



# *Electronic properties of solids excited with intermediate laser power densities*

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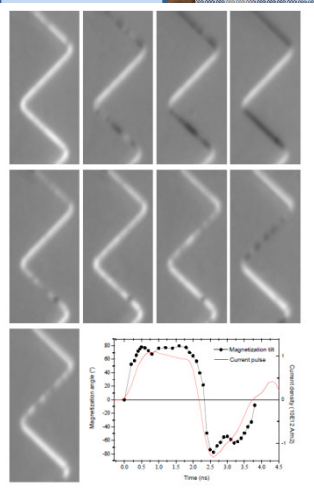


Beamline

1<sup>ère</sup> rencontre des utilisateurs Attolab 19-20/11/2015



# Time resolved Experiments

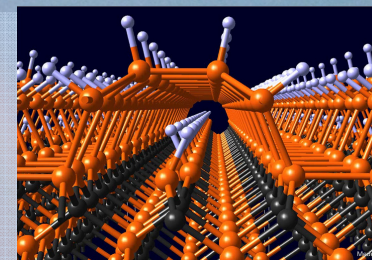


PRB, 2011, 83(2): art.n°020406

- Time resolved electronic structure:

- Real time experiments

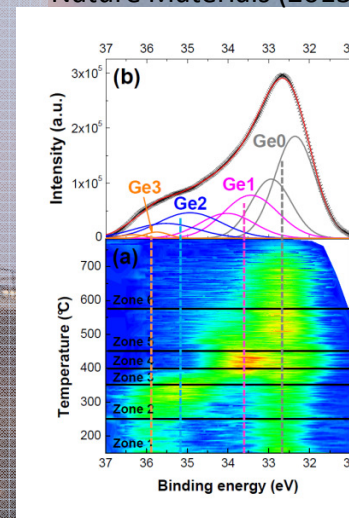
- Annealing processes
- Phase transitions
- Adsorption and desorption



Nature Materials (2013)

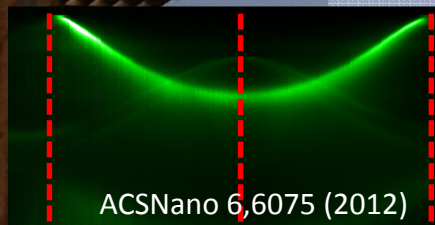
- Pump Probe experiments

- Spin injection & Magnetization dynamics
- Device operation
- Surface photovoltage
- Laser induced magnetization dynamics



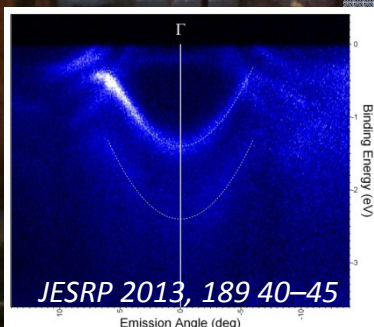
JAP, 2012, 112(9), 093508

E - EF (eV)



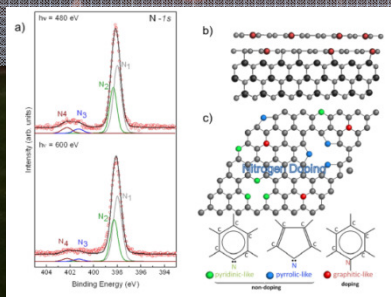
ACSNano 6,6075 (2012)

K'      Γ      K

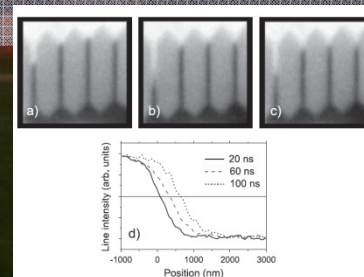


JESRP 2013, 189 40-45

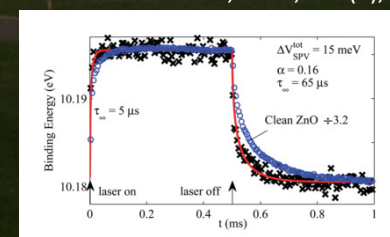
Emission Angle (deg)



ACS Nano, 2012, 6(12): 10893



PRL 108, 247202(2012)



Faraday Discuss. 2014, 171, 275

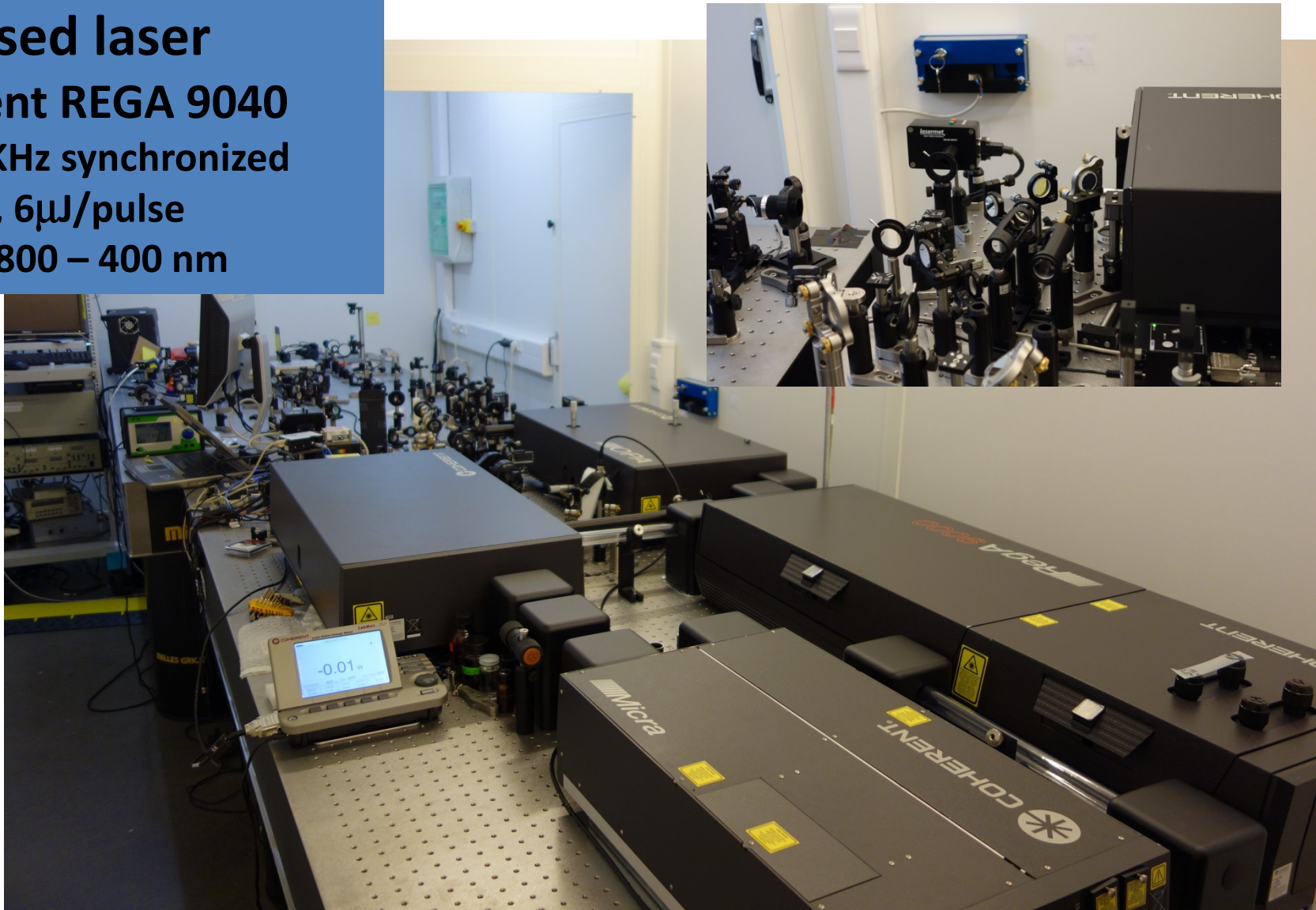


# Laser installation

## Fs Pulsed laser

### Coherent REGA 9040

- 282 KHz synchronized
- 40 fs, 6 $\mu$ J/pulse
- OPA 800 – 400 nm



# fs laser excitations in solid state

- Surface Chemical reactions
- Phase transitions
- Magnetization dynamics

Pump/probe experiments:

