Practicable enhancement of spontaneous emission using surface plasmons

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The authors develop a rigorous theory of the enhancement of spontaneous emission from a light emitting device via coupling the radiant energy in and out of surface plasmon polaritons (SPPs) on the metal-dielectric interface. Using the GaN/Ag system as an example, the authors show that using SPP pays off only for emitters that have a low luminescence efficiency. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2539745]

Recently, there has been a great deal of interest in enhancing the efficiency of spontaneous emission (fluorescence) using the surface plasmon polariton (SPP) in the vicinity of a metal-dielectric boundary. The first definite sign of improvement was attained in GaN photoluminescence by placing a thin Ag film atop the GaN.[1] 90-fold enhancement of the spontaneous recombination rate in a similar structure was subsequently demonstrated.[2] Since then, SPP enhancement of spontaneous emission has been shown in a large number of different light emitting media,[3–7] including Si emitters.[8] The coupling of normally nonradiative SPPs into the radiation mode has been accomplished with one-dimensional (1D) dielectric gratings,[3,4,7] or two-dimensional (2D) corrugated silver films,[6,9] or more complicated cavity-like structures.[10]

With all the results reported, it is far from clear whether the schemes involved in the aforementioned experiments are actually optimal for the given emitters and collection optics. There were a number of analytical[11,12] and numerical calculations[10] that describe the SPP coupling into the radiation mode. Some of the calculations[12] completely disregard the issues of Ohmic losses in SPPs and thus give an overly optimistic prognosis for the SPP enhancement. Others[10] require extensive numerical modeling for each particular case. To the best of our knowledge, there has been no comprehensive study aimed at answering two simple questions: What should be the optimum configuration of the SPP structure for an emitter with a given radiative efficiency in a given light-collection geometry and what, if any improvement, will such an apparatus offer?

This work is aimed precisely at developing such a theory: a framework that provides unambiguous answers about the maximum improvement in radiative efficiency that one can expect using SPP and about the optimal configuration in which such improvement can be achieved. The theory relies only upon two parameters, namely, the intrinsic luminescence efficiency of the emitter and the imaginary part of the dielectric constant of a metal.

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Using the GaN/Ag material system as an example, we consider the geometry of a typical SPP enhancement scheme,[10–12] shown in Fig. 1. The emitting layer (GaN) has an internal radiative efficiency $\eta_{rad}=\tau_{nrad}/(\tau_{nrad}+\tau_{rad})$, where the radiative lifetime τ_{rad} is determined mostly by the density of modes in the dielectric. When the emitter is placed in the vicinity of the metal layer, there appears another decay channel—into the high-density SPP modes with a decay rate $\tau_{SPP}^{-1}=F_P\tau_{rad}^{-1}$ enhanced by the Purcell factor F_P.[13] Thus the energy is very efficiently transferred from the emitter into the SPP mode because of the large values of F_P, but in order for energy to be emitted, it has to be coupled into the continuum of radiation modes by a grating. This radiative process characterized by a coupling strength κ_{pr} competes with a nonradiatve dissipation due to the Ohmic loss determined by the imaginary part of the SPP propagation constant β_p''. When the nonradiative process becomes dominant, the overall SPP enhancement of radiation efficiency can be severely reduced or even negated.

The electric field of the SPP at the interface between a metal layer with a dielectric function ε_M and a dielectric medium with a dielectric constant ε_D can be written as $\mathbf{E}_p(z,x)=A_p(z)\mathbf{e}_p(\beta_p,x)e^{j(\beta_p z-\omega t)}$, where the SPP eigenmode

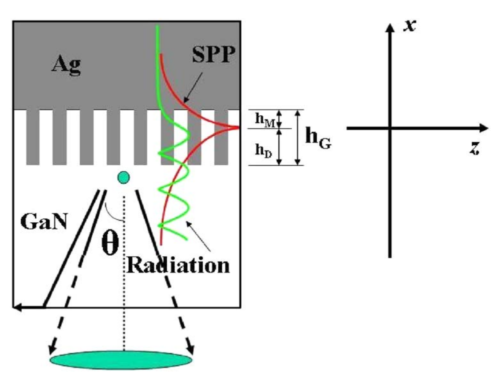

FIG. 1. (Color online) Light emission via coupling of SPP to radiation modes by a grating placed at the interface. The dot indicates an emitting center within the GaN layer.

