



HOKKAIDO
UNIVERSITY

Pump-Probe **2 beam** XAS spectra

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Two photons

1. One + One Photons

1. One phenomenon with two monitor beams (Two wave lengths)
2. Combination of different methods
 1. Diffraction + Absorption
 2. One photon + One positron
3. Pump (One photon) + Probe (One photon)

Important parameters. Wavelength, size and time, Intensity

2. Nonlinear process

1. Two photon absorption, Sum frequency, Non-linear process, Induced Raman, Induced Fluorescence, Resonance Raman.

Important parameters Phase, photon density + above.

In order to understand Catalytic Reaction

Catalytic reaction.

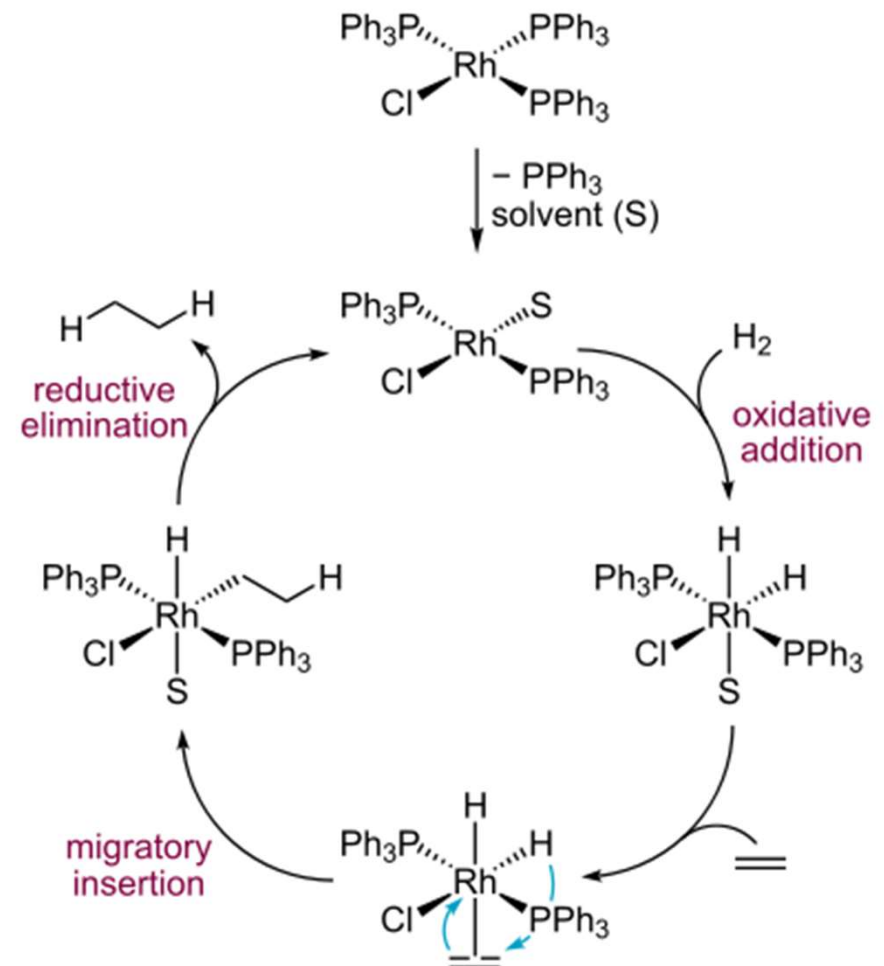
To decide the active site and determine its structure.

To determine the intermediate structure and reaction step.

To determine the kinetics and the mechanism

To write the reaction cycle.

For homogeneous catalyst, the active structure is isolated and a



Wilkinson Catalyst

Ni₂P 脱硫触媒へのIn situ EXAFSの応用 2001-2007

In situ EXAFS is a powerful tool.

The hydrodesulfurization reactions are often carried out in the presence of oil under high-pressure and high temperature conditions.



