Micro-photoluminescence mapping of surface plasmon-coupled emission from InGaN/GaN quantum wells

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Micro-photoluminescence (PL) mapping was investigated to elucidate the detailed mechanism of surface plasmon (SP)-enhanced emissions from InGaN/GaN quantum wells (QWs) with Ag or Al coating. The PL wavelengths were obviously red-shifted with Ag films, while the PL peak wavelengths were not changed with Al coating. The relationship between the PL peak intensity and the PL wavelength at each pixel showed a positive or negative correlation for the uncoated part of the InGaN/GaN QWs with blue or green emission, respectively. We found that these correlations disappeared in the Ag-coated regions. These results suggest that the energy transfer from the excitons to the SPs should be much faster than that in the exciton localization and charge screening processes of the piezoelectric field in QWs. These effects were not observed for the Al-coated regions because the mechanism of PL enhancement should be quite different as we have suggested previously.

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1. Introduction

Recently, it became known that surface plasmon (SP) resonance is a very promising method to increase emission efficiencies of several light-emitting materials. This approach had been theoretically predicted in 1999 by Ybronovich’s group,1,2 and was experimentally demonstrated for the first time by our group in 2004 for enhancing the visible emissions from InGaN/GaN quantum wells (QWs).3 The enhancement mechanism of light emissions using SP resonance is a type of Purcell effect. Spontaneous emission rates due to the radiative recombination processes of excitons in the active layer are enhanced when located in optical cavities. The SP resonance can act as an optical nano-cavity because it confines light waves at the metal/dielectric interface. The quality factors of plasmonic cavities are not so high because the free electron oscillations are easily damped; however, the mode volumes are very small. The SP resonance can confine light waves into nanospace much smaller than the wavelength. By this feature, plasmonics have been expected to develop a new class of optical nanotechnology and nanophotonics. We also experimentally observed that the SP resonance enhanced not only emission efficiencies but also emission decay rates.4,5 The obtained Purcell factor of blue emission of InGaN/GaN with Ag film reached 500 at 440 nm, which is the wavelength with the best SP resonance for Ag films.5

One of most important targets of SP-enhanced light emission is device applications.6 Recently, a great number of groups have tried to develop high-efficiency plasmonic light-emitting diodes (LEDs) based on InGaN/GaN QWs.7–12 In spite of a lot of efforts and reports for plasmonic LEDs, the enhancement factors have so far been only twice as large as that by electrical pumping, although more than tenfold enhancements were achieved for photopumped photoluminescence (PL). In order to achieve a greater enhancement factor from this method, the further optimization of both device and metal structures by deeply understanding the SP coupling mechanism is necessary. There are many reports on SP-enhanced light emission especially using InGaN/GaN QWs, but the detailed enhancement mechanisms related to the exciton emission mechanisms are still not so clear. For example, the internal quantum efficiencies of InGaN/GaN QWs strongly depend on the exciton dynamics in the active layers, such as exciton localization13,14 and the quantum confinement Stark effect (QCSE).15,16

Spatial resolved evaluations of the PL features using microscopic PL measurements are very useful to measure and elucidate the exciton dynamics which strongly affect the internal quantum efficiencies of emissions. For example, we found special inhomogeneties in both PL peak intensity and wavelength mapping using a scanning near-field optical microscope (SNOM),17 cathodoluminescence,18 and also a confocal scanning laser microscope.19 We also found that both the exciton localization and the QCSE can be evaluated by these microscopic measurements by the correlations between the PL peak and wavelength mapping.20 In this study, we used a normal fluorescent microscope with moving stage and evaluated the exciton dynamics under the SP resonance to understand its detailed mechanisms.

2. Experimental methods

The experimental setup of the micro-PL mapping system and the sample structures are shown in Fig. 1. Blue- and green-emitting InGaN/GaN (3 nm/10 nm) QWs were grown on undoped GaN (4 μm thick)/sapphire substrates by metal organic chemical vapor deposition. The thickness of the GaN spacer layers was 40 nm. 50-nm-thick Ag or Al films were deposited onto half of the sample wafer with a thermal vapor deposition system. PL spectra were measured with a fluorescence microscope using a mercury lamp for excitation (400–410 nm) and a spectrograph (Spectra Pro 2300i) and charge-coupled device camera (PIXIS100). A motorized
moving stage was controlled by a stepping motor for scanning with a 2 μm step. All of the PL spectra obtained at each pixel were fitted with a Gaussian function, and the values of the PL peak wavelength and intensity were extracted from the Gaussian fitting result.

