1 Optical Interactions in the Context of Nano Optics : Near-field optics

- We consider a dipole in front of a perfectly conductive interface. The boundary value problem can be solve using image charges. In other words, the charges of the dipole induce opposite charges in the perfect conductor as shown in the figure below.

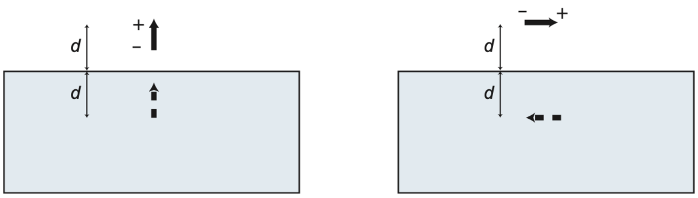


Figure 1: Dipoles near a perfect mirror with orientation perpendicular and parallel to the interface. The dashed arrows represent the image dipoles.

The modification of the emission rate can be easily obtained by calculating the power radiated by the real and image dipoles P, normalized by the power emitted by the real dipole in free space P_0, using the fact that $\Gamma/\Gamma_0 = P/P_0$ (see lecture notes). The results depends on the dipole orientation and it reads

$$\frac{\Gamma_\perp}{\Gamma_0} = 1 - 3\mathrm{Im}\left[\left(\frac{1}{(2kz)^3} - \frac{i}{(2kz)^2}\right)\exp(2ikz)\right], \tag{1}$$

$$\frac{\Gamma_\parallel}{\Gamma_0} = 1 + \frac{3}{2}\mathrm{Im}\left[\left(\frac{1}{(2kz)^3} - \frac{i}{(2kz)^2} + \frac{1}{(2kz)}\right)\exp(2ikz)\right], \tag{2}$$

where $\perp$ and $\parallel$ refer to the dipole orientations with respect to the interface, $k = \omega/c$ is the wave-vector and z is the distance of the real dipole from the perfect conductor.

When $z \to 0$, $\Gamma_\perp/\Gamma_0 \to 2$, because the two dipoles oscillate in phase, whereas $\Gamma_\parallel/\Gamma_0 \to 0$, because the two dipoles oscillate in opposite phase. The term $\exp(2ikz)$ is the interference between the two dipoles, which depends on $2z$, i.e. the distance between real and image charges. When $z \to \infty$, the decay rates approach the value Γ_0, but the perpendicular dipole approaches it faster, since it does not contain the far-field term $1/(2kz)$.

- In free-space, the partial local density of states ρ_μ is identical to the total density of states ρ. To show this, prove that

$$\left[n_\mu . \mathrm{Im}\left\{\overleftrightarrow{G}_0\right\} . n_\mu\right] = \frac{1}{3}\,\mathrm{Im}\left\{\mathrm{Tr}\left[\overleftrightarrow{G}_0\right]\right\}, \tag{3}$$

where $\overleftrightarrow{G}_0$ is the free-space dyadic.

In this problem we would like to prove

$$\left[\mathbf{n}_\mu \cdot \mathrm{Im}\{\overleftrightarrow{G}_0\} \cdot \mathbf{n}_\mu\right] = \frac{1}{3}\mathrm{Im}\{\mathrm{Tr}\left[\overleftrightarrow{G}_0\right]\}.$$

The free space dyadic Green function is given by

$$\overleftrightarrow{G}(R) = \left[(\overleftrightarrow{I} - \hat{R}\hat{R}) + \frac{i}{kR}(\overleftrightarrow{I} - 3\hat{R}\hat{R}) - \frac{1}{k^2R^2}(\overleftrightarrow{I} - 3\hat{R}\hat{R})\right] G_0,$$

with

$$G_0 = \frac{e^{-ikr}}{4\pi r}.$$

By using

$$\mathrm{Tr}(\overleftrightarrow{I}) = 3 \quad , \quad \mathrm{Tr}(\hat{R}\hat{R}) = 1$$

$$\mathbf{n}_\mu \overleftrightarrow{I} \mathbf{n}_\mu = 1 \quad , \quad \mathbf{n}_\mu(\hat{R}\hat{R})\mathbf{n}_\mu = 1/3$$

