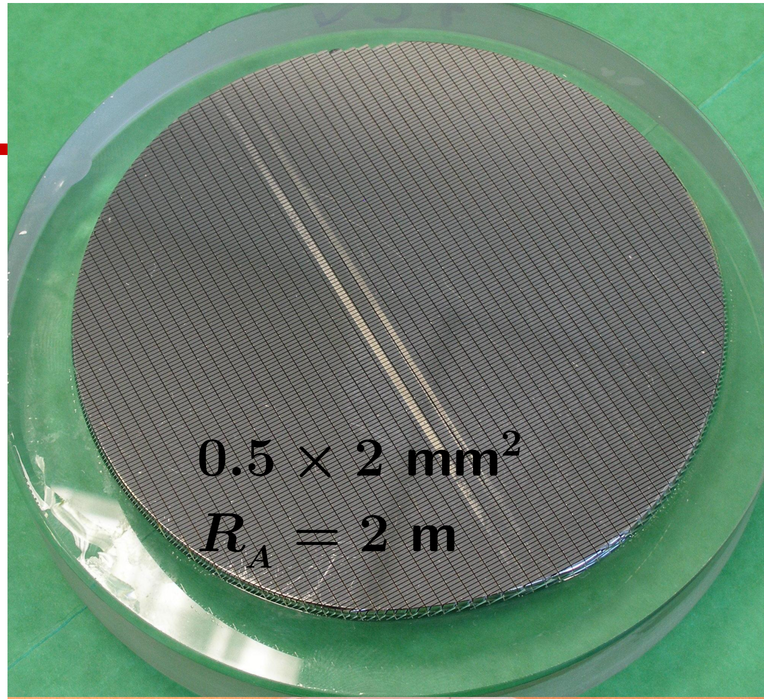
Expected: 90 meV

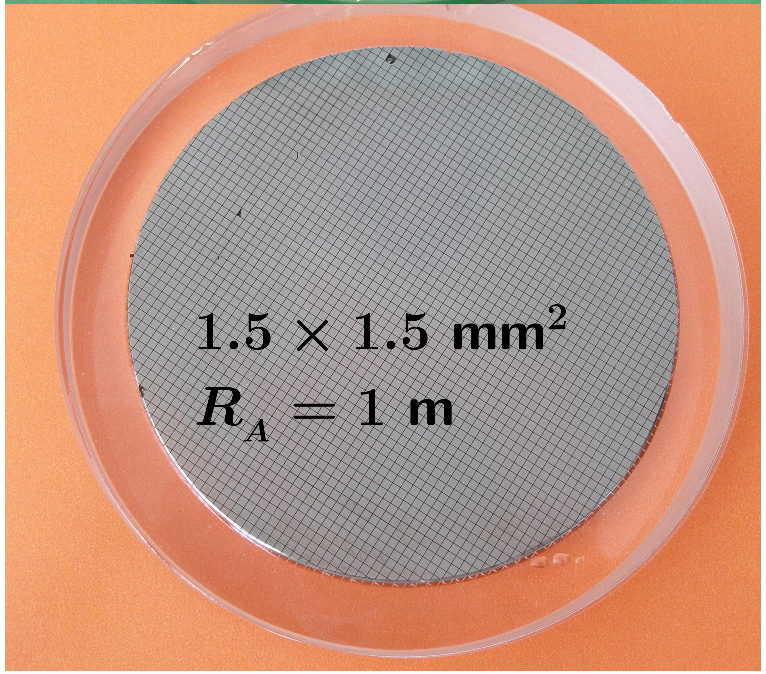
Deconvoluted width : 52 meV

Cu K-edge RIXS Analyzers

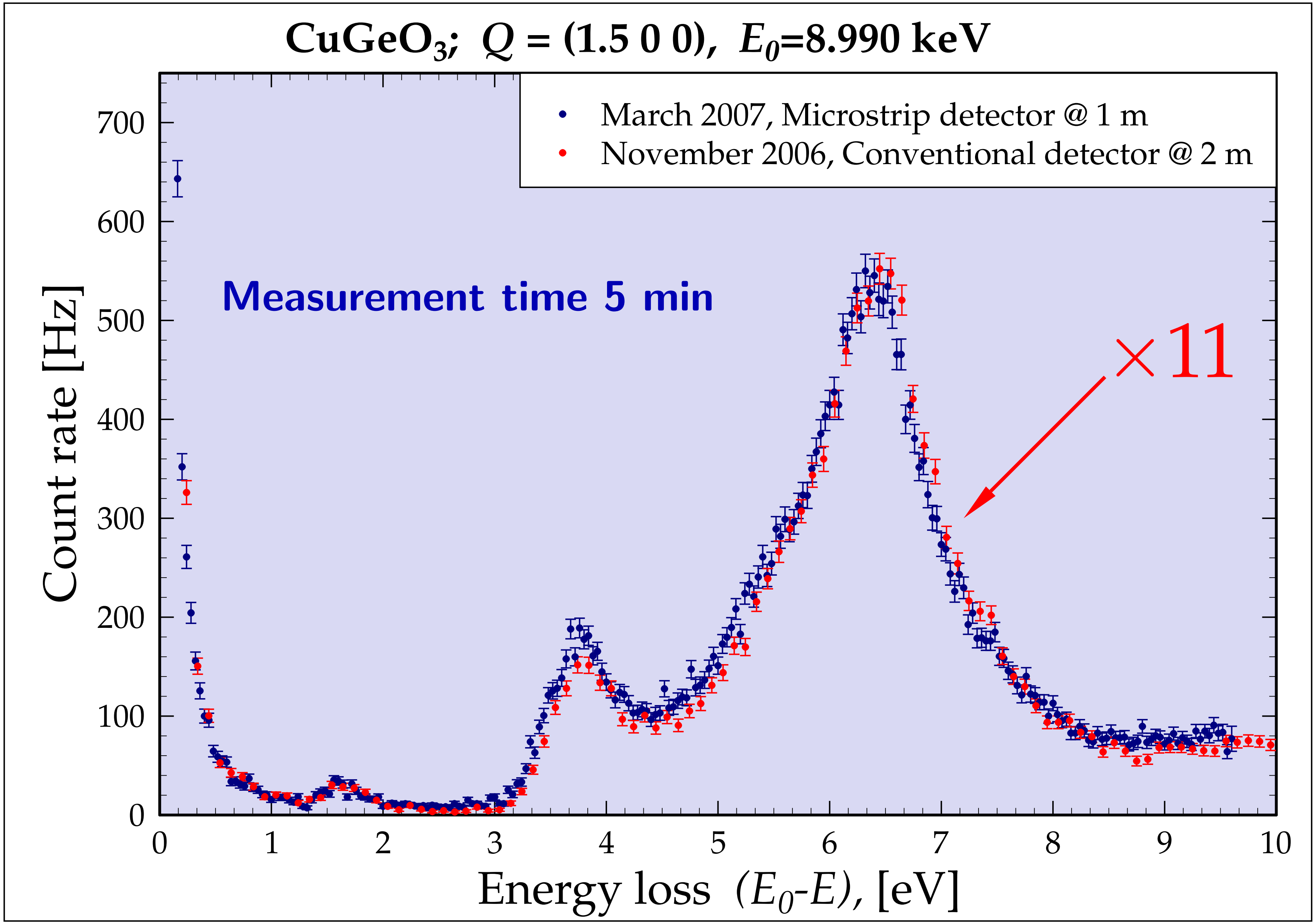
Ge(3 3 7) analyzer





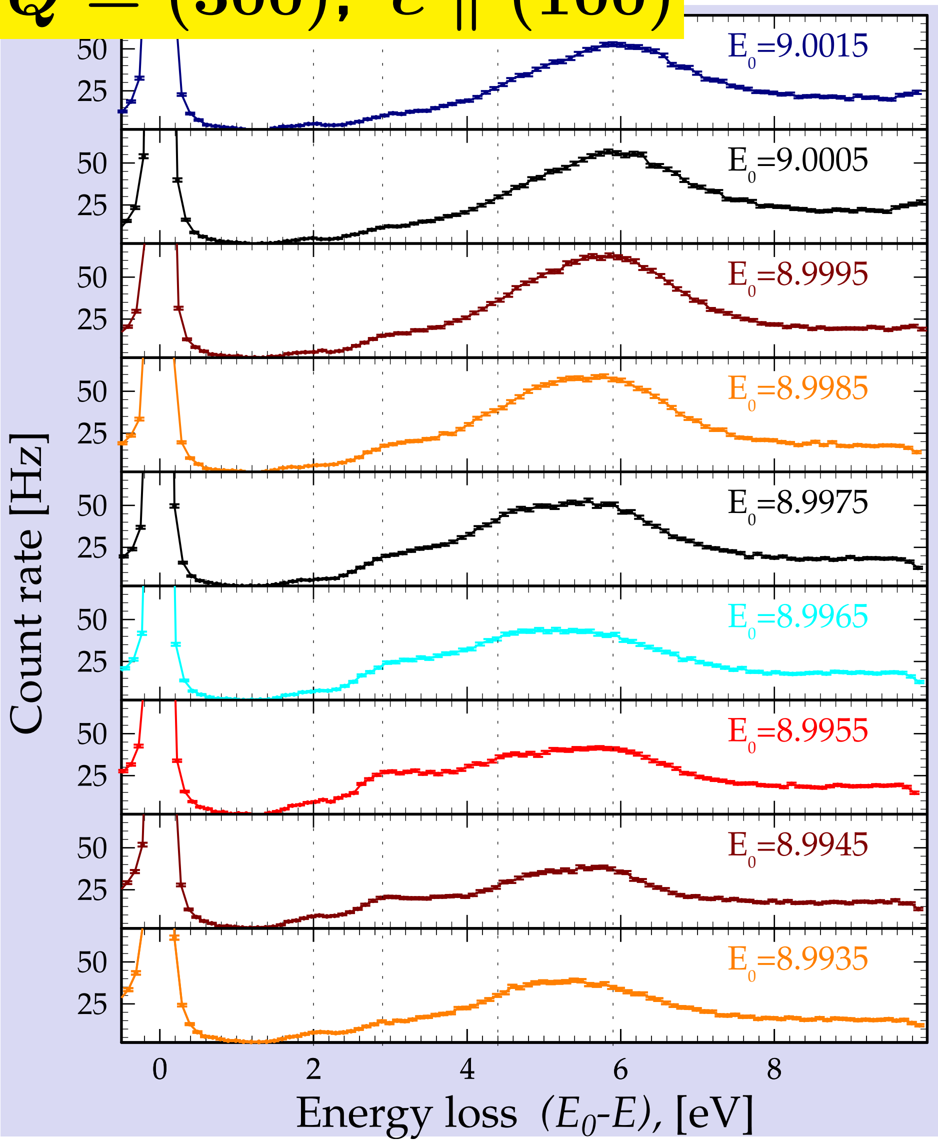


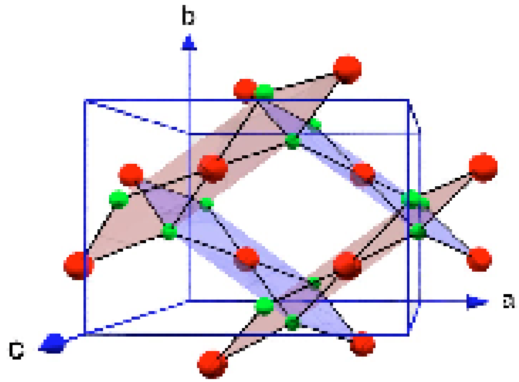
Dramatic Increase in Count Rate

