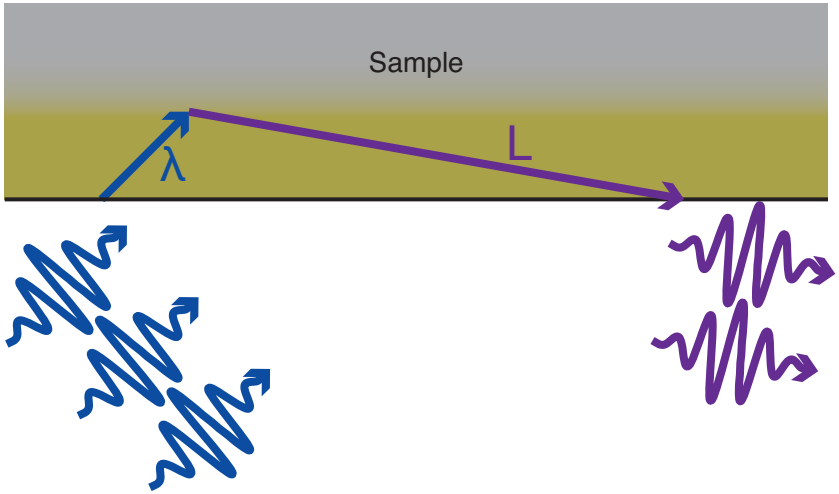
NON-LINEAR SOFT X-RAY OPTICS.



Martin Beye

Internal Workshop on tailored soft X-ray pulses
Villigen, May 11, 2016

HELMHOLTZ ASSOCIATION

DESY

To understand and control functionality

> Correlated materials:
-Metal-to-Insulator Transitions
-Superconductors
-Magnetic switching
-Ferroelectrics

Fradkin, Kivelson, Nat. Phys. 8, 864 (2012)

> Photochemistry /
Ultrafast chemical dynamics:
-Metal-to-Ligand charge transfer
-Artificial photosynthesis
-Solar fuels
-Heterogeneous catalysis

Valence electronic structure at the active site: Soft X-ray spectroscopy

$YBa_2Cu_3O_{6+y}$

Cr^{3+} (aq)

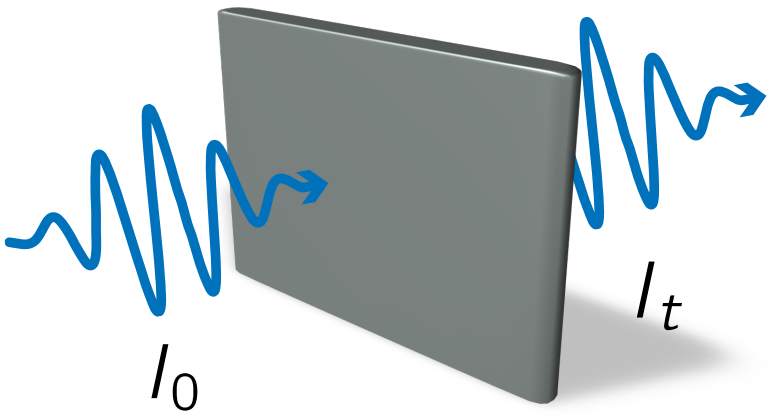
DESY

Motivation and Introduction

Unoccupied valence is „easy“ to do,
(XAS in transmission,
total / partial electron / fluorescence yield)

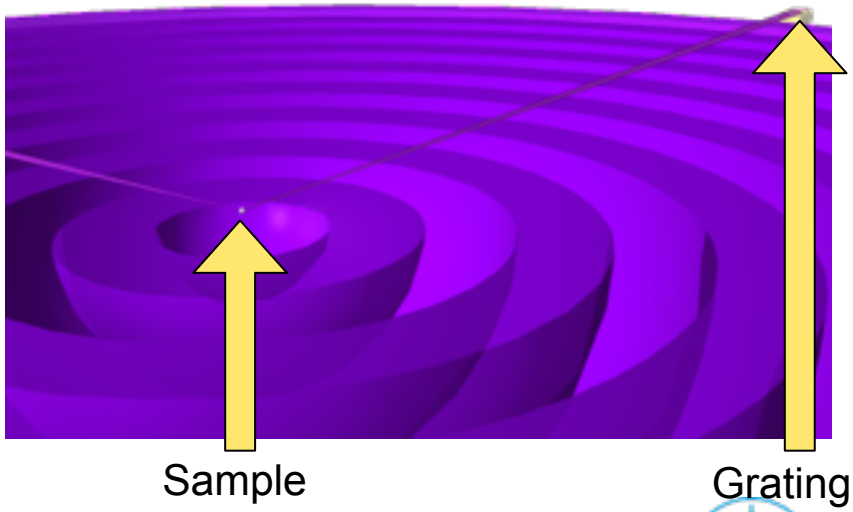
but hard to understand
(core hole in final state)

