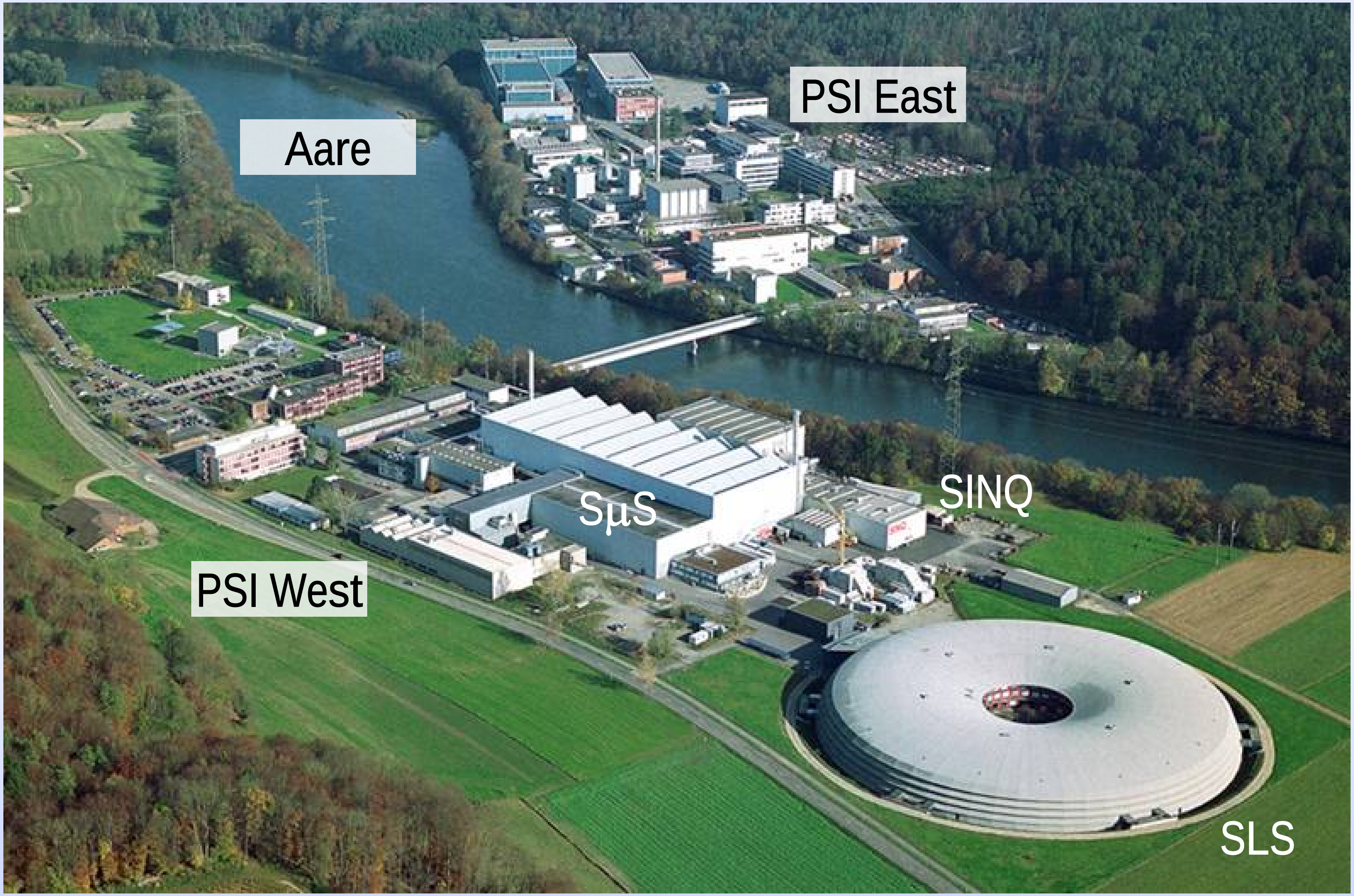
High-resolution soft-X-ray beamline ADRESS at Swiss Light Source for resonant X-ray scattering and angle-resolved photoelectron spectroscopies

V.N. Strocov and T. Schmitt

Swiss Light Source, Paul Scherrer Institute, Switzerland

Swiss Light Source @ Paul Scherrer Institute: Aerial view

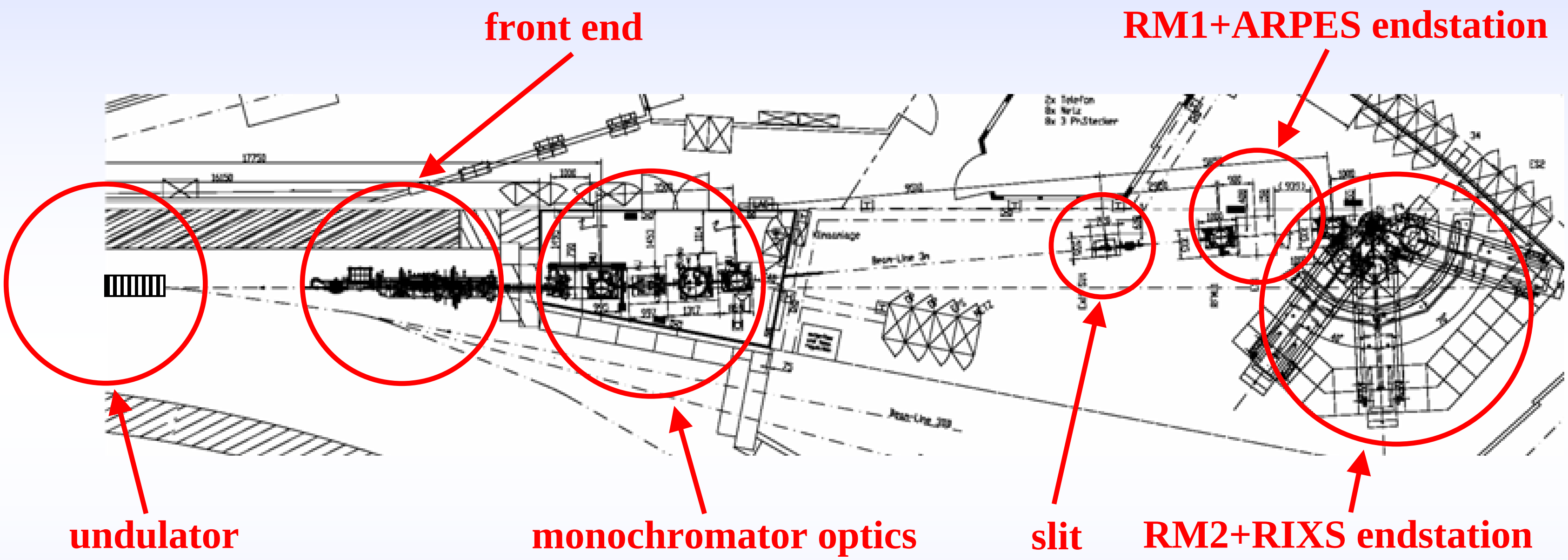


ADRESS (*ADvanced RESonant Spectroscopies*) beamline :

- soft-X-ray radiation with circular and 0-180^o variable linear polarizations
- energy range 300 – 1800 eV
- high resolution ΔE ~ 30 meV @ 1 keV
- collimated-light PGM optical scheme
- endstations:

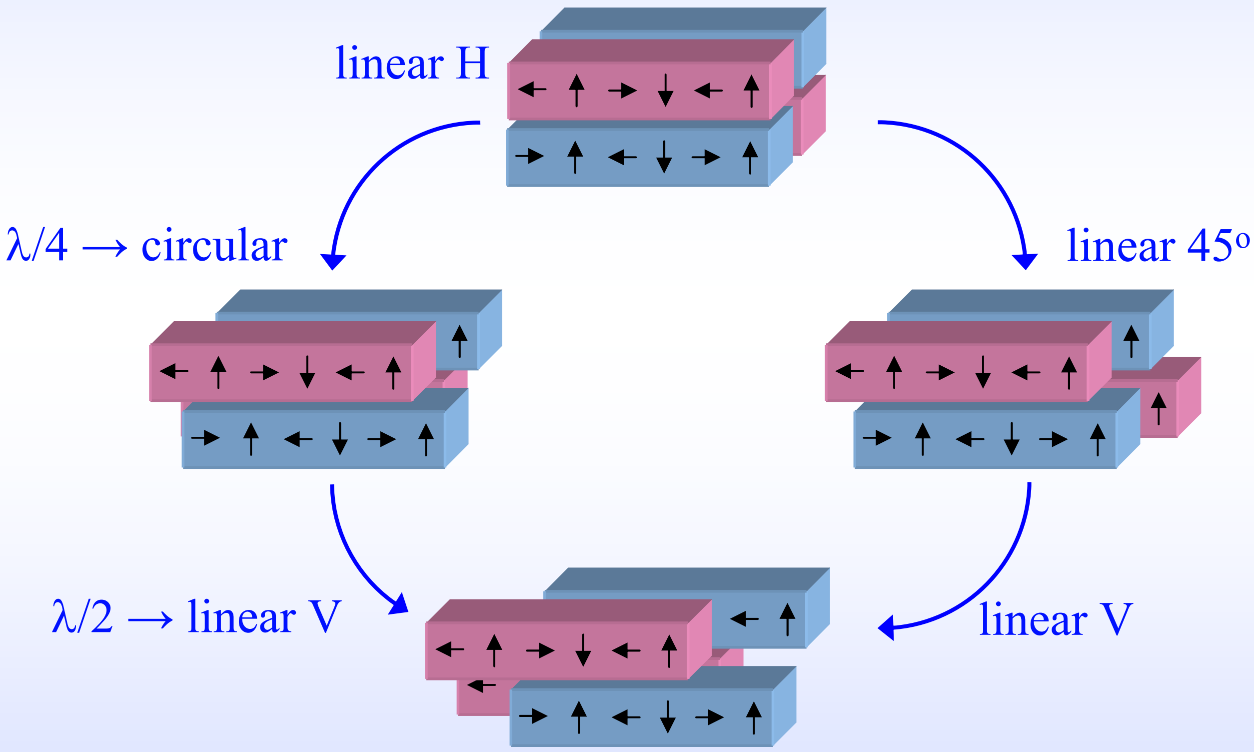
- resonant inelastic X-ray scattering (RIXS): ΔE~70 meV @ 1 keV
- angle-resolved photoelectron spectroscopy (ARPES)

Beamline layout



Undulator

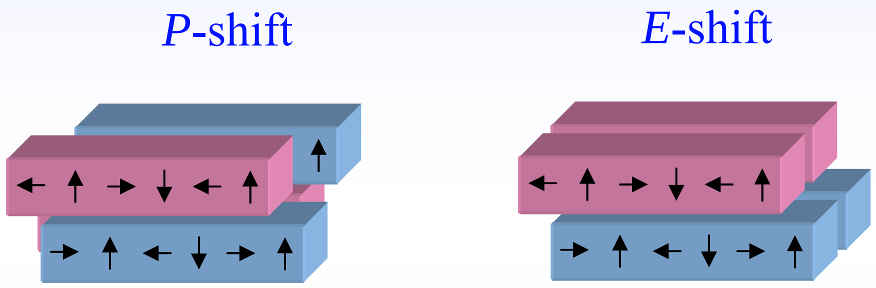
- Starting point: Apple-II type permanent magnet design



• 6 motors (*P*-shifts+gap), complicated design

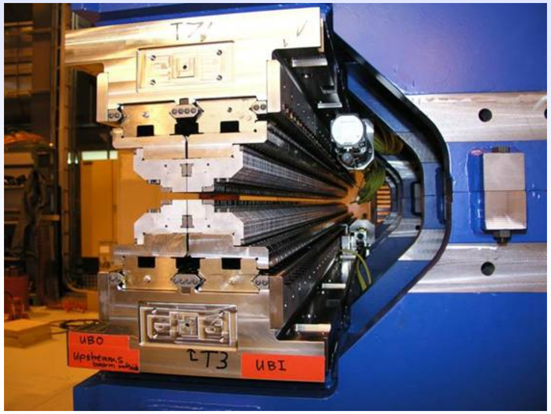
Undulator: Concept

- Apple-2 permanent magnet design with fixed gap (concept by R. Car)

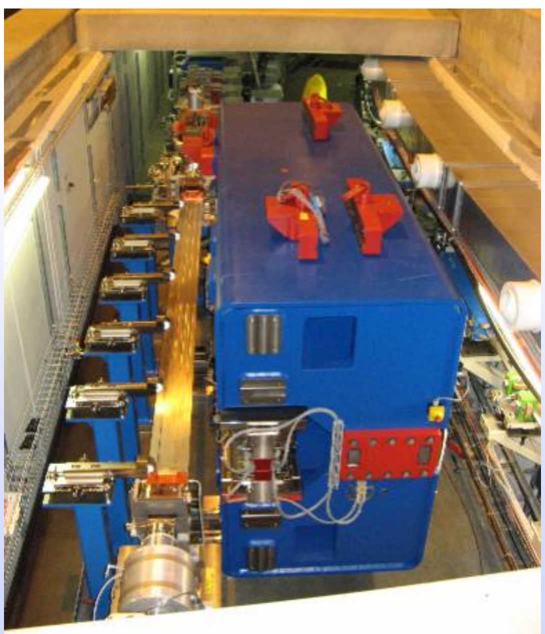


☺ full functionality (circular + linear 0-180° polarizations)
☺ simple and mechanically rigid design (4 motors)
☹ polarization and *E* coupling requires complicated mathematical models

Undulator: Design (T.Schmidt's group)