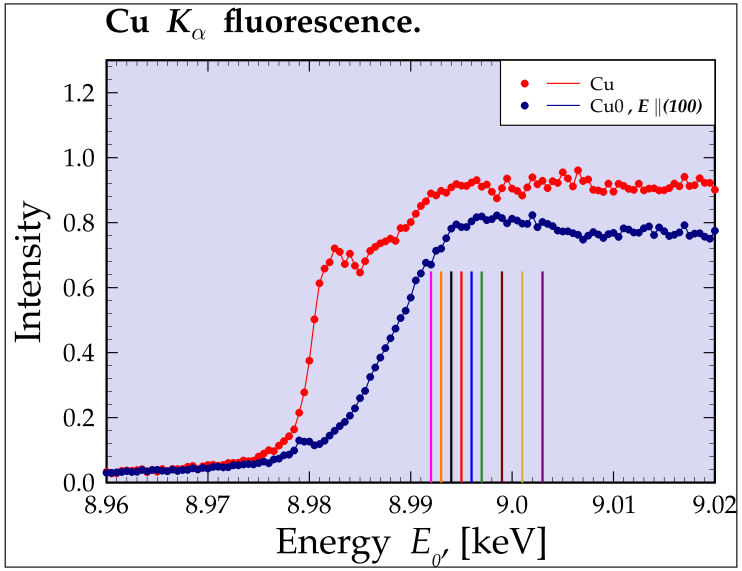


Cu K-edge RIXS in CuO

$$\vec{Q} = (300),\ \vec{e} \parallel (100)$$







Yu. Shvyd'ko, H. Yavas, E. Alp, D. Casa, A. Said (2007)

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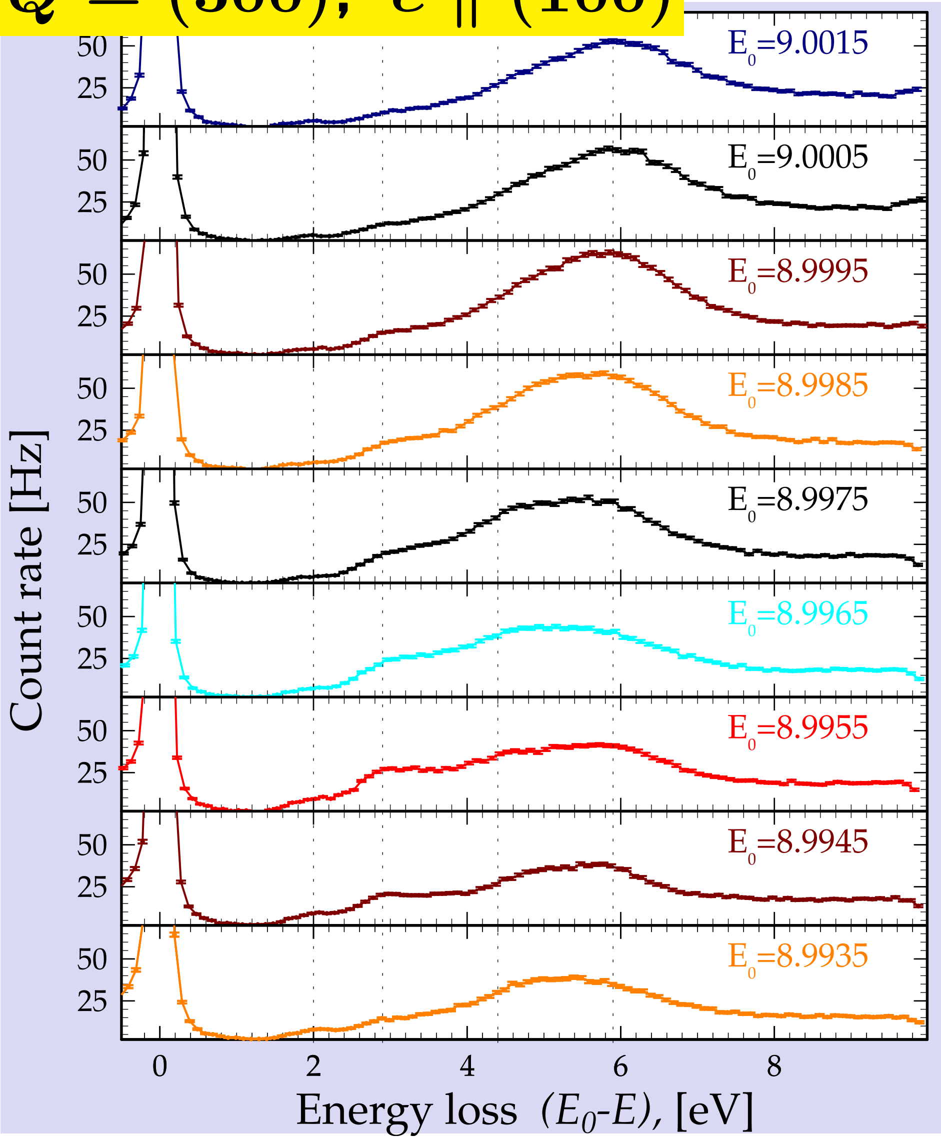
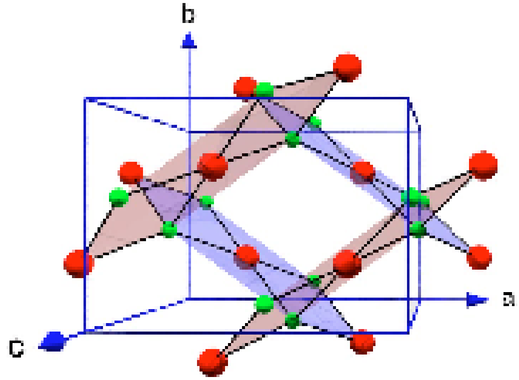
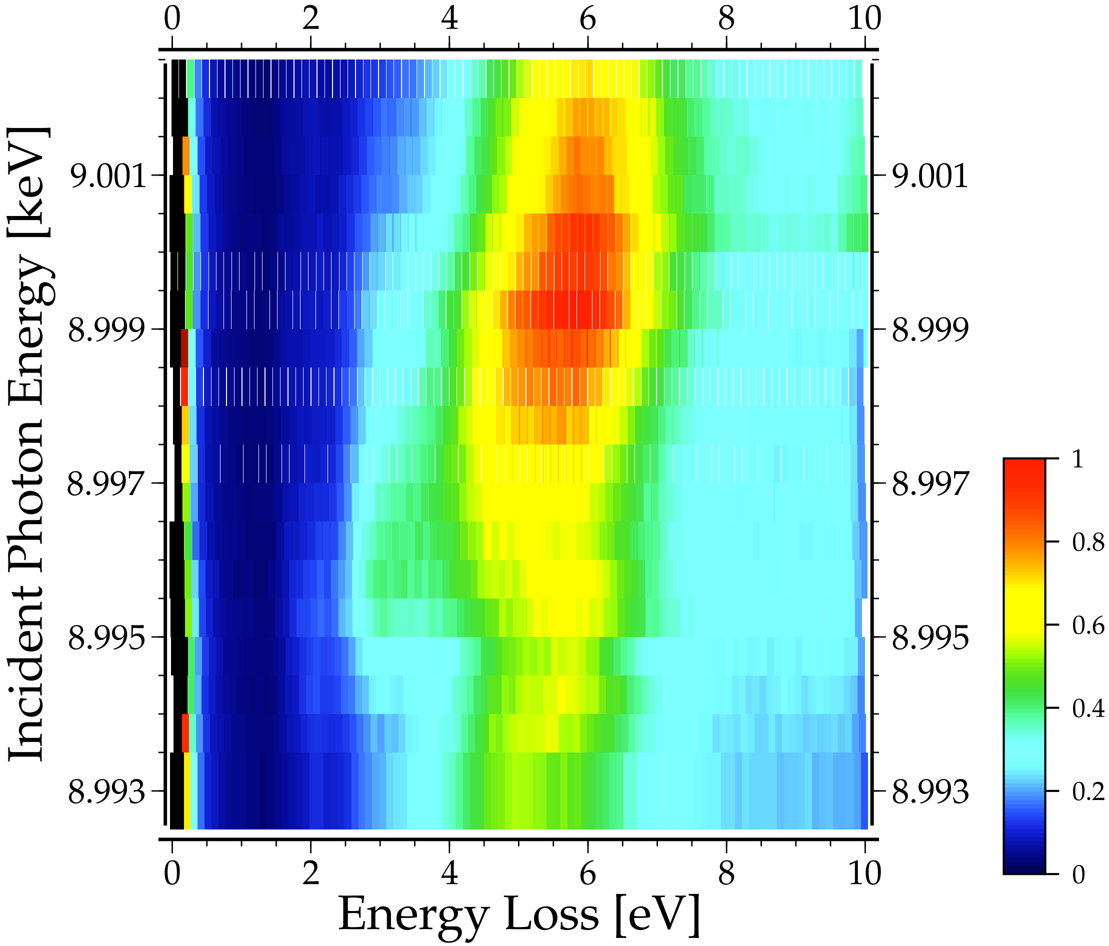






