



Nuclear quantum optics at an XFEL

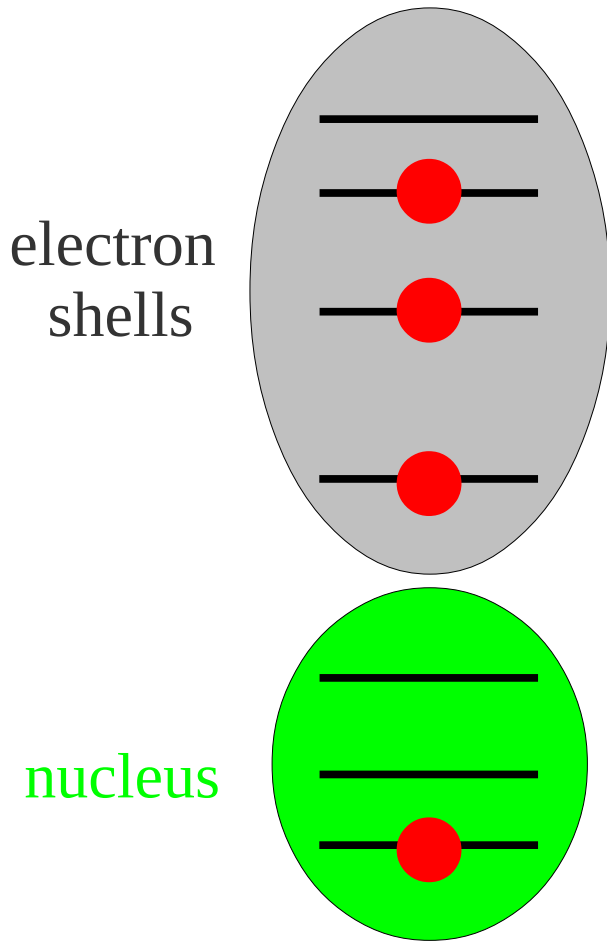
Jörg Evers

Max-Planck-Institut für Kernphysik, Heidelberg

Paul-Scherrer Institut, Villigen, Schweiz, 21.11.2012

Motivation

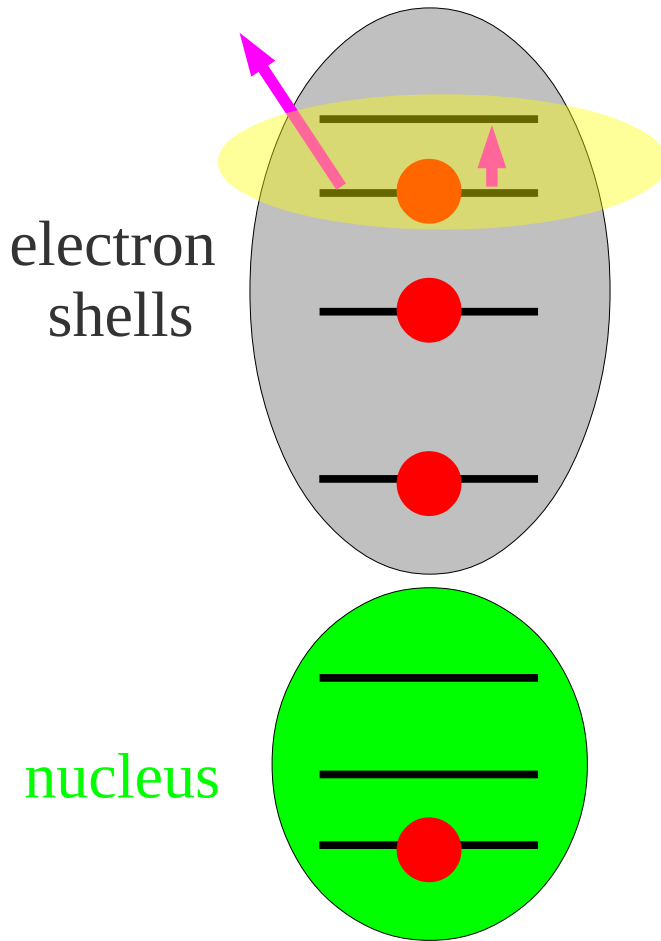
Light-matter interactions



Motivation

Light-matter interactions

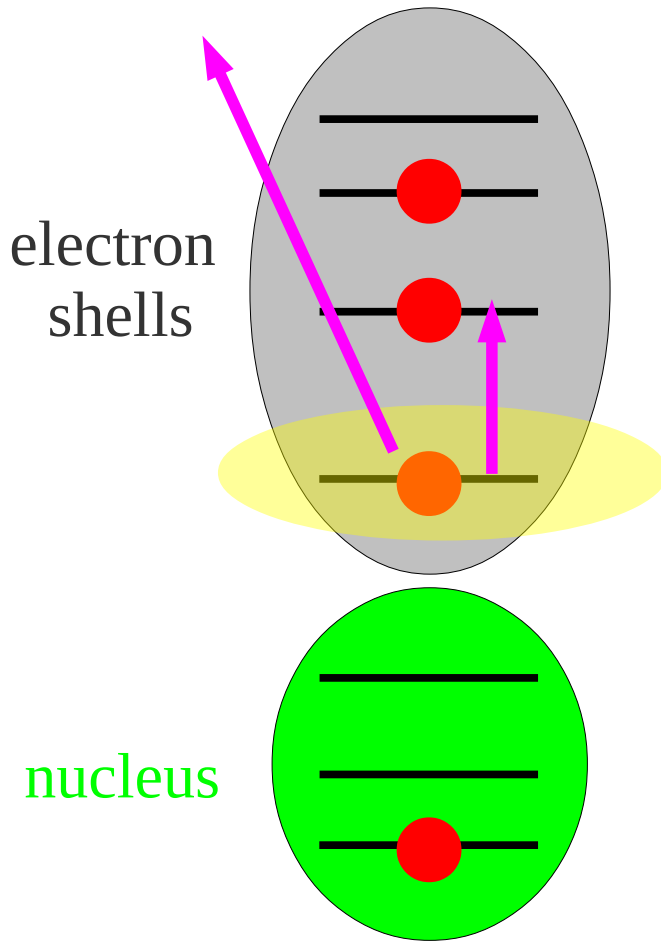
- ▶ optical driving fields:
excite/ionize outer electrons



Motivation

Light-matter interactions

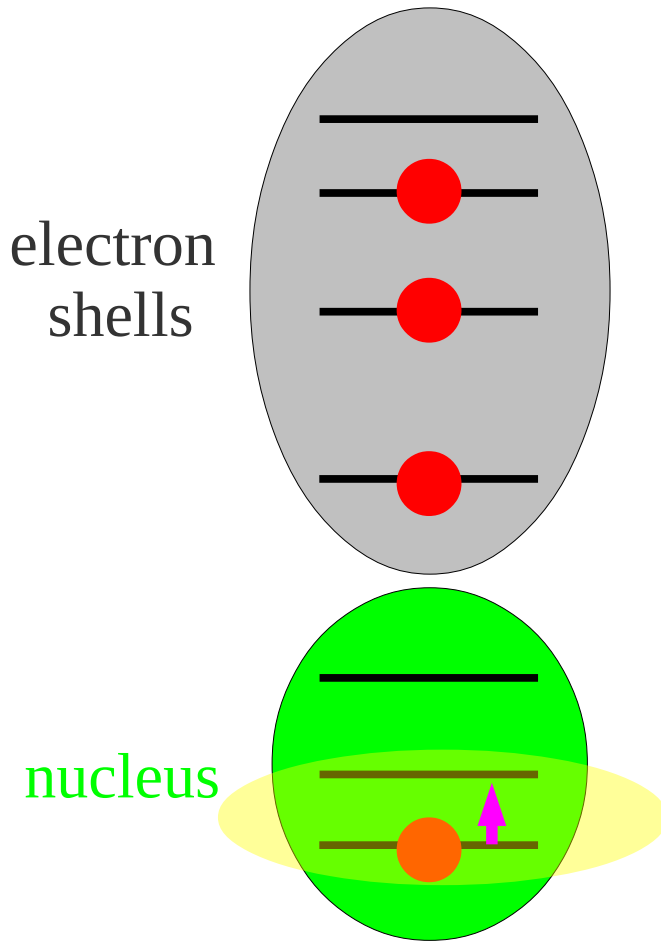
- ▶ optical driving fields:
excite/ionize outer electrons
- ▶ Higher frequencies/intensities:
excite / ionize core electrons



Motivation

Light-matter interactions

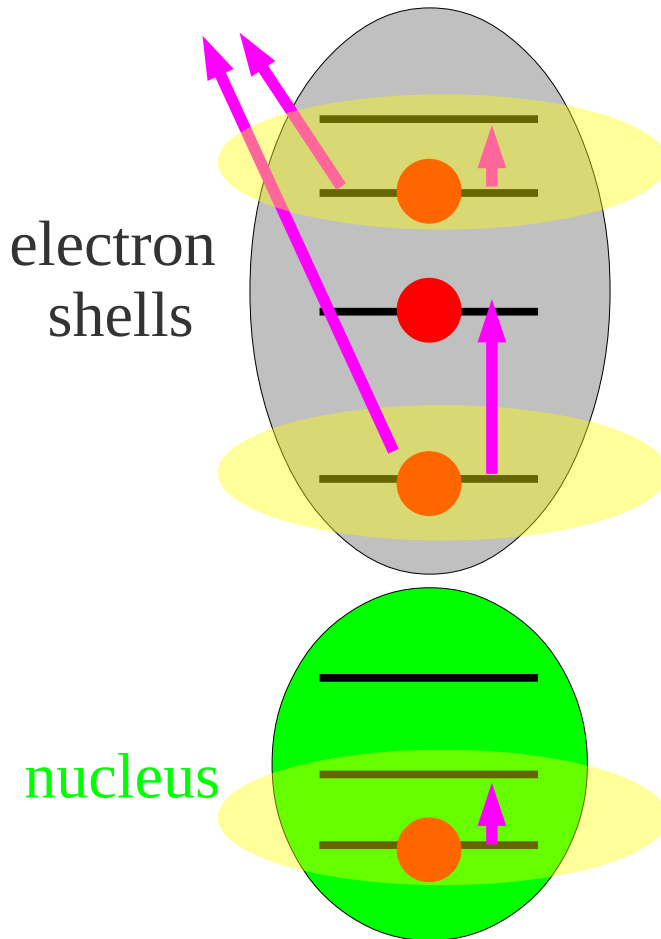
- ▶ optical driving fields:
excite/ionize outer electrons
- ▶ Higher frequencies/intensities:
excite / ionize core electrons
- ▶ Even higher frequencies/intensities:
excite nucleus



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

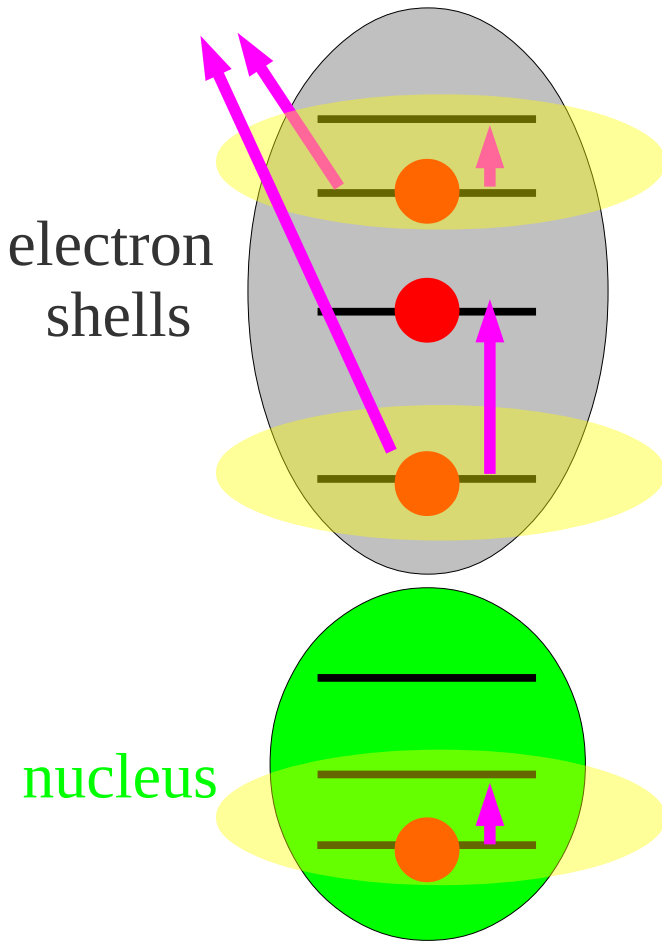
Light-matter interactions



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- λ 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 \rightarrow 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)

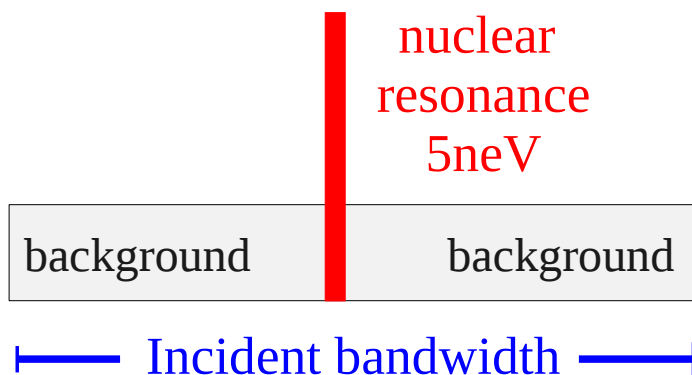
Our approach: Explore starting from experimentally proven setups, do not simply copy from optical case

Synchrotron radiation vs. seeded FEL beams

	Synchrotron	Seeded XFEL
Bunch separation (ns)	200ns	200ns (microbunch)
Avg Flux (ph/s/ Γ)	5×10^4	2×10^8
Fluence (ph/bunch/ Γ)	10^{-2}	6×10^3

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



Synchrotron radiation vs. seeded FEL beams

Two directions

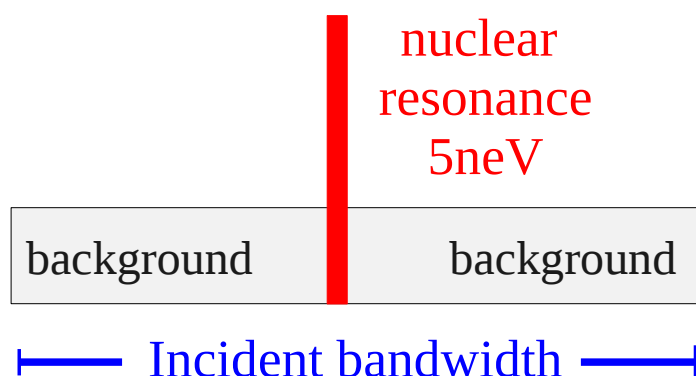
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photon hungry
("proven concepts
with higher
count rate")

Short, nonlinear,
coherent
("new ideas")

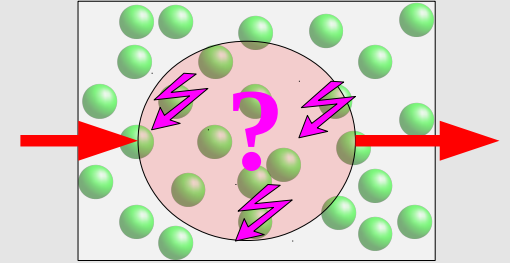
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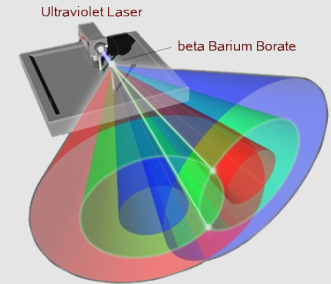


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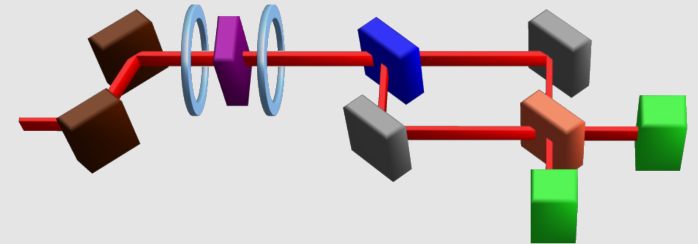
Introduction / NFS



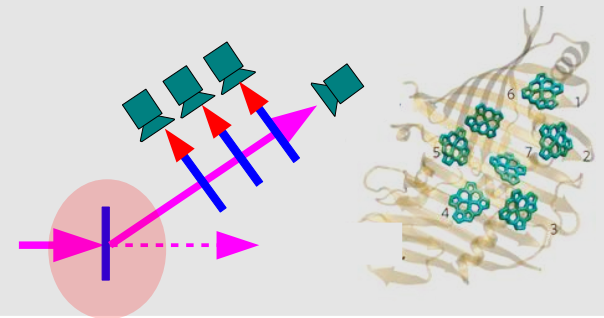
Quantum optics and information



Nuclear quantum optics

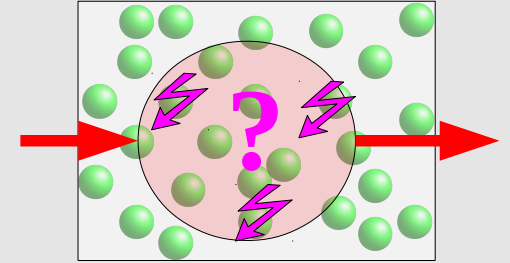


Future perspectives

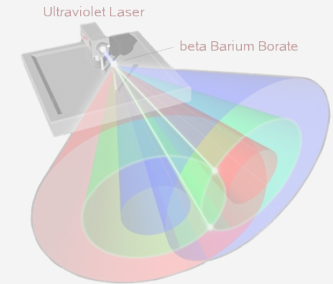


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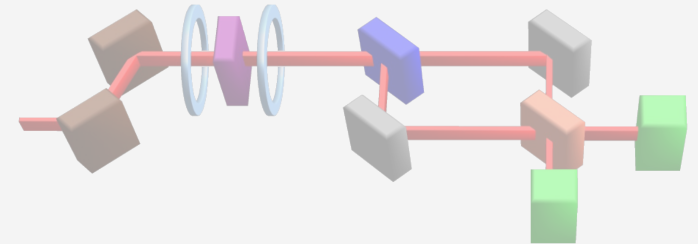
Introduction / NFS



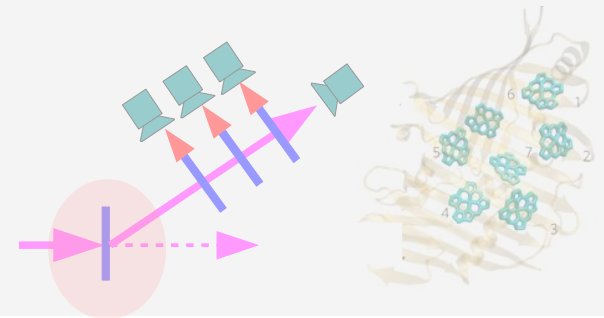
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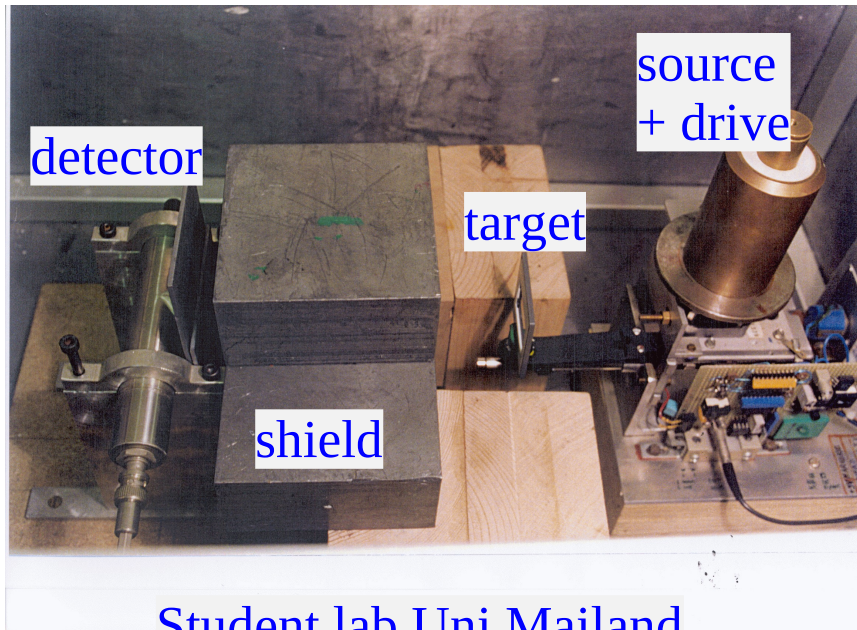
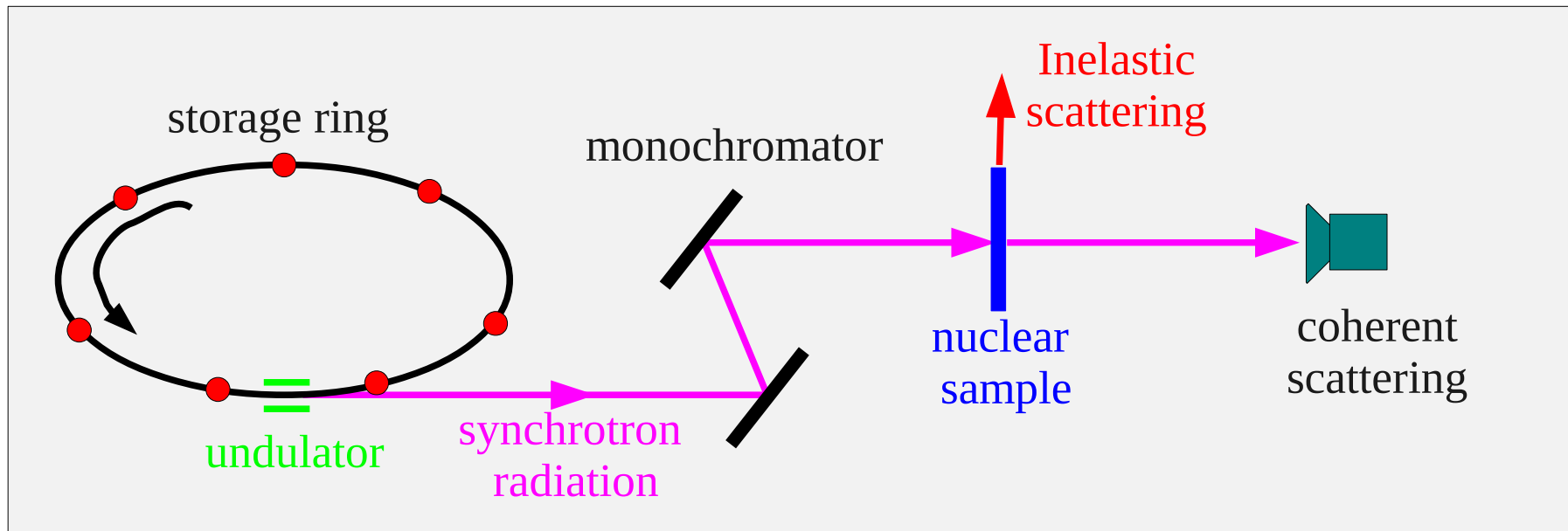
Nuclear quantum optics



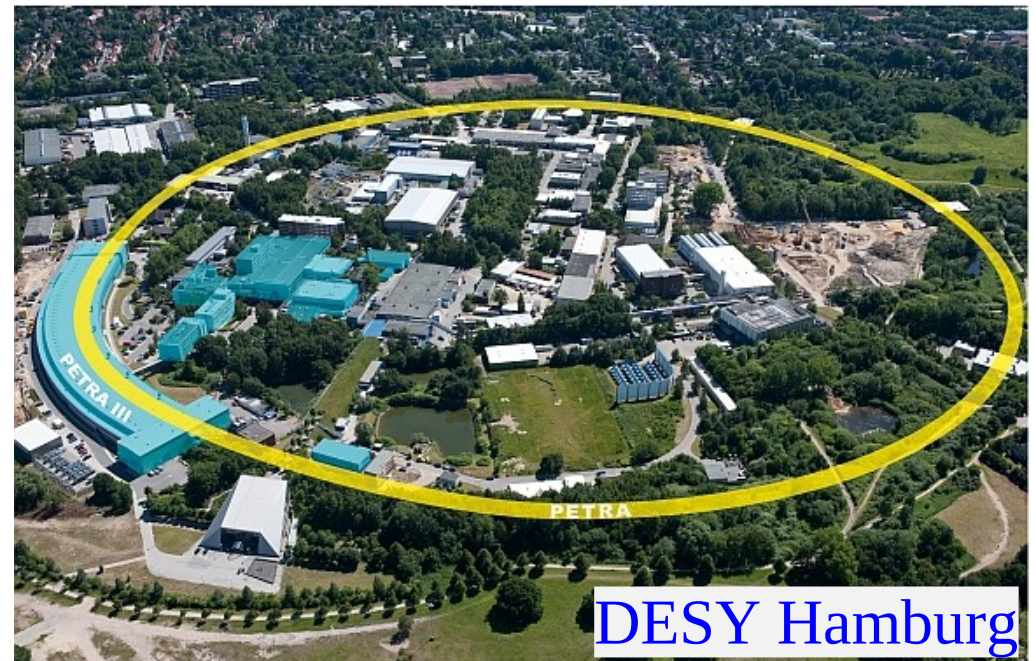
Future perspectives



Nuclear resonance scattering



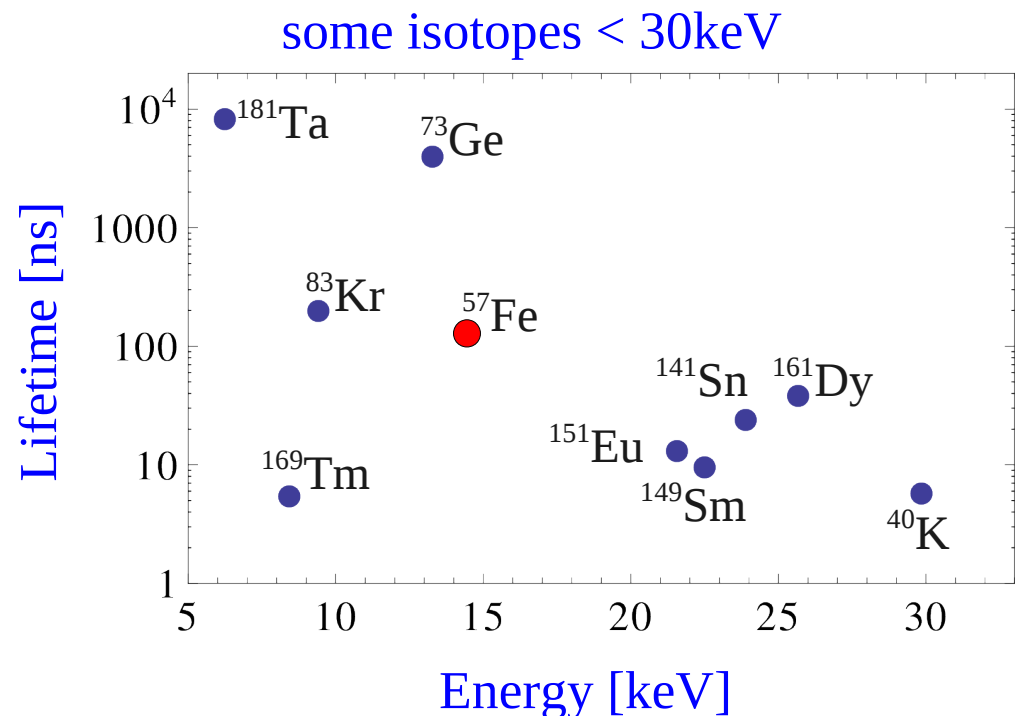
Student lab Uni Mailand



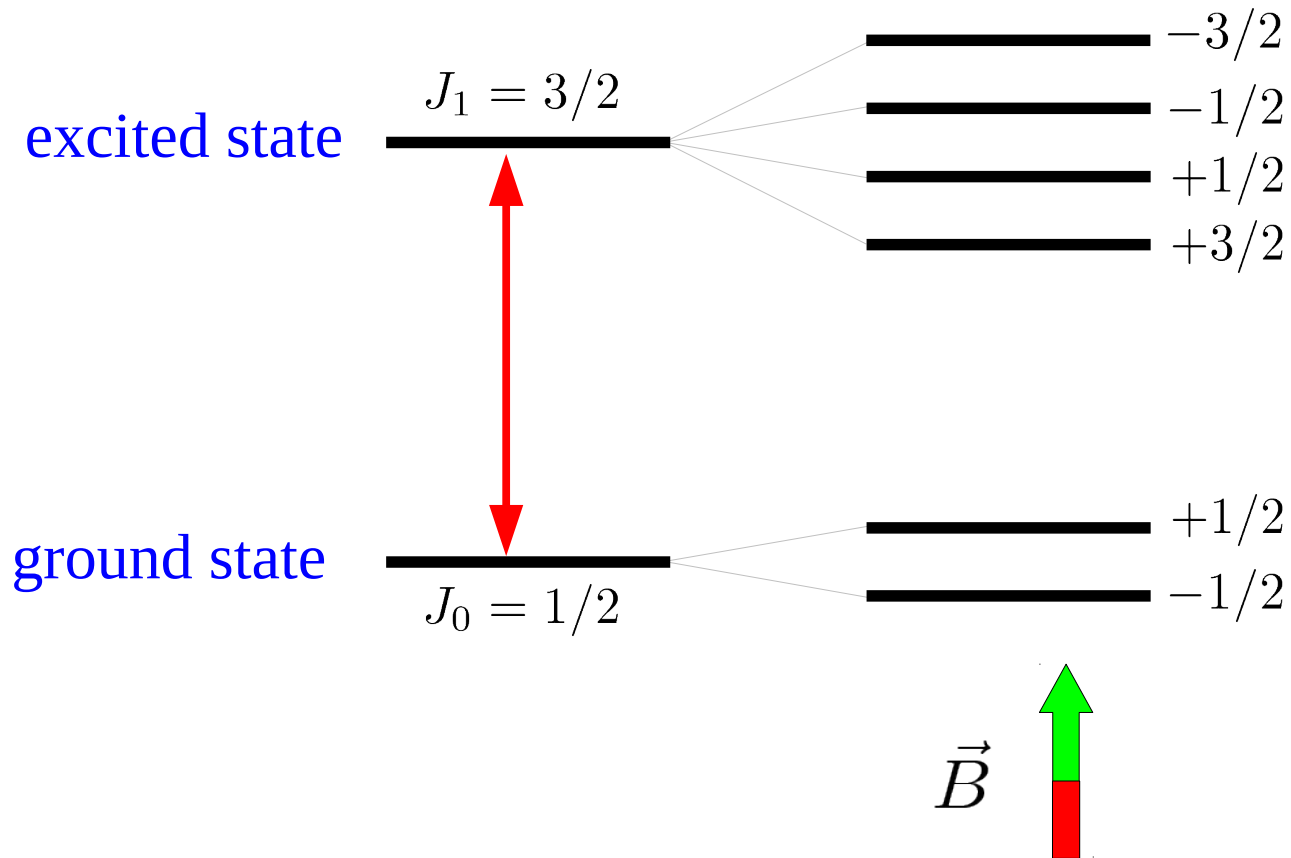
DESY Hamburg

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



^{57}Fe iron Mössbauer transition

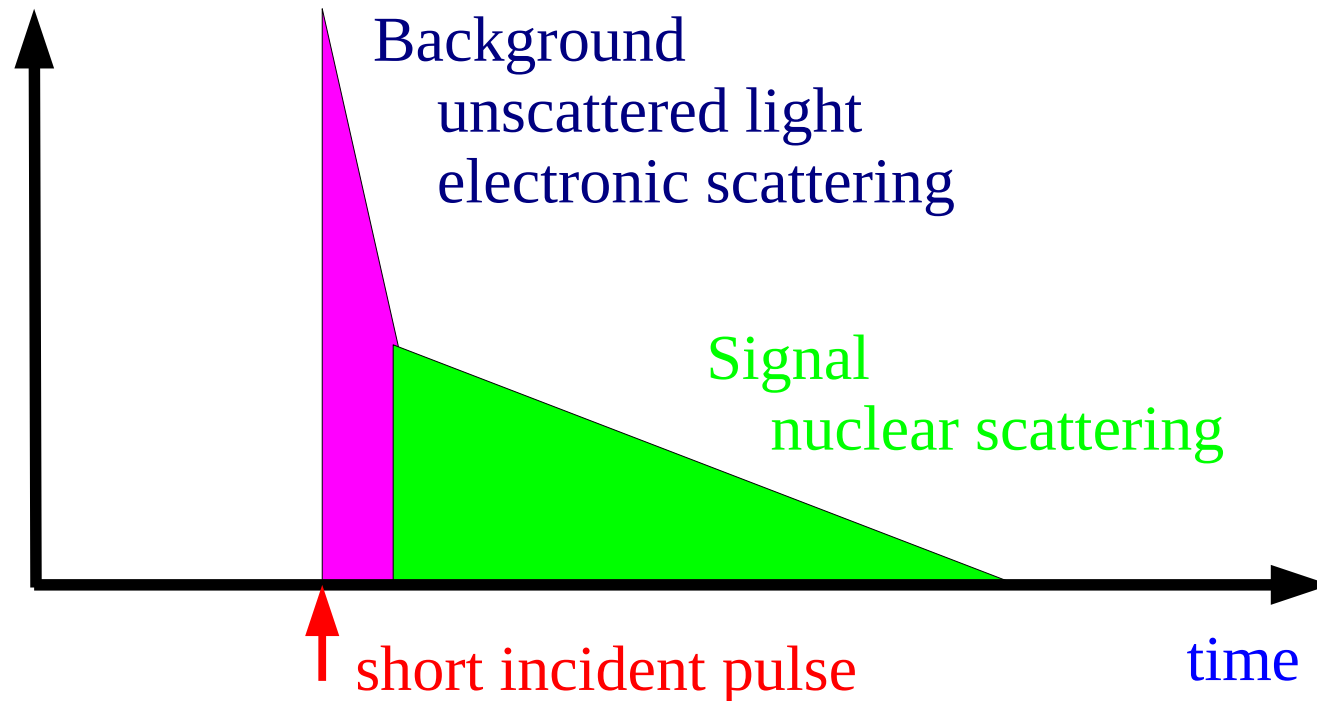


- ▶ Iron is of significance in biology, earth science, ...
- ▶ “Working horse” of nuclear resonance scattering
- ▶ $Q \sim \omega_0 / \Gamma \sim 10^{12}$

$$\lambda = 0.86 \text{ \AA}$$
$$\hbar\omega_0 = 14.4 \text{ keV}$$
$$\hbar\Gamma = 4.7 \times 10^{-9} \text{ eV}$$
$$1/\Gamma = 141 \text{ ns}$$

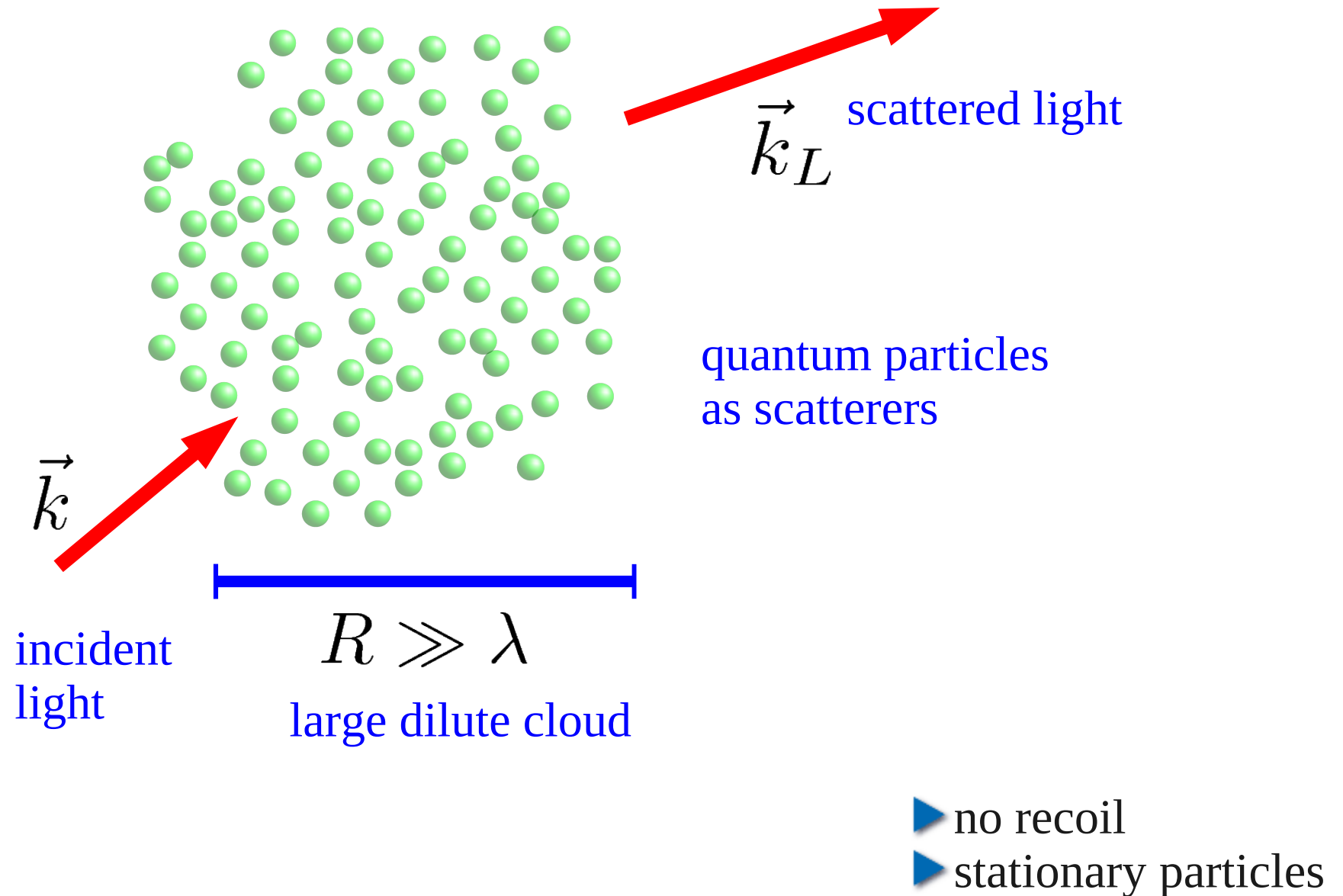
Separating signal and background

Scattered light intensity (log)

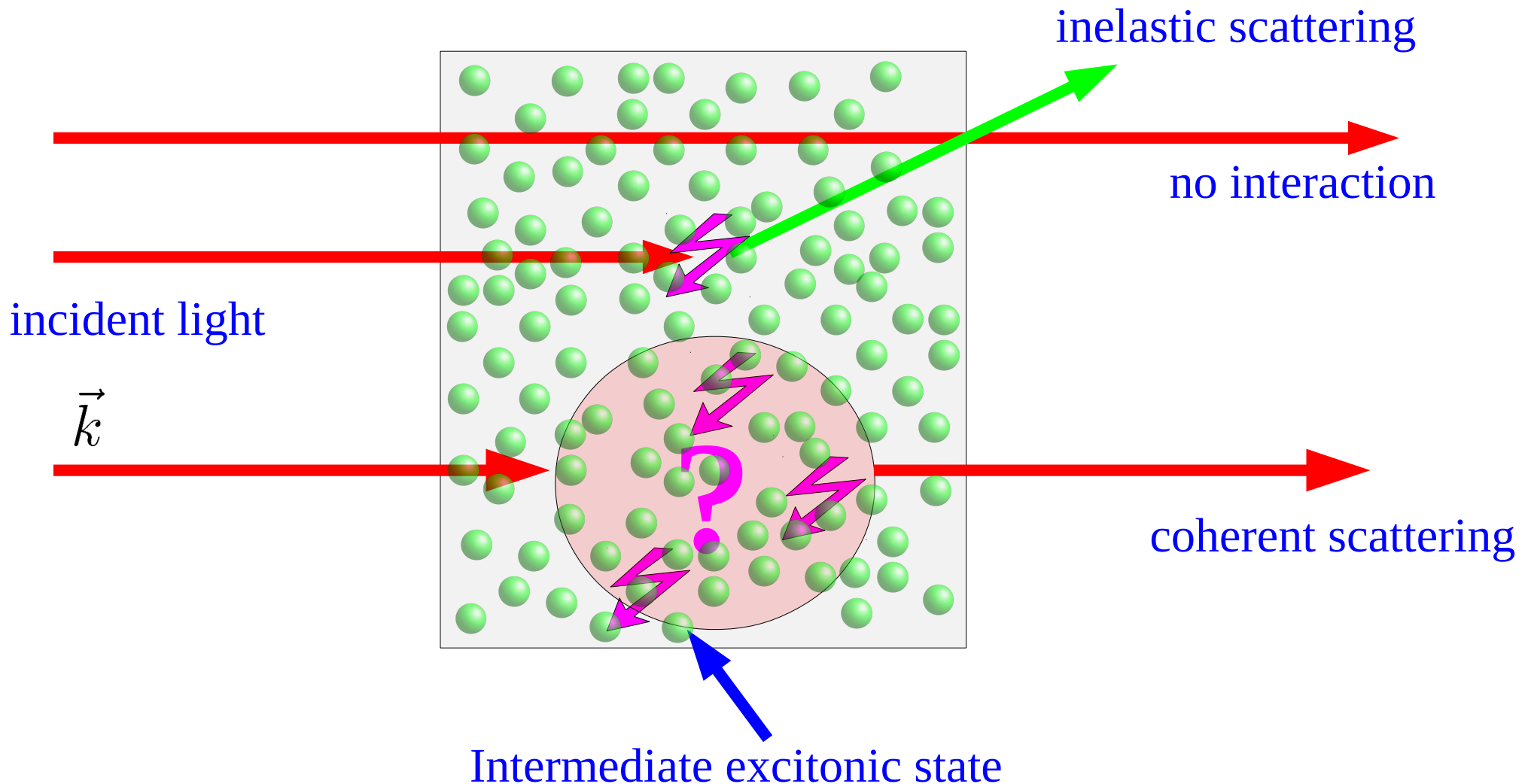


- ▶ Nuclear resonances very narrow (μeV - peV)
- ▶ Nuclear scattering has delayed tail on time scale $1/\Gamma$
Example (^{57}Fe): 141 ns
- ▶ Time-gating \rightarrow almost background-free
- ▶ Alternative methods available \rightarrow later

Cooperative light scattering

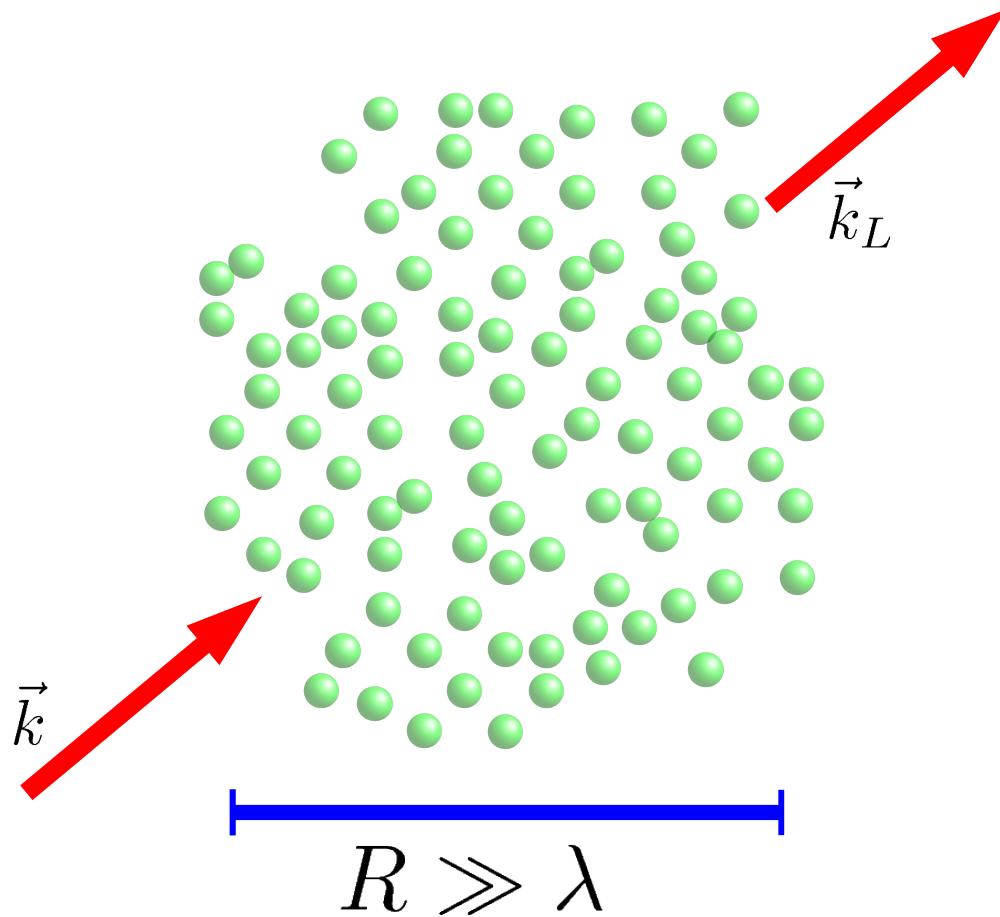


Elementary processes

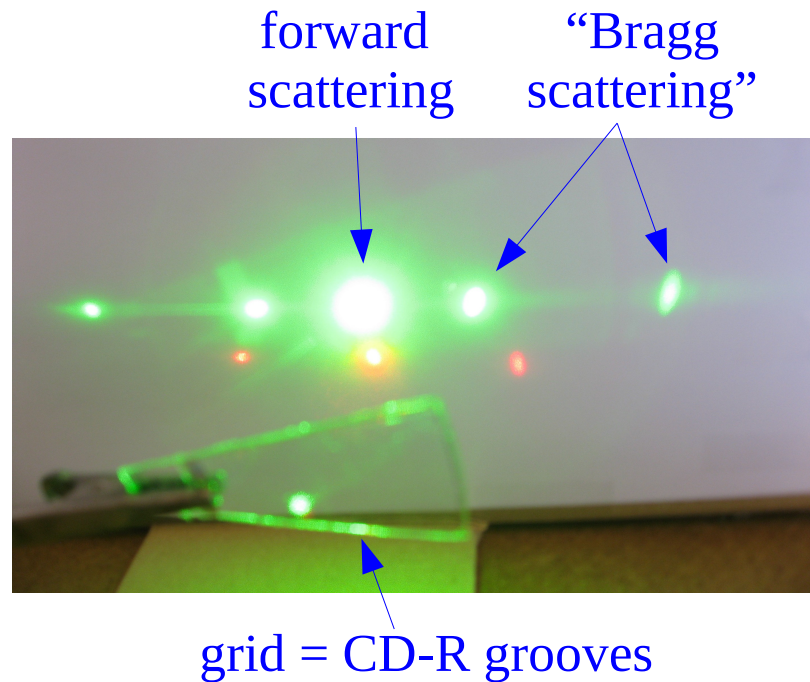


$$|\Psi\rangle = \frac{1}{\sqrt{N}} \sum_{i=1}^N e^{i\vec{k}\vec{r}_i} |g_1, \dots, g_{i-1}, e_i, g_{i+1}, \dots, g_N\rangle$$

Directionality of coherent scattering



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

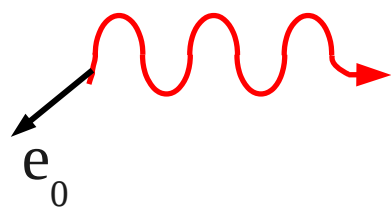


$$\lim_{N \rightarrow \infty} \sum_{i=1}^N e^{i(\vec{k} - \vec{k}_L) \cdot \vec{r}_i} \sim \delta(\vec{k} - \vec{k}_L)$$

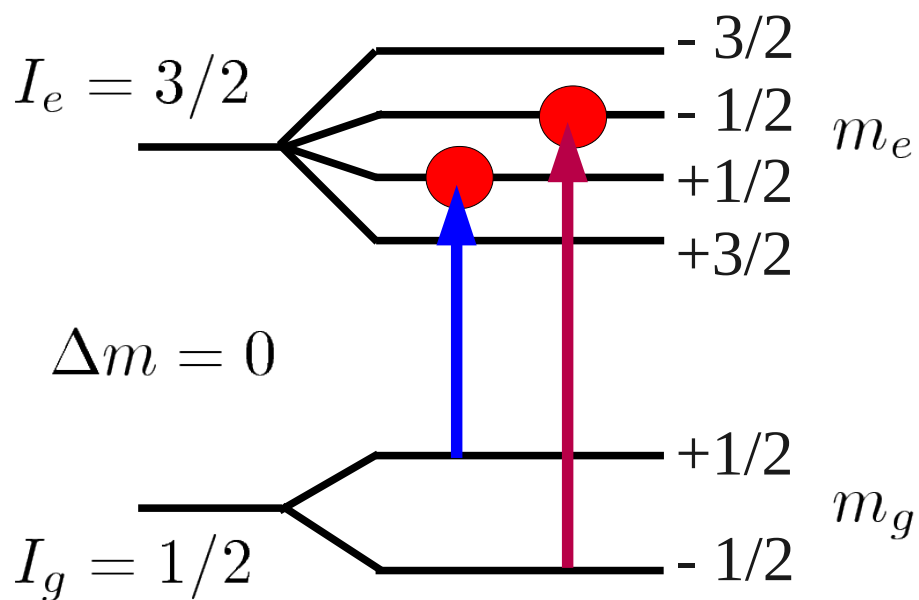
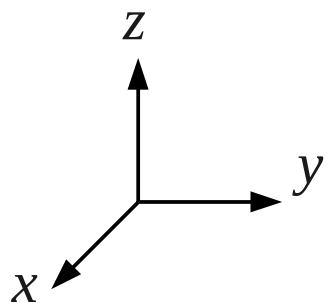
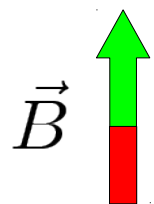
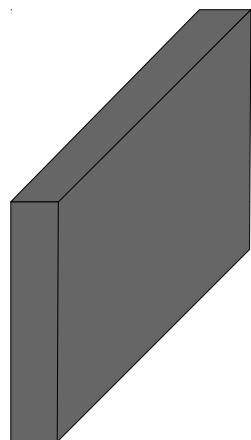
Temporal beats

^{57}Fe sample

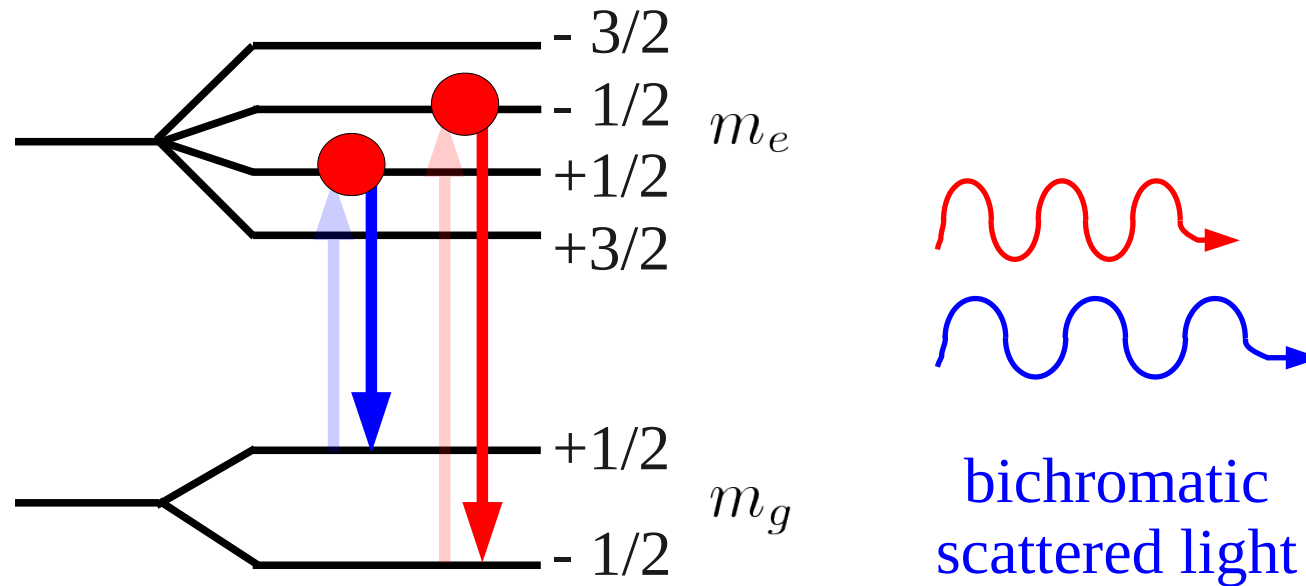
σ -polarized



“broadband”
excitation

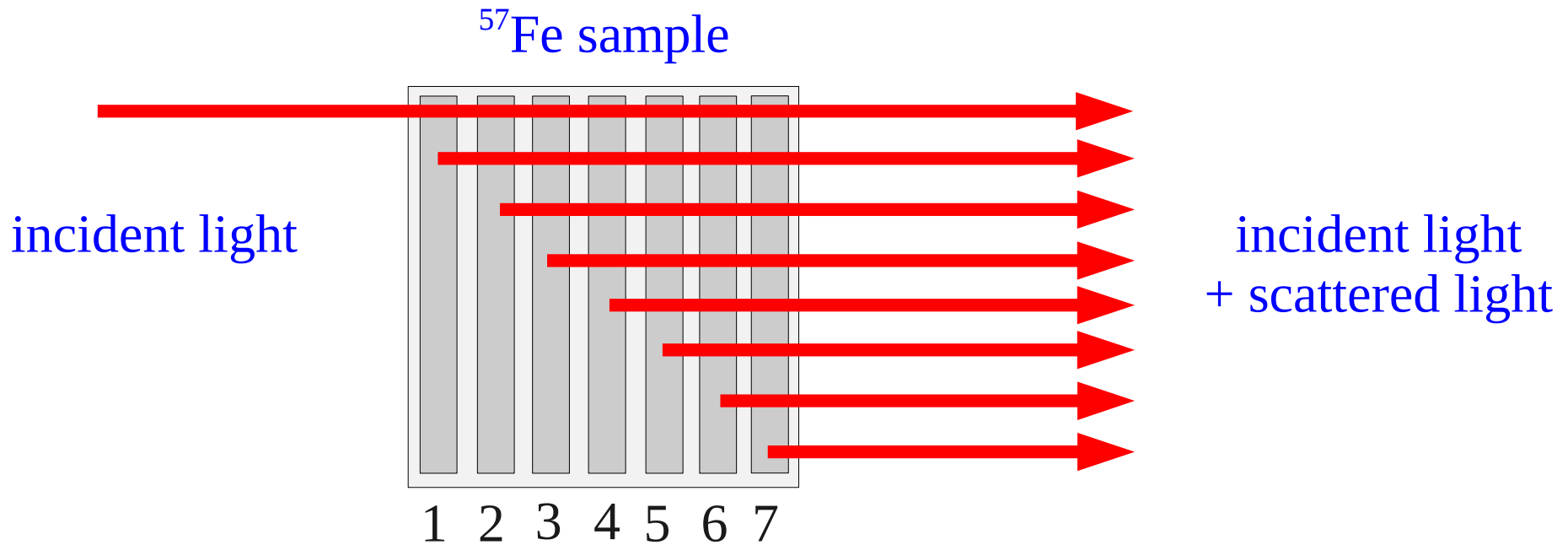


Temporal beats



- ▶ 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)

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

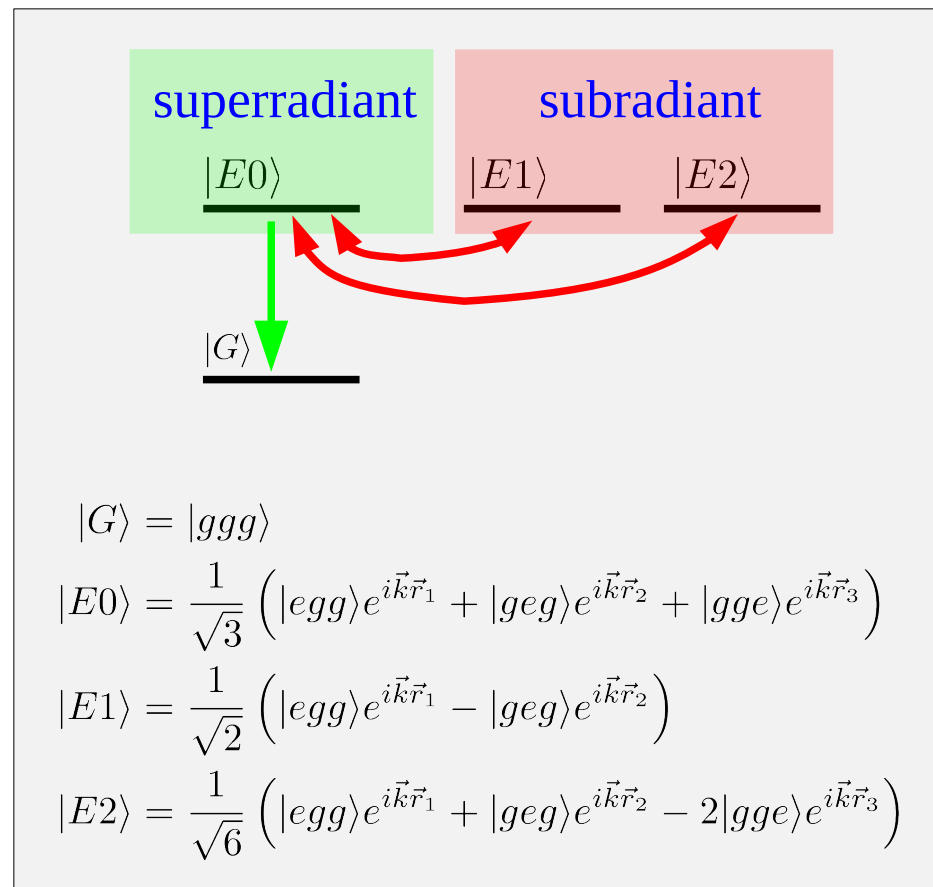
$$\langle G|\vec{d}|\Psi\rangle = \sqrt{N} \langle g_i|\vec{d}|e_i\rangle$$

$$\gamma \rightarrow N \gamma$$

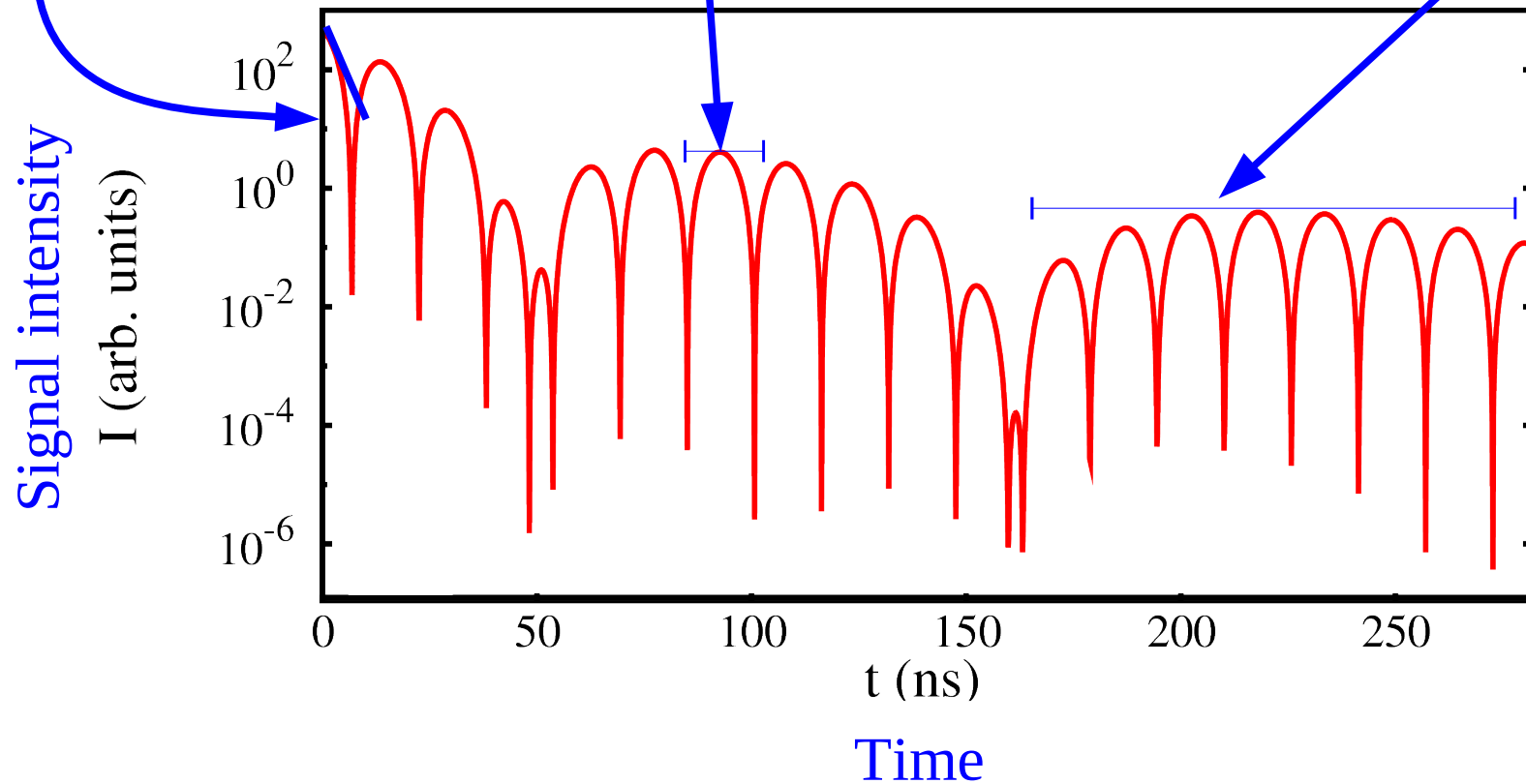
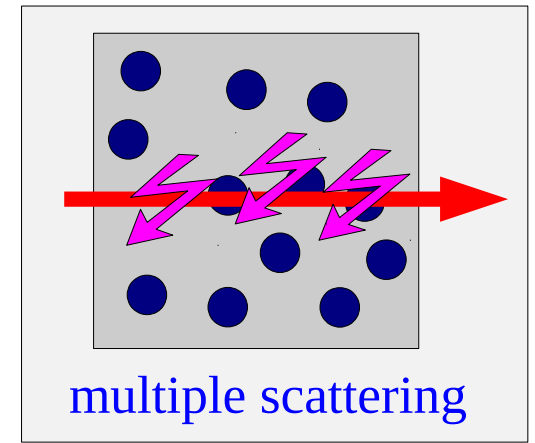
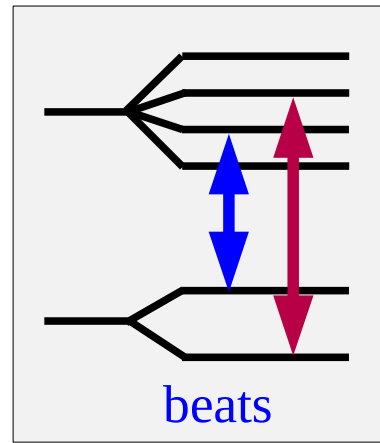
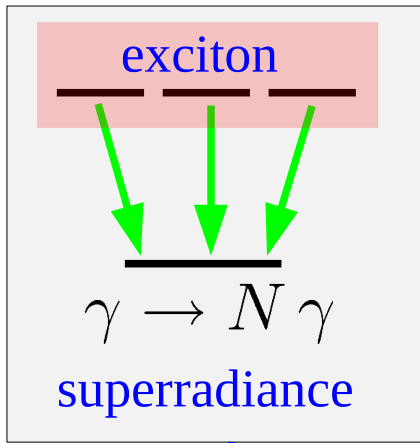
NFS case (large dilute sample)

