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## Axially Chiral Bis-Cycloplatinated Binaphthalenes and Octahydro-Binaphthalenes for Efficient Circularly Polarized Phosphorescence in Solution-Processed Organic Light-Emitting Diodes

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extension of chiral axis planes. They can be used as emitters in solutionprocessed organic light-emitting diodes to achieve luminance efficiency, asymmetry factor, and external quantum efficiency up to 5.4 cd  $A^{-1}$ , 3.0 ×  $10^{-3}$ , and 3.1%, respectively. Moreover, the essential relationships between their chemical structures and luminescence quantum efficiency and asymmetry factor are discussed, which affords explicit insights for designing circularly polarized luminescent materials and devices.

### INTRODUCTION

Recently, circularly polarized organic light-emitting diodes (CP-OLEDs) have received growing attention due to their efficient ability to generate a circularly polarized luminescence (CPL) directly and extensive applications in optical spintronics, optical data storage, and three-dimensional (3D) displays.<sup>1-5</sup> Since Meijer et al. demonstrated the first CP-OLEDs in 1997,<sup>6</sup> various organic polymers and small molecules have been successively employed as emitters for the application in CP-OLEDs. On the one hand, except a few thermally activated delayed fluorescence organic emitters, most of them are fluorescent emitters, and consequently their OLEDs usually are inefficient with an intrinsic internal quantum efficiency (IQE) of 25%.8,9 On the other hand, because of the McClure heavy-atom spin-orbit coupling effect,<sup>10</sup> phosphorescent transition-metal complexes, such as Ir(III) and Pt(II) complexes, can harvest not only singlet but also triplet excitons to realize a theoretical 100% IQE for OLEDs.<sup>1-5</sup> Although considerable progress has been made, few high-performance CP-OLEDs with both a high electroluminescent (EL) efficiency and asymmetry factor  $(g_{\rm FL})$  have been reported. In general, there is a trade-off between improving EL efficiency and increasing  $g_{EL}$ . For example, Ln(III) complexes-based CP-OLEDs have a very high  $g_{EL}$  up to 1.41 but a relatively low external quantum efficiency (EQE) of  $4.2 \times 10^{-3}$  %.<sup>11</sup> On the contrary, Ir(III) complexes have an extra high EQE of 23.6% but a low  $g_{\rm EL}$  of 5  $\times$  10<sup>-4</sup>.<sup>12</sup>

Therefore, the development of new chiral emitters for highperformance CP-OLEDs is still urgent and significant.

In the past two decades, the class of square-planar cyclometalated Pt(II) complexes has been extensively investigated because of their interesting phosphorescence nature,<sup>13-19</sup> but there are only a finite number of chiral Pt(II) complexes for the applications in CP-OLEDs.<sup>20-27</sup> In 2016. Brandt et al. used helicene-based Pt(II) complexes [Figure 1a,  $(C^N)$ \*Pt(O^O), C^N = (P)/(M)-pyridinyl-helicene, O^O =  $\beta$ -diketones] to construct solution-processed CP-OLEDs, which have a high  $g_{EL}$  (+0.22 and -0.38) but a low power efficiency ( $\eta_{\rm P} = 0.23$  lm W<sup>-1</sup>).<sup>20</sup> After that Yan et al. introduced -F and -CF<sub>3</sub> moieties at a pyridinyl-helicene ligand to increase the volatility and emission intensity for vacuum-evaporated CP-OLEDs (EQE = 18.8%,  $g_{\rm EL}$  = 1.6 ×  $10^{-3}$ ).<sup>21,22</sup> Han et al.<sup>23</sup> [(N^C^N)Pt( $\equiv R$ )\*, N^C^N = 1,3bis(2-pyridyl,  $\equiv R = (R)/(S)-(+)-\alpha$ -methylbenzylisocyanide, solution-processed CP-OLEDs, luminance efficiency  $\eta_{\rm L} = 0.84$ cd  $A^{-1}$ ,  $g_{EL} \approx 10^{-4}$ ], Lee et al.<sup>24</sup> [(N^C^N)\*Pt(Cl), N^C^N =

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Figure 1. Selected reported Pt(II) complexes for CP-OLEDs (a-c) and binuclear Pt(II) complexes (d) in this work.

