

Nanophotonics Local Density of Optical States (LDOS) Assignment

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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. We will demonstrate that the radiated power of a dipole in front of a mirror is enhanced or reduced, depending on the distance from the mirror, by resp. constructive and destructive interference of the field of the dipole with that of its mirror image.

1A. First 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 $-p \sin(\theta) \hat{\phi}$ and $\hat{r} \times \vec{p} \times \hat{r}$ reduces to $-p \sin(\theta) \hat{\theta}$. Show that this is true by evaluating the cross products. 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:

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

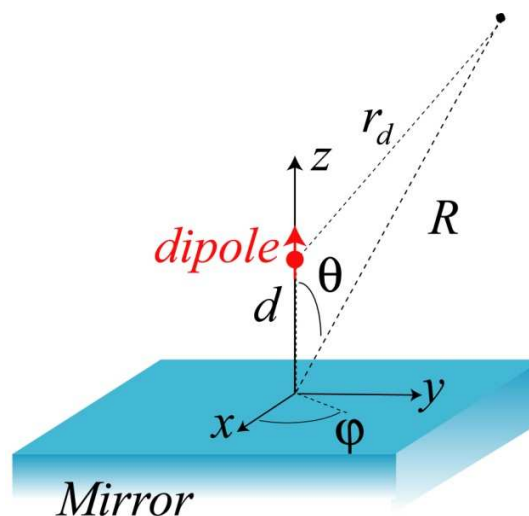
Calculate P_{free} . What is the order of magnitude of P_{free} in watts for a typical molecular-scale dipole emitting in the visible? How do you interpret this result?

Let us now 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.

1B. 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, expressed in terms of the distance d and the dipole moment \vec{p} of the real dipole?

1C. Do the same for an image dipole generated by a dipole in front of a mirror for a dipole oriented parallel to the mirror. Suppose d is small. What is the effective dipole moment in this case?



1D. 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 ϕ is in the plane of the mirror.

Here are some step-by-step hints:

- Integrate over a spherical surface of radius R as in the diagram. Do you need a full sphere ?
- Prove that: if $R \gg d : r_d \approx R(1 - \frac{d}{R} \cos \theta)$
- 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.
- 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_{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.

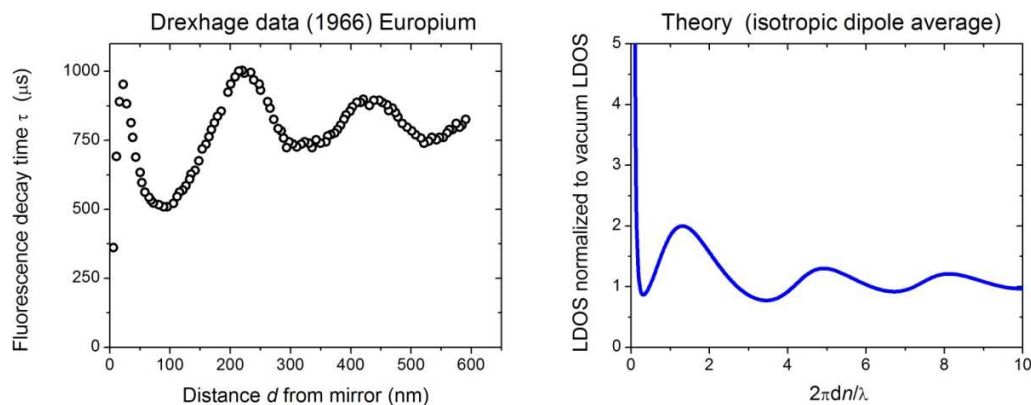
Your result for the change in emitted power of a classical point dipole is *equivalent* to the change of the spontaneous emission decay rate of a quantum dipole: an atom in front of a mirror, i.e. to the LDOS change relative to vacuum in vicinity of a mirror. It tells you that depending on geometry,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

1E. Consider the diagram you drew in 1C: do you expect the LDOS to be enhanced or reduced for the horizontally oriented dipole for small d ? Explain why. Sketch the radiated power versus distance.

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

2. Drexhage's Experiment

Drexhage performed his experiment by measuring the photoluminescence decay rate of Europium ions in front of a silver mirror. Below a graph is shown with Drexhage's data points and a plot of the average LDOS for randomly oriented dipoles in a medium with $n=1.5$ above a silver mirror. 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.



2A. Estimate the wavelength of emission of Europium based on the oscillation period of the data in the figure above. The ions are embedded in a medium of index $n \sim 1.5$. 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 the normalized LDOS (normalized with respect to the LDOS in vacuum) and Γ_0 , the decay rate in vacuum. Use Fermi's golden rule in your explanation.