3. Results and discussion

Before the PL measurements, we observed the microscopic reflection images to verify the borderline of the metal coating. The obtained reflection intensity mapping for Ag-coated (top) and Al-coated (bottom) green-emitting InGaN QWs is shown in Fig. 2. We can clearly see the metal-coated region and the borderline because the reflectivity of Ag and Al is much larger than that of GaN.

Figure 3 shows the obtained PL mappings of both PL peak intensities and wavelength for green-emitting InGaN/GaN QWs with half Ag coating. The obtained PL mapping exhibited some spatial fluctuation at the micro-meter scale mainly due to the inhomogeneities of the indium compositions. The dotted lines are the boundary between the metal-coated and uncoated regions detected by the reflectance mapping. Clearly the PL intensities were enhanced in the Ag-coated regions and also the PL peak wavelength was red-shifted.

Figure 4 shows a similar PL mapping for green-emitting InGaN/GaN QWs with Al coating. We found that the PL peak wavelengths were not changed by the SP resonance with Al, while the PL peak intensities became much larger. This result was quite different to that of the Ag-coated samples. This discrepancy should be due to the difference of the enhancement mechanism with Ag and Al films as previously reported. The shortest wavelength limit of the SP mode at the Ag/GaN interface is ~430 nm, estimated by the dispersion relationship of the propagation mode of the SP polaritons. The excitation light around 400 nm wavelength cannot resonate to the SP mode at the Ag interface, and the PL enhancement should be attributed to the SP resonance with green emissions. On the other hand, the shortest wavelength limit of the SP mode at Al/GaN is ~250 nm. For this reason, the SP resonance with Al film should be useful to enhance deep UV emissions. For example, we succeeded in producing a huge enhancement of the UV emission around 250 nm from AlGaN/AlN QWs with Al film. For a very broad wavelength region of the SP mode at the Al interface, the excitation light at 400 nm can also resonate with the SP mode and induce a giant electrical field to the Al film. This is likely to cause an enhancement of light absorption and exciton densities, and must be the main origin of the huge PL enhancements on the Al coating. On the other
The PL wavelength mapping was changed with Ag spontaneous emission rates. This should be the reason why the emission mechanism; for example, it increases the light. The SP resonance with visible emissions should modify emissions. The wavelength in deep UV regions is far from the visible should be not so effective because the vast resonant hand, the SP resonance with emission at the Al interface (black circles) part of blue- and green-emitting InGaN/GaN QWs.

Fig. 5. (Color online) Relationship between the PL peak intensity and the PL wavelength at each pixel for the Ag-coated (colored circles) and uncoated (black circles) part of blue- and green-emitting InGaN/GaN QWs.

Next, focusing on Ag coating, we consider the mechanism of the PL peak shift. In order to understand the mechanism in more detail, we plot the relationship between the PL peak intensities and the wavelength for blue and green emission in Fig. 5. For the uncoated regions shown as black marks, the regions with higher PL intensities seemed to have longer PL wavelengths for InGaN with blue emissions. This positive correlation is similar to the findings of our previous reports and should be due to the exciton localization effects. The generated excitons diffuse in the QW layer and are localized into indium-rich regions, which seem to have quantum dot structures. The localized excitons are well protected against the thermal loss and have high emission efficiencies. For this reason, the region of longer PL wavelength must be much brighter than the shorter wavelength regions in blue emission. In this study, we could observe the positive correlation of the PL intensity and wavelength even by using a normal fluorescent microscope with mercury lamp excitation. The spatial distributions of the indium composition may have several levels of spatial scales, namely, fractal structures. Therefore, our observed exciton dynamics with few-micron scale were similar to that with submicron scales usually observed by SNOM or cathodoluminescence.

Figure 5 shows that the PL peak wavelength was blue-shifted and the positive correlation disappeared by Ag coating for the blue-emitting InGaN/GaN. This result suggests that the energy transfer from the excitons to the SPs on Ag film are faster than the localization processes. If the SP resonance increases the emission rate faster than the exciton localization, the exciton localization effects should be canceled. Then the positive correlation should disappear and the PL wavelength is blue-shifted. In our previous reports, the spontaneous emission rate with the SP resonance was in the order of sub-nanoseconds, while the exciton localization process was nanoseconds, as measured by time-resolved SNOM. This fast decay feature of the SP resonance is a very promising property to apply to plasmonic optical devices. The Purcell factor can be increased further by optimizing the plasmonic nanostructures, and further improvement with picosecond-order decay may be possible. This would bring a ~100 GHz modulation speed using InGaN LEDs similar to the InGaN laser. The exciton localization process brings in many cases positive factors to improve the emission efficiencies of LEDs; however, it sometimes has negative factors when applied to laser diodes. A canceling feature for the exciton localizations of the SP-enhanced emissions would be useful to apply to low-threshold laser diodes in the future.

