Time-resolved ESCA and PED with the new 60° ARTOF at Slicing

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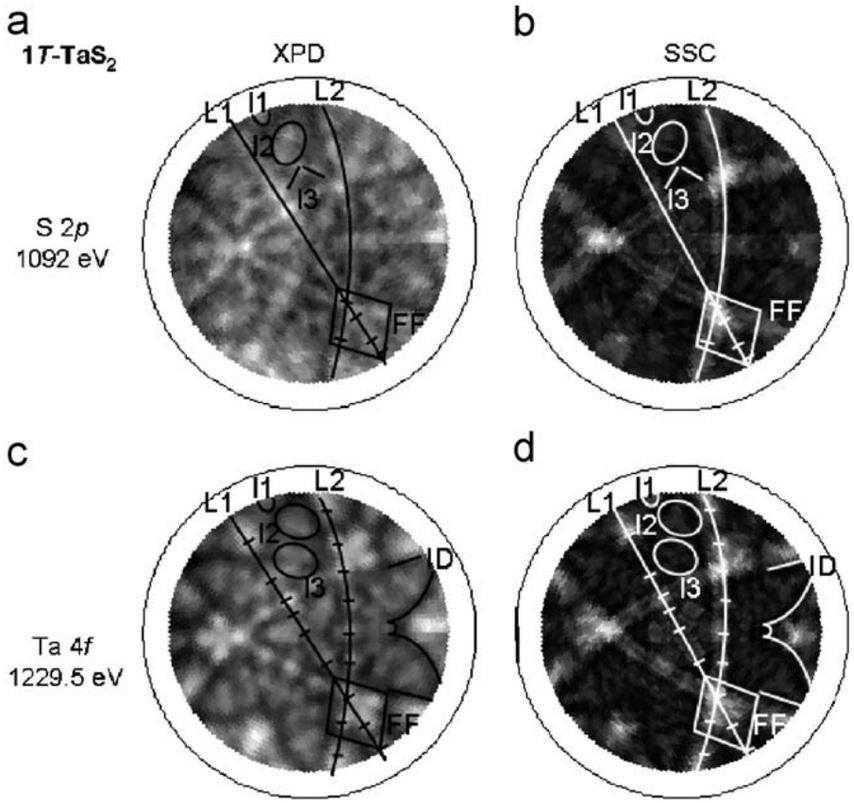
What can Photoemission provide?

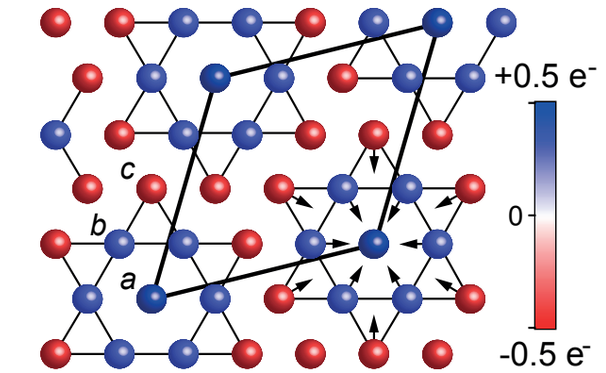
TaS2 - a layered CDW compound

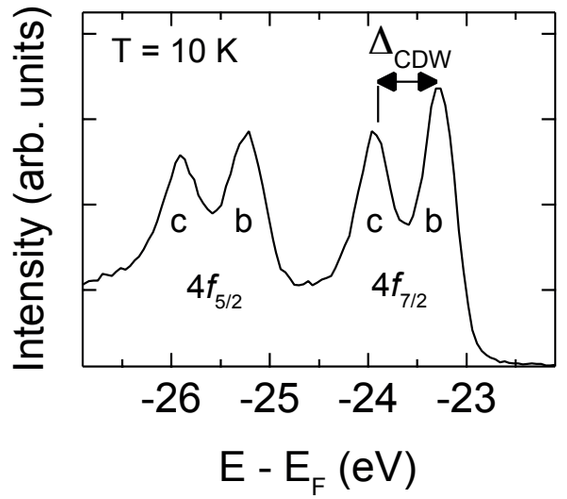
(Crystal) Structure via PED (@RT)



Stoltz, D. & Stoltz, S. E.,
Physica B: Condensed Matter 398, 172–177 (2007).

(Electronic) Structure via ESCA

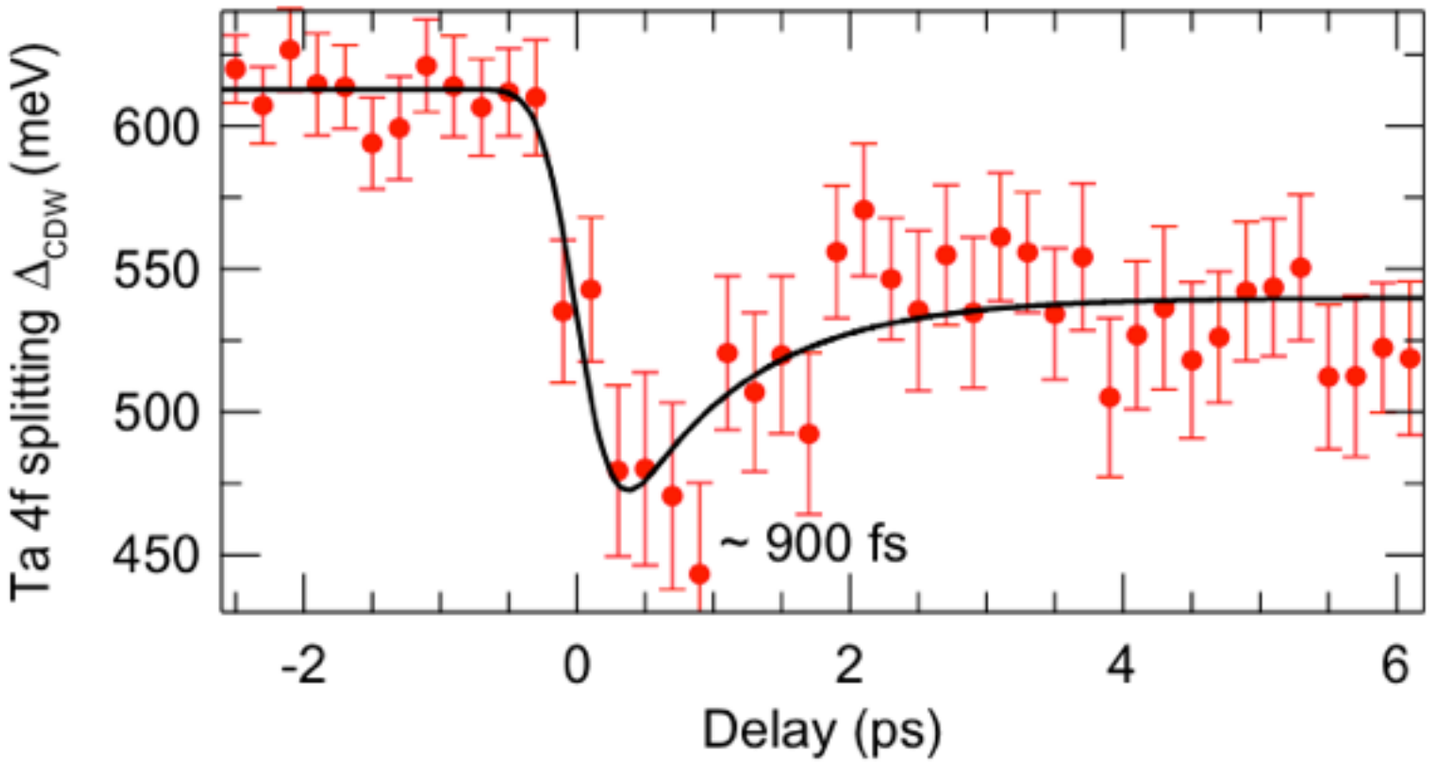


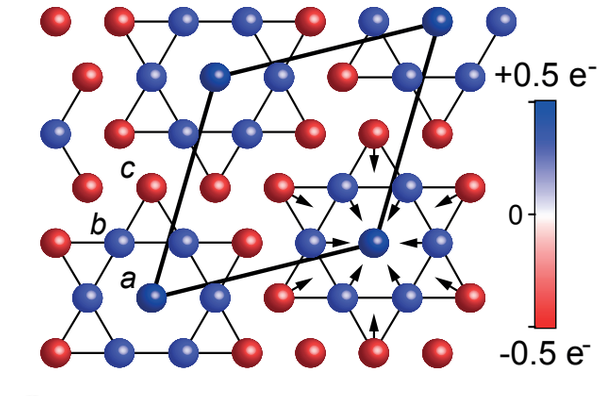


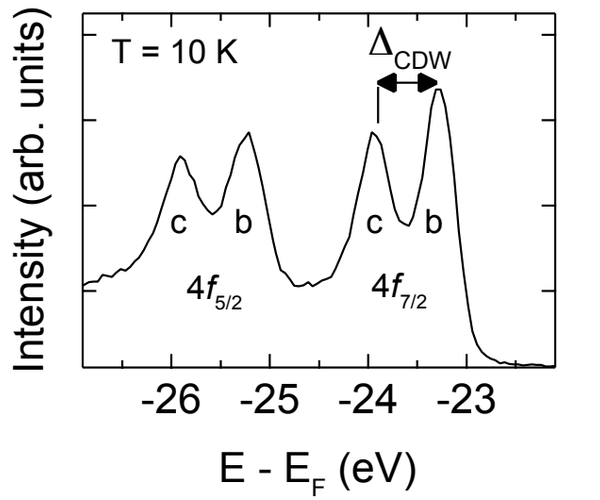
Hellmann, S. et al., New J. Phys. 14, 013062 (2012).

What can Photoemission provide?

Time-Resolved ESCA at FLASH



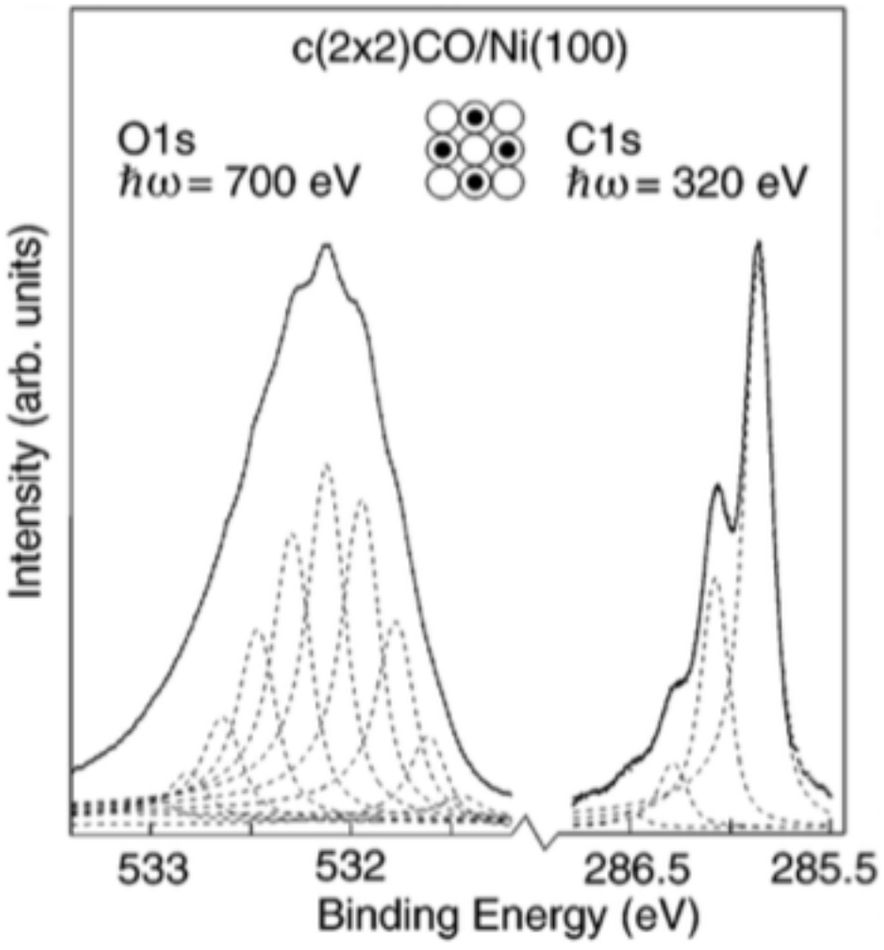


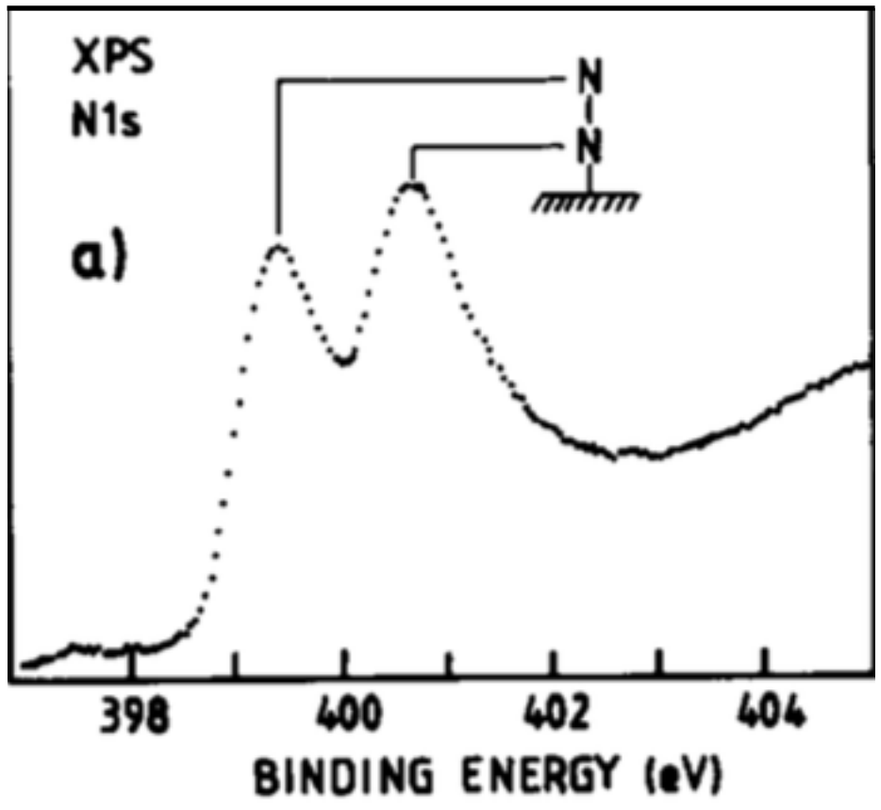


Hellmann, S. et al., New J. Phys. 14, 013062 (2012).

