Spin and magnetization dynamics studied with femtosecond VUV and XUV radiation

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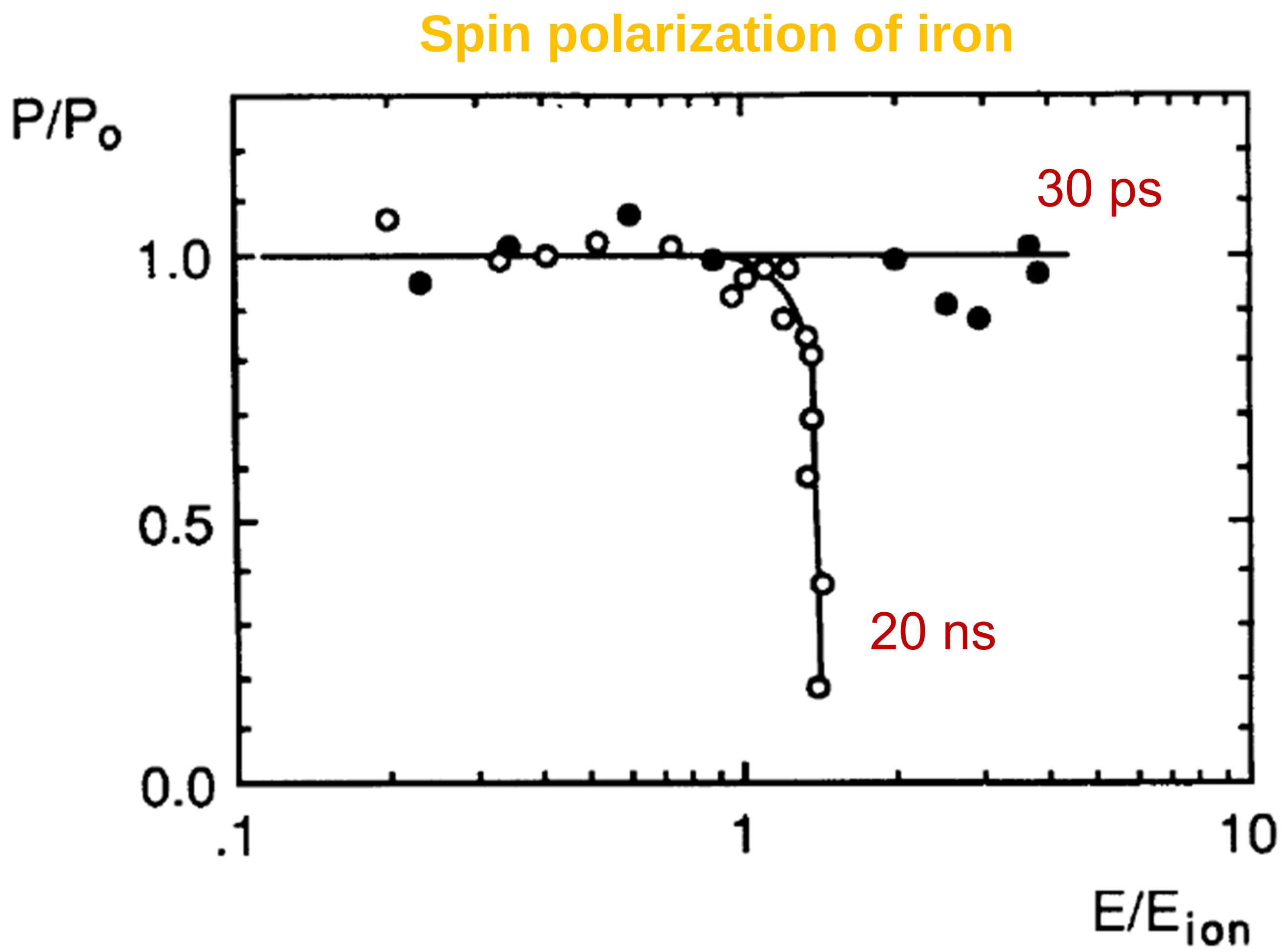
T.A. Ostler et al.,
Nature Communications 3 (2012) 666.

“Ultrafast heating as a sufficient stimulus for magnetization reversal in a ferrimagnet”

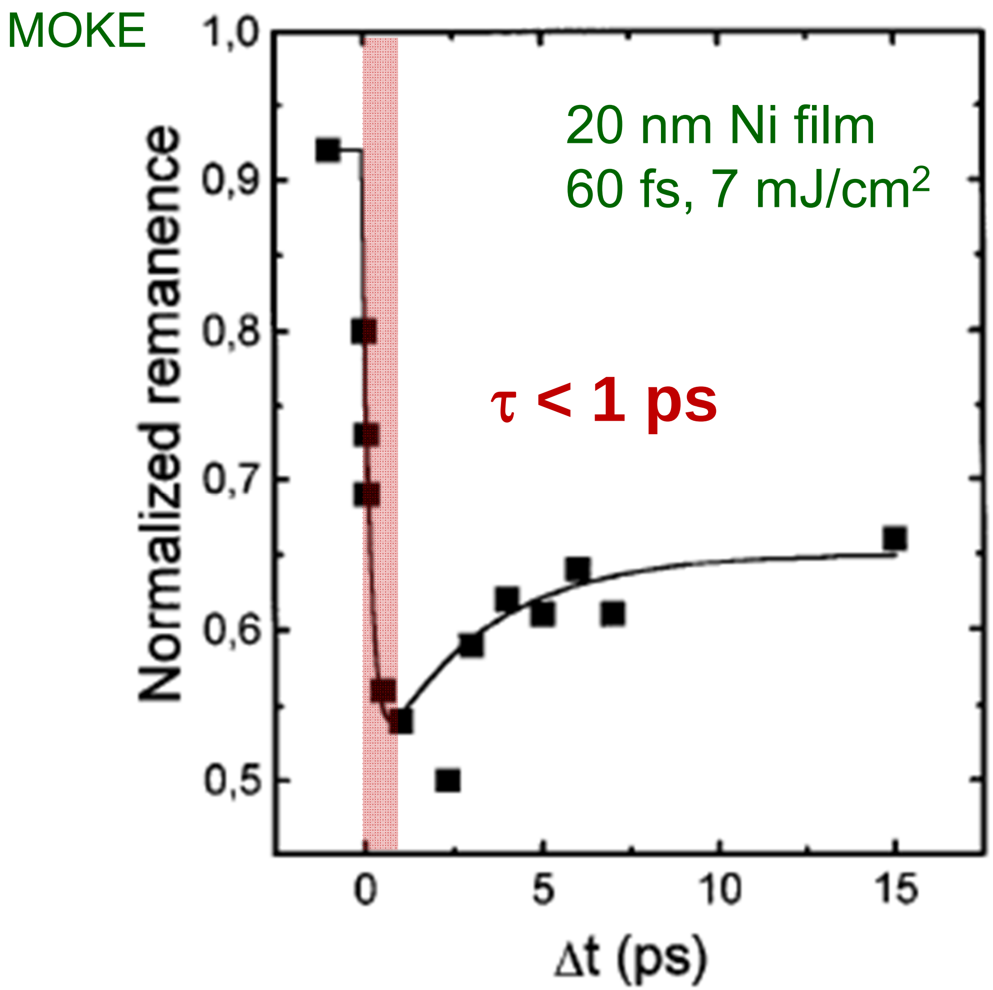
Spin–lattice relaxation time τ_{SL} in iron



τ_{SL} > 30 ps

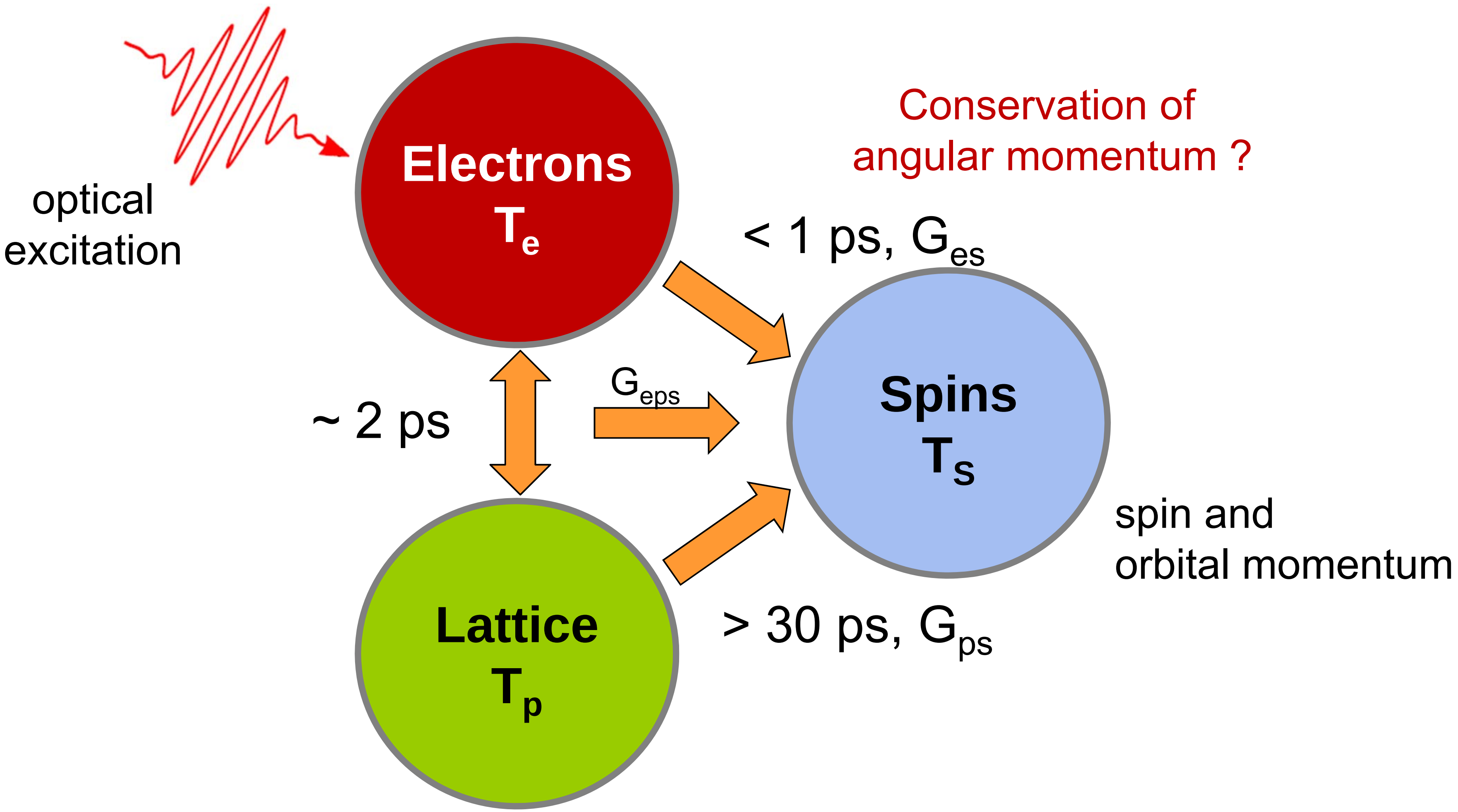
A. Vaterlaus, T. Beutler, D. Guarisco, M. Lutz, and F. Meier, Phys. Rev. B 46 (1992) 5280.

Ultrafast Spin Dynamics in Ferromagnetic Nickel



E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot., Phys. Rev. Lett. ***76*** *(1996) 4250.*

Three Temperature Model



S.I. Anisimov et al., Sov. Phys. JETP ***39****, (1974) 375*
A. Vaterlaus et al., Phys. Rev. Lett. ***67*** *(1991) 3314*
E. Beaurepaire et al.; Phys. Rev. Lett. ***76*** *(1996) 4250*
B. Koopmans et al., Nature Mat. ***9*** *259 (2010) 259*

Ultrafast demagnetization of metallic ferromagnets

„Despite some progress, the results are still being debated, both theoretically and experimentally.”

A. Kirilyuk, A. V. Kimel, and T. Rasing, Ultrafast optical manipulation of magnetic order, Rev. Mod. Phys. 82 (2010) 2731

Outline

- 2 slides on magnetic coupling in Gadolinium

- Magnetic linear dichroism in 4f core-level photoemission
  A. Melnikov et al., Phys. Rev. Lett. ***100****, 107202 (2008).*

- X-ray magnetic circular dichroism at the M_5 edge
  M. Wietstruk et al., Phys. Rev. Lett. 106 (2011) 127401.

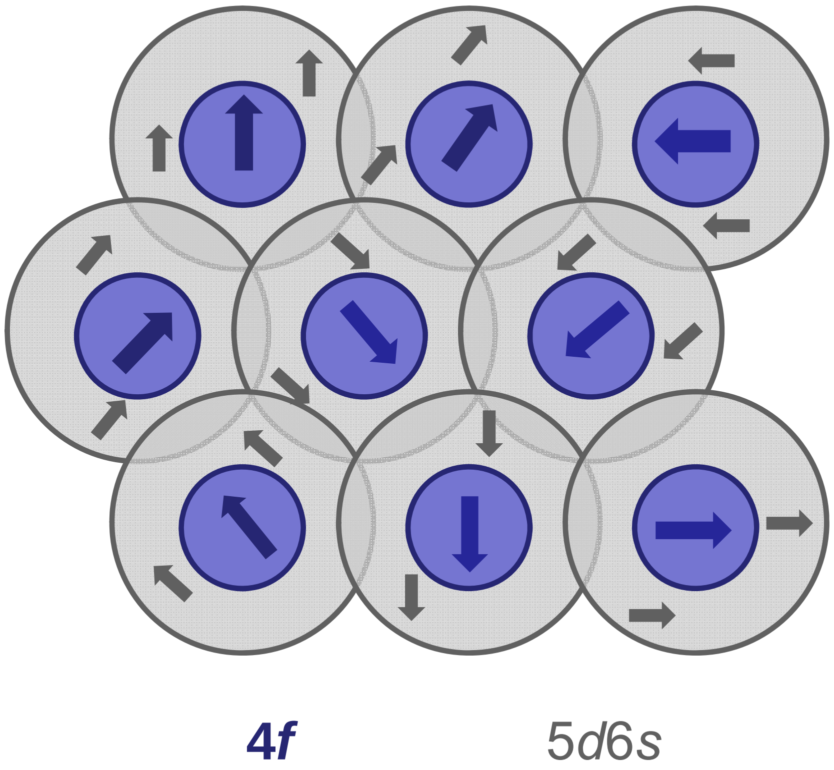
Collaboration with groups of U. Bovensiepen (FU Berlin / Univ. Duisburg-Essen) and H. Dürr (Bessy femtoslicing team, Helmholtz Zentrum Berlin / Standford)

- Transient bandstructure of Gadolinium by time- and angle-resolved photoemission
  R. Carley et al., Phys. Rev. Lett. (2012) in press.

