Nuclear quantum optics at XFELs

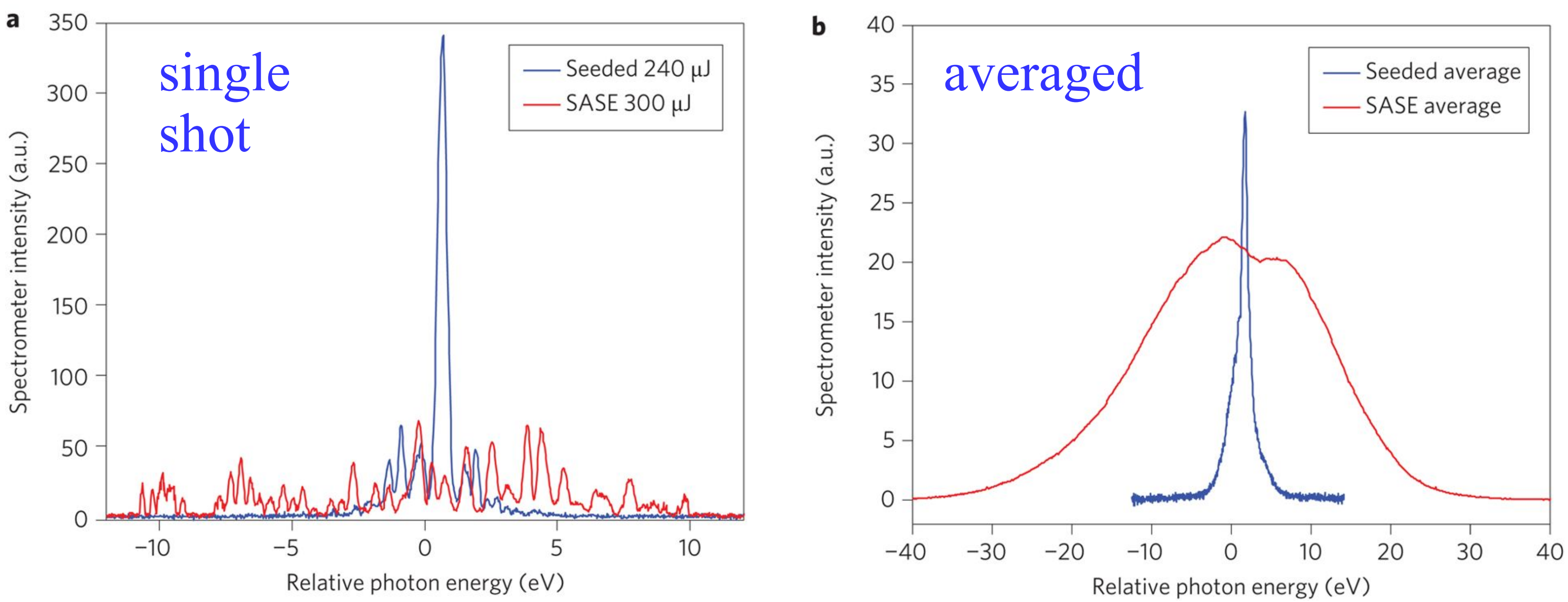
Jörg Evers

Max Planck Institute for Nuclear Physics
Heidelberg, Germany

New Scientific Capabilities at European XFEL (2019)

Longitudinal coherence

Typical association:



Amann et al., Nat. Phot. 6, 693 (2012)

Relevant technical concepts:
Seeding and x-ray oscillator (XFELO)

Longitudinal coherence in more detail

"Obvious" implications:

- high spectral photon density
  → qualitatively new parameter regimes

- pulse-to-pulse stability

- reduction of heat load by off-resonant pulse components

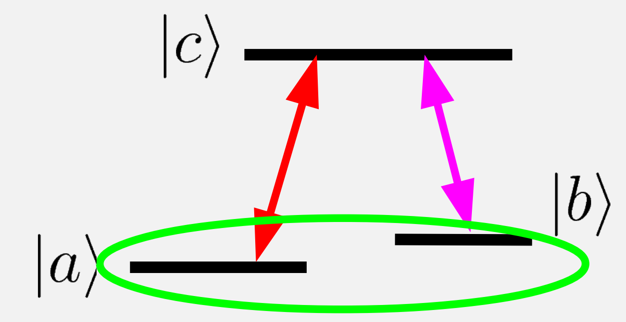
"Less obvious" implications:

- Longitudinal coherence affects the quantum dynamics of the target
  → possibility to affect/control quantum dynamics
  → advanced measurement / spectroscopy schemes

- Longitudinal coherence may enhance detection capabilities
  → interference in diffraction limited by longitudinal coherence
  → phase-sensitive "homodyne" measurements
  → Ramsey / multidimensional spectroscopy

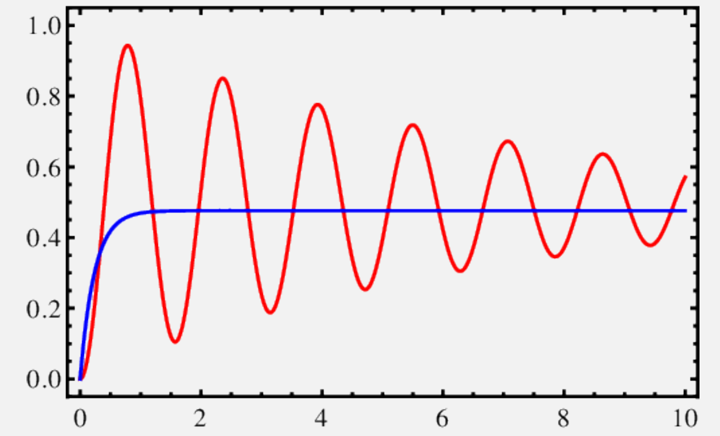
Examples

- EIT: Coherent laser fields create atomic coherence, which in turn modifies the interaction with the light
- Population dynamics: Rabi oscillation vs. incoherent rate dynamics
- Longitudinally coherent pulses yields stronger excitation than corresponding incoherent pulses



Electromagnetically induced transparency

population dynamics

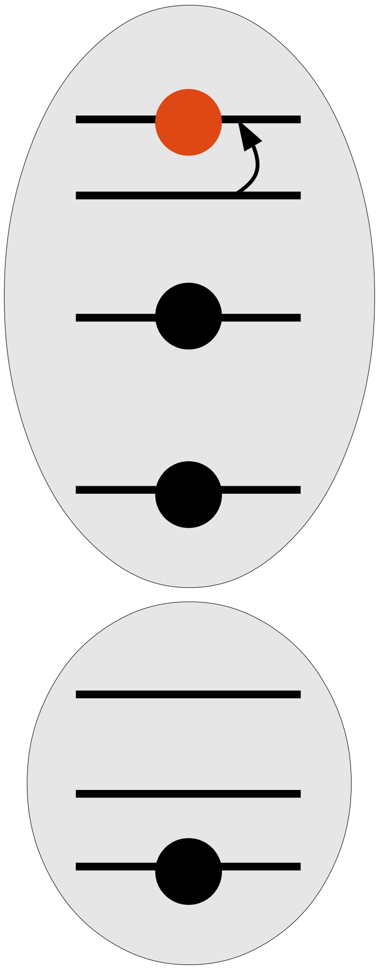


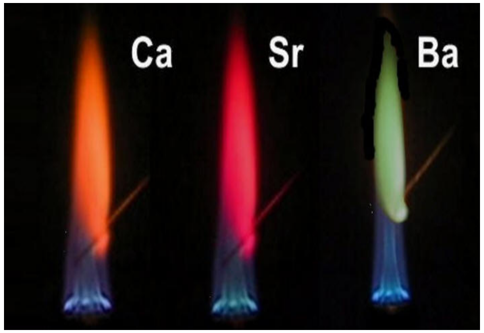
coherent light
incoherent light

longitudinally coherent light imprints coherence onto matter which favorably modifies the dynamics

160 years of light-matter interaction in one slide

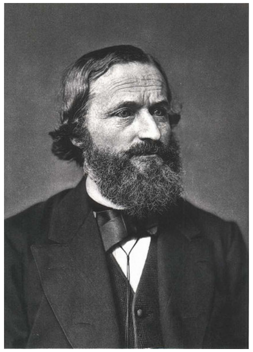
Atomic physics and quantum optics





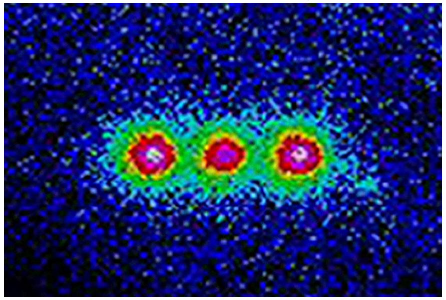
1859

“incoherent pump
and passive observation”



Bunsen Kirchhoff

today



“full quantum control”

Progress enabled via coherence, non-linearities, quantum effects
→ quantum optics

X-ray quantum optics

Light-matter interactions

different paradigms

uncontrolled pump
+ passive observation

full quantum
control

- X-ray physics could greatly benefit from moving more towards coherence/non-linear/quantum/control
- New light sources and upgrades
  → now is the right time
- Necessary to fully exploit new light sources

new tools in
x-ray physics

↔

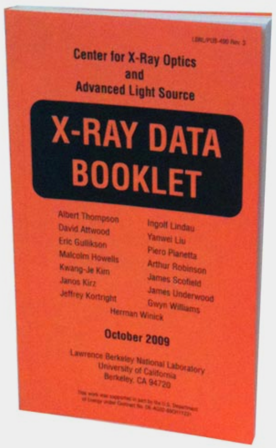
new platform for
quantum optics

Not entirely new:

many existing x-ray setups already rely on quantum optical concepts

Two branches of x-ray quantum optics

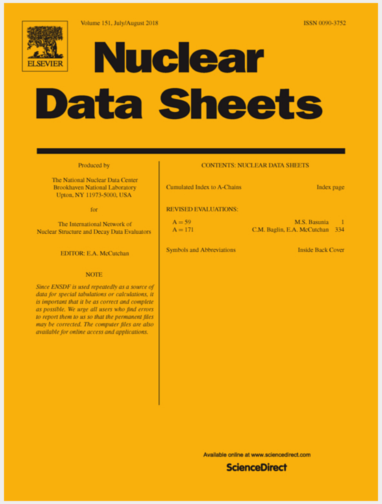
Electronic resonances (K-edge in ^{57}Fe)



$$E = 7.112 \text{ keV}$$
$$\sigma_e = 26 \times 10^{-20} \text{ cm}^2$$
$$\Gamma \approx 2 \text{ eV}$$
$$\tau \approx 0.33 \text{ fs}$$

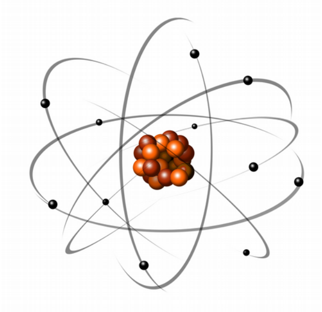
focus of XFEL research

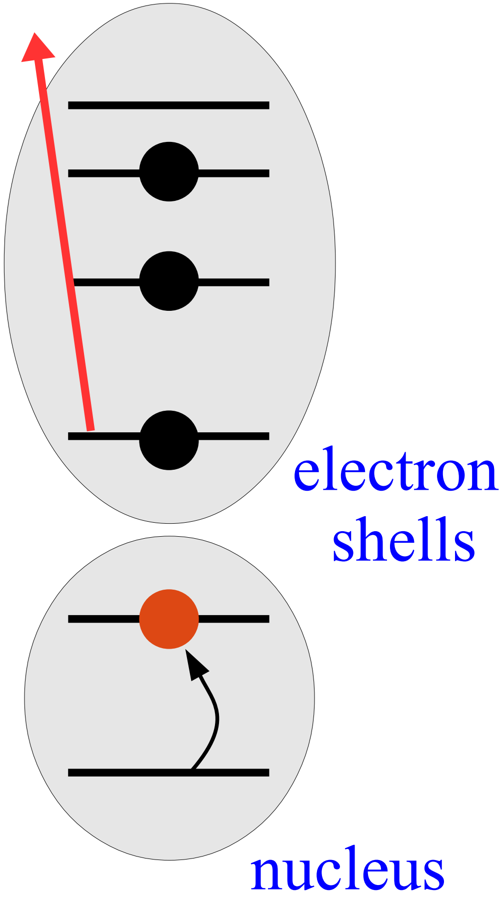
Nuclear resonances (Mössbauer transition in ^{57}Fe)



$$E = 14.412 \text{ keV}$$
$$\sigma_e = 256 \times 10^{-20} \text{ cm}^2$$
$$\Gamma = 4.7 \times 10^{-9} \text{ eV}$$
$$\tau = 141 \text{ ns}$$

Extremely narrow resonance





How could XFEL benefit from narrow resonances?

Extreme monochromatization

Precision spectroscopy and fundamental tests

Quantum optics and nonlinear science

coherent control techniques

Narrow resonances as a tool

Qualitatively new Mössbauer science

X-ray quantum optics

advanced spectroscopy techniques

Coherent bridge from ~ fs scales to ~ 100ns scales

Bridge between x-ray and visible

Correlations and (out-of-equilibrium) dynamics

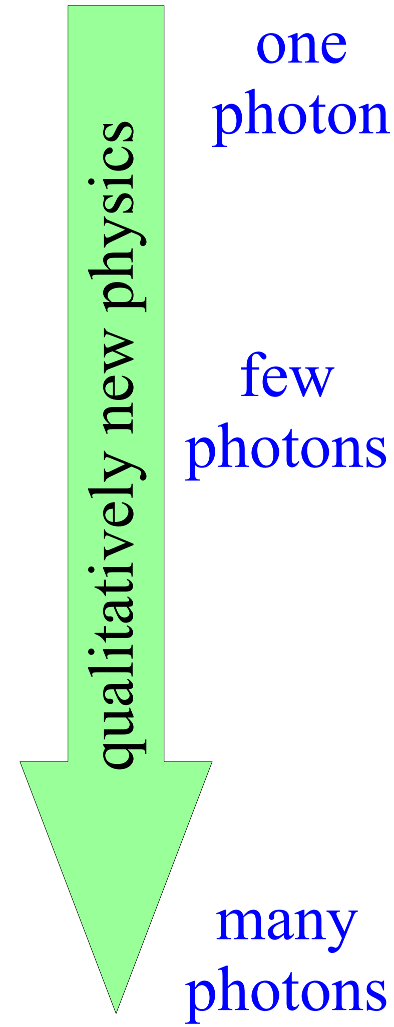
Complementary to science with electronic resonances

Mössbauer: Qualitatively new parameter regimes

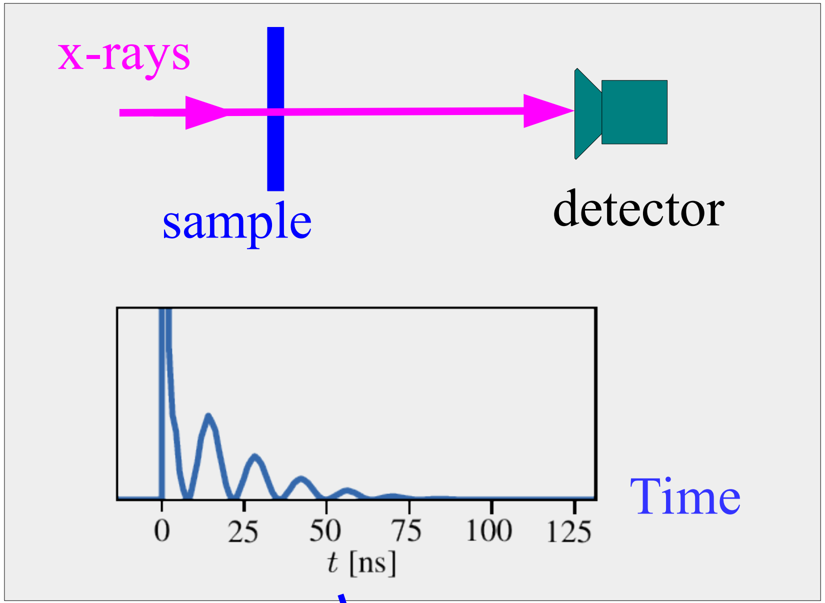
Example: ^{57}Fe

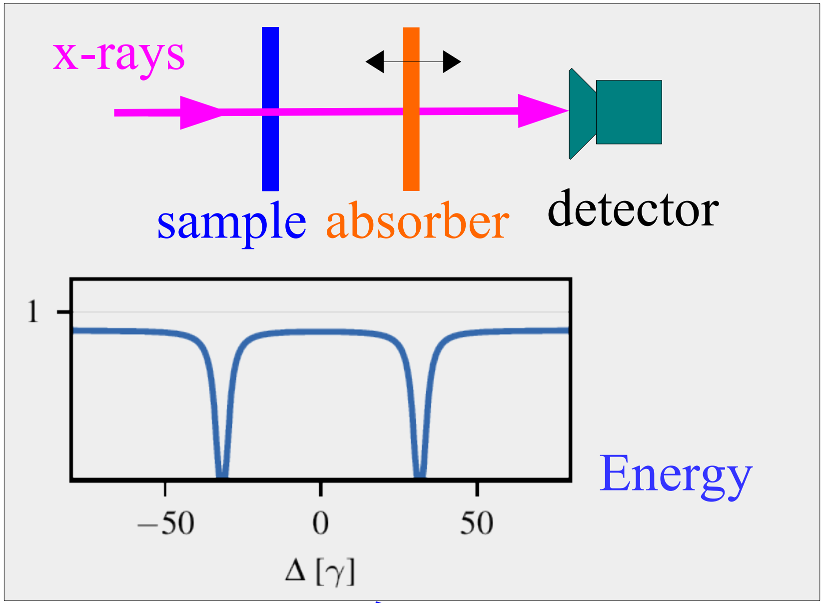
$$E = 14.4 \text{ keV} \qquad \gamma = 4.7 \text{ neV}$$
$$\tau = 141 \text{ ns}$$

