Non-equilibrium energy transfer dynamics by ultrafast solid state spectroscopy

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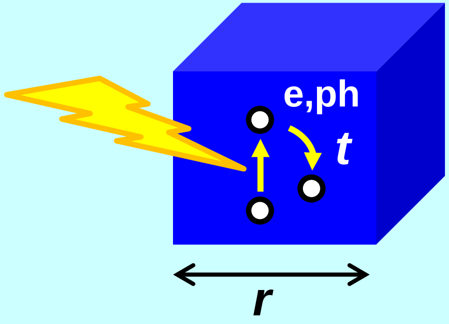
Univ. Duisburg-Essen, Germany

ECRYS
August 9, 2022

r < interaction length
local relaxation
isolated excitations

Project Area A



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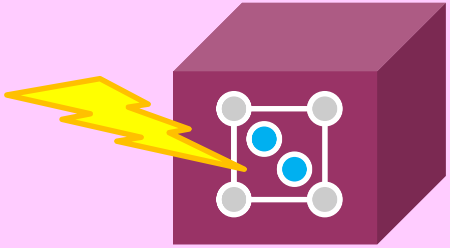
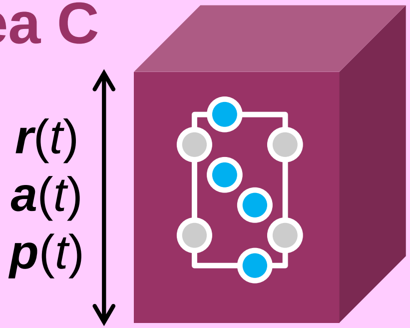
Non-equilibrium dynamics of condensed matter in the time domain

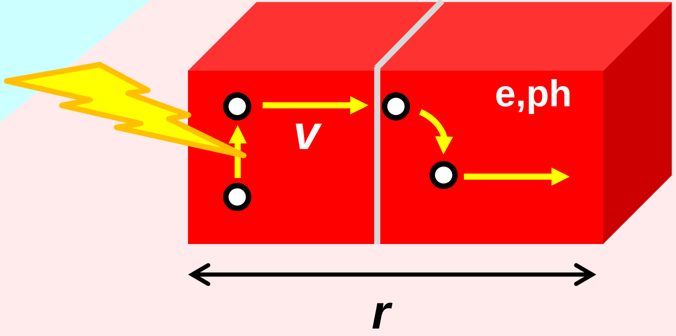
www.sfb1242.de



structural / cooperative dynamics

Project Area C



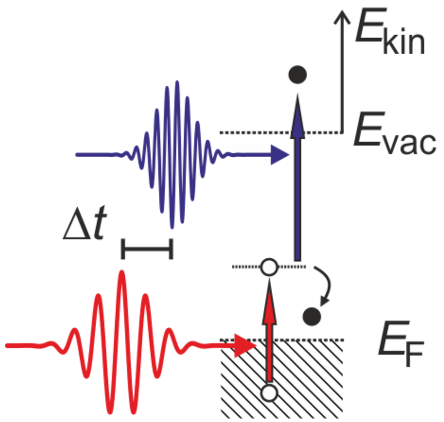


r > interaction length
propagation effects
extended excitations

Project Area B

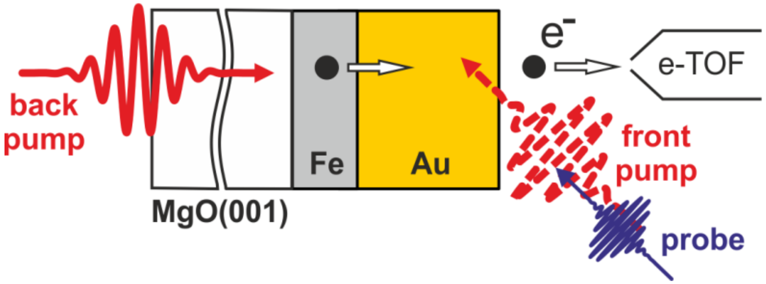
Electron Dynamics at Buried Media and Interfaces: Au/Fe/MgO(001)

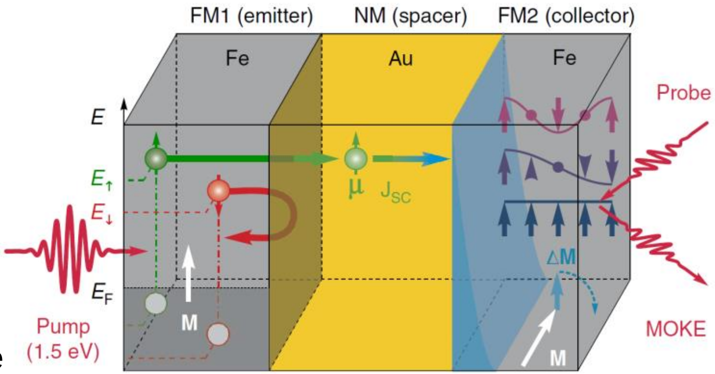
Y. Beyazit, J. Beckord, J. P. Meyburg, F. Kühne, D. Diesing, P. Zhou, M. Ligges
University of Duisburg-Essen, Germany



front pump / front probe

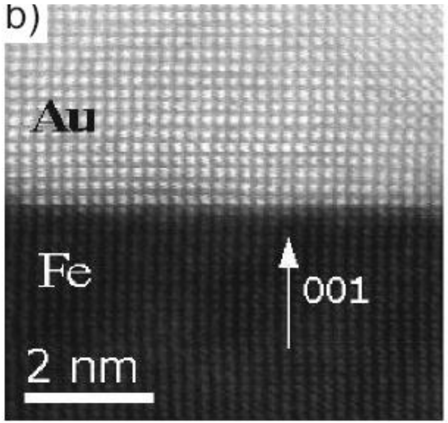
here compared to
back side pump / front side probe





Alekhin et al.,
Phys. Rev. Lett. **119**, 017202 (2017)

Razdolski et al.,
Nature Commun. **8**, 15008 (2016)

