

Electronic Supplementary Data for:

Assembling model tris(bipyridine)ruthenium(II) photosensitizers into ordered monolayers in the presence of the polyoxometallate $[\text{Co}_4(\text{H}_2\text{O})_2(\alpha\text{-PW}_9\text{O}_{34})_2]^{10-}$

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Fig. S1. ^1H NMR spectrum (400 MHz, CDCl_3) of compound **1**; * = residual CHCl_3 ; ** = water.

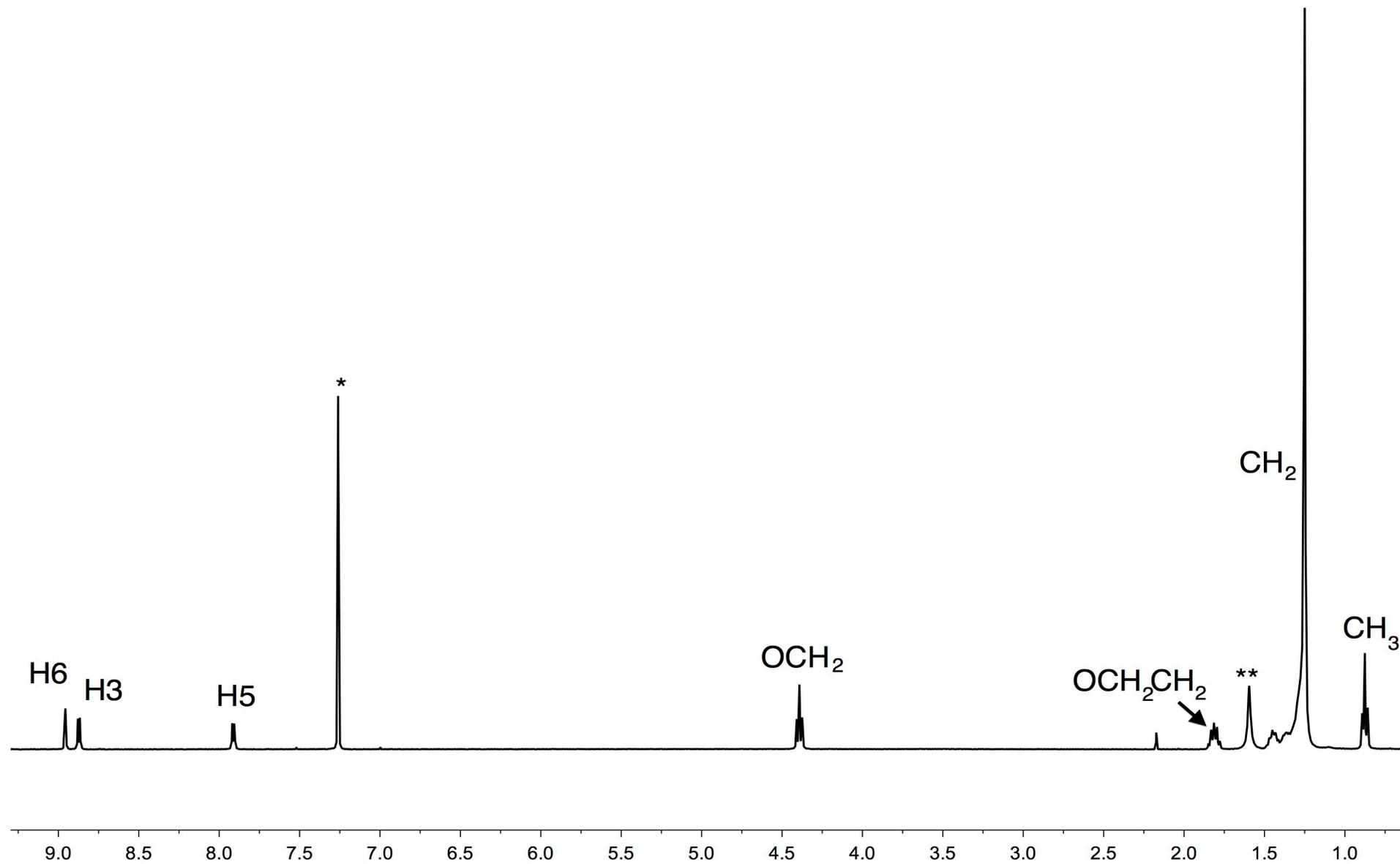


Fig. S2. Brewster angle microscopy images of *cis*-[Ru(**1**)₂Cl₂] on a water subphase at (a) 0.00 mN/m, (b) 0.10 mN/m (c) 3.32 mN/m, (d) 13.00 mN/m and (e) 24.00 mN/m.

