Ultrafast Dynamics in Solids Probed and Induced by Soft X-rays

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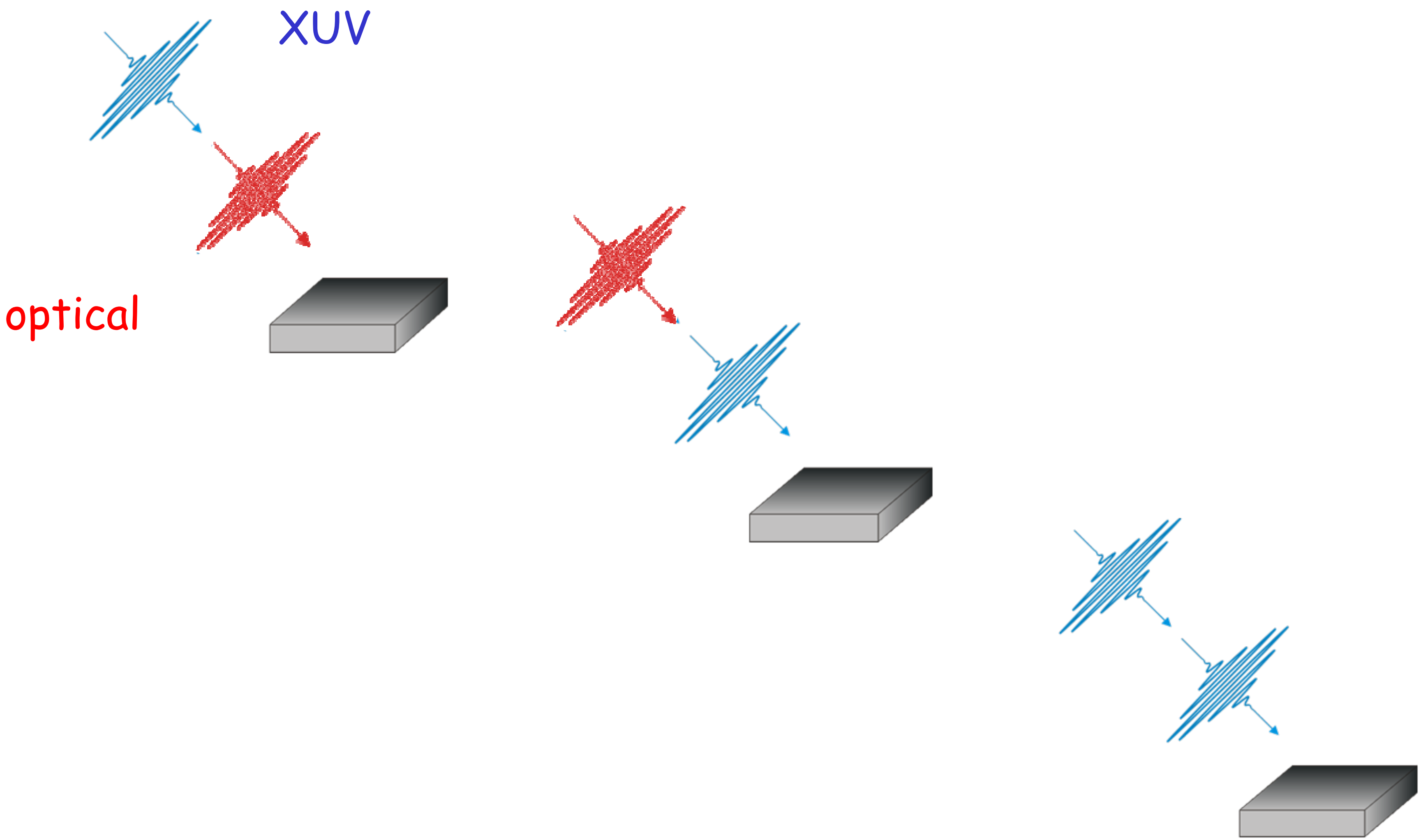
University of Hamburg

Applications of time-resolved spectroscopy at FLASH



Collaborators

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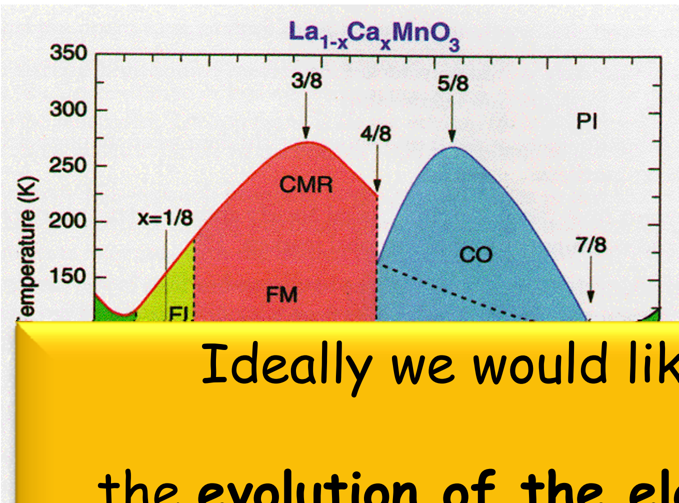
Priority program
FSP 301-FLASH

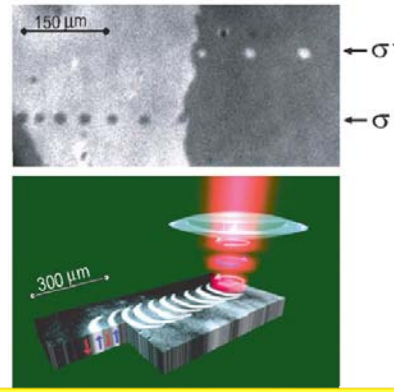
bmb+f
Großgeräte der physikalischen Grundlagenforschung

Some Questions in Soft X-ray Materials Science at FEL´s

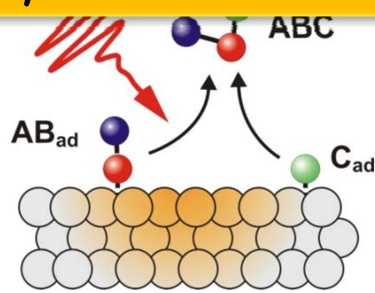
Can we understand and control strongly correlated electron systems ?

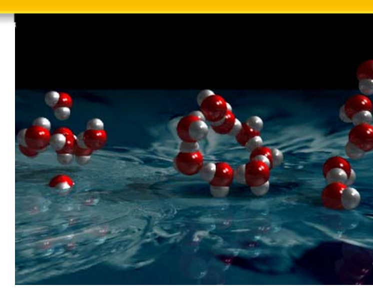


How fast can we switch magnetisation ?



Ideally we would like to follow the **position of the atoms**
<u>and</u>
the **evolution of the electronic states** at any given point in time, i.e. map out multi-dimensional energy surfaces and see how complex systems evolve on these surfaces





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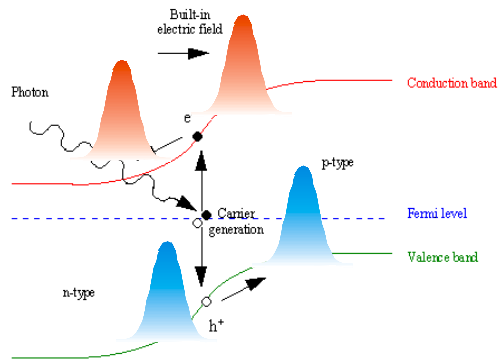
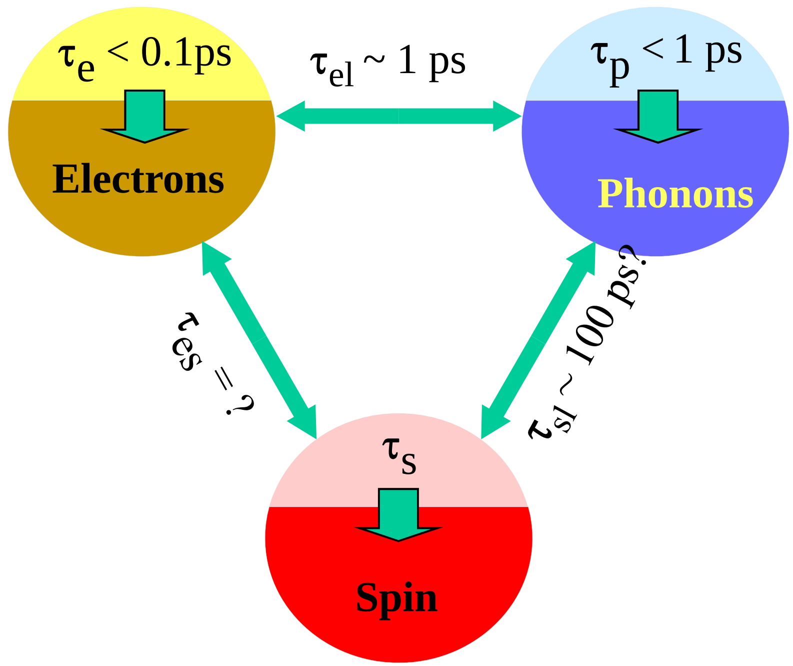
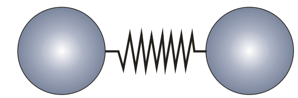
Surfaces

Liquids

Challenge:
Electron, spin, and nuclear dynamics and their interplay

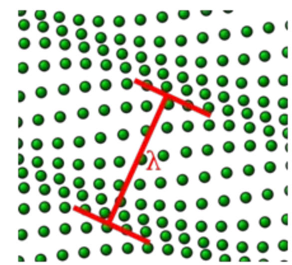






Molecular vibrations

phonons

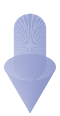


plasmon excitations (screening)

