X-ray photons: From information carriers to compact storage and nanoscale probing

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841. WE-Heraeus Seminar
Important Quantum Technologies

Julius-Maximilians-
UNIVERSITÄT
WÜRZBURG

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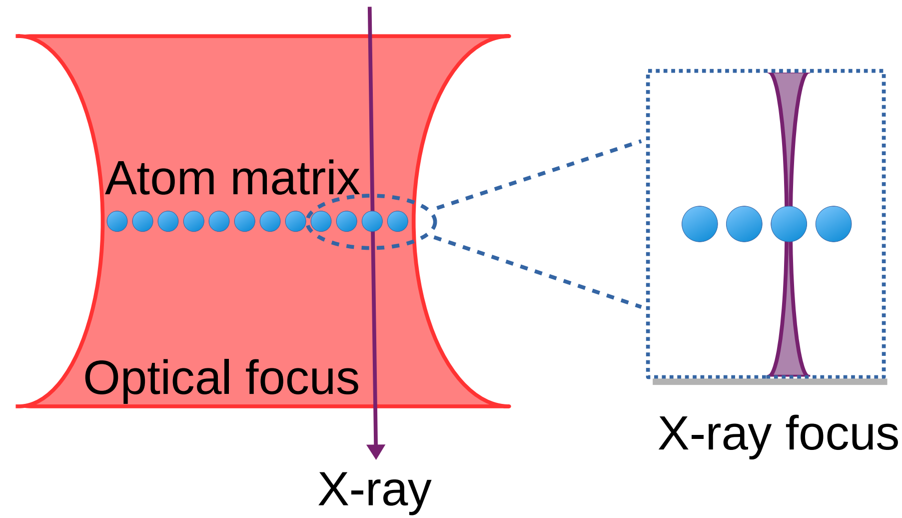
ct.qmat
Complexity and Topology in Quantum Matter

Incentives for x-ray quantum control

- Robustness, efficient detection
- Deeper penetration
- Focusing - diffraction limit

➡ *Ultimate miniaturization of photonic circuits*
Sensing with unprecedented spatial resolution



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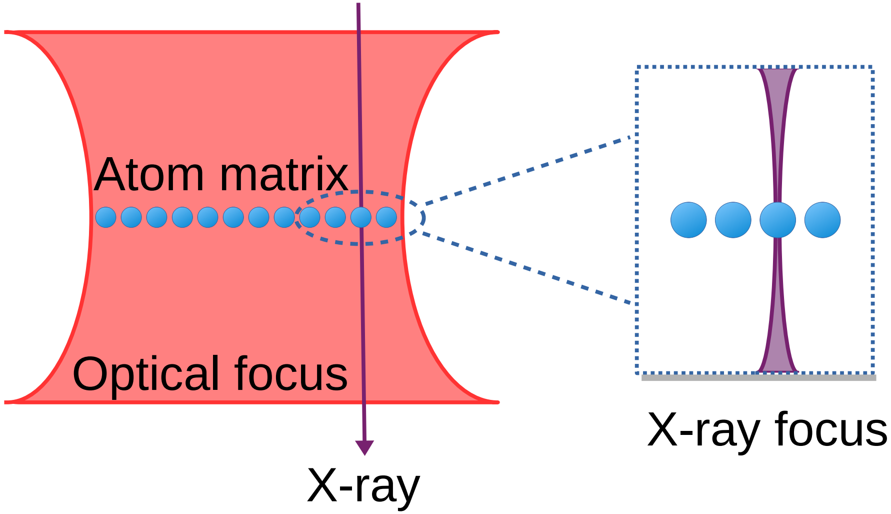
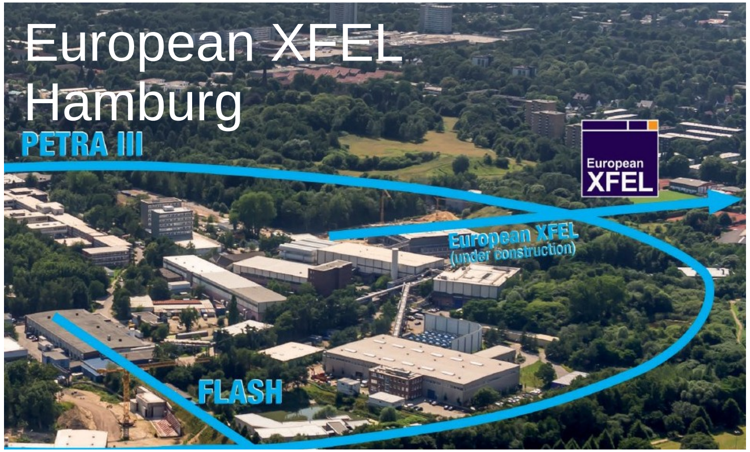
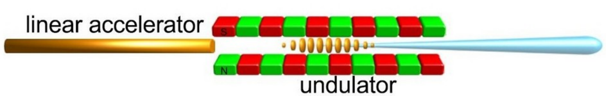


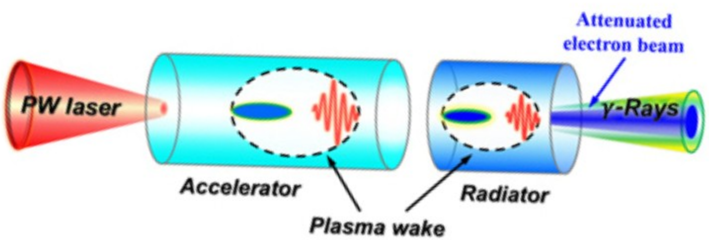


Vision

- Photonic devices (components that manipulate light) for x-rays, down to single photon
- X-ray control with unprecedented spatial and energy resolution

XFEL





Plasma-based sources

filter/control

Photonic device

- Control propagation
- Switches
- Stop and release
- Phase manipulation

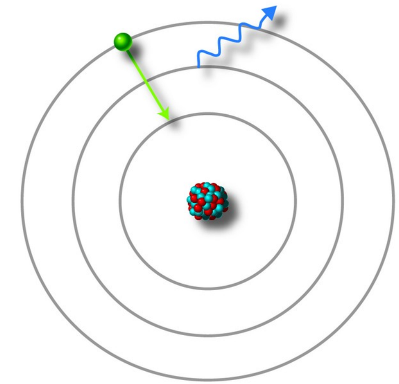
Applications

- Quantum technology: quantum imaging x-ray qubits
- X-ray nanolasers
- Material science
- Biochemistry
- ...

Challenges

So far insufficient control of x-ray photons

- Resonant transitions:
  X-rays are no longer resonant to
  atomic valence electron transitions



- Temporal coherence:
  XFELs lack coherence properties of optical lasers

- Cavities:
  Lack of high-finesse x-ray cavities

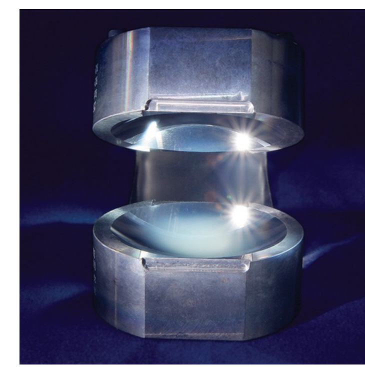
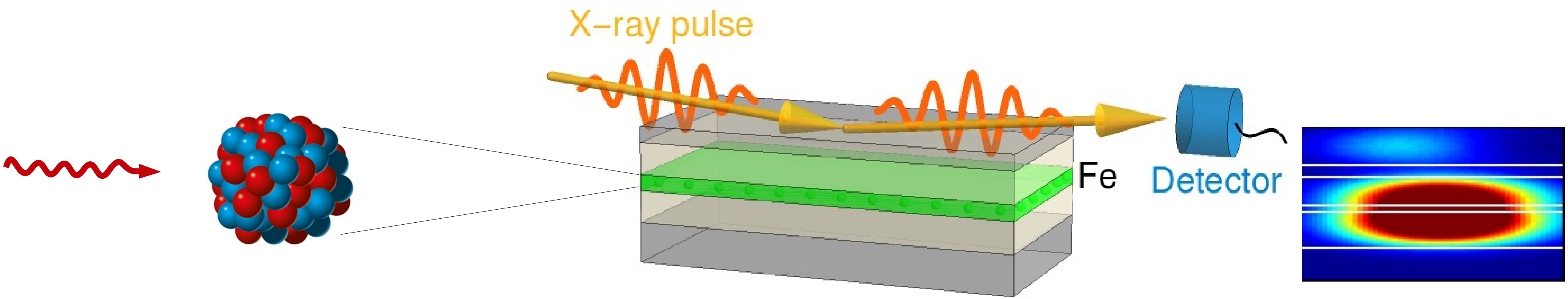


Photo: Nature 446 (2007)

Highly reflective cavity
Haroche group

Developments in x-ray quantum optics

- Use nuclear transitions resonant to x-ray photons
- Collective effects (superradiance) bring means of control
- Thin-film nanostructures with nm-thick nuclear layer(s)

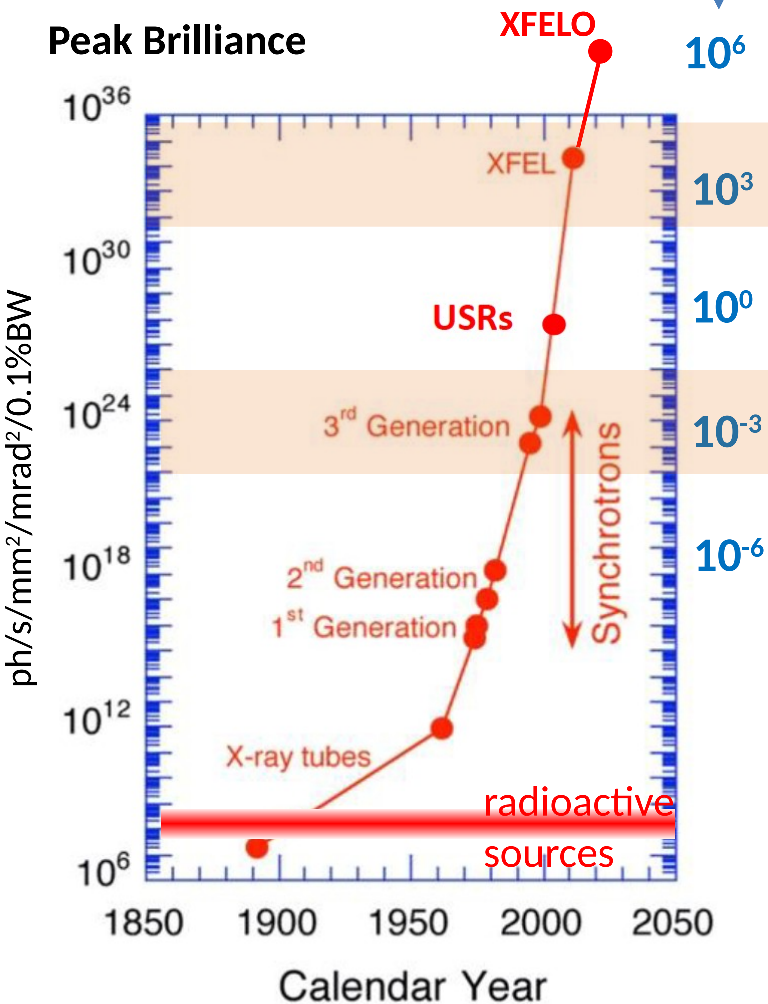


thin-film x-ray cavities
with embedded Fe layer(s)

resonant interaction of x-rays with Mössbauer nuclei – ^{57}Fe with 14.4 keV excitation

X-ray sources

Resonant photons/neV/pulse



14.4 keV resonance of ^{57}Fe

1000 resonant photons / neV / pulse

Game-changing step

1 resonant photon / neV / 1000 pulses

Excitation of ^{45}Sc 12.4 keV nuclear transition with XFEL:

*Y. Shvyd'ko et al, Nature **622**, 471 (2023)*

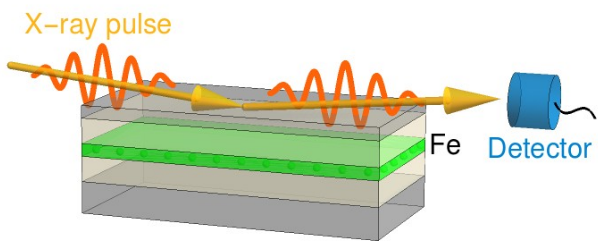
2023/4: Experiments with ^{57}Fe, team of R. Röhlsberger

Outline

I. X-ray thin film nanostructures

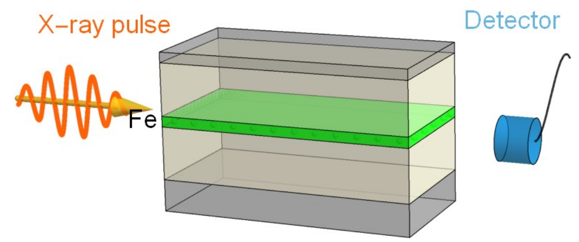
Collective effects and superradiant decay

II. Applications in cavity geometry (grazing incidence)



Storing a single x-ray photon
Playing ping-pong with a single x-ray photon
Dark states and topological effects

III. Applications in waveguide geometry (front coupling)



Front coupling theory and experiment

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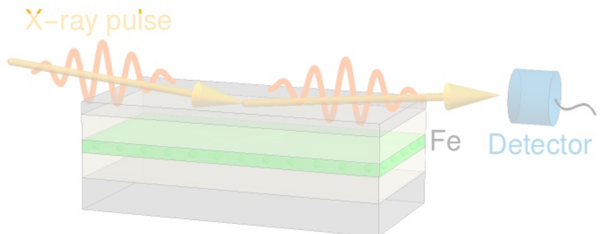

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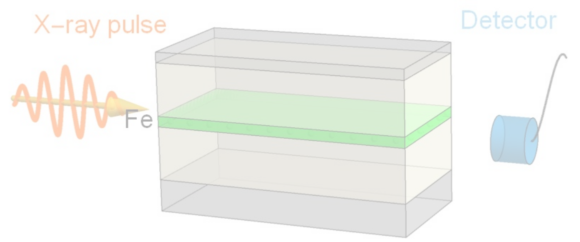
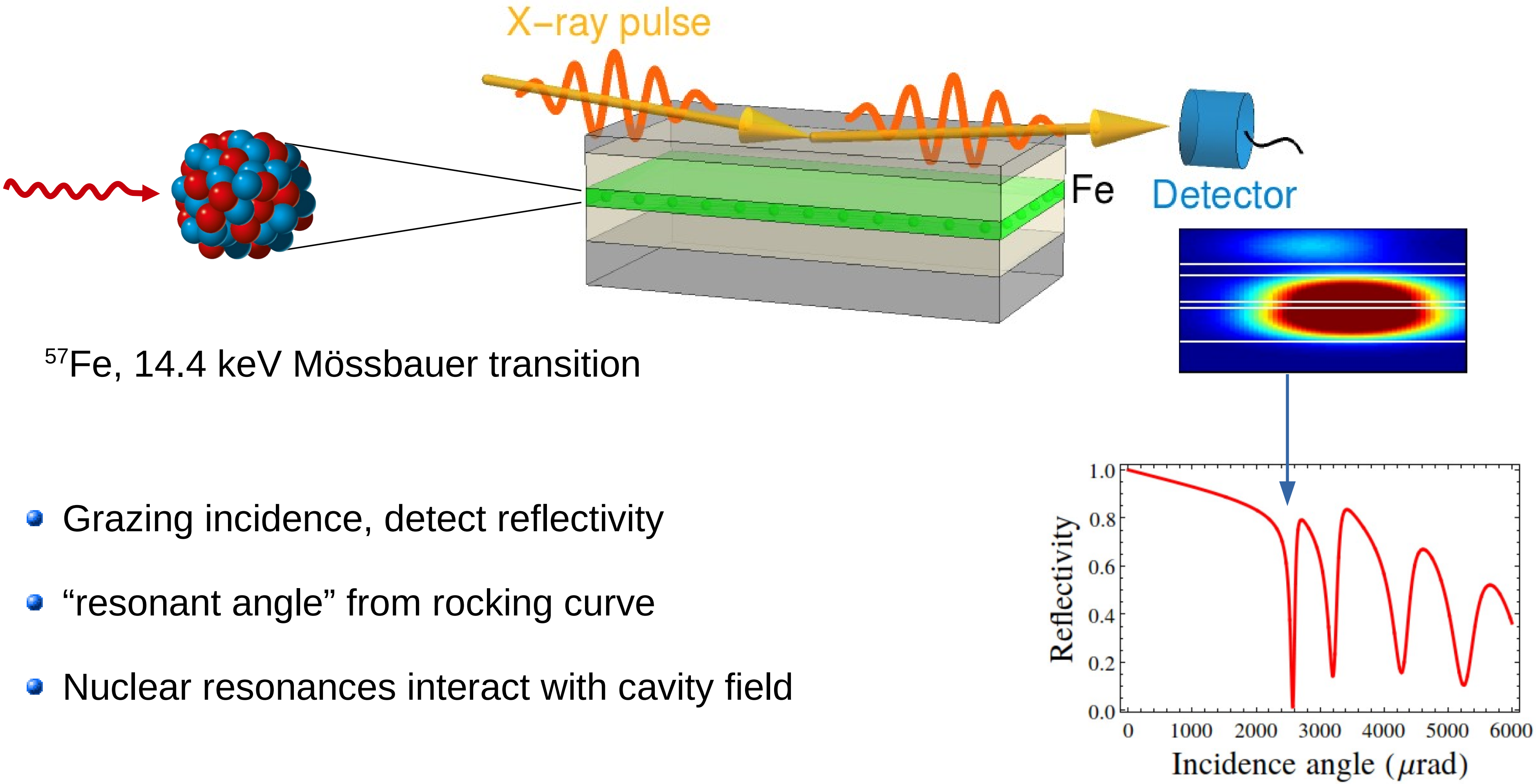


