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Surface plasmon dispersion engineering via double-metallic Au/Ag layers for III-nitride based light-emitting diodes

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Double-metallic Au/Ag layers deposited on top of InGaN/GaN quantum wells (QWs) are used to tune the Purcell peak enhancement of the radiative recombination rate for nitride light-emitting diodes. By modifying the Au/Ag thicknesses, the Purcell factor can be widely tuned between the surface plasmon frequencies of Au/GaN and Ag/GaN. Photoluminescence studies demonstrated the concept of the Purcell factor tuning by using the double-metallic Au/Ag layers. © *2011 American Institute of Physics*. [doi:10.1063/1.3580628]

III-nitride semiconductors have applications for solid state lighting and lasers,[1–10] power electronics,[11] thermoelectricity,[12–14] and solar cells.[15–18] The InGaN quantum wells (QWs) have been employed as active region in nitride light-emitting diodes (LEDs).[19–32] The internal quantum efficiency (IQE) in InGaN QWs LEDs is limited by the following: (1) high dislocation density leading to large nonradiative recombination rate, and (2) charge separation in the QW leading to reduction in the radiative recombination rate (R_{Rad}). Several approaches have been demonstrated to suppress the charge separation issue by employing QWs with improved electron-hole wave function overlap (Γ_{e_hh}).[19–32]

Another approach to enhance the R_{Rad} and IQE of InGaN QWs is by using surface-plasmon (SP) LEDs with single Ag metallic layer,[33–36] which is based on the increase in photon density of states near the SP frequency (ω_{sp}) from Purcell factor. The use of single metallic layer leads to strong enhancement near the ω_{sp}, and the enhancement will reduce for frequency further away from the ω_{sp}, with no enhancement obtained for frequency above the ω_{sp}.

Recent interesting approach based on metallodielectric stacked structures[37,38] had been proposed to tune the SP frequency. However, the Purcell factor based on this approach reduces for the frequency regime away from SP frequency of metal/GaN. Our approach based on the double-metallic layers can be practically implemented by single metallic deposition step.

In this letter, we present an approach to achieve wide-spectrum tuning of the SP resonant frequency by employing the double-metallic layers on GaN. The use of double metallic Au/Ag layers enables the tuning of the SP frequency over a large frequency range between the SP frequencies of the individual metals ($E_{sp\_Ag}=\hbar\omega_{sp\_Ag}$ and $E_{sp\_Au}=\hbar\omega_{sp\_Au}$) while maintaining a large Purcell factor throughout the frequency range. Photoluminescence (PL) studies were carried out as the proof-of-concept experiments for the Purcell factor tuning by using the double-metallic concept.

Figure 1 shows the SP dispersion curves of thin Ag (or Au) film on GaN substrate as a function of the Ag (or Au) thicknesses of 40, 10, and 5 nm. From Fig. 1, a single metal film deposited on GaN results in a particular SP frequency (i.e., $E_{sp\_Ag}=2.8$ eV; $E_{sp\_Au}=2.2$ eV), which cannot be tuned by modifying the single metal film thickness, although the dispersion relation shows different trend for different metal thicknesses.

The use of double-metallic layers (Au/Ag), with varying thickness combinations of Au (d_{Au}) and Ag (d_{Ag}), allows the Purcell factor tuning between the SP frequencies of Ag/GaN (ω_{sp_Ag}) and Au/GaN (ω_{sp_Au}). Figure 2(a) shows the SP dispersion curves of Au/Ag layers on GaN with d_{Au}/d_{Ag} values of 0 nm/20 nm (Ag-only), 10 nm/10 nm, 5 nm/15 nm, 3 nm/17 nm, 1 nm/19 nm, and 20 nm/0 nm (Au-only). The total thicknesses of the double metallic layers are kept constant ($d_{total}=20$ nm) in our analysis for comparison purpose. By modifying the ratio of the Au and Ag thicknesses, the dispersion curve can be engineered with different SP frequencies between ω_{sp_Ag} and ω_{sp_Au}.

Figure 2(b) plots the Purcell factor as a function of energy for double-metallic Au/Ag layers on a GaN substrate with Au/Ag layer thicknesses as described in Fig. 2(a). Figure 2(b) indicates that the Purcell factor can be tuned between the ω_{sp_Ag} and ω_{sp_Au} without decreasing the Purcell factor. Note that the ratio of the double metallic Au/Ag film thicknesses determines the SP resonant frequency (ω_{sp}). Thus, it is crucial to optimize the Au layer thickness to tune the ω_{sp} between ω_{sp_Ag} and ω_{sp_Au}.

To better illustrate the effect of the sequence of the double-metallic layers on the coupling between the QW and

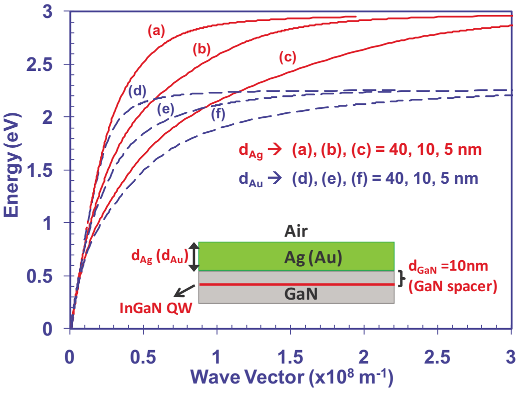

FIG. 1. (Color online) SP dispersion curves of thin Ag (Au) film on GaN substrate with Ag (Au) film thicknesses of 40, 10, and 5 nm.

