X-ray nuclear quantum optics I

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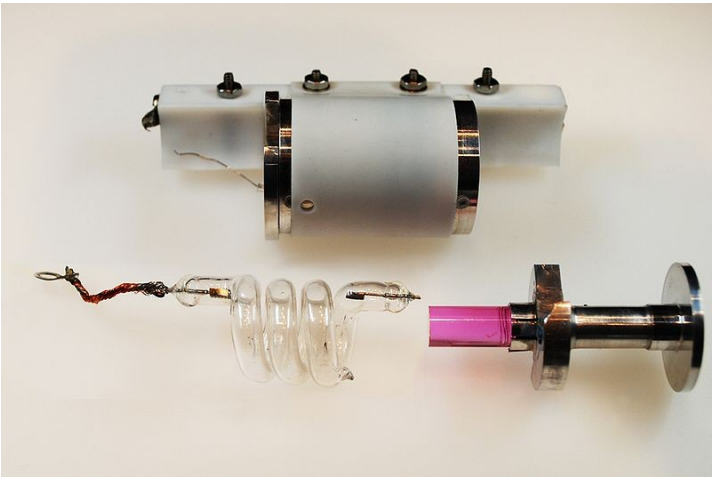
Hamburg, 6-7 October 2014

Historical perspective

1960, T. Maiman, first successful laser



Lasers mostly OPTICAL, based on and used to control ATOMS

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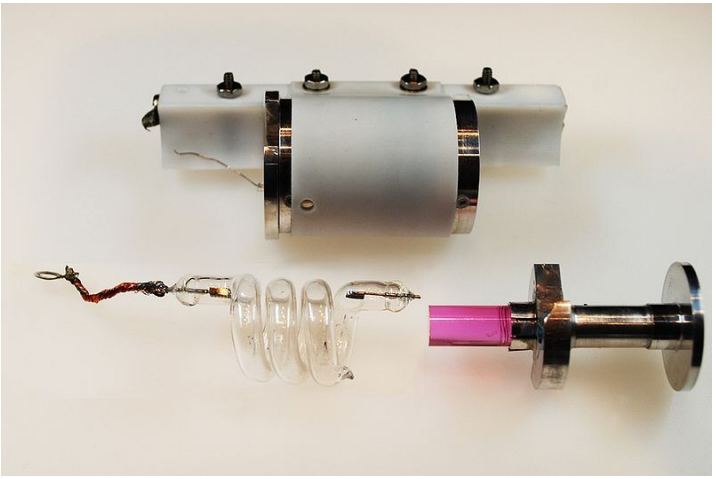
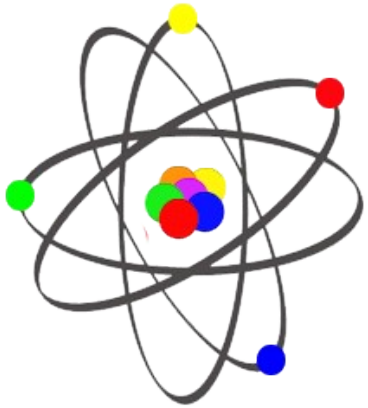
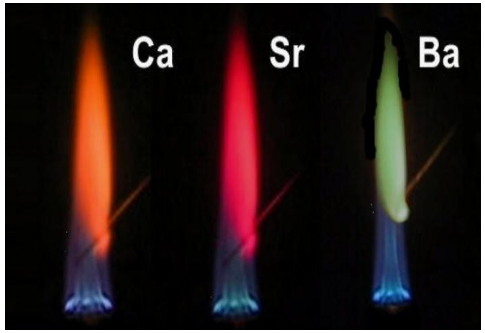




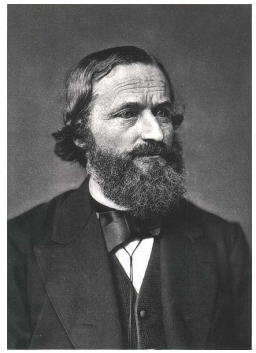
Lasers mostly OPTICAL, based on and used to control ATOMS

Atomic physics and quantum optics

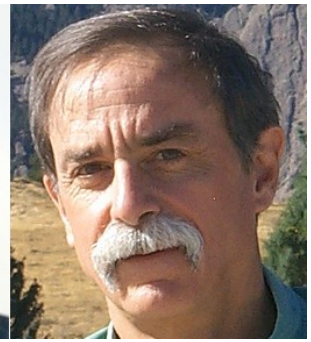


"uncontrolled pump
and passive observation"

1859

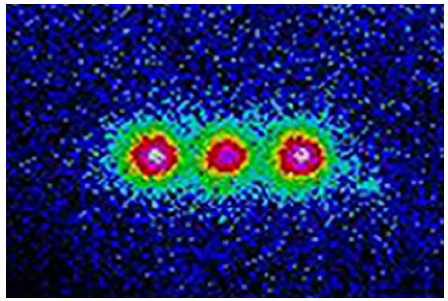


Bunsen Kirchhoff



Haroche Wineland

2012

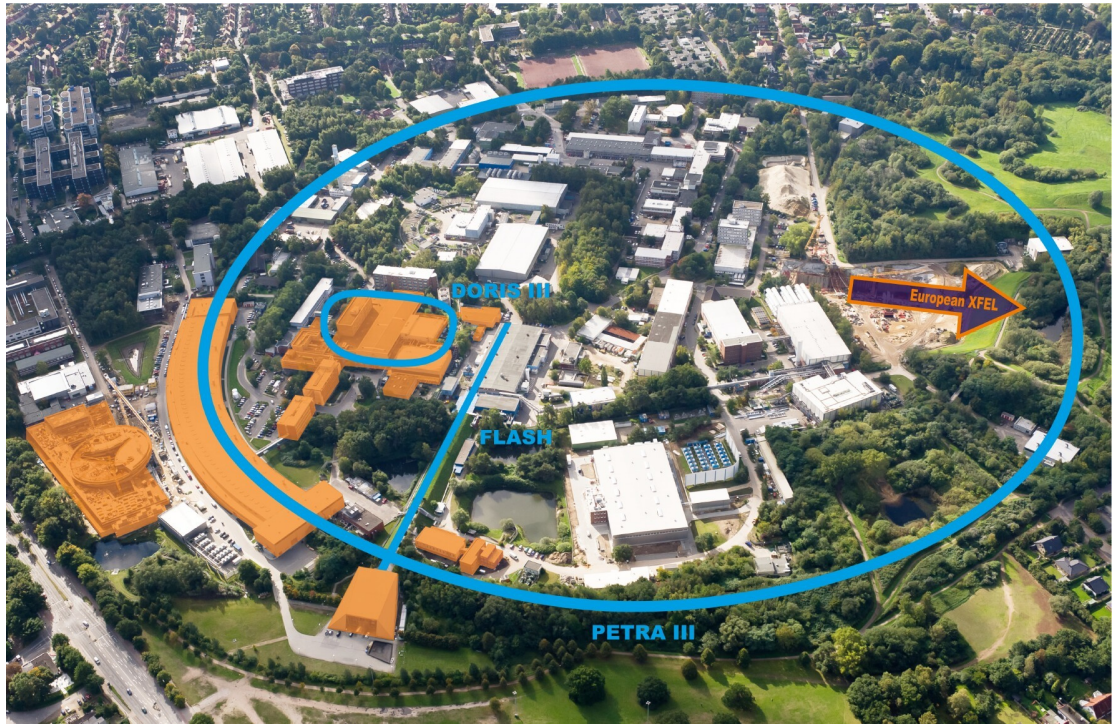


"full quantum control"

Progress enabled by coherence,
non-linearities, quantum effects

LCLS Stanford

2014 +



DESY Hamburg

SACLA Japan

ELI NP Bucharest

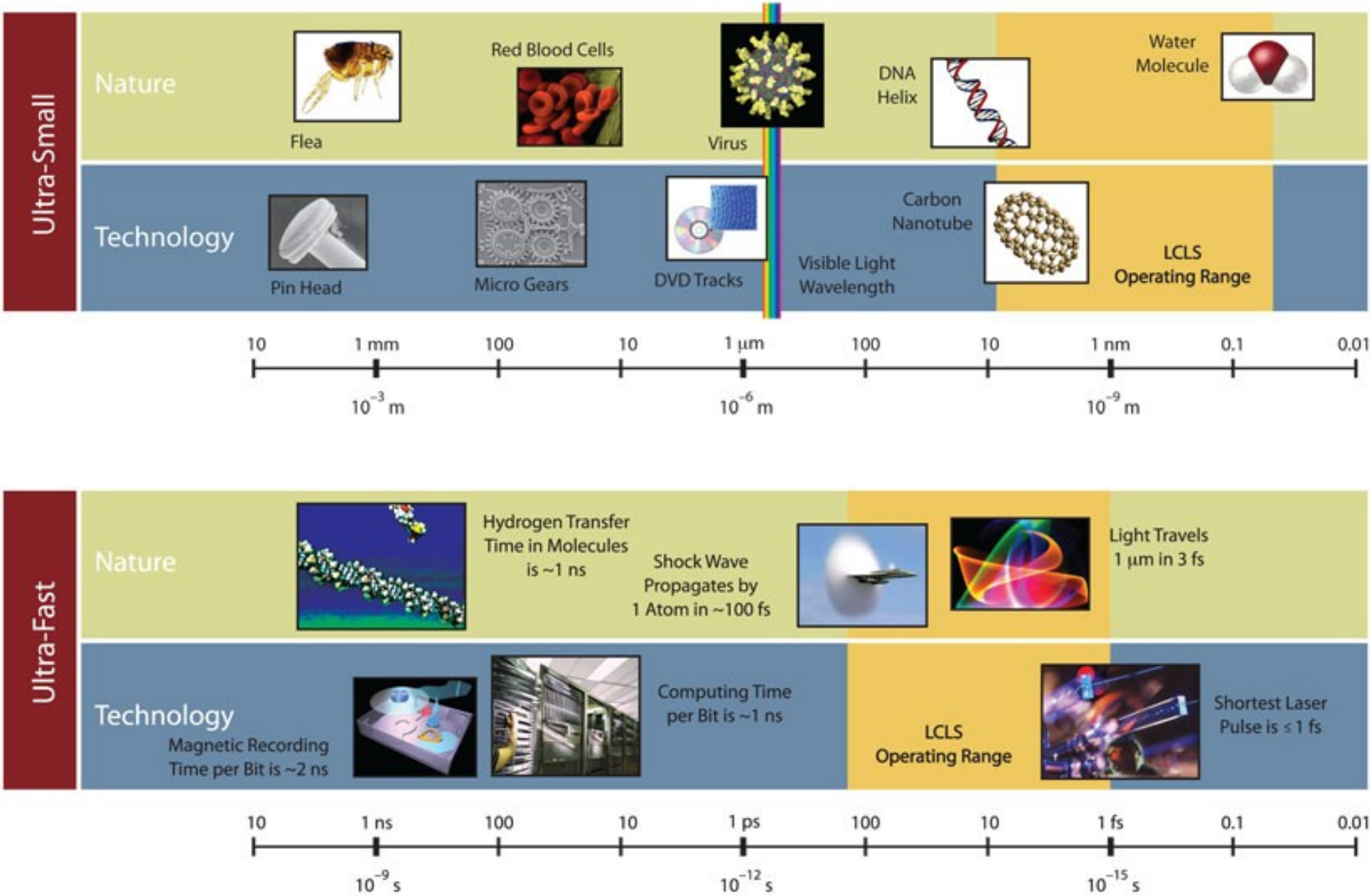

Ultra-Small
Nature
Flea
Red Blood Cells
Virus
DNA Helix
Water Molecule
Technology
Pin Head
Micro Gears
DVD Tracks
Visible Light Wavelength
Carbon Nanotube
LCLS Operating Range
10
1 mm
100
10
1 µm
100
10
1 nm
0.1
0.01
10^-3 m
10^-6 m
10^-9 m
Ultra-Fast
Nature
Hydrogen Transfer Time in Molecules is ~1 ns
Shock Wave Propagates by 1 Atom in ~100 fs
Light Travels 1 µm in 3 fs
Technology
Magnetic Recording Time per Bit is ~2 ns
Computing Time per Bit is ~1 ns
LCLS Operating Range
Shortest Laser Pulse is ≤ 1 fs
10
1 ns
100
10
1 ps
100
10
1 fs
0.1
0.01
10^-9 s
10^-12 s
10^-15 s

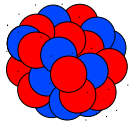
Image: LCLS

Resonant to nuclear or core-electron transitions

Since 2009 XFEL -

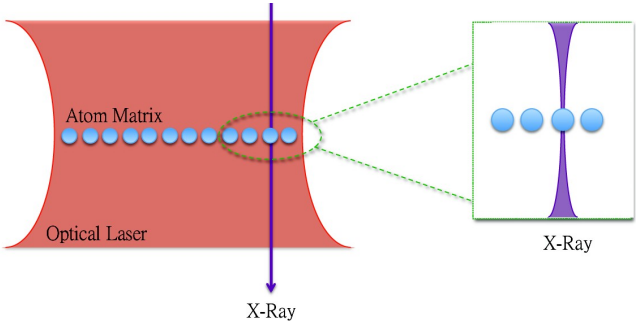
X-ray Free Electron Laser

?



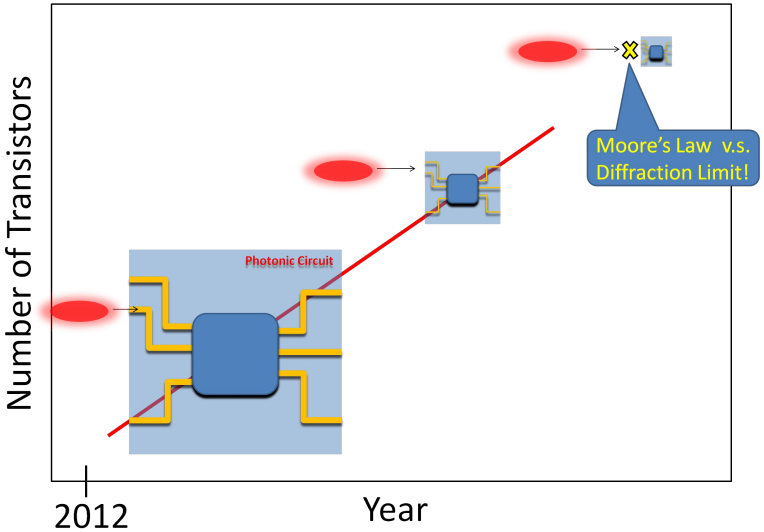
X-rays…

- More robust
- Better detection efficiency
- Better focus – see diffraction limit
- Deeper penetration
- Match NUCLEAR TRANSITIONS

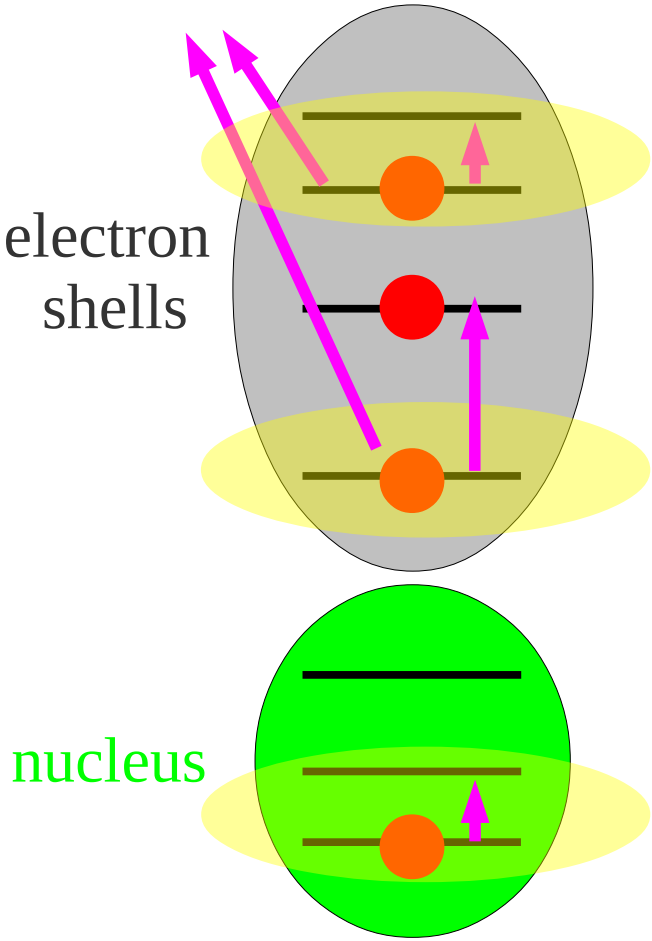


As qubits?

Photonic circuits?



Optical vs. x-ray



courtesy Jörg Evers

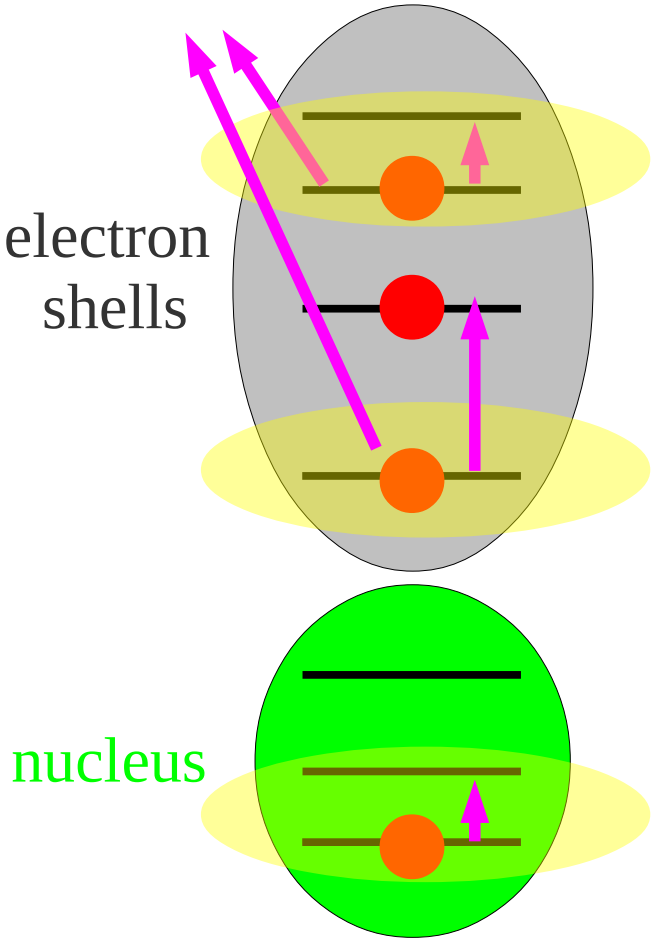
Light-matter interactions

- Optical laser fields
  control dynamics of 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

X-ray incentives



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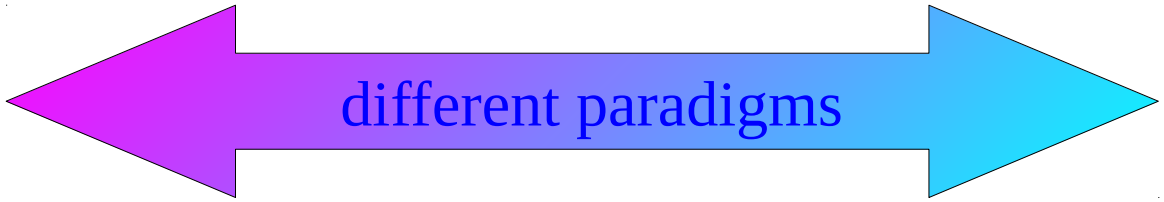


uncontrolled pump
+ passive observation

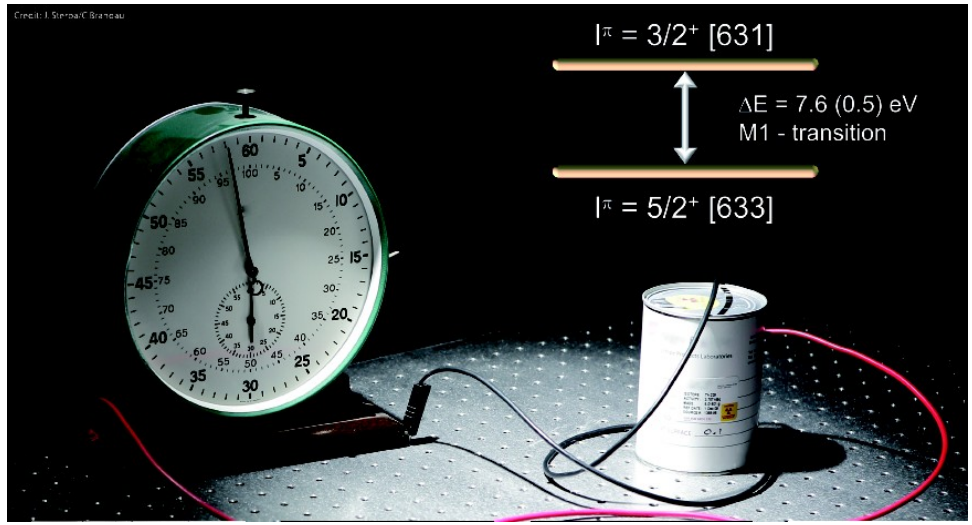
full quantum
control

- X-ray physics could greatly benefit from moving towards quantum/non-linear/control
- New light sources and upgrades now and in near future → now is the right time
- Such developments are necessary to fully exploit potential of new light sources

Special nuclear incentives ...

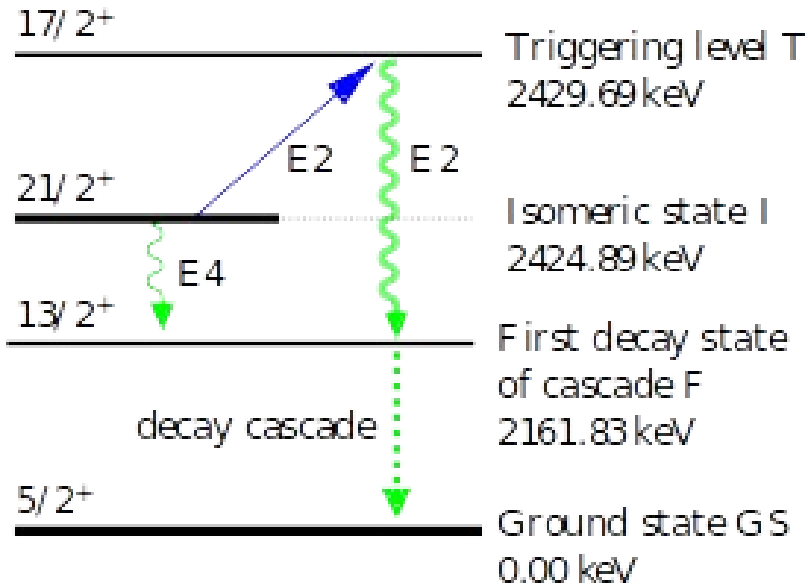
GAMMA-RAY LASERS

FREQUENCY STANDARDS



NUCLEAR ISOMERS

- long-lived nuclear states
- nuclear batteries
- astrophysical interest

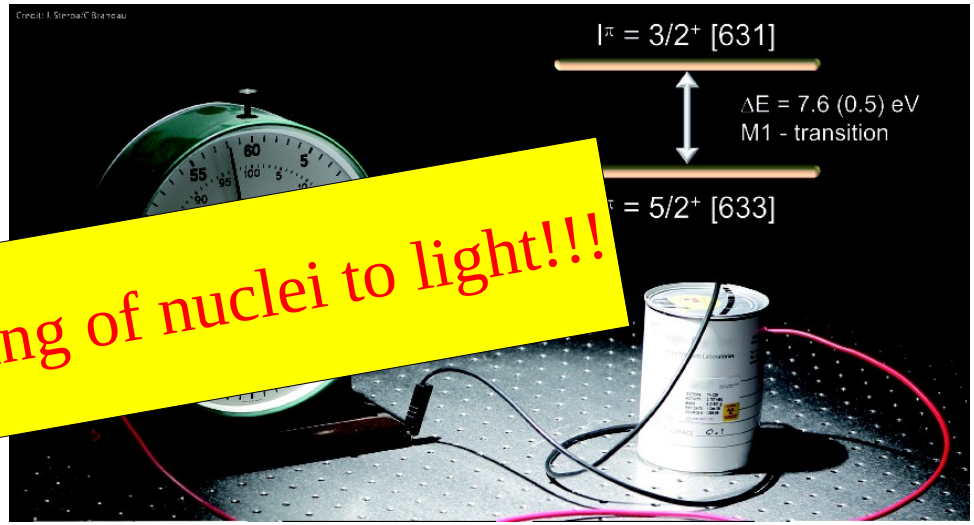


Special nuclear incentives ...

GAMMA-RAY LASERS

FREQUENCY

BUT … weak coupling of nuclei to light!!!



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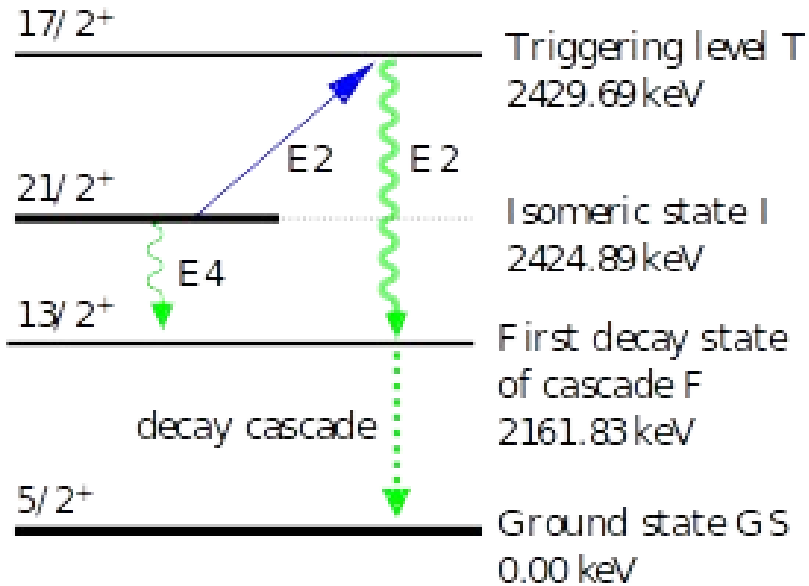
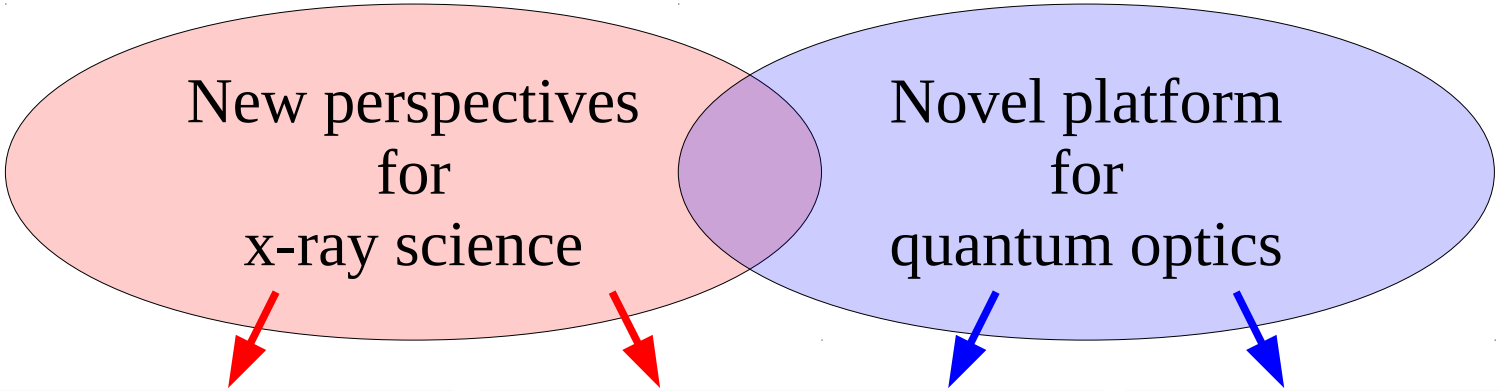


X-ray nuclear quantum optics!



Interesting alternative to electron shell

Nanostructuring + isotope selective addressing

FELs will enable entirely new parameter regime for nuclei

Conceptionally simple experiments

Exceptionally clean system

High Q of resonance

Potential for x-ray / optical hybrid systems

Experimental constraints "enforce" new ideas

Dynamically tunable multi-level schemes

Strong collective effects

courtesy Jörg Evers

Outline

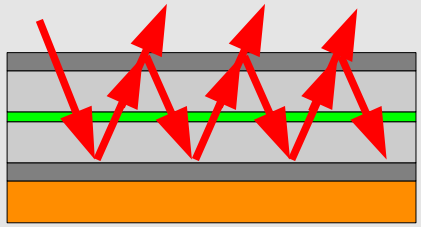
I. Photon-hungry quantum optics with nuclei

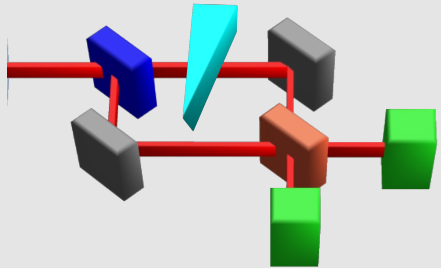
the source: XFEL
the nuclei: tricky
theory: density matrix
coherence vs. intensity
solid-state targets
accelerated ion targets
STIRAP

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

II. Single-photon quantum optics with nuclei

the source: SR
the nuclei: Mössbauer's ^{57}Fe
theory: Dicke, Maxwell-Bloch
nuclear forward scattering
nuclear waveguide cavities
storing a single photon
entanglement and slow light





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TODAY

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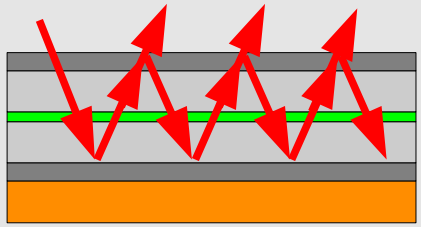

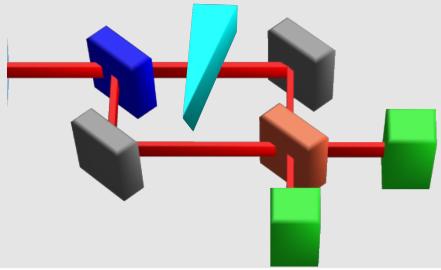

TOMORROW

Outline

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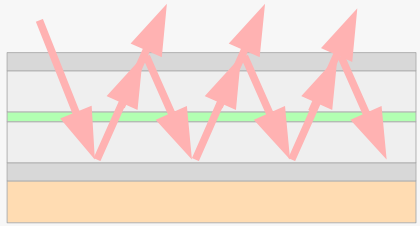
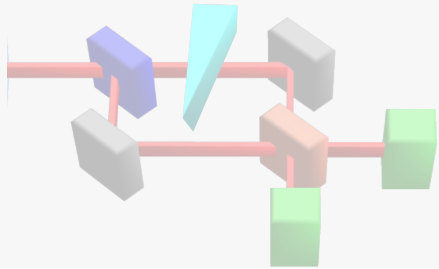




Previous experiences ...

- scaling in frequency from optical lasers + atoms

*coherent control**, *non-linear optics,*
dark states, slow light, entanglement,
electromagnetically induced transparency,
revolution in atomic physics, etc.

* relies on driving and control over most of the atomic state population!

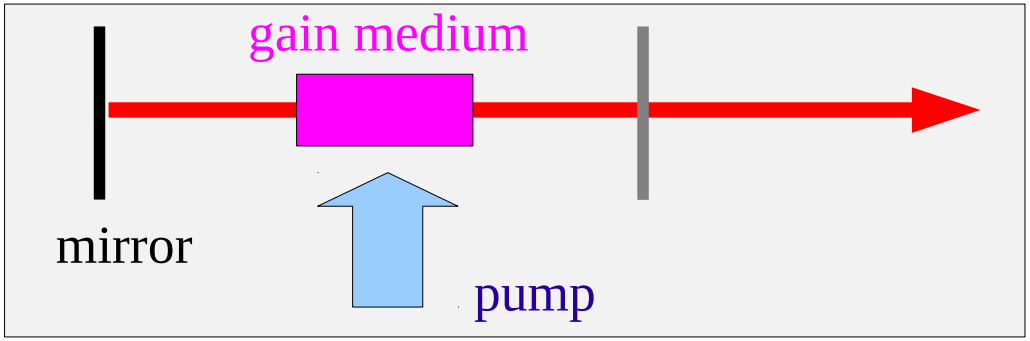
CAN WE ACHIEVE SOMETHING SIMILAR WITH NUCLEI AND X-RAYS?

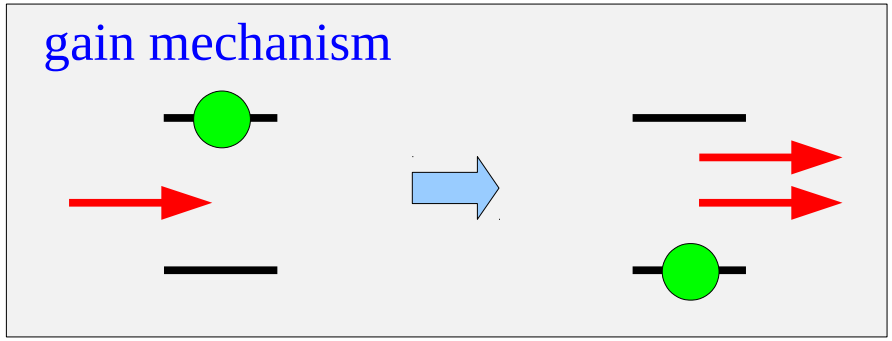
→ WE'LL NEED A LOT OF RESONANT PHOTONS FOR THAT!!!

The X-ray Free Electron Laser

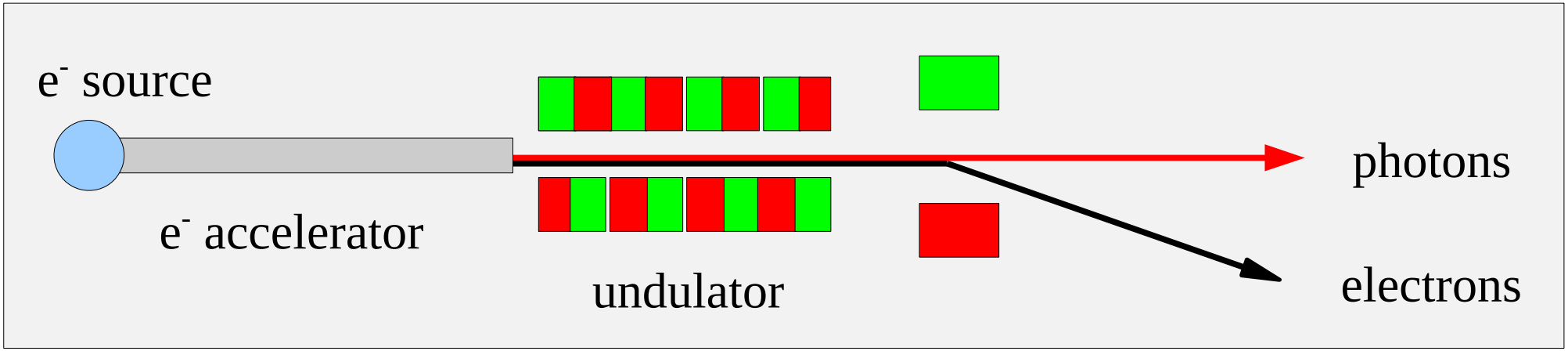
The XFEL

Conventional laser





Free electron laser

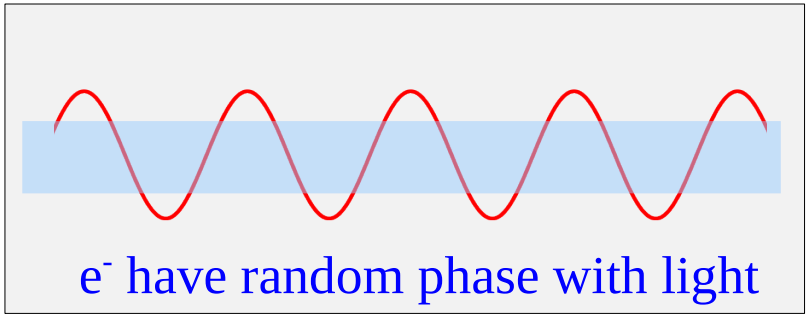


Light is amplified by converting electron energy into photons

XFEL vs. conventional undulator

Undulator

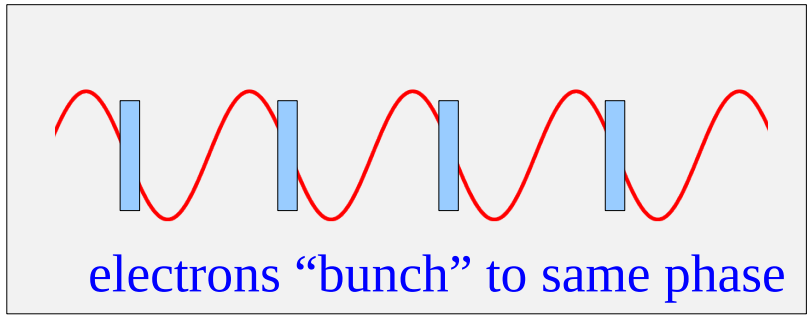
- Emissions of a single electron in different periods coherent
- Electrons uncorrelated



$$I \sim n_e$$

FEL

- Emissions of a single electron in different periods coherent
- Emission of different electrons coherent



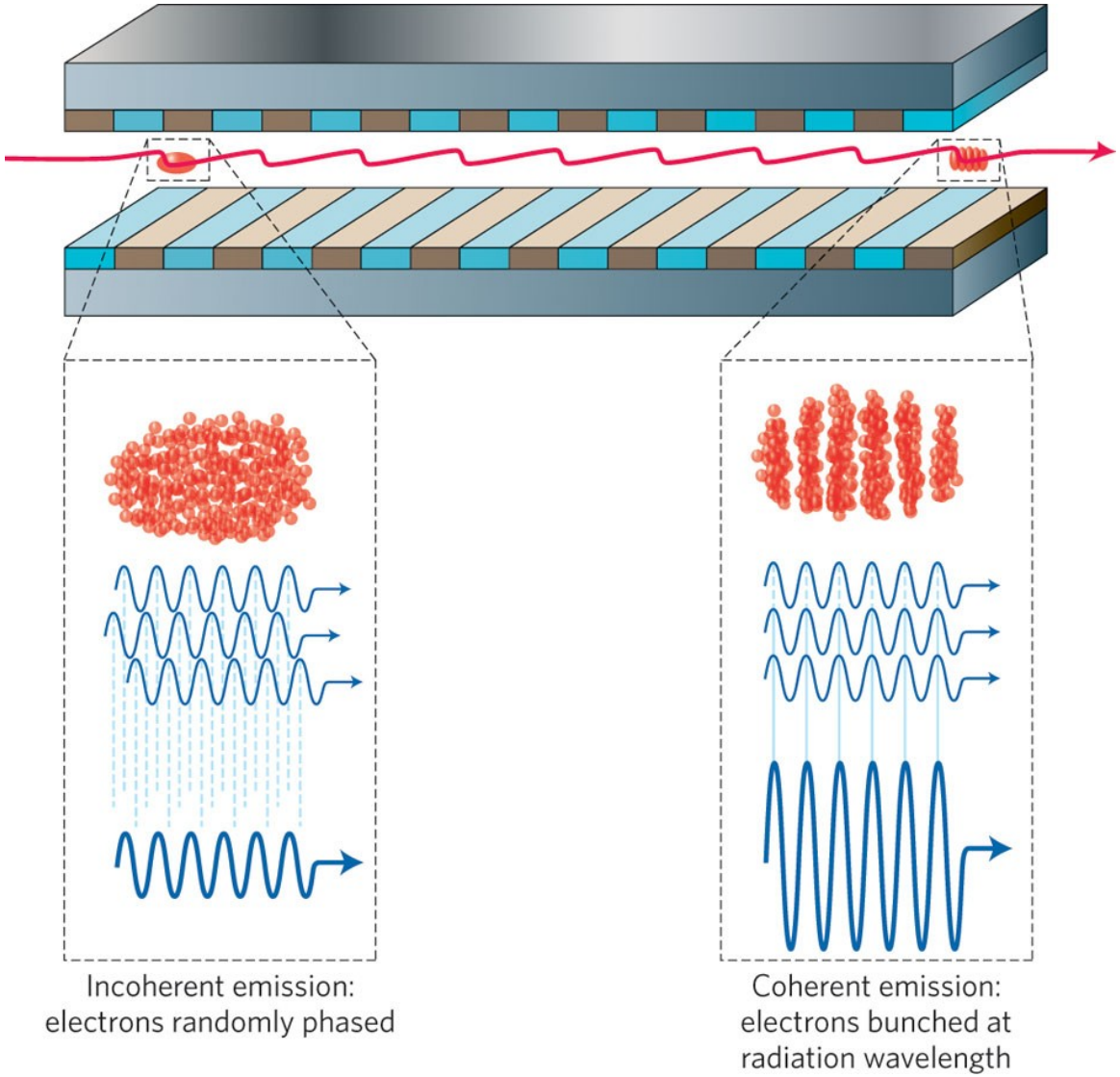
$$I \sim n_e^2$$

back-action of field on electrons leads to bunching

courtesy Jörg Evers

Microbunching

- Energy exchange depending on relative phase of electron and field
- Oscillation amplitude depends on electron energy
- This leads to to microbunching of electrons at the light wave length
- Therefore coherent emission of all electrons



Images: Nature Photonics 4, 814 (2010);

Some pictures

SLAC undulators (silver)

undulators magnets

Images: SLAC

Some numbers

- Photon energy up to 10-20 keV
- Full transverse coherence, upgrade to full longitudinal coherence possible
- High Brilliance - 10^{12} photons per pulse
- Short pulses - around 100 fs
- Bandwidth of approx. 0.08%

Some numbers

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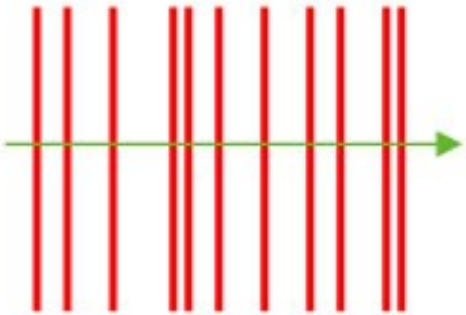
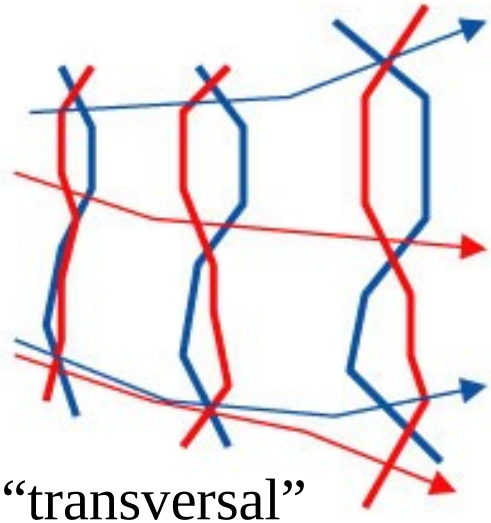
what do they mean?

Coherence



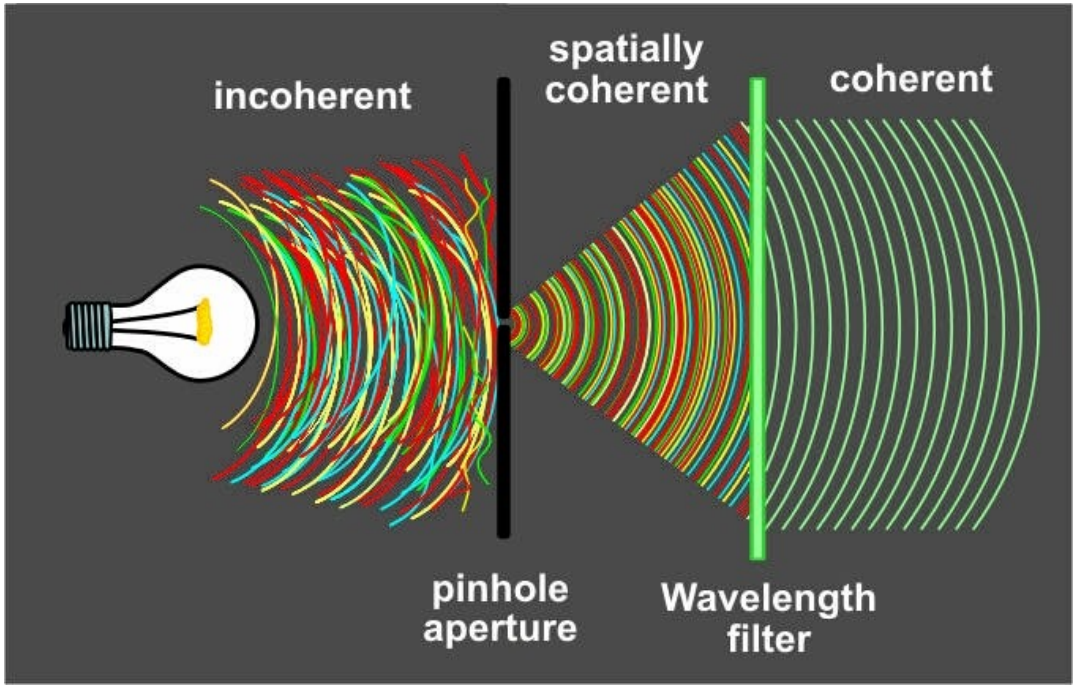
"longitudinal"

temporal coherence: random fluctuations in the *spacing* of the wavefronts

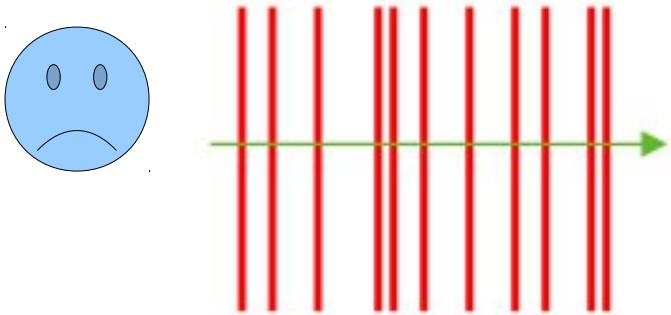


"transversal"

spatial coherence: random fluctuations in the *shape* of the wavefronts



Coherence for XFEL



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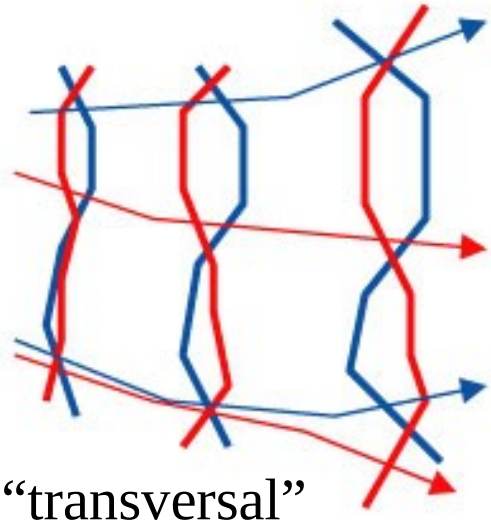

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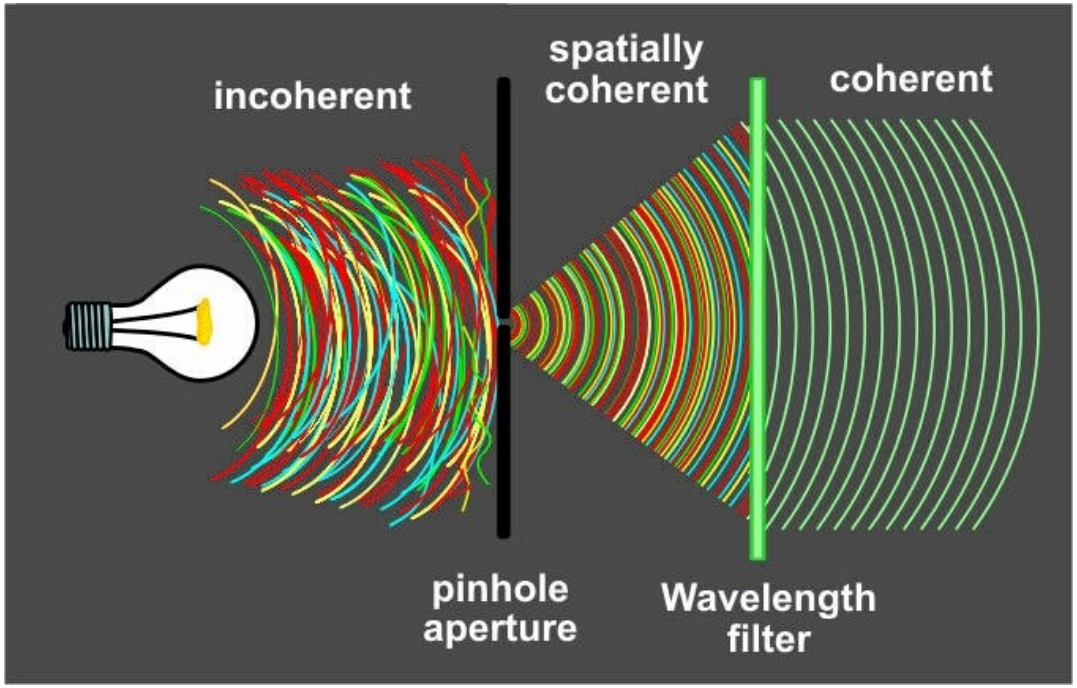
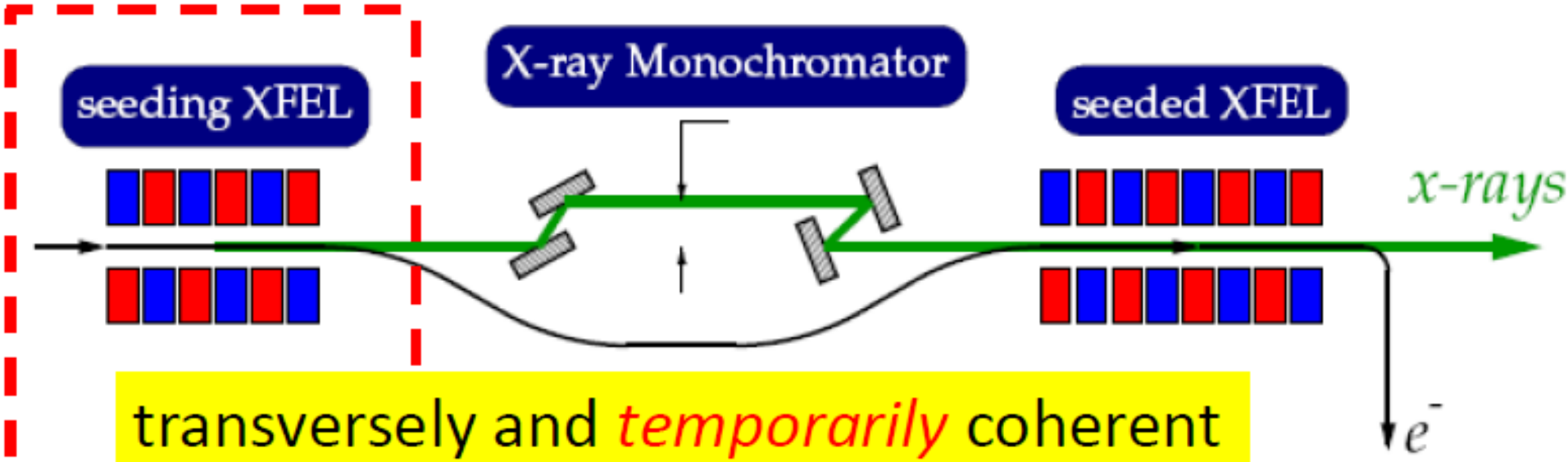


... seeded XFEL



transversely and *temporarily* coherent

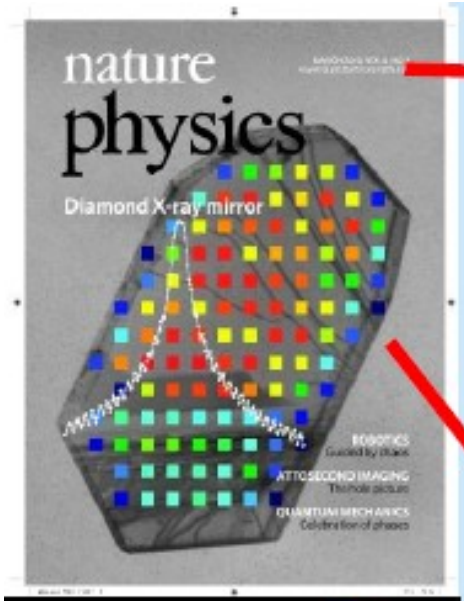
European XFEL

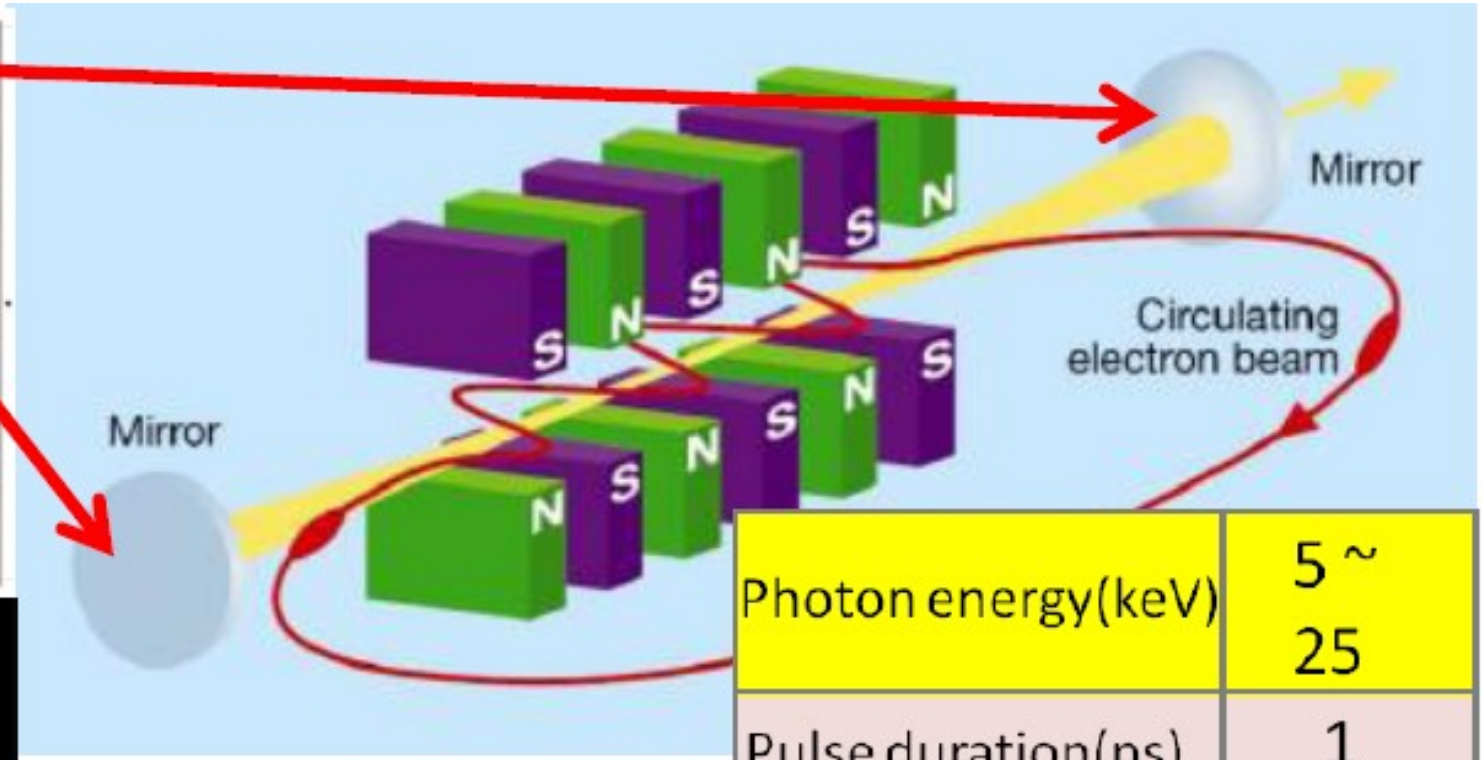
LCLS

Photon energy(keV)	0.2 ~ 12.4
Pulse duration(ps)	0.1
Band width (meV)	10

E. L. Saldin et al., **Nucl. Instrum. Meth. A 475, 357 (2001)**

... XFEL oscillator





Photon energy(keV)	5 ~ 25
Pulse duration(ps)	1
Band width (meV)	2

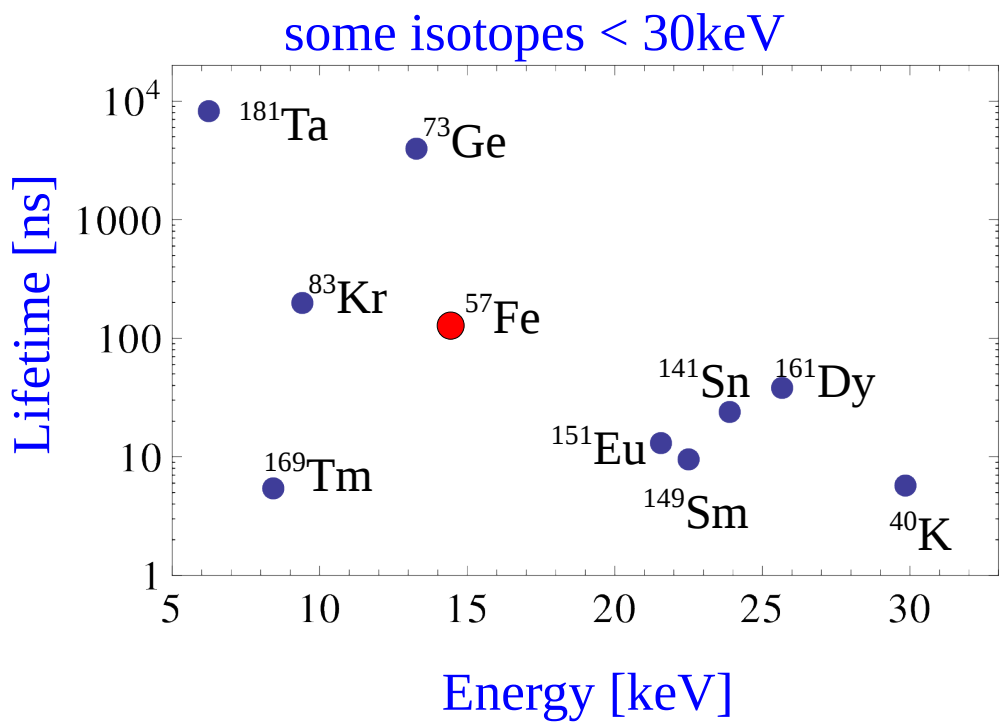
transversely and *temporarily* coherent

K.-J. Kim, Y. Shvyd'ko and S. Reiche, **PRL100, 244802 (2008)**

Y. Shvyd'ko et al., **Nature Phys. 6, 196 (2010)**

The nuclei

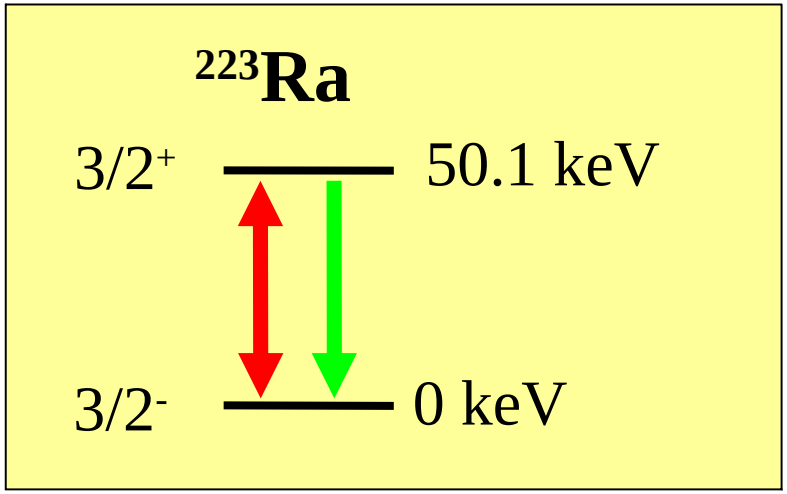
Nuclear transition energies



but many more with higher energies – use target acceleration!

