Supporting Information

Controlling Growth of Self-Propagating Molecular Assemblies

Leila Motiei,[a] Moran Feller,[a] Guennadi Evmenenko,[b] Pulak Dutta[b] and
Milko E. van der Boom*[a]

[a] *Department of Organic Chemistry, The Weizmann Institute of Science, Rehovot, Israel.
Fax: +972 8934 4142; Tel: +972 8934 2515; E-mail:milko.vanderboom@weizmann.ac.il*
[b] *Department of Physics and Astronomy, Northwestern University, Evanston, IL 60208-
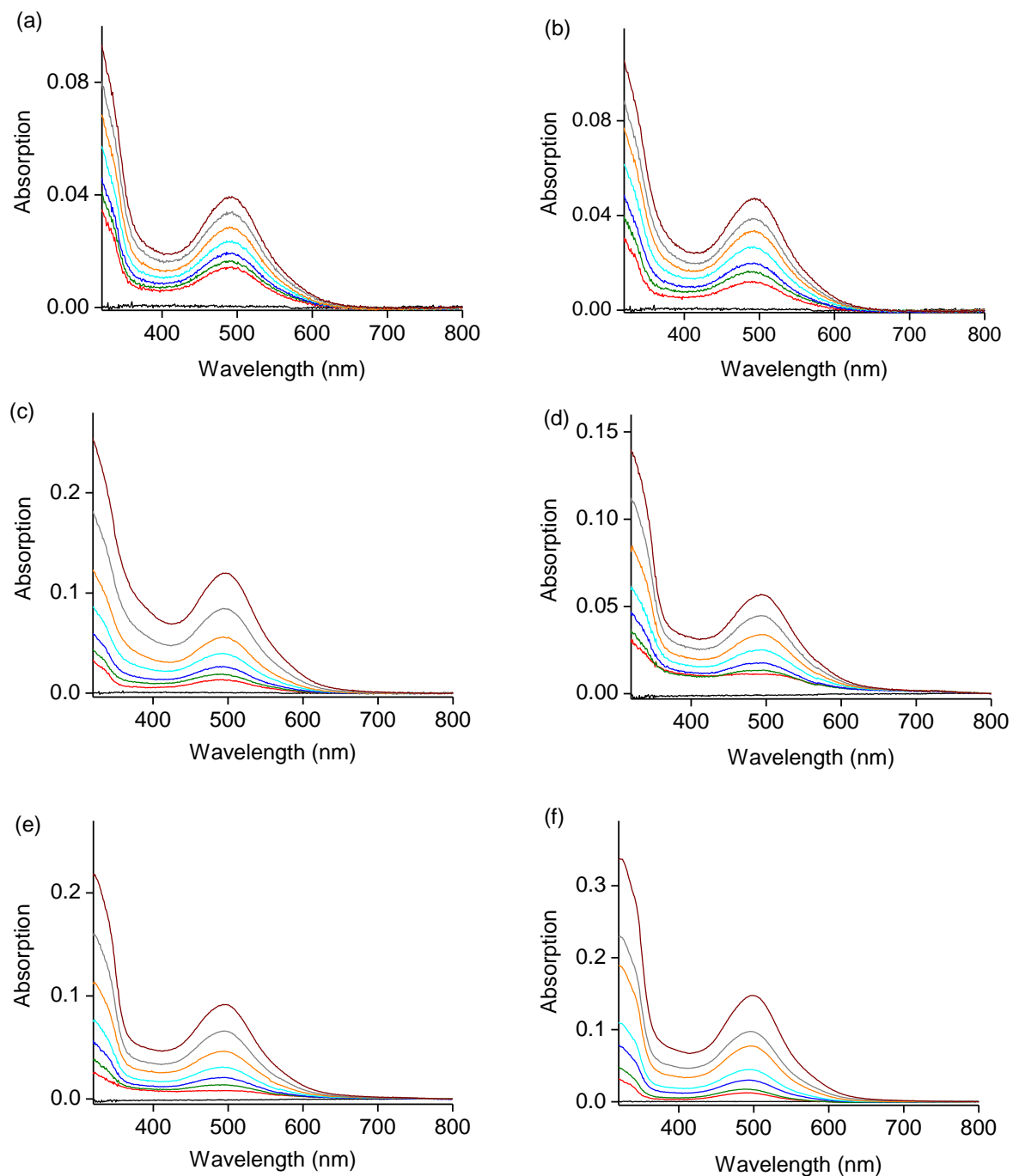3113, USA.*

