

# X-Ray Lasers



**Nina Rohringer**

21st meeting on Atomic Processes in Plasmas

HELMHOLTZ

DASHH

MAX  
PLANCK  
SCHOOL  
of  
photonics

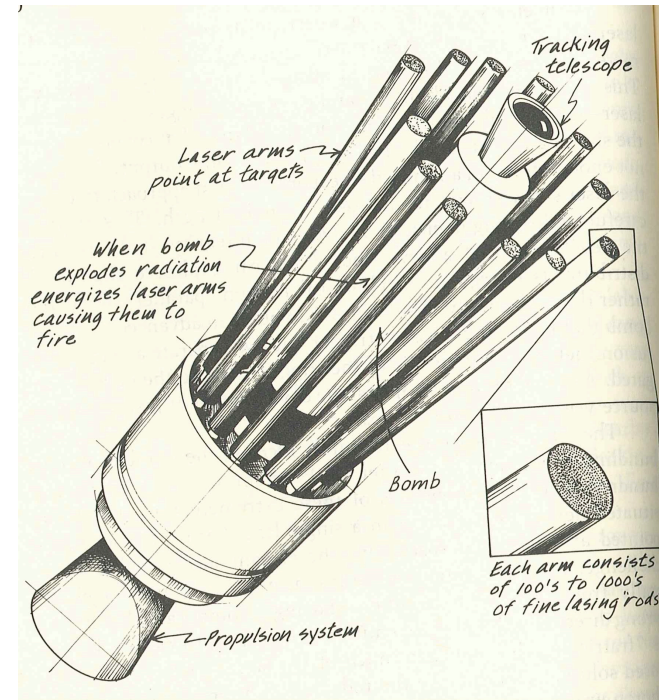
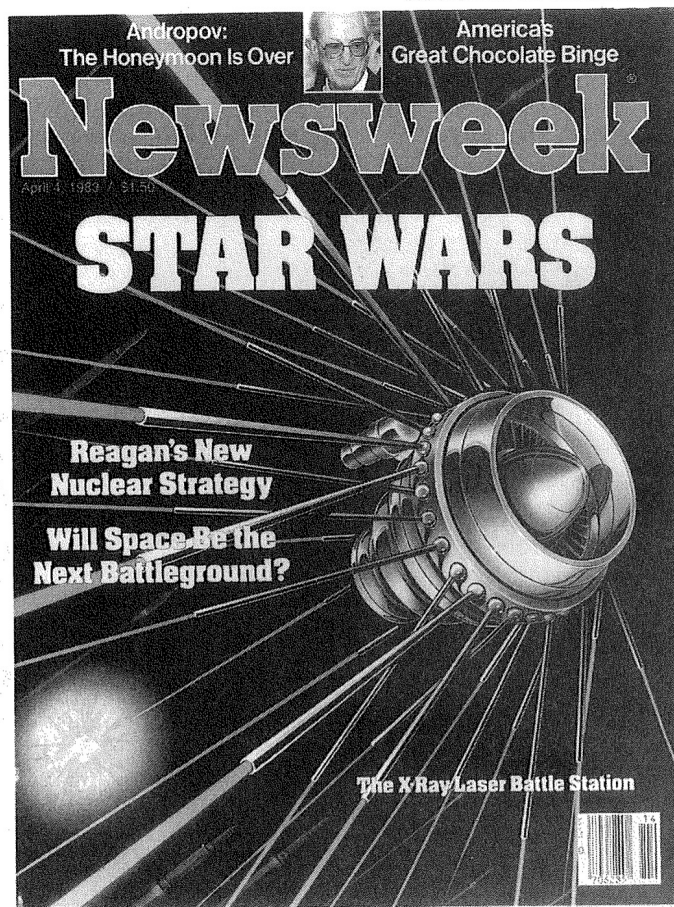
CFEL  
SCIENCE

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Universität Hamburg  
DER FORSCHUNG | DER LEHRE | DER BILDUNG



# US Strategic Defense Initiative (1984-1993)

## Project Excalibur: Nuclear-explosion pumped XRL for ballistic missile defense



William J. Broad,  
"Teller's War, The top-secret story  
behind the star wars deception"  
(Simon & Schuster, New York, 1992)

D. M. Ritson, Nature 328, 487 (1987)

N. Bloembergen et al., Rev. Mod. Phys. 59,1 (1987) 3.7 Milliarden \$ (1985-1991)

# X-ray lasers pumped by nuclear explosion

List of (presumably) performed experiment from 1978 to 1988

Code Name	Date	Result
Diablo Hawk	Sep. 13, 1978	Test apparatus fails
Dauphin	Nov. 14, 1980	1 <sup>st</sup> probable x-ray laser, though some experts say evidence is sketchy
Cabra	March 26, 1983	Sensors fail
Romano	De. 16, 1983	1 <sup>st</sup> hard X-ray laser evidence
Correo	Aug. 2, 1984	Laser fails
Cottage	March 23, 1985	1 <sup>st</sup> focusing attempt
Goldstone	Dec. 28, 1985	1 <sup>st</sup> good measure of brightness shows basic laser is dimmer than previously believed
Labquark	Sept. 30, 1986	More focusing tests
Delamar	Apr. 18, 1987	1 <sup>st</sup> fear that focusing has failed
Kenville	Feb. 15, 1988	1 <sup>st</sup> high-quality data on basic laser

# Pumping power requirements for x-ray lasers

Population inversion:  $\Delta N = \left( N_2 - N_1 \frac{g_2}{g_1} \right)$   $I(z) = I(0) \cdot e^{g \cdot z}$

Gain coefficient:  $g = \sigma \Delta N \cong \frac{A_{21} \lambda^2}{8 \pi \Delta \nu} \left( N_2 - N_1 \frac{g_2}{g_1} \right)$

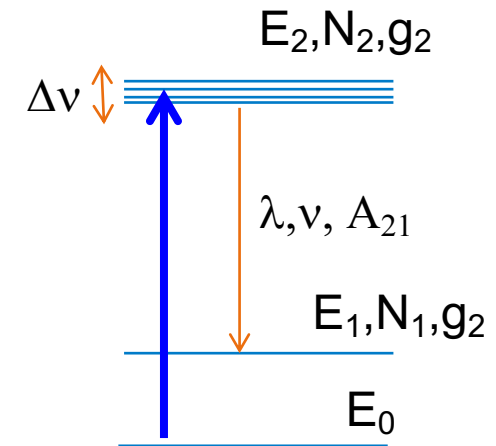
Einstein A coefficient:  $A_{21} \propto \lambda^{-2}$

Naturally broadened transition (rad. decay):  $\Delta \nu \propto A_{21}$

$\rightarrow g \propto \Delta N \lambda^2 \cong N_2 \lambda^2$

Required pump power density to compensate for level depletion:

$\rightarrow P = N_2 A_{21} \frac{hc}{\lambda} \propto N_2 \lambda^{-3}$



Pumping power to maintain a specific gain:  $P \propto g \lambda^{-5}$

# Pumping power requirements for x-ray lasers

Pump power to achieve a given gain  $g$  scales as:

$$P \propto g \lambda^{-5}$$

$$\frac{P_{x\text{-ray}}}{P_{\text{optical}}} = \left( \frac{\lambda_{\text{optical}}}{\lambda_{x\text{-ray}}} \right)^5 = \left( \frac{800 \text{ nm}}{1 \text{ nm}} \right)^5 = 3.3 \times 10^{14}$$

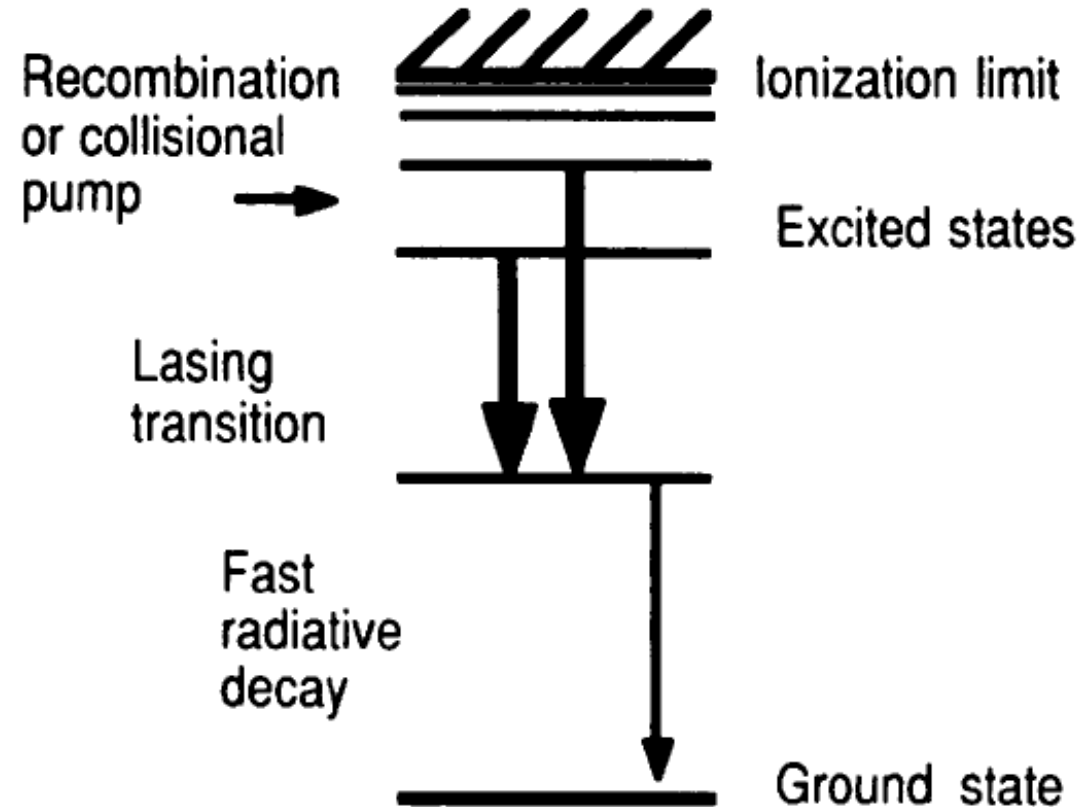
Optical Laser: 10 W            X-ray Laser :  $10^{15}$  W

Electric power production in Germany 2022 : (Source Statistisches Bundesamt).

**500 TWh =  $5 \times 10^{14}$  Wh**

# Mechanisms for population inversion on x-ray transitions

## Recombination or collisional pump

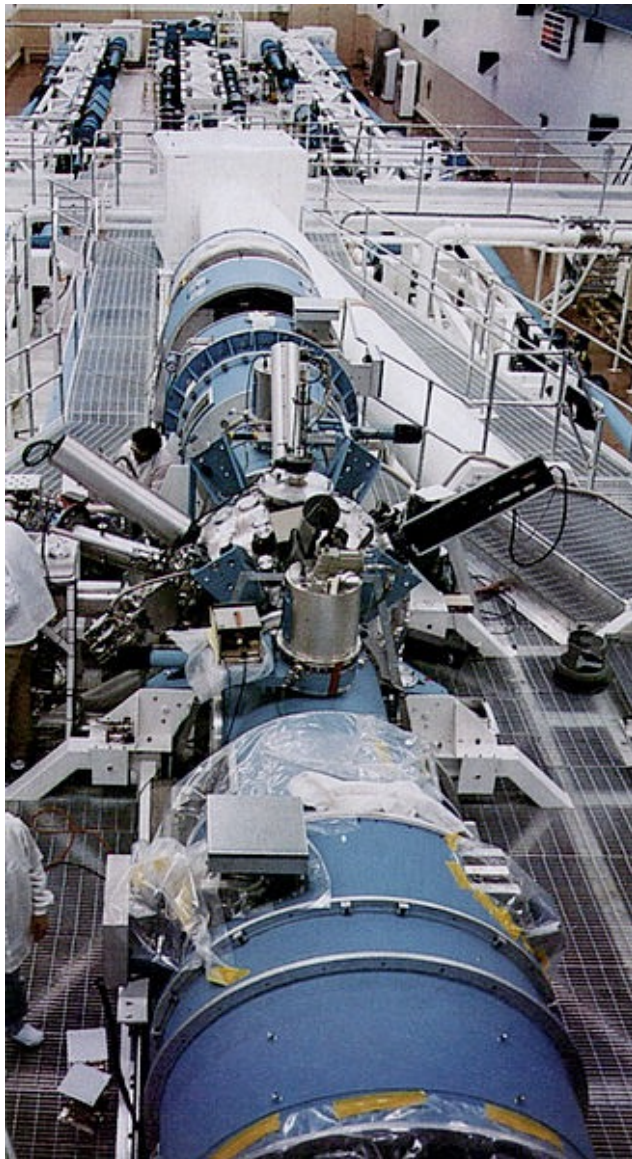


Create hot plasma, of high ionization degree

Electron temperatures:  $\sim$  keV

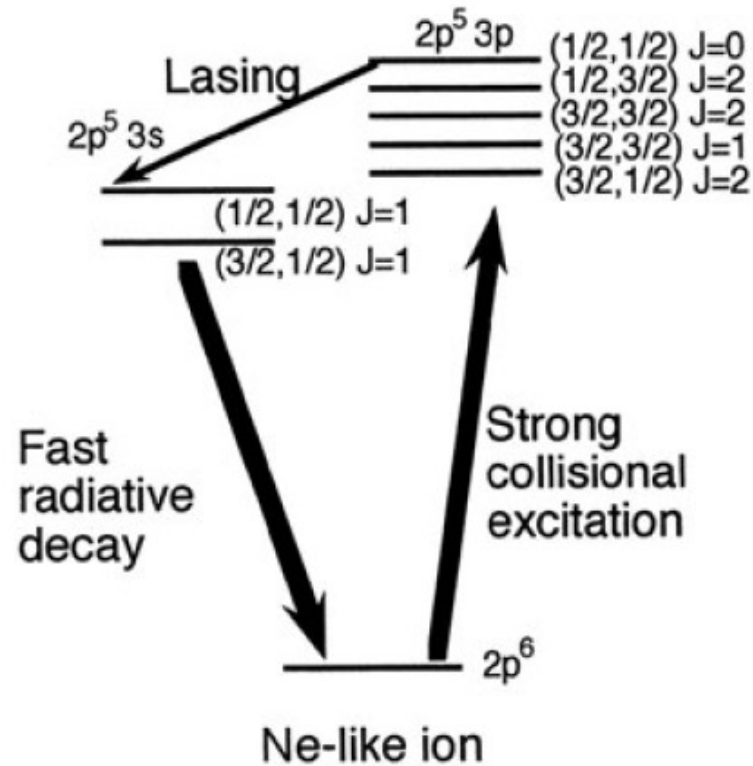
Densities:  $10^{21} - 10^{22} \text{ cm}^{-3}$

# 1<sup>st</sup> atomic XUV Laser at Novette Laser in 1984



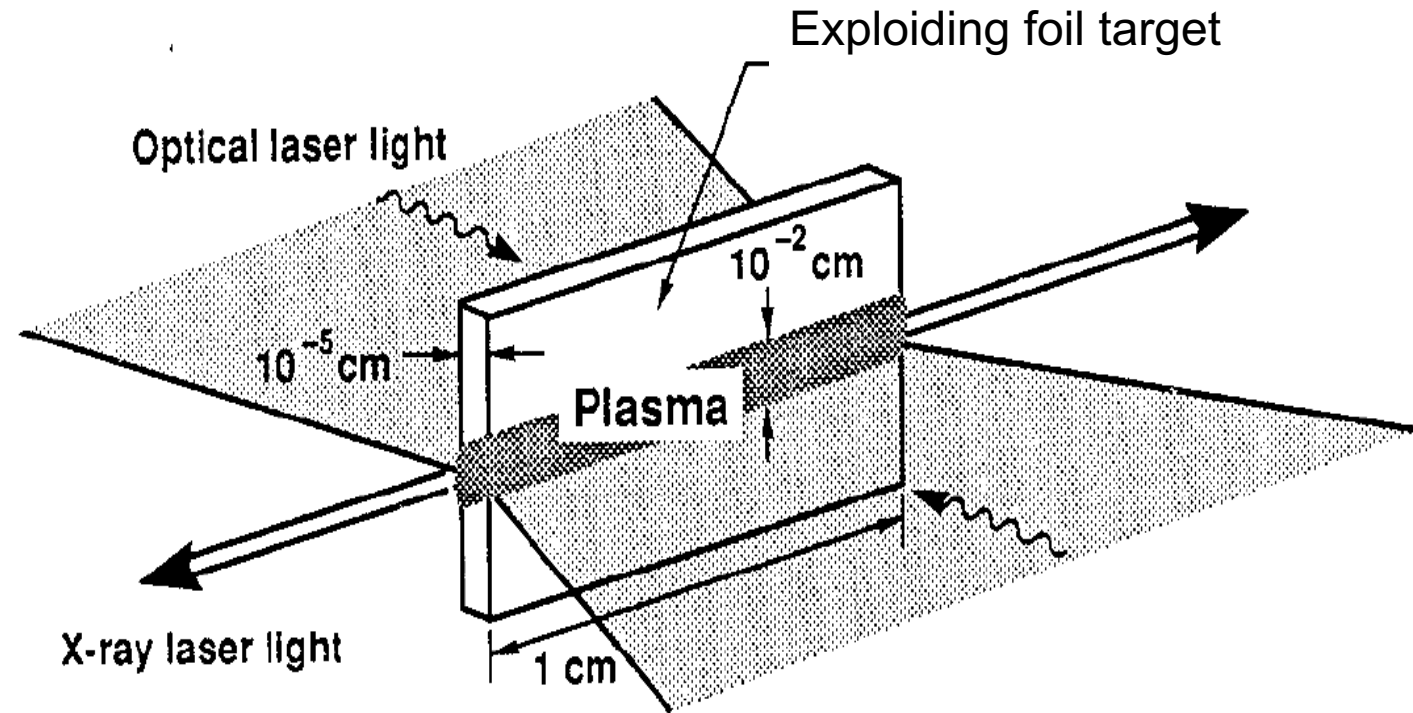
# Soft x-ray laser by electron impact excitation

1<sup>st</sup> experimental realization at LLNL in 1984: Ne-like Se laser 20 nm (60 eV, 20 nm)



Electron temperature:  $T = 1 \text{ keV}$   
 Density:  $N = 1 \times 10^{21} \text{ cm}^{-3}$

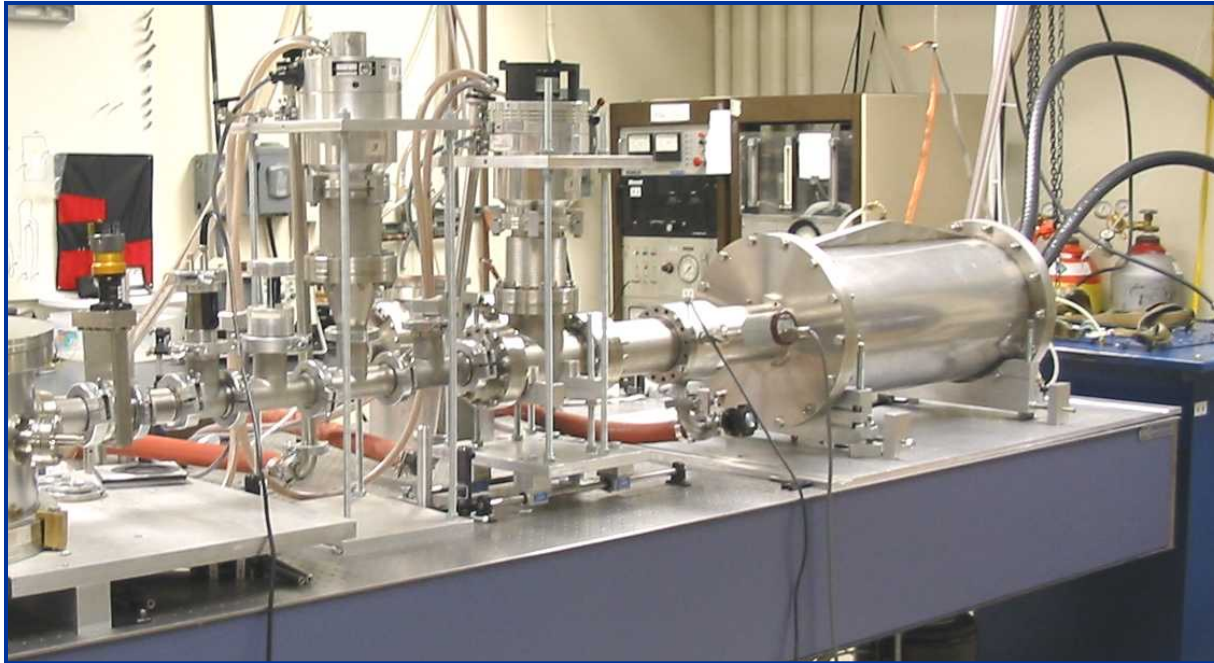
Transverse pumping:  
 Pump Energy 1 kJ  
 Line focus:  $0.02 \times 1.12 \text{ cm}$   
 Pump wavelength: 532 nm  
 Duration: 450 ps  
 Intensities on target:  $5 \times 10^{13} \text{ W/cm}^2$





