Plasmonic Enhanced Emission of InGaN Quantum Dots by Ag Nanoparticles

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***Abstract*—Quantum dots incorporated multiple quantum wells have been grown by metalorganic chemical vapor deposition using trimethylIndium treatment. The size distribution of quantum dots from 30 nm to 120nm leads to a broadened peak emission signal from 480 nm to 700 nm. Annealing of Ag film deposited on the multiple quantum well generated Ag nanoparticles which serves to enhance the emission especially for longer wavelength spectrum as well as reducing the fluorescence lifetime for carriers during transition.**

Keywords: InGaN quantum dots, mutliple Quantum Wells, plasmonic

I. INTRODUCTION

For the past decade, phosphor coated LEDs have remained dominant in their application of white LEDs [1]. However, extensive research has shown that phosphor coated LEDs have poor colour rendering index (CRI), yield production issues and limited luminescence lifetime expectancy as it degrades overtime leading to colour temperature changes [2]. Currently, alternatives have been pursued to overcome these drawbacks. Quantum dots in semiconductors are nanostructures with properties which differ from bulk materials due to the carrier confinement effect. Further research has shown that InGaN self-assembled QDs which uses Indium atoms as anti-surfactants enhances the incorporation of Indium to form connected QDs which emit in the yellow wavelength [3]. This can be seen as a replacement for yellow phosphor technology [4, 5]. By dual stacking these connected QDs with amber emission with MQWs emitting in the blue wavelength, cool white light emission can be attained. However, there is little tuning that can be done to the color rendering index of the white light, hence in this work, we incorporated the use of Ag nanoparticles to generate enhance emission in the amber emission spectrum.

II. SAMPLE PREPARATION

The MQWs were grown using Metal Organic Chemical Vapor Deposition (MOCVD). The first two layers of quantum wells was grown at 780ºC in the MOCVD reactor with Trimethylindium (TMIn) flow of 200 sccm and Trimethylgallium (TMGa) flow of 90 sccm treatment to obtain a emission at approximately 460nm. For the quantum dots incorporated layer, TMIn treatment was carried out by inhibiting the flow of Trimethylgallium for 5s to promote the growth of the quantum dots for the top layer of quantum well. The layer was subsequently capped by InGaN well layer grown at the same condition as the initial two quantum wells. The schematic diagram of the growth layer structure is as shown in Figure 1.

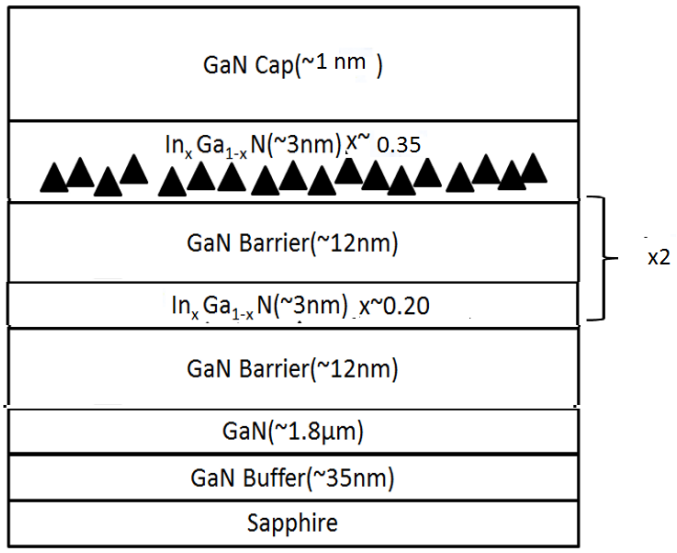

Figure 1 shows the schematic diagram of the growth MQWs structure with incorporated quantum dots

AFM scan was carried out for the sample to determine the morphology and the distribution of the quantum dots obtained for the sample. Figure 2a shows the flatten image of the surface morphology of the MQWs sample grown with a cap layer of GaN with thickness of 1 nm. The size of the smaller quantum dots range from 30 nm to 50 nm while the bigger size dots are about 70 nm to 120 nm. The height of the dots varies from 0.5 nm to 4nm as determined by its size

proportionality. This sample was then deposited with silver (Ag) film up to 50nm by e-beam deposition