Fig. S1 Representative transmission UV/Vis absorption spectra of the 6 SPMAs on glass substrates measured after each deposition step of complex **11** (Scheme 1). The spectra with the lowest absorption intensities are from the template layer (red line; TL, Scheme 2a) and the baseline (black line). (a) SPMA|Pt(PhCN)$_2$Cl$_2$, (b) SPMA|(PhCN)$_2$PtBr$_2$, (c) SPMA|(PhCN)$_2$PtI$_2$, (d) SPMA|(PhCN)$_2$Pd(SMe$_2$)$_2$Cl$_2$, (e) SPMA|Pd(COD)Cl$_2$, and (f) SPMA|Pd(PhCN)$_2$Cl$_2$. The spectra of the latter SPMA have been included in the main text as Fig. 1a.
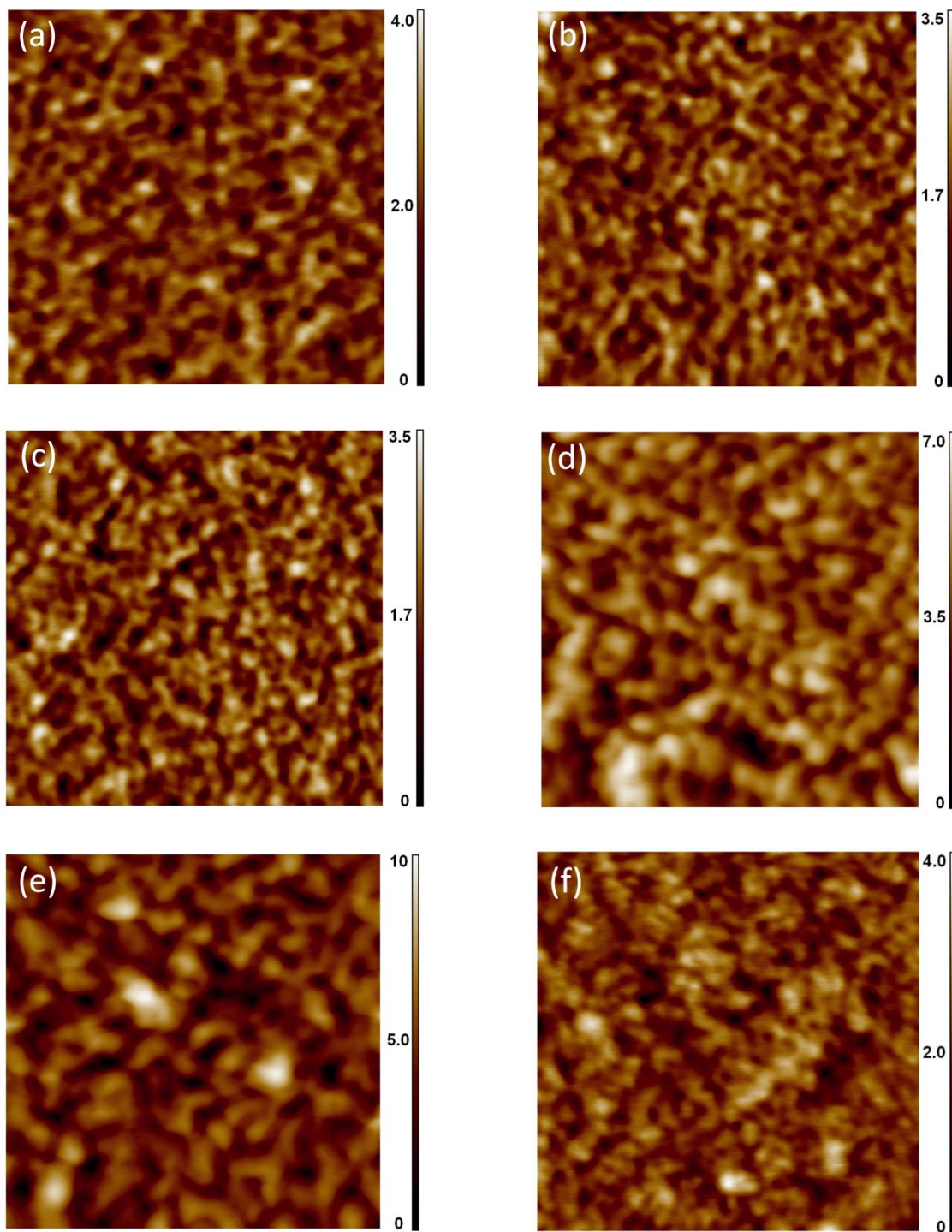
Fig. S2 Semicontact AFM images (500 nm × 500 nm scan areas) of (a) SPMA|Pt(PhCN)$_2$Cl$_2$ (R_{rms} = 0.4 nm), (b) SPMA|Pt(PhCN)$_2$Br$_2$ (R_{rms} = 0.5 nm), (c) SPMA|Pt(PhCN)$_2$I$_2$ (R_{rms} = 0.4 nm), and (d) SPMA|Pd(SMe$_2$)$_2$Cl$_2$ (R_{rms} = 1.1 nm), (e) SPMA|Pd(COD)Cl$_2$ (R_{rms} = 1.0 nm), and (f) SPMA|Pd(PhCN)$_2$Cl$_2$ (R_{rms} = 0.4 nm) after 13 deposition steps on silicon substrates. Deposition step 1 is the template layer (TL) (Scheme 2a). The values shown in the vertical scales are in nm. The image of SPMA|Pd(PhCN)$_2$Cl$_2$ (f) has been included in the main text as Fig. 2.
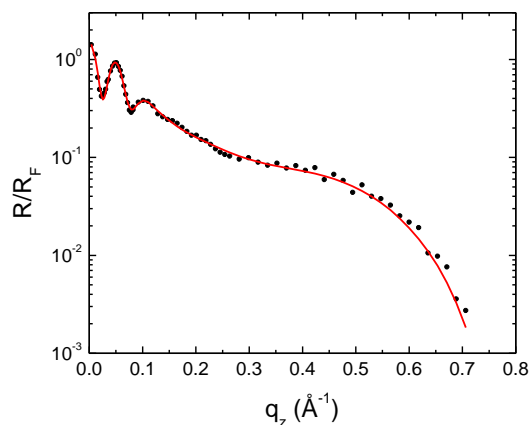
Fig. S3 Representative synchrotron X-ray reflectivity (XRR) spectrum of SPMA|Pd(COD)Cl$_2$ on a silicon substrate after 13 deposition steps. Deposition step 1 is the template layer (TL) (Scheme 2a). The red trace is a fit of the experimental data (black dots). For details regarding XRR and data fitting, see: G. Evmenenko, M. E. van der Boom, S. W. Dugan, J. Kmetko, T. J. Marks and P. Dutta, *J. Chem. Phys.,* 2001, **115**, 6722.

