

Improved electron injection and efficiency in blue organic light-emitting diodes using coupled electric field near cathode

Lingling Deng^{a,b}, Zhijie Zhou^a, Bolun Jia^a, Hongwei Zhou^a, Ling Peng^a, Wenjuan Shang^a, Jing Feng^c, Shufen Chen^{b,*}

^a School of Optoelectronic Engineering, Nanjing University of Posts and Telecommunications, Nanjing, 210023, China

^b Key Laboratory for Organic Electronics and Information Displays and Institute of Advanced Materials, Nanjing University of Posts and Telecommunications, Nanjing, 210023, China

^c State Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun, 130012, China

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ABSTRACT

In this paper, we report high-performance blue organic light-emitting diodes (OLEDs) based on evaporated silver nanoclusters (SNCs) located near the aluminum cathode. We investigate the influence of localized surface plasmon resonance (LSPR) and trapping effects of SNCs on the performances of blue OLEDs depending on the size, distribution, and location of the SNCs. The research results reveal when the SNCs are located in the electron transporting layer, the serious trapping effect would cause reduced current density and efficiency of our blue OLEDs. Fortunately, when the SNCs are close to the cathode, owing to the electric field coupling effect between the LSPR of SNCs and the surface polarization of aluminum cathode, the resonant electric field is concentrated near the cathode, thereby enhancing the electron injection. The balanced carrier injection and transport processes improve the performance of blue OLEDs. The optimized device with SNCs located 2 nm away from the aluminum cathode achieves the highest current efficiency of 40.7 cd/A, enhancing by 15% compared to the control device without SNCs.

1. Introduction

Localized surface plasmon resonance (LSPR) are electromagnetic surface modes activated by the coupling between the optical field and the surface charge in metal nanostructures particularly of silver (Ag) and gold (Au) nanoparticles (NPs). LSPR greatly enhances the intensity of the electromagnetic field around the NPs and then affects the fluorescence of dye molecules locating near the metal NPs. Due to these special properties, metal NPs have drawn considerable attention in a range of applications including bio-sensing, surface enhanced spectroscopy and optoelectronics [1–3].

Organic light-emitting diodes (OLEDs), as a promising candidate for next generation of display and illumination, have attracted much attention to improve their performance [4–6]. Over the past decades, metal NPs have been applied in OLEDs to improve the device efficiency, efficiency roll-off, and stability [7–12]. Several properties of metal NPs help to improve the performance of NP-based OLEDs. The main mechanism is fluorescence enhancement caused by LSPR. When the metal NPs are placed near the emission excitons, the enhanced electric field around NPs can accelerate the recombination of excitons and increase

the spontaneous radiation rate, thereby enhancing the luminous intensity. In this process, the proper distance between NPs and emission excitons is very important because too short a distance leads to exciton quenching, while the long distance cannot excite the interaction between NPs and excitons [7–10]. Secondly, the optical scattering of metal NPs is another feature that helps OLEDs improve performance. In conventional flat OLEDs, the total internal reflection occurring at the interface between different materials reduces the light extraction efficiency. When metal NPs are introduced into OLEDs, the special nanostructures help to enhance optical scattering and decrease internal reflection, which improves the light extraction [11,12].

However, the performance of metal NP-based OLEDs reported in previous work is relatively low. Some researchers revealed that one of the main negative effects comes from trapping effect of metal NPs, which captures carriers, breaks the carrier balance and reduces the internal quantum efficiency [13,14]. In other reports, metal NPs were placed on the electrodes to improve the carrier injection by enhancing the electric field or changing the work function [12,15,16]. Moreover, the LSPR effect of metal NPs is usually discussed in isolation, rarely considering the influence of surrounding materials in OLEDs. The

* Corresponding author.

E-mail address: iamsfchen@njupt.edu.cn (S. Chen).

working mechanism of metal NPs in OLEDs is complicated and still not illustrated clearly up to now. So the systematic studies on optical and electrical properties of metal NPs are necessary for the application of NPs in high-performance OLEDs.