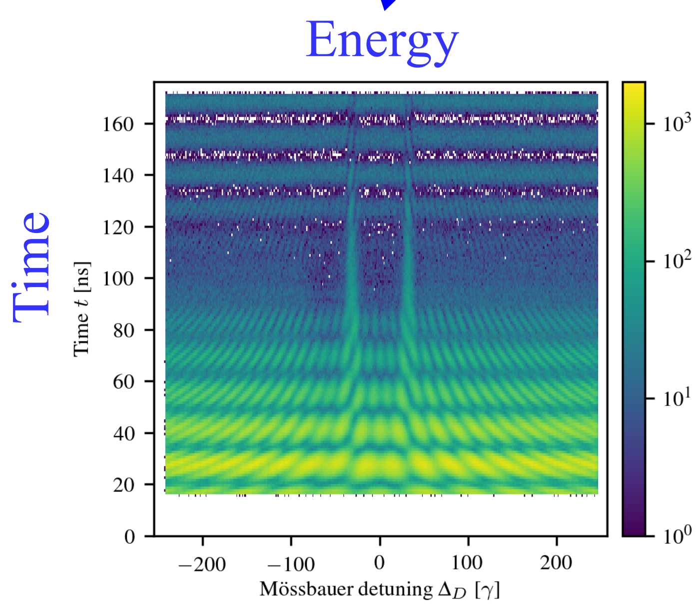
- P01 at Petra III:
  on average <1 resonant photon per pulse
- SASE XFEL: (10^{11} photons/pulse, $\Delta E/E \sim 10^{-3}$)
  on average ~30 resonant photons per pulse
- Self-seeding addition: (spectral brightness * 10)
  on average ~300 resonant photons per pulse
- XFELO: (2.2 mJ in 28meV at 12 keV, extrapolated to 14.4 keV)
  on average ~10^5 resonant photons per pulse
  potentially pulse-to-pulse coherence if stabilized



Exploiting correlations – single photon case







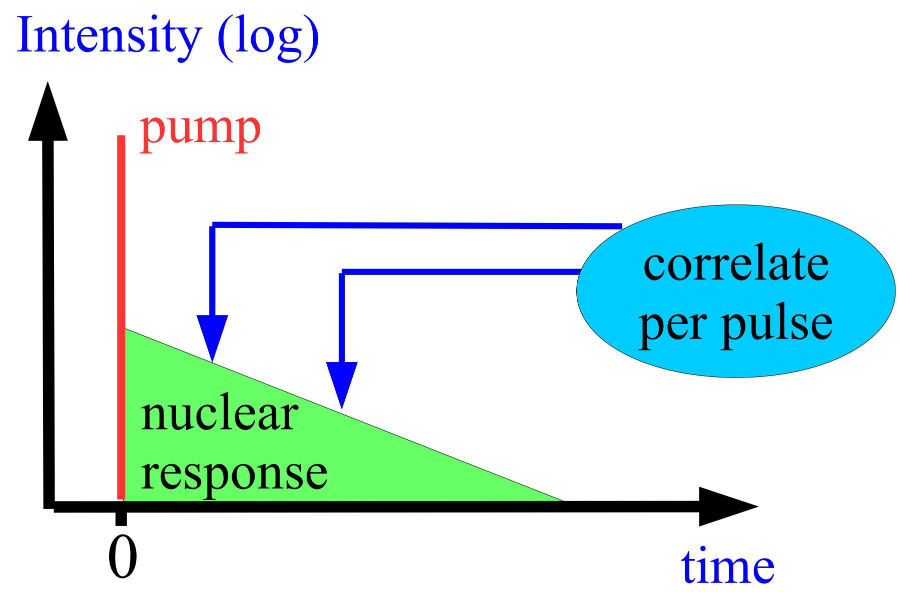
correlate
detection
at synchrotron

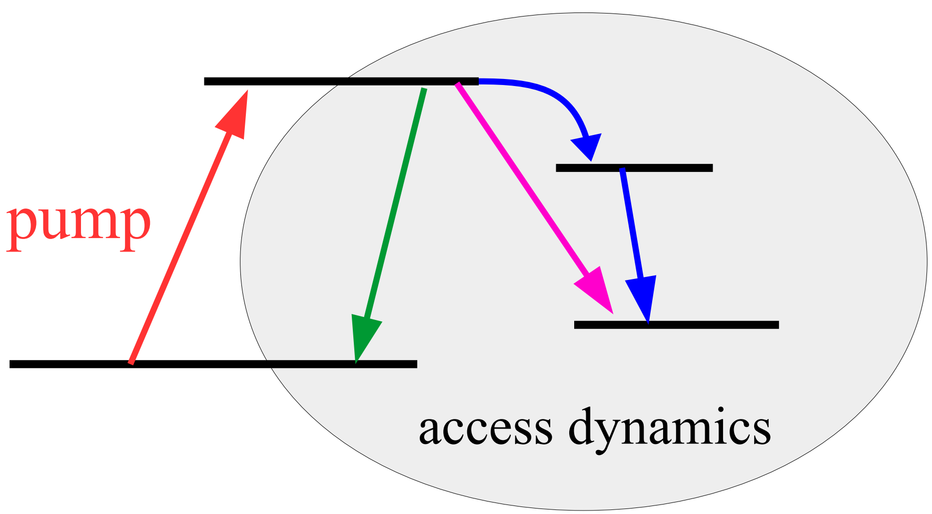
rich interference
structure provides
unique insight
into dynamics

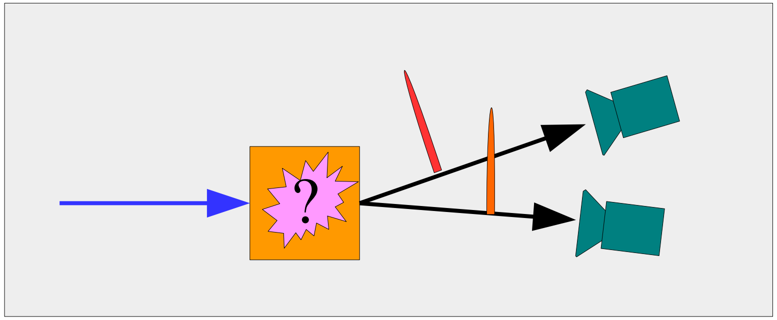
Exploiting correlations – multi-photon case

Few signal photons per pulse

- Study pump-induced dynamics using the delayed response
- Higher-order correlations characterizing the dynamics
- Distinguish different dynamics







spatial correlations?

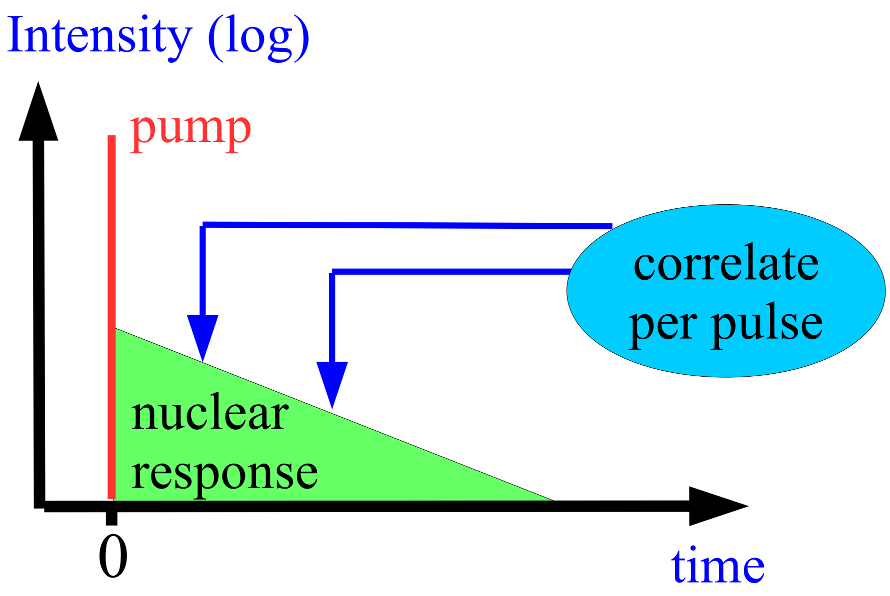
different final states

different pathways

Exploiting correlations – many photon case

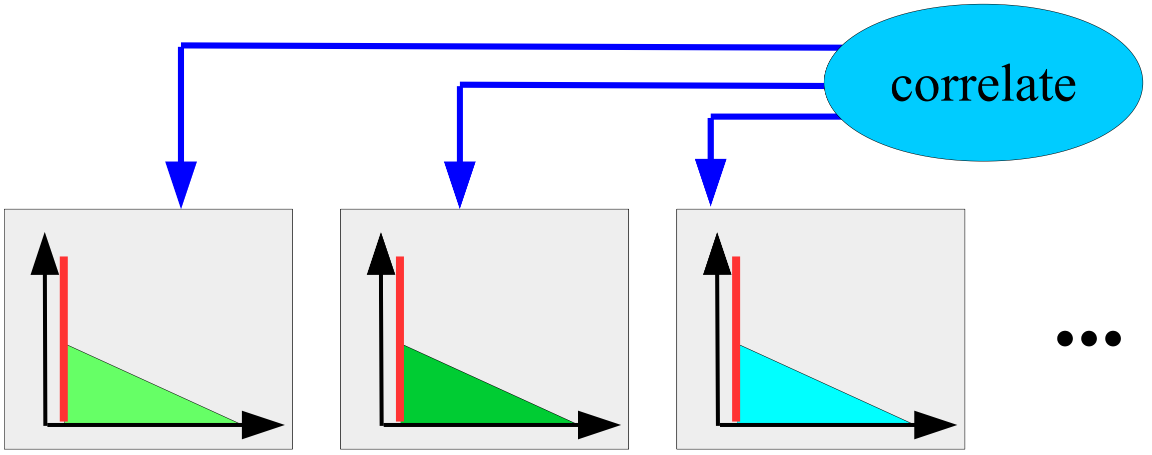
Few signal photons per pulse

- Study pump-induced dynamics using the delayed response
- Higher-order correlations characterizing the dynamics
- Distinguish different dynamics



Many signal photons per pulse

- “single shot spectra”
- compare/correlate different repetitions of pump-probe scheme
- out-of-equilibrium / non-cyclic/ non-ergodic dynamics

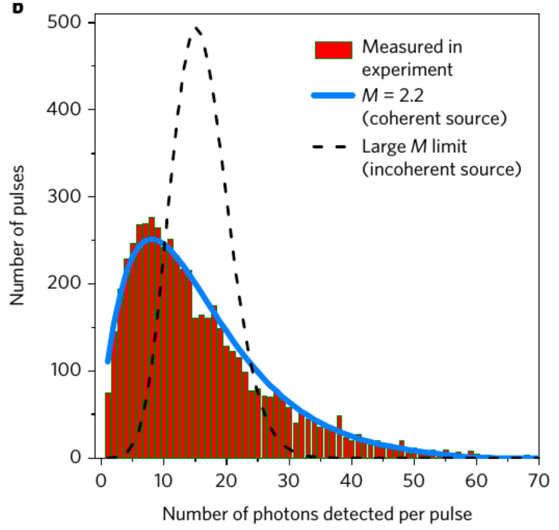


Proof-of-principle experiment

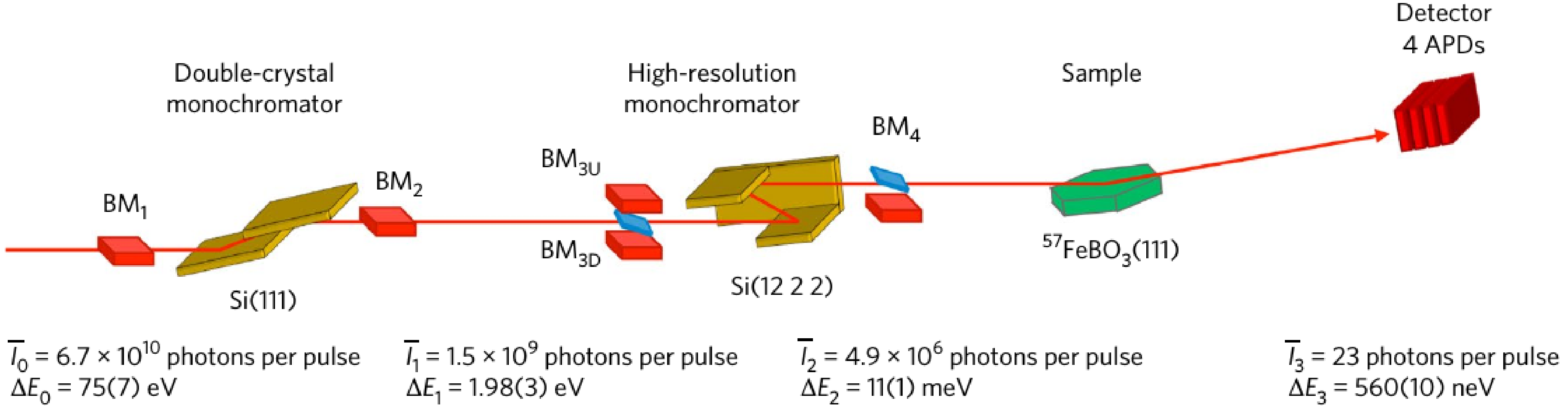
Superradiance of an ensemble of nuclei excited by a free electron laser

Aleksandr I. Chumakov, Alfred Q. R. Baron, Ilya Sergueev, Cornelius Strohm, Olaf Leupold, Yuri Shvyd'ko, Gennadi V. Smirnov, Rudolf Rüffer, Yuichi Inubushi, Makina Yabashi, Kensuke Tono, Togo Kudo & Tetsuya Ishikawa

Nature Physics **14**, 261–264 (2018) | Download Citation

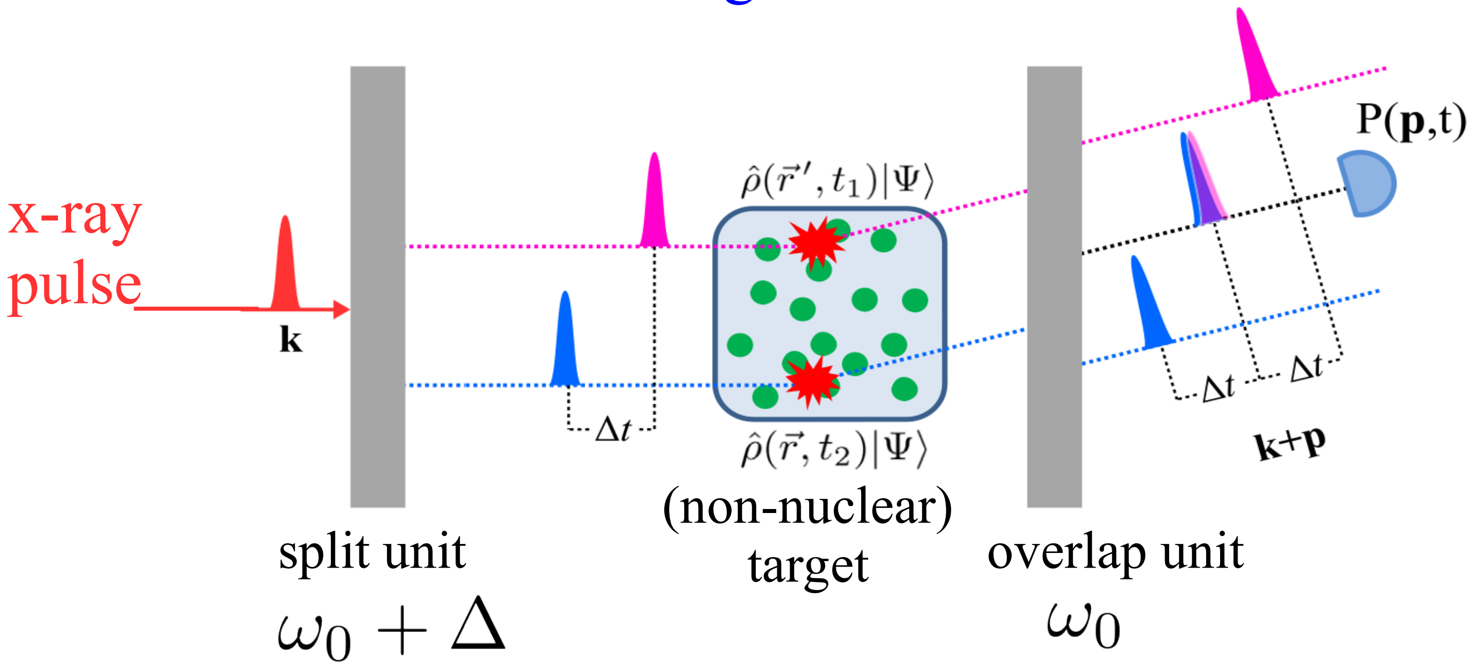


- First FEL experiment with Mössbauer nuclei (at SACLA)
- Observation of correlations between photons from each shot separately, detected "one at a time"
- Example question: How does the initial emission dynamics depend on the degree of excitation?