Now we have

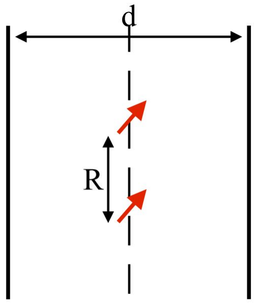
$$\mathrm{Tr}(\overleftrightarrow{G_0}) = \left[3 - 1 + \frac{i}{kr}(3-3) - \frac{1}{k^2r^2}(3-3)\right] G_0 = 2G_0,$$

$$\mathbf{n}_\mu \cdot \overleftrightarrow{G_0} \cdot \mathbf{n}_\mu = \left[(1 - \frac{1}{3}) + \frac{i}{kr}(1-1) - \frac{1}{k^2r^2}(1-1)\right] G_0 = \frac{2}{3}G_0.$$

$$\left.\begin{array}{l} \mathrm{Im}\left[\mathrm{Tr}(\overleftrightarrow{G_0})\right] = 2\mathrm{Im}(G_0) \\ \mathrm{Im}(\mathbf{n}_\mu \cdot \overleftrightarrow{G_0} \cdot \mathbf{n}_\mu) = (2/3)G_0 \end{array}\right\} \left[\mathbf{n}_\mu \cdot \mathrm{Im}\{\overleftrightarrow{G_0}\} \cdot \mathbf{n}_\mu\right] = \frac{1}{3}\mathrm{Im}\{\mathrm{Tr}\left[\overleftrightarrow{G_0}\right]\}.$$

- Two molecules, fluorescein (donor) and alexa green 532 (acceptor), are located in a plane centered between two perfectly conducting surfaces separated by the distance d. The emission spectrum of the donor (f_D) and the absorption spectrum of the acceptor (σ_A) are approximated by a superposition of two Gaussian distribution functions. Use the fit parameters from Section 8.6.2 in the text book Principles of Nano-Optics (Second edition) by Lukas Novotny.
  1. Determine the Green's function for this configuration.
  2. Calculate the decay rate γ_0 of the donor in the absence of the acceptor.
  3. Determine the transfer rate $\gamma_{D\to A}$ as a function of the separation R between donor and acceptor. Assume random dipole orientations.
  4. Plot the Förster radius R_0 as a function of the separation d.

Two molecules, fluorescein (donor) and alexa green 532 (acceptor), are located in a plane centered between two perfectly conducting surfaces separated by the distance d (see the figure).



1. To find the Green's function one can use angular spectrum representation for a dipole.

$$\frac{e^{ikr}}{r} = \frac{i}{2\pi}\iint\limits_{-\infty}^{\infty} \frac{e^{ik_x x + ik_y y + ik_z z}}{k_z} \mathrm{d}k_x \, \mathrm{d}k_y,$$

$$G_0(r, r') = \frac{e^{ik|r-r'|}}{4\pi|r-r'|},$$

$$\overleftrightarrow{G}(r, r') = \left[\mathbb{1} + \frac{1}{k^2}\nabla\nabla\right] G_0.$$

Then

$$G_0(r, r') = \frac{i}{8\pi^2}\int \frac{e^{ik_x x + ik_y y + ik_z z}}{k_z} \mathrm{d}k_x \, \mathrm{d}k_y,$$

where $k_z d = \pi n$ and as a result $k_z = \pi n/d$. We also have

$$\omega = kc = \sqrt{k_x^2 + k_y^2 + k_z^2}c = \sqrt{k_\parallel^2 + \frac{\pi^2 n^2}{d^2}},$$

$$G_0(r, r') = \sum_{n=1}^{\infty} \frac{1}{8\pi^2}\int \frac{e^{ik_x x + ik_y y}}{\frac{\pi n}{d}} \left[2i \sin\left(\frac{\pi n}{d} z\right)\right] \mathrm{d}k_x \, \mathrm{d}k_y,$$

where $e^{ik_z z} + e^{-ik_z z} = 2i \sin k_z z$.