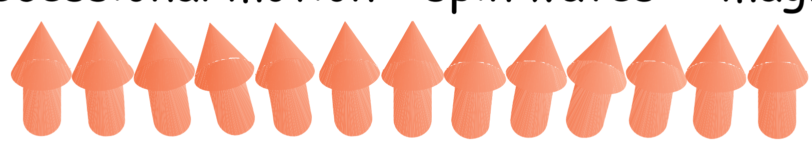
resonant electron transfer

e-h pair excitations

Stoner excitations

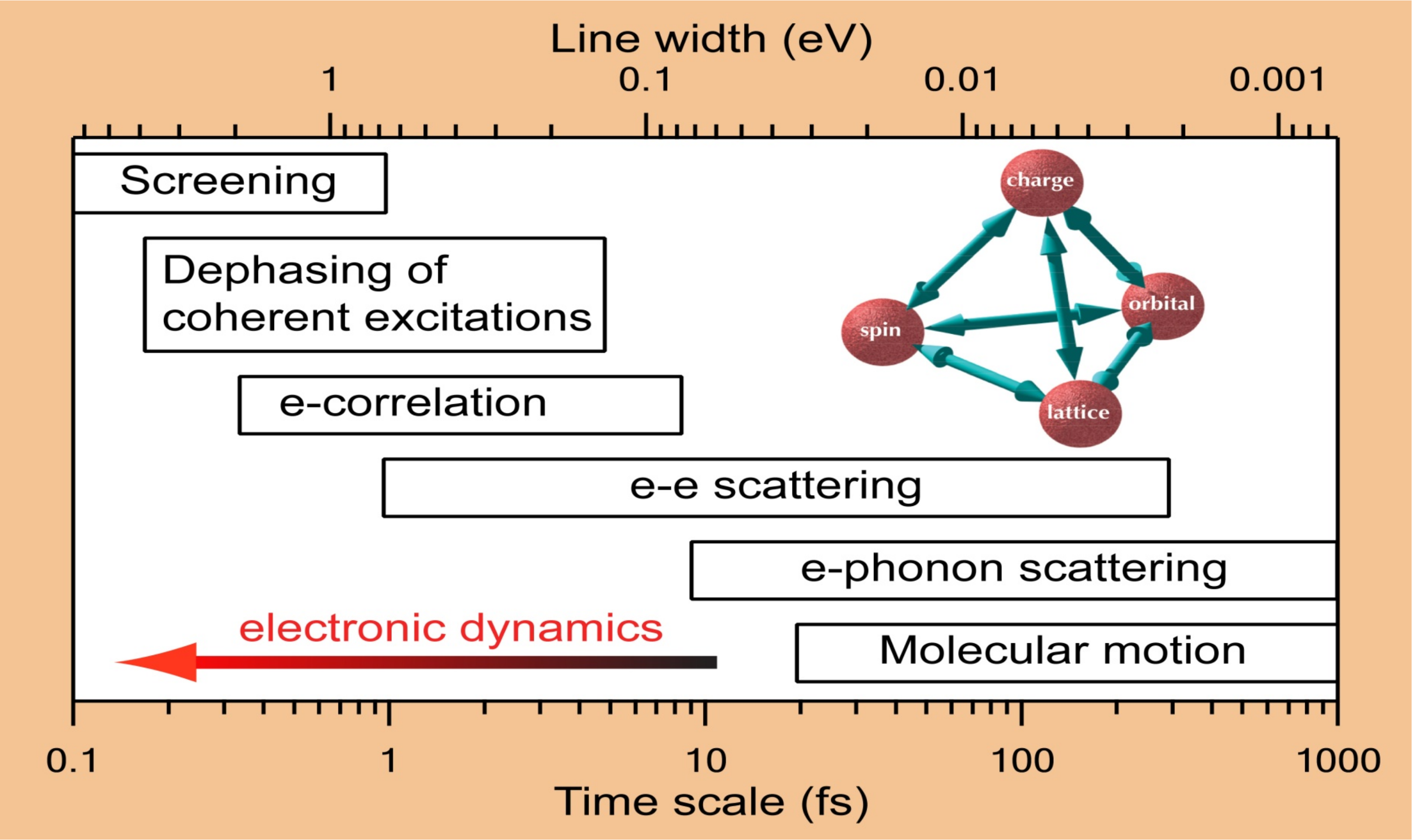


precessional motion – spin waves - magnons



Relevant timescales for electronic relaxation

(Neglecting spin)



Reproduced from Petek et al., Prog. Surf. Sci. **56**, 239 (1997).

What can soft-x-rays do for you?

probe electronic structure !

momentum

$$E(k, R_{nuc}, \sigma)$$

spin

atomic position

ARPES

ESCA

photoemission
x-ray absorption
x-ray emission

NEXAFS

XMCD - XMLD

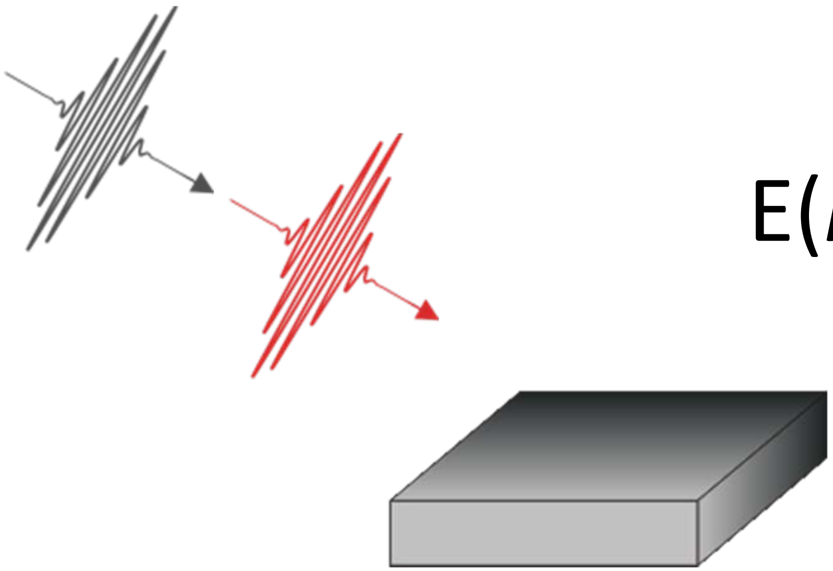
XES

RIXS

Stroboscopic (Pump-Probe) Experiments

From static to dynamic

$$E(k, R_{nuc}, \sigma, t)$$



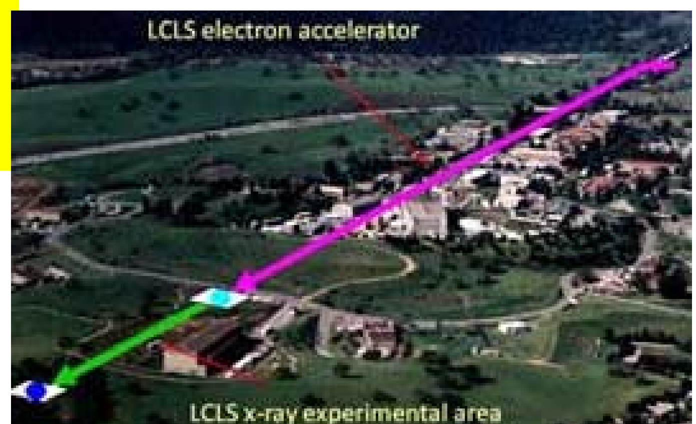
Opportunities at Free Electron Lasers

User facility since 2005

60 nm < λ < 4.45 nm

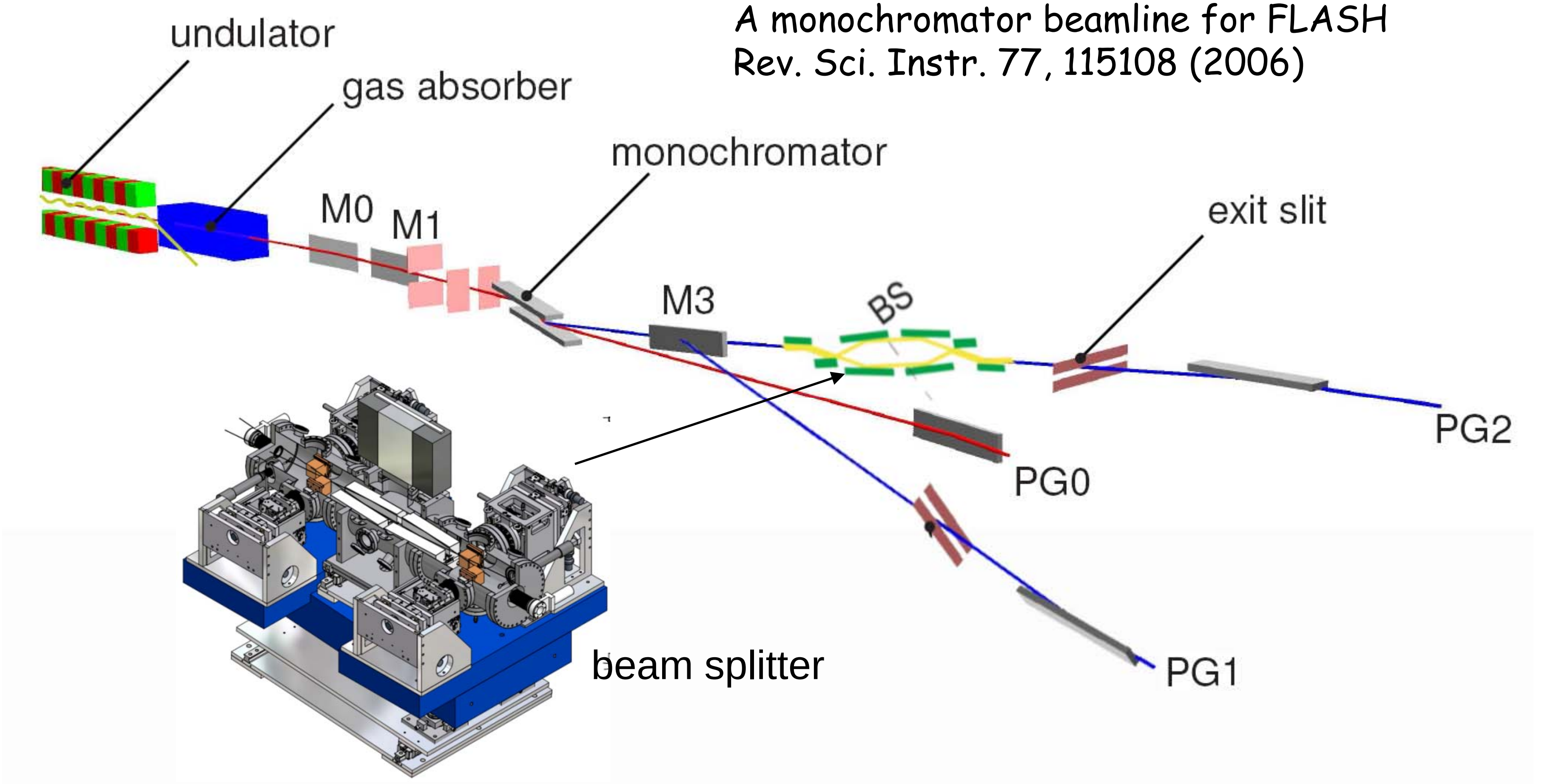
Soft X-ray Materials Science (SXR) at LCLS (since May 2010)

2.3 nm < λ < 0.6 nm



The PG-beamlines at FLASH

M. Martins, M. Wellhöfer, J.T. Hoeft, W. Wurth, J. Feldhaus, R. Follath
A monochromator beamline for FLASH
Rev. Sci. Instr. 77, 115108 (2006)



M. Wellhöfer, M. Martins, and W. Wurth, A. Sorokin and M. Richter
Performance of the monochromator beamline at FLASH
J. Opt. A: Pure Appl. Opt. 9 (2007) 749 - 756

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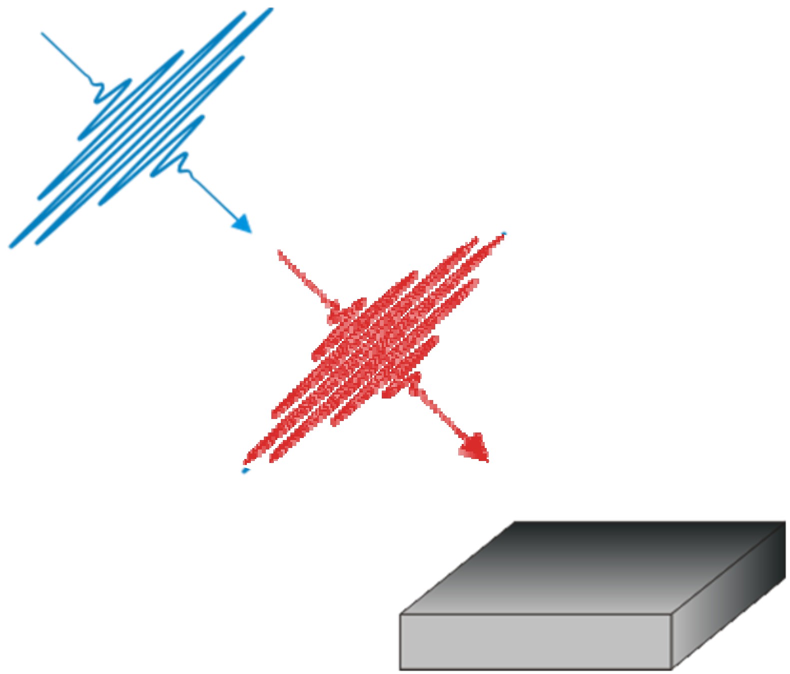
Großgeräte der physikalischen Grundlagenforschung

