Spectroscopy on strongly correlated electron systems

The collaboration between P01 and MPI CPfS Dresden

Dr. Martin Sundermann
Dr. Hlynur Gretarsson
Dr. Ilya Sergeev
Frank-Uwe Dill
Simon Mayer
Klas Pikul
Dr. Christoph Schlüter
Dr. Andrei Hloskovsky

DESY. Petra III Hamburg

Prof. Liu Hao Tjeng
Andrea Marino
Denise Christovam
Paulius Dolmantas
Dr. Chun-Fu Chang
Dr. Andrea Amorese
Dr. Brett Leedahl
Christoph Becker
Katharina Höfer
Dr. Berit Goodge
Edgar Abarca Morales

MPI CPfS Dresden

Dr. Andrea Severing

Uni Köln

Prof. Jochen Geck
Dr. Christopher Marein Rahn

TU Dresden

Prof. Bernhard Keimer
Vignesh Sundaramurthy

MPI FKF Stuttgart

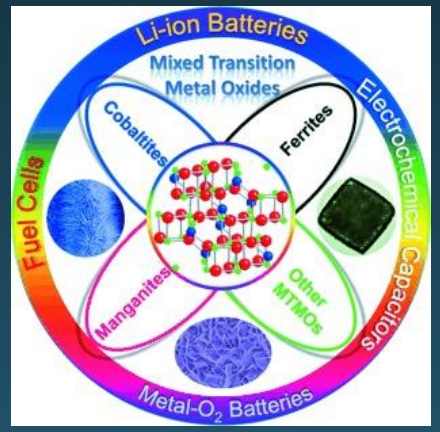
Prof. Maurits W. Haverkort

Uni Heidelberg

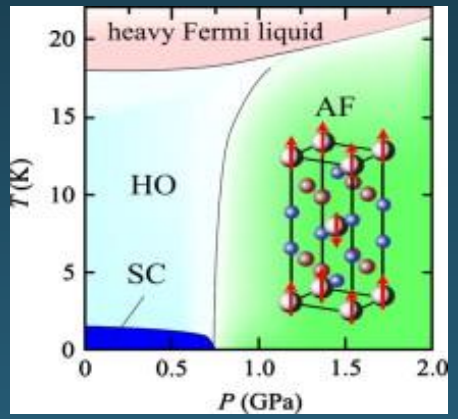
Spectroscopy on strongly correlated electron systems

Outline

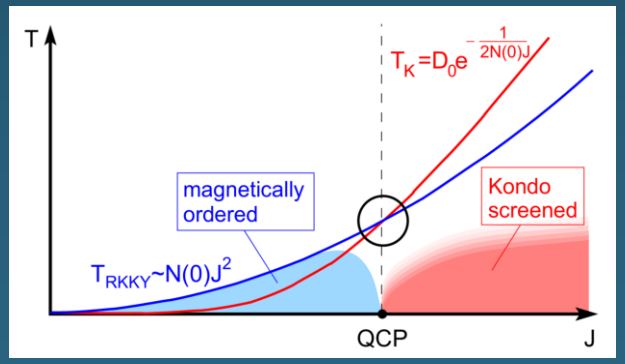
Materials



TM oxides



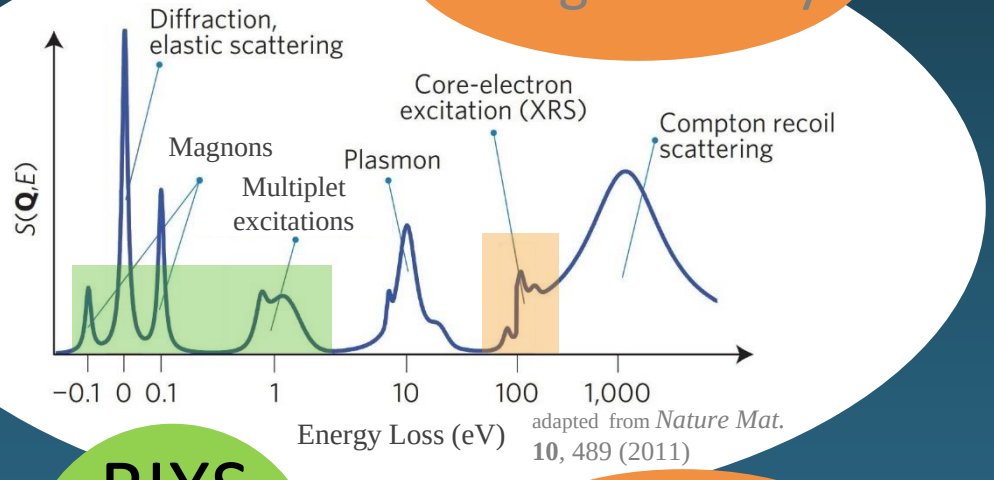
U intermetallics



RE compounds

Methods

NIXS
charge density



adapted from *Nature Mat.* **10**, 489 (2011)

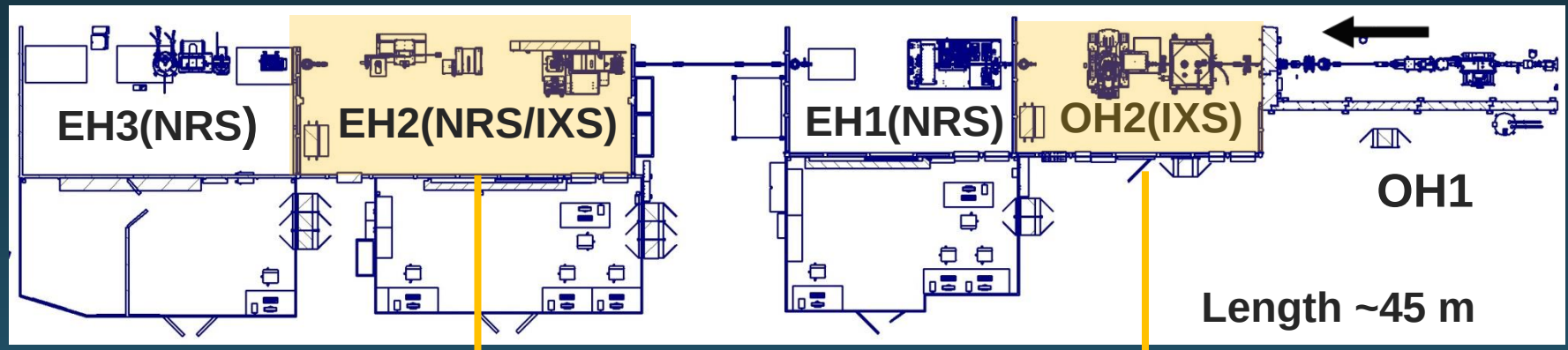
RIXS
low energy excitations

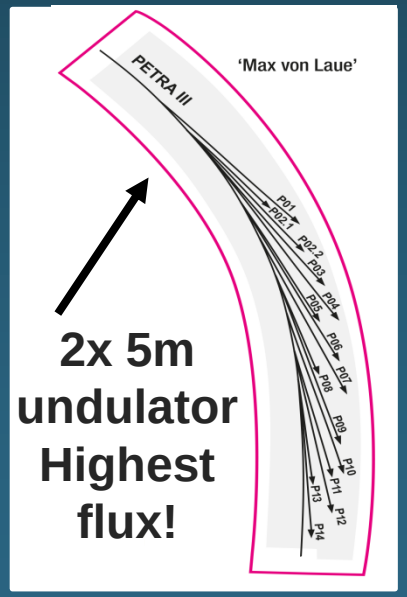
HAXPES
valence

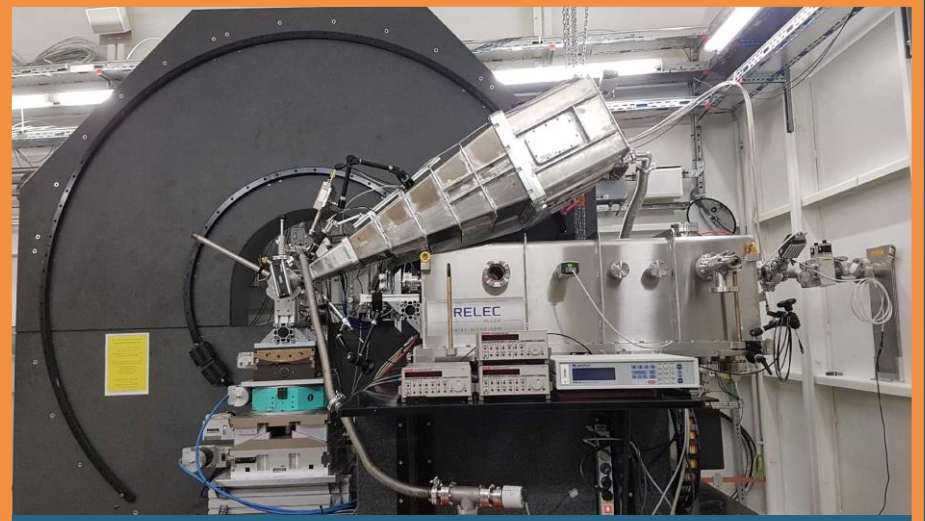
Spectroscopy on strongly correlated electron systems