Enough power to induce modification of physical properties

**But the solid sample is still there**

# Photoelectron spectroscopy

- Electronic structure =>
- all properties
  - physical, chemical, magnetic,
- Element and site specific
- Basis for all time resolved studies
  - In all phase transitions electrons, more mobile than nuclei could drive the observed behavior

Needed several  $\text{mJ}/\text{cm}^2$

REGA :  $6\mu\text{J}/\text{pulse}$   
with a focal spot of about 200 microns



$5 \text{ mJ}/\text{cm}^2$  @ 25 degrees incidence



High stability needed for both synchrotron and laser beamlines

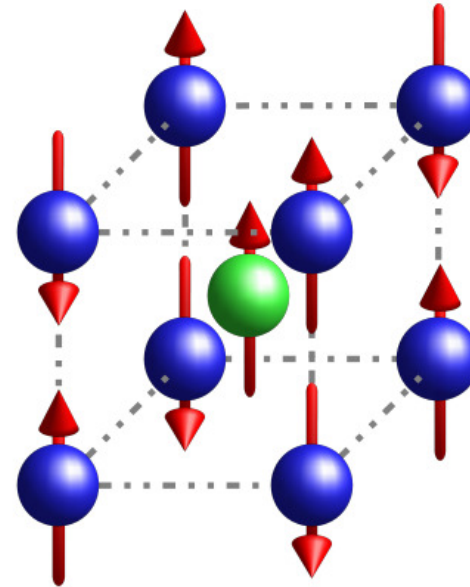


# The phase transition in FeRh

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

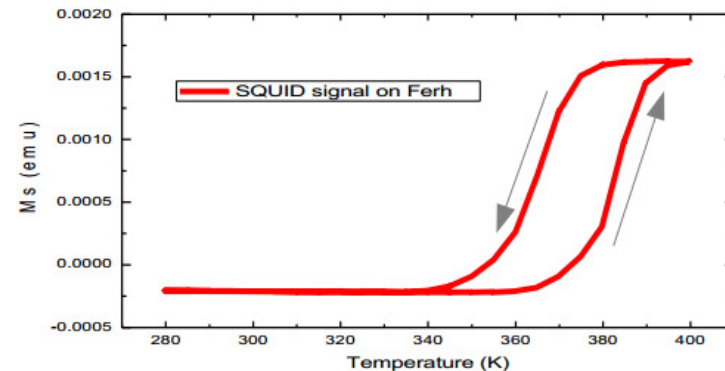
Below  $T_c$   
 $Fe = \pm 3.3 \mu_B$   
 $Rh = 0 \mu_B$

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;

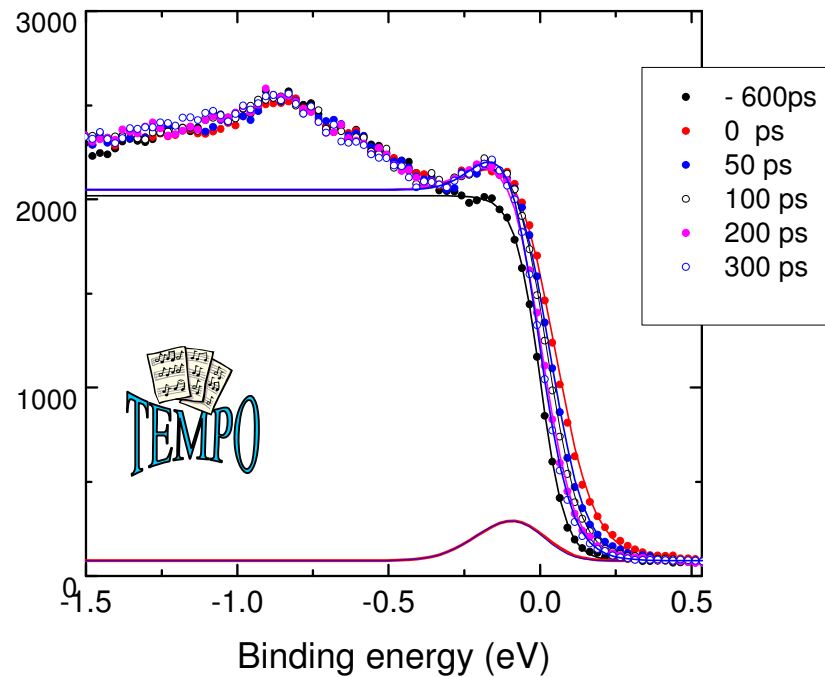
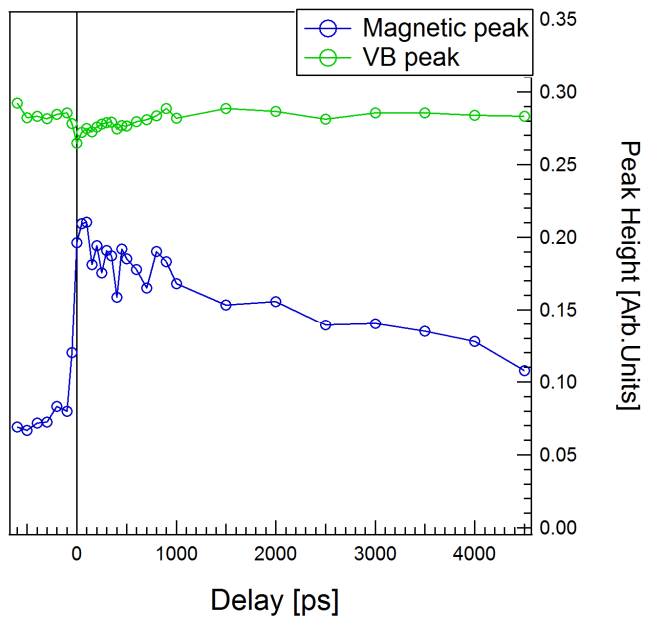
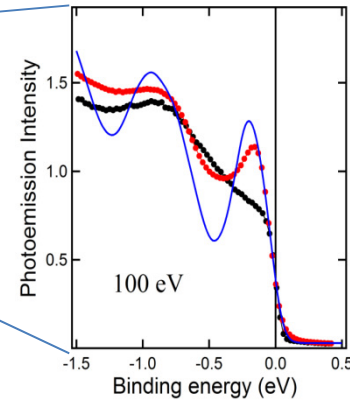
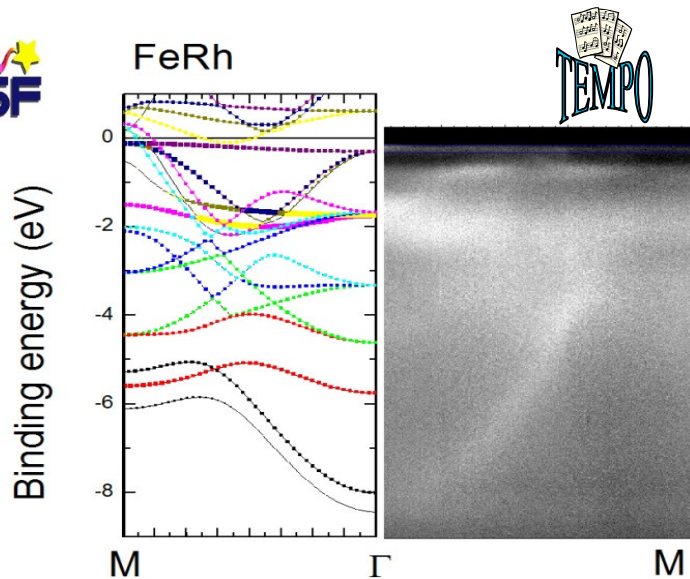
- Isotropically in bulk or in thick films;
- Along the out of plane direction for **thin films**



