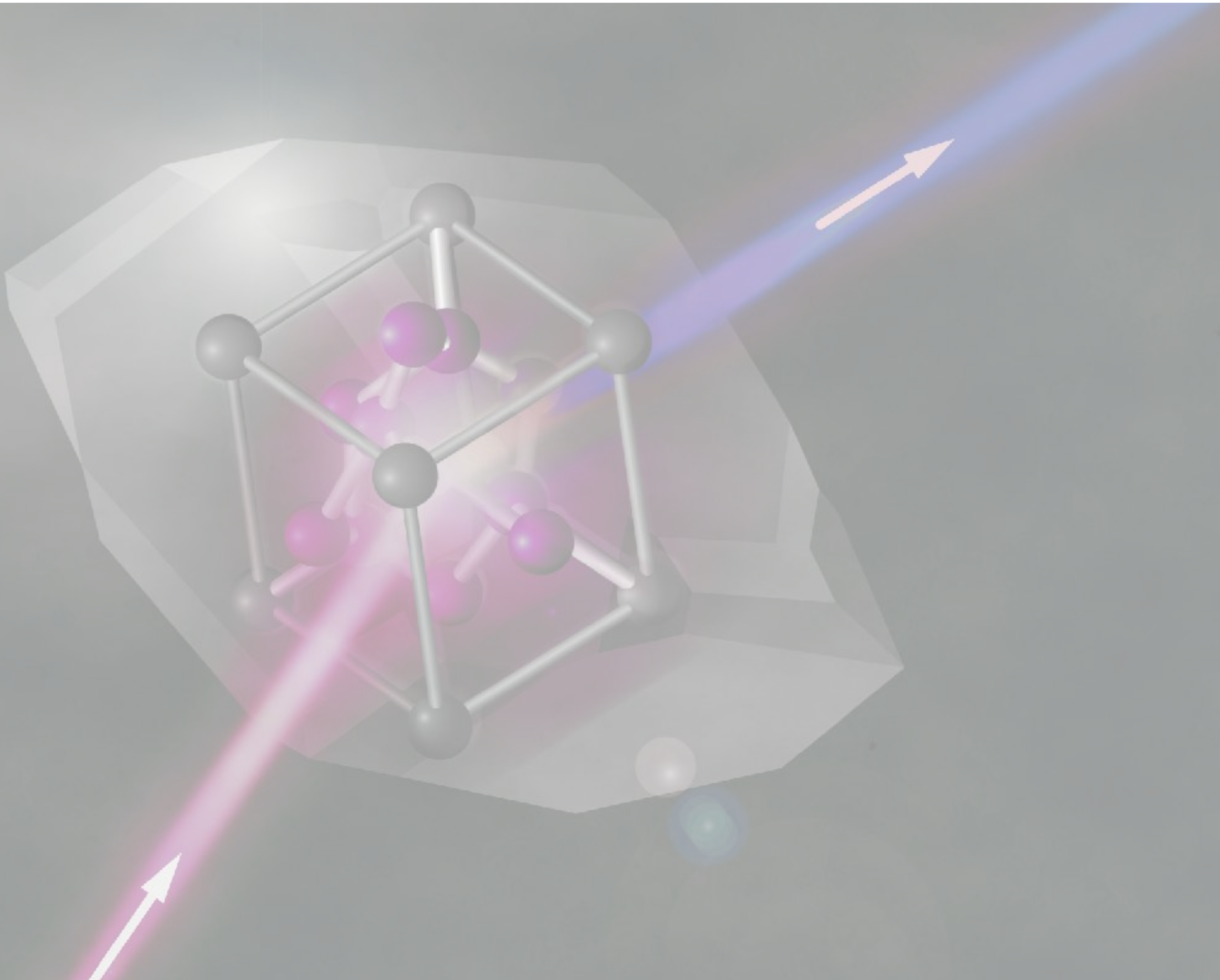
Non-linear processes in X-ray scattering experiments

School on Synchrotron and Free-Electron Laser Methods for Multidisciplinary Applications



Martin Beye,
ICTP Trieste,
May 18, 2018

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

DESY.

Outline

From theoretical field interactions to real experiments

Theory
- From Schrödinger to density matrix
- Field interactions vs. photons: coherence
- Fundamental dipole transitions and the vacuum
- Multi-wave mixing schemes

Experiment
- MUSIX
- Heterodyne detection with X-rays
- Amplified spontaneous emission
- Concurrent processes
- Stimulated emission

Concept of field interactions and coherences

Some theory

How does a laser pulse interact with a quantum system?

Time-dependent Schrödinger equation: $$i\hbar\frac{\partial}{\partial t}|\Psi\rangle = \hat{H}|\Psi\rangle$$

with electric field as perturbation: $$\hat{H} = \hat{H}_0 + \hat{W}(t) \qquad \hat{W}(t) = \hat{\mu}\mathcal{E}\cos(\omega t)$$

Assume Eigenstates: $$\hat{H}_0|n\rangle = E_n|n\rangle$$

General time-dependent (perturbation) solution: $$|\Psi(t)\rangle = \sum_n c_n e^{-iE_n t/\hbar}|n\rangle$$

Concept of field interactions and coherences

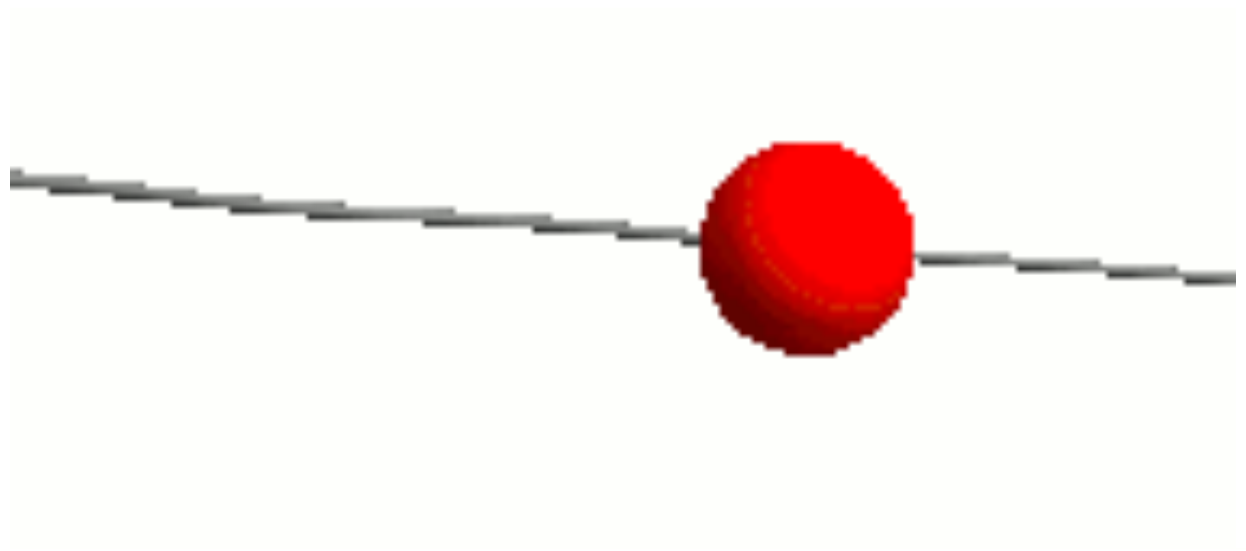
Laser pulse couples ground state and first excited state

After a (short) pulse coupling the ground and first excited states,
the system is in a coherent superposition:

$$|\Psi(t)\rangle = c_0 e^{-iE_0 t/\hbar} |0\rangle + i c_1 e^{-iE_1 t/\hbar} |1\rangle$$

$|0\rangle$ s-orbital

$|1\rangle$ p-orbital



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Concept of field interactions and coherences

Polarisation after such a pulse

$$|\Psi(t)\rangle = c_0 e^{-iE_0 t/\hbar}|0\rangle + ic_1 e^{-iE_1 t/\hbar}|1\rangle$$

$$\begin{aligned} P(t) = \langle\hat{\mu}\rangle = \langle\Psi(t)|\hat{\mu}|\Psi(t)\rangle \\ &= \left(c_0 e^{iE_0 t/\hbar}\langle 0| - ic_1 e^{E_1 t/\hbar}\langle 1|\right)\hat{\mu}\left(c_0 e^{-iE_0 t/\hbar}|0\rangle + ic_1 e^{-iE_1 t/\hbar}|1\rangle\right) \\ &= c_0 c_1 \langle 0|\hat{\mu}|1\rangle \sin(\omega_{01} t) + c_0^2\langle 0|\hat{\mu}|0\rangle + c_1^2\langle 1|\hat{\mu}|1\rangle \\ &\equiv c_0 c_1 \mu_{01}\sin(\omega_{01} t) \\ &\propto +\mu_{01}^2 \sin(\omega_{01} t) \end{aligned}$$

Polarisation oscillates with $\omega_{01} = (E_1 - E_0)/\hbar$

radiates field proportional to $-\mu_{01}^2\cos(\omega_{01} t)$

Out of phase with exciting field: exciting field is „attenuated“ (destructive interference)

Concept of field interactions and coherences

The radiation from this coherent state is a second interaction with the field and transfers population

$$|\Psi(t)\rangle = c_0 e^{-iE_0 t/\hbar}|0\rangle + i c_1 e^{-iE_1 t/\hbar}|1\rangle$$

This state can be represented in a density matrix.

Directly after the laser pulse, without dephasing or population relaxation, this yields:

$$\rho_{00}(t) = c_0^2 = \text{const.}$$
$$\rho_{11}(t) = c_1^2 = \text{const.}$$
$$\rho_{01}(t) = -i c_0 c_1 e^{+i\omega_{01} t}$$
$$\rho_{10}(t) = i c_0 c_1 e^{-i\omega_{01} t}$$

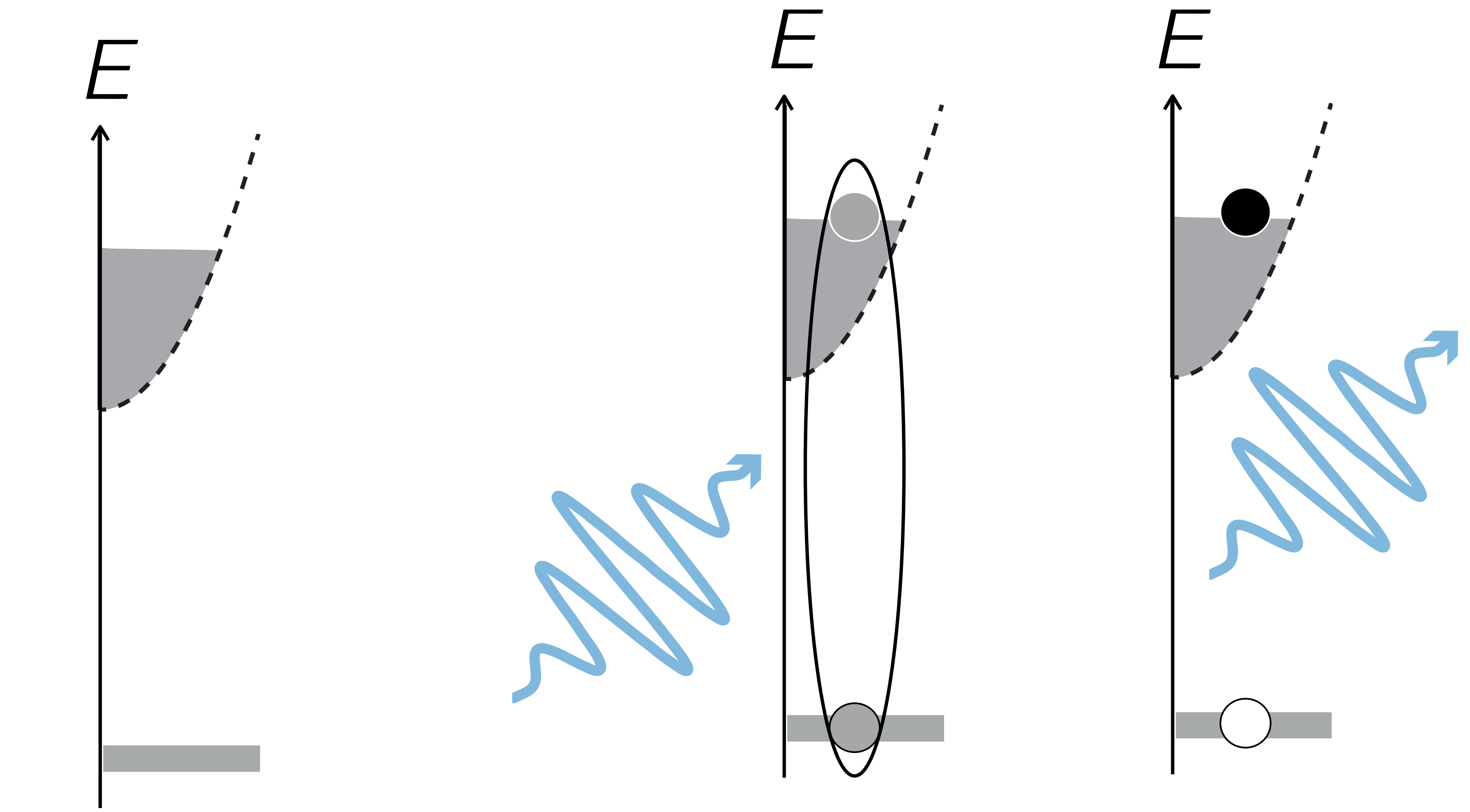
for maximal pulse strength:

$$\rho(t=0) = \begin{pmatrix} 1/2 & -i/2 \\ +i/2 & 1/2 \end{pmatrix}$$

Diagonal elements are called „populations“.
Off-diagonal elements are „coherences“.

Field interactions and level schemes (population spectroscopies)

Described one field interaction. Absorption becomes real with second field interaction



0. Ground state

$\rho_{11} = 0$

$\rho_{00} = 1$

1. Create coherence

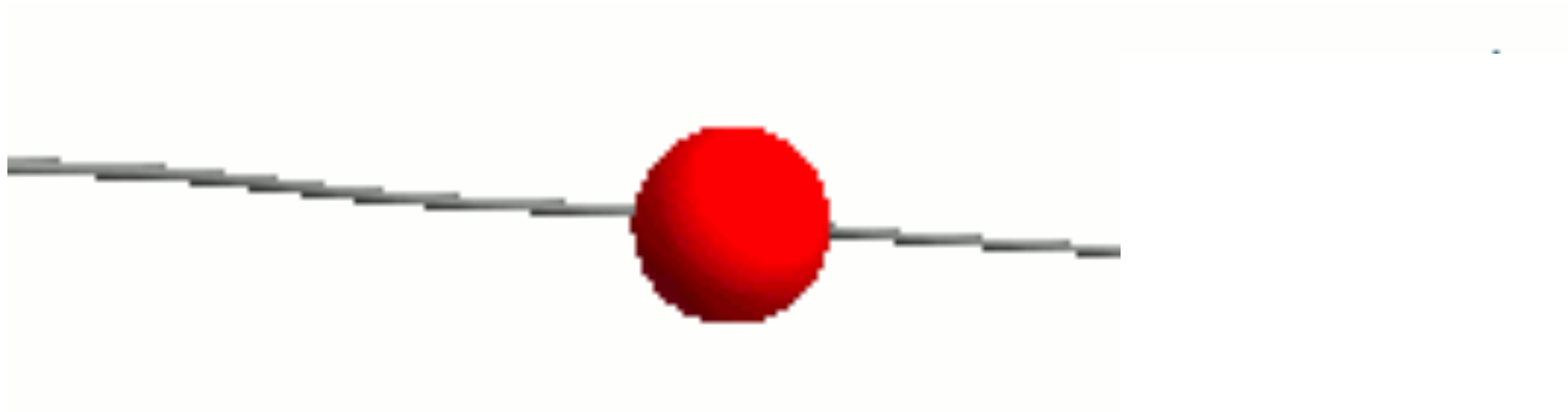
$\rho_{11} = 1/2$

$\rho_{00} = 1/2$

2. Transfer population

$\rho_{11} = 1$

$\rho_{00} = 0$



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What is a „coherence“?

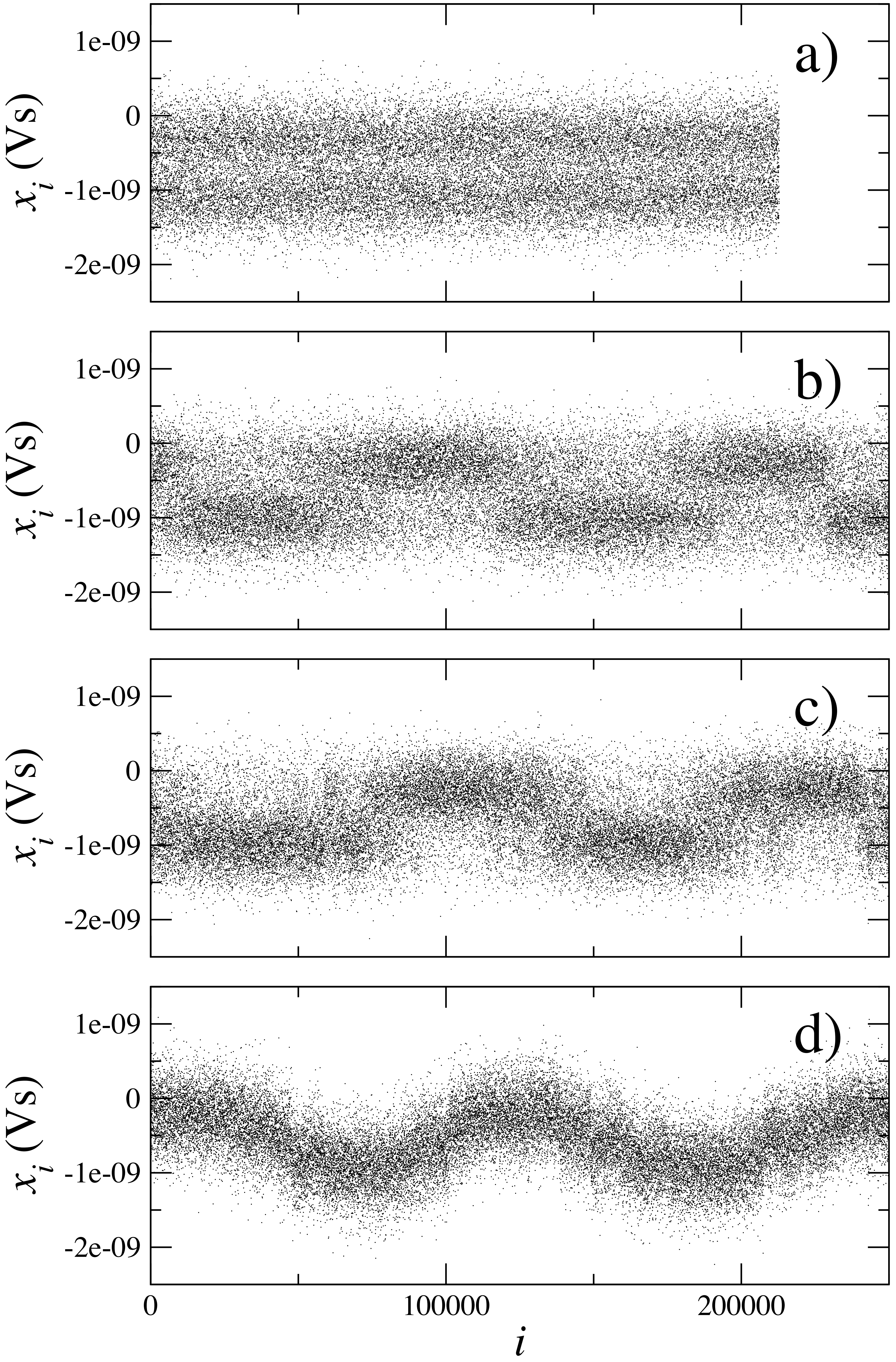
From quantum optics to experiment: Sidebands vs streaking

Coherence is a well-defined phase relation.