(R)/(S)-1-(2-oxazoline)-3-(2-pyridyl)phenylate, vacuumevaporated CP-OLEDs, EQE = 9.7%,  $g_{EL} = 1.2 \times 10^{-4}$ ], and Fu et al.<sup>25</sup> [Figure 1b, (C^N)Pt(N^O)\*, C^N = 1-(benzo[b]thiophen-2-yl)-isoquinoline, N^O = (R)/(S)-Schiff base ligands; solution-processed CP-OLEDs, EQE = 0.93%,  $g_{EL} \approx$  $10^{-3}$ ] adopted point-chirality-ligand-based Pt(II) complexes to fabricate CP-OLEDs. Qian et al. further utilized a liquid crystal to improve the EL performance of point-chirality-ligand-based Pt(II) complexes [(N^C)\*Pt(O^O), N^C = (R)/(S)-2phenylpyridine derivate, solution-processed CP-OLEDs, EQE = 11.3%,  $g_{EL} = 0.02$ ].<sup>26</sup> Recently, Jiang et al.<sup>27</sup> reported that axial-chirality-ligand-based Pt(II) complexes bearing aggregation-induced emission<sup>28-32</sup> are suitable for fabricating solution-processed CP-OLEDs [Figure 1c, (N^C)\*Pt(O^O), N^C = (R)/(S)-binaphthalene-derived N^C ligands, EQE = 2.15%,  $g_{EL} = 1.1 \times 10^{-3}$ ]. However, all the above Pt(II) complexes are mononuclear Pt(II) complexes. In comparison with mononuclear Pt(II) complexes, multinuclear Pt(II) complexes may have some unique luminescence properties,  $^{17,33-37}$  but they usually might suffer the problem of low solubility and aggregation-caused quenching (ACQ) through intramolecular and/or intermolecular interactions.

We are increasingly concerned about the preparation, optics, and sensing properties of axially chiral binaphthalenes (BINA).<sup>36-41</sup> Because of a restriction in the intramolecular rotations<sup>27,37</sup> or motions<sup>36</sup> of BINA, the reported BINA-based Pt(II) complexes usually have aggregation-induced emission (AIE)-active CPL. Moreover, since the dihedral angle of BINA is ~90°, BINA derivates are not planar molecules and have good solubility consequently. In the present work, a series of novel neutral and chiral BINA- or octahydro-BINA-bridging binuclear Pt(II) complexes  $[(O^{O})Pt(\mu - BINA - C^{N})*Pt (O^{O})$  or  $(O^{O})Pt(\mu - H_8 - BINA - C^{N}) * Pt(O^{O})$ , molecular weight up to 1342] (Figure 1d) were designed and prepared for efficient solution-processed CP-OLEDs. Because of the effect of a steric hindrance from bridging ligands and the 2,3position extension in chiral axis planes, the binuclear Pt(II) complexes emit an orange-red phosphorescence in both solution [emission quantum yield ( $\Phi$ ) up to 21% in degassed  $CH_2Cl_2$  and solid  $[\Phi$  up to 14% and 70% in powder and 4,4',4"-tris(carbazol-9yl)triphenylamine (TCTA) host, respectively]. Furthermore, the interrelationships between their chemical structures and  $\Phi/EQE/g$  are explored, which afford useful tips for the design of CPL materials and devices. As far as we are aware, this is the first example of binuclear Pt(II) complexes for the application in CP-OLEDs.

#### RESULTS AND DISCUSSION

**Synthesis and Characterization.** The synthetic methods of binuclear Pt(II) complexes are illuminated in Figure 2 and described in detail in the Supporting Information. The complexes have a good solubility in 1,4-dioxane, CH<sub>2</sub>Cl<sub>2</sub>,



Figure 2. Synthesis of binuclear Pt(II) complexes.

CHCl<sub>3</sub>, benzene, toluene, ethyl acetate (EtOAc), and tetrahydrofuran (THF) but a poor solubility in H<sub>2</sub>O, ether, hexane, ethanol, and CH<sub>3</sub>CN. Ten milliliters of toluene would dissolve 0.5 g of (R)-[tmd] (tmd = tetramethyl-l,2-dioxetane) and (R)-[H<sub>8</sub>-tmd] at least. The single crystals of (R)-[tmd] (CCDC No. 2086886) and (S)-[H8-acac] (CCDC No. 2086879) were obtained by a slow diffusion and evaporation of a CH<sub>3</sub>CN/toluene solution.

