APS Colloquium Series, March 1, 2006

Boosting the Light: X-Ray Physics in Confinement

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The Simpletons (Citizens of Schilda) Build a City Hall

- but they forget the windows

Proposal: Trap the light and carry it in

Themes

How to put light into boxes

→ Photonic resonators: Visible light → X-rays

When light meets matter … it scatters

→ Spontaneous emission and the Purcell effect

→ Coherent scattering: Accelerating the temporal evolution and boosting the intensity

Applications

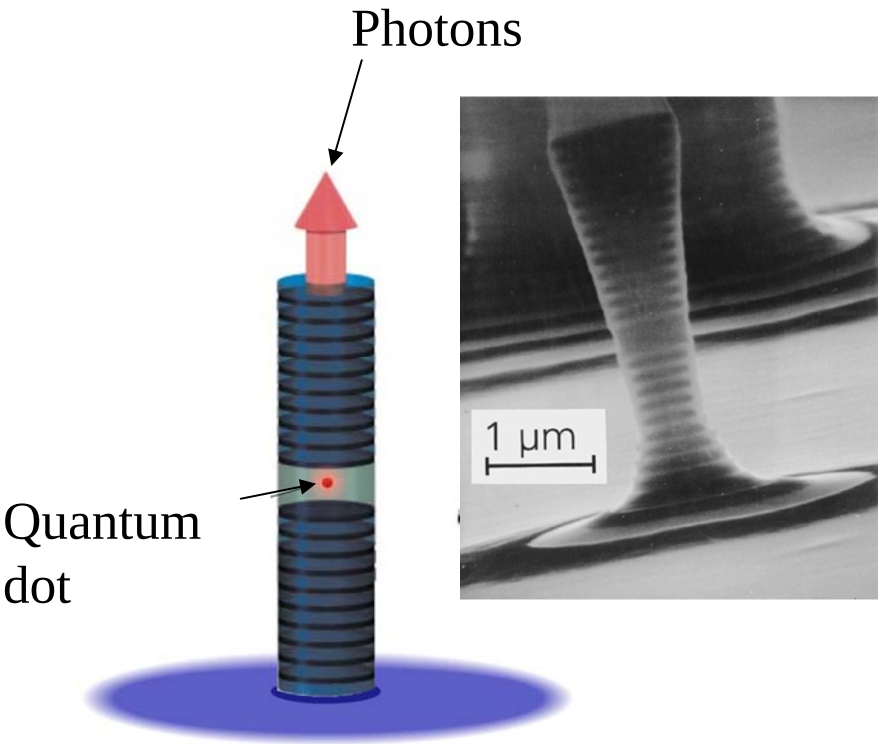
→ Probing the magnetic structure of exchange-coupled films

→ Amplification of coherent x-ray scattering

Putting Light into Boxes

Fabry-Perot microcavities

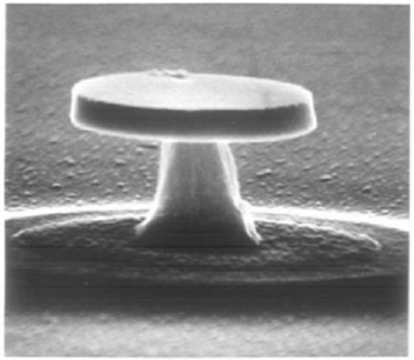
Two parallel low-loss mirrors
separated by a gap

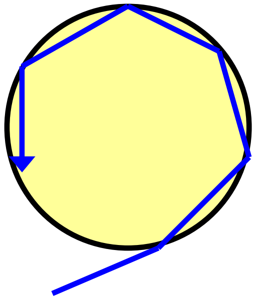


Q > 20000

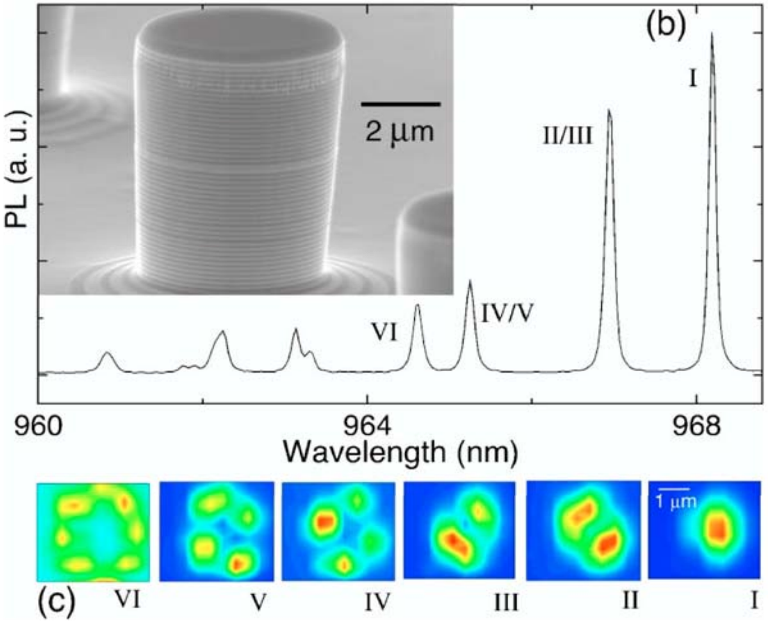
J. M. Gerard et al.,
Appl. Phys. Lett. 69, 449 (2001)

Whispering-gallery resonators





Q > 10000

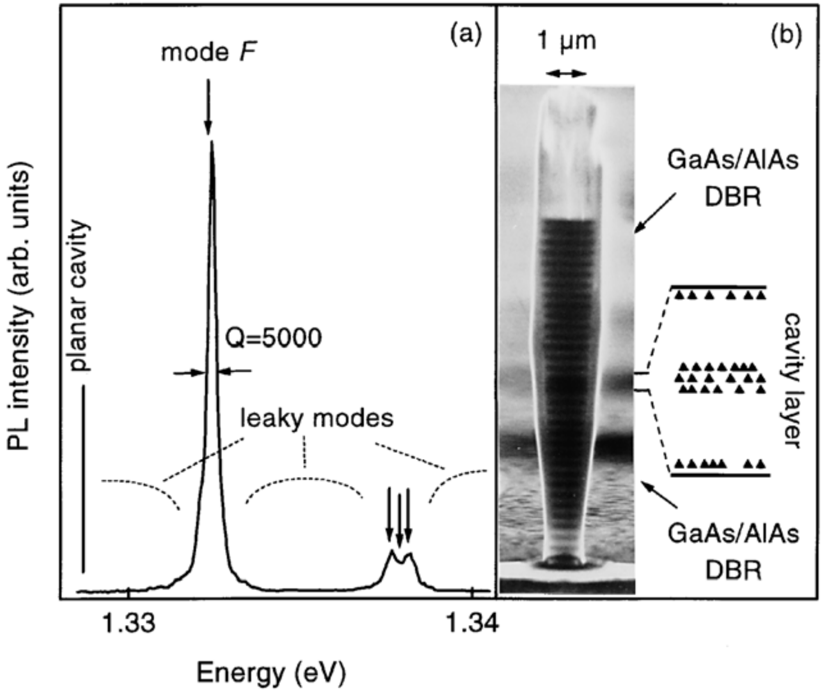


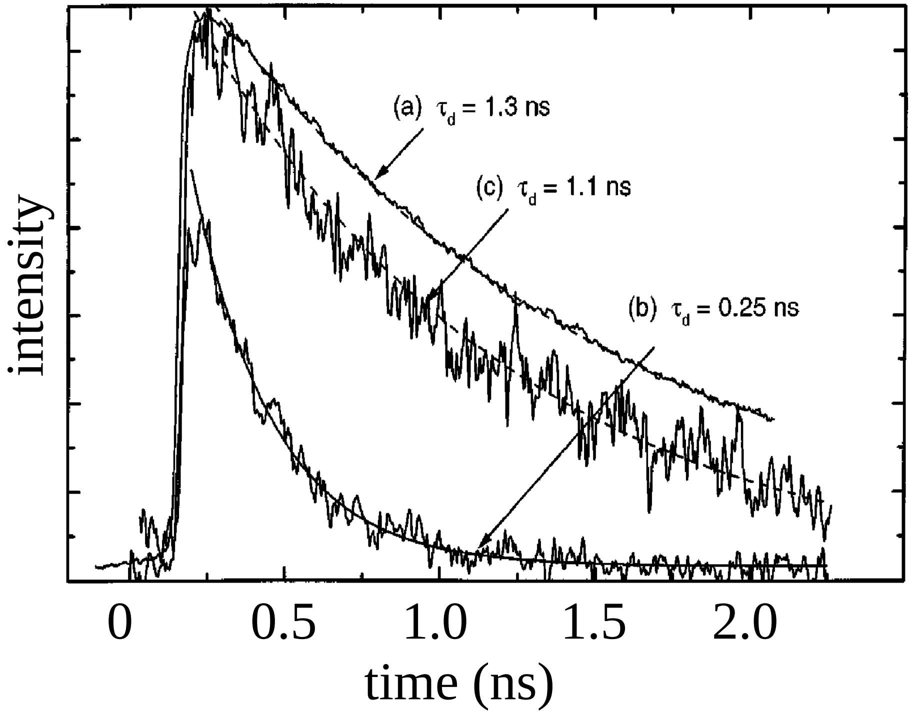
Resonance in a Resonator

Interaction of two-level emitters interacting resonantly with a single cavity mode

Time-resolved photoluminescence from quantum dots in a microcavity

J. M. Gerard et al., Phys. Rev. Lett. 81, 1110 (1998)

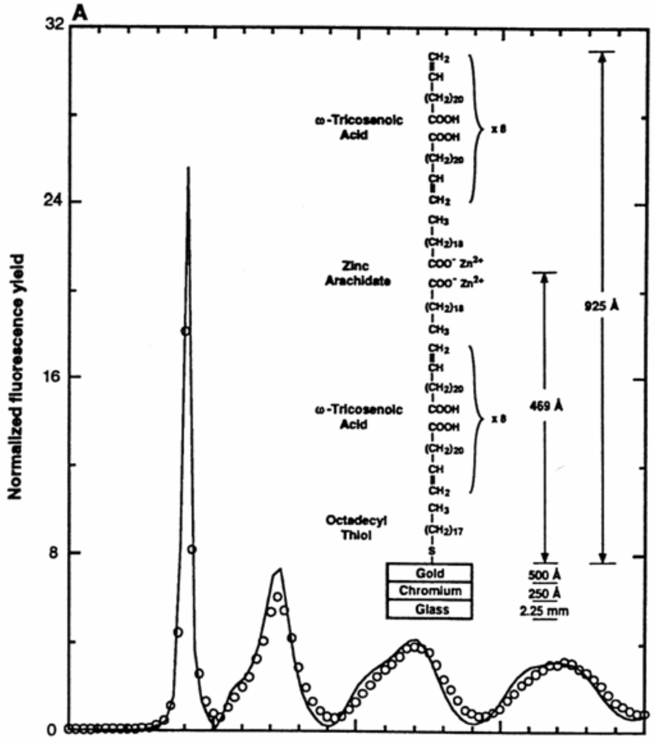




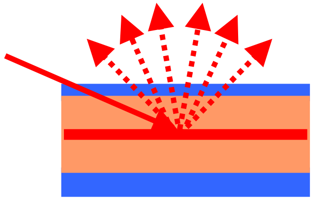
Applications:
- High-speed optical data processing
- Single-photon sources
- Low-threshold lasing

When x-rays meets matter in confinement: Increasing the fluorescence yield

Resonance enhanced x-rays in thin films: A structure probe for membranes, surface layers and materials in confinement

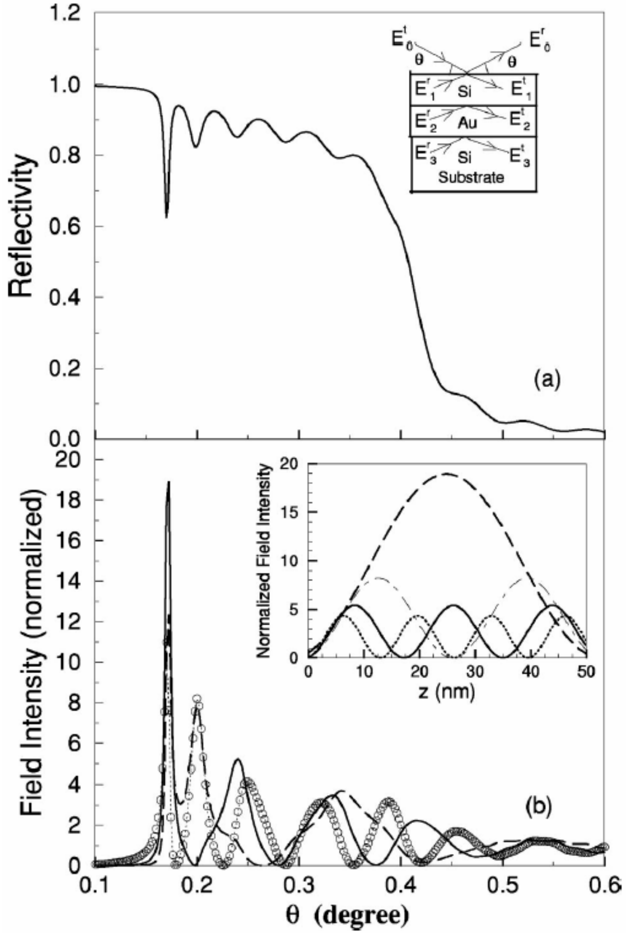


J. Wang, M. Bedzyk, and M. Caffrey, Science 258, 775 (1992)

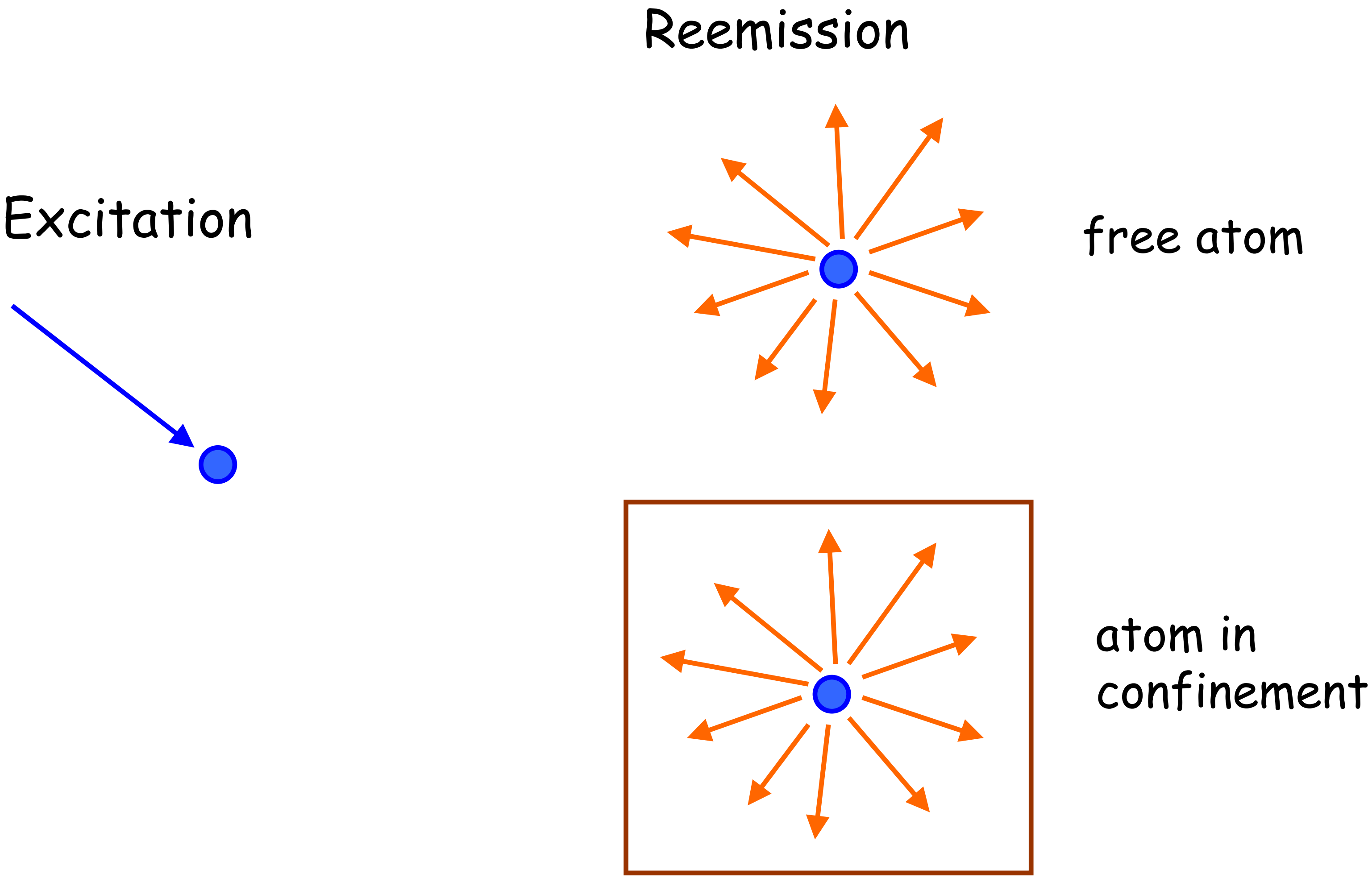


Resonance enhancement of x-rays and fluorescence yield from marker layers in thin films

S. K. Ghose, B.N. Dev, and A. Gupta, Phys. Rev. B 64, 233403 (2001)



How to treat photon emission in a resonator ?