Quantum optics of photon / electric field

Coherent state:
- Well-defined (electric field) phase
- Undefined photon number

Fock state (number state):
- Undefined phase
- Well-defined photon number



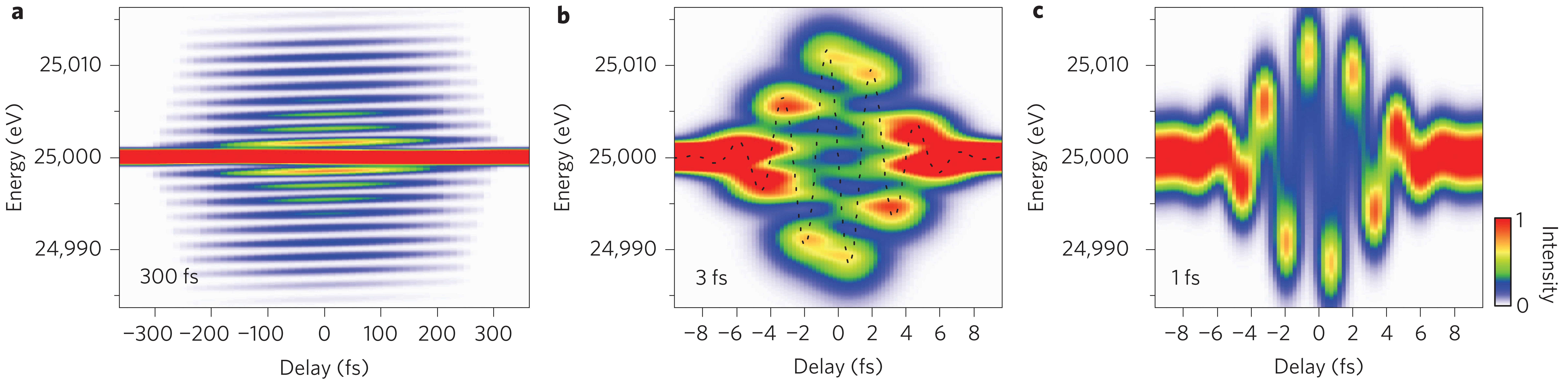
Bellini, Zavatta, Viciani, Proc of SPIE 5866, 278 (2005)

What is a „coherence“?

From quantum optics to experiment: Sidebands vs streaking

Interaction of an electric field (photons) with electrons:

Kirchner et al., Nat. Phot, 8, 52 (2013)



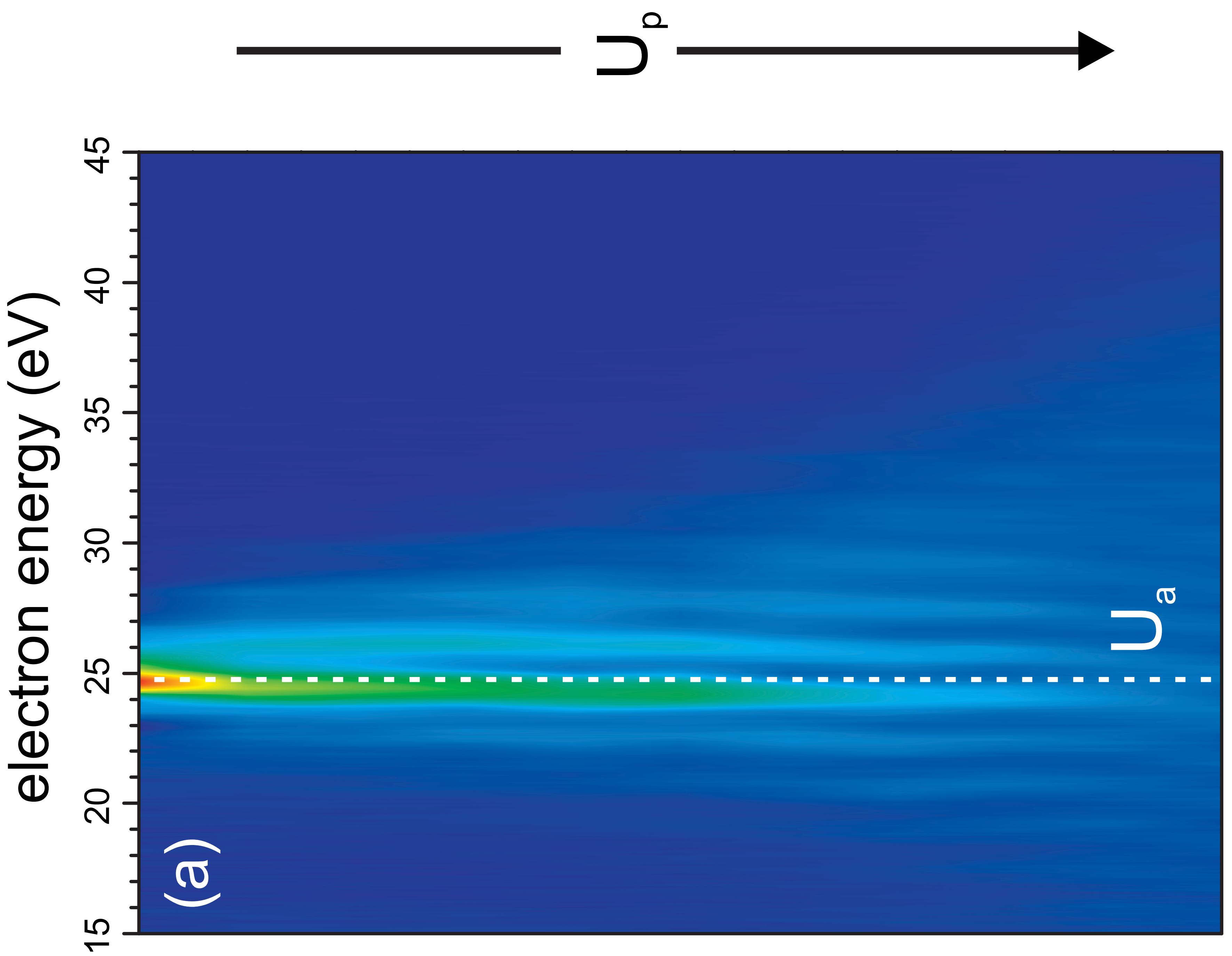
Sidebands:
Well-defined photon number
Undefined phase

Streaking:
Undefined photon number
Well-defined phase

What is a „coherence“?

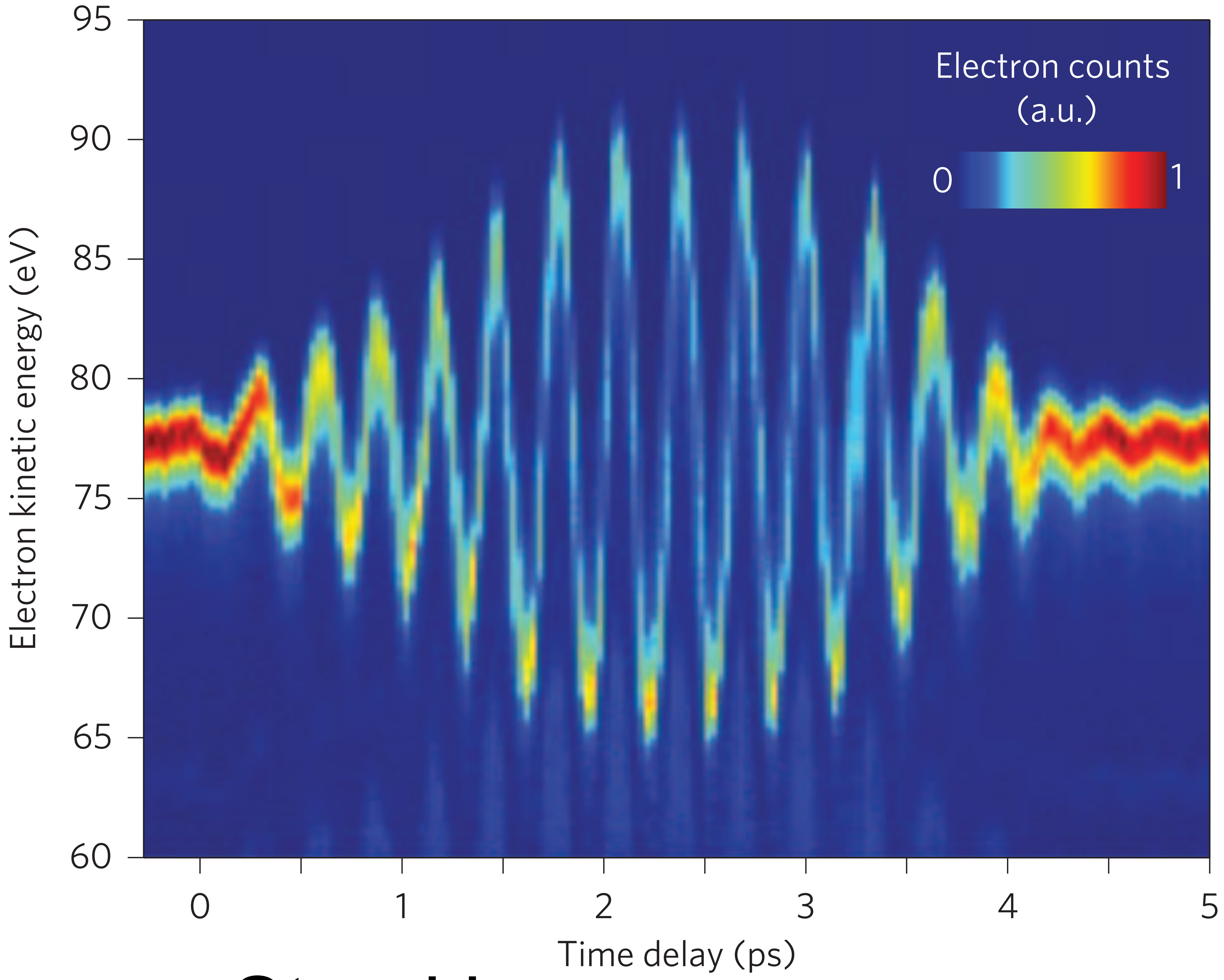
From quantum optics to experiment: Sidebands vs streaking

Experimental results from FLASH



Radcliffe et al., NJP 14, 043008 (2012)

Frühling et al., Nat. Phot. 3, 523 (2009)



Sidebands:
Well-defined photon number
Undefined phase

Streaking:
Undefined photon number
Well-defined phase

Field interaction vs. photon interaction

One photon interaction can be regarded as two field interactions ($cc^* = |c|^2$)

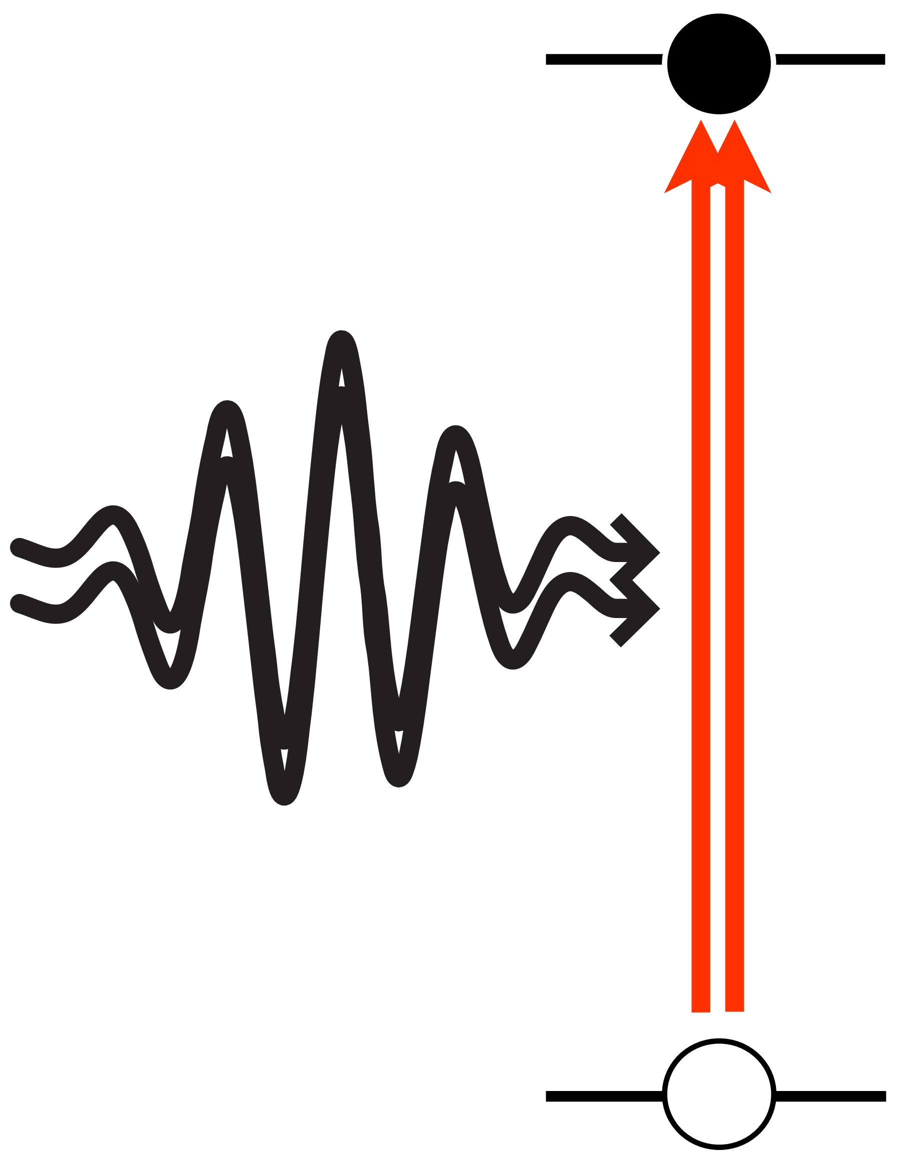
In the photon picture the phase is lost.

Population is transferred with two (identical) field interactions.

(Synchrotron spectroscopies are nearly always „population spectroscopies“:
-incoherent (no phase relation)
-well-defined number of transitions.
Coherence only becomes relevant at FELs:
-large number of coherent photons)

Fundamental dipole interactions / Photon transitions

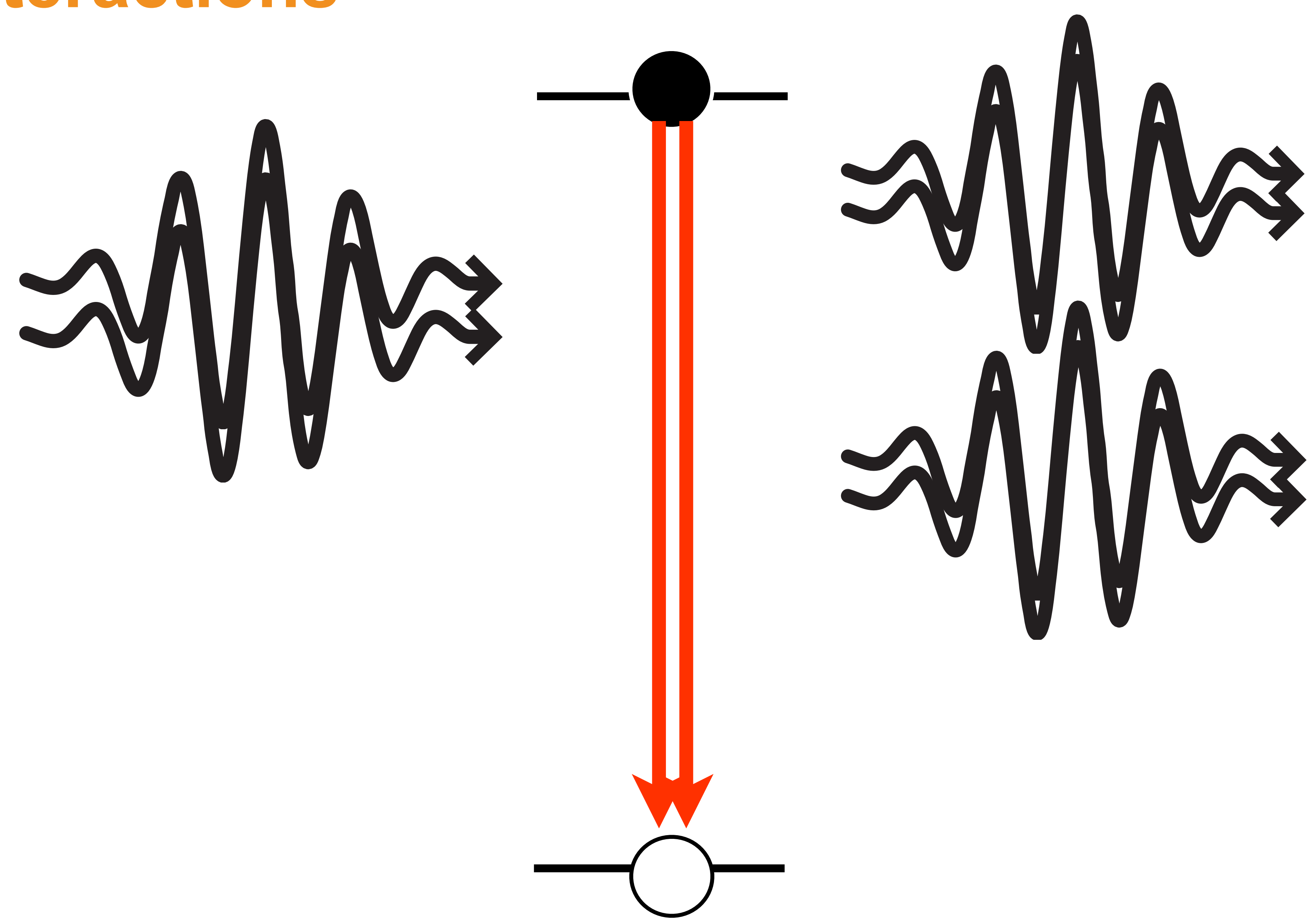
„Two-wave mixing“, two field interactions



