Nuclear quantum optics at an XFEL

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Motivation

Light-matter interactions

electron shells

nucleus
Motivation

Light-matter interactions

- optical driving fields: excite/ionize outer electrons

electron shells

nucleus
Motivation

Light-matter interactions

▶ optical driving fields: excite/ionize outer electrons

▶ Higher frequencies/intensities: excite / ionize core electrons
Motivation

**Light-matter interactions**

- Optical driving fields: excite/ionize outer electrons
- Higher frequencies/intensities: excite / ionize core electrons
- Even higher frequencies/intensities: excite nucleus
Motivation

Light-matter interactions

- Optical driving fields: excite/ionize outer electrons
- Higher frequencies/intensities: excite / ionize core electrons
- Even higher frequencies/intensities: excite nucleus

These scenarios appear similar

But the methods and applications are quite different
Motivation

Light-matter interactions

different paradigms

uncontrolled pump + passive observation

full quantum control

- X-ray physics could greatly benefit from moving more towards quantum control
- What can be done is to large degree determined by availability of light sources
- New light sources and upgrades now and in near future → now is the right time
### What could be the benefit?

| Quantum                   | - Quantum-enhanced measurements, e.g. sub-\(\lambda\) resolution, squeezing  
|                          | - Foundations of quantum mechanics, e.g. entanglement of macroscopic objects  
| Nonlinear                | - Enhanced spectroscopy and measurements  
|                          | - Probe fragile targets  
|                          | - Combine different frequencies, e.g. resonant photon + x-ray for high position resolution  
| Control                  | - Enhanced sample preparation  
|                          | - Design material properties  
|                          | - Separate signal and background/noise  

So far rough ideas only – essentially unexplored field
Where is the catch?

Simple transfer optical → x-ray?

- Limitations of light sources (e.g., resonant intensity, temporal coherence, bandwidth, phase-locked and synchronized multi-color, ...)
- Limitations of instrumentation
- Boring from conceptional point of view

Implementation often not as easy as it may appear on paper

- Example: gamma ray laser (e.g., despite many attempts, population inversion remains a challenge)

Our approach: Explore starting from experimentally proven setups, do not simply copy from optical case
### Synchrotron radiation vs. seeded FEL beams

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<td>200ns (microbunch)</td>
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<td>Avg Flux (ph/s/Γ)</td>
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<td>2×10⁸</td>
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<tr>
<td>Fluence (ph/bunch/Γ)</td>
<td>10⁻²</td>
<td>6×10³</td>
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#### Nuclear parameters
- (for $^{57}$Fe)
- Energy: 14.4 keV
- Linewidth: 5 neV

#### XFEL parameters
- 10¹² photons/pulse
- Rel. BW: 6×10⁻⁵
- Rep. rate: 30kHz

Geloni et al, arXiv:1111.5766
Synchrotron radiation vs. seeded FEL beams

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<td>5×10^4</td>
<td>2×10^8</td>
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<td>10^-2</td>
<td>6×10^3</td>
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**nuclear parameters** (for $^{57}$Fe)
- energy 14.4 keV
- linewidth 5 neV

**XFEL parameters**
- 10^{12} photons/pulse
- rel. BW $6×10^{-5}$
- rep. rate 30kHz

**Two directions**
- photon hungry ("proven concepts with higher count rate")
- Short, nonlinear, coherent ("new ideas")

Geloni et al, arXiv:1111.5766
Content

Introduction / NFS

Quantum optics and information

Nuclear quantum optics

Future perspectives
Content

Introduction / NFS

Quantum optics and information

Nuclear quantum optics

Future perspectives
Nuclear resonance scattering

DESY Hamburg

storage ring

monochromator

nuclear sample

Inelastic scattering

cohherent scattering

undulator

synchrotron radiation

source + drive

detector

shield

target

Student lab Uni Mailand

DESY Hamburg
Nuclear resonance scattering

- Tool to investigate magnetic, structural and dynamic properties of matter
- Small linewidth of nuclear resonances (μeV-peV) is both essential feature and technical challenge
- Mößbauer effect leads to recoilless absorption and emission

some isotopes < 30keV

Iron is of significance in biology, earth science, ...

“Working horse” of nuclear resonance scattering

\[ Q \sim \omega_0 / \Gamma \sim 10^{12} \]
Separating signal and background

Nuclear resonances very narrow (μeV-peV)
Nuclear scattering has delayed tail on time scale $1/\Gamma$
Example ($^{57}\text{Fe}$): 141 ns
Time-gating $\rightarrow$ almost background-free
Alternative methods available $\rightarrow$ later
Cooperative light scattering

incident light

large dilute cloud

scattered light

quantum particles as scatterers

$\vec{k} \quad R \gg \lambda$

no recoil

stationary particles
Elementary processes

incident light

\[ \vec{k} \]

Intermediate excitonic state

\[ |\Psi\rangle = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} e^{i \vec{k} \vec{r}_i} |g_1, \ldots, g_{i-1}, e_i, g_{i+1}, \ldots, g_N\rangle \]
Directionality of coherent scattering

- Coherent scattering occurs in forward direction
- Similarity to multi-slit / grid diffraction but constructive interference only in forward / Bragg direction

\[ R \gg \lambda \]

\[
\lim_{N \to \infty} \sum_{i=1}^{N} e^{i(\vec{k} - \vec{k}_L) \vec{r}_i} \sim \delta(\vec{k} - \vec{k}_L)
\]

grid = CD-R grooves
Temporal beats

\[ I_e = 3/2 \]

\[ \Delta m = 0 \]

\[ I_g = 1/2 \]

σ-polarized

“broadband” excitation

\[ {^{57}}\text{Fe sample} \]
Temporal beats

Scattering on two transitions with same dipole moment, but different transition frequencies

Expect beats in the time-dependent intensity
Multiple scattering

As a model, separate sample into thin layers

Due to forward scattering, first layer is driven only by incident field

Layer n > 1 is in addition driven by “upstream” layers, causing phase shifts

Initial phase synchronization due to incident pulse is dephased

Alternative view: synchrotron excitation does not correspond to radiation eigenmode of the sample → later

Superradiance

Dicke case (small dense sample)

\[ |\Psi\rangle = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} |g_1, \ldots, g_{i-1}, e_i, g_{i+1}, \ldots, g_N\rangle \]

\[ \langle G | \bar{d} | \Psi \rangle = \sqrt{N} \langle g_i | \bar{d} | e_i \rangle \]

\[ \gamma \rightarrow N\gamma \]

NFS case (large dilute sample)

\[ |\Psi\rangle = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} e^{i\vec{k} \cdot \vec{r}_i} |g_1, \ldots, g_{i-1}, e_i, g_{i+1}, \ldots, g_N\rangle \]

Superradiant state dynamically coupled to subradiant states

Imperfect preparation of superradiant state in thick samples → dephasing

Characteristic features in forward scattering

- Exciton
- Superradiance: $\gamma \rightarrow N\gamma$
- Beats
- Multiple scattering

![Graph showing signal intensity over time](Image)
Characteristic features in forward scattering

- Exciton
- Superradiance: $\gamma \rightarrow N\gamma$
- Beats
- Multiple scattering

All of these features are also intensely studied in quantum optics with atoms/visible light.
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Electromagnetically induced transparency
Optical response of a single resonance

\[ \Delta_{\text{probe}} \]

\[ \Omega_{\text{probe}} \]

\[ \Delta_{\text{probe}} \]
Electromagnetically induced transparency

Three-level $\Lambda$ system

Medium is rendered transparent by shining light on it!

EIT is an archetype quantum optical effect with a multitude of applications

Electromagnetically induced transparency

Interpretation as coherence/interference effect:

If EIT conditions are satisfied:

- Laser fields drive atom to coherent superposition of \( |a\rangle \) and \( |b\rangle \)
- Interference: amplitudes for \( |a\rangle \rightarrow |c\rangle \) and \( |b\rangle \rightarrow |c\rangle \) cancel

no excitation of the atom due to destructive interference
Key application: Slow light

- Linear dispersion with high slope
- Low absorption

Can modify group velocity of light

What is a light pulse?
What is a light pulse?
What is a light pulse?
What is a light pulse?