$$\mathbf{e}_p(\beta_p,x) = \begin{cases} \frac{1}{\varepsilon}(j\beta_p\hat{\mathbf{x}} + q_M\hat{\mathbf{z}})e^{-q_Mx}, & x>0 \\ (j\beta_p\hat{\mathbf{x}} - q_D\hat{\mathbf{z}})e^{q_Dx}, & x<0 \end{cases} \quad (1)$$

and $\varepsilon=\varepsilon_M/\varepsilon_D$. Here wave vectors β_p, q_D, and q_M, related by $\beta_p^2-q_D^2=1$ and $\beta_p^2-q_M^2=\varepsilon$, are all normalized to $k_D=\sqrt{\varepsilon_D}\omega/c$, while coordinates x and z are normalized to $1/k_D$, and are therefore all dimensionless. The SPP dispersion of the real (solid line) and imaginary parts (dashed line) of the propagation constant $\beta_p=\beta_p'+j\beta_p''=\sqrt{\varepsilon/(1+\varepsilon)}$ shown in Fig. 2 is obtained using the actual complex dielectric constant for Ag (Ref. 14) as well as the dispersion of index of refraction for GaN (Ref. 15) The propagation constant β_{p0} (dotted line) is obtained under the assumption of no loss: $\mathrm{Im}(\varepsilon_M)=0$. As one can see, the presence of loss in the metal not only engenders a substantial imaginary part β_p'' of the propagation constant, but also limits the maximum attainable value of the real part β_p' and, consequently, the Purcell factor F_P (Fig. 2), which is proportional to the density of SPP modes $\rho_p=\beta_p/(2\pi w_p v_g)$, where $v_g=\partial\omega/\partial\beta_p$ is the group velocity, and w_p is the SPP effective width. If loss is disregarded, huge Purcell factors[2,12] can be obtained when group velocity becomes very small at large β_p'. But with real metal, even Ag with low loss, the maximum Purcell factor $F_{P,\max}\approx 10^3$ is limited by the minimum value of the group velocity $v_g\approx 2\times 10^6$ m/s.

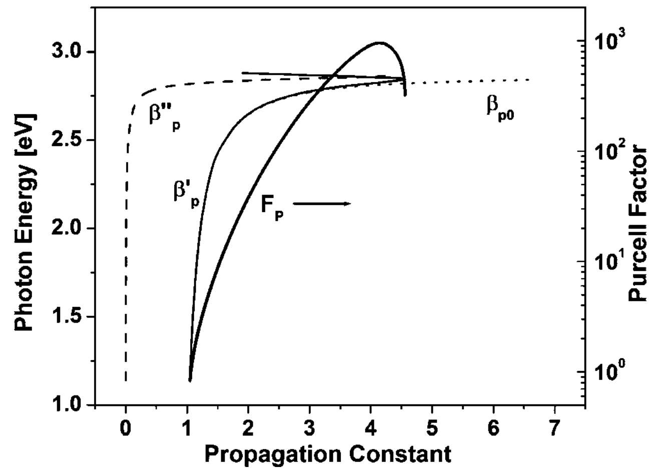

FIG. 2. SPP dispersion curve and Purcell factor F_P.

The electric field of TM radiation mode on the metal-dielectric boundary can be expressed as $\mathbf{E}_r(z,x)=A_r(z)\mathbf{e}_r(\beta_r,x)e^{j(\beta_r z-\omega t)}$, where the radiation eigenmode

$$\mathbf{e}_r(\beta_r,x) = \begin{cases} (j\beta_r\hat{\mathbf{x}} + q_r\hat{\mathbf{z}})e^{-q_rx}, & x>0 \\ j\beta_r[\varepsilon\cos(k_rx) - (q_r/k_r)\sin(k_rx)]\hat{\mathbf{x}} + [\varepsilon k_r\sin(k_rx) + q_r\cos(k_rx)]\hat{\mathbf{z}}, & x<0, \end{cases} \quad (2)$$

where $\beta_r^2+k_r^2=1$ and $\beta_r^2-q_r^2=\varepsilon$. In order for the SPP mode to couple into this radiation mode, either 1D or 2D grating is necessary to match their in-plane propagation constants $\mathbf{G}=\boldsymbol{\beta}_p-\boldsymbol{\beta}_r$ (Fig. 3). To achieve the most efficient coupling into the radiation mode normal to the surface, it is desirable to have the magnitude of $\mathbf{G}$ equal to that of $\boldsymbol{\beta}_p$. The SPP modes are located on the circumference of a ring with a radius $\beta_p>1$ while the radiation modes are confined within a smaller circle with a radius of 1. To obtain coupling between the discrete SPP state and the radiation continuum, we first evaluate the coupling between the SPP mode [Eq. (1)] and one of the radiation modes [Eq. (2)], and then integrate over the 1D density of radiation modes $\rho_r(\beta_r,\omega)=1/2\pi\omega\sqrt{1-\beta_r^2}$ to obtain the following equation for the radiative decay of the SPP mode:

$$\frac{\partial}{\partial z}|A_p(z)|^2 = -\kappa_{\mathrm{pr}}(\beta_p,\beta_r)|A_p(z)|^2, \quad (3)$$

where the coupling strength is

$$\kappa_{\mathrm{pr}}(\beta_p,\beta_r) = \frac{\pi\omega^2\rho_r(\beta_r,\omega)}{8v_g}f_G^2\left|\int \delta\varepsilon(x)\mathbf{e}_p(x)\mathbf{e}_r^*(x)dx\right|^2, \quad (4)$$

where the in-plane ($x=0$) Fourier amplitude $f_G=1/\pi$ for the 1D grating and $f_G=0.087$ for the 2D grating, $\delta\varepsilon(x)$ is the perpendicular (along x axis) dielectric variation of the grating, and the integral is evaluated over the grating height h_G.

The radiative efficiency of a given SPP mode propagating in the direction defined by an in-plane angle φ is then $\chi_{\mathrm{pr}}(\varphi)=\kappa_{\mathrm{pr}}/(\kappa_{\mathrm{pr}}+2\beta_p'')$. It is clear from Fig. 3 that SPP modes propagating in different directions do have different coupling efficiencies. Depending on the method of collection, the overall radiation efficiency should be integrated within an emission cone (Fig. 1) with a maximum collection angle $\theta_m=\sin^{-1}\beta_{r,\max}$, where the maximum radiation propagation constant $\beta_{r,\max}$ is related to the in-plane angle $\varphi_m=2\sin^{-1}(\beta_{r,\max}/2\beta_p)$ (Fig. 3). The overall radiation efficiency should therefore be evaluated as $\eta_{\mathrm{pr}}=N_G/2\pi\int_{-\varphi_m}^{\varphi_m}\chi_{\mathrm{pr}}(\varphi)d\varphi$, where $N_G=2$ for the 1D grating and $N_G=6$ for the 2D grating. We finally obtain the overall efficiency of the SPP-assisted emission process,

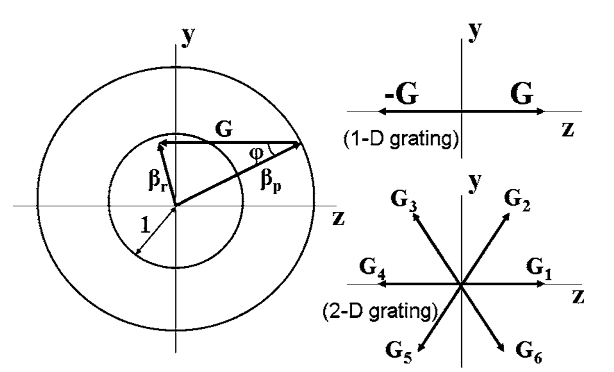

FIG. 3. SPP–radiation coupling by 1D and 2D basic grating vectors $\mathbf{G}$ in reciprocal space.