Absorption

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

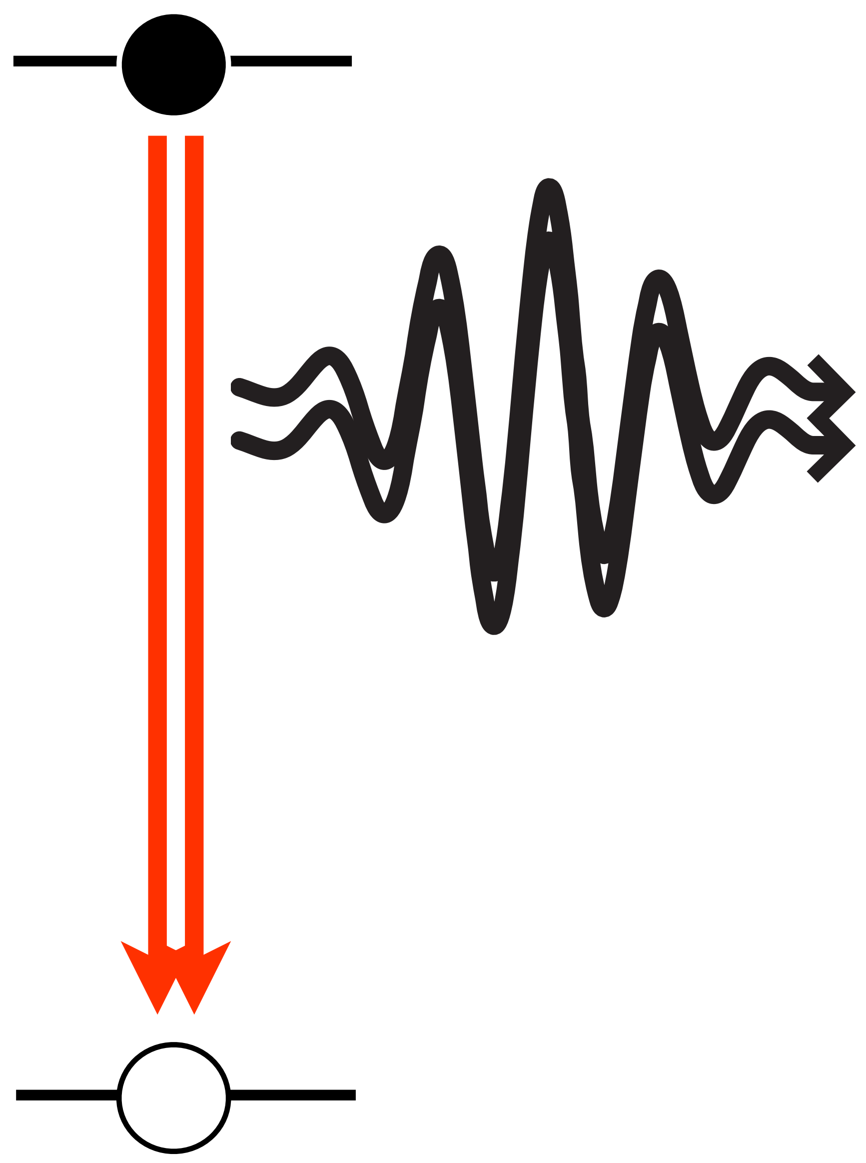
$$P_{abs} = N \sigma \rho_g d$$



Stimulated Emission

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

$$P_{stim} = N \sigma \rho_e d$$

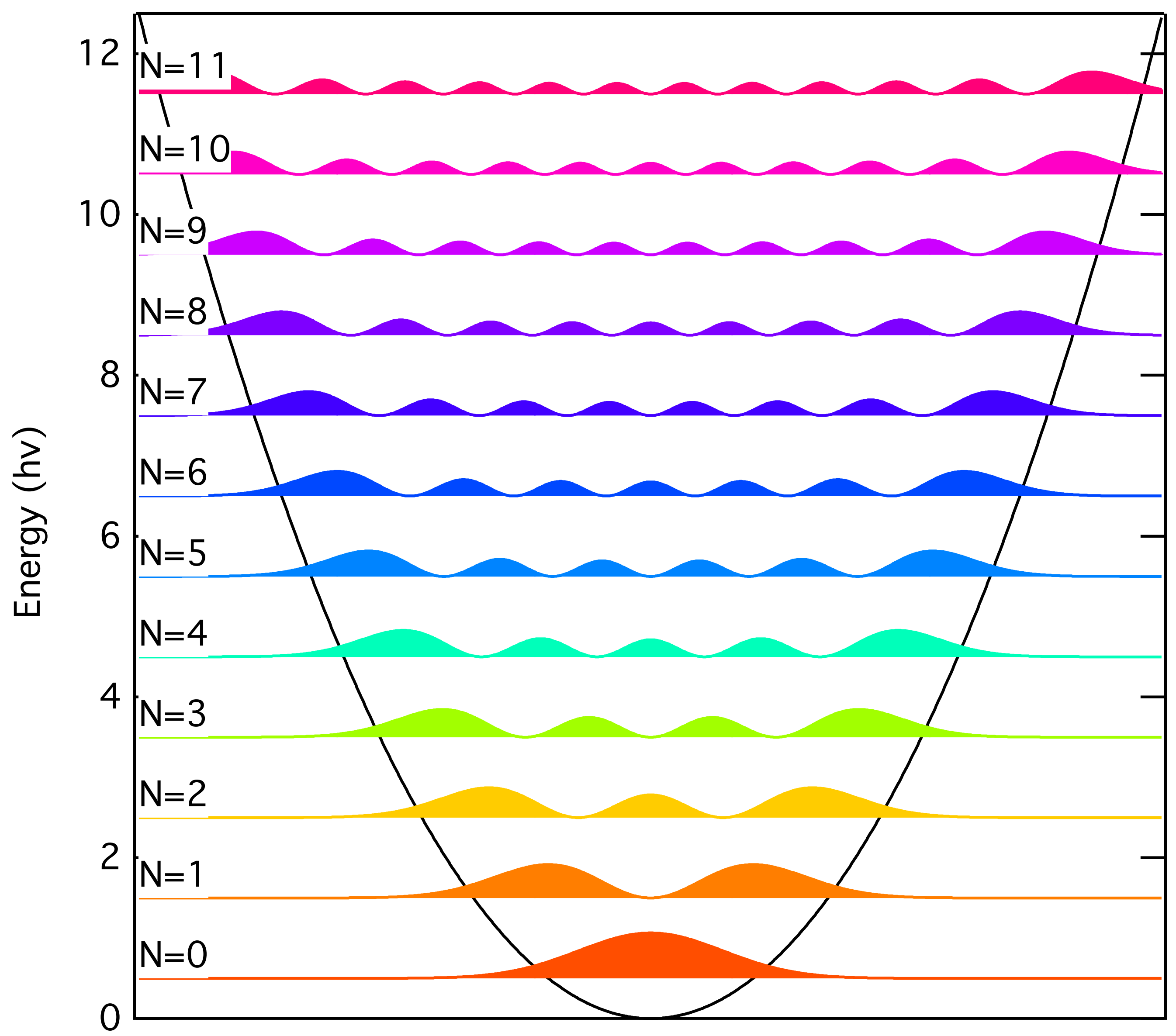


Spontaneous Emission

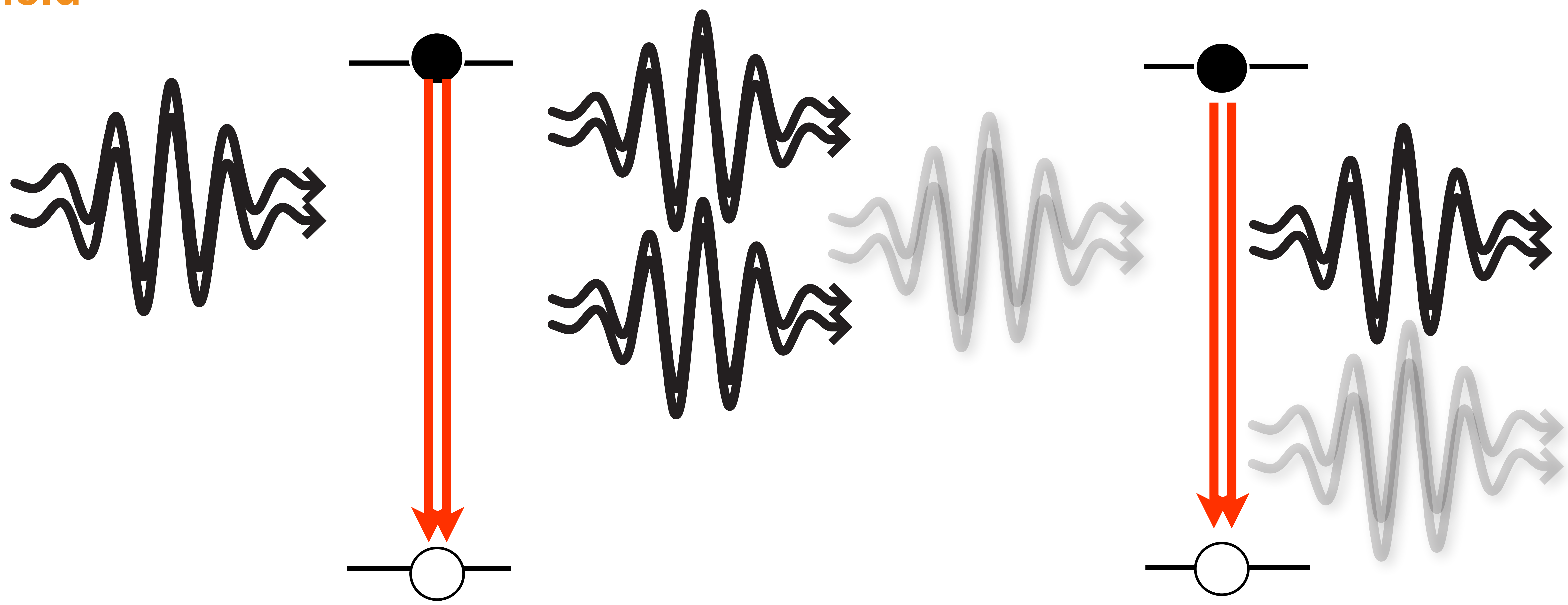
$$\left\langle g, 1 \middle| \vec{d} \middle| e, 0 \right\rangle$$

Spontaneous emission

Is stimulated by the zero-point field



$$E_n = \hbar\omega\left(N + \frac{1}{2}\right)$$



Stimulated Emission

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

Spontaneous Emission

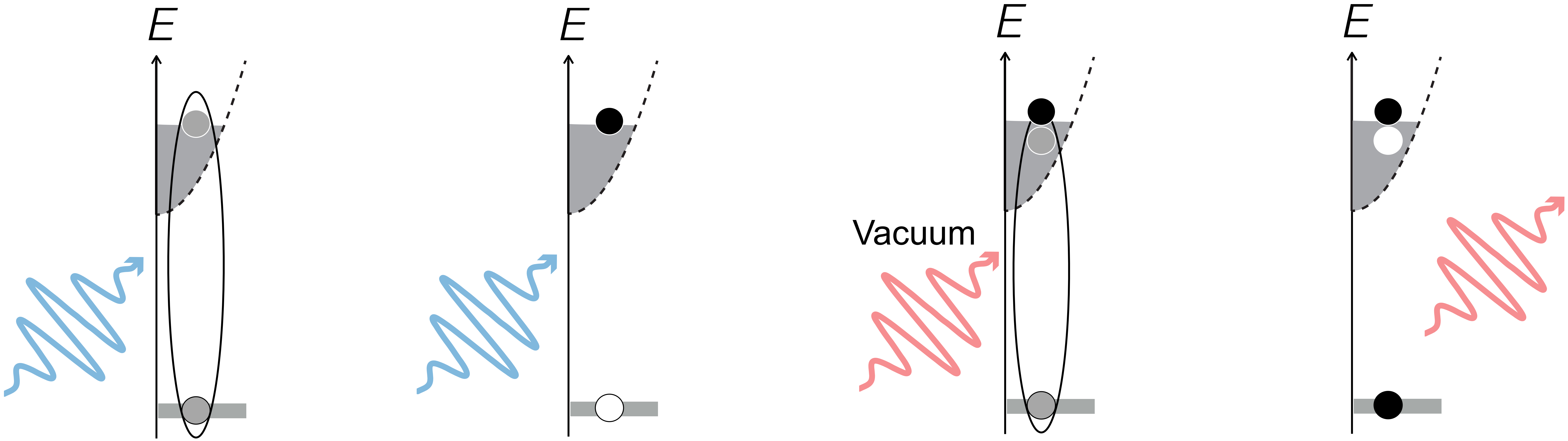
$$\left\langle g, 1|\vec{d}|e, 0\right\rangle$$

$$P_{stim} = N\sigma\rho_e d > P_{spon} = N_{vac}\sigma\rho_e d$$

$$N > N_{vac}$$

Everything is four-wave mixing

RIXS / XES (population spectroscopies - incoherent, zero-point field)



1. Create coherence

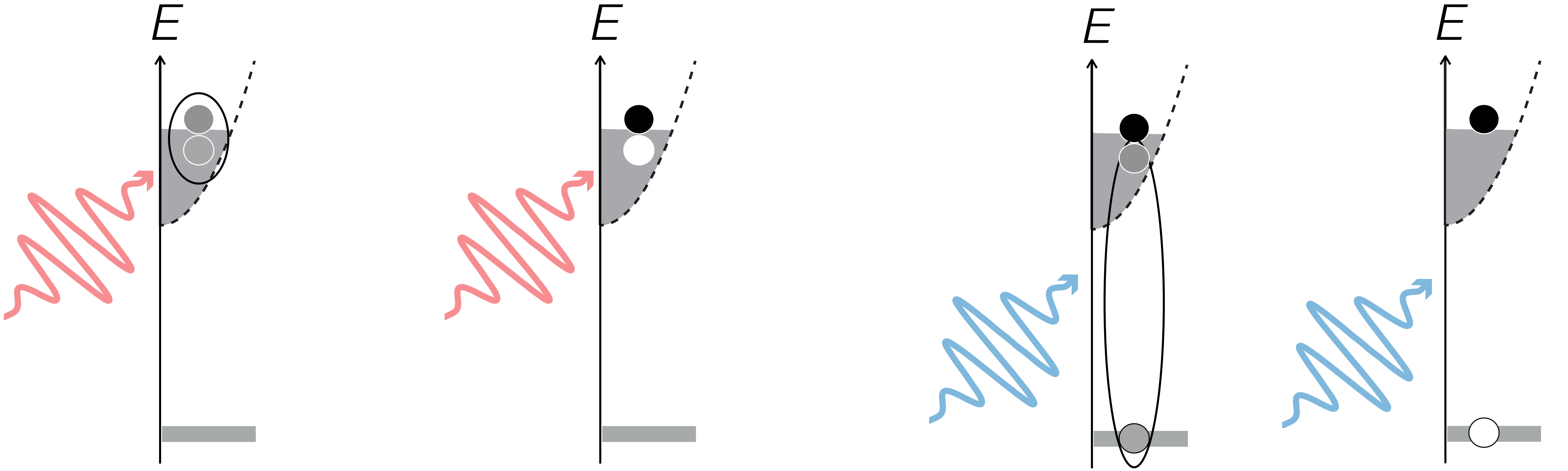
2. Transfer population

3. Create coherence

4. Emit photon

Everything is four-wave mixing

Pump probe—XAS (population spectroscopies - incoherent)



1. Create coherence

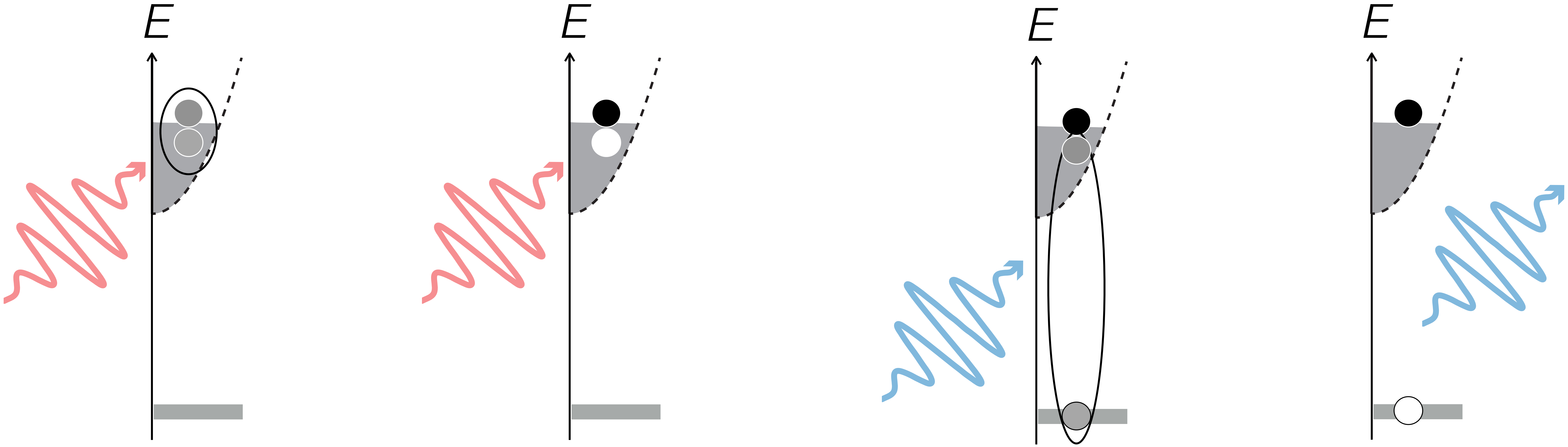
2. Transfer population

3. Create coherence

4. Absorb photon

Everything is four-wave mixing

Claudio's transient gratings (but now coherent!)