Destructive interference

Constructive interference
Motion in vacuum

All wavelengths have the same speed
All wavelengths have the same speed.
Motion in vacuum

All wavelengths have the same speed
All wavelengths have the same speed
Motion in dispersive medium

Different wavelengths have different speeds
First experiment

Experiment by Lene Hau (Harvard)

- pulse without atoms
  - $T = 450 \text{ nK}$
  - $\tau_{\text{delay}} = 7.05 \pm 0.05 \mu\text{s}$
  - $L = 229 \pm 3 \mu\text{m}$
  - $\nu_g = 32.5 \pm 0.5 \text{ m s}^{-1}$

- pulse with atoms
  - delay
  - time

Image: Diagram of the experiment setup with labels for components such as Detektor, control, Laser, and CCDs.
What would be desirable?

- Broad transparency window to propagate short input pulses
- Steep dispersion slope for strong effect on propagated pulse
- \((\text{time delay}) \cdot (\text{transparency bandwidth})\) is constant → need to tune for best trade-off
- More general level schemes offer wide range of applications
- Example: Strongly enhanced non-linear response
Nonlinear effects enhanced by EIT

- Destructive interference in 1st order susceptibility i.e. low absorption
- Constructive interference in 3rd order susceptibility
- Strong non-linearities possible down to single photon level
- Coherence is the key to these enhancements

Giant Kerr nonlinearity

Four-wave mixing

Example: Frequency conversion

- Coherence greatly enhances conversion efficiency
- Extra coherence modifies source term in propagation equation
- Interpretation: stringent phase matching conditions are alleviated

Consider a single photon:

- Focus on polarization, neglect other degrees of freedom (momentum, angular momentum, frequency, ...)
- The polarization space has the basis $|H\rangle$, $|V\rangle$
- A general state is given by a linear superposition:

$$|\Psi\rangle = \alpha|H\rangle + \beta|V\rangle$$
Entanglement

A basis for the two photon space is given by the product space

\[ |HH\rangle = |H_A\rangle |H_B\rangle, \quad |VH\rangle = |V_A\rangle |H_B\rangle, \]

\[ |HV\rangle = |H_A\rangle |V_B\rangle, \quad |VV\rangle = |V_A\rangle |V_B\rangle \]

All of these basis states are of the form

(state of photon 1) * (state of photon 2)

But due to the superposition principle, there are states which cannot be written as such a product. For example:

\[ |\Psi\rangle = \alpha |HH\rangle + \beta |VV\rangle \]
Entanglement of pure states

Consider two quantum objects A and B with basis

\[ \{ |i\rangle_A | i = 1, 2, \ldots \} \quad \text{and} \quad \{ |j\rangle_B | j = 1, 2, \ldots \} \]

The most general state is a superposition of all product states:

\[ |\Psi\rangle = \sum_{i,j} c_{i,j} |i\rangle_A |j\rangle_B \]

This state is called a **separable state** iff it can be written as

\[ |\Psi\rangle = \left( \sum_i c_i^A |i\rangle_A \right) \left( \sum_j c_j^B |j\rangle_B \right) \]

If not, then the state is an **entangled state**
Examples

\[ |\Psi\rangle = |HV\rangle + |VV\rangle = (|H\rangle_A + |V\rangle_A)|V\rangle_B \]

\[ |\Psi\rangle = |HH\rangle - |VH\rangle - |HV\rangle + |VV\rangle = (|H\rangle_A - |V\rangle_A)(|H\rangle_B - |V\rangle_B) \]

\[ |\Psi\rangle = |HV\rangle + |VH\rangle \]

\[ |\Psi\rangle = |HH\rangle + |VV\rangle \]

\[ |\Psi\rangle = \sqrt{2}|HH\rangle - 2|VH\rangle + |HV\rangle - \sqrt{2}|VV\rangle \]

Separable – Entangled - ????
Measurements on entangled states

Suppose the two photons are in the entangled state

$$\left| \Psi \right\rangle = \left| H V \right\rangle + \left| V H \right\rangle$$

First imagine a measurement on photon A. The outcome would be random: 50% probability \( |H\rangle \), 50% probability \( |V\rangle \)

Next imagine a measurement on photon B. The outcome would be random: 50% probability \( |H\rangle \), 50% probability \( |V\rangle \)

But if the measurement on A is actually performed:
   If measured state of A is \( |H\rangle \), then photon B with certainty is in \( |V\rangle \)
   If measured state of A is \( |V\rangle \), then photon B with certainty is in \( |H\rangle \)

The two photons are correlated, and a (local) measurement on A changed the state on B, independent of the separation of A and B (→ EPR argument)
Mode (single photon) entanglement

- Single photon impinging on 50/50 beam splitter gives output
  \[ |\Psi\rangle = \frac{1}{\sqrt{2}} (|0\rangle_A |1\rangle_B + |1\rangle_A |0\rangle_B) \]

- The single photon entangles the two field modes A and B - the photon itself is not entangled

- Applications like Bell violation, teleportation etc. have been proposed

- Can be converted to other forms, e.g. “regular” entanglement between atoms
  \[ |\Psi\rangle = \frac{1}{\sqrt{2}} (|g_1 e_2\rangle + |e_1 g_2\rangle) \]
Why bother about entanglement?

spooky action at a distance
In what context is entanglement considered?

Entanglement is one defining element of QM, and not an exception! But in many cases, effects are hidden (e.g. by decoherence).

Foundations of physics:

- The relation of the classical and the quantum world
- Is quantum mechanics a real/complete/meaningful theory?
- What are the ultimate limits for preparation, measurement, control?

Applications:

- Essential ressource for all quantum information/communication protocols
- Measure and structure beyond “classical” or standard quantum limit
Foundations: The EPR argument


- If a measurement outcome can be predicted with certainty without perturbing the state, then it corresponds to an element of reality (fulfilled by classical observables like mass)

- A complete theory should account for all elements of reality (fulfilled, e.g., by electrodynamics)

- Two subsystems can be separated such that a measurement on one does not immediately change the second (locality assumption)

- Thought experiment => position and momentum of a particle are both elements of reality

- Since QM does not allow to know both simultaneously, there is a contradiction. EPR concluded: QM is incomplete
Bell inequalities

- Bell started from EPR assumptions (reality and locality)

- These properties are seen as cornerstones of a “classical theory”

- Result: If a theory is (real and local), then correlations between measurements of non-commuting observables obey certain inequalities, the Bell inequalities

- Quantum mechanics violates these inequalities in theory and experiment: Thus either realism and/or localism has to be abandoned
Loopholes

Problem:

- So far, no unambiguous experiment on Bell inequalities, due to experimental problems

- Example: Detection efficiency
  Need detection efficiency near 100%, difficult with optical photons

- Example: Communication
  No speed-of-light communication may be possible during measurement time (optical experiments: > 100km separation)

How might x-rays help?

- Perform experiments with other loopholes?

- Close some loopholes, e.g., via high photon detection efficiency?

