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Remarkable increase in the efficiency of N,N′-dimethylquinacridone dye heavily doped organic light emitting diodes under high current density

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We demonstrated a remarkable rise of external quantum efficiency (EQE) of N,N′-dimethylquinacridone (DMQA) heavily doped organic light-emitting diodes with tris-(8-hydroxyquinolinato) aluminum as host at large current density. The EQE of 5.0 wt % DMQA doped device increased 42% as the current density rises from 20 to 300 mA/cm^2. At 300 mA/cm^2, the EQE of 1.8 wt % DMQA doped device equals to that of the optimal concentration (0.8 wt %) doped device, which behaves a flat trend curve with the increase of current density. Whereas, another green fluorescent dye (Coumarin6) doped device with the same structure indicates straight falling dependence of EQE on increasing current density. The EQE increase is attributed to the dissociation of excimer species formed by interaction of higher concentration DMQA molecules into DMQA monomer excitons under high current density. © *2010 American Institute of Physics*. [doi:10.1063/1.3332478]

Since the thin film organic light-emitting diode (OLED) and fluorescent dye doped OLEDs were reported,[1,2] OLEDs have been gradually matured and industrially in panel displays and solid state lightings, such as portable electronic devices and OLED televisions. While more researchers have devoted to further study on the electroluminescence (EL) mechanism because there are still some barriers for display applications such as the fall of luminescent efficiency and the degradation of passive matrix displays at larger current. Various high yield fluorescent dyes were doped into host materials with optimized transport and luminescent properties, which can lead to EL of desirable hues with very high efficiencies. Another advantage of the doped emitter system in OLED is the enhancement of its operational stability by transferring electrogenerated excitons to the high efficient and stable dopant site.[3] Altogether, the technique based on dye doped emitter system would be significant for architecture design of high performance OLEDs.

In order to achieve high performance OLED, the high external quantum efficiency (EQE) at high drive current density would be very crucial. However, the EQE of electrofluorescent OLEDs generally decreases with the rising of drive current density, caused by the quenching of the singlet excited state of the dopant,[4] which is similar to the behavior of triplet-triplet annihilation in electrophosphorescence devices.[5] Besides, high current in OLEDs sometimes lead to the high local Joule heat that generally reduces EL efficiency.[6] Murata *et al.*[7] reported a device with temperature-independent EQE which selected quinacridone derivation (N,N′-diethylquinacridone) as emitter dopant. In addition, Shi and Tang have reported that N,N′-dimethylquinacridone (DMQA) doped tris-(8-hydroxyquinolinato) aluminum (AlQ) hosted OLED (DMQA doped device, hereafter) showed higher device stability.[3] So we fabricated the DMQA, especially high concentration, doped devices to investigate the influence factors on EQE of OLEDs at larger drive current density.

In this letter, we reported considerably increases in EQEs of 1.8–5.0 wt % DMQA doped OLED devices in high current density range. The findings are unlike with that of DMQA doped device with an optimal concentration of 0.8 wt % and OLEDs doped with other fluorescent dye, such as, Coumarin6 (C6). It is surprising that EQEs of 1.8–5.0 wt % DMQA doped devices markedly rise with the drive current density. Especially, the EQE of 5.0 wt % device rises up to 42% as the current density rises from 20 to 300 mA/cm^2, and at 300 mA/cm^2, the EQE of 1.8 wt % DMQA doped device equals to that of the optimal concentration doped device which behaves a flat trend curve of EQE with increase of the current density.

The OLED devices with structure of ITO/NPB (40 nm) /AlQ: DMQA (*x* wt %, 30 nm)/AlQ (60 nm) /LiF (1 nm)/Al (150 nm) were fabricated, here NPB is N,N′-bis-(1-naphthyl)-N,N′-diphenyl-1,1′-biphenyl-4,4′-diamine and denotes the hole-transporter, and *x* is 0, 0.6, 0.8, 1.8, 2.5, and 5.0, respectively. All chemicals are commercially available. The deposition of organic film, device fabrication, and the test processes are mainly accorded to the previous reports of our group.[8,9] The photoluminescence (PL) decay curve and spectrum were measured by a two channel TEKTRONIX TDS-3052 oscilloscope and a Spectrometer with Spex 1403 photomultiplier with a boxcar averager under excitation of a 355 nm YAG:Nd^{3+} laser with a repetition frequency of 10 Hz and pulse duration of 10 ns. The response time of the measurement system is about 30 ns.