Time domain interferometry: Nuclei as a tool

Correlations in "non-nuclear" targets:



$$P(\vec{p}, t) \propto |f(t)|^2 \left[S(\vec{p}, 0) + S(\vec{p}, t) \cos(\Delta t)\right]$$

Access to intermediate scattering function in "gap region" from ~1- ns to ~100 ns with neV energy resolution and essentially without background

(TDI proposed for applications at XFELs in SwissFEL science case)

A. Baron et al., Phys. Rev. Lett. 79, 2823 (1997)

Science cases

Promote Mössbauer-based science to the study of time-dependent out-of-equilibrium phenomena

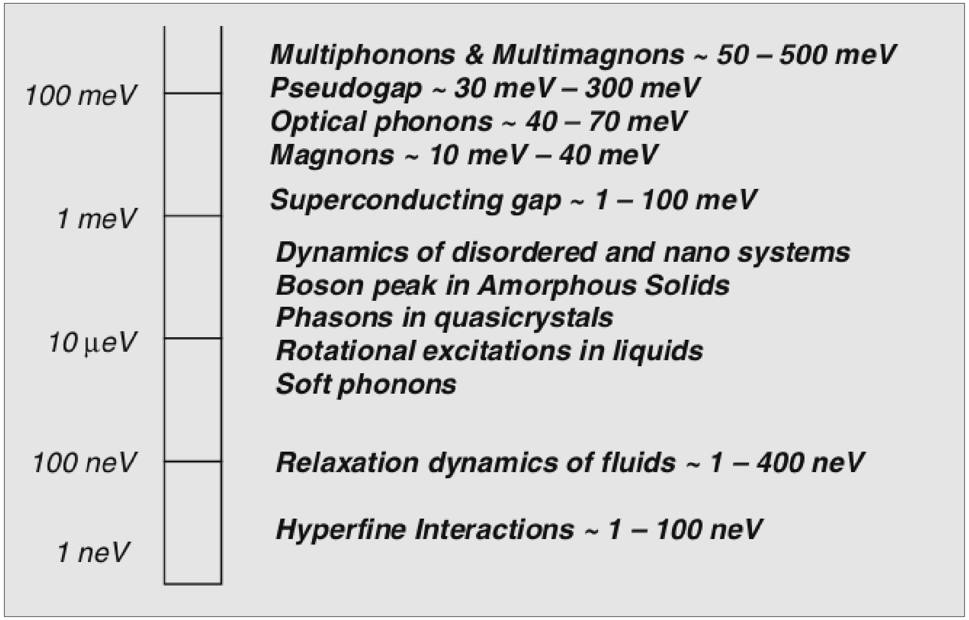
- Pump via (x-ray / optical / heat / pressure / elm. Fields / …), probe via nuclear response

  → e.g., optically excited molecular switches
  → e.g., magneto-optical nanomaterials

Access low-energetic condensed-matter excitations on neV-meV and nm-μm scales

- Probe low-energetic condensed-matter excitations on neV-meV and nm-μm scales

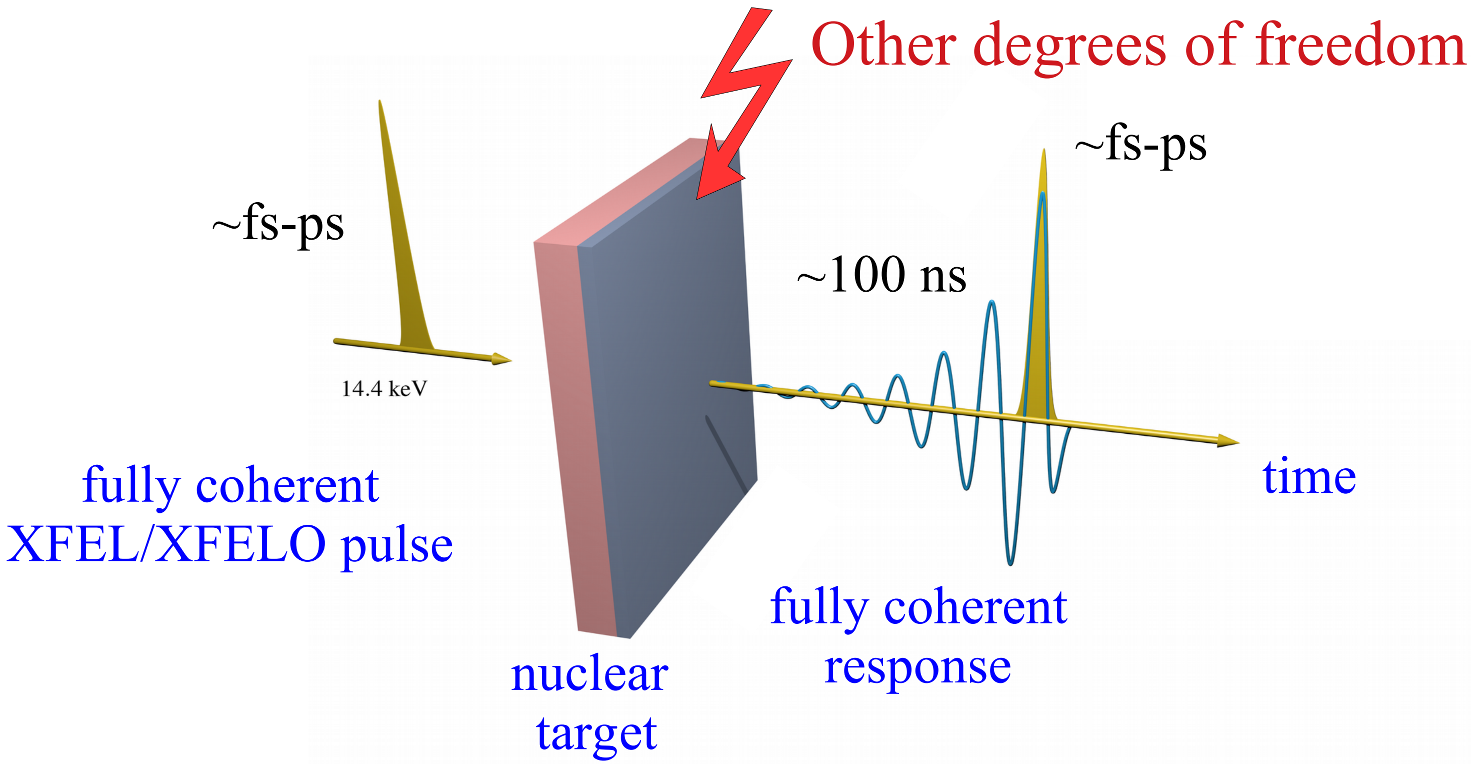
  → e.g., physics of glasses
  → e.g., mesoscopically structured materials
  → diffusion phenomena



Report: Scientific opportunities with XFELOs arXiv:1903.09317 [physics.ins-det]

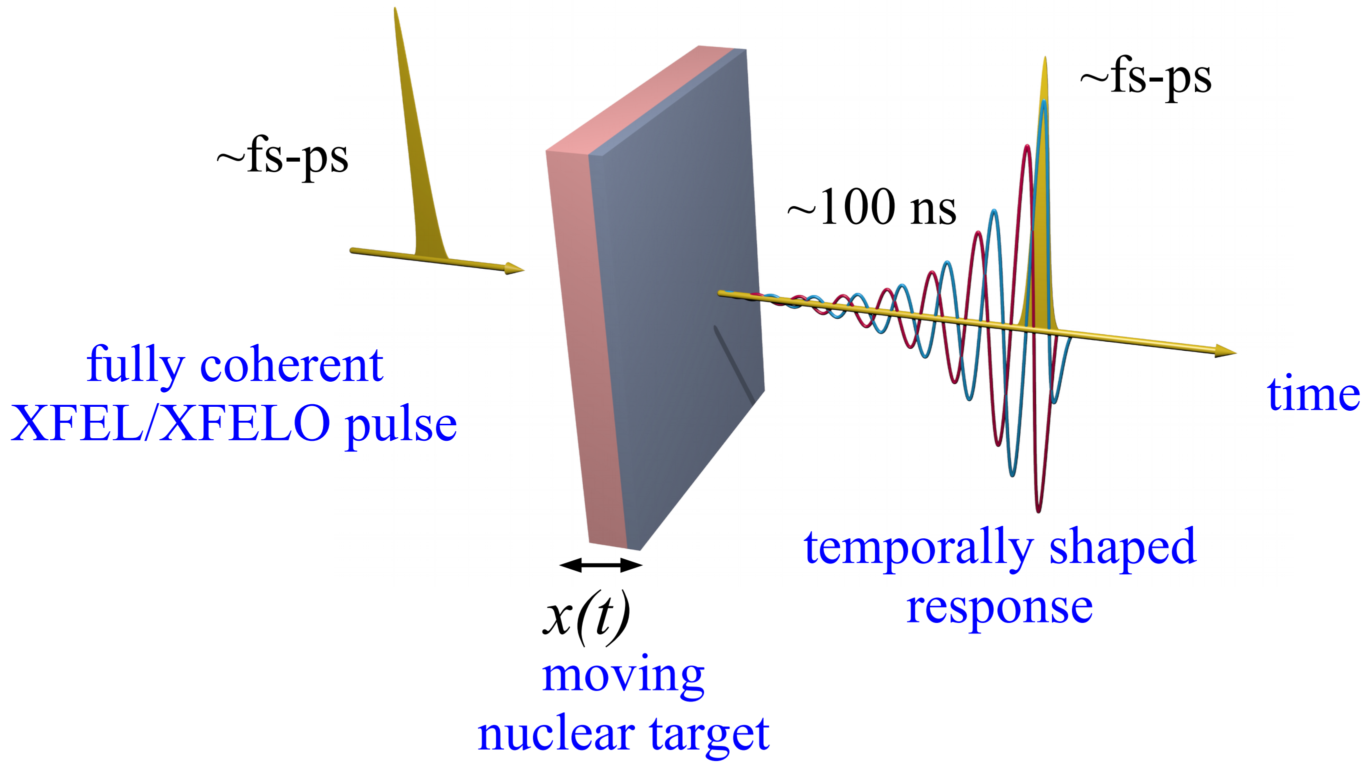
Shenoy&Röhlsberger, Hyperf. Int. 182, 157 (2008)

X-ray optical control of nuclei



- Nuclear resonance scattering naturally extends the longitudinal coherence to the ~100ns timescale
- Interference between pump pulse and response can be used for "homodyne" detection
- Can pump-probe-like ideas be used directly on nuclei?
  → probe external influence on nuclei (e.g. coupling to other subsystems)
  → important tool for nuclear quantum optics

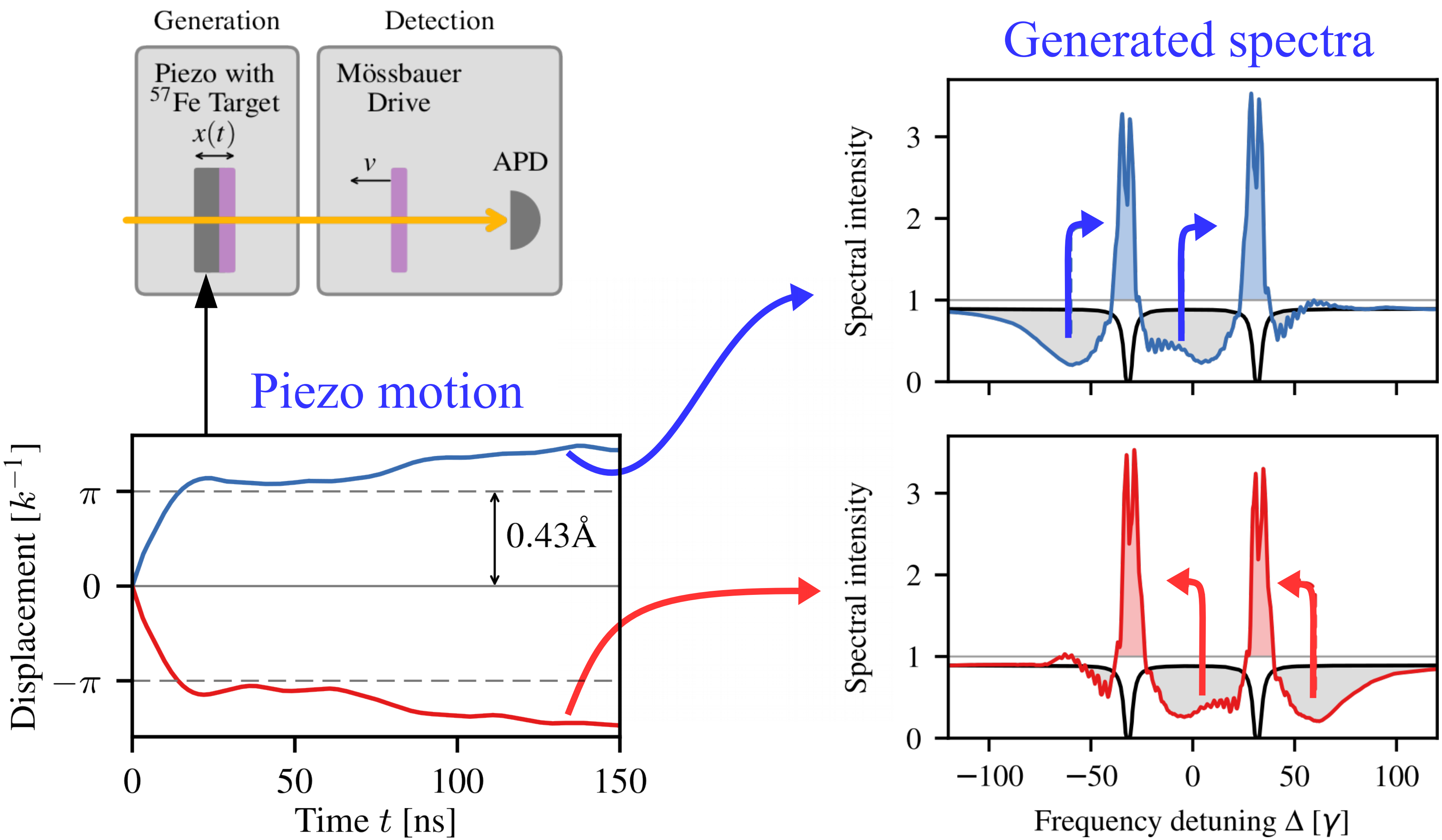
First step: X-ray pulse shaping



- Motion of the nuclear target imprints a time-dependent phase onto the nuclear response
- Effectively: controlled dynamical boost of real part of index of refraction by orders of magnitude without affecting absorption/imaginary part

$$n = 1 - \delta[x(t)] + i\beta$$

"X-ray afterburner" (ID 18, ESRF)



