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Introduction

Organic light-emitting diodes (OLEDs) have received considerable attention in recent years because of their promising potential in flexible flat-panel displays and solid-state lighting.^{1–3} High-performance OLEDs rely on the efficient conversion of both electronically excited singlet (25%) and triplet (75%) excitons into photons for electroluminescence with high external quantum

Intermolecular locking design of red thermally activated delayed fluorescence molecules for high-performance solution-processed organic light-emitting diodes[†]

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Design of high-performance red thermally activated delayed fluorescence (TADF) materials remains a great challenge owing to their small energy bandgaps with severe nonradiative decay for low luminous efficiency. Introducing rigid and fused moieties is an effective way to enhance the luminescence of red emitters, but the solubility is also significantly reduced, prohibiting inevitably their applications in solution-processed TADF organic light-emitting diodes (OLEDs). Herein, we propose an intermolecular locking strategy to improve both the solution processibility and photoluminescence efficiency of red TADF emitters by using a highly-soluble flexible difluoroboron β -diketonate unit with exposed and easily reachable fluorines that can form hydrogen bonds to induce strong intermolecular locking in the solid state for high luminescent efficiency. The thus designed emitters show excellent device performance in solution-processed TADF OLEDs with a low turn-on voltage of 4.4 V, red electroluminescence around 600 nm, a high external quantum efficiency up to 8.2% and a small efficiency roll-off of 9.0% at 1000 cd m⁻², which are among the best results of solution-processed red OLEDs. These results demonstrate that the intermolecular locking strategy by directly addressing the internal conflicts between solubility and luminescent efficiency provides important clues in developing highly efficient and solution-processible red emitters for high-performance OLEDs.

efficiency (EOE).⁴⁻⁷ To overcome the limitation imposed by the spin-forbidden triplet exciton emission, many efforts have been made by using noble-metal complexes with spin-allowed triplet exciton emission for phosphorescent OLEDs,8,9 thermally activated delayed fluorescence (TADF) molecules to transform triplet excitons to singlet ones for TADF OLEDs,¹⁰⁻¹³ and even stable organic radicals with extraordinary doublet emission for radicalbased OLEDs.14,15 Among them, metal-free purely organic TADF materials with efficient reverse intersystem crossing (RISC) under a significantly reduced singlet-triplet splitting energy ($\Delta E_{\rm ST}$) to harvest both singlet and triplet excitons for 100% internal quantum efficiency of the device are the most attractive since their report in 2009,16 and great progress has been achieved in both blue and green TADF OLEDs with EQEs over 30%.17-19 Unfortunately, red TADF emitters lag far behind in both investigations and device performance, mainly owing to their narrow bandgaps with substantial energy loss via nonradiative decay as governed by the energy gap law.^{20,21}

Generally, rigid and fused moieties have been proved to be effective to suppress nonradiative transitions in designing high-efficiency red TADF materials (Fig. 1a).^{22,23} Red TADF OLEDs fabricated by vacuum thermal deposition technology

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Fig. 1 (a) Design of highly efficient red TADF materials for solution processing by the intermolecular locking strategy. (b) Molecular structures of the designed red TADF emitters.

show high EQEs over 20%.^{24–26} But, the rigid molecular framework usually leads to poor solubility, which significantly prohibits their application in solution-processed OLEDs, although solutionprocessing has been widely recognized to have significant advantages, especially in low-cost and large-area production for commercial manufacture.²⁷ A few solution-processed red TADF OLEDs with EQE around 10% have been achieved by introducing the widely used *tert*-butyl group to improve the solubility of organic molecules.^{28–31} However, the turn-on voltages of these solution-processed devices are up to 7.0 V with large efficiency roll-off, owing to the steric hindrance for inferior carrier transportation and non-conductivity of the introduced *tert*-butyl group.²⁹