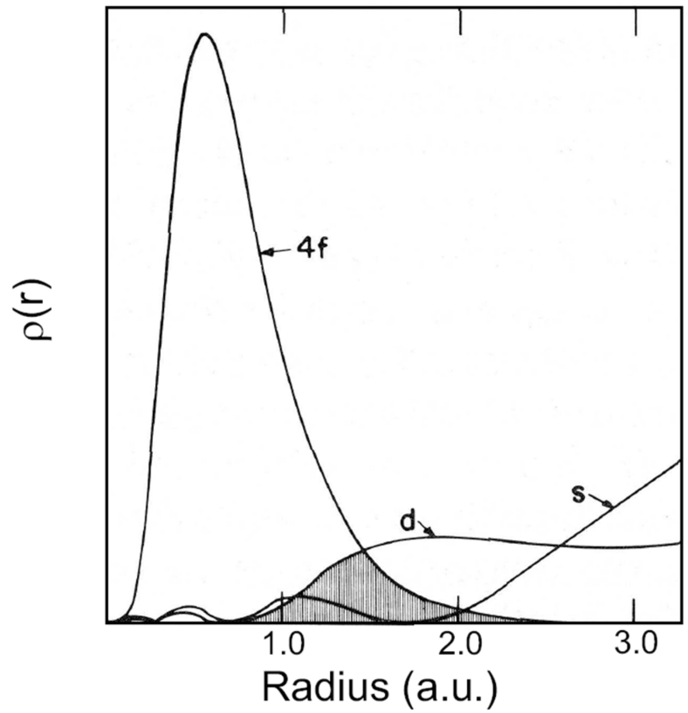
Magnetic coupling in Gadolinium

Gd: [Xe] $4f^7\ 5d^1\ 6s^2$

RKKY interaction

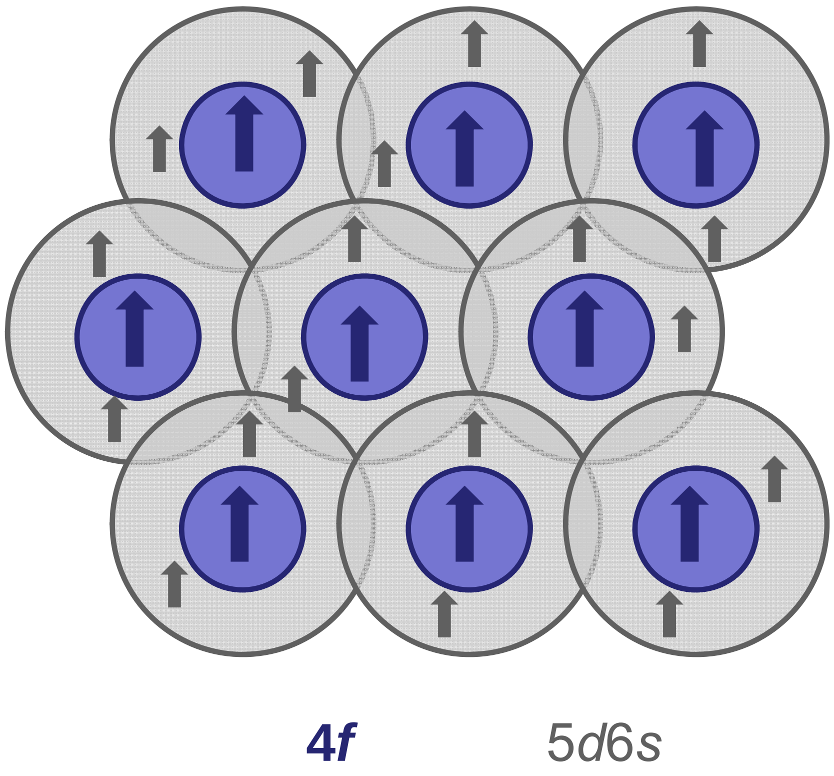


- 4f shell half-filled, $S_f = 7/2$, $L_f = 0$
- localized 4f moments $7\mu_B$
- polarized valence electrons 0.55 μB



Magnetic coupling in Gadolinium

RKKY interaction



- 4f shell half-filled
- localized 4f moments $7\mu_B$
- polarized valence electrons 0.55 μB
- T_C = 293 K
- exchange coupling J ~ 88 meV

R. Ahuja et al., Phys. Rev. B 50 (1994) 5147.

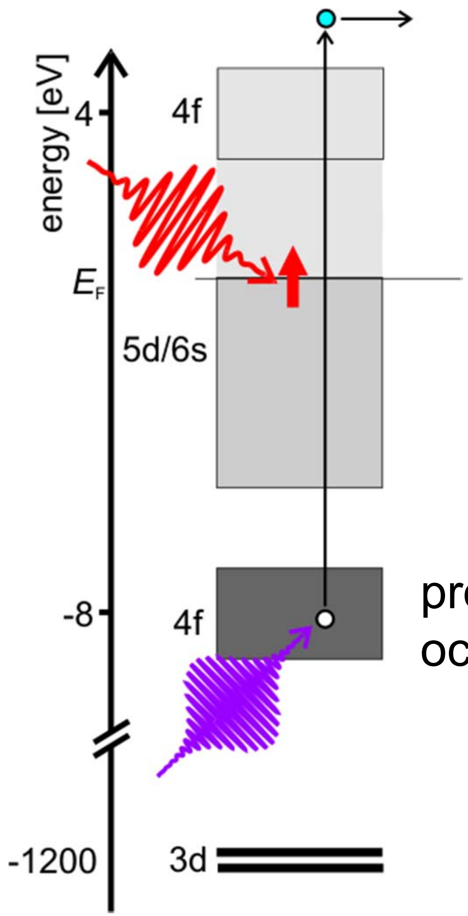
Magnetization dynamics of the two spin subsystems: $4f^7$ and $(5d6s)^3$

A. Vaterlaus, T. Beutler, and F. Meier, Phys. Rev. Lett. ***67*** *(1991) 3314.*

$\tau_{SL} = 100 \pm 80$ ps

$4f^7$ magnetization dynamics

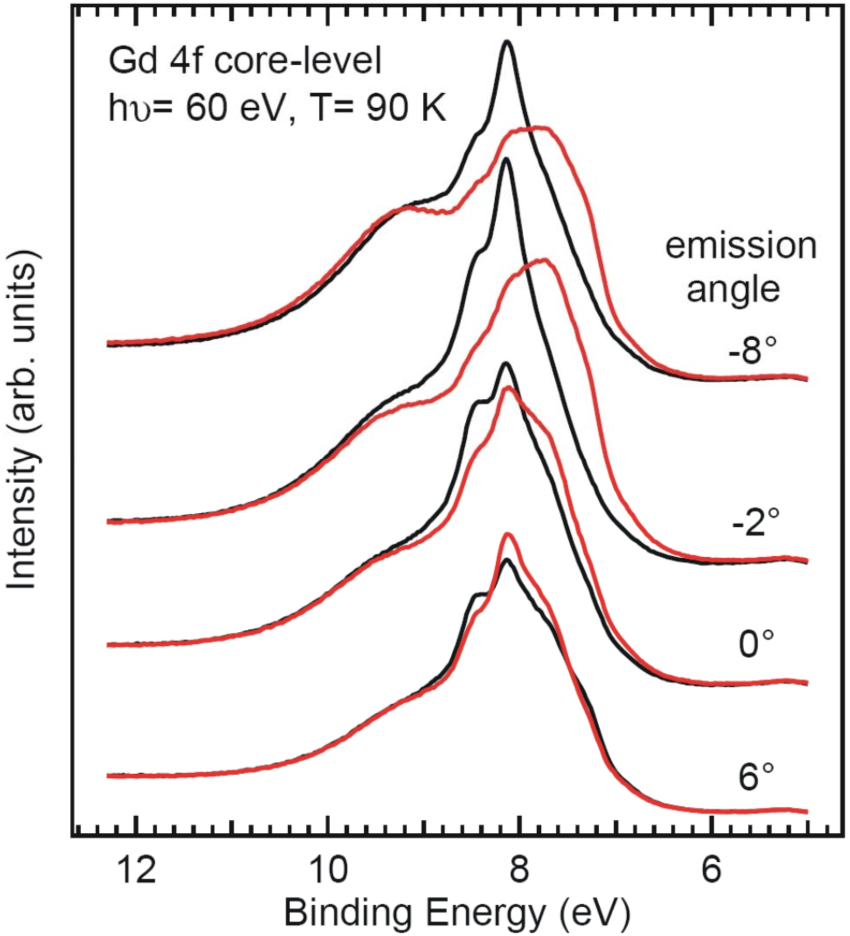
Magnetic linear dichroism (MLD)
in time-resolved photoemission

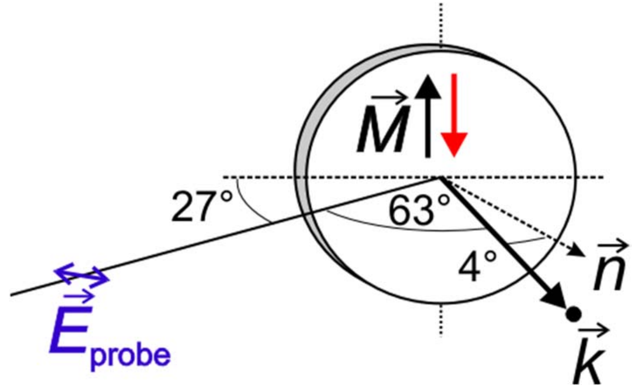


probing
occupied 4f-levels

Magnetic linear dichroism in 4f photoemission

100 Å Gd(0001)/W(110)

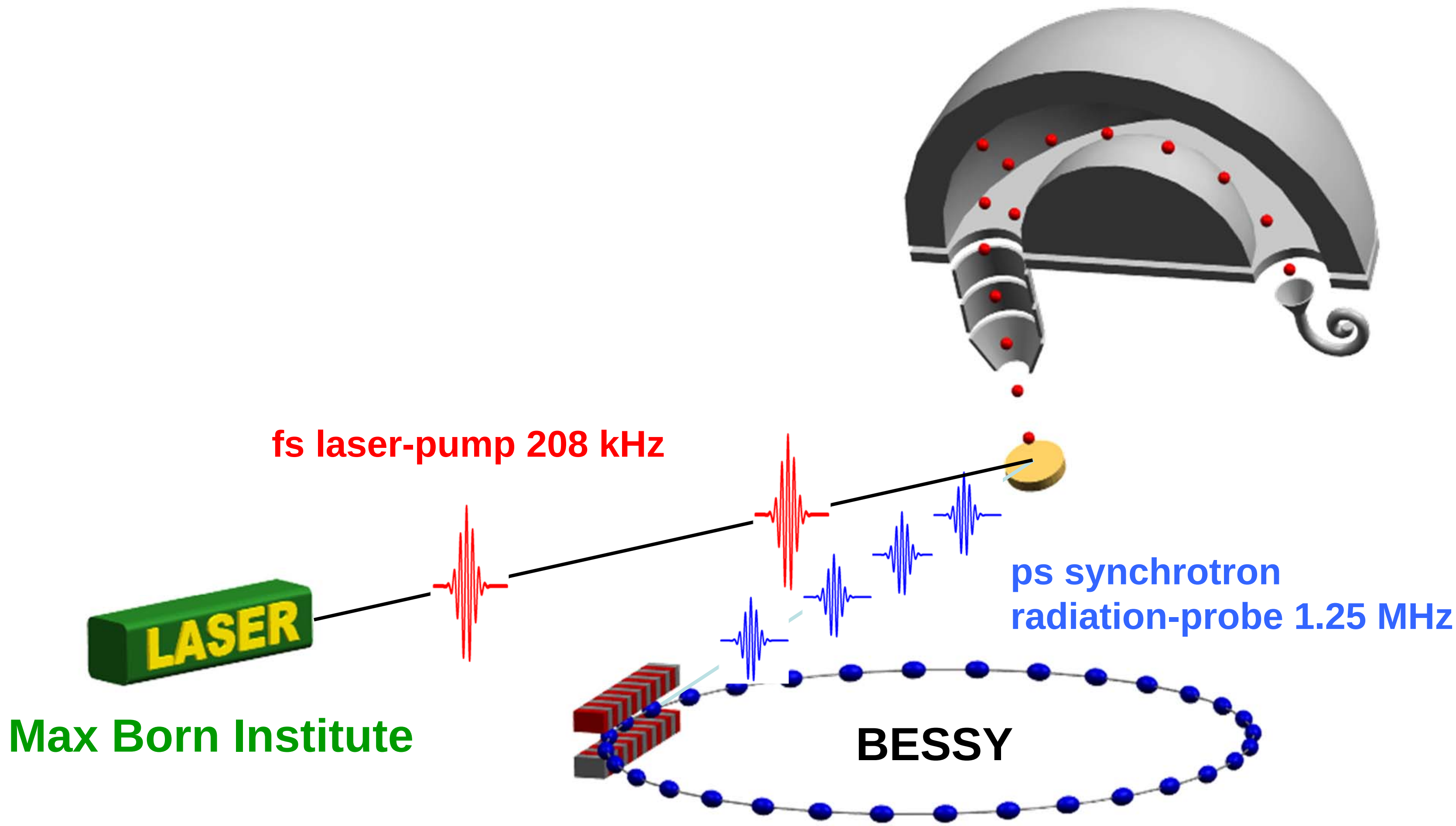




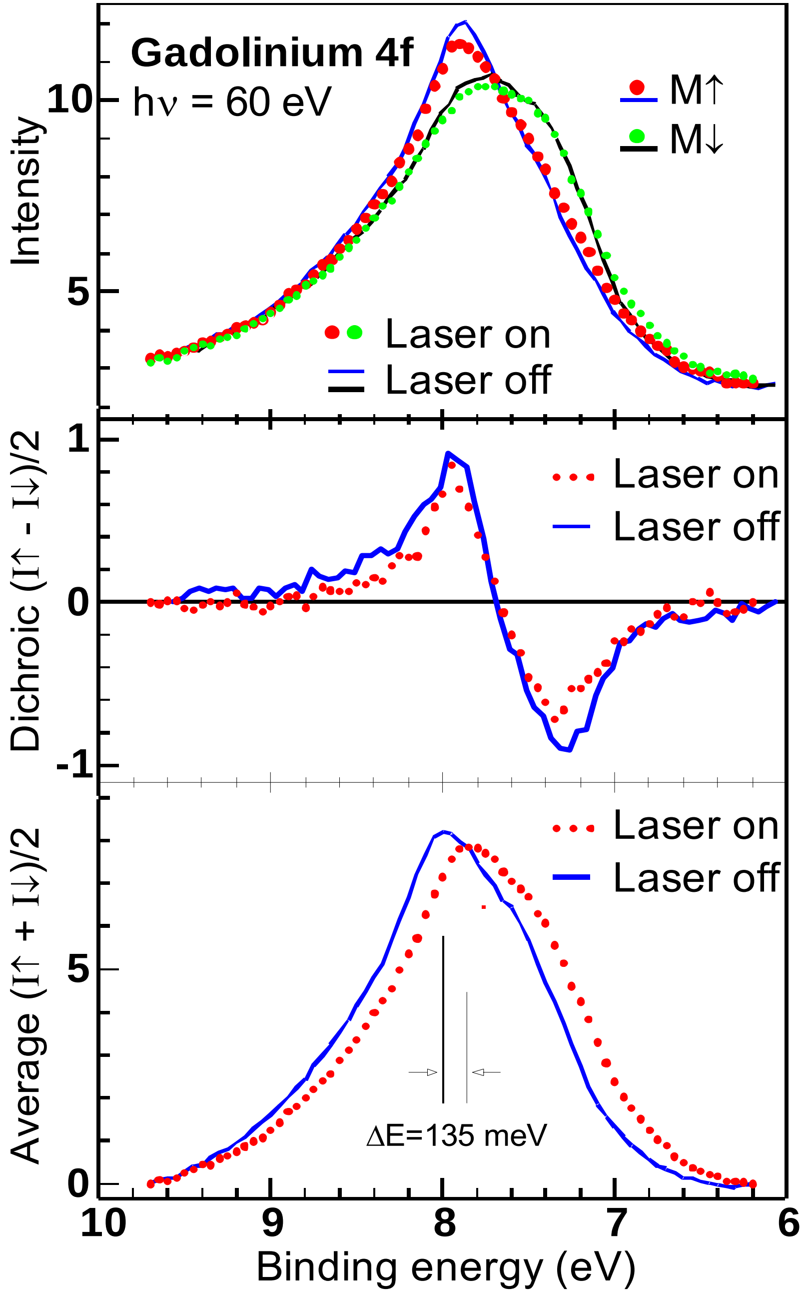
Dichroic contrast:

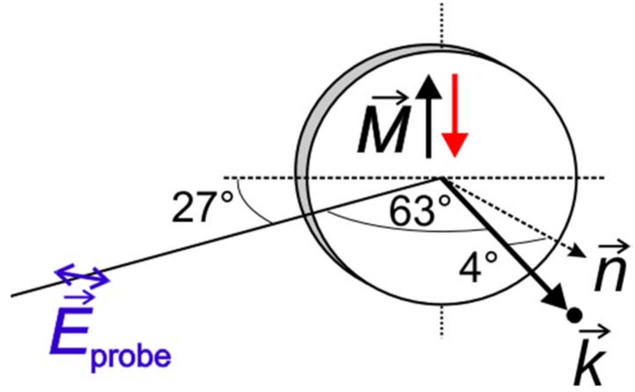
$$\frac{I_{\uparrow} - I_{\downarrow}}{I_{\uparrow} + I_{\downarrow}}$$

Time-resolved photoelectron spectroscopy



Magnetic linear dichroism in x-ray PE from Gd 4f-shell

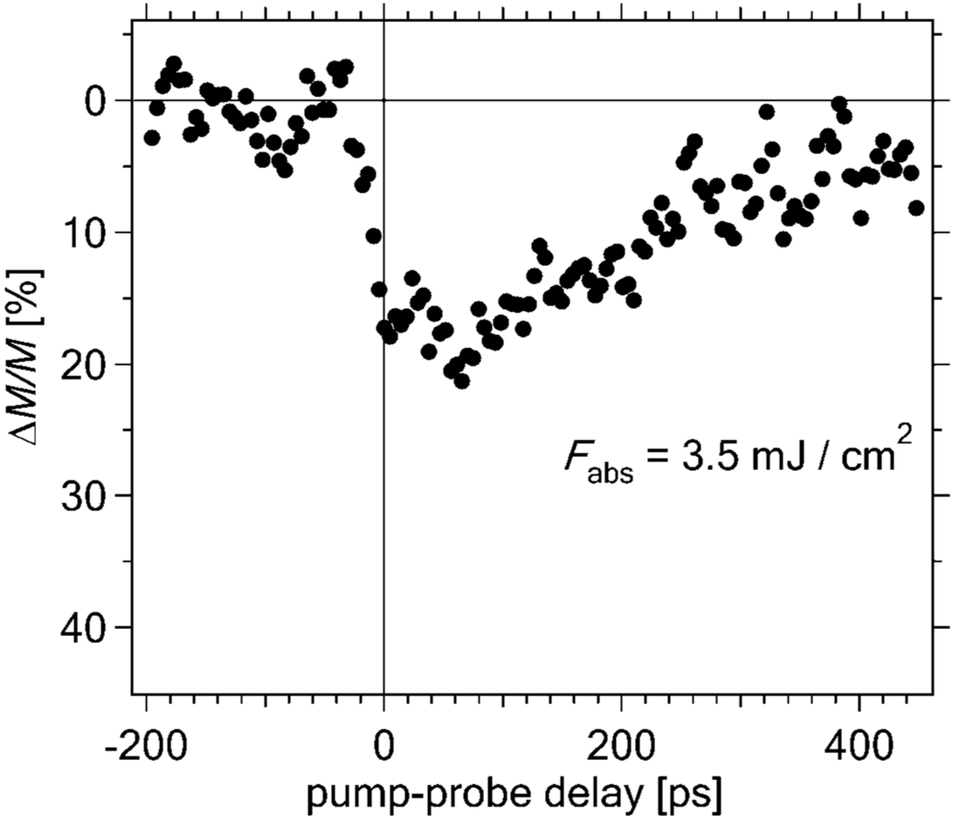




MLD contrast:

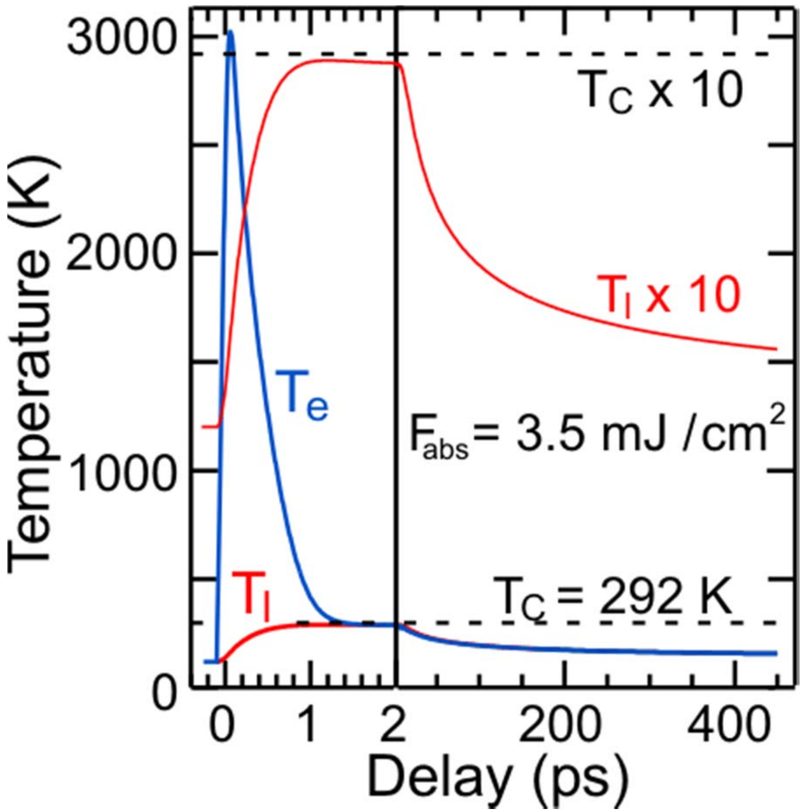
$$M_D = \sqrt{\frac{\int\left[I\uparrow(E) - I\downarrow(E)\right]^2 dE}{\int\left[I\uparrow(E) + I\downarrow(E)\right]^2 dE}} = \gamma M$$

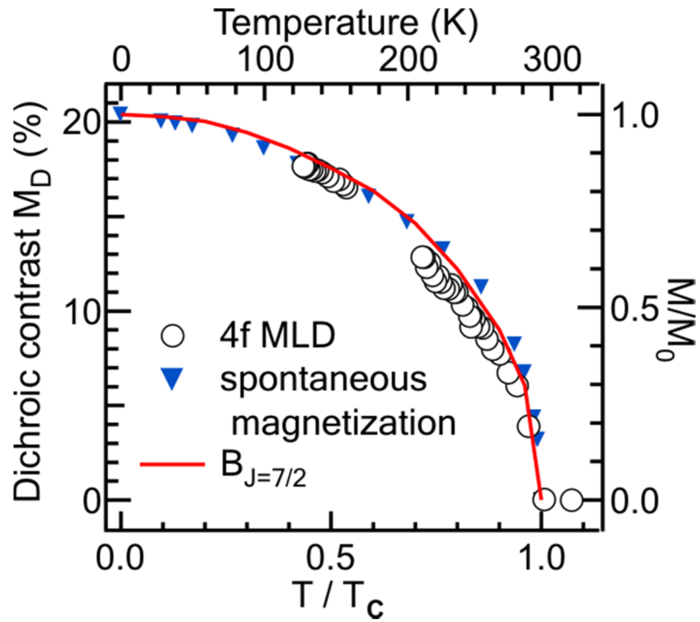
Spin-lattice relaxation in Gd 4f system



Model of coupled heat baths

2-temperature model



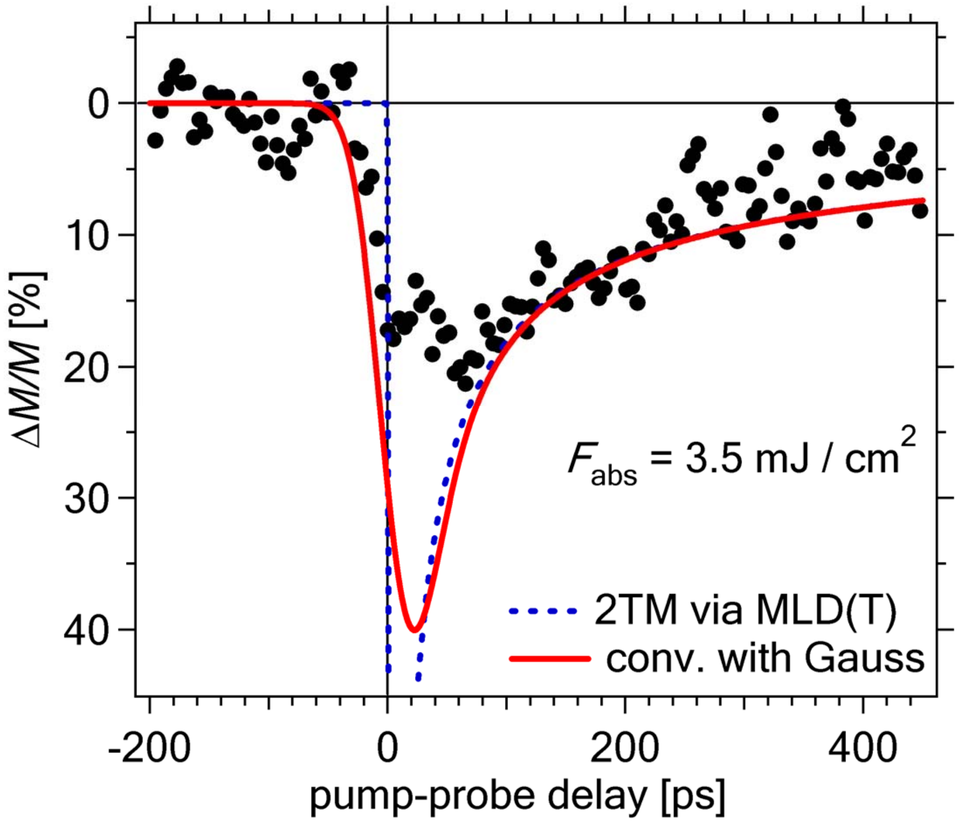


- assumption of quasi-equilibrium of lattice and spin system on time-scale of SR pulse length

time evolution of $\Delta M_D/M_D$

- lattice temperature almost reaches T_C
- within probe pulse length T_e=T_l

Spin-lattice relaxation in Gd 4f system



magnetic contrast does not follow lattice temperature for ~100 ps

→ spin-lattice relaxation time

τ_{SL} > 30 ps

Phys. Rev. Lett. ***100***, *107202 (2008).*

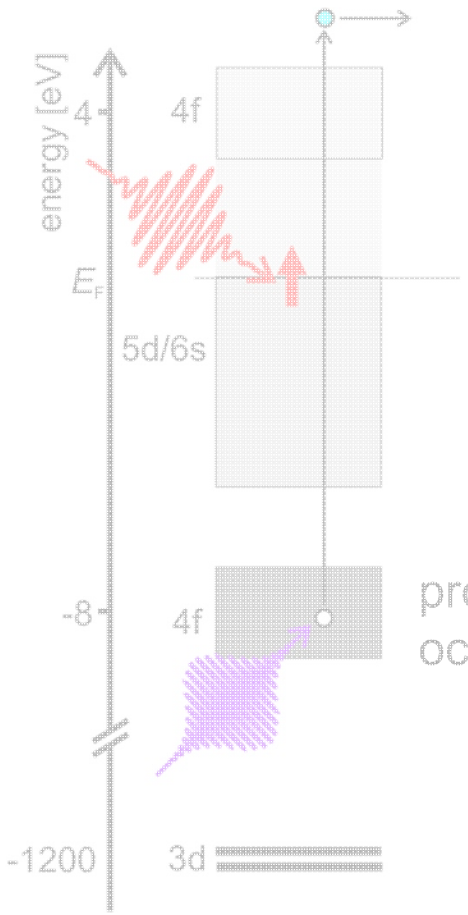
but: ultrafast time scale not resolved due to limited time resolution

Scientific case

- MLD in photoemission requires only moderate energy resolution
- Transform-limited 10 fs Gaussian pulse → 200 meV
- Intrinsic time-scale of RKKY coupling ?
- Space charge effects in dichroism easier manageable + higher photon energies
  ~10^8 photons / pulse, *A. Pietzsch et al., New Journal of Physics 10, 033004 (2008)*
- Contrast enhancement for resonant photoemission
- 4f surface and bulk components can be separated (multiplet splitting in Tb $4f^8$)

Gd-4f magnetization dynamics

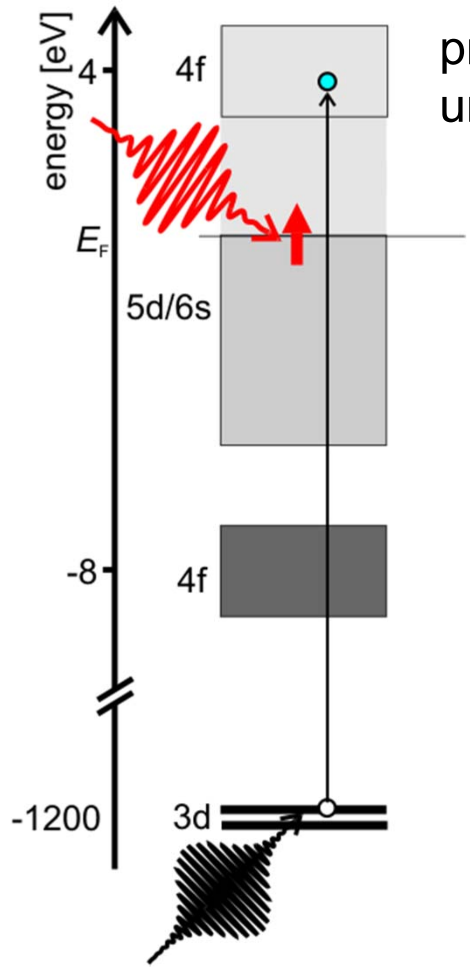
Magnetic linear dichroism (MLD) in time-resolved photoemission



probing
occupied 4f-levels

•surface and bulk sensitiv

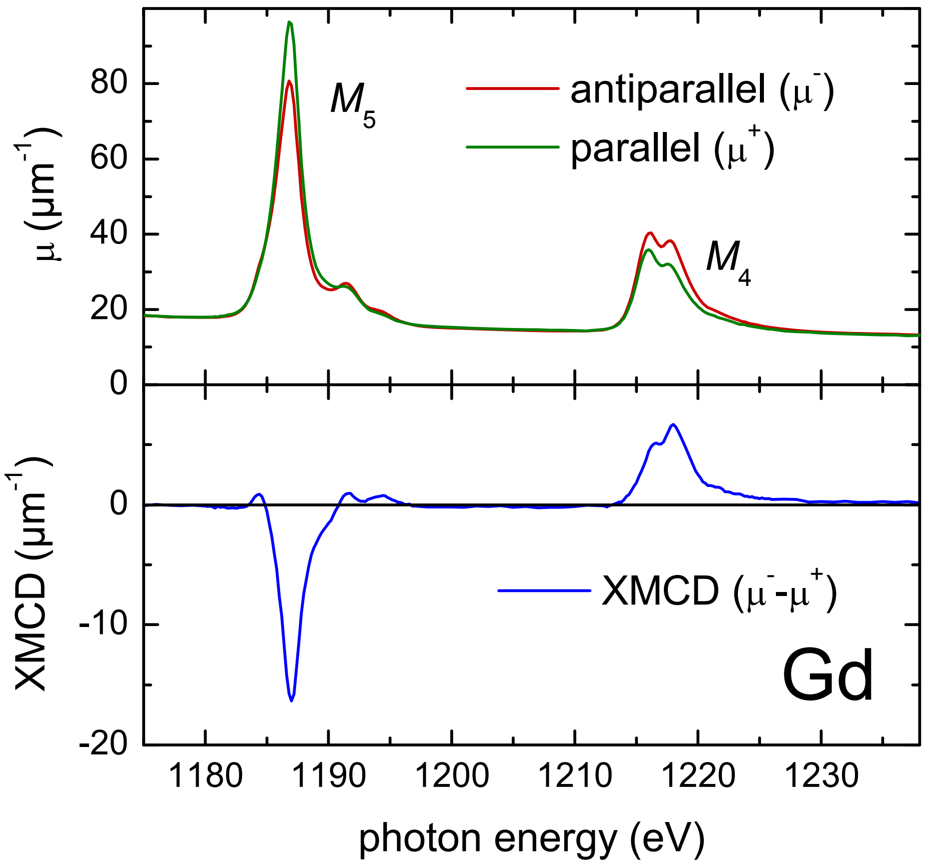
X-ray magnetic circular dichroism (XMCD) in time-resolved transmission

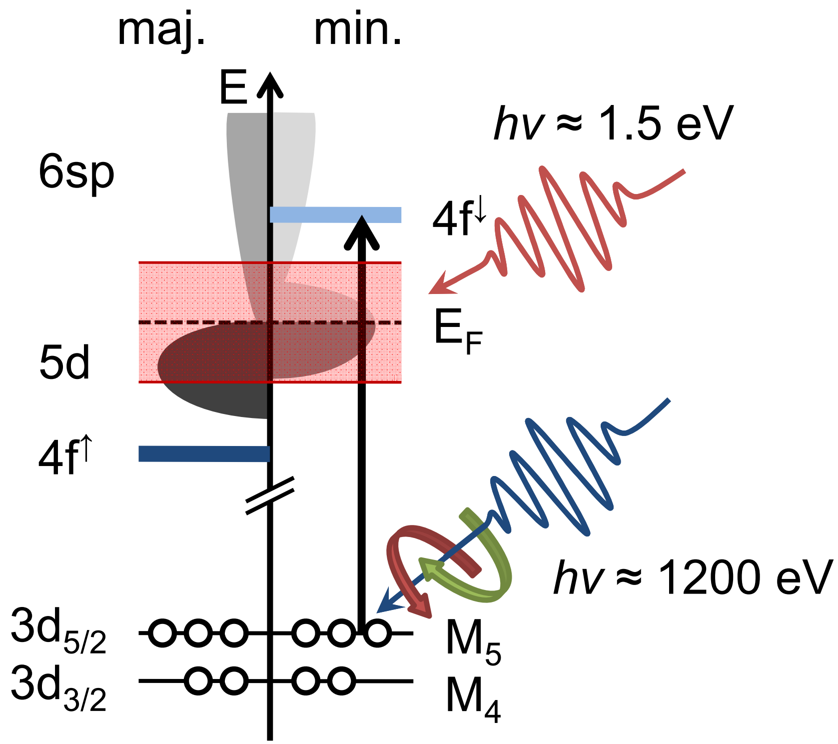


probing
unoccupied 4f-levels

• bulk sensitiv

X-ray Magnetic Circular Dichroism (XMCD)





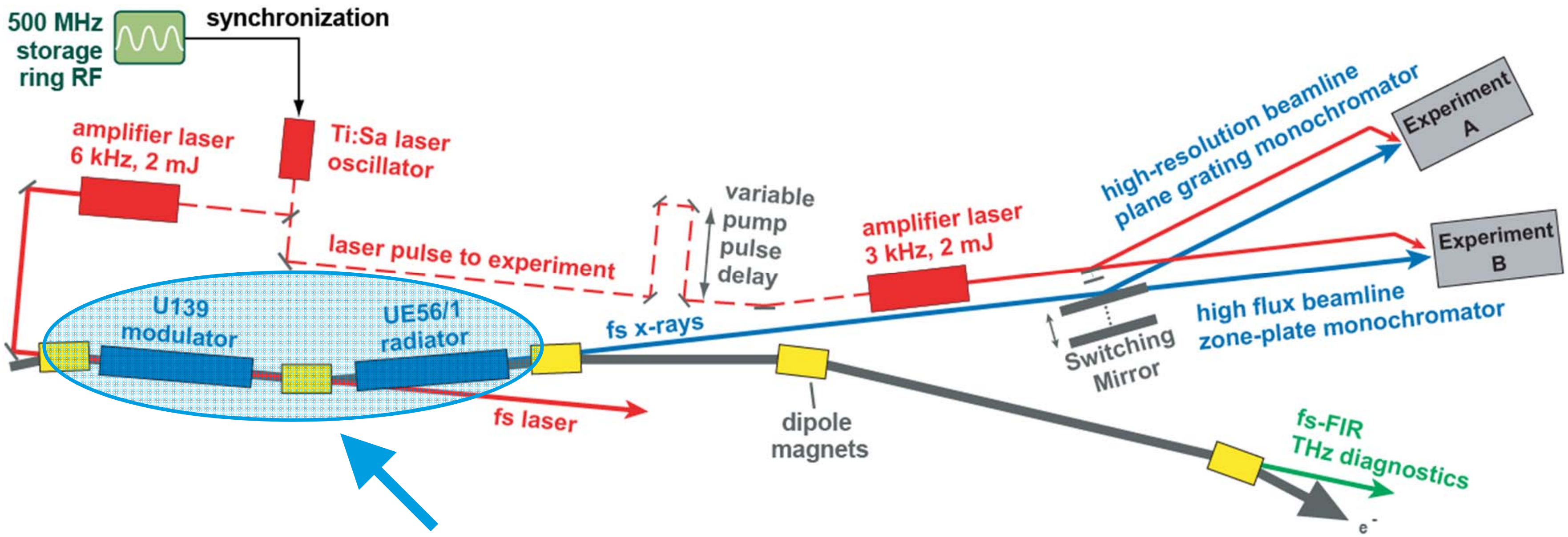
XMCD $\mu^{-} - \mu^{+} \propto M$

[Erskine, Stern PRB **12**, 5016 (1975)]