Front coupling theory and experiment

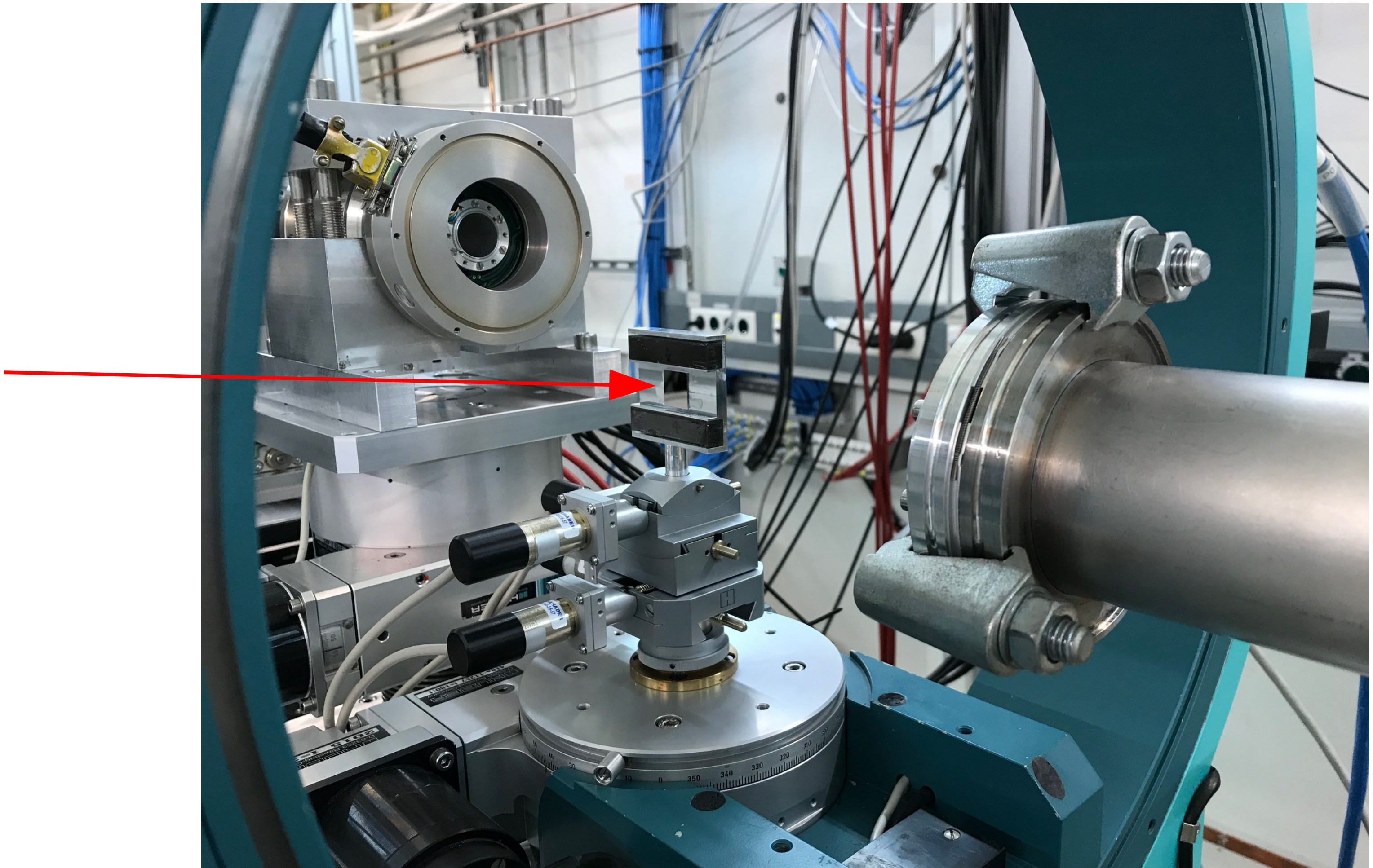
Thin-film x-ray nanostructures



^{57}Fe, 14.4 keV Mössbauer transition

- Grazing incidence, detect reflectivity
- "resonant angle" from rocking curve
- Nuclear resonances interact with cavity field

Thin-film x-ray nanostructures

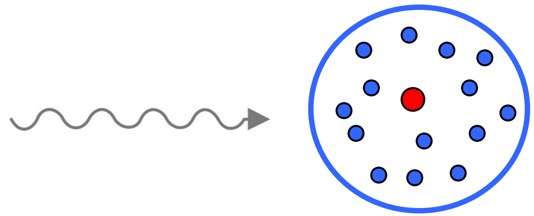


Setup at PETRA III, DESY, Hamburg

Number of excitations in the system

… Synchrotron radiation

~ 0.01 resonant photons/pulse

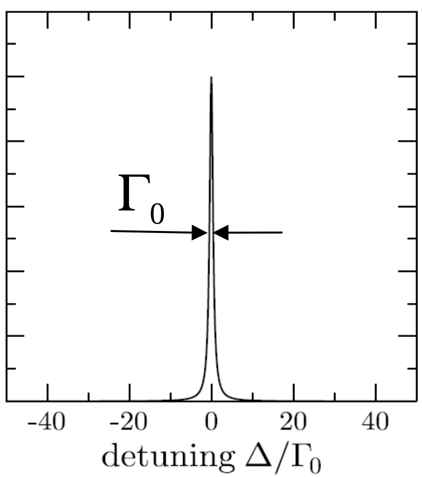


Single-photon collective effects

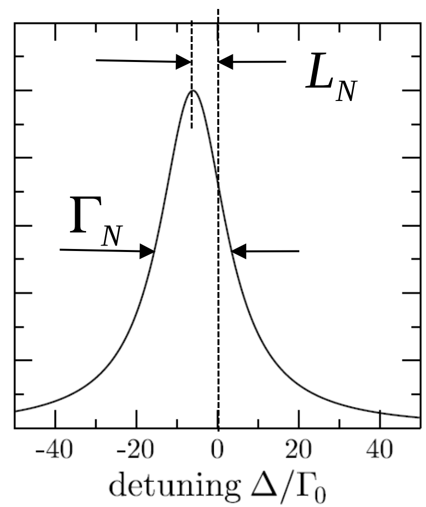
- Single photon superradiance
- Collective Lamb shift

R. Röhlsberger, K. Schlage, B. Sahoo, S. Couet and R. Rüffer, **Science 328, 1248 (2010)**

Single-photon, single-atom



Single-photon, multi-atom



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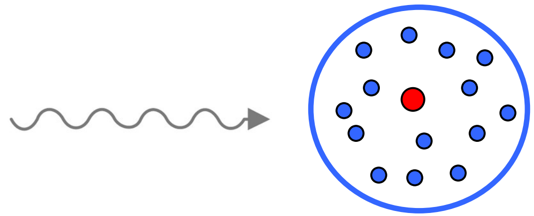

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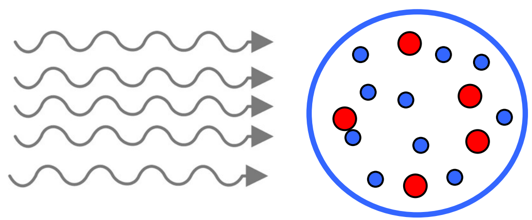
Multi-photon, multi-atom scattering

… XFEL HXRSS radiation

Several 100 resonant photons/pulse



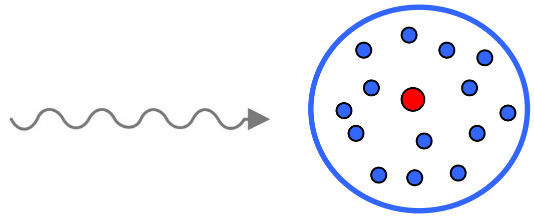
New phenomena !
- **Photon correlations**
- **Nonlinear effects**
- **Entanglement, nonclassical states**
- **Collective effects (e.g. multiphoton collective Lamb shift)**

Multiphoton nuclear resonant scattering

Number of excitations in the system

… Synchrotron radiation

~ 0.01 resonant photons/pulse



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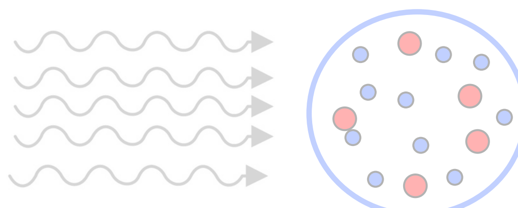

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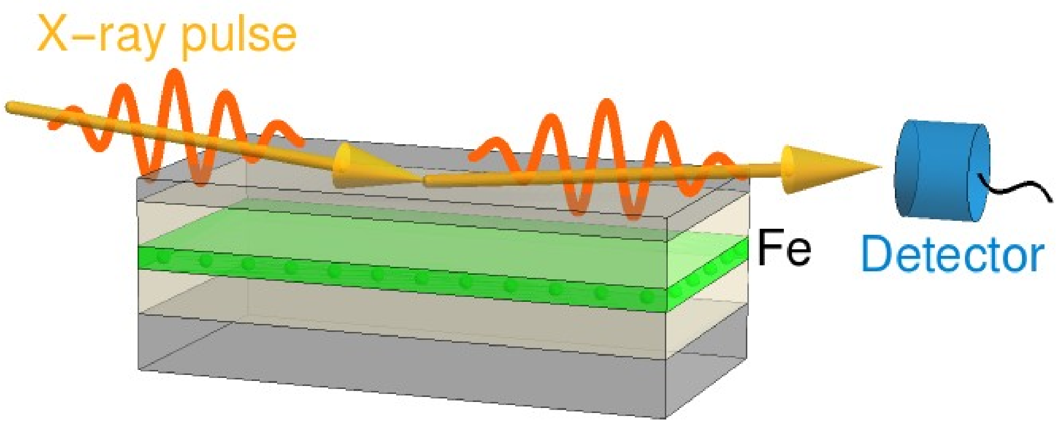
Multiphoton nuclear resonant scattering

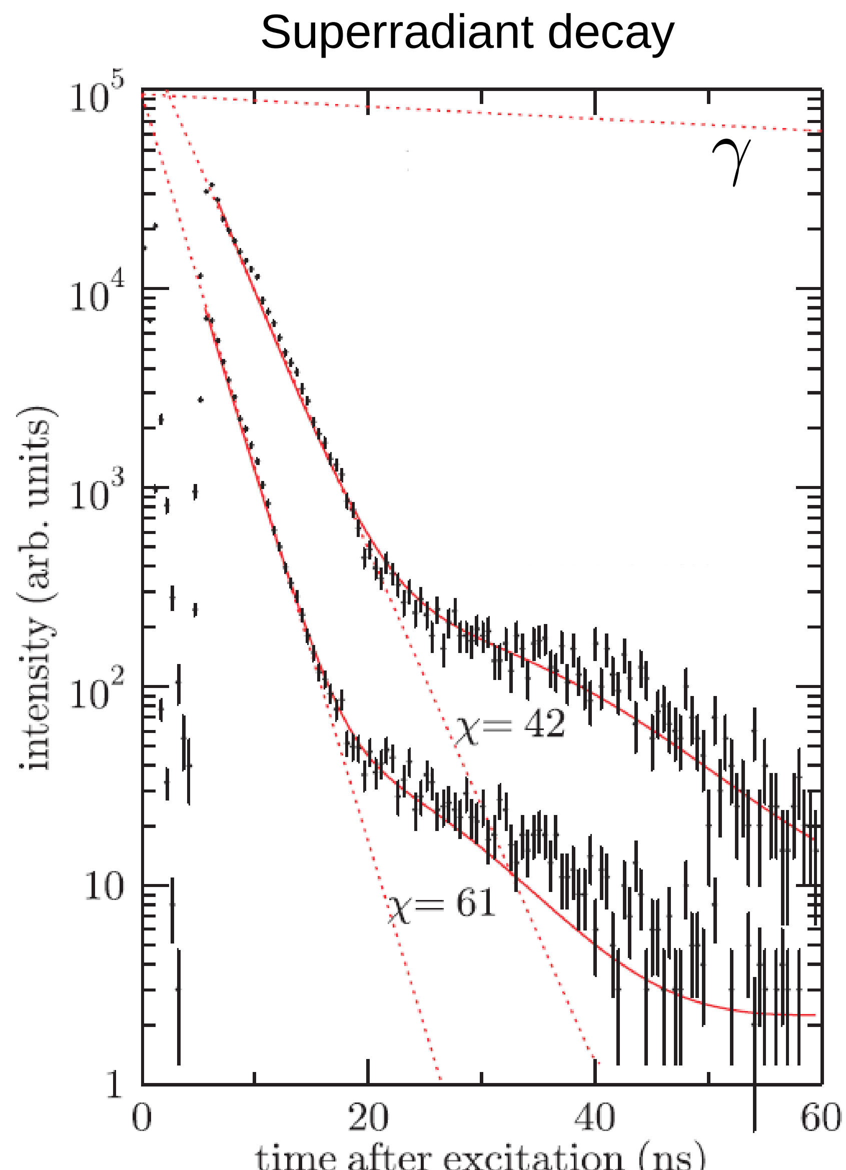
Superradiant decay



- Cavity field excites radiative eigenmode
- χ is the coherent decay enhancement factor

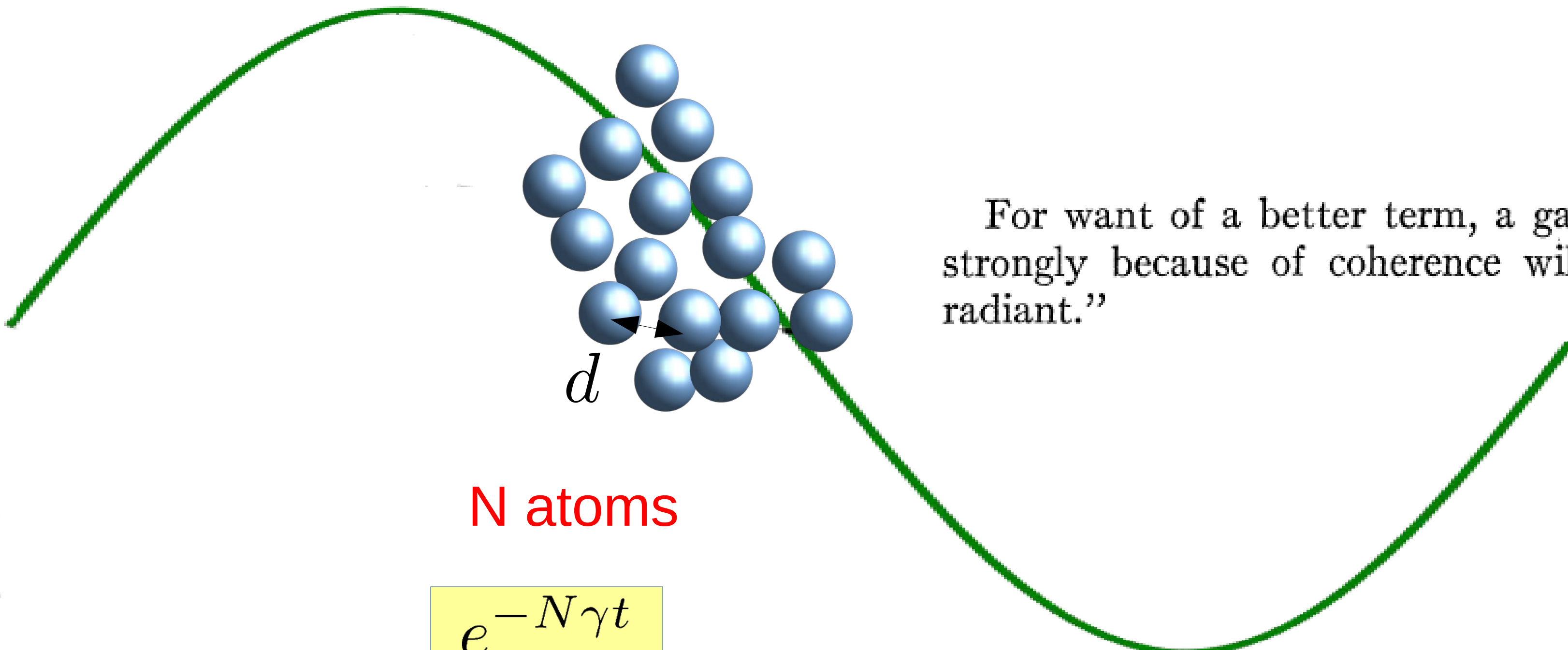
The collective effects are our "control knob" on the system!

*R. Röhlsberger et al, Science **328**, 1248 (2010)*



Dicke model

R. Dicke, Phys. Rev. ***93****, 99 (1954)* $\lambda \gg d$ theory



For want of a better term, a gas which is radiating strongly because of coherence will be called "super-radiant."

N atoms

$$e^{-N\gamma t}$$

Superradiant decay increased by factor N!

