Electronic Supplementary Information for

Rapid improvements in charge carrier mobility at ionic liquid/pentacene single crystal interfaces by self-cleaning

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Fig. S1: Transfer characteristics of the air-gap FET.

Fig. S2: FM-AFM images of the pentacene surface in air.

Fig. S3: FM-AFM images of pentacene and rubrene surfaces.



Fig. S1. (a) Schematic of air-gap FET. (b) Transfer characteristics of the air-gap FET. Note that this result was obtained just before putting BMIM-TFSI into the air-gap to perform EDL-FET measurements shown in Fig. 2.



Fig. S2. FM-AFM images of the pentacene surface in air. (a)(b) $3 \times 3 \mu m^2$, $\Delta f = -100$ Hz, $A_{p-p} = 1.0$ nm, and $f_0 = 302.7$ kHz, and (c)(d) $1 \times 1 \mu m^2$, $\Delta f = -100$ Hz, $A_{p-p} = 0.9$ nm, and $f_0 = 302.7$ kHz. Z-ranges of color scale bar are (a) 160, (b) 3, (c) 30, and (d) 6 nm. The line profiles along the solid lines are depicted at the bottom.



Fig. S3. FM-AFM images of (a) fresh pentacene, (b) fresh rubrene, and (c) intentionally oxidized rubrene surfaces, taken in air. Characteristics of the images: (a) area: $3 \times 3 \mu m^2$, $\Delta f = -100 \text{ Hz}$, $A_{p-p} = 1.0 \text{ nm}$, and $f_0 = 302.7 \text{ kHz}$; (b) area: $1.5 \times 1.5 \mu m^2$, $\Delta f = -300 \text{ Hz}$, $A_{p-p} = 4.7 \text{ nm}$, and $f_0 = 263.3 \text{ kHz}$; and (c) (single crystal was stored under ambient air for a month) area: $300 \times 300 \text{ nm}^2$, $\Delta f = -200 \text{ Hz}$, $A_{p-p} = 4.4 \text{ nm}$, and $f_0 = 260.7 \text{ kHz}$. The details of the rubrene surfaces were reported in the previously conducted report [Y. Yokota, H. Hara, Y. Morino, K. Bando, A. Imanishi, T. Uemura, J. Takeya and K. Fukui, *Phys. Chem. Chem. Phys.*, 2015, **17**, 6794]. (d)–(f) Line profiles along the solid lines in (a)–(c), respectively. Z-ranges of color scale bar are (a) 160, (b) 0.7, and (c) 1.2 \text{ nm}.