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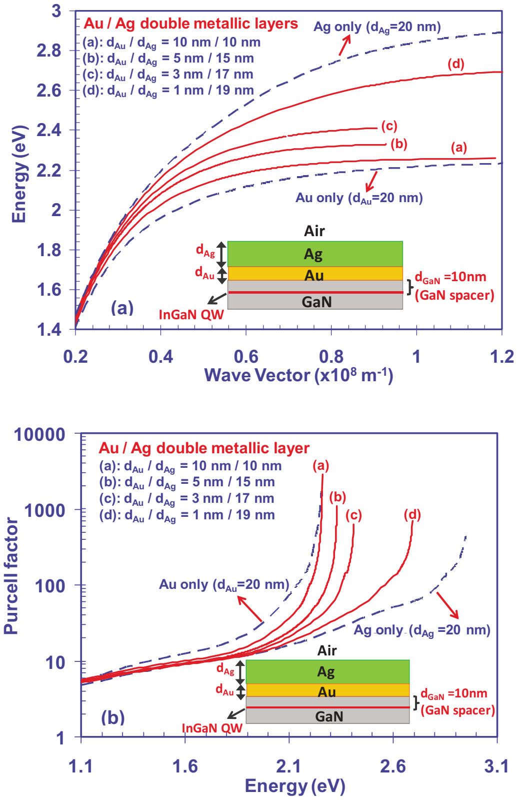

FIG. 2. (Color online) (a) SP dispersion curves and (b) Purcell factor as a function of energy for double-metallic Au/Ag layers on GaN substrate with Au/Ag layer thicknesses of 10 nm/10 nm (E_{sp}=2.25 eV), 5 nm/15 nm (E_{sp}=2.33 eV), 3 nm/17 nm (E_{sp}=2.41 eV), and 1 nm/19 nm (E_{sp}=2.69 eV). The corresponding SP dispersion curves and Purcell factors for Ag-only/GaN (d_{Ag}=20 nm, E_{sp}=2.88 eV) and Au-only/GaN (d_{Au}=20 nm, E_{sp}=2.23 eV) are also plotted.

the SP electric field, Figs. 3(a) and 3(b) shows the electric field for the double-metallic layers of Au (3 nm)/Ag (17 nm) and Ag (3 nm)/Au (17 nm) on InGaN/GaN, respectively. The SP electric field of Au/Ag double-metallic layers is strongly coupled to the InGaN QW, while the SP electric field of Ag/Au double-metallic layers shows almost no coupling to the InGaN QW. From Fig. 3(a), the SP electric field interacts with both Au and Ag films, which results in an average SP resonant energy of Au/GaN and Ag/GaN.

Experimental studies were conducted to demonstrate the concept of the SP frequency tuning using the double-metallic layers on four-period InGaN/GaN QWs samples (λ_{peak} $\sim$465 nm) grown by metalorganic vapor phase epitaxy on u-GaN (3 μm)/double-side polished sapphire. On top of the last QW, the GaN spacer thickness is 10 nm. The metals with total thickness of 50 nm were deposited on the QW samples by thermal evaporation. PL measurements were conducted by exciting the QWs with a 410 nm InGaN laser from the bottom of the substrate [Fig. 4(a)], and silicon photodetector was used to collect the emission from the sample. PL measurements were performed for InGaN QWs deposited with different metallic layers as follows: (1) 50 nm Ag, (2) 2 nm Au/48 nm Ag, (3) 4 nm Au/46 nm Ag, (4) 6 nm Au/44 nm Ag, (5) 8 nm Au/42 nm Ag, (6) 10 nm Au/40 nm Ag, (7) 12 nm Au/38 nm Ag, and (8) 50 nm Au. The PL measurements for these samples were compared with the corresponding control InGaN QWs without metal deposition.

The surface roughnesses of the metallic layers are keys for coupling the SP mode to the radiation mode. The atomic force microscopy results show the rms surface roughnesses of Au-only (50 nm) and Au (8 nm)/Ag (42 nm) films as 3.5 nm and 2.7 nm, respectively (area=5 μm$\times$5 μm).