# Table-top XUV lasers in Ne-like Ar at 46.9 nm (26 eV)

Coherent average power is comparable to synchrotron beam line



(By courtesy of J.J. Rocca,  
Colorado State University)

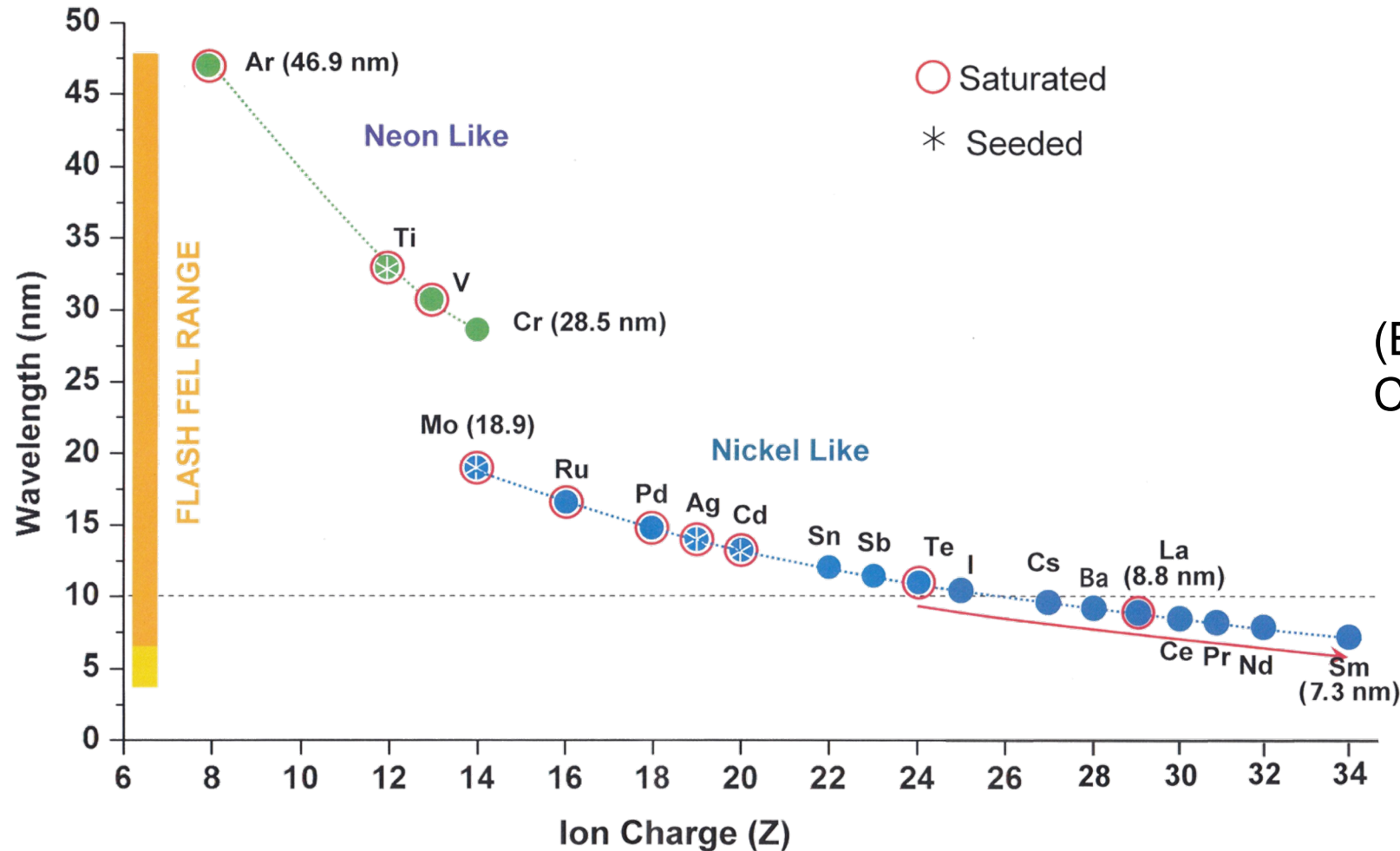
Discharge current pulse:  
37 kA, duration 70 ns

High average power: up to 3 mW  
High pulse energy: 0.1 mJ - 0.8 mJ @4 Hz  
Narrow spectral bandwidth:  $\Delta\lambda/\lambda = 3 \times 10^{-5}$   
Beam divergence:  $\theta = 4.5$  mrad

B. Benware et al. Phys.Rev.Lett. **81**, 5804, (1998)

# Gain-saturated table-top soft X-ray Lasers

Wavelength region from 7.3 nm to 47 nm



(By courtesy of J.J. Rocca, Colorado State University)

Review Articles:

J. J. Rocca, 'Table-top soft x-ray lasers', Rev. Scient. Instr. **70**, 3799 (1999).

S. Suckewer and P. Jaegle, Laser Phys. Lett. **6**, No. 6, 411–436 (2009)

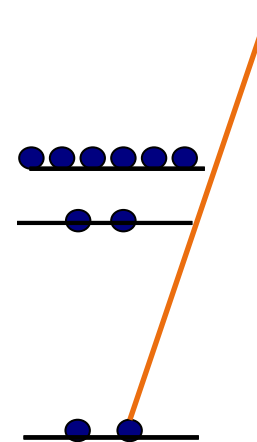
# 1<sup>st</sup> theoretical concept of an atomic inner-shell X-ray laser

## Population inversion by inner-shell photoionization

1<sup>st</sup> X-ray laser proposed back in 1967:

Duguay & Rentzepis,  
Appl. Phys. Lett. **10**, 350 (1967).

1<sup>st</sup> realization in the optical regime (blue laser):  
Silfvast et al., Opt. Lett. **8**, 551 (1983).

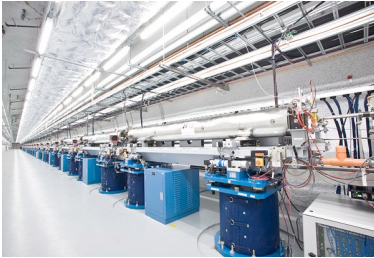


**Ultrafast ionization of  
inner-shell electrons**

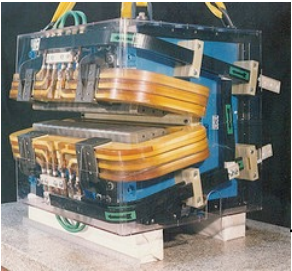
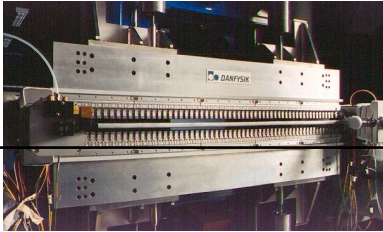
**Fast, powerful x-ray pump required to beat Auger decay !**

# Evolution of accelerator-based x-ray sources over time

Free-electron Lasers

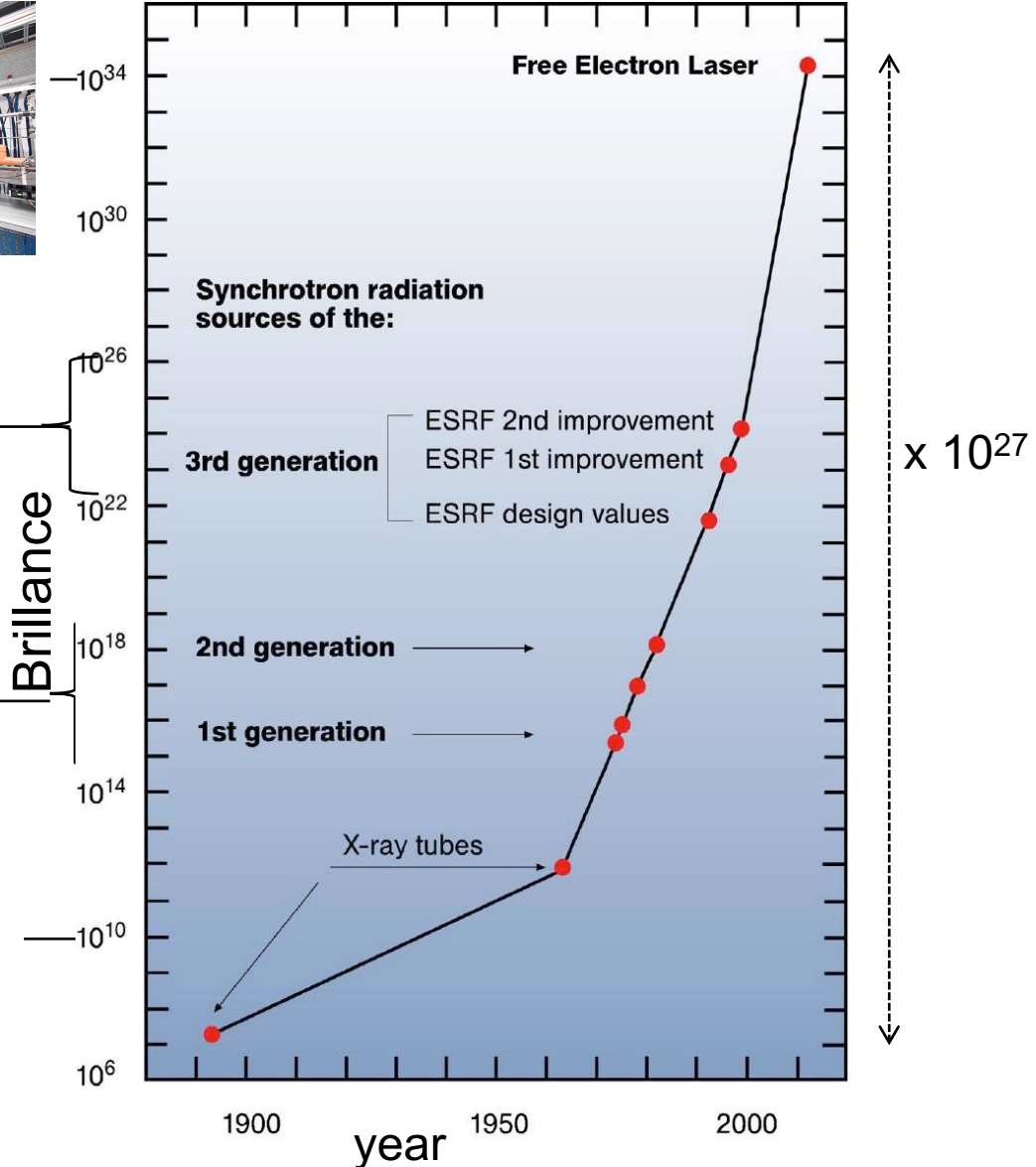


Undulators



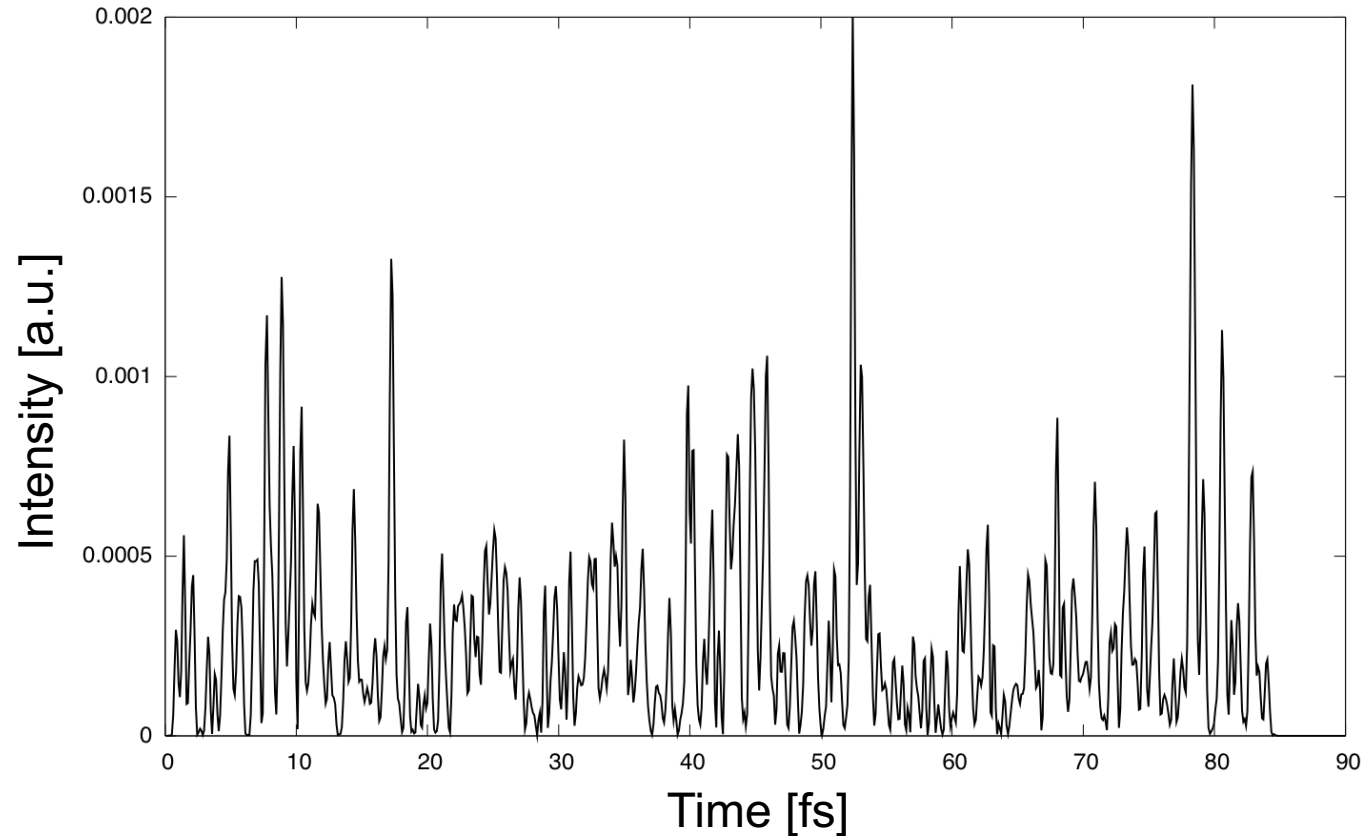
Bending magnets

X-ray tubes



# Self Amplified Spontaneous Emission (SASE)

SASE XFELs have limited temporal coherence



Achievable intensities through focusing:

$10^{18}$  W/cm<sup>2</sup> @ 1 nm

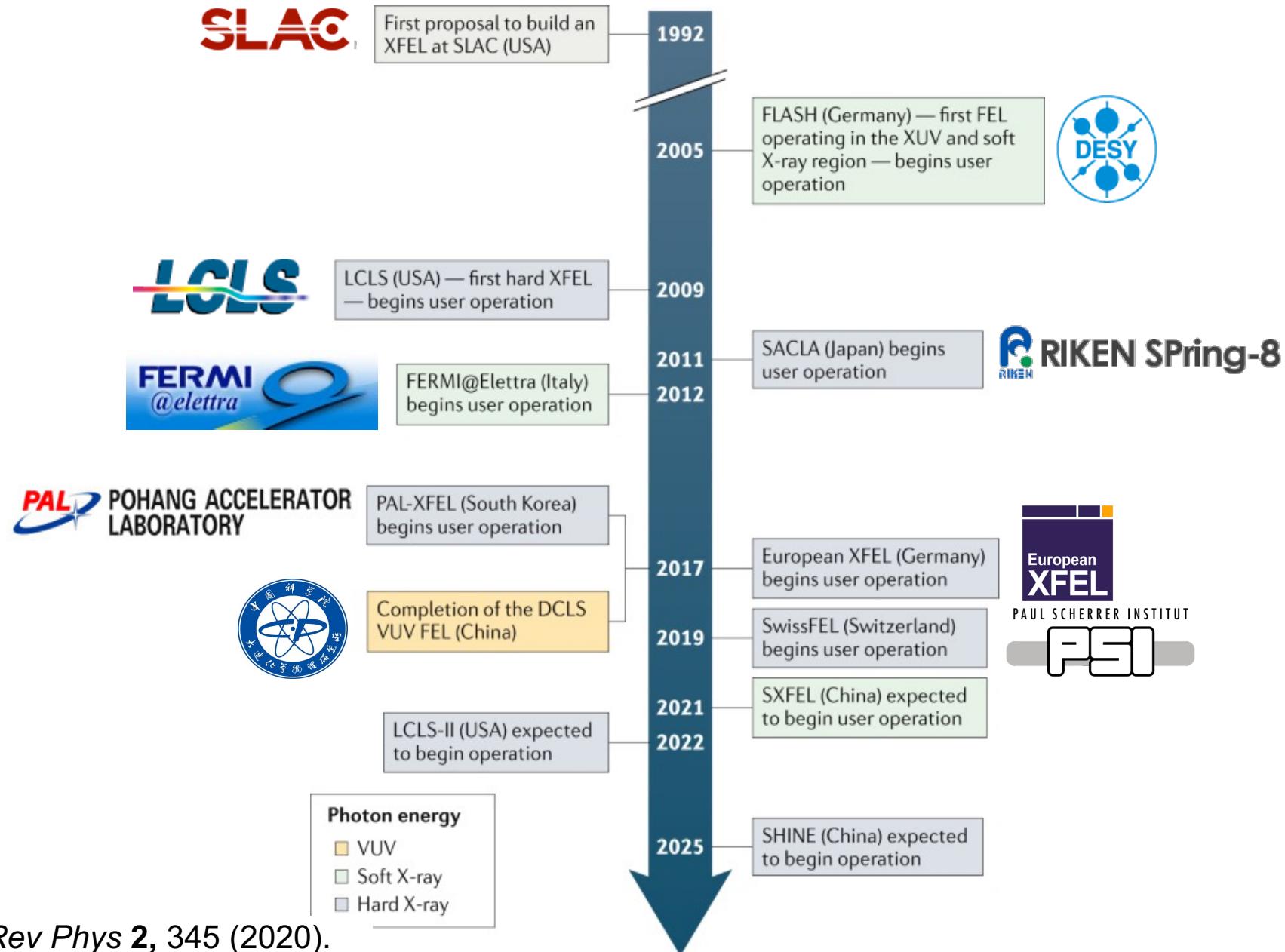
$10^{20}$  W/cm<sup>2</sup> @ 0,1 nm

bandwidth @ 1keV photon energy:  $\Delta\omega = 6-9$  eV

Coherence time: 0.3 - 0.5 fs

Pulse Energy: 1 mJ (for 20 fs pulse duration)