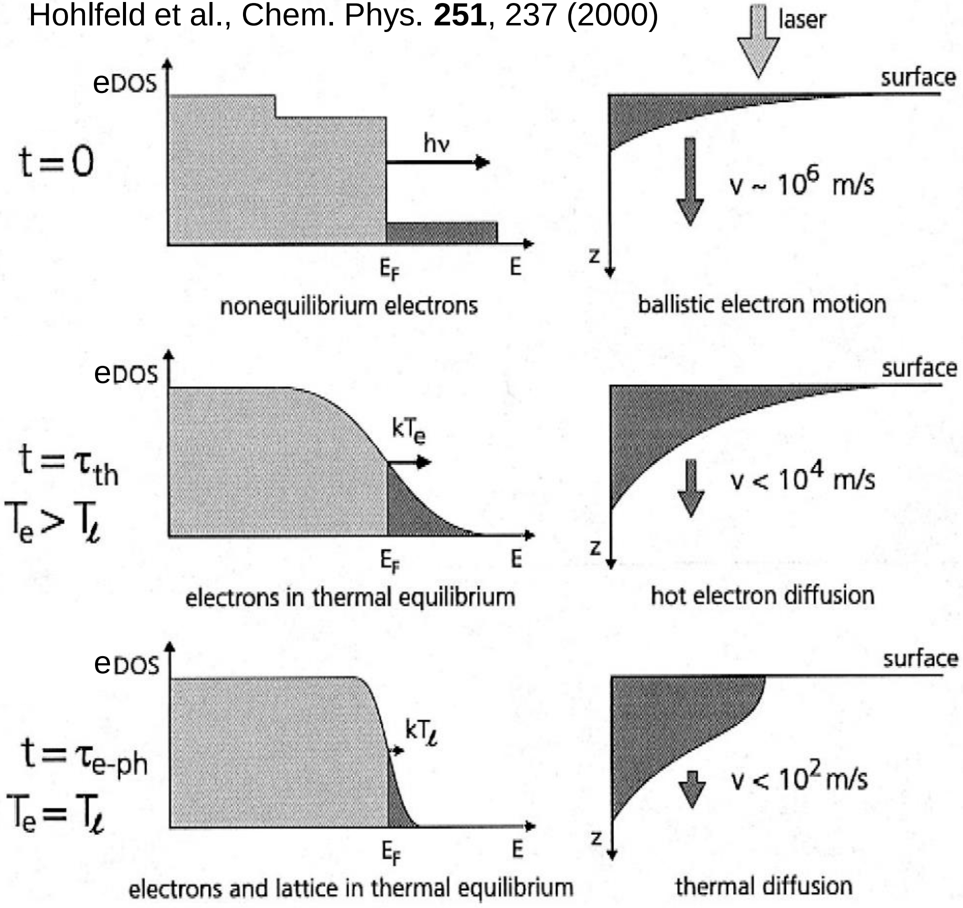


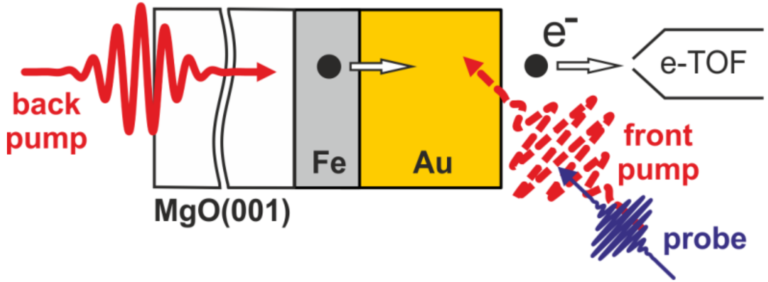
Au/Fe/MgO(100)

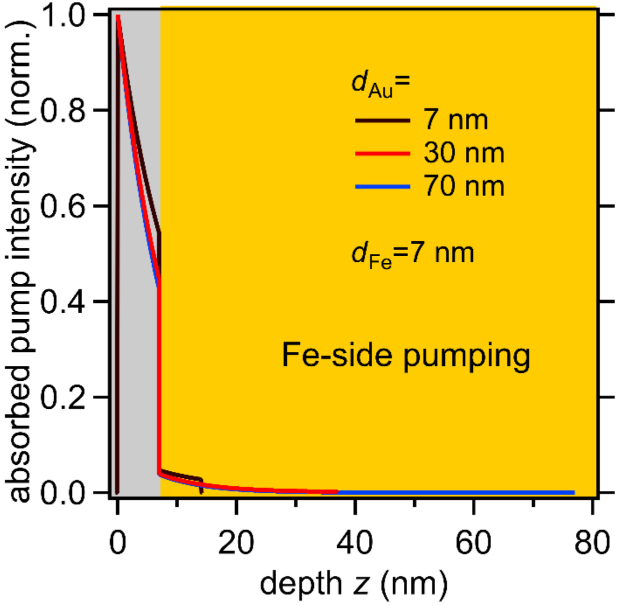
Melnikov et al.,
Phys. Rev. Lett. **107**, 076601 (2011)

Driving transport processes by Optical Pumping

Hohlfeld et al., Chem. Phys. **251**, 237 (2000)