P01 IXS end stations

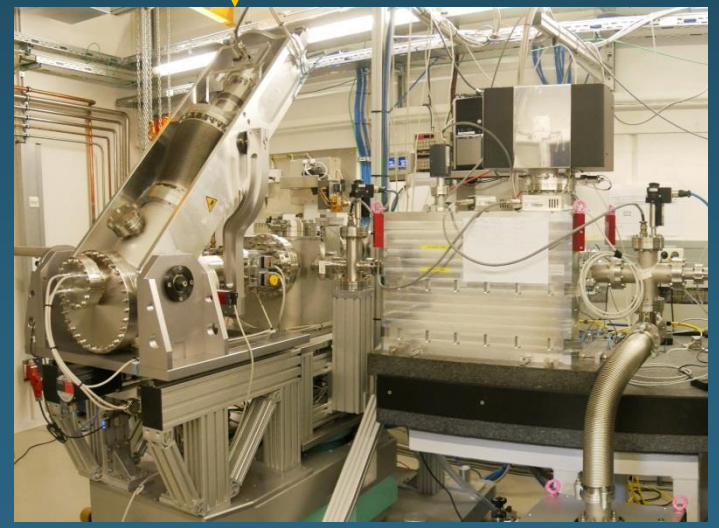
Beamtime share:
50% NRS
20% IXS MPI CPfS
20% IXS MPI FKF
10% IXS Proposals







NIXS – MPI CPfS



RIXS – MPI FKF

Spectroscopy on strongly correlated electron systems

Non-resonant Inelastic X-ray Scattering (NIXS)

Soft x-ray absorption spectroscopy
BUT: Using hard x-rays

- 700meV FWHM at 10keV

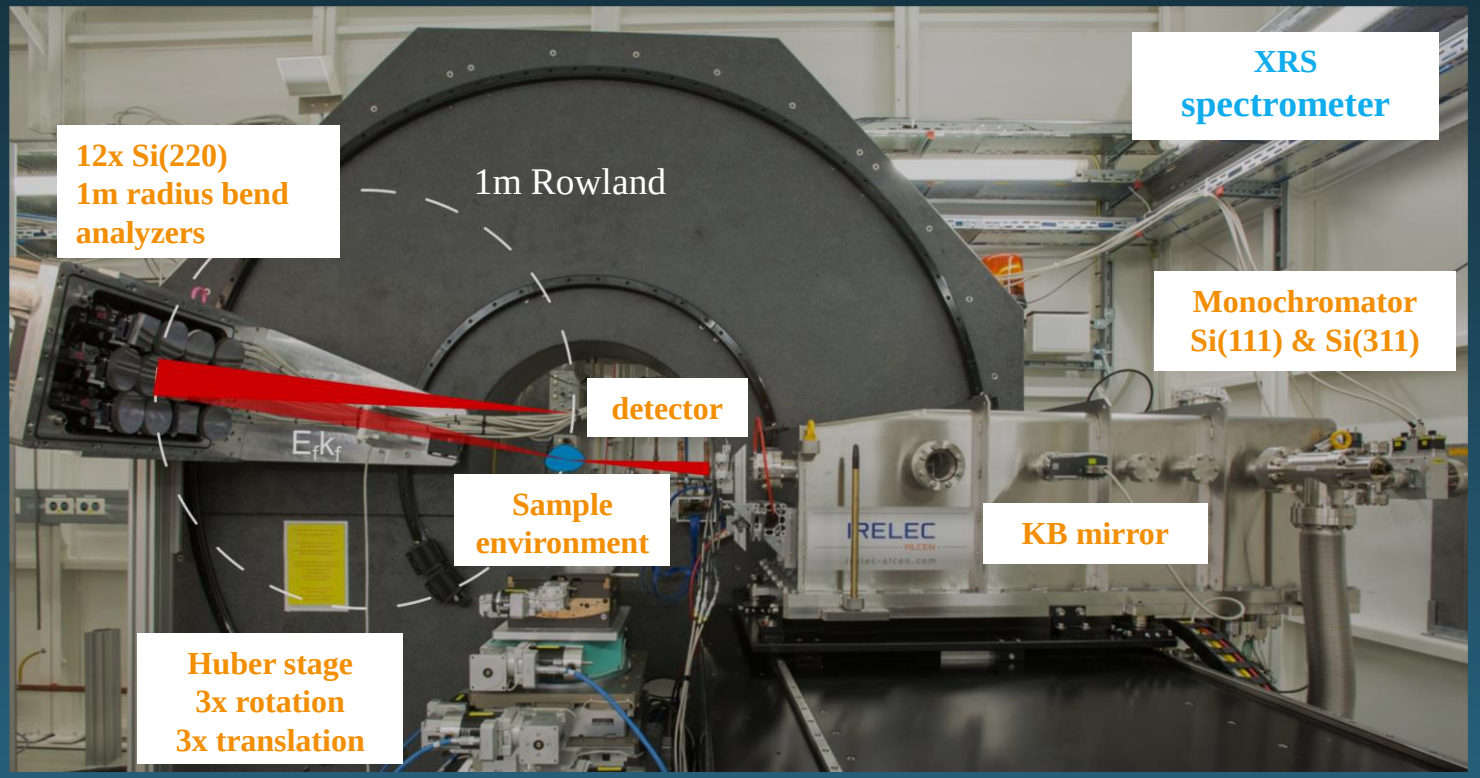
Bulk sensitive

- Complex environments
- In-situ experiments
- No cleaving

Beyond dipole

- New excitonic states
- Dichroism beyond 2-fold

Photon hungry



Medipix3 detector (x-spectrum)

Photon counting 2D-detector

Dynaflow cryostat (ESRF)

T down to 3K

360° rotation

Oven – custom design

T up to 850K

Spectroscopy on strongly correlated electron systems

Side remark: NIXS for chemistry or geoscience

Soft x-ray absorption spectroscopy

BUT: Using hard x-rays

- 700meV FWHM at 10keV

Bulk sensitive

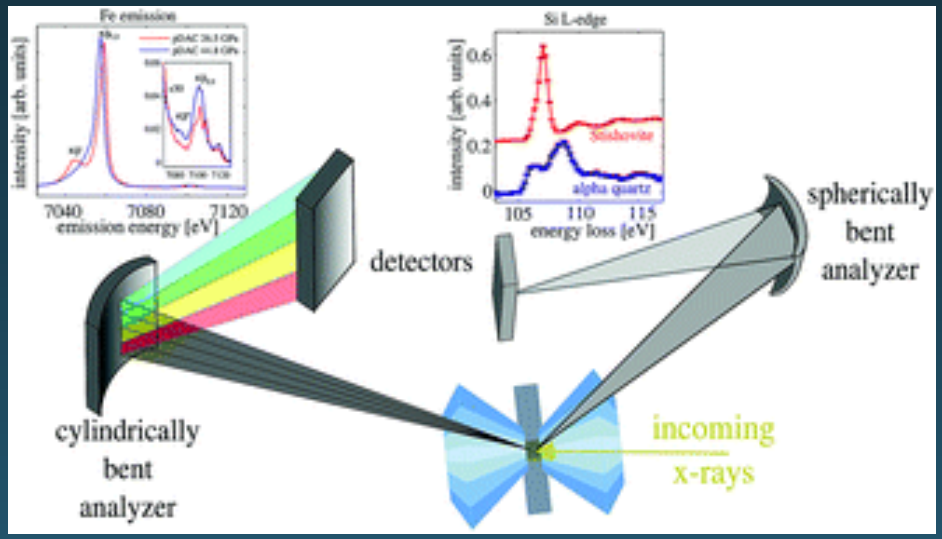
- Complex environments
- In-situ experiments
- No cleaving

Beyond dipole

- New excitonic states
- Dichroism beyond 2-fold

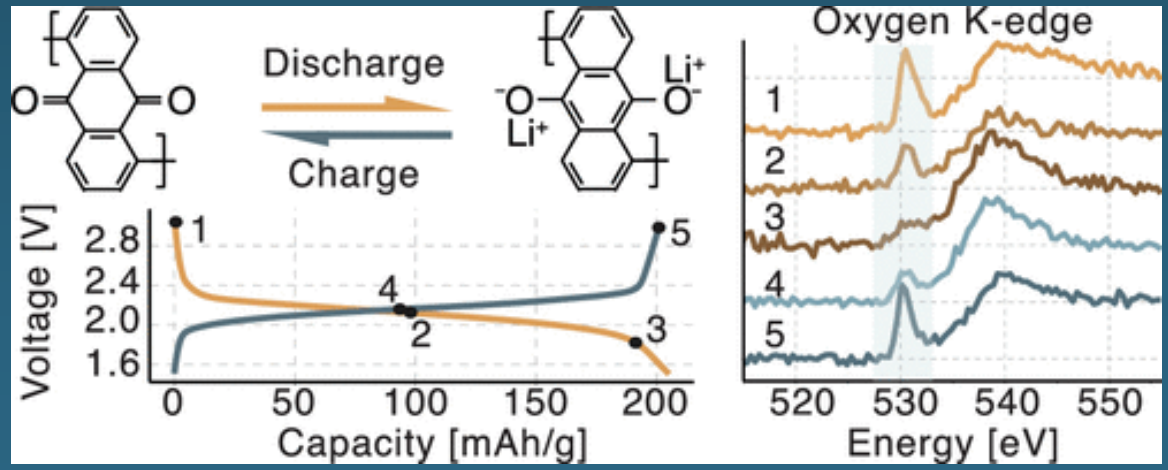
Photon hungry

Geoscience:
Extreme conditions



C. Weis, *et al.*, **J. Anal. At. Spectrom. 34**, 384 (2019)

Batteries:
In-situ / in-operando



Ava Rajh, *et al.*, **J. Phys. Chem C 126**, 5435 (2022)

