Summary of Ultrafast Chemistry/AMO Breakout Session

Shin-ichi Adachi & Tetsuo Katayama

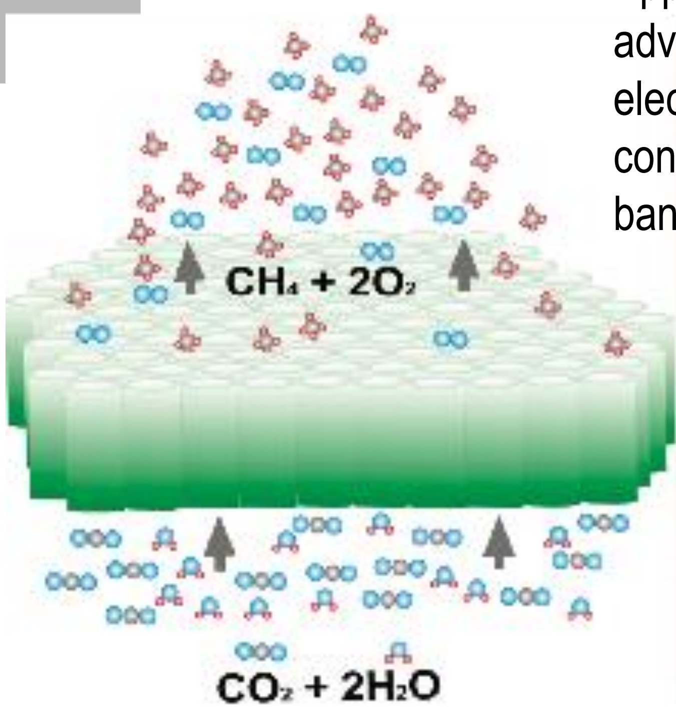
Session 2. Ultrafast Chemistry/AMO

"Perspectives of Chemical Applications of Ultrafast X-ray/XUV Spectroscopy & Scattering at SACLA" organized by S. Adachi (KEK) and T. Katayama (SACLA)

Presenter	Affiliation	Title
C. Milne	SwissFEL	Applications of X-ray scattering and spectroscopy at SwissFEL for investigation of ultrafast chemical dynamics
S. Adachi	KEK	Chemical Applications of Ultrafast X-ray/XUV Spectroscopy & Scattering
H. Fukuzawa	Tohoku Univ.	Ultrafast dynamics induced by FEL or optical laser in atoms, molecules, and clusters
F. Lima (T. Katayama)	European XFEL	Ultrafast ligand exchange reactions in iron complexes

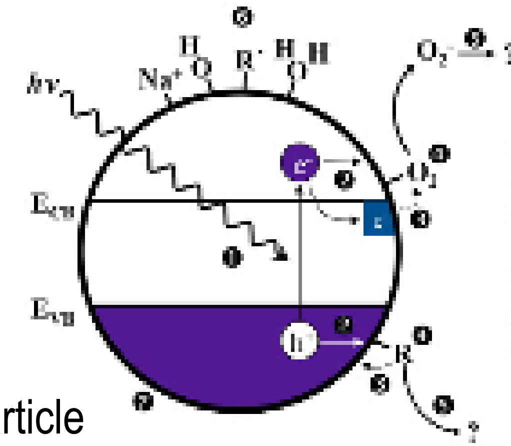
Photocatalysis and energy conversion

Metal-oxide nanoparticles are commonly used in dye-sensitized solar cells (DSSCs) and as photocatalysts



S.C. Roy et al. *ACS Nano* **4**, 1259 (2010)

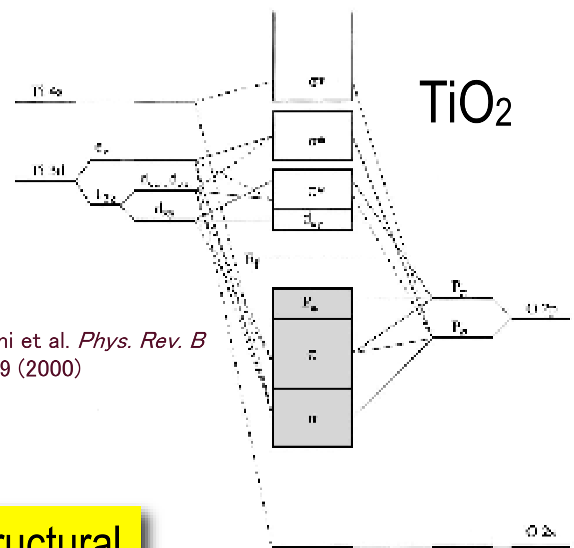
Applications take advantage of an electron/hole in the conduction/valence band of the nanoparticle



