Stimulated X-ray Emission Spectroscopy with hard X-rays

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X-Ray Emission Spectroscopy

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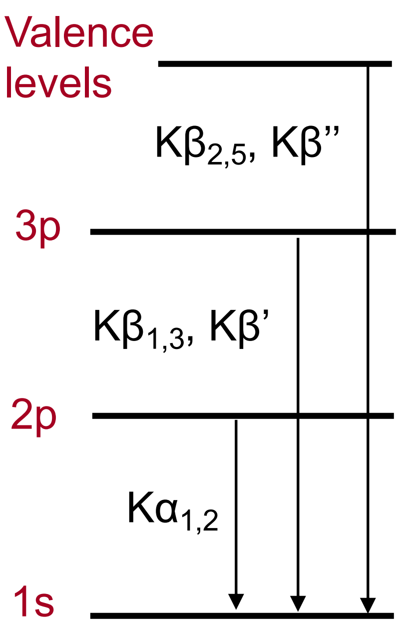
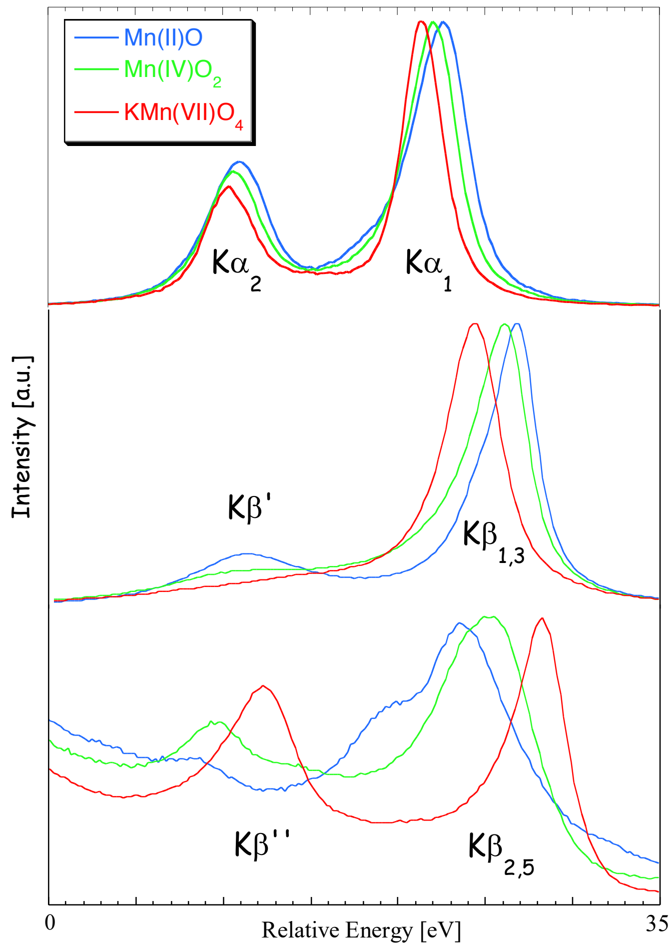
Level Diagram





Spin/oxidation state of transition metals

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

Bergmann et al, *J Synchr Rad*. 8, 199 (2001)

Stimulated X-ray Emission Spectroscopy

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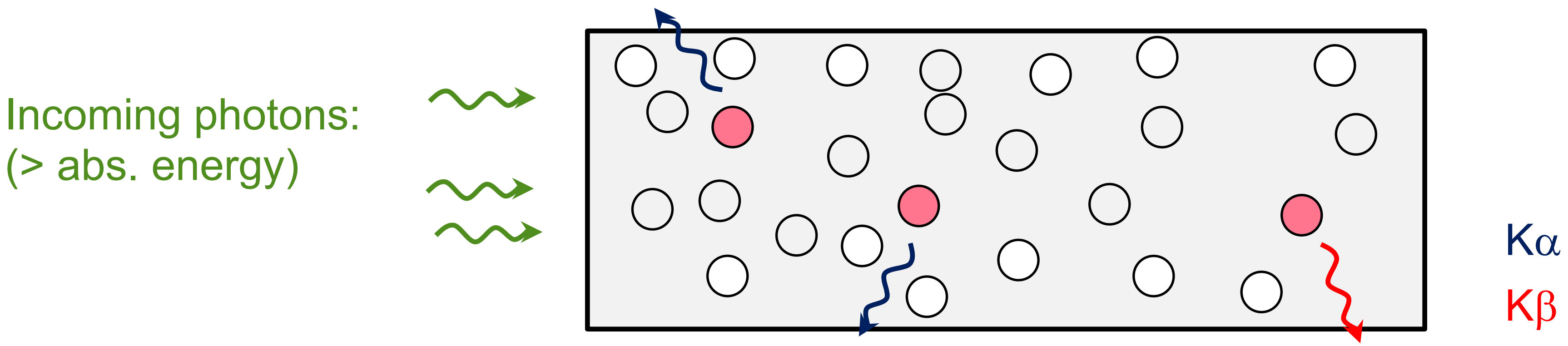
Stimulated (non-linear) emission:

Spontaneous (conventional) emission



- Low photon flux density (only a few photons at a time in the sample)
- Only spontaneous emission in 4π direction