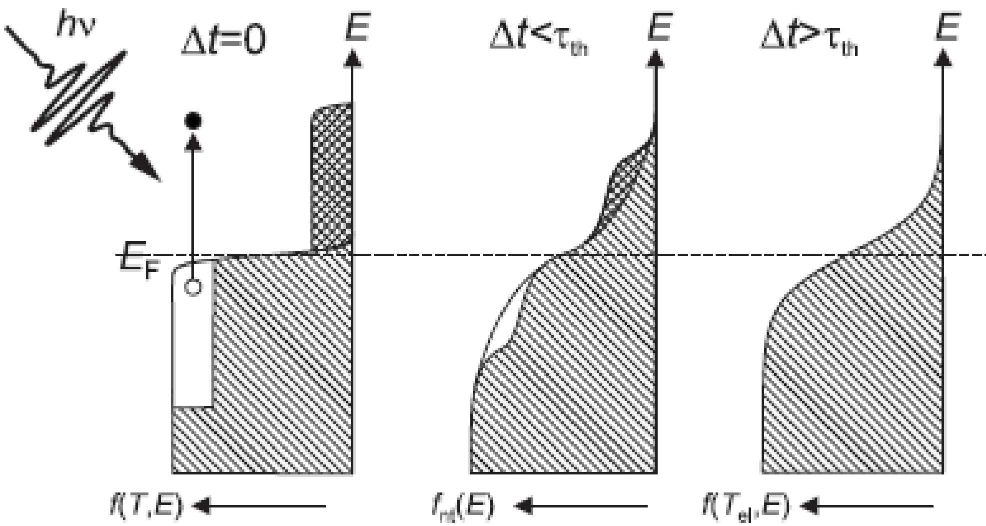
Optical-pump XUV-probe



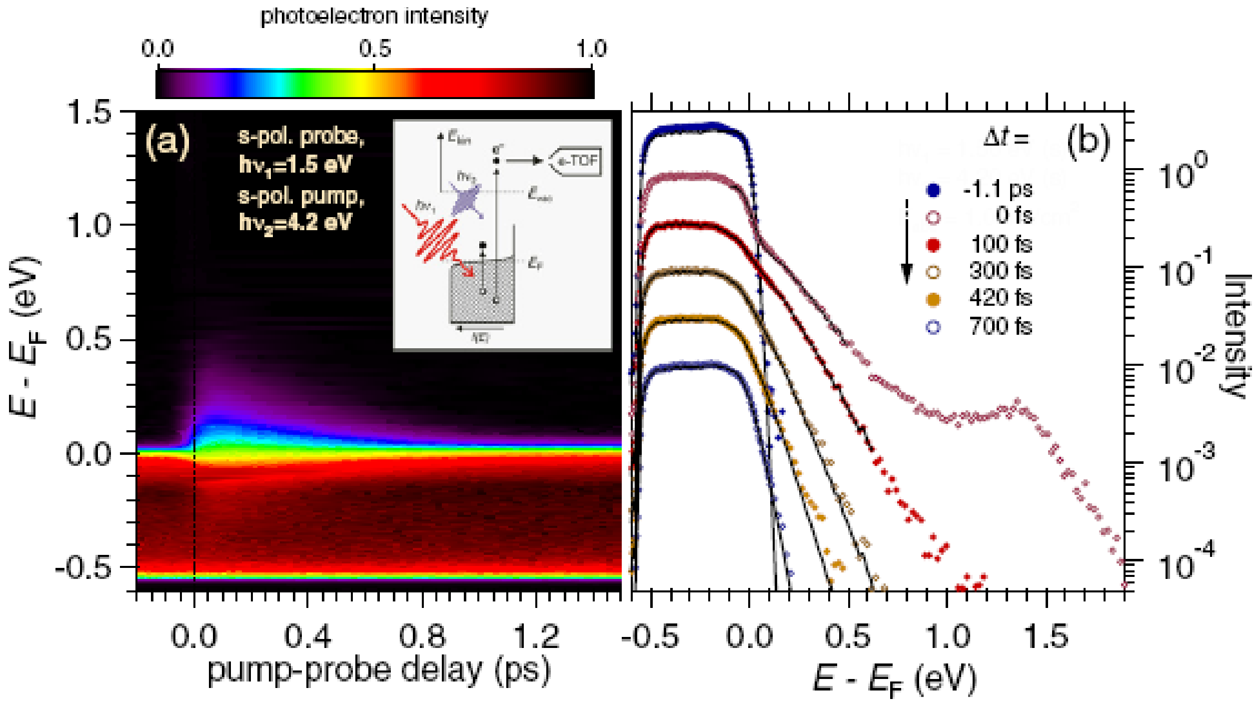
An atomic view on the dynamics of charge order in TaS_2
from time-resolved core-level photoemission

Snapshots of electronic structure relaxation
in highly photoexcited Si
taken with time-resolved x-ray emission spectroscopy

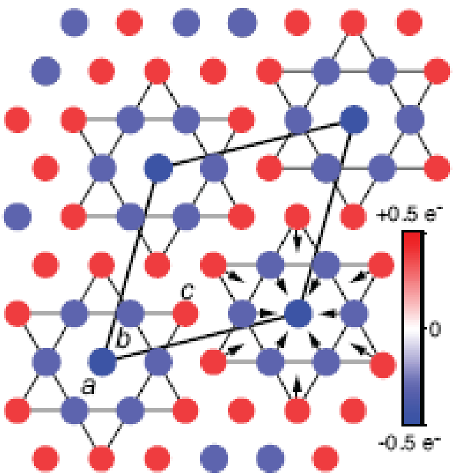
Optical pumping – „heating the electrons“ ?



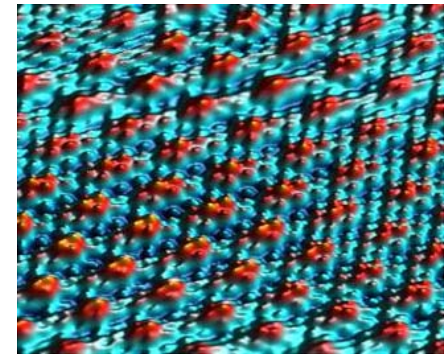
From U. Bovensiepen
J. Phys.: Condens. Matter
19, 083201, (2007)



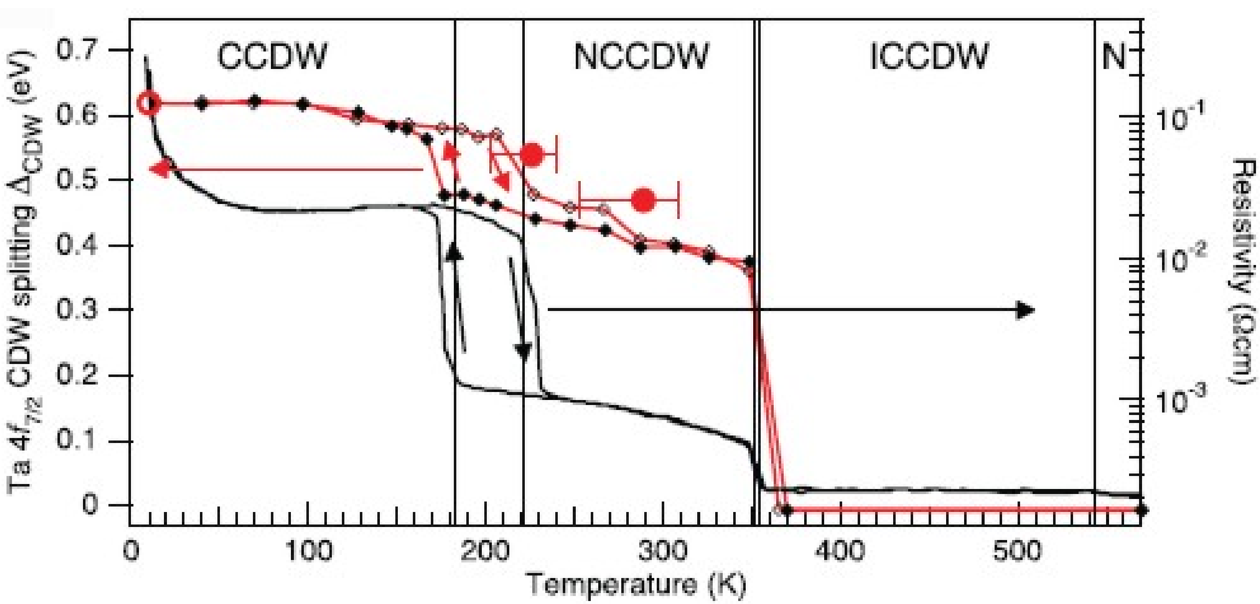
1T-TaS_2 – a layered CDW / Mott-insulator

