# Manipulation of Pt···Pt Interaction in Platinum Complex by Methyl Group to Achieve Single-Doped White OLEDs: An Approach to Simulation of Daylight from Dawn until Dusk

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ABSTRACT: White organic light-emitting diodes (WOLEDs) possess excellent electro-optic conversion efficiency for state-of-the-art light sources and play an important role in energy consumption and carbon emission reduction. Because of the multiple cells or multiple emitters, the structure of normal WOLEDs is sophisticated, resulting in high fabrication costs and low long-term stability. To overcome these hurdles, WOLEDs based on platinum (Pt) complexes with a single light-emitting layer demonstrate promising potential. Herein, we report the regulation of Pt···Pt interaction of Pt(II) complexes by methylation of the ligand tuning the emission of monomer and excimer, resulting in realizing correlated color temperature (CCT) adjustable WOLEDs. Single-doped WOLEDs based on Im2PhPtCl with CIE coordinates of (0.32, 0.36), color rendering index (CRI) over 83, high external quantum efficiency (EQE) of 24.4%, and low-efficiency roll-offs (EQE =

23.9% at 1000 cd m^{-2}, EQE = 20.7% at 5000 cd m^{-2}) are accomplished by using exciplex as a host. More importantly, the CCT of the WOLEDs could be tuned between 2751 and 7061 K by simply changing the driving voltages and closely matching with daylight locus from dawn until dusk. The outstanding device performances illuminate the state-of-the-art for WOLEDs, and their further applications may let people enjoy sunbathing indoors.
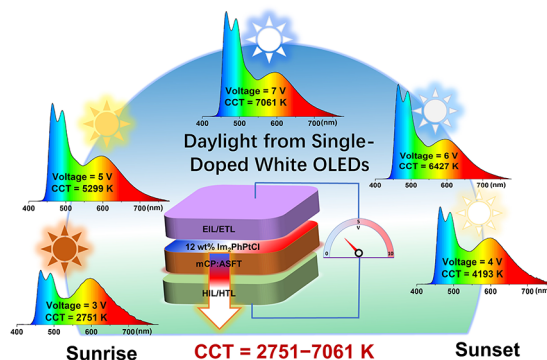
With the development and progress of science and technology, more and more artificial lighting sources, such as electricity/battery-driven incandescent bulbs and fluorescent tubes, have been invented and applied to illuminate the darkness.[1−3] However, compared with the natural source of light, sunlight, it is still a challenge for current lighting technologies to easily access high energy conversion efficiency, high color rendering index (CRI), and especially tunable correlated color temperature (CCT). From sunrise to sunset via noon, the CCT varies between 2500 and 7500 K in a northern country, and the CCT directly impacts human physiology and psychology.[4] High CCT light promotes the production of cortisol, a hormone that keeps people awake and active, while inhibiting the production of melatonin secretion, which can disorder the body's biological clock and may induce diseases. As a result, lighting sources with high CCT are suitable to be utilized for working time, and low CCT lights are preferred for night illumination.[5,6] Therefore,

developing a lighting source with easily controllable CCT features is valuable and necessary.

