Homework 4

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Nano Optics, Fall Semester 2018
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Having familiarized ourselves with the concept of the local density of optical states and its repercussions for spontaneous emission, the first problem of this homework illustrates the impact of the LDOS onto the optical response of a passive dipolar scatterer. In particular, we investigate the effects of radiation damping on a simple optical antenna. In the second problem, we turn to optical forces, yet another manifestation of light-matter interactions. Exploiting our knowledge about focussing of fields gained in the first part of the lecture, we consider optical trapping in a Gaussian focus.

Please stick to the following points:

- Hand in your solution on time (beginning of lecture) in legible hand-written form or typeset using LaTeX (do not provide any code of any programming language, do not send your solutions by email).
- Keep your solutions in the order of the problem set.
- Show explicitly how you arrived at your answer and comment on your thoughts. Focus on the presentation of your solutions as if you were to explain them to a reader who is not familiar with the topic. Be brief but to the point.
- If you decide to generate graphs with a computer (which is encouraged), please position them at the appropriate position within your solution.
- Always label graphs and axes and provide units where appropriate.
- You can receive a maximum of 100 points for the correct solutions to the problems. For the quality of your presentation, you can receive an additional 10 points.

You are strongly encouraged to discuss the problems with your peers. In case of remaining questions, feel free to approach the course instructors.

1 Optical antennas and the electrodynamic polarizability [60 pts.]

Optical antennas typically consist of scatterers with dimensions much smaller than the wavelength, whose response to electromagnetic fields can be treated in the dipolar approximation. The dipole moment $\boldsymbol{p}$ acquired by such a scatterer in response to an electric field $\boldsymbol{E}$ is then given as $\boldsymbol{p} = \overleftrightarrow{\boldsymbol{\alpha}}\ \boldsymbol{E}$. For simplicity we restrict ourselves to isotropic scatterers, such that the tensor $\overleftrightarrow{\boldsymbol{\alpha}}$ reduces to the scalar α. The polarizability determines how a particle scatters, absorbs, and extincts electromagnetic radiation. Due to energy conservation, the extinct power must equal the sum of absorbed and scattered power. Throughout the following, consider vacuum as a background medium. Be very careful with textbooks treating this topic. Most of them do not take radiation damping effects into account, which are essential to correctly describe strong scatterers.

(a) (5 pts.) The scattering cross section σ_{scat} is defined as the ratio of power scattered by a particle, normalized to the power flux per unit area of the impinging plane wave. Derive the scattering cross section of a particle with polarizability α. Besides α, your expression should contain only the angular frequency ω, the speed of light in vacuum c, the vacuum permittivity ε_0 and numerical factors.

(b) (5 pts.) The extinction cross section σ_{ext} is defined as the ratio of power extinct from the incoming field, normalized to the power flux per unit area of the impinging plane wave. Derive the extinction cross section of a particle with polarizability α. To get the extinct power, consider the work per unit time done by the driving field on the induced dipole moment. Besides α, your expression should contain only the angular frequency ω, the speed of light in vacuum c, and the vacuum permittivity ε_0.

(c) (4 pts.) Show that energy conservation requires $\operatorname{Im}\alpha \geq A\ |\alpha|^2$ with $A > 0$. Determine A and show that it is proportional to the imaginary part of the free space Green's function evaluated at the origin, which is nothing else but the LDOS.

We have just derived an inequality that any electrodynamic polarizability α needs to fulfill in order to not violate energy conservation. A common method to obtain the polarizability of a small particle is to consider the *static* dipole moment $\boldsymbol{p}_s$ a small particle with radius a of a material with dielectric constant ε acquires in vacuum when exposed to a *static* (i.e. $\omega = 0$) electric field $\boldsymbol{E}_s$, which reads $\boldsymbol{p}_s = \alpha_0 \boldsymbol{E}_s$ with $\alpha_0 = 4\pi\varepsilon_0 a^3 \frac{\varepsilon-1}{\varepsilon+2}$. One then commonly introduces the dielectric constant $\varepsilon(\omega)$ at optical frequencies into the expression for α_0.