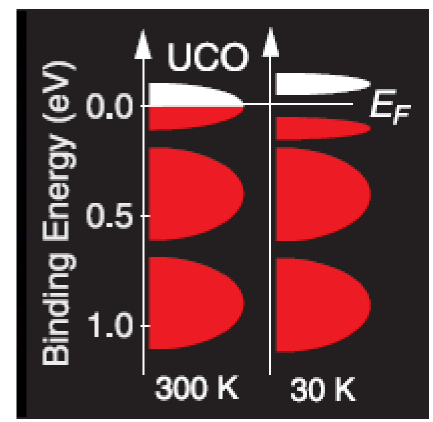


Charge Ordered State:
13 atom cluster
„Star of David“



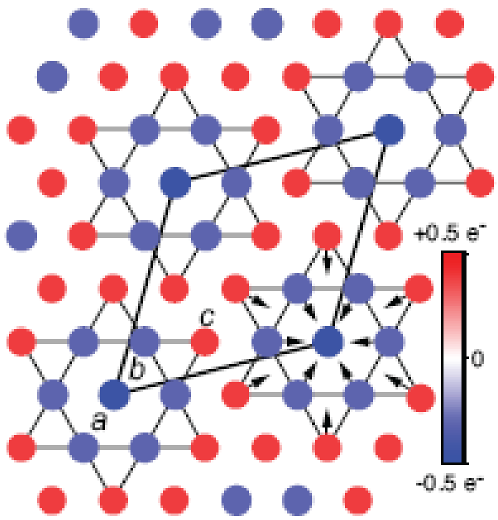
J. Wiebe, UniHH

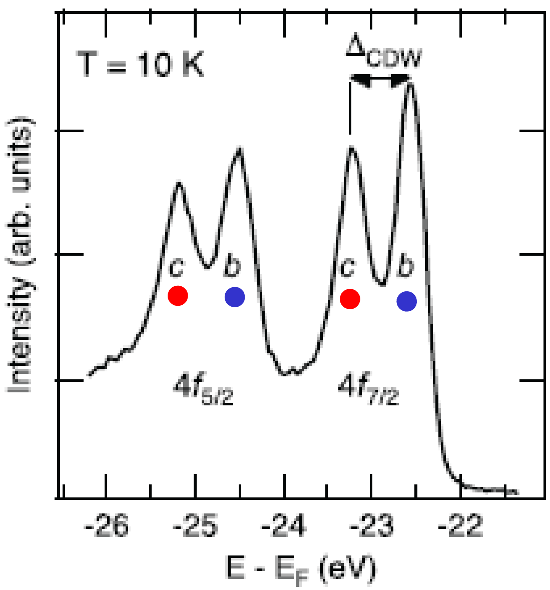




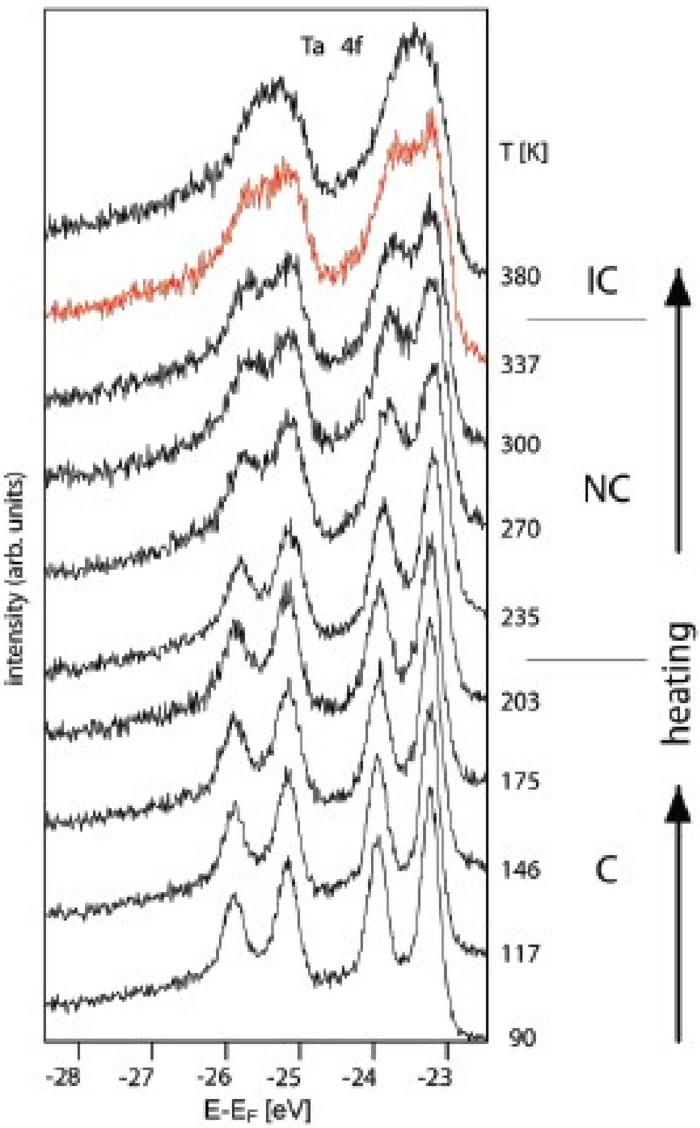
L. Perfetti et al.
PRL **97**, 067402 (2006)

Ta 4f photoemission – a probe for charge order in TaS_2

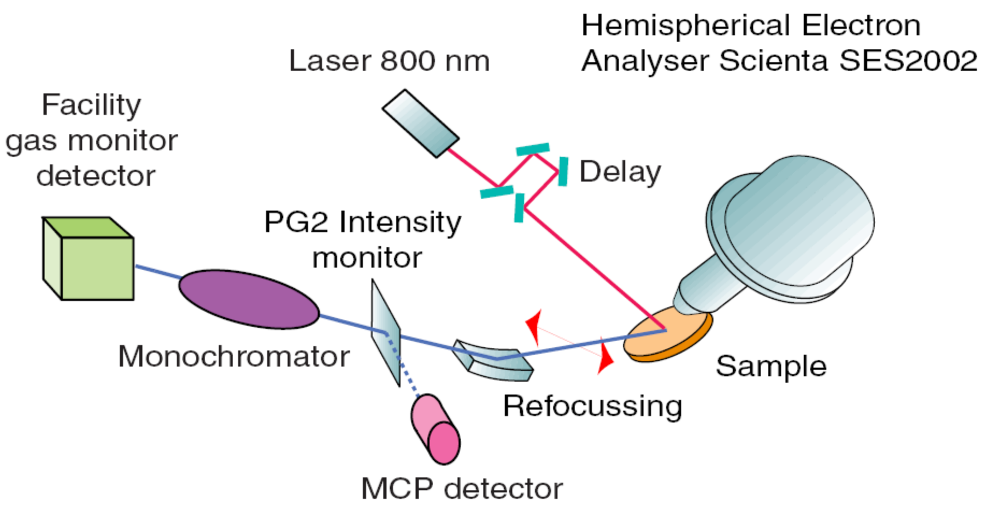




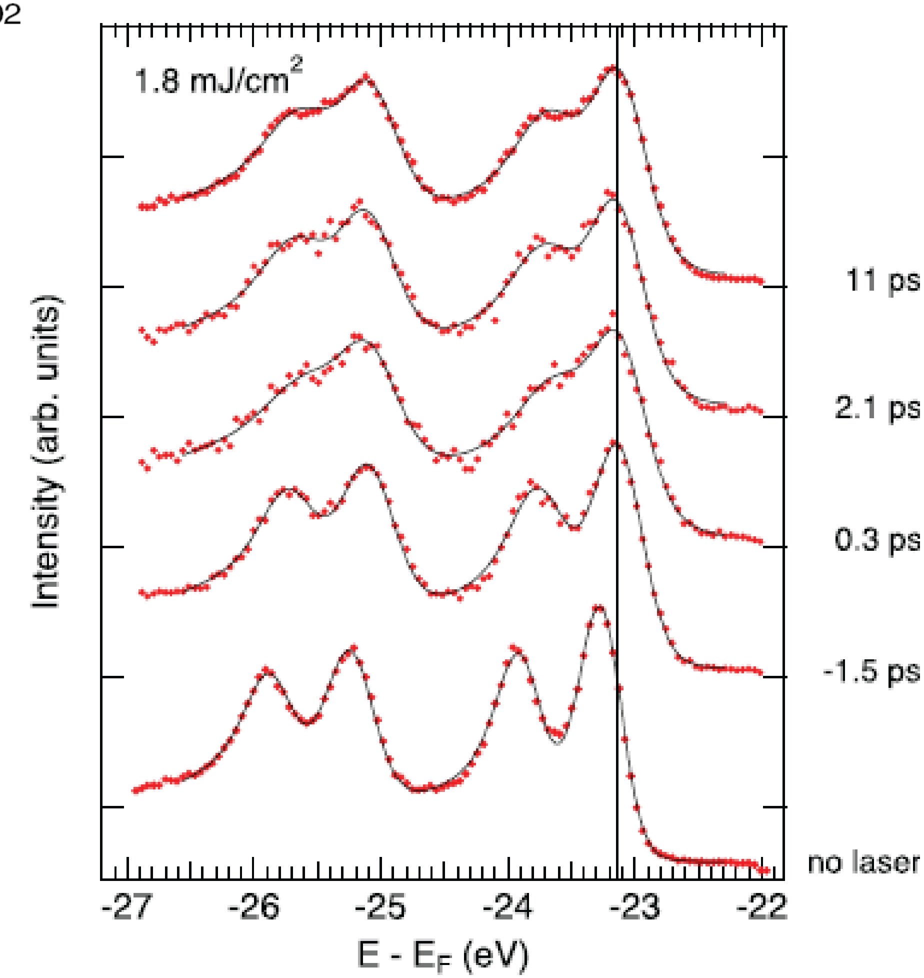
Equilibrium dynamics



Charge state dynamics after photoexcitation



A. Pietzsch et al., NJP **10** (2008) 033004



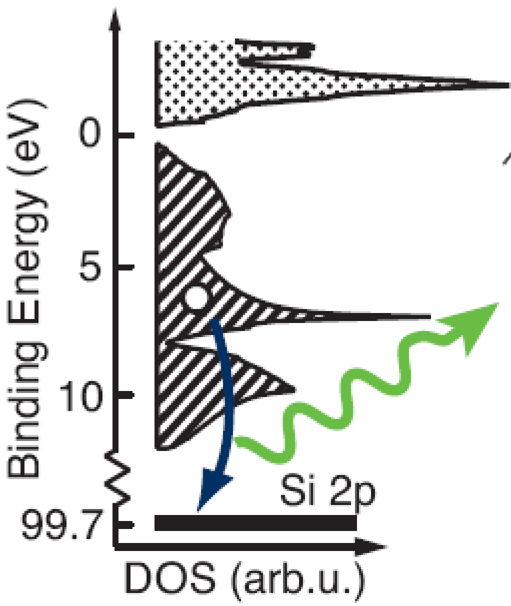
Optical pump: 800nm, 120fs
XUV-probe: 156 eV, 160fs, 3rd harm.

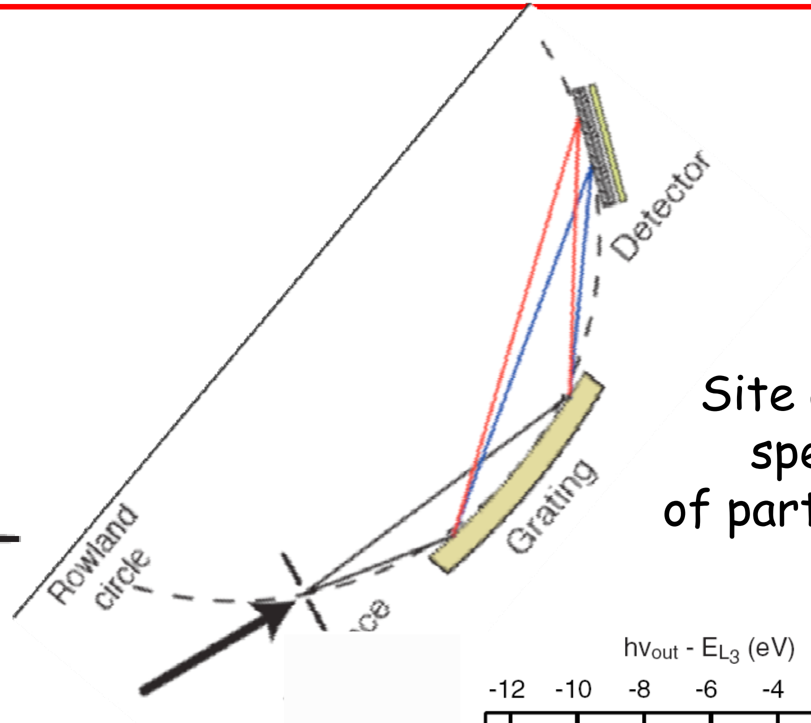
S. Hellmann, K. Rossnagel et al., cond-mat arXiv:1004.4790v1

Conclusions: Electronically driven transition

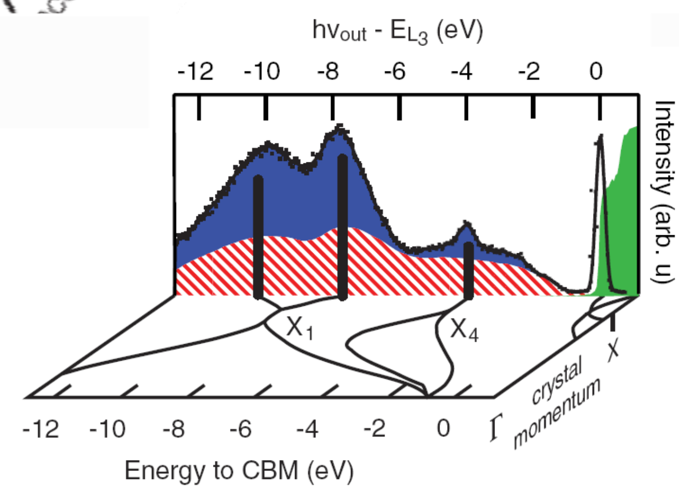
- Transient decoupling of CDW and lattice distortion
- Same time scale as Mott gap recovery
- Fast formation of domain walls (~ 1ps)

