Tunable surface plasmons in coupled metallo-dielectric multiple layers for light-emission efficiency enhancement

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The coupling of spontaneously emitted photons to surface plasmons in metal films is a promising technique to increase the efficiency of light-emitting devices. Here we propose and theoretically investigate the use of metallo-dielectric multiple layers to engineer the surface-plasmon density of states, so as to introduce tunable resonances in the emission efficiency through the anticrossing of modes localized on neighboring interfaces. To illustrate, large enhancements in the radiative recombination rate at tunable wavelengths are predicted for a GaN-based light-emitting device, using a $Ag/Si_3N_4/Au/Si_3N_4$ structure with different combinations of the layers thicknesses.
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Solid-state sources of visible light, such as semiconductor and organic light-emitting diodes (LEDs), are the subject of considerable interest for many applications, including ultimately the replacement of light bulbs for white lighting. Towards this goal, several techniques are being investigated to enhance both their internal quantum efficiency and their light extraction efficiency. An approach that is attracting a lot of attention is the use of surface plasmons (SPs) to increase the radiative recombination rate.[1–8] SPs are collective charge oscillations at the interface between a metal and a dielectric; the coupling of light to these oscillations results in guided polariton modes that are confined at, and propagate along, the interface.[9] Due to the tightly bound nature of these modes, near the interface their fields are highly enhanced compared to radiative modes. Thus, if a metal film is deposited in close proximity of an LED active layer, the device quantum efficiency is correspondingly increased through emission of SP polaritons at the metal surface. These guided modes can then be converted into radiative waves (the useful output of the LED) by means of a grating, or even by the roughness of the metal film.[8]

Recently, large enhancements of the photoluminescence efficiency have been demonstrated in silver-coated organic[7] and GaN (see Ref. 8) devices based on SP-mediated light emission. However, for this approach to be effective, the LED emission frequency should be closely matched to the SP resonance frequency ω_{SP}, where the SP density of states (SP-DOS) is maximum. For a single planar metallic overlayer, this frequency is entirely determined by the dielectric functions of the metal and emitter material; if the emission frequency differs from ω_{SP} the SP efficiency enhancement is reduced, as was observed in Ref. 8. Thus, the practical development of SP-enhanced LEDs will require a technique to effectively tune this resonance frequency.

A possible technique is the use of metallic gratings instead of planar films, to break up the SP dispersion relation into a series of bands.[3,5] Through a careful choice of the grating period, this can lead to a band edge—and hence an increased SP-DOS—in the wavelength region of interest. Tunable SPs have also been demonstrated using metallic nanospheres and nanoshells,[10,11] whose geometry allows for extremely wide tuning ranges. However, all these approaches require a precise control of the metal features size and shape, leading to demanding fabrication processes. Here we propose a different technique based on coupled SPs in metallo-dielectric multiple layers. Through the careful choice of the layers thicknesses, this can also lead to tunable singularities in the SP-DOS, in this case through the hybridization and anticrossing of the SP dispersion curves of neighboring interfaces. Metallo-dielectric multiple layers have already been studied as promising photonic band-gap materials,[12] but their use to engineer the SP-DOS has not yet been explored.

To introduce the proposed technique we first consider a single metallic layer, for example, silver, on a GaN-based LED, as shown in the inset of Fig. 1(a). Also shown in this figure are the SP dispersion curves $\hbar\omega(k)$—where k is the in-plane wave vector—for different thicknesses of the Ag film. These were computed by solving Maxwell's equations in the GaN substrate, Ag film, and air, and matching the solutions with the electromagnetic boundary conditions. A simple Drude model was used for the dielectric function of Ag; that is, $\varepsilon(\omega)=\varepsilon_\infty[1-\omega_p^2/(\omega^2+i\gamma\omega)]$, with $\hbar\omega_p=3.76$ eV and $\varepsilon_\infty=9.6$ extrapolated from experimental data[4,13] (γ is sufficiently small to have negligible effect on the SP dispersion[9]). For each thickness of the Ag film two disper-