Opposite to the blue emission, negative correlations were observed for InGaN QWs with green emissions. Also, the energy distributions of the PL peak wavelength were much broader compared with those of the blue emission. We already reported these properties of green-emitting InGaN/GaN QWs as due to an increase in the number of defects as non-radiative recombination centers and also the QCSE. With increasing the indium composition, the indium-rich region become larger and larger by the phase separation. In these cases, the exciton localization process became ineffective. The excitons captured into indium-rich regions still can diffuse inside the indium-rich region and are easily trapped in...
the non-radiative recombination centers such as threading dislocations, point defects, and impurities. Then, the emission efficiencies of the exciton in the indium-rich regions are dramatically reduced, which leads to the broad PL peak distributions and also the negative correlation between the PL peak intensities and wavelength. Moreover, the QCSE can also be the origin of these optical properties. In the indium-rich regions, the internal piezoelectric field becomes larger and separates the spatial interactions between the wave functions of electrons and holes of excitons. Then, the emission efficiencies become much lower, the spontaneous emission rates become slower, and the bandgap energies of the InGaN QW become smaller. This is likely to be the reason for the negative correlation. It is well known that the QCSE can be screened when the exciton densities are very high after the photo excitation. After the excitation, with decreasing the exciton density, the screening effect was reduced and the QCSE become more remarkable. This is likely to be the reason for the very broad distribution of the PL peak wavelength.

Figure 5 shows that the negative correlation also disappeared with the SP resonance with the Ag coating. One contribution of the SP resonance is similar to that of the blue emissions; the SP resonance canceled the exciton localization because the SP-enhanced emission rate should be faster than the exciton localization. For the green emission, the exciton localization causes the negative correlation, so the SP resonance can cancel it and also induce a blue-shift of the PL peak wavelength. Figure 5 shows that the main part of the PL peak distribution was blue-shifted by the Ag coating. However, some parts of the PL peak distribution were red-shifted. The other important process is the influence of the SP resonance on the QCSE. As described above, the SP resonance increases the decay rates of the excitons. The excitons generated in the QW should rapidly decay by the SP resonance, therefore the exciton density becomes much lower immediately. This should prevent the screening of the QCSE, and the QCSE becomes much stronger. This should cause the red-shift of the PL peak wavelength shown in Fig. 5. Therefore, the contribution of the SP resonance to the green-emitting InGaN/GaN is more complicated than that of the blue emissions. The SP resonance canceled both the exciton localization and the screening of the QCSE. By these effects, the wavelength distribution of the PL peak wavelength becomes very narrow for the green emission. This is also a very important effect to consider in the photonic application of plasmonics.

All these effects of the SP resonance with Ag were not observed for the SP resonance with Al because the enhancement mechanisms are quite different. Opposite to the SP resonance with Ag, the screening of the QCSE can be enhanced by the increasing of the exciton density by the light absorption enhancement by the SP resonance with the excitation light at the Al interface. The giant electric field generated at the Al interface by the SP resonance is also able to cancel the QCSE. We have already observed such preliminary results using Al films although we didn’t include these results in this study. We will report the details of the canceling QCSE effect of the SP resonance with Al at the next opportunity.

4. Conclusions
We investigated the micro-PL measurement for blue and green emissions of InGaN/GaN QWs with Ag or Al in order to understand the detailed mechanism of the emission enhancement. The obtained PL peak intensity and peak mapping with Ag and Al were quite different as a result of the difference in the emission enhancement mechanism. Especially for the Ag-coated samples, we found a remarkable change in the correlation between the PL peak intensities and wavelength. By analyzing the results, we found that the SP resonance with Ag coating canceled both the exciton localization effect and the QCSE because the SP resonance enhanced the PL decay rate faster than the exciton localization process and reduced the screening of the QCSE. In this study, we found that the SP resonance could control not only the emission rates and efficiencies but also both the exciton localization and the QCSE in the QWs. These exciton dynamics are very important factors to determine the optical properties and functions of the InGaN/GaN QWs. Based on this method, much more flexible control and tuning of the optical properties and functions should be available by further optimization of these effects.

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