



Challenges and opportunities for pump-probe spectroscopy at synchrotron radiation sources

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SOLEIL- Synchrotron)



TREES, Trieste- December 3rd 2018

Summary

- Which pump which probe
- Examples
 - Magnetization dynamics
 - In real and reciprocal space
 - Electronic structure & phase transition
 - in FeRh and half metal manganite
- Discussion & Conclusion

Which Pump, which probe

Exciting Pump:

- Magnetic pulse
- **Electric/current pulse**
- **Laser pulse**

Measuring probe:

- **Synchrotron Pulse**
- Laser Pulse
- HHG
- FEL

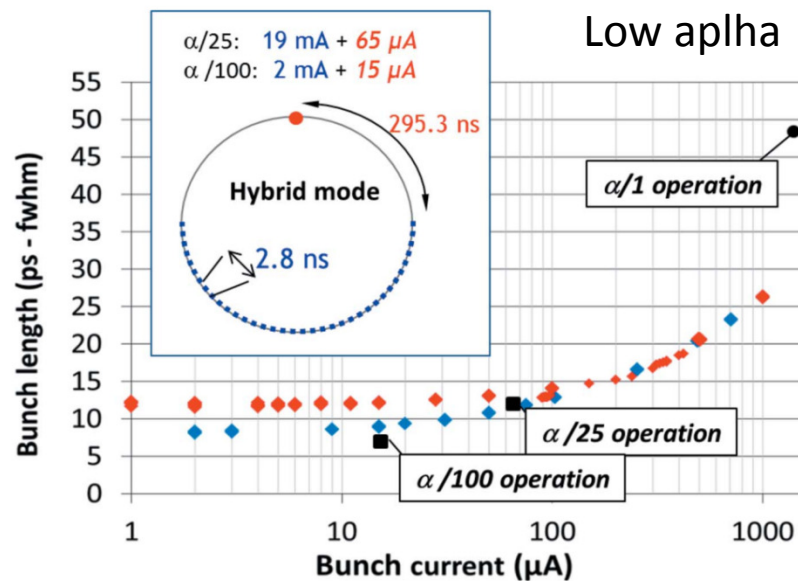
Repetition rate
Energy/pulse



Repetition rate
Photons/pulse

Excitation is essential to define the experiment
Changing excitation is like changing the sample

Short pulses: slicing \leftrightarrow low alpha



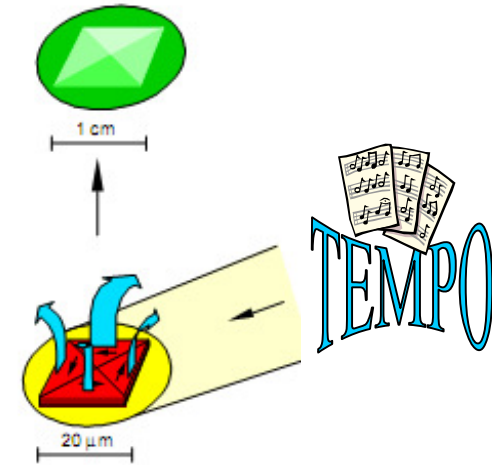
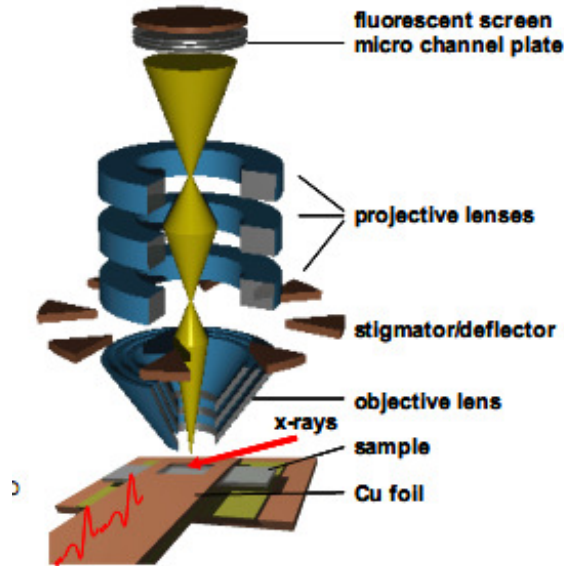
Slicing photon flux ?
 $10^6 - 10^7$?

J. Synchrotron Rad. (2017). 24, 886–897

- Total flux
- Repetition rate

Magnetization Dynamics

J. Vogel Institut Néel
V. Uhler
S. Pizzini
N. Rougemaille
L. Ranno
O. Fruchart
V. Cros
E. Jimenez
J. Camarero
C. Tieg



CNRS/Thales

Universidad Autonoma de Madrid,
ESRF

Phys. Rev. B, 2010, 81(22),224418

Phys. Rev. B, 2011, 83(2),020406

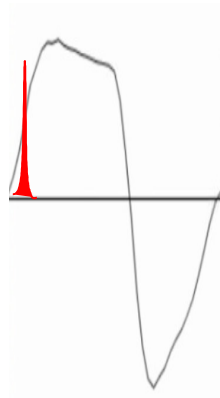
Phys. Rev. Lett., 2012, 108(24),247202

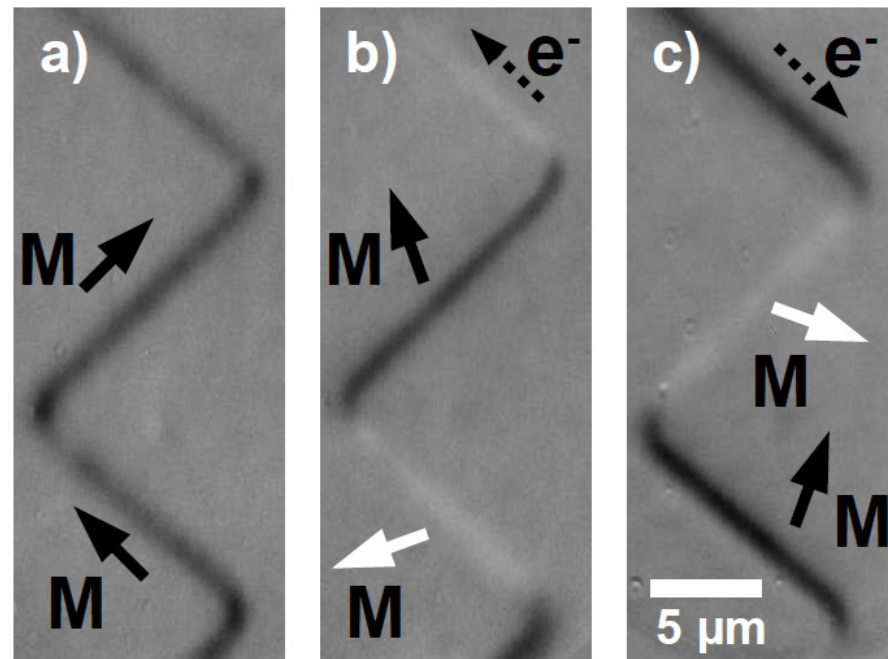
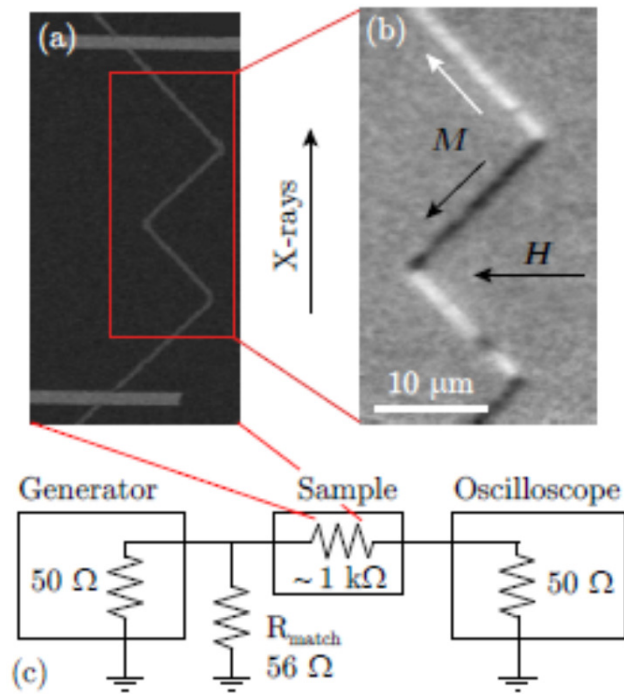


