**Observation of whispering gallery modes in InGaN/GaN multi-quantum well microdisks with Ag plasmonic nanoparticles on Si pedestals**

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**Abstract**

We fabricate the plasmonic free-standing InGaN/GaN multi-quantum wells (MQWs) microdisks on the Si (111) pedestals using wet-chemical undercut etching, followed by decorating of Ag nanoparticles on microdisks to improve whispering gallery modes (WGMs) resonance emission. The enhancement results from the plasmonic coupling effect between excitons in MQWs and localized surface plasmons of Ag. The radial resonance WGMs from optically pumped microdisk cavities are observed in the photoluminescence spectra at threshold optical pumping power density of 4.7 kW/cm2 with WGMs mode spacing *Δλ* = 1.3 nm.

**Keywords:** Microdisks**,** Whispering gallery modes, InGaN/GaN multi-quantum wells

1. **Introduction**

Over the past decade, III-nitride semiconductors have been achieved remarkable success in high power light-emitting diodes (LEDs) and laser diodes (LDs). Many researchers have made extraordinary strides in improving the efficiency of LEDs for solid-state lighting applications, i.e., defect reduction techniques as effective strain, polarization, bandgap engineering of LED heteroepitaxy structures, and various light extraction techniques as texturing, photonic crystals, and localized metal plasmons [1].In sympathy with defect reduction of epitaxial nitride films and fabrication techniques of cleaved cavity mirror, the great successes of commercially available GaN-based Fabry-Perot (F-P) edge-emitting LDs have been reported and realized in blue, green, and ultraviolet wavelength [2,3].However, III-nitride semiconductors have not yet come close to achieving its full potential and a new possibility in the field of novel conception LDs, e.g., distributed feedback (DFB) edge-emitting F-P LDs and vertical-cavity surface-emitting laser (VCSEL) with distributed Bragg reflectors (DBR) [4,5].

GaN-related microdisk cavities, one of the challenges in optical resonance cavity with nitride semiconductors, are of interest for studies of cavity quantum electrodynamics as well as for their unconventional lasing characteristics which offer several benefits over the edge emitting devices, including enhanced quantum efficiency, high quality factor, and reduced lasing threshold, due to the whispering gallery modes (WGMs). Microdisks or micro-rings have early been fabricated in GaN-based semiconductors, leaving a thicker sapphire substrate, and have shown a small volume cavity mode lasing [6,7].The milestone progress has began from the successful fabrication of freestanding GaN/InGaN multi quantum wells (MQWs) microdisk has a surrounding air index, using by a lateral bandgap-selective photo-electro-chemical (PEC) etch process at InGaN/InGaN sacrificial superlattice or oxidized AlInN sacrificial layer [8,9]. Also, nonpolar InGaN/GaN MQWs microdisks were reported that have a threshold optical pumping power as low as 400 W/cm2 and a quality factor as high as 2000, because of the lack of a Stokes shift in *m*-plane oriented MQWs [10].In case of GaN epitaxial growth on Si substrate [11],the microdisks which have resonance cavities medium of thin GaN film [12],GaN/InGaN MQW templates [13],and embedded GaN/AlGaN quantum dots in AlN stacks [14], have been produced on Si (111) substrates due to extreme higher chemically reaction etching selectivity between nitrides and Si substrates. The WGMs stimulated emissions with high quality factors have been observed in aforementioned structures. In this regard, herein, free-standing InGaN/GaN MQW microdisks on Si pedestals have been fabricated. With incorporating Ag nanoparticles, emitted light from microdisks was enhanced by surface plasmons effect. Particularly, the WGMs behavior in free-standing InGaN/GaN MQW microdisks with Ag nanoparticles has been also observed.