Herein, we propose an intermolecular locking strategy to solve the above dilemma by combining a flexible molecular architecture for good solubility in solution and strong intermolecular interactions in aggregation to improve the emission efficiency for highly efficient solution-processed red TADF OLEDs (Fig. 1a). Strong intramolecular charge transfer (CT) characteristics for red emission and typical TADF features with small $\Delta E_{\rm ST}$ and efficient RISC processes were facilely realized by using a strong acceptor of difluoroboron β-diketonate.³² This rarely used acceptor unit is rather flexible to enable high solubility of the TADF materials in common organic solvents. But, more importantly, this unit with an exposed acceptor core of difluoroboron enables abundant and multiple intermolecular hydrogen bond interactions between the easily reachable fluorine on the difluoroboron group and the surrounding hydrogen atoms of the aromatic donor units on the nearby molecules to suppress nonradiative transitions. Thus, the photoluminescence quantum yield (PLQY) increases significantly from 22% in dichloromethane (CH2Cl2) to 54% in the pure film, and to 97% when doped (5 wt%) into 1,3-bis(N-carbazolyl)benzene (mCP). The solution-processed red (~ 600 nm) TADF OLEDs based on these red emitters exhibit excellent performance with a low turn-on voltage of 4.4 V, an EQE up to 8.2% and small efficiency roll-offs of 3.7% and 9.0% at 100 and 1000 cd m^{-2} , respectively. These performances are comparable to the best results of red TADF OLEDs (Table S1 and Scheme S1, ESI⁺).

Results and discussion

Molecular design and synthesis

To construct the TADF molecules with intermolecular interlocking, difluoroboron β-diketonate with strong electronwithdrawing ability is selected as an acceptor unit and the widely used phenylcarbazole or triphenylamine as a donor unit in a donor-acceptor-donor (D-A-D) architecture, typically shown in DPhCzB and DTPAB (Fig. 1b). The thus designed compounds were facilely synthesized through a conventional three-step reaction of Claisen condensation, C-N coupling, and a borylation reaction with high total yields up to 39% (Scheme S2, ESI[†]), and were fully characterized by ¹H and ¹³C NMR spectroscopy, high resolution mass spectrometry, and singlecrystal X-ray diffraction (Fig. S1-S6 and Table S2, ESI⁺). Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) reveal that these D-A-D molecules have excellent thermal stability with high decomposition temperatures (T_d) over 380 °C, glass-transition temperatures (T_g) over 130 °C and melting temperatures (T_m) over 200 °C (Fig. S7, ESI†), which are among the highest thermal stabilities of organic boron-containing optoelectronic materials. Also, the compounds exhibit excellent solubility in common solvents (Table S3, ESI[†]). Especially, DTPAB has superior solubility (>100 mg mL⁻¹) and a smooth film morphology with a small root-mean-square (RMS) roughness of 0.206 nm when spin-coated on an ITO substrate owing to its flexible diphenylamine compared to the rigid carbazole in DPhCzB (Fig. S8, ESI⁺). Therefore, promising applications of these molecules in solution-processed OLEDs can be highly expected.

Photophysical properties

The photophysical properties of DPhCzB and DTPAB were studied using ultraviolet-visible (UV-vis) absorption and photoluminescence (PL) spectra in dilute toluene solution $(10^{-5} \text{ mol L}^{-1})$ and solid films. Both emitters show similar UV-vis absorption spectra in toluene solution (Fig. 2a), exhibiting one distinct absorption band peaking at around 295 nm ascribed to the carbazole or diphenylamine-centered π - π * transition, and a weaker absorption at around 340 nm that can be assigned to the n- π^* transition of the entire molecule.³³ The strongest absorption bands of DPhCzB and DTPAB were at 468 and 502 nm owing to the facile CT transition from the donor of carbazole or diphenylamine to the acceptor of the difluoroboron β -diketonate group. DTPAB shows a more red-shifted CT absorption peak because of the stronger electron-donating ability of the diphenylamine unit than carbazole for a stronger CT transition. This CT state also shows a broad and structureless CT emission band peaking at 538 and 560 nm for DPhCzB and DTPAB in toluene solution, and was further confirmed by the large variations of their emission bands in solvents with different polarities (Fig. S9, ESI⁺). When turned into the solid thin film state, DPhCzB and DTPAB exhibit significantly red-shifted (up to 33 nm) CT absorption bands to 501 and 528 nm compared to those in solution owing to the extended π -conjunction and strong intermolecular interaction (Fig. 2b). The red-shift (up to 100 nm) of their PL spectra is even larger in the film state for typical red emission at 637 and 650 nm, indicating

