Radiation-matter interaction

Classical dipoles
- Dipole radiation
- Power radiated by a classical dipole in an inhomogeneous environment
- The local density of optical states (LDOS)

Quantum emitters
- Lifetime of quantum emitters
- Fluorescence lifetime measurements
- Fermi’s Golden Rule and decay rate engineering by shaping LDOS

Spontaneous emission control
- Drechsel’s experiment
- The Purcell effect
- Microcavities

Optical antennas
- Radiation reaction in dipolar scatterers
- Decay rate engineering with optical antennas
Previously ... the Green function

\[ E(r) = \omega^2 \mu_0 \mu \mathbf{G}(r, r_0) \mathbf{p} \]

Field at \( r \) generated by dipole at \( r_0 \)

In a homogeneous medium:

\[ \mathbf{G}(r, r_0) = \mathbf{G}_0(r, r_0) \]
Previously ... the Green function

\[ E(\mathbf{r}) = \omega^2 \mu_0 \mu \mathbf{G}(\mathbf{r}, \mathbf{r}_0) \mathbf{p} \]

Field at \( \mathbf{r} \) generated by dipole at \( \mathbf{r}_0 \)

In an inhomogeneous environment:

\[ \mathbf{G}(\mathbf{r}, \mathbf{r}_0) = \mathbf{G}_0(\mathbf{r}, \mathbf{r}_0) + \mathbf{G}_s(\mathbf{r}, \mathbf{r}_0) \]
Previously ... the power radiated by a dipole

\[
E(r) = \omega^2 \mu_0 \mu \hat{G}(r, r_0) p
\]

\[
E(r_0) = \omega^2 \mu_0 \mu \hat{G}(r_0, r_0) p
\]

\[
\langle P \rangle = \frac{\omega}{2} \text{Im} [p^* \cdot E(r_0)]
\]

Dipole does work against its own field!

\[
\langle P \rangle = \frac{\omega}{2} |p|^2 \left\{ n_p^\top \text{Im} \left[ \omega^2 \mu_0 \mu \hat{G}(r_0, r_0) \right] n_p \right\}
\]

In a homogeneous medium:

\[
\hat{G}_0(r_0, r_0)
\]

\[
n_p^\top \text{Im} \left[ \omega^2 \mu_0 \mu \hat{G}_0(r_0, r_0) \right] n_p = \frac{k}{6\pi}
\]

\[
\langle P_0 \rangle = \frac{\omega k^3}{12\pi \varepsilon \varepsilon_0} |p|^2
\]
Previously ... the power radiated by a dipole

\[ E(r) = \omega^2 \mu_0 \mu \vec{G}(r, r_0) p \]

\[ E(r_0) = \omega^2 \mu_0 \mu \vec{G}(r_0, r_0) p \]

\[ \langle P \rangle = \frac{\omega}{2} \text{Im} [p^* \cdot E(r_0)] \]

Dipole does work against its own field!

\[ \langle P \rangle = \frac{\omega}{2} |p|^2 \left\{ n_p^\top \text{Im} \left[ \omega^2 \mu_0 \mu \vec{G}(r_0, r_0) \right] n_p \right\} \]

In an inhomogeneous environment:

\[ \vec{G}_0(r_0, r_0) \]

\[ \vec{G}_s(r, r_0) \]

\[ r_0 \]

\[ \vec{G} = \vec{G}_0 + \vec{G}_s \]
Light sources

Radiating sources at 1000 THz (visible):

Atoms

Dye molecules

Quantum dots

“Optics/visible” @ 1 eV, with $h$ and $c$: $\lambda \approx 1\mu m$, “sub-\(\lambda\) optics” is “Nano-Optics”

Thermal noise

$kT$

100 meV

1 eV

Ionizing

Ry

10 eV

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Quantum emitters

Radiating sources at 1000 THz:

Atoms

Dye molecules

Quantum dots

Optical emitters have discrete level scheme. Let’s focus on the two lowest levels. How long will the system remain in its excited state?
Probability to find the system in the excited state decays exponentially with rate $\gamma$.

$$p_e(t) = p(0) \exp[-\gamma t]$$

How can we measure the population of the excited state?
Fluorescence lifetime measurements

The probability to detect a photon at time $t$ is proportional to $p(t)$:

$$p_e(t) = p_e(0) \exp \left[-\gamma t\right]$$

1. Prepare system in excited state with light pulse at $t=0$
2. Record time delay $t_1$
3. Repeat experiment many times
4. Histogram arrival time delays
Calculation of decay rate $\gamma$

Fermi’s Golden Rule:

$$\gamma = \sum_f \frac{2\pi}{\hbar} |\langle f | \hat{\mathcal{H}} | i \rangle|^2 \delta(E_i - E_f)$$