Synchrotron radiation Time structure

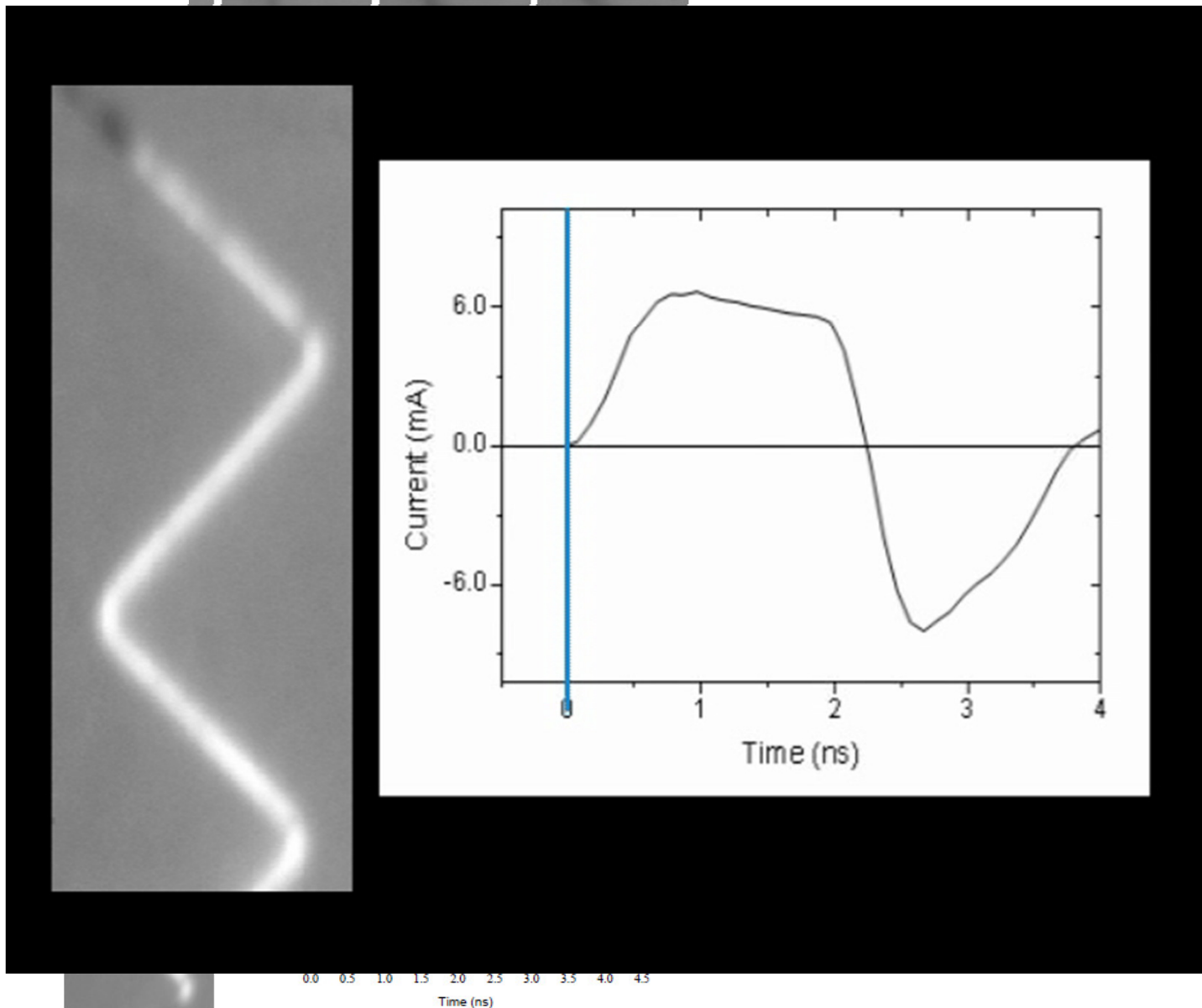


Synchrotron pulse duration: 50 ps
High repetition rate: 6 MHz ,

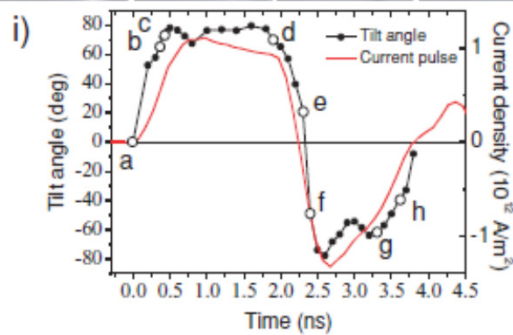
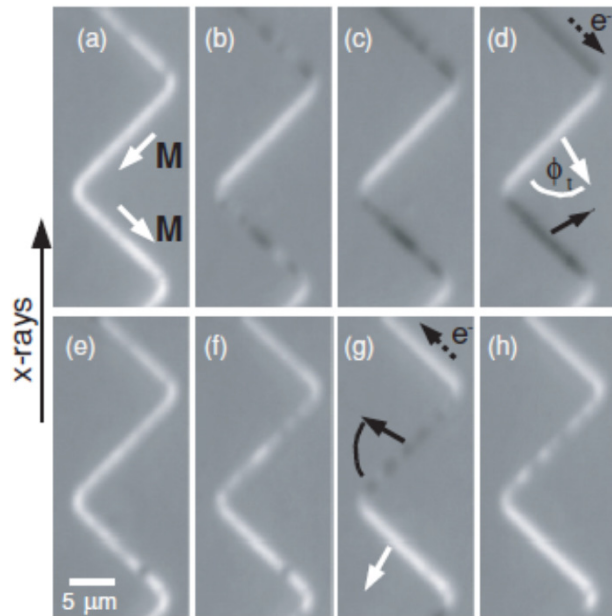




Magnetization Dynamics



Electron microscopy with polarized X-rays



Study of laser excitations in nanostructures:
Electron transport, heat transport

Imaging secondary electrons
XMCD contrast – element specific

Could be done in low alpha mode,
Slicing only with high repetition rate

Samples are the challenge

Magnetization Dynamics



ps Time Resolved Resonant Magnetic Scattering Using SOLEIL's Low-Alpha Mode

Synchrotron SOLEIL (SEXTANTS)

Horia Popescu, Victor Lopez-Flores, Maurizio Sacchi, Nicolas Jaouen



Synchrotron SOLEIL (TEMPO)

Mathieu Silly, Christian Chauvet, Fausto Sirotti, Philippe Hollander



LCPMR, Paris VI

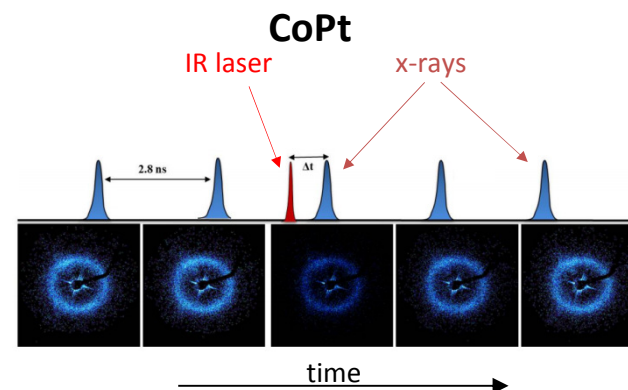
B. Tudu, R. Delaunay, J. Lüning



Synchrotron SOLEIL (Sources)

Marie-Agnès Tordeux, Amor Nadji

ELETTRA (Detectors and Instrumentation
Luigi Stebel, Rudi Sergo, Giuseppe Causero



IPCMS, Strasbourg

Christine Boeglin

LPS, Orsay

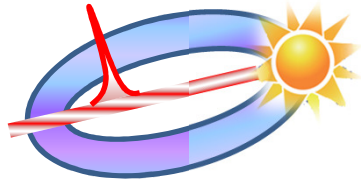
Gregory Malinowski

SPINTEC, Grenoble

Marina Tortarolo

CSNSM, Orsay

Cédric Baumier, F. Fortuna



FemtoSlicing

Specificities TEMPO

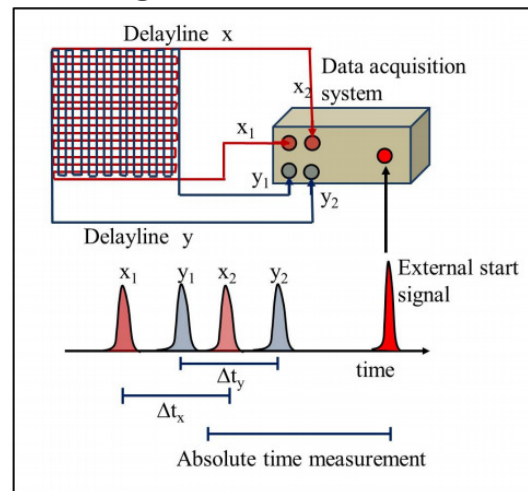


2D time resolved detector (Elettra)

MCP



Single count detector :



4 Mcounts/s

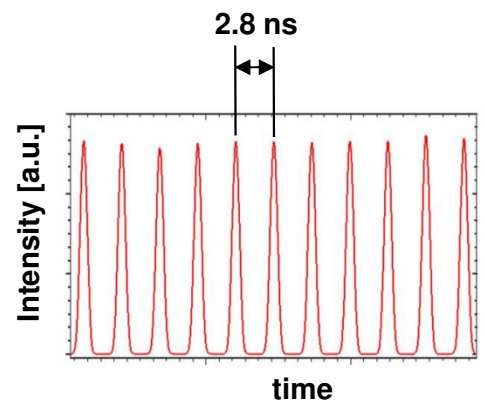
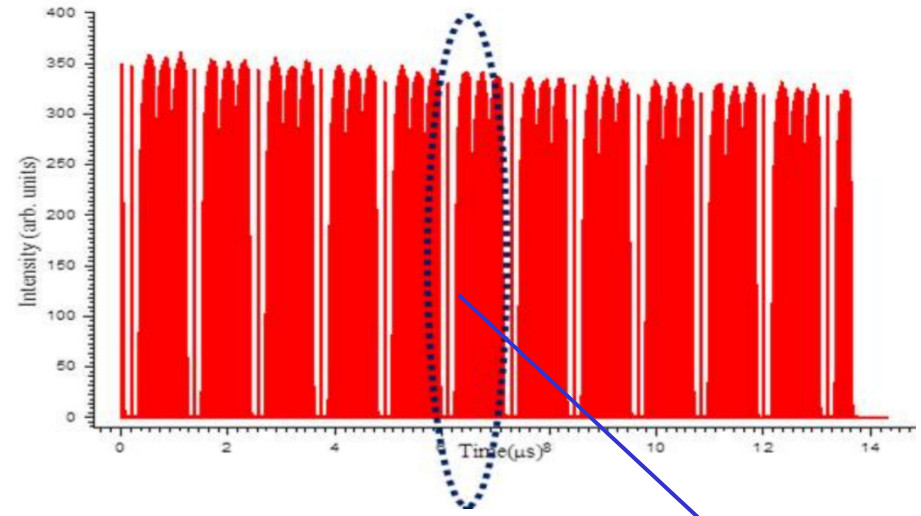
27 ps temporal resolution