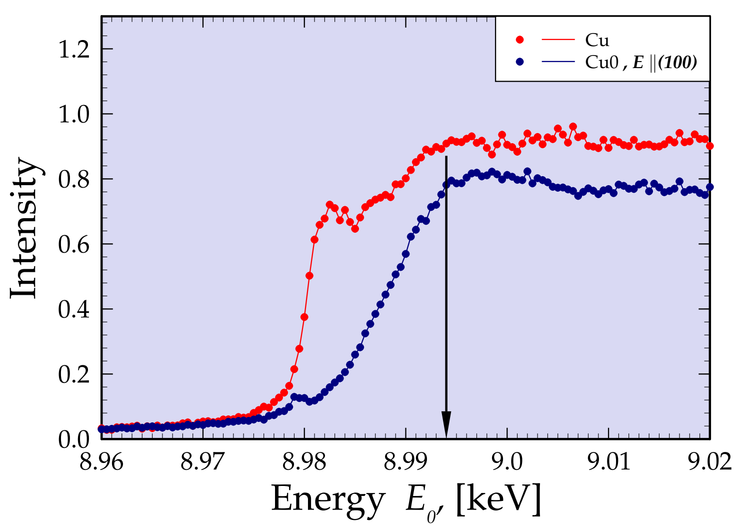
Yu. Shvyd'ko, H. Yavas, E. Alp, D. Casa, A. Said (2007)

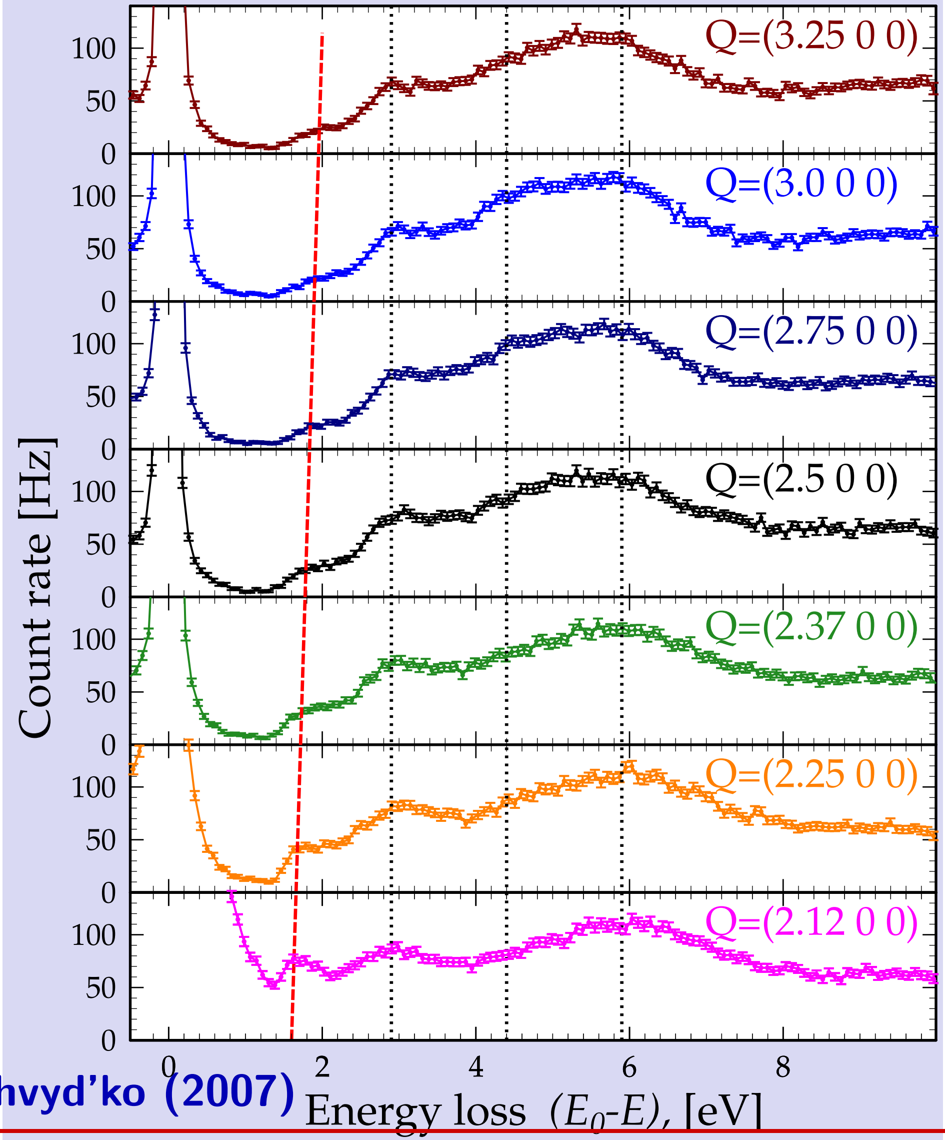
Cu K-edge RIXS in CuO: $\vec{Q}$-Dependence

$E = 8.994$ keV, $\vec{e} \parallel (100)$

Measurement time 25 min/spectrum

Cu K_α fluorescence.





H. Yavas, E. Alp, D. Casa, A. Said, Yu. Shvyd'ko (2007)

Collective Charge Modes in "Spin-Charge separation" cuprates $SrCuO_2$ measured at MERIX (2007)

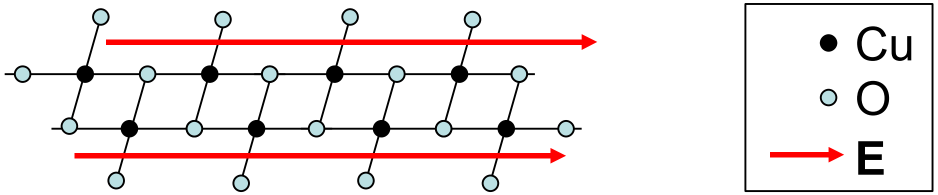
Resonant inelastic x-ray scattering signal at the copper k-edge has been measured for quasi-one dimensional (1D) ***spin-charge separation materials class $SrCuO_2$*** at MERIX and SPring-8 BL11XU.

Enhanced resolution and count rate at MERIX (1) revealed new features not seen before and (2) clearly resolved multiple features in the spectrum not possible in any previous study of such cuprates.

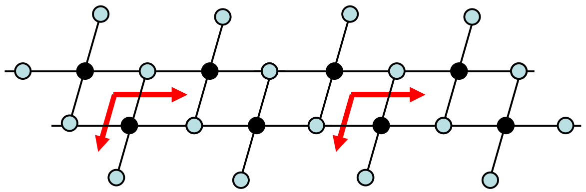
A small angle geometry could be chosen at MERIX to allow pure incident polarization along the 1D chain direction, while the available moderate resolution (~400meV) at SPring-8 necessitated a 90^{o} scattering geometry with mixed polarization in the Cu-O plaquette to obtain signals.

Polarization conditions:

MERIX: **E** parallel to chain, in plaquette



SPring-8: **E** 45^{o} to a- and b-axes, in plaquette



Wray et.al. (2007)

Resonance Profiles measured on $SrCuO_2$ at SPring-8 and MERIX

SPring-8 BL11XU

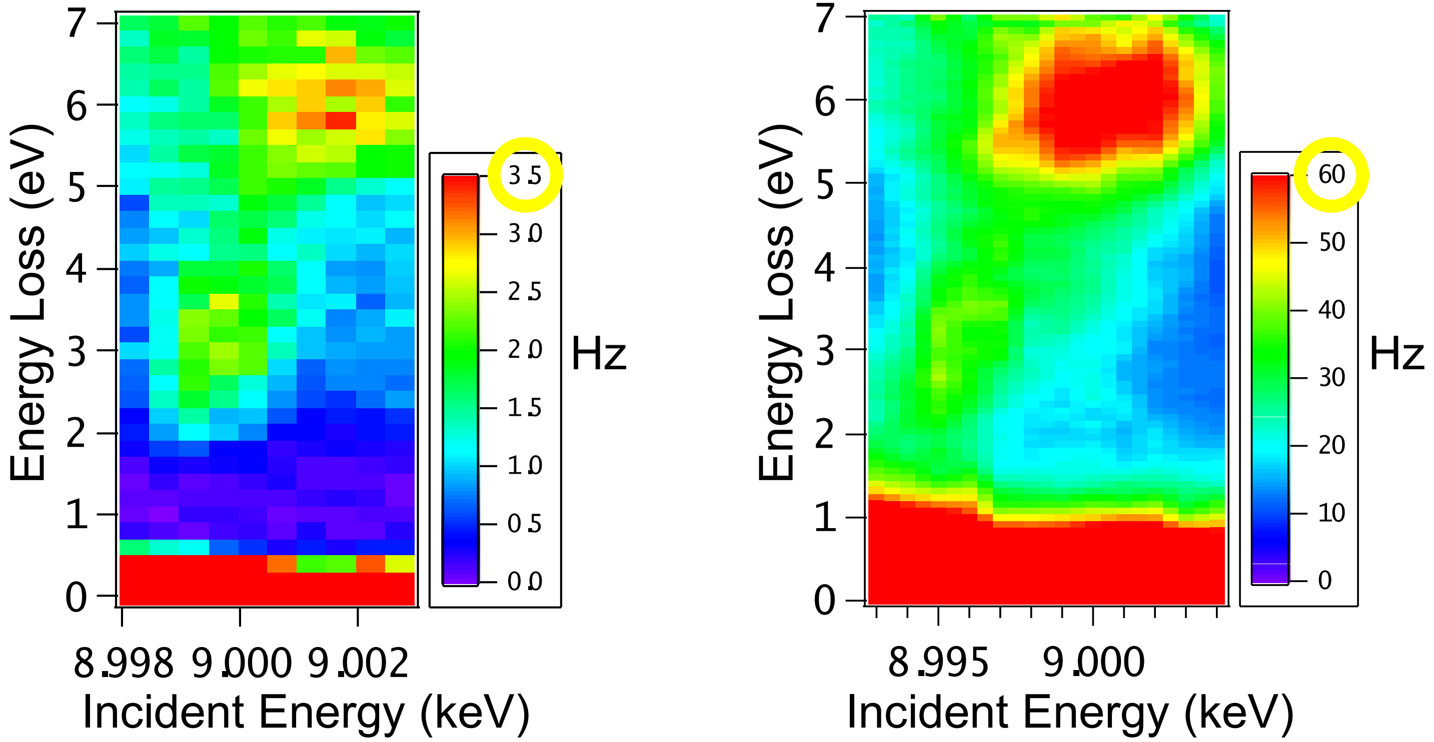
45 seconds/point

(90° geometry)

