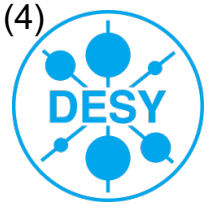
Direct imaging of orbitals using inelastic X-ray scattering

Hasan Yavaş

Co-authors:
L.H. Tjeng[1], M. Haverkort[2], A. Severing[3], M. Sundermann[1,3,4], A. Amorese[1,3], B. Leedahl[1], Hlynur Gretarsson[1,4]

Additional help: M. Harder, F-U. Dill, S. Mayer, D. Ketenoglu, H-C. Wille
C. Sternemann, M. Wilke, G. Spiekermann



SLAC NATIONAL ACCELERATOR LABORATORY

Outline – basis of this talk

SLAC

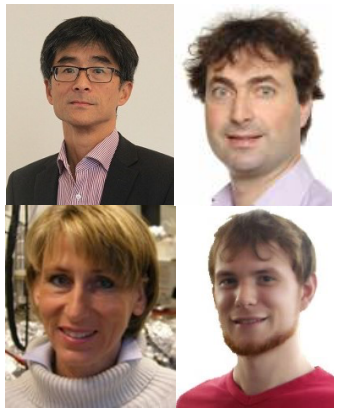
nature physics

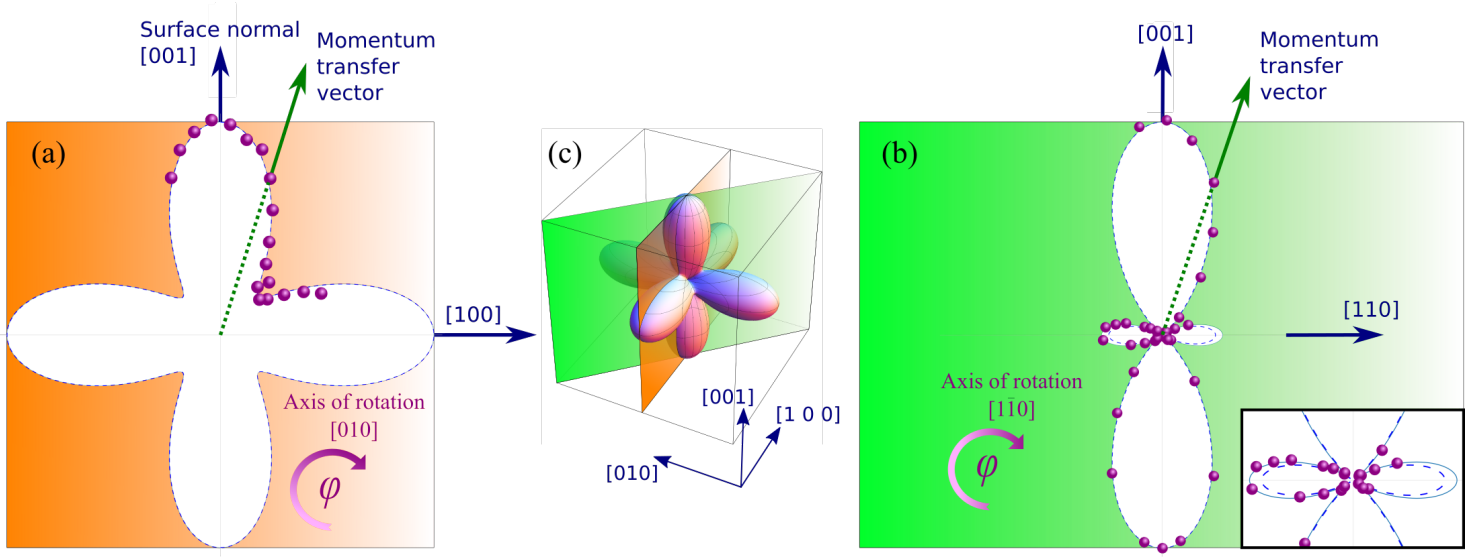
ARTICLES

https://doi.org/10.1038/s41567-019-0471-2

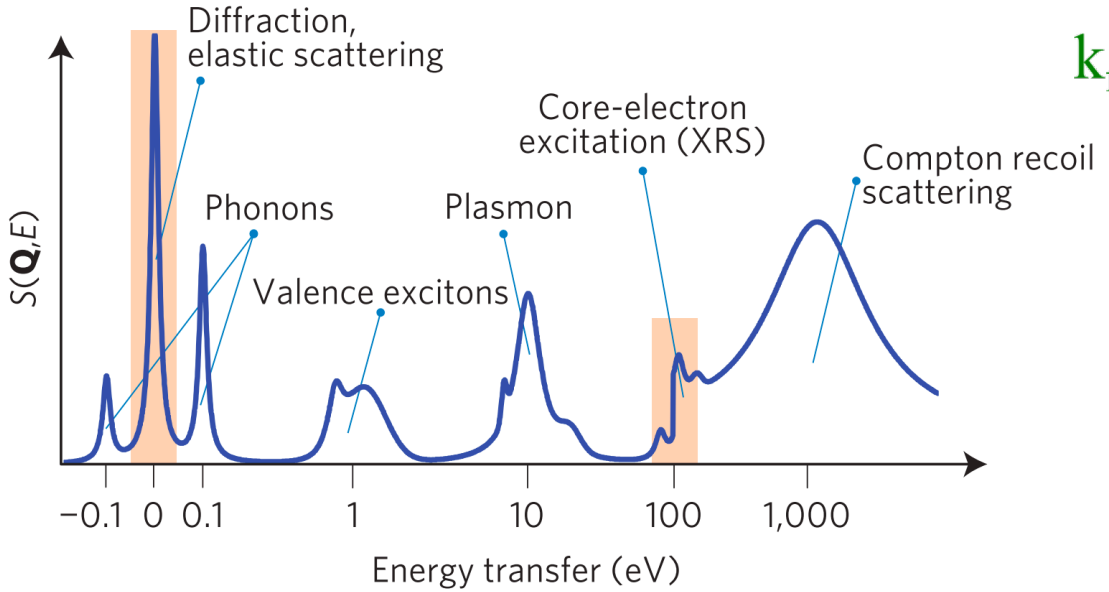
Direct imaging of orbitals in quantum materials