- New experimental approaches?
Quantum-assisted measurements

Consider interferometer

- The sample induces phase shift in one of the arms
- The phase shift leads to a pattern on the screen
- The two arms are entangled!

\[
\text{input} \quad |1\rangle \quad \rightarrow \quad |1\rangle_A |0\rangle_B + |0\rangle_A |1\rangle_B \\
\text{sample} \quad \rightarrow \quad |1\rangle_A |0\rangle_B e^{i\phi} + |0\rangle_A |1\rangle_B
\]
N00N states

\[ |N\rangle_A |0\rangle_B + |0\rangle_A |N\rangle_B \]

\[ \rightarrow |N\rangle_A |0\rangle_B e^{iN\phi} + |0\rangle_A |N\rangle_B \]

- The N00N state leads to a phase shift multiplied by N
- This leads to a N-fold enhancement of the resolution
- The N00N state is a highly non-classical, entangled state
- \( |2002\rangle \) can be produced by Hong-Ou-Mandel effect
How can entanglement be created?
Down conversion

- High-frequency photon split in two photons with different polarization, emitted in two cones
- In directions where cones intersect, entangled photon pair
- Not deterministic, high vacuum contribution, inefficient
Coherent control

- Deterministic, but need strong coupling ("Pi pulses")
- Other related implementations (Quantum dots, ...)
- Coherent control approach for entanglement generation in NFS

Haroche et al.

Rempe et al.
Introduction / NFS

Quantum optics and information

Nuclear quantum optics

Future perspectives
Magnetic switching

The level structure depends on applied magnetic field: Zeeman splitting

In certain crystals (e.g. FeBO$_3$), the hyperfine field is very strong ($\sim 30$ T), and can be aligned via weak external fields (few Gauss)

This allows to switch the direction of a very strong effective magnetic field in few ns in the lab

---

**Storage of Nuclear Excitation Energy through Magnetic Switching**


1II. Institut für Experimentalphysik, Universität Hamburg, D-22761 Hamburg, Germany
2Physik-Department E15, Technische Universität München, D-85748 Garching, Germany
3RRC, “Kurchatov Institute”, SU-1123182 Moscow, Russia

(Received 8 May 1996)

The decay rate of $^{57}$Fe nuclei in an $^{57}$FeBO$_3$ crystal excited by 14.4 keV synchrotron radiation pulses was controlled by switching the direction of the crystal magnetization. Abrupt switching some nanoseconds after excitation suppresses the coherent nuclear decay. Switching back at later times restores it, starting with an intense radiation spike. The enhanced delayed reemission is due to the release of the energy stored during the period of suppression. Suppression and restoration originate from drastic changes of the nuclear states and of the interference within the nuclear transitions.

HASYLAB F4 beam line

Two “ingredients”

\[ e_0 \]

\[ \sigma\text{-polarized} \]

\[ \vec{B} \]

\[ ^{57}\text{Fe sample} \]

\[ I_e = 3/2 \]

\[ -3/2 \]

\[ -1/2 \]

\[ +1/2 \]

\[ +3/2 \]

\[ \Delta m = 0 \]

\[ I_g = 1/2 \]

\[ +1/2 \]

\[ -1/2 \]

\[ m_e \]

\[ m_g \]

coherence

\[ |a\rangle \rightarrow |b\rangle \]

\[ |c\rangle \]
Coherent control of the exciton

Excite the sample

Rotate quantization axis

- Rotate applied magnetic field
- Experiment: 30T in 5ns possible in certain crystals

Deexcitation

- Destructive interference of all pathways possible
- Analogy to electromagnetically induced transparency
Exciton storage

Experimental verification:

- Control of coherent NFS possible
- The coherent decay is (almost) fully suppressed after switching
- Revival of coherent decay after switching back
- Primary limitation: incoherent decay with natural lifetime

X-ray entanglement generation
keV single photon entanglement

Motivation

- Build up on experimentally demonstrated technique of nuclear switching
- Establish coherent control of x-rays on the single photon level
- First step towards nonlinear and quantum x-ray science
- High photon momentum: x-ray optomechanics, entanglement with more macroscopic objects
- More general: New parameter ranges, more complex quantum systems, more robust photons, less thermal background noise
Advanced magnetic switching schemes

Rotation angle

- Determines new quantization axis and superposition states

Timing

- Important due to different transition energies
- Determine whether constructive/destructive interference occurs
- Example: Suppression at $t_1$, how does $t_2$ affect further evolution?

Transition amplitudes

- Linear: $\left\{ \frac{1}{2} \to \frac{1}{2}, -\frac{1}{2} \to -\frac{1}{2} \right\}$
- Circular: $\left\{ -\frac{1}{2} \to \frac{1}{2}, \frac{1}{2} \to -\frac{1}{2} \right\}$
- Circular: $\left\{ \frac{3}{2} \to \frac{1}{2}, -\frac{3}{2} \to -\frac{1}{2} \right\}$

Step 1: Synchrotron excitation

Initially, magnetic field is in $z$ direction.
Step 2: Canceling coherent decay

- Initially, magnetic field is in $z$ direction
- At time $t_1$, cancel decay by rotating into $y$ direction

**Graph:**
- Initial magnetic field $\vec{B}$
- Graph showing decay over time $t$ with $I$ in arbitrary units
- Two graphs comparing 'no switching' and 'switching' scenarios

**Diagram:**
- Axes: $x$, $y$, $z$
- Magnetic field $\vec{B}$ pointing along $z$ axis
- Time $t$ in nanoseconds
- Decay intensity $I$ on a logarithmic scale
Step 3: Releasing circular polarization

- Initially, magnetic field is in $z$ direction

- At time $t_1$, cancel decay by rotating into $y$ direction

- At time $t_2$, enable decay on $\Delta m = \pm 1$
  but continue to suppress $\Delta m = 0$

\[
\begin{align*}
\left\{ \frac{1}{2} \rightarrow \frac{1}{2}, \quad -\frac{1}{2} \rightarrow -\frac{1}{2} \right\} \\
\left\{ -\frac{1}{2} \rightarrow \frac{1}{2}, \quad \frac{1}{2} \rightarrow -\frac{1}{2} \right\} \\
\left\{ \frac{3}{2} \rightarrow \frac{1}{2}, \quad -\frac{3}{2} \rightarrow -\frac{1}{2} \right\}
\end{align*}
\]
Step 4: Canceling coherent decay

- Initially, magnetic field is in z direction.
- At time $t_1$, cancel decay by rotating into y direction.
- At time $t_2$, enable decay on $\Delta m = \pm 1$ but continue to suppress $\Delta m = 0$.
- At time $t_3$, cancel decay by rotating into y direction.
Step 5: Releasing linear polarization

- Initially, magnetic field is in \( z \) direction
- At time \( t_1 \), cancel decay by rotating into \( y \) direction
- At time \( t_2 \), enable decay on \( \Delta m = \pm 1 \) but continue to suppress \( \Delta m = 0 \)
- At time \( t_3 \), cancel decay by rotating into \( y \) direction
- At time \( t_4 \), enable decay on \( \Delta m = 0 \)
Temporal mode entanglement

Design advanced coherent control scheme:

- Coherently control exciton decay such that single excitation is distributed into three pulses
- Neglecting the background, the two signal pulses are time bin entangled
- Can extract signal from background and convert it to spatial mode entanglement using x-ray optics

\[ |\Psi\rangle = \alpha |0\rangle_A |1\rangle_B + \beta |1\rangle_A |0\rangle_B \]
How to extract signal pulse?

- Problem: One part of signal has same polarization as background pulse.

- Time gating not useful if following setup should be protected from high-intensity background; lighthouse effect difficult because of precise timing of nuclear switching.

- PSM: Piezo electric steering mirror or sub-ns control device based on crystal lattice deformation.\(^1\)

- Have about 180 ns “steering time” because of magnetic switching.

