

Surface plasmon enhanced InGaN light emitter

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ABSTRACT

We report a dramatic increase in the photoluminescence (PL) emitted from InGaN/GaN quantum wells (QW), obtained by covering these sample surface with thin metallic films. Remarkable enhancements of PL peak intensities were obtained from In_{0.3}Ga_{0.7}N QWs with 50 nm thick silver and aluminum coating with 10 nm GaN spacer. These PL enhancements can be attributed to strong interaction between QWs and surface plasmons (SPs). No such enhancements were obtained from samples coated with gold, as its well-known plasmon resonance occurs only at longer wavelengths. We also showed that QW-SP coupling increase the internal quantum efficiencies by measuring the temperature dependence of PL intensities. QW-SP coupling is a very promising method for developing the super bright light emitting diodes (LEDs). Moreover, we found that the metal nano-structure is very important factor to decide the light extraction. A possible mechanism of QW-SP coupling and emission enhancement has been developed, and high-speed and efficient light emission is predicted for optically as well as electrically pumped light emitters.

Keywords: surface plasmon, InGaN/GaN, light emitting diode, quantum well, internal quantum efficiency, solid-state light source

1. INTRODUCTION

Since 1993, InGaN quantum wells (QW)-based light emitting diodes (LEDs) have been continuously improved and commercialized as light sources in the ultraviolet and visible spectral regions.¹⁻³ Moreover, white light LEDs, in which a blue LED is combined with a yellow phosphor, have been commercialized and offer a replacement for conventional incandescent and fluorescent light bulbs.⁴ However, the promise of inexpensive solid state lighting has so far been delayed by the relatively poor extraction efficiency of light from semiconductor light sources. We believe that the development of efficient and bright white LEDs will rapidly result in commercialization of efficient solid state illumination sources. The most important requirement for a competitive LED for solid state lighting is the development of new methods to increase its quantum efficiency of light emission.

The external quantum efficiency (η_{ext}) of light emission from an LED is given by the light extraction efficiency (C_{ext}) and internal quantum efficiency (η_{int}). η_{int} in turn is determined by the ratio of the radiative (k_{rad}) and nonradiative (k_{non}) recombination rates of carriers.

$$\eta_{ext} = C_{ext} \times \eta_{int} = C_{ext} \times \frac{k_{rad}}{k_{rad} + k_{non}} \quad (1)$$

Often, k_{non} is faster than k_{rad} at room temperature, resulting in modest η_{int} . There are three methods to increase η_{ext} : (1) increase C_{ext} , (2) decrease k_{non} , or (3) increase k_{rad} . Previous work has focused on improving C_{ext} from InGaN LEDs by using the patterned sapphire substrates and mesh electrodes.⁵ However, further improvements of extraction of light through these methods are rapidly approaching fundamental limitations. Although much effort has recently been placed into reducing k_{non} by growing higher quality crystals,⁶⁻⁷ dramatic enhancements of η_{ext} have so far been elusive.⁸⁻⁹ On the other hand, there have been very few studies focusing on increasing k_{rad} ,¹⁰⁻¹¹ though that could prove to be most effective for development of high η_{ext} light emitters. In this article, we propose the enhancement of k_{rad} by coupling

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between surface plasmon (SP) and the InGaN QWs. If the plasmon frequency is carefully selected to match the QW emission frequency, the increase of the density states resulting from the SP dispersion diagram can result in large enhancements of the spontaneous emission rate. Therefore, energy coupling between QW and SP as described in this article is one of the most promising solutions to increase k_{rad} .

SPs, excited by the interaction between light and metal surfaces,¹²⁻¹³ are known to enhance absorption of light in molecules¹⁴, increase Raman scattering intensities¹⁵⁻¹⁶ and light transparencies¹⁷⁻¹⁸, and also generate photonic band gap¹⁹⁻²⁰. Since 1990, SPs have also received much attention when used in LEDs²¹⁻³⁰. Gianordoli et al optimized the emission characterization of GaAs-based LED by SP.²⁵ Vuckovic et al. reported the SP enhanced LED analyzing by both theoretically and experimentally.²⁶ Thus, great attention has been focused on SP enhanced emission. Hobson et al. reported the SP enhanced organic LEDs.²⁷ For InGaN QWs, Gontijo and co-workers reported the coupling of the spontaneous emission from QW into the SP on silver thin film²⁸ and showed increased absorption of light at the SP frequency. Neogi et al. confirmed that the recombination rate in an InGaN/GaN QW could be significantly enhanced by the time-resolved PL measurement.²⁹ However, in this early work, light could not be extracted efficiently from the silver/GaN surface. Therefore, the actual PL enhancement of InGaN/GaN by coupling into SP had not so far been observed directly. Quite recently, we have reported for the first time large photoluminescence (PL) increases from InGaN/GaN QW material coated with metal layers.³⁰ In order to design even more efficient structures and to fabricate electrical pumped LED devices by using SP coupling, we have to understand and optimize both mechanism and dynamics of energy transfer and light extraction. Here we fabricate and test nano-structured metal layers to explore the dependence of the plasmon enhancement on metal composition, thickness and grain shapes and sizes. The purpose of this work is to predictably use our control over metal geometries and composition to improve light emission and localization.