What can Photoemission provide?

Chemical Shift of Surface Adsorbates



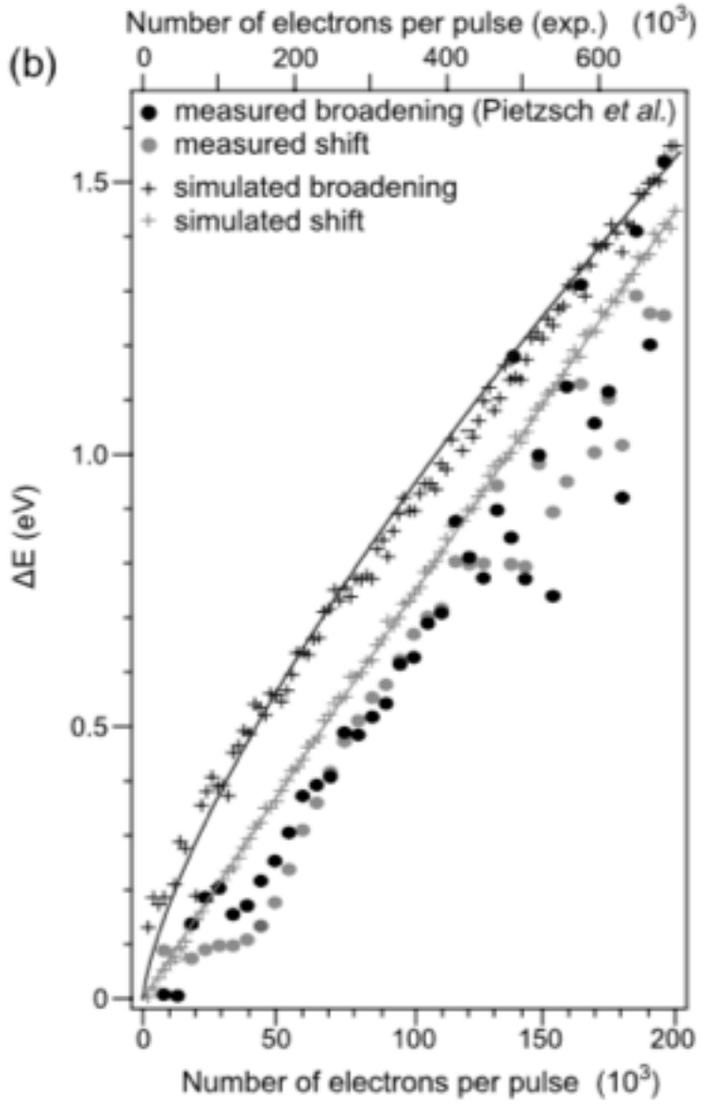


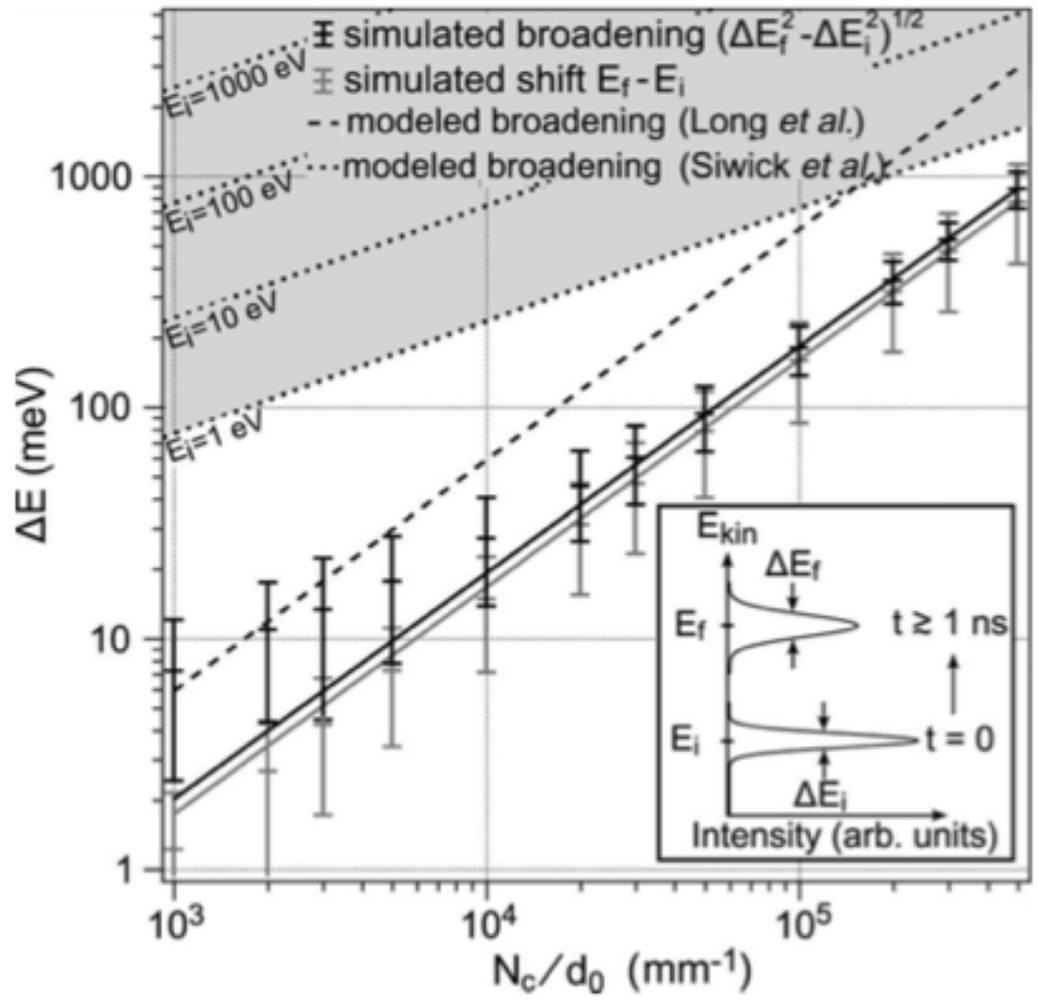
1. Föhlisch, A. et. al., Physical Review 81, 1730–1733 (1998).

Mårtensson, N. & Nilsson, A.,
J. of Elec. Spect. and Rel. Phen. 72, 1–8 (1995).

What can Photoemission provide?

Space-charge effects are problematic

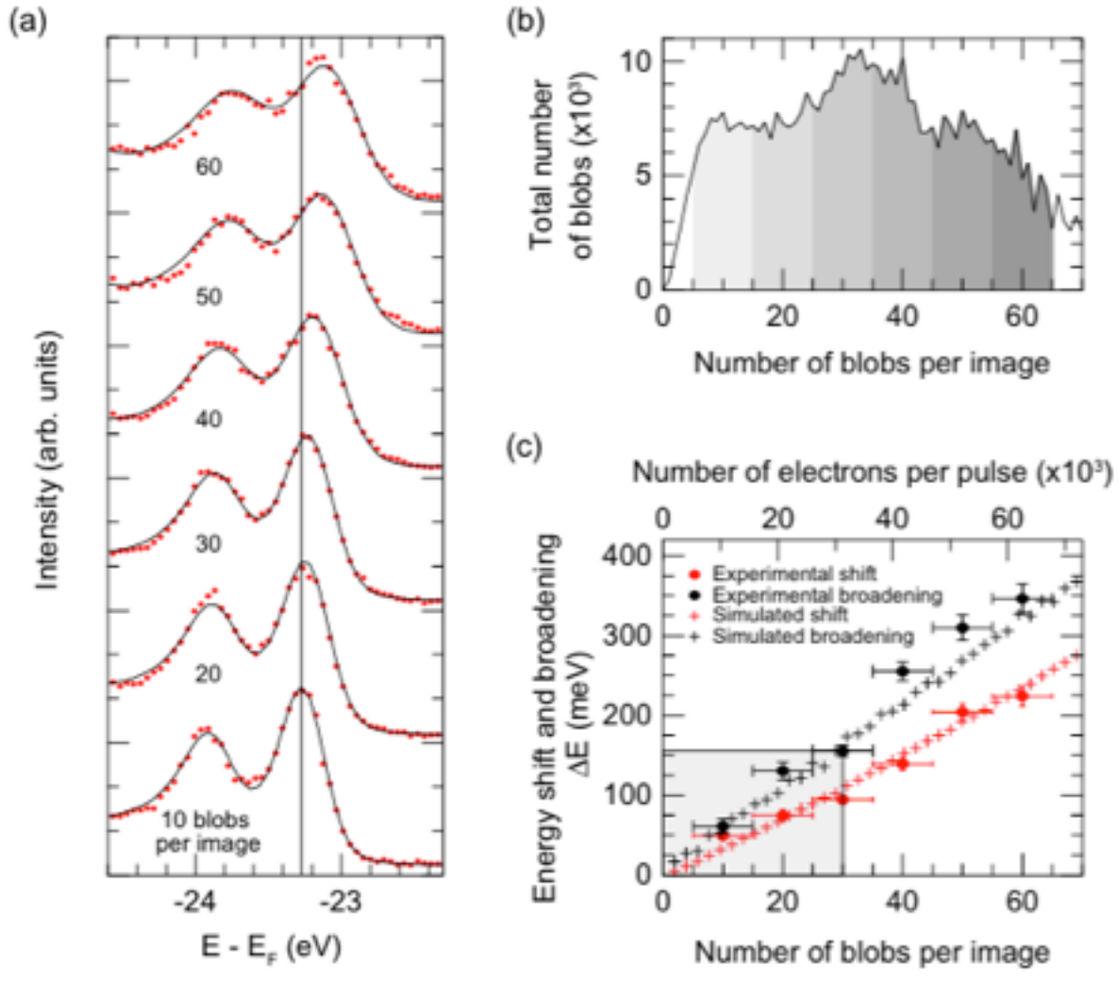




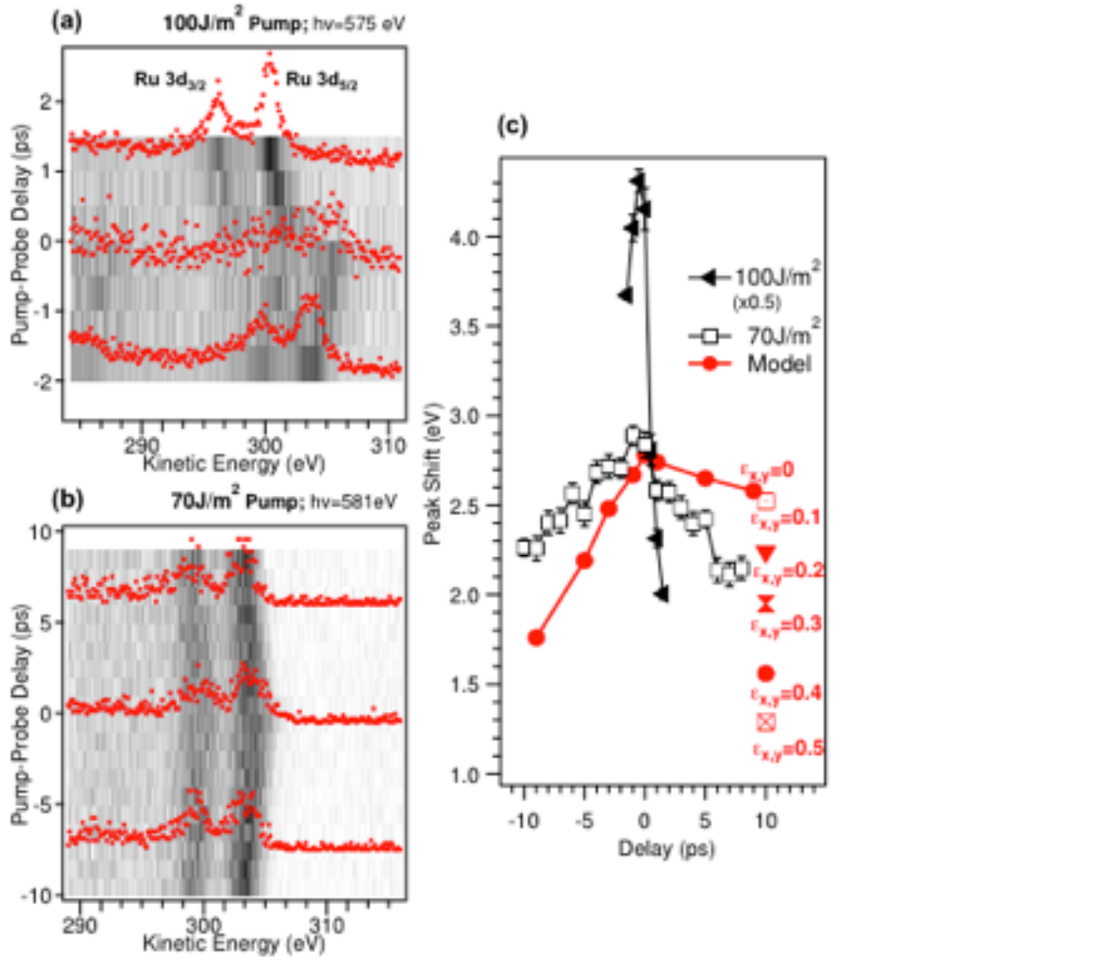
Hellmann, S., Rossnagel, K., Marczynski-Bühlow, M. & Kipp, L., *Phys. Rev. B* **79,** 035402–12 (2009).