# A new era of coherent, bright x-ray sources



# 1<sup>st</sup> realization of an atomic inner-shell x-ray laser

## Colorado State University:

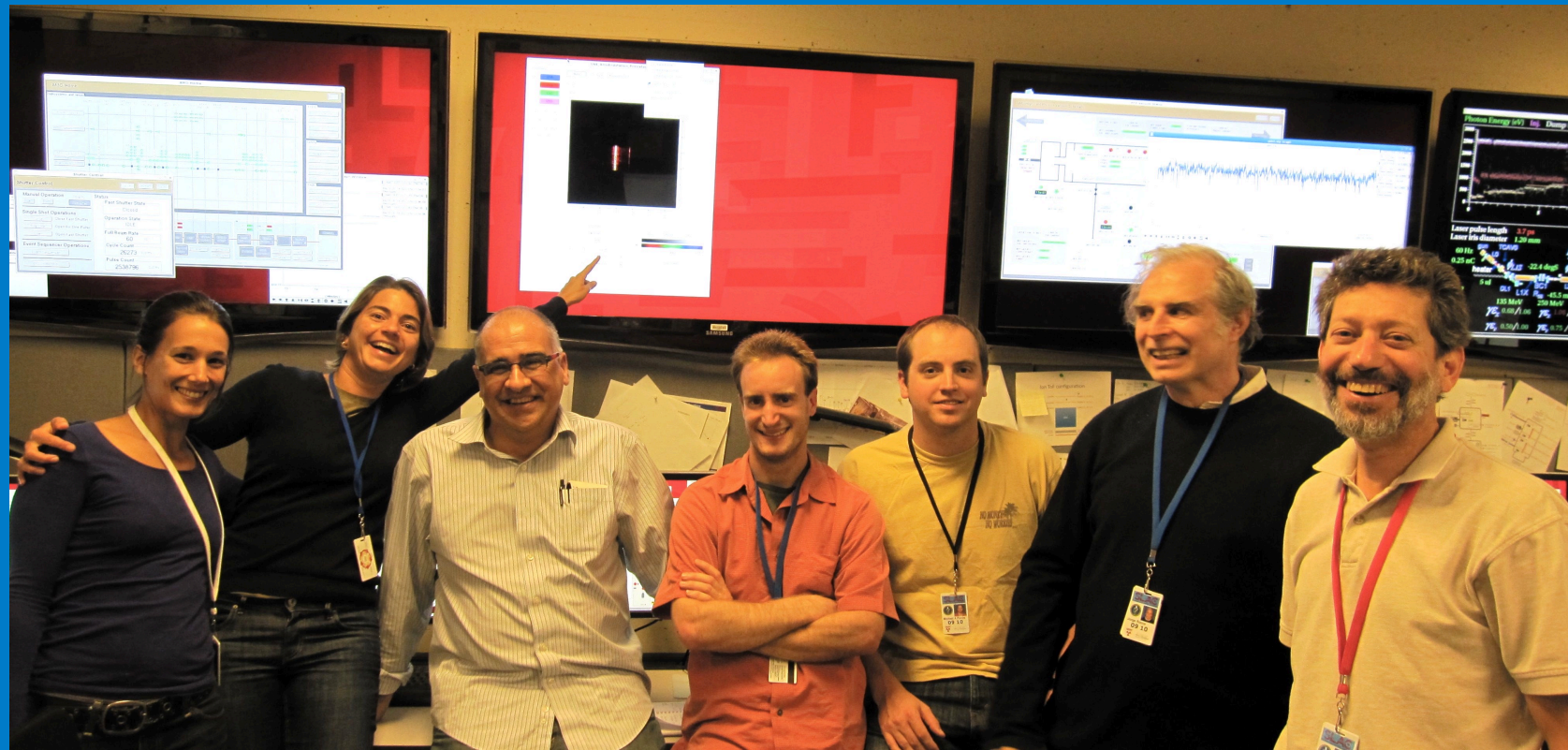
J.J. Rocca, D. Ryan, M. Purvis

## LLNL:

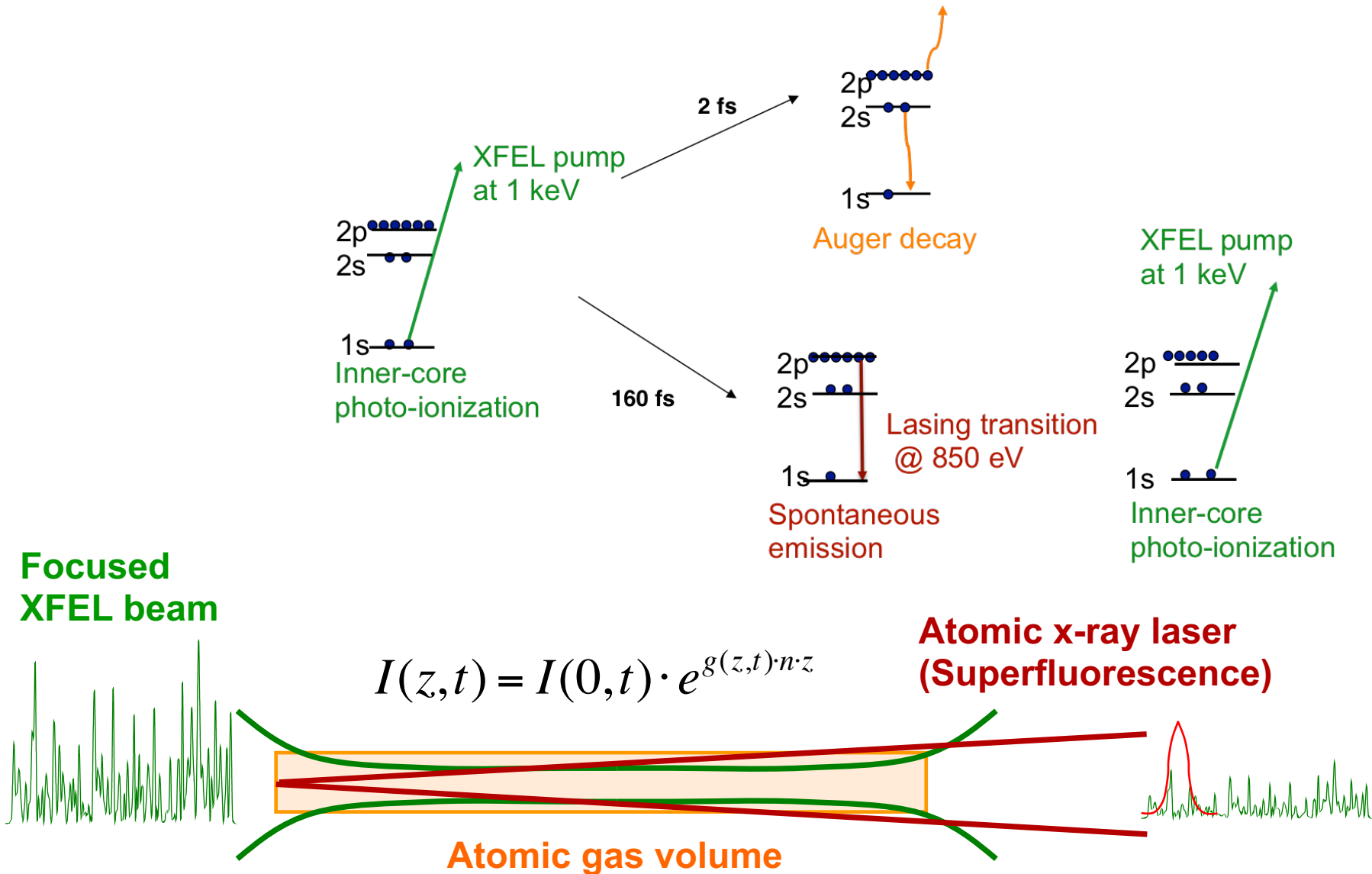
R. London, F. Albert, J. Dunn

## SLAC:

J. Bozek

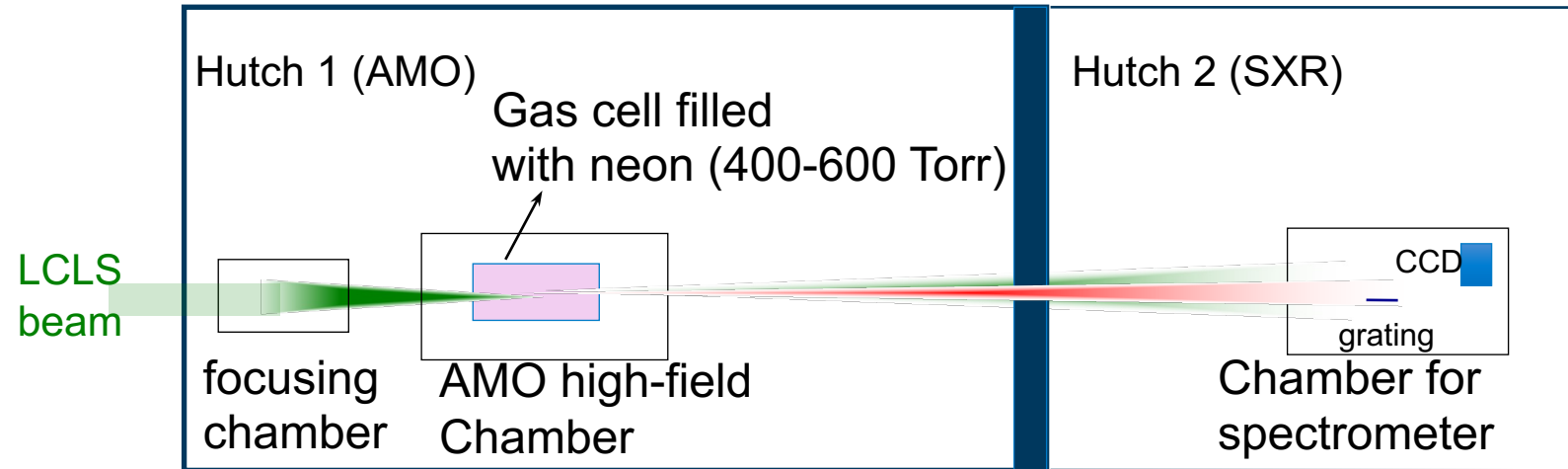


# Photo-ionization inner-shell x-ray laser, Neon



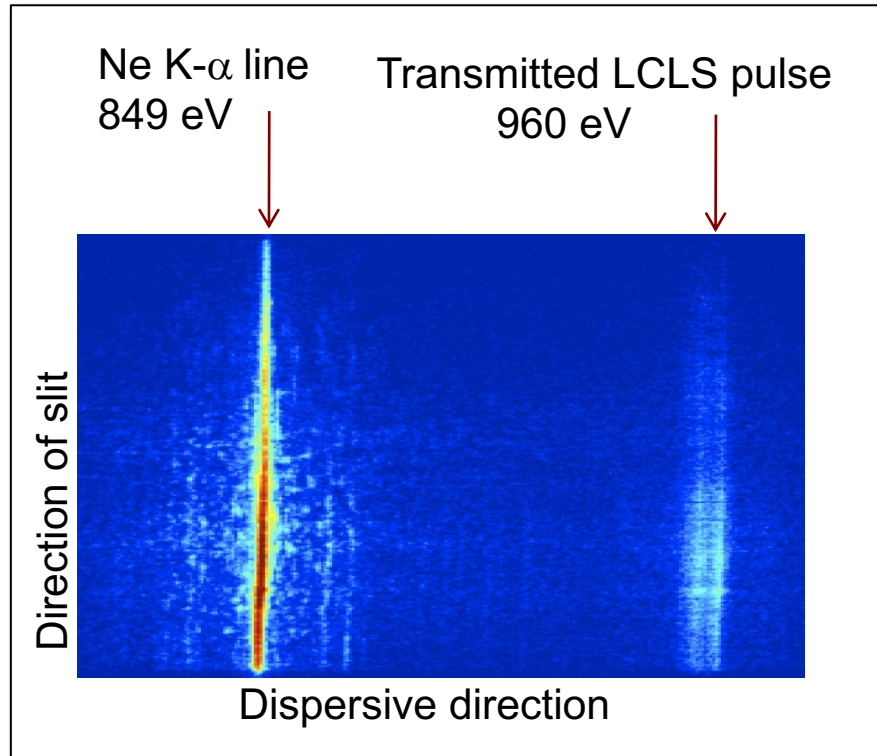


# Experiment at the LCLS XFEL @ SLAC, Sept. 2010



# $8 \times 10^9$ photons in Ne K- $\alpha$ line in a single shot

1,1  $\mu\text{J}$  of energy in K-a line, corresponding to gain-length product of 21-23



$$I(z, t) = I(0, t) \cdot e^{g(z, t) \cdot n \cdot z}$$

$$g(z, t) = n_U(z, t) \sigma_{stim} - n_L(z, t) \sigma_{abs}$$

conversion efficiency:  
 $\approx 4 \times 10^{-3}$

Input:

LCLS pump at 960 eV

pulse energy: 1.4 mJ (0.25 mJ on target)

focus diameter:  $\approx 4$  micron

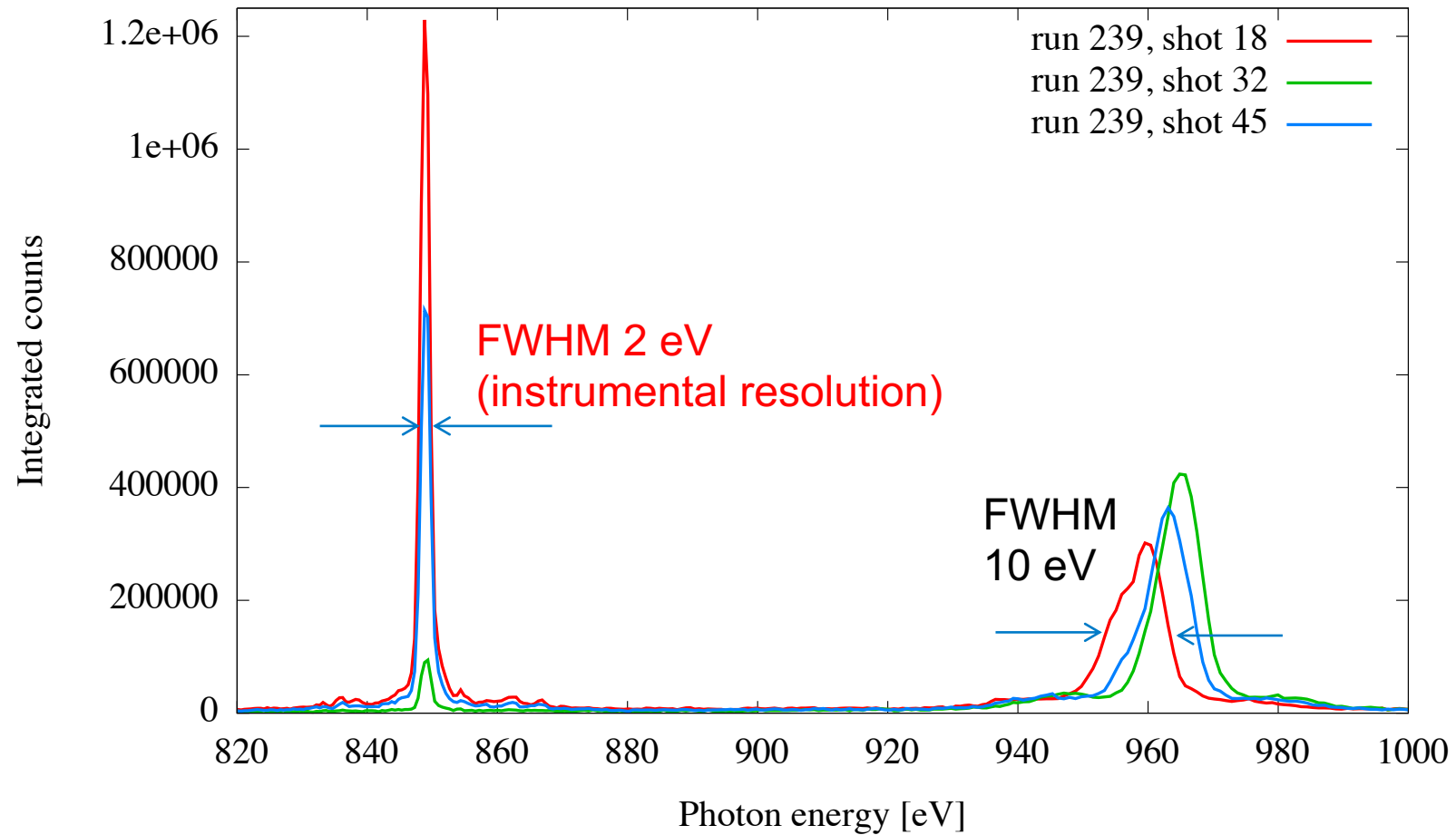
Pulse duration: 40 fs

Gas pressure: 500Torr

Interaction length: 1.6 cm

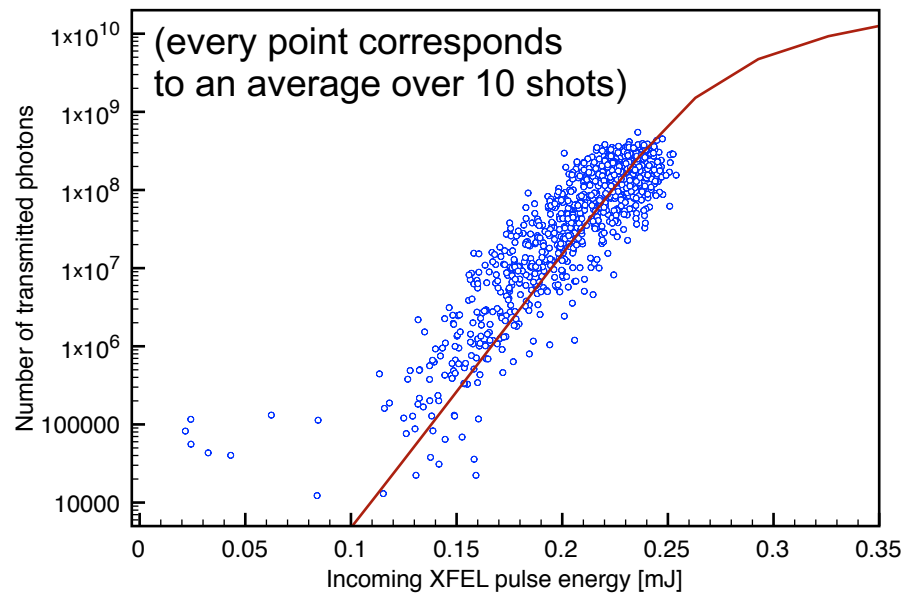
# Integrated spectrum for three sample shots

Stable spectrum, but high pulse-energy fluctuations from pulse to pulse



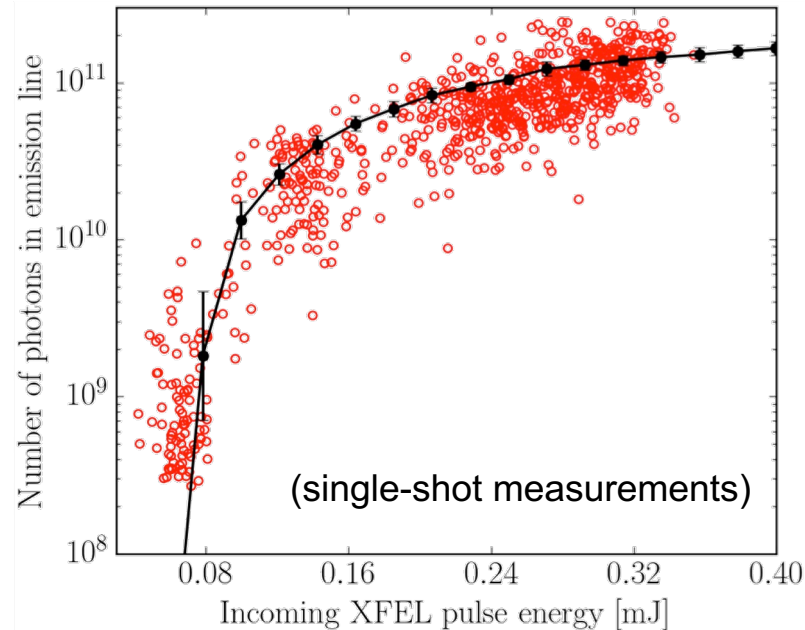
# Pumping-power dependence of Ne K- $\alpha$ transition

High gain-length products of 19-21 @ 0,25 mJ FEL pulse energy



1st experiment, Sept. 2010

Saturation of the amplification,  
> 10<sup>11</sup> detected photons per pulse



2nd experiment, Aug. 2011

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

Weninger et al., *Phys. Rev. Lett.* **111**, 233902 (2013).