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

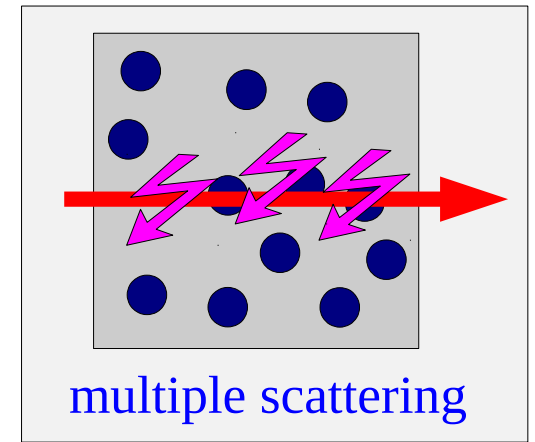
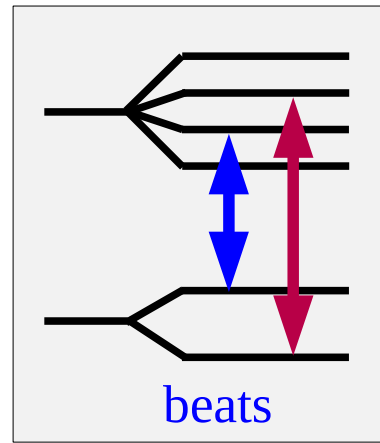
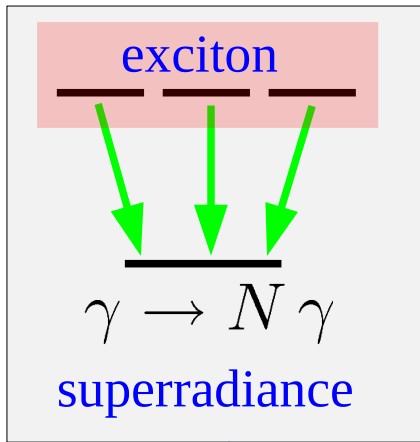
- ▶ Superradiant state dynamically coupled to subradiant states
- ▶ Imperfect preparation of superradiant state in thick samples → dephasing



Characteristic features in forward scattering



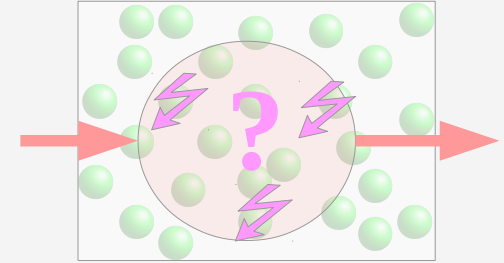
Characteristic features in forward scattering



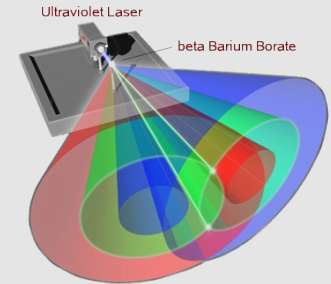
All of these features
are also intensely studied
in quantum optics
with atoms/visible light

Content

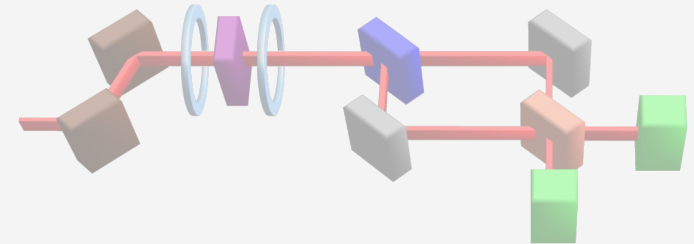
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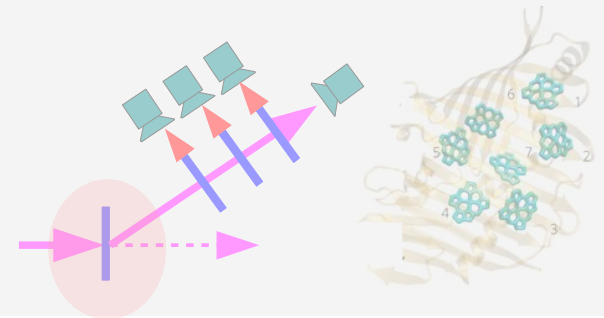
Quantum optics and information



Nuclear quantum optics



Future perspectives



Electromagnetically induced transparency

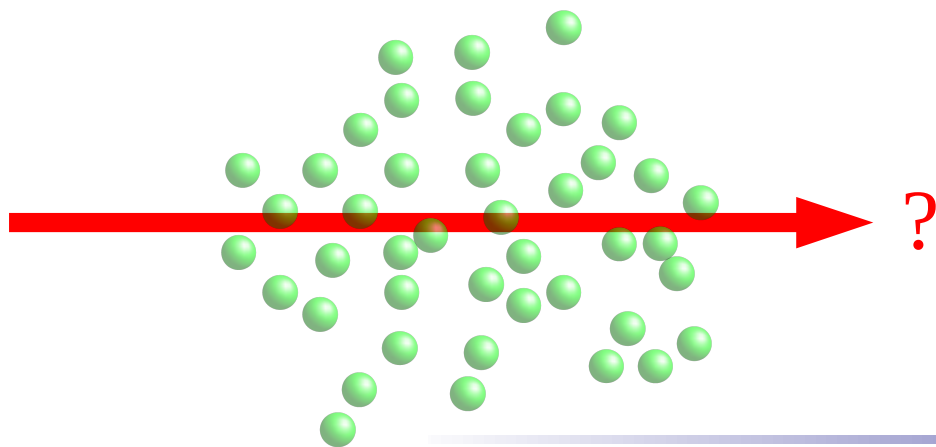
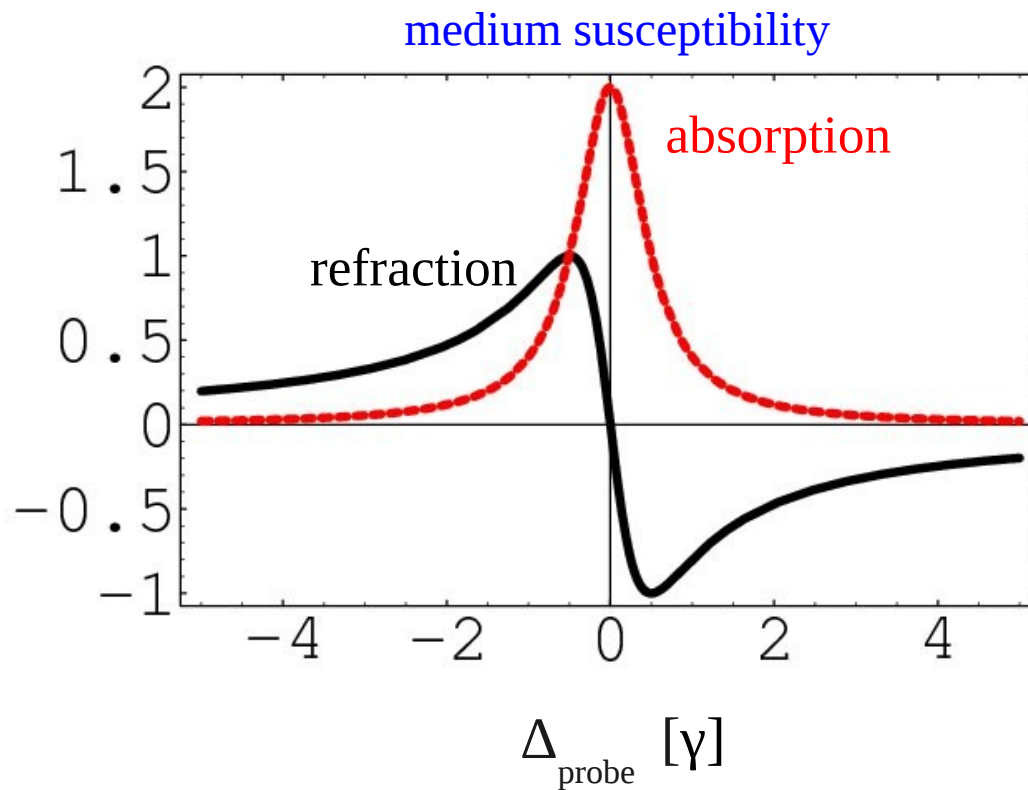
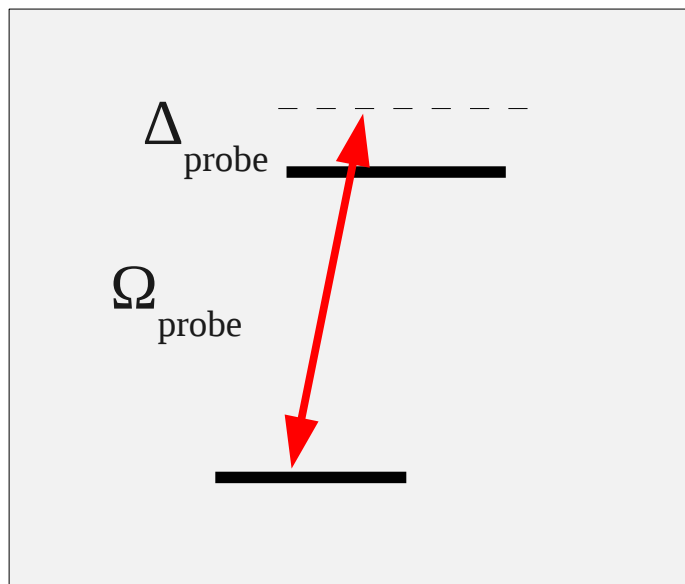


Gilt auch für
Licht

???

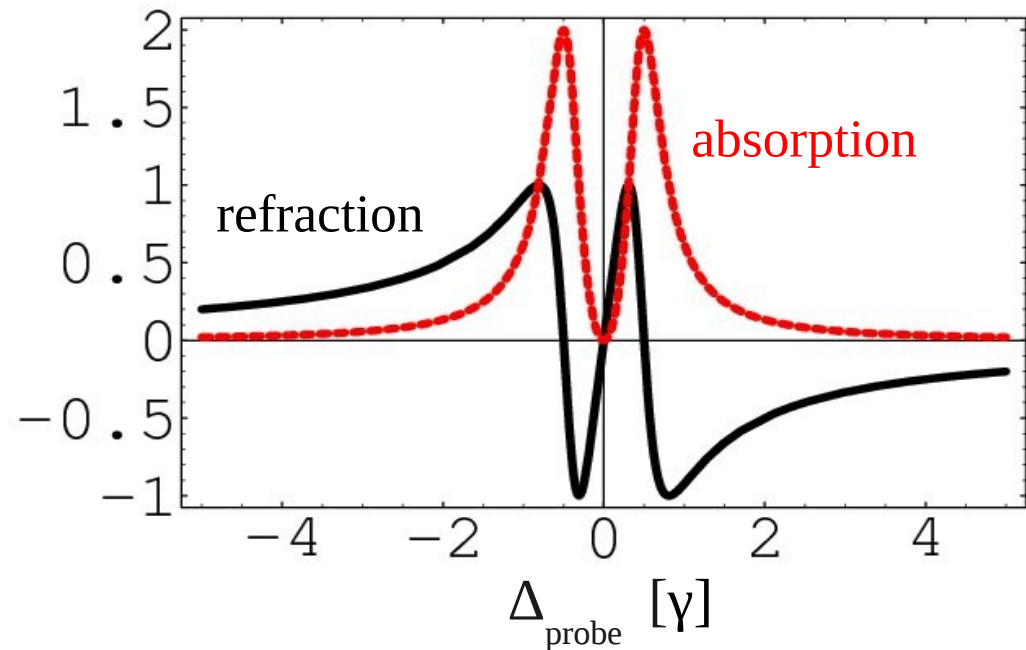
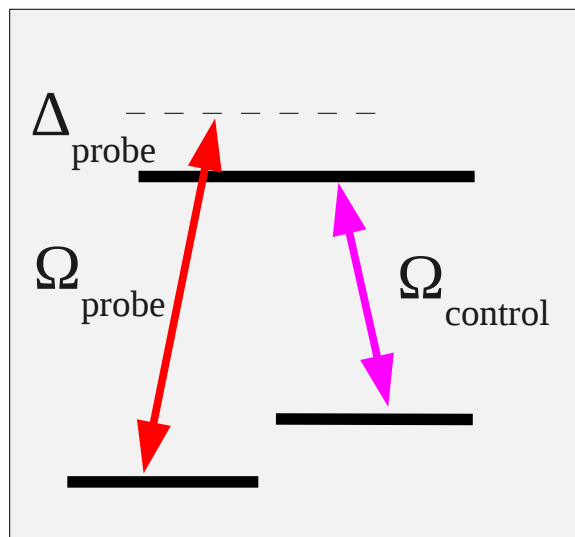


Optical response of a single resonance



Electromagnetically induced transparency

Three-level Λ system

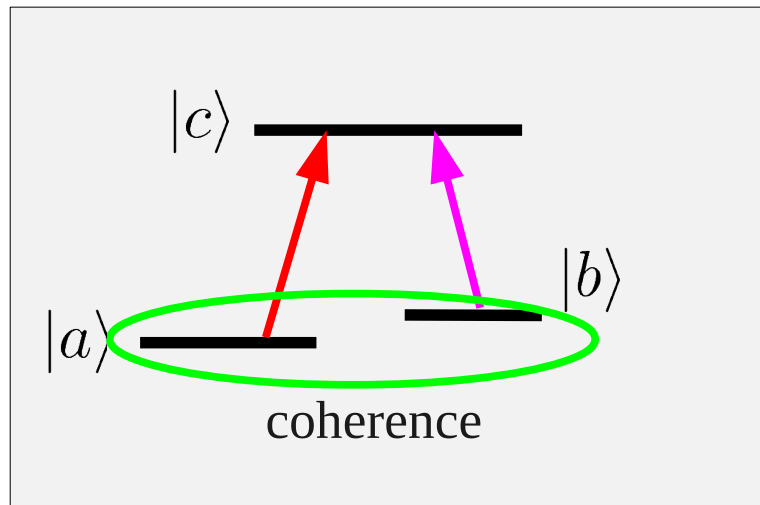


Medium is rendered transparent by shining light on it!

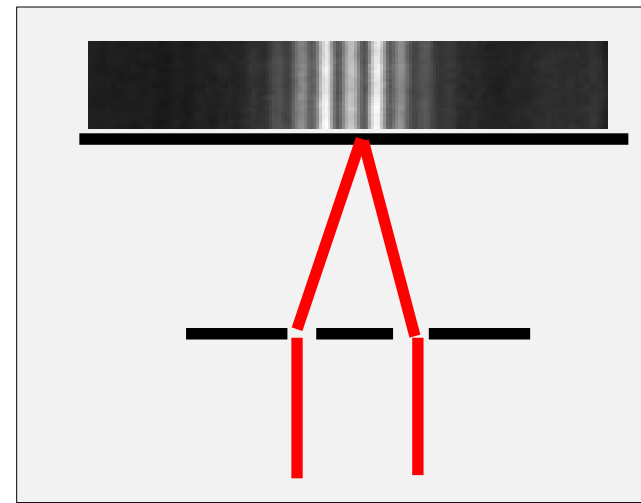
EIT is an archetype quantum optical effect with a multitude of applications

Electromagnetically induced transparency

Interpretation as coherence/interference effect:



EIT



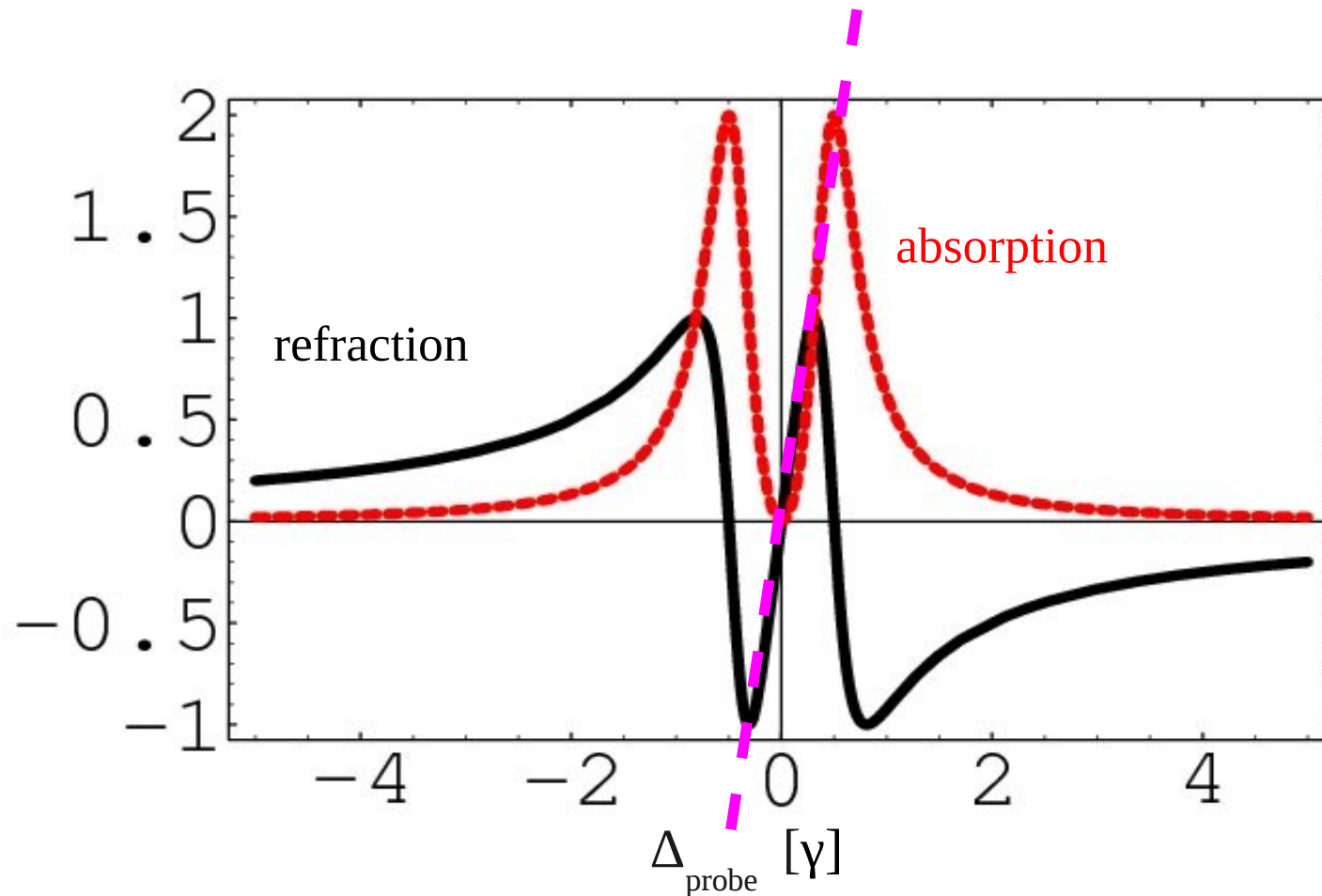
double slit

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



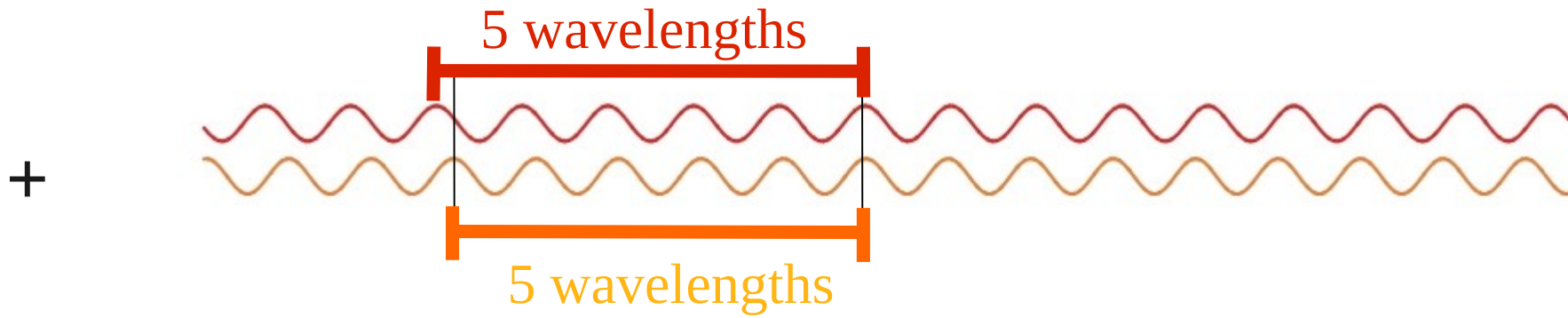
- ▶ Linear dispersion with high slope
- ▶ Low absorption

Can modify group
velocity of light

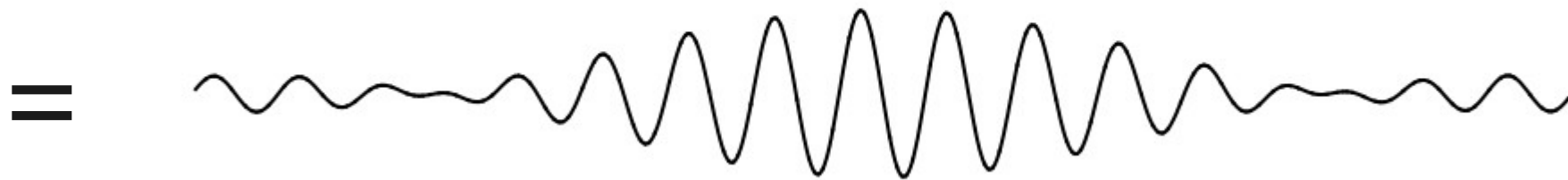
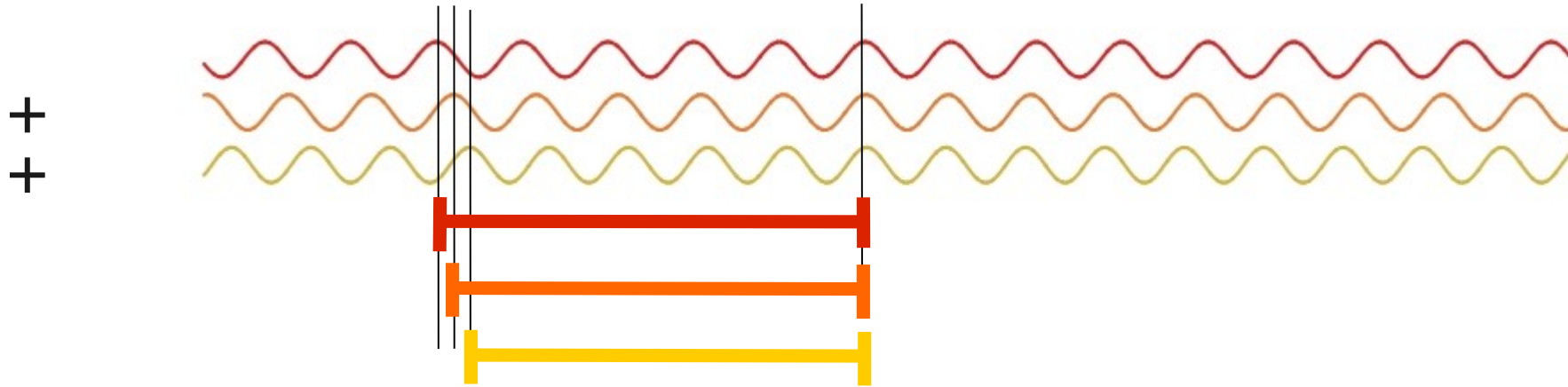
What is a light pulse?



What is a light pulse?



What is a light pulse?

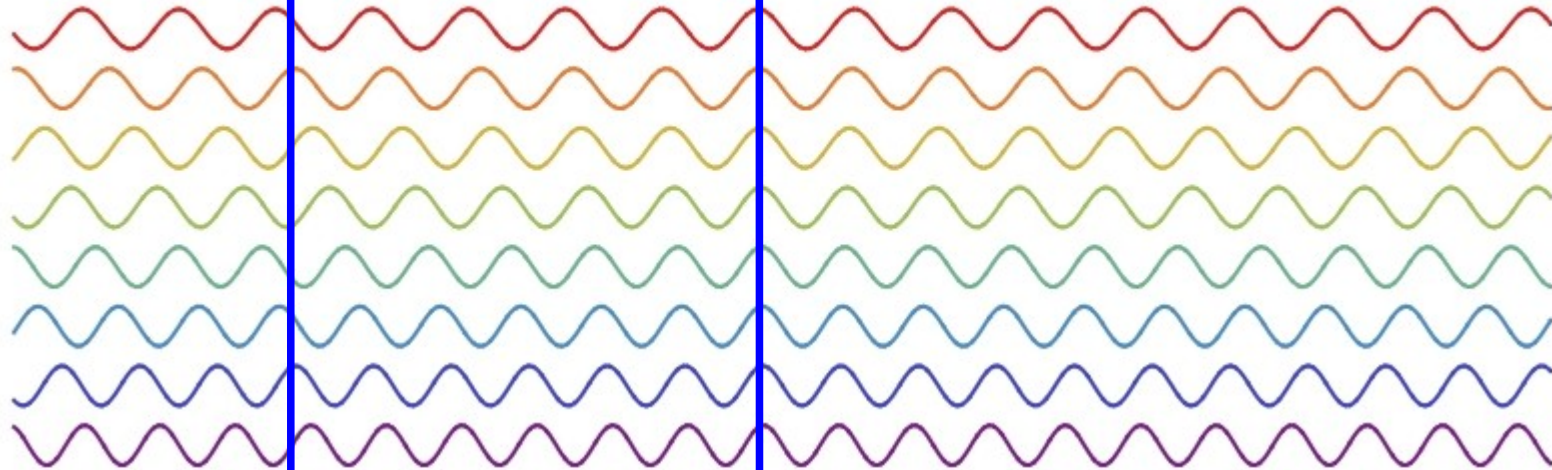


What is a light pulse?

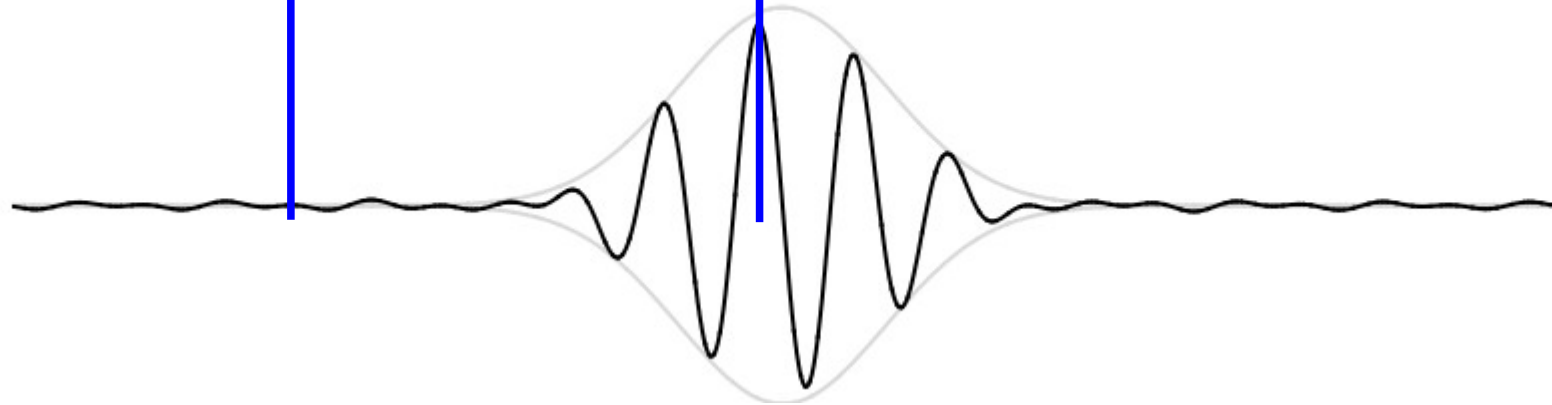
Destructive interference

Constructive interference

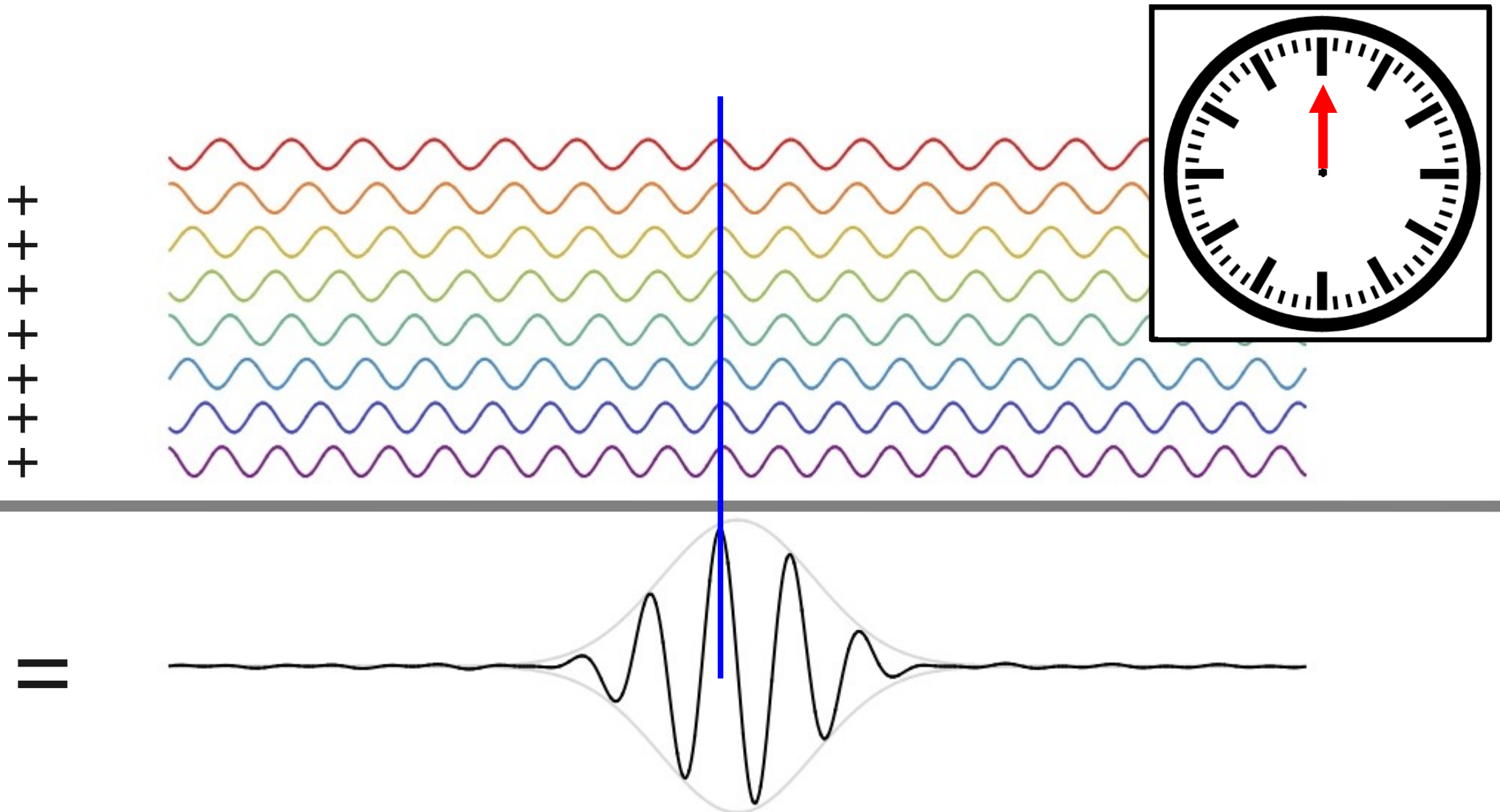
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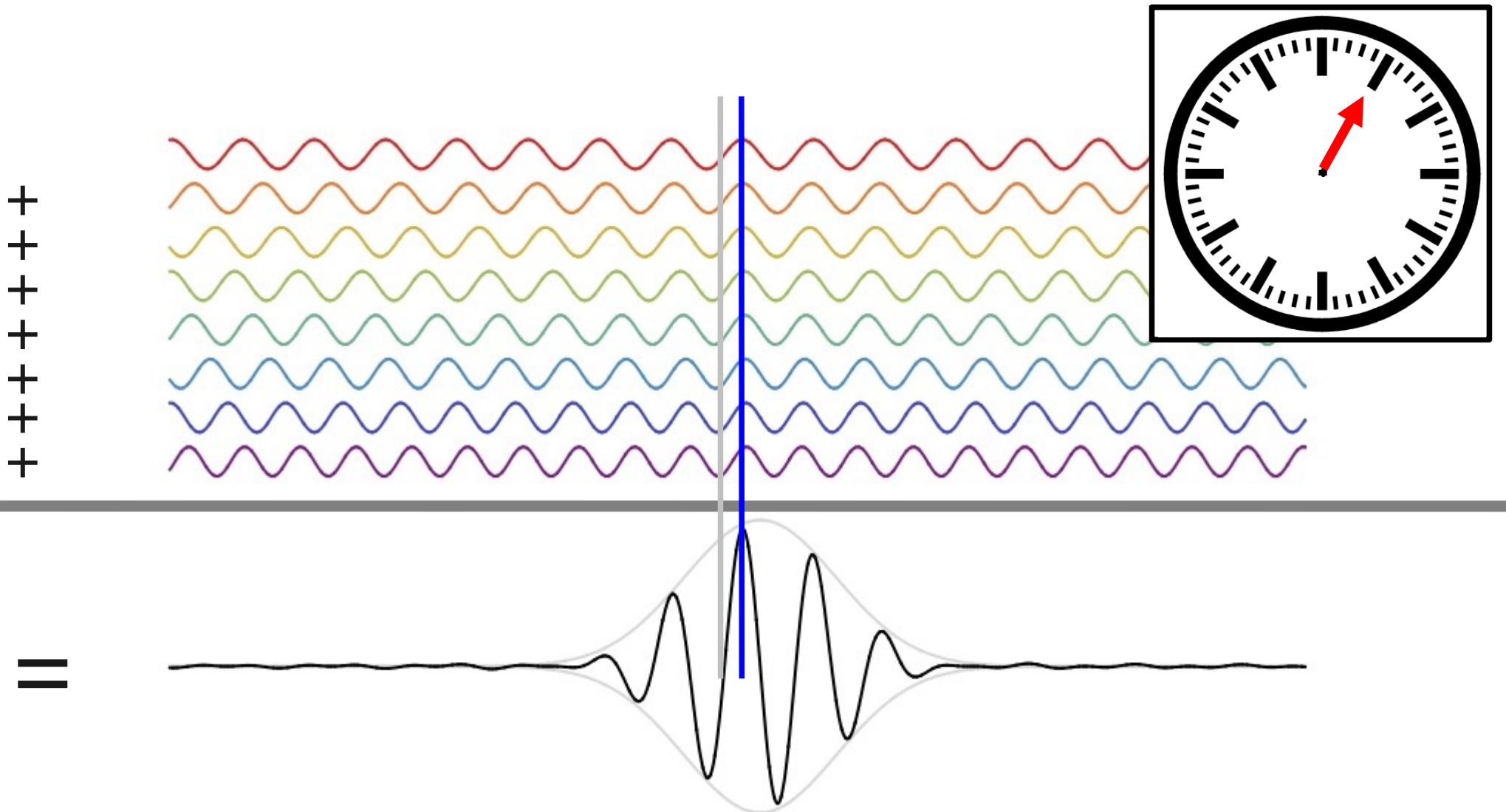


Motion in vacuum



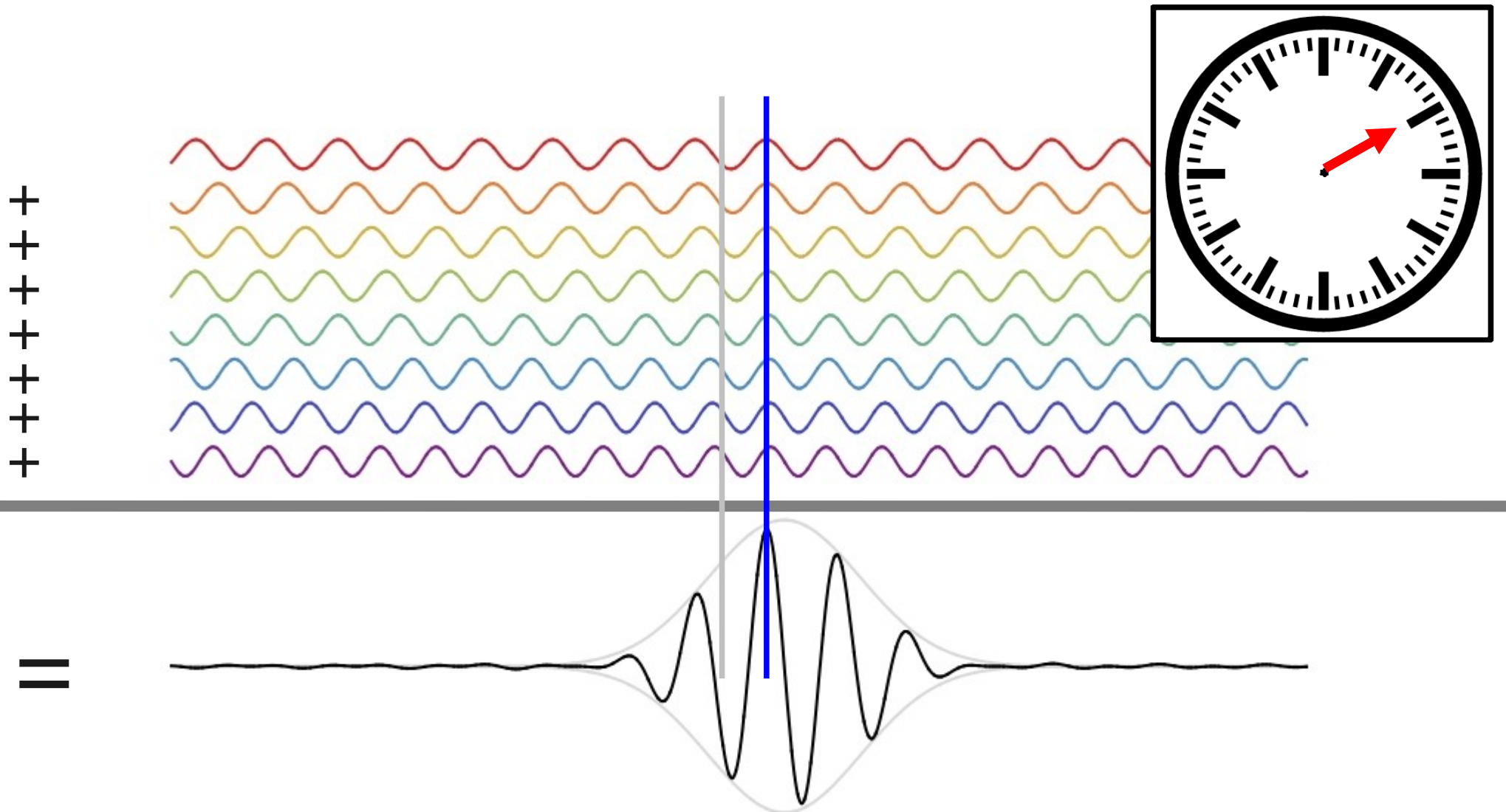
All wavelengths have the same speed

Motion in vacuum



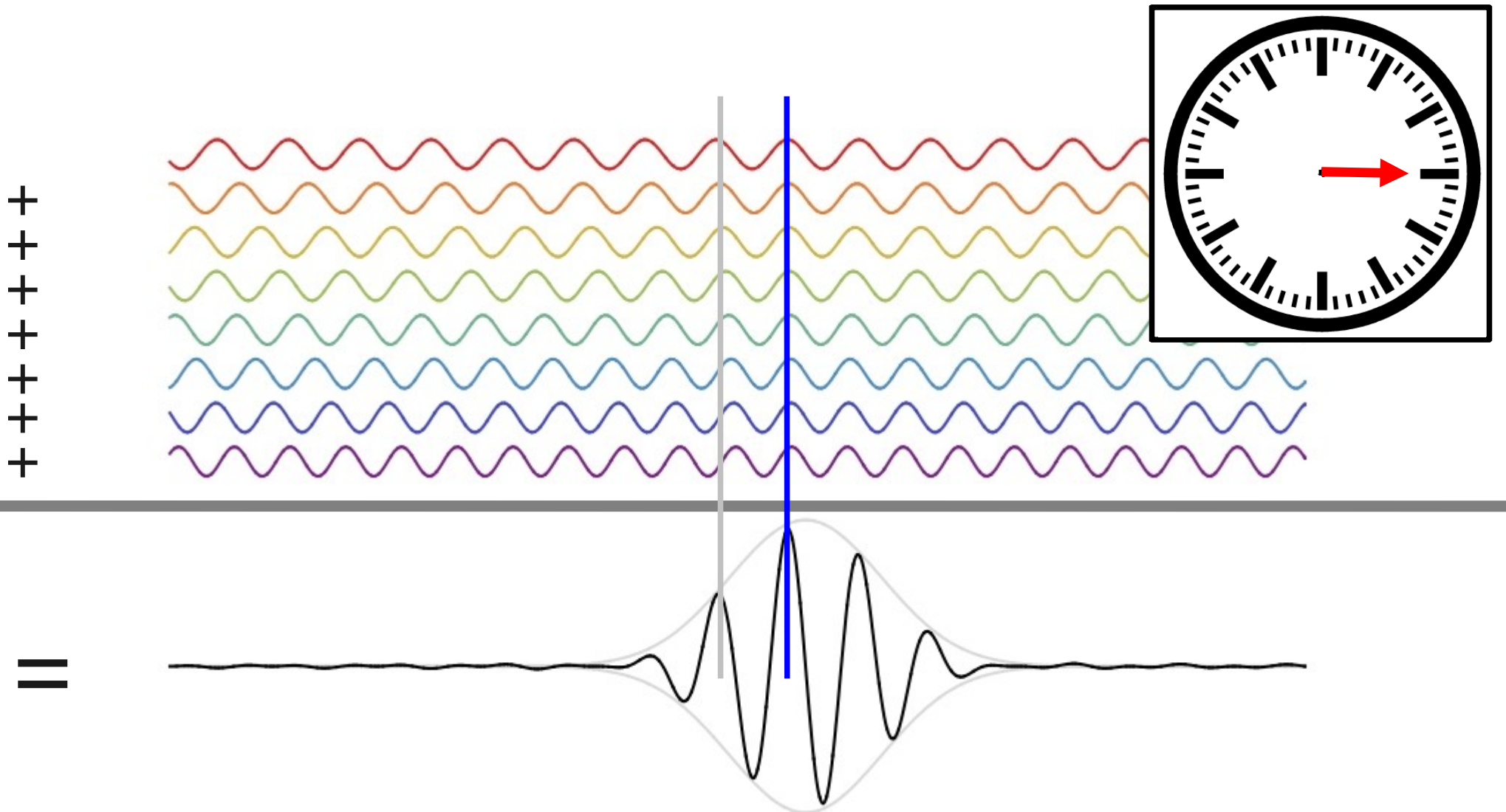
All wavelengths have the same speed

Motion in vacuum



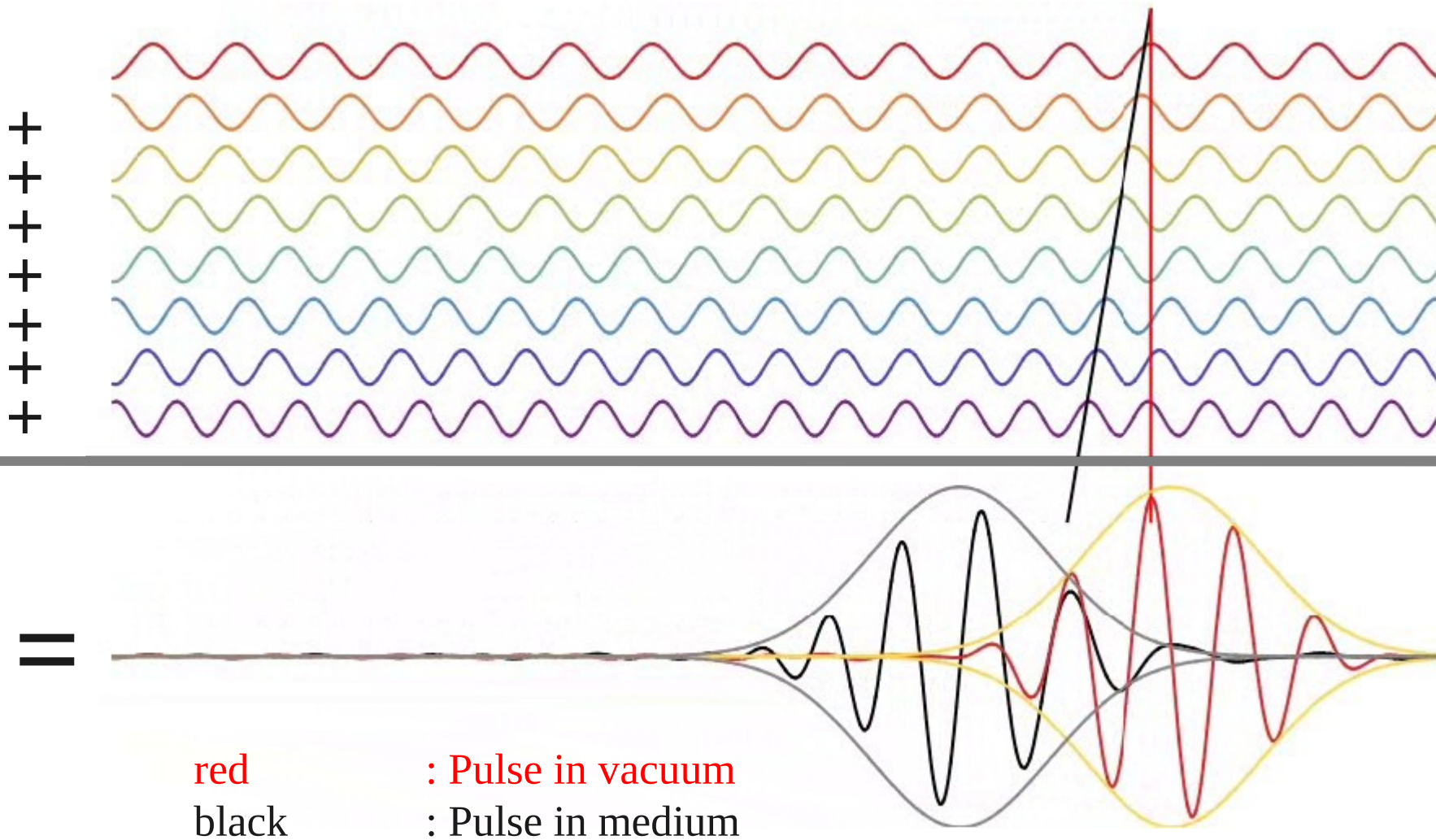
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All wavelengths have the same speed

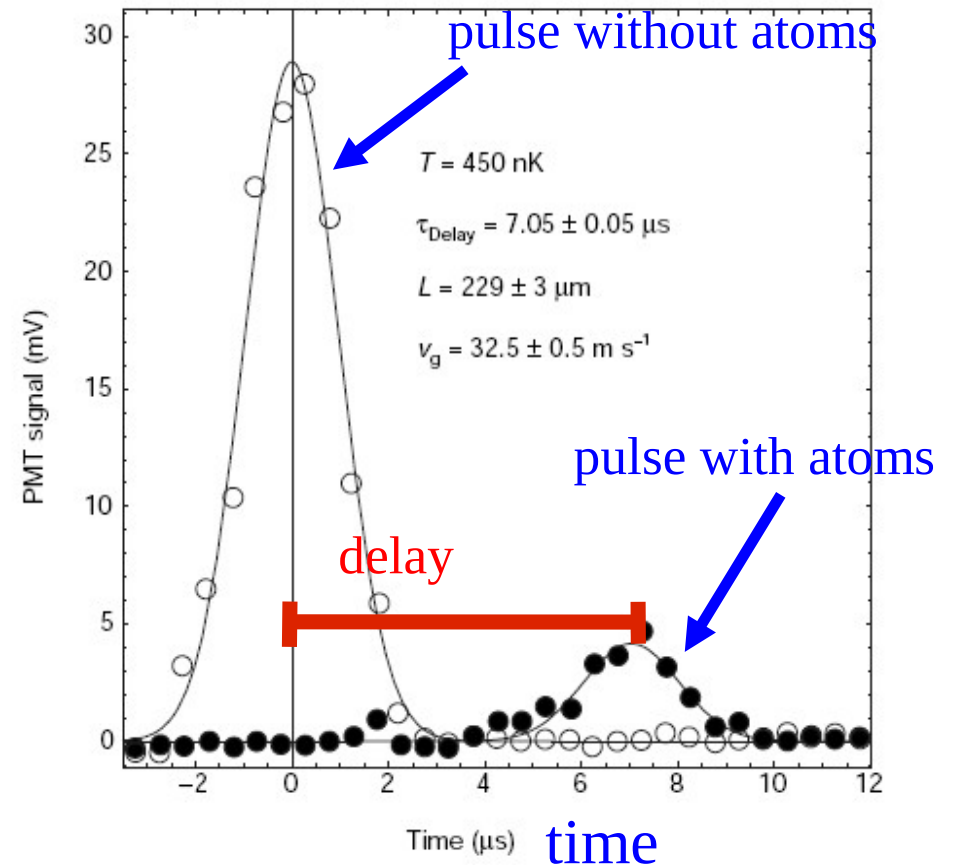
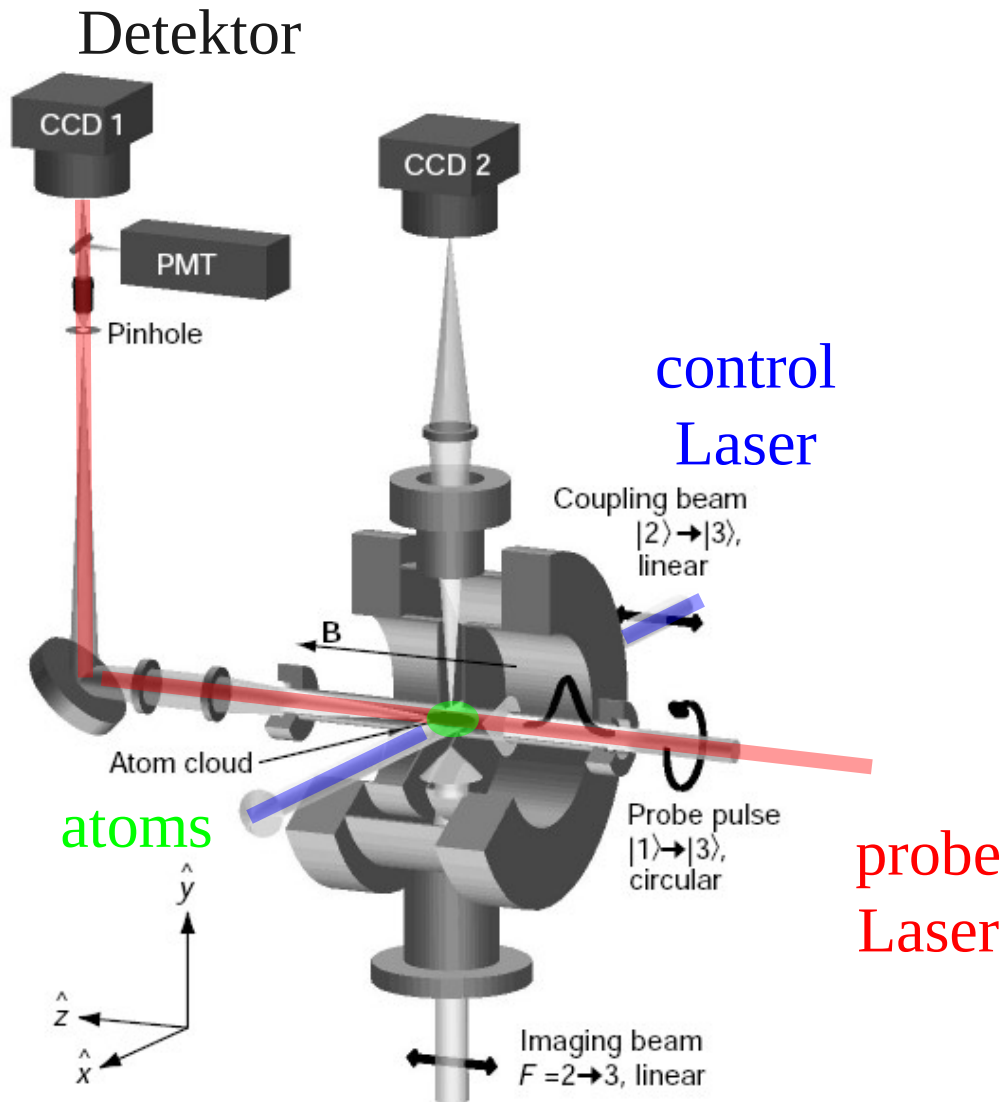
Motion in dispersive medium



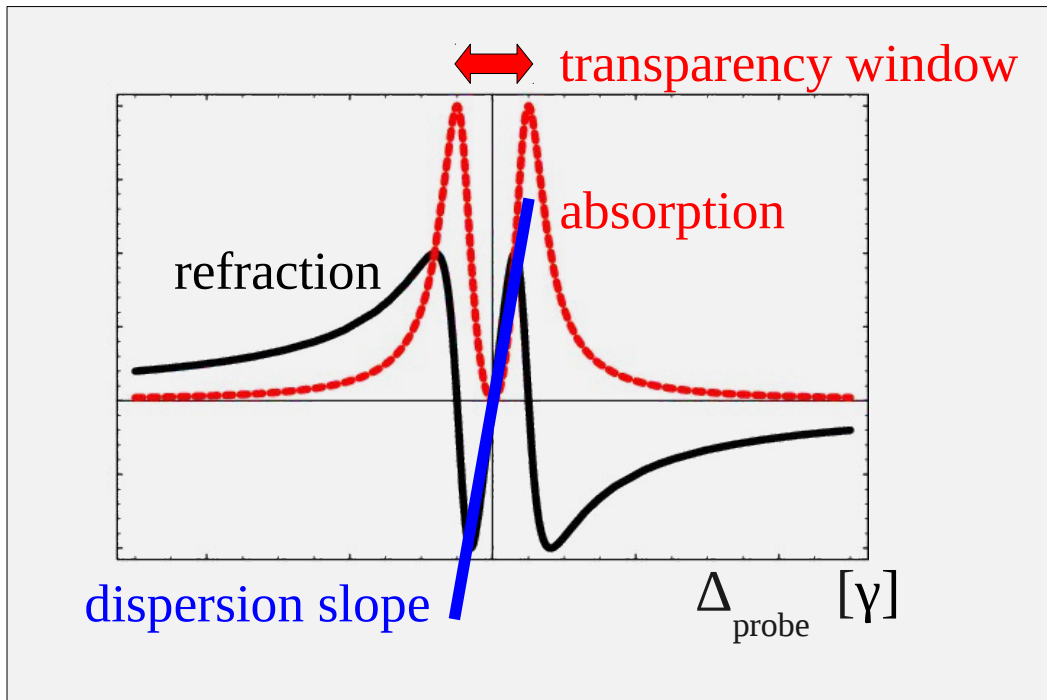
Different wavelengths have different speeds

First experiment

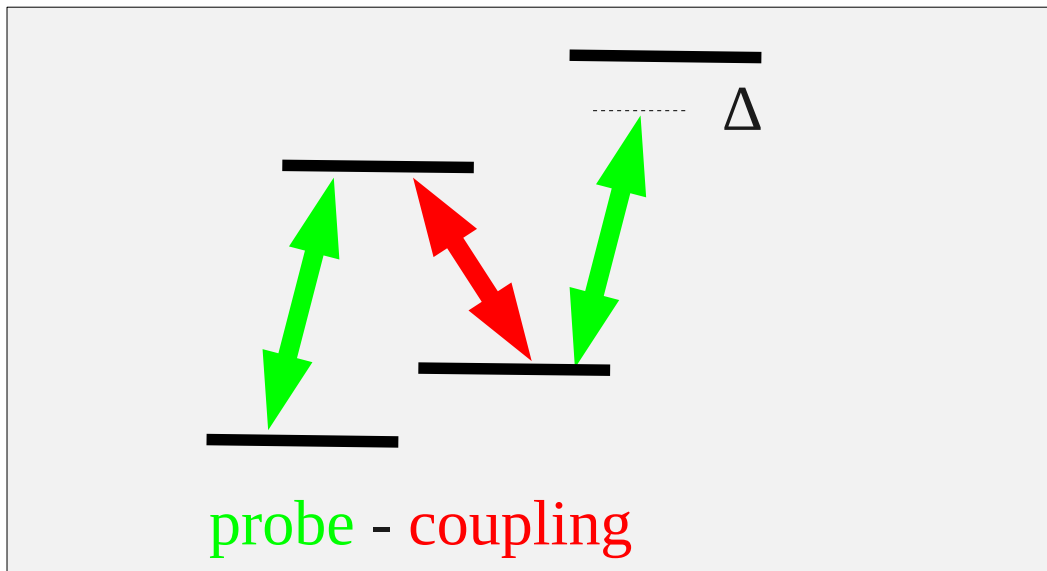
Experiment by Lene Hau (Harvard)



What would be desirable?



