

Surface plasmon enhanced bright light emission from InGaN/GaN

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Surface plasmon (SP) coupling technique was used to enhance blue and green light emissions from InGaN/GaN quantum wells (QWs). Large enhancement of photoluminescence (PL) of both blue and green emissions was observed with silver coated samples, whereas the enhancements were not so effective for the gold coated samples. We could obtain well enhanced green emission by tuning the matching condition of QW-SP coupling with nano-grating structures at gold layers. This method should be useful to design even more efficient structures and to fabricate super bright light emitting devices.

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1 Introduction

InGaN/GaN quantum well (QW) based light-emitting diodes (LEDs) have been developed and expected to eventually replace traditional illumination sources. However, the emission efficacy of commercial white LEDs is still substantially lower than that of fluorescent tubes. Recently, we reported a method for enhancing the light-emission efficiency from InGaN QWs by controlling the energy transfer between QWs and surface plasmons (SPs) [1]. The SP has been used for many optical devices [2] and the idea of SP enhanced light emission was previously described [3–5]. For the first time, we directly measured significant enhancement of internal quantum efficiency (IQE) of InGaN/GaN by SP coupling [1]. We also observed a 32-fold increase in the spontaneous emission rate of InGaN/GaN at 440 nm probed by time-resolved photoluminescence [6]. Likewise, we have also reported similar light enhancement for dye-molecule doped polymers [7] and CdSe-based quantum dot nano-crystals [8].

In this paper, we also try to enhance the green emission of InGaN/GaN QW. The SP coupling technique can lead to enhancement of light emission at any wavelength region just by choosing the appropriate metal layer and geometry. High efficient InGaN emission with RGB colours should be useful to create a white light LED without a yellow phosphor.

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2 Experimental

GaN(10 nm)/InGaN(3 nm)/GaN(4 μm) QWs were grown on a sapphire substrate by a metal-organic chemical vapour deposition. A 50 nm thick silver or gold film was evaporated on top of the surface of wafers. After polishing the bottom surface of the samples, we photoexcite and detect emission from the backside of the samples through the transparent substrate. Such back side access to the QWs permit us the rapidly compare the PL from QWs with and without the influence of SPs. Photoluminescence (PL) measurements were performed by exciting the QW with a 406 nm diode laser and detecting the emission with a multi-channel spectrometer. Metal grating structures were fabricated by electron beam lithography on a 50 nm thick polymethylmethacrylate (PMMA) mask coated on the metal surface. The pattern was transferred into the top metal layer by using Ar ion milling.

3 Results and discussions

Figure 1(a) shows PL spectra from an InGaN/GaN QW which has blue emission with silver, gold, or no-metal coated [1]. The PL peak of the uncoated wafer at 470 nm is normalized to 1, and a 14-fold enhancement in peak PL intensity is observed from the silver coated emitter. No such enhancements were obtained from samples coated with gold. Figure 1(b) shows enhanced PL spectra with Ag and Au coating taken for an InGaN/GaN QW with green emission. 7-fold and 2-fold enhancements were obtained with Ag and Au coated samples. These PL enhancements should be attributed to QW-SP coupling. Electron-hole pairs excited within the QW couple to electron vibrations at the metal/semiconductor interface when the energies of electron-hole pairs in InGaN and of the metal SP are similar. Then, electron-hole recombination may produce SPs instead of photons, and this new recombination path increases the spontaneous recombination rate.

We obtained evidences that suggest the existence of the QW-SP coupling through several measurements and observations. (1) Enhanced PL intensities decrease exponentially with increasing GaN spacer thickness. As the SP is an evanescent wave that exponentially decays with distance from the metal surface, only electron-hole pairs located within the near-field of the surface can couple to the SP mode. (2) Obtained wavelength dependence of the PL enhanced ratios is clearly correlated to the dispersion diagrams of the SPs calculated with the dielectric functions. The dispersion diagrams of the SP modes at the metal/GaN interfaces are calculated by the dielectric functions of metal and GaN. (3) In order to separate the SP enhancement from other possible effects (mirror effect, photon recycling, increased scattering, resonant cavity effects, etc.), we have estimated the IQE of emission by the temperature dependence of the PL intensity. We found that the IQE values were actually increased, which is explainable only by spontaneous recombination rate enhancements through SP coupling. The IQE is a fundamental property and does not depend on the pumping method. Therefore, this technique should improve the efficiency of

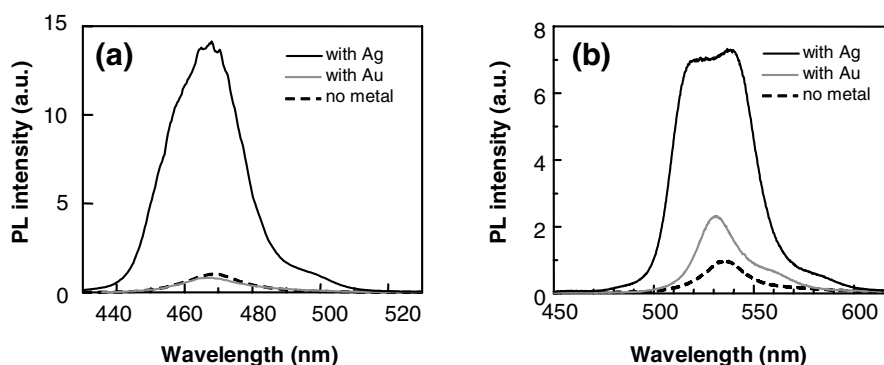


Fig. 1 SP enhanced (a) blue and (b) green emission from InGaN/GaN QWs coated with Ag or Au.