MERIX

5 seconds/point

(small angle geometry)



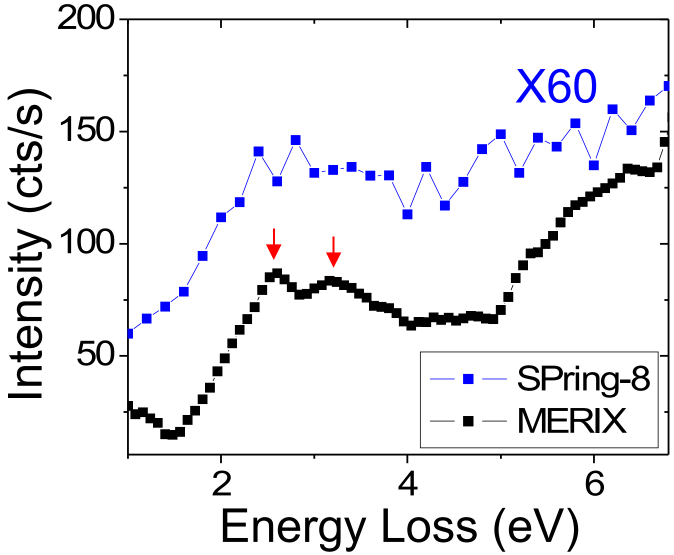
Wray et.al. (2007)

Intensity shown in counts/s

Excitonic Modes in a 1D Cuprate Chain

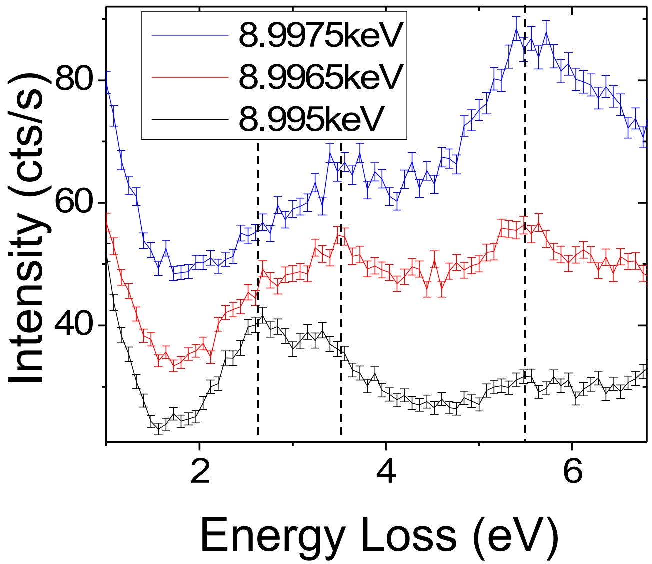
Clearly resolve features for the first time

Comparison of MERIX and SPring-8 data with energy and polarization tuned for optimal enhancement of the 2-4eV signal:



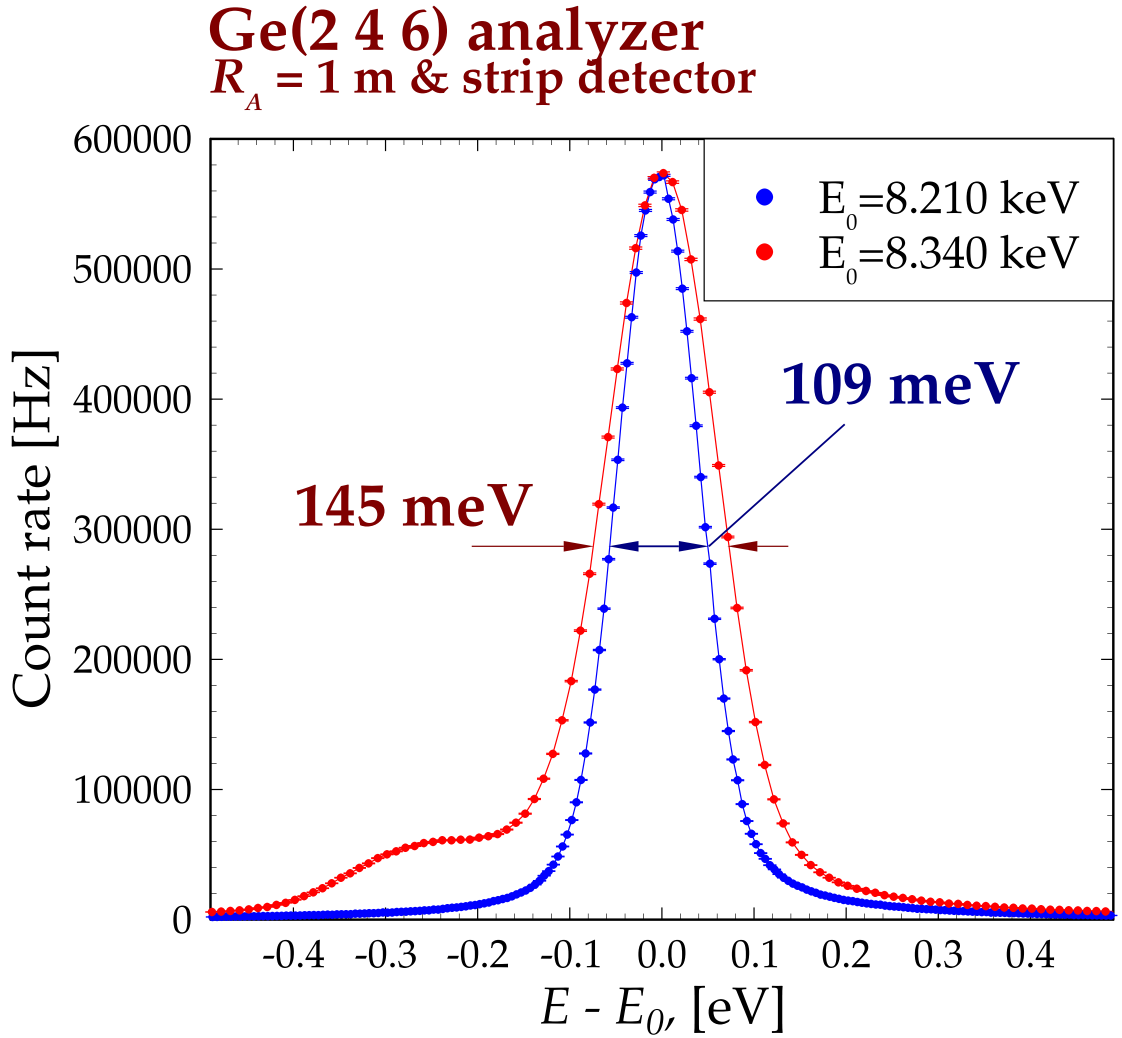
Showed new features As E_{in} is varied

Using different incident energies, in conjunction with high count rate and resolution, allows identification of diverse features at MERIX:



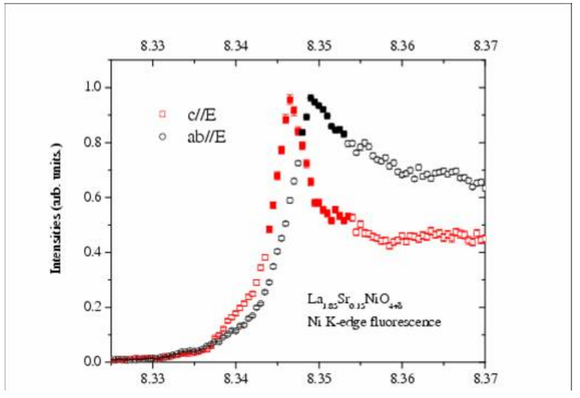
Wray et.al. (2007)