2. EXPERIMENT

Fig 1 shows the setup of the PL measurement and the sample structure. In_{0.3}Ga_{0.7}N/GaN QW wafers were grown on a (0001) oriented sapphire substrate by a metal-organic chemical vapor deposition (MOCVD). The QW heterostructure consists of a GaN (4 μm) buffer layer, an InGaN QW (3 nm) and a GaN cap layer (10, 40 or 150 nm), and the PL peak wavelength of the wafers is located at 470 nm. A 50nm thick silver film was evaporated on top of the surface of these wafers. After polishing the bottom surface of the QW 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, and to measure the dependence of the emission intensity on the distance between the QW and the metal films by changing the GaN spacer thickness. Topography measurements were performed by a twin-SNOM system manufactured by OMICRON. Fluorescence microscopy was used with X40 objective, a mercury lamp, and a color CCD camera. 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.

To perform the photoluminescence (PL) measurements, a cw-InGaN diode laser (406 nm) was used to excite

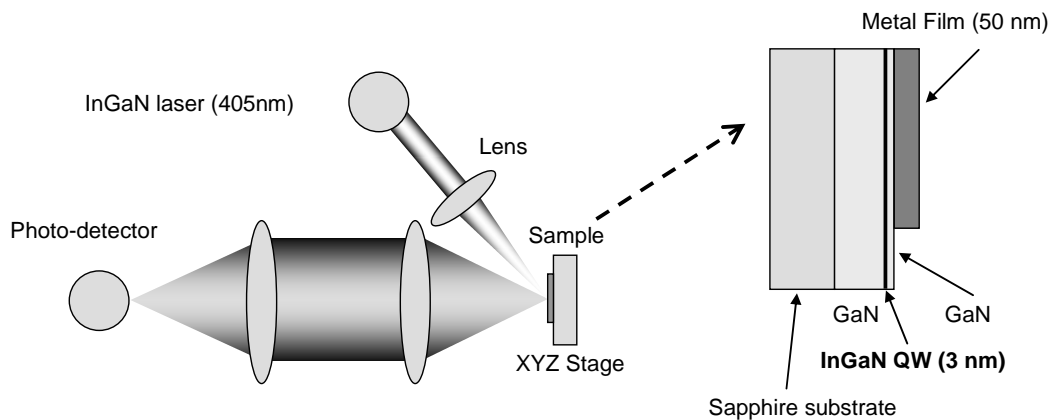


Fig. 1 Experimental setup of the photoluminescence measurement and the sample structure.

the QWs from the bottom surface of wafer. Luminescence was collected and focused into an optical fiber and subsequently detected with a multichannel spectrometer (Ocean optics). Neutral density filters were used to vary the excitation power (from 0.18 to 4.5 mW) to determine the power dependence of the luminescence intensities, and their temperature dependence was studied by using a cryostat with the ability of cooling from room temperature to 6K. To perform time-resolved PL measurements, frequency doubled beams of a mode-locked $\text{Al}_2\text{O}_3:\text{Ti}$ laser pumped by an Ar^+ laser were used to excite the QW from the backside of the wafer. A 1.5 ps pulse width, 400 nm pump wavelength, and 80 MHz repetition rate were chosen to excite luminescence in the QW. A streak camera system (Hamamatsu) was used as the detector.

3. RESULTS AND DISCUSSIONS

3.1 Enhanced photoluminescence spectra

Fig. 2a shows typical emission spectra from InGaN/GaN QW samples covered with silver layers. As the PL peak of the uncoated wafer at 470 nm was normalized to 1, it is clear that a dramatic enhancement in the PL intensity from the silver coated InGaN QWs can be obtained when the cap layer thicknesses is limited to 10 nm. On the other hand, the PL intensities are no longer strongly influenced from the silver in samples with 150 nm thick cap layers. The enhancement ratios of 10 nm capped QW samples covered with silver are 14-fold at the peak wavelength and 17-fold when comparing the luminescence intensity integrated over the emission spectrum with un-coated InGaN samples. We also compared the PL spectra of our QW samples after coating them with silver, aluminum, and gold layers (Fig. 2b). For InGaN QWs with a 10nm cap, such measurements indicate that a 8-fold peak intensity and 6-fold integrated intensity enhancement is obtained after coating with aluminum, and no enhancement in PL is found to occur in gold-coated samples. In such a measurement, a small ($2\times$) increase in the luminescence efficiency could be expected after metallization as the deposited metal reflects light back into the QW, and this may double the effective path-length of the incident pump light. Although the reflectivity of gold at 470 nm is smaller than that of silver or aluminum, this difference alone cannot explain the large difference in the enhancement ratio of each metal.