**Photophysical Properties.** The UV/visible absorption and phosphorescence data of all binuclear Pt(II) complexes at room temperature are listed in Table S1 and Figures 3–6 and



Figure 3. Absorption spectra of (*R*)-binuclear Pt(II) complexes in  $CH_2Cl_2$  (5.0 × 10<sup>-6</sup> mol dm<sup>-3</sup>).



**Figure 4.** Experimental and calculated absorption spectra (a) and energy-level diagram and frontier molecular orbitals (b) of (R)-[**tmd**] (in CH<sub>2</sub>Cl<sub>2</sub>).

(S1–S3 Supporting Information). Except for the circular dichroism (CD) and CPL properties, the optical properties of five pairs of (S)/(R) enantiomers are similar (Figures S1–S3), as in our previous reports.<sup>35–41</sup> As shown in Figure 3, the BINA-based Pt(II) complexes have similar absorption spectra with the same low-energy absorption band at  $\lambda_{abs} = 460$  nm, even though they bear different O^O auxiliary ligands of acetylacetone (acac), dibenzoylmethane (dbm), and tetra-



Figure 5. (a) Emission spectra of (*R*)-binuclear Pt(II) complexes in powder and TCTA (10%). Corresponding photos (b, c) in degassed CH<sub>2</sub>Cl<sub>2</sub> ( $5.0 \times 10^{-6}$  mol dm<sup>-3</sup>), (d) in PVK (5.0%), and (e) in powder under sunlight and 360 nm UV lamp.



**Figure 6.** Emission spectra  $(5.0 \times 10^{-6} \text{ mol dm}^{-3} \text{ in CH}_2\text{Cl}_2)$  of (R)-[**tmd**]. (inset) Calculated T<sub>1</sub> single spin density of (R)-[**tmd**].

methyl-l,2-dioxetane (tmd). The similar results were observed in the previous work on simple (C^N)Pt(O^O) complexes.<sup>42,43</sup> Since the H<sub>8</sub>–BINA-based Pt(II) complexes have a smaller  $\pi$ -conjugation system than BINA-based Pt(II) complexes, H<sub>8</sub>–BINA-based Pt(II) complexes exhibit blueshifted absorption spectra ( $\lambda_{abs} = 445$  nm).

To understand the mechanism of the excited states and transitions, density functional theory (DFT) and timedependent (TD) DFT were performed by the Gaussian 09 program package. In a dilute  $CH_2Cl_2$  solution, both absorption spectra bands and molar absorption coefficients ( $\varepsilon$ ) of (R)-[**tmd**] are predicted well by a theoretical calculation (Figure 4). The low-energy absorption band ( $\lambda_{abs} = 460$  and 453 nm for experiment and calculation, respectively) is contributed by the highest occupied molecular orbital (HOMO)  $\rightarrow$  lowest unoccupied molecular orbital (LUMO) (12.3%), HOMO  $\rightarrow$ LUMO+1 (39.2%), and HOMO-1  $\rightarrow$  LUMO (40.7%) transitions with an oscillator strength ( $f_{OSC}$ ) of 0.135. The energy-level diagram and frontier molecular orbitals of (R)-[tmd] (Figure 4) reveal that the electron clouds of HOMO-1, HOMO, LUMO, and LUMO+1 are symmetrically distributed over the two sides of the BINA. For the HOMO-1 and HOMO, the electron clouds are mainly located at electron-rich naphthalene rings and Pt(II) ions, but they are primarily contributed from not only naphthalene rings but also electrondeficient quinoline rings for LUMO and LUMO+1. Thereby, the low-energy absorption band can mainly be ascribed to both a singlet intraligand charge transfer (<sup>1</sup>ILCT) in a BINA bridging ligand and a singlet metal-to-ligand charge transfer (<sup>1</sup>MLCT) from Pt(II) ions to a BINA bridging ligand. Similar transitions are found for (R)-[dbm] (Figure S4) and (S)-[H<sub>8</sub>acac] (Figure S5) as well. Clearly, for different energy levels of (R)-[tmd], (R)-[dbm], and (S)-[H<sub>8</sub>-acac], the  $\pi$ -conjugated skeletons of O^O ligands have a small contribution to the electron clouds, but R groups (CH<sub>3</sub>, t-Bu, and Ph) have no contribution. This might be reason that BINA-based ( $\lambda_{abs}$  = 460 nm) and H<sub>8</sub>-BINA-based ( $\lambda_{abs}$  = 445 nm) Pt(II) complexes show alike low-energy absorption bands, even if they have different R groups.