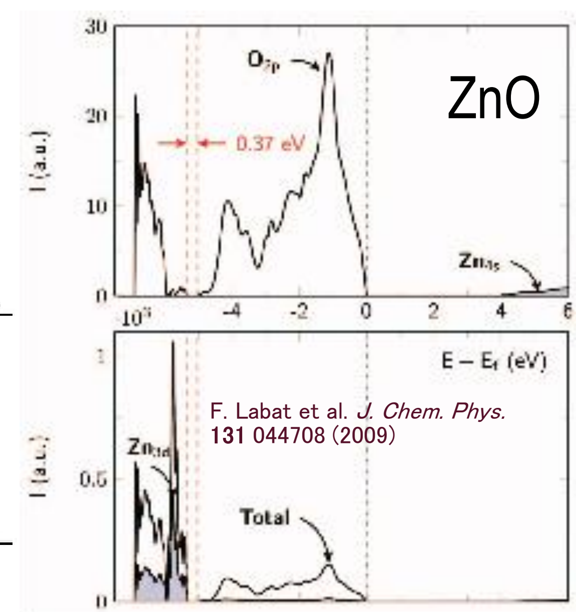
M. Henderson *Surf. Sci. Rep.* **66**, 185 (2011)

Important issues
- excitation
- charge transport and trapping
- charge transfer
- molecular adsorption
- reaction mechanisms
- poisons and promoters
- surface and material structure

Valence and conduction bands have distinct atomic character



R. Asahi et al. *Phys. Rev. B* **61** 7459 (2000)



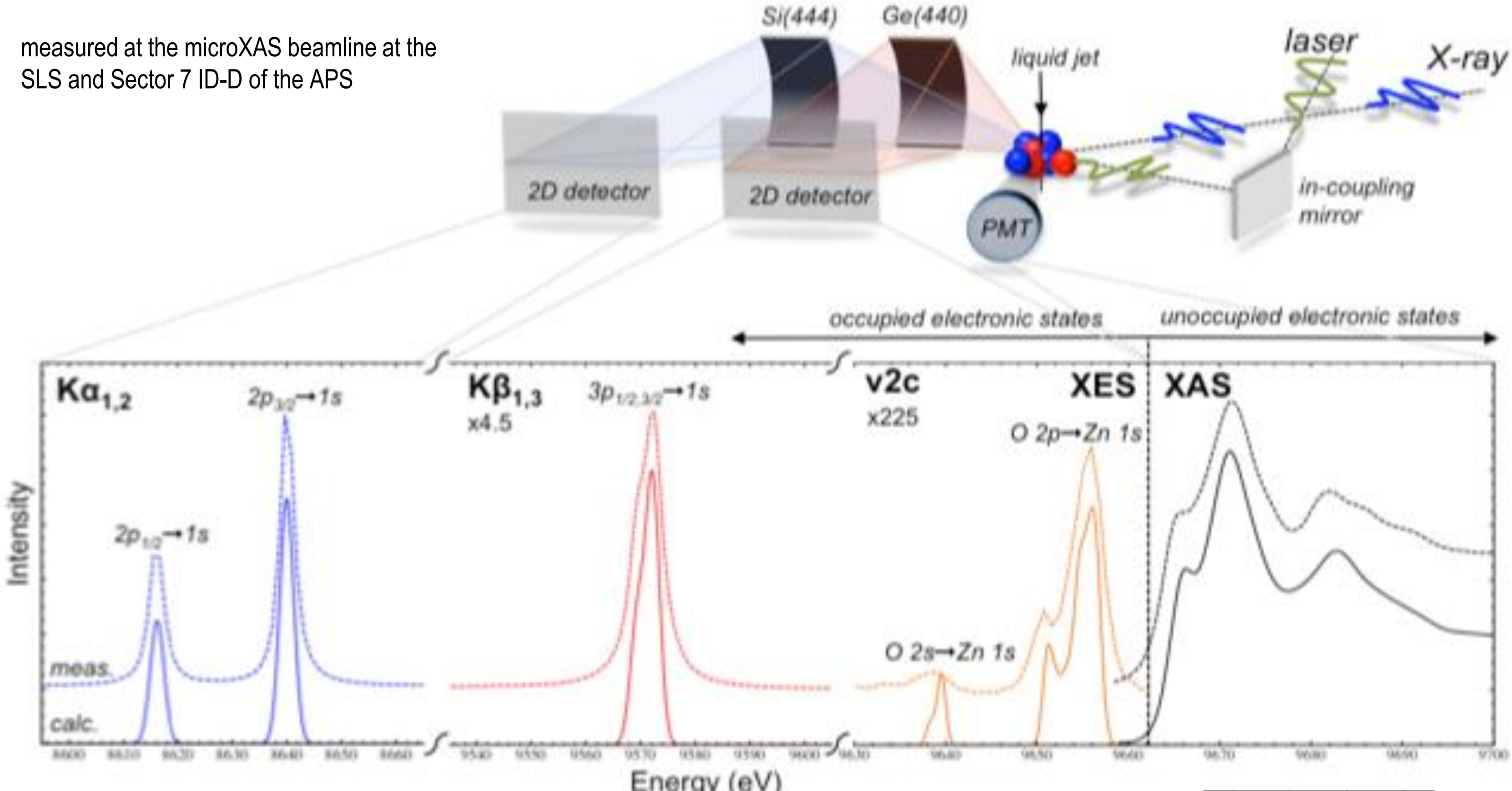
F. Labat et al. *J. Chem. Phys.* **131** 044708 (2009)

