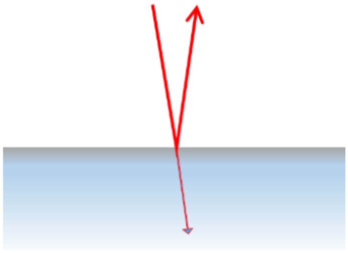
Exercise 3.1 (skin depth of metals)

In the low frequency range ($\omega/\Gamma \ll 1$) the reflectance of metals is very high and approaches 1 for metals with high conductivity. Nevertheless light is transmitted through the metal layer and strongly attenuated. The penetration depth is called skin depth.



a) Provide an expression for the skin depth δ of a metal as a function of plasma frequency ω_p and relaxation time τ. Introduce the DC conductivity σ_0 to replace ω_p and τ in this expression.

b) Determine the skin depth δ for aluminum for the incoming light frequencies of 100 MHz as well as 10^{15} Hz using the DC conductivity of $\sigma_0 = 3.69 \cdot 10^7\ \Omega^{-1}\text{m}^{-1}$ for aluminum.

c) Compare metallic reflection to the phenomenon total reflectance (compare the wavelength to the penetration depth/skin depth in both cases).

Exercise 3.1 (skin depth of metals). Solution

From the lecture we know that complex dielectric permittivity expressed as:

$$\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega)$$

$$\varepsilon_1(\omega) = 1 - \frac{\omega_p^2}{\Gamma^2 + \omega^2}$$

$$\varepsilon_2(\omega) = \frac{\omega_p^2 \Gamma}{\omega(\Gamma^2 + \omega^2)}$$

And Γ=1/τ for the case $\frac{\omega}{\Gamma} \ll 1\,, or\ \omega\tau \ll 1$ respectively. Following the lecture results we also have:

$$n = \frac{1}{\sqrt{2}}\sqrt{\varepsilon_1 + \sqrt{\varepsilon_1^2 + \varepsilon_2^2}}$$

$$k = \frac{1}{\sqrt{2}}\sqrt{-\varepsilon_1 + \sqrt{\varepsilon_1^2 + \varepsilon_2^2}}$$

For low frequencies $\omega\tau \ll 1$:

$$\varepsilon_2 \gg \varepsilon_1$$

$$n \approx k \approx \sqrt{\frac{\varepsilon_2}{2}}$$

$$\varepsilon_2(\omega) = \frac{\omega_p^2 \Gamma}{\omega(\Gamma^2 + \omega^2)} \approx \frac{\omega_p^2 \tau}{\omega}$$

For dimensional absorption coefficient α:

$$\alpha = \frac{4\pi k}{\lambda} = \frac{2\omega \mathrm{k}}{c} \approx \frac{2\omega}{c}\sqrt{\frac{\varepsilon_2}{2}} = \sqrt{\frac{2\omega_p^2 \tau\omega}{c^2}}$$

Since $\omega_p^2\tau = \frac{\sigma_0}{\varepsilon_0}$:

$$\alpha = \sqrt{\frac{2\sigma_0\omega}{\varepsilon_0 c^2}}$$

We see that the absorption coefficient is proportional to the square root of DC conductivity σ_0 and frequency ω. So, high frequency field has high absorption coefficient and therefore can only penetrate a short distance into a conductor such as metal. This phenomenon is called the skin effect.

We remember that material absorbs the light passing through: $I(z) = I_0 e^{-\alpha z}$

If the field strength decays as: $E(z) = E_0 e^{-\frac{z}{\delta}}$

Then the intensity decays as E^2: $I(z) \sim |E^2| \sim e^{-\frac{2z}{\delta}}$

So, we can connect the characteristic length of field decay δ with the absorption coefficient α:

$$\delta = \frac{2}{\alpha}$$

The coefficient δ is also known as skin depth.