As the reported BINA-based Pt(II) complexes usually have AIE-active photoluminescence (PL),<sup>27,36,37</sup> it is unexpected that all of them are highly luminescent in degassed CH<sub>2</sub>Cl<sub>2</sub> and powder (Table S1 and Figures 5, S2, and S3).<sup>41</sup> In a dilute solution, all the BINA-based binuclear Pt(II) complexes show similar emission spectra ( $\lambda_{em} = 620$  nm,  $\Phi = 7.5 - 8.2\%$ ). At the same time, all the emission spectra of H<sub>8</sub>-BINA-based binuclear Pt(II) complexes blue shift to  $\lambda_{em} = 610$  nm with higher  $\Phi$  (19–21%). Compared with solution samples, powder samples give a bit of a red shift in the emission band ( $\lambda_{em}$  = 635-643 and 625-630 for BINA- and H<sub>8</sub>-BINA-based binuclear Pt(II) complexes, respectively) along with a slight emission quenching ( $\Phi = 3.2-14\%$ ). If the binuclear Pt(II) complexes are doped in the host of polyvinylcarbazol (PVK) or TCTA, their  $\Phi$  values would sharply increase into 51% and 70%, respectively, through a host-guest energy transfer (Figure S6). Among all the binuclear Pt(II) complexes, tmdbased complexes have the highest  $\Phi$ , because of their biggest effect of t-Bu steric hindrance.

Because of the long emission decay lifetimes  $(0.50-7.9 \ \mu s)$ and the large Stokes shifts (~160 nm) between the absorption and emission band (Table S1 and Figures S7 and S8), the emission of binuclear Pt(II) complexes originates from triplet excited states (T). This can be further verified by the fact that the phosphorescence of a degassed solution can be efficiently quenched by oxygen (Figure 6). A single-electron spin density that is the density difference between  $\alpha$ - and  $\beta$ -electrons is useful to describe a triplet electron transition.<sup>44,45</sup> As depicted in Figures 6 and S9, the spin density distribution of the T<sub>1</sub> state for (*R*)-[**tmd**] and (*S*)-[**H**<sub>8</sub>-**acac**] is dominated by the upper side of the BINA bridging ligand and the upper Pt(II) ion, indicating that its phosphorescence originates mainly from <sup>3</sup>ILCT and <sup>3</sup>MLCT.

X-ray Single-Crystal Structures. The chemical structures and arrangements of organic molecules play a key in their emission characters. The X-ray single-crystal structures and packing of (R)-[tmd] and (S)-[H<sub>8</sub>-acac] are shown in Figure 7. There are some disorder problems in the single-crystal



Figure 7. X-ray single-crystal structures of (R)-[tmd] (a, c) and (S)-[H<sub>8</sub>-acac] (b).