The dramatic PL enhancement of samples after coating with Ag and Al can be attributed to the strong interaction between the QW and SPs. We propose a possible mechanism of QW-SP coupling and light extraction shown in Fig. 3a. Electron-hole pairs created in the QW can couple to the electron vibration at the metal/semiconductor surface when the bandgap energy ($\hbar\omega_{\text{BG}}$) of InGaN active layer is close to the electron vibration energy ($\hbar\omega_{\text{SP}}$) of SP. Then, electron-hole recombination may produce a SP instead of a photon, and this new path of the recombination increases the spontaneous recombination rate. If the metal/semiconductor surface were perfectly flat, it would be difficult to extract light emission from the SP, since it is a non-propagating evanescent wave. However, in evaporated metal coatings, light emission can be observed as the SP is scattered through roughness and imperfections in the metal layers. The coupling

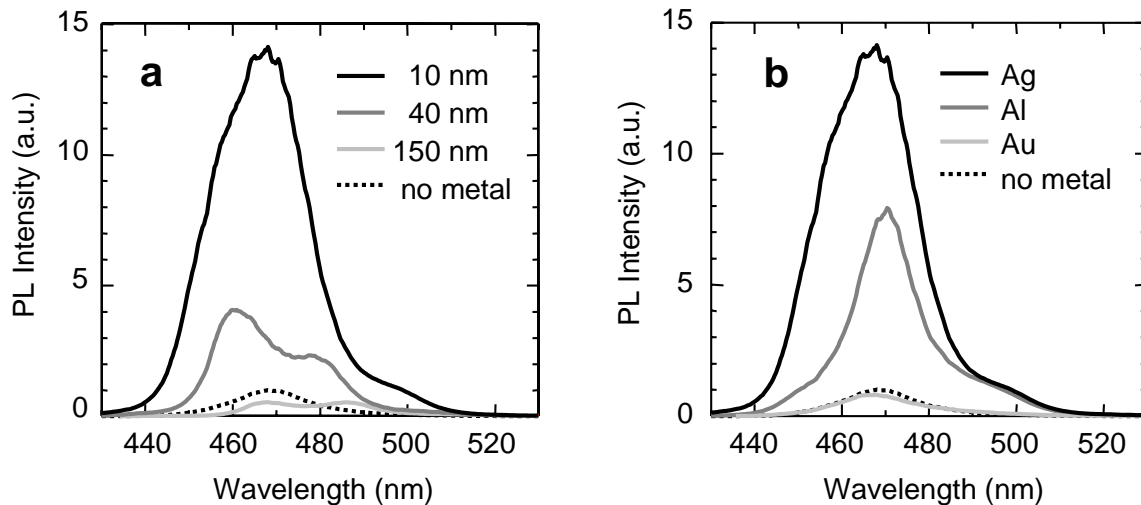


Fig. 2 a, PL spectra of InGaN/GaN QWs coated with silver layers with 10 nm, 40 nm and 150 nm thick GaN spacers. b, PL spectra of InGaN/GaN coated with Ag, Al and Au with 10 nm GaN spacers.

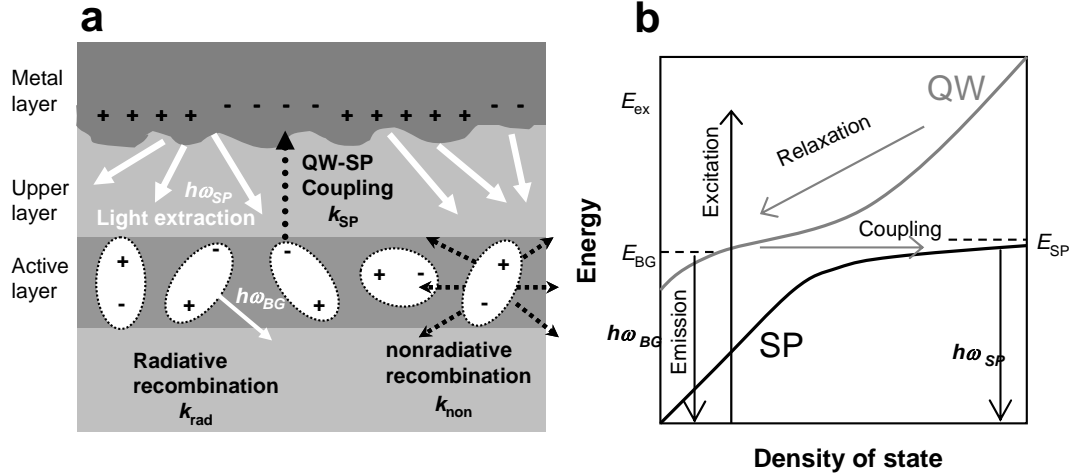


Fig. 3 a, Schematic diagram of the electron-hole recombination and QW-surface plasmon (SP) coupling mechanism. b, Energy diagram of excitation, emission and QW-SP coupling.