-> broadband pulses helpful

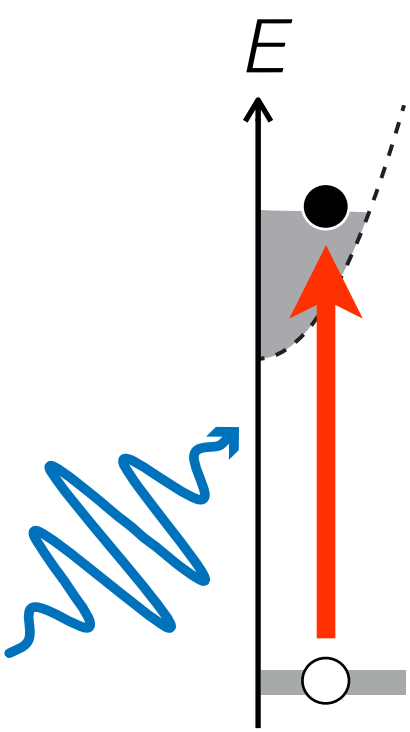


Geometrical Spectrometer Acceptance
10^{-5} to 10^{-4} of 4π

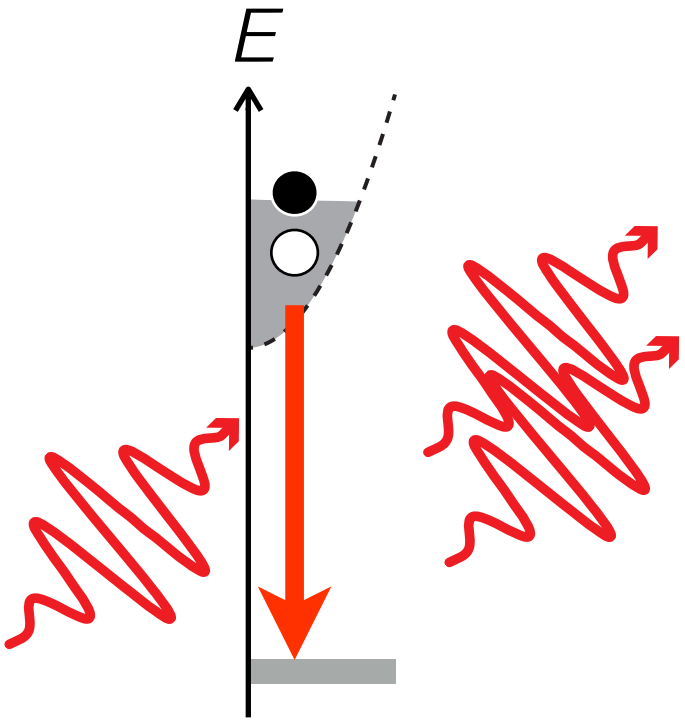
Occupied valence is harder to do
(XES, RIXS):
<1% fluorescence yield
low spectrometer acceptance



Stimulated Emission

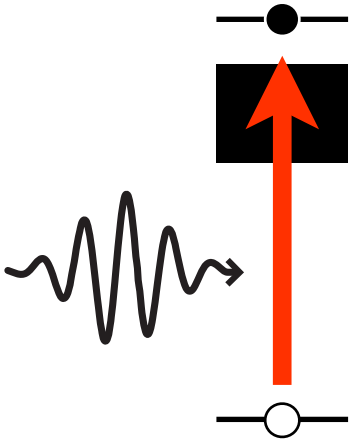


Core
Excitation

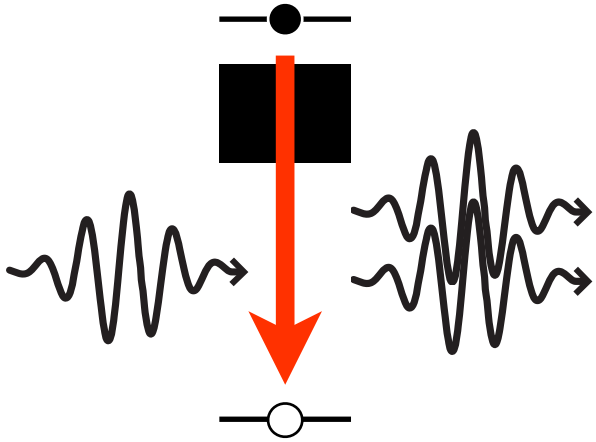


Stimulated
Emission

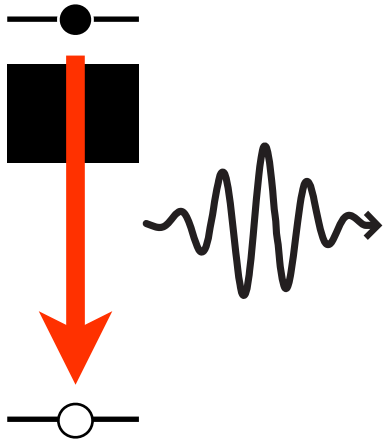
Fundamental Dipole Transitions



Absorption



Stimulated Emission



Spontaneous Emission

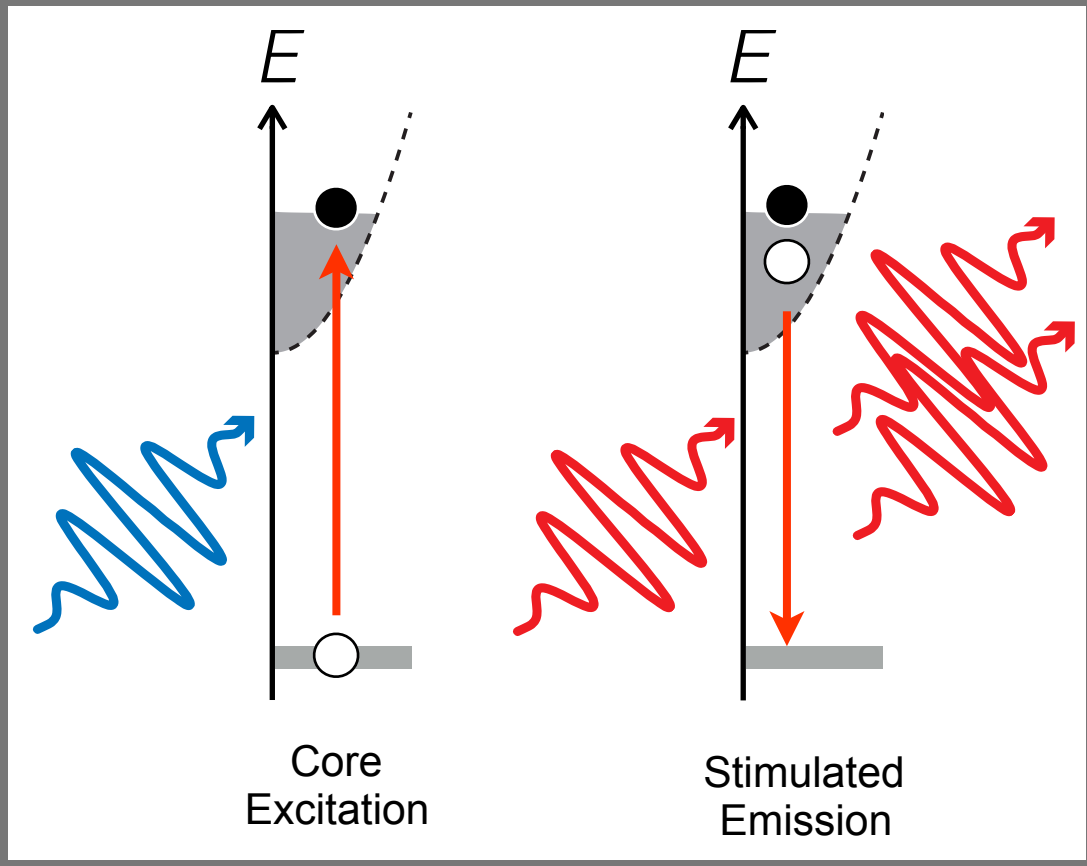
$$\left\langle e, N-1 | \vec{d} | g, N \right\rangle \qquad \left\langle g, N | \vec{d} | e, N-1 \right\rangle \qquad \left\langle g, 1 | \vec{d} | e, 0 \right\rangle$$