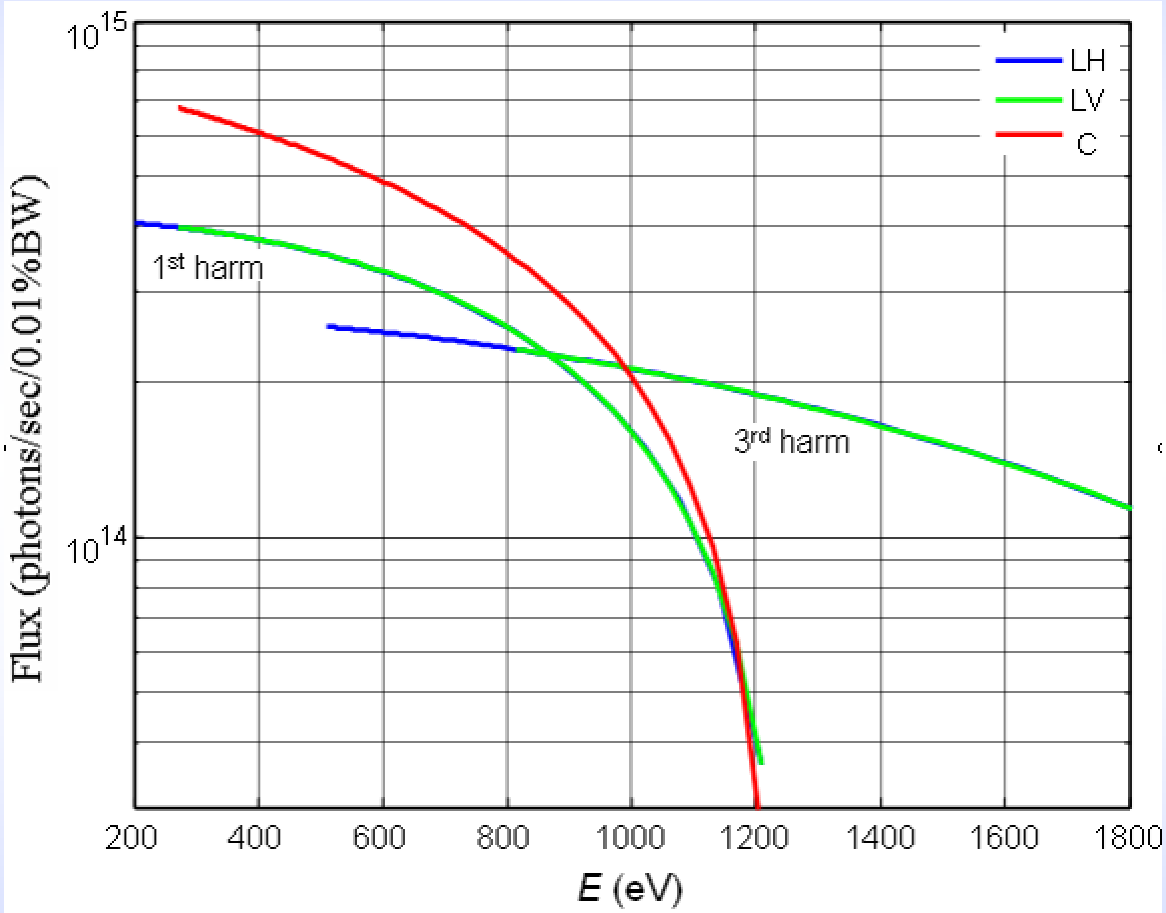


- mechanically rigid C-like construction
- λ=44 mm (optimized for *hv* = 400-1800 eV), *L*=3.5 m



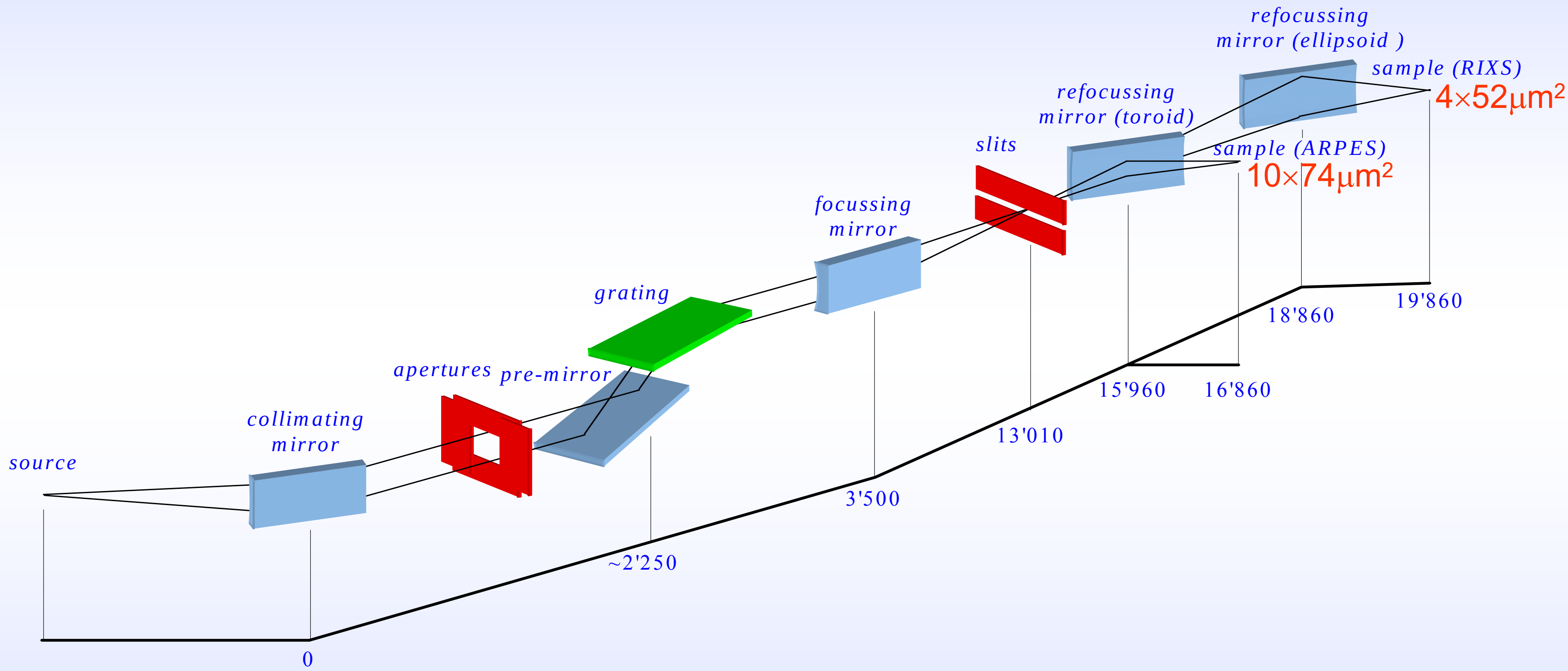
- world's first fixed gap undulator

Undulator: Performance



- gap reduced to 11 mm => no V-pol flux discontinuity around 1000-1200 eV (Zn,Ga,Ge $2p_{3/2}$; La,Ce $3p_{3/2}$)
- source @1000 eV: $\sigma_X \times \sigma_Z = 0.107 \times 0.014$ mm, $\sigma'_X \times \sigma'_Z = 0.047 \times 0.014$ mrad

Optical scheme : Collimated-light PGM



- high resolution
- no entrance slit: high flux
- wide energy range
- resolution, flux and HIOS optimization by C_{ff}
- proven design and flawless operation @ SLS

Monochromator optics: Resolution optimization

- goal: $E/\Delta E > 30\ 000$ @ 1 keV
- tools: ray tracing code PHASE (J. Bahrdt, U. Flechsig)

Slope errors optimization

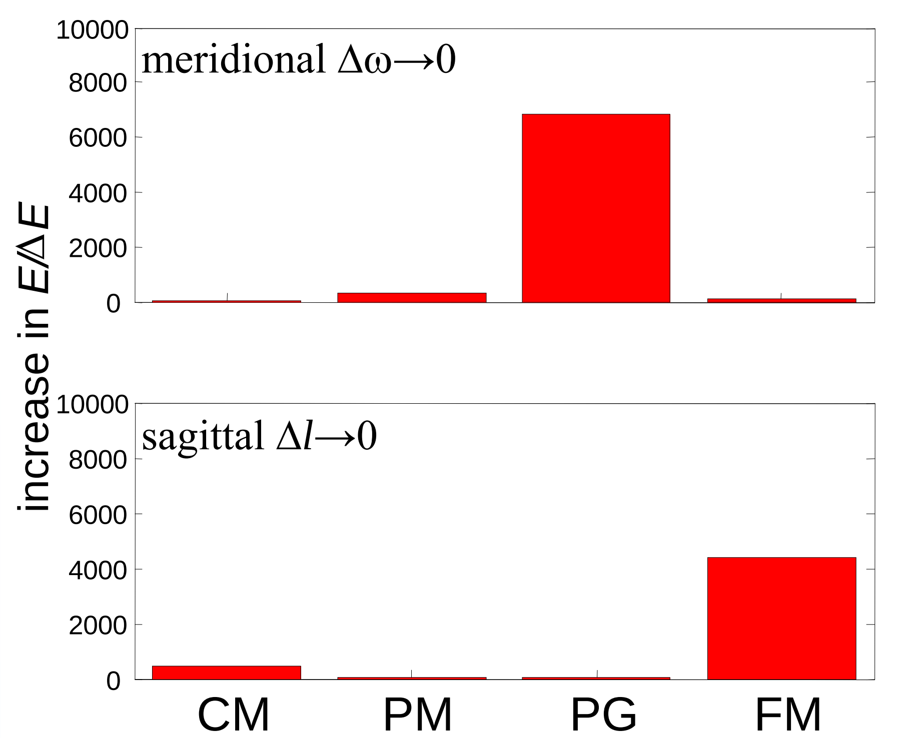
- starting point: 4800/mm grating in 1st order, $f = 10\ 000$ mm

- ideal optics → ***E*/Δ*E* = 65000**;
  real optics $(\Delta\omega/\Delta l)_{PO} = 0.5/5\mu$rad, $(\Delta\omega/\Delta l)_{TO} = 2.5/25\mu$rad → ***E*/Δ*E* = 16700**

- which are the most critical elements?

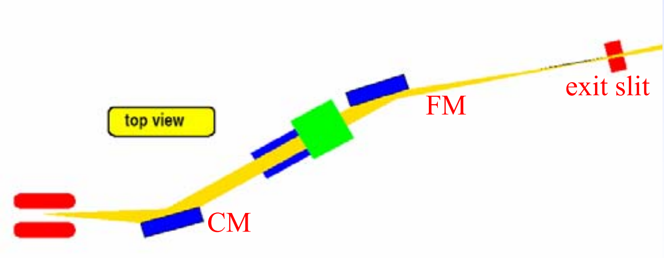
• most critical are $\Delta\omega_{PG}$ and Δl_{FM}

- vendors: $\Delta\omega_{PG} = 0.375$ µrad,
$\Delta l_{FM} = 7.5$ µrad possible → ***E*/Δ*E* ~ 30000**

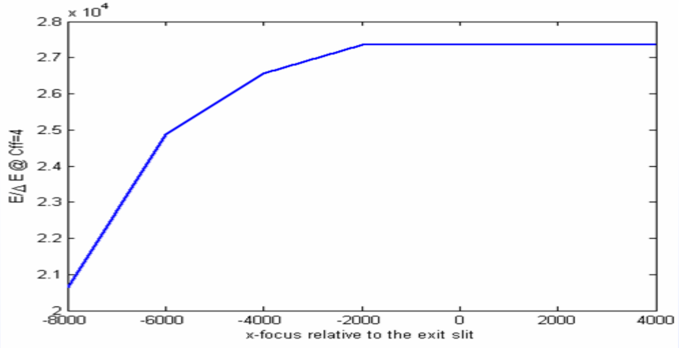


Beamline geometry optimization

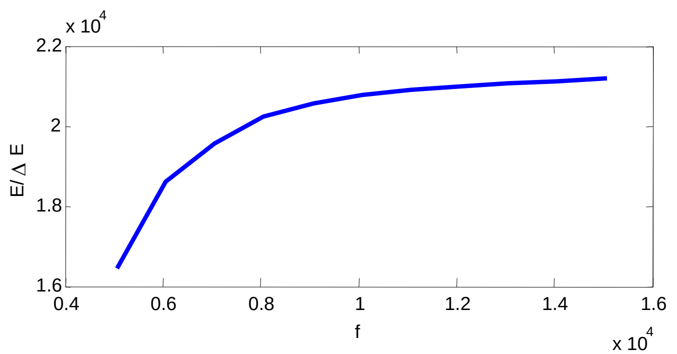
(1) horizontal focussing schemes
- collimation by CM + focusing by FM
- cylinder CM, focusing by FM
- focusing by CM, cylinder FM
• $E/\Delta E$ improves by ~1000



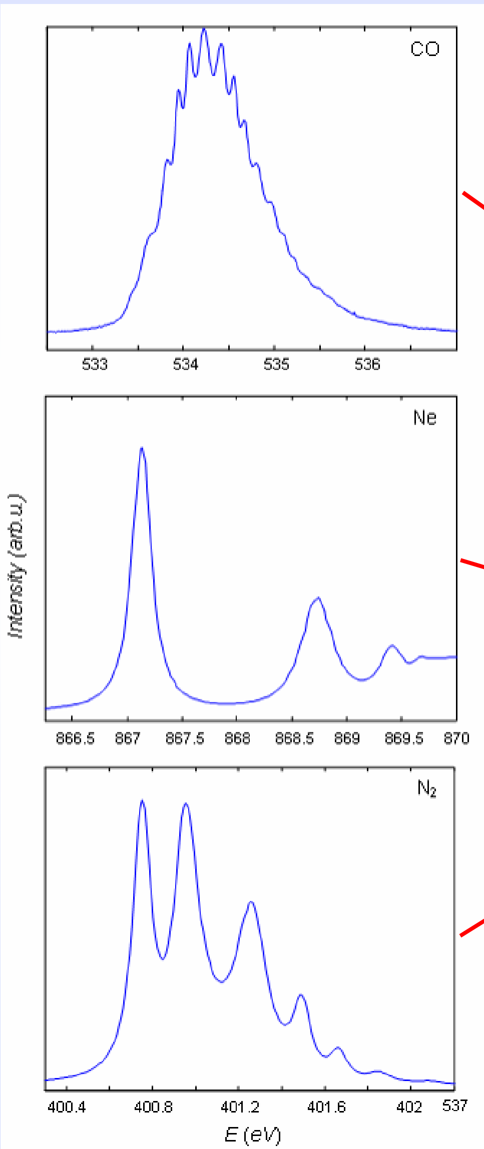
(2) astigmatism
• best $E/\Delta E$ @ stigmatic focus