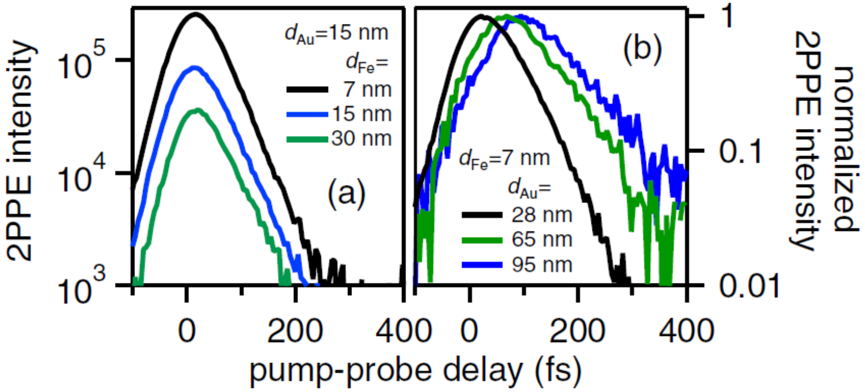


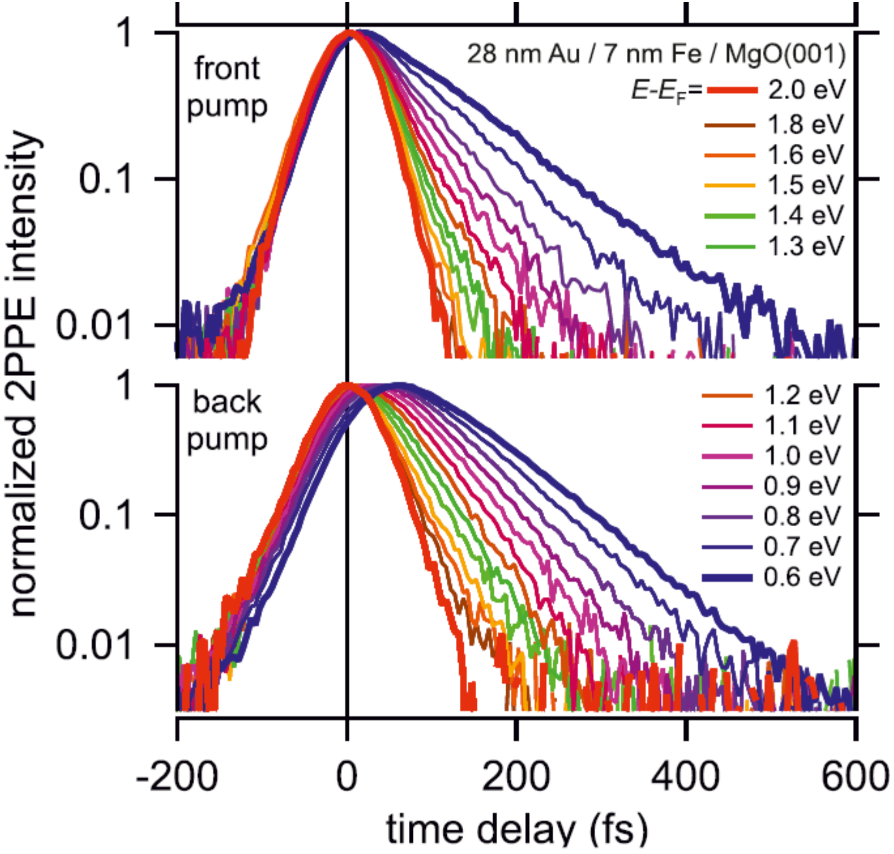




Comparison of front and back side pumping in 2PPE

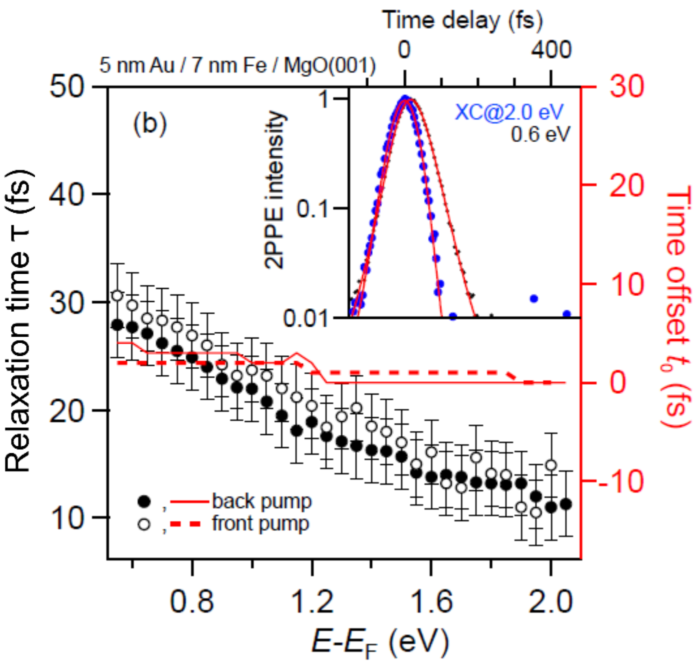
- qualitatively comparable data for front vs. back pumping
- delayed relaxation due to transport
- single exponential relaxation dynamics

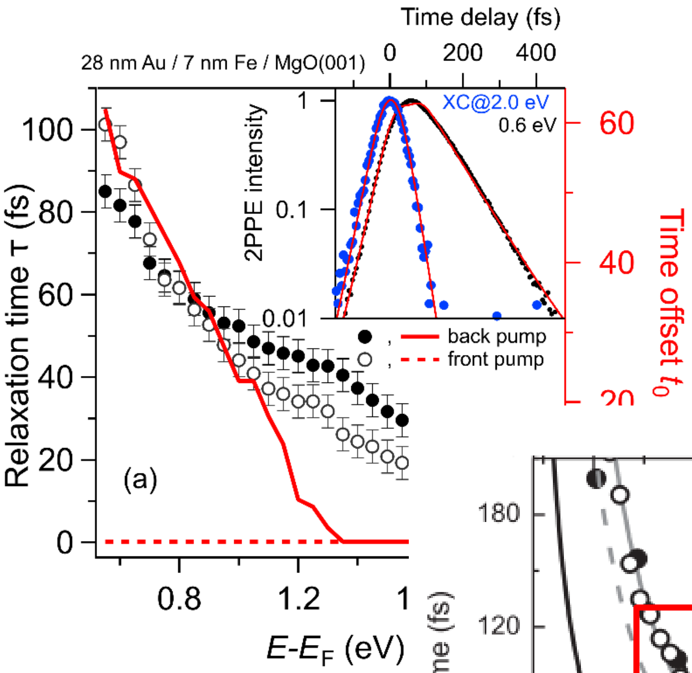




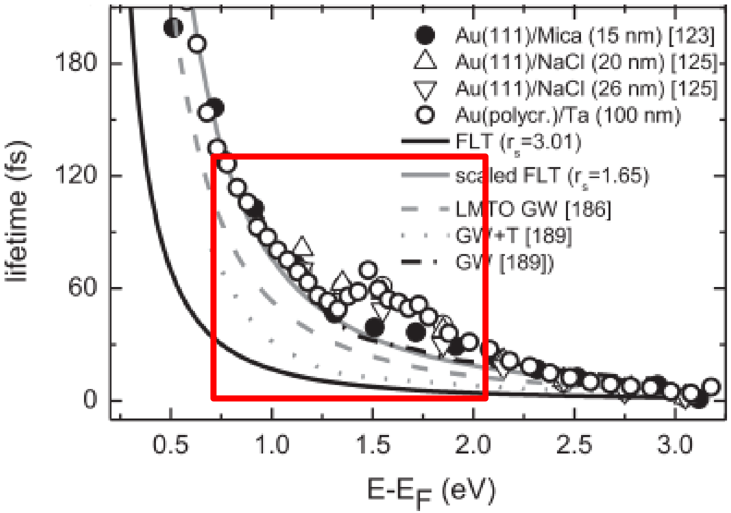
Beyazit et al., Phys. Rev. Lett **125**, 076803 (2020)

Data analysis by single exponential relaxation



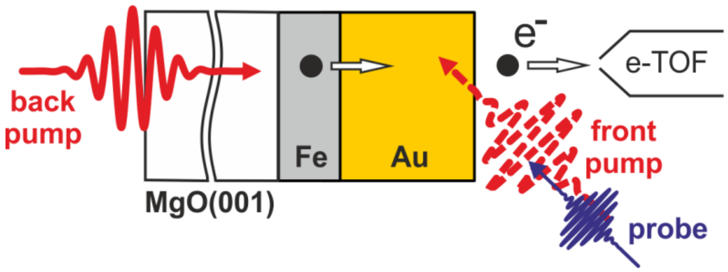


Bauer et al.,
Prog. Surf. Sci. **90**, 319 (2015)



- no / small difference in relaxation time for back vs. front pump
- importance of Au film thickness
- clear difference in time offset for d_{Au}=28 nm
- time offset ≈ relaxation time: superdiffusive regime

Separation of scattering times in Fe and Au



scattering probability τ^{-1} increases linearly with d_i

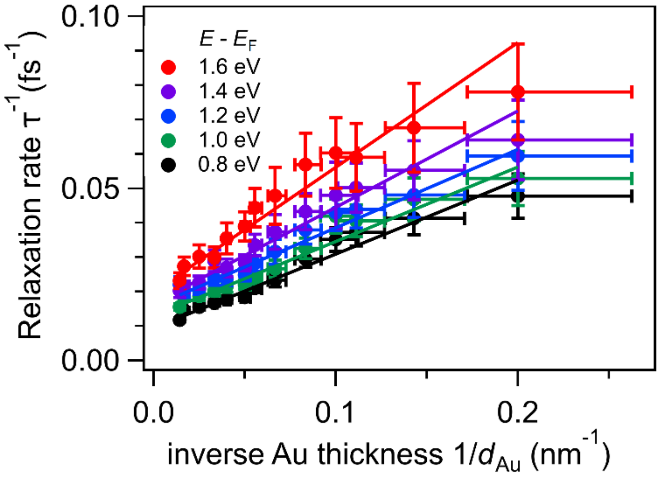
$$\int_0^{d_{Fe}+d_{Au-Fe}+d_{Au}} \frac{dz}{\tau(z)} = \frac{d_{Fe}}{\tau_{Fe}} + \frac{d_{Au-Fe}}{\tau_{Au-Fe}} + \frac{d_{Au}}{\tau_{Au}}$$