structures, which are normal for alkyl groups.<sup>34,46,47</sup> No racemization is observed in the X-ray single-crystal structures of (R)-[tmd] and (S)-[H<sub>8</sub>-acac], revealing that the (R) and (S) axial chirality is stable enough for a chemical synthesis. Except the peripheral atoms of methyl groups and cyclohexyl for (R)-[tmd] and (S)-[H<sub>8</sub>-acac], respectively, all their other atoms are almost in two chiral axis planes, because the figuration of the 4-coordinated Pt(II) ion is square-planar. This is much different from the previous results<sup>27,36,37<sup>1</sup></sup> and would help to improve the CPL by extending axially chiral planes and achieve a strong emission in solution. These molecules have propeller-type structures with a dihedral angle within the BINA and  $H_8$ -BINA of 90.8 and 90.5° for (R)-[tmd] and (S)-[H<sub>8</sub>acac], respectively, and thus there are no intramolecular  $\pi - \pi$ stacking and Pt-Pt interactions. However, for (R)-[tmd], strong face-to-face intermolecular  $\pi - \pi$  stacking interactions (3.33 Å) are observed between the closest molecules, which might contribute to its low  $\Phi$  in a powder. The shortest intermolecular Pt–Pt distance is 8.01 Å,  $^{17,35}$  revealing that there is no intermolecular Pt-Pt interactions for (R)-[tmd]. On the contrary, (S)-[H<sub>8</sub>-acac] molecules have strong intermolecular Pt-Pt interactions (3.50 Å) but weak face-toface  $\pi - \pi$  stacking interactions. The comprehensive effect of two factors would result in (S)-[ $\mathbf{H}_8$ -acac] having a higher  $\Phi$ . Moreover, with the help of chirality and strong intermolecular interactions, (R)-[ $\mathbf{tmd}$ ] can self-assemble to form helix-like structures in a crystal.<sup>36,37</sup> No intramolecular  $\pi - \pi$  stacking and Pt-Pt interactions combine the fact that the  $\pi$ -conjugated system of binaphthalenes is broken by an axial chirality, which leads to the similar photophysical properties between binaphthalenes-based mononuclear Pt(II) complexes (Figure 1c) and binuclear Pt(II) complexes.

**Chiroptical Properties.** The chirality properties of all binuclear Pt(II) complexes in a dilute  $CH_2Cl_2$  solution (5.0 ×  $10^{-6}$  mol dm<sup>-3</sup>) are investigated by CD spectra (Figure S10). As an example, the CD spectra of (R)/(S)-[tmd] exactly mirrored each other (Figure 8), revealing that they are a pair of



Figure 8. Experimental and computational CD spectra of (R)/(S)-[tmd] and (R)/(S)-[H<sub>8</sub>-tmd] in CH<sub>2</sub>Cl<sub>2</sub> (5.0 × 10<sup>-6</sup> mol dm<sup>-3</sup>).

enantiomers. The computed CD spectrum of (*R*)-[**tmd**] in dilute CH<sub>2</sub>Cl<sub>2</sub> is almost identical to the experimental spectrum, revealing that the absolute (*R*) stereochemistry of (*R*)-[**tmd**] is reserved in a dilute solution. For (*R*)/(*S*)-[**H**<sub>8</sub>-**tmd**], similar results are observed. On the basis of previous work,<sup>36-41</sup> the high-energy (<350 nm) and lower-energy (>350 nm) CD bands can be mainly assigned to chiral binaphthyl itself and chiral binaphthyl-induced <sup>1</sup>MLCT/<sup>1</sup>ILCT, respectively. It is noteworthy that the CD signals at 465 nm of BINA-based Pt(II) complexes are much bigger than those of H<sub>8</sub>-BINA-based Pt(II) complexes, meaning that BINA-based Pt(II) complexes have much bigger absorption asymmetry factors [ $g_{abs} = 4.3 \times 10^{-3}$  and  $1.2 \times 10^{-3}$  for (*R*)-[**tmd**] and (*R*)-[**H**<sub>8</sub>-**tmd**], respectively, Table S1].

(*R*)-[**tmd**] and (*R*)-[**H**<sub>8</sub>-**tmd**] were doped in PVK to examine their CPL properties, because their cast films have high  $\Phi$  values up to 51% (Figure 9). The dissymmetric factors of  $g_{\rm PL}$  are determined up to  $-4.2 \times 10^{-3}$  and  $-1.9 \times 10^{-3}$  for (*R*)-[**tmd**] and (*R*)-[**H**<sub>8</sub>-**tmd**], respectively, which are in agreement with the CD absorption data.