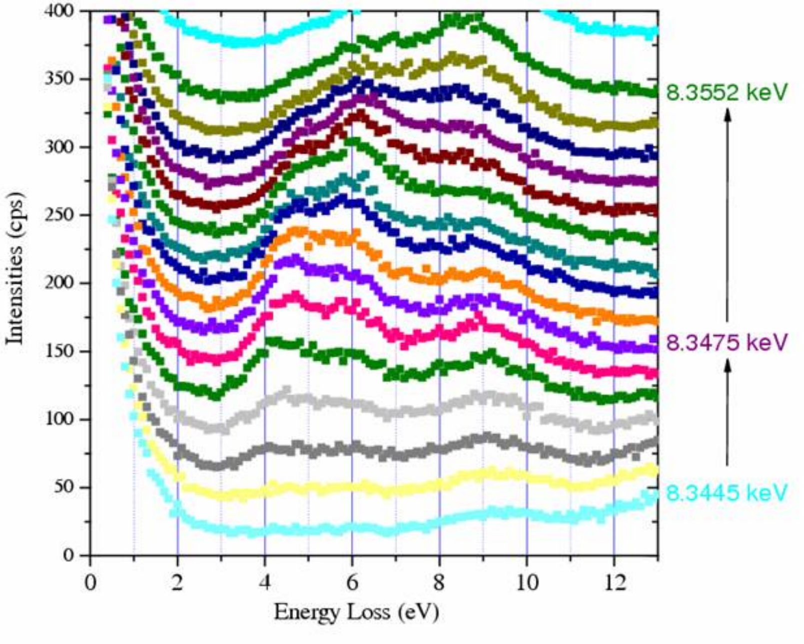
Ni K-edge RIXS Analyzer

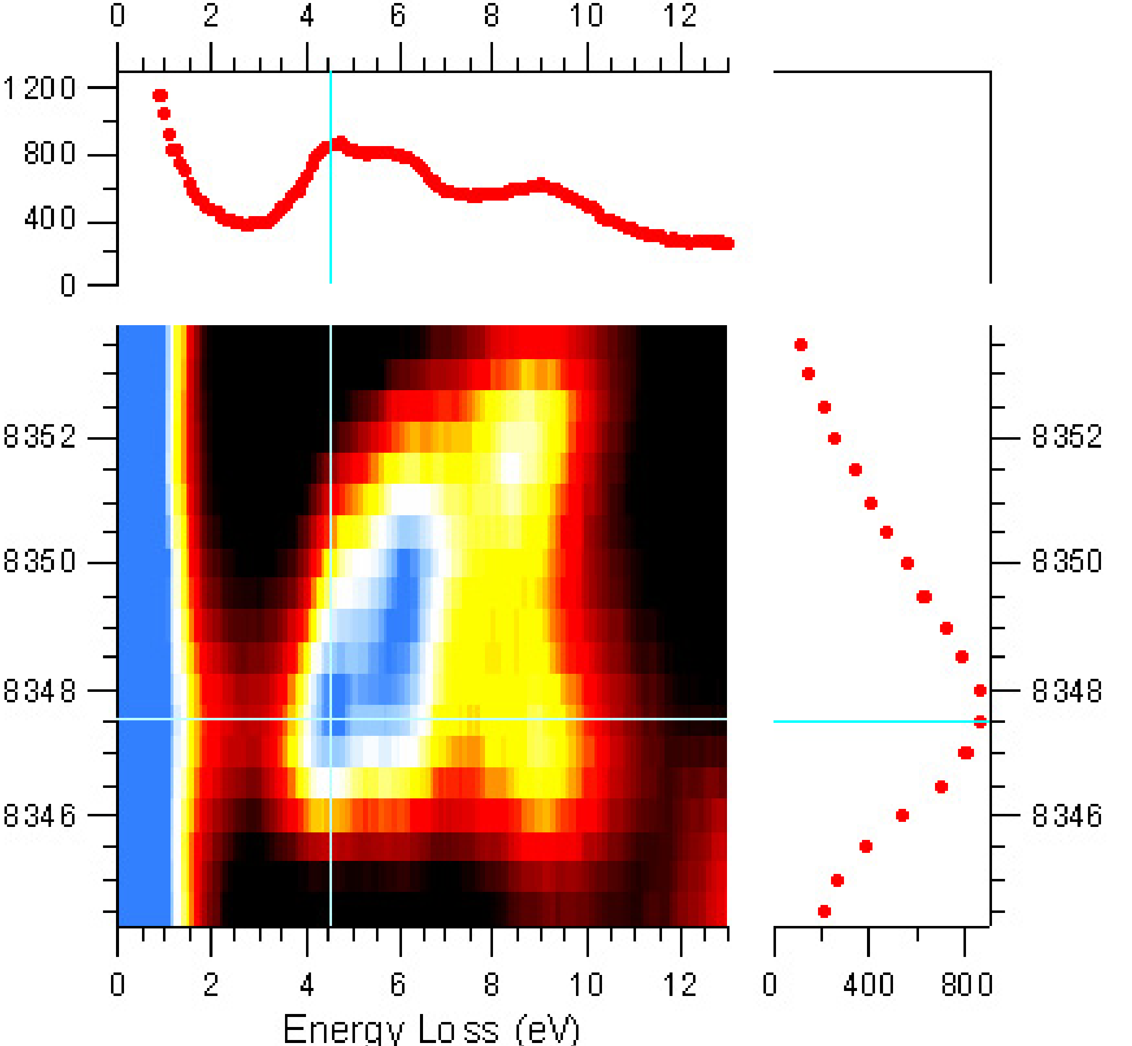


Expected: 123 meV @ 8.340 keV and 105 meV @ 8.210 keV, respectively

Ni K-edge RIXS in $La_{1.85}Sr_{0.15}NiO_{4+\delta}$: $\vec{e} \parallel \vec{c}$

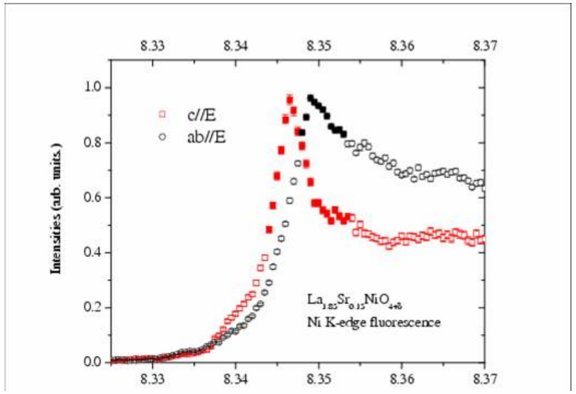


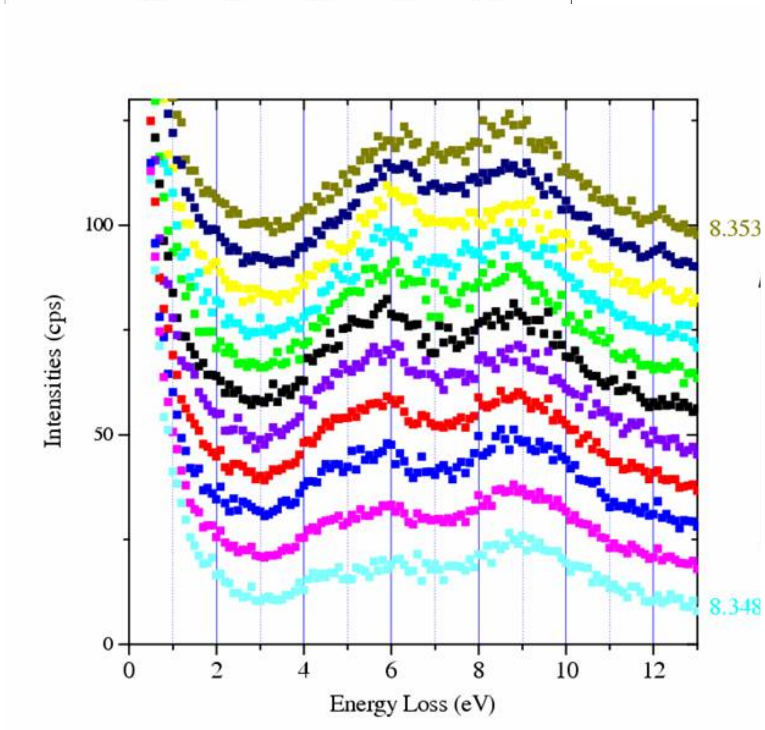


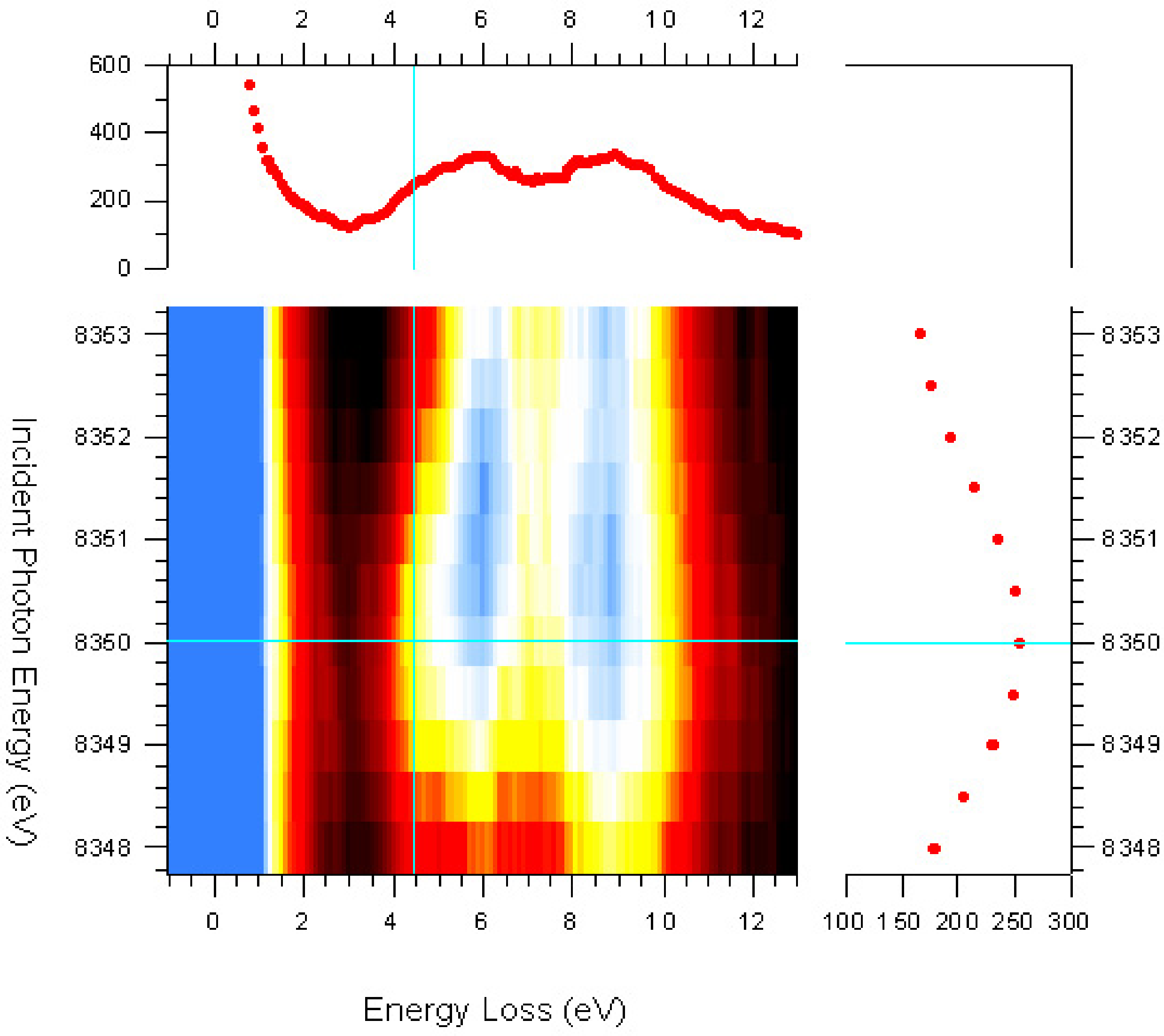


J.H. Kim, D. Ellis, Y.-J. Kim, E. Alp, A. Said, Yu. Shvyd'ko (2007)

Ni K-edge RIXS in $La_{1.85}Sr_{0.15}NiO_{4+\delta}$: $\vec{e} \parallel (\vec{a}, \vec{b})$



