A soft X-ray view on ultrafast demagnetization

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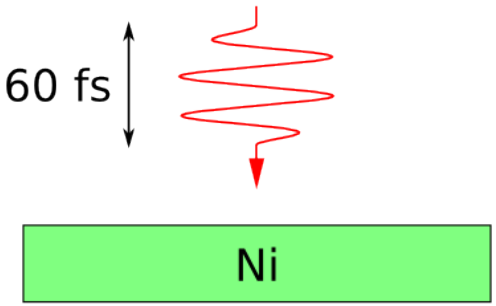
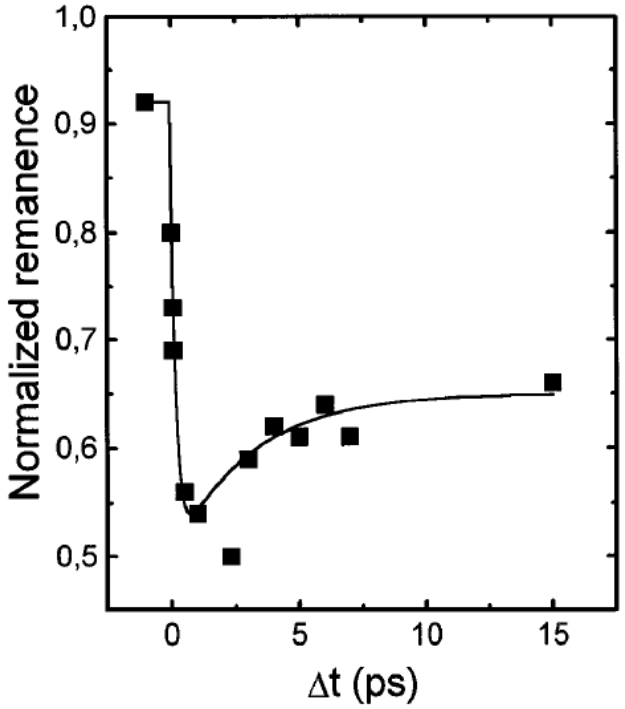
Palaiseau, France

Motivation : understanding ultrafast demagnetization





Beaurepaire et al., Phys. Rev. Lett., 76, 4250 (1996), Kirilyuk 82, 2731 (2010)

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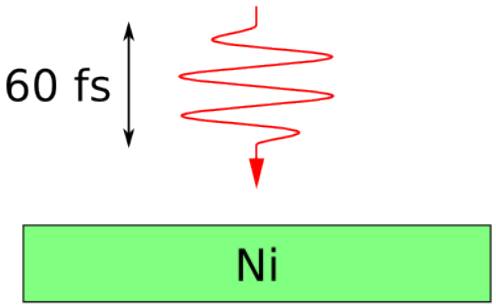

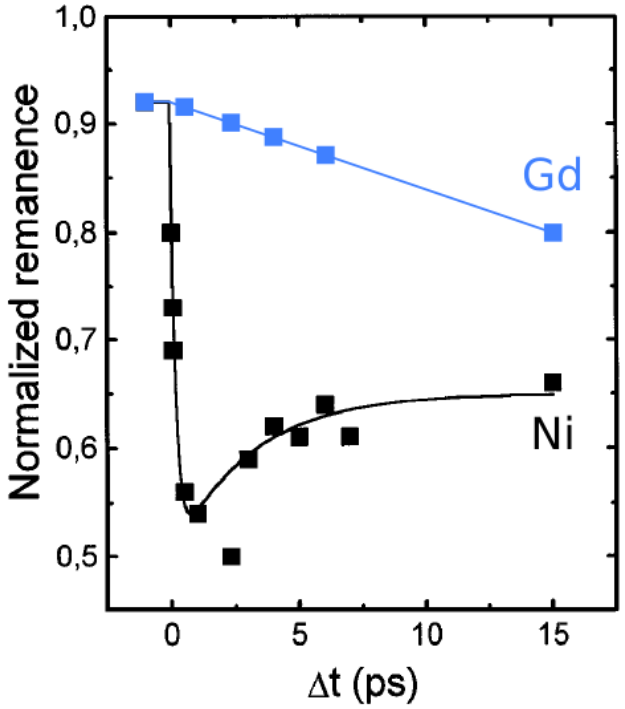

- contradiction with previous observations (and expectations)

Beaurepaire et al., Phys. Rev. Lett., 76, 4250 (1996), Kirilyuk 82, 2731 (2010)
Vaterlaus et al., Phys. Rev. Lett., 67, 3314 (1991)

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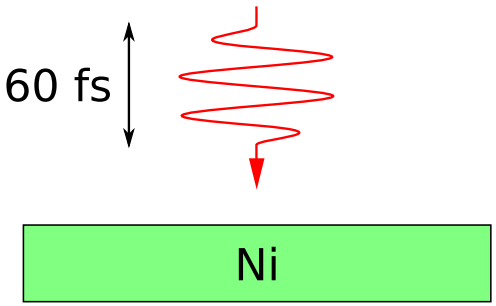

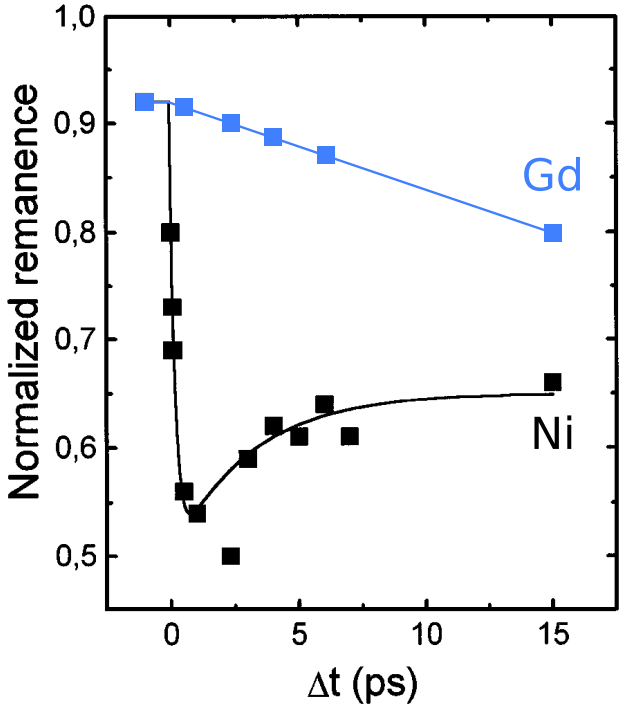

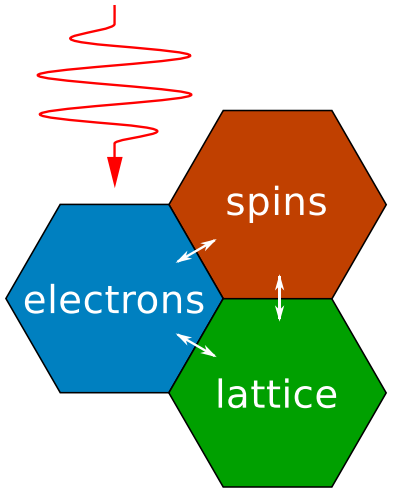

- contradiction with previous observations (and expectations)
- no satisfactory physical explanation (angular momentum transfer ?)

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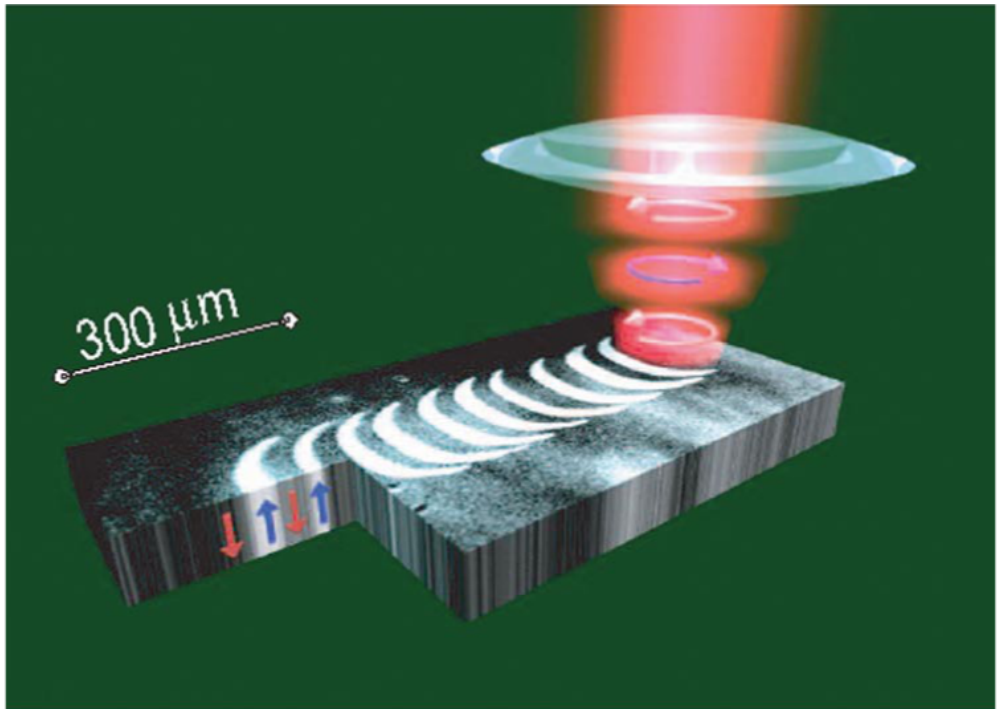
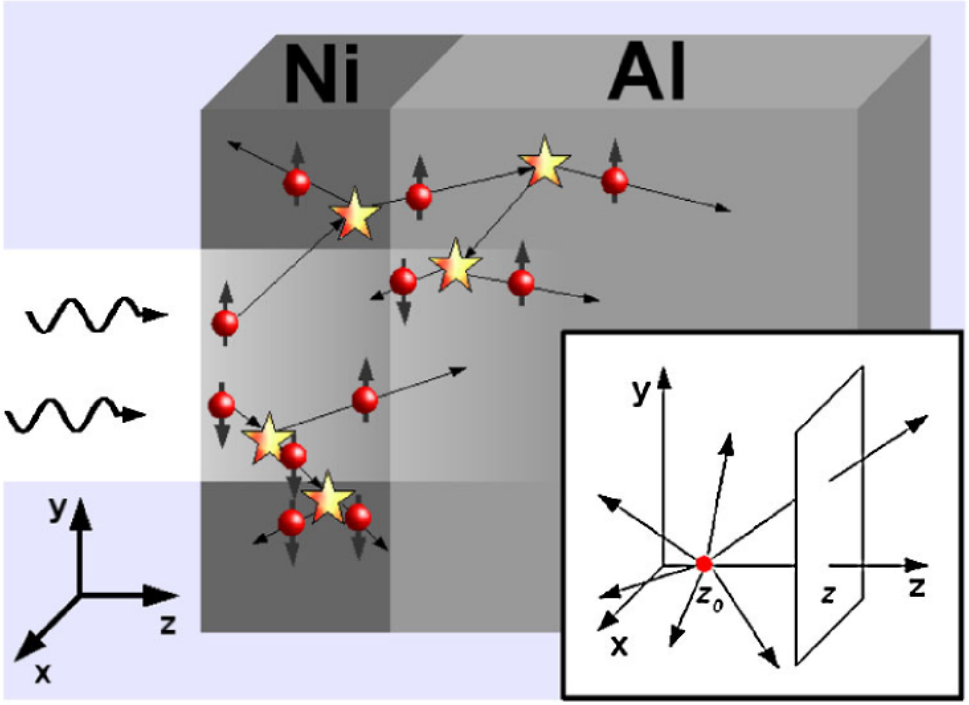


- contradiction with previous observations (and expectations)
- no satisfactory physical explanation (angular momentum transfer ?)
- femtosecond control of magnetization (application)

Beaurepaire et al., Phys. Rev. Lett., 76, 4250 (1996), Kirilyuk 82, 2731 (2010)
Vaterlaus et al., Phys. Rev. Lett., 67, 3314 (1991)
Stanciu et al., Phys. Rev. Lett., 98, 207401 (2007)

Superdiffusive spin transport



Direct spin transfer (hot electrons)

- conservation of angular momentum
- can be tested experimentally

Battiato et al., Phys. Rev. Lett., 105, 027203 (2010)

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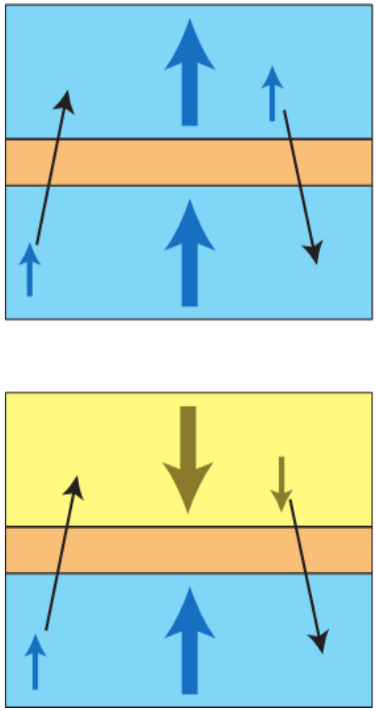