---

Proof-of-principle experiment

- Do not extract signal, use time gating to remove background
- Switching → two entangled overlapping pulses with opposite polarization
- Correlation measurement with interferometer, violate Bell-like inequality*)
- Need to eliminate “which-way”-information hidden in polarization
- “loophole”: explanation of results also possible by non-local classical theory

X-ray branching ratio control

\[
\frac{\gamma_1}{\gamma_2} = ?
\]
Motivation

- Prepare specific initial state
  - single magnetic ground-state sub-level
  - metastable excited state (isomer)

- Release excited state on demand

- Modify/control chain of decays, e.g., for preparation of specific states/isotopes

- Assist the control over quantum dynamics in more advanced setups
Branching ratio

Single particle branching ratio:
- Determines ratio of spontaneous emission channels
- Property of the particle only

Branching ratio in ensembles
- Have cooperative modification of excitation and decay
- Determined by particle, ensemble and excitation properties, varies with time
- Need to define cooperative branching ratio
Motivation

- Aim: Efficiently pump from ground state $|G\rangle$ to isomeric state $|I\rangle$
- Cooperativity leads to enhanced excitation to $|E\rangle$, but also to fast decay
- In effect, little transfer to $|I\rangle$

Idea:

- Suppress cooperative emission
- Then cooperativity leads to enhanced excitation, but decay proceeds with single particle branching ratio
- In effect, enhanced pumping to $|I\rangle$

The ideal case

- Assume purely superradiant decay with rate $\xi \cdot \gamma$
- Assume perfect coherent control of cooperative decay

Result:

$$\frac{b_c^C}{b_c^{NC}} = \xi + 1$$

- Cooperative branching ratio is larger by factor $\xi + 1$
- In addition, cooperative enhancement of excitation
How to control?

**Magnetic switching:**

- Turn off cooperative decay by interference
- The incoherent decay with single-particle branching ratio remains

**Destroy phase coherence:**

- Use short pulse of incoherent light, spatially inhomogeneous magnetic field, or similar to destroy spatial coherence
- Without the coherence, uncorrelated decay without cooperative enhancement
- Can be done immediately after excitation, does not require sophisticated pulse control
The magnetic switching case

superradiant decay to initial state

population of sub-radiant states levels off decay to initial state → limit to enhancement

Switching improves result, but significant decay before trapping can be achieved → better results with phase destruction

The magnetic switching case

- Branching ratio time dependent as expected
- Cooperative branching ratio smaller than single-particle ratio due to superradiance
- After switching, single-particle branching ratio is achieved
- With destruction of phase coherence, single-particle ratio can immediately be achieved

Radiative eigenmodes
Temporal structure of scattered light

Where is the difference? Does it spoil the analogy?
Microscopic analysis of light scattering

- Multiple scattering in thicker samples
- All atoms are radiatively coupled
Radiative eigenmodes

Single excitation

\[ |\psi(t)\rangle = \sum_j \beta_j(t) |e_j, 0\rangle + \sum_k \eta_k(t) |G, 1_k\rangle \]

Equations of motion

\[ \dot{\beta}_j(t) = -\frac{\Gamma_0}{2} \beta_j(t) - \sum_{j' \neq j} \frac{\Gamma^{(j)}_{j'}}{2} \beta_{j'}(t) \]

Diagonalization of these equations leads to radiative eigenmodes

Radiative eigenmodes

Decompose initial states in radiative eigenmodes

$$|\psi(0)\rangle = \sum_{n=1}^{N} C_n |\nu^{(n)}\rangle$$

what the x-rays prepare

single eigenmode

Time evolution

$$|\psi(t)\rangle = \sum_{n} C_n e^{-\lambda_n t} |\nu^{(n)}\rangle$$

eigenvalue

Each radiative mode decays exponentially with specific frequency shift and decay rate

Dominant eigenmode vs. ensemble size

- **Small volume**: One dominating eigenvalue, strong exponential superradiance
- **Intermediate volume**: Several equivalent modes
- **Large volume**: Many competing modes, complicated temporal structure (sub/superradiant mixed)

Selective excitation of radiative modes

“Standard” x-ray scattering with thick target

- Incident pulse significantly excites many different radiative eigenmodes
- Complicated temporal structure inevitable

X-ray scattering with structured targets

- In Bragg geometry, a single radiative eigenmode is excited

\[ |T_{k_f} \rangle = \frac{1}{\sqrt{N}} \sum_j e^{ik_f \cdot r_j} |e_j, 0 \rangle \]

\( ^{57}\text{Fe sample} \)

Nuclei

Hannon/Trammell review, Evers et al in preparation
Exciting single radiative eigenmodes

Bragg geometry

X-ray thin film cavities effectively realize Bragg case

Flexible design ⇒ more possibilities

geometry crucial

Evers et al in preparation
Thin film x-ray cavities
Thin film x-ray cavities

- nm-sized thin film cavity
- Cavity resonances give field enhancement
- Nuclear resonances in Fe can interact with cavity field, observable in reflection

![Diagram of thin film x-ray cavity](image)

Movie 1

Movie 2

Reflection vs. angle graph

Field intensity graph
A single iron layer

Effectively acts as a two level system!
Cooperative effects

But with modified properties: superradiance + cooperative Lamb shift
Tailoring the light-matter interaction

Iron nuclei strongly couple to cavity field
- Accelerated decay
- High excitation probability

Iron nuclei weakly couple to cavity field
- Decelerated decay
- Low excitation probability

Effective properties of the nuclei can be tailored!
Two iron layers

Reflection spectrum

Looks like EIT!
Two iron layers

$^{57}\text{Fe}$

Reflection spectrum

It is EIT!

same nuclei acquire different properties

$|\text{cavity}\rangle$

$|\text{layer 1}\rangle$

superradiantly enhanced

cavity coupling

$|\text{layer 2}\rangle$

probe

natural decay

What would be desirable?

- Broad transparency window to propagate short input pulses.
- Steep dispersion slope for strong effect on propagated pulse.
- \((\text{time delay}) \cdot (\text{transparency bandwidth})\) is constant → need to tune for best trade-off.

- More general level schemes offer wide range of applications.
- Example: Strongly enhanced non-linear response.

J. Evers, work in progress
General quantum optical theory
Exploit the hyperfine structure

- So far, operated nuclei as 2-level systems
- Next, apply magnetic field to exploit magnetic hyperfine structure
- Many degrees of freedom: polarization, magnetization

Find quantum optical model to interpret results, and to include nonlinear/quantum effects
Constructing a quantum optical model

Find level scheme and set up master equation

\[
\frac{d}{dt} \rho = -\frac{i}{\hbar} [H, \rho] + \mathcal{L}[\rho]
\]

density matrix
coherent evolution
incoherent evolution

Kiffner, Macovei, Evers, Keitel, Progress in Optics 55, 85 (2010)

Use input-output formalism to calculate cavity response

\[
R = \frac{\langle a_{out} \rangle}{\langle a_{in} \rangle}
\]

reflectance

Constructing a quantum optical model

Limit of linear nuclear response to classical field:

Nuclear scattering formalism

reflectivity

\[ R \sim \frac{1}{\Delta + \frac{i}{2} \gamma - iC \cdot pq} \]

Quantum optics formalism

susceptibility

\[ \chi \sim \frac{1}{\Delta + \frac{i}{2} N \cdot \gamma + \Delta_{LS}} \]

analytical equivalence
Towards a quantum optical model

Find level scheme, field configuration and master equation such that

We have applied this approach for the general case with hyperfine splitting and arbitrary input and output polarizations and material magnetization

A single quantum optical model to rule it all!
Unexpected spectral signatures

What's this? Only interference can create zeros in overlapping resonances. But can't be EIT – only one layer!