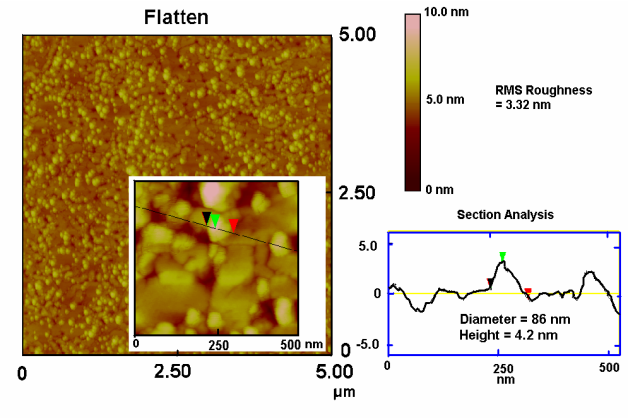

Figure 2 shows the AFM image of the quantum dots with a distribution of size from 30 nm-50nm for small quantum dots and 70 nm to 120 nm for bigger quantum dots distributed in the QWs structure. The inset shows a magnified image to determine the size of the dots.

III RESULTS AND DISCUSSION

A. *PL Measurement for quantum dots incorporated Mulitple Quantum Well*

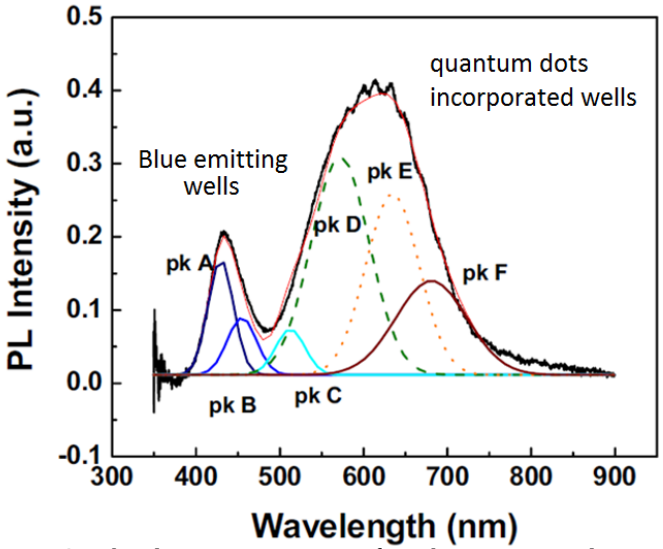

Figure 3 Photoluminescence scan for the quantum dots incorporated quantum wells with the fitted peaks as attributed by the different well layers

Photoluminescence (PL) measurement was carried out for the sample from 350 nm to 900 nm. Figure 3 shows the PL spectrum with the fitting done for the respective peaks identified. The peak A is attributed to the blue emission from the two blue quantum wells layer grown. The broaden peak Peak D, E and F are likely to be attributed to the varying emission spectrum from different size categories of the quantum dots identified. The fitted red emitting spectrum is likely to be attributed to the bigger size quantum dots as a result of agglomeration of Indium while the peak E and Peak D are attributed to the smaller size dots giving amber and yellow emission respectively. There are some transition fitted peaks (peak B and C) due to down conversion of the blue emission from the two base layers of quantum wells.

B. *Surface Plasmon Resonance*

Plasmonic metal nanoparticles have great potential for chemical and biological sensor applications, due to their sensitive spectral response to the local environment of the nanoparticle surface and ease of monitoring the light signal due to their strong scattering or absorption. [6,7]

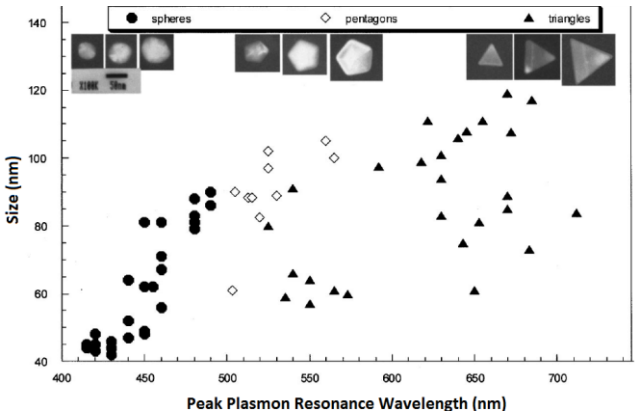

Figure 4 Plot of 3D images versus spectral peak wavelength for a diverse collection of individual silver particles of different size and shapes. The plot is obtained from Ref [6].