$$P_{abs} = N\sigma\rho_g d \qquad P_{stim} = N\sigma\rho_e d$$

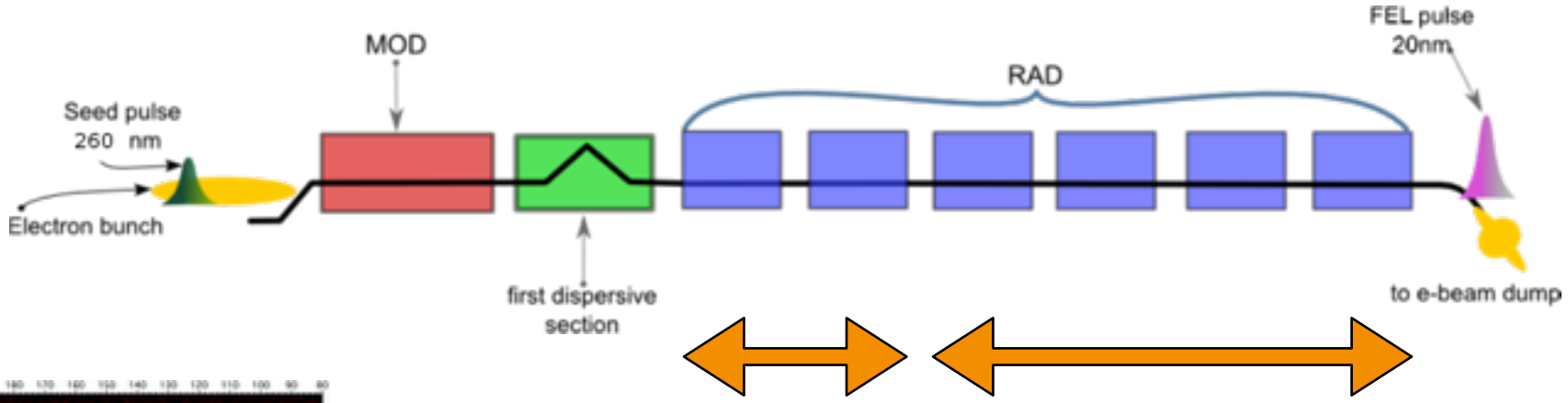
DESY

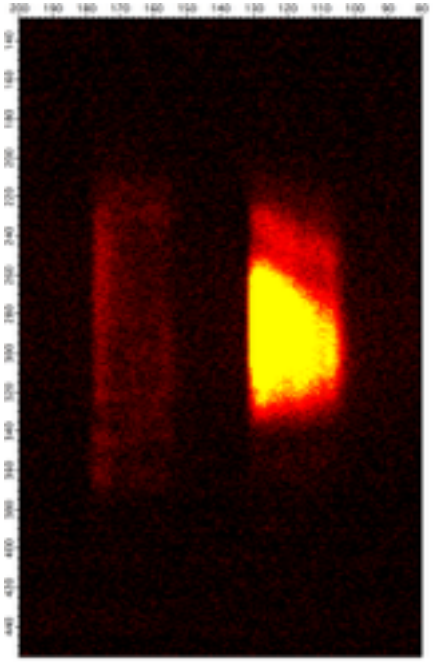
Two-Color X-rays @ FERMI

M. Beye
R. Mincigrucci
E. Giangrisostomi
M. Hantschmann
A. Battistoni
F. Bencivenga
L. Giannessi
N. Mahne
L. Raimondi
C. Svetina
M. Zangrando
C. Masciovecchio
A. Föhlisch
E. Principi