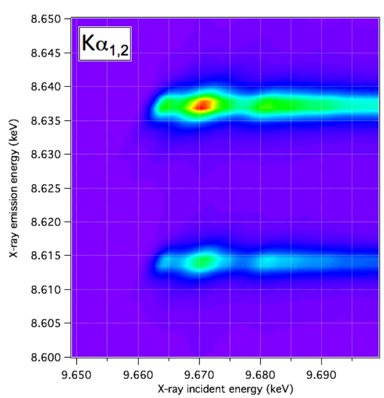
We want time-resolved electronic and structural information on these systems as they function

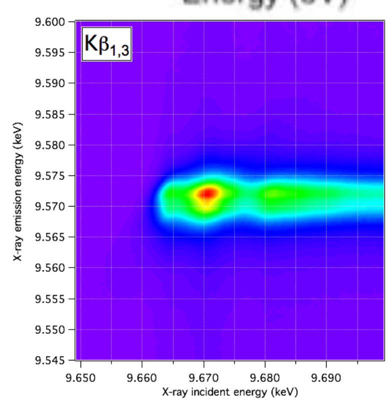
PAUL SCHERRER INSTITUT PSI

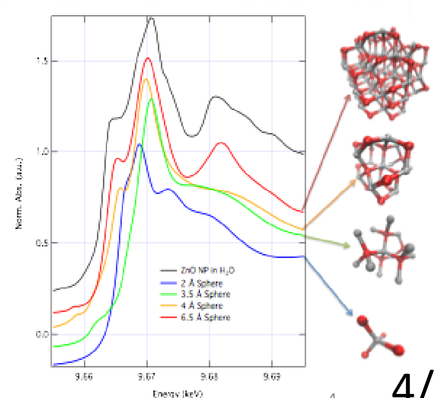
High-repetition rep rate RXES at the APS