White organic light-emitting diodes (WOLEDs), as one of the most promising lighting technologies, possess the potential to act as an ideal lighting source with tunable CCT.[7−10] To realize this property, a straightforward strategy is to combine multiple sub-OLED cells in parallel or tandem and independently control them, as reported by Leo and co-workers,[11] Chou and co-workers,[12] and Kim and co-workers.[13] Because of the multiple cells, the structure of these devices is sophisticated, resulting in high fabrication costs and low long-
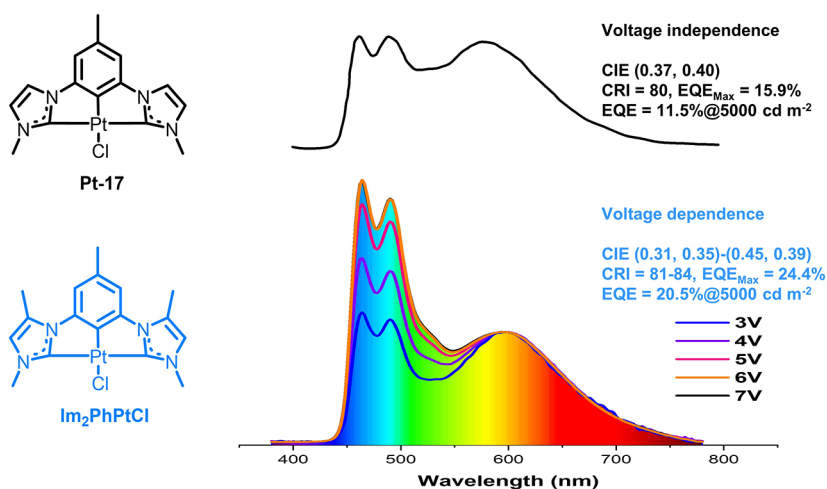
Figure 1. Molecular structures and emission spectra of Pt-17 and Im$_2$PhPtCl with their device performances.
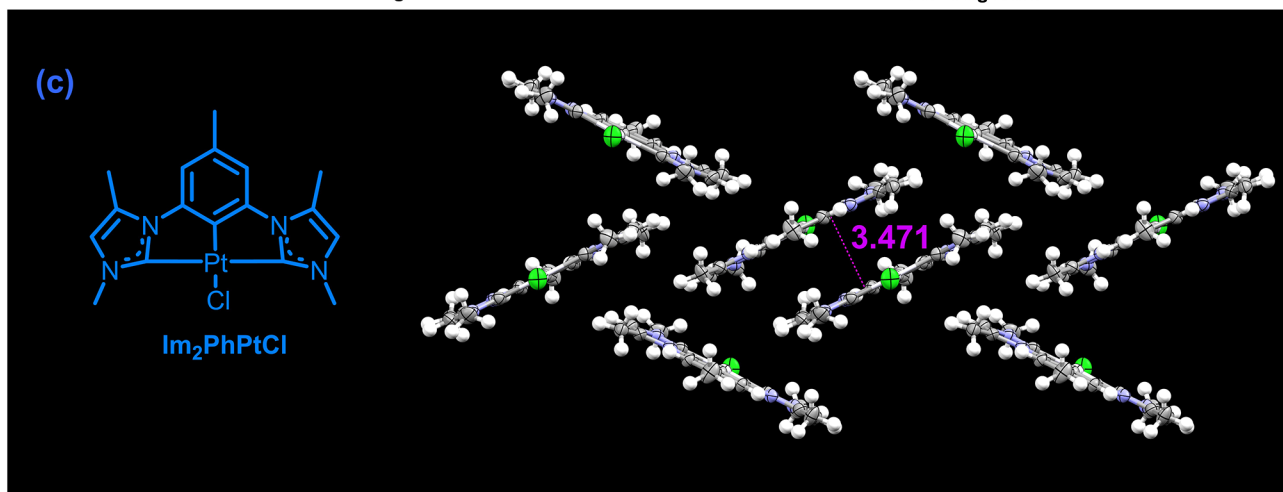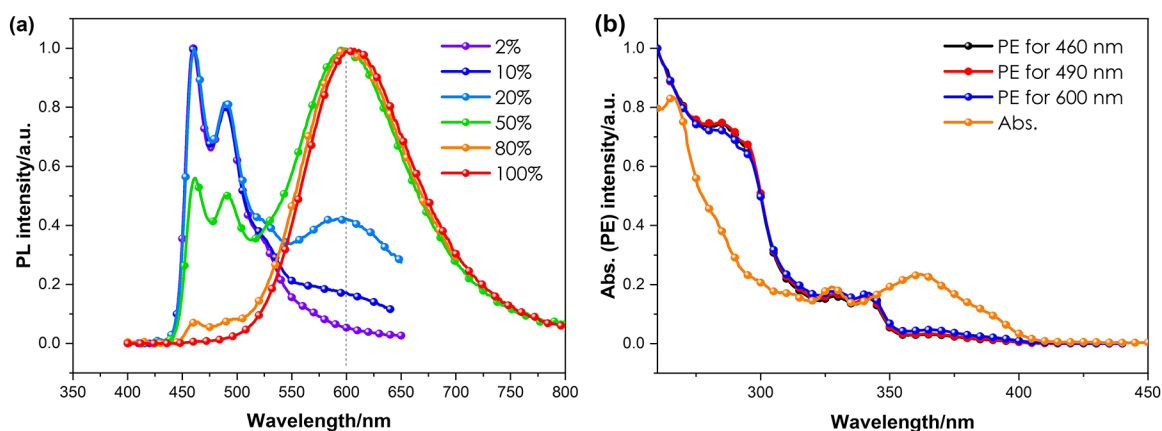

Figure 2. (a) PL spectra of Im$_2$PhPtCl in mCP with different doping concentrations. (b) Absorption spectrum, excitation spectra of emission at 460, 490, 600 nm of Im$_2$PhPtCl. (c) Chemical structure of Im$_2$PhPtCl and molecular packing in a single crystal.

term stability. A single-cell device structure with multiple emitters or layers, which can be selectively activated at different driving voltages, is a simpler choice for CCT-tunable OLEDs.[14−17] To further simplify the device structure, a single cell with a single emitter and emissive layer is highly desired for CCT-tunable WOLEDs.