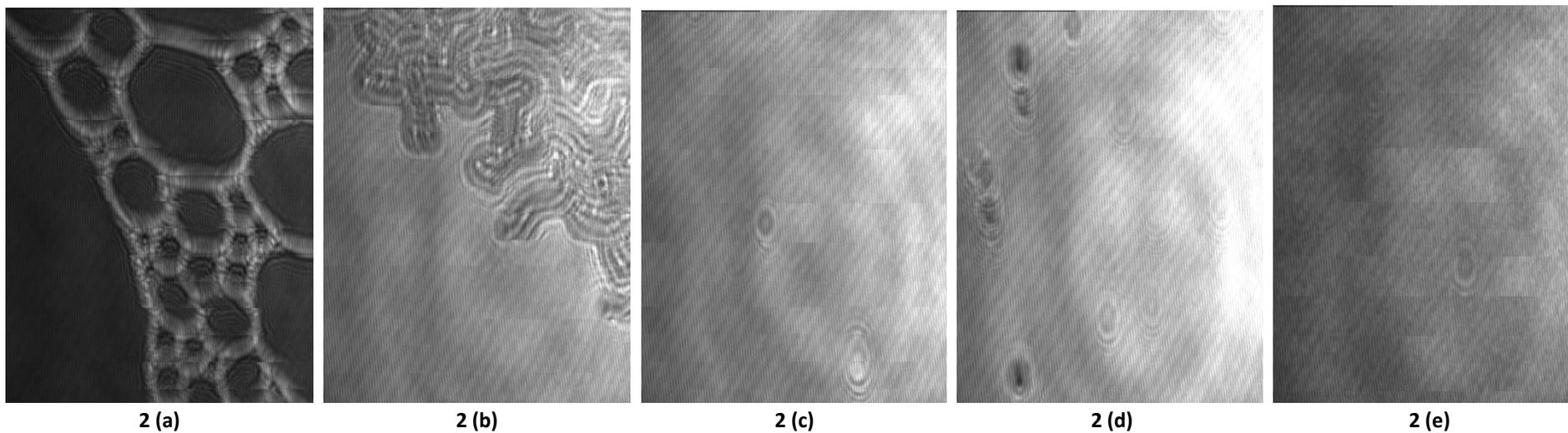


Fig. S3. Brewster angle microscopy images of [Ru(**1**)₂(bpy)][PF₆]₂ on a water subphase at (a) 0.001 mN/m, (b) 0.06 mN/m (c) 0.08 mN/m, (d) 1.87 mN/m and (e) 53.69 mN/m.

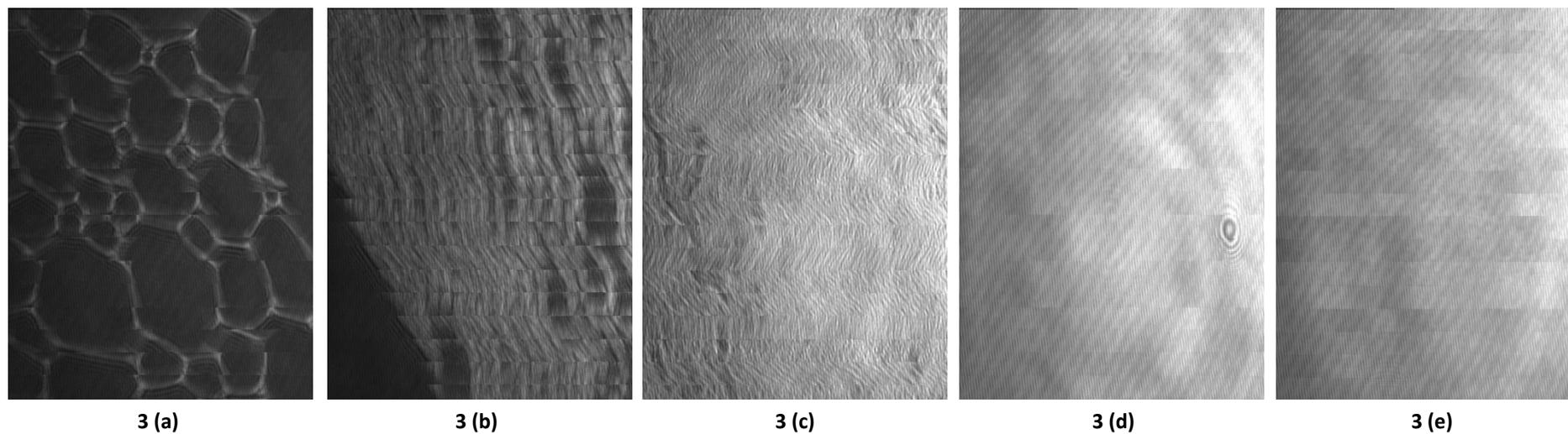


Fig. S4. Brewster angle microscopy images of *cis*-[Ru(**1**)₂Cl₂] on an aqueous KCl subphase at (a) 0.00 mN/m, (b) 1.27 mN/m (c) 5.33 mN/m, (d) 15.23 mN/m and (e) 26.90 mN/m.

