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Optical Modeling of Plasmonic Nanoparticles Enhanced Light Emission of Silicon Light-Emitting Diodes

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Abstract Significant enhancement of radiative efficiency of thin-film silicon light-emitting diodes achieved by placing the active layer in close proximity to silver (Ag) nanoparticles has been observed. In this paper, optical properties including transmission, reflection, and absorption of a random assembly of Ag nanoparticles are theoretically investigated using the effective medium model. Furthermore, the influence of Ag nanoparticles on light emission of silicon light-emitting diodes is studied by an improved effective mode volume model we propose here. The normalized line shape of dipole oscillation is calculated directly using Lorentz–Drude model without using any approximation. Thus, it results in more accurate calculation of the enhanced Purcell factor in comparison with the conventional approach. We show that an enhancement of radiative efficiency of silicon light-emitting diodes can be achieved by localized surface plasmons on metal nanoparticles. The calculated result of optimal Ag nanoparticle size to enhance light emission of silicon light-emitting diodes at 900 nm wavelength is in very good agreement with those obtained from the experimental result. The model is useful for the design of metallic nanoparticles enhanced light emitters.

Keywords Plasmonic nanoparticles · Silicon light-emitting diodes

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Introduction

Silicon-on-insulator (SOI) light-emitting diodes (LEDs) are very promising light sources for optical communication technologies. However, the development of these components is hampered by their low efficiencies. Efforts to improve the limitations of the LEDs efficiency have, among others, made use of plasmonic nanostructures [1]. Large efficiency enhancement of luminescence was achieved by placing metal nanoparticles close to light emitters. This has attracted intense research interests devoted to demonstrating such an enhancement in various media [2, 3]. Since the surface plasmon resonance frequency is determined by various parameters such as shape, size, and distribution of nanoparticles, an accurate theoretical model is very important in design of nanoparticles enhanced light emitters.

There exist various methods developed to investigate the efficiency enhancement of electroluminescence. Among these, the recently introduced method based on the effective mode volume theory is known as a systematic and rigorous model to predict emission enhancement of quantum well LEDs in the vicinity of metal nanoparticles [4]. However, the method failed to explain the experimental results of an enhanced light emission of SOI-LEDs due to silver (Ag) nanoparticles. While the experiment demonstrated that a significant enhancement in electroluminescence from SOI-LEDs at 900 nm via excitation of surface plasmons in silver nanoparticles was obtained at the average particle size of a radius of 50–70 nm [1], those obtained by this approach (referred to as the conventional approach thereafter in this work) for the particle radius is only around 20–30 nm. This is attributed to the use of Drude approximation in determining the enhanced Purcell factor. To overcome this problem, the Lorentz–Drude model for metal dispersion, which is well-known as a more accurate model [5], combined with a

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calculation without approximations of the enhanced Purcell factor due to nanoparticles are proposed in this paper. The calculated results by the improved approach are shown to be in very good agreement with the experiment.

Furthermore, a random assembly of isolated nanoparticles may hold an advantage over the ordered arrays for light emitter devices of finite area [4]. In this paper, we present a theoretical model for the optical properties of such nanoparticles using the effective medium theory as means for optimal design of light emitters. The validity of the model is tested via a comparison to the Mie theory.

An Improved Model of Plasmonic Nanoparticles Enhanced Light Emitters

The theoretical model employed to evaluate a radiation enhancement of SOI-LEDs is probably described as a two-step process. The first step is dedicated to describe an energy transfer from a matter to surface plasmon polaritons (SPPs) mode at the metal–dielectric interface. Owing to the high mode density in the SPP, the Purcell factor $\boldsymbol{F}_p$ [6] is significantly enhanced. The second step is a combination of the energy from the confined SPP modes to actual propagation modes with a certain coupling rate, and this process competes with nonradiative loss due to absorption in metal nanoparticles. Typically, tightly confined large wave vector SPPs are more difficult to couple to the outside world. Hence, the overall radiative efficiency from the excited matter to the free-propagating wave has a complicated dependence on the SPP characteristics. It was shown that for a given original radiative efficiency, η_{rad}, there exists an exact value of SPP enhancement [4].