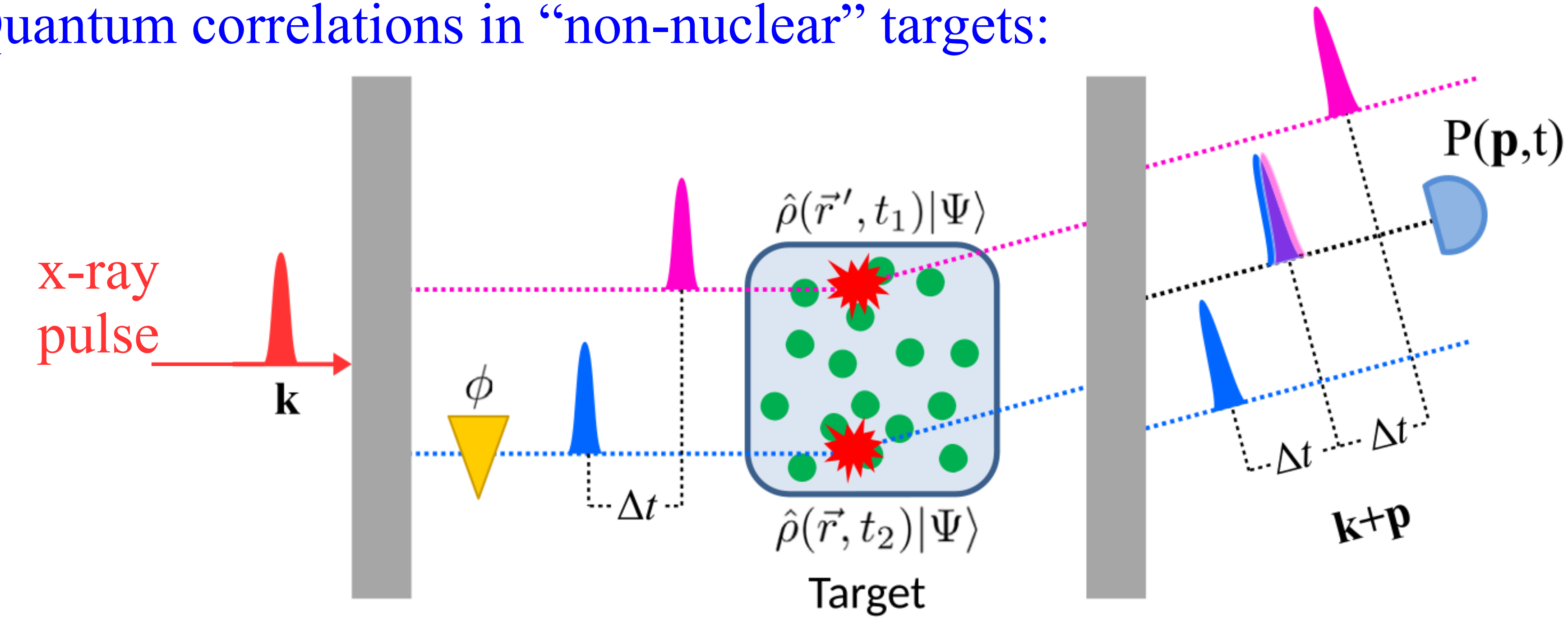
Resonant "gain" exceeding all loss channels
Mechanical motion controlled and measured on ns / sub-Å level

Black line:
no motion

Heeg et al., Science **357**, 375 (2017)

Immediate application: Phase-control in TDI

Quantum correlations in “non-nuclear” targets:

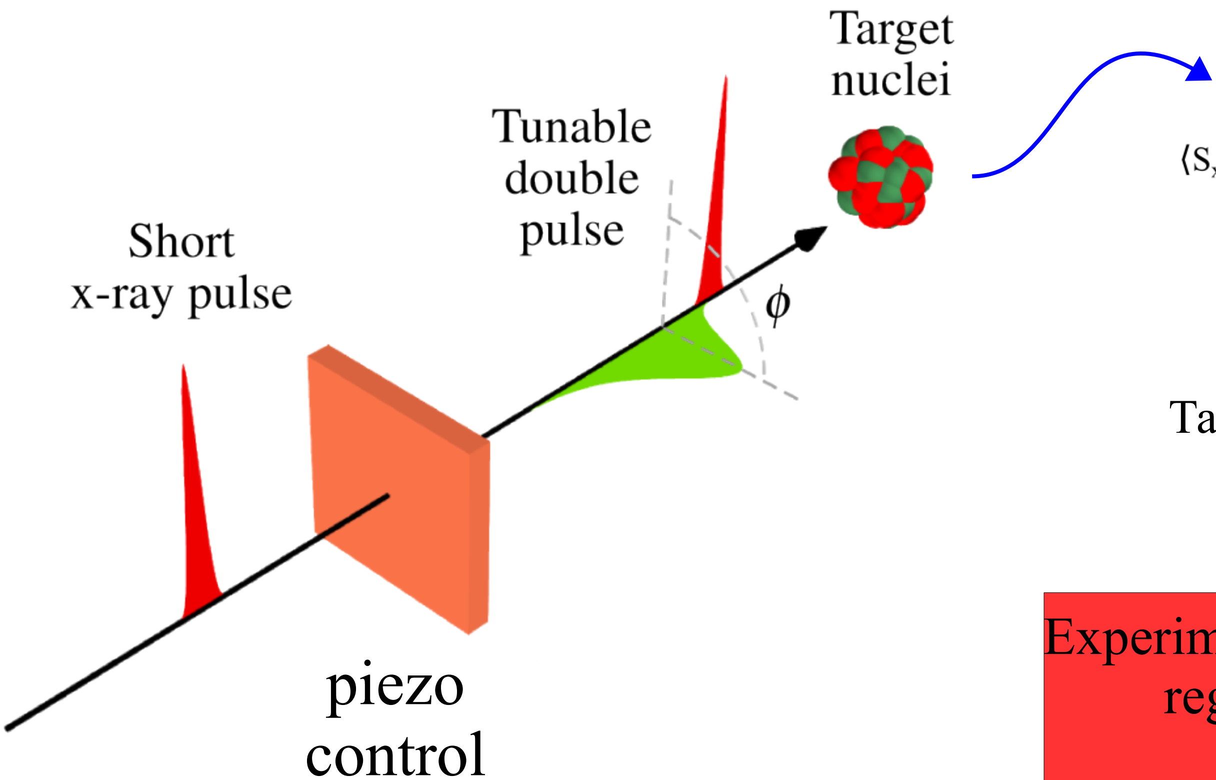


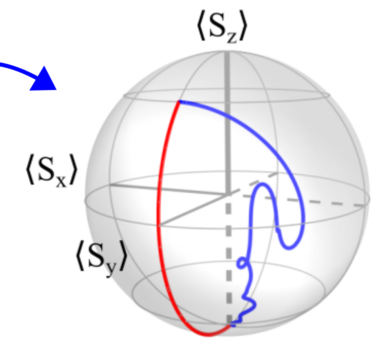
$$P(\mathbf{p},t) \propto f(t)^2 \left(\sum_{j=1,2} S_{qu}(\mathbf{p},t_j,t_j) \; + 2\{\cos[\phi] S^R_{qu}(\mathbf{p},t_1,t_2) - \sin[\phi] S^I_{qu}(\mathbf{p},t_1,t_2)\} \right)$$

real part — imaginary part

Phase-control of one of the two scattering pathways provides access to real and imaginary parts of the complex intermediate scattering function
→ quantum mechanical correlations

Towards nuclear pump-probe experiments





Tailor nuclear dynamics on
Bloch sphere

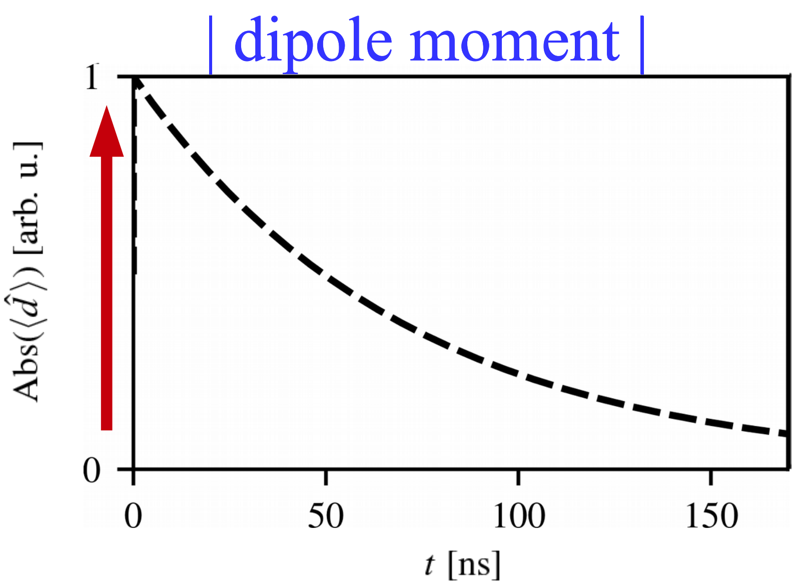
Experiment so far in single-photon regime at synchrotrons: linear response

”true” pump-probe requires strong driving → XFEL(O)

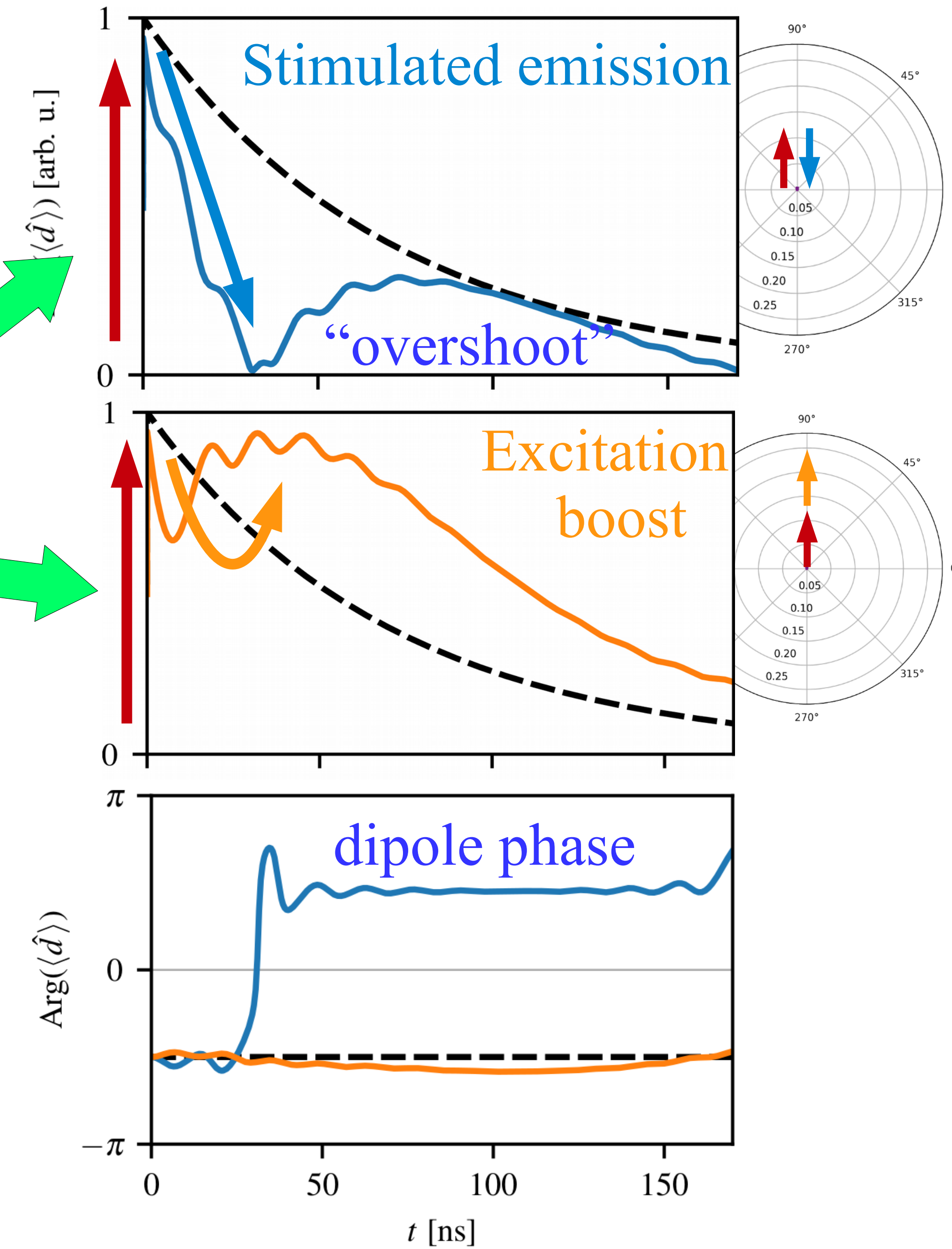
- First pulse excites nuclear target
- Piezo control shapes second pulse part
- Double-pulse determines the dynamics of the nuclear target

Experimental results: x-ray optical control of nuclei

Preparation pulse
excites the system:



Regular decay without
second pulse



X-ray optical control of
nuclear dynamics

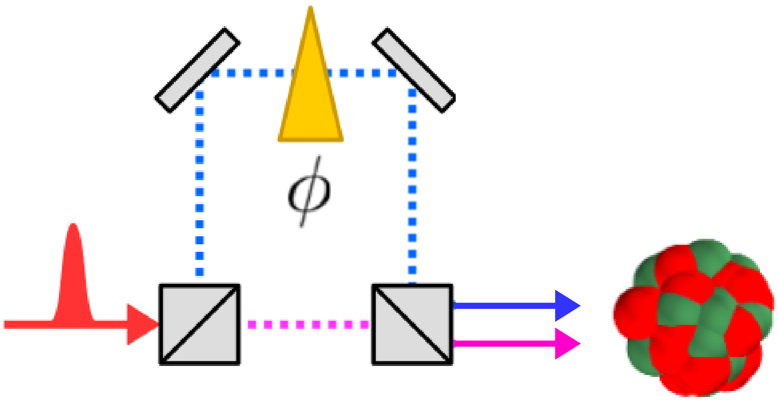
stable to fractions of x-ray wavelength

K. P. Heeg et al, submitted

Why is the control so stable?

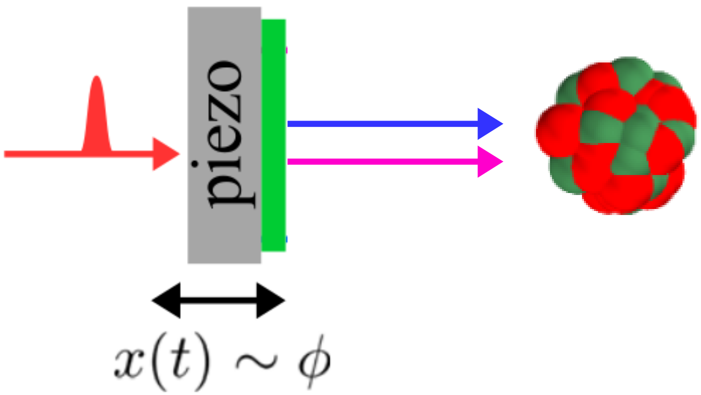
“Conventional approaches”

- Interfering pathways spatially separated
- Geometry must be stabilized throughout the entire long accumulation of statistics



Piezo control with mechanical motion

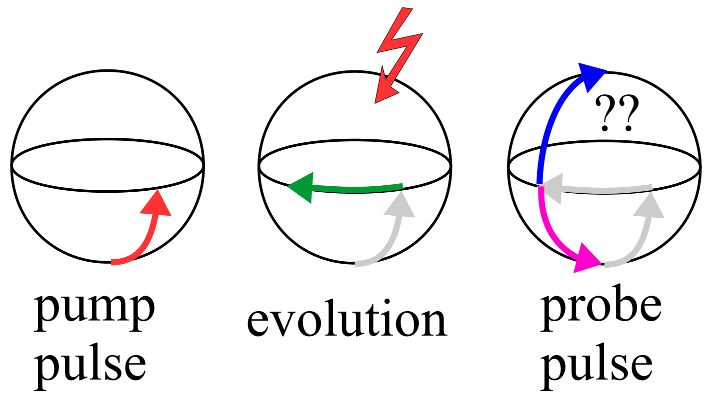
- Interfering pathways coincide in space
- Control depends on motion relative to the geometry at the time of excitation
- Geometry only needs to be stable for a ~ 200 ns measurement interval after each x-ray pulse
- All other drifts / noise do not matter!