X-ray Emission Spectroscopy



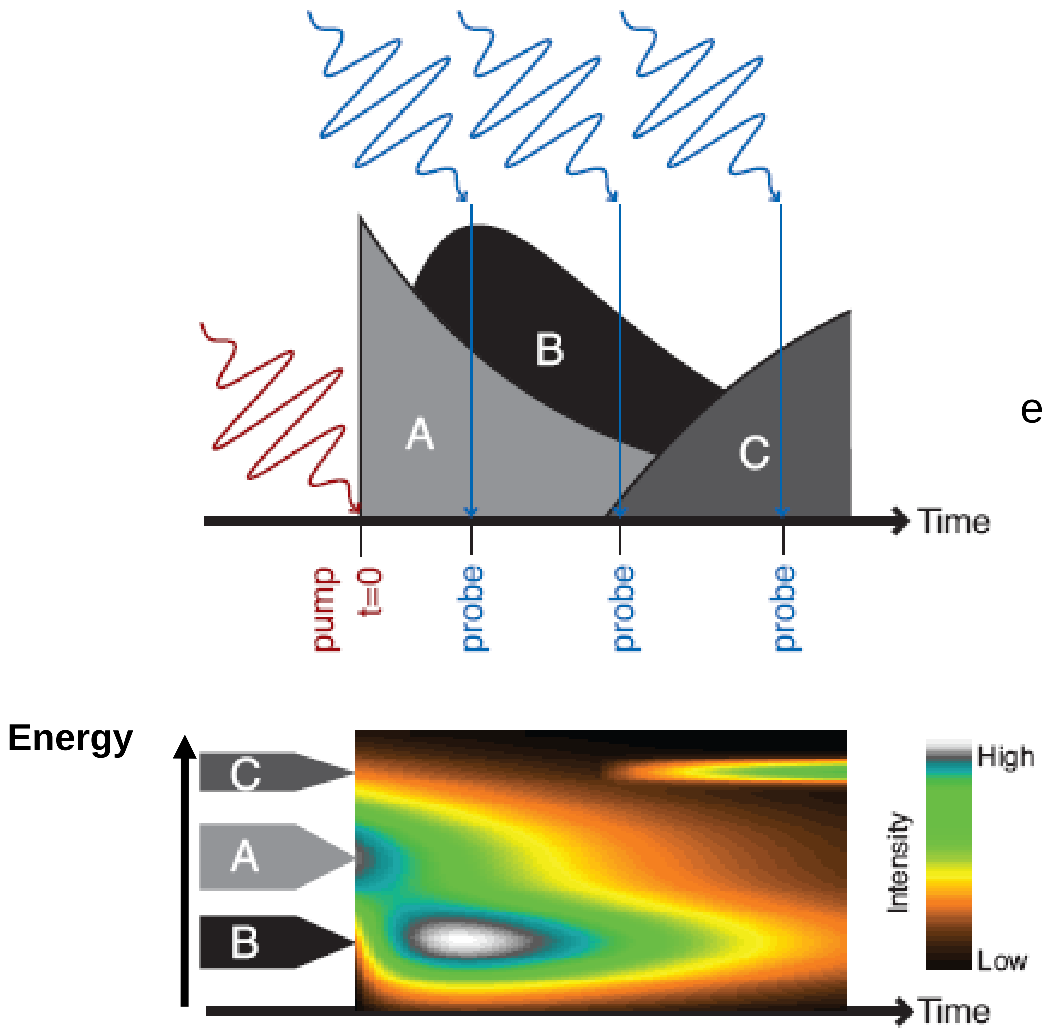


Site and element specific map of partial density of states



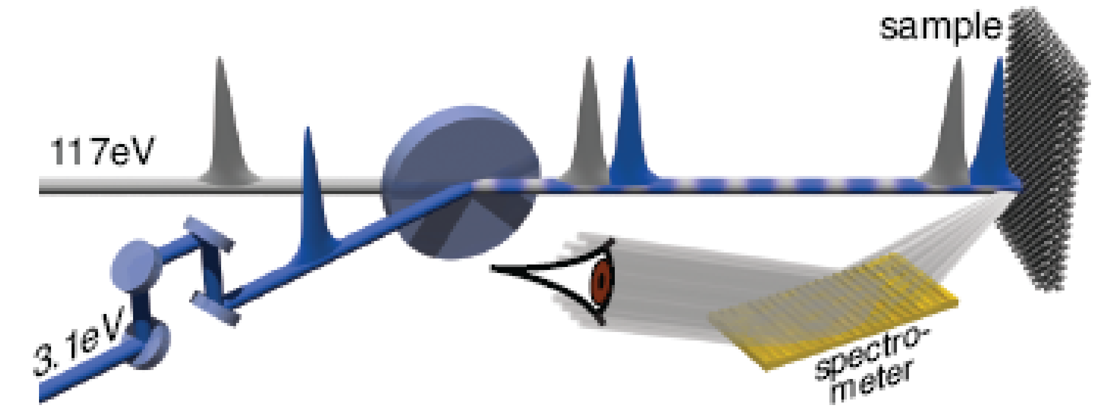
- time-resolved

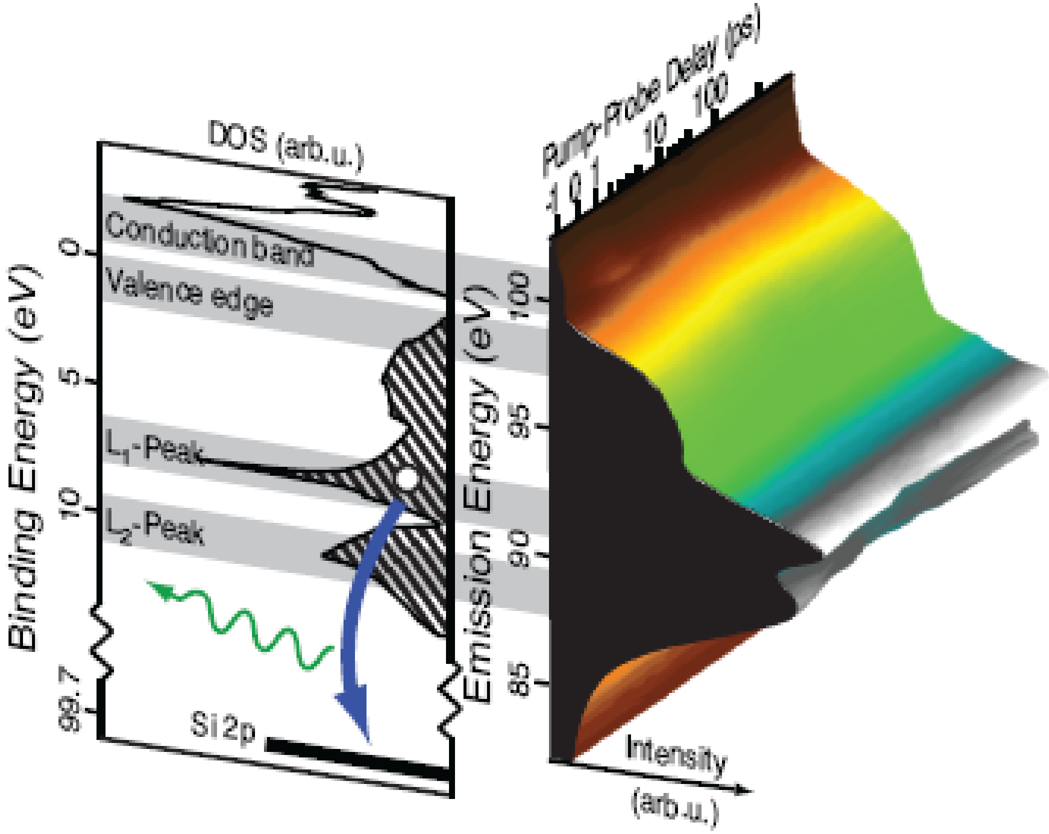
Time-resolved electronic structure maps



electronic states A, B, C

Dynamics of highly photoexcited silicon





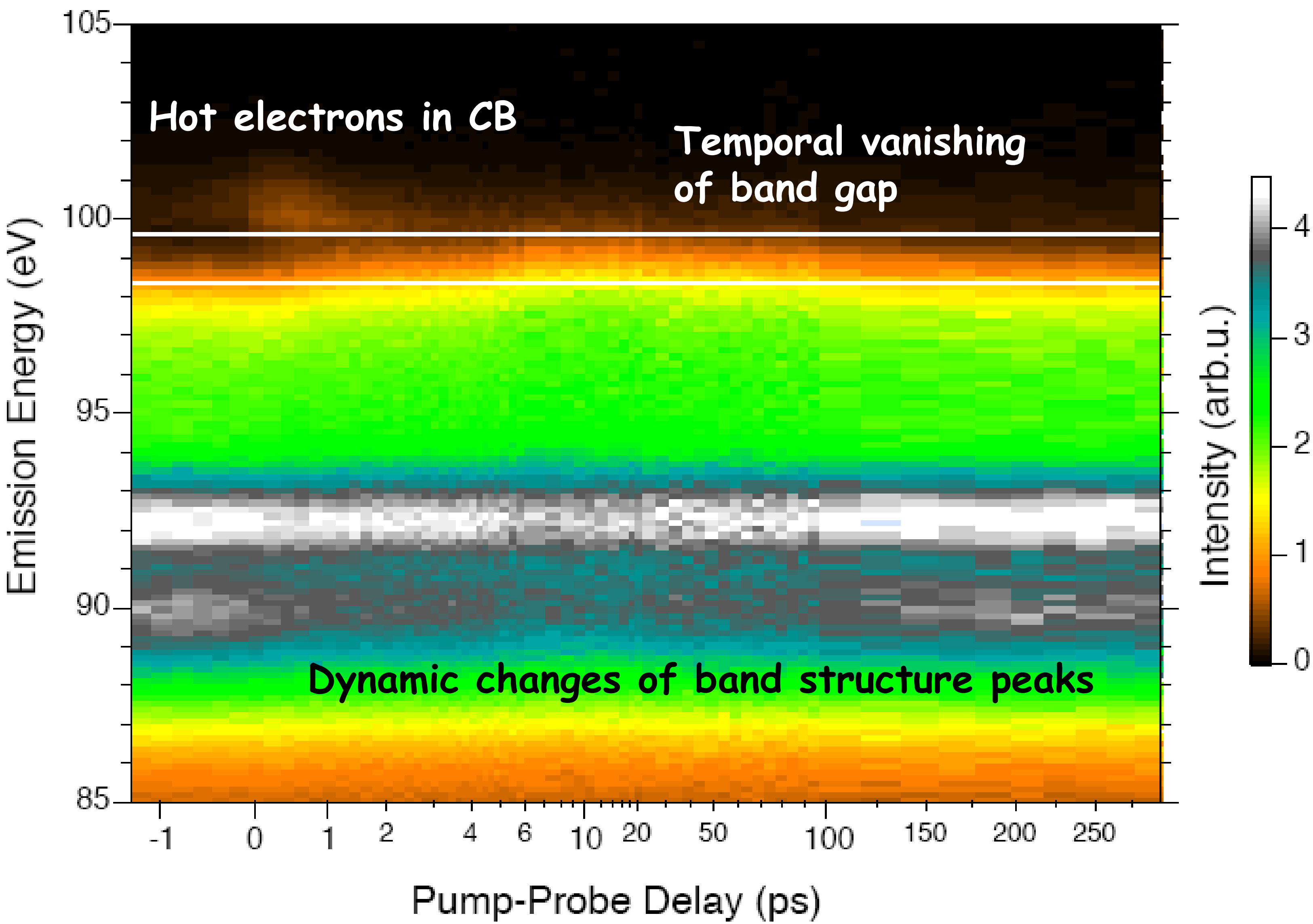
FLASH:

- 117eV Photons
- 30 bunches@250kHz
- every 200ms
- around 40µJ per pulse
- 30fs pulse length
- attenuated
- ~80mJ/cm^2

