Supplementary Information

Snapshot angle-resolved spectroscopy and its application for study of highly efficient polariton OLEDs

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Wavelength calibration of CMOS image

Wavelength dimension (x-axis) of the CMOS image was calibrated using the Hg-Ne lamp and laser diodes as light source. Figure S1a shows the CMOS image of the Hg-Ne lamp placed vertically in front of the objective lens, exhibiting three straight lines at the x-axis pixel values corresponding to the characteristic wavelengths of 546 nm, 577 nm, and 579 nm. Figures S1b show the CMOS images of 473 nm, 532 nm, and 635 nm laser diodes as incident into the system at 0 degree. The calibration result determines the whole detected wavelength range of 449-641 nm, and the wavelength resolution estimated from the FWHM of the spectral line is 1.8 nm (Figure S1a).

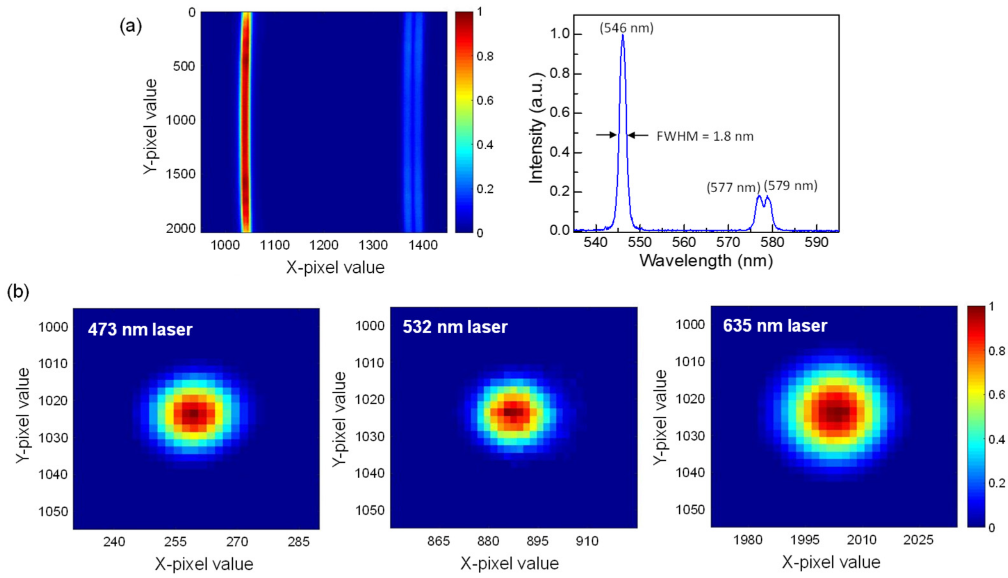

Figure S1. (a) CMOS image and extracted spectrum of the Hg-Ne lamp. (b) CMOS images of 473 nm, 532 nm, and 635 nm laser diodes.

EL and PL intensity calibration of CMOS image and goniometric angle-resolved spectroscopy system

In this work, EL and PL intensity calibration is based on correlating the CMOS image and the angle-resolved spectra measured from a typical goniometric system. For EL calibration, we used a blue-green OLED (ITO/m-MTDATA/NPB/DPVBi/Alq_3

/LiF/Ag) with broadband and high stability as the light source. Figure S2a shows the raw CMOS image of the blue-green OLED (5×5 mm^2) captured in 1 sec integration time. On the other hand, the angle-resolved spectra obtained by a goniometric system was employed as a calibration reference (see Figure 2d). We installed the goniometric system in a nitrogen glovebox to avoid device degradation induced by oxygen and moisture during the measurement, and scanned repeatedly to check the emission stability. In this setup, the OLED was placed on a platform with the ITO glass side facing up and precisely aligned at the rotation center. The OLED emission at a specific angle was collected by a collimating lens, which was mounted on a motorized rotation stage with a 7 cm radius of gyration and coupled to an optical fiber connected with a spectrometer (HR4000, Ocean Optics). The collimating lens was set to rotate from -60° to 60° relative to the device normal at the speed of 1°/sec (i.e., about 2 minutes in total), while the EL spectrum was recorded for every one degree. Since the effective area of emission collected by the collimating lens would change with the rotation angle, the OLED was prepared in a sufficiently small size (1×1 mm^2) for this setup to ensure collection of the parallel light from the entire device area at different angles. Therefore, this condition can be regarded as an approximation of point source measurement. For comparison, the CMOS image and angle-resolved spectra of the blue-green OLED were converted into the same matrix form of 121(angle)×2048(wavelength), where the maximum intensity was normalized to unity. By dividing the angle-resolved spectra in Figure 2d with the raw CMOS image in Figure S2a, we obtained the EL intensity correction matrix (EL-ICM), as shown in Figure S2b.