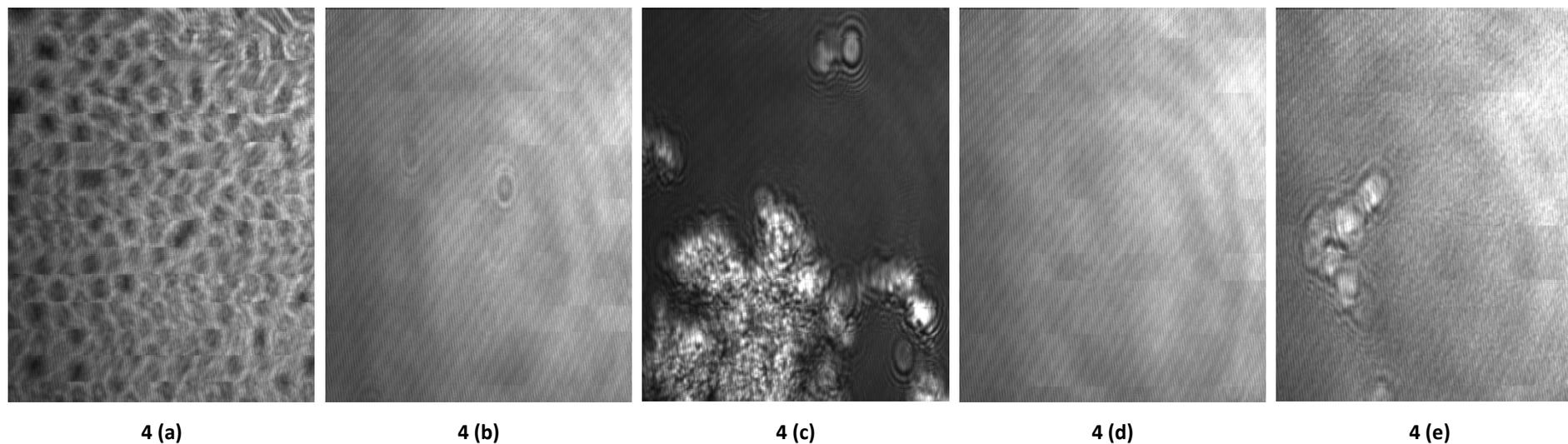


Fig. S5. Brewster angle microscopy images of [Ru(**1**)₂(bpy)][PF₆]₂ on an aqueous KPF₆ subphase at (a) 0.02 mN/m, (b) 1.08 mN/m (c) 8.20 mN/m, (d) 28.55 mN/m and (e) 51.42 mN/m.

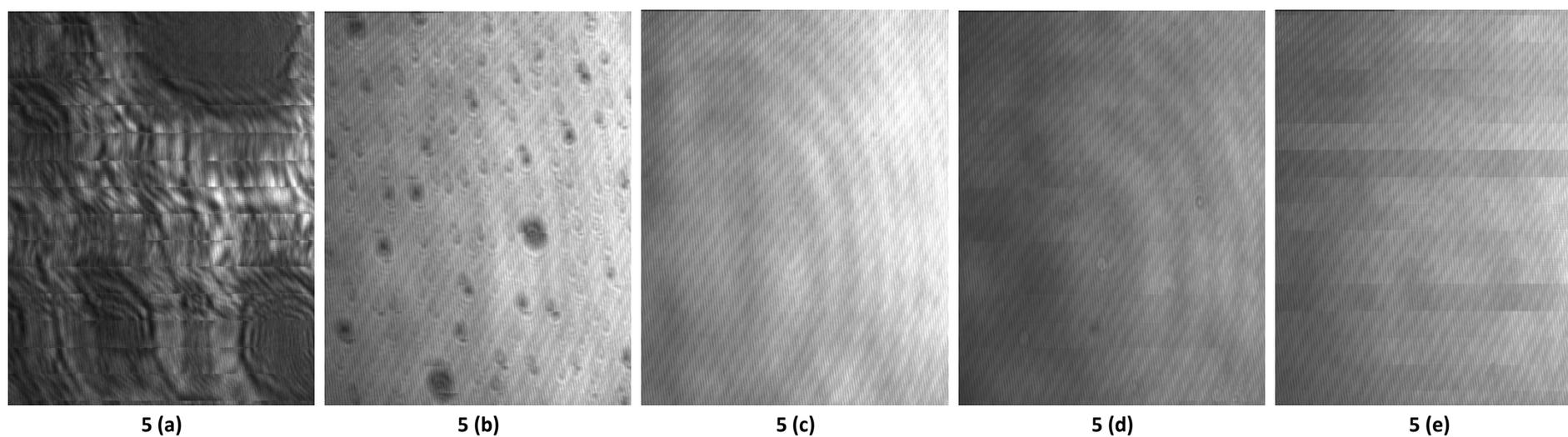


Fig. S6. Brewster angle microscopy images of cis-[Ru(**1**)₂Cl₂] on a Co₄POM subphase at (a) 0.00 mN/m, (b) 0.95 mN/m (c) 3.31 mN/m, (d) 11.04 mN/m and (e) 27.10 mN/m.