N. Skribanowitz et al, Phys. Rev. Lett. ***30****, 309 (1973)* experiment

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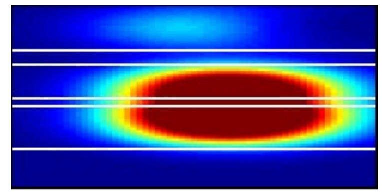
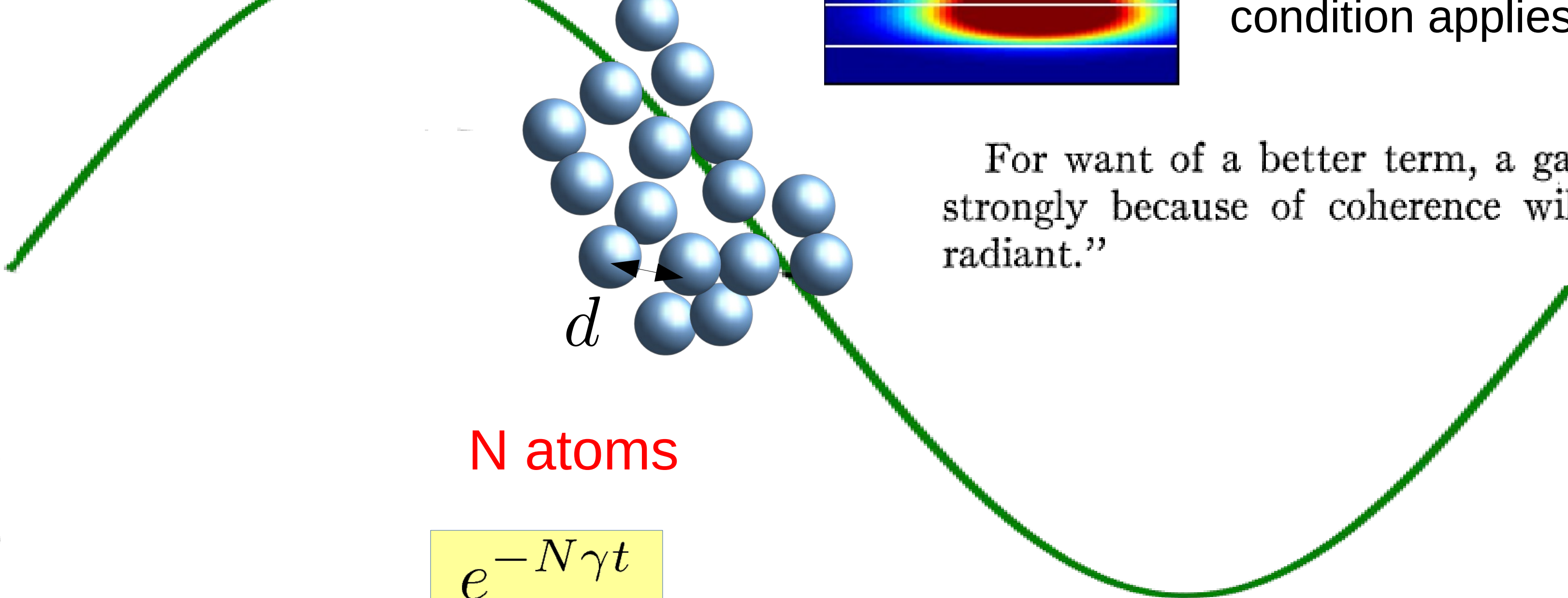


For the thin-film nanostructure, condition applies for standing wave



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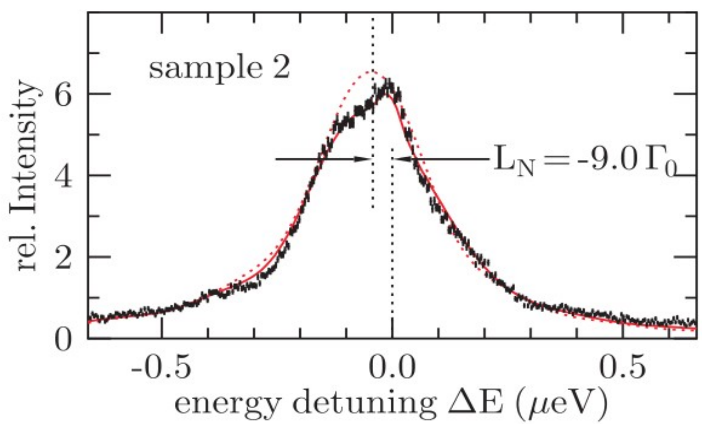
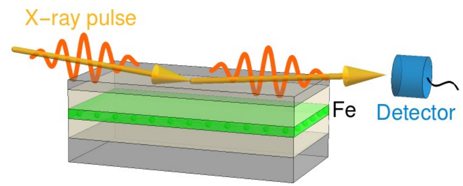
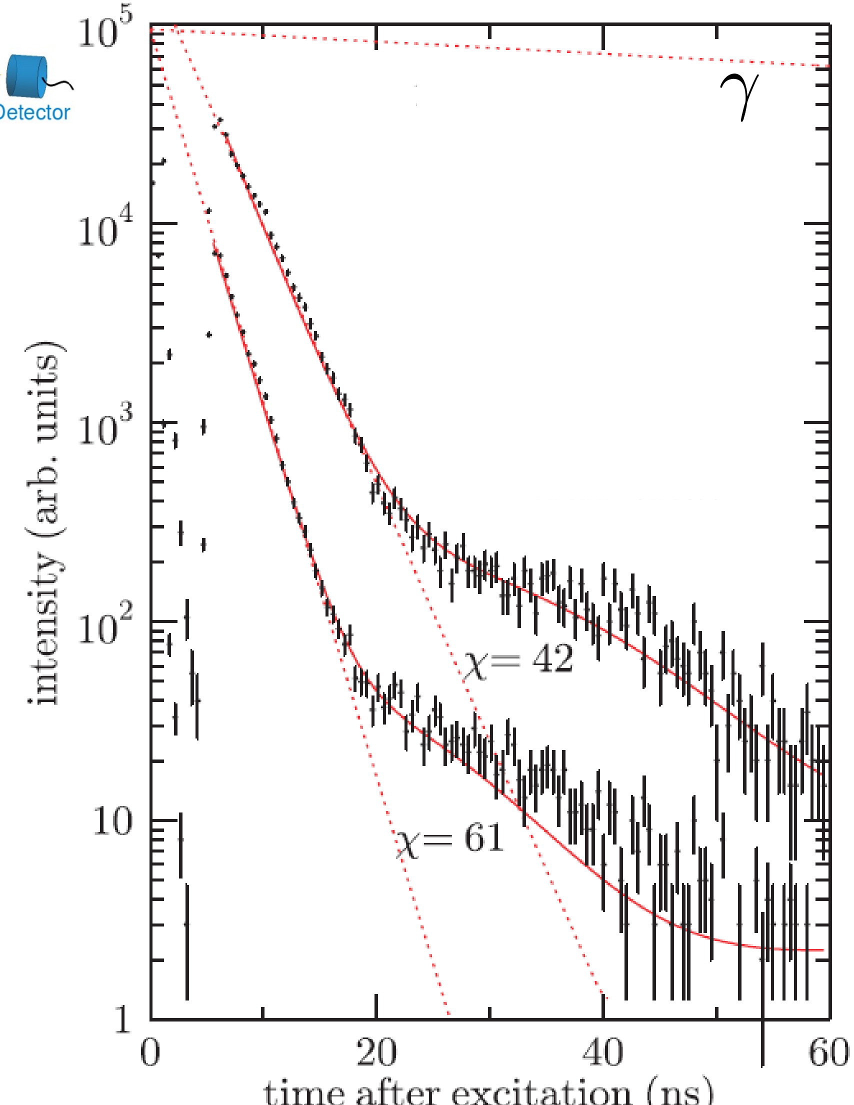
Thin-film x-ray cavities

Collective Lamb shift





Superradiant decay



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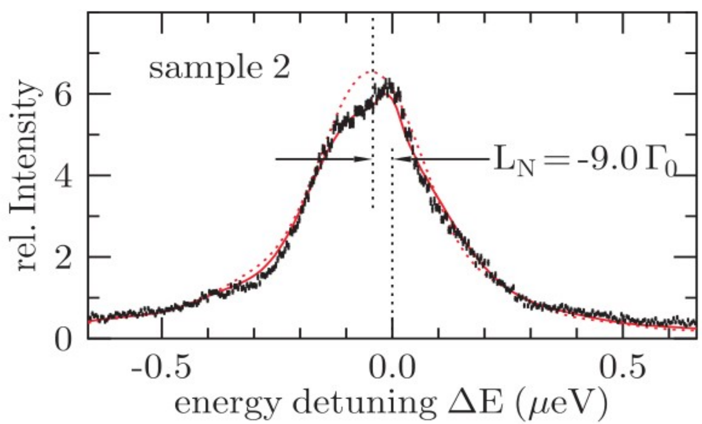
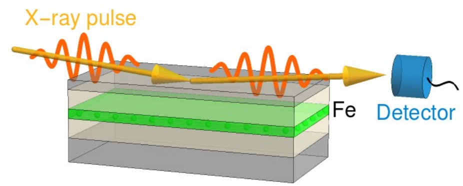
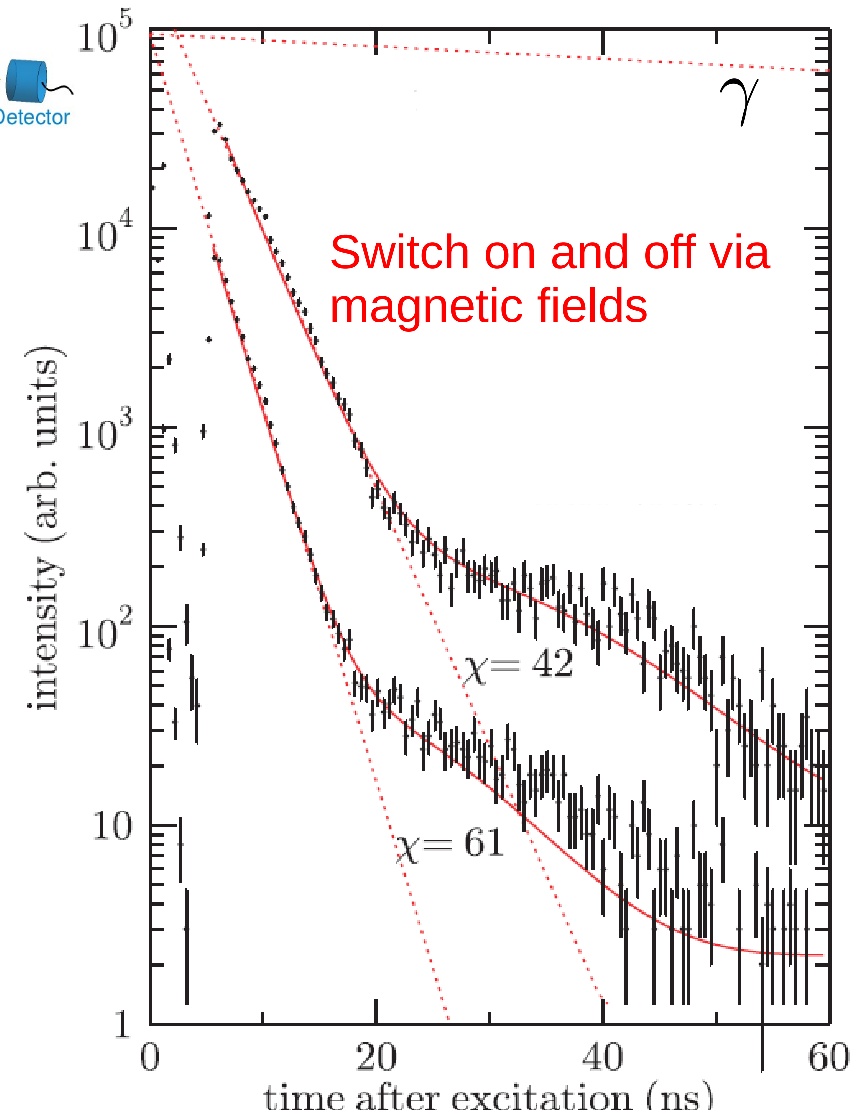




Superradiant decay



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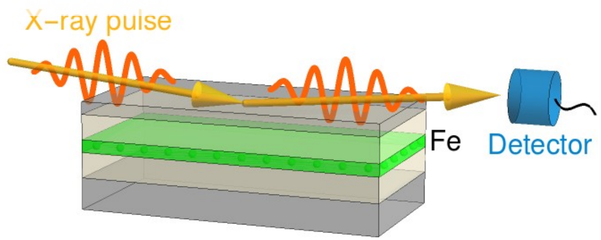

Storing a single x-ray photon
Playing ping-pong with a single x-ray photon
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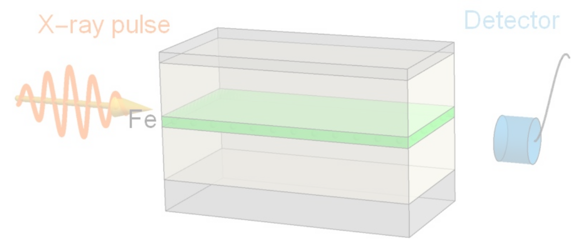
III. Applications in waveguide geometry (front coupling)



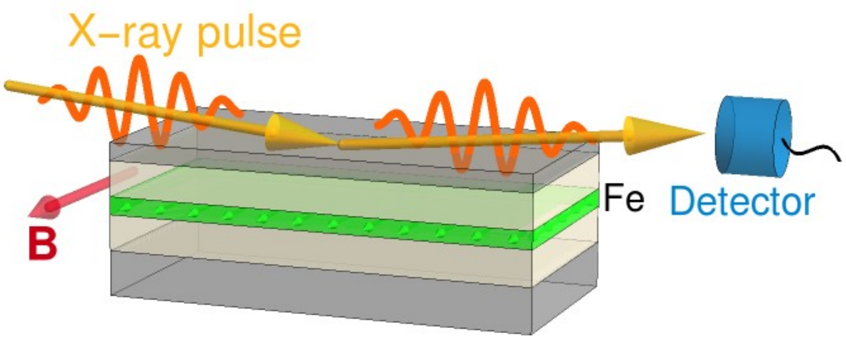
Front coupling theory and experiment

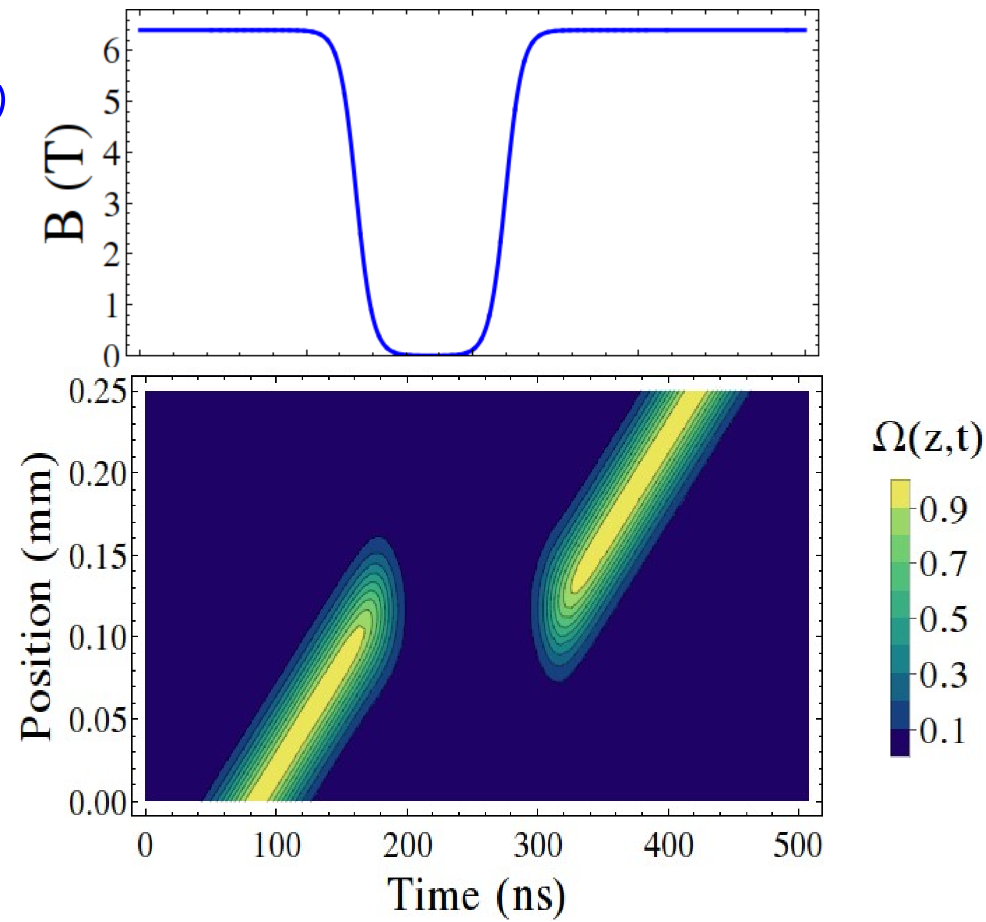
1. Storing single x-ray photons

Switch off the coherent (superradiant) decay via magnetic switching

- Thick targets, normal incidence
  Liao, Pálffy, Keitel, Phys. Rev. Lett. ***109****, 197403 (2012)*

- Thin-film cavities: stopping light mechanism
  Pulse mapped to nuclear coherences
  Kong and Pálffy, Phys. Rev. Lett. ***116****, 197402 (2016)*





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“strong coupling” with x-rays

interpreted as “x-ray ping-pong”

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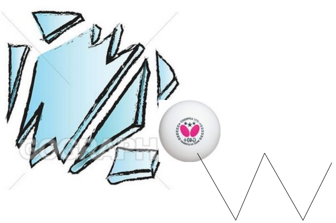


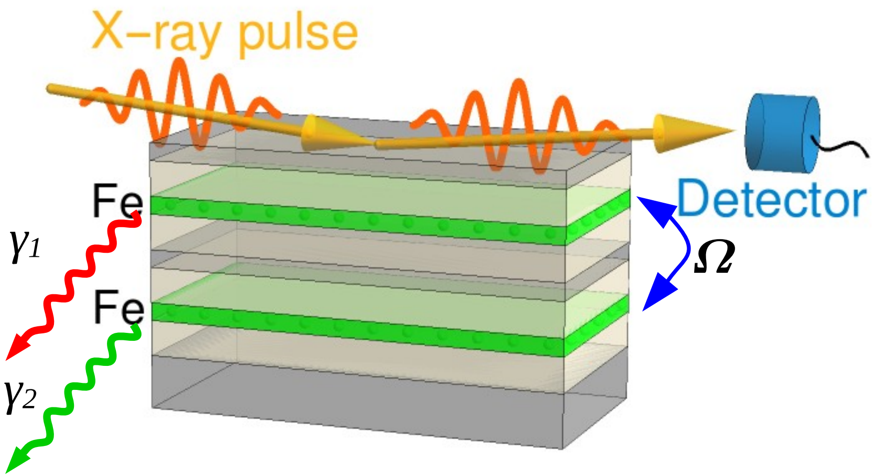
interpreted as “x-ray ping-pong”

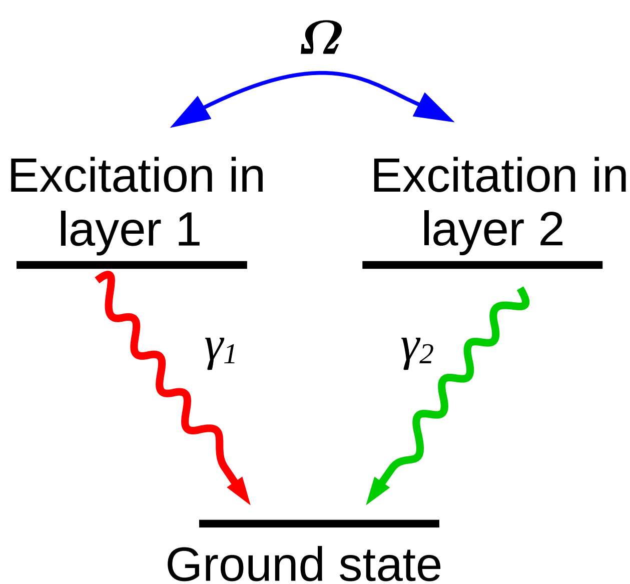
2. Mimicking strong coupling

“strong coupling” with x-rays
= many successful exchanges
until ball lost

interpreted as “x-ray ping-pong”

