General Properties of Local Plasmons in Metal Nanostructures

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Under the quasistatic approximation, the characteristics of a local plasmon resonance of a metal nanostructure exhibit several general properties. The resonance frequency depends on the fraction of plasmon energy residing in the metal through the real dielectric function of the metal. For a given resonant frequency, the Q factor of the resonance is determined only by the complex dielectric function of the metal material, independent of the nanostructure form or the dielectric environment. A simple result describing the effect of optical gain on the Q factor is also obtained.

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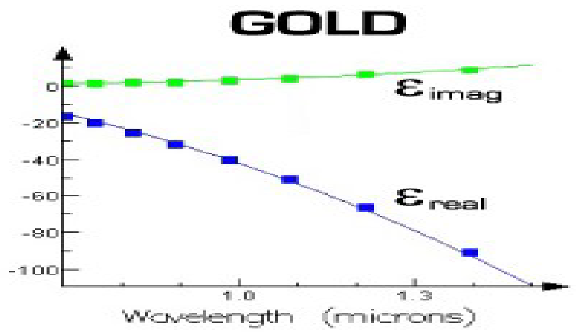
'Quasi-static' refers to the regime for which the finite speed of light can be neglected and field treated as if they propagate instantaneously.

In metal nanostructure, $L/\lambda = kL \ll 1$

Where L is the size of the nanostructure which is much smaller than the wavelength. Then local plasmon resonance is purely electric.

$$\varepsilon = \varepsilon' + i\varepsilon'' = N^2 = (n + i\kappa)^2 = n^2 - \kappa^2 + 2n\kappa i$$

$$n^2 - \kappa^2 = 1 - \frac{\omega_p^{\ 2}}{\omega^2 + \tau^{-2}} \qquad 2n\kappa = \frac{\omega_p^{\ 2}}{\omega^2 + \tau^{-2}}\left(\frac{1}{\omega\tau}\right)$$



http://www.lumerical.com/fdtd_multicoefficient_material_modeling.php

In the frequency region below bulk plasma frequency ω_p

$\varepsilon = \varepsilon' < 0$, dispersive

assume that $\varepsilon'' \ll |\varepsilon'|$ (low loss condition)

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Time-averaged energy in dispersive metal is,

$$\bar{U}_m = \int_{\Omega_{metal}} \frac{1}{8\pi}\left[\frac{d(\omega\varepsilon)}{d\omega}\bar{E}^2\right]dV$$

Whereas dielectric matter is assumed to be lossless and non-dispersive.

(1)

$$\bar{U}_d = \int_{\Omega_{dielectric}} \frac{1}{8\pi}\left[\varepsilon_d \bar{E}^2\right]dV$$

Take the large volume faraway from the resonant local field.

$$\int_{\Omega} \varepsilon_d \vec{E}\cdot\vec{E}\,dV = \int_{\Omega} \vec{D}\cdot\vec{E}\,dV = -\int_{\Omega} \vec{D}\cdot\vec{\nabla}\Phi\,dV$$

$$= -\int_{\Omega}[\vec{\nabla}\cdot(\Phi\vec{D}) - \Phi(\vec{\nabla}\cdot\vec{D})]dV = -\oint_{S_\Omega} \Phi(\vec{D}\cdot\hat{n})ds = 0$$

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with Ω divided into Ω_{metal}, $\Omega_{dielectric}$,

$$\Rightarrow \int_{\Omega_{dielectric}} [\varepsilon_d \bar{E}^2] dV = -\int_{\Omega_{metal}} [\varepsilon_m \bar{E}^2] dV \longrightarrow (2)$$

And by Kramer - Kronig relation,

$$\Rightarrow \varepsilon'(\omega) = 1 + \frac{2}{\pi} P.V \int_0^{\infty} \frac{x\varepsilon''}{x^2 - \omega^2} dx$$

$$\Rightarrow \frac{d(\omega\varepsilon'_m(\omega))}{d\omega} = -\varepsilon'_m + 2 + \frac{2}{\pi} P.V \int_0^{\infty} \frac{2x^3\varepsilon''}{(x^2 - \omega^2)^2} dx > -\varepsilon'_m$$