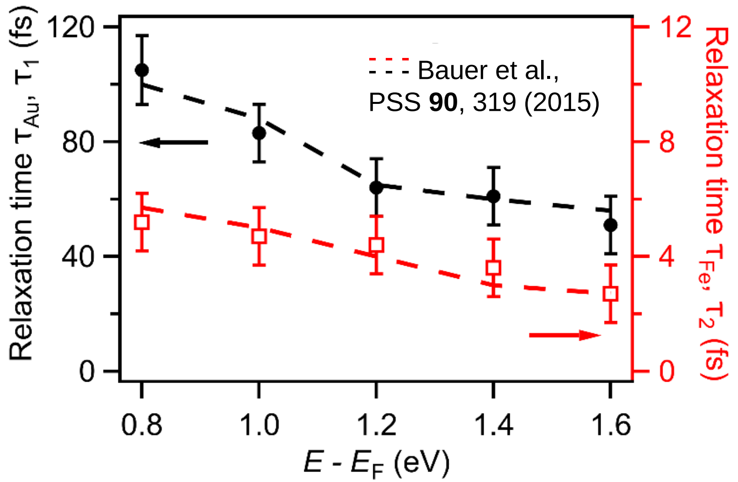
variation of d_{Au}: separation of two rates

$$\frac{1}{\tau(d_{Au})} = \qquad = A + \frac{B}{d_{Au}}$$

observation: $\tau_1 = \tau_{Au}$, $\tau_2 = \tau_{Fe}$ (bulk lifetimes of hot electrons)

ongoing work: buried interfaces with TMDC



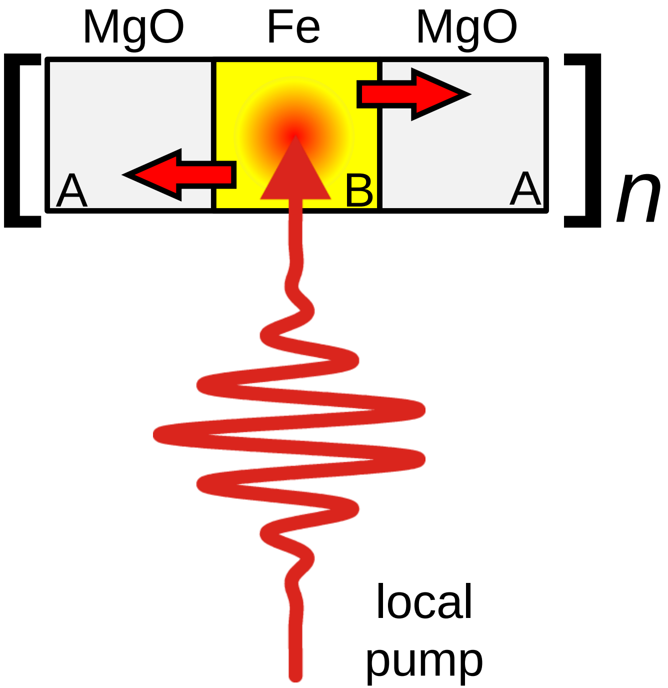


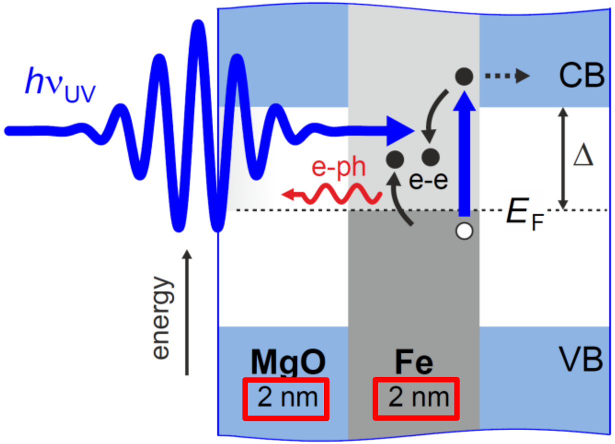
Microscopic energy flow dynamics in a simple heterostructure

N. Rothenbach, M. Gruner, K. Ollefs, S. Salamon, R. Pentcheva, P. Zhou,
H. Wende, K. Sokolowski-Tinten, A. Eschenlohr *Univ. Duisburg-Essen*