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Fig. 2 (a and b) UV-vis absorption and photoluminescence spectra in (a) toluene solution and (b) solid films, (c) fluorescence and phosphorescence (delay 5 ms) spectra in toluene at 77 K and (d) transient PL decay curves of these intermolecular locking molecules (5 wt%) doped in *m*CP films at room temperature of **DPhCzB** (black) and **DTPAB** (red).

again strong intermolecular interactions of these molecules in the solid state.

From the onset thresholds of the absorption spectra in solid films, the optical band gaps ($E_{\rm g}$ s) of **DPhCzB** and **DTPAB** were identified to be as narrow as 2.03 and 1.96 eV, respectively (Table 1). From the PL and time-resolved phosphorescence spectra measured at 77 K in toluene, a small $\Delta E_{\rm ST}$ of 0.15 and 0.17 eV can be evaluated for **DPhCzB** and **DTPAB**, respectively (Fig. 2c). With such a small $\Delta E_{\rm ST}$, effective RISC for TADF can be expected. Indeed, the transient PL decay curves of the two D–A–D molecules doped into *m*CP films (5 wt%) exhibit a prompt decay followed by a delayed decay with corresponding lifetimes of 15.7 ns and 19.5 µs for **DPhCzB** and 3.7 ns and 55.7 µs for **DTPAB**, respectively (Fig. 2d and Fig. S10, ESI†), providing direct evidence of their TADF characteristics. Moreover, the two new red TADF emitters of **DPhCzB** and **DTPAB** show acceptable PLQYs of 22% and 44% in

 CH_2Cl_2 owing to the flexible molecular structures with severe molecular vibrations in the single-molecule state, but exhibit much increased values to 54% and 56% in pure films, illustrating obvious aggregation enhanced emission (AEE) behavior owing to the suppressed nonradiative decays in solid films. When the red emitters were doped (5 wt%) into *m*CP, the PLQYs further increase to 87% and 97% for **DPhCzB** and **DTPAB**, respectively. The high PLQYs of these red TADF emitters in either pure or doped films should be highly attractive for high-efficiency OLEDs.

Intra- and inter-molecular interaction investigation

To understand the significantly red-shifted UV absorption and PL emission and increased PLQYs of the red TADF molecules in the solid state, the intra- and intermolecular interactions were investigated. The reduced density gradient (RDG) isosurface calculations of **DTPAB** and **DPhCzB** in the single molecule state based on

Table 1 Photophysical and electrochemical properties of DPhCzB and DTPAB												
	λ_{abs} (nm)		$\lambda_{\rm em} ({\rm nm})$				τ^d				CV (eV)	
Compound	Tol.	Film	$E_{\rm g}^{\ a}$ (eV)	Tol.	Film	ΔE_{ST} (eV)	$\tau_{\rm PF}\left(ns\right)$	$\tau_{\rm DF}~(\mu s)$	PLQY		номо	LUMO ^e
DPhCzB DTPAB	292, 468 300, 501	294, 501 301, 528	2.03 1.96	538 560	637 650	0.15 0.17	15.7 3.7	19.8 55.7	$\begin{array}{c} 0.54^b \\ 0.56^b \end{array}$	0.87 ^c 0.97 ^c	$\begin{array}{c} -5.41 \\ -5.36 \end{array}$	$-3.38 \\ -3.40$

 $^{a}E_{g}$ was estimated from the absorption onset in neat films. b Absolute PLQY measured in pure films at room temperature. c Absolute PLQY measured in doped films at room temperature. d Measured in doped films. e LUMO energy level estimated by adding E_{g} to the HOMO energy level.