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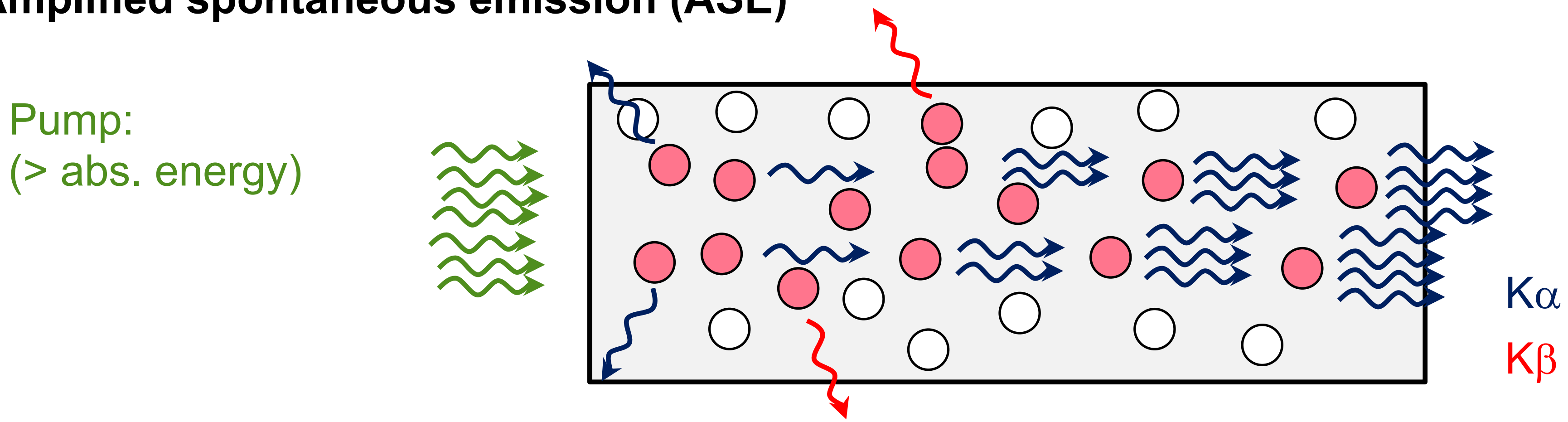
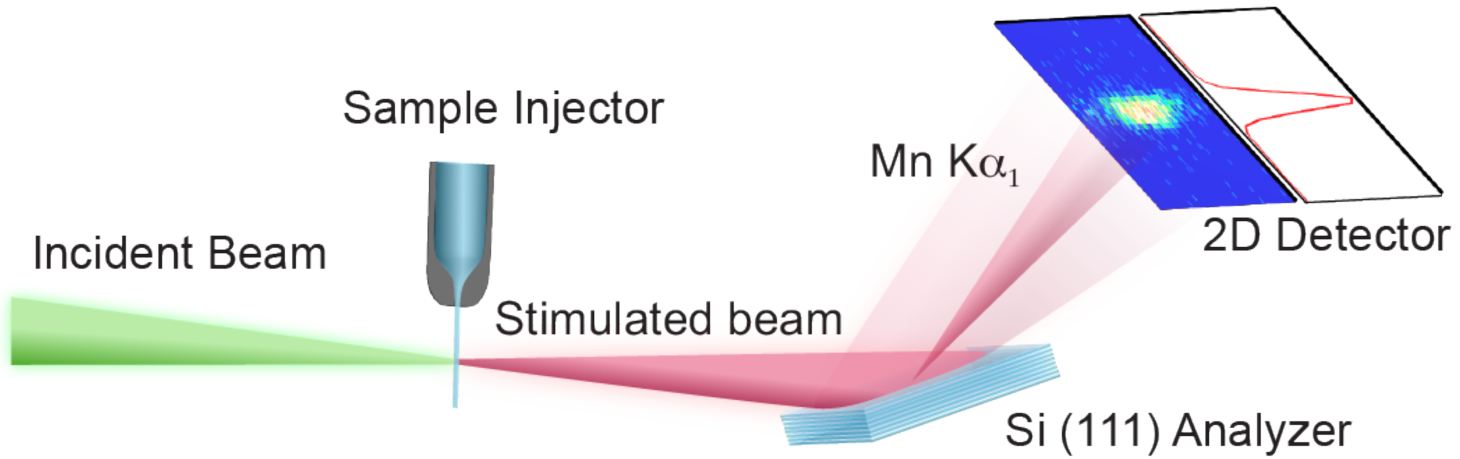
Stimulated (non-linear) emission:

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

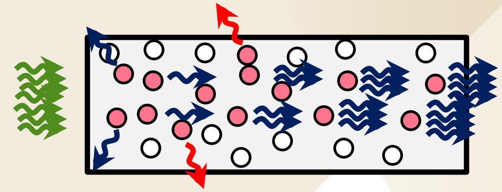


- Highly focused 10 fs beam (100 x 150 nm^2)
- Flat crystal analyzer in forward direction