Reemission

Excitation

free atom

atom in confinement

Spontaneous emission

leads to most of the radiation that surrounds us

The rate of spontaneous emission is given by Fermi's Golden rule:

$$\boxed{\Gamma = \frac{1}{\tau} = \frac{2\pi}{\hbar}\,\rho(\vec{r},\omega_0)\,\left|\langle g,1|\,\mathbf{H}\,|e,0\rangle\right|^2}$$

Transition matrix element

Density of (final) photon states

→ Spontaneous emission (SE) is not an inherent property of atoms, but depends on geometry

→ SE rate will be increased in the vicinity of boundaries and interfaces where the density of modes of the electromagnetic field is enhanced

→ This effect is most pronounced if emitters are placed in resonators

The Photonic Density of States in Cavities

a) in free space →

$$\rho_0 = \frac{1}{V}\frac{dN}{dE} = \frac{2}{\pi}\left(\frac{\omega^2}{c^3}\right)$$

b) in a cavity →

$$\rho_c = \frac{1}{V\,\Delta\nu}$$

Volume V,
Quality factor Q,
Spectral width $\Delta\nu$

$$\Delta\nu = \frac{\nu}{Q}, \quad V = \left(\frac{\lambda}{2}\right)^3$$

↓

$$\rho_c = \frac{2}{\pi^2}\frac{\omega^2}{c^3}Q$$

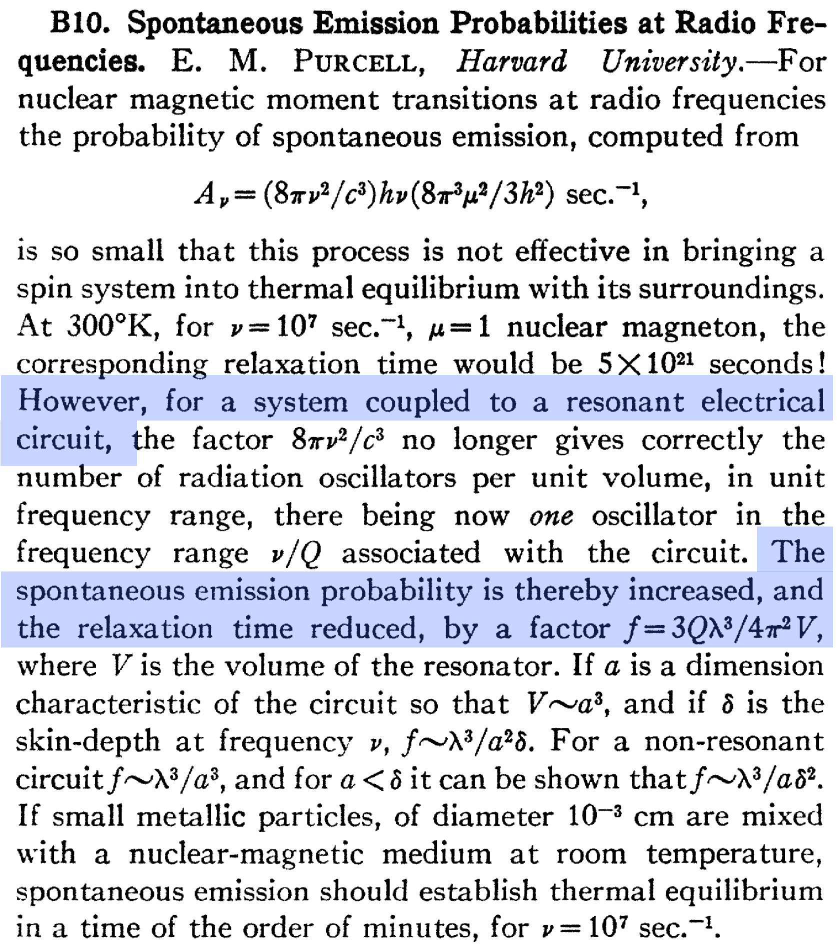
Enhancement of the radiative decay rate

$$\boxed{\frac{\Gamma}{\Gamma_0} = \frac{\rho_c}{\rho_0} = \frac{Q}{\pi}}$$

The Purcell Effect

The Purcell effect

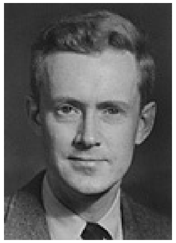
Phys. Rev. 69, 681 (1946)



B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. Purcell, *Harvard University*.—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

$$A_\nu = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2)\ \text{sec.}^{-1},$$

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu = 10^7$ sec.$^{-1}$, $\mu = 1$ nuclear magneton, the corresponding relaxation time would be 5×10^{21} seconds! However, for a system coupled to a resonant electrical circuit, the factor $8\pi\nu^2/c^3$ no longer gives correctly the number of radiation oscillators per unit volume, in unit frequency range, there being now *one* oscillator in the frequency range ν/Q associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor $f = 3Q\lambda^3/4\pi^2 V$, where V is the volume of the resonator. If a is a dimension characteristic of the circuit so that $V \sim a^3$, and if δ is the skin-depth at frequency ν, $f \sim \lambda^3/a^2\delta$. For a non-resonant circuit $f \sim \lambda^3/a^3$, and for $a < \delta$ it can be shown that $f \sim \lambda^3/a\delta^2$. If small metallic particles, of diameter 10^{-3} cm are mixed with a nuclear-magnetic medium at room temperature, spontaneous emission should establish thermal equilibrium in a time of the order of minutes, for $\nu = 10^7$ sec.$^{-1}$.



NOBEL

Prize in Physics 1952

Acceleration of spontaneous emission from atoms in a cavity

Purcell enhancement factor for spontaneous emission :

$$\boxed{F_P = \frac{\tau_0}{\tau} = \frac{3}{4\pi^2}\left[\frac{(\lambda/n)^3}{V}\right] Q}$$

Q = quality factor

λ = wavelength

n = index of refraction

V = cavity volume

What happens to light and its interaction with matter when it is trapped in a box ?

→ Cavity Quantum Electrodynamics (CQED)

Manipulating the light-matter interaction in confining geometries

Experimental challenges:

Achieve a high Purcell factor via

- Development of high-Q cavities
- Cavity dimensions in the order of the wavelength
- 3D photon confinement → Highly reflective boundaries

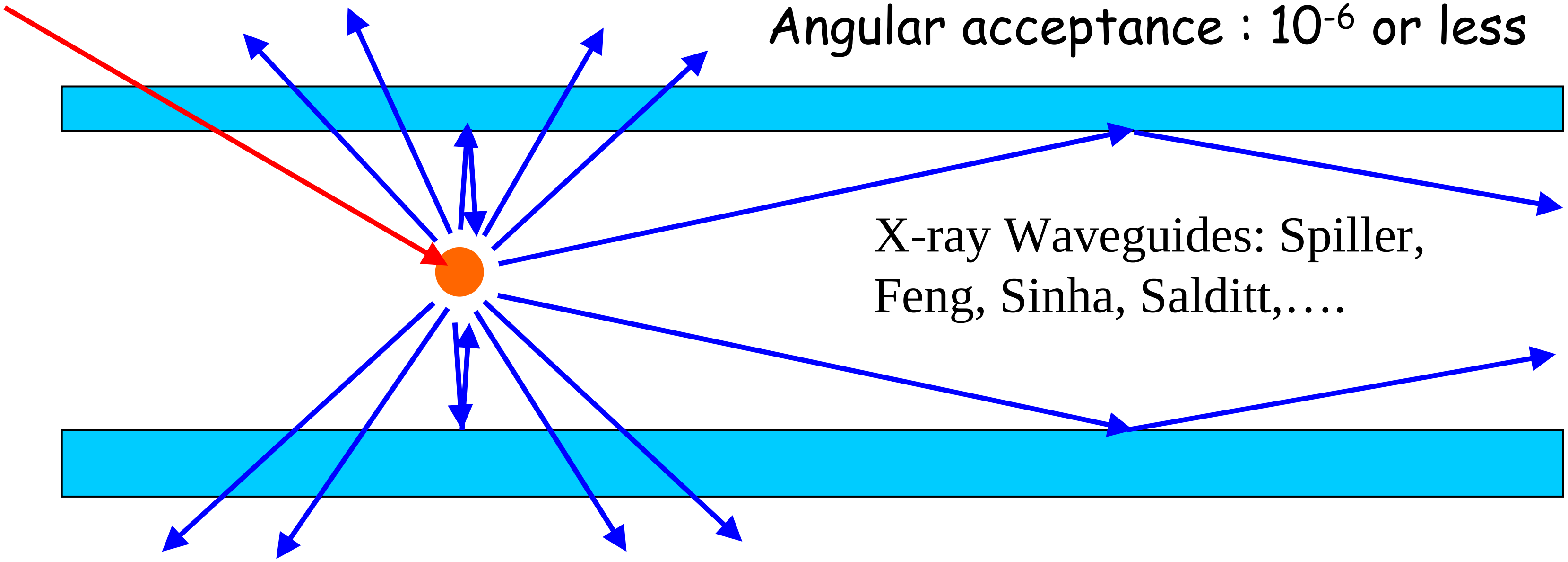
Accurate placement of emitters in the cavity

How to trap x-rays in a box ?

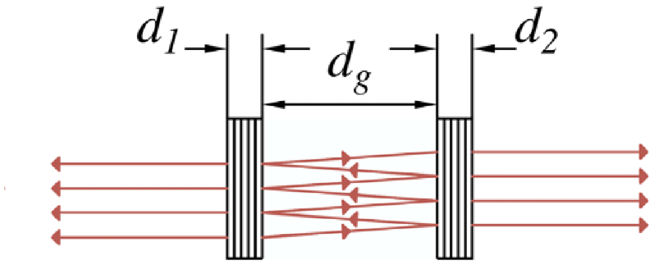
Seems to be difficult because of

- → Narrow angular acceptance
- → Low reflectivities of interfaces

Angular acceptance : 10^{-6} or less



X-ray Waveguides: Spiller, Feng, Sinha, Salditt,….



X-ray Fabry-Perot Resonators:

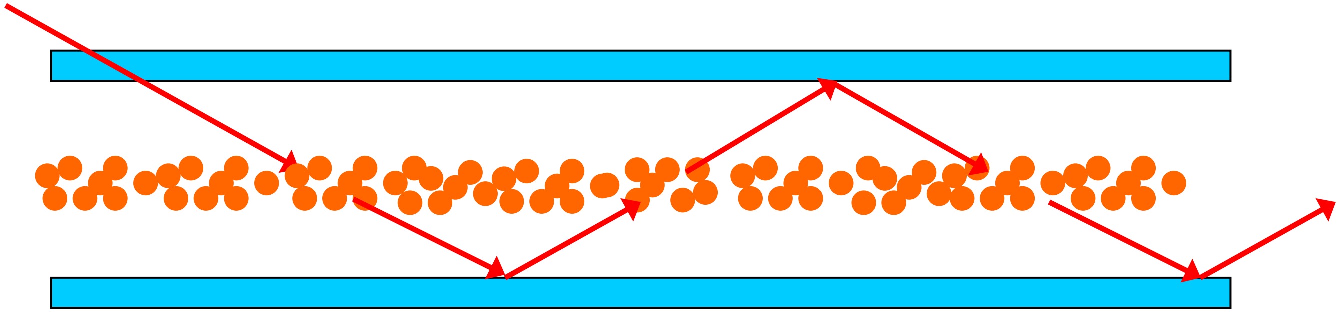
Yu. V. Shvyd‘ko et al., PRL 90, 013904 (2003)

S. L. Chang et al., PRL 94, 174801 (2005)

Towards Cavity Quantum Electrodynamics with X-rays

Efficient trapping of spontaneously emitted x-rays:

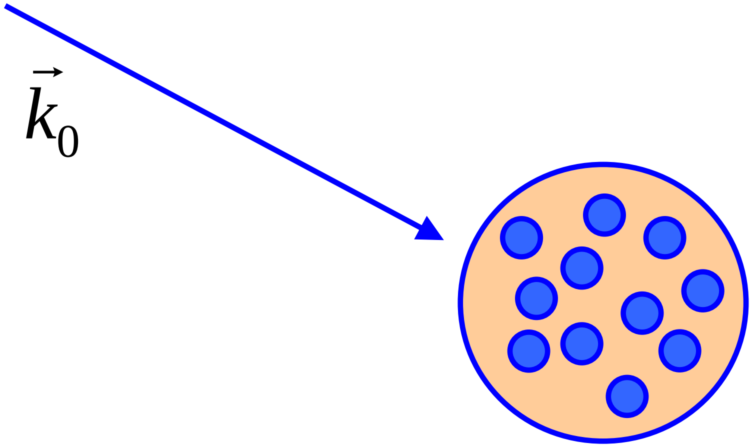
(1) Employ highly directional coherent scattering from many atoms instead of incoherent emission from single emitters



(2) Employ x-ray scattering in grazing-incidence geometry where the reflectivities of interfaces are high

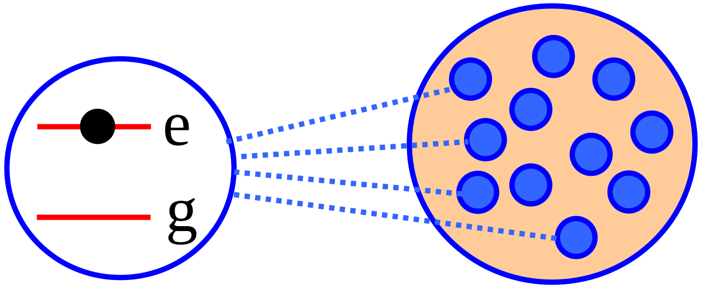
(3) Employ atomic transitions with an experimentally accessible lifetime: The 14.4 keV transition of ^{57}Fe with τ = 141 ns, excited with a short-pulsed radiation source