(d) (5 pts.) Water has a refractive index around $n(\omega) = 1.4$ throughout the optical spectrum. Plot the scattering cross section (calculated from the electrostatic expression for the polarizability) of a very small water droplet of diameter 5 nm as a function of frequency throughout the visible part of the spectrum. Do your findings offer a reason why the sky appears blue?

(e) (3 pts.) Calculate the extinction cross section of the water droplet (using the electrostatic expression for the polarizability). Comment on your result. What does that mean for the validity of α_0?

A recipe to obtain a well defined *electrodynamic* polarizability is to add a radiation correction according to $\alpha^{-1} = \alpha_0^{-1} - \mathrm{i}\,\text{LDOS}$. This correction adds a radiation damping term LDOS that (besides numerical factors) equals the local density of optical states.

(f) (7 pts.) Sketch the derivation of the above correction. To this end, consider that the particle is subjected to its own scattered field. Point out the role of the real part of the Green function. Assume an isotropic environment, so you can neglect the tensorial nature of the Green function. Determine the factor LDOS as a function of the Green function. Formulate the factor LDOS for a particle in vacuum using solely k and constants.

(g) (4 pts.) Apply the correction recipe to the static polarizability of the water droplet of problem (d). Check that the dynamic polarizability fulfills the relation found in (c). What is the relation between A and LDOS?

(h) (4 pts.) The correction formula establishes an upper bound for $\mathrm{Im}\,\alpha$, which can never exceed LDOS^{-1}. This limit is sometimes called the unitary limit, since here the ratio $\sigma_{\text{scat}}/\sigma_{\text{ext}}$ is unity. Energy conservation forbids that the polarizability of any dipolar scatterer exceeds the unitary limit! Show that the extinction cross section of a dipolar scatterer at the unitary limit is $\sigma_{\text{ext}}^{UL} = \frac{3}{2\pi}\lambda^2$.
Comment: Make it clear to yourself that you just found that the *optical* size of a dipolar scatterer, given by σ_{ext}, can largely exceed its *geometrical* size, and that the optical size at the unitary limit essentially corresponds to the diffraction limit! Accordingly, sub-wavelength nanoparticles of noble metals, with material (plasmonic) resonances, which can be very strong scatterers at the unitary limit, have an optical interaction cross section largely exceeding their geometrical size.

So far, we have considered a small dielectric particle without any intrinsic resonance, which is a weak scatterer. Subwavelength metallic particles are strong scatterers in the visible thanks to resonances of their free electrons. Such metal particles are therefore often called plasmonic scatterers. The dielectric function of a metal can be described by the Drude theory as $\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega+\mathrm{i}\gamma)}$ with the plasma frequency ω_p and the Ohmic loss rate γ.

(i) (7 pts.) Calculate $\alpha_0(\omega)$ for a metallic particle. Introduce the resonance frequency $\omega_0 = \omega_p/\sqrt{3}$. Plot both real and imaginary part of the optical volume in μm^3, i.e. the normalized polarizability $\alpha_0/(4\pi\varepsilon_0)$, as a function of wavelength for a particle of radius 30 nm and a resonance in the orange part of the spectrum at 550 nm. A plot range of 500 to 600 nm should be suitable. Assume a material damping rate $\gamma = 6.8 \cdot 10^{13}\ \text{s}^{-1}$. Add the unitary limit, i.e., the maximally possible value for $\mathrm{Im}\,\alpha$ to your plot. What do you observe?
Hint: It will be helpful to bring the polarizability into the form

$$\alpha_0 \propto \frac{1}{\omega_0^2 - \omega^2 - \mathrm{i}\gamma\omega}. \tag{1}$$

(j) (3 pts.) Apply the radiation correction to the polarizability of the metallic particle to obtain an electrodynamically valid α. Show that the radiation loss adds to the material loss and convince yourself, that the radiation loss is proportional to the LDOS at the scatterer's position.