Phosphorescent platinum (Pt) emitters have proven to be satisfactory candidates to realize monochromatic OLEDs, thanks to their excellent structural flexibility that allows the

ground state and excited state to be easily altered by employing various cyclometalating ligands.[18−22] Moreover, Pt(II) complexes can form a broad excimer emission due to their square-planar geometry that favors face-to-face interactions between molecules. The strong Pt···Pt interaction can destabilize the highest occupied molecular orbital (HOMO), resulting in the concentration-dependent formation of excimers whose emission is red-shifted. The combination of bluish monomer emission and reddish exciter emission are expected to obtain
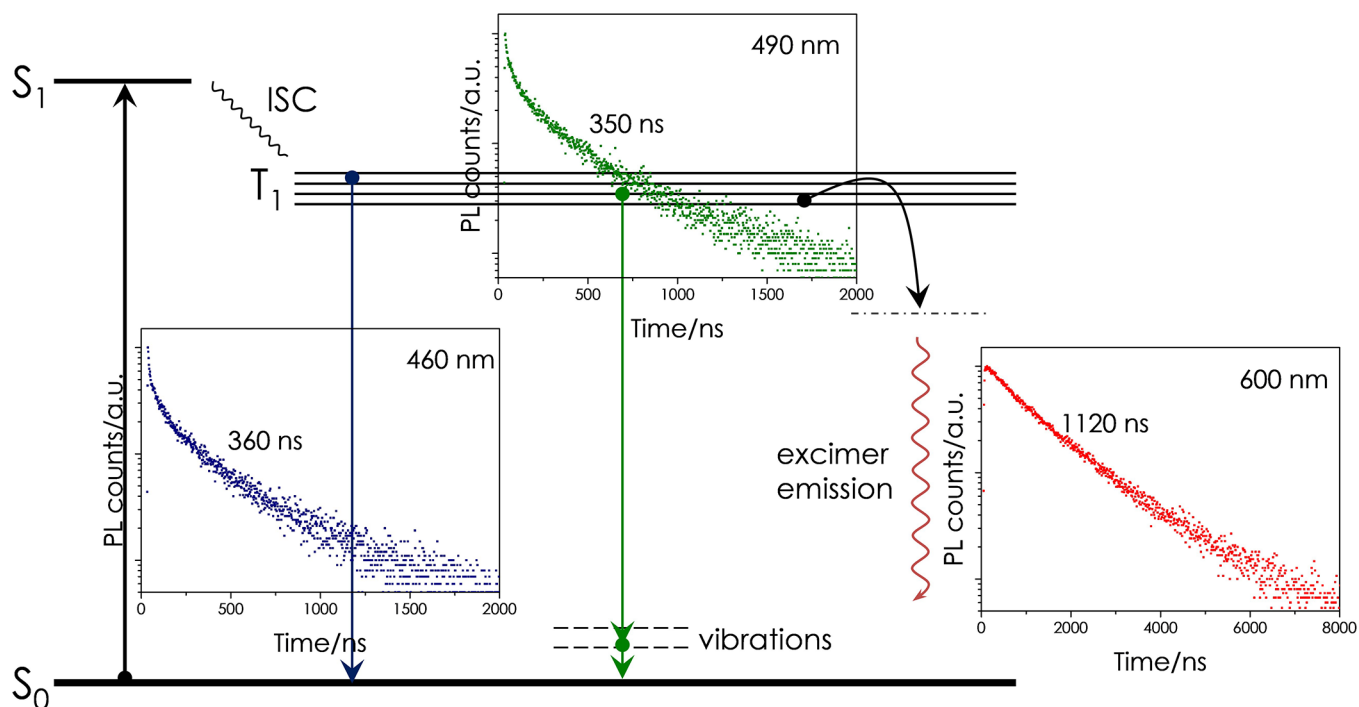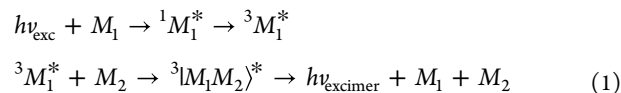
Figure 3. Transient decay and suggested energy level diagram of $Im_2PhPtCl$ doped in mCP.

white light from single-emitting species.[23−27] Li and co-workers reported a series of Pt-based WOLEDs and realized high performances with a maximum external quantum efficiency (EQE_{max}) of >20% and CRI of 80.[28,29] However, almost no CCT change was observed in these WOLEDs. Very recently, CCT-tunable WOLEDs based on two emissive layers with different Pt(II) emitter doping concentrations were reported by Che, Cheng, and co-workers, which achieve EQE_{max} values of ∼23.23% with a CIE coordinate being shifted from (0.56, 0.43) at 3 V to (0.42, 0.55) at 11 V.[30] These results show promising potential to access WOLEDs with tunable CCT based on Pt(II) emitters with better CRI.

Although the CCT could be tuned by using Pt(II) complexes as mentioned above, the parameters of CIE coordinates, CRI, CCT change range, and efficiency rolloff could not satisfy the requirement of WOLEDs for practical applications. To pursue a simpler device structure, higher efficiency, CRI, and wider CCT change range to act as a sunlight-like lighting source, herein, we employed Pt(II) [1,1′-(5-methyl-1,3-phenylene)bis(3,5-dimethyl-1H-imidazol-2-ylidene)] chloride (**$Im_2PhPtCl$**) as an emitter in a single-emissive-layer device.[31] Through utilizing the blue emission produced from **$Im_2PhPtCl$** monomer and red emission from its excimers, single-doped WOLEDs with CIE coordinates of (0.32, 0.36), CRI values of >83, high EQE values (24.4%), and low-efficiency rolloff (EQE = 23.9% at 1000 cd m^{-2}, EQE = 20.7% at 5000 cd m^{-2}) were accomplished. It is noteworthy that comparing with devices based on Pt-17, the extra methyl groups were introduced on imidazole for **$Im_2PhPtCl$**, which could drastically improve the performances of WOLEDs (Figure 1).[28] Simultaneously, the CCT of these WOLEDs could also be easily tuned between 2751 and 7061 K by changing driving voltages, and the CCT values closely matched with daylight locus from dawn until dusk.