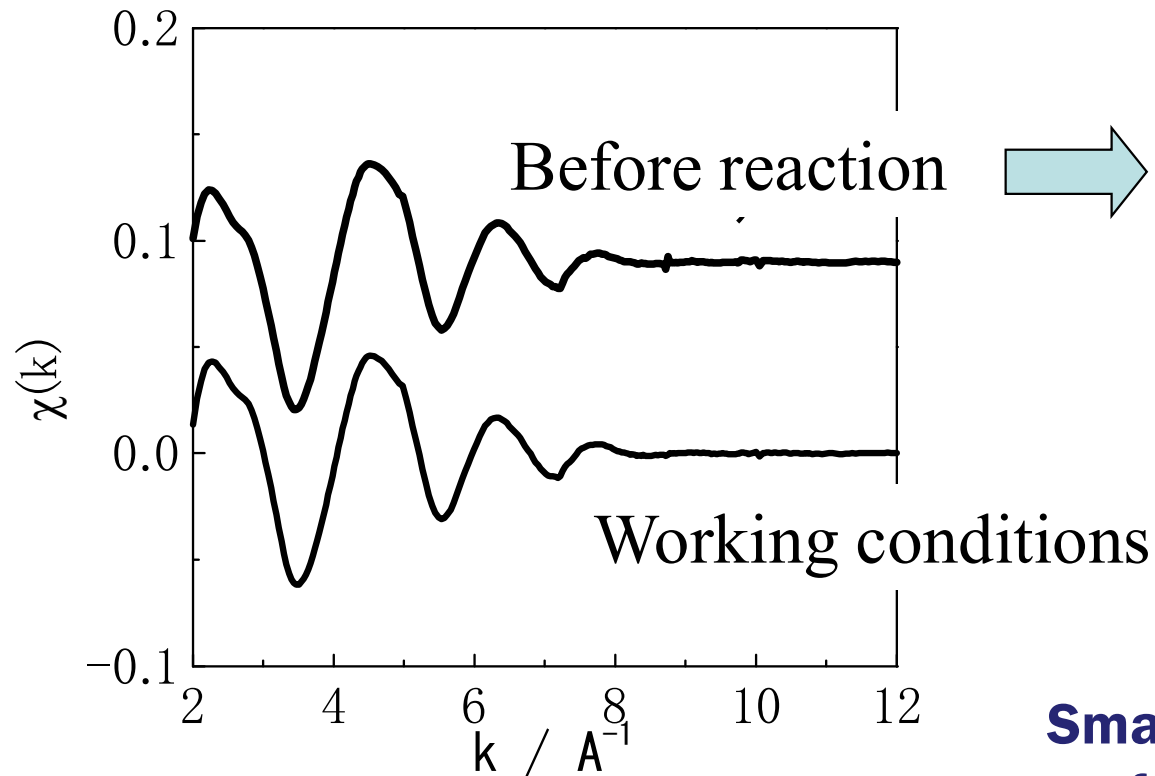
油から脱硫反応を起こしている最中を測定する。

油はX線を吸収し、熱を伝える。

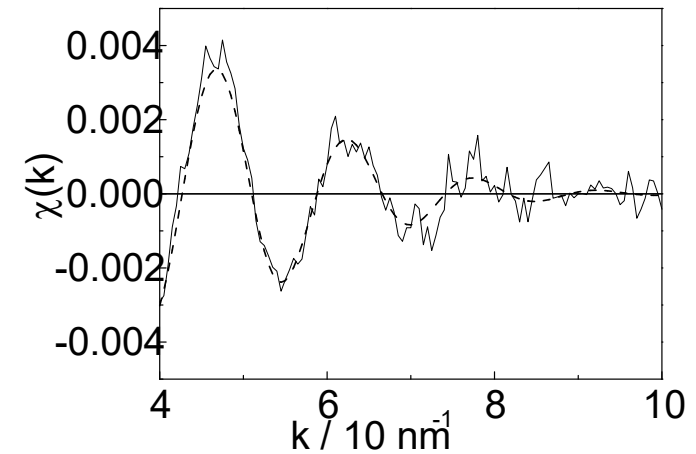
窓と炉を離すことが不可能

Oyama, S. T.; Wang, X.; Requejo, F. G.; Sato, T.; Yoshimura, Y., Hydrodesulfurization of petroleum feedstocks with a new type of nonsulfide hydrotreating catalyst. *J.Catal* 2002, 209, 1.