Spectroscopy on strongly correlated electron systems

NIXS: Unique insights to matter (part 1)

Soft x-ray absorption spectroscopy

BUT: Using hard x-rays

- 700meV FWHM at 10keV

Bulk sensitive

- Complex environments
- In-situ experiments
- No cleaving

Beyond dipole

- New excitonic states
- Dichroism beyond 2-fold

Photon hungry

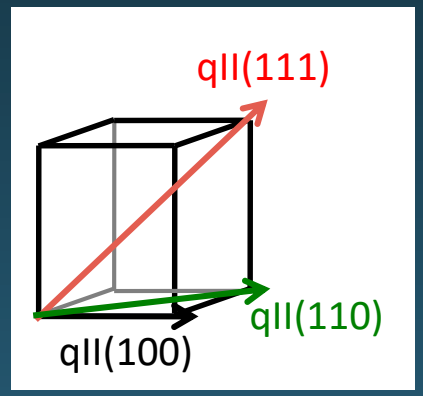
Exploit the transitions not accessible by soft XAS
→ Unique new probe of matter

Spectroscopy on strongly correlated electron systems

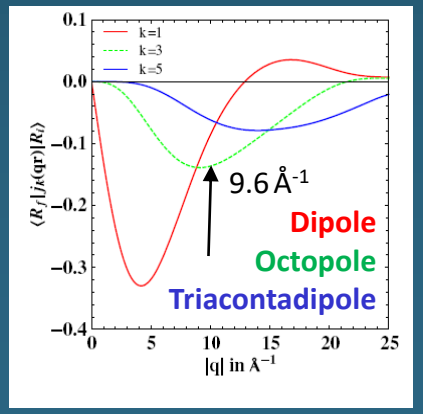
NIXS on rare earth (RE) intermetallics: 4*f* crystal-field ground state

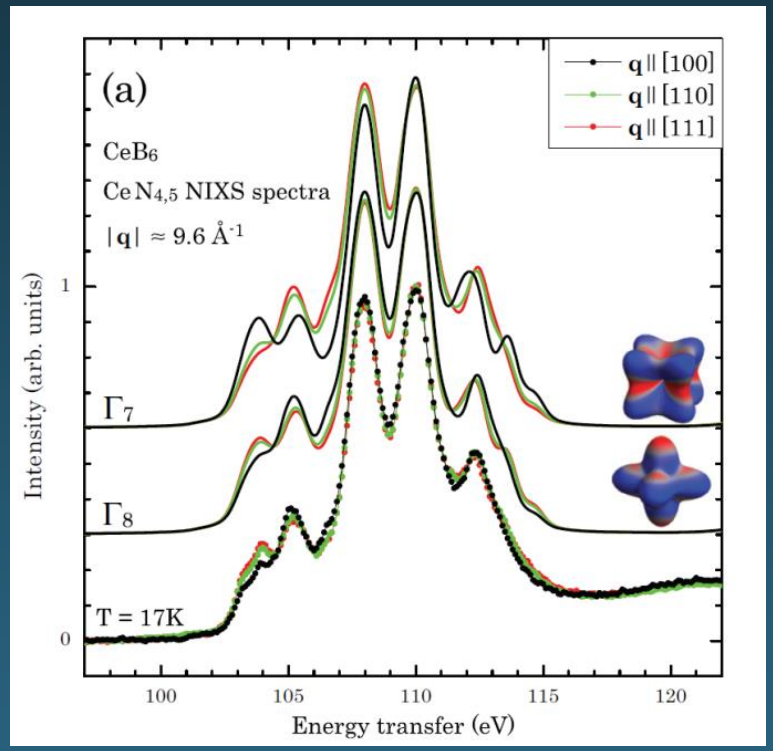
Question: ground state of Ce^{3+} or Sm^{3+}: Γ_8 or Γ_7?

Dichroism in cubic systems



possible trough
beyond-dipole transitions

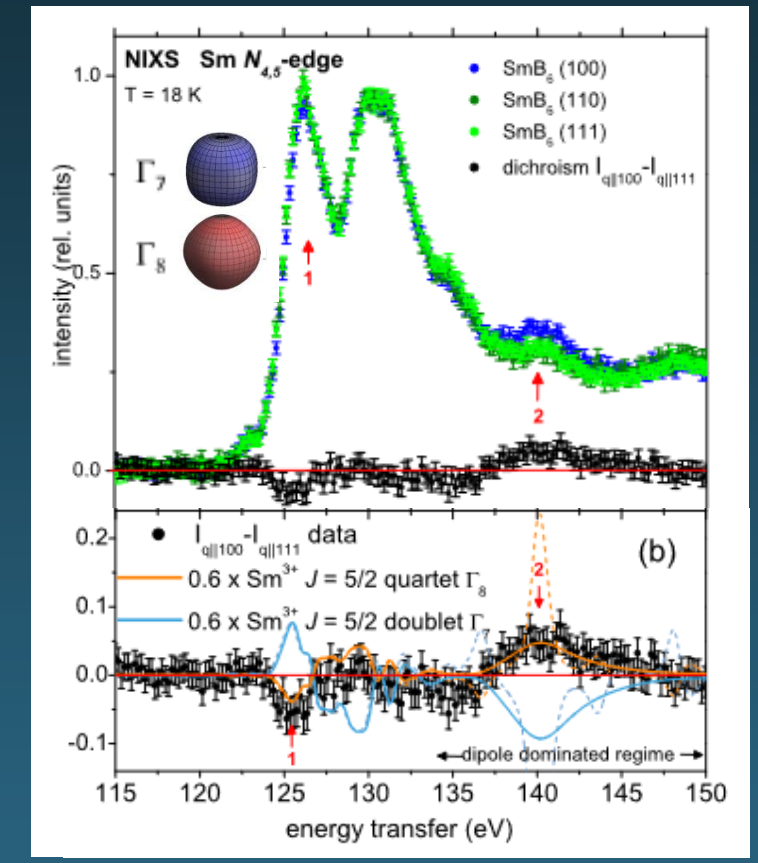




CeB_6: multipolar order compound

We confirm the Γ_8 ground state!!

EPL 117, 17003 (2017)



SmB_6: Kondo insulator and putative strongly correlated topological insulator.
$Sm^{2.6+}$: mixture of Sm^{3+} and Sm^{2+}
only Sm^{3+} contributes to dichroism
PRL 120, 016402 (2018)

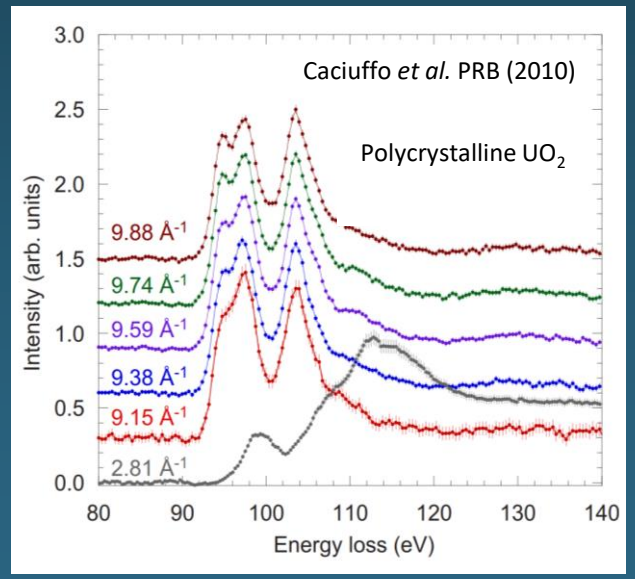
Spectroscopy on strongly correlated electron systems

