Enhanced optical characteristics of light-emitting-diode by localized surface plasmon of Ag/SiO₂ nanoparticles

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1. Introduction

The surface plasmon (SP) coupling is known to have interesting optical characteristics due to the excitation of resonant collective oscillations of the electrons by electromagnetic radiation [1]. The nano-sized metallic particles are used to obtain the phase matching and strong enhancement of the SP, known as localized surface plasmon (LSP) [2]. Recently, these LSP have attracted great interest in light-emitting-diode (LED), because the optical properties can be greatly enhanced by LSP coupling between the metallic particles and the quantum well (QW) [1-5]. This distinctive phenomenon has been intensively studied in In-GaN/GaN based LED using various metals [1-3, 5]. Generally, various metal films were deposited on the surface with near active layer and then annealed to form the metal nanostructure [1-6]. However, the revealed metal nano-particles in air are easily oxidized and the absorbance spectrum of LSP is changed [7].

In this study, we fabricated the Ag/SiO₂ core/shell nano-particle (NP) by sol-gel method to prevent the oxidation and changing the absorbance spectrum. Also, we investigated the optical characteristics applied to InGaN/GaN based LED by LSP coupling.

2. Experimental procedure

Ag core/SiO₂ shell

All chemicals used were Aldrich grade including tetraethoxysilane (TEOS), silver nitrate hydroxide. De-ionized water was used in all experiments. A typical preparation procedure was performed as followed [8]: a 500 mL beaker was filled with 180 mL of aqueous solution including cetyl trimethyl ammonium bromide (0.145g) under vigorous magnetic stirring. A freshly prepared aqueous solution of silver nitrate (0.1 M, 10 mL) was then added to the mixture solution. 20 mL of ascorbic acid aqueous solution as reducing agent was added to the mixture solution in 5 min. After the mixture was further stirred for 10 min, sodium hydroxide (0.1 M) was added to speed the reaction and the pH of the mixture solution was about 5. Subsequently, 50 mL of ethanol and 1 mL of TEOS was added into the above-mentioned silver colloids, respectively. The solution was stirred for another 3 h at room temperature (RT).

InGaN/GaN based LED

The InGaN/GaN multiple-quantum-well (MQW) epitaxial structure was grown by metal organic chemical vapor deposition (MOCVD) technique. Trimethylgallium (TMGa), trimethylindium (TMIn) and NH₃ were used as precursors for Ga, In and N, respectively. A thermal annealing of c-plane sapphire substrate was carried out at 1100 °C for 10 min, followed by the growth of a low temperature GaN buffer layer. A 400 nm thick undoped GaN layer was grown at 1060 °C. Then, five pairs of In-GaN/GaN MQW were grown on the undoped GaN layer. The GaN barriers and InGaN wells were grown at temperatures of 850 °C and 750 °C, respectively. A 10 nm thick undoped GaN layer was grown on it as the spacer layer.

The morphology of the Ag/SiO₂ NPs was investigated by transmission electron microscopy (TEM). The UV-visible spectroscopy and photoluminescence (PL) and low temperature PL measurement were carried out to study the optical properties. A 25mW, He-Cd laser (325 nm) was used as the excitation source for the RT. Time-resolved PL (TRPL) was performed under excitation of frequency-doubled (398 nm) laser pulses from a Ti:sapphire mode locked laser and the signals were analyzed by streak camera with an overall resolution of 15 ps.

3. Results and discussion

Figure 1 shows the typical TEM images of Ag/SiO2 NPs. It is clearly seen that Ag NPs were spherical and completely covered by SiO_2 shell. As shown in Fig. 1, Ag core and SiO_2 shell had a size of around 50 nm and 20 nm,

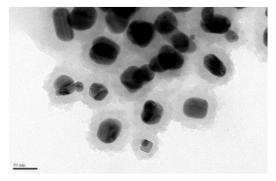


Fig.1. TEM images of the SiO₂-coated Ag nanoparticles.

respectively.

The synthesized Ag/SiO₂ NPs were coated on the surface of the LED structure by droplet with a 10 mL and drying at RT. The back-side PL responses from the MOWs with Ag/SiO₂ NPs and absorbance spectra of NPs are shown in Fig. 2. Compared with the uncoated sample (reference), the Ag/SiO₂ coated sample showed a remarkable increase in the PL by about 50 %. This optical enhancement is associated with the LSP coupling between the MQWs and Ag/SiO₂ NPs [1-6]. As evident from Fig. 2, Ag/SiO₂ NPs exhibited the absorbance spectra at 475 nm and suitable for LSP coupling to blue emission. To clearly investigate the LSP coupling, the TRPL measurements were performed in Fig. 3. The decay time of reference sample and Ag/SiO_2 NPs coated sample were 139 ns and 104 ns, respectively. These results mean that the faster decay time is obtained from the LSP coupling of Ag/SiO₂ NPs [3, 5] and the enhanced PL result is related to the increase in the spontaneous emission rate, suggesting effective coupling of LSP-MQW.

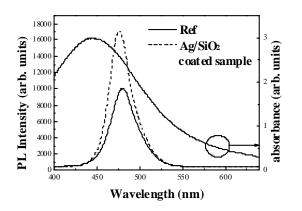


Fig.2. Room temperature PL spectra from the reference and the Ag/SiO₂-NP-coated sample, and the absorbance spectra of the NPs.

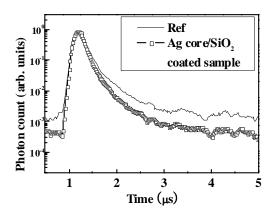


Fig.3. Time resolved PL spectra of the reference and the Ag/SiO_2 -NP-coated sample at room temperature.

3. Conclusions

We investigated the LSP coupling behavior in In-GaN/GaN based LED by Ag/SiO_2 NPs. The PL intensity of Ag/SiO_2 coated sample was enhanced by 50 % and the absorbance spectra was suitable for LSP coupling to blue emission. From the TRPL measurements, the faster decay time of Ag/SiO_2 NPs coated sample indicates that the enhanced luminescence intensity was attributed to the energy transfer from MQW to Ag/SiO_2 NPs by LSP coupling.

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