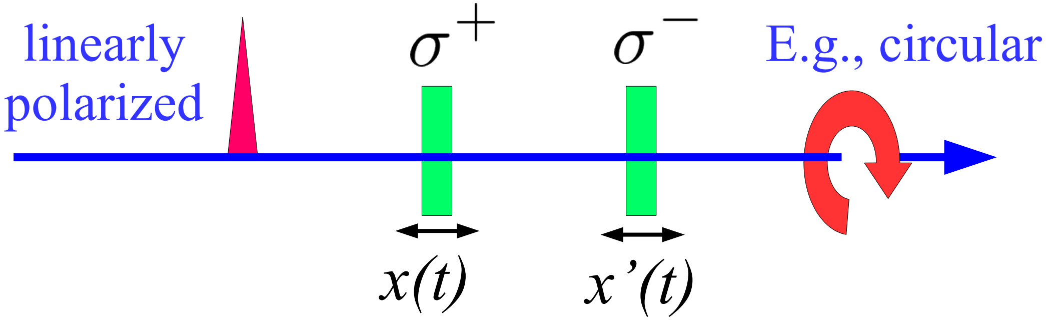
Piezo control for nuclear quantum optics

Advanced spectroscopy (e.g., Ramsey)

- Robust method to precisely measure frequencies, dynamics of coherences, external couplings
- First step towards multidimensional spectroscopy

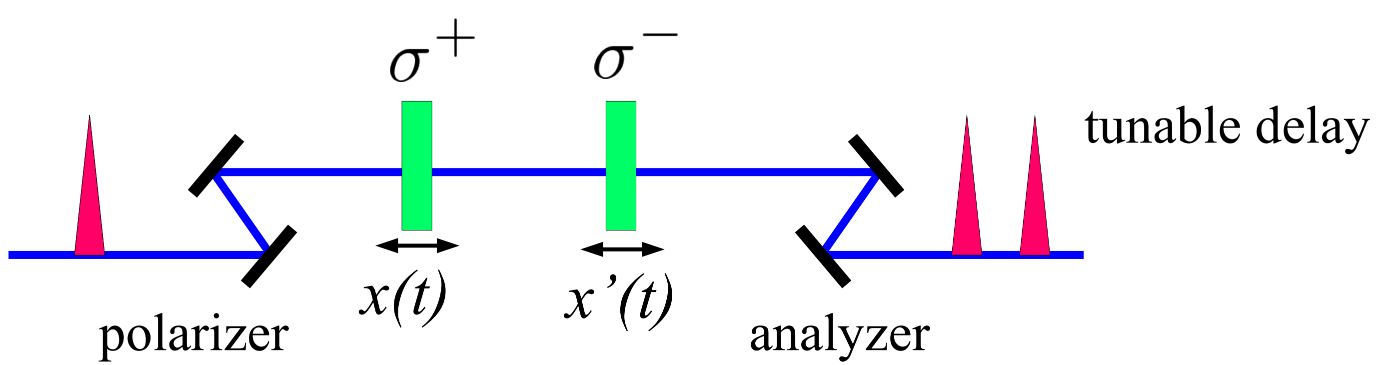


Dynamical polarization control



Motion-induced birefringence/ ”waveplates”

“True” double pulses



Crucial: Target design

- Smaller targets / higher resolution

  → interfaces (magnetism, friction, ...)

  → spatially resolved dynamics (heat, magnetization, chemistry)

- Structured targets

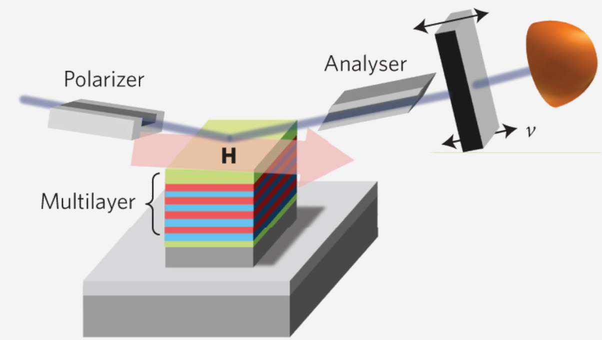
  → design artificial quantum systems (e.g., EIT with two-level systems)

  → enhance interaction between light and matter (e.g., via cavities, waveguides)

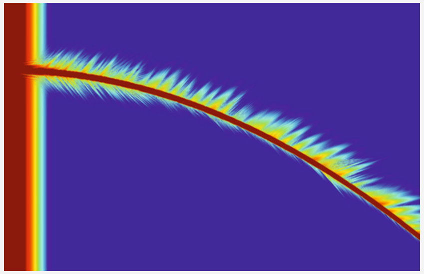
  → x-ray nanophotonics: photonic crystals / light-matter quasiparticles / x-ray photonic circuits

  → decoherence control via engineering of environment (e.g. SGC)

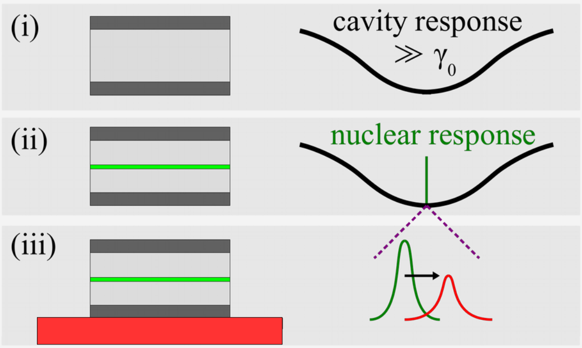
  → evanescently coupled sensing schemes



Röhlsberger et al., Nat. Phot. 10, 445 (2016)

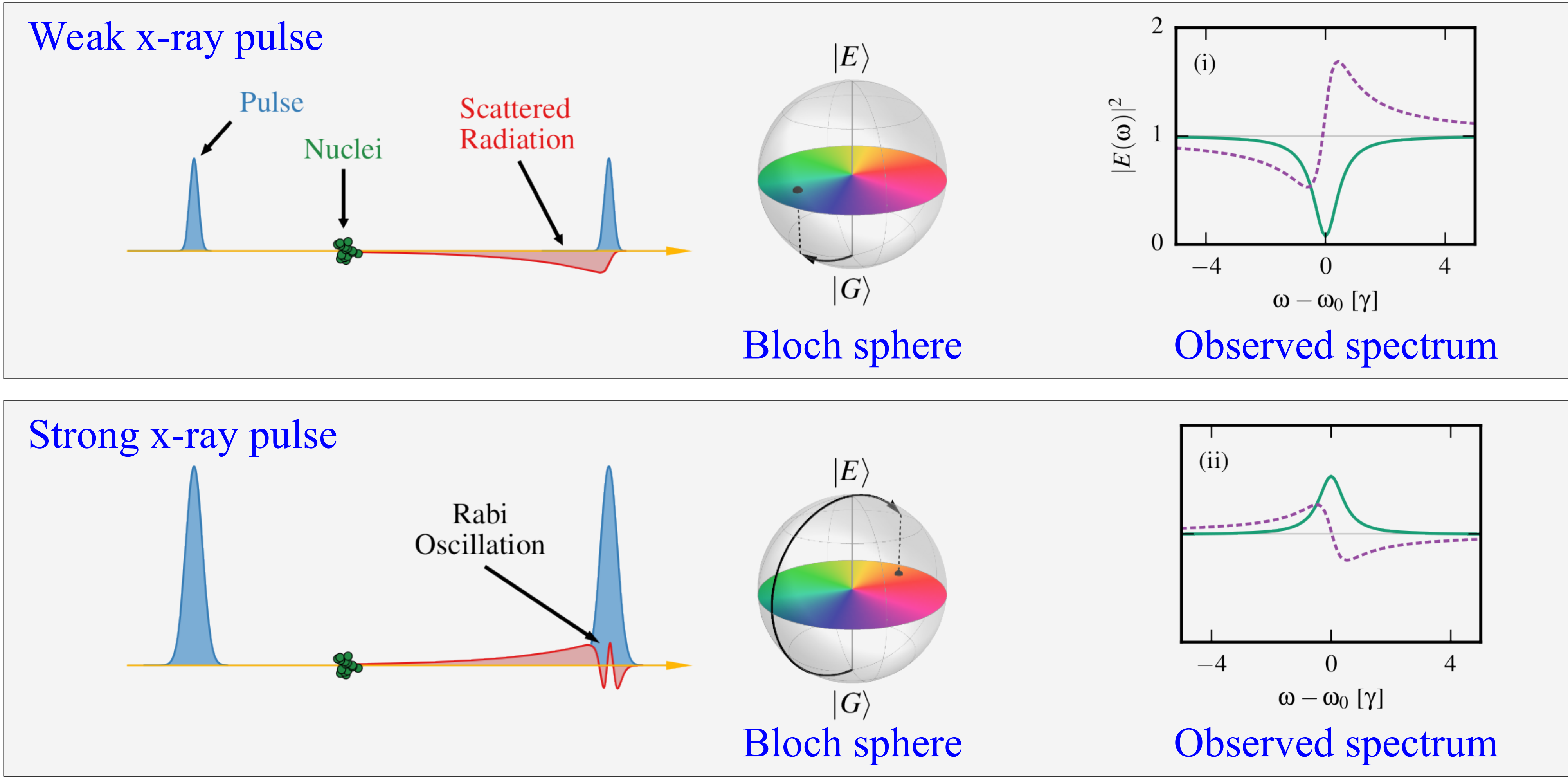


nanoscale waveguide
Salditt et al., PRL 2015



Evers et al., in preparation

Towards non-linear science with nuclei: Rabi flopping



Dipole phase sign change upon each half Rabi cycle leads to flip of spectra

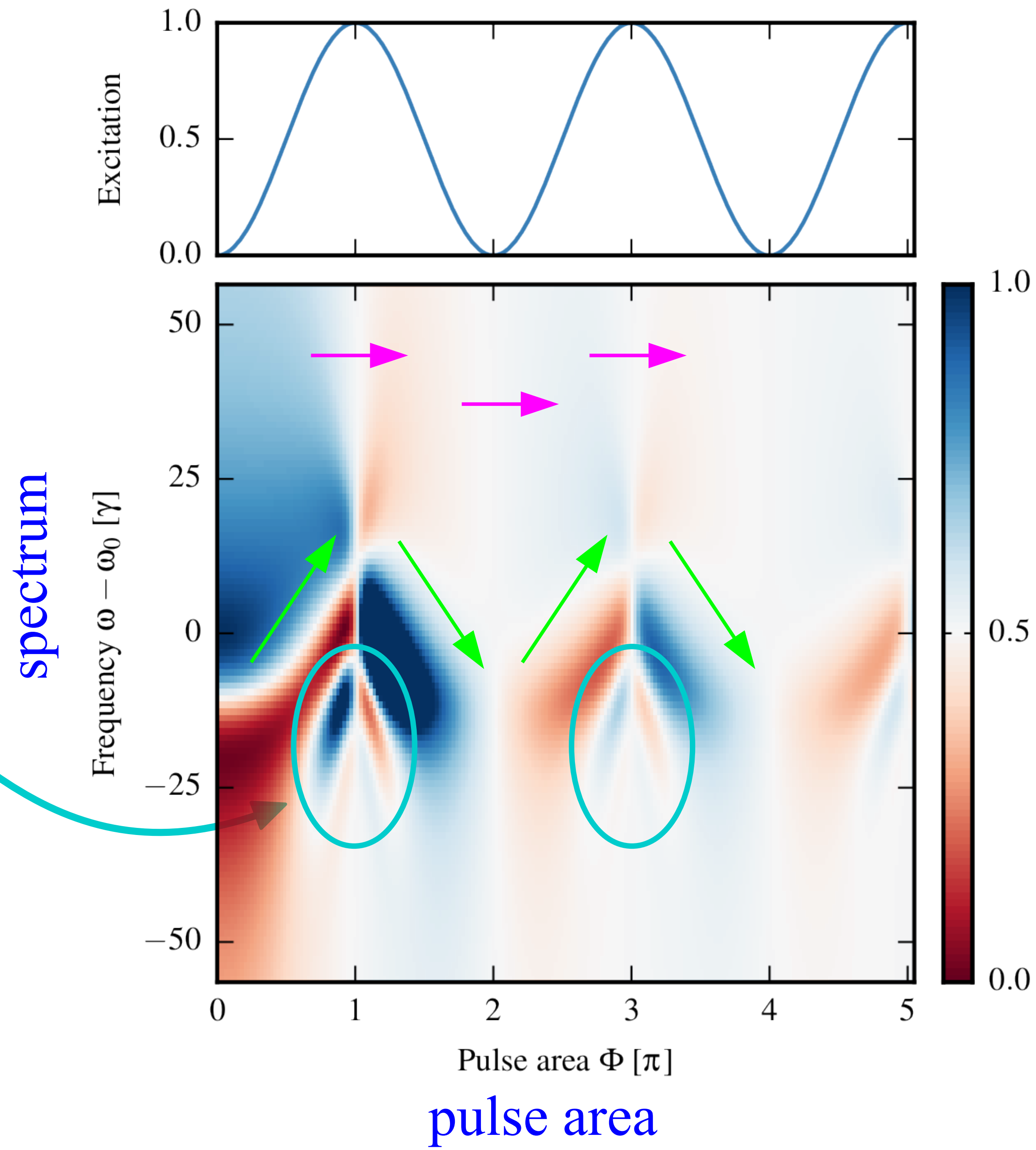
$$E(t) = A_0\delta(t) - \frac{\beta d}{2}\sin\left(\Phi\right)\theta(t)e^{-\frac{\gamma}{2}t}$$

Heeg, Keitel, Evers, arXiv:1607:04116 [quant-ph]

Nonlinear nuclear quantum optics

Signatures of strong excitation of nuclei in x-ray cavities

- Symmetry flips of the spectra of the delayed photons
- Corrections due to collective Lamb shifts
- Additional spectral signatures due to non-exponential collective decay



Non-linear signatures may be within reach with XFELO, focusing, optimized sample design

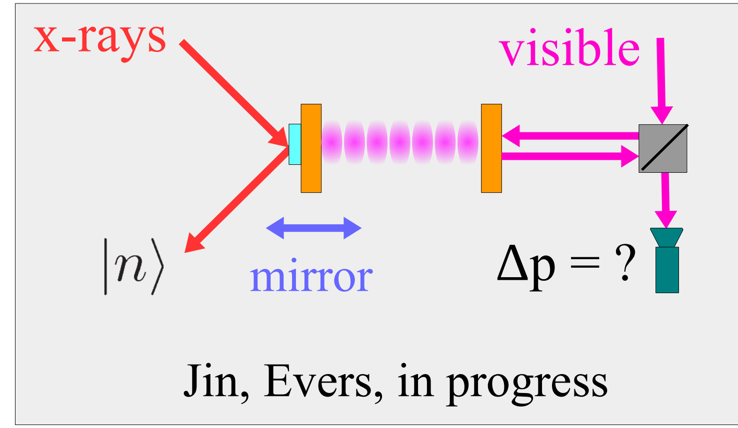
also c.f. Chumakov FEL experiment

Quantum interfaces linking x-rays and "low frequency"

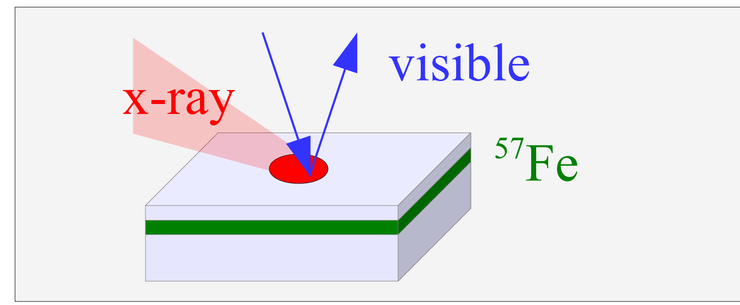
Vision: coherent link between x-rays and lower frequencies to combine best of both "worlds"

