Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Locking Excitons in Two-Dimensional Emitting Layers for Efficient Monochrome and White Organic Light-Emitting Diodes

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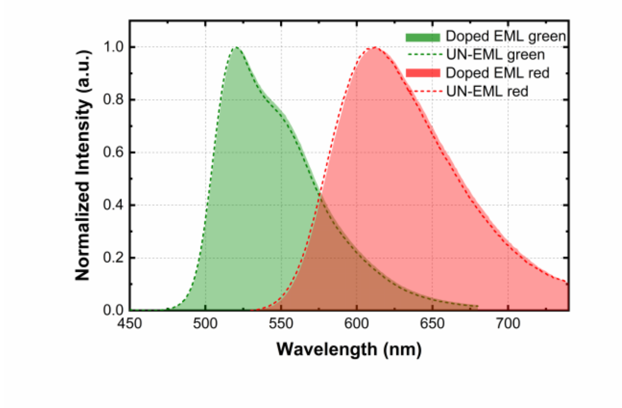

Fig. S1 PL spectra of UN-EMLs and doped EMLs. The device structure is quartz/50 nm mCP:B3PYMPM:Ir(ppy)$_2$(acac) (1:1, 8 wt%) for doped green EML, quartz/50 nm mCP:B3PYMPM:Ir(MDQ)$_2$(acac) (1:1, 10 wt%) for doped red EML, quartz/3 nm mCP:B3PYMPM (1:1)/[0.1 nm Ir(ppy)$_2$(acac)/5 nm mCP:B3PYMPM (1:1)]$_5$/0.1 nm Ir(ppy)$_2$(acac)/3 nm mCP:B3PYMPM (1:1) for green UN-EML and quartz/3 nm mCP:B3PYMPM (1:1)/[0.1 nm Ir(MDQ)$_2$(acac)/5 nm mCP:B3PYMPM (1:1)]$_5$/0.1 nm Ir(MDQ)$_2$(acac)/3 nm mCP:B3PYMPM (1:1) for red UN-EML, respectively.

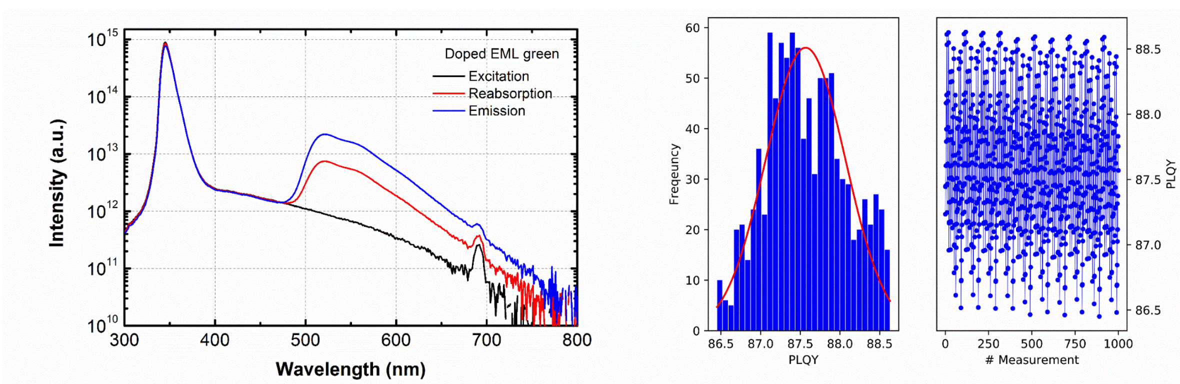

Fig. S2 PLQY of green doped EML with a structure of quartz/50 nm mCP:B3PYMPM:Ir(ppy)$_2$(acac) (1:1, 8 wt%).