Facility's Ti:Sa LASER:

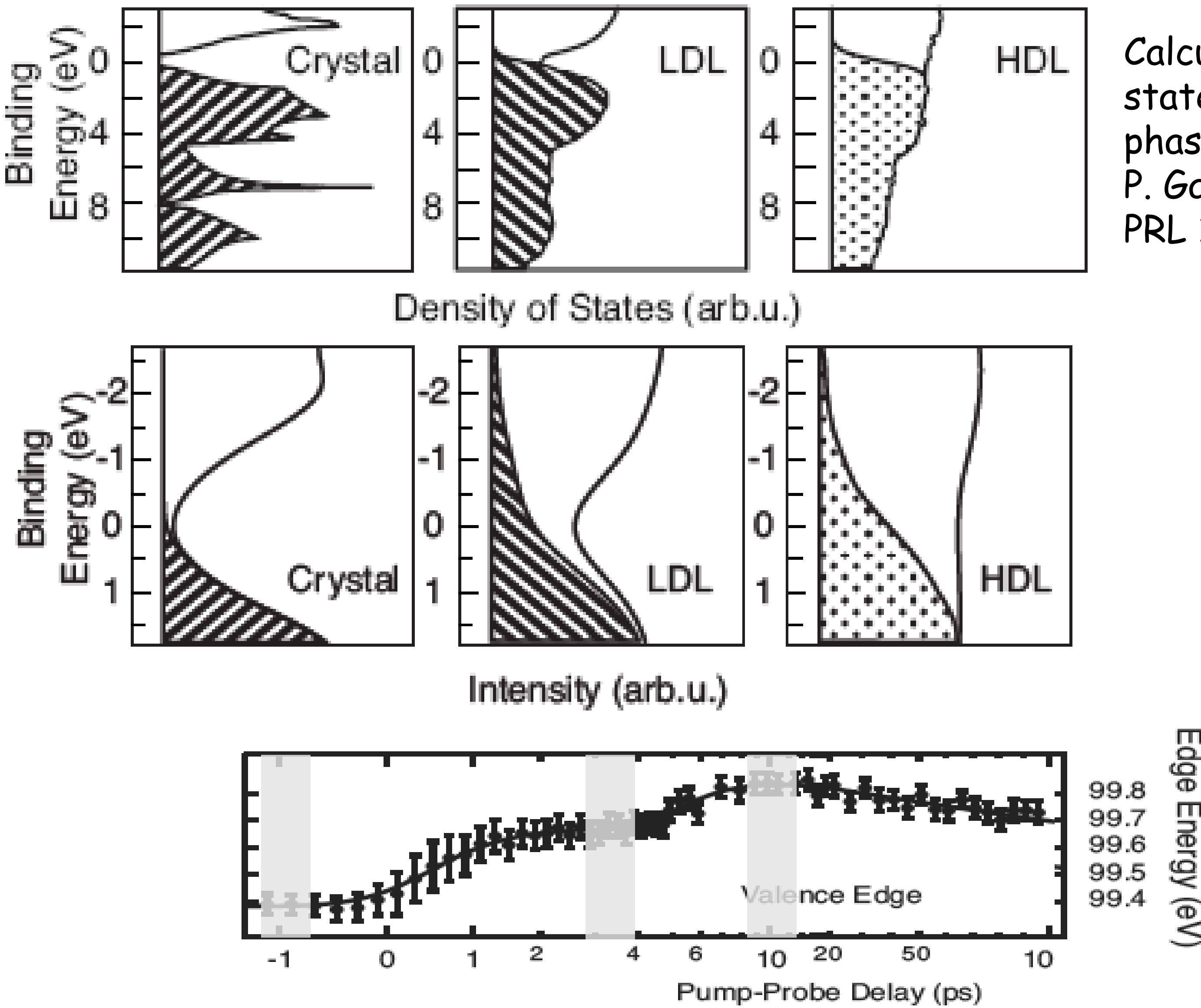
- 400nm
- same time structure
- 260mJ/cm^2 on sample
- 120fs pulse length
- 10^{22}/cm^3 excitation density

Evolution of electronic structure after strong photoexcitation



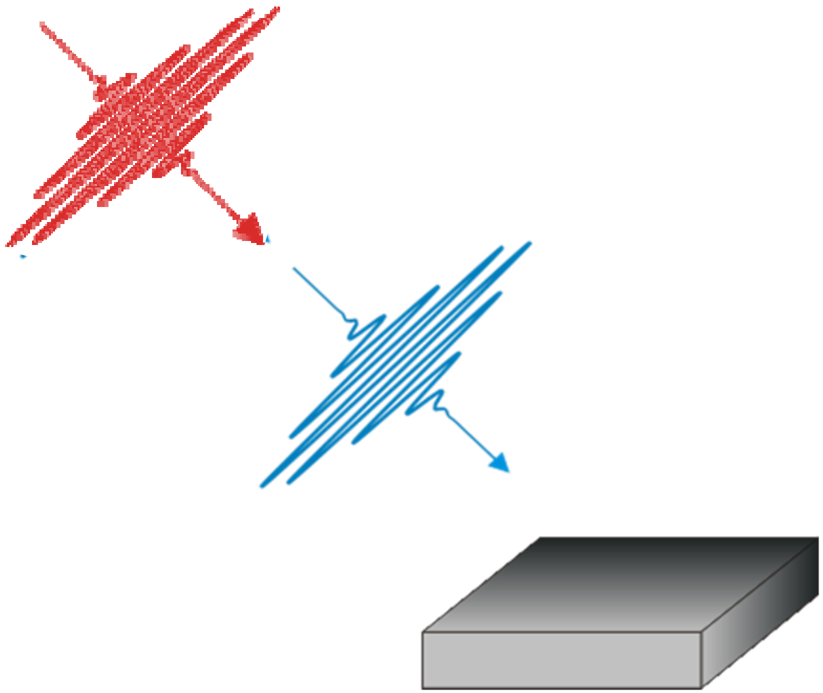
M. Beye et al., PNAS published online August 30, 2010, doi:10.1073/pnas.1006499107

Conclusion: Liquid-liquid transition in Si



Calculated density of states for different phases of silicon
P. Ganesh and M. Widom, PRL 102, 075701 (2009)

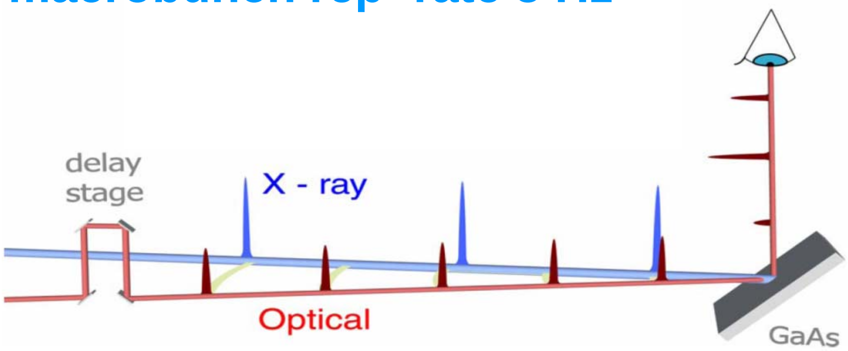
XUV-pump Optical-probe



XUV induced transient reflectivity change in GaAs

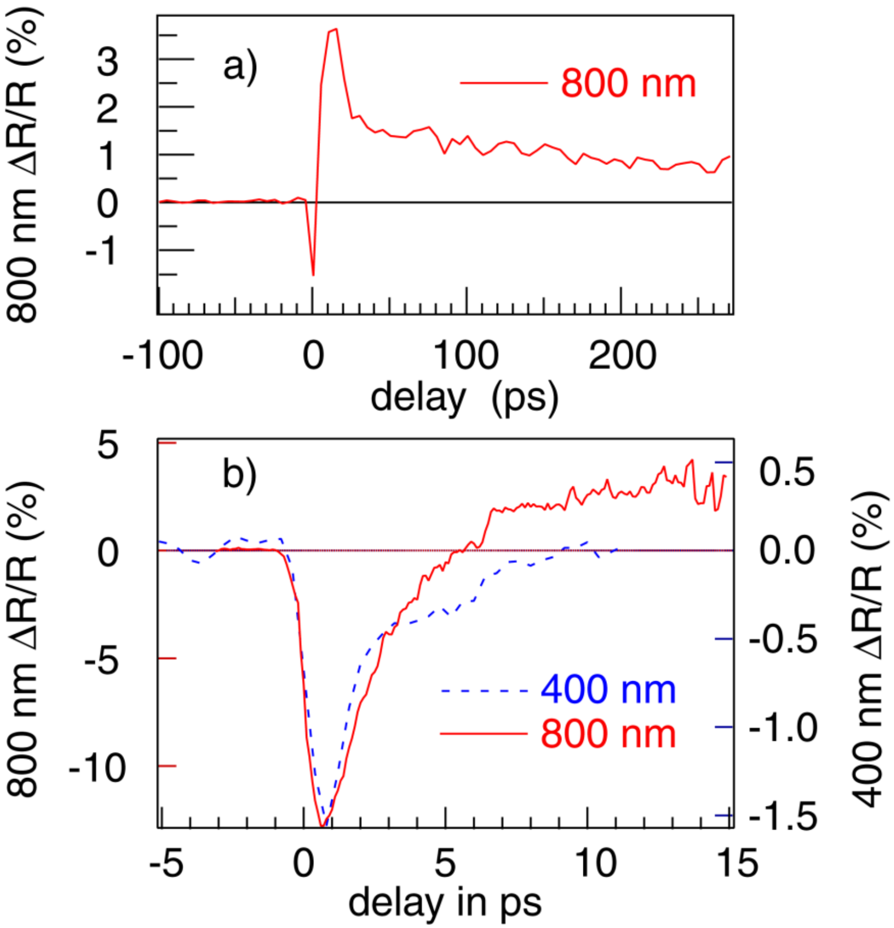
Experiment

FLASH: 39.5 eV
< 25fs, <16µJ/pulse
30 pulses/macrobunch @ 500 kHz
macrobunch rep- rate 5 Hz



Optical Laser: 800 nm or 400 nm,
~ 120fs, < 10 nJ,
60 pulses/macrobunch @ 1MHz

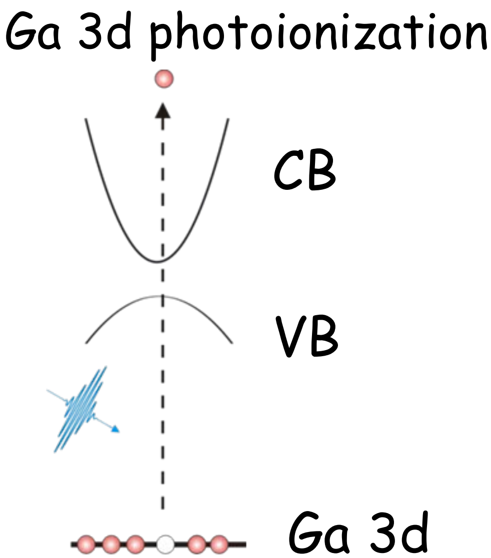
C. Gahl et al. Nature Photonics 2, 165 (2008)
T. Maltezopoulos et al., NJP 10 (2008) 033026



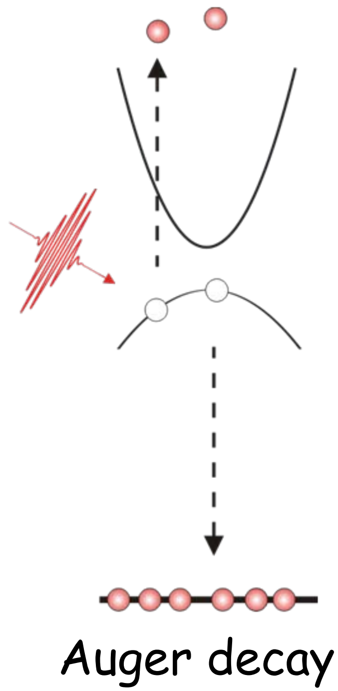
Quasi-instantaneous drop in optical reflectivity !

