Nonlinear X-ray Optics and Extreme Metrology

Light Matters

Ralf Röhlsberger
DESY

Workshop on Evolution and Control of Complexity: Key Experiments Using Sources of Hard X-rays

Advanced Photon Source, 11. - 13. October 2010

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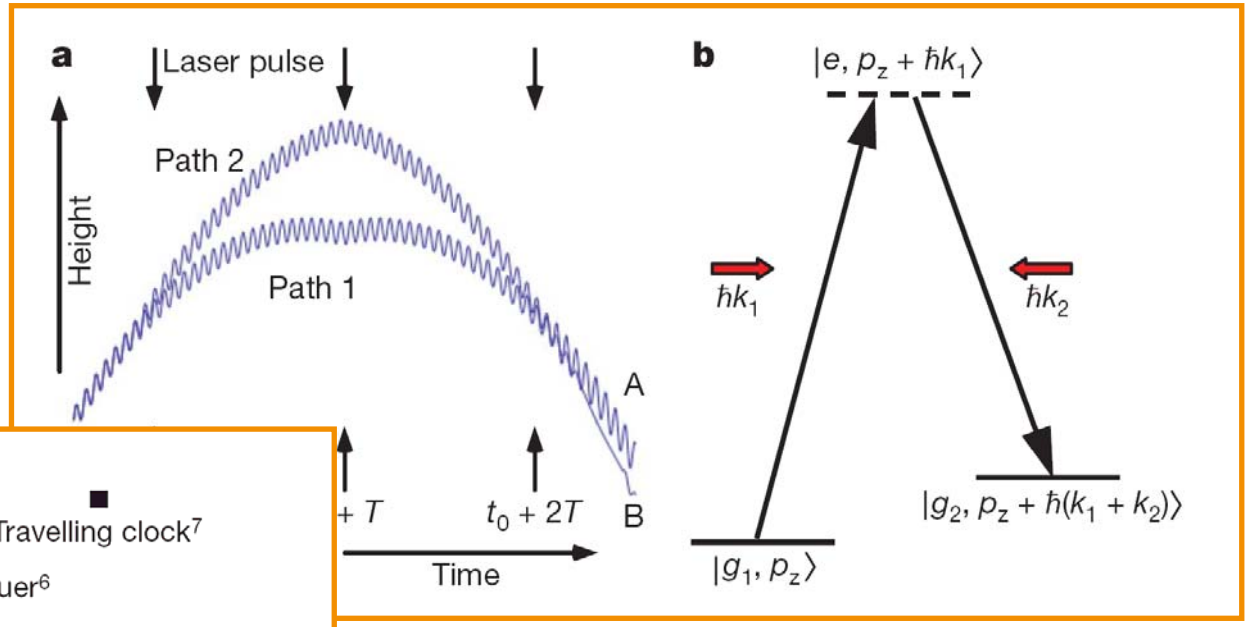
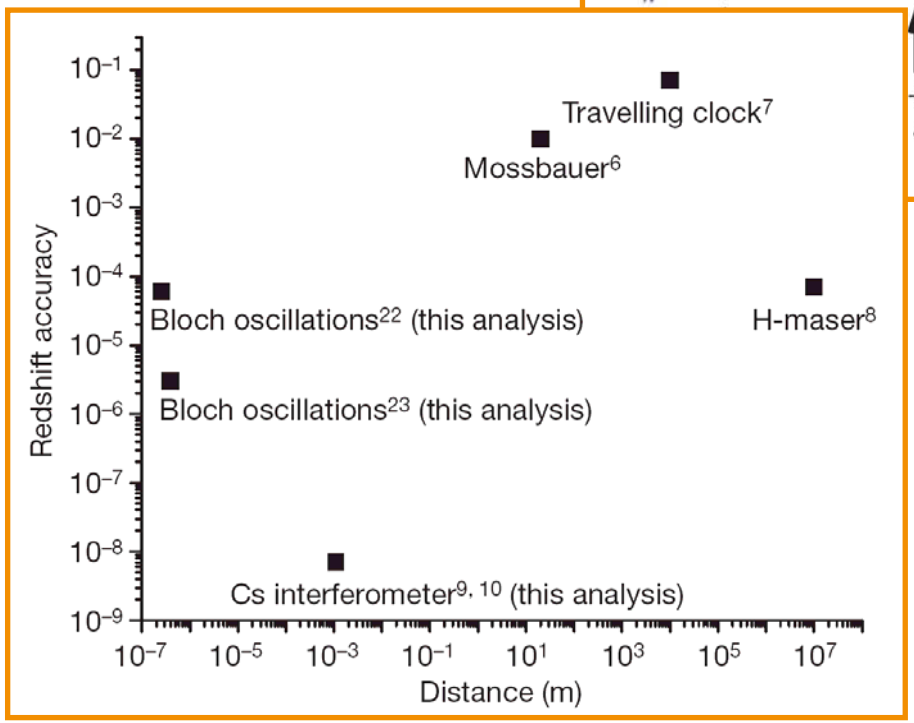
Precision metrology: Matter waves vs. Nuclear transitions