Table S1 Slopes derived by linear fitting of the light absorption intensity at $\lambda \approx 465$ nm versus the SPMA thickness (Fig. 5; $R^2 \geq 0.98$).

entry	M	L	X	Slope (x 10^{-4})
1	Pt	PhCN	Cl	6.4
2	Pt	PhCN	Br	7.1
3	Pt	PhCN	I	6.8
4	Pd	PhCN	Cl	8.1
5	Pd	½ COD	Cl	10
6	Pd	SMe$_2$	Cl	9.0

Table S2 The estimated molecular density of complex **11** for the SPMAs in molecules/nm^3. Deposition step 1 is the Template Layer (TL) (Scheme 2a).

Deposition step	Pt(PhCN)$_2$Cl$_2$	Pt(PhCN)$_2$Br$_2$	Pt(PhCN)$_2$I$_2$	Pd(PhCN)$_2$Cl$_2$	Pd(SMe)$_2$Cl$_2$	Pd(COD)Cl$_2$
1	1.4	1.0	1.4	1.1	1.1	0.6
3	1.3	1.1	1.1	1.0	0.9	0.8
5	1.2	1.0	1.1	1.2	0.9	0.9
7	1.2	1.1	1.1	1.3	1.1	0.9
9	1.1	1.1	1.0	1.1	1.1	1.0
11	1.1	1.1	1.0	1.2	1.2	1.1
13	1.1	1.1	1.1	1.2	1.3	1.2