measured at the microXAS beamline at the SLS and Sector 7 ID-D of the APS



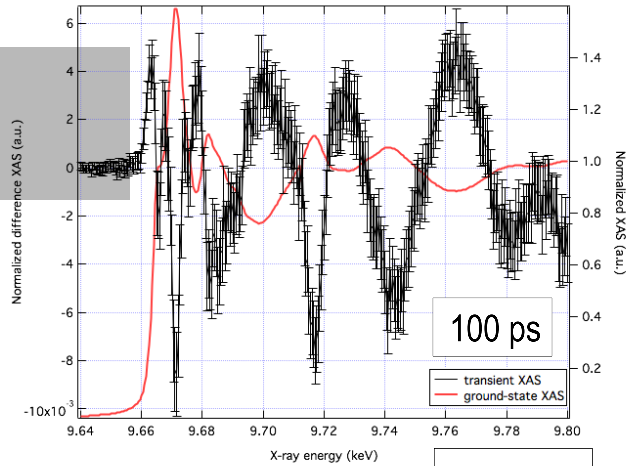




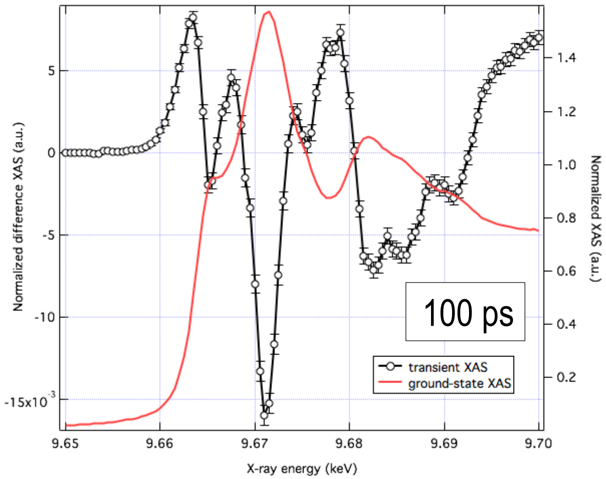


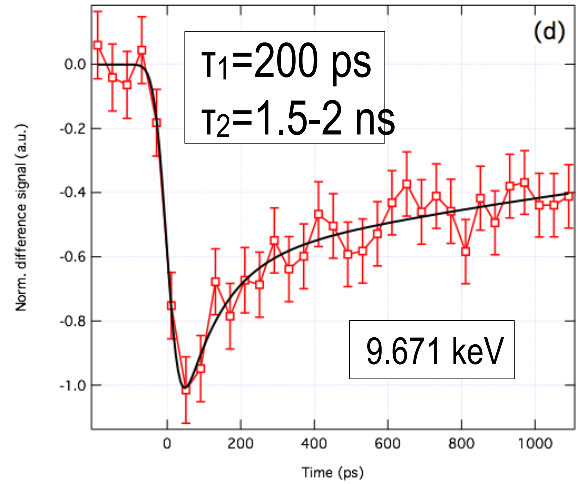
ZnO pump-probe X-ray spectroscopy

EXAFS

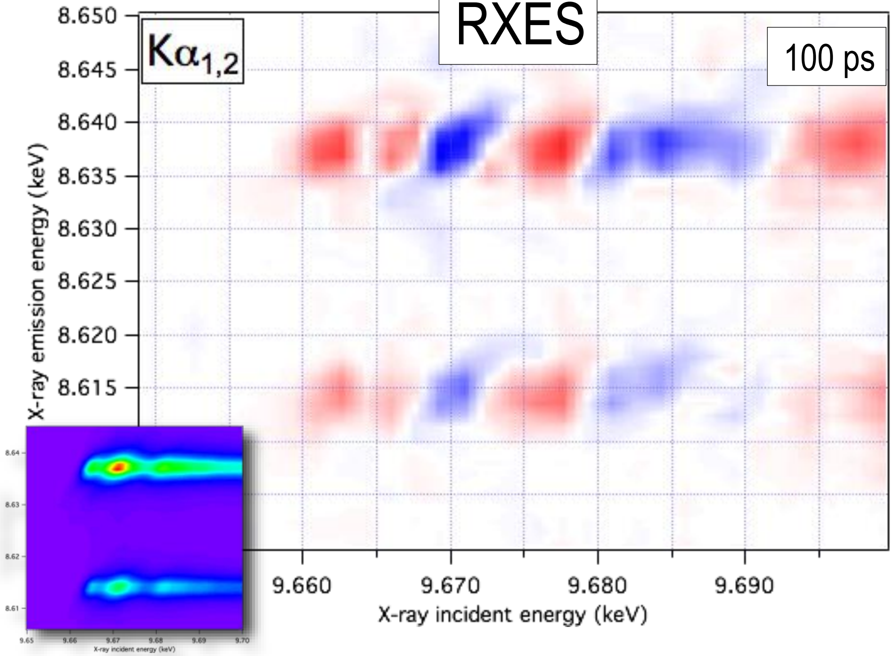


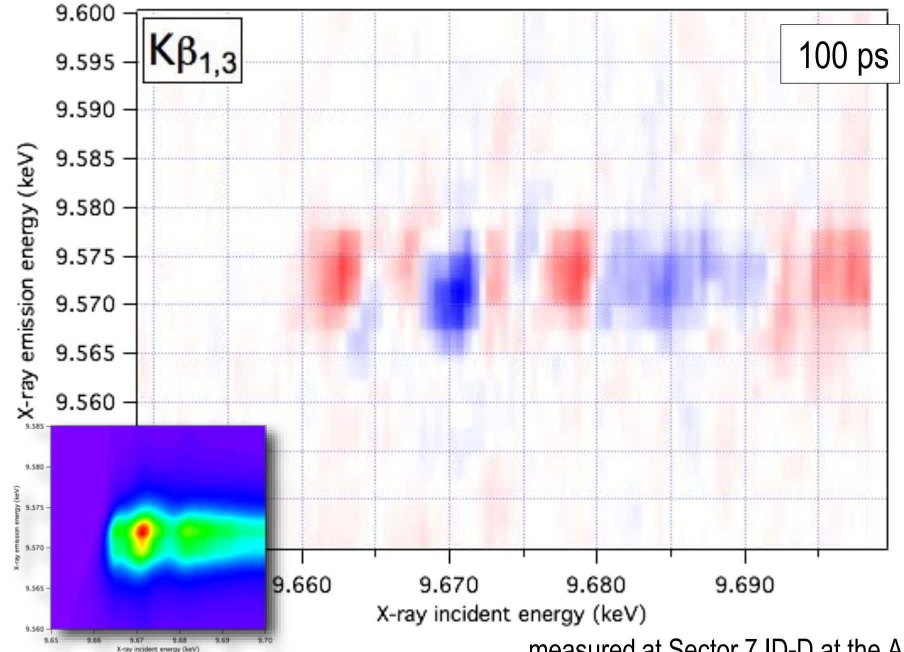