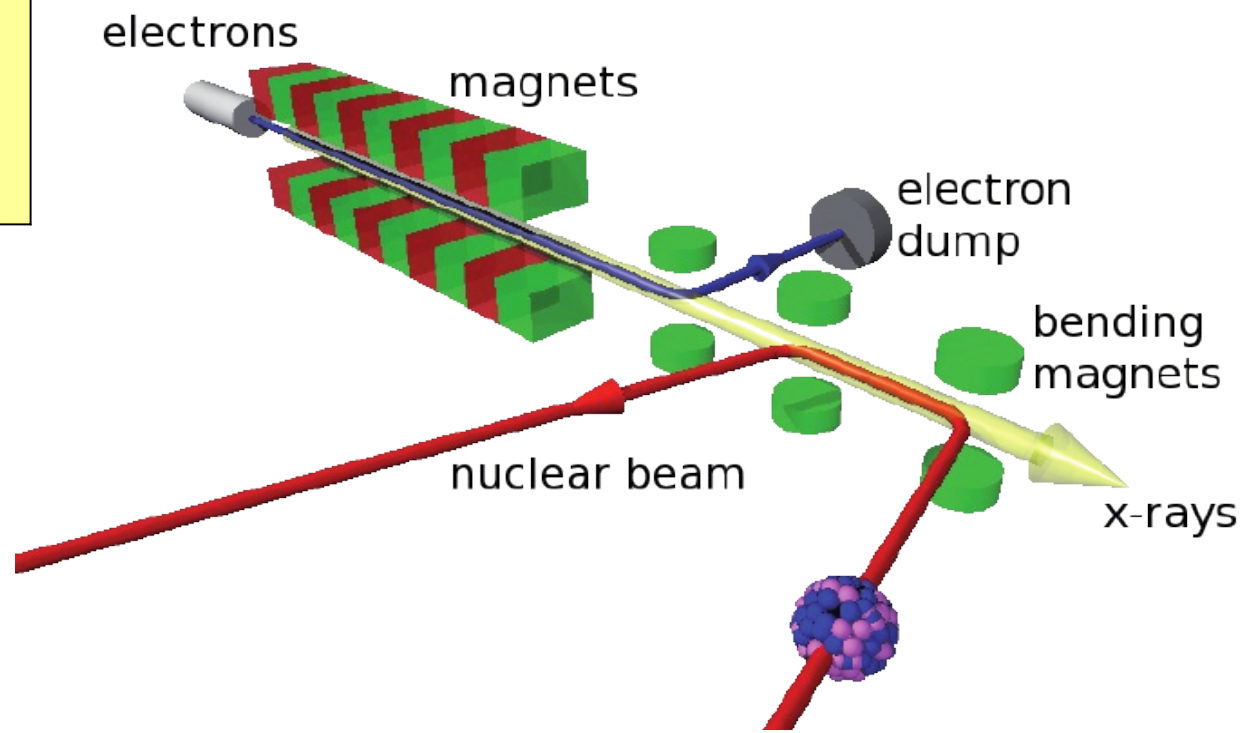
Accelerated nuclei as targets

bridge the gap between nuclear excitation and photon energies...



Doppler shift

$$E_n = (1+\beta)\gamma E_L$$



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

Accelerated nuclei as targets

bridge the gap between nuclear excitation and photon energies...

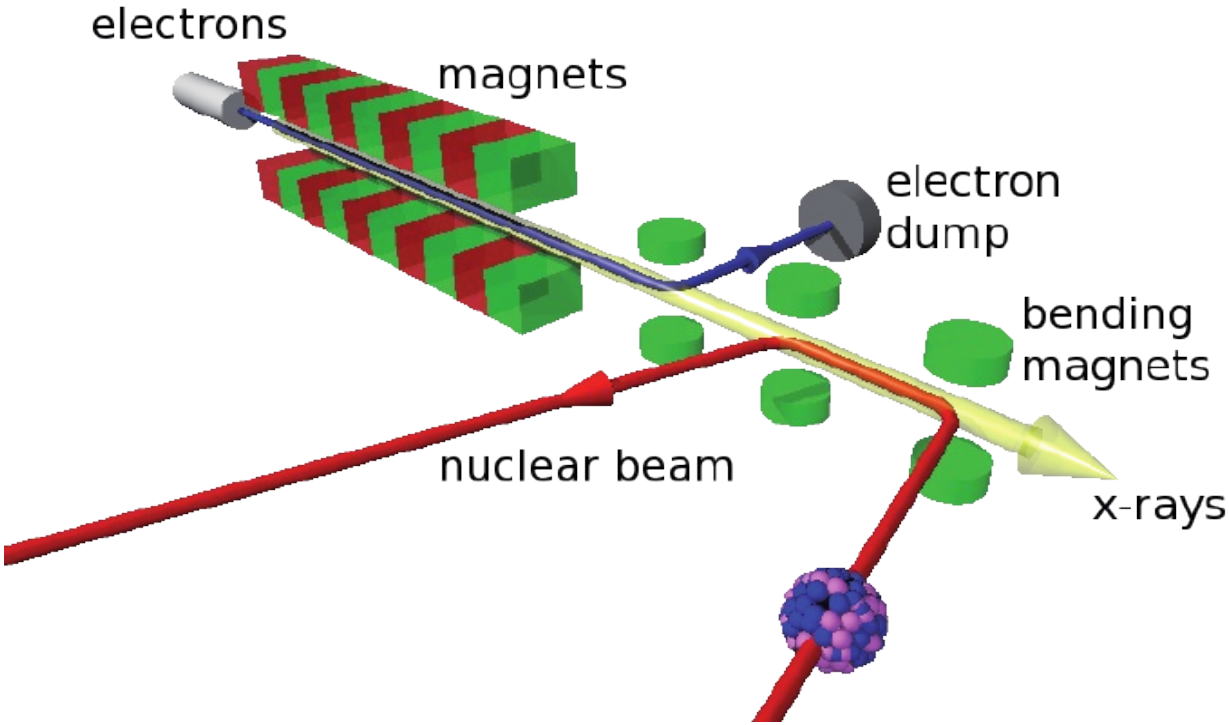
Doppler shift

$$\beta = \frac{v}{c} \qquad \gamma = \frac{1}{\sqrt{1-\beta^2}}$$

$$t_b = \frac{t_l}{(1+\beta)\gamma}$$

$$I_b = I_l(1+\beta)\gamma$$

$$E_b = E_n = (1+\beta)\gamma E_l$$



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

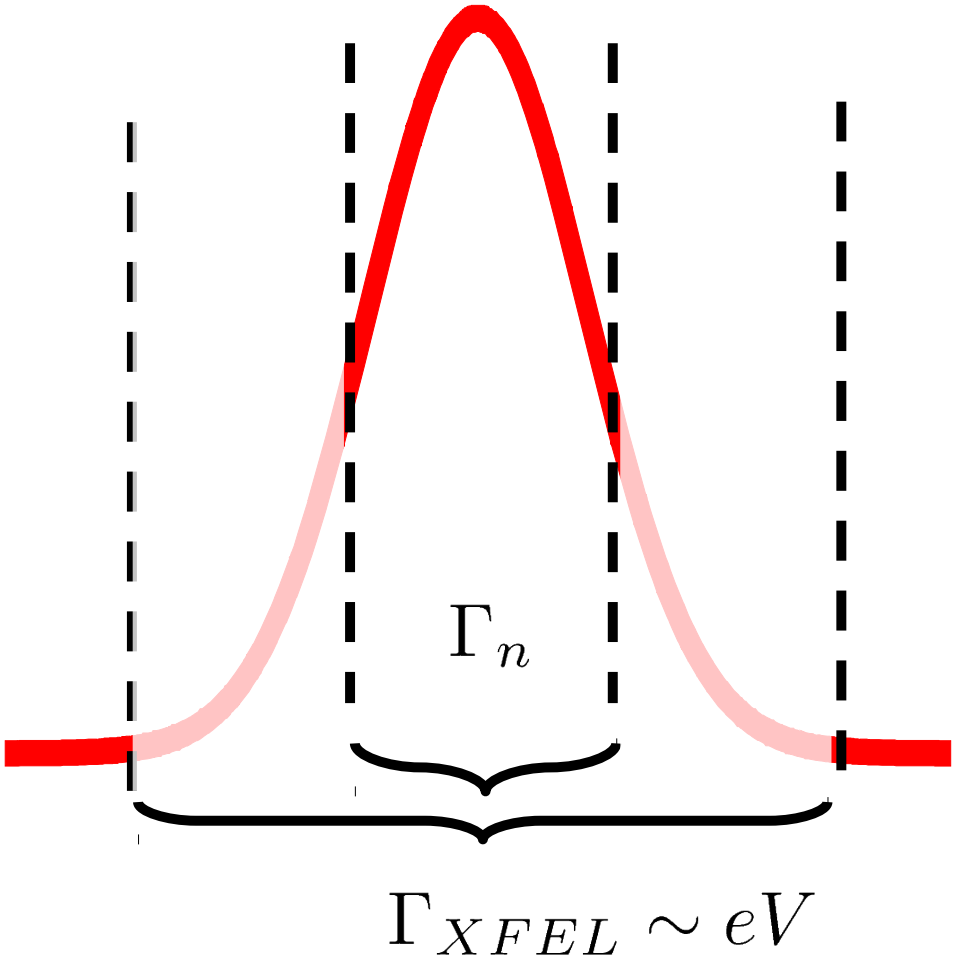
Nuclear transition widths

Nuclear widths are very narrow!

$$\Gamma_n \sim 10^{-8} - 10^{-5}\ eV$$

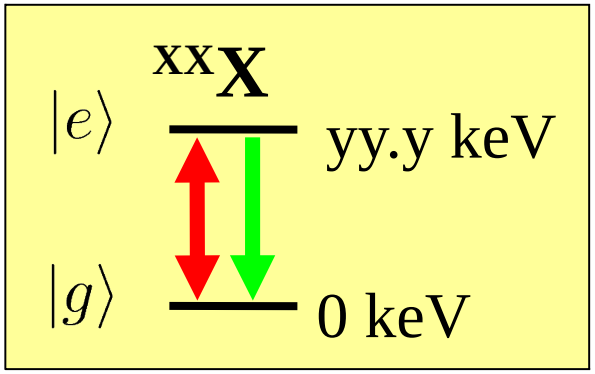
$$I_{eff} = I \frac{\Gamma_n}{\Gamma_{XFEL}}$$

effective intensity!



Quantum optics approach

Density matrix formalism



$$\rho = |\Psi\rangle\langle\Psi| \qquad |\Psi\rangle = c_g|g\rangle + c_e|e\rangle$$

→ so-called Bloch equations

$$\frac{\partial\rho}{\partial t} = \frac{i}{\hbar}[H_o, \rho] - \underbrace{\frac{\gamma_{SE}}{2}\left([A_{eg}, A_{ge}\rho] + H.c.\right)}_{spontaneous \quad emission} - \underbrace{\gamma_d\left([A_{ee}, A_{ee}\rho] + H.c.\right)}_{decoherence}$$

$$A_{ij} = |i\rangle\langle j|, \quad i, j \in \{e, g\}$$

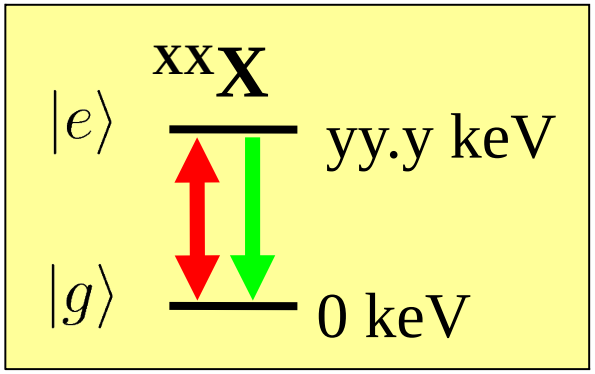
$$H_0 = -\frac{1}{c}\int d^3r\ \vec{j}(\vec{r}) \cdot \vec{A}(\vec{r}, t)$$

AP, J. Evers and C. H. Keitel, **PRC 77, 044602 (2008)**

Nuclear transition might be “forbidden”

Atomic quantum optics – electric dipole E1 transitions

Low-lying nuclear transitions - M1, E2, rarely E1



$$\frac{\partial \rho}{\partial t} = \frac{i}{\hbar}[H_o, \rho] - \underbrace{\frac{\gamma_{SE}}{2}\left([A_{eg}, A_{ge}\rho] + H.c.\right)}_{spontaneous \quad emission} - \underbrace{\gamma_d\left([A_{ee}, A_{ee}\rho] + H.c.\right)}_{decoherence}$$

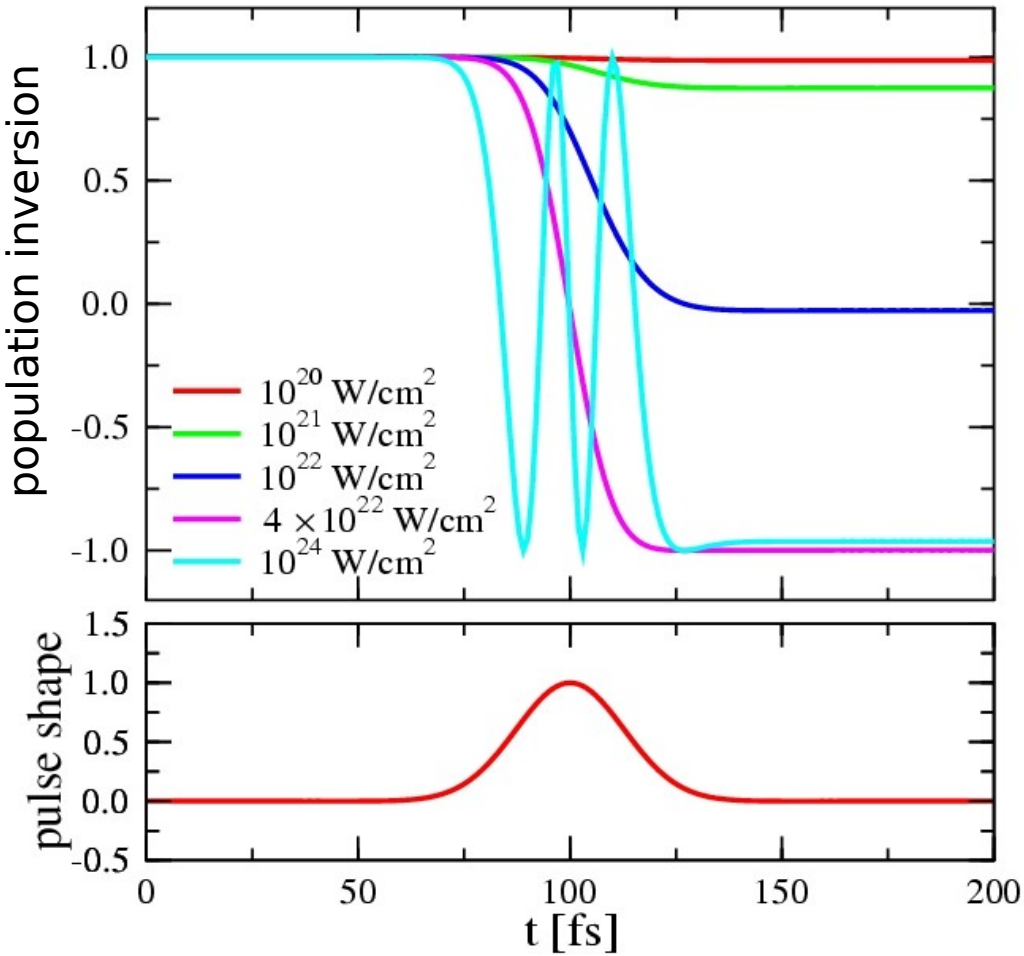
Beyond the dipole approximation!

$$A_{ij} = |i\rangle\langle j|, \quad i, j \in \{e, g\}$$

$$H_0 = -\frac{1}{c}\int d^3r \; \vec{j}(\vec{r}) \cdot \vec{A}(\vec{r}, t) \propto \mathfrak{M}_{LM} \neq -e\vec{r} \cdot \vec{E}$$

AP, J. Evers and C. H. Keitel, **PRC 77, 044602 (2008)**

Nuclear Rabi flopping



Population inversion in ^{223}Ra

Population inversion ~

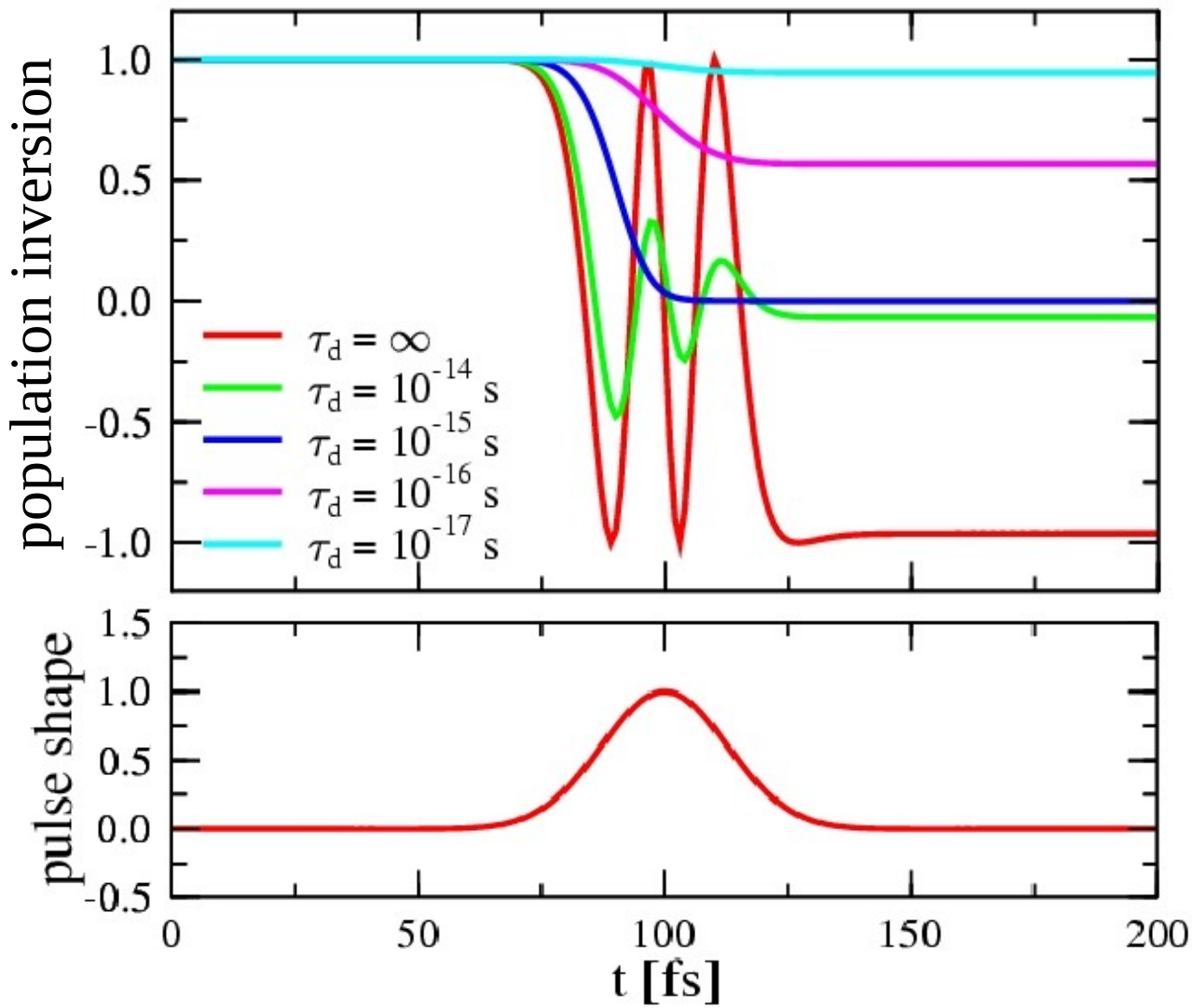
$$\langle g|\rho|g\rangle - \langle e|\rho|e\rangle$$

- resonant laser-nucleus interaction allows to induce Rabi flopping of nuclear population
- detection e.g. via scattered light, state-selective measurements
- potential application: model-free determination of nuclear parameters

T. Bürvenich, J. Evers and C. H. Keitel, Phys. Rev. Lett. 96, 142501 (2006)

Coherence in nuclear Rabi floping

coherence ⟷ intensity



strongly incoherent

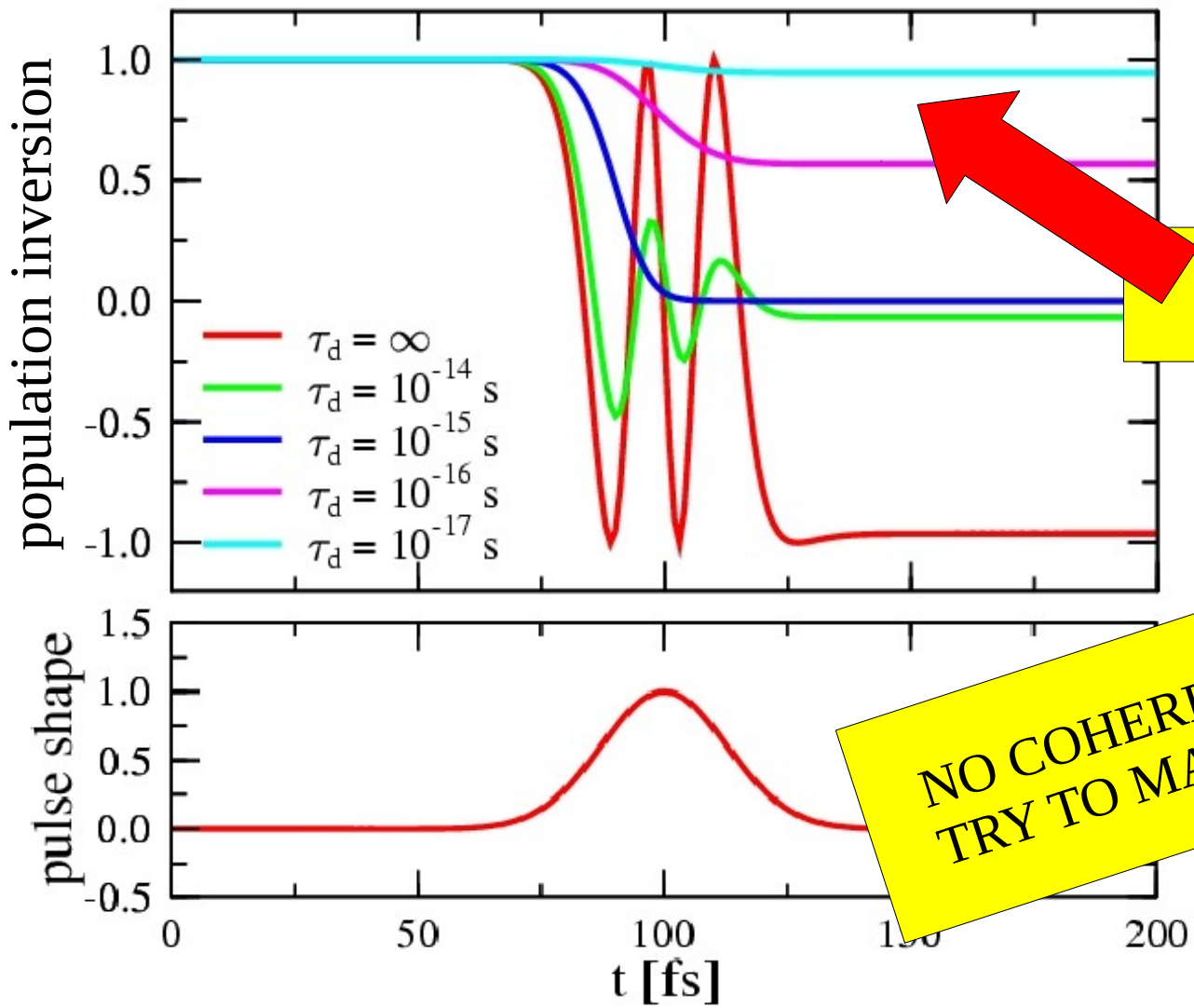
partially coherent

coherent

T. Bürvenich, J. Evers and C. H. Keitel, Phys. Rev. Lett. 96, 142501 (2006)

Coherence in nuclear Rabi floping

coherence ⟷ intensity



strongly incoherent

WE ARE HERE!

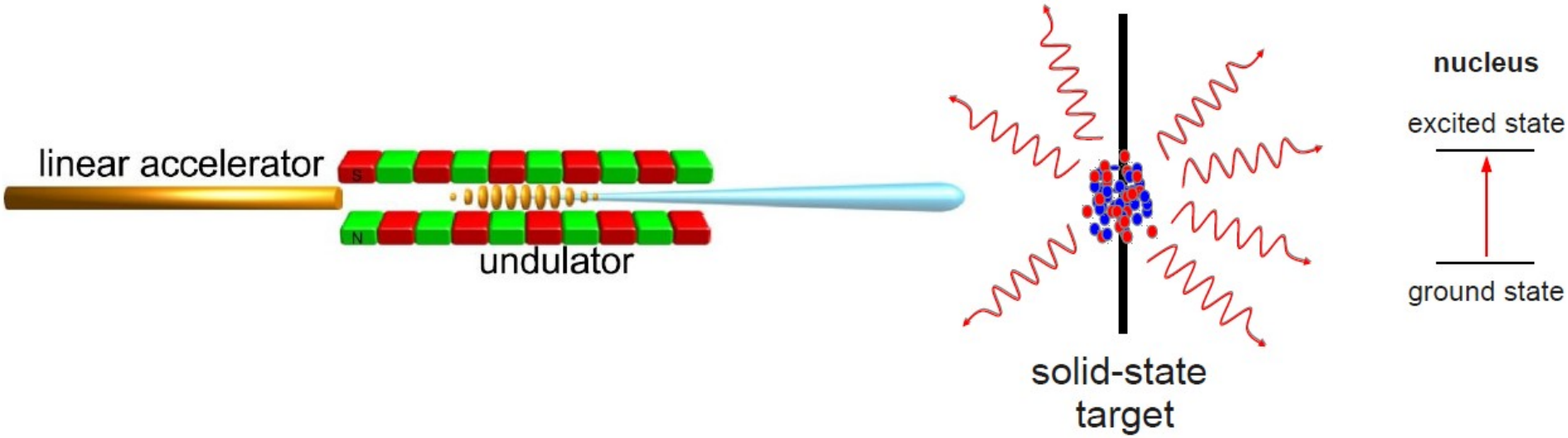
coherent

NO COHERENT EFFECTS YET
TRY TO MAXIMIZE EXCITATION

T. Bürvenich, J. Evers and C. H. Keitel, Phys. Rev. Lett. 96, 142501 (2006)

Maximizing excitation by high-density targets

Solid-state targets

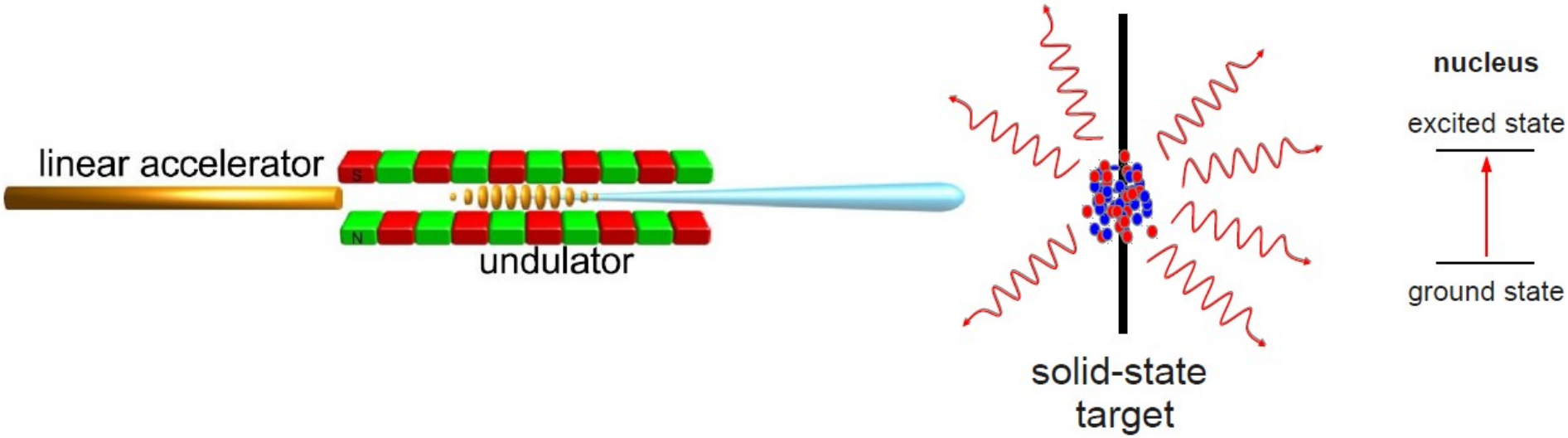


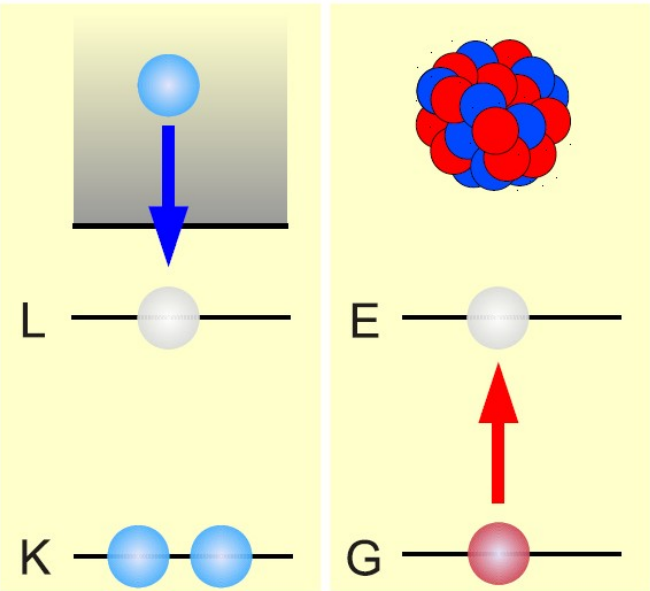
solid-state density $10^{22} - 10^{23} \ \mathrm{cm}^{-3}$

Secondary nuclear processes become possible in the plasma environment:

- secondary photoexcitation
- Coulomb excitation
- coupling to the atomic shell (NEET, NEEC)

Solid-state targets





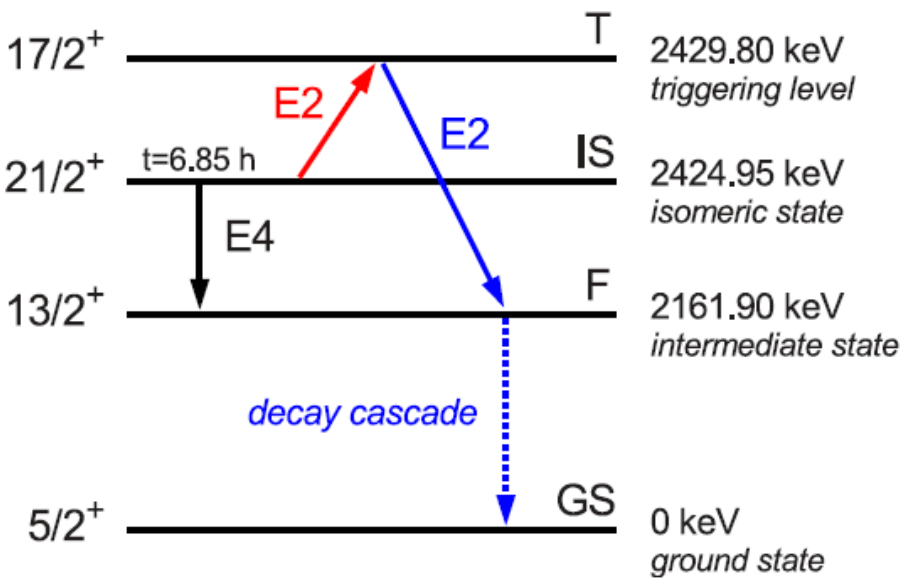
Nuclear Excitation by Electron Capture

- time–reversed process of internal conversion (IC)
- resonant process

Direct vs. secondary excitation

Idea of isomer triggering

Depopulate long–lived isomeric state by excitation to a triggering level which is linked to freely radiating states.

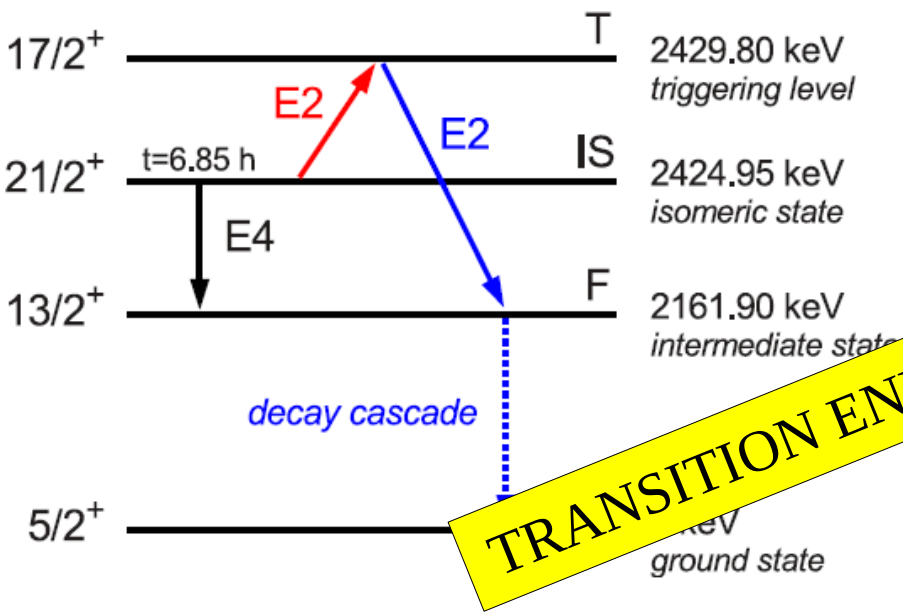


- IS - T: theoretically predicted
- T - F: experimentally confirmed
- transition energy: 4.85(8) keV
- level width of T: $\sim 10^{-7}$ eV
- decay cascade: 1 MeV photon = signature for triggering
- isomer production: ${}^{93}_{41}$Nb(p,n) ${}^{93m}_{42}$Mo reactions

Direct vs. secondary excitation

Idea of isomer triggering

Depopulate long–lived isomeric state by excitation to a triggering level which is linked to freely radiating states.



TRANSITION ENERGY UNCERTAINTY 80 eV!!

- IS - T: theoretically predicted
- T - F: experim onfirmed
- trans gy: 4.85(8) keV
  width of T: $\sim 10^{-7}$ eV
- decay cascade: 1 MeV photon = signature for triggering
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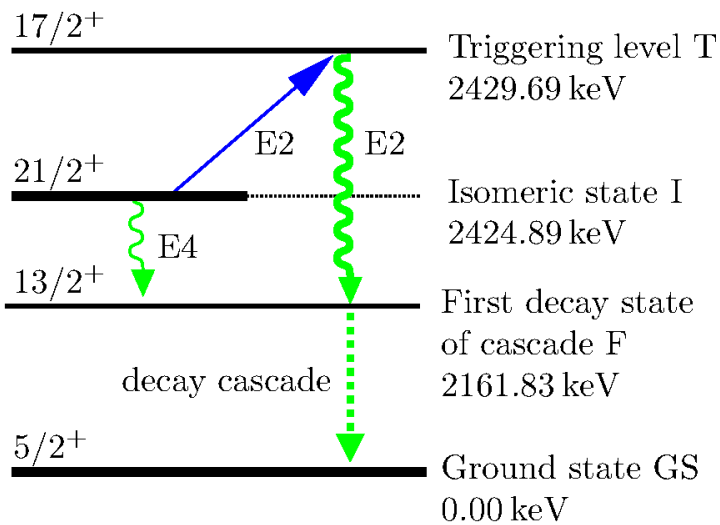
Direct vs. secondary excitation



Partial level scheme of $^{93}_{42}\mathrm{Mo}$

Triggering mechanisms

- Photoexcitation
- NEEC

Typically, $\sigma_{neec} \simeq 100 \times \sigma_{ph}$

for low-lying triggering levels

Competition in the nuclear excitation process between

resonant XFEL photons – direct photoexcitation
plasma electrons – NEEC

(Rough) plasma estimates

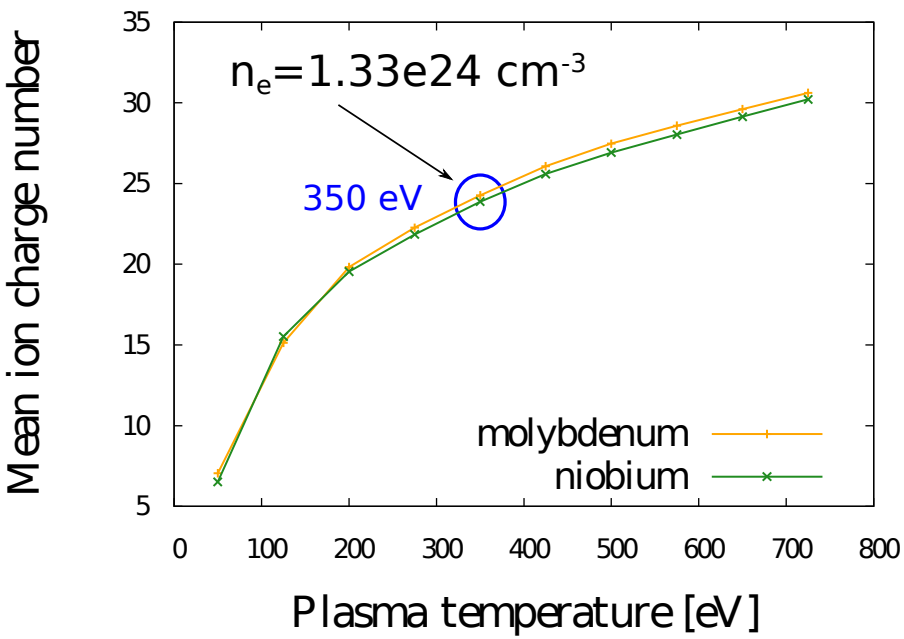
XFEL-induced plasma:
inner-shell holes, uniform radiation,
rapid heating, cold ions

LETTER

doi:10.1038/nature10746

Creation and diagnosis of a solid-density plasma with an X-ray free-electron laser

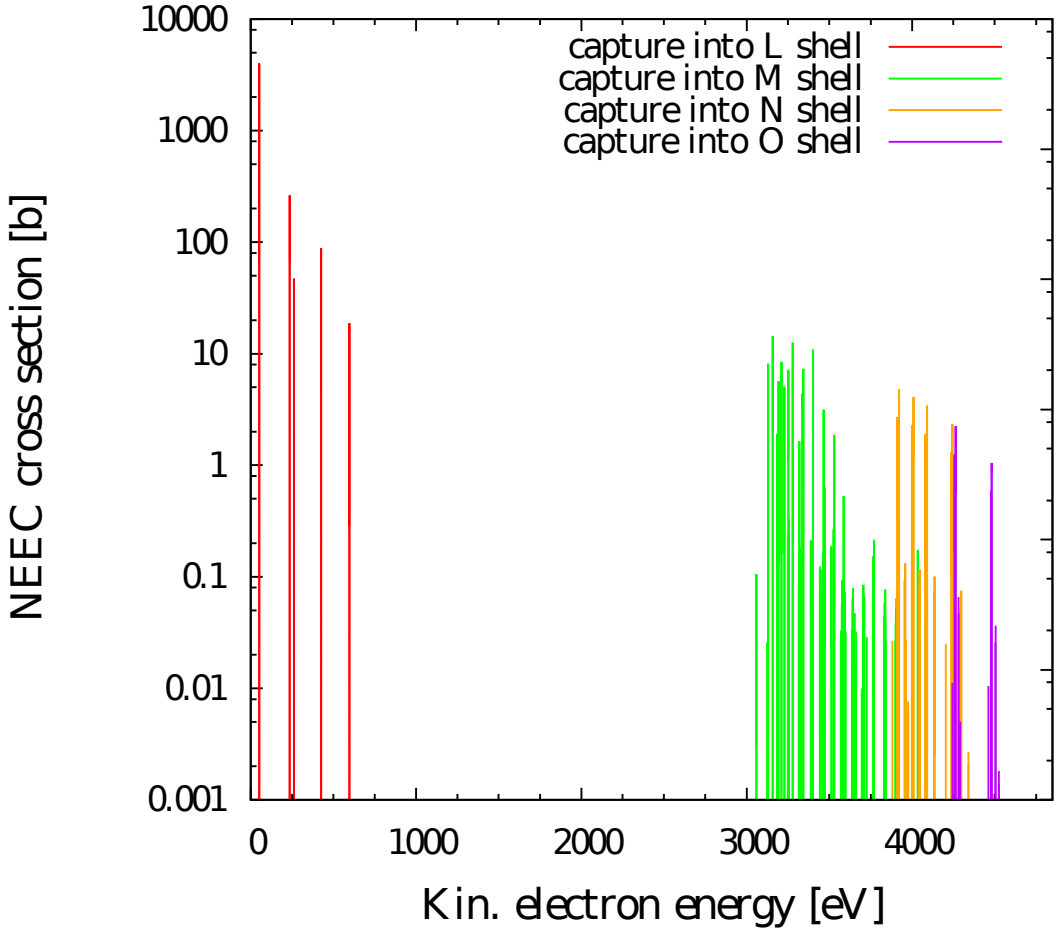
S. M. Vinko[1], O. Ciricosta[1], B. I. Cho[2], K. Engelhorn[2], H.-K. Chung[3], C. R. D. Brown[4], T. Burian[5], J. Chalupský[5], R. W. Falcone[2,6], C. Graves[7], V. Hájková[5], A. Higginbotham[1], L. Juha[5], J. Krzywinski[7], H. J. Lee[7], M. Messerschmidt[7], C. D. Murphy[1], Y. Ping[8], A. Scherz[7], W. Schlotter[7], S. Toleikis[9], J. J. Turner[7], L. Vysin[5], T. Wang[7], B. Wu[7], U. Zastrau[10], D. Zhu[7], R. W. Lee[2], P. A. Heimann[2], B. Nagler[7] & J. S. Wark[1]



- electrons equilibrate quickly
- uniform electron temp. T_e and density n_e
- T_e estimate from deposited laser energy
- FLYCHK: calculation of charge state distribution and n_e (rate equation model)
- ions stay at room temperature and solid–state density
- hydrodynamic expansion is neglected
- plasma lifetime $\sim$ 100 ps

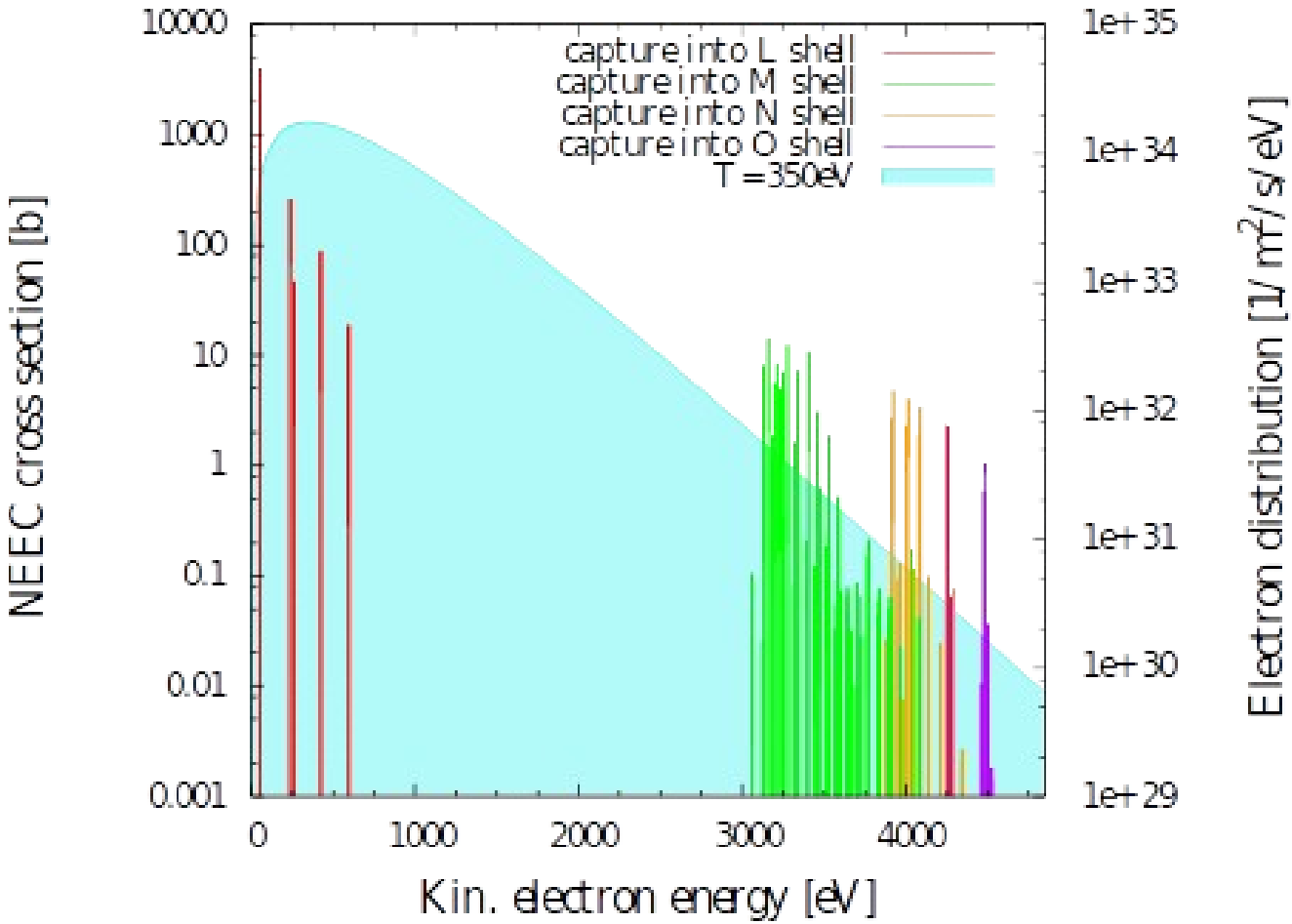
FLYCHK
H. K. Chung et al. HEDP 1, 3 (2005)