**2. Experimental Method**

Firstly, InGaN/GaN MQWs epitaxial templates were grown on highly tensile-strained GaN films on Si (111) substrate using MOCVD as shown in Fig. 1. In general, the growth of GaN on Si substrate often leads to cracks and crystalline defects due to a high lattice and thermal mismatch between GaN and Si. In order to overcome these problems, InGaN/GaN MQWs epitaxial templates were constituted mainly three parts, namely: (i) Al pre-seeding process for nucleation, (ii) AlN/GaN/AlN strain compensation structure, and (iii) SixNy dislocation blocking layer. Our previous work reported that blue emission and crack-free InGaN/GaN MQWs were successfully grown on Si (111) substrates by optimized growth parameters [15,16]. For the fabrication of 20 μm diameter microdisk cavity, a 0.5 μm thick SiO2 layer, which acts as selective mask material for dry-etching and for avoiding surface damages, was deposited on InGaN/GaN MQWs templates. The SiO2 layer was wet-etched by conventional photolithography process, and the whole thickness of nitride layers etched using by inductive coupled plasma reactive ion etching (ICP-RIE). The microdisks mesas were formed as a trapezoid side facet due to anisotropic dry etching. For fabrication of completed free-standing microdisks, the samples were undercut etched with HF acid solution. A HF acid is used to etch the Si substrate area without attacking GaN epilayers. Finally, 50 nm thick of SiO2 layer was deposited on microdisk surface for passivation of surface damages using PECVD system, followed by spray-coating of Ag NPs on passivated microdisk surface for investigating of a new possibility of a microdisk cavity fused with surface plasmonic nature. The surface morphology and structure of prepared microdisks were investigated using scanning electron microscopy (SEM). The excitation power dependent photoluminescence (PDPL) and time-resolved photoluminescence (TRPL) measurement system was used to observe the optical properties of fabricated microdisks. The PDPL spectra were investigated from one microdisk using He-Cd laser (λ = 325 nm) with an excitation laser total power ranging from 11 mW to 45 mW for spot diameter of about 30 μm, which are corresponded power densities from 1.6 kW/cm2 to 6.3 kW/cm2, respectively, at room temperature. Also, the TRPL were measured using a streak camera detector at room temperature. TRPL used a pulsed 266 nm laser generated by the second-harmonic generation of a mode-locked femto-second Ti: sapphire laser, with an average output power of more than 100 mW and a pump power of 4 W. The pulse width and repetition rate were 100 fs and 76 MHz, respectively.

**3. Results and discussion**

Figure 2 (a-d) shows a schematic illustration of the fabrication process for free-standing InGaN/GaN MQWs-based microdisks together with SEM images for each step, respectively. It is clear to see that a microdisk structures are formed on top of tiny Si pedestals as illustrated in Fig. 2(c). Here, we proposed a novel microdisk for improving WGMs resonance emission originated by surface plasmon effect of incorporated Ag nanoparticles. The most notable phenomenon occurring with the metal nanoparticles on insulators is electromagnetic resonances due to collective charge oscillations of the metal’s free electrons, termed localized surface plasmons (LSPs). These plasmonic features have attracted interest for the enhancement of quantum efficiency of excitonic emitters. The radiative scattering path through the coupling with LSPs occurs much faster than other non-radiative phonon scattering due to the closer extinction energy and strong localized fields of LSPs. It has been reported that LSPs coupled with InGaN MQWs of nitride based LEDs enhance both the photoluminescence intensity and light out power of LEDs through the efficient and rapid resonance with excitonic oscillation energy in MQWs [17].In particular, the Plasmon oscillation energy of Ag nanoparticles is suitable for surface Plasmon coupling to blue emission associated with MQWs [18]. Inherited from these reports, we fabricated Ag nanoparticles incorporated microdisk as shown in Fig. 2(d) for investigating of a new possibility of a microdisk cavity fused with surface plasmonic nature. Before incorporating Ag nanoparticles, 50 nm thick SiO2 surface passivation layer was deposited over both top surface and sidewalls of microdisks because the side walls of the microdisk were sloped through etch process. The SiO2 deposited sidewalls revealed smoother surface due to fill the striation roughness and passivated the surface imperfections. Finally, the Ag nanoparticles were randomly sprinkled on the whole of microdisk surface.

The SEM image of spray-coated Ag nanoparticles on the quartz substrate is shown in Fig. 3(a). The average particle size is found to fluctuate from 60 to 100 nm. The UV-Vis absorption spectrum of the synthesized Ag nanoparticles is studied to determine particle information and its properties. It can be seen from Fig. 3(b) that the UV-Vis absorption spectrum exhibits a broad maximum absorbance peak at 450 nm. Wiley et al. reported that metallic silver with a mean diameter of 80 nm give a surface plasmon resonance band in the 320−800 nm region [19]. Therefore, the observed peak at 450 nm is ascribed as the typical plasmon absorption peak of Ag nanoparticles. This peak is likely shifted to the red side with a broader shape as compared to other reports [20–22]. It is possibly due to the increase in size and inhomogeneous distribution of synthesized Ag nanoparticles. There are reports in the literature that, as the interparticle distances become smaller than the dimensions of the spherical particle, or even when aggregation occurs, the plasmon resonance redshift, and often an additional shoulder peak at 350 nm appears [21]. We also observe an addition shoulder peak at 350 nm. This observation implies that synthesized Ag nanoparticles have inhomogeneous sizes varied from 60 to 100 nm, which are consistent with the SEM image (Fig. 3(a)).