Photoluminescent (PL) characteristics of **$Im_2PhPtCl$** was studied in doped N,N-dicarbazolyl-3,5-benzene (mCP) thin films with concentrations increasing from 2 to 100 wt % (neat **$Im_2PhPtCl$** thin film). As shown in Figure 2, the structured PL spectra with main emission peaks at 460 and 490 nm were observed at low concentration (2 wt %). The first high-energy emission (peak at 460 nm) corresponds to the transition from the lowest excited triplet state to the ground state ($T_1 \rightarrow S_0$), which is slightly red-shifted, compared with its analogs, Pt-16 and Pt-17.[28] The low-energy emission peak at 490 nm could be ascribed to the dominant vibrational stretch of molecules. When the concentration was increased by >10%, a new emission peak located at ∼600 nm was observed. As the concentration increases, the intensity of the red emission gradually increases. Its neat film (100 wt % **$Im_2PhPtCl$**) only show the emission peak at 600 nm, which could be ascribed to the excimer emission as previously reported.[18,23,28,29] To confirm its excimer emission of 600 nm at films, we measured the excitation spectra of the emission at 460, 490, and 600 nm, respectively. It was found that the excitation spectrum of the broadband (600 nm) is identical to that of the monomeric emission (Figure 2b), indicating that it originates from the excitation of isolated molecules (i.e., M_1 and M_2) that subsequently form the triplet emission excimer species ($^3|M_1M_2\rangle^*$):[32]

$$h\nu_{exc} + M_1 \rightarrow {}^1M_1^* \rightarrow {}^3M_1^*$$

$$^3M_1^* + M_2 \rightarrow {}^3|M_1M_2\rangle^* \rightarrow h\nu_{excimer} + M_1 + M_2 \quad (1)$$

The intermolecular interactions are weak at low concentrations, because the molecules are far apart, so the triplet excitons transit directly to the ground state and give out blue emission (λ = 460, 490 nm). As the concentration increased, more $Im_2PhPtCl$ molecules were involved in the film and the distance between them was shortened. The shortest intermolecular distance may be close to 3.471 Å, which is the distance between **$Im_2PhPtCl$**, as shown in the crystal packing (Figure 2c). The short distance could enhance
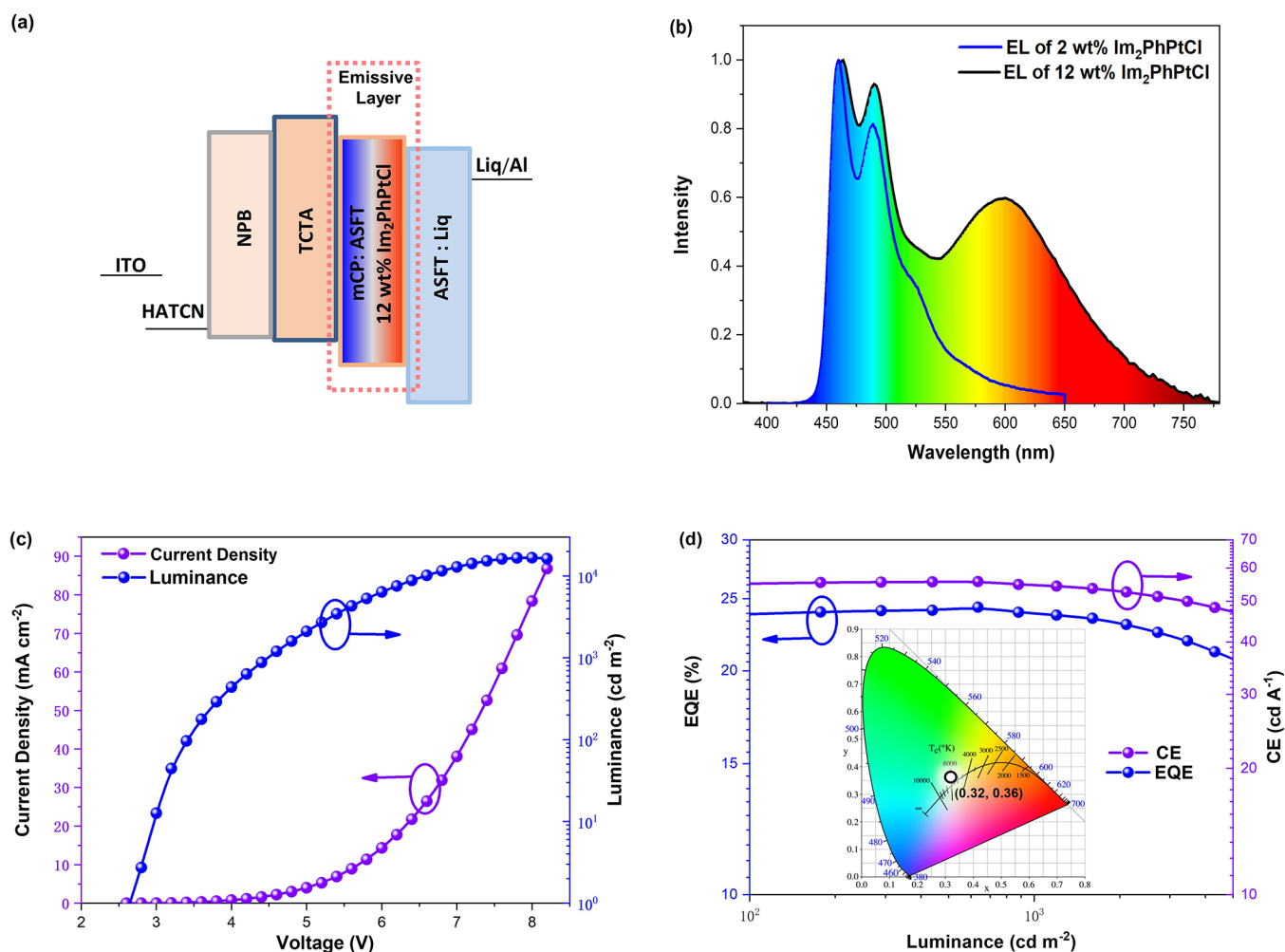
Figure 4. (a) Device architecture of single-doped WOLEDs. (b) EL spectra for blue and white devices at 4 V. (c) Current density−voltage− luminance characteristics (J−V−L) for the white devices. (d) EQE and CE versus luminescence curves for white devices.