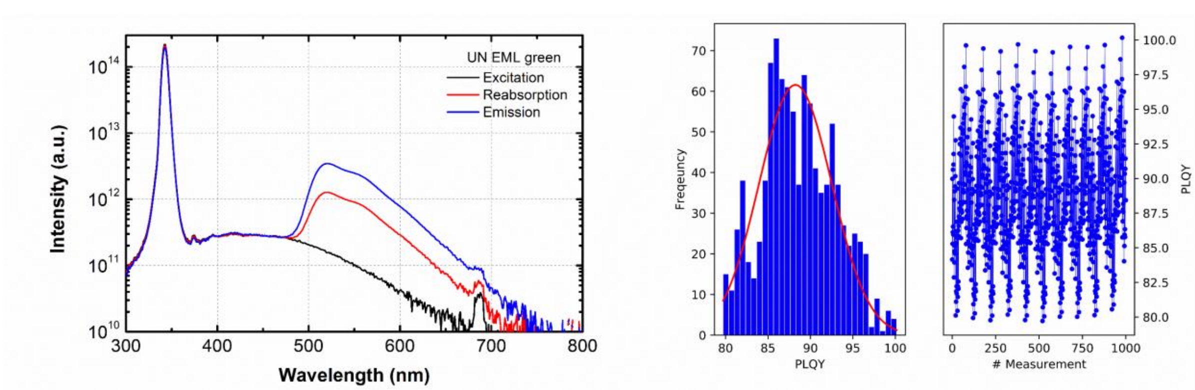

Fig. S3 PLQY of green UN-EML with a structure of quartz/3 nm mCP:B3PYMPM (1:1)/[0.1 nm Ir(ppy)$_2$(acac)/5 nm mCP:B3PYMPM (1:1)]$_5$/0.1 nm Ir(ppy)$_2$(acac)/3 nm mCP:B3PYMPM (1:1).

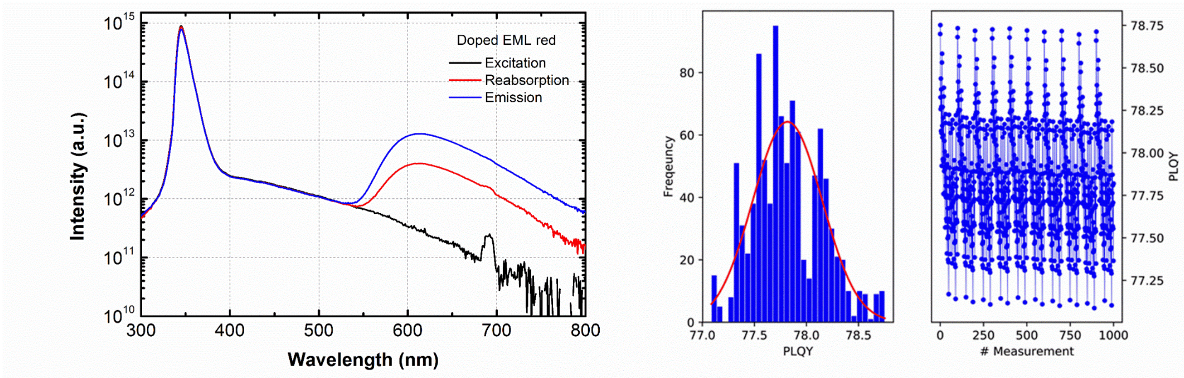

Fig. S4 PLQY of red doped EML with a structure of quartz/50 nm mCP:B3PYMPM:Ir(MDQ)$_2$(acac) (1:1, 10 wt%).