**Electrochemical Properties.** Electrochemical spectra were measured by cyclic voltammetry (CV) in  $CH_2Cl_2$  solutions to examine the redox properties of (*R*)-[tmd] (Figure S11) and (*R*)-[H<sub>8</sub>-tmd] (Figure S12). The energy levels of the HOMO and LUMO in the corresponding complexes were calculated by the first oxidation potential and the onset of the absorption spectra. The band gap and energy levels of HOMO and LUMO are -2.44, -5.46, & -3.02 and -2.53, -5.50, & -2.97 eV for (*R*)-[tmd] and (*R*)-[H<sub>8</sub>-tmd], respectively. These data are a bit higher than the TD-DFT computational data (Figure 4), but their trends are coincident.



Figure 9. PL (top) and CPL spectra (middle) and  $g_{PL}$  (bottom) of dye-doped PVK films (5.0%) based on (a) (R)-[tmd] and (b) (R)-[ $H_8$ -tmd].

**CP-OLEDs Properties.** Combining high emission quantum yields and asymmetry factors, (R)/(S)-[tmd] and (R)/(S)-[H<sub>8</sub>-tmd] were used as dopants in an emitting layer (EML) to fabricate cost-effective and large-area CP-OLEDs by a wet method. To avoid the triplet-triplet annihilation of phosphorescent complexes, TCTA and 1,3-di-9-carbazolylbenzene (mCP) were selected as hole-transport host material, and 1,3-bis(5-(4-(tert-butyl)phenyl)-1,3,4-oxadiazol-2-yl)benzene (OXD-7) was selected as electron-transport host materials, which possess high triplet state energy (2.5-3.3 eV) to ensure an efficient energy transfer from the host to phosphorescent complexes.<sup>48-53</sup> Besides, mixed hole-transport and electrontransport host materials could effectively promote the carrier balance in OLEDs. The HOMO and LUMO energy levels of binuclear Pt(II) complexes lie between the host materials TCTA, mCP, and OXD-7, which further indicates that they can be used as host materials in CP-OLEDs. The device architecture and chemicals used in CP-OLEDs are listed in Figure 10. The optimal device structure is indium tin oxide (ITO) glass/poly(2,3-dihydrothieno-1,4-dioxin)-poly-(styrenesulfonate) (PEDOT:PSS) (30 nm)/blended host materials: Pt(II) complex (50 nm)/1,3,5-tris(1-phenyl-1Hbenzo[d]imidazol-2-yl)benzene (TPBI) (30 nm)/LiF (1 nm)/ Al (100 nm). PEDOT:PSS, TPBI, and LiF served as a holeinjection layer (HIL), electron-transporting layer (ETL)/holeblocking layer, and electron-injecting layers, respectively. The EMLs were prepared through spin-coating from a chlorobenzene solution of Pt(II) complex (10% in weight) in the mixed host of TCTA/mCP/OXD-7 (1,1:1).

In general, OLED layers are amorphous. The morphology of the spin-coated HIL and EML were characterized by an atomic force microscope (AFM). (R)-[tmd]- and (R)-[H<sub>8</sub>-tmd]- containing EMLs were deposited on the HIL of PEDOT:PSS. The HIL of PEDOT:PSS has a rough surface with a root-mean-square (RMS) roughness of 1.31 nm (Figure S13). If further depositions of EML were made, the RMS surface roughness would reduce to 0.61 and 0.47 nm for (R)-[tmd] and (R)-[H<sub>8</sub>-tmd], respectively. It is evident that the EML would help to generate a smothered surface morphology with fewer islandlike features. This would prevent the formation of electrical shorts and nonemissive dark spots and, eventually, improve the performance of solution-processed OLEDs.



Figure 10. Device architecture (a, c) and chemical structures (b) of the materials used in CP-OLEDs.