What can Photoemission provide?

Space-charge effects are problematic



Hellmann, S. et al., New J. Phys. 14, 013062 (2012).



Dell'Angela, M. et al., Struct. Dyn. 2 025101 (2015)

What can Photoemission provide?

High peak flux is not necessarily the best since

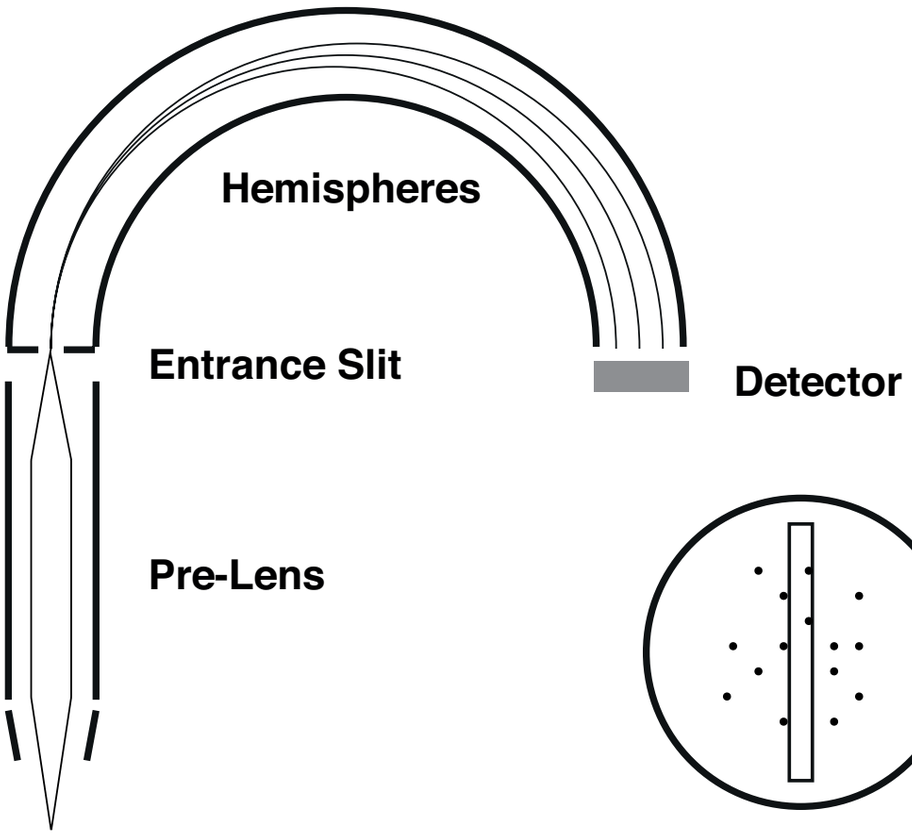
- **space charge effects need to be taken into account hence FELs might not be the ideal source (only SC linacs can compensate with high rep. rate)**
- **adsorbates on single crystals can desorb when exposed to too much flux**

A high repetition rate source (i.e. a synchrotron) is an ideal source for photoemission. However for some applications we need to improve the detection efficiency

Principles and Operation of the ArTOF

Hemispherical analyzers

- map the kinetic energy and one angle onto a 2D detector (in angular mode)
- are equipped with an entrance slit necessary for a decent resolution
- have energy resolutions typical in the range from a few meV to sub 100meV



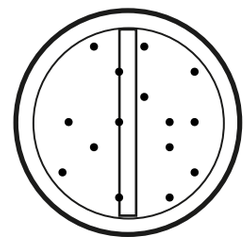
Some electrons do not pass the entrance slit!

Has been the working horse in the past

Principles and Operation of the ArTOF

Modern Time of Flight spectrometers

- **map both angles and the kinetic energy onto a 3D volume (x,y,t)**
- **are slit less which gives a gain in count rate of $\frac{\pi l}{4s}$ as compared to a hemispherical analyzer**

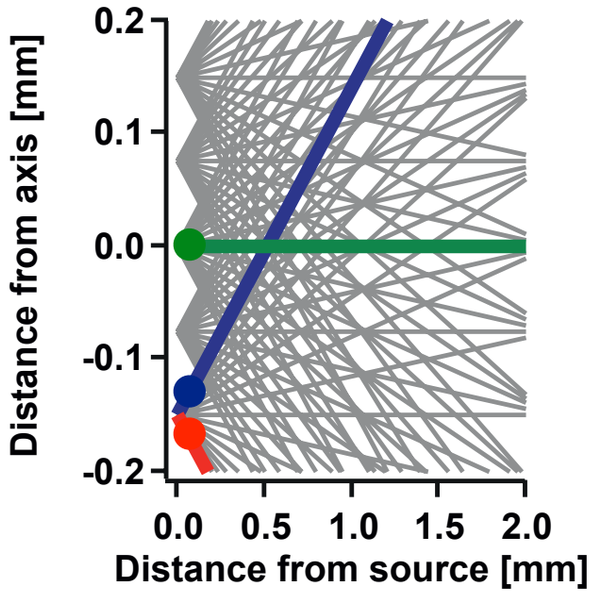


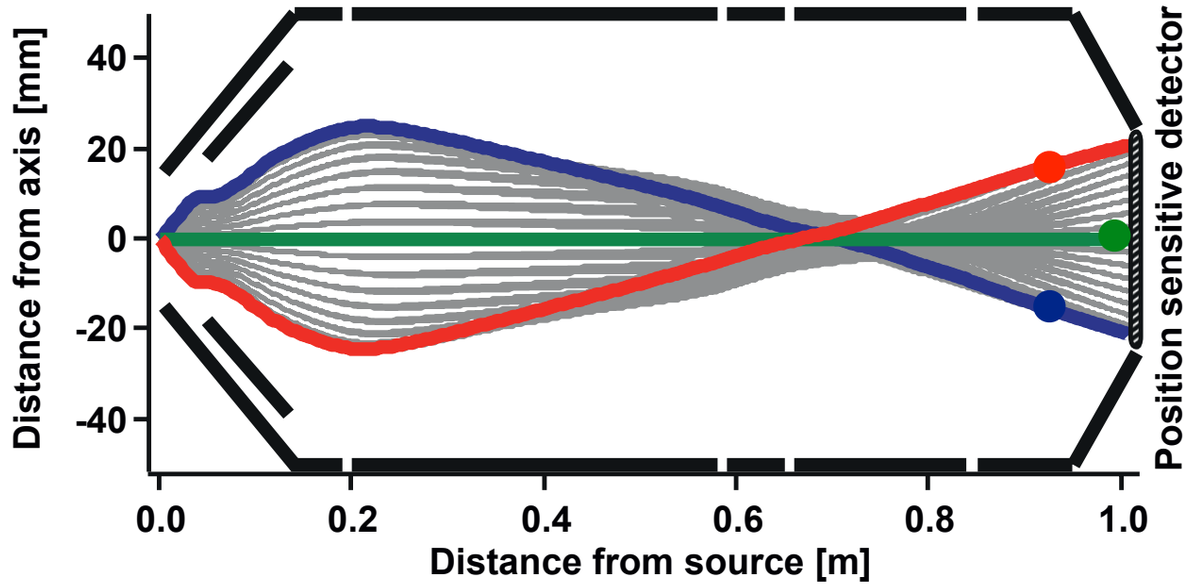
- **have energy resolutions typical in the range from a few meV to 100meV**

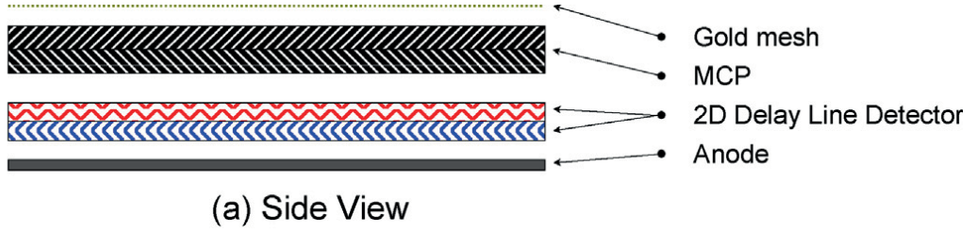
However, they need

- **a good timing resolution and stability (< 200 ps)**
- **a pulsed source, i.e. a synchrotron**
- **a new detector scheme capable of measuring position in 2D and flight time**
- **sophisticated simulation and analysis software**

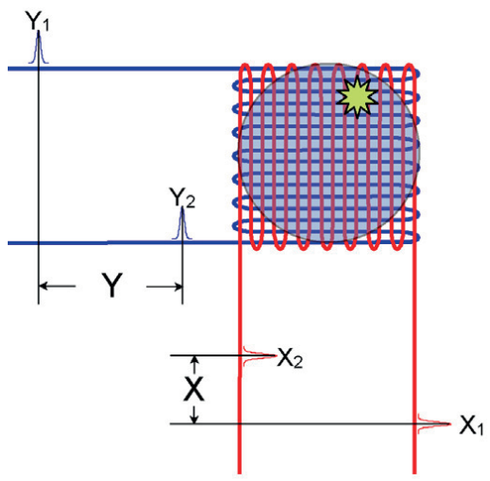
Principles and Operation of the ArTOF







(a) Side View



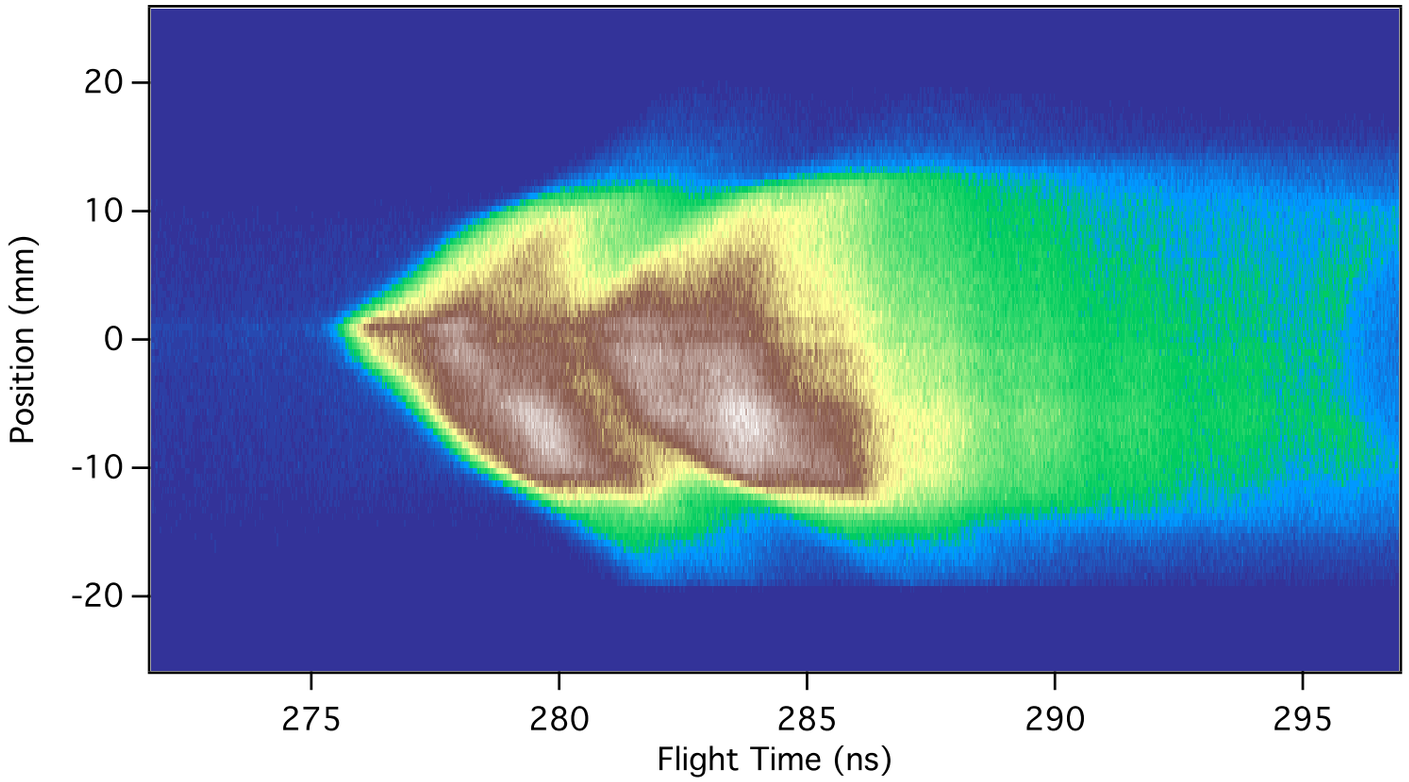
(b) Top View

The time of arrival for a pulse at each end of the wires (Y_1,Y_2,X_1,X_2) is measured. Then the position will be proportional to the difference of arrival time:

$$Y = Y_1 - Y_2$$
$$X = X_1 - X_2$$

Ovsyannikov, R. et al. Journal of Electron Spectroscopy and Related Phenomena, 191, 92–103 (2013).