Fig. 3 (a) Molecular packing and inter/intra-molecular interactions in the **DPhCzB** crystal. (b) PLQY values of *m*CP blend films doped with **DPhCzB** and **DTPAB** at various doping concentrations (in wt%). (c) Reduced density gradient (RDG) *versus* sign(λ_2) ρ with the view of the RDG isosurface of the **DPhCzB** dimer.

their optimized ground molecular structures (Fig. S11, ESI⁺) show heavy intramolecular interactions, especially in DPhCzB. From the single crystal structure analyses, abundant intra/intermolecular interactions with short distances can be observed with strong intermolecular hydrogen bonds of C-H···F and C-H···O, owing to the exposed difluoroboron β-diketonate group with multiple fluorine and oxygen atoms in forming hydrogen bonds (Fig. 3a). The heavy intra- and intermolecular interactions in the solid state offer a rigid environment to dramatically suppress the nonradiative decay for high luminescent efficiency, which explains the bathochromic shift of the PL peaks and much improved PLQY in film states. Further, the PLQYs of the doped films with different concentrations were measured (Fig. 3b). The increased PLQY at reduced doping concentration indicates the strong intermolecular interactions between the emitters and mCP to well disperse the emitters in mCP owing to the easily reachable fluorine on the difluoroboron group for hydrogen bonding. Furthermore, the intermolecular interaction was theoretically verified by RDG isosurface calculations based on the single crystal structure of DPhCzB.³⁴ Strong intermolecular attractive and repulsive interactions with denser interaction regions were theoretically revealed (Fig. 3c). Therefore, the intermolecular locking strategy solves the intrinsic competition between solubility and luminescent efficiency, resulting in extraordinary excellent solubility, good film morphology, and high luminescent efficiency for highperformance solution-processed red OLEDs.

Electrochemical and computational investigations

The electrochemical properties of the red TADF emitters were investigated by cyclic voltammetry (CV) using tetrabutyl-

ammonium hexafluorophosphate (TBAPF₆) as the supporting electrolyte and ferrocene as the internal standard (Fig. S12, ESI⁺). From the onset of the oxidation wave, the highest occupied molecular orbital (HOMO) energy levels of DPhCzB and DTPAB were measured to be -5.41 and -5.36 eV, respectively. With the aid of the optical band gaps, the lowest unoccupied molecular orbital (LUMO) energy levels were estimated to be -3.38 and -3.40 eV. It should be noted that these HOMO and LUMO energy levels are very close to those of widely used hole-transporting and electron-transporting materials, suggesting their high potential in optoelectronic device applications. To evaluate their frontier molecular orbital distributions and electronic structures, density functional theory (DFT) computations were performed (Fig. 4). Similar to typical TADF molecules, the HOMOs of these molecules are dominantly located on the donor segments of phenylcarbazole or triphenylamine, while the LUMOs are mainly distributed on the acceptor segments of difluoroboron β-diketonate, leading to partly separated frontier orbital distributions with moderate overlap extents $(I_{H/L})$ of 37.34% and 48.52% between the HOMO and LUMO for small ΔE_{ST} and high PLQYs. Moreover, by comparing to the theoretically optimized single molecule geometry in the ground state, the geometry in the crystal becomes more planar with a reduced dihedral angle between difluoroboron β -diketonate and phenyl from 12.7° to 7.53° (Scheme S3, ESI[†]), indicating that the intermolecular hydrogen bonding interaction can also conformationally turn the molecules more planar for a higher luminescent efficiency and AEE phenomenon.

Solution-processed red TADF OLEDs

In light of the excellent solubility and high PLQYs of the red TADF molecules designed by the intermolecular locking



Fig. 4 Experimental frontier molecular orbital energy levels of **DPhCzB** and **DTPAB** as well as their simulated frontier orbital distributions and overlap extents $(I_{H/L})$.

strategy, solution-processed OLEDs using these red TADF materials as emitters were fabricated in a device configuration of ITO/PEDOT:PSS (40 nm)/mCP:5 wt% red TADF emitter (50 nm)/DPEPO (10 nm)/TmPyPB (50 nm)/Liq (1 nm)/Al (100 nm) (Fig. 5a and Fig. S13, ESI†). In these devices, indium tin oxide (ITO) and Al act as the anode and the cathode, respectively; poly(3,4-ethylenedioxythiophene):poly (styrenesulfonic acid) (PEDOT:PSS) and 8-hydroxyquinolinolatolithium (Liq) serve as the hole- and electron-injecting layer, respectively; and *m*CP was selected as the host material, bis [2-(diphenylphosphino)phenyl]ether oxide (DPEPO) as the exciton-blocking layer, and 1,3,5-tri[(3-pyridyl)-phen-3-yl] benzene (TmPyPB) as the hole-blocking and electron-transporting material.

