



# Quantum optics with novel coherent light sources

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#### Light-matter interactions



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IR/optical driving fields: excite/ionize outer electrons





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- Higher frequencies/intensities: excite / ionize core electrons
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- Even higher frequencies/intensities: excite nucleus

What can be done is to a large degree determined by the availability of light sources

full quantum control



uncontrolled pump + passive observation

#### Free electron laser



Working principle

- Photon energy up to few keV
- Full transverse coherence, upgrade to full longitudinal coherence possible



Short pulses



SLAC linear accelerator

(image from SLAC)

### Synchrotron



(image from DESY)

### Novel light sources in China



National Synchrotron Radiation Laboratory (USTC, Hefei)

#### Beijing Synchrotron Radiation Facility (CAS)



There are 3 experimental halls (12#, 13#, 15#), 5 insertion devices and 14 beamlines and stations in BSRF



Shanghai synchrotron facility Free electron laser (Shanghai, CAS)

> New machines always bring new opportunities

# Applications in the x-ray range

#### Quantum

- Note: The set of the
- 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

# X-ray and $\gamma\text{-ray}$ quantum optics @ MPIK



### Outline

Introduction

X-ray entanglement generation

X-ray branching ratio control

Outlook: Engineering advanced level schemes











#### Cooperative light scattering



 $ec{k}_L$  scattered light

quantum particles as scatterers



#### Elementary processes



#### Coherent forward scattering



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

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



grid = CD-R grooves

### <sup>57</sup>Fe iron Mößbauer transition



### Temporal beats





bichromatic scattered light

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

J. P. Hannon and G. T. Trammell, Hyperf. Int. 123/124, 127 (1999)

### Superradiance

#### Dicke case (small dense sample)

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

$$\langle G|\vec{d}|\Psi
angle = \sqrt{N} \langle g_i|\vec{d}|e_i
angle$$
  
 $\gamma \longrightarrow N \gamma$ 

#### NFS case (large dilute sample)

$$\Psi \rangle = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} e^{i\vec{k}\vec{r}_{i}} |g_{1}, \dots, g_{i-1}, e_{i}, g_{i+1}, \dots, g_{N} \rangle$$

- Superradiant state dynamically coupled to subradiant states
- Imperfect preparation of superradiant state in thick samples  $\rightarrow$  dephasing



M. O. Scully et al., Phys. Rev. Lett. 96, 010501 (2006)

#### (Some) characteristic features in NFS spectra



#### Experimental realization





Student lab Uni Mailand



#### Example: Coherent control via magnetic switching

The level structure depends on applied magnetic field: Zeeman splitting

- In certain crystals (e.g. FeBO<sub>3</sub>), the magnetic crystal field is very strong (~ 30 T), and can be aligned using a weak external field (few Gauss)
- This allows to switch the direction of a very strong effective magnetic field in few ns in the lab



### Optical response of a single resonance





# 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

S. Harris, Physics Today 50, 36 (1997); M. Fleischhauer et al., Rev. Mod. Phys. 77, 633 (2005)

# 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

### 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



 $\vec{R}$ 



# Control of coherent NFS

#### 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



Yu. V. Shvyd'ko et al., Phys. Rev. Lett. 77, 3232 (1996)



#### No switching

Apply switching Switch back Decay with natural life

time

#### 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





Röhlsberger et al, Science 328, 1248 (2010)

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### 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

# Single photon entanglement

Single photon impinging on 50/50 beam splitter gives output

$$|\Psi\rangle = \frac{1}{\sqrt{2}} \left(|0\rangle_A |1\rangle_B + |1\rangle_A |0\rangle_B\right)$$

- 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  $|\Psi\rangle = \frac{1}{\sqrt{2}} (|g_1 e_2\rangle + |e_1 g_2\rangle)$ 





S. J. van Enk, Phys. Rev. A 67, 022303 (2003)

### 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?



A. Palffy and J. Evers, J. Mod. Opt. 57, 1993 (2010)

#### Step 1: Synchrotron excitation







### Step 2: Canceling coherent decay



# Step 3: Releasing circular polarization



### Step 4: Canceling coherent decay



- At time t<sub>1</sub>, 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<sub>3</sub>, cancel decay by rotating into y direction





## Step 5: Releasing linear polarization

- Initially, magnetic field is in z direction
- At time t<sub>1</sub>, 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<sub>3</sub>, 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



#### A. Palffy, C. H. Keitel, J. Evers, Phys. Rev. Lett. 103, 017401 (2009)

### How to extract signal pulse ?

- Priblem: 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 prcise timing of nuclear switching
- PSM: Piezo electric steering mirror or sub-ns control device based on crystal lattice deformation<sup>1)</sup>
- Have about 180 ns "steering time" because of magnetic switching



1) A. Grigoriev et al., Appl. Phys. Lett. 89, 021109 (2006)

# Proof-of-principle experiment

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



\*) H.-W. Lee and Kim, Phys. Rev. A 63, 012305 (2000)

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# Application: Isomer triggering

#### Nuclear isomers:

long-lived nuclear states
 may "store" much energy

#### Motivation:

- "nuclear batteries"
- gamma-ray laser
- fundamental questions in astro- and nuclear physics



How to efficiently populate and trigger isomers?