rate (k_{SP}) between the QW and SP is expected to be much faster than k_{rad} as a result of the large electromagnetic fields introduced by the large density of states (Fig. 3b). Actually, we observed such the enhanced spontaneous emission rates by the time-resolved PL measurement. All profiles could be fitted to single exponential functions and PL lifetimes (τ_{PL}) were obtained. We found that the time-resolved PL decay profiles of the Ag-coated sample strongly depend on the wavelength and become faster at shorter wavelengths, whereas those of the uncoated sample show little spectral dependence.³¹ We attribute the increases in both emission intensities and decay rates from Ag-coated samples to the coupling of energy between the QW and the SP.

3.2 Surface plasmon dispersion diagram

The dispersion diagrams of the SP modes at the metal/GaN interfaces are shown in Fig. 4a. The SP wave-vector (momentum) $k(\omega)$ was obtained by the following equation.¹²⁻¹³

$$k(\omega) = \frac{\omega}{c} \sqrt{\frac{\epsilon'_{metal}(\omega)\epsilon'_{GaN}(\omega)}{\epsilon'_{metal}(\omega) + \epsilon'_{GaN}(\omega)}} \quad (2)$$

where, $\epsilon'_{metal}(\omega)$ and $\epsilon'_{GaN}(\omega)$ are the real part of the dielectric functions for metal and GaN, respectively. The plasmon energy ($\hbar\omega_p$) of silver is well known as 3.76 eV.³² The SP energy ($\hbar\omega_{SP}$) must be modified for a silver/GaN surface, and can be estimated to be approximately ~ 2.8 eV (~ 440 nm) (Fig. 4a) when using the dielectric constant of silver³³ and GaN³⁴. $k(\omega)$ approaches infinity around ~ 2.8 eV by $\epsilon'_{metal}(\omega) + \epsilon'_{GaN}(\omega) \sim 0$. We have plotted a typical measured PL spectrum from the InGaN QW in Fig. 4a. The position of the PL peak was very close to $\hbar\omega_{SP}$, and large SP enhancements in luminescence intensity were observed especially at the higher energy side of the PL spectrum. This observation supports the existence of the QW-SP coupling phenomenon. Thus, silver is suitable for SP coupling to blue emission, and we attribute the large increases in luminescence intensity from Ag-coated samples to such resonant SP excitation. In contrast, the estimated $\hbar\omega_{SP}$ of gold on GaN is below ~ 2.2 eV (~ 560 nm), and no measurable enhancement is observed in Au-coated InGaN emitters as the SP and QW energies are not matched. In the case of aluminum, the $\hbar\omega_{SP}$ is higher than ~ 5 eV (~ 250 nm), and the real part of the dielectric constant is negative over a wide wavelength region for visible light.³⁵ Thus, a substantial and useful PL enhancement is observed in Al-coated samples, although the energy match is not ideal at 470 nm and a better overlap is expected at shorter wavelengths. Fig. 4b shows the enhancement ratios of PL intensities with metal layers separated from the QWs by 10 nm spacers as a function of wavelength. We find that the enhancement ratio increases at shorter wavelengths for Ag samples, while it is independent of wavelength for Al coated samples. The clear correlation between Figs.4a and 4b suggests that the obtained emission enhancement with Ag and Al is due to SP coupling.

