Single molecule controlled emission in planar plasmonic cavity

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Light/matter interaction at the nanoscale

\[ \frac{\lambda}{2} \sim 10^{-10} \text{cm}^2 \left( 10^4 \text{nm}^2 \right) \]

\[ \sigma \sim 10^{-15} \text{cm}^2 \]

(0.1 nm²)
Strategies (at ambient T°C)

Cavity quantum electrodynamics (cQED)

<table>
<thead>
<tr>
<th>Fabry-Perot</th>
<th>Whispering gallery</th>
<th>Photonic crystal</th>
</tr>
</thead>
<tbody>
<tr>
<td>Q: 2,000</td>
<td>V: 5 (μm)²</td>
<td></td>
</tr>
<tr>
<td>Q: 12,000</td>
<td>V: 6 (μm)²</td>
<td></td>
</tr>
<tr>
<td>Q: 7,000</td>
<td>V: 1,3 x 10⁶</td>
<td></td>
</tr>
<tr>
<td>Q: 13,000</td>
<td>V: 1,2 (μm)³</td>
<td></td>
</tr>
</tbody>
</table>

Surface enhanced spectroscopies (SERS, SEF)

Purcell factor

\[
 F_p = \frac{\Gamma}{n_1 \Gamma_{tot}} = \frac{3}{4 \pi^2} \left(\frac{\lambda}{n_1}\right)^3 \frac{Q}{V_{eff}}
\]

Interaction volume

\[
 V_{SPP} \sim \frac{4}{3} \pi R^3 \ll \left(\frac{\lambda}{n_1}\right)^3
\]

Optical microcavities (Review)

Resonance quality, radiative/ohmic losses and modal volume of Mie plasmons,
Derom et al, EPL 98, 47008 (2012)
Strategies (at ambient T°C)

Cavity quantum electrodynamics (cQED)

Surface enhanced spectroscopies (SERS, SEF)

Interaction volume

Interaction Duration (high Q)

Resonance quality, radiative/ohmic losses and modal volume of Mie plasmons, Derom et al., EPL 98, 47008 (2012)

Purcell factor

\[ F_p = \frac{\Gamma}{\Gamma_{tot}} = \left( \frac{\lambda}{n_1} \right)^3 \]

Optical microcavities (Review)

Low threshold/thresholdless laser

\[ \beta = \frac{\Gamma_{cav}}{\Gamma_{tot}} \approx \frac{F_p}{1 + F_p} \]

Fig. 15.14. Light-power-versus-current curves for single spatial-mode emission from (i) conventional laser, (ii) a high-\(\beta\)-factor laser, and (iii) a thresholdless laser. The conventional laser has a distinct current threshold. The high-\(\beta\)-factor laser has a less distinct threshold. It would be noticeable in the spectrum and device modulation speed, however. A hypothetical thresholdless laser would have a \(\beta\) close to 1, and would somehow suppress all other lossy emission until the carrier density required for gain (or at least transparency) was achieved.

A photonic crystal nanocavity with ultralow threshold
Nomura, Iwamoto, Arakawa, SPIE (2007)
Gain-assisted propagation

Gain-Assisted Propagation in a Plasmonic Waveguide at Telecom Wavelength
Grandidier et al, Nano Letters 9, 2935 (2009)
In-plane plasmonic cavity

Submicrometer In-Plane Integrated Plasmon Cavities


Gong et al, APL 94, 013106 (2009)

Mode confinement
Light extraction (LED, ...)