60 μm spatial resolution

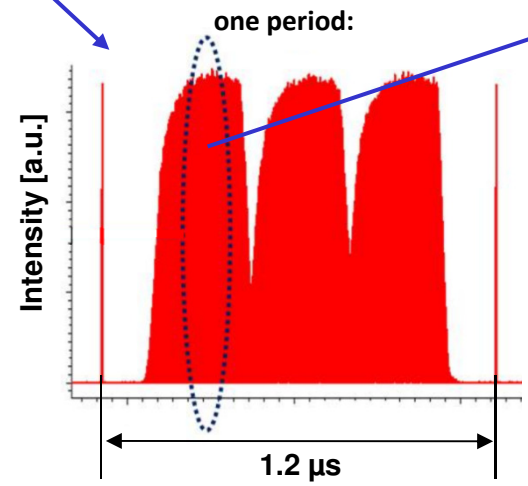


Low-Alpha time histogram as seen by the detector

70 kHz: 12 synchrotron periods

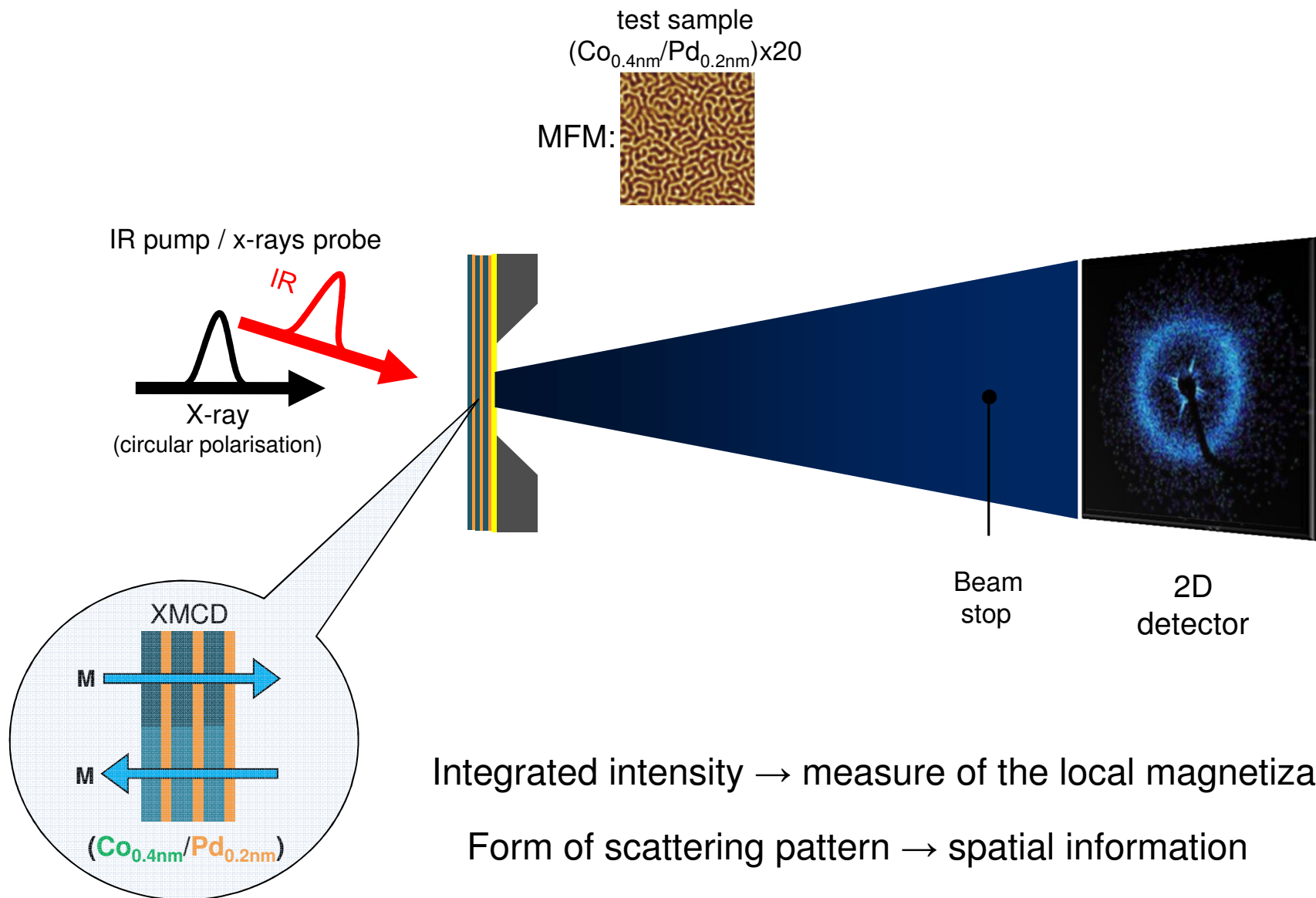


70 kHz : the laser frequency was decreased in order to avoid the sample overheating

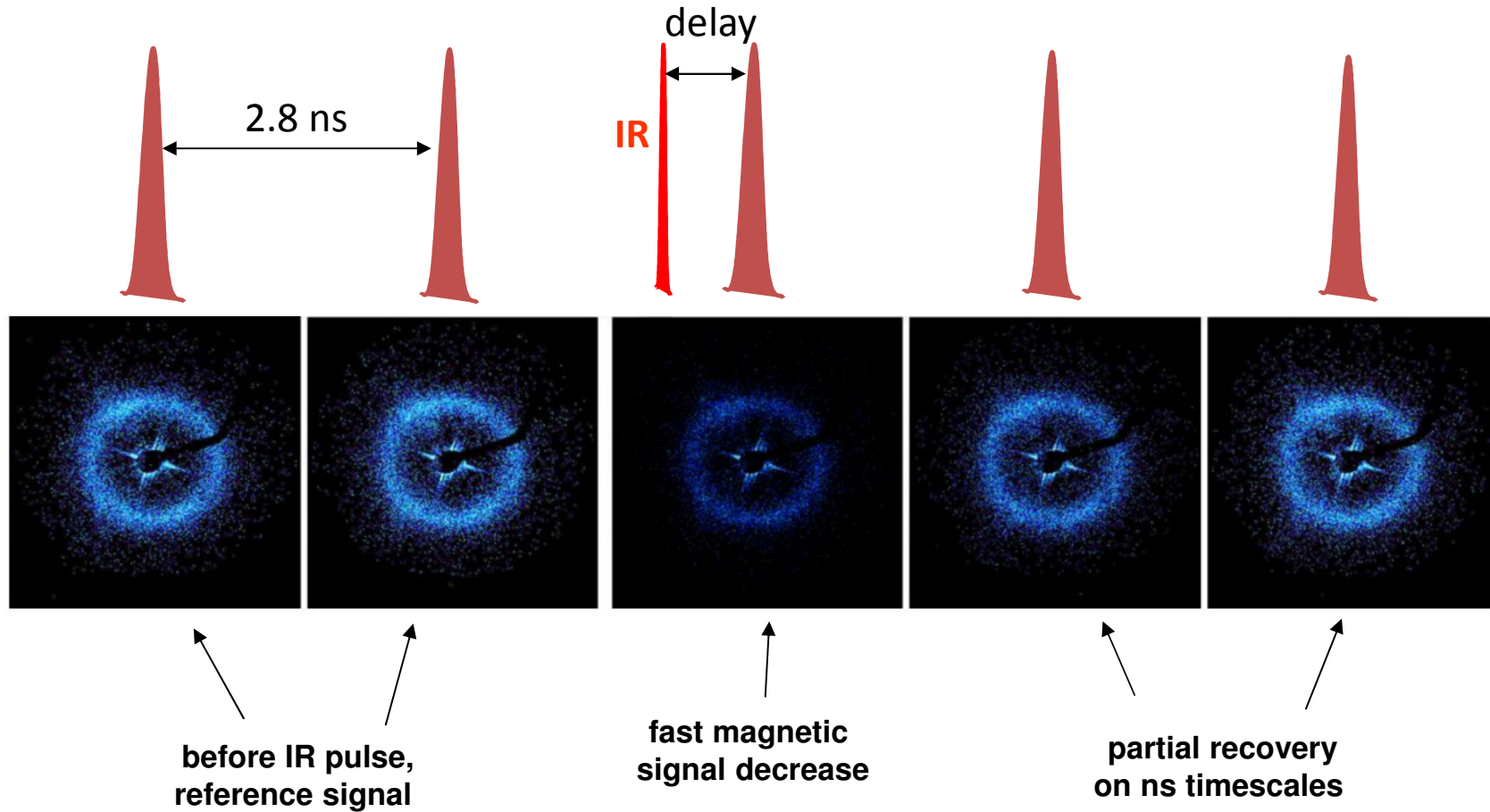


312 bunches
65 μA / bunch

Magnetization dynamics using small angle x-rays scattering (SAXS)