B) Skin depth for aluminum:

frequencies of 100 MHz as well as 10^{15} Hz

$$\sigma_0 = 3.69 \cdot 10^7\ \Omega^{-1}\mathrm{m}^{-1}$$

$$\frac{\omega_1}{2\pi} = 100\ MHz$$

$$\frac{\omega_2}{2\pi} = 10^{15}\ Hz$$

$$\varepsilon_0 = 8.854 \cdot 10^{-12}\ F{\cdot}m^{-1}$$

$$\alpha_1 = \sqrt{\frac{2\sigma_0\omega_1}{\varepsilon_0 c^2}} = \sqrt{\frac{4\pi\sigma_0 f_1}{\varepsilon_0 c^2}} = \sqrt{\frac{4\pi \cdot 3.69 \cdot 10^7 \cdot 10^8}{8.854 \cdot 10^{-12} \cdot 9 \cdot 10^{16}}} = 240 \cdot 10^3\ m^{-1}$$

$$\delta_1 = 8.3\ um$$

$$\alpha_2 = \sqrt{\frac{2\sigma_0\omega_2}{\varepsilon_0 c^2}} = \sqrt{\frac{4\pi\sigma_0 f_2}{\varepsilon_0 c^2}} = \sqrt{\frac{4\pi \cdot 3.69 \cdot 10^7 \cdot 10^{15}}{8.854 \cdot 10^{-12} \cdot 9 \cdot 10^{16}}} = 762 \cdot 10^6\ m^{-1}$$

$$\delta_2 = 2.6\ nm$$

C) Let’s compare metal absorption to total reflection and to the wavelength

Wavelength λ	Skin depth δ
$\lambda_1 = 3\ m$	$\delta_1 = 8.3\ um$
$\lambda_2 = 300\ nm$	$\delta_2 = 2.6\ nm$

$$\lambda = \frac{c}{\nu}$$

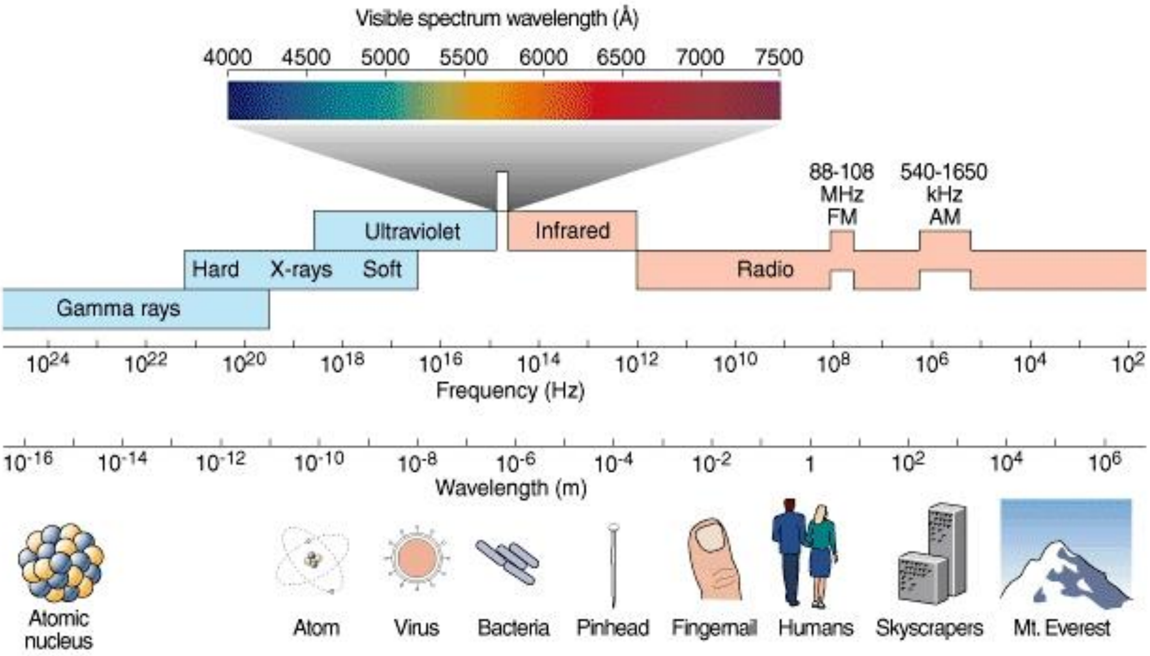
Penetration depth in total reflection d:

$$d = \frac{\lambda}{2\pi\sqrt{n_1^2 \sin^2\theta_i - n_2^2}} = \frac{\lambda}{2\pi\sqrt{\Delta n^2}} \sim \lambda$$

Skin depth:

$$\delta = \sqrt{\frac{\lambda\varepsilon_0 c}{\pi\sigma_0}} \sim \sqrt{\lambda}$$

Now we can compare obtained values with the EM field frequency range and corresponding sizes:



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Exercise 3.2 (Plasmon absorption and scattering of metal nanoparticles)

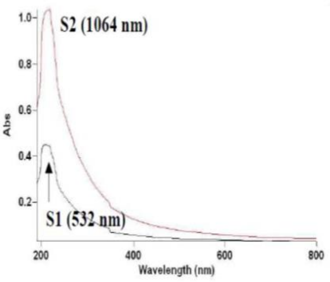
Interaction of light with small metal nanoparticles at the resonance frequency result in the excitation of surface plasmons leading to both absorption and scattering.

a) From the absorption cross section, determine the resonance frequency of small metallic nanospheres.

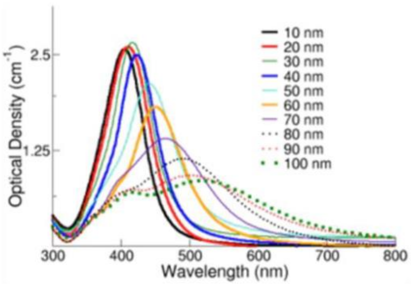
Hint: For the metal use the dielectric function derived from the Drude Zener model. Surface plasmon resonance occurs close to the bulk plasma frequency, therefore $\omega/\Gamma \gg 1$ applies.

b) Determine the wavelength of maximum absorption for small (< 10 nm diameter) aluminum nanospheres suspended in ethanol (estimate the plasma frequency of aluminum from the reflectance vs wavelength figure in the lecture notes).

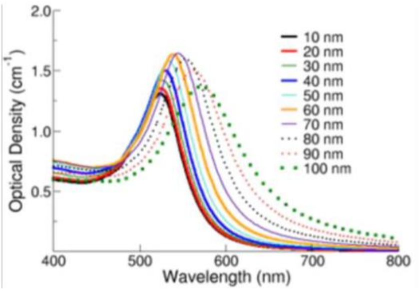
c) Calculate the maximum absorption wavelength for Ag and Au nanospheres suspended in water by obtaining the plasma frequency from the DC conductivity (Au: $4.5 \cdot 10^{7}\ \Omega^{-1}\mathrm{m}^{-1}$, Ag: $6.3 \cdot 10^{7}\ \Omega^{-1}\mathrm{m}^{-1}$) and relaxation times (Au: $3 \cdot 10^{-14}\mathrm{s}^{-1}$, Ag: $4 \cdot 10^{-14}\mathrm{s}^{-1}$). Discuss your results.



