Modeling the optical properties of metallic nanoparticles

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We present the theoretical models for the optical properties of metallic nanoparticles. Optical absorption and scattering efficiency, and transmission and reflection coefficient of a single metallic nanoparticle are analytically calculated by Mie theory while those of a random assembly of nanoparticles are investigated by the effective medium theory. Via these models promising applications of metallic nanoparticles in enhancing light emission and absorption are discussed.

Introduction

The optical properties of metallic particles have been studied long time ago. In recent decades a great deal of interest has intensified, in particular due to the discovery that Raman scattering can be increased by orders of magnitude through the utility of metallic nanoparticles. These noble metallic nanoparticles strongly enhance so-called localized surface plasmon resonances (or nanoparticle plasmon resonances) which make them excellent scatters and absorbers of visible light [1-3]. This leads to a considerable increase in research activities directed toward applications of metallic nanoparticles in efficiency enhancement of light absorber and light emitter.

Since the plasmon resonance of metallic nanoparticles is strongly sensitive to the particle size, shape and the dielectric properties of the surrounding medium (thus resulting in their tunable optical properties by varying the size and shape), rigorous theoretical and numerical methods to describe nanoparticle optical properties play an important role in optimal design of light absorber and emitter. In this paper, we present theoretical models for the optical properties of single metallic nanosphere and isolated nanospheres as means to optimize light absorber and emitter design.

Calculation method

The optical properties of metallic nanospheres were qualified in terms of their calculated optical absorption and scattering efficiency, transmission and reflection coefficient, and their optical resonance wavelength. For the description of metallic nanoparticles in the frequency range of interest, the complex dielectric function of particles is calculated by Lorentz-Drude model. It is well-known that for nanospheres with diameters well below the wavelength of light, these optical properties were well-described by Mie theory for homogeneous spheres. The Mie absorption and scattering cross-section efficiency (Q_{abs} and Q_{scat}) is given by [4]:

$$Q_{abs} = \frac{C_{abs}}{\pi a^2},\ Q_{scat} = \frac{C_{scat}}{\pi a^2} \tag{1}$$

$$C_{abs} = \left(\frac{2\pi}{\lambda}\right)\mathrm{Im}[\alpha],\ C_{scat} = \frac{1}{6\pi}\left(\frac{2\pi}{\lambda}\right)^4 |\alpha|^2 .$$

where $\alpha = 3V\left[\dfrac{\varepsilon_p - \varepsilon_m}{\varepsilon_p + 2\varepsilon_m}\right]$ is the polarizability of the particle, V is the particle volume, ε_p is the dielectric function of the particle and ε_m is the dielectric function of the embedding medium.

The scattering and absorption efficiency determine the conversion of the incident light into the corresponding quantities. However, they give no information about the directionality of the scattered field. Through the transmission and reflection of light interacting with a single nanosphere a better understanding of the process can be obtained. If the transmitted light through a single nanoparticle is only collected along the incidence axis, the transmission is expressed as [2]

$$T = 1 - \eta\left(Q_{scat}^{b} + Q_{abs}\right). \tag{2}$$

and the reflection is given by

$$R = 1 - T - \eta Q_{abs} \tag{3}$$

where Q_{scat}^{b} is the backward scattering efficiency and η is the normalized surface coverage.

For a random assembly of isolated nanoparticles their transmission and reflection rates can be expressed as the transmission and reflection rates through a slab of nanoparticles. In this case the optical properties of the slab of non-interacting nanospheres embedded in the host material are well-described by the Maxwell-Garnet theory based on the concept of mean-field inside and outside the nanoparticles. With a low volume fraction of isolated nanoparticles, the effective dielectric permittivity is given by

$$\varepsilon_{eff} = \left[\frac{1 + 2f\gamma}{1 - f\gamma}\right]\varepsilon_m,\ \gamma = \frac{\varepsilon_p - \varepsilon_m}{\varepsilon_p + 2\varepsilon_m}, \tag{4}$$

and the reflection and transmission of the slab of nanoparticles are calculated by

$$R = \left|\frac{r\left(1 - \exp(ikn_{eff}h)\right)}{1 - r^2\exp(ikn_{eff}h)}\right|^2, m = \frac{n_{eff}}{n_m},\ r = \frac{1-m}{1+m}, \tag{5}$$

$$T = \left|\frac{4m}{(1+m)^2}\frac{\exp(-ikn_m h)}{\left[\exp(-ikn_{eff}h) - r^2\exp(ikn_{eff}h)\right]}\right|^2, \tag{6}$$

where k is the wavevector of an electromagnetic wave propagating in free space, n_m and n_{eff} is the refractive index of the surrounding medium and the effective refractive index of the slab of nanoparticles, respectively, and h is the slab thickness.