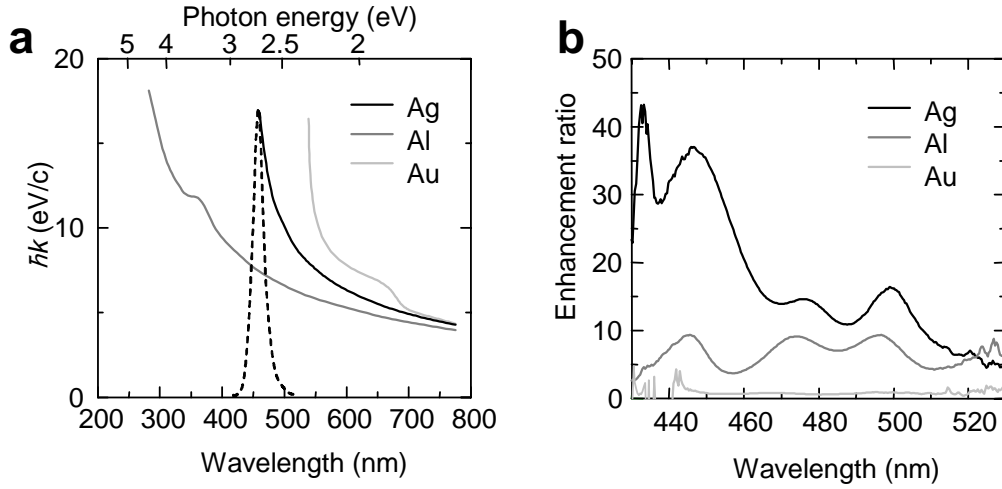


Fig. 4 a, Dispersion diagrams of surface plasmons generated on metal/GaN. The dashed line was the PL spectrum of InGaN/GaN. b, PL enhancement ratios at several wavelengths for same samples with Fig. 2b.

3.3 Spacer thickness and excitation power dependences

PL intensities of Al and Ag coated samples were also found to strongly depend on the distance between QWs and the metal layers, in contrast to Au coated samples. Fig. 5a shows this dependence of the PL enhancement ratios taken for three different GaN spacer thicknesses (of 10nm, 40nm, and 150nm) with each metal coating. These show an exponential increase in intensity as the spacer thickness is decreased for Ag and Al, but no significant improvement in the PL intensity for samples coated with gold. This figure suggests that coupling between SP should be main component to contribute to the PL enhancement, because the SP is an evanescent wave, which decays exponentially with increasing distance from the metal surface. Only electron-hole pairs located within the near-field from the surface can couple to the SP mode. The penetration depth $Z(\omega)$ of the SP fringing field into GaN from metal can be calculated from¹¹⁻¹²

$$Z(\omega) = \frac{c}{\omega} \sqrt{\frac{\epsilon'_{\text{GaN}}(\omega) - \epsilon'_{\text{metal}}(\omega)}{\epsilon'_{\text{metal}}(\omega)^2}} \quad (3)$$

$Z(\omega)$ is predicted to be $Z=47, 77,$ and 33 nm for Ag, Al, and Au, respectively at 470 nm. The inset of Fig. 5a shows good agreement between these calculated penetration depths (dashed lines) and measured values for Ag and Al coated samples. This again indicates that the emission enhancement results from QW-SP coupling.

We also find that the luminescence enhancement ratio increases with increasing excitation power (Fig. 5b). In InGaN QWs, electron-hole pairs are often localized by spatial modulations in bandgap energy produced by fluctuations of indium composition, QW width, or piezoelectric field. Such localization centers serve as radiative recombination centers for electron-hole pairs and explain the strong emission and insensitivity to growth defects in InGaN/GaN QW material. The emission efficiency may be reduced at high excitation intensities by saturation of these localization centers. When metal layers are coated within the near field of the QW, both localized and un-localized electron-hole pairs can immediately couple to the SP mode. In that situation, the saturation of the localized centers can be avoided and this leads to high emission efficiencies even under intense excitation. We consider this very advantageous in light emitting diodes, since generally the emission efficiencies of such emitters are reduced under the high current pumping. Thus, by using the SP coupling, higher current operation and brightness should be achievable.

3.4 Internal quantum efficiencies and Purcell enhancement factor

We expect that the SP coupling will increase the efficiency (η_{int}) by enhancing the spontaneous recombination rate. In order to estimate the η_{int} and to separate the SP enhancement from other effects (mirror effect, photon recycling, etc.), we have also measured the temperature dependence of the PL intensity. Fig. 6a shows the linear and Arrhenius plots of the integrated photoluminescence intensities of InGaN-SQWs coated with Ag and Al and compares these to un-coated