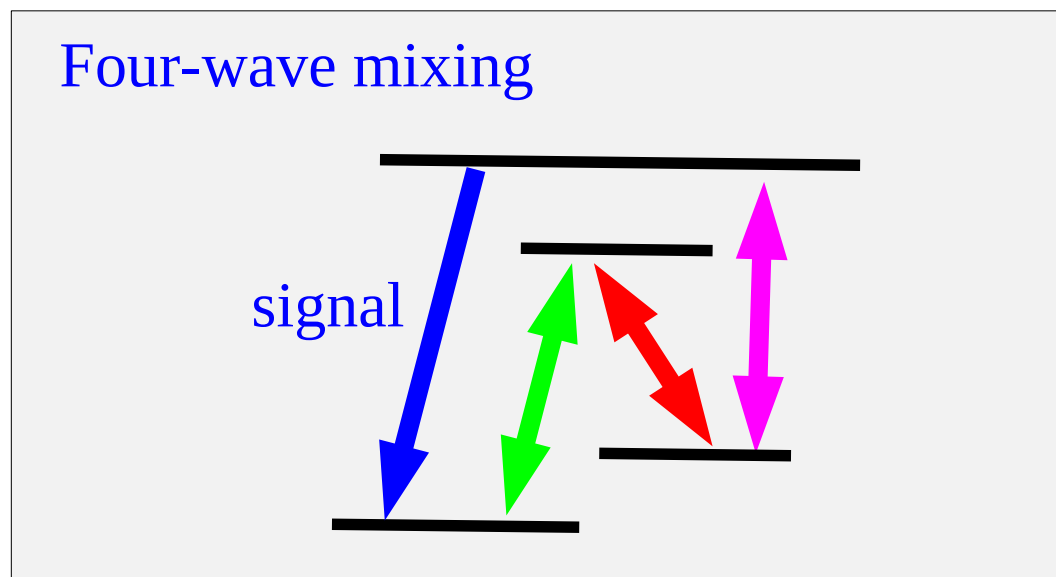
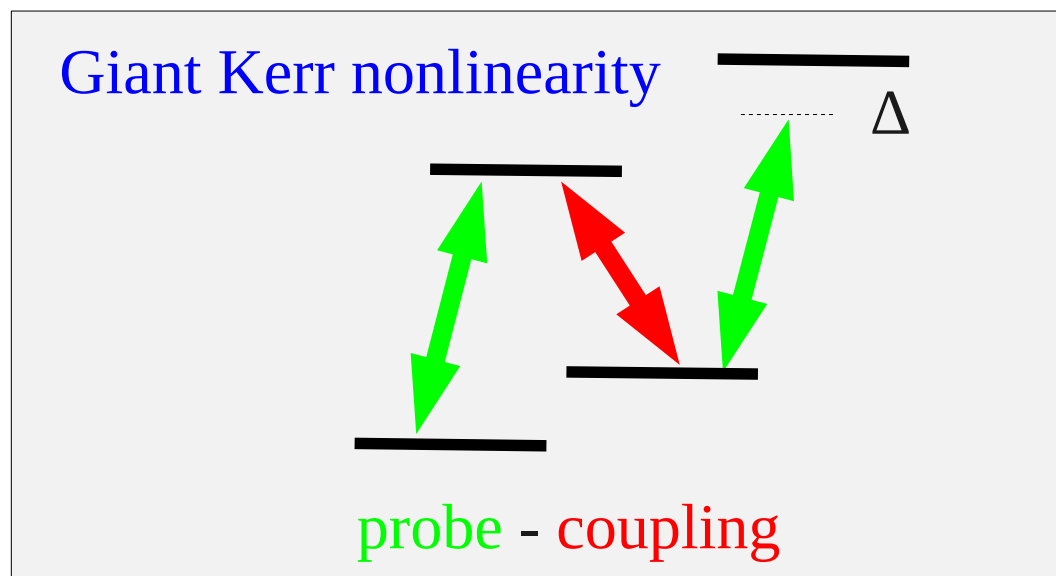
- ▶ Broad transparency window to propagate of short input pulses
- ▶ Steep dispersion slope for strong effect on propagated pulse
- ▶ (time delay)·(transparency bandwidth) is constant
→ need to tune for best trade-off



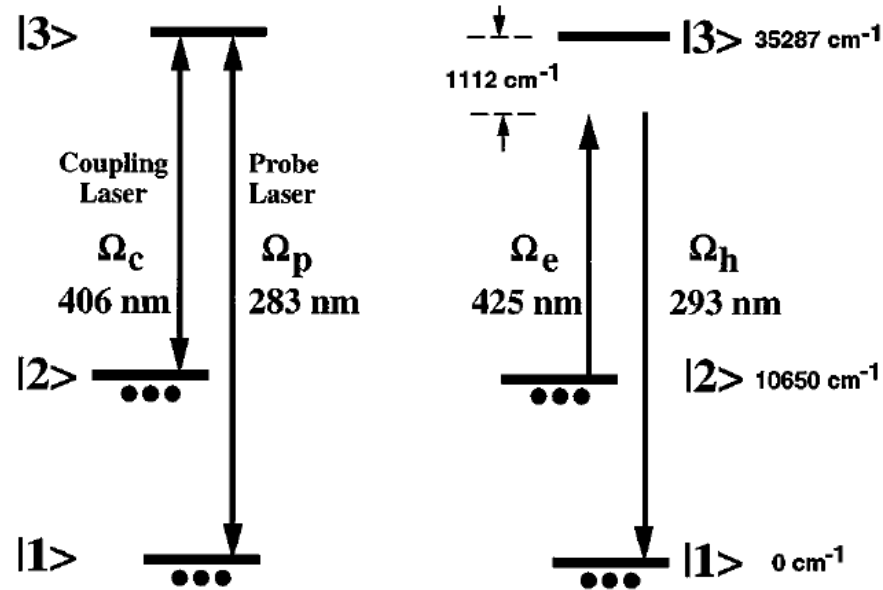
- ▶ More general level schemes offer wide range of applications
- ▶ Example: Strongly enhanced non-linear response

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

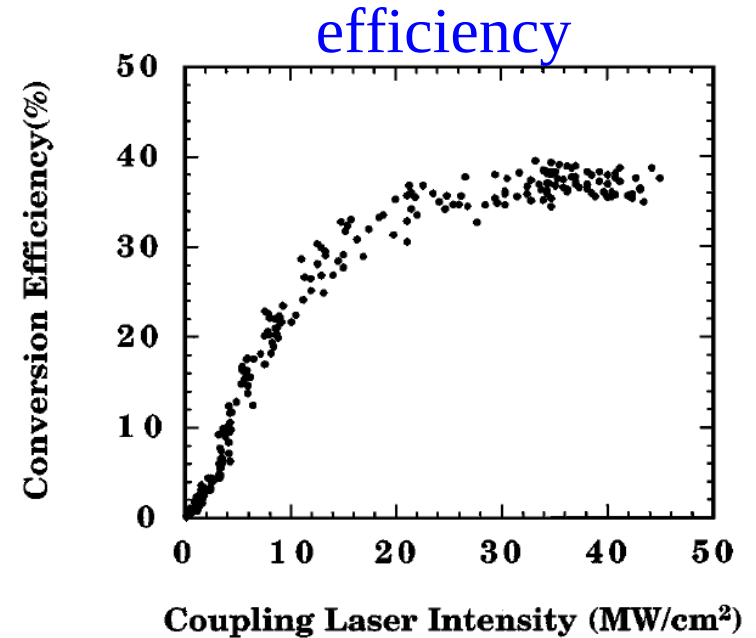


Example: Frequency conversion



coherence
preparation

conversion



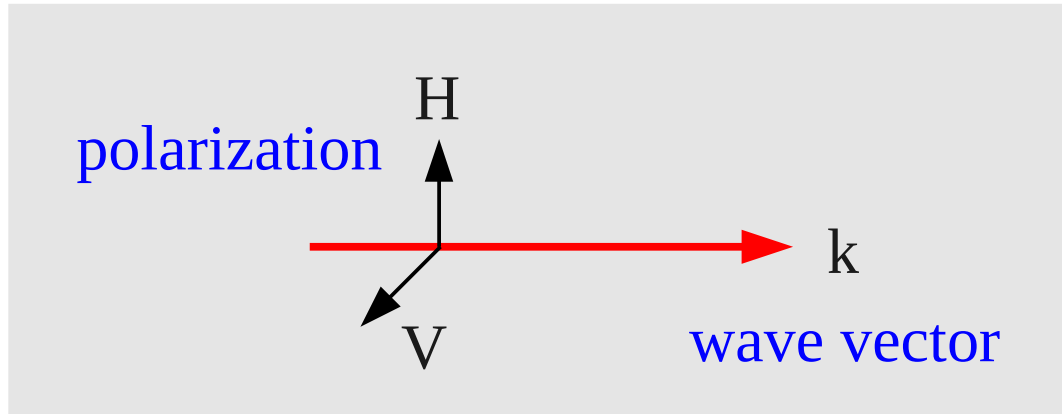
- ▶ Coherence greatly enhances conversion efficiency
- ▶ Extra coherence modifies source term in propagation equation
- ▶ Interpretation: stringent phase matching conditions are alleviated

Entanglement



Quantum mechanical superposition principle

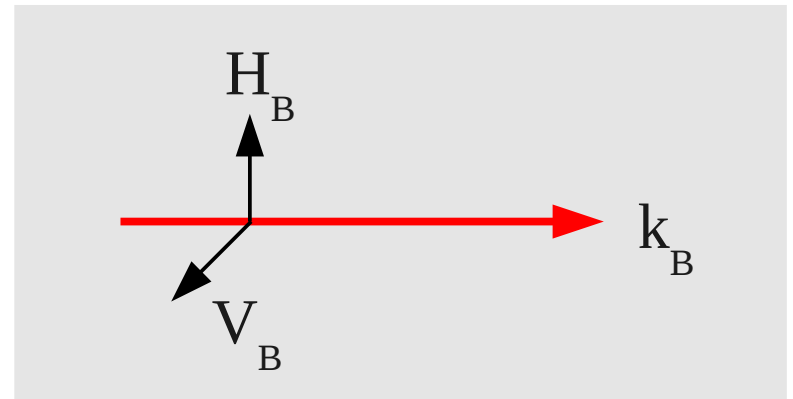
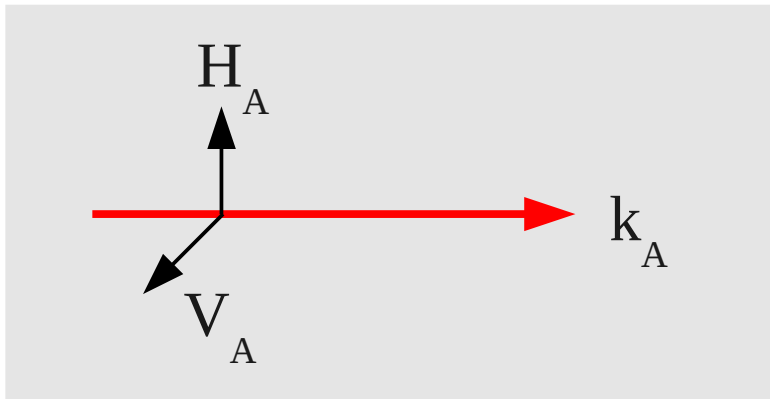
Consider a single photon:



- ▶ 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

$$\begin{aligned} |HH\rangle &= |H_A\rangle|H_B\rangle, & |VH\rangle &= |V_A\rangle|H_B\rangle, \\ |HV\rangle &= |H_A\rangle|V_B\rangle, & |VV\rangle &= |V_A\rangle|V_B\rangle \end{aligned}$$

- ▶ 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

$$\{|i\rangle_A | i = 1, 2, \dots \} \quad \text{and} \quad \{|j\rangle_B | j = 1, 2, \dots \}$$

- ▶ 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

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

$$\begin{aligned} |\Psi\rangle &= |HH\rangle - |VH\rangle - |HV\rangle + |VV\rangle \\ &= (|H\rangle_A - |V\rangle_A)(|H\rangle_B - |V\rangle_B) \end{aligned}$$

$$|\Psi\rangle = |HV\rangle + |VH\rangle$$

$$|\Psi\rangle = |HH\rangle + |VV\rangle$$

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

Separable – Entangled - ???

Measurements on entangled states

- ▶ Suppose the two photons are in the entangled state

$$|\Psi\rangle = |HV\rangle + |VH\rangle$$

- ▶ 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)
-

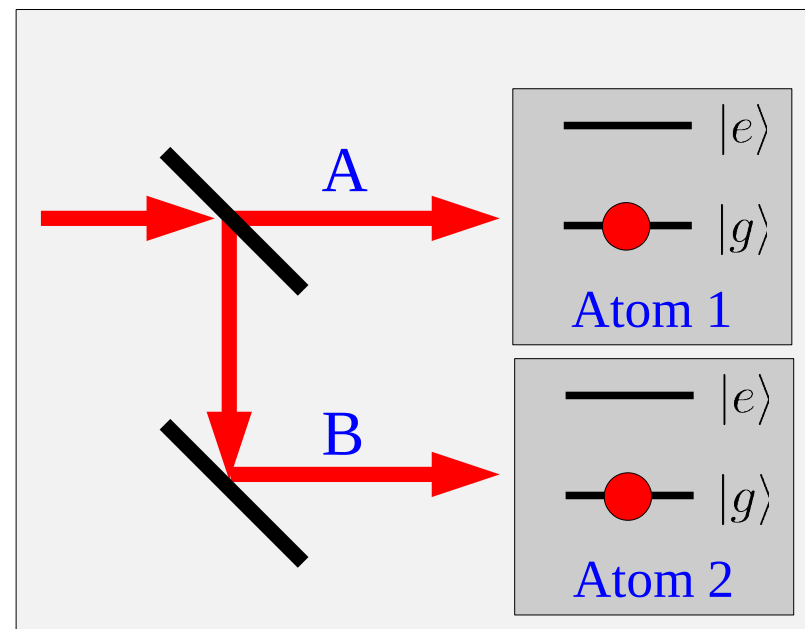
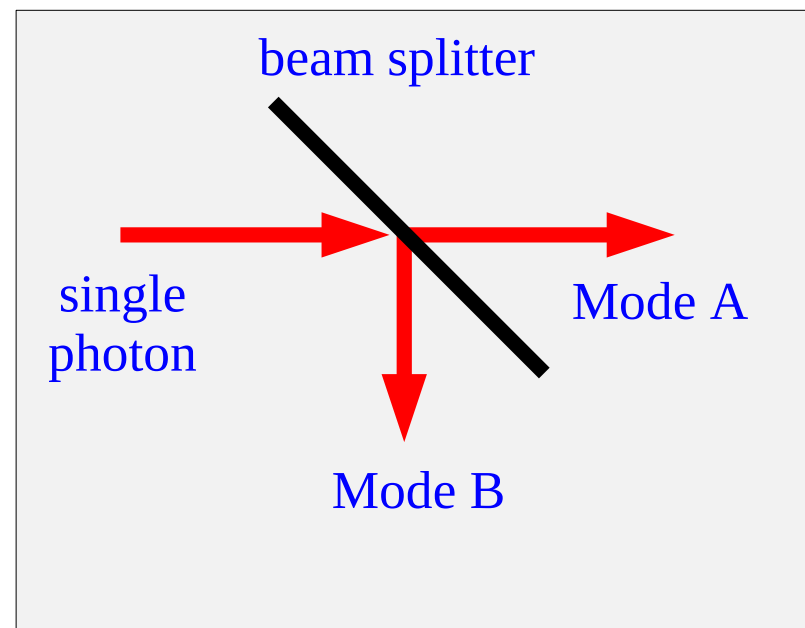
Mode (single photon) entanglement

- ▶ 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 resource for all quantum information/communication protocols
 - ▶ Measure and structure beyond “classical” or standard quantum limit
-

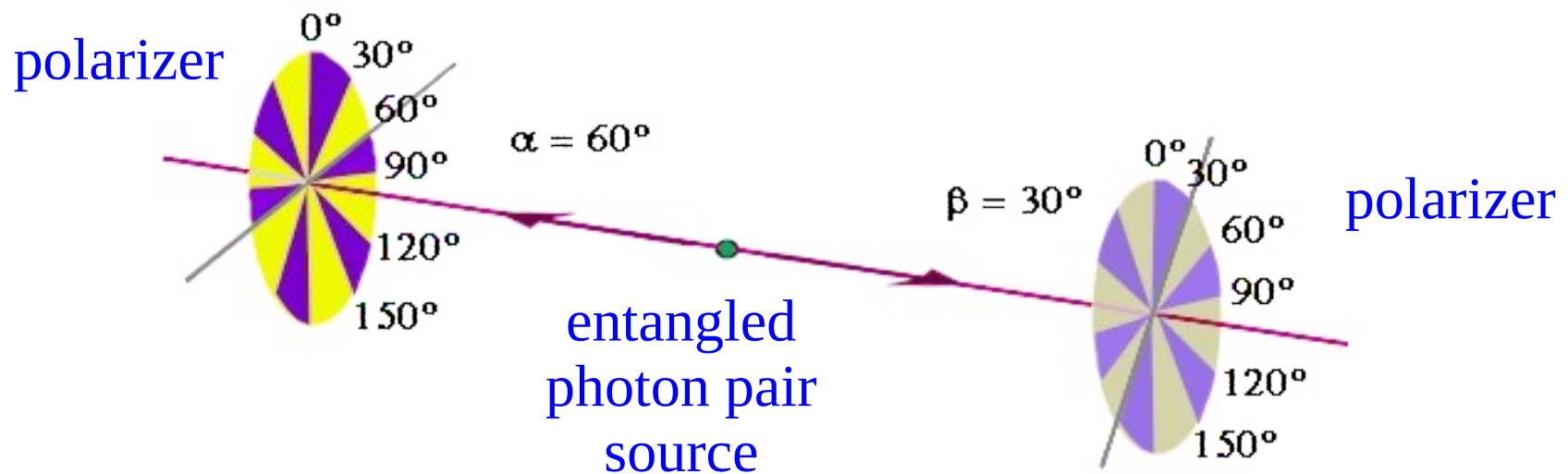
Foundations: The EPR argument

A. Einstein, B. Podolsky, N. Rosen, Phys. Rev. 47, 777 (1935)

- ▶ 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 \Rightarrow 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: $> 100\text{km}$ separation)

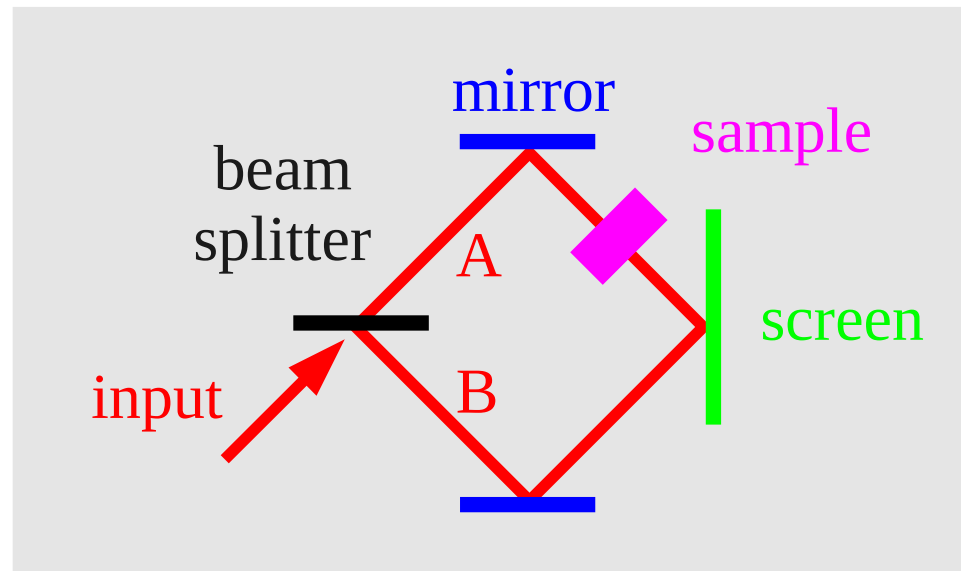
How might x-rays help?

- ▶ 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!



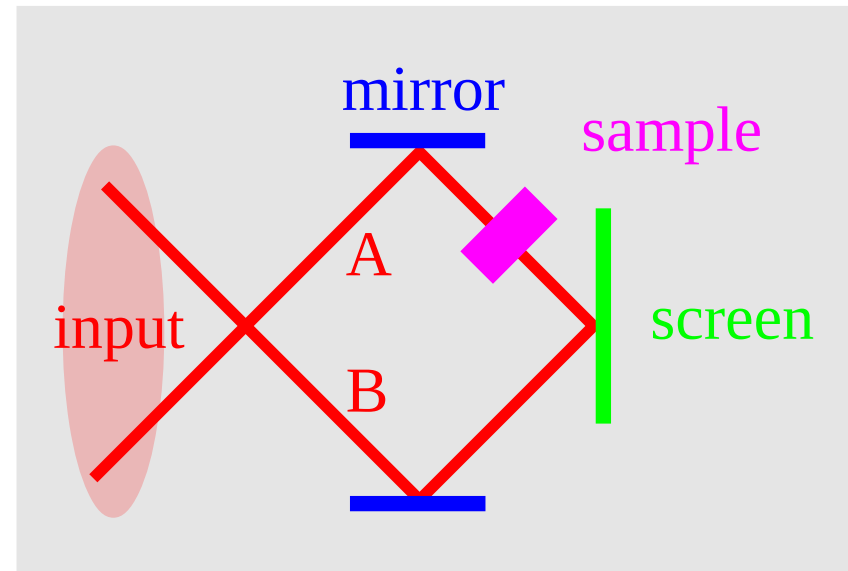
$$\begin{array}{l} \text{input } |1\rangle \xrightarrow{\text{beam splitter}} |1\rangle_A |0\rangle_B + |0\rangle_A |1\rangle_B \\ \text{sample} \xrightarrow{\quad\quad\quad} |1\rangle_A |0\rangle_B e^{i\phi} + |0\rangle_A |1\rangle_B \end{array}$$

N00N states

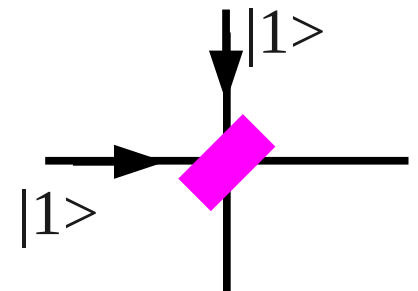
$$|N\rangle_A |0\rangle_B + |0\rangle_A |N\rangle_B$$

sample

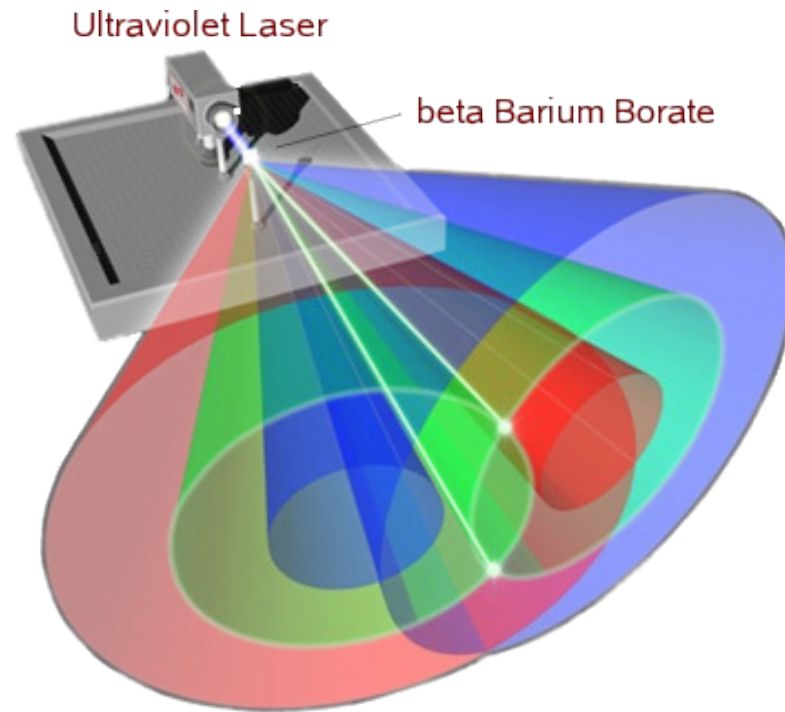
$$\rightarrow |N\rangle_A |0\rangle_B e^{iN\phi} + |0\rangle_A |N\rangle_B$$



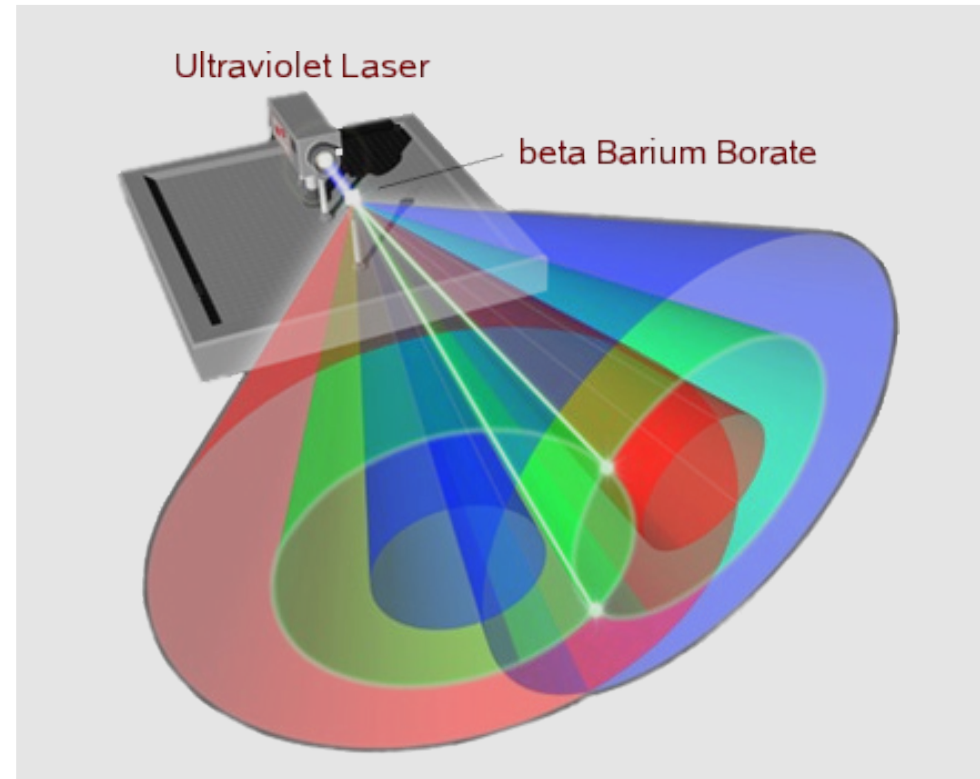
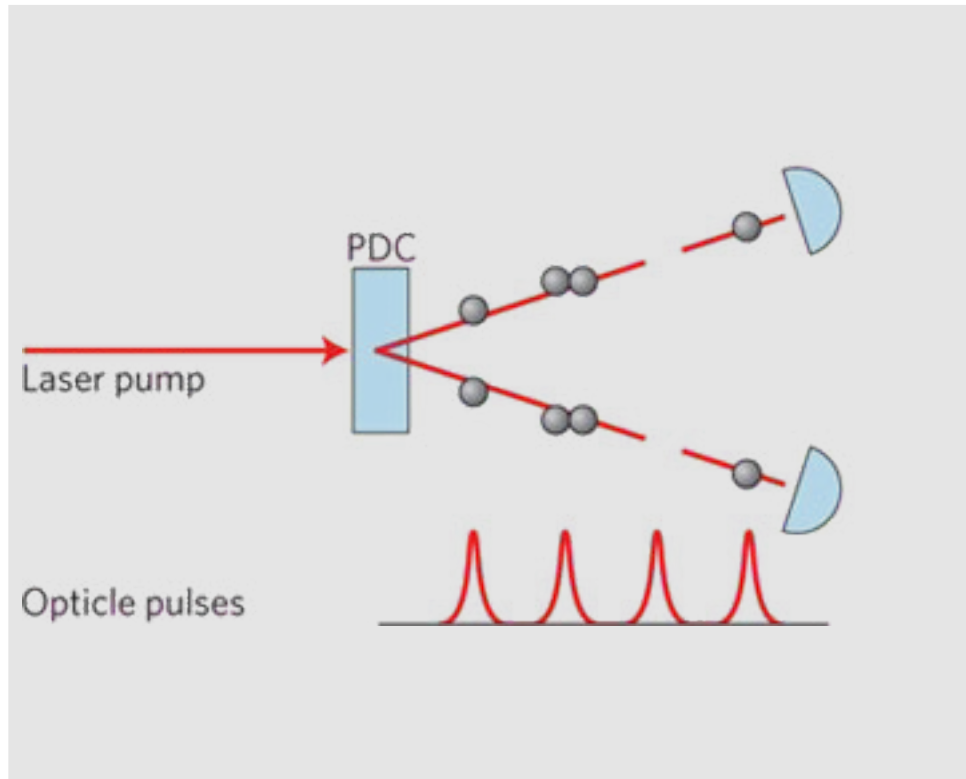
- ▶ The N00N state leads to a phase shift multiplied by N
- ▶ This leads to a N-fold enhancement of the resolution
- ▶ The N00N state is a highly non-classical, entangled state
- ▶ $|2002\rangle$ can be produced by Hong-Ou-Mandel effect



How can entanglement be created?

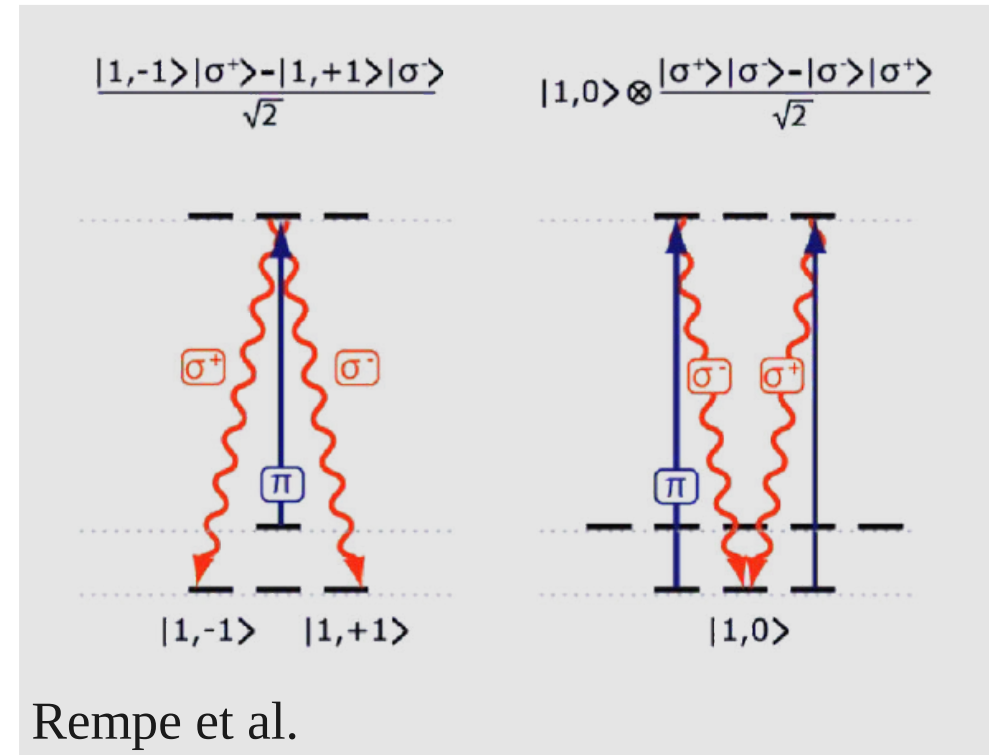
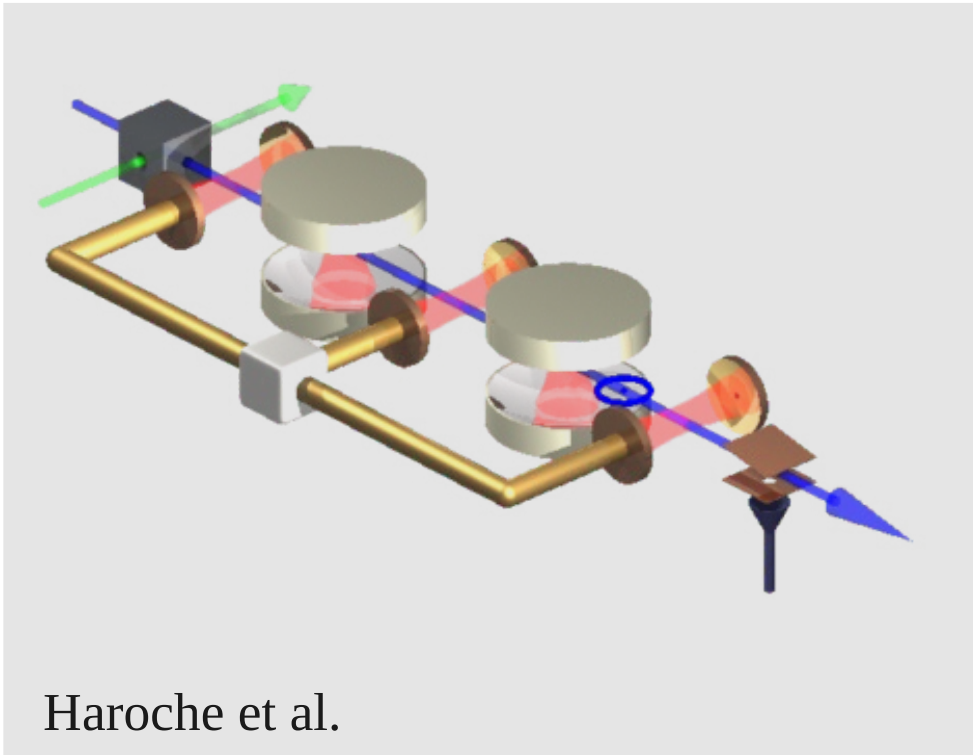


Down conversion



- ▶ 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

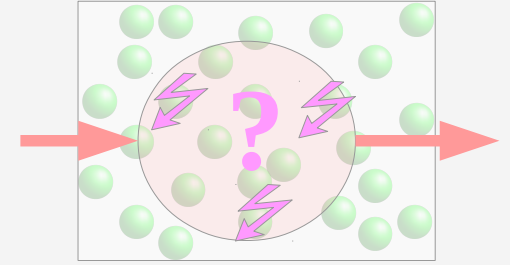
Coherent control



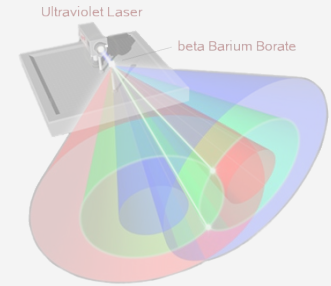
- ▶ Deterministic, but need strong coupling (“Pi pulses”)
- ▶ Other related implementations (Quantum dots, ...)
- ▶ Coherent control approach for entanglement generation in NFS

Content

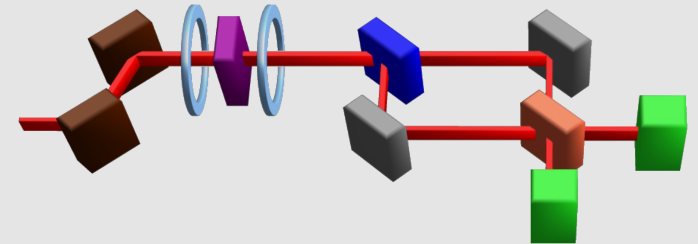
Introduction / NFS



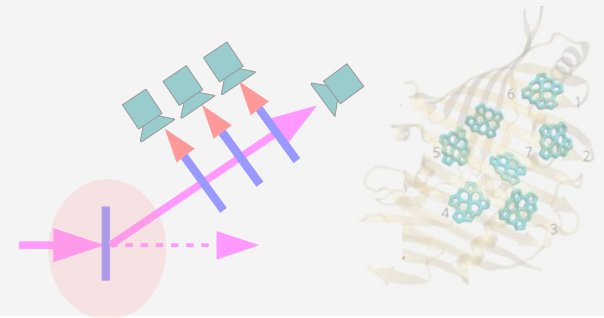
Quantum optics and information



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 (~ 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

VOLUME 77, NUMBER 15

PHYSICAL REVIEW LETTERS

7 OCTOBER 1996

Storage of Nuclear Excitation Energy through Magnetic Switching

Yu. V. Shvyd'ko,¹ T. Hertrich,² U. van Bürck,² E. Gerdau,¹ O. Leupold,¹ J. Metge,¹ H. D. Rüter,¹ S. Schwendy,¹
G. V. Smirnov,³ W. Potzel,² and P. Schindelmann²

¹*II. Institut für Experimentalphysik, Universität Hamburg, D-22761 Hamburg, Germany*

²*Physik-Department E15, Technische Universität München, D-85748 Garching, Germany*

³*RRC, "Kurchatov Institute", SU-1123182 Moscow, Russia*

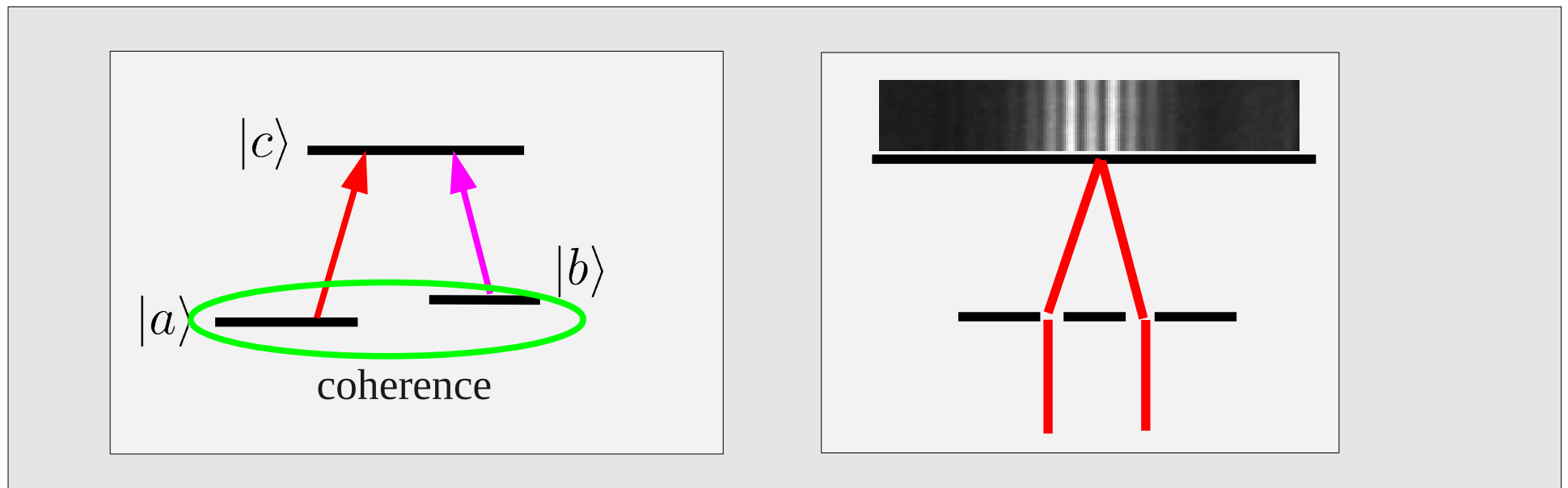
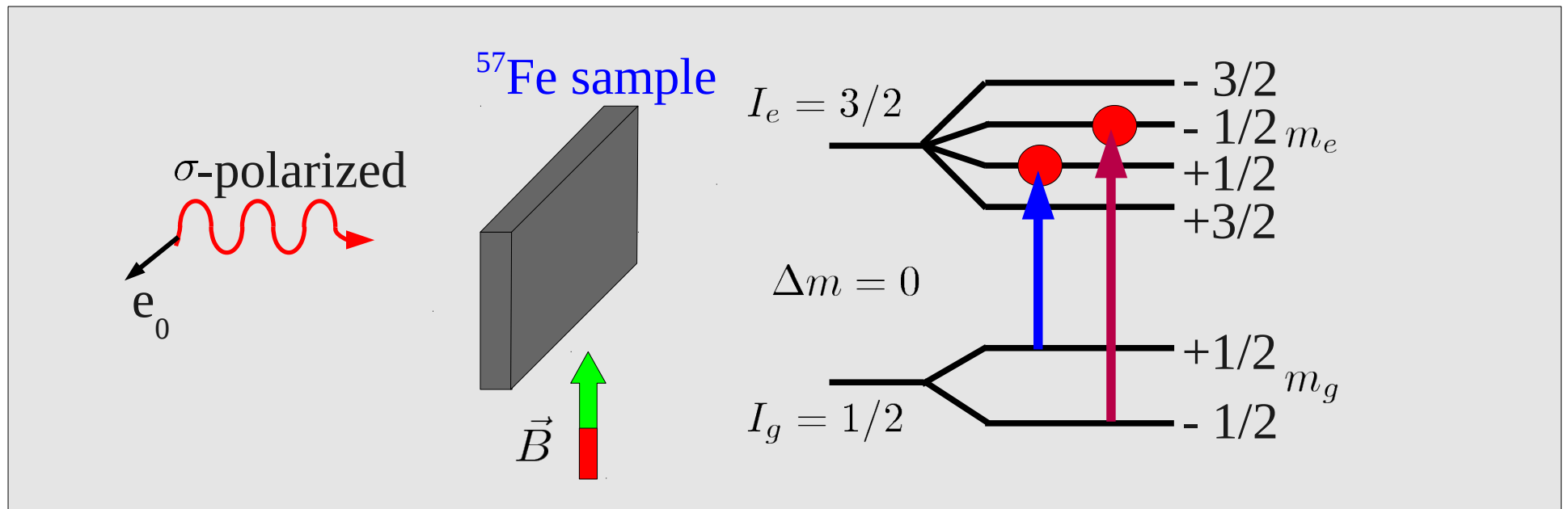
(Received 8 May 1996)

The decay rate of ^{57}Fe nuclei in an $^{57}\text{FeBO}_3$ 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

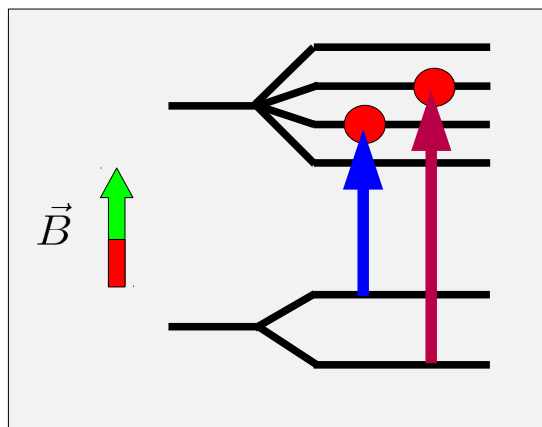
Phys. Rev. Lett. 77, 3232 (1996)

Two “ingredients”



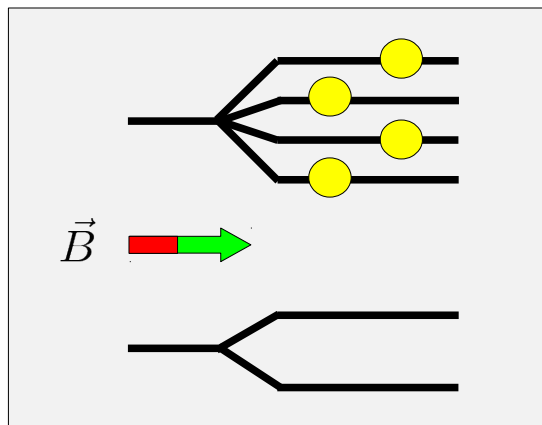
Coherent control of the exciton

Excite the sample



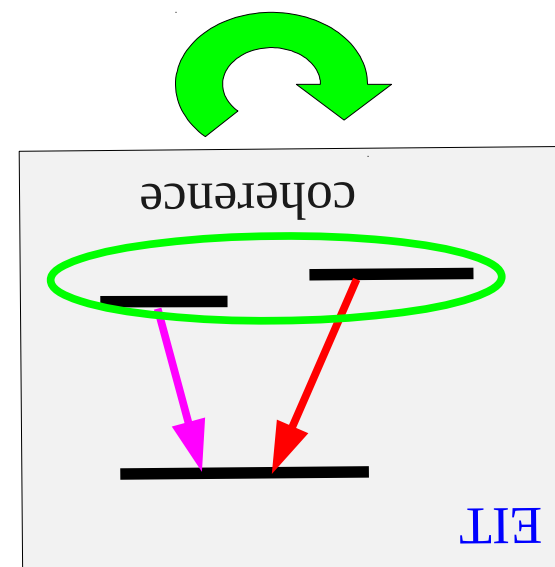
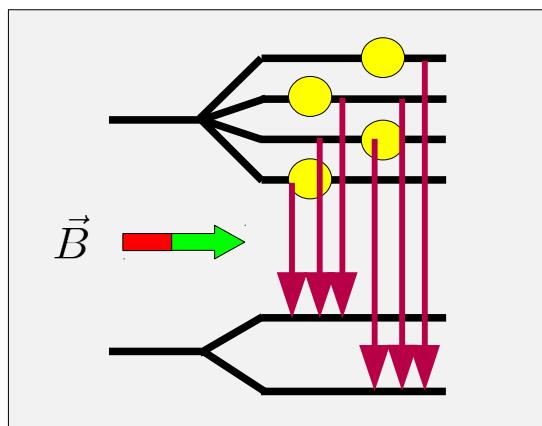
Rotate quantization axis

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



Deexcitation

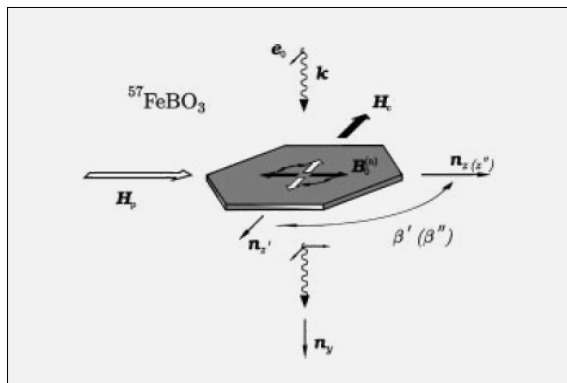
- ▶ Destructive interference of all pathways possible
- ▶ Analogy to electromagnetically induced transparency



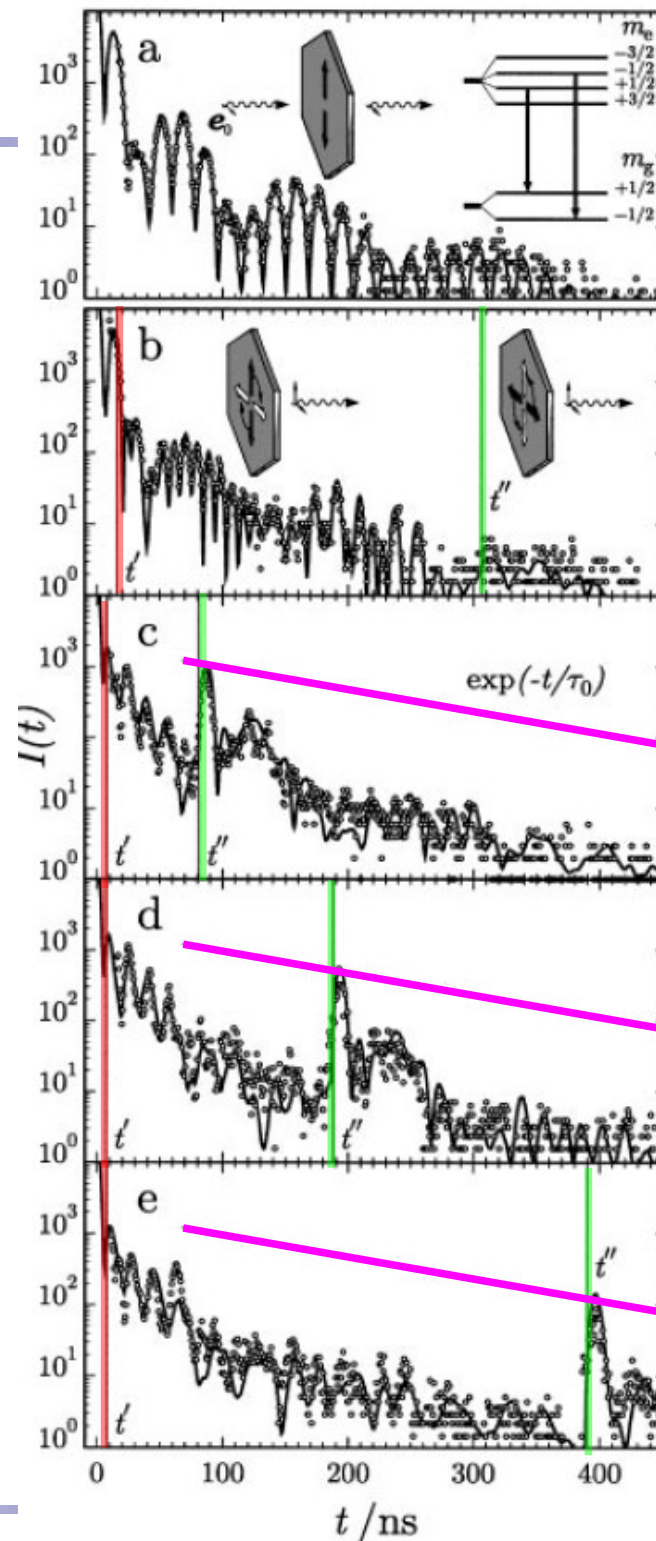
Exciton storage

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



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



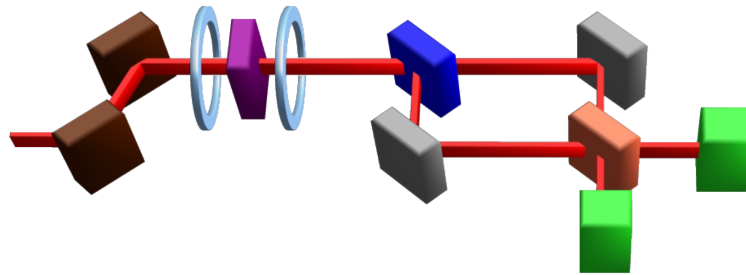
No switching

Apply
switching

Switch back

Decay with
natural life
time

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
-

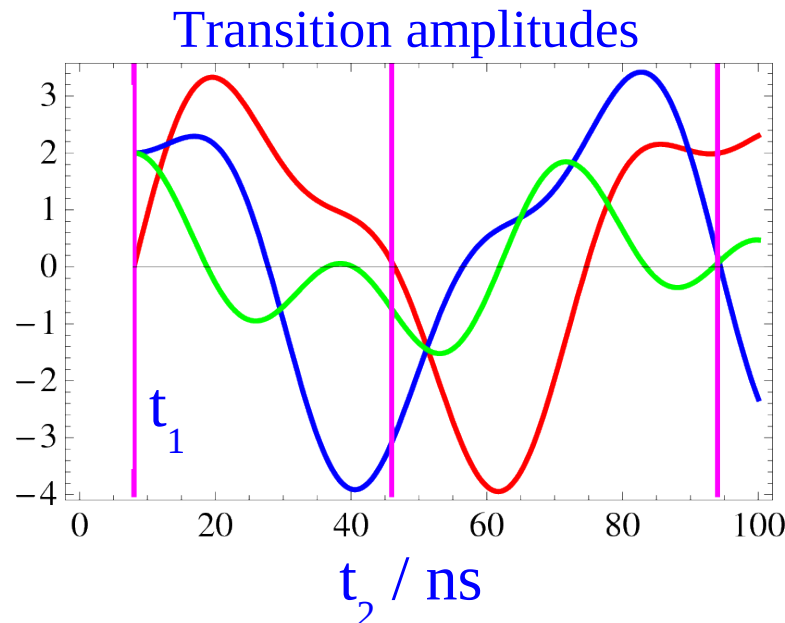
Advanced magnetic switching schemes

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?



$$\left\{ \frac{1}{2} \rightarrow \frac{1}{2}, -\frac{1}{2} \rightarrow -\frac{1}{2} \right\}$$

linear

$$\left\{ -\frac{1}{2} \rightarrow \frac{1}{2}, \frac{1}{2} \rightarrow -\frac{1}{2} \right\}$$

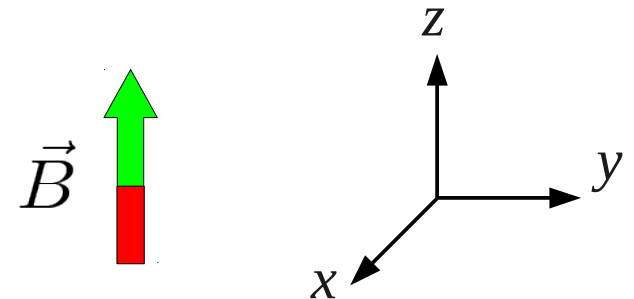
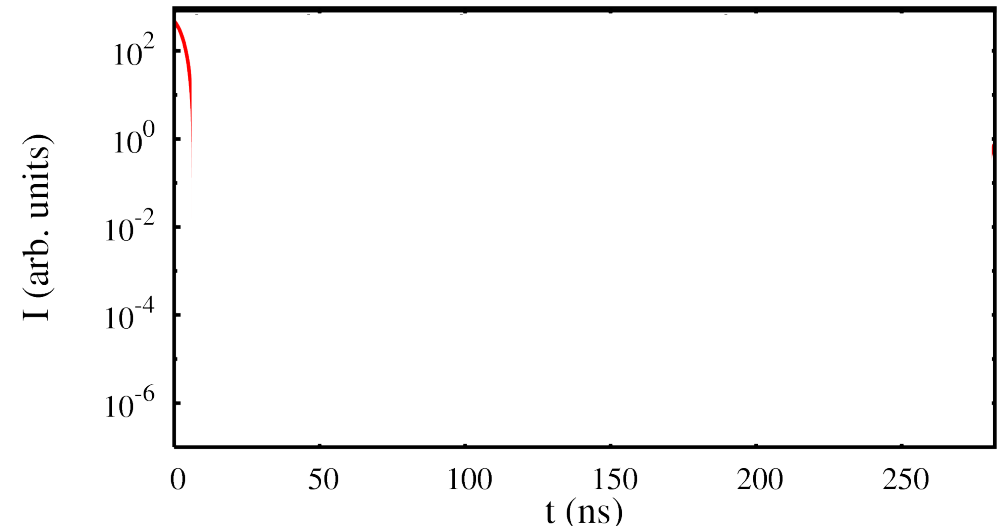
circular

$$\left\{ \frac{3}{2} \rightarrow \frac{1}{2}, -\frac{3}{2} \rightarrow -\frac{1}{2} \right\}$$

circular

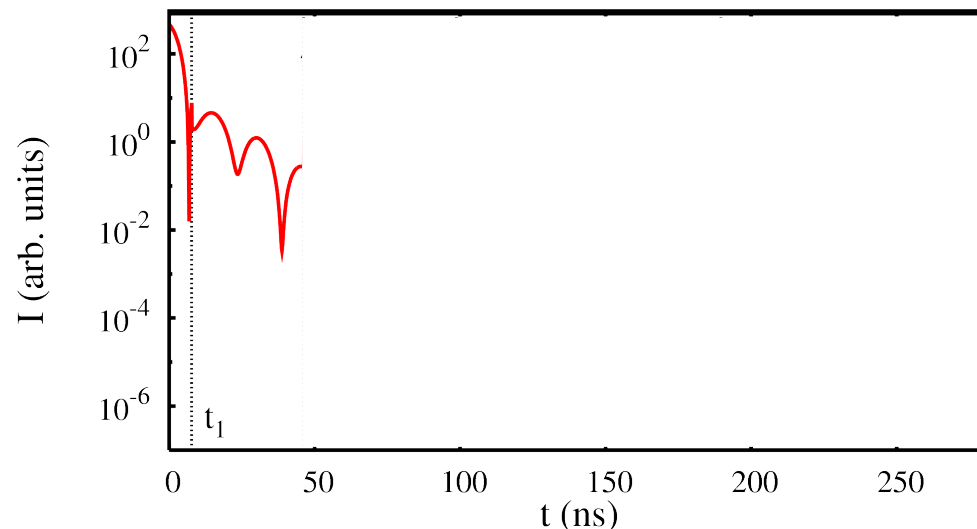
Step 1: Synchrotron excitation

- ▶ Initially, magnetic field is in z direction

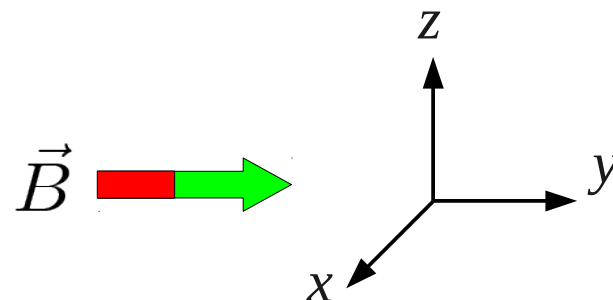
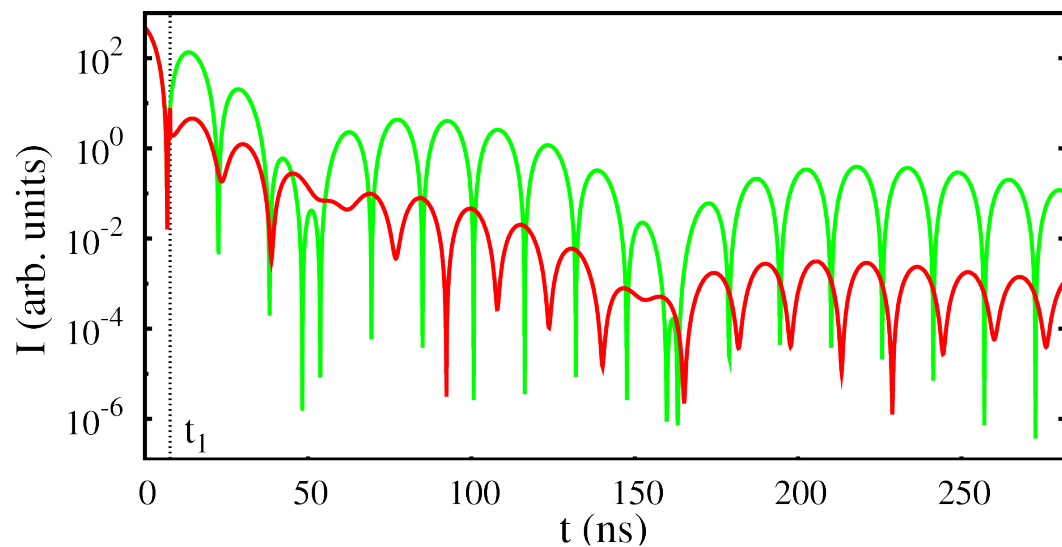


Step 2: Canceling coherent decay

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

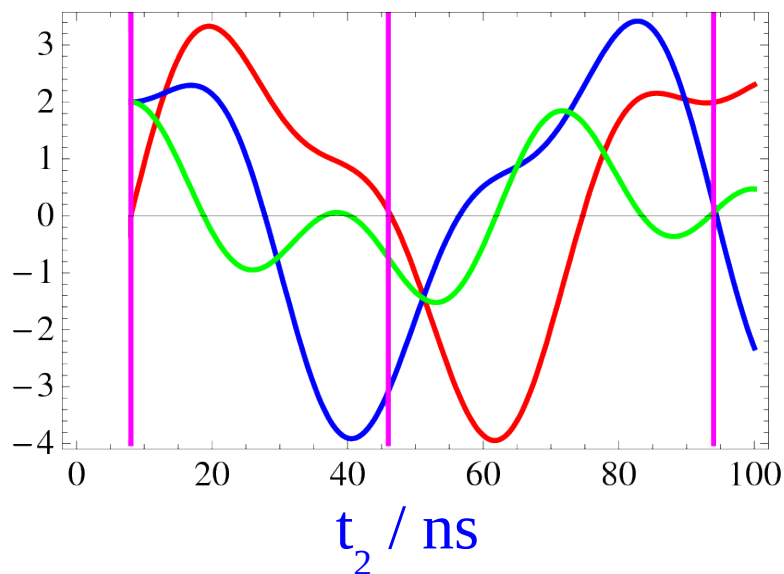
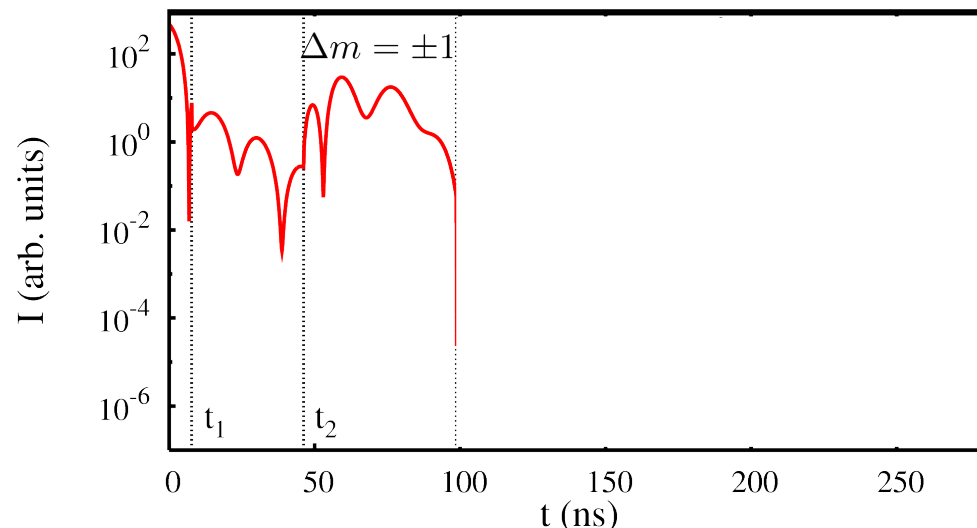


no switching - switching



Step 3: Releasing circular polarization

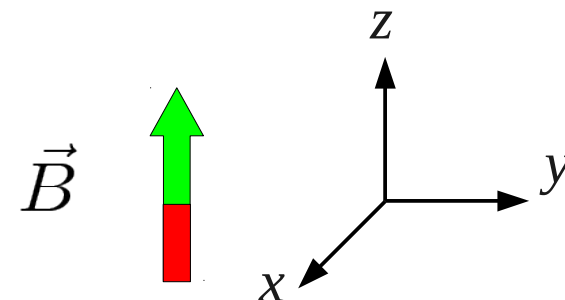
- Initially, magnetic field is in z direction
- At time t_1 , cancel decay by rotating into y direction
- At time t_2 , enable decay on $\Delta m = \pm 1$ but continue to suppress $\Delta m = 0$



$$\left\{ \frac{1}{2} \rightarrow \frac{1}{2}, -\frac{1}{2} \rightarrow -\frac{1}{2} \right\}$$

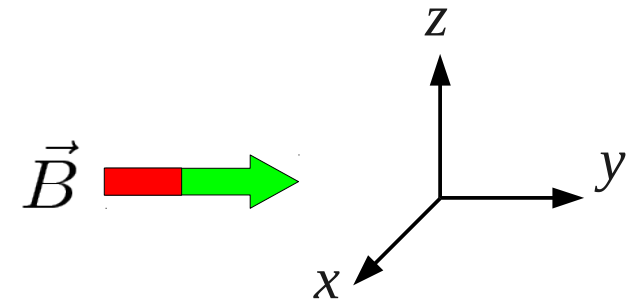
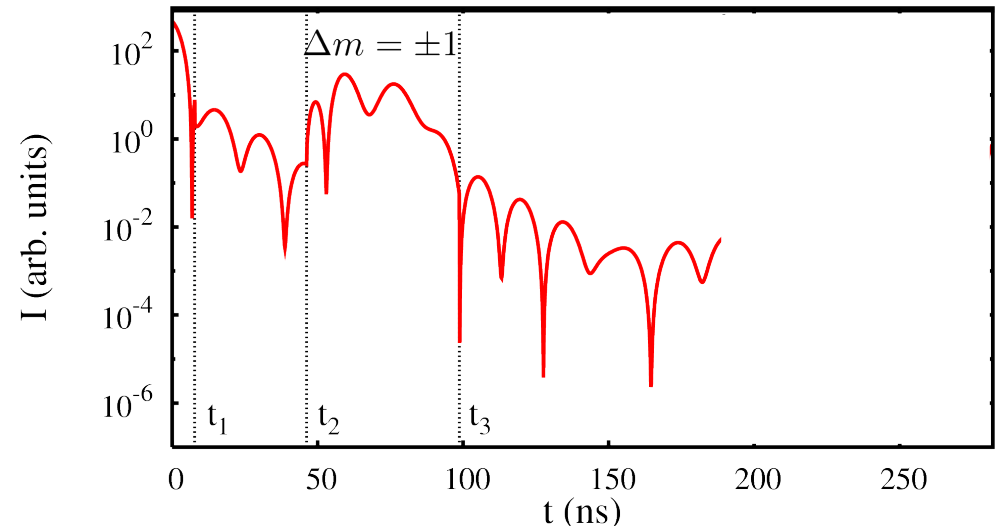
$$\left\{ -\frac{1}{2} \rightarrow \frac{1}{2}, \frac{1}{2} \rightarrow -\frac{1}{2} \right\}$$

$$\left\{ \frac{3}{2} \rightarrow \frac{1}{2}, -\frac{3}{2} \rightarrow -\frac{1}{2} \right\}$$



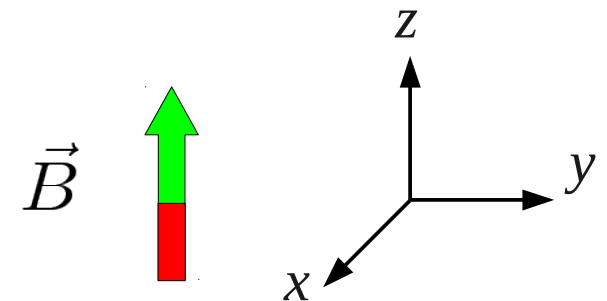
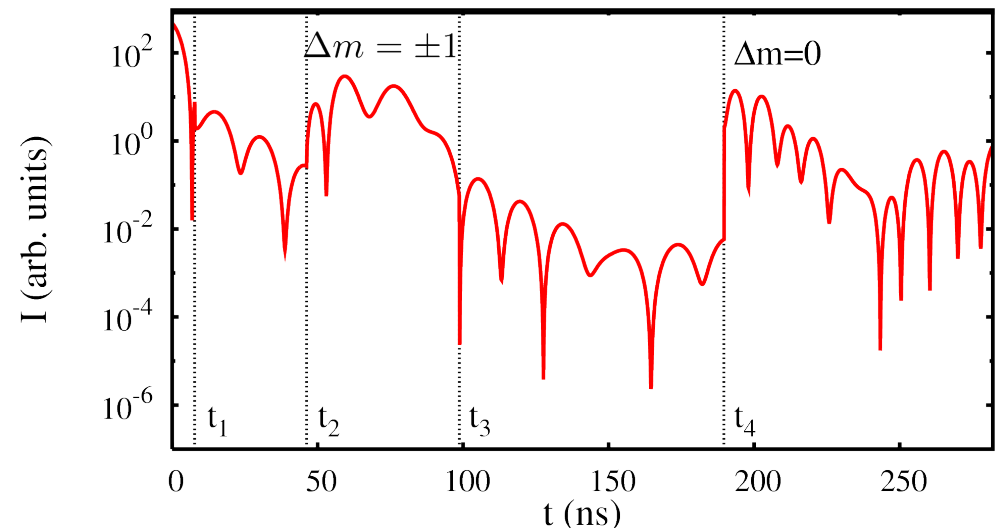
Step 4: Canceling coherent decay

- ▶ 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

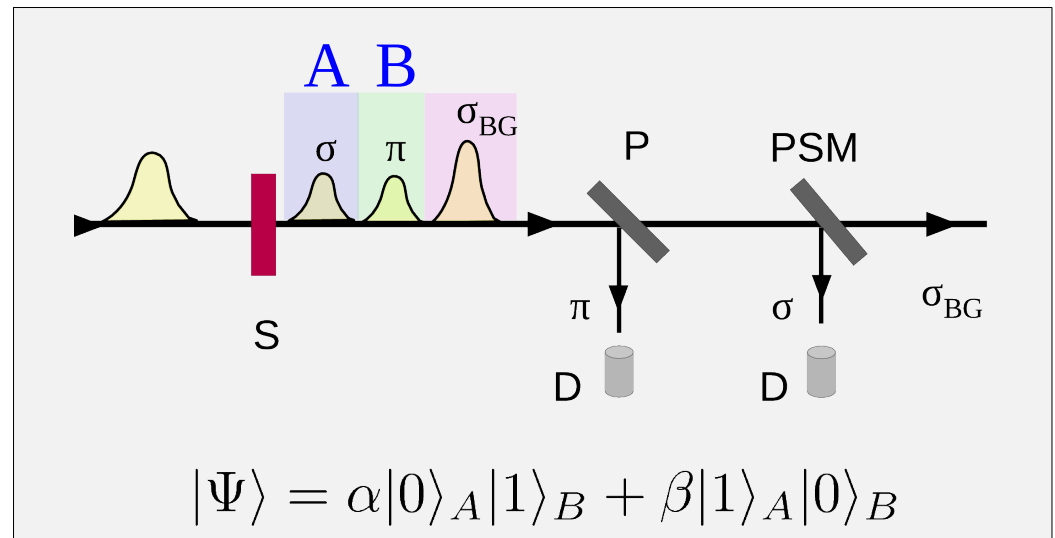
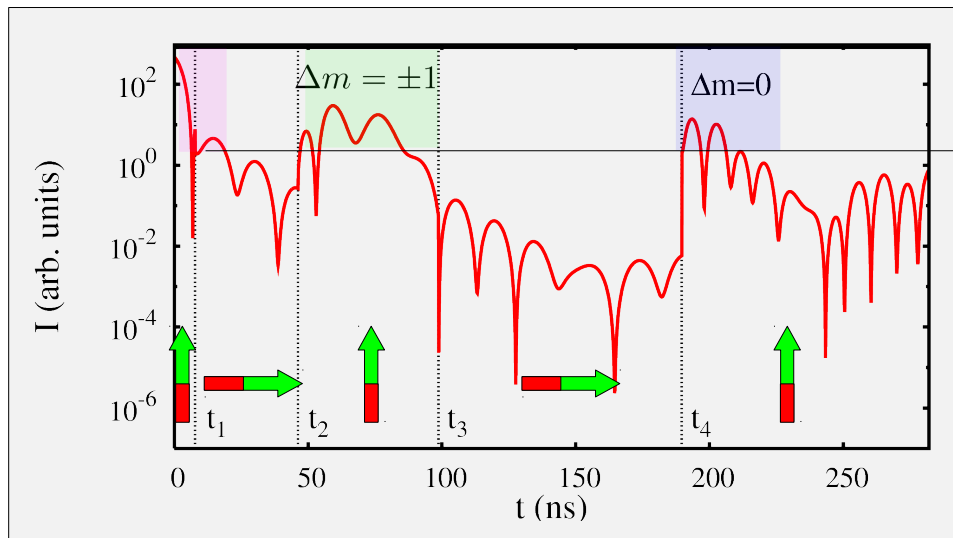
- Initially, magnetic field is in z direction
- At time t_1 , cancel decay by rotating into y direction
- At time t_2 , enable decay on $\Delta m = \pm 1$ but continue to suppress $\Delta m = 0$
- At time t_3 , cancel decay by rotating into y direction
- At time t_4 , enable decay on $\Delta m = 0$



Temporal mode entanglement

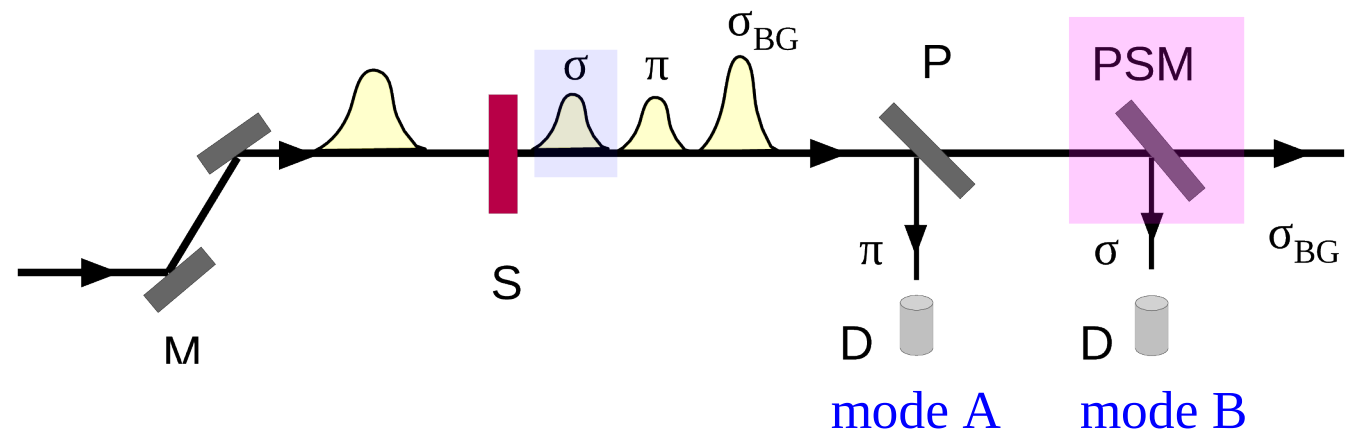
Design advanced coherent control scheme:

- ▶ 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



How to extract signal pulse ?