Setup at FERMI



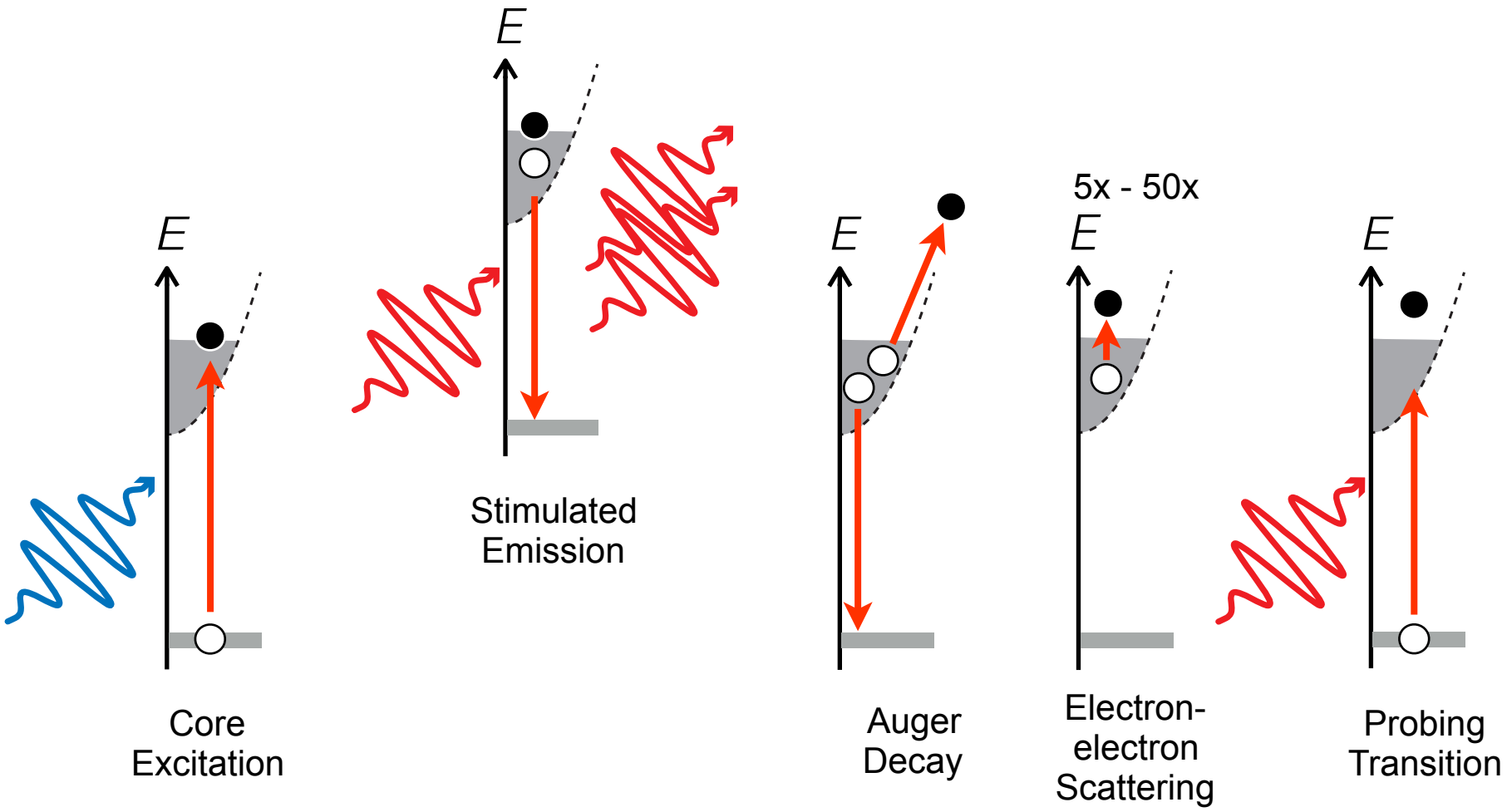


Probe
Harmonic 15:
71eV

Pump
Harmonic 16:
76eV

TIMEX beamline with additional spectrometer
to separate pump and probe in transmitted beam
~µJ pulse energies, ~10µm focal size, ~100fs pulse length

FERMI explanation



Time

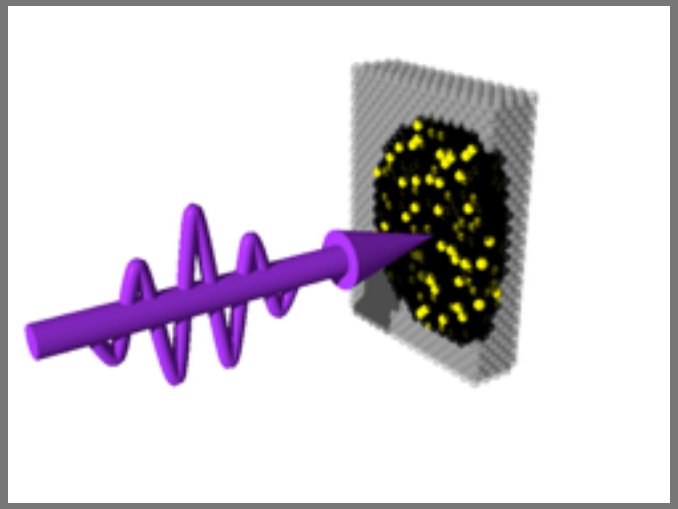
Short pulses are absolutely crucial
for X-ray induced non-linear processes
(because of Auger affecting valence states)

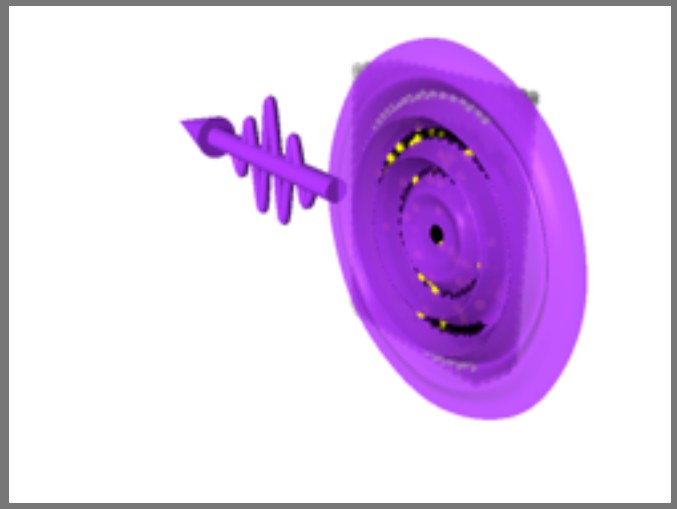
short is ~1/2 core hole lifetime
< 1 fs

maybe SASE is not so bad…
(when time structure is known…)