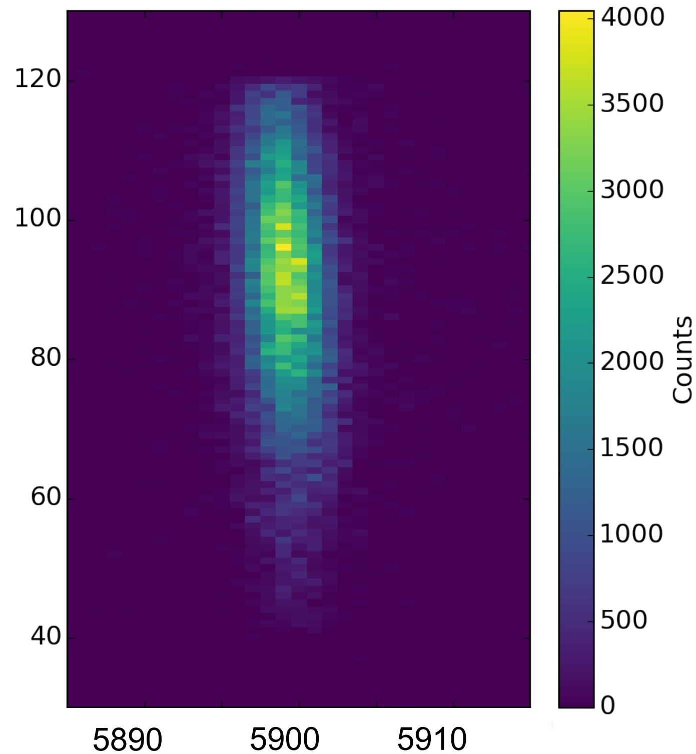
Objectives

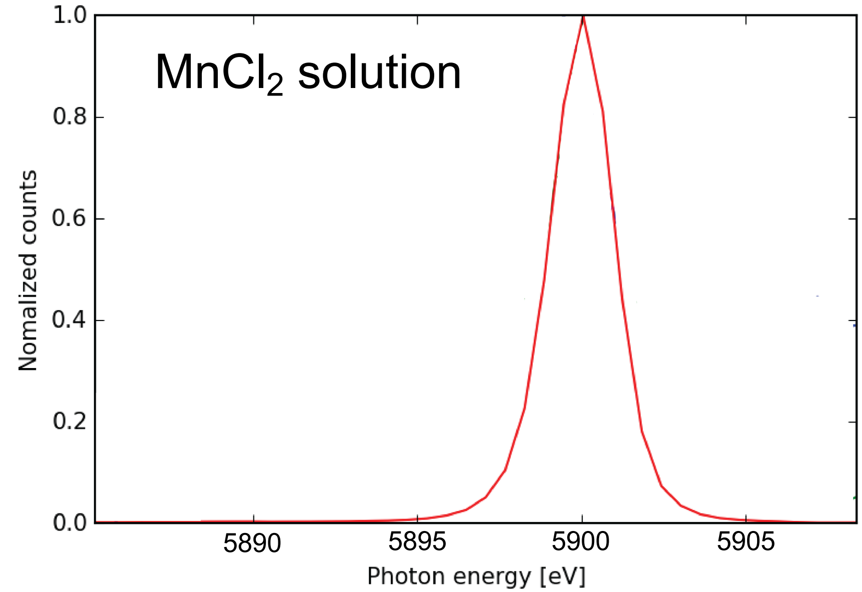
- Can stimulated XES be applied to transition metal solutions?
- What is fundamentally new information provided by stimulated XES techniques that is not achievable with other techniques?
- Can stimulated XES be used for chemically sensitive X-ray emission lines? Is the chemical information preserved?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

Single shot spectra (Kα-ASE)



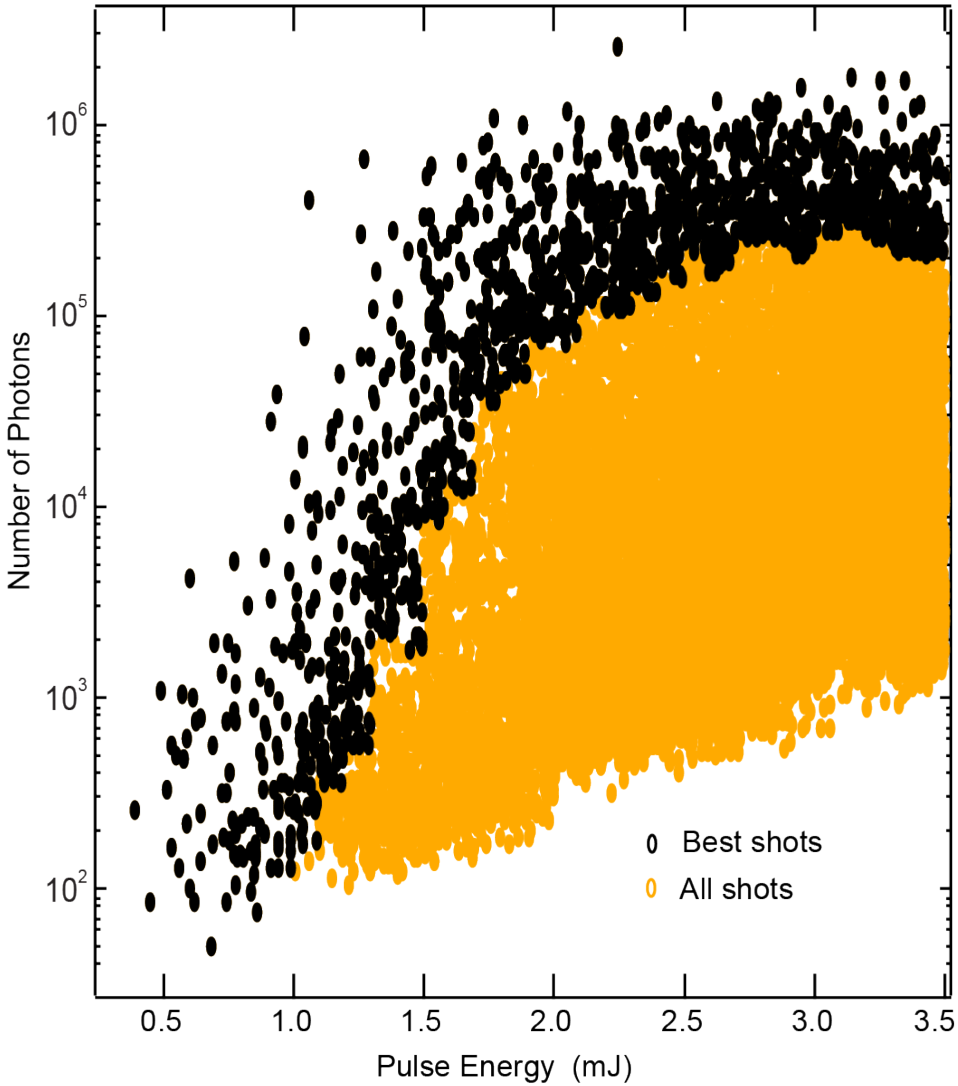
Single shots:

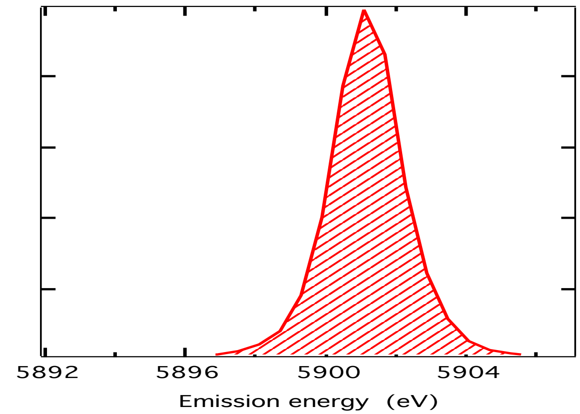




- Clean curve in single shot
- Only the Kα_1 visible
- FWHM < normal Kα width

Lasing in $MnCl_2$ solution



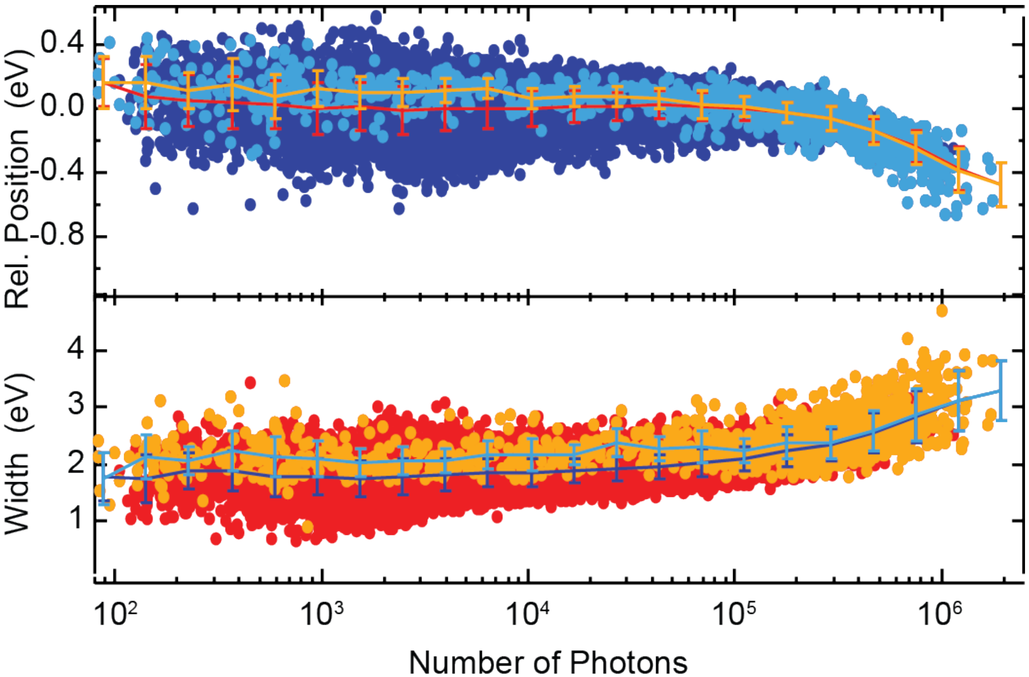


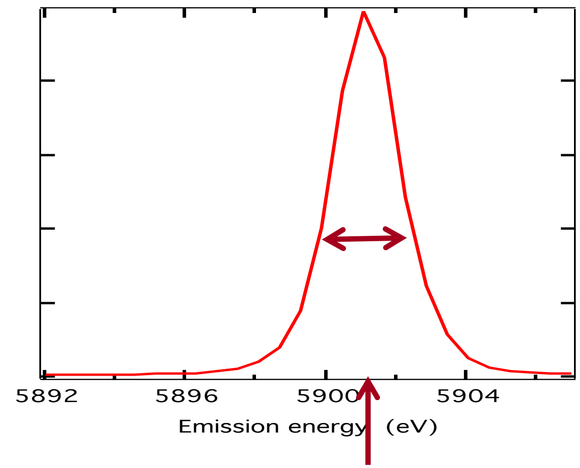
Proof of Lasing:

- Exponential increase
- Linear gain regime below ~2 mJ
- Saturation reached > 2 mJ