Initial state (excited atom, no photon):

$$|i\rangle = |e, 0\rangle$$

Final state (de-excited atom, 1 photon in state $k$ at frequency $\omega$):

$$|f\rangle = |g, 1\omega_k\rangle$$

Interaction Hamiltonian:

$$\hat{\mathcal{H}} = -\hat{p} \cdot \hat{E}$$
Calculation of decay rate $\gamma$

Fermi’s Golden Rule:

$$
\gamma = \sum_f \frac{2\pi}{\hbar} \left| \left\langle f \mid \hat{H} \mid i \right\rangle \right|^2 \delta(E_i - E_f)
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$$
|f\rangle = |g, 1_{\omega_k}\rangle
$$

Interaction Hamiltonian:

$$
\hat{H} = -\hat{p} \cdot \hat{E}
$$

Sum over final states is sum over photon states ($k$) at transition frequency $\omega$.

$$
\gamma = \frac{\pi \omega}{3\hbar\epsilon_0} \left| \hat{p} \right|^2 \rho_n(r_0, \omega)
$$

Atomic part:

transition dipole moment (quantum)

$$
\left| \hat{p} \right|^2 = \left| \left\langle g \mid \hat{p} \mid e \right\rangle \right|^2
$$

Field part:

Local density of states (classical)

$$
\rho_n(r_0, \omega) = \frac{6\omega}{\pi c^2} \left\{ n_p \Im \left[ \hat{G}(r_0, r_0; \omega) \right] n_e \right\}
$$
Decay rate engineering

\[ \gamma = \frac{\pi \omega}{3 \hbar \varepsilon_0} \left| \hat{p} \right|^2 \rho_n(r_0, \omega) \]

**Emitter**
Transition dipole moment:
Wave function engineering by synthesizing molecules, and quantum dots
Chemistry, material science

**Environment**
LDOS: Electromagnetic mode engineering by shaping boundary conditions for Maxwell’s equations
Physics, electrical engineering

antennaking.com, Wikimedia, emory.edu

www.photonics.ethz.ch
Transition dipole moment is NOT classical dipole moment, but Classical electromagnetism CANNOT make a statement about the absolute decay rate of a quantum emitter.

BUT: Classical electromagnetism CAN predict the decay rate enhancement provided by a photonic system as compared to a reference system.

\[
\gamma = \frac{\pi \omega}{3 \hbar \epsilon_0} |\hat{p}|^2 \rho_n(r_0, \omega)
\]

\[
\langle P \rangle = \frac{\pi \omega^2}{12 \epsilon \epsilon_0} |p|^2 \rho_n(r_0, \omega)
\]

\[
\frac{\gamma}{\gamma_0} = \frac{P}{P_0}
\]
Drexhage’s experiment (late 1960s)

First observation of the local (!) character of the DOS!

Drexhage, J. Lumin. 1,2; 693 (1970)

www.photonics.ethz.ch
Drexhage’s experiment (late 1960s)

Emitter sees its own mirror image. 
$G_s$ is given as the field generated by the mirror dipole.
1946 - E. M. Purcell predicts modification of spontaneous emission rates in complex media

Edward M. Purcell
1912-1997

Phys. Rev. 69, 681 (1946)
Density of states in the lossless resonator

How many modes in frequency band $[\omega, \omega + \Delta \omega]$ and resonator volume $V$?

\[ \rho(\omega) = \frac{1}{V} \frac{dN(\omega)}{d\omega} \]

$\rho(\omega)$: density of states
$\omega$: angular frequency
$V$: resonator volume
$N$: number of modes
Density of states in the lossless resonator

How many modes in frequency band $[\omega, \omega+\Delta\omega]$ and resonator volume $V$?

$$N(\omega) = V \frac{\omega^3 n^3(\omega)}{3\pi^2 c^3}$$

$N(\omega)$: number of modes
$V$: resonator volume
$n(\omega)$: refractive index
$c$: speed of light

$$\rho(\omega) = \frac{1}{V} \frac{dN(\omega)}{d\omega}$$

$\rho(\omega)$: density of states

In free space (large resonator):

$$\rho(\omega) = \frac{\omega^2 n^3(\omega)}{\pi^2 c^3}$$
Density of states in a realistic resonator

How many modes in frequency band \([\omega, \omega+\Delta\omega]\) and resonator volume \(V\)?

\[
N(\omega) = V \frac{\omega^3 n^3(\omega)}{3\pi^2 c^3}
\]

\[
\rho(\omega) = \frac{1}{V} \frac{dN(\omega)}{d\omega}
\]

- \(V\): resonator volume
- \(N\): number of modes

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In free space (large resonator):

\[
\rho(\omega) = \frac{\omega^2 n^3(\omega)}{\pi^2 c^3}
\]
Density of states in a realistic resonator

How many modes in frequency band \([\omega, \omega + \Delta\omega]\) and resonator volume \(V\)?