Figure 4(b) show the PDPL spectra of the InGaN/GaN MQW microdisk without Ag nanoparticles. The spectra only show multiple F-P interference peaks at shown 440 nm and 480 nm occurring in vertical reflection at interface between air and GaN-related template. The F-P interference peaks position is independent with the optical power. It should be noted that the distance between two peaks strongly depends on the thickness of the disk as well as effective refractive indices of InGaN and GaN materials. However, the WGMs stimulated emissions is absence in PDPL spectra even at a high optical pumping rate. It was attributed that higher defect density in MQWs and relatively lager active volume and microdisk diameter, and the striation shaped sidewalls caused by the erosion of photoresist and ion damages during the dry etching. Although the microdisk is isolated from substrate with thin Si pedestal, there still remains indispensable thicker GaN buffer template, which induces optical loss. As results, microdisks have not shown the WGMs emission as not only increased non-radiative scattering internal losses in MQWs but also cavity mirror losses at striation formed sidewalls.

Figure 5(a) exhibits the SEM image of Ag nanoparticles incorporated 20 μm diameter microdisk. The PDPL spectra of Ag plasmonic nanoparticles incorporated microdisk were investigated at varying excitation power densities as shown in Fig. 5(b). For microdisks without Ag nanoparticles, the maximum values for the F-P band are around 480 nm, as shown in Fig. 4b. When microdisks are coated with Ag nanoparticles, the peak shifts to 470 nm, as seen in Fig. 5b. The change in total thickness and effective reflective index of microdisks associated to Ag nanoparticles was blamed for the peak movement. The F-P peak about 440 nm was thought to be heavily reliant on excitation power, however, peak position is independent of optical power.

The intensity of LSPs coupling related peak increases much more rapidly with the optical pumping than that of F-P interference peak. Also, when the excitation exceeds the threshold value of a 4.7 kW/cm2, a sharp spectral feature emerged at a center wavelength of ~442.3 nm and ~443.6 nm as shown in Fig. 5(c). The observed peaks suggest the presence of WGMs at the round edge of microdisks due to low optical loss for a total internal reflection. The effective cavity length of *πR* imposed by the periodic boundary condition results in WGMs eigen mode condition of *πRneff = mλ*, for large integer *m*, and the mode spacing is given by *ΔλWGM = λ2/πRneff*, where *λ* is the wavelength of light propagating light, *neff* is effective index of refraction, and *R* is the diameter of the microdisk. Calculation of the expected mode spacing value and representative values of *R* = 20 μm and *neff* = 2.6 reveals that the observed mode spacing, *ΔλWGM* = 1.3 nm, correspond well to the WGMs resonance emission of microdisks. However, the threshold optical pumping power density for WGMs stimulated emission is too higher as over 4.7 kW/cm2, while the recent works recorded the threshold power density for WGMs lasing of GaN-based microdisks as low as a few hundred W/cm2 range [8,10].We also believe that the low threshold condition WGMs emitters can be improved by size down of microdisk diameter, improved smoothness of sidewalls, and reduced diameter of Si pedestals through improving fabrication techniques.