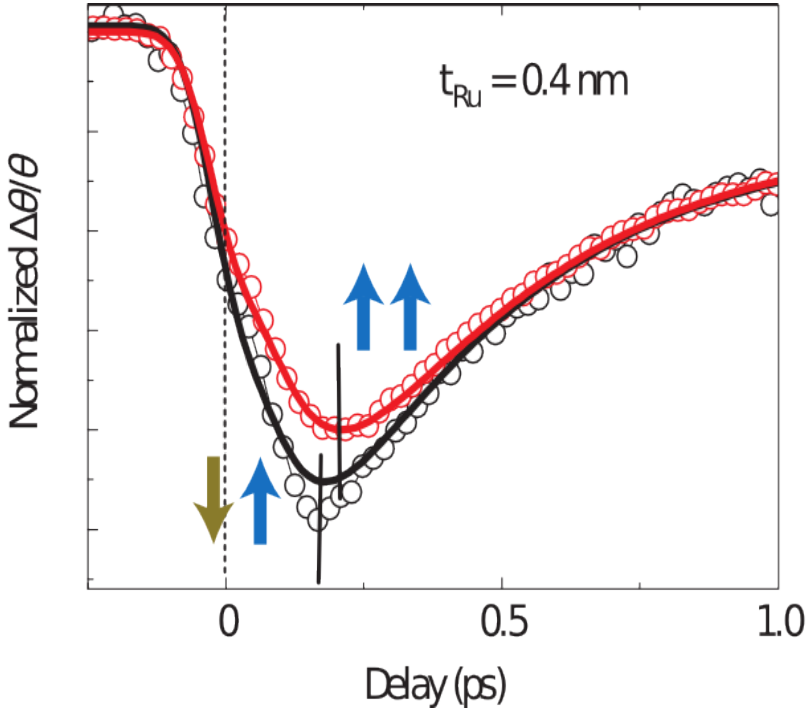

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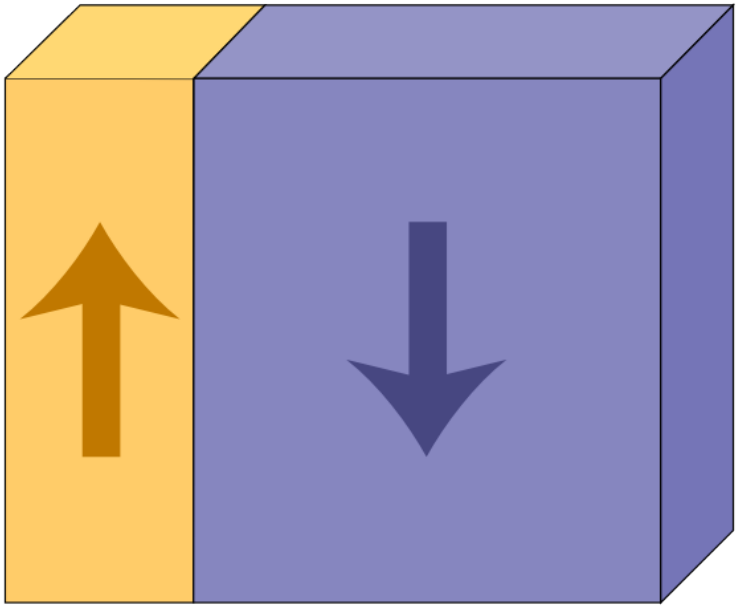
Malinowski et al., Nature Phys., 4, 855, (2008)

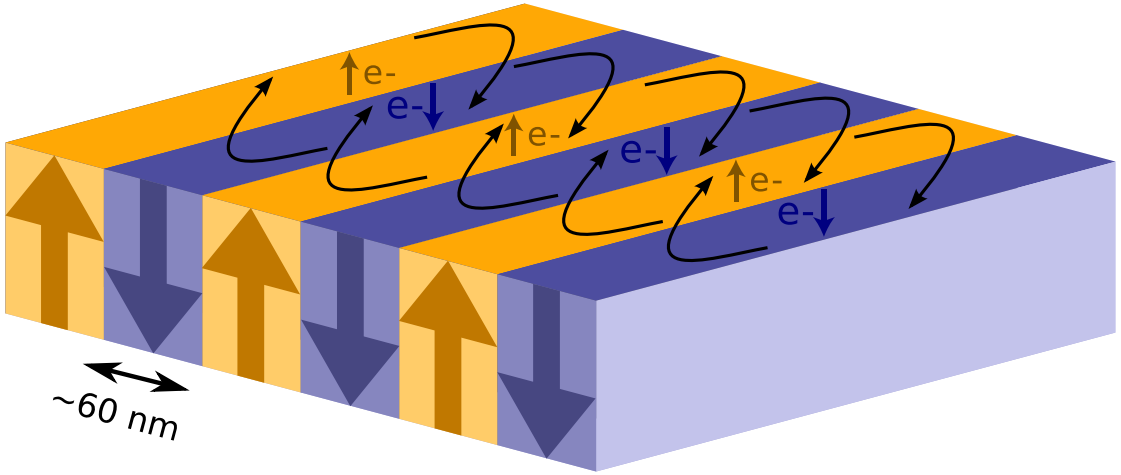
How to experimentally observe direct spin transfer ?



Antiparallel bilayer configuration

- higher demagnetization efficiency

How to experimentally observe direct spin transfer ?



Appears in multilayers with perpendicular magnetic anisotropy

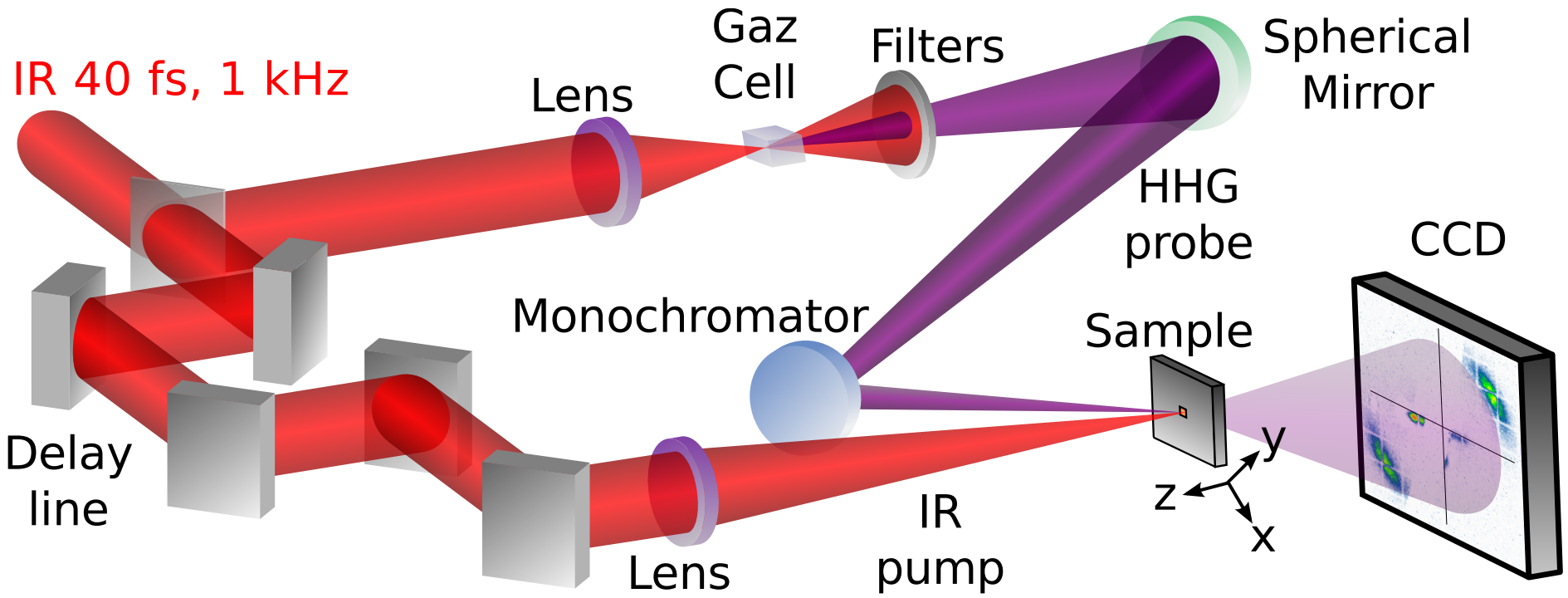
- antiparallel nanometric magnetic domains

Requires pulse duration < 50 fs $+$ wavelengths < 100 nm

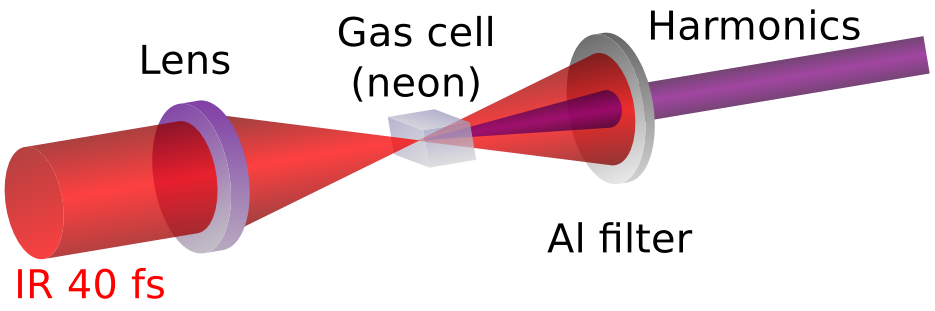
$\Rightarrow$ femtosecond soft X-rays sources

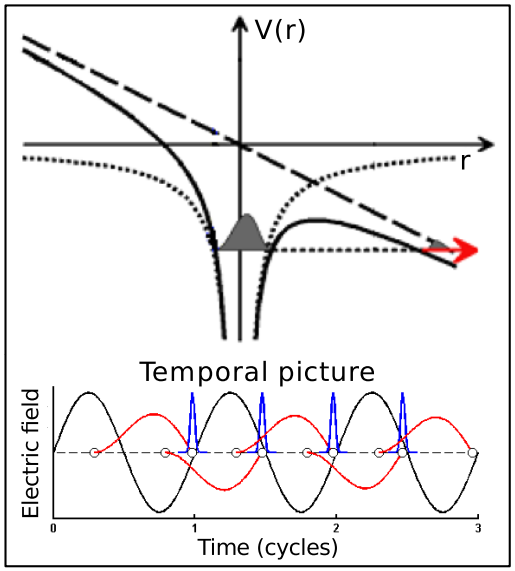
Experimental setup for the study of Co/Pd multilayers

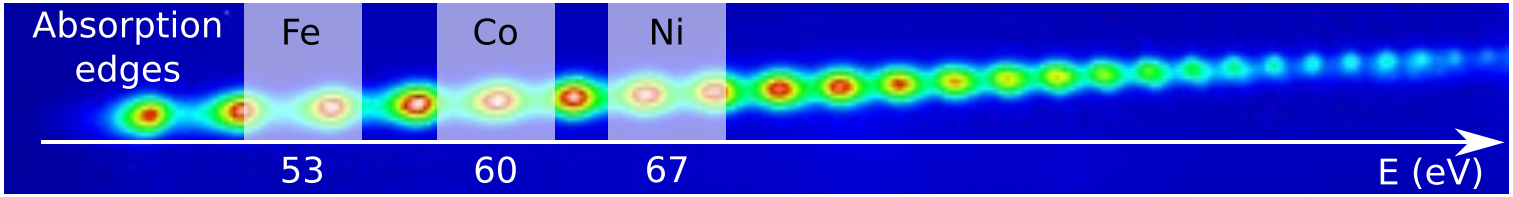
IR pump/soft X-ray probe geometry



High order harmonics generation (HHG)





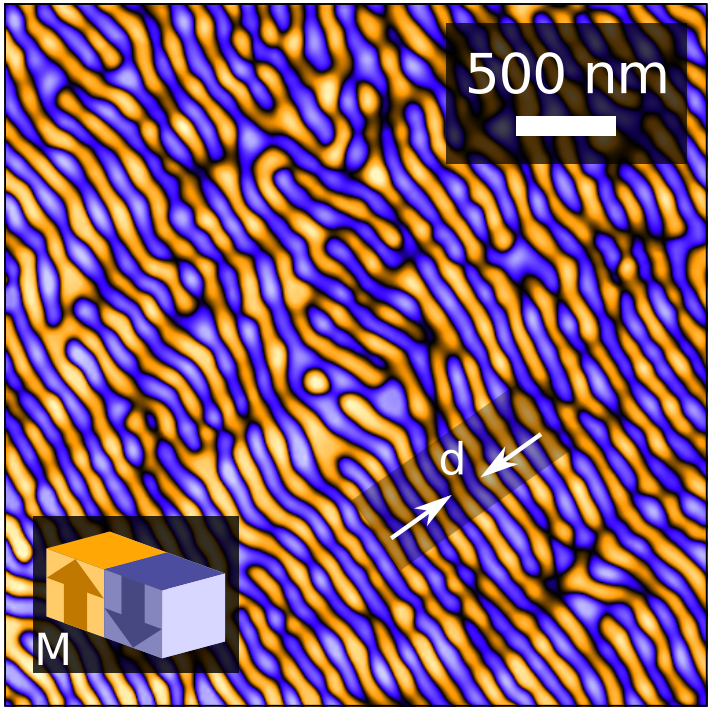


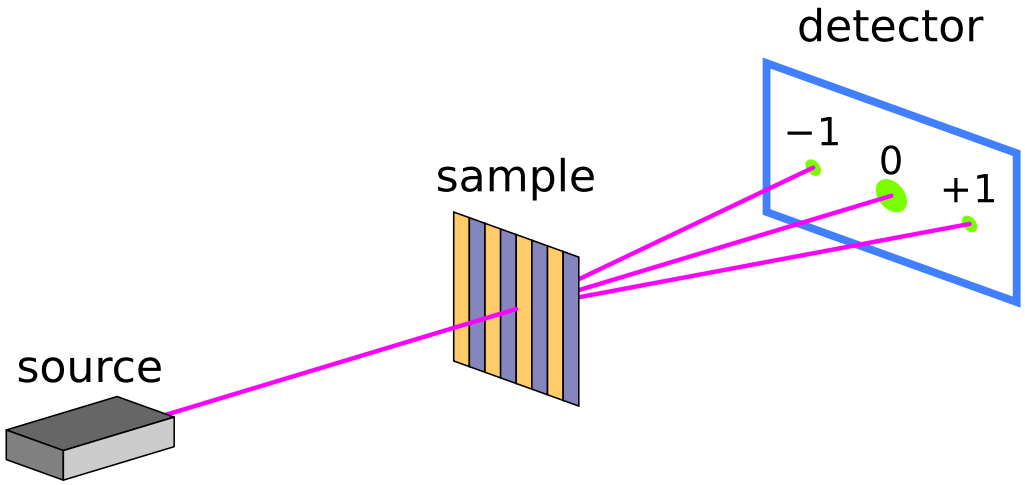
HHG properties :