1. Create coherence

2. Transfer population (spatially modulated)

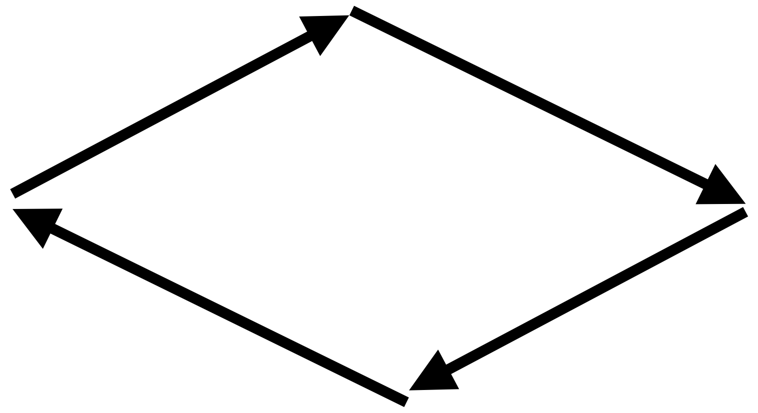
3. Create coherence

4. Scatter photon

Everything is four-wave mixing

Many more:
CARS (coherent anti-Stokes Raman spectroscopy)
CSRS (coherent Stokes Raman spectroscopy)
Stimulated Raman
Inverse Raman
…

Coherent spectroscopies need phase matching!
k-vectors need to add up
> condition for angles
> angular separation of signal possible
> angular focussing of signal



Everything is four-wave mixing

Polarisation expansion

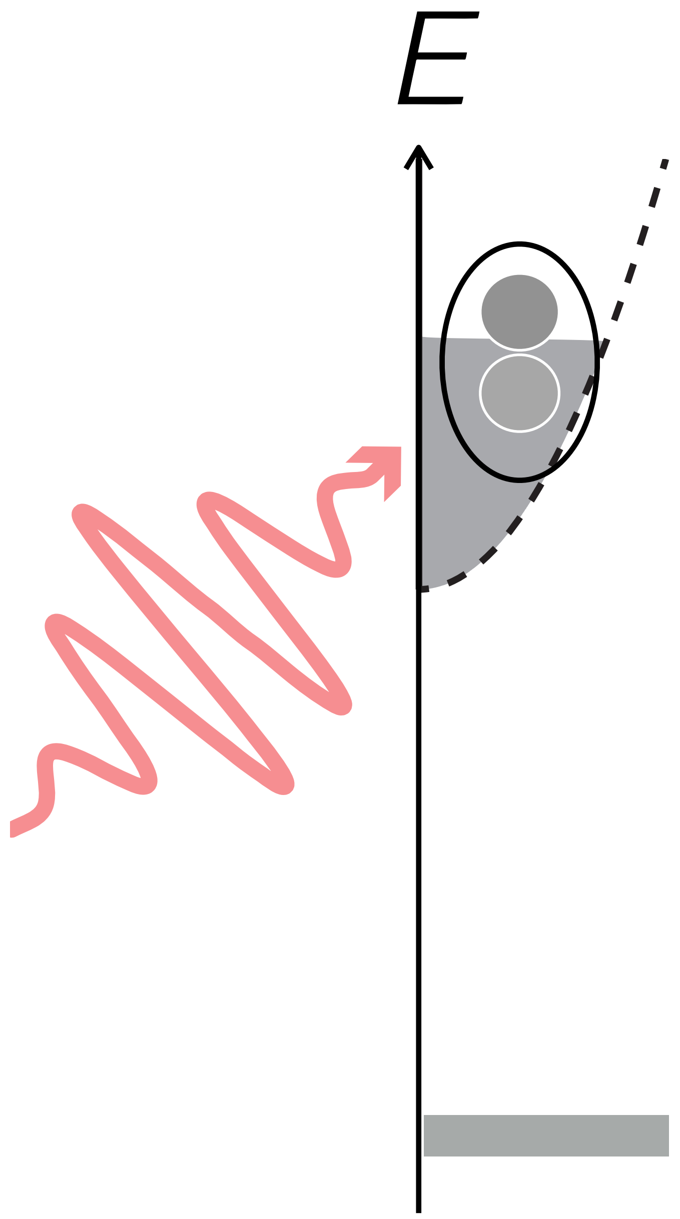
from Claudio:

$$P = \varepsilon_0 \cdot \left[\left(\sum_i \chi^{(1)} \cdot \boldsymbol{E}_i \right) + \left(\sum_{i,j} \chi^{(2)} \cdot \boldsymbol{E}_i \cdot \boldsymbol{E}_j \right) + \left(\sum_{i,j,k} \chi^{(3)} \cdot \boldsymbol{E}_i \cdot \boldsymbol{E}_j \cdot \boldsymbol{E}_k \right) + \cdots \right]$$

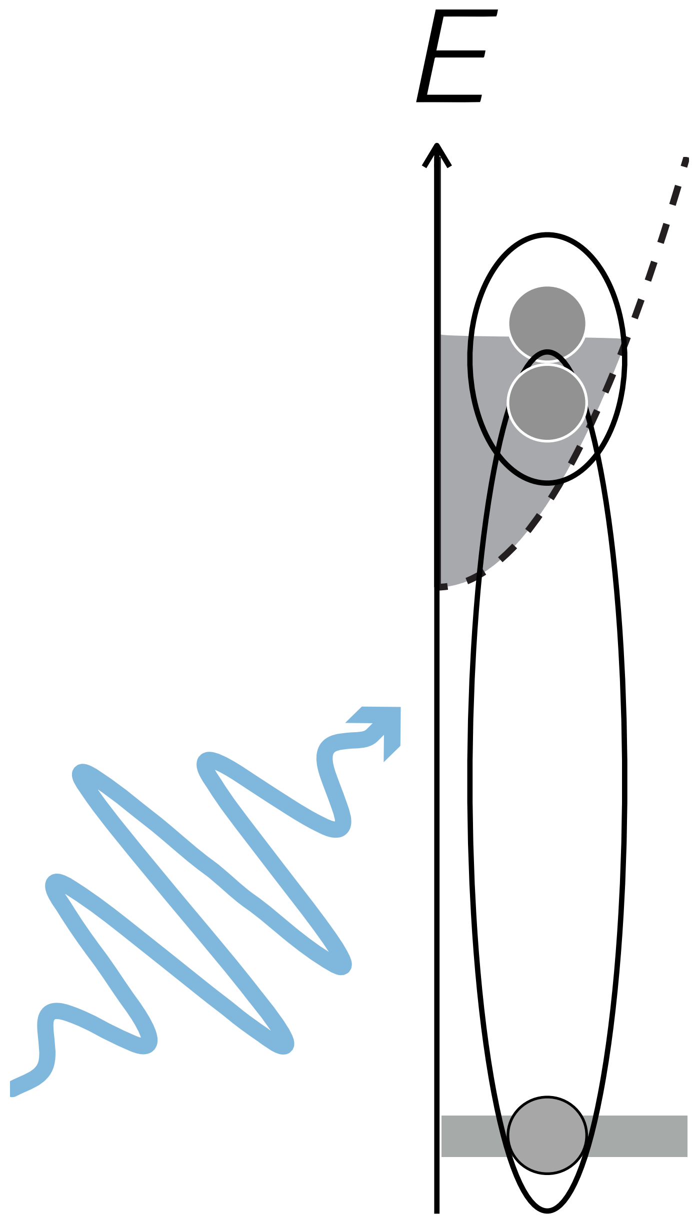
one-wave mixing does not exist… (only a pure coherence cannot be observed)
two-wave mixing is a $\chi^{(1)}$ process
three-wave mixing is a $\chi^{(2)}$ process
four-wave mixing is a $\chi^{(3)}$ process
…

Three-wave mixing

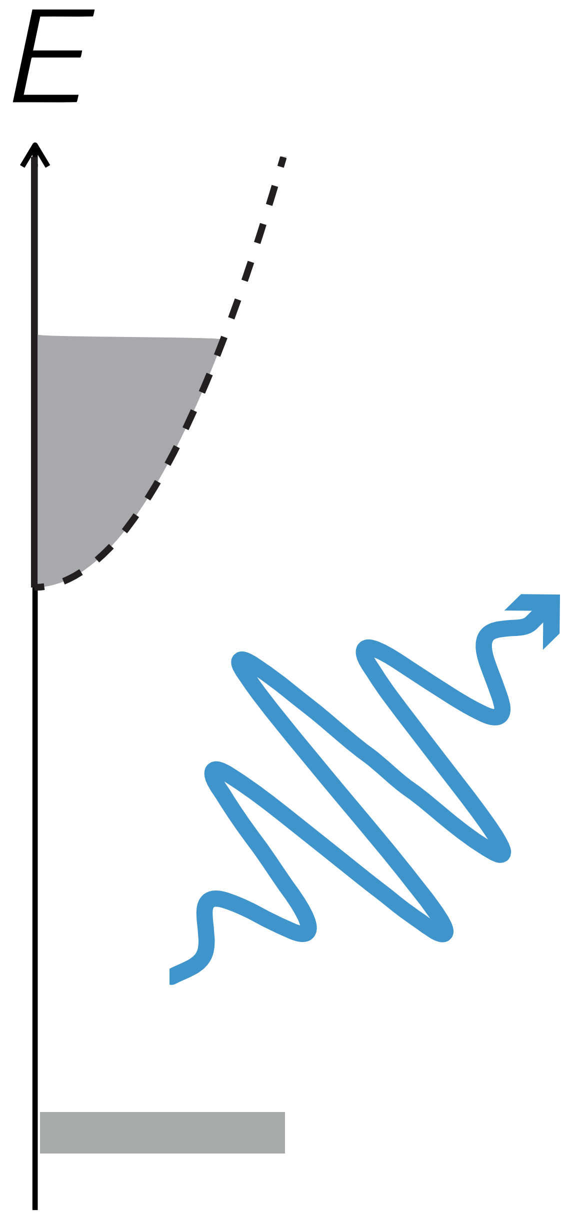
SFG (SHG, DFG)



1. Create coherence



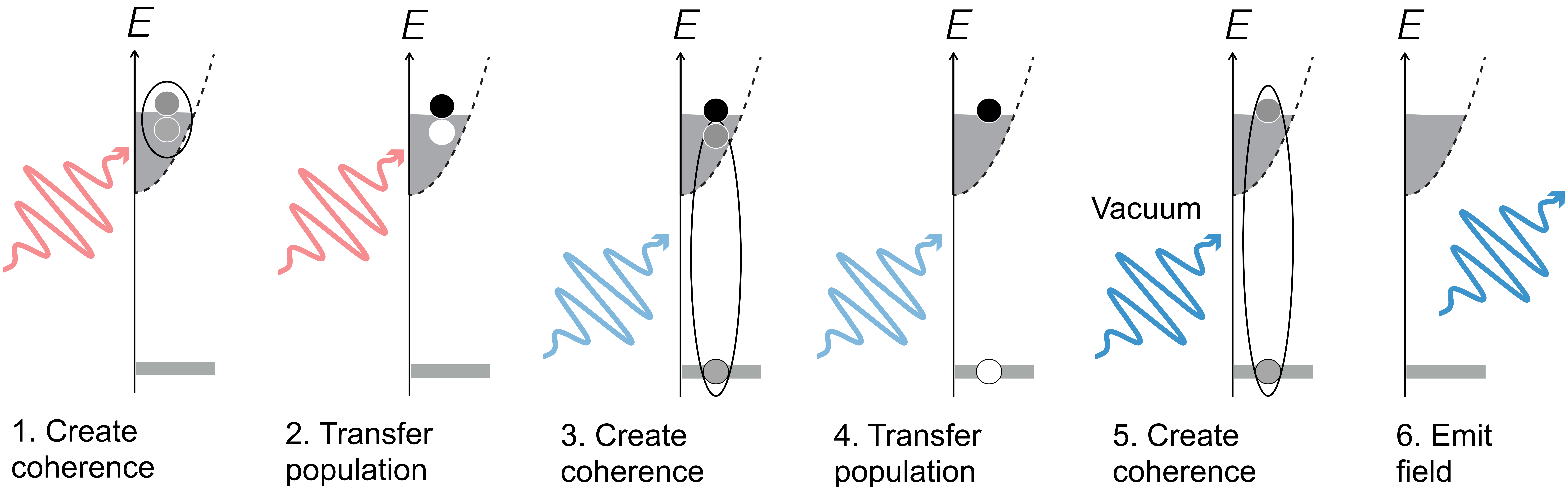
2. Transfer coherence



3. Emit field

Six-wave mixing

Pump-probe (anti-Stokes) RIXS



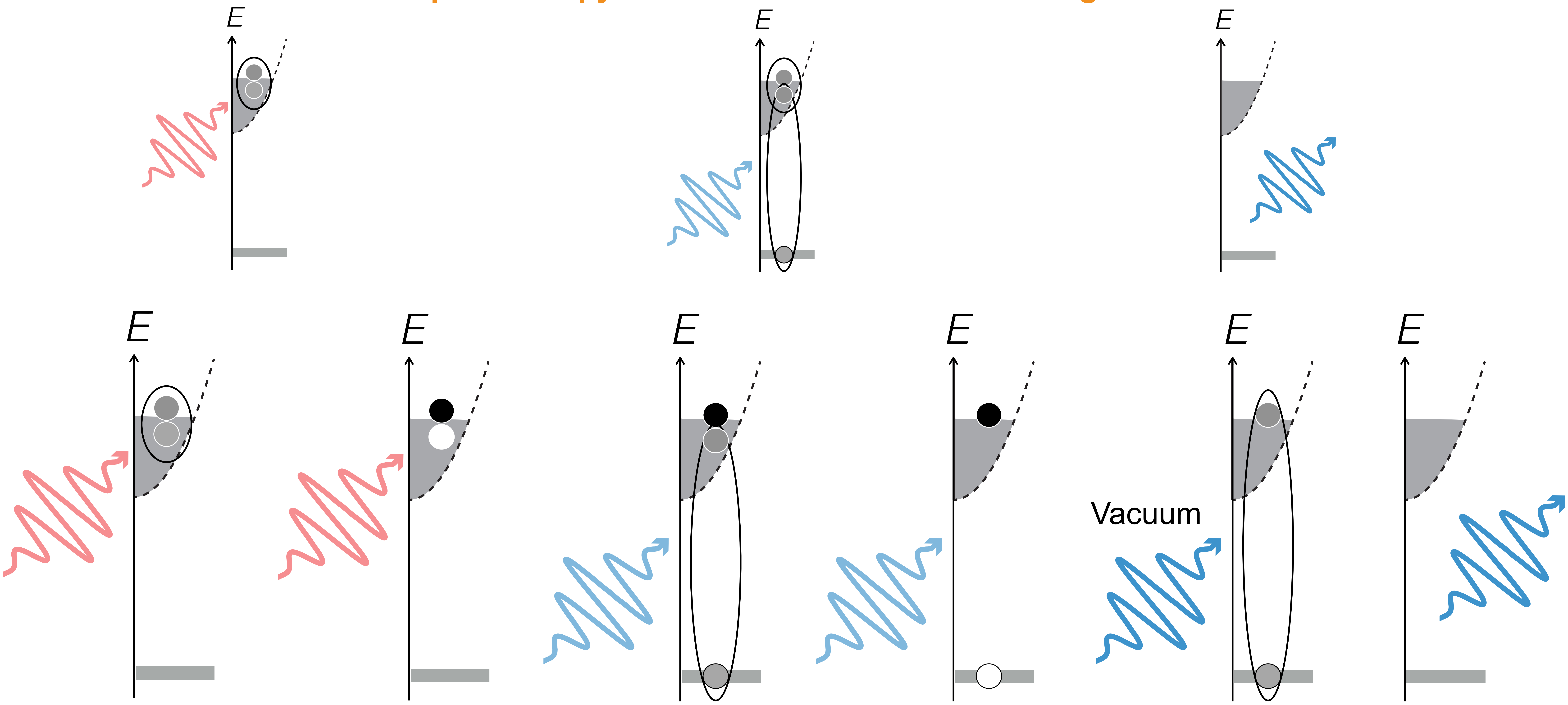
Experimental designs

to realise wave-mixing spectroscopies

Things to consider:
-phase matching condition (angles, geometry)
-concurrent processes and their relative strengths