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



# FeRh : electronic structure -> + laser induced phase transition

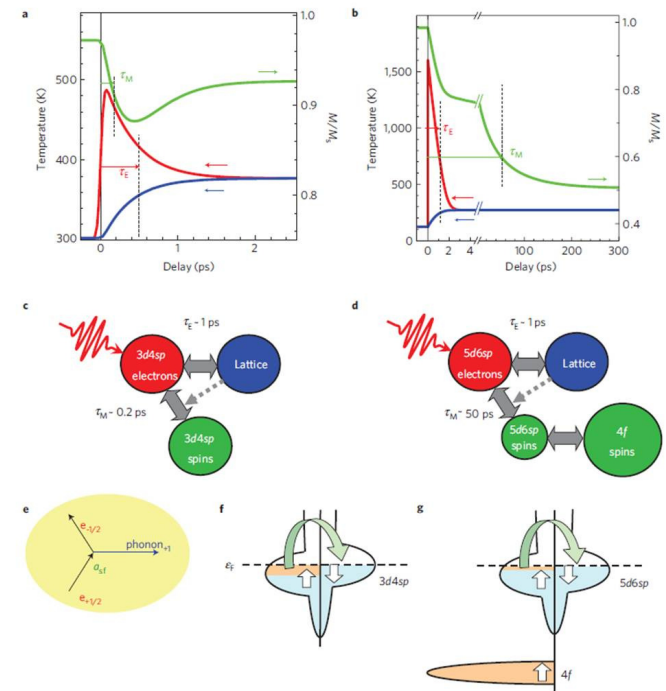


(F. Pressacco, M. Gatti, A. Nicolaou, D. Krizmancic)



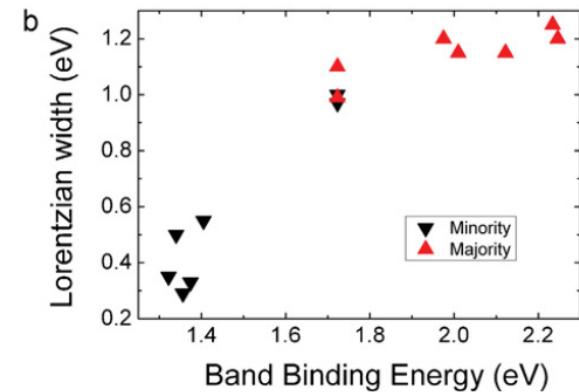
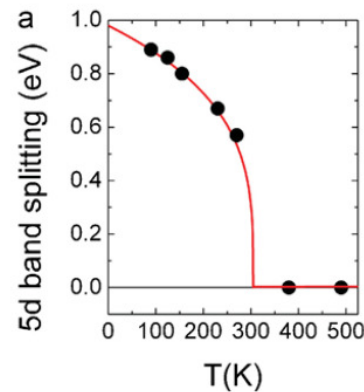
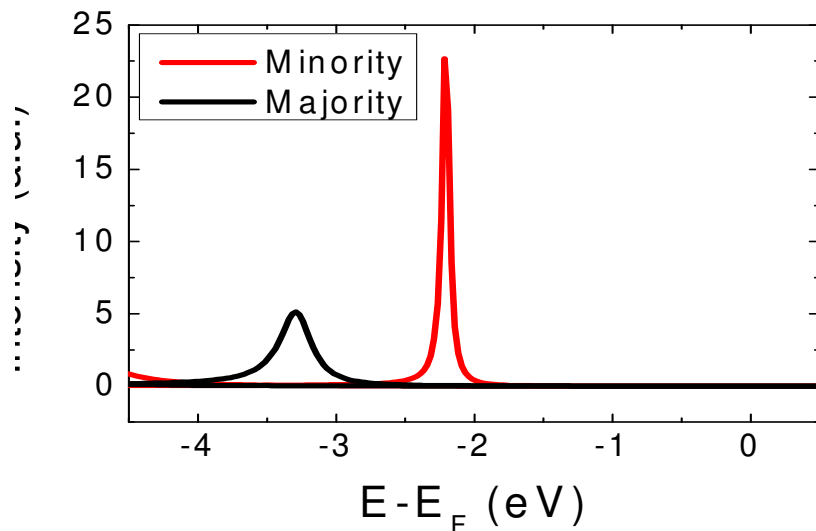
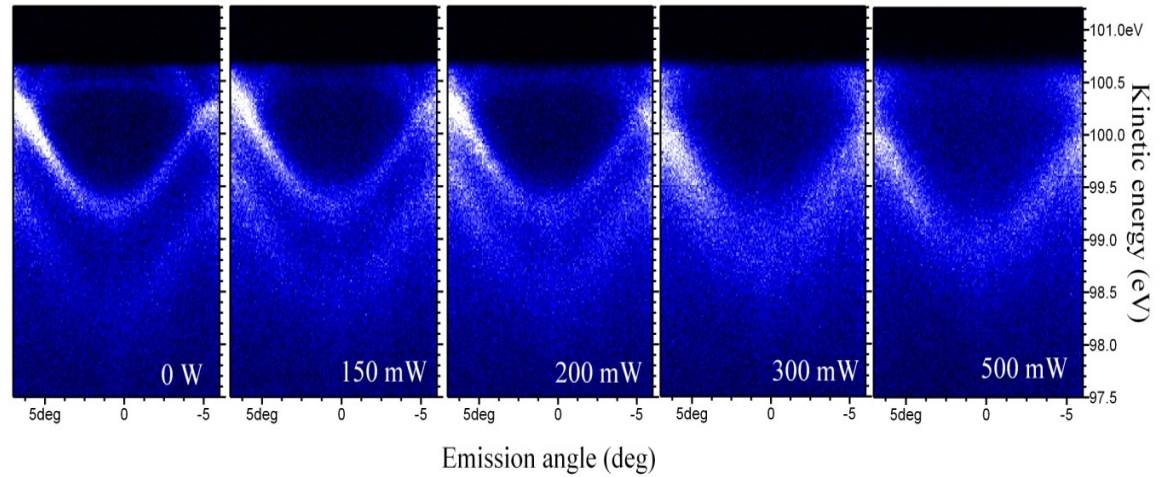
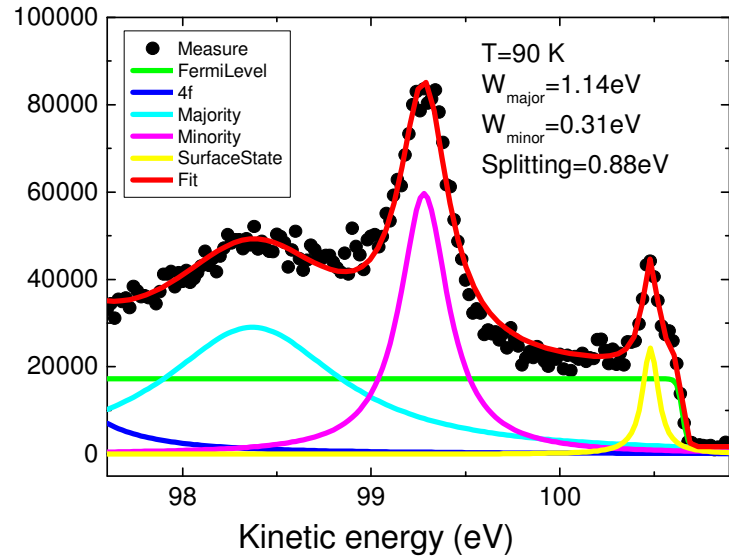
# Magnetization dynamics