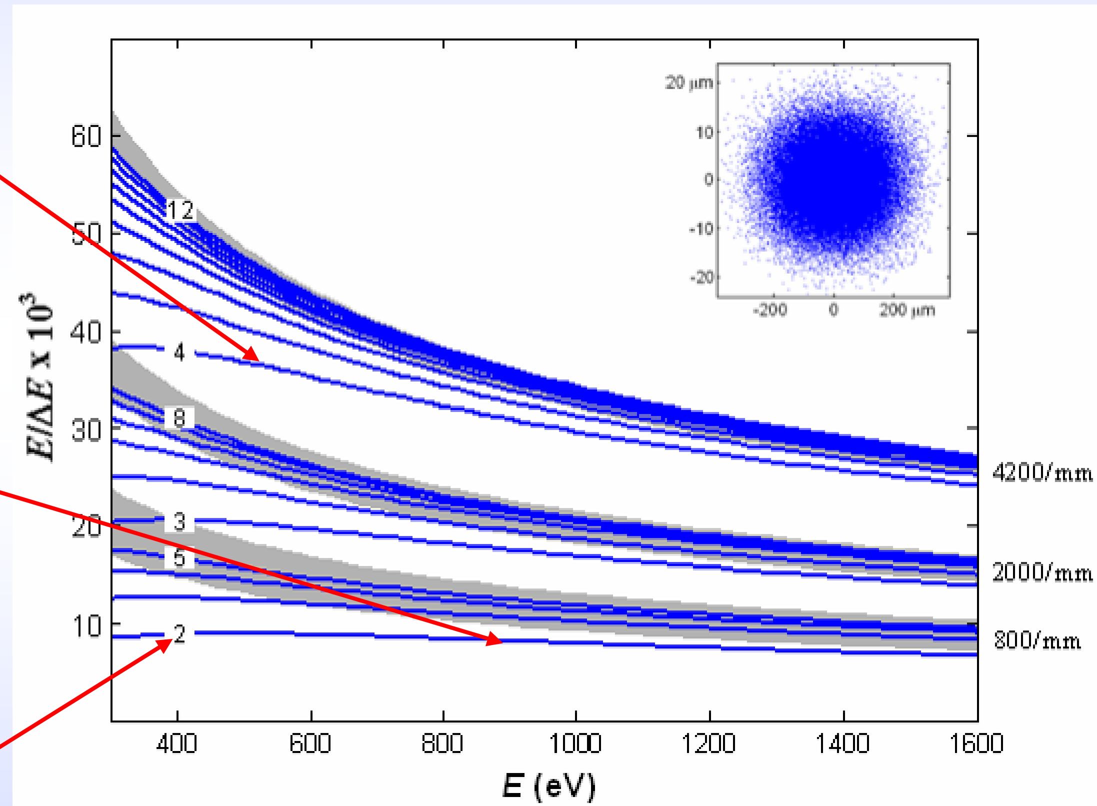
(3) dispersion arm
• saturation @ ~14 m (~10 m available)



Resolution with the optimized parameters



- diffraction contribution $E/\Delta E = N$ included



- FWHM spot size = 14.1×228 µm^2, almost no aberrations
- $E/\Delta E$ up to 33000 @ 4200/mm, C_{ff}=10
- optimal resolution coverage with 800, 2000 and 4200/mm

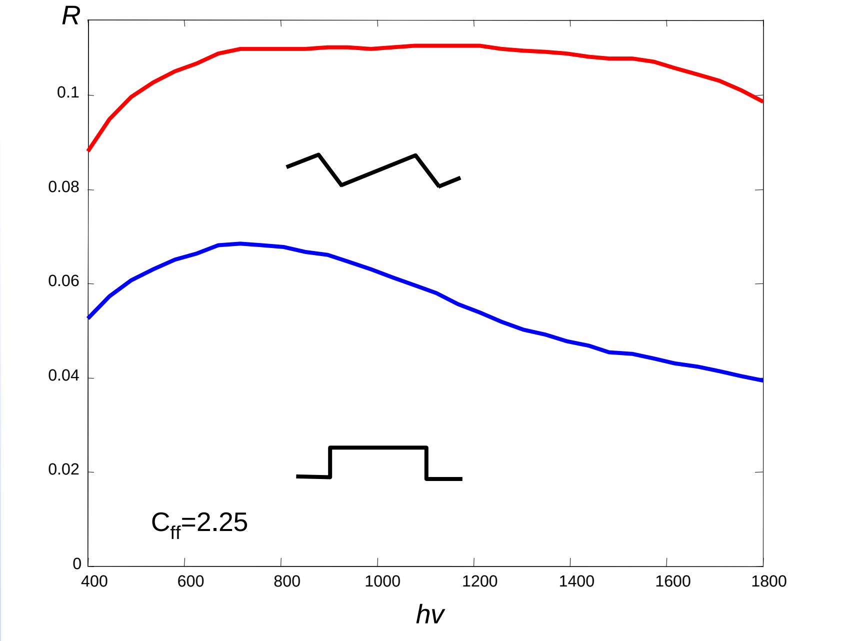
Gratings: Flux optimization

- Lamellar or blazed? if lamellar, *h* and *c*/*d* ?
- Tools: Grating efficiency code REFLEC (Nevier+BESSY)

Blazed *vs* lamellar

- 2000/mm ideal blazed (ϕ_{blaze}=1.3^{o} optimized @ 930 eV, C_{ff}=2.25)
vs ideal lamellar (*h*=5.5nm, *c*/*d*=0.6 optimized @ 700-1100 eV, C_{ff}=2.25)

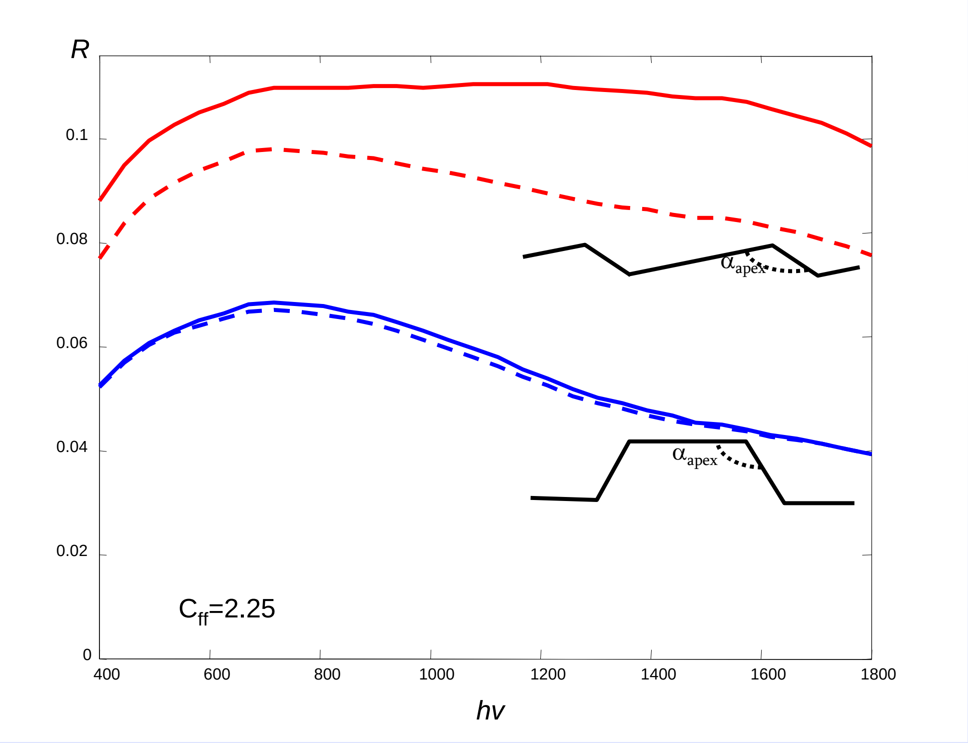
- the blazed betters on flux + flatness of the energy dependence



Gratings: Blazed *vs* lamellar

- 2000/mm realistic profile: α_{apex}~170° for blazed, 164° for lamellar

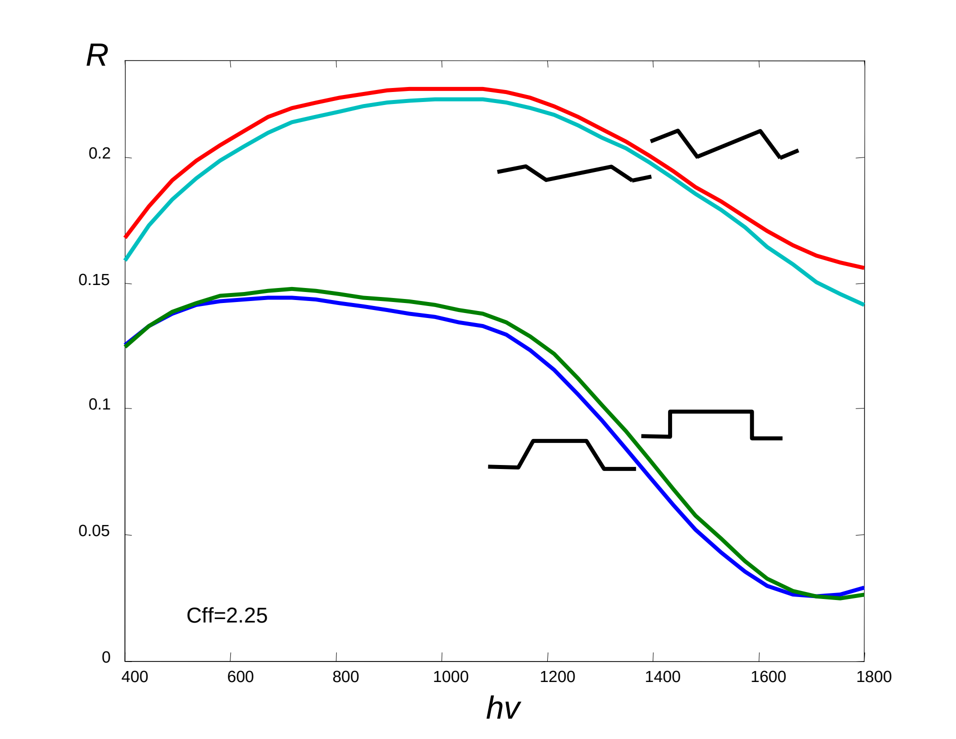
• advantages of the blazed on flux and flatness degrade



Gratings: Blazed *vs* lamellar

- 800/mm blazed (ϕ_{blaze}=0.8°) *vs* lamellar (*h*=11 nm, *c*/*d*=0.69), ideal and realistic

• for lower l/mm advantages of the blazed on flux and flatness preserve



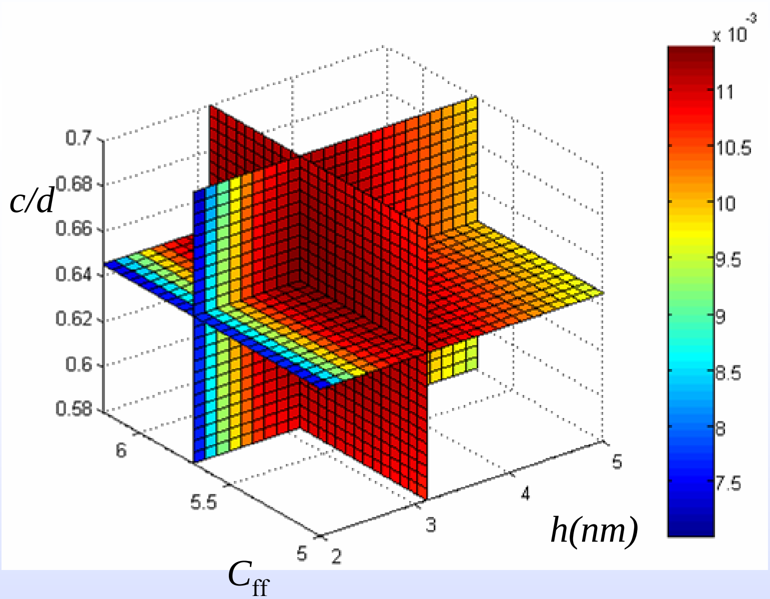
=> blazed 800/mm (high flux, low res + HIOS) = **'flux'** grating;
lamellar 2000/mm (low flux, high res + HIOS) = **'workhorse'** grating
lamellar 4200/mm (lowest flux, highest res) = **'hi-res'** grating

Gratings: Optimization of lamellar gratings

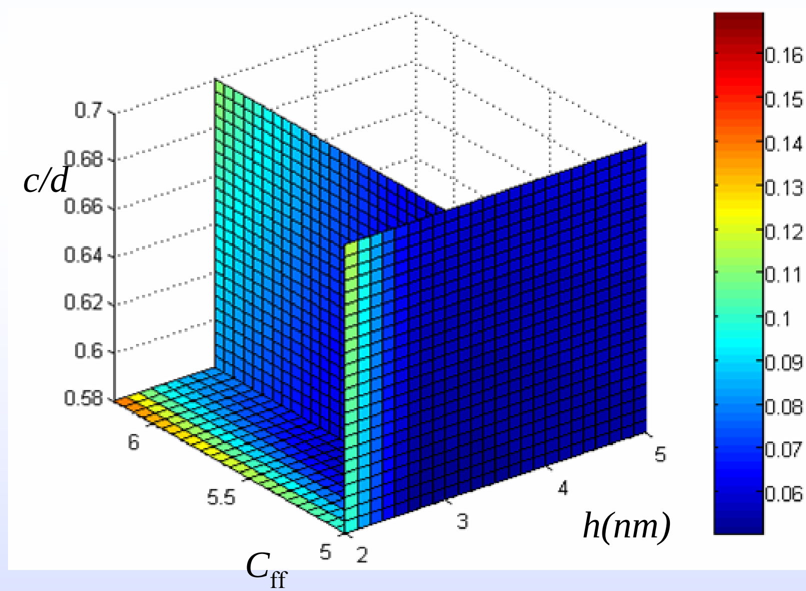
- *h, c/d*, C_{ff} to optimize the flux, energy dependence flatness, HIOS interplay
- PM(C_{ff}) to be included

- realistic 2000/mm (α_{apex}=164^o), *hv*=700-1200 eV

<reflectivity>



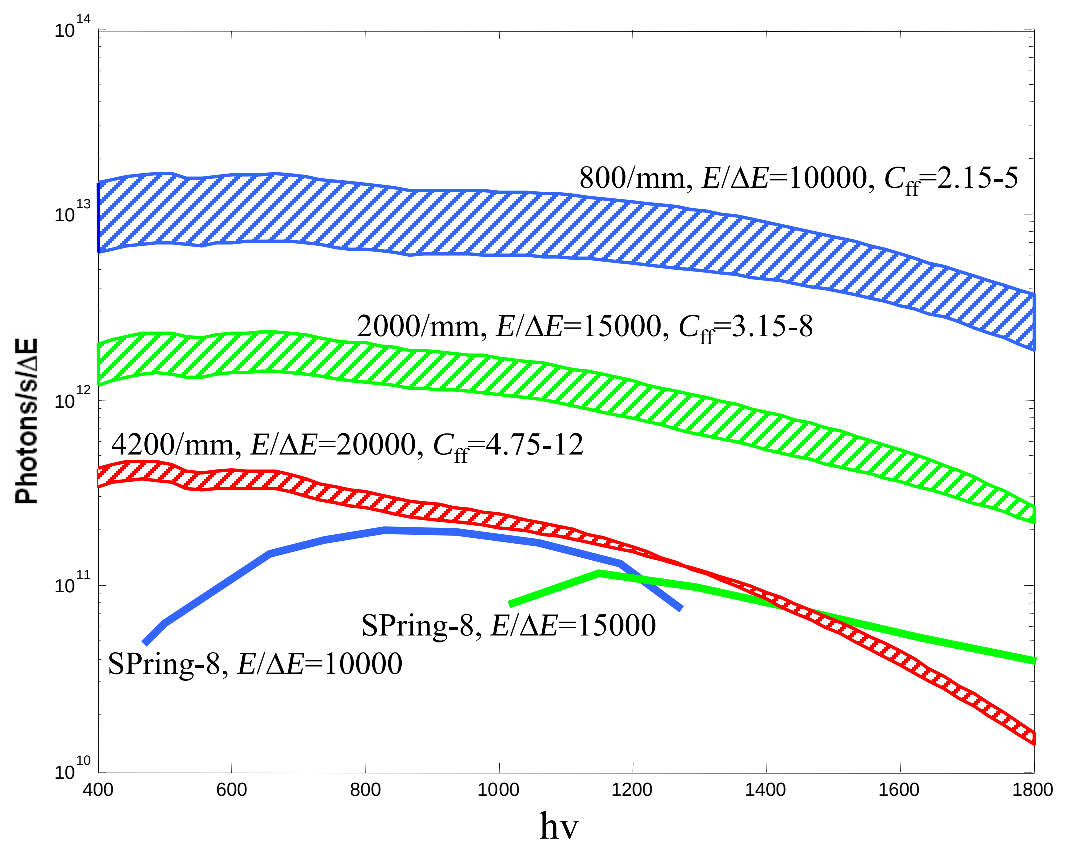
<reflectivity variation>



- optimal *h, c/d*, C_{ff} taken slightly shifted from the flux maximum towards better flatness + HIOS