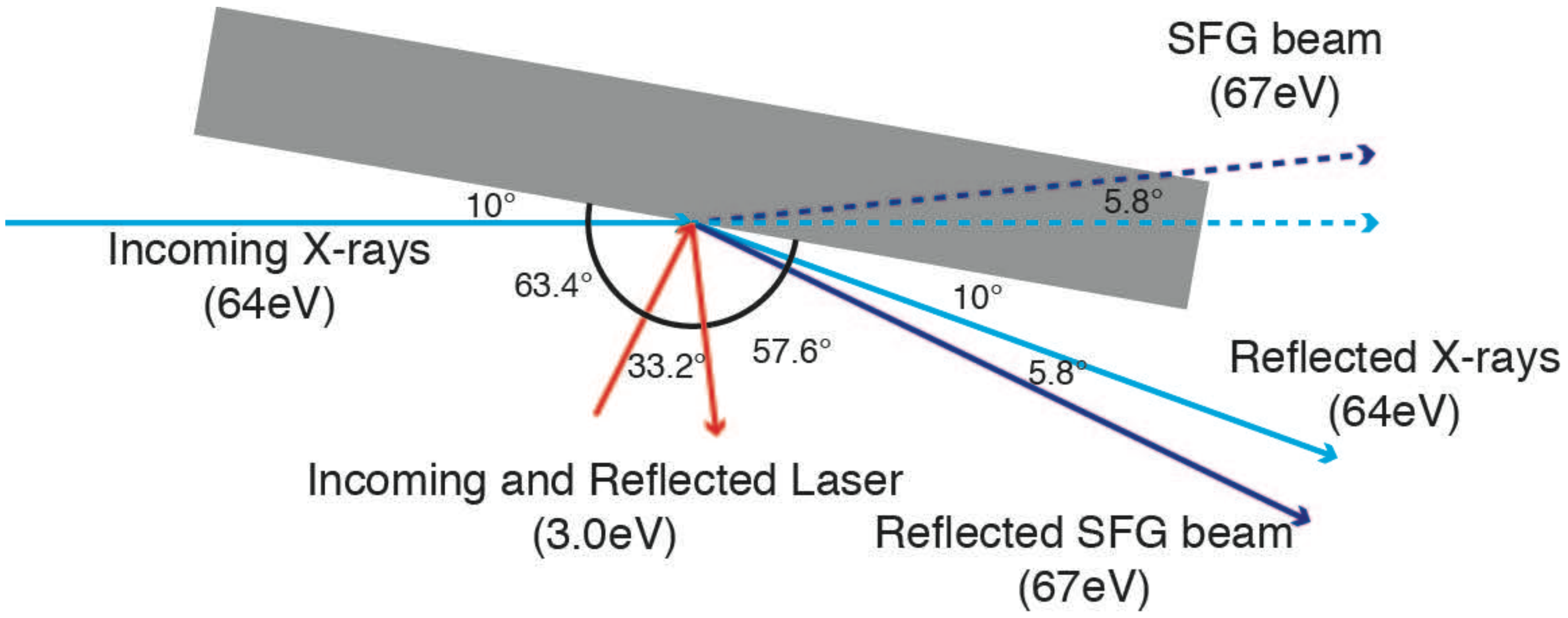
SFG vs. anti-Stokes RIXS

Coherent vs. incoherent spectroscopy / three-wave vs. six-wave mixing



Possible SFG geometry at an M-edge

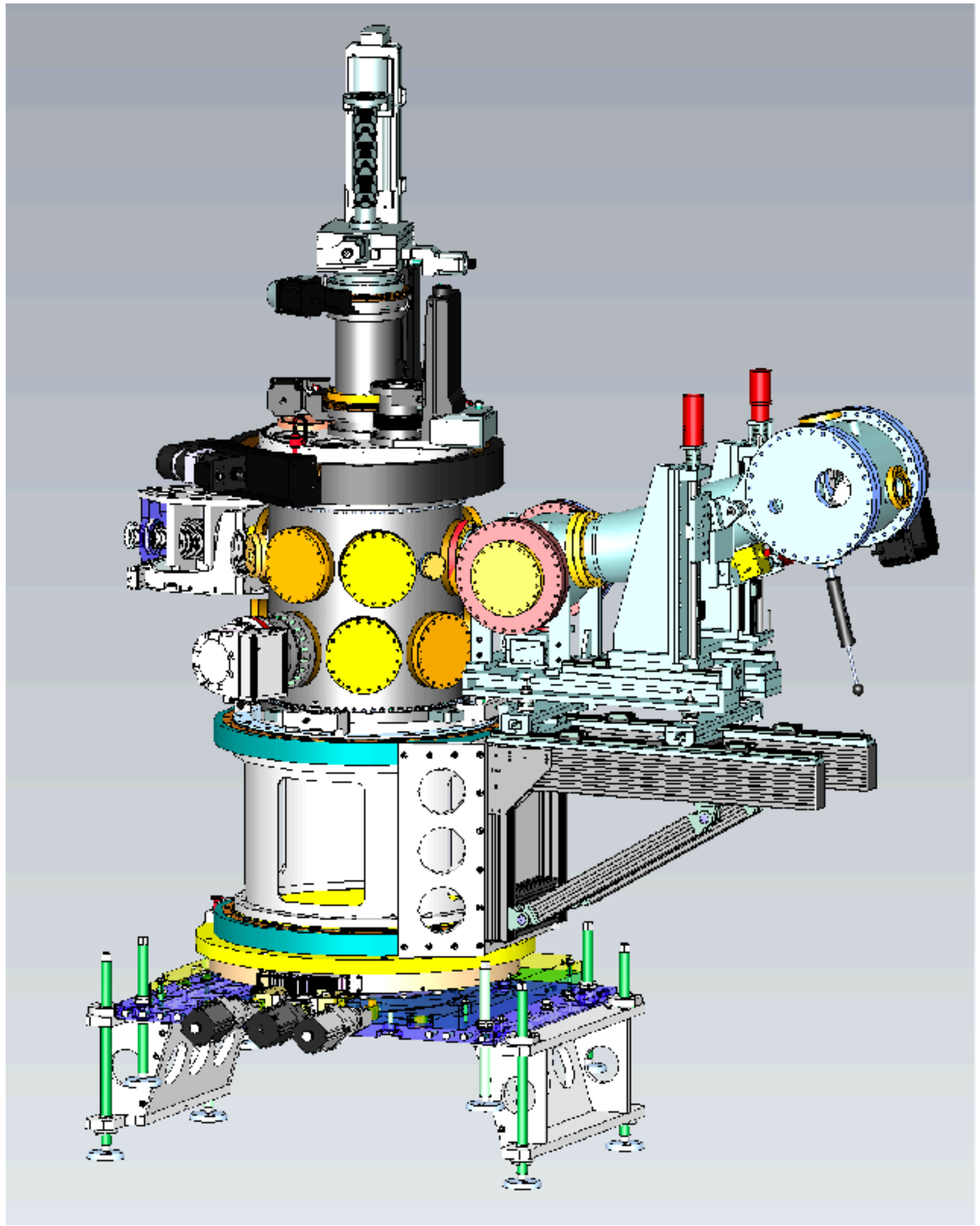
Anti-Stokes RIXS has no phase matching condition



Experiments

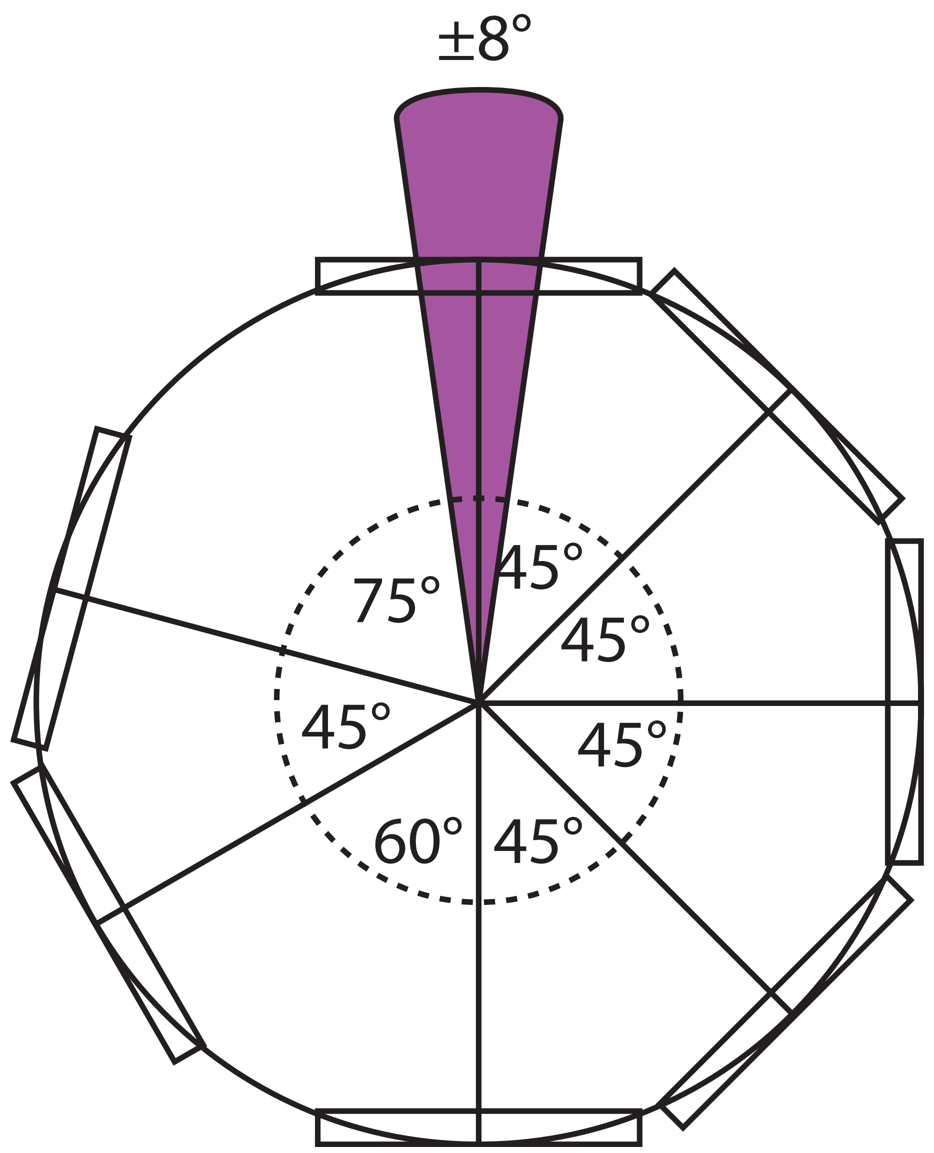
MUSIX

Multi-dimensional spectroscopy and (in-)elastic X-ray scattering instrument



- Designed at DESY
- UHV diffractometer modified from Uni Köln design, in use at BESSY
- spectrometer design in house
- in (full) operation within the next two weeks

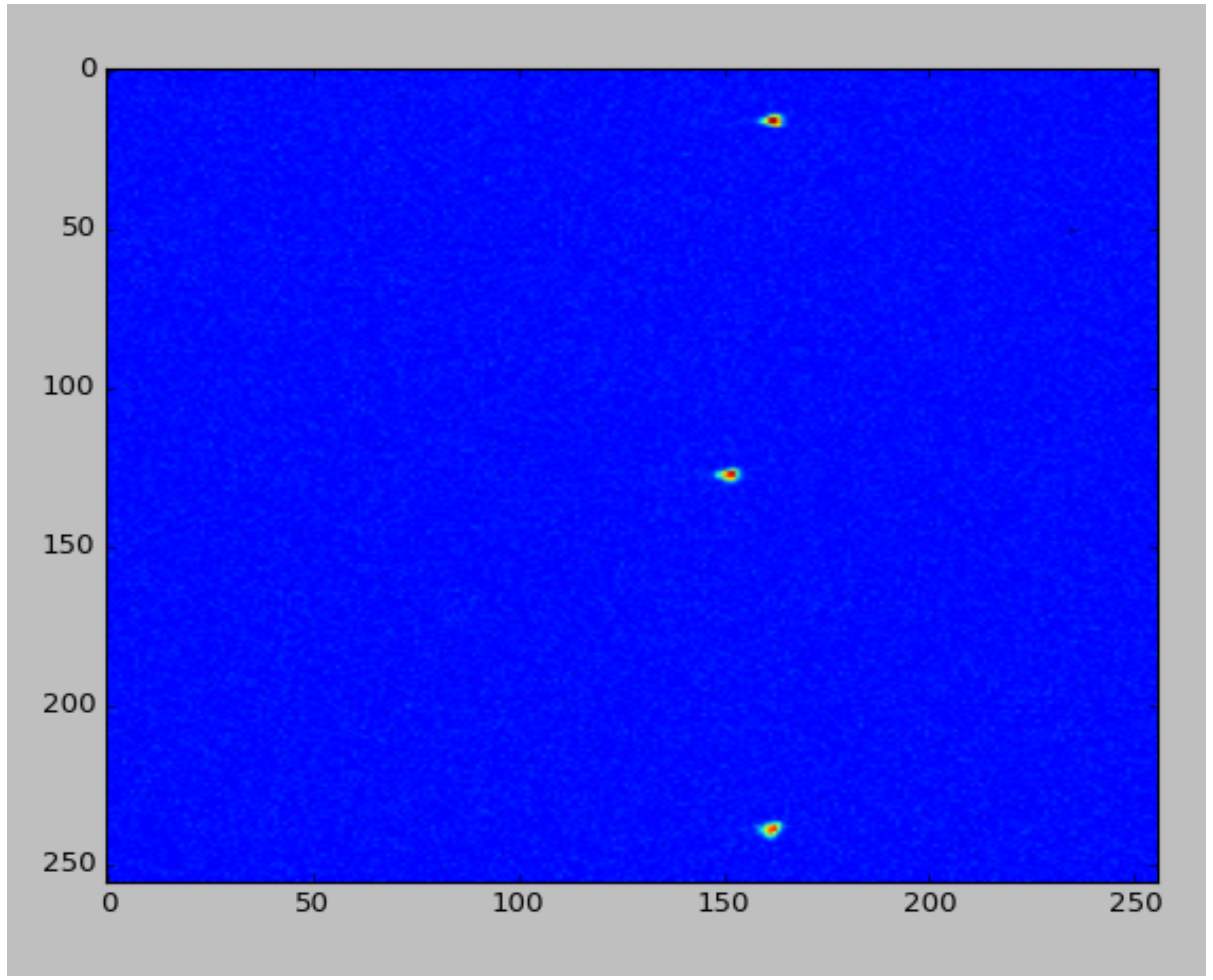
- flexible for various beam lines / light sources
- fulfill phase matching condition
- spectral analysis
- materials' science environment
- open for collaborations @ FLASH / PETRA (P04) or elsewhere

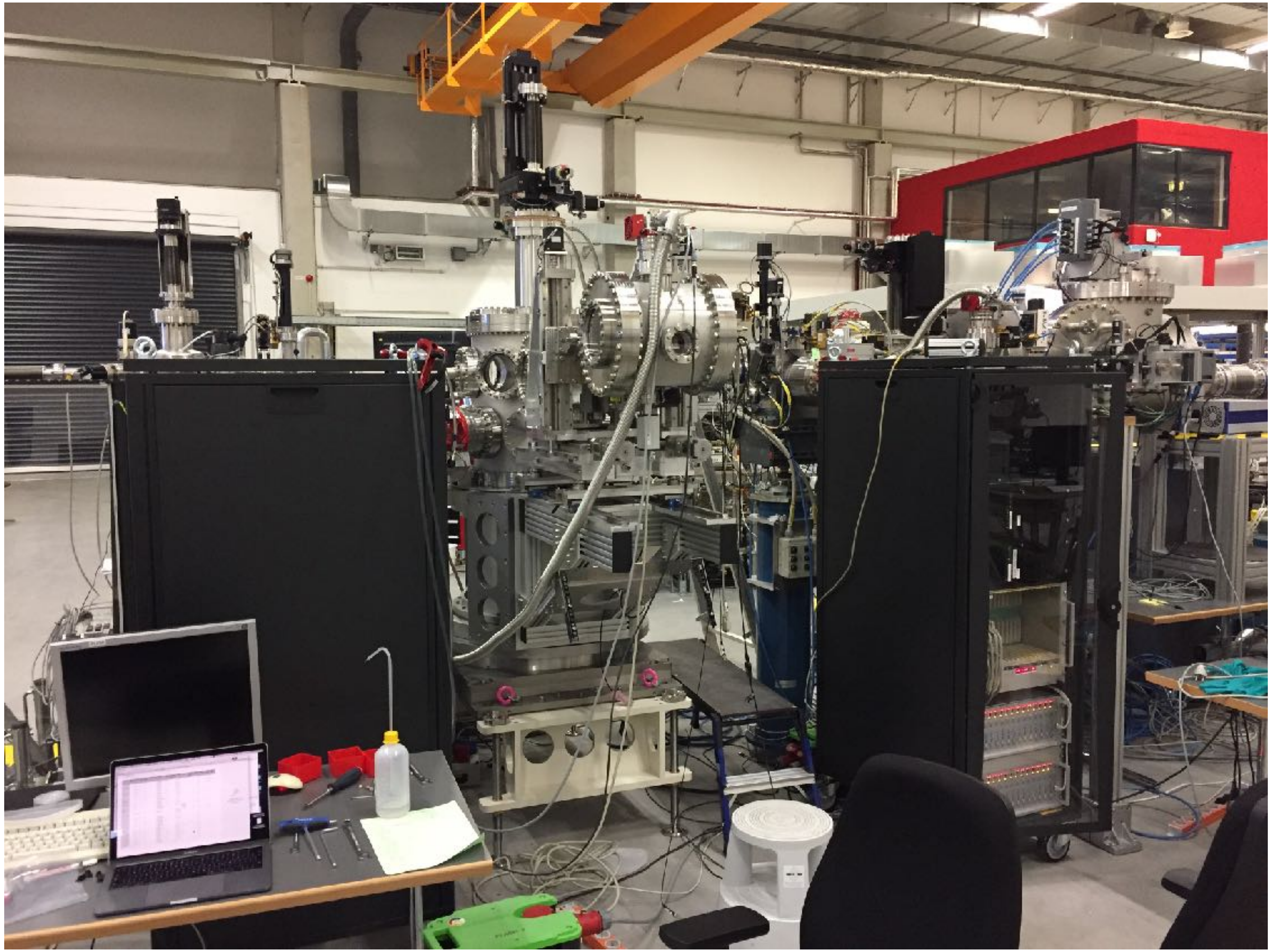


MUSIX in reality

Completed in next two weeks, ideas for future collaborations welcome

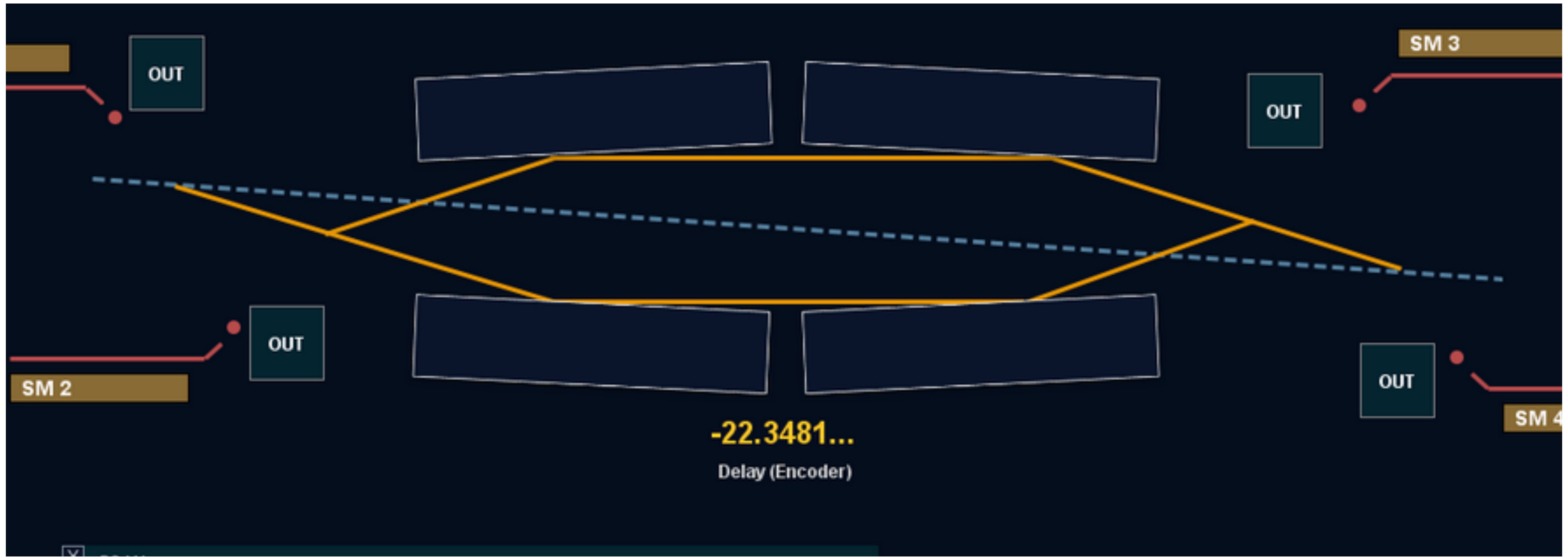
-Nitrogen K-edge (400eV) XES
-Triple RZP analyser
-Resonant excitation with third harmonic @ FLASH2