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 Γ_{nonrad} and Γ_0 by comparing theory and experiment. Calculate the reduced χ^2 statistic and interpret the result.

Hint:

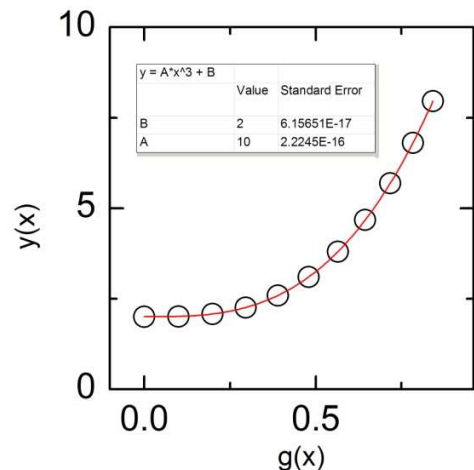
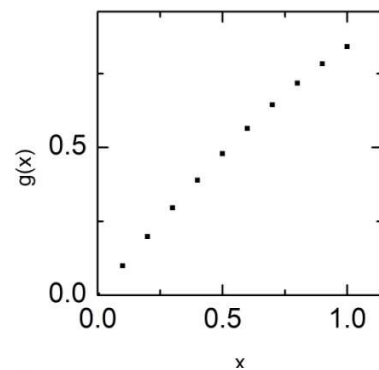
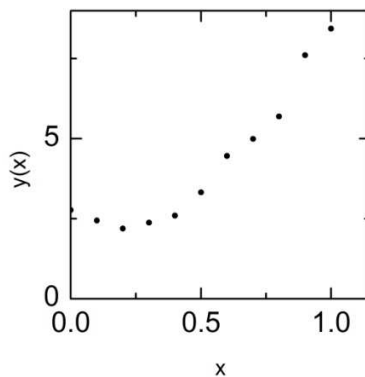
You may be more familiar with fitting data with functions rather than tabulated data. Here is a small exercise to help you with 2B above.

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.

2C. The classical calculations of exercise 1 should match the experimental results of Drexhage. In exercise 1 you only calculated the power for a vertically oriented dipole, whereas the measurements of Drexhage were done on a isotropic dipole average.

To be able to compare his measurements with theory, Drexhage in addition calculated the intensity for a horizontal dipole. He then averaged over the 2 dipole orientations, and corrected for the non radiative decay term. His calculation together with the measurement are shown below.

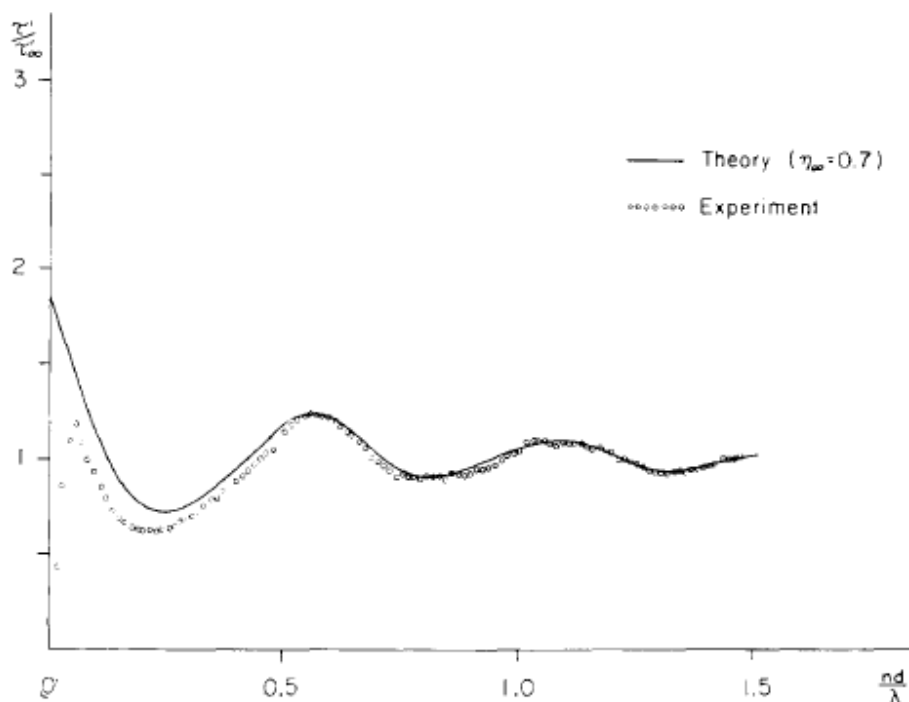


Fig. 5. Comparison of theory and experiment for decay time of Eu^{3+} in front of silver mirror.

There is a small mismatch between theory and the measurement. Give a possible explanation for this mismatch.