Amplified Spontaneous Emission @ FLASH

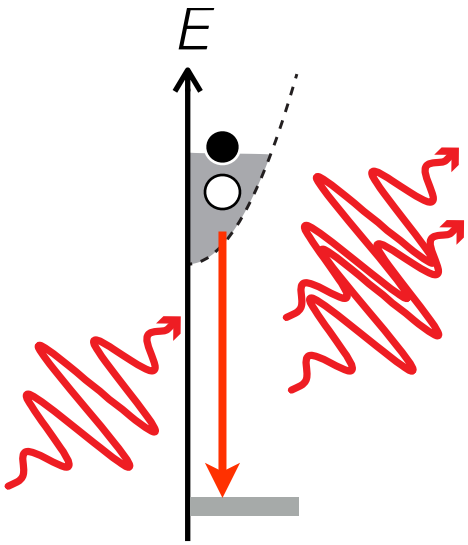
Martin Beye
Simon Schreck
Florian Sorgenfrei
Christoph Trabant
Niko Pontius
Christian Schüssler-Langeheine
Wilfried Wurth
Alexander Föhlisch





Beye et al., Nature 501, 191 (2013)

The trick



Stimulated Emission

$$P_{stim} = \sigma_{stim}\rho_{ch}d \stackrel{!}{=} 1$$

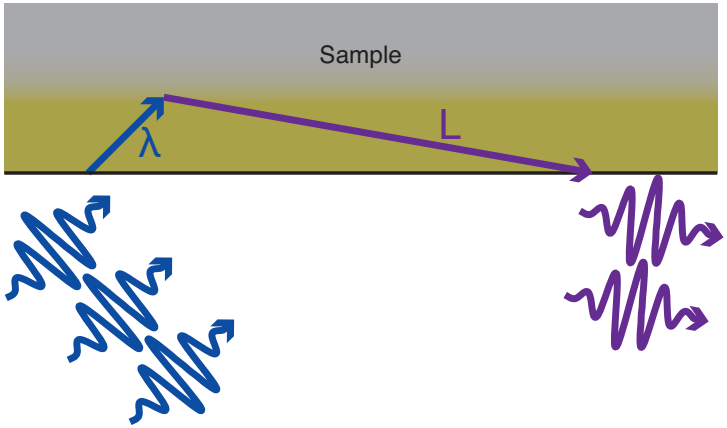
$$\sigma_{stim} \approx \sigma_{abs}$$

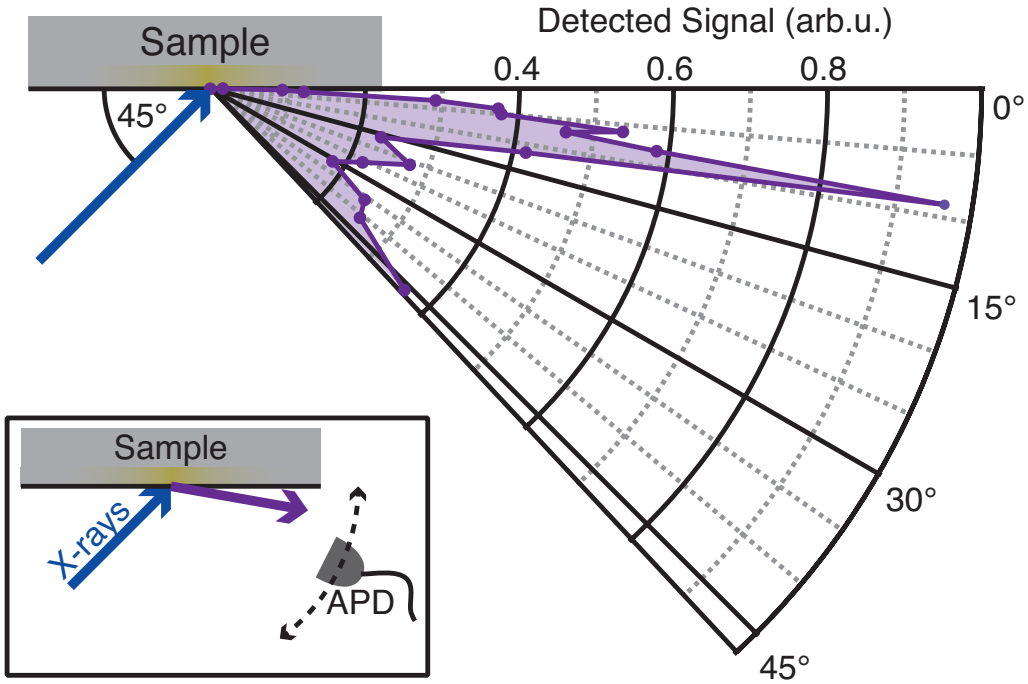
$$\sigma_{abs} = \frac{1}{\lambda_{abs}\rho_{atom}}$$

