

## Nanophotonics Local Density of Optical States (LDOS) Assignment

The TA for this assignment is Robb Walters <walters@amolf.nl>. Please ask questions in English.

### 1. Dipole in front of a mirror

In 1966, Drexhage was the first to show that the spontaneous emission lifetime of atoms changes near a mirror due to the local density of states (LDOS). In this exercise we will do a classical calculation to explain his results.

Let us first consider an electrostatic problem: a single charge is positioned a distance  $d$  from a perfectly conducting surface (the mirror). It is well known that the electric field in the rightmost half space is the sum of that of the charge and that of an *image charge*, of opposite sign, located a distance  $d$  behind the mirror.

**1A.** Draw a diagram of the *image dipole* generated by a dipole in front of a mirror for a dipole oriented perpendicular to the mirror. Suppose  $d$  is small. What is the effective dipole moment?

**1B.** Now we consider an oscillating dipole in a uniform medium that does not contain a mirror. To calculate how much power the dipole radiates you must integrate the Poynting vector over a surface enclosing the dipole. The formulas for the electric and magnetic fields of a dipole are:

$$\vec{E} = \frac{1}{4\pi\epsilon_0} \left\{ \frac{\omega^2}{c^2 r} \hat{r} \times \vec{p} \times \hat{r} + \left( \frac{1}{r^3} - \frac{ik}{r^2} \right) [3\hat{r}(\hat{r} \cdot \vec{p}) - \vec{p}] \right\} e^{i(kr - \omega t)} \quad (1)$$

$$\vec{B} = \frac{1}{4\pi\epsilon_0} \frac{\omega^2}{c^3} \hat{r} \times \vec{p} \left( \frac{1}{r} + \frac{i}{kr^2} \right) e^{i(kr - \omega t)} \quad (2)$$

In the “far-field” region ( $r \gg R_0$ ), terms that decay as  $r^{-1}$  dominate. Find an expression for  $R_0(k)$  and give a typical value in nm. If we choose the  $z$  axis to be aligned with the dipole moment  $\vec{p}$ , then the quantity  $\hat{r} \times \vec{p}$  reduces to  $p \sin(\theta) \hat{\theta}$  and  $\hat{r} \times \vec{p} \times \hat{r}$  reduces to  $p \sin(\theta) \hat{\phi}$ . Write down simplified expressions for  $\vec{E}$  and  $\vec{B}$  in the far-field when the dipole is aligned with the  $z$  axis. Recall that  $\mu_0 \epsilon_0 = 1/c^2$  and show that the time-averaged Poynting vector  $\langle \vec{S} \rangle$  is:

$$\langle \vec{S} \rangle = \frac{1}{2\mu_0} \vec{E} \times \vec{B}^* = \frac{p^2 \omega^4}{(4\pi\epsilon_0) \cdot 8\pi \cdot c^3} \frac{\sin^2(\theta)}{r^2} \hat{r} \quad (3)$$

The radiated power is the integrated Poynting vector flux through the surface of an enclosing volume. The symmetry of the problem suggests we integrate over a sphere:

$$P_{\text{free}} = \oint_{\text{surface}} \langle \vec{S} \rangle \cdot \hat{n} = \int_0^{2\pi} d\phi \int_0^\pi d\theta r^2 \sin(\theta) \langle \vec{S} \rangle \quad (4)$$

Calculate  $P_{\text{free}}$ . What is the order of magnitude of  $P_{\text{free}}$  in watts for a typical molecular-scale dipole emitting in the visible? How do you interpret this result?

**1C.** Calculate the radiated power in case of a vertically oriented dipole on the  $z$ -axis in front of a horizontal mirror that is located in the  $xy$  plane. Refer to the diagram for the geometry. The angle  $\varphi$  is in the plane of the mirror.