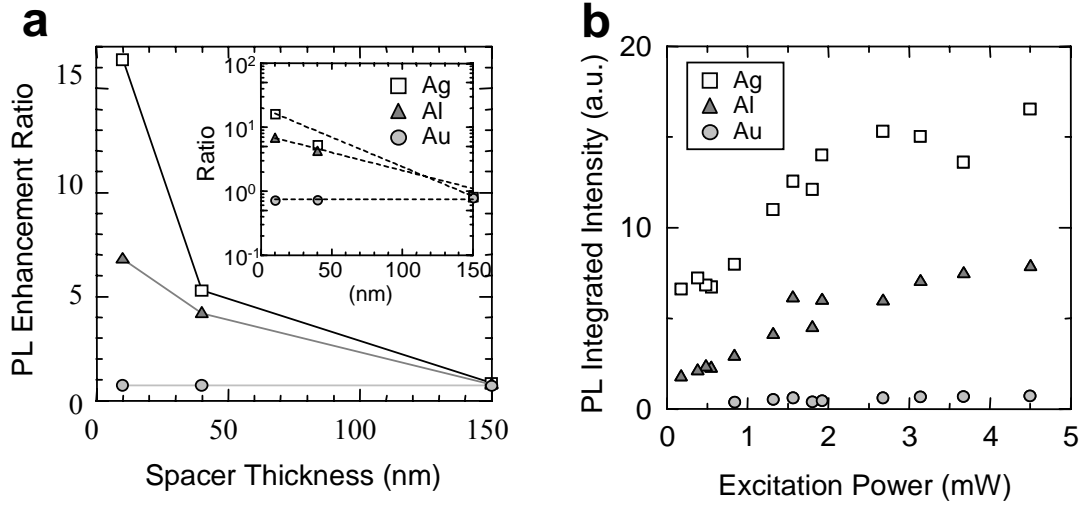


Fig. 5. GaN spacer thicknesses (a) and excitation power (b) dependence of the PL enhancement ratios.

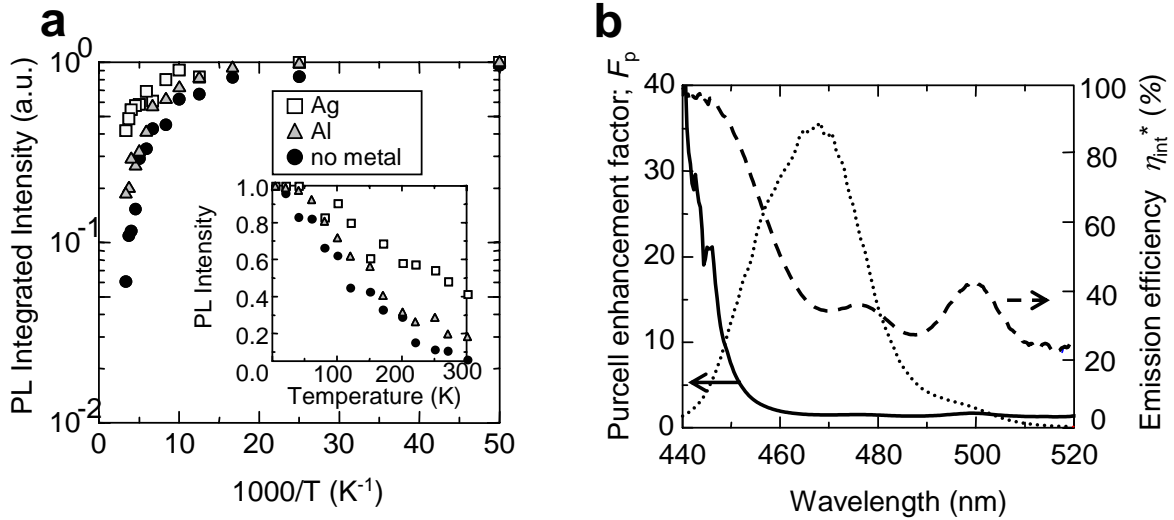


Fig. 6 a, Temperature dependence of integrated PL intensities. b, Wavelength dependent emission efficiencies [$\eta_{int}^*(\omega)$] was plotted as dashed line. The Purcell enhancement factor $F_p(\omega)$ estimated by $\eta_{int}^*(\omega)$ was also plotted (solid line). The dotted line was the PL spectrum of the same sample.

samples with 10 nm GaN spacer layer thicknesses. The η_{int} values of un-coated InGaN was estimated as 6 % at room temperature by assuming $\eta_{int} \sim 100\%$ at ~ 6 K.³⁶ We found that the η_{int} values were increased by 6.8 times (41%) by Ag coating and by 3 times (18%) by Al coating. We expect this actual enhancement of the η_{int} values to be a result of the enhancement of the spontaneous recombination rate of electron-hole pairs by SP coupling. 6.8-fold increasing of η_{int} means that 6.8-fold improvement of the efficiency of electrically pumped LED devices should be achievable because η_{int} is a fundamental property and not depend on the pumping method. Such improved efficiencies of the white LEDs, in which a blue LED is combined with a yellow phosphor, are expected to be larger than those of current fluorescent lamps or light bulbs. The luminous efficacy of commercial white LEDs is 25 lm/W under a current of 20 mA at room temperature.³⁷ This value is still lower than that of fluorescent tubes (75 lm/W). A 3-fold improvement is necessary to exceed the current fluorescent lamps or light bulbs. We expect that the SP coupling technique is very promising for even larger improvements of solid-state light source.