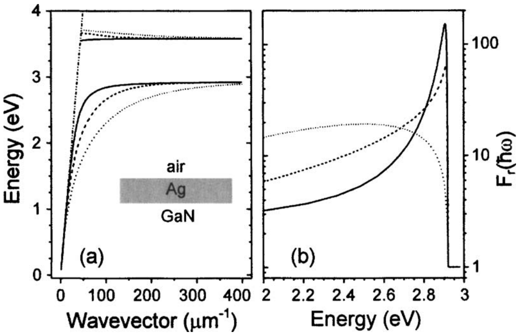

FIG. 1. (a) Dispersion curves of the bound SP polaritons of the structure shown in the inset, for Ag thicknesses of 100 nm (solid curve), 10 nm (dashed curve), and 5 nm (dotted curve). The dash-dotted straight line is the light line in GaN. (b) Spectra of the radiative recombination rate ratio corresponding to the dispersion curves of (a).

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sion curves are obtained, one (at lower frequencies) for the SPs at the GaN/Ag interface, the other for the SPs at the Ag/air interface. As the film thickness is decreased, the solutions at the two interfaces couple to each other leading to symmetric and anti-symmetric hybrid modes,[3,4,6,7] and correspondingly the frequency separation between the two dispersion curves increases.[14]

A simple figure of merit that can be used to quantify the SP-induced LED efficiency enhancement is the radiative recombination rate ratio[4,6]

$$F_r(\hbar\omega) = \frac{\Gamma_0(\hbar\omega) + \Gamma_{\mathrm{SP}}(\hbar\omega)}{\Gamma_0(\hbar\omega)}, \tag{1}$$

where Γ_0 and Γ_{SP} are the spontaneous emission rates into radiation modes and SP polaritons, respectively (F_r is the same as the Purcell factor in the limit of negligible nonradiative recombination). Γ_{SP} can be calculated using Fermi golden rule[4,6]

$$\Gamma_{\mathrm{SP}}(\hbar\omega) = \frac{2\pi}{\hbar}|\mathbf{d}\cdot\mathbf{E}_{\mathbf{act}}(\hbar\omega)|^2\left[\frac{A}{4\pi}\frac{d(k^2)}{d(\hbar\omega)}\right], \tag{2}$$

where $\mathbf{d}$ is the dipole moment matrix element, which will be assumed to be isotropic for simplicity; the quantity in brackets is the SP-DOS on a surface of area A; and $\mathbf{E}_{\mathbf{act}}(\hbar\omega)$ is the electric field of the SP mode of frequency ω, normalized to the vacuum fluctuation energy $\hbar\omega/2$, and evaluated at the location of the active layer. To compute $\mathbf{E}_{\mathbf{act}}(\hbar\omega)$, a separation of 10 nm between the active layer and the metal film, for example, due to a GaN cap layer, will be used in all the calculations presented in this article. Finally, Γ_0 is computed using the classical formula[15]

$$\Gamma_0(\hbar\omega) = \frac{4nd^2\omega^3}{3\hbar c^3}, \tag{3}$$

where n is the refractive index of the emissive material.

This model is based on several simplifying assumptions, and in particular it does not account for the complexities of semiconductor active layers, nor for the broadening of $\Gamma_{\mathrm{SP}}(\hbar\omega)$ due to damping of the electronic motion in the metal. On the other hand, it has the advantage of requiring a minimal set of input parameters and thus it provides a very convenient design tool. In fact, after substitution of Eqs. (2) and (3) into Eq. (1), F_r only depends on the layers' dielectric functions and thicknesses through $\mathbf{E}_{\mathbf{act}}$ and $\omega(k)$. From a device perspective, the more important parameters are the LED internal efficiency with and without SP enhancement. These are given by $\eta=(\Gamma_0+\Gamma_{\mathrm{SP}})/(\Gamma_0+\Gamma_{\mathrm{SP}}+\Gamma_{\mathrm{NR}})$ and $\eta_0=\Gamma_0/(\Gamma_0+\Gamma_{\mathrm{NR}})$, respectively, where Γ_{NR} is the nonradiative recombination rate and unit probability of SP conversion into radiation modes has been assumed. Eliminating Γ_{NR} from these two expressions and using Eq. (1) yields

$$F_r = \frac{\eta}{1-\eta}\frac{1-\eta_0}{\eta_0}. \tag{4}$$

Thus, for example, a value of $F_r=100$ corresponds to an efficiency enhancement from $\eta_0=10\%$ to $\eta=92\%$, or from $\eta_0=50\%$ to $\eta=99\%$, and so forth.