Here are some step-by-step hints:

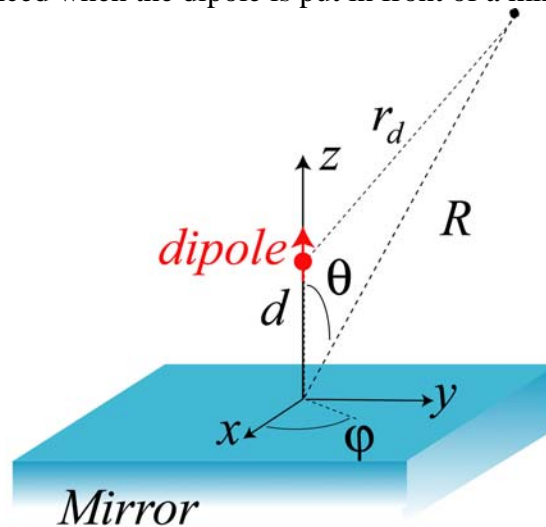
a. Integrate over a spherical surface of radius  $R$  as in the diagram. Do you need a full sphere?

b. Prove: if  $R \gg d$  then:  $r_d \approx R \left( 1 - \frac{d}{R} \cos(\theta) \right)$

c. You can assume that  $r_d=R$  is sufficiently precise for the denominator in the electric field (see Eq. (1)) but that you need hint b in the complex phase factors.

d. Mathematica knows how to do  $\int_0^\pi d\theta \sin^m(\theta) \cos^n(\alpha \cos(\theta))$  numerically using

`NIntegrate`. Plot your result normalized to  $P_{\text{free}}$ . This ratio tells you by how much the emitted power of a dipole antenna that is driven by a given current is enhanced when the dipole is put in front of a mirror.



Your result for the increase in emitted power of a classical point dipole is *equivalent* to the enhancement of the spontaneous emission decay rate of a quantum dipole: an atom in front of a mirror, i.e. to the LDOS enhancement relative to vacuum in vicinity of a mirror. It tells you that depending on geometry, an atom can decay faster or slower due to constructive or destructive interference with its own mirror image.

**1D.** Interpret the result for  $d=0$  and interpret the period of the oscillations. Why do the oscillations die out for large  $d$ ? (If you didn't manage 1C: try to guess the value at  $d=0$  and at infinite  $d$  and explain your guesses).

**1E.** Draw an image dipole diagram for a dipole in front of a mirror that is oriented along the mirror surface. Do you expect the LDOS near the mirror to be enhanced or reduced for this dipole orientation? Sketch the LDOS for this dipole orientation.

Note if you are curious: you can calculate the LDOS for this case as in 1C. Put the dipole on the y-axis, pointing along z, and place the mirror in the xz plane. You can not get rid of the integral over  $\varphi$  by hand, so you will need Mathematica.

## 2. Drexhage's Experiment

Drexhage performed his experiment by measuring the photoluminescence decay rate of Europium ions in front of a silver mirror. You will now analyze similar data from a more recent experiment reported by Amos and Barnes in Phys. Rev. B in 1997. You may download a copy of this paper from <http://www.erbium.nl/nanophotonics/>. We will look carefully at the lifetime data shown in Figure 2 of this work, reproduced below. Note that the authors estimate the uncertainty in their lifetime measurements at ~1%. The dipole moment of the Europium ions is not aligned in any specific orientation relative to the mirror, so we will compare the experimental data to calculations averaged over an isotropic distribution of dipoles (below right).