Wavelength depended enhanced efficiencies $\eta_{int}^*(\omega)$ can be related the coupling rate $k_{sp}(\omega)$ between QWs and SPs by the relationship:

$$\eta_{\text{int}}^*(\omega) = \frac{k_{\text{rad}}(\omega) + C'_{\text{ext}}(\omega)k_{\text{SP}}(\omega)}{k_{\text{rad}}(\omega) + k_{\text{non}}(\omega) + k_{\text{SP}}(\omega)} \quad (4)$$

where $C'_{\text{ext}}(\omega)$ is the probability of photon extraction from the SPs energy and is decided by the ratio of light scattering and dumping of electron vibration through non-radiative loss. Fig. 6b shows the $\eta_{\text{int}}^*(\omega)$ of Ag coated sample estimated from PL enhancement ratio (Fig. 6b) by normalizing the integrated η_{int}^* should be 41%. We find that $\eta_{\text{int}}^*(\omega)$ increases at shorter wavelengths where the plasmon resonance more closely matches the QW emission, and reaches almost 100% at 440 nm.

The Purcell enhancement factor F_p^{38} quantifies the increase in the spontaneous emission rate of a mode for a particular mode, and can be described by $\eta_{\text{int}}(\omega)$ and $\eta_{\text{int}}^*(\omega)$ when $C'_{\text{ext}} \approx 1$:

$$F_p(\omega) = \frac{k_{\text{rad}}(\omega) + k_{\text{non}}(\omega) + k_{\text{SP}}(\omega)}{k_{\text{rad}}(\omega) + k_{\text{non}}(\omega)} \approx \frac{1 - \eta_{\text{int}}(\omega)}{1 - \eta_{\text{int}}^*(\omega)} \quad (5)$$

Fig.6b also shows $F_p(\omega)$ estimated at each wavelength by assuming a constant $\eta_{\text{int}}(\omega) = 6\%$. $F_p(\omega)$ is significantly higher at wavelengths below 470 nm, well in agreement with previous work²⁸⁻²⁹. The PL spectrum shape (plotted as dotted line) also indicates that $F_p(\omega)$ values are higher at the shorter wavelength region. That should be a possible reason for the asymmetry in the luminescence peak of Fig. 2. Fig. 6b suggests that a InGaN QW with a peak position at around 440 nm should be best matched for SP enhancement from a silver layer. In that case, the enhanced $\eta_{\text{int}}^*(\omega)$ value is expected to approach 100% throughout the PL spectrum. The SP frequency could be geometrically tuned to match our $\lambda \sim 470$ nm QW by fabricating nanostructures, for example, using a grating structure, or using alloys.

3.5 Surface roughness and grating structures

The SP energy can be extracted as light by providing roughness or nano-structuring the metal layer. Such roughness allows SPs of high momentum to scatter, lose momentum and couple to radiated light.³⁹ $C'_{\text{ext}}(\omega)$ in Eq.(4) should depend on the roughness and nano-structure of the metal surface. We succeeded in controlling the grain structure within nano-sizes. Such roughness in the metal layer was observed from topographic images obtained by shear-force microscopy of the original GaN surface (Fig. 7a) and the coated Ag surface (Fig. 7b). The depth profiles along the dashed line in the topographic images are also plotted.

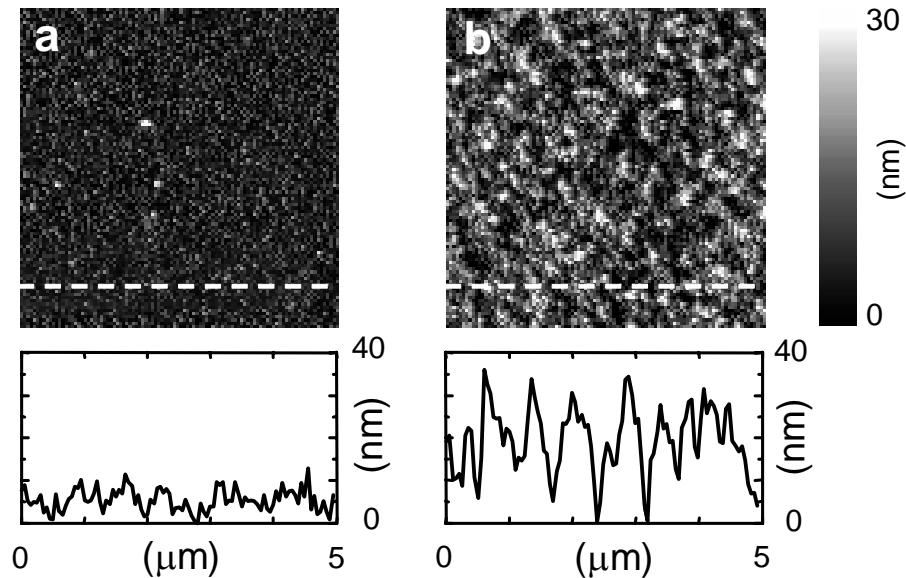


Fig. 7 Topographic image of the uncoated GaN surface (a) and the 50 nm thick Ag film evaporated on GaN (b). The depth profile along the dashed line in the topographic images are also plotted.