In situ XAFS of Ni₂P in the reaction.



Ni₂P bulk structure is stable.



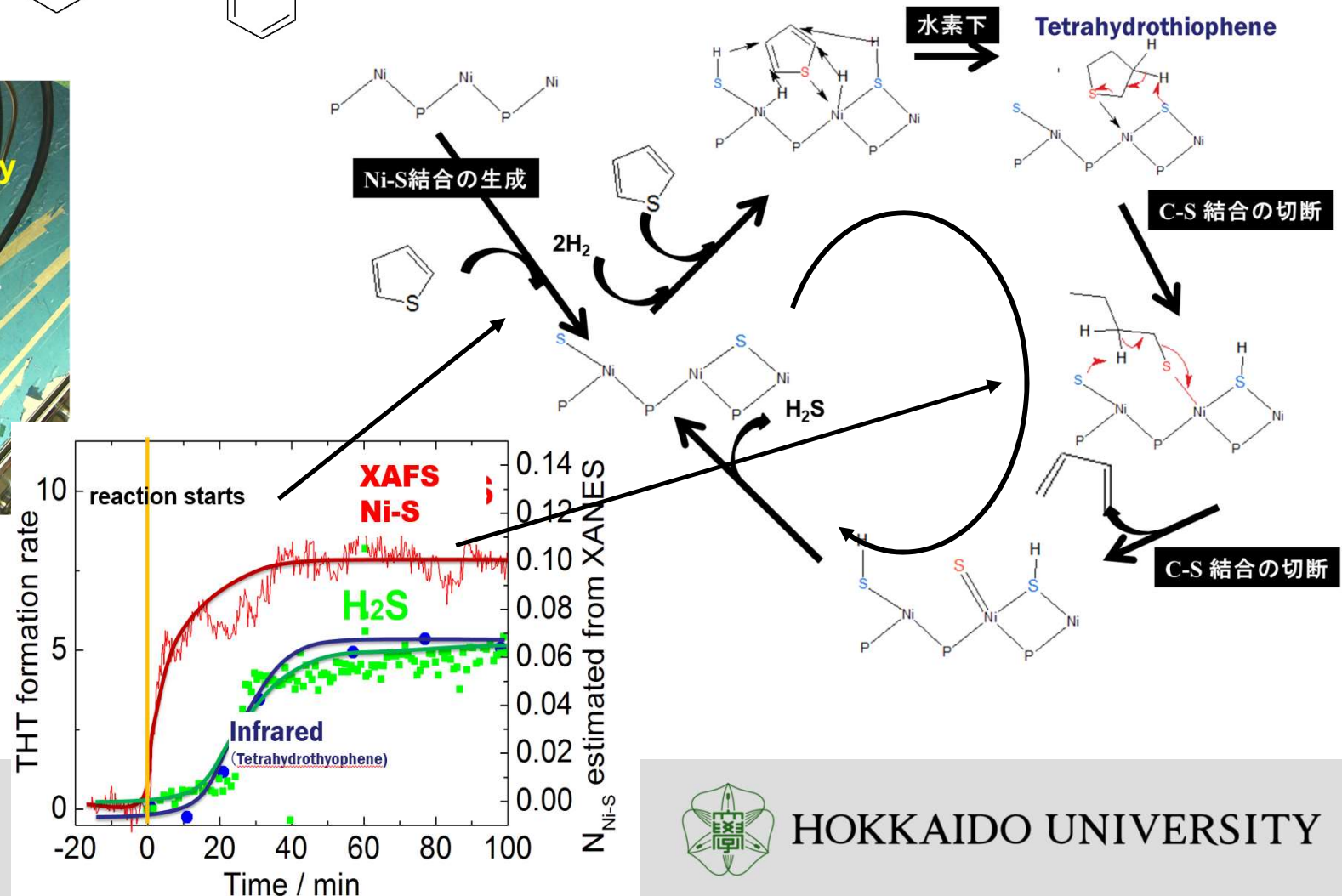
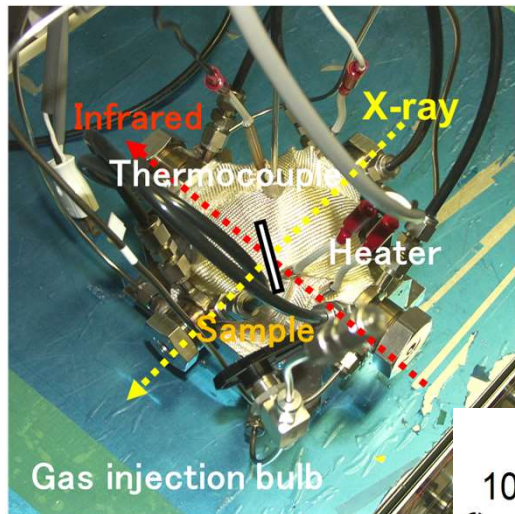
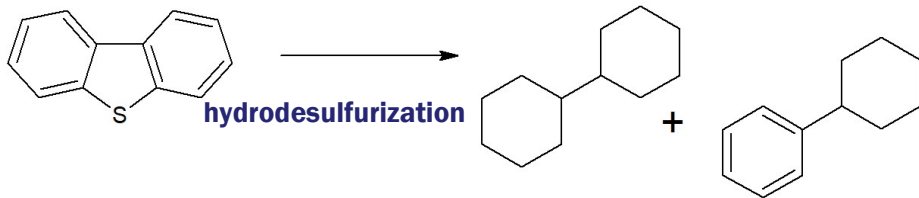
Difference spectrum.