C. Schmitz-Antoniak *FZ Jülich* N. Pontius, R. Mitzner, K. Holldack, C. Schüssler, *HZ Berlin*

S. Weathersby, J. Yang, X.J. Wang *Stanford Linear Accelerator Center, USA*

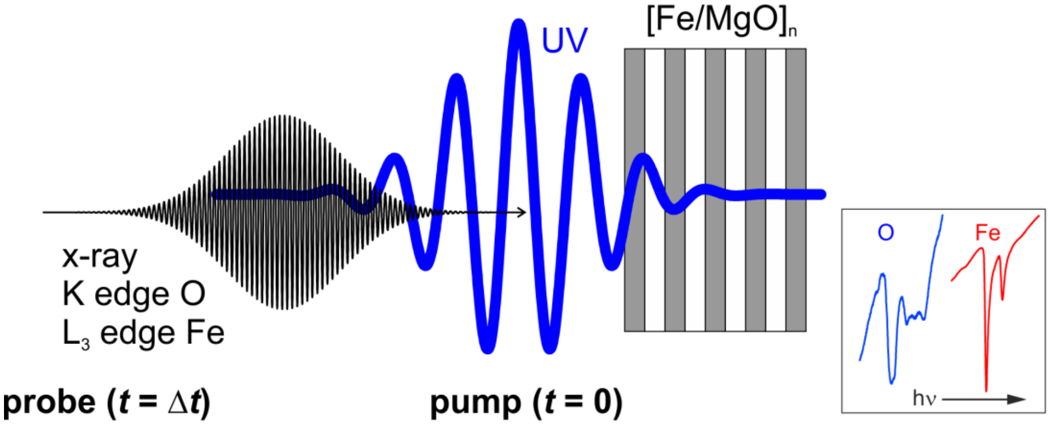


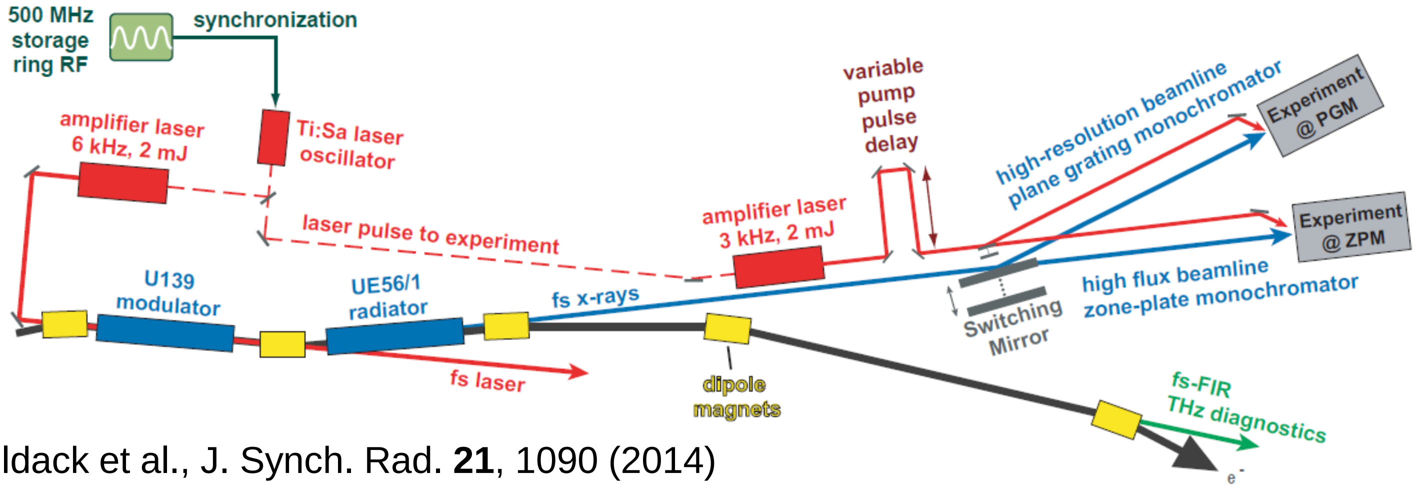


competing processes at the nanoscale

- electron transfer
- electron-electron scattering
- electron-phonon scattering

Element specific probing by soft x-ray spectroscopy



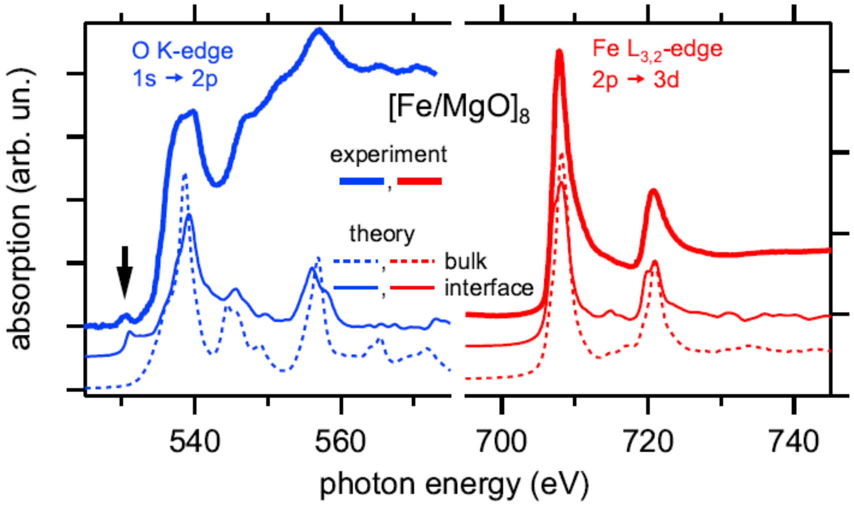


Federal Ministry of Education and Research

HZB Helmholtz Zentrum Berlin

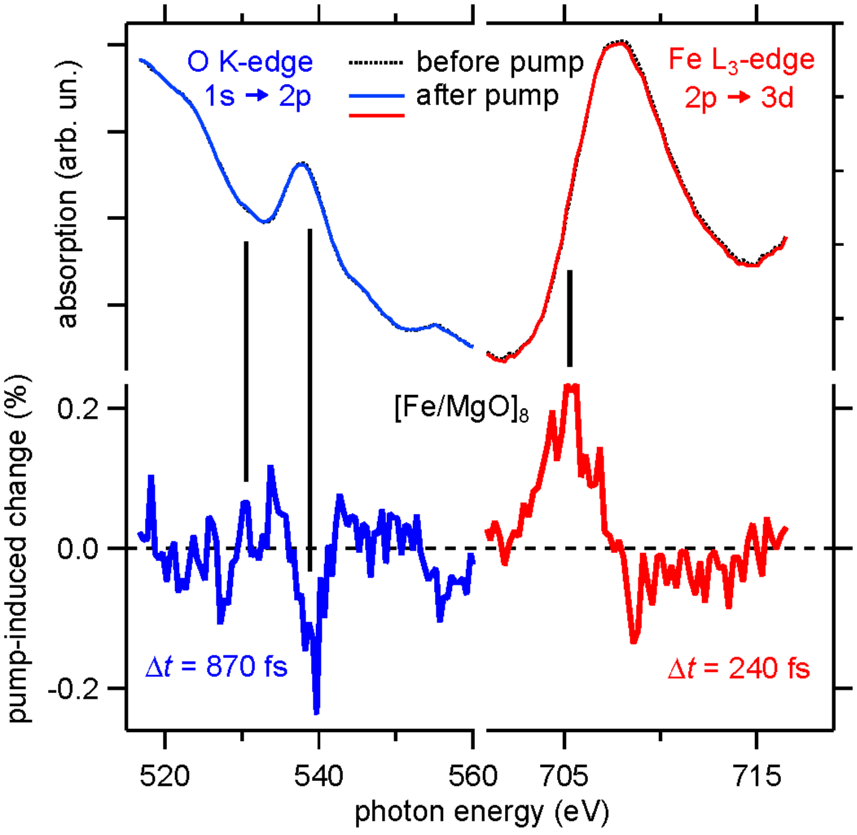
Holldack et al., J. Synch. Rad. **21**, 1090 (2014)

Near edge soft x-ray spectroscopy



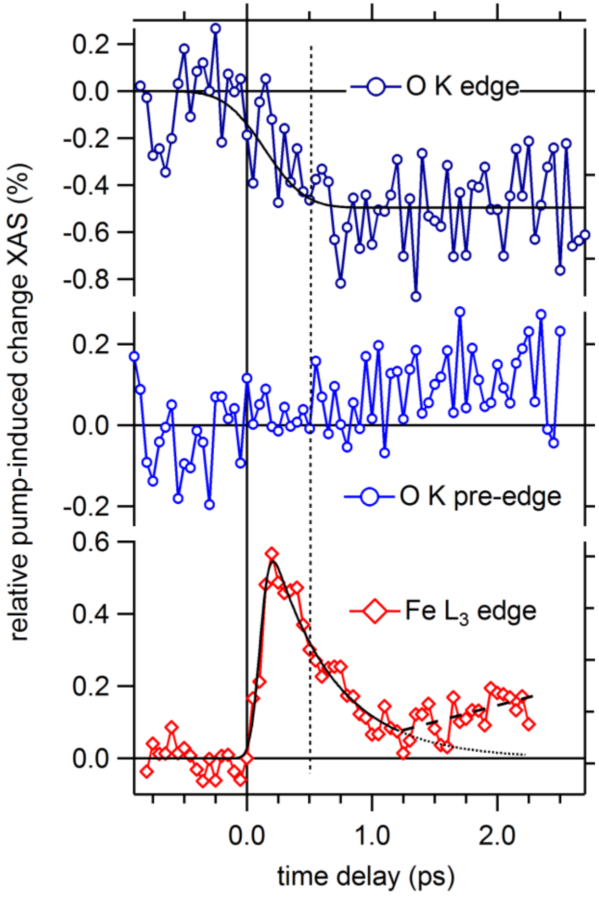
- bulk signatures of O and Fe
- feature of Fe-O interface state