Electrons for NEEC



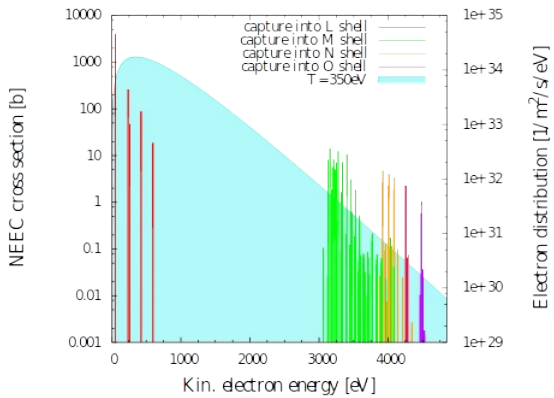
J. Gunst, Y. A. Litvinov, C. H. Keitel and AP, PRL 112, 082501 (2014)

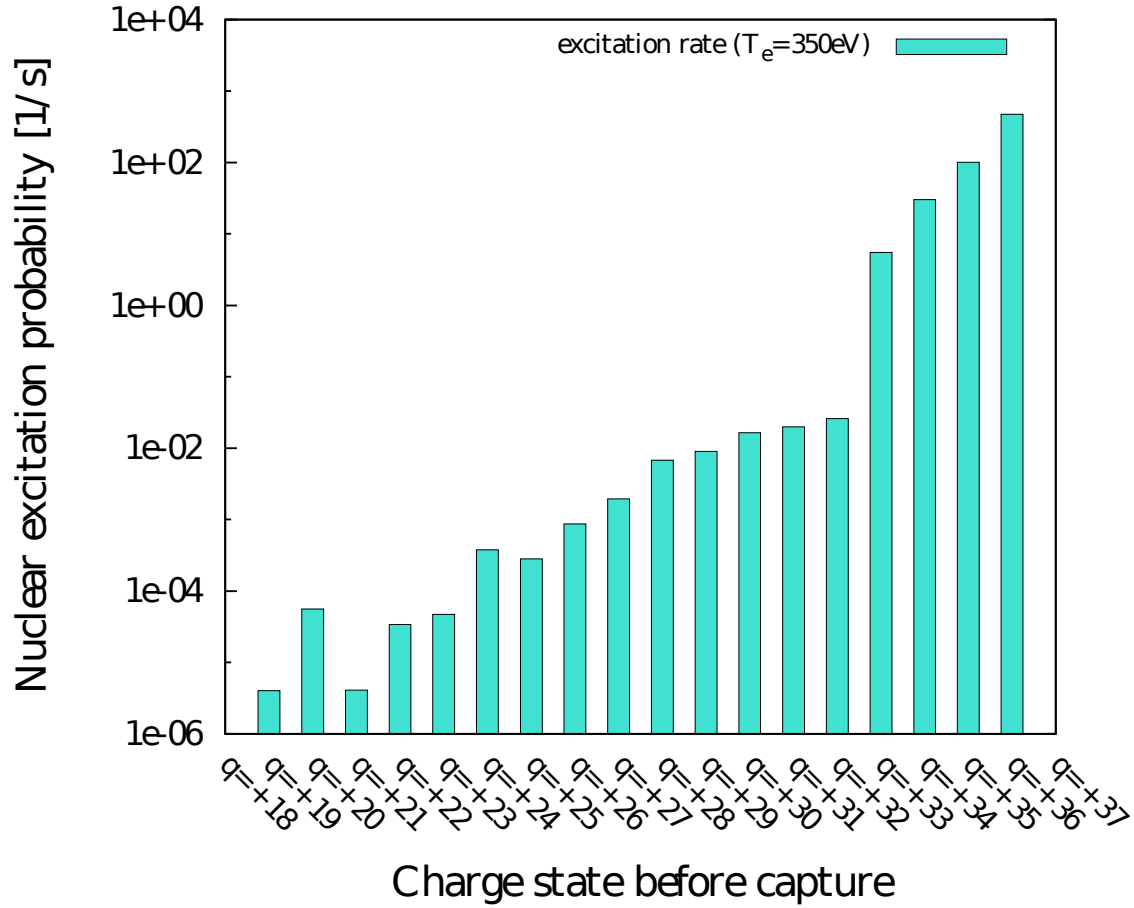
Electrons for NEEC



J. Gunst, Y. A. Litvinov, C. H. Keitel and AP, PRL 112, 082501 (2014)

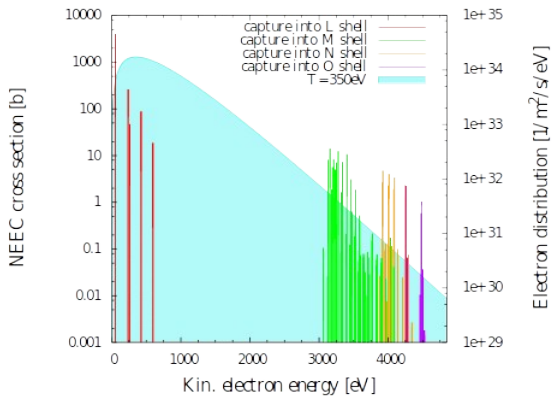
Electrons for NEEC

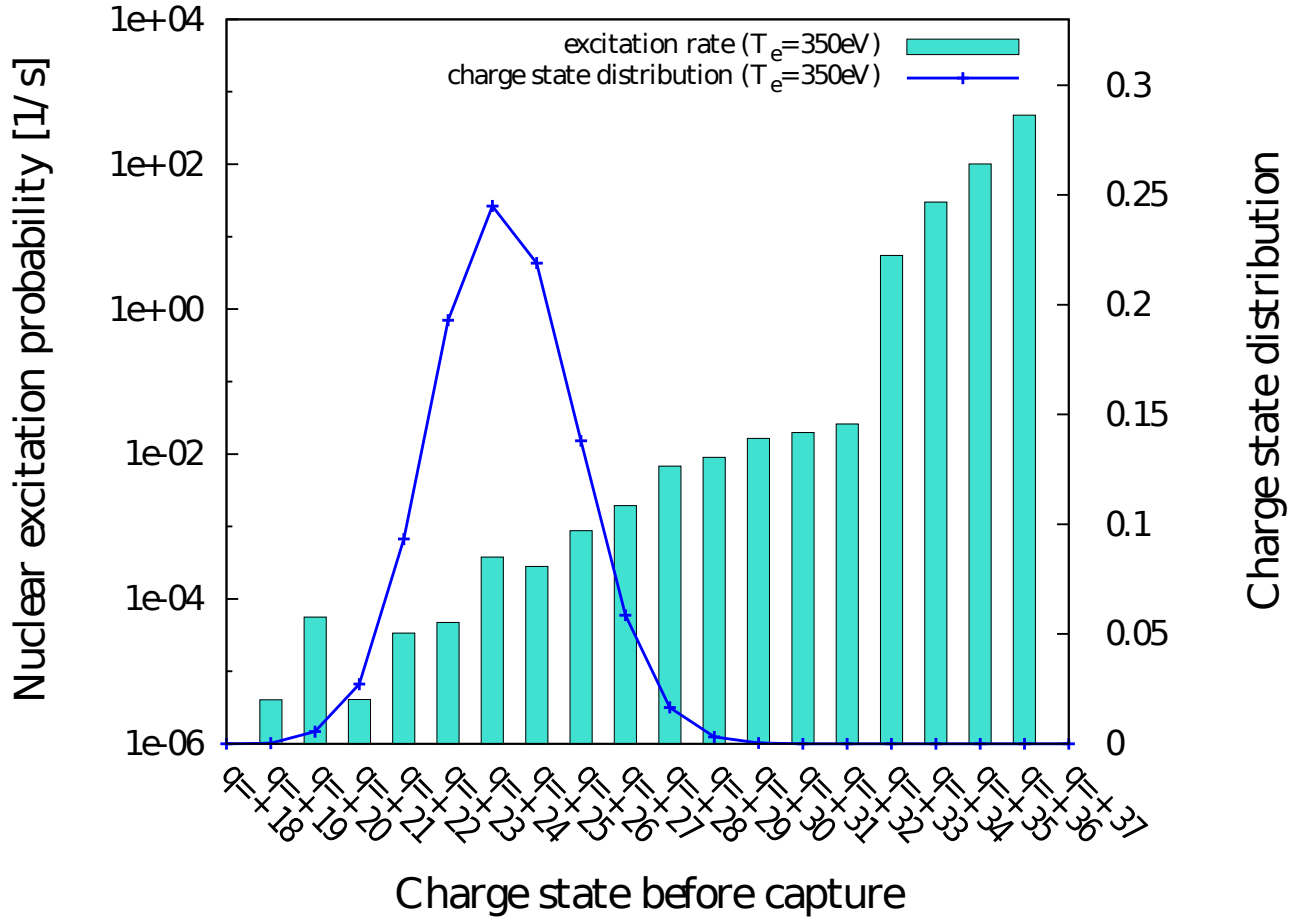




J. Gunst, Y. A. Litvinov, C. H. Keitel and AP, PRL in press (2014)

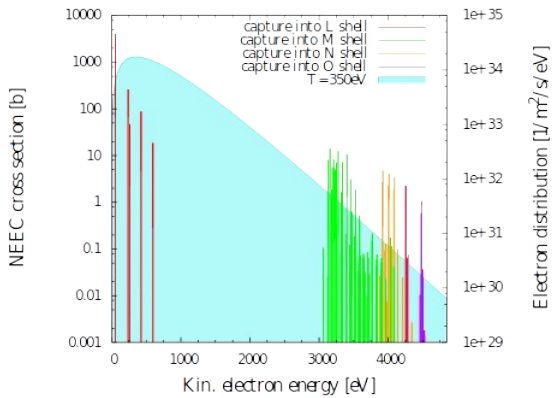
Electrons for NEEC

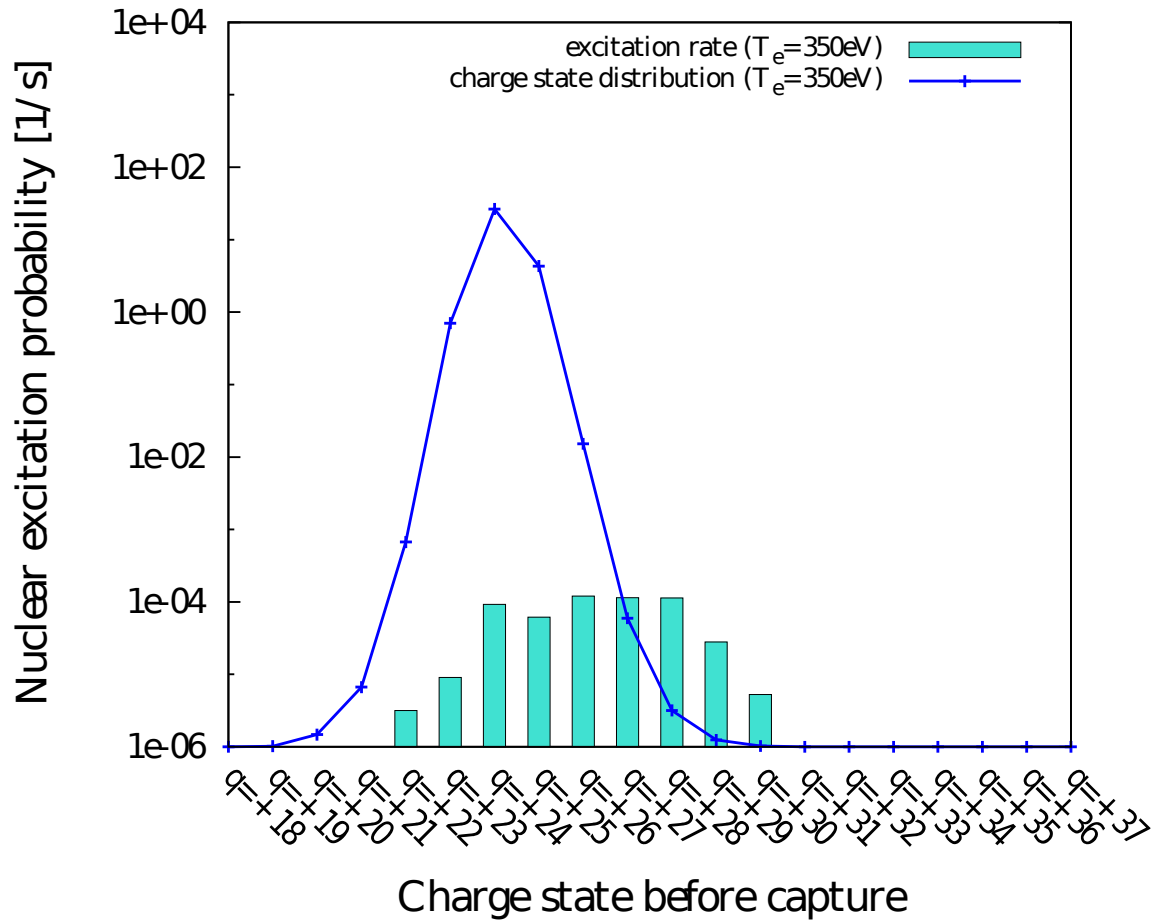




J. Gunst, Y. A. Litvinov, C. H. Keitel and AP, PRL 112, 082501 (2014)

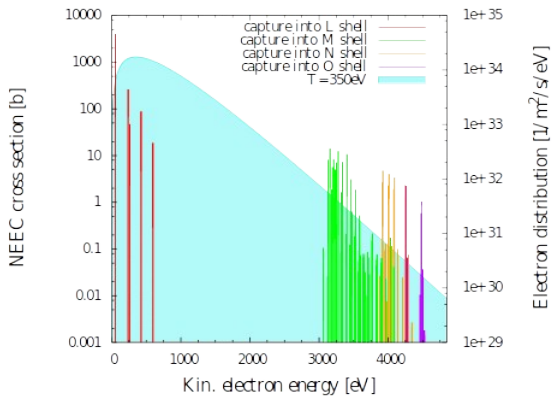
Electrons for NEEC

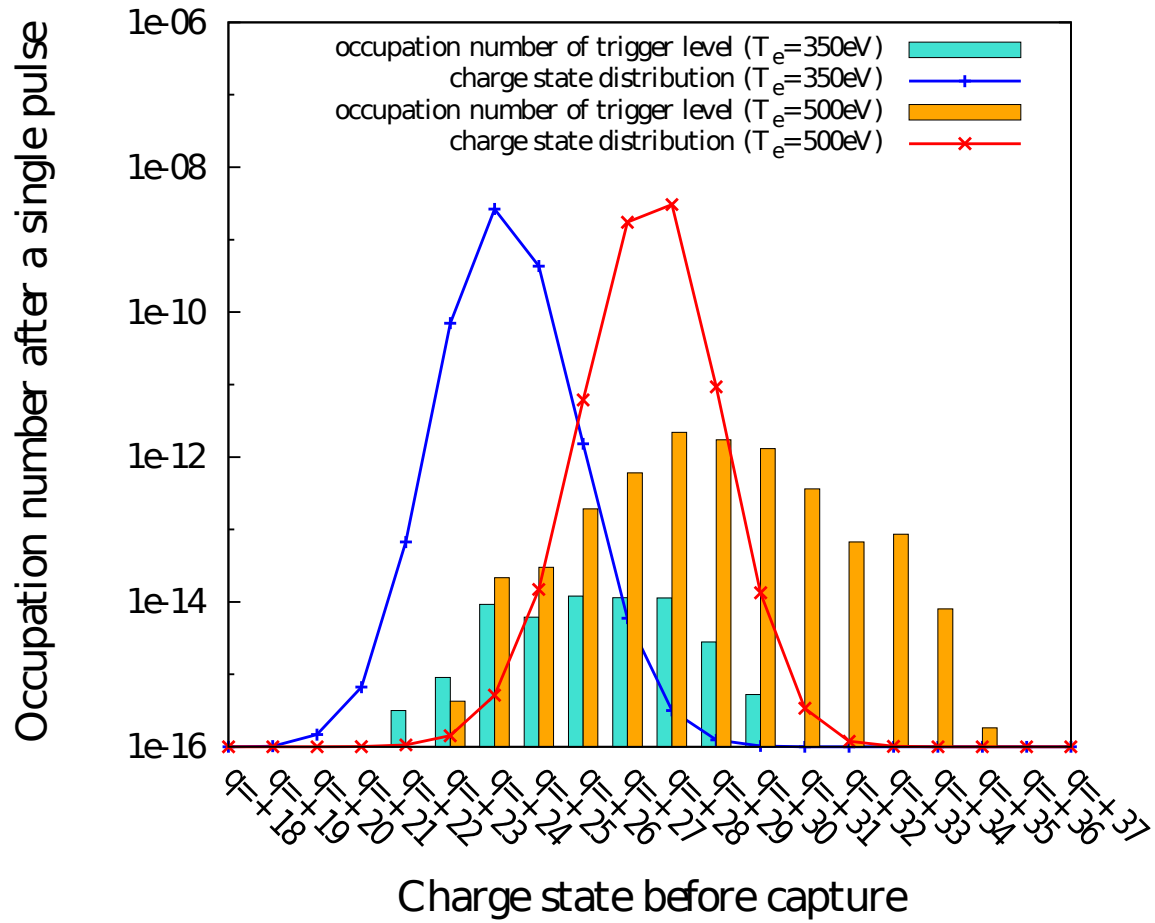




J. Gunst, Y. A. Litvinov, C. H. Keitel and AP, PRL 112, 082501 (2014)

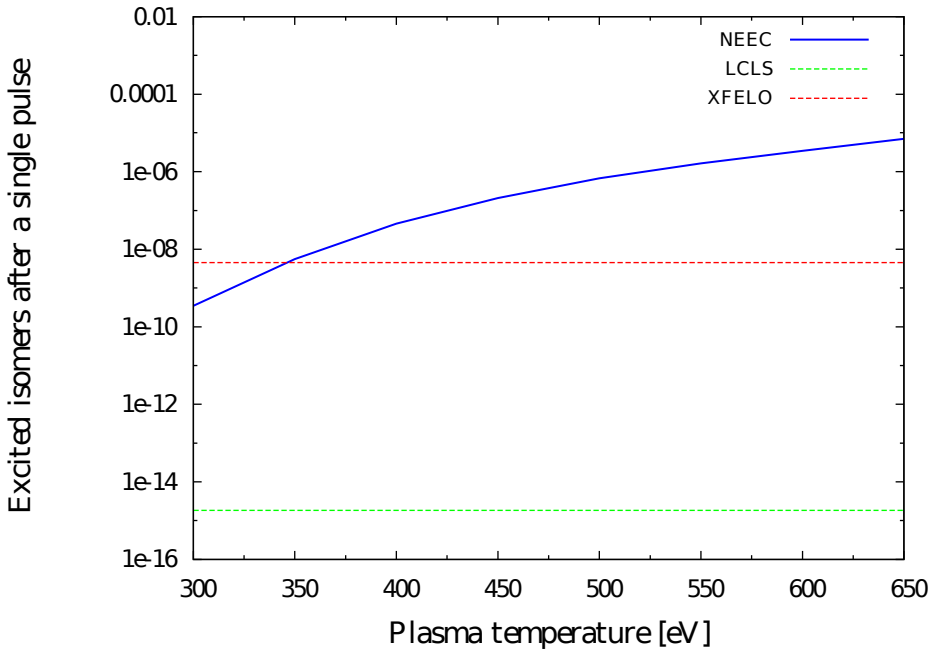
Electrons for NEEC





J. Gunst, Y. A. Litvinov, C. H. Keitel and AP, PRL 112, 082501 (2014)

Comparison



- coherent XFEL would be great
- uncertain transition energy complicates things
- secondary nuclear excitation dominates for most transitions available for XFEL

THINGS DON'T LOOK TOO GOOD FOR COHERENT CONTROL WITH XFEL!

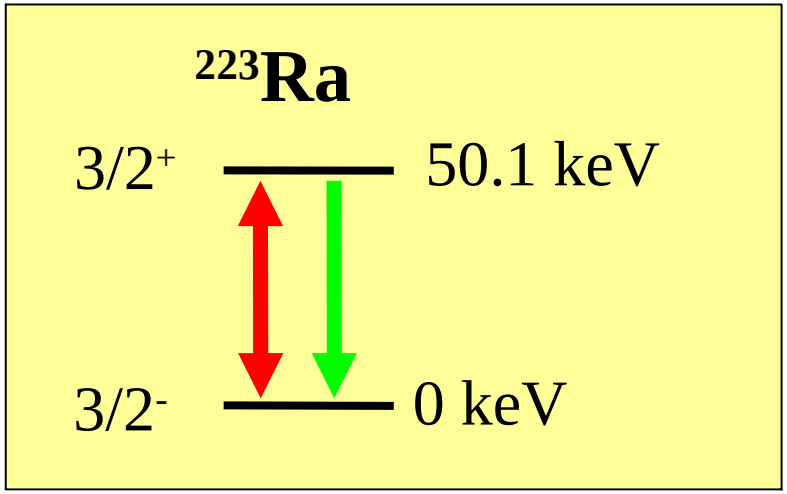
→ laser coherence/intensity not sufficient

→ secondary nuclear excitation – incoherent channel

Accelerated ion targets

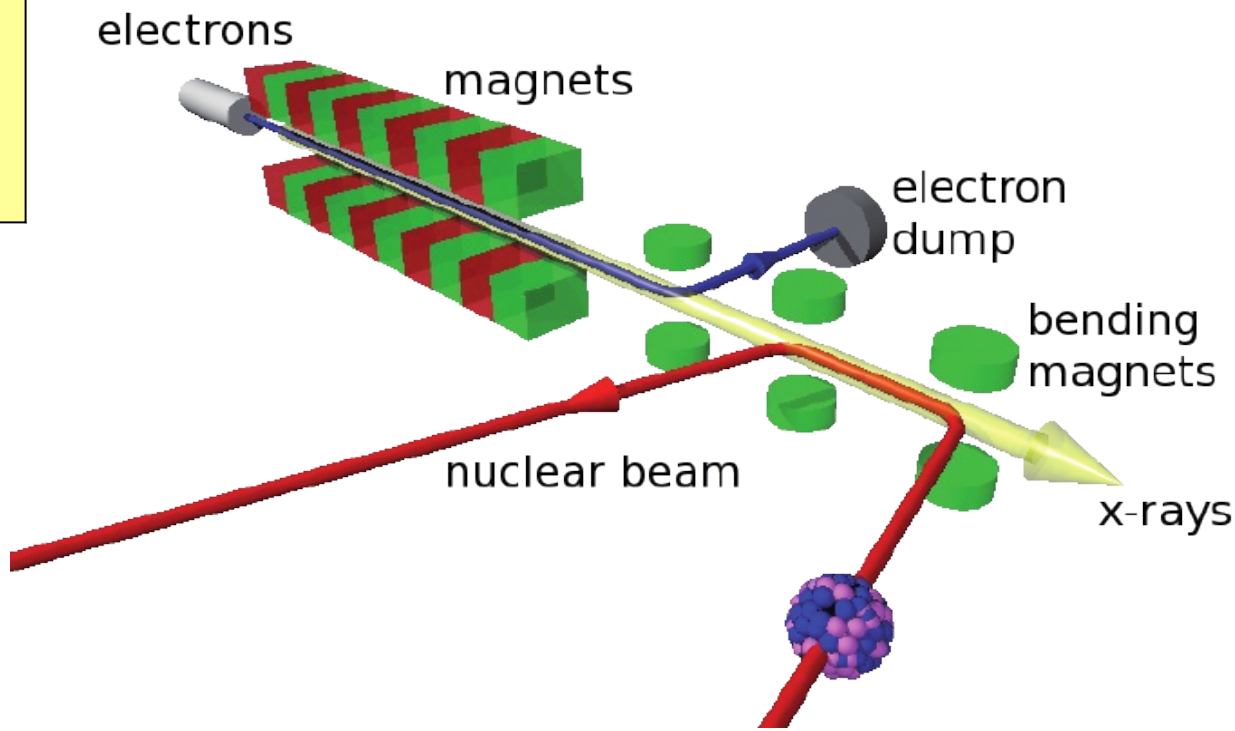
Nuclear excitations and the XFEL

bridge the gap between nuclear excitation and photon energies...



Doppler shift

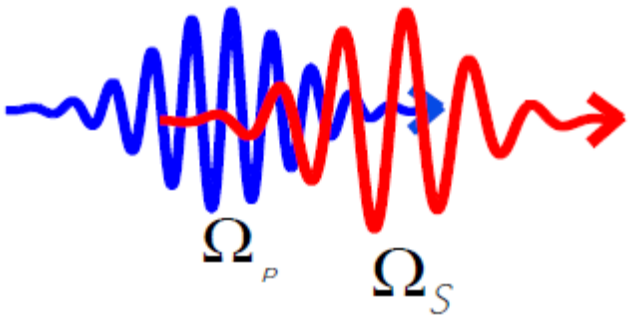
$$E_n = (1+\beta)\gamma E_L$$

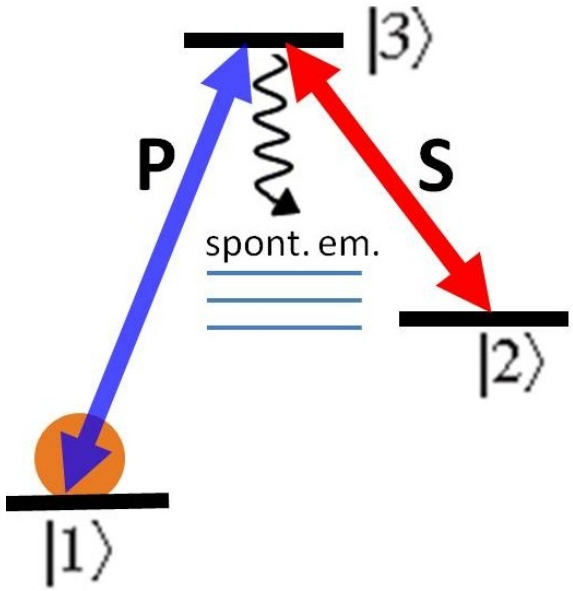


T. Bürvenich, J. Evers and C. H. Keitel, PRL 96, 142501 (2006)

STIRAP and the coherent XFEL

STImulated Raman Adiabatic Passage



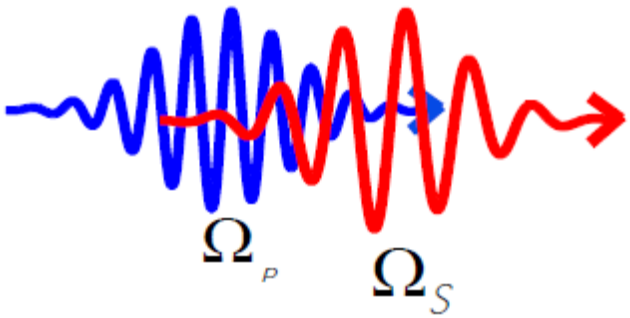


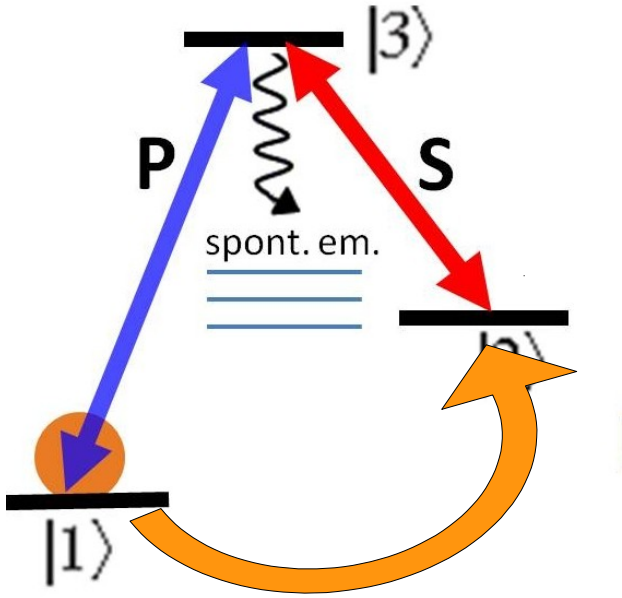
$$|D\rangle = \frac{\Omega_s}{\sqrt{\Omega_p^2 + \Omega_s^2}}|1\rangle - \frac{\Omega_p}{\sqrt{\Omega_p^2 + \Omega_s^2}}|2\rangle$$

K. Bergmann, H. Theuer, B.W. Shore, Rev. Mod. Phys. 70, 1003 (1998) 1003

STIRAP and the coherent XFEL

STImulated Raman Adiabatic Passage

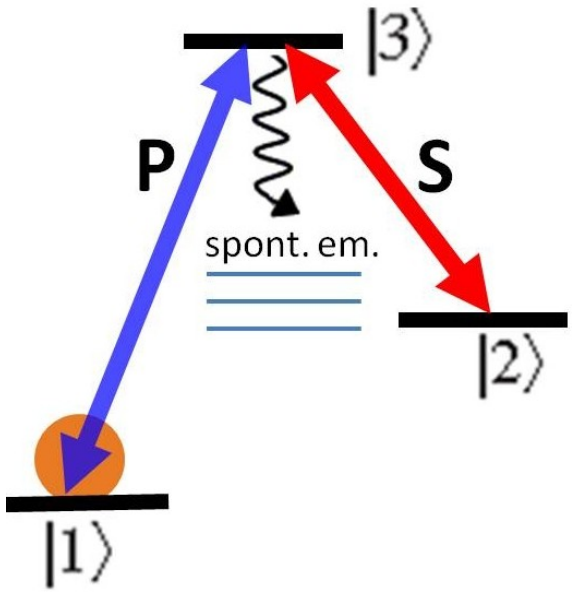


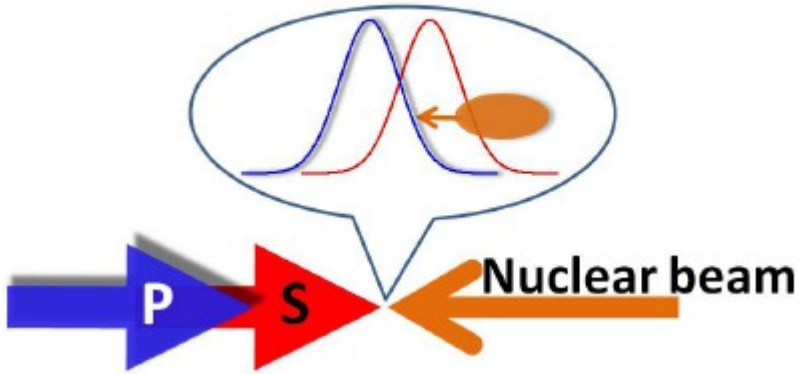


Robust coherent population transfer → **ISOMER TRIGGERING?**

$$|D\rangle = \frac{\Omega_s}{\sqrt{\Omega_p^2 + \Omega_s^2}}|1\rangle - \frac{\Omega_p}{\sqrt{\Omega_p^2 + \Omega_s^2}}|2\rangle$$

STIRAP and the coherent XFEL



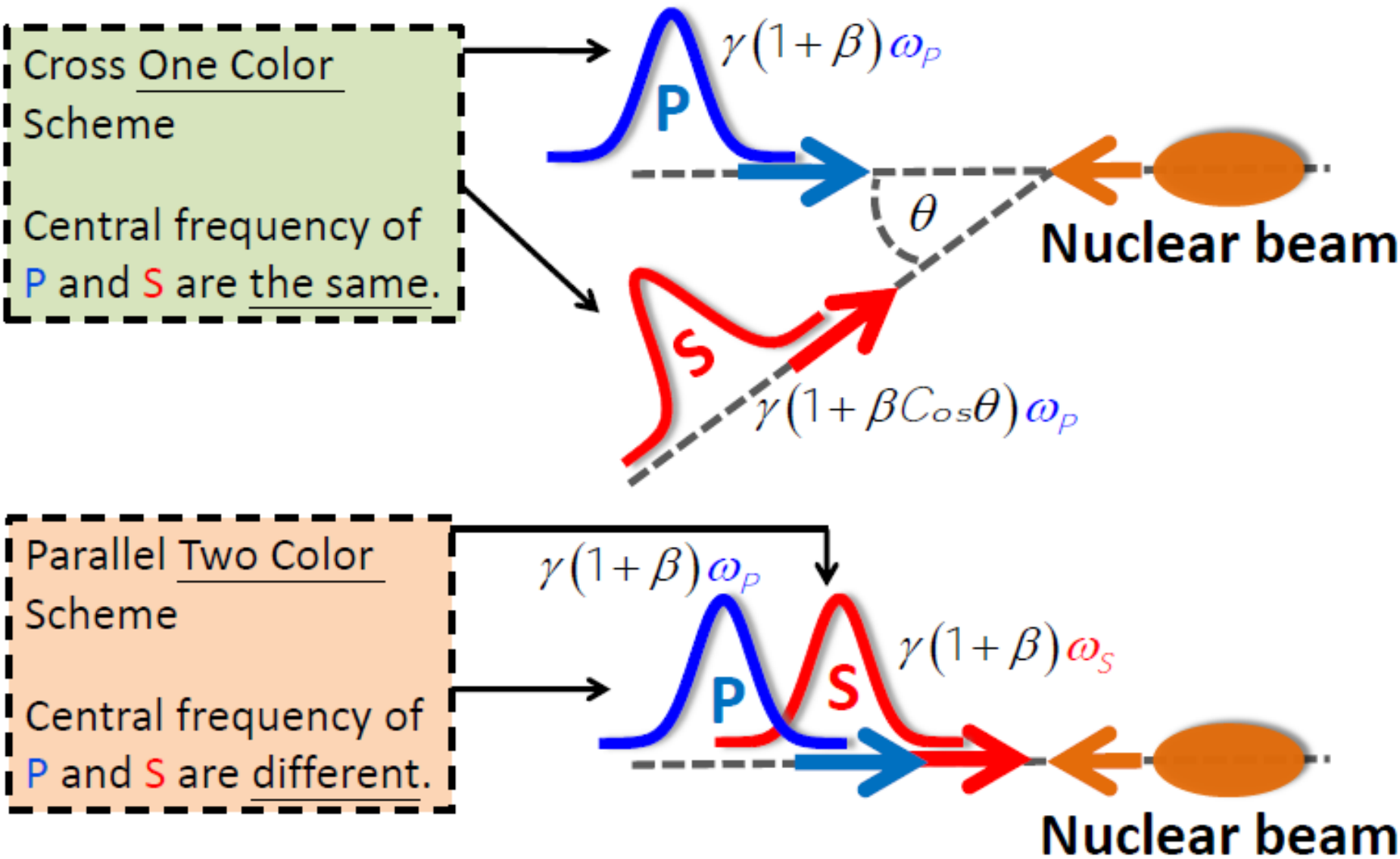


$$|D\rangle = \frac{\Omega_s}{\sqrt{\Omega_p^2 + \Omega_s^2}}|1\rangle - \frac{\Omega_p}{\sqrt{\Omega_p^2 + \Omega_s^2}}|2\rangle$$

Requirements:

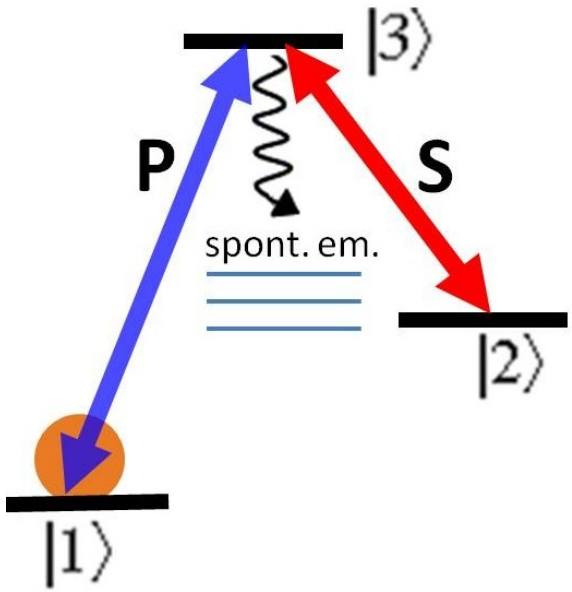
- **accelerated nuclei**
- **two-color**
- **full temporal coherence**

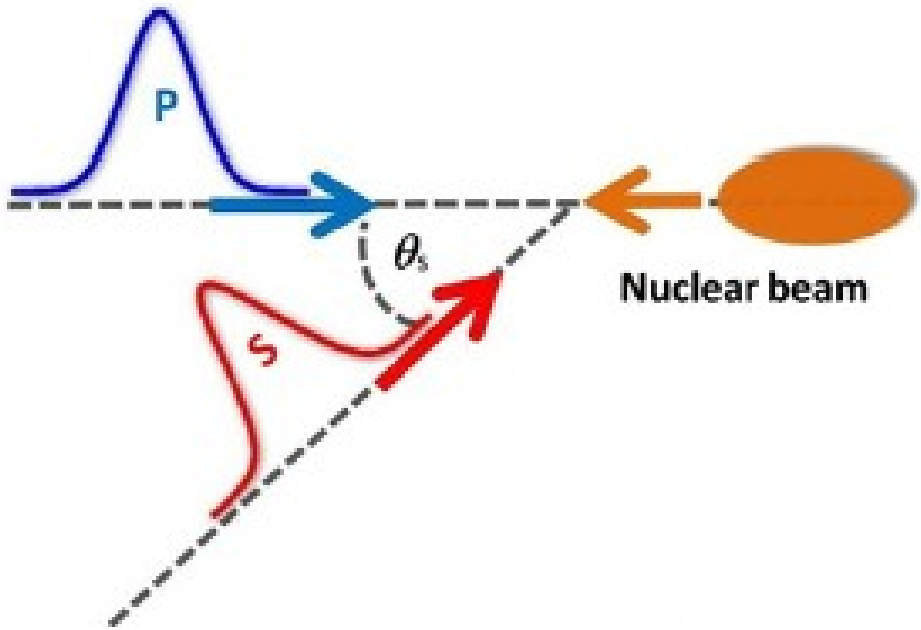
STIRAP and the coherent XFEL



W.-T. Liao, AP and C. H. Keitel, Phys. Lett. B 705, 134 (2011)

STIRAP and the coherent XFEL



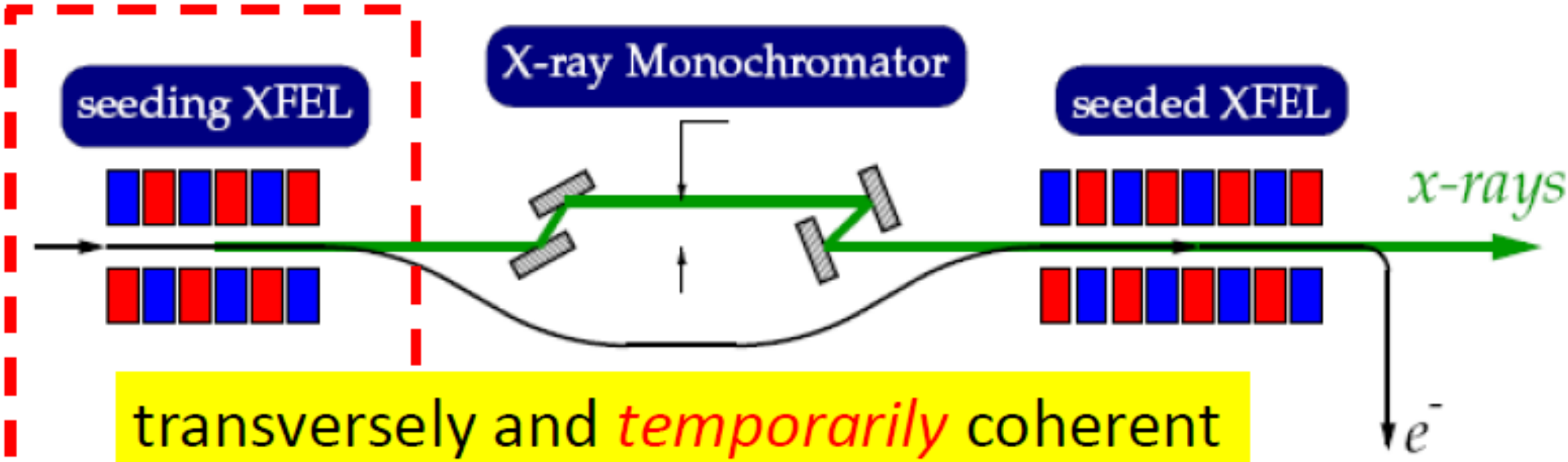


$$|D\rangle = \frac{\Omega_s}{\sqrt{\Omega_p^2 + \Omega_s^2}}|1\rangle - \frac{\Omega_p}{\sqrt{\Omega_p^2 + \Omega_s^2}}|2\rangle$$

Requirements:

- **accelerated nuclei**
- ~~**two-color**~~
- **full temporal coherence**

... seeded XFEL



transversely and *temporarily* coherent

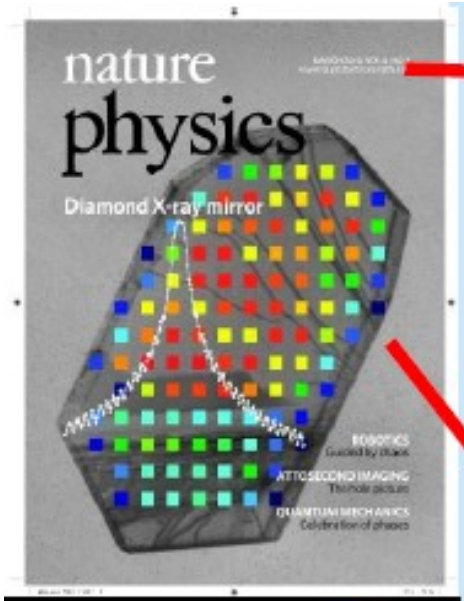
European XFEL

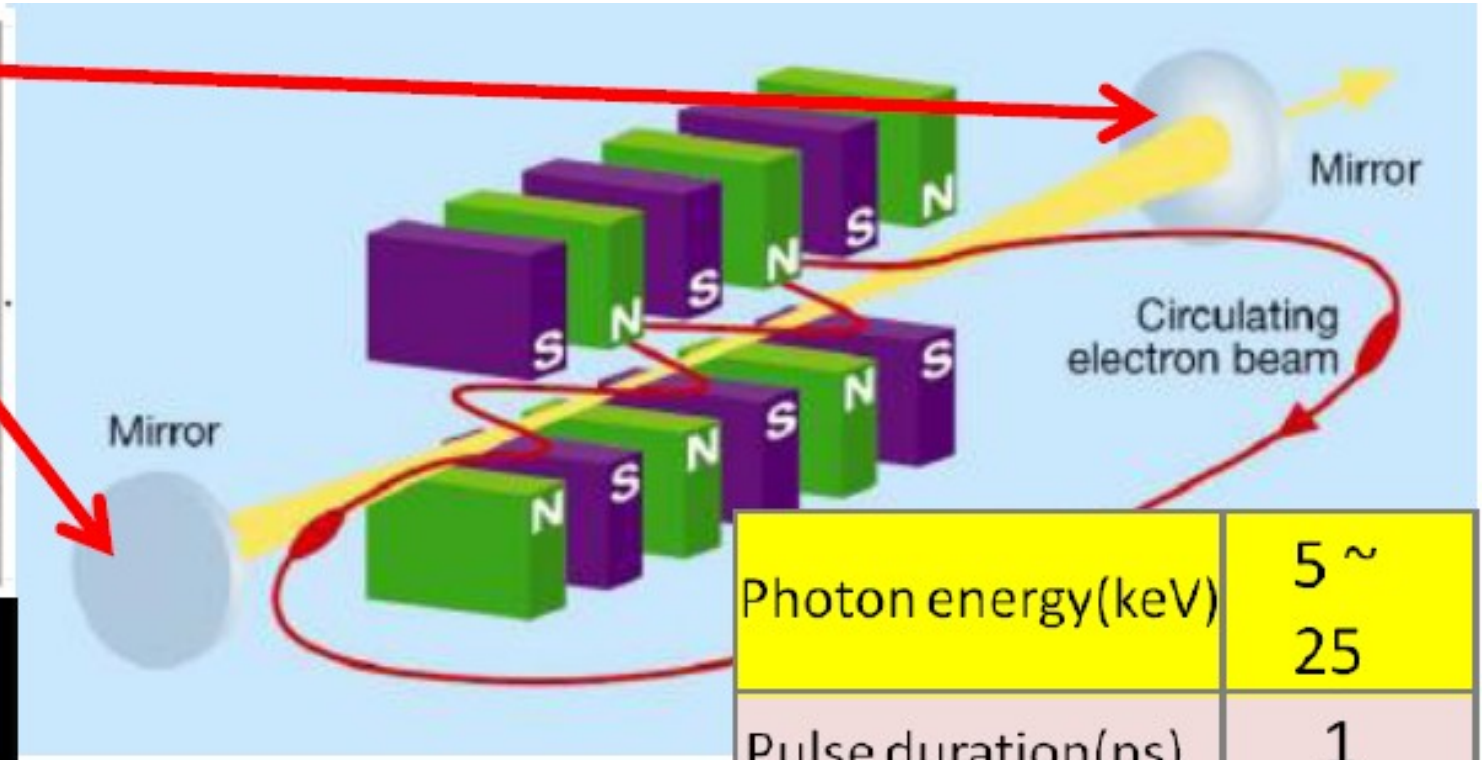
LCLS

Photon energy(keV)	0.2 ~ 12.4
Pulse duration(ps)	0.1
Band width (meV)	10

E. L. Saldin et al., **Nucl. Instrum. Meth. A 475, 357 (2001)**

... XFEL oscillator





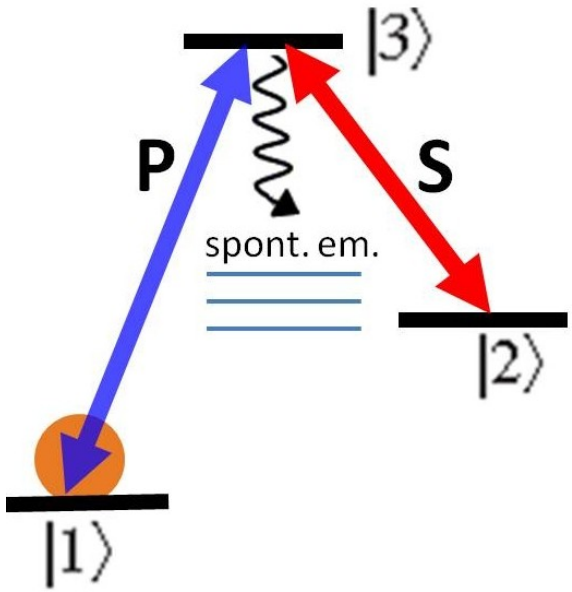
Photon energy(keV)	5 ~ 25
Pulse duration(ps)	1
Band width (meV)	2

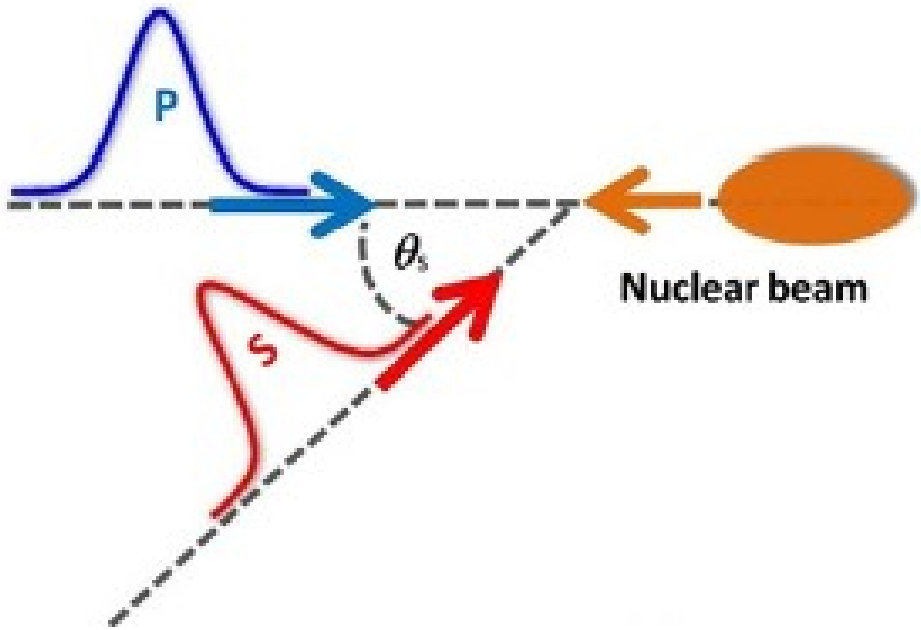
transversely and *temporarily* coherent

K.-J. Kim, Y. Shvyd'ko and S. Reiche, **PRL100, 244802 (2008)**

Y. Shvyd'ko et al., **Nature Phys. 6, 196 (2010)**

STIRAP and the coherent XFEL





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Requirements:

- **accelerated nuclei**
- ~~**two-color**~~
- **Coherence: SXFEL, XFELO**

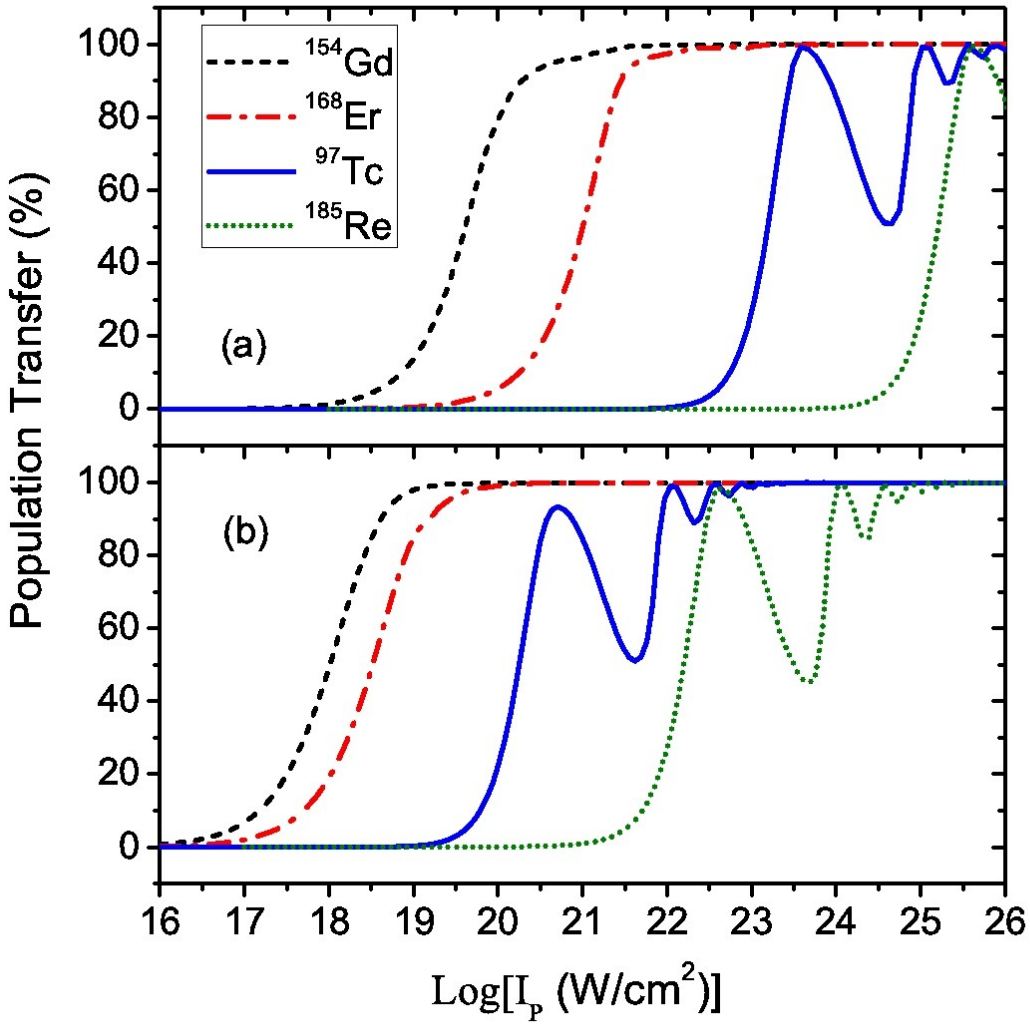
STIRAP and the coherent XFEL

Seeded XFEL

XFELO

TABLE I. The nuclear data.