So,

$$\Rightarrow \frac{\bar{U}_m}{\bar{U}_d} = \frac{\int_{\Omega_{metal}} \frac{1}{8\pi} [\frac{d(\omega\varepsilon)}{d\omega} \bar{E}^2] dV}{-\int_{\Omega_{metal}} [\varepsilon_m \bar{E}^2] dV} \geq 1 \longrightarrow (3)$$

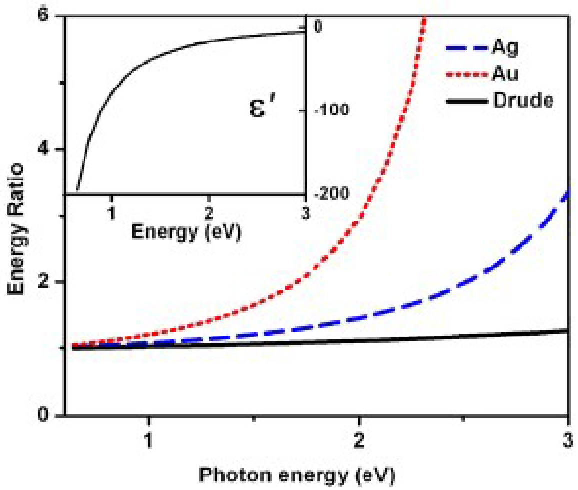


Fig (1)

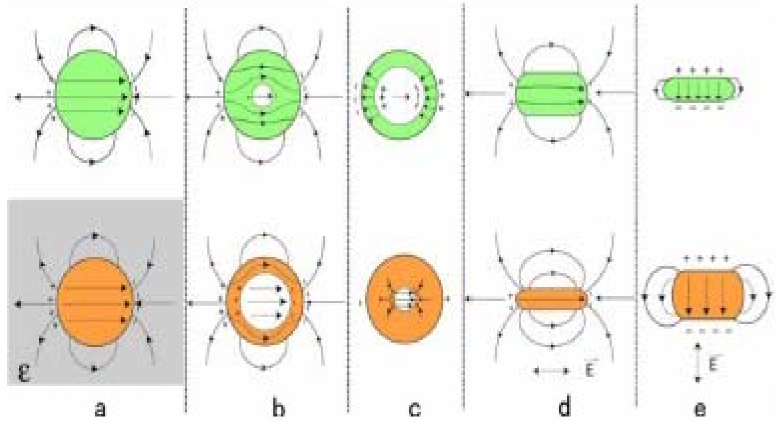


Fig (2)

From (2),

$$-\varepsilon_m = \int_{\Omega_{dielectric}} [\varepsilon_d \bar{E}^2] dV \Big/ \int_{\Omega_{metal}} \bar{E}^2 \, dV \longrightarrow (4)$$

Means that the energy ratio get larger for higher ε_d value Fig (2)-a

And the decrease of the thick of nanostructure redshift the plasmon resonance frequency then cause increase of $\frac{d(\omega\varepsilon)}{d\omega}$
Compensate the decreasing of metal volume Fig (2)-b Fig (2)-c

Aside from the resonance frequency, the other important parameter for plasmon resonance is Q(quality factor)

$$Q \equiv \frac{\text{total energy}}{\text{energy loss per one cycle}} = \frac{\omega \bar{U}_{total}}{d\bar{U}_{total}/dt}$$

$$= \frac{\int_{\Omega_{metal}} \frac{d(\omega\varepsilon_m')}{d\omega} \bar{E}^2 dV + \int_{\Omega_{dielectric}} \varepsilon_d \bar{E}^2 dV}{2\int_{\Omega_{metal}} \varepsilon_m'' \bar{E}^2 dV} = \frac{\omega \frac{d\varepsilon_m'}{d\omega}}{2\varepsilon_m''} \longrightarrow (5)$$