XANES





RXES





measured at Sector 7 ID-D at the APS

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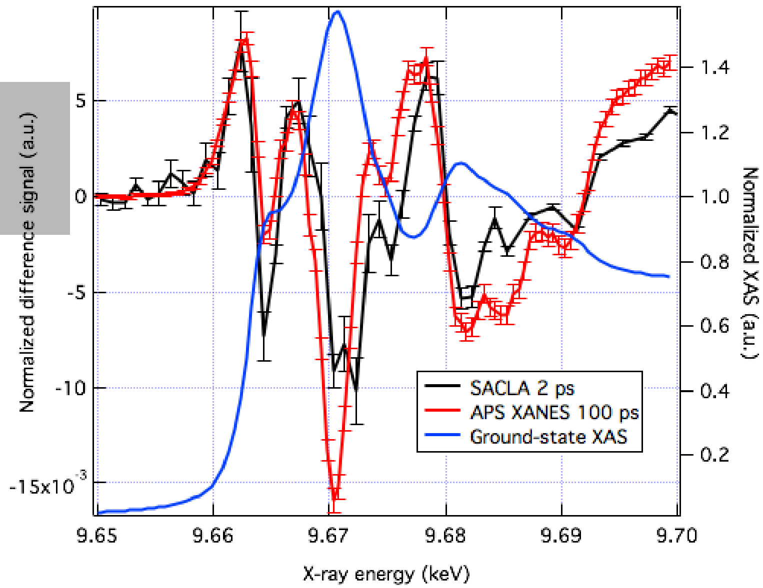
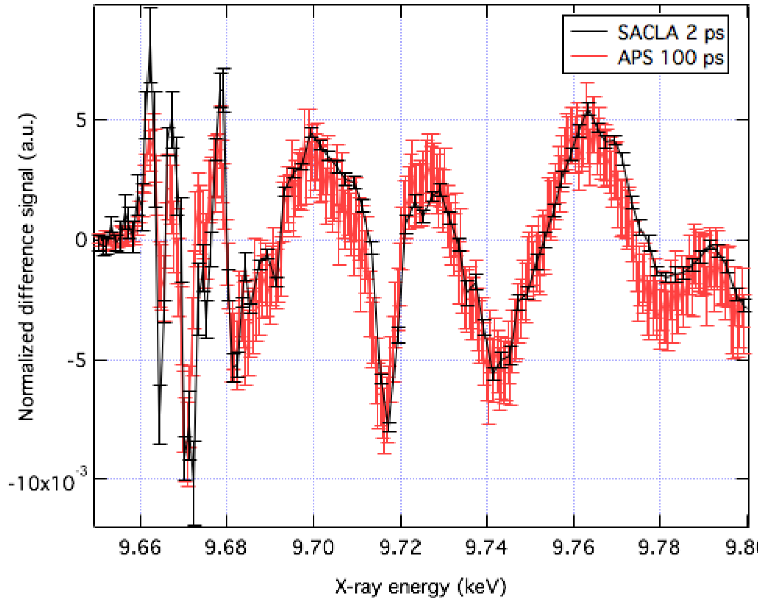
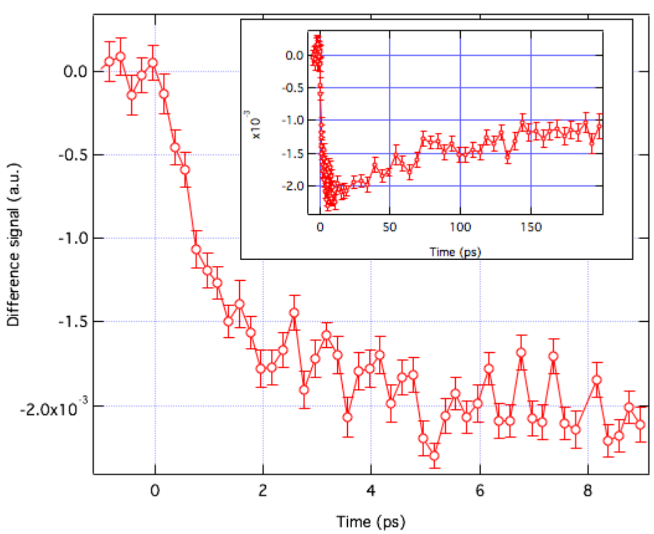
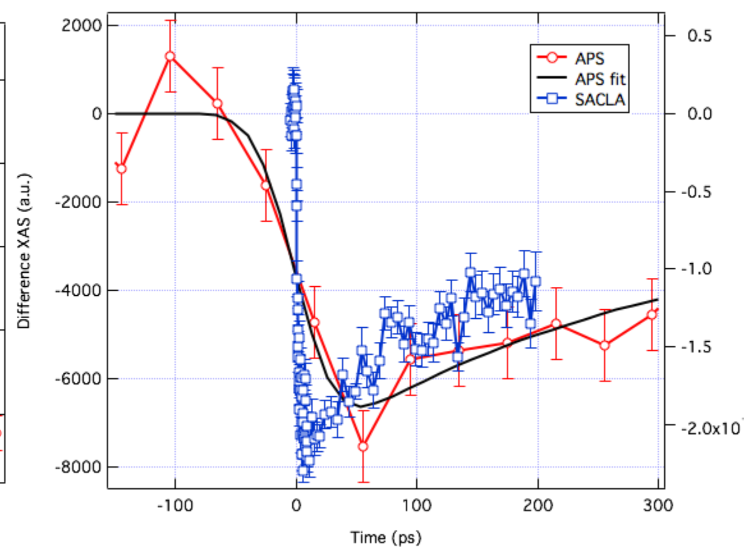
ZnO at SACLA: fs X-ray spectroscopy