- Three temperature model
- Role of hot electrons
- IR laser excitation



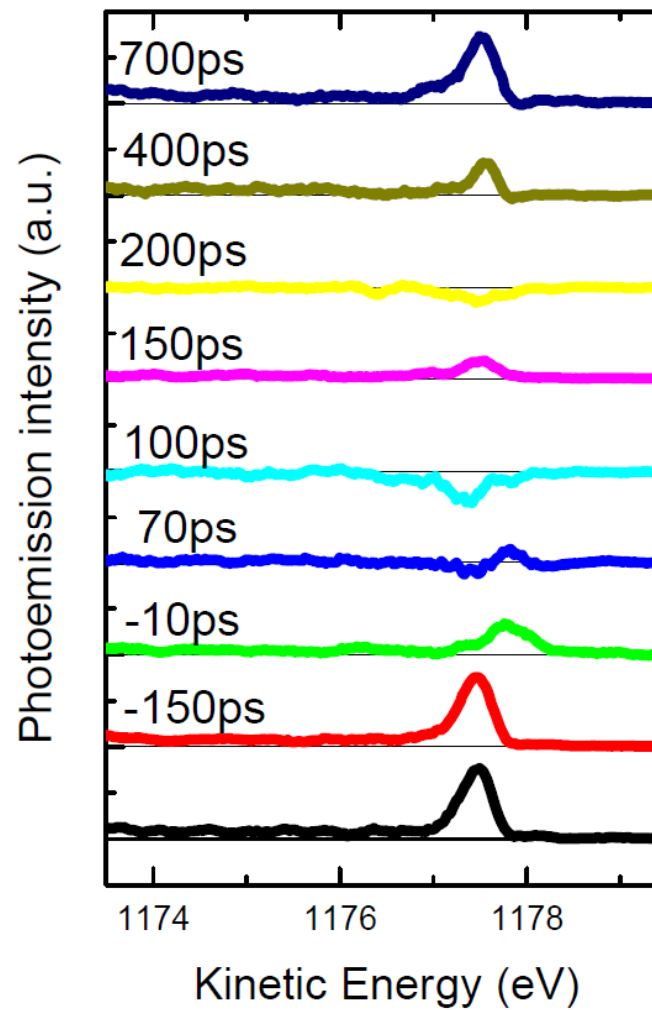
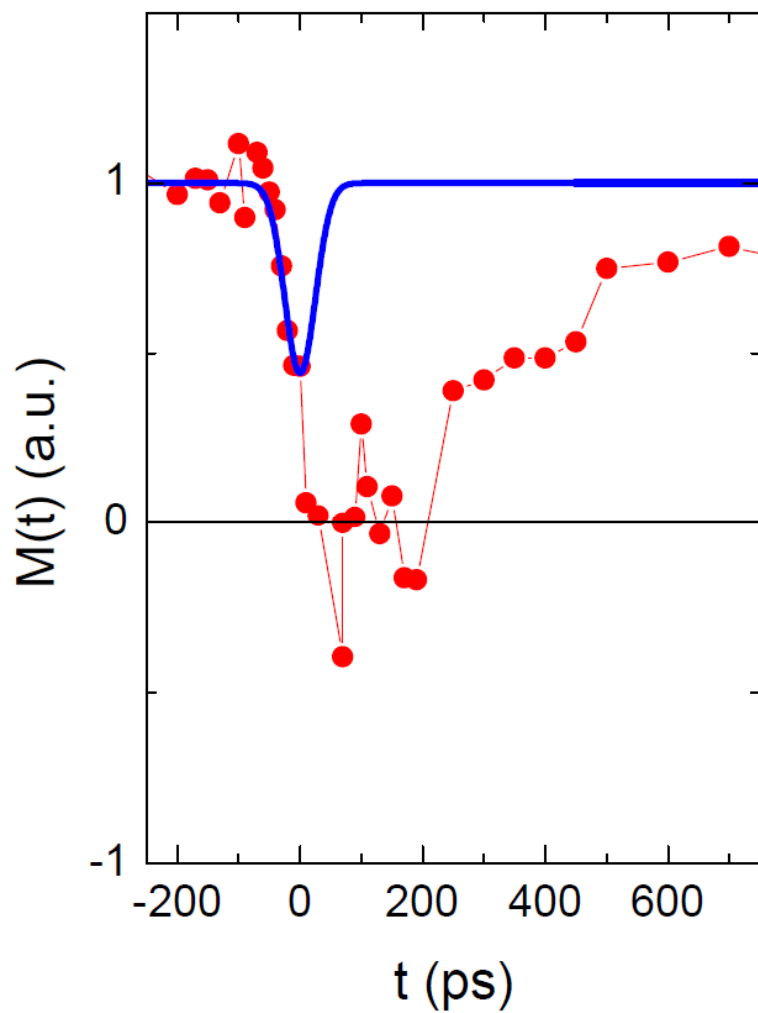
extracted from [Nature Materials Vol: 9, Pg: 259–265 (2010)].