# Theory Intermezzo

Coherent x-ray pulses of narrow spectral bandwidth & high intensity



**S. Chuchurka**

A. Benediktovitch  
V. Sukharnikov



**A. Halavanau**



**Š. Krušič**



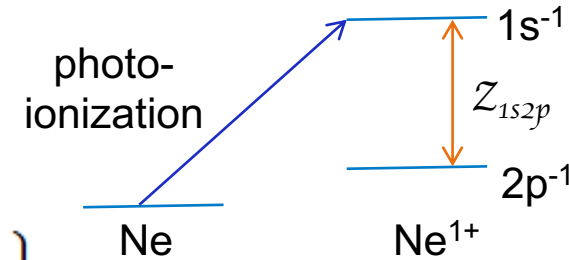
**C. Weninger**

# Maxwell-Bloch approach for open quantum system

Ionisation of ground state creates inner-shell holes in ionic density matrix

Spatio-temporal evolution of ground-state population and ionic density matrix:

$$\begin{aligned} \frac{\partial \rho_0(\tau, z)}{\partial \tau} &= -\sigma_{1s} J_p(\tau, z) \rho_0(\tau, z) - \sigma_{2p} J_p(\tau, z) \rho_0(\tau, z) \\ \frac{\partial \rho_{1s1s}(\tau, z)}{\partial \tau} &= \sigma_{1s} J_p(\tau, z) \rho_0(\tau, z) - \Gamma_{1s} \rho_{1s1s}(\tau, z) + \left\{ i \rho_{1s2p}^*(\tau, z) \frac{\mu_{\mathcal{E}}(\tau, z)}{2} + c.c. \right\} \\ \frac{\partial \rho_{2p2p}(\tau, z)}{\partial \tau} &= \sigma_{2p} J_p(\tau, z) \rho_0(\tau, z) + \left\{ i \rho_{1s2p}(\tau, z) \frac{\mu_{\mathcal{E}}^*(\tau, z)}{2} + c.c. \right\} \\ \frac{\partial \rho_{1s2p}(\tau, z)}{\partial \tau} &= -\frac{\Gamma_{1s}}{2} \rho_{1s2p}(\tau, z) - i(\rho_{1s1s}(\tau, z) - \rho_{2p2p}(\tau, z)) \frac{\mu_{\mathcal{E}}(\tau, z)}{2}, \end{aligned}$$



Macroscopic polarization:  $\mathcal{P} = 2nZ_{1s2p}\rho_{1s2p}$

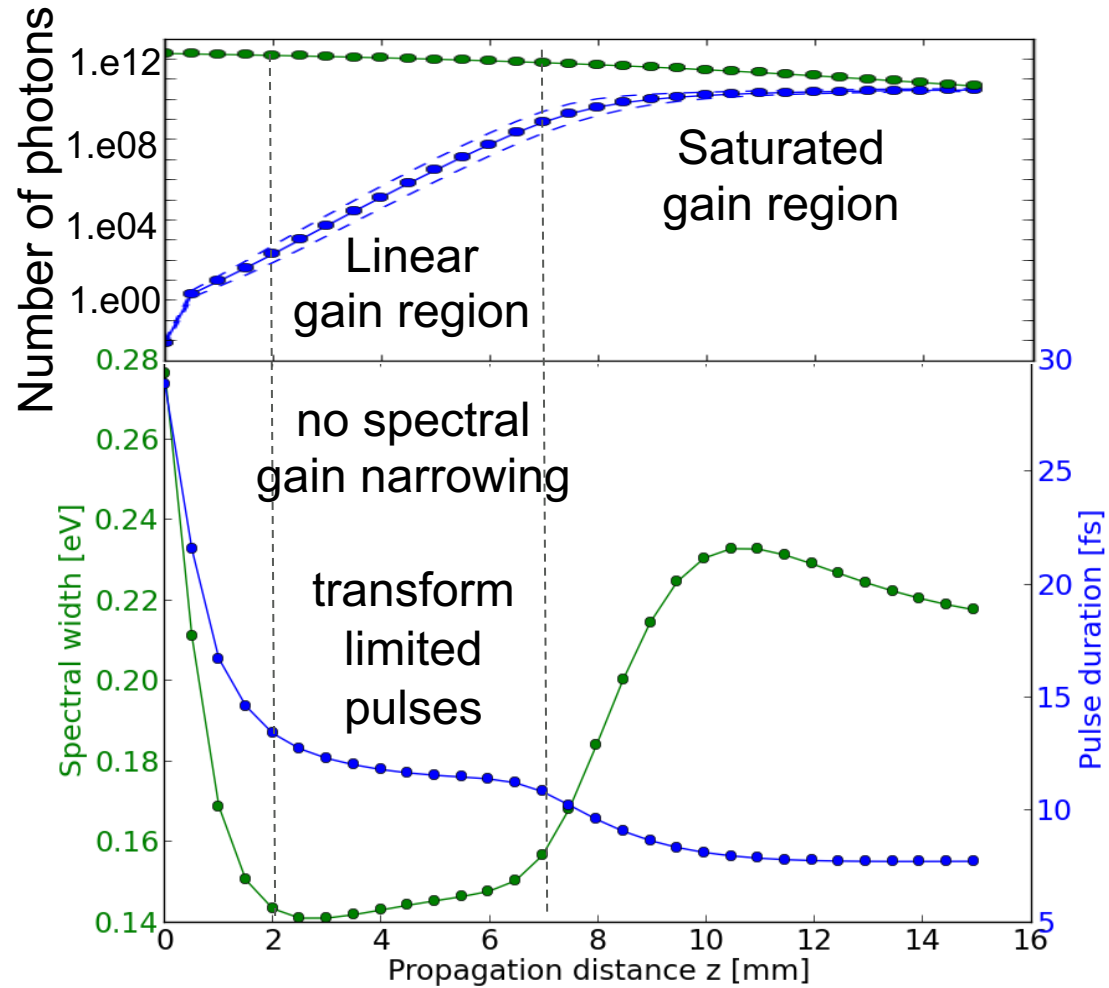
1D Maxwell equations (slow varying envelope, paraxial wave approximation)

$$\frac{\partial \mathcal{E}}{\partial z} + \frac{1}{c} \frac{\partial \mathcal{E}}{\partial t} = i \frac{\mu_0 \omega^2}{2k} \mathcal{P} + \mathbf{S} \quad \frac{\partial J_p(\tau, z)}{\partial z} = -\rho_0(\tau, z) n (\sigma_{1s} + \sigma_{2p}) J_p(\tau, z)$$

(solved in moving frame of propagating pump light)

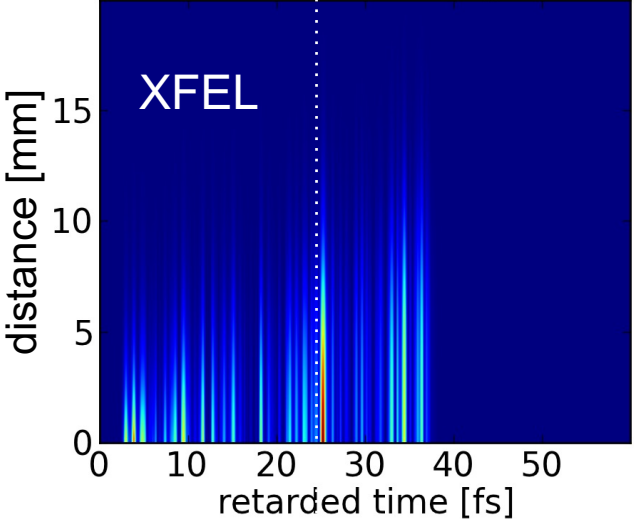
# The build-up of transform-limited pulses

Gaussian pulse, 40 fs,  $2 \times 10^{12}$  photons; Length: 16 mm, Density:  $1.6 \times 10^{19} \text{ cm}^{-3}$

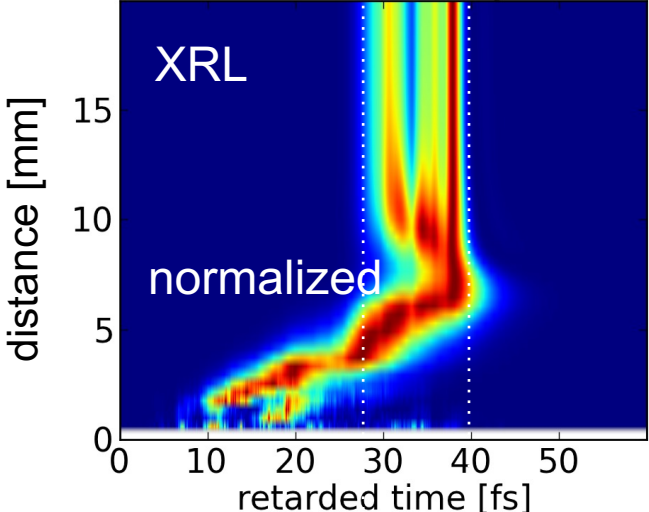


# Prediction: phase-stable pulses of fs duration

Temporal structure of SASE pump Pulse

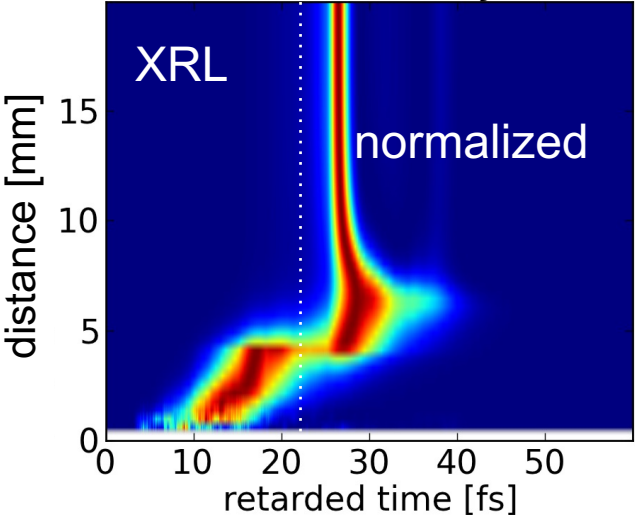


Temporal structure of emitted XRL

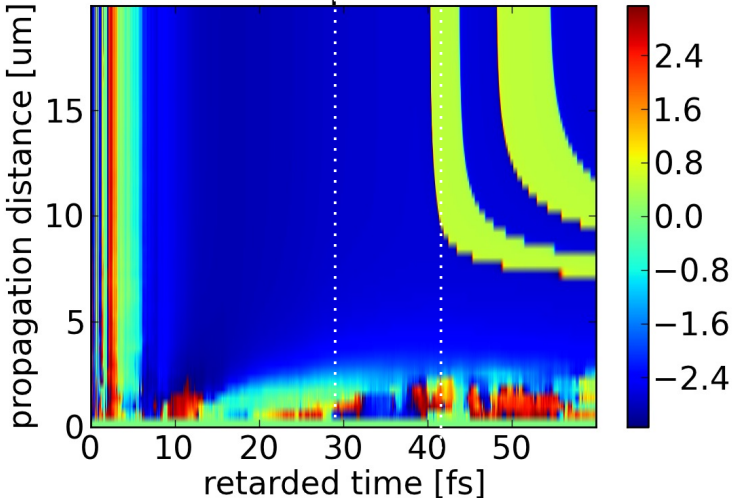


Example of 3 emission bursts

Temporal structure of emitted XRL



XRL phase



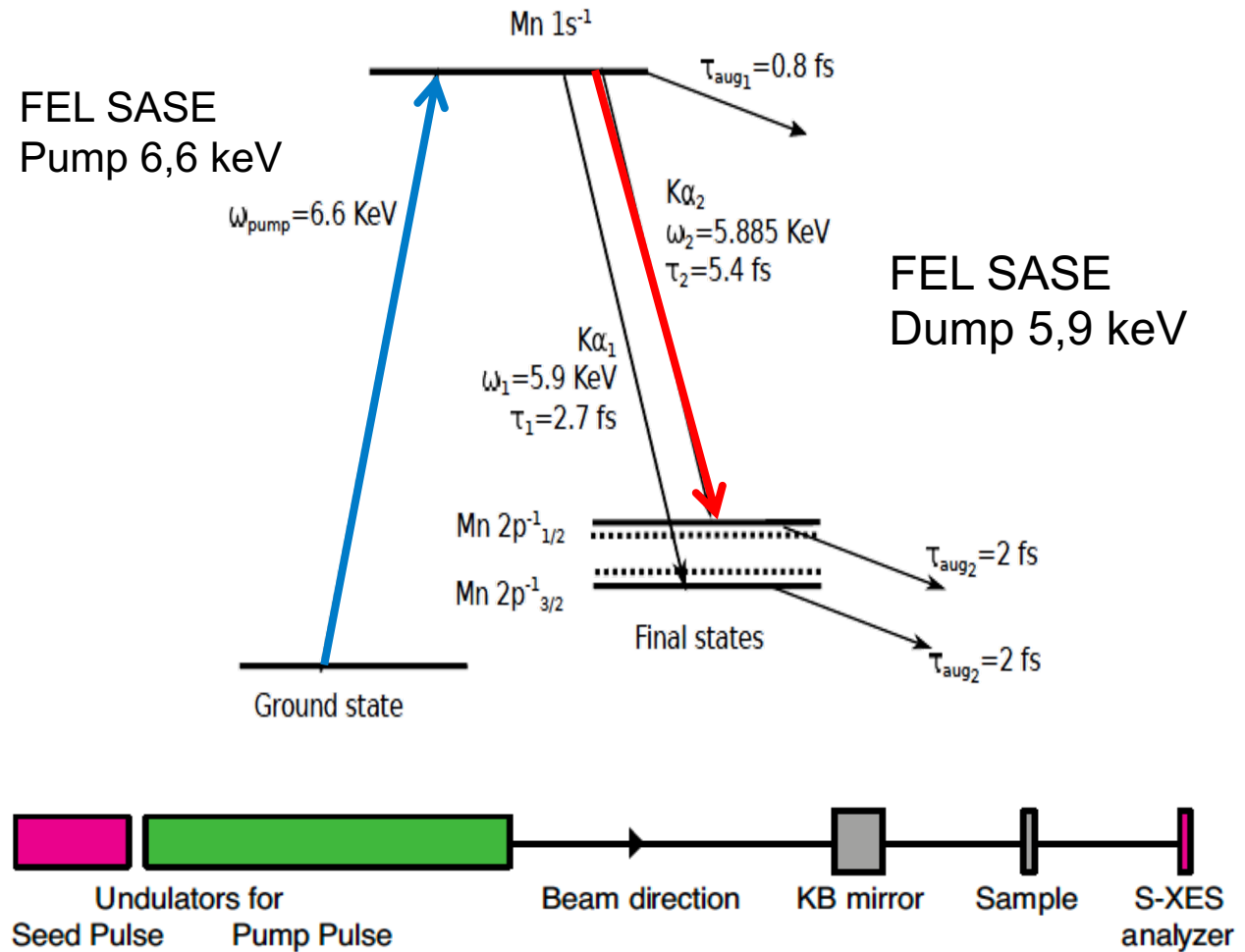
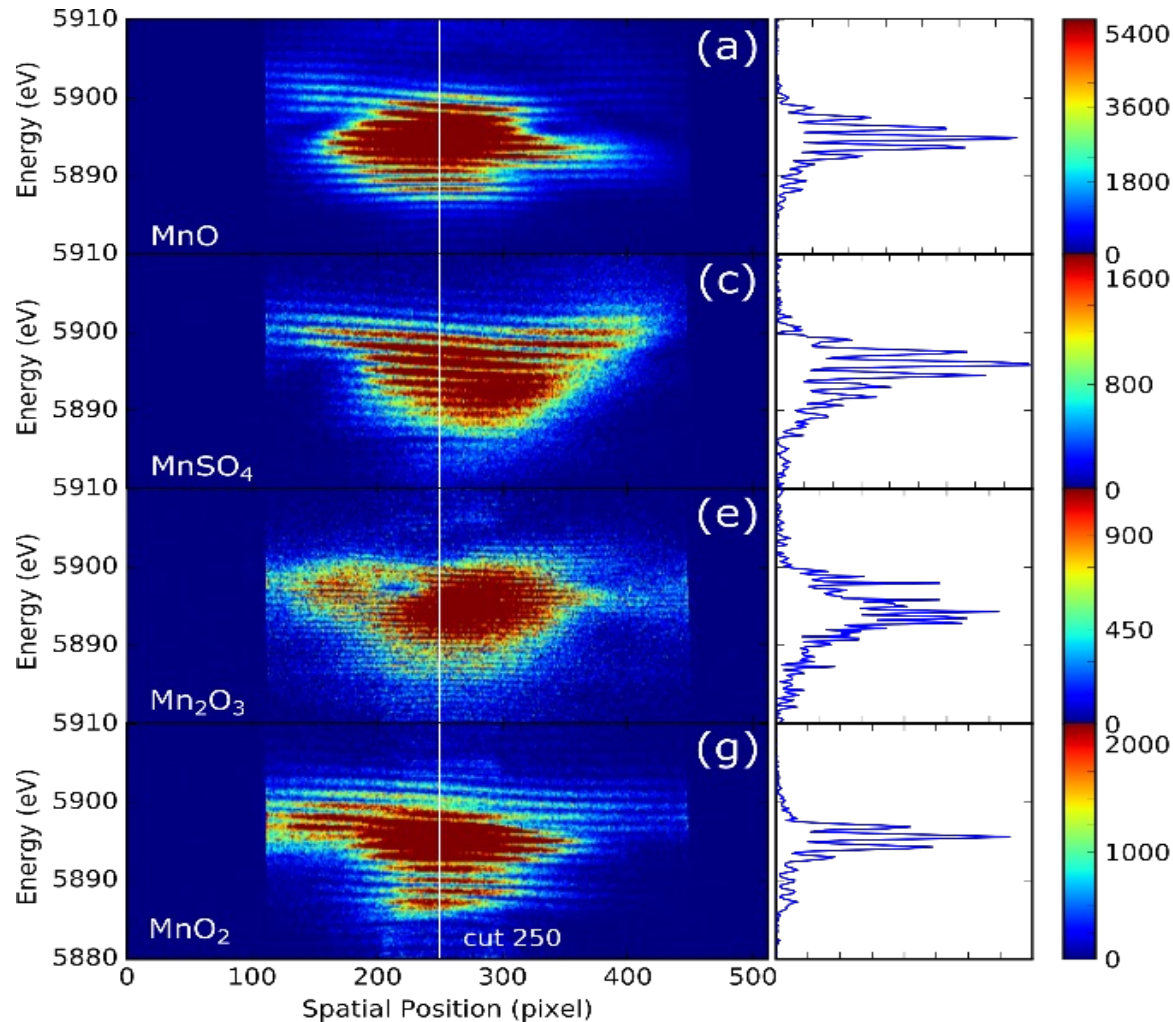
Emission bursts are phase stable



# Recent results: Stimulated K- $\alpha$ emission

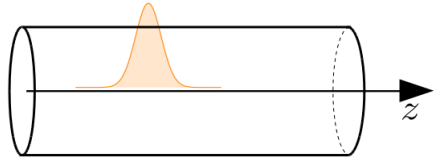
Interference pattern points to emission of 2 phase stable fs pulses of fs separation