Ni-S=0.227 nm

Small change (probably in surface) occurs.

Correlation = simultaneous measurement

Ni2P



Ni₂P with two colors.

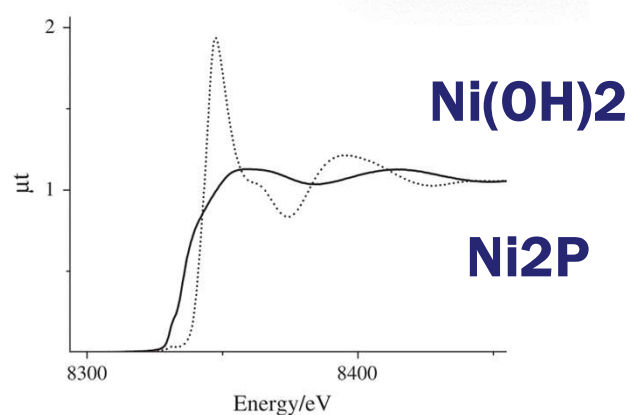
2 colors:

Ni K-edge

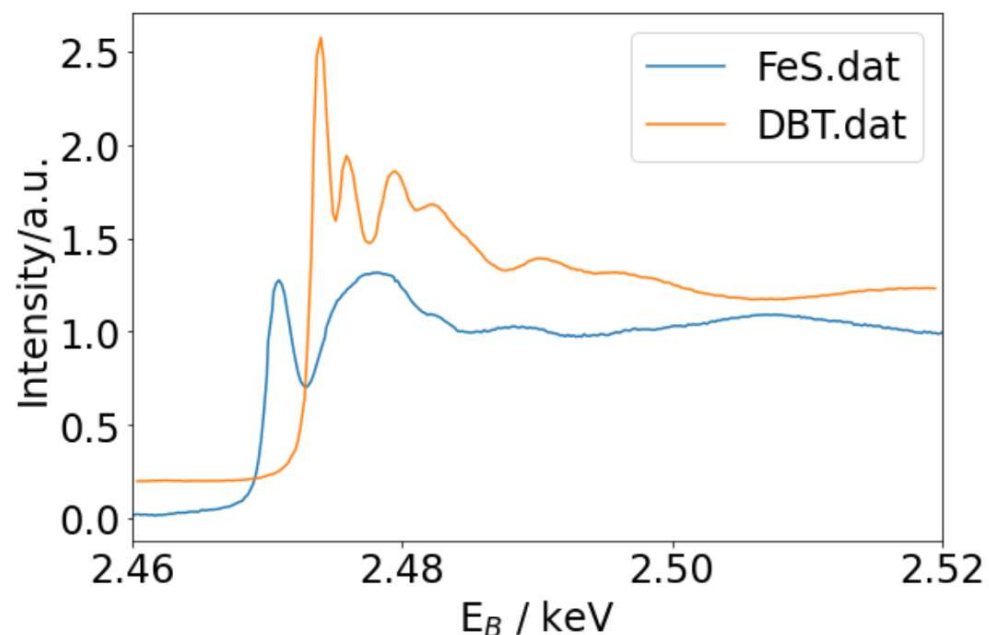
P K and S K edges

S(Thiophene), S(THT)

S-Ni



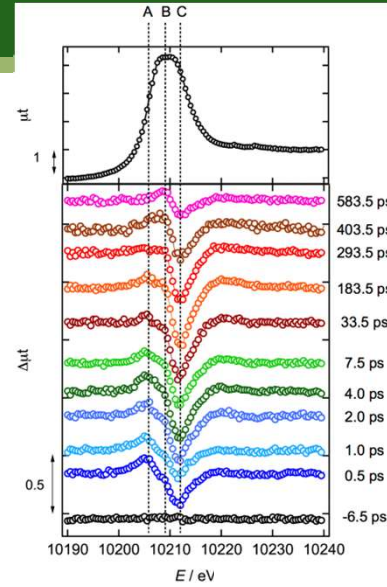
