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Effect of coupling between excitons and gold nanoparticle surface plasmons on emission behavior of phosphorescent organic light-emitting diodes

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ABSTRACT

Enhanced efficiency and reduced efficiency roll-off in phosphorescent organic light-emitting diodes (PhOLEDs) are realized by interposing a solution-processed gold nanoparticle (GNP)-based interlayer between the anode and the hole-injection layer. Transient photoluminescence measurements elucidate that a reduced lifetime of the triplet excitons was observed for samples having a GNP-interlayer as compared to a control sample without the GNP-interlayer. The decrease in the triplet exciton lifetime, caused by the coupling between the triplet excitons and the localized surface plasmons (LSPs) excited by the GNPs, enables reducing the triplet–triplet and triplet–polaron annihilation processes, thereby a reduced efficiency roll-off in PhOLEDs. The presence of a GNP-interlayer also acts as an optical out-coupling layer contributing to the efficiency enhancement and was demonstrated by the theoretical simulation.

1. Introduction

Phosphorescent organic light-emitting diodes (PhOLEDs) have attracted a significant attention for application in flat panel displays and solid-state lighting [1,2]. Many progresses have been made in improving the performance of PhOLEDs, including high power efficiency tandem structure incorporating bulk heterojunction organic bipolar charge generation layer [3]. The accomplishment in bi-directional and symmetrical illumination semitransparent white PhOLEDs also offer additional features and design freedoms for application in planar diffused lighting [4]. Phosphorescent iridium complex based emitters, e.g., fac-tris(2-phenylpyridinato-N,C2′)iridium(III) [Ir(ppy)$_3$] doped in 4,4′-bis(9-carbazolyl)-1,1′-biphenyl (CBP) host, have been widely used. The phosphorescent emitters allow harvesting both singlet and triplet excitons achieving 100% internal quantum efficiency [5], opening up a plethora of opportunities for making highly efficient PhOLEDs. However, there is still a room for further improvement in the efficiency via device optimization. Efficiency roll-off in PhOLEDs, a decrease in the electroluminescence (EL) efficiency at a high current density or luminance, still remains an open challenge. Efficiency roll-off in PhOLEDs is predominantly caused by the triplet–triplet annihilation (TTA) and the triplet–polaron annihilation (TPA) due to the long radiative lifetime of the triplet excitons [6,7]. Different methods have been proposed to reduce the efficiency roll-off in PhOLEDs [8–13]. It is well known that the exciton quenching process due to TTA is proportional to the square of the density of triplet excitons, while the exciton quenching arising from TPA increases linearly with the density of the triplet excitons [9]. Under operation condition, the exciton density is in proportion to the exciton lifetime [7]. Consequently, the TTA is proportional to the square of lifetime of triplet exciton and the TPA scales linearly with the lifetime of triplet exciton. Zhang et al. reported using a short-lived organoeuropium emitter to reduce efficiency roll-off [14], revealing the impact of decrease in the lifetime of triplet excitons on reduced efficiency roll-off in PhOLEDs.

Noble metal nanoparticles have generated considerable interest in recent years due to their unique optical properties that are clearly different from those of the bulk [15–19]. One of these properties is localized surface plasmon (LSP) resonance, which strongly affects the kinetic characteristics of nearby molecules [20]. The incorporation of gold nanoparticles (GNPs) in organic light-emitting diodes (OLEDs) has shown to enhance device performance, due to the LSP effect [21–24]. The effect of GNPs on light out-coupling efficiency in OLEDs is less reported. A systematic study is a prerequisite for better understanding the effect of GNPs on the overall performance of PhOLEDs.

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In this work, the performance of GNP-incorporated PhOLEDs, with a distribution of GNPs over the diameter range from 9 to 15 nm on ITO/glass substrates, was investigated. Time-resolved photoluminescence (PL) measurements were used to study the change in the decay rate of triplet excitons in $Ir(ppy)_3$ doped in CBP host, an archetypical electrophosphorescent material system, due to the interaction between triplet excitons and GNPs. Out-coupling effect of GNP-interlayer in PhOLEDs was measured and analyzed using theoretical simulation.