Element	Line width of |3> (meV)	E_3 (keV)	E_2 (keV)	E_1 (keV)
^{185}Re	0.04	284.200	125.359	0
^{97}Tc	0. 60	656.900	324.476	96.57
^{154}Gd	300.00	1241.291	123.071	0
^{168}Er	130.00	1786.123	79.804	0



W.-T. Liao, AP and C. H. Keitel, Phys. Lett. B 705, 134 (2011)

STIRAP and the coherent XFEL

TABLE II. The nuclear data.

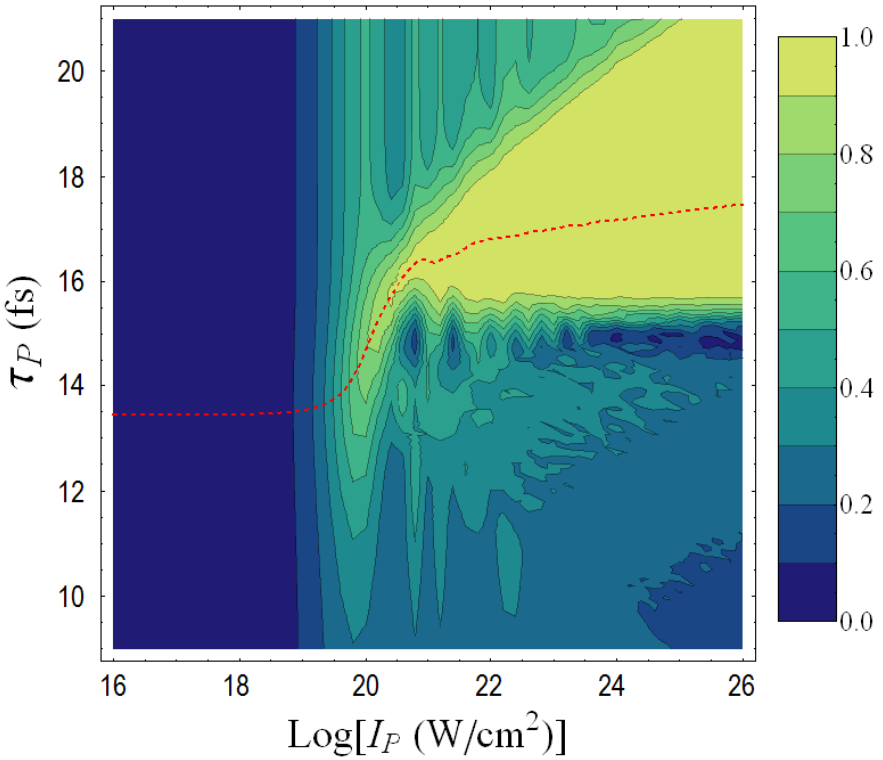
Element	(a) Seeded XFEL				(b) X-FELO			
	γ	Stokes angles (rad)	$\frac{I_p^{eff}}{I_p}$	$\frac{I_S^{eff}}{I_S}$	γ	Stokes angles (rad)	$\frac{I_p^{eff}}{I_p}$	$\frac{I_S^{eff}}{I_S}$
^{185}Re	11.482	1.45440	0.085	0.048	5.728	1.45963	0.211	0.118
^{97}Tc	22.605	1.38357	2.710	1.608	11.229	1.38482	6.722	3.988
^{154}Gd	50.057	0.64069	2963.200	2669.410	24.836	0.64079	2465.290	2000.670
^{168}Er	72.025	0.42598	1876.080	1792.260	35.730	0.42601	4652.680	4444.800

ISSUES:

geography

ion beam resolution

ion beam focus



W.-T. Liao, AP and C. H. Keitel, Phys. Rev. C 87, 054609 (2013)

STIRAP and the coherent XFEL

Seeded XFEL

XFELO

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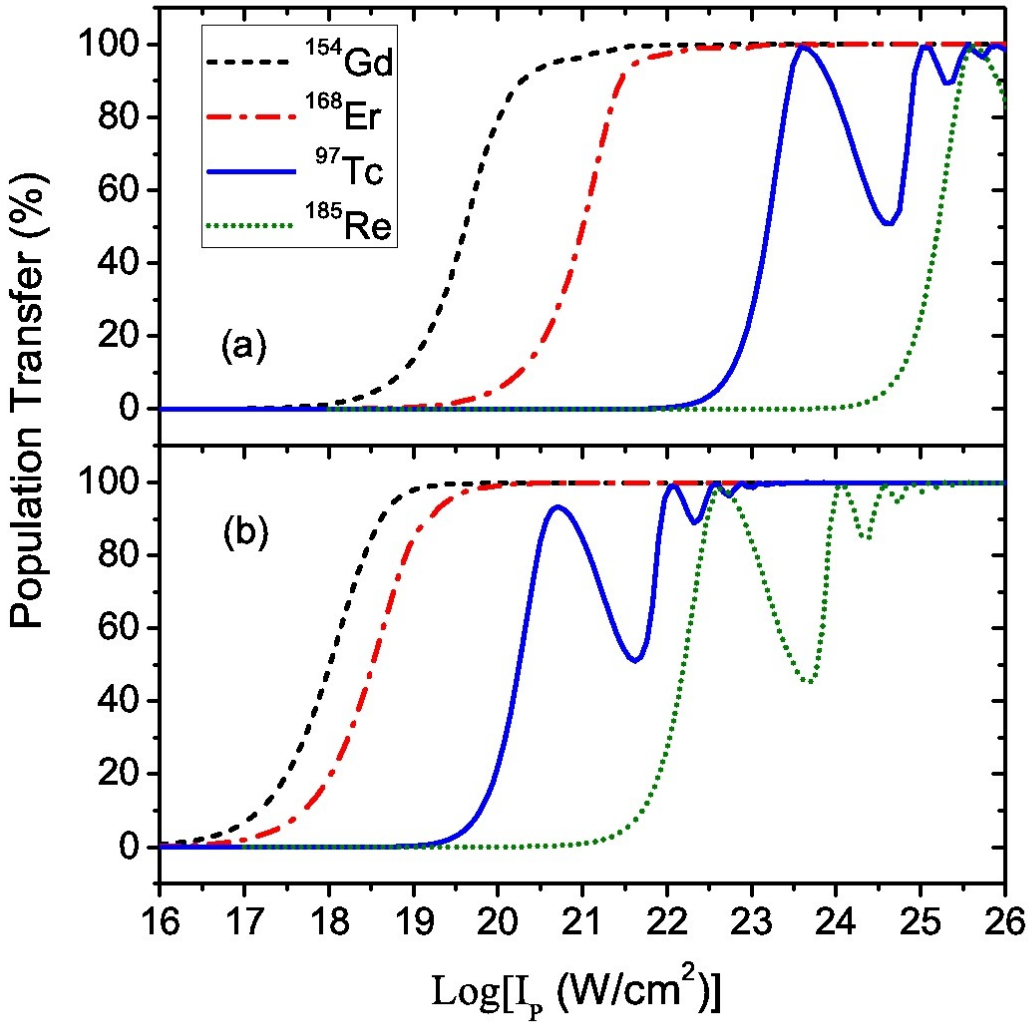

W.-T. Liao, AP and C. H. Keitel, Phys. Lett. B 705, 134 (2011)

Conclusions

I. Photon-hungry quantum optics with nuclei

the source: XFEL
the nuclei: tricky
theory: density matrix
coherence vs. intensity
solid-state targets
accelerated ion targets
STIRAP

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

TODAY

- Indeed photon hungry because of very narrow resonances!
- Temporal coherence of source is crucial
- Difficult to achieve the coherent control regime for nuclei!

Conclusions

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TODAY

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COMPLETELY DIFFERENT APPROACH TOMORROW!

TOMORROW

Conclusions

TODAY

I. Photon-hungry quantum optics with nuclei

the source: XFEL
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theory: density matrix
coherence vs. intensity
solid-state targets
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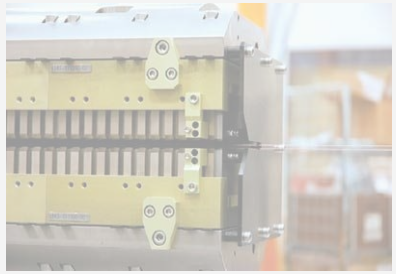
STIRAP



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

THANK YOU FOR YOUR ATTENTION AND SEE YOU TOMORROW!

TOMORROW

- Indeed photon hungry because of very narrow resonances!
- Temporal coherence of source is crucial
- Difficult to achieve the coherent control regime for nuclei!

COMPLETELY DIFFERENT APPROACH TOMORROW!

X-ray nuclear quantum optics II

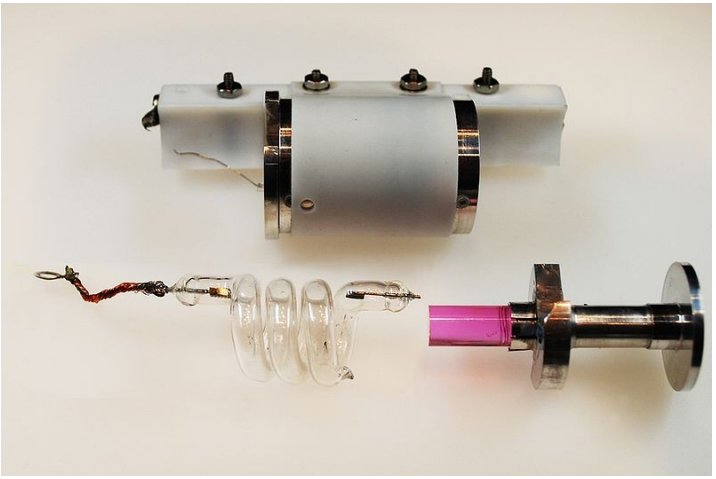
Adriana Pálffy
palffy@mpi-hd.mpg.de
Max Planck Institute for Nuclear Physics, Heidelberg, Germany

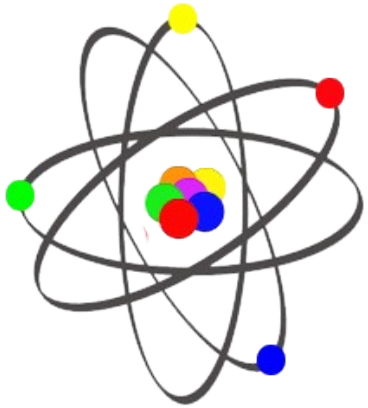
PIER Helmholtz Graduate School

Hamburg, 6-7 October 2014

Historical perspective

1960, T. Maiman, first successful laser

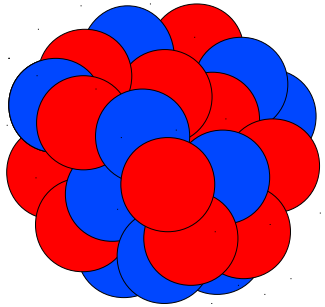




Lasers mostly OPTICAL, based on and used to control ATOMS

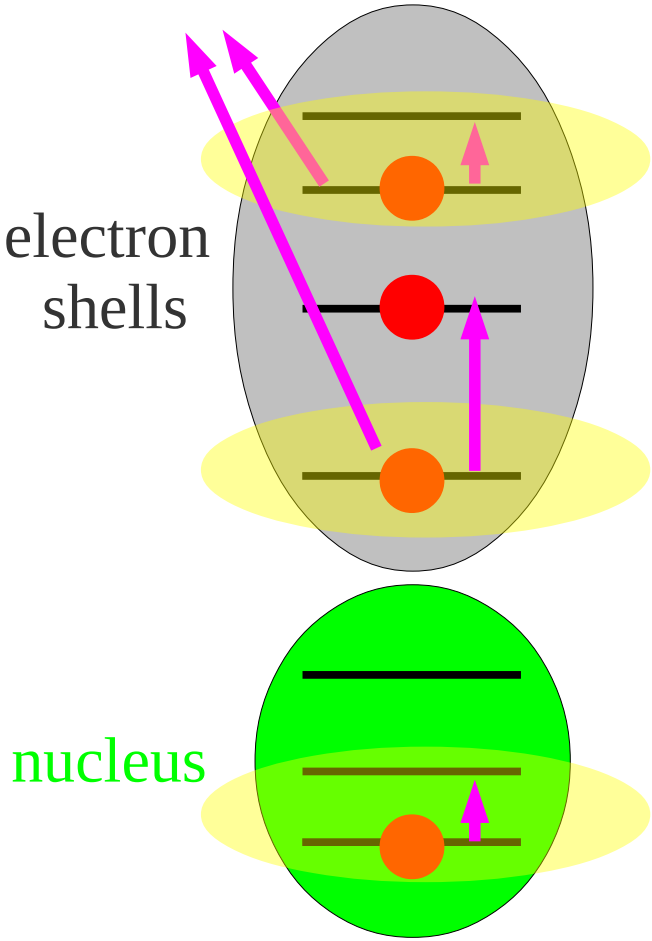
Historical perspective

since 2009, XFEL



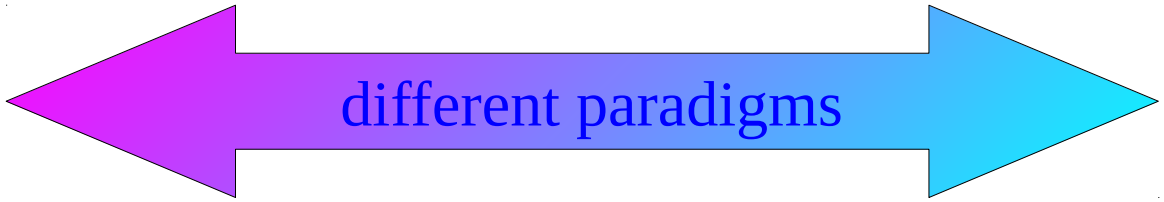
Driving and control of NUCLEAR transitions?
Mutual control?

X-ray incentives



courtesy Jörg Evers

Light-matter interactions



uncontrolled pump
+ passive observation

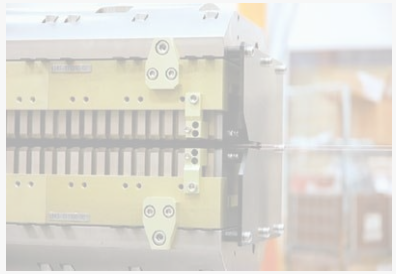
full quantum
control

- X-ray physics could greatly benefit from moving towards quantum/non-linear/control
- New light sources and upgrades now and in near future → now is the right time
- Such developments are necessary to fully exploit potential of new light sources

Outline

I. Photon-hungry quantum optics with nuclei

the source: XFEL
the nuclei: tricky
theory: density matrix
coherence vs. intensity
solid-state targets
accelerated ion targets
STIRAP

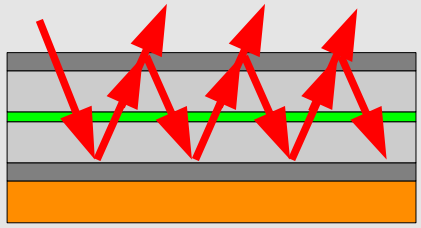


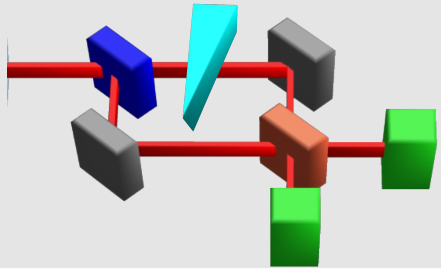
$$\frac{d}{dt}\rho = -\frac{i}{\hbar}\left[H,\rho\right] + \mathcal{L}[\rho]$$

YESTERDAY

II. Single-photon quantum optics with nuclei

the source: SR
the nuclei: Mössbauer's ^{57}Fe
theory: Dicke, Maxwell-Bloch
nuclear forward scattering
nuclear waveguide cavities
storing a single photon
entanglement and slow light





TODAY

Previous experiences ... DISCUSSED YESTERDAY

- scaling in frequency from optical lasers + atoms

*coherent control**, *non-linear optics,*
dark states, slow light, entanglement,
electromagnetically induced transparency,
revolution in atomic physics, etc.

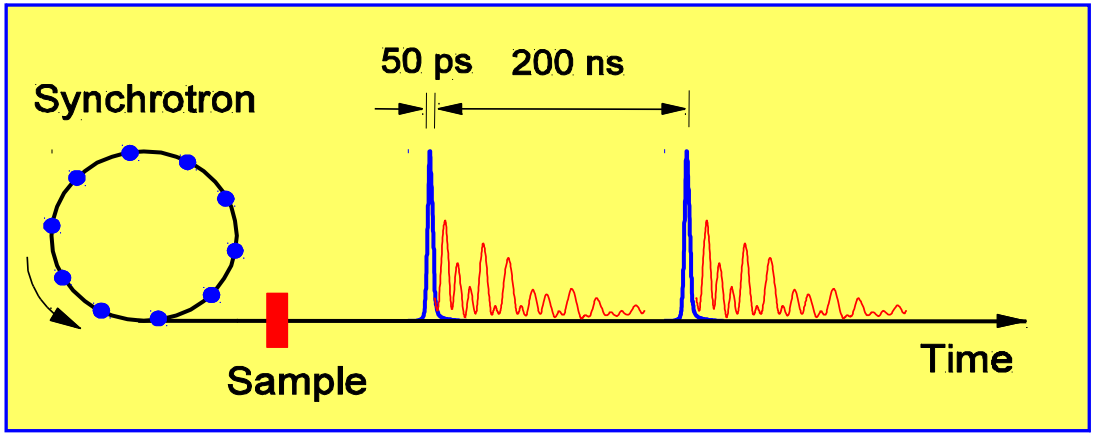
* relies on driving and control over most of the atomic state population!

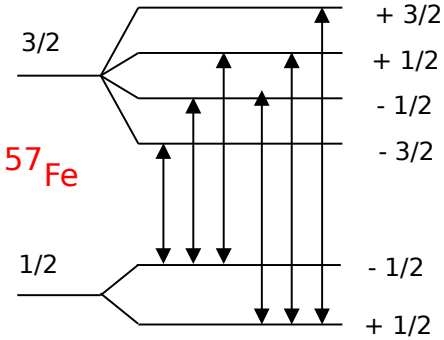
CAN WE ACHIEVE SOMETHING SIMILAR WITH NUCLEI AND X-RAYS?

→ WE'LL NEED A LOT OF RESONANT PHOTONS FOR THAT!!!

Previous experiences … TODAY'S TOPIC!

• scaling in brilliance from synchrotrons + nuclei





@ 14.413 keV

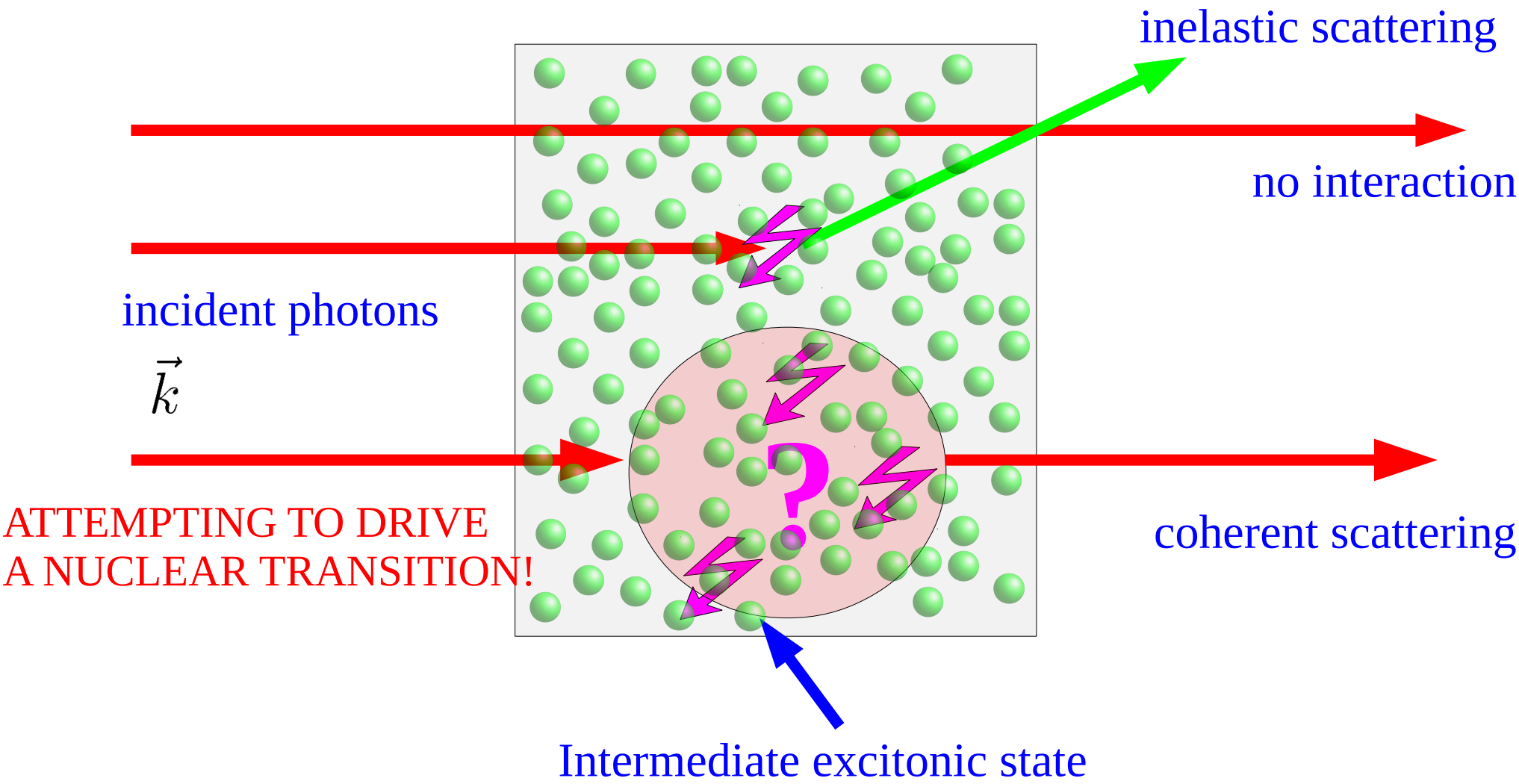
Nuclear Forward Scattering (NFS) of Synchrotron Radiation

nuclear condensed matter physics based on the Mössbauer effect,
*control of nuclear decay for ensembles of nuclei**, *storing single x-ray photons*

* relies on weak excitation, a single nucleus only!

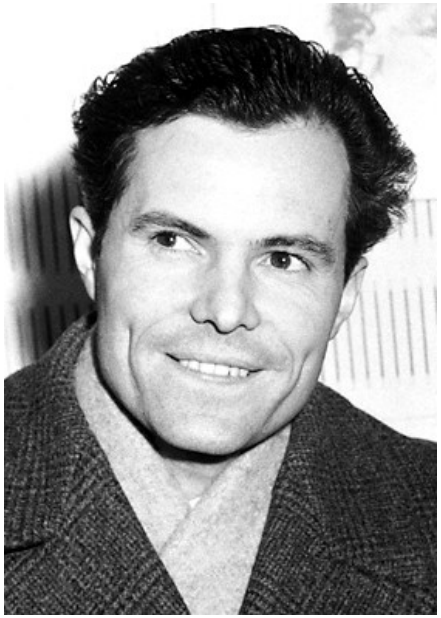
Nuclear resonance scattering

Nuclear resonance scattering



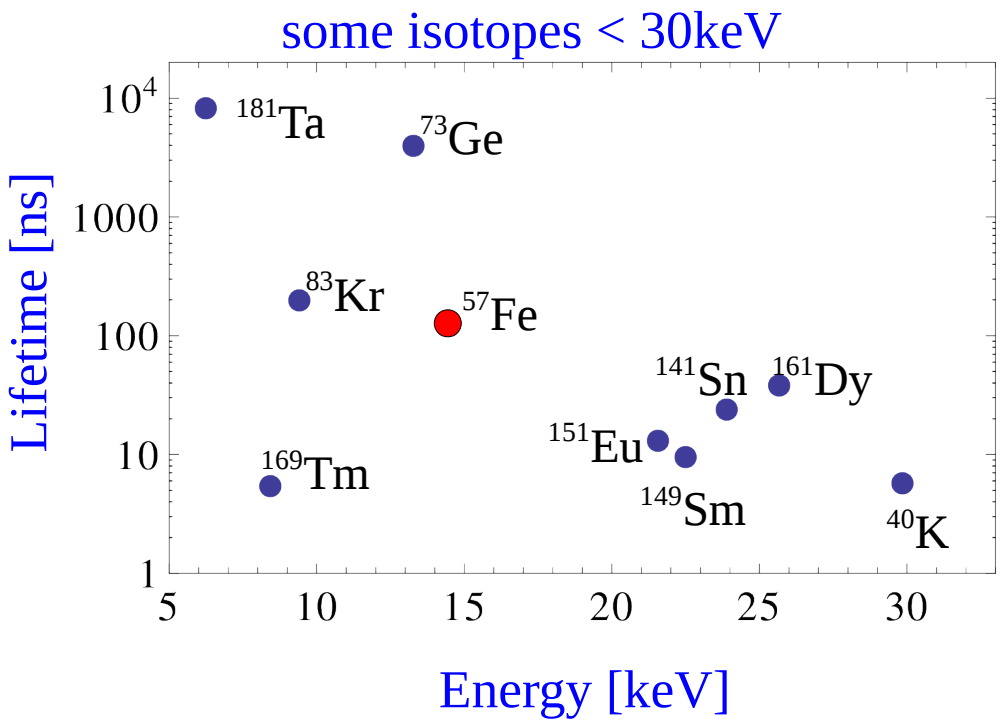
Nuclear resonance scattering

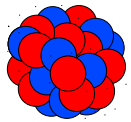
- Tool to investigate magnetic, structural and dynamic properties of matter
- Small linewidth of nuclear resonances (μeV-peV) is both essential feature and technical challenge
- Mössbauer effect leads to recoilless absorption and emission



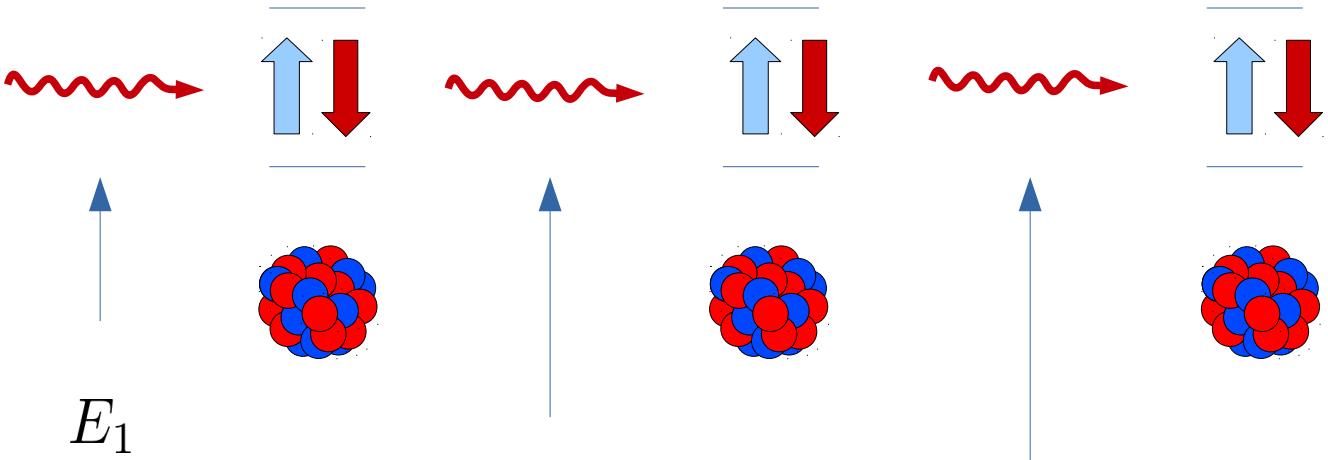
R. Mössbauer
1955-57 phd work
in Heidelberg

(image from Wikipedia)





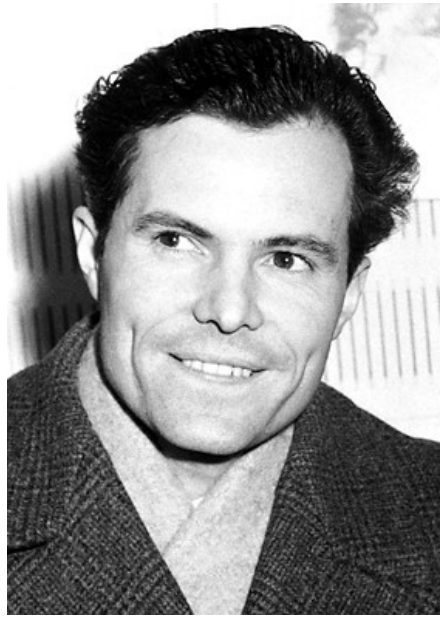
Recoilless nuclear transitions



E_1

$$E_2 = E_1 - 2E_{rec}$$

$$E_3 = E_2 - 2E_{rec}$$



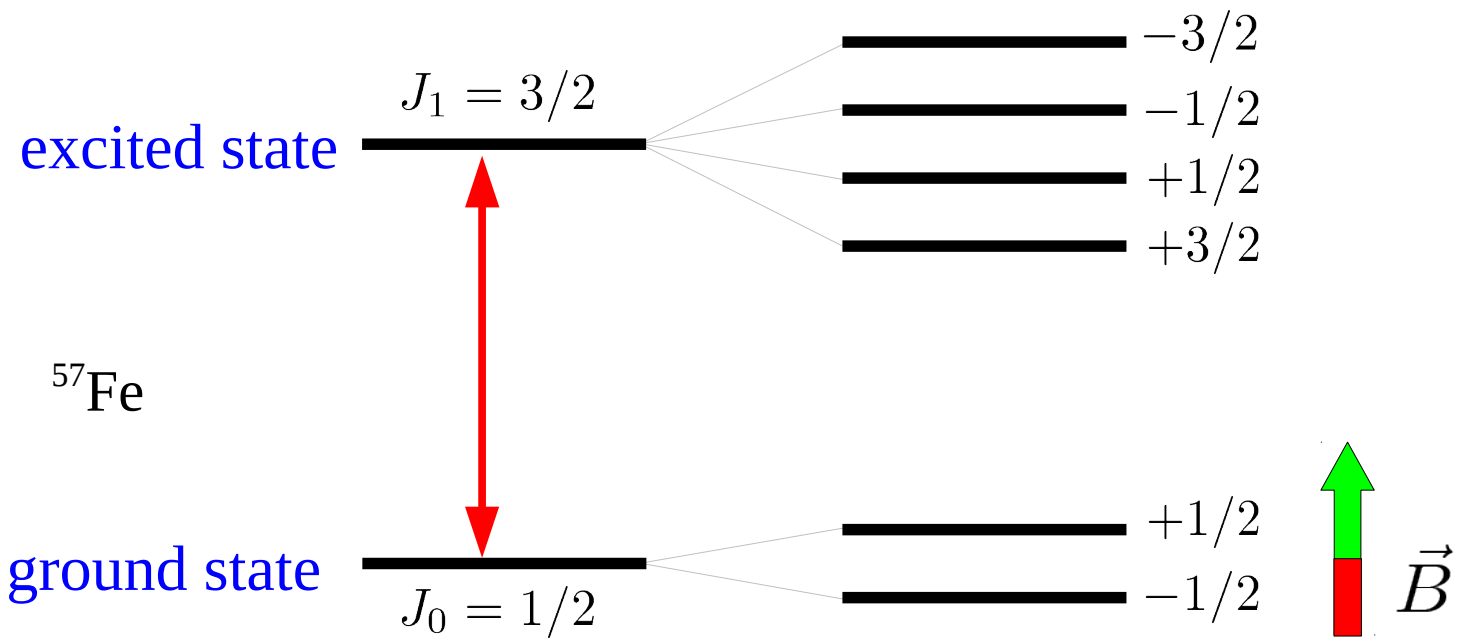
→ reabsorbtion of the photon works only if $2E_{rec} < \Gamma$ and $E_{rec} < \Gamma$

Mössbauer effect:

in certain solid-state targets, the recoil is distributed over the whole lattice!

$$E_{rec} \sim \frac{1}{M} \sim 0$$ → REABSORPTION WORKS!!!

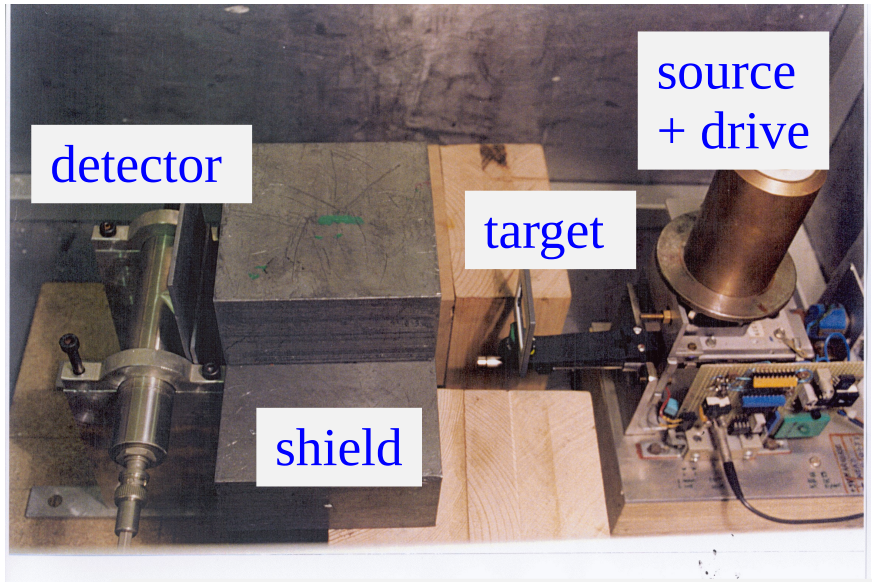
The iron age



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

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

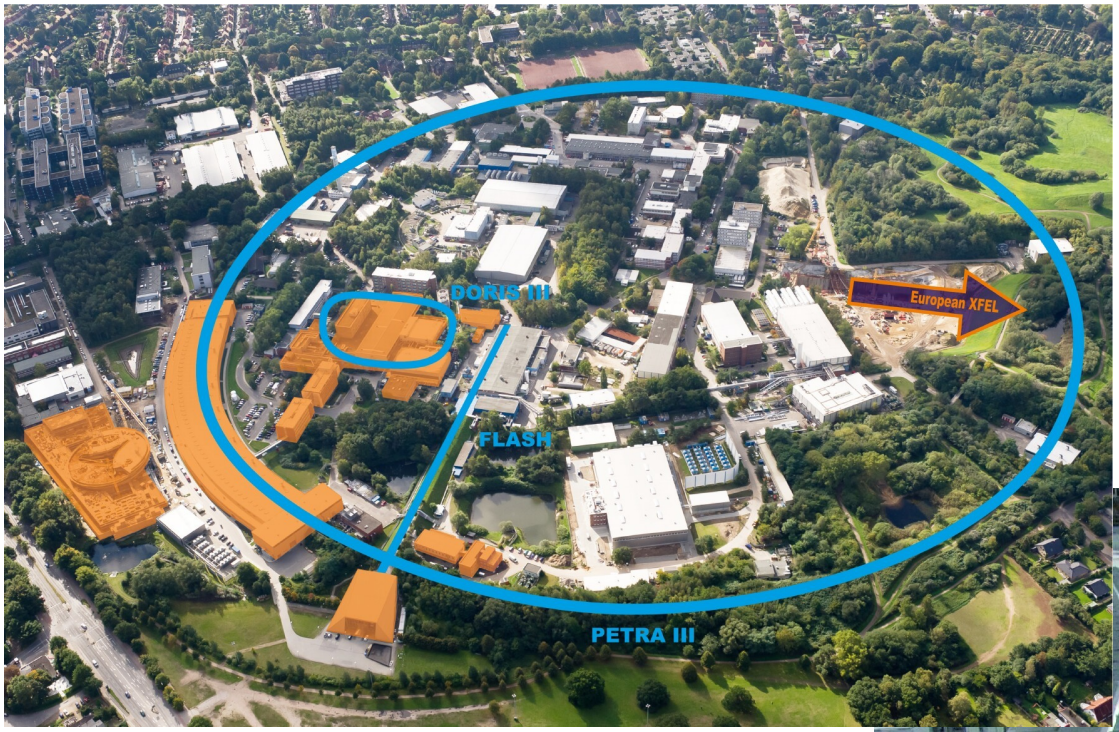
Mössbauer traditional setup

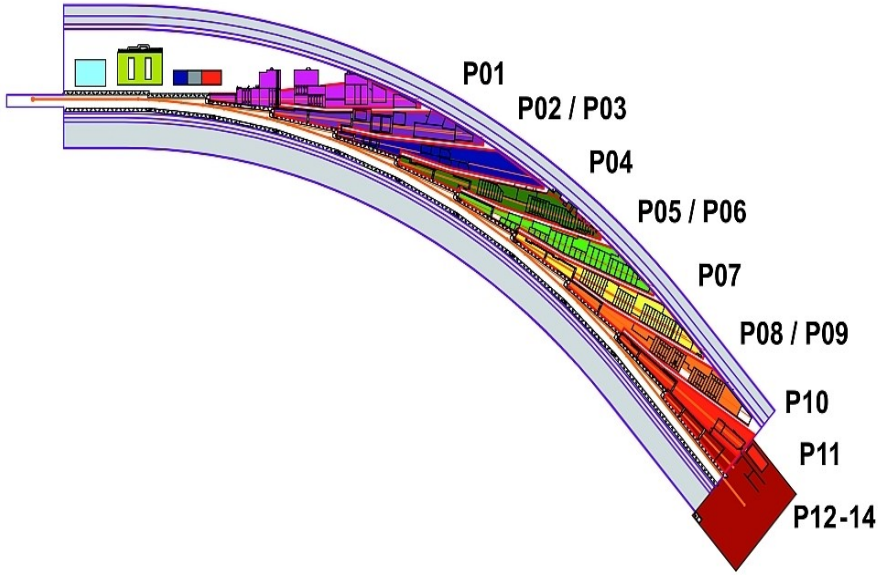


Student lab in Milan

courtesy Jörg Evers

The upgrade

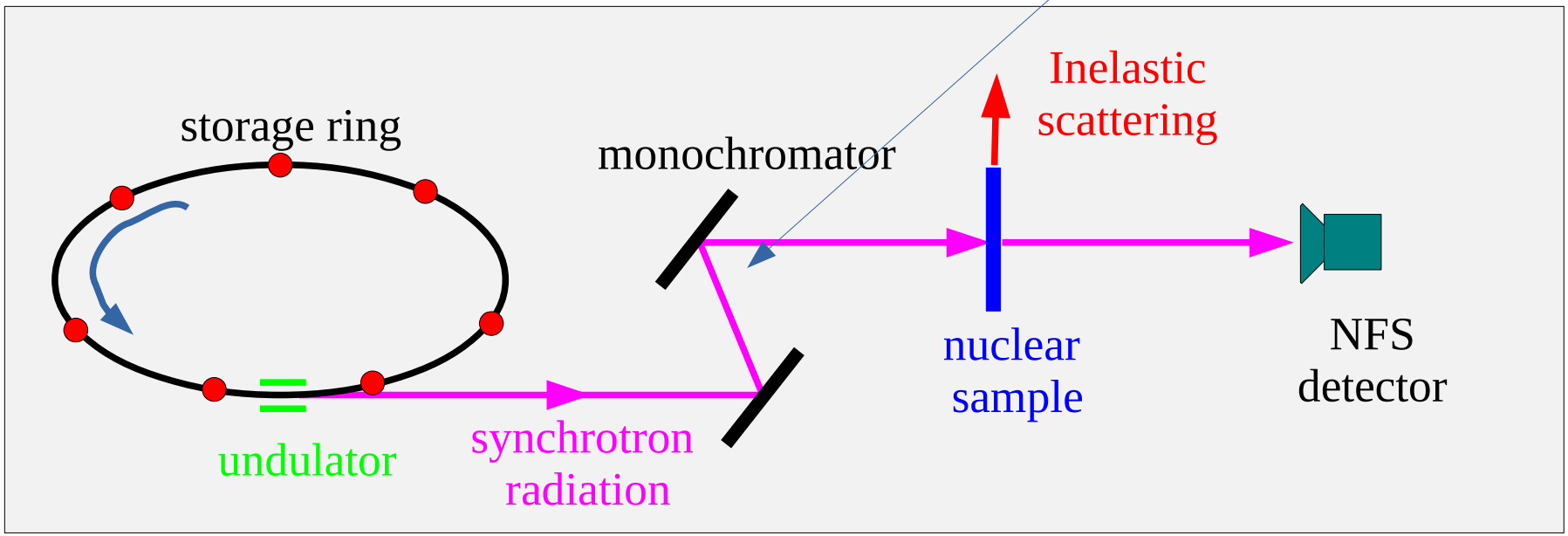




Images: DESY

Nuclear forward scattering

filtering out photons
with meV resolution

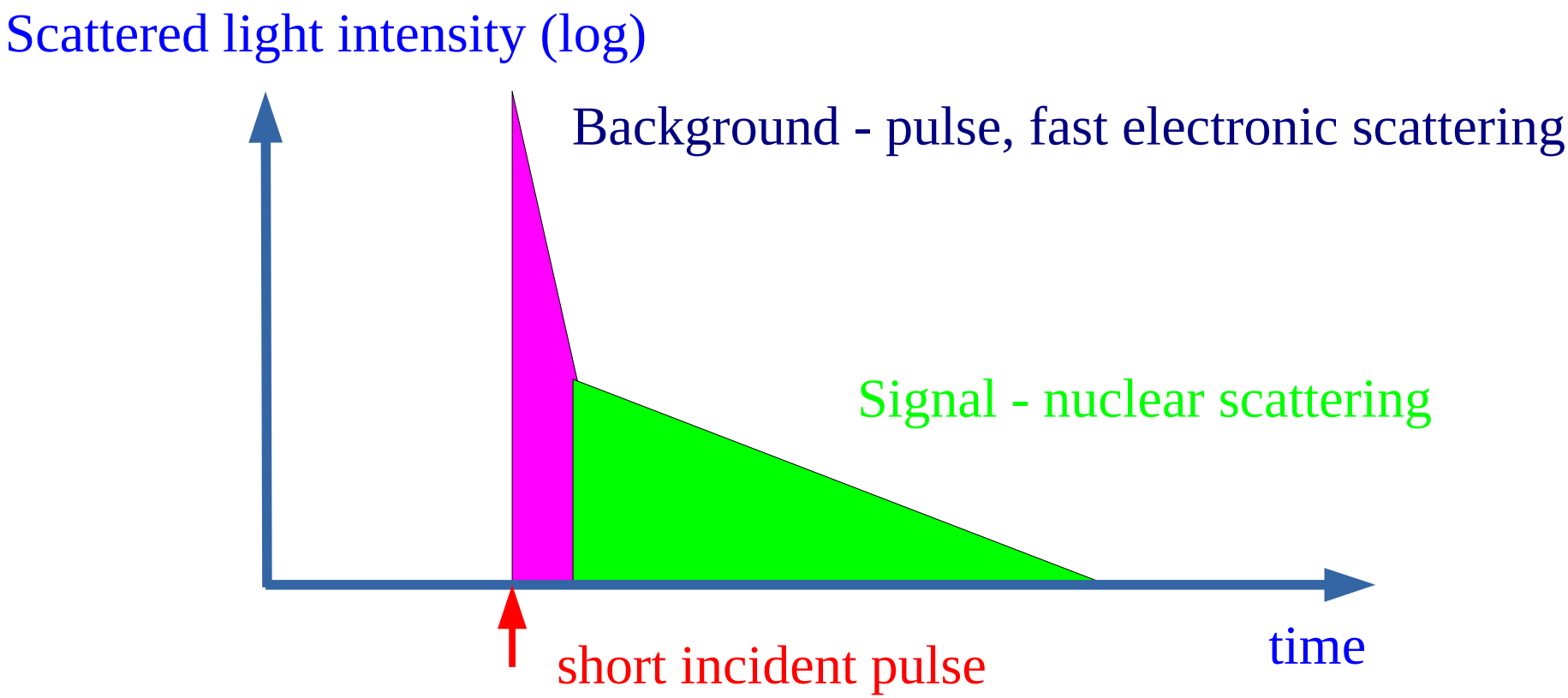


MHz repetition rate
10^9 photons/s after monochromator!
nuclear width approx. 5 neV

substantially less photons
per pulse, but better
repetition rate than XFEL!

WEAK EXCITATION – A SINGLE RESONANT PHOTON PER PULSE AT MOST!

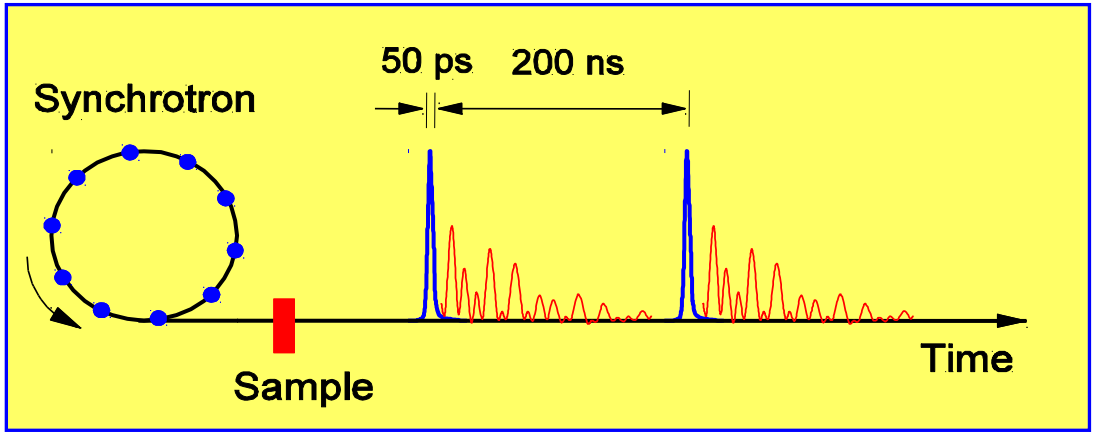
Nuclear forward scattering

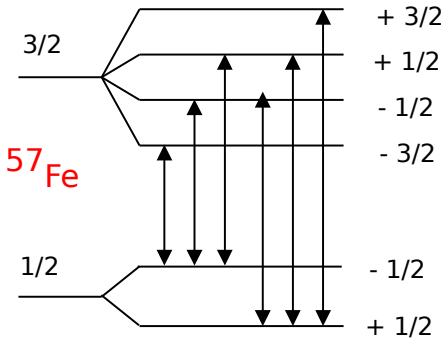


- Nuclear scattering has delayed tail on time scale 1/Γ
  Example (^{57}Fe): 141 ns
- This allows to measure almost background-free using time gating

Nuclear forward scattering

DETECT THE SCATTERED SIGNAL IN TIME

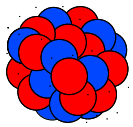




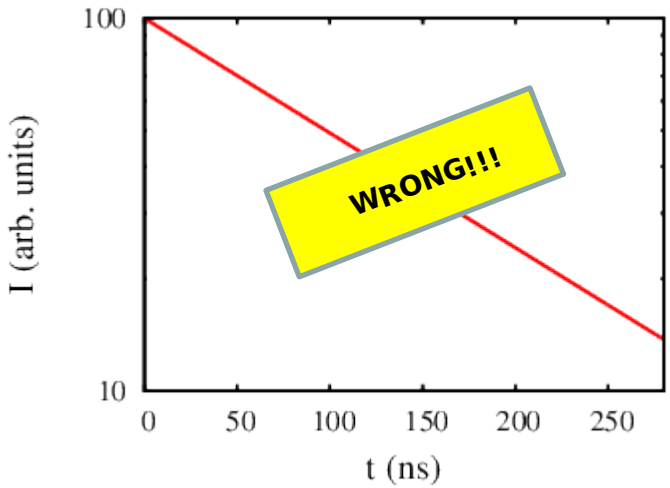
@ 14.413 keV

Nuclear forward scattering via recoilless transitions

At most, a single nucleus is excited!

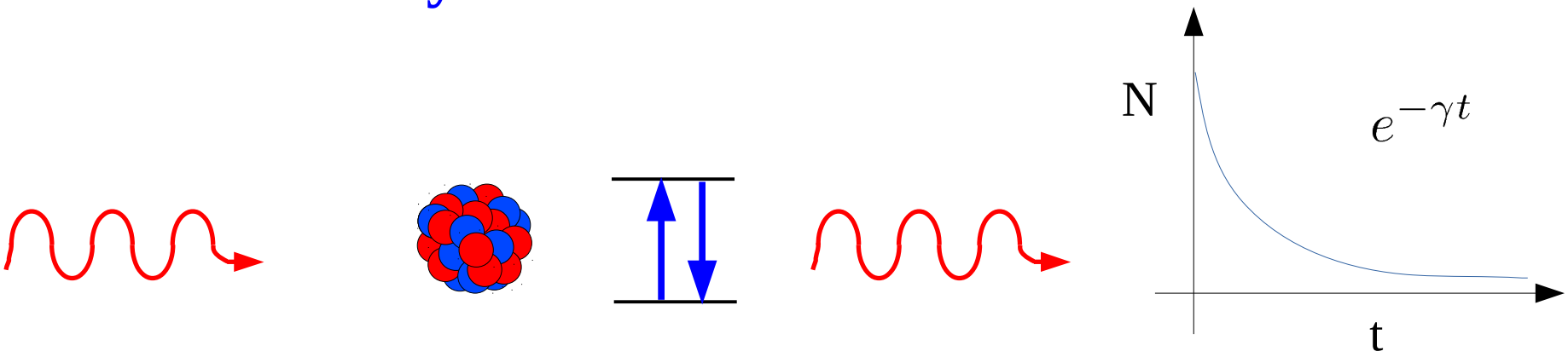


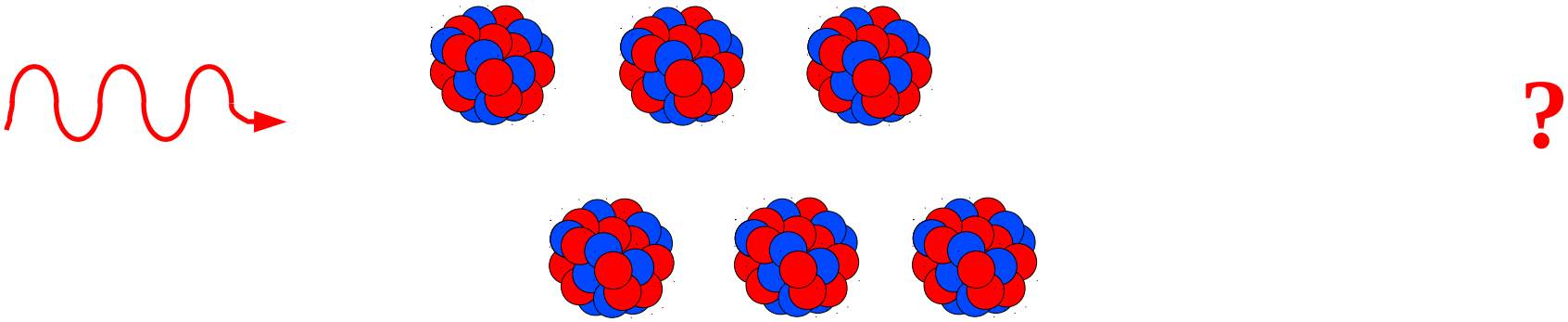
Expect exponential decay?