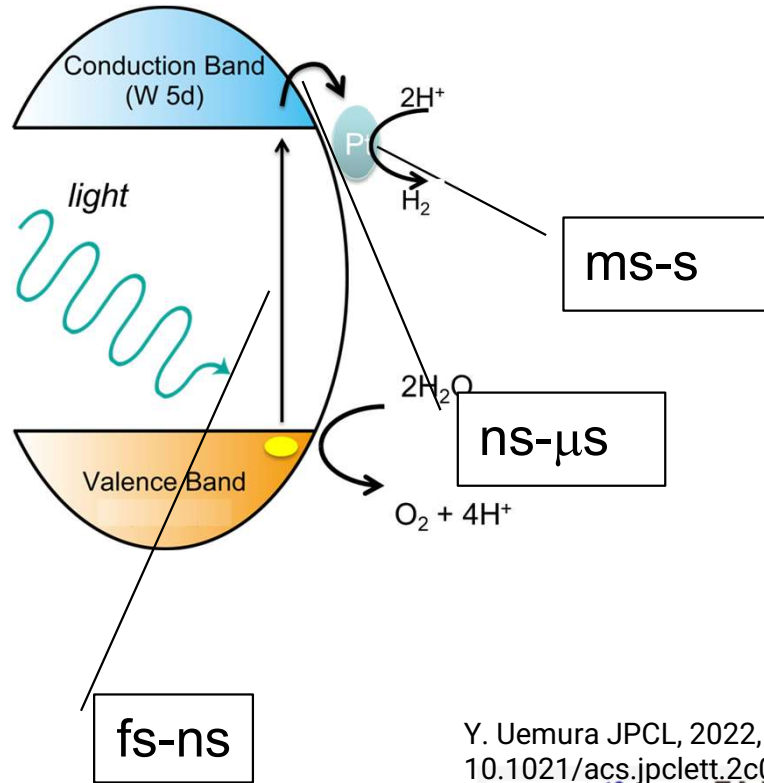
Time-resolved XANES
studies will be enough
for Kinetic studies



<http://www.esrf.eu/home/UsersAndScience/Experiments/XNP/ID21/php.html>

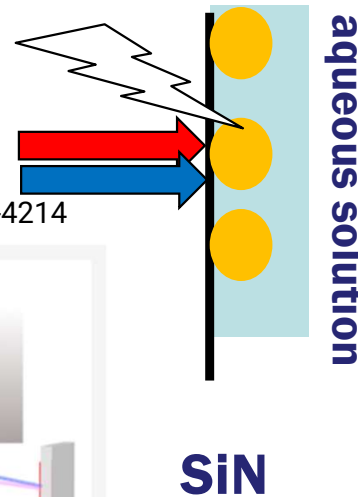
Proposal: Photoexcitation process and carrier

Y. Uemura AgCIE 2016, 55, 1364-1367



Simultaneous measurement of O and Co, Pt. with 2 beam.