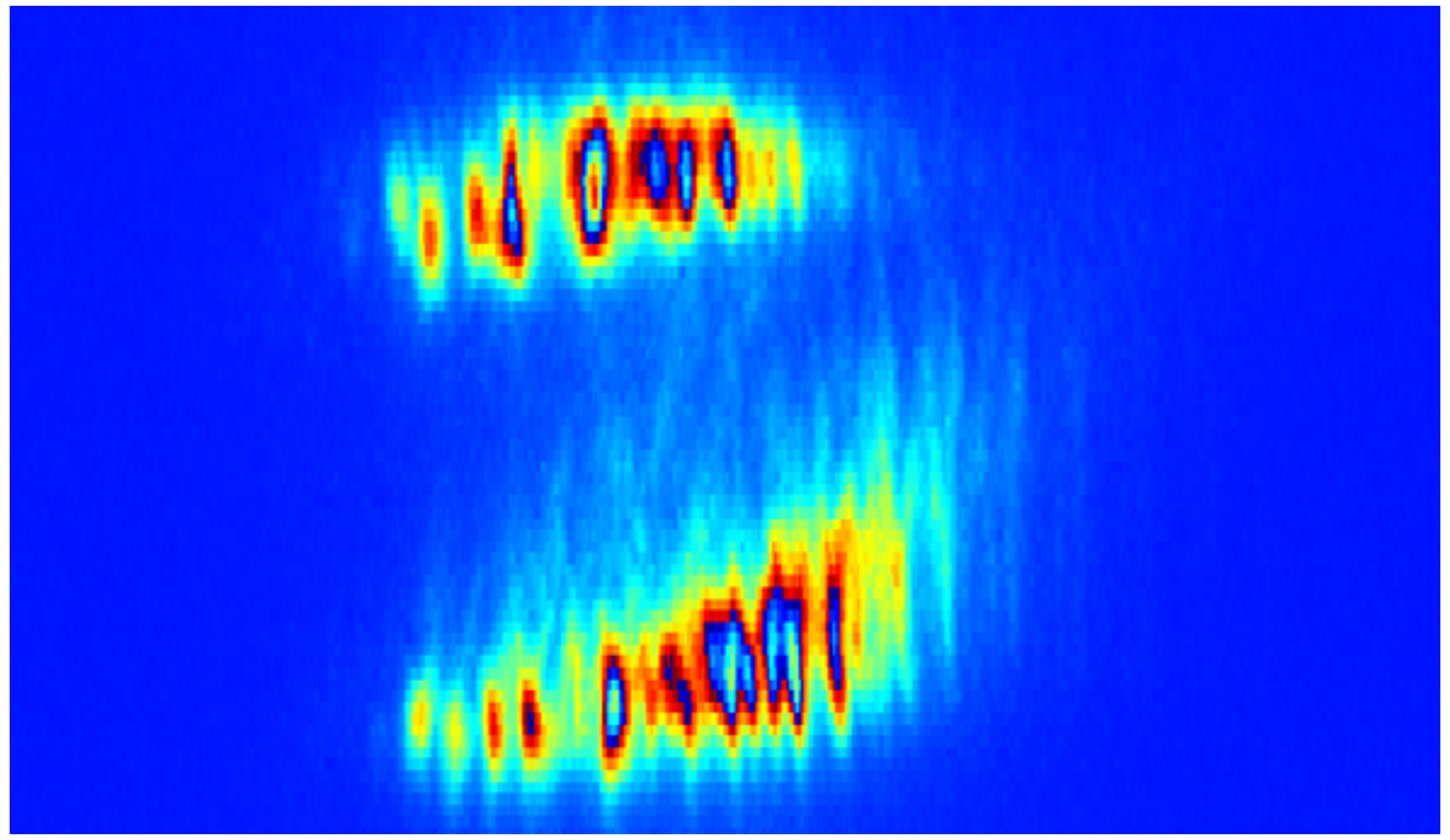


Heterodyne detection: Coherent reference beams

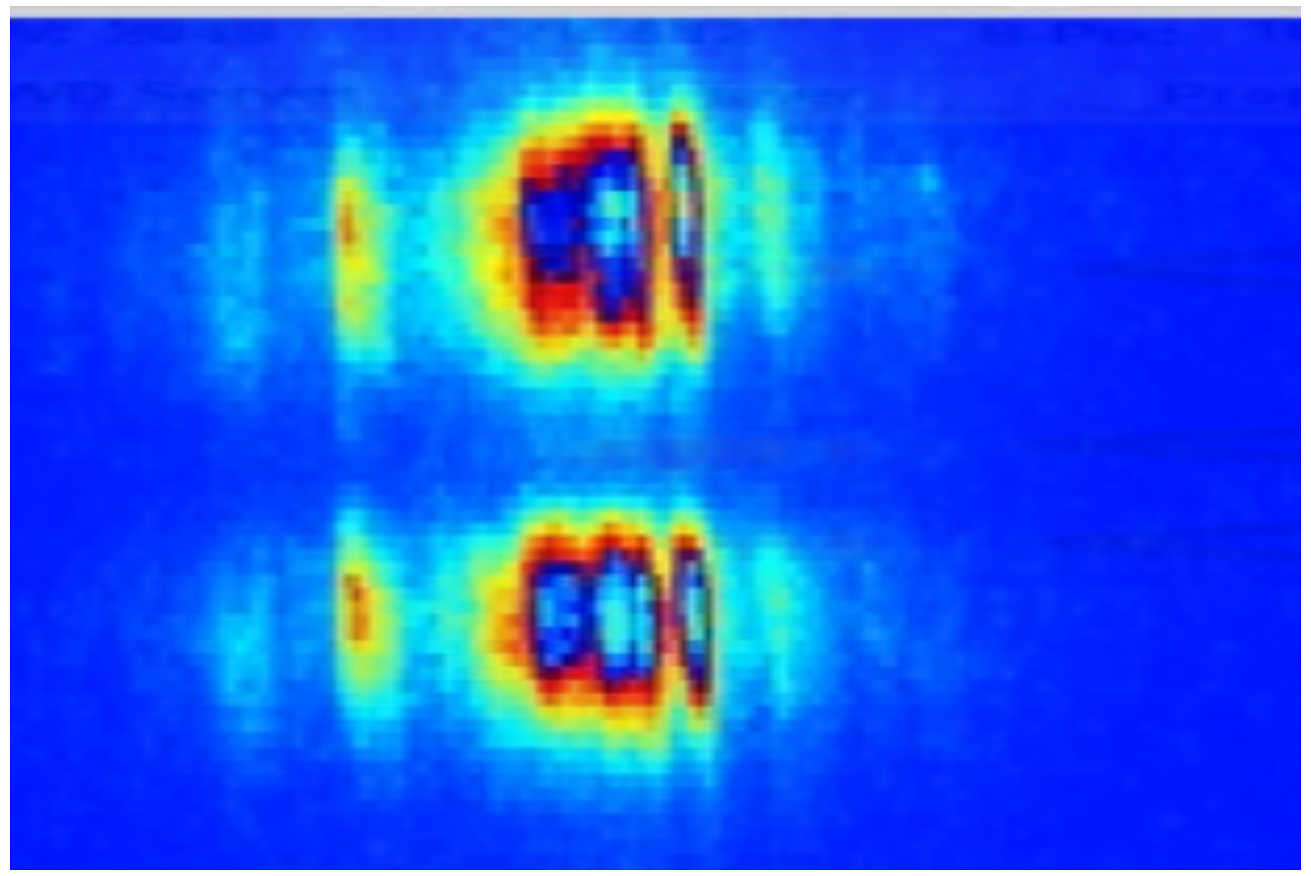
Optimised split and delay units



Standard scheme:
Split with edge of mirror



New scheme:
Split with transmission grating

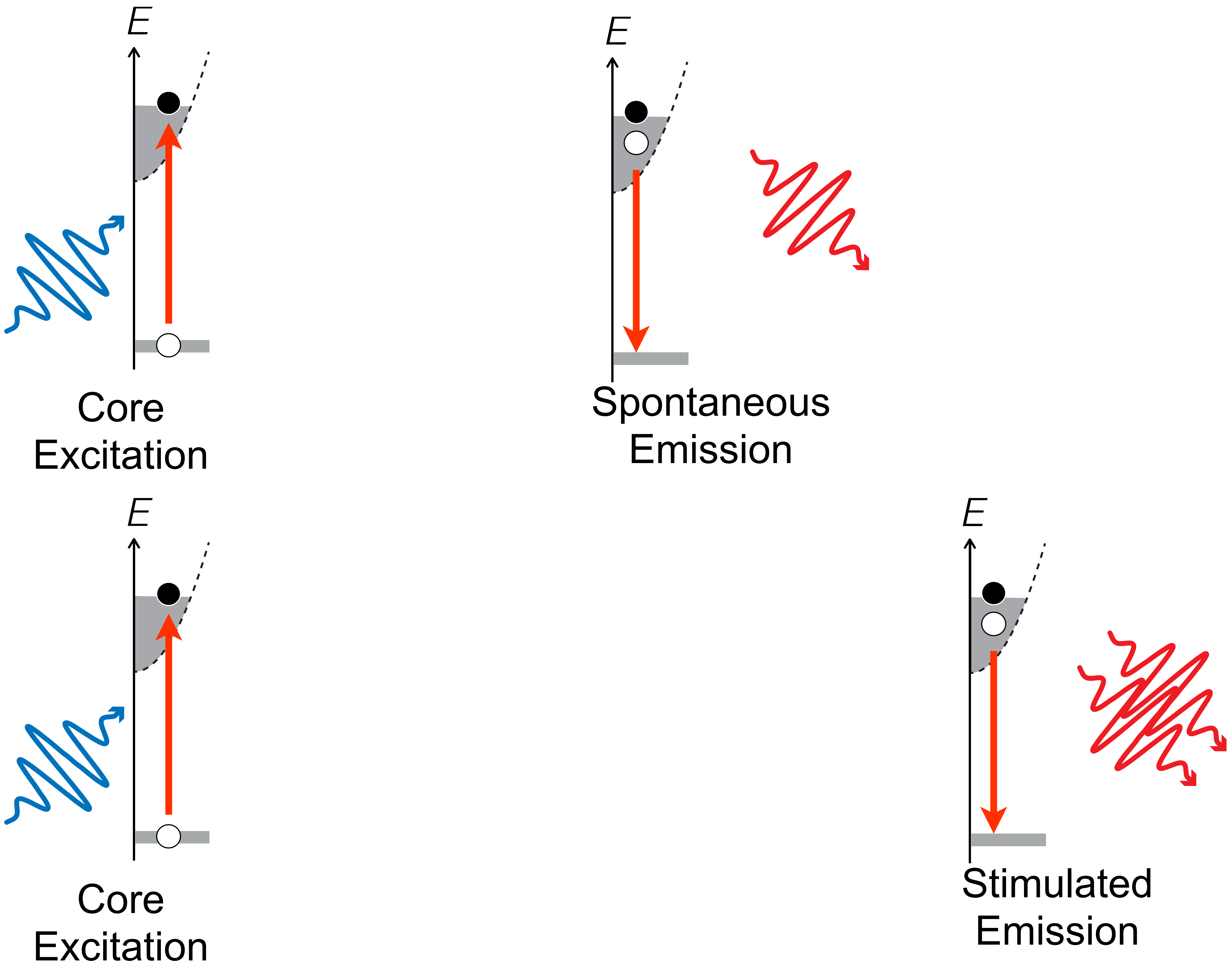


Collaboration with G. Brenner (FLASH) et al. + C. David (PSI) et al.

Amplified spontaneous emission @ FLASH

Silicon at high intensities

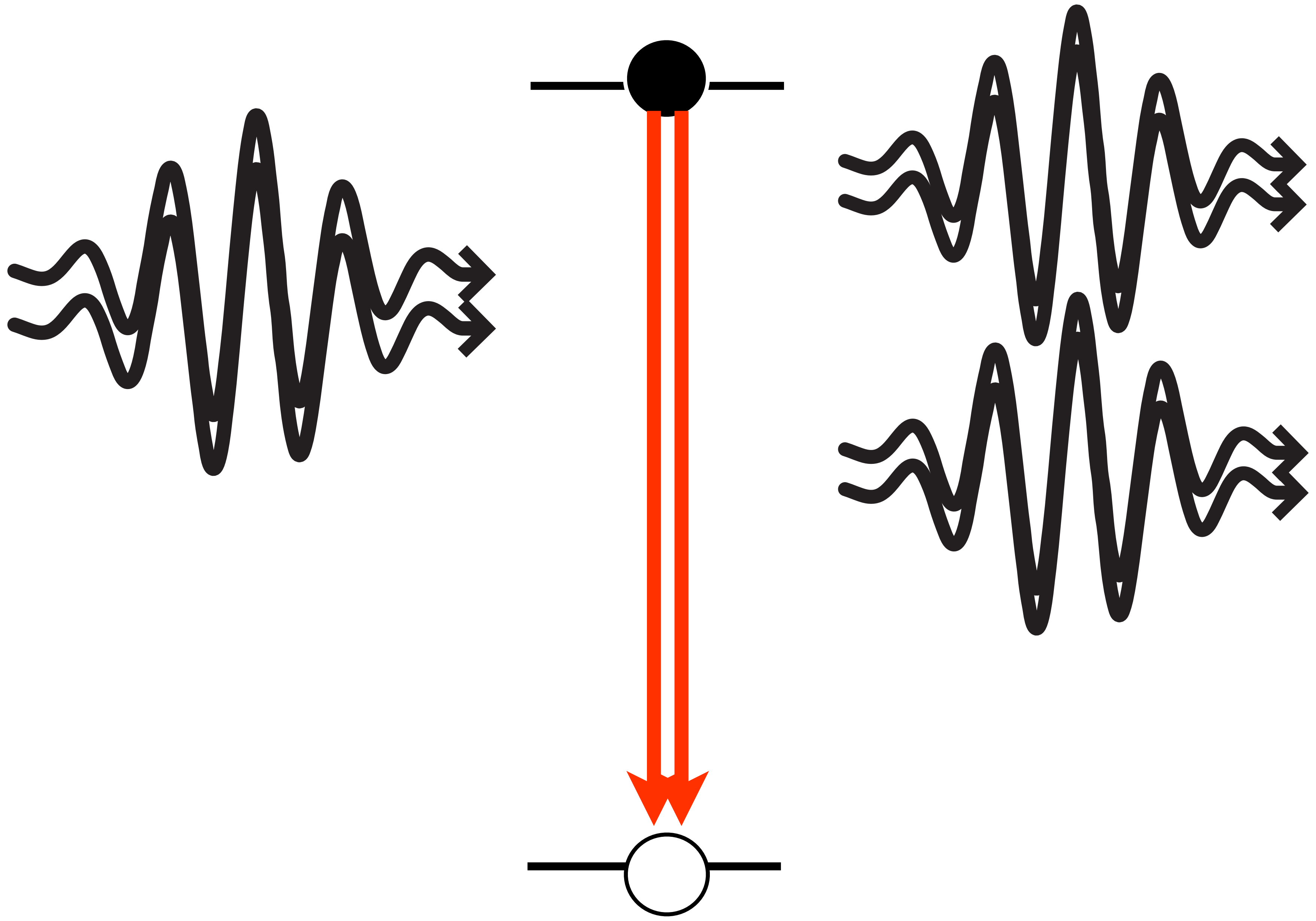
Martin Beye
Simon Schreck
Florian Sorgenfrei
Christoph Trabant
Niko Pontius
Christian Schüssler-Langeheine
Wilfried Wurth
Alexander Föhlisch



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

Geometry optimisation

Maximise interaction length with core-excited volume



Stimulated Emission

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

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

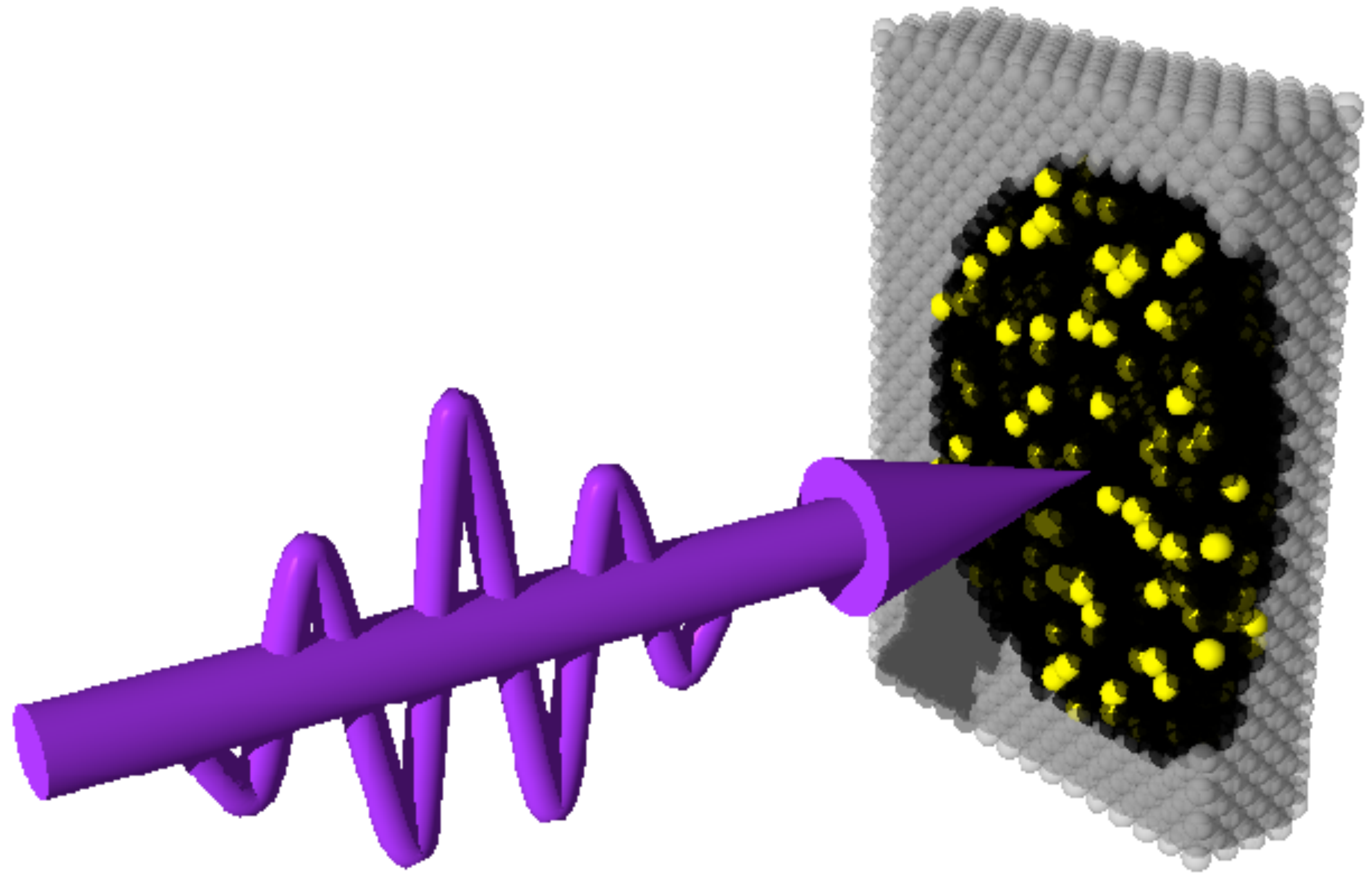
$$\sigma_{abs} = \frac{1}{\lambda_{abs}\rho_{atom}}$$