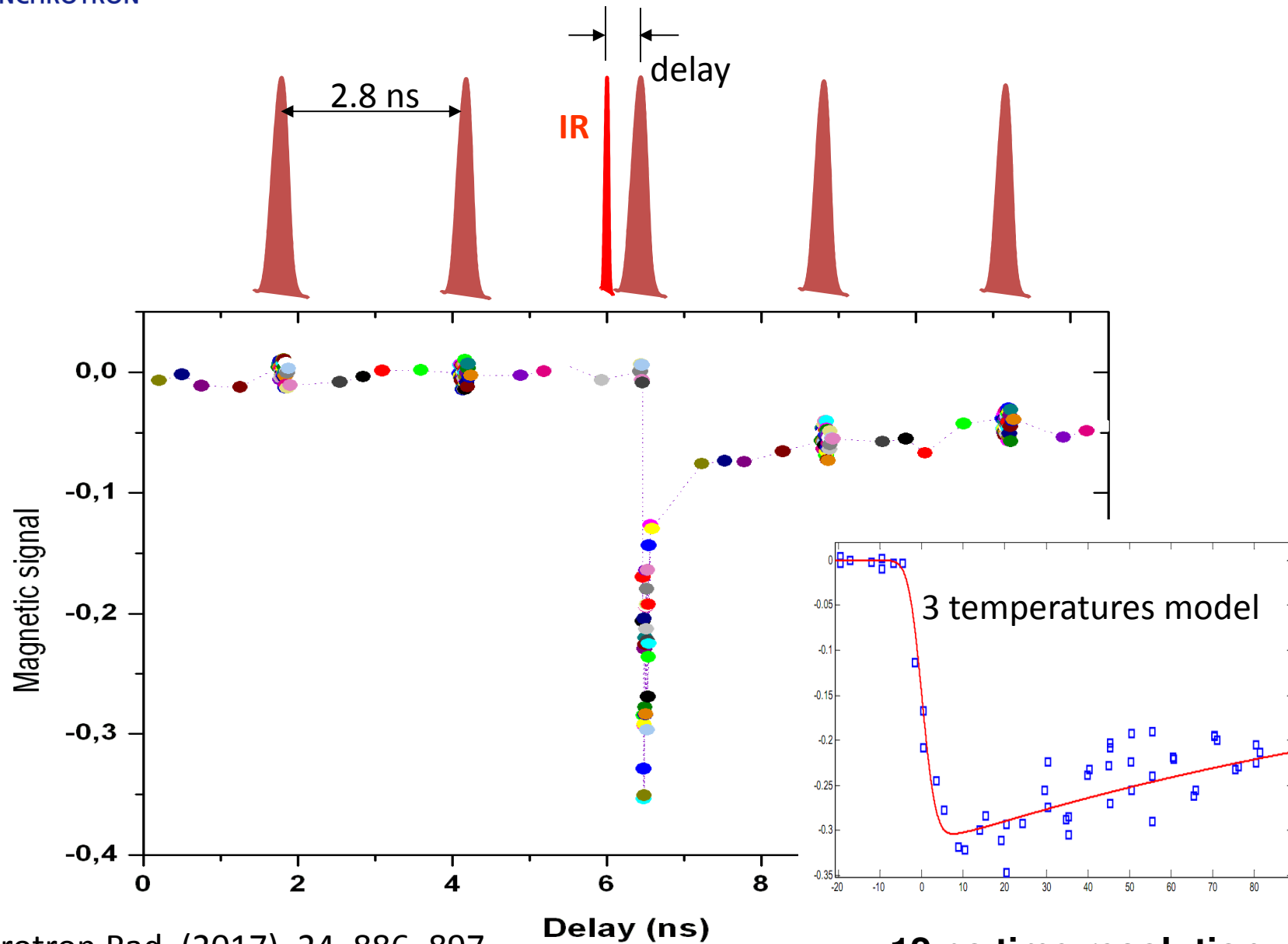
Resonant (Co L₃) magnetic small angle scattering pattern



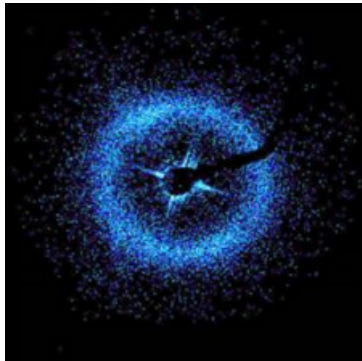
Laser fluence: 15 mJ/cm²

Integration time: 3 min at 70 kHz

Low-Alpha multi-bunch: fast (ps) and slow (ns) dynamics



Time resolved resonant magnetic scattering



Study of magnetization dynamics in nanostructures:

Coherent imaging

XMCD contrast – element specific

Low alpha mode, Slicing

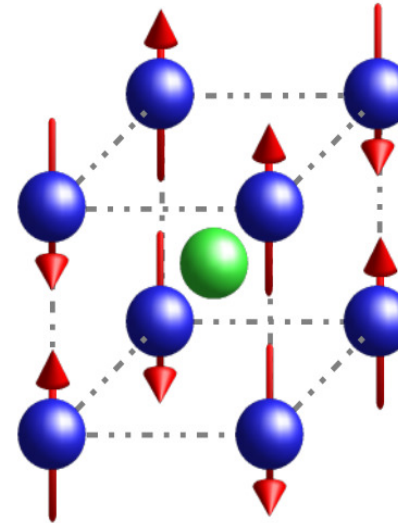
FEL - collaborations

The phase transition in FeRh

In FeRh the transition involves both the **Magnetic Order** and the **Lattice Structure**;

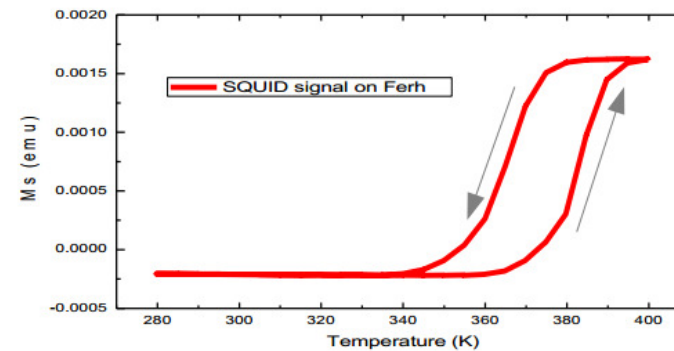
Below T_c
 $Fe = \pm 3.3 \mu_B$
 $Rh = 0 \mu_B$

Above T_c
 $Fe = 3.1 \mu_B$
 $Rh = 0.9 \mu_B$



The volume is expanded of about 1% bulk samples or in thick films;

- Isotropically in bulk or in thick films;
- Along the out of plane direction for **thin films**



$$T_{FM \rightarrow AFM} = 385 \text{ K} \quad T_{AFM \rightarrow FM} = 395 \text{ K}$$

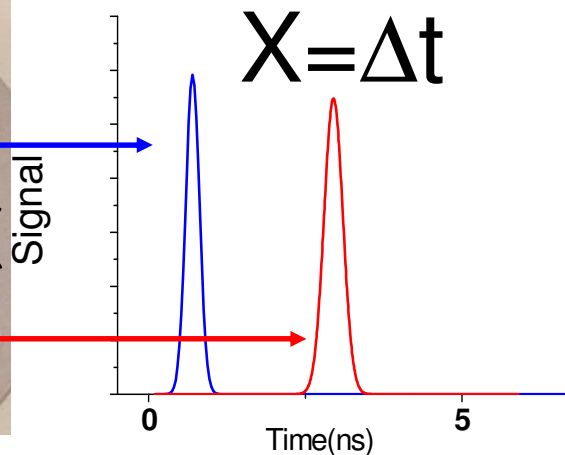
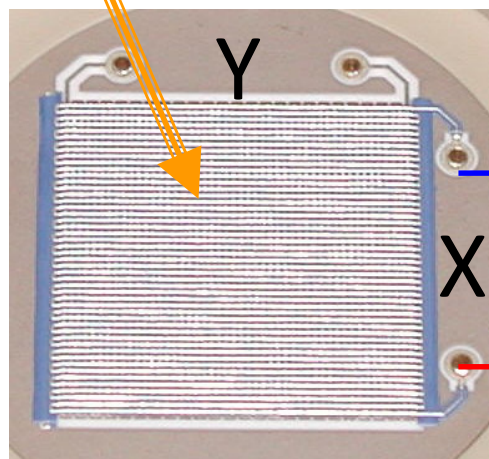
2D detector