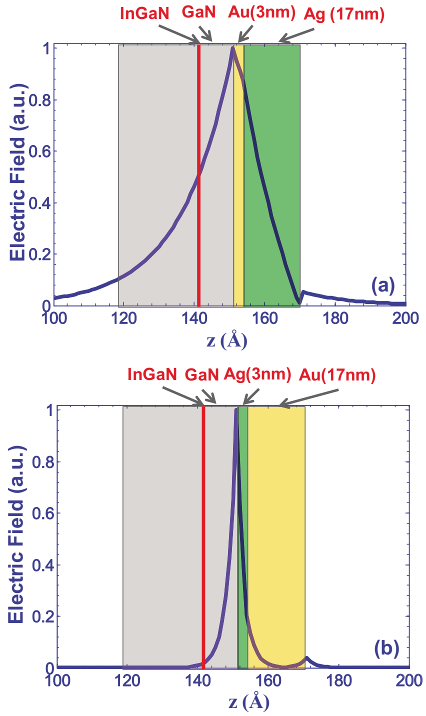

FIG. 3. (Color online) Electric field for double-metallic (a) Au (3 nm)/Ag (17 nm) and (b) Ag (3 nm)/Au (17 nm) layers on InGaN/GaN substrate.

Figure 4(a) shows the PL spectrum for the InGaN/GaN QWs coated with 6 nm Au/44 nm Ag, which is compared with the control InGaN QW sample without metal coating. From Fig. 4(a), significant PL intensity enhancement is observed by depositing the double metallic Au (6 nm)/Ag (44 nm) on top of InGaN/GaN QWs at peak emission wavelength of 490 nm. Note that the peak emission wavelength of the sample coated with Au/Ag layers shows redshift by 10 nm as compared to that of the control sample.

Figure 4(b) plots the enhancement ratios of PL intensities of the InGaN QWs with and without metal coating as a function of wavelength. The Ag-coated InGaN QW sample shows larger enhancement at shorter wavelength due to the higher SP frequency. By using the double-metallic Au/Ag layers on top of InGaN QWs, the peak enhancement ratio shifts to the longer wavelength region as shown in Fig. 4(b). From Fig. 4(b), as the Au layer thickness increases from 2 nm, 4 nm to 6 nm, the peak enhancement ratio increases in the green regime. The trend shows good agreement with theory. The InGaN QWs coated with Au (6 nm)/Ag (44 nm) shows as the optimized structure for green-emitting LED. The peak enhancement ratio for the InGaN QWs coated with the double-metallic layers decreases as the thickness of the gold layer increases further to 8, 10, and 12 nm. The enhancement ratio of the InGaN QWs coated with Au (12 nm)/Ag (38 nm) shows minimum enhancement in the green emission region, which is similar to that of the InGaN QWs coated with 50 nm gold layer. The Au-coated InGaN QWs shows the enhancement ratio between 1.1 and 1.6, which is due to the reflection from the Au layer.

All the proof-of-concept experiments on the tuning of the SP frequency by using the double-metallic layers were performed with total metallic layer thickness of 50 nm and

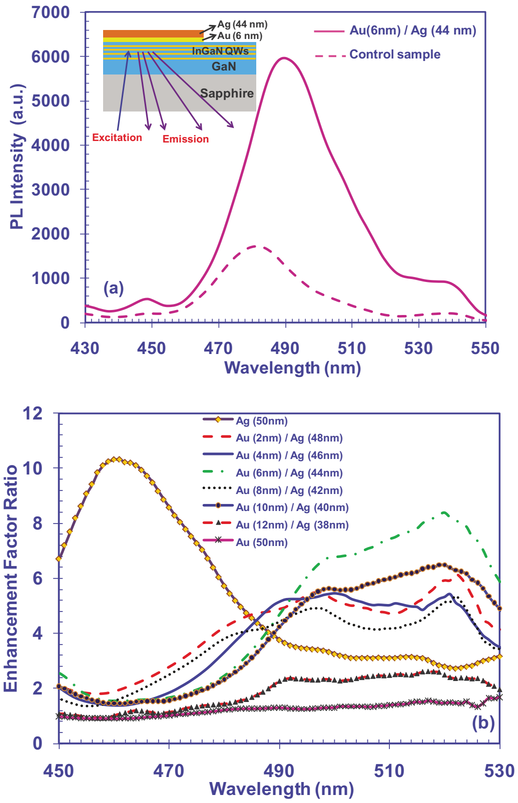

FIG. 4. (Color online) (a) PL spectra comparison of InGaN QWs coated with Au (6 nm)/Ag (44 nm) and control sample (with no metal coating), and (b) the enhancement ratios of PL intensities of the InGaN QWs with metal coating and without metal coating as a function of the wavelength for various double-metallic Au/Ag layers thicknesses.

GaN spacer layer thickness of 10 nm. Future works on the optimized total metallic layer thickness and GaN spacer thickness for nitride LEDs need to be carried out.

The results from Fig. 4(b) clearly indicate that the use of Ag-only SP structure resulted in enhancement factor with decreasing value as the wavelength is away from the ω_{sp}. However, the use of Au/Ag double metallic layer structure leads to increase in Purcell factor for emission wavelength between the two SP frequencies, with enhancement up to 8.3-times in the green spectral regime.

In summary, the use of double-metallic layers on top of semiconductor was proposed and demonstrated as an approach to tune the SP frequency and Purcell factor between the two SP frequencies of the single metal on top of the semiconductor. The concept of the SP frequency tuning using double-metallic layers can be extended to tune SP frequency from UV up to red spectral regime. This approach has the potential to realize SP-based LEDs with enhanced IQE in a visible spectral range.

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