Spatial profile: several transverse modes



# 3D model in paraxial symmetry for a 2-band electronic system

Field in paraxial approximation, in retarded time, in Rabi frequency units

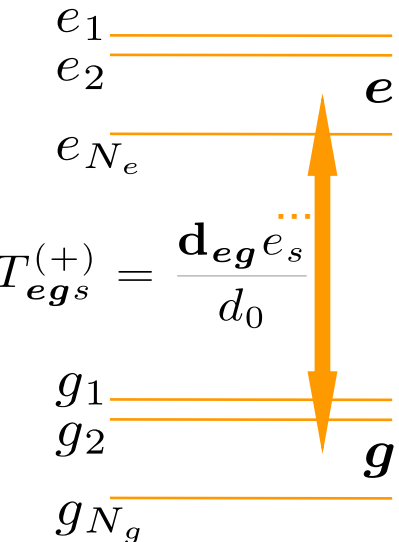


$$\mathbf{D}(\mathbf{r}, t) = \sum_s \left( D_s^{(+)}(\mathbf{r}, t) e^{i(k_0 z - \omega_0 t)} \mathbf{e}_s + D_s^{(-)}(\mathbf{r}, t) e^{-i(k_0 z - \omega_0 t)} \mathbf{e}_s^* \right),$$

$$\Omega_s^{(\pm)}(\mathbf{r}, t) = d_0 D_s^{(\pm)}(\mathbf{r}, t + z/c) / (\hbar \epsilon_0)$$

$$\left[ \frac{\partial}{\partial z} - \frac{i}{2k_0} \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) \right] \Omega_\sigma^{(+)} = -\frac{\mu}{2} \Omega_\sigma^{(+)} + i \frac{3}{8\pi} \lambda^2 \Gamma_{sp} \left( n T_{ges}^{(-)} \rho_{eg}^{(+)} + f_s^{(+)} \right)$$

Atomic gain medium: two set of levels, rotating-wave approximation



$$\frac{\partial}{\partial t} \rho_{ee'} = i \left( \Omega_s^{(+)} T_{egs}^{(+)} \rho_{ge'}^{(-)} - \Omega_s^{(-)} \rho_{eg}^{(+)} T_{ge's}^{(-)} \right)$$

$$\frac{\partial}{\partial t} \rho_{eg}^{(+)} = -i \Omega_s^{(+)} \left( \rho_{ee'} T_{e'gs}^{(+)} - T_{eg's}^{(+)} \rho_{gg'} \right) + f_s^{(-)*} \rho_{ee'} T_{e'gs}^{(+)}$$

$$\frac{\partial}{\partial t} \rho_{ge}^{(-)} = +i \Omega_s^{(-)} \left( T_{ge's}^{(-)} \rho_{e'e} - \rho_{gg'} T_{g'es}^{(-)} \right) + f_s^{(+)*} T_{ge's}^{(-)} \rho_{ee'}$$

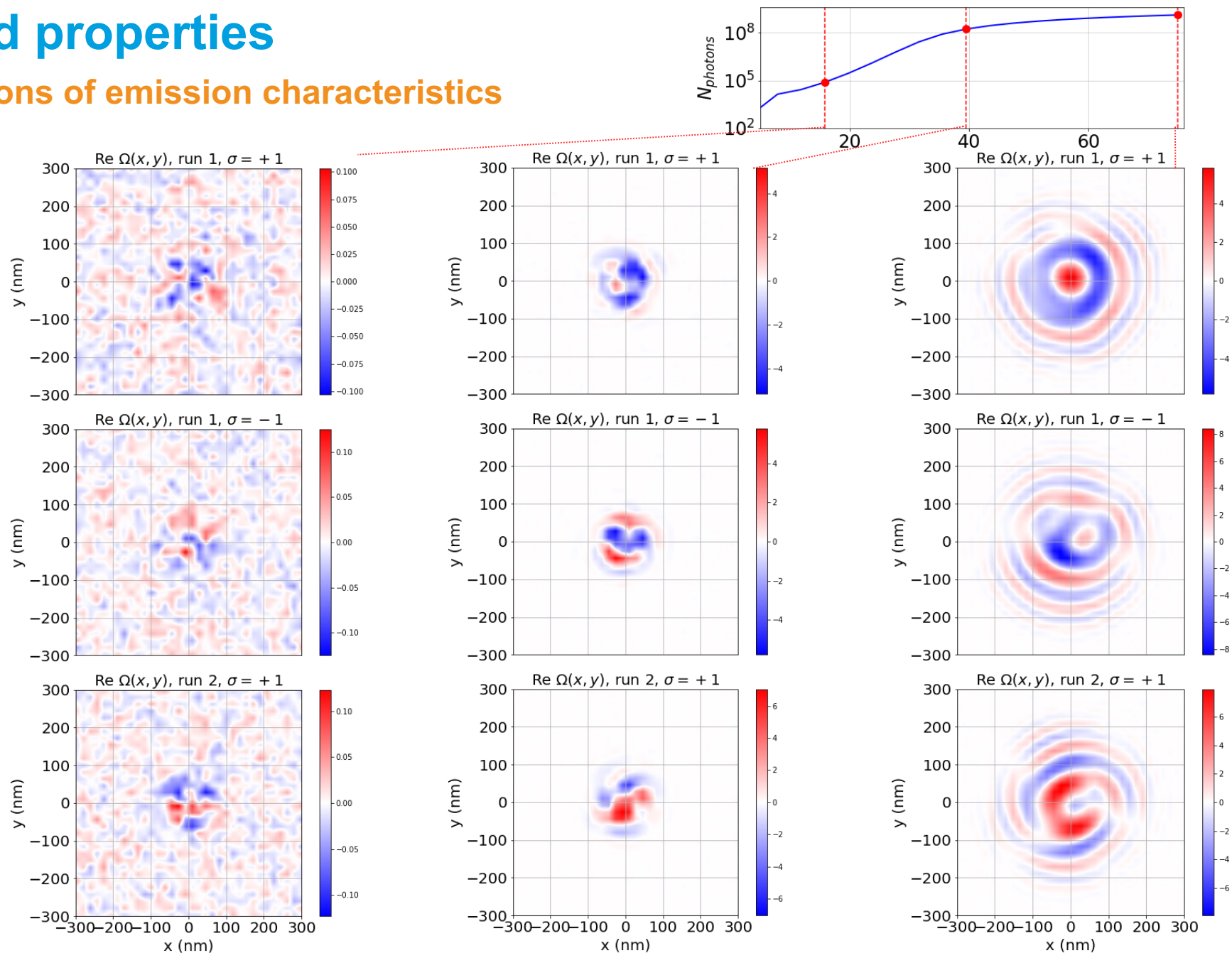
$$\frac{\partial}{\partial t} \rho_{gg'} = i \left( \Omega_s^{(-)} T_{ges}^{(-)} \rho_{eg'}^{(+)} - \Omega_s^{(+)} \rho_{ge}^{(-)} T_{eg's}^{(+)} \right) + f_s^{(+)*} T_{ges}^{(-)} \rho_{eg'}^{(+)} + f_s^{(-)*} \rho_{ge}^{(-)} T_{eg's}^{(+)}$$

**Noise correlation properties:**

$$\langle f_s^{(\pm)}(\mathbf{r}, t) f_{s'}^{(\pm)*}(\mathbf{r}', t') \rangle = \delta_{(It_0)}(t + z/c - t' - z'/c) \delta_{(space)}(\mathbf{r} - \mathbf{r}') \delta_{ss'}$$

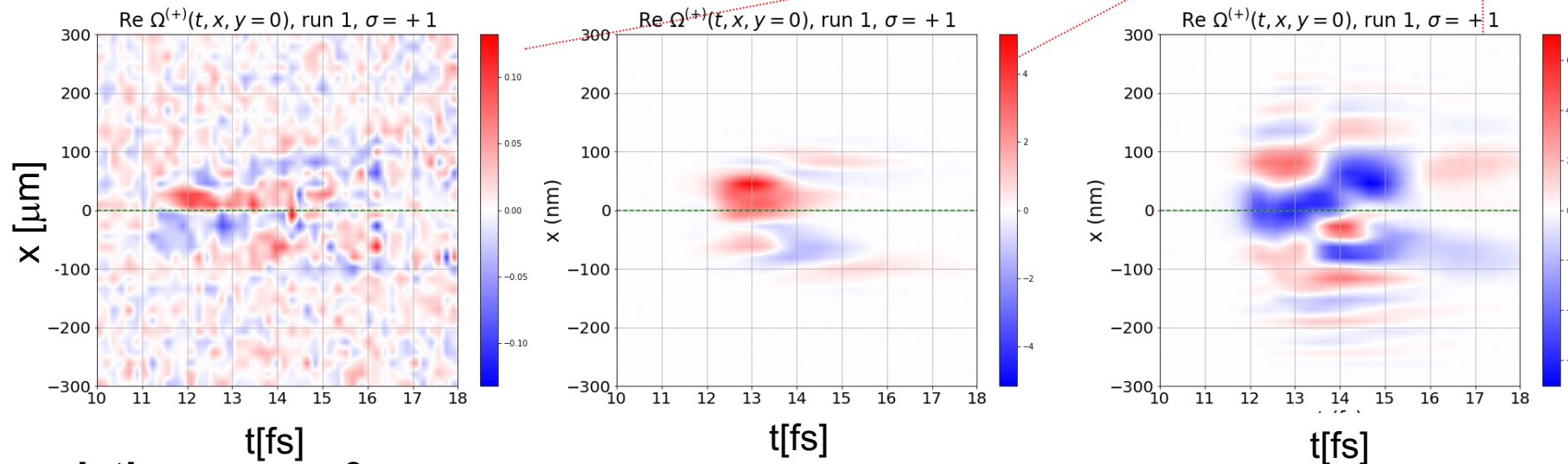
# Transverse field properties

## Shot-to-shot fluctuations of emission characteristics

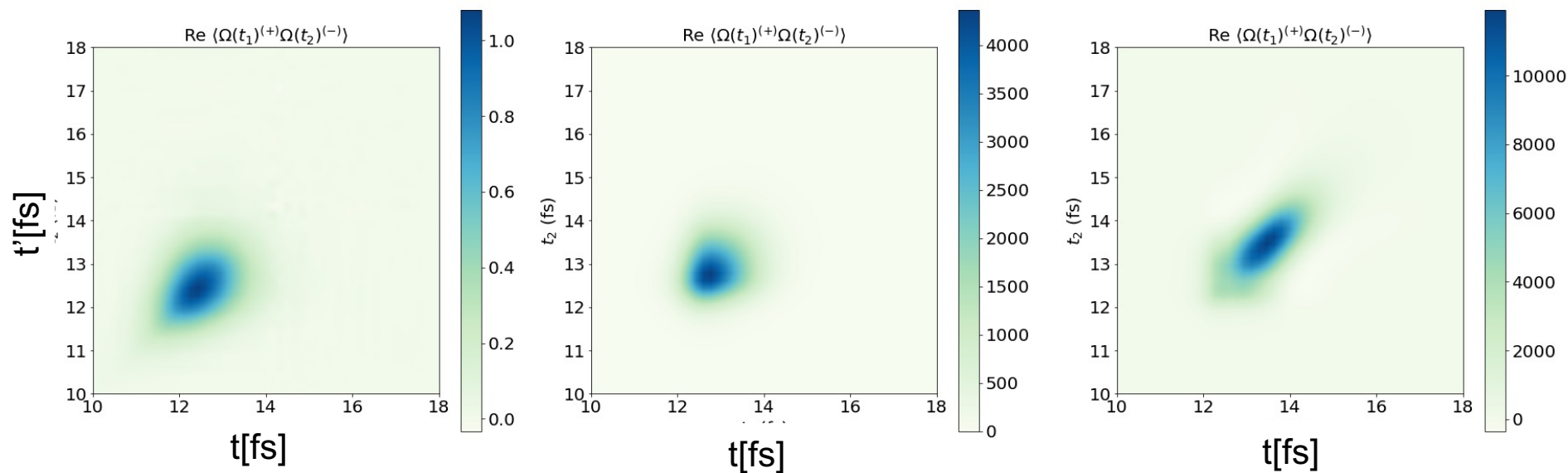


# Spatio-temporal field properties

## Temporal field envelope versus x



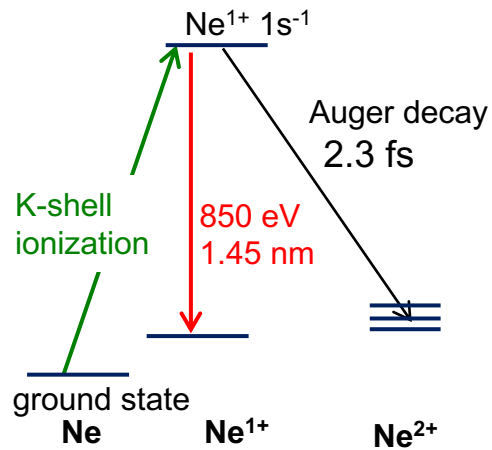
## Temporal correlation at $x=y=0$



# Amplified spontaneous x-ray emission (X-ray superfluorescence)

## Photoionization K- $\alpha$ laser – from 1<sup>st</sup> demonstration to chemical analysis

Ne 849 eV (gas)

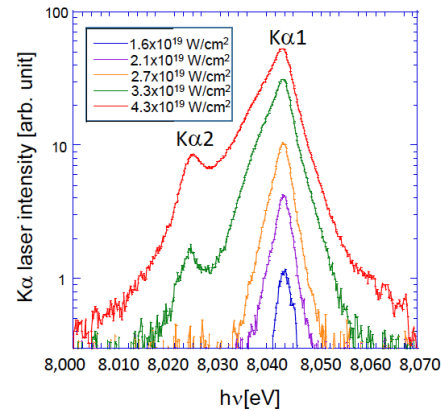


Rohringer et al.,  
*Nature* **481**, 488 (2012).

Scheme first proposed by  
Duguay and Rentzepis,  
*Appl. Phys. Lett.* 10, 350 (1967).

Emission in forward direction,  
up to e<sup>21</sup> amplification of  
spontaneous K- $\alpha$  emission

Cu 8,04 keV (solid)

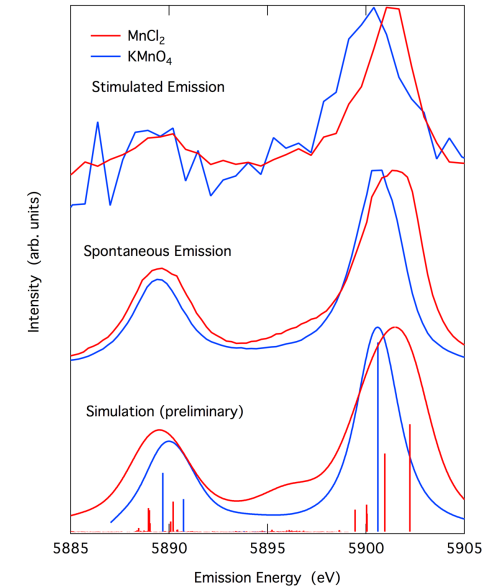


Yoneda et al.,  
*Nature* **524**, 446 (2015).

Hard x-ray laser,  
seeded by 2-color FEL  
operation

Si L lines 70-100 eV (solid)  
M. Beye et al.,  
*Nature* **501**, 191 (2013)

Mn 5,9 keV (liquid)



Kroll et al.,  
Manuscript in prep. (2016).

Stimulated emission for  
Chemical analysis under  
single FEL pulse  
exposure

# Stimulated Emission Spectroscopy

Characterization of the catalytic pathways and intermediates in bio catalysis

X-ray supfluorescence maintains chemical sensitivity



U. Bergmann

T. Kroll

A. Marinelli

A. Lutman

S. Boutet

A. Aquila

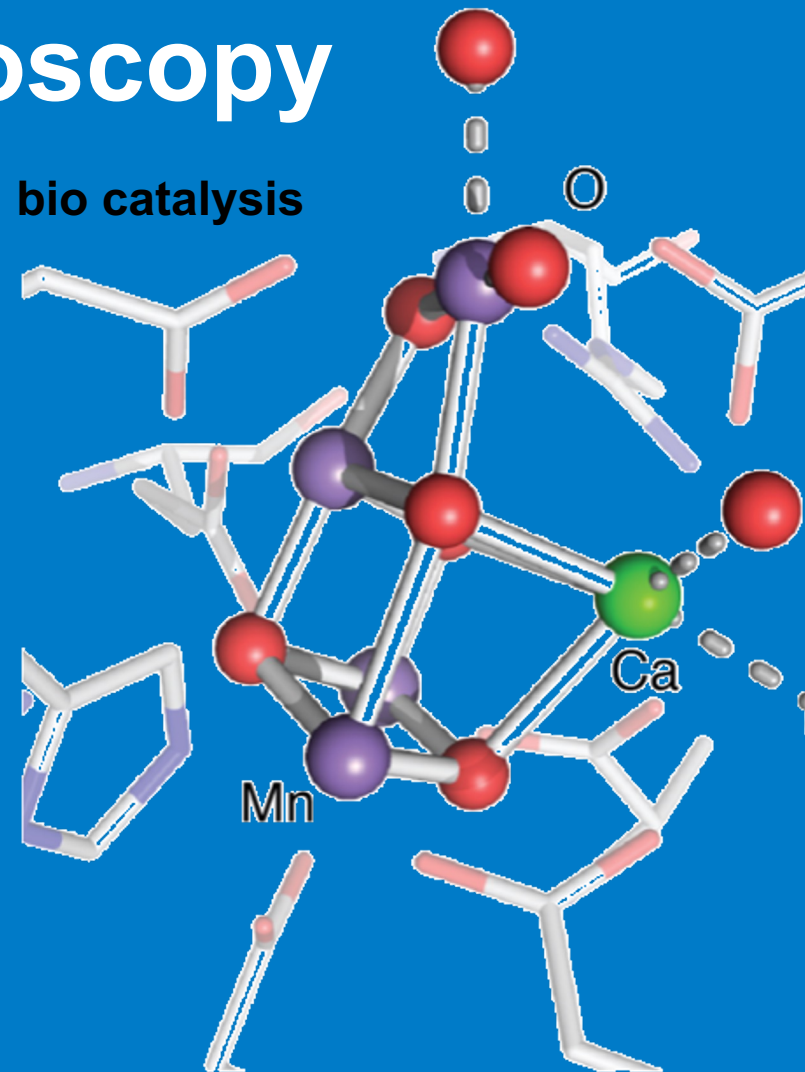
C. Weninger

J. Yano

V. K. Yachandra

J. Kern

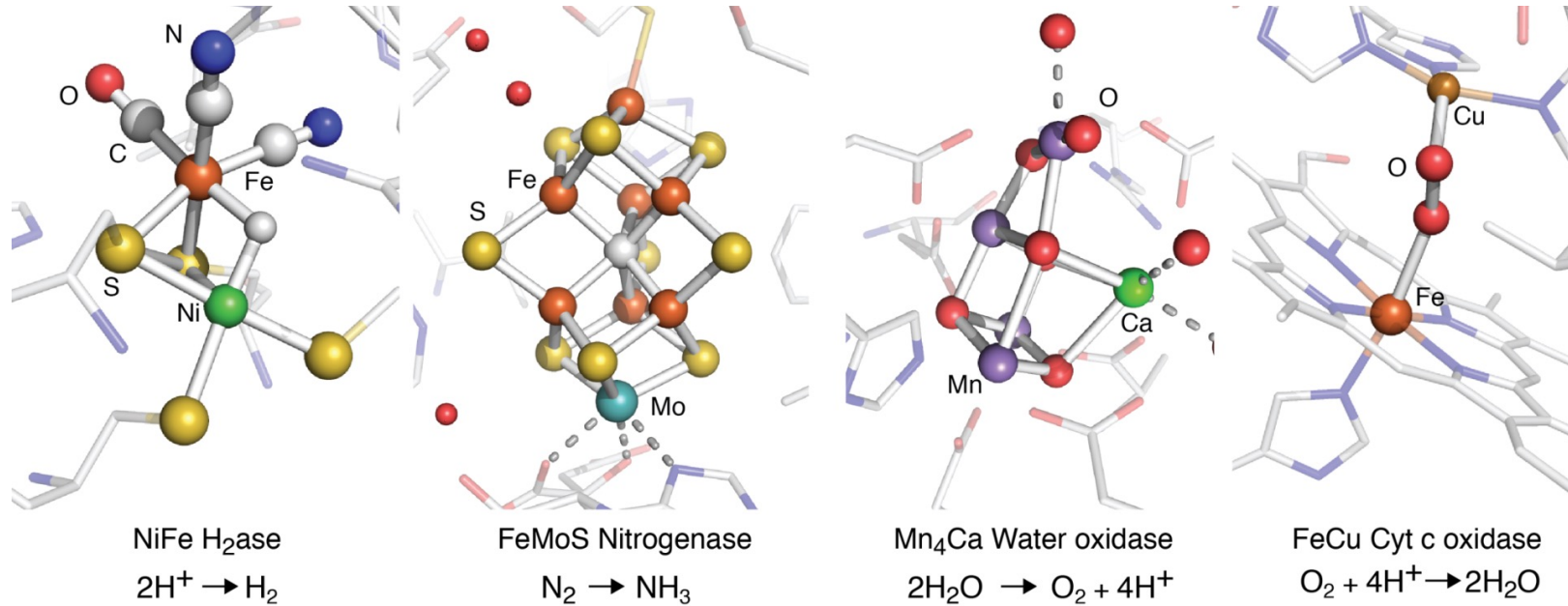
A. Benediktovitch



Mn<sub>4</sub>Ca Water oxidase



# Importance of Metals in Biological Systems

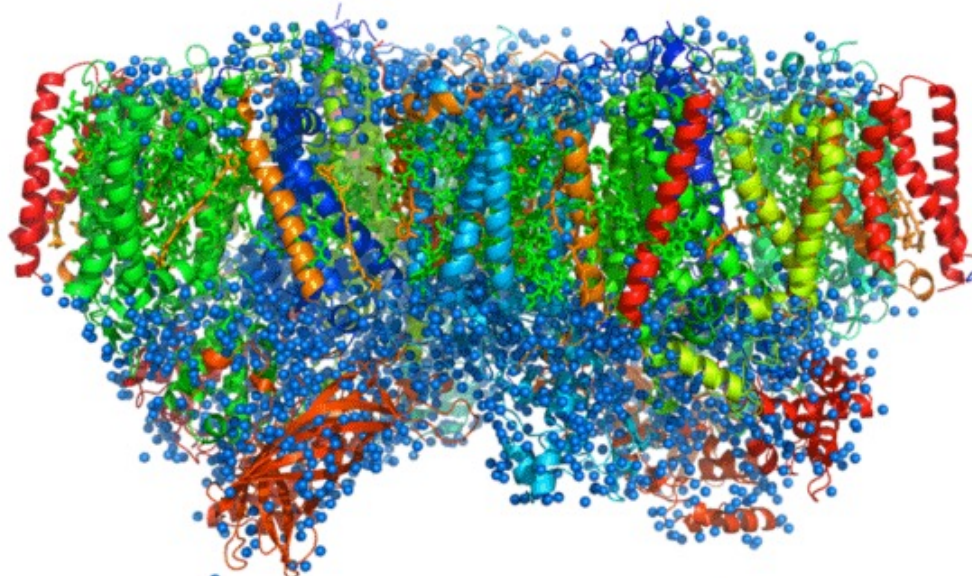


Respiration  
Photosynthesis  
Nitrogen fixation  
RNA synthesis, Detoxification...