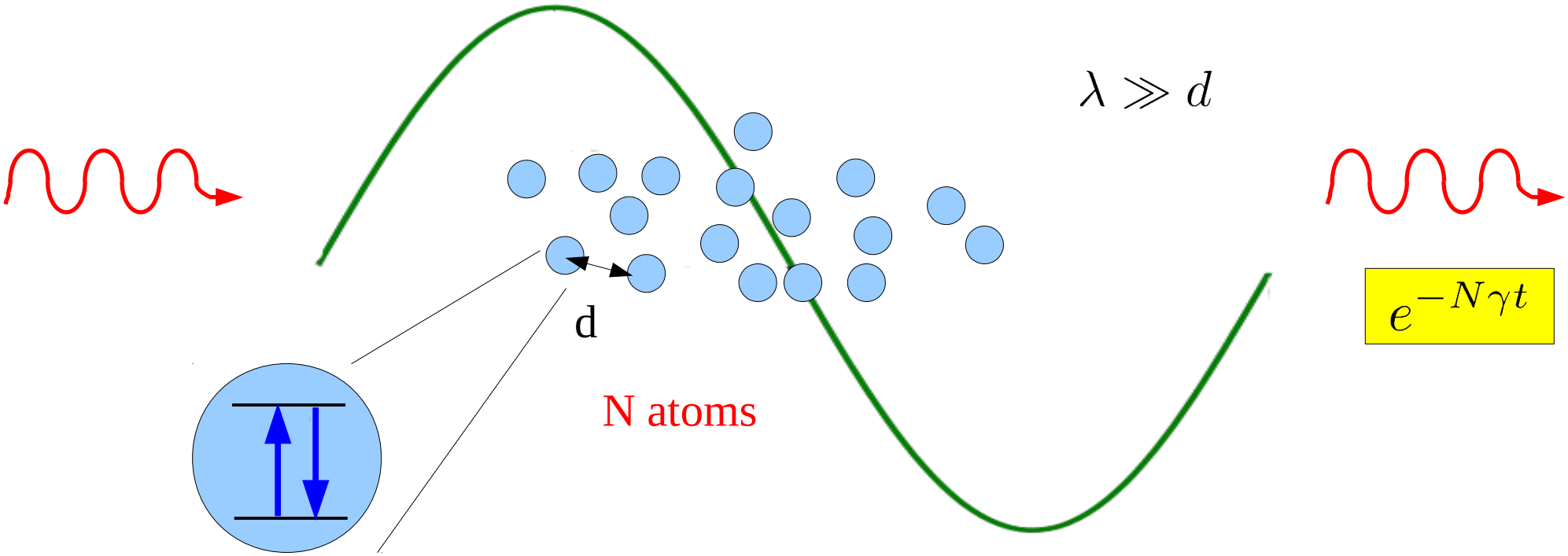
The beauty of collective

One versus many





Superradiance in atoms



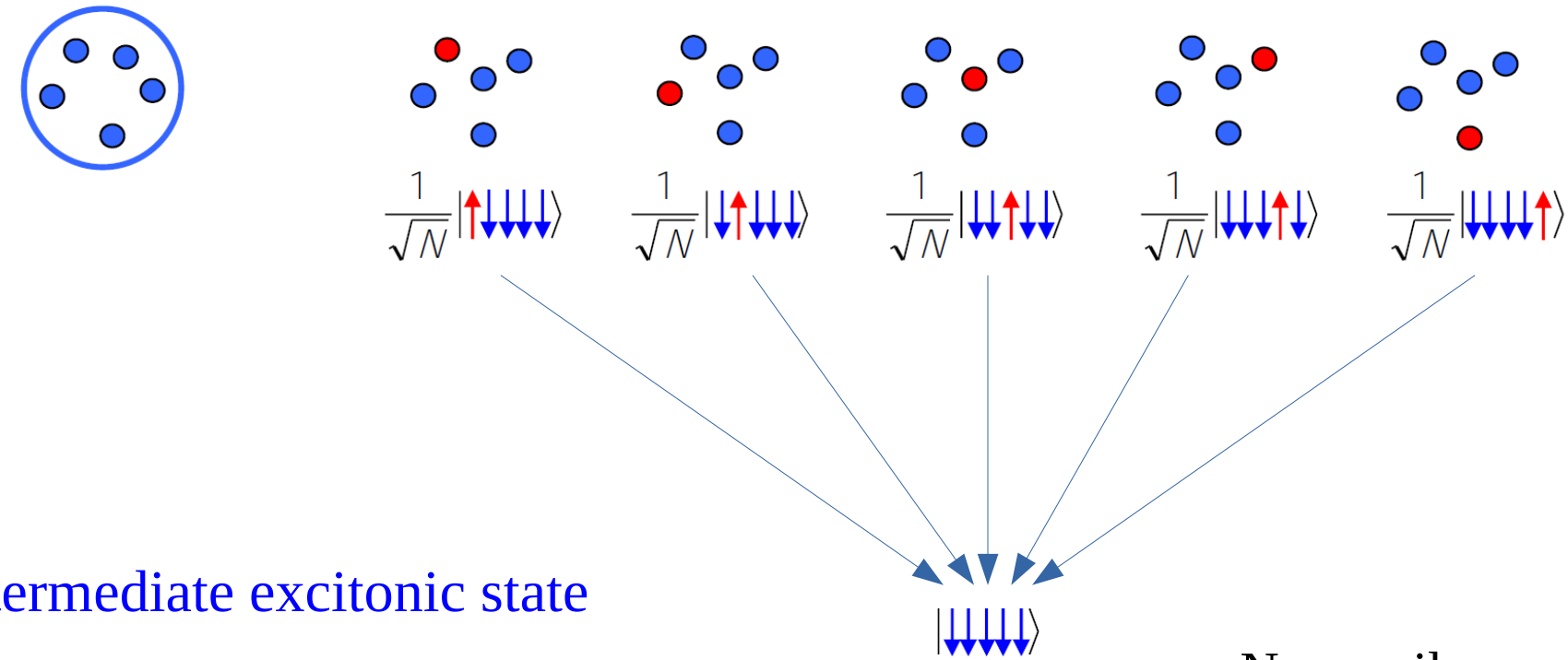
Many atoms are in the excited state, we don't know which one decays ...

Nuclei

→ Internuclear distance comparable to the wavelength $\lambda \simeq d$

→ Only one nucleus excited throughout the sample,

but we do not know which one!



Intermediate excitonic state

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

- No recoil
- No spin flip
- No internal conversion

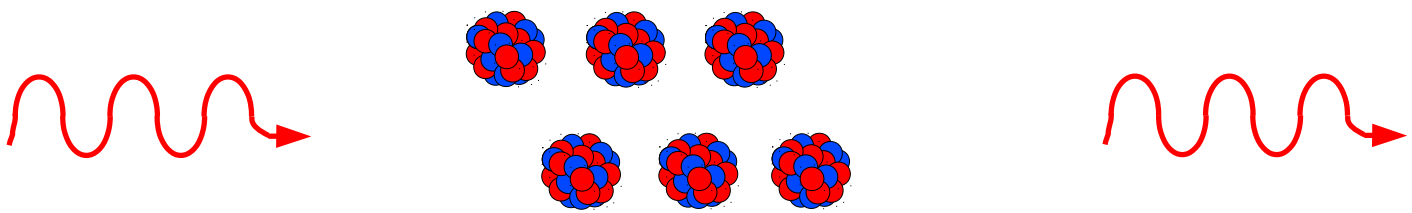
Privileged directions

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

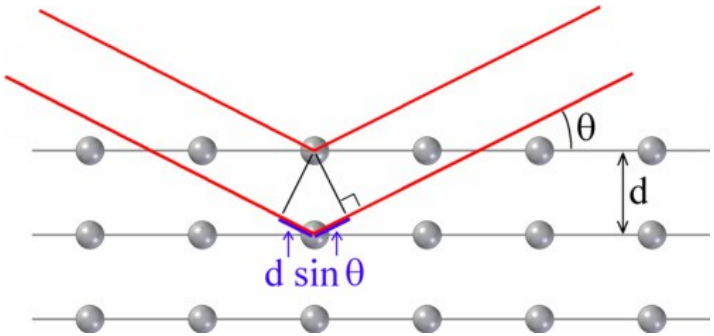
$$\boxed{e^{-N\gamma t}}$$

The phases add up constructively for

→ the forward direction



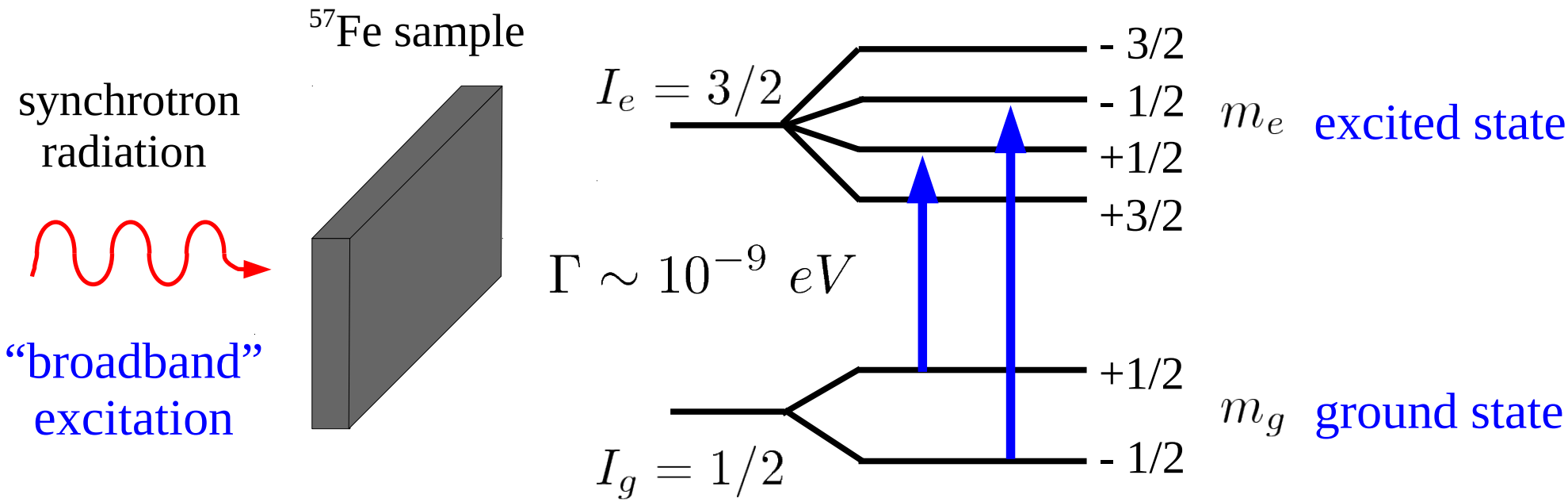
→ the Bragg direction in single crystals



$$\lambda = 2d \sin\theta$$

Nuclear forward scattering

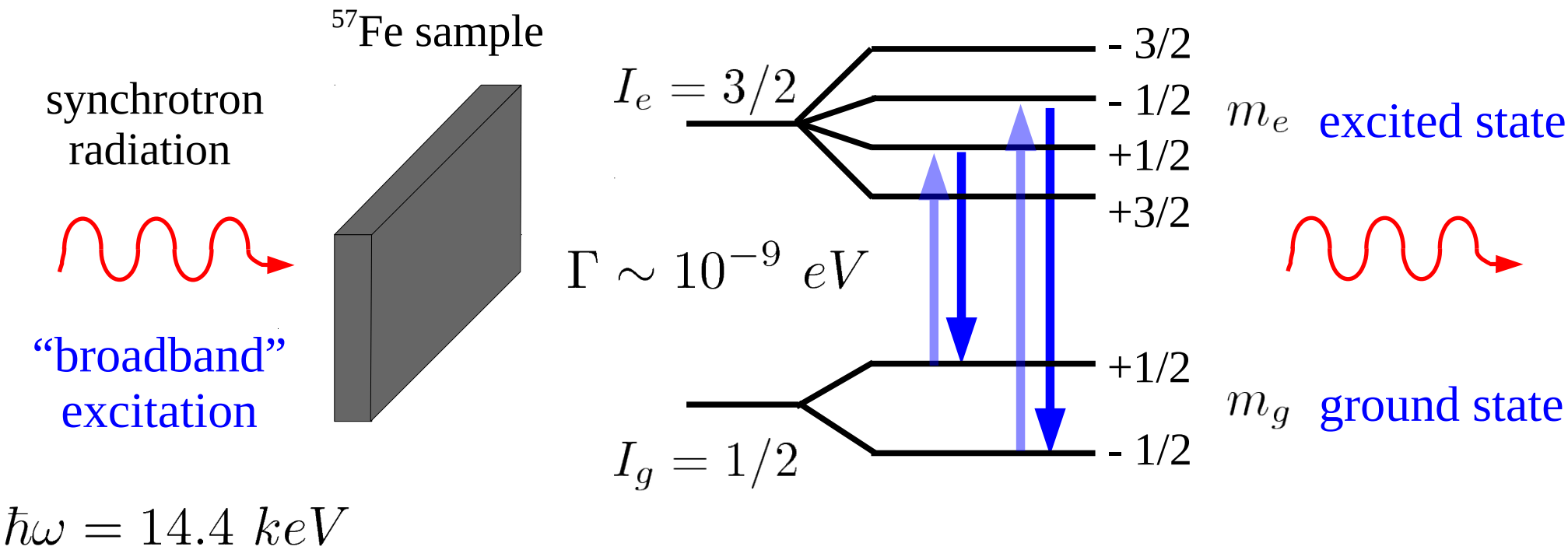
SCATTERING VIA AN EXCITONIC STATE



$\hbar\omega = 14.4\ keV$

Nuclear forward scattering

SCATTERING VIA AN EXCITONIC STATE



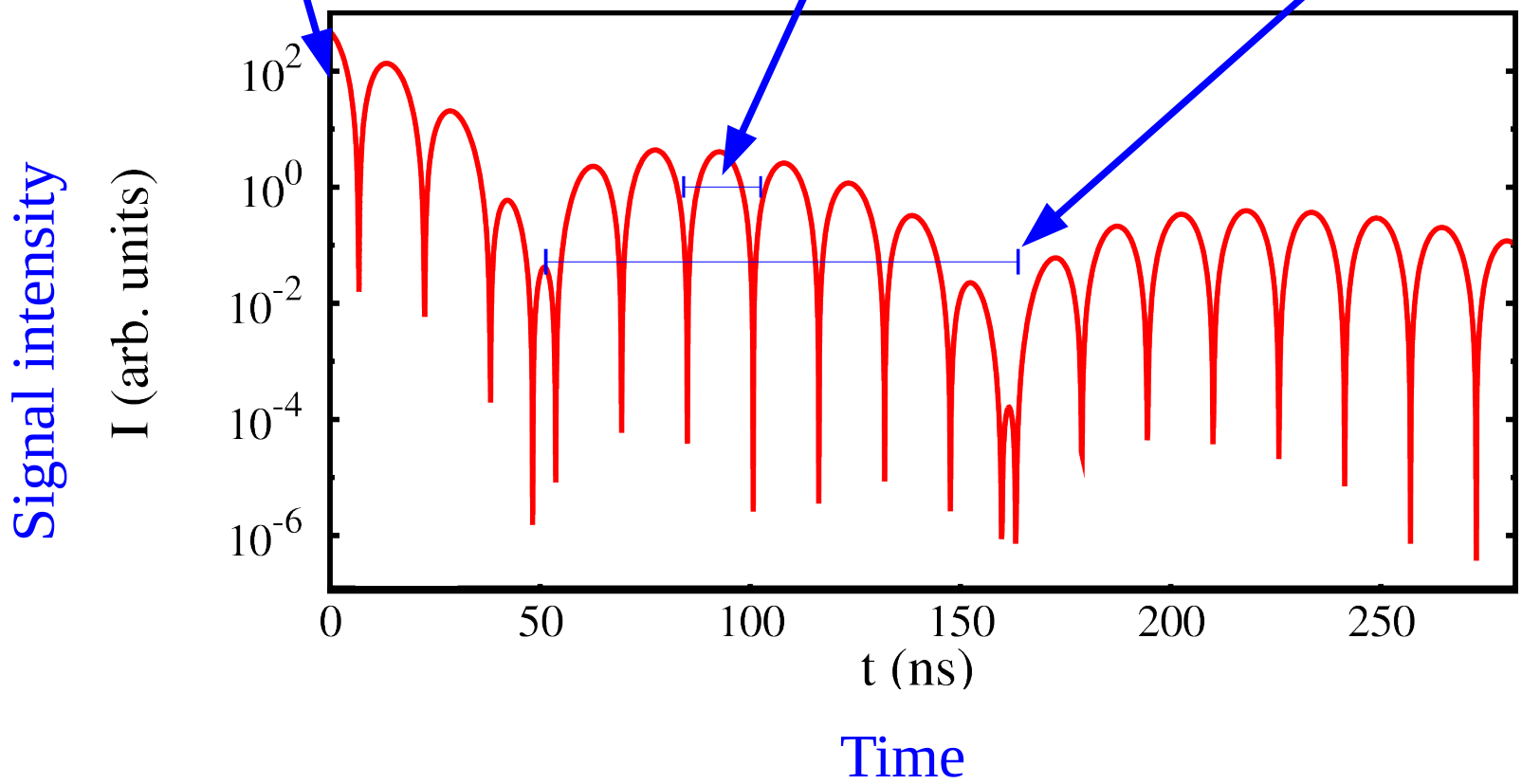
In coherent scattering, the nuclei return to their original state

→ scattering pathway unknown

t=0: almost instantaneous excitation, prompt (electronic) scattering → background

quantum beats due to different transition frequencies in single or different nuclei

dynamical beat (Bessel function) in thicker samples due to multiple scattering



Natural lifetime $t_0 = 141$ ns

Theoretical intermezzo

Wave equation

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

Slowly varying envelope approximation

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

Nuclei as source term (2nd order)

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

Final wave equation

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

sum over transitions

de-excitation

excitation

Iterative solution, incident pulse

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

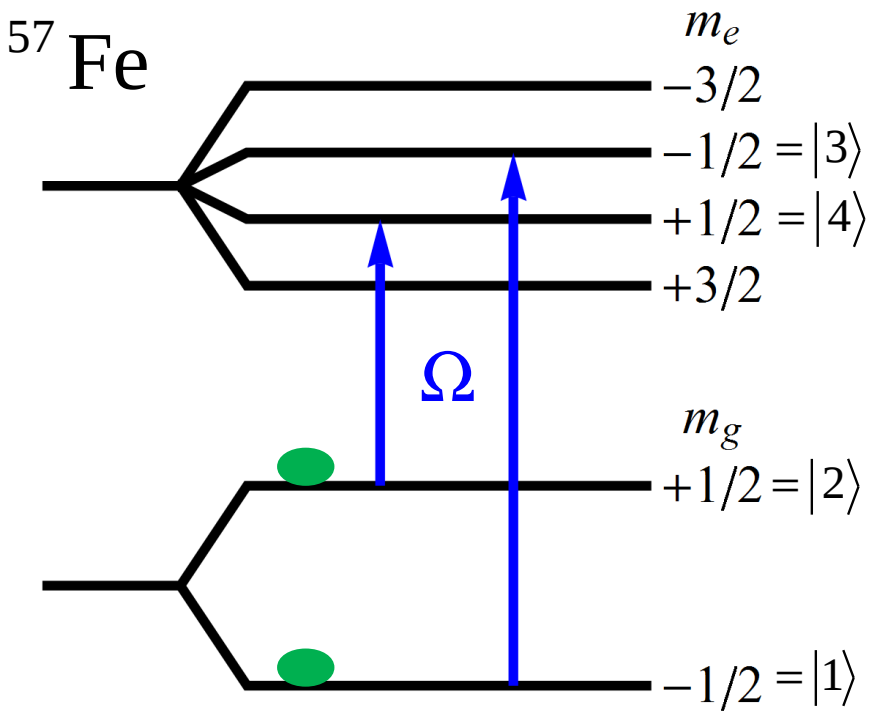
Y. V. Shvydko, Hyperf. Int. 123/124, 275 (1999)

Maxwell-Bloch equations

$$\partial_t \widehat{\rho} = \frac{1}{i\hbar}\left[\widehat{H}, \widehat{\rho}\right] + \widehat{\rho}_s \,,$$

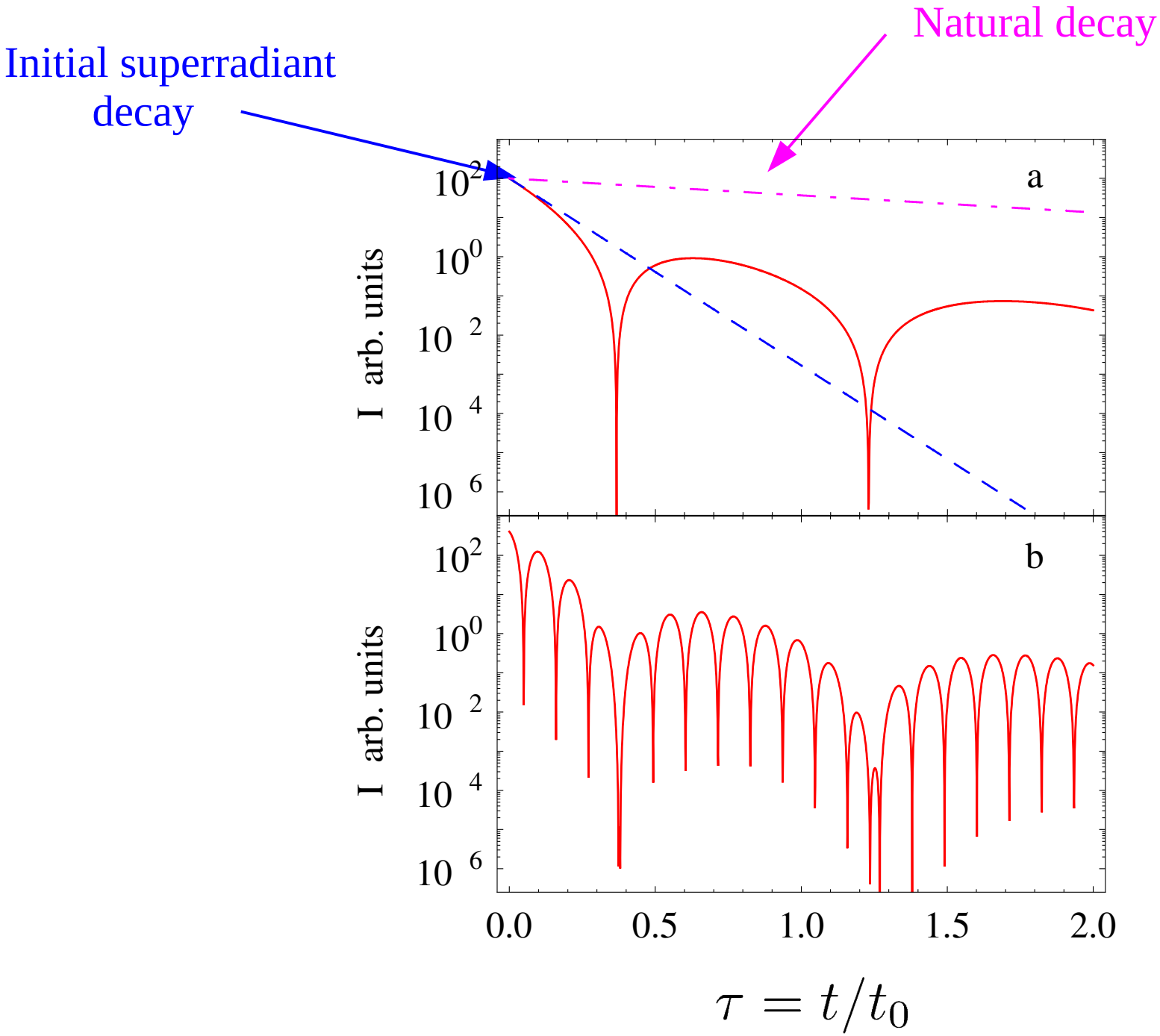
$$\frac{1}{c}\partial_t \Omega + \partial_y \Omega = i\eta\left(\rho_{31} + \rho_{42}\right)$$

Transition Currents



In both cases, classical fields although single photons!
Theory describes surprisingly well the experiments!

W.-T. Liao, AP and C. H. Keitel, Phys. Rev. Lett. 109, 197403 (2012)
X. Kong, W.-T. Liao and AP, New J. Phys. 16, 013049 (2014)


Natural decay
Initial superradiant decay
a
b
I arb. units
I arb. units
10^2
10^0
10^{-2}
10^{-4}
10^{-6}
0.0
0.5
1.0
1.5
2.0
$\tau = t/t_0$

CAN BE CONTROLLED!

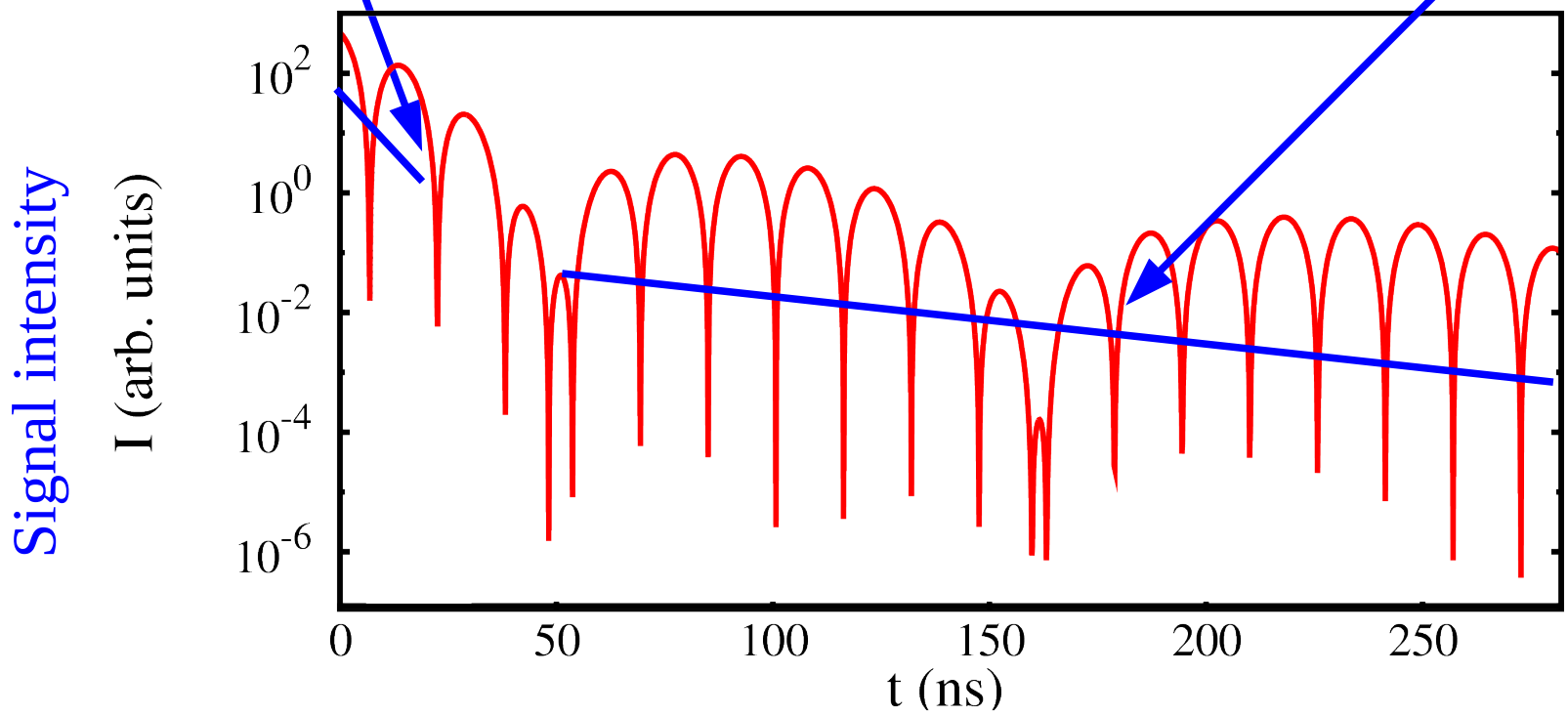
superradiant mode
populates subradiant modes

THICK SAMPLE

Superradiant behaviour

$$\gamma \rightarrow N\,\gamma$$

slower long-time decay
(non-exponential)



Time

superradiant mode
populates subradiant modes

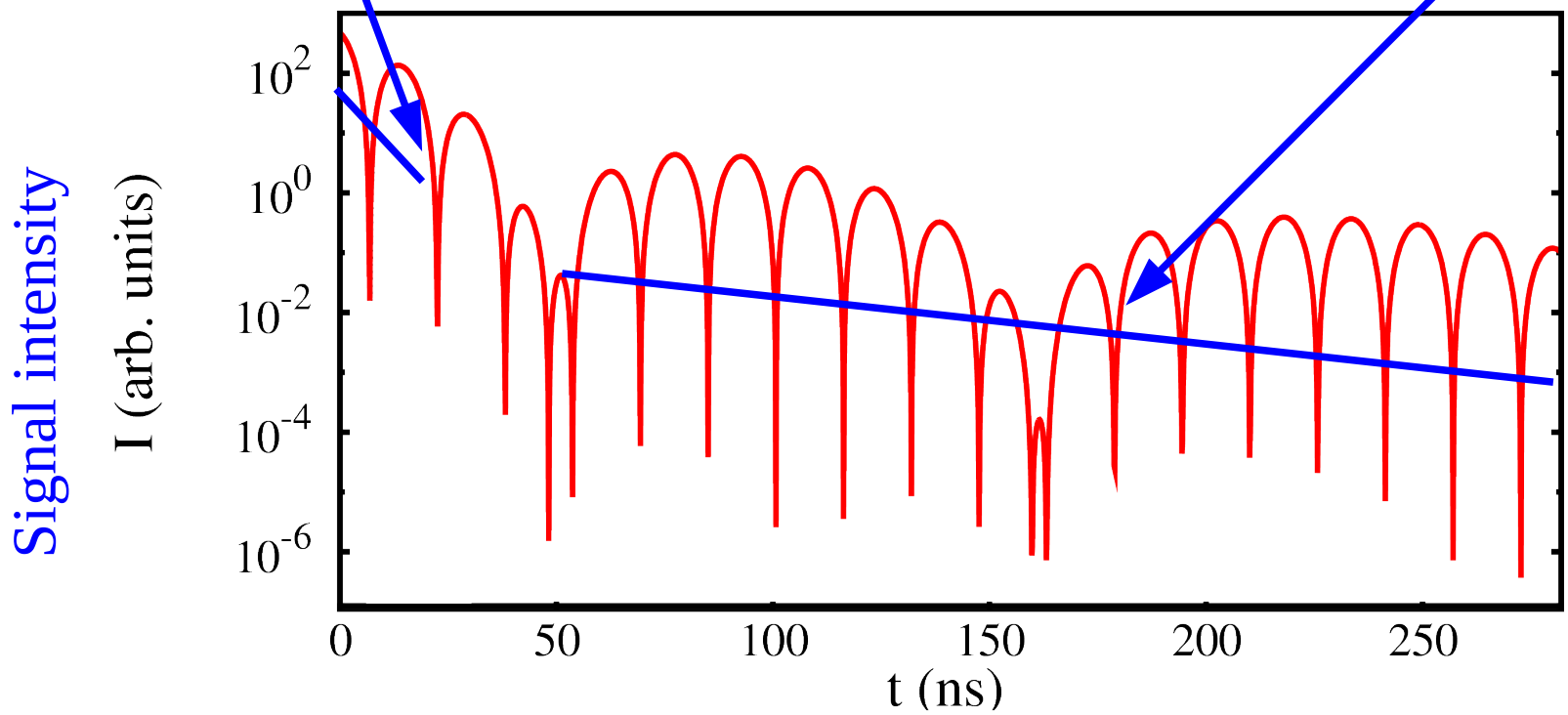
CAN BE CONTROLLED!

THICK SAMPLE

Superradiant behaviour

$$\gamma \rightarrow N\,\gamma$$

slower long-time decay
(non-exponential)



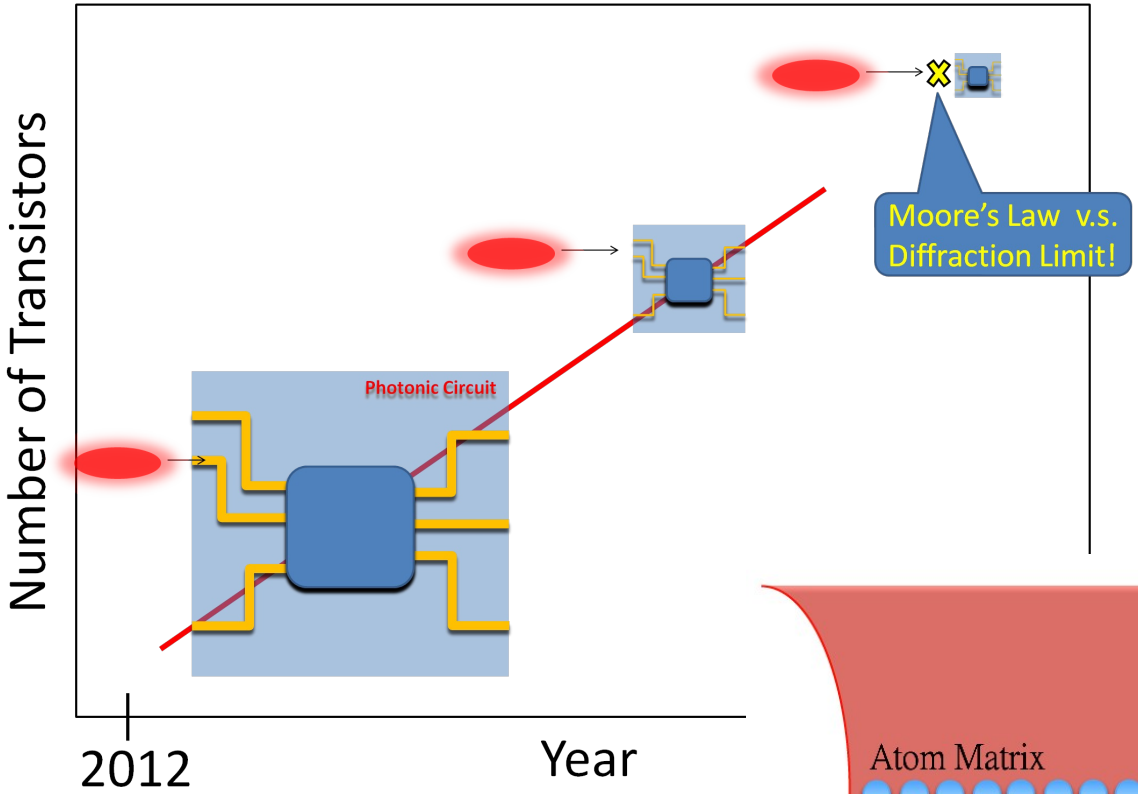
"THIN" SAMPLE

Time

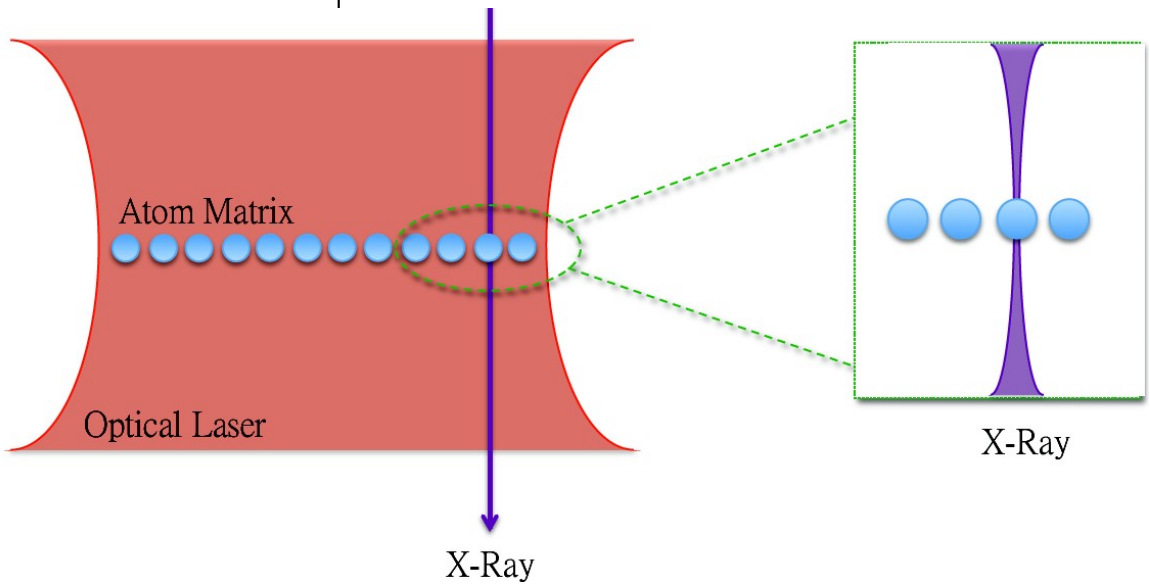
NANOLAYER CAVITIES EXPLOIT SUPERRADIANT PART - R. Röhlsberger

Control of single x-ray photons!

the information carriers of tomorrow?



photons circuits
robust x-ray qubits
well focusable

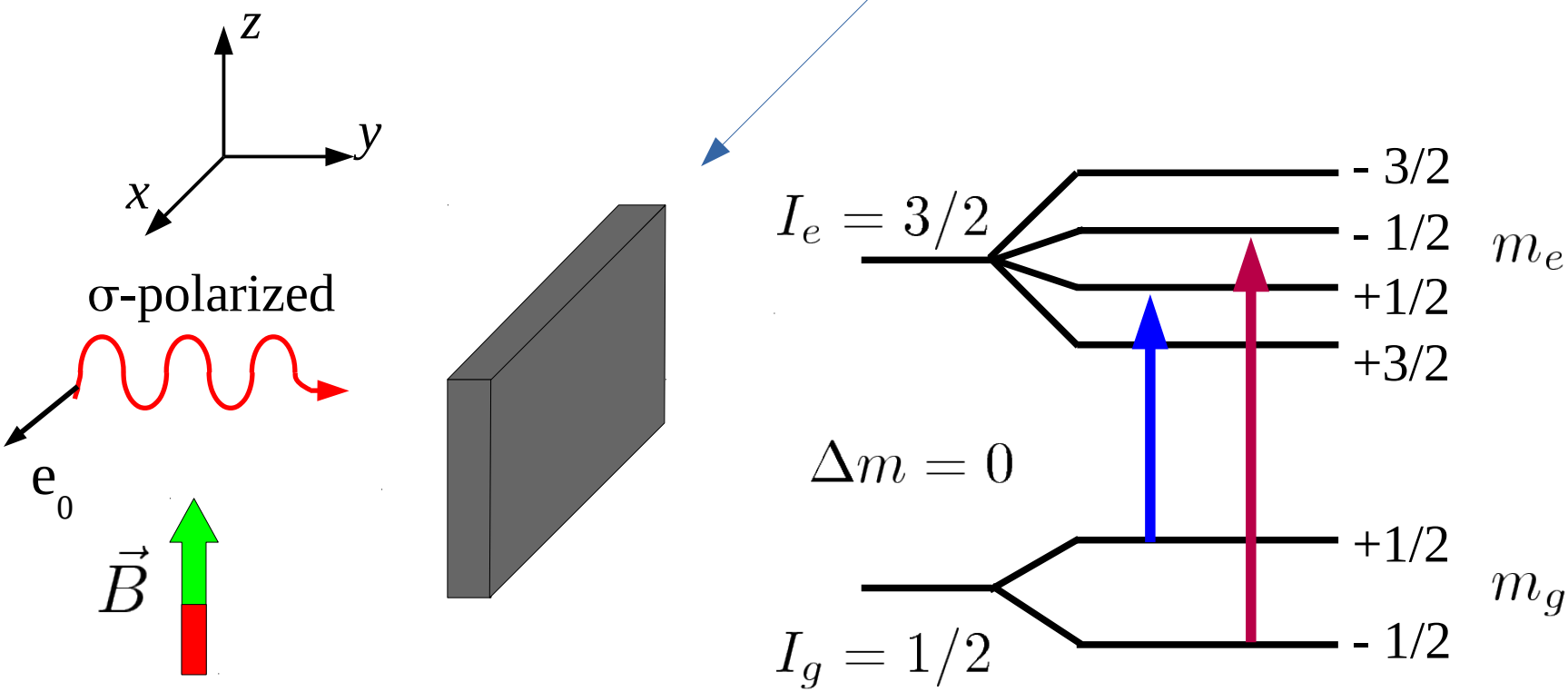


X-ray nuclear quantum optics with “thick” samples

- Storing and “shaping” single photons
- Single-photon entanglement

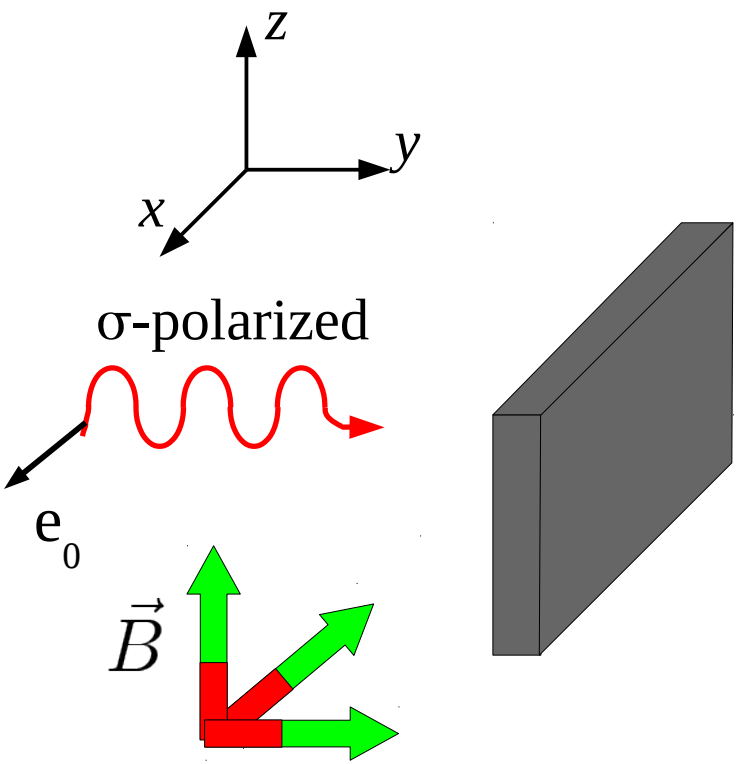
How to control the decay

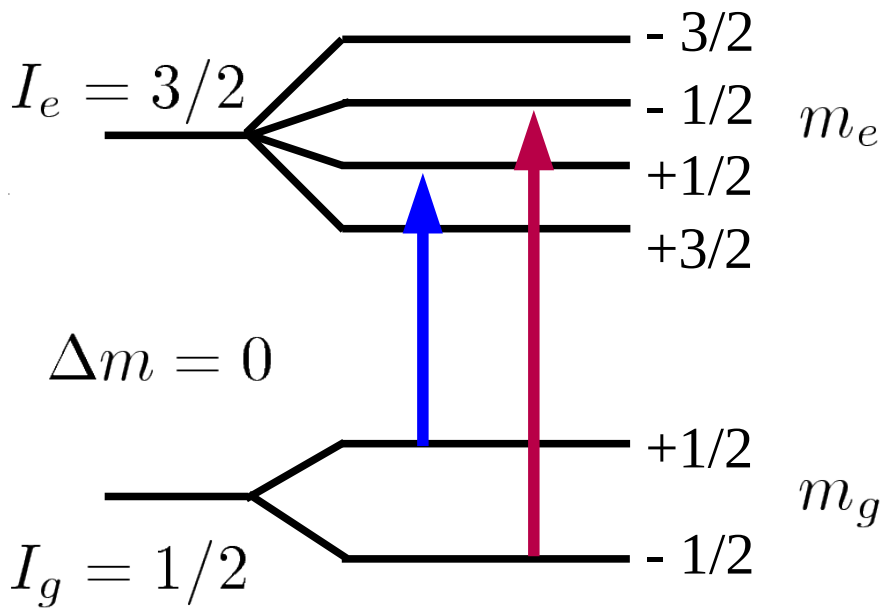
typically 10-micron-thick!



rotation of the nuclear hyperfine magnetic field

Magnetic switching

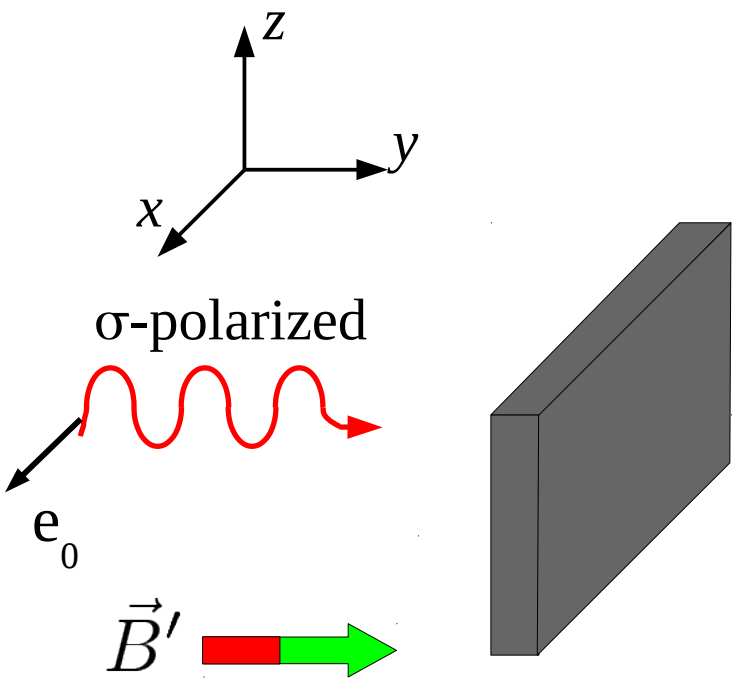


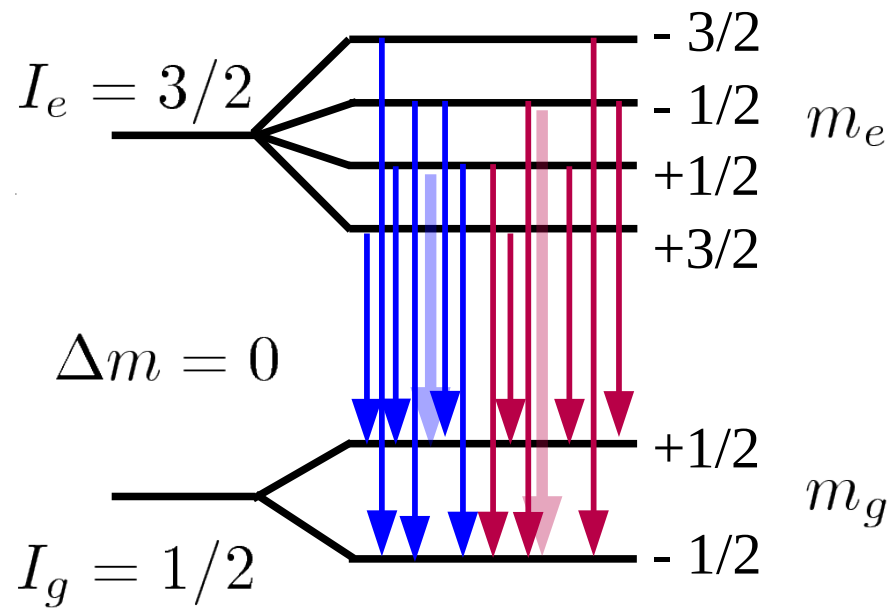


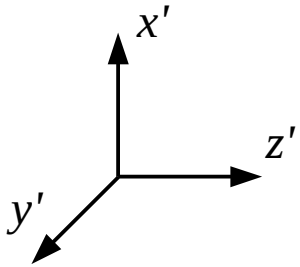
rotation of the nuclear hyperfine magnetic field

Magnetic switching

each monochromatic transition is transformed into a sextet!



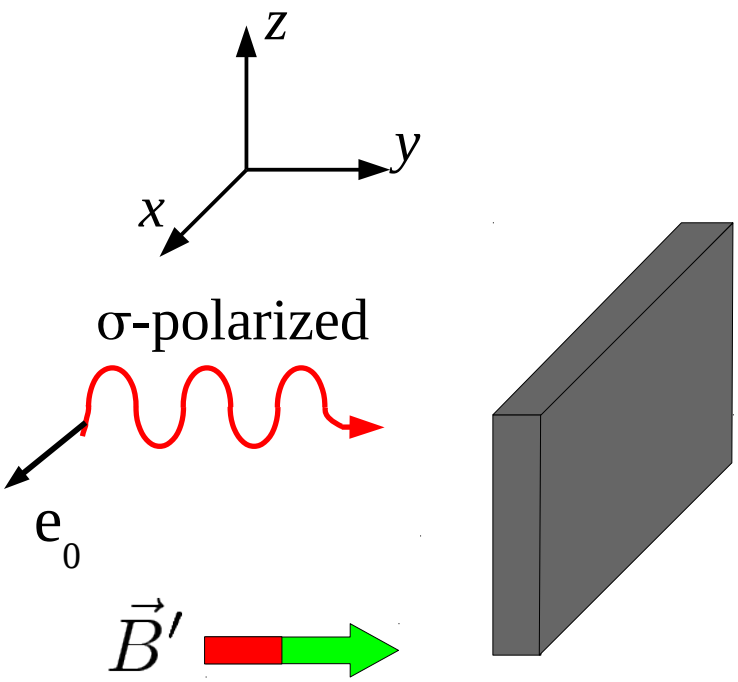


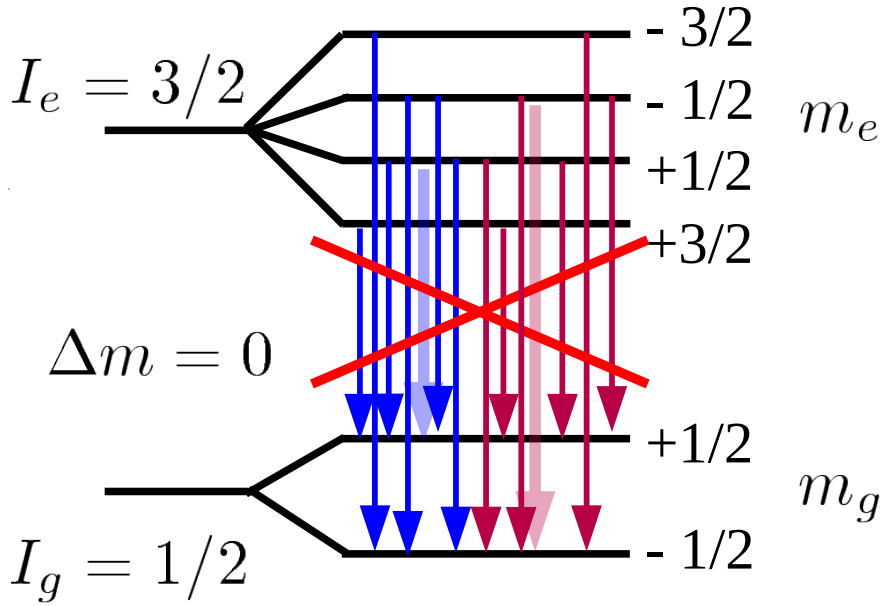


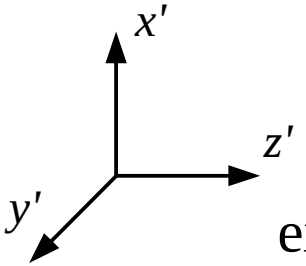
- redistribution of nuclear state population
- the new transition currents interfere

Magnetic switching

for certain switching TIME and ANGLE, the currents interfere destructively!