Elettra

L. Stebel
R. Sergio
P. Pittana
G. Cauero

Electrons cascade from Micro channel plate

4 Mcounts/s
27 ps temporal resolution
60 μm spatial resolution



research papers

Journal of
Synchrotron
Radiation

ISSN 0909-0495

Received 2 March 2010
Accepted 13 December 2010

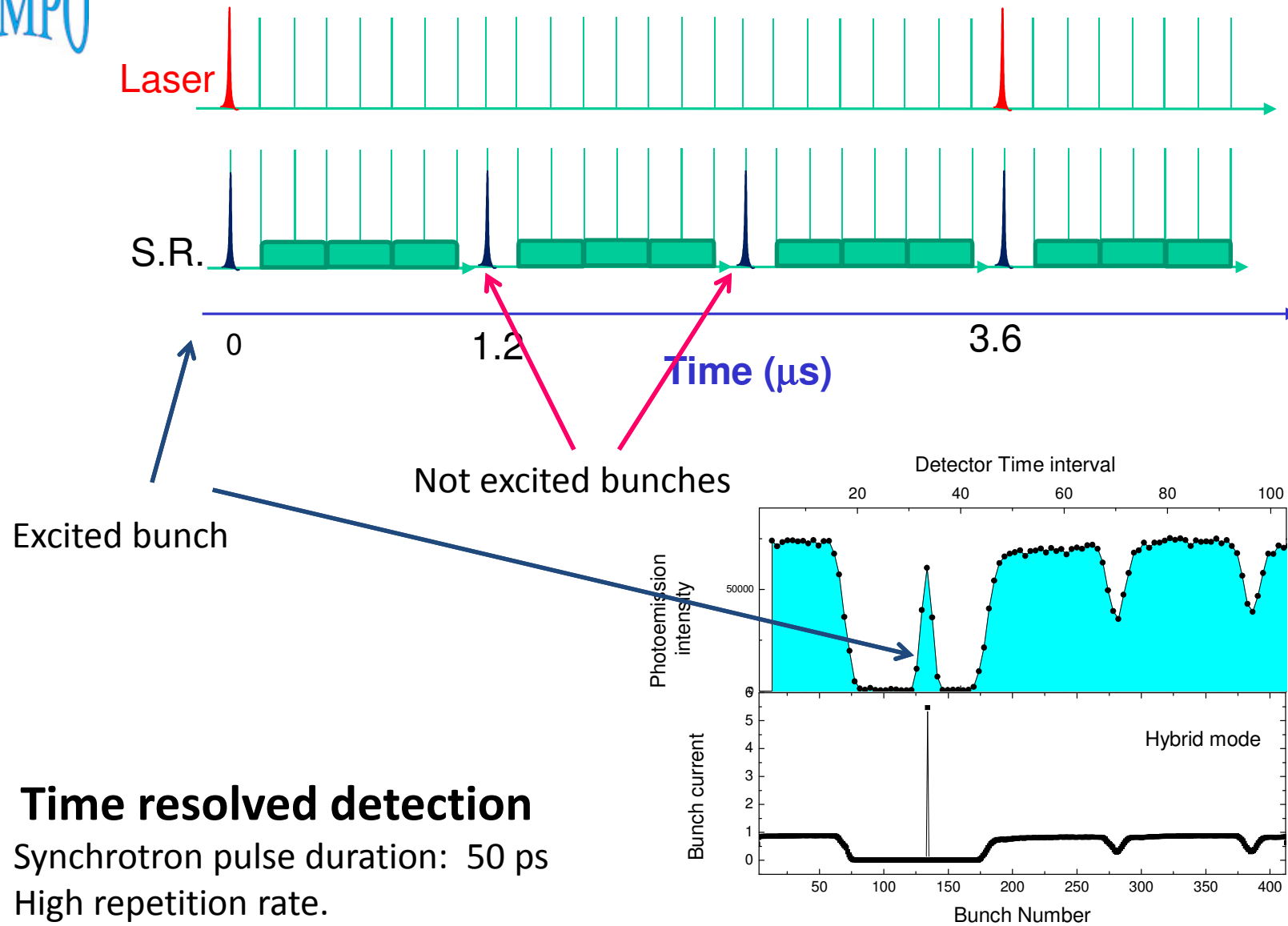
**Time-resolved photoelectron spectroscopy using
synchrotron radiation time structure**

N. Bergéard,^a M. G. Silly,^a D. Krizmancic,^b C. Chauvet,^a M. Guzzo,^{a,c} J. P. Ricaud,^a
M. Izquierdo,^a L. Stebel,^d P. Pittana,^d R. Sergio,^d G. Cauero,^d G. Dufour,^e
F. Rochet^e and F. Sirotti^{a*}

Journal of Synchrotron Radiation, 2011, 18(2): 245-250



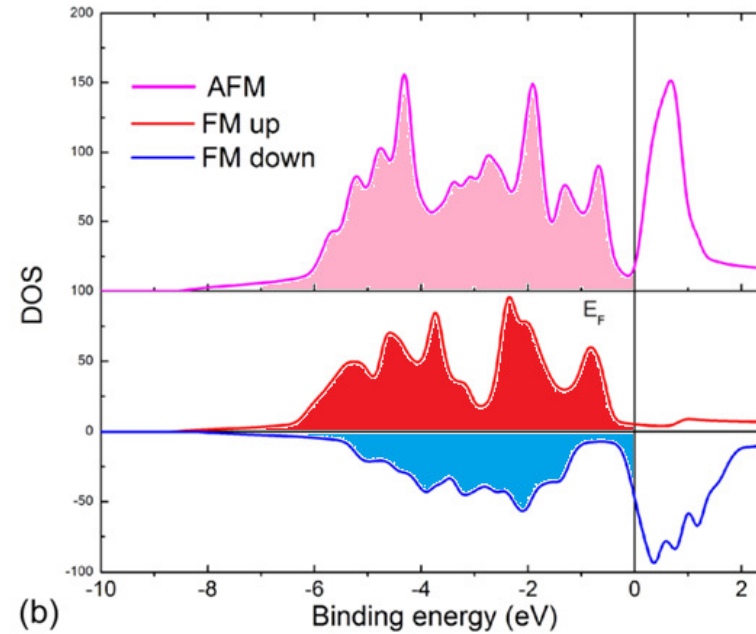
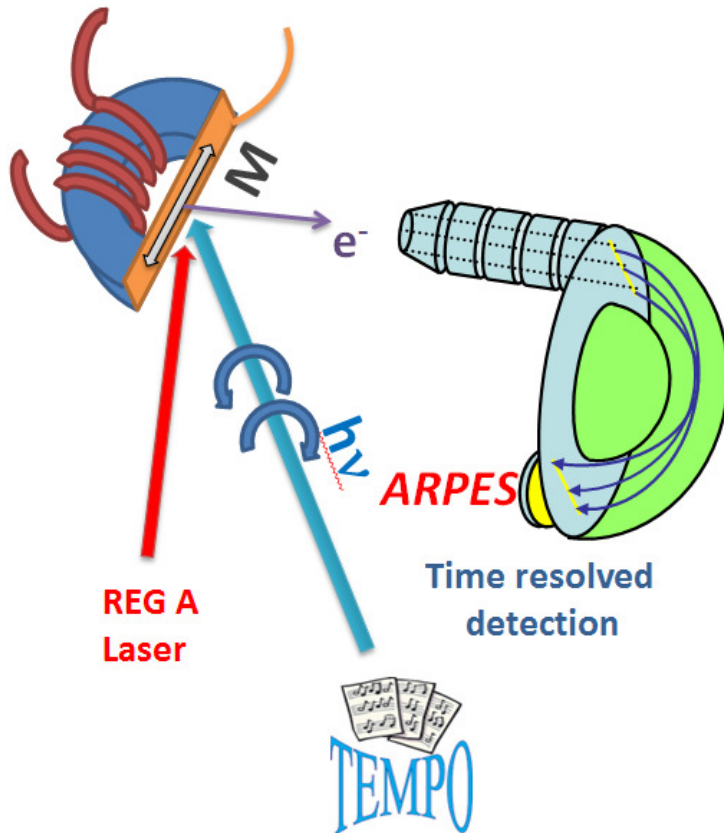
Photoemission experiments





FeRh: magnetism and electronic structure

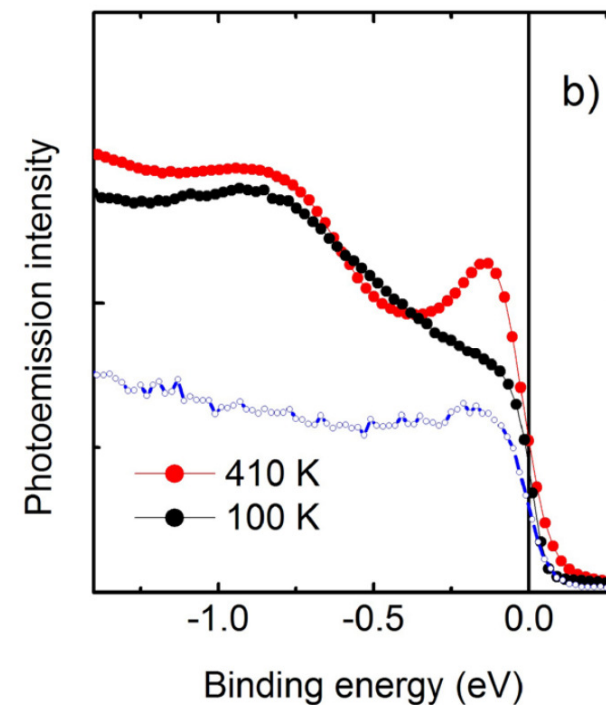
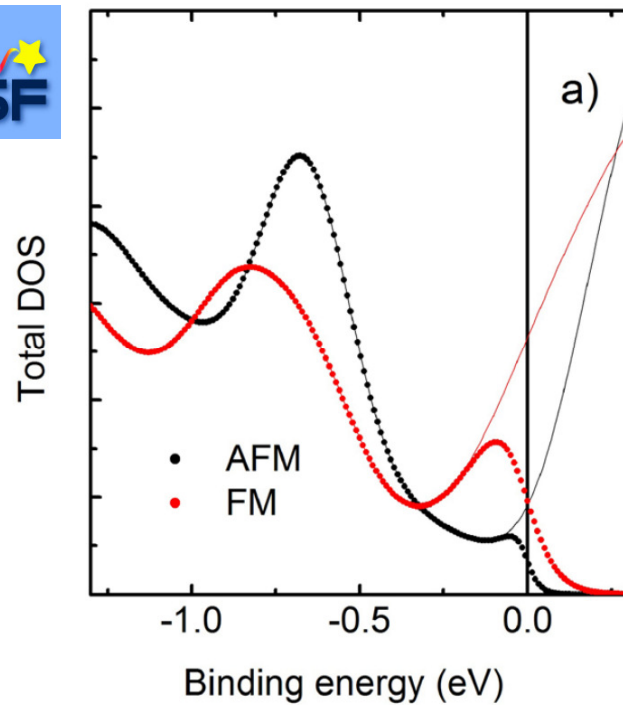
Variable T