Coherent Resonant Forward Scattering from an ensemble of atoms



Absorption of one photon with wavevector $\vec{k}_0$

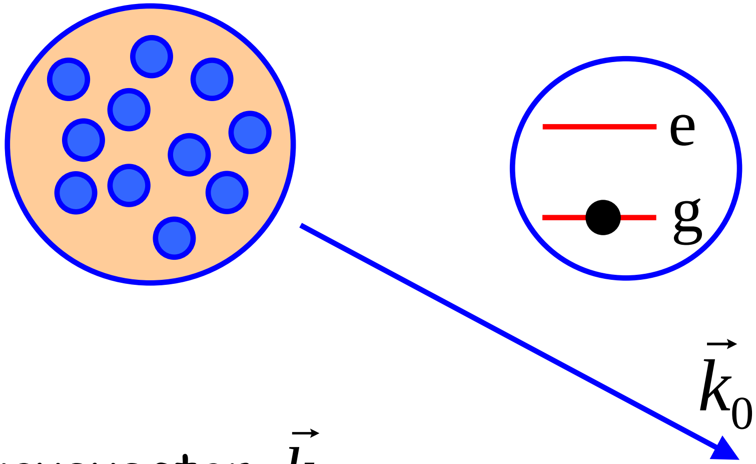
Coherent Resonant Forward Scattering from an ensemble of atoms



Absorption of one photon with wavevector $\vec{k}_0$

→ One atom can be excited, but we do not know which one

Coherent Resonant Forward Scattering from an ensemble of atoms



Absorption of one photon with wavevector $\vec{k}_0$

→ One atom can be excited, but we do not know which one

→ The ensemble of atoms radiates as a coherent superposition of classical dipole oscillators

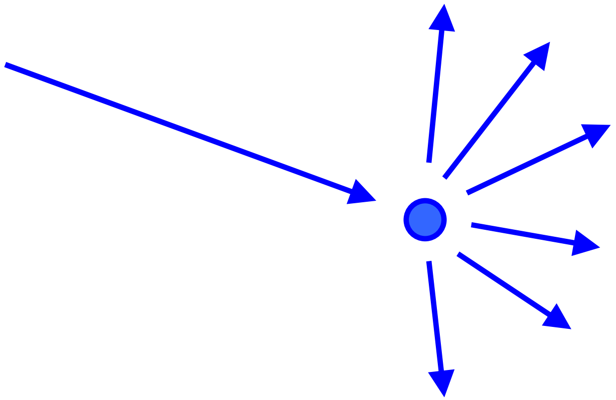
→ Reemission in direction of $\vec{k}_0$

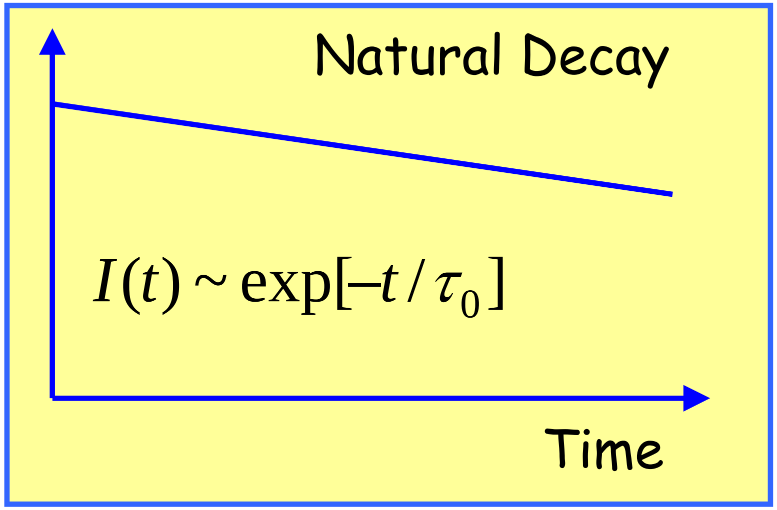
Nuclear forward scattering of synchrotron radiation:

J. B. Hastings et al.,
Phys. Rev. Lett. 66, 770 (1991)

Temporal Evolution of Scattering Processes (1)

Incoherent Scattering

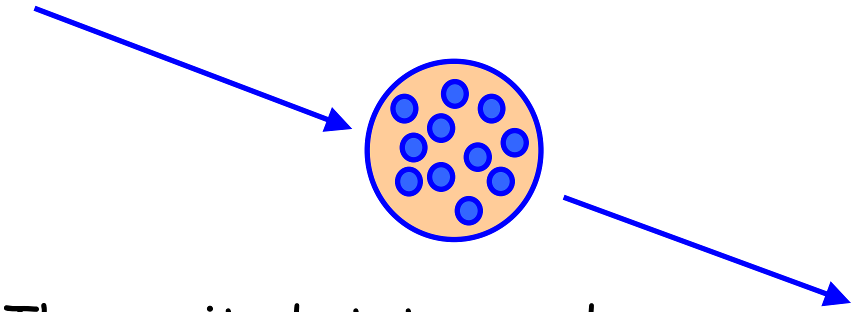




$$I(t) \sim \exp[-t/\tau_0]$$

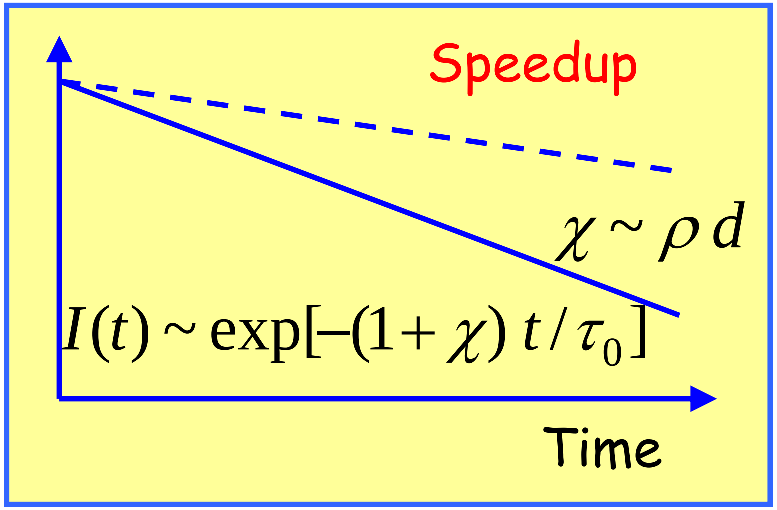
Temporal Evolution of Scattering Processes (2)

Coherent scattering



The excited state can decay through many atoms

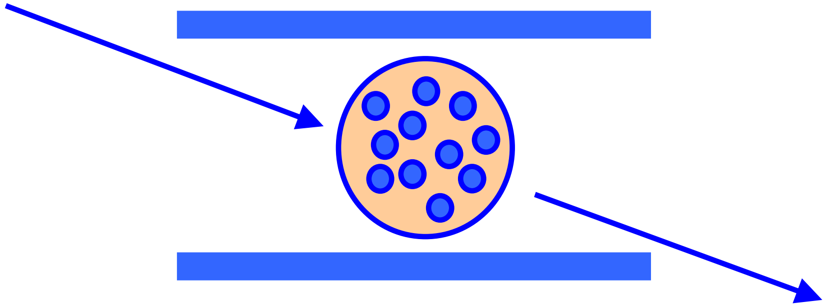
(U. van Bürck et al, PRB 46, 6207 (1992))



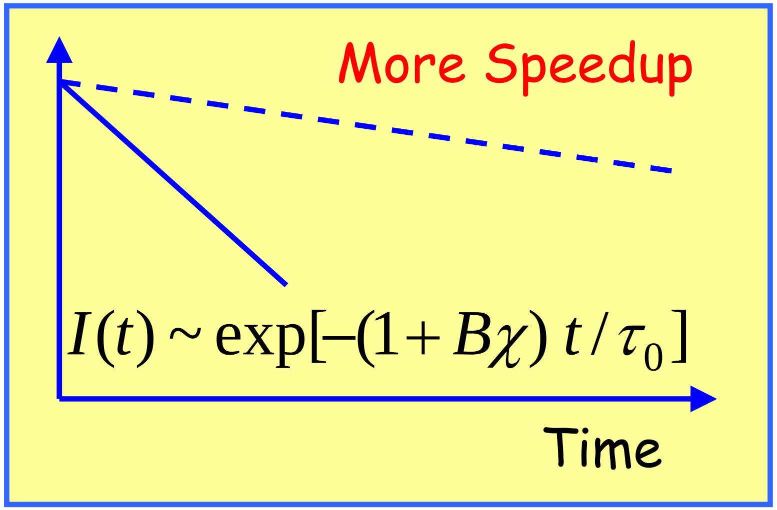
$$\chi \sim \rho\, d$$

$$I(t) \sim \exp[-(1+\chi)\, t/\tau_0]$$

Coherent scattering in confinement



The excited state can decay through many photonic states

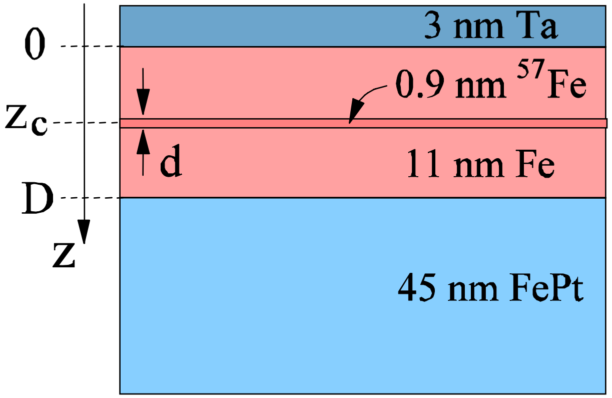


$$I(t) \sim \exp[-(1+B\chi)\, t/\tau_0]$$

The additional enhancement factor is determined by the photonic density of states in the cavity:

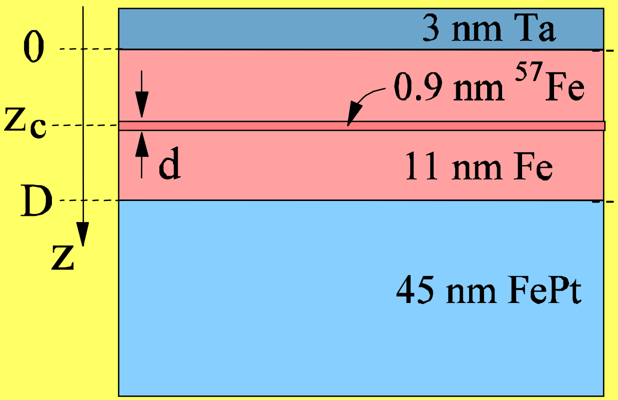
$$\boxed{B = \frac{\rho_c}{\rho_0}}$$

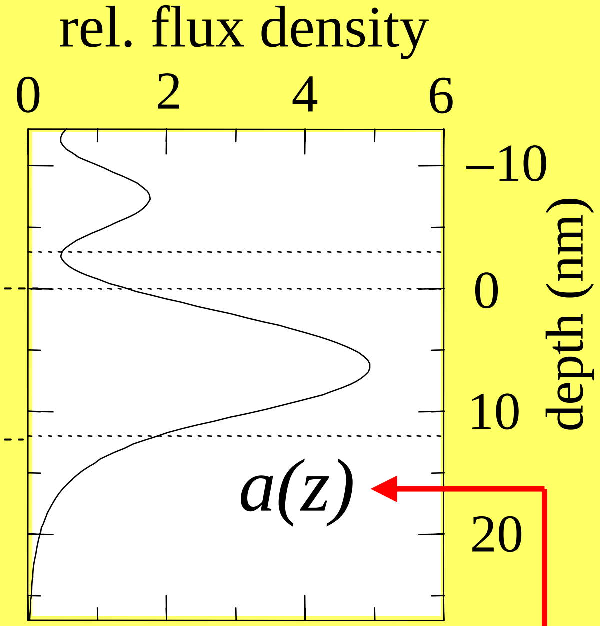
Experiment: An ultrathin layer of ^{57}Fe in an x-ray waveguide