K. P. Heeg, R. Röhlsberger, J. Evers, in preparation
Quantum optical model: Master equation

Two-level system

$$\frac{\partial}{\partial t} \rho = -\frac{\gamma}{2} \left( |e\rangle \langle e| \rho + \rho |e\rangle \langle e| - 2 |g\rangle \langle e| \rho |e\rangle \langle g| \right)$$

$$\frac{\partial}{\partial t} \rho_{ee} = -\gamma \rho_{ee} \quad \frac{\partial}{\partial t} \rho_{eg} = -\frac{\gamma}{2} \rho_{eg}$$

Three-level system

$$\frac{\partial}{\partial t} \rho = -\frac{\gamma}{2} \left( |1\rangle \langle 1| \rho + \rho |1\rangle \langle 1| - 2 |g\rangle \langle 1| \rho |1\rangle \langle g| \right) - \frac{\gamma}{2} \left( |2\rangle \langle 2| \rho + \rho |2\rangle \langle 2| - 2 |g\rangle \langle 2| \rho |2\rangle \langle g| \right)$$
Quantum optical model: Master equation

Two-level system

\[
\frac{\partial}{\partial t} \rho = -\frac{\gamma}{2} \left( |e\rangle \langle e| \rho + \rho |e\rangle \langle e| - 2 |g\rangle \langle e| \rho |e\rangle \langle g| \right)
\]

\[
\frac{\partial}{\partial t} \rho_{ee} = -\gamma \rho_{ee} \\
\frac{\partial}{\partial t} \rho_{eg} = -\frac{\gamma}{2} \rho_{eg}
\]

Three-level system

\[
\frac{\partial}{\partial t} \rho = -\frac{\gamma}{2} \left( |1\rangle \langle 1| \rho + \rho |1\rangle \langle 1| - 2 |g\rangle \langle 1| \rho |1\rangle \langle g| \right)
\]

\[
-\frac{\gamma}{2} \left( |2\rangle \langle 2| \rho + \rho |2\rangle \langle 2| - 2 |g\rangle \langle 2| \rho |2\rangle \langle g| \right)
\]

\[
-\frac{\gamma_C}{2} \left( |1\rangle \langle 2| \rho + \rho |1\rangle \langle 2| - 2 |g\rangle \langle 1| \rho |2\rangle \langle g| \right)
\]

\[
-\frac{\gamma_C}{2} \left( |2\rangle \langle 1| \rho + \rho |2\rangle \langle 1| - 2 |g\rangle \langle 2| \rho |1\rangle \langle g| \right)
\]

Find additional terms!
Fundamental light-matter interactions

- **Spontaneous coherences** can be generated by virtual photon exchange involving different states in the same atom.
- Desirable consequences, but usually forbidden e.g. by selection rules.
- Literally hundreds of theory papers on this topic.
- So far no experimental observations of these V-type SGC.

---

- **Decay, Lamb shift** (same atom, same transition)
- **Dipole-dipole interaction** (other atom)
- **Spontaneous coherences** (same atom, different transition)
Conditions for SGC

Requirements for SGC

\[ |1\rangle \xrightarrow{\text{SGC}} |2\rangle \]

**Condition I**

\[ \vec{d}_1 \cdot \vec{d}_2 \neq 0 \]

non-orthogonal
dipole moments

**Condition II**

\[ E_1 \approx E_2 \]

approx. same
transition energy

Re-absorption to \(|1\rangle\) and \(|2\rangle\) should be indistinguishable

Conditions not met, e.g., in atoms
Such SGC so far not observed!
Quantum optical model: Master equation

Two-level system

\[
\frac{\partial}{\partial t} \rho = -\frac{\gamma}{2} \left( |e\rangle \langle e| \rho + \rho |e\rangle \langle e| - 2 |g\rangle \langle e| \rho |e\rangle \langle g| \right)
\]

\[
\frac{\partial}{\partial t} \rho_{ee} = -\gamma \rho_{ee} \quad \frac{\partial}{\partial t} \rho_{eg} = -\frac{\gamma}{2} \rho_{eg}
\]

Three-level system

\[
\frac{\partial}{\partial t} \rho = -\frac{\gamma}{2} \left( |1\rangle \langle 1| \rho + \rho |1\rangle \langle 1| - 2 |g\rangle \langle 1| \rho |1\rangle \langle g| \right)
\]

\[
-\frac{\gamma}{2} \left( |2\rangle \langle 2| \rho + \rho |2\rangle \langle 2| - 2 |g\rangle \langle 2| \rho |2\rangle \langle g| \right)
\]

\[
-\frac{\gamma_c}{2} \left( |1\rangle \langle 2| \rho + \rho |1\rangle \langle 2| - 2 |g\rangle \langle 1| \rho |2\rangle \langle g| \right)
\]

\[
-\frac{\gamma_c}{2} \left( |2\rangle \langle 1| \rho + \rho |2\rangle \langle 1| - 2 |g\rangle \langle 2| \rho |1\rangle \langle g| \right)
\]

Find additional terms!
Quantum optical model: Master equation

Two-level system

\[ \frac{\partial}{\partial t} \rho = -\frac{\gamma}{2} (|e\rangle \langle e| \rho + \rho |e\rangle \langle e| - 2|g\rangle \langle e| \rho |e\rangle \langle g|) \]

\[ \frac{\partial}{\partial t} \rho_{ee} = -\gamma \rho_{ee} \quad \frac{\partial}{\partial t} \rho_{eg} = -\frac{\gamma}{2} \rho_{eg} \]

Three-level system with SGC, diagonalized

\[ \frac{\partial}{\partial t} \rho = -\frac{\gamma + \gamma_C}{2} (|S\rangle \langle S| \rho + \rho |S\rangle \langle S| - 2|g\rangle \langle S| \rho |S\rangle \langle g|) \]
\[ -\frac{\gamma - \gamma_C}{2} (|A\rangle \langle A| \rho + \rho |A\rangle \langle A| - 2|g\rangle \langle A| \rho |A\rangle \langle g|) \]

\[ |S\rangle = \frac{1}{\sqrt{2}} (|1\rangle + |2\rangle) \quad \text{constructive interference} \]

\[ |A\rangle = \frac{1}{\sqrt{2}} (|1\rangle - |2\rangle) \quad \text{destructive interference} \]

dark line in spectrum
Susceptibility with and without SGC

Without SGC

With SGC

Incoherent sum of two resonances

Interference → observed with nuclei

SGC is essential in our setup.

Reflectance (log scale)

Results with / without SGC differ strongly
Experiment at PETRA III (DESY Hamburg)
Experiment at PETRA III (DESY Hamburg)
Experimental results

Experiment+theory

(including exp. details)

First observation of such SGC!

K. P. Heeg, R. Röhlsberger, J. Evers, in preparation
Experimental results

really zero -
clean system, 
no decoherence!

This indicates 
that essentially 
no decoherence 
occurs 
IDEAL CASE
Why can SGC be observed in nuclei?

First mechanism: “Quantum simulator”

In cavity: many-body system which “microscopically” shows no SGC

probed from outside appears as single system with SGC

Second mechanism: “Anisotropic vacuum”

SGC can appear in atoms in anisotropic environments (proposal by G. S. Agarwal)

Cavity together with superradiance leads to effective coupling already in single nuclei

K. P. Heeg, R. Röhlsberger, J. Evers, in preparation
X-ray waveguides: Present status

- Superradiance
- Cooperative Lamb shift (first observation)

- EIT
- Novel mechanism to tailor level schemes

- Externally tunable level schemes
- Implementation and first direct observation of SGC
X-ray waveguides: Present status

- Superradiance
- Cooperative Lamb shift (first observation)
- EIT
- Novel mechanism to taylor level schemes
- Externally tunable level schemes
- Implementation and first direct observation of SGC
X-ray waveguides: Present status

Nuclear resonance scattering

CONUSS (numerical) → layer formalism (analytical)

very good agreement

Quantum optics

Master equation

analytical equivalence

\[ R \sim \chi \]

new input to both fields

promising basis for implementation of advanced quantum optical techniques in hard x-ray range

K. P. Heeg, R. Röhlsberger, J. Evers, in preparation
Nuclear resonance scattering

CONUSS (numerical)  layer formalism (analytical)

very good agreement

Quantum optics

Master equation

quantized light
non-linear light-matter coupling

promising basis for implementation of advanced quantum optical techniques in hard x-ray range

K. P. Heeg, J. Evers, work in progress
Crossed polarimeter setup

Developed at Uni Jena, installed at Petra III (DESY Hamburg)
Alternatives to nuclei
Inner-shell electrons