Theory:
M. Gatti, LSI & ETSF Palaiseau

Modification of the electronic structure

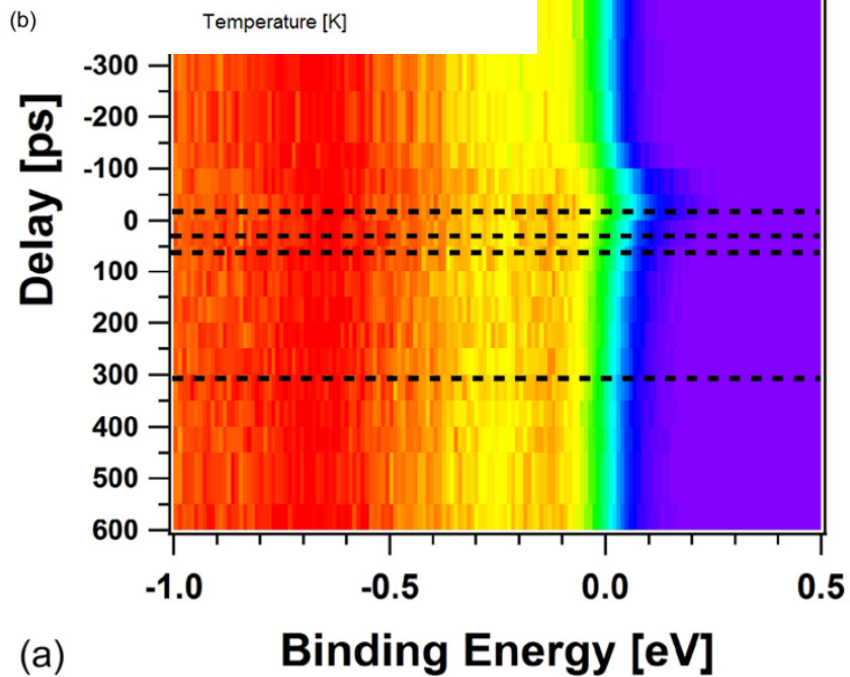
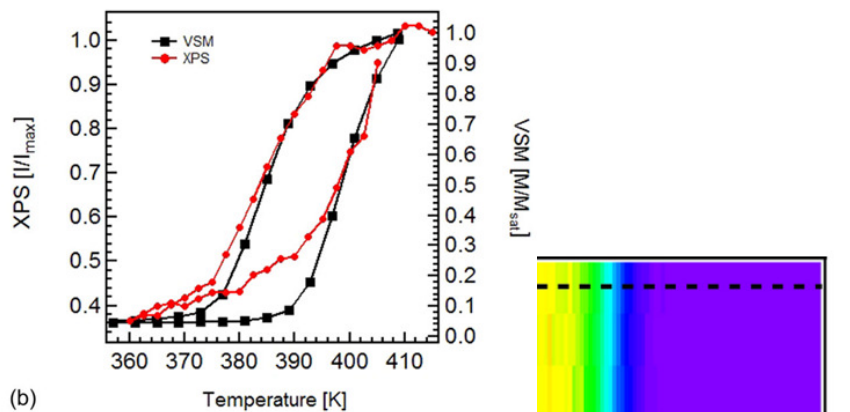
Modification of the electronic structure



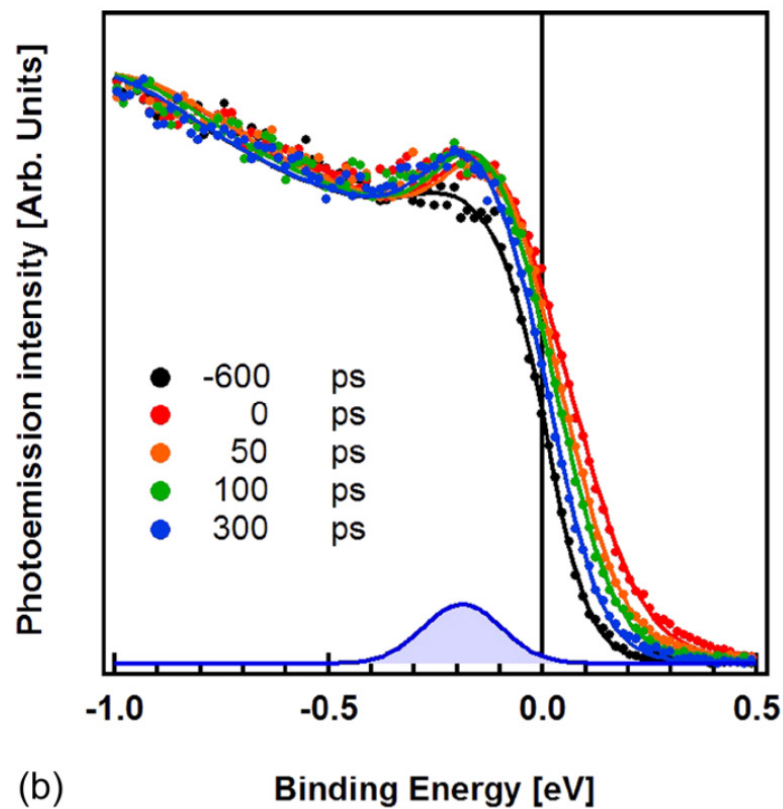
Shape of band structure well characterize the magnetic phase



FeRh: magnetism and electronic structure



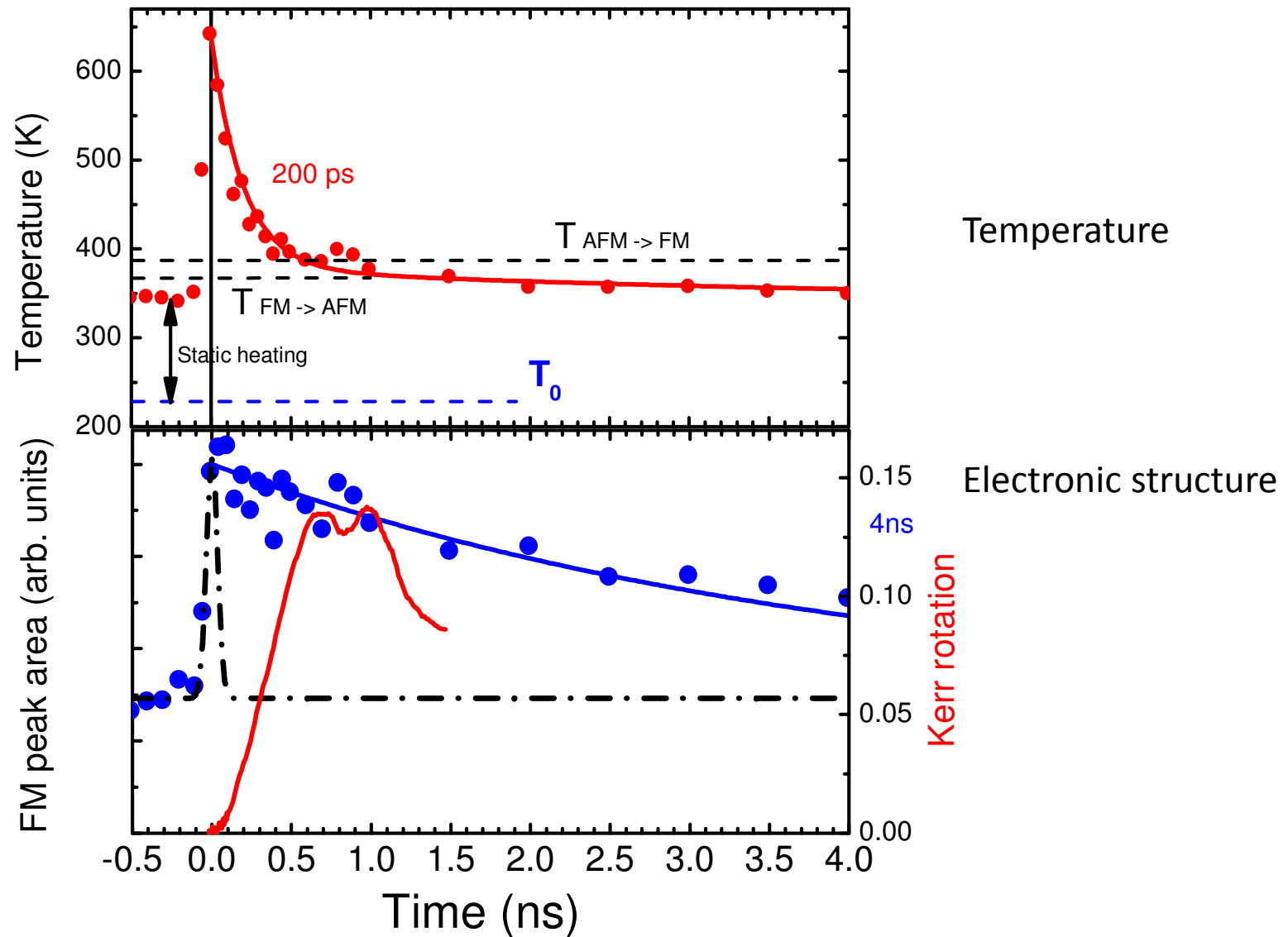
Laser excited
Phase transition



F. Pressacco et al. Struct. Dyn.5, 034501 (2018)

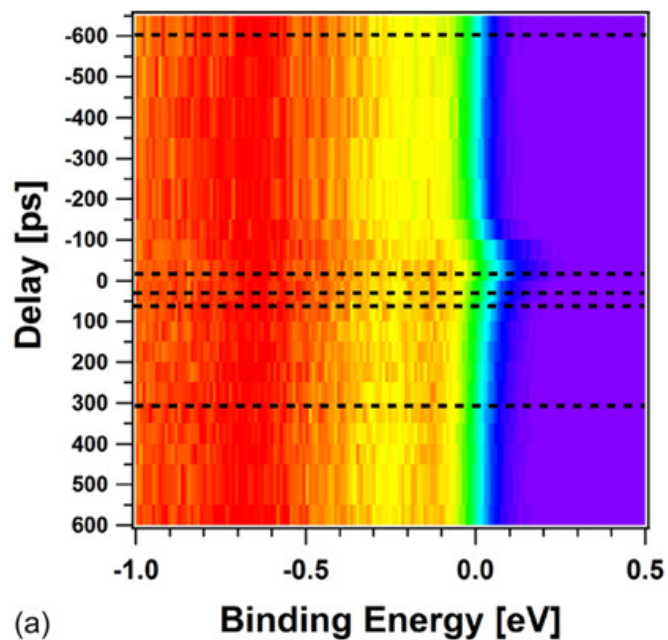


FeRh: magnetism and electronic structure





Valence Band photoemission



Electronic & magnetic properties

Study of recovery process.

