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ABSTRACT

We investigated the surface plasmon coupling behavior in InGaN/GaN multiple quantum wells at 460 nm by employing Ag nanostructures on the top of a roughened p-type GaN. After the growth of a blue light emitting diode structure, the p-GaN layer was roughened by inductive coupled plasma etching and the Ag nanostructures were formed on it. This structure showed a drastic enhancement in photoluminescence and electroluminescence intensity and the degree of enhancement was found to depend on the morphology of Ag nanostructures. From the time-resolved photoluminescence measurement a faster decay rate for the Ag-coated structure was observed. The calculated Purcell enhancement factor indicated that the improved luminescence intensity was attributed to the energy transfer from electron-hole pair recombination in the quantum well to electron vibrations of surface plasmon at the Ag-coated surface of the roughened p-GaN.

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1. INTRODUCTION

GaN-based light-emitting diodes (LEDs) with InGaN/GaN multiple quantum well (MQW) structures, ranging from short-wavelength to long-wavelength part of the visible spectrum have been intensely developed [1-3]. However, crystal defects, dislocation and piezoelectric fields induce the non-radiative recombination energy in the InGaN/GaN MQW structures, which suppress the external quantum efficiency of GaN-based LEDs [1-4]. To achieve the high-efficiency LEDs, it is necessary to increase the optical extraction efficiency and internal quantum efficiency. Many interesting studies have been proposed to achieve this, such as the application of surface roughness, photonic crystal, carrier confinement and surface plasmon (SP) [4-10]. Especially, SP has attracted recent research interest of GaN-based LEDs requiring high quantum efficiency.

When a metal deposited on the surface of semiconductor (dielectric), the electro-magnetic field by oscillation of electron gas in metal is formed, the surface plasmon. If the SP energy matched well with the photon energy of LEDs, electron-hole pairs can transfer to the SP mode beside the radiative- and non-radiative recombination process in the active layer of LEDs [11-13]. In other words, SP provides the additional emission process by energy coupling between the SP and electron-hole pair, and suppress the non-radiative recombination process of injected carrier in the active layer. As a result, the internal quantum efficiency of LEDs can be increased. From these SP mechanisms, the joint researchers of CalTech and Nichia firstly reported the SP effect at the InGaN based blue LED structure [11]. The spacer thickness is a important factor for SP energy transfer, and it can be determined to calculate the penetration depth of SP. The penetration depth for the effective energy coupling into the semiconductor is given by $Z=\lambda/2\pi[(\varepsilon'_{\mathrm{GaN}}-\varepsilon'_{\mathrm{metal}})/\varepsilon'^{2}_{\mathrm{GaN}}]^{1/2}$, where $\varepsilon'_{\mathrm{GaN}}$ and $\varepsilon'_{\mathrm{metal}}$ are the real part of the dielectric constants of the semiconductor and metal, respectively [11]. Using the real part of the dielectric constant of the GaN and Ag metal, the penetration depth Z at an emission energy of 2.7 eV (459.2 nm) for a blue LED was estimated to be 42 nm for SP-MQW coupling, i.e., the Ag structure should be within 42 nm from the active layer of LEDs. However p-GaN can be substituted for the spacer layer in the conventional LEDs with thickness greater than 76 nm in a blue LEDs to obtain the efficient carrier injection [4]. But the thickness of spacer layer is found to be too thick for the energy transfer by SP-MQW coupling. Till date, there has been no report on the SP energy coupling deposited on the conventional LEDs having thick p-GaN.

In this paper, we approached a structural way to obtain the energy coupling by SP as well as the carrier injection of the conventional LEDs. We fabricated a 150 nm-thick p-GaN layer for the carrier injection and partially etched p-GaN layer for the efficient SP coupling. This structure showed a remarkable increase in PL and EL intensities. From the time resolved PL (TRPL) measurements, a faster decay rate and the calculated Purcell enhancement factor of partially etched LED with Ag nanostructures represented the energy transfer by SP-MQW coupling.