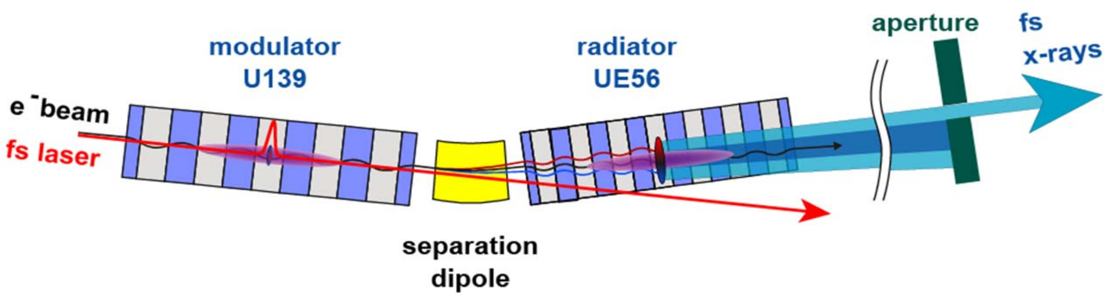
courtesy of Marko Wietstruk

BESSY II Femtoslicing Source

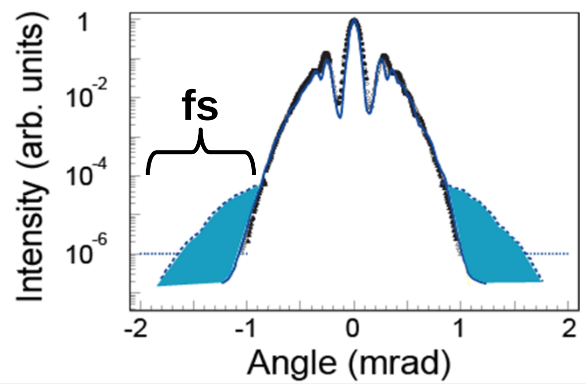
Generation of fs x-ray pulses



angular separation scheme



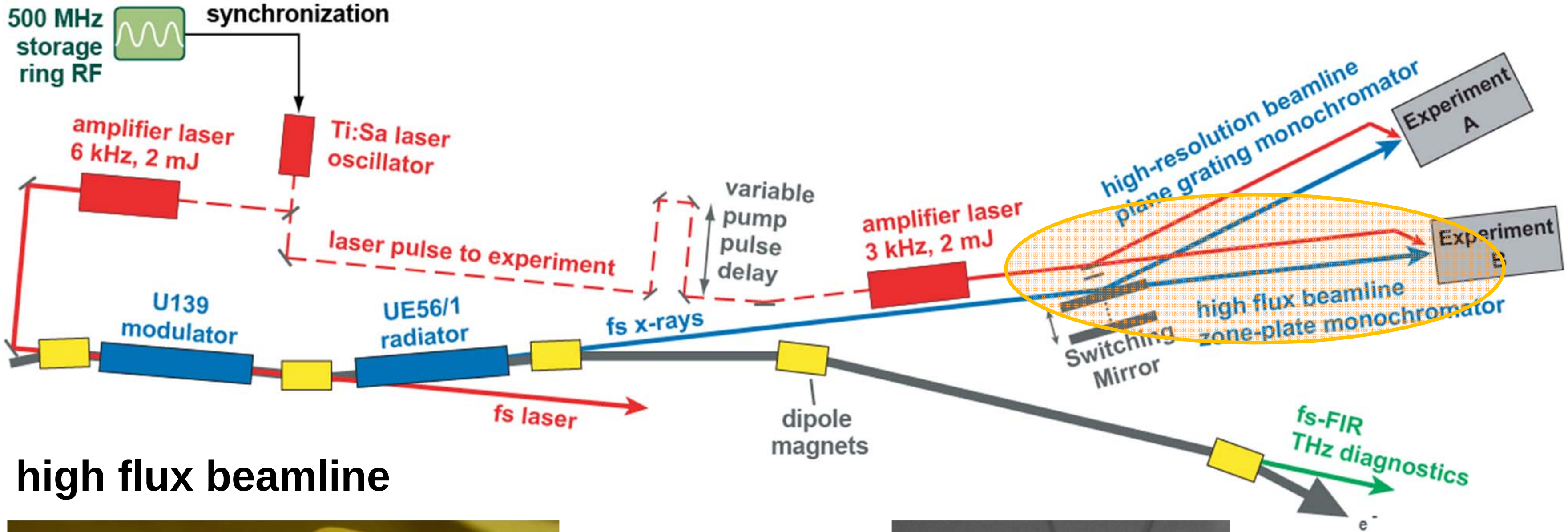
intensity distribution



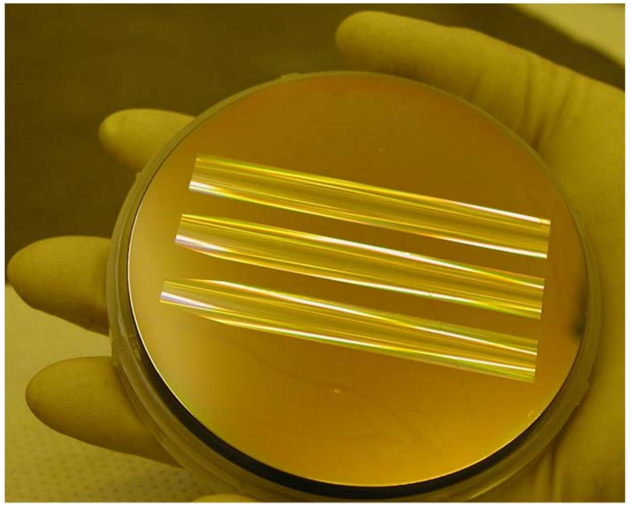
courtesy of Marko Wietstruk

BESSY II Femtoslicing Source

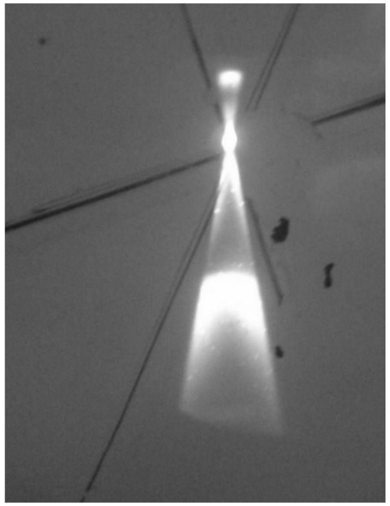
fs laser pump – fs x-ray probe



high flux beamline



Fresnel zone plate

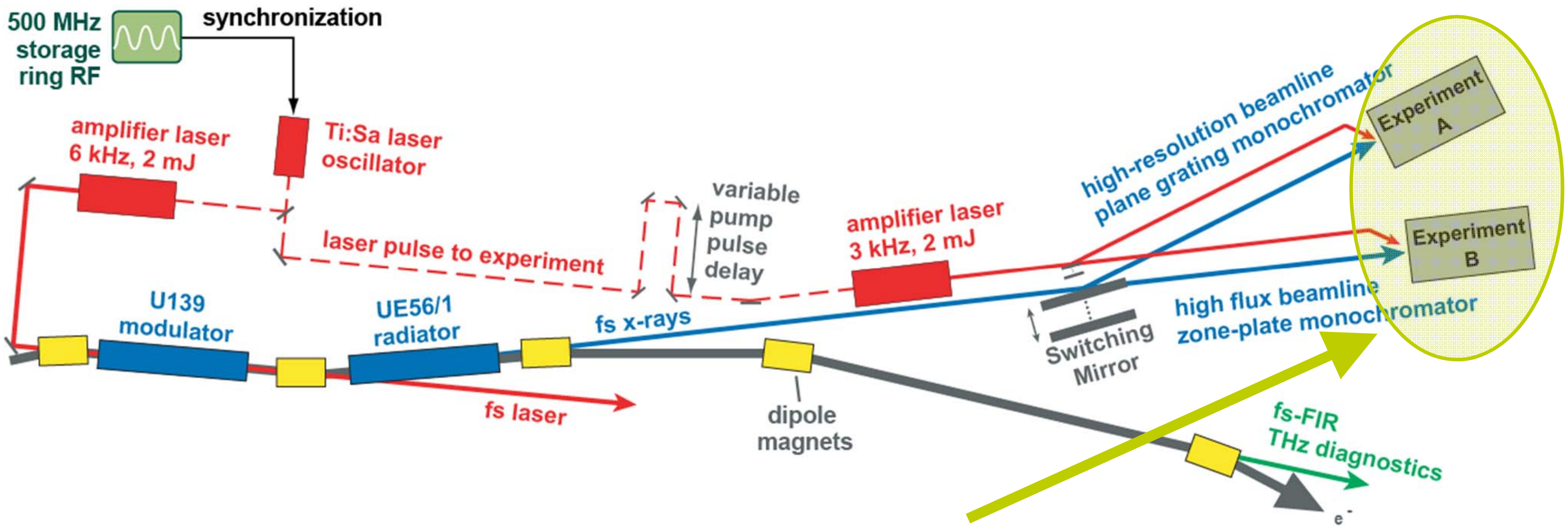


X-ray beam profile

courtesy of Marko Wietstruk

BESSY II Femtoslicing Source

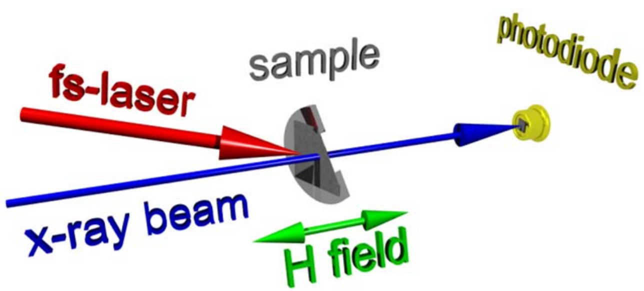
fs laser pump – fs x-ray probe



pump with 3 kHz laser pulses
probe with 6 kHz x-ray pulses

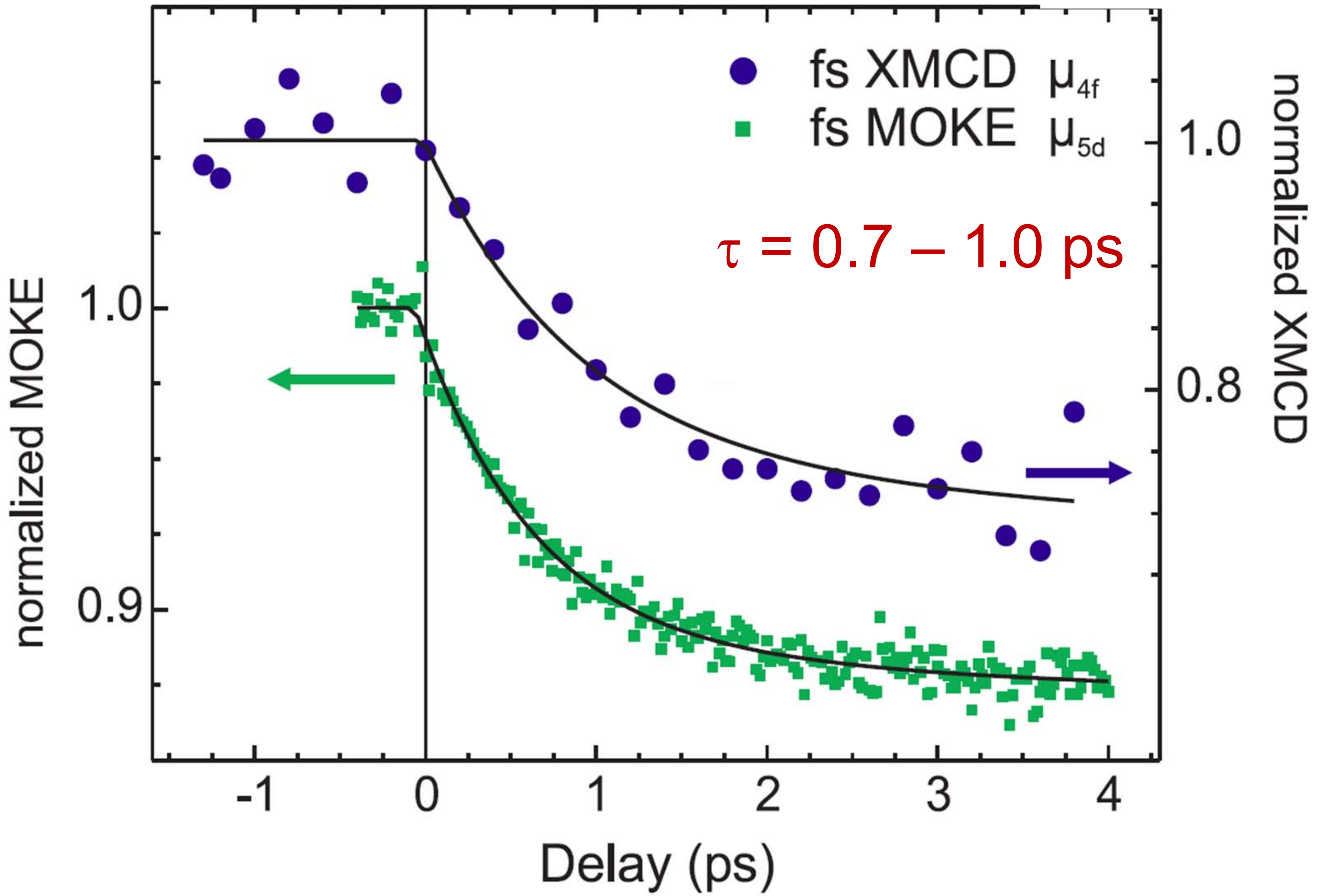
- **pump-probe delay**
- **x-ray photon energy**
- **x-ray polarization**
- **magnetic field**

x-ray transmission setup



courtesy of Marko Wietstruk

XMCD vs. MOKE -- 4f vs. 5d



„Comparable“ ultrafast dynamics (± 0.2 ps)

M. Wietstruk et al., PRL 106 (2011) 127401.

Scientific case

- Photon in – photon out
- High time resolution

BUT:

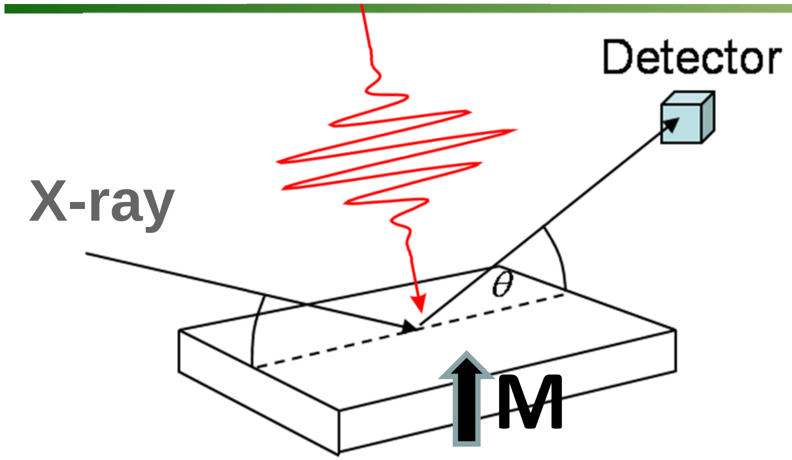
- Heat problem in thin film samples
- Sample quality less defined
- Switching of magnetic field requires time