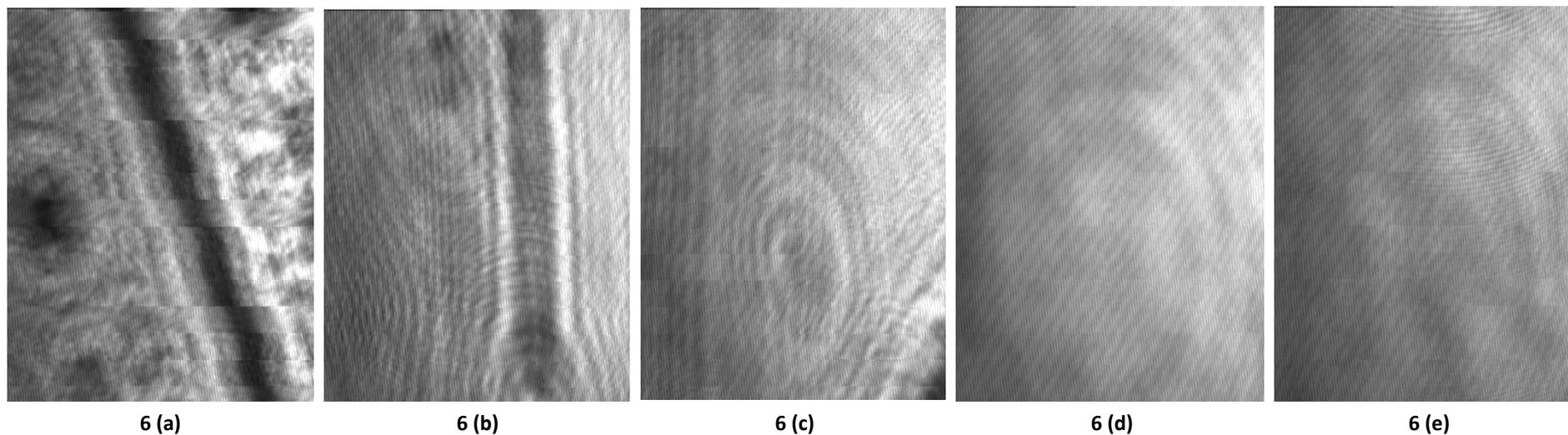


Fig. S7. Brewster angle microscopy images of [Ru(**1**)₂(bpy)][PF₆]₂ on a Co₄POM subphase at (a) 0.06 mN/m, (b) 0.17 mN/m (c) 2.00 mN/m, (d) 15.28 mN/m and (e) 21.34 mN/m.

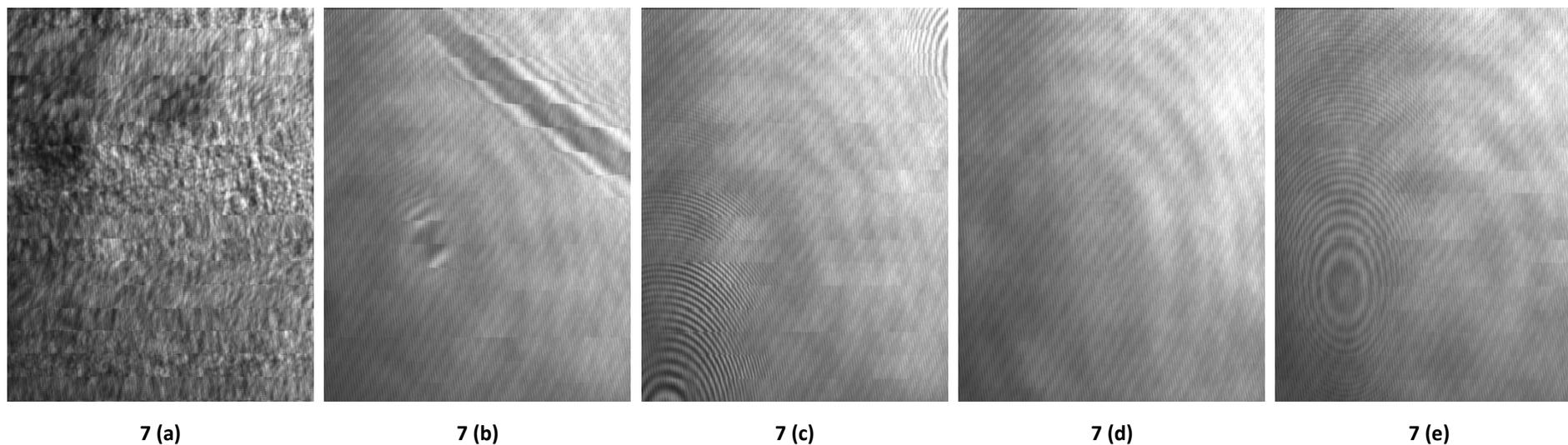


Fig. S8. Brewster angle microscopy images of $\text{Ru}(\mathbf{1})_2(\text{bpy})][\text{PF}_6]_2 : \text{DODA} (1:0.5)$ on a water subphase at (a) 0.06 mN/m, (b) 0.12 mN/m (c) 1.71 mN/m, (d) 25.27 mN/m and (e) 66.13 mN/m.

