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Enhancement of light-extraction efficiency of SiN$_x$ light emitting devices through a rough Ag island film

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Based on an ITO/Ag/SiN$_x$/p$^+$-Si/Al structure, a significant enhancement of the external quantum efficiency was achieved compared with the device without Ag island film. Analysis showed that the increase of the light-extraction efficiency resulted from the surface roughening of ITO electrode has a main contribution to this enhancement. The increase of the internal quantum efficiency induced by the enhancement of spontaneous emission rate and the carrier injection level also has an instructive contribution. Our results demonstrate that localized surface plasmons enhanced SiN$_x$-based light emitting devices show great promise for the development of efficiency Si-based optical device.

Recent years, SiN$_x$-based light emitting devices (LEDs) have received extensive attentions as a candidate of light source for silicon photonics due to the lower turn-on voltage and higher luminescence intensity resulting from the smaller barriers for carrier injection compared to SiO$_2$-based LEDs. Also, the preparation of the SiN$_x$-based LEDs can be compatible with the current complementary metal-oxide semiconductor (CMOS) techniques. However, the extraction efficiency of electroluminescence (EL) from SiN$_x$-based LEDs is still too low to meet the demands of optical interconnections.

Approaches proposed to improve the extraction efficiency of EL from SiN$_x$-based LEDs can be divided into two classes depending on whether it changes the spontaneous emission rate: (1) increase of the flow of light emitted from the active layer by the modification of the surface morphology and (2) enhancement of the internal quantum efficiency (IQE) by the modification of spontaneous emission rate.

Among all the approaches employed in these references, the application of localized surface plasmons (LSPs, the collective oscillation of excited free electrons confined to metallic nanoparticles) has recently received more and more attentions. By employing the structure with Ag nanoparticles underneath the active layer, Kim et al. enhanced the EL intensity of SiN$_x$-based LEDs by 4.34 times. It would be favorable to the integration of current CMOS technique for the structure with Ag on the active layer. Meanwhile, this structure would experience a simple technological process. Therefore, we proposed the use of Ag island film deposited on the active layer to increase the light extraction efficiency of SiN$_x$-based LEDs.

SiN$_x$ film was deposited onto the p/p$^+$-Si (100) substrate (the thickness and resistivity of the epitaxial layer were 18.5 ± 1 μm, 9 ± 1.5 Ω cm, respectively) by plasma enhanced chemical vapor deposition (PECVD), in which N$_2$-diluted 10% SiH$_4$ and NH$_3$ were used as the reactant gas sources with the flow rate ratio fixed at 10:1 (Si/N = 1). The rf power, chamber pressure and substrate temperature for the growth were maintained at 20 W, 0.1 Torr, and 400°C, respectively. The thickness of SiN$_x$ film was about 50 nm measured by ellipsometry (J. A. Woollam M-2000D). After the deposition of SiN$_x$ film, the sample was annealed in N$_2$ at 800°C for 1 h. An Ag layer was deposited by magnetron sputtering afterwards. Ag island film, with the optimal particle dimension regulated by the sputtering time, was formed after the rapid thermal process (RTP) in Argon at 500°C for 60 s. Finally, a top transparent ITO circular electrode and a rear Al metal contact were deposited by magnetron sputtering and e-beam evaporation, respectively. For comparison measurements, a SiN$_x$-based LED without Ag island film was also fabricated. Same samples without Al rear electrode were prepared on Si and quartz substrates for the test of photoluminescence (PL) and extinction spectra, respectively. An optical gap of 3.5 eV can be obtained from the fitting of extinction spectra by Tauc plot.

The current-voltage (I-V) characteristics of the SiN$_x$-based LEDs were measured using a Keithley 4200 SCS semiconductor parameter analyzer. A HITACHI U-4100 spectrophotometer was used to measure the extinction spectra of the devices. Atomic force microscope (AFM) was used to study the size and shape of Ag island film, also the surface morphology of ITO with and without Ag.

Figs. 1(a) and 1(b) show the schematic diagram of the SiN$_x$-based LED structure and the surface morphology of Ag nanoparticles on SiN$_x$ without ITO contact, respectively. As shown in Fig. 1(b), Ag island film was formed after the treatment of RTP, with the dimension of Ag particles was around 100 nm in diameter. The device with Ag particles of this size was the optimal one as it has the maximum external quantum efficiency (EQE) (not shown here). After the deposition of ITO, the root means square (RMS) roughness of the surface decreased significantly, from 17.5 nm to 5.5 nm, as shown in Fig. 1(c). However, the surface of ITO without Ag island film was quite smooth with the RMS roughness of 1.9 nm only, as shown in Fig. 1(d).

This surface roughening of ITO electrode induced by the introduction of Ag island film can reduce the
inner-reflection of light and significantly increase the light extraction efficiency of the device.\textsuperscript{11,12} We estimated the contribution of this roughening on the enhancement of the light extraction efficiency of LEDs quantitatively based on the total integrated scatter (TIS), which was defined as the total power scattered into a hemisphere divided by the incident power.\textsuperscript{4} Under the assumption that the scattering mainly occurs at the regular reflection direction, the TIS is given as

\[
\text{TIS} = \left( \frac{2 \pi \delta \Delta n}{\lambda} \right)^2,
\]

where $\delta$ is the RMS roughness of the ITO surface and $\Delta n$ is the refractive index difference between the ITO and the luminescence medium.\textsuperscript{4} Theoretically, an eight-fold enhancement of the EL extraction efficiency of SiNx-based LEDs can be achieved by this roughening.