$$\frac{\rho_{ch}d}{\lambda_{abs}\rho_{atom}} = 1$$

$$\frac{\rho_{ch}}{\rho_{atom}} = \frac{\lambda_{abs}}{d}$$

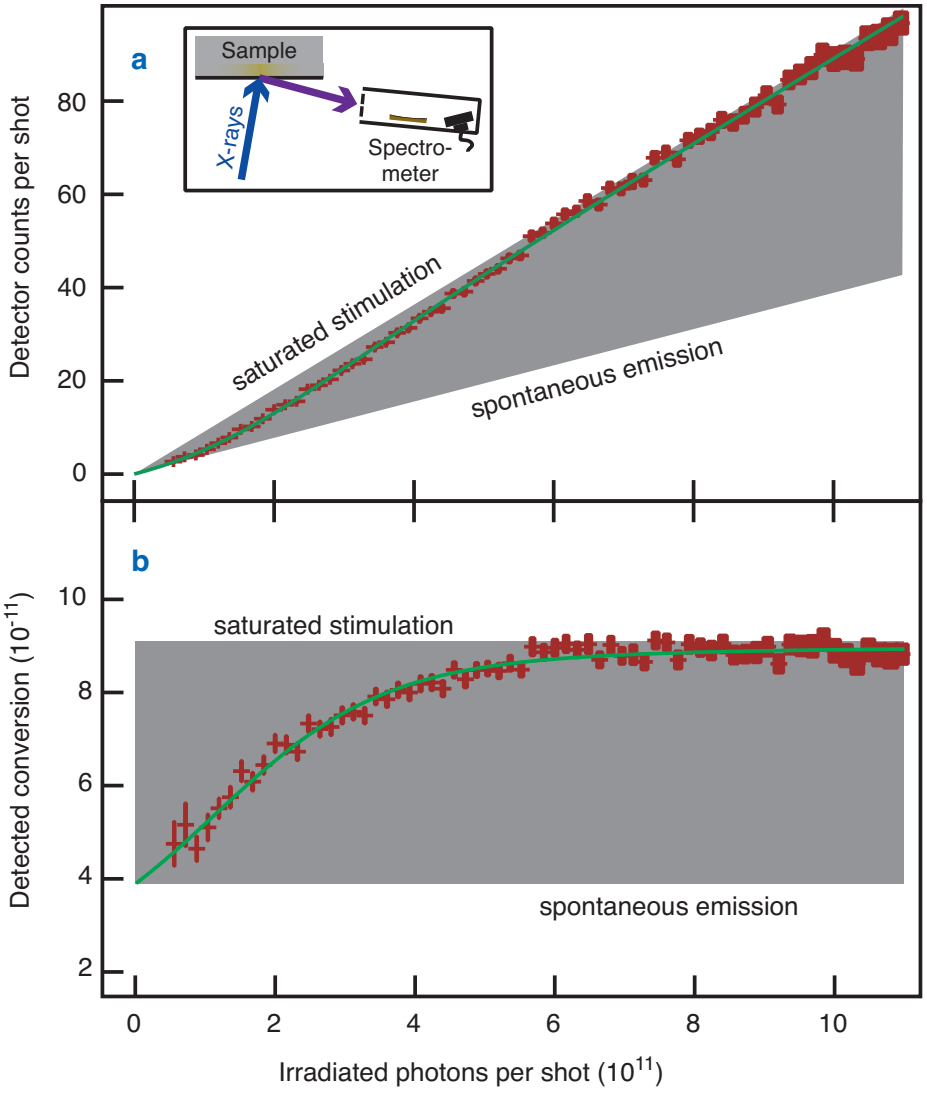
Angular dependence





Si (100), 115eV (10.8nm)
1J/cm^2, 30fs

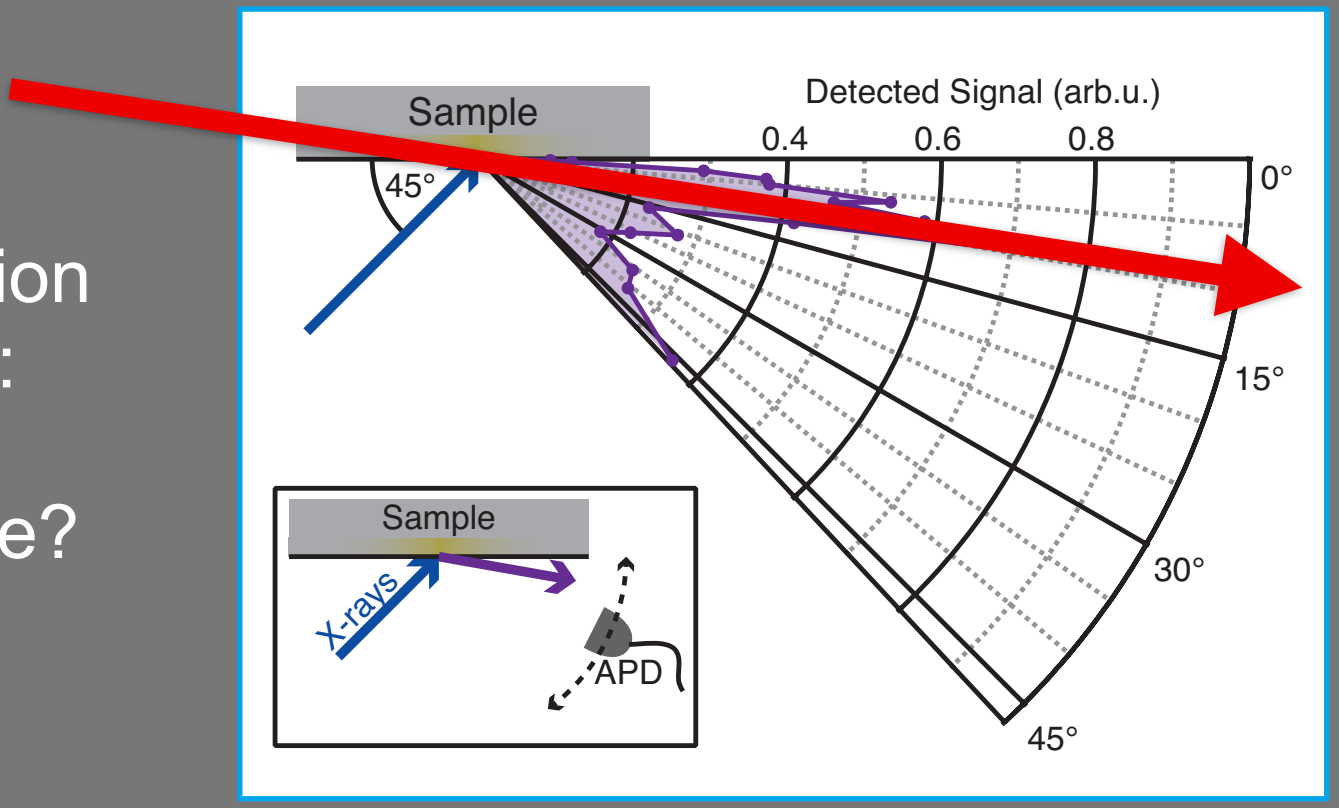
Amplified Spontaneous Emission at Soft X-ray Energies