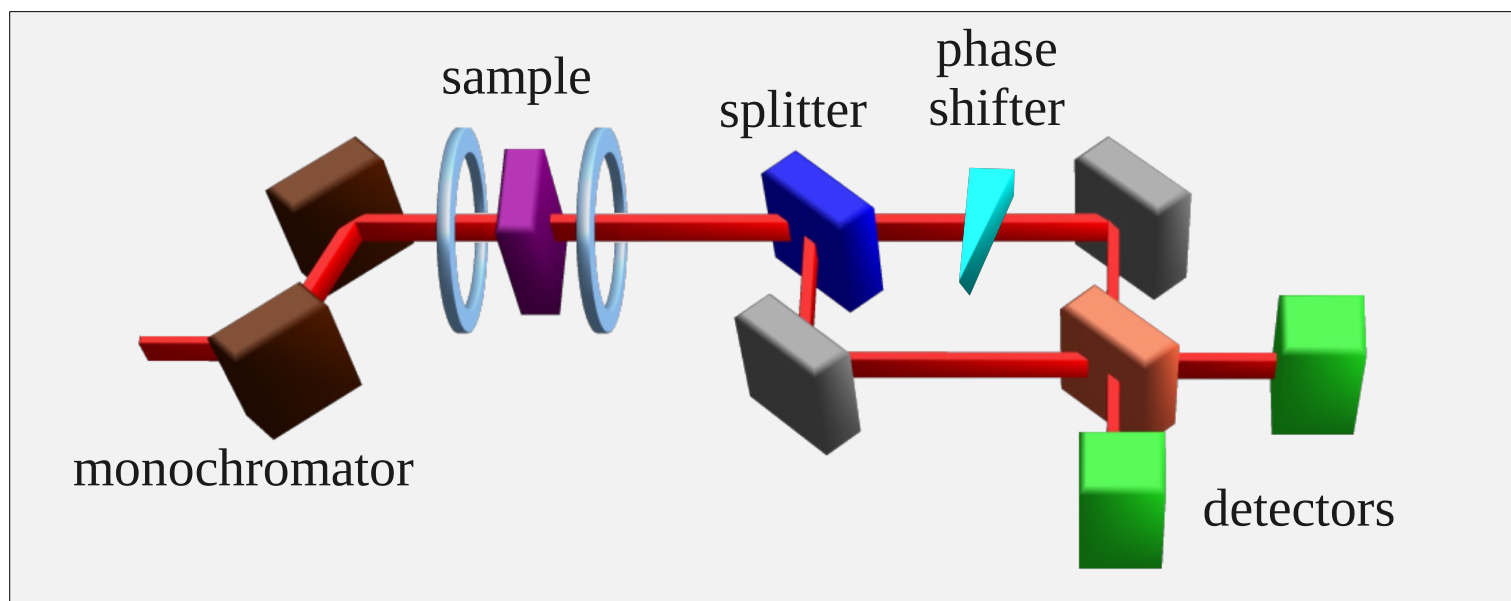
- ▶ Problem: One part of signal has same polarization as background pulse
- ▶ Time gating not useful if following setup should be protected from high-intensity background; lighthouse effect difficult because of precise timing of nuclear switching
- ▶ PSM: Piezo electric steering mirror or sub-ns control device based on crystal lattice deformation ¹⁾
- ▶ Have about 180 ns “steering time” because of magnetic switching



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

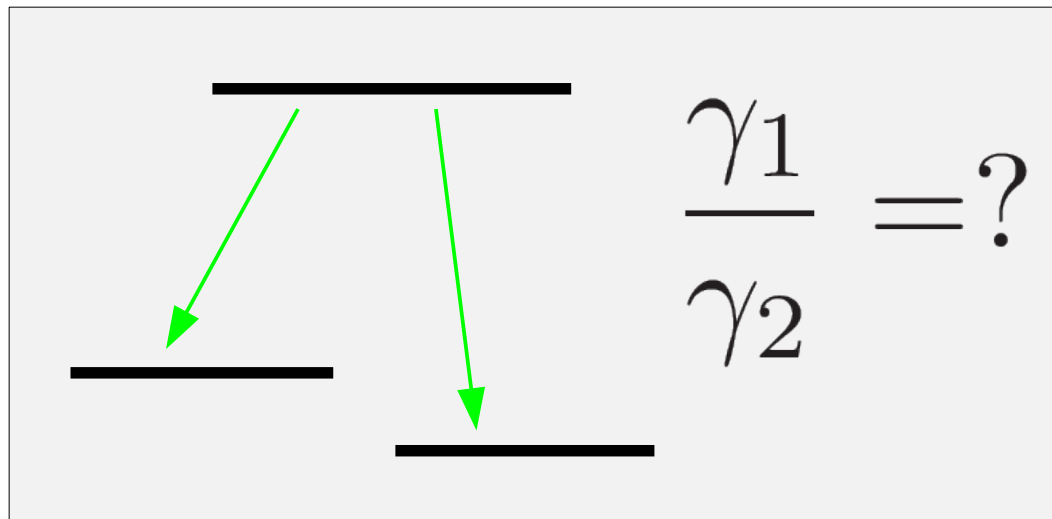
Proof-of-principle experiment

- ▶ Do not extract signal, use time gating to remove background
- ▶ Switching → 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



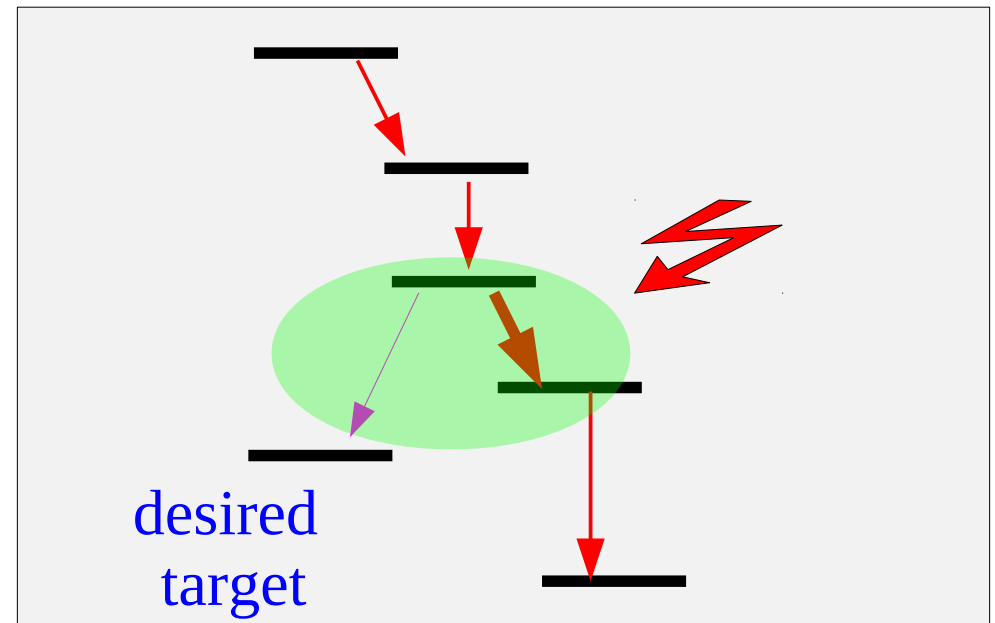
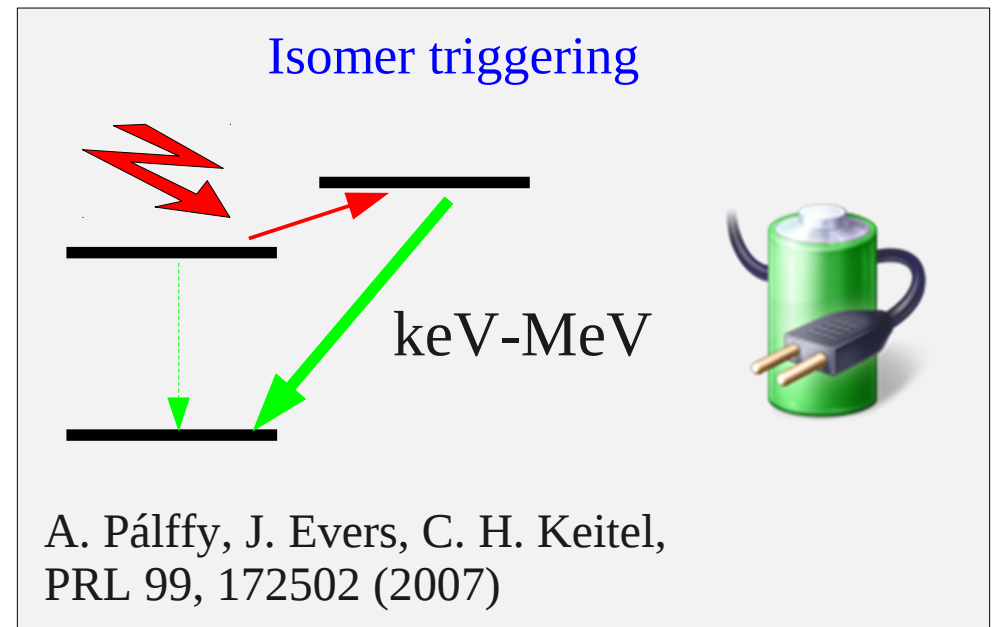
^{*)} H.-W. Lee and Kim, Phys. Rev. A 63, 012305 (2000)

X-ray branching ratio control



Motivation

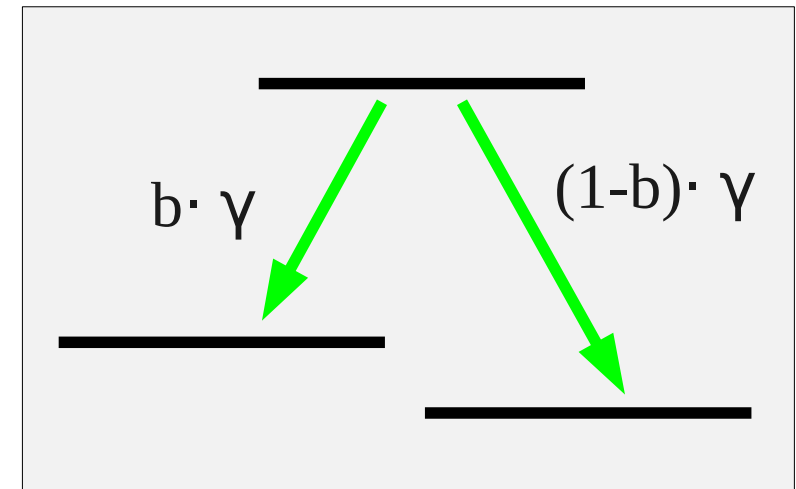
- ▶ Prepare specific initial state
 - single magnetic ground-state sub-level
 - metastable excited state (isomer)
- ▶ Release excited state on demand
- ▶ Modify/control chain of decays, e.g., for preparation of specific states/isotopes
- ▶ Assist the control over quantum dynamics in more advanced setups



Branching ratio

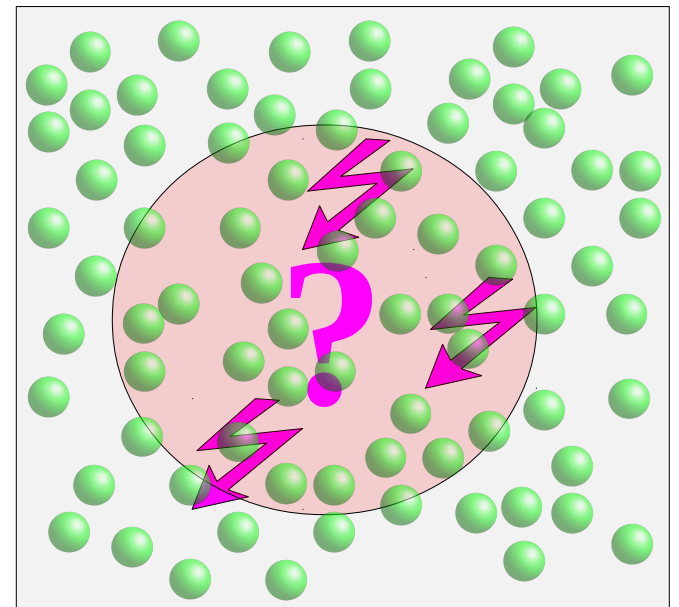
Single particle branching ratio:

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



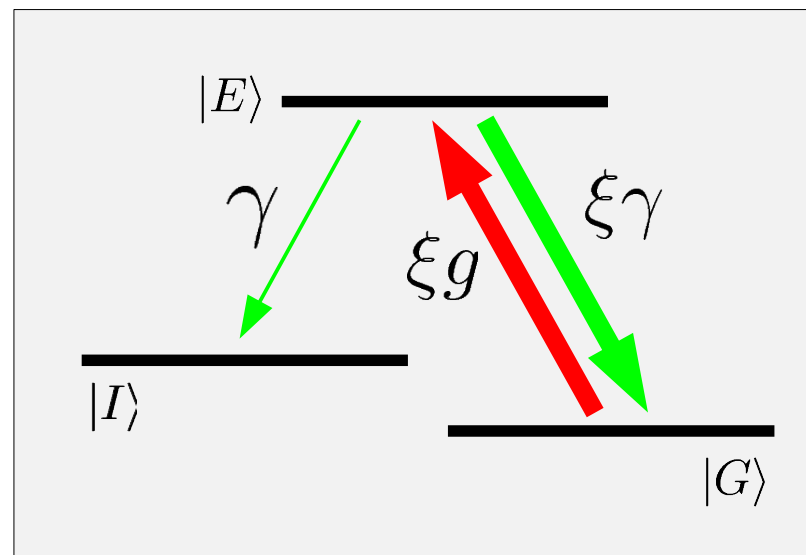
Branching ratio in ensembles

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



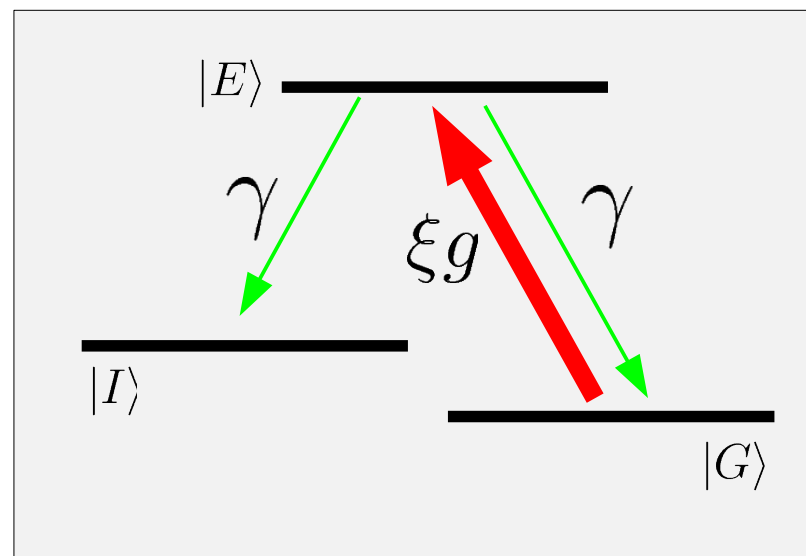
Motivation

- ▶ 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 $|I\rangle$



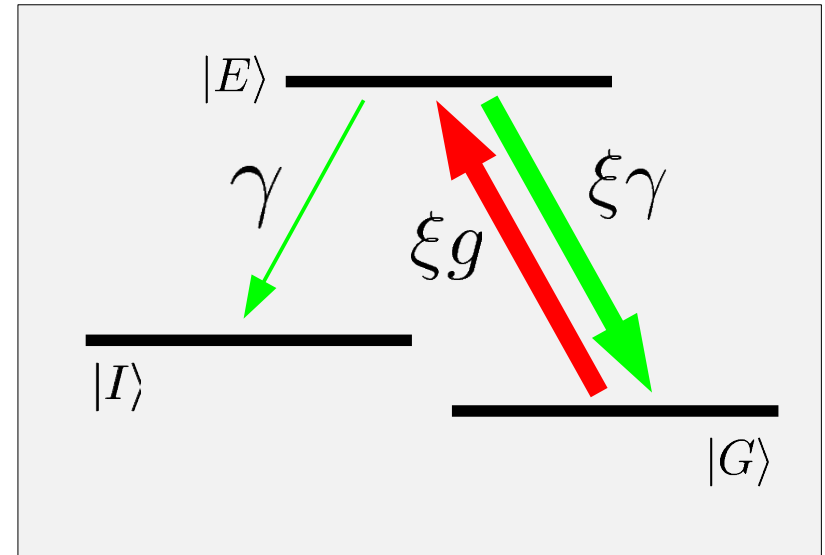
Idea:

- ▶ 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:

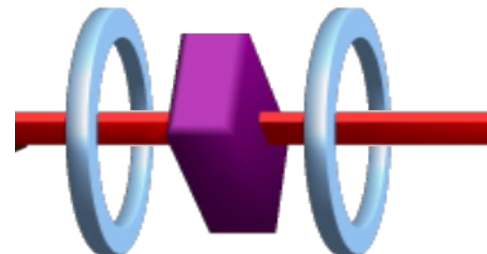
$$b_c^C / b_c^{NC} = \xi + 1$$

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

How to control?

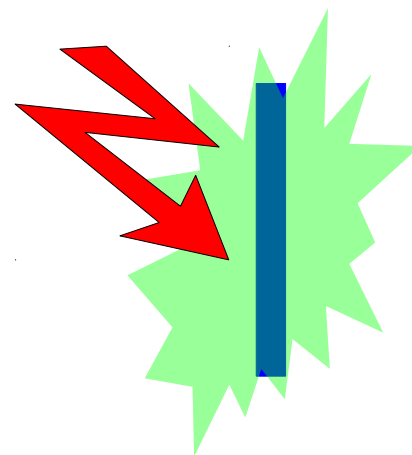
Magnetic switching:

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

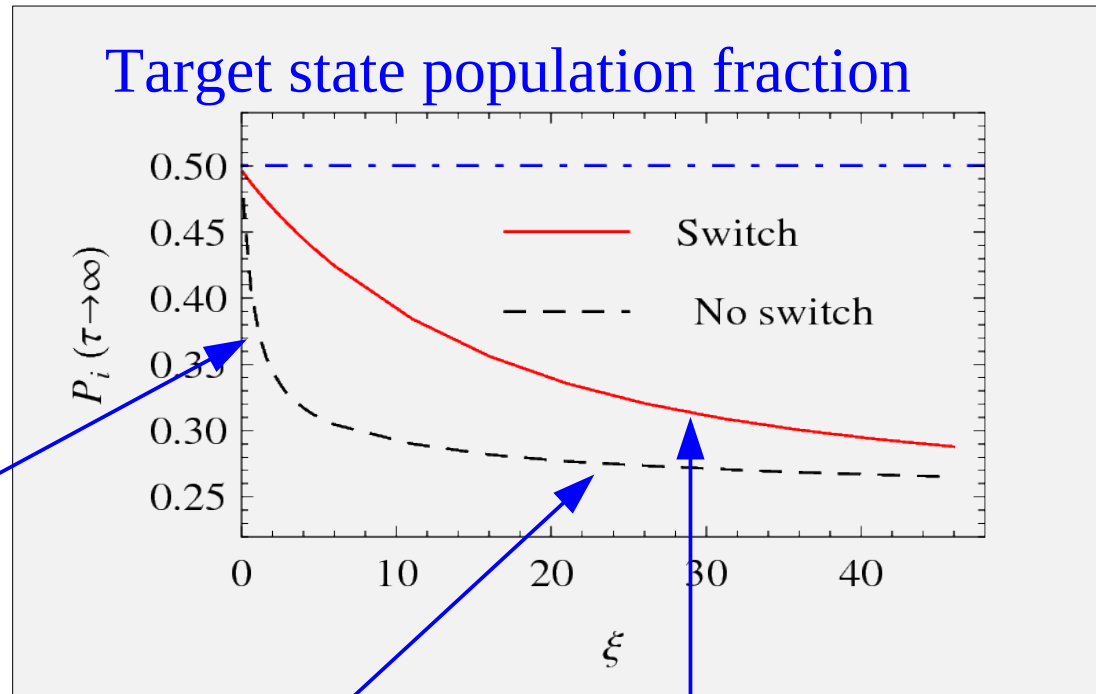
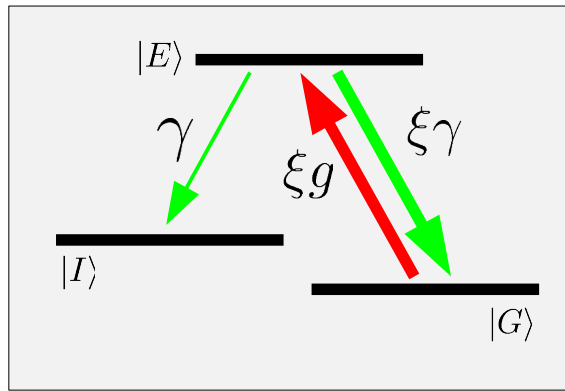


Destroy phase coherence:

- ▶ 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

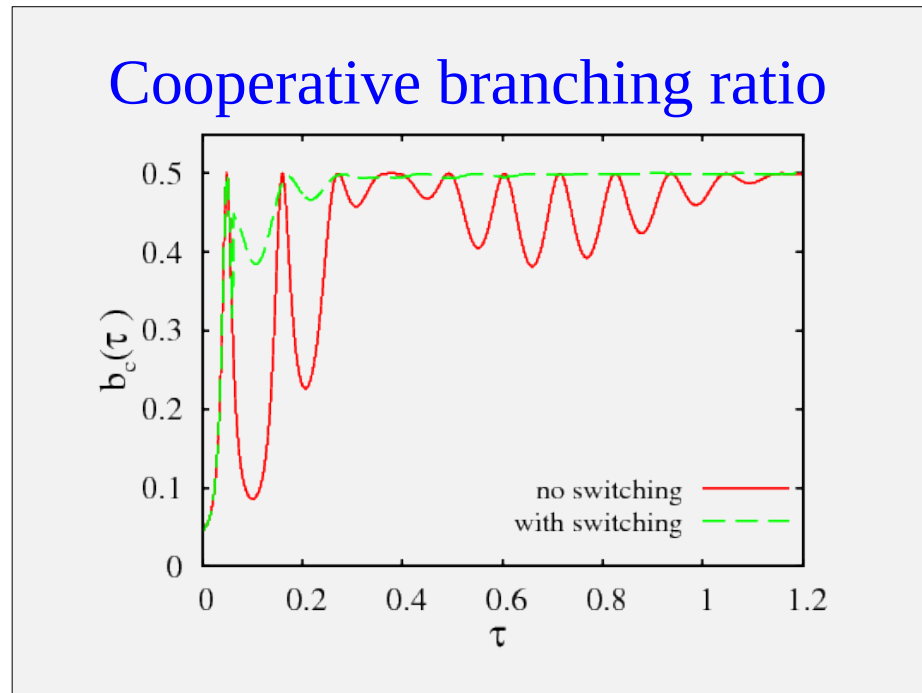


superradiant
decay to initial
state

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

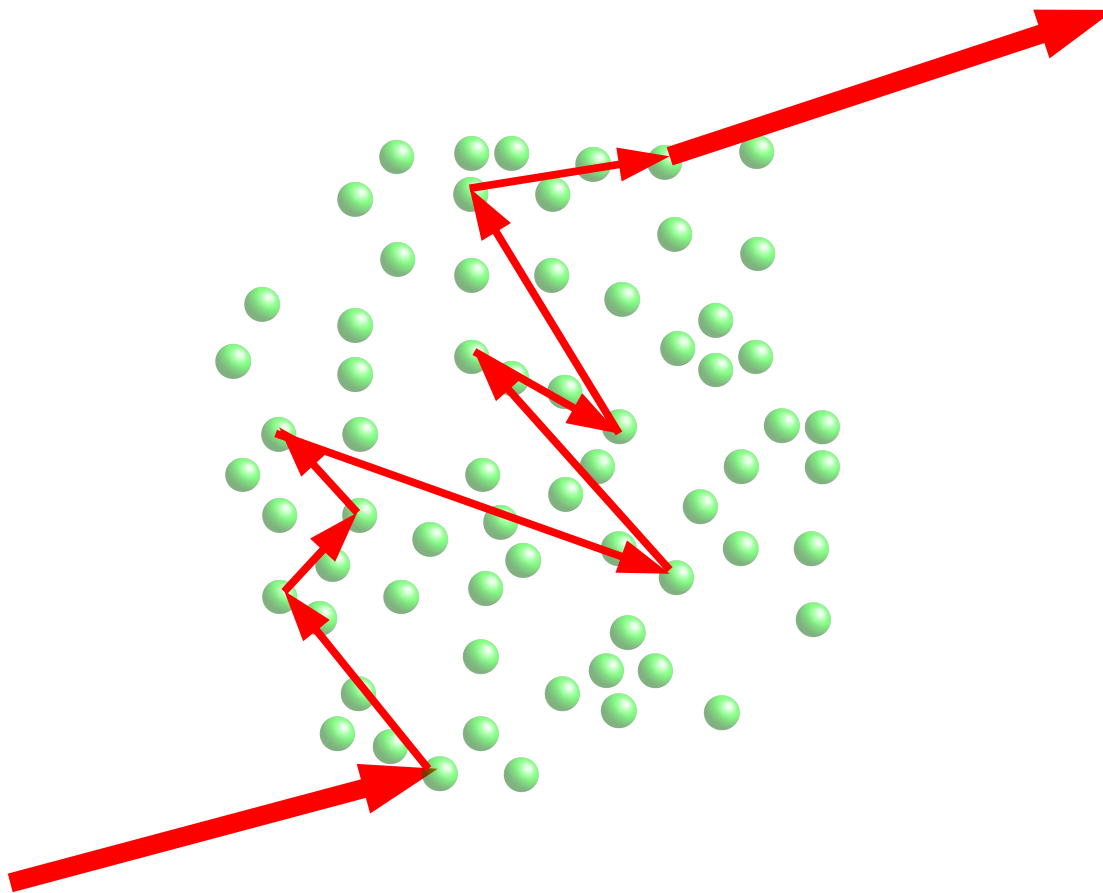
Switching improves result,
but significant decay before
trapping can be achieved
→ better results with
phase destruction

The magnetic switching case



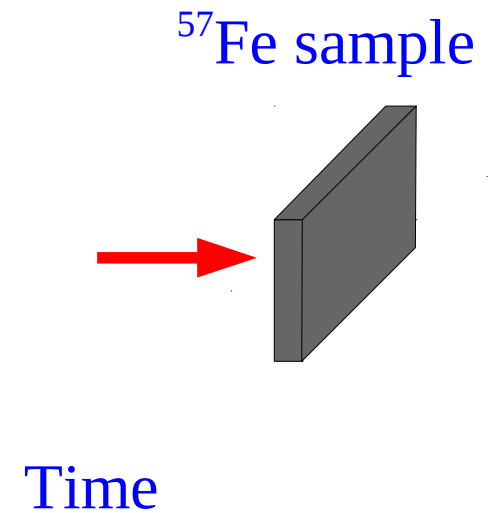
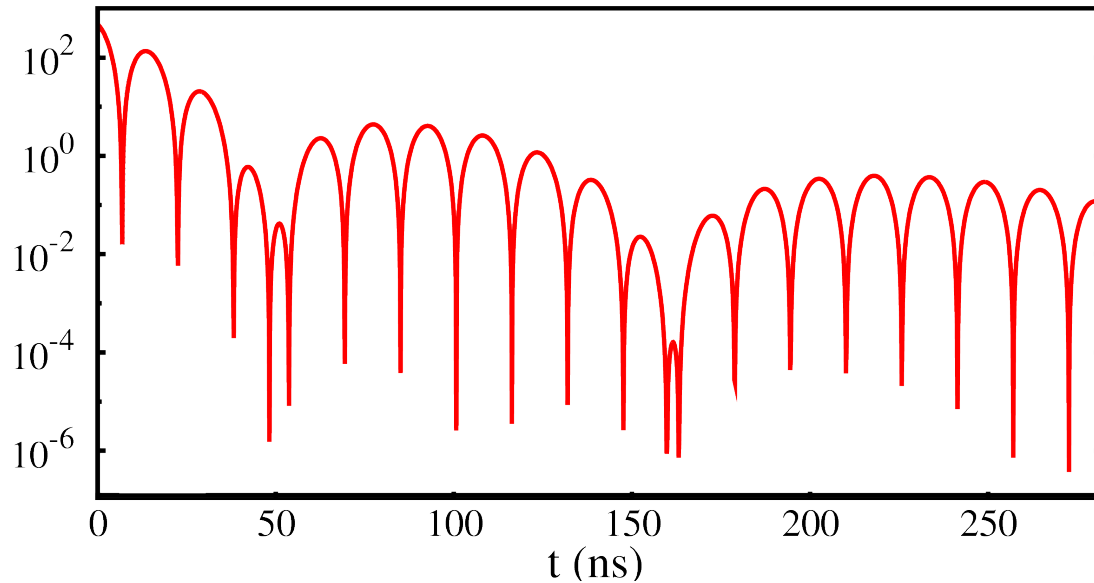
- ▶ Branching ratio time dependent as expected
- ▶ Cooperative branching ratio smaller than single-particle ratio due to superradiance
- ▶ After switching, single-particle branching ratio is achieved
- ▶ With destruction of phase coherence, single-particle ratio can immediately be achieved

Radiative eigenmodes

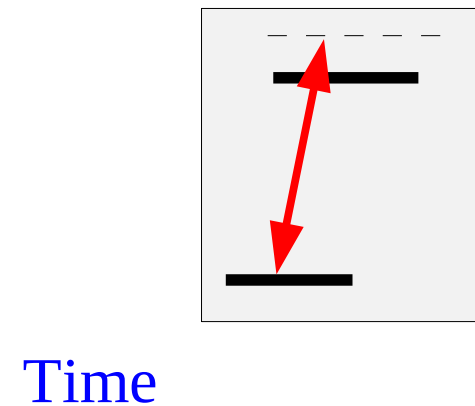
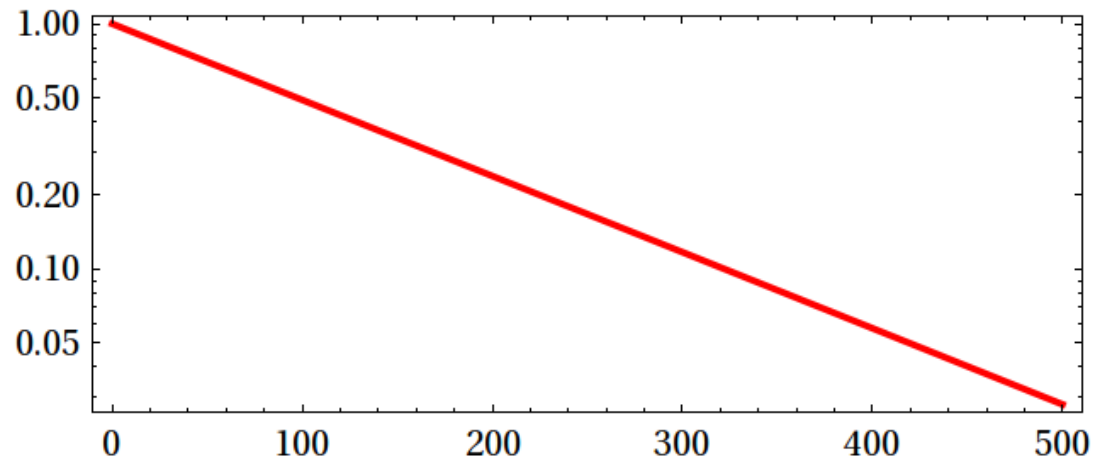


Temporal structure of scattered light

Signal intensity
 I (arb. units)

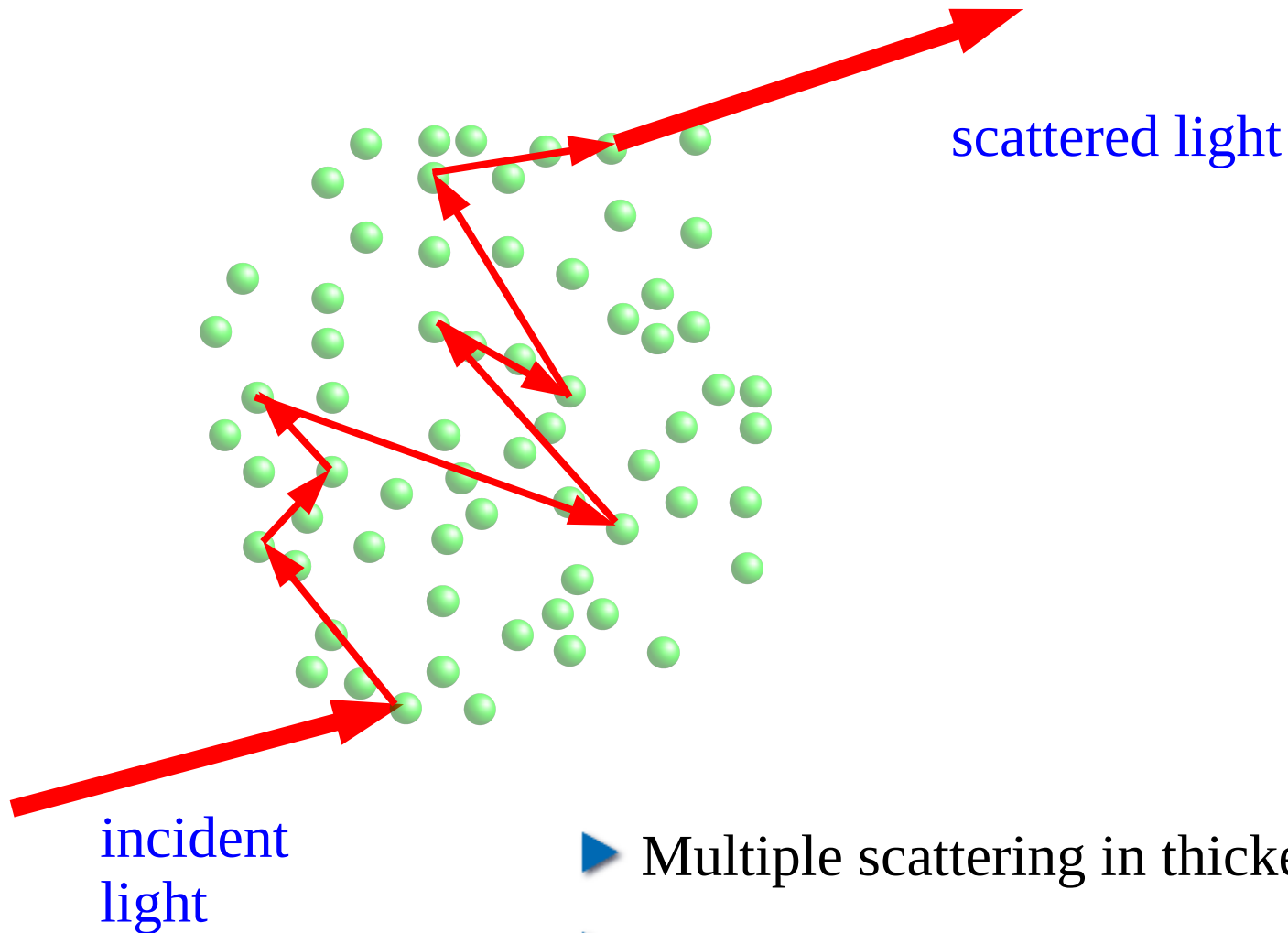


Signal intensity



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

$$|\psi(t)\rangle = \sum_j \beta_j(t) |e_j, \mathbf{0}\rangle + \sum_{\mathbf{k}} \eta_{\mathbf{k}}(t) |G, \mathbf{1}_{\mathbf{k}}\rangle$$

nucleus j
excited photon in
mode k

Equations of motion

$$\dot{\beta}_j(t) = -\frac{\Gamma_0}{2}\beta_j(t) - \sum_{j'(\neq j)} \frac{\Gamma_{j'}^{(j)}}{2}\beta_{j'}(t)$$

single nucleus
decay coupling

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 \swarrow \nwarrow single eigenmode

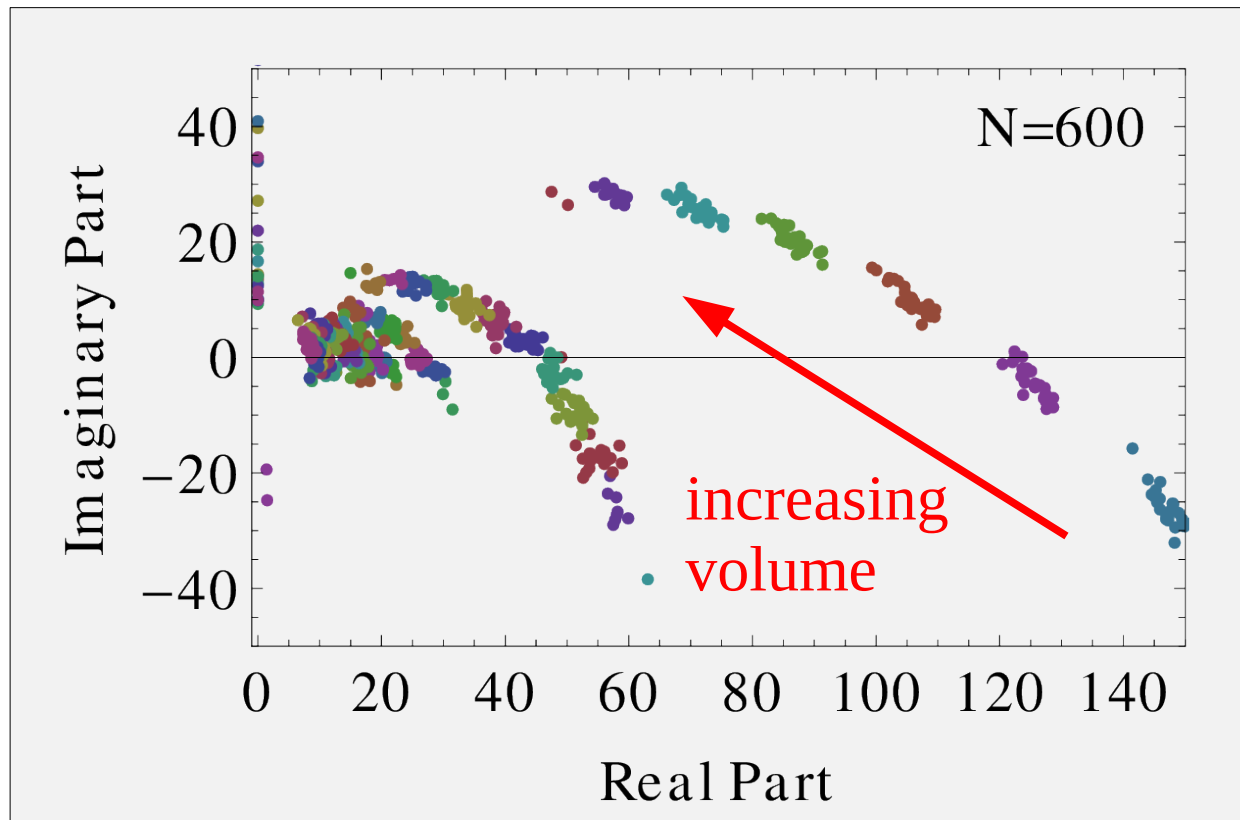
Time evolution

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

\uparrow eigenvalue

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

Dominant eigenmode vs. ensemble size

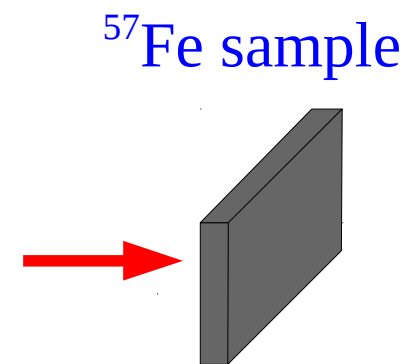


- ▶ Small volume: One dominating eigenvalue, strong exponential superradiance
- ▶ Intermediate volume: Several equivalent modes
- ▶ Large volume: Many competing modes, complicated temporal structure (sub/superradiant mixed)

Selective excitation of radiative modes

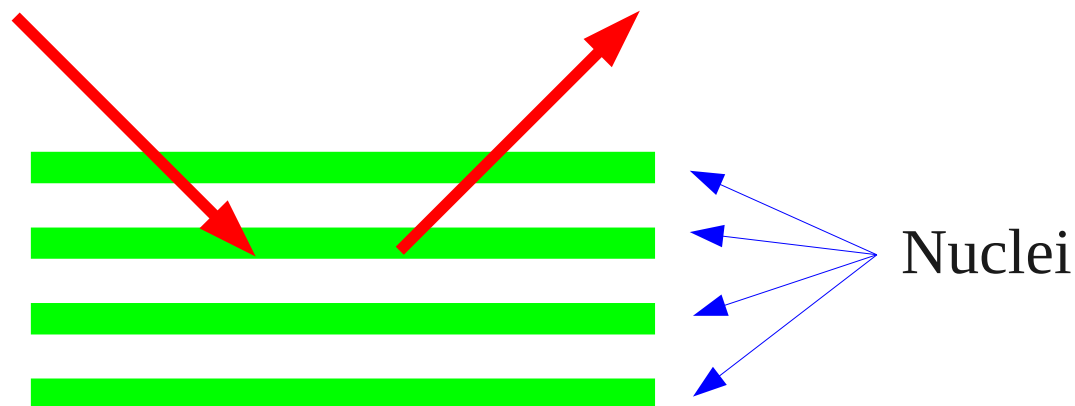
“Standard” x-ray scattering with thick target

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



X-ray scattering with structured targets

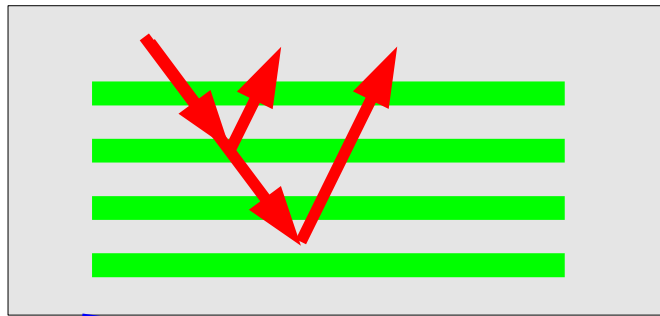
- ▶ In Bragg geometry, a single radiative eigenmode is excited



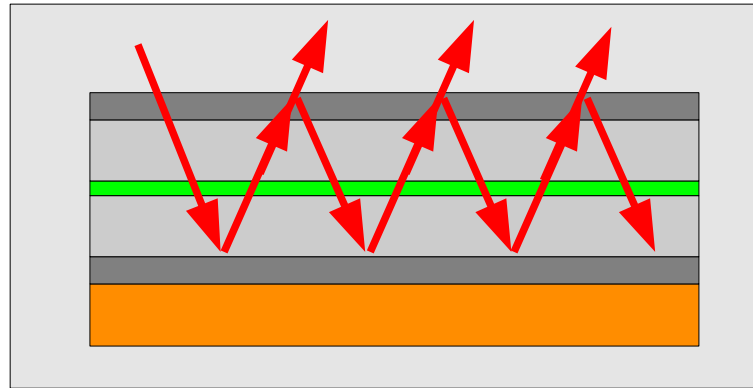
$$|T_{\mathbf{k}_I}\rangle = \frac{1}{\sqrt{N}} \sum_j e^{i\mathbf{k}_I \cdot \mathbf{r}_j} |e_j, \mathbf{0}\rangle$$

timed Dicke state

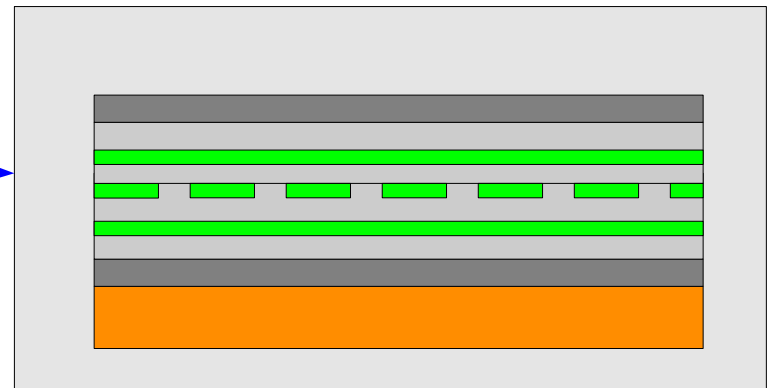
Exciting single radiative eigenmodes



Bragg geometry



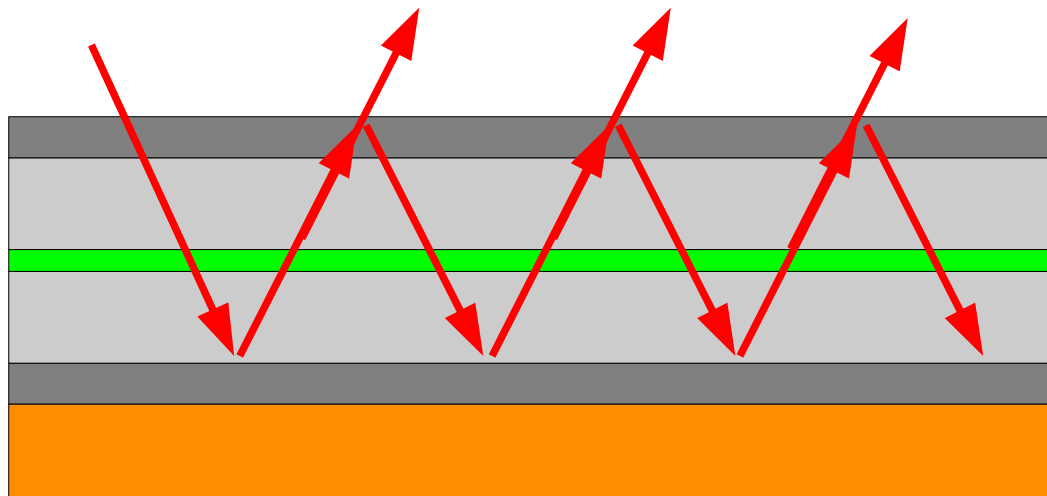
X-ray thin film cavities effectively realize Bragg case



Flexible design \Rightarrow more possibilities

geometry crucial

Thin film x-ray cavities

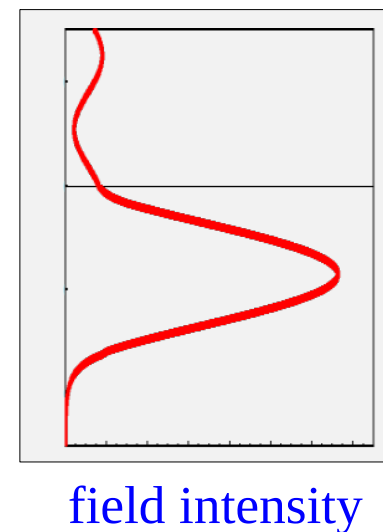
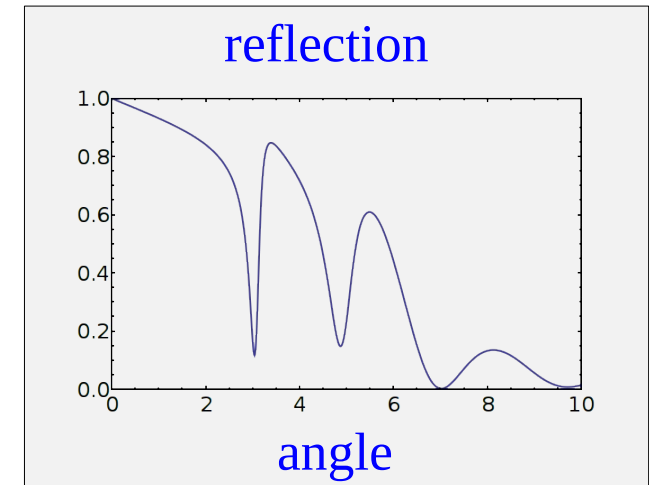
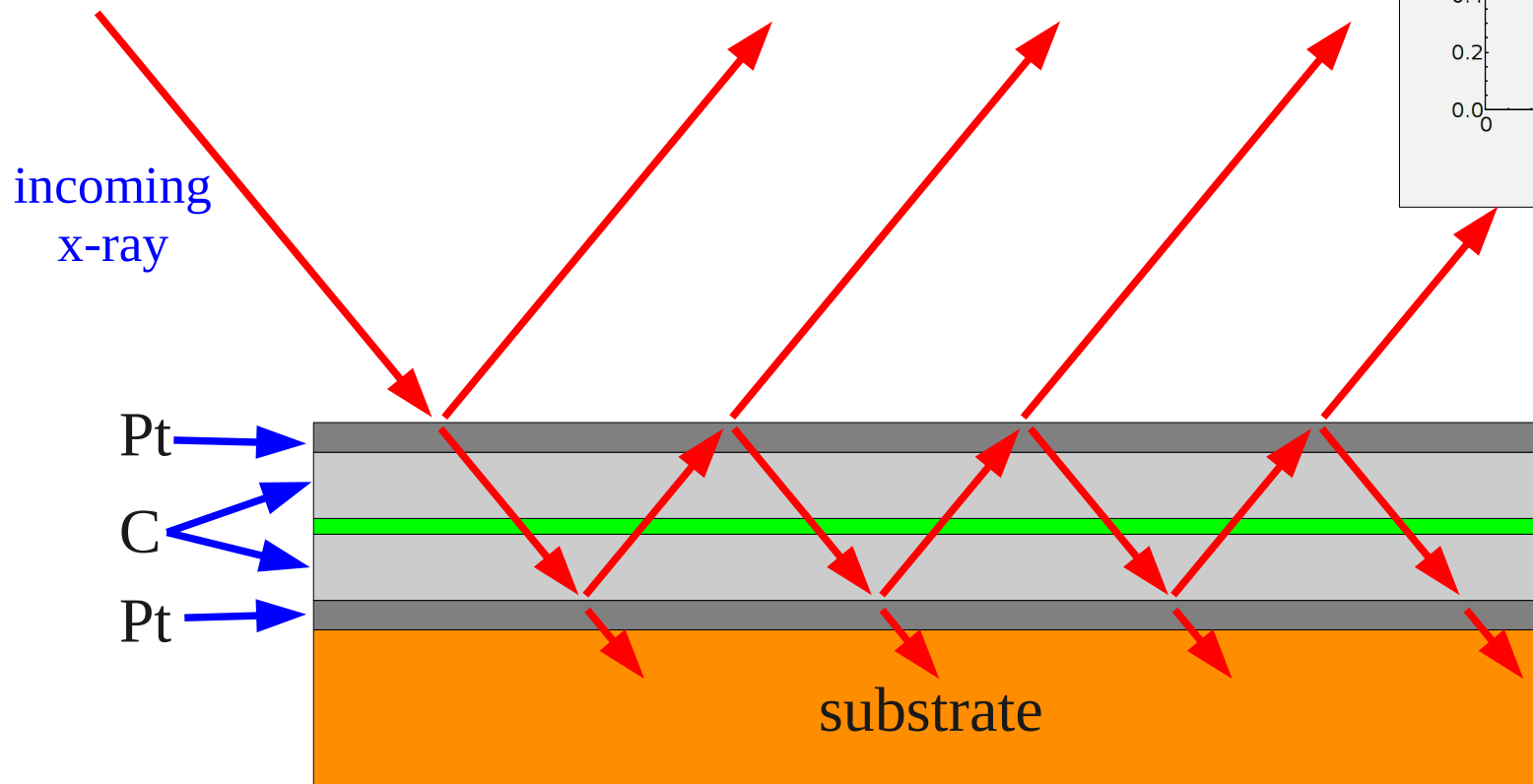


Thin film x-ray cavities

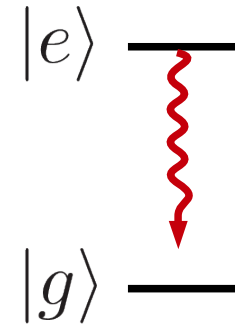
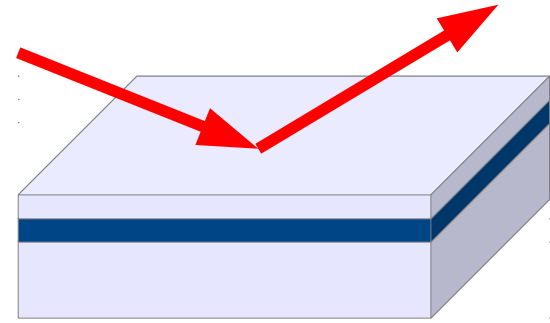
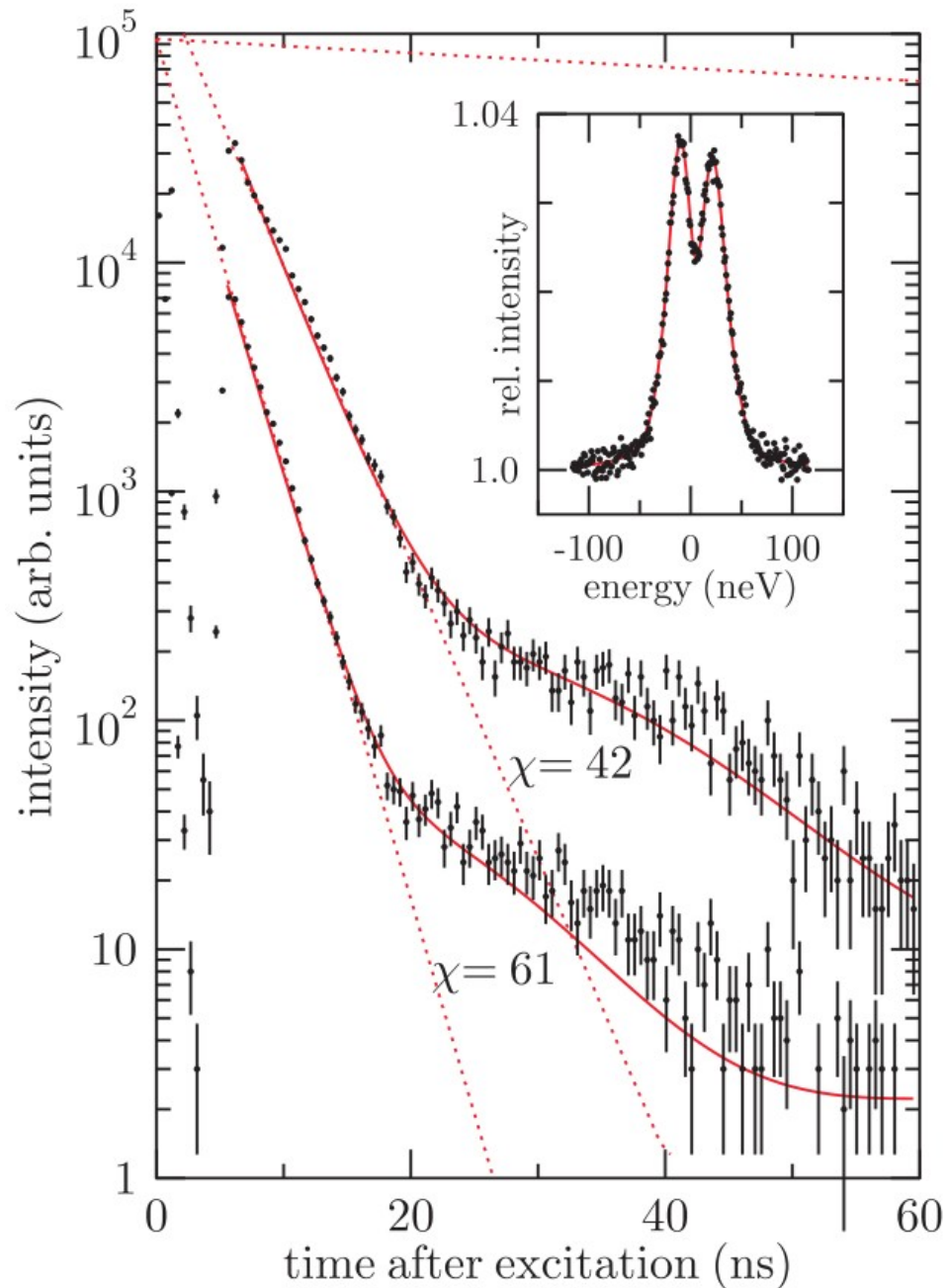
Movie 1

Movie 2

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

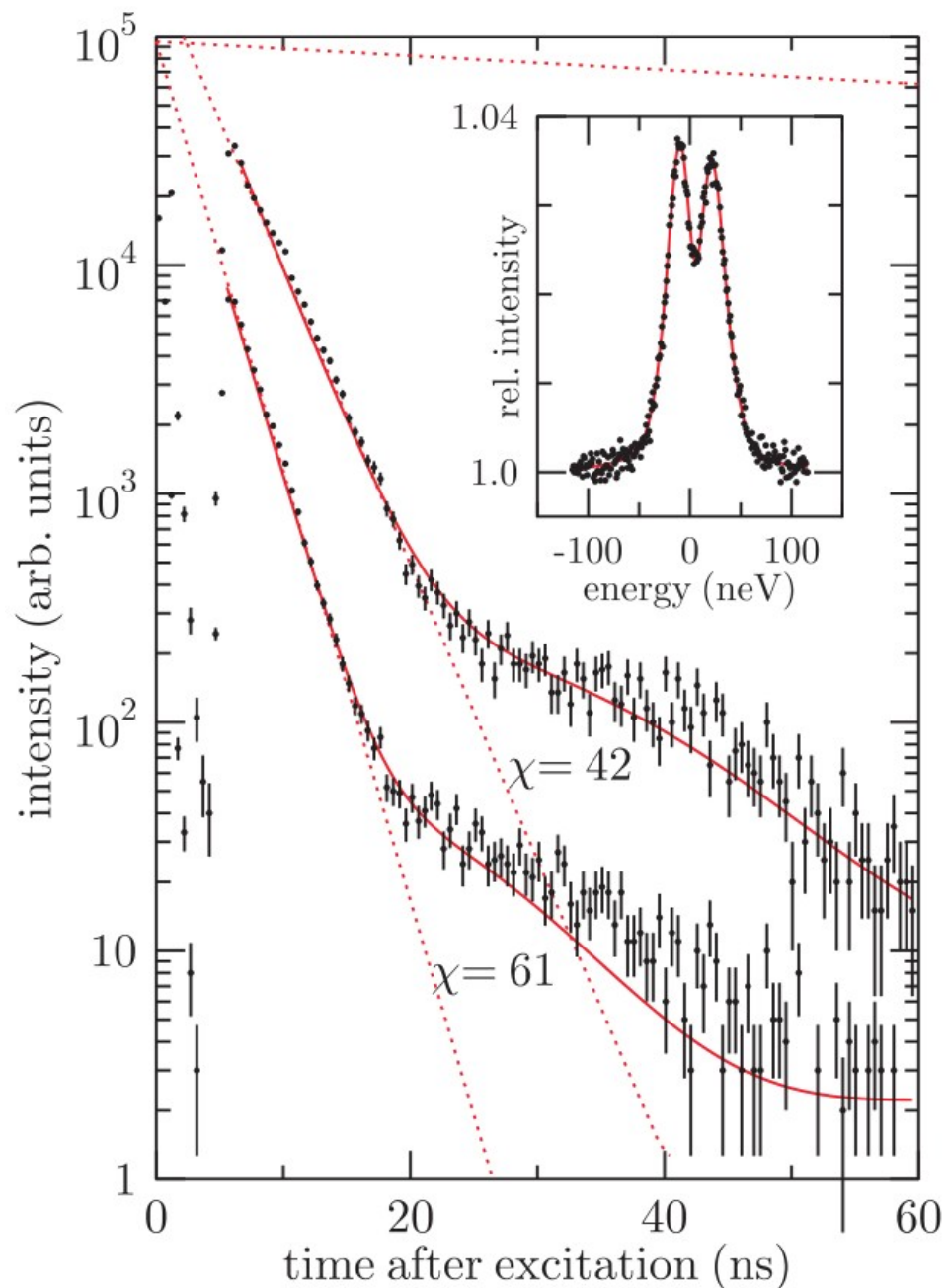


A single iron layer

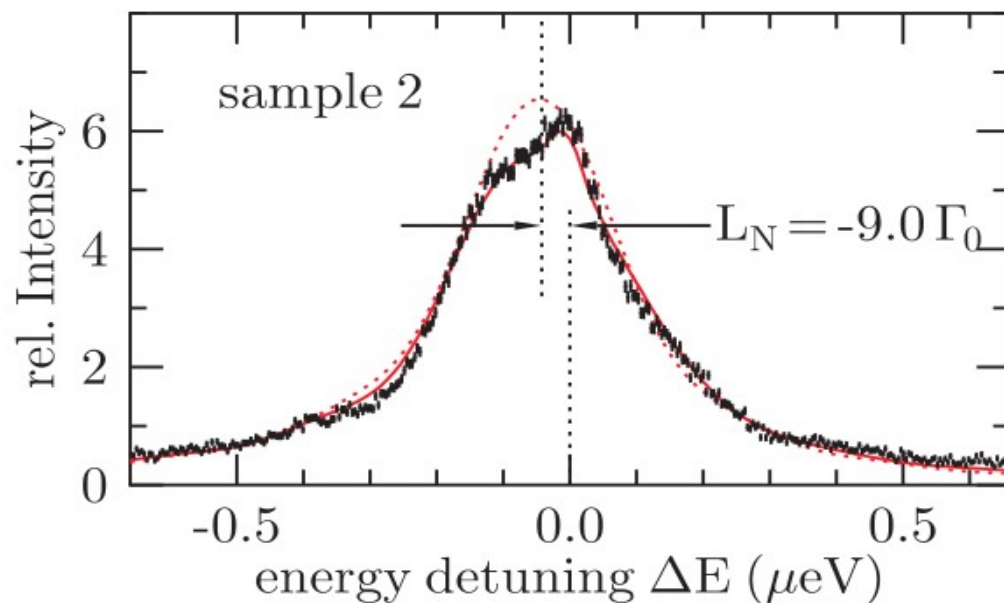


Effectively acts
as a two level
system!