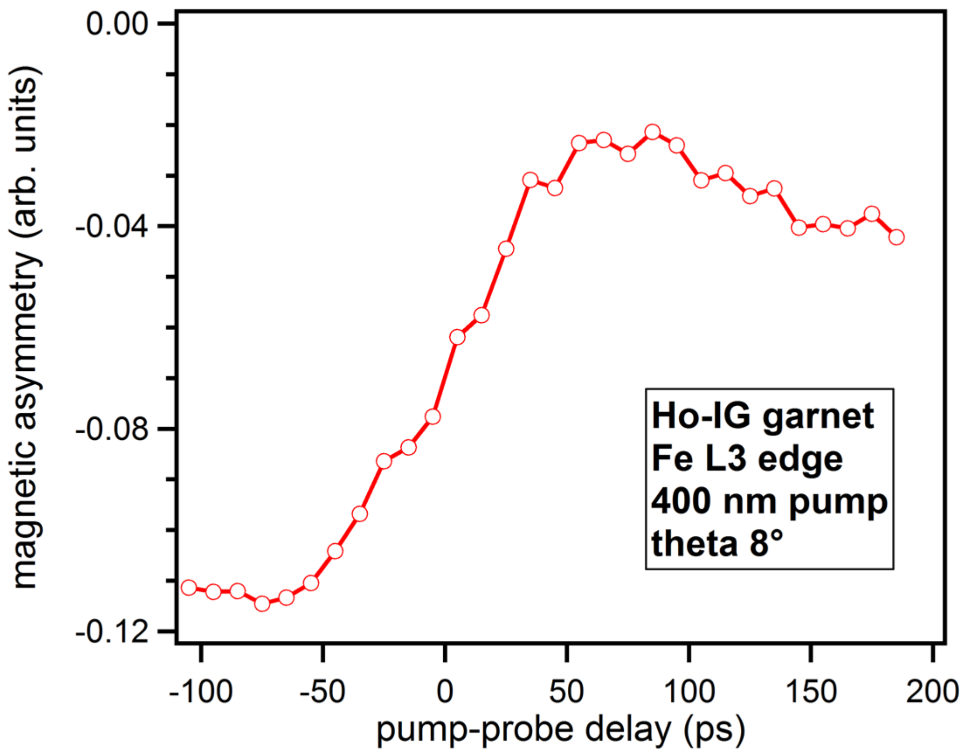
Planned BMBF proposal

Experimental endstation

- X-ray diffraction and reflection
- TMOKE
- variable orientation of fast switchable, high magnetic field (< 2 Tesla)
- preparation chamber with MBE / PLD deposition and RHEED
- easy to move to synchrotron, FEL, HHG and laser sources

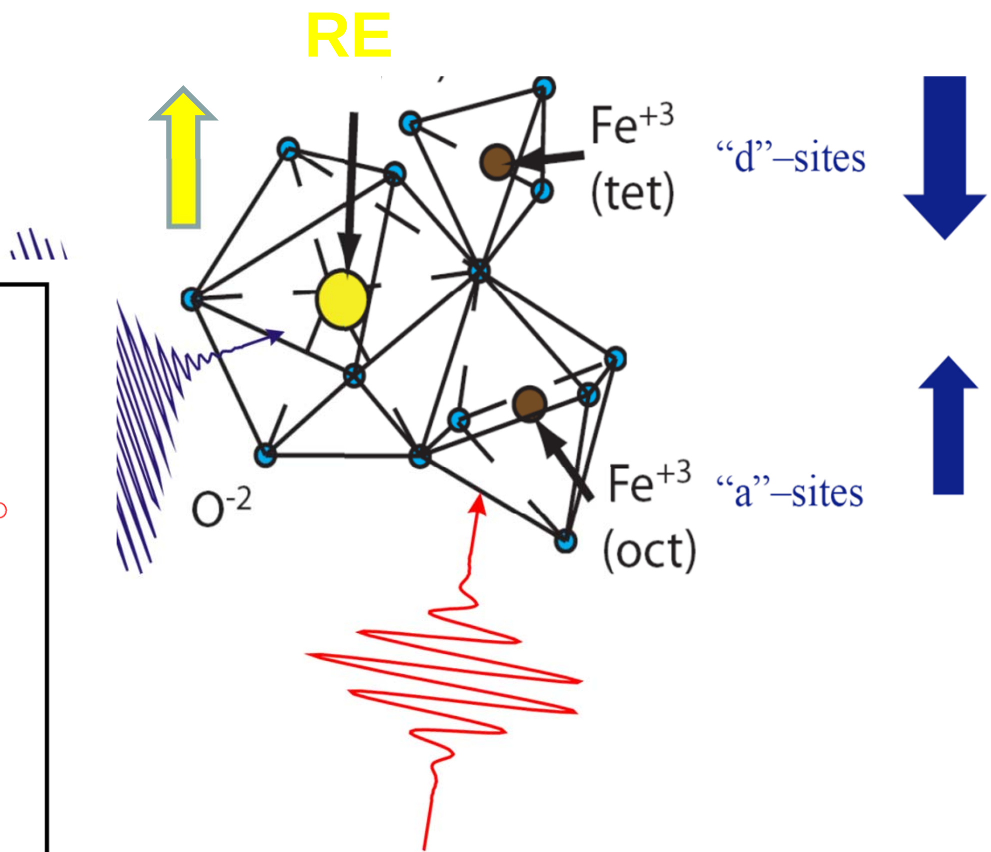
Ongoing @ Slicing: X-ray Resonant Magnetic Reflectivity





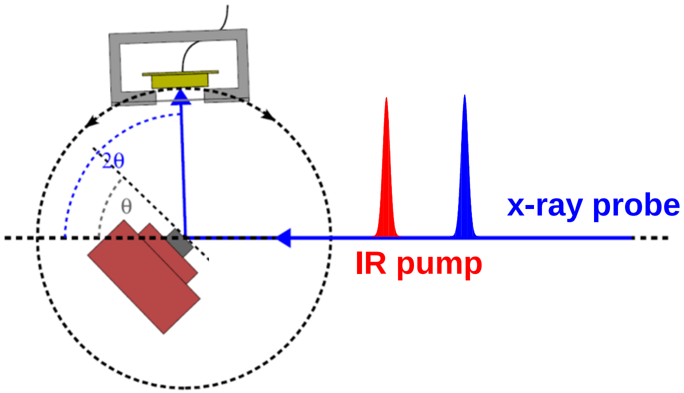
Ferrimagnetic Oxides

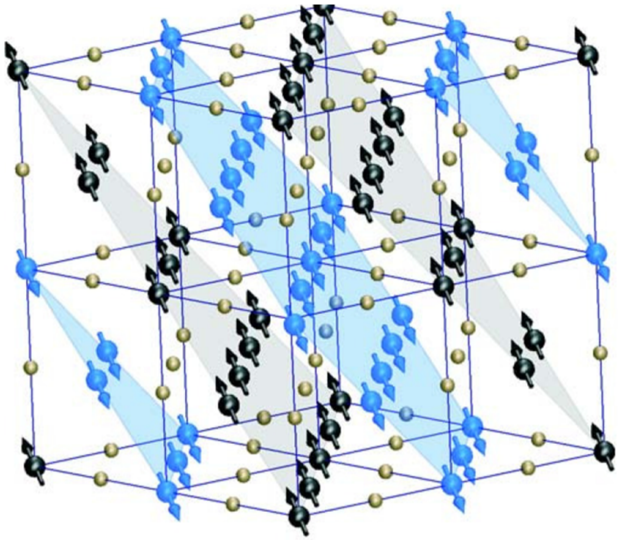
- selective excitation
- resonant probing / imaging



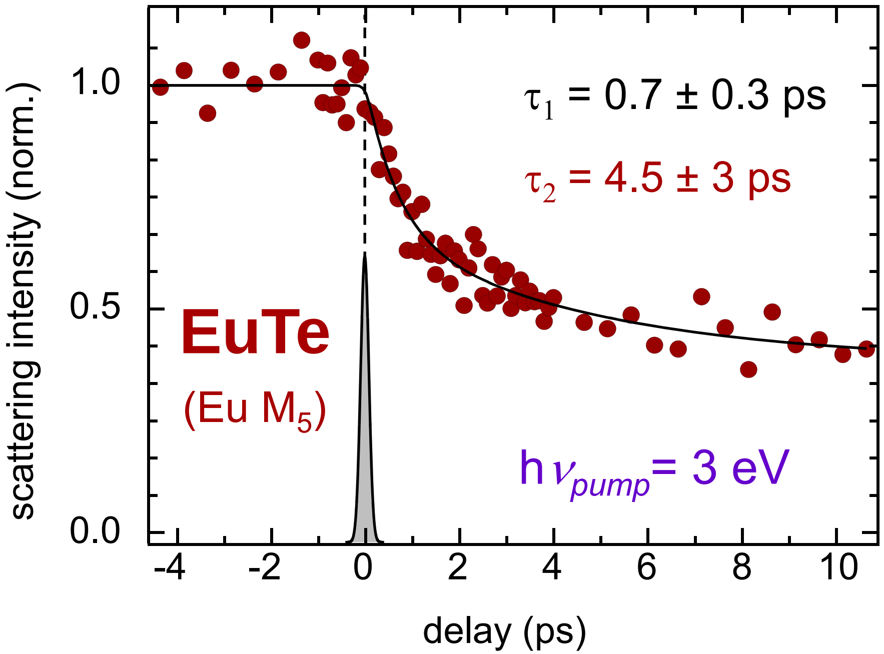
courtesy of Ilie Radu

Antiferromagnetic semiconductor EuTe

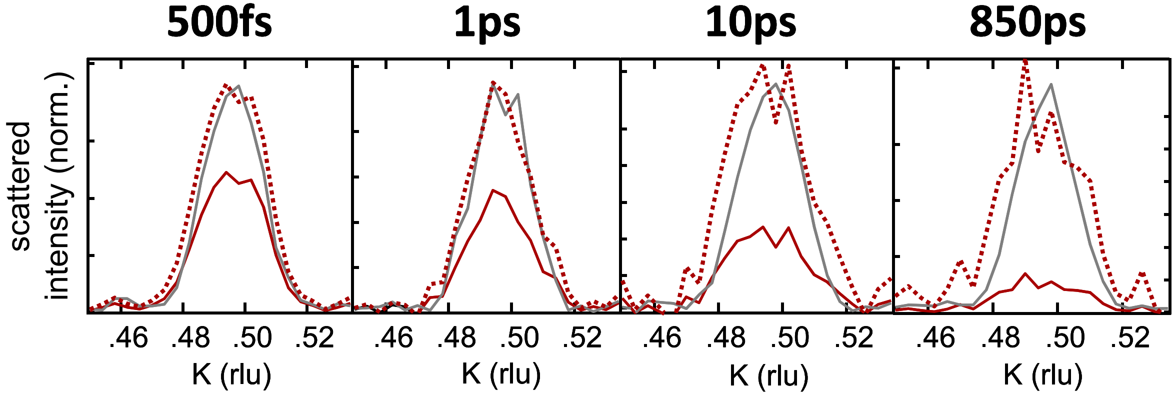




K. Holldack et *al.*,
APL **97**, 062502 (2010)



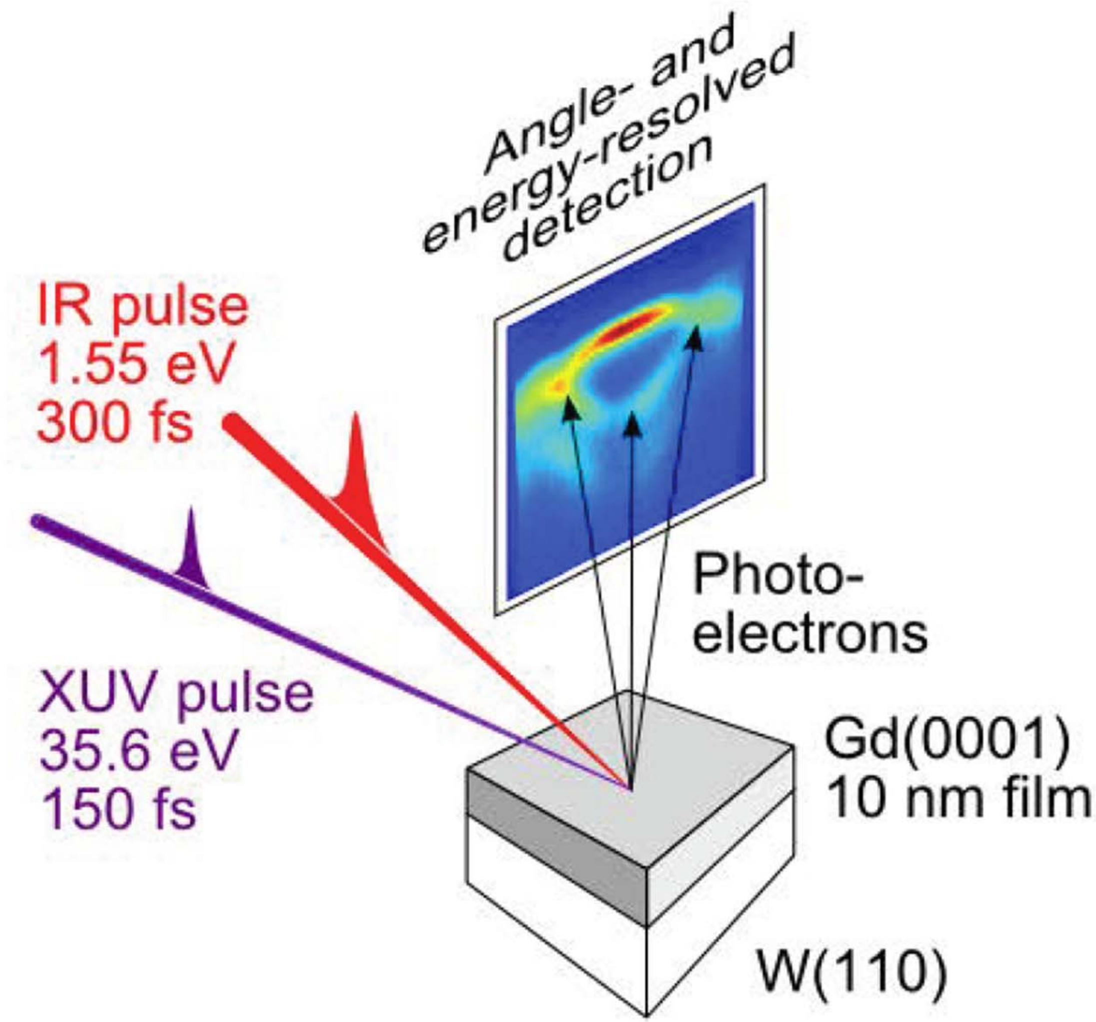
Ultrafast antiferromagnetic dynamics (½, ½, ½)



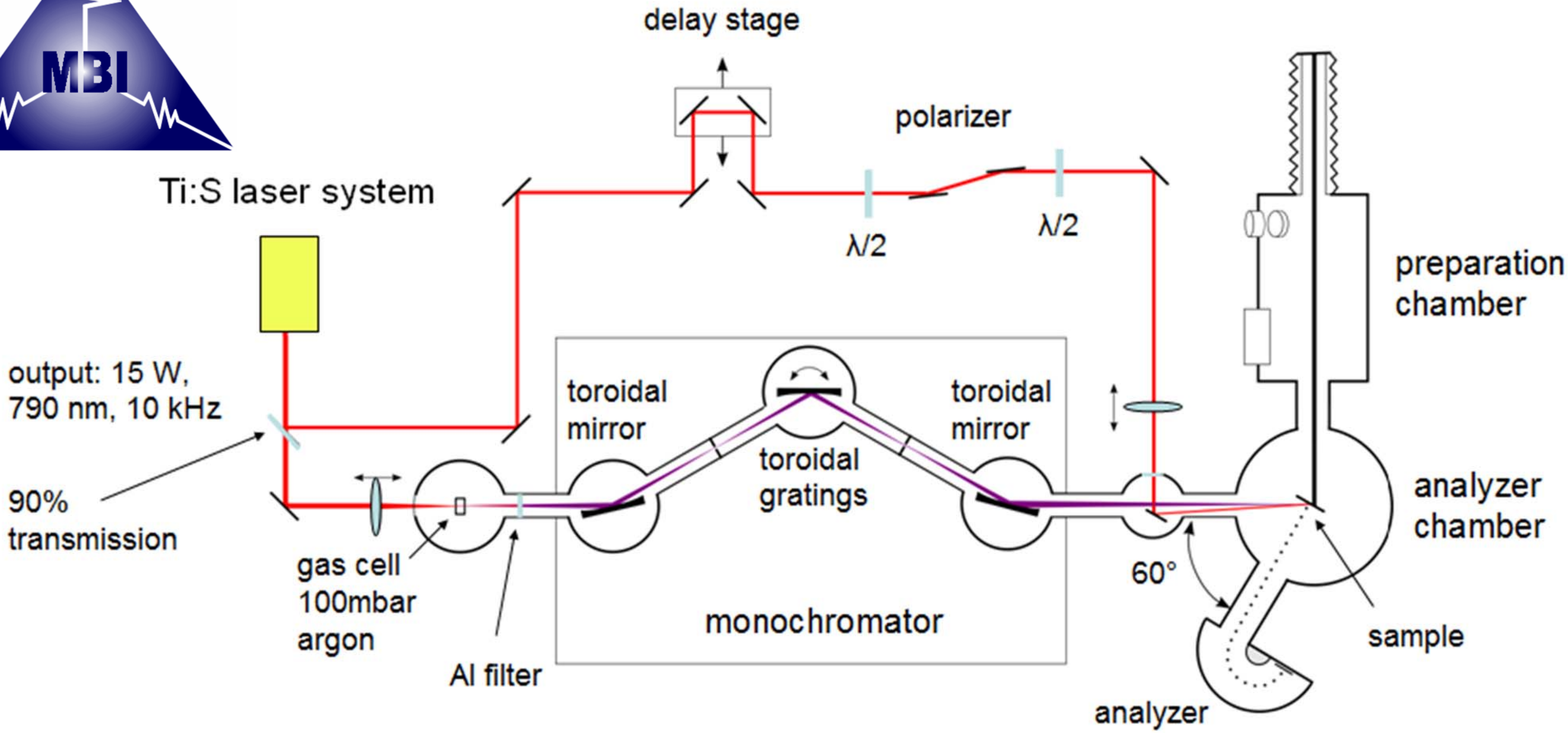
Diffraction peak shape reveals transient depth profile