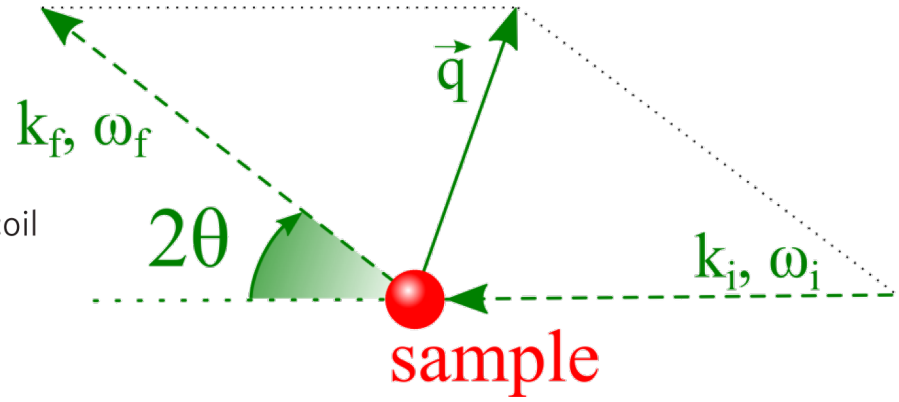
Hasan Yavaş[1,2,5], Martin Sundermann[1,3], Kai Chen[3,6], Andrea Amorese[1,3], Andrea Severing[1,3], Hlynur Gretarsson[1,2], Maurits W. Haverkort[4] and Liu Hao Tjeng[1*]





Inelastic X-ray scattering



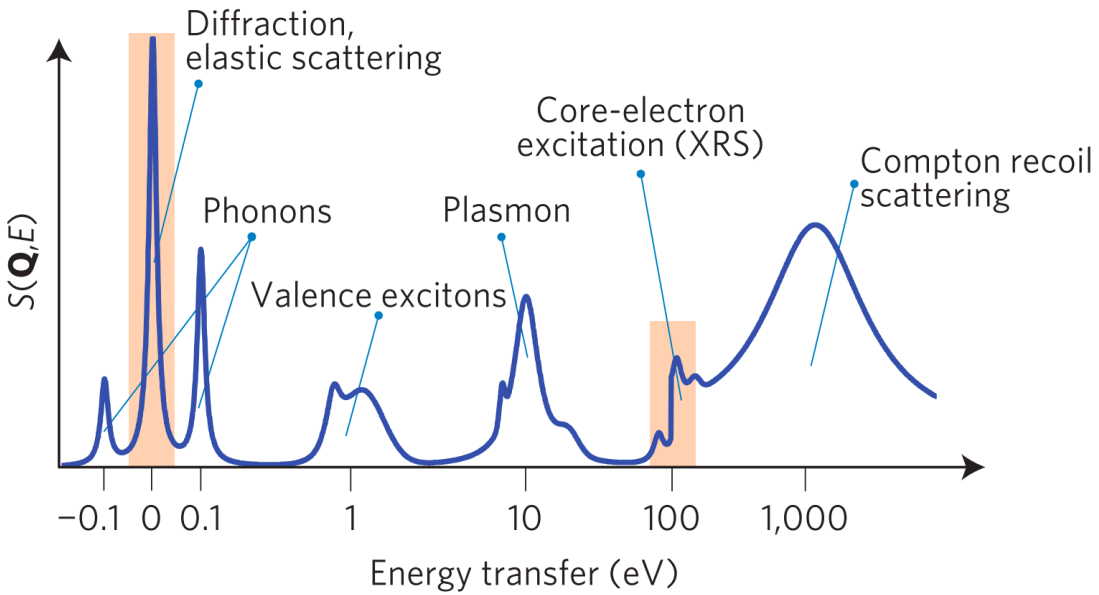


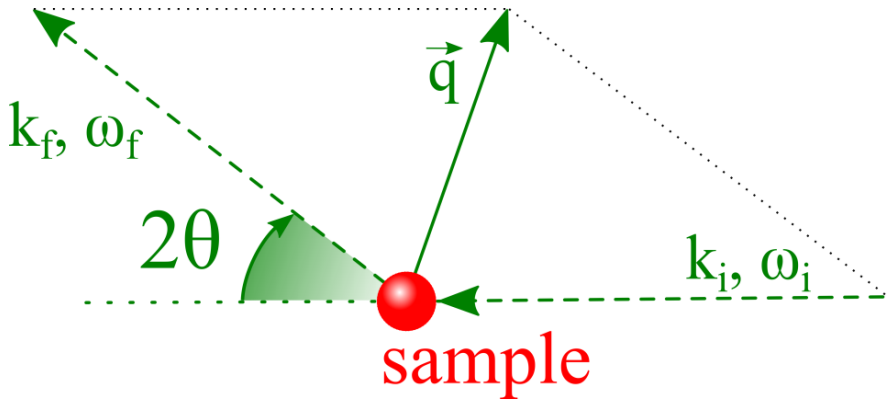
Inelastic: $\omega_i \neq \omega_f$

Elastic: $\omega_i = \omega_f$

W. Schülke, "Electron dynamics by inelastic x-ray scattering" Oxford University Press (2008)
M. van Veenendaal, "Theory of inelastic scattering and absorption of X-rays" Chambridge U. P. (2015)
C. J. Sahle, et al. *J. Synchrotron Radiat.*, 22, 1–10 (2015)
S. Huotari, et al. Nat. Mat. 10, 489 (2011)

Flavors of inelastic X-ray scattering





$$S(\vec{q},\omega) = \sum_f |\langle f|e^{i\vec{q}\cdot\vec{r}}|i\rangle|^2 \, \delta(\hbar\omega_i - \hbar\omega_f - \hbar\omega)$$

$$\vec{q} = \vec{k_i} - \vec{k_f}$$

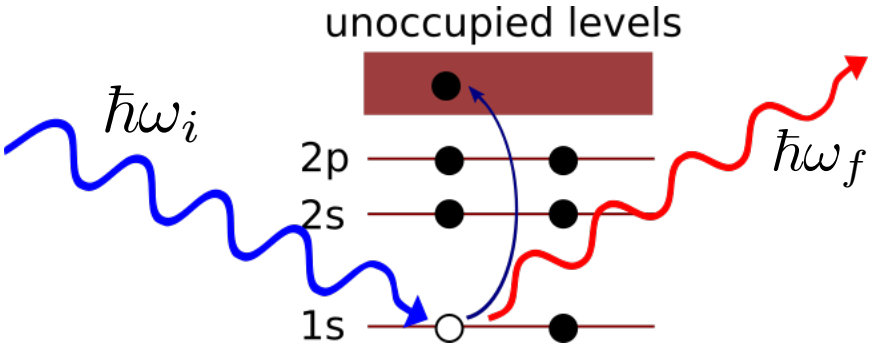
$$\hbar\omega = \hbar\omega_i - \hbar\omega_f$$