- short pulses (attosecond !)
- energies up to 150 eV (but can go up to 2 keV !), short wavelenghts
- tunable, element specificity
- coherent, no jitter with IR, linearly polarized,etc

Magnetic resonant scattering

Magnetic contratst : XMCD at the Co *M* edge (60 eV, 20 nm)





Magnetic domain network = magnetic diffraction grating

Magnetic resonant scattering

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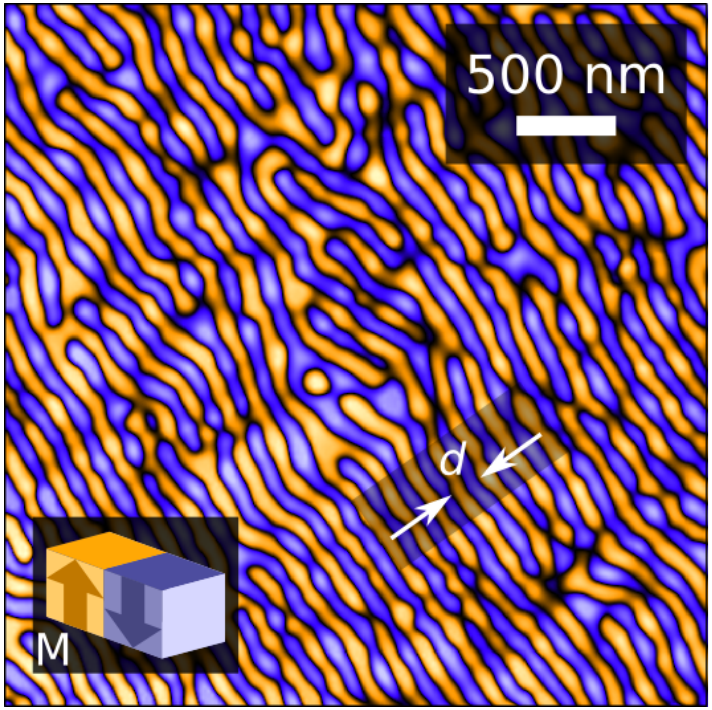
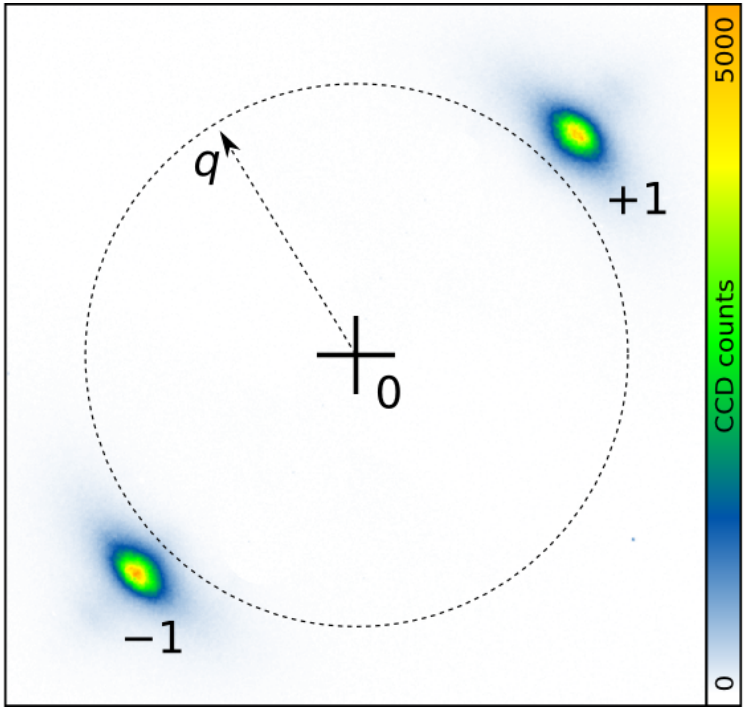




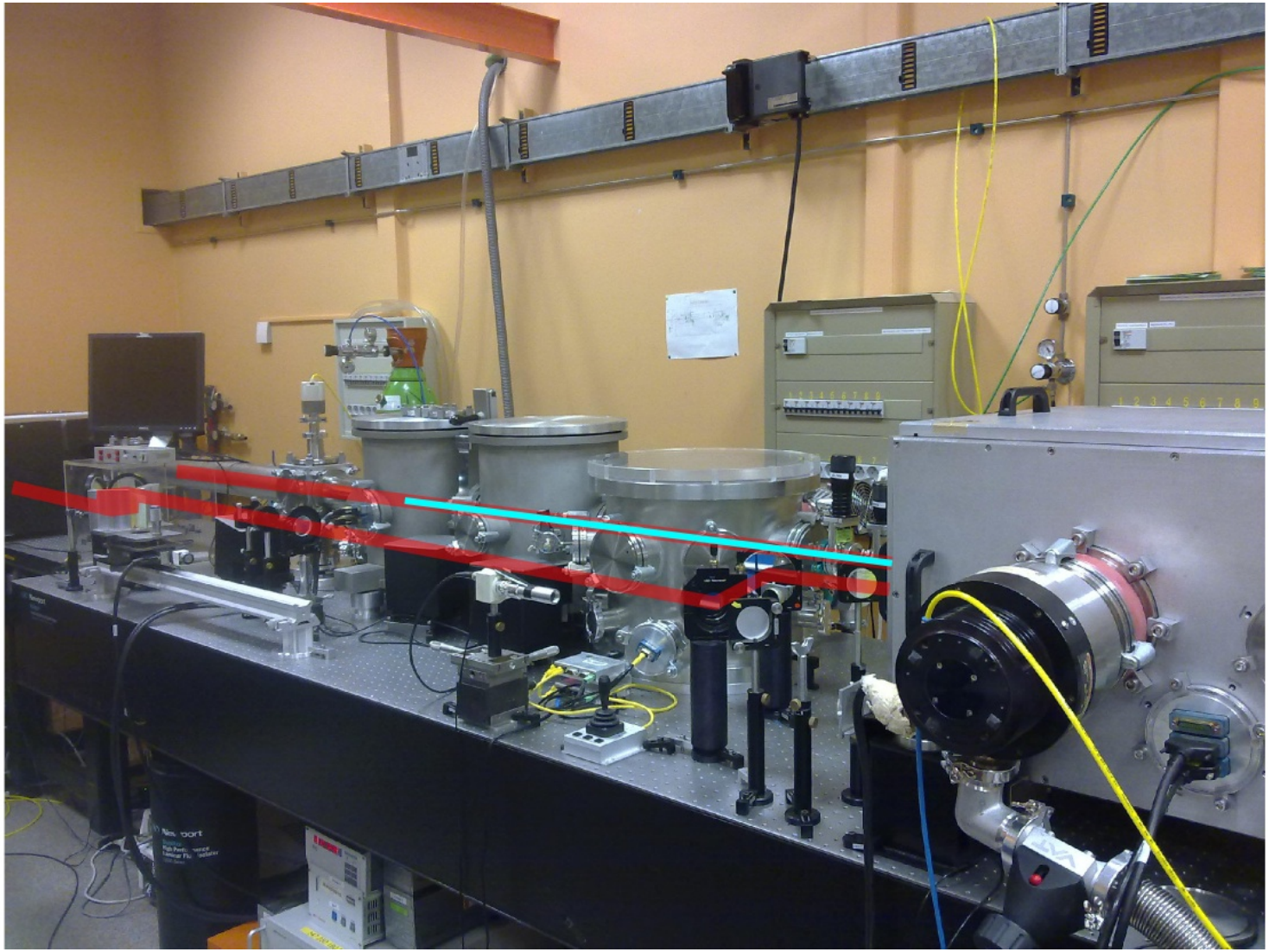
Magnetic domain network = magnetic diffraction grating

- scattering intensity → domains magnetization ($I \sim M^2$)
- spots position and shape → domains size and orientation ($q \sim \frac{1}{d}$)

Linear polarization ok ($L = H^+ + H^-$) : identical $H+$ and $H-$ patterns

Vodungbo et al., EPL, 94, 54003 (2011)

Experimental setup



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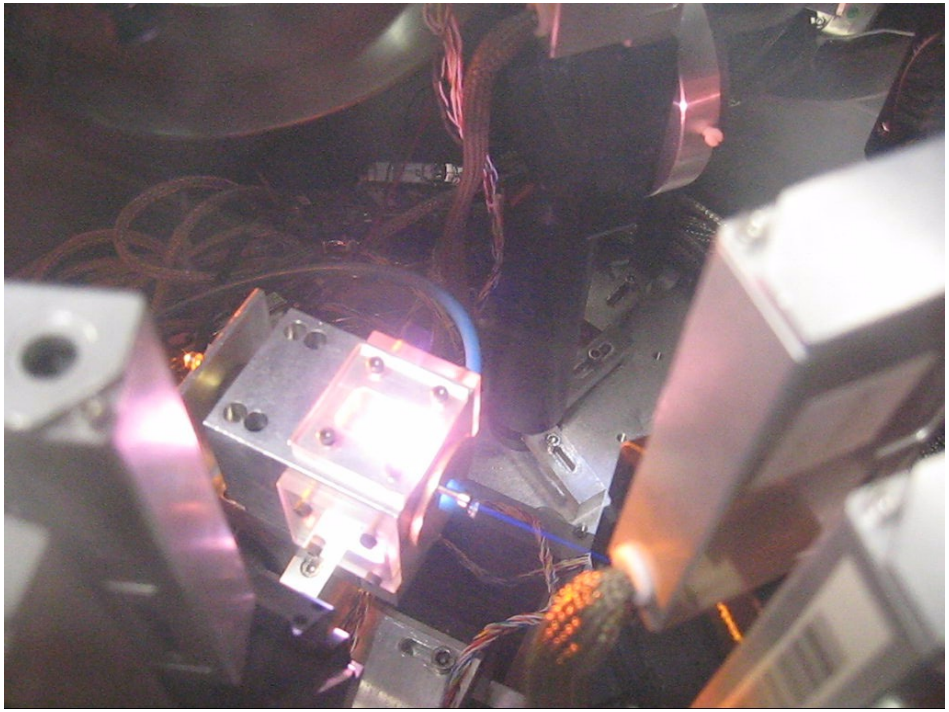