courtesy of Christian Schüßler-Langeheine

Time-resolved photoemission



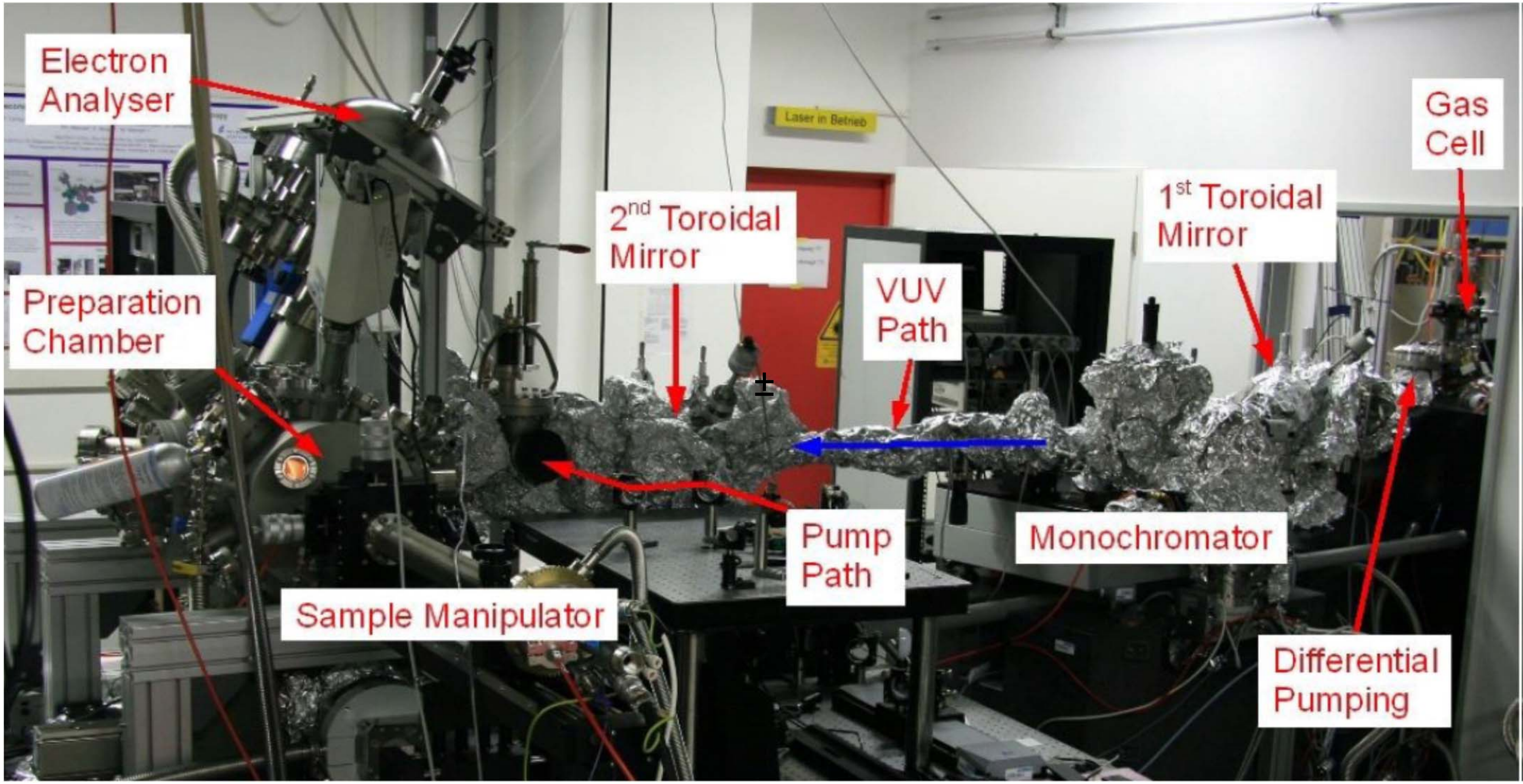
Experimental Apparatus



Pump: 1.5 eV at 300 fs FWHM (s polarized, streched)
Probe: 35.6 eV at 100 fs FWHM (p polarized)

XUV photoemission beamline

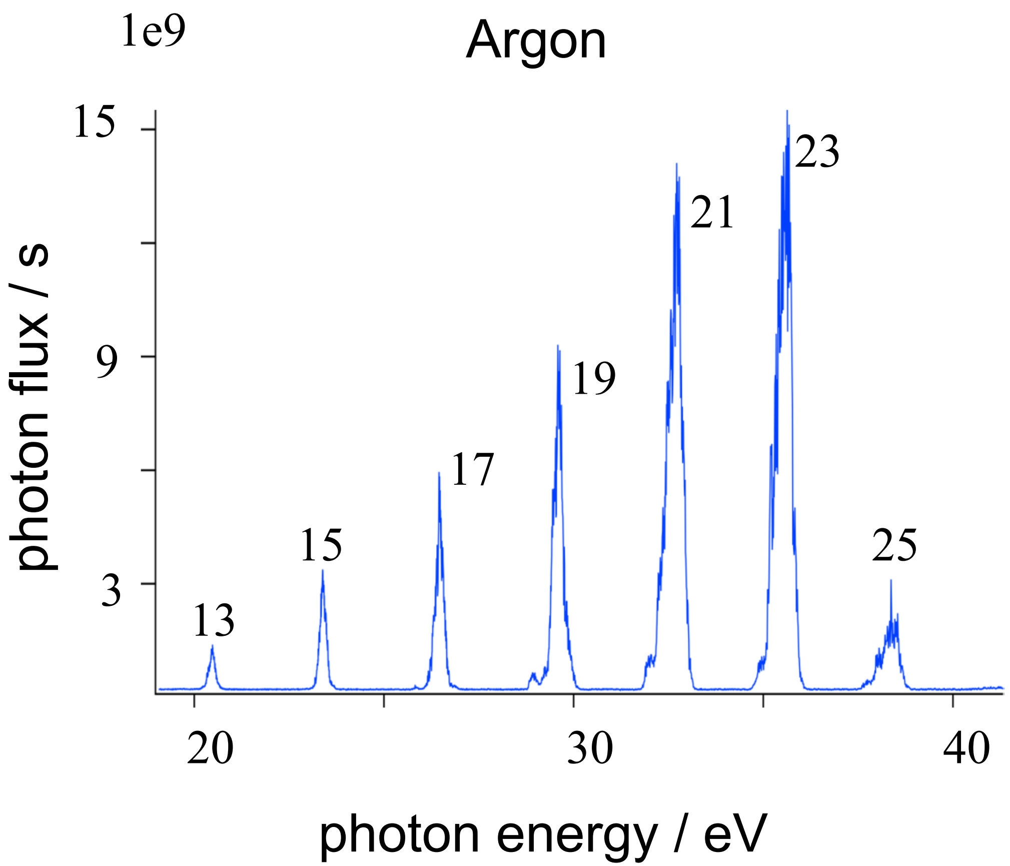
Freie Universität Berlin

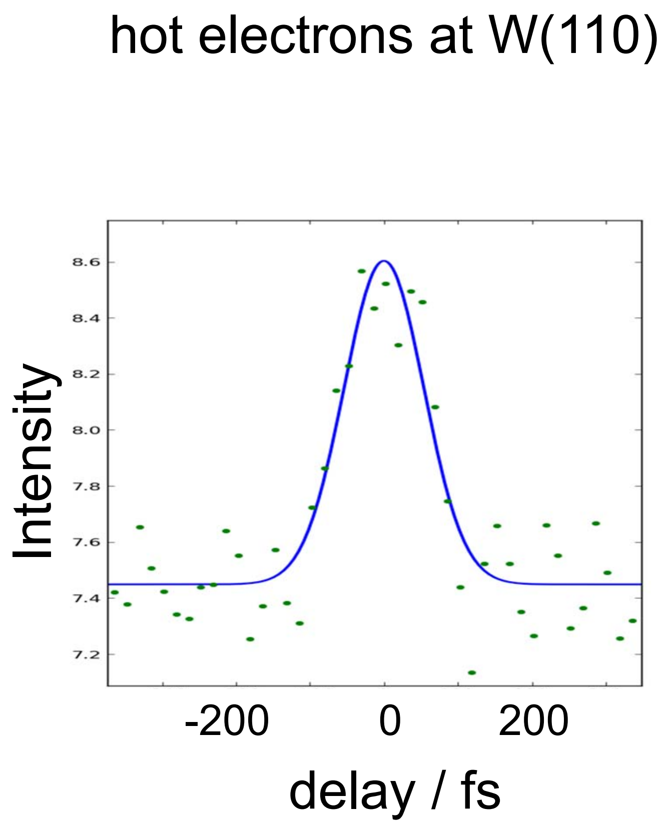


Robert Carley, Martin Teichmann, Björn Frietsch, Kristian Döbrich

German Research Foundation, DinL (WGL graduate school), Helmholtz Virtual Institute

Beamline Characterization



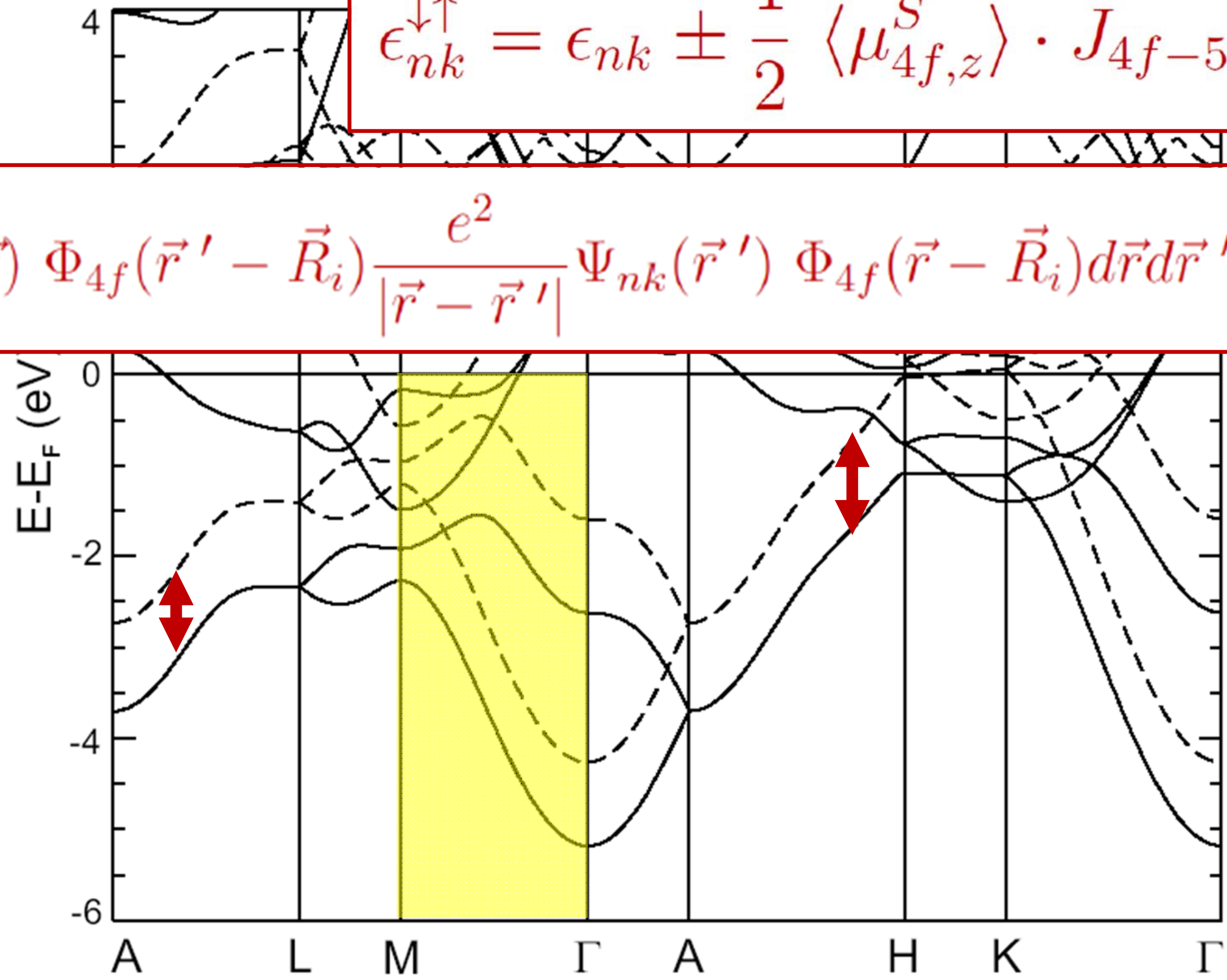


100 fs XUV pulse duration

Electronic band structure of Gd

$$\epsilon_{nk}^{\downarrow\uparrow} = \epsilon_{nk} \pm \frac{1}{2}\,\langle \mu_{4f,z}^{S} \rangle \cdot J_{4f-5d}(n\vec{k}, n\vec{k})$$

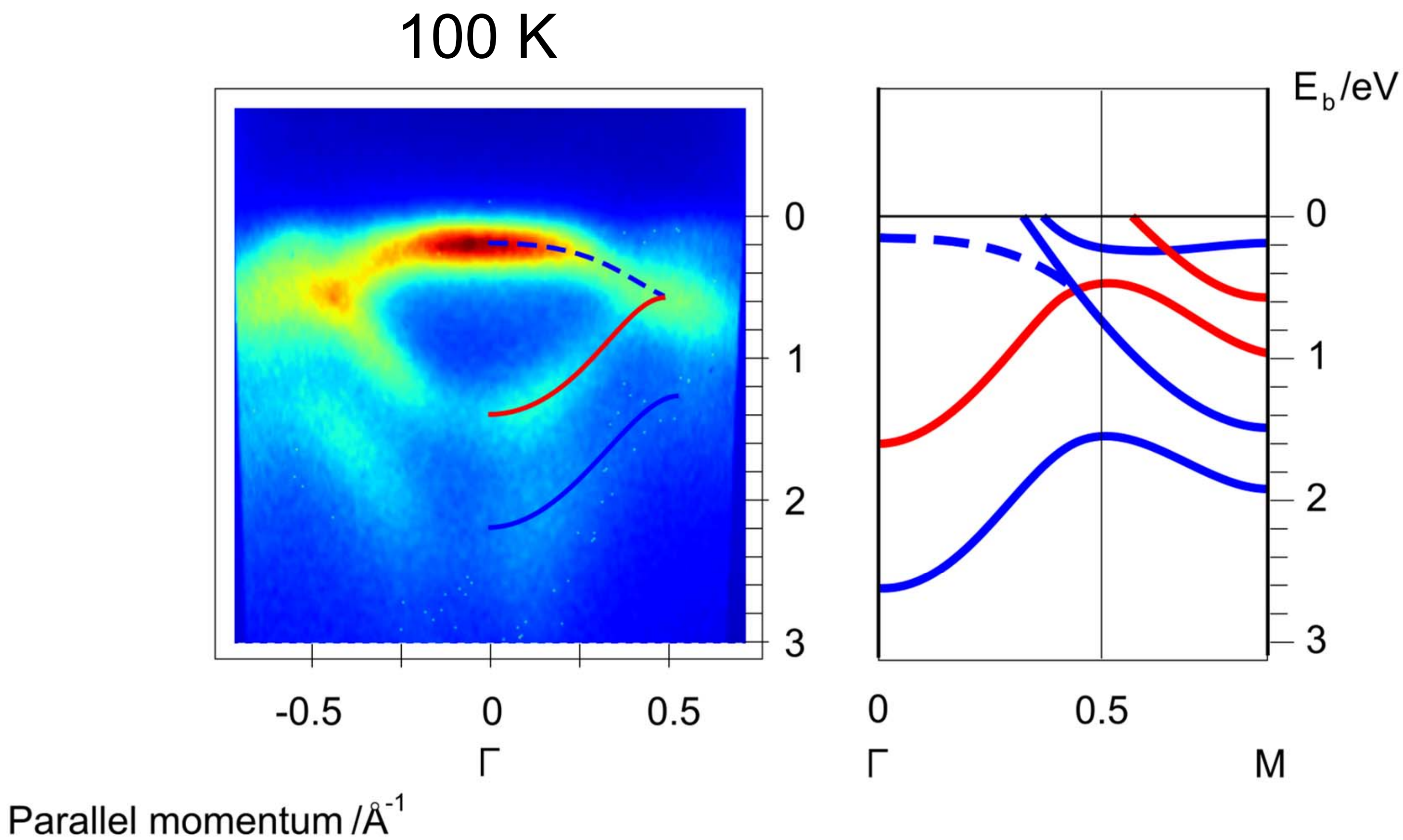
$$\int \Psi_{n'k'}(\vec{r})\ \Phi_{4f}(\vec{r}\,' - \vec{R}_i)\frac{e^2}{|\vec{r} - \vec{r}\,'|}\Psi_{nk}(\vec{r}\,')\ \Phi_{4f}(\vec{r} - \vec{R}_i)d\vec{r}d\vec{r}\,' \simeq 88\ \mathrm{meV}$$