intermolecular interactions. As a result, some of the monomer triplet excitons are transferred to the lower triplet state of the excimer, resulting in red emission. When the concentration of the emitter is increased, the proportion of red emission increases, with only red emission observed in neat **Im₂PhPtCl** film.

For excimers, the two molecules showed strong Pt···Pt interaction in the excited state. During relaxation back to the ground state, they dissociate and are thus reduced to monomeric molecules. Because of this, there is no ground state of excimers. Therefore, upon excitation, energy can be efficiently transferred from the monomer molecule to the bound emission center. The absorption spectrum of **Im₂PhPtCl** dissolved in dichloromethane solution was also performed to confirm this process (Figure 2b). The intense absorption below 300 nm can be assigned to $^1\pi-\pi^*$ transitions, largely centered at the cyclometalating ligands. The bands at a longer wavelength (300−400 nm) were assigned to metal-to-ligand charge-transfer (MLCT) transitions.[28]

Transient decay of the 20% **Im₂PhPtCl**-doped mCP thin film was measured, and the corresponding lifetimes of emission registered at λ = 460, 490, and 600 nm were 360, 350, and 1120 ns, respectively. Figure 3 shows the suggested energy level diagram and lifetimes in this process. Because of the lower

lowest unoccupied molecular orbital (LUMO) and the higher HOMO levels of **Im₂PhPtCl** than those of mCP host material, some of the **Im₂PhPtCl** triplet excitons may be formed through direct carrier trapping by **Im₂PhPtCl** dopants. The other **Im₂PhPtCl** triplet excitons should be formed by effective triplet energy transfer from the triplet states of the host materials to the **Im₂PhPtCl** triplet states. The higher triplet energy level of mCP may facilitate this transfer. The transition from the triplet state to the ground state of the monomer was performed at a high rate with a short lifetime (350 and 360 ns), but the energy transfer from the triplet states of **Im₂PhPtCl** to its excimer states may spend more time due to the interaction indicated in eq 1, resulting in a longer lifetime of the excimer emission process (1120 ns). When examined in degassed 2-methyltetrahydrofuran solution, a similar trend was demonstrated (Figure S1 in the Supporting Information), which remains consistent with the reported FPt.[23]

The emission color change of Pt(II) complexes with different doping concentrations inspired us to explore its electroluminescent performances. The emission peaks of **Im₂PhPtCl** are observed at 460 and 490 nm with a PLQY of 78% at low doping concentration (2 wt % in mCP), which can be used as a dopant for blue OLED. In addition, we measured it in mCP:ASFT (1:1) and found the PLQY could be increased to 87%. Indium tin oxide (ITO) and Liq/Al were