Precision test of the gravitational redshift

H. Müller, A. Peters and S. Chu, Nature 463, 926 (2010)





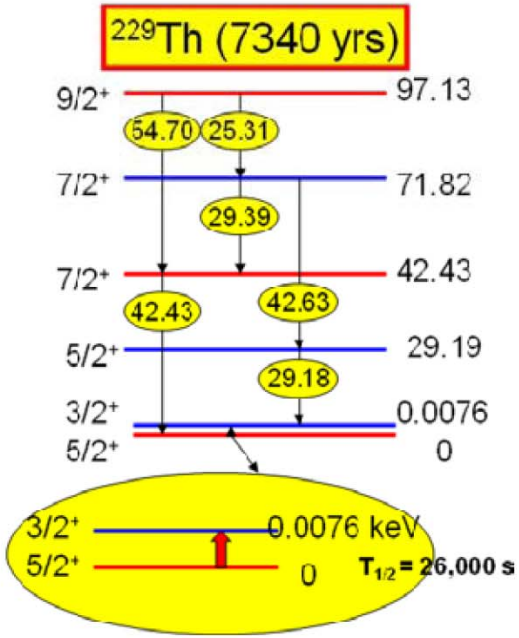
$$\Delta\phi = \int \omega_C \, d\tau$$

With $\omega_C = 10^{25}\ s^{-1}$, the Compton frequency of the atoms

Matter waves are quantum clocks operating at a frequency 10^{25} Hz

$$\frac{\Delta\phi_{meas}}{\Delta\phi_0} - 1 = 7 \cdot 10^{-9}$$

An Optical Frequency Reference Based on the ^{229}Th Nucleus



Nuclide	^{229}Th
Abundance, (%)	7340 yrs (100%)
Resonance, E_o	7.6 eV
Transition (I_g - I_e)	$5/2^+$ - $3/2^+$
Multipolarity	M1
$T_{1/2}$	26,000 s
Bandwidth, Γ	17.5zeV
$Q = \Gamma/E_o$	2.30×10^{-21}
σ_o cm^2	2.56×10^{-18}

Optical clock based on a nuclear transition with

$$\frac{\Delta f}{f} = 2 \cdot 10^{-16}$$

Precise determination of the transition energy with a VUV frequency comb

Rellergert et al.,
Constraining the Evolution of the Fundamental Constants with a Solid-State Optical Frequency Reference Based on the ^{229}Th Nucleus,
Phys. Rev. Lett. 104, 200802 (2010)

Question: Can excitation by hard x-rays help to efficiently populate the 7.6 eV level ?