2. EXPERIMENTAL PROCEDURE

The InGaN/GaN MQW epitaxial structure was grown by metal organic chemical vapor deposition (MOCVD) technique. Trimethylgallium (TMGa), trimethylindium (TMIn) and NH_3 were used as precursors for Ga, In and N respectively. A thermal annealing of c-plane sapphire substrate was carried out at 1100 ^{o}C for 10 min, followed by the growth of a low temperature GaN buffer layer. A 1 μm-thick undoped GaN layer and a 2 μm-thick n-type GaN layer were grown at 1060 ^{o}C. Then, five pairs of InGaN/GaN MQW were grown on high quality GaN epitaxial layers. The GaN barriers and InGaN wells were grown at temperatures of 850 ^{o}C and 750 ^{o}C respectively. Two types of GaN layers were grown as the spacer layer between metal and MQWs, (i) 10 nm-thick undoped GaN layer was directly grown on MQW to investigate the SP-MQW energy coupling by the Ag nanostructures and (ii) 150 nm-thick p-GaN layer was grown on for electrical characterization. The schematic diagram of the GaN LEDs structures are shown in Fig. 1 (a) and Fig. 1 (b). In order to fabricate the partially etched p-GaN layer, a 100 nm-thick SiO_2 and 10 nm-thick Ni mask were deposited on the sample by plasma-enhanced CVD and e-beam evaporator [14]. The sample was subsequently annealed at 800 ^{o}C for 1 min under N_2 ambient to form the Ni clusters. Finally, the SiO_2 and p-GaN layer were etched for 120 sec using an ICP-reactive ion etching system. The etch depth of the p-GaN was ~ 130 nm. For the fabrication of Ag nanostructure, Ag thin film was deposited on the etched p-GaN layer of LED and subjected to rapid thermal annealing (RTA) under Ar ambient at 250 ^{o}C for 10 min.

The surface morphology of the deposited Ag was investigated by scanning electron microscopy (SEM, HITACHI S4300SE). PL and EL measurements were carried out to study the optical properties. The electrical luminescence characteristics were quantified through the back side photo detector by on-wafer probing of the devices. A 25mW, He-Cd laser (325nm) was used as the excitation source for the room temperature (RT) PL measurements. We carried out RT-TRPL measurements (using PicoQuant PicoHarp 300) to clarify the SP-MQW coupling. The optical apparatus consists of a commercially available InGaN/GaN based 375 nm laser diode used in pulsed mode operation, a monochromator, a photomultiplier tube, a high speed photodetector and controller electronics. All decay lifetimes are calculated by the software of Pico-Quant (FLUOFIT).

3. RESULTS and DISCUSSION

Figure 2 (a)-(d) shows the SEM images of the Ag nanostructures formed on the 10 nm undoped GaN spacer after annealing the Ag films with thickness of 10 nm, 15 nm, 20 nm and 25 nm at 250 ^{o}C for 10 min. A variety of morphologies ranging from irregular islands to regular clusters were obtained. The patch-shaped large Ag islands were noticed for the 25 nm-thick Ag film and the curved rod-shaped Ag clusters were observed for the 15–20 nm-thick Ag films. The 10 nm-thick Ag film resulted in the formation of regularly shaped, densely distributed Ag nanostructures with an average size of ~100 nm in diameter.

The front side PL responses from the MQW with the Ag nanostructures obtained from 10–25 nm-thick Ag films are shown in Fig. 3. Multiple interference peaks were clearly observed for the bare LEDs, indicating the presence of Fabry-

Perot (FP) effect caused by the mirror-like surface. However, the PL spectra of all the Ag-coated samples showed weak FP effects due to the light scattering caused by the Ag nanostructures at the Ag-GaN interface [15, 16]. Compared with the uncoated sample, the MQW coated with 10 and 25 nm-thick Ag films exhibited the decrease in PL intensities, whereas the 15 nm-thick Ag film showed a slight increase in PL intensity. In a sharp contrast, the sample coated with 20 nm-thick Ag films showed a remarkable increase in the PL intensity by about 80 %. In the SP mechanism, the phase matching between SP and photon energy is the important factor for the effective light emission [17]. The phase matching is controlled by the size and shape of the nanostructures as well as dielectric property of metal [18, 19]. With the increasing Ag thickness, the nanostructure size after the thermal annealing process was increased and the SP resonance wavelength gets red-shifted. The suitable thickness of Ag layer for efficient SP coupling at 460 nm is a 20 nm and it showed a good phase matching condition [20]. The results are found to be consistent with our enhanced PL intensity.