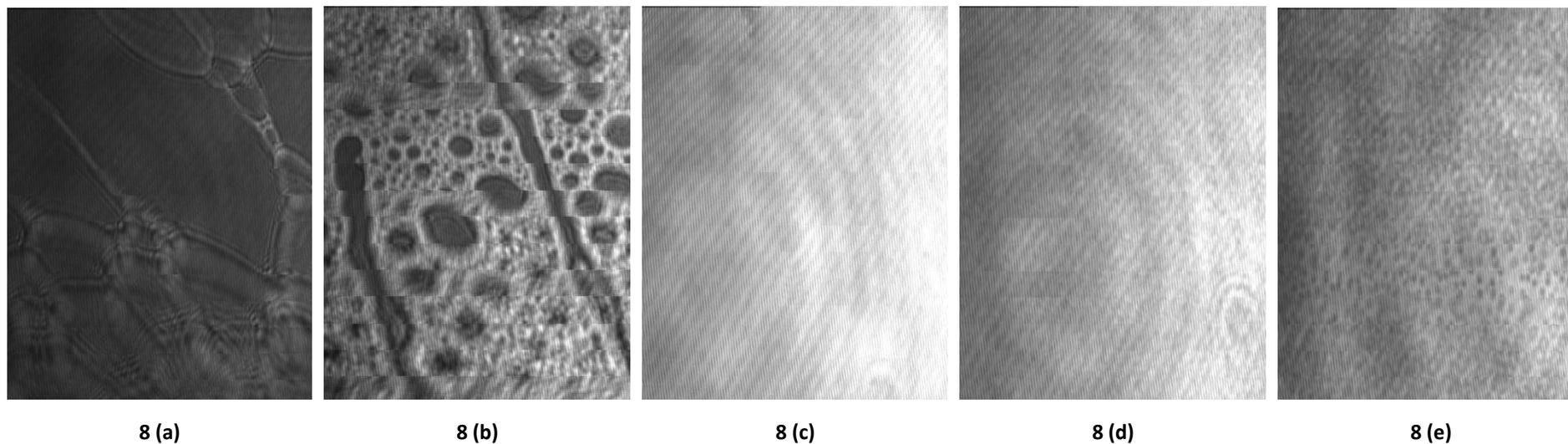


Fig. S9. Brewster angle microscopy images of $[\text{Ru}(\mathbf{1})_2(\text{bpy})][\text{PF}_6]_2 : \text{DODA} (1 : 0.5)$ on a Co_4POM subphase at (a) 0.04 mN/m, (b) 0.15 mN/m (c) 0.63 mN/m, (d) 2.65 mN/m and (e) 19.12 mN/m.