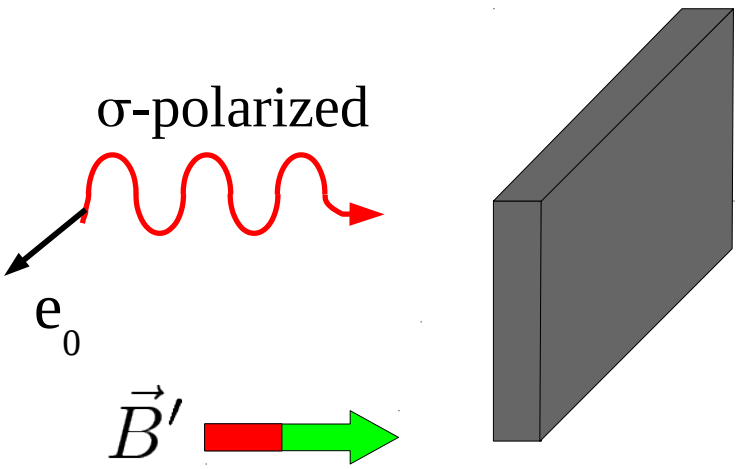


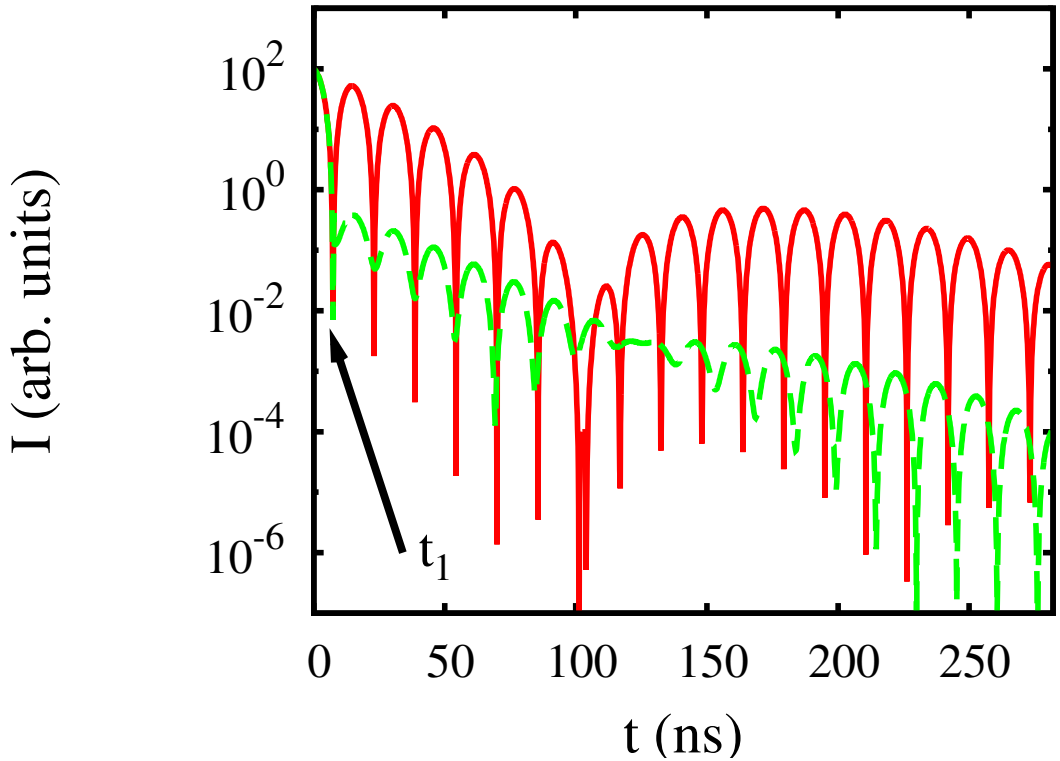


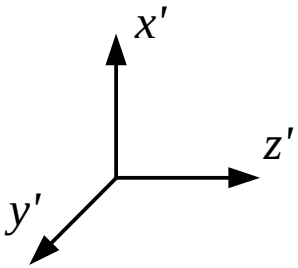
THE COHERENT DECAY IS SUPPRESSED!

experimentally observed: Yu. V. Shvyd'ko *et al.*, PRL 77 (1996) 3232

Magnetic switching



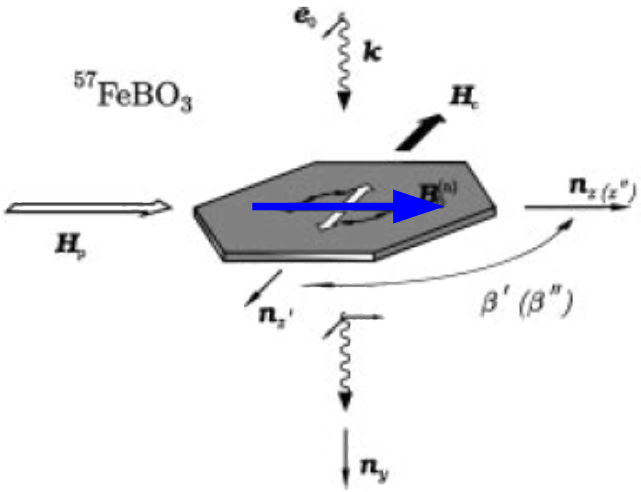




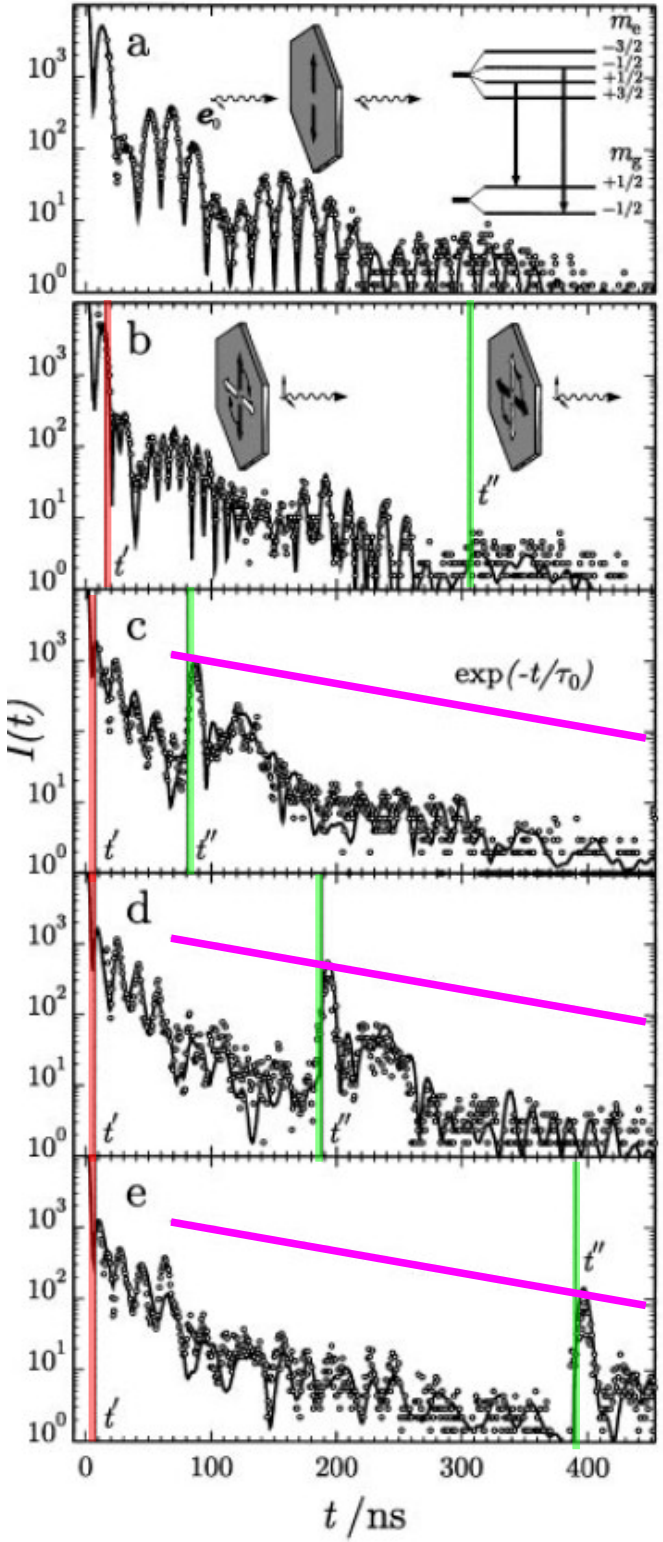
switching at the minima, complete suppression of dominant first order scattering is achieved

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

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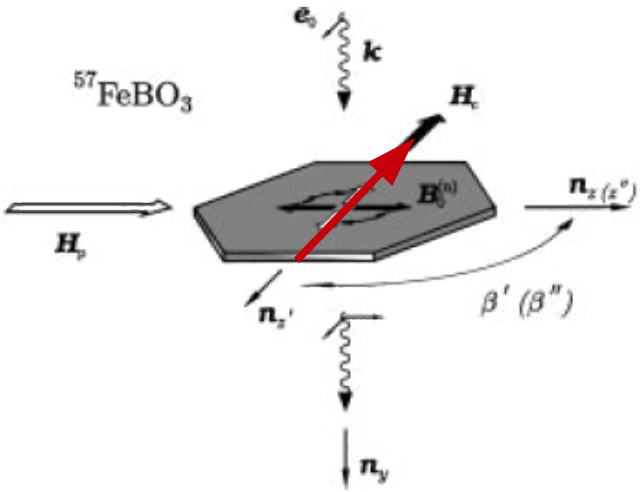

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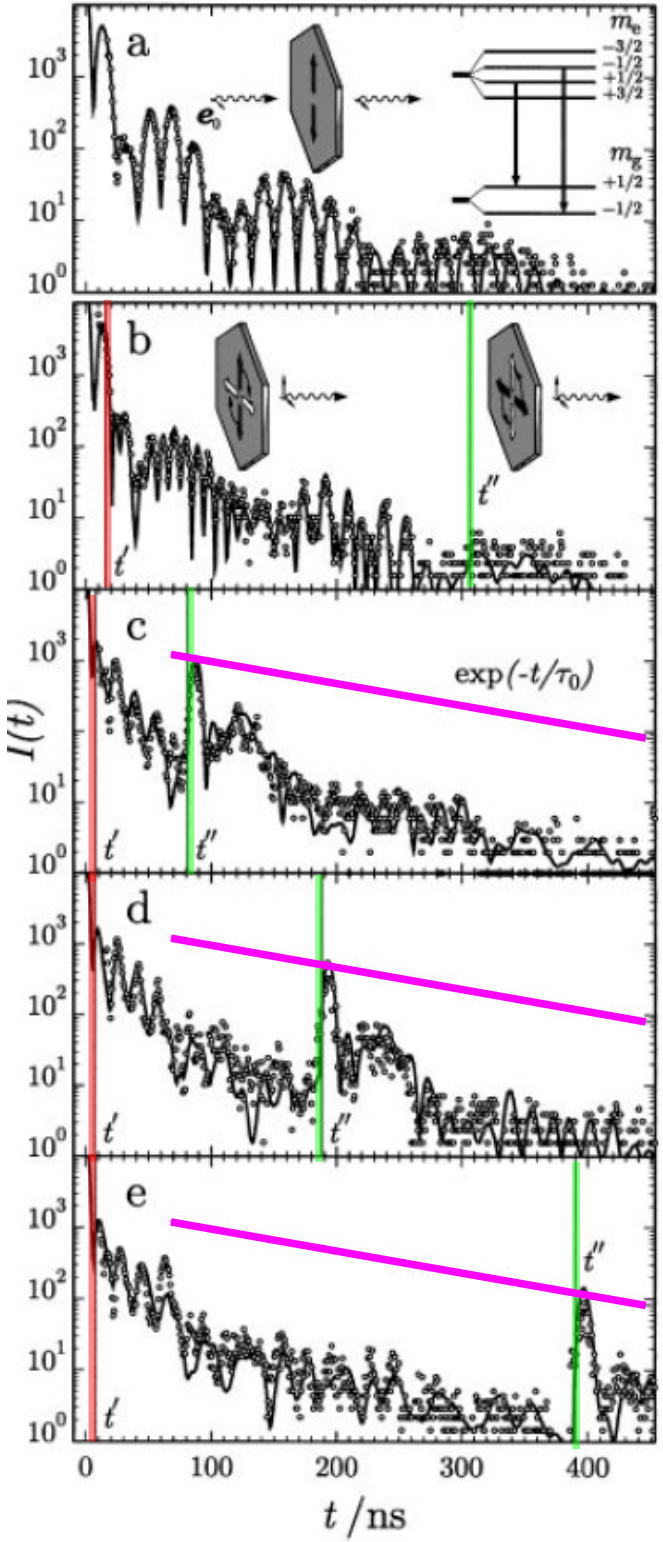

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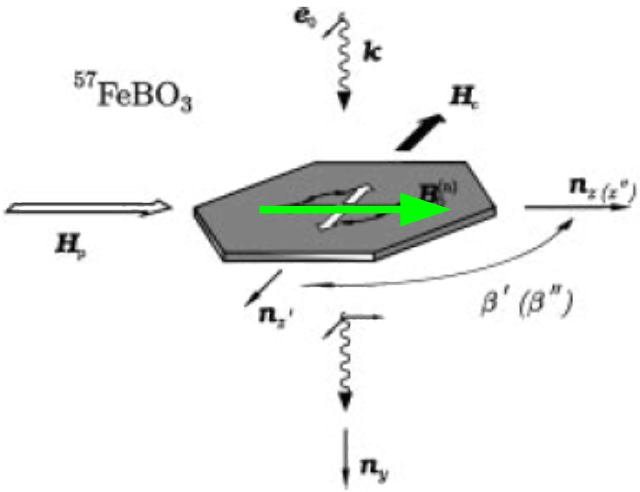

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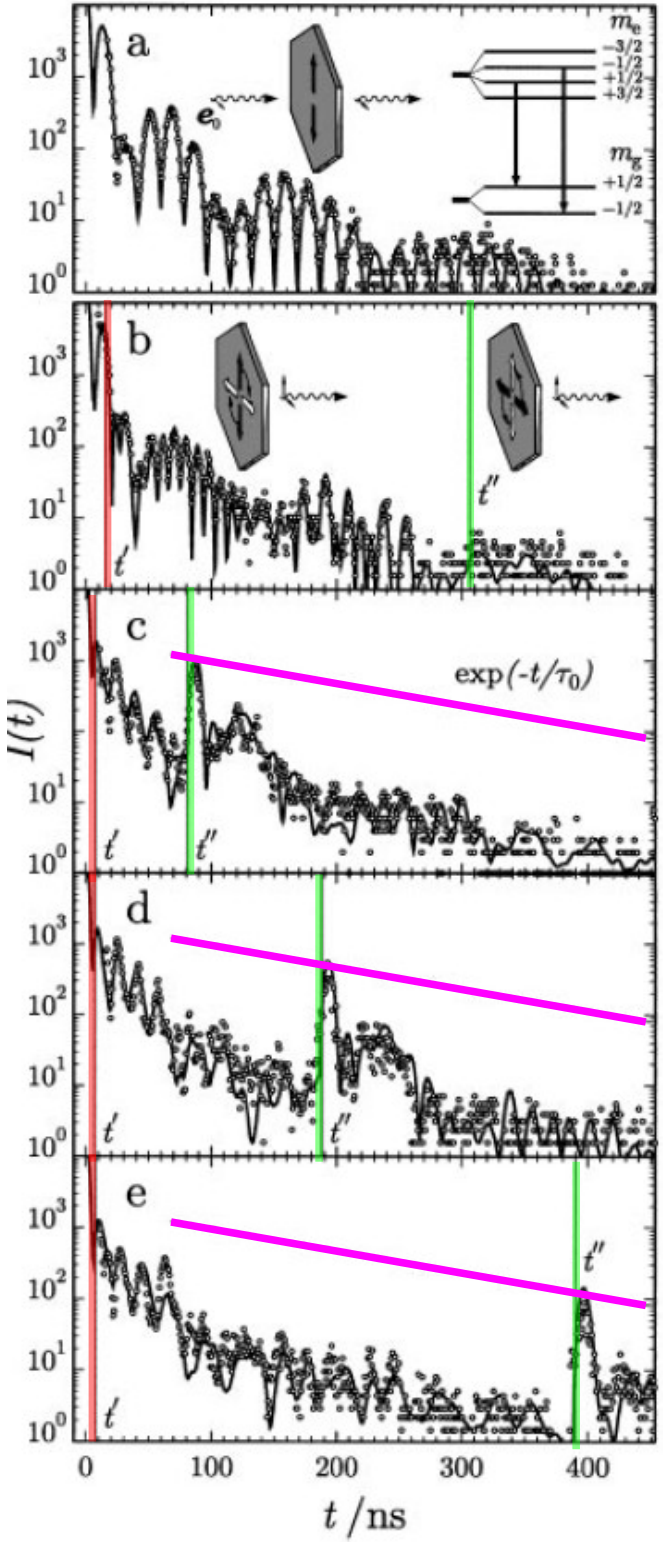


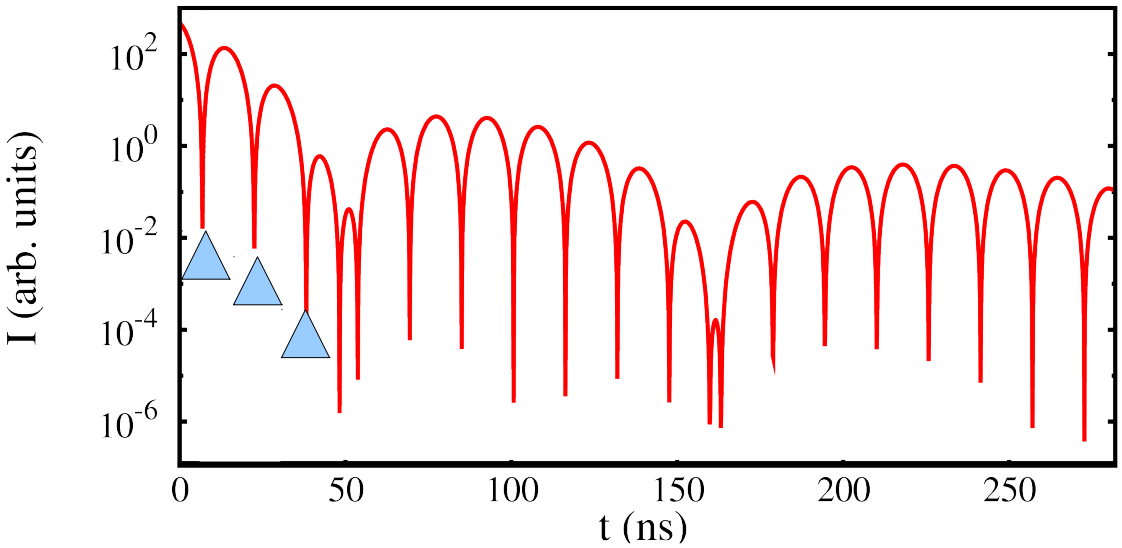
No switching

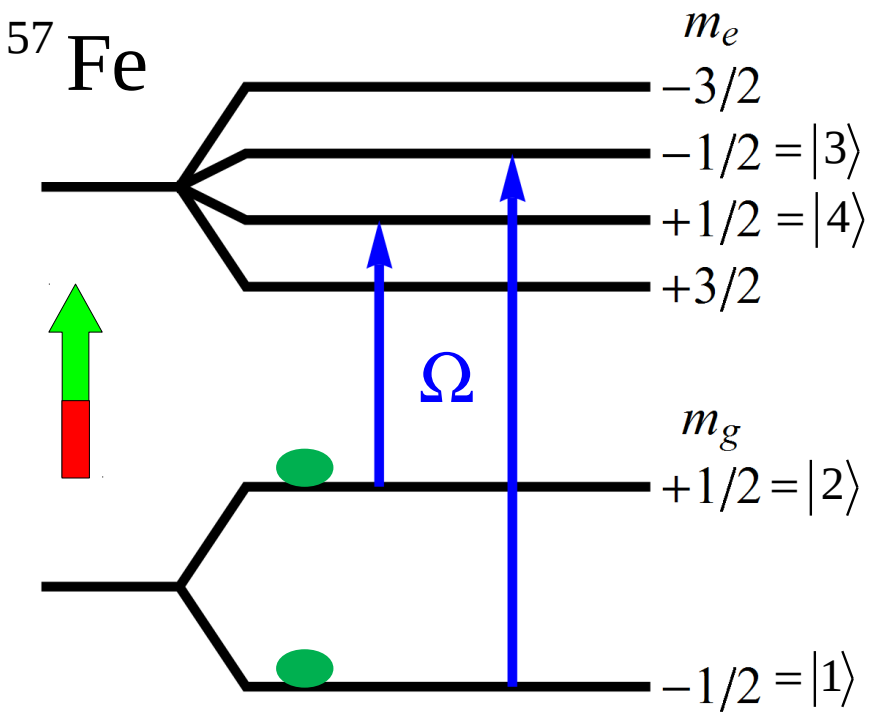
Apply switching

Switch back

Decay with natural life time

Upgrade for phase - Coherent storage





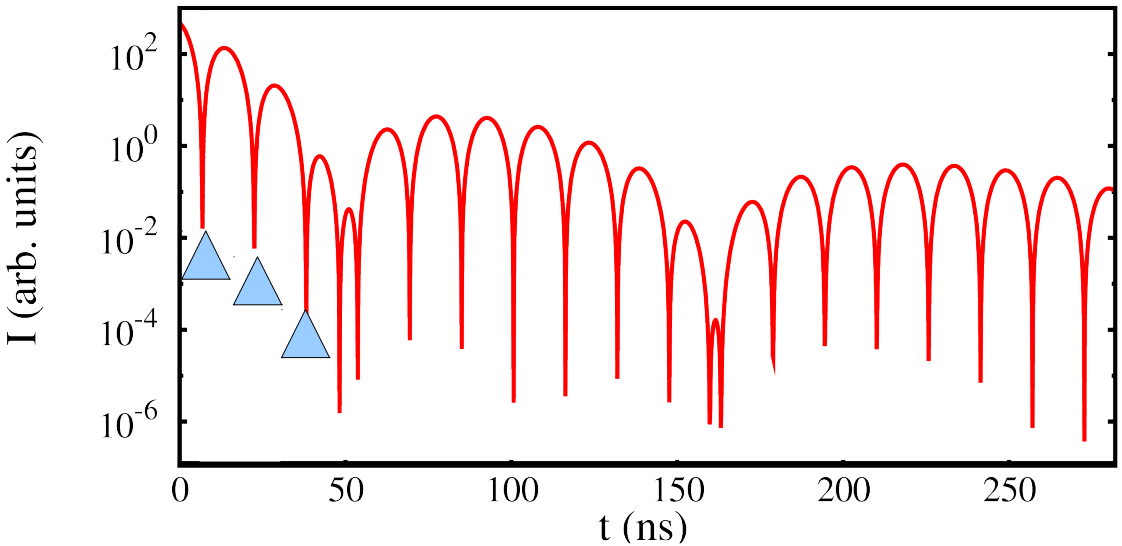
Quantum beat minima – moments of storage!

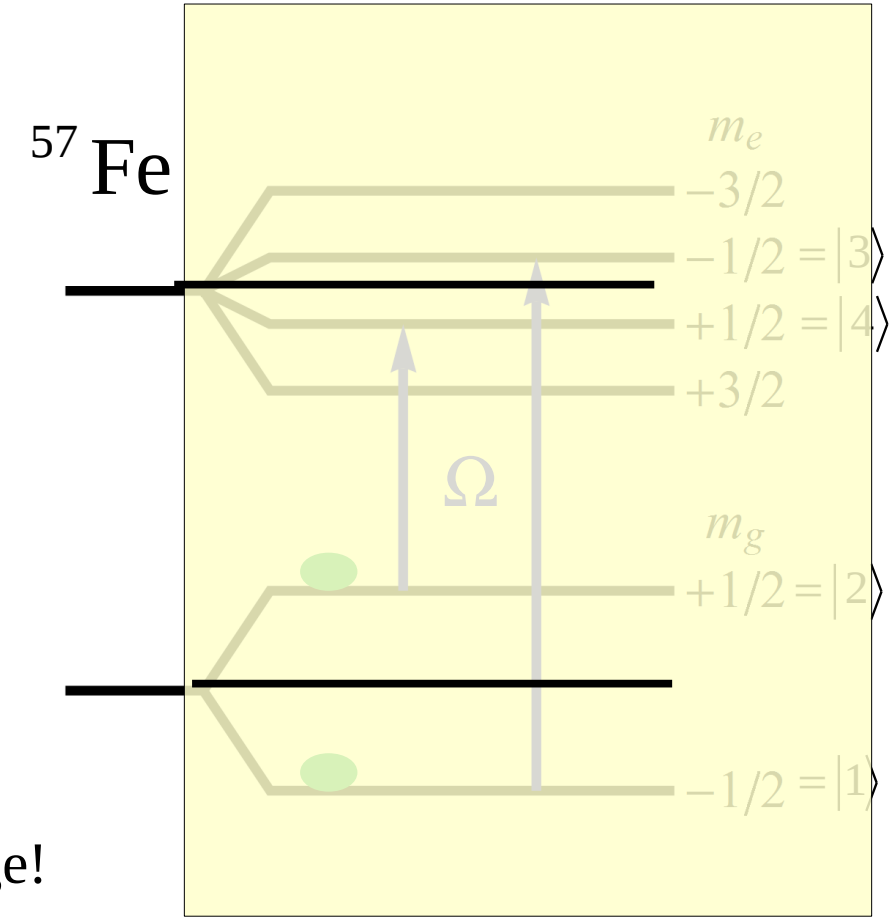
$$\partial_t \widehat{\rho} = \frac{1}{i\hbar}\left[\widehat{H}, \widehat{\rho}\right] + \widehat{\rho}_s \,,$$

Transition currents become imaginary

$$\frac{1}{c}\partial_t \Omega + \partial_y \Omega = i\eta\left(\rho_{31} + \rho_{42}\right)$$

Coherent storage – a proposal





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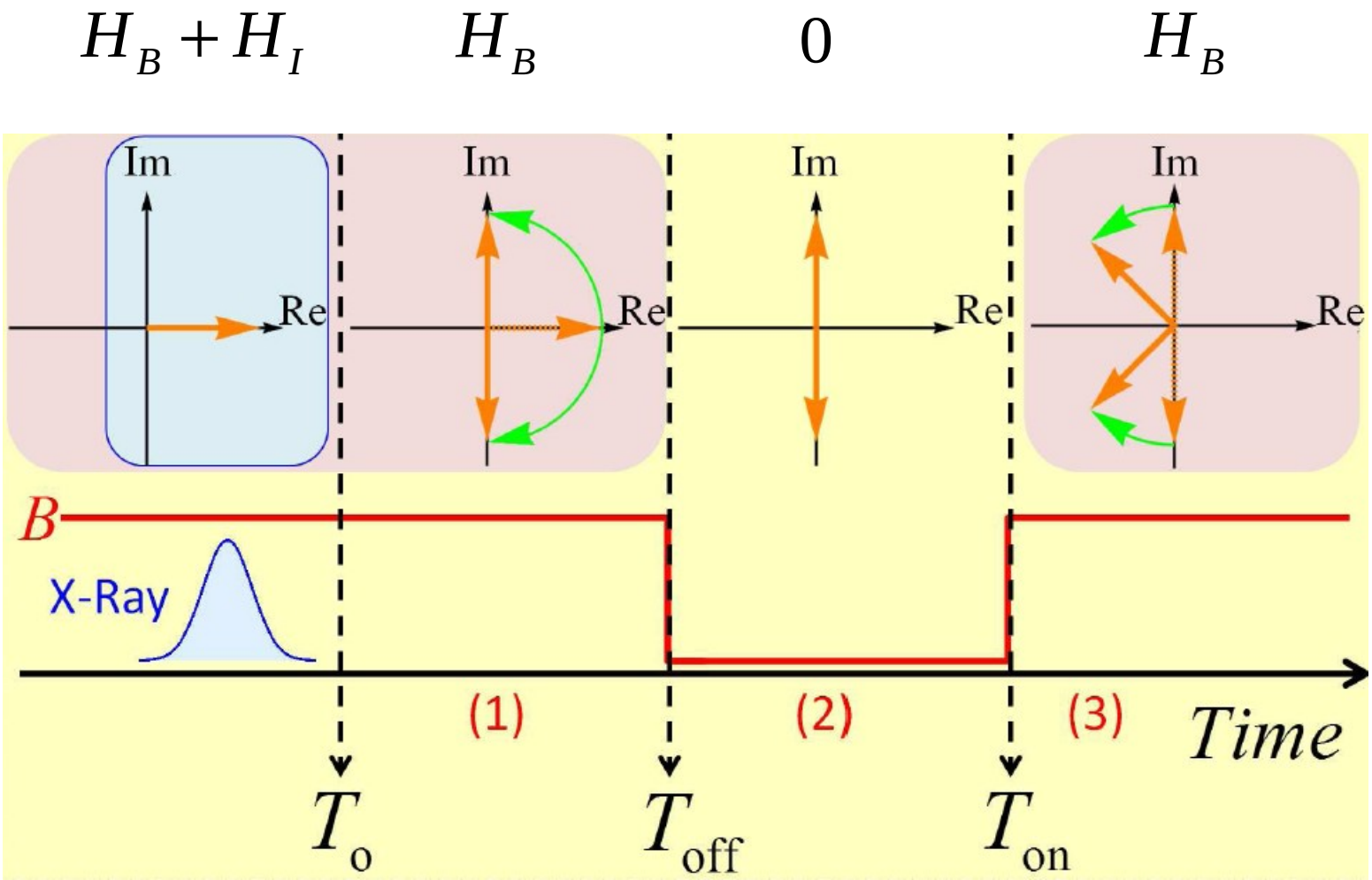
Transition currents become imaginary

Coherent storage

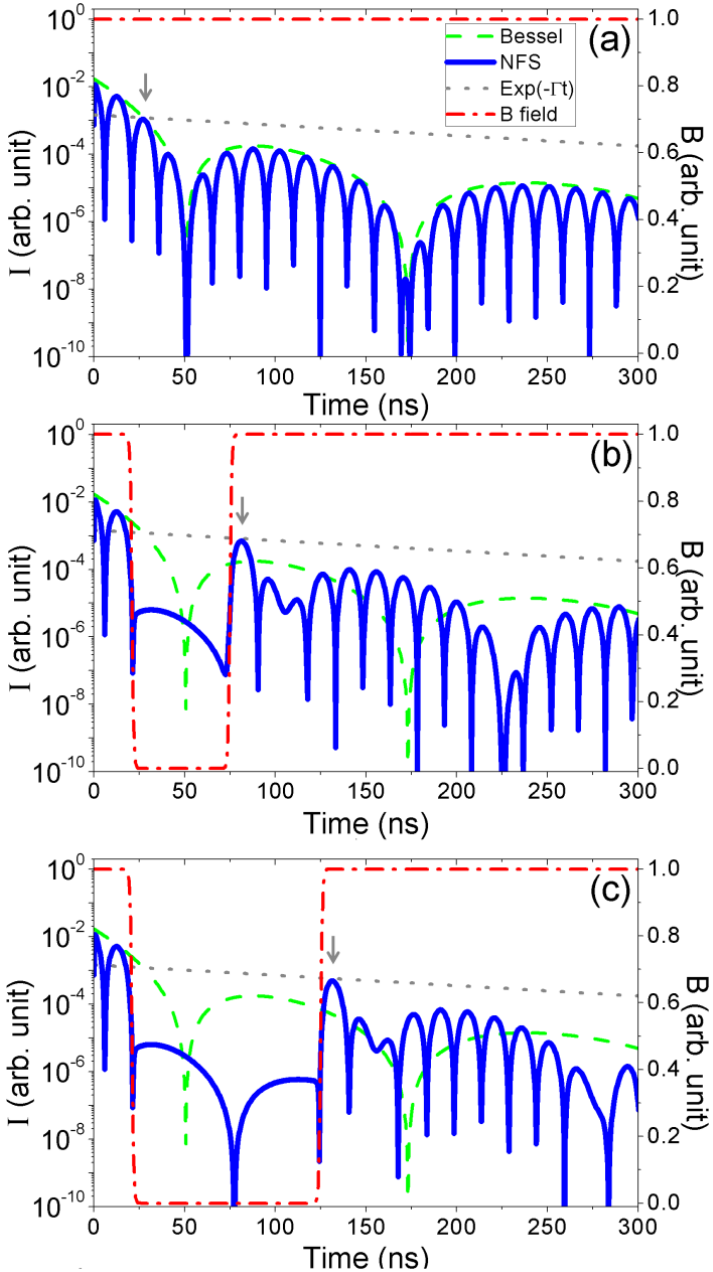


W.-T. Liao, AP and C. H. Keitel, PRL 109, 197403 (2012)

Coherent storage

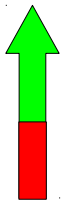
Both polarization and phase are conserved!!!

→ Coherent storage of a single x-ray photon

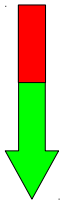


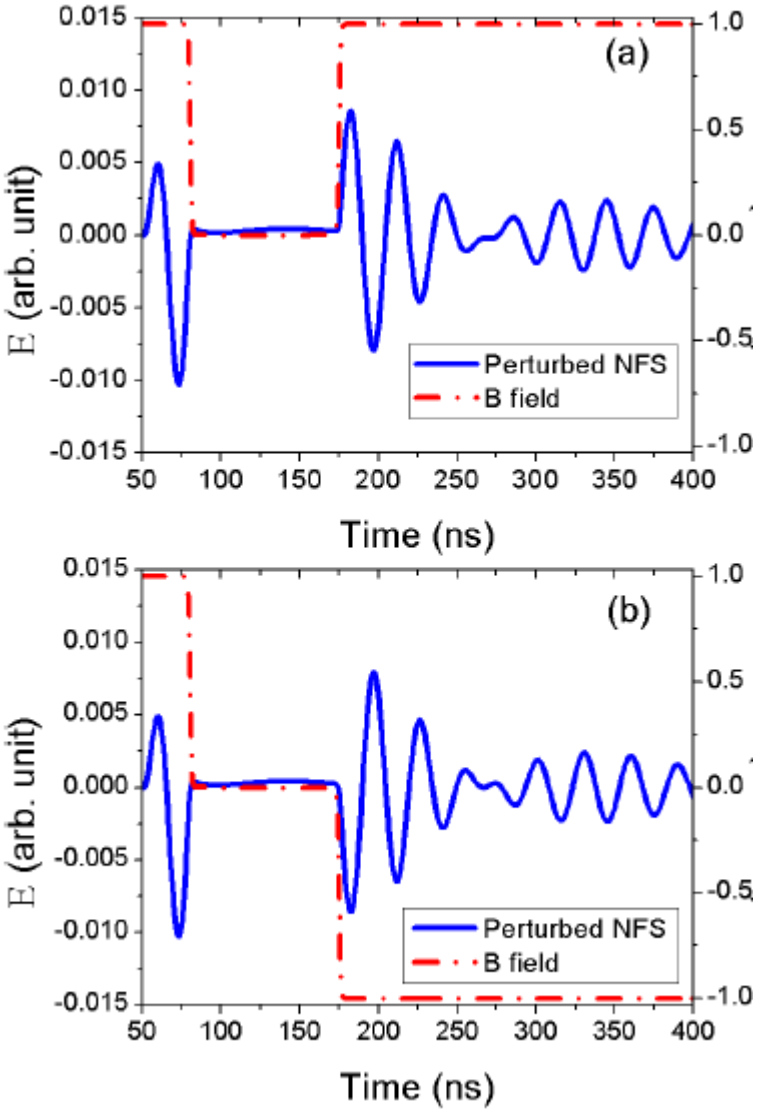
W.-T. Liao, AP and C. H. Keitel, PRL 109, 197403 (2012)

Coherent storage



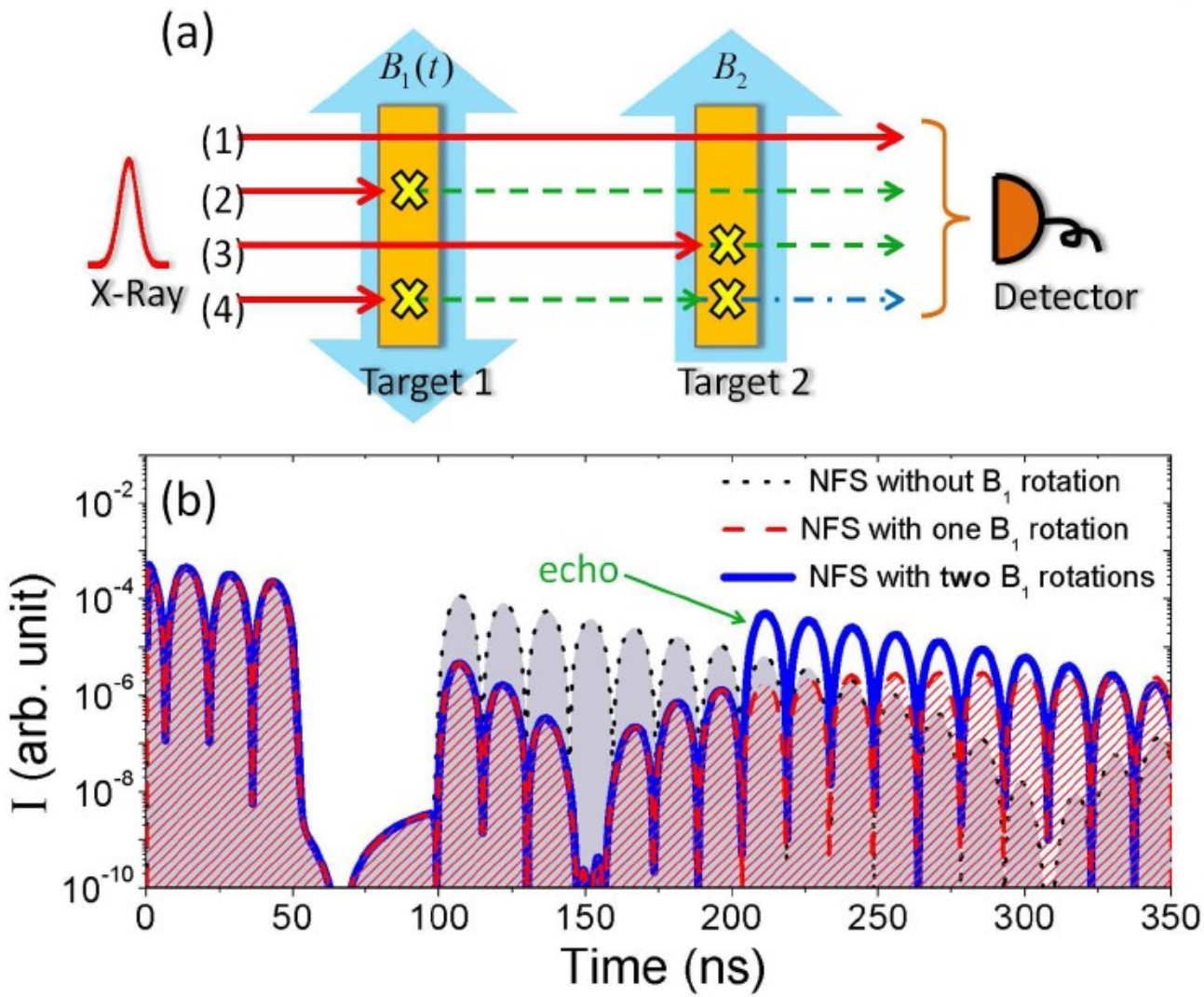
Phase is modulated by π by a 180° rotation of the reviving magnetic field!





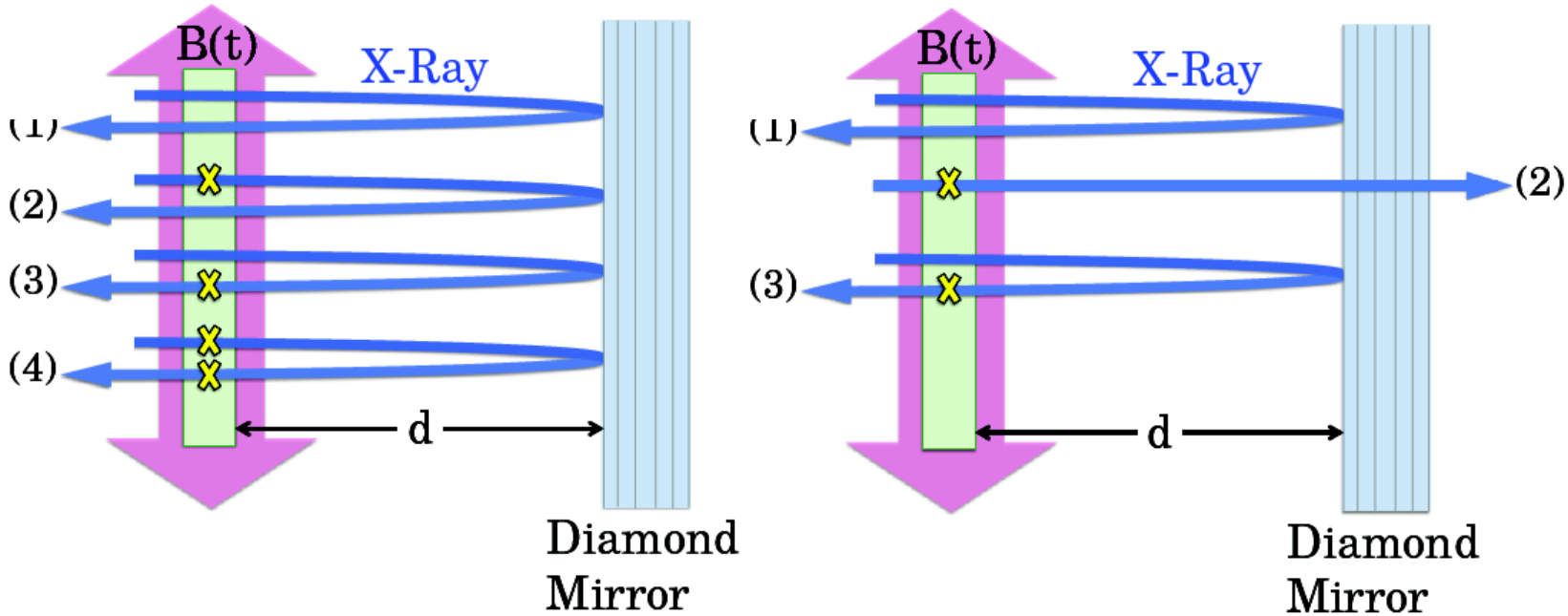
W.-T. Liao, AP and C. H. Keitel, PRL 109, 197403 (2012)

Phase modulation check



W.-T. Liao, AP and C. H. Keitel, PRL 109, 197403 (2012)

Shaping of x-ray photon wavepackets

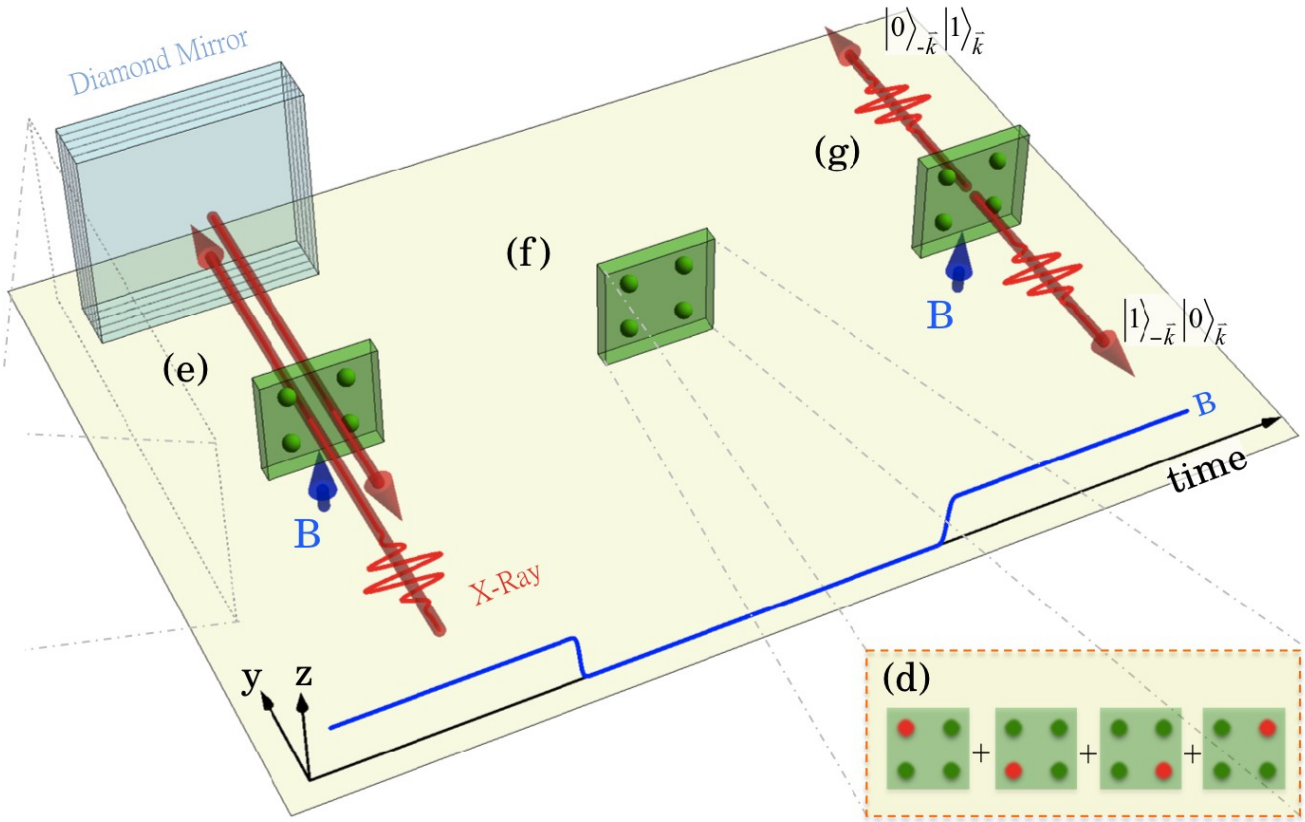


Single-pass Nuclear Polariton

$$|\mathrm{NP}\rangle = \frac{\alpha(t)}{\sqrt{N}} \sum_{\ell=1}^{N} |g\rangle|e_\ell\rangle e^{i\vec{k}\cdot\vec{r}_\ell} \otimes |0\rangle + \beta(t)|G\rangle \otimes |1\rangle_{\vec{k}}$$

W.-T. Liao and AP, PRL 112, 057401 (2014)

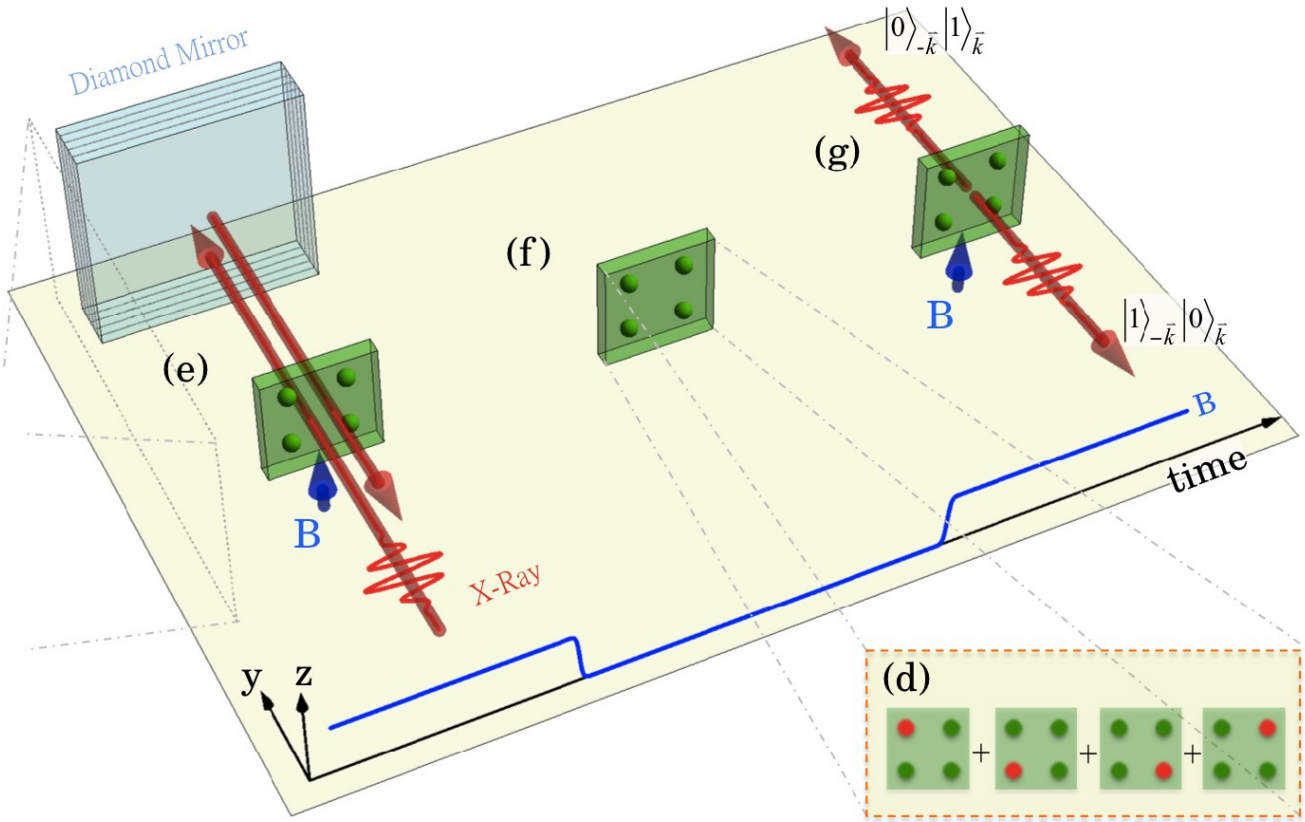
Shaping of x-ray photon wavepackets



$$|\mathrm{NPE}\rangle = \frac{\alpha(t)}{2\sqrt{\pi N}} \sum_{\ell=1}^{N} |g\rangle|e_\ell\rangle \left(e^{i\vec{k}\cdot\vec{r}_\ell} \pm e^{-i\vec{k}\cdot\vec{r}_\ell}\right) \otimes |0\rangle$$
$$+ \frac{\beta(t)}{\sqrt{2}} |G\rangle \otimes \left(|1\rangle_{-\vec{k}}|0\rangle_{\vec{k}} \pm |0\rangle_{-\vec{k}}|1\rangle_{\vec{k}}\right) .$$

W.-T. Liao and AP, PRL 112, 057401 (2014)

Shaping of x-ray photon wavepackets

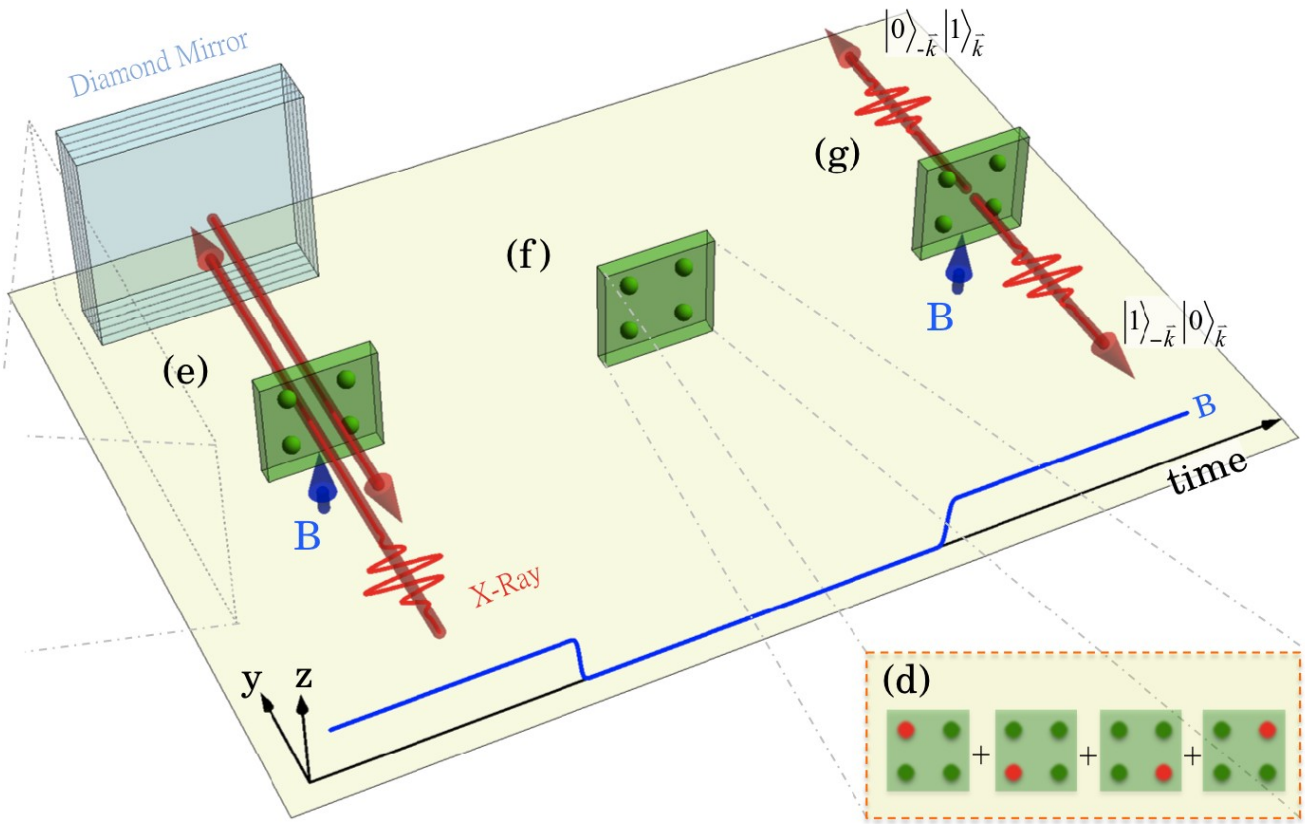


standing wave excitation pattern

$$|\mathrm{NPE}\rangle = \frac{\alpha(t)}{2\sqrt{\pi N}} \sum_{\ell=1}^{N} |g\rangle|e_\ell\rangle \boxed{\left(e^{i\vec{k}\cdot\vec{r}_\ell} \pm e^{-i\vec{k}\cdot\vec{r}_\ell}\right)} \otimes |0\rangle$$
$$+ \frac{\beta(t)}{\sqrt{2}} |G\rangle \otimes \left(|1\rangle_{-\vec{k}}|0\rangle_{\vec{k}} \pm |0\rangle_{-\vec{k}}|1\rangle_{\vec{k}}\right) .$$