Preparation of tools for FEL & HHG

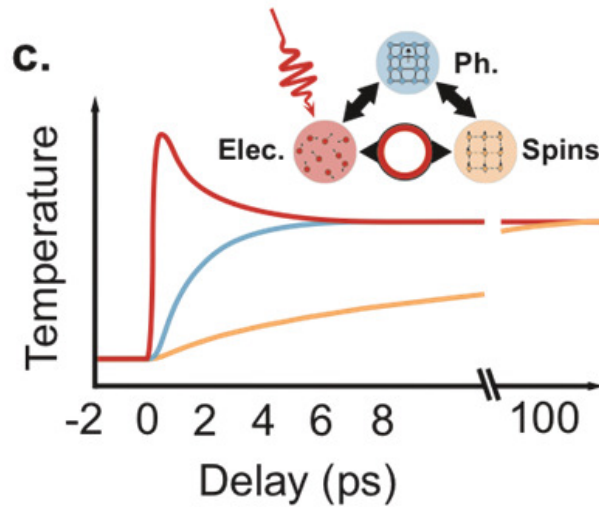
Studies motivated by pump/probe

50 ps & Low alpha mode

Synchrotron operation needed

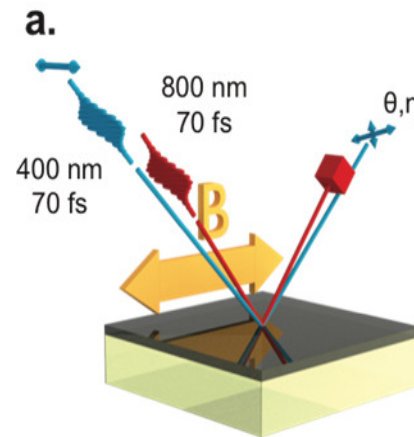
FEL & HHG

Magnetization dynamics of half-metallic manganite

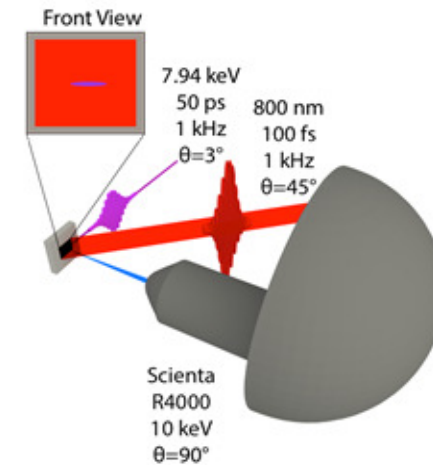


T. Pincelli, R. Cucini, A. Verna, F. Borgatti, M. Oura, K. Tamasaku, H. Osawa, T.L. Lee, C. Schlueter, S. Gunther, C.H. Back, M. Dell'Angela, P. Orgiani, A. Petrov, F. Sirotti, R. Ciprian, V. A. Dediu, I. Bergenti, P. Graziosi, F. Miletto Granozio, Y. Tanaka, M. Taguchi, H. Daimon, J. Fujii, G. Rossi, and G. Panaccione

In a half-metallic system, the direct spin-electron de-excitation channel is almost suppressed due to the absence of available spin-flip processes.



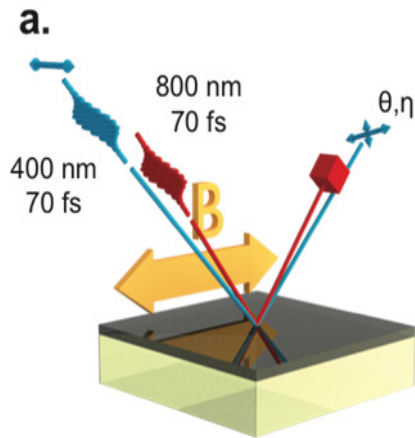
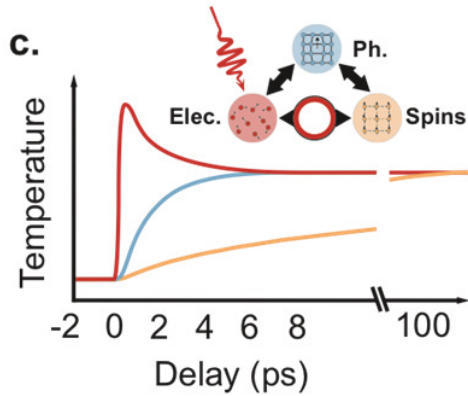
Pump-probe MOKE setup.



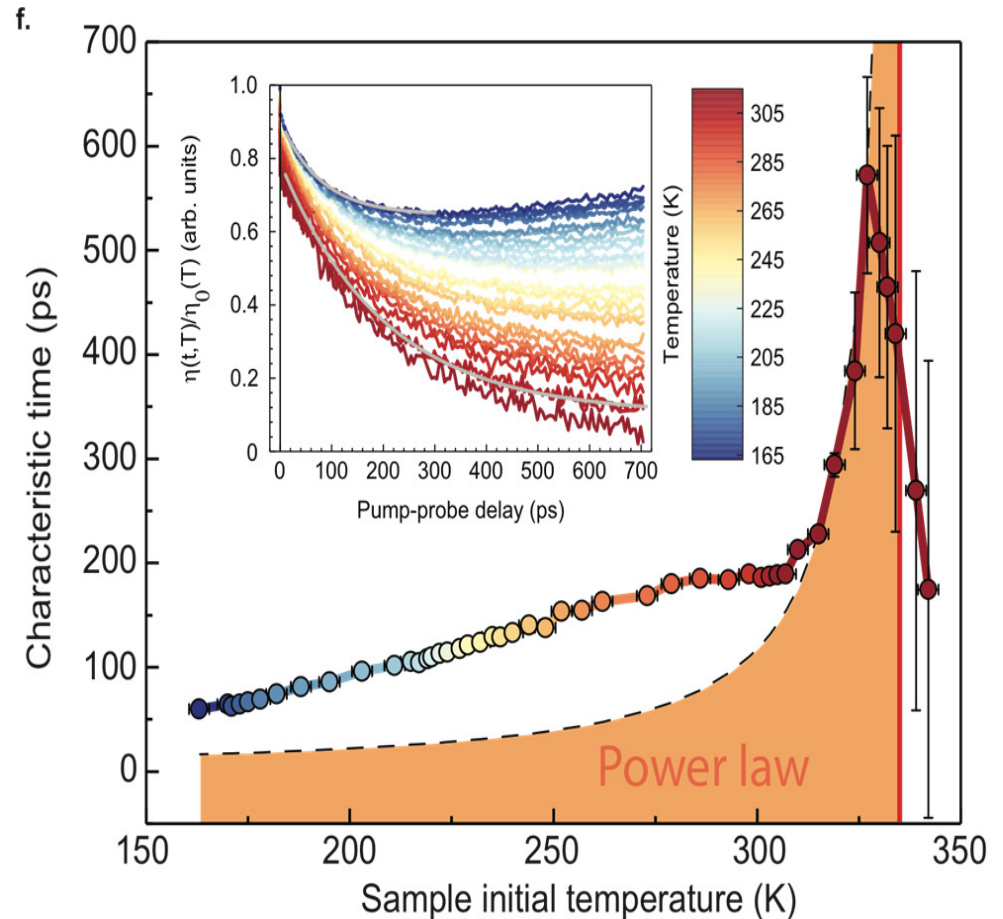
Pump-probe **HAXPES** setup

Magnetization dynamics of half-metallic manganite

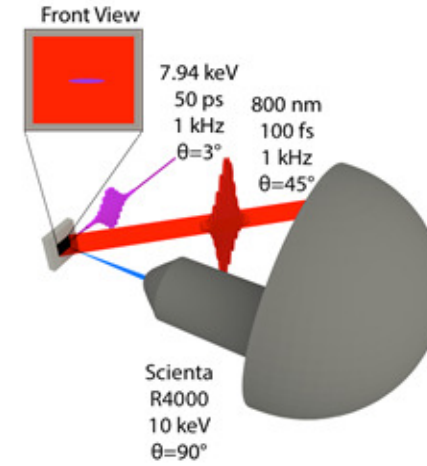
In a half-metallic system, the direct spin-electron de-excitation channel is almost suppressed due to the absence of available spin-flip processes.



