Stimulated X-ray Emission Spectroscopy with hard X-rays

Thomas Kroll

SLAC National Accelerator Laboratory
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X-Ray Emission Spectroscopy

Valence levels

3p

Kβ_{2,5}, Kβ''

Kβ_{1,3}, Kβ'

2p

Kα_{1,2}

1s

Level Diagram

Spin/oxidation state of transition metals

Valence orbitals: ligand type, structure, covalency, ligand protonation, etc

Stimulated X-ray Emission Spectroscopy

Stimulated (non-linear) emission:

Spontaneous (conventional) emission

- Low photon flux density (only a few photons at a time in the sample)
- Only spontaneous emission in $4\pi$ direction
Stimulated X-ray Emission Spectroscopy

Stimulated (non-linear) emission:
Amplified spontaneous emission (ASE)

- Very high flux density (~$10^{11}$ ph / 10 fs / 100x150 nm$^2$)
- Creation of population inversion
- Non-linear effects become dominant
- Cascade like decay
Experimental Setup

- Highly focused 10 fs beam (100 x 150 nm²)
- Flat crystal analyzer in forward direction
Objectives

- Can stimulated XES be applied to transition metal solutions?
- What is fundamentally new information provided by stimulated XES techniques that is not achievable with other techniques?
- Can stimulated XES be used for chemically sensitive X-ray emission lines? Is the chemical information preserved?
- Can we stimulate the weaker Kβ emission?
Single shots:

- Clean curve in single shot
- Only the $K\alpha_1$ visible
- FWHM < normal $K\alpha$ width

MnCl$_2$ solution
Lasing in MnCl$_2$ solution

Proof of Lasing:
- Exponential increase
- Linear gain regime below ~2 mJ
- Saturation reached > 2 mJ

Peak Position and Width

- Low to mid high photon numbers:
  - Constant broadening and position
- High photon numbers:
  - Spectral broadening
  - Shift to lower energies
- Variations through beam position, temporal shape, lasing condition

Line broadening due to saturation effects
Additional final states become visible
Peak Position and Width

- Mn Kα₁ life-time broadening: 1.48 eV (Krause and Oliver, 1979)
- Lowest observed S-XES peak width: < 1.0 eV
- Darwin width of Si (111): 0.77 eV
- Lowest S-XES lifetime broadening: < 0.5 eV
Stimulated (non-linear) emission:

Amplified spontaneous emission (ASE) + seeded stimulated emission

Pump: > abs. energy
Seed: Kβ energy

- Kβ Seed pulse outruns Kα emission → seeded stimulated Kβ emission
- Requirement: Overlap in time, space and energy
Experimental Setup

- Highly focused 10 fs beam (100 x 150 nm²)
- Use part of the undulators for seed pulse
- Two flat crystals in series in forward direction for simultaneous detection of Kα and Kβ
Seeded Stimulated Kβ emission

Biggest issue:
- Seed pulse and stimulated emission are at the same energy
- How to separate the stimulated signal from the SASE seed pulse?

Pump:
> abs. energy

Seed:
Kβ energy
Conclusions

• Strongly stimulated XES in Mn solutions has been observed
• Gain curves for X-ray lasing have been measured
• Gain narrowing to less than the natural line width
• Chemical information appears to be preserved!
• Seeded stimulated Kβ emission observed

Future

• Improved diagnostics:
  • especially in time domain
  • Seeded beam diagnostic
Stimulated X-ray Emission has been observed

Past achievements:

Soft X-rays
- Stimulated soft X-ray Ne laser
  (Rohringer et al., *Nature* 481, 488 (2012), LCLS)
  - Ne gas
- Stimulated X-ray Emission in Silicon L Lines (70-100 eV)
  (Beye et al., *Nature* 501, 191 (2013), LCLS)
  - Si

Hard X-rays
- Stimulated hard X-ray Fe laser
  (Yoneda et al., *Nature* 524, 446 (2015), SACLAL)
  - Fe foil
High number of photons:

Varying pulse energy:

- Increase of $K\alpha_2$ for high fluences
- Can be explained by final state effects

MnCl$_2$ FWHM:
- Green: 2.2 eV
- Blue: 3.3 eV
- Orange: 4.6 eV

Cu foil (Yoneda et al., *Nature* 524, 446 (2015))

Chemical Sensitivity

Compare solution spectra:

- Stimulated data retain the expected chemical shift
- Need to address stimulated photon / matter interaction in more detail

Experimental Setup

X-ray Free Electron Laser: LCLS

LCLS @ SLAC Linear Accelerator Laboratory
- > 3 km long building
- Electrons are accelerated
- Undulators create photons
- Pulsed and highly focused beam
- Large number of photons per pulse:
  - $10^{11}$ per pulse of 10 fs