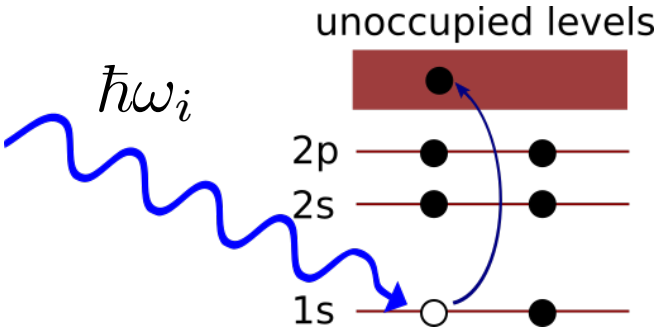
$qa \gg 1 \quad ; \quad \hbar\omega \gg E_B$ Compton

$qa \sim 1 \quad ; \quad \hbar\omega \approx E_B$ Core-shell

W. Schülke, “Electron dynamics by inelastic x-ray scattering“ Oxford University Press (2008)
K. Hamalainen and S. Manninen, *J. Phys. Condens. Matter* 13, 7539 (2001)

Inelastic process vs absorption





electron	$\hbar\omega = E_f - E_i$	$\hbar\omega = E_f - E_i$
photon	$\hbar\omega = \hbar\omega_i - \hbar\omega_f$	$\hbar\omega = \hbar\omega_i$

$$S(\vec{q},\omega) = \sum_f |\langle f|e^{i\vec{q}\cdot\vec{r}}|i\rangle|^2\,\delta(\hbar\omega_i - \hbar\omega_f - \hbar\omega)$$

$$e^{i\vec{q}\cdot\vec{r}} \approx 1 + i\vec{q}\cdot\vec{r} - \frac{(\vec{q}\cdot\vec{r})^2}{2} + \cdots$$

$q : small$

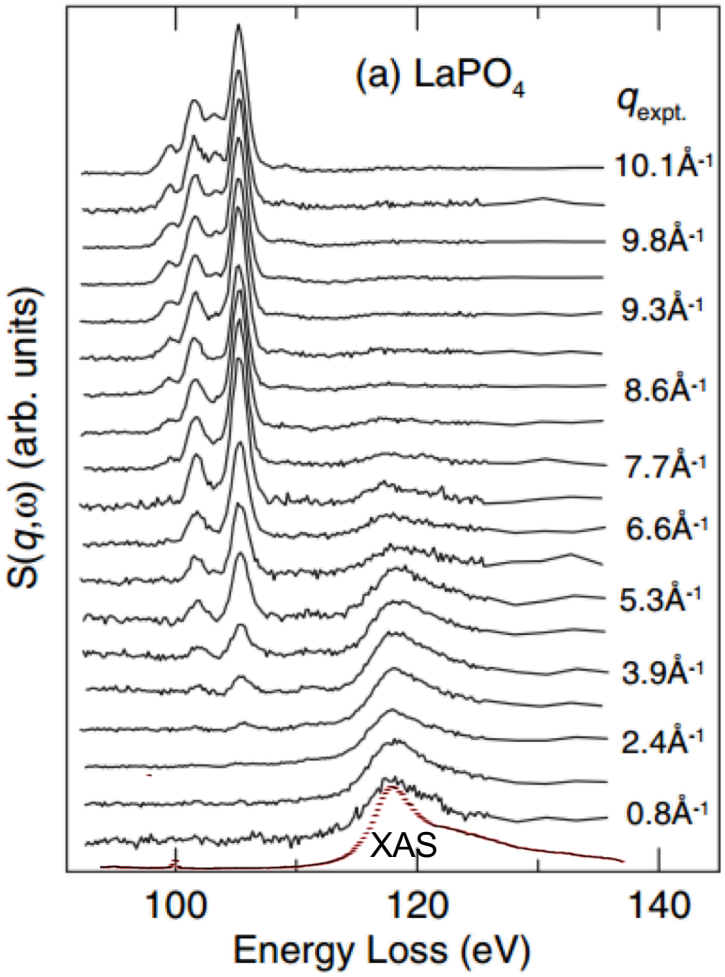
$$S(\vec{q},\omega) \approx \sum_f |\langle f|\vec{q}\cdot\vec{r}|i\rangle|^2\,\delta(\hbar\omega_i - \hbar\omega_f - \hbar\omega) \qquad \mu \propto \sum_f |\langle f|\vec{\epsilon}\cdot\vec{r}|i\rangle|^2\,\delta(E_f - E_i - \hbar\omega)$$

$\vec{q}$ & $\hbar\omega_i$: independent parameters to tweak

"Almost" All sample dependent parameters

IXS as X-ray absorption spectroscopy

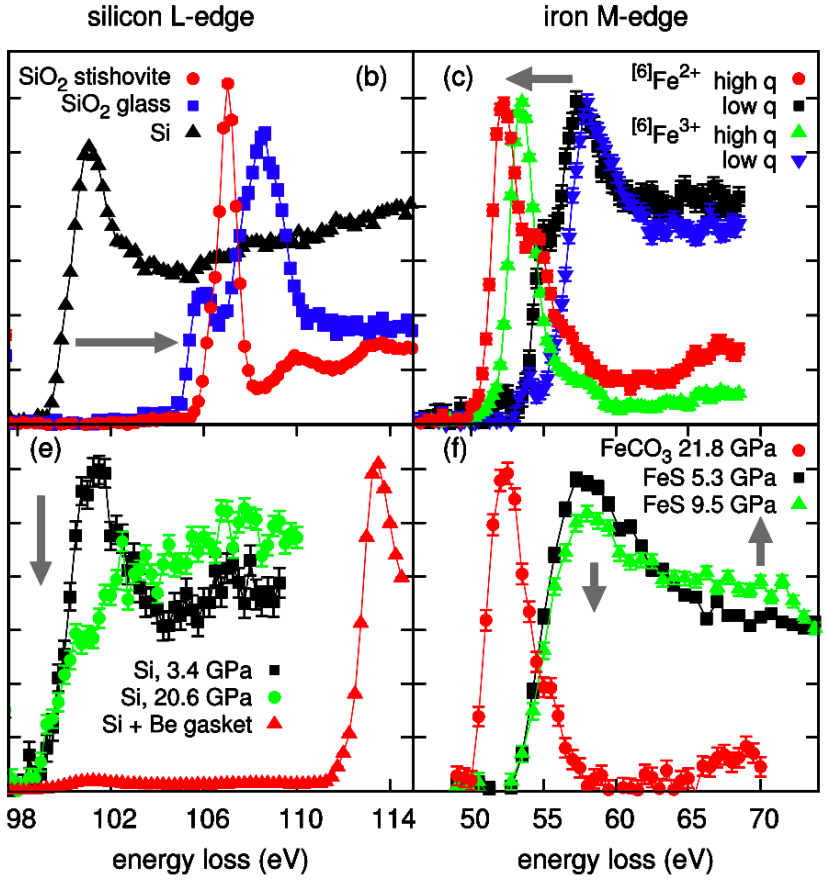
M. Haverkort, et al. PRL (2007)
R. A. Gordon, et al., EPL (2008)