W.-T. Liao and AP, PRL 112, 057401 (2014)

Shaping of x-ray photon wavepackets



$$|\mathrm{NPE}\rangle = \frac{\alpha(t)}{2\sqrt{\pi N}} \sum_{\ell=1}^{N} |g\rangle|e_\ell\rangle \left(e^{i\vec{k}\cdot\vec{r}_\ell} \pm e^{-i\vec{k}\cdot\vec{r}_\ell}\right) \otimes |0\rangle + \frac{\beta(t)}{\sqrt{2}} |G\rangle \otimes \boxed{\left(|1\rangle_{-\vec{k}}|0\rangle_{\vec{k}} \pm |0\rangle_{-\vec{k}}|1\rangle_{\vec{k}}\right)}.$$

W.-T. Liao and AP, PRL 112, 057401 (2014) *single-photon entanglement*

X-ray nuclear quantum optics with “thick” samples

- Storing and “shaping” single photons
- Single-photon entanglement

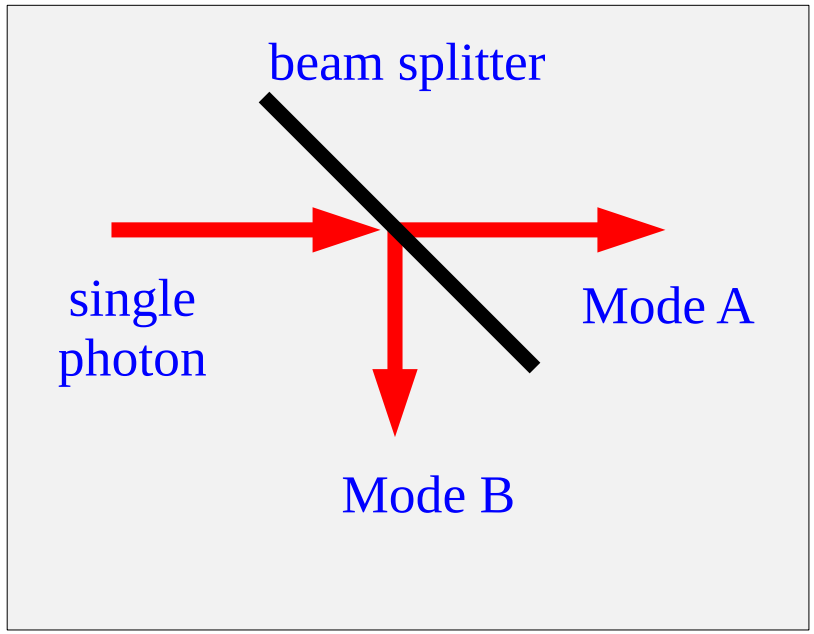
Introducing single-photon entanglement

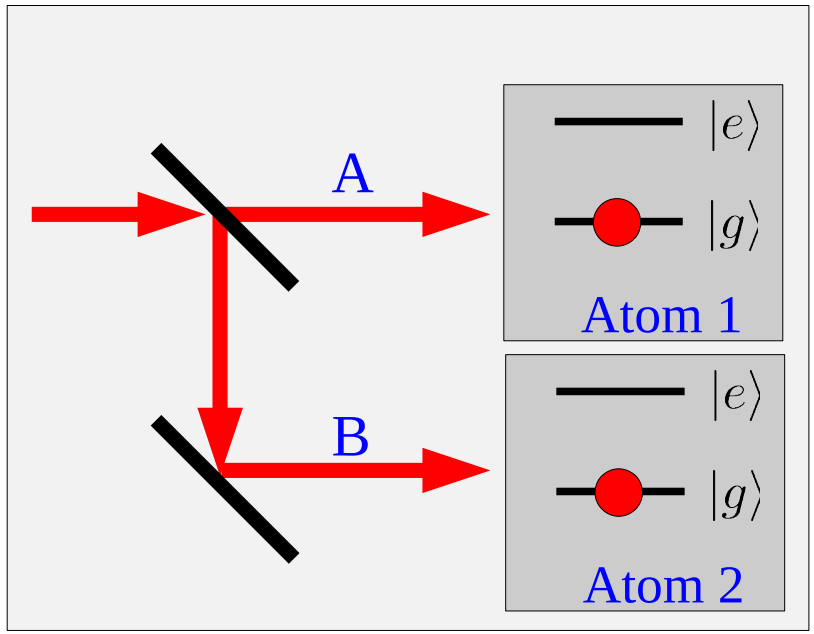
- Single photon impinging on 50/50 beam splitter gives output
$$|\Psi\rangle = \frac{1}{\sqrt{2}}\left(|0\rangle_A|1\rangle_B + |1\rangle_A|0\rangle_B\right)$$

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

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

- Can be converted to other forms, e.g. “regular” entanglement between atoms
$$|\Psi\rangle = \frac{1}{\sqrt{2}}\left(|g_1 e_2\rangle + |e_1 g_2\rangle\right)$$

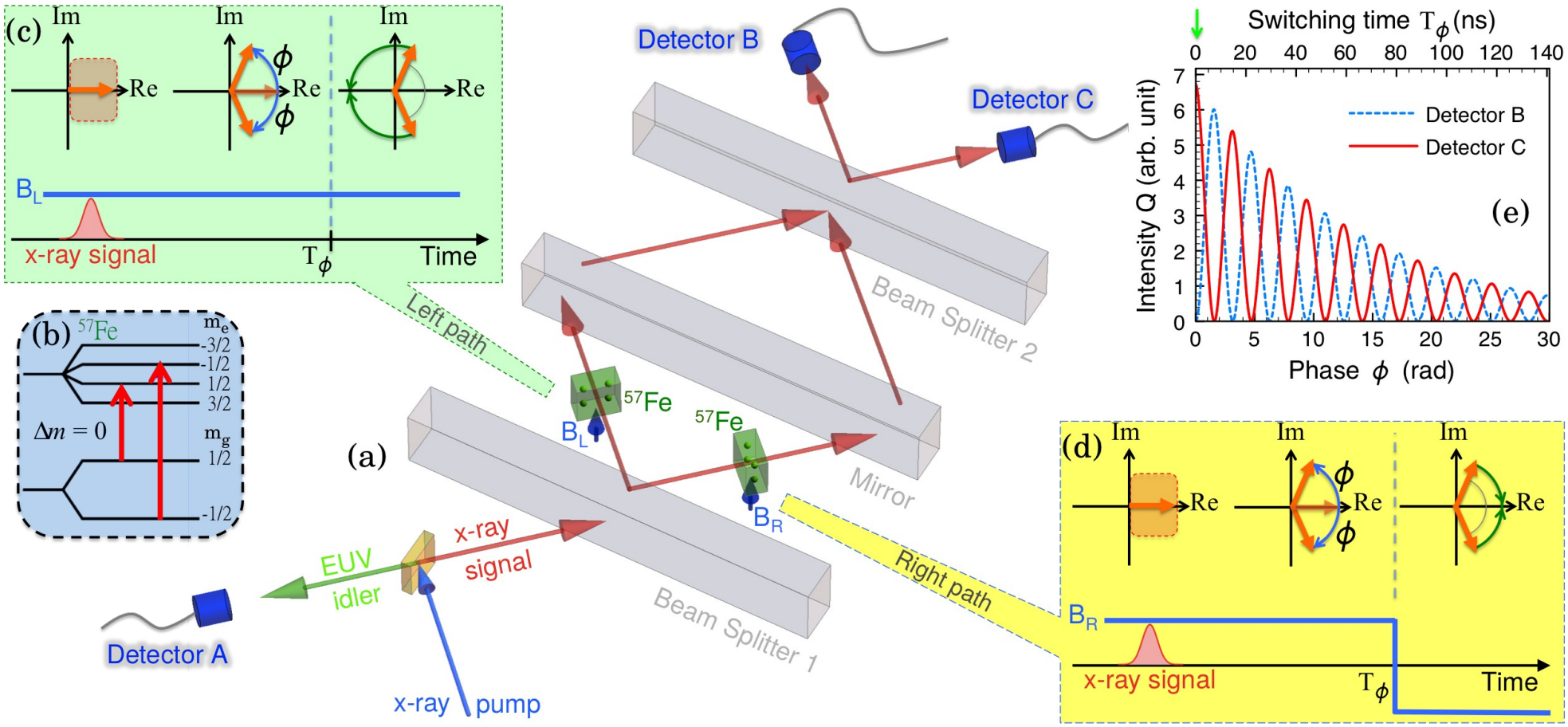




S. J. van Enk, **Phys. Rev. A 67, 022303 (2003)**

Entangling instead macroscopical samples

for decoherence test, boundary between classical & quantum



→ check entanglement via quantum state tomography

W.-T. Liao, C. H. Keitel and AP, arXiv:1407.3292 (2014)

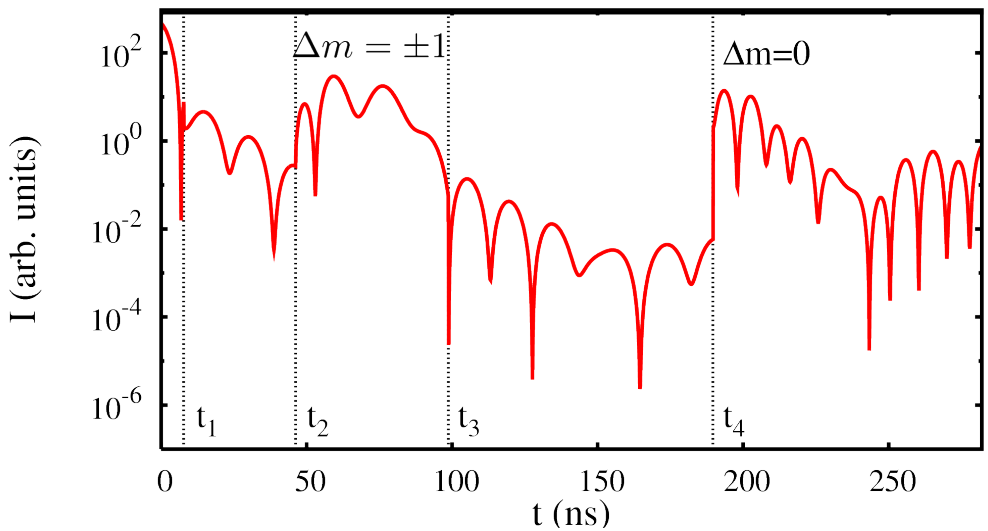
Polarization entanglement

switching time controls light polarization

$0 < t < t_1$ σ-polarized

$t_2 < t < t_3$ π-polarized

$t_4 < t$ σ-polarized



AP, C. H. Keitel and J. Evers, **PRL 103, 017401 (2009)**

AP and J. Evers, **J. Mod. Opt. 57, 1993 (2010)**

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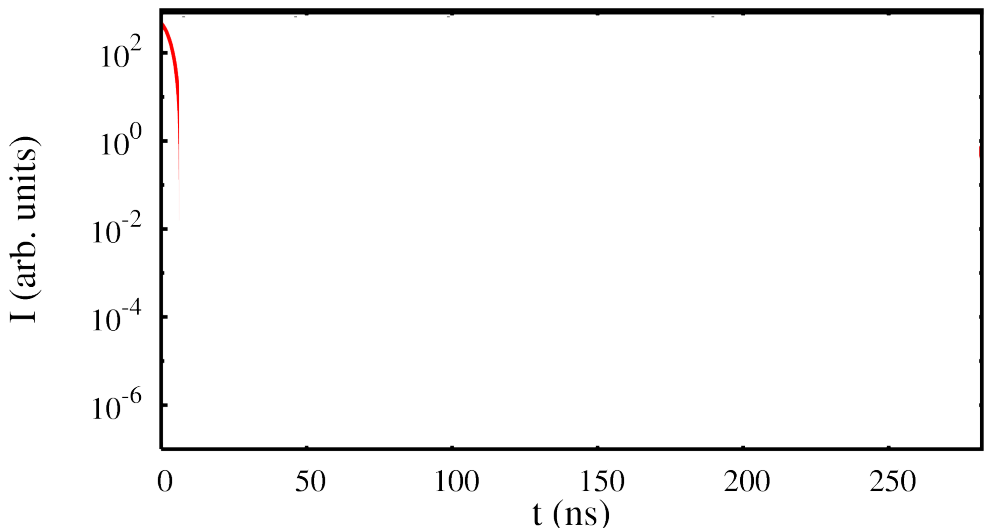

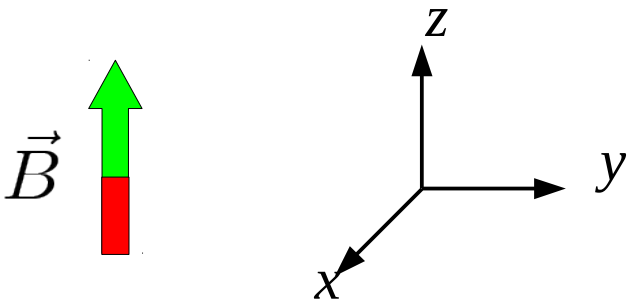

AP, C. H. Keitel and J. Evers, **PRL 103, 017401 (2009)**

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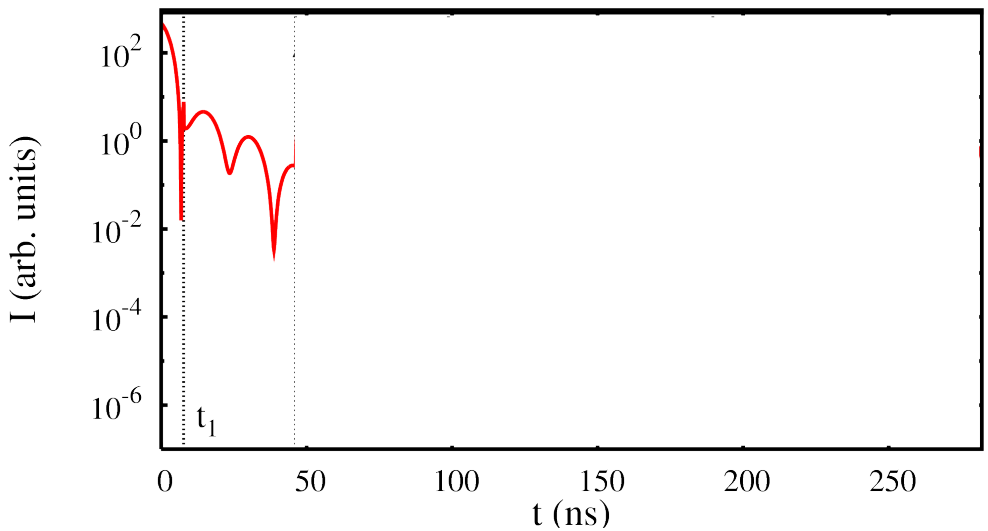

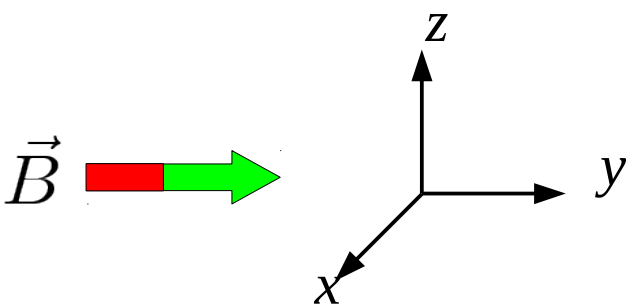

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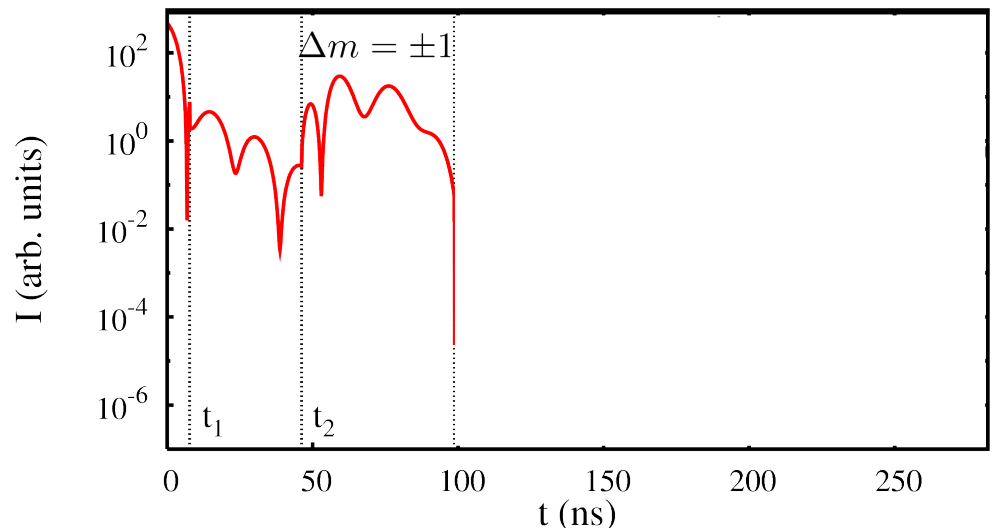

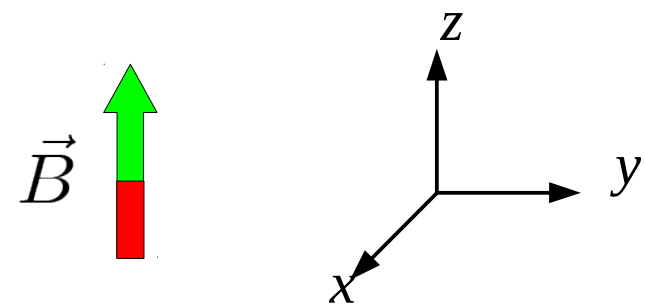

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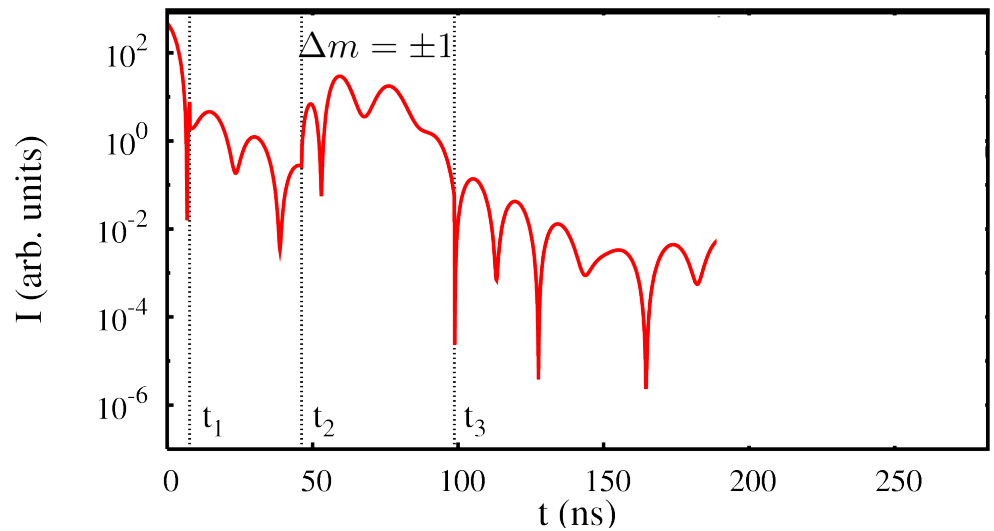

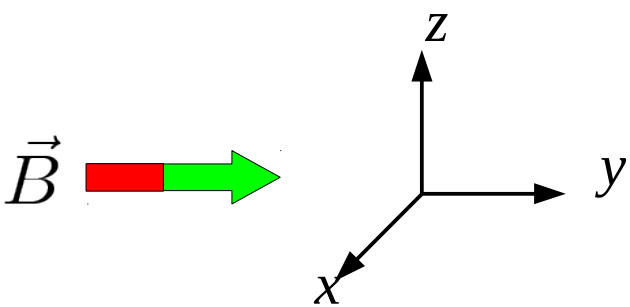

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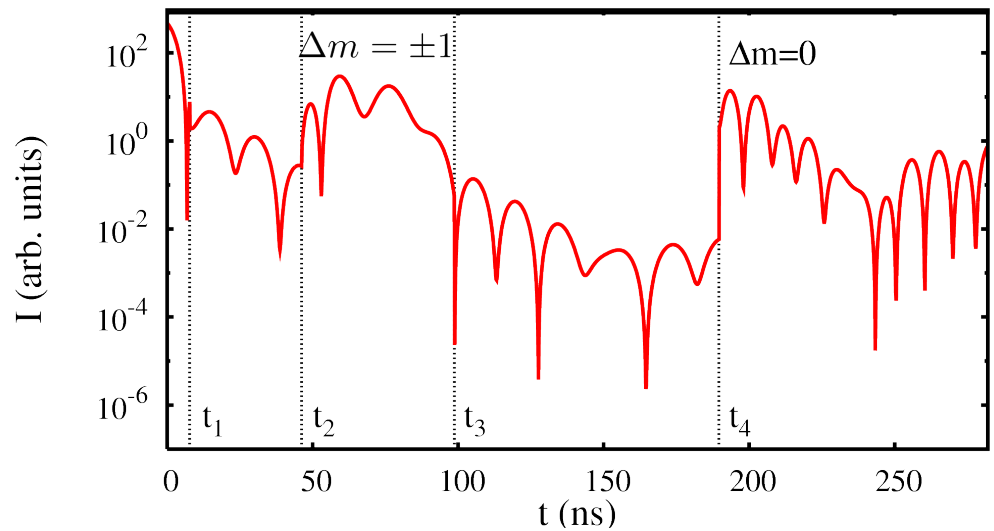

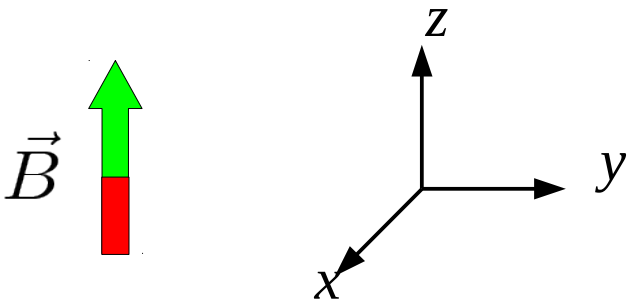

AP, C. H. Keitel and J. Evers, **PRL 103, 017401 (2009)**

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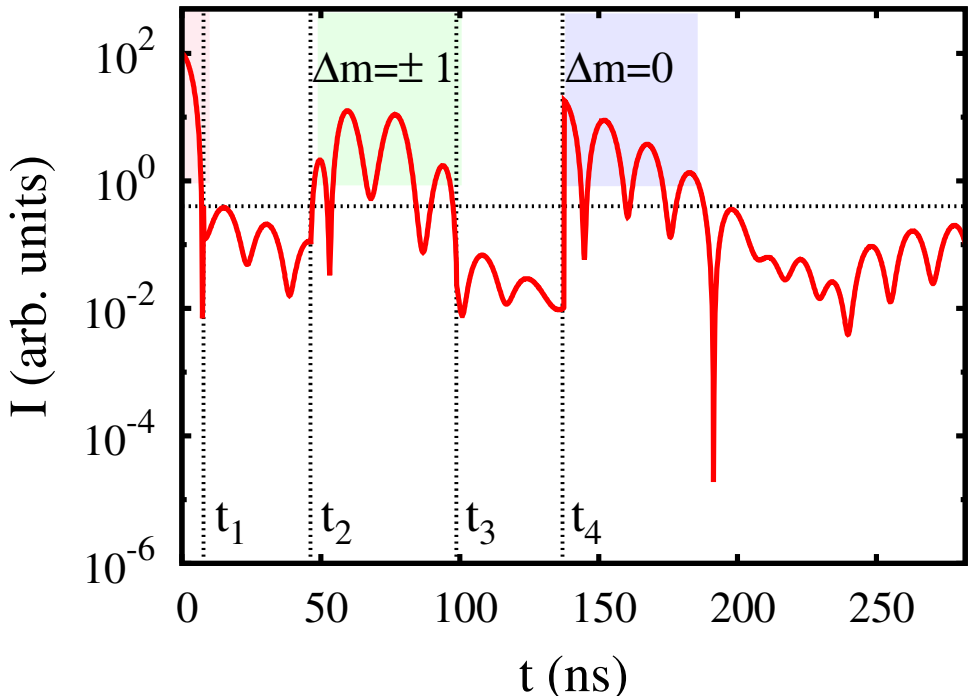
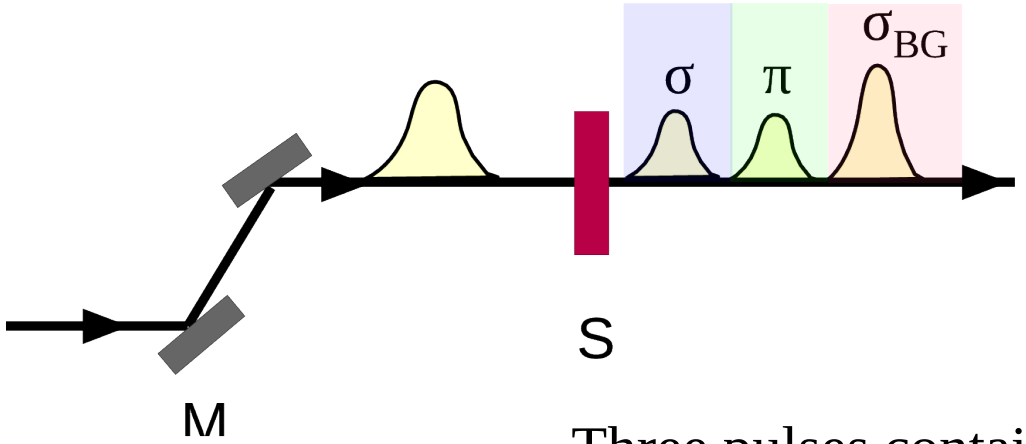
single-photon entanglement with keV photons



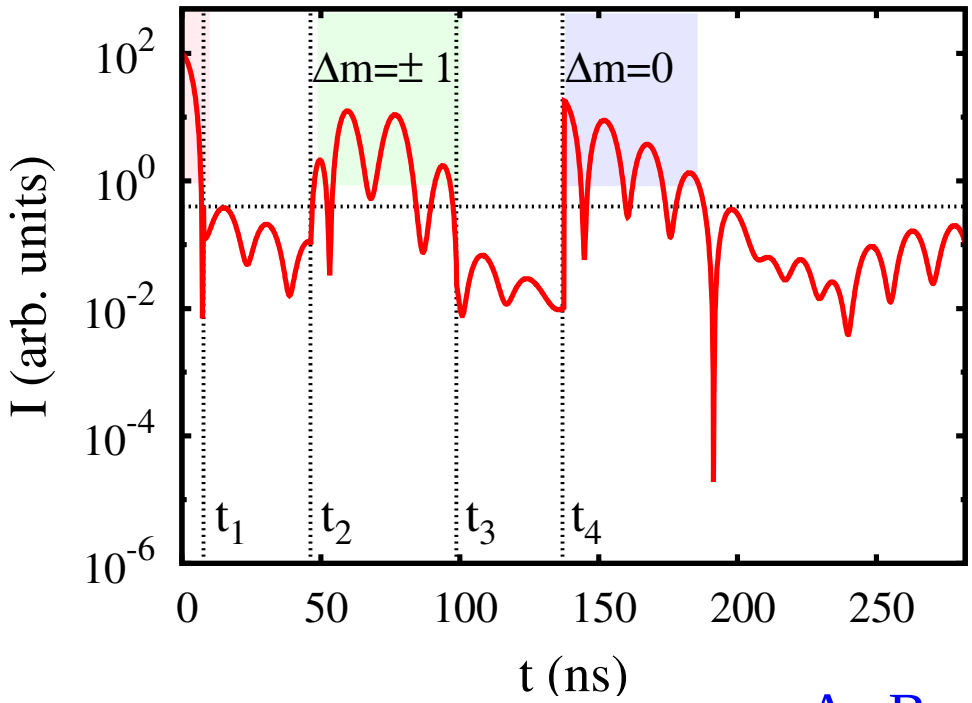
AP, C. H. Keitel and J. Evers,
PRL 103, 017401 (2009)



Three pulses contain one single photon*!

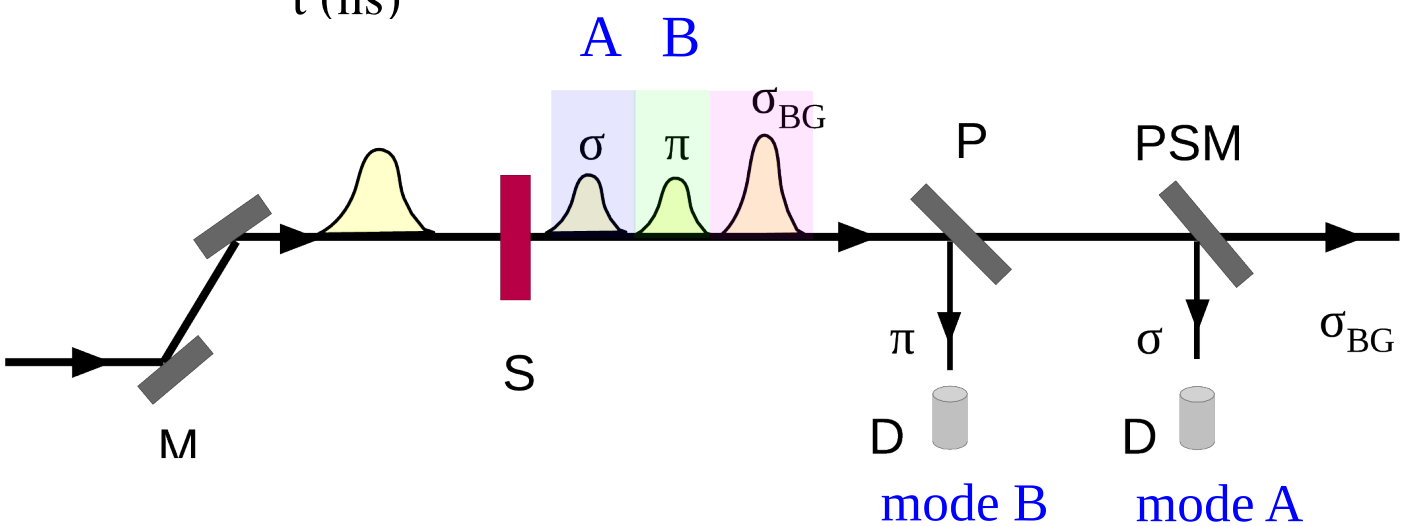
*not really a Fock state unless postselection!

single-photon entanglement with keV photons



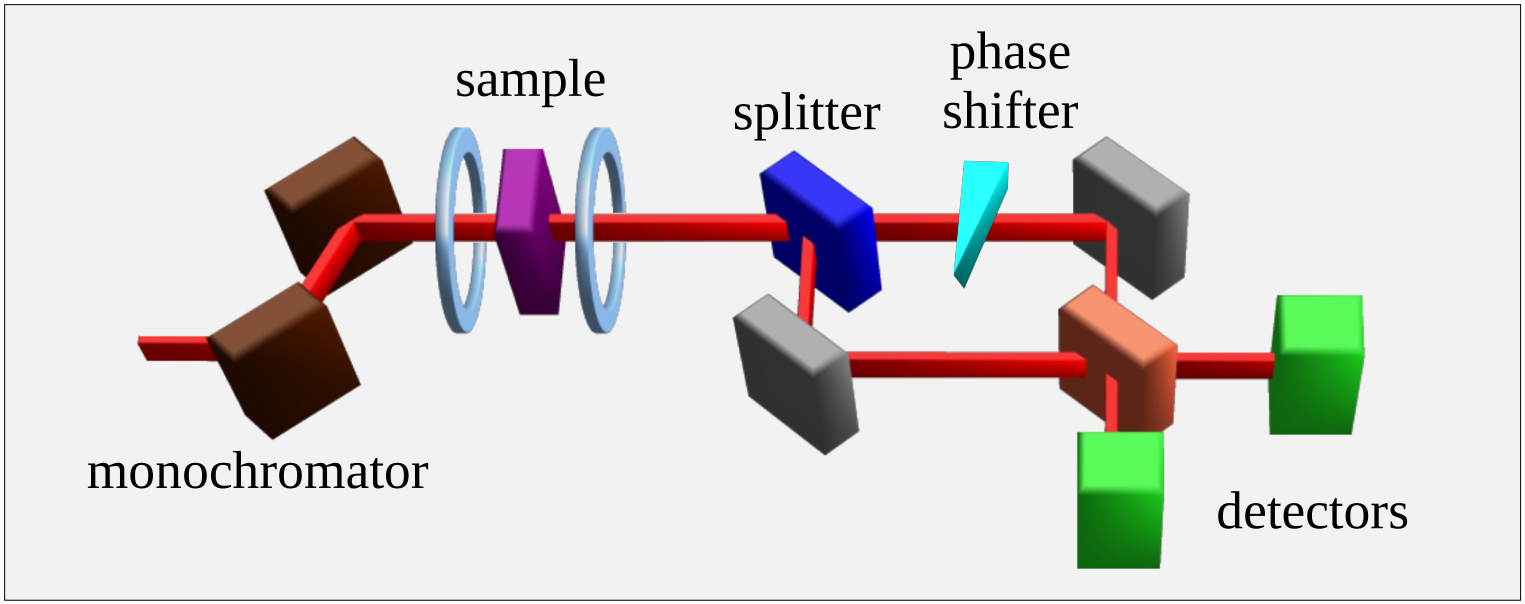
AP, C. H. Keitel and J. Evers,
PRL 103, 017401 (2009)

$$|\Psi\rangle = \alpha|0\rangle_A|1\rangle_B + \beta|1\rangle_A|0\rangle_B$$



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



courtesy Jörg Evers

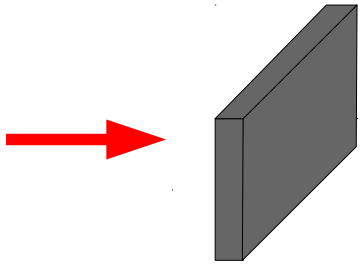
*) H.-W. Lee and Kim, Phys. Rev. A 63, 012305 (2000)

X-ray nuclear quantum optics with “thin” samples

- The beauty of nanocavities
- Lamb shift, EIT, SGC

Thin vs. thick

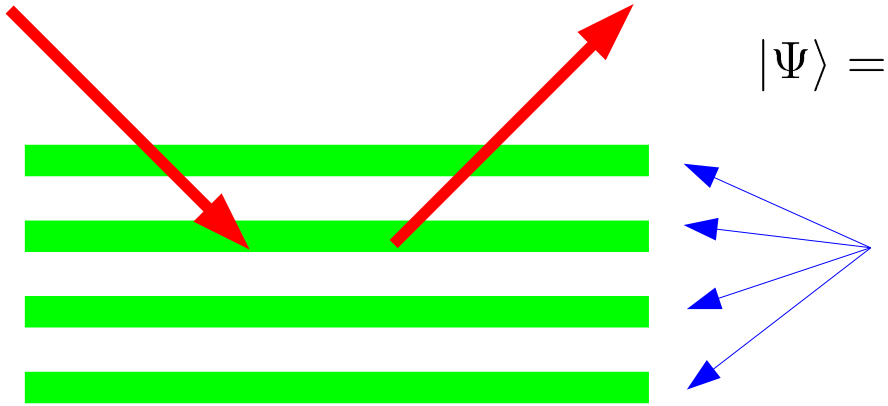
“Standard” forward scattering with thick target



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

Light scattering on structured targets

- In Bragg geometry, a single radiative eigenmode is excited

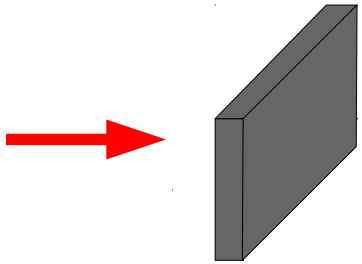


$$|\Psi\rangle = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} e^{i\vec{k}\vec{r}_i} |g_1, \ldots, g_{i-1}, e_i, g_{i+1}, \ldots, g_N\rangle$$

timed Dicke state

Thin vs. thick

“Standard” forward scattering with thick target



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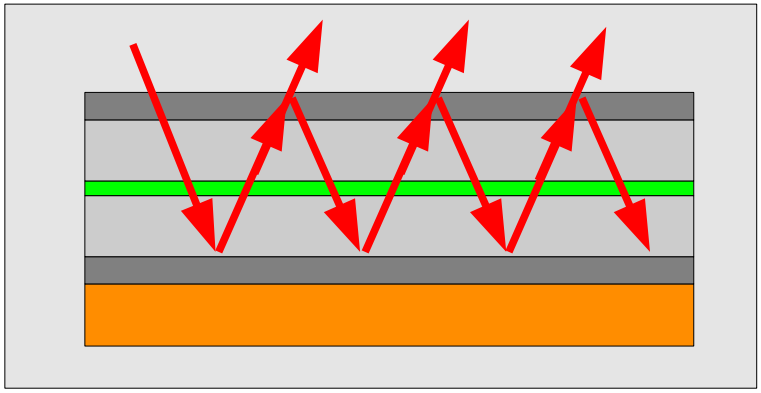
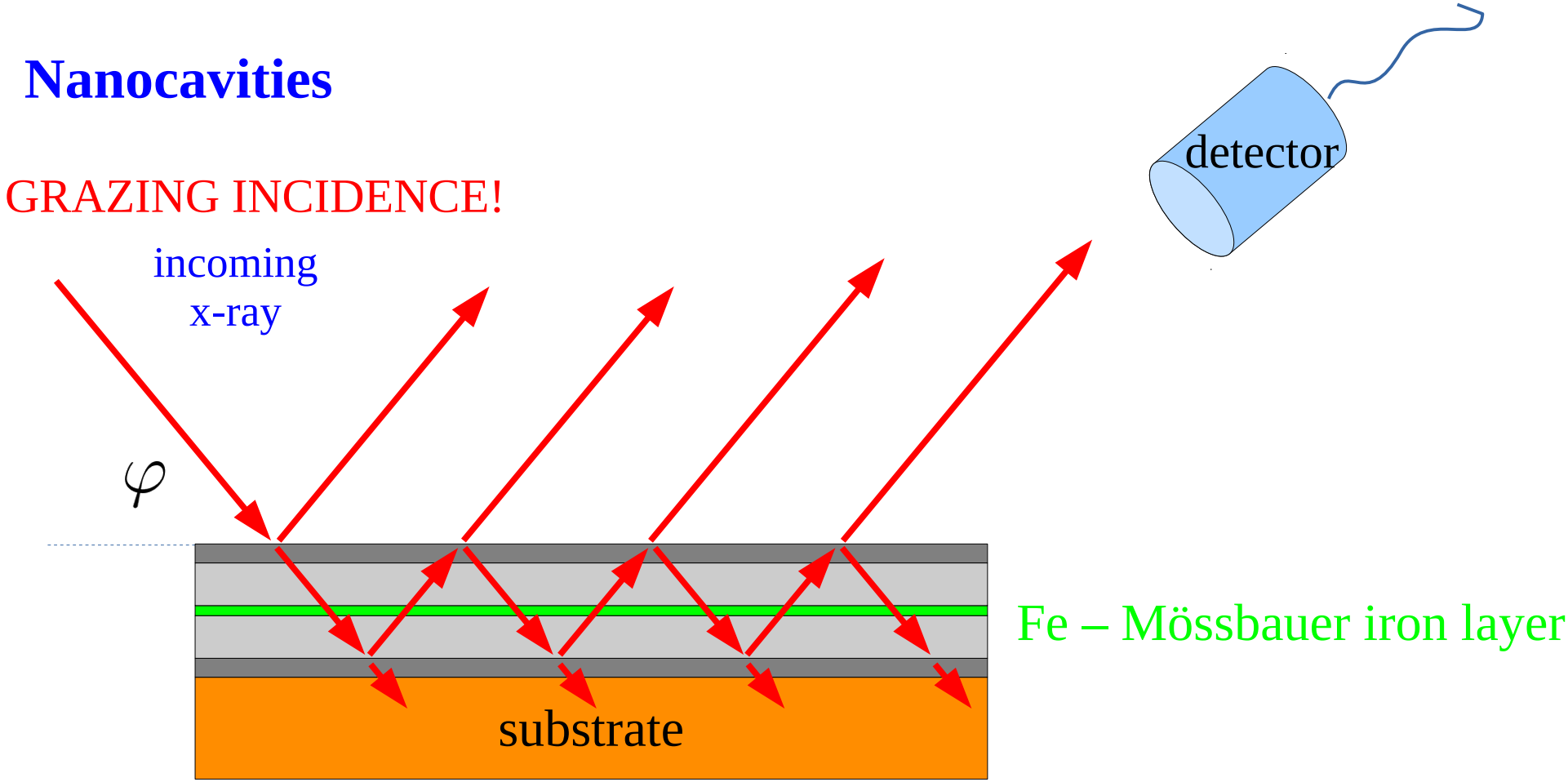


planar cavities effectively realize Bragg case

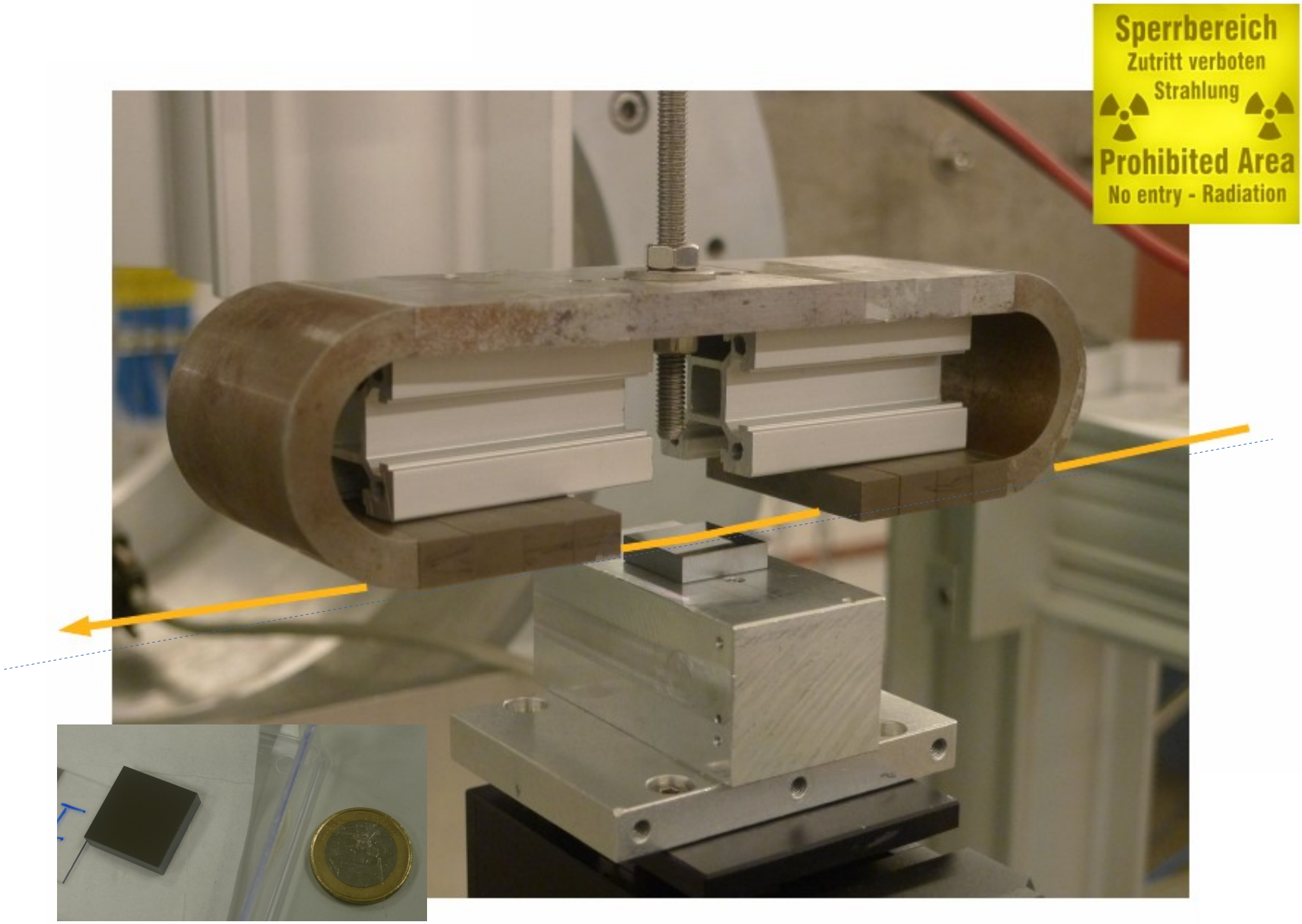
The cavity geometry allows to selectively excite a single radiative eigenmode→ purely exponential decay

Nanocavities



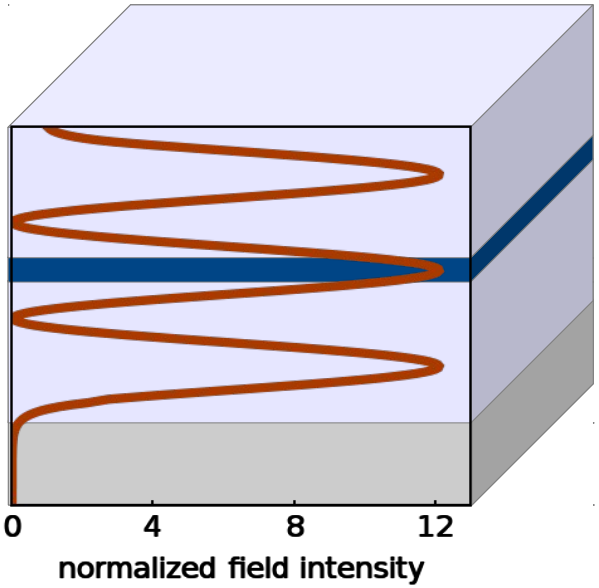
- nm-sized thin film cavity
- Cavity resonances give field enhancement
- Nuclear resonances in Fe can interact with cavity field, observable in reflection

Experiments at Petra III



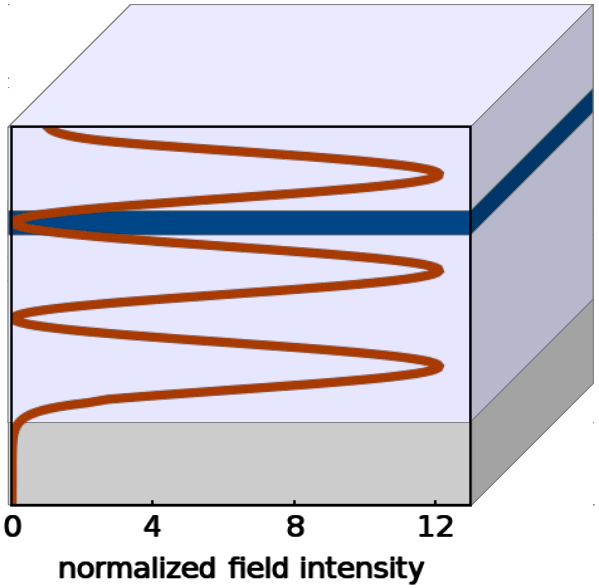
courtesy Jörg Evers

Positioning in the cavity



Iron nuclei strongly couple to cavity field

- Accelerated decay
- High excitation probability



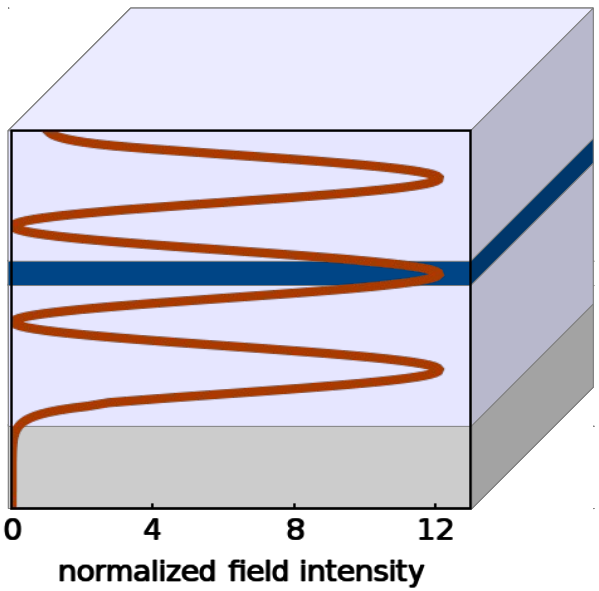
Iron nuclei weakly couple to cavity field

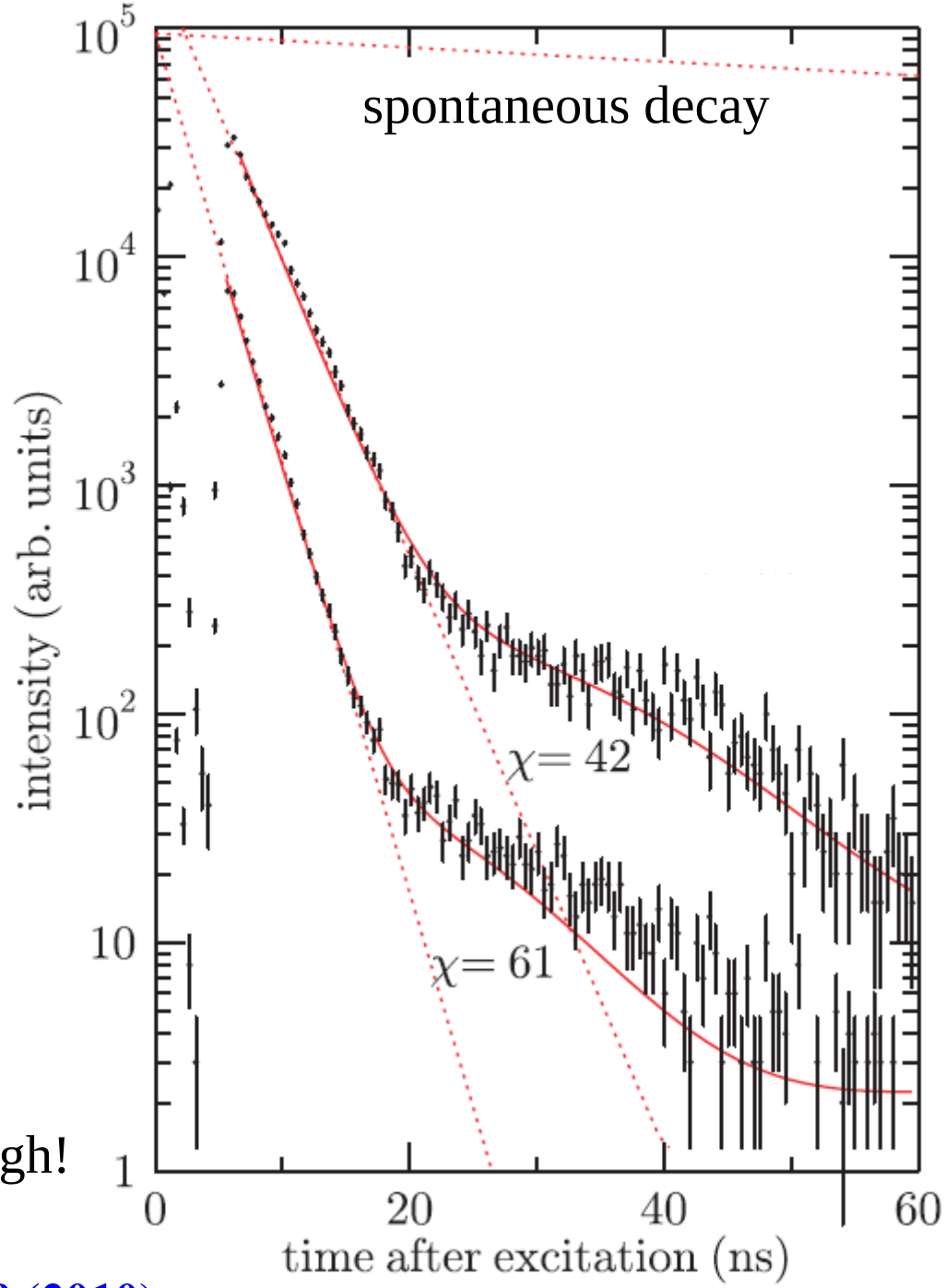
- Decelerated decay
- Low excitation probability

Effective properties of
the nuclei can be tailored!

courtesy Jörg Evers

Faster exponential decay





χ is the coherent decay enhancement factor

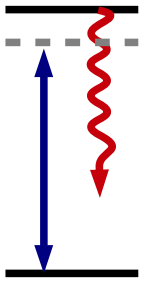
… even here not forever, but long enough!