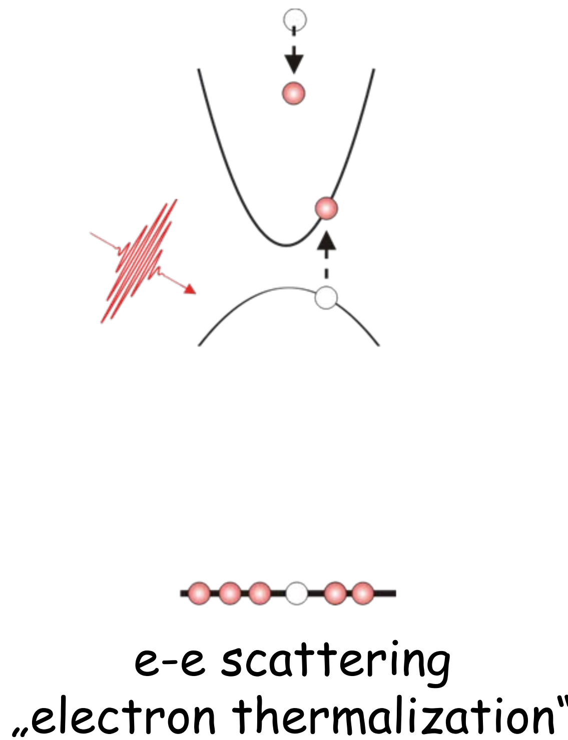
Model: ultrafast electron relaxation

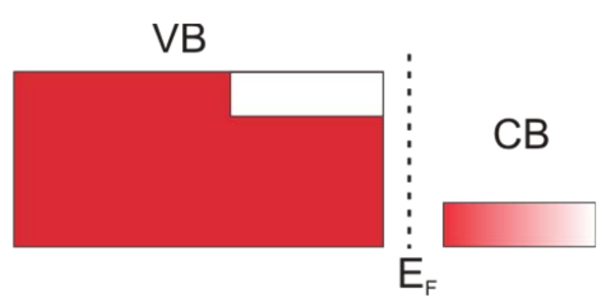
Pump



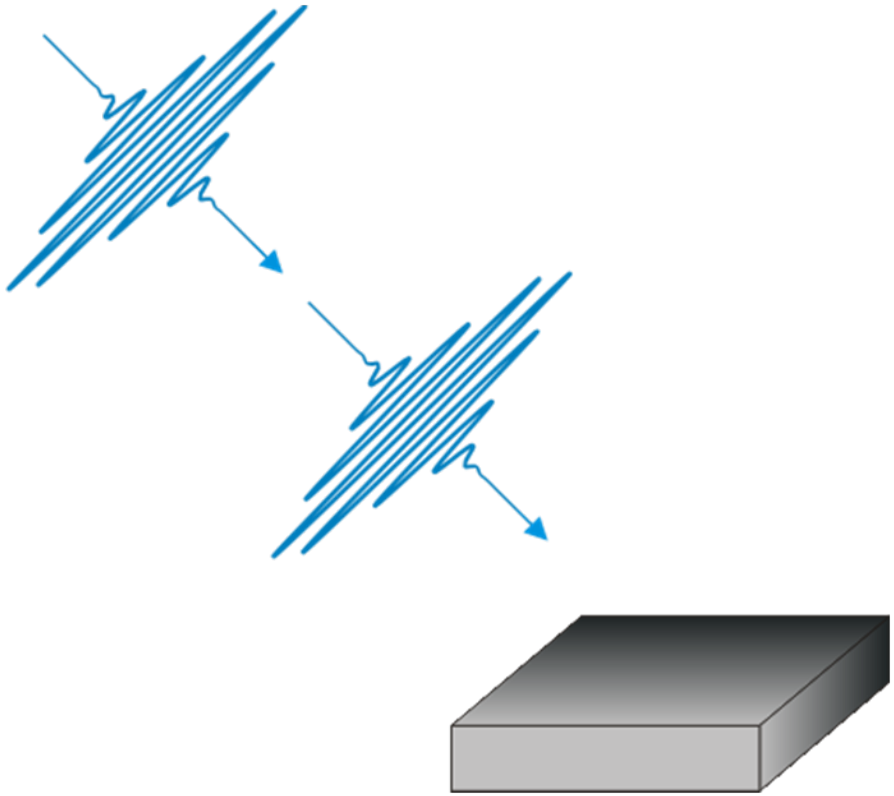
Probe







XUV-pump XUV-probe

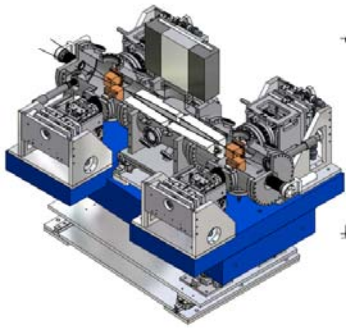


XUV split and delay unit

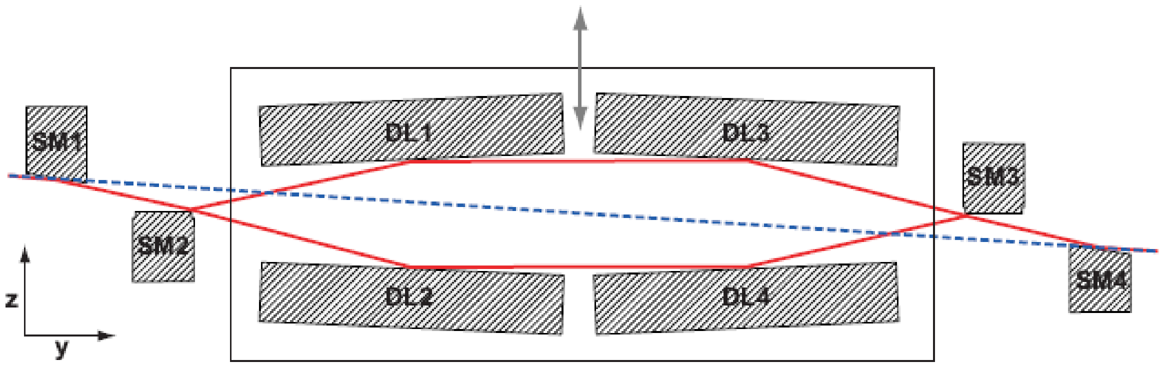
Molecular wave packet dynamics – N_2

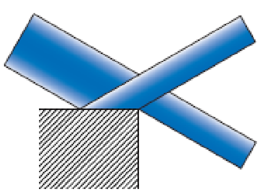
Hot electron dynamics in Si

Source properties- Interferometry

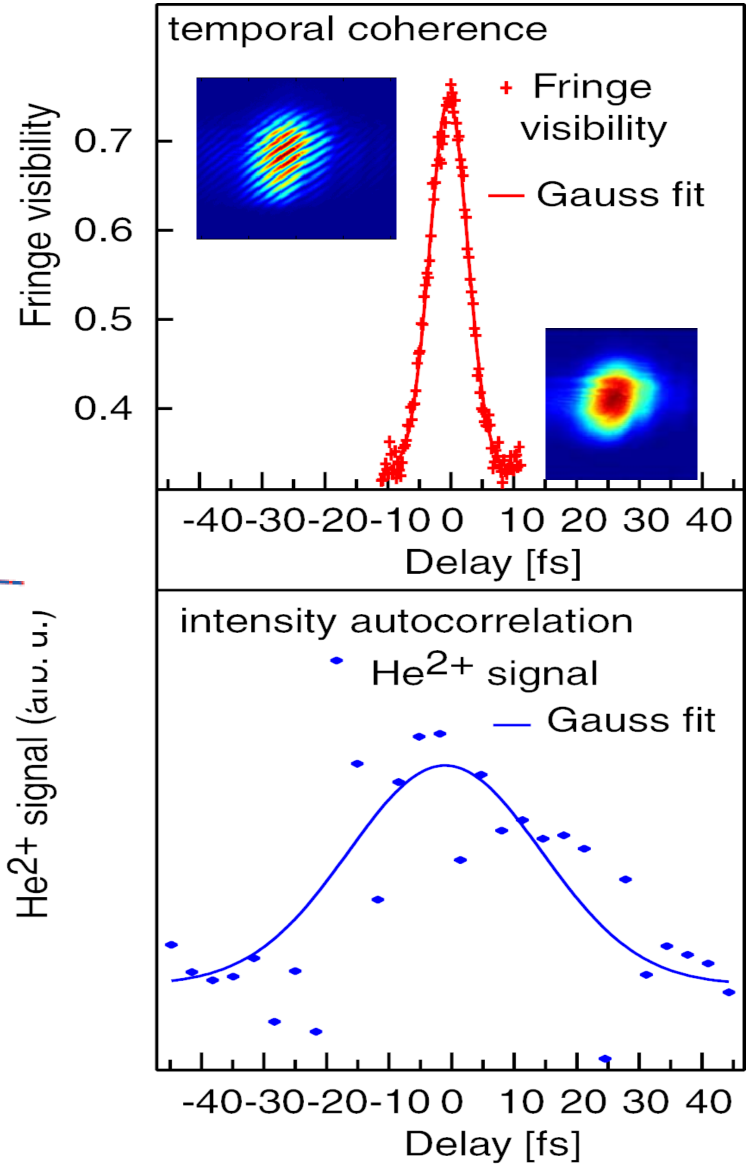


Mach-Zehnder-type XUV interferometer



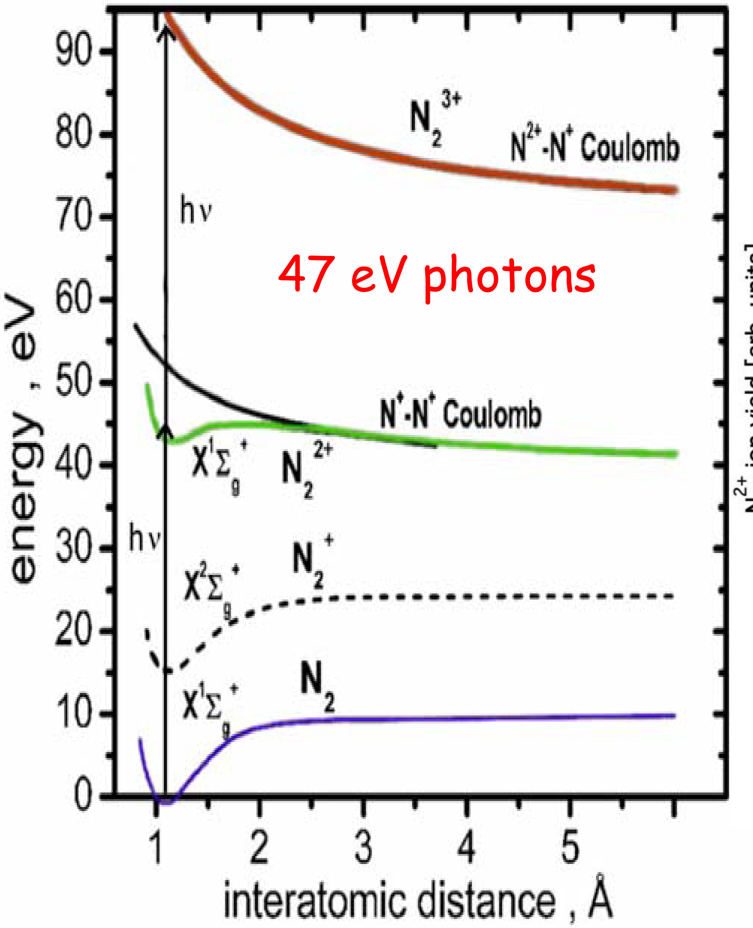


F. Sorgenfrei et al., RSI 81, 043107 (2010)
W. F Schlotter et al. Optics Letters 35, 372 (2010)

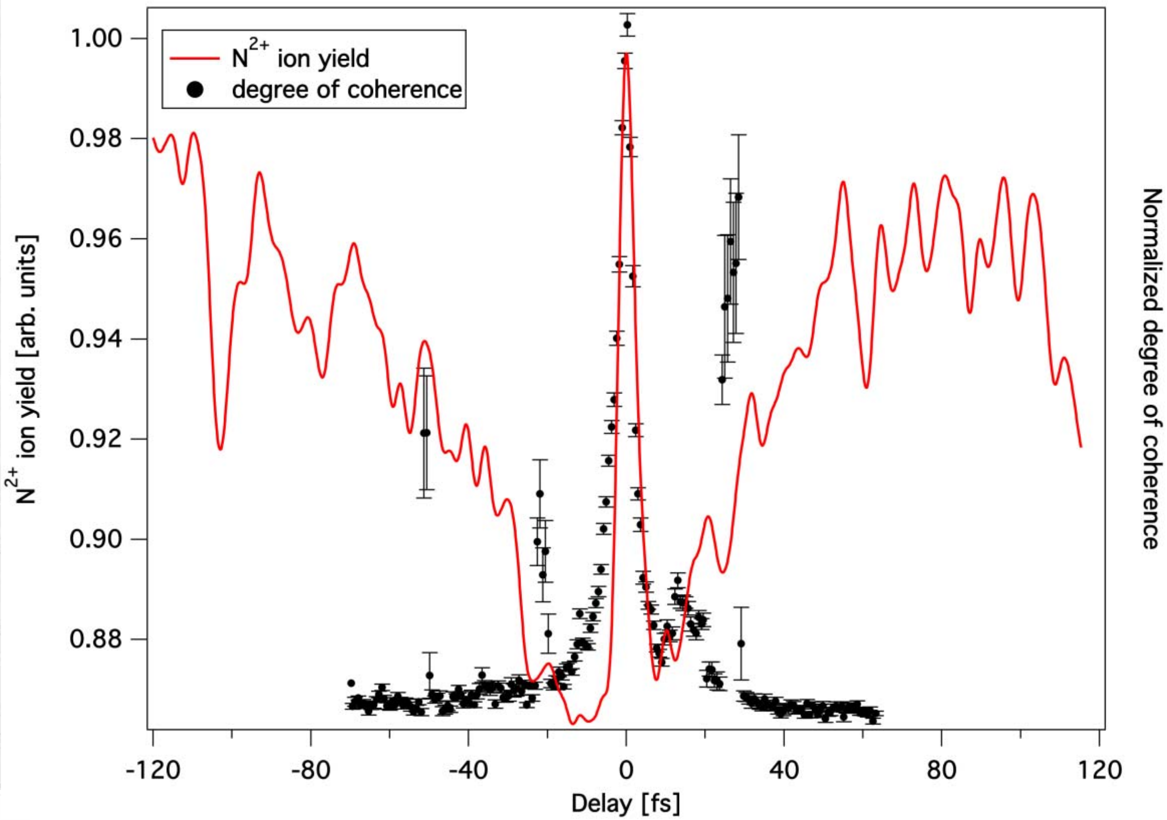


See also R. Mitzner et al. Optics Express 16, 19909 (2008), PRA 80, 025402 (2009)

N_2 wave packet dynamics



Potential energy curves N_2 (Franceschi et al. J. Chem. Phys. (2007))



Oscillations of the N^{2+} ion yield

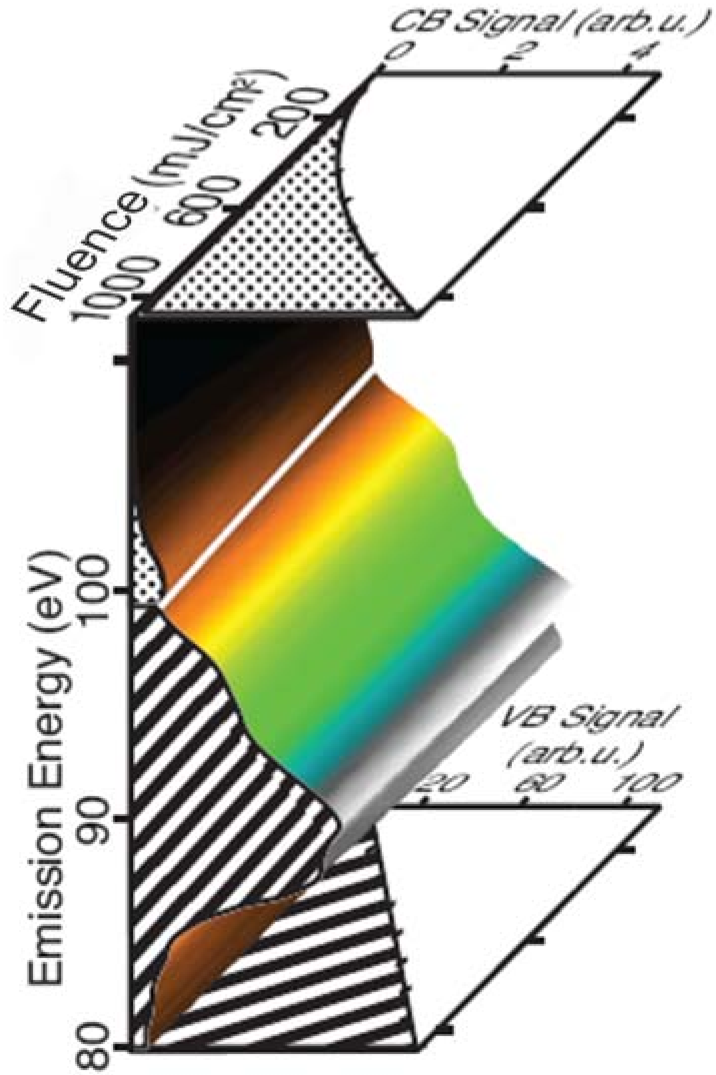
„Coherence peak" ??

F. Sorgenfrei et al. to be published

XUV induced charge carriers in Si
Fluence dependence

FLASH:

- undoped Si(100) sample
- 117eV Photons
- 30 bunches@250kHz
- every 200ms