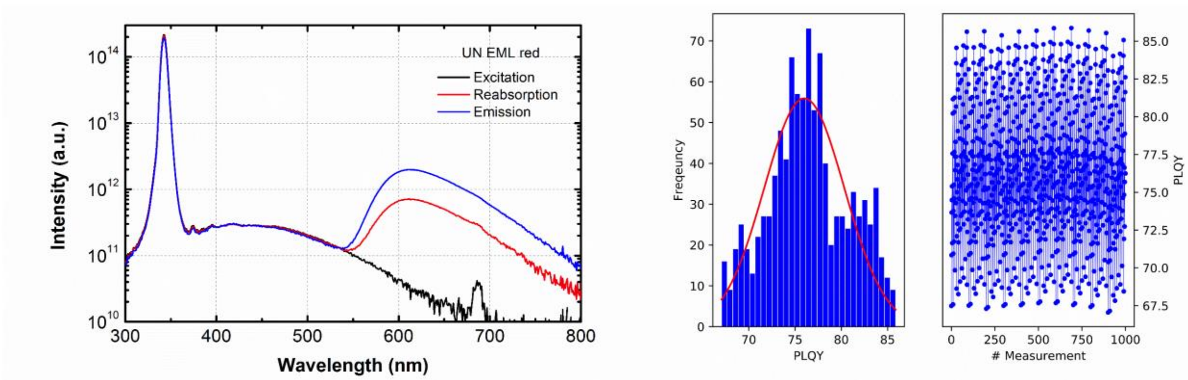

Fig. S5 PLQY of green UN-EML with a structure of quartz/3 nm mCP:B3PYMPM (1:1)/[0.1 nm Ir(MDQ)$_2$(acac)/5 nm mCP:B3PYMPM (1:1)]$_5$/0.1 nm Ir(MDQ)$_2$(acac)/3 nm mCP:B3PYMPM (1:1).