Results and discussion

In this section we start to investigate the optical properties of various single metallic nanoparticles and a random assembly of these nanoparticles. The nanoparticles used for calculation are gold (Au) and silver (Ag) embedded in the medium of silica (SiO_2) and GaN, respectively. The calculated spectra of the efficiency of optical absorption and scattering of these metallic nanoparticles with respect to the wavelength of light are shown in Fig. 1.

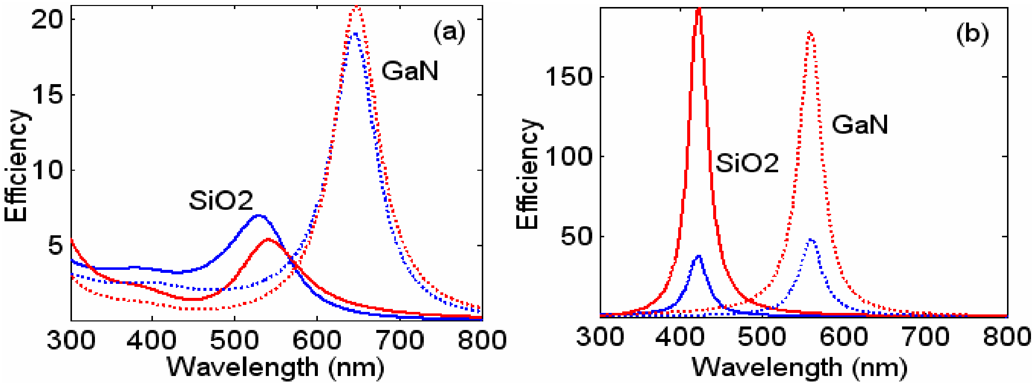

Fig. 1. Calculated spectra of the efficiency of absorption (blue lines) and scattering (red lines) for single 120 nm diameter Au (a) and Ag (b) particles embedded in SiO_2 (solid lines) and GaN (dotted lines).

The dimensionless of the scattering and absorption efficiencies can be converted to the corresponding cross-sections scattering and absorption by multiplication with the cross-sectional area of the particle. These cross-sections can also be directly related to efficient light absorbers. It is seen that at certain resonance frequency an enhanced absorption is observed. Corresponding to the optical absorption and scattering of these single nanoparticles the reflection and transmission coefficient are also calculated from Eq. (2) and (3), and shown in Fig. 2.

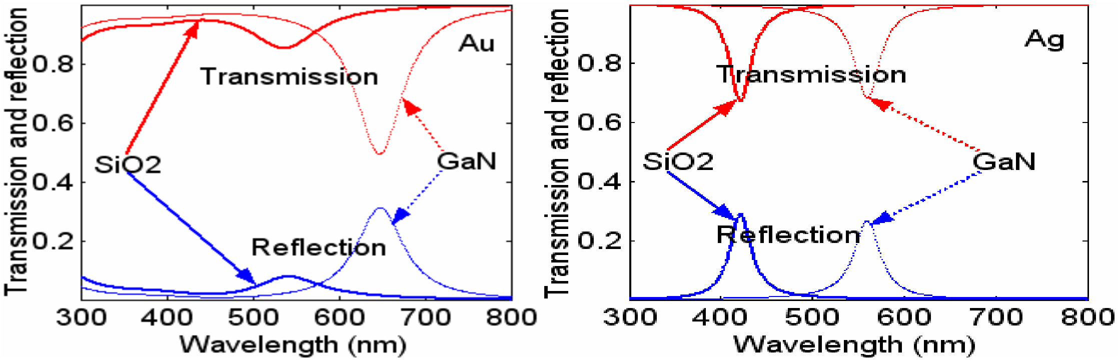

Fig. 2. Calculated spectra of the transmission (red lines) and reflection (blue lines) for single 120 nm diameter Au and Ag particles embedded in SiO_2 (solid lines) and GaN (dotted lines).