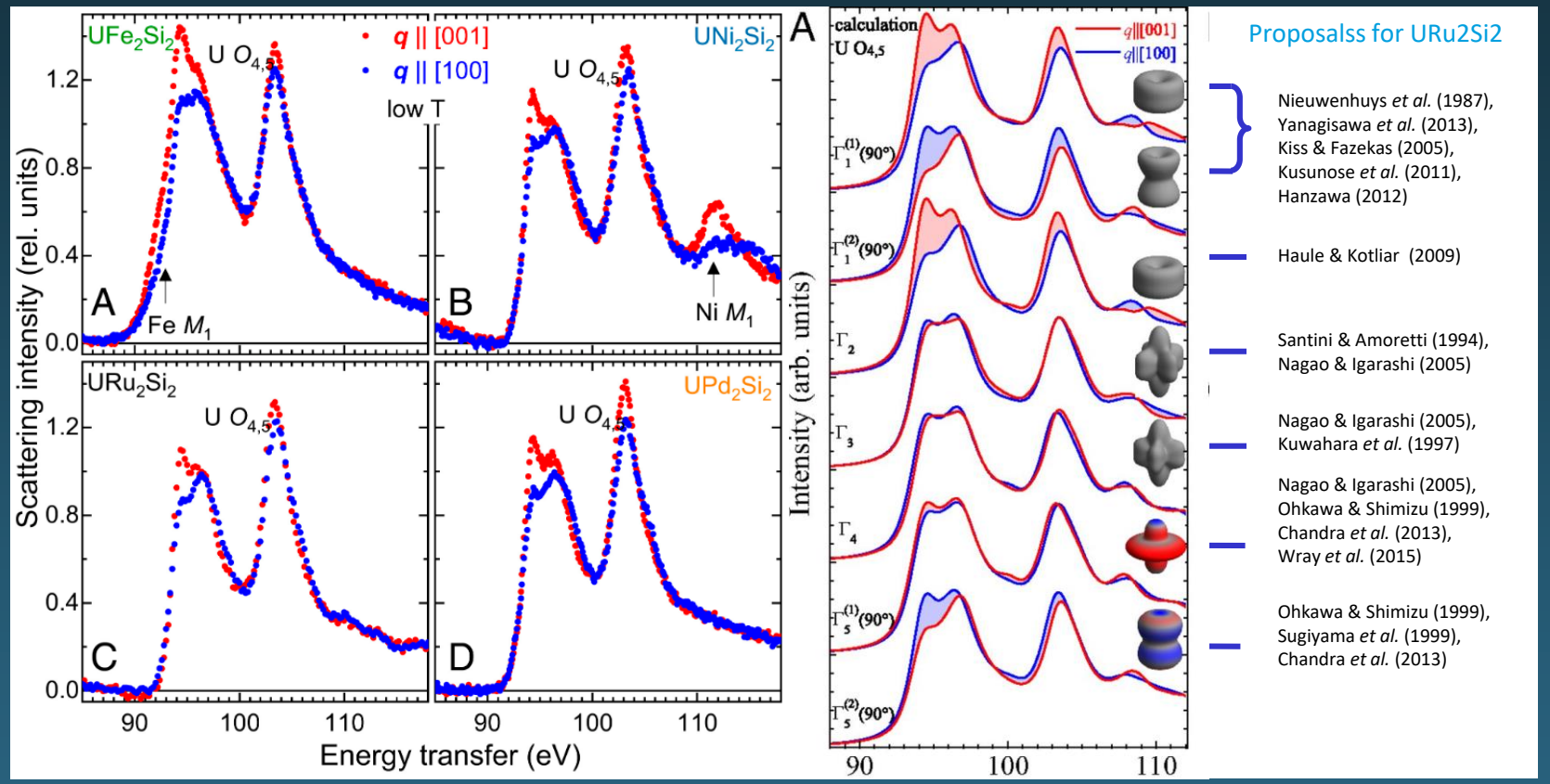
NIXS on Uranium: Excitonic resonances in intermetallic compounds

Question: CF ground state?

Beyond-dipole transitions give access to excitonic multiplets!

No cleaving required!





UM_2Si_2: From hidden order (HO) to magnetism

Same symmetry forms HO and magnetism!!

A. Amorese *et al*, **PNAS 117**, 30220 (2020)

URu_2Si_2: Hidden order compound

Restrict scenarios of the ordering!!

PNAS 113, 13989 (2016)

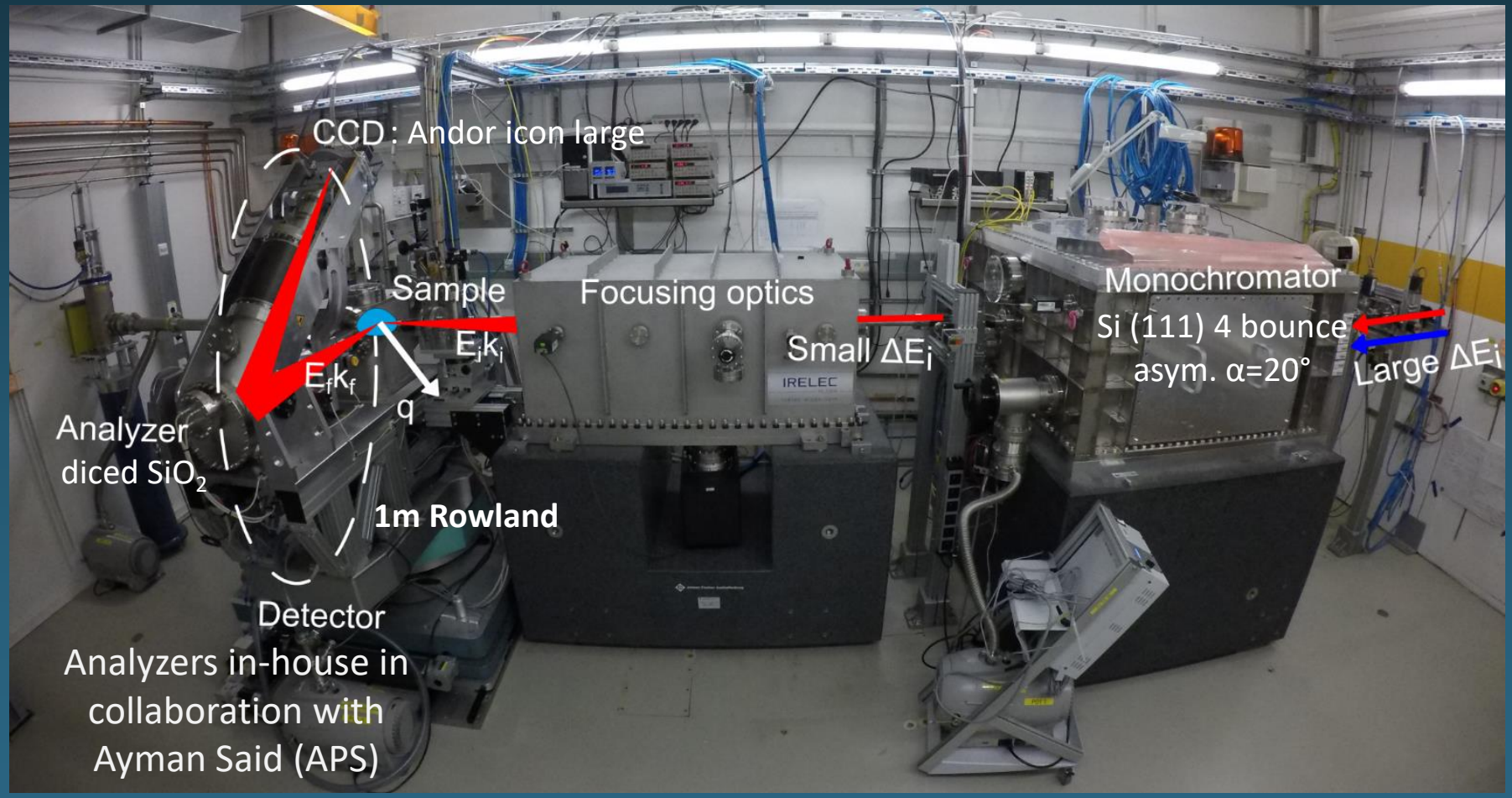
Spectroscopy on strongly correlated electron systems

Tender Resonant Inelastic X-ray Scattering (RIXS)

MPI FKF Stuttgart
Prof. Bernhard Keimer
Build for RIXS at Ru L_3

Edge	Energy (eV)	FWHM (meV)
Ru L_3	2840	75
Rh L_3	3005	90
Rh L_2	3146	150
U M_5	**3550**	**70**
U M_4	**3725**	**150**

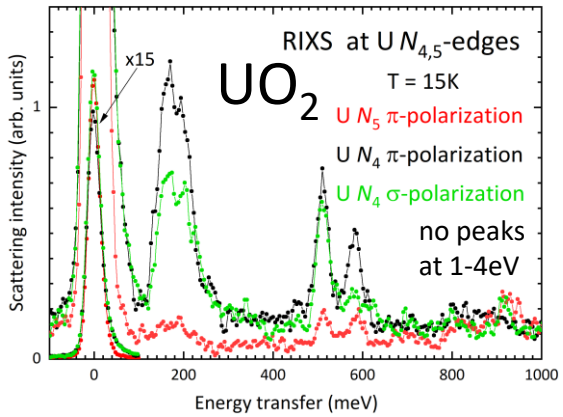
Upcoming: S K-edge 2475 eV
T. Liu et al., Energy Storage Materials (2020)



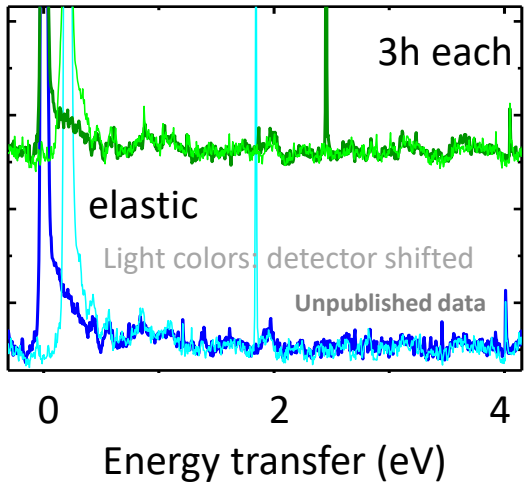
Spectroscopy on strongly correlated electron systems