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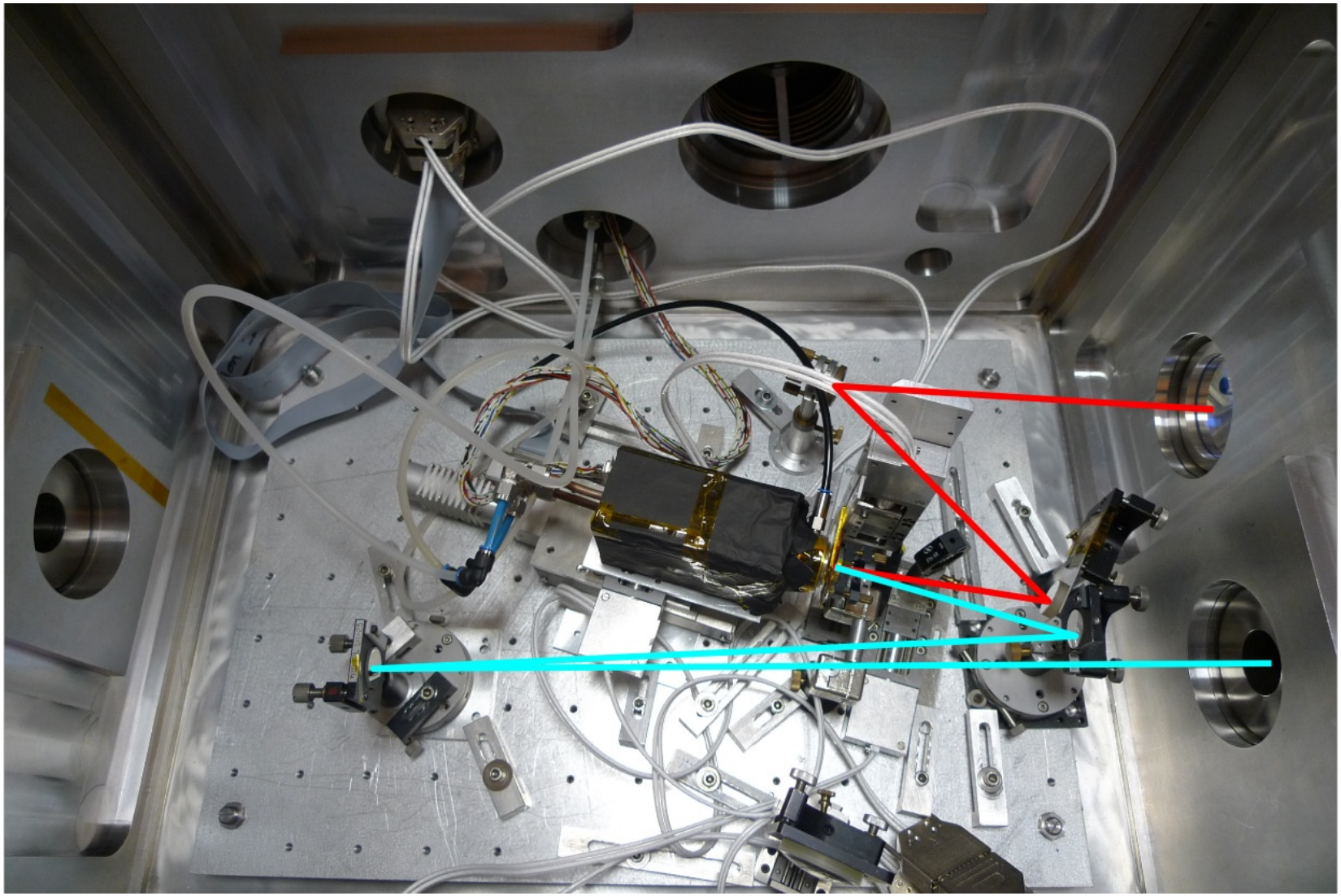
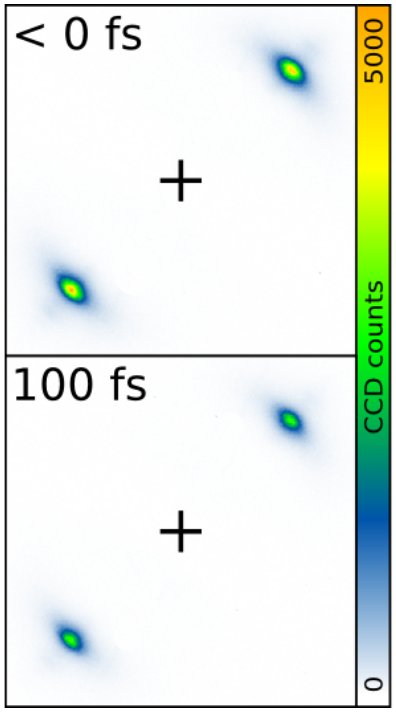
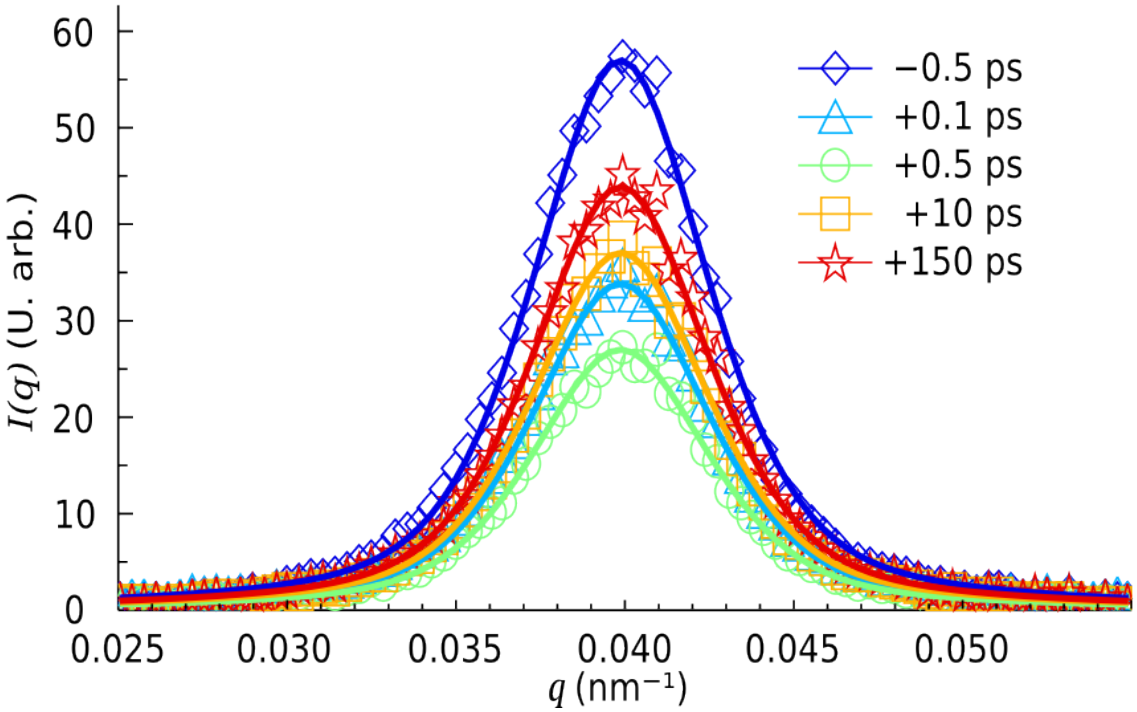


Ultrafast demagnetization at the magnetic domains level





Vodungbo et al., Nat. Commun., 3, 999 (2012)

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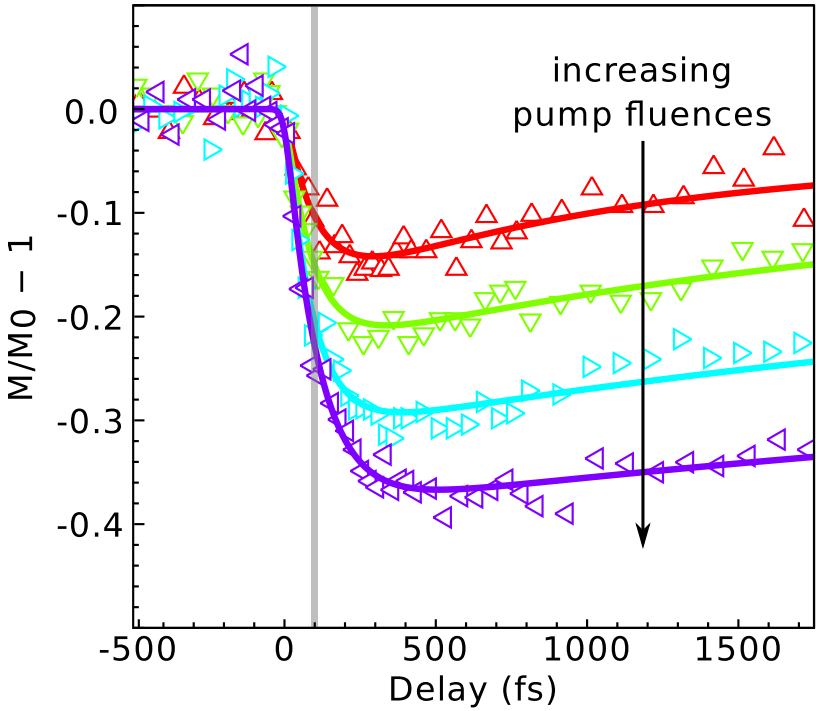

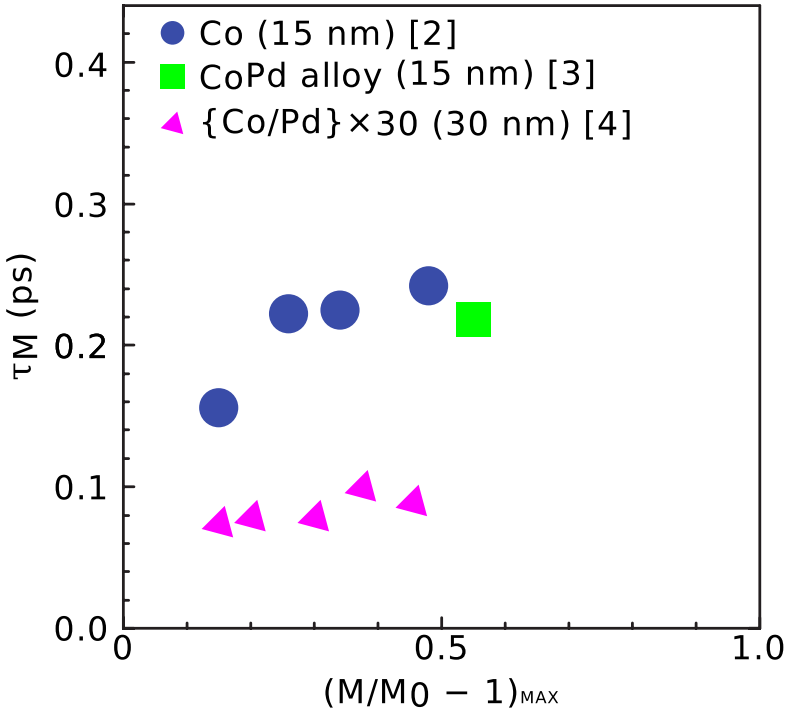

- faster demagnetization ($\sim$ 100 fs) than without domains ($>$ 200 fs)

Vodungbo et al., Nat. Commun., 3, 999 (2012)

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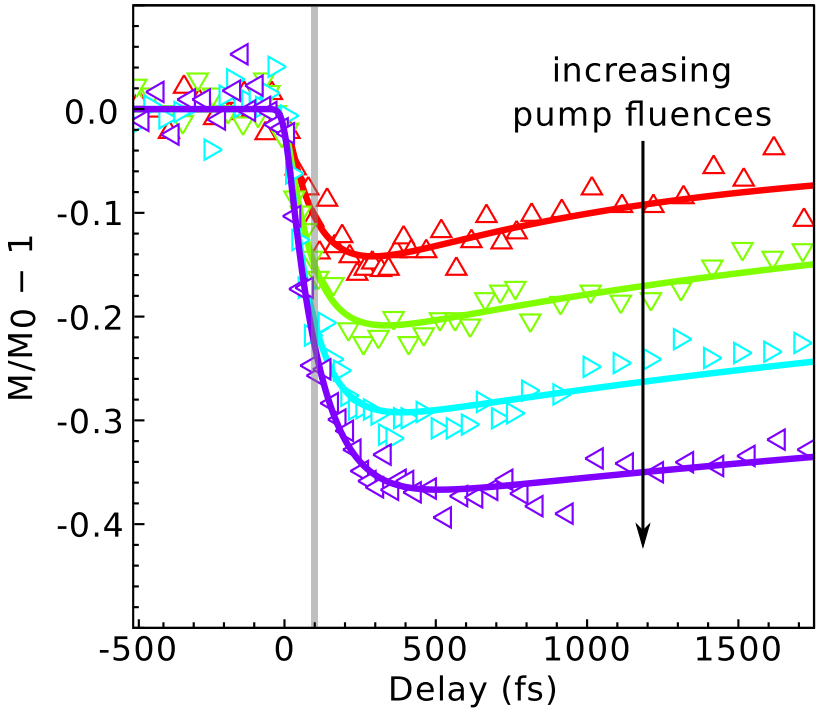
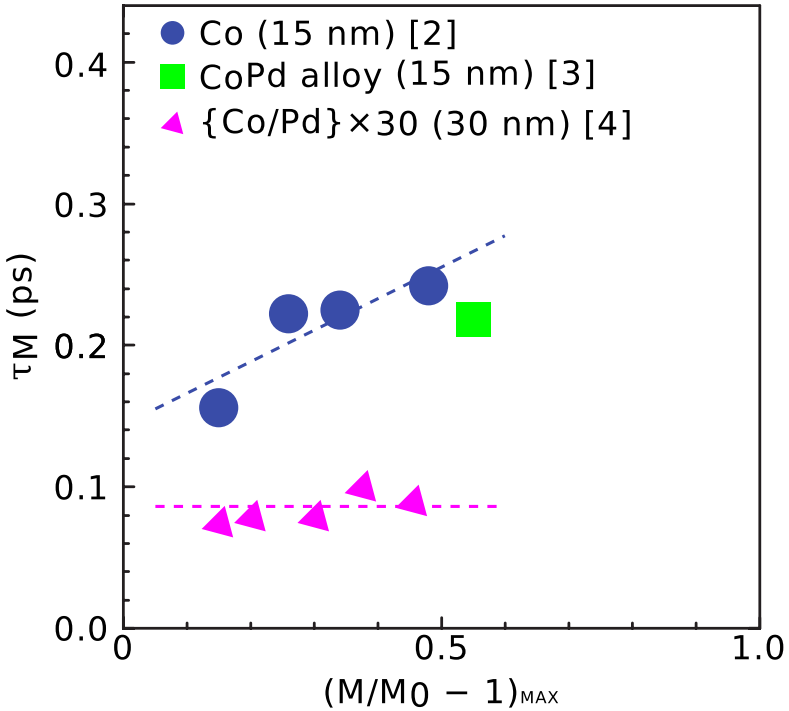