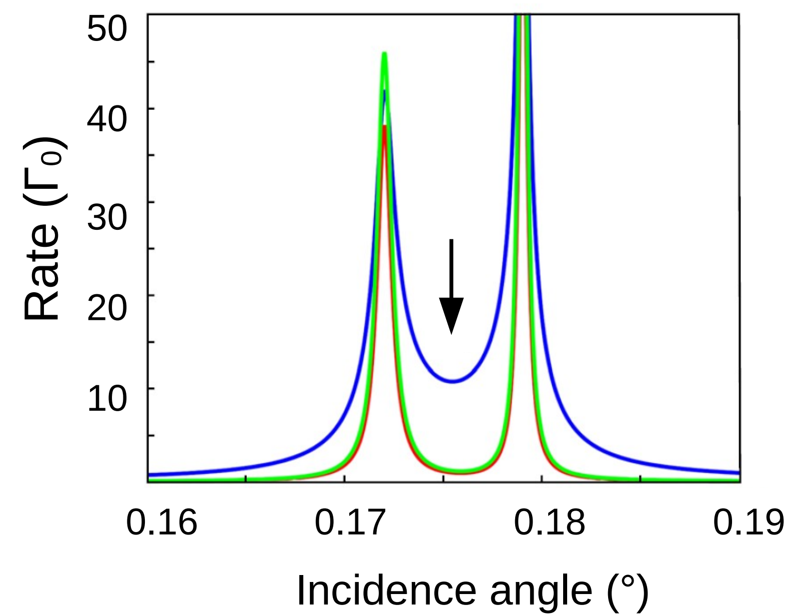
Control of inter-layer coupling



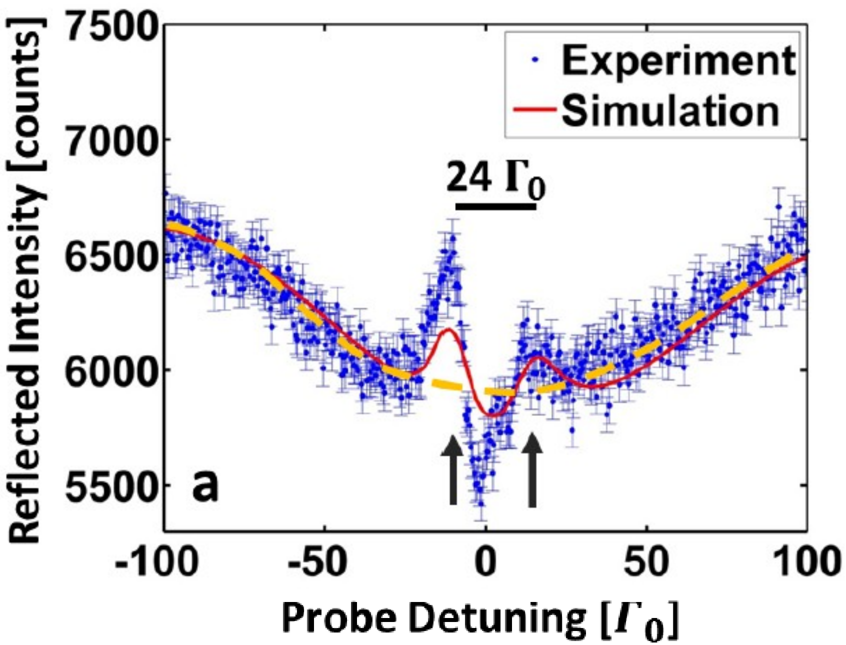


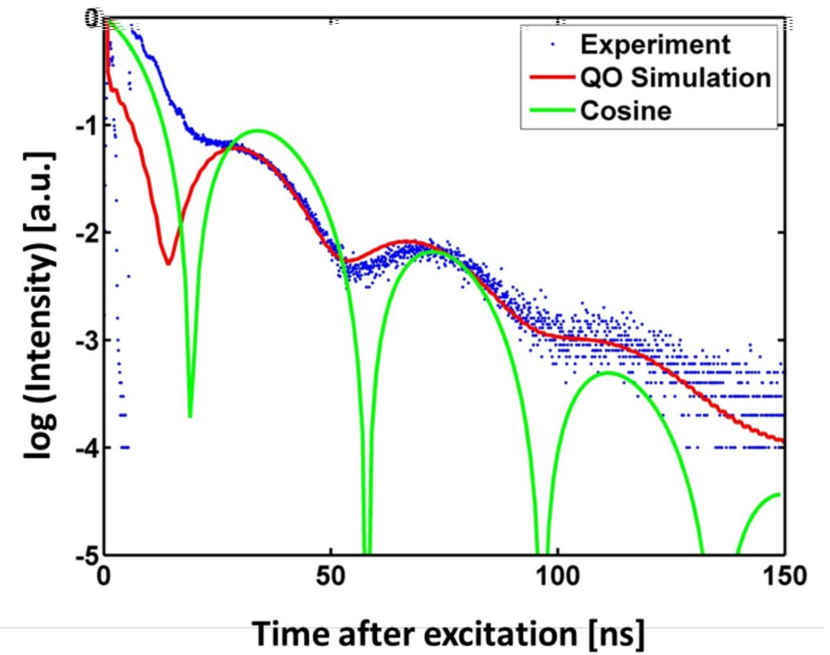
 resonant x-ray photon

 excitation in layer 1 or 2



Experimental confirmation





The resonance line is split and one can observe Rabi oscillations as known from the strong coupling regime!

Haber, ... Pálffy, Röhlsberger*,*
Nature Photon. ***11****, 720 (2017)*

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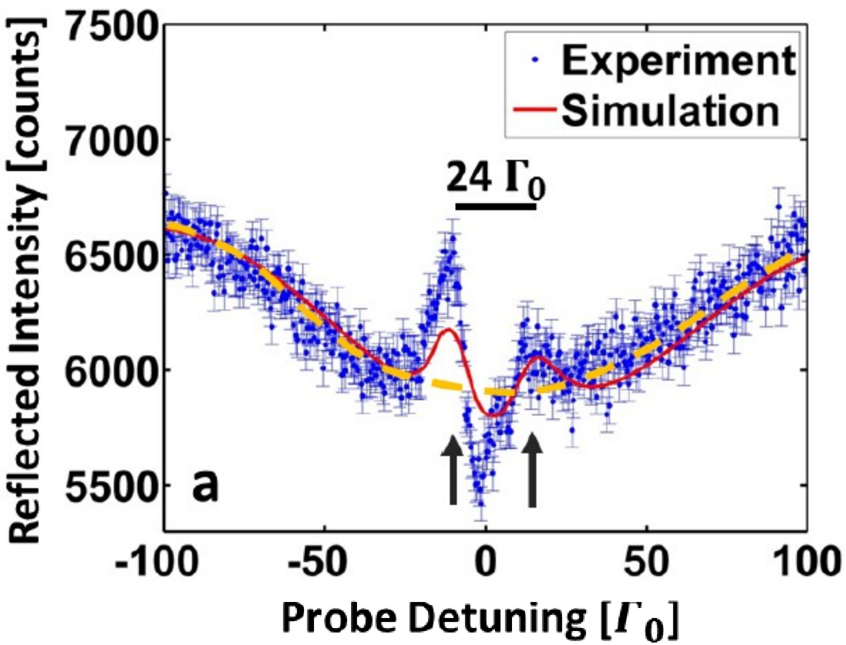

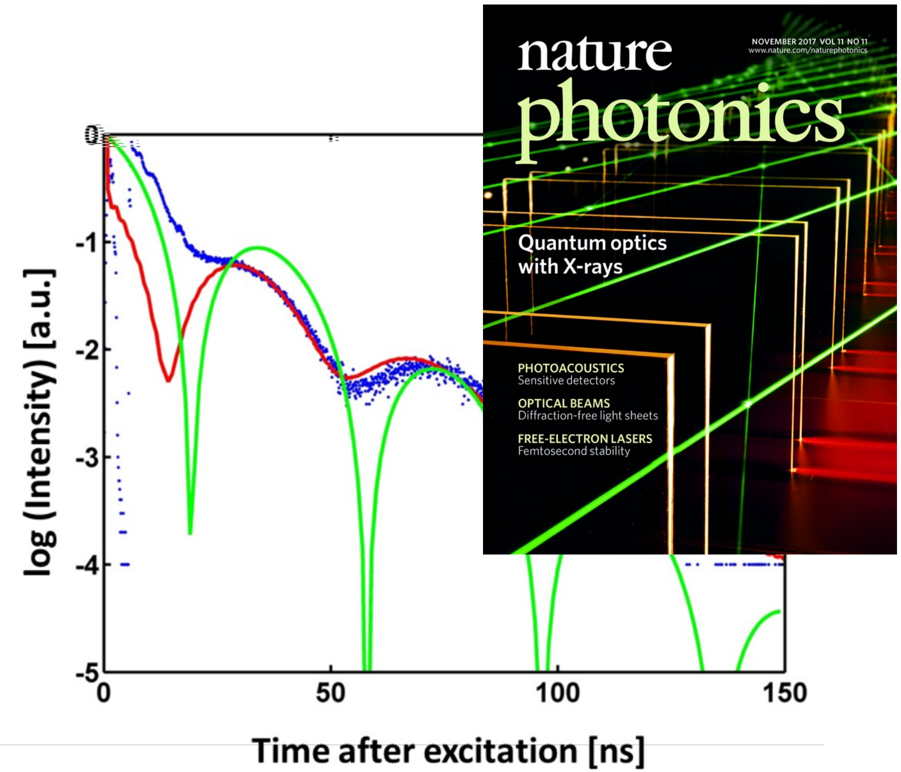

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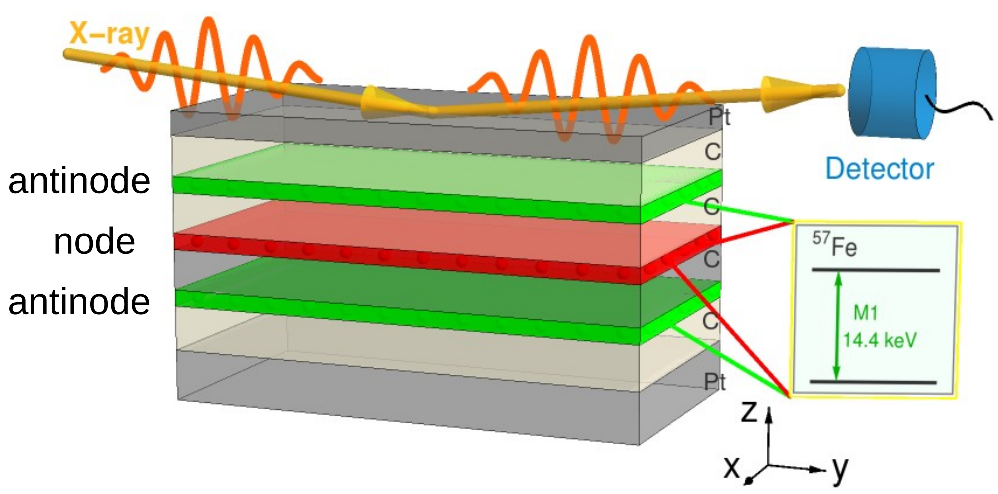
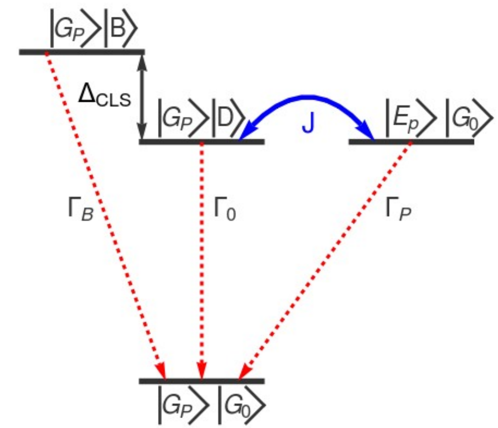
3. Dark states in many-layer structures

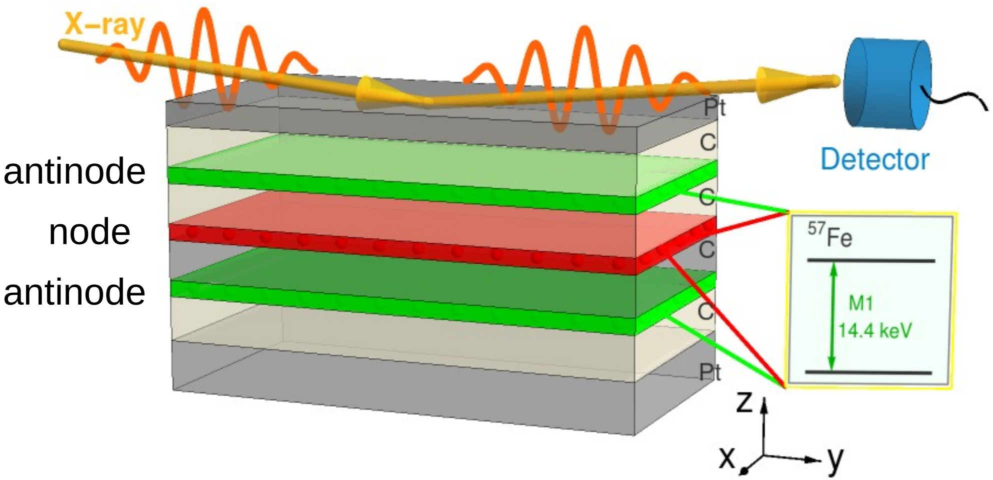




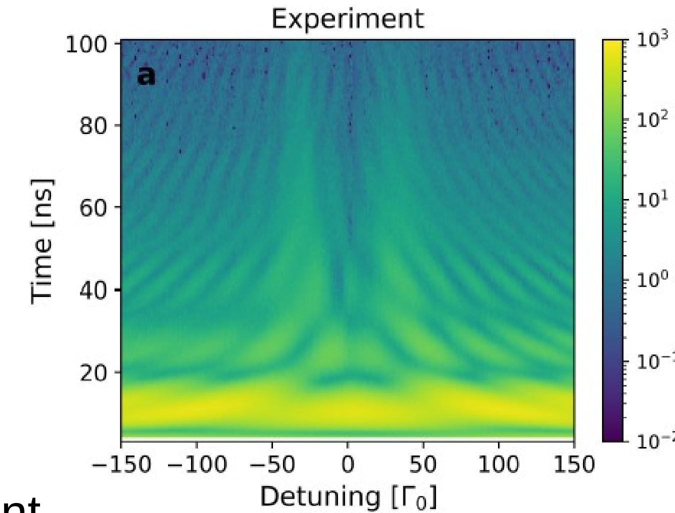
- Effective Jaynes-Cummings cavity QED Hamiltonian
- Strong-coupling regime via the dark-state field
- Effective photon-photon interaction, generation of non-classical states of light

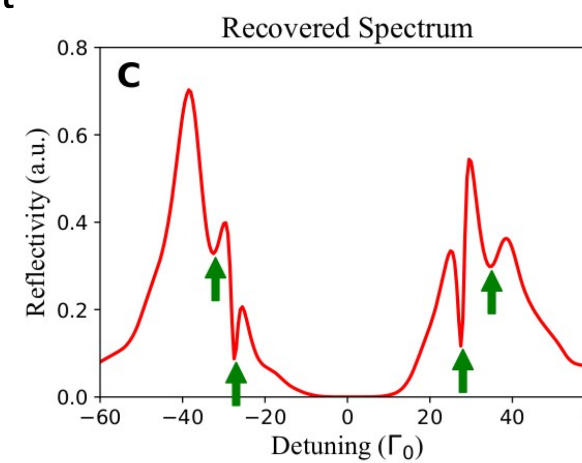
Preparation: Green's function formalism *X. Kong, D. Chang, AP, PRA **102**, 033710 (2020)*
*P. Andrejić, AP, PRA **104**, 033702 (2021)*

3. Dark states in many-layer structures



Spring-8
Experiment



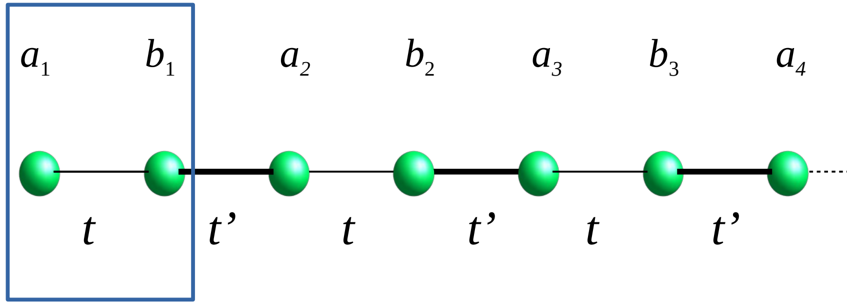


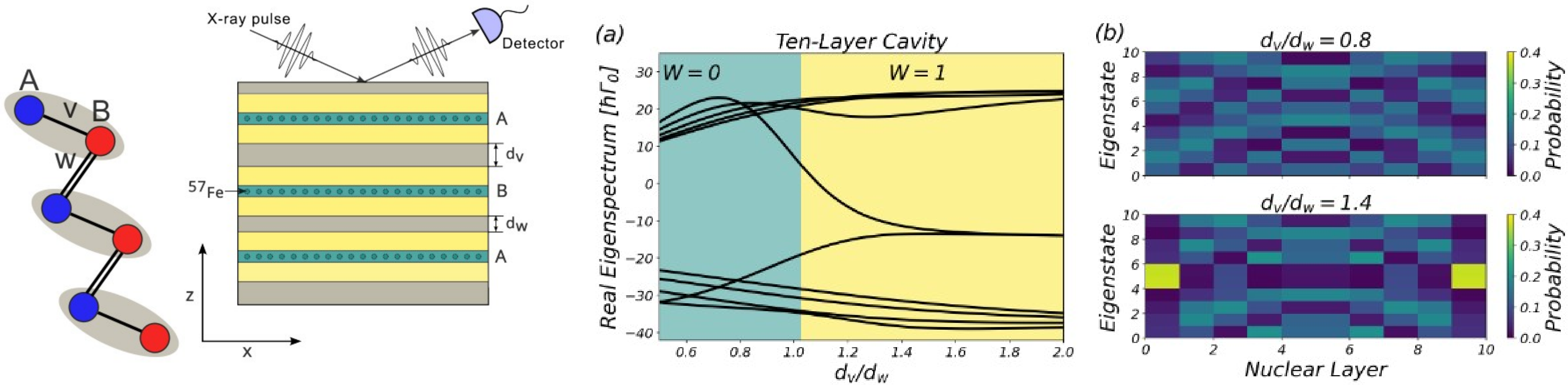
- Proof-of-principle for 3 layers
- Look for unequal couplings in generic 2N+1 layer systems