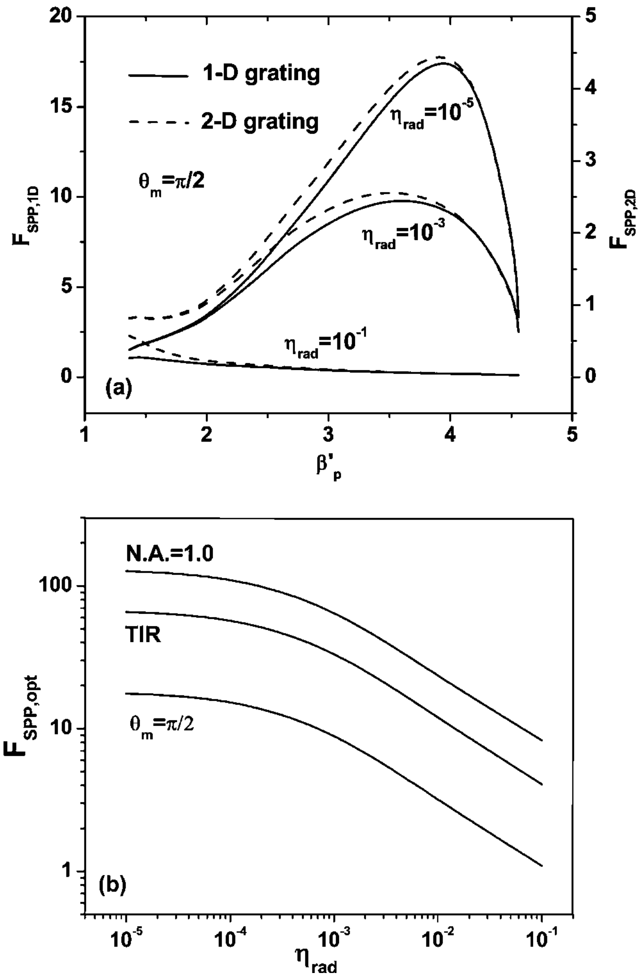

FIG. 4. (a) SPP enhancement factor by 1D and 2D gratings with $\theta_m=\pi/2$. (b) The optimal SPP enhancement by 1D grating as a function of the original radiative efficiency of the emitter at optimal value $\beta'_{p,\mathrm{opt}}$.

$$\eta_{\mathrm{SPP}} = \eta_{\mathrm{pr}} \frac{F_P \tau_{\mathrm{rad}}^{-1}}{F_P \tau_{\mathrm{rad}}^{-1} + \tau_{\mathrm{nrad}}^{-1}}, \tag{5}$$

and the enhancement due to SPP

$$F_{\mathrm{SPP}} = \frac{\eta_{\mathrm{SPP}}}{\eta_{\mathrm{rad}}} = \frac{\eta_{\mathrm{pr}}}{\eta_{\mathrm{rad}} + (1-\eta_{\mathrm{rad}})F_P^{-1}} < \frac{\eta_{\mathrm{pr}}}{\eta_{\mathrm{rad}}}. \tag{6}$$

Thus, no matter how high the Purcell factor, in the end, the SPP enhancement F_{SPP} is limited by the ratio of the SPP's radiative coupling efficiency to the original efficiency of the emitting source.

We have considered three different cases of the collection methods that are limited by different maximum output emission angles θ_m. First, we assume that all radiation modes escape the dielectric eventually by avoiding total internal reflection with the use of surface roughness or domed structures ($\theta_m=\pi/2$). Second, we look at the situation in which the angle of the emission cone is limited by the total internal reflection ($\theta_m=\sin^{-1}(1/\sqrt{\varepsilon_D})$). Finally, for the case in which the emission cone is limited by the numerical aperture (NA) of collection lens, the maximum emission angle can be given as $\theta_m=\sin^{-1}(\sin[\tan^{-1}(\mathrm{NA}/2)]/\sqrt{\varepsilon_D})$.

In Fig. 4(a) we show the SPP enhancement obtained using 1D and 2D gratings with $\theta_m=\pi/2$ as a function of the SPP propagation constant. The enhancement factor clearly experiences a peak at some optimal values $\beta'_{p,\mathrm{opt}}$ for a given original radiative efficiency η_{rad} and that optimum value depends on the original radiation efficiency. The 1D grating consistently outperforms 2D grating by a factor close to 4 due to the smaller value of 2D Fourier amplitude f_G.

The main result of our work—the optimal enhancement factor $F_{\mathrm{SPP,opt}}=\eta_{\mathrm{SPP}}(\theta_m,\beta'_{p,\mathrm{opt}})/\eta_{\mathrm{rad}}(1-\cos\theta_m)$ evaluated at optimal values of the propagation constant $\beta'_{p,\mathrm{opt}}$ for a wide range of η_{rad}—is shown in Fig. 4(b) for the three different cases of collection methods with the 1D grating. For a fair comparison between the different collection methods, we have factored in $\Omega/2\pi=1-\cos\theta_m$, where Ω is the solid angle of the collection cone for either the total internal reflection or lens collection, assuming a rear reflector is used in the absence of the metal layer. Overall, significant enhancement is attainable only with a low original radiative efficiency. For instance, when collection is limited by the total internal reflection and $\eta_{\mathrm{rad}}=5\%$, an eightfold improvement is feasible—which is in excellent agreement with the experimental observation in the same InGaN/Ag material system.[3,4] Once the original radiative efficiency surpasses 10%, no SPP enhancement efficiency is feasible.

In summary, we have obtained easy-to-interpret analytical results that unequivocally indicate that SPP enhancement of spontaneous radiation is most noticeable only if the original radiative efficiency of the emitter is very small, less than 1%. Even then, the SPP enhancement is not substantially higher than ten fold. For this reason, it does not appear that SPP offers any advantage for light emitting diodes, with the only possibly exception of Si emitters whose original radiative efficiency is very low. The main application of SPP enhancement should remain as improving the efficiency of weak photoluminescence and nonlinear processes for the purpose of detecting small amounts of different substances.

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