Catalyze thermodynamically demanding reactions under physiological conditions with minimized driving force!

**Holy Grail:**  
**Characterization of the Catalytic Pathways and Intermediates**

# Structure – Function Relationship



## Coherent diffractive imaging:

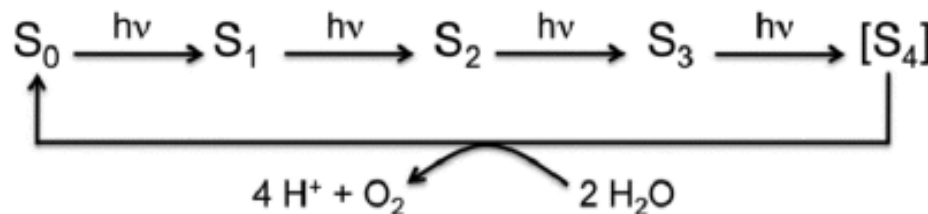
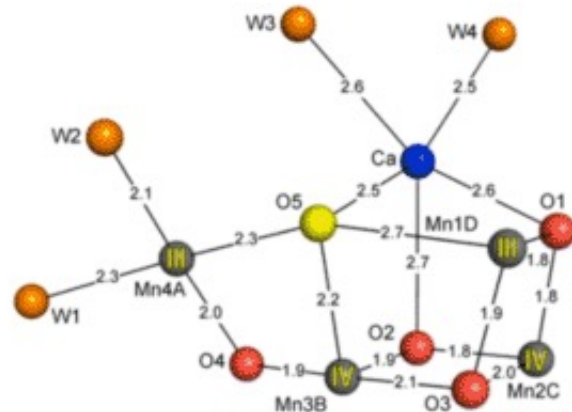
Molecular structure in reciprocal space

Structure (atomic positions) doesn't give us the complete picture.

Consider for example:

Pure electronic excitation transfer, energy transfer to reaction centers

Change of valence electron distribution (chemistry) during catalytic reaction



## X-ray spectroscopy

a complementary tool to

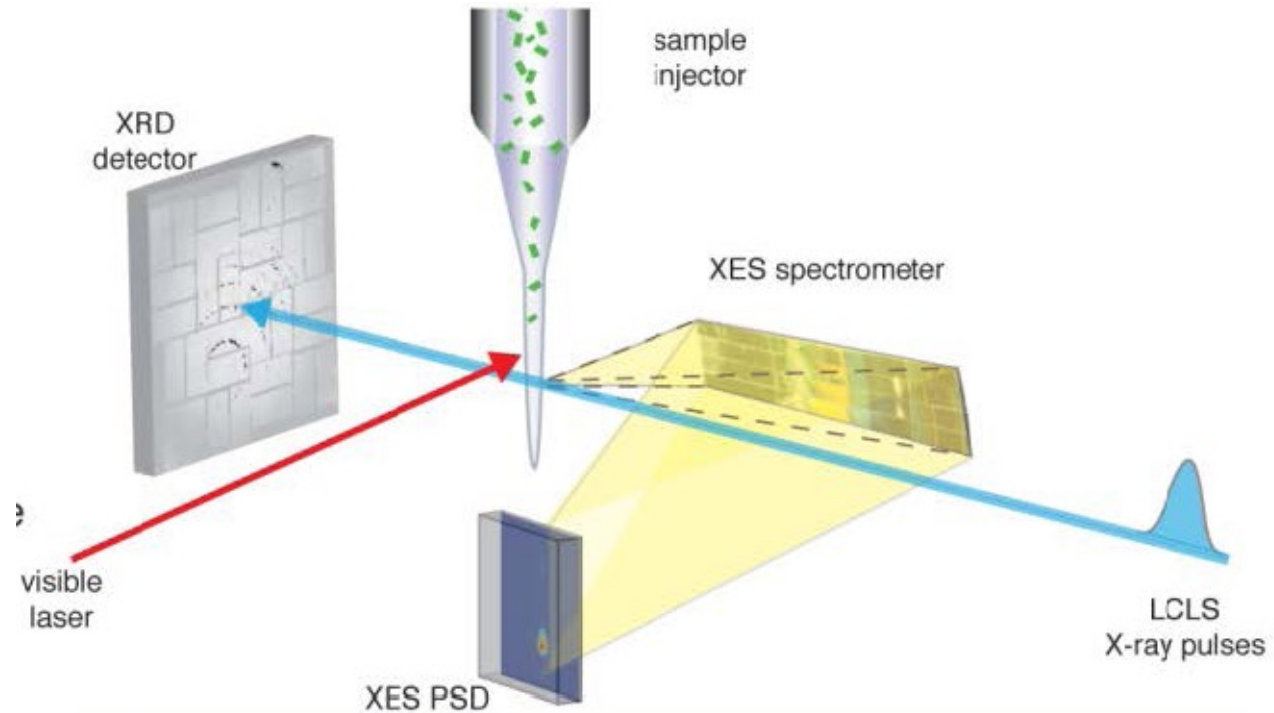
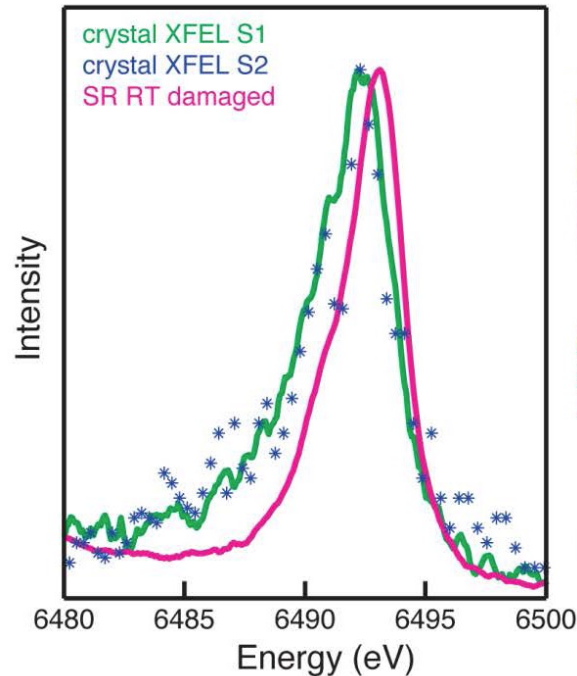
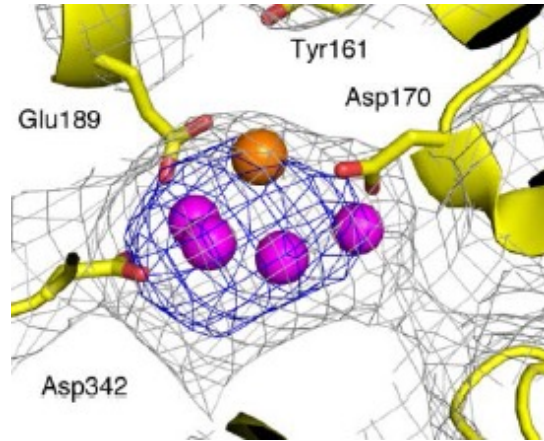
## x-ray diffraction

to tackle these questions



# Amplification of x-ray emission signal

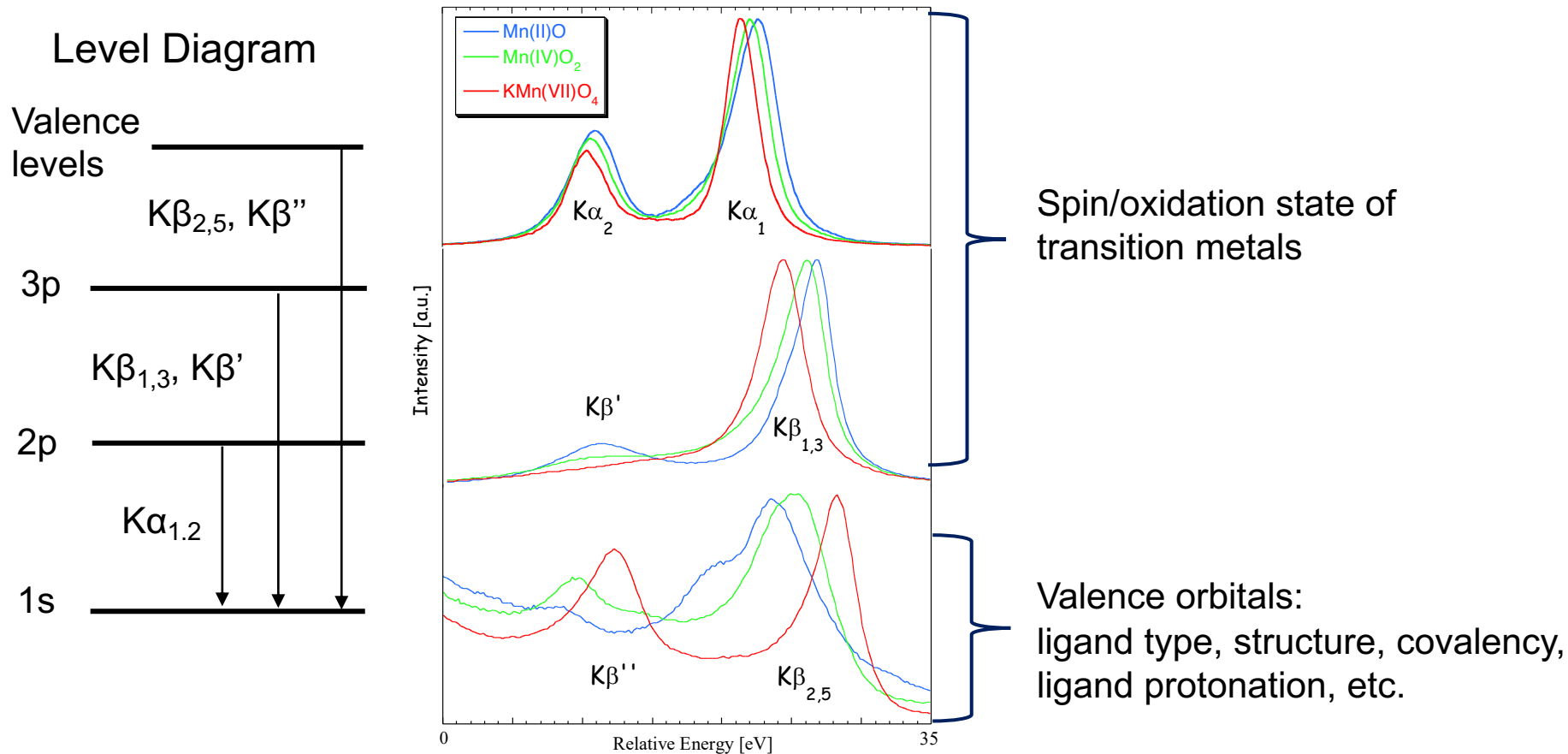
## Single-shot high-resolution spectroscopy



Probing the electronic structure of the  $Mn_4CaO_5$  cluster in the oxygen-evolving complex of PS II

# Towards stimulated emission spectroscopy

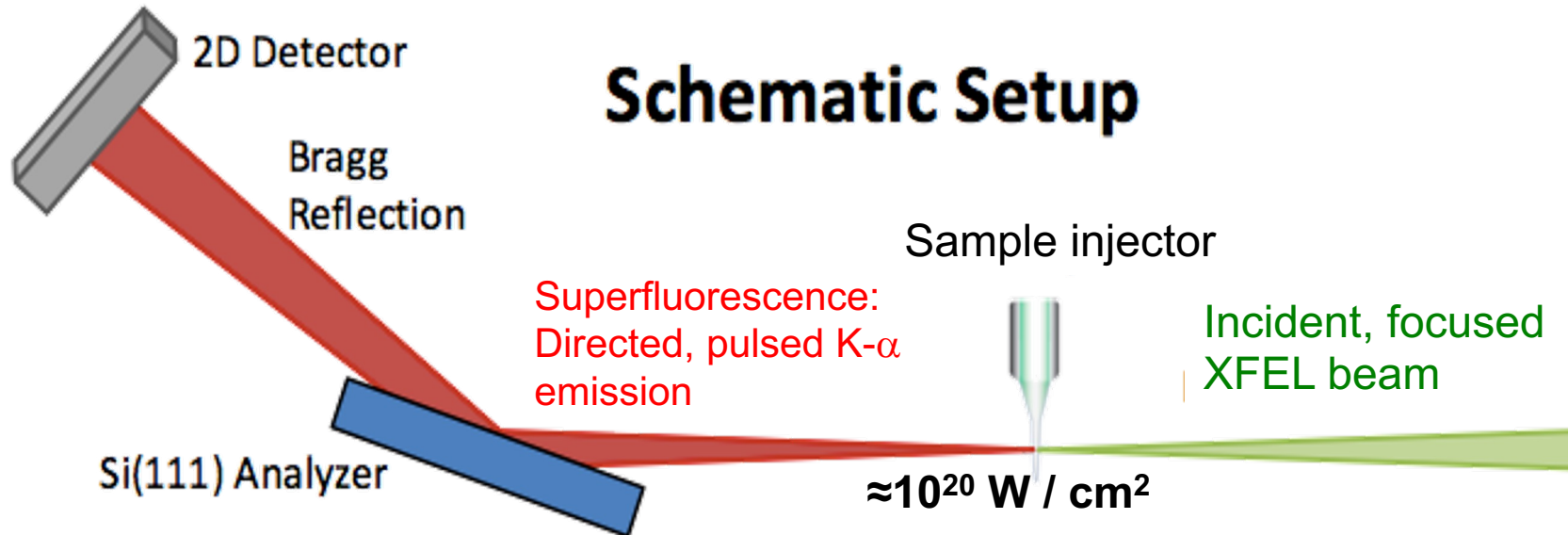
## X-ray emission reveals chemically relevant information



For reviews see: Glatzel & UB, *Coord. Chem. Rev.*, **249**, 65-95, (2005)  
Pollock & DeBeer, *Accounts of Chemical Research* (2015)

# K- $\alpha$ lasing of Mn-salt aqueous solutions

$10^{20}$  W/cm<sup>2</sup> on target creates population inversion on K- $\alpha$  transition



$\approx 150$  nm focus  
 $\approx 10^{11}$  ph/pulse  
 $\approx 50$  fs pulse duration

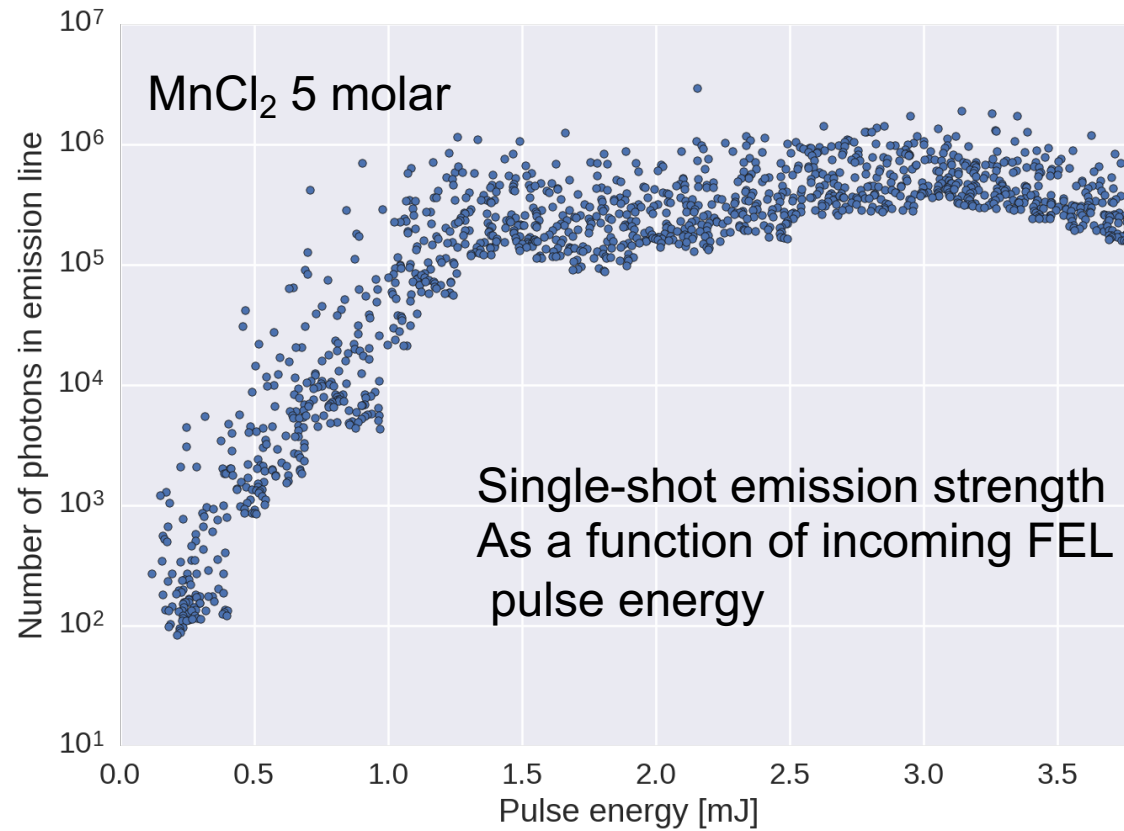
Samples: MnCl<sub>2</sub> solution (5 and 1 molar), KMnO<sub>4</sub> (0.4 molar)