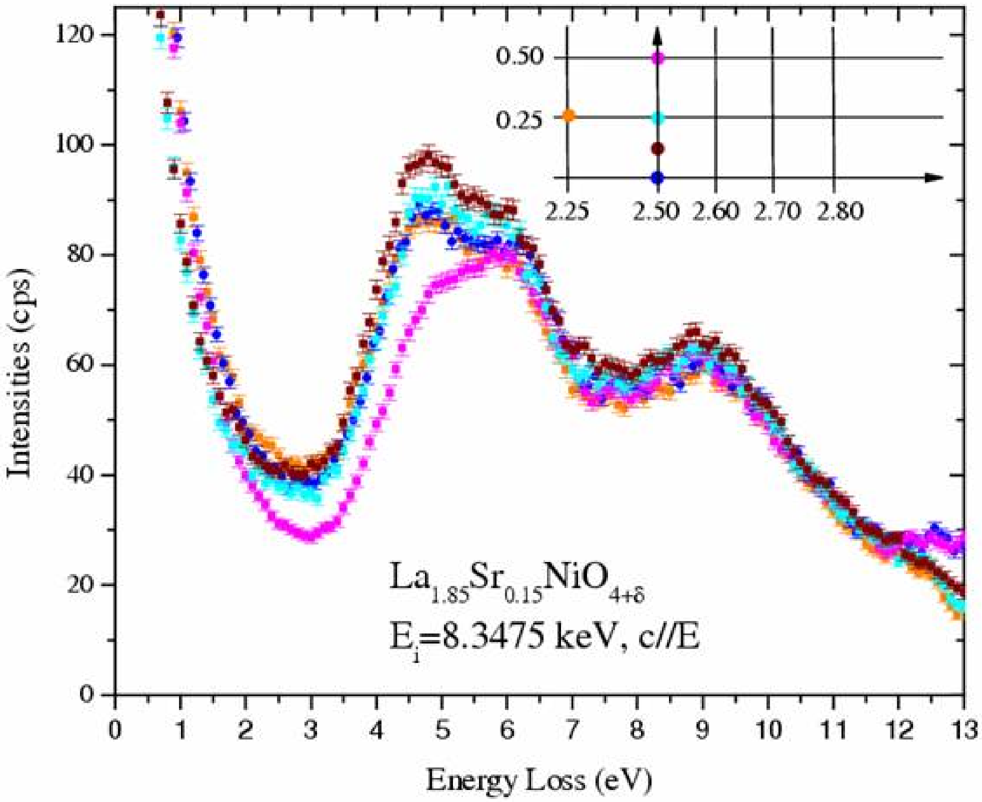


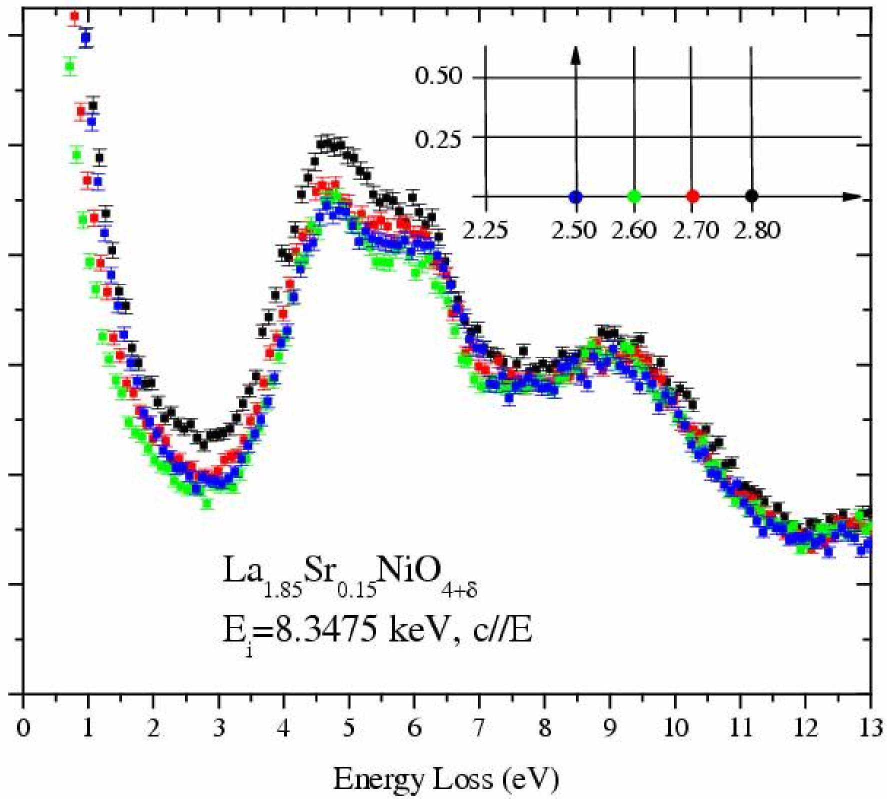


J.H. Kim, D. Ellis, Y.-J. Kim, E. Alp, A. Said, Yu. Shvyd'ko (2007)

Ni K-edge RIXS in $La_{1.85}Sr_{0.15}NiO_{4+\delta}$: $\vec{Q}$-Dependence

$$\vec{Q} = (h, k, 0)$$

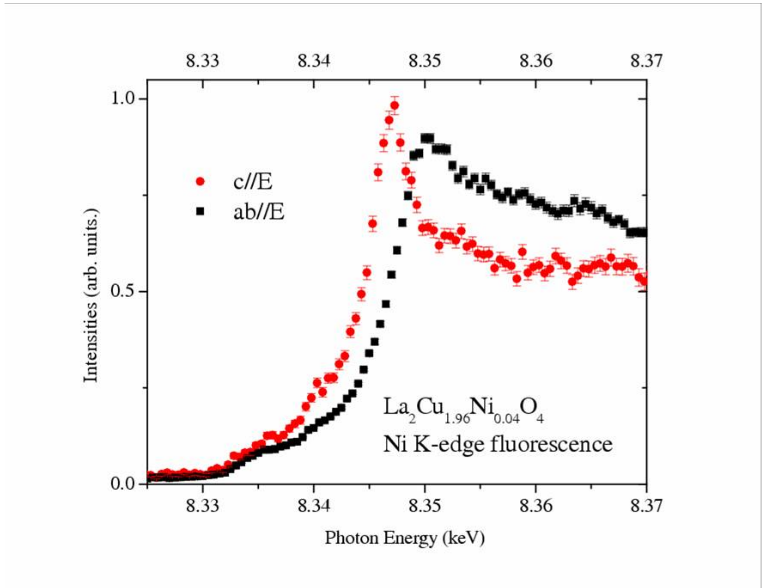


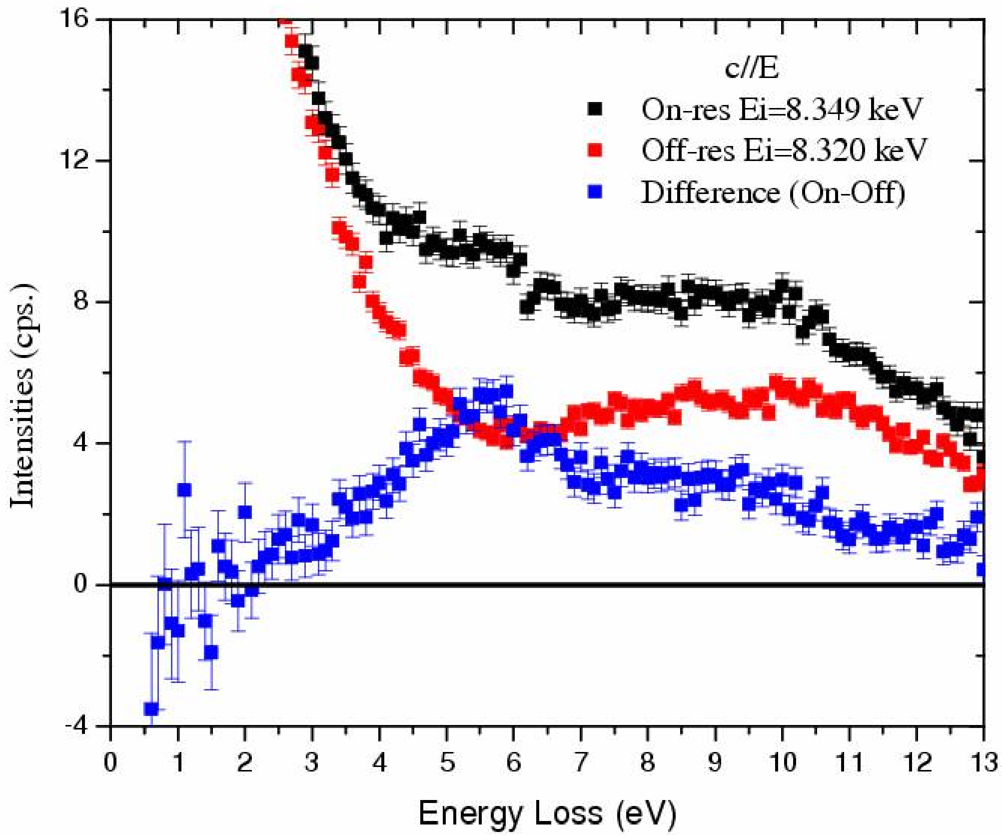


J.H. Kim, D. Ellis, Y.-J. Kim, E. Alp, A. Said, Yu. Shvyd'ko (2007)