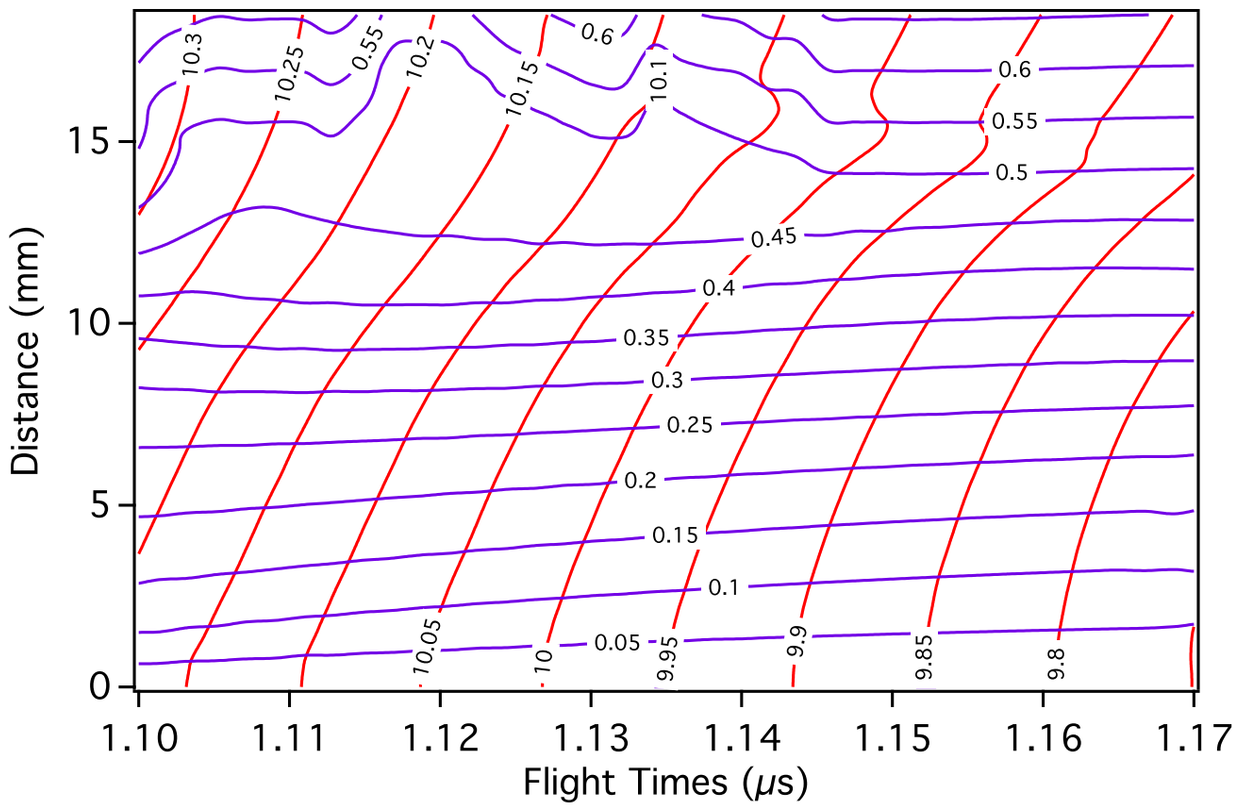
Principles and Operation of the ArTOF



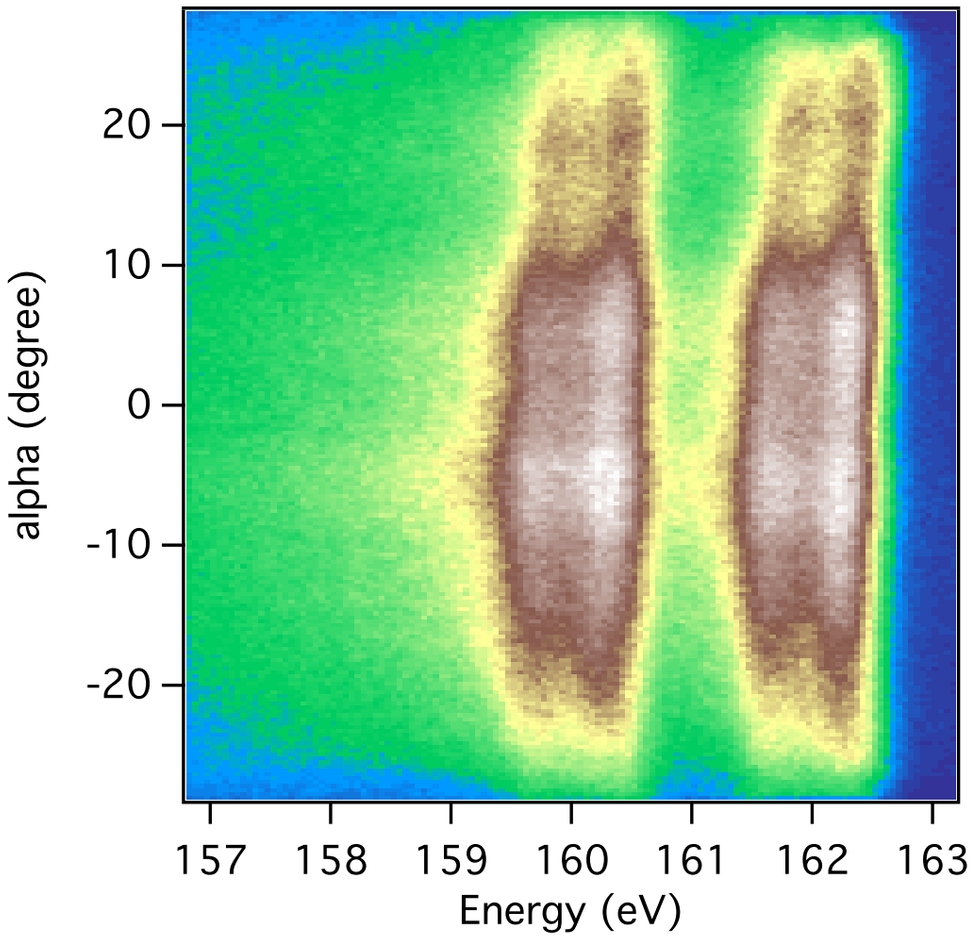
Transformation Matrix

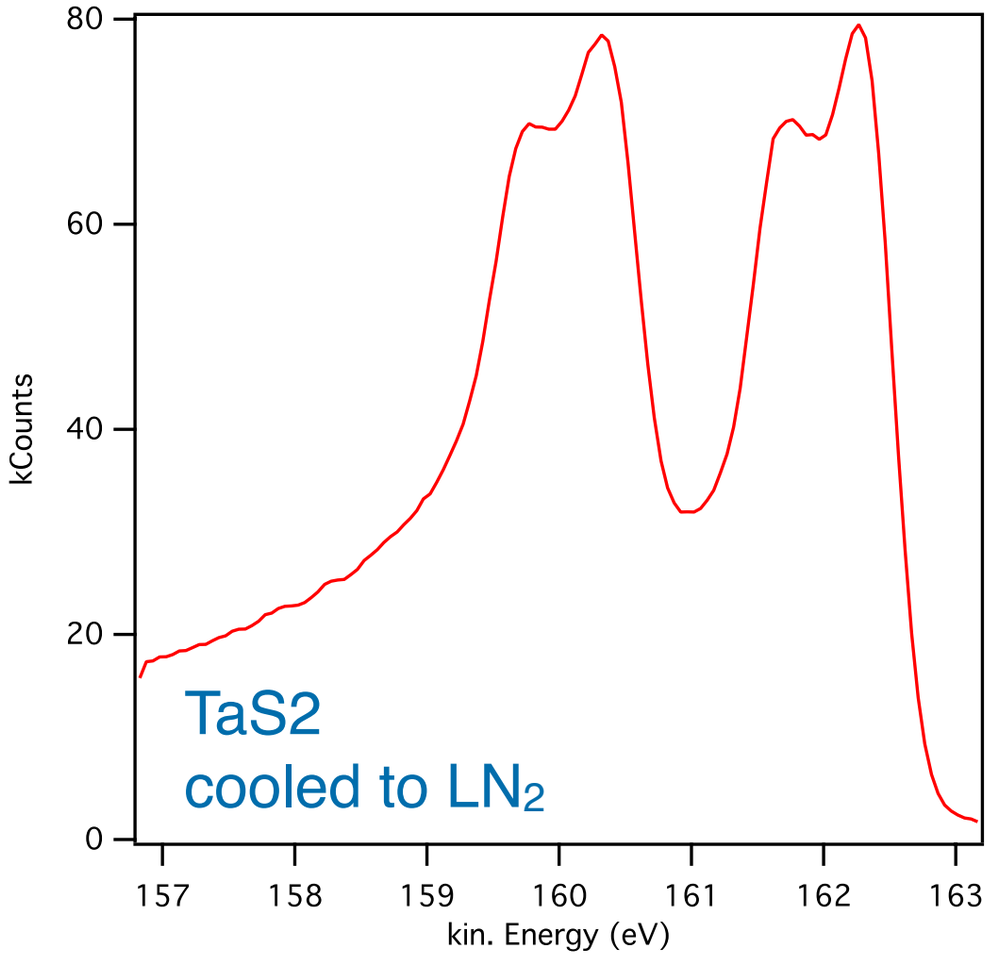
$$\begin{pmatrix} E_k \\ \theta \\ \phi \end{pmatrix} = \mathbf{T} \begin{pmatrix} t \\ x \\ y \end{pmatrix}$$

available from the known trajectories



Principles and Operation of the ArTOF





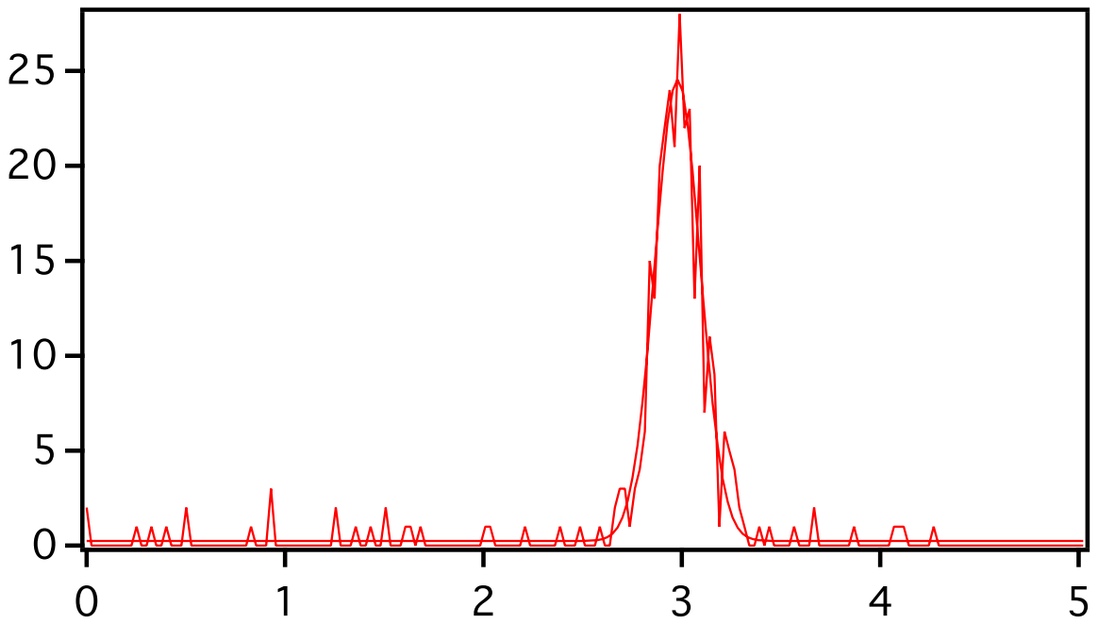
Transformation matrix works only within an