Sample geometry

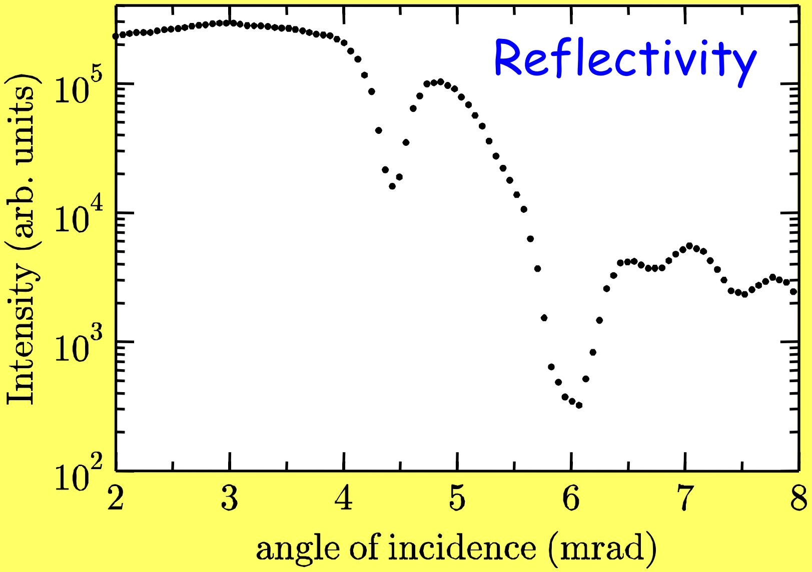
^{57}Fe in a single-mode waveguide



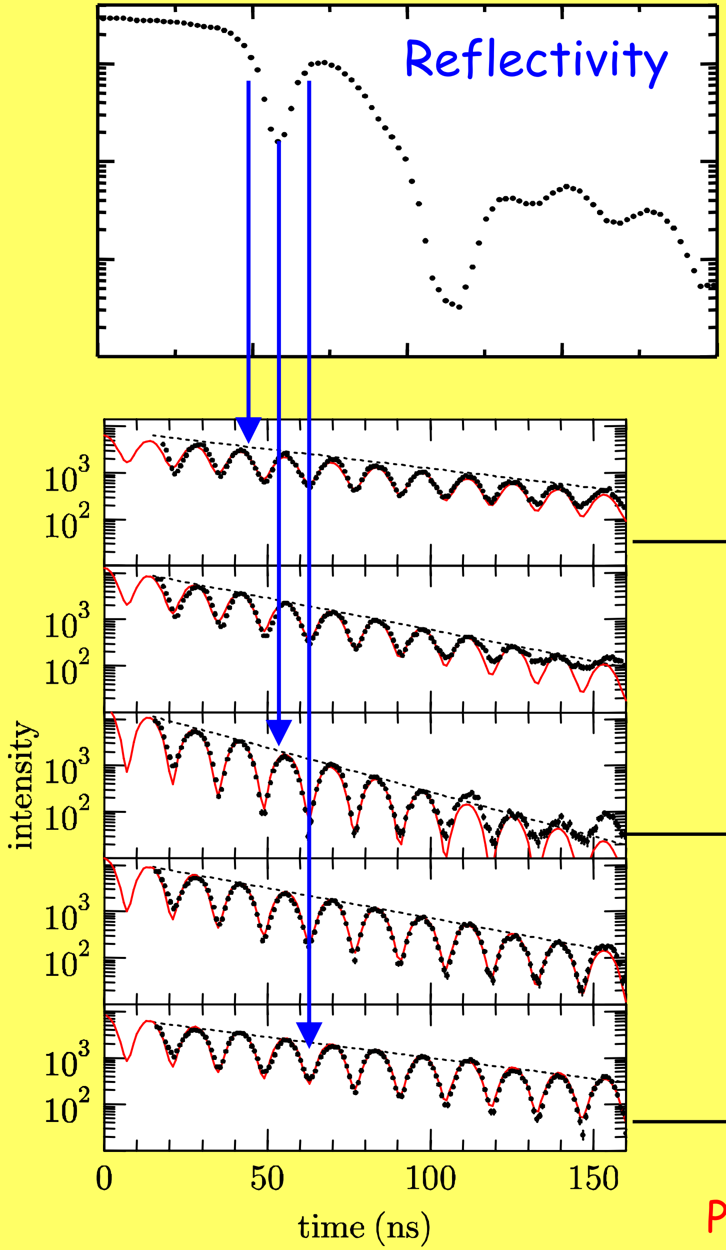


$$\rho(z) = [1 + a(z)]\rho_0$$

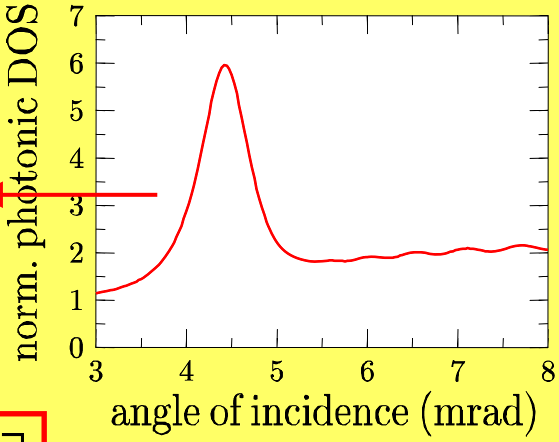
Photonic density of states



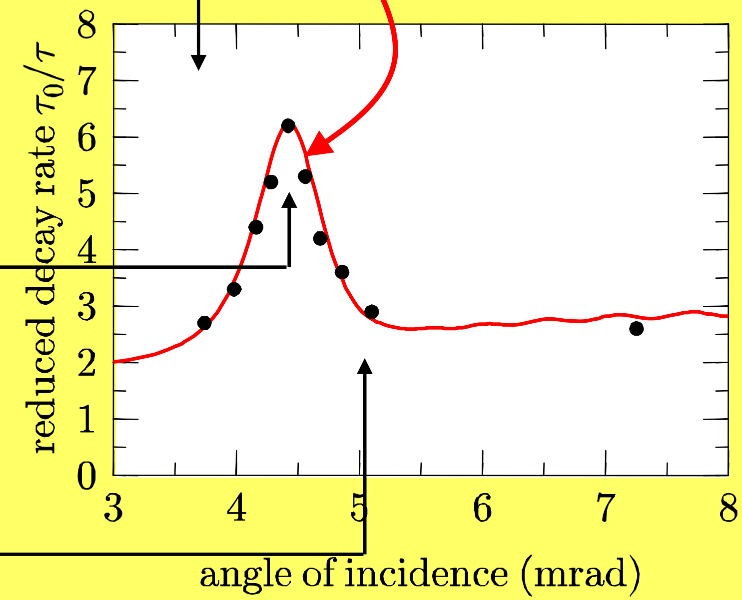
The time-delayed signal from the ^{57}Fe nuclei



$$\frac{\rho(z)}{\rho_0}=[1+a(z)]$$



$$\boxed{\frac{\tau_0}{\tau}=1+\chi\left[\frac{\rho(z_c)}{\rho_0}\right]}$$

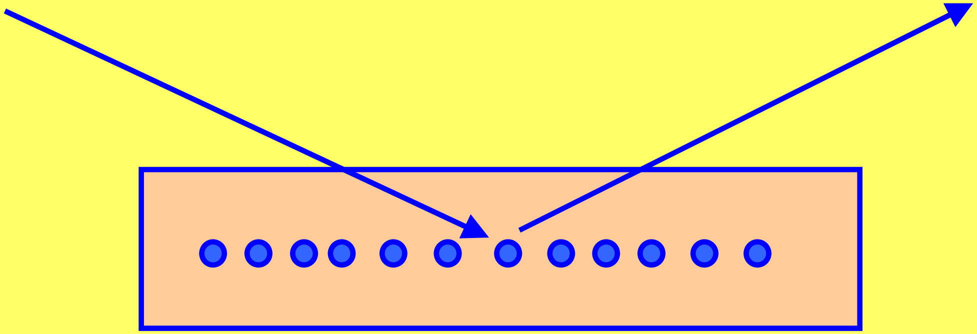


PRL 95, 097601 (2005)

Coherent Scattering and the Photonic Density of States

Temporal Dependence

$$\boxed{I(t) \sim \exp[-(1+B\chi)\, t/\tau_0]}$$



with $B = \dfrac{\rho_c}{\rho_0}$

Total Intensity

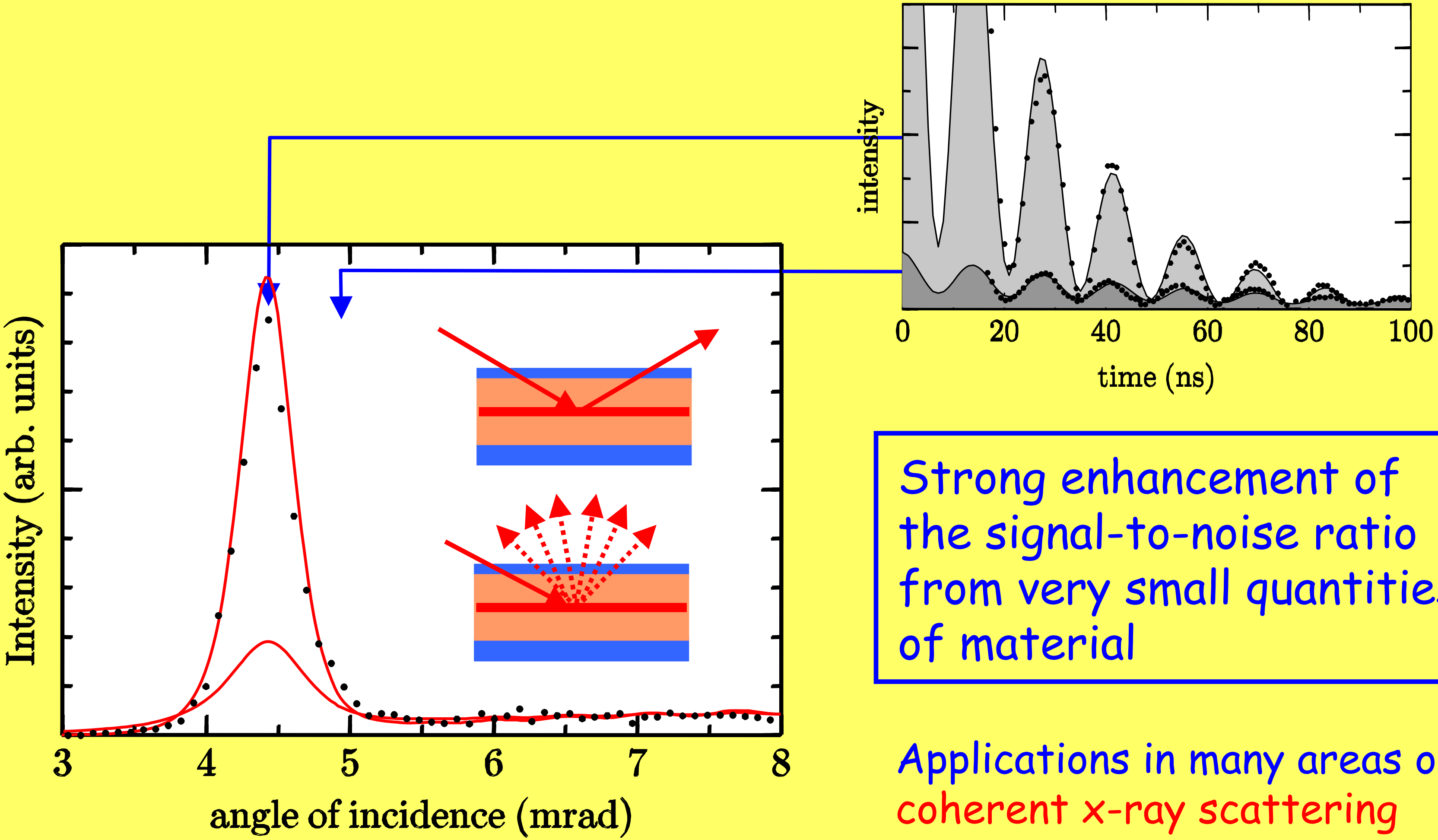
Coherent Scattering Amplitude

$$f_{coh} \sim \frac{\rho_c}{\rho_0} = B$$

$$\boxed{I_{tot} \sim |f_{coh}|^2 \sim B^2}$$

The intensity of coherent x-ray scattering scales quadratically with the photonic density of states at the position of the atoms

Boosting the Intensity of Coherent X-ray Scattering



PRB 69, 235412 (2004)

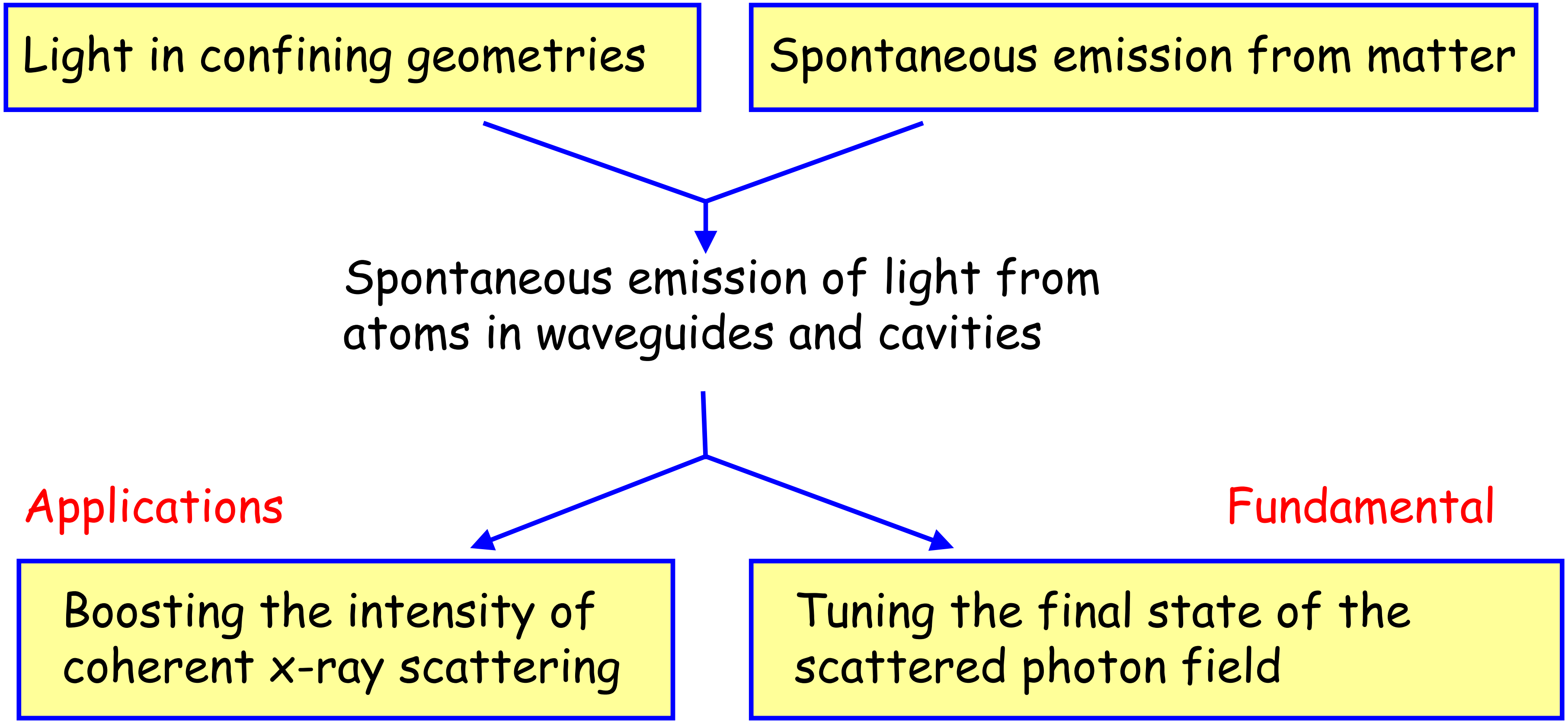
Strong enhancement of the signal-to-noise ratio from very small quantities of material

Applications in many areas of coherent x-ray scattering from layered systems:

GISAXS, GID, XRMS, XPCS

What happens if one puts light into a box:

Light-matter interaction in confining geometries



Light in confining geometries

Spontaneous emission from matter

Spontaneous emission of light from atoms in waveguides and cavities

Applications

Fundamental

Boosting the intensity of coherent x-ray scattering

Tuning the final state of the scattered photon field

The Spin Structure of Hard/Soft - Magnetic Bilayers

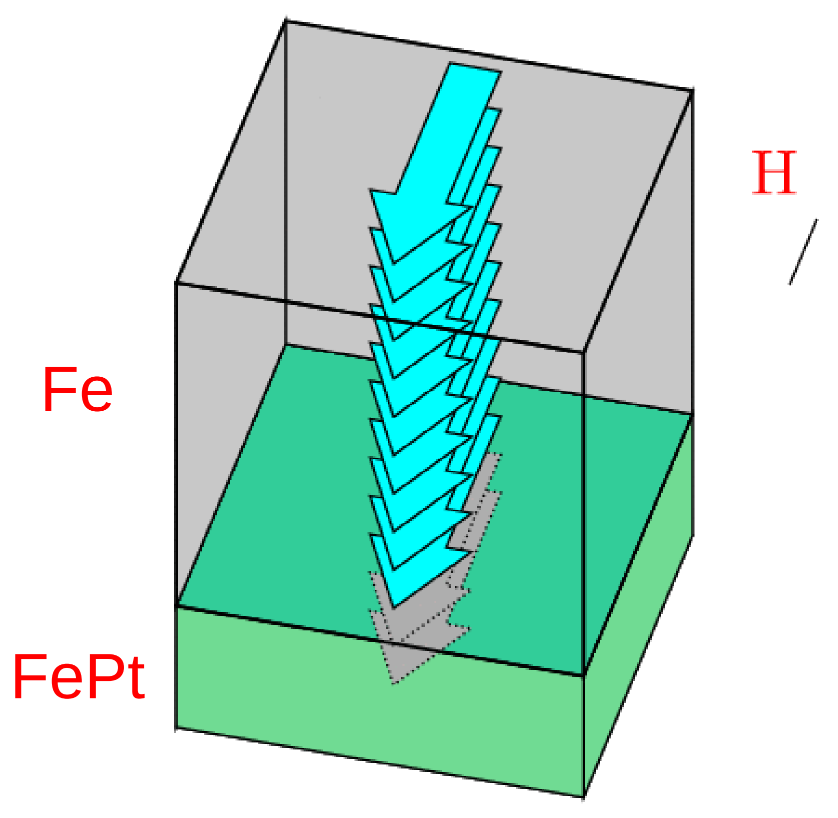
Fe on FePt

Soft - magnetic Fe

Hard - magnetic FePt
with uniaxial anisotropy

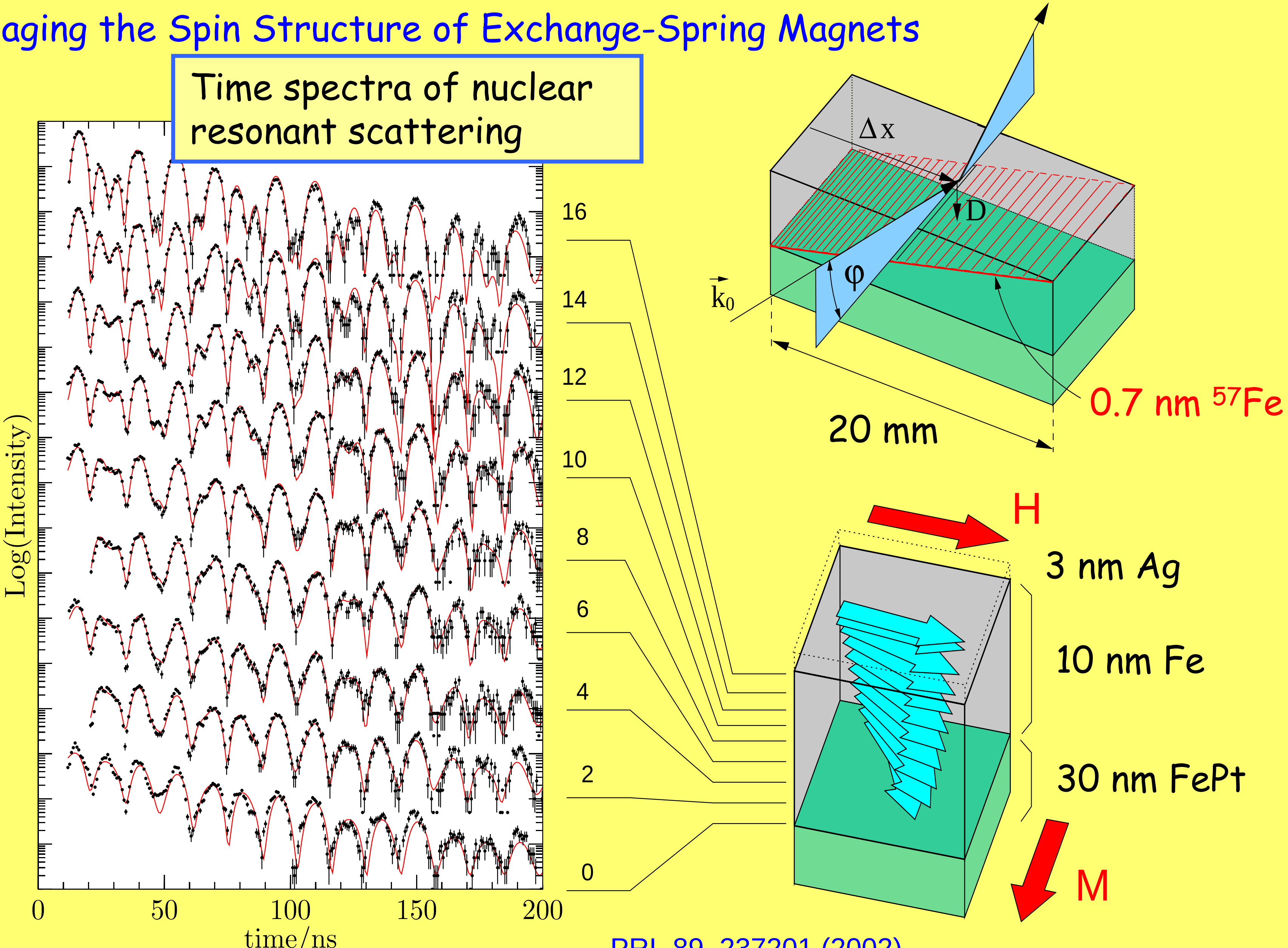
- Exchange coupling at the interface: Parallel alignment of Fe and FePt moments
- With increasing distance from the interface: Coupling becomes weaker
- External field H induces spiral magnetization
- Return to parallel alignment for H = 0

Exchange-Spring magnets



Amplifying the Intensity of Coherent X-ray Scattering (1)

Imaging the Spin Structure of Exchange-Spring Magnets



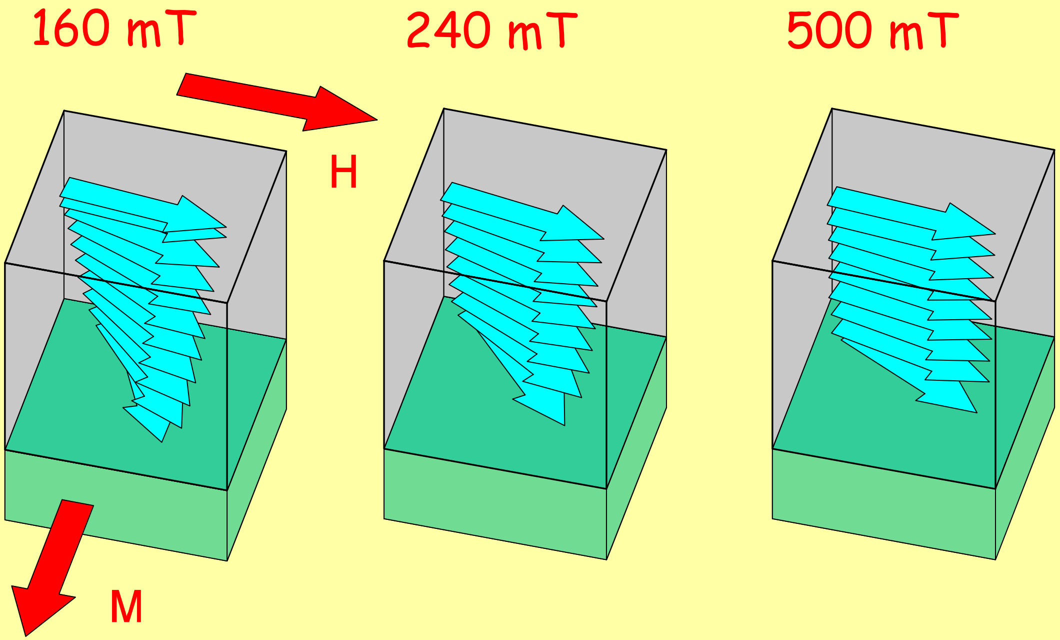
Time spectra of nuclear resonant scattering

PRL 89, 237201 (2002)

The Internal Spin Structure of Exchange-Spring Bilayers :

Fe on FePt

(with Ag capping layer)



Micromagnetic model

Exchange

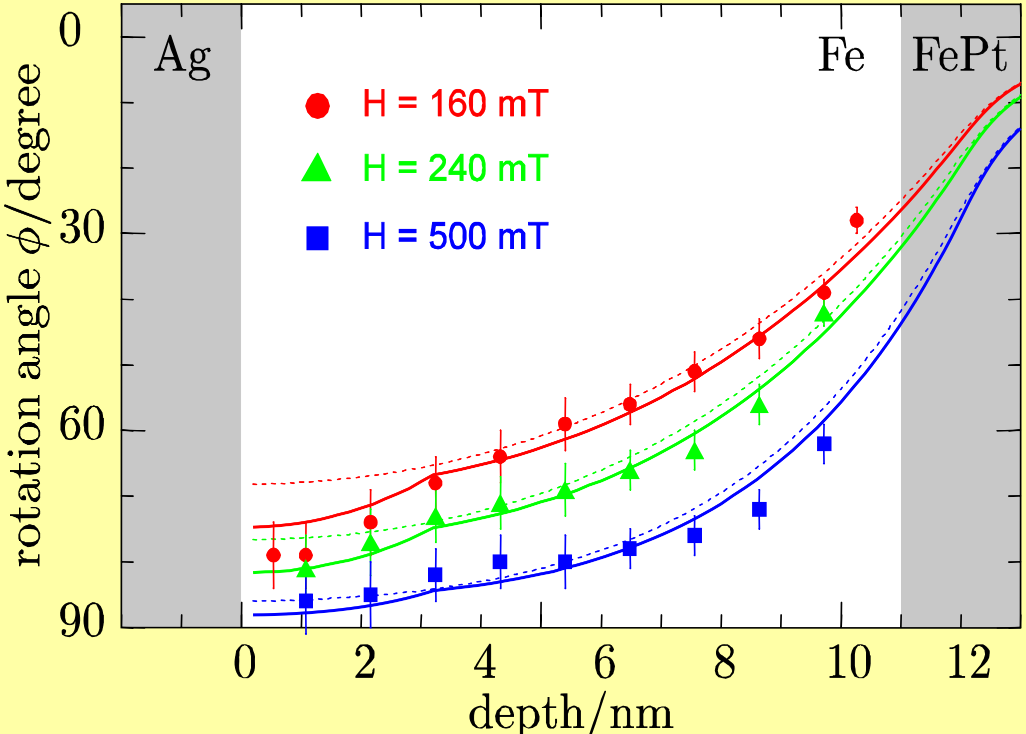
$$E = -\sum_{i=1}^{N-1} \frac{A_{i,i+1}}{d^2} \cos(\varphi_i - \varphi_{i+1})$$

Anisotropy

$$-\sum_{i=1}^{N} K_i \cos^2 \varphi_i$$

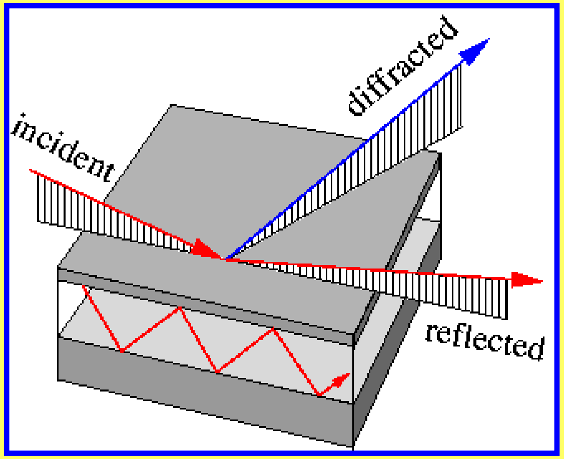
Dipole

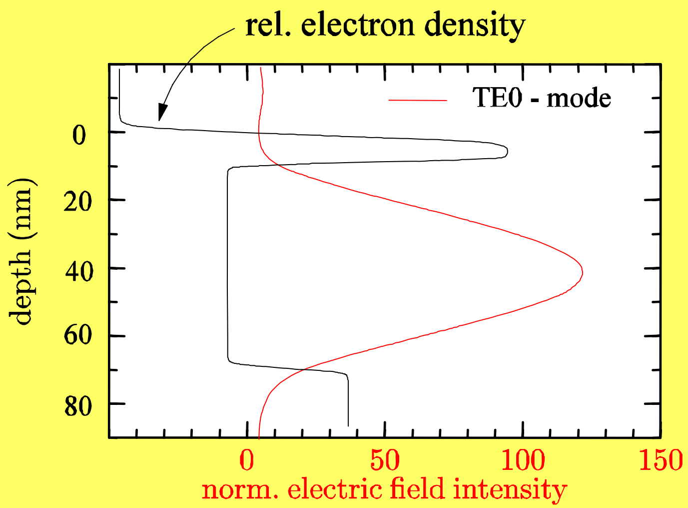
$$-\sum_{i=1}^{N} H M_i \cos(\varphi_i - \varphi_H)$$

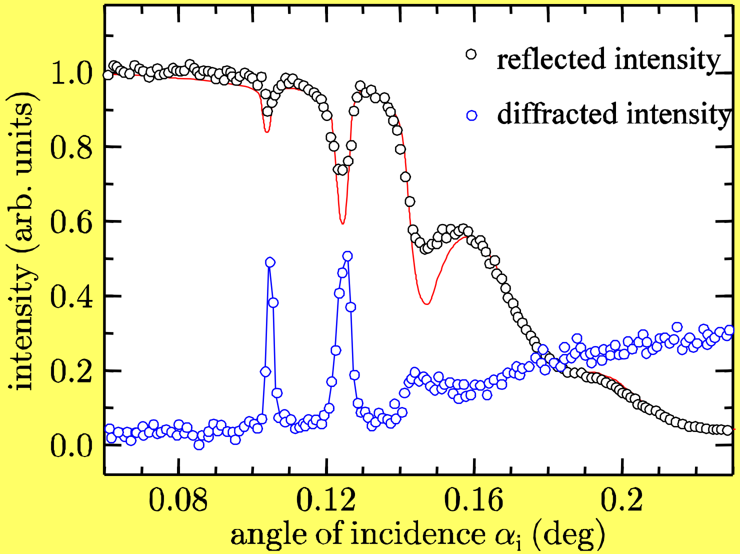


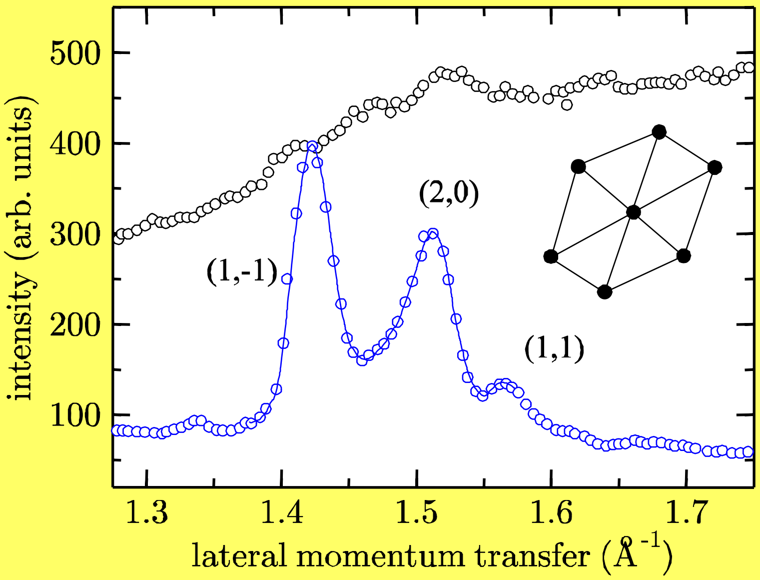
Amplifying the Intensity of Coherent X-ray Scattering (2)