Work has done on the effects of shape variation of silver nanoparticles on the different regime of surface plasmon enhancement.

Based on Figure 4, to achieve plasmon resonance for the red emission peak, the size of the Ag nanoparticles must ideally be above 50 nm with triangular structure while to enhance the yellow emission, the size requirement remains unchanged but the Ag nanoparticles should be pentagon in structure.

In our work, e-beam deposited Ag film was annealed with varying annealing recipe and step condition to achieve the triangular profile for the Ag nanoparticles. The annealed temperature caused the Ag particles to ball up with thermal diffusion of the Ag metal. It was observed that the balling of the silver nanoparticles will occur at the position in close proximity to the quantum dots due to change in the surface energy at the dots and flat surfaces. The effect of varying

annealing condition to generate different structure will be reported in other journal article.

C. *Time Resolved Photoluminescence for quantum dots incorporated Mulitple Quantum Well*

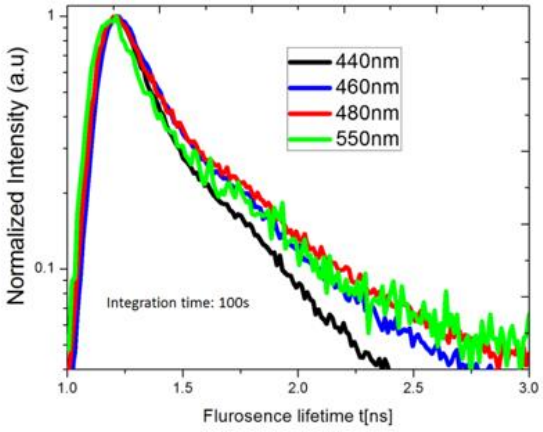

Figure 5 Fluorescence lifetime measurement by TRPL system for quantum dots incorporated quantum wells

Time Resolved Photoluminecence (TRPL) was conducted on the sample before Ag deposition to observe the decay rate of the quantum dots sample. The results are as shown in Fig 5 fitting with the exponential function as shown in Eqn1

$$I(t) = I(0)\, exp\left[-\left(\frac{t}{\tau}\right)\right]^{\beta} \qquad (1)$$

Where β is the stretching parameter and $I(t)$ is the emission intensity as a function of time. The stretched exponential function or the Kohlrausch function has been constantly applied to study the effects of photoluminescence decay on disordered semiconductors such as porous silicon [8,9]. Our samples (InGaN MQWs with QDs) displayed similar characteristic to porous silicon as they are both disordered semiconductors. The half-life approximation was used in our analysis of the PL decay rate and the result is plotted in Table I.

Table I reports the half-life of the fluorescence lifetime after being fitted

Wavelength	**Half-life Time [ps] (Approx.)**
440nm	118.80
460nm	134.80
480nm	135.15
550nm	135.11

Based on the results attained in Fig.18, we report that the fluorescence lifetime is reduced at shorter wavelengths. The change after 460nm is not very significant and stagnated after 480 nm. Hence, it can be deduced that the radiative recombination rate of the free and donor-bound excitons is faster at shorter wavelengths. (Approx. < 480nm). The faster response can be attributed to small amount of quantum dots with blue emitting signals (peak B) as shown in Fig 3.