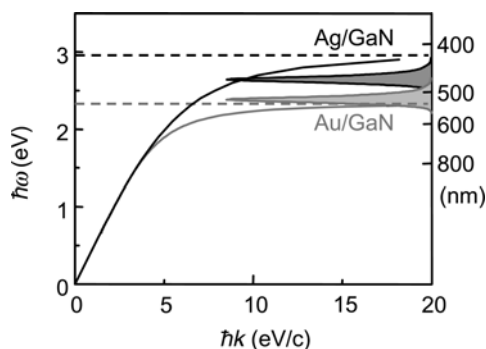


Fig. 2 Dispersion diagrams of the SP at Ag/GaN and Au/GaN interfaces. Blue and green emissions of InGaN/GaN QW are also plotted.

electrically pumped LED devices. (4) We also found that the spontaneous emission rates were also dramatically increased by the time-resolved PL measurement [6]. This high-speed emission property is also very important and would make possible desirable new technologies such as optical logical circuits and optical computing in the near future. These evidences give support to our proposed coupling mechanism.

Figure 2 shows the dispersion diagrams of SP on metal/GaN surfaces calculated by the dielectric functions. The SP frequencies of Ag/GaN and Au/GaN were calculated as ~ 3 eV and ~ 2.3 eV, respectively. This dispersion diagram shows that the density states of the SP mode become dramatically larger with approaching to the SP frequency. This large density of states increases the energy transfer from QW to SP and enhances the spontaneous emission rates, and this increases the IQE of emission. Since the green emission energy is a little far from the SP frequency of Ag/GaN, the SP enhancement ratio of green emission (7 fold) is smaller than that of blue emission (14 fold). The SP frequency of Au/GaN is lower than the energy of both blue and green emissions. Thus the PL intensities of both blue and green emissions are not enhanced with gold coating.

Other techniques to enhance the InGaN emission rates have already been reported by Walterelt and co-workers, who pioneered piezo-electric field free GaN/AlGaIn QW grown on M-plane of GaN substrate and observe about 10 times faster PL decay [9]. Wierer and co-workers have also reported InGaN/GaN LEDs within a photonic crystal, and report ~ 1.5 fold increases in light extraction [10]. Compared with these reports, our surface plasmon coupling method is much simpler and easier. We used just an evaporated metal layer, but we obtained still remarkable enhancement. Special nano-structural geometries or crystal growth techniques are not necessary for our method. For the SP coupling technique, the enhancement ratios are decided by the matching condition between the SP frequency and emission wavelength. Therefore, next most important problem is how to controllable the coupling conditions. There are several methods to tune the surface plasmon mode. For example, Paiella has proposed the method to tune the SP mode using metallo-dielectric multiple layers for emission enhancement [11]. Another method is using nano-structures of metal layers to localize the SP mode. The metal nano-structures are very important for the plasmon-photon coupling process. Metal nano-structures allow SPs of high momentum to scatter, lose momentum, and couple to radiative photon [12]. We believe that the nano-metal structures may be also useful to localize the SP modes and tune the SP frequencies.

Figure 3 shows the SP enhancement ratios of green light with nano-grating structured Au layers on InGaN/GaN. There is very clear coloration between the grating periods and the PL intensities. We believe that the nano-grating structures modify the SP frequencies and bring the good coupling condition between the SP at Au/GaN and green emission. The PL enhancement ratio reaches the maximum with 200 nm periods grating and this ratio (~ 7 fold) is similar to that of Ag/GaN (Fig. 1b). The nano-grating may influence also plasmon-photon coupling conditions (light extraction, scattering). Quantitative analysis is now in progress by using the finite difference time domain (FDTD) simulation of the localized SP modes. This may bring the more understanding of the mechanism and optimization of more effective geometry. We should be able to control both the QW-SP coupling and the SP-photon coupling by changing the widths and periods of nano-grating structures.

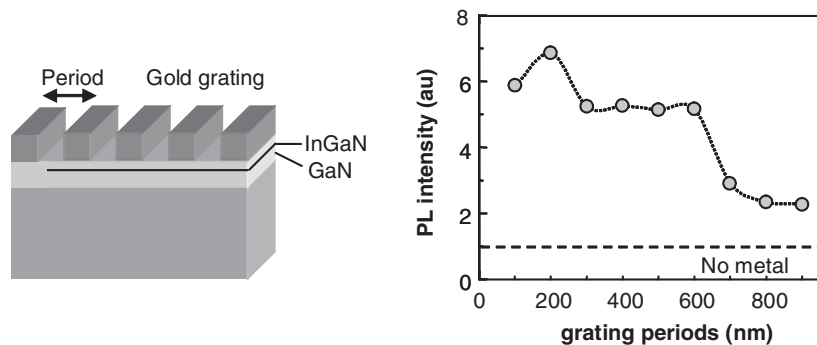


Fig. 3 Grating periods dependence of the SP enhanced ratios of green InGaN/GaN with Au.