Table 1. Electroluminescent Device Performances of Blue/White OLEDs Based on Im₂PhPtCl and Some Representative Pt-Based WOLEDs

device	V_{on} [V]	CE_{max} [cd A⁻¹]	PE_{max} [lm W⁻¹]	EQE [%]	CIE (x, y)	CRI	CCT (K)
Im₂PhPtCl	2.9	56.9	50.3	25.9[a]	(0.16, 0.27)	–	–
Im₂PhPtCl	2.7	55.7	53.4	24.4[a]/23.9[b]/20.7[c]	(0.32, 0.36)[d]	83[b]	2751−7061
Pt-17[35]	4.0	–	37.8	15.9/14.6/11.5	(0.37, 0.40)	80	–
Pt-16[28]	3.5	–	51.0	20.1/16.0/−	(0.33, 0.33)	80	–
Pt-X-4[30]	3.0	43.7	29.2	23.2/23.2/21.0	(0.56, 0.43)[e]	–	–
Pt2O2[36]	–	59.0	46.1	24.6/21.0/−	(0.48, 0.48)	72	–
Pt7O7[29]	–	–	55.7	24.5/17.7/−	(0.37, 0.43)	70	–
FPt[37]	4.0	42.5	29.0	18.0/15.9/−[f]	(0.46, 0.47)	69	–

[a]At maximum. [b]At 1000 cd m⁻². [c]At 5000 cd m⁻². [d]At 635 cd m⁻² (EQE_max). [e]At 3 V (0.42, 0.55) at 11 V. [f]At 500 cd m⁻².
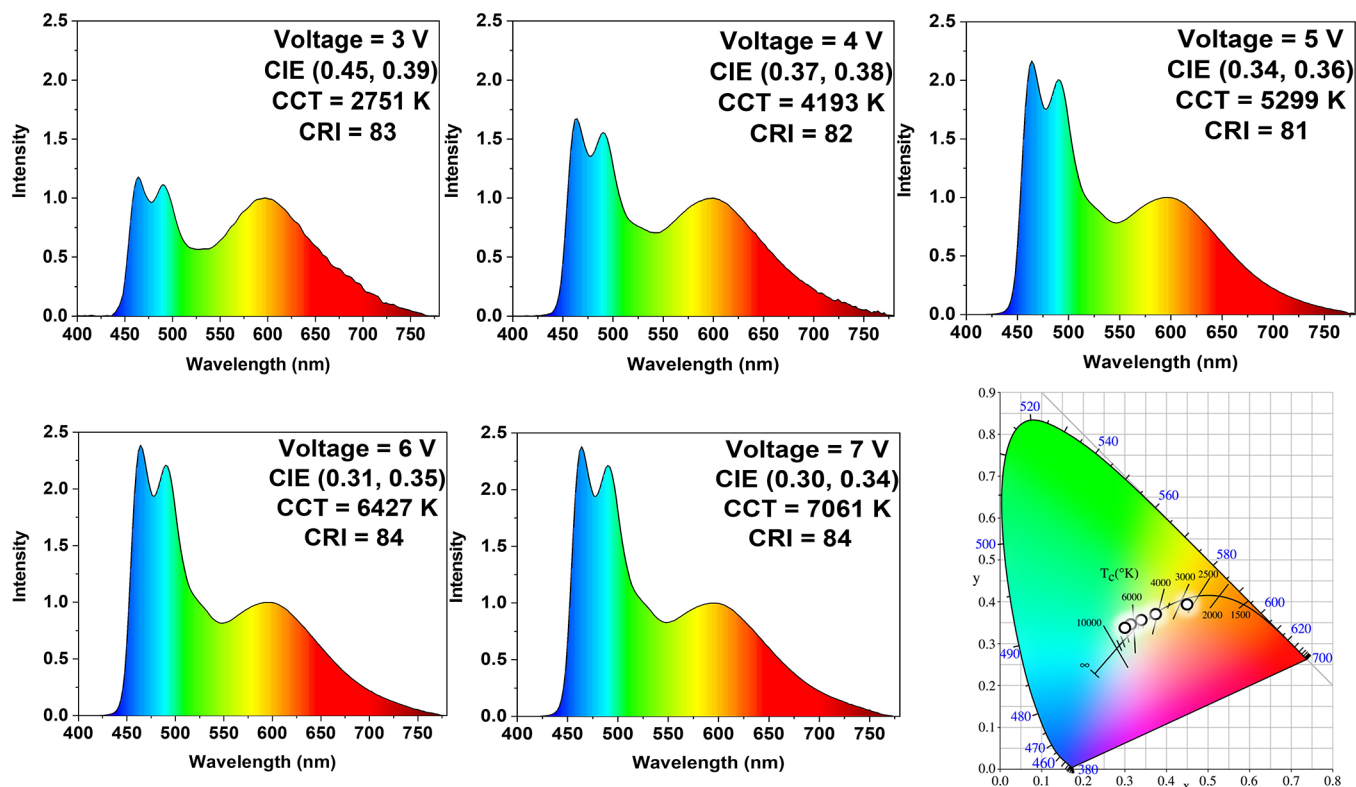
Figure 5. Emission spectra and CIE coordinates of WOLEDs based on Im₂PhPtCl at 3−7 V.