…and beyond!

$$S(\vec{q},\omega) \approx \sum_f |\langle f|\vec{q}\cdot\vec{r}|i\rangle|^2\,\delta(\hbar\omega_i - \hbar\omega_f - \hbar\omega)$$

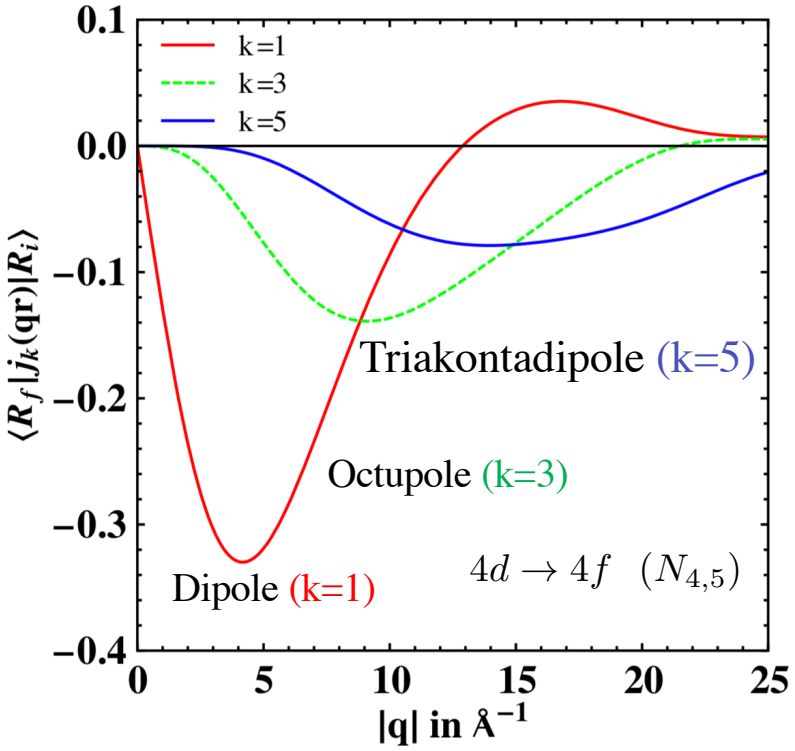
$$S(\vec{q},\omega) = \sum_f |\langle f|e^{i\vec{q}\cdot\vec{r}}|i\rangle|^2\,\delta(\hbar\omega_i - \hbar\omega_f - \hbar\omega)$$



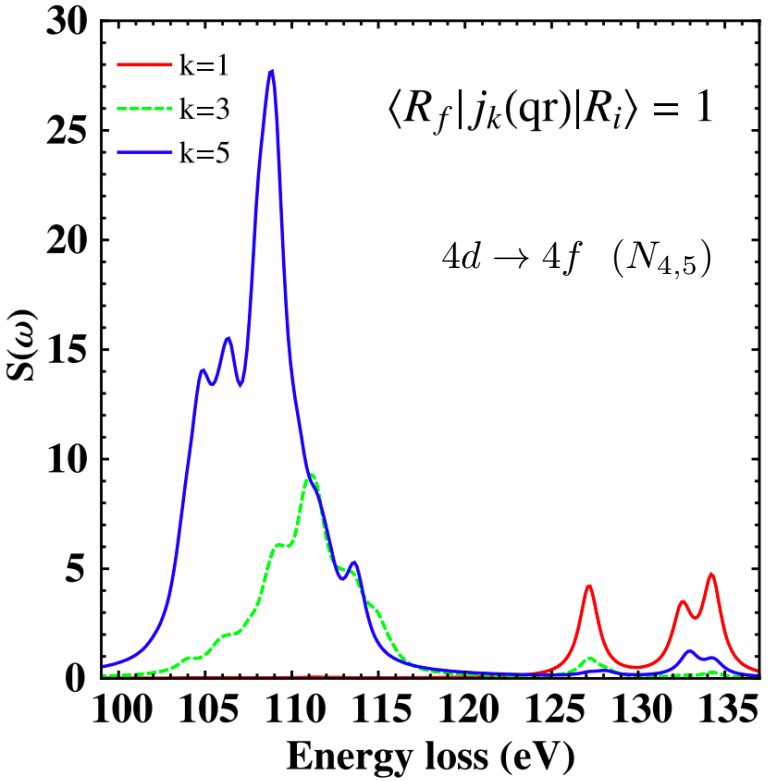
C. Sternemann and M. Wilke, High Pressure Research (2016)

Higher order terms

$$e^{i\vec{q}\cdot\vec{r}} = 4\pi \sum_{k}^{\infty} \sum_{m=-k}^{k} i^k j_k(qr) Y^*_{km}(\theta_q, \phi_q) Y_{km}(\theta_r, \phi_r)$$



T. Willers *et al.*, *PRL* 109, 4, 046401 (2012)



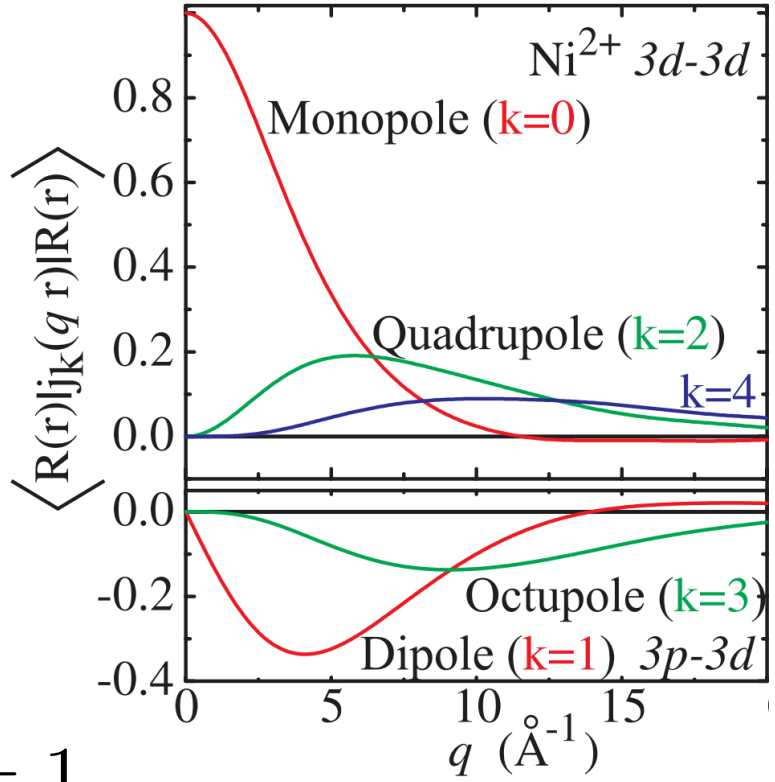
M. W. Haverkort, PRL 99, 25, 257401 (2007)