Nonlinear optics with hard x-rays

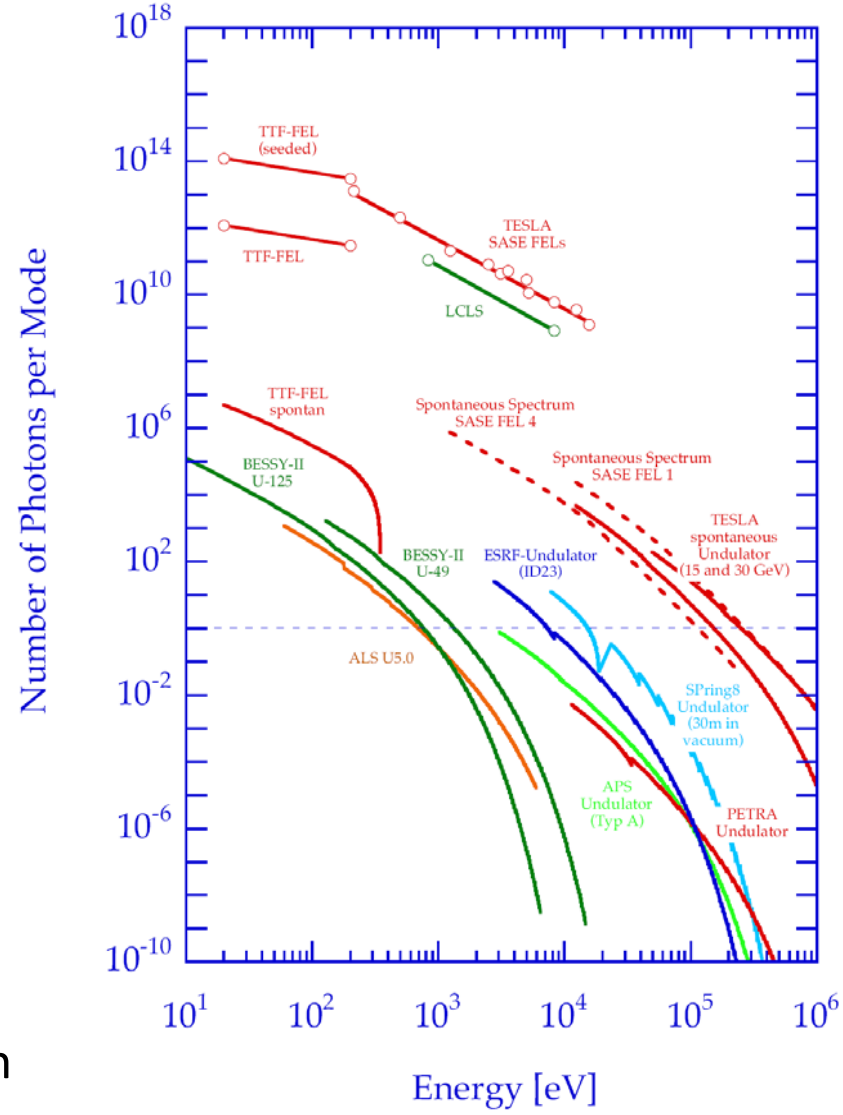
→ Photons modify the state of the sample that is experienced by subsequent photons

→ this requires a sufficiently high number of photons per mode of the radiation field (photon degeneracy)

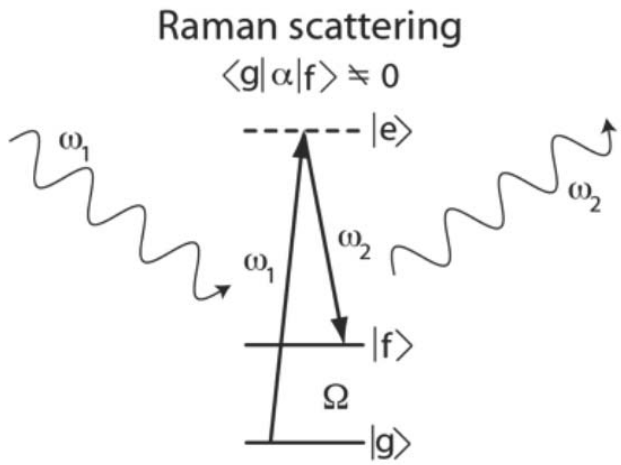
Photon degeneracy δ is the number of photons in a coherence volume, given by the transverse coherence area times the longitudinal coherence length:

$$\delta = \frac{\lambda^3}{4c} B$$

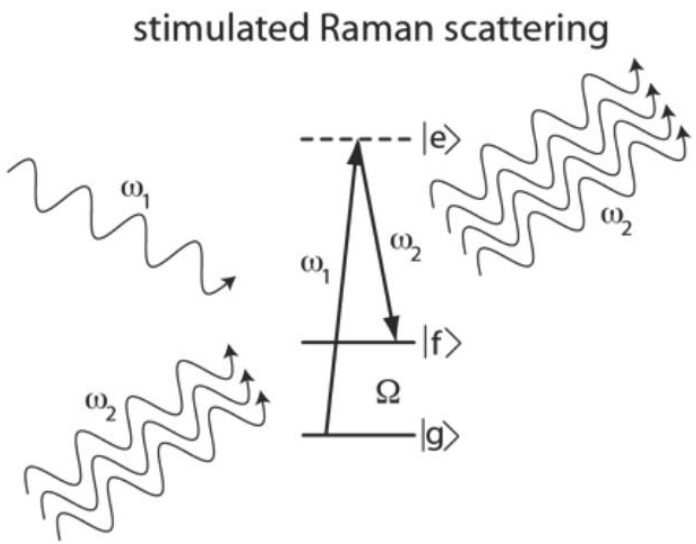
A new process that emerges at high photon degeneracies: Stimulated emission



Stimulated Raman scattering



B. D. Patterson, SLAC Technical Note SLAC-TN-10-026 (2010)



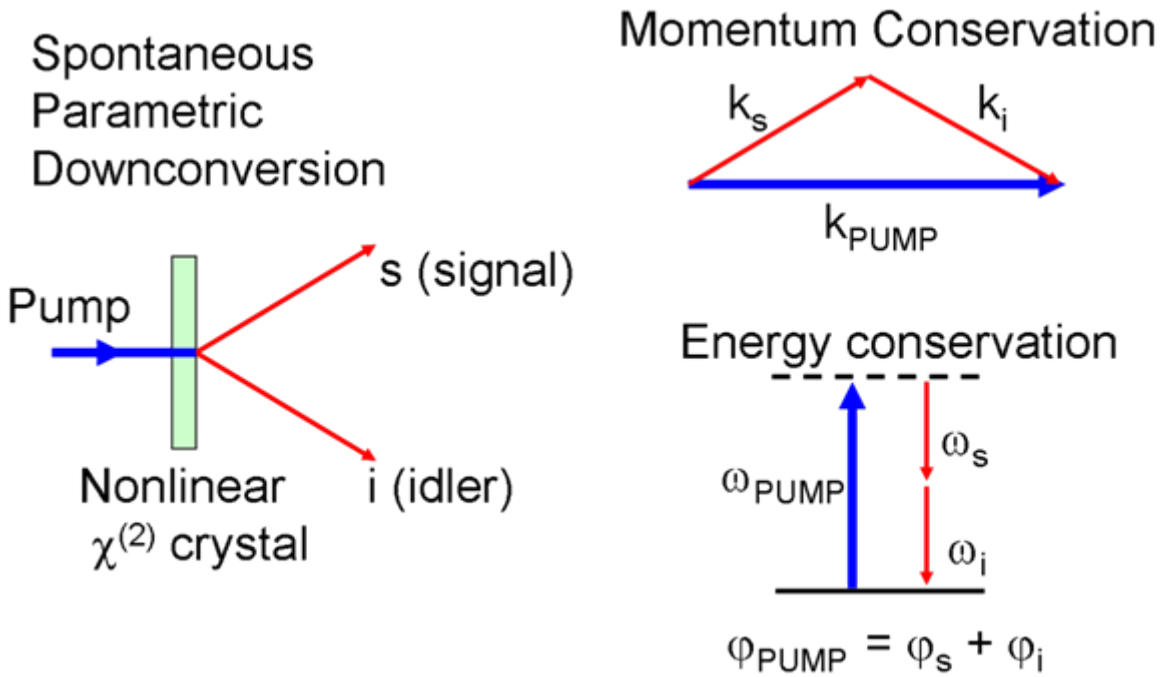
Stimulated Raman scattering outweighs the spontaneous Raman scattering when the incident photon flux contains more than one photon per mode.

→ Boosting the sensitivity of the Raman signal !!

Question: Can this scheme be converted into a high-resolution, scanning spectroscopy at x-ray energies ?