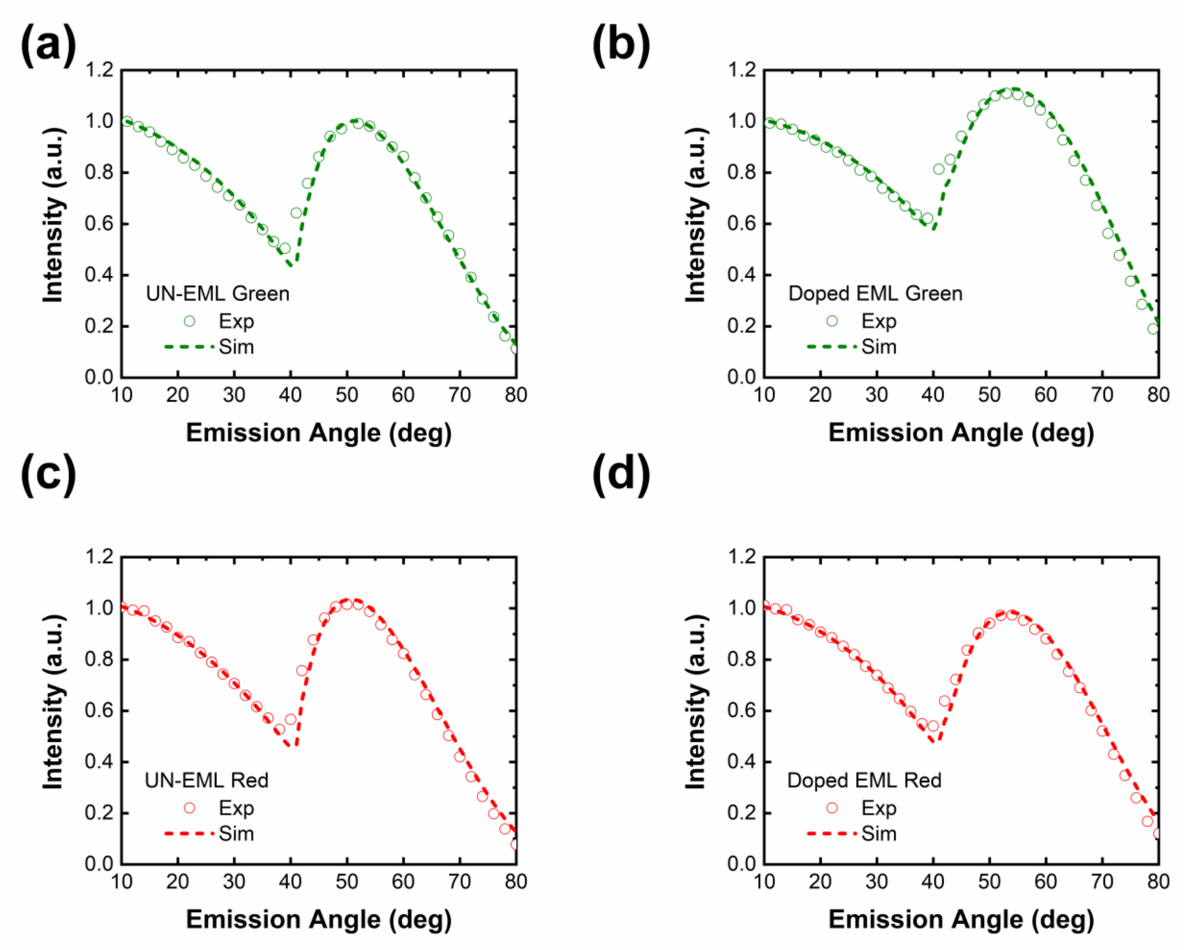

Fig. S6 Measured (dots) and fitted (dashed lines) angular emission spectra of (a) green UN-EML, (b) green doped EML, (c) red UN-EML and (d) red doped EMLs. The fits are performed at the emission peak wavelengths which are 520 nm for the green and 610 nm for the red emitter.

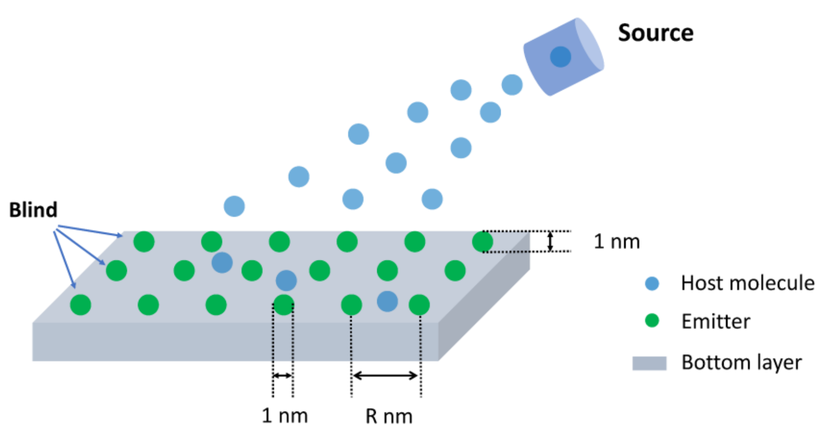

Fig. S7 Deposition process of UN-EML. Roughly, the 0.1 nm sub monolayer reaches 10 mol% by treating all molecules as nanoballs with 1 nm diameter and assuming only 1 nm is needed to form a continuous film.

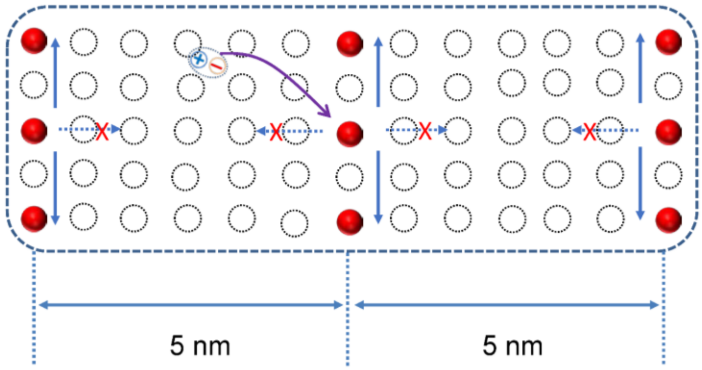

Fig. S8 Exciton diffusion and transfer routes in the UN-EML. The red ball and the black circle represent the emitter and CT-host, respectively. The purple arrow indicates the exciton transfer from charge-transfer hosts to emitters. After the excitons captured by the emitters, exciton diffusion between emitters occurs, which is a prerequisite for TTA via Dexter transfer. Due to the ultrathin body of the emitting layer and high triplet energy of both donor and acceptor materials of the CT-host system, the exciton diffusion in the direction perpendicular to the substrate is suppressed (blue dash arrows). Therefore, the exciton diffusion is locked in two-dimensional planes (blue arrows).