Controlling X-rays with light

T. E. Glover¹, M. P. Hertlein¹, S. H. Southworth², T. K. Allison¹, J. van Tilborg¹, E. P. Kanter², B. Krässig², H. R. Varma², B. Rude³, R. Santra², A. Belkacem¹ and L. Young²

control x-ray absorption with light?
Temporal coherence

- All quantum optical effects rely on coherence and interference.
- Synchrotron experiments operate at the single photon level, and single photons interfere with themselves.
- But: strong and coherent driving is key to most quantum optical effects.
- Availability of temporally coherent pulse with many resonant photons within nuclear linewidth would enable entirely new possibilities.
Challenges

Lifetimes

- Two ground states should be stable
- Here, they are not: ionization, fast decay of core-holes
- Solution: Intense control with Rabi flopping faster than loss

Level structure

- Ideally, 3-level Λ system
- Here, potentially many levels coupled

Fast timescale

- Core hole life time 2.4fs
- Need ultrafast x-ray and control laser
- Need to synchronize the two laser
Without control laser

- Rydberg series with clear absorption on transition 1s-3p can be seen
With control laser

- With control laser the transparency is increased
- The higher the control intensity, the higher the increase
- EIT could not be established due to high decoherence

![Diagram showing energy levels and x-ray transition](image)
<table>
<thead>
<tr>
<th>Content</th>
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<tbody>
<tr>
<td><strong>Introduction / NFS</strong></td>
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<tr>
<td><strong>Quantum optics and information</strong></td>
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<tr>
<td><strong>Nuclear quantum optics</strong></td>
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<tr>
<td><strong>Future perspectives</strong></td>
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</tbody>
</table>
## Synchrotron radiation vs. seeded FEL beams

<table>
<thead>
<tr>
<th></th>
<th>Synchrotron</th>
<th>Seeded XFEL</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bunch separation (ns)</td>
<td>200ns</td>
<td>200ns (microbunch)</td>
</tr>
<tr>
<td>Avg Flux (ph/s/Γ)</td>
<td>$5 \times 10^4$</td>
<td>$2 \times 10^8$</td>
</tr>
<tr>
<td>Fluence (ph/bunch/Γ)</td>
<td>$10^{-2}$</td>
<td>$6 \times 10^3$</td>
</tr>
</tbody>
</table>

### Nuclear parameters (for $^{57}$Fe)
- Energy: 14.4 keV
- Linewidth: 5 neV

### XFEL parameters
- 10$^{12}$ photons/pulse
- Rel. BW: $6 \times 10^{-5}$
- Rep. rate: 30 kHz

### Two directions
- Photon hungry ("proven concepts with higher count rate")
- Short, nonlinear, coherent ("new ideas")

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Geloni et al, arXiv:1111.5766
Nonequilibrium lattice dynamics

- Nuclei can not only monochromatize to sub-meV
- fs pulses capture snapshots of fast dynamics
- XFEL can produce double pulses with low jitter (< 5 fs)
- Small focus/isotope selective absorption provide high spatial resolution
- Long signal tail alleviates background / detection problems
- Example application: Heat transfer on nano scale

Nuclear lighthouse effect

- Exploit long lifetime of exciton to map between time and space
- Temporal resolution better than the incident pulse duration possible
- Can resolve high internal magnetic fields i.e. fast beat periods

Low-energy condensed matter excitations

- Polarizer/Analyzer blocks all light

- Only exception: polarization-rotating scattering via iron nuclei

- This process is restricted to narrow linewidth of iron (few neV)

- Tunable via Doppler shift due to mirror rotation

- Method was shown to work, but not enough signal from synchrotron sources for inelastic scattering

Low-energy condensed matter excitations

- Spectroscopy with μeV bandwidth tunable over ~meV scale

- Advantage of x-rays:
  - very high energy and angular resolution
  - reach more parts of phase space due to high brilliance
  - smaller samples accessible

- XFEL could make this feasible for inelastic scattering

<table>
<thead>
<tr>
<th>Energy (meV)</th>
<th>Excitations</th>
</tr>
</thead>
<tbody>
<tr>
<td>100 meV</td>
<td>Multiphonons &amp; Multimagnons ~ 50 – 500 meV</td>
</tr>
<tr>
<td>1 meV</td>
<td>Pseudogap ~ 30 meV – 300 meV</td>
</tr>
<tr>
<td>0.1 meV</td>
<td>Optical phonons ~ 40 – 70 meV</td>
</tr>
<tr>
<td>100 neV</td>
<td>Magnons ~ 10 meV – 40 meV</td>
</tr>
<tr>
<td>1 neV</td>
<td>Superconducting gap ~ 1 – 100 meV</td>
</tr>
<tr>
<td></td>
<td>Dynamics of disordered and nano systems</td>
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<tr>
<td></td>
<td>Boson peak in Amorphous Solids</td>
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<tr>
<td></td>
<td>Phasons in quasicrystals</td>
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<tr>
<td></td>
<td>Rotational excitations in liquids</td>
</tr>
<tr>
<td></td>
<td>Soft phonons</td>
</tr>
<tr>
<td></td>
<td>Relaxation dynamics of fluids ~ 1 – 400 neV</td>
</tr>
<tr>
<td></td>
<td>Hyperfine Interactions ~ 1 – 100 neV</td>
</tr>
</tbody>
</table>

Exciton manipulation without material motion

- Steer / control / split / focus light
- Method: Apply controlled phase patterns to stored excitons

Possible application: “Virtual” interferometer

Operation possible without need to stabilize material parts???
Can we enter the non-linear regime?

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

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

**EIT case:** Kerr effect

\[ n = n_0 + I_P n_2 \quad \chi = \chi^{(1)} + 3I_P \chi^{(3)} \]

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

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

---

EIT: no linear absorption, strong enhancement via advanced schemes possible.

---

What would nonlinear effects be good for?

Nonlinear spectroscopy / imaging

- CARS: Signal from ensemble of scatterers coherently adds up
- Better spatial resolution for nonlinear imaging
- Directed signal emission due to phase matching
- Decouple probe wavelength and x-ray spatial resolution
  

- Downconversion/wave mixing recently observed
  
Immediate applications of multiple photons

Separate coupling and probe
- Drive multiple modes simultaneously
- Beams could be individually and mutually temporally coherent

Quantum information and fundamental tests
- QIP protocol with qubit photons and quantum channel photons
- Entangled pairs of photons (downconversion or scheme by Rempe)

State preparation and pumping
- Isomer triggering
- X-ray induced emission with nuclei

Immediate applications of multiple photons

Separate coupling and probe

- Drive multiple modes simultaneously
- Beams could be individually and mutually temporally coherent
- Advanced level schemes?
- Two-photon entanglement, photon cross-correlations?

probe - coupling
Light propagation in cavities

- Use EIT or SGC to coherently control light propagation in thin film cavities
- Long propagation times with low losses achievable
- Applications:
  - nm sized x-ray sources (c.f. T. Salditt, Göttingen)
  - Enhanced light/matter or light/light interaction
  - embed target in cavity
  - deposit cavity on target “evanescent field coupling”

J. Evers et al, in preparation
Climbing up the Dicke ladder

Go beyond single excitation

- Much richer dynamics
- Can one stay in maximally superradiant branch?
- Dynamical beats with many excitations?
- Interactions between different excitons?
- Borrow ideas from solid state physics?

![Diagram showing ladder-like structure with excitons](image)
Direct laser driving of nuclei

- “Nuclear Rabi flopping”
- Nuclear and light frequencies could be matched using target acceleration
- Conceptionally most direct analogy to Quantum optics (boring?)
- But: Challenging to achieve significant inversion probably even with seeded FELs

More recent work by A. Palffy et al
Quantum-assisted measurements

\[ |N\rangle_A |0\rangle_B + |0\rangle_A |N\rangle_B \]

\[
\rightarrow |N\rangle_A |0\rangle_B e^{iN\phi} + |0\rangle_A |N\rangle_B
\]

- The N00N state leads to a phase shift multiplied by N
- This leads to a N-fold enhancement of the resolution
- The N00N state is a highly non-classical, entangled state
- |2002> can be produced by Hong-Ou-Mandel effect
X-ray optomechanics

Fundamental physics with mechanical resonators

feasible at all?