In order to elucidate the nature of the LSPs coupled emission enhancement of the Ag nanoparticles incorporated microdisks, TRPL were measured using a streak camera detector at room temperature as shown in Figs. 6(a) and 6(b). The photoluminescence spectrum of Ag nanoparticles incorporated microdisk showed two dominant peaks at ~440 nm and ~480 nm due to F-P interference effect as same as without Ag nanoparticles, whereas the photoluminescence intensity of around ~440 nm wavelength peaks are more enhanced comparing with the other wavelength region. Further insight is provided by TRPL peaks intensity decay measurements as presented in Fig. 6(c). Table 1 also shows the predicted carrier lifetime of the Ag nanoparticles integrated microdisk and comparison of bare GaN/InGaN MQWs microdisk. The TRPL spectra decay curves are not of a single exponential form, but can be approximated by a superposition of two exponential decays with differing relaxation times *τ*1 and *τ*2. A double-exponential decay function [*I*(t) = *A*1 exp (−*t*/*τ*1) + *A*2 exp (−*t*/ *τ*2)] was used to fit the decay curves for GaN samples, and then the decay times, *τ*1 and *τ*2, were calculated using the optimized fitting. In general, the fast decay component *τ*1 is thought to represent effective non-radiative recombination at room temperature, whereas the slow decay component *τ*2 is thought to represent the free exciton radiative lifetime. At 440 nm wavelength, the measured decay times for the microdisk combined Ag nanoparticles were *τ*1 = 0.36 ns and *τ*2 = 3.36 ns, respectively, while at 480 nm wavelength, they were *τ*1 = 0.48 ns and *τ*2 = 5.81 ns, respectively. These decay periods are shorter than those from a microdisk without Ag nanoparticles, with *τ*1 = 0.97 ns and *τ*2 = 19.67 ns at 440 nm wavelength and *τ*1 = 1.08 ns and *τ*2 = 8.57 ns at 480 nm, respectively. The initial fast decay component of TRPL decay curves is caused by exciton recombination, while the slow component is caused by localized carrier recombination caused by the quantum-confined Stark effect. The relatively short decay time could be taken as evidence for a significant excitation transfer process to nonradiative recombination channels in the GaN related epitaxial layer, such as the dislocations, point defects and their complexes. It is also linked to a decrease in PL intensity. However, even if the decay time of Ag nanoparticle incorporated microdisk, which is related to nonradiative recombination, is much faster than that of microdisk without Ag nanoparticle, the photoluminescence efficiency of Ag nanoparticle incorporated microdisk is significantly higher. This contradictory result, which includes a shorter lifetime and increased photoluminescence intensity, suggests that the effects are due to an increase in the spontaneous emission rate caused by resonance coupling between LSPs in Ag nanoparticles and excitons in MQWs, rather than a nonradiative relaxation process. Because the LSPs coupling path happens significantly faster than another non-radiative defect or phonon scattering, it increases the quasi-radiative exciton recombination and spontaneous emission efficiency in MQWs. The emission mechanisms involved in LSPs, in particular, reveal that the exciton energy is non-radiatively coupled to LSPs before radiative dispersion into the far-field by surface plasmon polaritons (SPPs). However, photon extraction from microcavity via surface SPPs remains a subject of contention for stimulated emission. While including Ag nanoparticles into this light extraction path increases emitting efficiency, it also reduces total internal reflection in cavities to obtain optical gain and mirror losses for stimulated emission.

**4. Conclusion**

In conclusion, we have fabricated plasmonic free standing InGaN/GaN microdisks on the Si (111) pedestals using wet-chemical undercut etching and incorporated Ag nanoparticles for improving WGMs resonance emission originated by LSPs effect. The radial resonance WGMs in optically pumped microdisk cavities have been observed. We believe that the low threshold condition WGM emitters can be enhanced by creating smooth side finesse, and reducing the diameter of the micro-disks. The presence of cavity modes in these InGaN/GaN MQW microdisks has benefits for wide applications of various lasing emitters.

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**Conflict of Interest**

The authors declare that they have no conflict of interest.

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**Figure captions**

FIG. 1. Schematic diagram and SEM image of hetero-epitaxial structure of GaN-based material used to make microdisks.

FIG. 2. Schematic diagrams of fabrication process flow for InGaN/GaN microdisks with incorporating plasmonic Ag nanoparticles on Si (111) substrates.

FIG. 3. SEM image of synthesized Ag nanoparticles (a), the absorption spectra of Ag nanoparticles stand-alone (b)

FIG. 4. A 20 μm microdisks without Ag nanoparticles: (a) SEM images, (b) TDPL spectra only show multiple F-P interference peaks without WGMs peaks.

FIG. 5. A 20 μm microdisks incorporated plasmonic Ag nanoparticles: (a) SEM images, (b) TDPL spectra show the WGMs resonance emission peaks at centered 442.3 nm. (c) The magnified area of WGMs lasing peaks.

FIG. 6. TRPL measured from a microdisks: Temporal and spectroscopic profile with (a) and without plasmonic Ag nanoparticles (b), and (c) decay lifetimes of both F-P interference emission peak (~480 nm) and LSPs coupled emission peak (~440 nm). Inset of (c) shows the expanded guided line for lifetime τ1,where the overlapped dashed line shows the TRPL fitting data using a biexponential model.

**Table captions**

Table I. TRPL fitting data using a biexponential model for the microdisks with and without plasmonic Ag nanoparticles.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| PL peak | | τ1 (ns) | A1 (%) | τ2 (ns) | A2 (%) |
| With Ag  nanoparticles | 440 nm | 0.36 | 78.6 | 3.36 | 21.4 |
| 480 nm | 0.48 | 76.0 | 5.81 | 24.1 |
| Without Ag  nanoparticles | 440 nm | 0.97 | 74.5 | 10.67 | 26.5 |
| 480 nm | 1.08 | 79.0 | 8.57 | 21.0 |