- element-specific
- site-specific



maximal pump-induced change at different Δ*t* for Fe and O

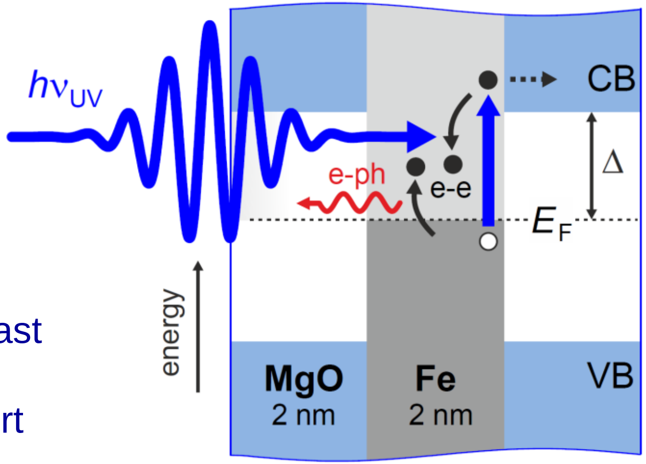
Time-resolved soft x-ray spectroscopy

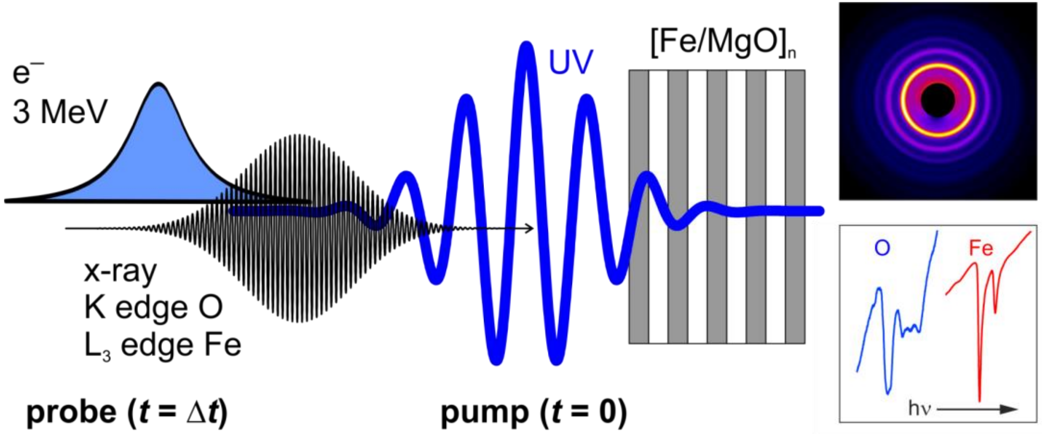


Fe L edge
ultrafast response due to electronic (<200 fs) and phononic excitations

O K edge
delayed response suggests lattice dynamics, but rather fast

hot electron lifetimes too short to generate sizable signal



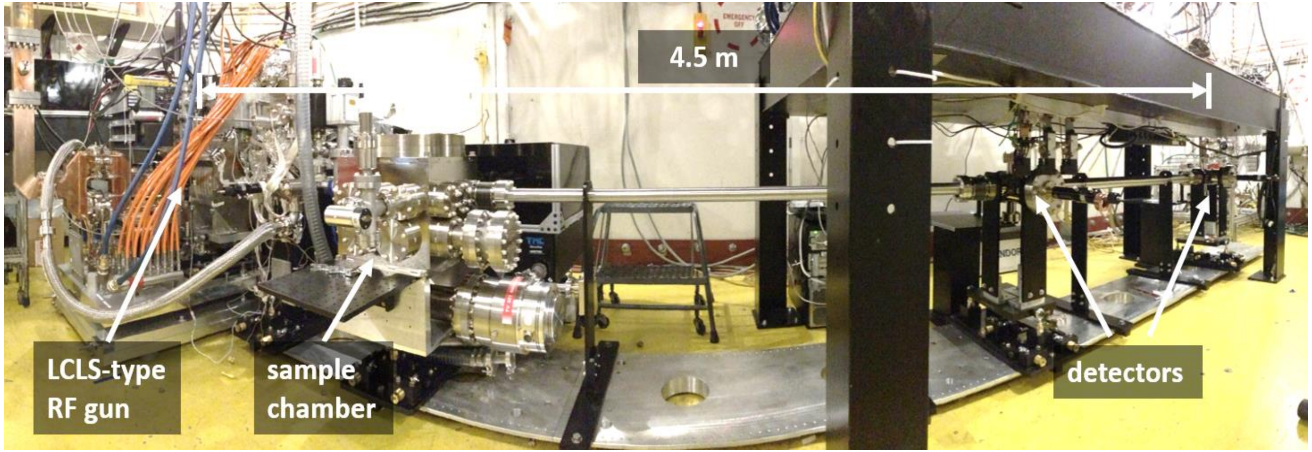


Rothenbach et al., PRB **100**, 174301 (2019)