I. Pikovski et al., Nature Physics 8, 393 (2012)
Where could this lead?

**Optical analogues of general relativity**

- Interesting effects arise if medium moves faster than speed of light in the medium
- Difficult to move macroscopic objects at speed of light – thus make light slow
- Can create optical analogues of event horizons, black holes, Hawking radiation, ...
- Solid state nuclear systems are good candidates:  
  - background free measurements  
  - fast rotation and motion of nuclear media has already been exploited  
  - slow light is likely to occur in existing systems, but not yet verified

Where could this lead?

Quantum transport

- Designer quantum channels
- Start from a clean system, then add decoherence / dephasing at will
- Model complex bath by perturbing the transport sites independently using laser, E/B field, vibrations, ...
- Does optimal transport require coherence/ decoherence/ entanglement/...?
- What are experimental signatures applicable to complex transport systems?
- How can we control quantum mechanical energy transport to exploit it for applications?
- Need many photons to monitor transport “online”

Excitation: coherently controlled sample

NFS detector

array of nano-structured targets as transport medium

J. Evers, work in progress
“Wish list”

Exciting possibilities, but

- Resonant driving of Mößbauer nuclei mandatory, $^{57}$Fe requires 14.4 keV
- X-ray distribution system should be compatible with nuclear resonances
- Many photons per nuclear linewidth to achieve qualitative difference to synchrotrons
- Long pulses / low initial bandwidth favorable for “non-ultrafast” applications (more photons in resonance)
- Temporally coherent single or mutually coherent double pulses desirable for advanced quantum optical schemes

Geloni, arXiv:1111.5766
## The team

<table>
<thead>
<tr>
<th>Name</th>
<th>Position</th>
</tr>
</thead>
<tbody>
<tr>
<td>Martin Gärttner</td>
<td>PhD student</td>
</tr>
<tr>
<td>Qurrat-ul-Ain Gulfam</td>
<td>PhD student</td>
</tr>
<tr>
<td>Kilian Heeg</td>
<td>PhD student</td>
</tr>
<tr>
<td>Paolo Longo</td>
<td>PostDoc</td>
</tr>
<tr>
<td>Andreas Reichegger</td>
<td>Master student</td>
</tr>
<tr>
<td>David Schönleber</td>
<td>Master student</td>
</tr>
<tr>
<td>Lida Zhang</td>
<td>PhD student</td>
</tr>
</tbody>
</table>

## Collaboration (DESY)

- Ralf Röhlsberger
- Hans Christian Wille
- Kai Schlage

## Funding:

- MPG, DFG, DAAD,
- IMPRS-QD, CQD
Summary

X-ray entanglement

Quantum transport

Tunable nuclear level schemes

spontaneously generated coherences

Thank you!
Scattering function in the time domain

Setup with NRS in two $^{57}$Fe foils

Two interfering scattering pathways

\[ E_m(\mathbf{q}, t) \propto G_1(t)e^{-i(\omega_0 + \Omega)t} \int \rho_m(\mathbf{r}, t)e^{i\mathbf{q} \cdot \mathbf{r}} d\mathbf{r} \]
\[ + G_2(t)e^{-i\omega_0 t} \int \rho_m(\mathbf{r}, t = 0)e^{i\mathbf{q} \cdot \mathbf{r}} d\mathbf{r} \]

Spatial coherence and large resonant flux could enable position and time resolved study of scattering function over large parameter space

Probing fast dynamics at the nanoscale

- Scattering is characterized by the scattering function $S$ and transition rate $R \sim \left| S(\bar{Q}, \omega) \right|^2$

- Measurements in energy domain not favorable if
  - scattering medium changes with time (diffusion, molecular motion, short-lived quasiparticles, ...)
  - strong interaction leads to broadening of resonances

Then it is favorable to measure in time domain:

$$S(\bar{Q}, t) = \int S(\bar{Q}, \omega) e^{i\omega t} \, d\omega$$

- Need high Q and t range, large signal/noise ratio

- Example application: correlated electron materials

---

A. Q. R. Baron et al, Phys. Rev. Lett. 79, 2823 (1997); SwissFEL Science Case
X-ray and γ-ray quantum optics @ MPIK

Direct laser driving of nuclei


Isomer triggering


Yoctosecond physics


X-ray cooperative light scattering

Possible proof-of-principle experiment

- Without phase shifts: All \( N \) photons go to \( C \) (\( G_N \))

- With phase shift by Alice:
  \[ N_A = \sin^2(\phi_A/2) \, N \] photons go to \( D \) (\( G_A \))

- With phase shift by Bob:
  \[ N_B = \sin^2(\phi_B/2) \, N \] photons go to \( D \) (\( G_B \))

- With both phase shifts:
  \[ N_{AB} = \sin^2[(\phi_A - \phi_B)/2] \, N \] go to \( D \) (\( G_{AB} \))

Locality assumption: photons which arrive at \( C \) both
- if (Alice shifts but not Bob) and if (Bob shifts but not Alice)
- will still arrive at \( C \) if (Alice and Bob shift)

\[
(G_N - G_A) \cap (G_N - G_B) \subseteq (G_N - G_{AB})
\]

\[ N_{AB} \leq N_A + N_B \] violated for some phase shifts

Experimental evidence with local oscillator

Visibility (91 ± 3)% with background correction
Visibility (66 ± 2)% without background correction
71% limit for violation of Bell inequality

Single photon entanglement teleportation scheme

Sender

Alice

classical communication

Possible verification at receiver side

Source

entanglement generation

input state

single photon

Teleportation algebra

\[ |\Psi\rangle = \frac{1}{\sqrt{2}} (|1\rangle_A|0\rangle_B + |0\rangle_A|1\rangle_B)(a|1\rangle_C + b|0\rangle_C) \]

\[ = \frac{1}{2} |0\rangle_E|1\rangle_F (a|1\rangle_B + b|0\rangle_B) \]

\[ + \frac{1}{2} |1\rangle_E|0\rangle_F (a|1\rangle_B - b|0\rangle_B) \]

\[ + \frac{1}{2} \left( \frac{1}{2} |0\rangle_E|2\rangle_F - \frac{1}{2} |2\rangle_E|0\rangle_F + \frac{1}{\sqrt{2}} |0\rangle_E|0\rangle_F \right) (a|0\rangle_B + b|1\rangle_B) \]

\[ + \frac{1}{2} \left( \frac{1}{2} |0\rangle_E|2\rangle_F - \frac{1}{2} |2\rangle_E|0\rangle_F - \frac{1}{\sqrt{2}} |0\rangle_E|0\rangle_F \right) (a|0\rangle_B - b|1\rangle_B) \]

measurement Alice
Efficiency estimate

- Assumed rate of excited nuclei: \( \sim 10^6 / \text{s} \)
- Of stored excitation, 70% background, 30% signal
- Loss at polarizer: Only about 10% of photons are kept
- Single photon entanglement rate: \( \sim 10^3 / \text{s} \)

Signal and background separated!

