

*Electronic Supplementary Information for*

**Molecularly Clean Ionic Liquid/Rubrene Single-Crystal Interfaces  
Revealed by Frequency Modulation Atomic Force Microscopy**

*Yasuyuki Yokota,<sup>a</sup> Hisaya Hara,<sup>a</sup> Yusuke Morino,<sup>a</sup> Ken-ichi Bando,<sup>a</sup>*

*Akihito Imanishi,<sup>a</sup> Takafumi Uemura,<sup>b, c</sup> Jun Takeya<sup>b, c, \*</sup>, and Ken-ichi Fukui<sup>a, \*</sup>*

<sup>a</sup> Department of Materials Engineering Science, Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan

<sup>b</sup> Department of Advanced Materials Science, Graduate School of Frontier Science, The University of Tokyo, Kashiwa, Chiba 277-8561, Japan

<sup>c</sup> The Institute of Scientific and Industrial Research (ISIR), Osaka University, Ibaraki, Osaka 567-0047, Japan

**Figs. S1 and S2:**

The FM-AFM images used for counting the number of defect sites.

**Fig. S3:**

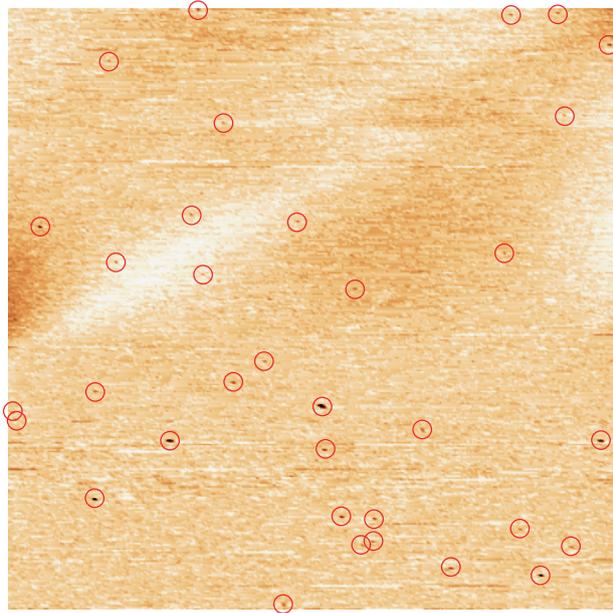
An FM-AFM image of rubrene (001) taken in water.

**Fig. S4:**

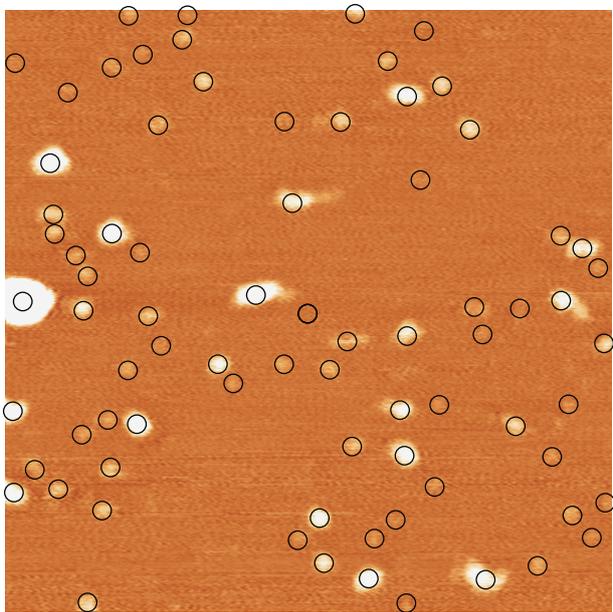
The force plotted with respect to the tip-sample distance calculated from the frequency shift curves in Fig. 4.

**Reference S1:**

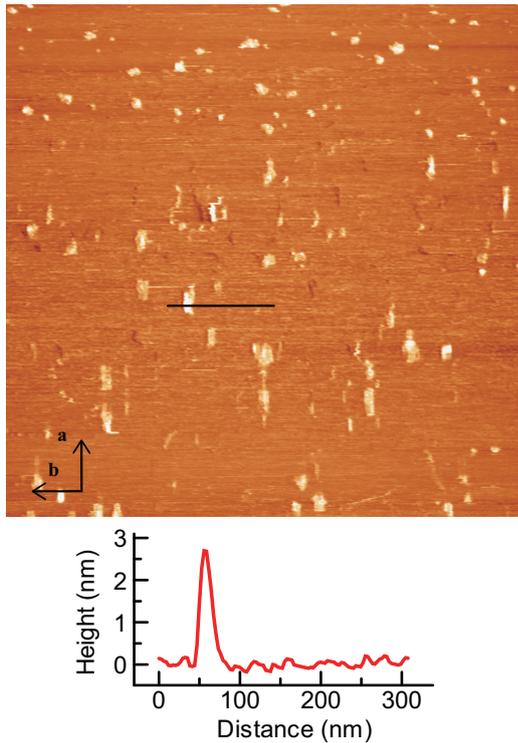
J. E. Sader and S. P. Jarvis, *Appl. Phys. Lett.*, 2004, **84**, 1801.



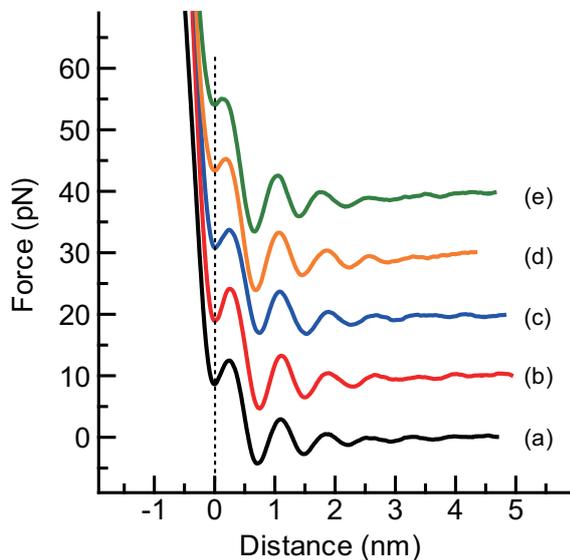
**Fig. S1.** The FM-AFM image of Fig. 1(b) ( $2 \times 2 \mu\text{m}^2$ ) with circles showing the position of defect sites. There were 34 identified defects, resulting in a defect density of  $8.5 \times 10^8 / \text{cm}^2$ .



**Fig. S2.** The FM-AFM image of Fig. 1(c) ( $300 \times 300 \text{nm}^2$ ) with circles showing the position of defect sites. There were 77 identified defects, resulting in a defect density of  $8.6 \times 10^{10} / \text{cm}^2$ .



**Fig. S3.** Typical FM-AFM image ( $1.5 \times 1.5 \mu\text{m}^2$ ) of the rubrene surface taken in water. Line profile along the solid line in the image is shown below. The following imaging conditions were used;  $\Delta f = +100 \text{ Hz}$ ,  $f_0 = 138.8 \text{ kHz}$ , and  $A_{p-p} = 0.50 \text{ nm}$ .



**Fig. S4.** (a)–(e) The force plotted with respect to the tip–sample distance calculated from the frequency shift curves in Fig. 4 using the method of Sader and Jarvis.<sup>S1</sup> Each curve is laterally shifted to align the local minimum nearest to the rubrene surface and vertically shifted by  $10 \text{ pN} \times n$  ( $n = 0, 1, 2, 3, 4$ ) for clarity.