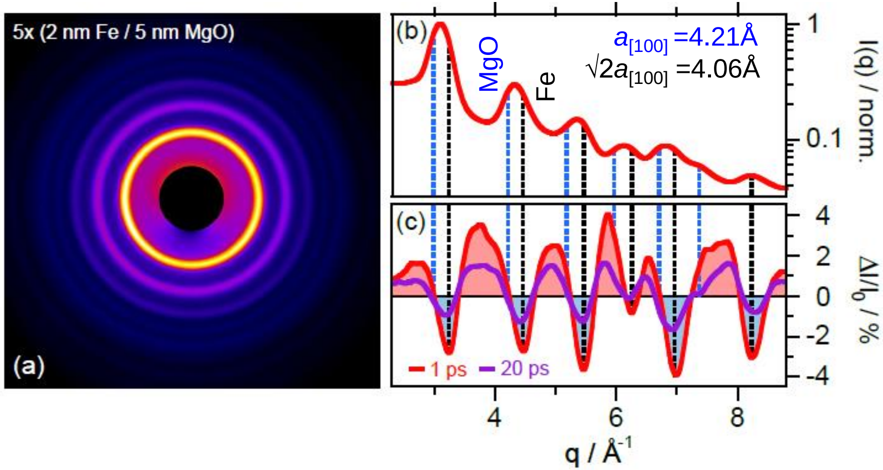
Ultrafast electron diffraction

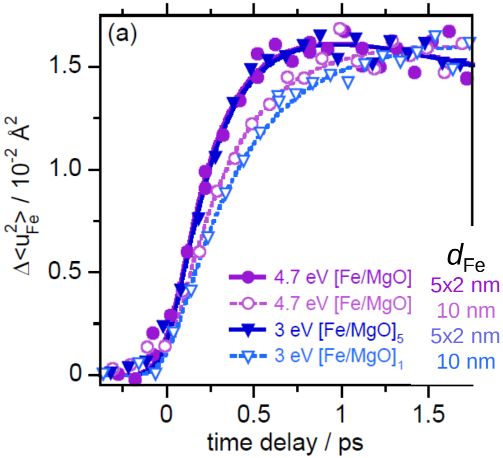
SLAC NATIONAL ACCELERATOR LABORATORY

E_{kin} = 3 - 5 MeV
pulse duration 50 - 250 fs
10^6 e^-/pulse



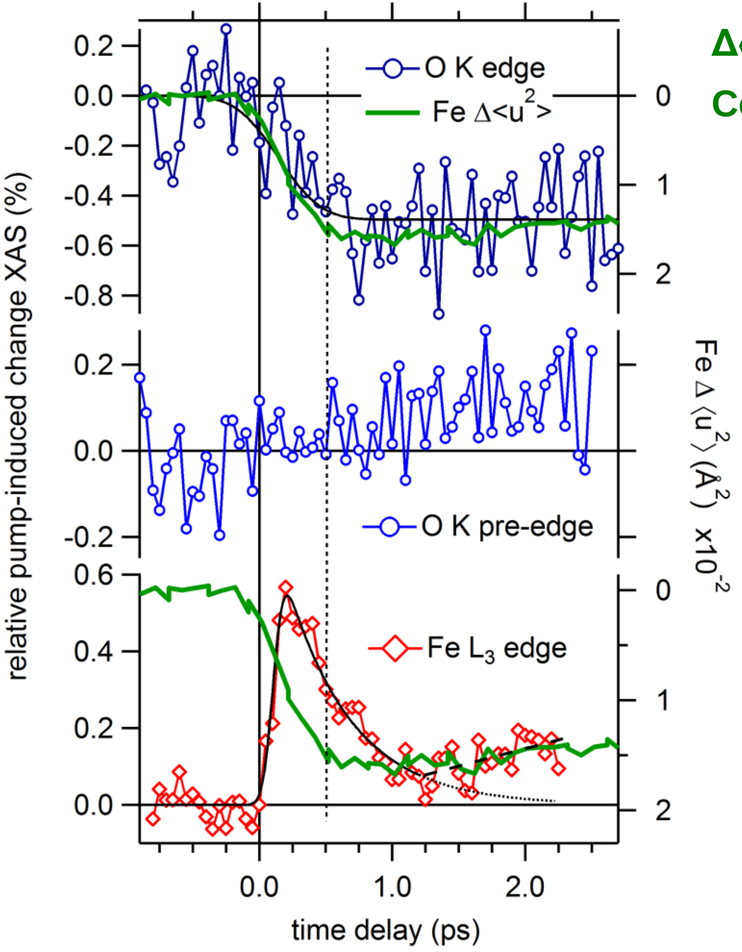
Weathersby et al., Rev. Sci. Instr. **86**, 073702 (2015)





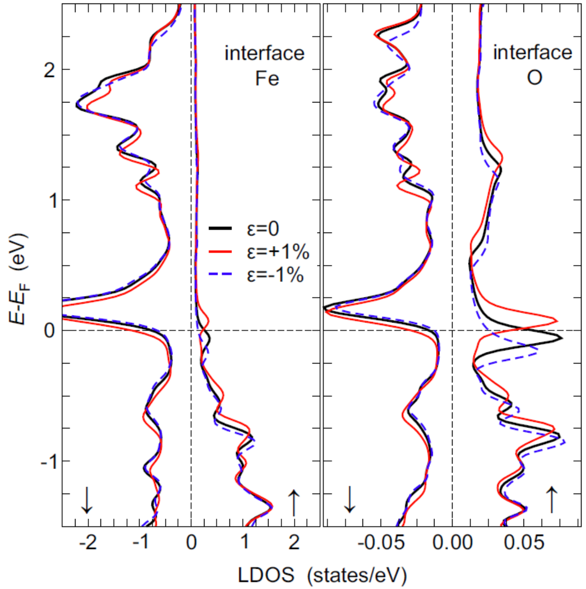
- analysis of mean square displacement $\Delta\langle u^2\rangle$ of Fe
- interface mediated energy transfer
- identical dynamics for pumping at $h\nu$ = 4.7 and 3 eV

Monitoring electronic and phononic response



Δ⟨u^2⟩ of Fe nuclei: electronic response in MgO follows phonons in Fe

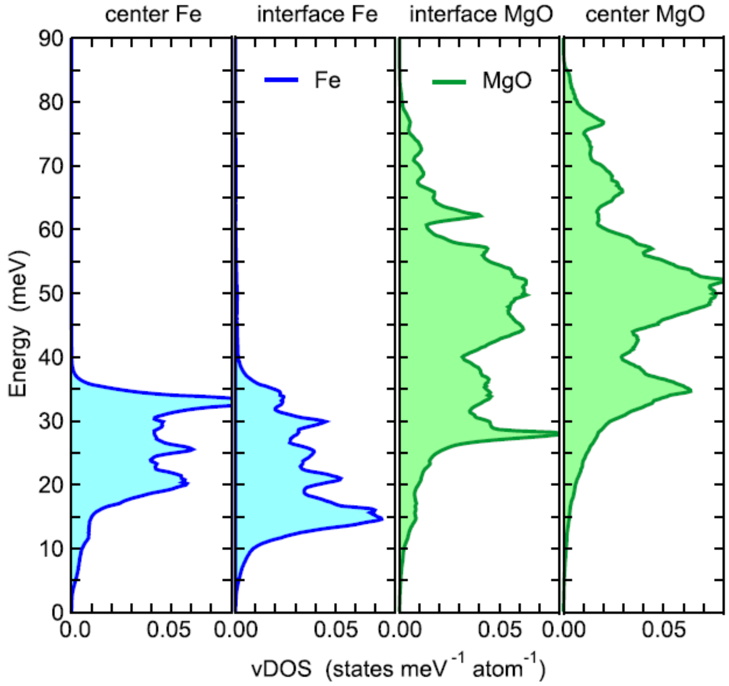
Coupling to MgO phonons: electronic Kapitza resistance



effect of lattice expansion and compression ε in eDOS

Δt > 1 ps
local lattice distortion and heterostructure thermalization

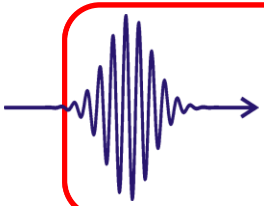
Energy transfer across interface by non-thermal phonons



time scale ~ 1 ps
transfer of excitations from Fe to MgO mediated by strong interaction of phonons at the interface at ħΩ ≥ 15 meV

non-thermally populated interface phonons

interface polaron?