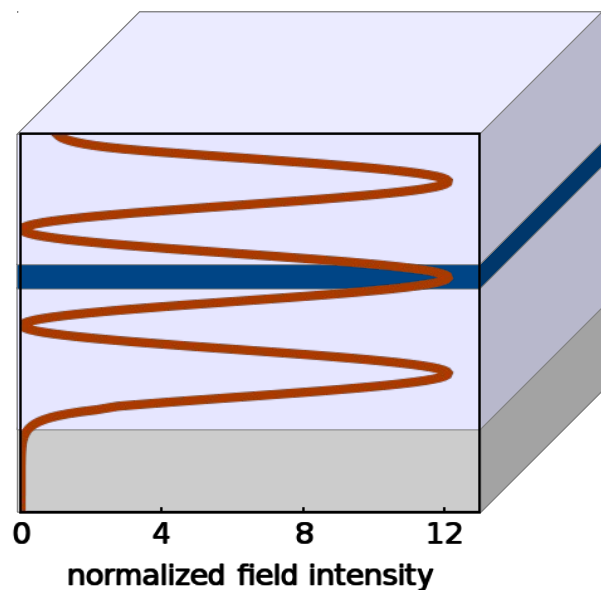
Cooperative effects



But with modified properties:
superradiance +
cooperative Lamb shift

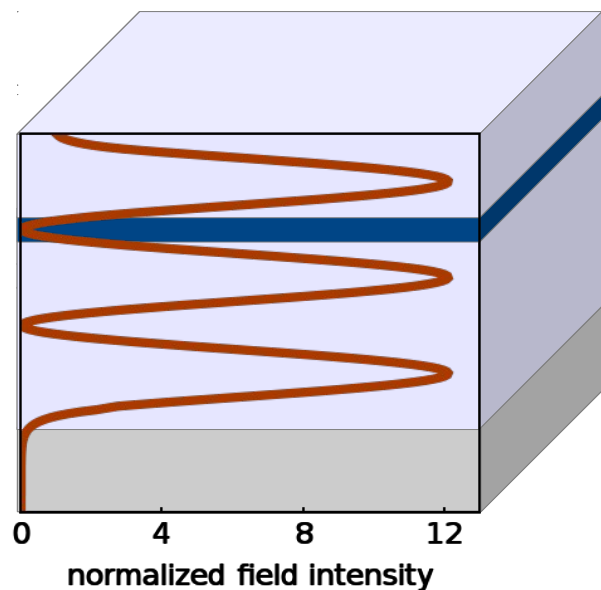


Tailoring the light-matter interaction



Iron nuclei strongly couple to cavity field

- ▶ Accelerated decay
- ▶ High excitation probability

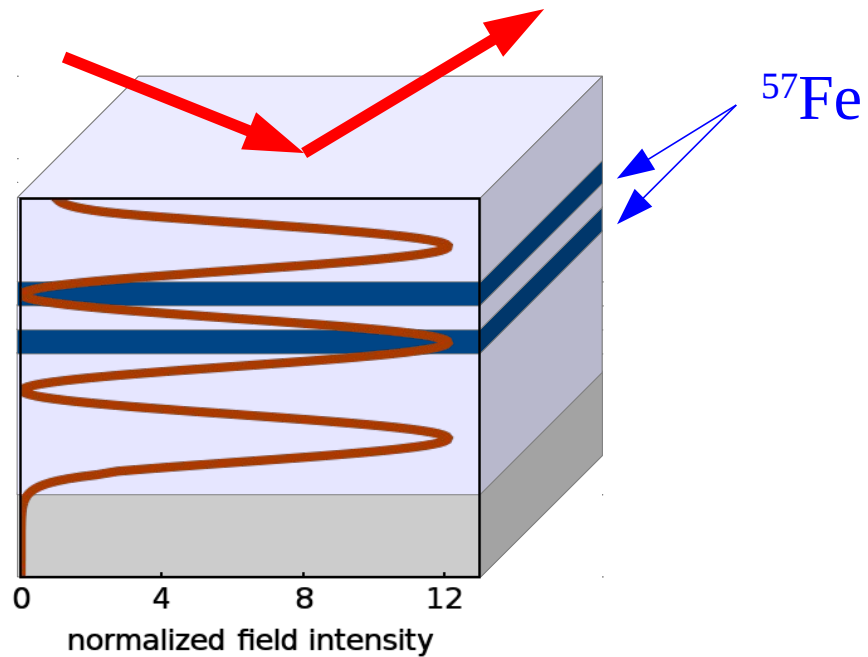


Iron nuclei weakly couple to cavity field

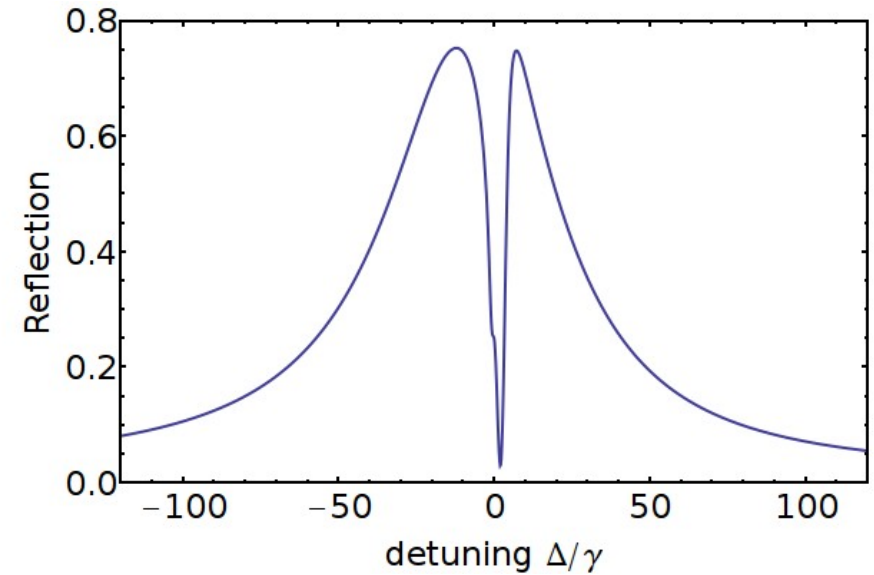
- ▶ Decelerated decay
- ▶ Low excitation probability

Effective properties of
the nuclei can be tailored!

Two iron layers

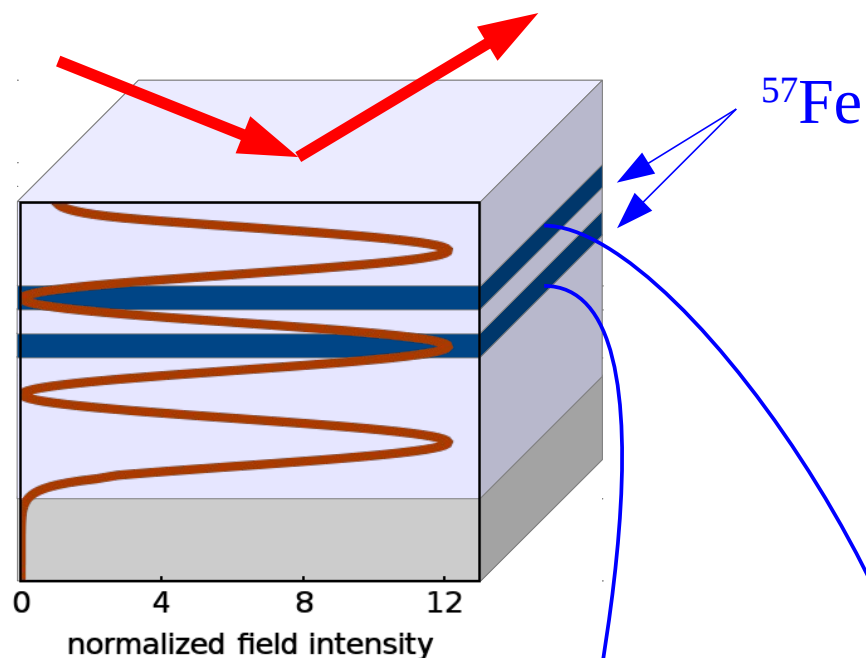


Reflection spectrum

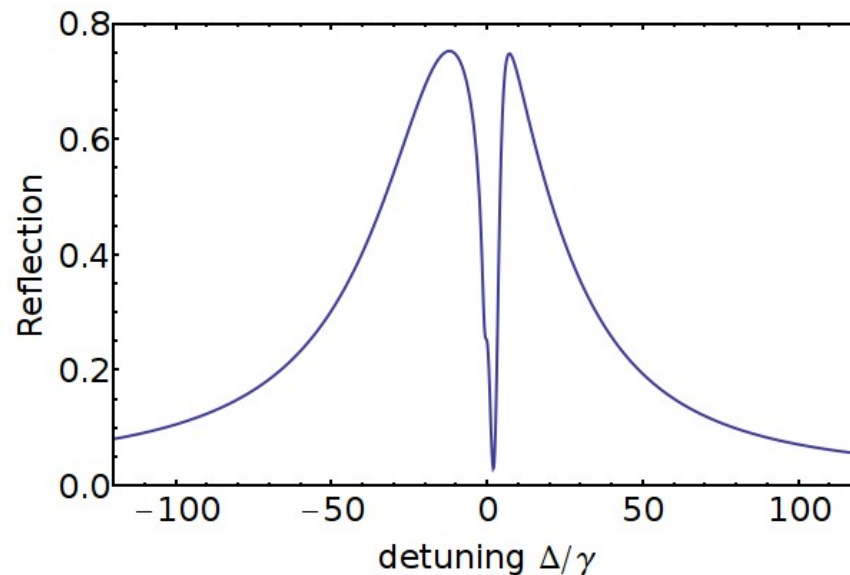


Looks like EIT!

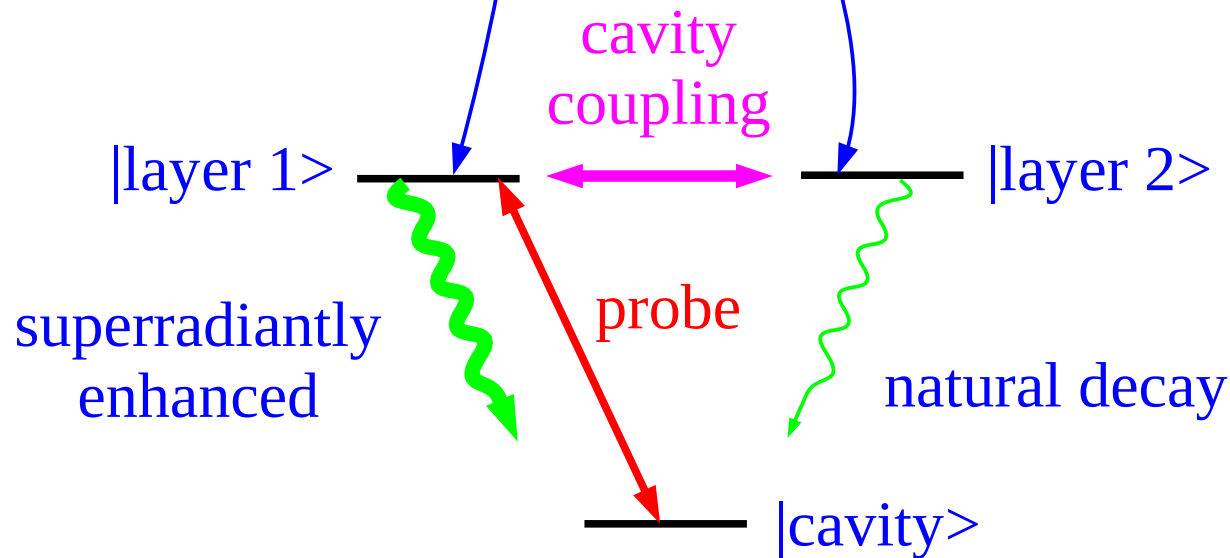
Two iron layers



Reflection spectrum

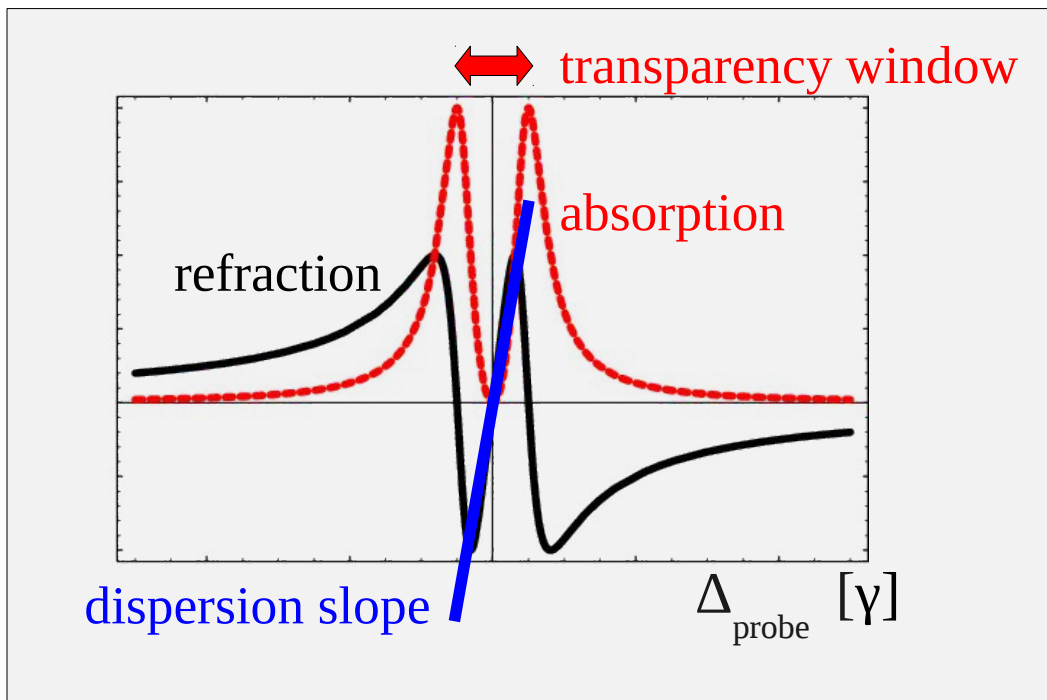


It is EIT!

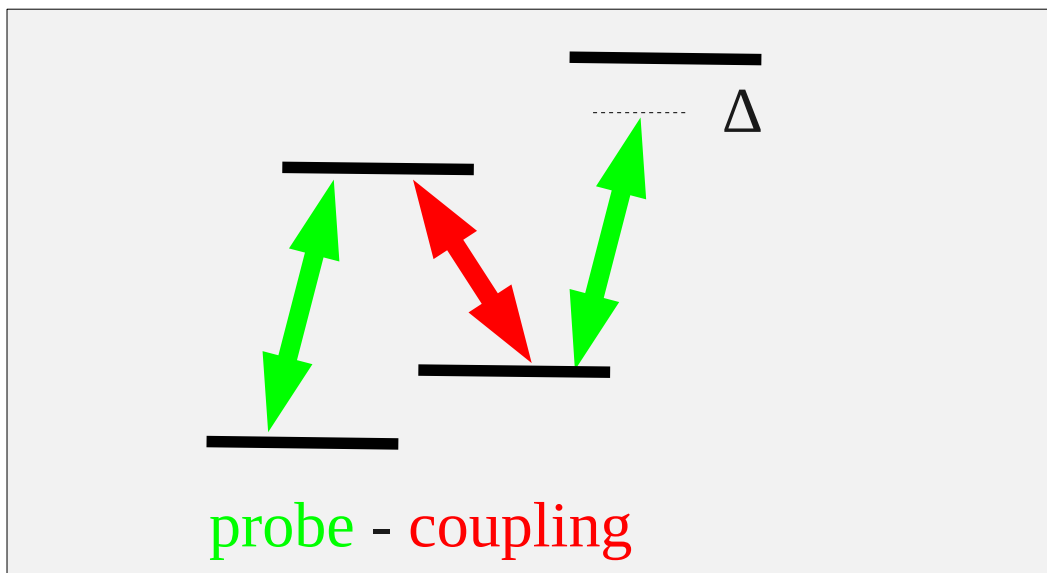


same nuclei
acquire
different
properties

What would be desirable?

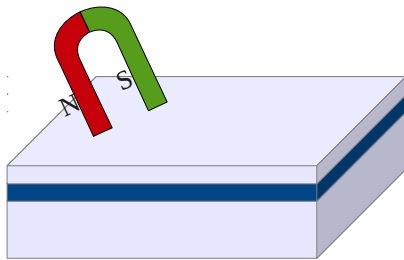


- ▶ Broad transparency window to propagate of short input pulses
- ▶ Steep dispersion slope for strong effect on propagated pulse
- ▶ (time delay)·(transparency bandwidth) is constant
→ need to tune for best trade-off

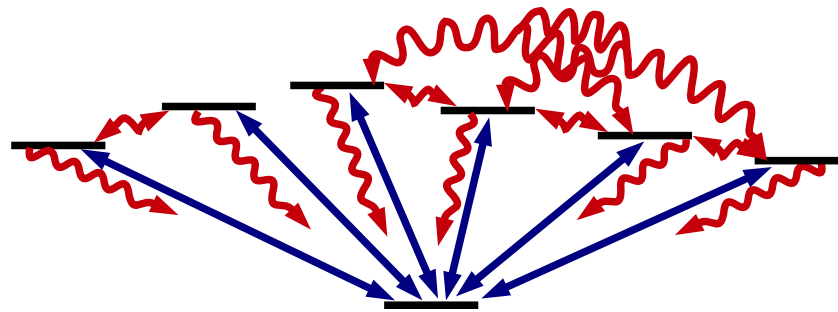


- ▶ More general level schemes offer wide range of applications
- ▶ Example: Strongly enhanced non-linear response

General quantum optical theory

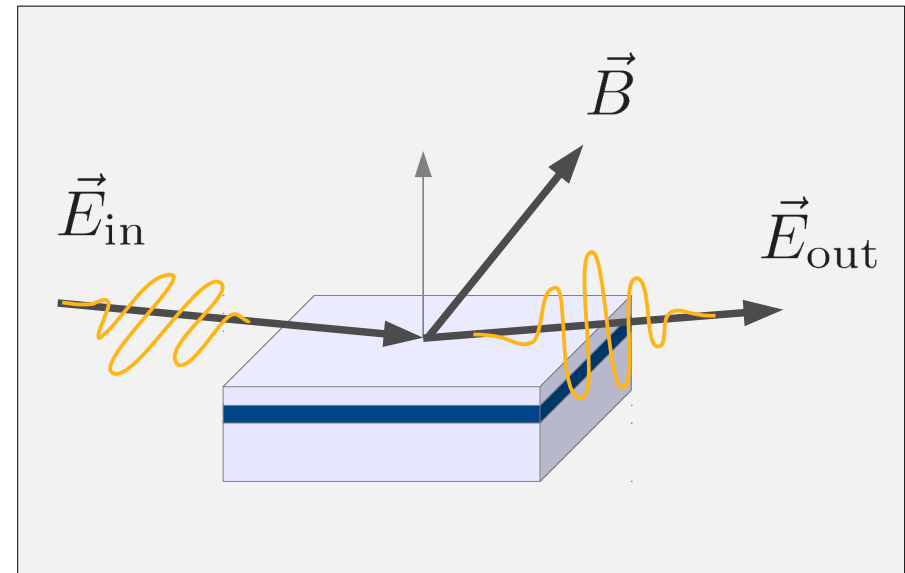
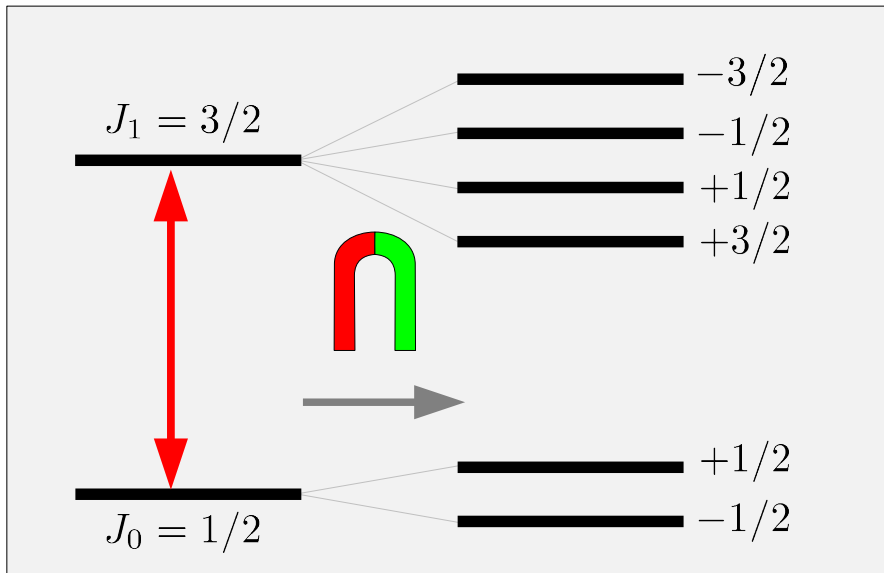


$\hat{\Delta}$



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

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

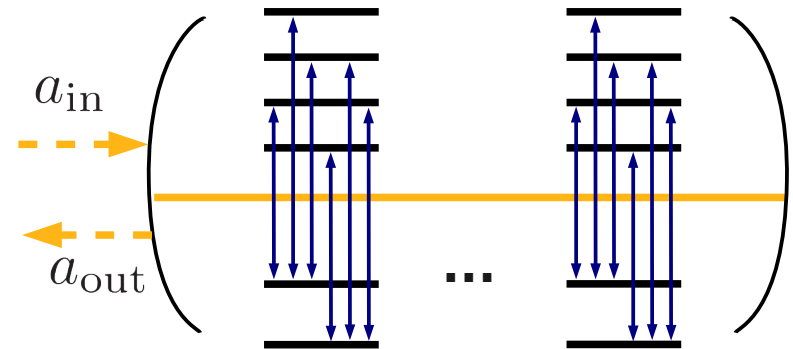
density matrix coherent evolution incoherent evolution

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

Use input-output formalism to calculate cavity response

$$R = \frac{\langle a_{\text{out}} \rangle}{\langle a_{\text{in}} \rangle}$$

reflectance



Gardiner, Zoller, *Quantum Noise*, Springer (2000)

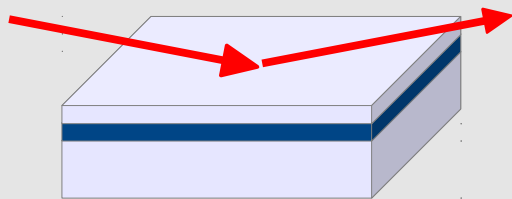
Constructing a quantum optical model

Limit of linear nuclear response to classical field:

Nuclear scattering
formalism

reflectivity

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

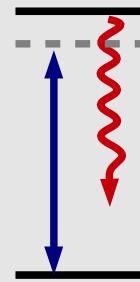


$\hat{=}$

Quantum optics
formalism

susceptibility

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



detuning Δ_{LS}

spontaneous
emission $N \cdot \gamma$

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!

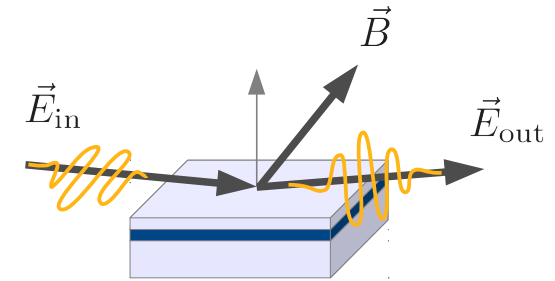
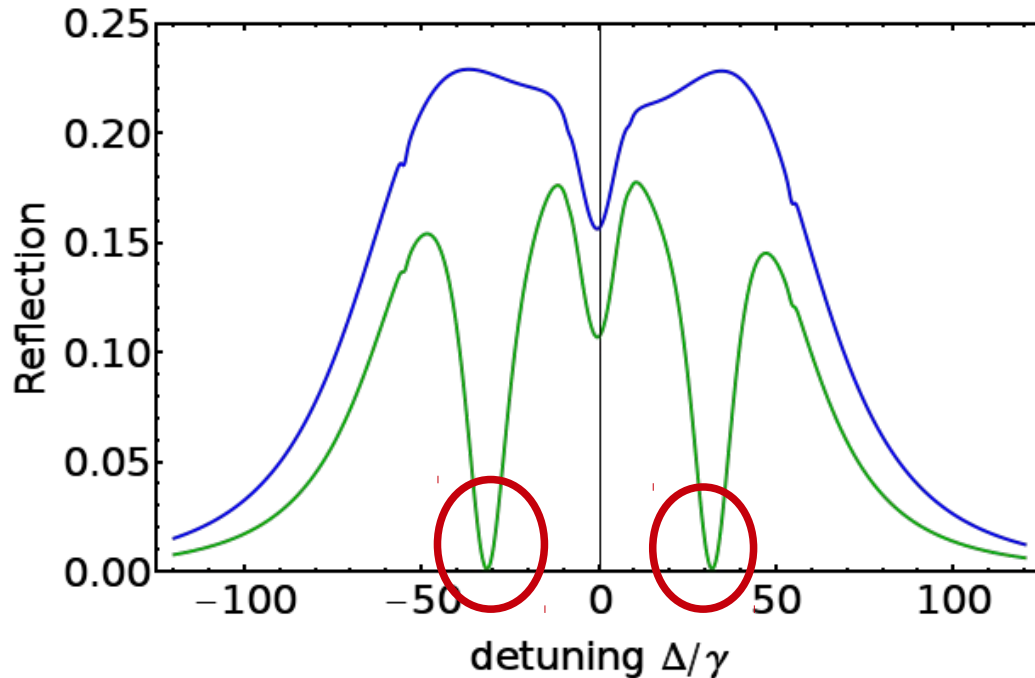
Δ_{LS}

Δ_{LS}

$N \cdot \gamma$

emission

Unexpected spectral signatures



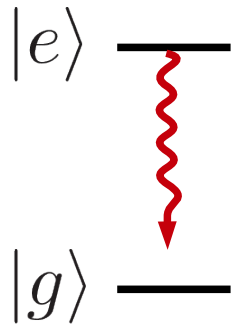
$$\vec{B} \parallel \hat{k}$$

$$\vec{B} \parallel \hat{k} + \hat{E}_{in}$$

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

Quantum optical model: Master equation

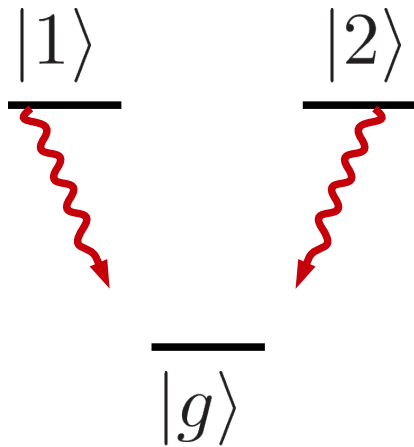
Two-level system



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

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

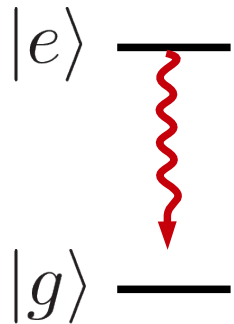
Three-level system



$$\begin{aligned} \frac{\partial}{\partial t}\rho = & -\frac{\gamma}{2} (|1\rangle\langle 1|\rho + \rho|1\rangle\langle 1| - 2|g\rangle\langle 1|\rho|1\rangle\langle g|) \\ & -\frac{\gamma}{2} (|2\rangle\langle 2|\rho + \rho|2\rangle\langle 2| - 2|g\rangle\langle 2|\rho|2\rangle\langle g|) \end{aligned}$$

Quantum optical model: Master equation

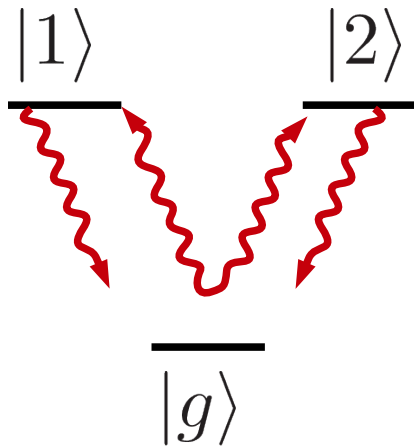
Two-level system



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

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

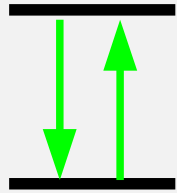
Three-level system



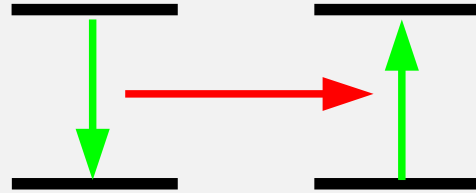
$$\begin{aligned} \frac{\partial}{\partial t}\rho = & -\frac{\gamma}{2}(|1\rangle\langle 1|\rho + \rho|1\rangle\langle 1| - 2|g\rangle\langle 1|\rho|1\rangle\langle g|) \\ & -\frac{\gamma}{2}(|2\rangle\langle 2|\rho + \rho|2\rangle\langle 2| - 2|g\rangle\langle 2|\rho|2\rangle\langle g|) \\ & -\frac{\gamma_C}{2}(|1\rangle\langle 2|\rho + \rho|1\rangle\langle 2| - 2|g\rangle\langle 2|\rho|1\rangle\langle g|) \\ & -\frac{\gamma_C}{2}(|2\rangle\langle 1|\rho + \rho|2\rangle\langle 1| - 2|g\rangle\langle 1|\rho|2\rangle\langle g|) \end{aligned}$$

Find additional terms!

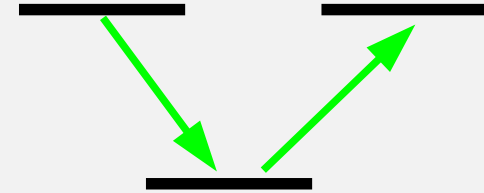
Fundamental light-matter interactions



decay, Lamb shift
(same atom, same transition)



dipole-dipole interaction
(other atom)

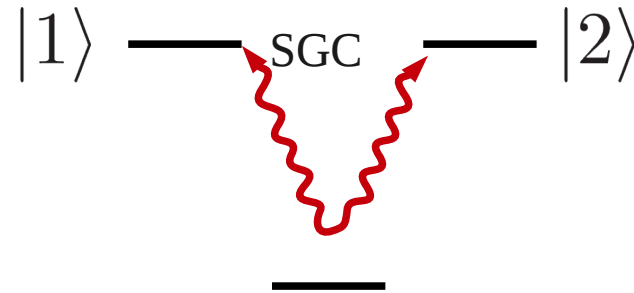


spontaneous coherences
(same atom, different transition)

- ▶ **Spontaneously generated 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
-

Conditions for SGC

Requirements for SGC



Condition I

$$\vec{d}_1 \cdot \vec{d}_2 \neq 0$$

non-orthogonal
dipole moments

Condition II

$$E_1 \approx E_2$$

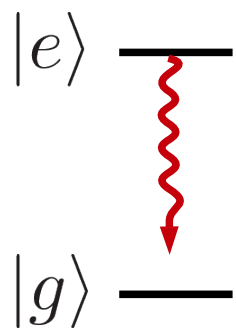
approx. same
transition energy

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

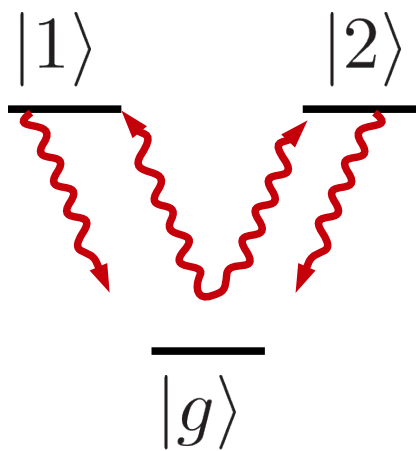
Two-level system



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

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

Three-level system



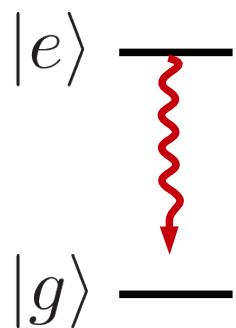
$$\begin{aligned} \frac{\partial}{\partial t} \rho = & -\frac{\gamma}{2} (|1\rangle\langle 1|\rho + \rho|1\rangle\langle 1| - 2|g\rangle\langle 1|\rho|1\rangle\langle g|) \\ & -\frac{\gamma}{2} (|2\rangle\langle 2|\rho + \rho|2\rangle\langle 2| - 2|g\rangle\langle 2|\rho|2\rangle\langle g|) \end{aligned}$$

$$\begin{aligned} & -\frac{\gamma_C}{2} (|1\rangle\langle 2|\rho + \rho|1\rangle\langle 2| - 2|g\rangle\langle 2|\rho|1\rangle\langle g|) \\ & -\frac{\gamma_C}{2} (|2\rangle\langle 1|\rho + \rho|2\rangle\langle 1| - 2|g\rangle\langle 1|\rho|2\rangle\langle g|) \end{aligned}$$

Find additional terms!

Quantum optical model: Master equation

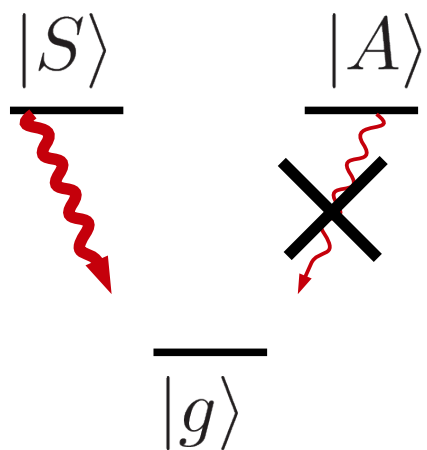
Two-level system



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

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

Three-level system with SGC, diagonalized



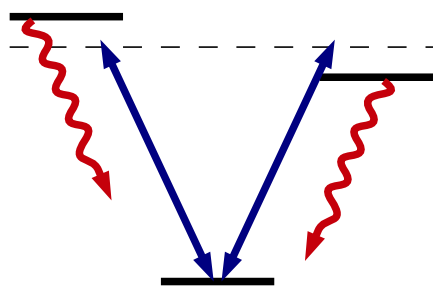
$$\frac{\partial}{\partial t} \rho = -\frac{\gamma + \gamma_C}{2} (|S\rangle\langle S|\rho + \rho|S\rangle\langle S| - 2|g\rangle\langle S|\rho|S\rangle\langle g|) - \frac{\gamma - \gamma_C}{2} (|A\rangle\langle A|\rho + \rho|A\rangle\langle A| - 2|g\rangle\langle A|\rho|A\rangle\langle g|)$$

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

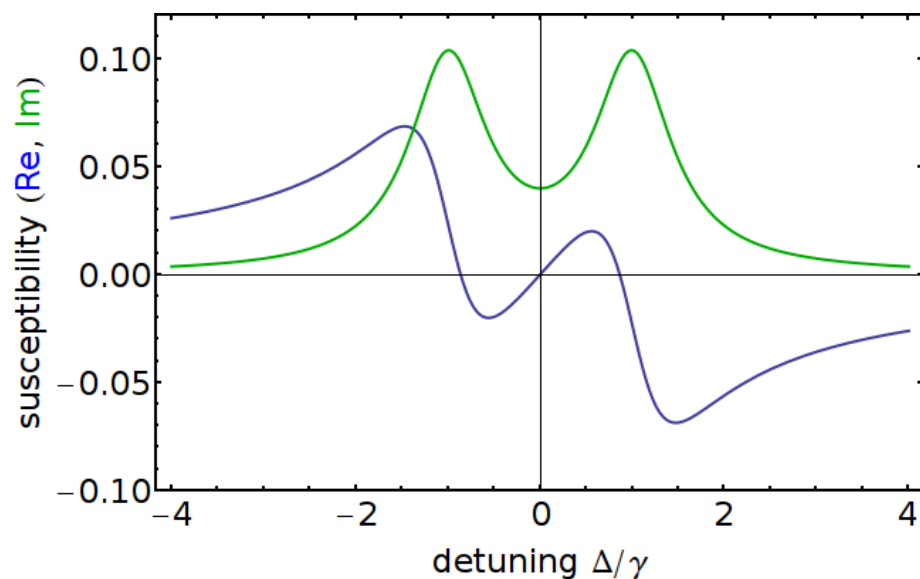
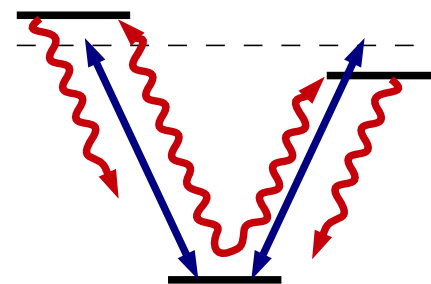
$$|A\rangle = \frac{1}{\sqrt{2}} (|1\rangle - |2\rangle) \quad \text{destructive interference} \\ \text{dark line in spectrum}$$

Susceptibility with and without SGC

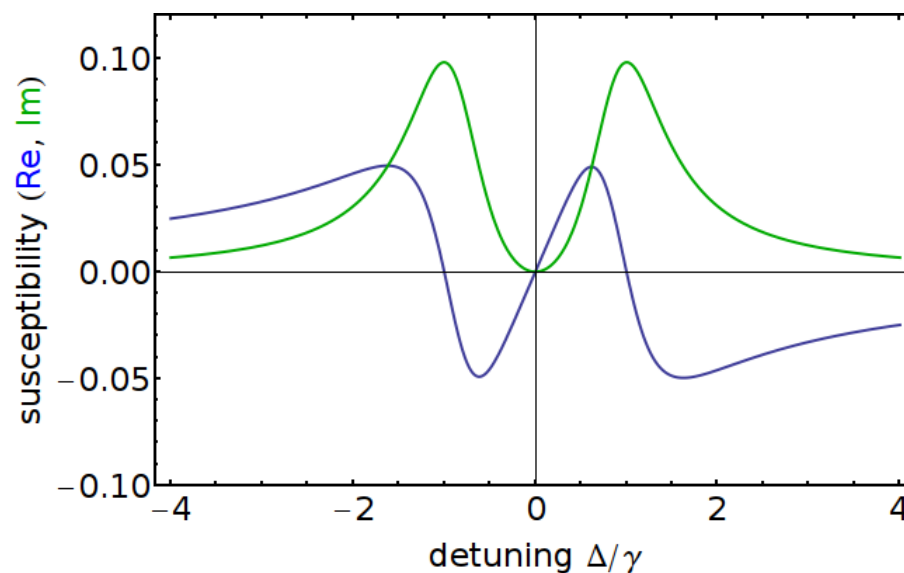
Without SGC



With SGC



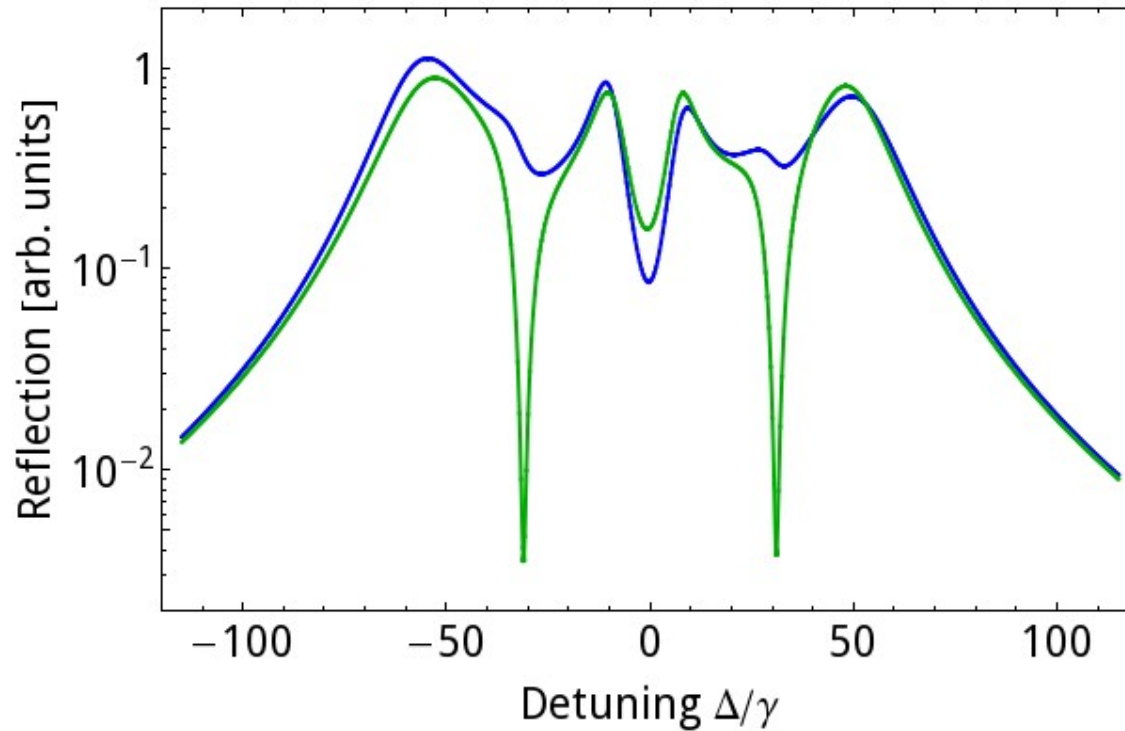
Incoherent sum
of two resonances



Interference
→ observed with nuclei

SGC is essential in our setup

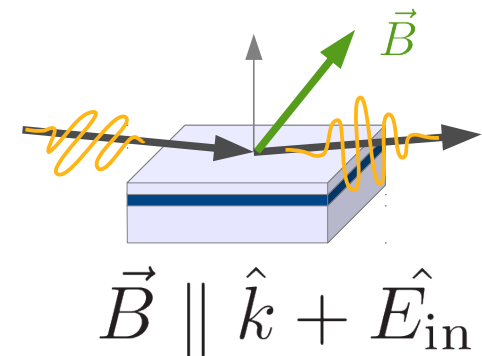
Reflectance (log scale)



Detuning

With SGC

Without SGC

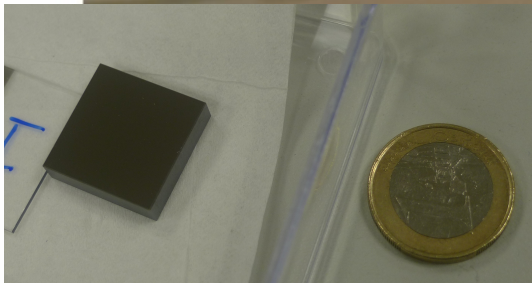
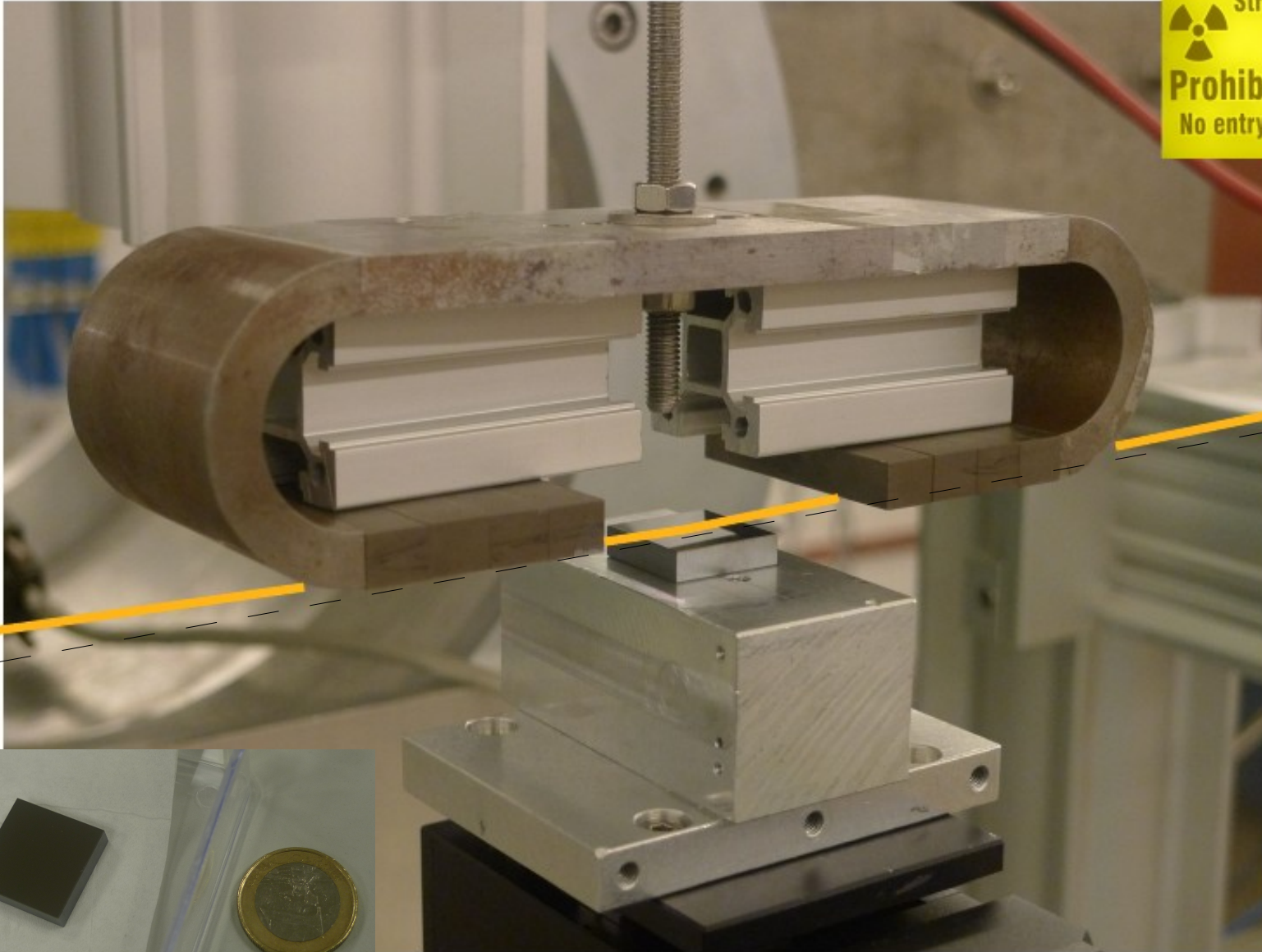


Results with / without SGC
differ strongly

Experiment at PETRA III (DESY Hamburg)



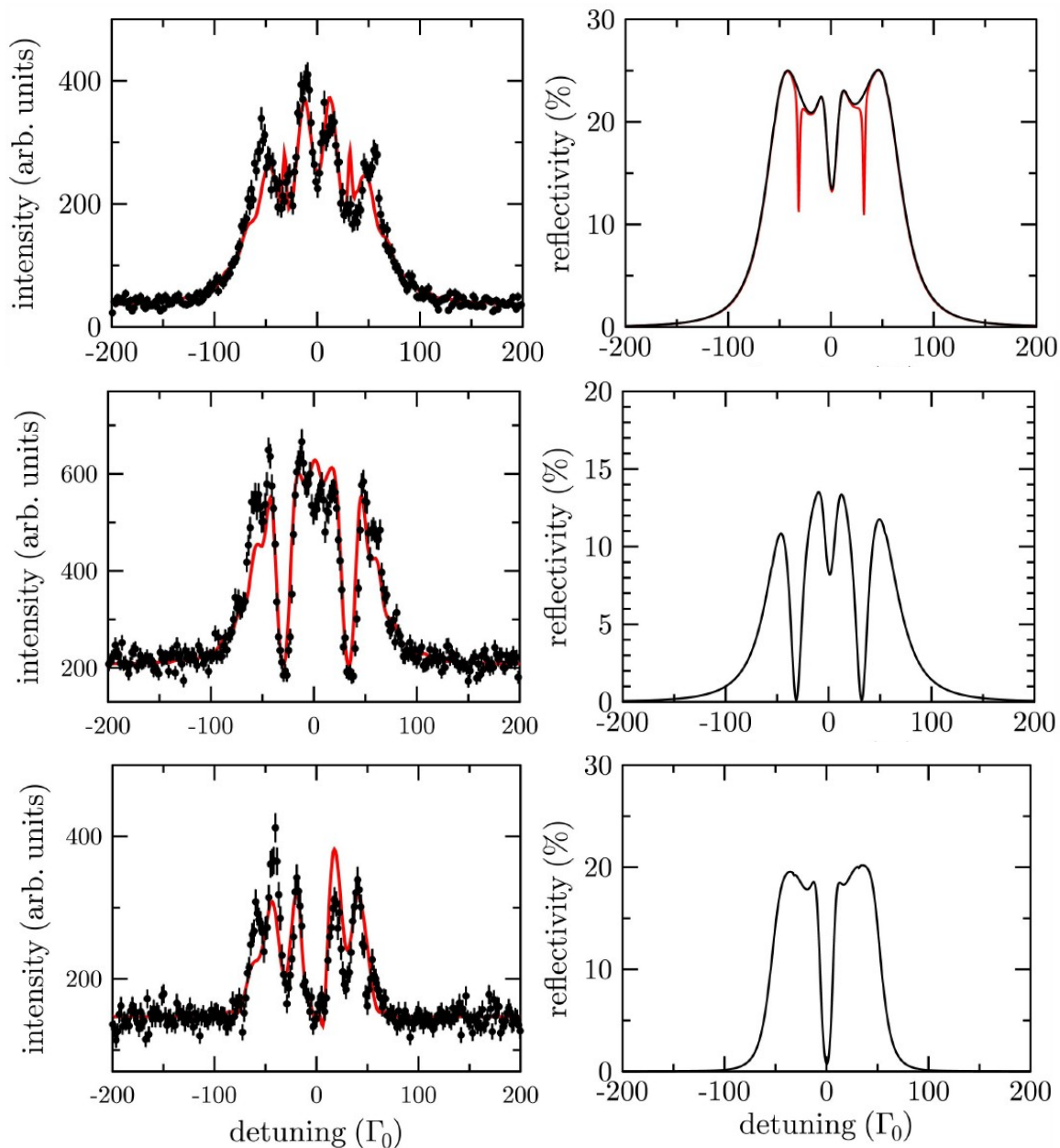
Experiment at PETRA III (DESY Hamburg)



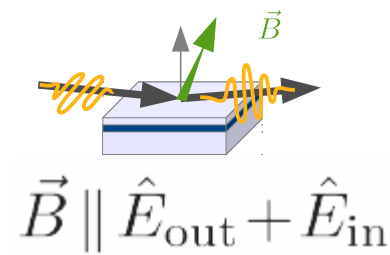
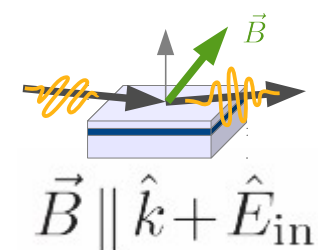
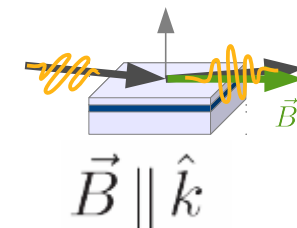
Experimental results

Experiment+theory

(including exp. details)

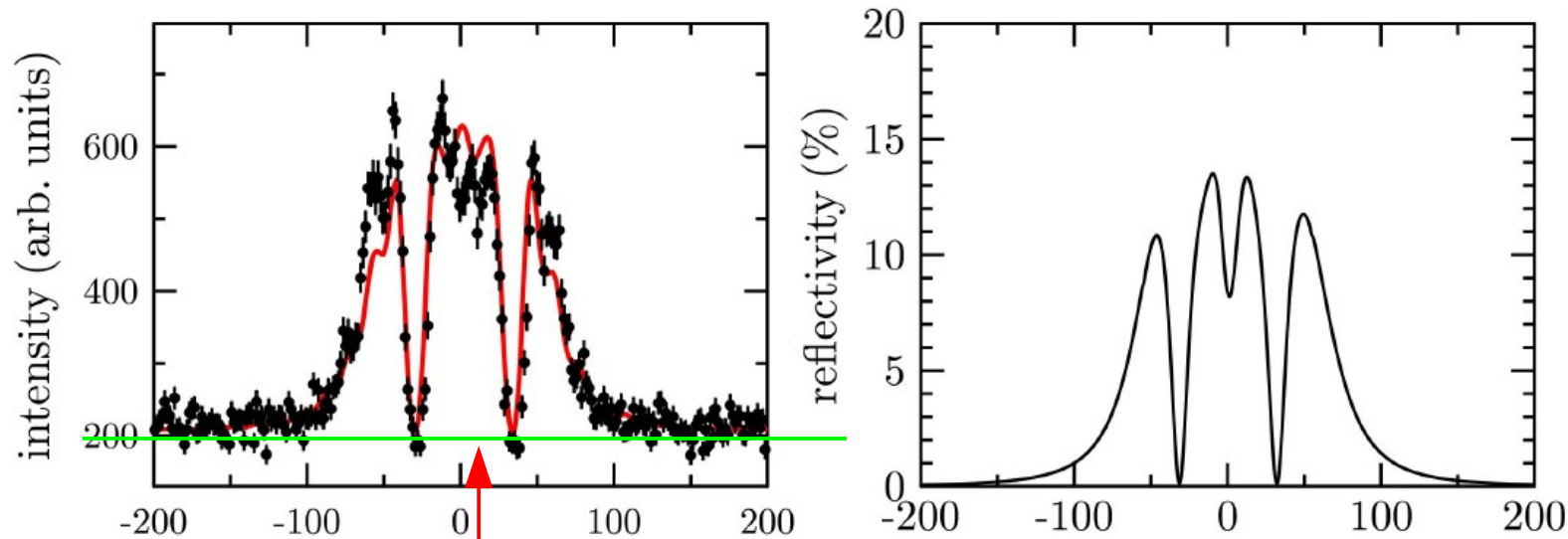


Plain theory



First observation of such SGC!