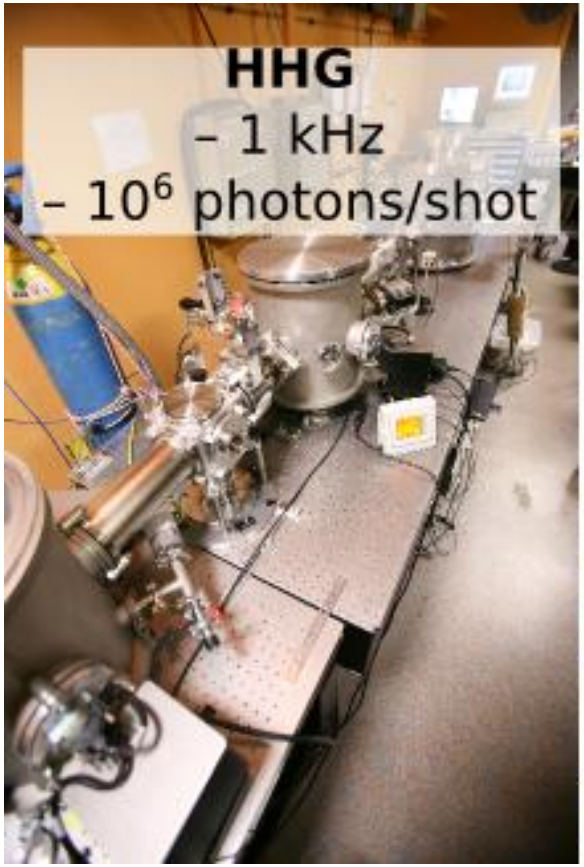


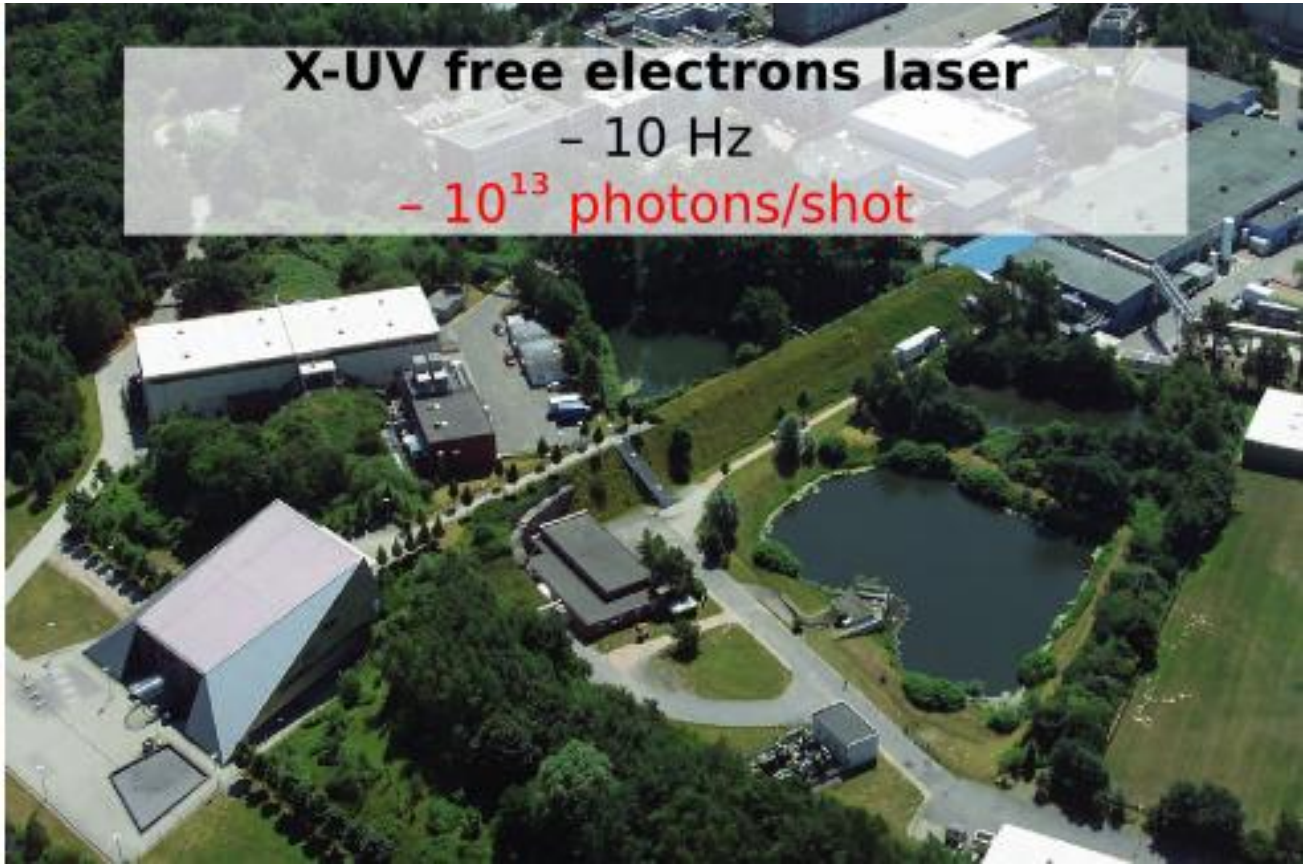
- faster demagnetization ($\sim$ 100 fs) than without domains ($>$ 200 fs)
- demagnetization time independent of the pump fluence

$\Rightarrow$ Consistent with direct spin transfer between domains

Vodungbo et al., Nat. Commun., 3, 999 (2012)

From kHz (HHG) to single shot (XFEL) experiments



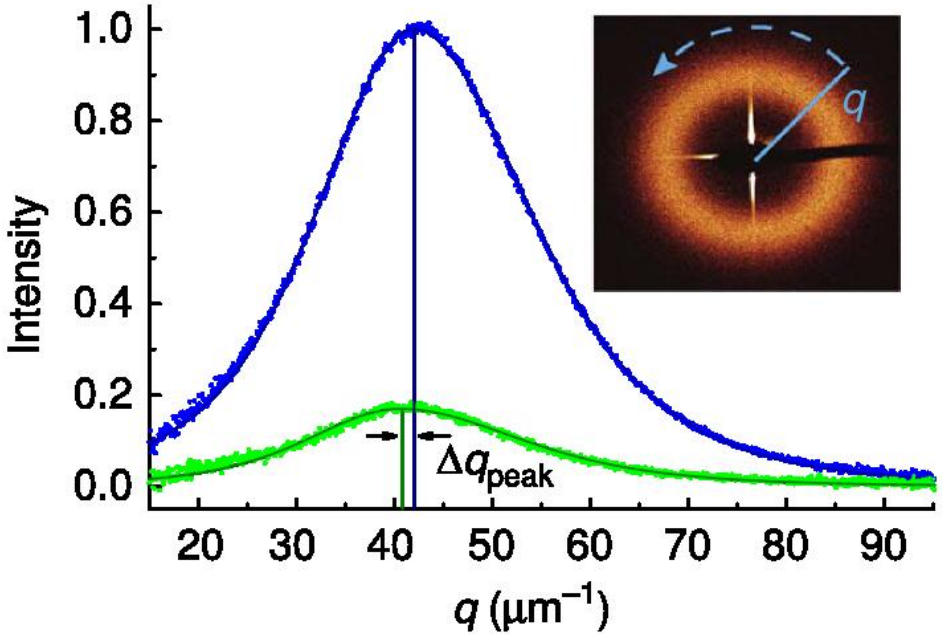


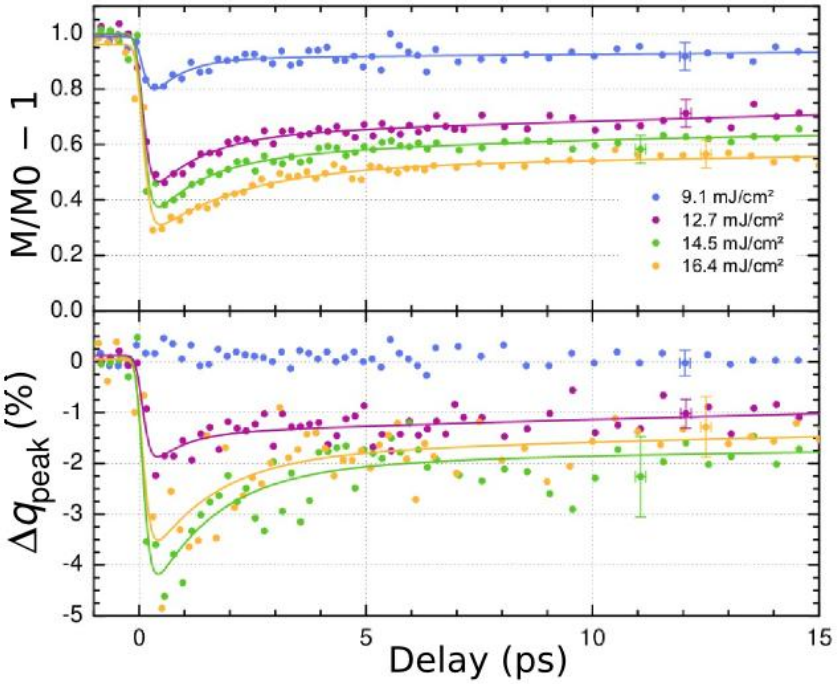
New perspectives

- very high pump fluence ($>$ destruction treshold)
- irreversible phenomena

Demagnetization in the high fluence regime

International collaboration (FLASH, Hamburg)





Shift of the magnetic scattering peak

- ultrafast modification of the magnetic domain structure

Pfau et al., Nat. Commun., 3, 1100 (2012)

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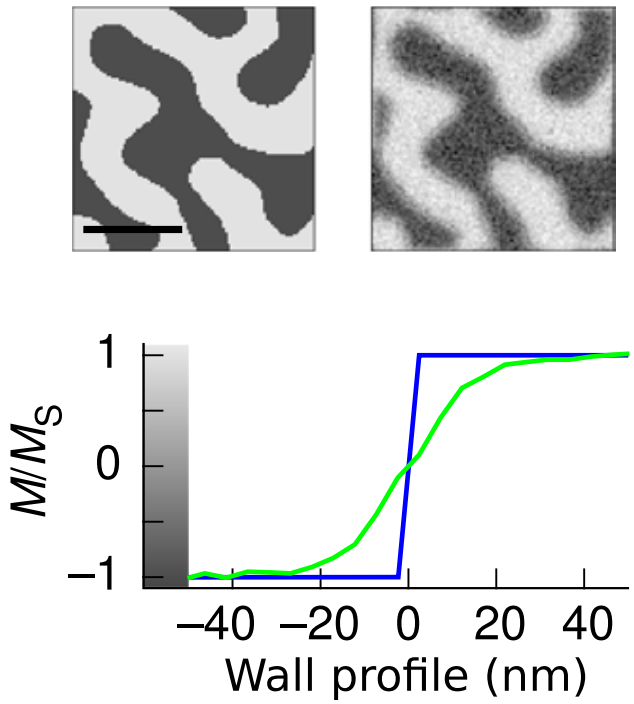
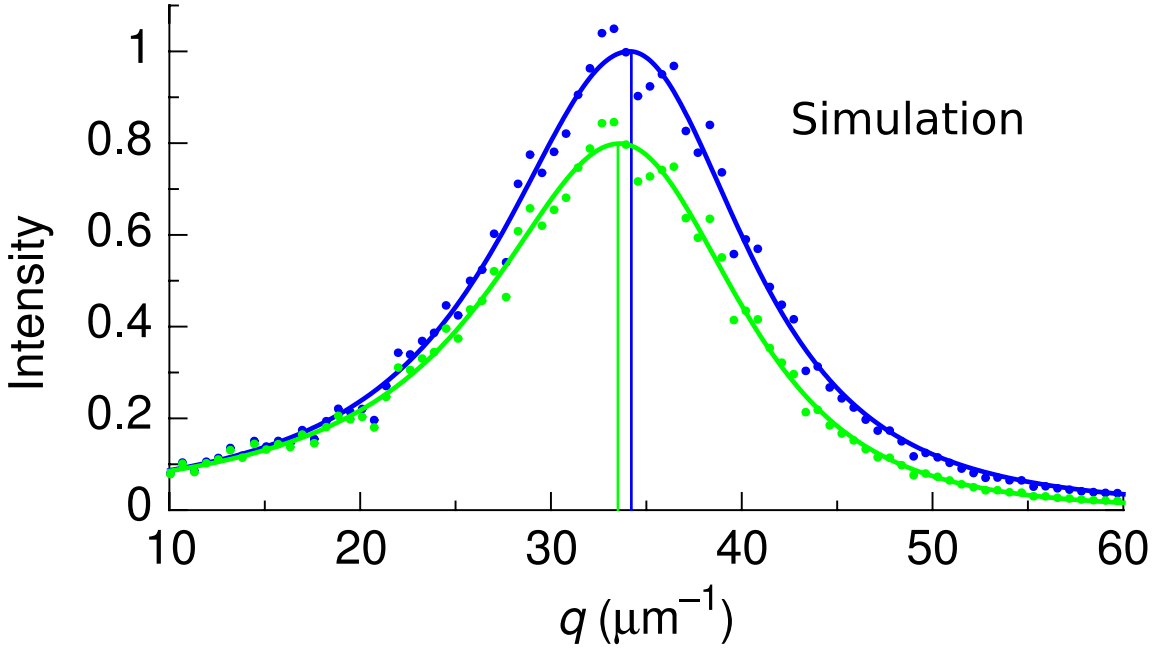