Based on these results, the 20 nm-thick Ag film layer was formed on the LEDs with a 150 nm-thick p-GaN layer by depositing and annealing as shown in Fig. 1 (b). Figure 4 shows the EL spectra of different LED structures, the bare LED without Ag (not etched) (sample A), the ICP-etched LED without Ag (sample B), and the Ag-coated LED after ICP (sample C). The peak wavelengths of the EL spectra were observed at 465, 460 and 462 nm for sample A, sample B and sample C at operating current of 20 mA. The inset of Fig. 4 shows the tilted SEM image of sample C. The forward voltages were 7.8, 7.7 and 8.3 V for the sample A, B and C respectively, at the operating current of 20 mA. The integrated EL intensity of the sample B compared with sample A was enhanced about 10%. The enhancement of the sample B is due to the increase of the light extraction by partially etched p-GaN [10, 21]. However, the integrated EL intensity of the sample C with a higher forward voltage was enhanced about 53 %. The significant EL enhancement is related to the SP energy coupling by the deposited Ag nanostructures on thin p-GaN after ICP etching as well as the increase of the light extraction by ICP etching.

The TRPL measurements were performed to investigate the SP coupling at the maximum emission wavelength of each sample and the results are shown in Fig. 5. After etching LEDs by ICP, the decay time was observed to decrease from 5.89 ns to 5.1 ns due to the increase of the non-radiative recombination rate in p-GaN layer. The relatively larger non-radiative recombination rate was caused by the ICP etching damages [21]. The average decay curve of the sample C was shorter than the sample A and B which was 3.3 ns. However it shows a multi-stage decay behavior. The fast and slow decay time of the sample C were found to be 290 ps and 4.16 ns, respectively. Normally, the fast decay time results from exciton recombination while the slow decay time attributed to localized carrier recombination by the quantum-confined Stark effect [21]. When the exciton energy transferred to the SP, the exciton energy of the MQW (fast decay) is close to the electron vibration energy of the SP [4, 11-13]. It means that the 290 ps decay time is obtained from the SP energy coupling of Ag nanostructures on the partially etched p-GaN layer. To describe the increase in spontaneous emission rate (R_{se}), the increase in Purcell enhancement factor (F_P) was calculated using the equation:

$$F_P(\omega)=\tau_{\mathrm{PL}}(\omega)/\tau^*_{\mathrm{PL}}(\omega)=k_{\mathrm{PL}}{}^*(\omega)/k_{\mathrm{PL}}(\omega)$$

where $k_{\mathrm{PL}}(\omega)$ and $k_{\mathrm{PL}}^{*}(\omega)$ are the original and changed PL decay rate, respectively [22]. From the equation, the F_P value was found to be 1.54 at emission energy of 2.68 eV (462 nm) for the EL improvement of the sample C. Therefore, the high F_P indicates that the improved EL intensity is attributed to the increase in spontaneous emission rate suggesting the effective coupling of SP-MQW.

4. Conclusion

We investigated the SP coupling behavior in InGaN/GaN MQWs by employing Ag nanostructures on the top of roughened p-type GaN. From the results of PL and EL measurements, the luminescence efficiency was enhanced by 80 % and 53 % for an input current of 20 mA. The calculated Purcell enhancement factor by faster decay time of partially etched LED with Ag nanostructures indicated that the enhanced luminescence intensity was attributed to the energy transfer from electron-hole pair recombination in MQW layer to electron vibrations of SP at the Ag-coated surface on the roughened p-GaN.