While the reflection and transmission spectra of the slab of the nanoparticles calculated by Maxwell-Garnet theory using Eq. (5) and (6) are plotted in Fig. 3. From the figure the resonance frequency is observed at certain diameter of particles. This will be a promising way to enhance light absorption and emission by nanoparticles. For instance an enhanced light emission of silicon light emitting diodes (LEDs) is observed by using Ag nanoparticles with diameter of 120 nm as seen in Fig. 4 where the enhanced Purcell factor is obtained by the localized surface plasmon resonance around the Ag particle.

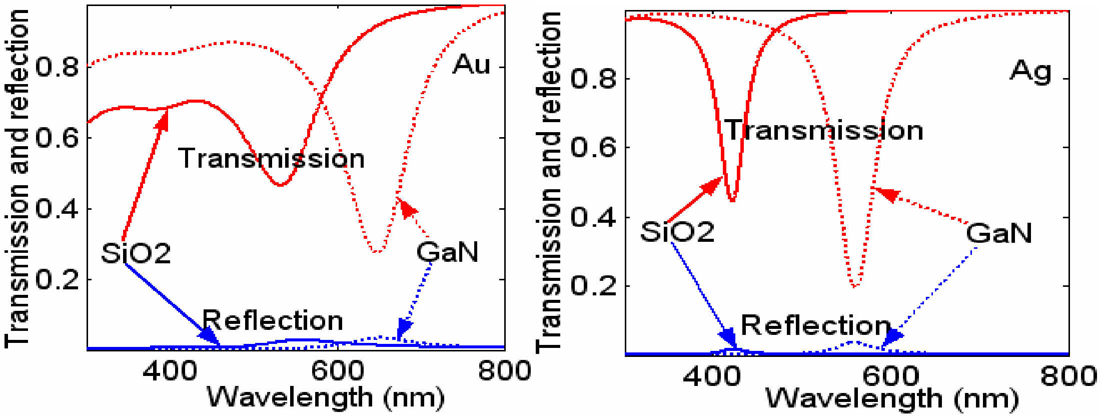

Fig. 3. Calculated spectra of the transmission (red lines) and reflection (blue lines) for slab of 120 nm diameter Au and Ag particles embedded in SiO_2 (solid lines) and GaN (dotted lines).

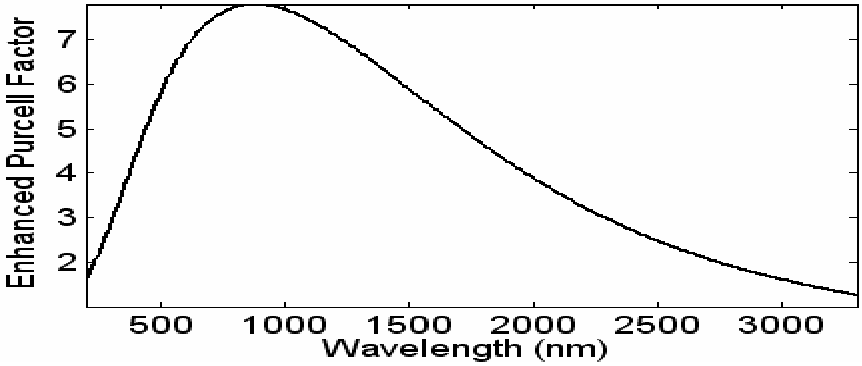

Fig. 4. Calculated enhancement of Purcell factor of silicon LED due to a 120 nm diameter Ag particle.

Conclusion

In this paper the optical properties of metallic nanoparticles have been modeled. Through these properties the localized surface plasmon resonance of certain nanoparticles embedded in the surrounding medium supporting for enhanced light absorption and emission have been observed. These models are very useful for optimal design of light absorber and emitter by using metallic nanoparticles.

References

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