(k) (5 pts.) To visualize the impact of the radiation correction, plot $\mathrm{Im}\,\alpha$ and $\mathrm{Im}\,\alpha_0$ for particles of 10, 30 and 50 nm radius. For which particle size does radiation damping play a role and why?

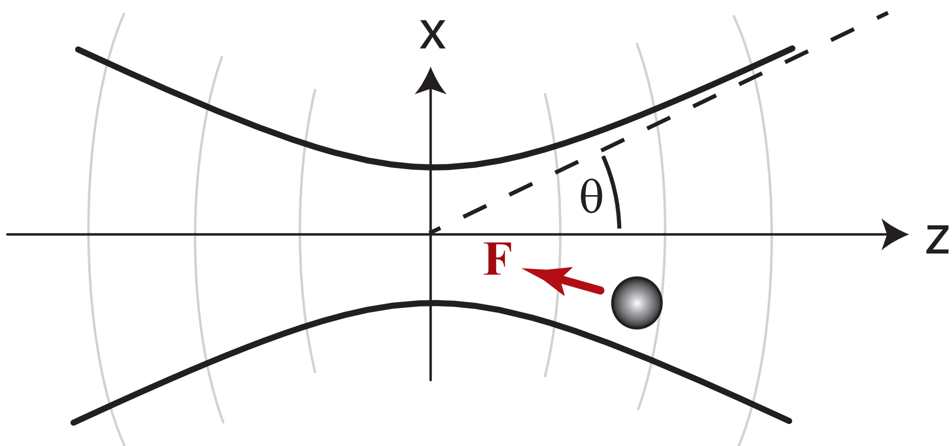
(l) (5 pts.) Calculate the maximum extinction cross section of the 50 nm radius particle. Compare this *optical* size of the particle to its *geometric* size and to the diffraction limit at the resonance frequency. What does this mean for the measurement of the particle size in an optical microscope? To this end, think about imaging a sub-wavelength sized glass particle and a gold particle in an optical microscope. In which sense does the image of the gold particle (not) differ from that of the glass particle?

(m) (3 pts.) Consider the case of no material loss. Show that the relation derived in (c) is fulfilled. At which frequency is it fulfilled with the equality sign?

2 Optical tweezers [40 pts.]

In 1971, Arthur Ashkin investigated dielectric particles exposed to a tightly focused laser beam. He discovered that particles could be trapped in the laser focus. By now, this technique termed *optical tweezers* has found widespread applications in biology and physics for manipulation of tiny objects and measurement of minute forces. Arthur Ashkin was awarded the Nobel Prize in Physics 2018 for his ground-breaking work on the principles of optical trapping. In this exercise we consider a dielectric particle (with no material loss) in the focus of a Gaussian beam with angular frequency ω. The particle is significantly smaller than the wavelength λ and we therefore model it as a dipolar scatterer with purely real electrostatic polarizability

$$\alpha_0 \; = \; 4\pi\varepsilon_0 \, a^3 \, \frac{\varepsilon - 1}{\varepsilon + 2}. \tag{2}$$



(a) (2 pts.) Using the electrodynamic correction to the electrostatic polarizability derived in the previous problem, show that the electrodynamic polarizability is, to first order in the electrodynamic correction, given by

$$\alpha = \alpha' + \mathrm{i}\alpha'', \tag{3}$$

where the real part α' is given by the electrostatic polarizability α_0. Derive the imaginary part α'' and express it using the wavenumber k, the electrostatic polarizability, and numerical constants.