RIXS at the U $M_{4,5}$ edges: Low energy excitations

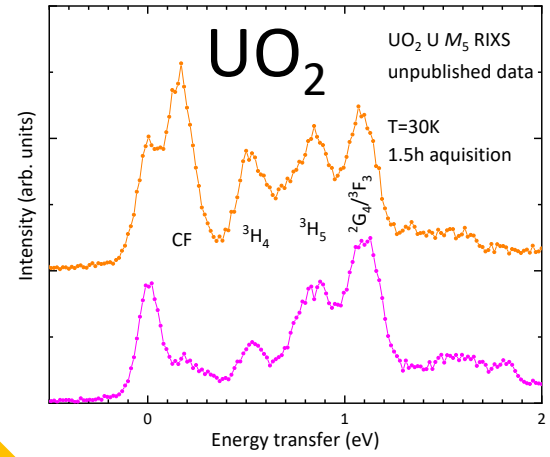
RIXS Uranium N_4 edge

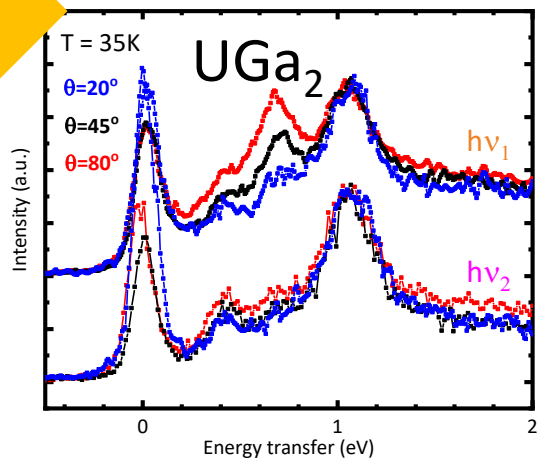


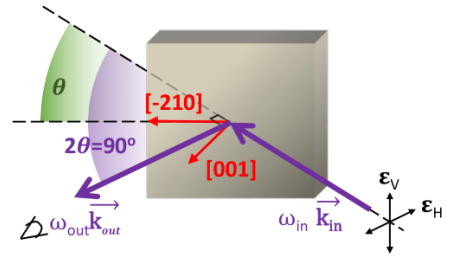
G.H. Lander *et al.:* J. Phys.: Condens. Matter 33 (2021) 06LT01

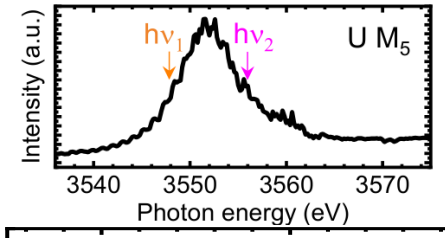


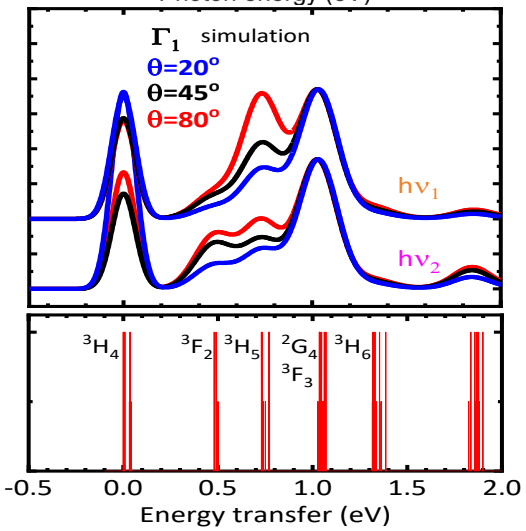
soft – tender











A. Marino *et al*, **PRB 108**, 045142 (2023)

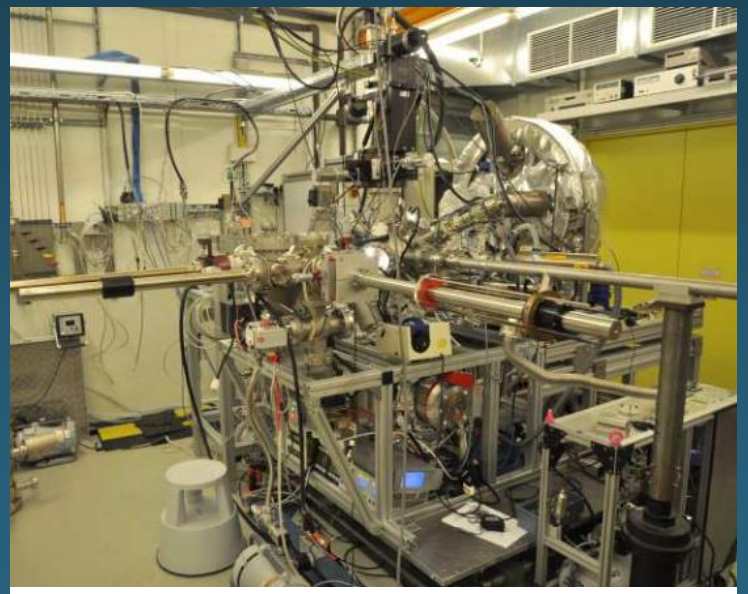
Spectroscopy on strongly correlated electron systems

P22: Hard X-ray Photoelectron spectroscopy (HAXPES) on *f*-electron systems

Question: valence of RE / U?

P22 via proposal

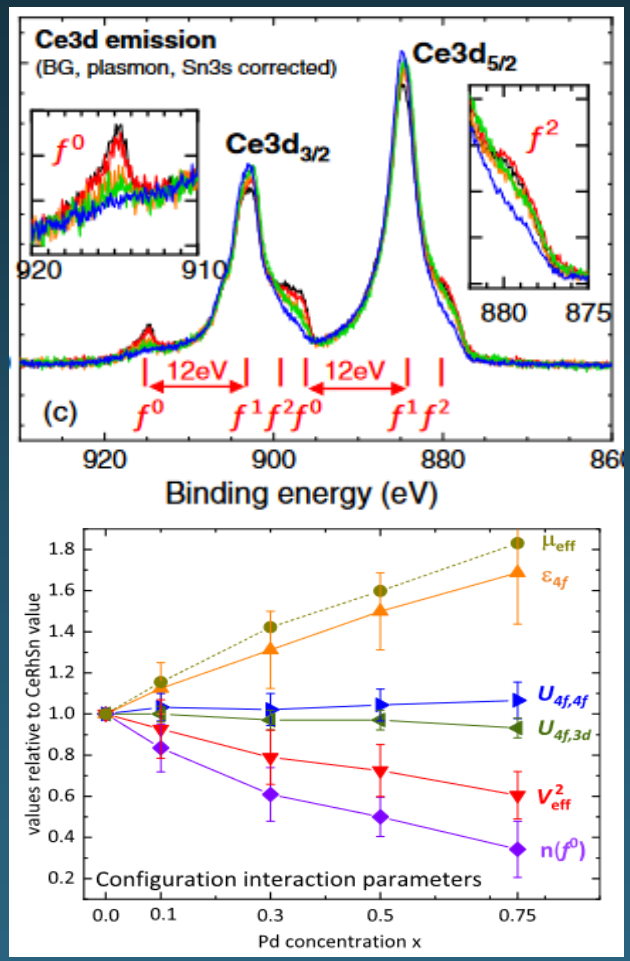
dedicated UHV chamber for U cleaving



The new dedicated HAXPES beamline P22 at PETRAIII

Cite as: AIP Conference Proceedings **2054**, 040010 (2019); https://doi.org/10.1063/1.5084611
Published Online: 16 January 2019

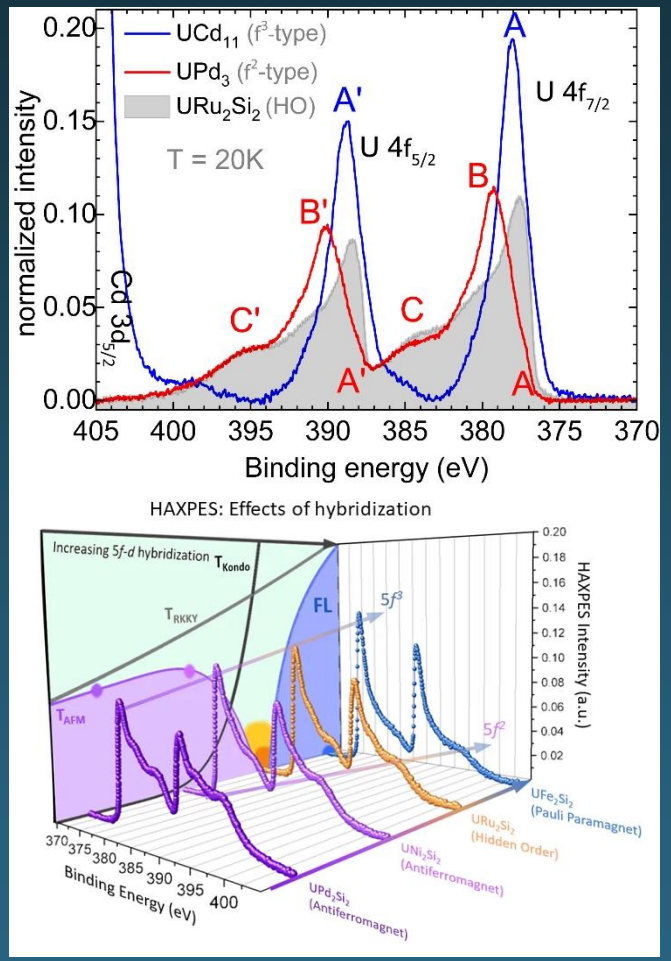
C. Schlueter, A. Gloskovskii, K. Ederer, I. Schostak, S. Piec, I. Sarkar, Yu. Matveyev, P. Lömker, M. Sing, R. Claessen, C. Wiemann, C. M. Schneider, K. Medjanik, G. Schönhense, P. Amann, A. Nilsson, and W. Drube



Intermediate valence in rare earth intermetallics

Gunnarson Schönhammer model yields CI parameters!!

PRB 104, 235150 (2021)



Uranium intermetallics

Valence change across the phase diagram!!