Beamline flux performance with the optimized gratings

- flat energy dependence with all gratings including 800/mm blazed
- flux-optimal C_{ff} increases with l/mm and energy
- 3×10^{11} to 1×10^{13} ph/s/0.01%BW (experimentally confirmed): factor of 10 to 100 flux increase or ~2 improvement in $E/\Delta E$ compared to BL25SU@SPring-8



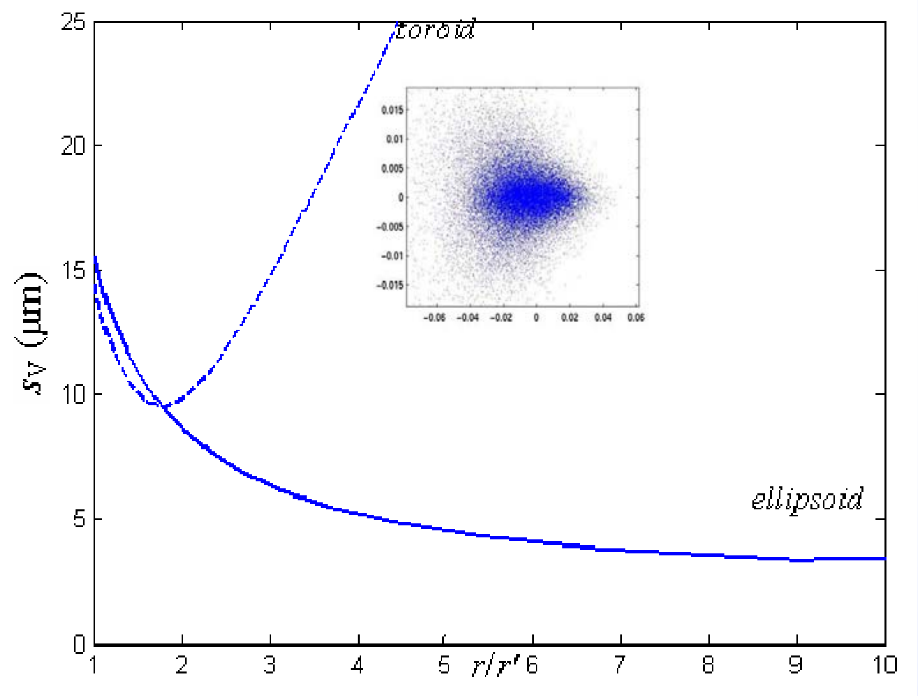
- excellent flux by virtue of (1) 2.4 GeV ring optimal for soft X-rays; (2) glancing angles on the mirrors; (3) minimal l/mm; (4) blazed/lamellar and profile optimization of gratings

Refocusing optics

- vertical spot size << 10 μm required for slitless operation of the RIXS spectrometer

Toroidal vs Ellipsoidal mirror

- ray tracing:
focused spot size at the exit slit 14.1μm, $r+r'$ = 7000 mm, grazing angle 89º, $\Delta\omega/\Delta l$ slope errors 0.5/1.5 μrad for TM and 1.5/4.5 μrad for EM

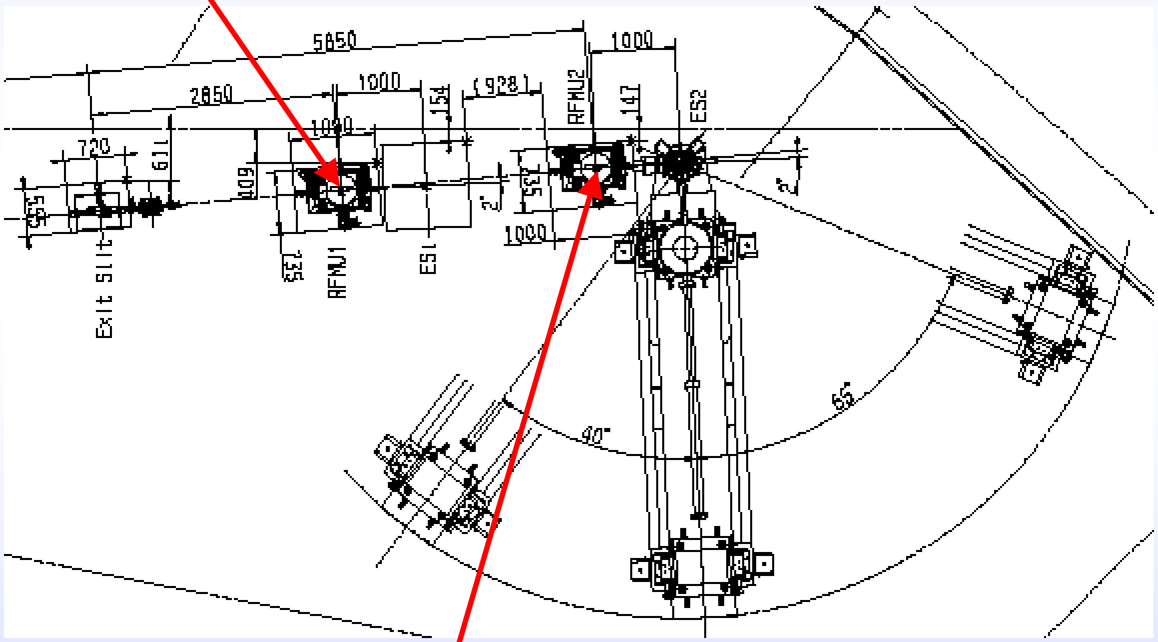


TM: aberrations for large r/r'; minimal s_v ~ 10 μm @ r/r' ~ 1.8 − inacceptable ☹

EM: decrease of s_v carries on towards ~ 3.4 μm @ r/r' ~ 9 − slitless operation of the RIXS spectrometer possible ☺

Refocusing optics layout

ARPES: moderate spot size and available *r/r'* ⇒ **TM**
- actual s_v ~ 10 μm @ *r/r'* ~ 2

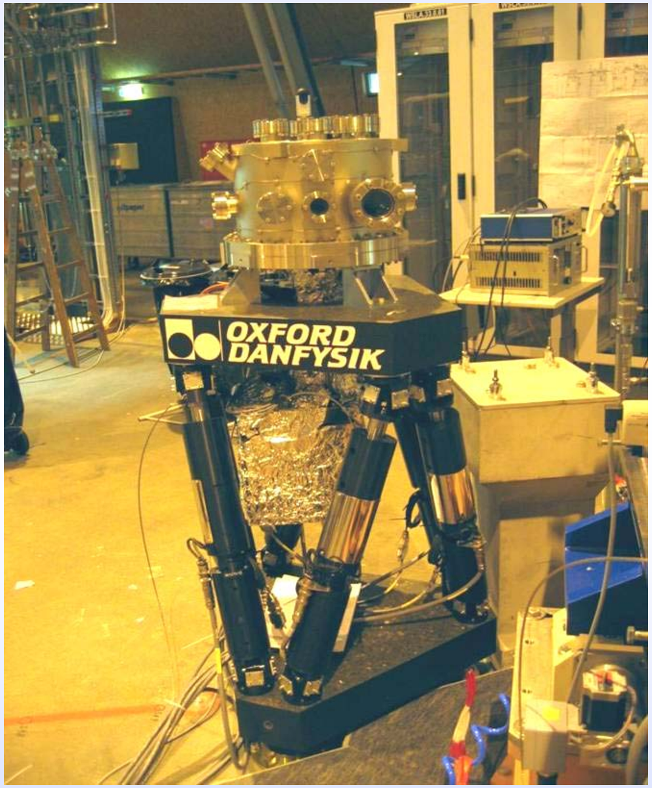


RIXS: maximal demagnification ⇒ **EM**
- due to limitation of *r'* the actual s_v ~ 3.9 μm @ *r/r'* ~ 5.85
- slope errors are crucial: EM from ZEISS with $\Delta\omega/\Delta l$ = 1.5/7.5 μrad

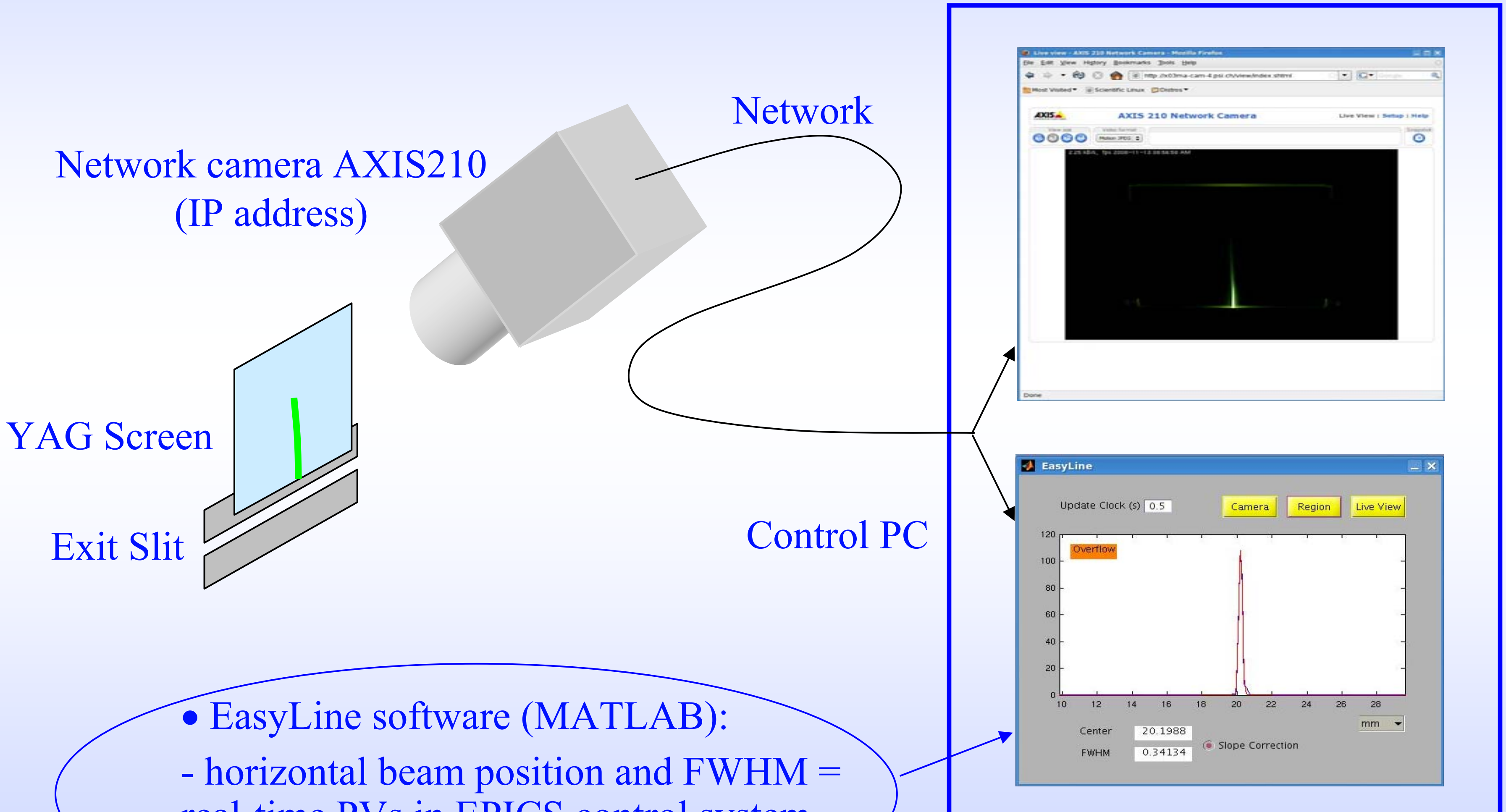
Refocusing mechanics

- hexapod systems (OXFORD-DANFYSIK):

- 3 translational + 3 soft-axis angular DOFs
- high setability of 1 μm and 1 μrad
- soft axes: mirror center 100 mm downstream



Alignment tools: Horizontal beam profile monitor

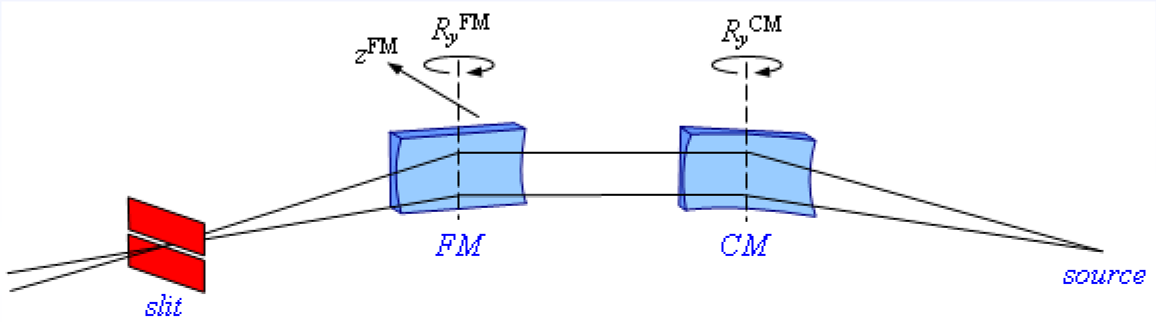


- EasyLine software (MATLAB):