Al nanospheres in ethanol



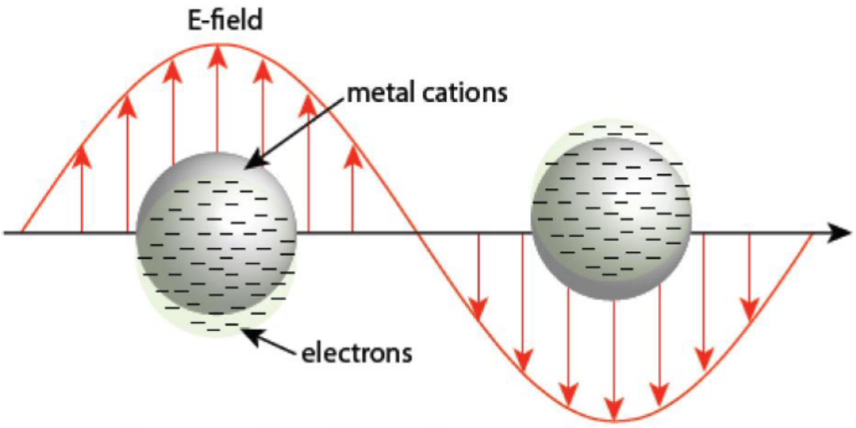
silver nanospheres in water



gold nanospheres in water

Exercise 3.2 (Plasmon absorption and scattering of metal nanoparticles). Solution

A) In this problem we consider metal nanoparticles with the radius R excited by incoming electromagnetic wave E with frequency ω:



sphere of radius R

For the absorption cross section, we know from the lecture (also see [1, 2]):

$$\sigma_{abs} = \frac{8\pi^2}{\lambda} R^3 \mathrm{Im}\left[\frac{\tilde{\varepsilon}_M - \tilde{\varepsilon}_D}{\tilde{\varepsilon}_M + 2\tilde{\varepsilon}_D}\right]$$

Where R is the radius of the spherical metallic particle, $\tilde{\varepsilon}_M$ is the permittivity of the particle, ε_D is the permittivity of the dielectric medium.

In general case, the permittivity depends on wavelength ω, and we need to consider the cross section as function of ω:

$$\sigma_{abs}(\omega) = \frac{4\pi\omega}{c} R^3 \mathrm{Im}\left[\frac{\tilde{\varepsilon}_M(\omega) - \tilde{\varepsilon}_D(\omega)}{\tilde{\varepsilon}_M(\omega) + 2\tilde{\varepsilon}_D(\omega)}\right]$$

For the resonance, we need to maximize the cross section σ_{abs} over the variable of exciting wave frequency ω. Since we have a fraction, we can maximize it by setting the denominator to zero. However, it is important to note that the condition of maximum of the expression

$$\frac{\tilde{\varepsilon}_M(\omega) - \tilde{\varepsilon}_D(\omega)}{\tilde{\varepsilon}_M(\omega) + 2\tilde{\varepsilon}_D(\omega)}$$

is not equal to the condition of maximum of the expression

$$\mathrm{Im}\left[\frac{\tilde{\varepsilon}_M(\omega) - \tilde{\varepsilon}_D(\omega)}{\tilde{\varepsilon}_M(\omega) + 2\tilde{\varepsilon}_D(\omega)}\right]$$

$$\frac{\tilde{\varepsilon}_M(\omega) - \tilde{\varepsilon}_D(\omega)}{\tilde{\varepsilon}_M(\omega) + 2\tilde{\varepsilon}_D(\omega)} = A + iB$$

$$A = \frac{{\varepsilon_1^M}^2 + \varepsilon_1^M \varepsilon_1^D - 2{\varepsilon_1^D}^2 + {\varepsilon_2^M}^2 + \varepsilon_2^M \varepsilon_2^D - 2{\varepsilon_2^D}^2}{(\varepsilon_1^M + 2\varepsilon_1^D)^2 + (\varepsilon_2^M + 2\varepsilon_2^D)^2}$$

$$B = 3\frac{\varepsilon_1^D \varepsilon_2^M - \varepsilon_1^M \varepsilon_2^D}{(\varepsilon_1^M + 2\varepsilon_1^D)^2 + (\varepsilon_2^M + 2\varepsilon_2^D)^2}$$

The expression

$$\mathrm{Im}\left[\frac{\tilde{\varepsilon}_M(\omega) - \tilde{\varepsilon}_D(\omega)}{\tilde{\varepsilon}_M(\omega) + 2\tilde{\varepsilon}_D(\omega)}\right]$$

Reaches maximum when the denominator of B turns to zero:

$$(\varepsilon_1^M + 2\varepsilon_1^D)^2 + (\varepsilon_2^M + 2\varepsilon_2^D)^2 = 0$$

As you can see, this expression is different from the denominator of the initial fraction $\tilde{\varepsilon}_M(\omega) + 2\tilde{\varepsilon}_D(\omega)$.