The geometry of isolated metal nanospheres being placed on top of SOI-LED is shown in Fig. 1. The effective volume of the SP mode supported by the metal sphere is given as follows [4]:

$$V_{\mathrm{eff}} = \frac{4}{3}\pi a^3\left(1+\frac{1}{2\varepsilon_D}\right) \tag{1}$$

with a being the radius of the nanoparticle and ε_D being the dielectric constant of the surrounding media.

For the dipole positioned at the distance d from the particle surface and oriented in z direction normal to the surface, the effective density of the SP modes is

$$\rho_{\mathrm{SP}} = \frac{L(\omega)}{V_{\mathrm{eff}}}\left(\frac{a}{a+d}\right)^6, \tag{2}$$

where the normalized line shape of the dipole oscillation is

$$L(\omega) = \frac{\mathrm{Im}\left[(\varepsilon_M(\omega)+2\omega_D)^{-1}\right]}{\int \mathrm{Im}\left[(\varepsilon_M(\omega)+2\omega_D)^{-1}\right]d\omega} \tag{3}$$

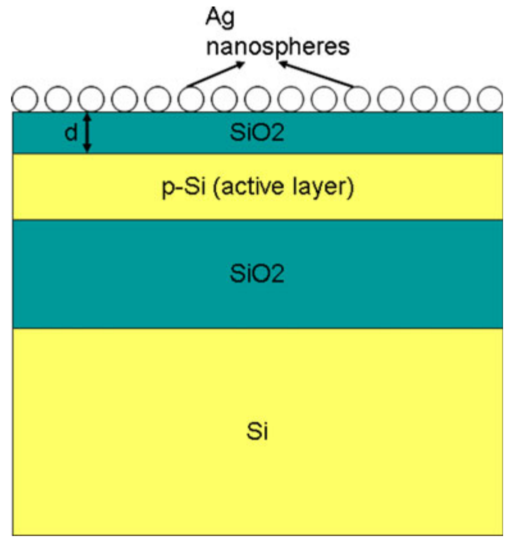

Fig. 1 Geometry of silicon-on-insulator LEDs with Ag nanospheres embedded on top

with $\varepsilon_M$being the metal dispersion. In [4], Eq. (3) was calculated by a Drude approximation. This is the main reason the method failed to explain the enhancement of the light emission from SOI-LEDs due to large nanoparticles. However, this can be remedied by considering the more accurate Lorentz–Drude model [5] combined with no approximation of the line shape of the dipole oscillation, which is performed in this work.

For Ag nanoparticles embedded in a silica ($\mathbf{SiO_2}$) medium, the enhanced fields that occur near metal particles as a result of SP resonance give rise to an enhanced absorption. According to a generalized form of Kirchoff's law which is valid for luminescent emission, enhanced absorption corresponds to an enhanced emission [1]. Figure 2 shows the scattering and absorption cross-sections for 100 nm diameter of Ag sphere embedded in

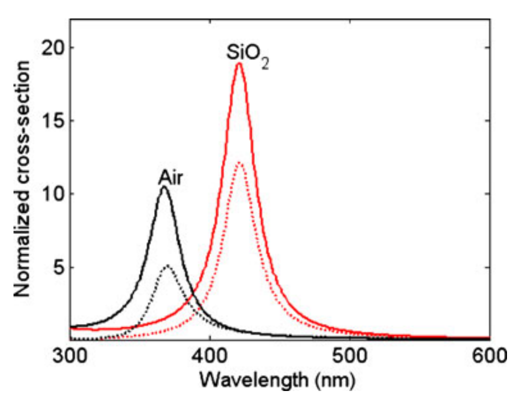

Fig. 2 Scattering (*solid lines*) and absorption (*dotted lines*) cross-sections for a 60-nm diameter Ag sphere embedded in air (*black*) and SiO_2 (*red*) normalized by the projected area of the sphere

air and $\mathbf{SiO_2}$ calculated by Mie theory [7]. Cross sections are normalized to the geometry particle cross section. For each embedded medium, a dipole resonance is observed. At resonance frequency ω_0, the Purcell factor $F_p(\omega_0)$can be estimated as a ratio of the effective density of the SP modes to that of the radiation components as follows:

$$F_p(\omega_0) = \frac{\rho_{\rm SP}}{\rho_{\rm rad}} = \frac{L(\omega_0)}{V_{\rm eff}}\left(\frac{a}{a+d}\right)^6\left[\frac{1}{3\pi^2}\left(\frac{2\pi}{\lambda_D}\right)^3\frac{1}{\omega_0}\right]^{-1}, \quad (4)$$

with $\lambda_D = \lambda/n$ being the emission wavelength in the dielectric.