- horizontal beam position and FWHM = real-time PVs in EPICS control system

- effective horizontal focusing tool

Alignment strategies: Vertical focusing scheme

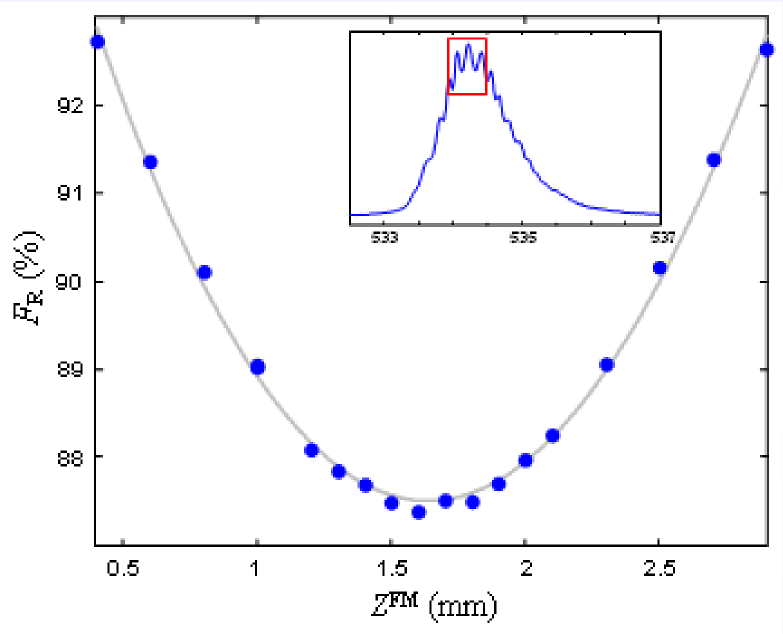


• Beam position at the slit + aperture matching constrains =>
R_y^{FM}, z^{FM} and R_y^{CM} are entangled in one *combined focalization motion* (CFM)

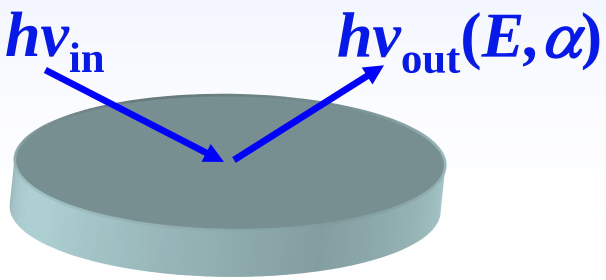
• 3 DOFs (R_y^{FM}, z^{FM} and R_y^{CM}) reduced to 1 DOF (CFM) parametrized by z^{FM} =>
- fast and unambiguous focalization
- maximal transmission
- maximal resolution due center of the optical surface

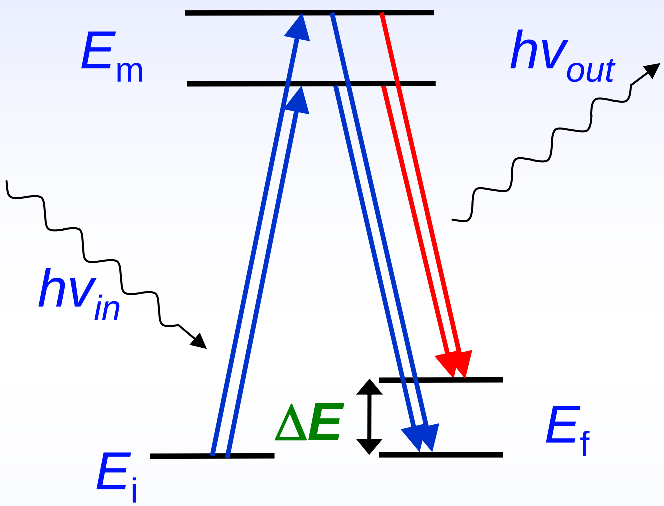
Alignment strategies: Example of focalization

- Typical focalization curve (1-2 hrs)



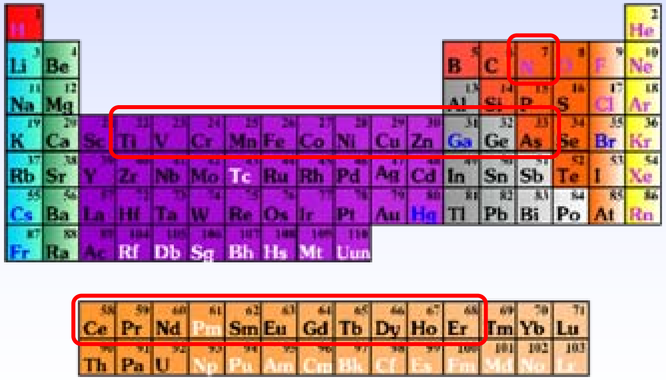
RIXS endstation: Technique



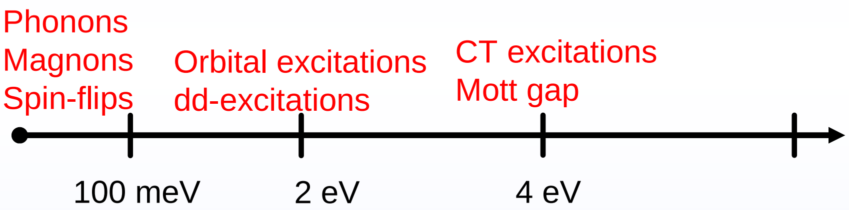


- ΔE difference between hv_{in} and hv_{out} $\Rightarrow$ spectrum of low-energy excitations in correlated materials
- probing depth ~300 nm: bulk properties, buried nanostructures…
- element specific electronic structure

High-resolution RIXS endstation: Concept

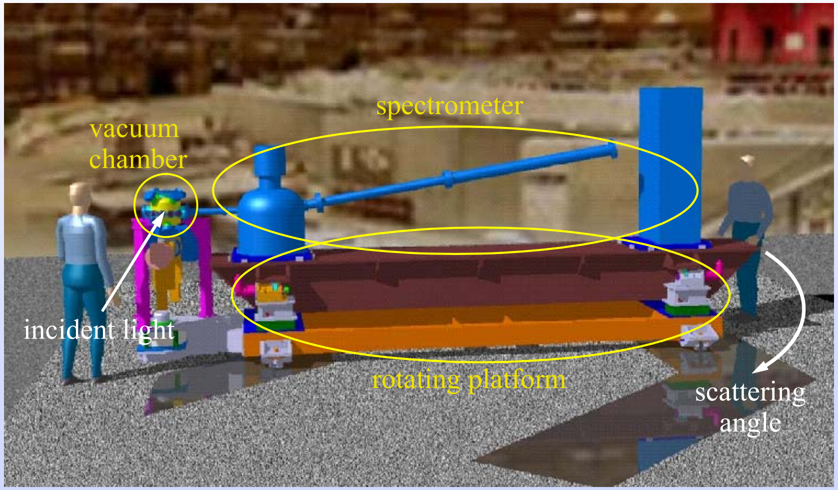


- *hν* = 300-1800 eV:
  - N *K*-edge, Ga,Ge,As *L*-edges: microelectronics…
  - TMs *L*-edges, REs *M*-edges: correlated systems (superconductivity, CMR, metal-insulator transitions…)



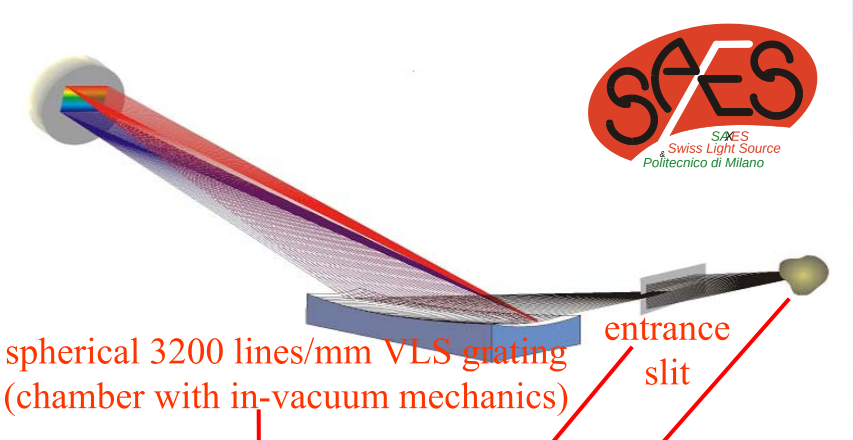
- Δ*E* ~100 meV @ 1 keV to go from *d-d* and *f-f* excitations towards magnons and phonons

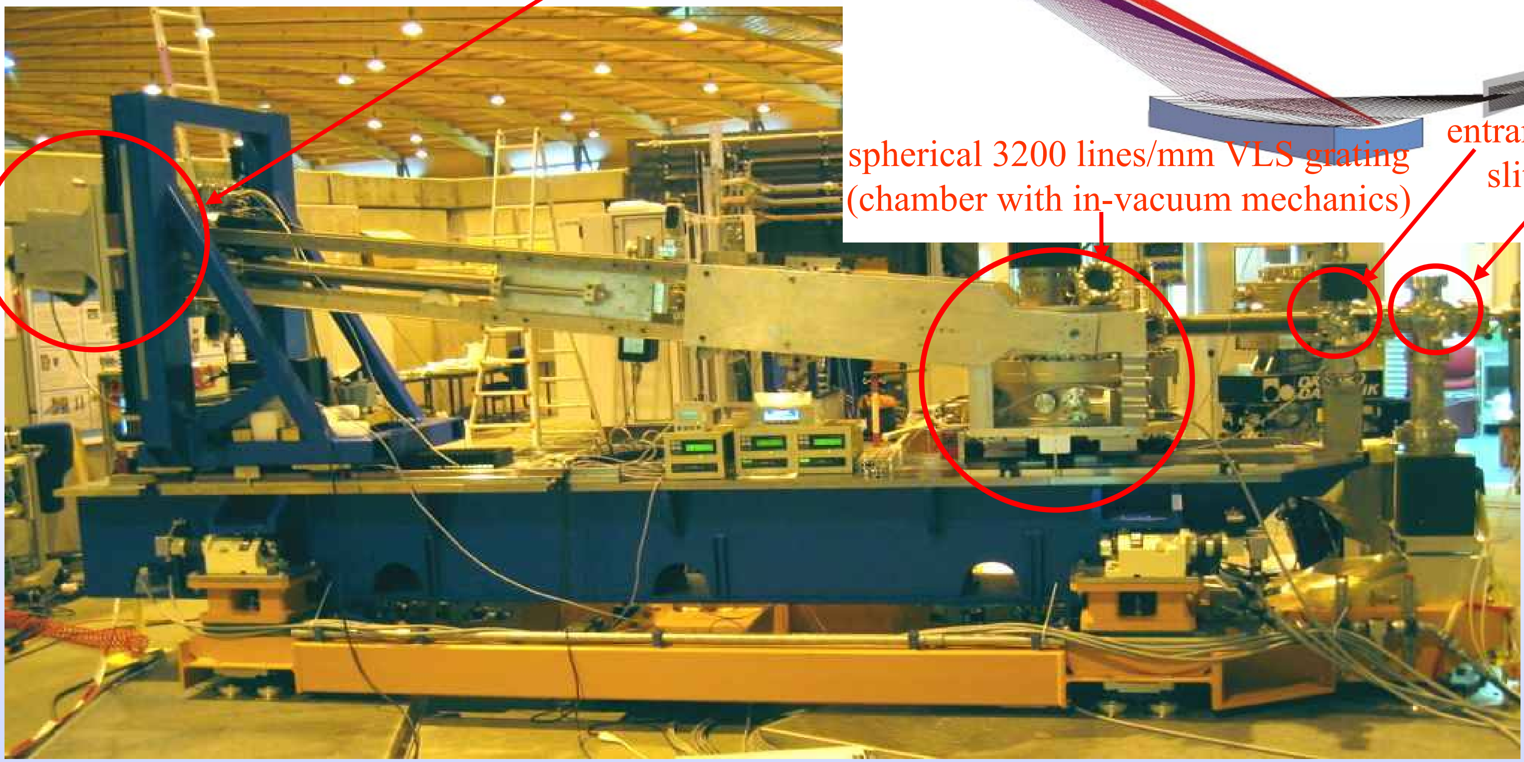
- variable scattering angle to study ***q***-dependences



RIXS endstation: Super Advanced X-ray Spectrometer (SAXES)

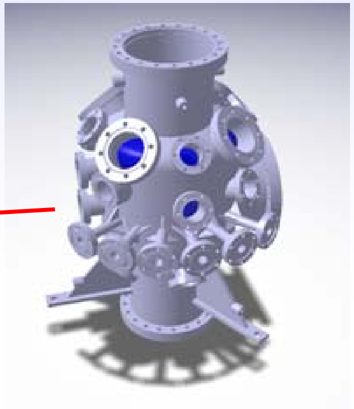
- optics by Politechnico di Milano (group of G. Ghiringhelli and L. Braicovich)
- resolving power $E/\Delta E \sim 12000$ @1 keV





G. Ghiringhelli *et al*, Rev. Sci. Instrum. **77** (2006) 113108

RIXS endstation: Rotating platform/vacuum chamber



Vacuum chamber

- 20^{o} steps in angle
- L-He_2 cryostat

Rotating platform on air cushions

- rigid I-shape (bending<7 µm)

Actuator

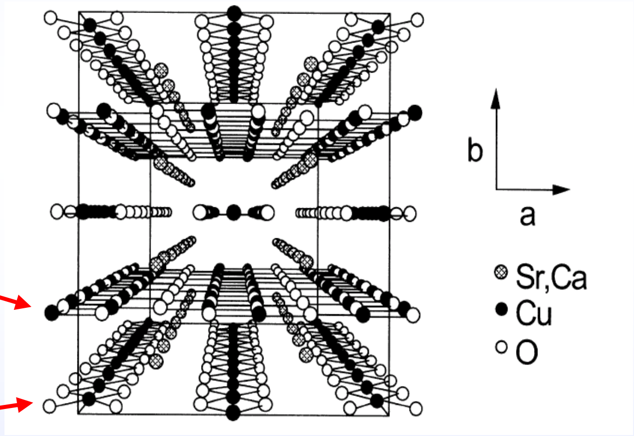
- 5 DOFs, accuracy 5 µm

A case study: 'telephone number' compound $Sr_{14}Cu_{24}O_{41}$ by Cu L_3-edge RIXS