Triangular condition & parity rule

$$e^{i\vec{q}\cdot\vec{r}} = 4\pi \sum_{k}^{\infty} \sum_{m=-k}^{k} i^k j_k(qr) Y^*_{km}(\theta_q, \phi_q) Y_{km}(\theta_r, \phi_r)$$

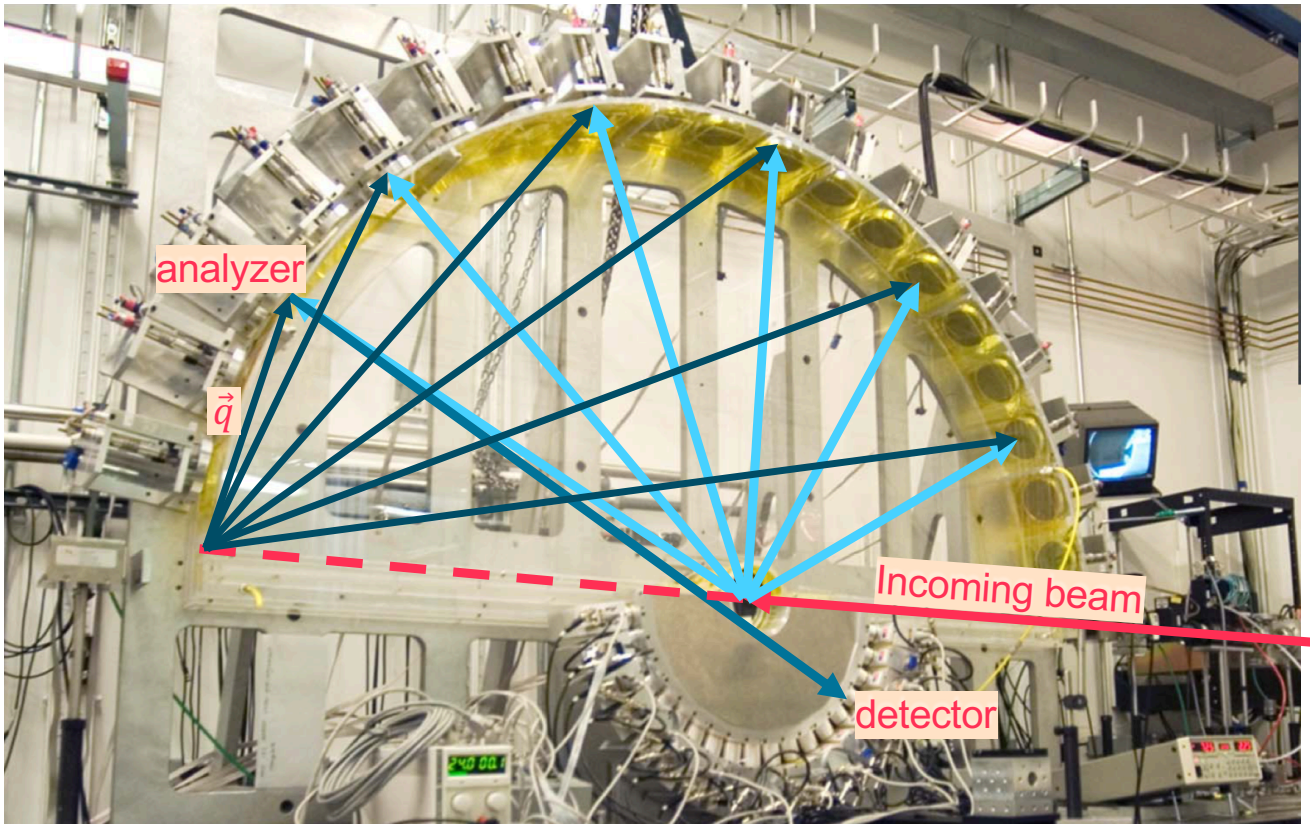
$$|l_f - l_i| \leq k \leq |l_f + l_i|$$

$$l_i + l_f + k = even$$



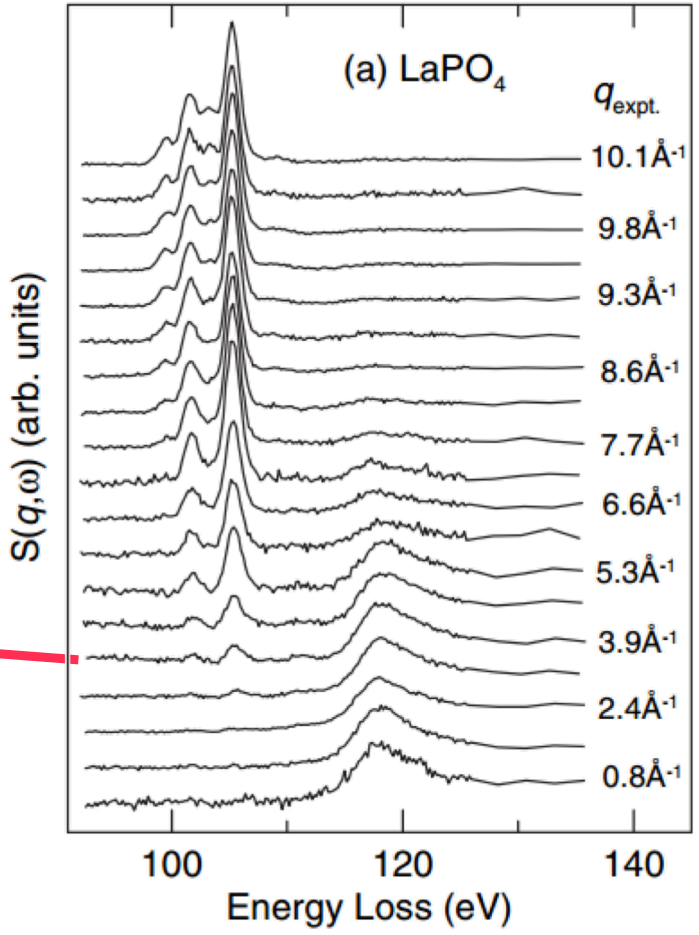
$$s \rightarrow p \;\; ; \;\; l_i = 0 \rightarrow l_f = 1 \;\; ; \;\; k = 1$$
$$p \rightarrow d \;\; ; \;\; l_i = 1 \rightarrow l_f = 2 \;\; ; \;\; k = 1, 3$$
$$s \rightarrow d \;\; ; \;\; l_i = 0 \rightarrow l_f = 2 \;\; ; \;\; k = 2$$
$$d \rightarrow f \;\; ; \;\; l_i = 2 \rightarrow l_f = 3 \;\; ; \;\; k = 1, 3, 5$$