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

X-ray nuclear quantum optics with “thin” samples

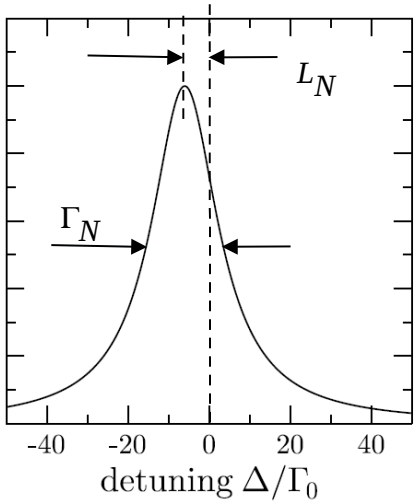
- The beauty of nanocavities
- Lamb shift, EIT, SGC

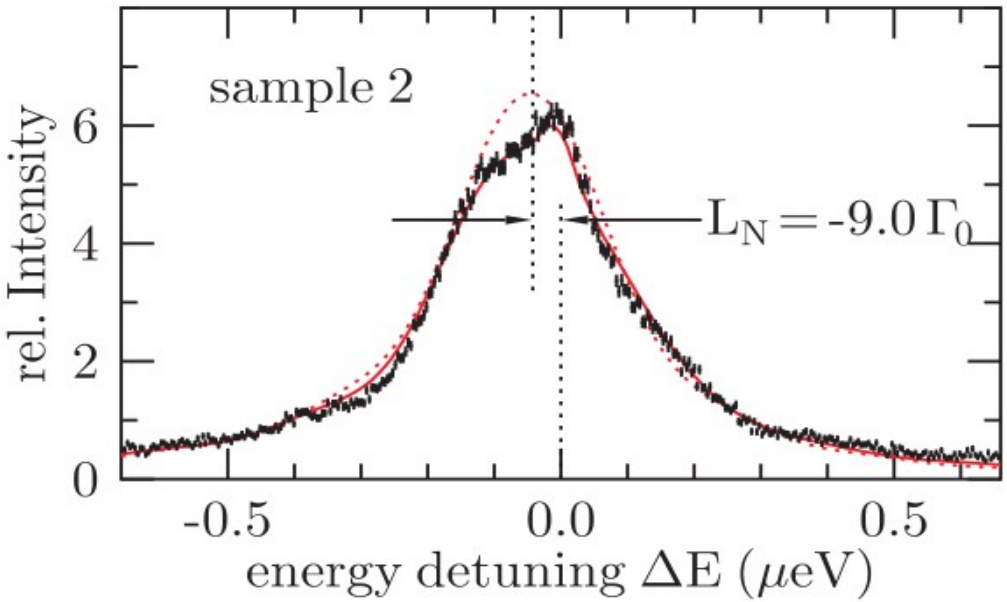
Collective Lamb shift



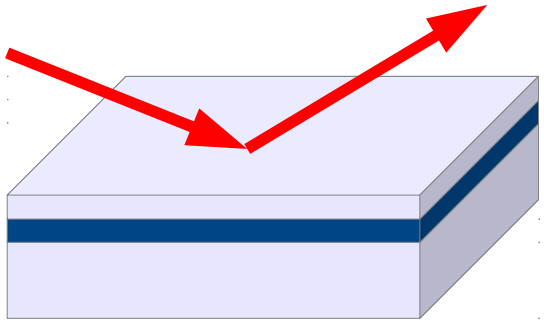
Lamb shift – interaction with virtual photons

Collective Lamb shift – interaction with virtual photons between identical nuclei



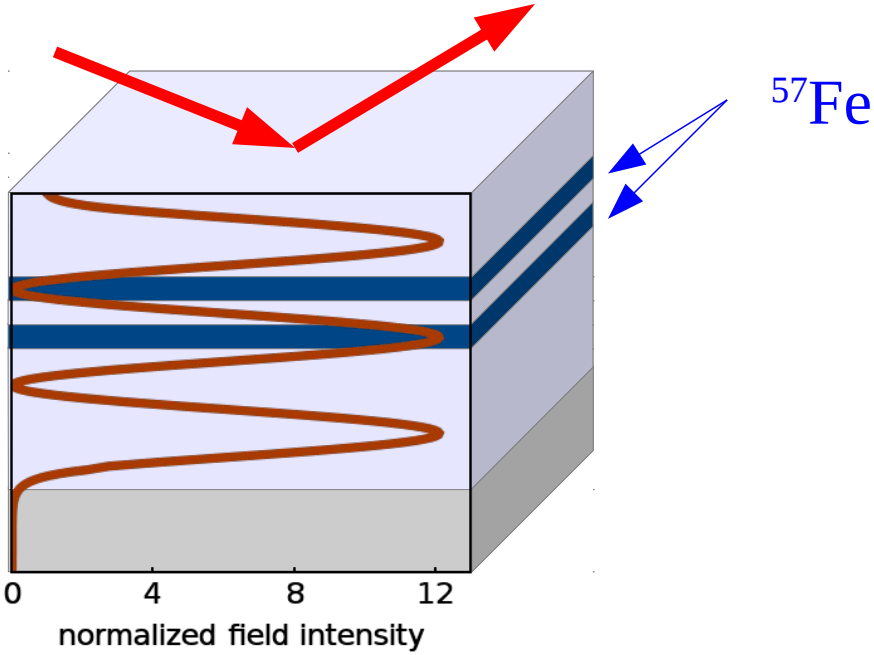


$$L_N \sim N\left[\frac{\lambda}{R}\right]^3 \Gamma_0 = \rho\lambda^3\Gamma_0$$

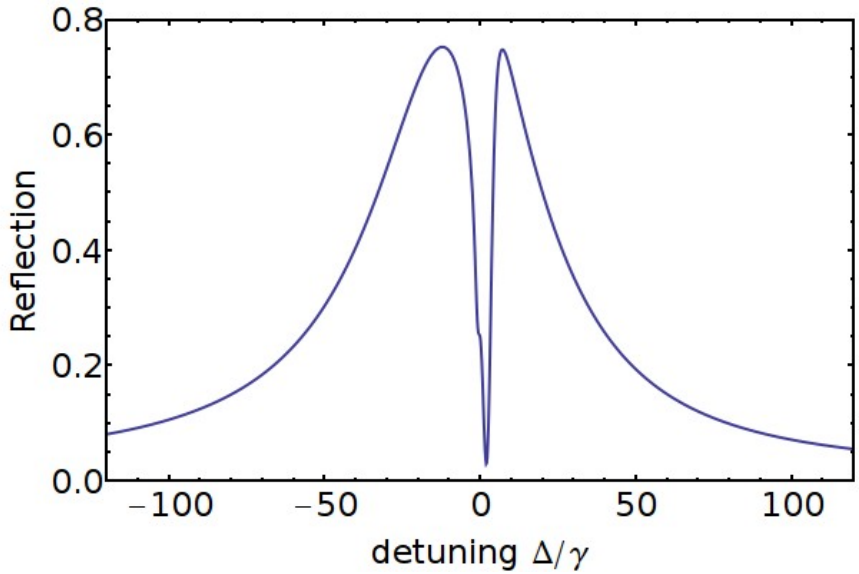


R. Röhlsberger et al., Science 328, 1248 (2010)

Two iron layers



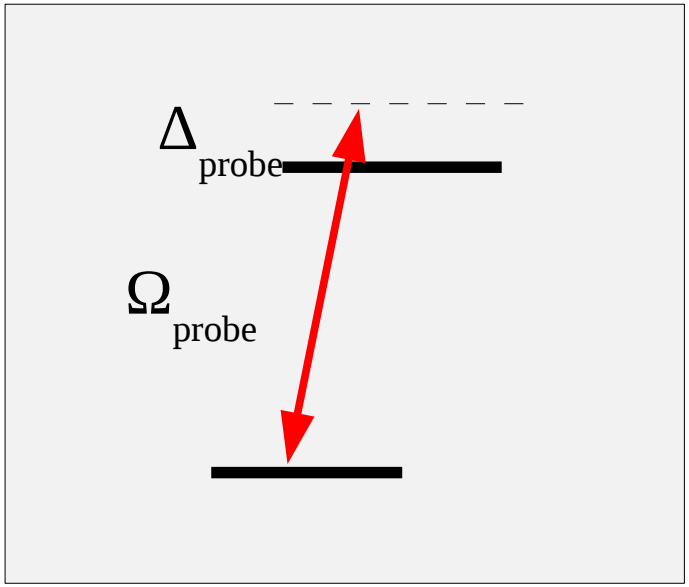
Reflection spectrum

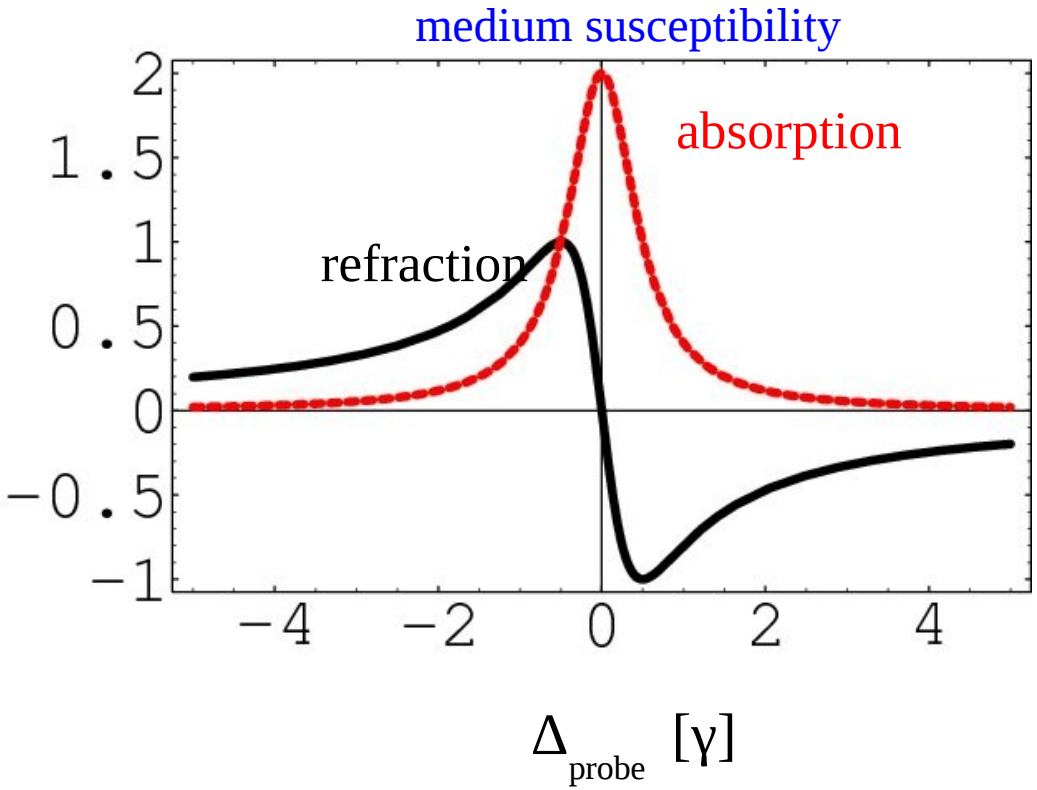


Looks like EIT!

R. Röhlsberger et al., Nature 482, 199 (2012)

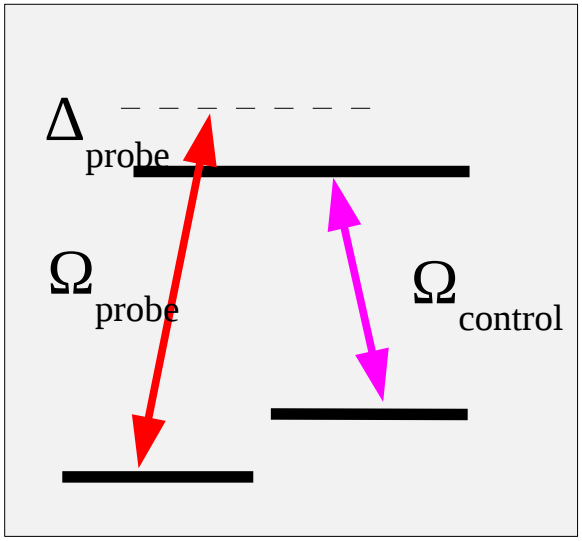
Optical response of a single resonance

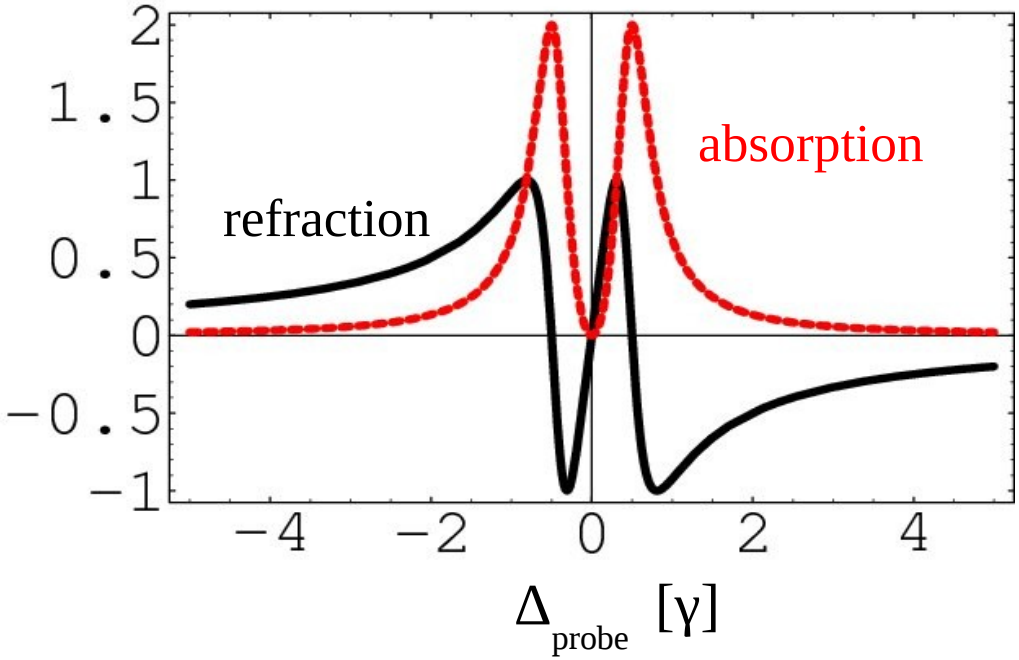




Electromagnetically induced transparency

Three-level Λ system



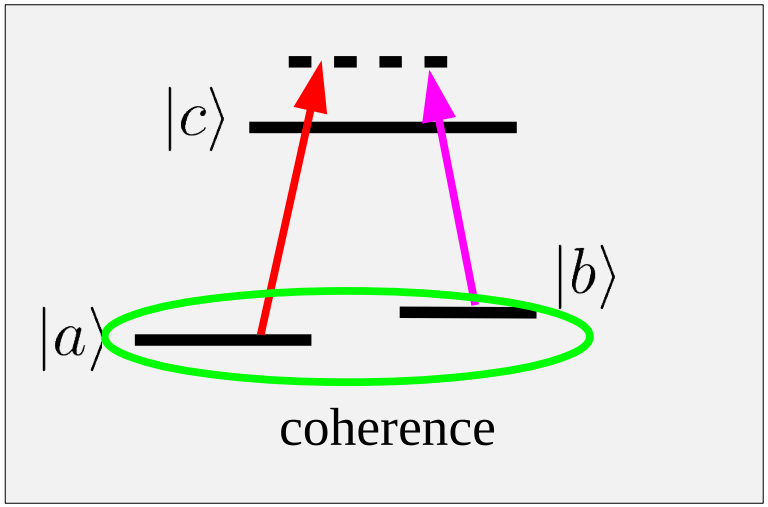


Medium is rendered transparent by shining light on it!

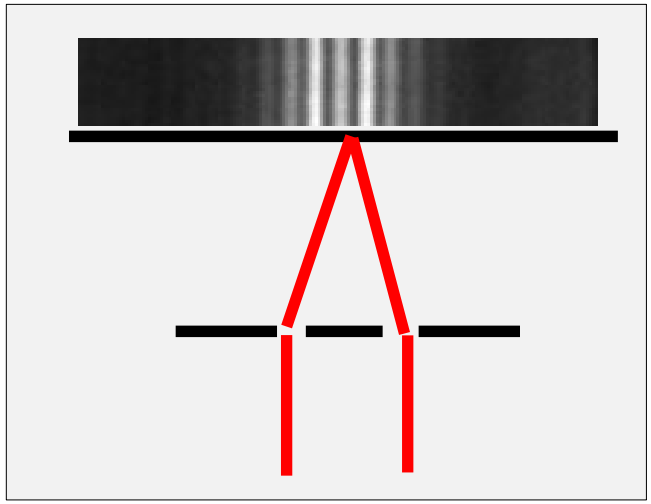
S. Harris, Physics Today 50, 36 (1997); M. Fleischhauer et al., Rev. Mod. Phys. 77, 633 (2005)

Electromagnetically induced transparency

Interpretation as coherence/interference effect:



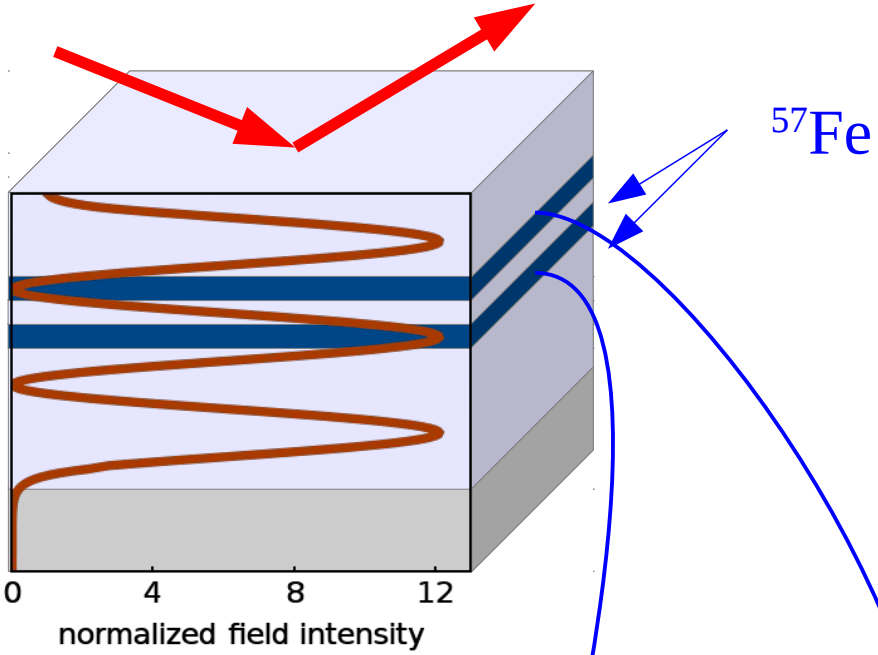
EIT



double slit

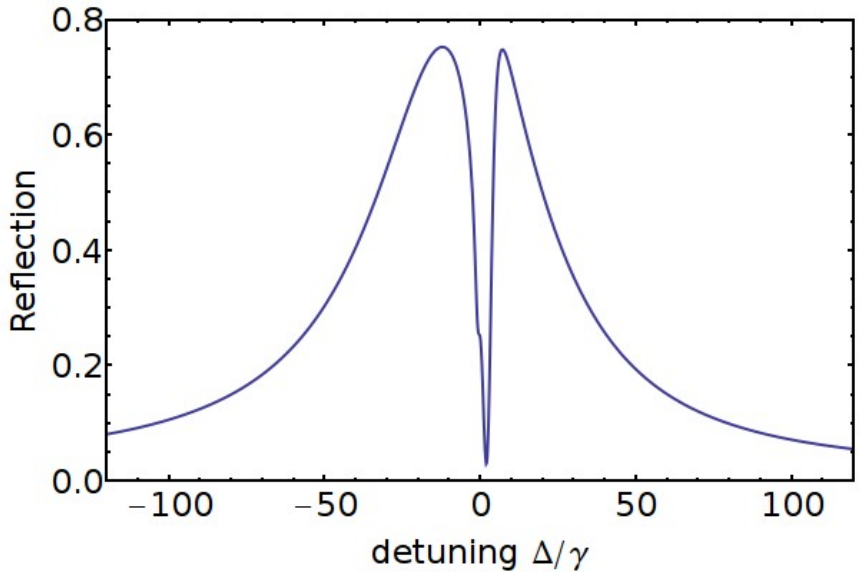
no excitation of the atom due to destructive interference

Two iron layers

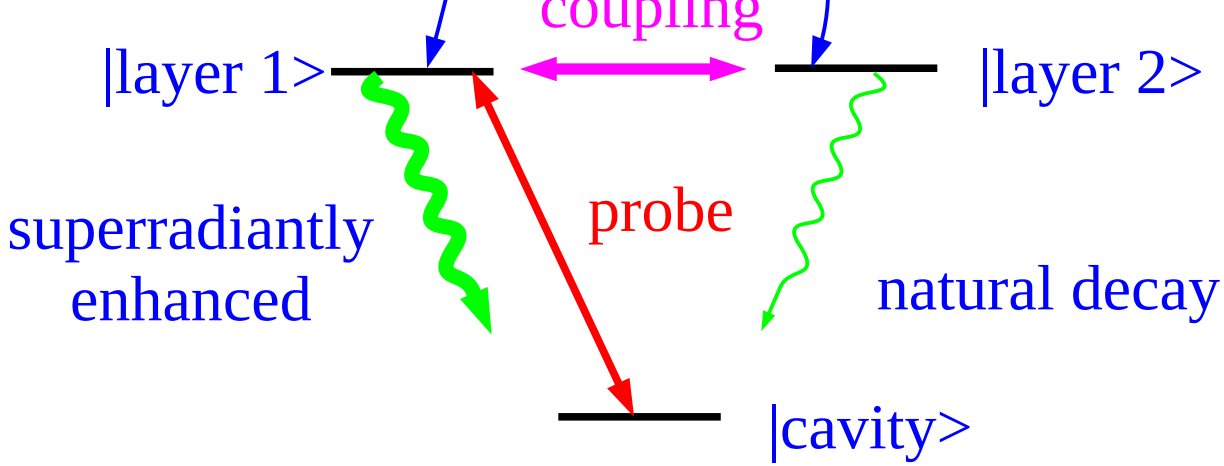


^{57}Fe

Reflection spectrum



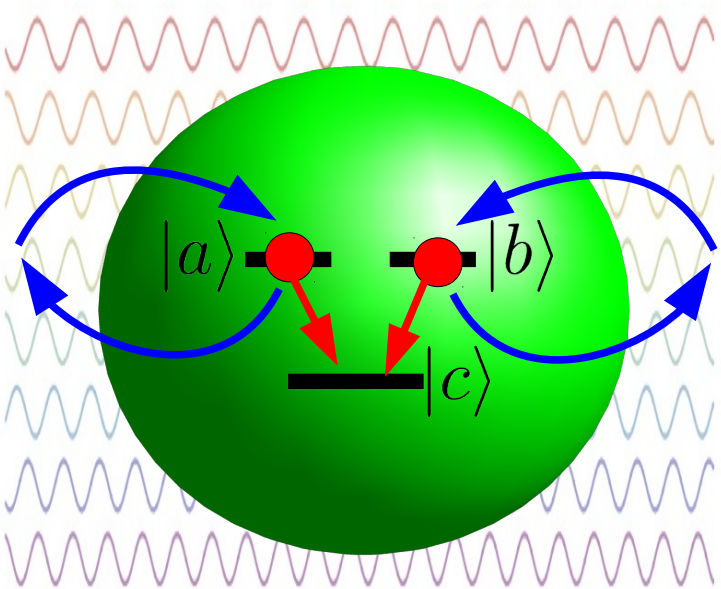
It is EIT!

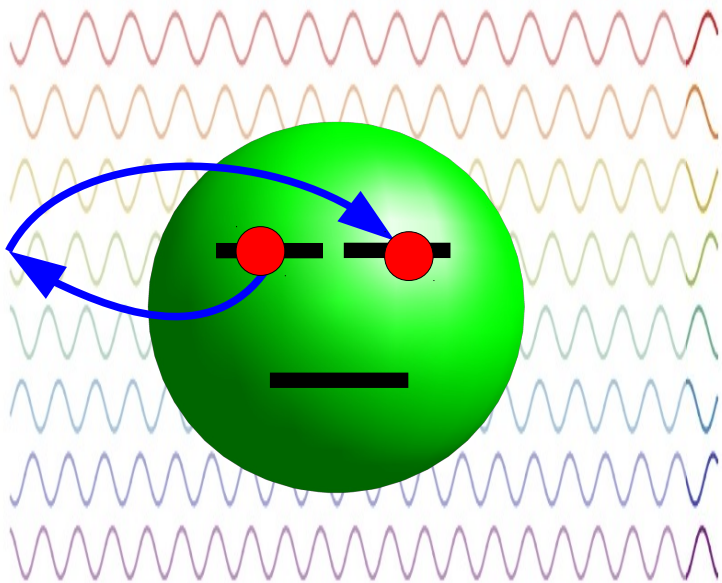


same nuclei acquire different properties

R. Röhlsberger et al., Nature 482, 199 (2012)

Spontaneously generated coherence





interaction with vacuum modes
creates decoherece – spontaneous decay

in very special cases,
interaction with vacuum
can bring coherence!

Condition I

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

non-orthogonal
dipole moments

Condition II

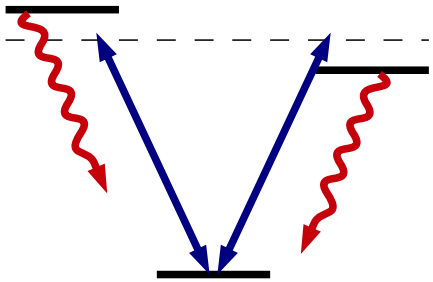
$$E_1 \approx E_2$$

approx. same
transition energy

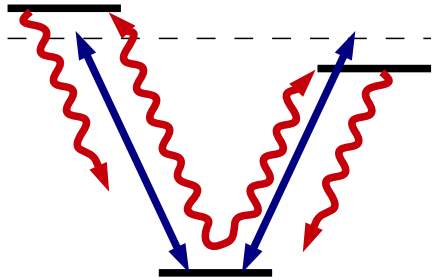
courtesy Jörg Evers

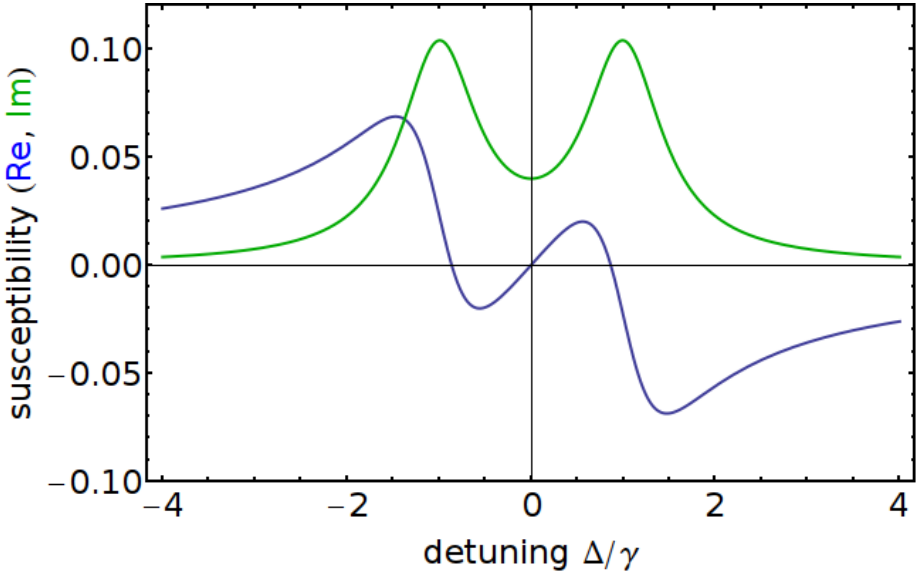
Susceptibility

Without SGC

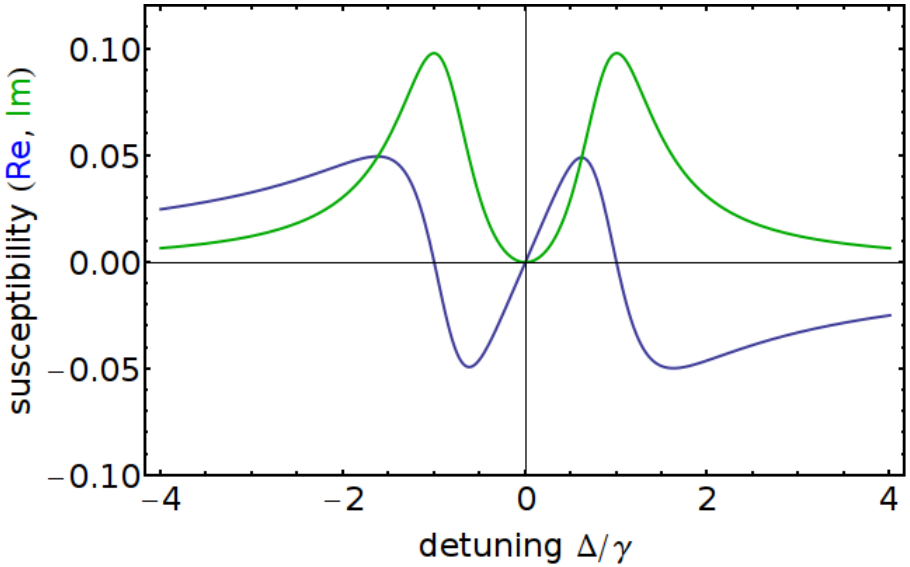


With SGC





Incoherent sum
of two resonances



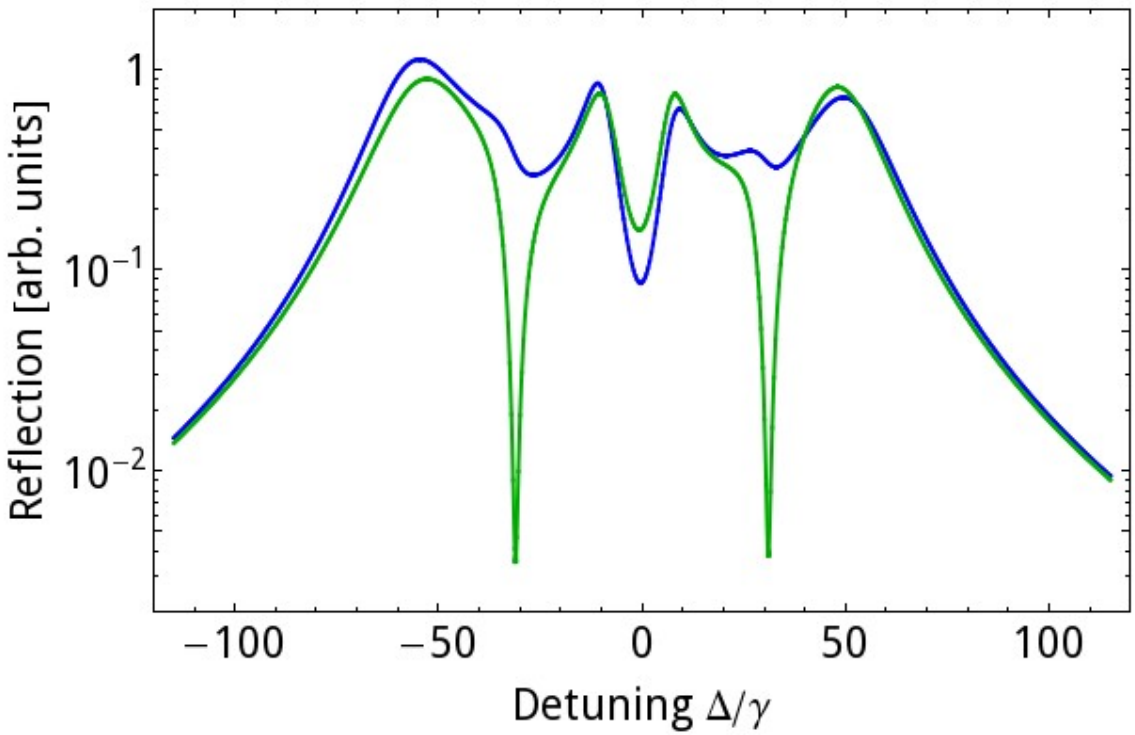
Interference

courtesy Jörg Evers

S.Y. Zhu, R.C.F. Chan, C.P. Lee, Phys. Rev. A 52, 710 (1995)

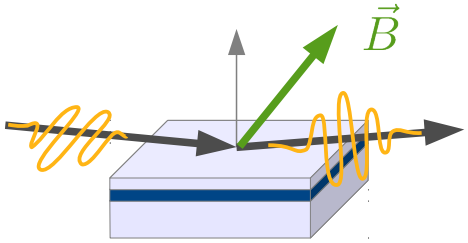
In theory

Reflectance (log scale)



With SGC

Without SGC



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

Detuning

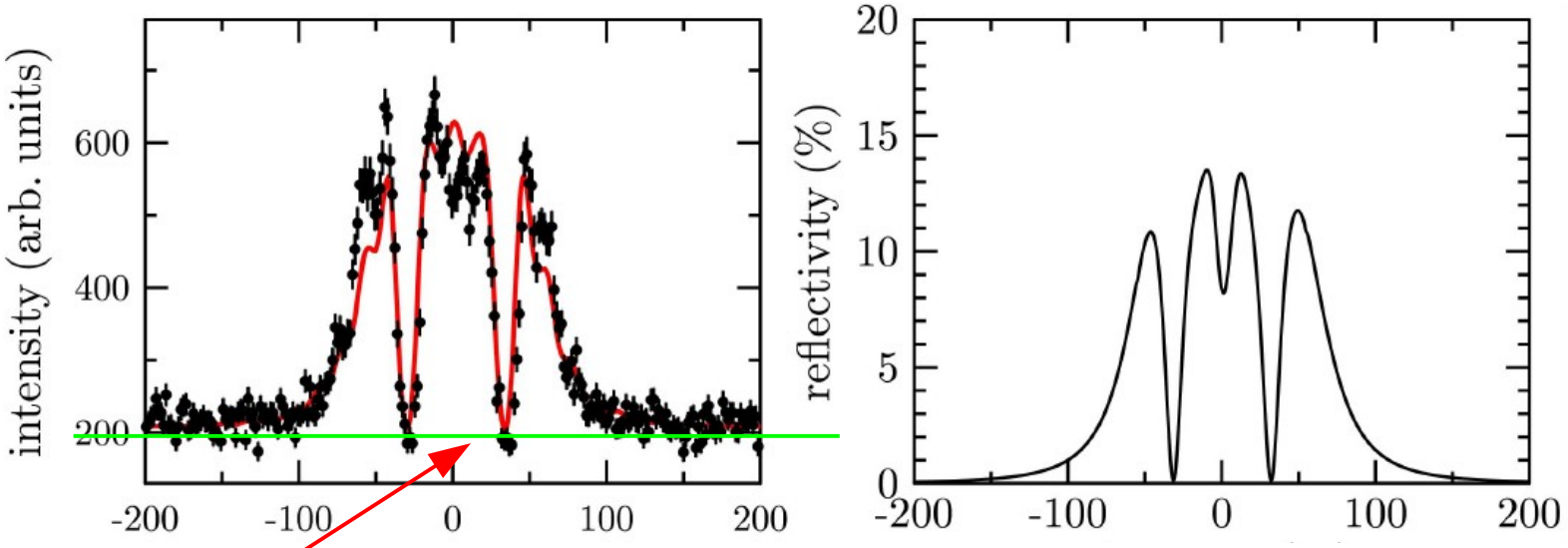
Identify and separate all contributing processes
Spectral “dips” are due to SGC

K. P. Heeg et al, Phys. Rev. Lett. 111, 073601 (2013)

courtesy Jörg Evers

In experiment!

Groups of Jörg Evers (Heidelberg) and Ralf Röhlsberger (DESY)



really zero -
clean system!

essentially
no decoherence

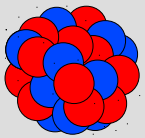
crucial advantage compared
to other quantum optical setups

K. P. Heeg et al, Phys. Rev. Lett. 111, 073601 (2013)

courtesy Jörg Evers

Vision

X-ray nuclear quantum optics

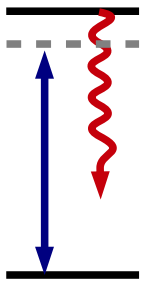


"clean" systems

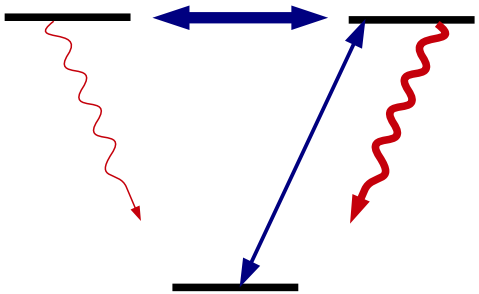
unperturbed by environment

Q factor – ratio transition energy/width

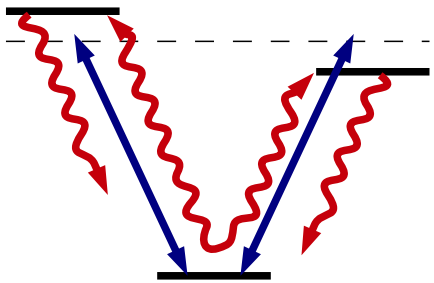
very successful quantum optics at a single-photon level



Collective Lamb shift



EIT



Spontaneously generated coherences

The future

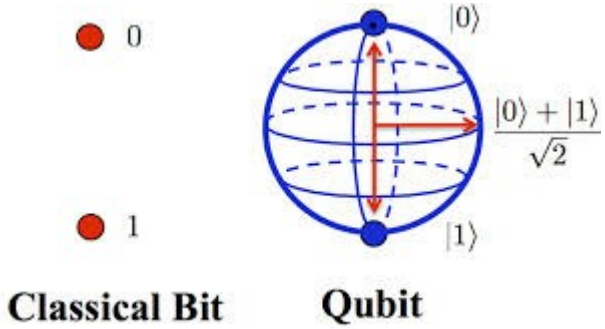
Metrology

Precision experiments @ x-ray time/space scales

nuClock

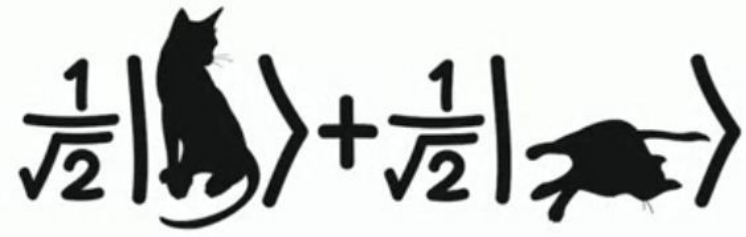
Photon qubits

Photonic circuits



Decoherence studies

Boundary between quantum and classical

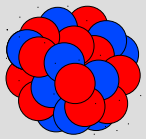


Help for phase reconstruction in imaging?

Iron is everywhere!

Conclusions

very successful quantum optics at a single-photon level

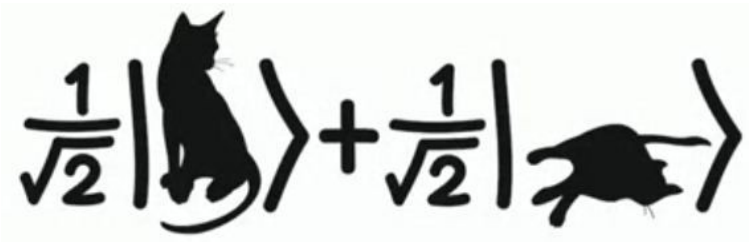


"clean" systems

unperturbed by environment

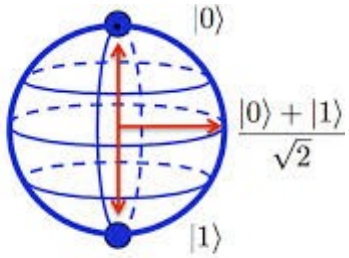
Q factor – ratio transition energy/width

exciting possibilities for the future



Iron is everywhere!

nuClock

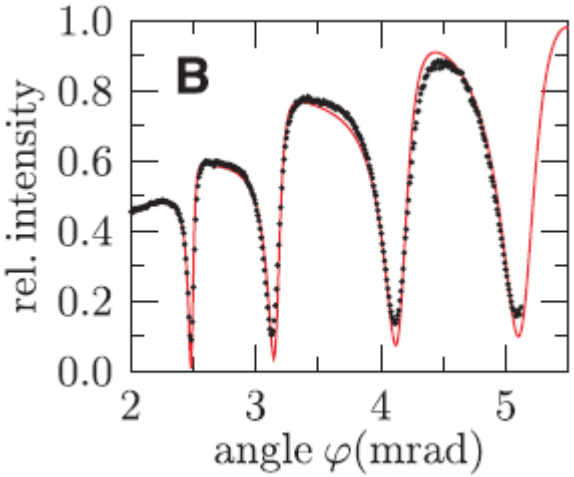


Qubit

THANK YOU FOR YOUR ATTENTION!

Extra

Reflectivity



Direct photoexcitation

Parameter	LCLS	SACLA	European XFEL	XFELO
E_{max} (eV)	10332	19556	24800	25000
BW	$2\text{–}5\times10^{-3}$	$<10^{-3}$	8×10^{-4}	1.6×10^{-7}
T_{pulse} (fs)	100	100	100	1000
T_{coh} (fs)	2	10	0.2	1000
P_{peak} (W)	4×10^{10}	10^{10}	2×10^{10}	4.1×10^{9}
I_{peak} (W/cm^2)	3.9×10^{17}	9.8×10^{16}	2.0×10^{17}	4.0×10^{16}
f_{rep} (Hz)	30	10	4×10^{4}	10^{8}
I_{eff} (W/cm^2)	5.2×10^{9}	1.2×10^{9}	6.5×10^{9}	6.7×10^{12}
ρ_{trig}	1.8×10^{-20}	1.7×10^{-20}	2.4×10^{-21}	4.4×10^{-14}
R_γ (1/s)	5.6×10^{-14}	1.7×10^{-14}	9.6×10^{-12}	4.5×10^{-1}

Acceleration of charged particles

Basic facts

- Radiated power depends on acceleration $P_s \sim \frac{dp_\mu}{d\tau}\frac{dp^\mu}{d\tau}$
- Radiation from non-relativistic motion or from linear acceleration can be neglected
- Radiation from circularly accelerated highly relativistic particles:

$$P_s = \frac{e^2}{3\epsilon_0 (m_0 c^2)^4}\frac{E^4}{R^2}$$

From this:

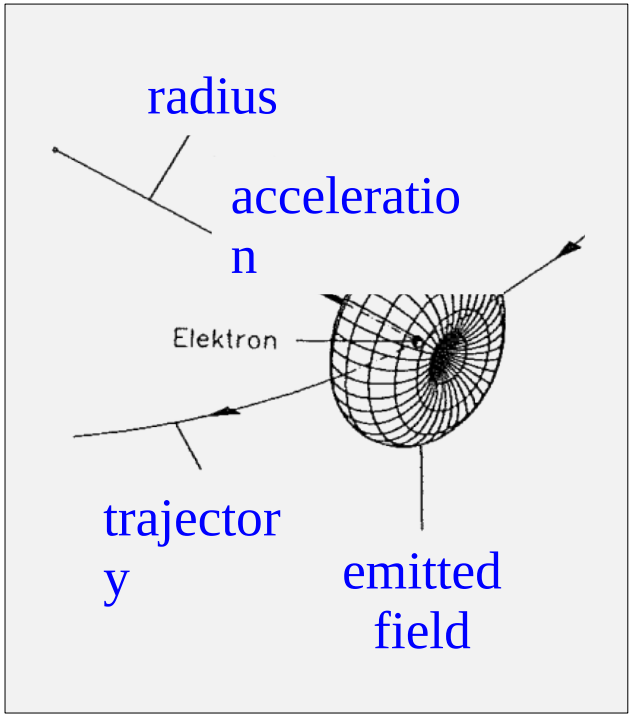
- $P_s \sim E^4/m_0^4$ → need highly relativistic particles
- $P_s \sim m_0^{-4}$ → use light particles to gain high power
- $P_s \sim R^{-2}$ → unwanted radiation loss in bends requires large scale devices (~ km circumference)

use circular accleration of highly relativistic electrons

courtesy Jörg Evers

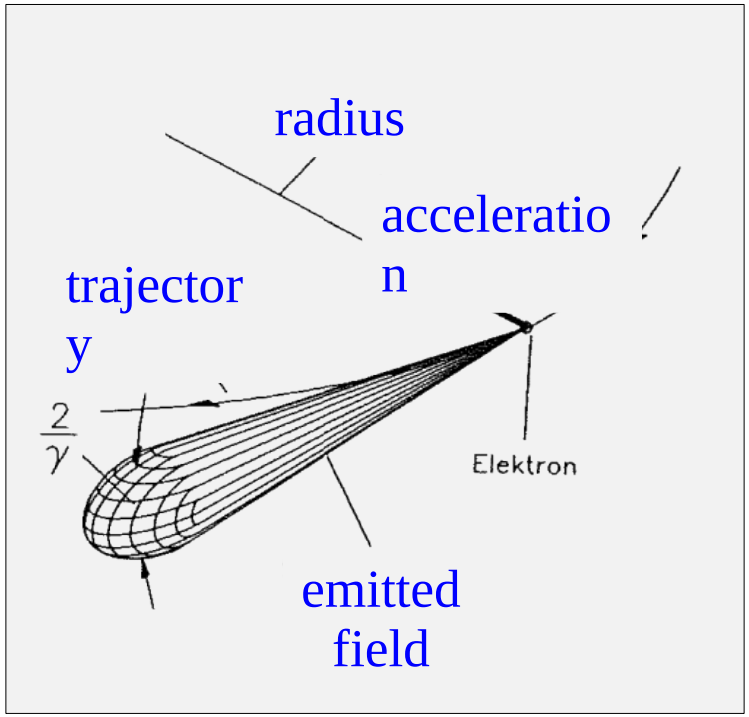
Angular distribution of emitted light

Electron rest frame



dipole emission pattern

Laboratory frame

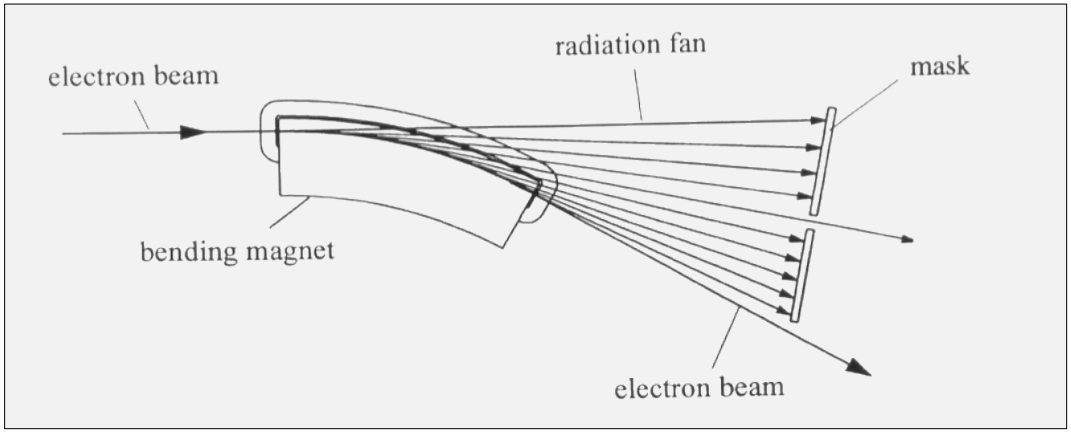


tightly bunched in forward direction due to Lorentz trafo

Images: Book by K. Wille

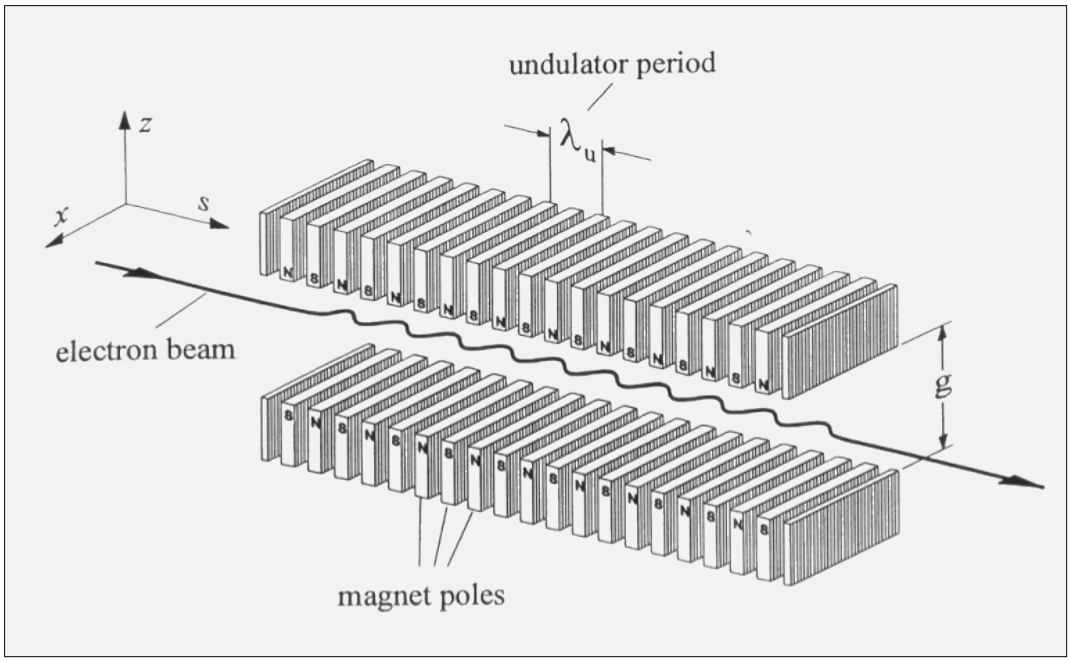
Bending magnets and insertion devices

Bending magnet



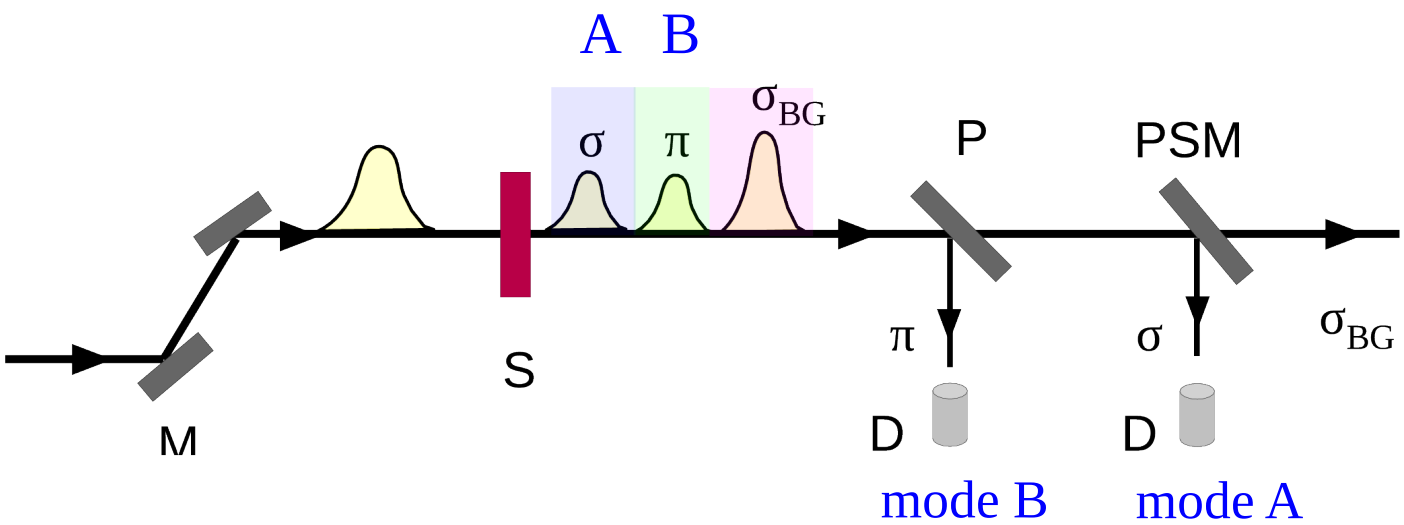
- radiation in beam bends
- Could be used in non-dedicated storage rings
- Large fan of radiation bad for small targets
- Broadband spectrum

Insertion devices



- Magnet arrays in linear sections of dedicated storage rings
- Different emission properties depending on parameters
- Wigglers, undulators, FELs

Images: Book by K. Wille

single-photon entanglement with keV photons

- Generate short photon wave packets to facilitate coincidence measurements
- Extend entanglement research to new parameter ranges, more complex quantum systems (c.f. entanglement of macroscopic objects)
- Photons with higher energy and momentum are considered “more robust”, high detection efficiency