hot electrons in Fe

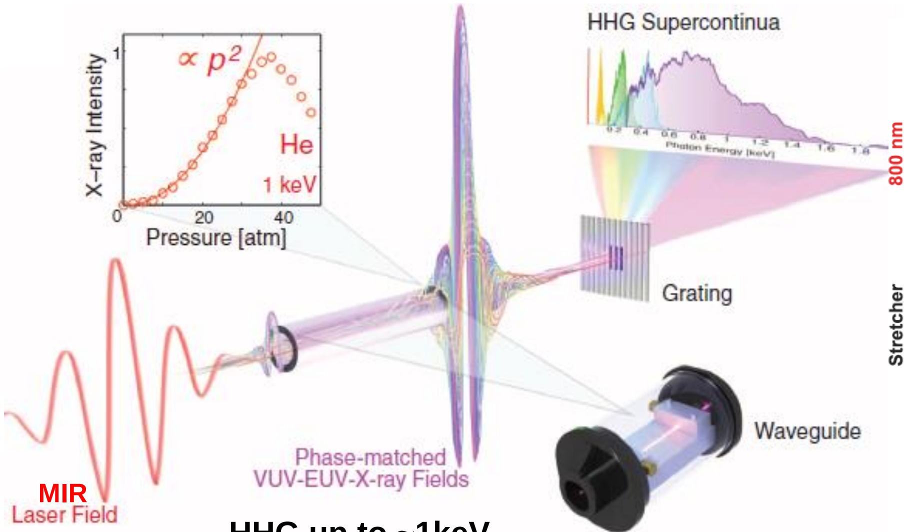
< 1 ps

hot high energy interface hybrid phonons

> 1 ps

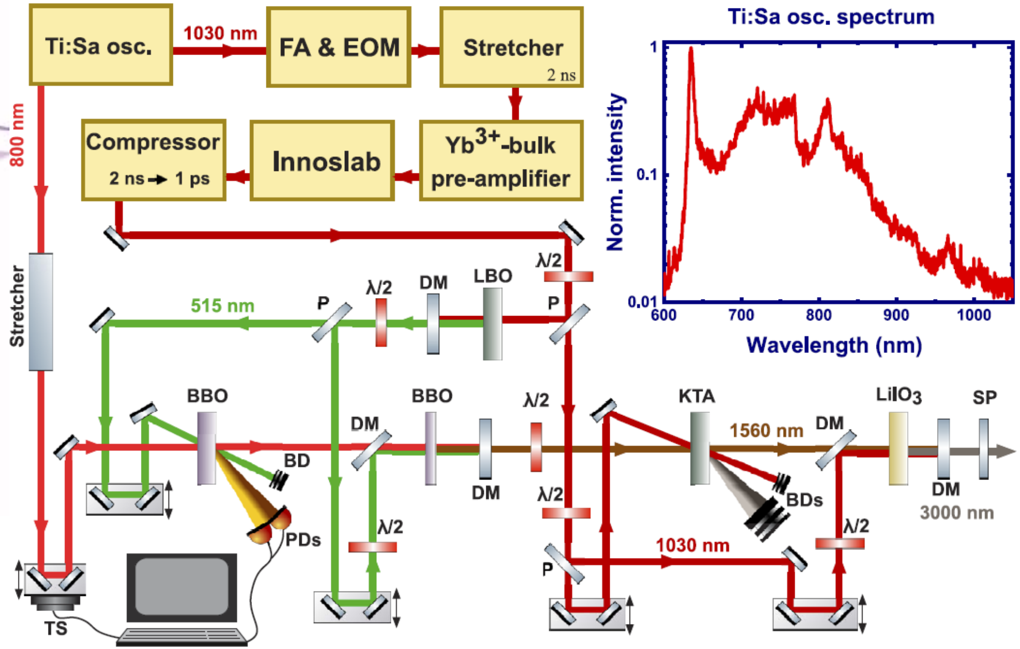
acoustic phonons, thermalization of the heterostructure

Towards table top soft x-ray pulses



HHG up to ~1keV
Popmintchev et al.
Science **336**, 1287 (2012)

OPCPA @ 1.5 and 3 µm



Bridger et al., Optics Express **27**, 31330 (2019)

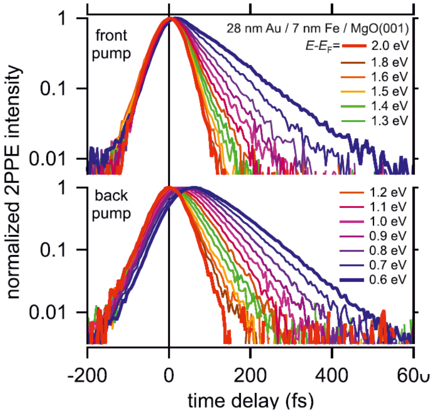
Conclusions

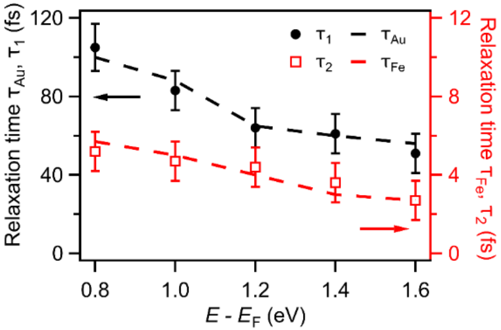
ultrafast photoelectron spectroscopy

dynamics at interfaces
analyze microscopic interactions in charge transfer and transport at complex interfaces

access to buried media
back-/ frontside excitation of hot electrons in Au/Fe/MgO(001)

separation of inelastic relaxation in Fe and Au
superdiffusive transport of hot e^- through Au

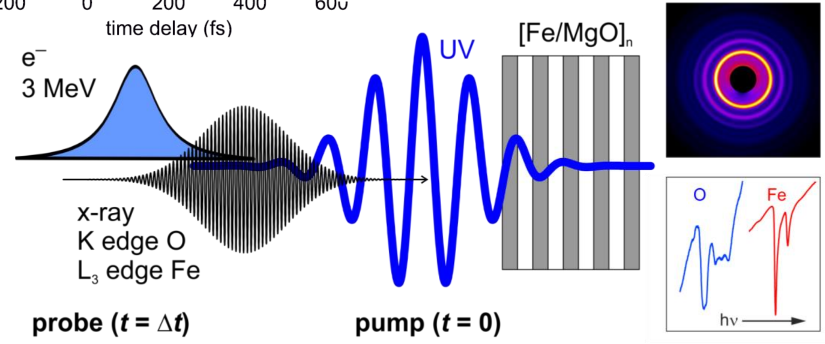




ultrafast soft x-ray spectroscopy

femtosecond slicing @ BESSY II and electron diffraction

[Fe/MgO]$_n$ energy transfer across interfaces
mediated by e-ph coupling with interface phonons



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