2. The decay rate γ_0 of the donor in the absence of the acceptor is related to

$$\gamma \propto \mathrm{Im}[\mathrm{Tr}\, \overleftrightarrow{G}] = \mathrm{Im}[G_{xx} + G_{yy} + G_{zz}] = \mathrm{Im}[3G_0 + \frac{1}{k^2}\nabla^2 G_0] = \mathrm{Im}[2G_0],$$

where

$$\nabla^2 G_0 = -\frac{1}{4\pi^2}\sum_{n=1}^{\infty}\int \frac{d}{\pi n}\left[-(k_x^2 + k_y^2 + \frac{\pi^2 n^2}{d^2})\right] e^{ik_x x + ik_y y} \sin\left(\frac{\pi n z}{d}\right) \mathrm{d}k_x \, \mathrm{d}k_y = -\frac{\omega^2}{c^2} G_0,$$

$$G_0(r, r') = \sum_{n=1}^{\infty} \frac{1}{4\pi^2}\int \frac{\cos(k_x x + k_y y) + i \sin(k_x x + k_y y)}{\frac{\pi n}{d}} \left[2i \sin\left(\frac{\pi n}{d} z\right)\right] \mathrm{d}k_x \, \mathrm{d}k_y,$$

where

$$k_{\parallel}^2 = k_x^2 + k_y^2,$$
$$\mathrm{d}k_x\, \mathrm{d}k_y = k_{\parallel}\, \mathrm{d}\Phi\, \mathrm{d}k_{\parallel},$$
$$i(k_x x + k_y y) = ik_{\parallel}\rho.$$

Thus we have

$$G_0(r,r') = \frac{2\pi}{4\pi^2}\sum_{n=1}^{\infty}\int k_{\parallel}\frac{\cos\big(k_{\parallel}\rho\big) + i\sin\big(k_{\parallel}\rho\big)}{\frac{\pi n}{d}}\left[\sin\Big(\frac{\pi n}{d}z\Big)\right]\mathrm{d}k_{\parallel},$$

$$\mathrm{Im}[2G_0] = -\frac{1}{\pi}\sum_{n=1}^{\infty}\left[\int_0^{\sqrt{\frac{\omega^2}{c^2}-\frac{n^2\pi^2}{d^2}}} k_{\parallel}\frac{\cos\big(k_{\parallel}\rho\big)\sin\big(\frac{n\pi z}{d}\big)}{\frac{n\pi}{d}}\mathrm{d}k_{\parallel} + \int_{\sqrt{\frac{\omega^2}{c^2}-\frac{n^2\pi^2}{d^2}}}^{\infty}\mathrm{d}k_{\parallel}\cdots\right]$$

Since $\sin(\pi nz/d)$ for $z=0$, it makes the second integral zero. At the end we have

$$\gamma \propto \frac{1}{\pi}\sum\int_0^{\sqrt{\frac{\omega^2}{c^2}-\frac{n^2\pi^2}{d^2}}}\frac{k_{\parallel}}{\frac{n\pi}{d}}\sin^2(\frac{n\pi z}{d})\mathrm{d}k_{\parallel},$$

which contains only the terms above cutoff (the decay rate is determined by the cavity modes).

3. The transfer rate $\gamma_{D\to A}$ as a function of the separation R between donor and acceptor is

$$\frac{\gamma_{D\to A}}{\gamma_0} = \frac{1}{R^6}\int_0^{\infty}\frac{f_0(\omega)\sigma(\omega)}{n^4(\omega)\omega^4}T(\omega)\mathrm{d}\omega,$$

where

$$T(\omega) = 16\pi^2 k^4 R^6\left|n_A\cdot\overleftrightarrow{G}\cdot n_D\right|^2.$$

Since the absolute value for random orientation is equal to $\mathrm{Tr}[\overleftrightarrow{G}]$, as a result we have

$$T(\omega) = k^4 R^6\left|\sum_n\int k_{\parallel}\frac{e^{ik_{\parallel}\rho}\sin\big(\frac{n\pi z}{d}\big)}{\frac{n\pi}{d}}\mathrm{d}k_{\parallel}\right|^2.$$

We remark that here the integration includes both near (evanescent) and far field contributions of $k_{\parallel}$.

4. The Förster radius R_0 is related to transfer rate

$$\frac{\gamma_{D\to A}}{\gamma_0} = \left[\frac{R_0}{R}\right]^6,$$

$$R_0^6 \approx R^6\left|\sum_n\int k_{\parallel}\frac{e^{ik_{\parallel}\rho}\sin\big(\frac{n\pi z}{d}\big)}{\frac{n\pi}{d}}\mathrm{d}k_{\parallel}\right|^2.$$

References

Principles of Nano-Optics (Second edition) by Lukas Novotny and Bert Hecht
Molecular scattering and fluorescence in strongly confined optical fields by Mario Agio.