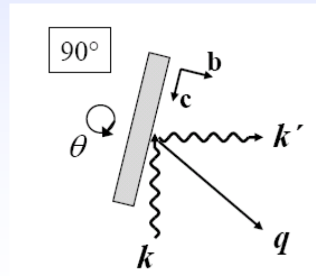
Kojima *et al*, JES **117** (2001) 237

quasi-1D spin ladders:
AFM exchange *J*~100 meV

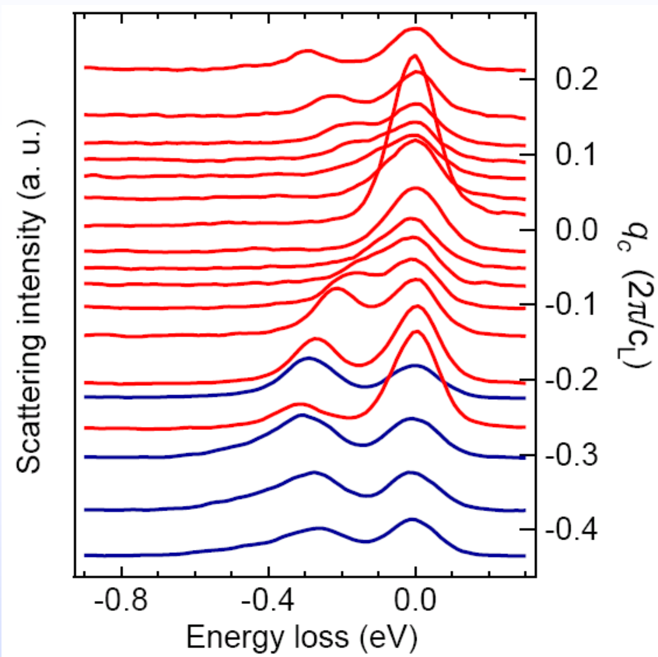
1D spin-chains:
FM exchange *J*~10 meV

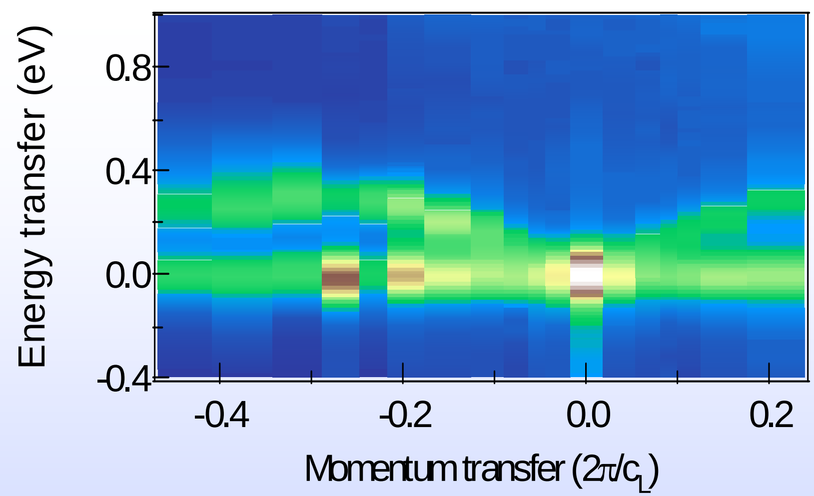


Case study: q-dispersion of magnetic excitations in 'telephone number' compound $Sr_{14}Cu_{24}O_{41}$ by Cu L_3-edge RIXS



J. Schlappa, T. Schmitt *et al*, PRL **103** (2009) 047401

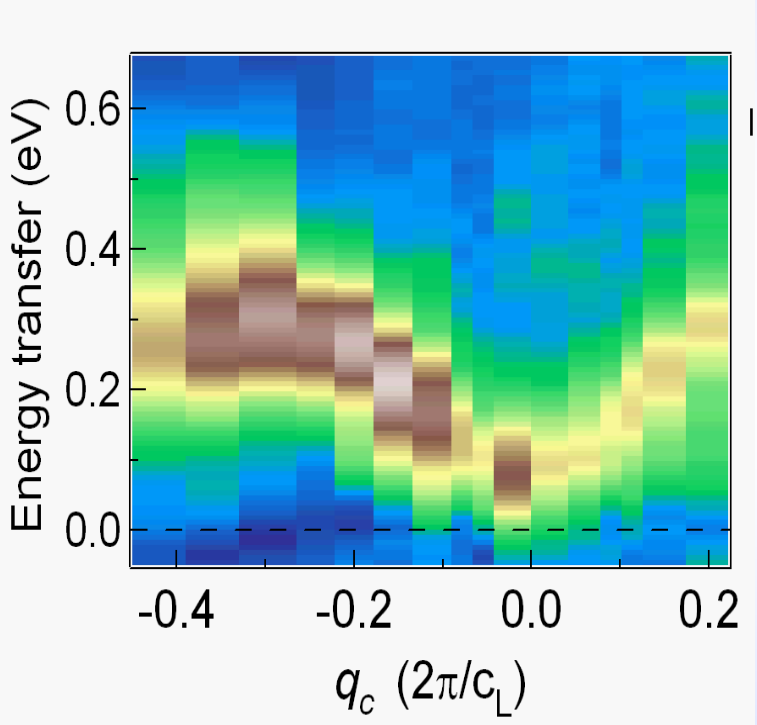




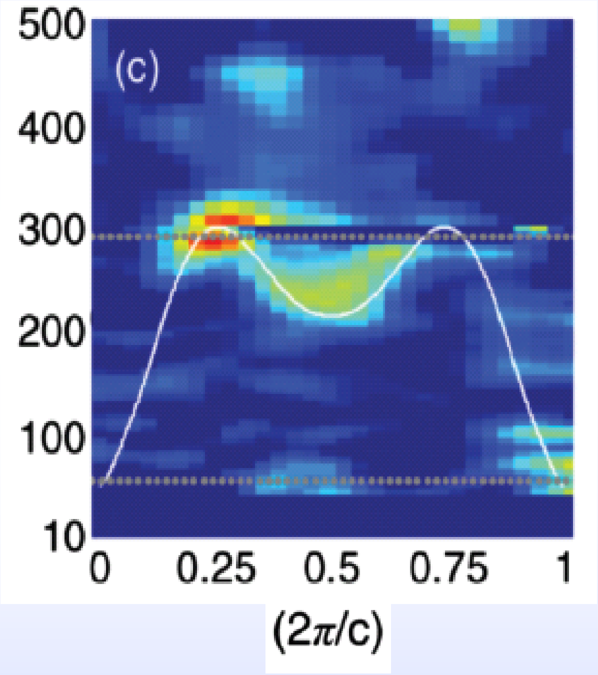
- two-triplon excitations in the ladder subsystem (AFM exchange *J*~100 meV)

RIXS vs Inelastic Neutron Scattering (INS)

RIXS from $Sr_{14}Cu_{24}O_{41}$



INS from $La_4Sr_{10}Cu_{24}O_{41}$
S. Notbohm *et al*, PRL **98** (2007) 027403



- flat cross-section over the full BZ
- ΔE ~100 meV and E-scale up to 3 eV

- low sensitivity for small q-transfer
- ΔE ~10 meV and E-scale up to ~ 500 meV

RIXS is complementary to INS on the energy scale and q-transfer region

Design of spherical VLS grating spectrometers

- Dedicated ray-tracing software TraceVLS allowing fast optimization of the grating parameters and spectrometer geometry

- Example: Model spectrometer with $E/\Delta E$=15000 @ 930 eV

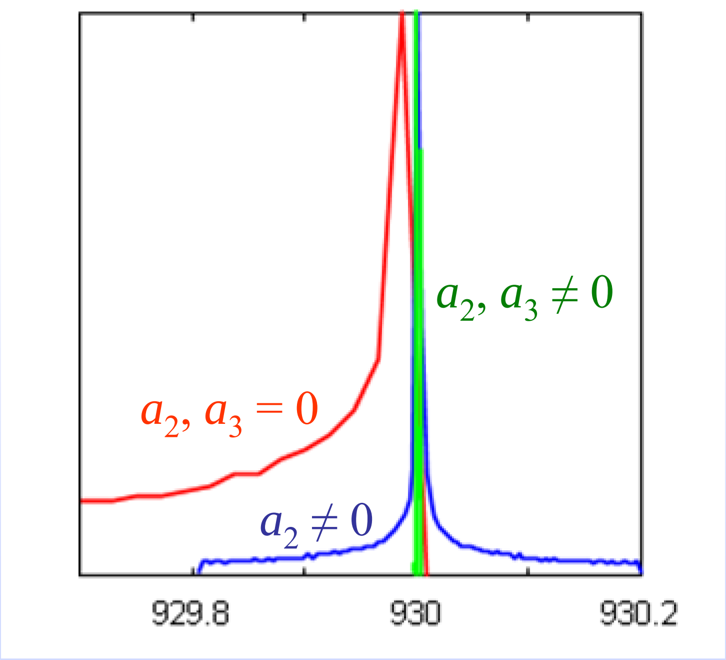
Step 1: Optimization of the grating parameters for reference E=930eV

Groove density $a(\omega) = a_0 + a_1\omega + a_2\omega^2 + a_3\omega^3 + \ldots$

-R and a_1: the focal distance r_1 and focal curve inclination γ (analytically) $\Rightarrow$ inclination reduces the effective detector pixel size

- a_2: profile asymmetry (coma) cancellation (numerically) – bug in SHADOW fixed in 2010!

- a_3: reduction of symmetric broadening (numerically) $\Rightarrow$ *increase of aberration-free vertical acceptance by a factor of 5*



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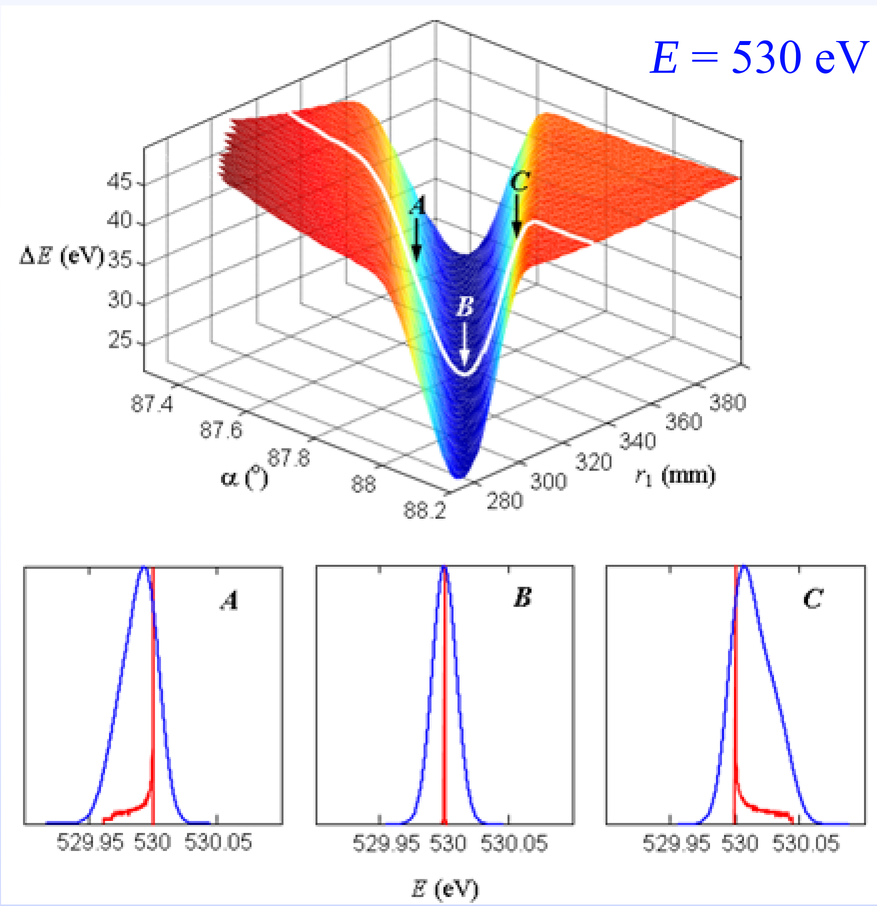
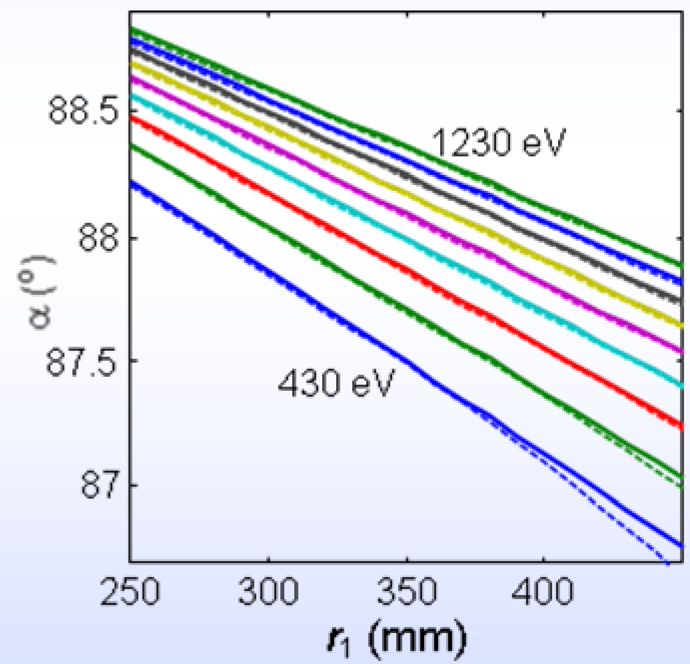
Design of spherical VLS grating spectrometers

Step 2: Optimization of the spectrometer geometry away from reference *E*

- How do we adjust r_1, α, r_2 to keep symmetric profile and thus best resolution?