Requirements for nonlinear spectroscopies with x-rays:
Multicolor XFEL sources

Spontaneous parametric down conversion



‚parametric' means: The state of the crystal is left unchanged in the process

A nonlinear crystal splits incoming photons into pairs of undistinguishable photons of lower energy

$$|\psi_{AB}\rangle = \frac{1}{\sqrt{2}}\left(|v\rangle_A|h\rangle_B + |h\rangle_A|v\rangle_B\right)$$

These two photons are entangled !

Measurement of the state of photon A determines the state of photon B

→ Test of quantum mechanics, Bell's theorem, applications in quantum cryptography

Spontaneous parametric down conversion of x-rays

Experiments reported (selection):

P. Eisenberger and S. McCall, Phys. Rev. Lett. **26**, 684 (1971),

Y. Yoda et al., J. Synchrotron Rad. **5**, 980 (1998),

B. W. Adams et al., J. Synchrotron Rad. **7**, 81 (2000).

Efficiency of PDC at x-ray energies is low

Max. 0.01 s^{-1} at third generation sources, up to 100 s^{-1} at XFELs

Experiments with nonclassical states of x-rays

Requirements:

- A deterministic source of single photons
- A source of high-intensity entangled pairs of photons.

Generation of Nonclassical states of Light in the X-ray regime

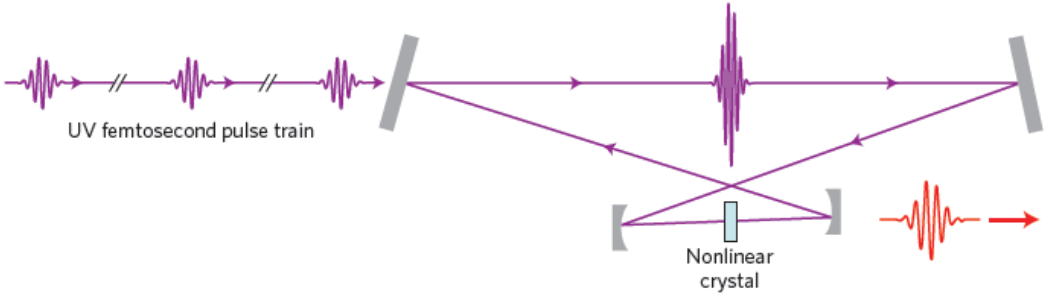
QUANTUM OPTICS

Enhanced quantum light generation

The use of a specially designed cavity to enhance the intensity of femtosecond ultraviolet pulses dramatically increases the rate at which non-classical states of light can be produced.

R. Jason Jones

A number of quantum networking protocols, such as quantum teleportation and secret sharing, require the use of multiphoton entangled states of light[1]. Such exotic quantum states are often produced from spontaneous parametric down-conversion (SPDC) in a nonlinear crystal. In this scheme, the crystal splits a single photon into multiple pairs of entangled photons of



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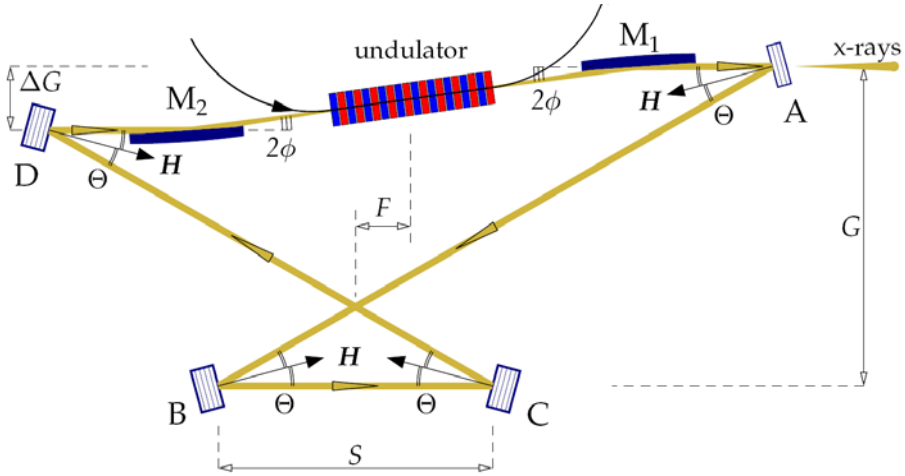
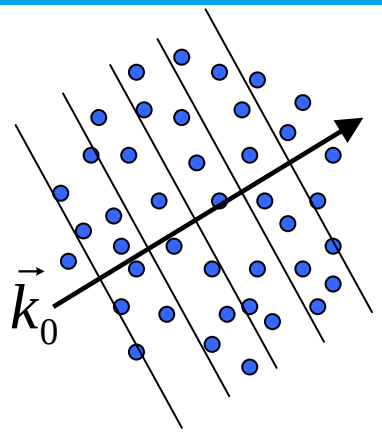
Compare with XFEL oscillator (XFELO)

Kwang-Je Kim, Yuri Shvydko et al.