Higher order transitions are allowed and q dependent



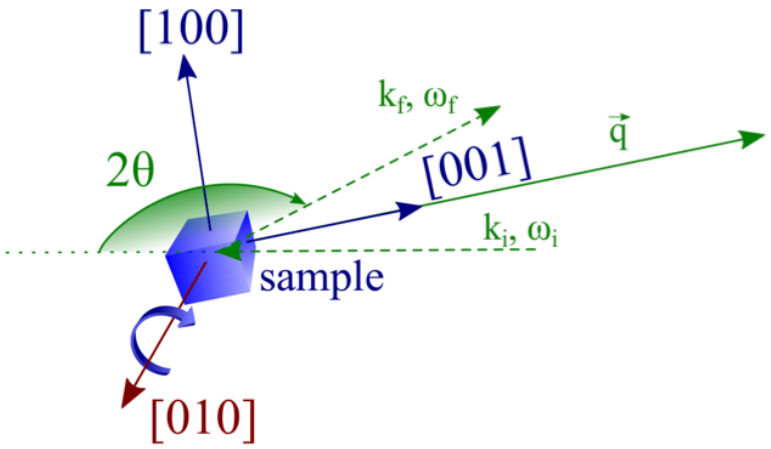
LERIX – APS 20-ID

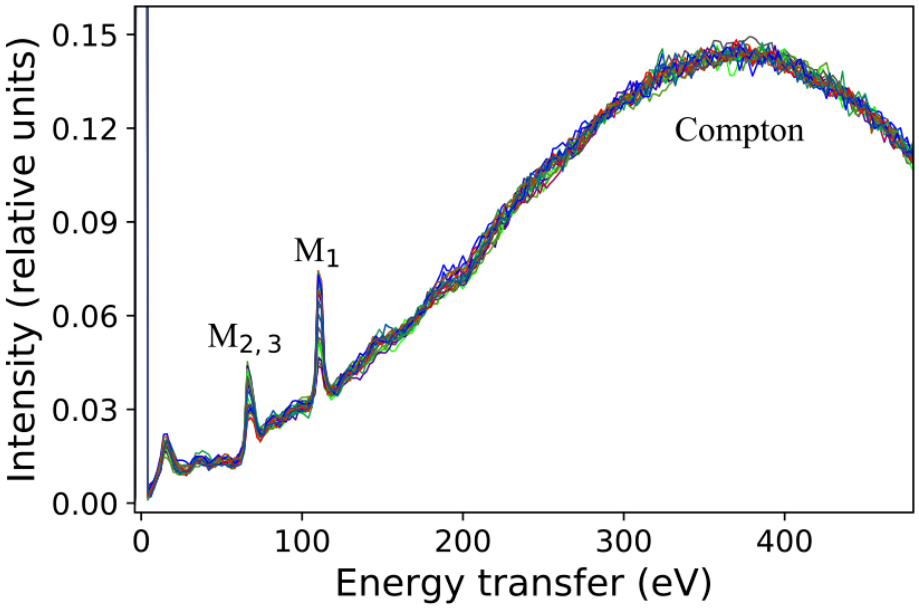
T. Fister, *et al.*, RSI (2006)