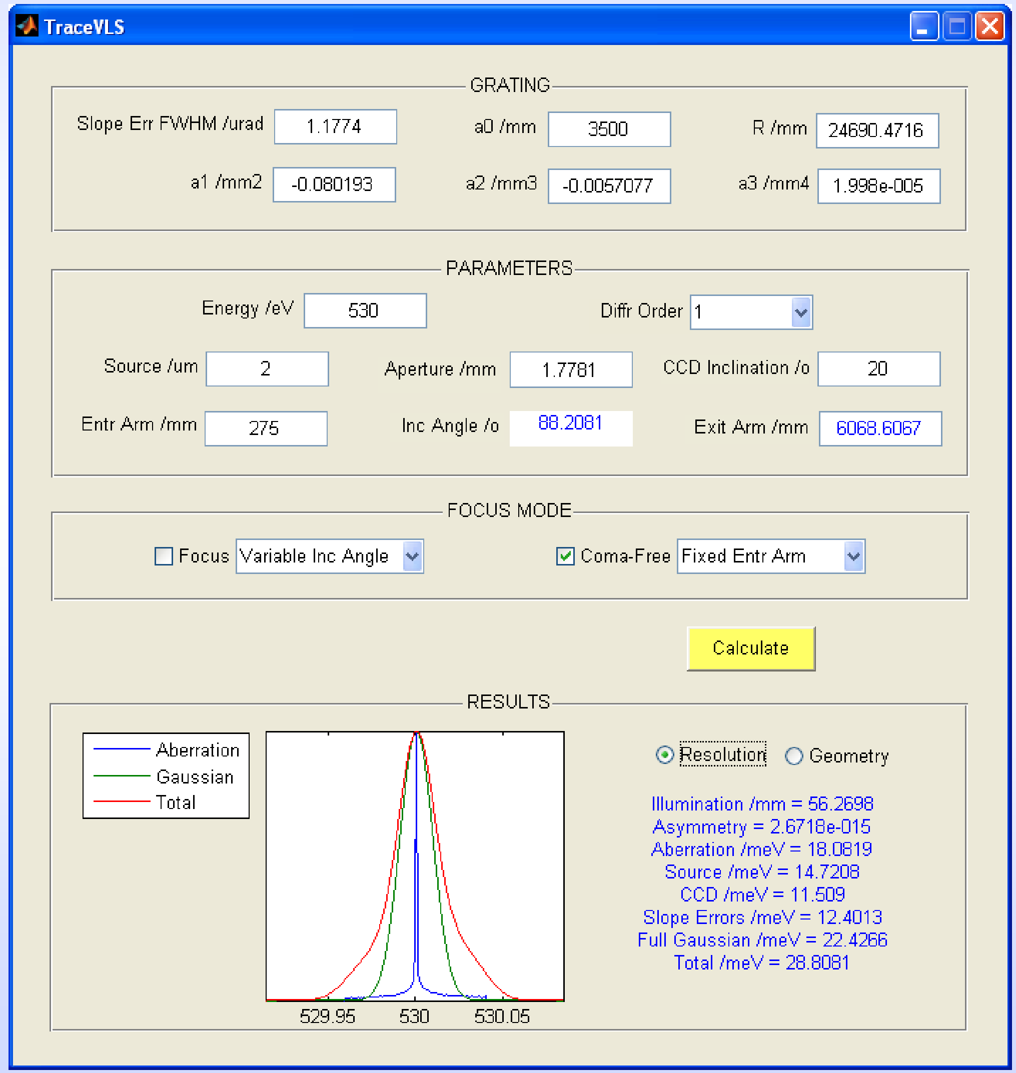




- symmetric profile can be maintained for *any energy* by adjustment of r_1 *or* α

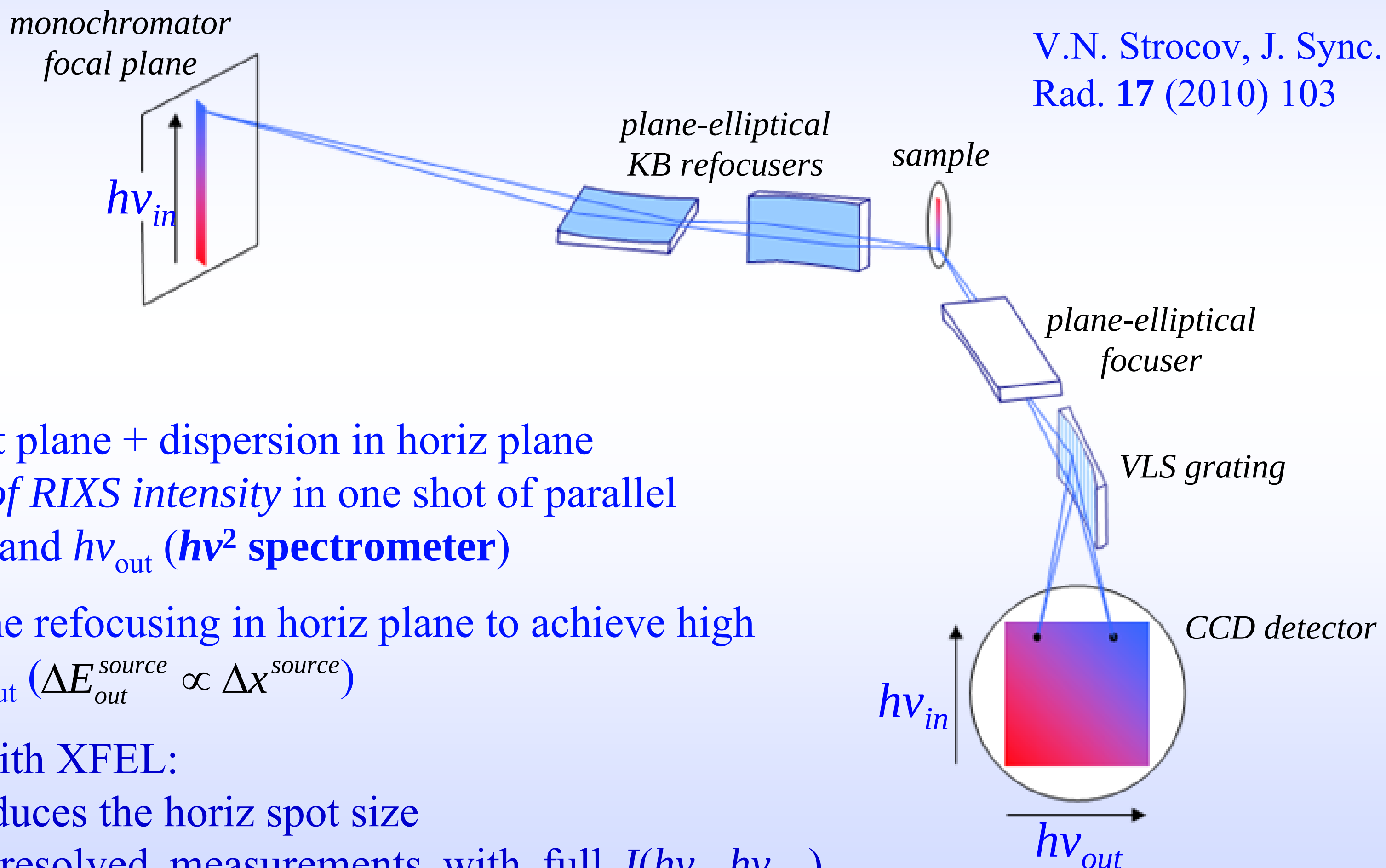
Online software to determine the optimal spectrometer settings

- the focal and symmetric-profile focal α, r_1 and r_2 in a fraction of second



Perspectives of RIXS instrumentation:
$h\nu^2$-spectrometer with simultaneous detection in $h\nu_{in}$ and $h\nu_{out}$

V.N. Strocov, J. Sync. Rad. **17** (2010) 103

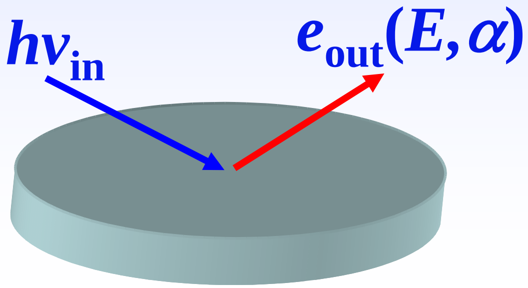


- imaging in vert plane + dispersion in horiz plane
$\Rightarrow$ *full 2D-map of RIXS intensity* in one shot of parallel detection in $h\nu_{in}$ and $h\nu_{out}$ (**$h\nu^2$ spectrometer**)

- critical: extreme refocusing in horiz plane to achieve high resolution in $h\nu_{out}$ ($\Delta E_{out}^{source} \propto \Delta x^{source}$)

- combination with XFEL:
- *round beam* reduces the horiz spot size
- efficient time-resolved measurements with full $I(h\nu_{in},h\nu_{out})$ snapshot in *one instant of time*: crucial for chemical reactions

ARPES endstation: Concept



• hole spectral function $A(E,\mathbf{k})$ resolved in E and $\mathbf{k}$

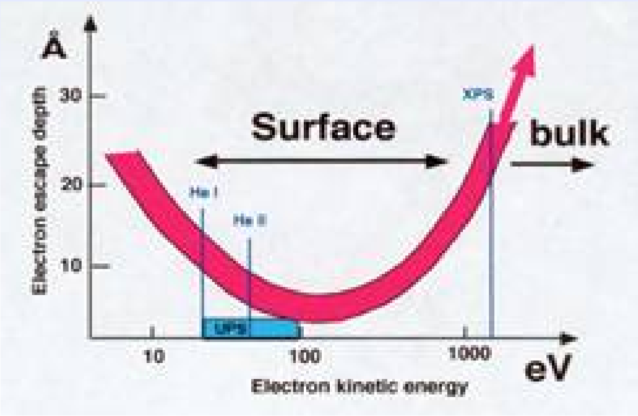
• soft X-rays vs hard X-rays to keep angular resolution

• combining with PLD

• electronic structure of complex materials (perovskites…) with enhanced bulk sensitivity and resolution in 3-dim **k**-space

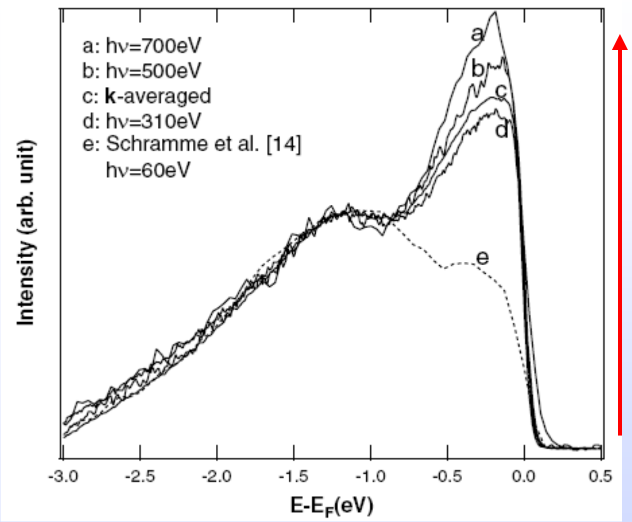
Why going from UV to Soft-X-Rays ?

Reason 1: Surface sensitivity



• 2-3 times increase in probing depth
⇒ through the distorted surface layer towards deeper atomic layers with bulk properties

Mott-Hubbard metal-insulator transition in V_2O_3
(Mo et al 2003)



bulk sensitivity increases with *hν*

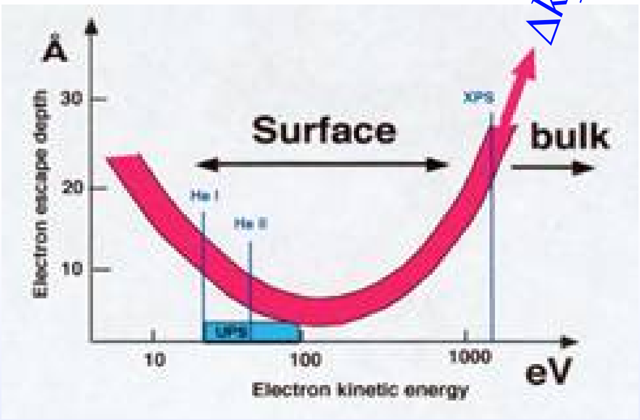
• quasiparticle peak in the paramagnetic phase develops only in bulk

⇒ **soft-X-ray energy range** to increase bulk sensitivity

Reason 2: Improvement of the intrinsic resolution in $k_\perp$

• photoelectron wavefunction confined by $\lambda \Rightarrow$ broadening $\Delta k_\perp = \lambda^{-1}$ ← **intrinsic $k_\perp$ resolution**

$\Delta k_\perp$ improvement



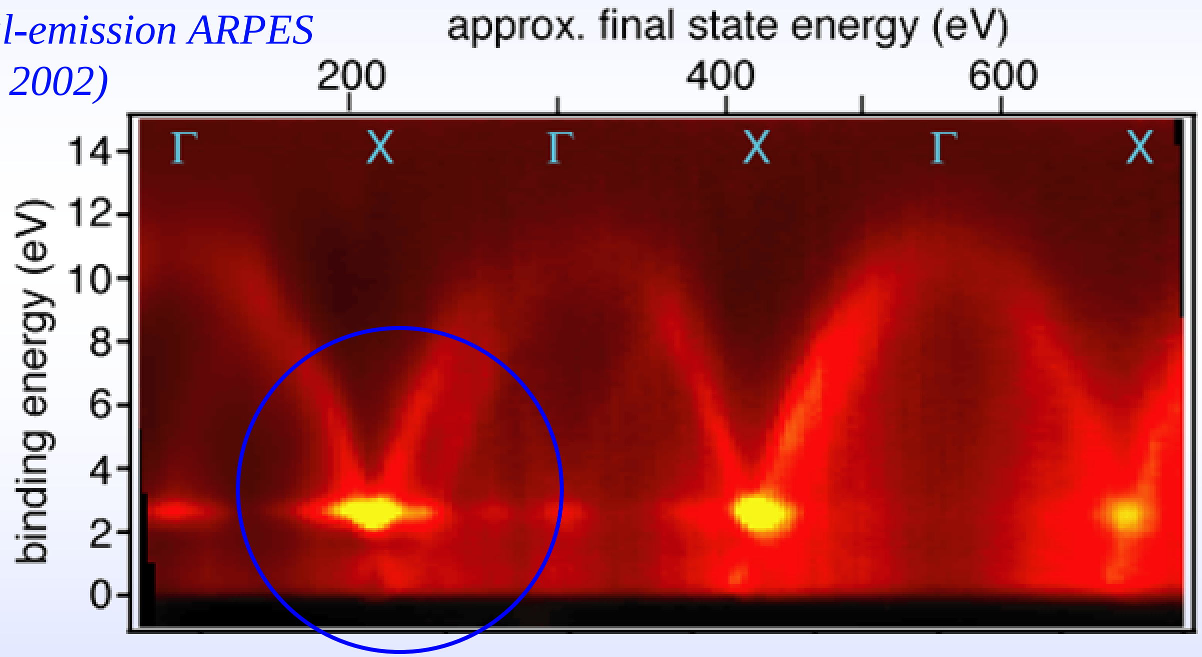
$\Rightarrow$ **soft-X-ray energy range** to increase the resolution in $k_\perp$

Reason 3: Free-electron final states

- Final-state $E(\mathbf{k})$ is required to resolve valence band $E(\mathbf{k})$ in 3-dimensional **k**

- How far in energy do the non-free-electron effects carry on?