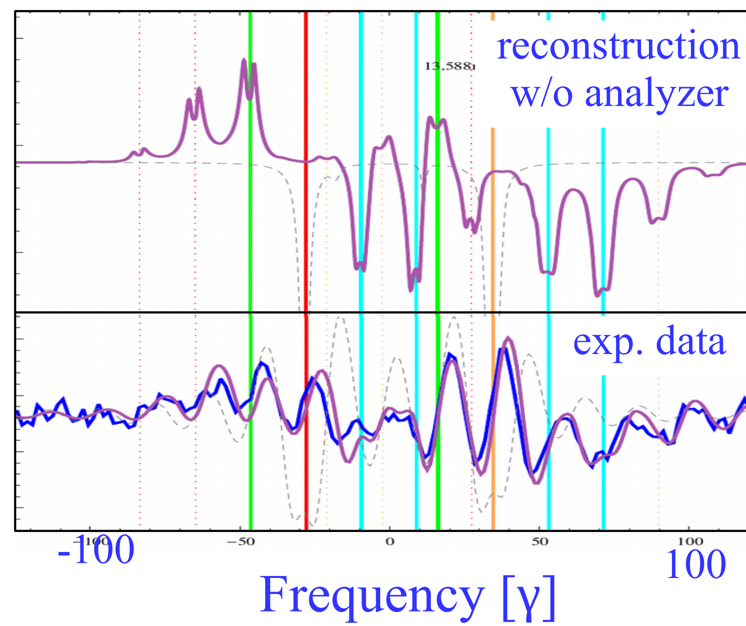
4.7 neV / h = 1.1 MHz – comparable scales

- X-ray optomechanics
  - → Manipulate x-rays using optical field (proposal Palffy et al, 2016)
  - → Macroscopic x-ray Fock state generation
- Coupling via common target
  - → x-ray detection of laser-induced motion (e.g. Kocharovskaya et al)
  - → visible-pump / nuclear probe (e.g. Schünemann et al)
- Nuclear frequency combs
  - → Modulate nuclear spectra using RF-fields (higher frequencies?) (Kocharovskaya et al, Evers, Pfeifer, Röhlsberger et al)



Jin, Evers, in progress





Summary

- Narrow nuclear resonances at XFELs:
  - useful as a tool
  - qualitatively new Mössbauer science
  - platform for x-ray quantum optics
- Seeding/XFELO schemes provide access to qualitatively different regimes for nuclear resonances
- Promote nuclear resonance scattering to time resolved (pump-probe) study of dynamical (out-of-equilibrium) processes
- Access to low-energetic solid-state excitations
- Coherent x-ray optical control of nuclei demonstrated in single-photon regime
  → nuclear pump-probe with seeding/XFELO?
  → Ramsey / multidimensional spectroscopy?
  → probe external influence on nuclei, other subsystems
- Interfaces between x-ray and "low frequency"

Acknowledgements:

Heeg, Kaldun, Strohm, Reiser, Ott, Subramanian, Lentrodt, Haber, Wille, Goerttler, Rüffer, Keitel, Röhlsberger, Pfeifer, Evers,
Science 357, 375 (2017)

Castrignano & Evers,
PRL 122, 025301 (2019)

MPIK Heidelberg, Germany

DESY, Hamburg, Germany

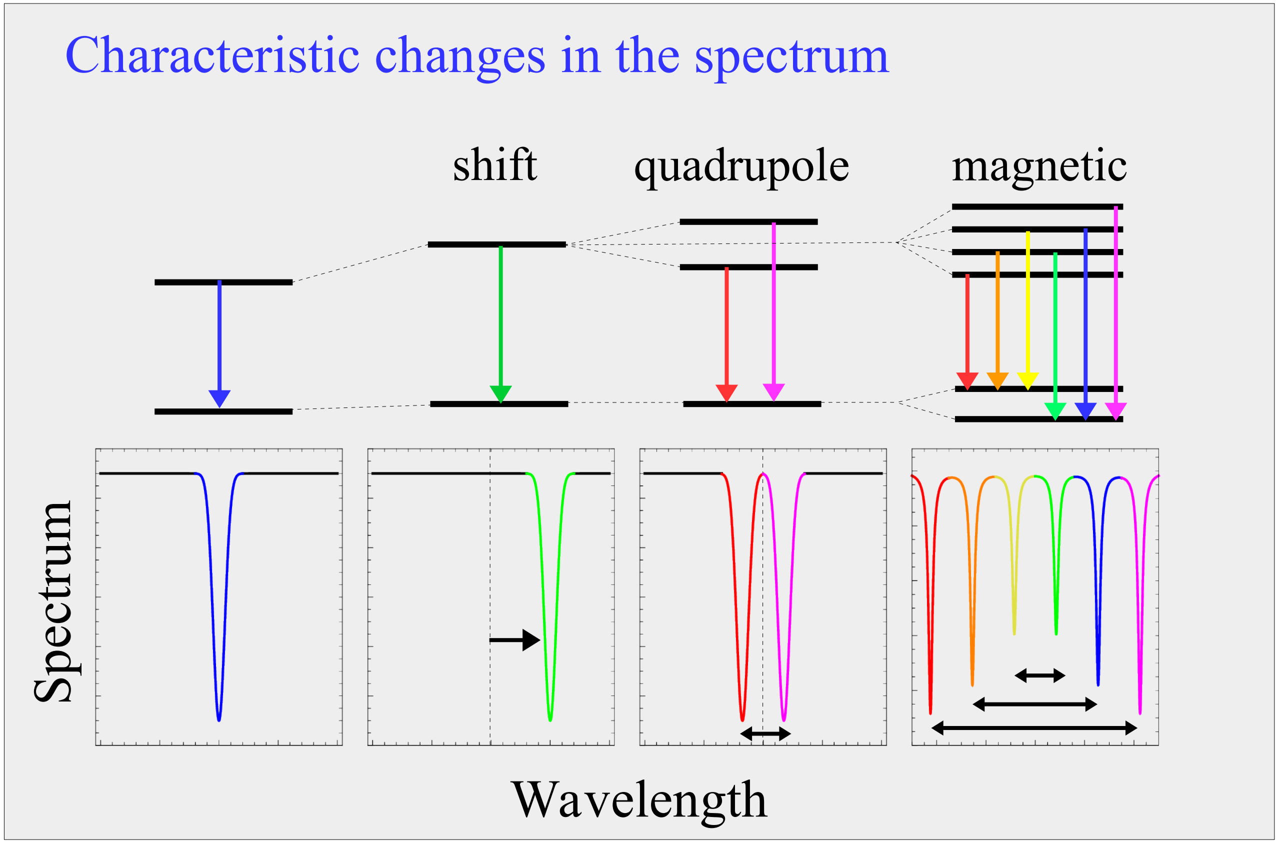
ESRF, Grenoble, France

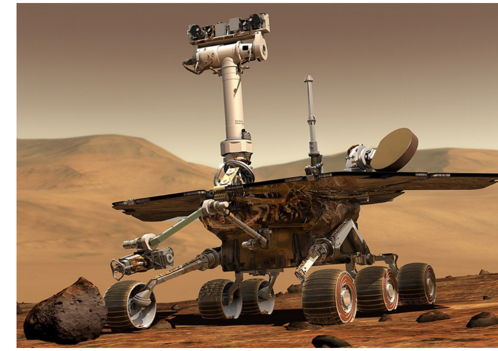
Report on the 2016 SLAC workshop on the scientific opportunities of XFELOs
arXiv:1903.09317 [physics.ins-det]

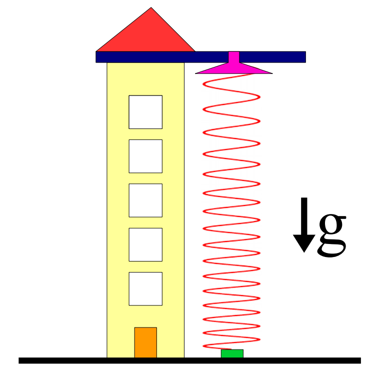
Shenoy&Röhlsberger,
Hyperf. Int. 182, 157 (2008)

Discussions with many colleagues

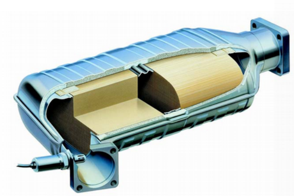
Mössbauer nuclei: sensitive probe of the environment





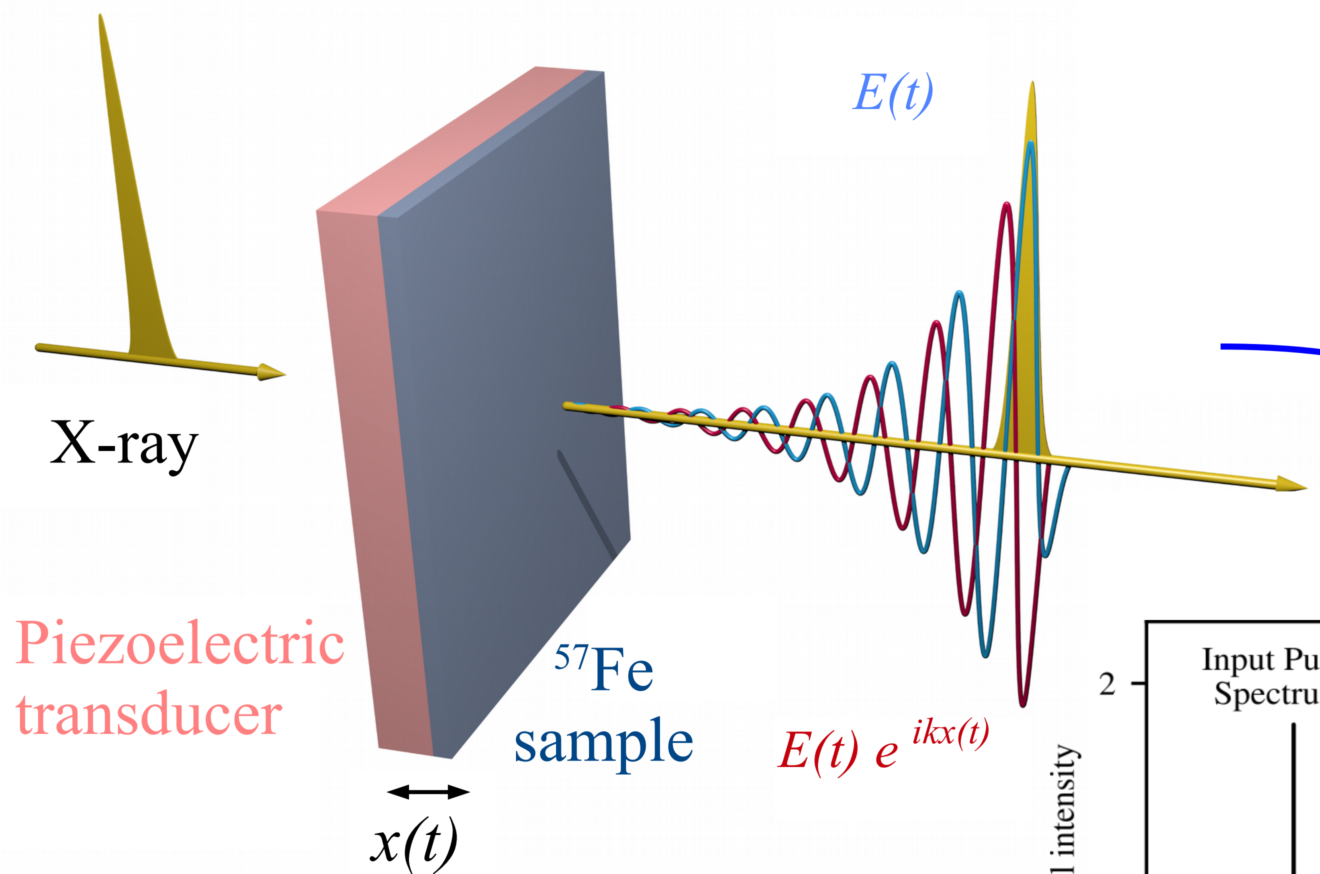


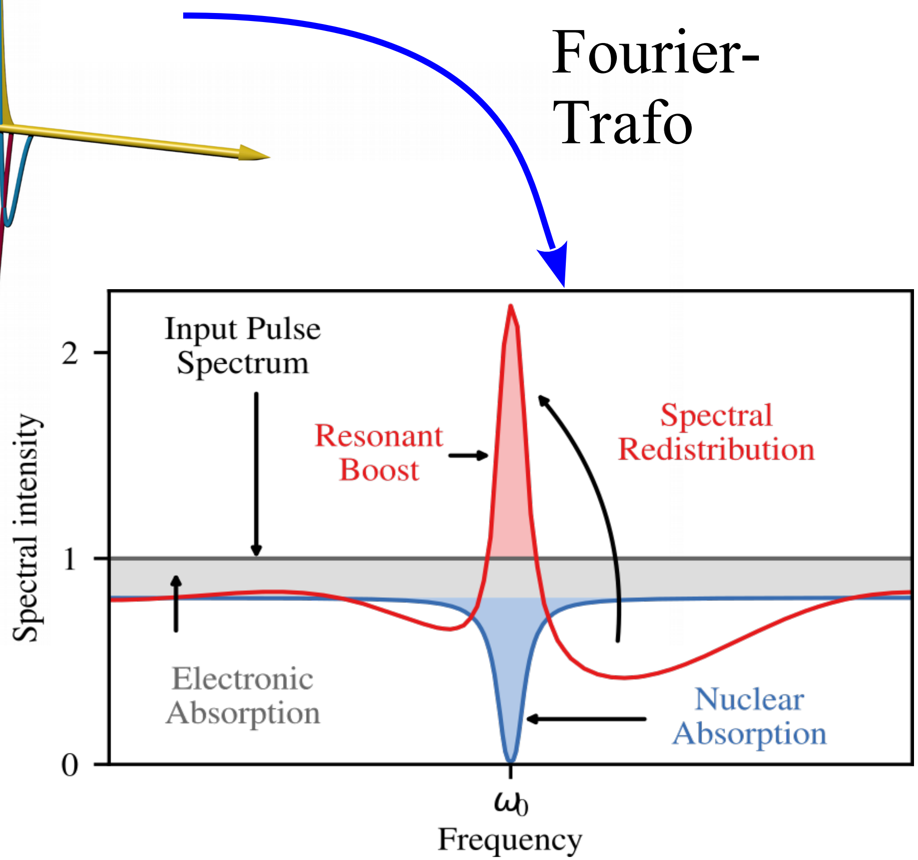
- identification of chemical, spin, and magnetic state
- relaxation phenomena
- coordination to neighbors
- value and direction of magnetic field



+ inelastic scattering: phonon density of states, dispersion relations, relaxation, ...