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Figures

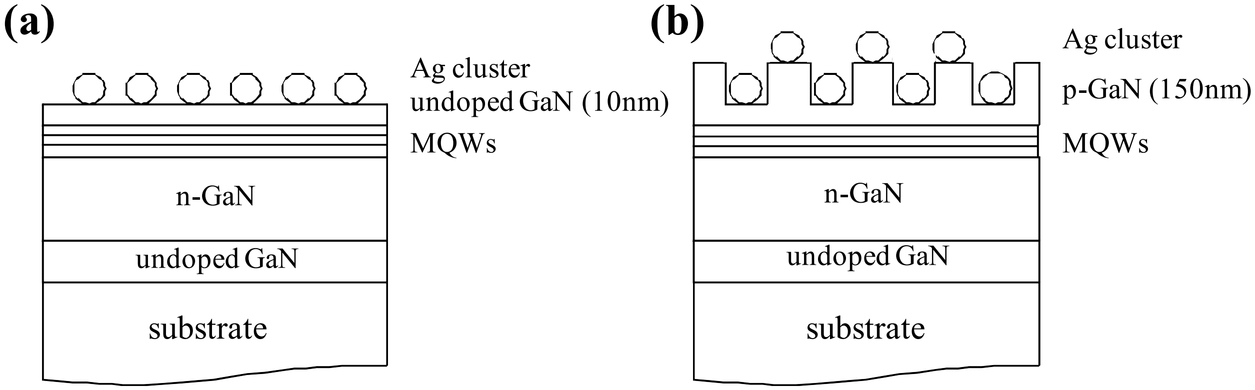

Fig. 1. Schematic sample structures for SP-QW coupling with 10 nm thick undoped GaN (a) and partially etched p-GaN (b).

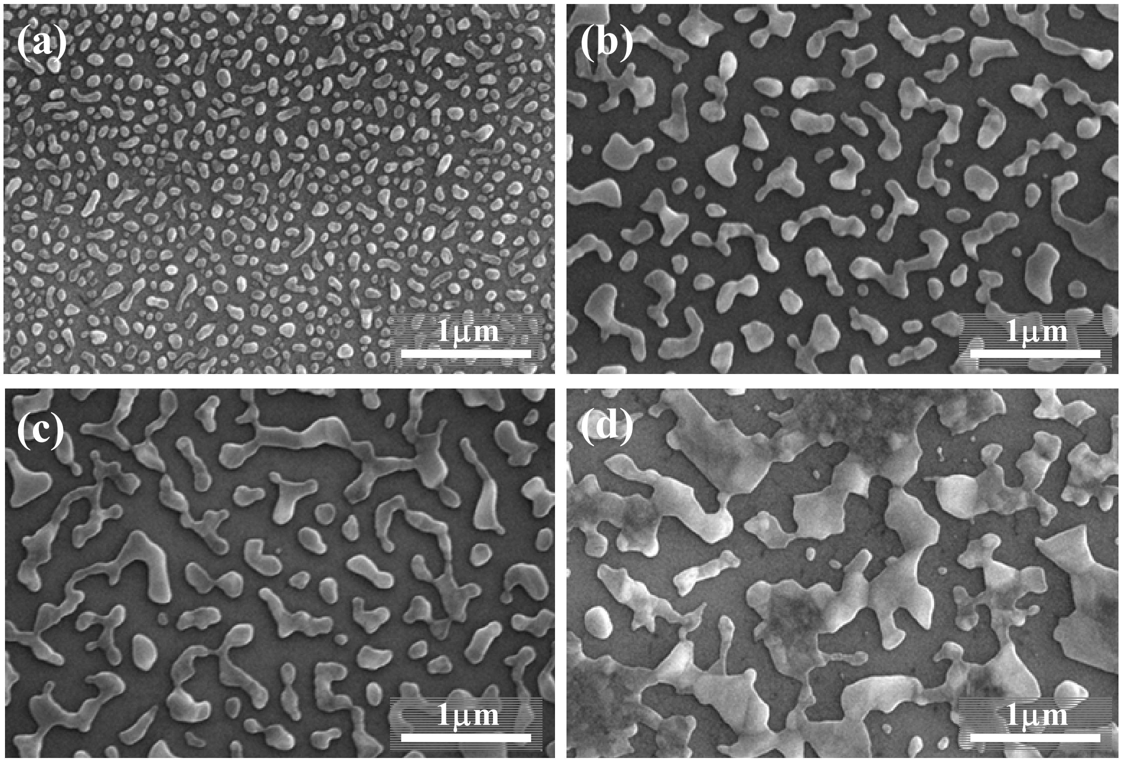

Fig. 2. SEM images of the Ag nanostructures formed on the 10 nm undoped GaN spacer after annealing the Ag films with thickness of 10 nm (a), 15 nm (b), 20 nm (c) and 25 nm (d).