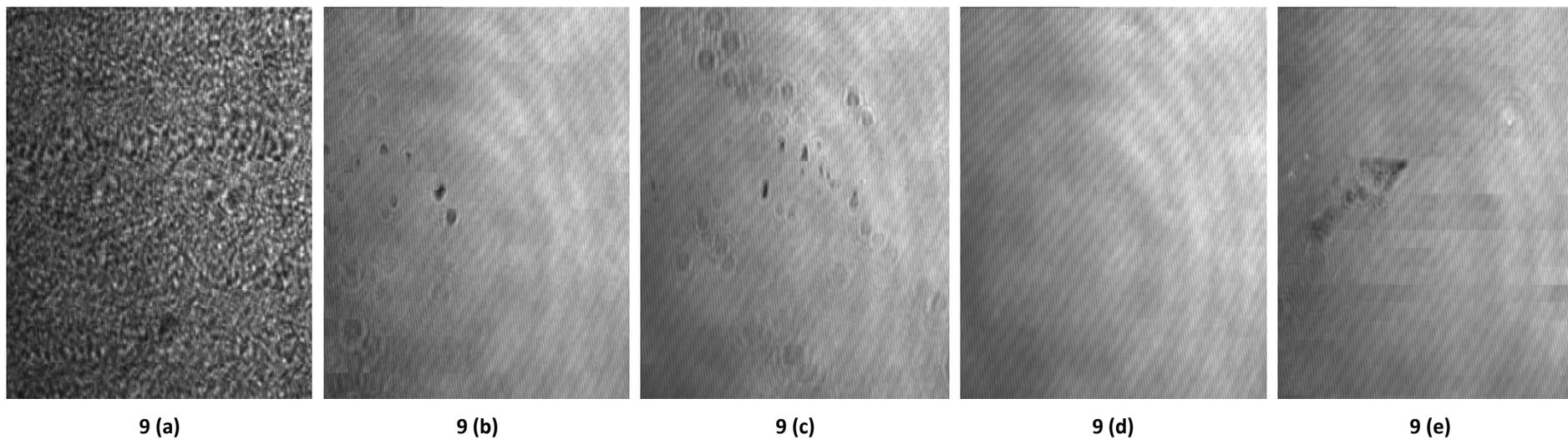


Fig. S10. Brewster angle microscopy images of $\text{Ru}(\mathbf{1})_2(\text{bpy})][\text{PF}_6]_2$:DODA (1:5) on a water subphase at (a) 0.11 mN/m, (b) 3.31 mN/m (c) 7.55 mN/m, (d) 31.54 mN/m and (e) 46.36 mN/m.

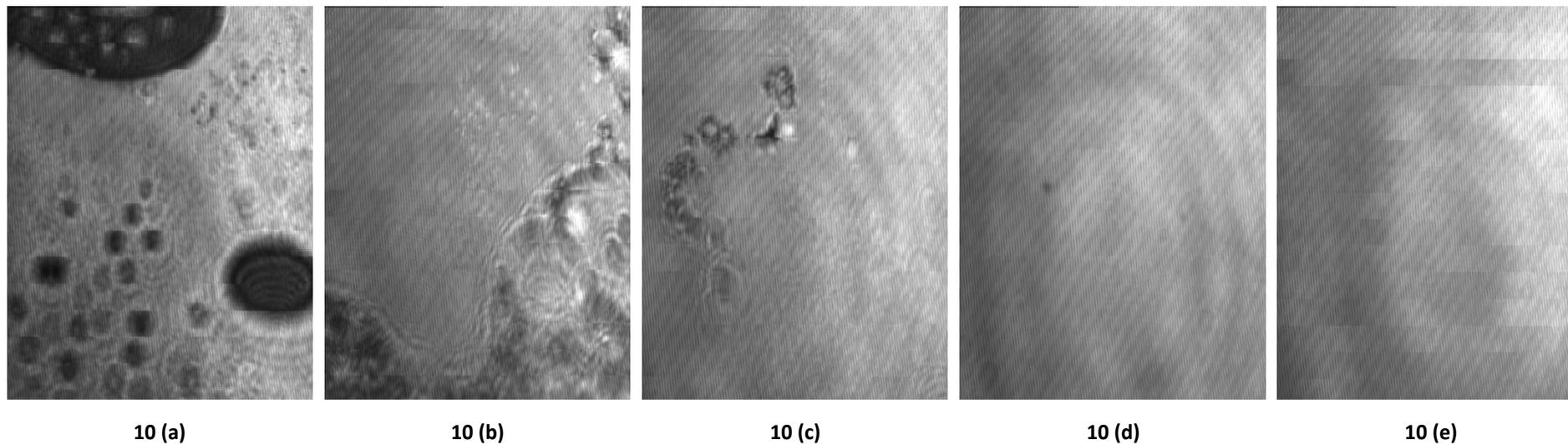


Fig. S11. Brewster angle microscopy images of $[\text{Ru}(\mathbf{1})_2(\text{bpy})][\text{PF}_6]_2$: DODA (1:20) on a Co_4POM subphase at (a) 0.05 mN/m, (b) 0.27 mN/m (c) 4.93 mN/m, (d) 52.08 mN/m and (e) 58.64 mN/m.