Figure 1(a) plots EQE as a function of current density and the inset shows the increase ratios of EQEs for 0.8–5.0

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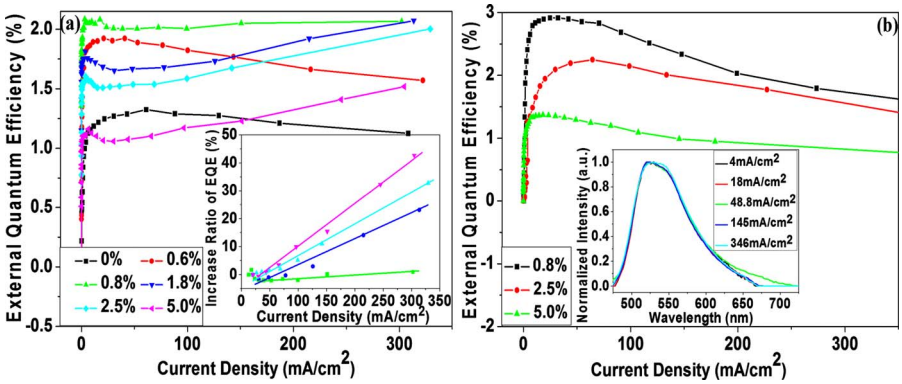

FIG. 1. (Color online) (a) The dependences of EQE on current density of DMQA doped devices with various concentrations. Inset: The EQE increase ratios for 0.8–5.0 wt % DMQA doped devices as the drive current density rises from 20 to 300 mA/cm^2. (b) The relations of EQE and current density of C6 doped devices with various concentrations. Inset: EL spectra of 5.0 wt % C6 doped device at different current densities. The structure and fabrication process of C6 doped devices is the same with DMQA doped OLED devices.

wt % DMQA doped devices at high current density comparing with that of 20 mA/cm^2. It can be seen that 0.8 wt % device represents the highest EQE of about 2.1% at 20 mA/cm^2 and behaves nearly flat response in a wide range of drive current density. However, it is interestingly found that for DMQA heavily doped devices with higher concentration (1.8–5.0 wt %), EQEs obviously increase with the rising of current density. The increase ratio is larger for 5.0 wt % doped device (42%) than for 1.8 wt % doped device (23%) as the drive current densities of devices rise from 20 to 300 mA/cm^2 [inset of Fig. 1(a)].

In order further to affirm the special dependence of EQE on current density for DMQA heavily doped devices, another green fluorescent dye, C6, doped devices with the same structure were also demonstrated. Figure 1(b) indicates the relations of EQE and current density of C6 doped devices with various doping concentrations. We note that EL results of C6 doped devices are far different from that of DMQA doped devices. Comparing with Fig. 1(a), it can be seen that only falling EQE dependences on current density for 0.8, 2.5, and 5.0 wt % devices are observed, which is similar to previous report of the efficiency dependence on drive current density at 1–100 mA/cm^2.[10] It is also noticed that the EL spectra have little change under different drive current densities [inset of Fig. 1(b)].

To explain why the EQEs increase under larger current density for higher DMQA doped devices, EL spectral figures of DMQA doped devices were compared, as described in Fig. 2. With rising of DMQA concentration, 575 nm shoulder peak increases and the spectral figure of the shoulder peak waveband is similar to that of the inset [Fig. 2(a)]. This indicates that there is additional emission band except for the DMQA monomer emission. The PL lifetime of the shoulder emission is about 369 ns while the lifetime and spectrum of the monomer emission band could not be detected, i.e., it should be smaller than 30 ns, which is a well known feature of fluorescence emission. Therefore, it could be expected that 540 and 575 nm emission bands should originated from DMQA monomer emitter and DMQA excimer emitter formed between QMQA molecules. It is well known that an excimer is a two molecule complex of the same compound, which is bound only in the excited state and rapidly dissociates into two separate molecules (monomers) after relaxation to the ground state.[11] Furthermore, from Figs. 2(b) and 2(c), we observe that the relative intensity of 575 nm emission of 5.0 wt % DMQA doped device decreases with the rise of current density but for 0.8 wt % DMQA device the 575 nm intensity keeps almost constant with current density.

Basing on above findings and arguments the increase in EQE of heavily DMQA doped devices with current density