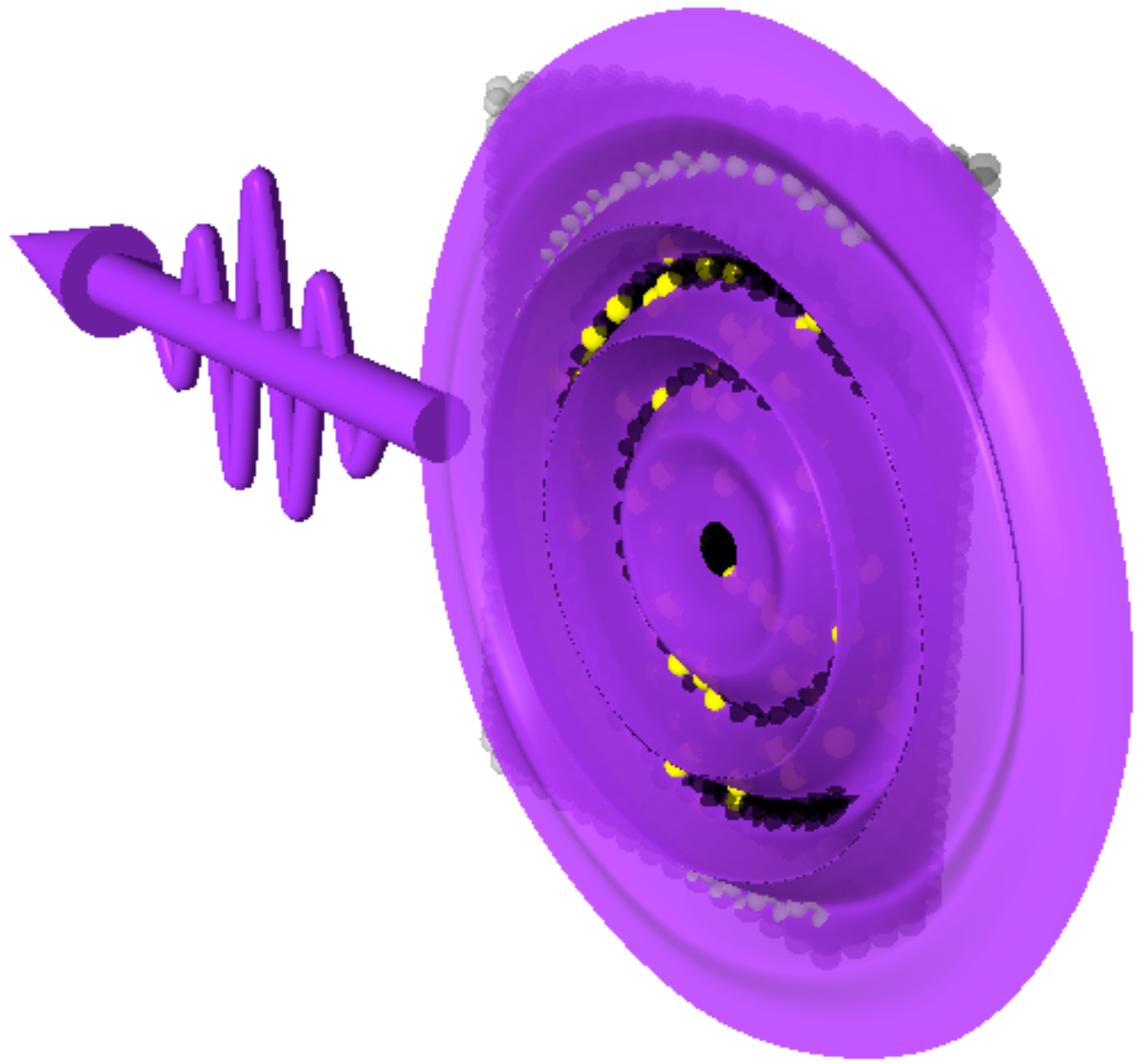
$$\frac{\rho_{ch}d}{\lambda_{abs}\rho_{atom}} = 1$$

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

Stimulation front

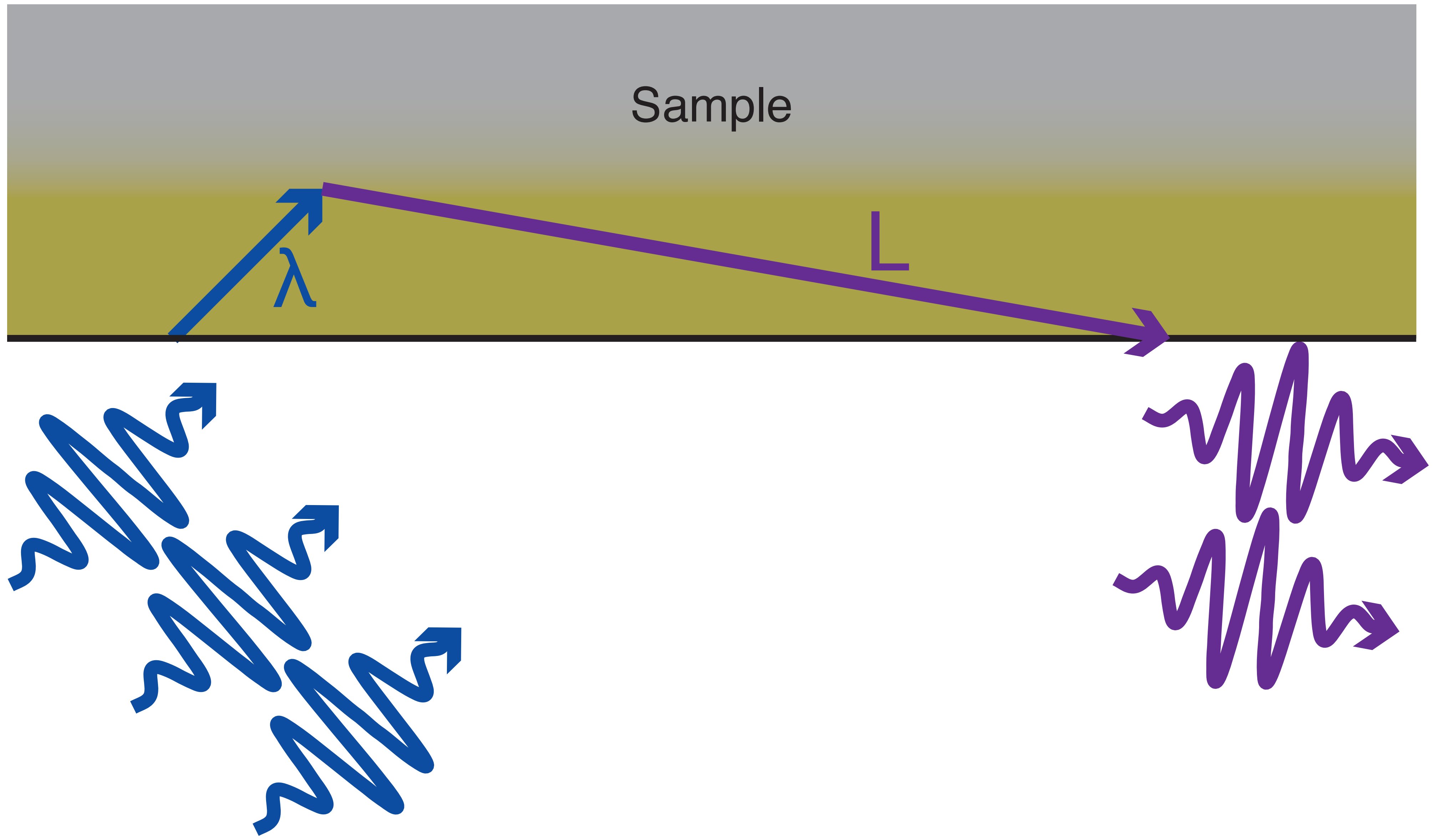
Primarily along the surface for soft X-rays on solids