Collaboration with Argonne, EPFL, XFEL, SACLA, DTU, Newcastle and Tohoku









Preliminary analysis of XAS indicates we populate the oxygen vacancies within 1.2 ps

ACS DIVISION OF PHYSICAL CHEMISTRY

Chemical Applications of Ultrafast X-ray/XUV Spectroscopy and Scattering

Josh Vura-Weis
Renske van der Veen
Philippe Wernet

Organizers

Boston Convention Center
August 19-23, 2018

5-day symposium

Room 206A
(*Room 257A on Thursday*)

Cosponsored by the Division of Inorganic Chemistry

SUNDAY MORNING
Small-Molecule Photophysics
R. van der Veen, J. Vura-Weis, P. Wernet, Presiding

8:30 (8). Photoinduced reaction mechanisms from time-resolved X-ray spectroscopy of ligand- and heteroatoms in ligands and organic molecules. **M. Ochmann**, A. Hussain, I. von Ahnen, A. Cordones, K. Hong, J.H. Lee, R. Ma, K. Adamczyk, T.K. Kim, R.W. Schoenlein, O. Vendrell, **N. Huse**
9:10 (9). Deconvoluting the isotropic and anisotropic ultrafast x-ray scattering of gas-phase N-methylmorpholine following Rydberg excitation. **B.M. Stankus**, J.M. Ruddock, H. Yong, N. Zotev, D. Bellshaw, T.J. Lane, S. Boutet, M. Liang, S. Carbajo, J.S. Robinson, J.E. Koglin, A. Aquila, Y. Zhang, W. Du, N. Goff, Y. Chang, M.P. Minitti, A. Kirrander, P.M. Weber
9:30 (10). Ultrafast x-ray molecular dynamics. **S.R. Leone**
10:10 (11). Time-resolved gas-phase X-ray scattering to reveal transients in photodissociation reactions. **J.M. Ruddock**, B.M. Stankus, H. Yong, W. Du, D. Bellshaw, N. Zotev, T.J. Lane, M. Liang, M.P. Minitti, S. Boutet, A. Kirrander, P.M. Weber
10:30 INTERMISSION
10:50 (12). Probing chemical dynamics by soft-X-ray transient absorption and XUV photoelectron spectroscopy. **H. Wörner**
11:30 (13). Non-adiabatic coherent electron dynamics in iodine monobromide probed by XUV attosecond transient-absorption spectroscopy. **Y. Kobayashi**, K. Chang, T. Zeng, M. Reduzzi, H. Timmers, M. Sabbar, D.M. Neumark, S.R. Leone
11:50 (14). Soft-X-ray spectroscopy of the amine group: Hydrogen bond motifs in alkylamine/alkylammonium acid-base pairs. M. Ekimova, M. Kubin, M. Ochmann, J. Ludwig, N. Huse, P. Wernet, M. Odelius, **E. Nibbering**
12:10 (15). Femtosecond dynamics in the iodomethane cation investigated by XUV-IR pump-probe ion imaging. **L. Banares**, G. Reitsma, M.L. Murillo-Sanchez, R. de Nalda, M. Corrales, S. Marggi Poullain, J. Gonzalez-Vazquez, M. Vrakking, O. Kornilov

HHG
Laser
XUV

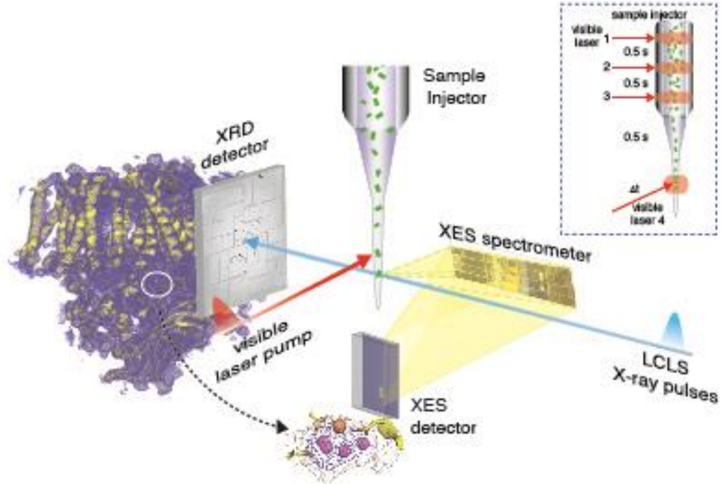
SUNDAY AFTERNOON
Biological Applications
R. van der Veen, J. Vura-Weis, P. Wernet, Presiding