T. Kroll et al. Phys. Rev. Letter 120, 133203 (2018)

Peak Position and Width

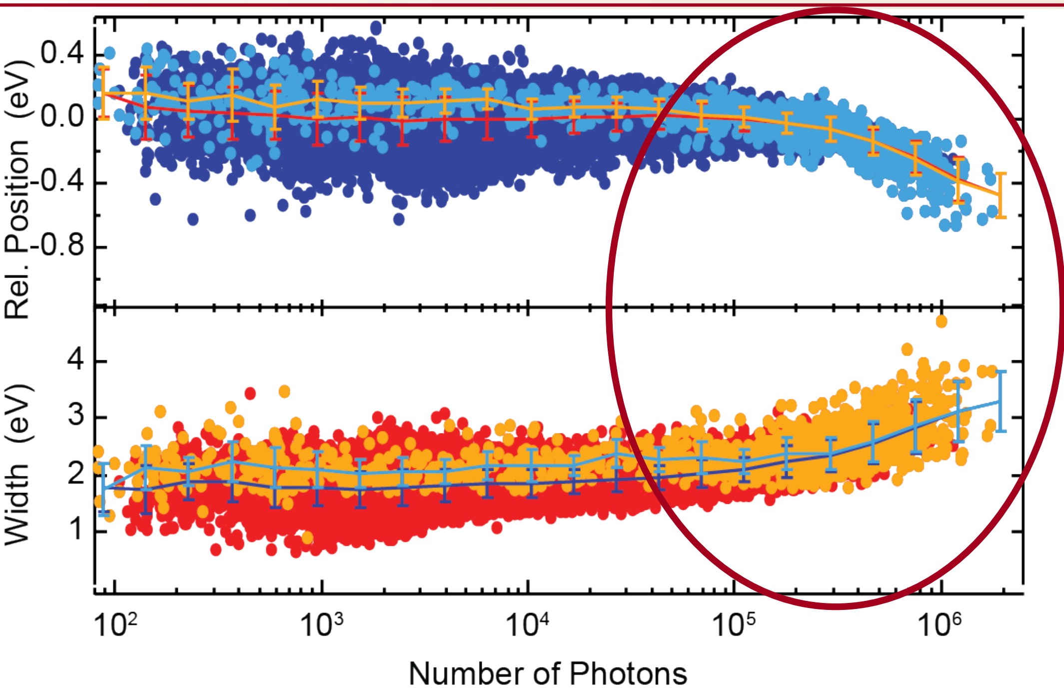


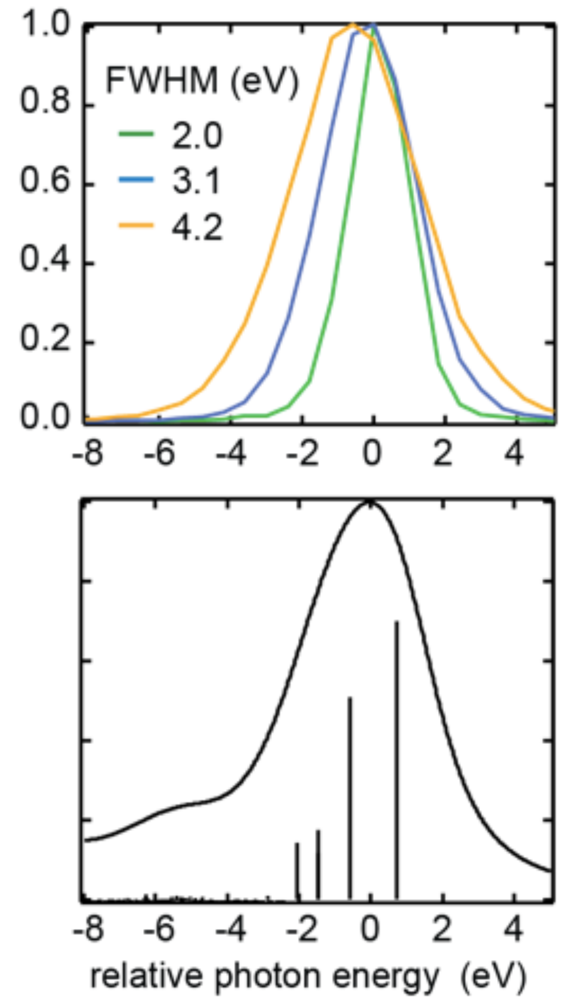


- Low to mid high photon numbers:
  - Constant broadening and position
- High photon numbers:
  - Spectral broadening
  - Shift to lower energies
- Variations through beam position, temporal shape, lasing condition