ENERGY WINDOW

selected as

FRACTION of selected CENTER ENERGY

Principles and Operation of the ArTOF



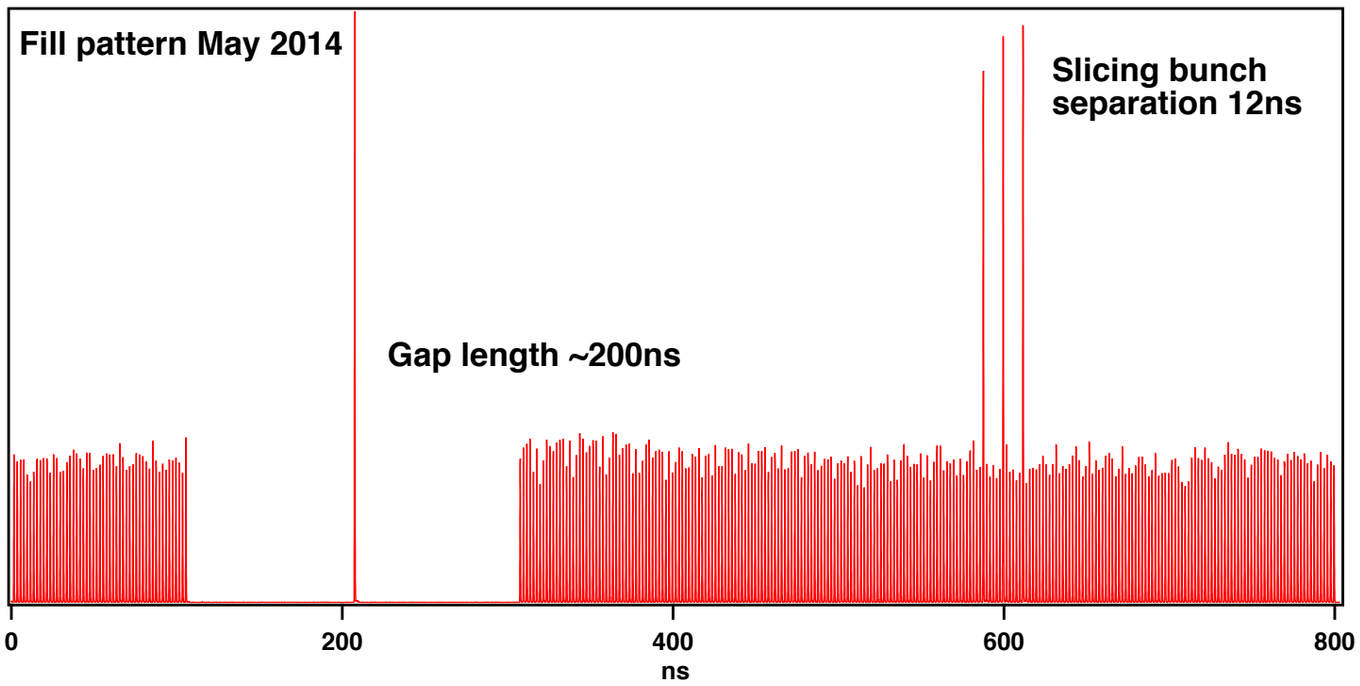
Width of the scattered photon signal at BESSY II ~250ps

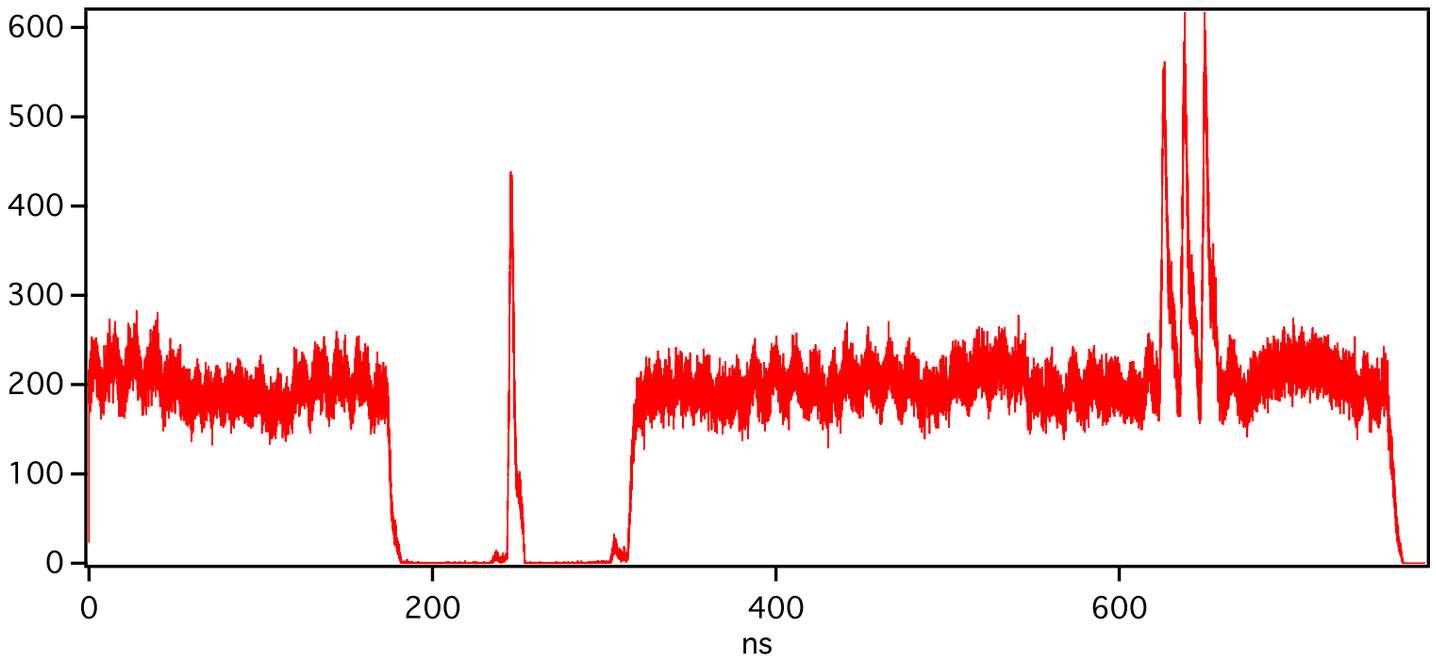
Broadened by the electronics

$$\Delta E = \sqrt{\left(\alpha E^{\frac{3}{2}} \Delta t\right)^2 + \left(\beta \Delta d^{\gamma} E\right)^2}$$

The resolution is related to the transformation matrix, the time resolution and the detector resolution

Pulse Picking at BESSY II





Detector can measure every single bunch

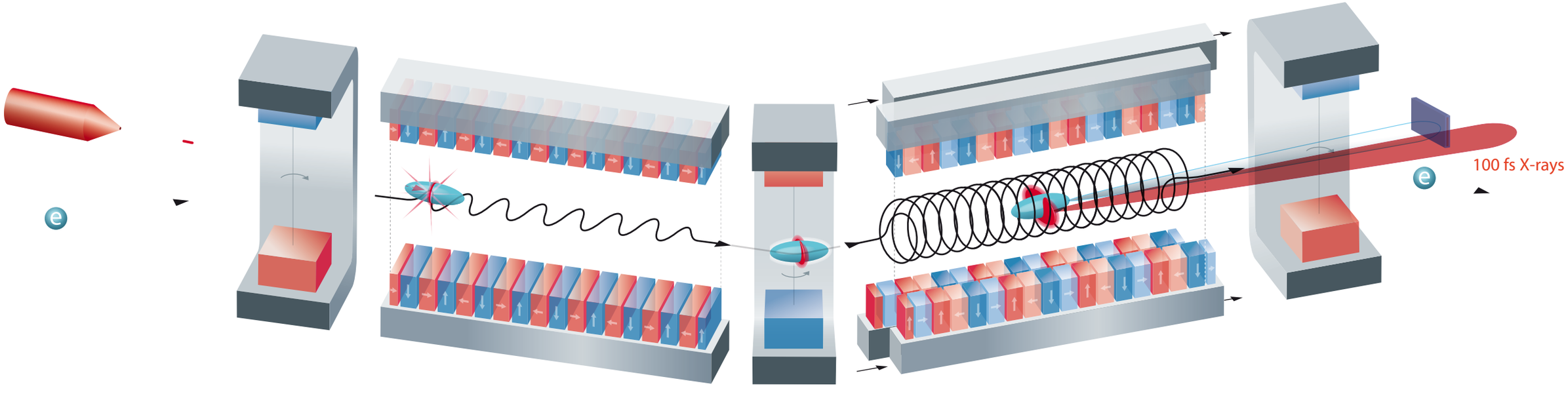
However, due to the ToF only the hybrid bunch is usable.

MB background creates high load on sample and MCP

Need for SB operation or pulse picking methods if one wants to use the ArTOF at a synchrotron