A. Amorese *et al*, **PNAS 117**, 30220 (2020)

Spectroscopy on strongly correlated electron systems

NIXS: Unique insights to matter (part 2)

Soft x-ray absorption spectroscopy

BUT: Using hard x-rays

- 700meV FWHM at 10keV

Bulk sensitive

- Complex environments
- In-situ experiments
- No cleaving

Beyond dipole

- New excitonic states
- Dichroism beyond 2-fold

Photon hungry

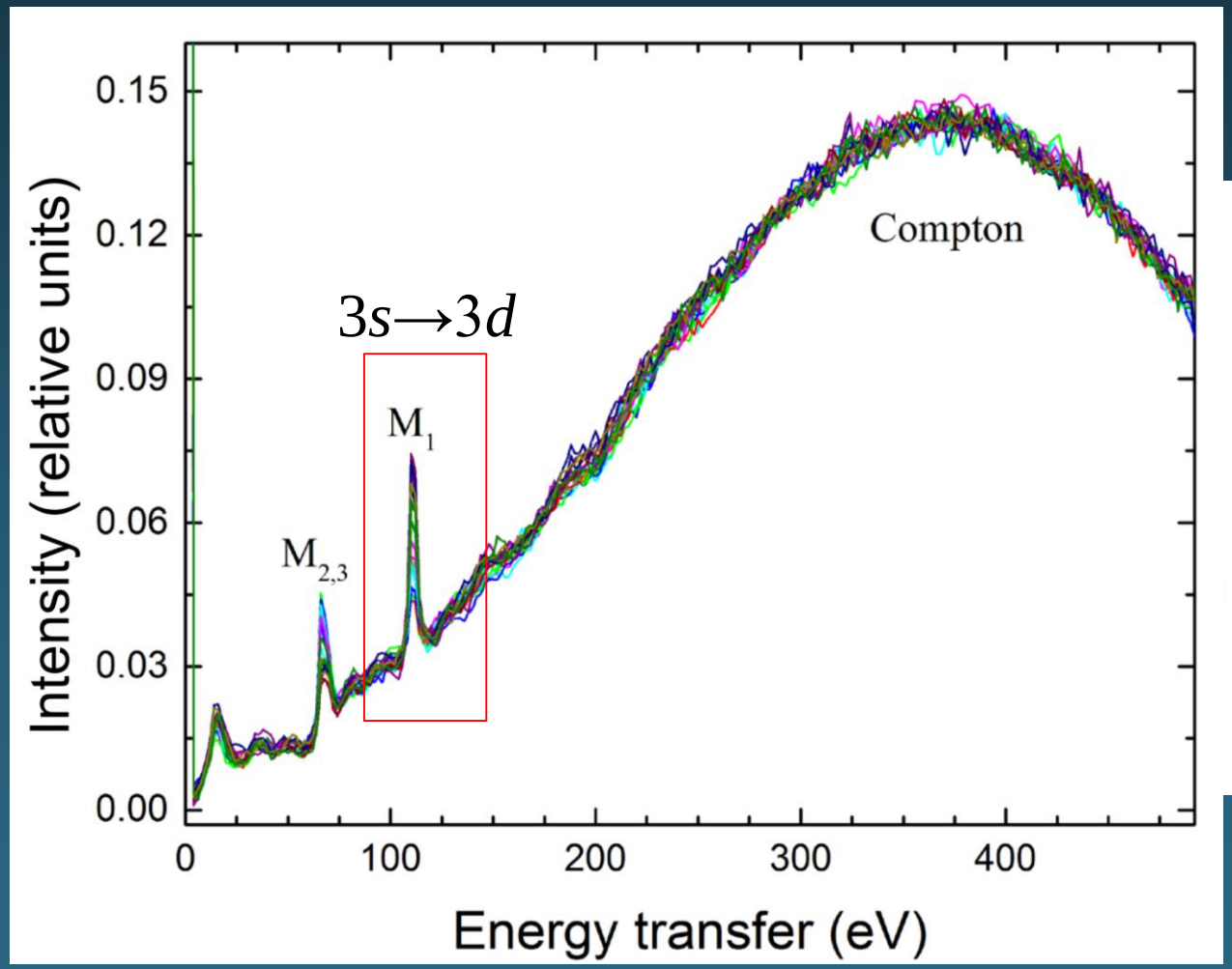
Exploit the transitions not accessible by soft XAS

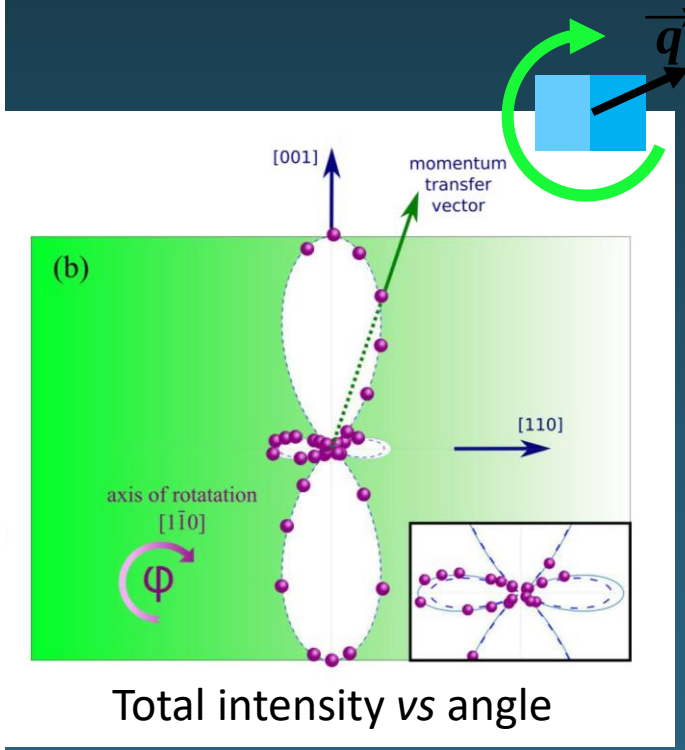
→ Unique new view on matter

Now: Direct imaging of orbitals

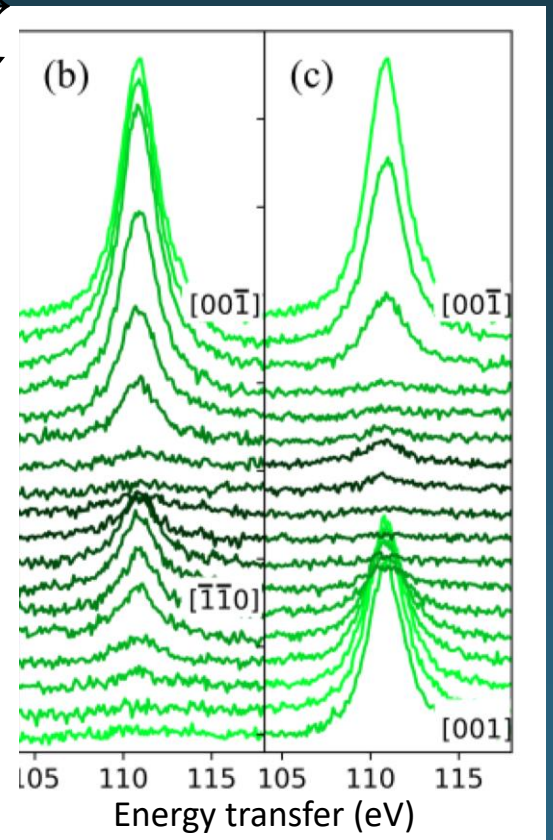
Spectroscopy on strongly correlated electron systems