Stoner-like exchange splitting: 0.6 – 0.9 eV

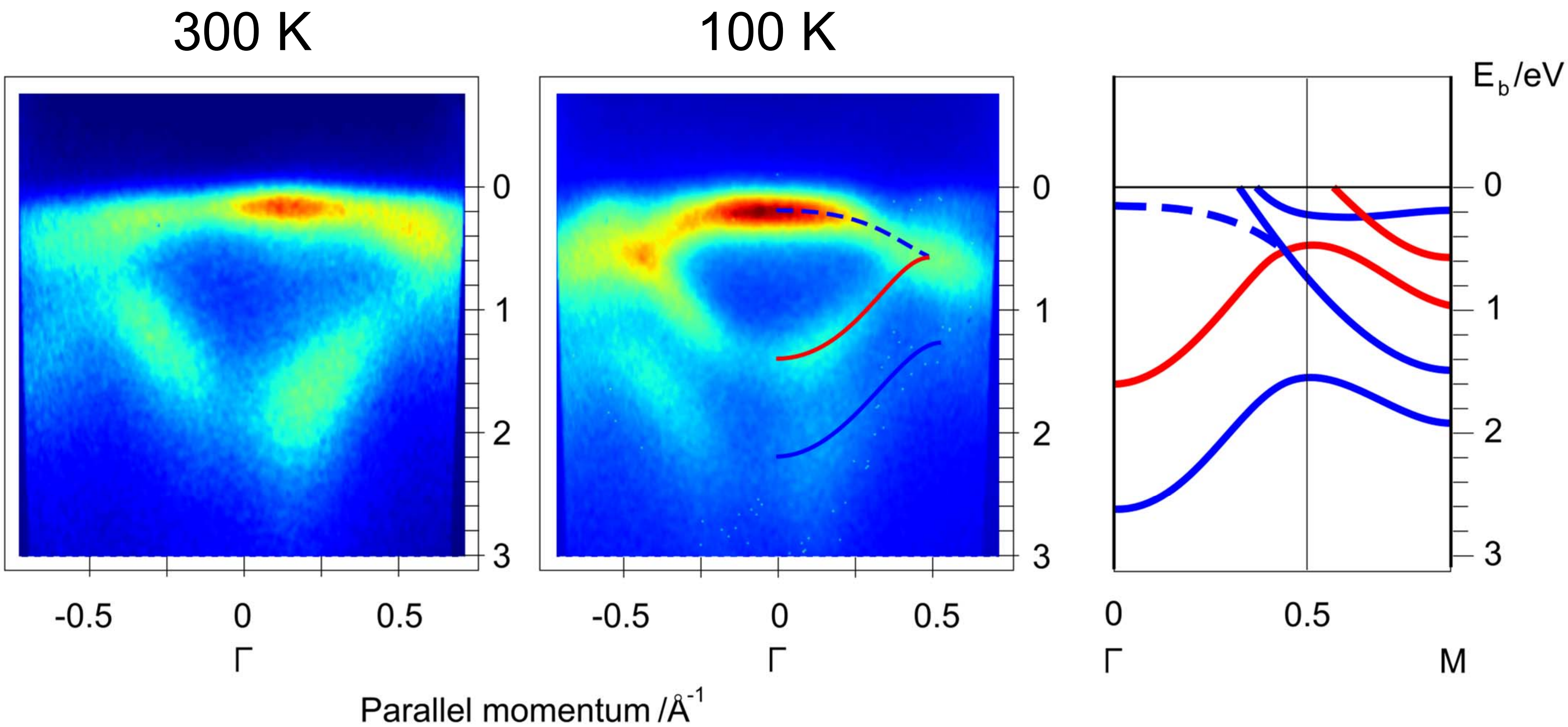
P. Kurz, G. Bihlmayer, S. Blügel; J. Phys.: Condens. Matter, 14 (2002) 6353.

Band Structure of Gd(0001) on W(110)



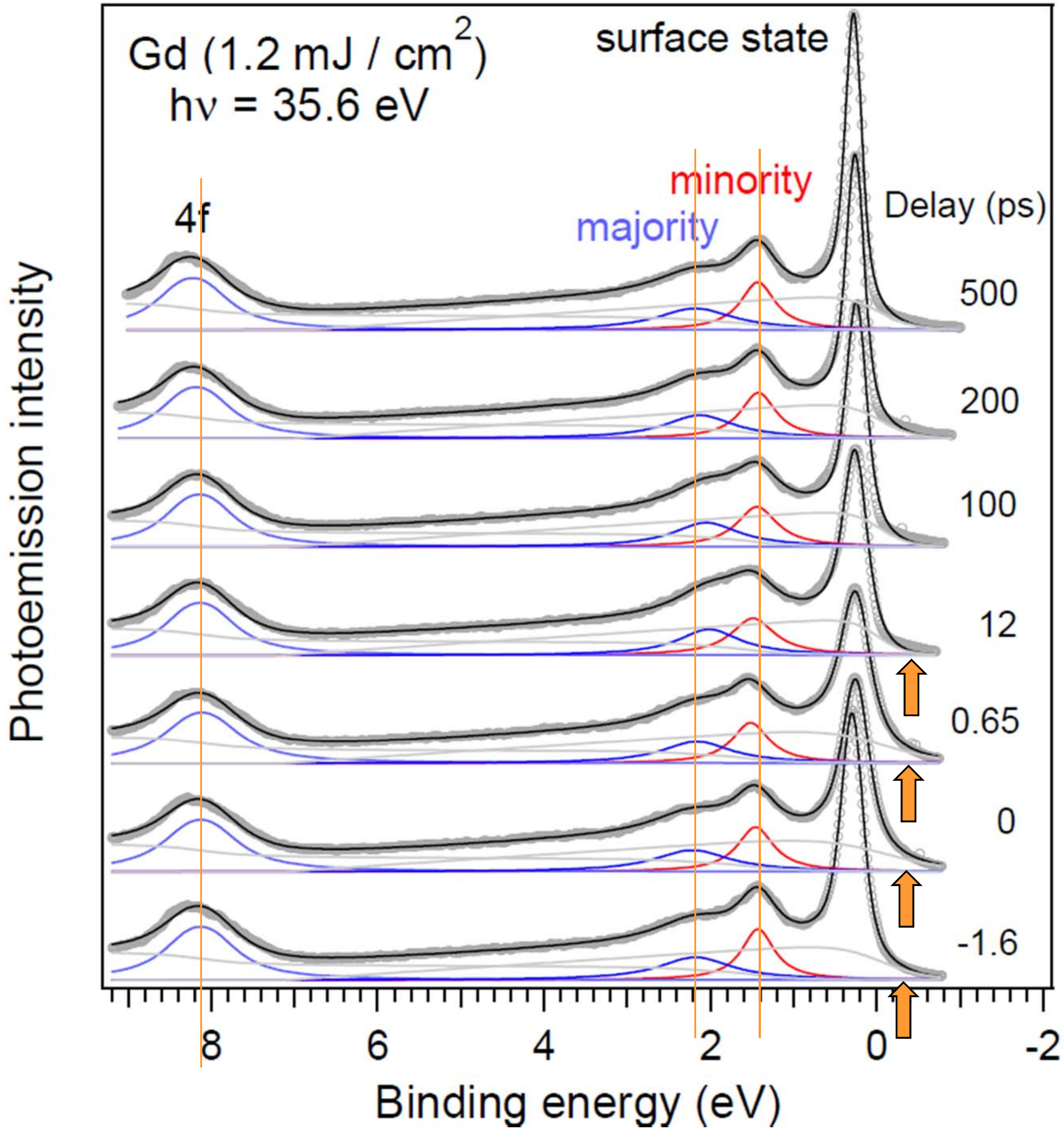
3rd Brillouin zone – hν = 35 eV

Band Structure of Gd(0001) on W(110)