used as the anode and the cathode, respectively; *N,N'*-di(naphthalen-1-yl)-*N,N'*diphenyl-benzidine (NPB) and 4,4′,4″-tris(carbazol-9-yl)triphenylamine (TCTA) were used as the hole-transporting layers. The designed and optimized blue OLED device structure is as follows: ITO/HATCN (10 nm)/NPB (20 nm)/TCTA (10 nm)/2 wt % **Im₂PhPtCl** in mCP:ASFT (1:1, 10 nm)/ASFT:Liq (1:1) (20 nm)/Liq (1 nm)/Al (80 nm) (Figure S2 in the Supporting Information). The emissive layer (EML) was constructed through vacuum codepositing **Im₂PhPtCl** (2 wt %) with mCP:ASFT (1:1). The exciplexes constructed between mCP/ASFT were used as a host for **Im₂PhPtCl** dopant.[33,34] The EL spectra showed two peaks at ∼460 and 490 nm, consistent with its PL in 2 wt % mCP (Figure 4b). No other emission peaks were observed, suggesting a complete energy transfer from the exciplex host to **Im₂PhPtCl**. The blue OLED with CIE coordinates of (0.16, 0.27), maximum current efficiency (CE_max) of 56.9 cd A⁻¹, power efficiency (PE_max) of 50.3 lm W⁻¹, and EQE_max of 25.9% were obtained (Figure S2 in the Supporting Information, and Table 1). To the best of our knowledge, the EQE_max of 25.9%

is one of the highest values in blue OLEDs based on Pt(II) complexes. Similar to the devices based on Pt-16 and Pt-17, the blue device using **Im₂PhPtCl** as an emitter also exhibited almost voltage independence in the emission color.

As mentioned above, the red emission component gradually increases with rising concentration. At the optimized concentration of 12 wt % in mCP, it exhibited PLQY 75% and emission peaks at 460, 490, and 600 nm, so it possesses the potential to be used as an emitter for single-doped WOLED. In the mCP:ASFT cohost, the PLQY was 85%. Compared with Pt-17 (PLQY of 48% with excimer), the higher PLQY of **Im₂PhPtCl** with excimer can be ascribed to the steric hindrance of the methyl group in the 1-position of imidazole unit, which could hinder the concentration quenching of excimers, resulting in high PLQY. The EL devices with 12 wt % of **Im₂PhPtCl** as emitters were constructed to pursue high-performance WOLED. The device structure was similar to the previous blue device, but the concentration of **Im₂PhPtCl** was increased to 12 wt % (Figure 4a). As expected, the device exhibited white emission with CIE coordinate of (0.32, 0.36)
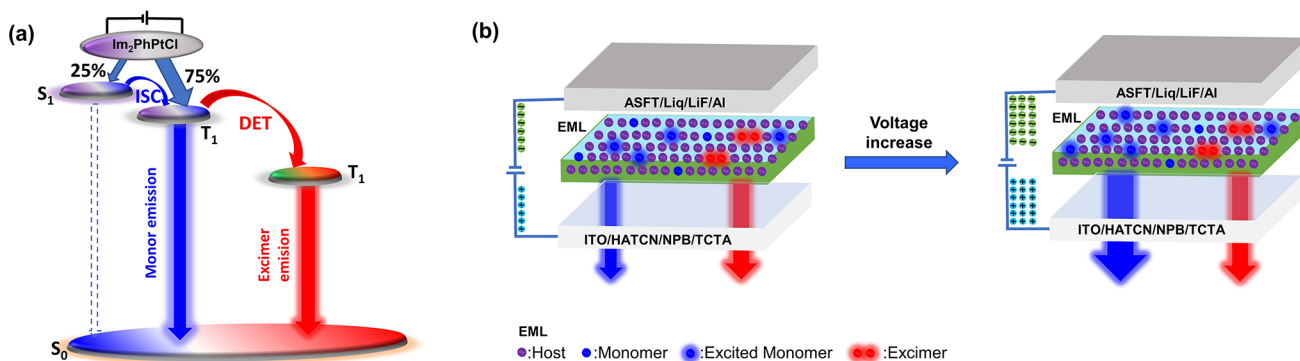
Figure 6. (a) Energy transfer of **Im₂PhPtCl**. (b) Monomer and excimer emission in EML at low and high driving voltages.

at 635 cd m^{-2}, CE$_{max}$ of 55.7 cd A^{-1}, PE$_{max}$ of 53.4 lm W^{-1}, and EQE$_{max}$ of 24.4% (see Figure 4 and Table 1). The CIE coordinates are very close to the standard requirement of NTSC for WOLED at CIE (0.33, 0.33). The CRI of the WOLED was 83, which is the best among that of FPt (75), Pt-16 (80), and Pt-17 (80).[23,28] The excellent CIE coordinates and CRI value demonstrate the high quality of white color based on **Im₂PhPtCl**. For practical luminance of 1000 cd m^{-2}, the EQE still maintained at 23.9%. Even at 5000 cd m^{-2}, the efficiency rolloff could also be inhibited effectively, leading to an EQE of 20.7% at this high luminance, which is the best performance so far for single-emitter and single-emissive-layer WOLEDs (see Table 1, as well as Table S1 in the Supporting Information). These results fully show the promising potential of **Im₂PhPtCl** for high-quality WOLEDs. Additionally, considering the smaller efficiency roll-offs of the WOLED based on **Im₂PhPtCl** (EQE$_{max}$ = 24.4%; EQE = 23.9% at 1000 cd m^{-2}, rolloff = 2.0%, and EQE = 20.7% at 5000 cd m^{-2}, rolloff = 15.2%) than that of devices reported by Li (EQE$_{max}$ = 24.5%; EQE = 17.7% at 1000 cd m^{-2}, rolloff = 27.7%), which exhibited a device lifetime to 50% of the initial luminance (LT$_{50}$) of 10 000 h, we believe that our WOLED device should also have a good performance in device operational stability. The deep exploration of this part is ongoing in our group.