Shift of the magnetic scattering peak

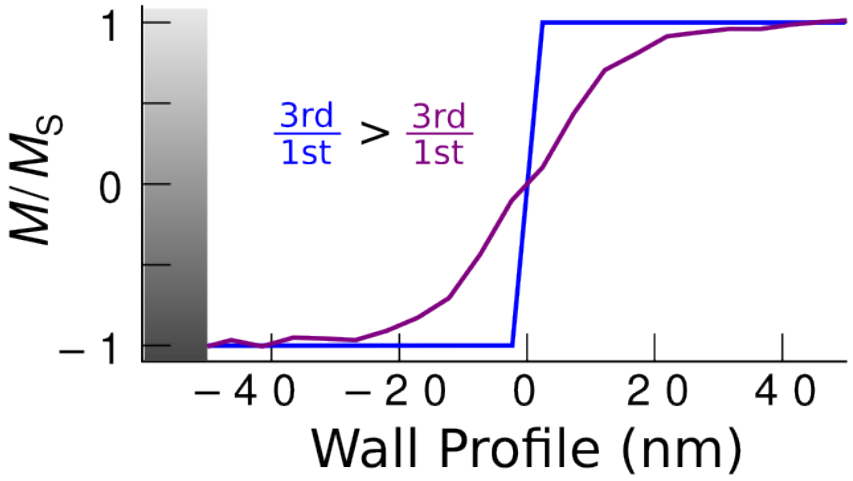
- ultrafast modification of the magnetic domain structure
- good agreement with simulation of electron motion

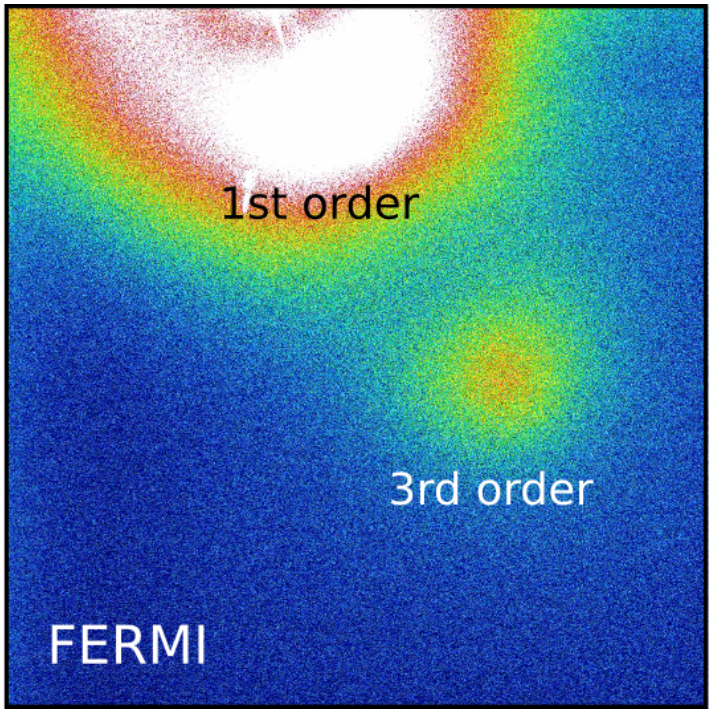
⇒ Confirmation of spin transfer between domains

Pfau et al., Nat. Commun., 3, 1100 (2012)

Can we get more informations ?

Observation of higher scattering orders





More precise measurement of the domain wall profile

- first beamtime at FERMI (Trieste), muliple-shots, 02/2013
- new beamtime at LCLS, single-shot, 03/2014

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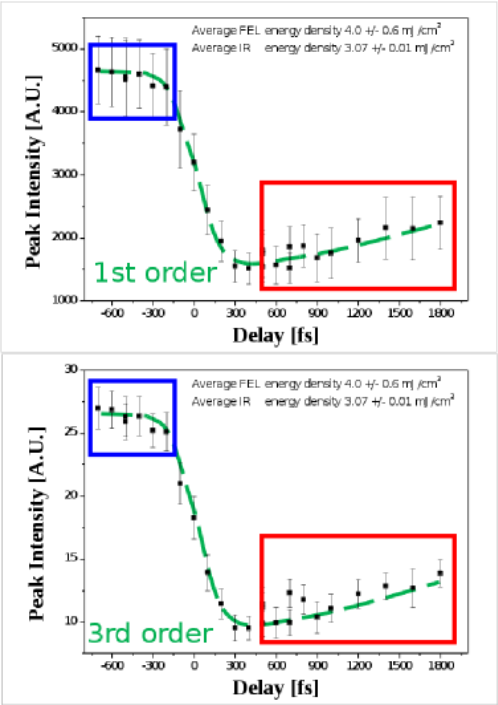

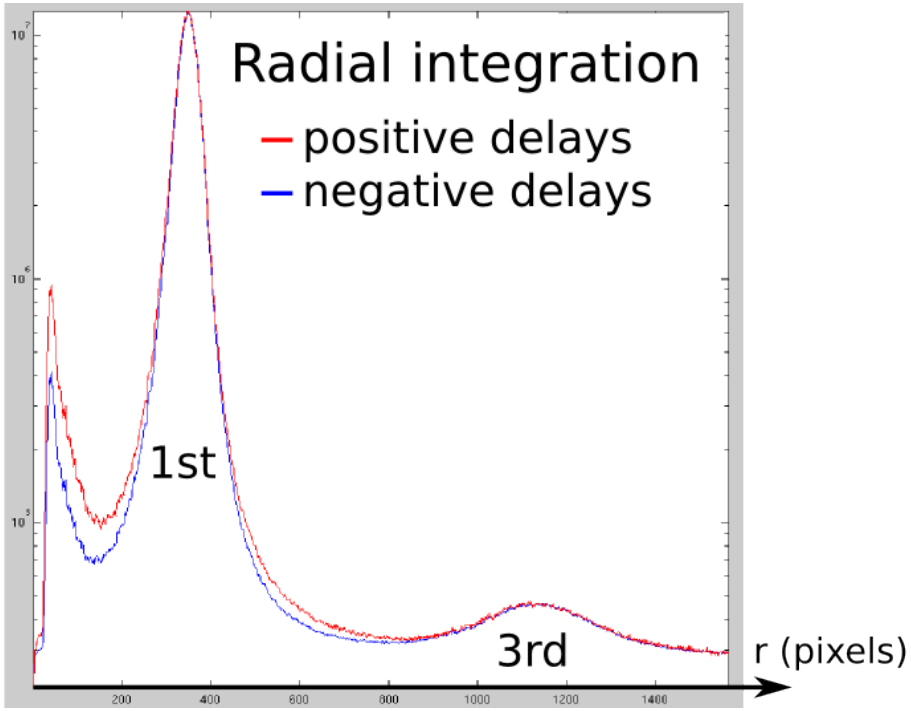

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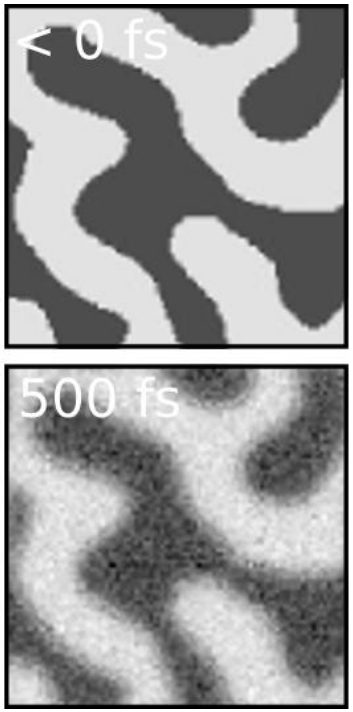
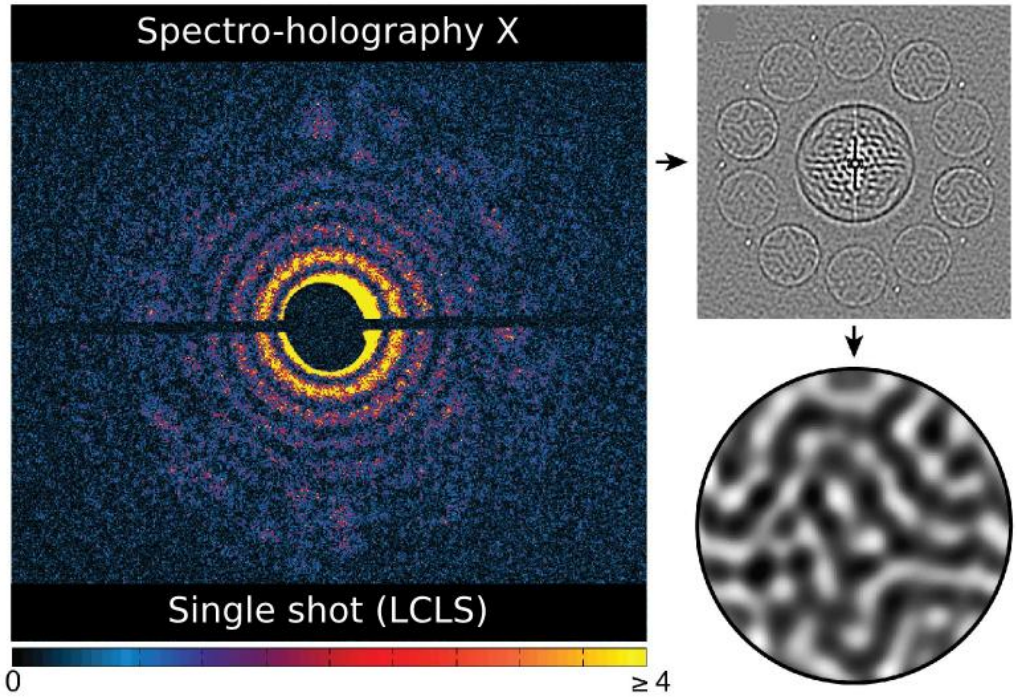
Real space observation of the domain wall evolution



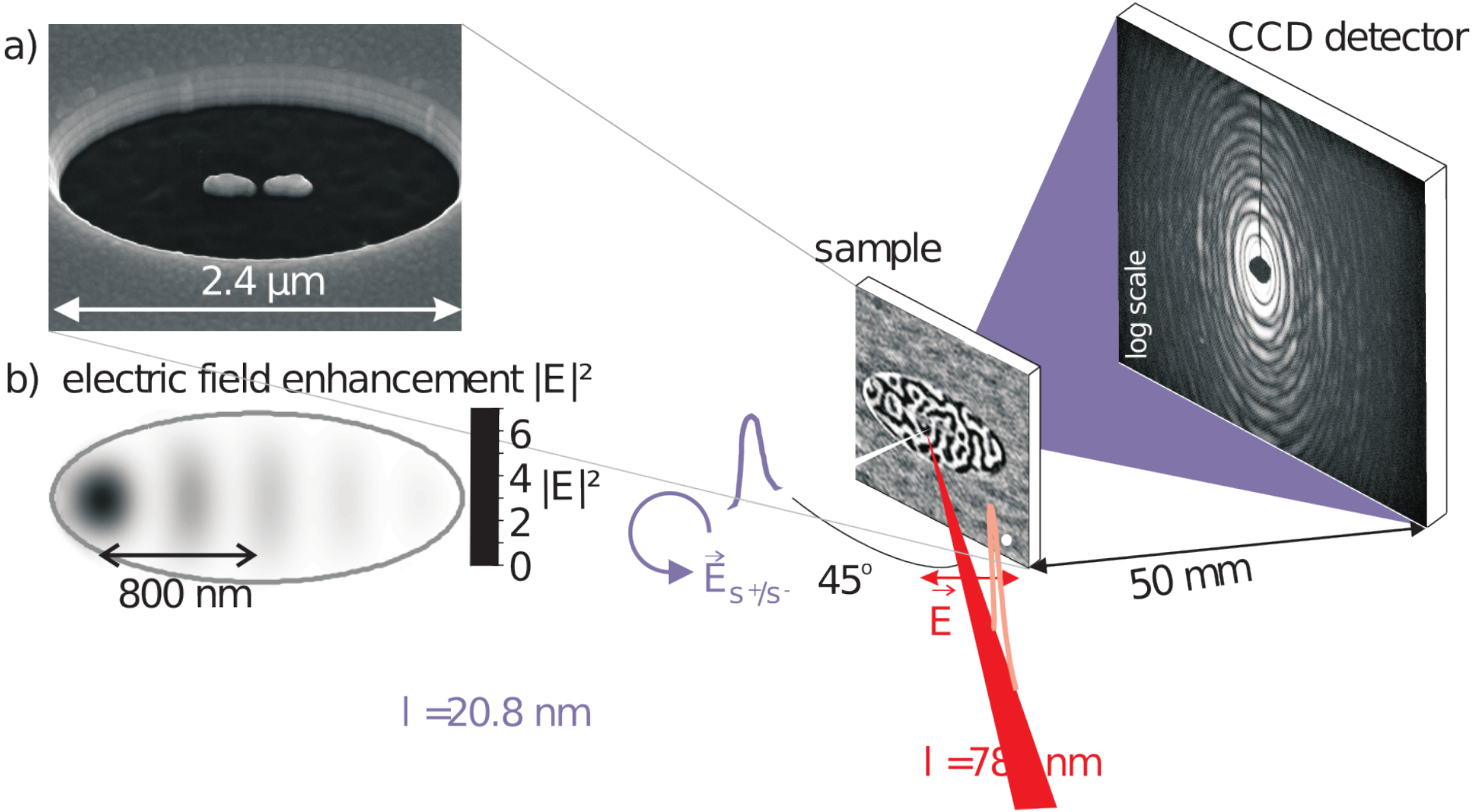


Time resolved magnetic imaging

- single-shot magnetic imaging Ok
- to be implemented in pump-probe with better resolution