Preparation: Phase retrieval method *Z. Yuan, …, AP, X. Kong, Nature Commun.* ***16****, 3096 (2025)*

4. Topological effects in thin-film cavities

- 1D Su-Schrieffer-Heeger model
- Unequal couplings t and t’
- Excitation is “collected” at the lateral side of the sample – topological edge state





H. Zimmermann, J. Sturm. I. C. Fulga, J. van den Brink, AP, arXiv:2506.10588

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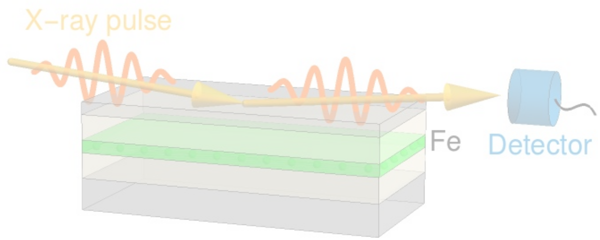

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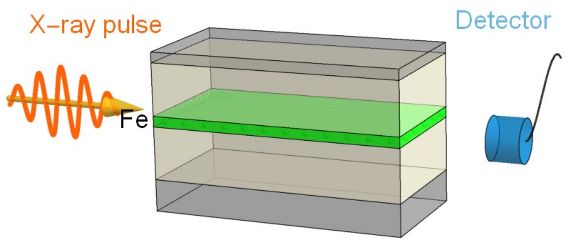
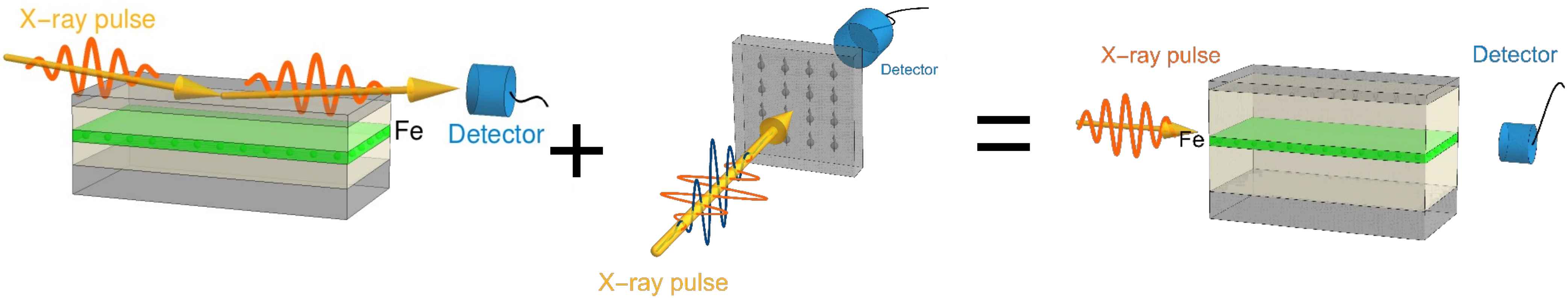
Dark states and topological effects

III. Applications in waveguide geometry (front coupling)



Front coupling theory and experiment

Waveguide geometry



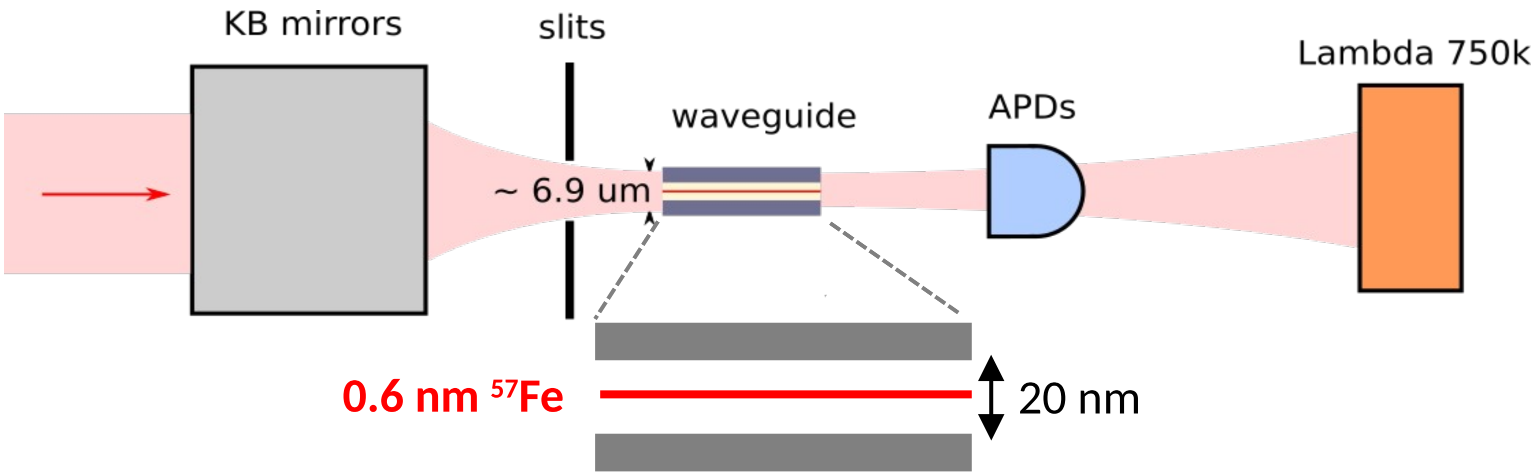
Front coupling

- Less explored regime: waveguide propagation in thin film nanolayers
- Same system as grazing incidence, but same boundary condition as forward scattering!

*P. Andrejić, L. Lohse, AP, Phys. Rev. A **109**, 063702 (2024)*

Front coupling experiment

Lohse, Andrejic, …, AP, Salditt, Röhlsberger
Phys. Rev. Lett. ***135****, 053601 (2025)*



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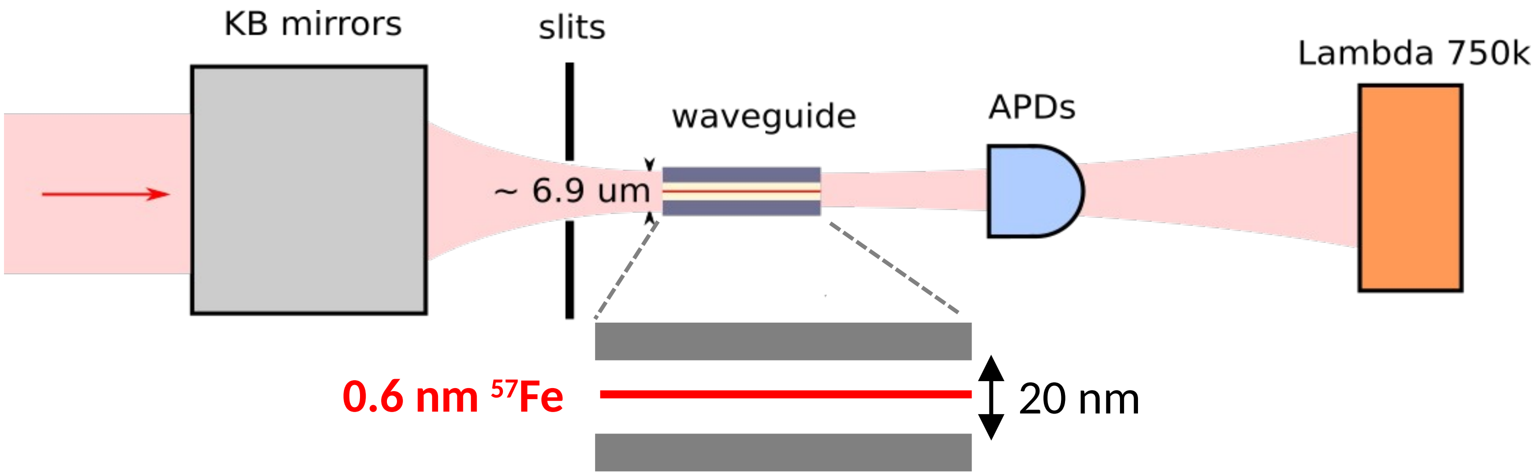
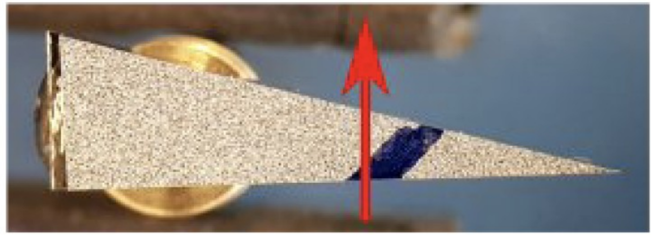


Shape of sample (top view)



Front coupling experiment

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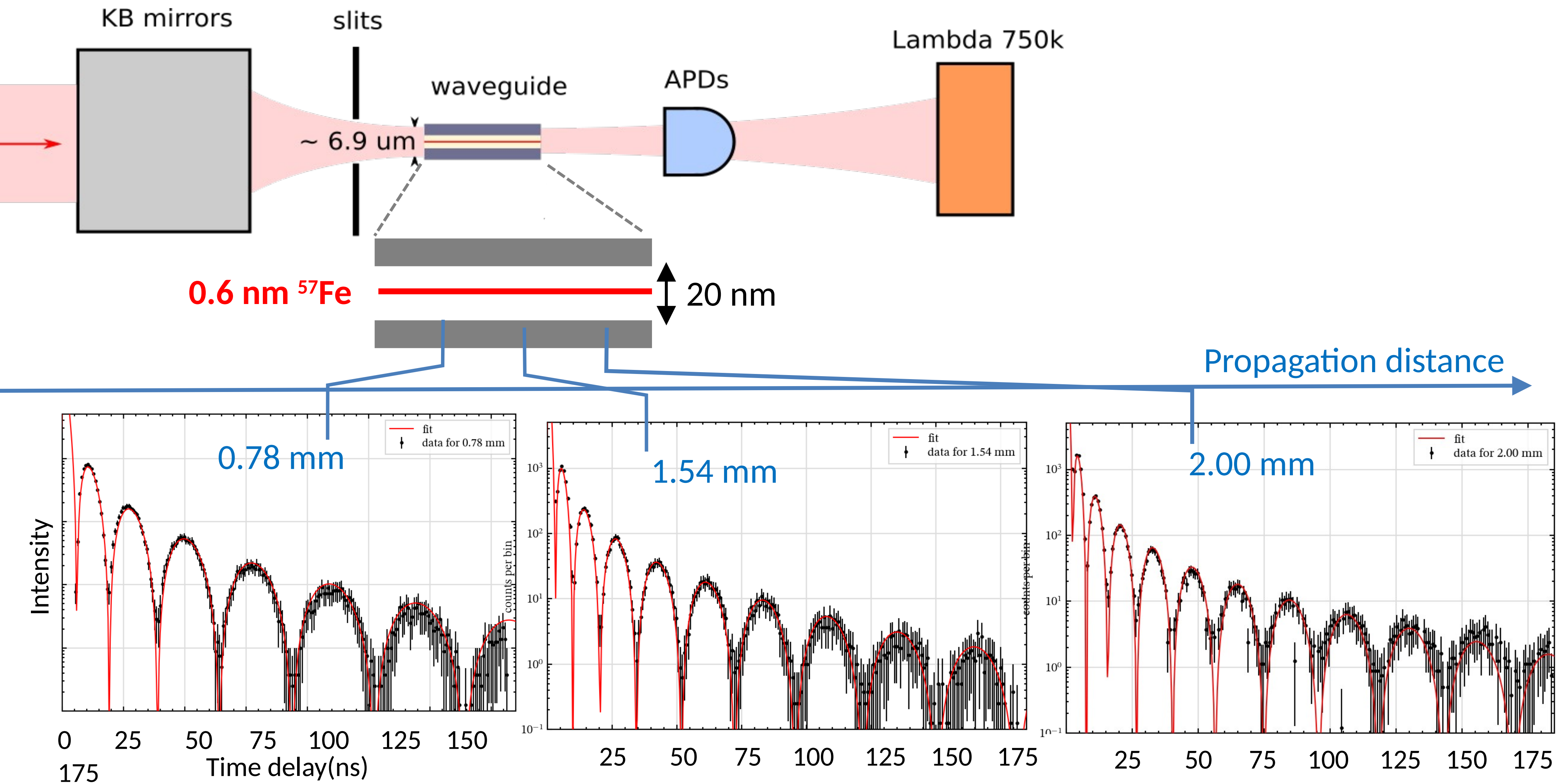

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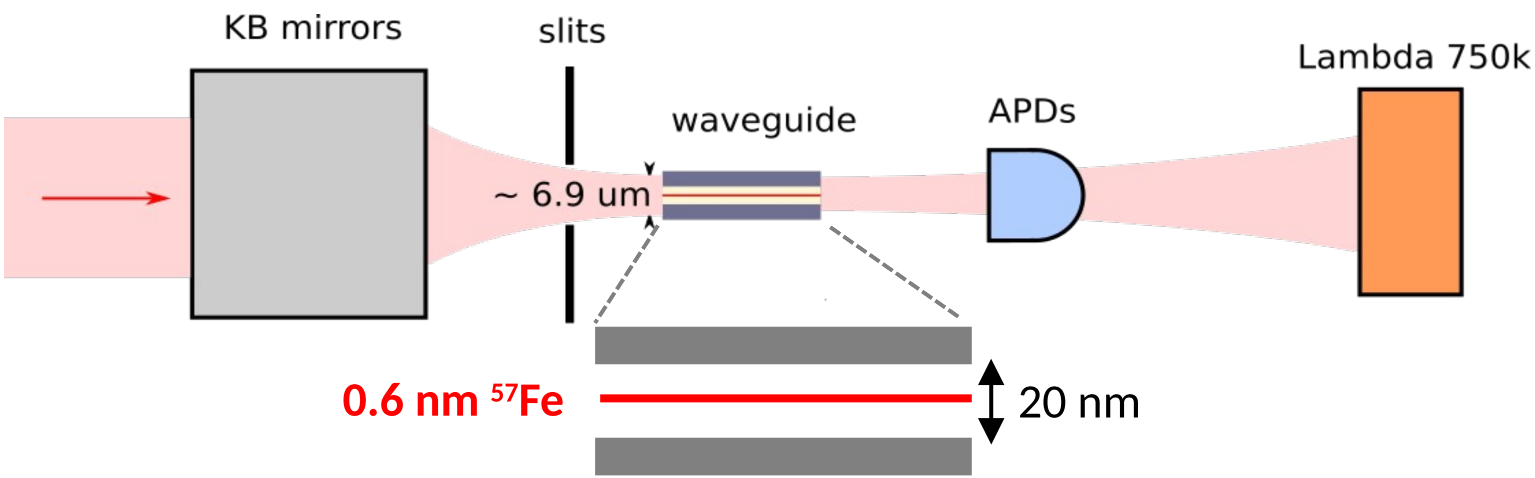
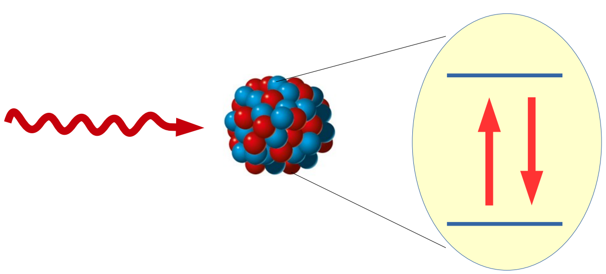


- Features of both grazing incidence and nuclear forward scattering
- Guided modes as in grazing incidence, each behaving as in nuclear forward scattering
- Guided modes can have very long attenuation lengths, up to mm scale!

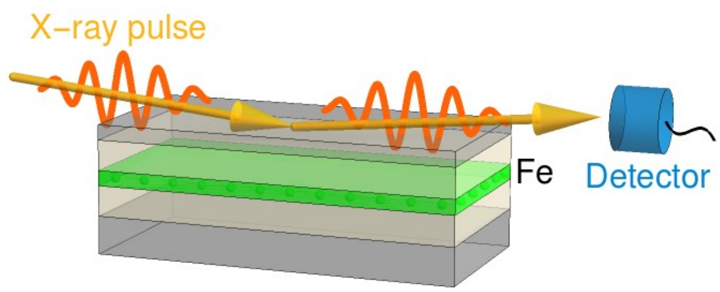
Conclusions

How to control of a single x-ray photon using Mössbauer nuclei:

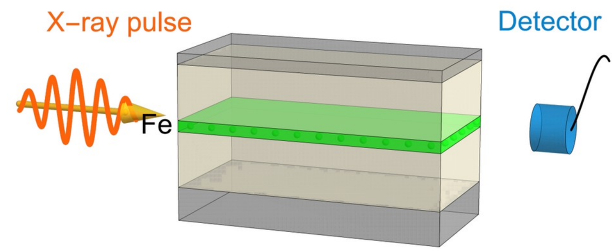
- Exploiting collective effects (superradiance) and external magnetic fields
- Cavity QED effects in grazing incidence geometry
- Long attenuation lengths in waveguide geometry
- Combine front coupling and topology for spatial propagation in edge states



clean quantum optical system



cavity geometry



waveguide geometry

Conclusions

How to control of a single x-ray photon using Mössbauer nuclei:

- Exploiting collective effects (superradiance) and external magr
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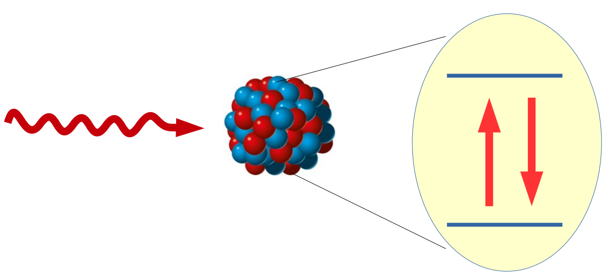
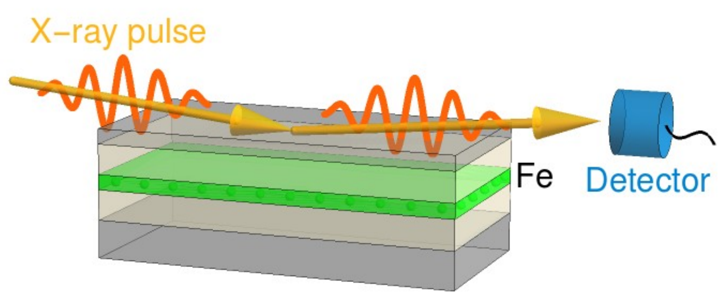
- Long attenuation lengths in waveguide geometry
- Combine front coupling and topology for spatial propagation in

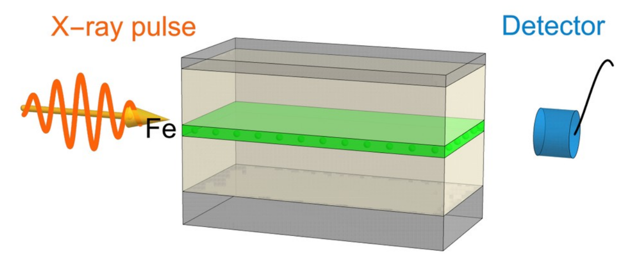
- Control propagation
- Switches
- Stop and release
- Phase manipulation



clean quantum optical system



cavity geometry



waveguide geometry

Thanks

Xiangjin Kong

Ralf Röhlsberger
Leon Lohse
Lars Bocklage
Sven Velten
Olaf Leupold

DFG

THANK YOU FOR YOUR ATTENTION!