X-ray pulse shaping





Control time-dependent phase
of scattered x-ray pulse

Mechanical motion
simulates control laser field

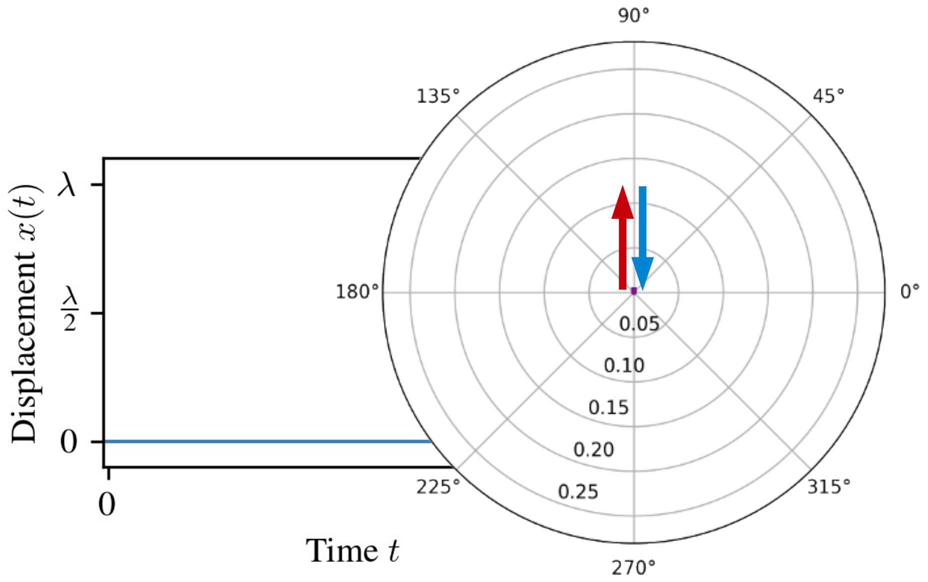
Demonstrate control with two special cases

- “Stimulated emission of excitons”

  Two pulses with opposite phase:

  First preparation pulse
  excites exciton

  Second control pulse
  de-excites target

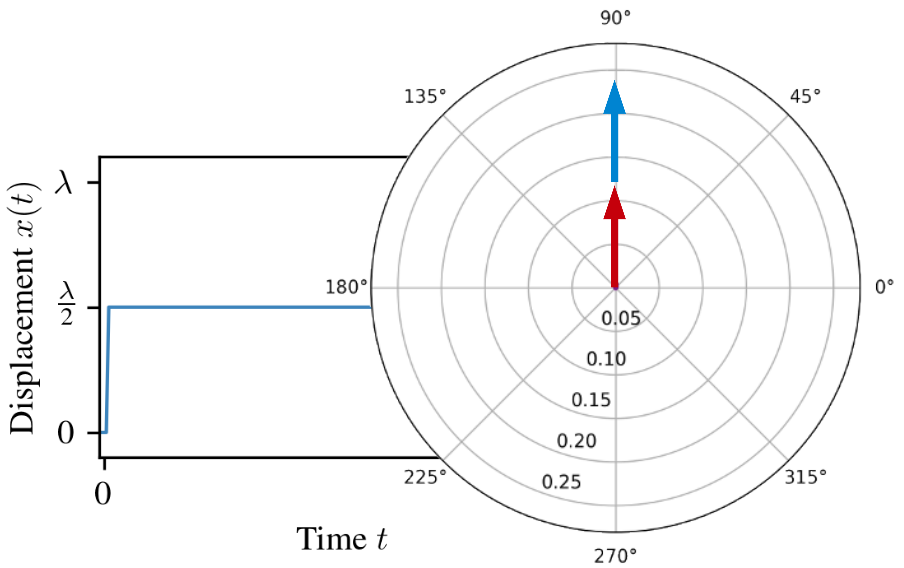


- “Coherent boost of excitation”

  Two pulses with same phase:

  First preparation pulse
  excites exciton

  Second control pulse
  further excites target

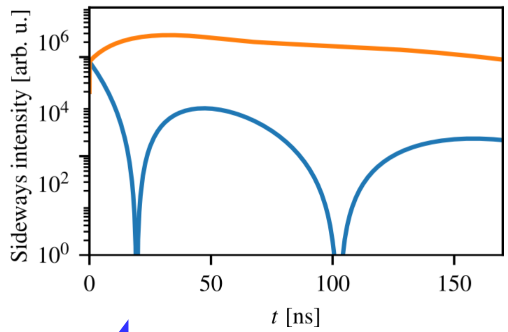


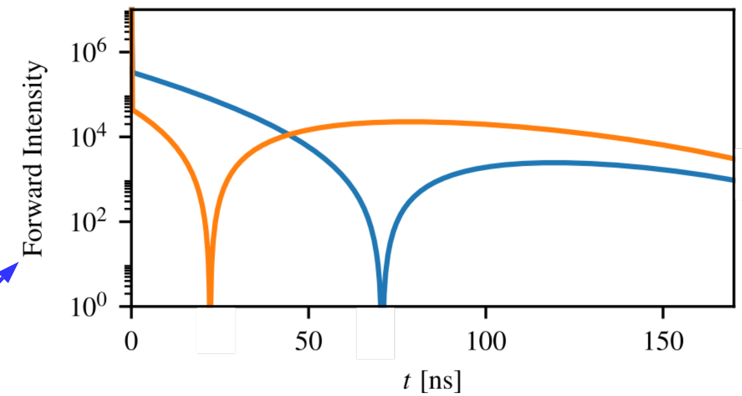
How to observe?

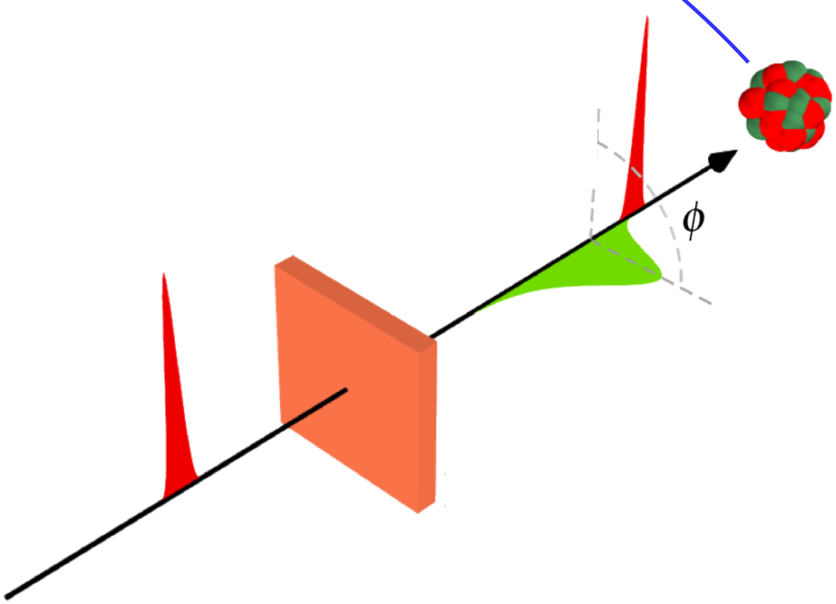
Stimulated emission
Excitation boost

Sideways:

- Only scattered light
- Clear signature
- Low count rate





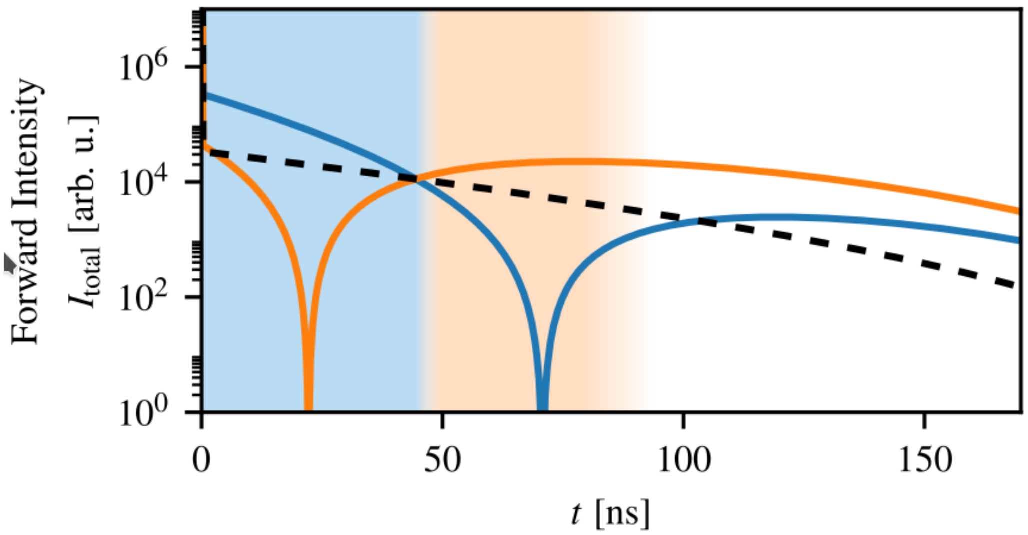


Forward direction:

- Interference with incident pulses
- Phase-sensitive
- High count rate
- Signature of dynamics?

Signature for spontaneous emission and boost

Theory model for single resonance in target



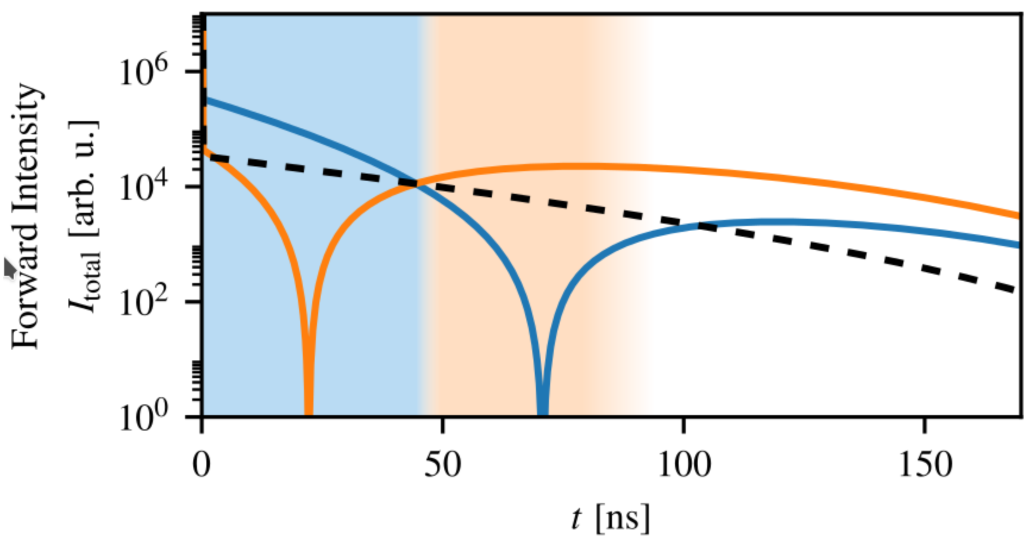
- Initially, higher intensity with SE, due to faster decay
- Afterwards, lower intensity with SE, since nuclei have already decayed

Stimulated emission

Excitation boost

Experimental results (ID 18, ESRF)

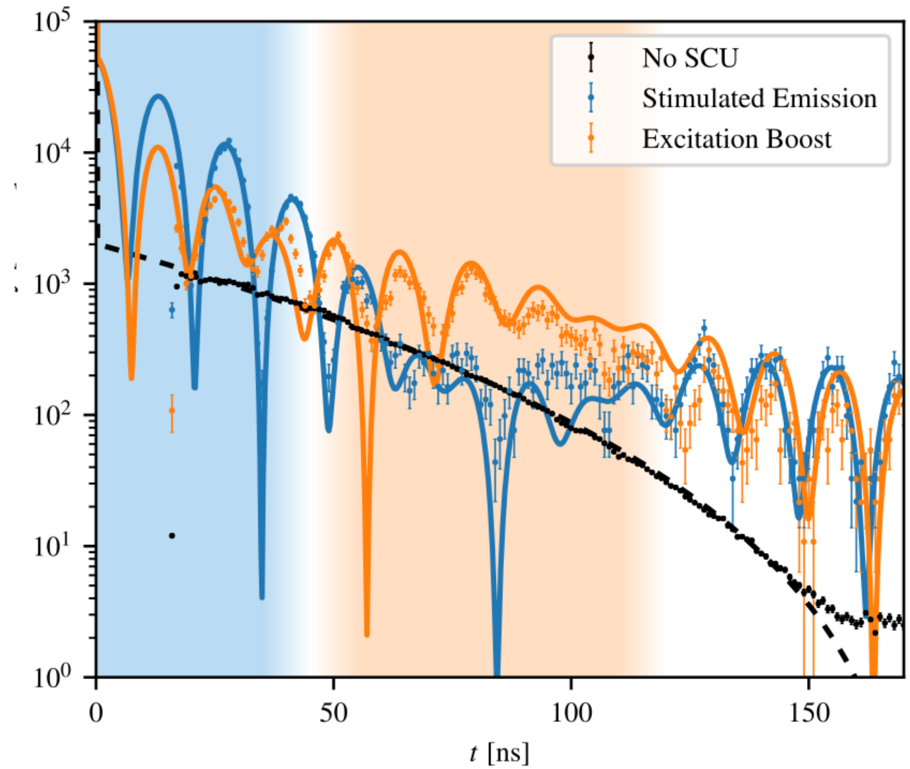
Theory model for single resonance in target



- Initially, higher intensity with SE, due to faster decay
- Afterwards, lower intensity with SE, since nuclei have already decayed

Stimulated emission
Excitation boost

Experimental results with fit



- Characteristic alternating intensities observed
- Fast oscillations due to additional beating with two resonances

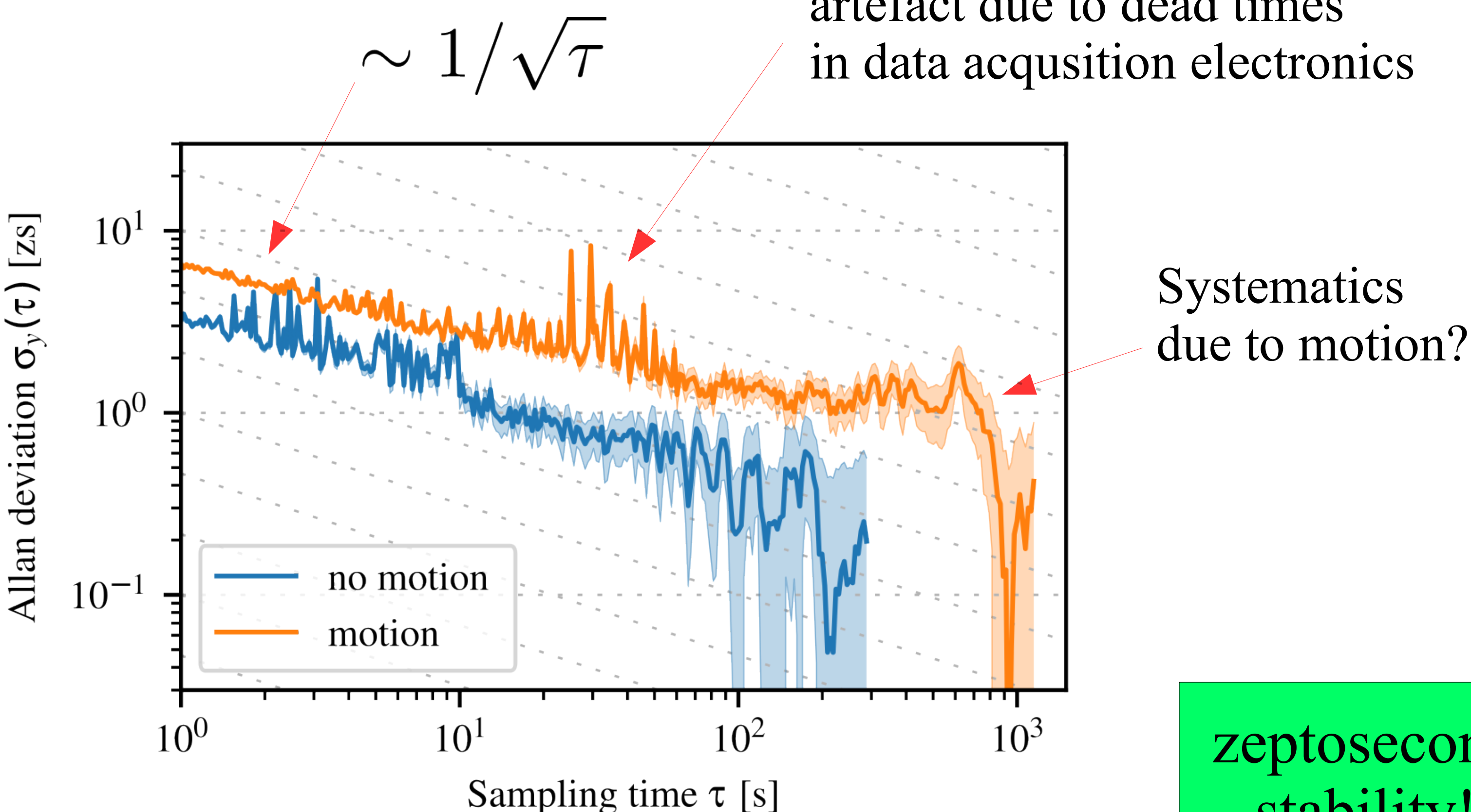
K. P. Heeg et al, submitted

How stable is the phase control?