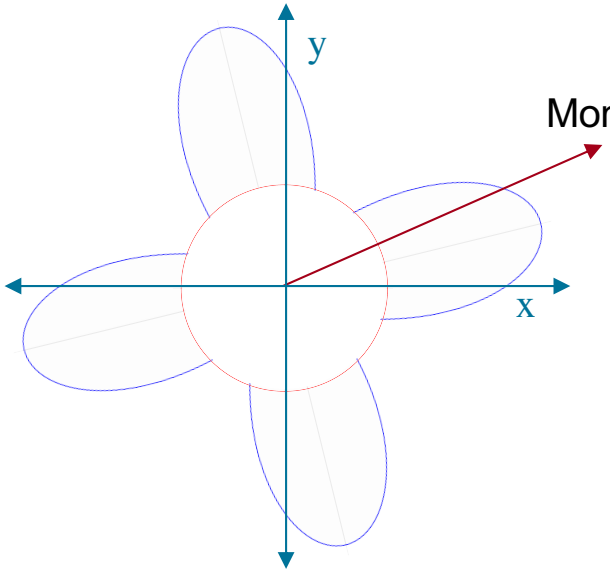


R. A. Gordon, et al., EPL (2008)

Measurement and results

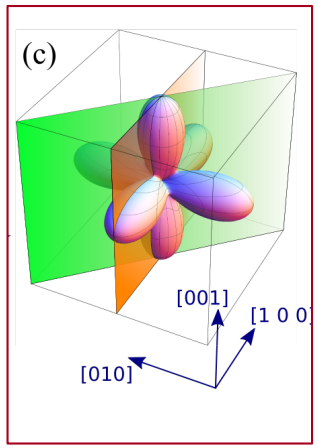


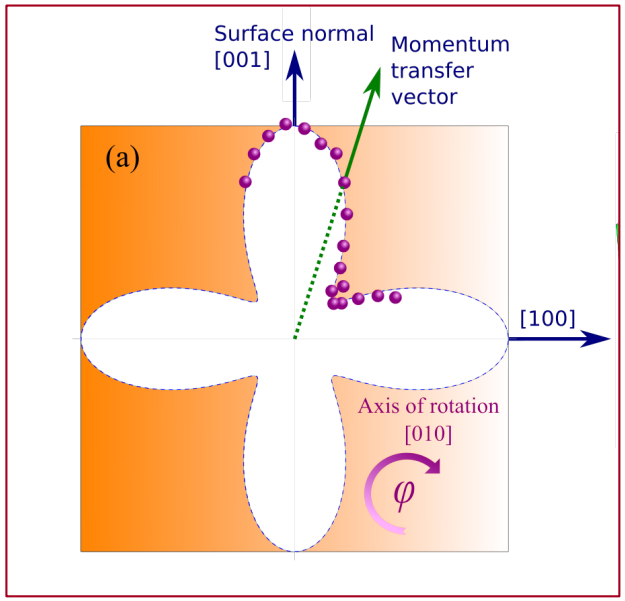


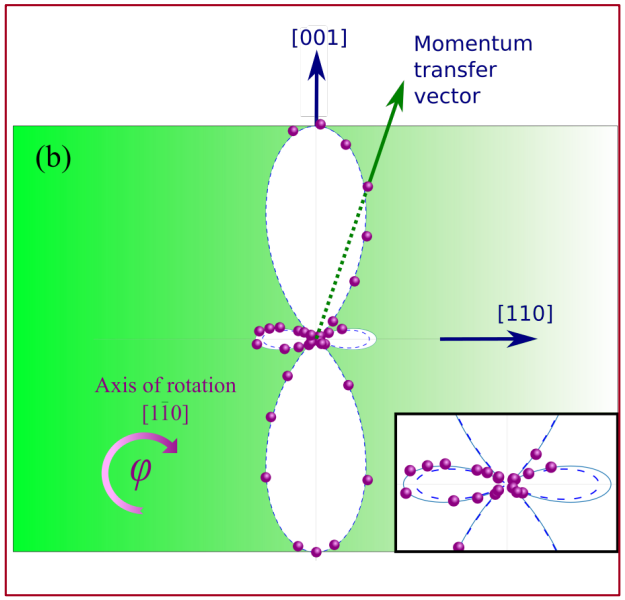


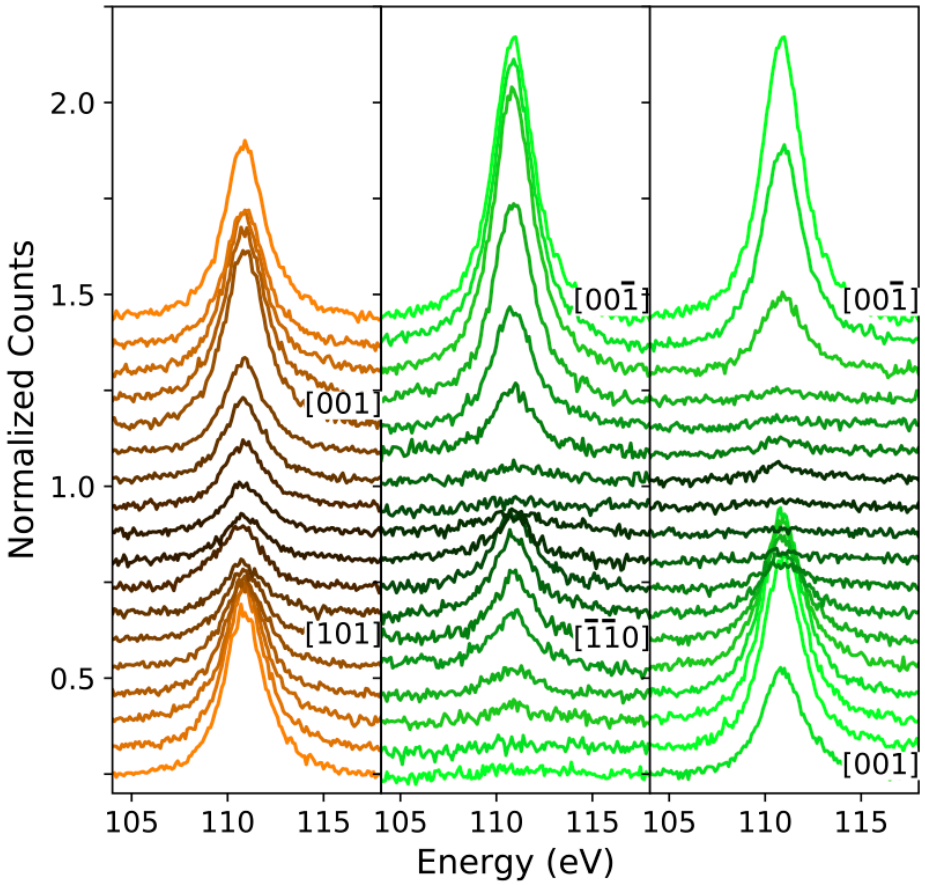
$M_1 : s \rightarrow d$

Measurement and results





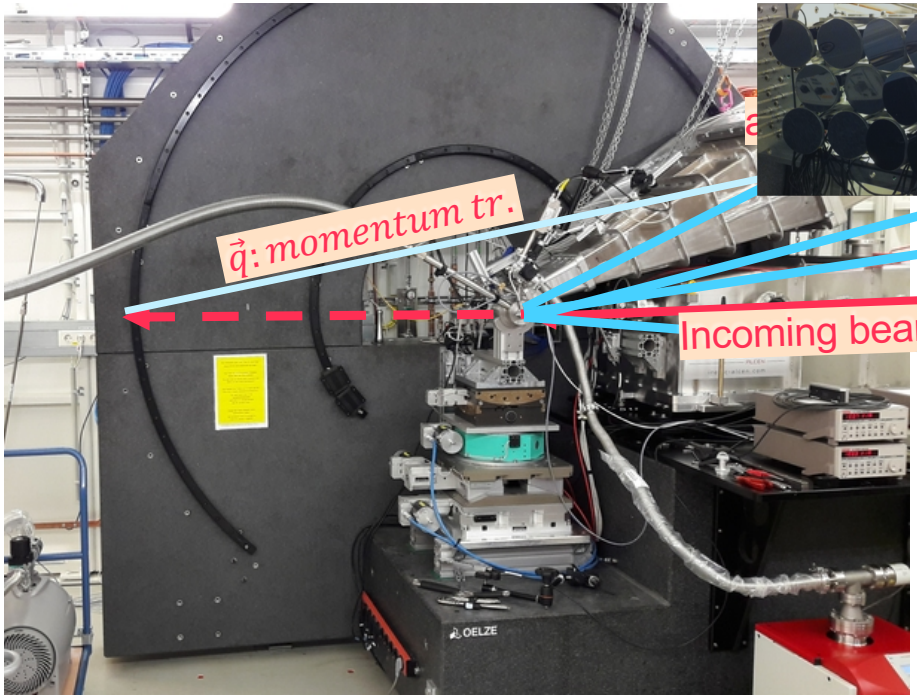


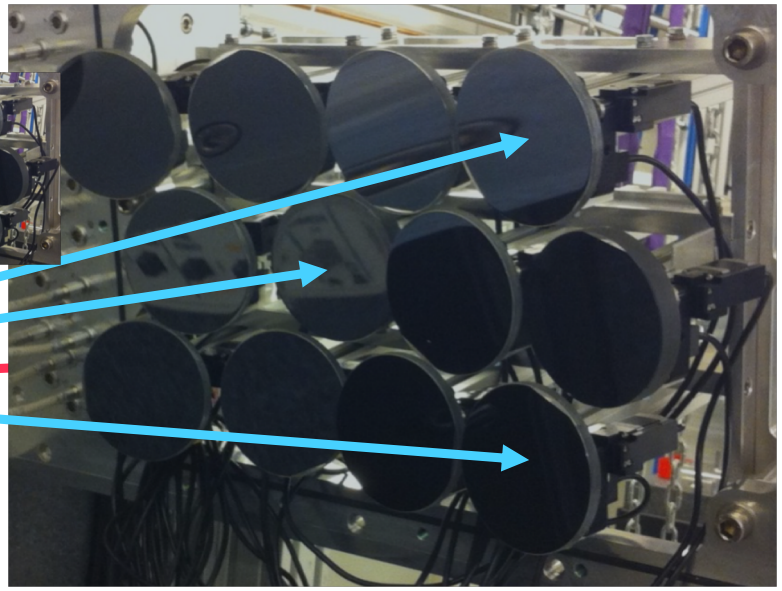


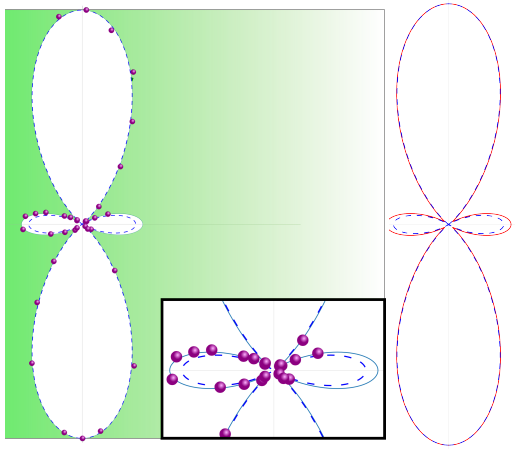