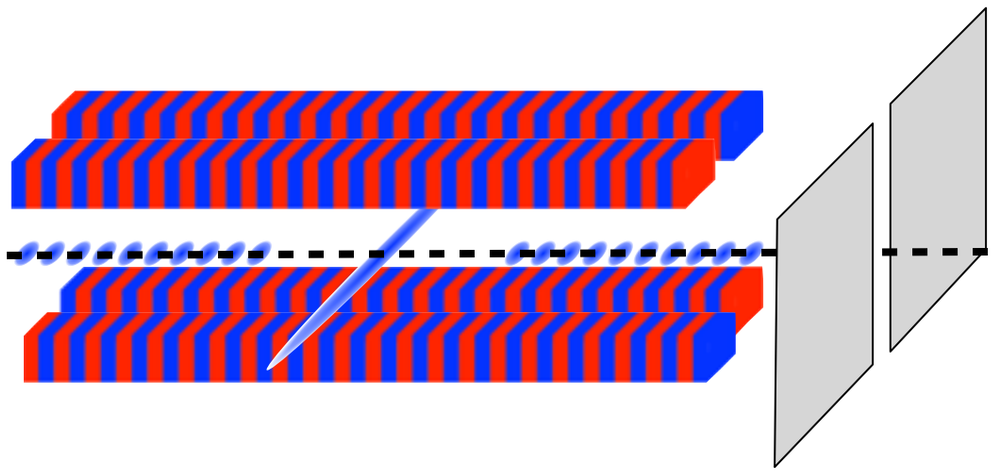
Pulse Picking at BESSY II

Sketch of the Slicing Beamline

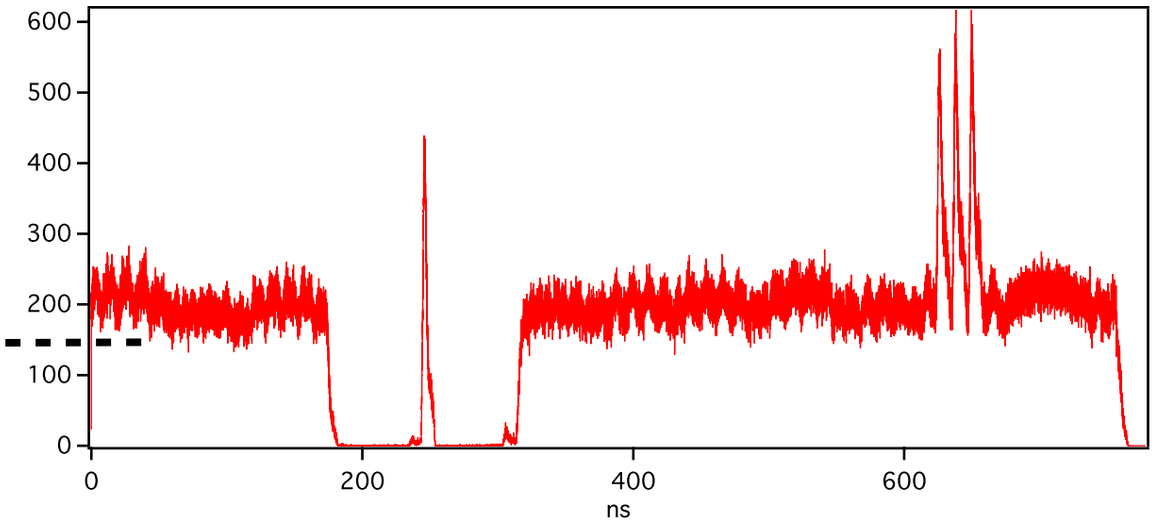


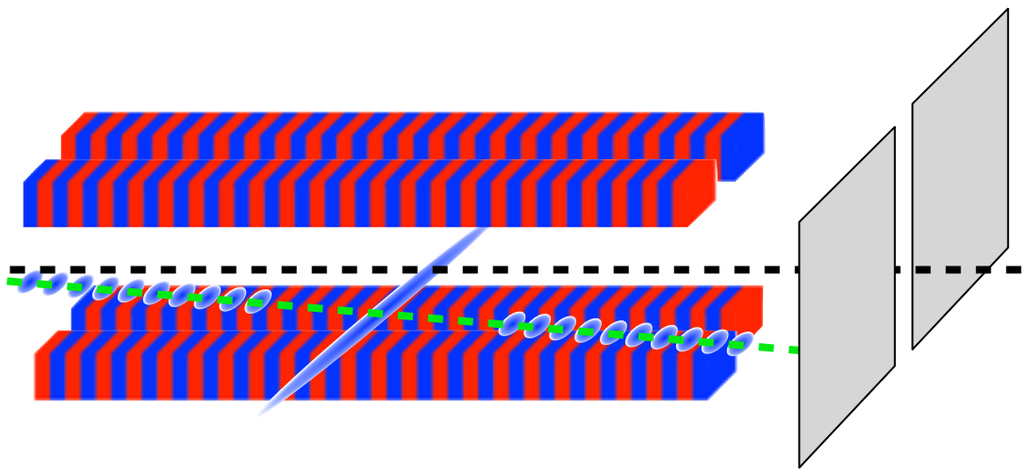
courtesy K. Holldack

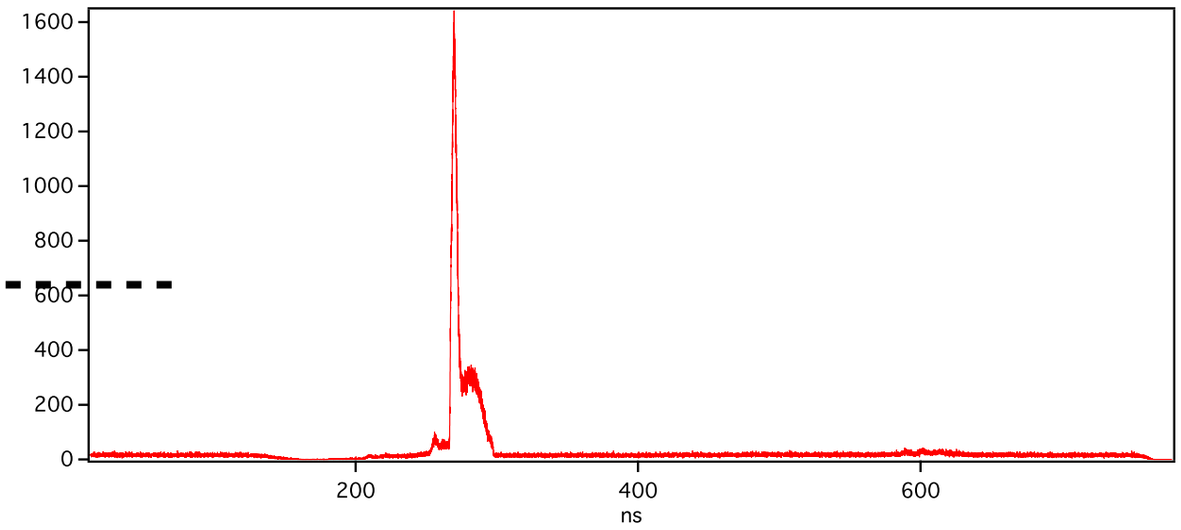
Pulse Picking at BESSY II



Electron Time of Flight measurements



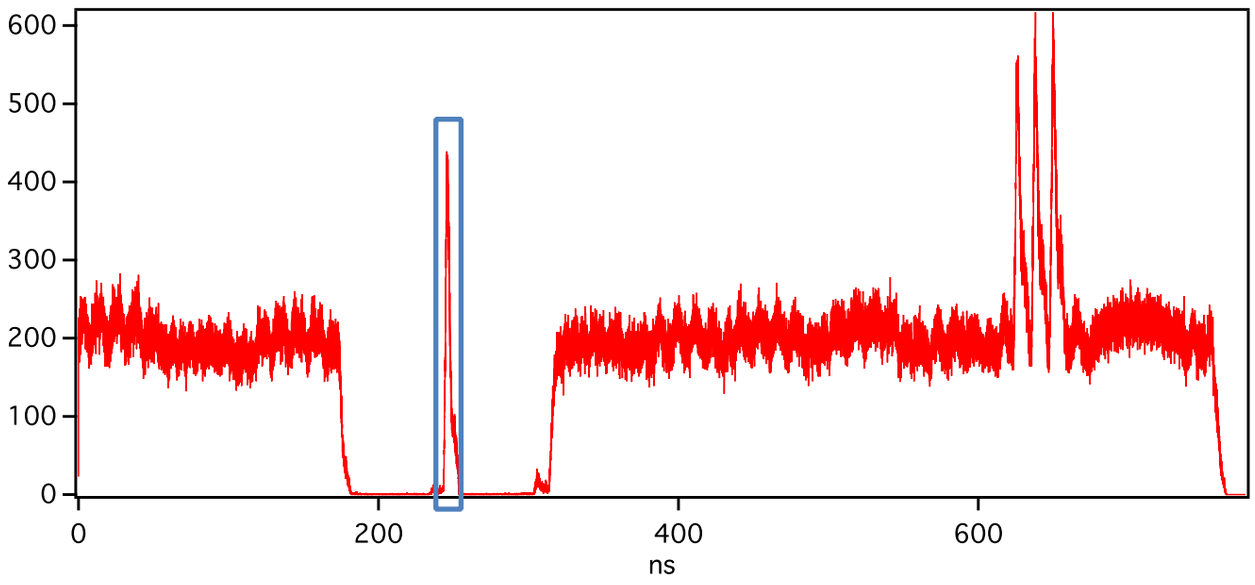




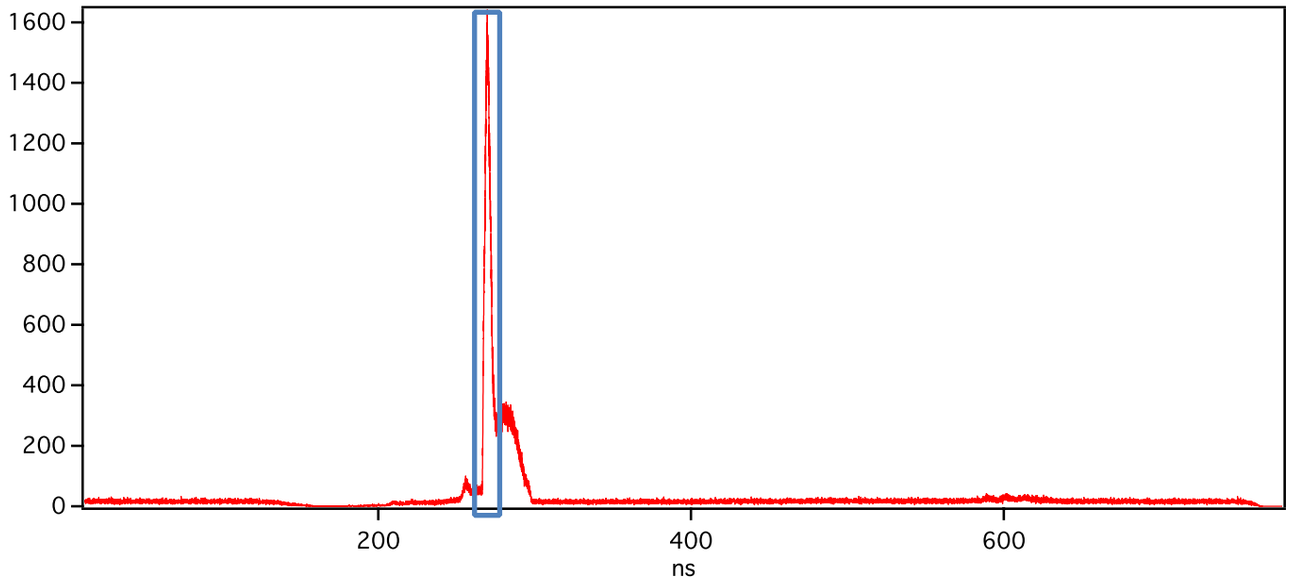
Electron Signal Purity ~100
additional hybrid bunch at the end of the gap

K. Holldack, et al Nat. Commun. 5, 4010 (2014).
Images: courtesy R. Ovsyannikov

Pulse Picking at BESSY II



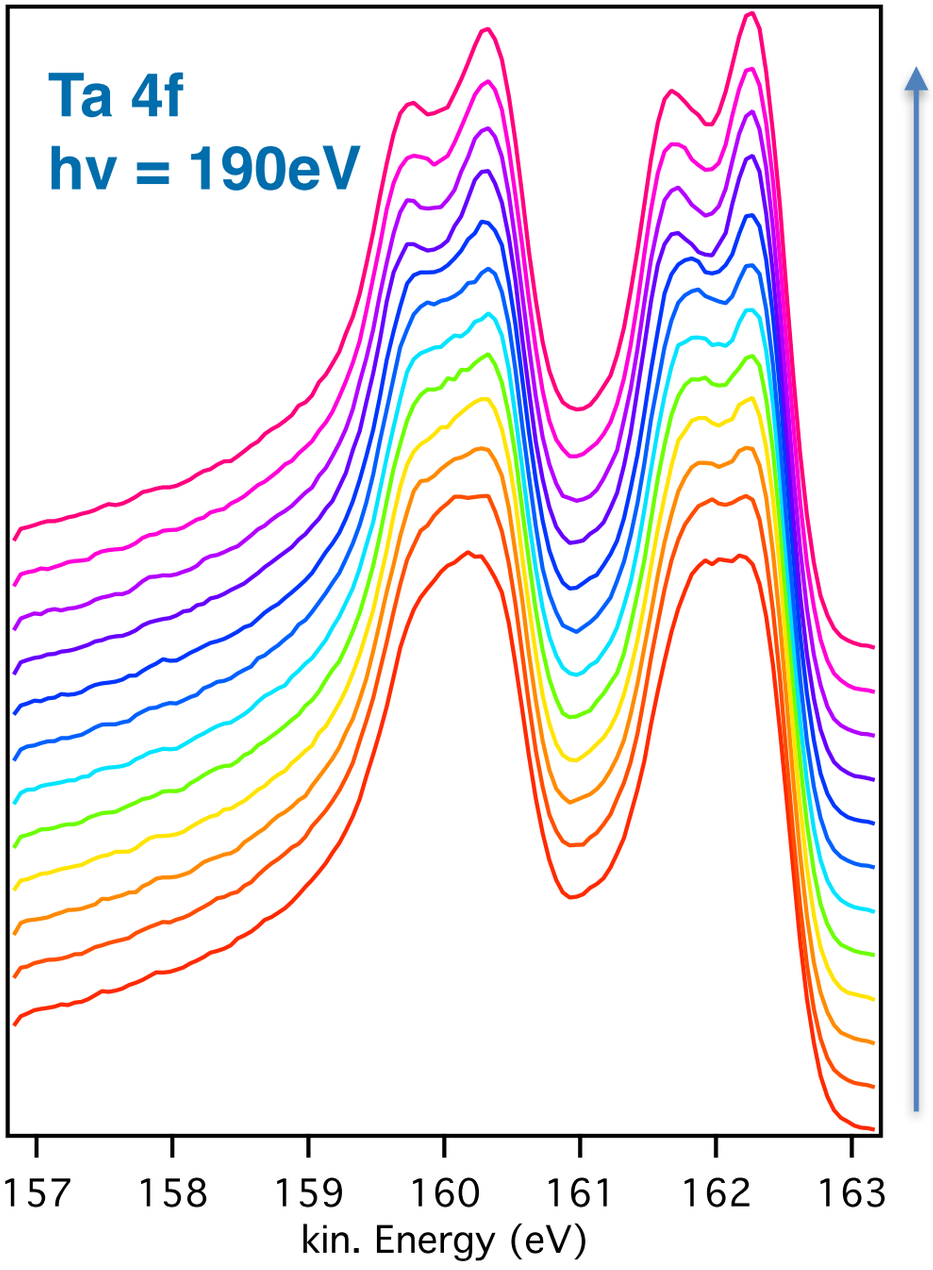
~31kHz count rate full turn
~330Hz usable count rate in the energy window

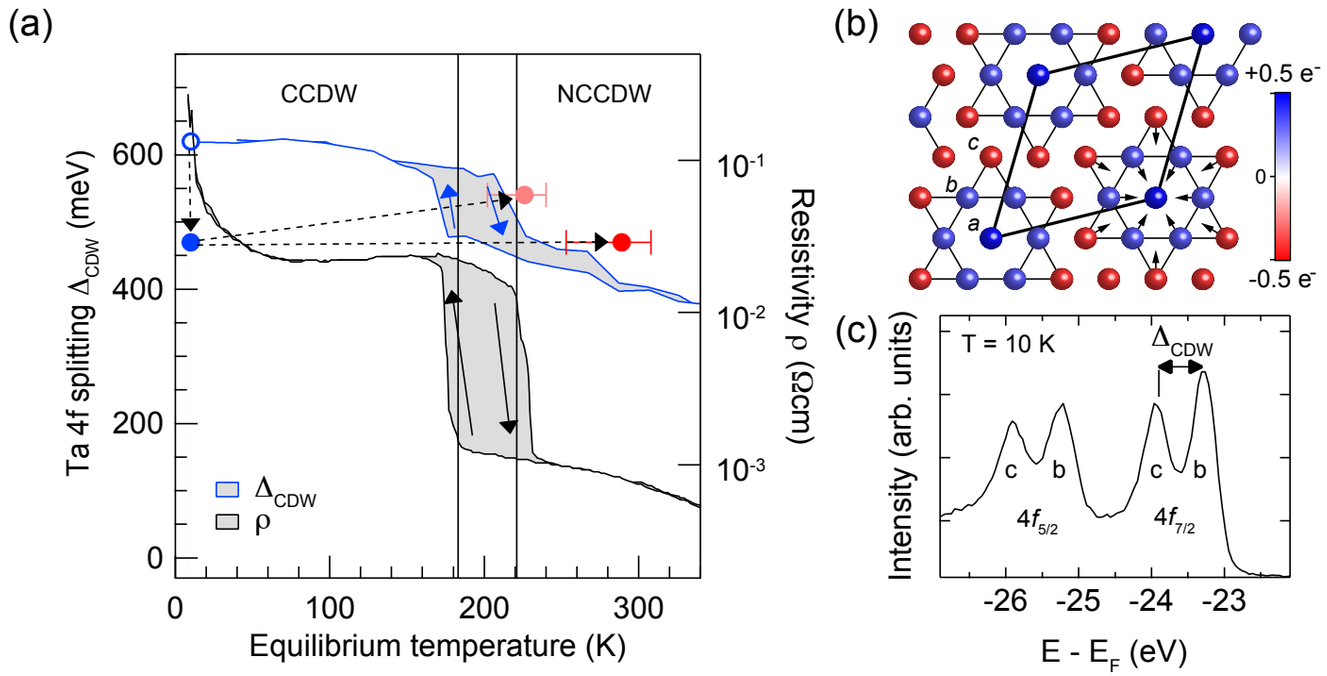


~2.3kHz count rate full turn
~600Hz usable count rate in the energy window

Higher efficiency
protects sample and detector
excellent for samples that demand low dose of photons