Similarly, the PL intensity was calibrated based on the angle-resolved fluorescence spectra of a reference sample (PFO:1 wt% F8BT blend film) measured by the goniometric system. However, unlike the EL which is homogeneously excited over the entire device area, PL is locally excited by the focused UV beam in the objective lens bore. To compare the two systems under the same excitation, the goniometric setup was installed in front of the snapshot system, and the rotation center was aligned at the objective lens bore. Figure S3a,b show the raw CMOS image of the PFO:F8BT blend film and its angle-resolved spectra measured with the goniometric system. By dividing the two, we obtained the PL intensity correction matrix (PL-ICM), as shown in Figure S3c.

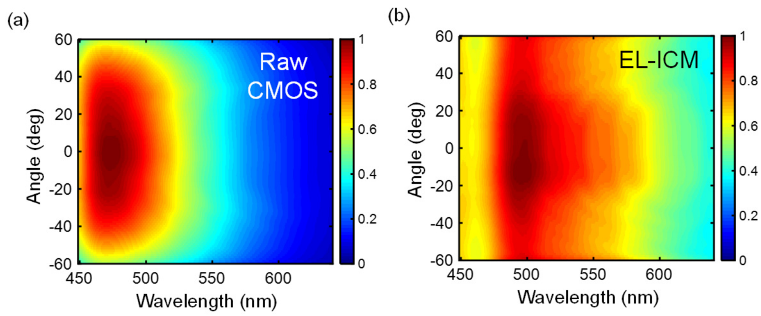

Figure S2. (a) Raw CMOS image of the blue-green OLED, and (b) EL intensity correction matrix (EL-ICM) extracted by dividing the angle-resolved spectra in Figure 2d with the CMOS image in (a).

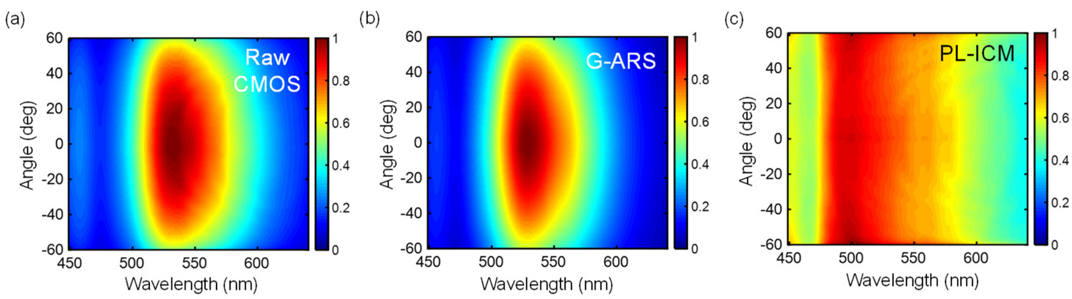

Figure S3. (a) Raw CMOS image of the PFO:F8BT blend film and (b) corresponding angle-resolved spectra measured with the goniometric system. (c) PL intensity correction matrix (PL-ICM) extracted by dividing the angle-resolved spectra in (b) with the CMOS image in (a).

Angle-resolved spectra of NPB/Alq3 OLED

We measured and simulated the angle-resolved spectra of NPB/Alq_3 OLED, which was configured as glass/ITO (150 nm)/MoO_3 (10 nm)/NPB (50 nm)/Alq_3 (50 nm)/LiF (1 nm)/Al (100 nm). The calibrated CMOS image well agrees with the angle-resolved spectra measured by the goniometric system (see Figure 3), and is also consistent to the simulation with the finite-difference time-domain (FDTD) method, as the result shown in Figure S4. In the FDTD simulation, we positioned a dipole point source near the NPB/Alq_3 interface, where electrons and holes were expected to recombine. There is a slight intensity difference between the measured and simulated spectra at high angles, possibly because the interference effect between multiple dipoles and the edge emission due to the guided mode were not considered in the simulation.