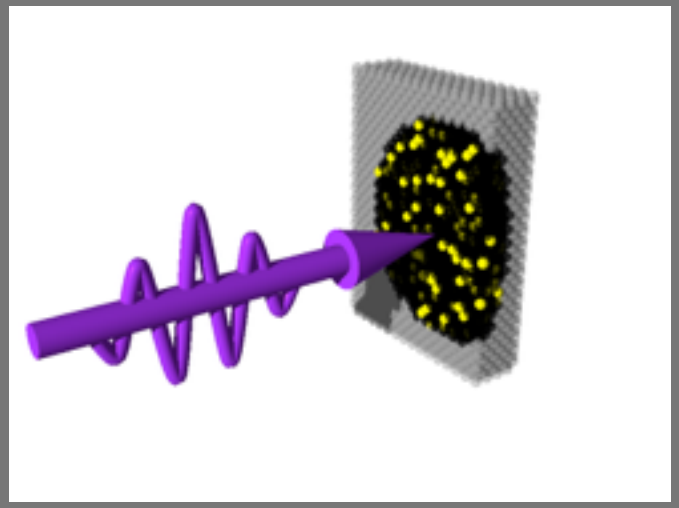
Geometry

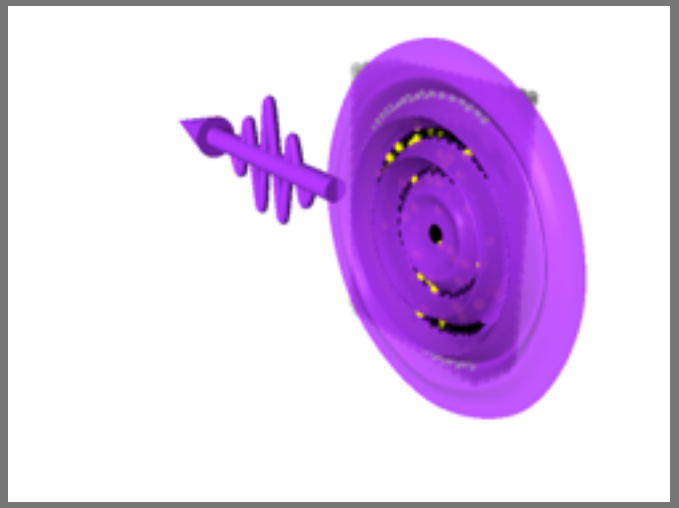
Optimal direction for stimulation:

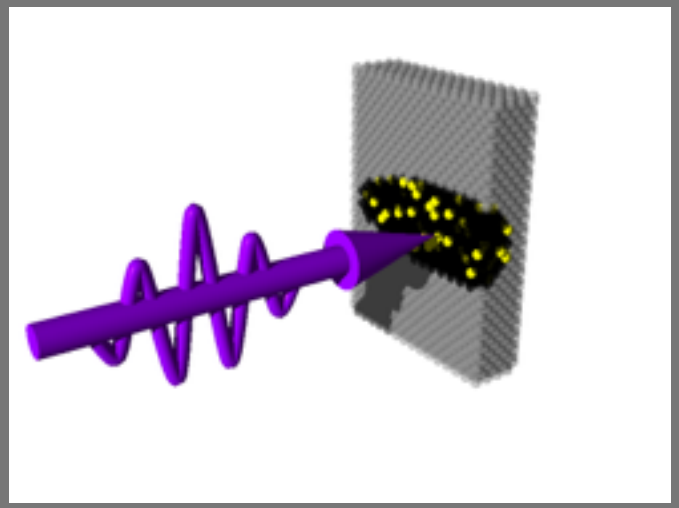
2nd color pulse?