Plotted in Fig. 1(b) are the calculated spectra of F_r corresponding to the dispersion curves of Fig. 1(a). As shown by the solid line, for a thick (100 nm) Ag film, F_r reaches a maximum value of ≈160 near the asymptotic energy $\hbar\omega_{\mathrm{SP}}$ (≈2.9 eV for Ag on GaN), where the SP-DOS diverges. At energies above $\hbar\omega_{\mathrm{SP}}$, the interface no longer supports SP guided modes and as a result F_r sharply decreases to unity. At lower energies, a gradual reduction is observed, due to the increasing slope of the dispersion curve $\omega(k)$—and hence decreasing SP-DOS—with decreasing energy. These results are in general agreement with previously published calculations and experimental data.[4,6,8] If a thinner Ag film is used, the dispersion curve of the SPs at the GaN/Ag interface is pushed to lower energies by the coupling with the SPs at the Ag/air interface (while the asymptotic energy $\hbar\omega_{\mathrm{SP}}$ remains unchanged[14]), and correspondingly the spectrum of F_r is broadened. Thus, as shown by the dotted and dashed lines in Fig. 1(b), thinner metal films can be used to increase the efficiency enhancement at energies below $\hbar\omega_{\mathrm{SP}}$. However, this approach leads to only a modest gain in the SP-DOS and hence in F_r, and therefore it is not effective in tuning the SP resonance, unlike, for example, in the case of metal nanoshells.[11]

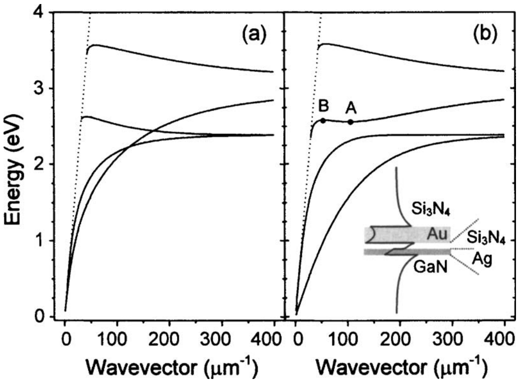

FIG. 2. Dispersion curves of the bound SP polaritons of the structure shown in the inset. The thicknesses of the intermediate Ag/Si$_3$N$_4$/Au layers are 5/100/14 (a) and 5/5/14 nm (b). Both the GaN substrate and the Si$_3$N$_4$ overlayer are taken to be infinitely thick. The dotted straight line in each plot is the light line in GaN. Also shown in the inset is the transverse field component of the SP mode at point A.

To obtain more significant enhancements at tunable photon energies we propose to use multiple metallo-dielectric layers. The general idea is to introduce singularities in the SP-DOS at the energies of interest through the anticrossing of SP modes of different metal films. To illustrate, we consider the structure shown in the inset of Fig. 2(b) consisting of a (5 nm) Ag film deposited over the LED, followed by a (14 nm) Au film sandwiched between two Si$_3$N$_4$ layers. This structure contains four metallo-dielectric interfaces and therefore supports four SP branches, plotted in Figs. 2(a) and 2(b) for a thickness of the intermediate Si$_3$N$_4$ layer of 100 and 5 nm, respectively ($\hbar\omega_p=2.87$ eV and $\varepsilon_\infty=8.9$ for Au were used in these calculations).[8,13] In order of increasing asymptotic energy, these branches are associated with the two Au/Si$_3$N$_4$ interfaces, the Ag/GaN interface, and the Ag/Si$_3$N$_4$ interface, respectively.