Heisenberg-Programm
DFG Deutsche Forschungsgemeinschaft

TRR 306
QuCoLiMa
Quantum Cooperativity of Light and Matter

ct.qmat
Complexity and Topology in Quantum Matter

Thanks

FUDAN UNIVERSITY 1905

Xiangjin Kong

DESY.

Ralf Röhlsberger
Leon Lohse
Lars Bocklage
Sven Velten
Olaf Leupold

Jonathan Sturm

Fabian Richter

Petar Andrejić

Hanns Zimmermann

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QuCoLiMa

Quantum Cooperativity of Light and Matter

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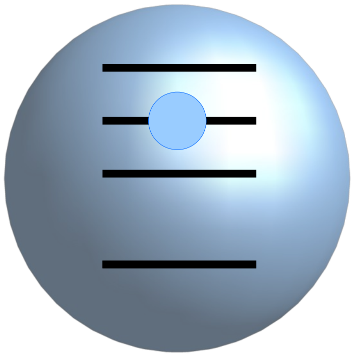
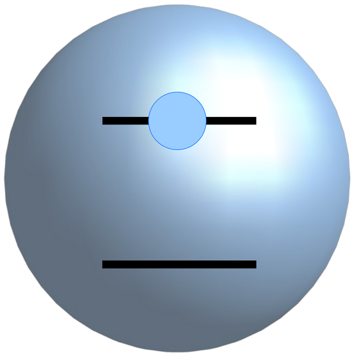
Complexity and Topology in Quantum Matter

Spontaneous decay of single atom



- Classical field description can't explain spontaneous decay
- Quantized field description introduces the vacuum – infinite empty modes
- Interaction with vacuum modes explains spontaneous decay
- Many identical atoms interacting with the same vacuum?

Spontaneous decay of single atom



2-level system

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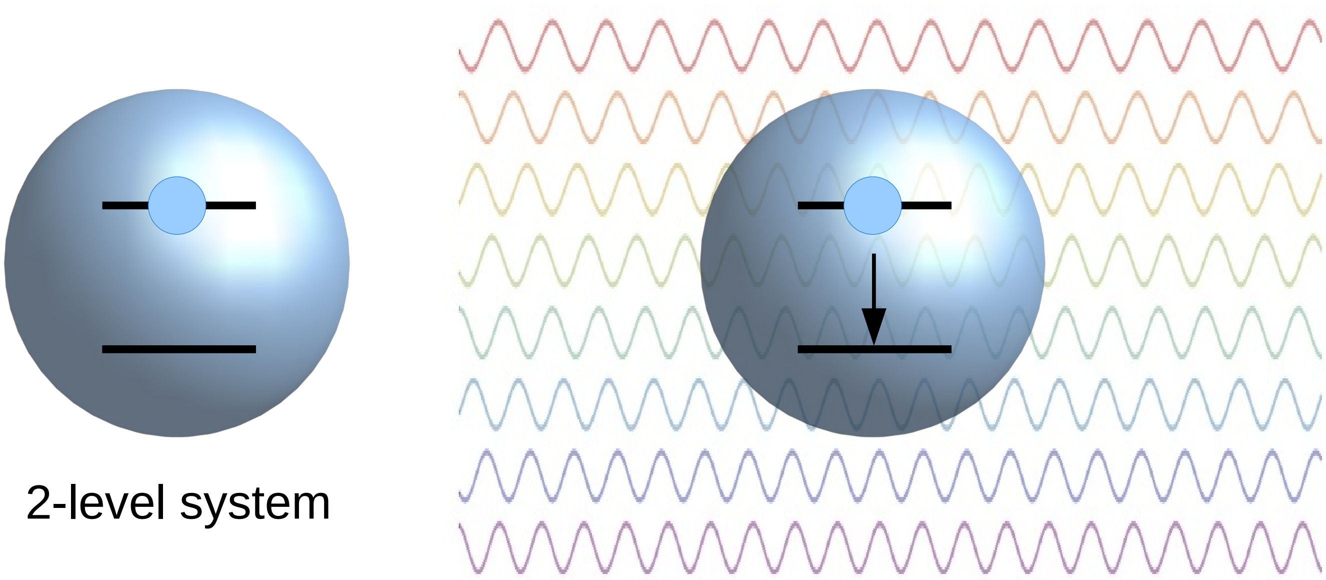

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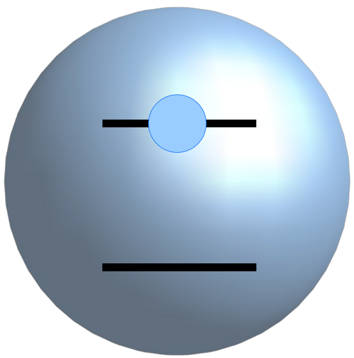
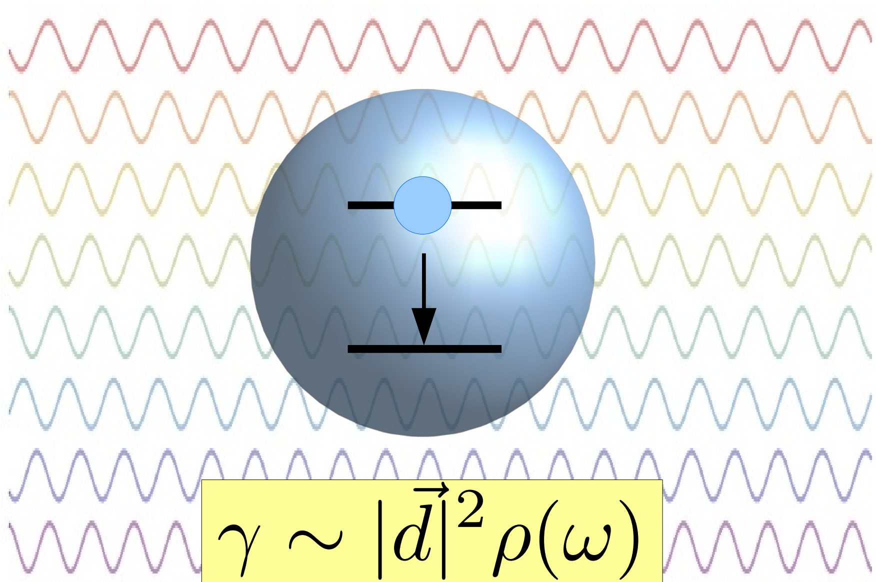


2-level system



$$\gamma \sim |\vec{d}|^2 \rho(\omega)$$

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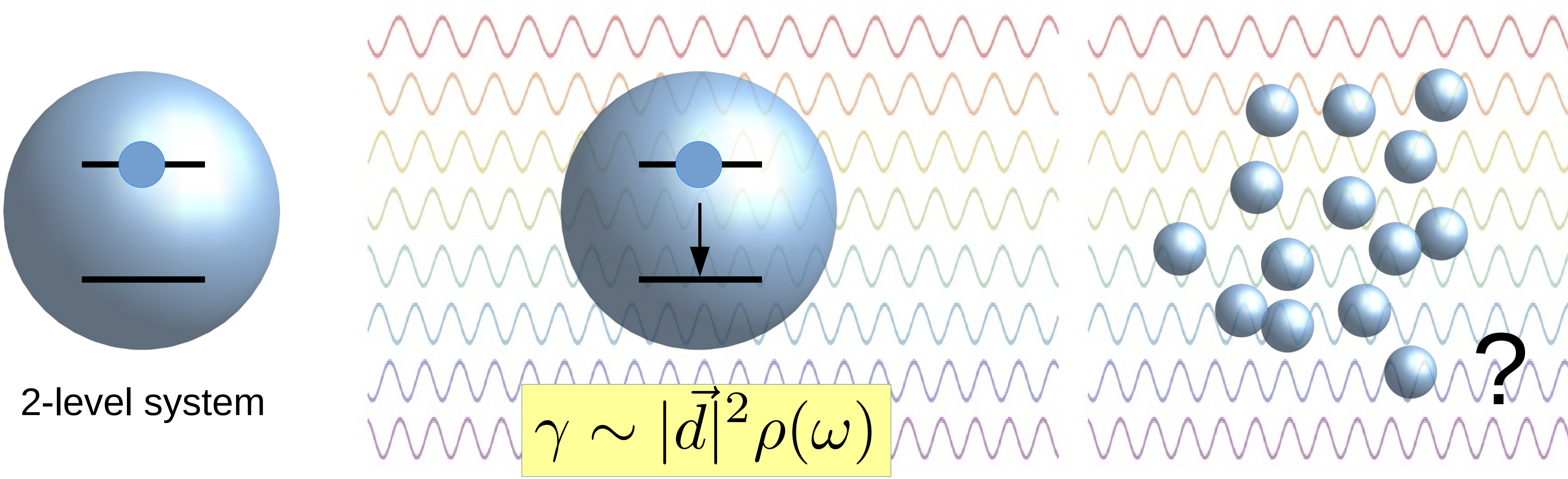

$$\gamma \sim |\vec{d}|^2 \rho(\omega)$$

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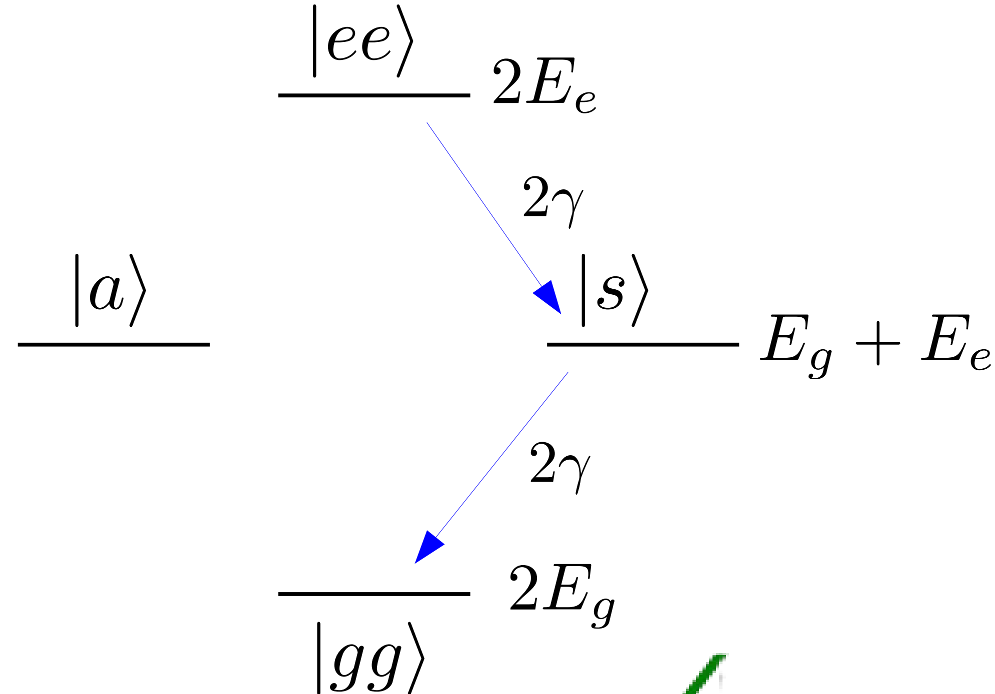
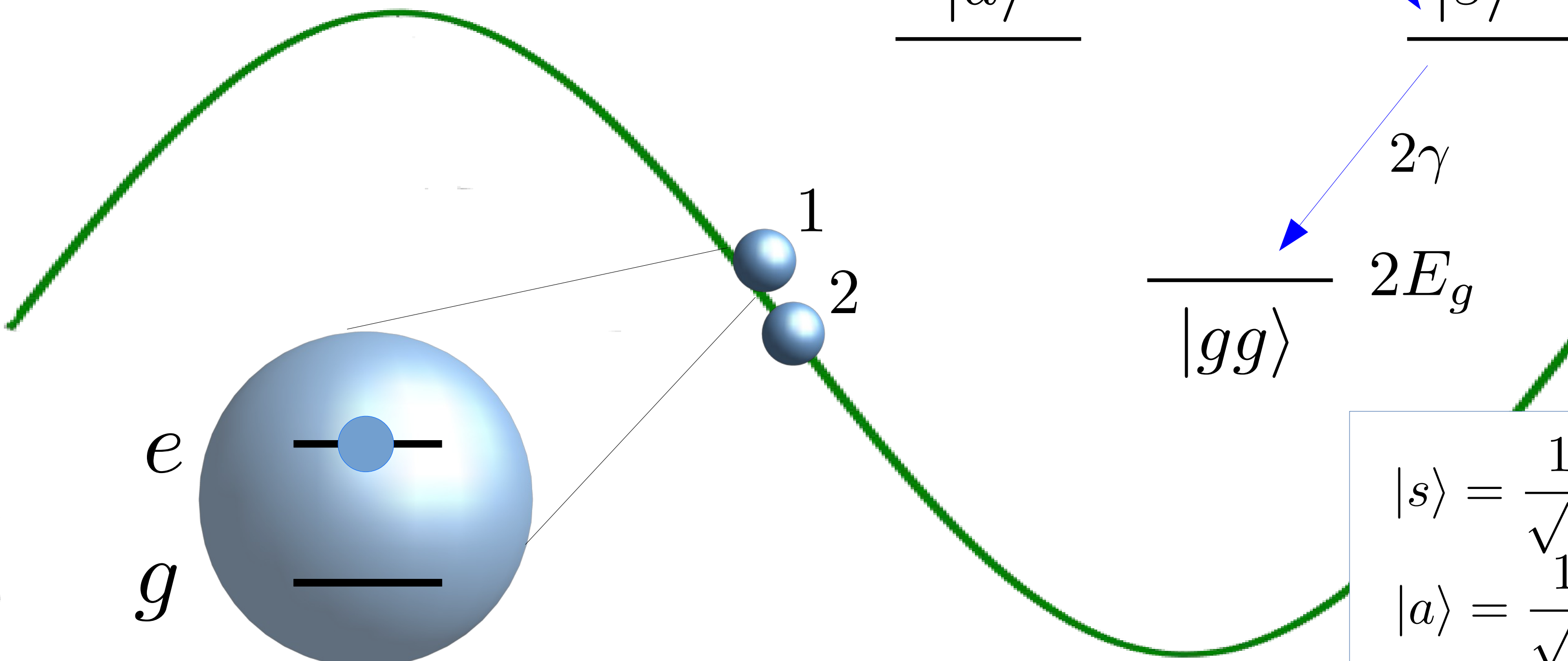
- Quantized field description introduces the vacuum – infinite empty modes
- Interaction with vacuum modes explains spontaneous decay
- Many identical atoms interacting with the same vacuum?

Example

2 non-interacting 2-level atoms



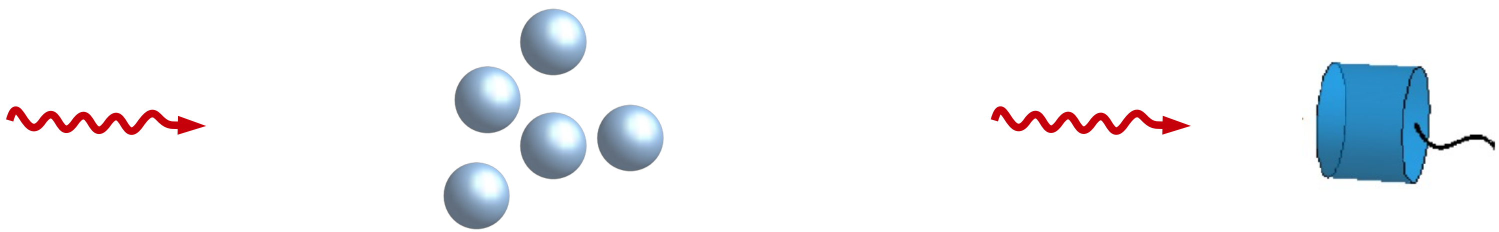


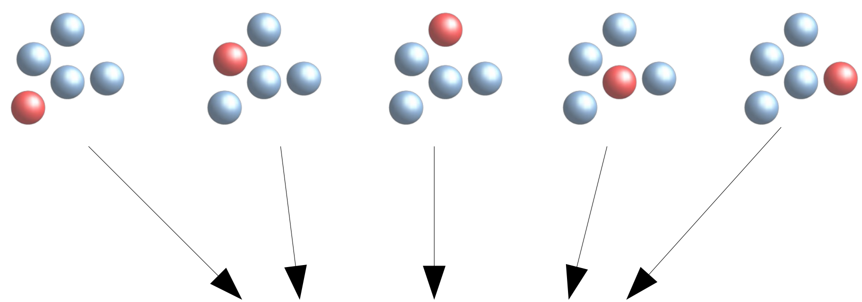
$$|s\rangle = \frac{1}{\sqrt{2}}(|g_1 e_2\rangle + |g_2 e_1\rangle)$$

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

Total dipole moment operator $\hat{\vec{d}} = \hat{\vec{d}}_1 + \hat{\vec{d}}_2$ invariant to transposition of atoms!