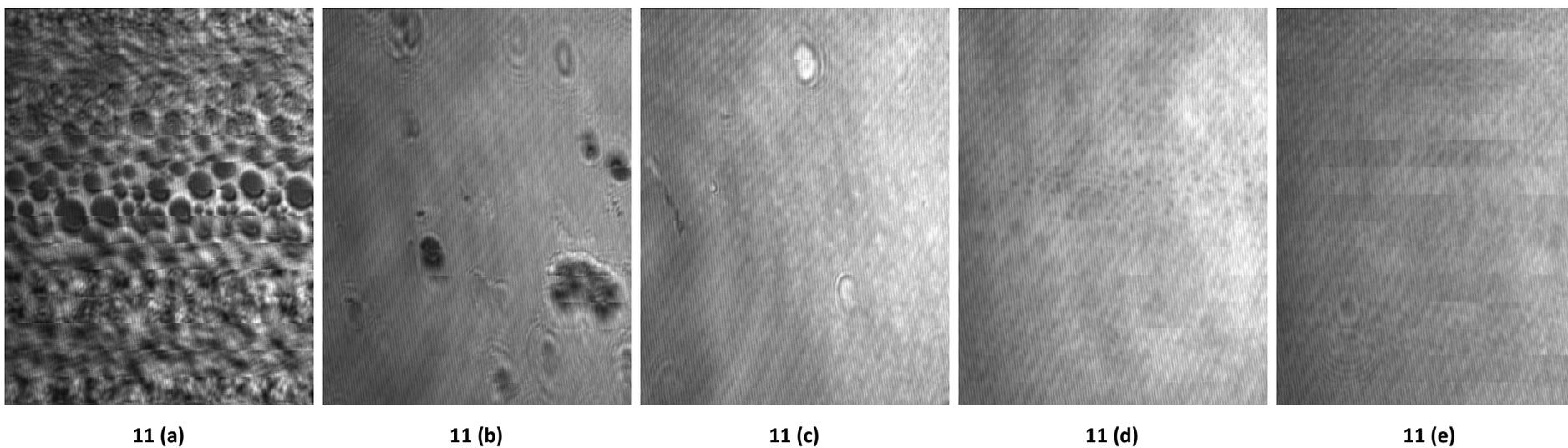
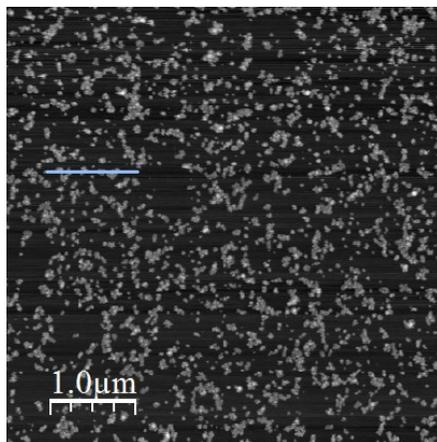
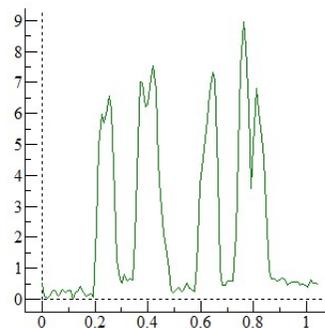


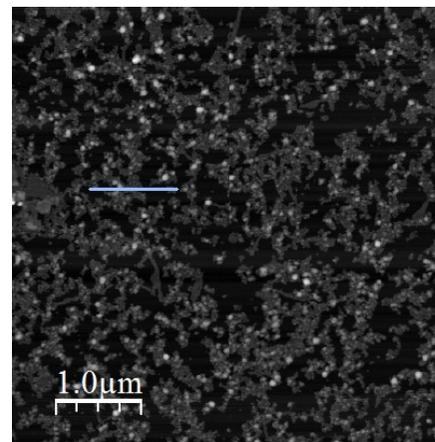
Fig. S12. AFM height images of *cis*-[Ru(1)₂Cl₂] LB film on mica, transferred from a monolayer formed on a Co₄POM subphase from (a) one dipping cycle, (b) line profile corresponding to the blue line drawn in (a), (c) three dipping cycles (d) Line profile corresponding to the blue line drawn in (c), and (e) five dipping cycles (f) Line profile corresponding to the blue line drawn in (e)



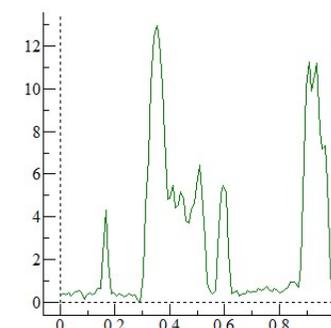
12 (a)



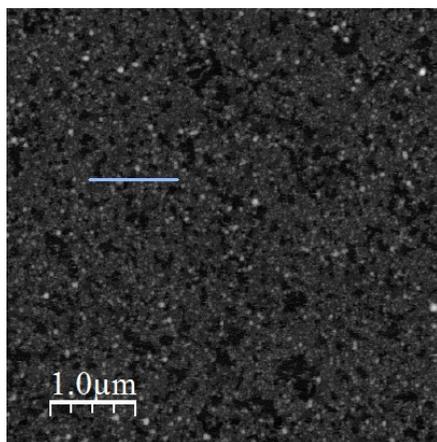
12 (b)



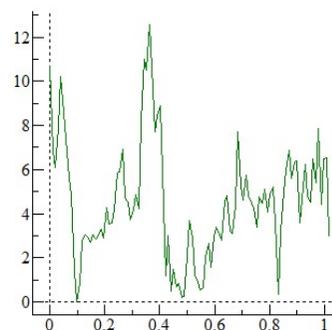
12 (c)



12 (d)

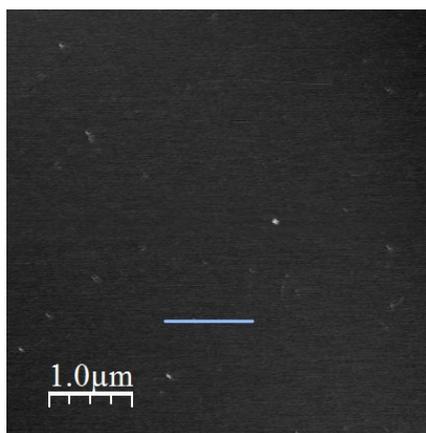


12 (e)