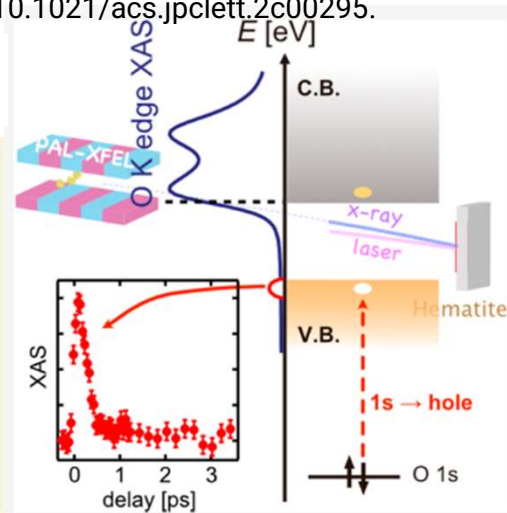
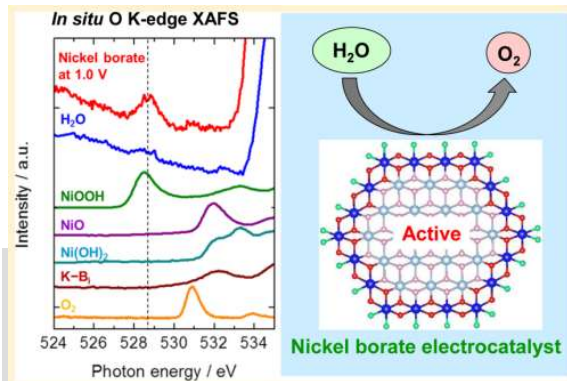
CoOOH



electron hole kinetics.

1 1 M. Yoshida, JPCC 2015, 119, 19279-19286 10.1021/acs.jpcc.5b06102.

Y. Uemura JPCL, 2022, 13, 4207-4214
10.1021/acs.jpcllett.2c00295.



Necessary conditions

Two beams have the same size to monitor the same position,

Two beams have the same timing after the pump beam.