Efficient red electroluminance (EL) peaking at 587 nm and 605 nm was observed in the OLEDs, suggesting the exothermic energy transfer from mCP to the guest molecules (Fig. 5b), and only the emission bands of the TADF emitters were observed in light of the consistent EL emission with the PL spectra of their doped (5 wt%) mCP films (Fig. S14, ESI⁺). Excitingly, these devices exhibit impressive high performance with low turn-on voltages (V_{on}) of 4.7 and 4.4 V, maximum EQEs of 6.7% and 8.2%, maximum current efficiencies (CEs) of 17.0 and 16.4 cd A^{-1} , and maximum power efficiencies (PEs) of 5.9 and 7.4 lm W^{-1} for OLEDs based on **DPhCzB** and **DTPAB**, respectively (Fig. 5c and d). Moreover, both devices have low efficiency roll-offs, which are as low as 3.7% and 9.0% at 100 and 1000 cd m⁻², respectively. The low efficiency roll-off could be related to the short lifetime of excitons to avoid triplet excited state-related quenching effects; **DPhCzB** exhibits a short lifetime of 19.5 µs, while DTPAB has a lifetime of 55.7 µs (Table 1). In addition, the strong intermolecular interactions can also facilitate the



Fig. 5 (a) Device configuration, (b) electroluminescence spectra at 7.0 V, (c) current density-voltage (open symbols) and luminance-voltage (solid symbols) curves and (d) efficiency-current density curves of solution-processed red TADF OLEDs.

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transportation/injection of charge carriers and improve the film stability of the emission layer, resulting in good device stability. The maximum EQEs of the OLEDs based on the two TADF emitters are significantly higher than the fluorescence efficiency limit of 5%, indicating that the 75% electronically excited triplet excitons are successfully harvested for EL by their facile RISC in the TADF mechanism with an exciton utilization efficiency of 39.0% and 57.7% in the **DPhCzB**- and **DTPAB**-based OLEDs,³⁵ respectively. It should be noted that this performance is among the best results of solution-processed red OLEDs.

Conclusions

In summary, we propose a new strategy, namely inter-molecular locking, to design highly efficient red TADF emitters with excellent solubility for high-performance solution-processed OLEDs. Based on flexible difluoroboron β -diketonate with exposed and easily reachable fluorine in forming hydrogen bonds with the surrounding hydrogen atoms, two novel red TADF molecules of DPhCzB and DTPAB were facilely prepared, exhibiting excellent solubility in common organic solvents and red emission beyond 630 nm with high PLQYs over 55% in neat films due to the rigidified molecules in the solid state by intermolecular hydrogen bonding to suppress the nonradiative decay significantly. Using DPhCzB and DTPAB as emitters in a simple solution-processed device structure, highly efficient red TADF OLEDs with a low turn-on voltage of 4.4 V, high EQE up to 8.2% and low efficiency roll-off of 9.0% at 1000 cd m^{-2} were realized. These impressive device performances with the aid of the intermolecular locking strategy to address the inherent contradictions between the solubility and luminescent efficiency of organic molecules represent an important step forward in designing highly efficient solution-processible organic optoelectronic materials, especially red light emitting TADF molecules, for advanced device applications.

Conflicts of interest

There are no conflicts to declare.

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Notes and references

 X. Tang, L. S. Cui, H. C. Li, A. J. Gillett, F. Auras, Y. K. Qu, C. Zhong, S. T. E. Jones, Z. Q. Jiang, R. H. Friend and L. S. Liao, *Nat. Mater.*, 2020, **19**, 1332–1338.

