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M2 Quantum, Light, Matter, Nanosciences

Fundamentals of Nanophotonics - Tutorial 3
Spontaneous Emission engineering

Rabi oscillations of a molecule coupled to a plasmonic nanoantenna

We consider a two–level molecule coupled to a single mode of a plasmonic cavity. The regime of coherent energy exchange (Rabi oscillations) requires that the light–matter coupling strength g dominates over cavity losses. A commonly used necessary condition for strong coupling is

$$2g > \kappa,$$

where κ is the cavity decay rate.

Light–matter coupling strength

The interaction Hamiltonian between a two–level emitter and the electric field of the cavity mode reads

$$\hat{H}_{\text{int}} = -\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}.$$

The single-photon coupling strength is defined by

$$\hbar g = dE,$$

where d is the transition dipole moment of the molecule and E is the electric field per photon of the cavity mode.

For a cavity mode of angular frequency ω, mode volume V, and relative permittivity ε_r, the electric field per photon is

$$E = \sqrt{\frac{\hbar\omega}{2\varepsilon_0\varepsilon_r V}}.$$

Thus,

$$g = \frac{d}{\hbar}\sqrt{\frac{\hbar\omega}{2\varepsilon_0\varepsilon_r V}} = d\sqrt{\frac{\omega}{2\hbar\varepsilon_0\varepsilon_r V}}.$$

Dipole moment and oscillator strength

The transition dipole moment can be expressed in terms of the oscillator strength f as

$$d^2 = \frac{f\hbar e^2}{2m\omega},$$

where e is the elementary charge and m the electron mass.

Substituting this expression into the formula for g^2, we obtain

$$g^2 = \frac{f\hbar e^2}{2m\omega} \cdot \frac{\omega}{2\hbar\varepsilon_0\varepsilon_r V} = \frac{fe^2}{4m\varepsilon_0\varepsilon_r V}.$$

Therefore,

$$g = \sqrt{\frac{fe^2}{4m\varepsilon_0\varepsilon_r V}}.$$

Ratio $2g/\kappa$

The cavity decay rate κ is related to the quality factor Q by

$$\kappa = \frac{\omega}{Q}.$$

Hence,

$$\frac{2g}{\kappa} = \frac{2Q}{\omega}\sqrt{\frac{fe^2}{4m\varepsilon_0\varepsilon_r V}}.$$

We now introduce the classical electron radius

$$r_e = \frac{e^2}{4\pi\varepsilon_0 mc^2},$$

which allows us to write

$$\frac{e^2}{m\varepsilon_0} = 4\pi r_e c^2.$$

Substituting this into the expression above gives

$$\frac{2g}{\kappa} = \frac{2Q}{\omega}\sqrt{\frac{f\pi r_e c^2}{\varepsilon_r V}}.$$

Using $\omega = 2\pi c/\lambda$, we finally obtain

$$\boxed{\frac{2g}{\kappa} = Q\sqrt{\frac{f\lambda^3}{V}\frac{r_e}{\pi\varepsilon_r\lambda}}}$$

which is the desired result.

Numerical estimate

We now evaluate this expression using the parameters :

$$\lambda = 600\ \text{nm}, \quad Q \sim 10, \quad V = 30\ \text{nm}^3, \quad \varepsilon_r = 2, \quad f \sim 1.$$

Using $r_e = 2.82 \times 10^{-15}$ m and converting all quantities to SI units, we estimate

$$\frac{\lambda^3}{V} = \frac{(600\ \text{nm})^3}{30\ \text{nm}^3} \approx 7.2 \times 10^6.$$

Furthermore,

$$\frac{r_e}{\pi\varepsilon_r\lambda} \approx \frac{2.82 \times 10^{-15}}{\pi \times 2 \times 600 \times 10^{-9}} \sim 7.5 \times 10^{-10}.$$

Combining all factors,

$$\frac{2g}{\kappa} \sim Q\sqrt{f \times 7.2 \times 10^{6} \times 7.5 \times 10^{-10}} \sim 10 \times \sqrt{5.4 \times 10^{-3}} \sim \mathcal{O}(1).$$

For a plasmonic nanoantenna with nanometric mode volume and moderate quality factor, the condition

$$2g > \kappa$$

can be satisfied. This demonstrates that **strong coupling and Rabi oscillations are achievable for a single molecule coupled to a plasmonic cavity**, despite the relatively low Q of plasmonic resonators.