(a) Intensity vs. wavelength
(b) Pump, polarizer, objective, detector
(c) Beam Splitter, quartz, Au Si-NC

Q~50
Plasmonic Purcell factor

**Flat film**

\[ Q = \frac{k_{SPP}}{\Delta k_{SPP}} = k_{SPP} L_{SPP} \approx 100 \]

\[ V_{SPP} \approx \delta L_{SPP}^2 \approx 30 \left( \frac{\lambda}{n} \right)^3 \]

**Single mode cavity**

\[ V_{SPP} \approx \delta L_{SPP} L_{cav} \approx 0.5 \left( \frac{\lambda}{n} \right)^3 \]

\[ F_p \approx \frac{\Gamma_{SPP}}{n_1 \Gamma} = \frac{3}{4 \pi^2} \left( \frac{\lambda}{n_1} \right)^3 \frac{Q}{V_{SPP}} \approx 15 \]

see also
Coupling of a dipolar emitter into one-dimensional surface plasmon
Plasmonic Bragg mirror
Surface plasmon coupled emission near a Bragg mirror

Single-molecule controlled emission in planar plasmonic cavities

Mirror efficiency
Surface plasmon coupled emission near a Bragg mirror

Spatial coherence of the localized nanosource
Mirror bandgap

Large bandwidth system (675 nm < λ < 790 nm)
In plane SPP cavity

\[ \lambda_{\text{exc}} = 635 \text{ nm} \]

\[ \lambda_{\text{em}} = 670 \text{ nm} \]
In plane SPP cavity

emitted power
$P(k_y)$

decay rate
$\gamma \propto \int P(k_y) dk_y$

$L_{cav} = \lambda_{SPP}/2$
In plane SPP cavity
In plane SPP cavity

\[ L_{cav} = 3 \lambda_{SPP}/2 \]
Comparison between planar and bulk cavities

(a) 

(b)
Optical µcavity

Rate an efficiency of spontaneous emission in metal-clad µcavities Worthing et al., J. App. Phys. 89, 615 (2001)

See also The Single Molecule Probe: Nanoscale Vectorial Mapping of Photonic Mode Density in a Metal Nanocavity Hoogenboom et al., Nano Letters 9, 1189 (2009)
Plasmonic (planar) cavity

Weeber et al., Nano Let. 7, 1352 (2007)

\[ \lambda_0 = 690 \text{ nm} \]
\[ \lambda_{SPP} = 665 \text{ nm (n_{eff} = 1.035)} \]

Single-molecule controlled emission in planar plasmonic cavities
In plane SPP cavity

Decay rate depends on
- the cavity length
- the molecule position
- the molecule orientation
In plane SPP cavity

DiD molecule

$\lambda_{\text{emission}} = 670$ nm

Photon event

$\tau_{\text{flu}} = 21$ ns

$\tau_{\text{flu}} = 1.3$ ns
Lifetime measurement

137 molecules into the cavities

Reference:
75 molecules far from cavities

Bi exponential fitting
- short components (~10 ps)
  background signal (notably, gold photoluminescence)
- single molecule fluorescence lifetime (~ ns)
Polar representation

\[ u(\omega) = \frac{\int_0^T \cos(\omega t) I(t) dt}{\int_0^T I(t) dt} \]

\[ v(\omega) = \frac{\int_0^T \sin(\omega t) I(t) dt}{\int_0^T I(t) dt} \]

Generalization of the polar representation in time domain fluorescence lifetime imaging microscopy for biological applications: practical implementation
Effect of the cavity size

- strong dispersion at the single molecule level

- lifetime averaged over position for each cavity size: small variations related to coupling to the cavity modes
Molecule position

dispersion decay rate due to molecules orientation
Single cavity ($L_{cav} = 2 \lambda_{SPP} = 1,1\mu m$)

Extraction efficiency (leakage into the substrate)

$$\frac{\Gamma_{rad}}{\Gamma_{tot}} \approx 0.9$$
Summarize

**Plasmonic Bragg mirror**
- efficient reflexion of locally excited SPP over $\sim 40^\circ$
- large bandwidth
$\Rightarrow$ control emission at room temperature

**Planar plasmonic cavity**
- surface wave confinement
- planar analogous of bulk optical $\mu$ cavity
- $F_p \sim 7$ ($\beta \sim 85\%$)
- good extraction efficiency

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Single-molecule controlled emission in planar plasmonic cavities
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