# Gd: Temperature dependence of magnetization





# *Laser induced Fast demagnetization*



## Femtosecond Laser Excitation Drives Ferromagnetic Gadolinium out of Magnetic Equilibrium

Robert Carley,<sup>1,2</sup> Kristian Döbrich,<sup>1</sup> Björn Frietsch,<sup>1,2</sup> Cornelius Gahl,<sup>2</sup> Martin Teichmann,<sup>1,2</sup> Olaf Schwarzkopf,<sup>3</sup> Philippe Wernet,<sup>3</sup> and Martin Weinelt<sup>2,\*</sup>

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(Received 10 April 2012; published 31 July 2012)

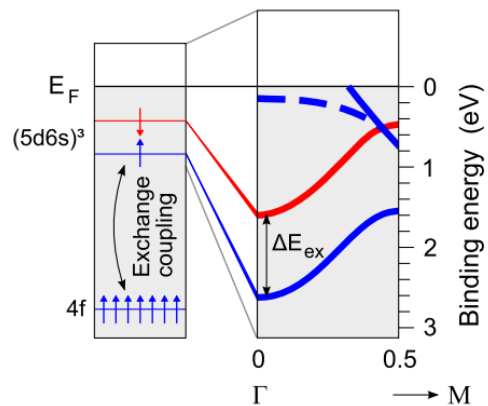
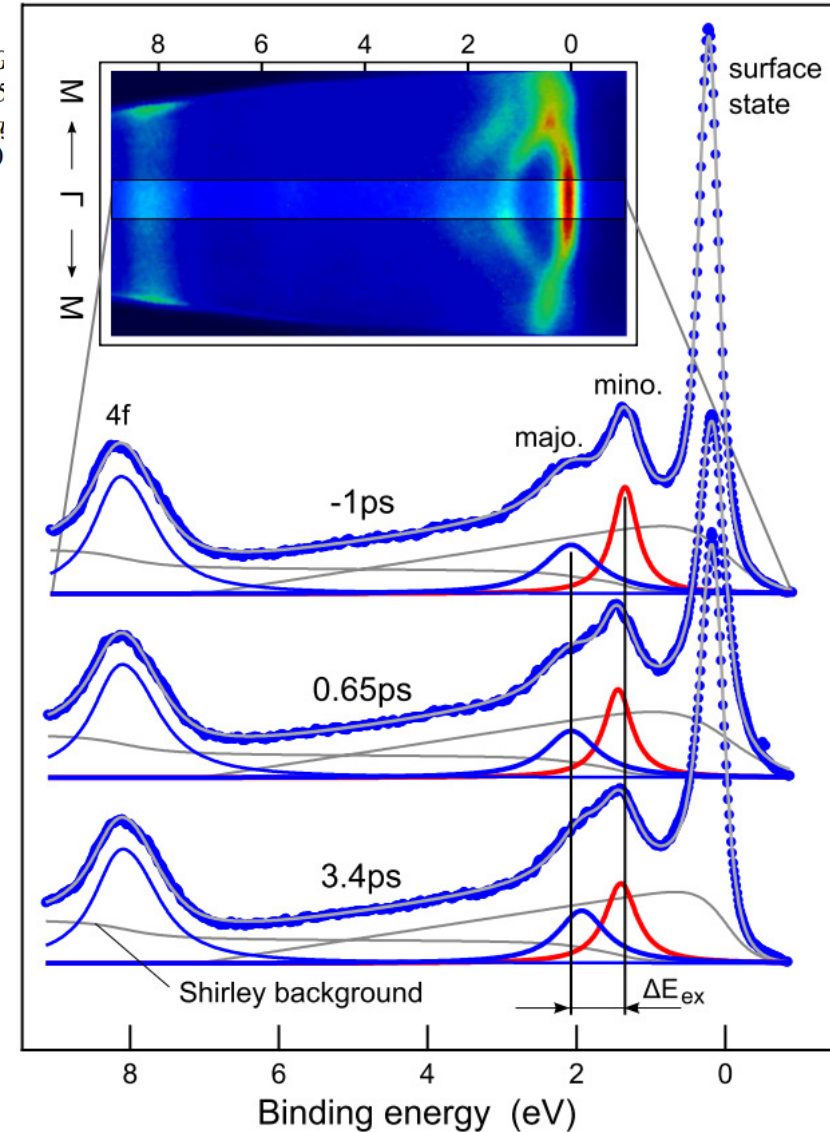
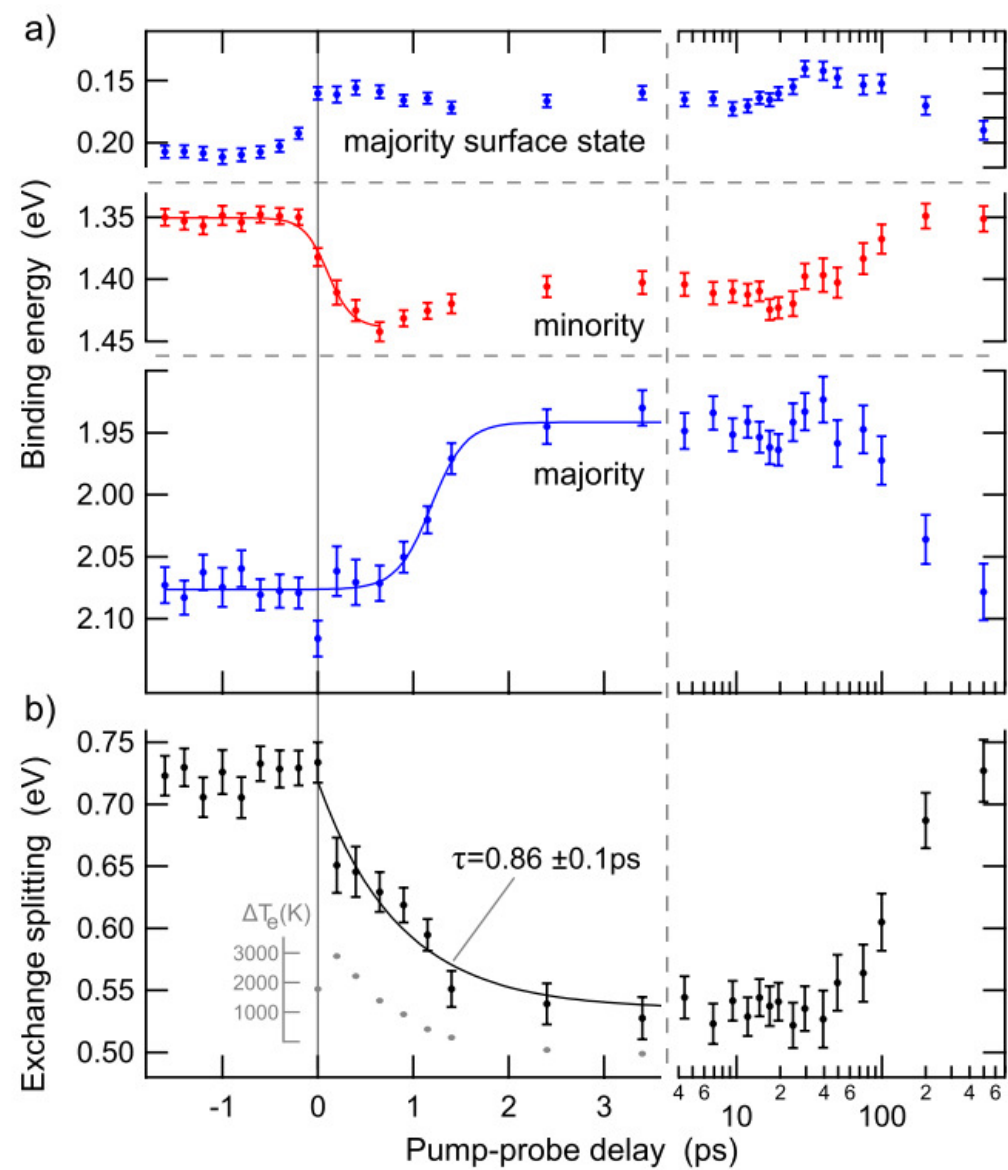


FIG. 1 (color online). Magnetic coupling (left) and calculated exchange-split valence band structure [31] of gadolinium (right). Majority spin band: blue, up arrows. Minority spin band: red, down arrows. The dashed line is the  $5d_{z^2}$  majority spin surface state. Bands not seen in ARPES at 36 eV photon energy have been omitted.