Now, with given original radiative efficiency, the expression for the enhancement factor due to a single metal nanoparticle is described as [4]:

$$F_{\rm single} = \frac{\eta_{\rm SP}}{\eta_{\rm rad}} = \frac{1+F_p\eta_{\rm pr}}{1+F_p\eta_{\rm rad}}, \quad (5)$$

where $\eta_{\rm pr}$ is the radiative coupling efficiency of the SP mode. For an example of InGaN quantum well LEDs, with isolated silver nanoparicles placed in close proximity to the active region, it was shown that the output enhancement due to isolated silver nanoparticles is significant while only modest enhancement can be achieved with an ordered array of nanoparticles. It was shown that a random assembly of isolated particles may hold an advantage over the ordered arrays for light emitter devices of finite area [4].

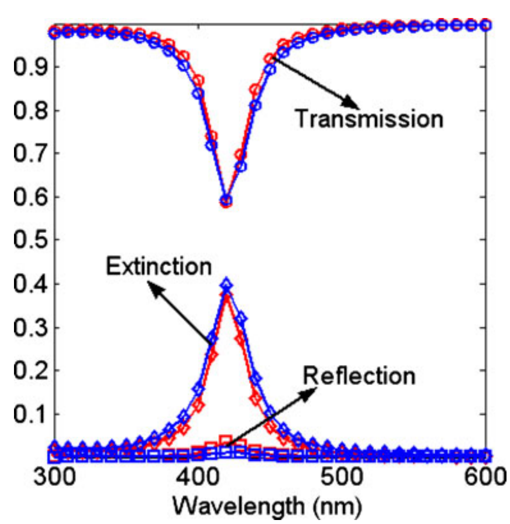

Fig. 3 Spectra of the transmission and reflection for slab of 60 nm diameter Ag particles embedded in SiO_2 calculated by the effective medium theory (*red lines*) and the Mie theory (*blue lines*)

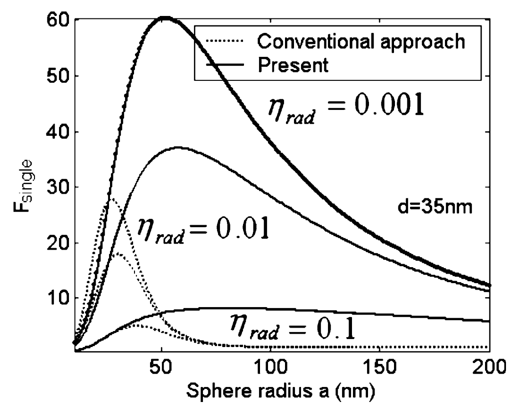

Fig. 4 Enhancement due to a single Ag sphere on SOI-LEDs with a distance of 35 nm from active layer to metal layer as a function of sphere radius for various original radiative efficiencies calculated by the conventional approach (*dotted lines*) and the present approach (*solid lines*)

Optical Properties of a Random Assembly of Nanoparticles

While the optical scattering and absorption efficiency determine the conversion of the incident light into the corresponding quantities, they give no information about the directionality of the scattered field. Through the transmission and reflection of light interacting with nanoparticles a better understanding of the process can be obtained. Using the effective medium theory the transmission and reflection of light through a random assembly of nanoparticles can be assumedly described by those 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.

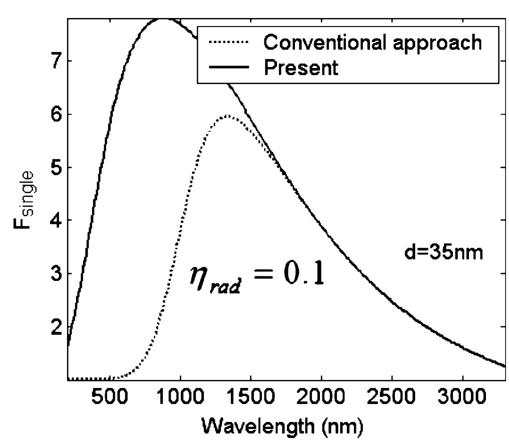

Fig. 5 Enhancement due to a single Ag sphere on SOI-LEDs with a distance of 35 nm from active layer to metal layer as a function of emission wavelengths calculated by the conventional approach (*dotted line*) and the present approach (*solid line*)