The most important advantage of the SP coupling technique is potential to apply not only to the III–V materials, but also to various materials that suffer from low quantum efficiencies, which include the indirect semiconductors (Si, Ge, SiC, etc.). Usually the emission efficiencies of such indirect semiconductors are quite low, but it is possible to enhance these efficiencies to values as large as those available from direct compound semiconductors by the SP enhancement. Indeed, this technique have been tried to apply to the various materials after our first report. For example, the same technique has been used to ZnO [13], InN [14], Si-based nano-crystals [15–17], and CdSe-based nano-crystals [18, 19]. These many reports suggest that our proposed technique is very powerful and promising to obtain very bright light emission. Undoubtedly, this technique would lead the new super bright solid-state light sources, which could be very cheap to make, easy to process, and would become commonly used light sources. In order to develop LED device structures based on this technique, we need to make p- or n-doping and ohmic contact with very thin (10 nm) layers. These processes are now in progress.

4 Conclusion

SP coupling is powerful methods for developing efficient solid-state light emitters. To design and fabricate even more efficient SP-enhanced optical devices for a wider spectral range, tuning of the SP mode is very important to achieve matching condition of the energy coupling. The SP could be geometrically tuned by fabricating nano-grating structures. By using this technique, high-efficiency and high-speed light emission is predicted for optically as well as electrically pumped light emitters. Super bright plasmonic light emitters would become the dominant white light sources, as an alternative to conventional fluorescent tubes.

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References

- [1] K. Okamoto, I. Niki, A. Shvortser, Y. Narukawa, T. Mukai, and A. Scherer, *Nature Mater.* **3**, 601 (2004).
- [2] W. L. Barnes, A. Dereux, and T. W. Ebbesen, *Nature* **424**, 824 (2003).
- [3] J. Vuckovic, M. Loncar, and A. Scherer, *IEEE J. Quantum Electron.* **36**, 1131 (2000).
- [4] I. Gontijo M. Broditsky, E. Yablonvitch, S. Keller, U. K. Mishra, and S. P. DenBaars, *Phys. Rev. B* **60**, 11564 (1999).
- [5] A. Neogi, C.-W. Lee, H. O. Everit, T. Kuroda, A. Tackeuchi, and E. Yablonvitch, *Phys. Rev. B* **66**, 153305 (2002).
- [6] K. Okamoto, I. Niki, A. Scherer, Y. Narukawa, T. Mukai, and Y. Kawakami, *Appl. Phys. Lett.* **97**, 071102 (2005).
- [7] T. D. Neal, K. Okamoto, and A. Scherer, *Opt. Express* **13**, 5522 (2005).

- [8] K. Okamoto, S. Vyawahare, and A. Scherer, *J. Opt. Soc. Am. B* **23**, 1674 (2006).
- [9] P. Walterelt, O. Brandt, A. Trampert, H. T. Grahn, J. Menniger, M. Ramsteiner, M. Reiche, and K. H. Ploog, *Nature* **406**, 865 (2000).
- [10] J. J. Wierer, M. R. Krames, J. E. Epler, N. F. Gardner, M. G. Craford, J. R. Wendt, J. A. Simmons, and M. M. Sigalas, *Appl. Phys. Lett.* **84**, 3885 (2004).
- [11] R. Paiella, *Appl. Phys. Lett.* **87**, 111104 (2005).
- [12] W. L. Barnes, *Nature Mater.* **3**, 558 (2004).
- [13] C. W. Lai, J. An, and H. C. Ong, *Appl. Phys. Lett.* **86**, 251105 (2005).
- [14] B. Monemar, P. P. Paskov, and A. Kasic, *Superlattices Microstruct.* **38**, 38 (2005).
- [15] J. S. Biteen, D. Pacifici, N. S. Lewis, and H. A. Atwater, *Nano Lett.* **5**, 1768 (2005).
- [16] J. Kalkman, H. Gersen, L. Kuipers, and A. Polman, *Phys. Rev. B* **73**, 075317 (2006).
- [17] J. S. Biteen, N. S. Lewis, H. A. Atwater, H. Mertens, and A. Polman, *Appl. Phys. Lett.* **88**, 131109 (2006).
- [18] A. G. Brolo, S. C. Kwok, M. D. Cooper, M. G. Moffitt, C. W. Wang, R. Gordon, J. Riordon, and K. L. Kavanagh, *J. Phys. Chem. B* **110**, 8313 (2006).
- [19] H. Y. Lin, Y. F. Chen, J. G. Wu, D. I. Wang, and C. C. Chen, *Appl. Phys. Lett.* **88**, 161911 (2006).