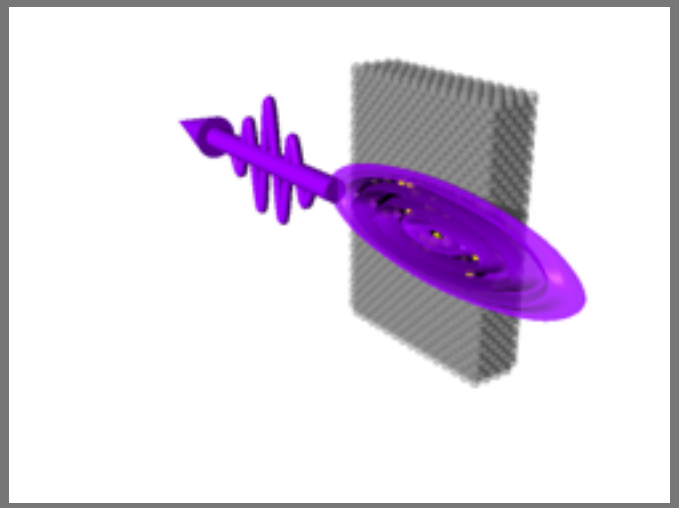


Focus shaping



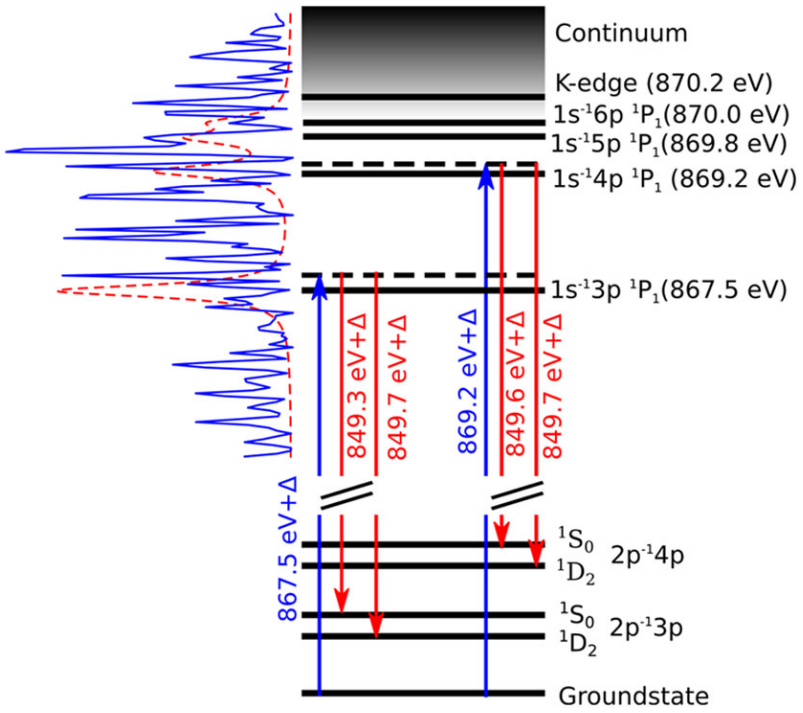






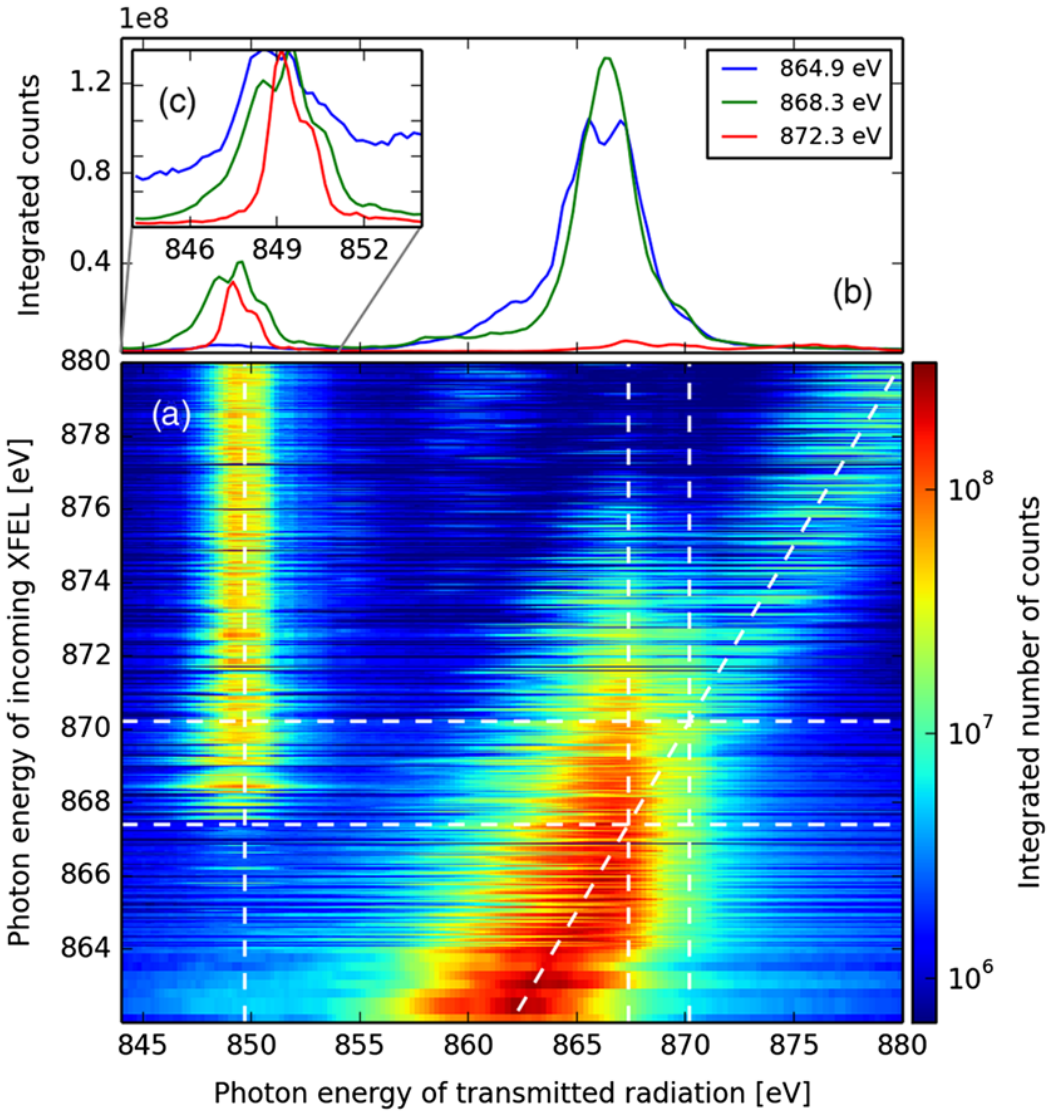
Maybe SASE is not so bad…

Two Colors from SASE bandwidth:



Weninger, C. *et al.* Stimulated Electronic X-Ray Raman Scattering. *Phys. Rev. Lett.* **111,** 233902 (2013)

When spectrum is known…



Weninger, C. *et al.* Stimulated Electronic X-Ray Raman Scattering. *Phys. Rev. Lett.* **111,** 233902 (2013)

THANK YOU FOR YOUR ATTENTION.