ESCA with the model system TaS2



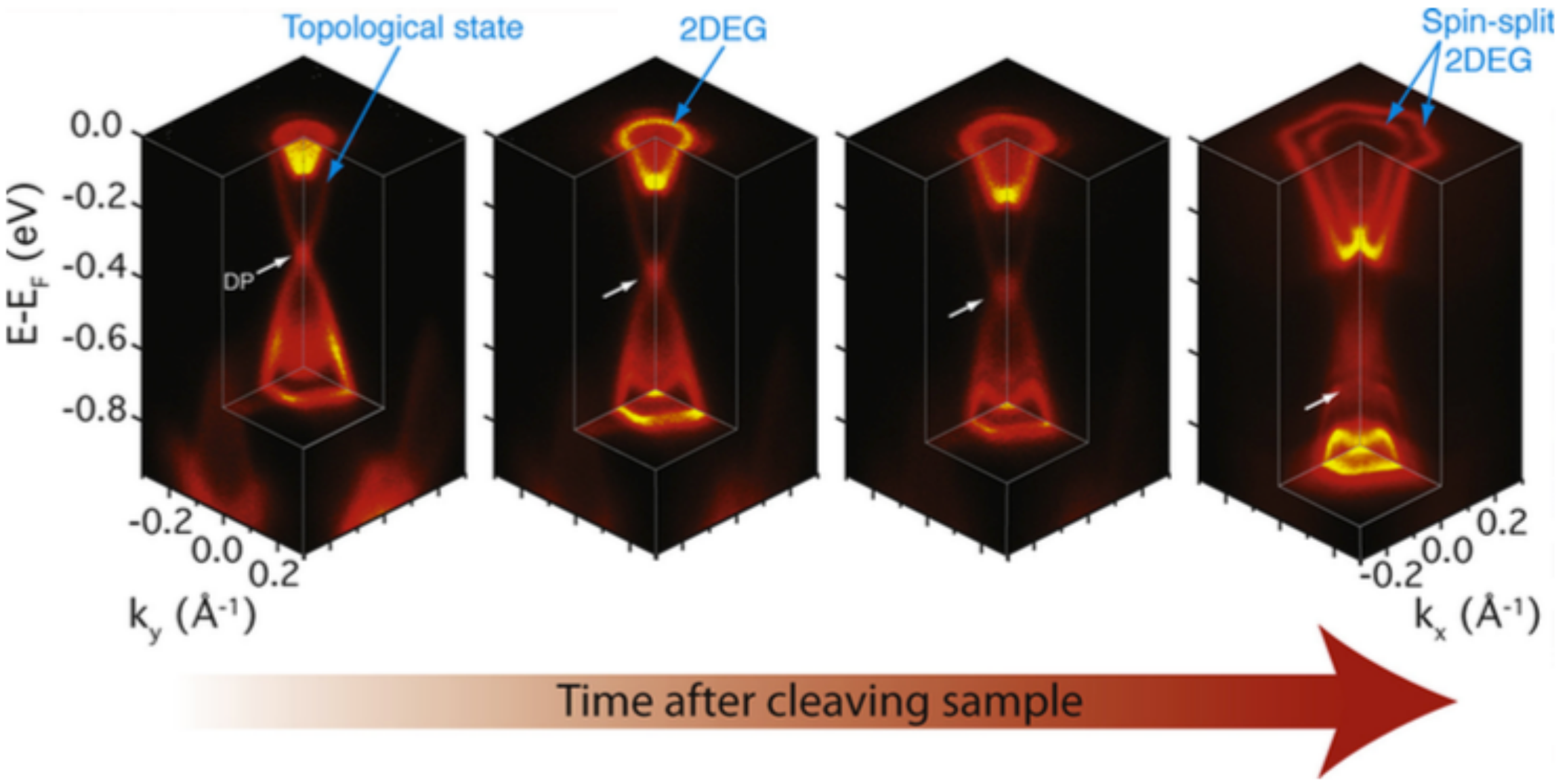


Hellmann, S. et al., New J. Phys. 14, 013062 (2012).

4 minutes steps - slow dynamics

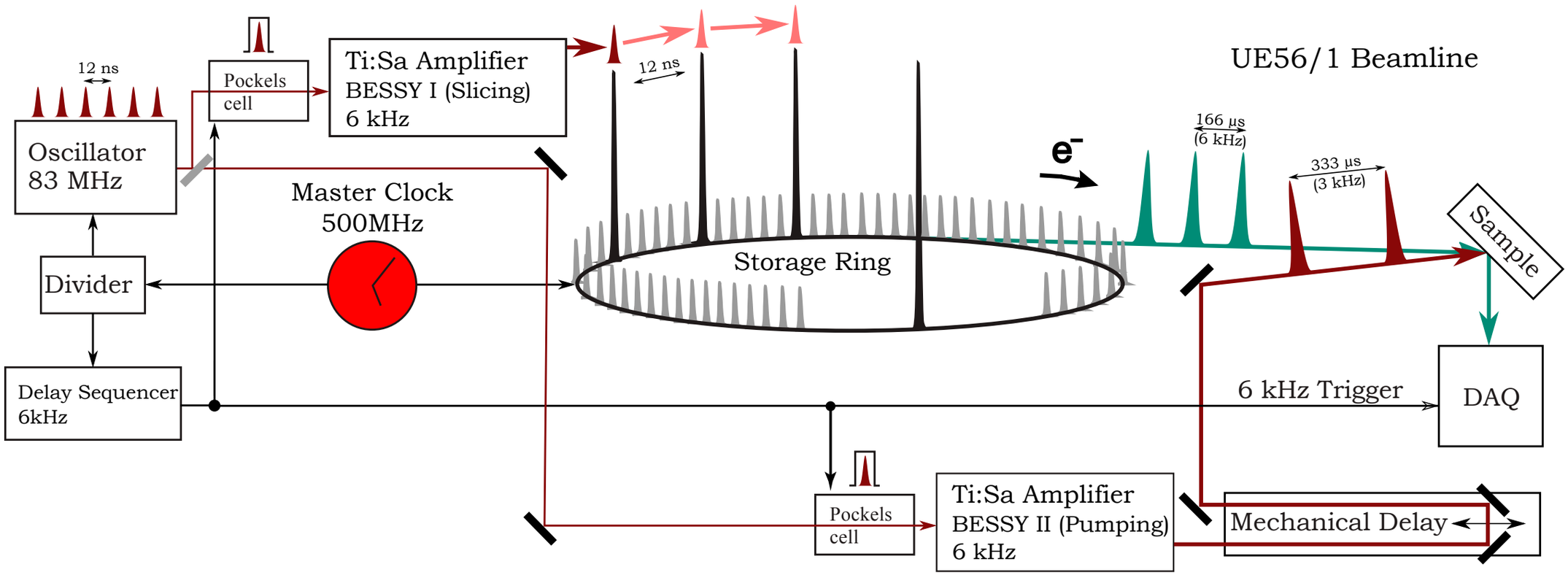
Slow trARPES

Time evolution of the Rashba splitting after cleaving a Bi2 Se3 crystal in vacuum



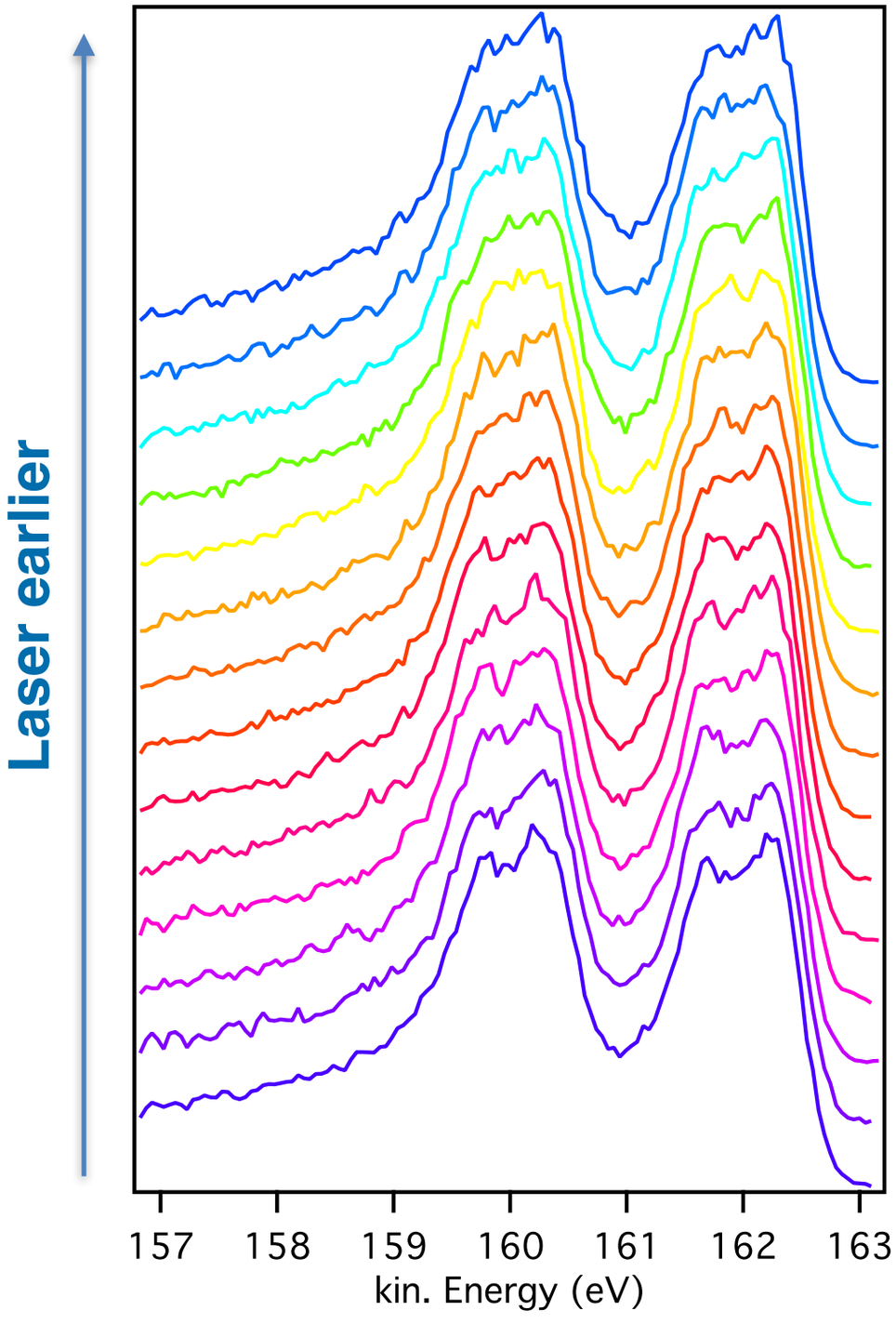
P.D.C. King et. al., Phys. Rev. Lett. 107 (2011) 096802.