The EL, current density,  $\eta_L$ , and EQE curves of CP-OLEDs are shown in Figure 11. The EL spectra of (R)/(S)-[tmd] are almost identical and have a good agreement with their PL spectra. (R)/(S)-[tmd]-based CP-OLEDs emit red light ( $\lambda_{em}$ = 605 and 642 nm) with the 1931 Commission Internationale de l'Eclairage (CIE 1931) coordinates of (0.62, 0.38), which are comparable to those of a pure red emission (0.65, 0.35). Their maximum EQE,  $\eta_L$ ,  $\eta_P$ , and brightness are 1.4%, 2.9 cd A<sup>-1</sup>, 1.7 lm W<sup>-1</sup>, and 1470 cd m<sup>-2</sup>, respectively. For (R)/(S)-[H<sub>8</sub>-tmd]-based CP-OLEDs, the EL spectra blue shift to  $\lambda_{em} =$ 597 nm (CIE 1931: 0.59, 0.40) with higher EL performances (EQE,  $\eta_{\rm L}$ ,  $\eta_{\rm P}$ , and brightness up to 3.1%, 5.4 cd A<sup>-1</sup>, 3.4 lm  $W^{-1}$ , and 3070 cd m<sup>-2</sup>, respectively). Compared with those of achiral mononuclear analogues (vacuum-evaporated OLEDs),<sup>54</sup> the EL performances of the binuclear Pt(II) complexes (solution-processed OLEDs) still have much room for further improvement.

The circularly polarized electroluminescence properties of OLEDs were studied as well. As displayed in Figure 12, (*R*)/(*S*)-[**tmd**] and (*R*)/(*S*)-[**H<sub>8</sub>-tmd**]-based CP-OLEDs emit strong and broad mirror-image CPEL signals (500–750 nm) with the maximum  $|g_{EL}|$  of 3.0 × 10<sup>-3</sup> and 2.0 × 10<sup>-3</sup>, respectively. These  $g_{EL}$  data are in the same region with those of the most reported CP-OLEDs.<sup>1–5</sup> In addition, a self-assembled supramolecule technology is widely used for chirality transfer and amplification.<sup>5S–57</sup> However, this stage might be not so efficient for CP-OLEDs. The EMLs of OLEDs usually are amorphous layers, and thus the electrons and holes can efficiently recombine in an EML to form excitons. Islandlike crystalline materials acting as dark spots would lead to electrical shorts and poor performances in OLEDs.<sup>58</sup>

#### CONCLUSIONS

In conclusion, a novel series of chiral BINA and  $H_8$ -BINAbridging binuclear Pt(II) complexes was designed and prepared for solution-processed CP-OLEDs. Because of the effect of steric hindrance from bridging ligands and 2,3position extension in chiral axis planes, the binuclear Pt(II) complexes are highly phosphorescent in both solution and solid states. Moreover, on the one hand, BINA-bridging complexes would have a higher asymmetry factor but a lower PL/EL efficiency. On the other hand,  $H_8$ -BINA-bridging complexes would have a contrary tendency. There is a trade-off between the PL/EL efficiency and the asymmetry factor. Therefore, we believe that the present work might potentially provide a new way to design chiral multinuclear complexes for CPL applications.

#### EXPERIMENTAL SECTION

Materials and Instrumentation. All reagents were obtained commercially and utilized directly. <sup>1</sup>H NMR (400 MHz) spectra were measured in CDCl<sub>3</sub> and C<sub>6</sub>D<sub>6</sub>. Tetramethylsilane was used as an internal standard to measure chemical shifts in parts per million. UV/ Vis absorption spectra were done by a U-5100 (Hitachi) spectrophotometer with quartz cuvettes of 1 cm path length. An F-7000 fluorescence spectrophotometer (Hitachi) was used to measure fluorescence spectra at room temperature; slit width = 5.0 nm, photon multiplier voltage = 400 V. CD spectra were done by a Chirascan plus qCD (Applied Photophysics) at room temperature. Emission decay lifetime spectra were measured by a Horiba Tempro-01 (Horiba Scientific). The CPL spectra were performed by a JASCO CPL-300 spectro-fluoropolarimeter at room temperature. High-resolution mass spectra (HR-MS) were obtained with a Shimadzu LCMS-IT-TOF (ESI). The synthetic routes of binuclear Pt(II) complexes were described in detail in the Supporting Information.

**Measurement of Cyclic Voltammetry.** CV was measured by a CHI 620E electrochemical analyzer in degassed  $CH_2Cl_2$  solutions with *n*-Bu<sub>4</sub>NPF<sub>6</sub> as the supporting electrolyte. The CV cell consisted of a glassy carbon electrode, a Pt wire counter electrode, and an Ag/AgCl reference electrode. The ferrocene/ferrocenium (Fc/Fc+) was used as a reference. The HOMO energy-level calculations of the corresponding complexes were performed using the first oxidation potential, and LUMO energy levels were calculated according to the equations LUMO (eV) =  $E_g$  + HOMO.