T. Kroll et al. Phys. Rev. Letter 120, 133203 (2018)

Peak Position and Width

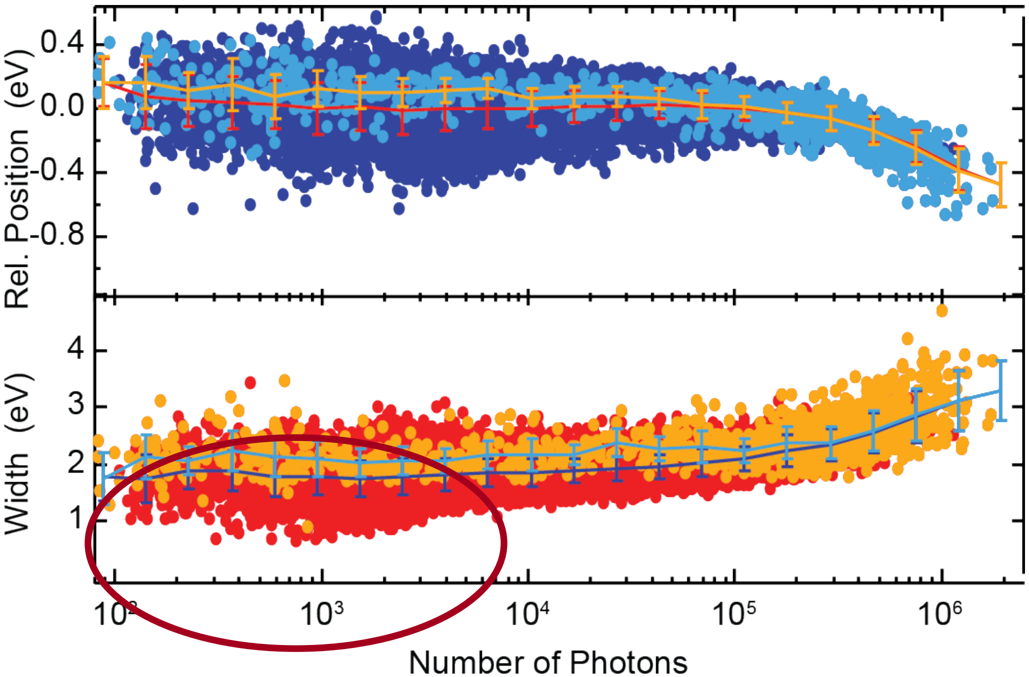


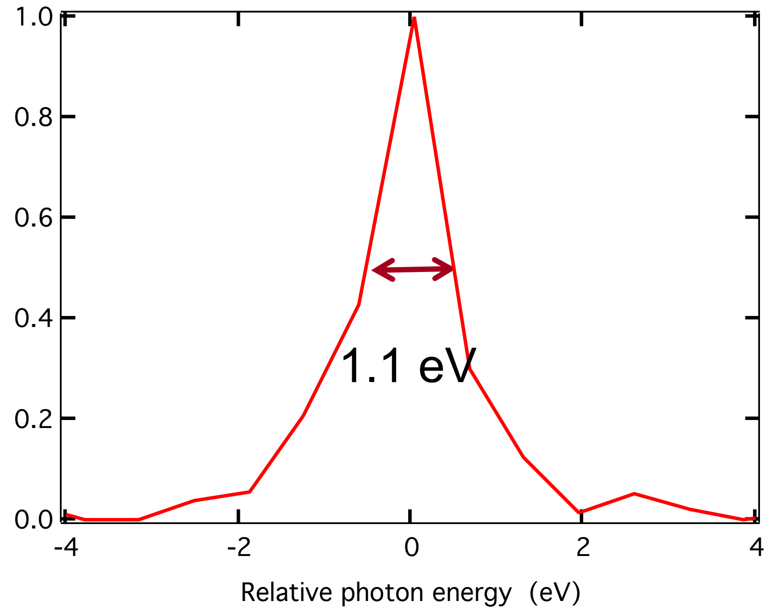


- Line broadening due to saturation effects
- Additional final states become visible

T. Kroll et al. Phys. Rev. Letter 120, 133203 (2018)

Peak Position and Width





- Mn Kα_1 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

T. Kroll et al. Phys. Rev. Letter 120, 133203 (2018)

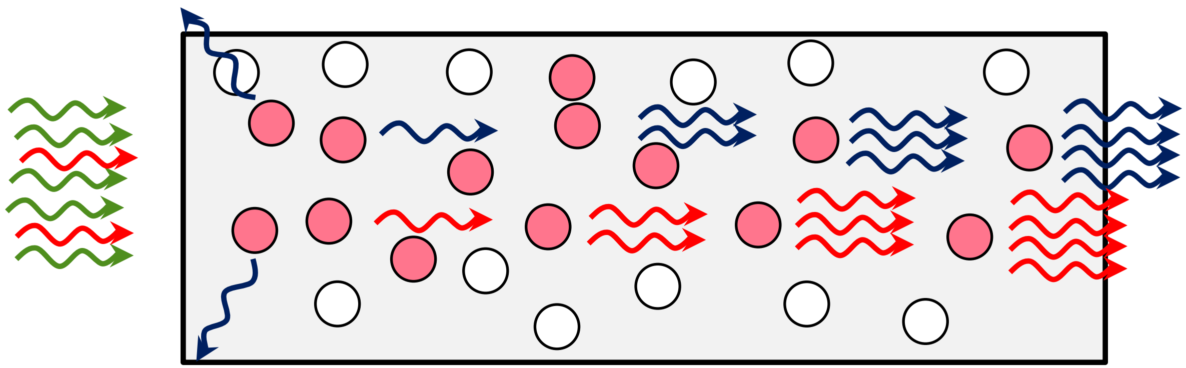
Stimulated X-ray Emission Spectroscopy

Stimulated (non-linear) emission:

Amplified spontaneous emission (ASE) + seeded stimulated emission

Pump:
> abs. energy

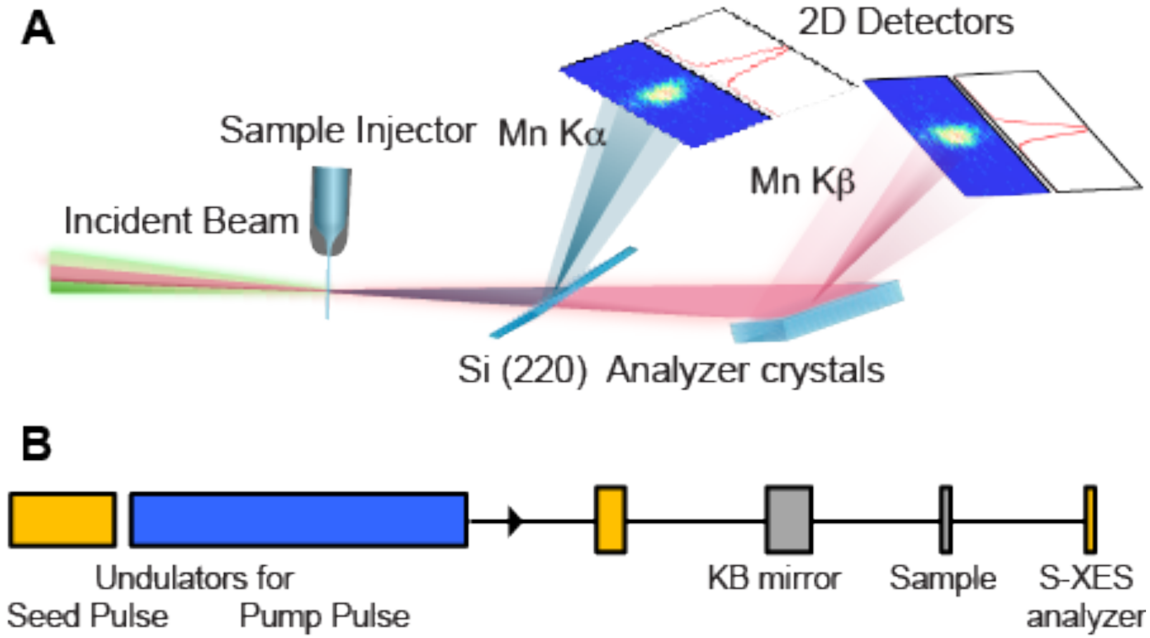
Seed:
Kβ energy



Kα
Kβ

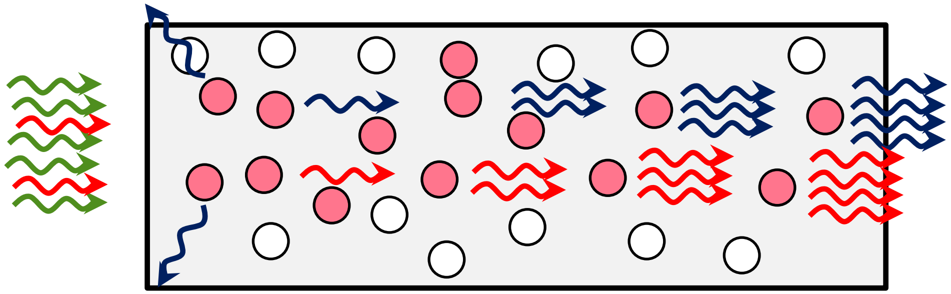
- Kβ Seed pulse outruns Kα emission → seeded stimulated Kβ emission
- Requirement: Overlap in time, space and energy

Experimental Setup



- Highly focused 10 fs beam (100 x 150 nm^2)
- Use part of the undulators for seed pulse
- Two flat crystals in series in forward direction for simultaneous detection of $K\alpha$ and $K\beta$

Seeded Stimulated Kβ emission



Pump:
> abs. energy

Seed:
Kβ energy

Biggest issue:

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

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

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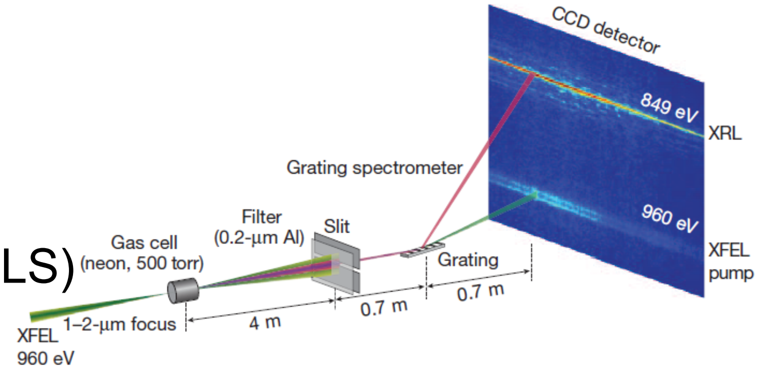
Past achievements:

Soft X-rays

- Stimulated soft X-ray Ne laser

  (Rohringer et al, *Nature* **481**, 488 (2012), LCLS)

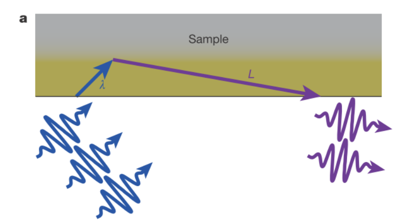
  - Ne gas



- Stimulated X-ray Emission in Silicon L Lines (70-100 eV)

  (Beye et al, *Nature* **501**, 191 (2013), LCLS)

  - Si

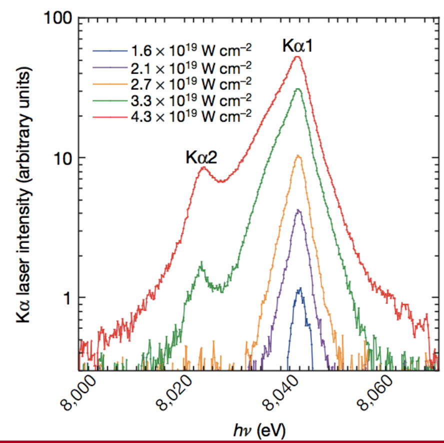


Hard X-rays

- Stimulated hard X-ray Fe laser

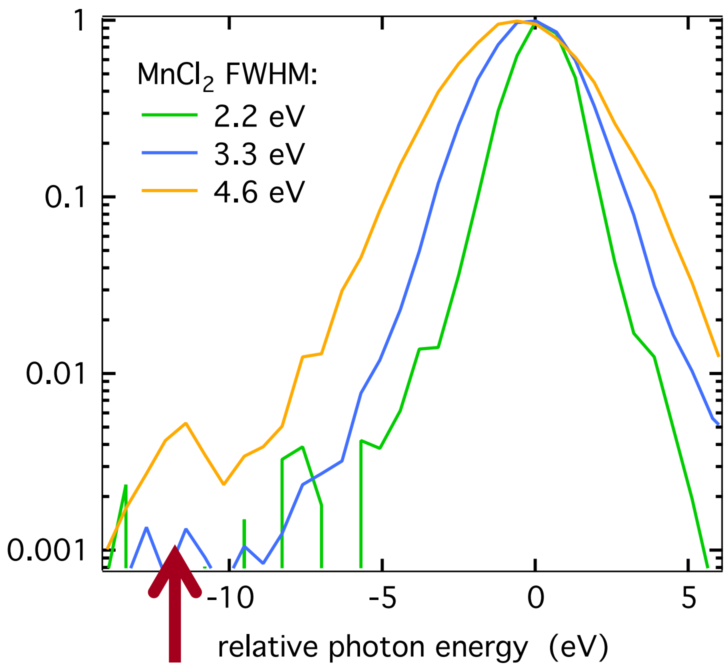
  (Yoneda et al., *Nature* **524**, 446 (2015), SACLA)