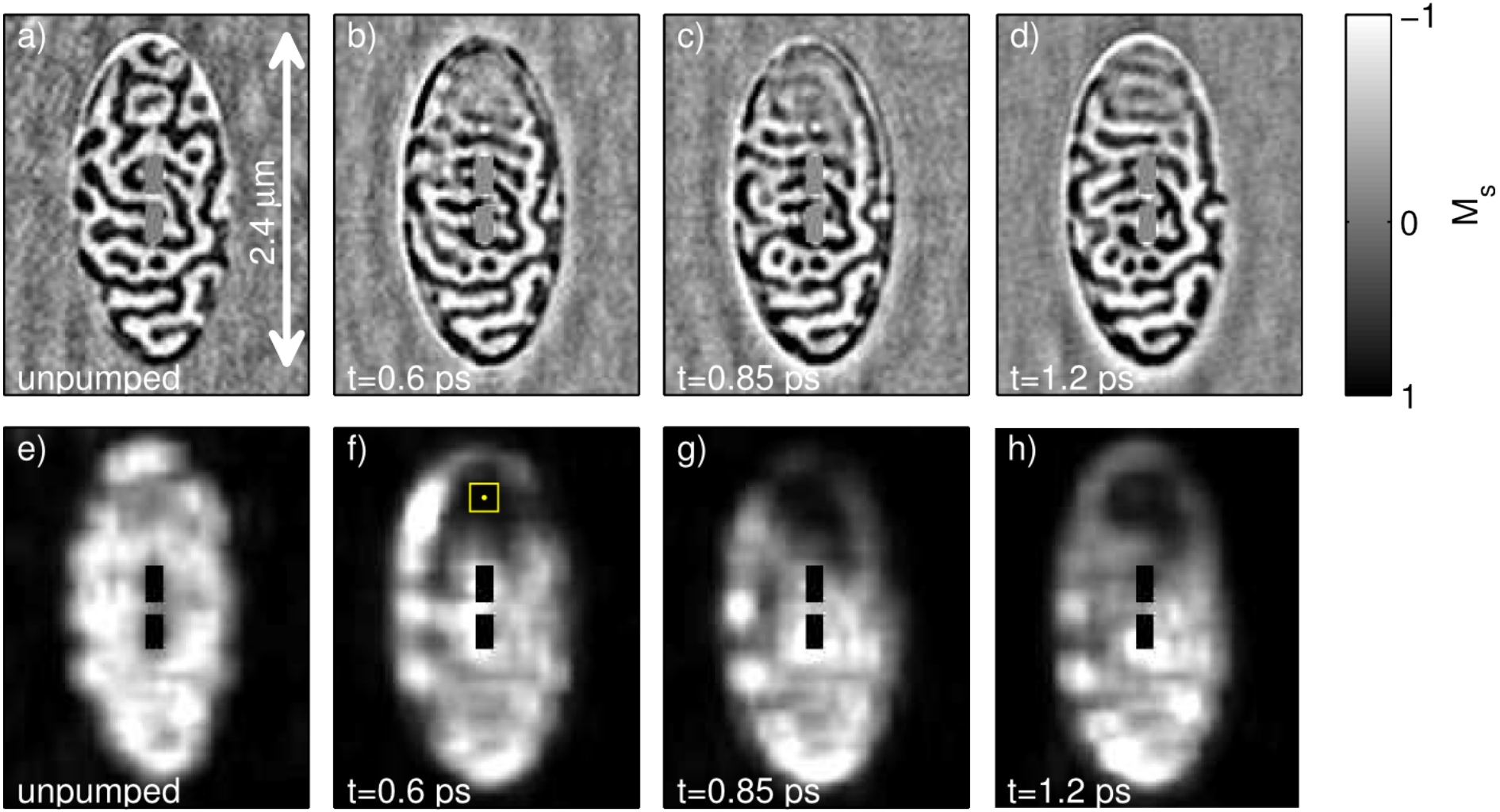
Wang et al., Phys. Rev. Lett., 108, 267403 (2012)

Time-resolved magnetic imaging : first results



Korff Schmising et al., Phys. Rev. Lett., under review

Time-resolved magnetic imaging : first results



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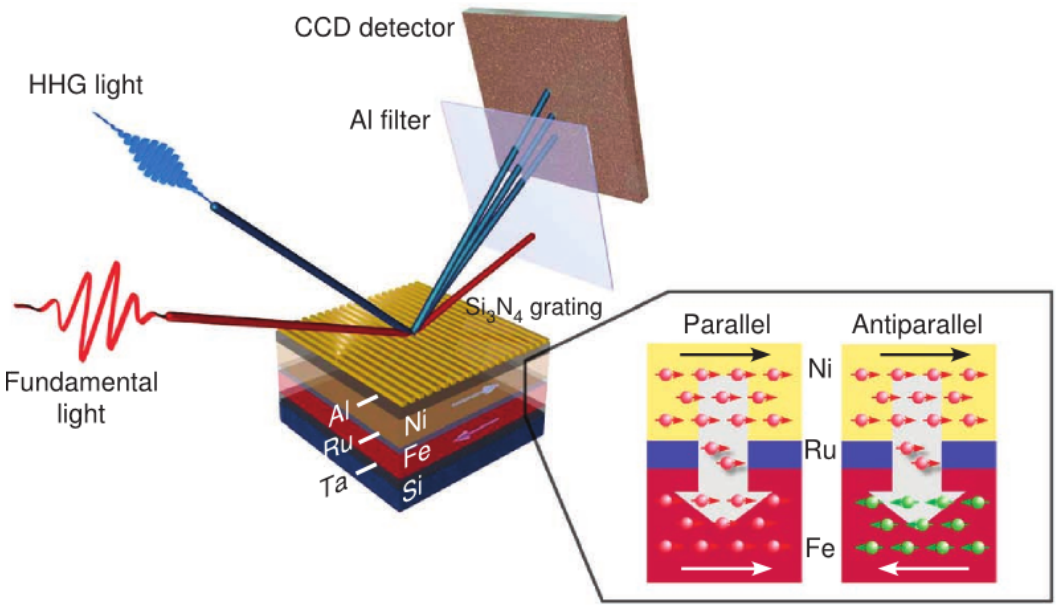
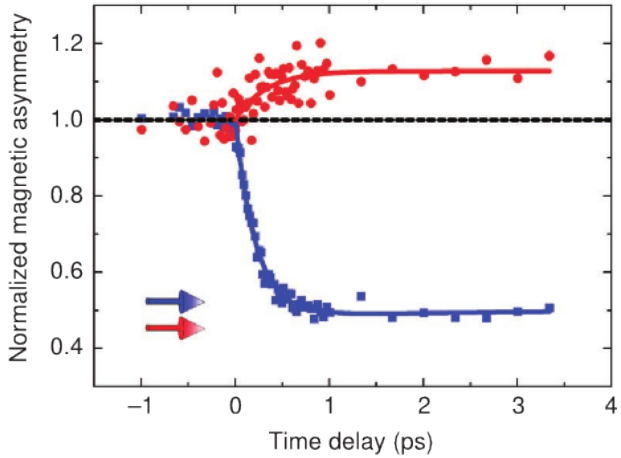
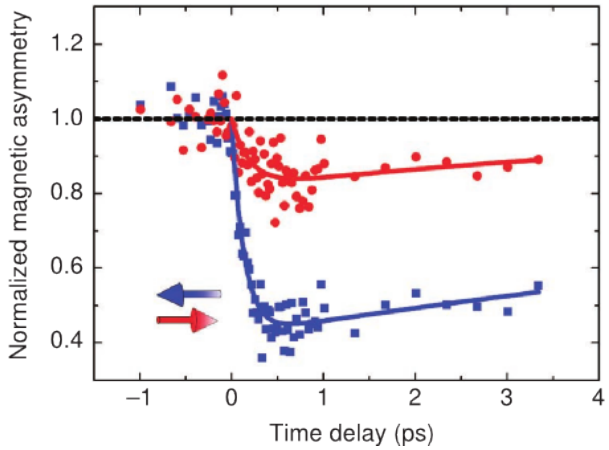
What about perpendicular spin transfer ?





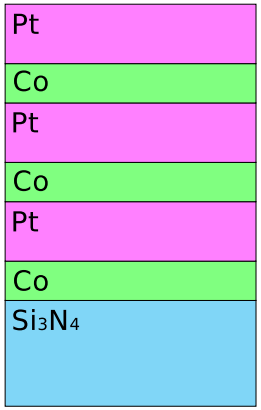


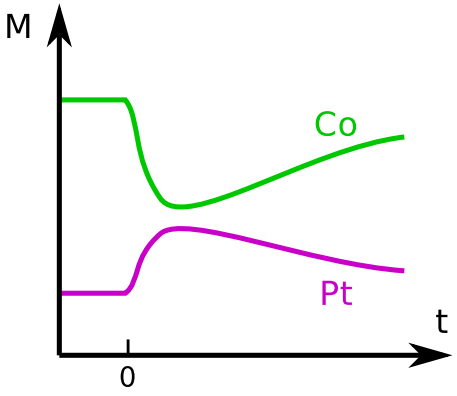
Other recent experimental observations :

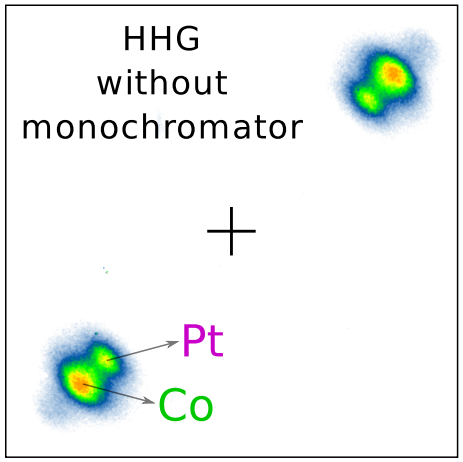
- *Rudolf et al., Nat. Commun., 3, 1037 (2012)*
- *Eschenlohr et al., Nature Mater., 12, 332 (2013)*
- *Kampfrath et al., Nature Nanotechnology, 8, 256 (2013)*

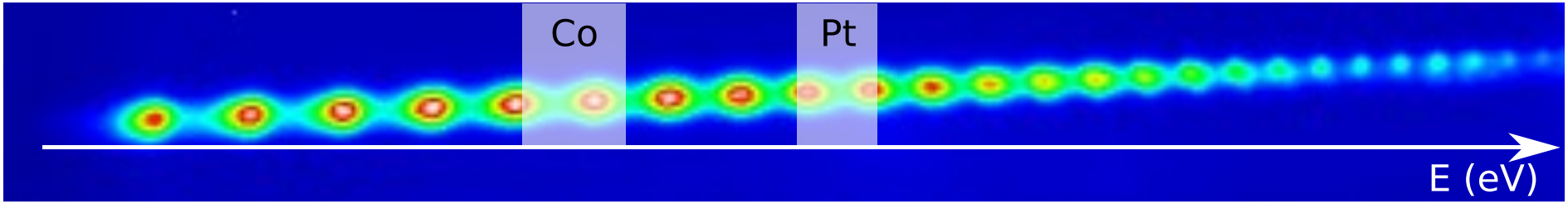
Is spin transfer the only mechanism for ultrafast demagnetization ?