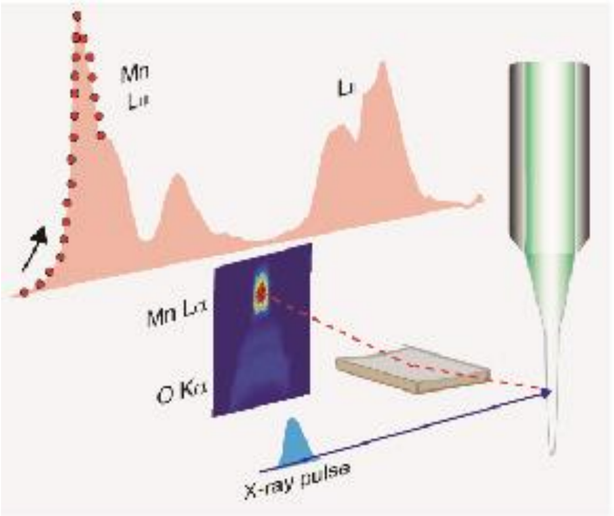
1:30 (54). Activating metal sites for biological electron transfer. **E.I. Solomon**
2:15 (55). Light, molecules, action: Ballistic excited state dynamics of cobalamins revealed by polarized fs-XANES. **R.J. Sension**
3:00 (56). Ultrafast photophysics of coordination complexes with tabletop XANES. **E. Ryland**, J. Vura-Weis
3:20 INTERMISSION
3:40 (57). Ultrafast reaction pathways in a metalloprotein revealed by optical polarization selected X-ray transient absorption spectroscopy and quantum mechanical calculations. M. Shelby, D. Hayes, P.J. Lestrange, H. Lemke, D. Zhu, X. Li, **L.X. Chen**
4:25 (58). Taking snapshots of reaction intermediates in metalloenzymes and catalysts at X-ray free electron lasers. J. Kern, F. Fuller, R. Chatterjee, S. Gul, M. Kubin, R. Mitzner, U. Bergmann, P. Wernet, V.K. Yachandra, **J. Yano**

Bio

Topics (Bio)

Taking snapshots of reaction intermediates in metalloenzymes and catalysts at X-ray free electron lasers
J. Yano (LBNL)





time-resolved Mn *L*-edge RIXS with zone-plate spectrometer at LCLS-II

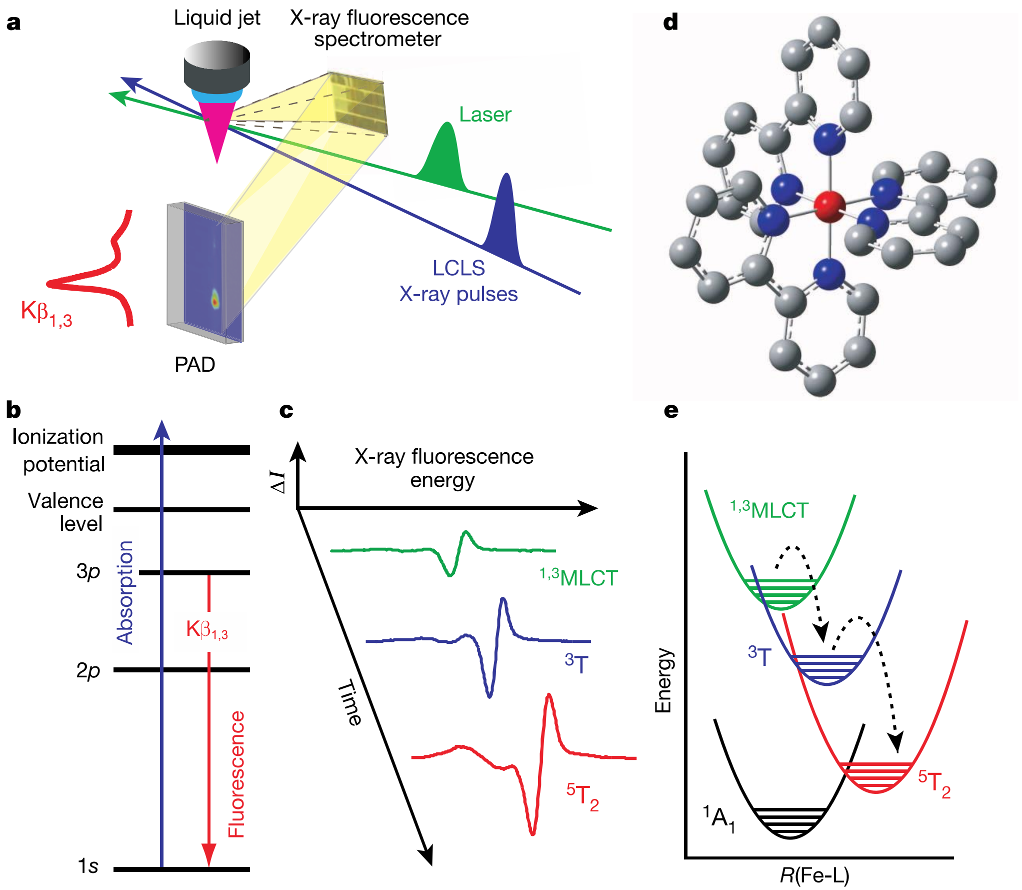
Young *et al.* (2016) Nature, 540, 453.

