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1 Supplementary Information

2 Crystalline organic thin films for crystalline OLEDs (II): weak epitaxy growth of

- 3 phenanthroimidazole derivatives
- 4 Dan Liu^{1,2}, Feng Zhu^{1,2*}, Donghang Yan^{1,2}
- 5 ¹State Key Laboratory of Polymer Physics and Chemistry, Changchun Institute of
- 6 Applied Chemistry, Chinese Academy of Sciences, Changchun 130022, China.
- 7 ²School of Applied Chemistry and Engineering, University of Science and Technology
- 8 of China, Hefei 230026, China.
- 9 *Corresponding author. Email: <u>zhufeng@ciac.ac.cn</u>
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21 1. Morphologies of BP1T films grown at different substrate temperatures



Fig. S1 AFM images of BP1T films grown at different substrate temperatures. (a-d) 0.6 ML BP1T grown at 62 °C (a), 82 °C (b), 102 °C (c), and 112 °C (d). (e-f) 1.4 ML (e) and 1.6 ML (f) BP1T grown at 112 °C.

(c) (a) (b) 0.4 ML 0.8 ML 1.2 ML double-layer Si/SiO₂ monolayer monolayer monolayer 5 µm 5 µm 5 µm (d) (e) (f) 1.4 ML 1.8 ML 2.2 ML double-layer double-layer 5 µm 5 µm 5 µm 28

27 2. Morphologies of different thicknesses BP1T

Fig. S2 AFM images of 0.4 (a), 0.8 (b), 1.2 (c), 1.4 (d), 1.8 (e), and 2.2 (f) monolayer
BP1T films at the substrate temperature of 102 °C.

- 32 3. Width of 2FPPICz strip-like crystal on BP1T
 - (a) 1.2 ML BP1T / 20 nm 2FPPICz



(b) 2.2 ML BP1T / 20 nm 2FPPICz



Fig. S3 (a-b) Width of 2FPPICz strip-like crystals grown on 1.2 (a) and 2.2 (b) monolayer BP1T.

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- 37 4. Height of 2FPPICz crystals on double-layer BP1T
 - (a)



39 Fig. S4 Height map of 2-nm thick 2FPPICz thin film deposited on 2.2-monolayer BP1T.

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41 5. Morphologies of different thicknesses 2FPPICz before and after annealing



Fig. S5 (a and b) AFM images of unannealing (a) and annealing for 2 hours (b) 2.0-nm
thick 2FPPICz thin film. (d and e) AFM images of unannealing (d) and annealing for 2
hours (e) 4.0-nm thick 2FPPICz thin film. (c and f) The corresponding zoom images of
(b) and (e).

48 6. Morphologies of different thicknesses 2FPPICz on BP1T



- 50 Fig. S6 Morphologies of 2FPPICz with different thicknesses on BP1T. The AFM images
- 51 of different thicknesses of 10 nm (a), 20 nm (b), and 30 nm (c).

53 7. Optical properties of 2FPPICz crystalline film



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Fig. S7 Optical properties of 2FPPICz crystalline film. (a) Normalized ultraviolet-visible (UV-vis) absorption and photoluminescence (PL) spectrum of 2FPPICz crystalline thin film from [J. Mater. Chem. C, 2021, 9, 2236-2242]. (b) Time-resolved photoluminescence spectra of 40 nm-thick 2FPPICz crystalline thin film. (c) Transient decay curves of 40 nm-thick 2FPPICz crystalline thin film.

Em (nm)	A ₁	τ_1 (ns)	A ₂	τ_2 (ns)
385	1148.42	0.74	8.7	3.56
405	1032.68	0.75	36.49	2.98
440	1025.86	0.97	122.11	3.94

Table S1 Fluorescence lifetimes (τ) of 2FPPICz crystalline film depend on wavelength.