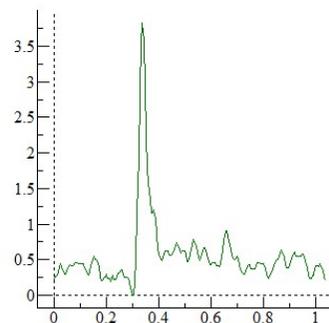


12 (f)

Fig. S13. AFM height images of $[\text{Ru}(\mathbf{1})_2(\text{bpy})][\text{PF}_6]_2$ LB film on mica, transferred from a monolayer formed on (a) a water subphase from one dipping cycle, (b) line profile corresponding to the blue line drawn in (a)

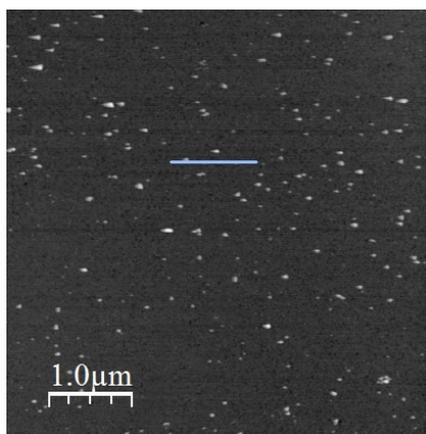


13 (a)

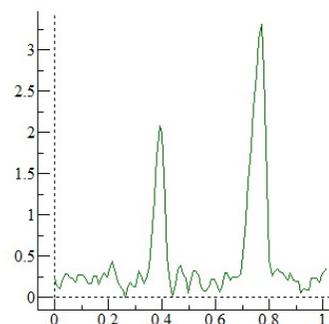


13 (b)

Fig S14. AFM height images of $[\text{Ru}(\mathbf{1})_2(\text{bpy})][\text{PF}_6]_2$ LB film on mica, transferred from a monolayer formed on a Co_4POM subphase from (a) five dipping cycles, with (b) Line profile corresponding to the blue line drawn in (a).

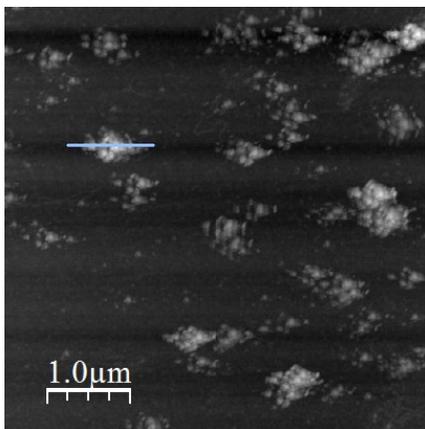


14 (a)

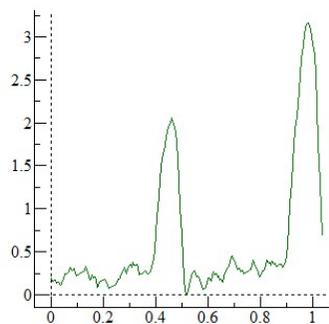


14 (b)

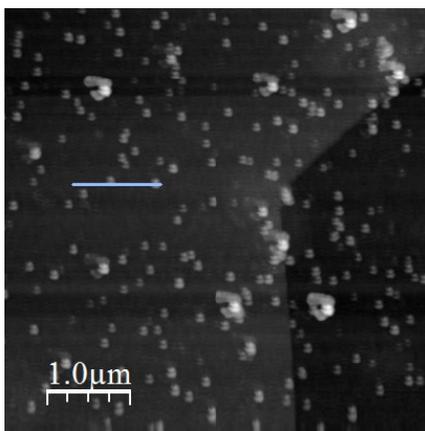
Fig S15. AFM height images of $[\text{Ru}(\mathbf{1})_2(\text{bpy})][\text{PF}_6]_2/\text{DODA}$ LB films (five dipping cycles) on mica transferred from monolayers formed on a Co_4POM subphase: (a) 1 : 5 $[\text{Ru}(\mathbf{1})_2(\text{bpy})][\text{PF}_6]_2$: DODA mole fraction, (b) Line profile corresponding to the blue line drawn in (a), (c) 1 : 20 $[\text{Ru}(\mathbf{1})_2(\text{bpy})][\text{PF}_6]_2$: DODA mole fraction, (d) Line profile corresponding to the blue line drawn in (c).



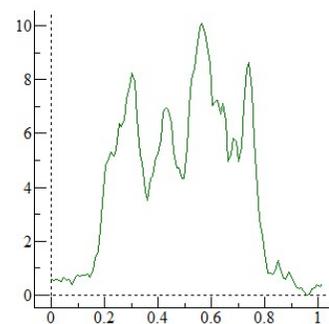
15 (a)



15 (b)



15 (c)



15 (d)