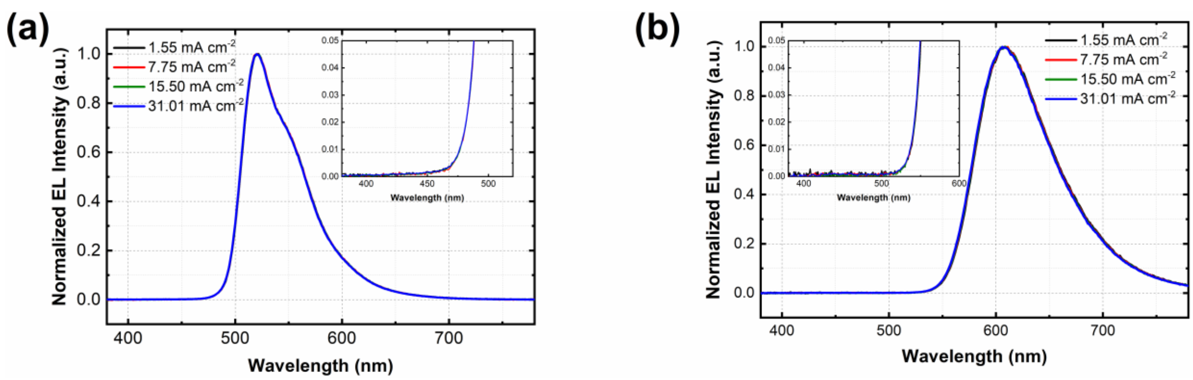

Fig. S9 Spectra of (a) green and (b) red OLEDs based on UN-EML under different current densities. Insets are the magnified plots.