- 30fs pulse length
- 8fs core hole lifetime
- $(50\mu m)^2$ spot

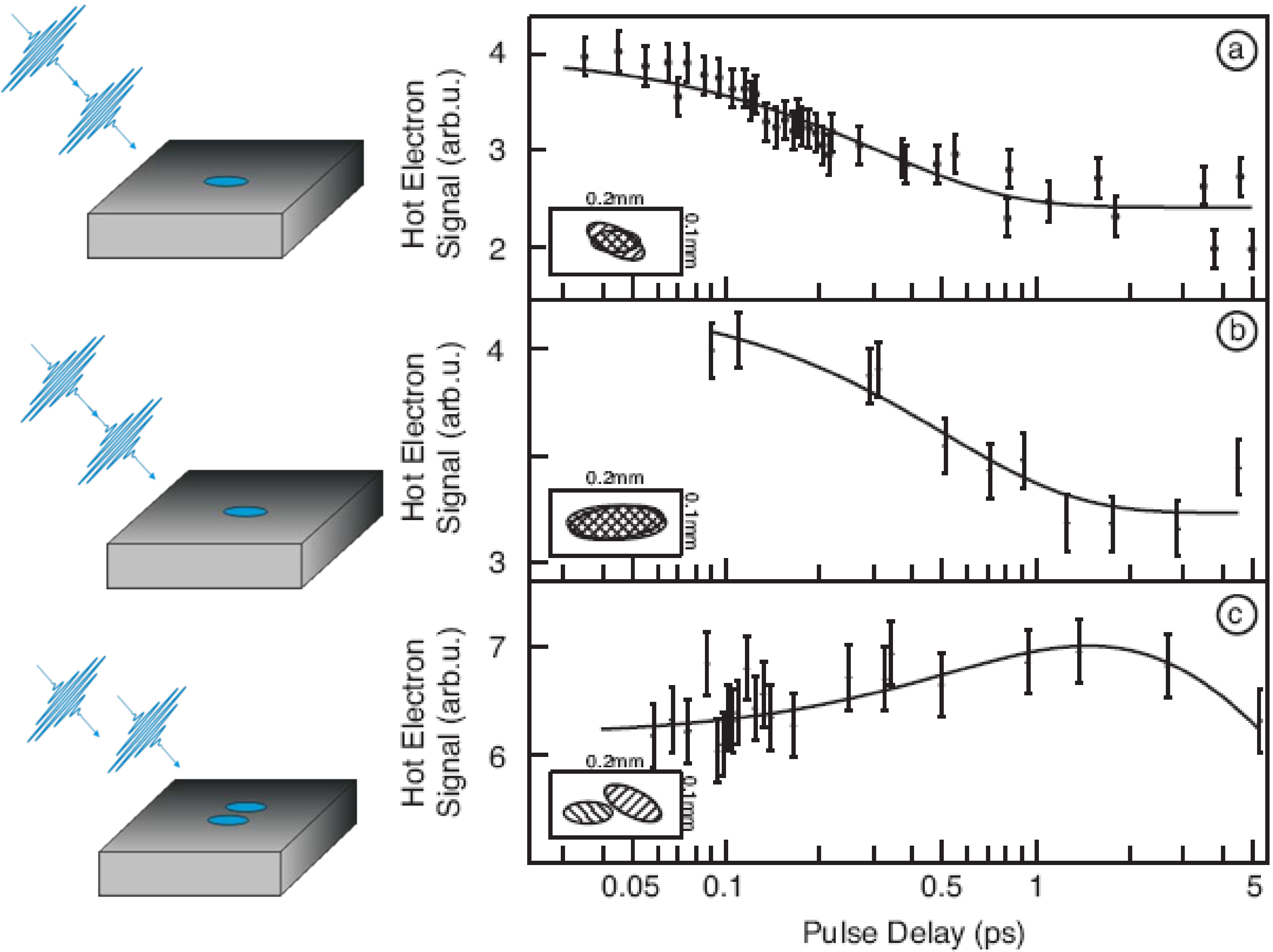


$CB \propto (Int)^{1.7}$

$VB \propto (Int)^{1}$

M. Beye et al. submitted

Hot electron dynamics



spot: 40μm x 85μm
τ~(310±50) fs

spot: 50μm x 150μm
τ~(500±150) fs

separation: 70μm
peak~1400 fs

M. Beye et al. submitted

Model: surface confined diffusion

- Fast creation of carriers through Auger decay (8fs) and inelastic e-e scattering

- Subsequent diffusion of hot carriers out of probing volume (probing depth ~20nm)

- Layer of charge carriers confined to surface

- extremely fast motion in high fields

Perspectives

Pump / Probe	IR or Optical	XUV- Soft X-rays
IR or Optical	Dynamics of low energy excitations (e.g. carrier dynamics, nuclear wave packet evolution,..)	Create well defined localized excited states in complex systems
XUV- Soft X-rays	Photo induced changes in electronic structure (e.g. photo switching, photo- and electrochemistry, magnetization dynamics,…..)	Nonlinear Processes (SHG, Resonant Raman, four-wave mixing…..)

shorter timescales are possible
☞ Sub - femtosecond pump - probe

Bright future for powerful short pulse soft x-ray sources !