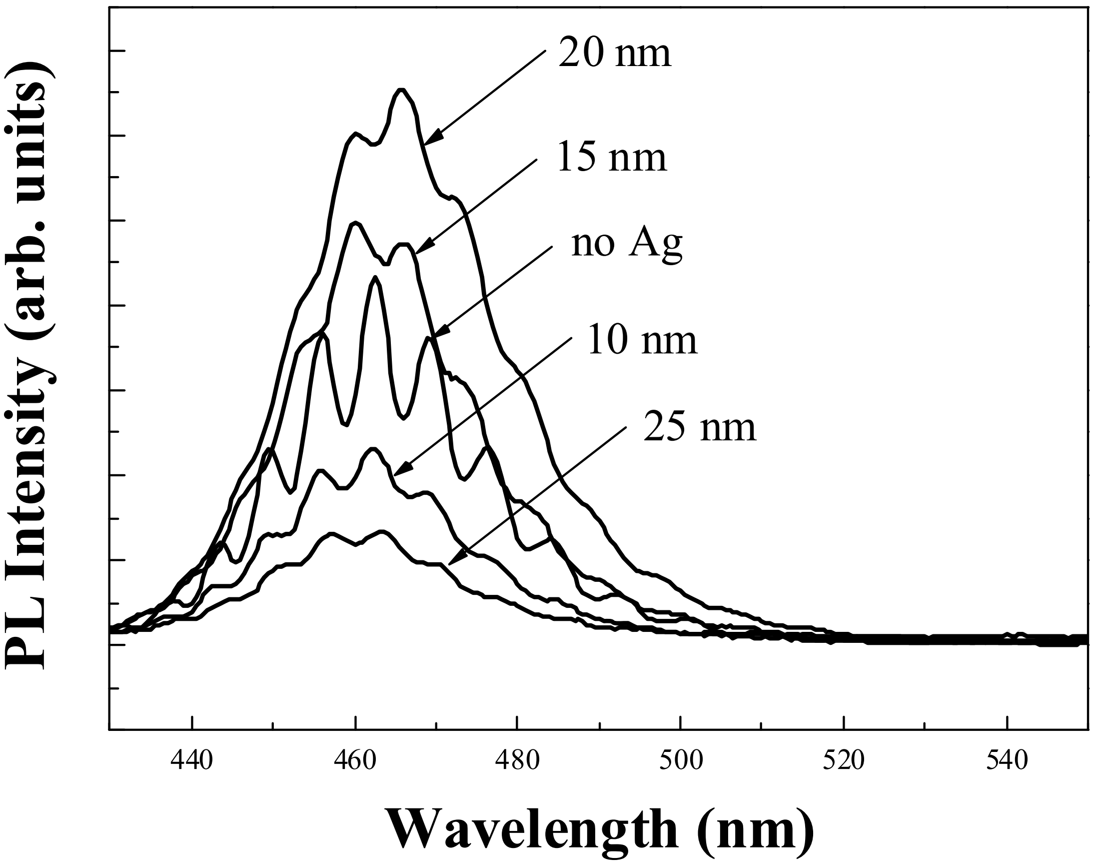

Fig. 3. Room temperature PL spectra from the samples with different Ag thickness.