Experimental results

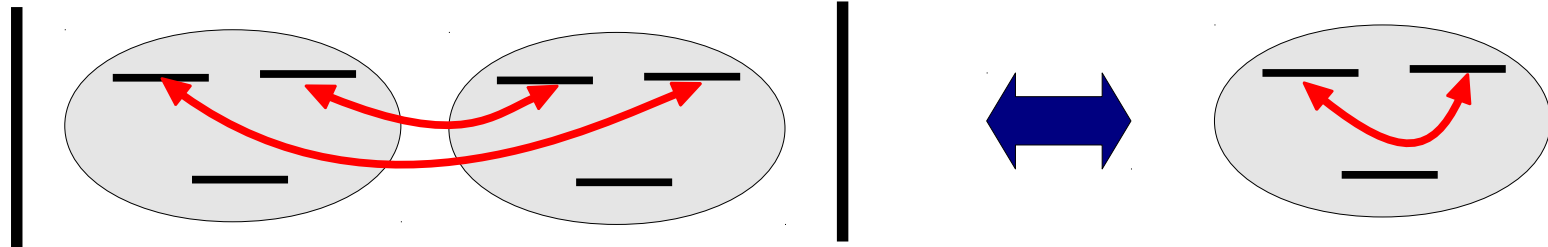


really zero -
clean system,
no decoherence!

This indicates
that essentially
no decoherence
occurs
IDEAL CASE

Why can SGC be observed in nuclei?

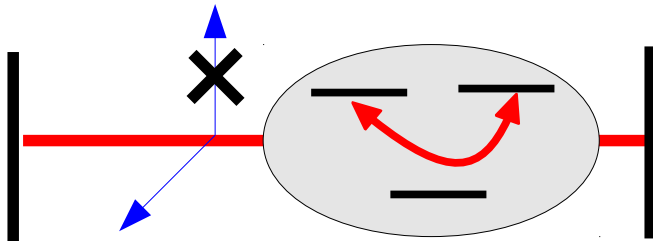
First mechanism: “Quantum simulator”



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

probed from outside appears as single system with SGC

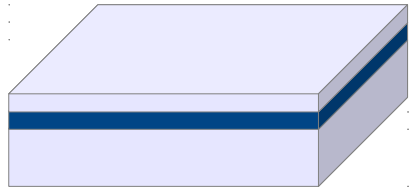
Second mechanism: “Anisotropic vacuum”



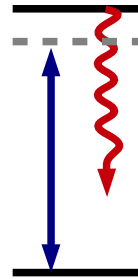
SGC can appear in atoms in anisotropic environments (proposal by G. S. Agarwal)

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

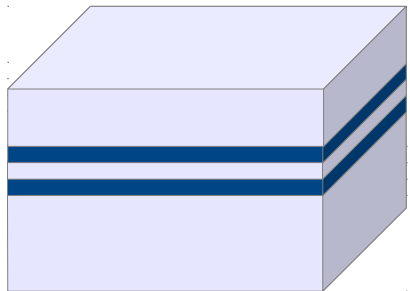
X-ray waveguides: Present status



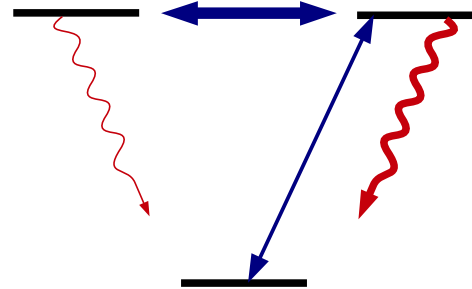
$\hat{=}$



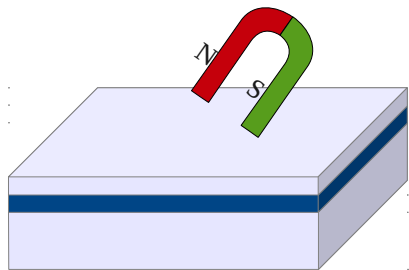
- Superradiance
- Cooperative Lamb shift (first observation)



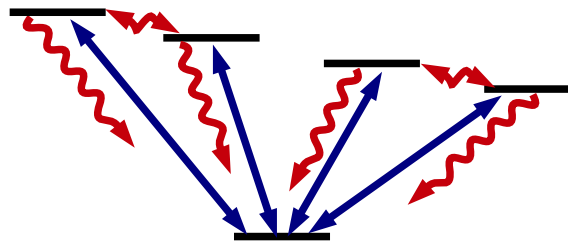
$\hat{=}$



- EIT
- Novel mechanism to tailor level schemes

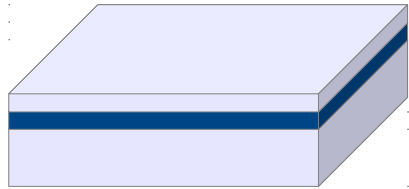


$\hat{=}$

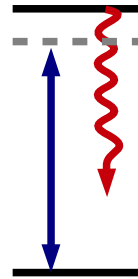


- Externally tunable level schemes
- Implementation and first direct observation of SGC

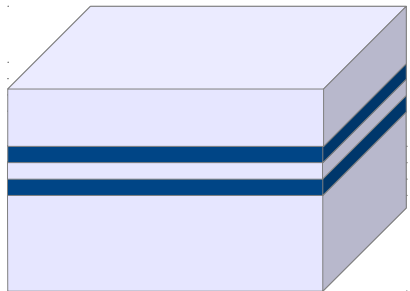
X-ray waveguides: Present status



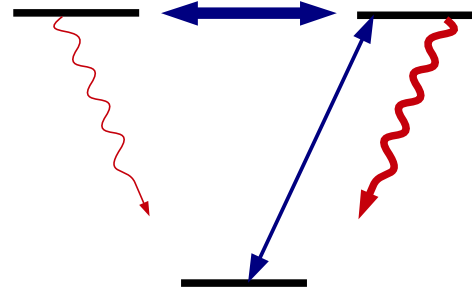
\cong



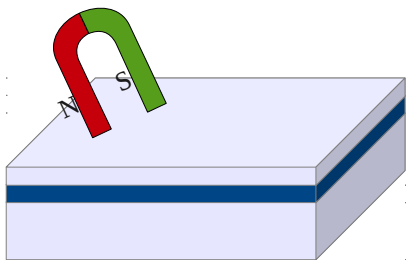
- Superradiance
- Cooperative Lamb shift (first observation)



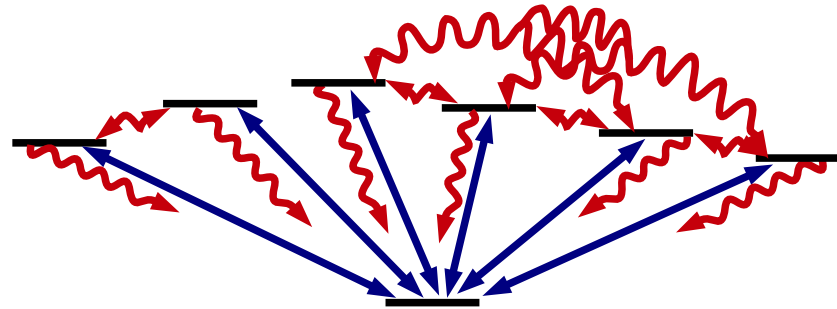
\cong



- EIT
- Novel mechanism to tailor level schemes

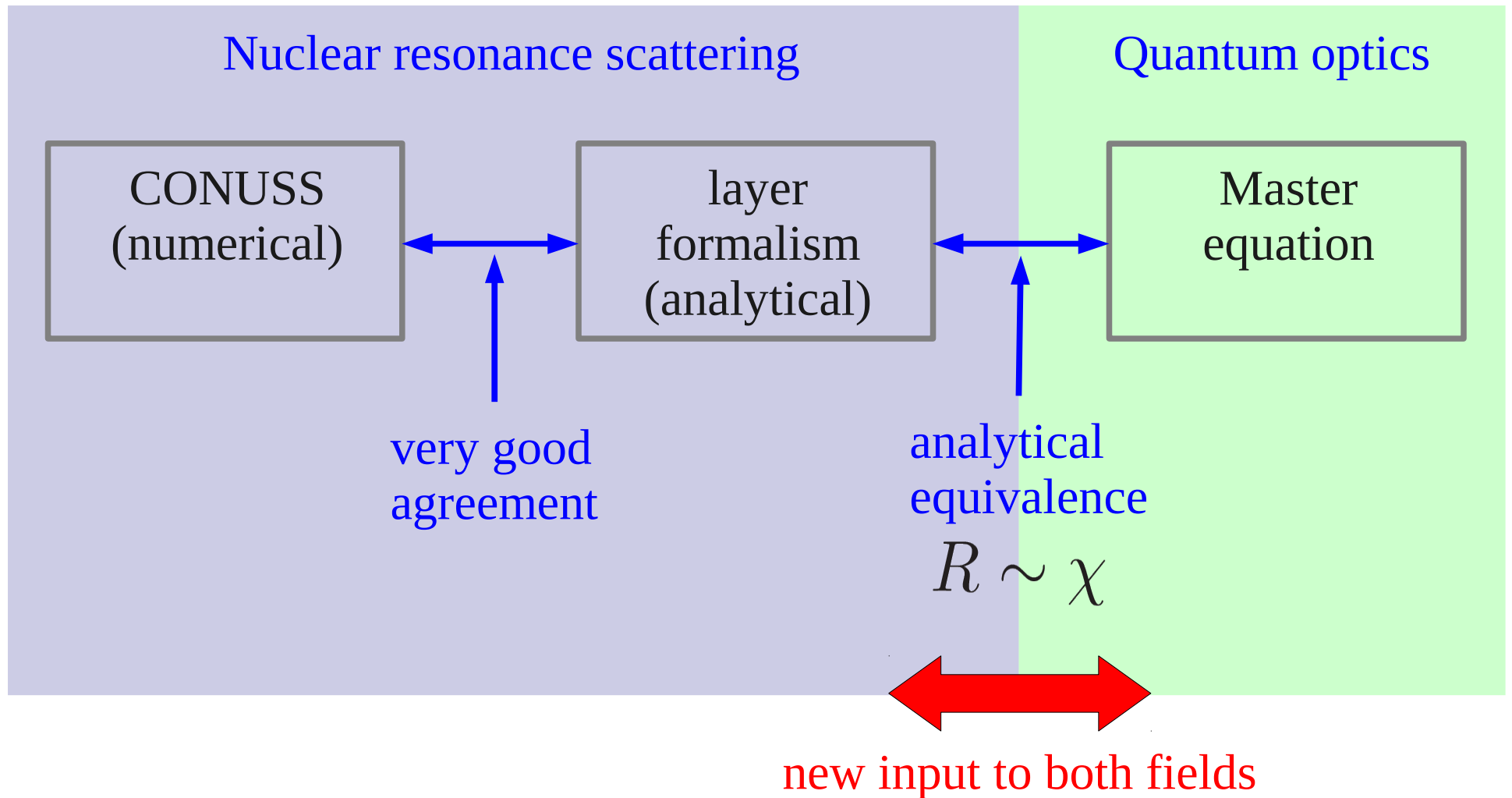


\cong



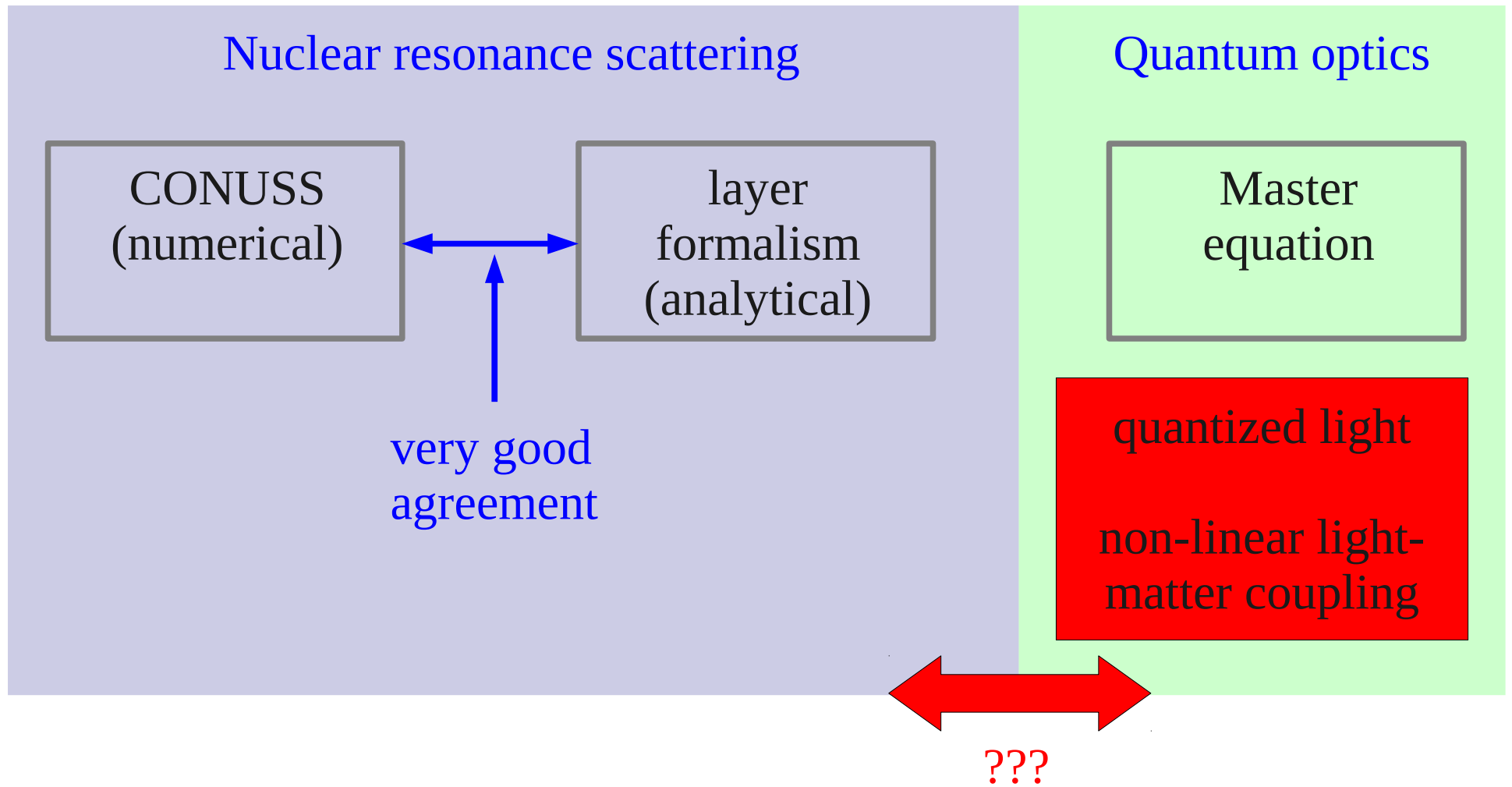
- Externally tunable level schemes
- Implementation and first direct observation of SGC

X-ray waveguides: Present status



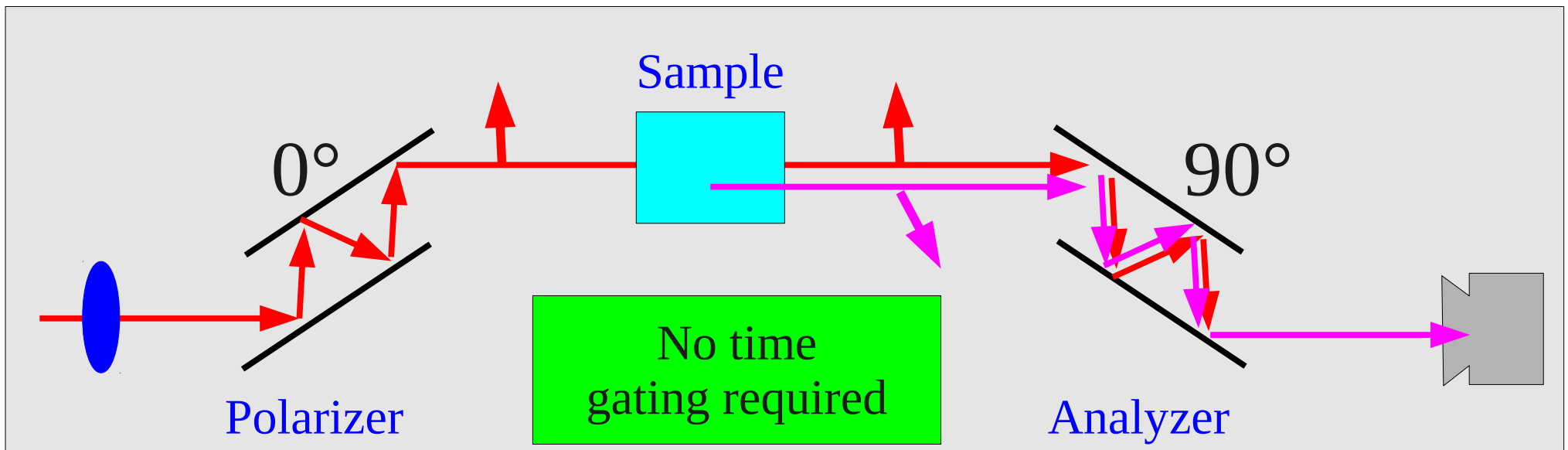
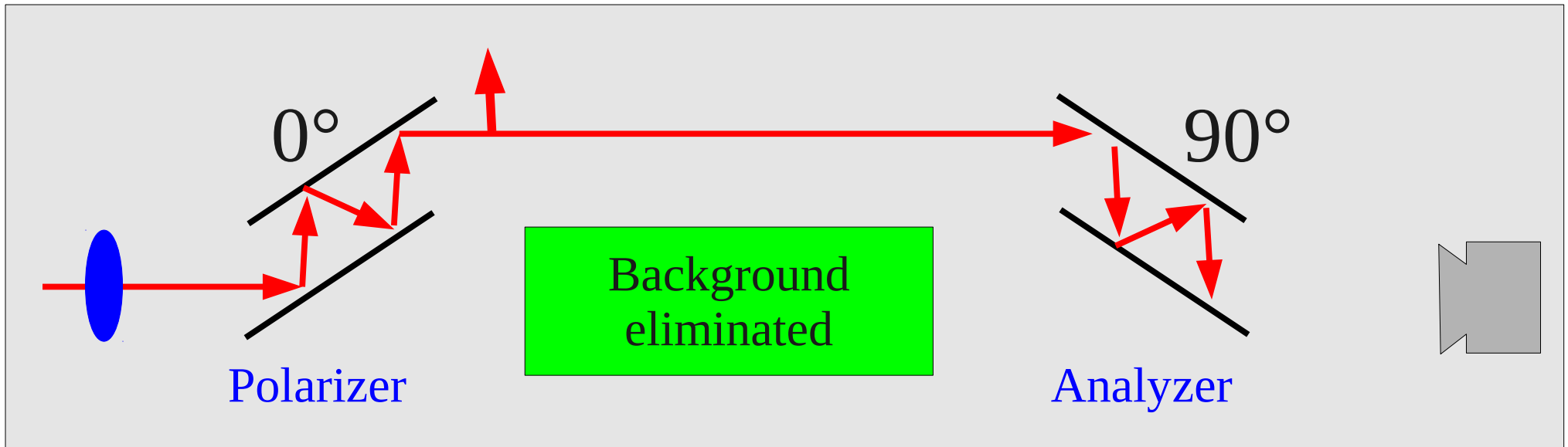
promising basis for implementation of advanced quantum optical techniques in hard x-ray range

X-ray waveguides: Present status

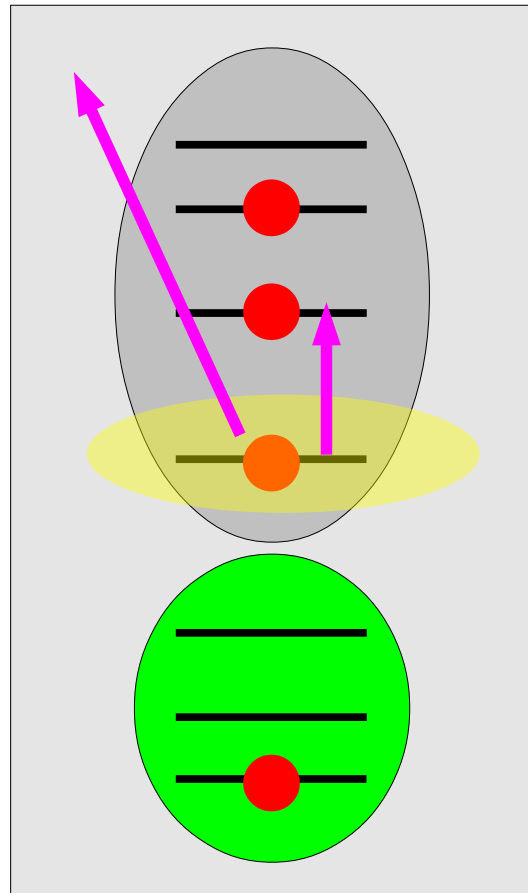


promising basis for implementation of advanced quantum optical techniques in hard x-ray range

Crossed polarimeter setup



Alternatives to nuclei



Inner-shell electrons

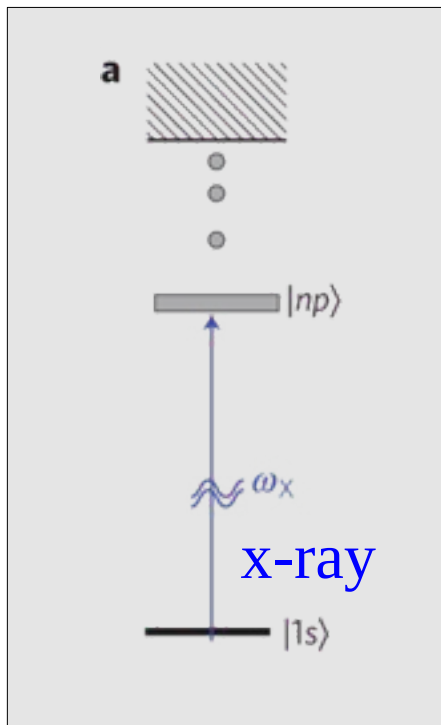
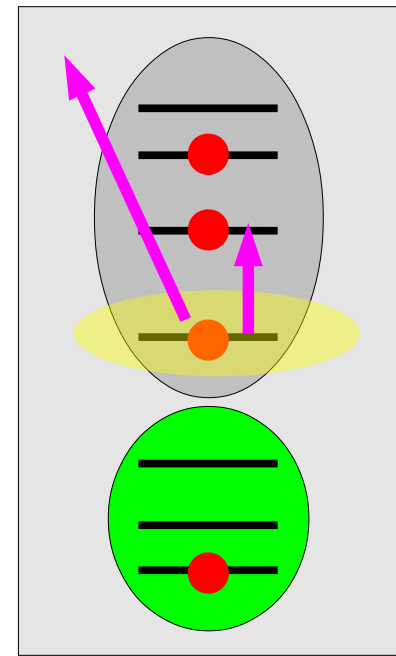
nature
physics

ARTICLES

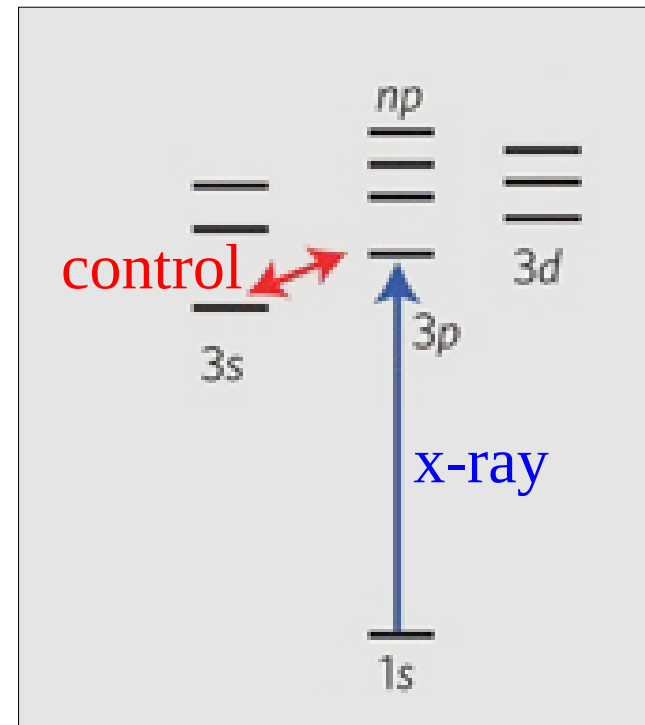
PUBLISHED ONLINE: 25 OCTOBER 2009 | DOI: 10.1038/NPHYS1430

Controlling X-rays with light

T. E. Glover¹, M. P. Hertlein¹, S. H. Southworth², T. K. Allison^{1,3}, J. van Tilborg¹, E. P. Kanter²,
B. Krässig², H. R. Varma², B. Rude¹, R. Santra^{2,4}, A. Belkacem¹ and L. Young^{2*}

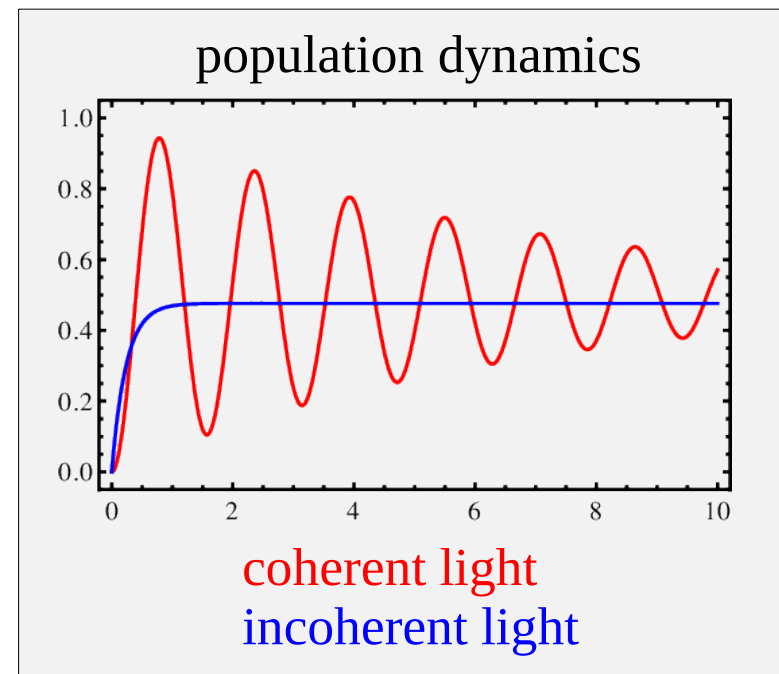
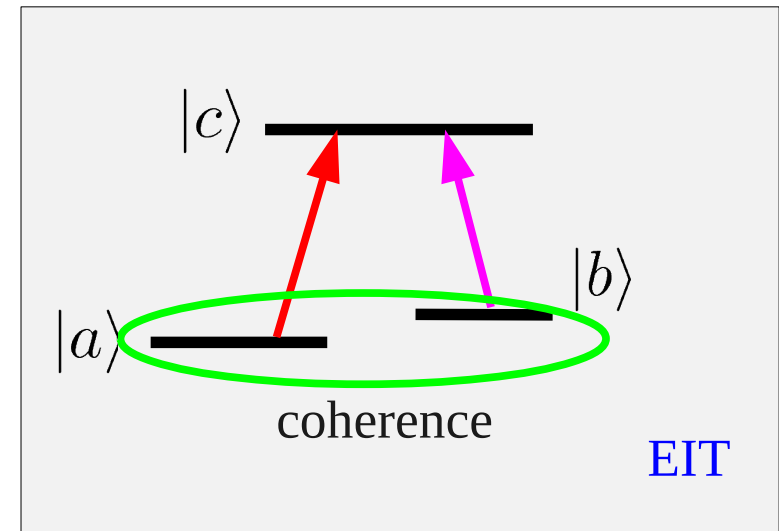


control x-ray
absorption
with light?



Temporal coherence

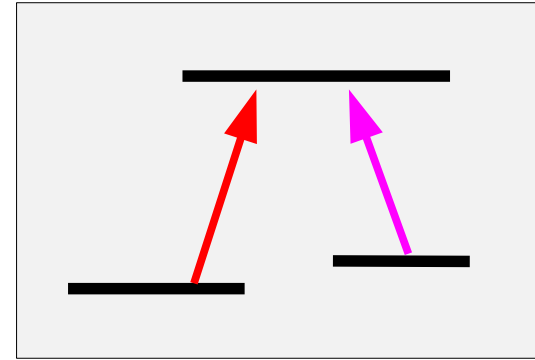
- ▶ All quantum optical effects rely on coherence and interference
- ▶ Synchrotron experiments operates at the single photon level, and single photons interfere with themselves
- ▶ But: strong and coherent driving is key to most quantum optical effects
- ▶ Availability of temporally coherent pulse with 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

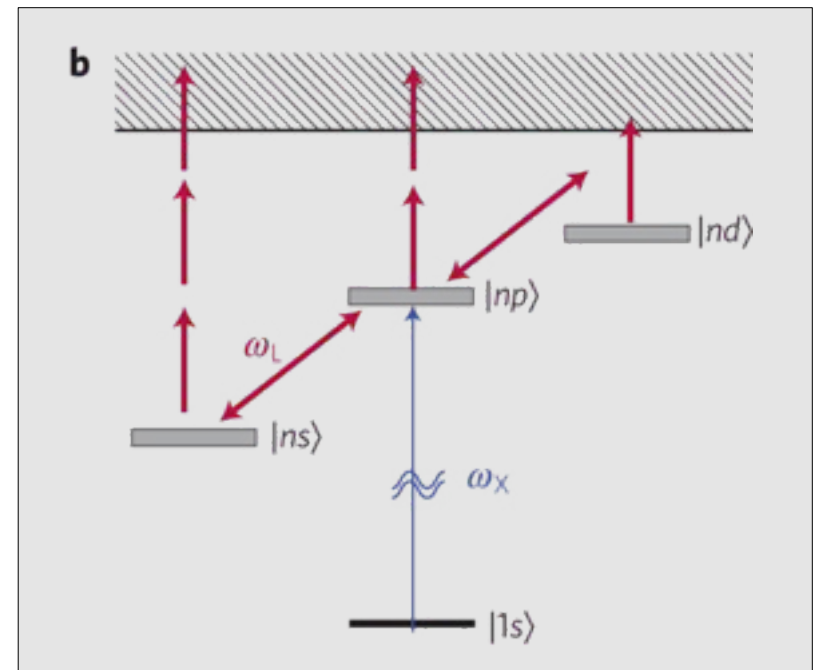


Level structure

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

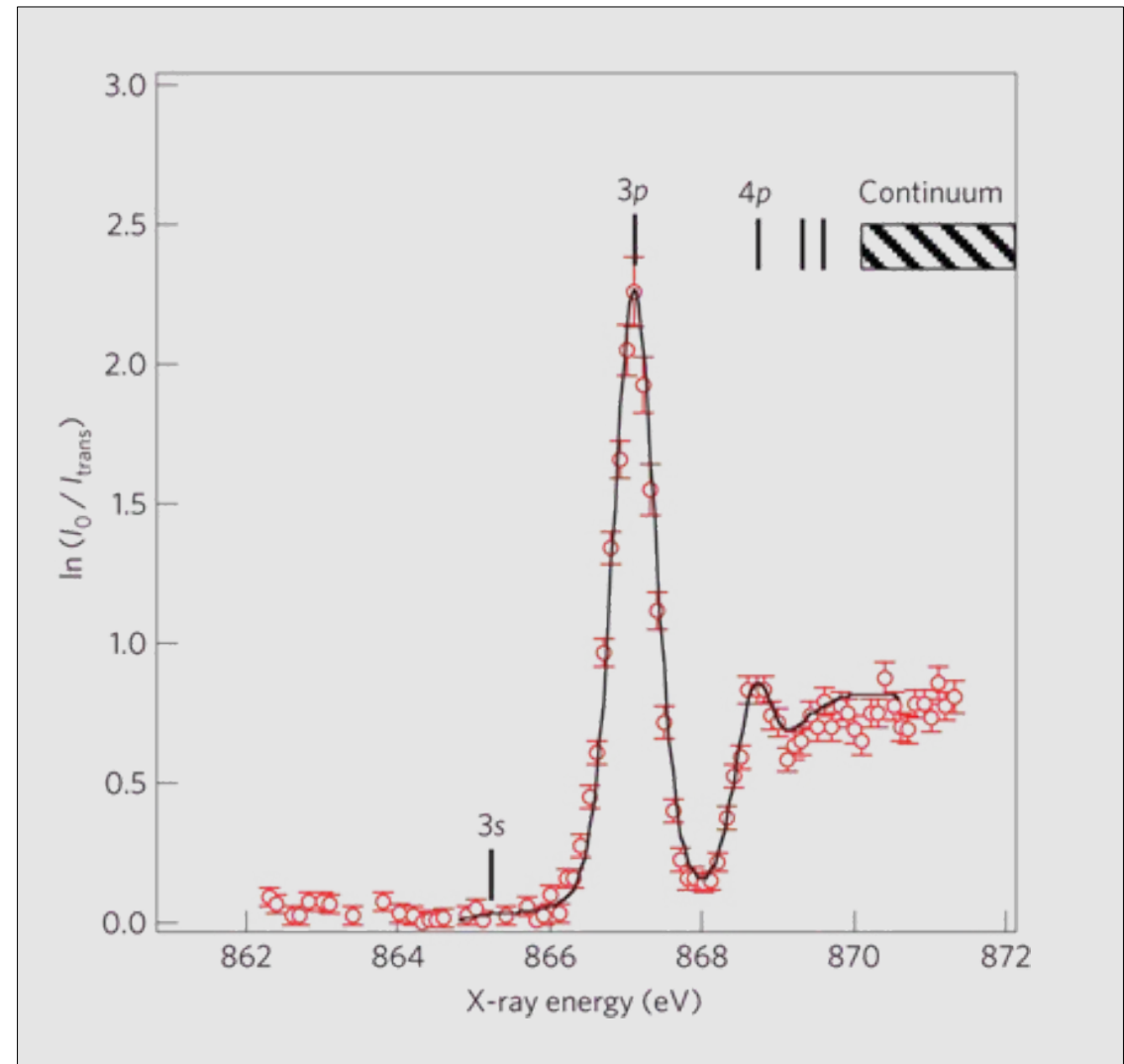
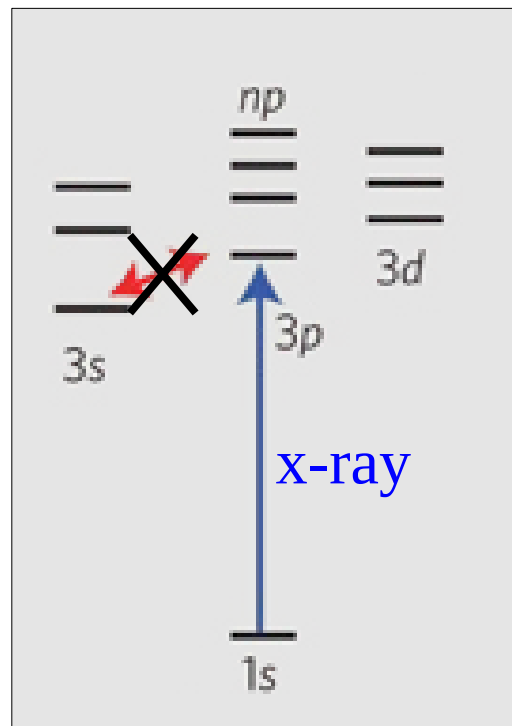
Fast timescale

- ▶ Core hole life time 2.4fs
- ▶ Need ultrafast x-ray and control laser
- ▶ Need to synchronize the two laser



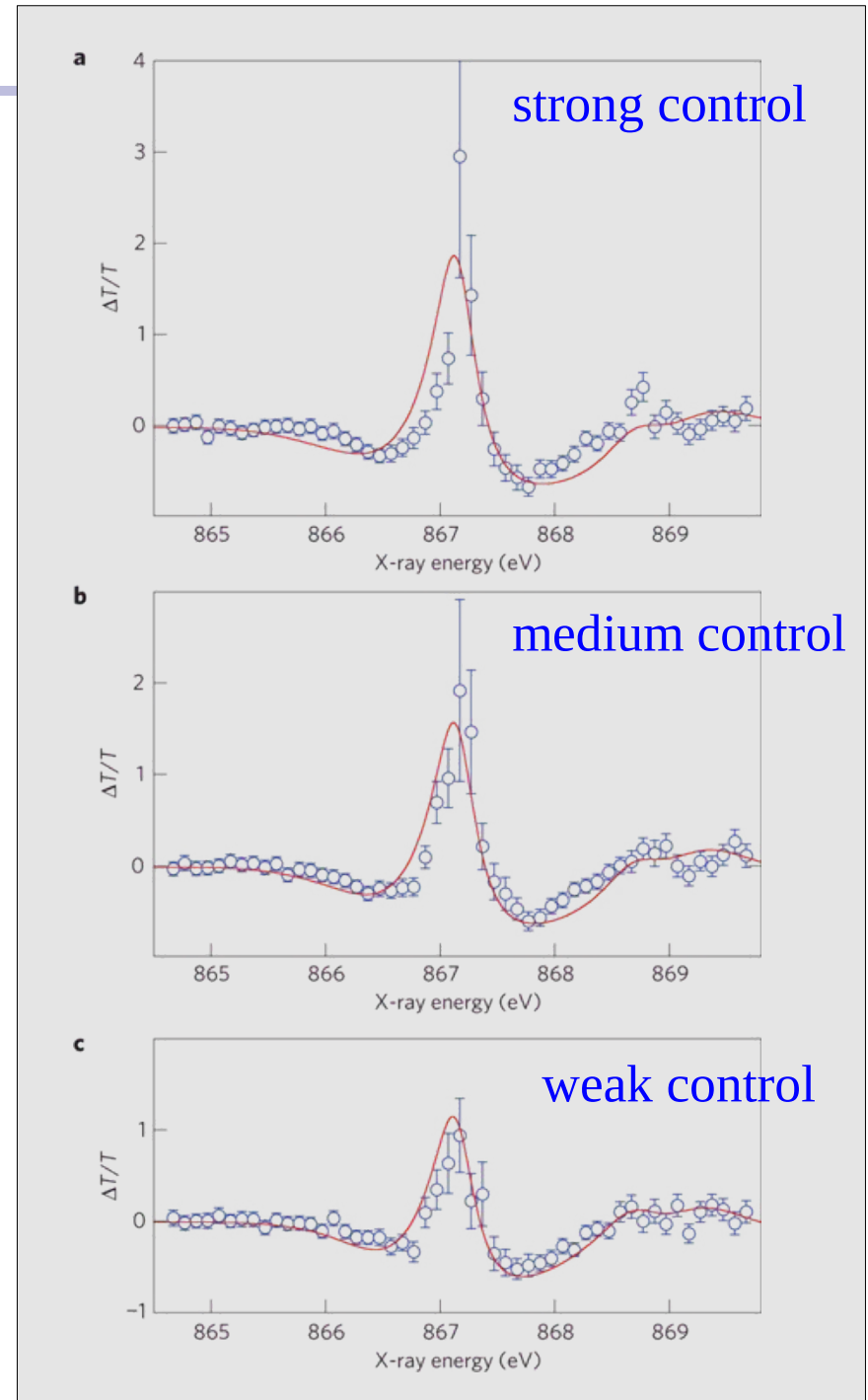
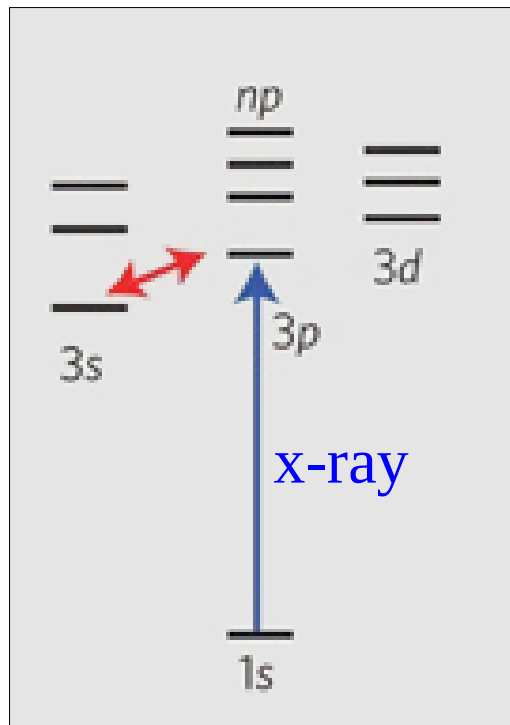
Without control laser

- ▶ Rydberg series with clear absorption on transition 1s-3p can be seen



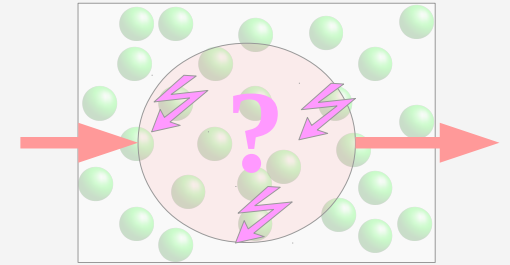
With control laser

- ▶ 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

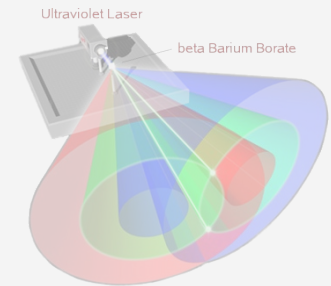


Content

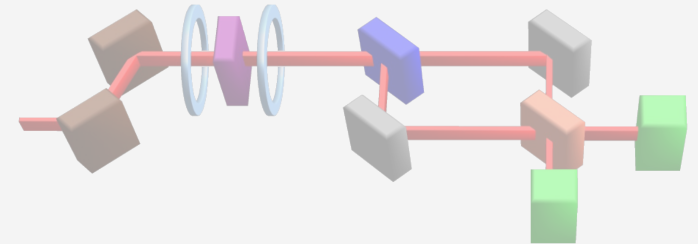
Introduction / NFS



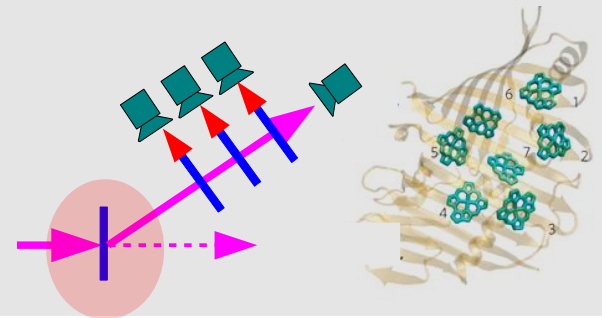
Quantum optics and information



Nuclear quantum optics



Future perspectives



Synchrotron radiation vs. seeded FEL beams

Two directions

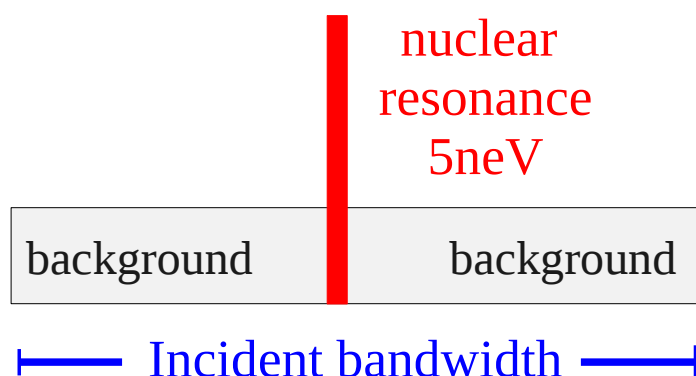
	Synchrotron	Seeded XFEL
Bunch separation (ns)	200ns	200ns (microbunch)
Avg Flux (ph/s/ Γ)	5×10^4	2×10^8
Fluence (ph/bunch/ Γ)	10^{-2}	6×10^3

photon hungry
("proven concepts
with higher
count rate")

Short, nonlinear,
coherent
("new ideas")

nuclear parameters
(for ^{57}Fe)
energy 14.4 keV
linewidth 5 neV

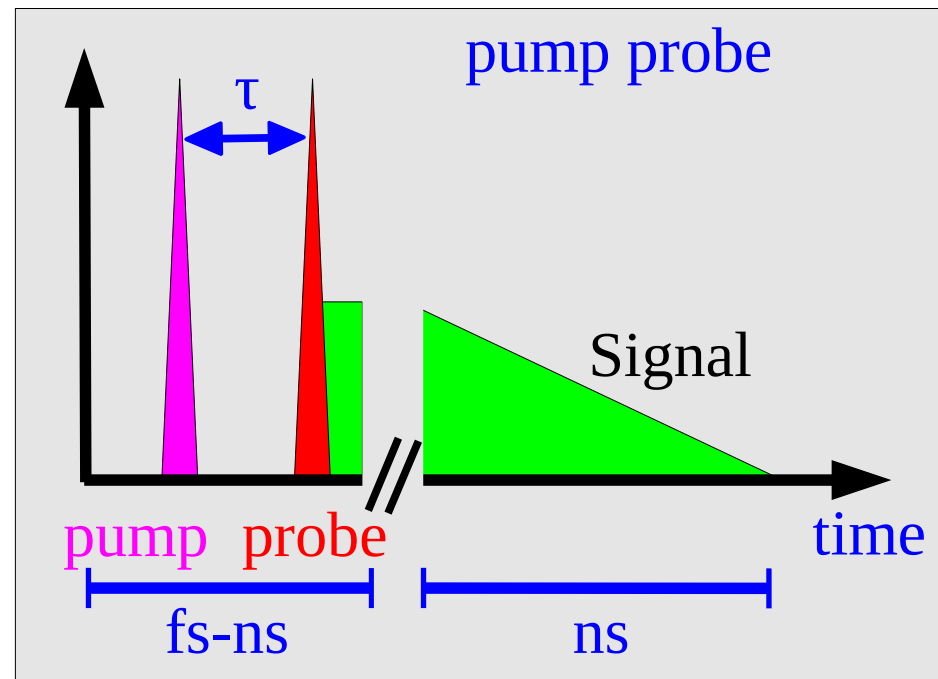
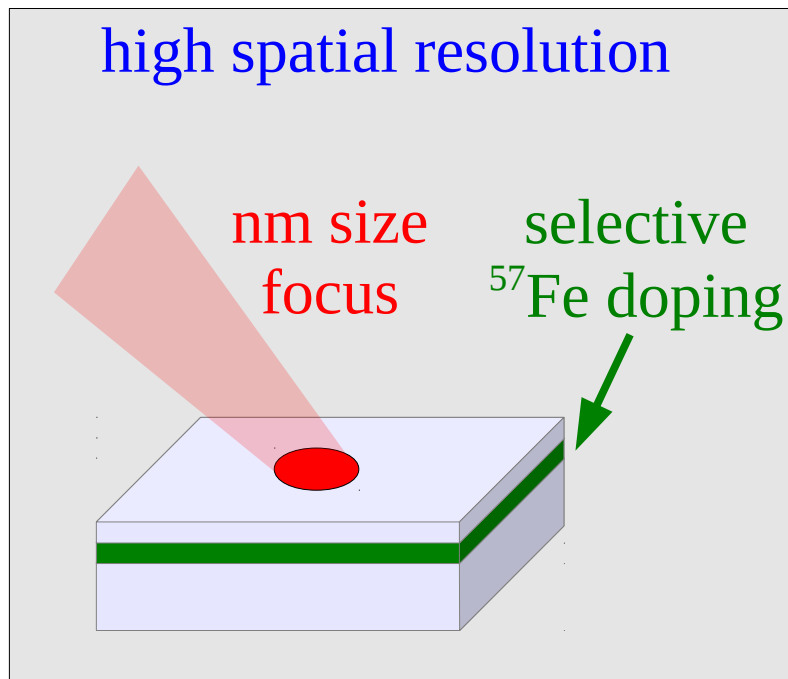
XFEL parameters
 10^{12} photons/pulse
rel. BW 6×10^{-5}
rep. rate 30kHz



Nonequilibrium lattice dynamics

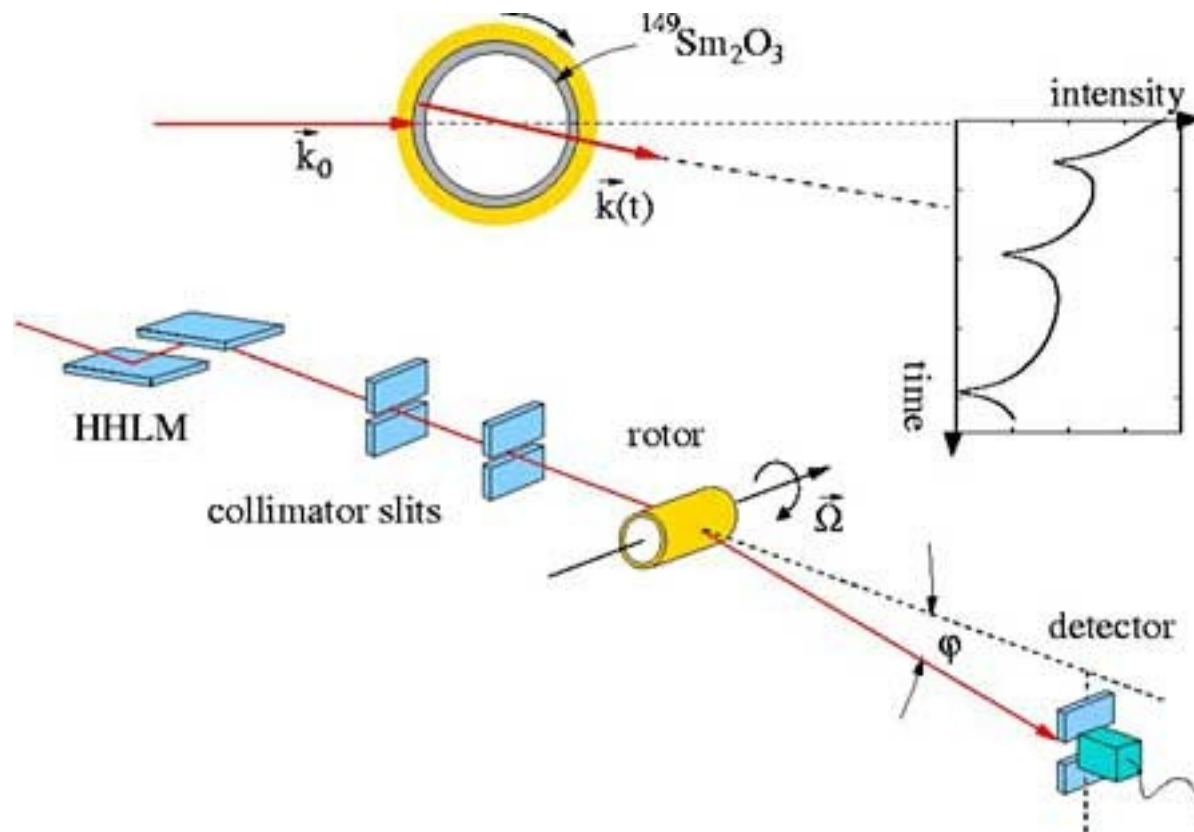
unique
XFEL/NRS
features

- ▶ 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



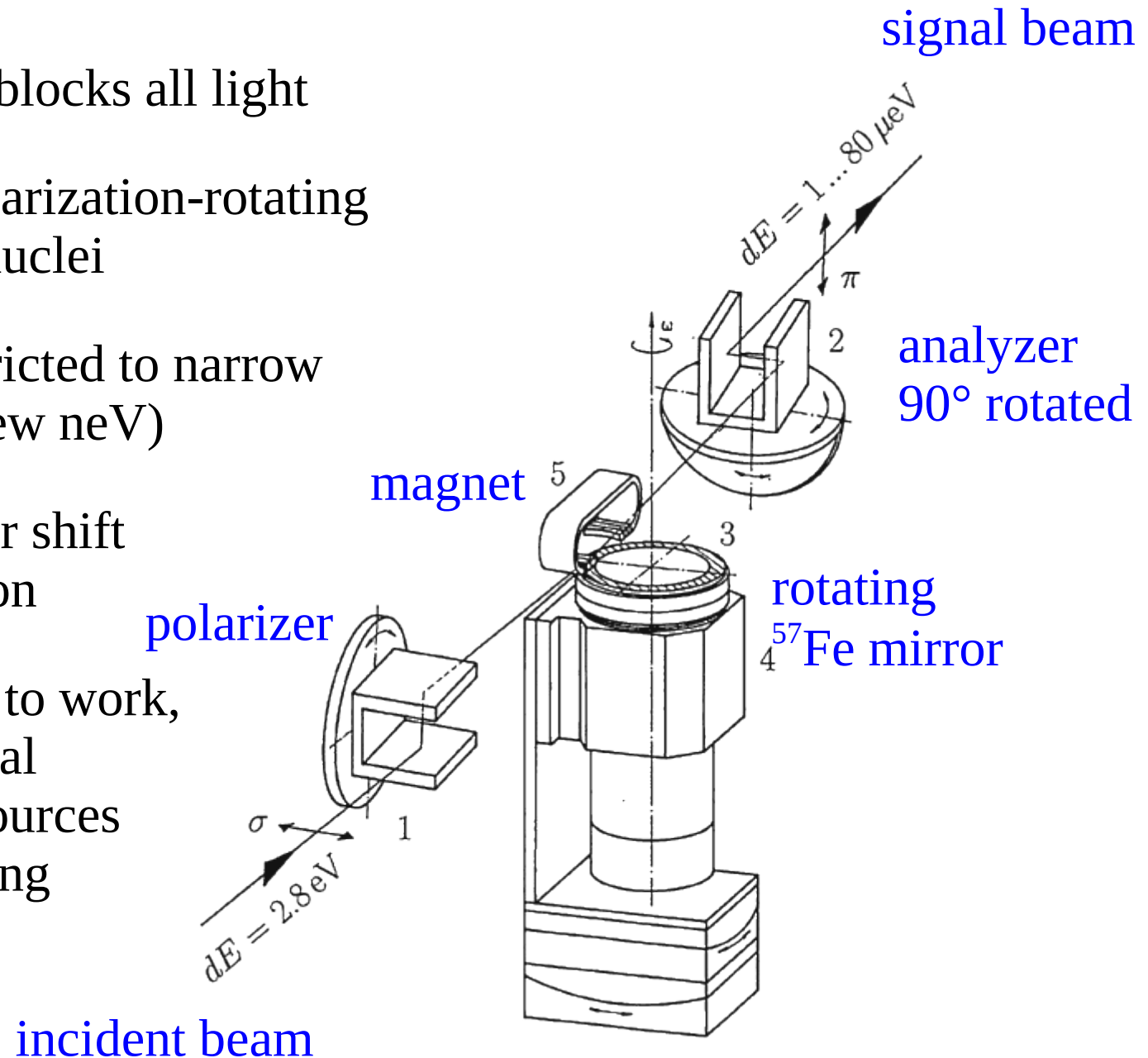
Nuclear lighthouse effect

- ▶ 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



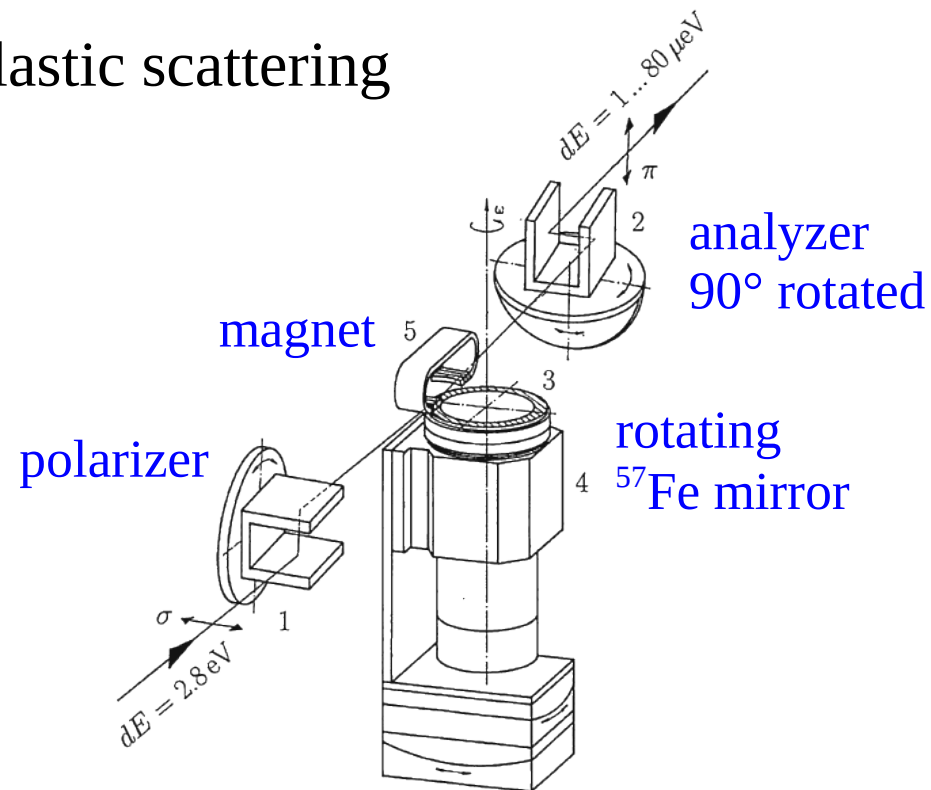
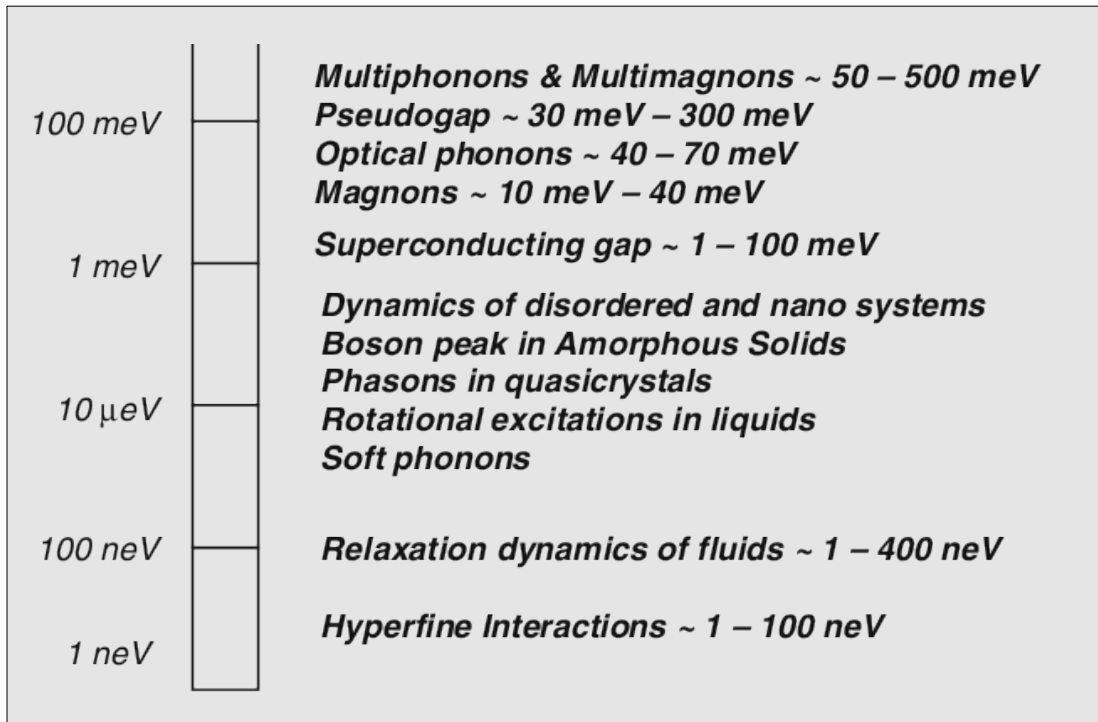
Low-energy condensed matter excitations

- ▶ 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



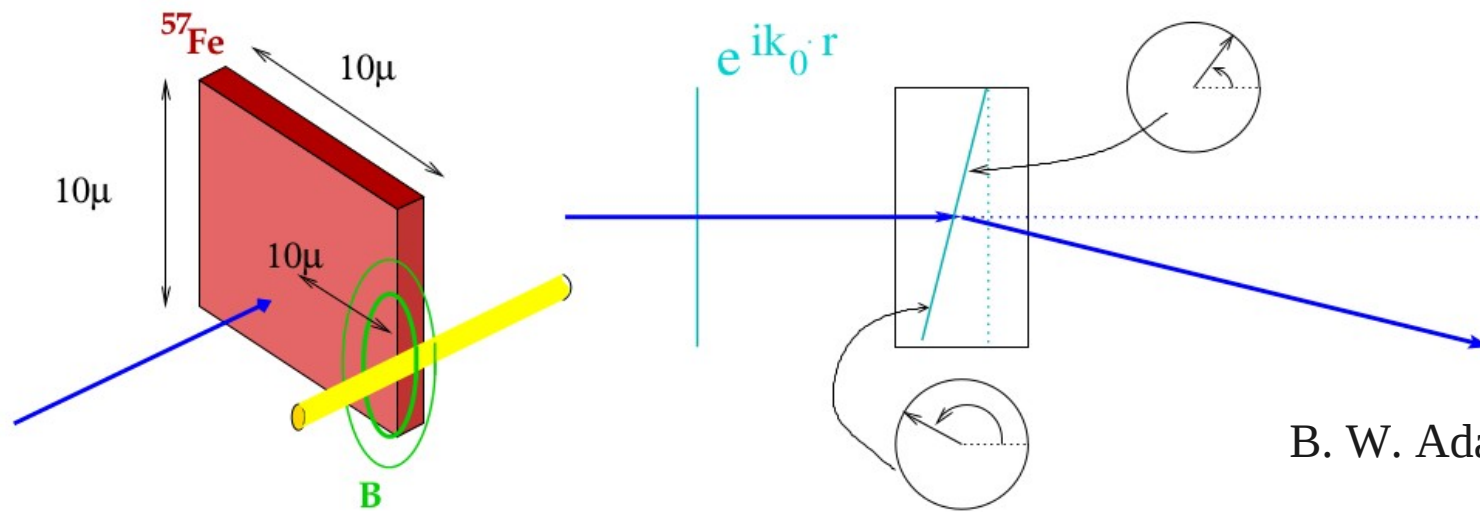
Low-energy condensed matter excitations

- ▶ Spectroscopy with μeV bandwidth tunable over $\sim\text{meV}$ 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



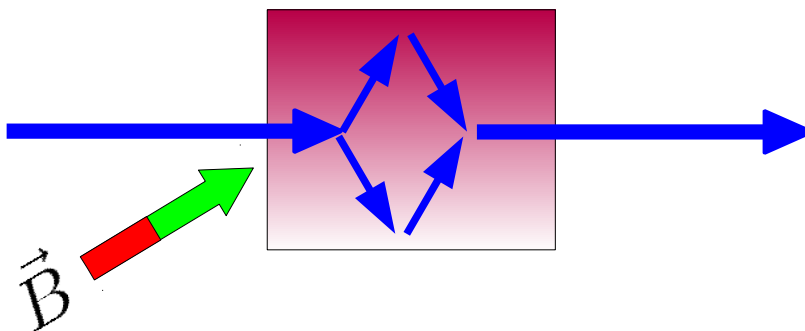
Exciton manipulation without material motion

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



B. W. Adams and others

Possible application: “Virtual” interferometer



Operation possible
without need
to stabilize
material parts???

