Ultrafast Soft-X-ray Absorption Spectroscopy

E. Seres*# & Ch. Spielmann*

*Department of Physics EP1
University of Würzburg
# Photonics Institute, Vienna University of Technology

Financial support DFG (SP 687/1-2) and Austrian Science Fund (F016 P03)
Pulsed X-ray sources

Photon Energy [eV]

Brilliance photons/(s mm² mrad² 0.1% BW)

10⁰ 10⁶ 10¹² 10¹⁸ 10²⁴

Ti:S laser
Undulator Bessy II
X-ray FEL (proposed)
Discharge pumped x-ray laser
Laser pumped x-ray laser
SPPS
High harmonic soft x-rays
Laser plasma x-ray line source

Pulse-duration

100 as
100 fs
100 ps
X-ray absorption spectroscopy

- **Pre-Edge**
  - Absorption coefficient $\mu(E)$

- **Post-Edge**

**XAMES**

X-RAY ABSORPTION MAIN EDGE STRUCTURE
- coordination number
- coordination geometry

**EXAFS (> 50 eV)**

X-RAY ABSORPTION EXTENDED FINE STRUCTURE
- atomic distances
- neighboring atomic species

**XANES (< 10 eV)**

X-RAY NEAR ABSORPTION EDGE SPECTROSCOPY
- band structure
- Molecular orbitals

Department of Physics, University of Würzburg, Germany
High Harmonic Generation (HHG)

Department of Physics, University of Würzburg, Germany

Femtosecond laser pulse (> $10^{14}$ W/cm$^2$)

Laser radiation is up-converted into spatially coherent soft x-ray radiation
HHG Spectrum

Department of Physics, University of Würzburg, Germany

odd harmonics of laser frequency

XUV continuum

Laser pulses with $\sim 12$ fs $>10^{16}$ W/cm$^2$ $\lambda = 780$ nm focused into Neon

Photons/s (10% BW): C K-edge (284 eV): $2 \times 10^7$
Ti-L-edge (453 eV): $1 \times 10^6$

HHG: Temporal Characteristic

HH coherent process:
XUV emission only within laser pulse duration

After spectral filtering:
train of sub-fs pulses

single attosecond pulse possible

$t_p = 100$ fs
XUV radiation

$t_p = 25$ fs

$t_p = 5$ fs
Time-resolved XAS

Department of Physics, University of Würzburg, Germany

Sample: polycrystalline (amorphous) Silicon (100nm thick)
Pump: 780nm, 20fs 2-5 mJ/cm² (10¹⁹cm⁻³ < N₀ < 10²⁰cm⁻³)
Probe: XUV continuum 100-600eV, < 20fs, spot size on sample 1mm x 1mm
XAMES @ Si L-edge

Si L_{II,III} absorption edge

Measured differential transmission spectra

Information about carrier distribution in the valence and conduction band

Two time constants observed:
- sub-ps: electron-phonon scattering
- several 10’s of ps: recombination
EXAFS Si L-edge

Calculated separation $r = 2.46\text{Å} \pm 10\%$

(Literature: amorphous Silicon: $2.37\text{Å}$)
T-EXAFS

Department of Physics, University of Würzburg, Germany

Energy (eV) 0 0.2 0.4 0.6 0.8 1

Delay (fs) 0 204 0 608 0 100

Energy 100 ..500eV
Delay 0 ..800fs, 20fs steps

Fourier-transform

Energy (meV)

TA – phonon

LO – phonon

Stampfli & Bennemann,
PRB 49 7299, 1994
Conclusion Outlook

• High harmonic radiation has been used for soft x-ray absorption spectroscopy with sub-20fs resolution
• Measured carrier dynamics with T-XAMES
• Measured lattice dynamics (coherent phonons) with T-EXAFS
• Improving the setup
• Time-resolved studies in the water window