- Y. C. Wei, S. F. Wang, Y. Hu, L. S. Liao, D. G. Chen, K. H. Chang, C. W. Wang, S. H. Liu, W. H. Chan, J. L. Liao, W. Y. Hung, T. H. Wang, P. T. Chen, H. F. Hsu, Y. Chi and P. T. Chou, *Nat. Photonics*, 2020, 14, 570–577.
- 3 Y. Wada, H. Nakagawa, S. Matsumoto, Y. Wakisaka and H. Kaji, *Nat. Photonics*, 2020, **14**, 643–649.
- 4 T. Wang, X. Su, X. Zhang, X. Nie, L. Huang, X. Zhang, X. Sun, Y. Luo and G. Zhang, *Adv. Mater.*, 2019, **31**, 1904273.
- 5 S. F. Wang, Y. Yuan, Y. C. Wei, W. H. Chan, L. W. Fu, B. K. Su, I. Y. Chen, K. J. Chou, P. T. Chen, H. F. Hsu, C. L. Ko, W. Y. Hung, C. S. Lee, P. T. Chou and Y. Chi, *Adv. Funct. Mater.*, 2020, **30**, 2002173.
- 6 M. C. Xie, C. M. Han, Q. Q. Liang, J. Zhang, G. H. Xie and H. Xu, *Sci. Adv.*, 2019, 5, eaav9857.
- 7 J. Song, H. Lee, E. G. Jeong, K. C. Choi and S. Yoo, Adv. Mater., 2020, 32, e1907539.
- 8 Y. Tao, L. Xu, Z. Zhang, R. Chen, H. Li, H. Xu, C. Zheng and W. Huang, J. Am. Chem. Soc., 2016, 138, 9655–9662.
- 9 K. Tuong Ly, R. W. Chen-Cheng, H. W. Lin, Y. J. Shiau, S. H. Liu, P. T. Chou, C. S. Tsao, Y. C. Huang and Y. Chi, *Nat. Photonics*, 2016, **11**, 63–68.
- 10 Y. Tao, K. Yuan, T. Chen, P. Xu, H. Li, R. Chen, C. Zheng, L. Zhang and W. Huang, *Adv. Mater.*, 2014, 26, 7931–7958.
- 11 Y. Liu, C. Li, Z. Ren, S. Yan and M. R. Bryce, *Nat. Rev. Mater.*, 2018, **3**, 18020.
- 12 W. Yuan, H. Yang, C. Duan, X. Cao, J. Zhang, H. Xu, N. Sun, Y. Tao and W. Huang, *Chem*, 2020, 6, 1998–2008.
- 13 S. Xu, Q. Yang, Y. Wan, R. Chen, S. Wang, Y. Si, B. Yang, D. Liu, C. Zheng and W. Huang, *J. Mater. Chem. C*, 2019, 7, 9523–9530.
- 14 X. Ai, E. W. Evans, S. Dong, A. J. Gillett, H. Guo, Y. Chen, T. J. H. Hele, R. H. Friend and F. Li, *Nature*, 2018, 563, 536–540.
- 15 H. Guo, Q. Peng, X. K. Chen, Q. Gu, S. Dong, E. W. Evans, A. J. Gillett, X. Ai, M. Zhang, D. Credgington, V. Coropceanu, R. H. Friend, J. L. Bredas and F. Li, *Nat. Mater.*, 2019, 18, 977–984.
- 16 A. Endo, M. Ogasawara, A. Takahashi, D. Yokoyama, Y. Kato and C. Adachi, *Adv. Mater.*, 2009, **21**, 4802–4806.
- 17 T. L. Wu, M. J. Huang, C. C. Lin, P. Y. Huang, T. Y. Chou, R. W. Chen-Cheng, H. W. Lin, R. S. Liu and C. H. Cheng, *Nat. Photonics*, 2018, **12**, 235–240.
- 18 D. H. Ahn, S. W. Kim, H. Lee, I. J. Ko, D. Karthik, J. Y. Lee and J. H. Kwon, *Nat. Photonics*, 2019, **13**, 540–546.
- 19 Y. Kondo, K. Yoshiura, S. Kitera, H. Nishi, S. Oda, H. Gotoh, Y. Sasada, M. Yanai and T. Hatakeyama, *Nat. Photonics*, 2019, 13, 678–682.
- 20 A. Zampetti, A. Minotto and F. Cacialli, *Adv. Funct. Mater.*, 2019, **29**, 1807623.
- 21 J. H. Kim, J. H. Yun and J. Y. Lee, Adv. Opt. Mater., 2018, 6, 1800255.
- 22 D. G. Congrave, B. H. Drummond, P. J. Conaghan, H. Francis, S. T. E. Jones, C. P. Grey, N. C. Greenham, D. Credgington and H. Bronstein, *J. Am. Chem. Soc.*, 2019, 141, 18390–18394.
- 23 Y. L. Zhang, Q. Ran, Q. Wang, Y. Liu, C. Hanisch, S. Reineke, J. Fan and L. S. Liao, *Adv. Mater.*, 2019, 31, e1902368.