J. Evers

Can we enter the non-linear regime?

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

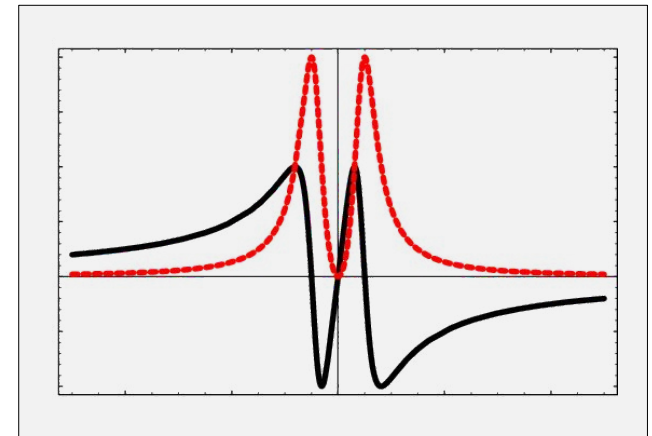
Seeded XFEL: $\frac{10^3 \text{ Photons @ } 14.4\text{keV}}{10\text{fs bunch} \times (\mu\text{m})^2 \times \Gamma} \Rightarrow I \sim 10^{10} \frac{\text{W}}{\text{cm}^2}$

EIT case: Kerr effect

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

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

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



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

What would nonlinear effects be good for?

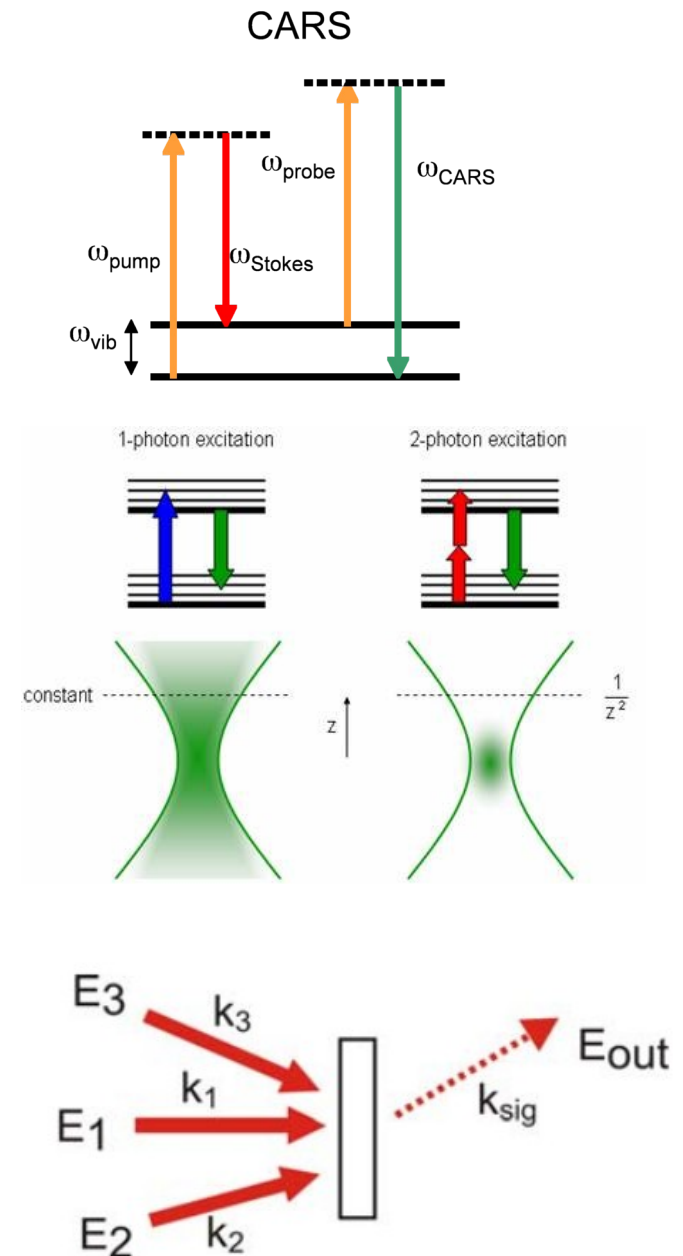
Nonlinear spectroscopy / imaging

- ▶ 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

K. Tamasaku et al, Nature Physics 7, 705 (2011)

- ▶ Downconversion/wave mixing recently observed

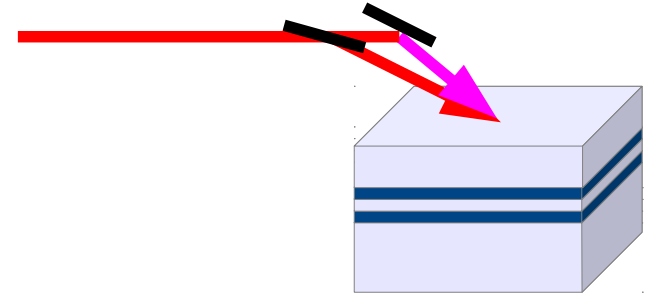
T. E. Glover, Nature 488, 603 (2012)



Immediate applications of multiple photons

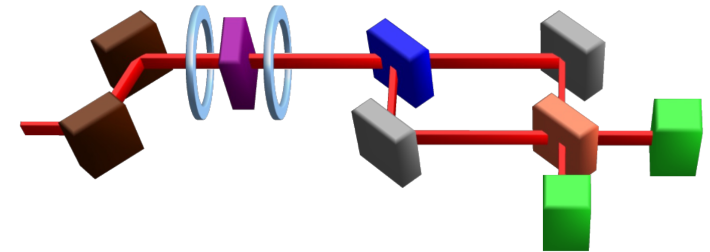
Separate coupling and probe

- ▶ Drive multiple modes simultaneously
- ▶ Beams could be individually and mutually temporally coherent



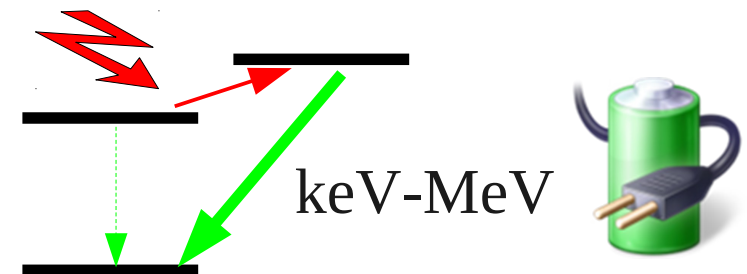
Quantum information and fundamental tests

- ▶ 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



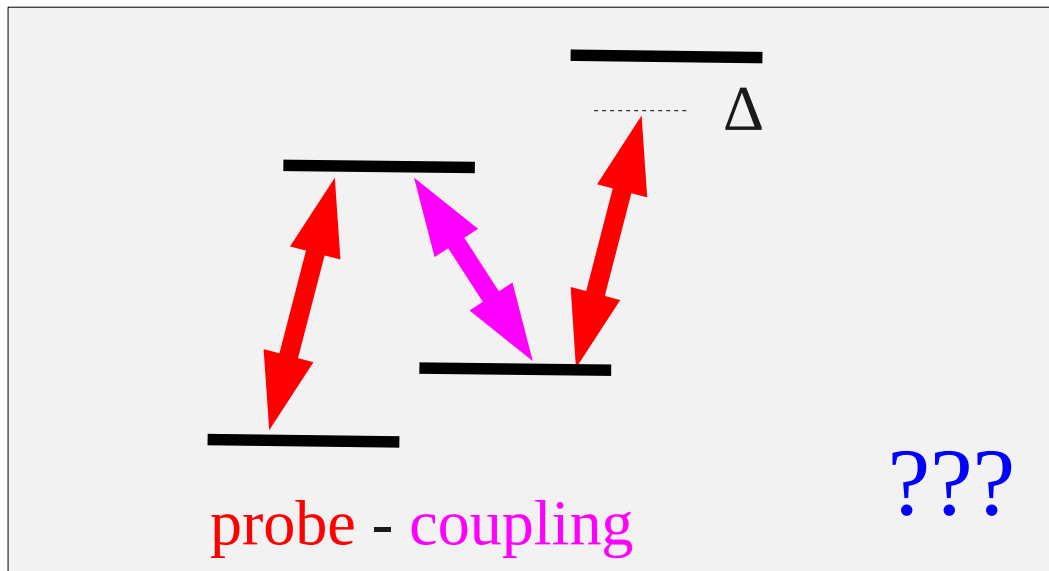
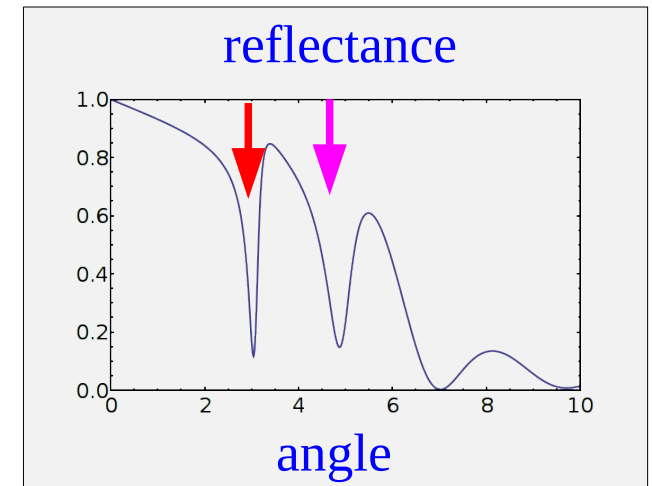
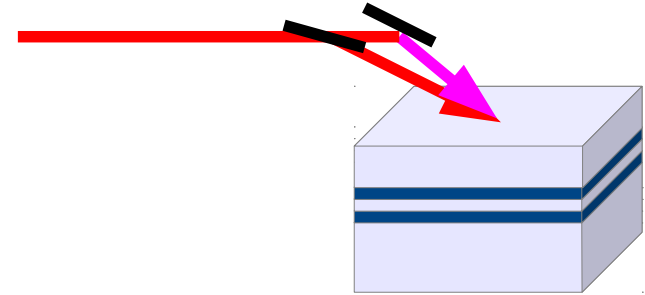
A. Pálffy, J. Evers, C. H. Keitel, *Phys. Rev. Lett.* 99, 172502 (2007)

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

Immediate applications of multiple photons

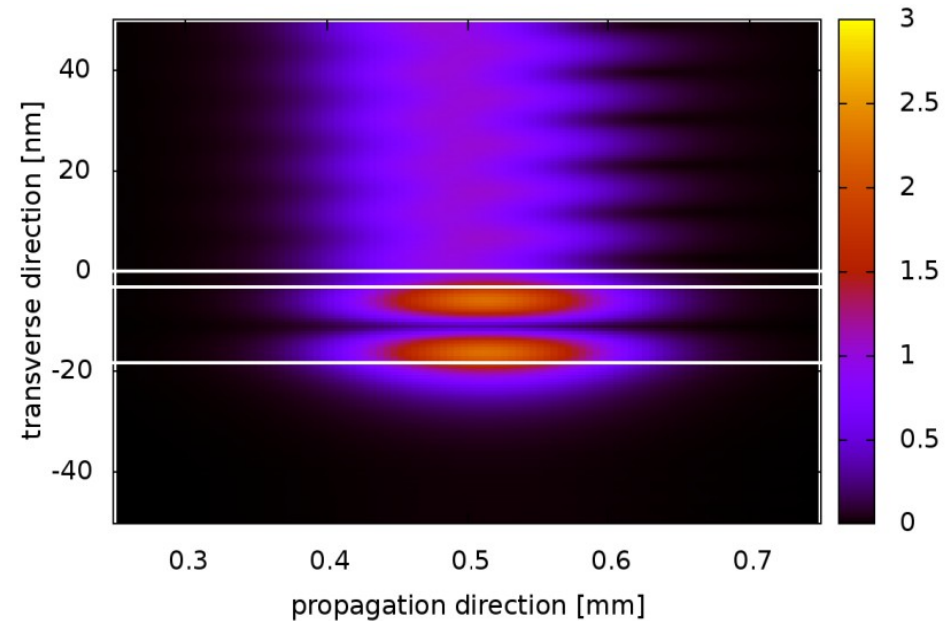
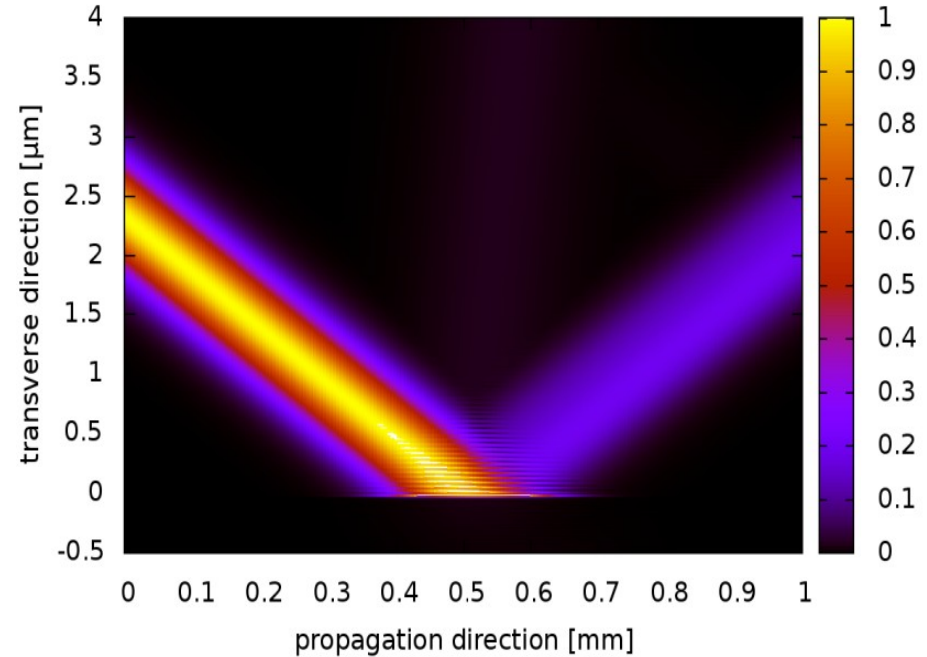
Separate coupling and probe

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



Light propagation in cavities

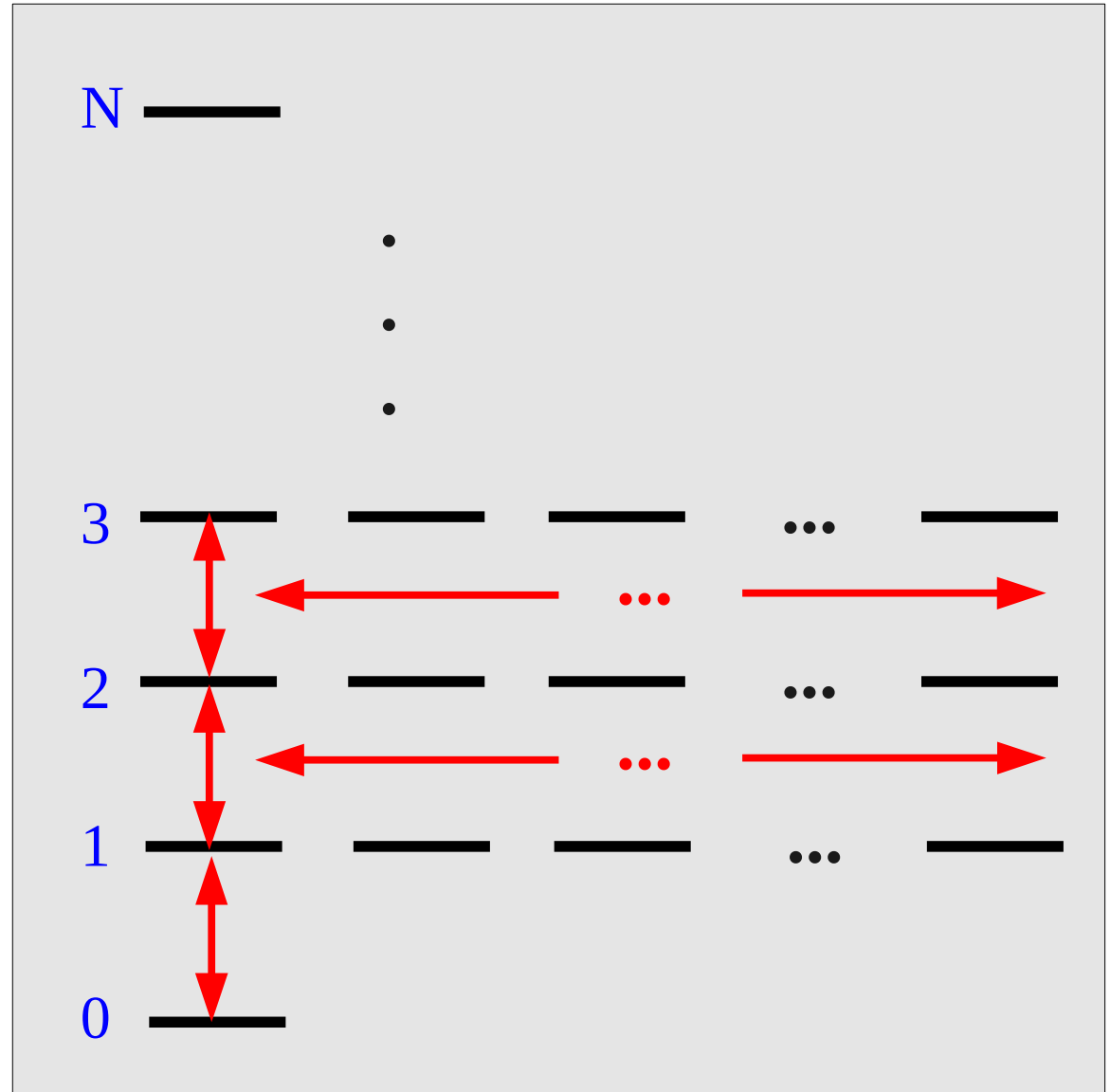
- ▶ Use EIT or SGC to coherently control light propagation in thin film cavities
- ▶ Long propagation times with low losses achievable
- ▶ Applications:
 - nm sized x-ray sources (c.f. T. Salditt, Göttingen)
 - Enhanced light/matter or light/light interaction
 - embed target in cavity
 - deposit cavity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



Climbing up the Dicke ladder

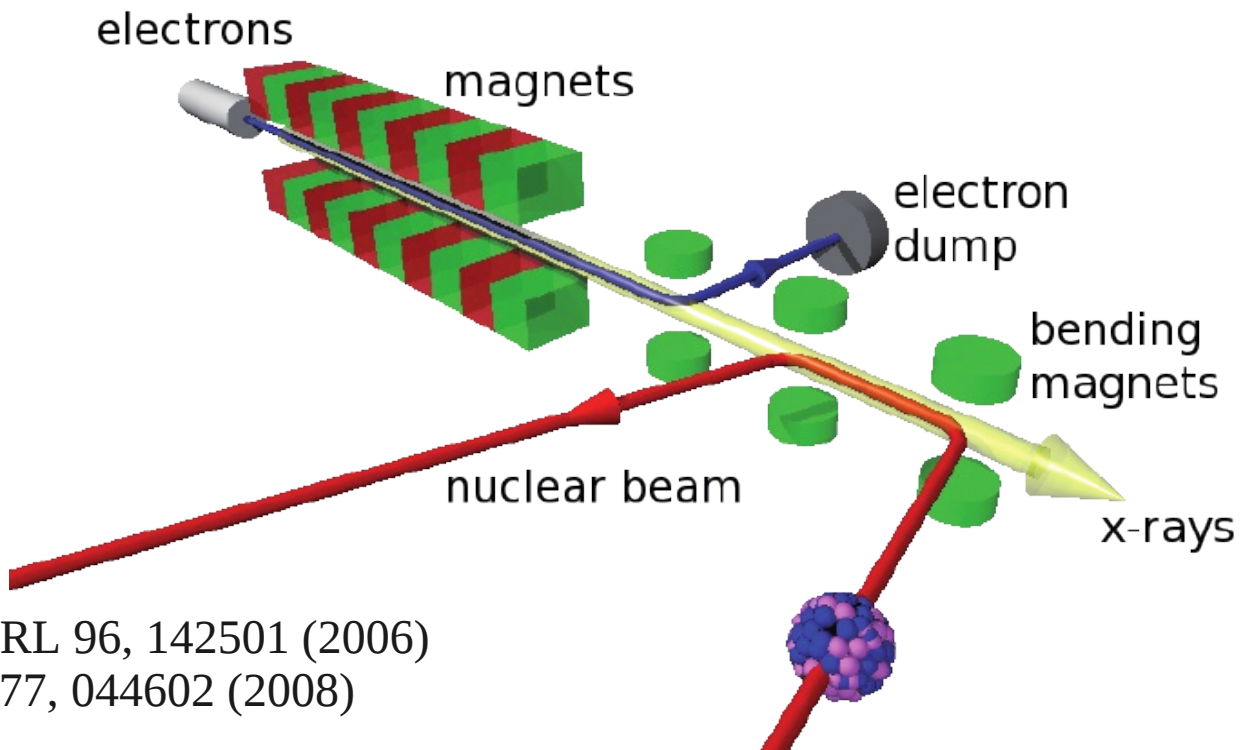
Go beyond single excitation

- ▶ 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?



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



T. Bürvenich, J. Evers, C. H. Keitel, PRL 96, 142501 (2006)

A. Palffy, J. Evers, C. H. Keitel, PRC 77, 044602 (2008)

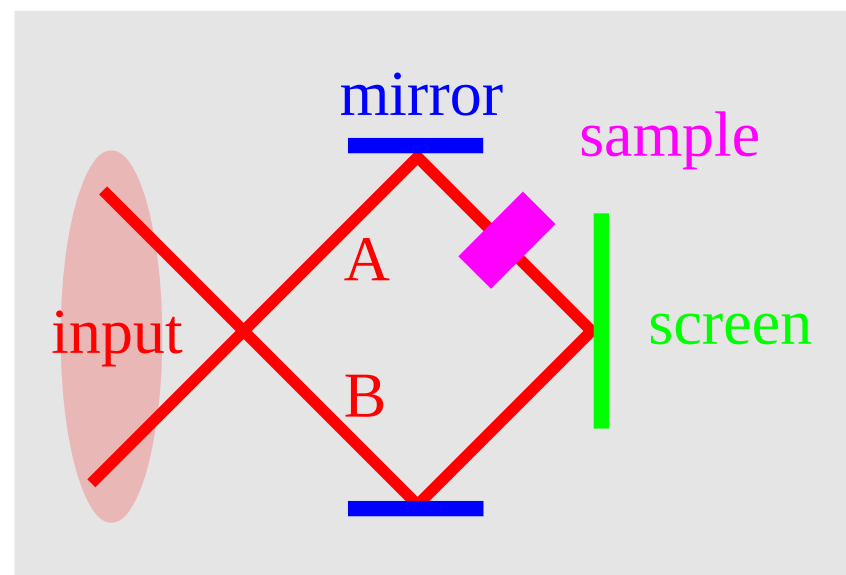
More recent work by A. Palffy et al

Quantum-assisted measurements

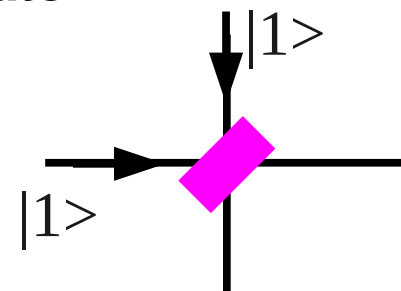
$$|N\rangle_A |0\rangle_B + |0\rangle_A |N\rangle_B$$

sample

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



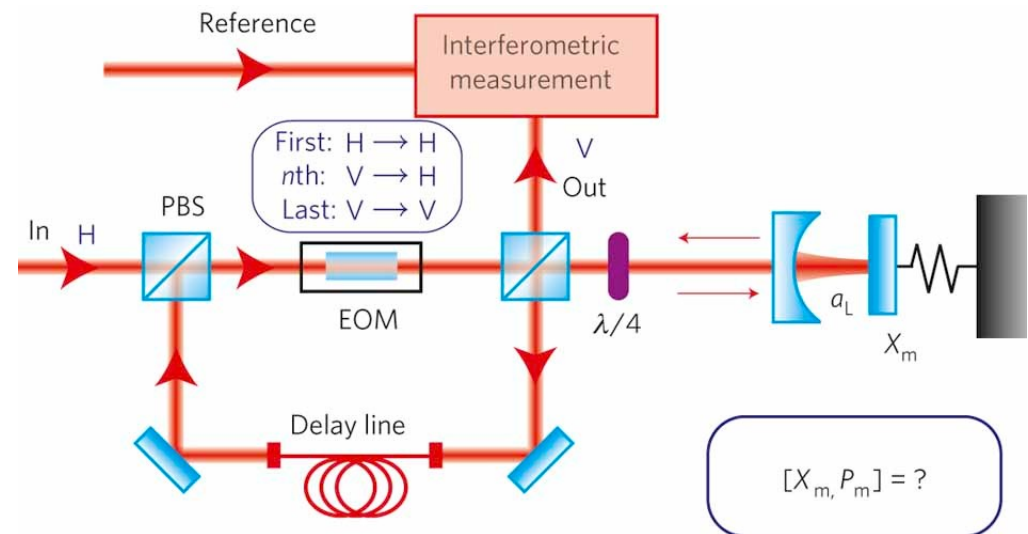
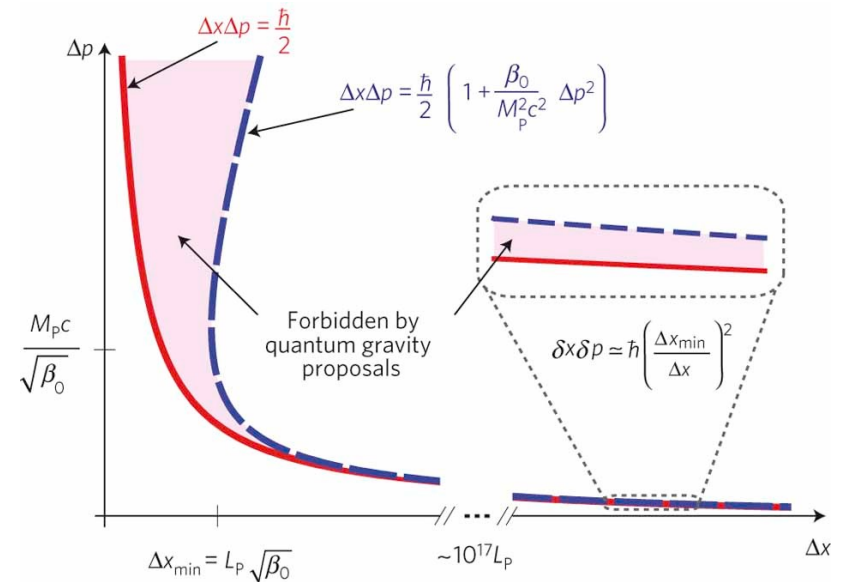
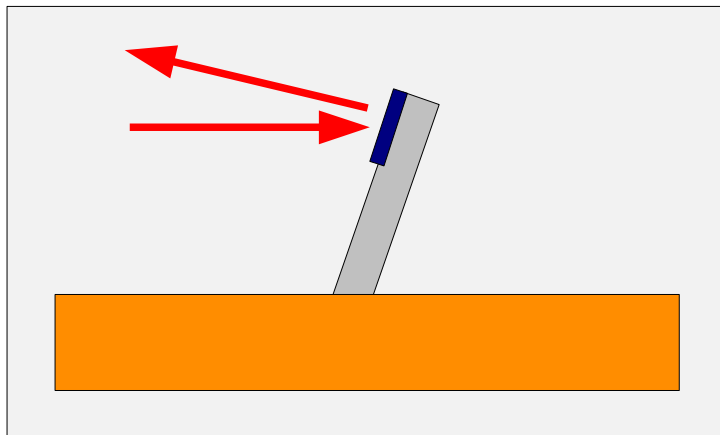
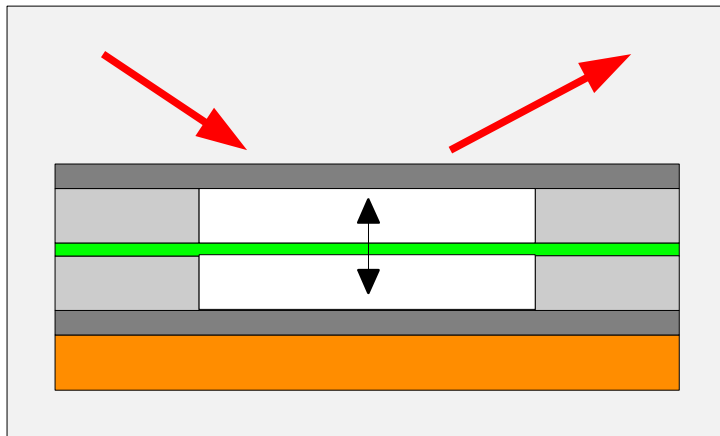
- ▶ The N00N state leads to a phase shift multiplied by N
- ▶ This leads to a N-fold enhancement of the resolution
- ▶ The N00N state is a highly non-classical, entangled state
- ▶ $|2002\rangle$ can be produced by Hong-Ou-Mandel effect



X-ray optomechanics

Fundamental physics with mechanical resonators

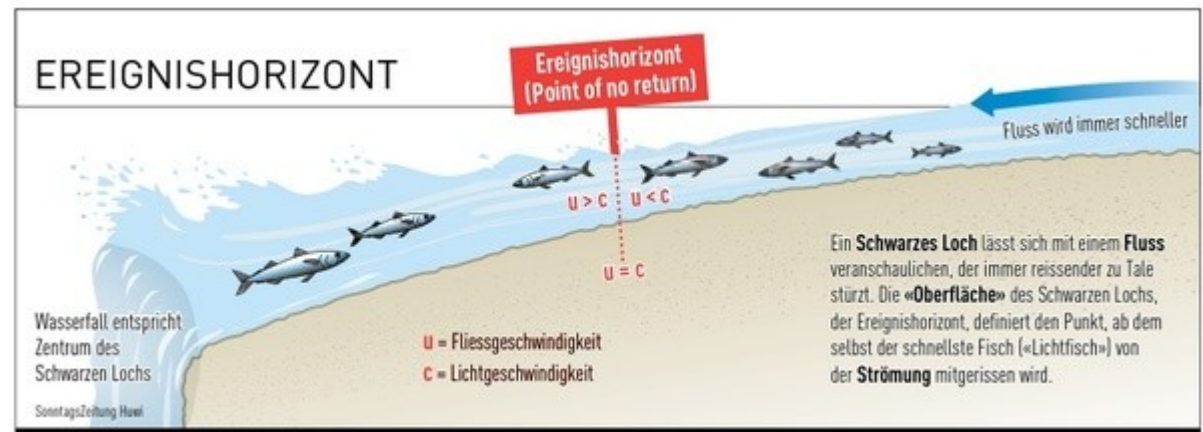
feasible at all?



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

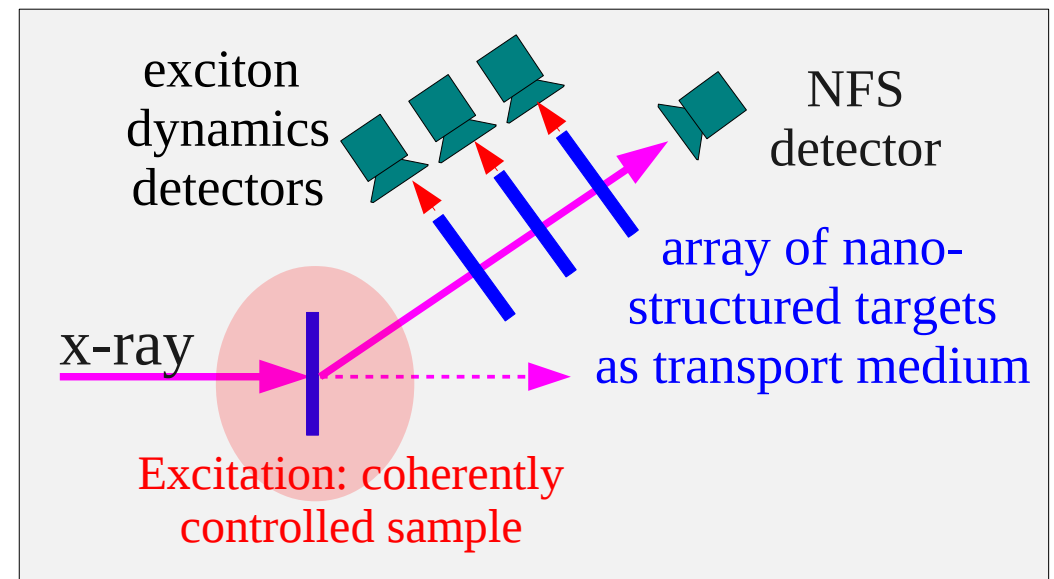
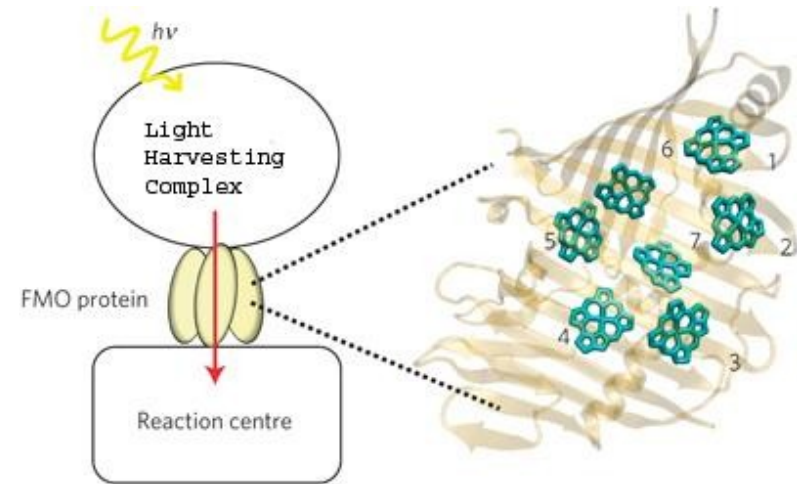


U. Leonhardt and T. G. Philbin, Prog. Opt. 53, 69-152 (2009)
G. Shenoy and R. Röhlsberger, Hyperf. Int. 182, 157 (2008)

Where could this lead?

Quantum transport

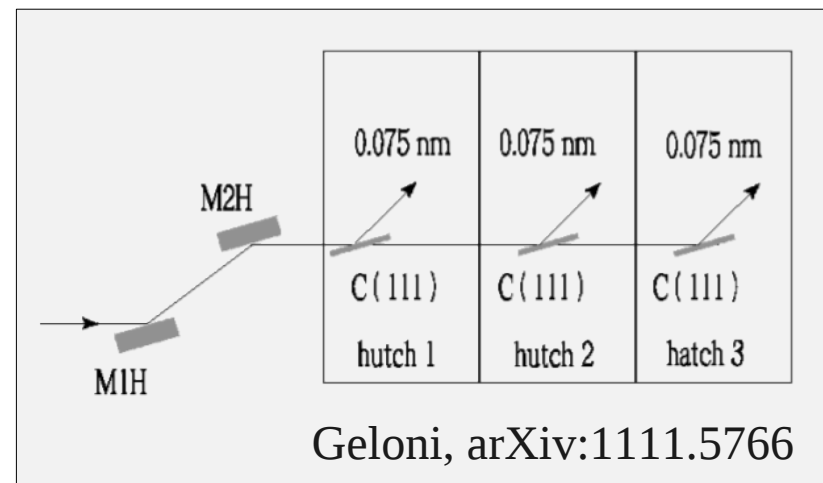
- ▶ Designer quantum channels
- ▶ Start from a clean system, then add decoherence / dephasing at will
- ▶ Model complex bath by perturbing the transport sites independently using laser, E/B field, vibrations, ...
- ▶ Does optimal transport require coherence/ decoherence/ entanglement/...?
- ▶ What are 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”



“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



The team

Martin Gärttner	PhD student
Qurrat-ul-Ain Gulfam	PhD student
Kilian Heeg	PhD student
Paolo Longo	PostDoc
Andreas Reichegger	Master student
David Schönleber	Master student
Lida Zhang	PhD student

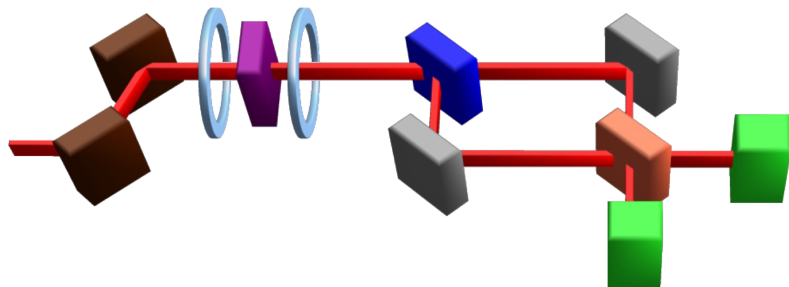
Collaboration (DESY)

Ralf Röhlsberger
Hans Christian Wille
Kai Schlage

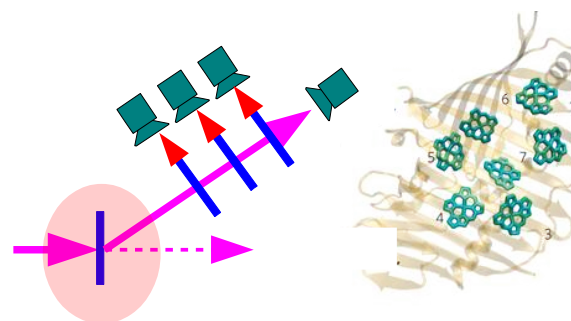
Funding: MPG, DFG, DAAD,
IMPRS-QD, CQD



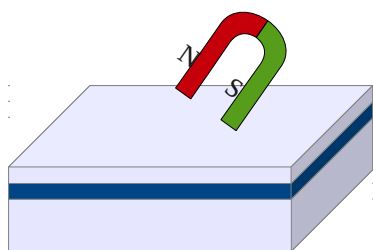
Summary



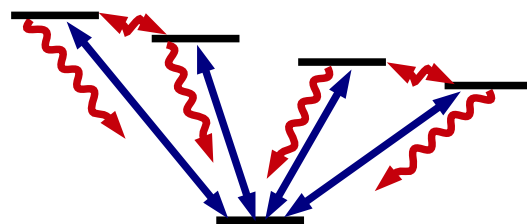
X-ray entanglement



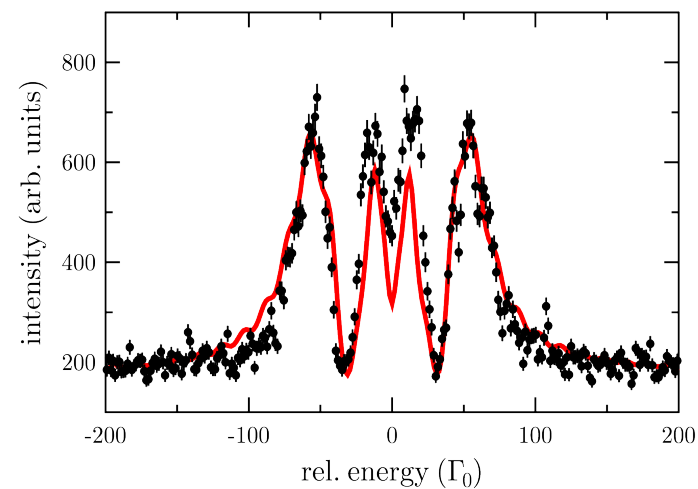
Quantum transport



\cong



Tunable nuclear level schemes



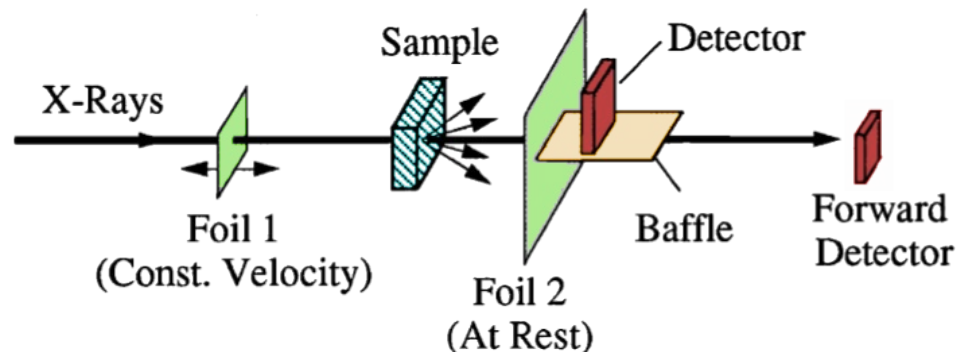
spontaneously generated
coherences

Thank you!

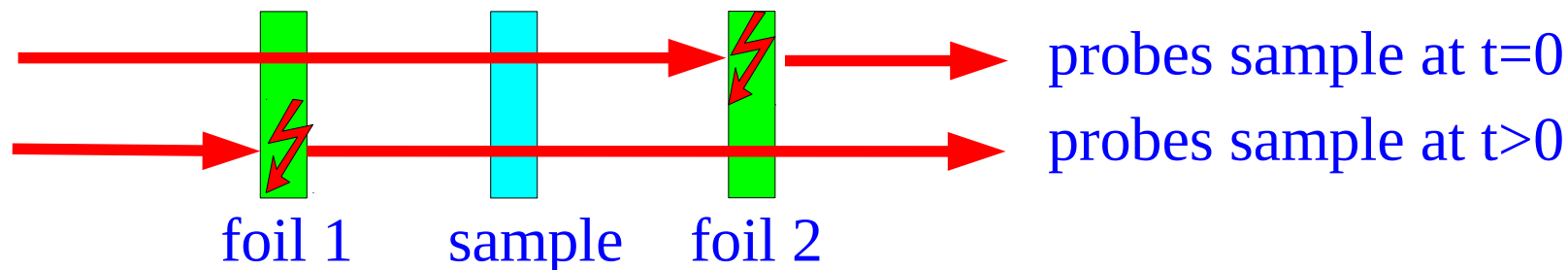


Scattering function in the time domain

Setup with NRS in two ^{57}Fe foils



Two interfering scattering pathways



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

beat pattern
(foil velocity)

electron density in sample

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**

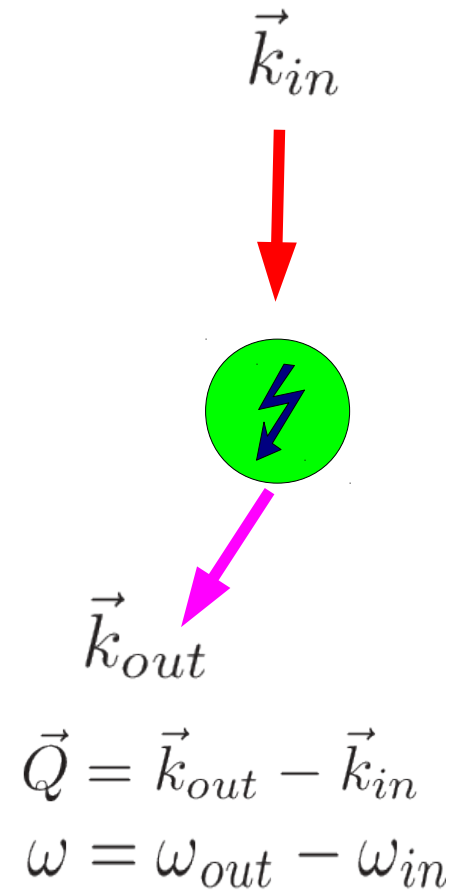
transition rate $R \sim \left| S(\vec{Q}, \omega) \right|^2$

- ▶ 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:

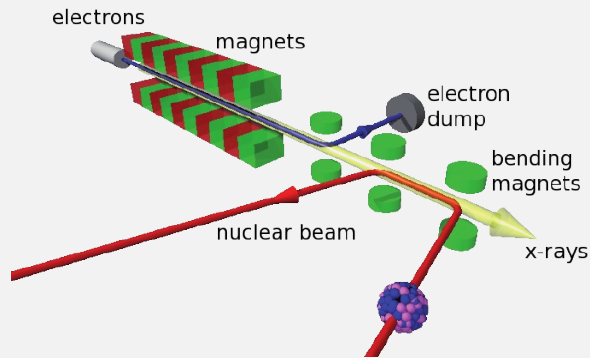
$$S(\vec{Q}, t) = \int S(\vec{Q}, \omega) e^{i\omega t} d\omega$$

- ▶ Need high Q and t range, large signal/noise ratio
- ▶ Example application: correlated electron materials



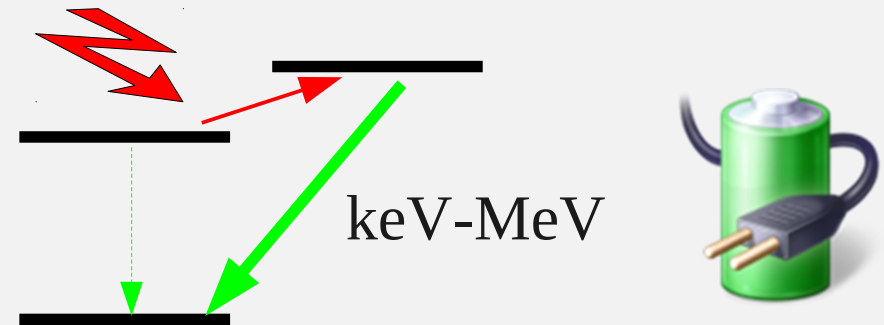
X-ray and γ -ray quantum optics @ MPIK

Direct laser driving of nuclei



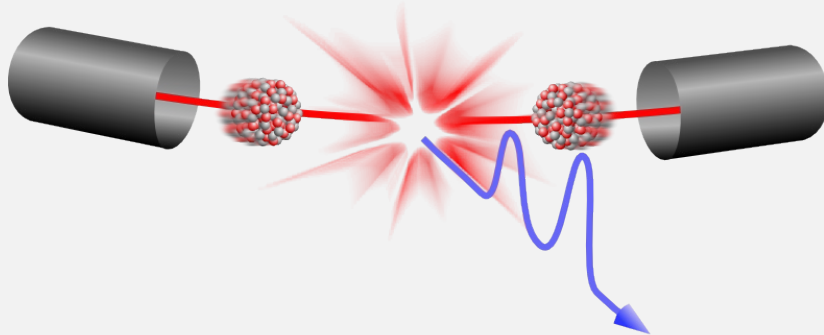
T. Bürvenich, J. Evers, C. H. Keitel,
PRL 96, 142501 (2006)

Isomer triggering



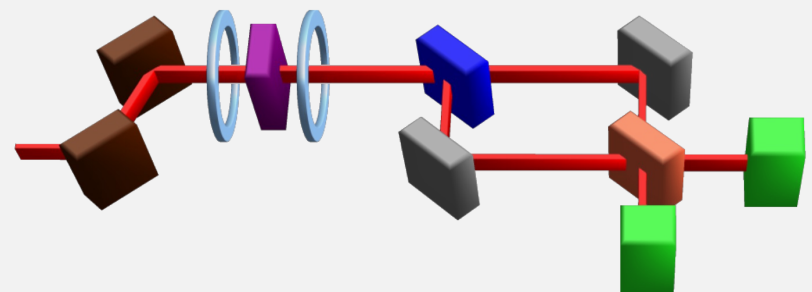
A. Pálffy, J. Evers, C. H. Keitel,
PRL 99, 172502 (2007)

Yoctosecond physics



A. Ipp, C. H. Keitel, J. Evers,
PRL 103, 152301 (2009)

X-ray cooperative light scattering



A. Pálffy, C. H. Keitel, J. Evers, PRL 103,
017401 (2009); PRB 83, 155103 (2011)

Possible proof-of-principle experiment

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

▶ With phase shift by Alice:

$$N_A = \sin^2(\phi_A/2) N \text{ photons go to D (} G_A \text{)}$$

▶ With phase shift by Bob:

$$N_B = \sin^2(\phi_B/2) N \text{ photons go to D (} G_B \text{)}$$

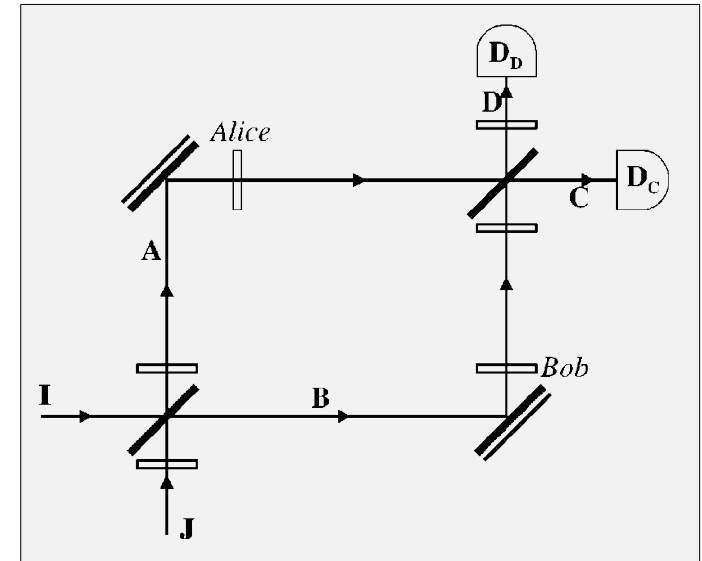
▶ With both phase shifts:

$$N_{AB} = \sin^2[(\phi_A - \phi_B)/2] N \text{ go to D (} G_{AB} \text{)}$$

▶ 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

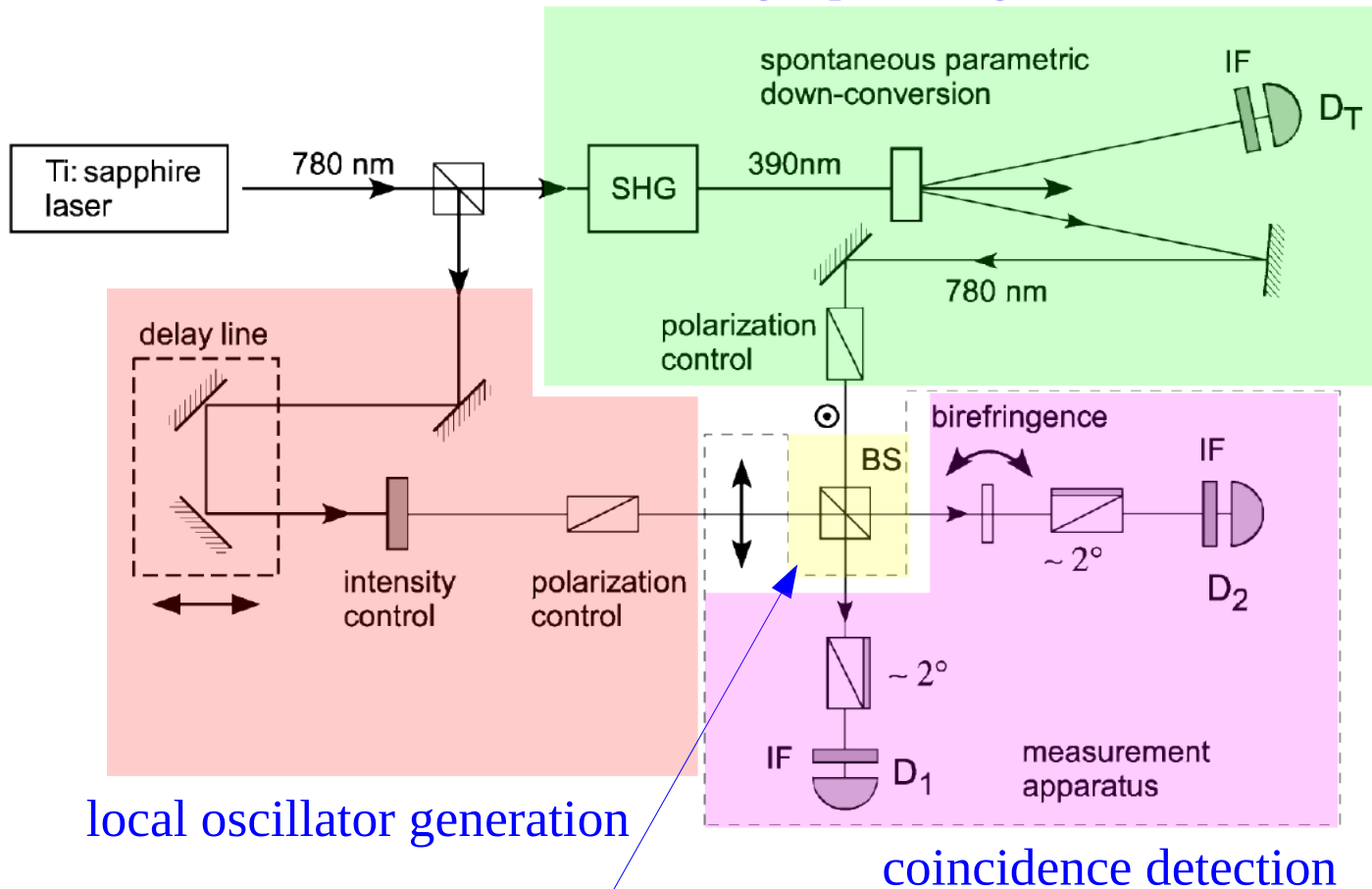
$$\text{(Alice and Bob shift)} \quad (G_N - G_A) \cap (G_N - G_B) \subseteq (G_N - G_{AB})$$



$$N_{AB} \leq N_A + N_B \text{ violated for some phase shifts}$$

Experimental evidence with local oscillator

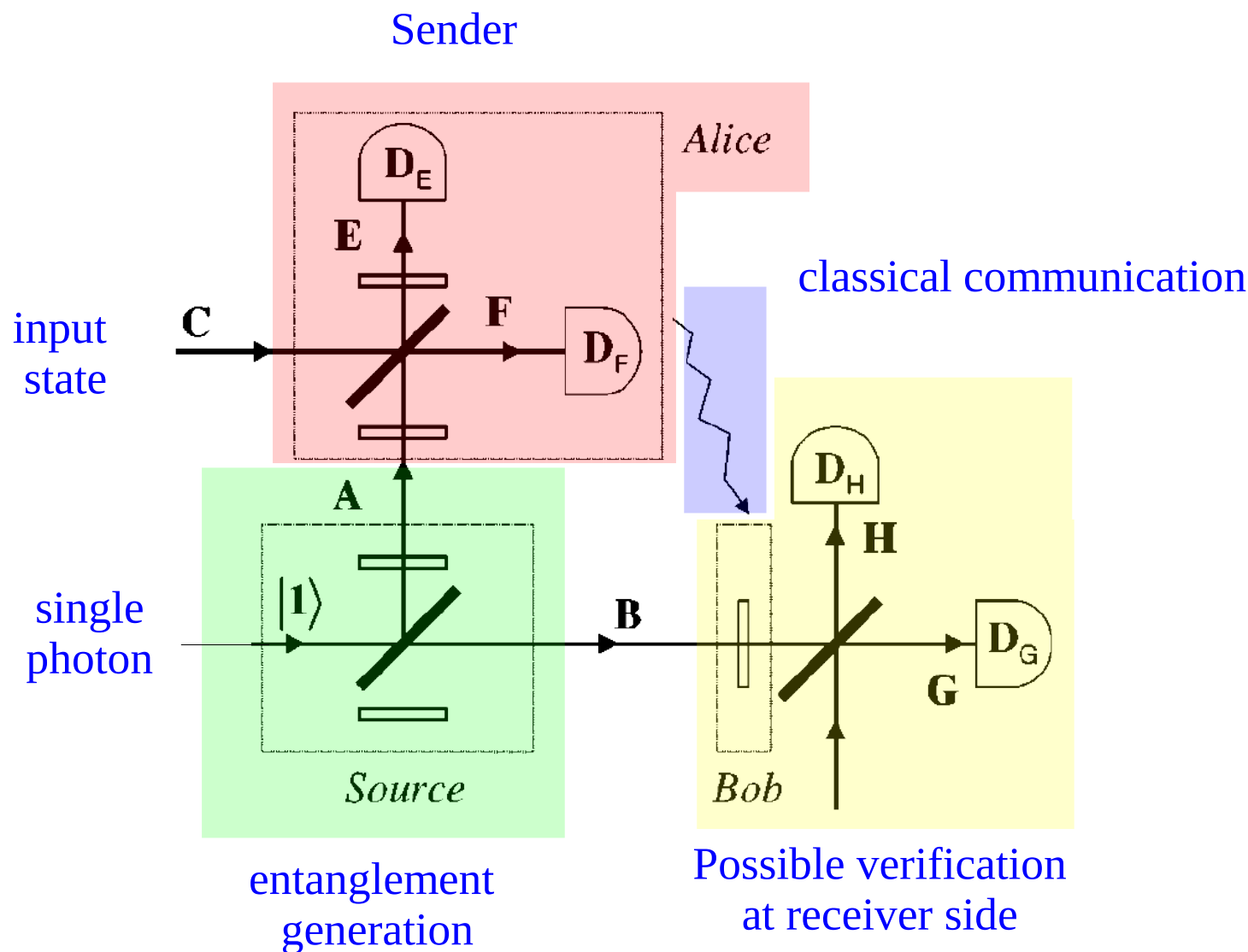
single photon generation



entanglement generation,
mixing with LO

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

Single photon entanglement teleportation scheme

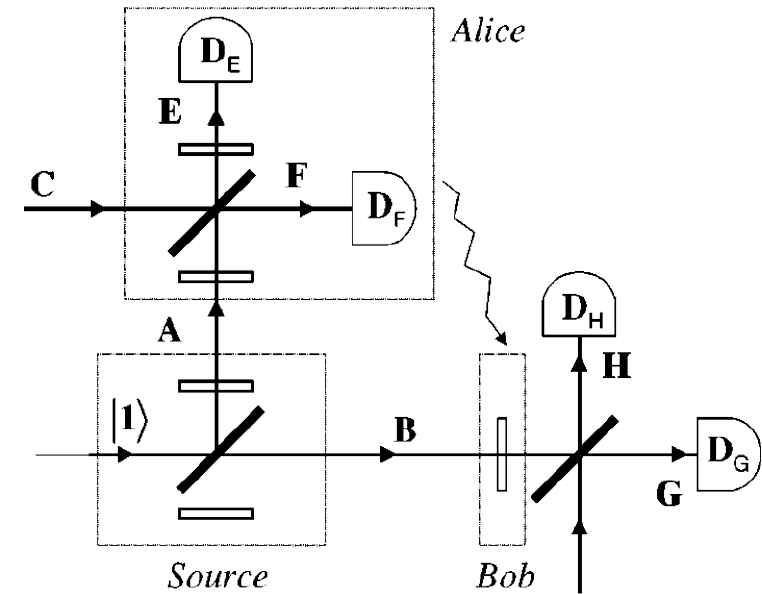


Teleportation algebra

$$\begin{aligned}
 |\Psi\rangle &= \frac{1}{\sqrt{2}} \left(|1\rangle_A |0\rangle_B + |0\rangle_A |1\rangle_B \right) (a|1\rangle_C + b|0\rangle_C) \\
 &= \frac{1}{2} |0\rangle_E |1\rangle_F (a|1\rangle_B + b|0\rangle_B) \\
 &+ \frac{1}{2} |1\rangle_E |0\rangle_F (a|1\rangle_B - b|0\rangle_B) \\
 &+ \frac{1}{2} \left(\frac{1}{2} |0\rangle_E |2\rangle_F - \frac{1}{2} |2\rangle_E |0\rangle_F + \frac{1}{\sqrt{2}} |0\rangle_E |0\rangle_F \right) (a|0\rangle_B + b|1\rangle_B) \\
 &+ \frac{1}{2} \left(\frac{1}{2} |0\rangle_E |2\rangle_F - \frac{1}{2} |2\rangle_E |0\rangle_F - \frac{1}{\sqrt{2}} |0\rangle_E |0\rangle_F \right) (a|0\rangle_B - b|1\rangle_B)
 \end{aligned}$$

entanglement
input

measurement Alice
teleported state

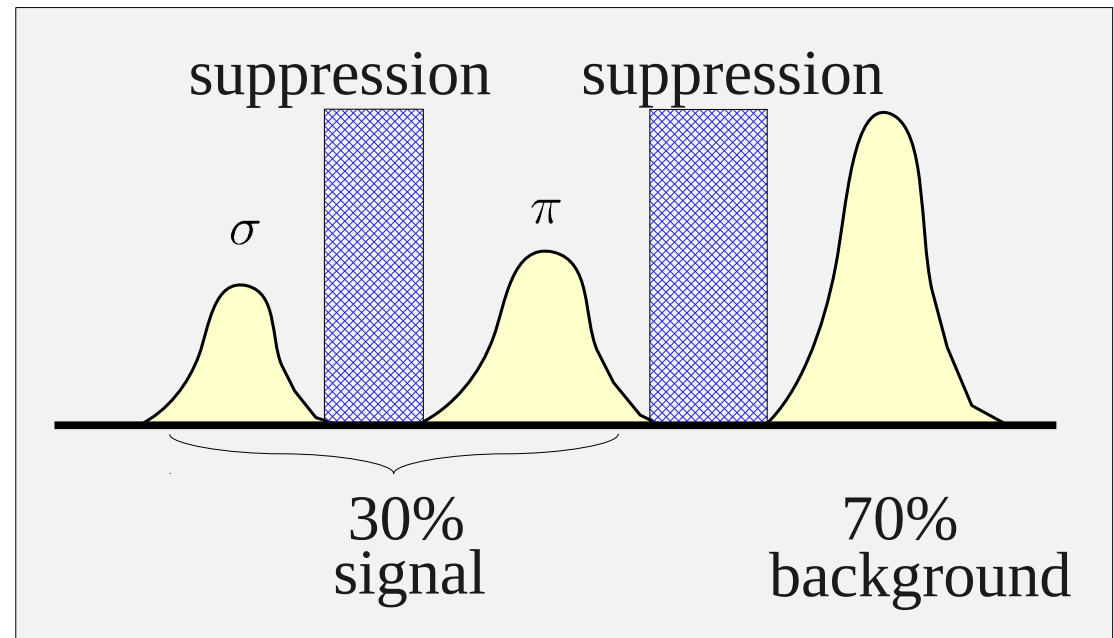


Efficiency estimate

- ▶ 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 (2nd order)