- Losses broaden delta-spike into Lorentzian
- Area under Lorentzian is unity
- The lower the loss, the higher the density of states on mode resonance
- Density of states on resonance exceeds that of free space

In free space (large resonator):

\[
\rho(\omega) = \frac{\omega^2 n^3(\omega)}{\pi^2 c^3}
\]
The Purcell effect

Free space: Density of states via Green function. Alternatively, count states in large box (see EM course).

In cavity: Lorentzian with one mode per $\Delta \omega$ and cavity volume $V$
The Purcell effect

Free space: Density of states via Green function.
Alternatively, count states in large box (see EM course).

In cavity: Lorentzian with one mode per $\Delta \omega$ and cavity volume $V$

$$\rho_0(\omega) = \frac{\omega^2 n^3}{\pi^2 c^3}$$

$$\rho_n(r_0, \omega) = \frac{6\omega n^2}{\pi c^2} \left\{ n_p^\dagger \text{Im} \left[ \hat{G}_0(r_0, r_0) \right] n_p \right\}$$

$$\rho_{cav} = \frac{\Delta \omega}{2\pi V} \frac{1}{(\omega - \omega_0)^2 + (\Delta \omega/2)^2}$$

$$\mathcal{F}_P = \frac{\rho_{cav}(\omega_0)}{\rho_0(\omega_0)} = \frac{3}{4\pi^2} \left( \frac{\lambda}{n} \right)^3 \frac{Q}{V}$$

$$Q = \frac{\omega}{\Delta \omega}$$
The Purcell effect

\[ \mathcal{F}_P = \frac{\rho_{\text{cav}}(\omega_0)}{\rho_0(\omega_0)} = \frac{3}{4\pi^2} \left( \frac{\lambda}{n} \right)^3 \frac{Q}{V} \]

The Purcell factor is the maximum rate enhancement provided by a cavity given that the source is
1. Located at the field maximum of the mode
2. Spectrally matched exactly to the mode
3. Oriented along the field direction of the mode

Caution: Purcell factor is only defined for a cavity. The concept of the LDOS is much more general and holds for any photonic system.
Observation of Cavity-Enhanced Single-Atom Spontaneous Emission

P. Goy, J. M. Raimond, M. Gross, and S. Haroche

Laboratoire de Physique de l'Ecole Normale Supérieure, F-75231 Paris Cedex 05, France
(Received 1 April 1983)

It has been observed that the spontaneous-emission lifetime of Rydberg atoms is shortened by a large ratio when these atoms are crossing a high-$Q$ superconducting cavity tuned to resonance with a millimeter-wave transition between adjacent Rydberg states.

Spontaneous atomic emission inside an electromagnetic cavity is expected to occur at a rate different from the same process in free space.$^{1-4}$ If the cavity is resonant with a transition between two atomic levels, the partial spontaneous emission rate associated with the transition is multiplied by $\eta_{cav} = 3Q\lambda^3/4\pi^2\nu$ where $Q$ is the cavity quality factor, $\nu$ its volume, and $\lambda$ the transition wavelength. This effect, first discussed in the context of radio frequencies by Purcell in 1946,$^1$ is due to the change of the number of radiator modes per unit volume and unit frequency induced by the presence of the cavity. It can equivalently be understood as resulting from the interaction between the atom and its electric images reflected in the cavity mirrors. This effect has never

FIG. 1. Experimental arrangement.
Inhibited Spontaneous Emission by a Rydberg Atom

Randall G. Hulet, Eric S. Hilfer, and Daniel Kleppner

Research Laboratory of Electronics and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 29 July 1985)

Spontaneous radiation by an atom in a Rydberg state has been inhibited by use of parallel conducting planes to eliminate the vacuum modes at the transition frequency. Spontaneous emission is observed to “turn off” abruptly at the cutoff frequency of the waveguidelike structure and the natural lifetime is measured to increase by a factor of at least 20.