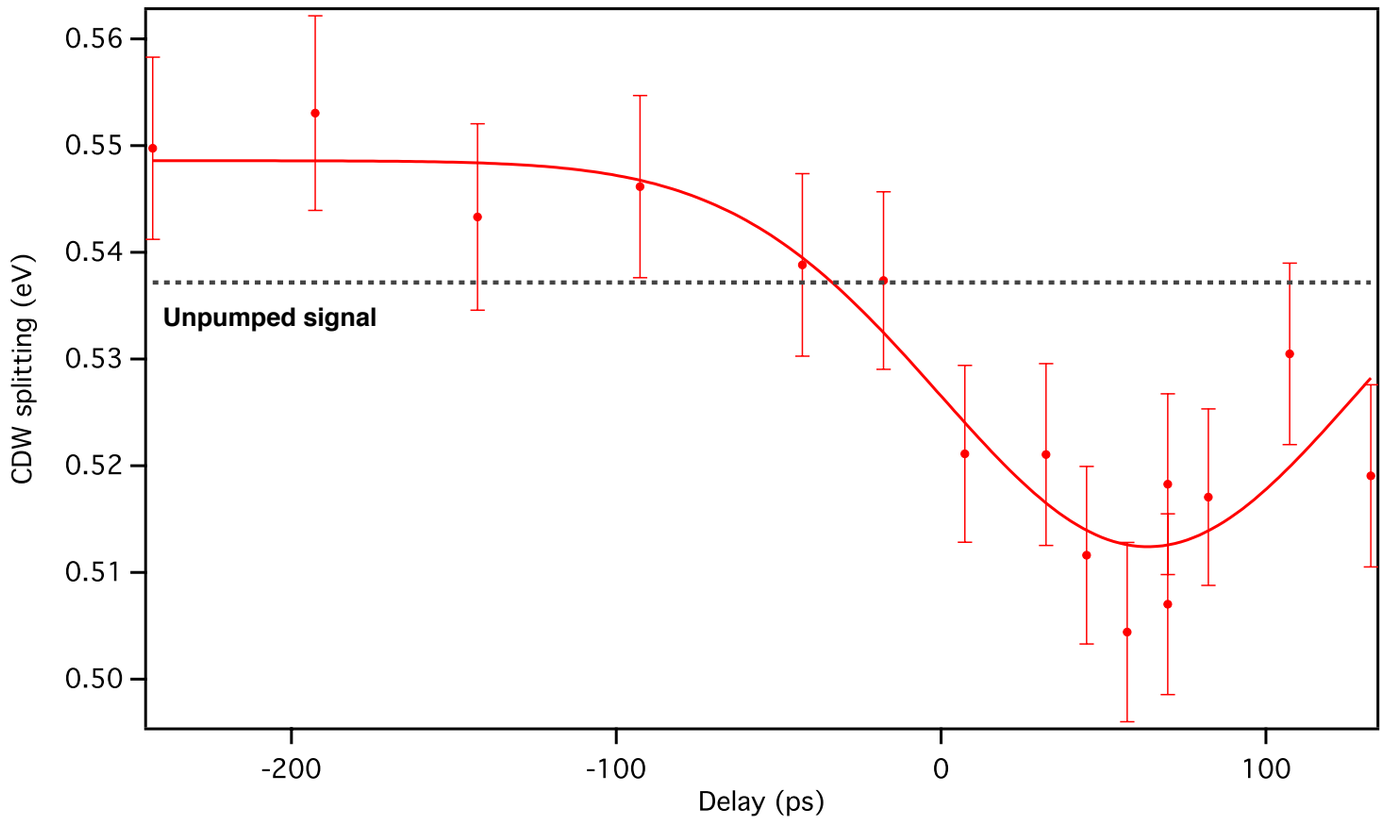
Time-Resolved ESCA at BESSY II



courtesy D. Schick

Time-Resolved ESCA at BESSY II

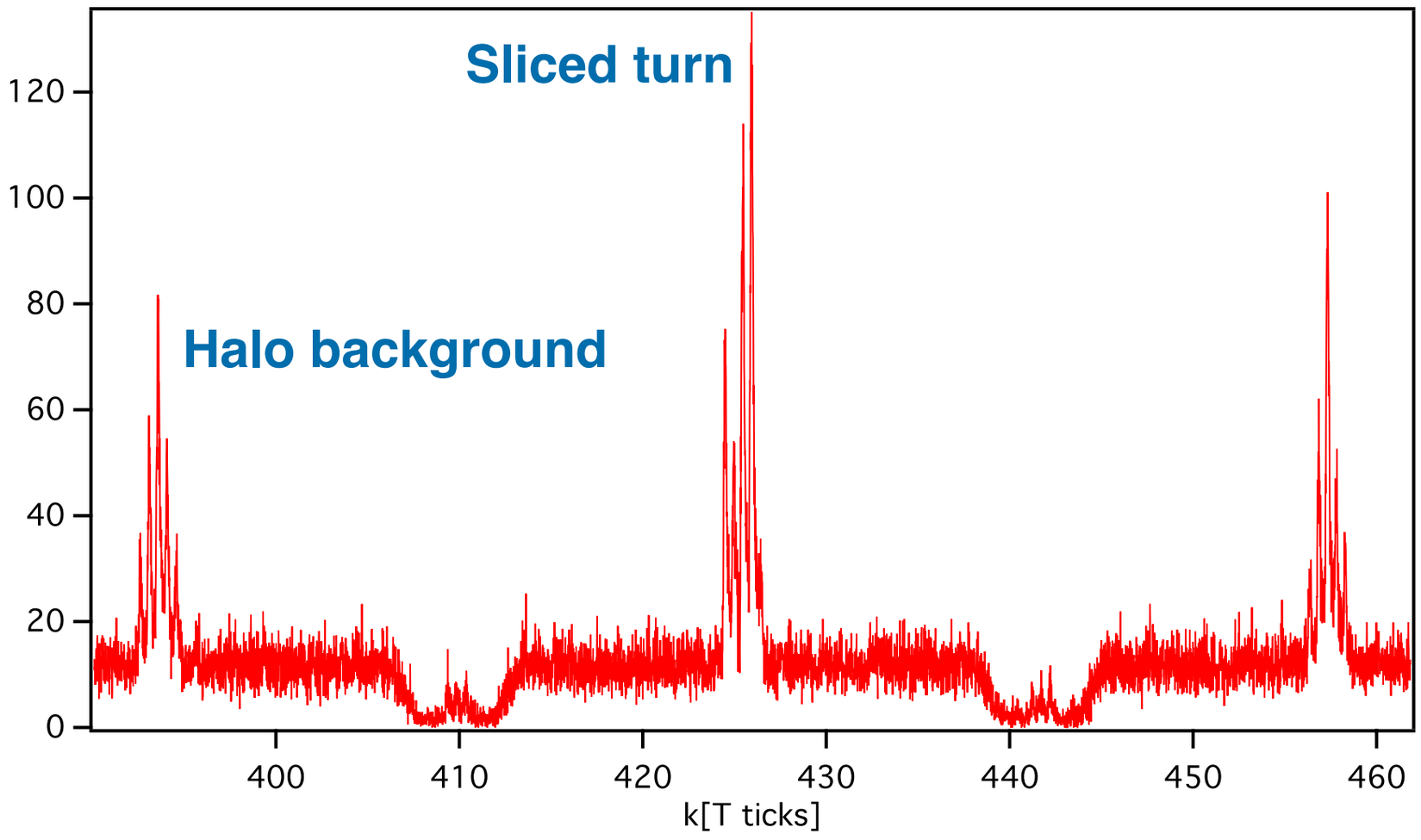




Ta 4f @ LN_2
hv = 190eV
150mW 800nm Laser
6kHz, SB operation
1200l/mm grating
~10min acquisition

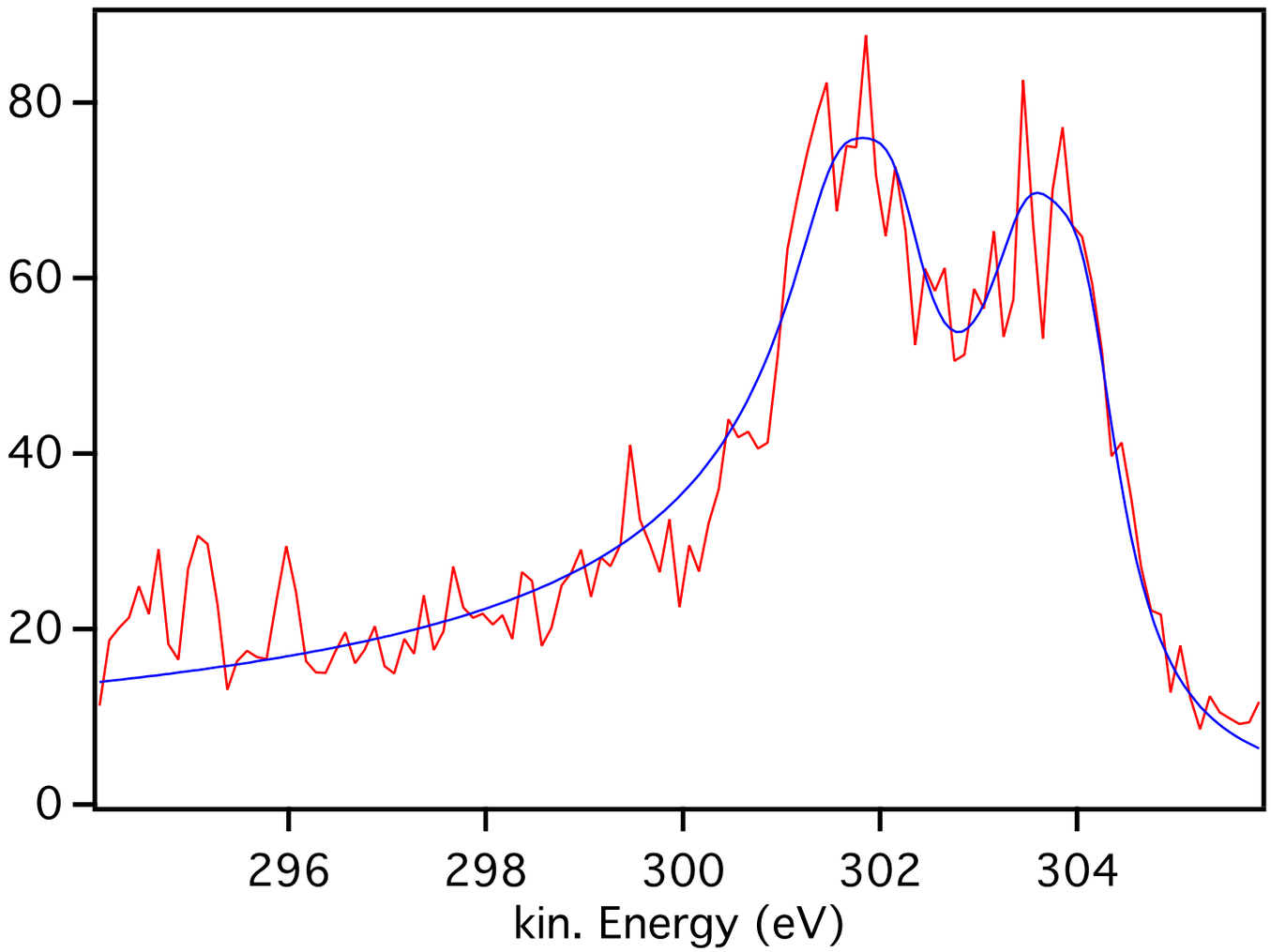
fs Time-Resolved ESCA at BESSY II?

It is very dark! But the ArTOF can help us here since the new 60° lens accepts ~0.84sr (compared to ~0.21 of the 30° lens)



We had better slice to halo ratios in the past - still work to be done

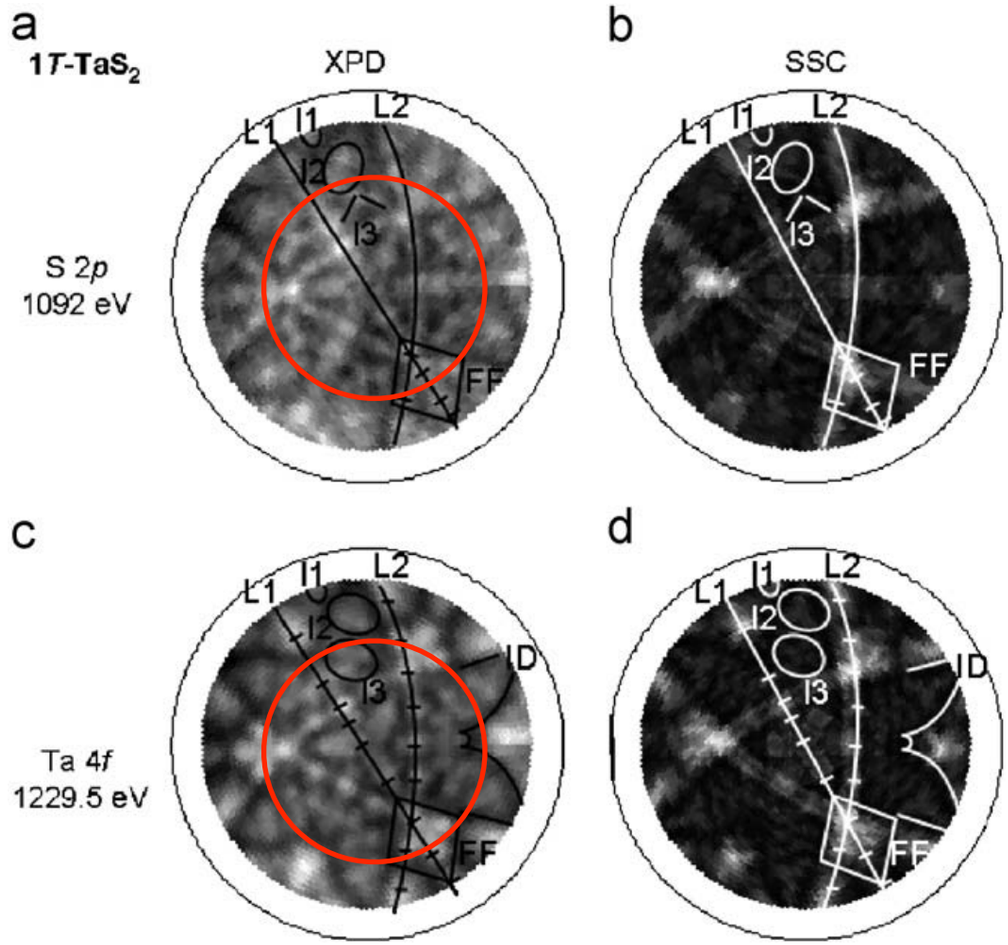
fs Time-Resolved ESCA at BESSY II?



Ta 4f @ RT
hv = 330eV
6kHz Slicing
150l/mm grating
2h40min acquisition

Fit gives a CDW splitting of ~550meV which is reasonable at RT
But there is also the Halo inside!

Further Perspectives using an ArTOF - (tr?)PED



XPD Ta 4f and S 2p @ RT
hv = 1253.6eV (Mg Kα)
5500 angular settings

Red circle marks roughly the acceptance of the 60° ArTOF

=> ~1 - 6 measurements sufficient

Stoltz, D. & Stoltz, S. E.,
Physica B: Condensed Matter 398, 172–177 (2007).

Conclusion

- The 60° ArTOF lens is working (still some improvements to be done)
- SB and/or pulse picking operation mode is needed for such an experiment
- relatively short pulses are needed for the energy resolution
- Due to the high acceptance also (fs) time-resolved studies are in reach
- Photo electron diffraction is a promising application
- New undulatory at Slicing will increase the flux
- BESSY VSR with the shorter pulse will give again a gain in flux

Acknowledgements

- Danilo Kühn, PhD student
- Karsten Holldack and Niko Pontius (Slicing)
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- Ruslan Ovsyannikov (ArTOF)
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- VG Scienta