Collect 100% of emission in forward direction

Use flat analyzer crystal – high efficiency

# Gain Curve for the 5.9 keV $K\alpha_1$ emission

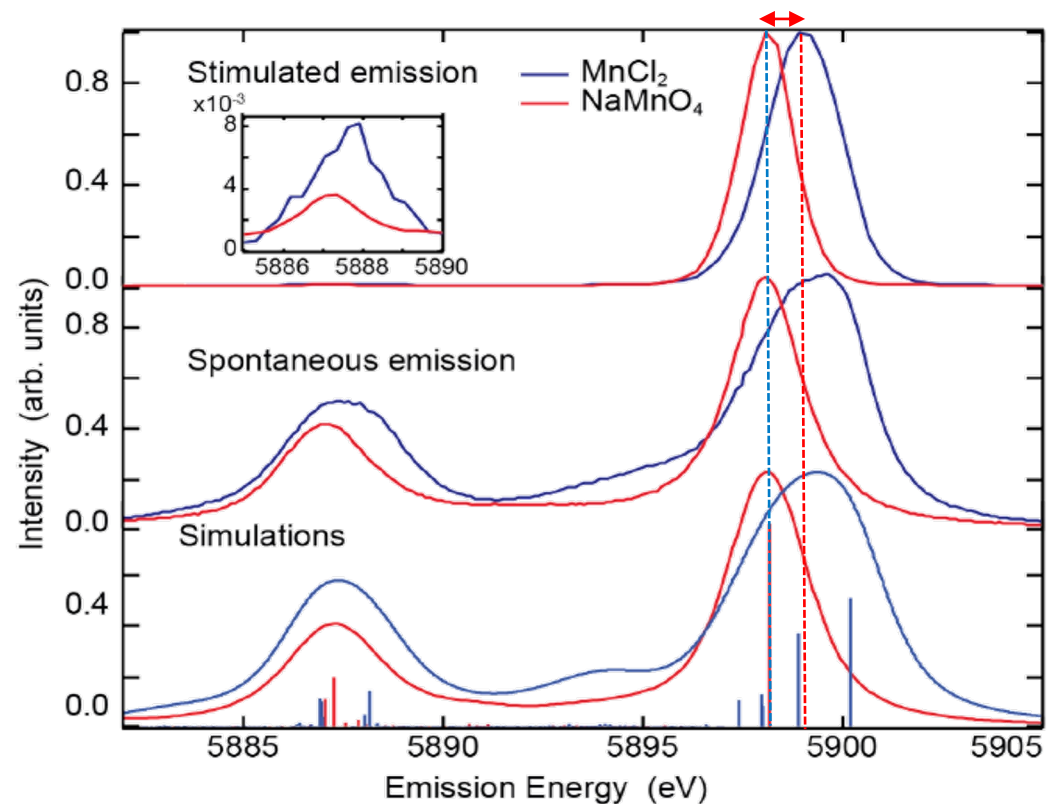
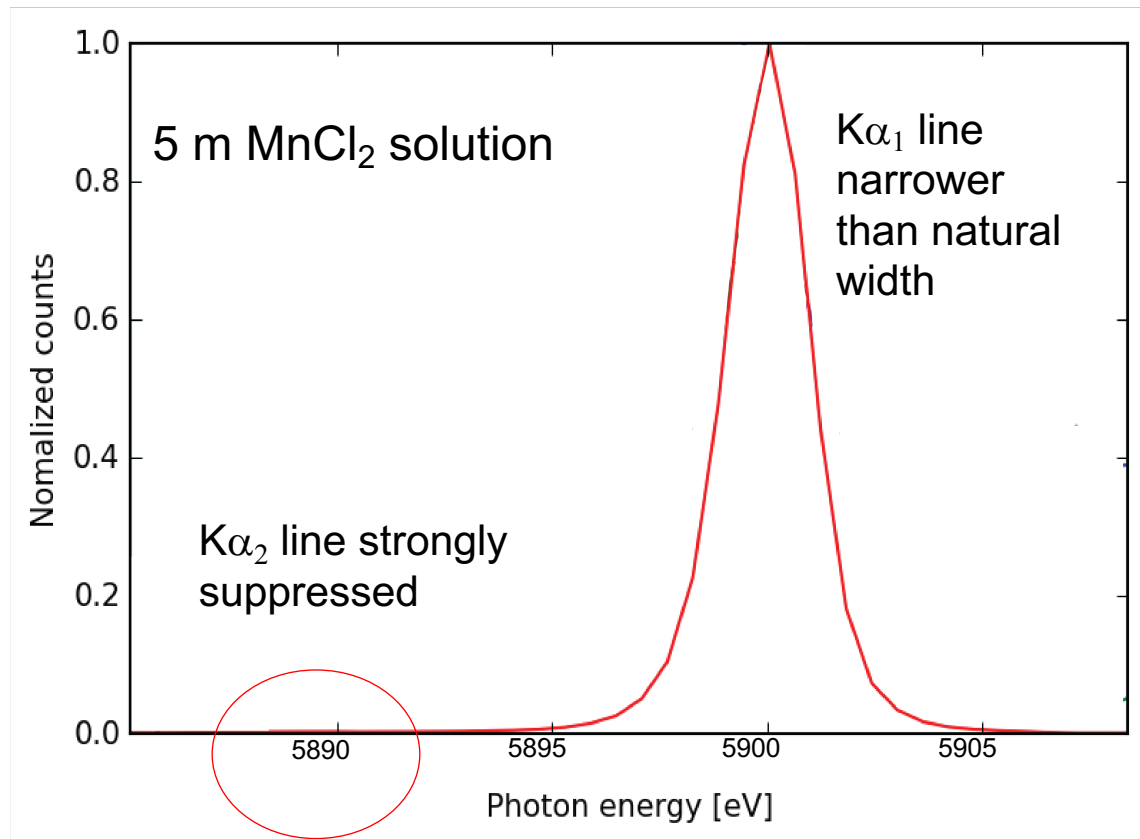
Exponential amplification over 4 orders of magnitude



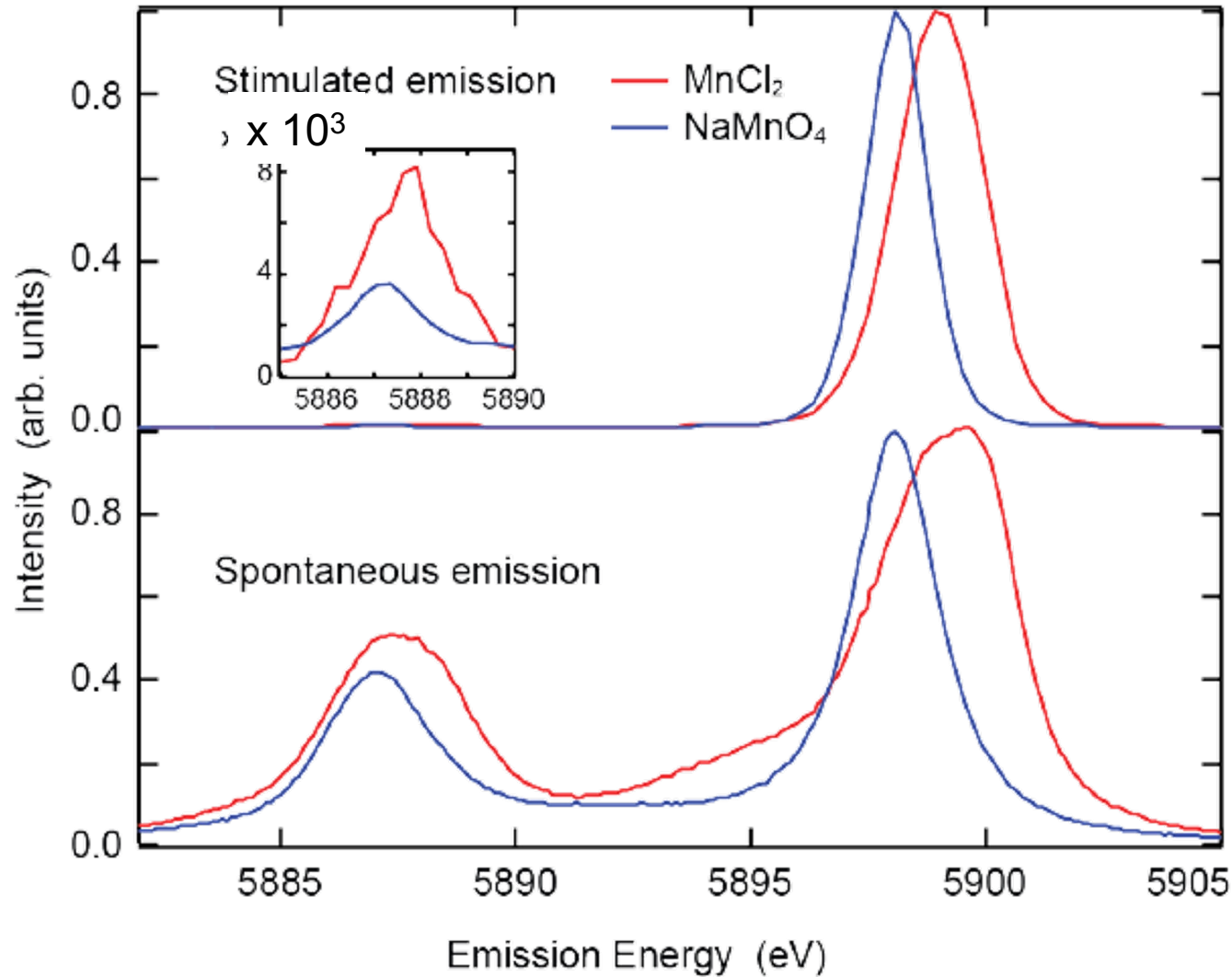
# Observation of strong lasing in Mn at 5.9 keV $K\alpha_1$

Strong gain up to  $10^6$  detected Mn  $K\alpha$  photons

Single shot spectrum



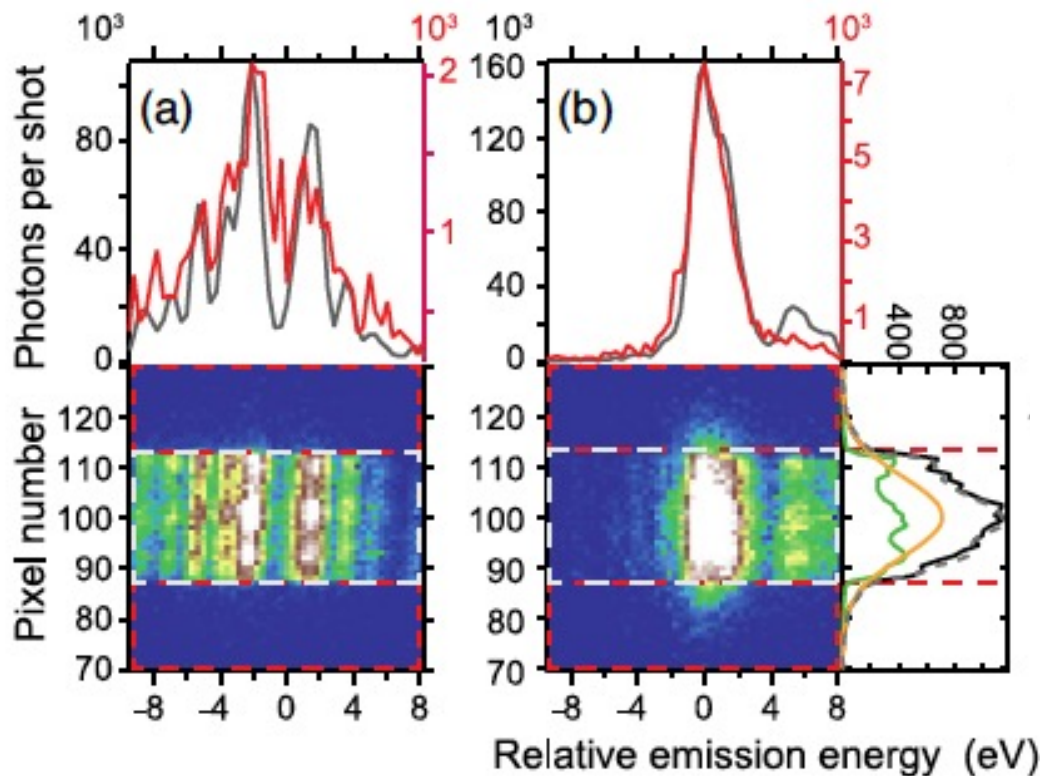
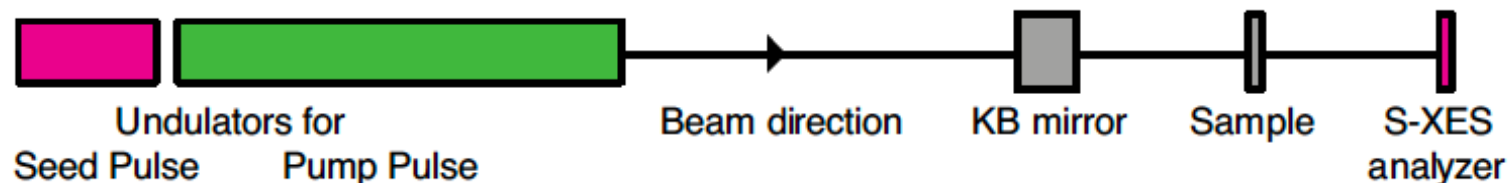
# Chemical Sensitivity



Stimulated emission retains the expected chemical shift !

# Stimulated K- $\beta$ emission in Mn salts – 2 colour FEL operation

## Statistical evidence of stimulated K- $\beta$ emission



Analysis:  
Spatial spectral profile

So far, proof-of-principle  
(no improvement  
on spectral information)

# X-ray Laser Oscillator (XLO)

Coherent x-ray pulses of narrow spectral bandwidth & high intensity



**C. Pellegrini**  
**A. Halavanau**  
Y. Feng  
Z. Huang  
A. Marinelli  
A. Lutman  
F. J. Decker  
S. Carbajo  
F. Fuller



**U. Bergmann**  
Noah Welke



A. Benediktovitch  
S. Chuchurka  
V. Sukharnikov

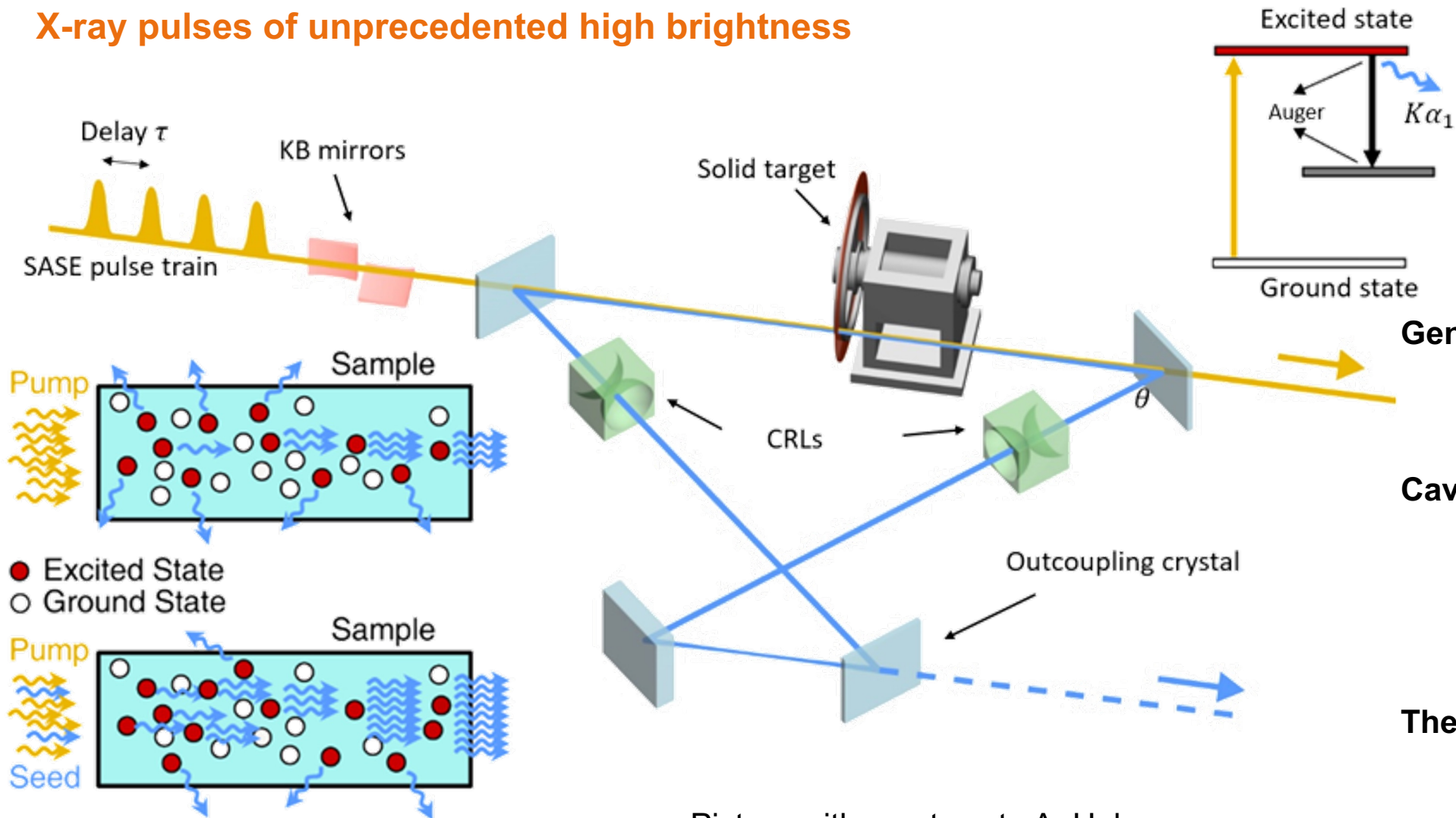


Š. Krušič



# Scheme of X-ray Laser Oscillator (XLO)

X-ray pulses of unprecedented high brightness



## Challenges

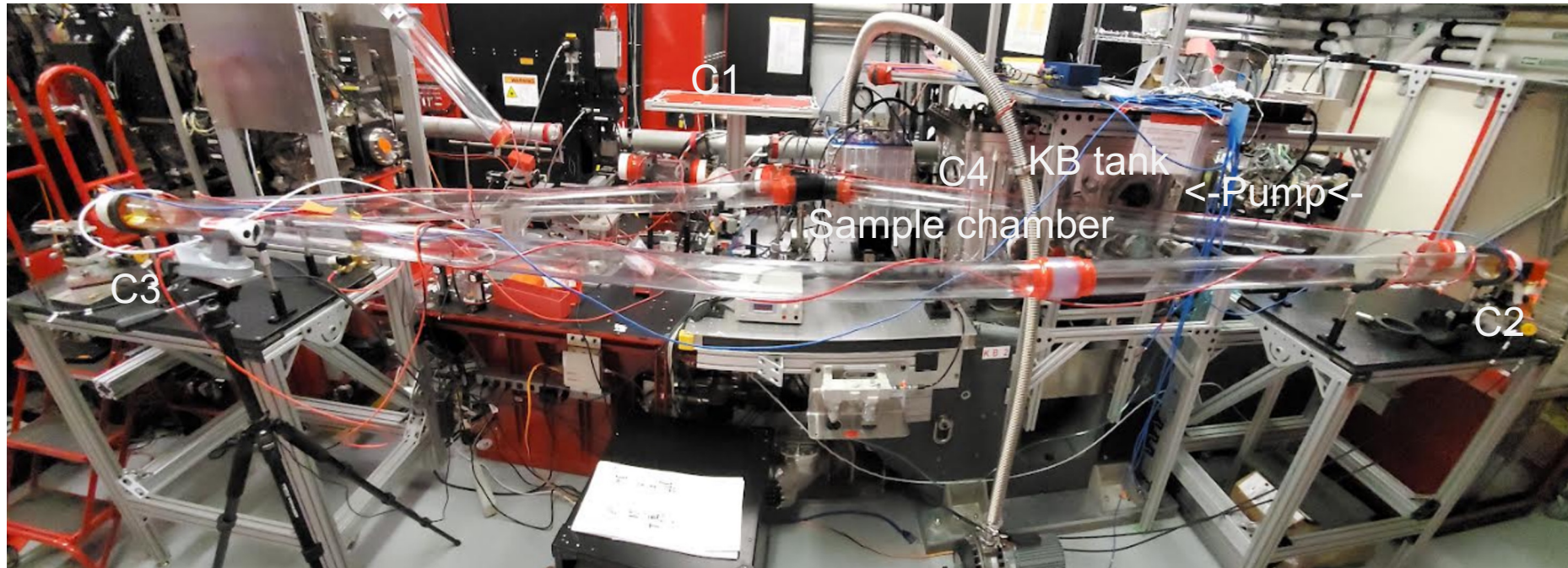
**Generation of multiple bunches**  
with 10 ns spacing  
acceleration in Cu linac

