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Surface plasmon-enhanced light-emitting diodes using silver nanoparticles embedded in p-GaN

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Abstract
We demonstrate the surface plasmon-enhanced blue light-emitting diodes (LEDs) using Ag nanoparticles embedded in p-GaN. A large increase in optical output power of 38% is achieved at an injection current of 20 mA due to an improved internal quantum efficiency of the LEDs. The enhancement of optical output power is dependent on the density of the Ag nanoparticles. This improvement can be attributed to an increase in the spontaneous emission rate through resonance coupling between the excitons in multiple quantum wells and localized surface plasmons in Ag nanoparticles embedded in p-GaN.

(Some figures in this article are in colour only in the electronic version)

1. Introduction
The group III-nitrides such as GaN, InN, AlN and their alloys have been demonstrated as materials for high efficiency light-emitting diodes (LEDs). Although III-nitride-based LEDs are already commercially available, further improvements in optical output power and external quantum efficiency (\(\eta_{\text{ext}}\)) are required [1]. The limited external quantum efficiency of LEDs is mainly attributed to the limited internal quantum efficiency (\(\eta_{\text{int}}\)). The \(\eta_{\text{int}}\) is strongly influenced by non-radiative recombination processes of threading dislocations [2] and also by a separation of the electron and hole wavefunctions by the polarization-induced electric field [3, 4].

Recently, among many approaches for the improvement of \(\eta_{\text{int}}\), surface plasmons (SPs) have been actively studied to enhance the luminescence efficiency of light-emitting materials and devices [5–12]. Surface plasmons are the collective oscillations of free electrons in a metal, which occur at the interfaces between metals and dielectrics. In particular, the collective oscillations of electrons in noble metal nanoparticles embedded in a dielectric matrix are localized surface plasmons (LSPs). The resonant excitation of LSPs leads to selective photon absorption and enhancement of local electromagnetic fields near the metal nanoparticles by orders of magnitude. However, this enhancement of local electromagnetic fields is strongly dependent on the distance between the quantum well (QW) and the metal nanoparticles. The penetration depth of the SP fringing field into the semiconductor is given by

\[
Z = \frac{\lambda}{2\pi \sqrt{|(\varepsilon'_{\text{GaN}} - \varepsilon'_{\text{metal}})/(\varepsilon'_{\text{GaN}})^2|^{1/2}},
\]

where \(\varepsilon'_{\text{GaN}}\) and \(\varepsilon'_{\text{metal}}\) are the real part of the dielectric constants of the semiconductor and the metal. The penetration depth is estimated to be 41 nm for QW–SP coupling at an emission energy of 2.75 eV (450 nm) for a blue LED when using the real part of the dielectric constant of the GaN and Ag metal [8, 9]. Therefore, when the QW is located close to the Ag nanoparticles within a distance of 41 nm, a strong resonant coupling between the QW and the...
LSPs is expected to take place. Kwon et al. reported on the SP-enhanced LEDs using Ag nanoparticles which were inserted in the n-GaN layer underneath the multiple quantum wells (MQWs) [8]. However, it is difficult to control the distance between the Ag nanoparticles and the MQWs. In addition, a large amount of Ag nanoparticles is evaporated at a high growth temperature for the overgrown n-GaN capping layer and the crystal quality of InGaN QWs is also deteriorated by Ag nanoparticles embedded in n-GaN. To overcome these problems, we demonstrate the SP-enhanced blue LEDs with Ag nanoparticles embedded in p-GaN. The optical properties of SP-enhanced LEDs with Ag nanoparticles were improved by the resonance coupling between the excitons in MQWs and LSPs in Ag nanoparticles. Furthermore, we observed that the optical and electrical properties of SP-enhanced LEDs were strongly dependent on the density of Ag nanoparticles.

2. Experimental procedures

Figure 1(a) shows a schematic of SP-enhanced blue LEDs with Ag nanoparticles embedded in a p-GaN layer. The LEDs were grown on a c-plane (0001) sapphire substrate by metal–organic chemical vapor deposition (MOCVD). After the growth of a 25 nm thick GaN nucleation layer at 550 °C, a 2 μm thick undoped GaN layer and a 2 μm thick n-GaN layer were grown at 1020 °C. Then five InGaN/GaN MQWs were grown at 770 °C, followed by a 30 nm thick p-GaN layer grown at 970 °C. In order to deposit Ag nanoparticles on the p-GaN layer, the samples were removed from the MOCVD chamber. To investigate the influence of the density of Ag nanoparticles, 0.3 (sample A) and 0.6 nm thick (sample B) Ag layers were deposited on the p-GaN layer by electron-beam evaporation. After the thermal annealing of the Ag layers, a 50 nm thick p-GaN layer was then grown on the Ag nanoparticles at 800 °C for 2.5 min as a capping layer. Finally, a 150 nm thick p-GaN layer was grown on the p-GaN capping layer at a temperature of 970 °C. Figure 1(b) shows the cross-sectional transmission electron microscopy (TEM) image of the SP-enhanced LEDs. As shown in figure 1(b), the Ag nanoparticles are embedded in the p-GaN layer. The diameters and heights of Ag nanoparticles are 50 ± 15 and 40 ± 10 nm, respectively. The TEM image also shows that the distance between MQWs and Ag nanoparticles is 32 ± 5 nm, which is close enough for QW–SP coupling. To fabricate LEDs, a p-GaN layer was etched by an inductively coupled plasma (ICP) etching process using Cl2/CH4/H2/Ar source gases until the n-GaN layer was exposed for n-type ohmic contact. Then the LEDs with a size of 300 × 300 μm2 were fabricated using indium tin oxide (ITO) with a thickness of 200 nm as a transparent current spreading layer and Ct/Au as an n-and p-pad electrode by e-beam evaporation, respectively.