2. Experiments

A set of PhOLEDs, with a structure of glass/ITO/GNPs/MoO_3 (2.0 nm)/CBP (13 nm)/CBP:$Ir(ppy)_3$ (12 wt%, 30 nm)/1,3,5-tri (1-phenyl-1H-benzo[d]imidazol-2-yl)phenyl (TPBi, 45 nm)/LiF (1.0 nm)/Al (100 nm), was fabricated. In this device configuration, MoO_3, CBP, CBP:$Ir(ppy)_3$, TPBi, and LiF/Al were used as the hole injection layer (HIL), hole transporting layer (HTL), green emission layer, electron transporting layer (ETL), and cathode, respectively. Following a previous work [24], a stack of a 2.0 nm thick MoO_3 HIL and a 13 nm thick CBP HTL was adopted, serving as a spacer to optimize the emission output of PhOLEDs. The layer thickness and deposition rate of the materials were monitored *in situ* using an oscillating quartz thickness monitor and the deposition rate of the functional organic materials, LiF and metal cathode was controlled at 0.2, 0.01, and ~0.5 nm/s, respectively. GNP-covered ITO/glass substrates with different GNP concentrations were formed by spin-coating GNP-solutions on the substrates following an annealing at 350 °C in air. Optical density (or absorbance) measurements were carried for solutions with different GNP concentrations. Optical density (OD) is a logarithmic ratio of light falling upon GNP solution to light transmitted through the solution. In this work, PhOLEDs made with GNP-covered ITO/glass substrates, using GNP solutions with different OD values estimated at light wavelength of 540 nm, e.g., devices A (OD = 26), B (OD = 20), and C (OD = 13), were fabricated. The GNPs were chemically synthesized according to the previous literature [25] and the values of the OD were estimated from the diluted GNP solution. A control device having an identical structure made with a bare ITO anode, device D, was also fabricated for comparison studies. The current density–voltage–luminance characteristics and EL spectra of the PhOLEDs were measured by a programmable Keithley model 2400 power supply and a Photo-research PR650 spectrometer at room temperature in air. The devices were not encapsulated. A set of samples having a stack of MoO_3 (2.0 nm)/CBP (13 nm)/CBP:$Ir(ppy)_3$ (30 nm) functional layers on GNP-coated ITO substrates, formed using GNP solutions having different OD values of 26 (S1), 20 (S2), 13 (S3), and a control stack without the GNP-based interlayer (S4) for comparison studies, were prepared. The absorption spectra were measured by an ultraviolet/visible spectrometer (UV 1700, Shimadzu). Transient PL measurements were carried out using Edinburgh Instruments FL920 Spectrometer, using a pulsed laser (average power of ~5 mW) with the wavelength of 405 nm and pulse duration of ~79 ps. All the GNP interlayers on substrates, formed with GNPs in toluene with different OD values, were spin-coated at 1600 rpm and annealed at 350 °C in a glove box (MBRAUN) for 30 min. The morphology of the GNP-covered ITO surfaces with different GNP densities was analyzed using Scanning Electron Microscope (SEM) (Hitachi S4800).

3. Results and discussion

SEM images measured for GNP-covered ITO/glass surfaces having different GNP concentrations, deposited using GNP solutions with different OD values of (a) 26, (b) 20 and (c) 13, are shown in Fig. 1. GNPs on ITO surface have a GNP distribution over the diameter range from 9 to 15 nm. It is clear that GNP-covered ITO with high GNP density generally form relatively larger-sized GNPs, induced by the aggregation of GNPs and the post-annealing process. The coverage of GNPs on ITO surface decreases with the reduction in the concentration of GNPs in the solution. Fig. 2a shows the normalized absorption spectra measured for samples

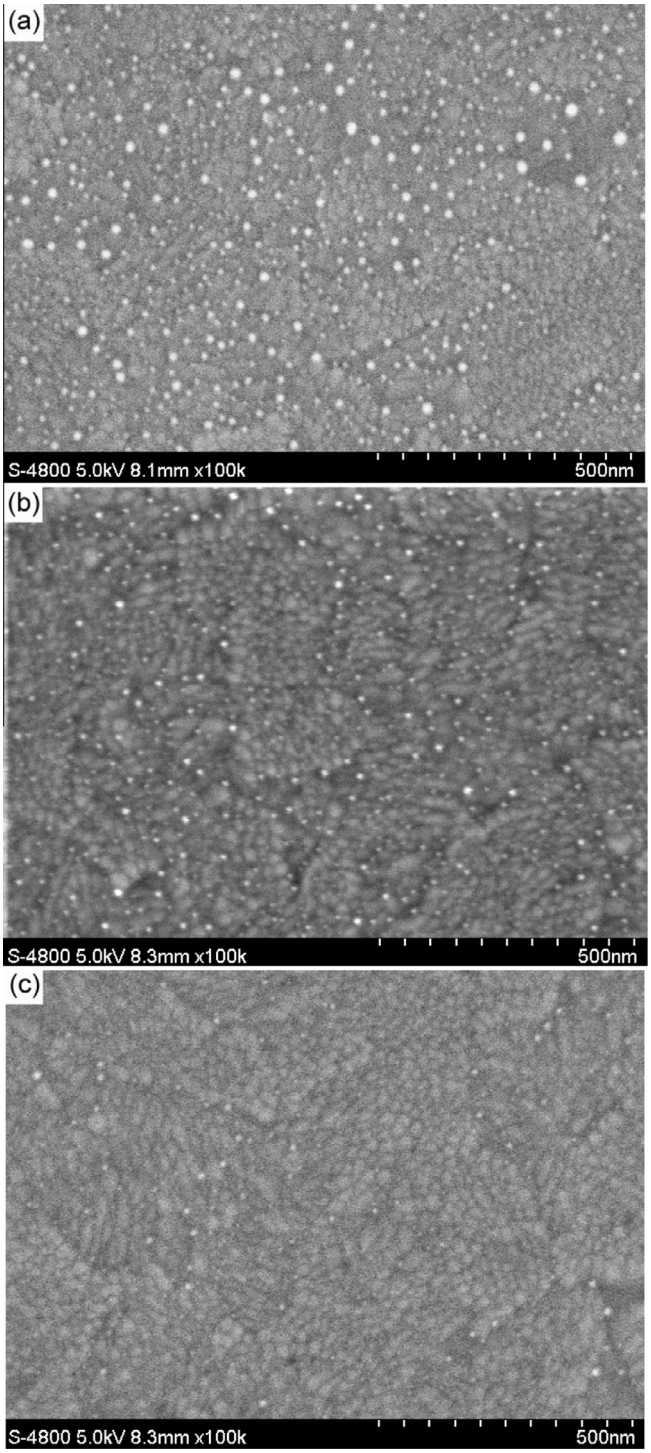

Fig. 1. SEM images measured for GNP-covered ITO/glass surfaces having different GNP concentrations, made by GNP solutions with different OD values of (a) 26, (b) 20 and (c) 13.