Entangled states between photons and atoms

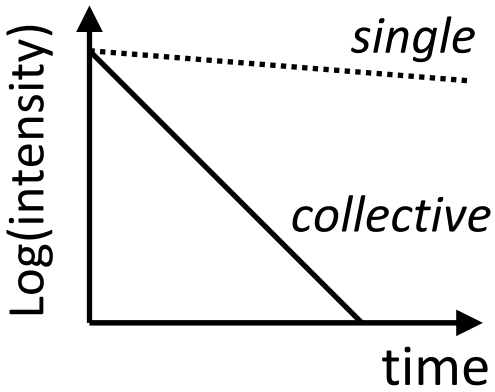
Cloud of N identical atoms, radius R, absorbing a single photon with resonant wavelength λ



$$\left|+\right\rangle_{k_0} = \frac{1}{\sqrt{N}} \sum_j e^{i\vec{k}_0 \cdot \vec{r}_j} \left|\downarrow_1 \downarrow_2 \ldots \uparrow_j \ldots \downarrow_N\right\rangle$$

One atom in the sample is excited, but we do not know which one

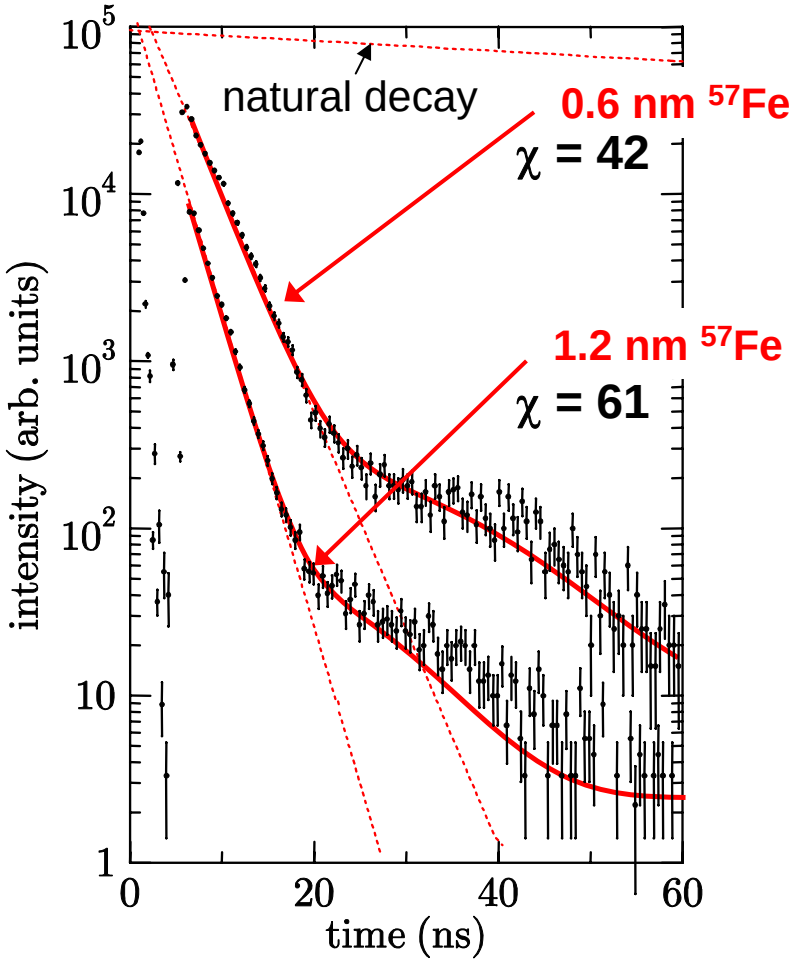
Maximum delocalization of photon energy → Superradiance !!



