Boosting the Light: X-Ray Physics in Confinement

Ralf Röhlsberger

HASYLAB @ DESY, Hamburg
The Simpletons (Citizens of Schilda) Build a City Hall

Proposal: Trap the light and carry it in

- but they forget the windows
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

Quantum dot
Q > 20000

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

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

How to treat photon emission in a resonator?

Excitation

Reemission

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:

\[
\Gamma = \frac{1}{\tau} = \frac{2\pi}{\hbar} \rho(\mathbf{r}', \omega_0) |\langle g, 1| \mathbf{H} |e, 0\rangle|^2
\]

\[\text{Transition matrix element}\]
\[\text{Density of (final) photon states}\]

→ 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
\[ \rho_c = \frac{1}{V} \frac{1}{\Delta \nu} \]
\[ \Delta \nu = \frac{\nu}{Q}, \quad V = \left( \frac{\lambda}{2} \right)^3 \]
\[ \rho_c = \frac{2}{\pi^2} \frac{\omega^2}{c^3} Q \]

Enhancement of the radiative decay rate
\[ \frac{\Gamma}{\Gamma_0} = \frac{\rho_c}{\rho_0} = \frac{Q}{\pi} \]

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

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

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300\(^{\circ}\)K, for \(\nu = 10^7\) sec\(^{-1}\), \(\mu = 1\) nuclear magneton, the corresponding relaxation time would be \(5 \times 10^4\) 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 \(\nu/Q\) associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor \(f = 3Q\lambda^3/4\pi^2V\), 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 \(\delta\) is the skin-depth at frequency \(\nu\), \(f \sim \lambda^3/a^2\delta\). 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\(^{-1}\).

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

\(Q\) = quality factor
\(\lambda\) = wavelength
\(n\) = index of refraction
\(V\) = cavity volume

Nobel Prize in Physics 1952
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 $^{57}\text{Fe}$ with $\tau = 141$ 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:

Temporal Evolution of Scattering Processes (1)

Incoherent Scattering

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

Coherent scattering in confinement

The excited state can decay through many photonic states

\[ I(t) \sim \exp\left[-\left(1 + \chi\right) \frac{t}{\tau_0}\right] \]

\[ \chi \sim \rho d \]

More Speedup

\[ I(t) \sim \exp\left[-\left(1 + B\chi\right) \frac{t}{\tau_0}\right] \]
The additional enhancement factor is determined by the photonic density of states in the cavity:

\[ B = \frac{\rho_c}{\rho_0} \]

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

$^{57}\text{Fe}$ 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

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

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

PRL 95, 097601 (2005)
Coherent Scattering and the Photonic Density of States

Temporal Dependence

\[ I(t) \sim \exp[-(1 + B\chi) \frac{t}{\tau_0}] \]

with \( B = \frac{\rho_c}{\rho_0} \)

Total Intensity

Coherent Scattering Amplitude

\[ f_{coh} \sim \frac{\rho_c}{\rho_0} = B \]

\[ I_{tot} \sim |f_{coh}|^2 \sim B^2 \]

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

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

PRB 69, 235412 (2004)
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**
- Boosting the intensity of coherent x-ray scattering

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

Imaging the Spin Structure of Exchange-Spring Magnets

Time spectra of nuclear resonant scattering

Log(Intensity)

0 50 100 150 200

time/ns

0.7 nm $^{57}$Fe

20 mm

3 nm Ag

10 nm Fe

30 nm FePt

PRL 89, 237201 (2002)
The Internal Spin Structure of Exchange-Spring Bilayers:
Fe on FePt
(with Ag capping layer)

Micromagnetic model

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

Exchange

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

Anisotropy

\[ - \sum_{i=1}^{N} HM_i \cos(\varphi_i - \varphi_H) \]

Dipole

The graph shows the rotation angle (\(\phi\)) in degrees against depth (in nm) for different magnetic fields (H = 160 mT, H = 240 mT, H = 500 mT). The colors correspond to the different magnetic fields, with red dots for 160 mT, green triangles for 240 mT, and blue squares for 500 mT.
Amplifying the Intensity of Coherent X-ray Scattering (2)

Grazing incidence diffraction from biomolecular membranes

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

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

\[ T_1 \text{ and } R_1 \text{ are small} \]

\[ T_1 \text{ and } R_1 \text{ are close to 1} \]
Intensity enhancement in waveguides

Intensity in the center of the guiding layer:

\[ I(z_c) = \left| \frac{T e^{ik_1zD/2}}{1 - R e^{2ik_1zD}} \right|^2 \]

- High reflectivity at the boundaries
- Low absorption in the guiding layer
- Direct coupling of nanobeams into the guiding layer
Cavity quality factor $Q$

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

Photoluminescence from quantum wells in a cavity

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

\( \Omega_{cx} \) = vacuum-field Rabi splitting

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

<table>
<thead>
<tr>
<th>Institution</th>
<th>Name</th>
</tr>
</thead>
<tbody>
<tr>
<td>Universität Rostock</td>
<td>Torsten Klein</td>
</tr>
<tr>
<td>Germany</td>
<td>Kai Schlage</td>
</tr>
<tr>
<td>Deutsches Elektronen Synchrotron (DESY)</td>
<td>Olaf Leupold</td>
</tr>
<tr>
<td>Hamburg, Germany</td>
<td></td>
</tr>
<tr>
<td>European Synchrotron Radiation Facility</td>
<td>Rudolf Rüffer</td>
</tr>
<tr>
<td>Grenoble, France</td>
<td></td>
</tr>
</tbody>
</table>
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

\[ \tau_0 = 141 \text{ ns} \]

\[ ^{57}\text{Fe} \]

14.4 keV

3/2

1/2

Temporal beats

natural decay

Determination of the magnetic structure of thin films

Sample

Synchrotron

50 ps 200 ns

Time

rel. intensity

rel. energy/\mu eV

rel. energy/\mu eV

time/ns

0 50 100 150
Spontaneous Emission in a Cavity: The Total Decay Rate

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

\[
\Gamma_c = \Gamma_\gamma (1 + \alpha) \chi = \Gamma_0 \chi
\]

\[
\chi = \varrho \sigma_0 f_{LM} d_{\parallel}/4
\]

describes ensemble of atoms

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

\[
\text{radiative decay width}
\]

\[
\Gamma_c = \left( \frac{\rho(z)}{\rho_0} \right) \chi \Gamma_0
\]

Natural linewidth \( \Gamma_0 \)

Coherent enhancement \( \Gamma_c \)

Internal conversion

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

Fe layer

PS = Polystyrene
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