Probing perpendicular spin transfer



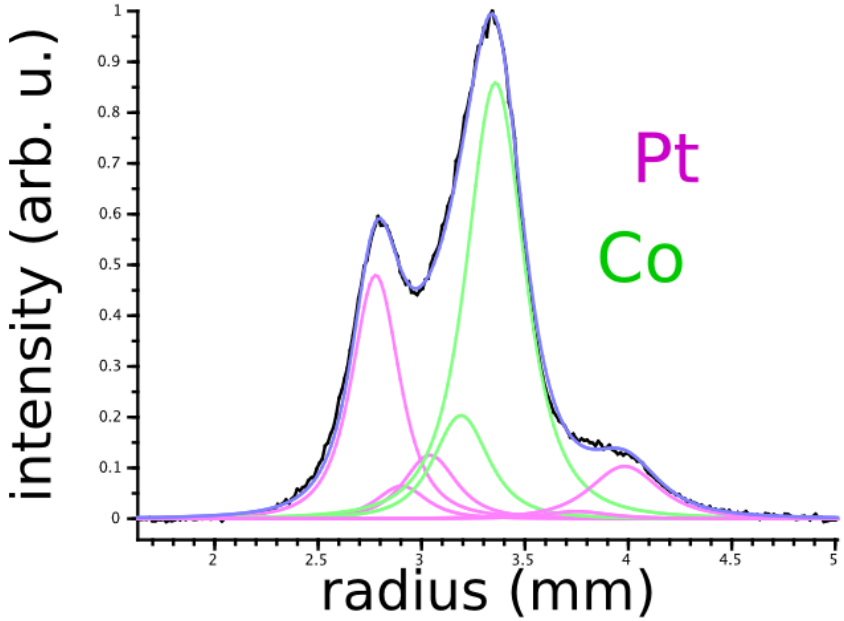


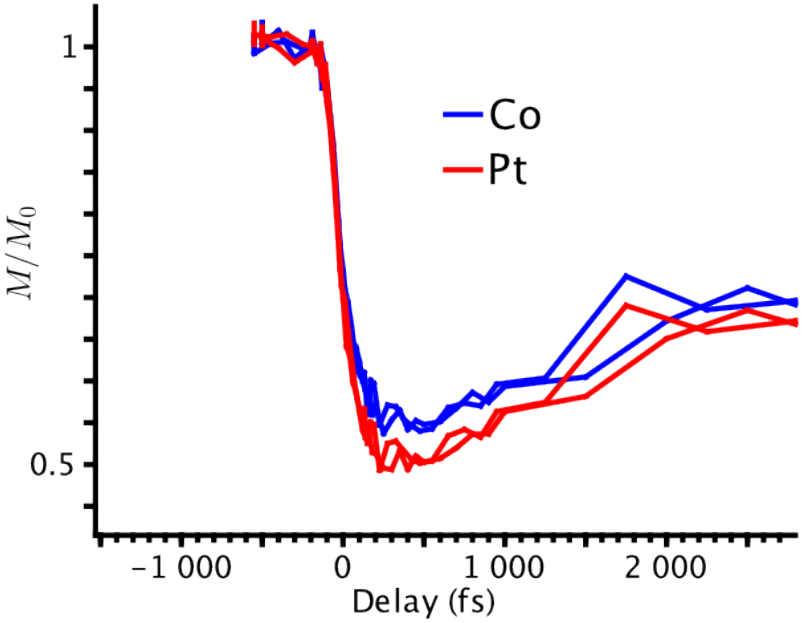




Looking at different elements in multilayers simultaneously

Probing perpendicular spin transfer



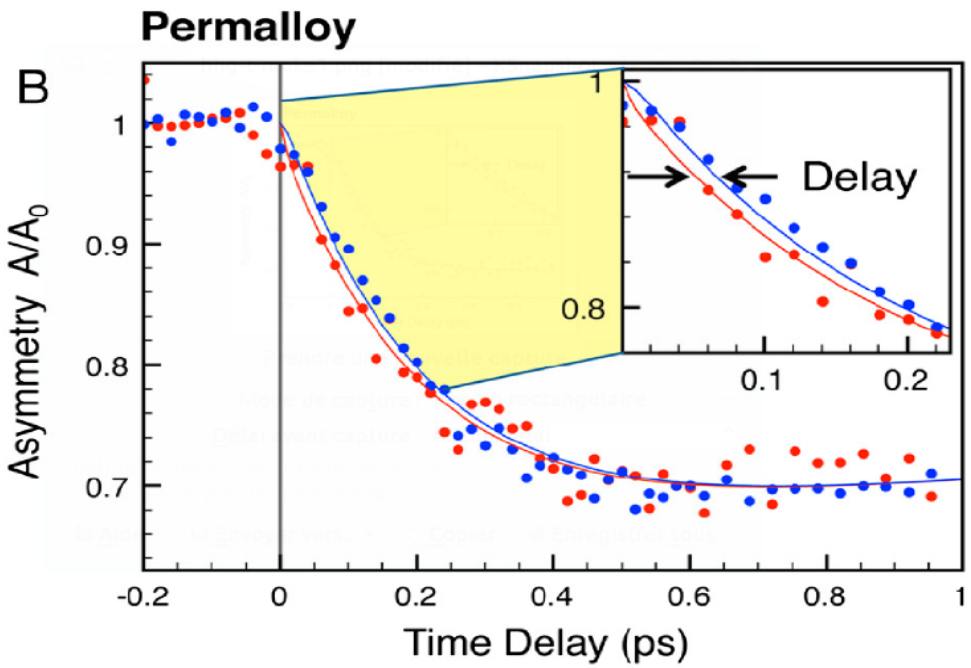


Looking at different elements in multilayers simultaneously

Conclusion

1. Superdiffusive spin transport contributes to ultrafast demagnetization
2. But it cannot explain all the observations

Outlook

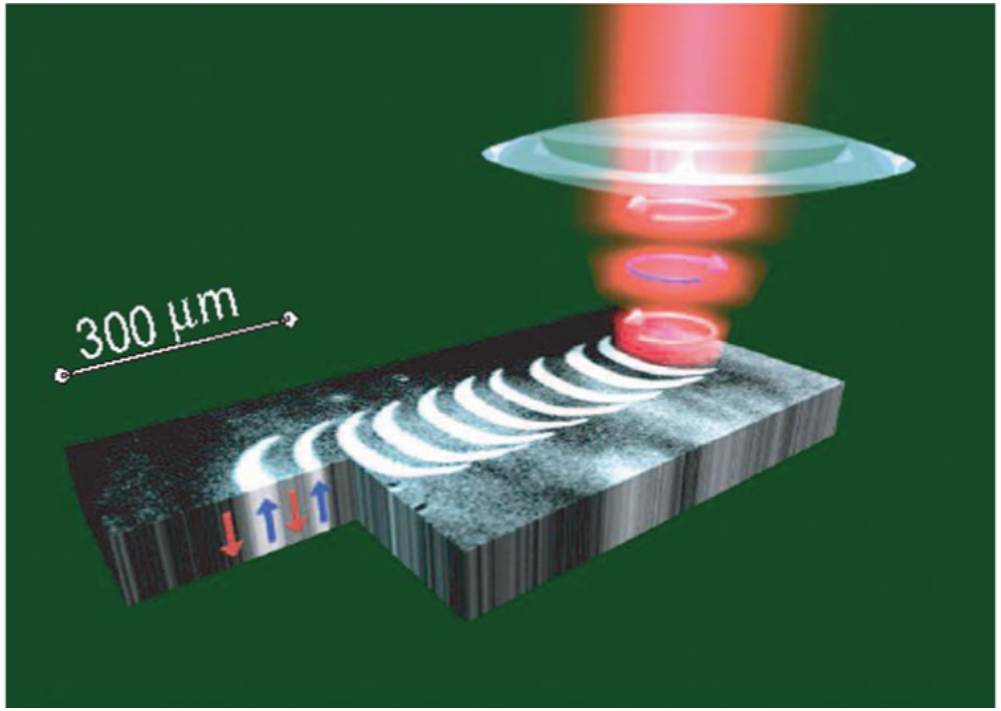


Requirements :

- Pushing the temporal resolution (shorter pulses)

Mathias et al., PNAS, 109, 4792 (2012)

Outlook

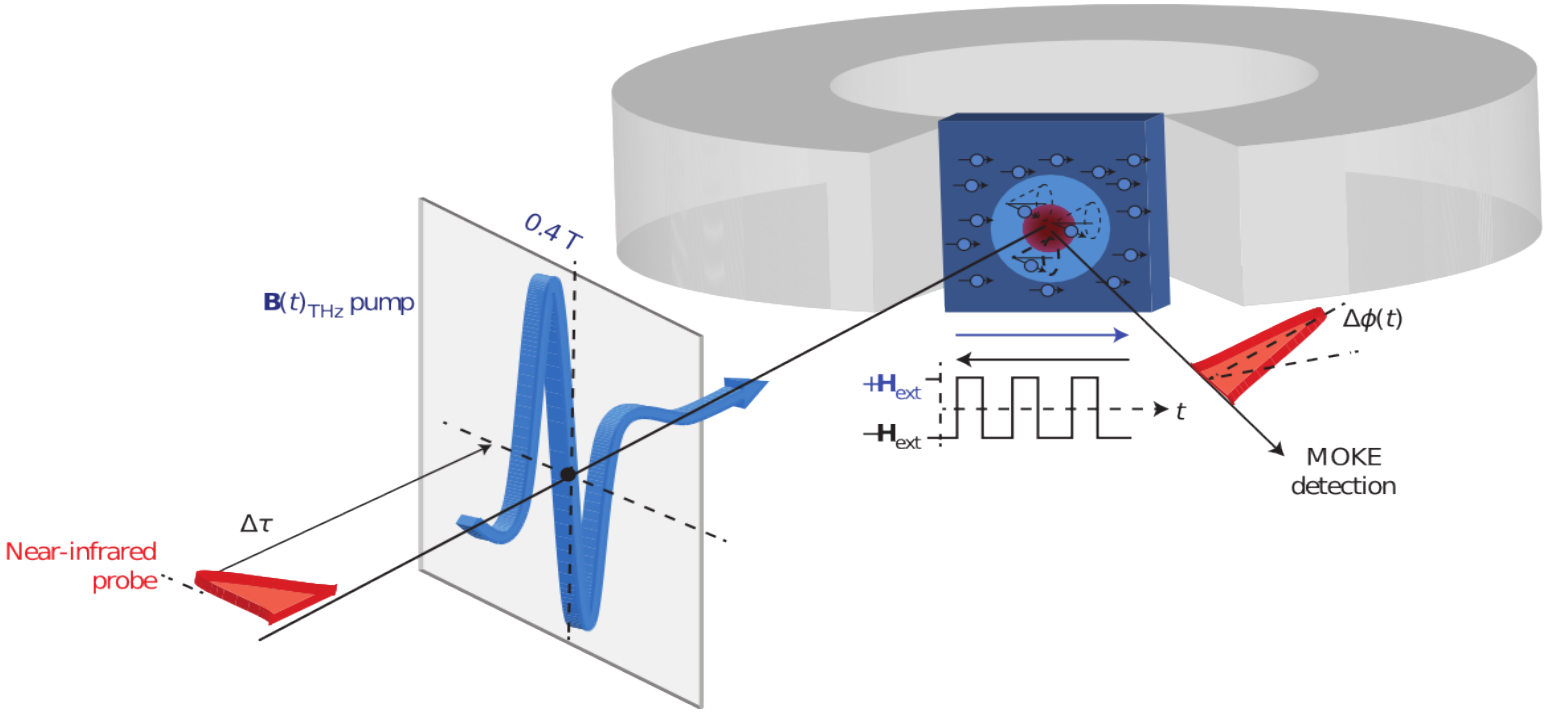


Requirements :

- Pushing the temporal resolution (shorter pulses)
- Higher energy harmonics $\sim$ 200 eV (rare earth absorption edges)

Stanciu et al., Phys. Rev. Lett., 98, 207401 (2007)

Outlook

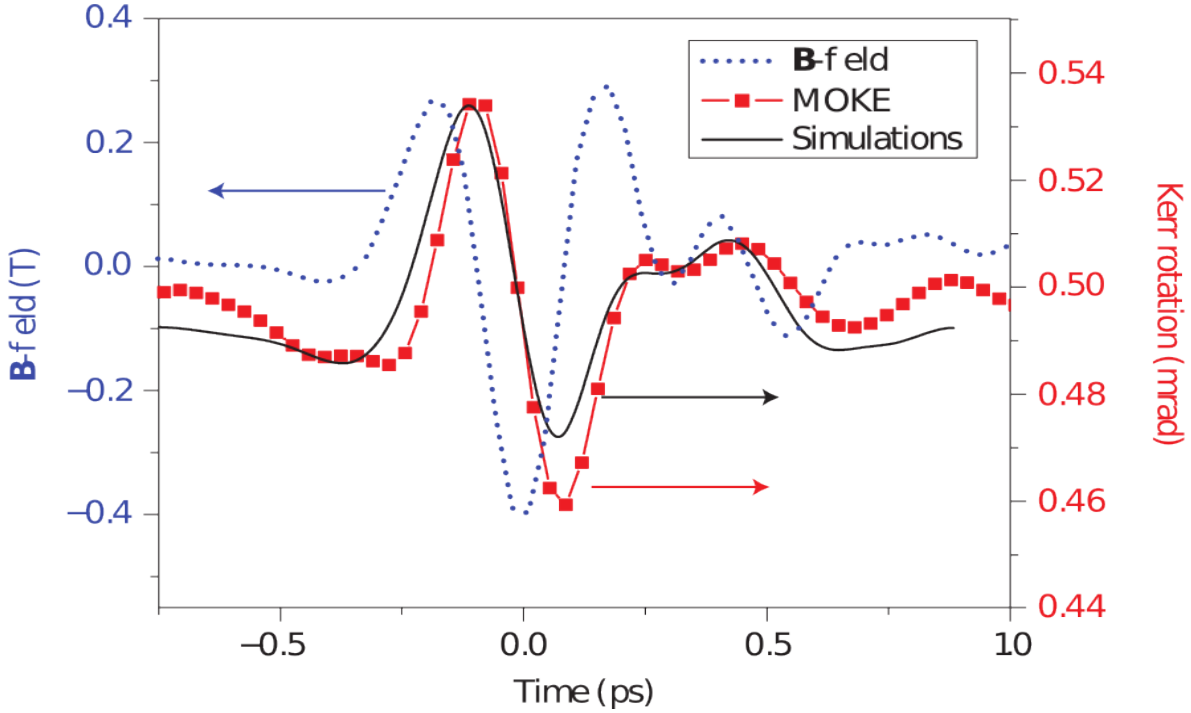


Requirements :

- Pushing the temporal resolution (shorter pulses)
- Higher energy harmonics $\sim$ 200 eV (rare earth absorption edges)
- High intensity THz source

Vicario et al., Nat. Photon., 7, 720 (2013)

Outlook



Requirements :

- Pushing the temporal resolution (shorter pulses)
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- High intensity THz source

Vicario et al., Nat. Photon., 7, 720 (2013)

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THz

- PSI, EPFL : C. Hauri ...