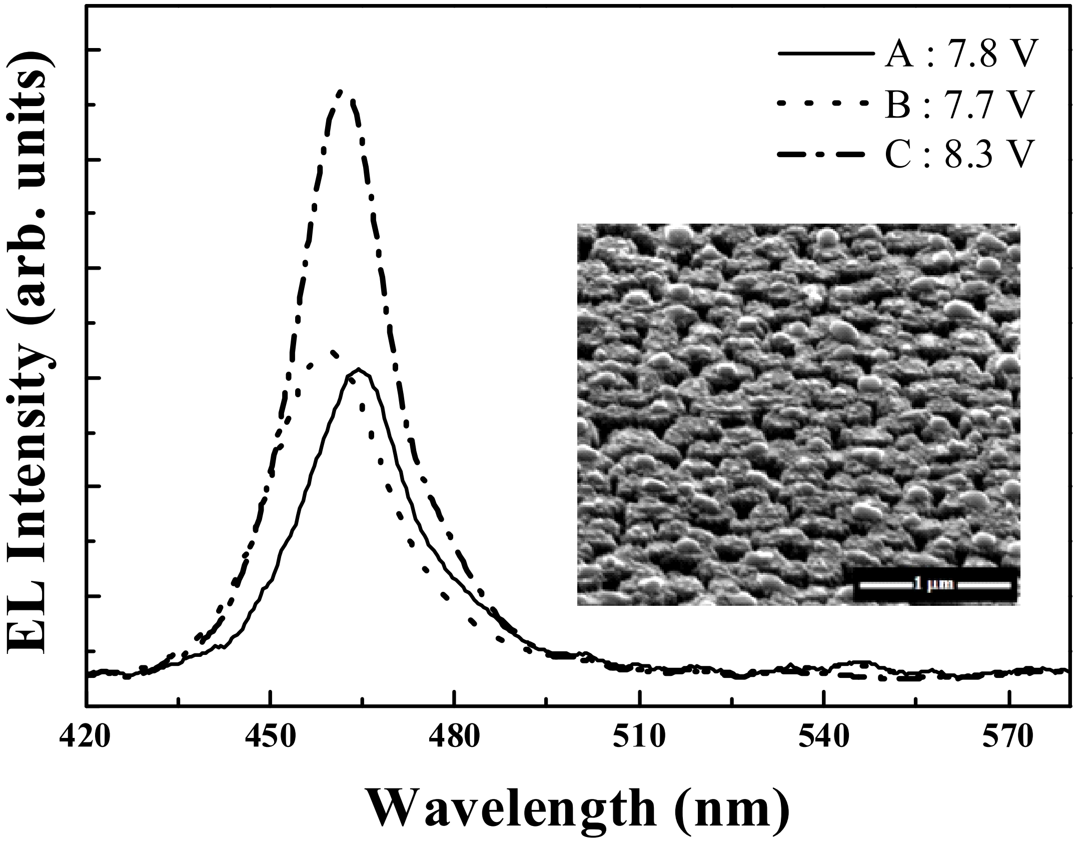

Fig. 4. EL spectra of the bare LED (A), ICP etched LED (B), and Ag coated LED after ICP etching (C). The inset shows the tilted surface morphology of the sample C by SEM.

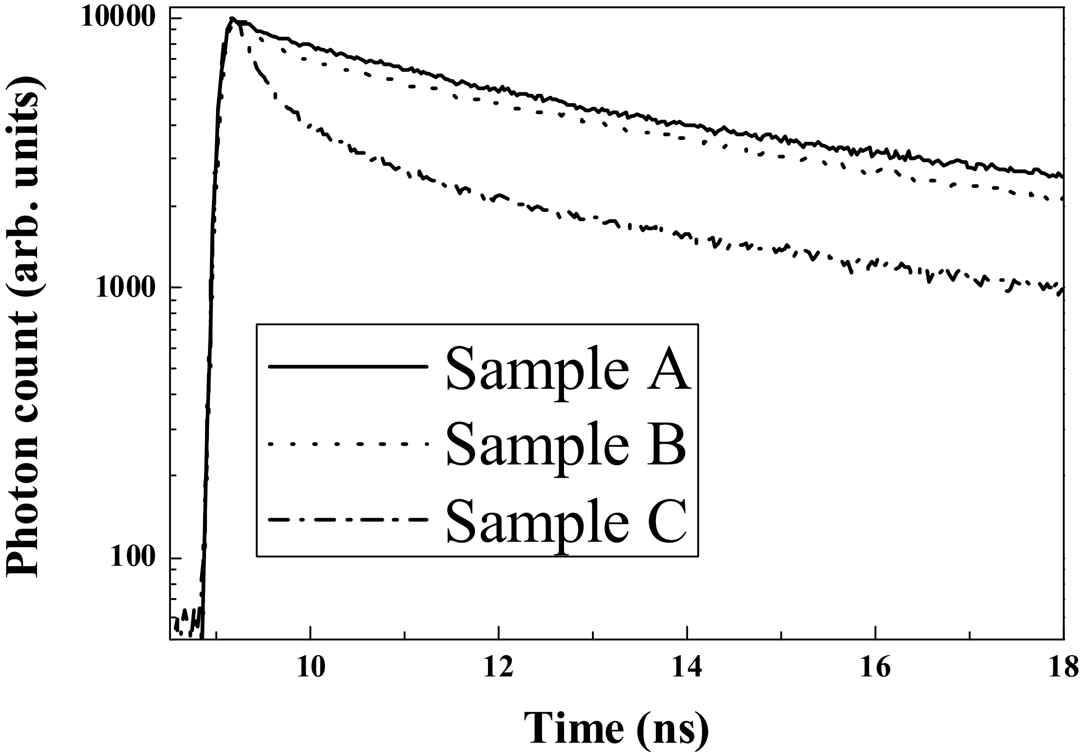

Fig. 5. Room temperature TRPL spectra of bare LEDs (A), ICP etched LEDs (B), and Ag coated LEDs after ICP etching (C).