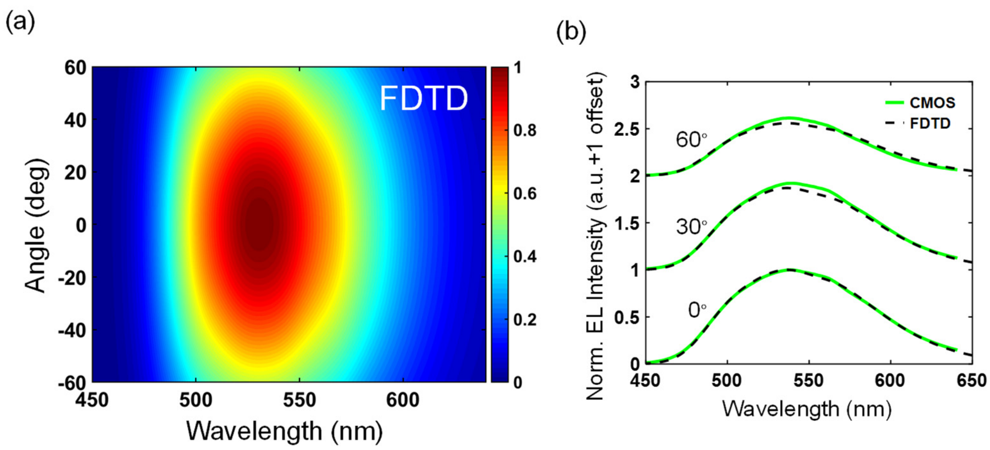

Figure S4. (a) FDTD simulation of angle-resolved spectra of NPB/Alq_3 OLED. (b) Comparison of the spectra extracted from the calibrated CMOS image and FDTD simulation.

Materials of polariton OLED with intracavity pumping architecture

In this work, we fabricated the polariton OLED with an intracavity pumping architecture, which mainly contains a cyanine dye DEDOC [5-chloro-2-(2-[(5-chloro-3-(3-sulfopropyl)-2(3H)-benzoxazolylidene)methyl]-1-butenyl)-3-(3-sulfopropyl)-

benzoxazolium inner salt, sodium salt] J-aggregate film as the strongly-coupled medium and a TXO-TPA [2-(4-(diphenylamino)phenyl)-10,10-dioxide-9H-thioxanthen-9-one] based thermally activated delayed fluorescence (TADF) OLED as the pumping emitter in a λ-thick cavity. The TADF OLED is configured as Au (15 nm)/MoO_3 (10 nm)/TAPC (40 nm)/mCP:2 wt% TXO-TPA (40 nm)/TmPyPb (40 nm)/LiF (1 nm)/Ag (150 nm). Figure S5 shows the absorption spectrum of DEDOC J-aggregate film and the EL spectrum of TADF OLED. The peak wavelength of TADF OLED nearly matches the low energy state of lower polariton mode, resulting in effective emission.

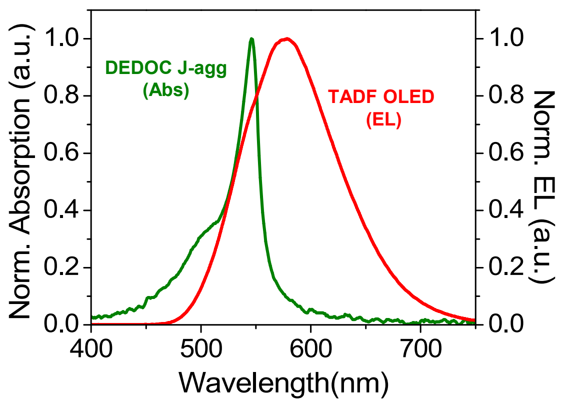

Figure S5. Absorption spectrum of DEDOC J-aggregate film and EL spectrum of TXO-TPA based TADF OLED.