Grazing incidence diffraction from biomolecular membranes







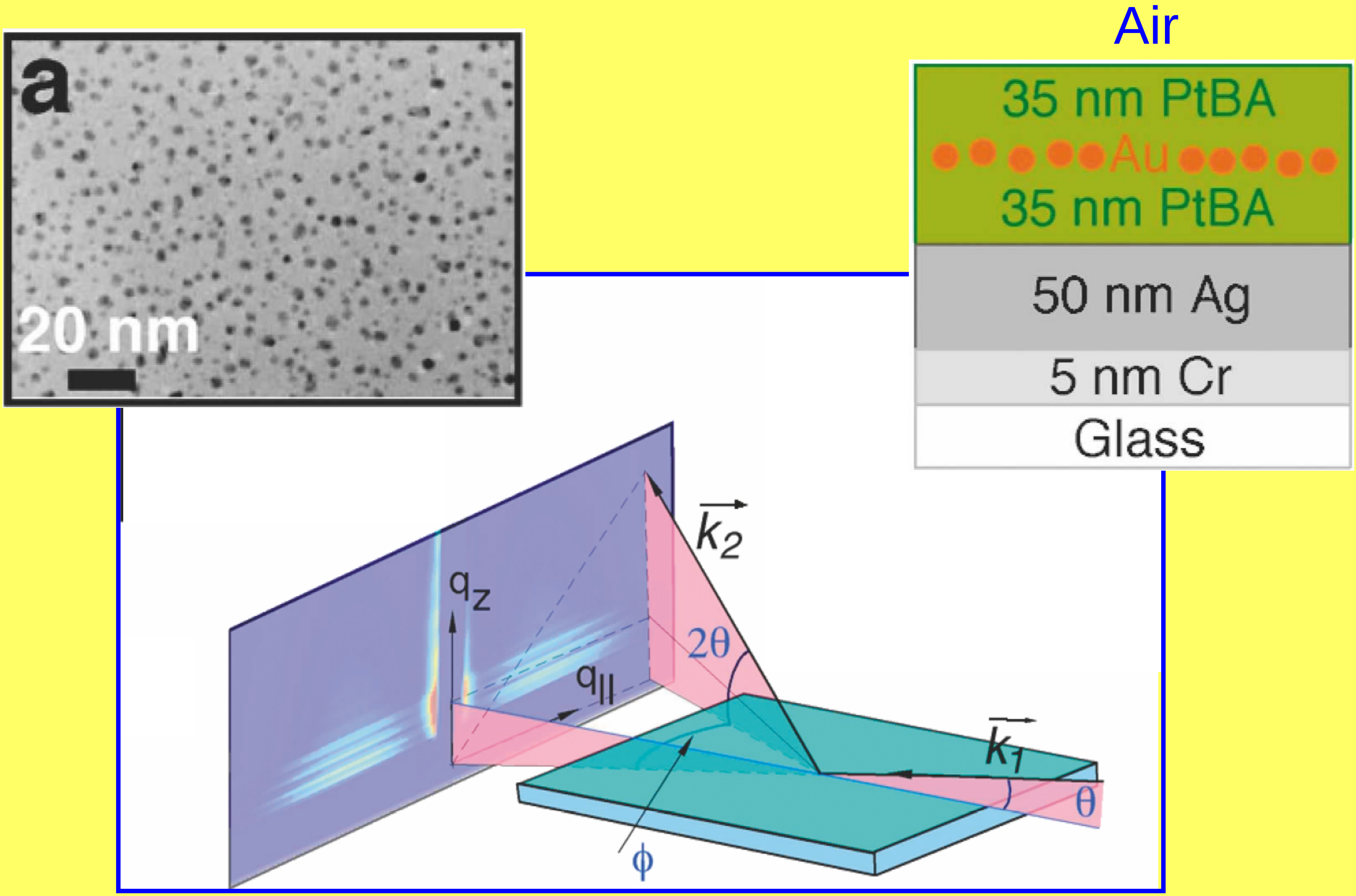


F. Pfeiffer et al., J. Appl. Cryst. 35, 163 (2002)

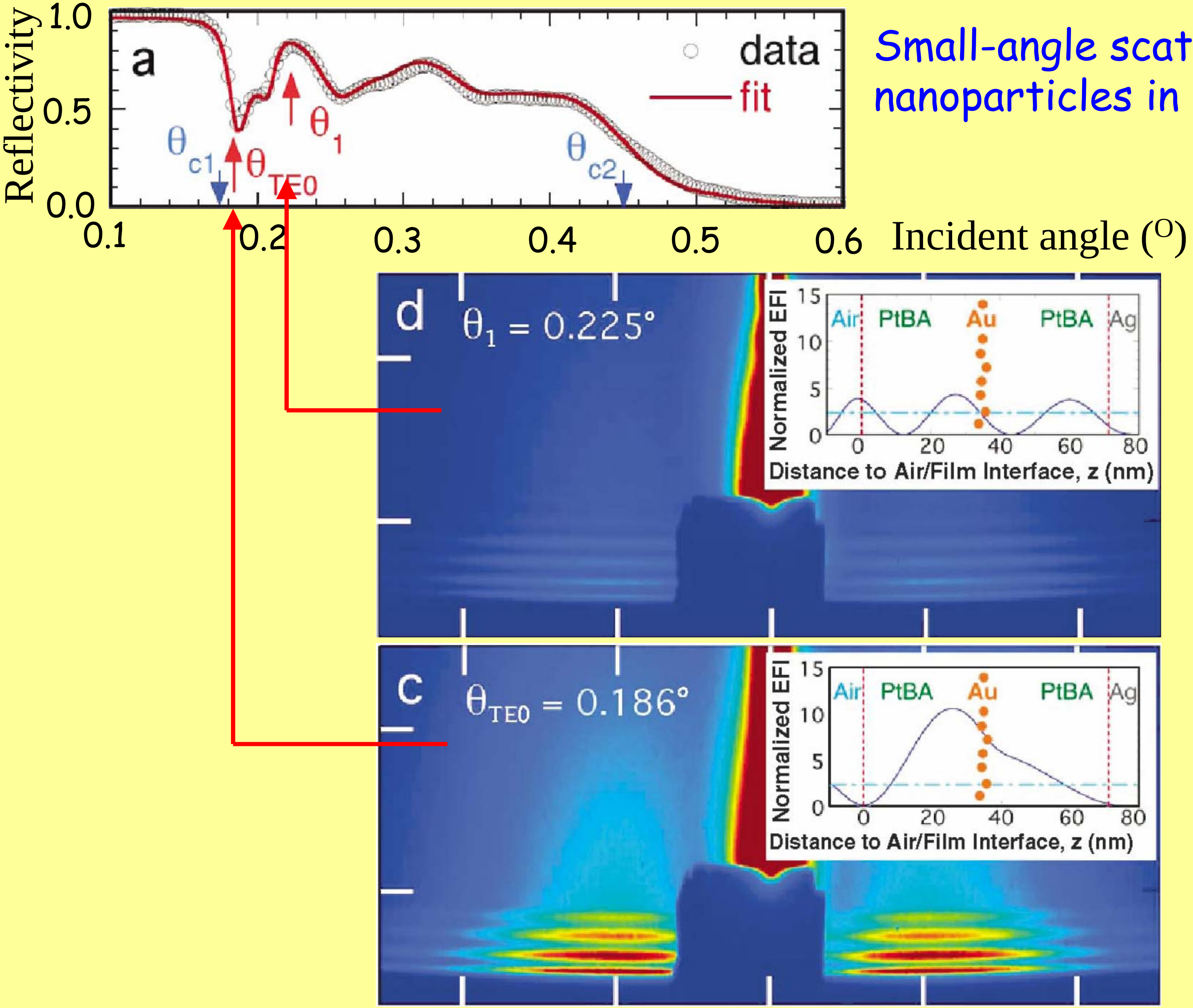
Amplifying the Intensity of Coherent X-ray Scattering (3)

⟶ Small-angle scattering from nanoparticles in a waveguide

S. Narayanan et al., PRL 94, 145504 (2005)



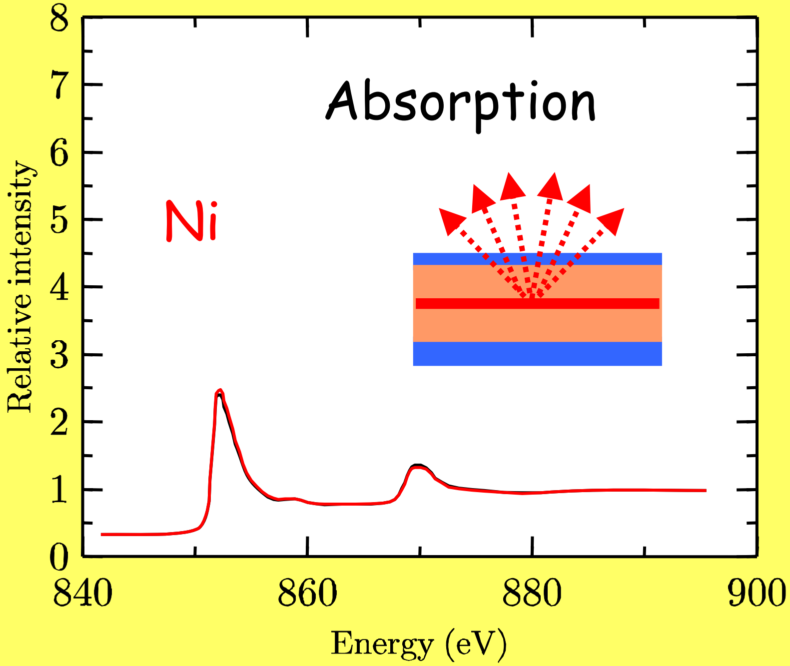
Small-angle scattering from nanoparticles in a waveguide

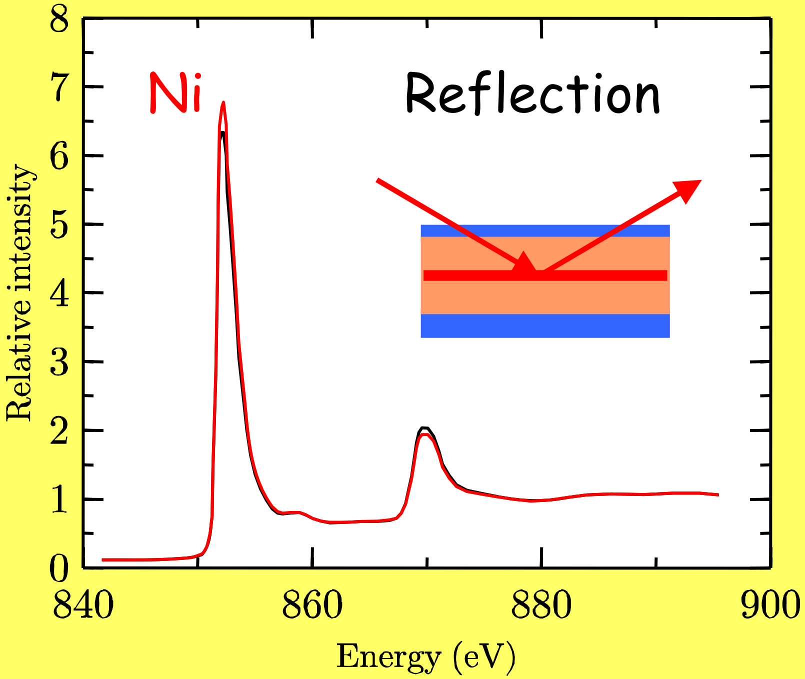


Amplifying the Intensity of Coherent X-ray Scattering (4)

Spectroscopic Reflectivity

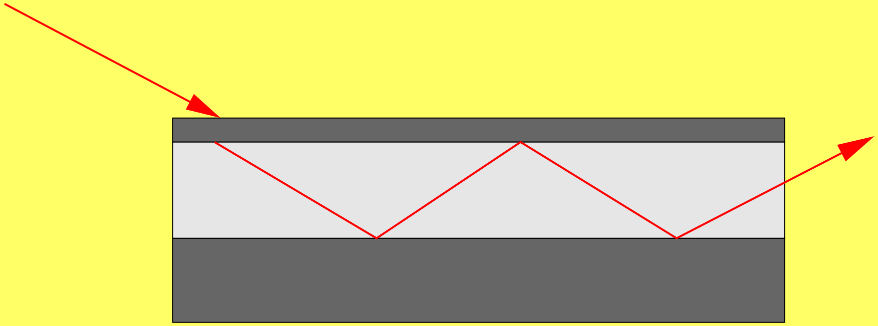
Enhancement of spectroscopic features compared to absorption mode





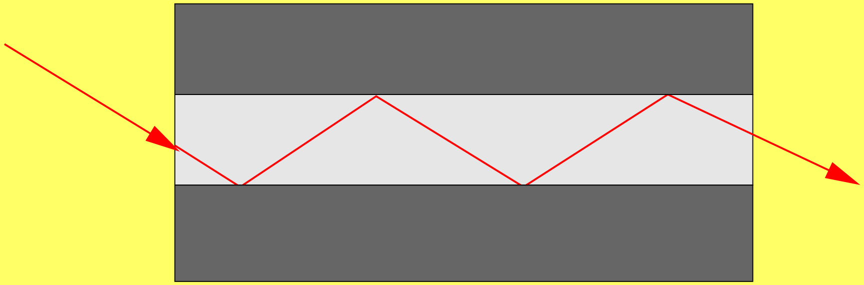
Increasing the photonic density of states in a waveguide

Evanescent Coupling vs. Direct (Front) Coupling



T_1 and R_1 are small

$$I(z) = \left| T_1 e^{ik_\perp z} \frac{1 + R_2\, e^{2ik_\perp (D-z)}}{1 - R_1 R_2\, e^{2ik_\perp D}} \right|^2$$

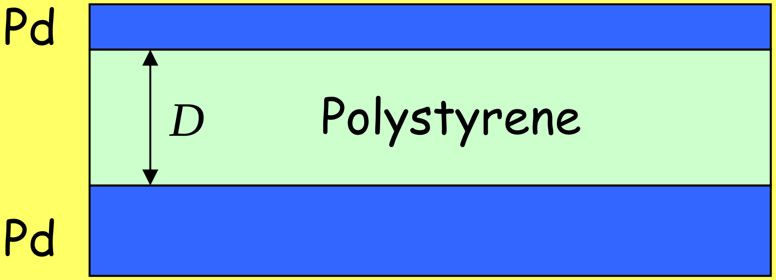


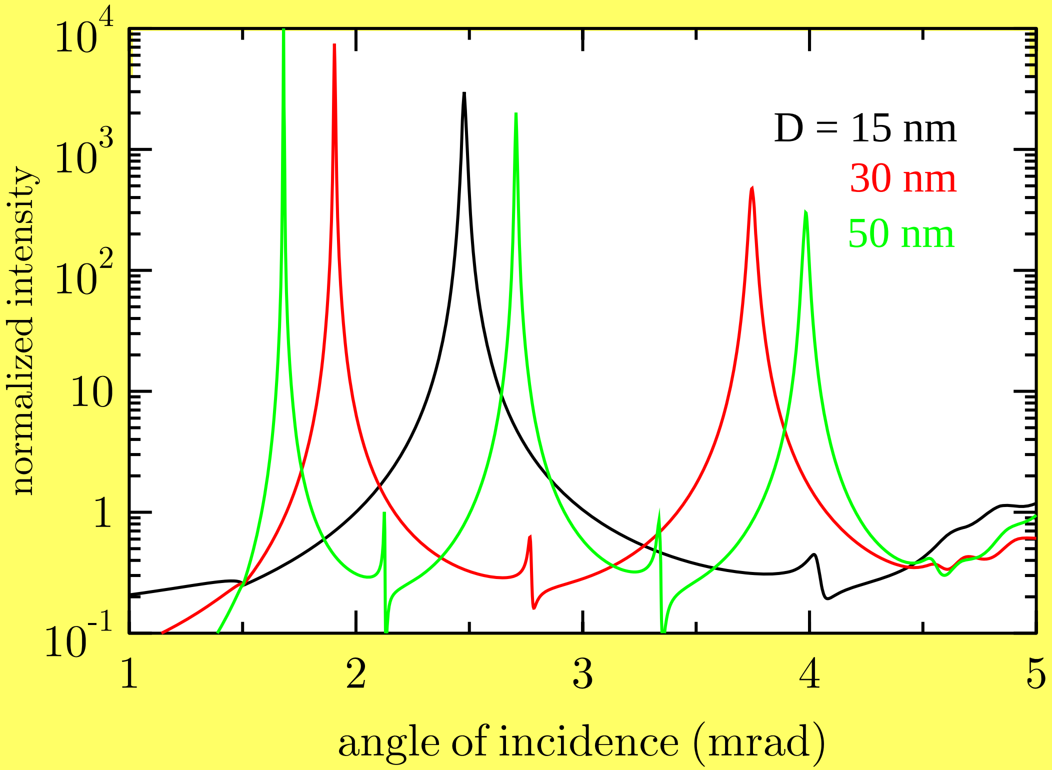
T_1 and R_1 are close to 1

Intensity enhancement in waveguides

Intensity in the center of the guiding layer:

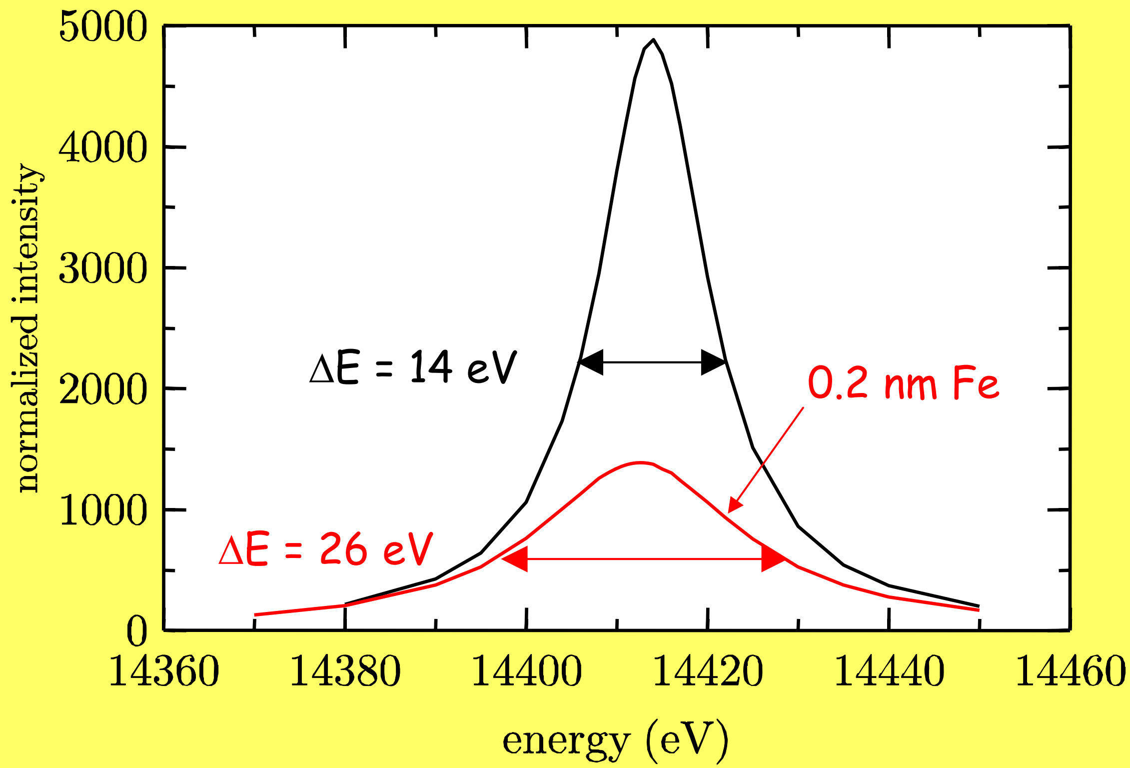
$$I(z_c) = \left| \frac{T\, e^{ik_{1z}D/2}}{1 - R\, e^{2ik_{1z}D}} \right|^2$$





- •High reflectivity at the boundaries
- •Low absorption in the guiding layer
- •Direct coupling of nanobeams into the guiding layer

Cavity quality factor Q



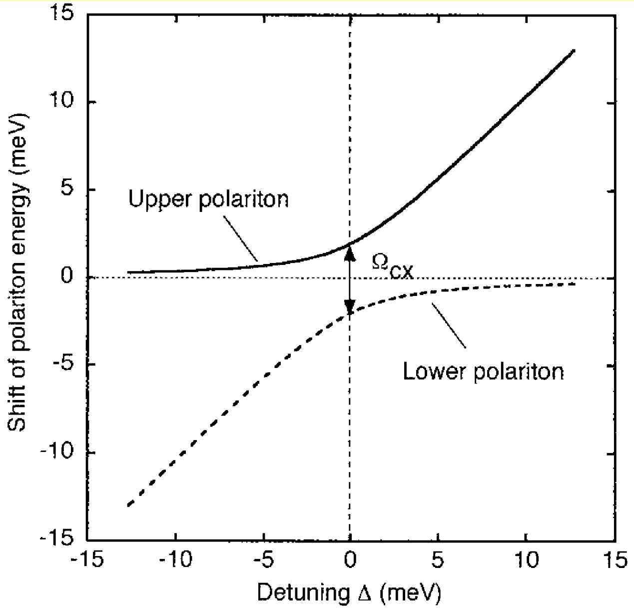
Q = 1000

Q = 550

Strong-coupling limit: Residence time of photons in the cavity is longer than lifetime of excited level

→ Photons are reabsorbed in the cavity

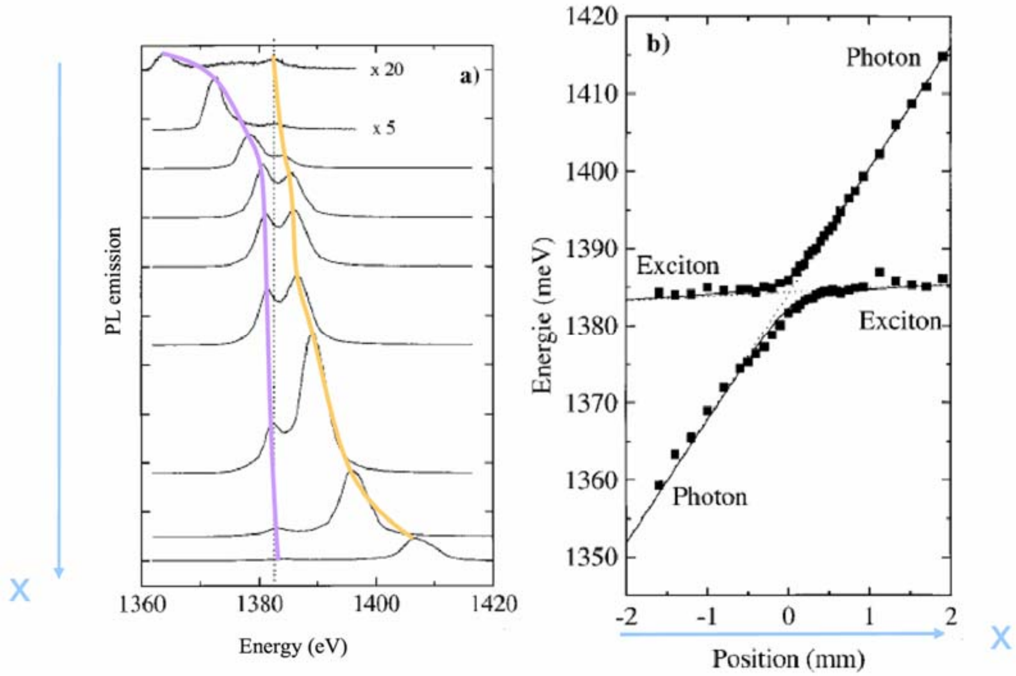
The cavity polariton

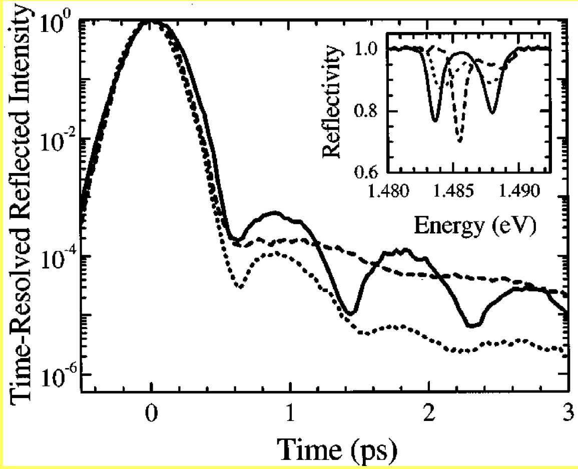


$$E_\pm = E_c + \frac{\Delta}{2} \pm \frac{1}{2}\sqrt{\Delta^2 + \Omega_{cx}^2}$$

Ω_{cx}= vacuum-field Rabi splitting

Photoluminescence from quantum wells in a cavity





Temporal evolution

Khitrova et al,
Rev. Mod. Phys.
71, 1591 (2004)

Conclusion and Outlook

- Resonant atoms in a cavity: Acceleration of the spontaneous emission

  → Time-resolved x-ray scattering with short-pulsed radiation sources

- Enhancement of the intensity of coherent scattering from ultrathin probe layers

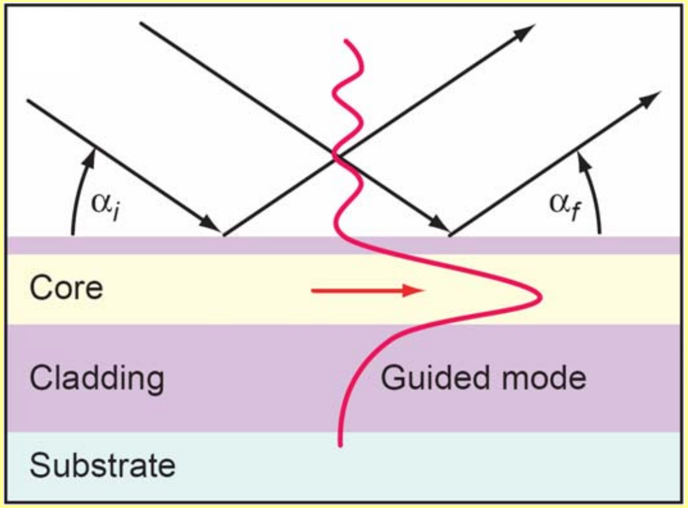
  → Investigation of very small amounts of material

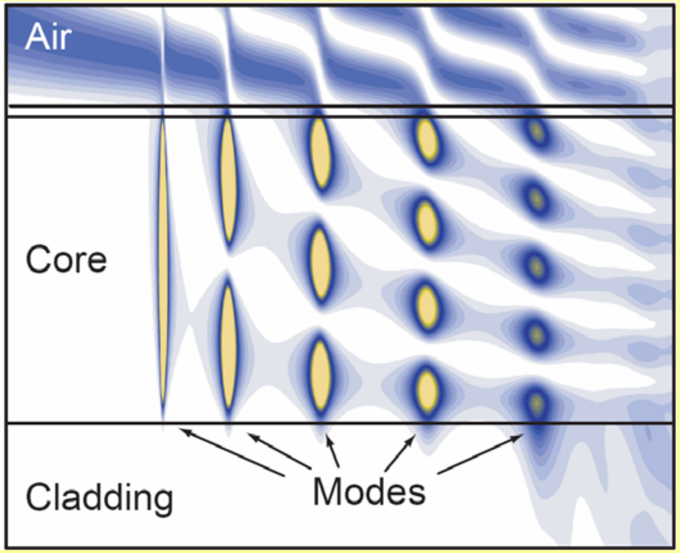
- Manipulating the final state of x-ray photon fields, e.g., increasing the mean number of photons per mode.

Coworkers

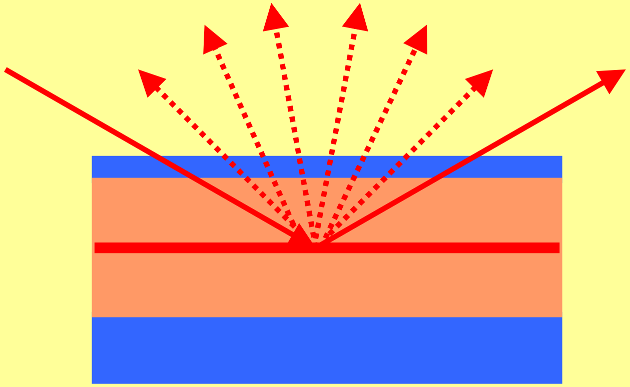
Universität Rostock Germany	Torsten Klein Kai Schlage
Deutsches Elektronen Synchrotron (DESY) Hamburg, Germany	Olaf Leupold
European Synchrotron Radiation Facility Grenoble, France	Rudolf Rüffer

Formation of guided modes in a layered system:

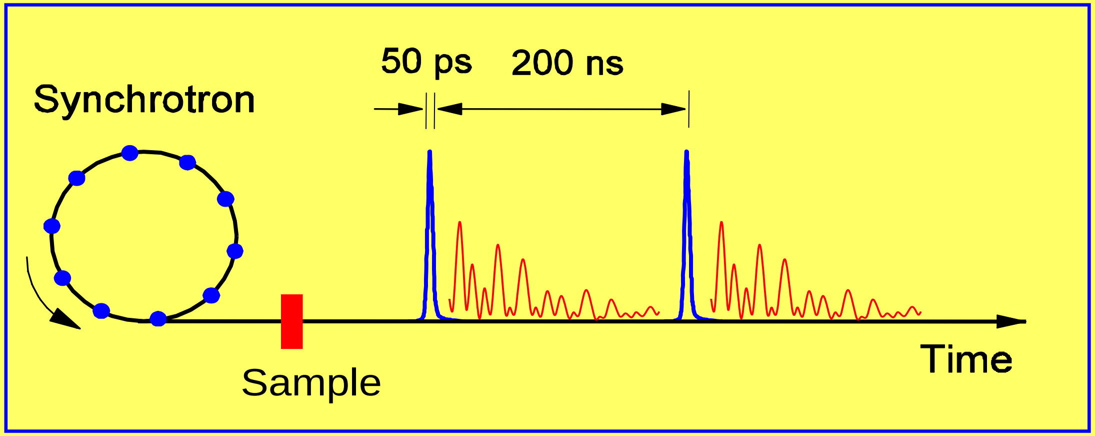




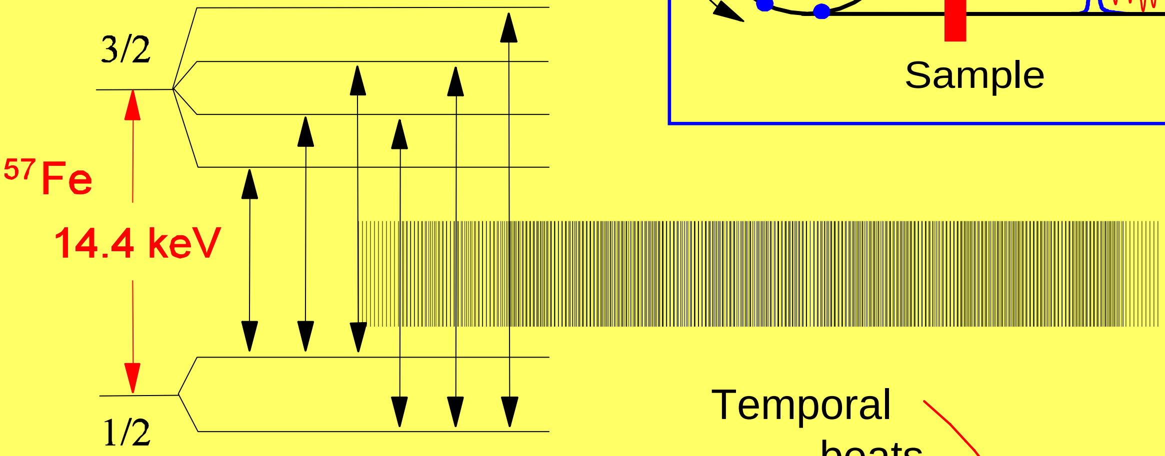
What is the impact on coherent scattering processes from material within the wavefield ?