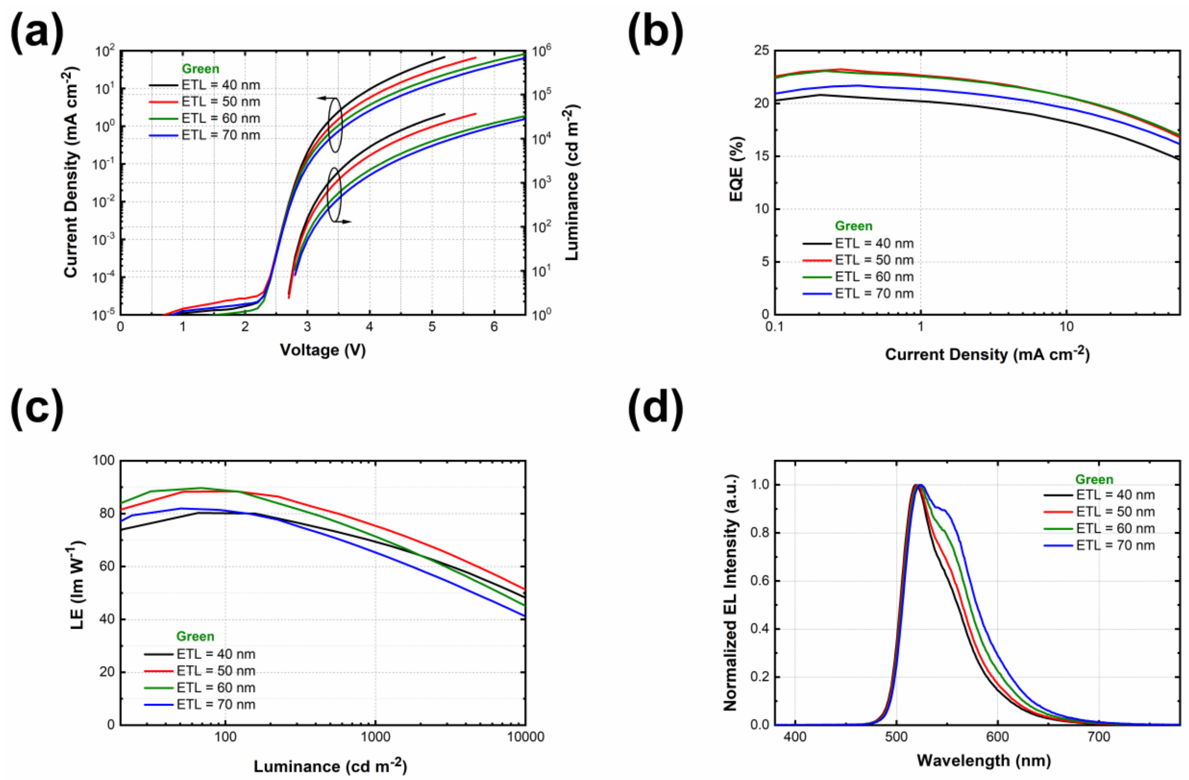

Fig. S10 (a) Current density-voltage-luminance curves, (b) EQE along with current density, (d) Luminous efficacy curves as function with luminance and (d) Normalized electroluminescent spectra at 15.4 mA cm^{-2} of monochrome green OLEDs with different thickness of the electron transport layer.

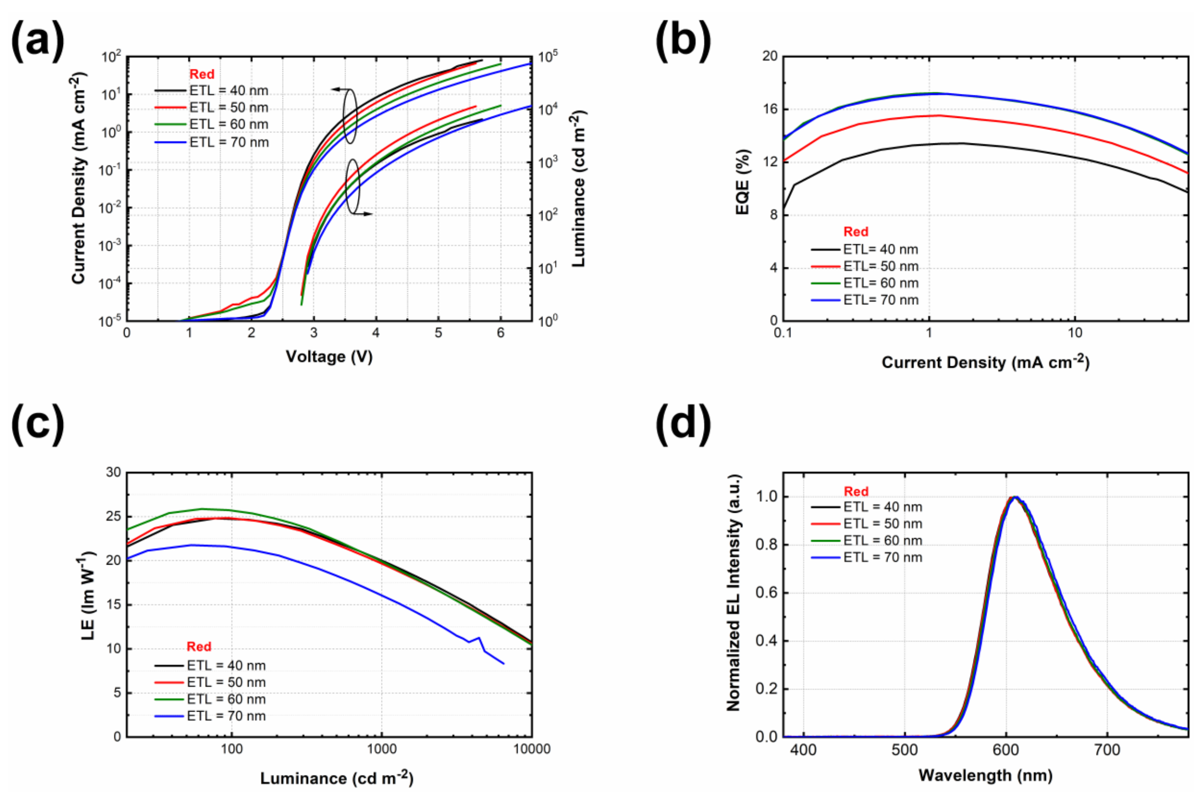

Fig. S11 (a) Current density-voltage-luminance curves, (b) EQE along with current density, (d) Luminous efficacy curves as function with luminance and (d) Normalized electroluminescent spectra at 15.4 mA cm^{-2} of monochrome red OLEDs with different thickness of the electron transport layer.