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

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
transitions

de-excitation

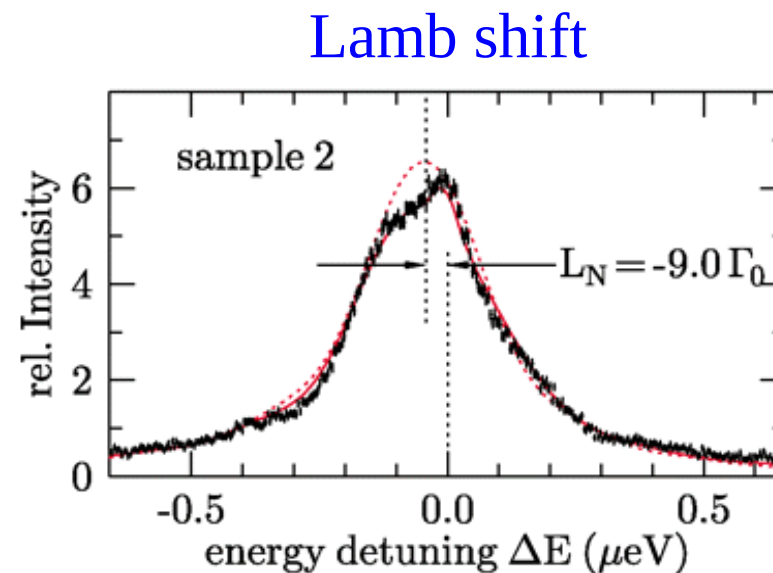
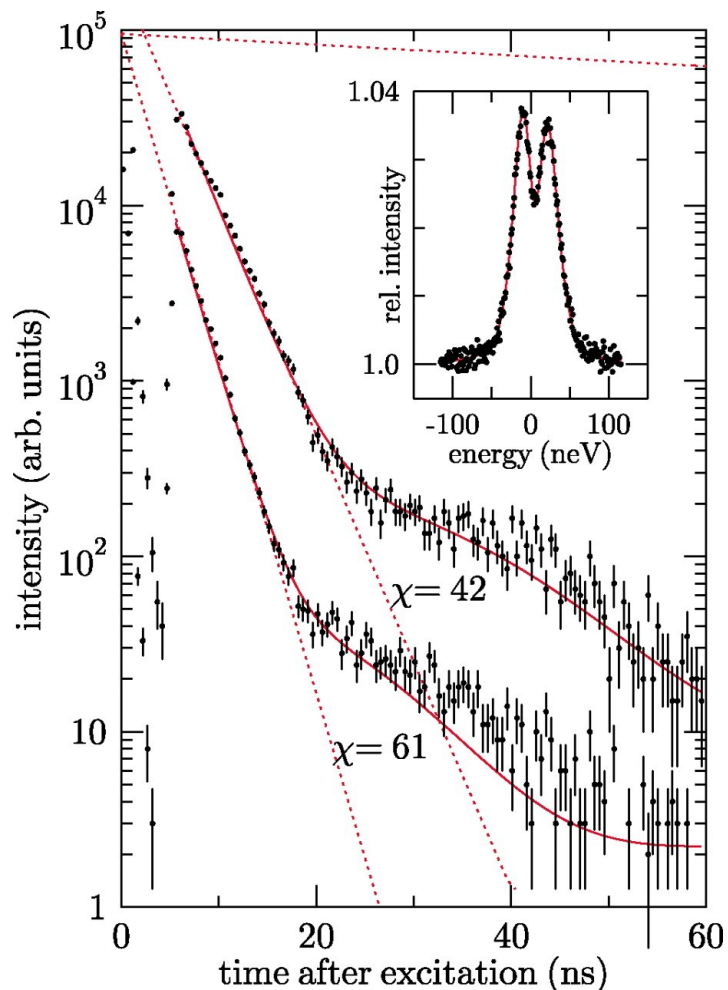
excitation

Iterative solution,
incident pulse

$$\mathcal{E}^{(0)} \sim \delta(t)$$

Recent experiment: Collective Lamb Shift

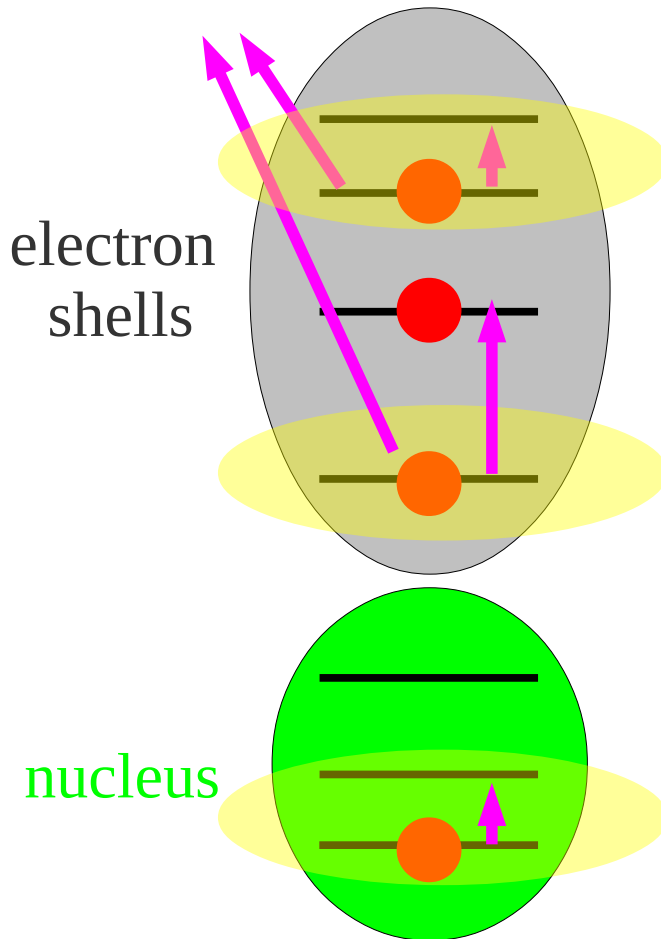
- ▶ Lamb shift due to virtual photon exchange in ensembles of atoms
- ▶ Experimentally observed with nuclei using forward scattering
- ▶ Experimental challenge: Prepare purely superradiant state in thick sample; solution: embed nuclei in low-q cavity



Motivation

Light-matter interactions

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

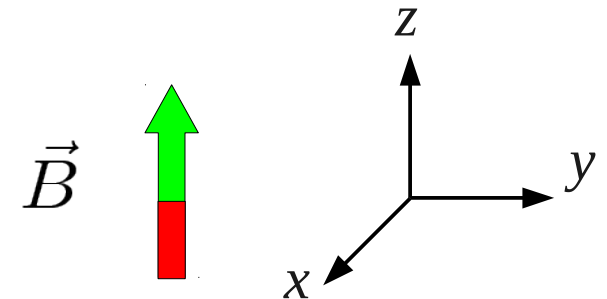
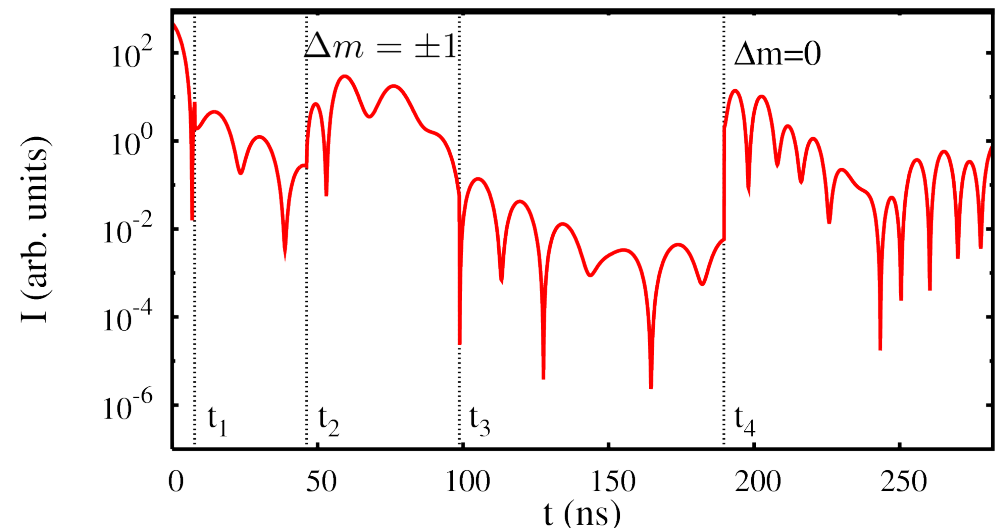


These scenarios appear similar

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 $\Delta m = 0$



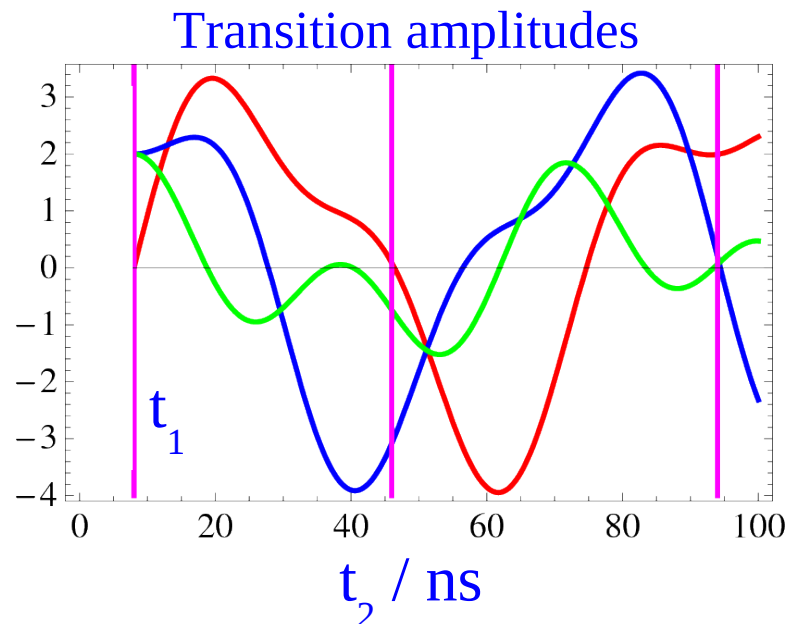
Advanced magnetic switching schemes

Rotation angle

- Determines new quantization axis and superposition states

Timing

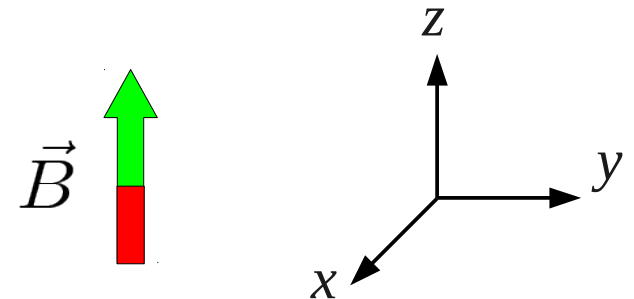
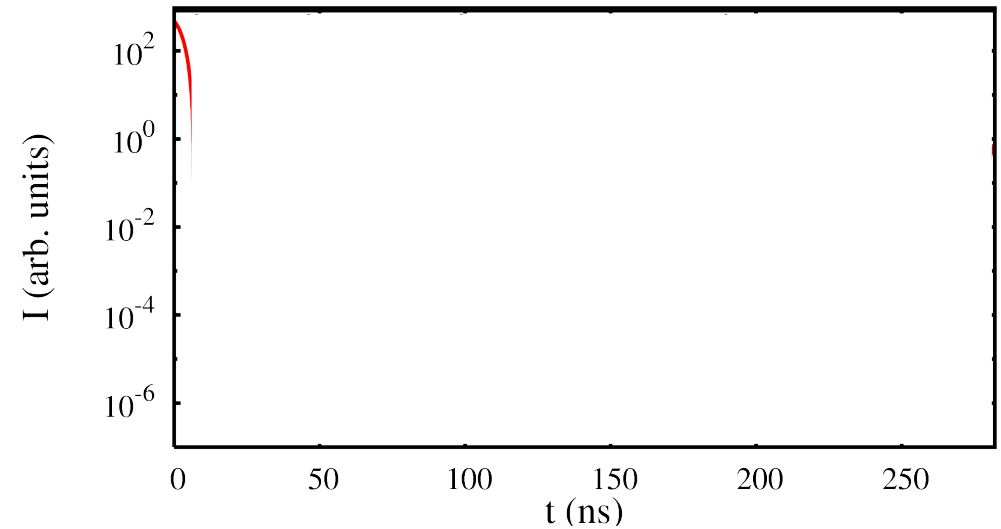
- Important due to different transition energies
- Determine whether constructive/destructive interference occurs
- Example: Suppression at t_1 , how does t_2 affect further evolution?



$$\left\{ \frac{1}{2} \rightarrow \frac{1}{2}, -\frac{1}{2} \rightarrow -\frac{1}{2} \right\} \quad \text{linear}$$
$$\left\{ -\frac{1}{2} \rightarrow \frac{1}{2}, \frac{1}{2} \rightarrow -\frac{1}{2} \right\} \quad \text{circular}$$
$$\left\{ \frac{3}{2} \rightarrow \frac{1}{2}, -\frac{3}{2} \rightarrow -\frac{1}{2} \right\} \quad \text{circular}$$

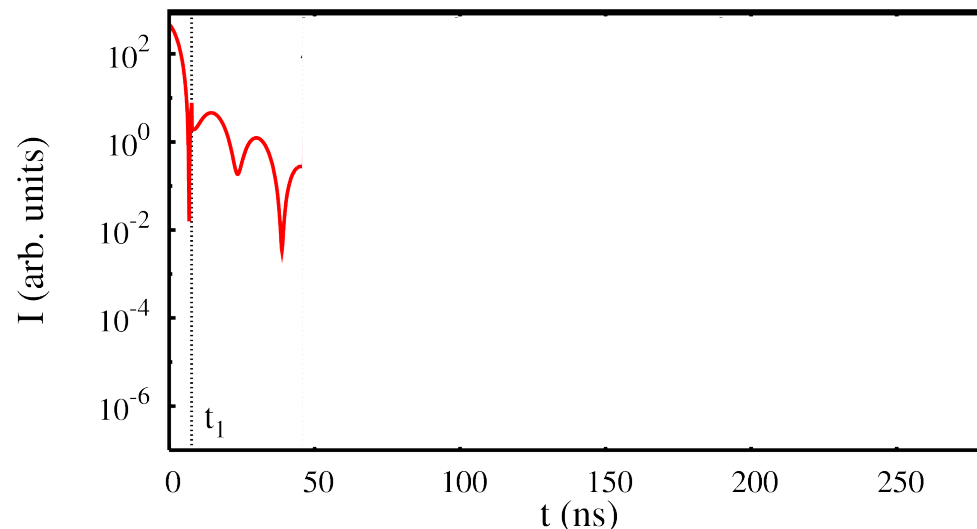
Step 1: Synchrotron excitation

- ▶ Initially, magnetic field is in z direction

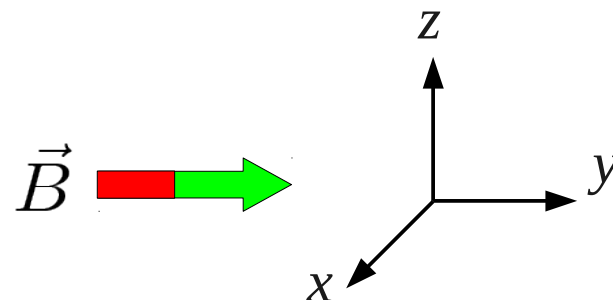
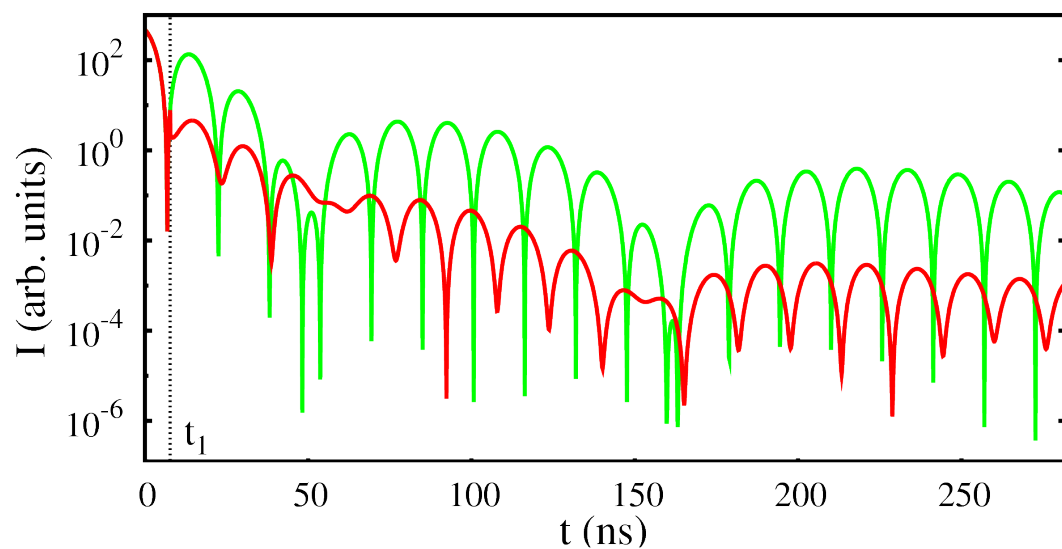


Step 2: Canceling coherent decay

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

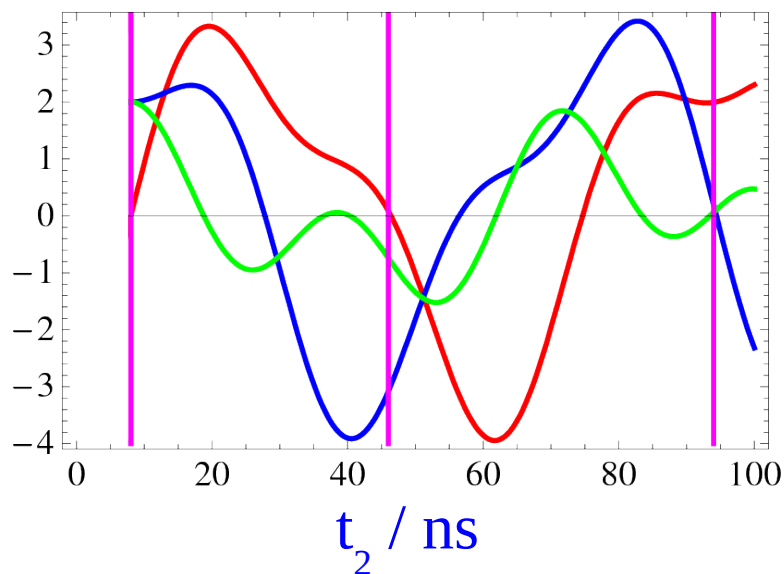
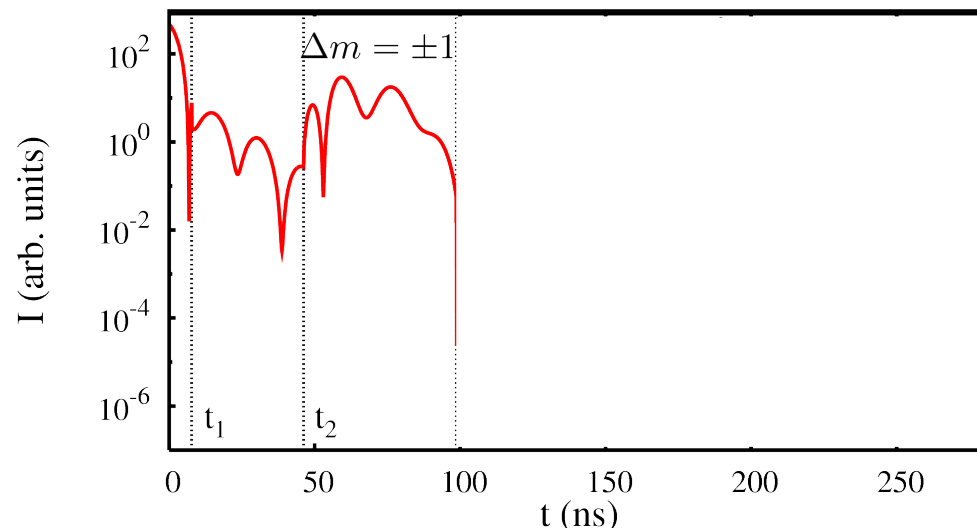


no switching - switching



Step 3: Releasing circular polarization

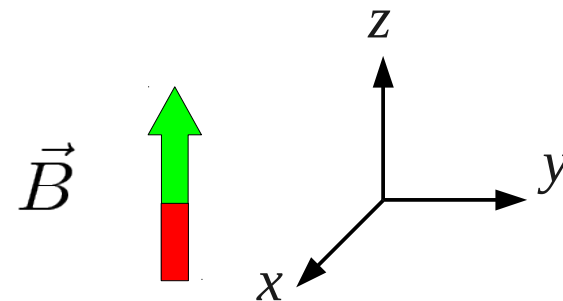
- Initially, magnetic field is in z direction
- At time t_1 , cancel decay by rotating into y direction
- At time t_2 , enable decay on $\Delta m = \pm 1$ but continue to suppress $\Delta m = 0$



$$\left\{ \frac{1}{2} \rightarrow \frac{1}{2}, -\frac{1}{2} \rightarrow -\frac{1}{2} \right\}$$

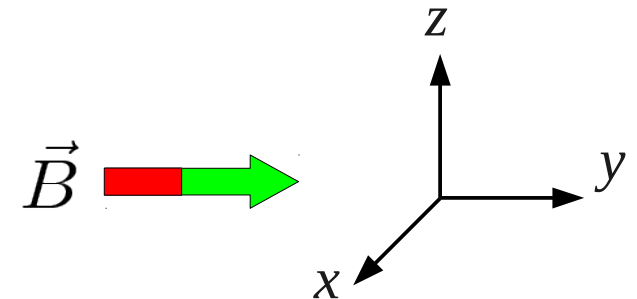
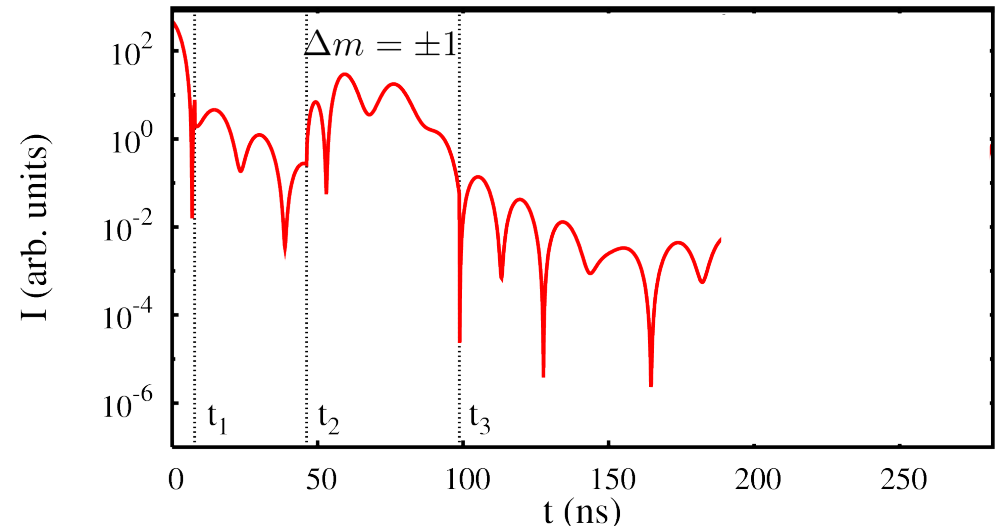
$$\left\{ -\frac{1}{2} \rightarrow \frac{1}{2}, \frac{1}{2} \rightarrow -\frac{1}{2} \right\}$$

$$\left\{ \frac{3}{2} \rightarrow \frac{1}{2}, -\frac{3}{2} \rightarrow -\frac{1}{2} \right\}$$



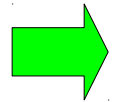
Step 4: Canceling coherent decay

- ▶ 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

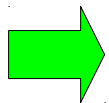
How to implement EIT in x-ray cavity?



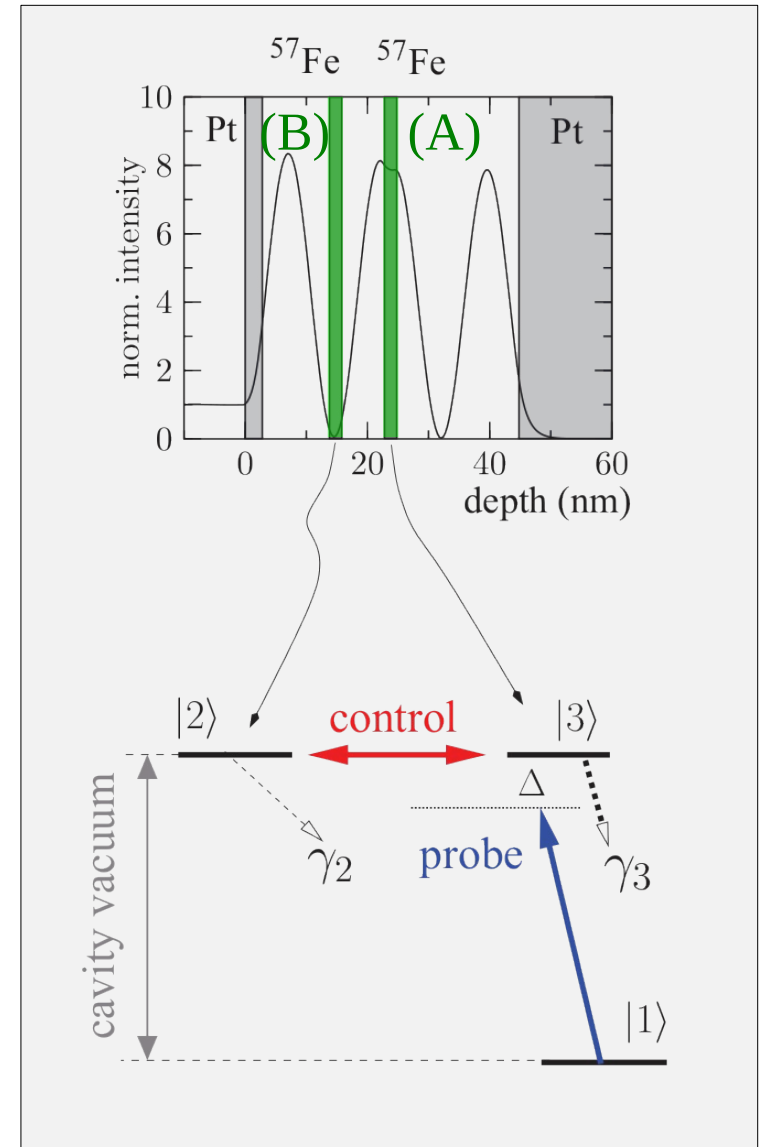
Next talk

How can one

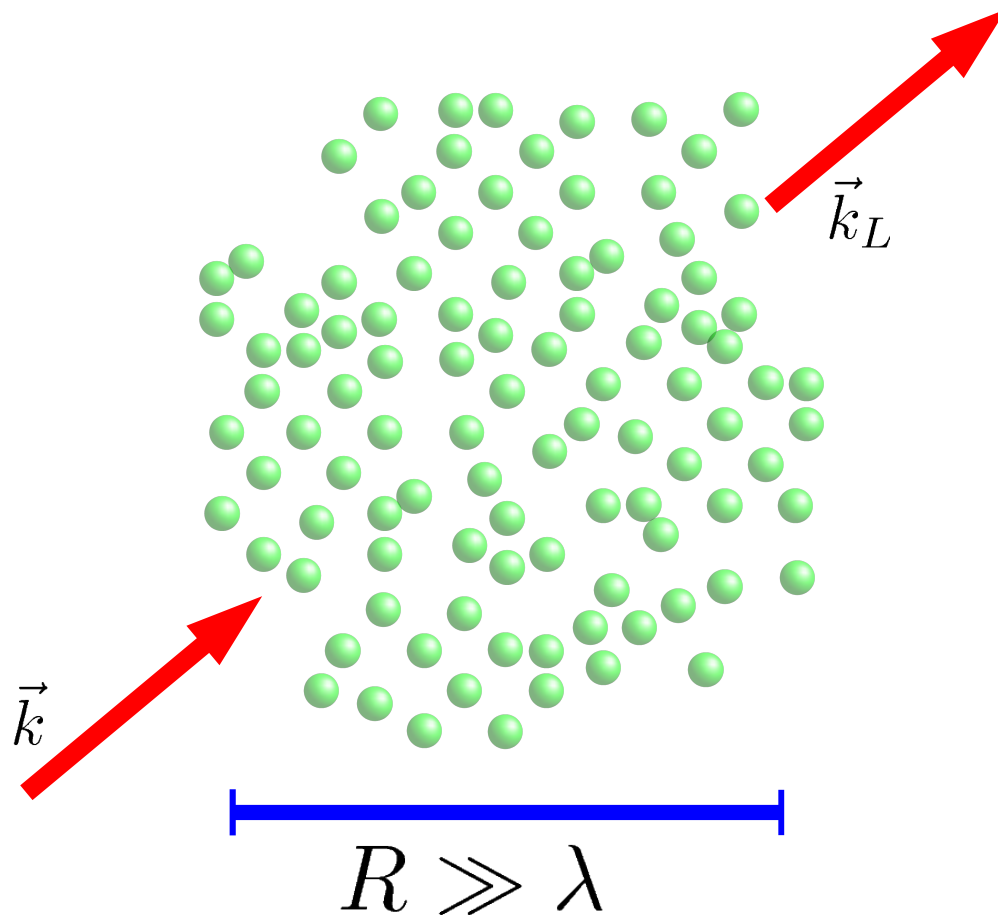
- ▶ Control and systematically study EIT without building many cavities?
- ▶ Engineer more complex level schemes?



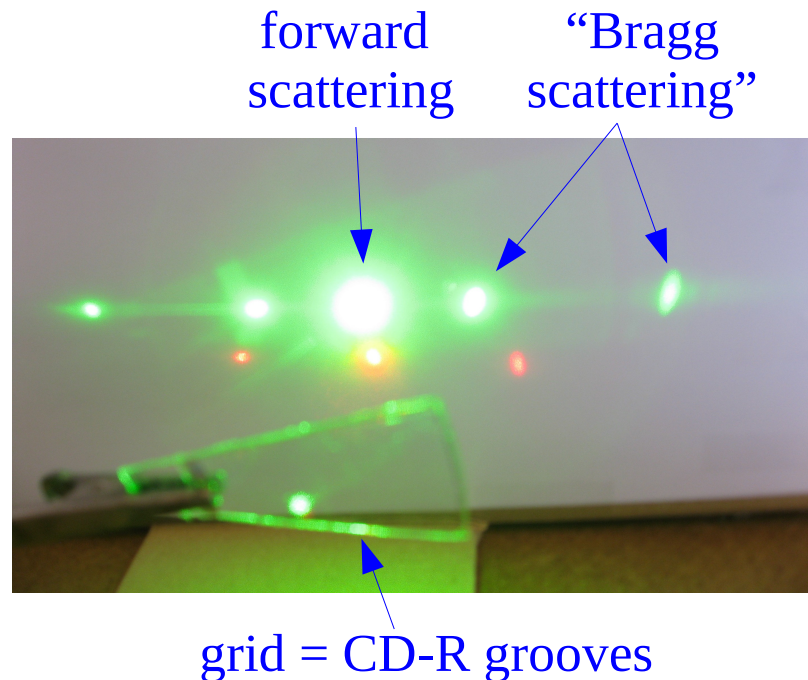
Poster by Kilian Heeg



Coherent forward scattering

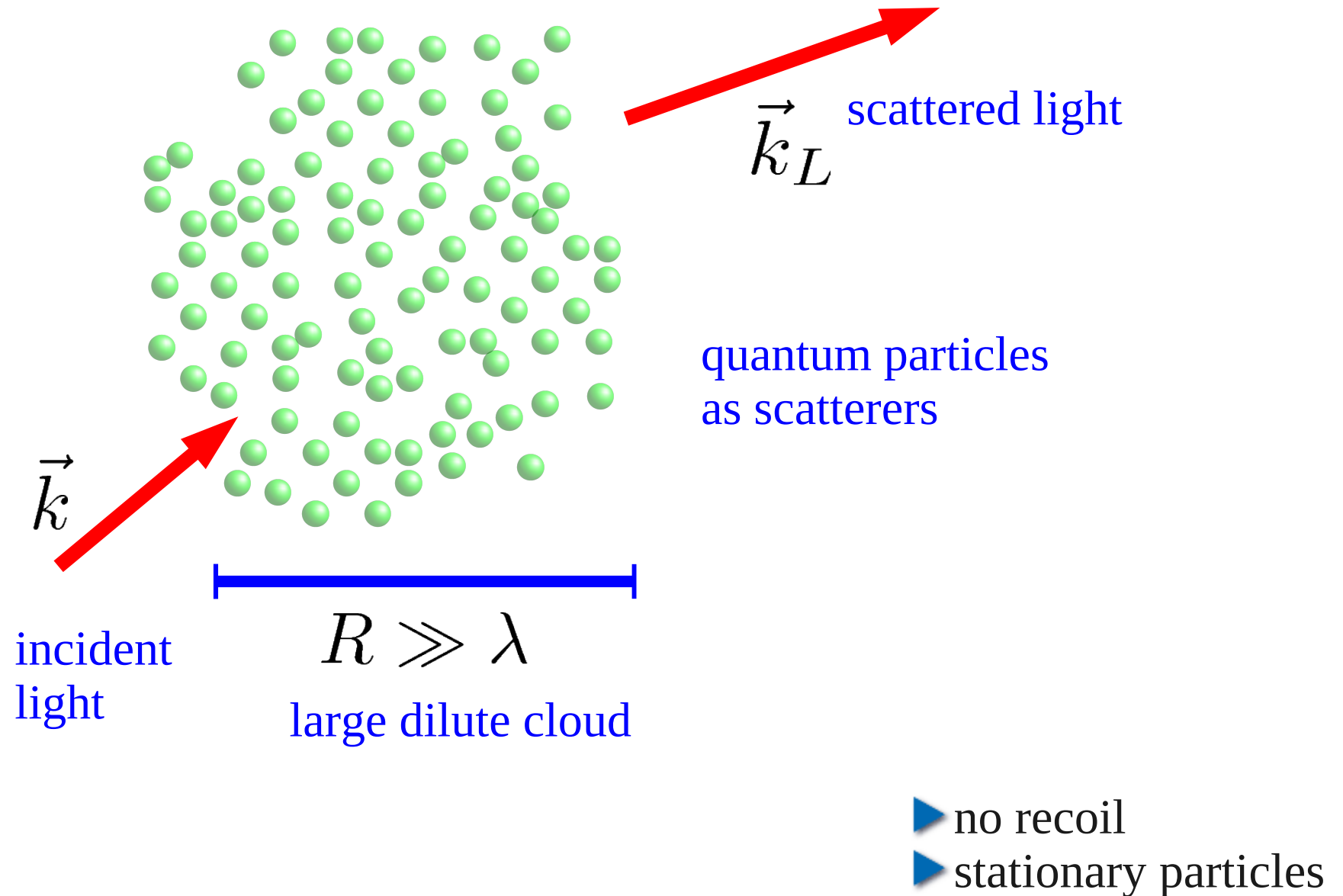


- ▶ Coherent scattering occurs in forward direction
- ▶ Similarity to multi-slit / grid diffraction but constructive interference only in forward / Bragg direction



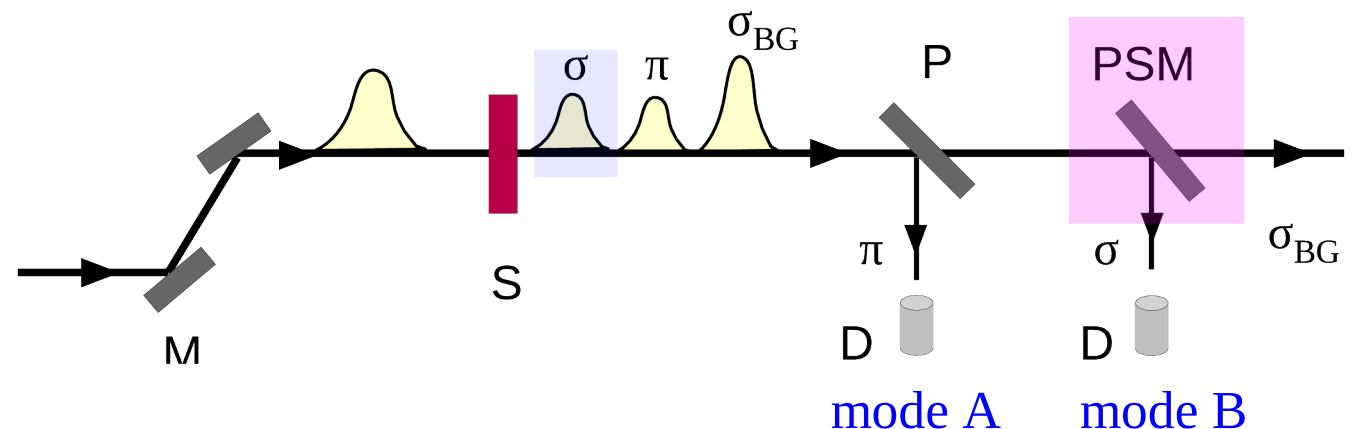
$$\lim_{N \rightarrow \infty} \sum_{i=1}^N e^{i(\vec{k} - \vec{k}_L) \cdot \vec{r}_i} \sim \delta(\vec{k} - \vec{k}_L)$$

Cooperative light scattering



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

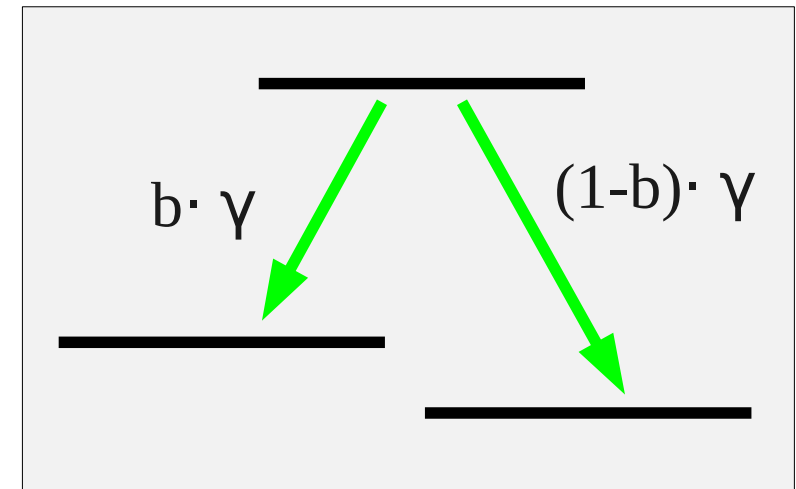


1) A. Grigoriev et al., Appl. Phys. Lett. 89, 021109 (2006)

Branching ratio

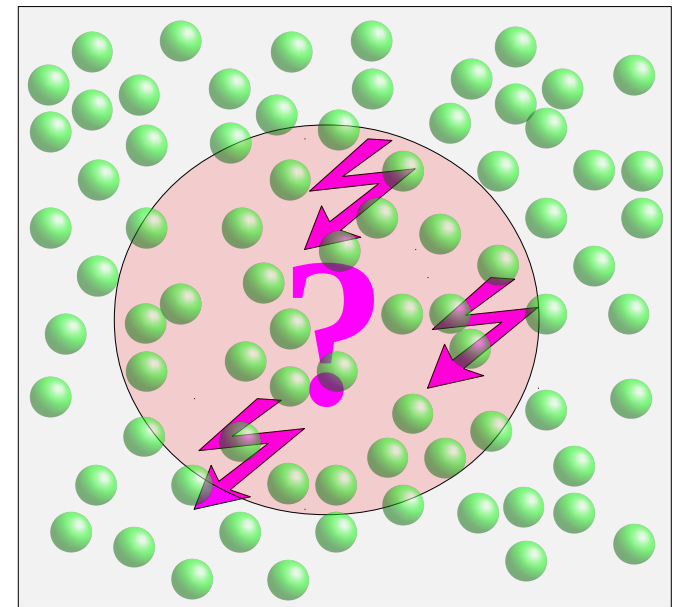
Single particle branching ratio:

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



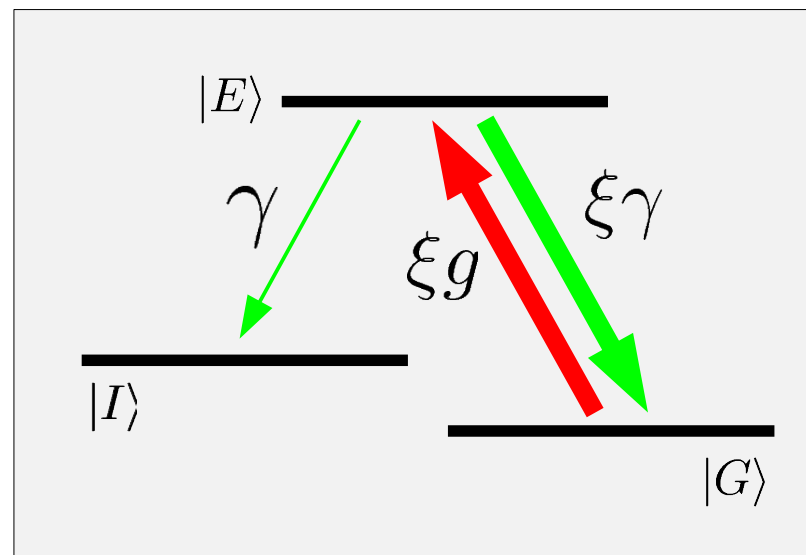
Branching ratio in ensembles

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



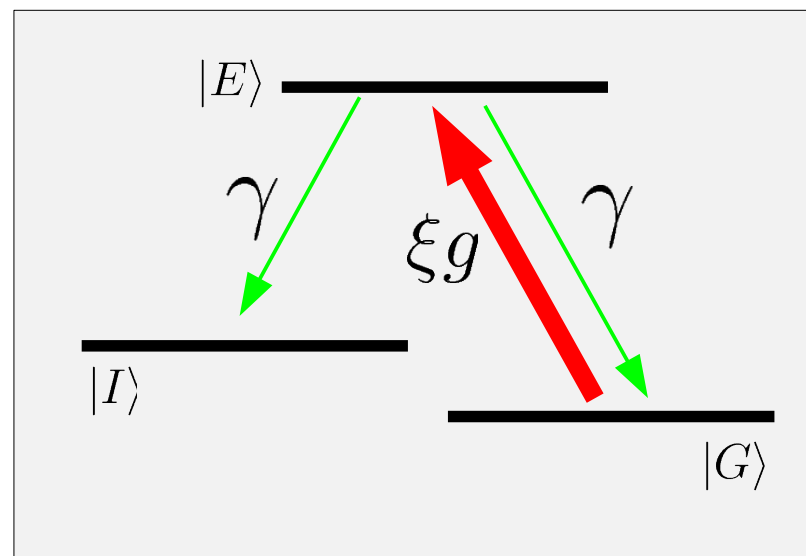
Motivation

- ▶ 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 $|I\rangle$



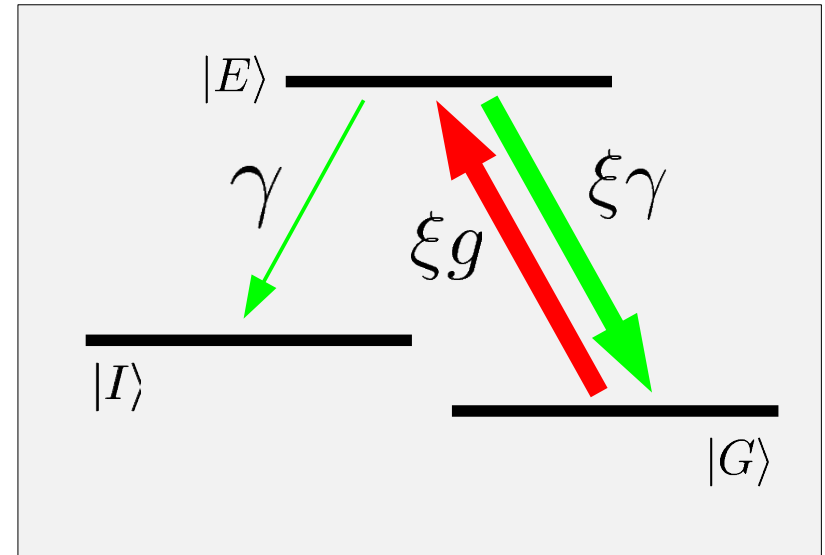
Idea:

- ▶ 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:

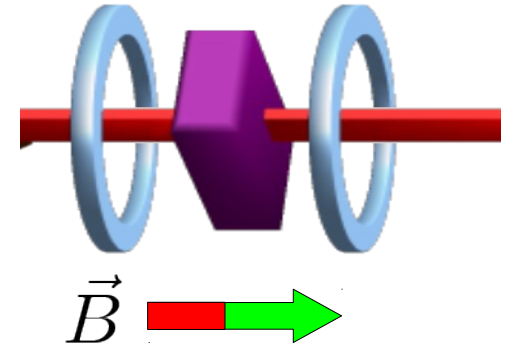
$$b_c^C / b_c^{NC} = \xi + 1$$

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

How to control?

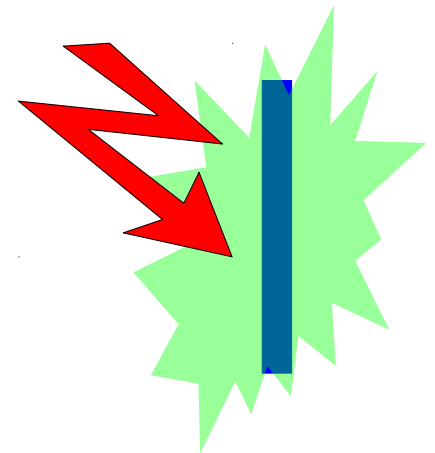
Magnetic switching:

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

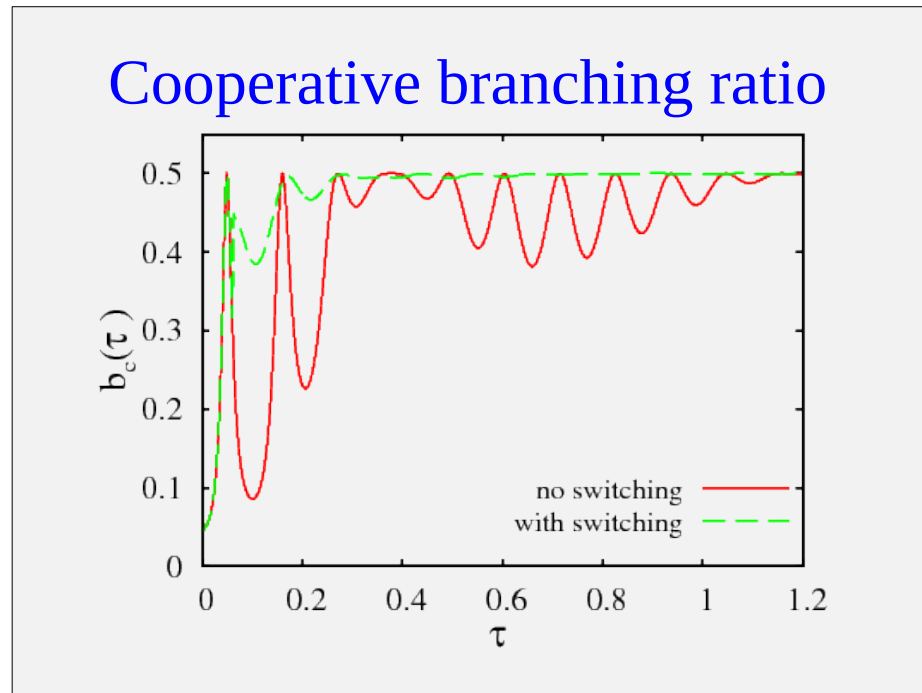


Destroy phase coherence:

- ▶ 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



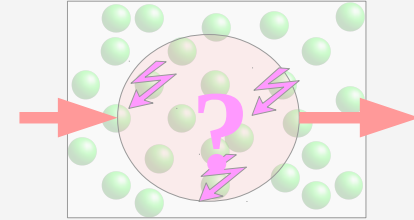
The magnetic switching case



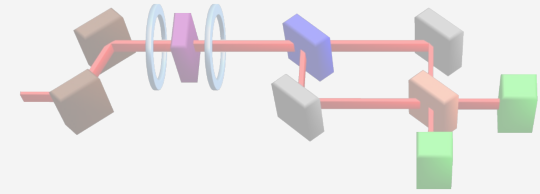
- ▶ Branching ratio time dependent as expected
- ▶ Cooperative branching ratio smaller than single-particle ratio due to superradiance
- ▶ After switching, single-particle branching ratio is achieved
- ▶ With destruction of phase coherence, single-particle ratio can immediately be achieved

Outline

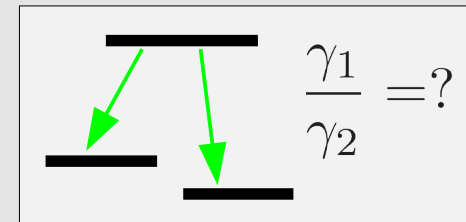
Introduction



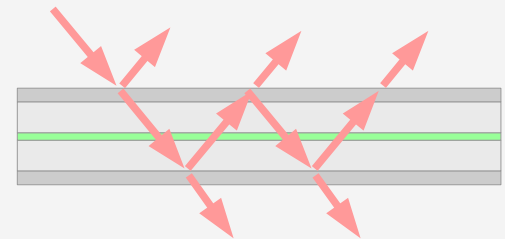
X-ray entanglement generation



X-ray branching ratio control

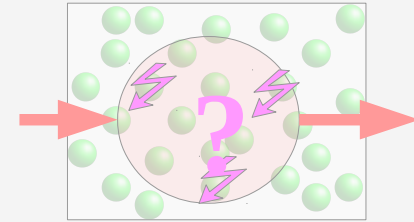


Outlook: Engineering advanced level schemes

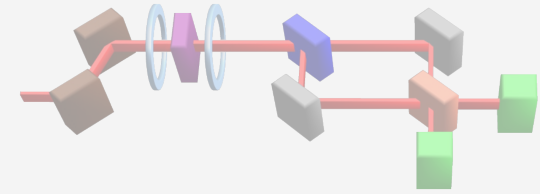


Outline

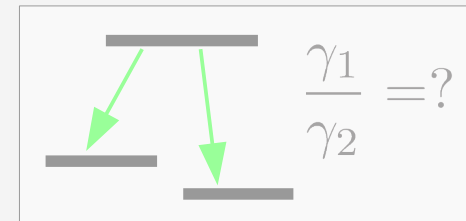
Introduction



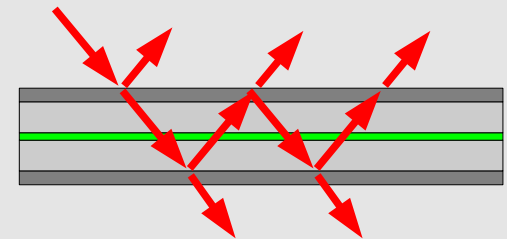
X-ray entanglement generation



X-ray branching ratio control

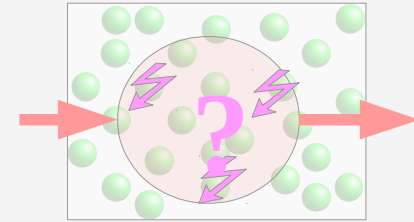


Outlook: Engineering advanced level schemes

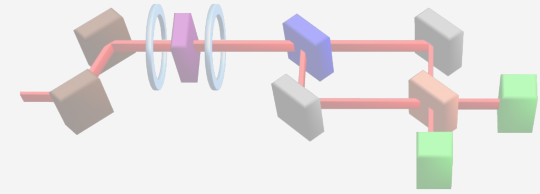


Outline

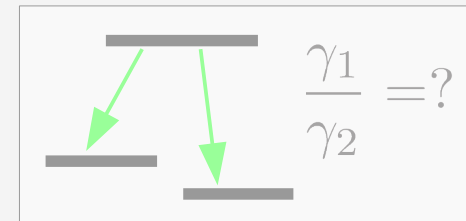
Introduction



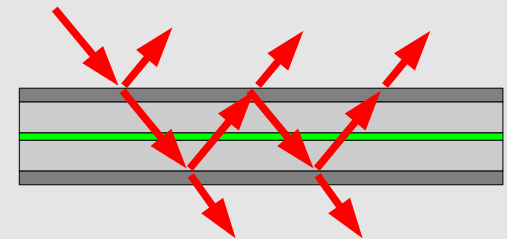
X-ray entanglement generation



X-ray branching ratio control



Outlook: Engineering advanced level schemes



Motivation

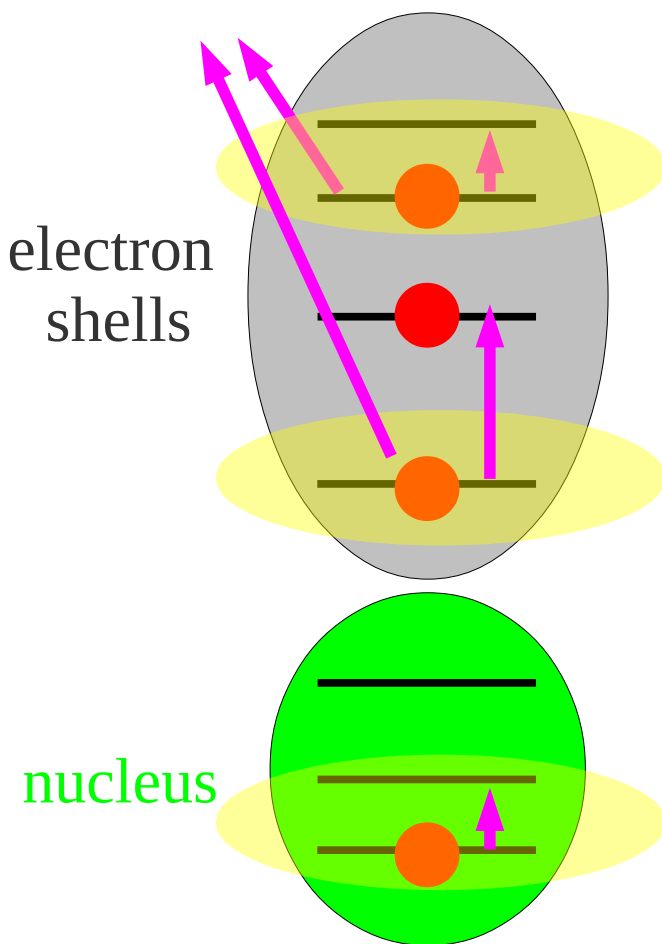
Light-matter interactions



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:

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

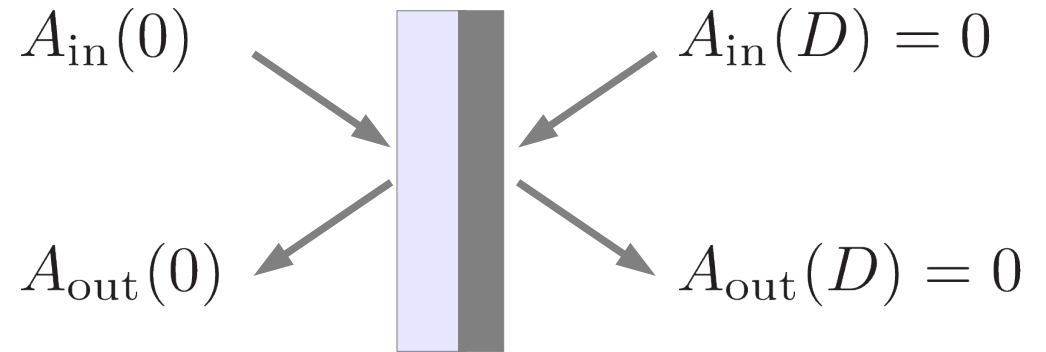
propagation equation:

$$\frac{d}{dz} \mathbf{A} = i\mathbf{F} \mathbf{A}$$

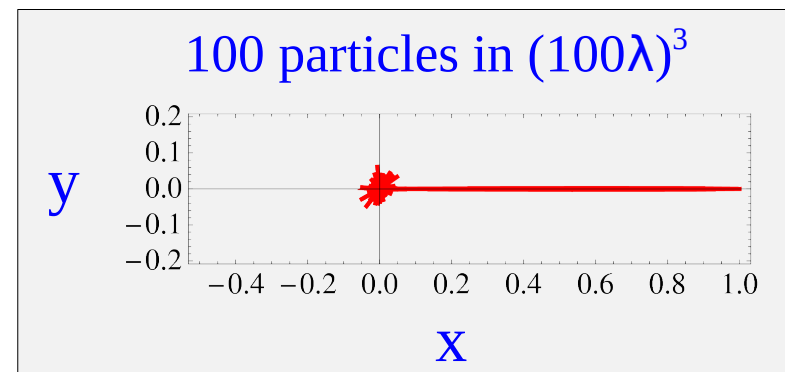
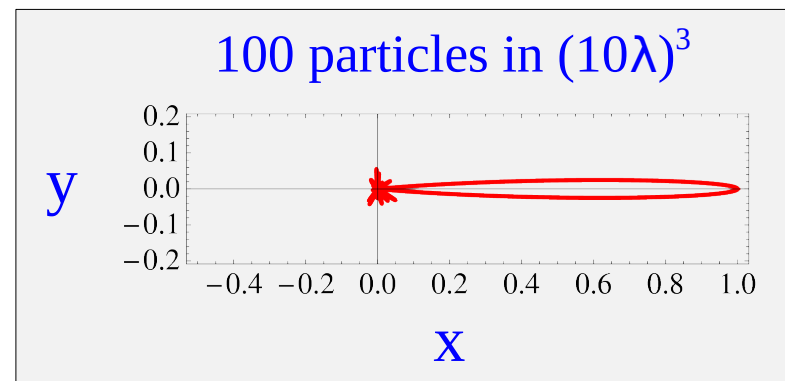
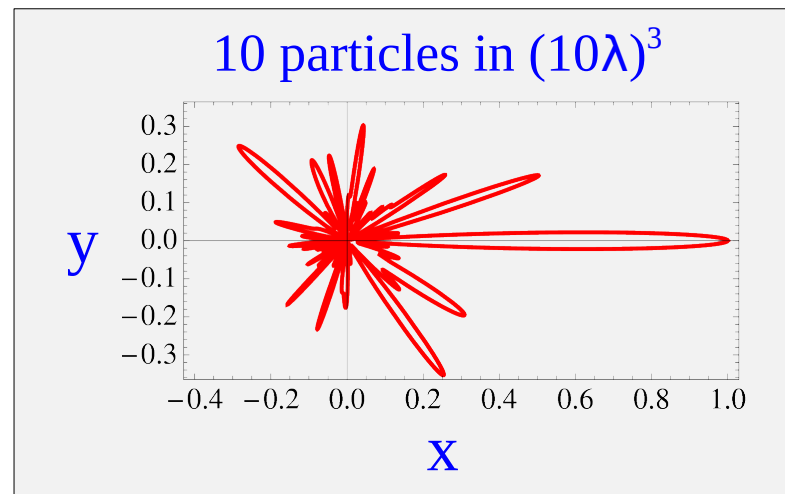
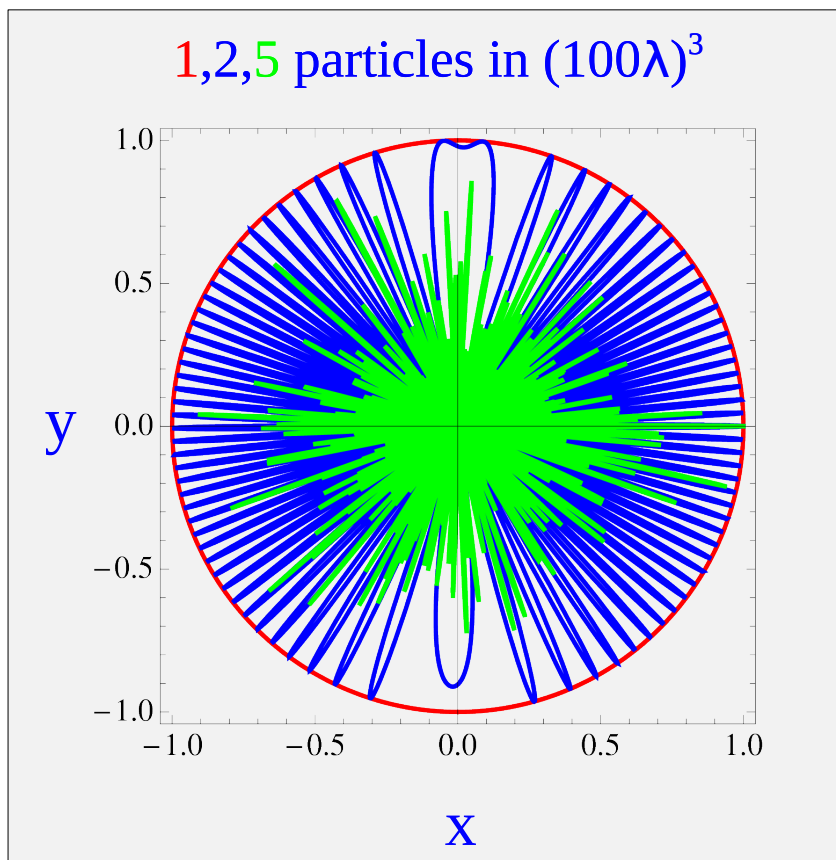
↑ scattering amplitudes

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

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



Directionality



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

Rough efficiency estimate

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

Signal and background
separated!

Incident photon flux
can be increased until
multiple excitations occur