OLED Device Fabrication and Characterization. Patterned indium-tin oxide (Lumtec) substrates were cleaned by a sonication in detergent, ethanol, acetone, and isopropyl alcohol continuously, followed by the UV-O3 treatment for 30 min. The hole injection layer (PEDOT: PSS, Al4083) was spin-coated on the preprocessed ITO substrates through a 0.22  $\mu$ m filter at a spin-coating rate of 4500 rpm for 40 s and oven-dried at 120 °C for 30 min to form a film of 30 nm thickness. The emitting layer was spin-coated through a filter with blend host materials and Pt(II) complex in chlorobenzene solution (20 mg/mL) and dried at 95 °C for 20 min. The thicknesses of spincoated layers were examined on a KLA-TENCOR D-100 profiler. After that, the substrates were transferred to a vacuum chamber. A 50 nm portion of an electron-transporting layer (TPBI), 1 nm of LiF, and 100 nm of Al were thermally evaporated consecutively under a vacuum ( $<1 \times 10^{-6}$  Torr, Mbraun MB200). The layer thickness was measured by quartz crystal monitors during the deposition. All



**Figure 11.** (a) Current density and luminance; (b)  $\eta_L$  and EQE; (c) EL of (*R*)/(*S*)-[**tmd**]. (d) Current density and luminance; (e)  $\eta_L$  and EQE; (f) EL of (*R*)/(*S*)-[**H**<sub>8</sub>-**tmd**]. (g) Photograph of CP-OLED.



Figure 12. CPEL properties of (R) (orange) /(S)-[tmd]-based (black) (a) and (R) (orange) /(S)-[H<sub>8</sub>-tmd]-based (black) (b) CP-OLEDs.

OLEDs were encapsulated by a UV adhesive under  $\rm N_2.$  The current–voltage–luminance data were done by Keithley 2400 and Photo Research PR680.

**Measurement of Atomic Force Microscopy.** Film morphologies were measured by an MFP3D-Stand Alone scanning probe in the tapping mode. The thin films were fabricated in a similar way to that of OLEDs fabrication.

**Measurement of Fluorescence Quantum Yield (\Phi).** The  $\Phi$  of degassed solution was done by the optical dilute method of Demas and Crosby.<sup>40</sup> The  $\Phi$  of powder and film was performed by an integrating sphere.

**X-ray Crystallographic Analysis.** The method of X-ray crystallographic analysis was similar to that of the previous work.<sup>40</sup>

**Computational Details.** DFT and TD-DFT calculations (PBE0/SDD/6-31G) were done by Gaussian 09 software. The ground-state geometry was optimized by DFT based on the X-ray single-crystal structure of Pt(II) complexes. After that, the optimized structure was used to calculate the UV/vis absorption and CD spectra by TD-DFT (CH<sub>2</sub>Cl<sub>2</sub> as the solvent, pcm method, 100 singlet—singlet transitions).

**Measurement of CP-EL Spectra.** CP-EL Spectra of CP-OLEDs were obtained by fixing the encapsulated OLED substrates in the groove of a JASCO circular polarization spectrophotometer (CPL-300) and applying a 6–7 V voltage under a scanning speed of 200 nm min<sup>-1</sup> with the "Continuous" mode. The measurement adopts the "slit" mode with an  $E_m$  slit width of 3000  $\mu$ m and multiple accumulations (three cycles).

#### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.inorgchem.1c01861.

General, materials, computational details, AFM images, CV spectra, optical spectra, (R)-[tmd] (CCDC: 2086886), and (S)-[H<sub>8</sub>-acac] (CCDC: 2086879) (PDF)

#### **Accession Codes**

CCDC 2086879 and 2086886 contain the supplementary crystallographic data for this paper. These data can be obtained free of charge via www.ccdc.cam.ac.uk/data\_request/cif, or by emailing data\_request@ccdc.cam.ac.uk, or by contacting The Cambridge Crystallographic Data Centre, 12 Union Road, Cambridge CB2 1EZ, UK; fax: +44 1223 336033.

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#### Notes

The authors declare no competing financial interest.

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