3. Results and discussion

In this experiment, the size of the Ag nanoparticles was controlled by thermal annealing in the MOCVD chamber. This process was performed at 800 °C for 3 min prior to the growth of the p-GaN capping layer. The average size of the as-deposited Ag nanoparticles on the p-GaN layer was 20 ± 10 nm. However, both the diameter and height of the Ag nanoparticles were increased by thermal annealing via Ostwald ripening [13]. Figures 2(a) and (b) show atomic force microscopy (AFM) images of the p-GaN capping layer grown at a temperature of 800 °C for 1.5 min, where Ag nanoparticles are embedded in the p-GaN layer. As shown in figures 2(a) and (b), sample A with an 0.3 nm thick Ag layer has GaN-covered Ag nanoparticles with a diameter of 80 ± 25 nm and a height of 25 ± 10 nm. The average diameter of Ag nanoparticles measured by AFM is larger than that of the TEM result. This result is attributed to the p-GaN capping layer grown on Ag nanoparticles for 1.5 min. In the case of sample B with 0.6 nm thick Ag layer, the diameter and height of the GaN-covered Ag nanoparticles are similar to those of sample A. The densities of the Ag nanoparticles of samples A and B are 8 × 109 cm−2 and 4 × 109 cm−2, respectively. This result indicates that the density of Ag nanoparticles is dependent on the thickness of the deposited Ag layer. Furthermore, the densities of Ag nanoparticles are much higher than those of Ag nanoparticles embedded in n-GaN due to the lower growth temperature for p-GaN [8]. Figures 2(c) and (d) show the surface morphology of p-GaN after the overgrowth of a 150 nm thick p-GaN layer. As shown in figures 2(c) and (d), full coalescence of p-GaN is achieved and the Ag nanoparticles are fully covered by the p-GaN epilayer in both samples. The root mean square (RMS) roughness of sample A is 0.37 nm, which
Figure 2. (a), (b) AFM images of Ag nanoparticles (0.3 and 0.6 nm thick Ag film) deposited on a p-GaN after the growth of capping layer for 1.5 min. (c), (d) AFM images of p-GaN after the overgrowth of 150 nm thick p-GaN layer.

is similar to that of the as-grown p-GaN epilayer. In the case of sample B, however, the RMS roughness is 0.45 nm, which is larger than that of sample A. This result can be attributed to the increase of coalesced regions of GaN on Ag nanoparticles in sample B. Therefore, as shown in figures 2(c) and (d), the RMS roughness and surface pit density are increased with increasing the density of Ag nanoparticles.