D. *TRPL Measurement for quantum dots incorporated Mulitple Quantum Well after formaiton of Ag nanoparticles*

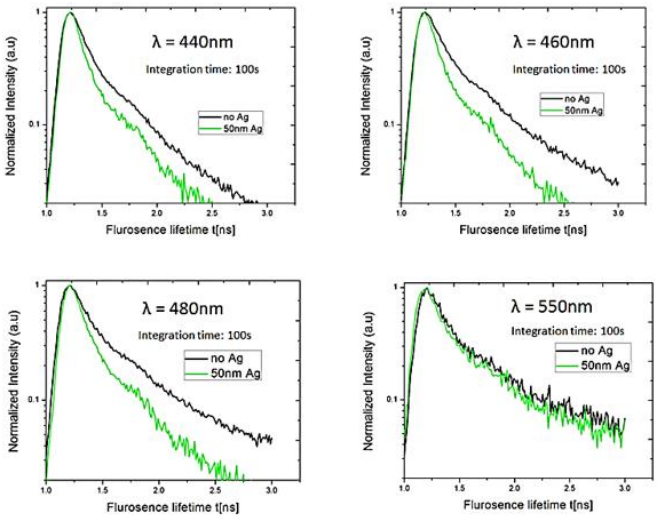

Figure 6 *shows the TRPL (Normalized Intensity vs Fluorescence lifetime [ns]) spectra of the (50nm Ag) sample (in green performed) at different wavelengths at room temperature compared to the original sample (no Ag) plotted in* black

Figure 6 illustrates the TRPL of the sample after 50nm deposition as compared with the measurement done for sample without the Ag deposition and annealing. Fitting is carried out for the TRPL curve using eqn (1) and half-life is reported in TABLE II

Table II reports the half-life of the fluorescence lifetime for the Ag nanoparticles after being fitted using eqn (1)

	440nm	**460nm**	**480nm**	**550nm**
Thickness	Half-life Time [ps] (Approx.)	Half-life Time [ps] (Approx.)	Half-life Time [ps] (Approx.)	Half-life Time [ps] (Approx.)
No Ag	118.8	135.8	135.2	135.1
50 nm Ag	91.0	91.1	102.1	103.2

The half-life which corresponds to their respective wavelength is detailed as shown in Table II. The radiative recombination rates of the samples are significantly improved as seen from the decrease in half-life throughout the different wavelengths. The highest improvements can be observed at higher wavelengths of 480 nm and 550 nm,

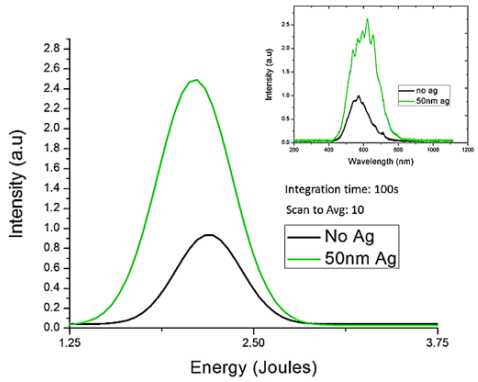

Figure 7 PL spectrum of the sample after depositing a layer of 50nm Ag. Inset gives the PL plot in nanometer

As shown in Fig 7, the 50nm Ag nanoparticles enhances the integrated PL intensity by more than two times and leads to a red-shift in the emission spectrum. A peak broadening was also observed as evident by the FWMH (0.52eV to 0.59eV) of the peak signal. The peak emission was also observed to have shifted from (2.193eV to 2.099eV).

III CONCLUSION

In summary, InGaN quantum dots have been grown and incorporated into blue quantum well structure. The size variation of the quantum dots results in the broaden peak emission which covers from green to red emission. Ag nanoparticles was generated by annealing the deposited Ag film and the result shows an enhancement in the PL emission spectra with a broadening of peak signal. This can be attributed to plasmonic coupling effect of the different size of quantum dots embedded in the uppermost well. A much faster radiative recombination rate has also been reported for all the wavelengths under investigation and this is attributed to the surface plasmon coupling effect. Localized surface plasmon offers a greater tunability that is the properties resulting from LSP can be easily be varied through alteration of the size and geometry of the Ag nanoparticles with optimized annealing process. This will enable the tuning of QDs LEDs to achieve better color rendering index. [10]

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