See, e.g., P. M. Walker and J. J. Carroll, Nuclear Physics News 17, 11 (2007)

# 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
- ln effect, little transfer to  $|I\rangle$

#### Idea:



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





A. Palffy, C. H. Keitel, and J. Evers, Phys. Rev. B 83, 155103 (2011)

### The ideal case

- Assume purely superradiant decay with rate  $\xi$ · γ
- Assume perfect coherent control of cooperative decay



#### **Result:**

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

**Cooperative branching ratio is larger by factor**  $\xi+1$ 

In addition, cooperative enhancement of excitation

#### 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



A. Palffy, C. H. Keitel, and J. Evers, Phys. Rev. B 83, 155103 (2011)

### 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

A. Palffy, C. H. Keitel, and J. Evers, Phys. Rev. B 83, 155103 (2011)

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### X-ray cavities

- nm-sized thin film cavity: Pt (electron rich) as mirror, C (little electrons) as spacer
- Cavity is probed in grazing incidence, because of low index of refraction change

1.0

B

Cavity resonances give field enhancement, can be observed in reflection





## Purcell effect and cooperativity

- Look at cavity with 2 active layers
  - (A) in cavity field maximum
  - (B) in cavity field minimum
- (A) couples strongly to the cavity, rapidly emits excitation into cavity field (Purcell effect + cooperative light emission)
- (b) has suppressed coupling to cavity because of intensity minimum

Lifetime of nuclear excitation in (B) much longer than that of one in (A)

Material is the same!

Image: Röhlsberger et al, Nature 482, 199 (2012)



# Engineering a 3-level $\Lambda$ level scheme

#### The level scheme

- State |1> : no excitations in (A), (B) but photon in cavity
- State |3>: excitation in (A), no photon in cavity
- State |2>: excitation in (B), no photon in cavity

#### Why is this a $\Lambda$ level scheme?

- |3> decays fast due to Purcell + cooperativity
- Compared to that, |2> and |1> metastable
- Control is generated by scattering between the layers
- Probe field by absorption of cavity photon by nucleus in (A)



Image: Röhlsberger et al, Nature 482, 199 (2012)

### Experiment: Nuclear EIT



EIT as an archetype quantum optical coherence effect observed with x-rays interacting with nuclei

EIT with a single light field due to clever cavity engineering

Image: Röhlsberger et al, Nature 482, 199 (2012)

#### Our current work: engineer advanced schemes





- Broad transparency window to propagate of broadband input pulses
- Steep dispersion slope for strong effect on propagated pulse (e.g. delay)
- (time delay)·(transparency bandwidth) is constant → need to tune for best trade-off

- More general level schemes offer wide range of applications
- Example: Strongly enhanced nonlinear response

K. P. Heeg, R. Röhlsberger, J. Evers, work in progress

#### The team

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Thank you!

PhD / PostDoc applications welcome in Heidelberg!







# NFS

### 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

$$\frac{\partial}{\partial z}\vec{\mathcal{E}} = -\frac{2\pi}{c}\vec{\mathcal{I}}$$

#### Nuclei as source term (2<sup>nd</sup> order)

$$\vec{I} = \operatorname{Tr}\left(\vec{j}\rho_{\mathrm{nuclei}}\right)$$

#### Final wave equation

$$\frac{\partial \vec{\mathcal{E}}(z,t)}{\partial z} = -\sum_{\ell} K_{\ell} \vec{J}_{\ell}(t) \int_{-\infty}^{t} d\tau \vec{J}_{\ell}^{\dagger}(\tau) \cdot \vec{\mathcal{E}}(z,\tau)$$
sum over de-excitation excitation transitions

Iterative solution, incident pulse

Y. V. Shvydko, Hyperf. Int. 123/124, 275 (1999)

#### A few numbers - classification of the system

- Incident light bandwidth ~meV, Fe transition width ~neV
   → on average typically less than 1 excited nucleus per shot, "single photon"
- Solid state densities (  $n \sim 10^{23} / \text{cm}^3$ ) but short wavelength (  $\lambda \sim 10^{-10} \text{ m}$ )  $\rightarrow n\lambda^3 \sim 0.1 \rightarrow \text{``dilute'' medium}$ 
  - High resonant scattering amplitude, Mößbauer effect
     → large optical depth, multiple scattering
- Sample of macroscopic size compared to wave length (R /  $\lambda \gg 1$ )
- ▶ Focus on coherent forward scattering
   → Treatment of cooperative effects much simplified (e.g., no radiation trapping)



# 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<sub>A</sub>)

With phase shift by Bob:  $N_B = \sin^2(\phi_B/2) N$  photons go to D (G<sub>B</sub>)

With both phase shifts:  $N_{AB} = \sin^2[(\phi_A - \phi_B)/2] N$  go to D (G<sub>AB</sub>)



► 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

H.-W. Lee and Kim, Phys. Rev. A 63, 012305 (2000)

### Experimental evidence with local oscillator

#### single photon generation



B. Hessmo et al, Phys. Rev. Lett. 92, 180401 (2004)

### Single photon entanglement teleportation scheme





H.-W. Lee and Kim, Phys. Rev. A 63, 012305 (2000)

#### Teleportation algebra



measurement Alice

H.-W. Lee and Kim, Phys. Rev. A 63, 012305 (2000)

### Efficiency estimate

- Assumed incoming flux after monochromator: 10<sup>9</sup> 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



A. Palffy, C. H. Keitel, J. Evers, Phys. Rev. Lett. 103, 017401 (2009)