The same pulse width

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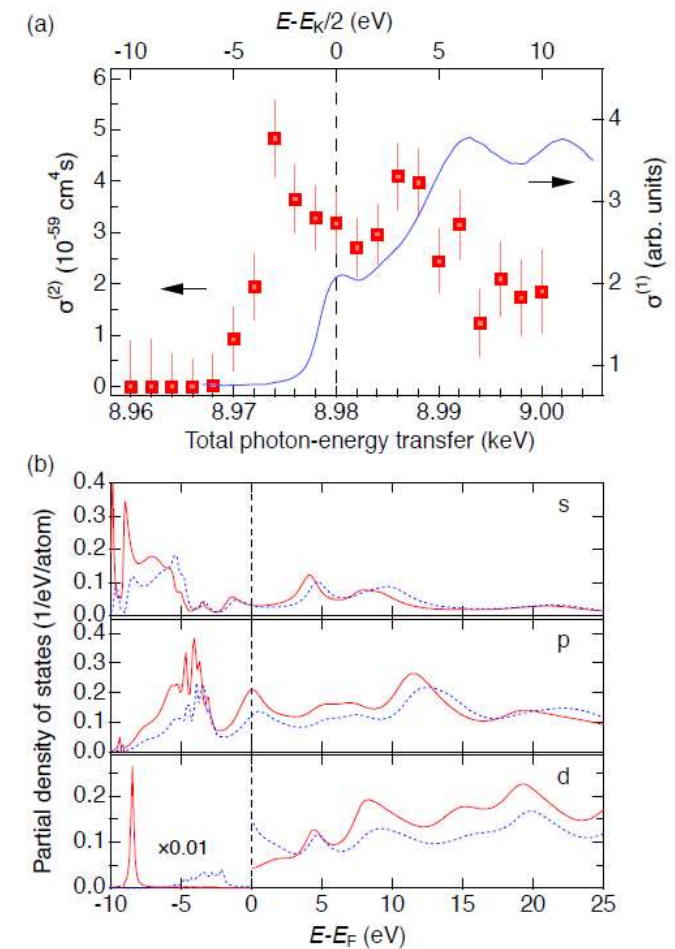
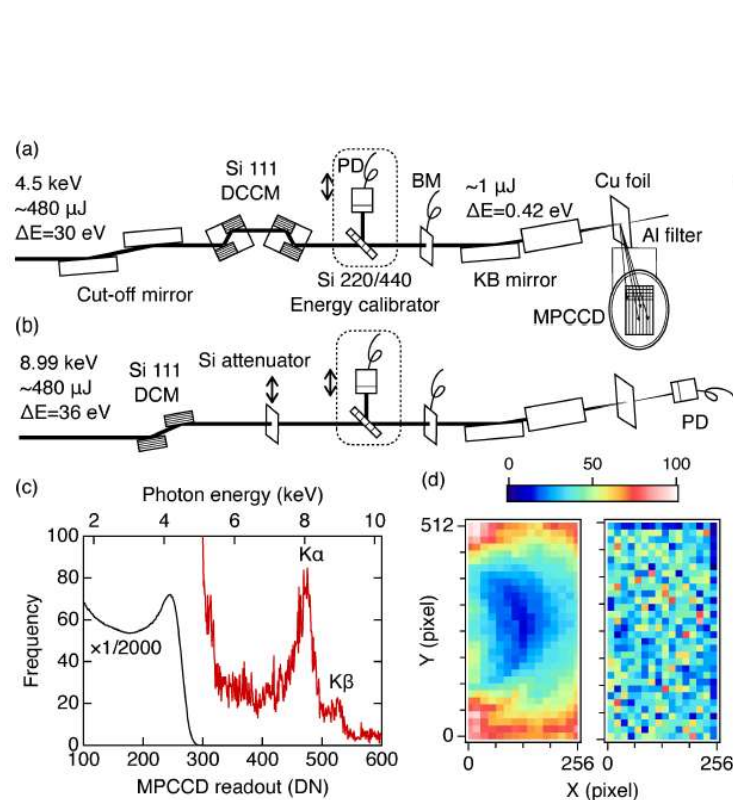
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Important parameters Phase, photon density + above coherence

Two Photon absorption



Tamasaku, K., Shigemasa, E., Inubushi, Y., Inoue, I., Osaka, T., Katayama, T., Yabashi, M., Koide, A., Yokoyama, T., Ishikawa, T., 2018. Nonlinear Spectroscopy with X-Ray Two-Photon Absorption in Metallic Copper. *Physical Review Letters* 121.. doi:10.1103/physrevlett.121.083901

X-ray induced Fluorescence

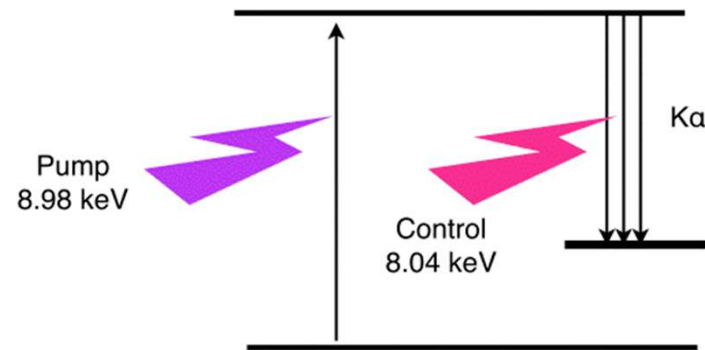


図 1. 本研究で目指した誘導過程の遷移図

弱い励起 X 線で銅原子の K 殻をイオン化し、強いコントロール X 線で $K\alpha_1$ 線を誘導放出させる。

DOI 10.18957/rr.10.4.405 SPring-8/SACLA 利用研究成果集