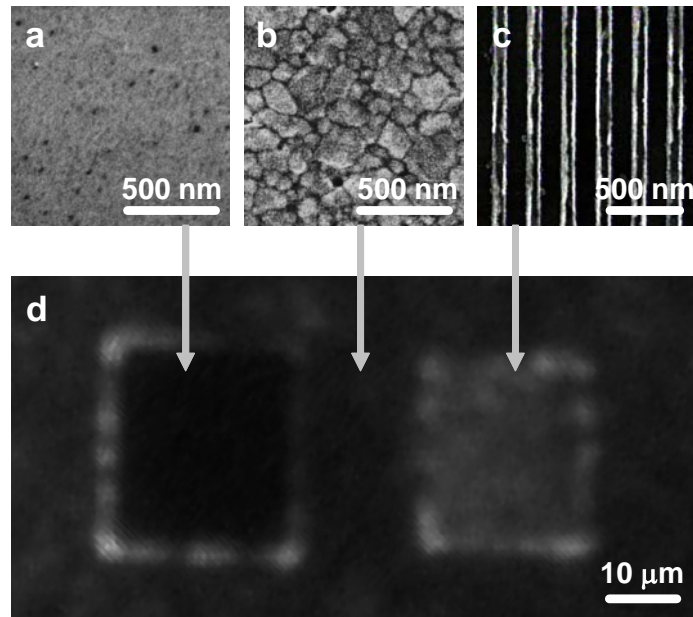


Fig. 8 SEM images of (a) the uncoated GaN surface, (b) the 50 nm Ag film on GaN, and (c) the grating structure with 33% duty cycle fabricated within a 50 nm thick Ag layer on GaN. (d) Micro-luminescence image including the areas of Figs. 3a-c.

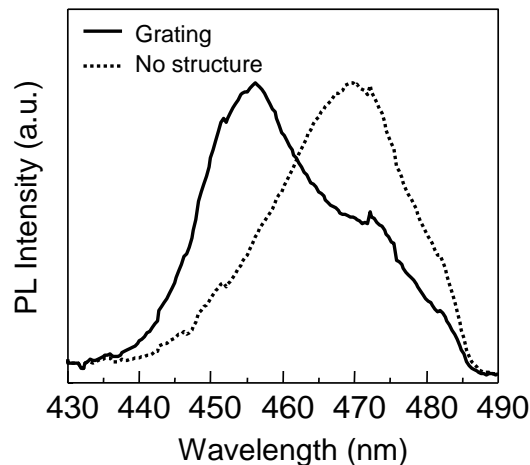


Fig. 9 PL spectra of InGaN/GaN QW with nano-grating structured and unstructured silver layers with 10 nm GaN

dashed lines of the Ag surface of approximately 30-40 nm while the GaN surface roughness was below 10 nm. Higher magnification SEM images of Ag and GaN surface are shown in Figs. 8a and 8b. The length scale of the roughness of Ag surface was determined to be a few hundred nanometers. Fig. 8c shows a fabricated metal grating, a geometry that has previously been used to couple SP and photons^{21, 23-26}. Micro-luminescence images of uncoated, coated, and patterned grating structures of Ag on InGaN QWs with 10 nm spacers are shown in Fig. 8d. We found a doubling of the emission from 133 nm wide Ag stripes forming a 400 nm period grating, whereas such an emission increase was not observed from 200 nm wide Ag stripes within a 600 nm period grating. This measurement suggests that the size of the metal structure determines the SP-photon coupling and light extraction. We also found that the PL peak position of grating structured regions was dramatically blue-shifted (Fig. 9). This suggests that the nano-grating structure modulate not only light extraction but also localized SP frequency. Such geometrical tuning of the SP frequency is one of the most important next subjects and is now on progress by experimentally and theoretically.

4. CONCLUSIONS

We conclude that the SP enhancement of PL intensities of InGaN is a very promising method for developing solid state light sources with high emission efficiencies. We have directly measured significant enhancements of η_{int} and the spontaneous recombination rate, and shown how distance and choice of patterned metal films can be used to optimize light emitters. Even when using un-patterned metal layers, the SP energy can be extracted by the submicron scale roughness on the metal surface. SP coupling is one of the most interesting solutions for developing efficient photonic devices, as the metal can be used both as an electrical contact and for providing high electromagnetic fields from SPs. We believe that this work provides a foundation for the rapid development of highly efficient and high-speed solid state light emitters alternative to conventional light bulbs.

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