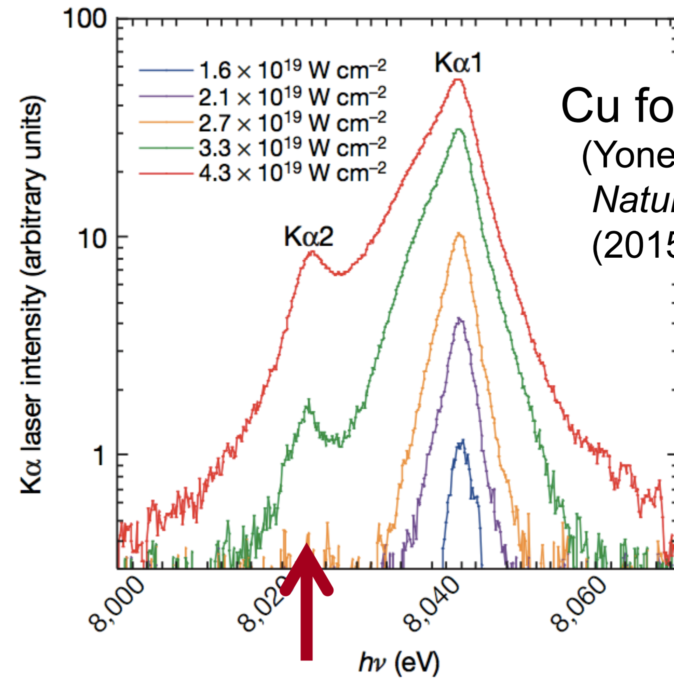
  - Fe foil



High number of photons:

Varying pulse energy:





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

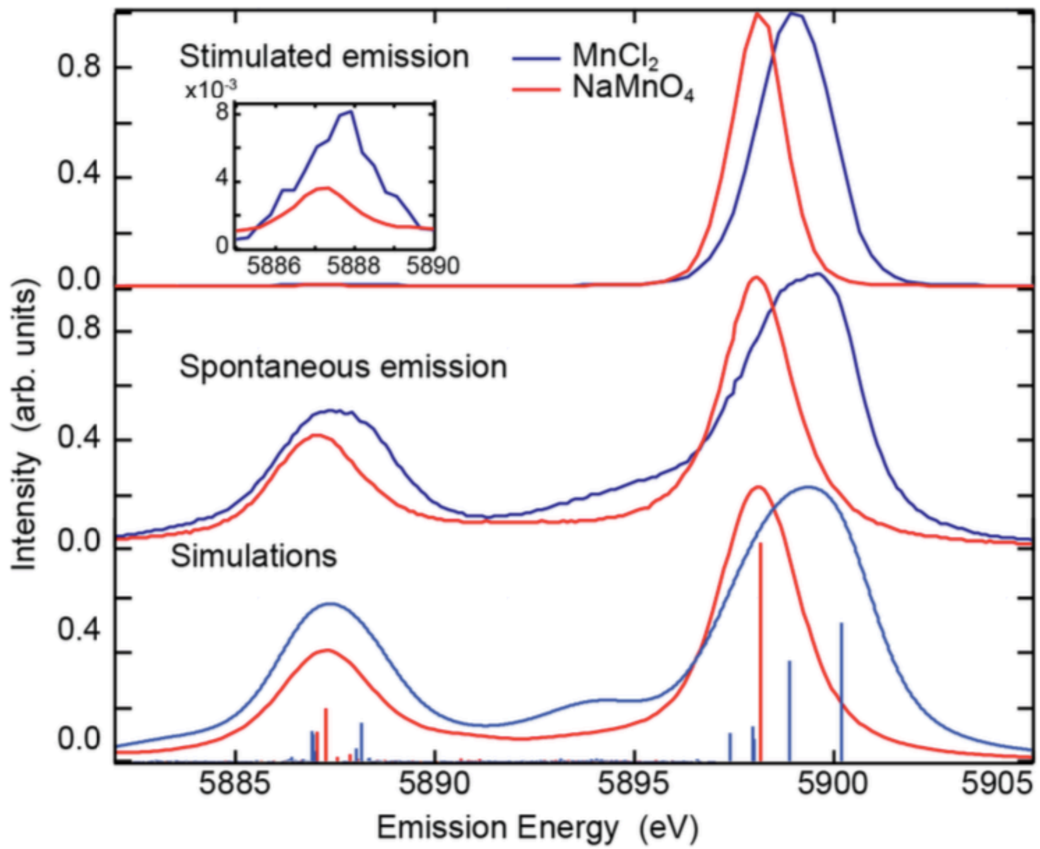
- Increase of Kα_2 for high fluences
- Can be explained by final state effects

T. Kroll et al. Phys. Rev. Letter 120, 133203 (2018)

Chemical Sensitivity

Compare solution spectra:

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



T. Kroll et al. Phys. Rev. Letter 120, 133203 (2018)

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