To simplify the expression, we can consider the dielectric without absorption, or in other words say that for the permittivity of the dielectric medium $\varepsilon_2^D \ll \varepsilon_1^D$, and therefore $\tilde{\varepsilon}_D \approx \varepsilon_1^D \approx n_D^2$, ($\kappa \approx 0$).

Then the condition of the resonance becomes

$$(\varepsilon_1^M + 2\varepsilon_1^D)^2 + {\varepsilon_2^M}^2 = 0$$

Then, we can consider the resonance for the metal particles. It is known, that surface plasmon resonance occurs close to the bulk plasma frequency (empirical fact), therefore: $\omega_{res} \approx \omega_p$

For the permittivity of metal particles, we have:

$$\varepsilon_1(\omega) = 1 - \frac{\omega_p^2}{\Gamma^2 + \omega^2}$$

$$\varepsilon_2(\omega) = \frac{\omega_p^2 \Gamma}{\omega(\Gamma^2 + \omega^2)}$$

And taking into account that we got $\frac{\omega_{res}}{\Gamma} \gg 1$:

$$\varepsilon_1(\omega) = 1 - \frac{\omega_p^2}{\Gamma^2 + \omega^2} \approx 1 - \frac{\omega_p^2}{\omega^2}$$

$$\varepsilon_2(\omega) = \frac{\omega_p^2 \Gamma}{\omega(\Gamma^2 + \omega^2)} \approx \frac{\omega_p^2 \Gamma}{\omega^3} \approx \frac{\Gamma}{\omega_{res}} \ll 1$$

$$\varepsilon_1(\omega) \gg \ \varepsilon_2(\omega)$$

$$\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega) \approx \varepsilon_1(\omega)$$

For the condition of resonance in this case we will have

$$(\varepsilon_1^M + 2\varepsilon_1^D)^2 + {\varepsilon_2^M}^2 \approx (\varepsilon_1^M + 2\varepsilon_1^D)^2 = 0$$

Finally, we get, that for the resonance we want to maximize the expression:

$$\varepsilon_1^M(\omega_{res}) + 2\varepsilon_1^D(\omega_{res}) = 0$$

If we neglect the dependence of ε_1^D on frequency ω (we assume very slow change of ε_1^D), then we have:

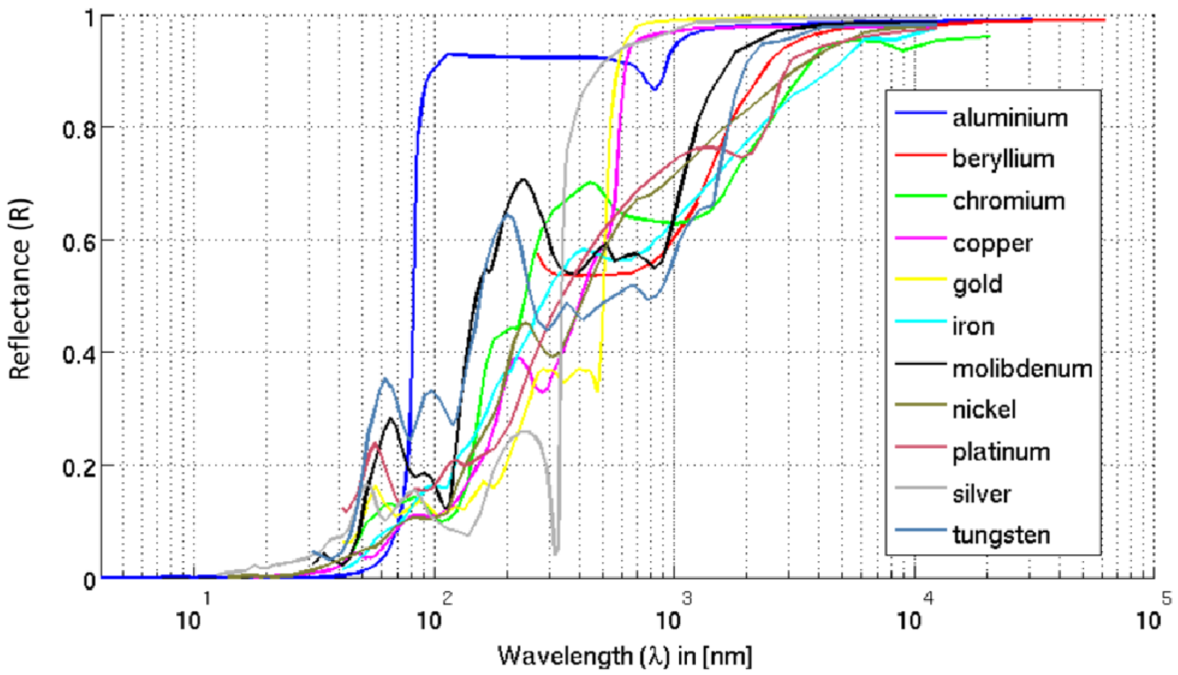
$$\varepsilon_1(\omega_{res}) + 2\varepsilon_D = 1 - \frac{\omega_p^2}{\omega_{res}^2} + 2\varepsilon_D = 0$$

$$\omega_{res}^2 = \frac{\omega_p^2}{1 + 2\varepsilon_D}$$

B) Refractive index for ethanol

$$n_{ethanol} \approx 1.36 = \sqrt{\varepsilon_D}$$

From the lecture we have the reflectance of different metals vs wavelength:



Since for metals the reflectance becomes $R \approx 1$ after $\omega \leq \omega_p$, then the plasma frequency for aluminum corresponds to $\lambda_p \approx 80$ nm. For the frequency:

$$\omega_p = \frac{2\pi c}{\lambda_p} \approx 2.36 \cdot 10^{16}\ \mathrm{s}^{-1}$$

The resonance frequency for aluminum:

$$\omega_{res}^2 = \frac{\omega_p^2}{1 + 2\varepsilon_D} = \frac{2.36^2 \cdot 10^{32}}{1 + 2 \cdot 1.8496} \approx 1.19 \cdot 10^{32}\ \mathrm{s}^{-2}$$

$$\omega_{res} = 1.09 \cdot 10^{16}\ \mathrm{s}^{-1}$$

$$\lambda_{res} = \frac{2\pi c}{\omega_{res}} = 173\ \mathrm{nm}$$

C) For the plasma frequency (see exercise 3.1):

$$\omega_p^2 \tau = \frac{\sigma_0}{\varepsilon_0}$$

Particle	σ_0, $\Omega^{-1}\mathrm{m}^{-1}$	τ, s	ω_p, s^{-1}	ω_{res}, s^{-1}	λ_{res}, nm
Au	$4.5 \cdot 10^7$	$3 \cdot 10^{-14}$	$1.31 \cdot 10^{16}$	$6.1 \cdot 10^{15}$	308
Ag	$6.3 \cdot 10^7$	$4 \cdot 10^{-14}$	$1.33 \cdot 10^{16}$	$6.25 \cdot 10^{15}$	301

References:

[1] van Dijk, M. A., Tchebotareva, A. L., Orrit, M., Lippitz, M., Berciaud, S., Lasne, D., ... & Lounis, B. (2006). Absorption and scattering microscopy of single metal nanoparticles. Physical Chemistry Chemical Physics, 8(30), 3486-3495.

[2] Hulst, H. C., & van de Hulst, H. C. (1981). Light scattering by small particles. Courier Corporation.