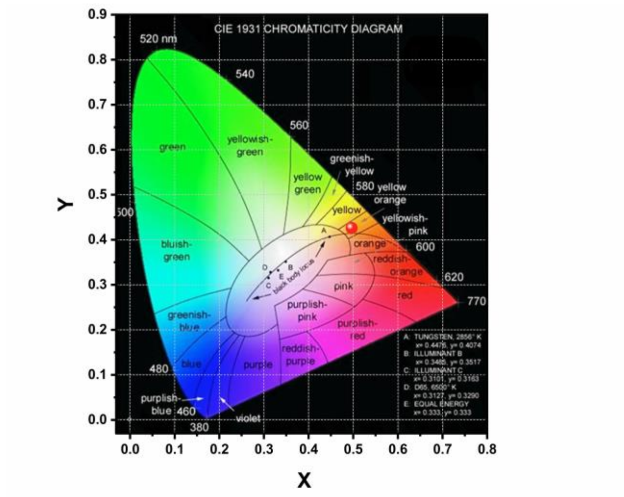

Fig. S12 CIE of white OLED W2.

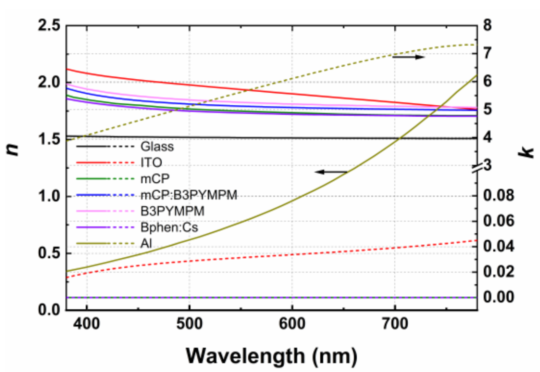

Fig. S13 Refractive index of the materials used in the simulation. The solid lines are the real parts of refractive index *n* and the dashed lines are the imaginary parts refractive index i.e. extinction coefficient *k*.

Table S1 Summary of parameters determined by the time-resolved PL curves at low and high excitation densities.

	Exciton density (10^{18} cm^{-3})	**Exciton lifetime τ (μs)**	**k_{TT} (10^{-12} cm^3 s^{-1})**
Doped EML /$Ir(ppy)_2(acac)$	0.7 ± 0.3	1.53 ± 0.05	0.8 ± 0.3
	2.3 ± 0.8	1.53 ± 0.05	0.6 ± 0.2
Doped EML /$Ir(MDQ)_2(acac)$	0.8 ± 0.3	2.00 ± 0.05	0.8 ± 0.3
	2.8 ± 1.0	2.07 ± 0.05	0.5 ± 0.2

Extreme case 1	UN-EML /Ir(ppy)$_2$(acac)	0.7 ± 0.3	1.65 ± 0.05	0.9 ± 0.4
		2.2 ± 0.8	1.70 ± 0.05	0.7 ± 0.3
	UN-EML /Ir(MDQ)$_2$(acac)	0.8 ± 0.3	2.14 ± 0.05	0.8 ± 0.3
		2.7 ± 1.0	2.32 ± 0.05	0.6 ± 0.2
Extreme case 2	UN-EML /Ir(ppy)$_2$(acac)	3.6 ± 1.2	1.63 ± 0.05	0.2 ± 0.1
		12 ± 4	1.70 ± 0.05	0.1 ± 0.1
	UN-EML /Ir(MDQ)$_2$(acac)	4.2 ± 1.5	2.14 ± 0.05	0.2 ± 0.1
		14 ± 5	2.31 ± 0.05	0.1 ± 0.1