Exciting the ensemble





Ground state $|g_1, g_2, g_3, g_4, g_5\rangle$

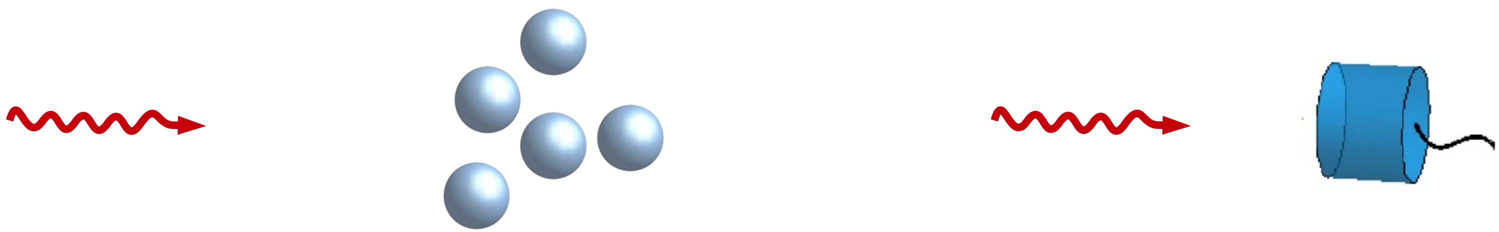
Dicke state

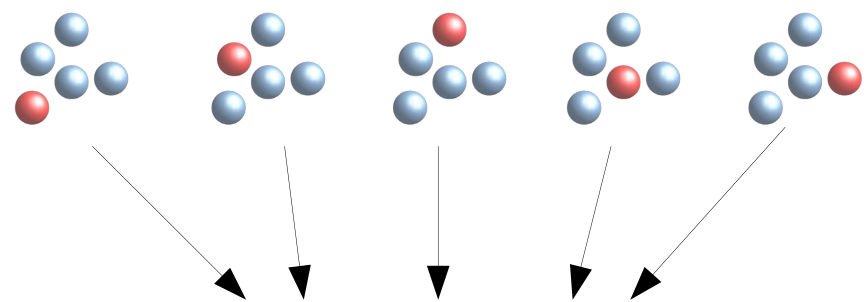
$$|\Psi\rangle = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} |g_1, g_2, \ldots, e_i, \ldots g_N\rangle$$

$\lambda \gg d$

N times faster decay!

Exciting the ensemble





Ground state $|g_1, g_2, g_3, g_4, g_5\rangle$

"timed" Dicke state

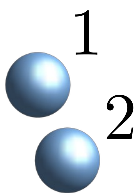
$$|\Psi\rangle = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} e^{i\vec{k}\cdot\vec{r}_i} |g_1, g_2, \ldots, e_i, \ldots g_N\rangle$$

$\lambda \ll d$

Extended sample, ???

Radiative eigenmodes

Can we have superradiance for extended samples?



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

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

If the laser excites the radiative eigenmode, yes - geometry and couplings

Temporal dynamics of radiative eigenmode:

$$\dot{\beta}_0 = -(\gamma + \Gamma_N + i\mathcal{L}_N)\beta_0$$

- Γ_N → superradiant decay rate
- $\mathcal{L}_N$ → "Collective Lamb shift"
- β_0 → probability amplitude

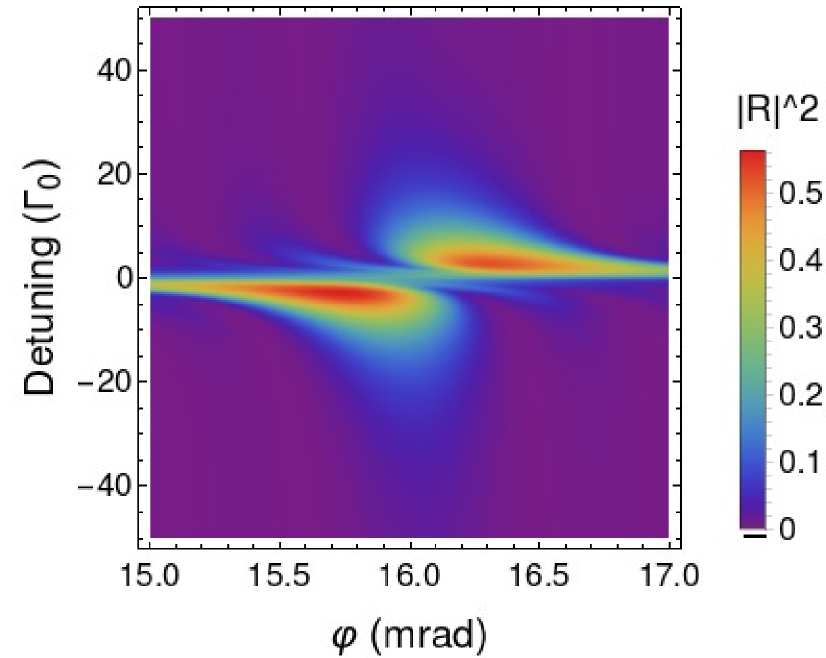
M. O. Scully, Phys. Rev. Lett. 102, 143601 (2009)

Green function formalism

- can be used for any multilayer structure
- fully quantum - quantized electromagnetic field
- Heisenberg eqs., input-output formalism
- Grazing incidence and front coupling

X. Kong, D. Chang, AP, PRA 102, 033710 (2020)
P. Andrejić, AP, PRA 104, 033702 (2021)

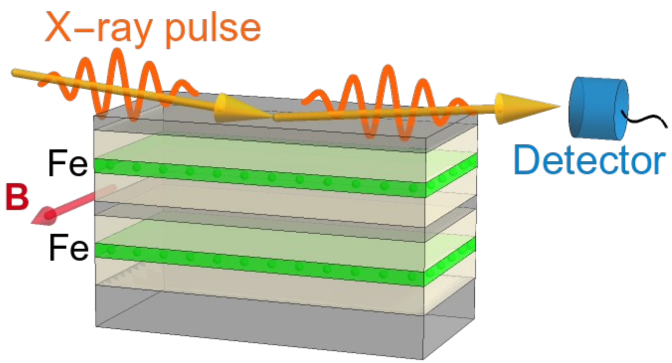
30-layer cavity



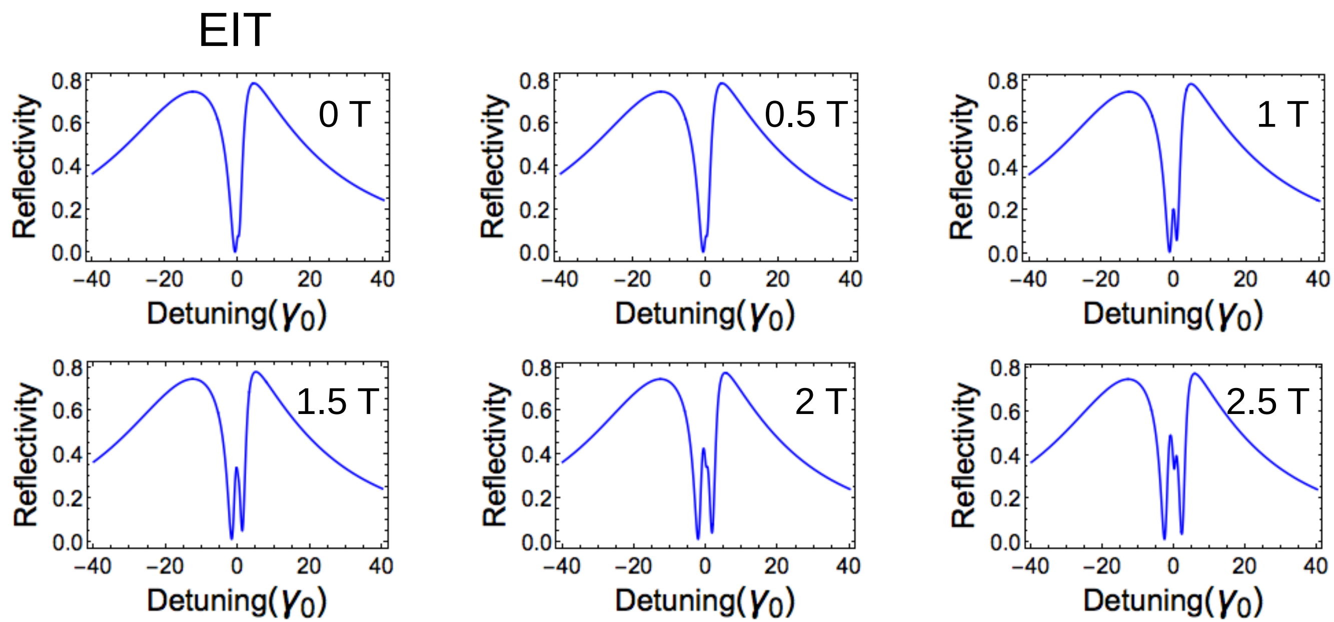
$$J_{lm} = \sqrt{N_l N_m}\left(\mu_0\omega_p^2/\hbar\right)\mathbf{d}^*\cdot\mathrm{Re}\left[\mathbf{G}_{1\mathrm{D}}\left(z_l, z_m, \omega_p\right)\right]\cdot\mathbf{d}$$ Spin-exchange - H

$$\Gamma_{lm} = \sqrt{N_l N_m}\left(2\mu_0\omega_p^2/\hbar\right)\mathbf{d}^*\cdot\mathrm{Im}\left[\mathbf{G}_{1\mathrm{D}}\left(z_l, z_m, \omega_p\right)\right]\cdot\mathbf{d}$$ Decay rate - $\mathcal{L}$

Multilevels and multilayers



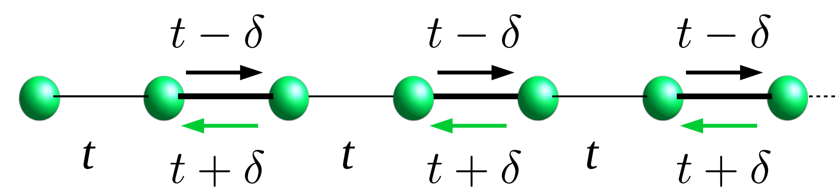
semiclassical results

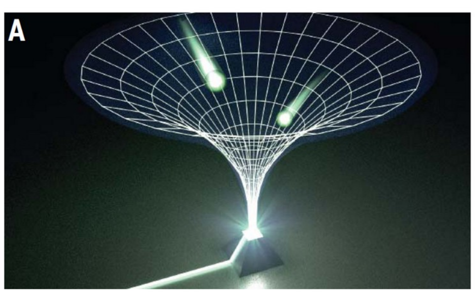


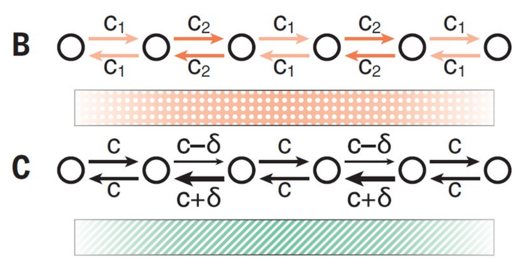
- Interplay between PT-symmetry, inter-layer coupling and superradiance
- What explanation have the occurring features? EP?
- Engineer desired response based on precise layer positioning

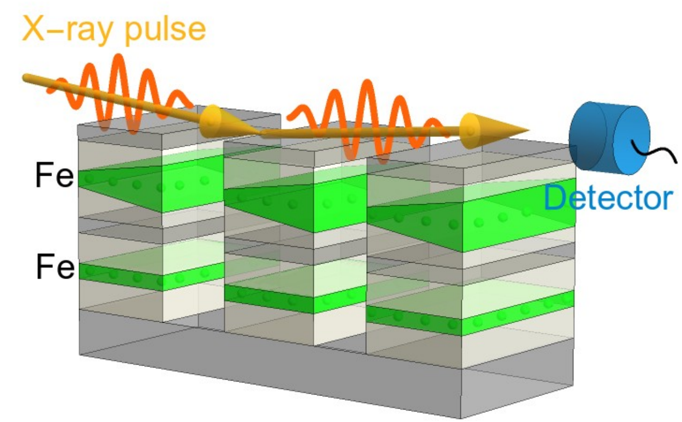
Non-Hermitian skin effect

- 1D Su-Schrieffer-Heeger model with anisotropic coupling
- Unequal coupings t, t+δ and t-δ
- Topological funneling of x-ray light





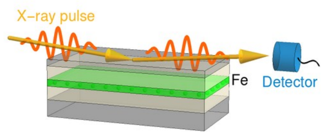
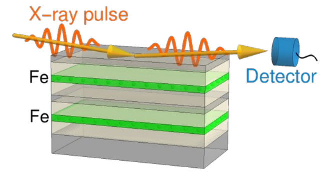
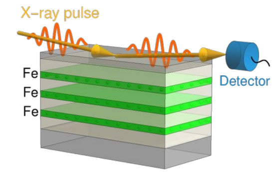
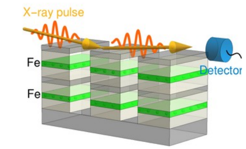
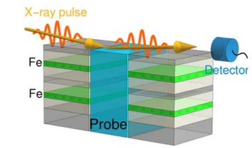




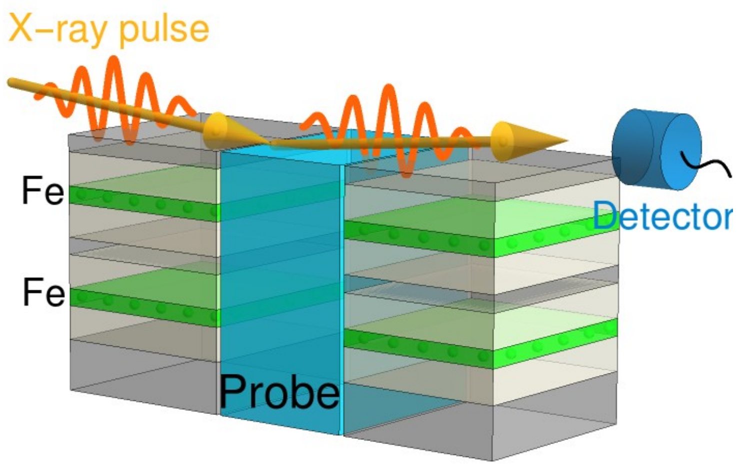
or inhomogeneous B field?

Weidemann, …, Thomale, Szameit, Science 368, 311 (2020)

Overview of nanostructure zoo

Structure					
$B = 0$	Superradiance	EIT/ Autler-Townes	Dark state nonlinearity	SSH topology	Probing technique
$B \neq 0$	PT symmetry	interplay?	??	??	

New probing techniques



- Microscopical probe embedded in cavity
- Taylor field via inter-layer coupling
- Novel technique for quasielastic x-ray scattering
- monitoring protein or spin dynamics

Combine with HAXPES?
Pump-probe type of experiment using x-ray and photoelectron response?

Ideas for ongoing/future work

- Robust x-ray transport and propagation control based on PT symmetry concepts
- Can we implement true gain in more complex structures?
- Quantum approach to reconcile PT-symmetry and reservoir engineering concepts from quantum optics
- X-ray topological photonics
- Exploit non-linearities for non-classical states of x-ray light
- Structure design to harness superradiance, layer coupling and PT symmetry interplay
- Development of new material probing techniques with x-rays and electrons?

1. Non-Hermitian photonics

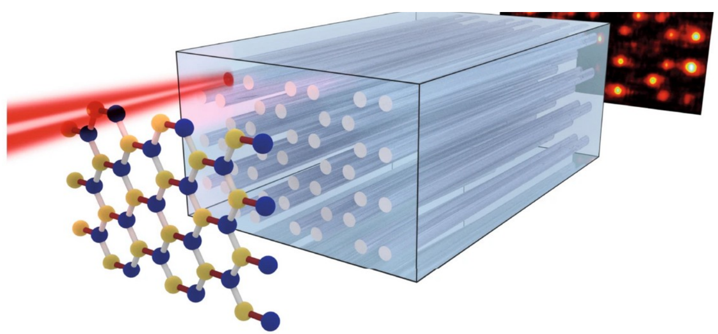
Schrödinger equation

Non-Hermitian Hamiltonian
PT symmetry
Real eigenstates

Electromagnetic wave propagation

Complex optical potential ε/μ
PT symmetry
Balanced gain - loss profile

- Extends photonics to the entire 4D space of complex ε / μ
- New functionalities: lasing, beam propagation, secondary emission, topological features



First 2D PT-symmetric crystal

Kremer, et al.
Nature Commun. 10, 435 (2019)

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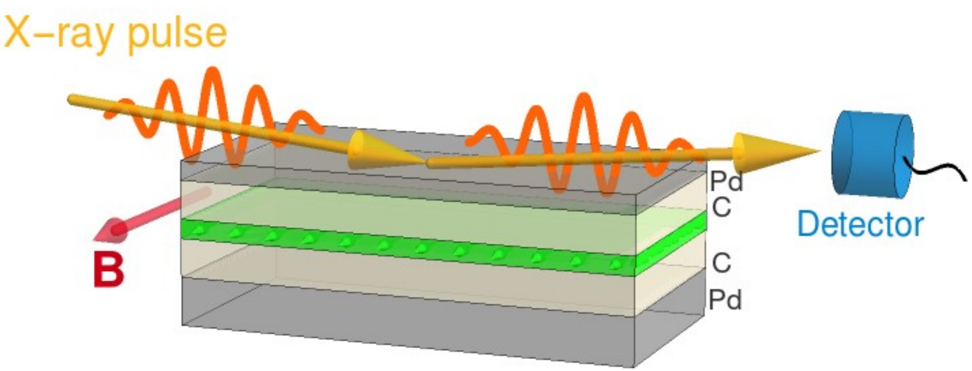
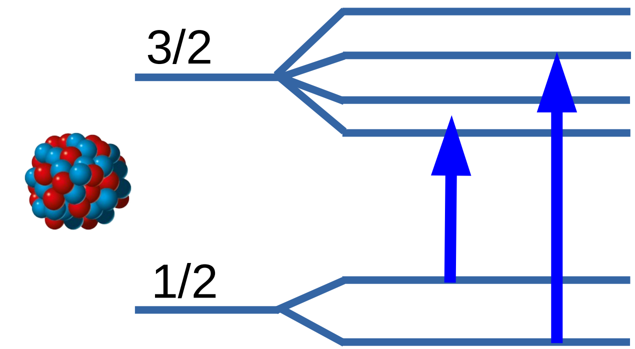
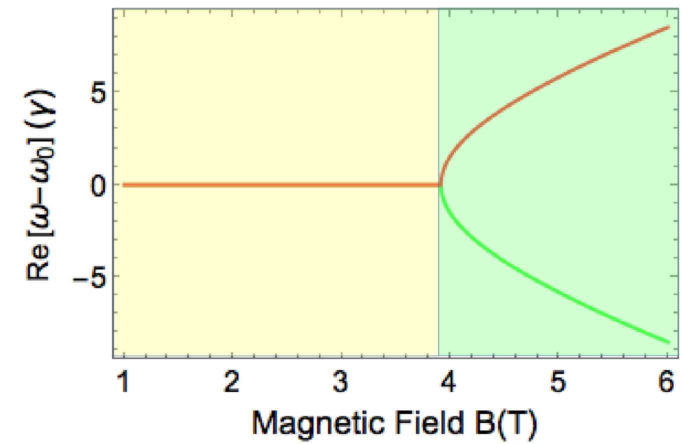
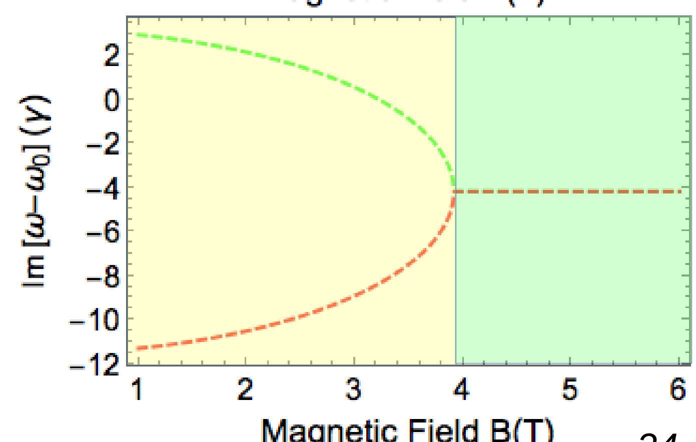
Complexity and Topology in Quantum Matter