In this paper, silver nanoclusters (SNCs) were introduced into the electron transporting layer (ETL) of a group of blue OLEDs. The size, distribution, and location of the SNCs were changed to systematically investigate the LSPR and trapping effects of SNCs in high-performance OLEDs. Particularly, the effect of SNCs near the aluminum (Al) cathode on the device performance was studied in detail and the distance between the SNCs and the Al cathode was optimized. Finally, the optimized blue OLED achieved the improved electron injection and current efficiency by using the coupled electric field between the SNCs and the Al cathode.

2. Experimental

The OLEDs were fabricated on glass substrates with patterned indium-tin oxide (ITO) layer with a sheet resistance of $10 \Omega/\square$. The ITO-coated glass substrates were cleaned with acetone, ethanol and deionized water sequentially for 15 min using an ultrasonic bath, and then dried completely in an oven at 100°C . After treated with UV-ozone for 6 min, the substrates were spin-coated with poly(3,4-ethylenedioxythiophene) doped with poly(styrenesulfonate) (PEDOT: PSS, AI 4083) at 2000 rpm for 1 min as hole transporting layer (HTL), and then annealed at 110°C for 30 min. Then, a blend solution of 2,6-bis(3-(9H-carbazol-9-yl)phenyl)pyridine (26DCzPPy), 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC), and iridium(III) [bis(4,6-difluorophenyl)pyridinato-N,C^{2'}] (Firpic) (the weight ratio of 26DCzPPy: TAPC: Firpic is 16:4:3) was dissolved in chlorobenzene and spin-coated at 2000 rpm for 1 min and then annealed at 80°C for 30 min to form the first emission layer (EML). The first EML containing the cohost of 26DCzPPy and TAPC helps the hole injection and reduces the operating voltage. Then, the substrates were loaded into the evaporation chamber to deposit the other films, including Firpic doped 26DCzPPy, 1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPB), LiF/SNCs, and LiF/Al. Here, Firpic doped 26DCzPPy acts as the second EML, which extends the exciton recombination zone and suppresses the efficiency roll-off. TmPyPB and LiF/Al are used as ETL and composite cathode respectively. LiF before the SNCs serves as the electron injection layer to suppress the trapping effect of the SNCs. During the deposition, the base pressure in the chamber was maintained as low as 6×10^{-4} Pa. The deposition rates of all organic layers were 0.1 nm/s, and Firpic guest was deposited at 0.01 nm/s to acquire a doping concentration of 10 wt%. The SNCs were deposited at 0.01 nm/s and the Al cathode was deposited at 0.5 nm/s. Thicknesses of all the deposited layers were monitored by a quartz crystal oscillators.

The device structures are ITO/PEDOT: PSS (45 nm)/26DCzPPy: 20 wt% TAPC: 13 wt% Firpic (30 nm)/26DCzPPy: 10 wt% Firpic (15 nm)/TmPyPB (50-x nm)/LiF (0.5 nm)/SNCs (y nm)/TmPyPB (x nm)/LiF (1 nm)/Al (130 nm), as shown in Fig. 1. Under the premise that the total thickness of TmPyPB layers keeps 50 nm in all OLEDs, we vary the thicknesses of TmPyPB layers before and after SNCs to investigate the influence of the SNC location. The thicknesses of SNCs are set as 0.1, 0.5 or 1 nm to obtain different sizes and distribution densities. The current density–luminance–voltage (J-L-V) characteristics of the devices were measured by the recombination of a Keithley 2400 (Keithley instruments Inc., USA) source measurement unit with a Photo-Research-655 (Photo Research Inc, USA) Spectroscan Colorimeter. The absorption spectra were measured by a U-3900 spectrophotometer (Hitachi, Japan). The photoluminescence (PL) spectra were measured by an IK series (Kimmon Koha, Japan). The scanning electron microscopy (SEM) images were measured with an S-4800 scanning electron microscopy (Hitachi, Japan). All the experimental results were measured at room temperature under air ambient.