K-edge RIXS in Strongly Diluted Samples

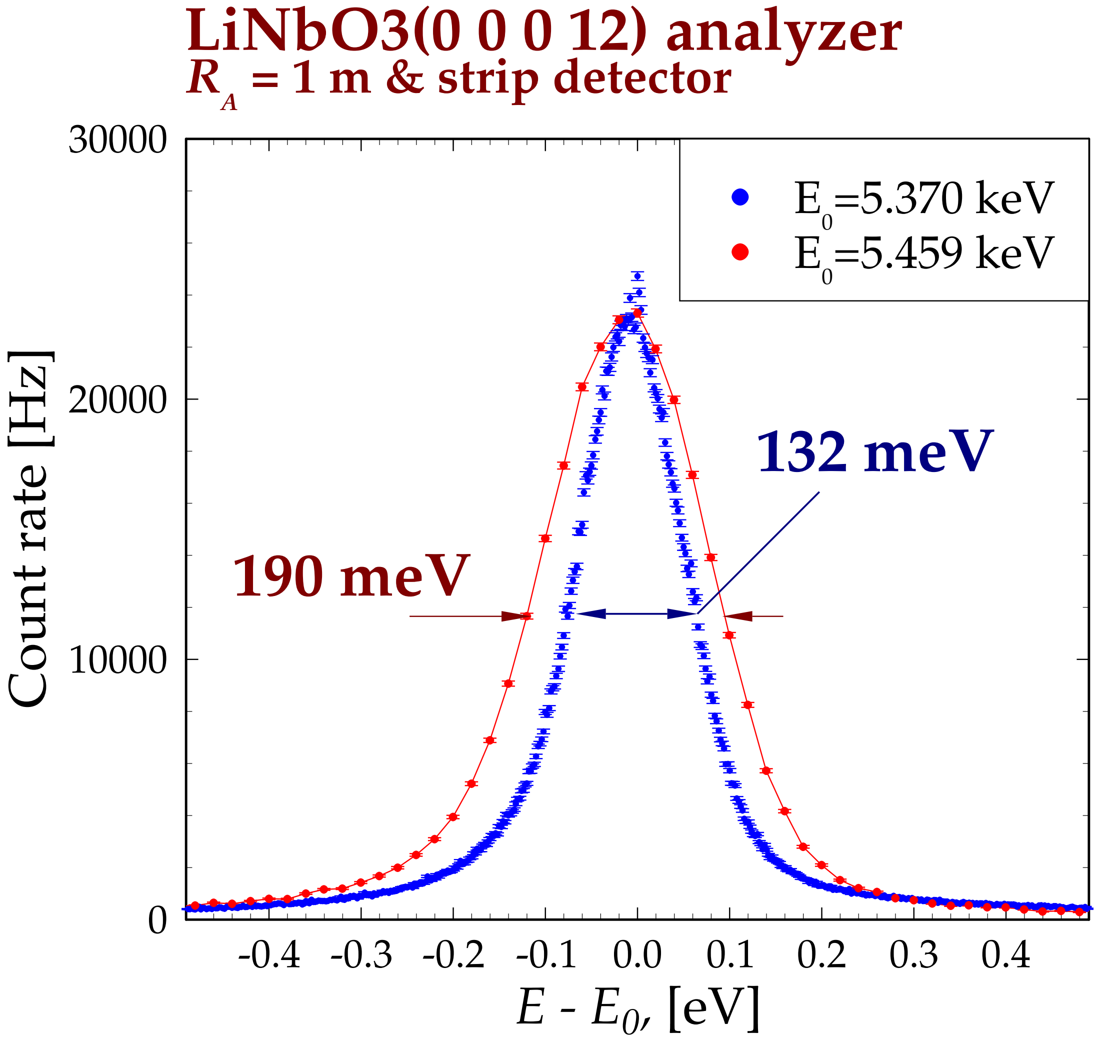
Ni K-edge RIXS in $La_2Cu_{0.96}Ni_{0.04}O_4$

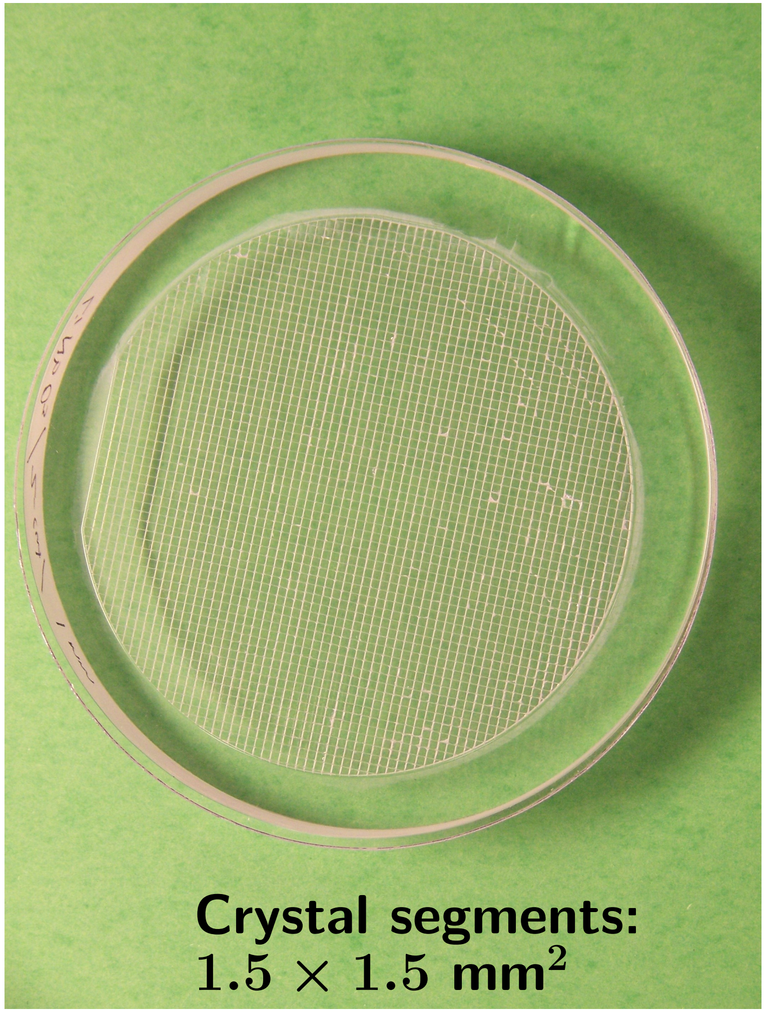




J.H. Kim, D. Ellis, Y.-J. Kim, E. Alp, A. Said, Yu. Shvyd'ko (2007)

V K-edge RIXS Analyzer





Expected: 145 meV @ 5.459 keV and 130 meV @ 5.370 keV, respectively

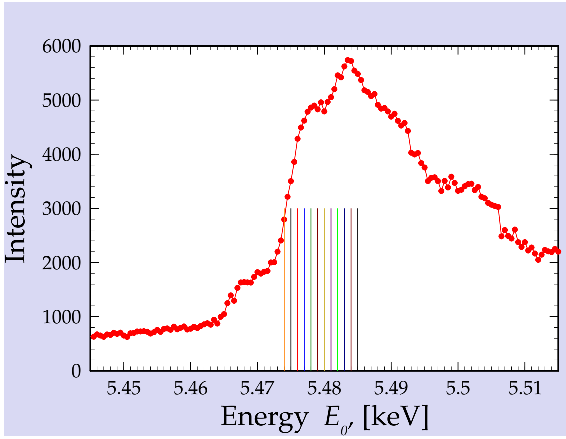
V K-edge RIXS in V_2O_3: E_0-dependence

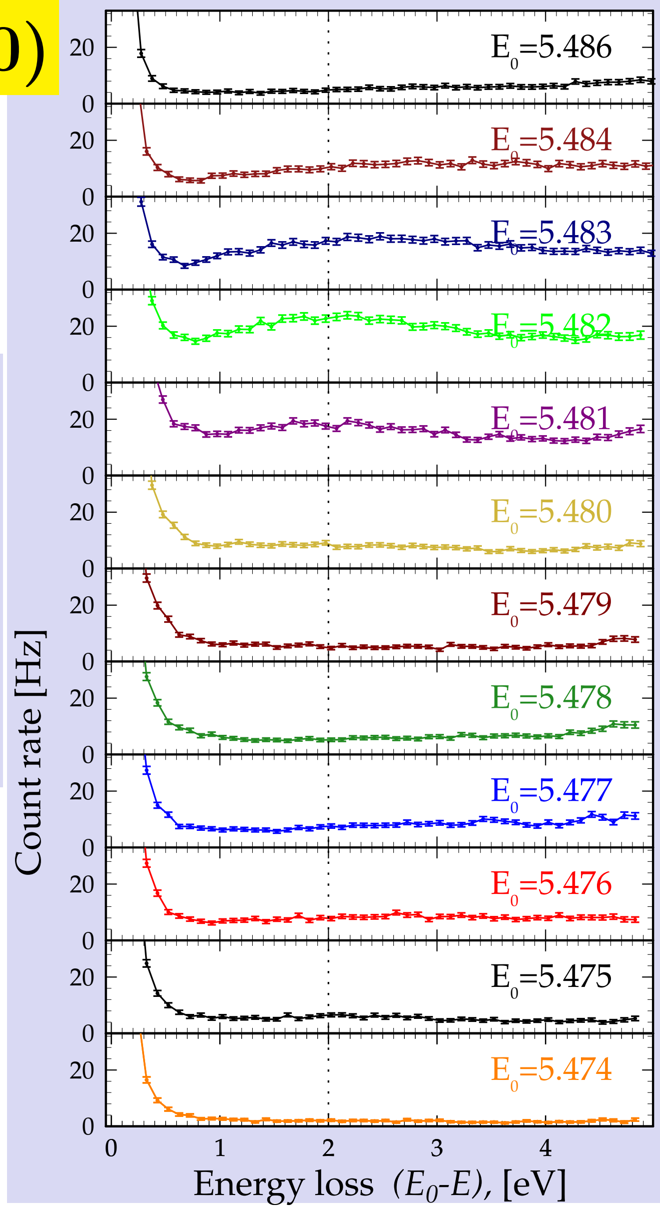
$\vec{Q} = (0.75\ 0.75\ 0)$
$\vec{e} \perp (001)$

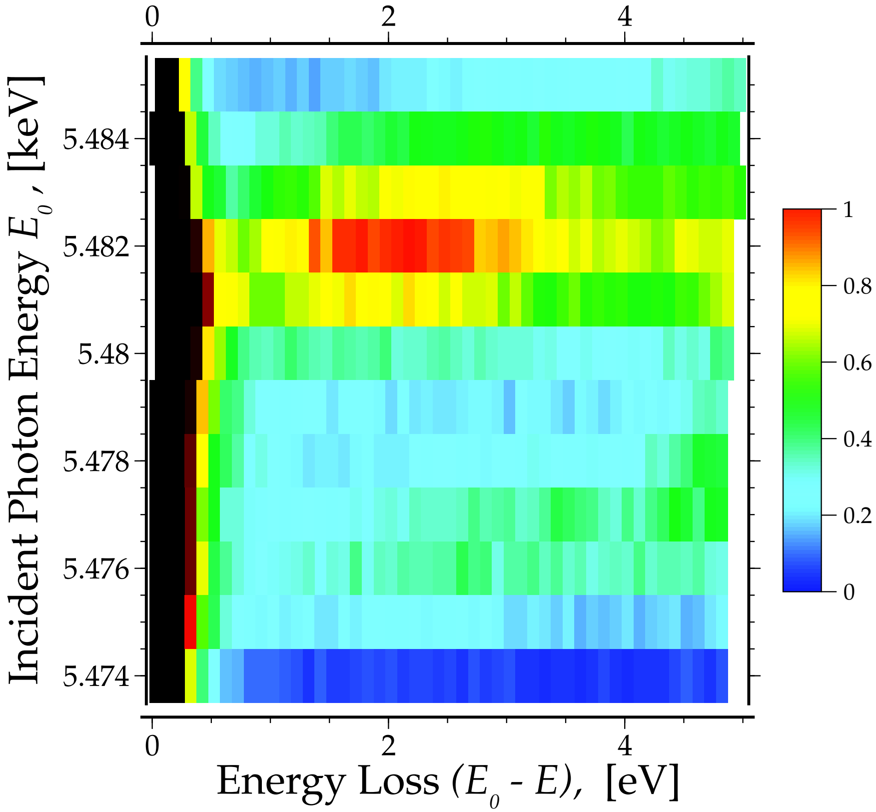
A. Said, C. Burns, E. Alp, Yu. Shvyd'ko (2007)