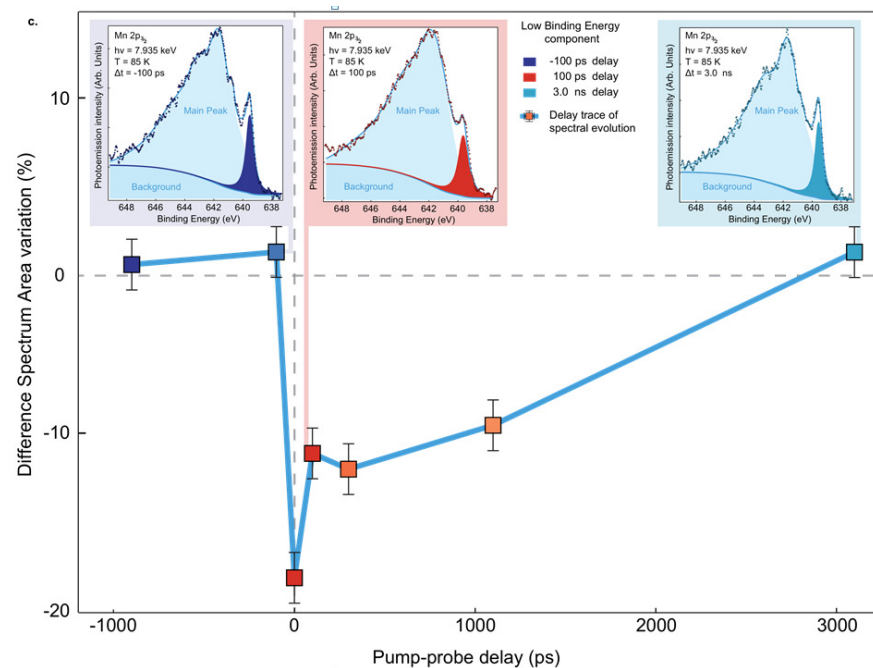
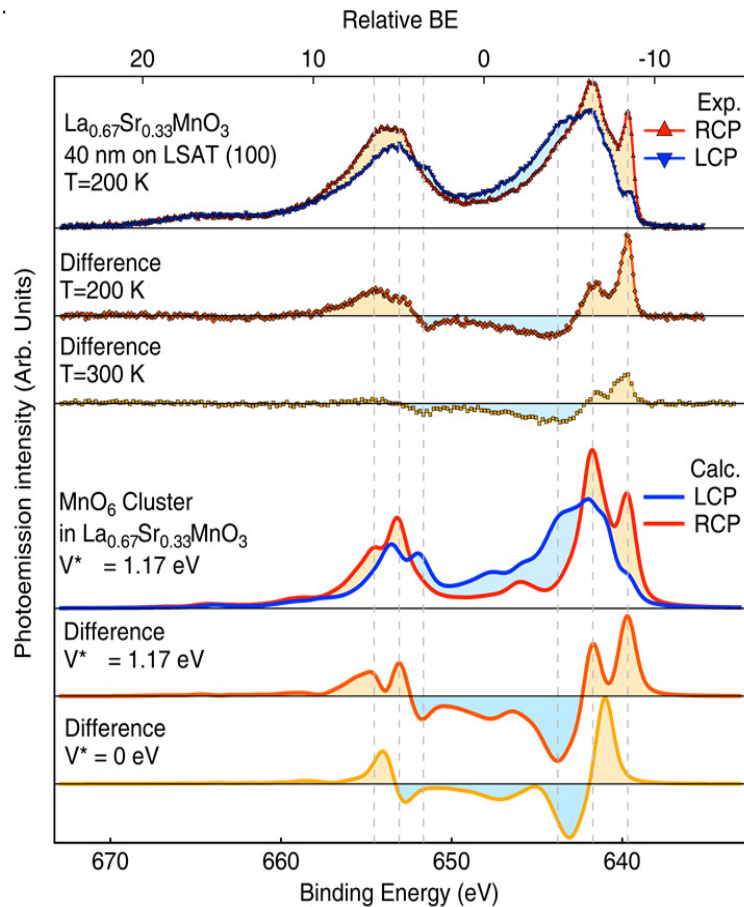
Pump-probe MOKE setup.



Magnetization dynamics of half-metallic manganite

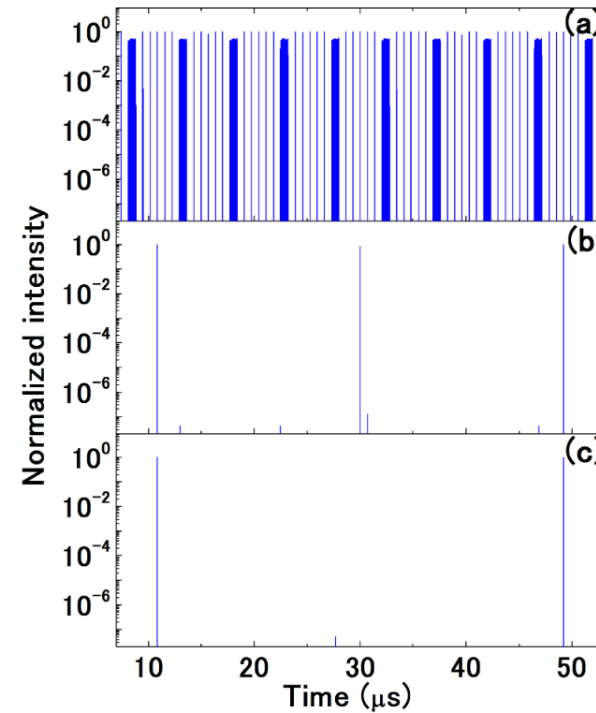
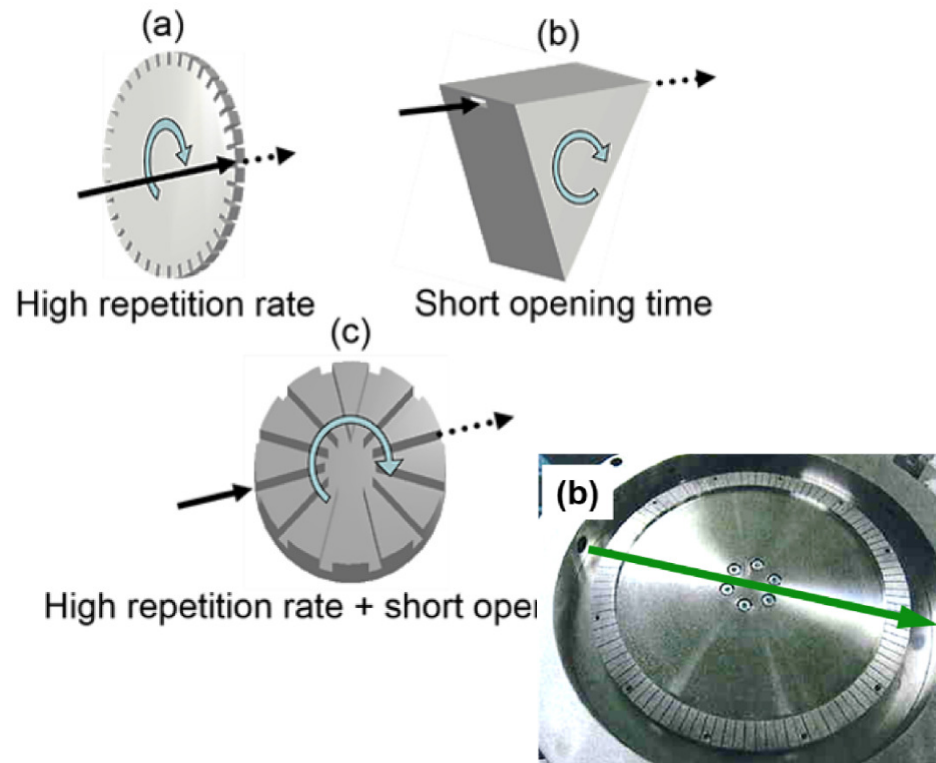


Pump-probe HAXPES setup
1 KHz – X-ray Chopper





Pump probe with hard X-rays



HAXPES Band photoemission

Electronic & magnetic properties

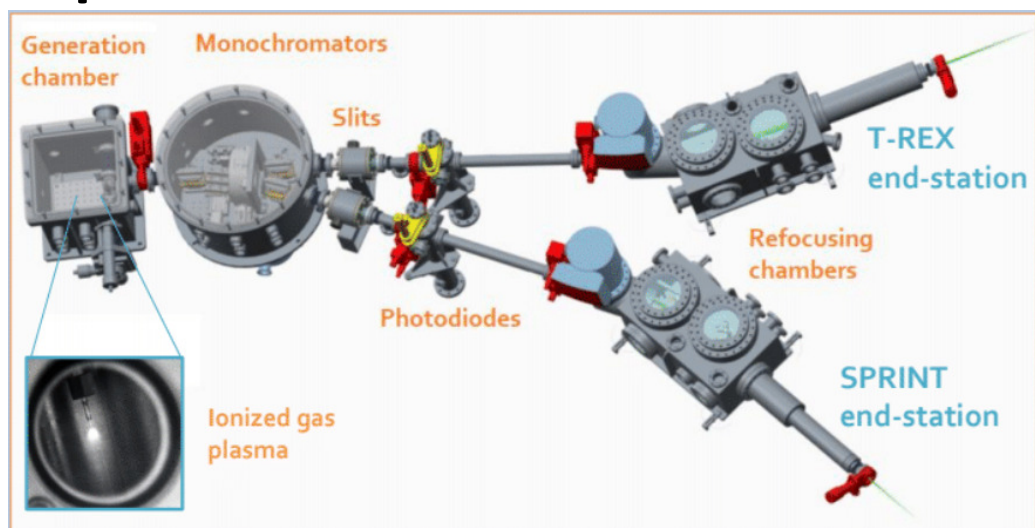
Studies motivated by pump/probe experiments

50 ps & Low alpha mode

Synchrotron operation

FEL & HHG

Sprint



Conclusions

Physical processes are characterized by **pump** characteristics (pulse duration, energy, polarization)

The pump laser is important in defining the initial state

- System dynamics can then be followed on different time scales

Synchrotron are complementary to FEL, HHG and laser

- Important to make the link between time scales and overlap time intervals of excitation and decay

- better sample characterization on synchrotrons

Strong and regular interaction with fs sources

Physics should drive collaborations to apply the most adapted source

Conclusions

Operation:

Ideally pump probe and normal synchrotron activity should be possible on demand -> Hybrid mode operation of storage rings.

Time resolved detection x-ray choppers
to switch between normal and time resolved mode


Short (few days) and rare (every six months) beamtime period allocated for special operating modes are not effective for scientific activity development

Conclusion

- TR-X-PEEM - Electron microscopy with polarized X-rays
- Time resolved resonant magnetic scattering
- ARPES and HAXPES can be used to study laser excited phase transitions in pump/probe experiments
- Electronic and magnetic properties measured at the same time
- Theoretical predictions are necessary to describe details of non equilibrium spectroscopy

Physics should drive collaborations to apply the most adapted source

Strong and regular interaction with fs sources

A photograph of a modern building with a curved wooden lattice structure in the foreground and a rainbow in the sky. The building has a dark, textured facade and a flat roof with a metal railing. The sky is a mix of blue and purple, suggesting dusk or dawn. The text "Thank you for your attention" is overlaid in the center in a bold, black, sans-serif font.

**Thank you
for your attention**