Figure 3(a) shows the photoluminescence (PL) spectra and wavelength-dependent PL enhancement of InGaN/GaN MQW LEDs without and with Ag nanoparticles in the p-GaN layer. The PL spectra were measured from the top side of the samples at room temperature using an He–Cd laser (λ = 325 nm) with an excitation laser power of 50 mW. As shown in figure 3(a), the PL intensity of the InGaN/GaN MQW LEDs with Ag nanoparticles is much higher than that of the LED without Ag nanoparticles. The integrated PL intensity of LEDs with Ag nanoparticles is increased by 2.86 times compared with that of LEDs without Ag nanoparticles. As shown in the inset of figure 3(a), the PL enhancement was increased with increasing the density of Ag nanoparticles. This enhancement can be attributed to the coupling between the MQWs and the LSPs due to the charge density oscillations of confined SP modes in the Ag nanoparticles [5–8]. To confirm the resonant wavelength of Ag nanoparticles in the GaN layer, the transmittance spectrum was measured. Figure 3(b) shows the transmittance spectrum of Ag nanoparticles embedded in the p-GaN layer. As shown in figure 3(b), the transmittance of Ag nanoparticles embedded in the p-GaN layer is higher than 90%, indicating that the increase in PL intensity is not due to the reflection of light by the Ag nanoparticle layer. It is also noted that the transmittance spectrum of the Ag nanoparticles embedded in the p-GaN layer exhibits a absorption peak at 452 nm, which resulted from the extinction by the excitation of dipole plasmon modes in the Ag nanoparticles [8, 14]. In addition, the 0.3 and 0.6 nm thick Ag layer samples show a similar absorption wavelength because the size of the Ag nanoparticles is almost the same in both samples. Figure 3(b) shows that the absorption intensity is increased with increasing the density of Ag nanoparticles. The similar peak positions of PL enhancement as shown in the inset of figure 3(a) and absorption by Ag nanoparticles embedded in the p-GaN layer indicate that the PL enhancement is due to an increase in the spontaneous recombination rate of the exciton dipole in MQWs through QW–LSP coupling [5–8]. To further confirm that the improvement of $\eta_{int}$ is due to an increase in the spontaneous recombination rate through QW–LSP coupling, the temperature-dependent PL was measured in a temperature range from 10 to 300 K. Figure 3(c) shows an Arrhenius plot of the normalized PL intensity for the LEDs without and with Ag nanoparticles in p-GaN. The $\eta_{int}$ of LEDs can be estimated by comparing the integrated PL intensities, assuming that the $\eta_{int}$ is 100% at 10 K [5]. The $\eta_{int}$ of LEDs without and with an Ag layer of 0.3 and 0.6 nm thickness is 6, 13 and
Figure 3. (a) Room temperature PL spectra of InGaN/GaN MQW LEDs with Ag nanoparticles (0.3 and 0.6 nm thick Ag film) and without Ag nanoparticles. The inset shows the wavelength-dependent PL enhancement. (b) Transmittance spectra of Ag nanoparticles embedded in p-GaN layer. (c) Temperature-dependent integrated PL intensity of InGaN/GaN MQW LEDs with Ag nanoparticles (0.3 and 0.6 nm thick Ag film) and without Ag nanoparticles.

22%, respectively. The $\eta_{\text{int}}$ of LEDs with Ag nanoparticles in p-GaN is much larger than that of LEDs without Ag nanoparticles. This improvement of $\eta_{\text{int}}$ is believed to be due to the fast spontaneous recombination rate of the exciton dipole in MQWs. Since the density of states of the LSP mode is much larger, the QW–LSP coupling rate is very fast and this new path of recombination can increase the spontaneous emission rate [5–7]. In particular, the improvement of $\eta_{\text{int}}$ is dependent on the density of Ag nanoparticles as shown in figure 3(c). It is noted that the probability of coupling between the excitons in MQWs and LSPs is increased with increasing the density of Ag nanoparticles in p-GaN.

In order to investigate the electrical and optical properties of SP-enhanced LEDs, the current–voltage ($I–V$) characteristics and optical output power were measured. As shown in figure 4(a), the forward voltages of LEDs with Ag nanoparticles in p-GaN are 3.8 V, which are almost the same as that of LEDs without Ag nanoparticles. These results indicate that the Ag nanoparticles embedded in p-GaN do not degrade the electrical properties. However, the series resistance of LEDs with Ag nanoparticles is slightly increased with increasing the density of the Ag nanoparticles. The $I–V$ curves in figure 4(a) show that the series resistances of LEDs without and with an Ag layer of 0.3 and 0.6 nm thickness are 19.6, 22.1, and 28.7 $\Omega$, respectively. Moreover, as shown in the inset of figure 4(a), the reverse-bias leakage current of SP-enhanced LEDs with Ag nanoparticles is higher than that of LEDs without Ag nanoparticles. The increased series resistance and reverse-bias leakage current of LEDs with
a high density of Ag nanoparticles can be attributed to the defects induced by Ag nanoparticles in p-GaN, as shown by the surface pits in the AFM images of figures 2(c) and (d) [2, 15]. Figure 4(b) shows the optical output power of LEDs both without and with Ag layers in p-GaN as a function of injection current. The output powers of LEDs with Ag nanoparticles are much higher than that of LEDs without Ag nanoparticles. It is noticed that a 38% enhancement in optical output power of an SP-enhanced LED with a 0.6 nm thick Ag layer is achieved at an injection current of 20 mA compared to that of LEDs without Ag nanoparticles. The large enhancement of optical output power can be attributed to an improvement of $\eta_{\text{int}}$ in MQWs owing to an increase in the spontaneous emission rate by the resonance coupling between the excitons in MQWs and the LSPs in the Ag nanoparticles.

4. Conclusions

In summary, we demonstrate the SP-enhanced LEDs using Ag nanoparticles embedded in p-GaN. The optical output power of SP-enhanced LEDs was enhanced by 38% at an injection current of 20 mA compared to that of LEDs without Ag nanoparticles and the enhancement of optical output power was dependent on the density of the Ag nanoparticles. The enhancement could be attributed to an improvement of $\eta_{\text{int}}$ in MQWs due to the coupling between the excitons in MQWs and the LSPs in Ag nanoparticles in p-GaN.

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