PT-symmetry with x-rays



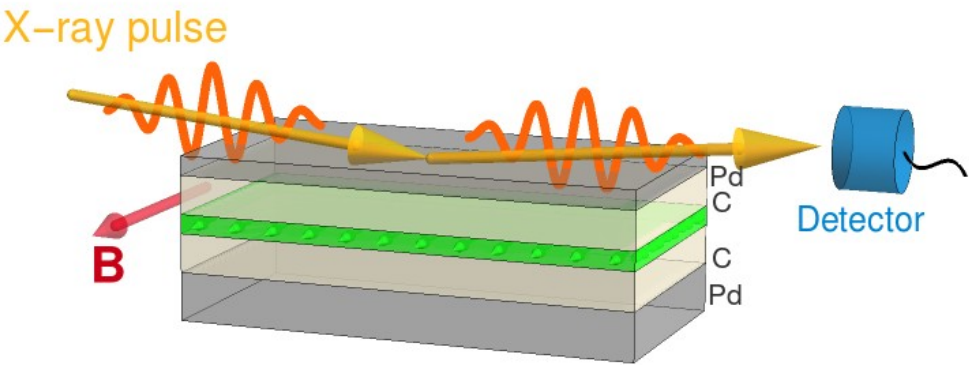


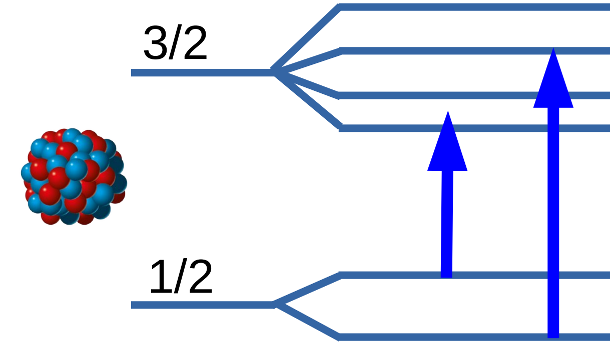




- Thin-film cavity displays PT-symmetry or PT-broken symmetry according to B value
- At exceptional point system very sensitive to external control
- Spatial interface can control x-ray propagation!

PT-symmetry with x-rays





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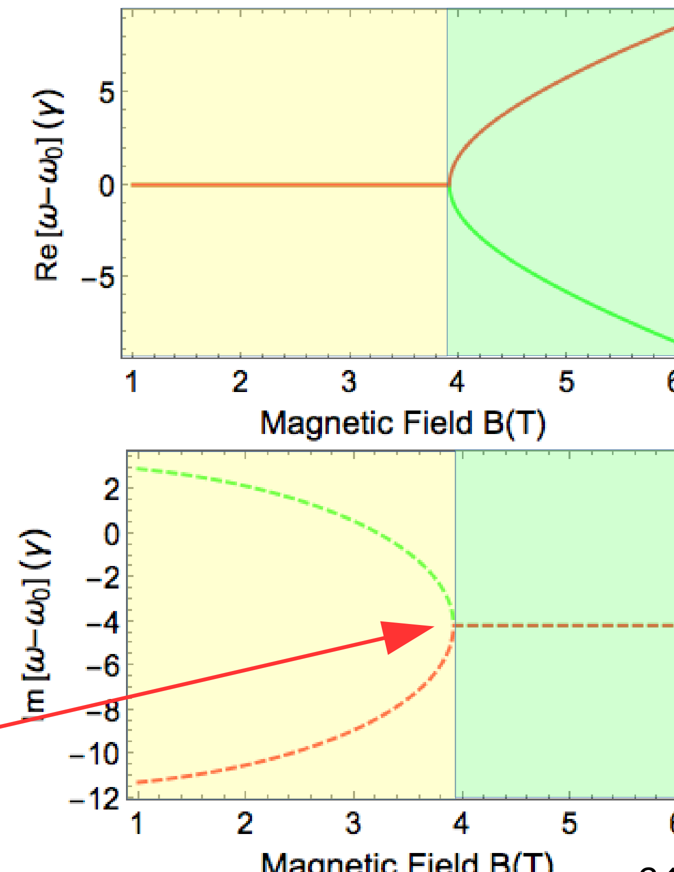
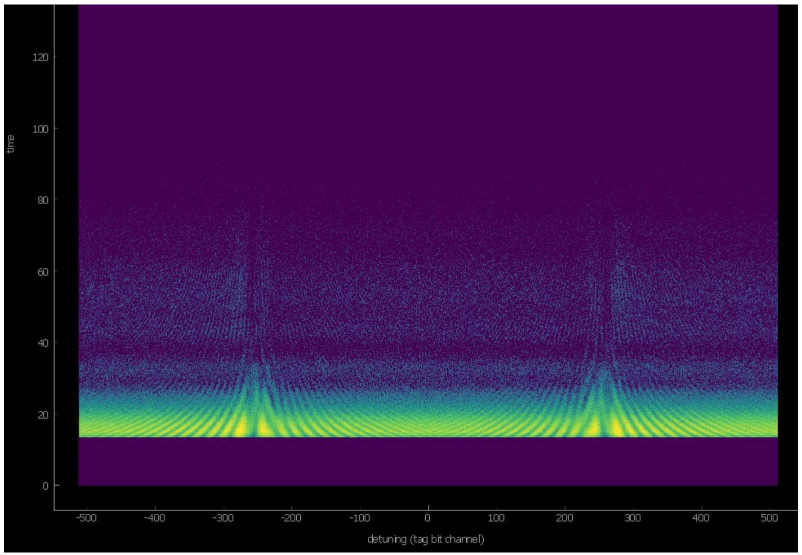


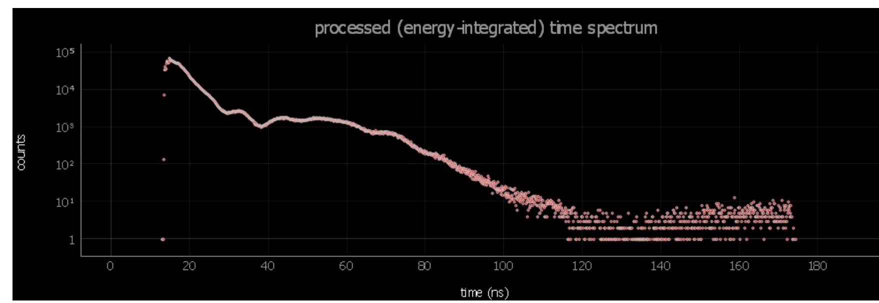
"Quasi-PT"
overall loss

Proof of principle

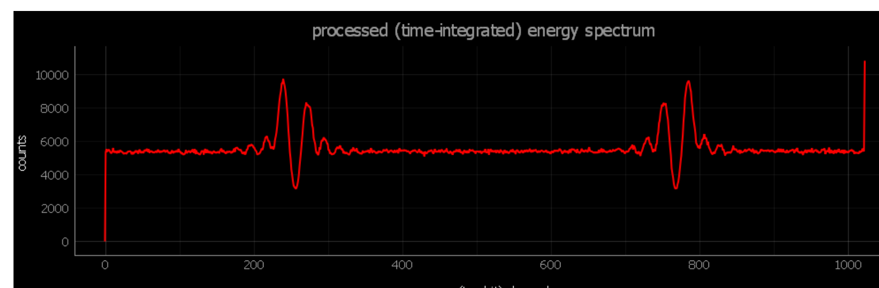
Experimental data on ^{119}Sn (November 2020) at PETRA III (P01)



- Extract the eigenvalues from energy or time spectra
- Proof existence of exceptional point

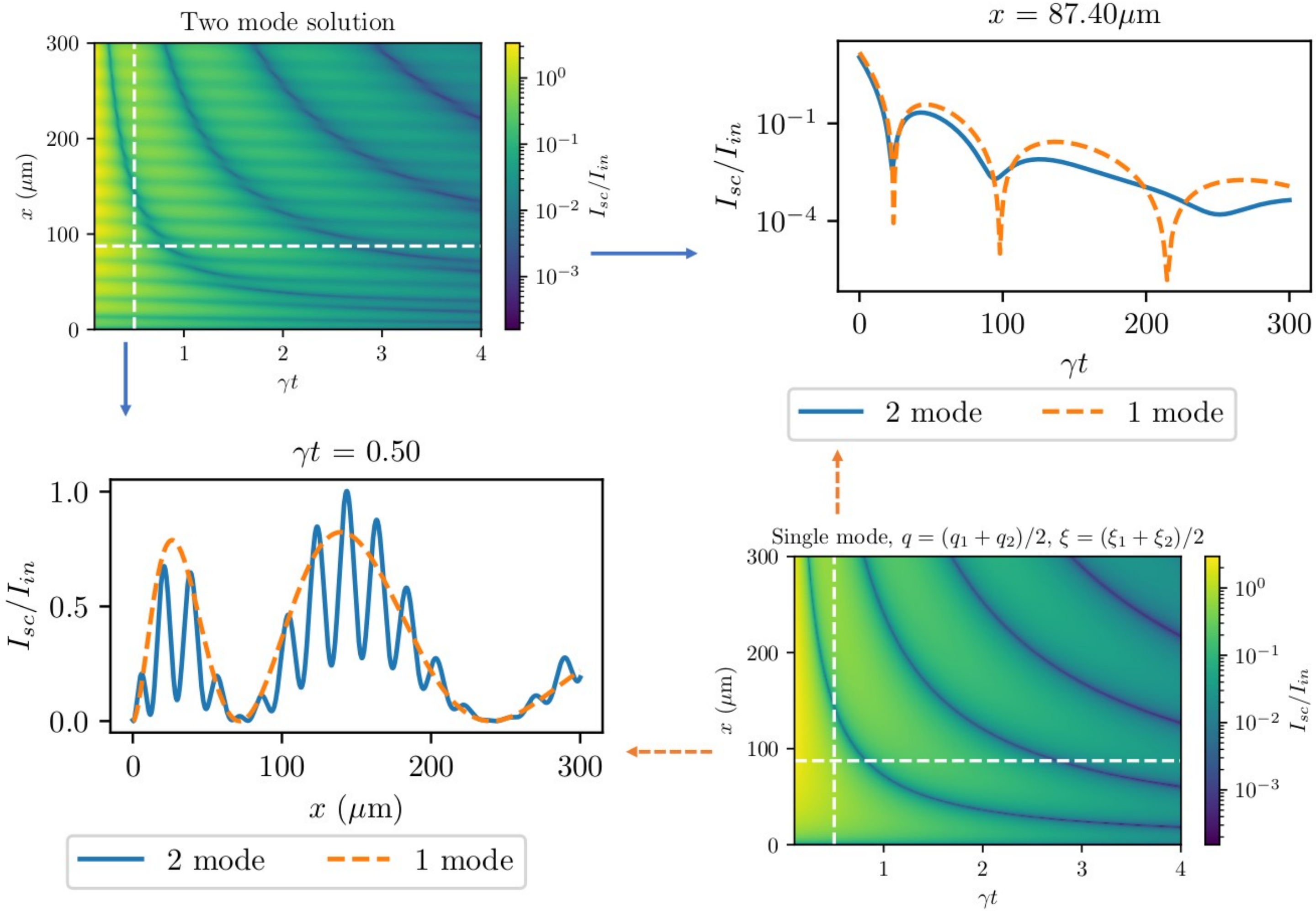


2D-spectra (time, energy) for several B values



evaluating data happily ever after
Recovery of energy spectra – phase retrieval
Z. Yuan et al., arXiv: 2204.06096

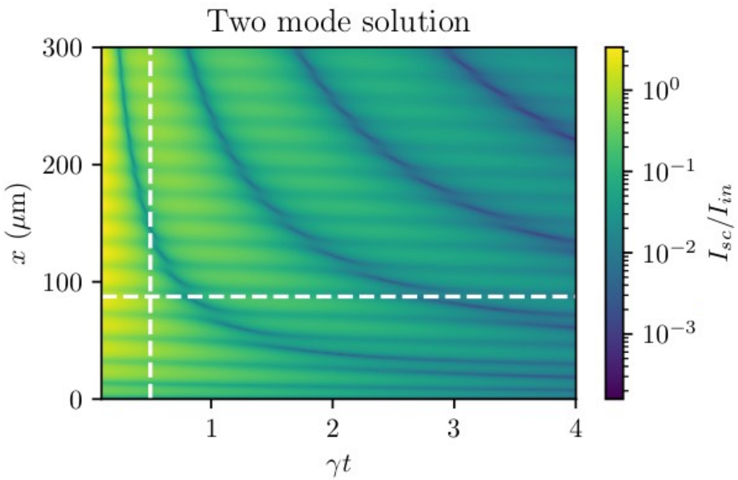
Two-mode interference

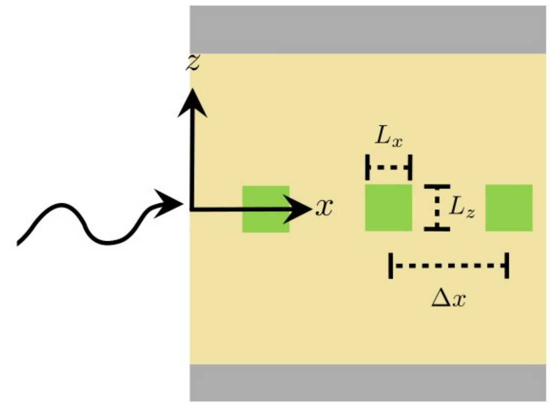


P. Andrejić, L. Lohse, AP, arXiv: 2305.11647 (2023)

Two-mode interference

Combine spatial interference pattern with micropatterning





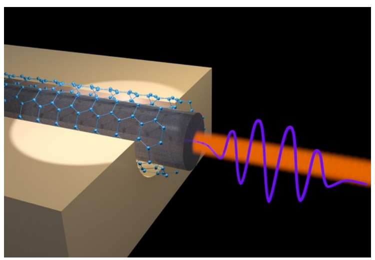
Sub-ensemble of even strips does not interact with sub-ensemble of odd strips!
Search for non-trivial topology?

P. Andrejić, L. Lohse, AP, arXiv: 2305.11647 (2023)

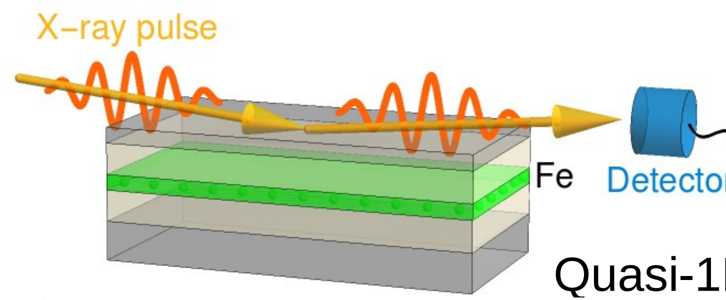
X-ray photonic devices for …

- Robust x-ray transport and propagation control based on PT symmetry concepts
- Nanoscale x-ray lasing for space- and energy-resolved probing of matter
- Miniaturized x-ray material probing

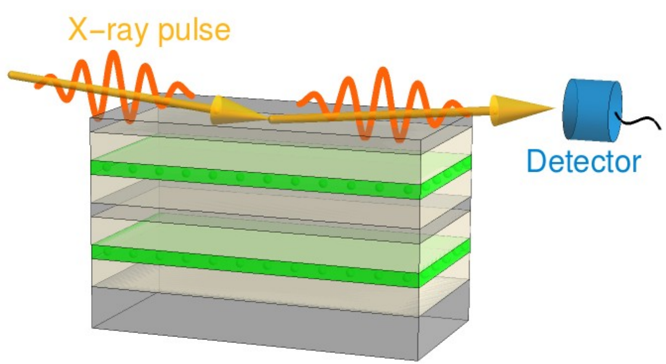
TP 1
TP 4
EP 4
EP 5



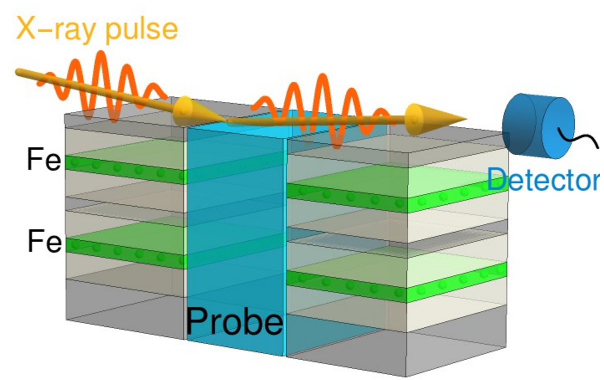
1D



Quasi-1D and 2D



More complicated layer structures



Develop new probing technique