Al(100) normal-emission ARPES (Hoffman et al 2002)



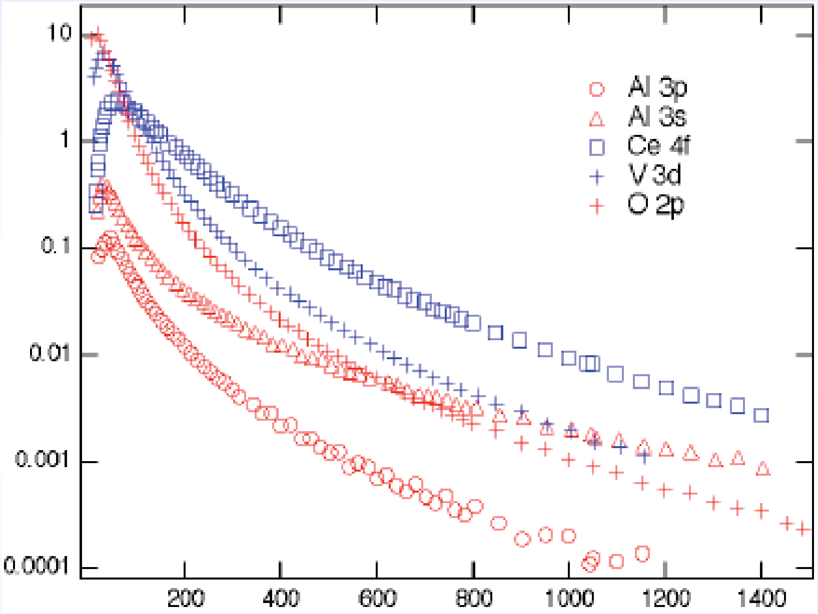
multiband final states (different $k_\perp$)

• failure of free-electron approximation despite the FE nature of Al and rather high *hv*
⇒ **soft-X-ray energy range** for free-electron final states

• Further reasons: Simplified matrix elements …

Problem: Photoexcitation crossection

• notorious problem of SX-ARPES: dramatic decrease of crossection, especially for *s*- and *p*-states



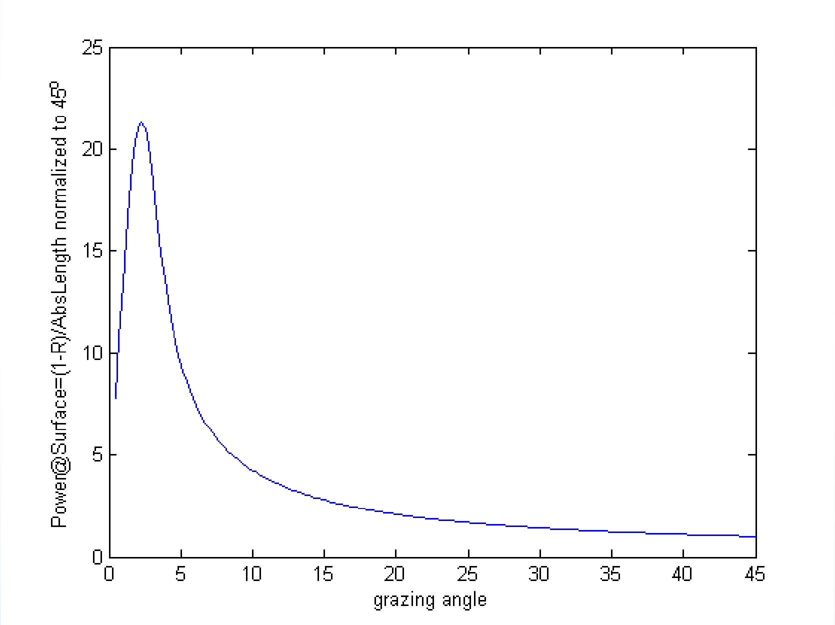
• the crossection problem is alleviated by 10 to 100 flux increase vs BL25SU @ Spring-8

Implementation of the SX-ARPES endstation

Experimental geometry concepts: Optimal light incidence angle

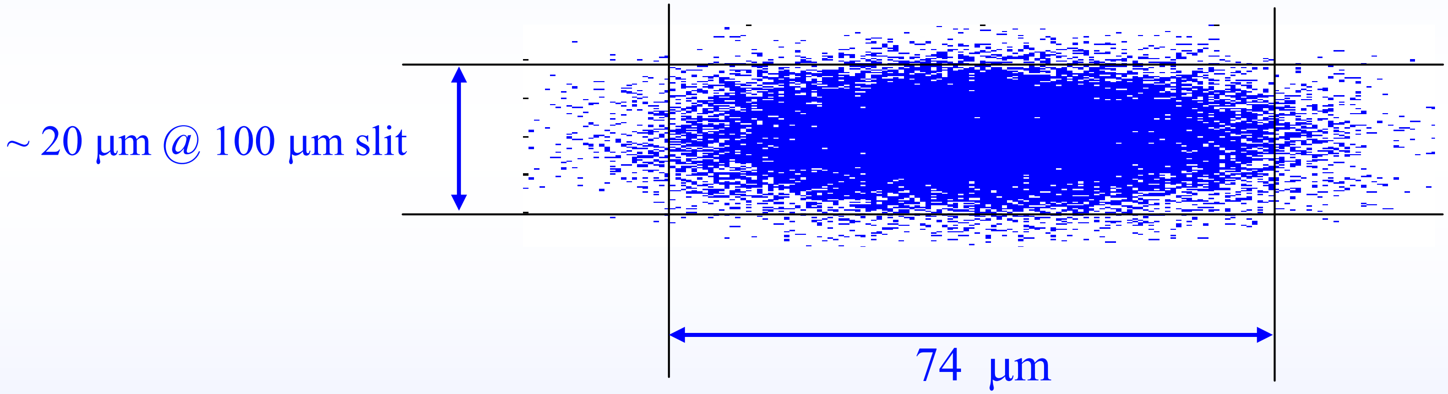
Photoelectron Yield $\propto (1-R)/\lambda_{hv}$

$(\lambda_e \ll \lambda_{hv})$



- photoelectron yield peak at glancing angles ~2.5°
- improvement of 2.1 @ 20° compared to standard 45°

Experimental geometry concepts: Alignment of the light footprint



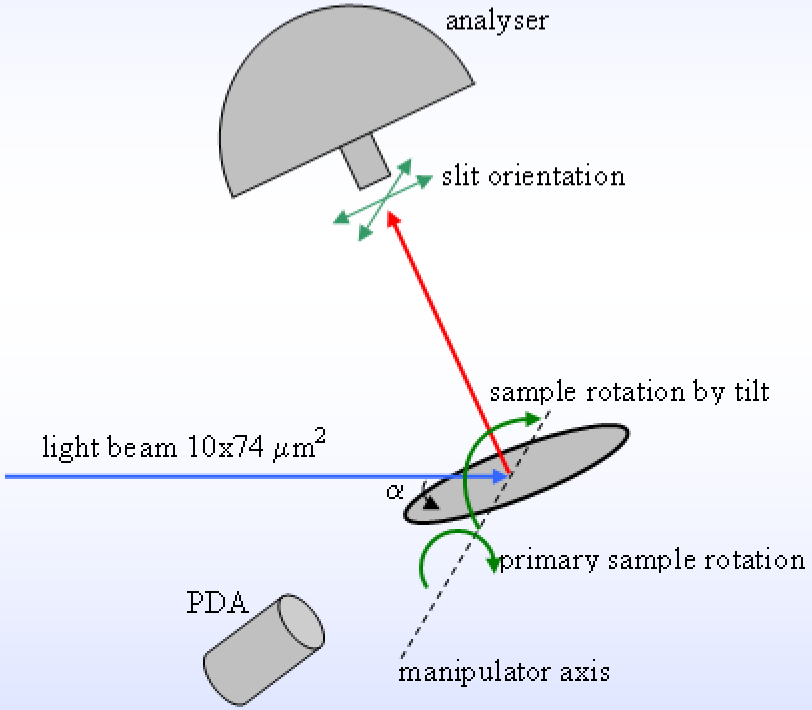
- *rotation around the horizontal axis* to align the horizontal and vertical spot size
- 100 µm slit => grazing incidence angle ~ 13.5°

Experimental geometry

• Grazing incidence at 20° // smaller vertical footprint with *horizontal* manipulator axis

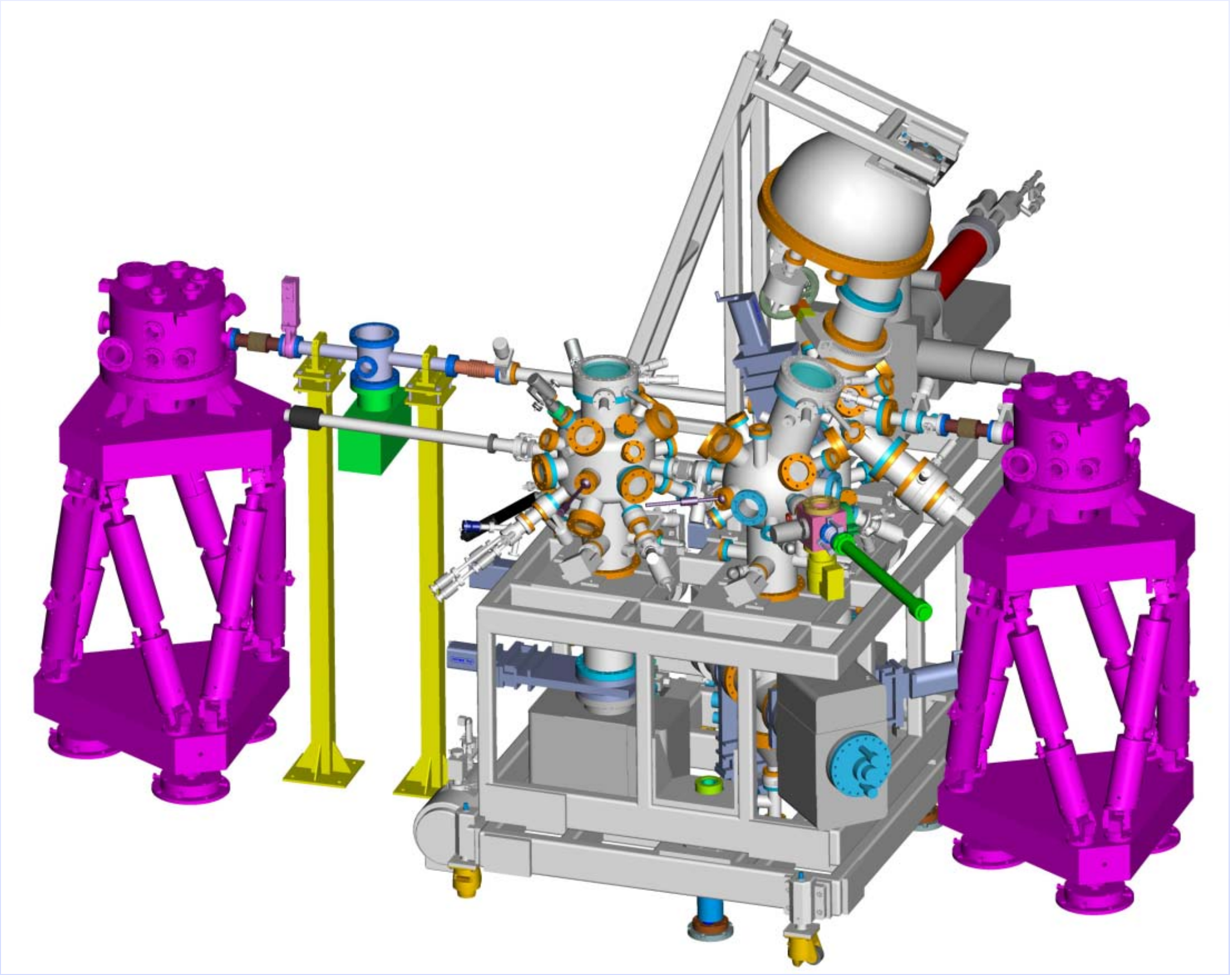
• 2 operation modes:
- analyser slit // beam (selection rules)
- analyser slit ⊥ beam (**k**-space sampling)

• Photoelectron Display Analyser (PDA) ~ photon-excitation LEED

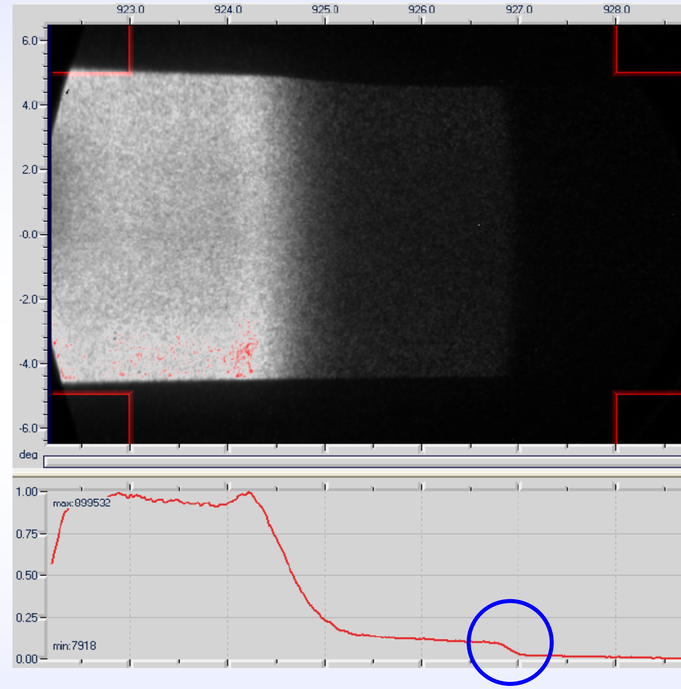


Technical realization

- analyzer PHOIBIOS 150 (SPECS)
- manipulator with 3 translation (resolution 5μm) + 3 angular (resolution 0.1°) DOFs and L-He_2 cooling to 10K
- analysis (AC) + transfer (TC) + preparation (PC) chambers + Load Lock (LL)
- sample preparation by cleavage, ion etching, thin film deposition
- only *one* sample transfer for cleaved samples
- compatibility with PLD



Status



E_F of Au @ $h\nu$ = 930 eV

- 10.5 K achieved
- *30 sec* data acquisition @ $h\nu$=930 eV, combined ΔE=100 meV
- Expert user operation from the end 2010

Summary

High-resolution soft-X-ray ADRESS beamline operating in the energy range 300 - 1800 eV:

• Fixed-gap undulator
- circular and 0-180° variable linear polarizations

• Collimated-light PGM with stigmatic focus
- ΔE~30 meV@1 keV
- flux up to 10^{13} photons/s/0.01%BW with optimized gratings (minimal l/mm, blazed/lamellar, optimized profiles, flux-optimal C_{ff})

• Ellipsoidal refocusing optics
- spot size below 4 μm

• RIXS spectrometer
- ΔE~70 meV@1 keV (energy scale of magnetic etc. excitations)
- variable scattering angle (momentum dependences)
- high-resolution RIXS complementary to INS
- further developments to optimize the acceptance and resolution

• ARPES spectrometer
- optimized experimental geometry (grazing light incidence, horizontal manipulator axis)
- rotatable analyser (selection rules vs **k**-space sampling)

People

Beamline scientists

V.N. Strocov (ARPES) and T. Schmitt (RIXS)

Beamline stuff

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U. Flechsig *Optics*

T. Schmidt, A. Imhof, B. Jakob *ID*

Q. Chen *Front End*

R. Betemps *Mechanics*

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