HHG source

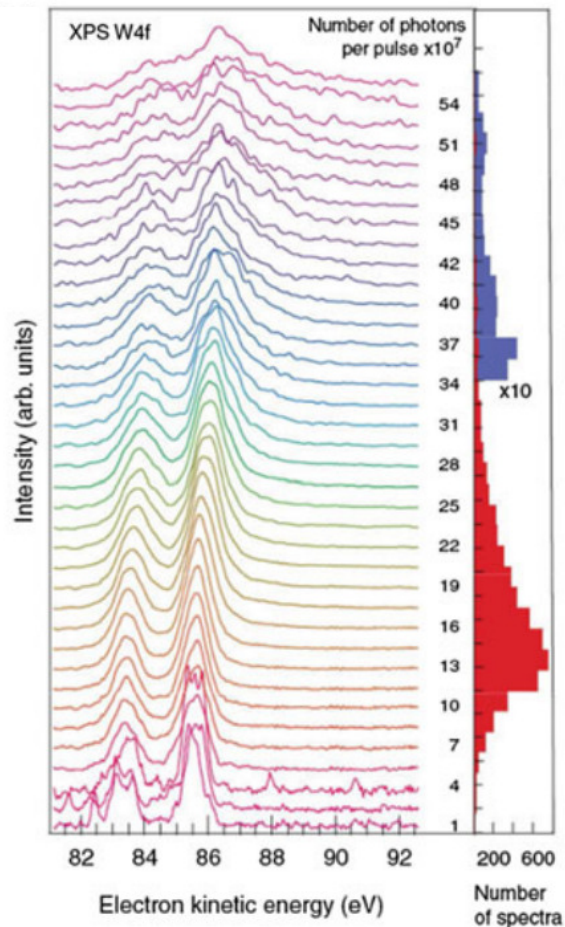




Relatively high power density  
Space charge created by pump  
pulse.



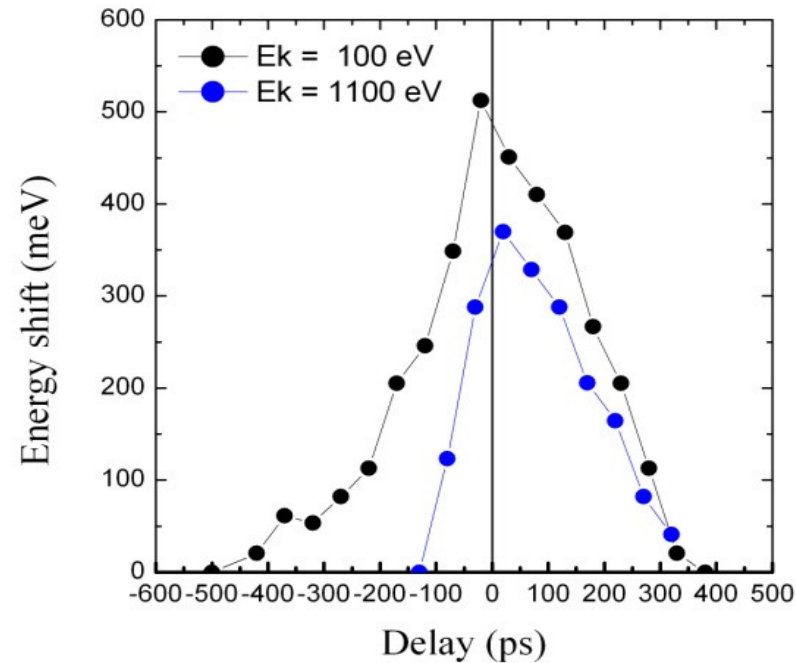
# Photoemission as a function of the photons per pulse



A. Pietzsch et al., *Towards time resolve core level photoelectron spectroscopy with femtosecond x-ray free-electron lasers*.  
New J. Phys. **10**, 033004 (2008)

FLASH

## Space charge effects occurring during fast demagnetization processes.



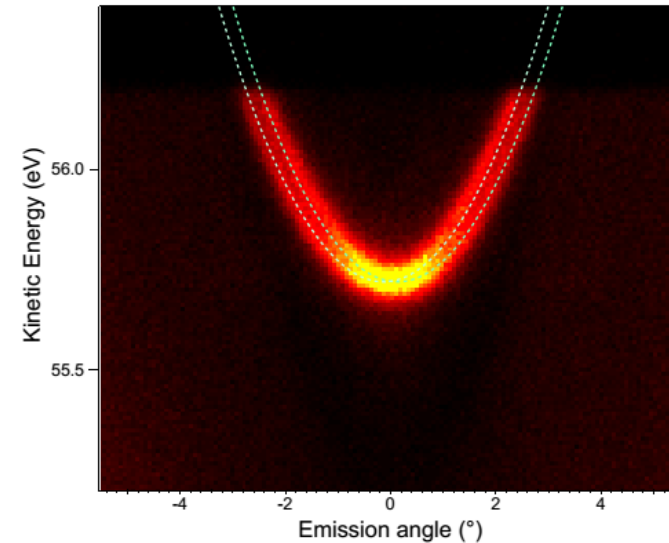




# Multiphoton Photoemission

F. Sirotti, N. Beaulieu, A. Bendounan, M. G. Silly, C. Chauvet TEMPO Beamline SOLEIL  
G. Malinowski Université de Lorraine, Nancy  
G. Fratesi CNISM U. Milano-Bicocca, Italy & ETSF  
G. Onida Dip. Fisica, U. Milano, Italy & ETSF  
Valerie Veniard, LSI Palaiseau & ETSF