s-NIXS
s-core-level non-resonant IXS

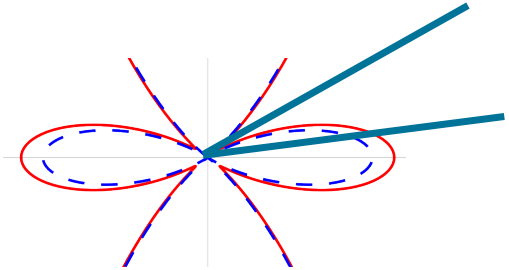
The instrument at P01 – PETRA III

M. Sundermann

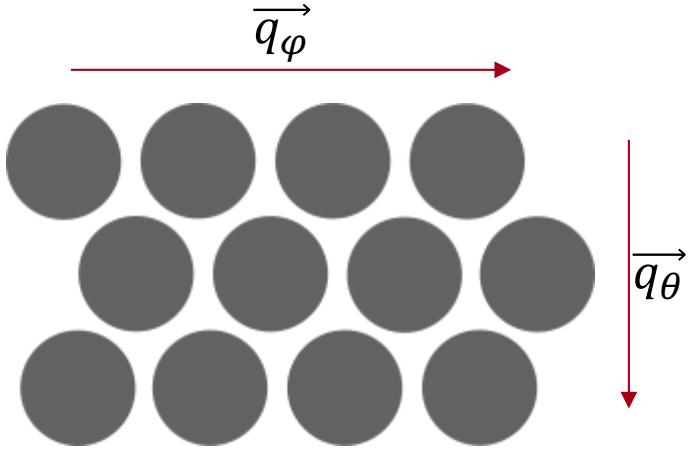






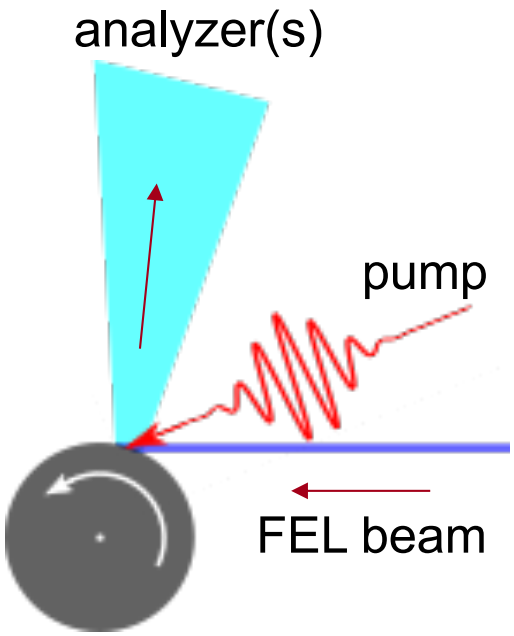


A. Amorese

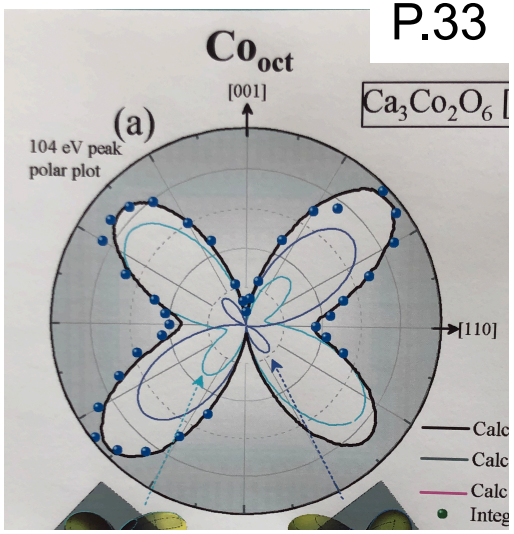


Present - Future

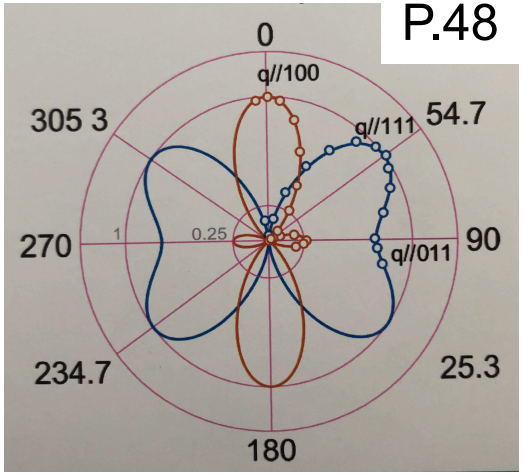
Time for time-resolved IXS?



P.33



P.48



MPI-Dresden

Ideas for
pioneering IXS studies
at FELs are welcome!