s-NIXS on transition metal (TM) oxides: direct imaging of orbitals



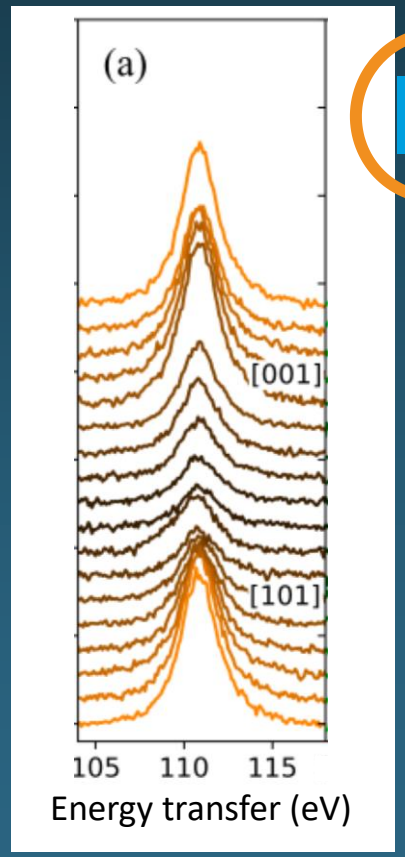


Total intensity *vs* angle

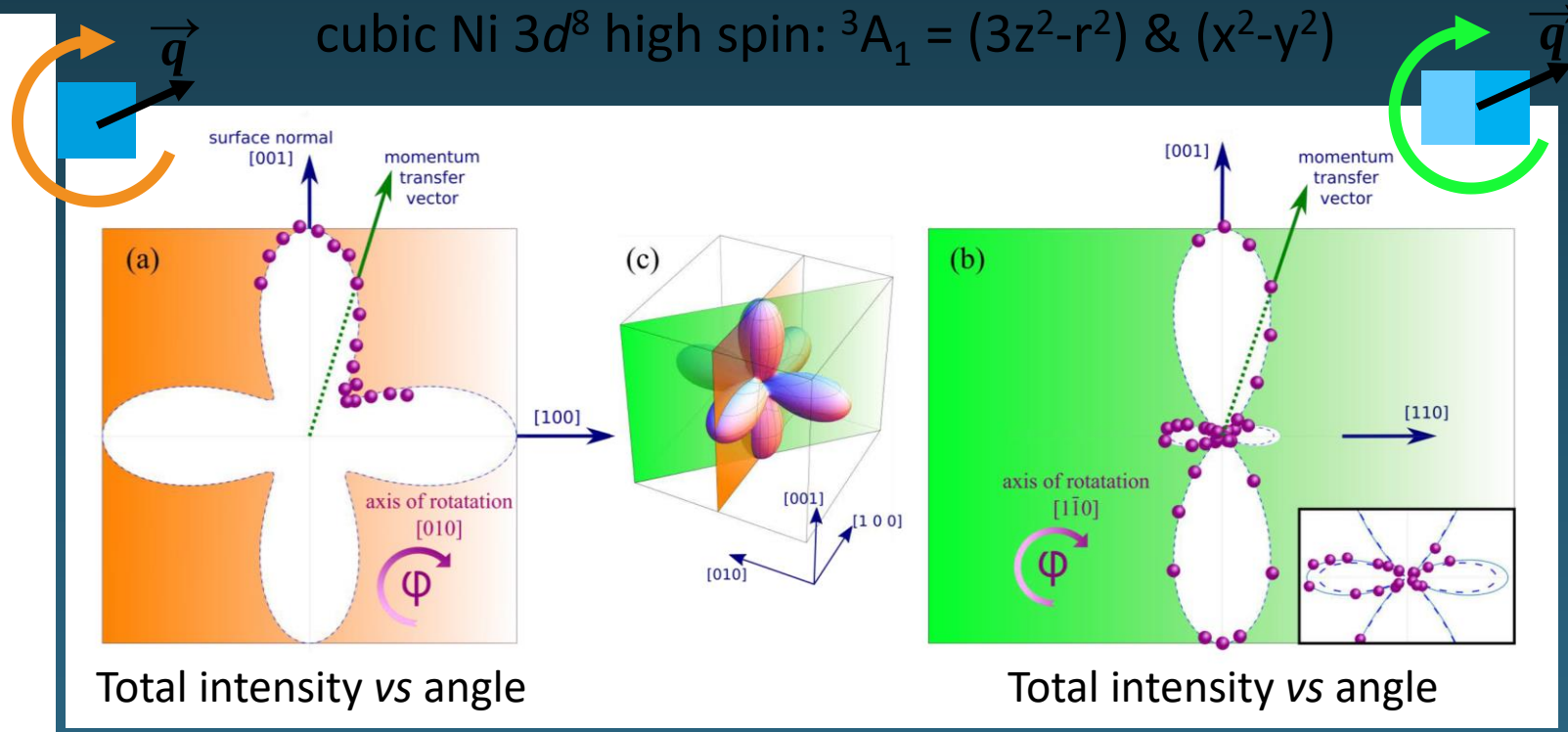


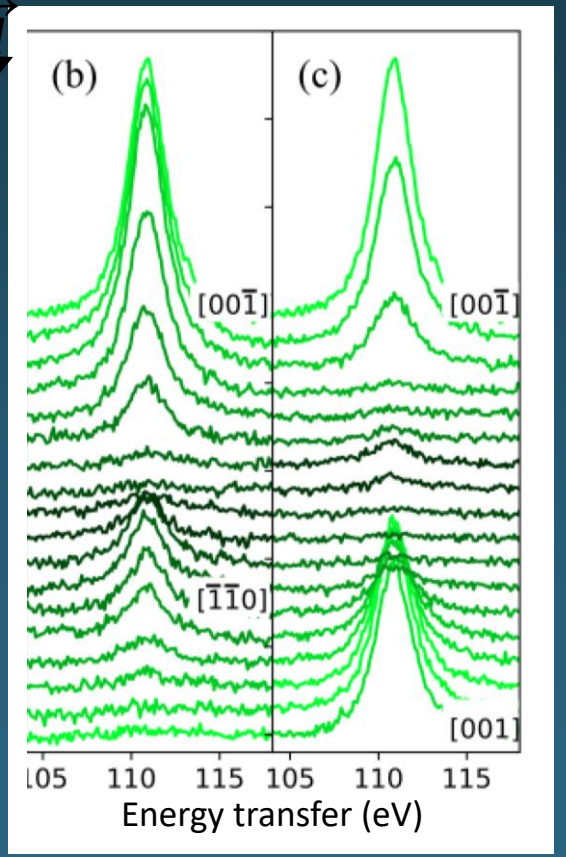
Spectroscopy on strongly correlated electron systems

s-NIXS on transition metal (TM) oxides: direct imaging of orbitals



cubic Ni $3d^8$ high spin: $^3A_1$ = $(3z^2\text{-}r^2)$ & $(x^2\text{-}y^2)$

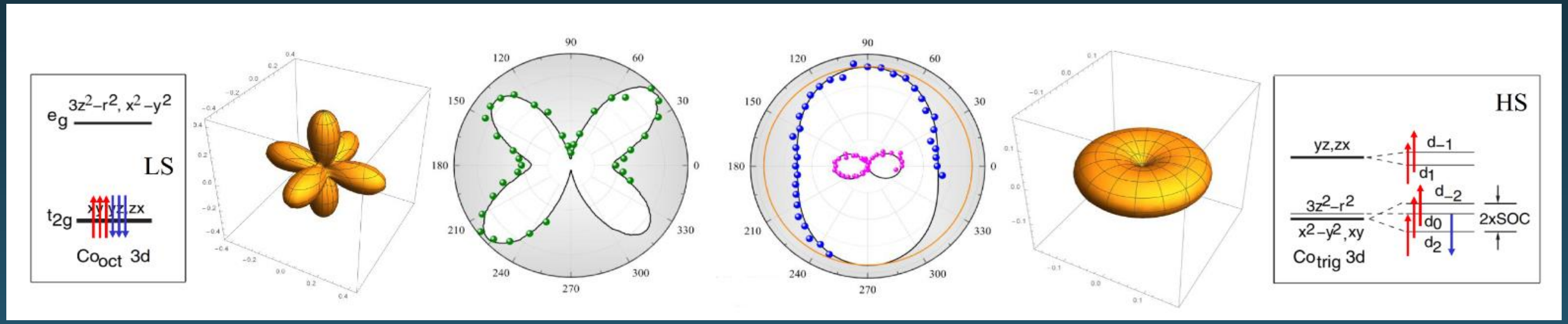




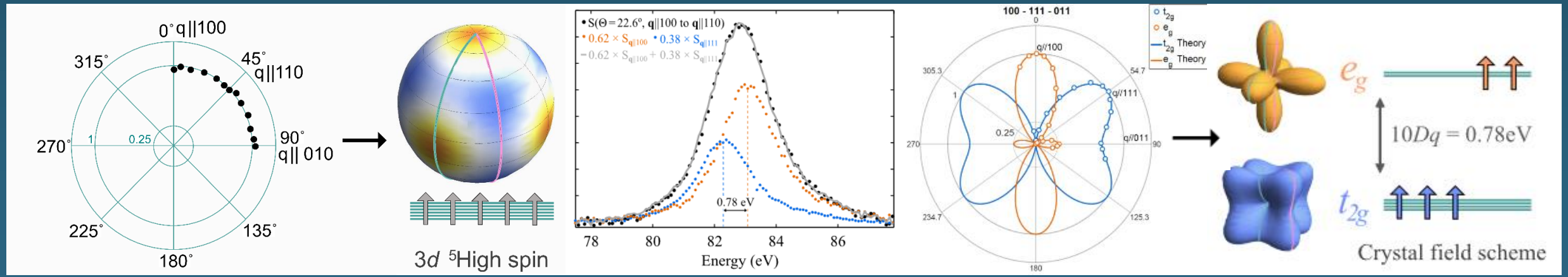
H. Yavas *et al.*, **Nature Physics 15**, 559 (2019)

Spectroscopy on strongly correlated electron systems

s-NIXS on transition metal (TM) oxides: direct orbital imaging



Leedahl *et al.*, **Nat. Comm. 10,** 5447 (2019)