Synchrotron radiation  
60 eV photon energy



Au(111) - 90 K

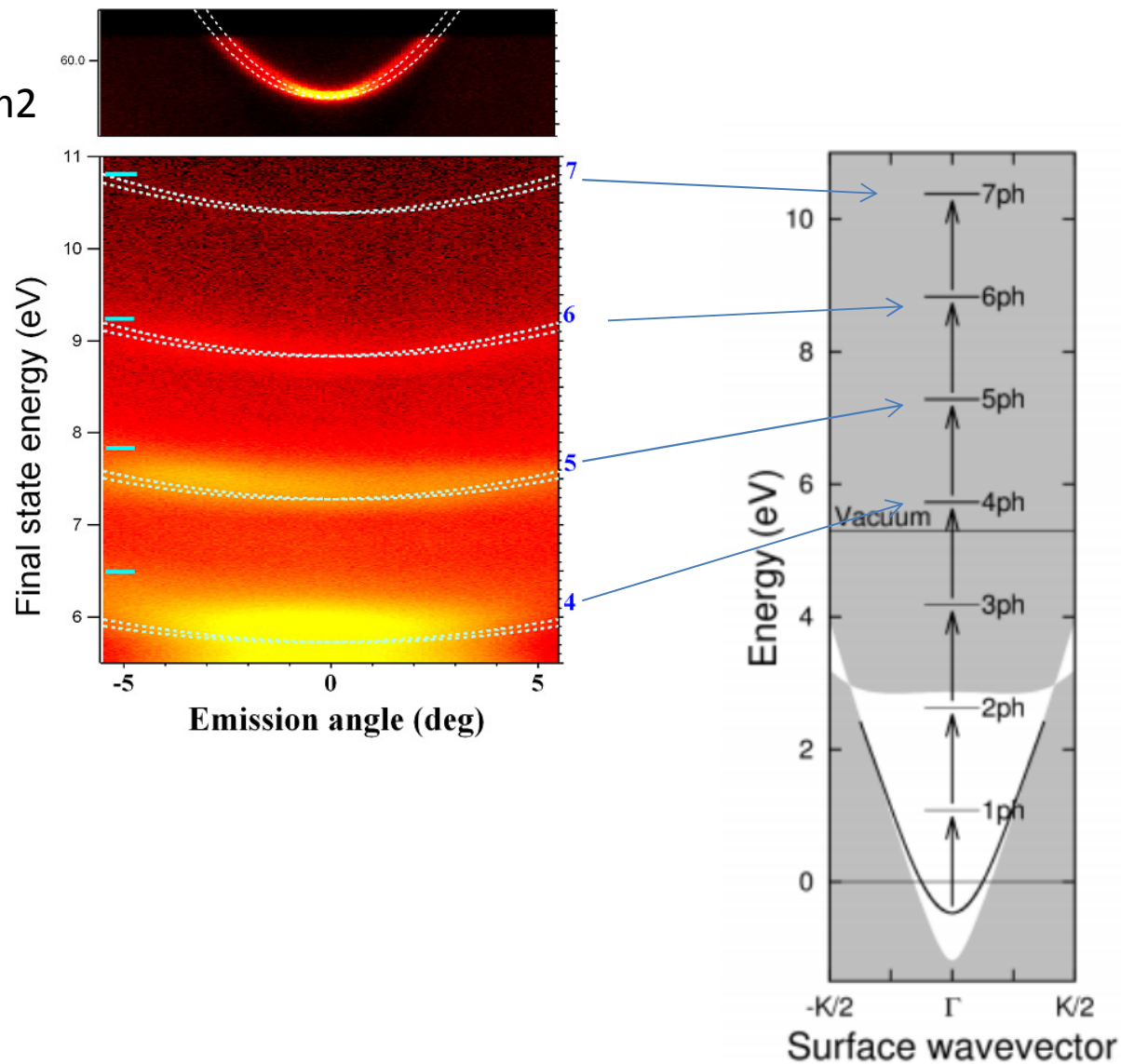
Phys Rev B 90, 035401 (2014)



# Multiphoton photoemission

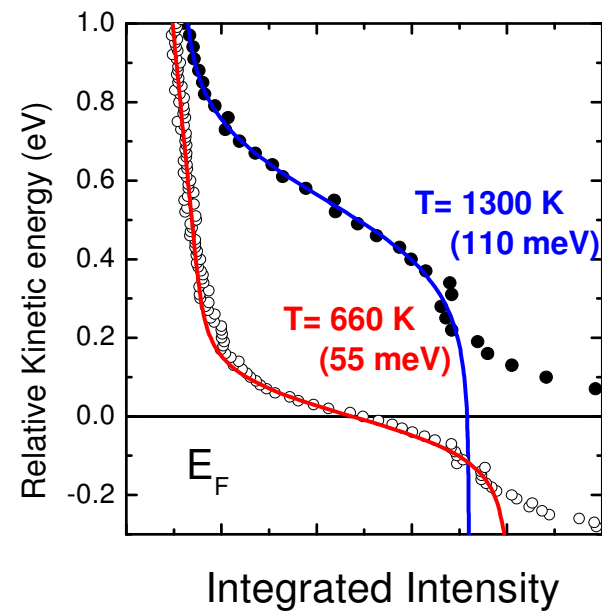
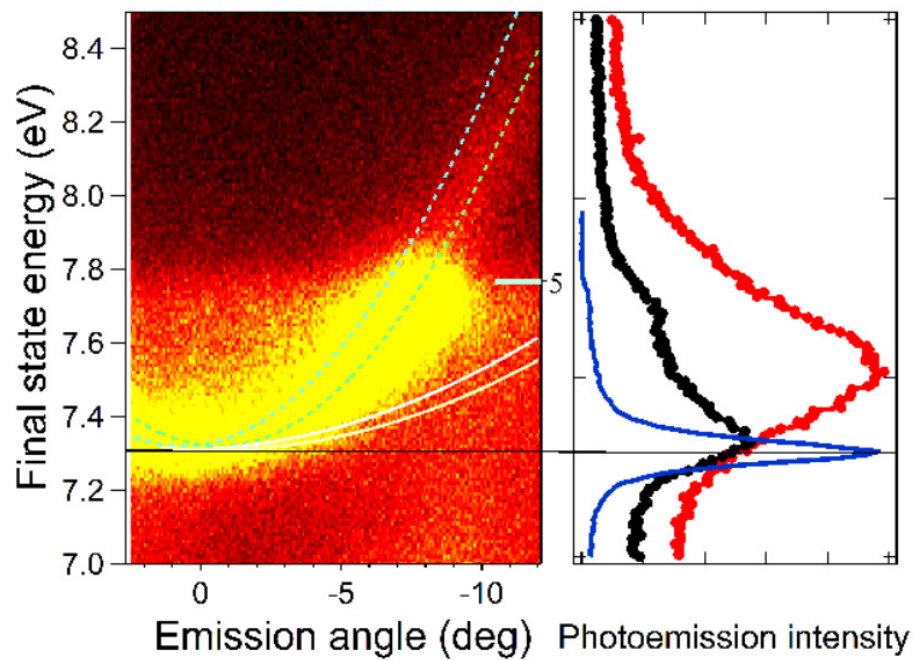
REGA : 2mJ/cm<sup>2</sup>