(b) (3 pts.) The force acting on a dipole $\boldsymbol{p}$ in an electromagnetic field is given by

$$\mathbf{F}(\mathbf{r}) \; = \; \sum_i \frac{1}{2} \mathrm{Re}\Big\{ p_i^* \, \nabla E_i(\mathbf{r}) \Big\} \tag{4}$$

where $i \in [x, y, z]$. Show that, for a scatterer wiht polarizability $\alpha = \alpha' + \mathrm{i}\alpha''$, this force can be written as

$$\mathbf{F}(\mathbf{r}) \; = \; \frac{\alpha'}{2} \sum_i \mathrm{Re}\Big\{ E_i^*(\mathbf{r}) \, \nabla E_i(\mathbf{r}) \Big\} \; + \; \frac{\alpha''}{2} \sum_i \mathrm{Im}\Big\{ E_i^*(\mathbf{r}) \, \nabla E_i(\mathbf{r}) \Big\}. \tag{5}$$

The first term in Eq. (5), which can be expressed as $\mathbf{F}_{\mathrm{grad}} = (\alpha'/4)\nabla(\mathbf{E}^* \!\cdot \mathbf{E})$, is referred to as the gradient force. From this expression, we see that $\mathbf{F}_{\mathrm{grad}}$ is the gradient of the potential $(\mathbf{E}^* \!\cdot \mathbf{E})$. Consequently, $\mathbf{F}_{\mathrm{grad}}$ is conservative, which means that $\nabla \times \mathbf{F}_{\mathrm{grad}} = 0$. The second term $\mathbf{F}_{\mathrm{scatt}}$ is referred to as the scattering force. It cannot be expressed as the gradient of a potential. Consequently, $\mathbf{F}_{\mathrm{scatt}}$ is nonconservative and accordingly $\nabla \times \mathbf{F}_{\mathrm{scatt}} \neq 0$.

Consider a Gaussian beam in vacuum polarized along the x-axis and focused under an angle $\theta = 53°$. This high numerical aperture stretches the limits of the paraxial approximation, but it turns out that our results are still a remarkably good representation of the physical reality. Use the small angle approximation of the trigonometric functions throughout the following. The wavelength is $\lambda = 1064\,\mathrm{nm}$ and the field strength at the focus is $E_0 = 2 \times 10^7\,\mathrm{V/m}$. The particle's radius is $a = 150\,\mathrm{nm}$ and its dielectric constant is $\varepsilon = 2.1$. The origin of our coordinate system is centered at the focus of the Gaussian beam.

(c) (8 pts.) Calculate the gradient force $\mathbf{F}_{\mathrm{grad}}(x, 0, 0)$ along the x-axis and plot it in the range $x = [-\lambda \ldots \lambda]$.

(d) (3 pts.) Expand $F^{(x)}_{\mathrm{grad}}(x, 0, 0)$ (x-component of the gradient force along the x-axis) to linear order in x. For small displacements from the focus, the gradient force can now be written as $F_{x_{\mathrm{grad}}} \approx -\kappa_x\, x$, i.e., the particle behaves as if it were attached to the focus by a spring with spring constant κ_x. Determine the spring constant κ_x. Your expression should contain only α', E_0, θ, and λ, besides numerical factors. Evaluate the numerical value of the spring constant κ_x for the parameters given in this problem. Units of μN/m should be appropriate.

(e) (3 pts.) In the linear approximation, the equation of motion for a particle moving along the x-axis is $m\ddot{x} + \kappa_x x = 0$. Assume that the specific density of the particle is $\rho = 2.5\,\mathrm{g/cm^3}$ and calculate the oscillation frequency Ω of the particle along the x-axis.

(f) (8 pts.) Give an expression for the gradient force $\mathbf{F}_{\mathrm{grad}}$ along the z-axis and plot it in the range $z = [-\lambda \ldots \lambda]$. Calculate the spring constant κ_z describing the restoring force in the linear approximation. Along which direction, x or z, is the restoring force acting on the particle stronger?