Allan deviation: Measure for stability

- Split data into N equal parts of duration τ
- y_i is the phase measured in interval i
- RMS instability of two measurements τ apart

$$\sigma_y(\tau) = \left(\frac{1}{2(N-1)} \sum_{i=1}^{N-1} (y_{i+1} - y_i)^2 \right)^{\frac{1}{2}}$$



X-ray period 287 zs

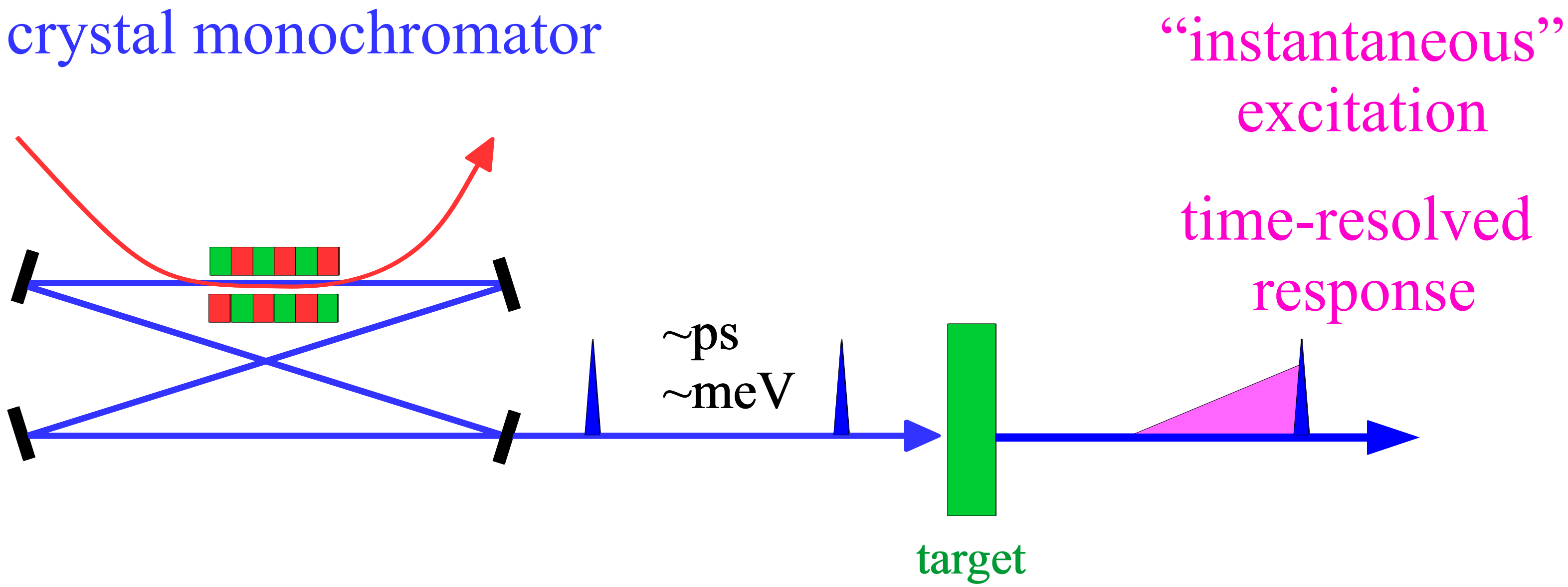
Measurement time ~ 45 min

zeptosecond stability!

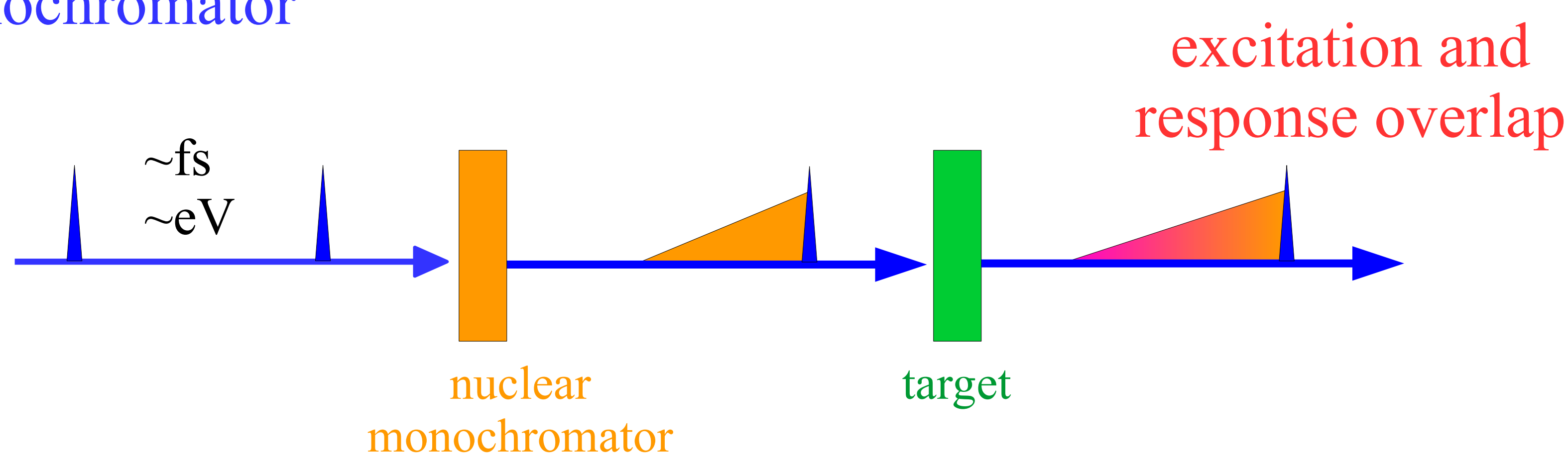
K. P. Heeg et al, submitted

Temporal pulse structure

XFELO / crystal monochromator



Nuclear monochromator



Can we enter the non-linear regime?

Synchrotron: $\dfrac{0.01 \text{ Photons @ 14.4keV}}{100\text{ps bunch} \times (\mu\text{m})^2 \times \Gamma} \Rightarrow I \sim 10^2 \dfrac{\text{W}}{\text{cm}^2}$

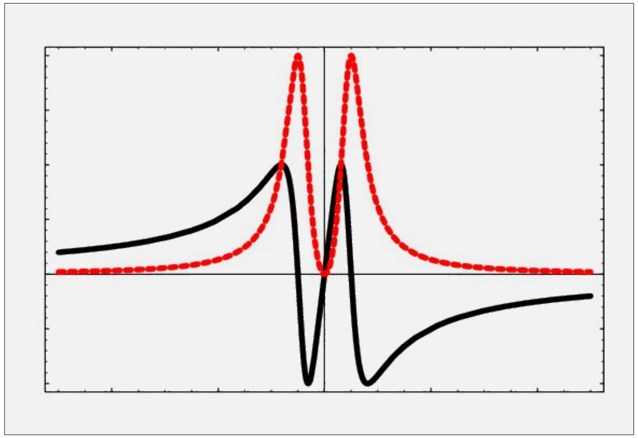
Seeded XFEL: $\dfrac{10^3 \text{ Photons @ 14.4keV}}{10\text{fs bunch} \times (\mu\text{m})^2 \times \Gamma} \Rightarrow I \sim 10^{10} \dfrac{\text{W}}{\text{cm}^2}$

EIT case: Kerr effect

$$n = n_0 + I_P n_2 \qquad \chi = \chi^{(1)} + 3 I_P \chi^{(3)}$$

$$\chi^{(3)} = 4.3 \times 10^{-22} m^2/V^2$$

$$\Rightarrow n_2 I_P \approx 10^{-7} \text{ for } 10^8 W/cm^2$$

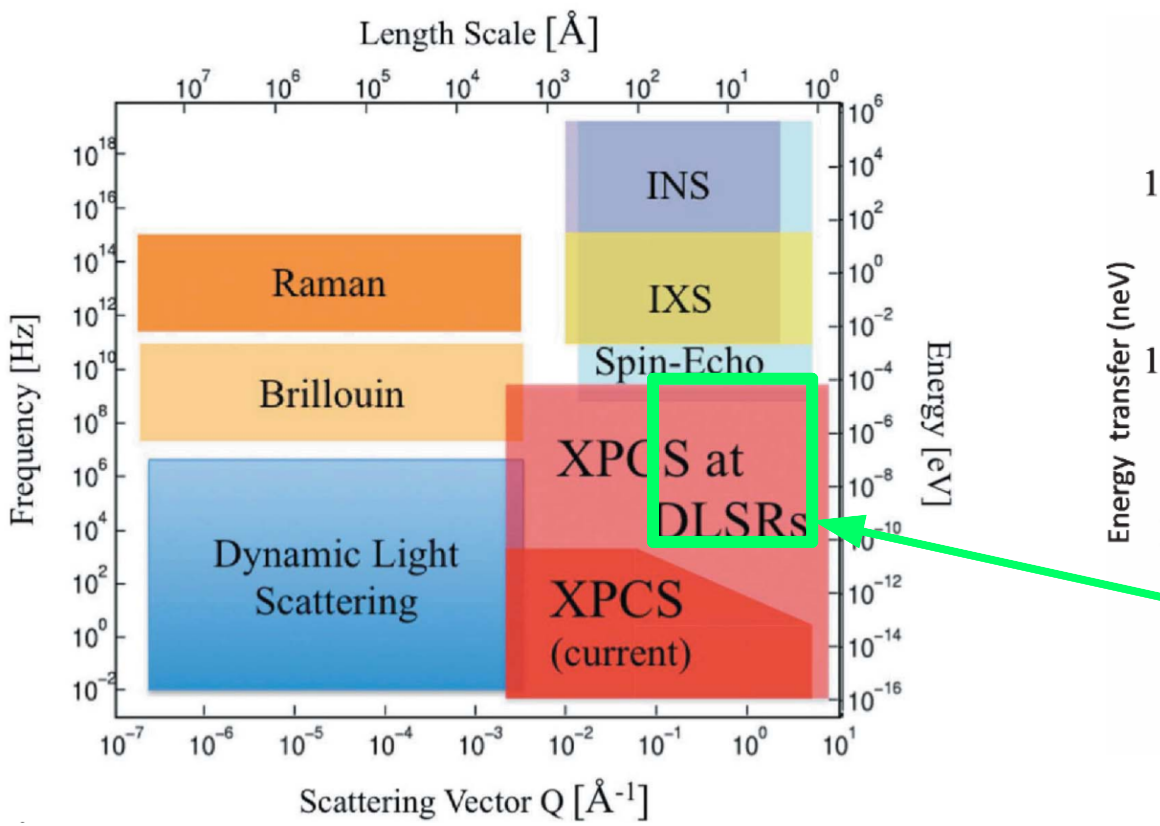


nonlinear phase shift ~ linear index achievable with seeded FEL
EIT: no linear absorption, strong enhancement via advanced schemes possible

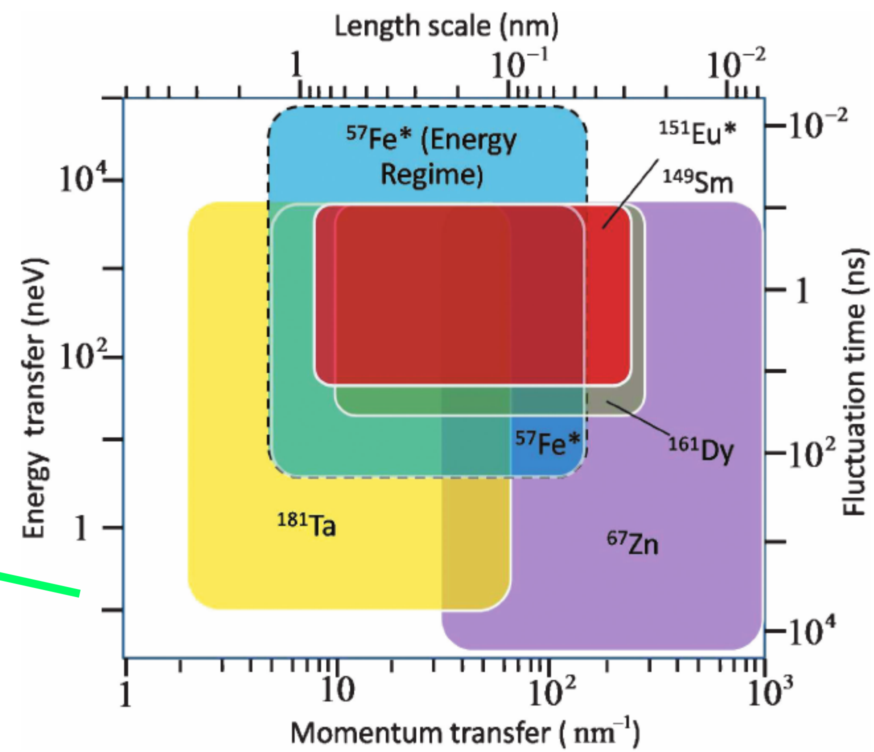
Röhlsberger et al, Nature 482, 199 (2012)

The temporal gap: nanoseconds to milliseconds

Time domain interferometry with nuclear resonances



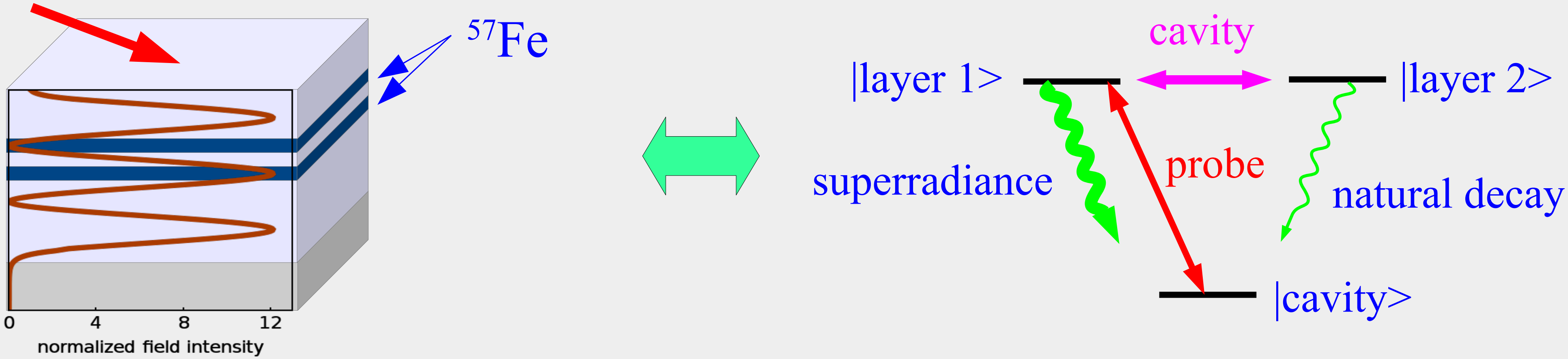
PETRA IV at DESY,
J. Synchrotron Rad. 25, 1277 (2018)



Saito et al., Appl. Phys. Express 2, 026502 (2009)

+ TDI proposed for applications at XFELs in SwissFEL science case

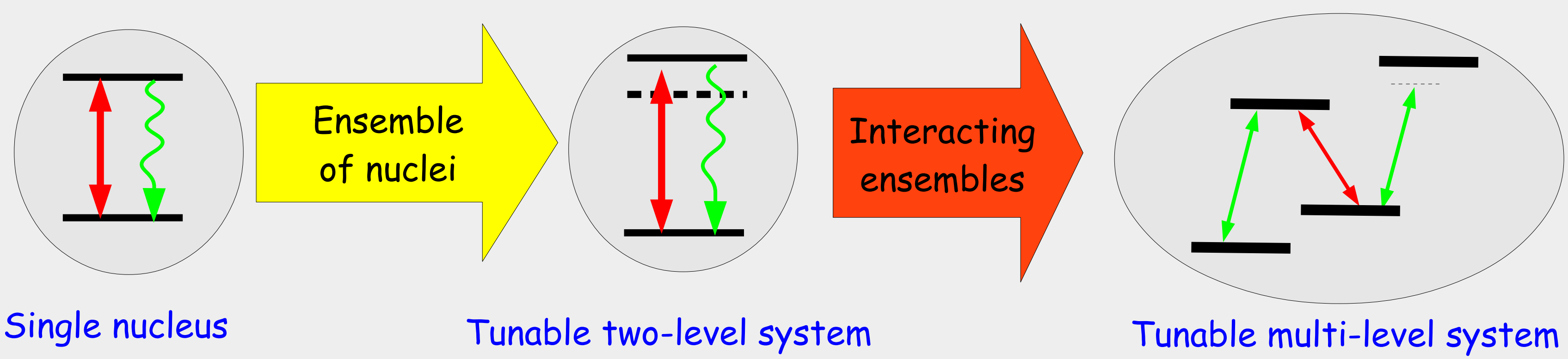
Design of artificial quantum systems



Röhlsberger et al, Nature 2012

→ create 3-level system otherwise not available
→ replace external control field by cavity
→ reduce docoherence via superradiance timescale

Next step: systematic approach to design level schemes



First steps: Longo, Keitel, Evers, Sci. Rep. 6, 23628 (2016)

→ "reverse engineering" of superradiance to determine required ensemble ensemble properties