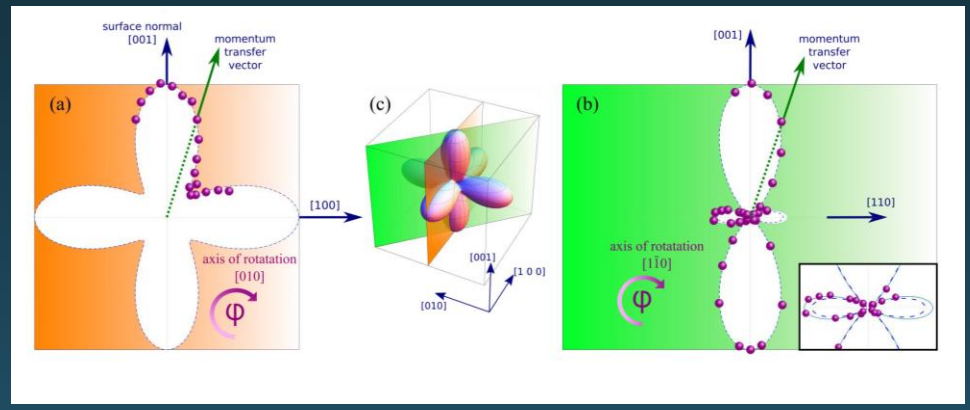


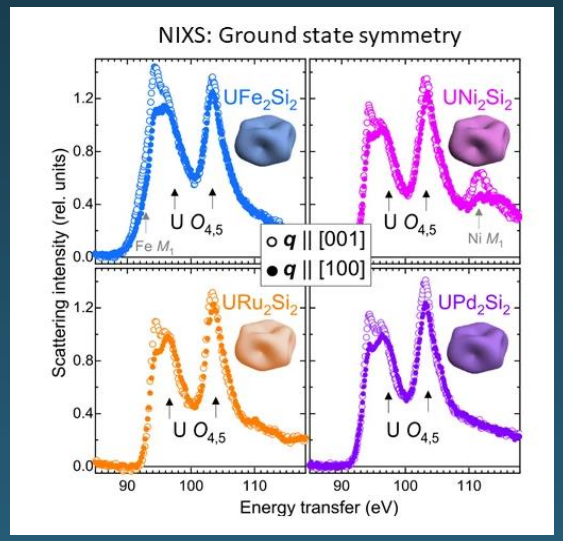
Amorese *et al.*, **PRX 11,** 011002 (2021)

Spectroscopy on strongly correlated electron systems

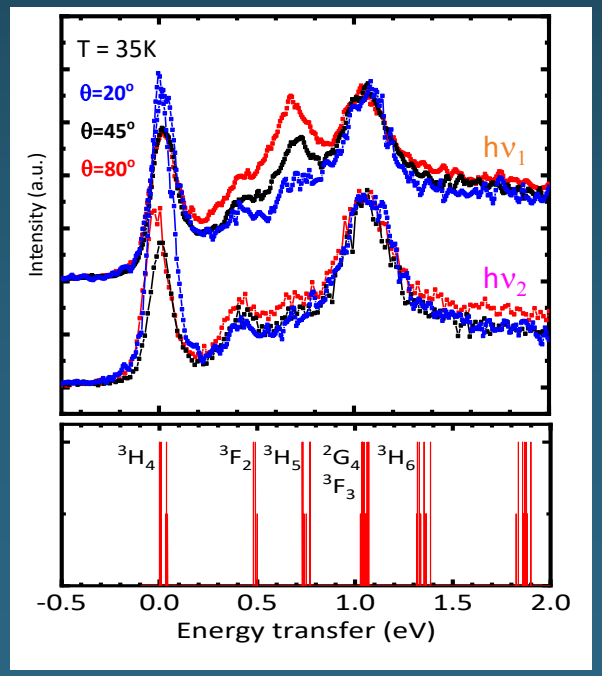
The collaboration between P01 and MPI CPfS Dresden

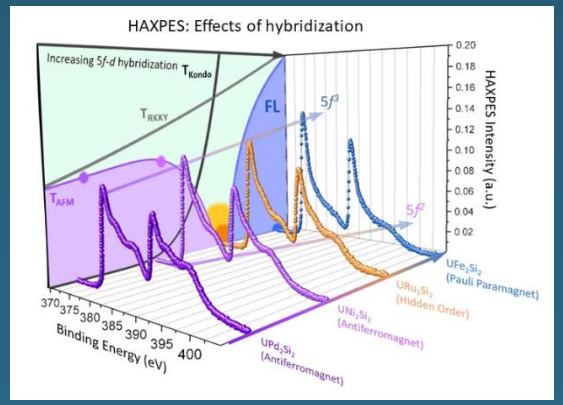
New method: s-NIXS:
direct imaging of charge densities
without sophisticated modeling





NIXS: Spectroscopy beyond dipole:
Ground state symmetry beyond 2-fold
Multiplets in metallic U compounds
No cleaving





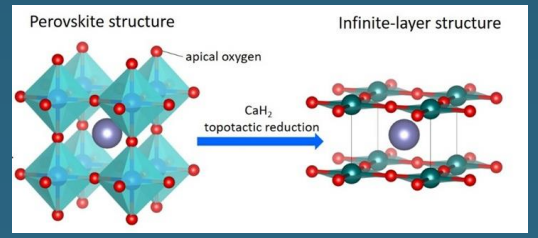
Tender RIXS at U $M_{4,5}$:
Low energy excitations

HAXPES:
Intermediate valence
4*f*: configuration interaction model

New collaborations

Prof. Jochen Geck
NIXS of Co $L_{2,3}$ in DAC

Dr. Berit Goodge
s-NIXS on nickelates



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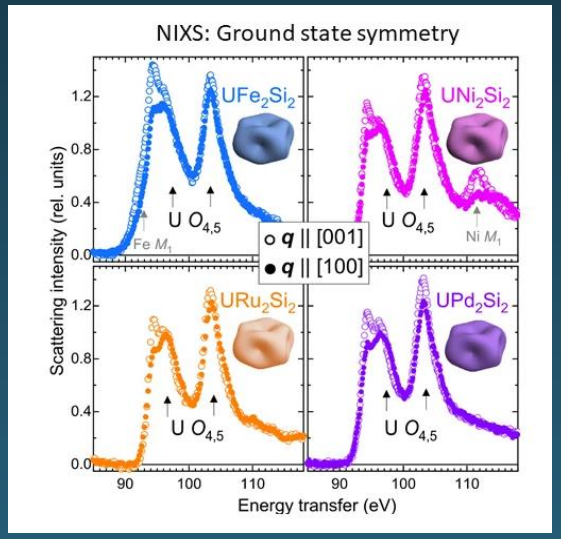
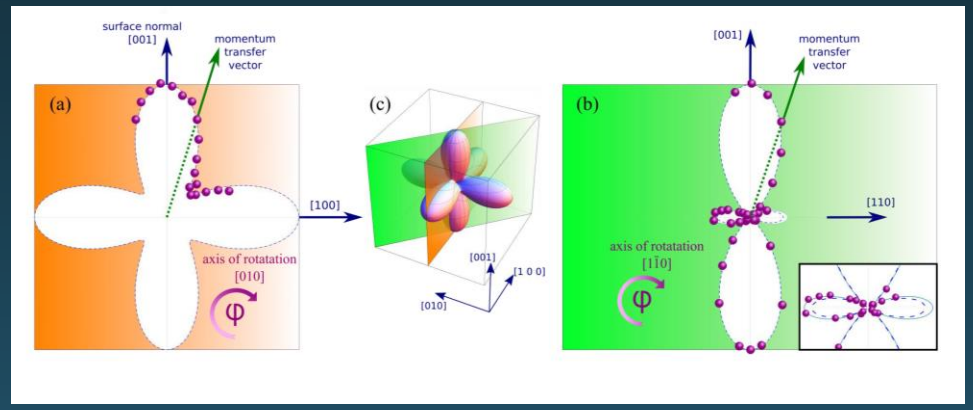
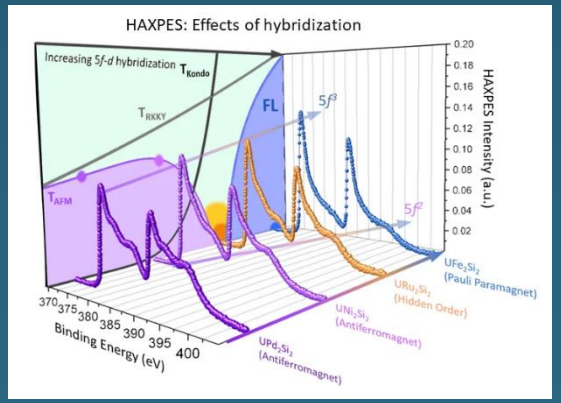




NIXS: Spectroscopy beyond dipole:
Ground state symmetry beyond 2-fold
Multiplets in metallic U compounds
No cleaving

Thank you!



Tender RIXS at U $M_{4,5}$:
Low energy excitations

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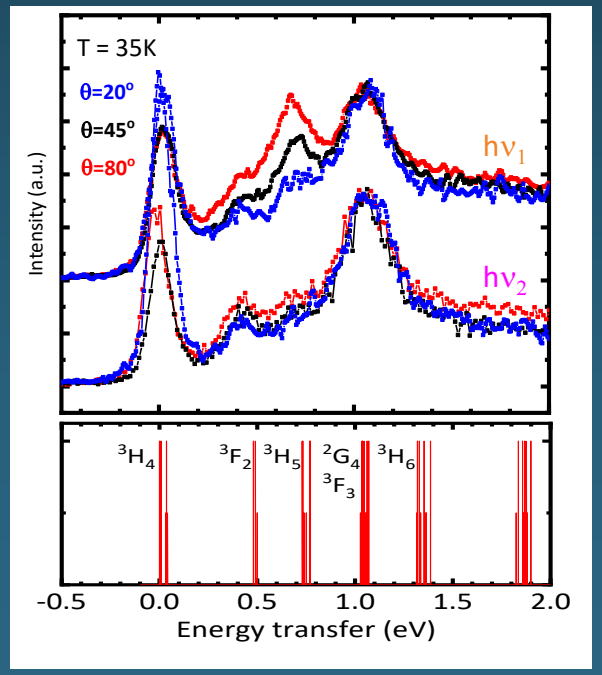
Intermediate valence
4*f*: configuration interaction model



New collaborations

Prof. Jochen Geck
NIXS of Co $L_{2,3}$ in DAC

Dr. Berit Goodge
s-NIXS on nickelates