Spontaneous emission is often regarded as an unavoidable consequence of the coupling between matter and space. However, as one of the authors has pointed out, by surrounding the atom with a cavity which has no modes at the transition frequency, spontaneous emission can be inhibited or “turned off.” Drexhage, in studies of fluorescence by dye molecules deposited on a dielectric film over a conducting plane, observed a decrease of up to 25% in the fluorescent decay rate due to cavitylike effects. Rydberg atoms provide the
Strong Purcell Effect for InAs Quantum Boxes in Three-Dimensional Solid-State Microcavities

Jean-Michel Gérard and Bruno Gayral

Vahala, Nature 424, 839
Micro-cavities in the 21st century

<table>
<thead>
<tr>
<th>Fabry–Perot</th>
<th>Whispering gallery</th>
<th>Photonic crystal</th>
</tr>
</thead>
<tbody>
<tr>
<td>High $Q$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$Q$: 2,000</td>
<td>$Q$: 12,000</td>
<td>$Q_{III-V}$: 7,000</td>
</tr>
<tr>
<td>$V$: 5 ($\lambda/n)^3$</td>
<td>$V$: 6 ($\lambda/n)^3$</td>
<td>$Q_{Poly}$: $1.3 \times 10^5$</td>
</tr>
<tr>
<td>$V$: 13,000</td>
<td>$V$: 1.2 ($\lambda/n)^3$</td>
<td></td>
</tr>
<tr>
<td>Ultrahigh $Q$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$F$: $4.8 \times 10^5$</td>
<td>$Q$: $8 \times 10^9$</td>
<td>$Q$: $10^8$</td>
</tr>
<tr>
<td>$V$: 1,690 $\mu$m$^3$</td>
<td>$V$: 3,000 $\mu$m$^3$</td>
<td></td>
</tr>
</tbody>
</table>

Vahala, Nature 424, 839

A cavity is a tool to increase light-matter interaction.
Radiation-matter interaction

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- Fluorescence lifetime measurements
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Spontaneous emission control
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- Microcavities

Optical antennas
- Radiation reaction in dipolar scatterers
- Decay rate engineering with optical antennas
Nanoparticles: resonators at optical frequencies

White light

nanoparticle

Scattered light

100 nm Au particle

"damping"

http://www.photonics.ethz.ch
Nanoparticles: resonators at optical frequencies

- Metal nano-particles show resonances in the visible

100 nm Au particle
Nanoparticles: resonators at optical frequencies

• Metal nano-particles show resonances in the visible

Lycurgus Cup (glass with metal nano-particles):
Green when front lit ➔
⇐ Red when back lit

How does that work?

100 nm Au particle

www.photonics.ethz.ch
The electrodynamic polarizability

\[ p_{\text{stat}} = \alpha_0 E_{\text{stat}}(r_0) \]

- Static polarizability: induced dipole moment due to static E-field
- For a sphere in vacuum:

\[ \alpha_0 = 4\pi \varepsilon_0 a^3 \frac{\varepsilon - 1}{\varepsilon + 2} \]
The electrodynamic polarizability

- $p_{\text{stat}} = \alpha_0 E_{\text{stat}}(r_0)$
- $p = \alpha_{\text{eff}} E_{\text{ext}}$
- $p = \alpha_0 \left[ E_{\text{ext}}(r_0) + \overrightarrow{G}(r_0, r_0)p \right]$
- $\alpha_{\text{eff}}^{-1} = \alpha_0^{-1} - \overrightarrow{G}(r_0, r_0)$
- $\overrightarrow{G} = \text{Re} \overrightarrow{G}_0 + \text{Re} \overrightarrow{G}_s + i \left[ \text{Im} \overrightarrow{G}_0 + \text{Im} \overrightarrow{G}_s \right]$

- Static polarizability: induced dipole moment due to static E-field
- Dynamic case: additional field generated by induced dipole moment
- Define effective electrodynamic polarizability “dressed” with Green function
- $\text{Re}G_0$ diverges at origin! Fact that we describe the scatterer as a mathematical point backfires. Choose to fit experimentally found resonance frequency.
- $\text{Re}G_s$ shifts resonance frequency depending on environment.
- $\text{Im}G$ represents radiation damping term: essential for energy conservation
The electrodynamic polarizability

\[ \alpha_{\text{eff}}^{-1} = \alpha_0^{-1} - i \text{Im} \mathcal{G}(r_0, r_0) \]

- This is a recipe to amend any electrostatic polarizability \( \alpha_0 \) with a radiation damping term to ensure energy conservation.
- Electrodynamic polarizability depends on position within photonic system.
- Radiation correction small for weak scatterers (small \( \alpha_0 \)).
- Radiation correction significant for strong scatterers (large \( \alpha_0 \)).
- Limit of maximally possible scattering strength.
The electrodynamic polarizability

\[ \alpha_{\text{eff}}^{-1} = \alpha_0^{-1} - i \text{Im} \hat{G}(r_0, r_0) \]

- Compare static and dynamic \( \alpha \)
- Static \( \alpha_0 \) may be huge, dynamic \( \alpha_{\text{eff}} \) is always bounded by inverse LDOS
- Radiation damping is a loss channel and dampens resonance
- Radiation damping is given by the LDOS at the scatterer’s position

Metallic particle (Drude model for \( \varepsilon \))

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