With a low volume fraction of isolated nanoparticles, the effective dielectric permittivity is given by

$$\varepsilon_{\mathrm{eff}} = \left[\frac{1+2f\gamma}{1-f\gamma}\right]\varepsilon_m \quad , \quad \gamma = \frac{\varepsilon_p-\varepsilon_m}{\varepsilon_p+2\varepsilon_m} \tag{6}$$

and the reflection and transmission of the slab of nanoparticles with an assumption of normal light incidence are calculated by

$$R = \left|\frac{r(1-\exp(ikn_{\mathrm{eff}}h))}{1-r^2\exp(ikn_{\mathrm{eff}}h)}\right|^2 \quad , \quad m = \frac{n_{\mathrm{eff}}}{n_m} \quad , \quad r = \frac{1-m}{1+m} \tag{7}$$

$$T = \left|\frac{4m}{(1+m)^2}\frac{\exp(-ikn_mh)}{[\exp(-ikn_{\mathrm{eff}}h) - r^2\exp(ikn_{\mathrm{eff}}h)]}\right|^2, \tag{8}$$

where k is the wave vector 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.

To validate the model the calculated transmission and reflection coefficients are compared to those obtained by the Mie theory. For small values of the surface coverage ($\eta \ll 1$), the transmission and reflection rates of the layer of nanoparticles can by calculated using the Mie cross-sections backward scattering and absorption efficiencies where multiple scattering has been neglected due to the low surface coverage [8]. Figure 3 shows the transmission and reflection coefficients (with η=0.02 and f=0.01) calculated by these two methods. From the figure, a good agreement between those methods is found as a mean to confirm the validation of the proposed model using the effective medium theory. The same figure shows that at the resonance frequency, the low transmission and reflection coefficients lead to the high extinction (scattering plus absorption) efficiency (E=1-T-R). That condition favors for an enhancement of radiative efficiency of LEDs.

Enhanced Light Emission by Isolated Nanoparticles

We now apply the developed model to evaluate the enhancement of the electroluminescence efficiency of silicon LEDs in which the active layer is placed in the vicinity of isolated Ag nanoparticles. The distance d from the active layer to Ag particle is 35 nm as in LED devices used in the experiments described in [9]. The calculated enhancement factor of the electroluminescence efficiency for a range of the original radiative efficiency of the emitter with respect to the sphere radius is shown in Fig. 4. We can see that the enhancement factor exhibits a strong dependence upon the nanosphere dimensions with the peak occurring when the radius is small enough to yield smaller effective mode volume for an enhanced Purcell factor, yet is still sufficiently large to assure strong radiative coupling of the SP mode. Furthermore, it is probably seen that the higher the original radiative efficiency, the more important grows the concern for the efficient energy transfer from the SP mode into free-space radiation modes. This situation favors larger nanoparticles that can emit the SP energy photons into the free space prior to get lost in the metal. At the emission wavelength of 900 nm, the enhancement of electroluminescence efficiency calculated by the present method can be found in range of the particle radius to be around 50–60 nm which is very good agreement with the experimental results reported in [9], whereas those obtained from the conventional approach is only 20–30 nm.

In addition, Fig. 5 shows the calculated enhancement factor of the electroluminescence efficiency with respect to emission wavelengths. It is seen that for a given original radiative efficiency of 0.1 the enhancement factor is observed at the emission wavelength of around 890 nm. It is in good agreement with those obtained by the experimental result of the emission wavelength at around 900 nm while those obtained by the conventional method is around 1,330 nm.

For a single nanosphere, although it may enhance light emission of only a very small emitter, an enhancement of light emission can be observed at certain optimal sizes. Therefore, with found optimal particle sizes, one can consider disordered arrays of these isolated nanospheres to achieve practical enhancement of such devices as SOI-LEDs.

Conclusions

The systematic and rigorous model based on the effective volume mode theory has been improved to evaluate the enhancement of the electroluminescence efficiency of SOI-LEDs due to isolated Ag nanoparticles. The improved model has well explained the plasmonic enhanced light emission of SOI-LEDs as reported in the experiment. In addition, the effective medium theory has been proposed for modeling optical properties of a random assembly of nanoparticles. Hence, it is very useful for the design of metal nanoparticles enhanced light emitters.

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