In Fig. 2(a), the Si$_3$N$_4$ layer between the two metals is thick relative to the SP decay length in Si$_3$N$_4$. Thus, the SP modes of the two metals are essentially uncoupled from each other and their dispersion curves can cross. On the contrary, in Fig. 2(b) the fields of these modes strongly overlap through the thin intermediate Si$_3$N$_4$ layer, leading to hybrid solutions near the crossing point. Therefore, an anticrossing behavior is observed which causes a flattening of the disper-

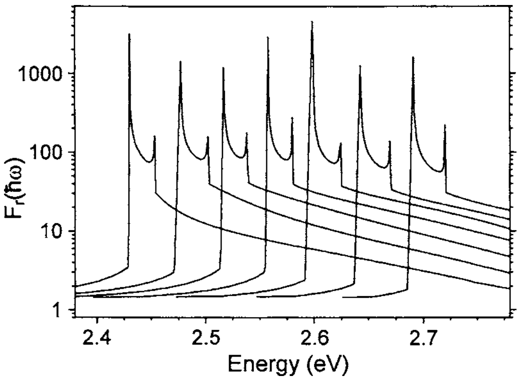

FIG. 3. Spectra of the radiative recombination rate ratio F_r of the GaN/Ag/Si_3N_4/Au/Si_3N_4 structure shown in the inset of Fig. 2, for different thicknesses of the intermediate Ag/Si_3N_4/Au layers. From left to right, these thicknesses are (in nm) 4/9/25, 4/7/20, 5/6/17, 5/5/14, 5/4/11, 6/4/9, and 6/4/6.

sion curves, and hence singularities in the SP-DOS (proportional to $dk/d\omega$), on both sides of the anticrossing. At the point labeled A in the figure, this singularity is also accompanied by a relatively large SP field at the LED active layer (the transverse component of this field is plotted in the inset). Thus, according to Eqs. (1) and (2), F_r is expected to be large in the spectral vicinity of point A. We emphasize that this spectral region can be tuned over a wide range by varying the metals thicknesses, which determine the location of the crossing point, and the intermediate Si_3N_4 thickness, which determines the size of the anticrossing.

The calculated spectrum of F_r for the structure of Fig. 2(b) is plotted in Fig. 3 (fourth curve from the left). Two sharp peaks are observed in this curve, corresponding to the points A and B in Fig. 2(b)—the latter being another point of singular SP-DOS and, in the presence of coupling, of a sizable SP field at the LED active layer. In the spectral region of these two peaks, F_r remains large over a range of several 10s meV; for example, if averaged over an LED bandwidth of 100 meV its maximum value is 79, corresponding to an increase in internal efficiency from, for example, 10% to 90% according to Eq. (4). This is comparable to the case of a single thick Ag layer [solid line in Fig. 1(b)], for which such maximum integrated value is 76, but at a different wavelength. Experimentally, the spectra of F_r will be broader than these curves by several tens of meV due to ohmic losses in the metals.[4] However, the above quoted efficiency enhancements integrated over a wide bandwidth will not be significantly altered by such broadening.

To illustrate the tunability allowed by this approach, the other curves in Fig. 3 correspond to the same multiple-layer structure but with different values of the layers thicknesses (listed in the caption). As shown, a tuning range of at least 300 meV is readily covered, with very similar spectra of F_r, and at energies far removed from the asymptotic SP energies of both the GaN/Ag and the GaN/Au interfaces ($\approx$2.9 and 2.2 eV, respectively). Various other wavelength regions of interest can be similarly accessed using different dielectrics and/or metals. More complex structures—for example, involving more than two metallic films—can also be designed to further optimize the SP-DOS for LED efficiency enhancement or other applications.

Finally, we address the issue of coupling the emitted SP polaritons into radiation modes. As shown in Ref. 8, light can be extracted quite efficiently from the SPs by the submicron roughness on the metal surface. Similar results can be expected in the proposed structures with intentionally introduced roughness in the uppermost dielectric layer. A more reliable method is the use of gratings in the metal film or upper dielectric layer. While a grating can also be used to tune the SP resonance,[3,5] one advantage of the present approach is that no stringent condition is imposed on its period. This could significantly simplify the device fabrication; alternatively, the grating may be designed to separately optimize the directionality of the emitted beam[1] and hence to maximize the LED light-extraction efficiency.

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[14] In the limit where ck/ω diverges, the frequency of each dispersion curve approaches its asymptotic value ω_{SP}, and the penetration depth of the SP modes in the metal becomes negligibly small (see Ref. 9). As a result, in this limit SPs at different interfaces do not couple to each other regardless of the metal film thickness, and therefore their asymptotic frequencies do not vary with metal thickness.
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