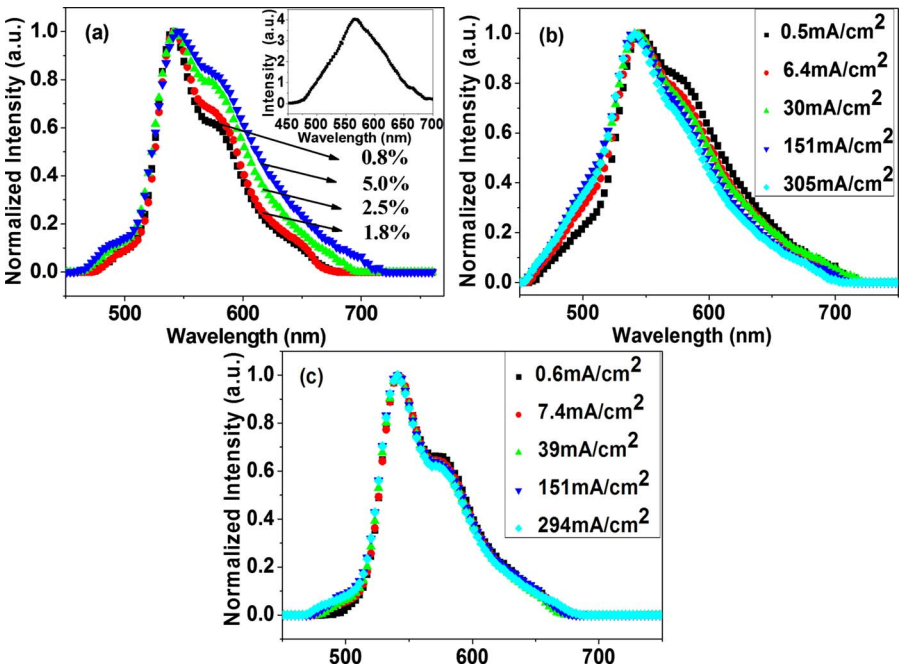

FIG. 2. (Color online) (a) EL spectra of DMQA doped devices at 20 mA/cm^2. Inset: The PL spectrum of 565 nm shoulder peak of 5.0 wt % DMQA doped AlQ film (100 nm). (b) EL spectra of 5.0 wt % DMQA doped device at different drive current densities. (c) EL spectra of 0.8 wt % DMQA doped device at different drive current densities.

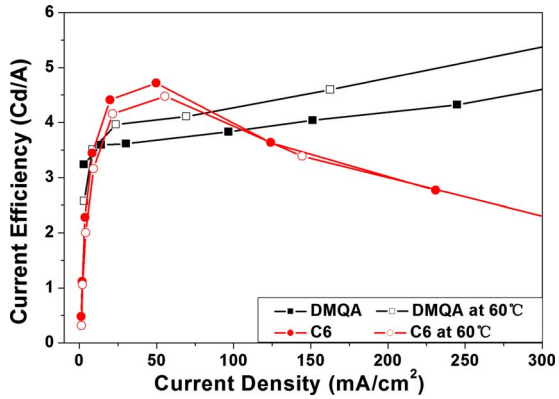

FIG. 3. (Color online) Relations of EQE and current density of 5.0 wt % DMQA and C6 doped devices before and after heating.

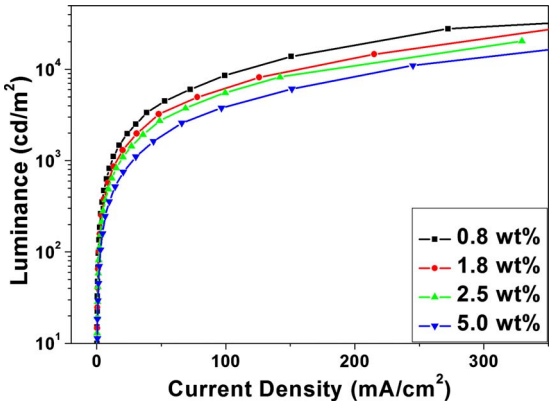

FIG. 4. (Color online) Plots of luminance as a function of current density of DMQA doped devices.

can be understood as follows. Note that drive of the device under larger current density implies that the device works under higher electrical field. The excimer emission appears to be easily quenched by the higher field, which is similar to exciplex emission due to their identical excited state feature.[12] That is, the decay dynamics of the excimer exciton is field-dependent but the monomer exciton is not.[13] For heavily doped devices, excimer and monomer excitons can be simultaneously formed, and the lower EQE at small current density [Fig. 1(a)] is due to the competition between excimer emission with lower efficiency and monomer emission. As the current density increases, the excimer is gradually quenched by the high electrical field and dissociates into DMQA monomer molecules which is again contribute to the monomer emission. Considering the efficiency of excimer emission is lower than monomer emission, the whole EQE of heavily DMQA doped device increases under large current density due to contribution of two emissions which resulted from the original DMQA monomer and DMQA monomer decomposed from excimer.

Figure 3 indicates the relations between EQE and current density of 5.0 wt % DMQA and C6 doped devices before and after heating. It can be seen that EQE of 5.0 wt % DMQA doped device at 60 °C not only increases with rising of current density but also is higher than that at room temperature in the whole current intensity range but the EQE of 5.0 wt % C6 doped device decreases at 60 °C. Thus it could be concluded that the higher resistibility of Joule heat resulted from local larger current density could be another reason for the increase in efficiencies of DMQA doped devices with current density. The smaller effect of Joule heat at larger current density on EQE of DMQA doped devices is perhaps attributed to weaker singlet-heat annihilation reported by Yamamoto *et al.*[14]

In addition, it is very valuable that 1.8–5.0 wt % DMQA doped devices behave acceptable luminance besides their high EQEs, as shown in Fig. 4. At 300 mA/cm^2, the luminance of 5.0 and 1.8 wt % DMQA doped devices are 13 000 and 22 000 cd/m^2, respectively.

We demonstrated remarkable increase in EQE for DMQA heavily doped devices with rising of current density. The 5.0 wt % DMQA doped device behaves the maximum rising amplitude up to 42% from 20 to 300 mA/cm^2. EQEs of 1.8 and 0.8 wt % DMQA doped devices are almost identical at 300 mA/cm^2. Whereas, EQE of the optimal DMQA doped device (0.8 wt %) exhibits a flat trend with increase of current density. The increase phenomenon of EQE may be attributed to the dissociation of DMQA excimer species into monomer excitons at high current density for heavily doped devices.

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