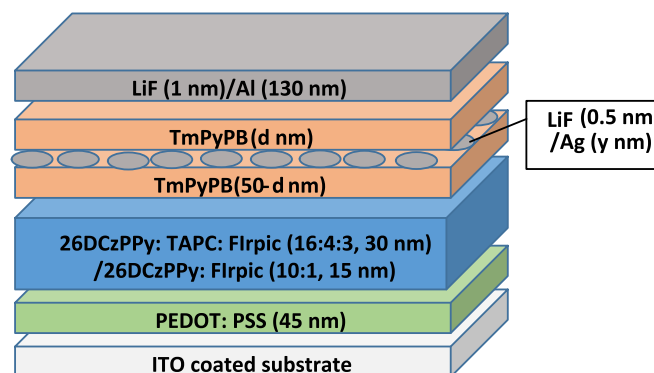


Fig. 1. Schematic structure of OLEDs with SNCs at different locations.

3. Results and discussion

3.1. Characteristics of SNC layers

SNCs were fabricated with different thicknesses of 0.1, 0.5, and 1 nm, and their SEM images in Fig. 2 (a)–(c) show that the SNCs spread discretely on the substrate. As the SNC thickness increases, the particle size increases, and the number of the bigger SNC also increases, which influences the LSPR effect of the SNCs. The absorption spectra in Fig. 2 (d) show the extinction peak is slightly red-shifted and the intensity is enhanced greatly with the increase of the SNC thickness, which demonstrates the absorption spectrum of SNCs is mainly determined by those bigger SNCs. Besides, we can find a significant overlap between the absorption spectra of the SNCs and the EL spectrum of Firpic, indicating the emission from Firpic can excite the LSPR of the SNCs.

For SNCs with a thickness of 0.1 nm, it exhibits a very weak absorption intensity. In order to figure out the LSPR effect of 0.1 nm SNCs, we fabricated a group of samples with structures of EML/TmPyPB (z nm)/Ag (0.1 nm)/TmPyPB (50-z nm), where $z = 5, 10, 20$, and tested their LSPR-enhanced photoluminescence (PL). Moreover, a control sample without SNCs was also prepared for comparison. The samples were illuminated with a monochromatic light with wavelength of 350 nm and their PL spectra were recorded in Fig. 2 (e). From the figure, we find the PL intensity decreases when the thickness of spacer layer between the SNCs and the EML is 5 nm due to the fluorescence quenching. With the spacer layer increased to 10 and 20 nm, the PL intensity was significantly enhanced because the enhanced fluorescence usually occurs when the distance between metal nanoparticles and exciton is 10–20 nm [18]. The PL results demonstrate there exists surface plasmon-exciton coupling between the excitons and the 0.1 nm SNCs, although the absorption of SNCs is very weak.

Besides LSPR, SNCs work as carrier traps, suppressing the carrier transport in OLEDs due to their lower energy levels. The influence of SNCs on the carrier transport is related to their distribution density [17]. We fabricated a series of blue OLEDs with different SNC densities (i.e., different SNC thickness) and a control device without SNCs. We find the current densities of the devices decrease obviously with the increase of SNC density (as shown in Fig. 3), which significantly affects the device performance. So in the following discussion, we used the 0.1 nm SNCs with weak trapping effect to investigate the influence of SNC position on LSPR enhanced fluorescence and carrier transport in OLEDs.

3.2. Electrical and efficiency properties of devices with SNCs locating near the EML

Fluorescence enhancement from metal NPs is critically related to the distance between the NPs and the emission excitons in OLEDs [18]. NPs are usually introduced near the EML to enhance emission, so we first investigate the LSPR and trapping effects of SNCs by changing the

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