Measurement time 35 min/spectrum

K_α fluorescence

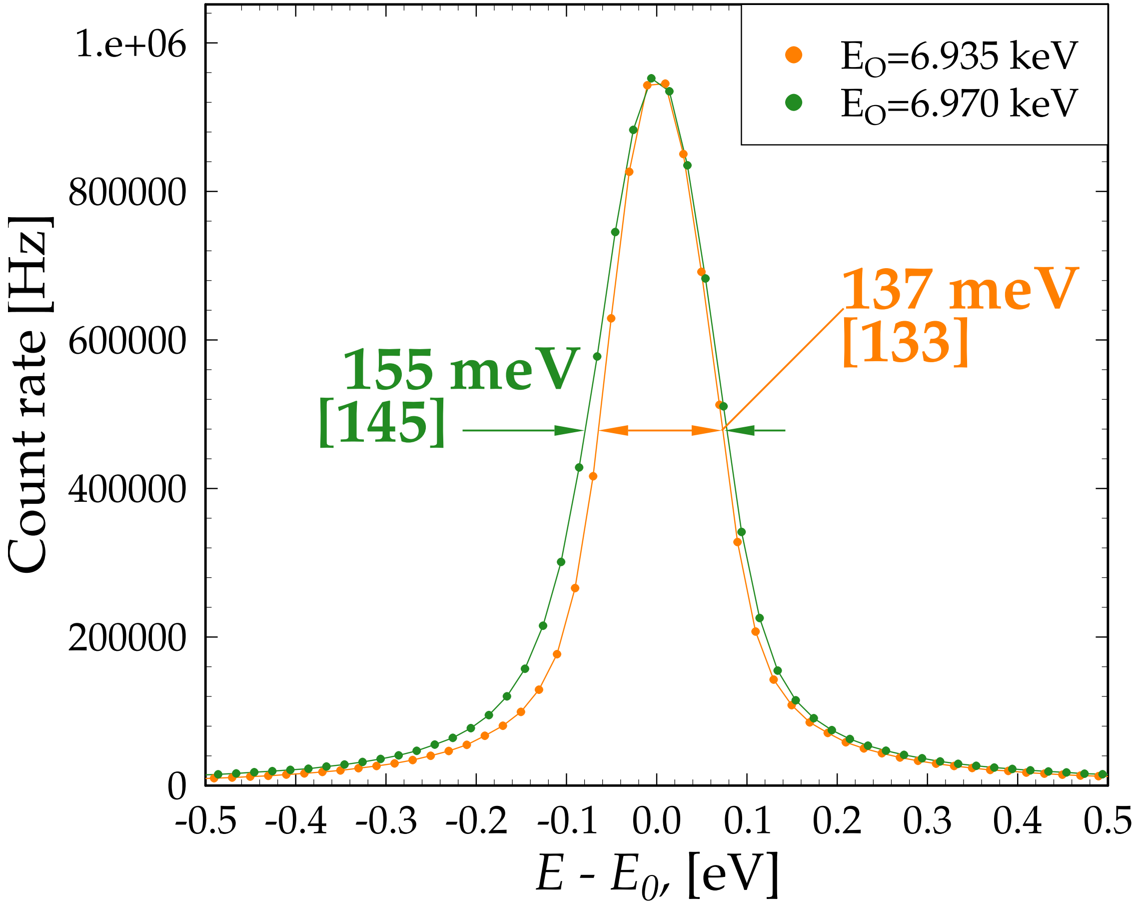


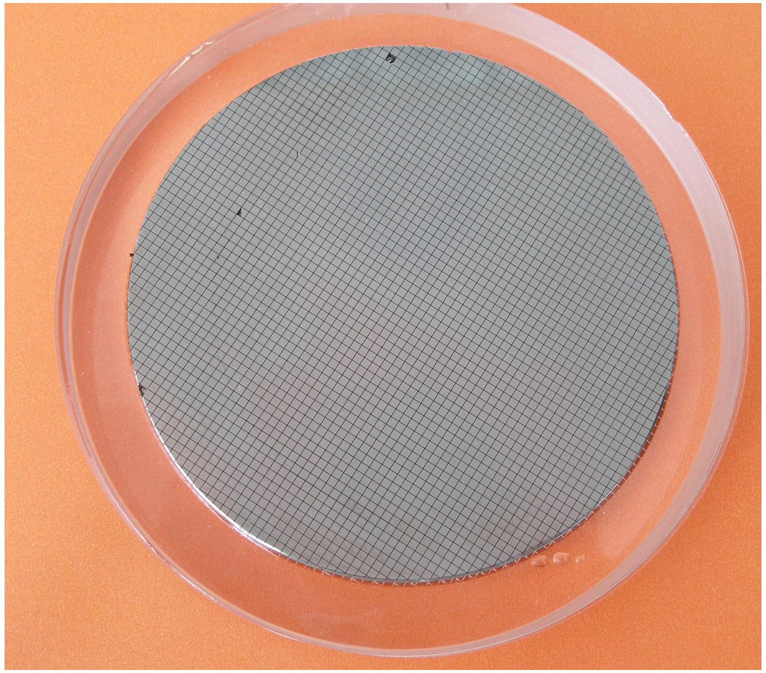




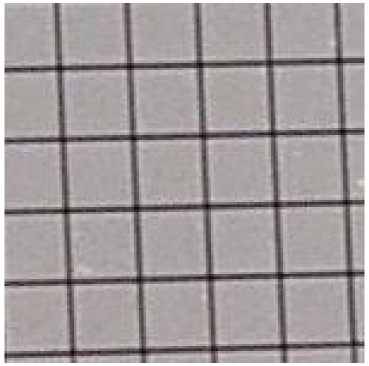
Eu L-edge RIXS Analyzer:

Ge(6 2 0) analyzer, R_A=1 m





Crystal segments:
1.5×1.5 mm^2



RIXS Analyzers for MERIX

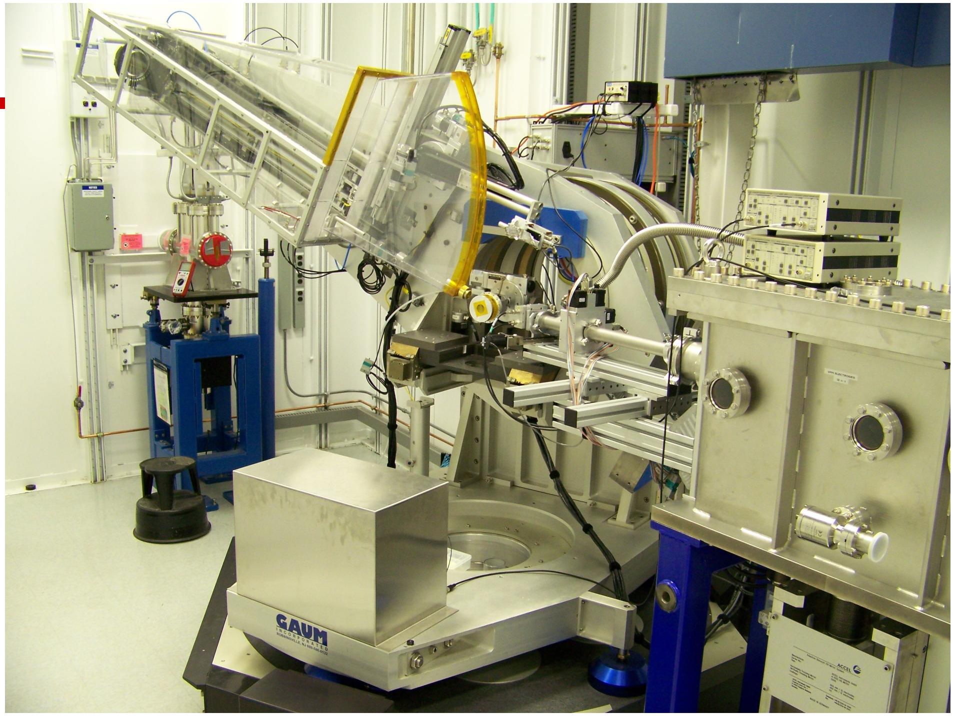
tested exist in production

Element	E [keV]	Crystal	Reflection	$\Delta E_{\rm i}$ intr. [meV]	$\Delta E_{\rm g}$ geom. [meV]	$\Delta E_{\rm tot}$ total [meV]
V(O)	5.480	$LiNbO_3$	$(0\ 0\ \bar{0}\ 12)$	109	71	130
Cr(O)	6.009	Si	(5 1 1)	52.2	61	81
Mn(O)	6.555	Si	(0 4 4)	62	72	95
Fe(O)	7.130	Ge	(6 2 0)	115	108	158
Co(O)	7.720	$LiNbO_3$	$(3\ 3\ \bar{6}\ 6)$	49	36	60
Ni(O)	8.345	$LiNbO_3$	$(0\ 6\ \bar{6}\ 0)$	50	19	54
		Ge	(2 4 6)	76	99	123
Cu(O)	8.990	Ge	(3 3 7)	42	41	59
Eu	6.977	Ge	(6 2 0)	112	51	123
Yb	8.944	Ge	(0 0 8)	64	131	145

Summary

- **November 2006: MERIX design parameters are achieved.**
- **February 2007: User operations started.**
- **February 2007: Next generation x-ray optics and detector is tested:**
    1. **Narrower spectral function (52 meV)**
    2. **Count rates increased by > 10, compared to the original design.**
- **K-edge RIXS spectroscopy in Cu, Ni, Co, Fe and V samples demonstrated.**
- **L-edge RIXS spectroscopy in Eu samples demonstrated.**
- **Analyzer tests and development is in progress**

Welcome to MERIX



MERIX became a user instrument from 2007.

The APS makes beamtime available to the international scientific community through the General User Program at:

www.aps.anl.gov/Users/Scientific_Access/General_User/index.html.

Deadline for proposals for run cycle 2008-1 is 5-NOV-07