Nanophotonics 2020 - Scattering & Microscopy

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Scattering

Plasmonic antennas are metal nanoparticles that have very large optical polarizabilities due to Localized Surface Plasmon Resonances (LSPR), and therefore they can very efficiently scatter light. This makes them interesting for applications such as nano-scale detectors and sensors, diffractive optical elements, and light trapping in solar cells. At the same time, they can absorb light and convert its energy into heat, which is used by researchers to locally heat and destroy tumor cells [1]. In this exercise we will look at scattering and absorption by dipolar plasmonic nanoparticles.

The polarizability α relates the induced dipole moment p to the incident field E_{inc}:

$$p = \alpha E_{\text{inc}}. \tag{1}$$

In case of a small spherical nanoparticle with relative permittivity ε_1, surrounded by a medium with relative permittivity ε_2, α can be approximated by the *static* polarizability:

$$\alpha_{\text{st}} = 3V\varepsilon_0\varepsilon_2\frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2}, \tag{2}$$

where $\varepsilon_0 = 8.854 \times 10^{-12}$ F/m, and V is the nanoparticle's volume. Assume that ε_1 is described by the Drude model:

$$\varepsilon_1 = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\omega\gamma}, \tag{3}$$

with parameters typical for silver: $\varepsilon_\infty = 5$, $\hbar\omega_p = 9$ eV, and $\hbar\gamma = 0.05$ eV.

Q1: Assume that the nanoparticle is embedded in PMMA polymer with $\varepsilon_2 = n^2$, where n is the refractive index ≈ 1.49 at 20°C and 1.48 at 80°C.

1a) Plot real and imaginary part of α_{st}/V as a function of wavelength λ in the frequency range 2.7-3.2 eV for both values of n. Use conversion formula: $\lambda[\text{nm}] = 1239/(\hbar\omega[\text{eV}])$.

1b) Resonant wavelength λ_{res} can be defined as the wavelength at which $\text{Re}(\alpha_{\text{st}}) = 0$. Assuming that n changes linearly with temperature, plot λ_{res} versus temperature from 0 to 100°C. What is the temperature shift of λ_{res} in nm per °C?

In Q2 and Q3 assume $n = 1.49$.

Q2: Optical theorem states that, apart from the intrinsic Drude damping (γ), the electromagnetic energy stored in a resonant nanoparticle is lost by radiation. This yields a modified expression for the so-called *dynamic* polarizability α_{dyn}:

$$\alpha_{\text{dyn}}^{-1} = \alpha_{\text{st}}^{-1} - \frac{ik^3}{6\pi\varepsilon_0\varepsilon_2}, \tag{4}$$

where $k = 2\pi n/\lambda$.

2a) Plot the wavelength dependence of $\text{Im}(\alpha_{\text{st}})$ and $\text{Im}(\alpha_{\text{dyn}})$ normalized to maximum, in the range 2-4 eV, for nanoparticles of diameters $d = 25$ and 50 nm.

2b) Plot the values of FWHM (full width at half maximum) of the peak in $\text{Im}(\alpha_{\text{dyn}})$ divided by the FWHM of the peak in $\text{Im}(\alpha_{\text{st}})$ as a function of d in the range 1-100 nm. What is the effect of radiation damping?

[1] Rizia Bardhan et al., Theranostic Nanoshells: From Probe Design to Imaging and Treatment of Cancer, Accounts of Chemical Research 44 (2011).

Q3: Total optical losses in plasmonic antennas are quantified by the extinction cross-section σ_{ext}:

$$\sigma_{\text{ext}} = \frac{k}{\varepsilon_0 \varepsilon_2} \text{Im}(\alpha_{\text{dyn}}), \tag{5}$$

while the radiative contribution to these losses is described by the scattering cross-sections σ_{scat}:

$$\sigma_{\text{scat}} = \frac{k^4}{6\pi \varepsilon_0^2 \varepsilon_2^2} |\alpha_{\text{dyn}}|^2, \tag{6}$$

and the absorption cross-section is simply the difference between the two:

$$\sigma_{\text{abs}} = \sigma_{\text{ext}} - \sigma_{\text{scat}}. \tag{7}$$

3a) The nanoparticle's geometric cross-section is: $S = \pi d^2/4$. Plot the wavelength dependence of σ_{scat}/S and σ_{abs}/S in the range 2-4 eV for $d = 25$ and 50 nm.

3b) Plot the peak values of σ_{scat}/S and σ_{abs}/S as a function of d in the range 1-100 nm. What d allows for the most efficient scattering per geometric area? What d leads to the most efficient absorption per geometric area? For which d absorption and scattering are equal?

Microscopy

Due to their strong scattering properties, individual plasmonic nanoparticles can be clustered, e.g., to form directional antennas. In the following exercise, we will use a small ensemble of dipolar sources to simulate the image formation in an aberration-free microscope.

In conventional microscopes, image formation is based on the light collected by an objective at a far distance from the source. The distribution of electric field E_{far} in the far zone is a function of the in-plane momentum components k_x and k_y (corresponding to certain radiation angles), and can be simulated from the spatial near field distribution (E_{near}) using Fourier transform:

$$E_{\text{far}}(k_x, k_y) = \int\int_{-\infty}^{+\infty} E_{\text{near}}(x, y) e^{ik_x x + ik_y y} dx dy \tag{8}$$

The image $|E_{\text{image}}(x, y)|^2$ is constructed by inverse Fourier transform of $E_{\text{far}}(k_x, k_y)$. Conventional microscopes can only collect propagating waves, i.e., waves with real momentum component in the propagation direction (k_z). Since $k_z^2 + k_x^2 + k_y^2 = k^2$, the maximum collected in-plane momentum is limited by: $k_x^2 + k_y^2 \leqslant k^2$. Furthermore, it is not possible to collect all angles from -90 to $+90°$, but only in the range from $-\theta_{\max}$ to $+\theta_{\max}$ inside the numerical aperture NA of a given objective: $\text{NA} = n \sin\theta_{\max}$. This leads to truncation of the Fourier space:

$$k_x^2 + k_y^2 \leqslant (k_0 \text{NA})^2 \qquad (\text{where } k_0 = 2\pi/\lambda) \tag{9}$$

which is the main factor limiting the imaging resolution in conventional optical microscopes.

Q4: Consider an array of 5 point sources arranged along the x axis ($y = 0$), with equal spacing a between them ($x = -2a, -a, 0, a, 2a$). Assume $E_{\text{near}} = 1$ at each of these points, and 0 otherwise. Set: $\lambda = 600$ nm, and $a = 500$ nm.

4a) Plot $|E_{\text{far}}|^2$ in the range $-3k_0 < k_x < 3k_0$, normalized to maximum, generated by E_{near}. Mark the truncation of the k_x range associated with $\text{NA} = 0.7$ and $\text{NA} = 1.4$.

4b) Plot $|E_{\text{image}}|^2$ for $-4a < x < 4a$, normalized to maximum, obtained with an objective of $\text{NA} = 0.7$. Does it show clearly all sources?

4c) Do the same for $\text{NA} = 1.4$. How is the image improved and why? (explain it with your previous plot of $|E_{\text{far}}|^2$). How one can achieve $\text{NA} > 1$ in real-life experiments? What other parameter can be changed to improve the resolution?