Moreover, we found that the EL spectra of WOLED based on **Im₂PhPtCl** changed as the driving voltage increased. As shown in Figure 5, the relative EL intensity of blue emission increased as the driving voltage increased from 3 to 7 V. With the voltage at 3 V, the device showed CIE coordinates of (0.45, 0.39) with a CRI of 83 and CCT of 2751 K. As the driving voltage increased, the percentage of blue emission increased. When the driving voltage was 7 V, the device exhibited CIE coordinates of (0.30, 0.34) with CRI of 84 and CCT up to 7061 K. Compared to the WOLEDs reported by Duan and co-workers[16] and Che and co-workers,[30] using multiemitter or multilayer emissive devices, our device realized CCT manipulation with the very simple device structure of a single-doped emissive layer (Figure 5a and Figure S4 in the Supporting Information). More importantly, the CCT could be tuned between 2751 and 7061 K by changing applied voltages, and compared with two very representative voltage-dependent WOLEDs; the results in our work are closer to the blackbody radiance curve with the percentage of ΔCIEy being below 10% (between −4.88% and +9.38%; see Figure S5 and Table S2 in the Supporting Information). This fully shows the potential of WOLEDs based on **Im₂PhPtCl** to mimic natural daylight covering the whole daytime variation from dawn until

dusk (CCT = 2500−7500 K), which is beneficial to future lighting applications.

For the CCT change phenomenon in **Im₂PhPtCl**-based WOLEDs, we suppose that it could be ascribed to the effective adjument of exciton concentrations in EL devices. As shown in Figure 6a, under low driving voltages, the current density is low and the exciton concentration is not very high. Because of the influence of the metal atoms, the generated singlet excitons of monomers are converted to triplet excitons by an intersystem crossing process. A part of the triplet exciton radiatively transits to the ground state, resulting in a single molecule of blue light. Simultaneously, because of its higher energy level, most of the triplet excitons of the monomer will be transferred to the lower triplet excitons of excimer, and they will eventually transit to the ground state with red emission. As the current density increases, the number of excitons in the device increases. The excitons that are transformed to the excimer for red emission by energy transfer will saturate due to a fixed concentration of excimer (Figure 6b). As a result, more triplet excitons of monomer become available, which transit directly to the ground state by radiative process, resulting in an increased proportion of blue emission. The emission color was changed from warm white to cold white. For **Im₂PhPtCl**, since the introduction of the methyl group changes the intermolecular interactions, it is possible to control the saturation of red emission from excimer, thereby realizing the change of luminescence. However, for Pt-16 and Pt-17, due to the small steric hindrance, resulting in strong intermolecular interactions, it is difficult to achieve controllable red emission saturation, which makes it unable to adjust the emission properties. In addition, under high voltage/high luminance, the efficiency rolloffs of devices based on Pt-16 and Pt-17 are more serious (18.4% for Pt-16, 5.7% for Pt-17; see Figure S6 and Table S3 in the Supporting Information) than **Im₂PhPtCl** (2.0%),[28,35] which shows that the excitons in these devices are more prone to be quenched. The newly generated extra excitons cannot be used to increase blue emission, resulting in an unchanged spectrum.

In summary, the advantages of Pt(II) complexes in blue and white OLEDs have gradually shifted from scientific research to industry. The simple introduction of a methyl group in Pt(II) complexes to regulate the interaction of Pt···Pt interactions provides a new approach for molecular design to achieve tunable single-doped WOLEDs. We report the first single-doped WOLED to realize the simulation of daylight from dawn until dusk. By combining the device engineering, CCT-tunable WOLEDs were successfully obtained with CIE coordinates of (0.32, 0.36), CRI of >83, a high EQE$_{max}$ of 24.4% with

extremely small efficiency rolloffs at 5000 cd m^{-2} (EQE = 23.9% at 1000 cd m^{-2}, rolloff = 2.0%, and EQE = 20.7% at 5000 cd m^{-2}, rolloff = 15.2%). It is noteworthy that the CCT of the white device could be tuned between 2751 K and 7061 K by simply changing the driving voltages, which closely matched the daylight locus on the CIE 1931 chromaticity diagram. This resulted in the ability of these devices to simulate the daylight from dawn until dusk in a northern country. Such promising device performance may allow us to develop sunlight-like lighting sources for indoor applications.

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Notes
The authors declare no competing financial interest.

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