Q factor is the parameter that determine the sharpness of Plasmon resonance spectrum

$$E(t) = e^{-\omega t/2Q} e^{i(\omega-\omega_0)t}$$

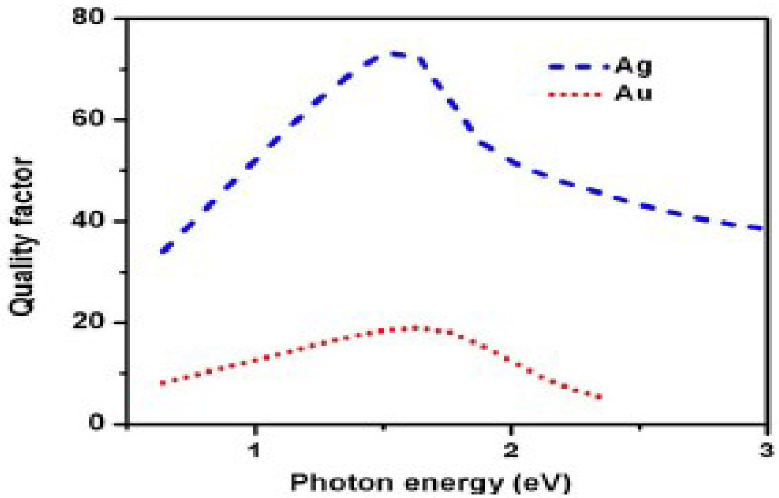
$$E(\omega) = \frac{1}{\sqrt{2\pi}} \int_0^{\infty} E_0 e^{-\omega t/2Q} e^{i(\omega-\omega_0)t} dt$$

$$\frac{\omega \bar{U}_{total}}{d\bar{U}_{total}/dt} = Q \rightarrow \bar{U}_{total}(\omega) = \bar{U}^{0}_{total} e^{-\omega t/Q}$$

Result of this Fourier transformation is Lorentizan model

$$|E(\omega)|^2 \propto \frac{1}{(\omega-\omega_0)^2 + (\omega_0/2Q)^2}$$

So, in quasi-static model the quality factor only depends on dielectric Function of material not geometrical shape.



Finally, investigate the effect of optical gain in dielectric material surrounding metal nanostructure.

assume that dielectric material is dispersive

and $\varepsilon_d = \varepsilon_g{}' + i\varepsilon_g{}'', \varepsilon_g{}'' < 0$

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$$Q = \frac{\int_{\Omega_{metal}} \frac{d(\omega\varepsilon_m^{'})}{d\omega} \bar{E}^2 \, dV + \int_{\Omega_{dielectric}} \frac{d(\omega\varepsilon_g^{'})}{d\omega} \bar{E}^2 \, dV}{2(\int_{\Omega_{metal}} \varepsilon_m^{"} \bar{E}^2 \, dV - \int_{\Omega_{dielectric}} \left|\varepsilon_g^{"}\right| \bar{E}^2 \, dV}$$

$$= \left(\frac{\varepsilon_m^{"}}{\left|\varepsilon_m^{'}\right|} - \frac{\left|\varepsilon_g^{"}\right|}{\varepsilon_g^{'}}\right)^{-1} \frac{\omega\left[\frac{d\varepsilon_m^{'}}{d\omega} + \frac{\left|\varepsilon_m^{'}\right|}{\varepsilon_g^{'}} \frac{d\varepsilon_g^{'}}{d\omega}\right]}{2\left|\varepsilon_m^{'}\right|} \longrightarrow \quad (7)$$

So, in specific case, $\frac{\varepsilon_m^{"}}{\left|\varepsilon_m^{'}\right|} - \frac{\left|\varepsilon_g^{"}\right|}{\varepsilon_g^{'}} \le 0$, the nano metal structure becomes oscillator or amplifier in principle.

Summary

- In quasi-static region, properties of metal nanostructure depend on purely it's dielectric function.
- Below the bulk plasma frequency, the reside energy ratio between metal and dielectric matter always larger than one.
- These relations constitute a useful frame work that encompasses results from wide range of studies on a specific nano structure.