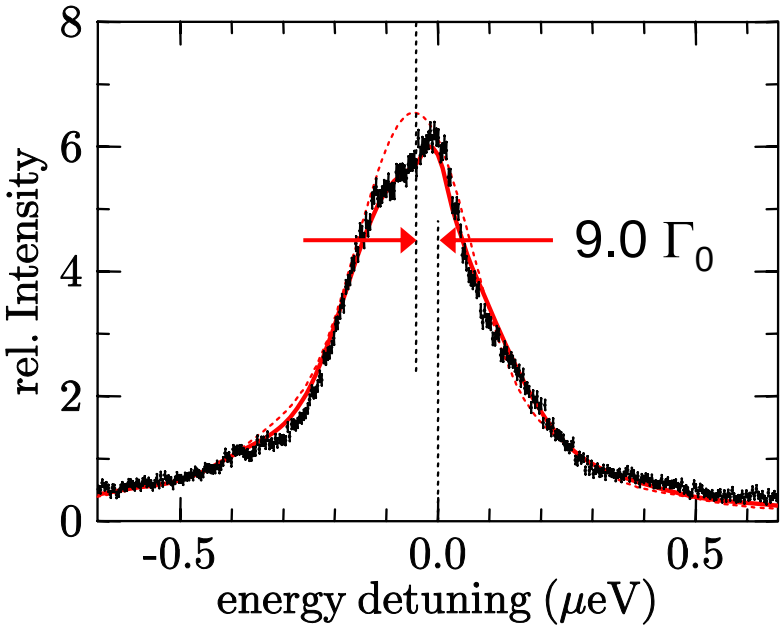
A collection of identical atoms radiates much faster than a single atom

Nuclear γ-ray Superradiance and the Collective Lamb Shift

Temporal and spectral response of ^{57}Fe nuclei embedded in a planar cavity, coherently excited with 14.4 keV x-rays



Redshift of spectral response



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

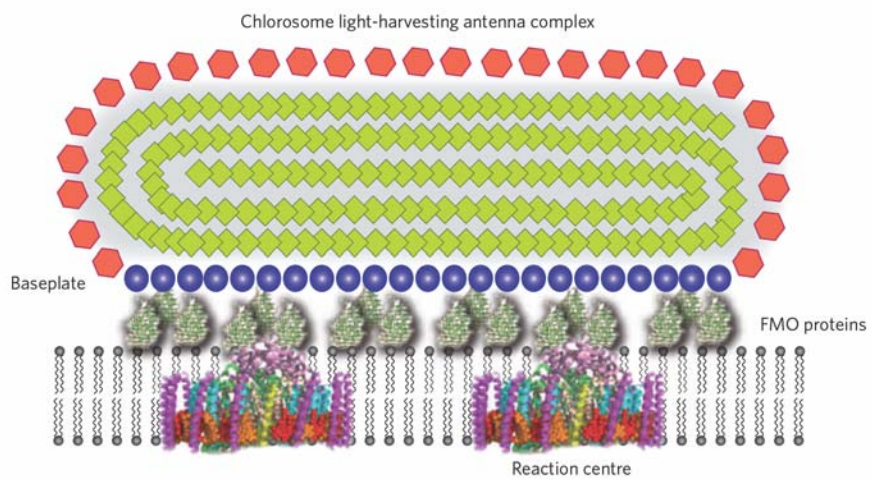
Superradiance as a probe of photonic (de)localization

Excitation of light-harvesting complexes

Entanglement is related to the extent of delocalization of the electronic excitation

Entanglement of excitations in a light-harvesting system might accelerate the transfer of the Sun´s energy to reaction centres

Observation of superradiant emission suggests that entangled states persist for the lifetime of the excited state



Photosynthetic machinery of green sulfur bacteria

M. Sarovar, A. Ishizaki,G. R. Fleming, B. Whaley, *Nature Physics* **6,** 462–467 (2010).

G. D. Scholes, *Nature Physics* 6, 402 (2010)

Learning from such systems can help in designing more efficient artificial light harvesting systems, i.e., solar cells.