(g) (4 pts.) The scattering force $\mathbf{F}_{\mathrm{scatt}}$ pushes the particle away from the focus along the optical axis (z). For the values given above, plot $\mathbf{F}_{\mathrm{scatt}}$ along the z-axis in the range $z = [-\lambda \ldots \lambda]$.
Optional: Find an analytical expression for $F^{(z)}_{\mathrm{scatt}}$.

(h) (4 pts.) Plot the total force $\mathbf{F}$ along the z-axis in the range $z = [-\lambda \ldots \lambda]$ and determine from your plot the displacement Δz of the equilibrium position due to the scattering force.

(i) (5 pts.) One finds experimentally that it is impossible to stably trap too large particles. Give a quantitative argument why this is the case by considering the radius dependence of the two force components discussed above.

3 The fluorescence lifetime of a hydrogen atom [optional]

We have understood that the spontaneous emission rate of a quantum emitter is (besides natural constants) governed by the product of the square of the transition dipole moment and the local density of optical states (LDOS) according to

$$\gamma = \frac{\pi\omega_0}{3\hbar\varepsilon_0} |\hat{\boldsymbol{p}}|^2 \rho(\boldsymbol{r}_0, \omega_0). \tag{6}$$

The transition dipole moment is an intrinsic quantum mechanical property of the emitter. In contrast, the LDOS is a purely classical quantity and a property of the electromagnetic environment. In the last exercise set, we encountered an example of a simple system (Drexhage's mirror) and its associated LDOS. In the current problem, we consider the transition dipole moment of a simple quantum emitter, the hydrogen atom in vacuum. While the calculation of such a quantum mechanical property is not at the heart of the Nano Optics lecture, this example shall serve us as a reference point to illustrate the very different character of the ingredients playing a role in the spontaneous emission process.
In this exercise, we consider a hydrogen atom in vacuum in the excited state characterized by the quantum numbers $(n, l, m) = (2, 1, 1)$.

(a) Derive an expression for the decay rate γ that, besides natural and numerical constants, only depends on the transition frequency ω_0 of the decay, and the transition dipole moment $\hat{\boldsymbol{p}}$. Make use of the definition of the LDOS you know from the lecture and your derivation of the LDOS in vacuum from the last homework problem set.

(b) For the hydrogen atom under consideration, which final electronic states are available to decay into?

(c) Determine the numerical value of the (angular) photon emission frequency ω_0 of the available transition. On the way, express the energy eigenvalues of the hydrogen atom using (besides numerical factors) only the electron mass, the speed of light, and the fine structure constant.
Hint: The energy eigenvalues of the hydrogen atom are $E_n = -E_0/n^2$ with the Rydberg energy $E_0 = me^4/(32\pi^2\varepsilon_0^2\hbar^2)$.

(d) We now turn to the calculation of the transition dipole moment $\hat{\boldsymbol{p}} = -e\boldsymbol{r}$. Calculate the expectation value of $\hat{\boldsymbol{p}} = (\hat{p}_x, \hat{p}_y, \hat{p}_z)$ for the transition under consideration.
Hint: The wavefunctions of the states are

$$\psi_{1,0,0}(\boldsymbol{r}) = \sqrt{\frac{1}{\pi a_0^3}} \exp\left[-r/a_0\right], \tag{7}$$

$$\psi_{2,1,1}(\boldsymbol{r}) = -\sqrt{\frac{1}{64\pi a_0^3}} \frac{r}{a_0} \sin\theta \exp\left[\mathrm{i}\phi\right] \exp\left[-r/(2a_0)\right], \tag{8}$$

with the Bohr radius $a_0 = \hbar/(cm\alpha)$.

(e) Give an expression for the decay rate γ for the transition under consideration that contains only the rest energy of the electron mc^2, Planck's constant, and the fine structure constant α.

(f) Calculate the numerical value of the fluorescence lifetime $\tau = 1/\gamma$ of the state $(n, l, m) = (2, 1, 1)$ of the hydrogen atom.