http://www2.lbl.gov/vkyachan/research.html

Topics (metal complexes in solution)

Ultrafast X-ray laser characterization of electronic excited states in first row transition metal complexes.

K. Gaffney, K. Kunnus (LCLS, PULSE)



Systematic study of the ligand field theory

$Fe(bpy)_3$ -> $(bpy)_2(CN)_2$ -> $(bpy)(CN)_4$ -> $(CN)_6$

Fe L-edge RIXS

High efficiency energy dispersive transition edge sensor (TES) detector

Zhang *et al.* Nature (2014) 509, 345.

Measurements (1/2)

- Existing methods to be improved:
  - X-ray scattering and diffraction, X-ray spectroscopies, i.e. XAS and XES, in particular EXAFS (!)
  - Particularly, EXAFS and diffraction experimental capability should be enhanced at SACLA.
- New methods to be considered:
  - Non-resonant valence-to-core XES, resonant XES, Inelastic X-ray Scattering, i.e. X-ray Raman…
  - Regarding new photon hungry methods, SACLA should not compete with high-rep-rate facilities (LCLS-II, European XFEL) directly.

Measurements (2/2)

- Exploit not only transverse coherence (imaging) but also longitudinal (temporal) coherence.
  - seeded pulses (transform-limited)
  - SACLA has realized the new scheme of self-seeding. For application, stable operation and easy tuning will be key points.
  - What applications? Time-resolved fixed-wavelength RIXS, SFX with small molecule nanocrystals, …
- Include as many different techniques as possible
  - complementary observables will help to disentangle the electronic and nuclear degrees of freedom
  - Combination of XAS, XES and scattering/diffraction is feasible at SACLA

Hardware

- Alternative schemes for “pumping” of chemical reactions:
  - Going beyond the optical wavelengths! Only a small fractions of (bio)chemical reaction can be triggered with light...
  - Bio-sample, dilute systems, using exotic lasers will be good examples to promote science at SACLA.
  - Use “exotic“ lasers to trigger the dynamics, i.e. mid-IR pulses, THz pulses, etc.

Software

- Software improvements:
  - Data reduction and analysis on-the-fly
  - On-line and off-line cluster access (before, during and after beam times)
  - Many aspects of “user friendliness” have improved over past 5 years, thanks!

Accelerator (1/1)

- New accelerator parameters:
  - two-color mode: X-ray pump/X-ray probe experiments
  - extremely ultrashort pulses → sub-5 fs down to sub 1-fs (attosecond)
  - Wavelength tunability in the hard X-ray range
  - broadband X-ray pulses: single-shot dispersive XAS for non-reversible experiment

SACLA-SPring-8 campus offers unique opportunities

- 100-ps and ns-resolved experiments at SR before applying for XFEL beamtime. Preferential access to synchrotron beamtime as part of the XFEL-related projects is preferable.
- On-site optical spectroscopy laboratories to carry out complementary optical measurements
- Utility support is also important.

AMO / H. Fukuzawa (Tohoku Univ.)

Request from AMO

Optical laser

1. Pointing stabilized optical laser. Active feed back system must be considered.
2. Short pulse duration of the optical laser (comparable with FEL).

FEL@BL1

0. We look forward to using the timing monitor @BL1
1. Shorter pulse duration for the FEL@BL1 (<10fs).
2. Split delay system @BL1 for FEL-pump–FEL-probe.
3. Shot-by-shot in-line spectrometer.
4. Seeded FEL

In order to use 3rd order light @BL1 (>300 eV)

1. Operation mode with relatively high intensity for higher-order light.
2. 1st, 2nd, and 3rd order light resolved FEL intensity monitor.

Software

1. Online (immediate) analysis of the arrival timing monitor.

Accessary

1. Precise alignment laser (smaller spot size, parallel to the FEL)

end