Incident photon flux can be increased until multiple excitations occur
Theoretical description

Wave equation
\[
\left( \Delta - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \vec{E} = \frac{4\pi}{c} \frac{\partial}{\partial t} \vec{I}
\]

Slowly varying envelope approximation
\[
\frac{\partial}{\partial z} \vec{\mathcal{E}} = -\frac{2\pi}{c} \vec{I}
\]

Nuclei as source term (2\textsuperscript{nd} order)
\[
\vec{I} = \text{Tr} \left( \vec{J} \rho_{\text{nuclei}} \right)
\]

Final wave equation
\[
\frac{\partial \tilde{\mathcal{E}}(z, t)}{\partial z} = -\sum_{\ell} K_{\ell} \vec{J}_\ell(t) \int_{-\infty}^{t} d\tau J^\dagger_\ell(\tau) \cdot \tilde{\mathcal{E}}(z, \tau)
\]

Iterative solution, incident pulse
\[
\mathcal{E}^{(0)}(t) \sim \delta(t)
\]

Recent experiment: Collective Lamb Shift

- Lamb shift due to virtual photon exchange in ensembles of atoms
- Experimentally observed with nuclei using forward scattering
- Experimental challenge: Prepare purely superradiant state in thick sample; solution: embed nuclei in low-q cavity

Motivation

Light-matter interactions

- optical driving fields: excite/ionize outer electrons
- Higher frequencies/intensities: excite / ionize core electrons
- Even higher frequencies/intensities: excite nucleus

These scenarios appear similar

But the methods and applications are quite different
Step 5: Releasing linear polarization

- Initially, magnetic field is in \( z \) direction

- At time \( t_1 \), cancel decay by rotating into \( y \) direction

- At time \( t_2 \), enable decay on \( \Delta m = \pm 1 \) but continue to suppress \( \Delta m = 0 \)

- At time \( t_3 \), cancel decay by rotating into \( y \) direction

- At time \( t_4 \), enable decay on \( \Delta m = 0 \)
Advanced magnetic switching schemes

Rotation angle
- Determines new quantization axis and superposition states

Timing
- Important due to different transition energies
- Determine whether constructive/destructive interference occurs
- Example: Suppression at $t_1$, how does $t_2$ affect further evolution?

Transition amplitudes

$\begin{align*}
\left\{ \frac{1}{2} \rightarrow \frac{1}{2}, -\frac{1}{2} \rightarrow -\frac{1}{2} \right\} & \quad \text{linear} \\
\left\{ -\frac{1}{2} \rightarrow \frac{1}{2}, \frac{1}{2} \rightarrow -\frac{1}{2} \right\} & \quad \text{circular} \\
\left\{ \frac{3}{2} \rightarrow \frac{1}{2}, -\frac{3}{2} \rightarrow -\frac{1}{2} \right\} & \quad \text{circular}
\end{align*}$

Step 1: Synchrotron excitation

Initially, magnetic field is in the z direction.
Step 2: Canceling coherent decay

- Initially, magnetic field is in $z$ direction
- At time $t_1$, cancel decay by rotating into $y$ direction
Step 3: Releasing circular polarization

- Initially, magnetic field is in \( z \) direction.
- At time \( t_1 \), cancel decay by rotating into \( y \) direction.
- At time \( t_2 \), enable decay on \( \Delta m = \pm 1 \) but continue to suppress \( \Delta m = 0 \).

\[ \left\{ \begin{array}{c}
\frac{1}{2} \rightarrow \frac{1}{2}, \quad -\frac{1}{2} \rightarrow -\frac{1}{2} \\
-\frac{1}{2} \rightarrow \frac{1}{2}, \quad \frac{1}{2} \rightarrow -\frac{1}{2} \\
\frac{3}{2} \rightarrow \frac{1}{2}, \quad -\frac{3}{2} \rightarrow -\frac{1}{2}
\end{array} \right\} \]

\( \Delta m = \pm 1 \)

\( t_1 \) to \( t_2 \) in nanoseconds.
Step 4: Canceling coherent decay

- Initially, magnetic field is in $z$ direction.
- At time $t_1$, cancel decay by rotating into $y$ direction.
- At time $t_2$, enable decay on $\Delta m = \pm 1$ but continue to suppress $\Delta m = 0$.
- At time $t_3$, cancel decay by rotating into $y$ direction.
Engineering multi-level schemes

How to implement EIT in x-ray cavity?

Next talk

How can one

- Control and systematically study EIT without building many cavities?
- Engineer more complex level schemes?

Poster by Kilian Heeg

Coherent forward scattering

- Coherent scattering occurs in forward direction
- Similarity to multi-slit / grid diffraction but constructive interference only in forward / Bragg direction

\[
\lim_{N \to \infty} \sum_{i=1}^{N} e^{i(k - k_L) \cdot \vec{r}_i} \sim \delta(k - k_{L})
\]

grid = CD-R grooves
Cooperative light scattering

- Incident light
- Large dilute cloud $R \gg \lambda$
- Scattered light $\vec{k}_L$
- Quantum particles as scatterers
- No recoil
- Stationary particles
How to extract signal pulse?

- Problem: One part of signal has same polarization as background pulse.

- Time gating not useful if following setup should be protected from high-intensity background; lighthouse effect difficult because of precise timing of nuclear switching.

- PSM: Piezo electric steering mirror or sub-ns control device based on crystal lattice deformation\(^1\).

- Have about 180 ns “steering time” because of magnetic switching.

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Branching ratio

Single particle branching ratio:

- Determines ratio of spontaneous emission channels
- Property of the particle only

Branching ratio in ensembles

- Have cooperative modification of excitation and decay
- Determined by particle, ensemble and excitation properties, varies with time
- Need to define cooperative branching ratio
Motivation

Aim: Efficiently pump from ground state $|G\rangle$ to isomeric state $|I\rangle$

Cooperativity leads to enhanced excitation to $|E\rangle$, but also to fast decay

In effect, little transfer to $|I\rangle$

Idea:

Suppress cooperative emission

Then cooperativity leads to enhanced excitation, but decay proceeds with single particle branching ratio

In effect, enhanced pumping to $|I\rangle$

The ideal case

- Assume purely superradiant decay with rate $\xi \cdot \gamma$
- Assume perfect coherent control of cooperative decay

**Result:**

$$\frac{b^C_c}{b^{NC}_c} = \xi + 1$$

- Cooperative branching ratio is larger by factor $\xi + 1$
- In addition, cooperative enhancement of excitation
How to control?

**Magnetic switching:**
- Turn off cooperative decay via interference
- The incoherent decay with single-particle branching ratio remains

**Destroy phase coherence:**
- Use short pulse of incoherent light, spatially inhomogeneous magnetic field, or similar to destroy spatial coherence
- Without the coherence, uncorrelated decay without cooperative enhancement
- Can be done immediately after excitation, does not require sophisticated pulse control
The magnetic switching case

Branching ratio time dependent as expected

Cooperative branching ratio smaller than single-particle ratio due to superradiance

After switching, single-particle branching ratio is achieved

With destruction of phase coherence, single-particle ratio can immediately be achieved

Outline

Introduction

X-ray entanglement generation

X-ray branching ratio control

Outlook: Engineering advanced level schemes
Outline

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Motivation

Light-matter interactions

different paradigms

uncontrolled pump + passive observation

full quantum control

- X-ray physics could greatly benefit from moving more towards quantum control
- What can be done is to large degree determined by availability of light sources
- New possibilities with seeded FEL?
Layer formalism

- How to calculate $R$?

**field amplitude:**

$$A(z) = \begin{pmatrix} A_{in}(z) \\ A_{out}(z) \end{pmatrix}$$

**propagation equation:**

$$\frac{d}{dz} A = iFA$$

- scattering amplitudes

**reflectivity:**

$$R = \frac{A_{out}(0)}{A_{in}(0)}$$

$$A_{in}(0) \rightarrow A_{in}(D) = 0$$

$$A_{out}(0) \rightarrow A_{out}(D) = 0$$

$$\int N \sim \frac{1}{\hbar \omega - E + i\gamma/2}$$
Directionality

\[
\lim_{N \to \infty} \sum_{i=1}^{N} e^{i(k - \vec{k}_L) \vec{r}_i} \sim \delta\left(\vec{k} - \vec{k}_L\right)
\]
Rough efficiency estimate

- Assumed incoming flux after monochromator: $10^9$ photons / s
- Assumed rate of excited nuclei: $5 \times 10^5$ / s
- Of stored excitation, 70% background, 30% signal
- Loss at polarizer: Only about 10% of photons are kept
- Single photon entanglement rate: $15 \times 10^3$ / s