800 nm





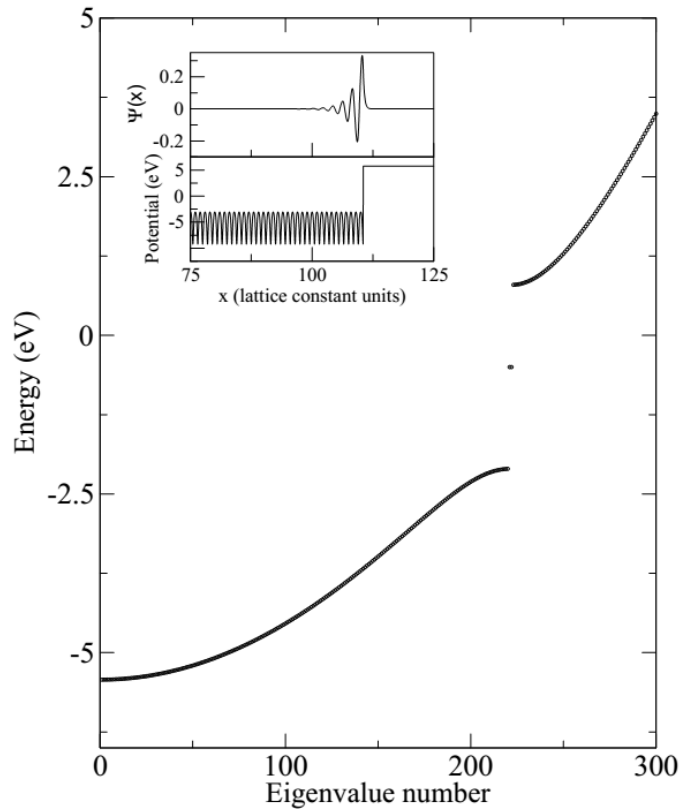
# Multiphoton photoemission





# Multiphoton photoemission

Simple model to describe the excitation of the electron in the surface state

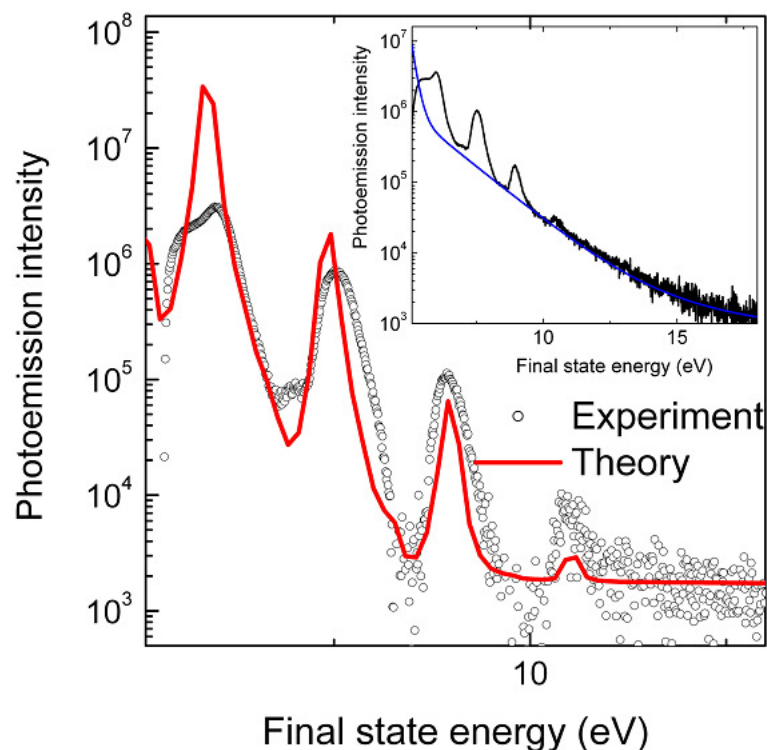


$$V(x) = \sum_n \frac{V_0}{\sqrt{(x - na_0)^2 + 1}} \exp(-k_{scr}|x - na_0|).$$

Time dependent Schrödinger equation  $\longrightarrow$



# Multiple electronic excitations with laser pulses at high power densities

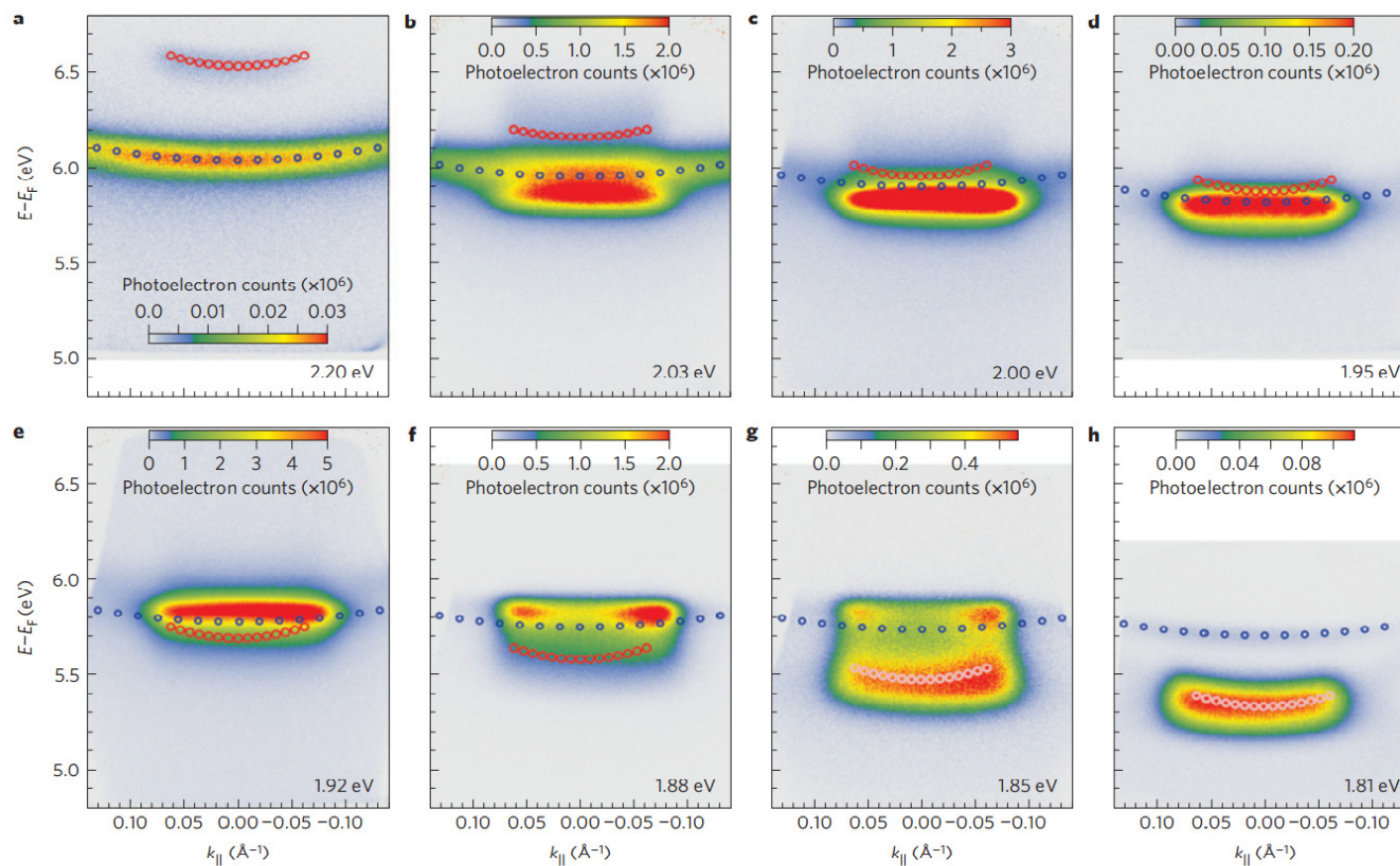


Good description of the  
Electronic excitation

Good agreement of laser power density

# Transient excitons at metal surfaces

Xuefeng Cui<sup>1</sup>, Cong Wang<sup>1</sup>, Adam Argondizzo<sup>1</sup>, Sean Garrett-Roe<sup>2</sup>, Branko Gumhalter<sup>3</sup>  
and Hrvoje Petek<sup>1\*</sup>



# Conclusion:

electronic structure:

photon energies in the range up to 100 eV

No need to destroy samples

- High repetition rate fs pulses
- Continuous reliable operation
- Dedicated optimized exp. stations

# Thanks to

M. Silly  
A. Bendounan  
C. Chauvet  
F. Pressacco  
N. Beaulieu



*Theory*  
*LSI Palaiseau & ETSF*

M.Gatti  
V. Veniard  
G. Onida  
G. Fratesi