**Cavity design:**  
minimize losses  
maximize acceptance angle  
thermal issues  
Outcoupling of X-ray pulses

**Theory:**  
3D simulations  
Quantitative predictions  
Optimization

# Current experimental setup at CXI

Time of flight – about 35 ns

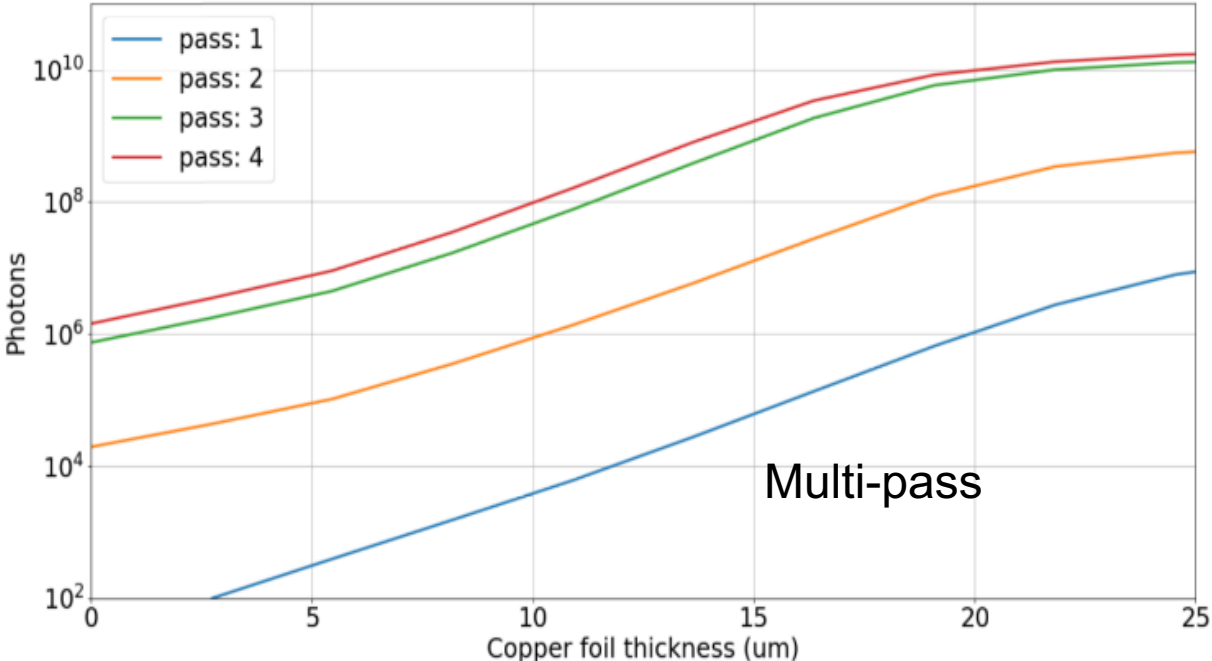
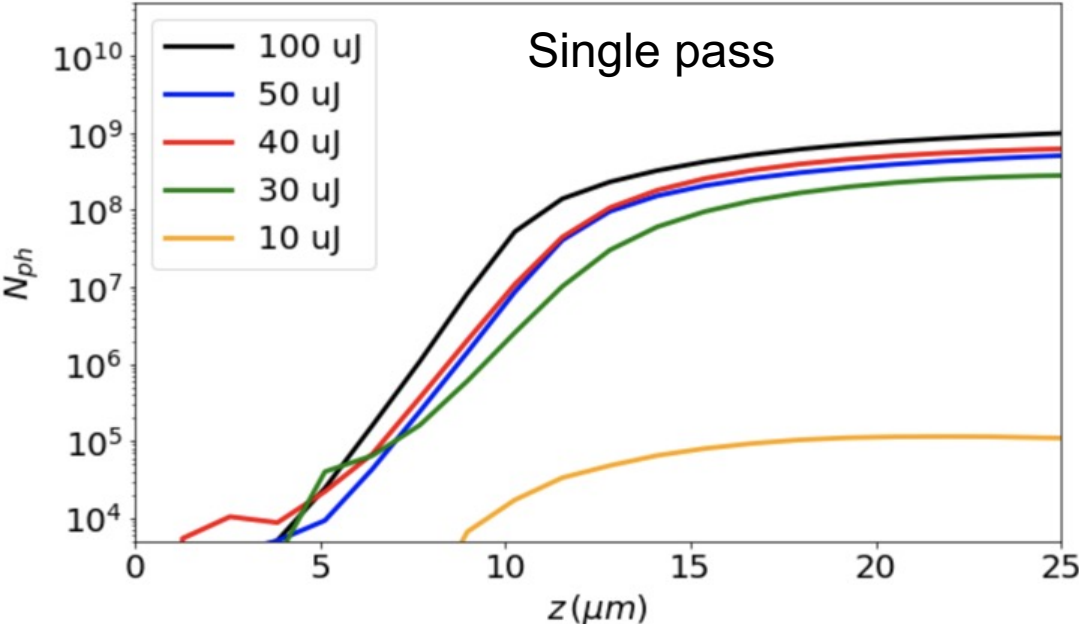


XLO v1.2, LCLS-CXI, November 15, 2022

# Numerical simulations – number of photons

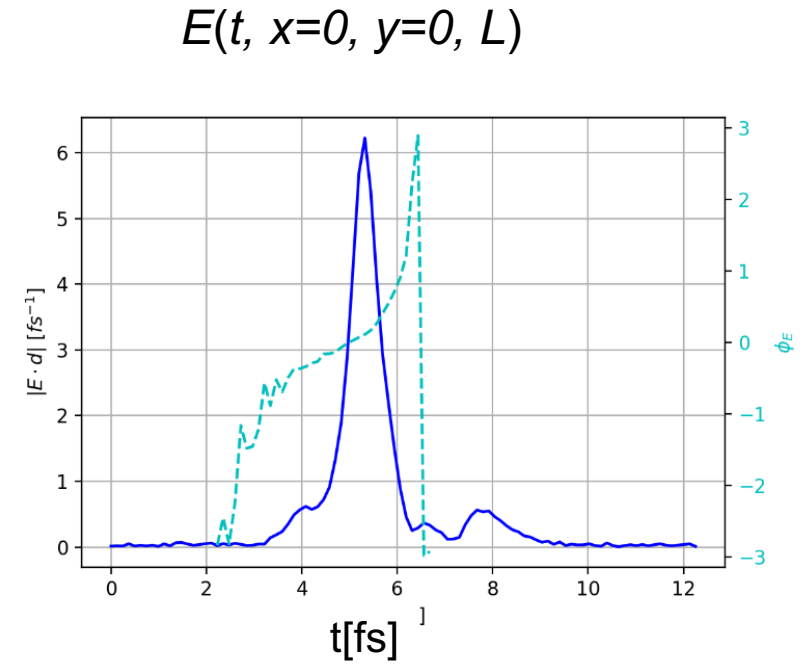
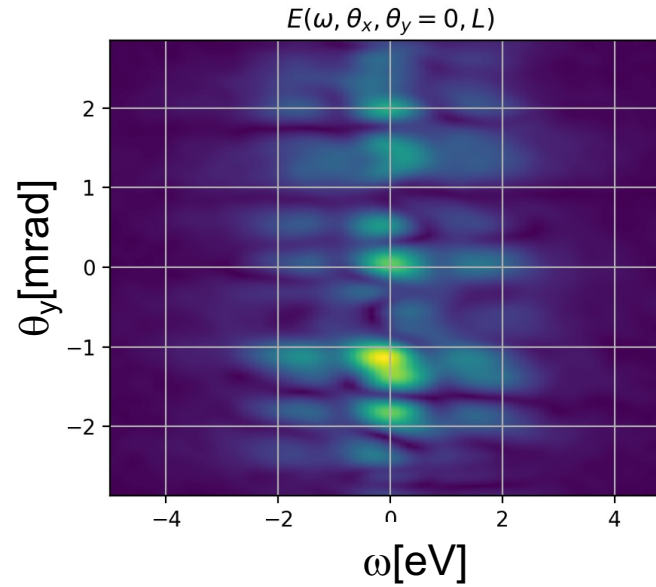
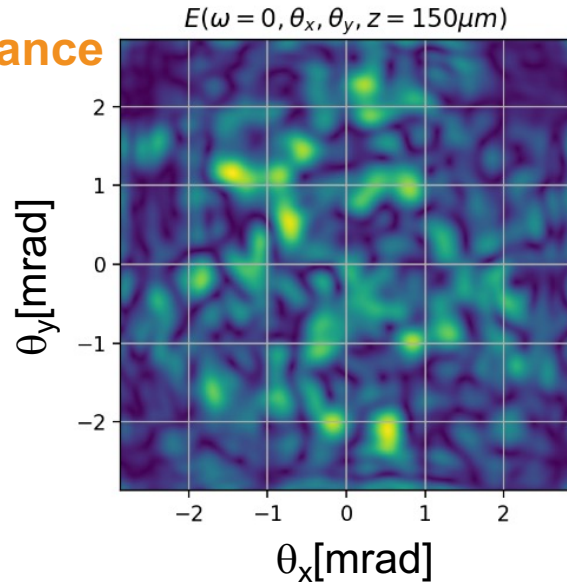


Number of emitted photons at the Cu  $K\alpha_1$  line as a fct. of target thickness for different pump-pulse energy

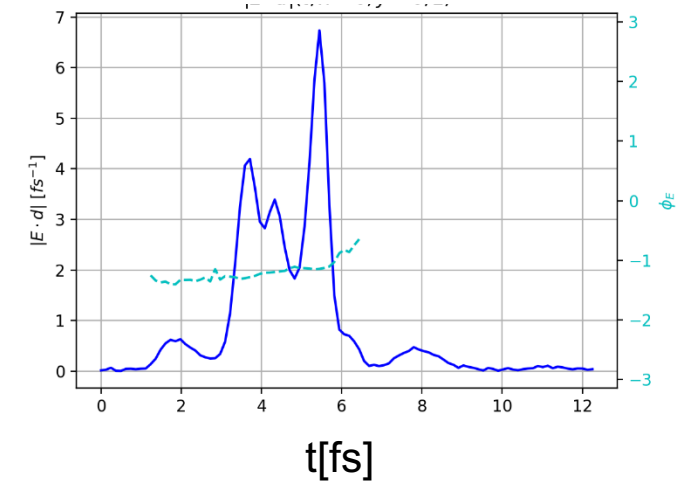
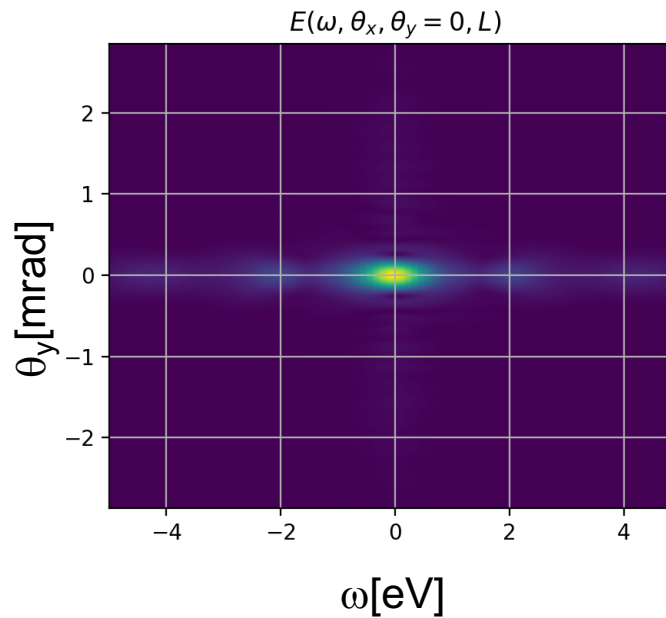
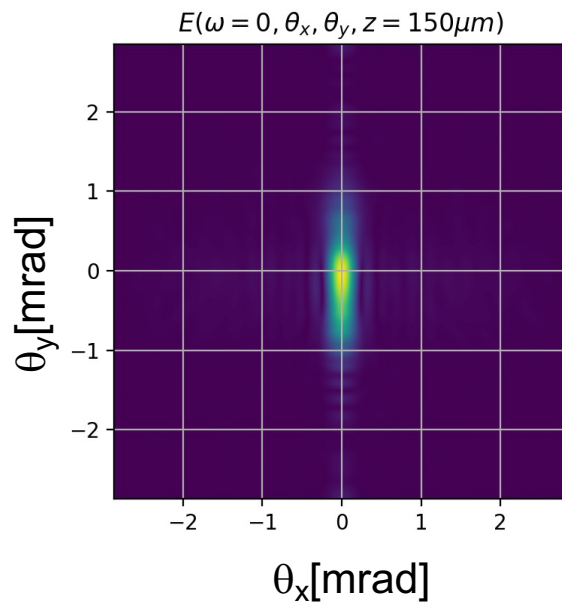


# Evolution of transverse pulse properties

## Superradiance



## After 3 rounds in the cavity:

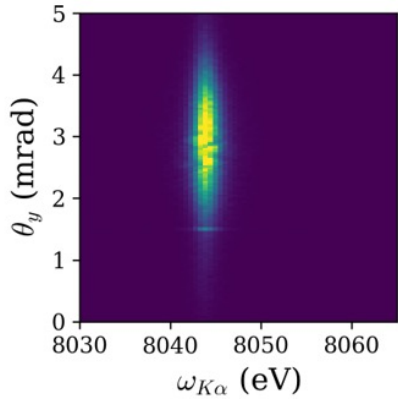


# Seeding x-ray superfluorescence for improved gain

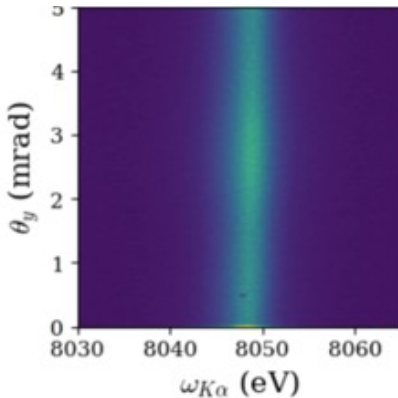
Slide with courtesy to  
A. Halavanau



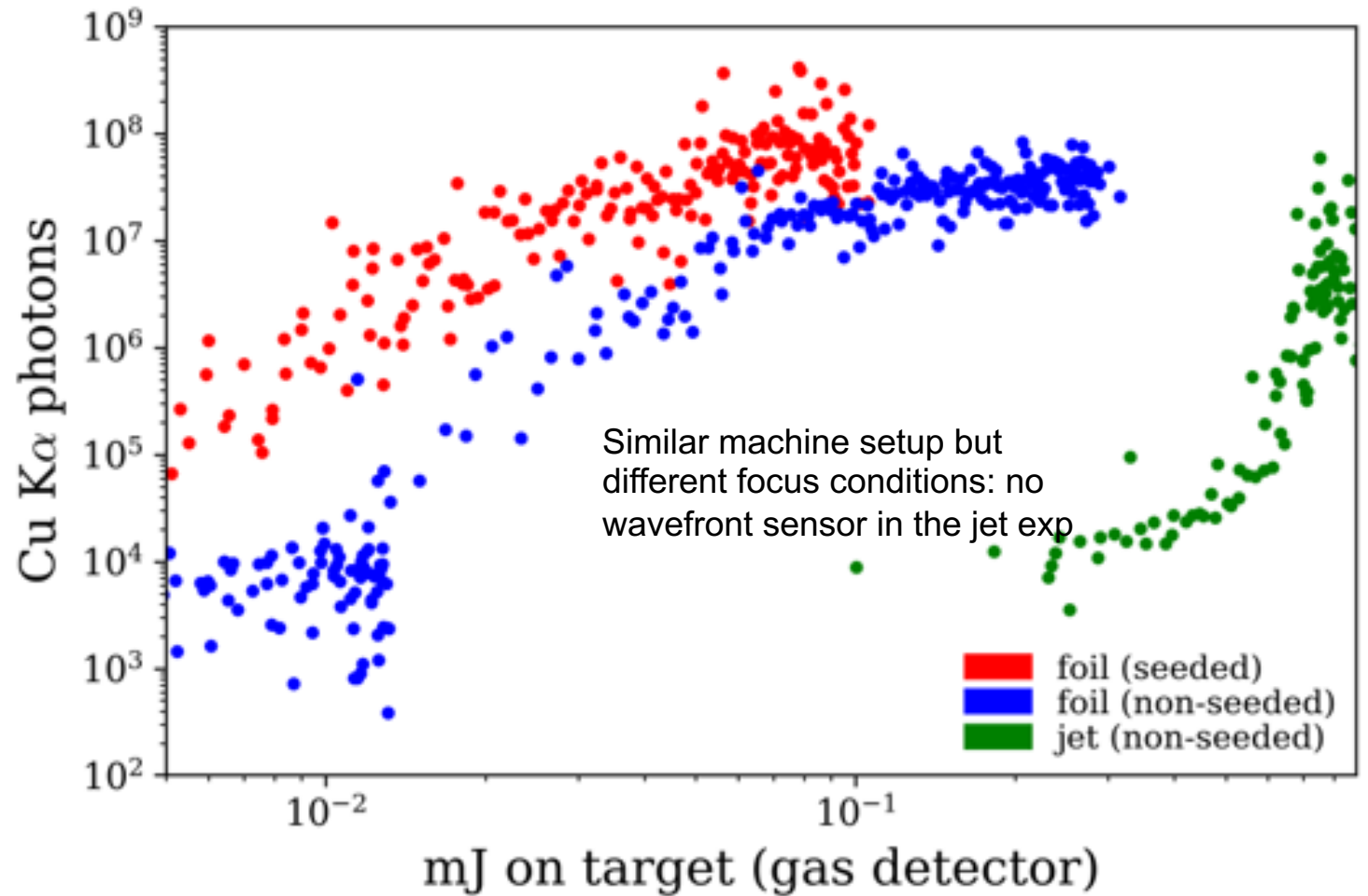
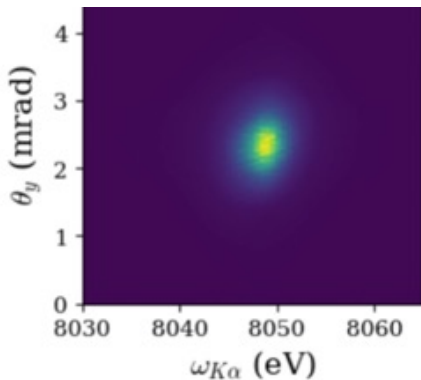
jet (ASE)  
Cu(NO<sub>3</sub>)<sub>2</sub>  
2.4 atoms/nm<sup>3</sup>  
up to 1 mJ pump



Solid foil  
Cu  
86 atoms/nm<sup>3</sup>  
up to 300 uJ pump



Solid foil (seeded)  
Cu  
86 atoms/nm<sup>3</sup>  
up to 100 uJ pump  
10<sup>4</sup>-10<sup>5</sup> seed photons



M. D. Doyle, A. Halavanau al., Optica 10, 513 (2023).

# Summary and Outlook

**XFELs open novel opportunities of nonlinear x-ray science**

## **Inner-shell x-ray lasing**

- Superfluorescence creates sub-fs transform limited x-ray pulses of high intensity
- Application in stimulated emission x-ray spectroscopy
- Source development: X-ray Laser Oscillator

**Open PhD position (theory) and PostDoc position (experiment)**

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Felicie Albert



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