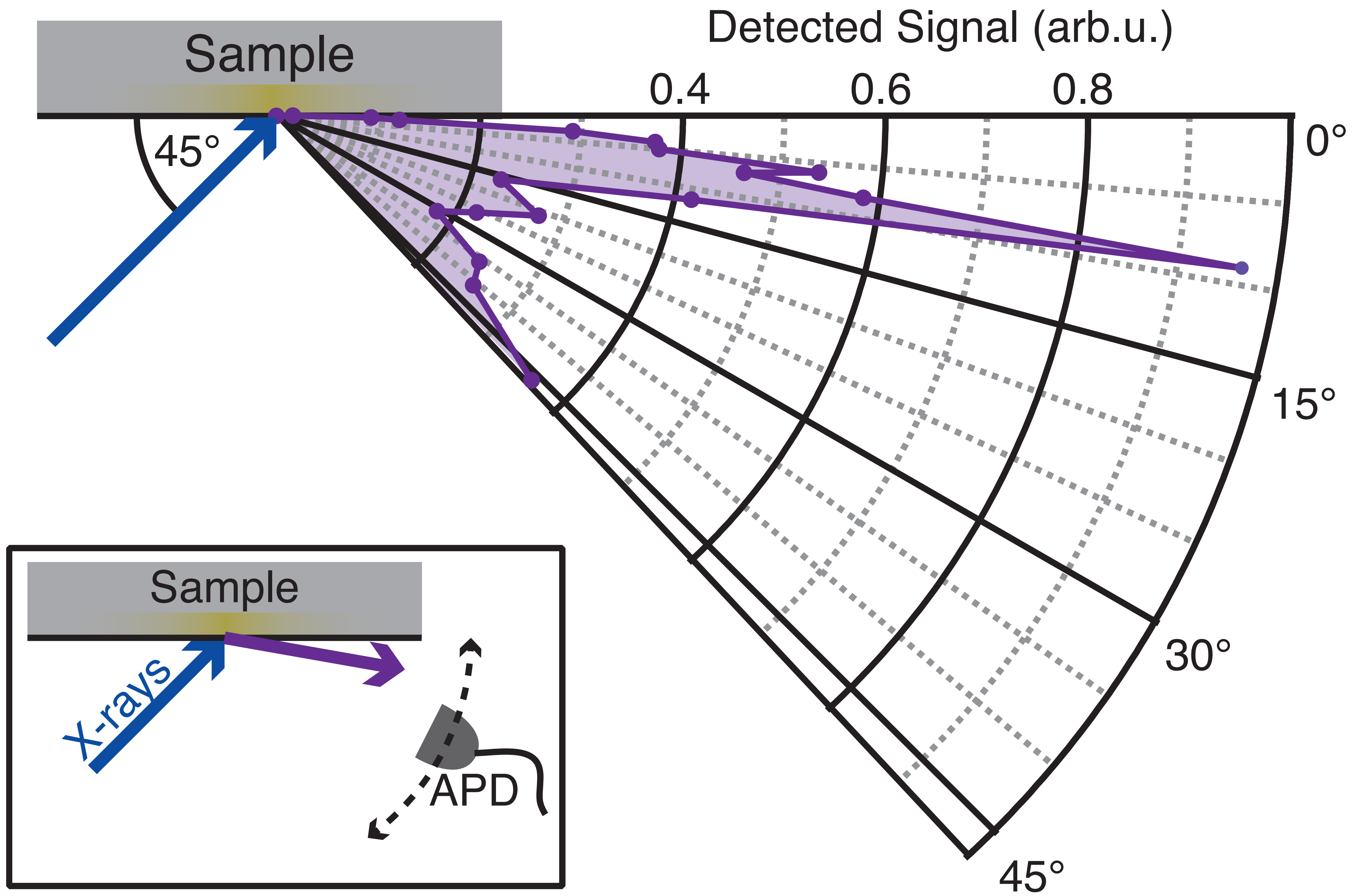




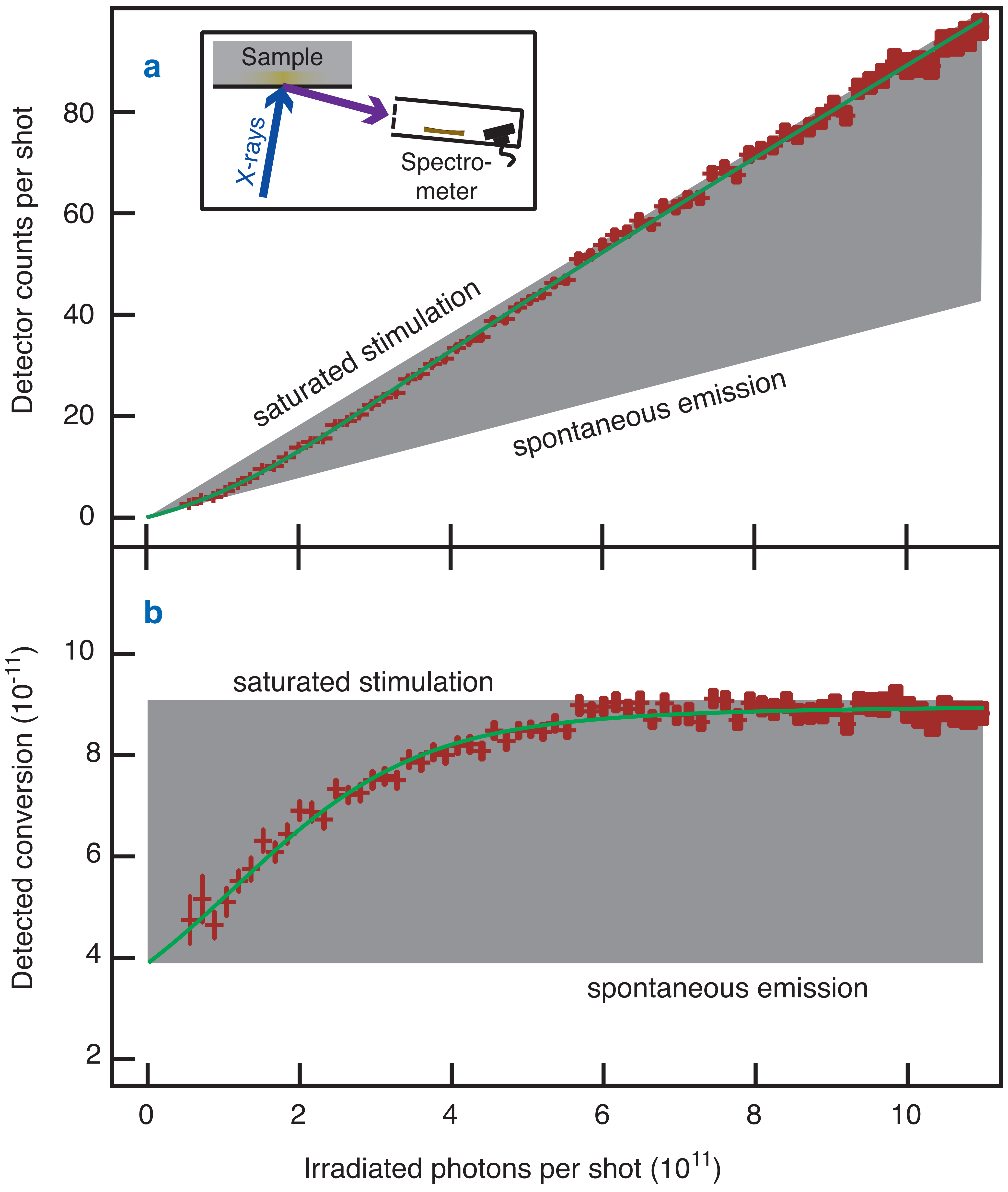
Angular dependence

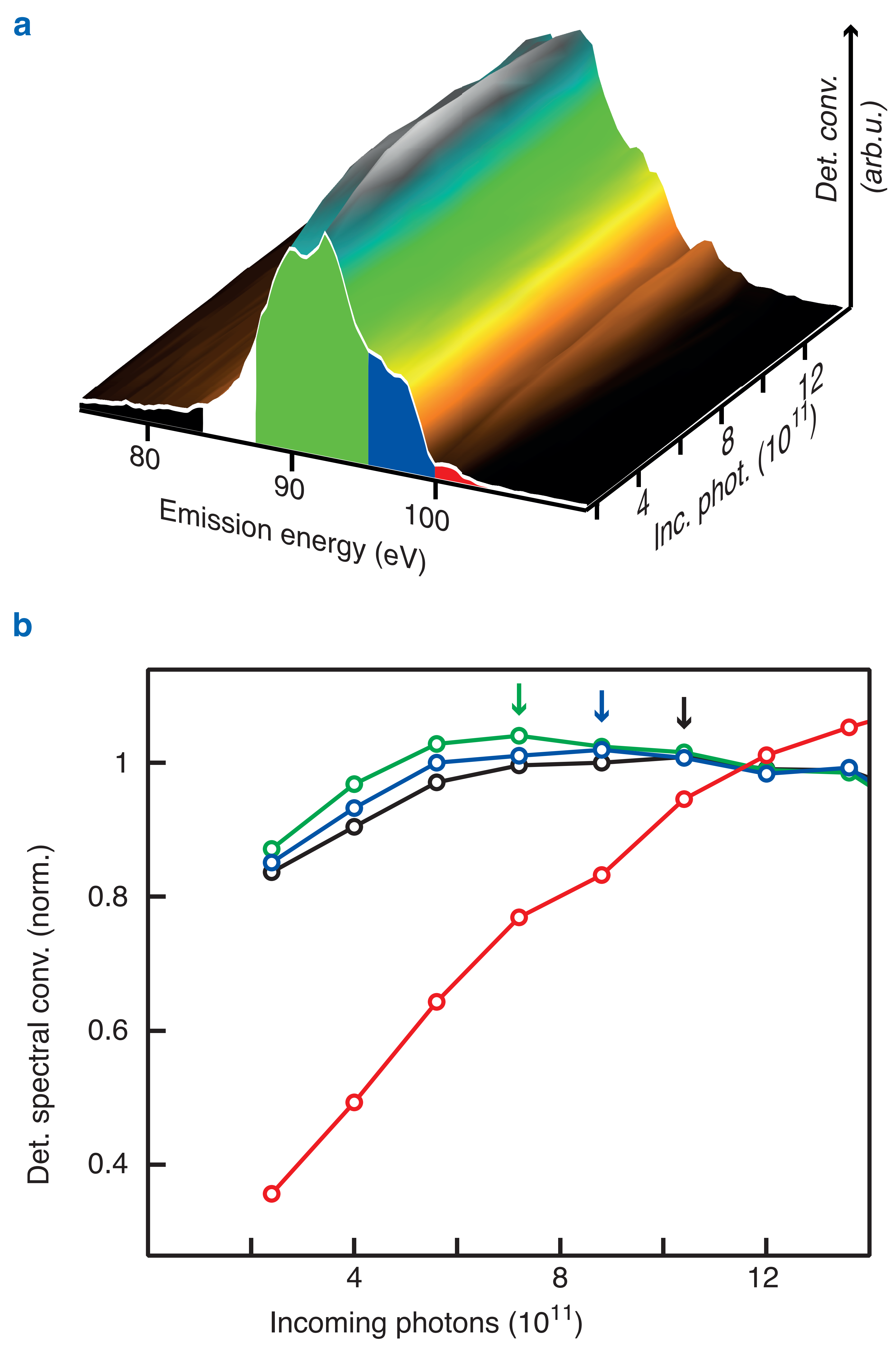
Si (100), 115eV, 1J/cm², 30 fs





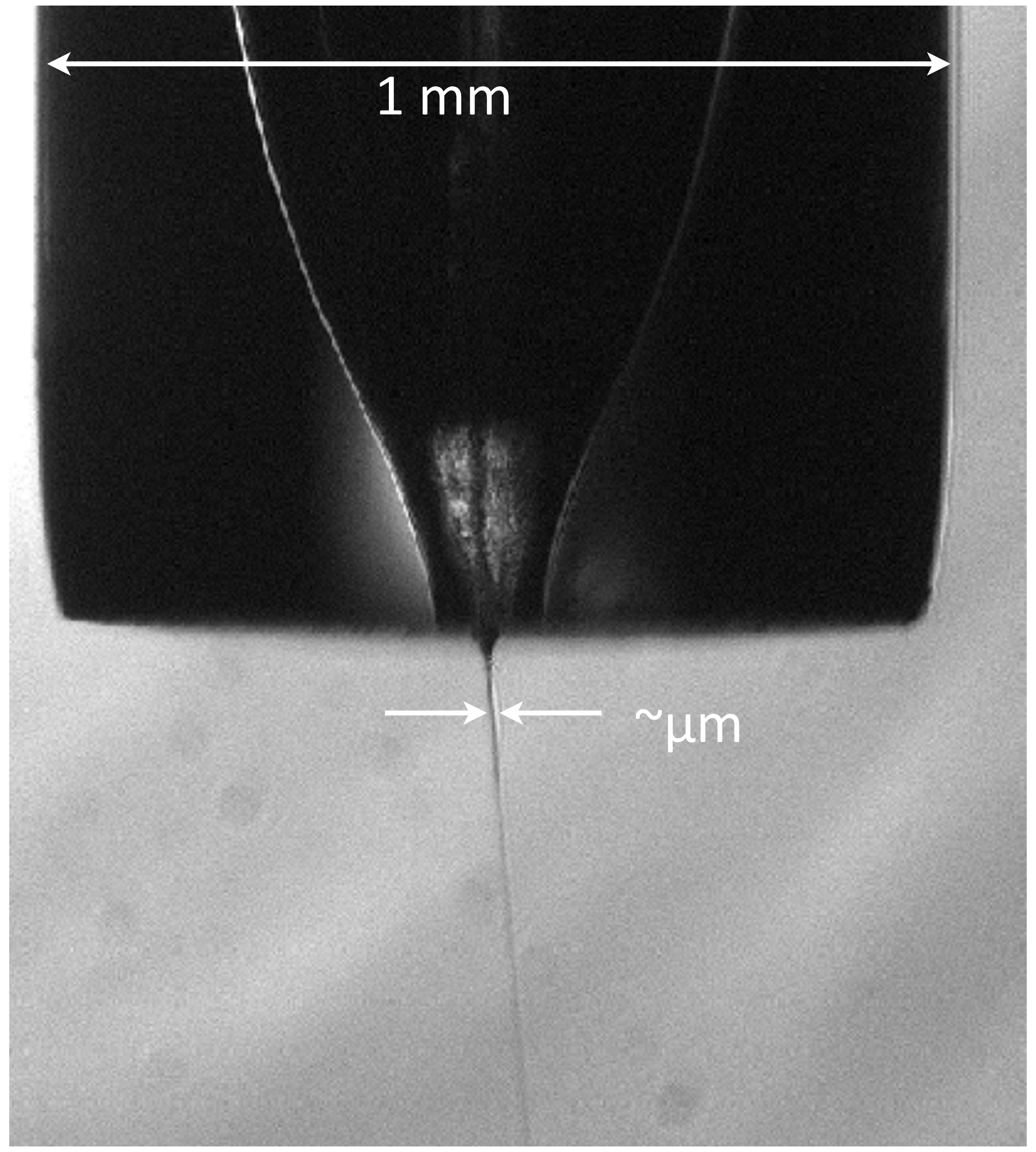
Fluence and spectral dependence





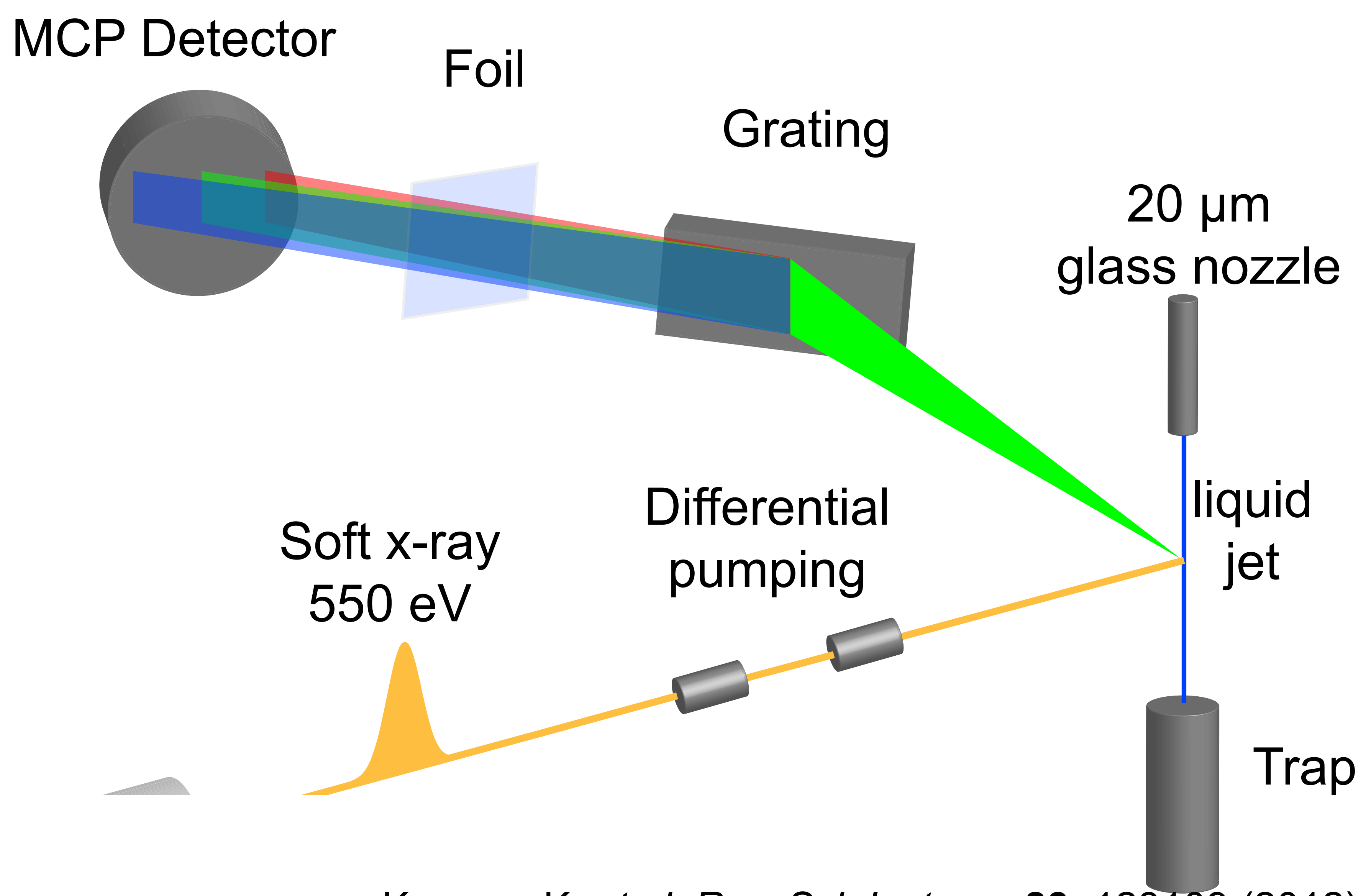
Amplified spontaneous emission from water?

Reabsorption of emission for long pulses and short core hole lifetimes



Liquid jet speed ~ 20 m/s
~ **kHz** to **MHz** rep. rate

The LJE/FlexRIXS endstation



Kunnus, K. *et al. Rev. Sci. Instrum.* **83,** 123109 (2012)

PRL **113,** 153002 (2014) PHYSICAL REVIEW LETTERS week ending 10 OCTOBER 2014

Reabsorption of Soft X-Ray Emission at High X-Ray Free-Electron Laser Fluences

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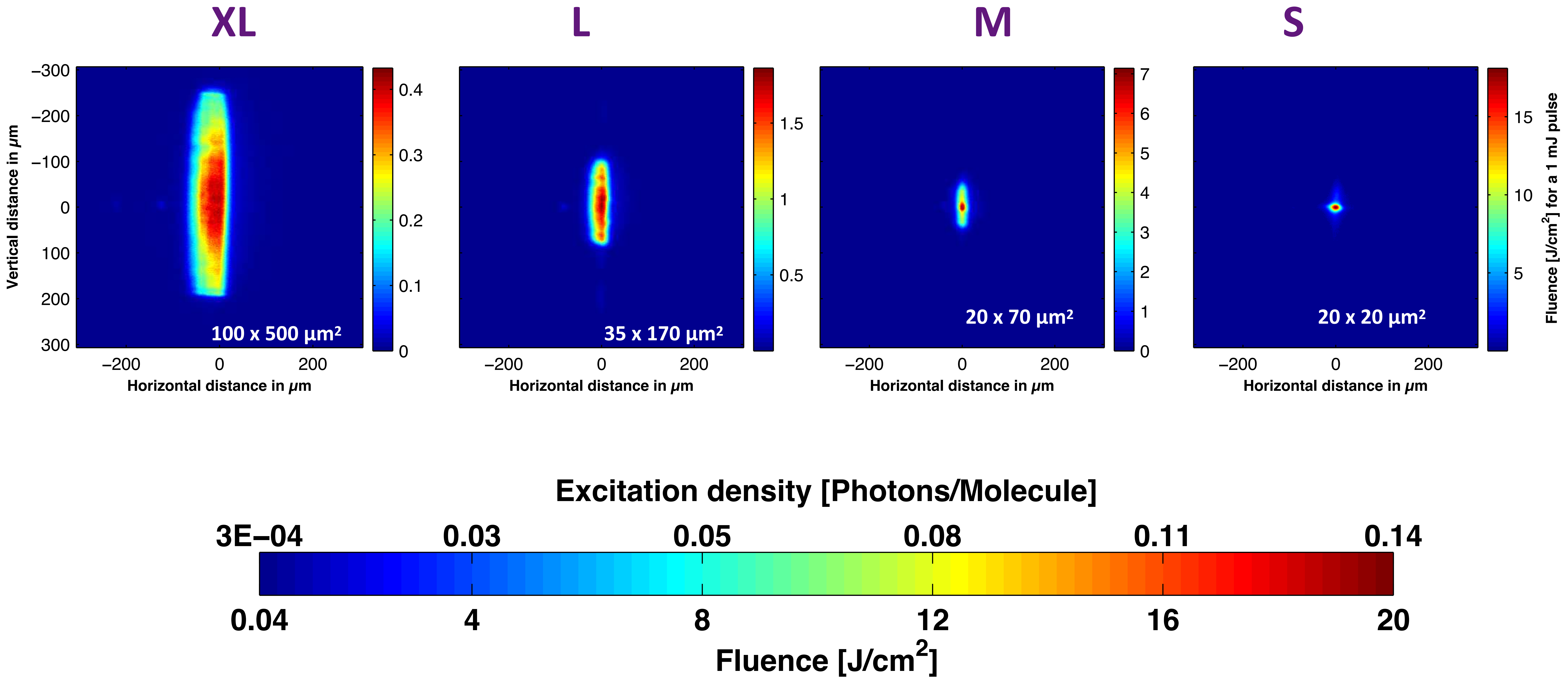
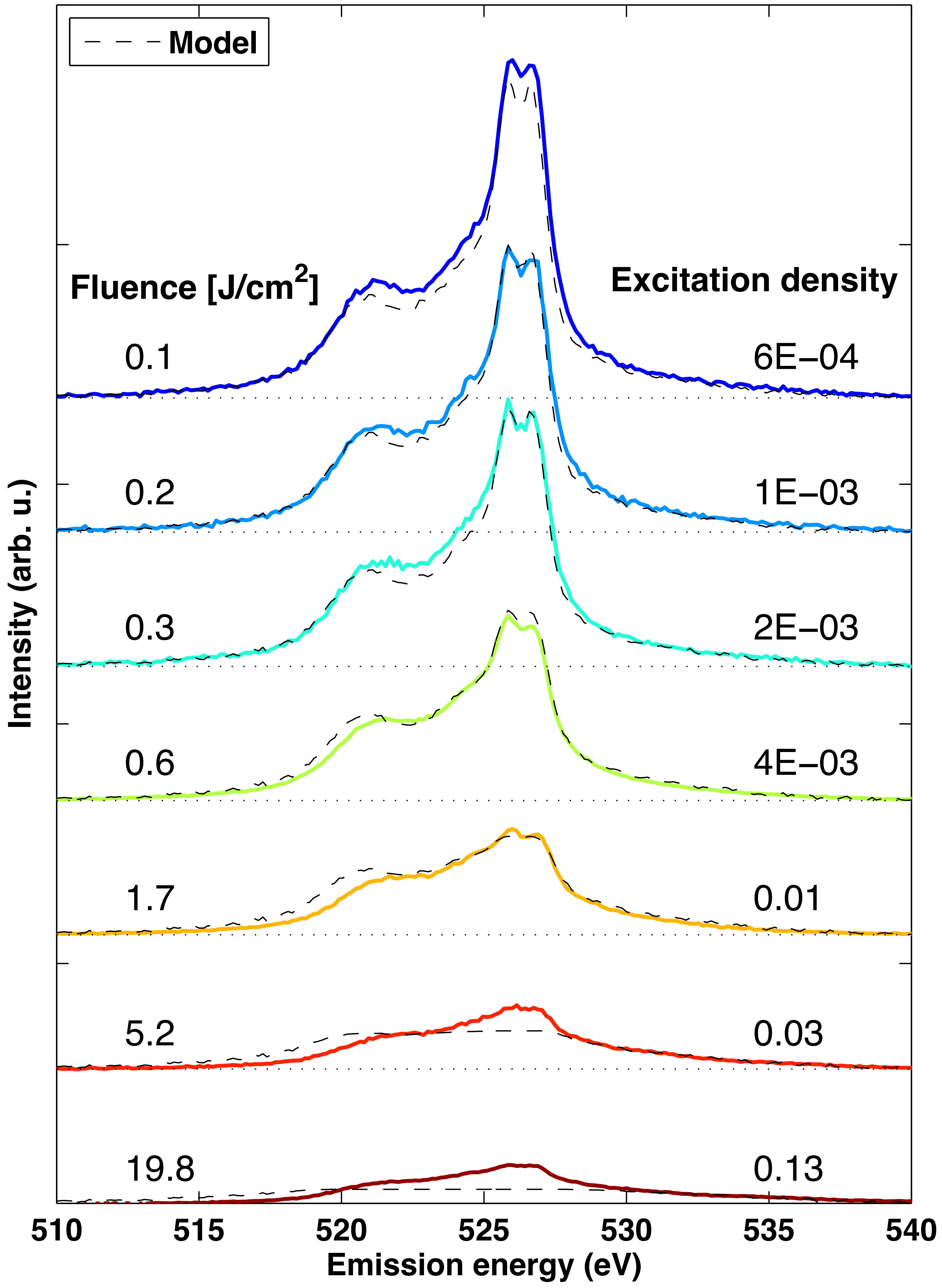
Continuous fluence variation by orders of magnitude

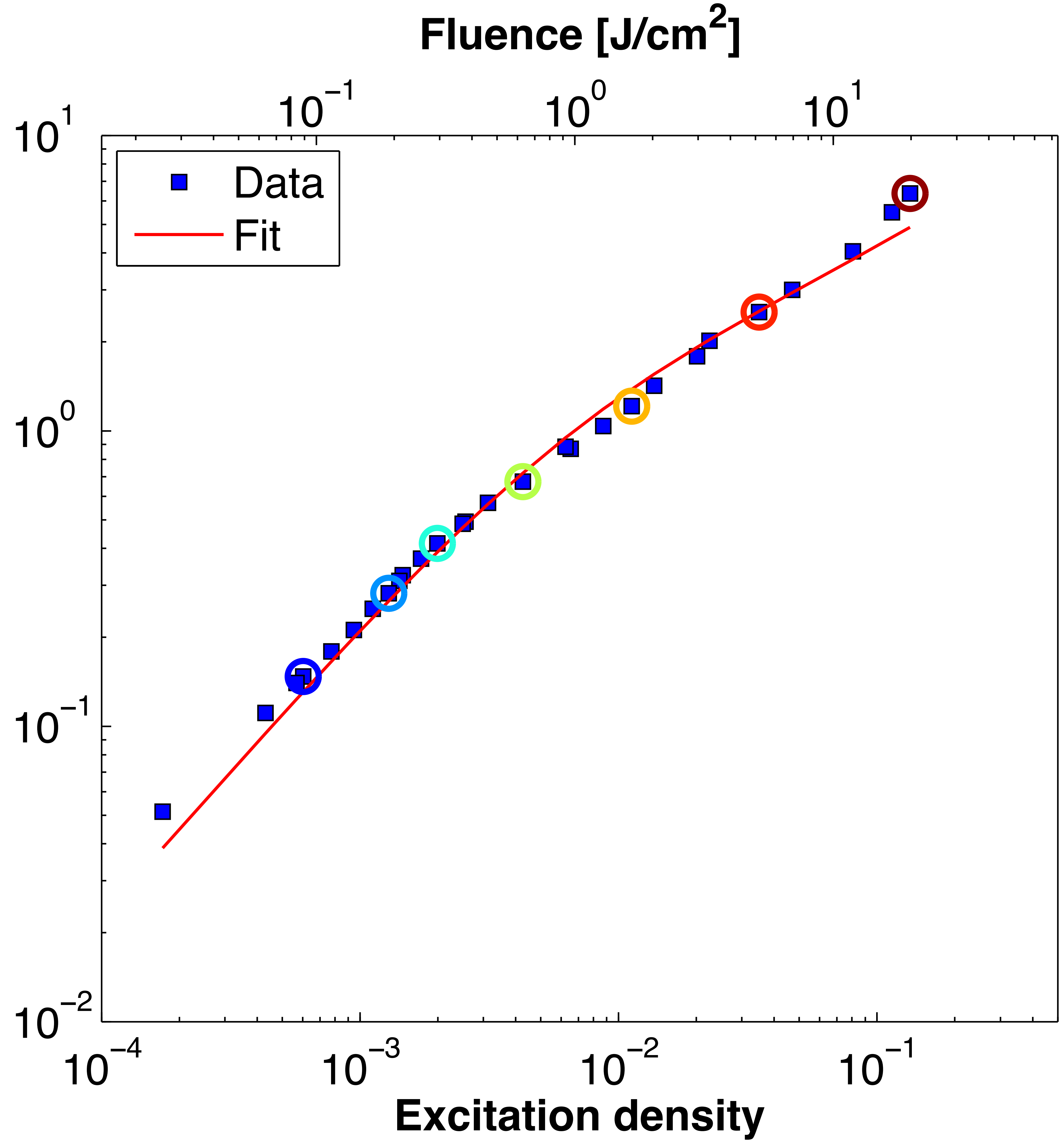
LCLS SXR has bendable KB-mirrors



Saturation in spectra and count rate

Simple model taking Auger cascade into account, models dependences quantitatively





Summary

From theoretical field interactions to real experiments

- Some non-linear processes have been observed
- Mostly population spectroscopy
- Electronic coherences in the X-ray regime still await discovery
- Potential to circumvent Auger cascade, if no population transfer
- Coherence lifetime undetermined, potentially very short
- WE NEED SHORTER PULSES!!!!

THANK YOU FOR YOUR ATTENTION.