- 24 J. X. Chen, K. Wang, C. J. Zheng, M. Zhang, Y. Z. Shi, S. L. Tao, H. Lin, W. Liu, W. W. Tao, X. M. Ou and X. H. Zhang, *Adv. Sci.*, 2018, 5, 1800436.
- 25 W. Zeng, H. Y. Lai, W. K. Lee, M. Jiao, Y. J. Shiu, C. Zhong, S. Gong, T. Zhou, G. Xie, M. Sarma, K. T. Wong, C. C. Wu and C. Yang, *Adv. Mater.*, 2018, **30**, 1704961.
- 26 J. X. Chen, W. W. Tao, W. C. Chen, Y. F. Xiao, K. Wang, C. Cao, J. Yu, S. Li, F. X. Geng, C. Adachi, C. S. Lee and X. H. Zhang, *Angew. Chem., Int. Ed.*, 2019, 58, 14660–14665.
- 27 Y. Tao, X. Guo, L. Hao, R. Chen, H. Li, Y. Chen, X. Zhang, W. Lai and W. Huang, *Adv. Mater.*, 2015, 27, 6939–6944.
- 28 J. X. Chen, W. W. Tao, Y. F. Xiao, K. Wang, M. Zhang, X. C. Fan, W. C. Chen, J. Yu, S. Li, F. X. Geng, X. H. Zhang and C. S. Lee, *ACS Appl. Mater. Interfaces*, 2019, **11**, 29086–29093.

- 29 W. Zeng, T. Zhou, W. Ning, C. Zhong, J. He, S. Gong, G. Xie and C. Yang, *Adv. Mater.*, 2019, **31**, 1901404.
- 30 Y. Zhou, M. Zhang, J. Ye, H. Liu, K. Wang, Y. Yuan, Y.-Q. Du,
 C. Zhang, C.-J. Zheng and X.-H. Zhang, *Org. Electron.*, 2019, 65, 110–115.
- 31 Y. Liu, Y. Chen, H. Li, S. Wang, X. Wu, H. Tong and L. Wang, *ACS Appl. Mater. Interfaces*, 2020, **12**, 30652–30658.
- 32 J. Jin, H. Jiang, Q. Yang, L. Tang, Y. Tao, Y. Li, R. Chen, C. Zheng, Q. Fan, K. Y. Zhang, Q. Zhao and W. Huang, *Nat. Commun.*, 2020, 11, 842.
- J. Jin, Y. Tao, H. Jiang, R. Chen, G. Xie, Q. Xue, C. Tao, L. Jin,
 C. Zheng and W. Huang, *Adv. Sci.*, 2018, 5, 1800292.
- 34 Y. Cheng, Y. Qi, Y. Tang, C. Zheng, Y. Wan, W. Huang and R. Chen, J. Phys. Chem. Lett., 2016, 7, 3609–3615.
- J. Liu, Z. Li, T. Hu, X. Wei, R. Wang, X. Hu, Y. Liu, Y. Yi,
 Y. Yamada-Takamura, Y. Wang and P. Wang, *Adv. Opt. Mater.*, 2018, 7, 1801190.