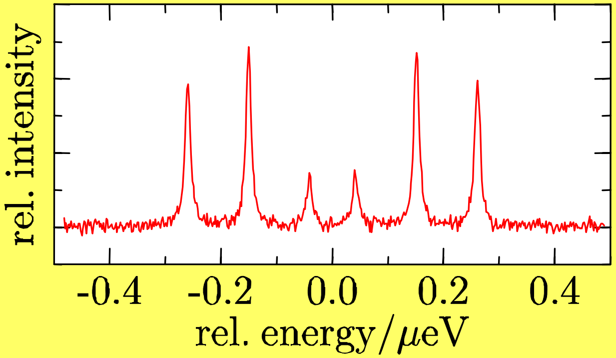


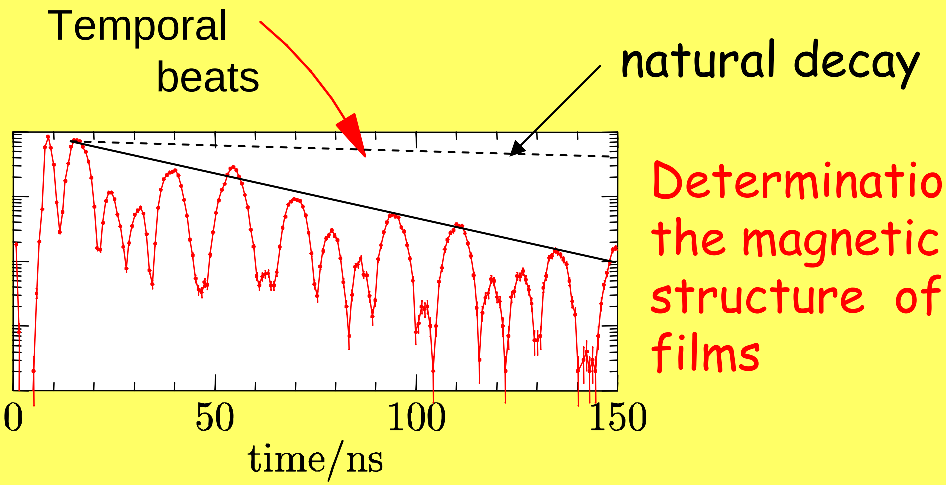
Nuclear Resonant Scattering of Synchrotron Radiation



$\tau_0 = 141$ ns







Determination of the magnetic structure of thin films

Spontaneous Emission in a Cavity: The Total Decay Rate

$$\boxed{I(t) \approx \left(\frac{\Gamma_c}{\Gamma_0}\right)^2 \exp[-(\Gamma_0 + \Gamma_c)\, t/\hbar]}$$

Natural linewidth Γ_0

Coherent enhancement Γ_c

$$\Gamma_c = \Gamma_\gamma\,(1+\alpha)\,\chi \;\; = \Gamma_0\,\chi$$

$$\chi = \varrho\,\sigma_0 f_{LM} d_{\|}/4$$

describes ensemble of atoms

internal conversion

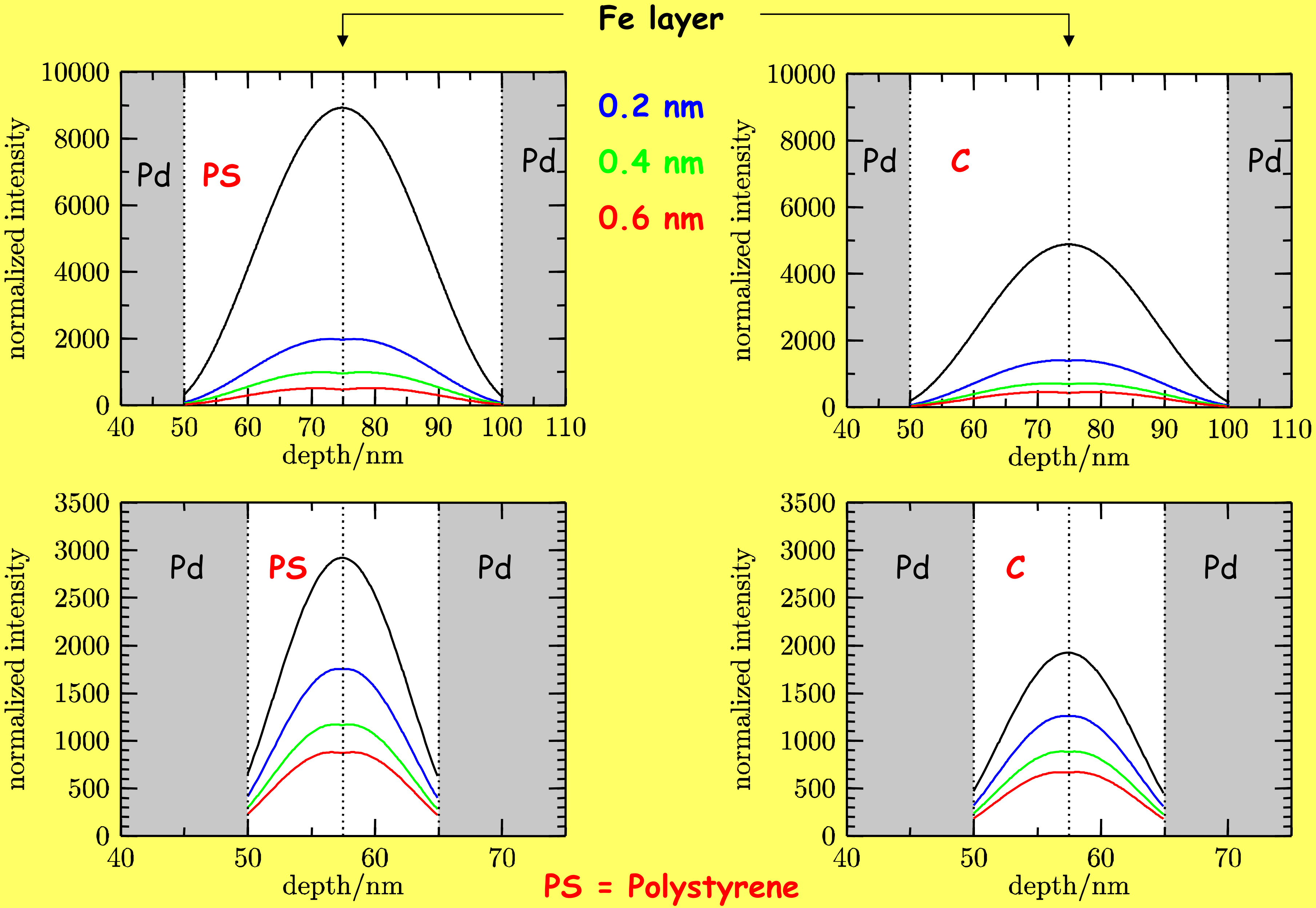
$$\Gamma_\gamma = \frac{2\pi}{\hbar^2}\,\rho(z,\omega_0)\,|\langle g,1|\,\mathbf{H}\,|e,0\rangle|^2$$

radiative decay width

Density of states

$$\boxed{\Gamma_c \;=\; \left(\frac{\rho(z)}{\rho_0}\right)\,\chi\,\Gamma_0}$$

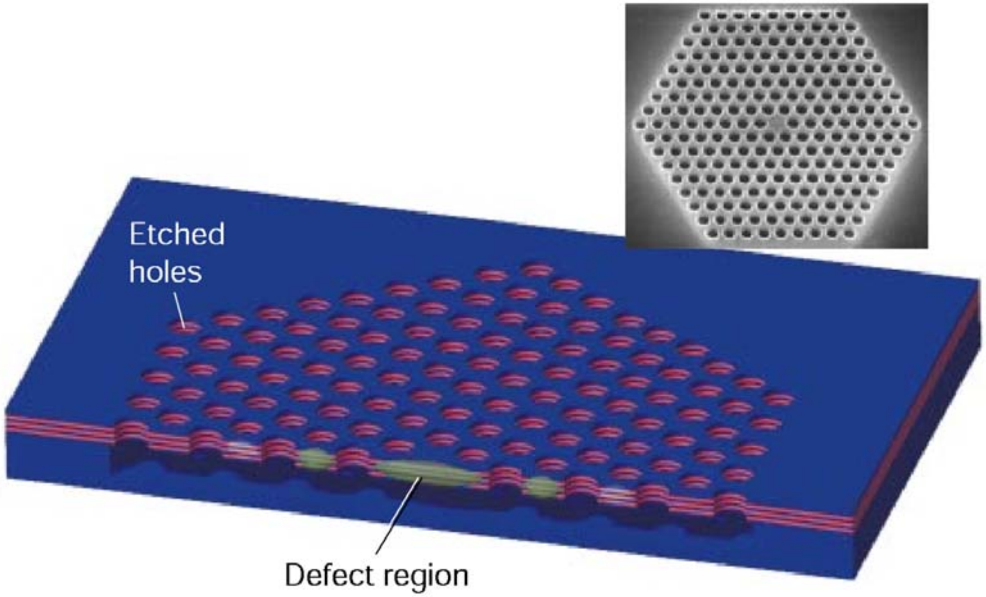
Direct Coupling of X-rays into a Planar Waveguide (2)

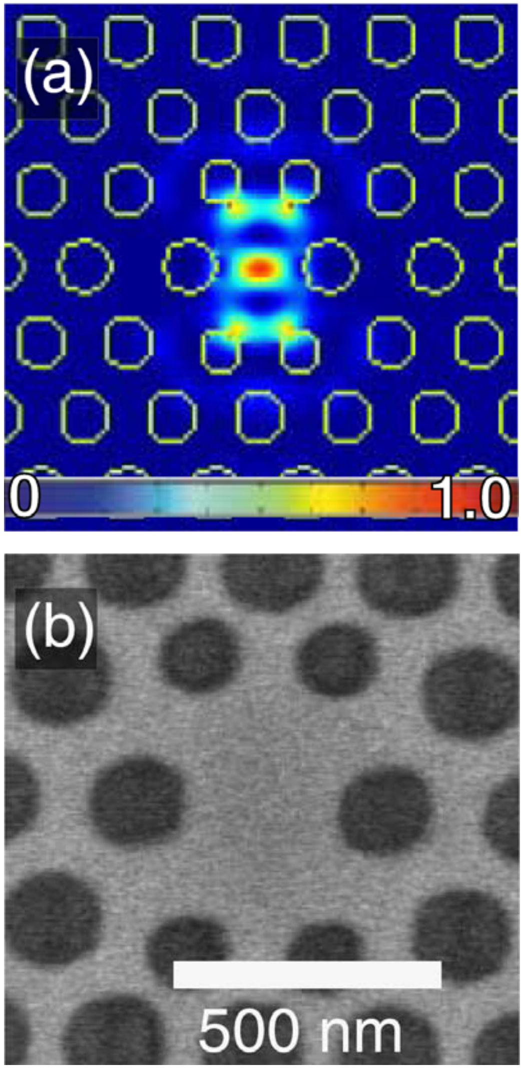


PS = Polystyrene

Putting Light into Boxes (2)

Photonic crystals: Microcavities as defects in the structure, leading to gap states in the photonic band structure

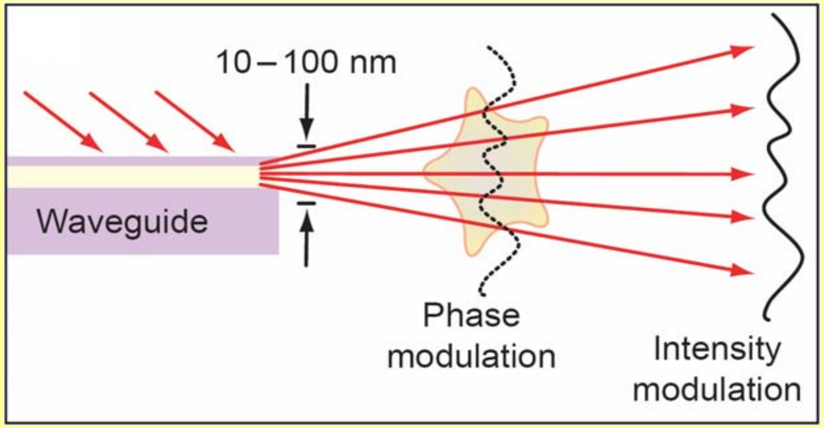




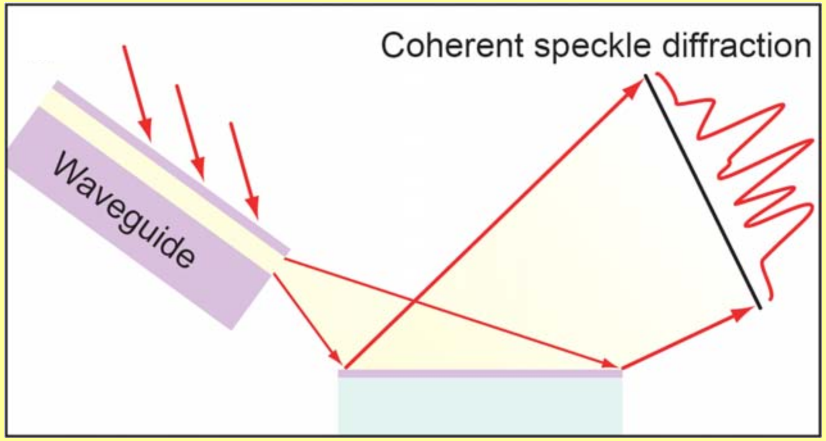
D. Englund et al.
PRL 95, 013904 (2005)

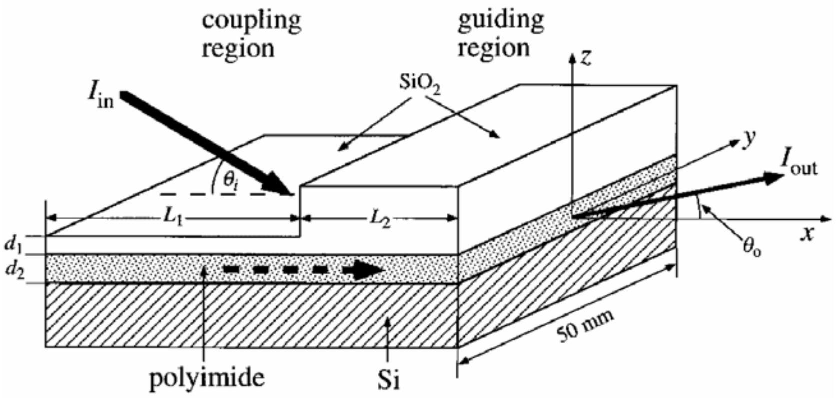
Cavities and Waveguides for X-ray Optical Applications

Phase contrast projection microscopy

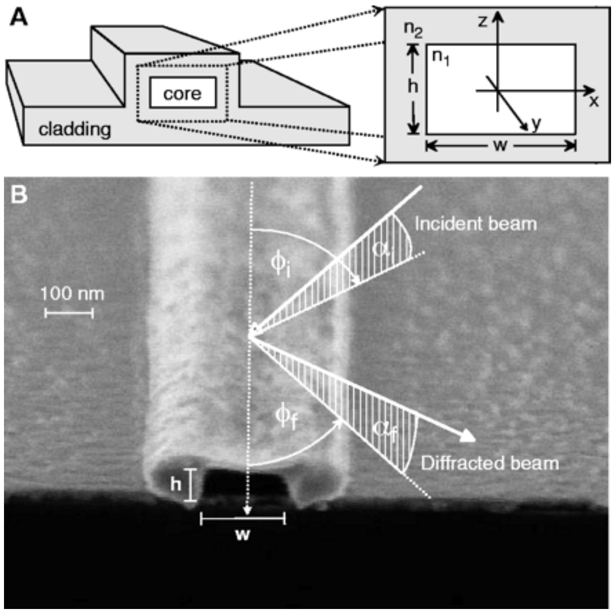


Coherent scattering, photon correlation spectroscopy





Y. P. Feng et al. Appl. Phys. Lett. 67, 3647 (1995)



F. Pfeiffer et al., Science **297**, 230 (2002).