The EL spectra of SiNx-based LEDs with and without Ag island film were measured under the same injected current (30 mA), as shown in Fig. 2(a). The EL for the reference device, with the peak wavelength at about 600 nm, was originated from the recombination of electrons and holes at defects levels in the SiNx bandgap which were in the range from 1.6 to 3.1 eV.\textsuperscript{13} With the introduction of Ag island film, the EL peak split up to two Gauss ones, at about 500 and 620 nm in wavelength, and the intensity increased significantly. We attributed both peaks to the radiative recombination between the defects levels in the SiNx bandgap.\textsuperscript{13–15} The appearance of these two EL peaks and the broadening of the EL band width might be resulted from the improved electron injection of ITO side after the addition of Ag island film as it can induce a large enhancement of the localized electric field at the interface.\textsuperscript{16} The radiative recombination between the electrons located at conduction band tail and holes confined in the center of $\equiv \text{Si}^0$ became obvious after the addition of Ag. A maximum EQE enhancement of around 6.5 times can be observed by the introduction of Ag, as shown in Fig. 2(b). Meanwhile, the stability of the device was improved significantly as it can suffer a higher breakdown voltage and a higher injection current comparing with the reference device. An electrical breakdown was occurred to the reference device as we applied the voltage larger than 11 V.

As have been mentioned above, LSPs also have an instructive effect on the enhancement of the EL extraction efficiency via the modification of spontaneous emission rate.\textsuperscript{4,8} Fig. 3(a) compares the enhancement factor of the intensity of EL spectra with that of PL spectra, which was defined by the ratio of the luminescence intensity of the device with Ag island film to the values of the reference one. The wavelength of maximum EL enhancement was very close to that of PL one, indicating that the enhancement of PL and EL...
were originated from the same source, which can be attributed to the excitons-LSPs coupling. This coupling can introduce an additional path of recombination, which can increase the spontaneous emission rate via Purcell effect. The extinction spectra shown in Fig. 3(b) further confirmed this conclusion as the consistency between the peak of resonance absorption and that of luminescence enhancement at long wavelength. The peak of luminescence enhancement at around 480 nm might be resulted from the improvement of the flow of light emitted from the active layer. This improvement was resulted from the minimum absorption of the Ag island film, as shown in Fig. 3(b). Therefore, we concluded that the increase of the spontaneous emission rate resulted from the coupling between excitons and LSPs also has an instructive contribution to the EQE enhancement of the SiN-based LED. As have been mentioned above, the origin of the enhancement of PL was mainly from the increase of IQE resulted from the enhancement of spontaneous emission rate, and the improvement of light-extraction efficiency. The absorption, backscattering, and the surface coverage of Ag particles should also be considered. Hence, this increase of IQE might not be very substantial as the enhancement of PL intensity was not obvious, as shown in Fig. 3(a).

From the analyses above, the contributions of the ITO surface roughening and the excitons-LSPs coupling on the enhancement of EL extraction efficiency have already exceeded the overall increase of the EQE of the device. However, the serious backscattering induced by the high surface coverage (>60%) of Ag particles would make it difficult for light to escape from the active layer. This phenomenon would be more serious by considering the absorption of Ag island film. Consequently, there might be other factors which would affect the EL extraction efficiency of SiN-based LEDs. As the EQE is the product of the IQE, the light-extraction efficiency, and the carrier injection efficiency, the injection of carriers should also be considered.

For this reason, J-V characteristics were carried out for the devices with and without Ag island film as shown in Fig. 4. The threshold voltage of the device with Ag was significantly lower than that of the reference one. Meanwhile, a larger current density under the same operating voltage was achieved in this improved device, indicating a great enhancement in carrier injection level. We attributed this improvement in carrier injection to the increased surface roughness due to the addition of Ag island film, which would induce a large enhancement of the localized electric field at the interface. Due to the existence of an additional path of recombination mentioned above, carriers can transfer their energies into LSPs rapidly keeping the carrier density in the luminescence layer low even though the injection level was improved significantly. As a result, the device with Ag island film can suffer a higher injection current than the reference device accompanied by the improvement of the stability of the device as mentioned above. However, the accumulation of carriers at the interface states between the contact electrode and the active material would screen the applied electric field, for which the effective injection was limited. Consequently, the improvement of the carrier injection might also have an instructive contribution to the enhancement of EL extraction efficiency.

In summary, the enhancement of EL intensity and EQE were observed for SiN-based LED with Ag island film deposited on it. A maximum EQE enhancement of 6.5 times was achieved by this improved device. We attributed this significant increase mainly to the improvement in light extraction efficiency, which was caused by the surface roughening of ITO electrode induced by the addition of Ag island film. The increase in radiative recombination rate of excitons in SiN and the improvement of the carrier injection level also played a partial role in this distinct enhancement. Further improvement of the EL performance of SiN-based LED can be achieved by the optimization of the surface coverage of Ag particles. Our experimental results provide an alternative approach towards the fabrication of SiN-based or Si-based LEDs with high EL extraction efficiency.

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