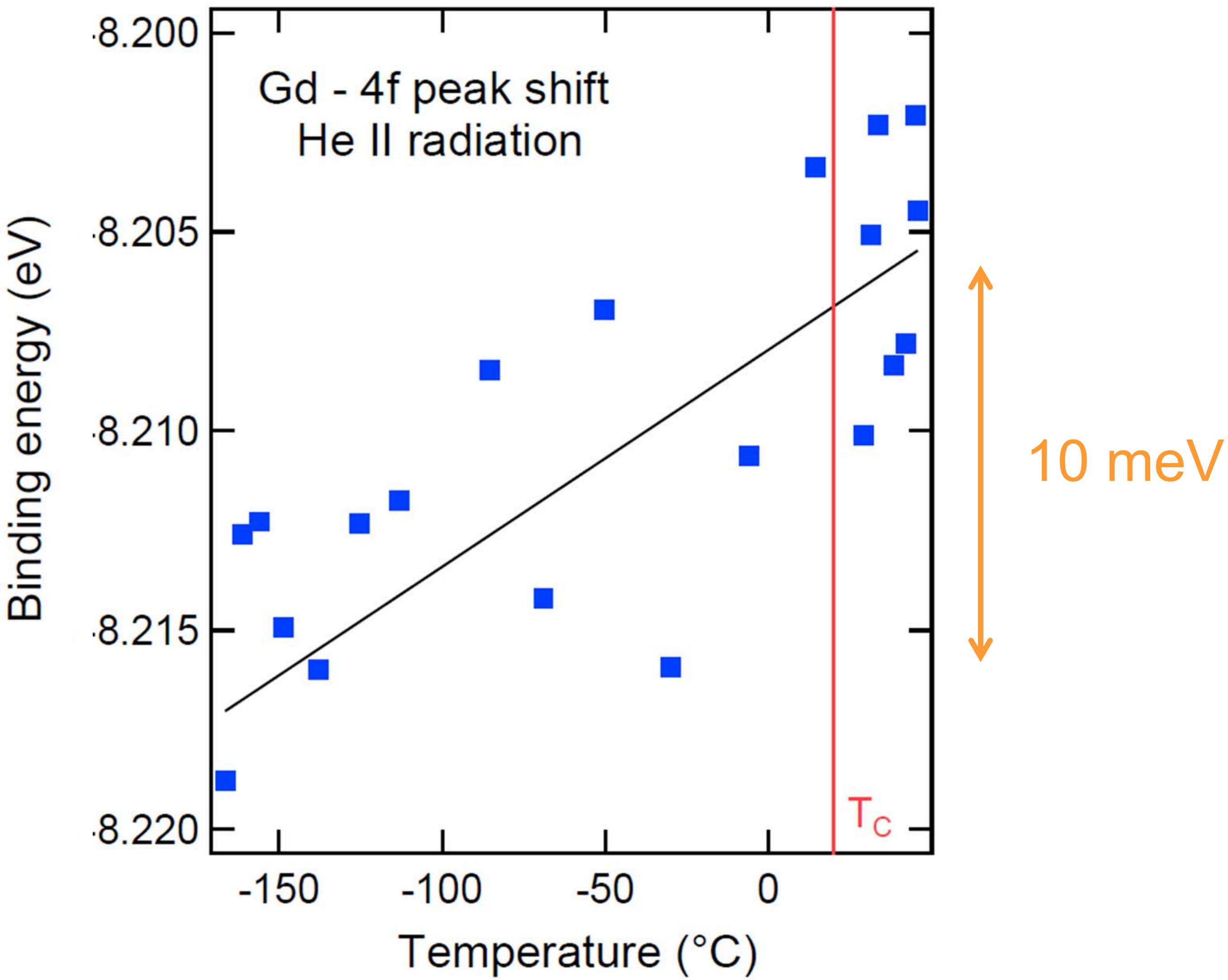


3rd Brillouin zone – hν = 35 eV

Energy-resolved spectra

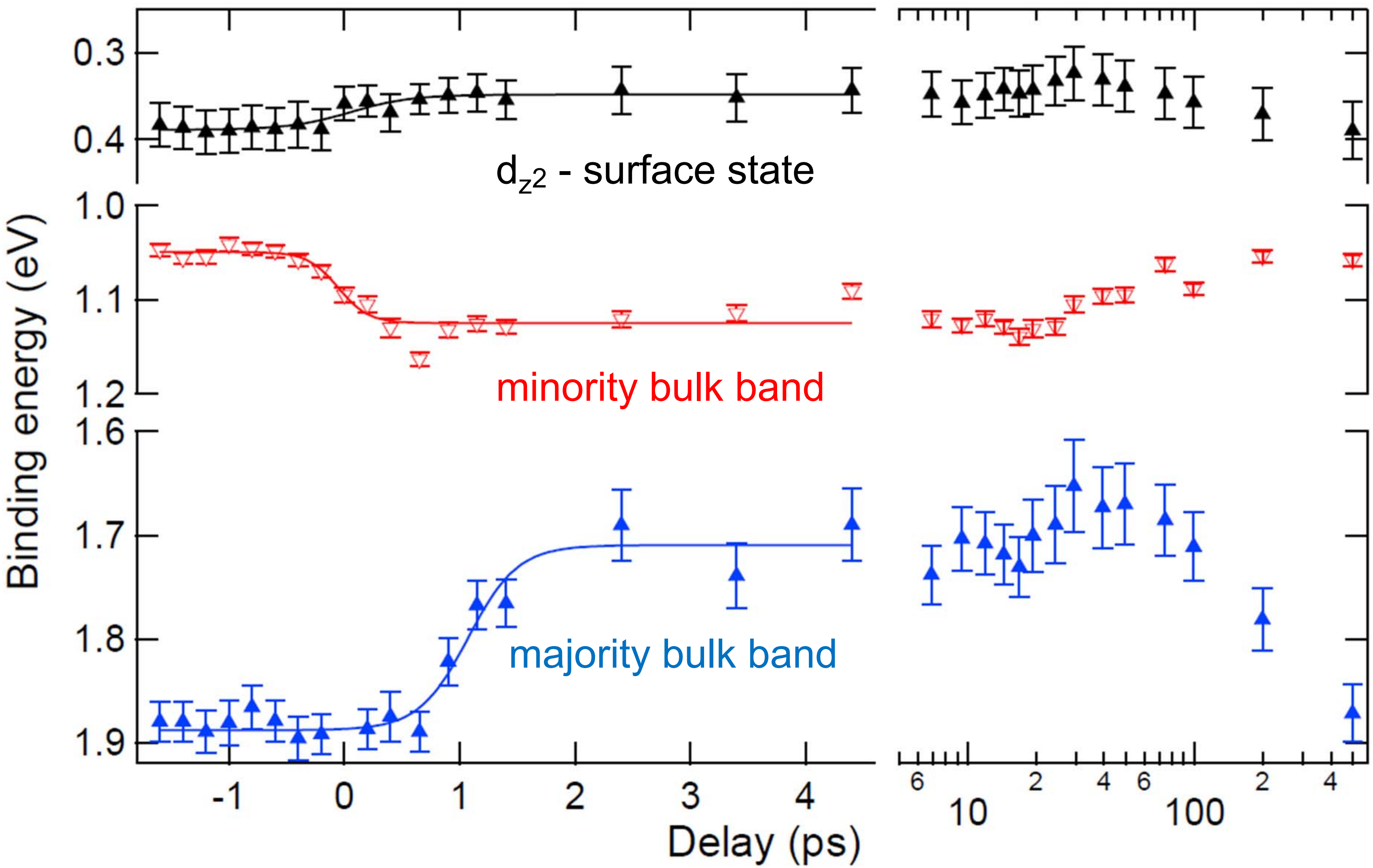


4f position – thermal demagnetization

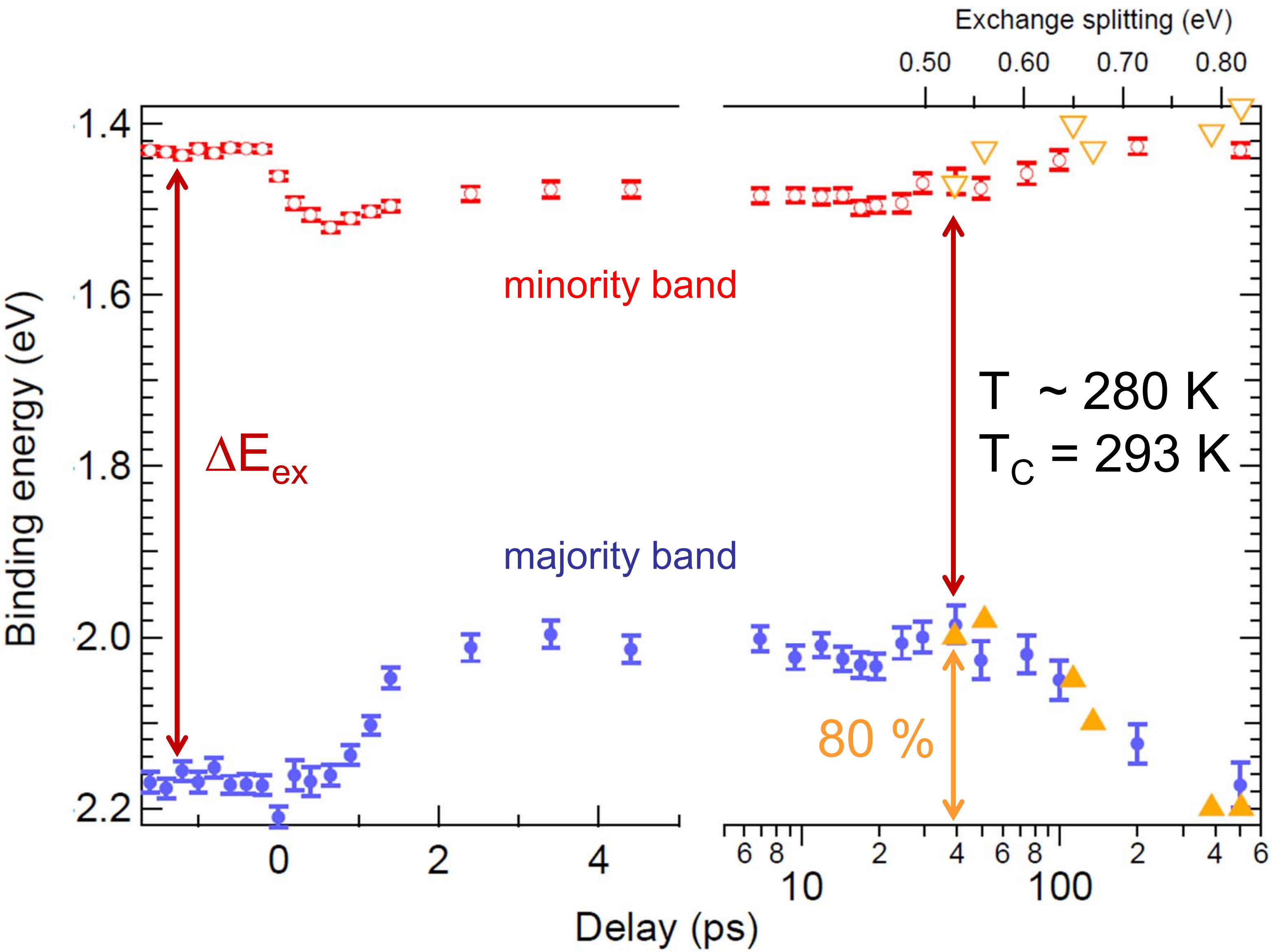


No significant shift of 4f level

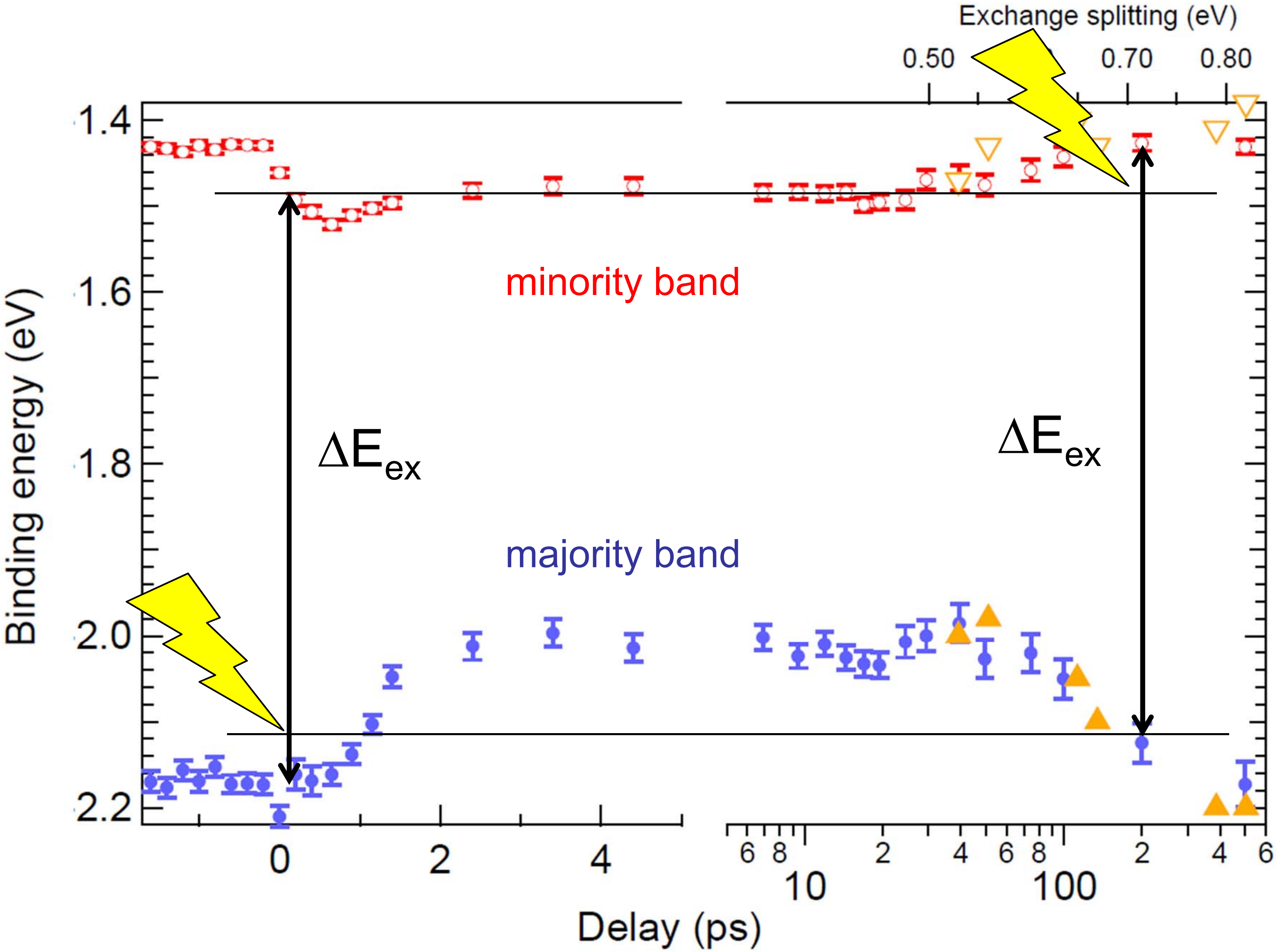
Optically-driven demagnetization (1.2 mJ / cm^2)



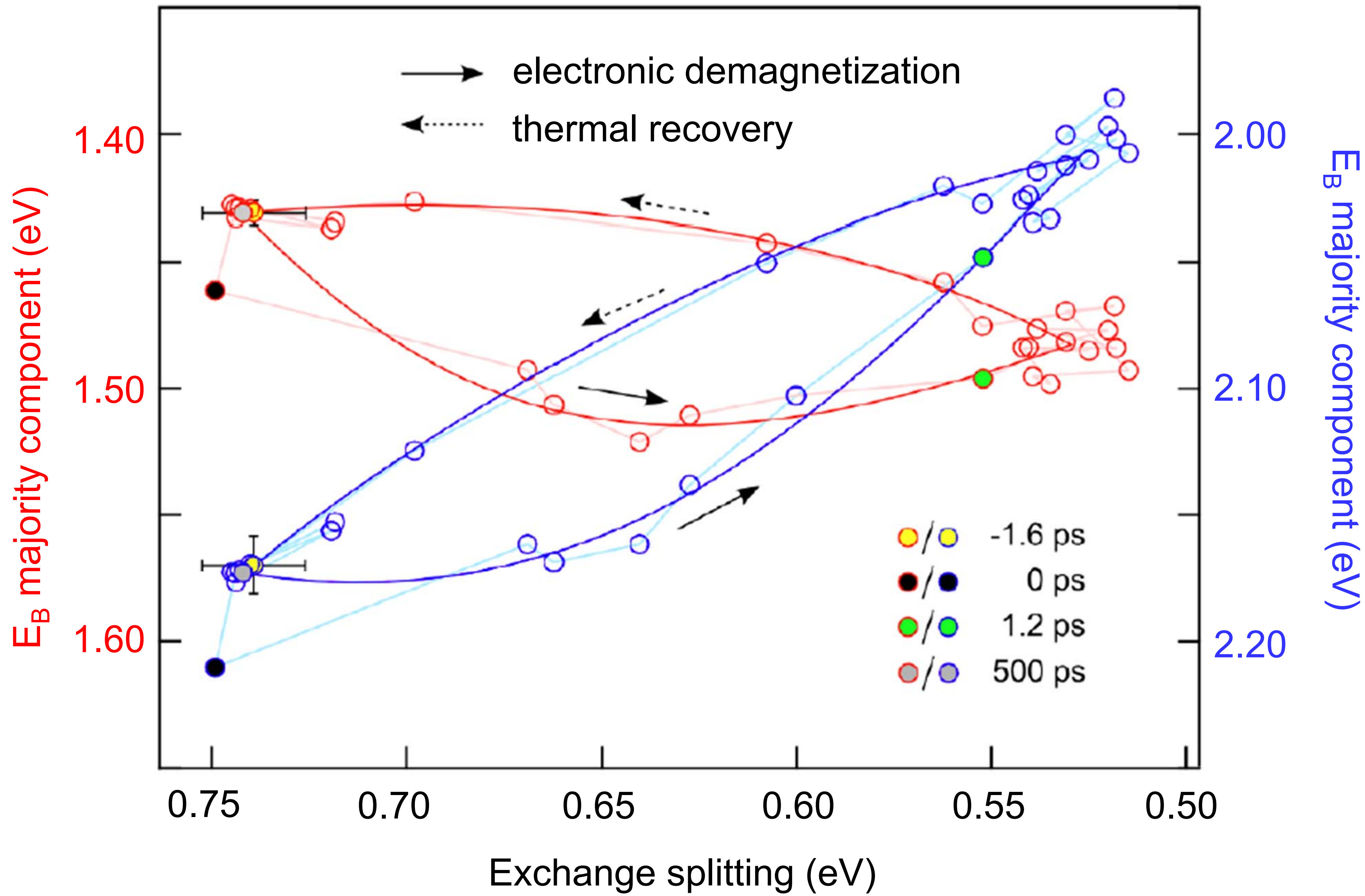
Optically-driven demagnetization (1.2 mJ / cm²)



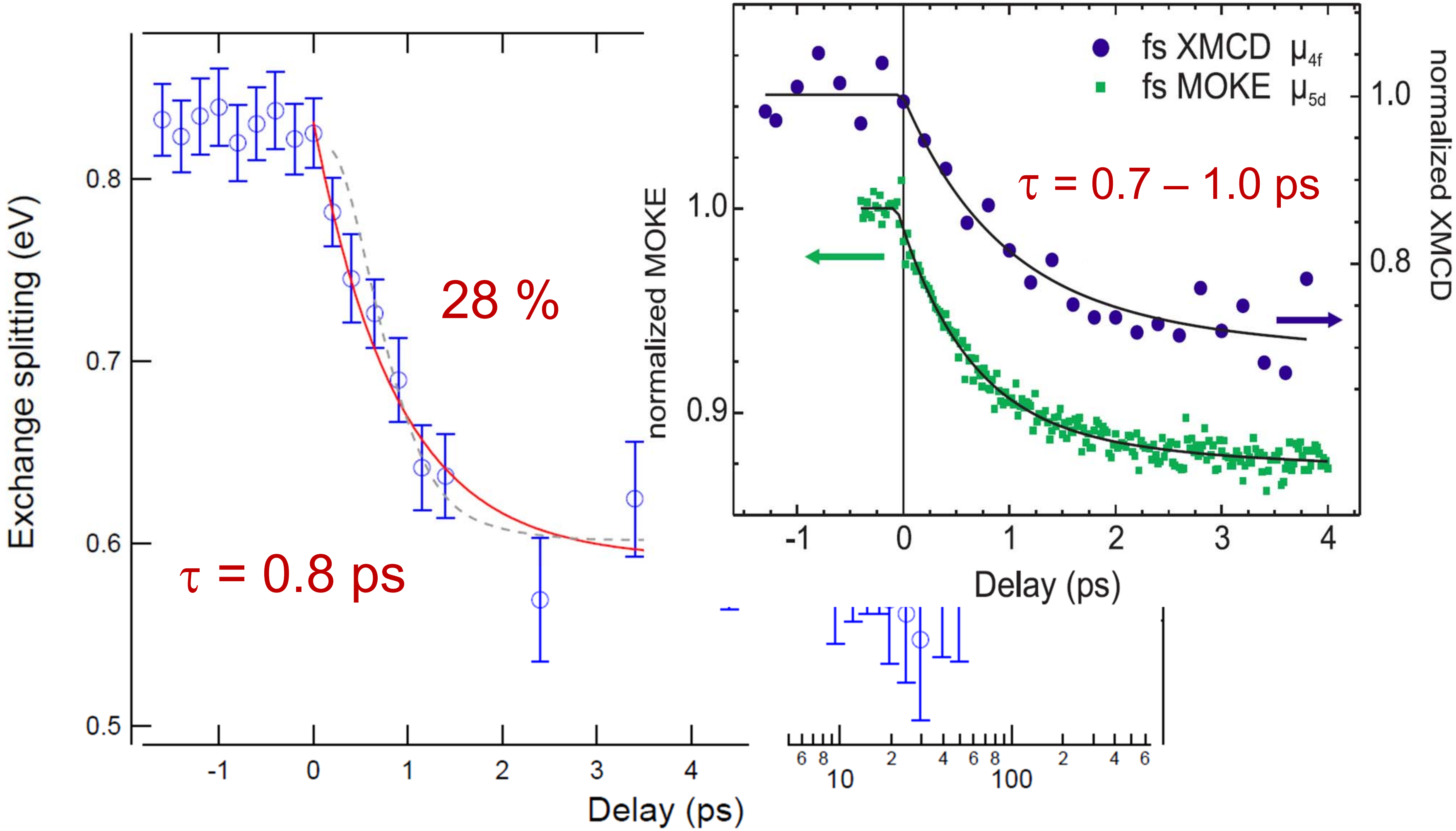
Optically-driven demagnetization (1.2 mJ / cm²)



Hysteresis - Gadolinium (1.2 mJ/cm²)

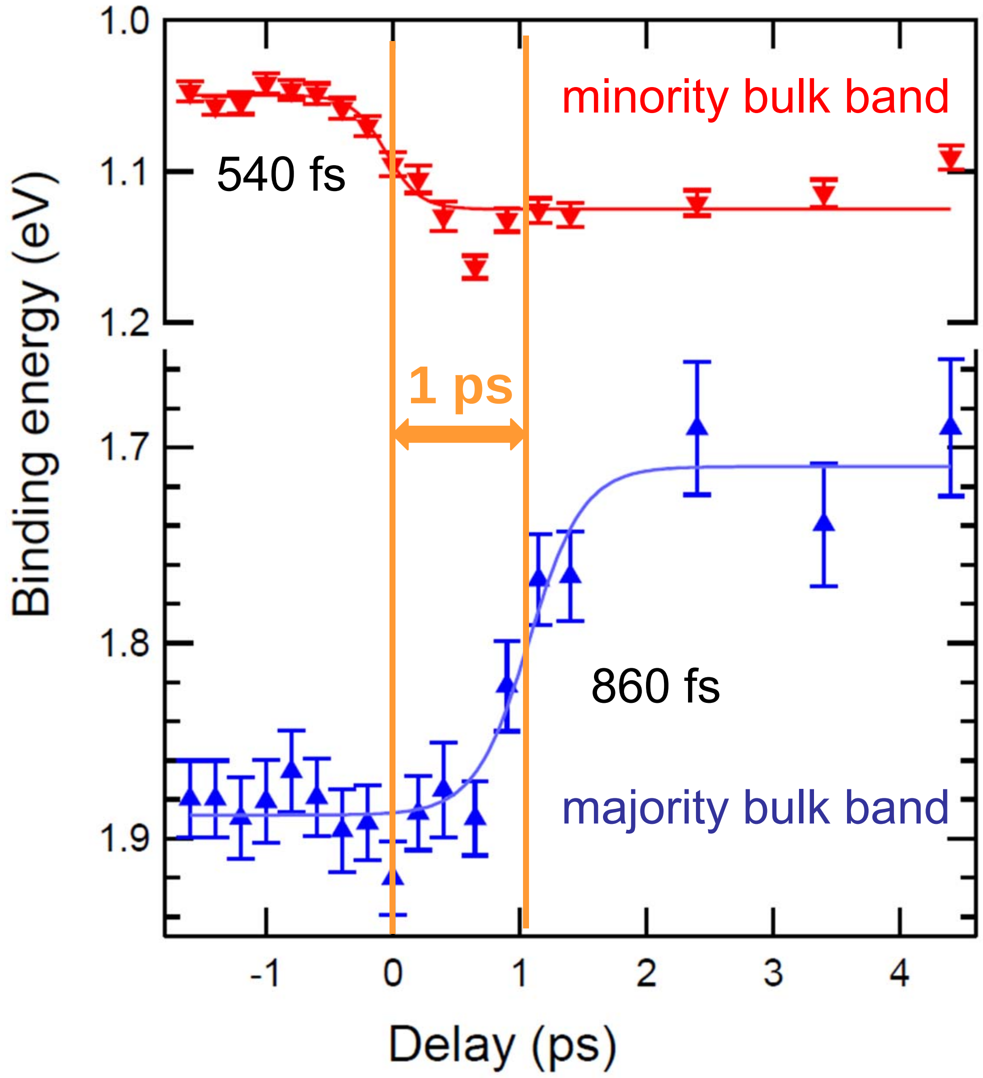


Exchange splitting ΔE_{ex}



Transient Δ_{ex} is a measure of (de)magnetization dynamics !

Non-equilibrium dynamics (1.2 mJ / cm²)



Minority spin band responds faster than majority spin band

Scientific Case

- Photoemission gives detailed insight into the transient electronic structure
- Transform-limited 10 fs Gaussian pulse → 200 meV energy resolution ok
- Ideally tunable and transform limited
- Pump-probe scheme and synchronisation ?
- Angle-resolved photoemission at higher photon energies, e.g., 200 eV
  less space charge, more k-space, complete dispersion $E(k_x,k_y,k_z)$
- Brillouin zone maps require efficient detection