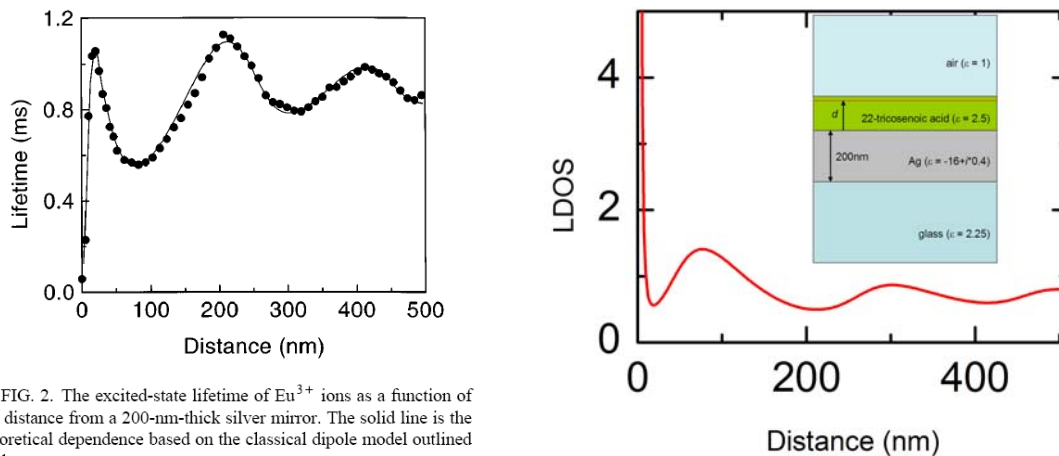


FIG. 2. The excited-state lifetime of  $\text{Eu}^{3+}$  ions as a function of the distance from a 200-nm-thick silver mirror. The solid line is the theoretical dependence based on the classical dipole model outlined in the text.

**2A.** Estimate the wavelength of emission of Europium based on the oscillation period of the data in Figure 2. The ions are embedded in a medium of index  $n \sim 1.58$ . Please explain how you make your estimate.

The europium ions do not only decay by spontaneous emission, but also by non-radiative processes that are not altered by the LDOS. The total decay rate measured in the experiment is the sum of rates of both processes:

$$\frac{1}{\tau} = \Gamma_{\text{total}} = \Gamma_{\text{rad}} + \Gamma_{\text{nonrad}}$$

Write down the formula for calculating the total emission rate ( $1/\tau$ ) as a function of the LDOS and  $\Gamma_0$ , the decay rate in vacuum. (The LDOS data provided have been normalized to the LDOS in a uniform medium with  $\epsilon=2.5$ .)

**2B.** Data extracted from Figure 2 and a calculation of the exact LDOS factor for the experimental geometry are available online at <http://www.erbium.nl/nanophotonics/>. Using the equation you have determined in 2A and the least-squares curve-fitting program of your choice, estimate values and standard deviations for  $\Gamma_{\text{nonrad}}$  and  $\Gamma_0$  by comparing theory and experiment. Calculate the reduced  $\chi^2$  statistic and interpret the result.

*Hint:*

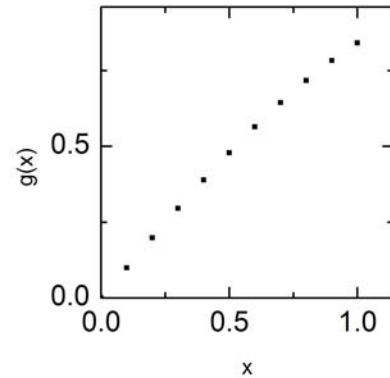
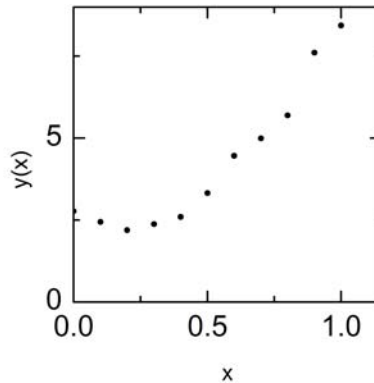
You may be more familiar with fitting data with functions rather than tabulated data. Here is a small exercise to help you with 2B above.

Consider a function:

$$y(x) = A \cdot (g(x))^3 + B \quad (5)$$

And the tabulated data and plots of  $y(x)$  and  $g(x)$ :

x	y(x)	g(x)
0	0	2
0.1	0.09983	2.00995
0.2	0.19867	2.07841
0.3	0.29552	2.25808
0.4	0.38942	2.59054
0.5	0.47943	3.10195
0.6	0.56464	3.8002
0.7	0.64422	4.67361
0.8	0.71736	5.69151
0.9	0.78333	6.8065
1	0.84147	7.95823



What you need to do is first plot  $y(x)$  vs.  $g(x)$  and then fit these points to the equation. Often you will find that you have calculated data for  $g(x)$  over a set of  $x$  values that do not exactly match the experiment's  $x$  values. In this case you must first interpolate values for  $g(x)$  for the  $x$  positions in the experiment. I have made this interpolation for you for this assignment.

