Electronic Supplementary Material (ESI) for Faraday Discussions.

Crystal growth of the core and rotated epitaxial shell of a heterometallic metal-organic framework revealed with atomic force microscopy

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Electronic Supporting Information

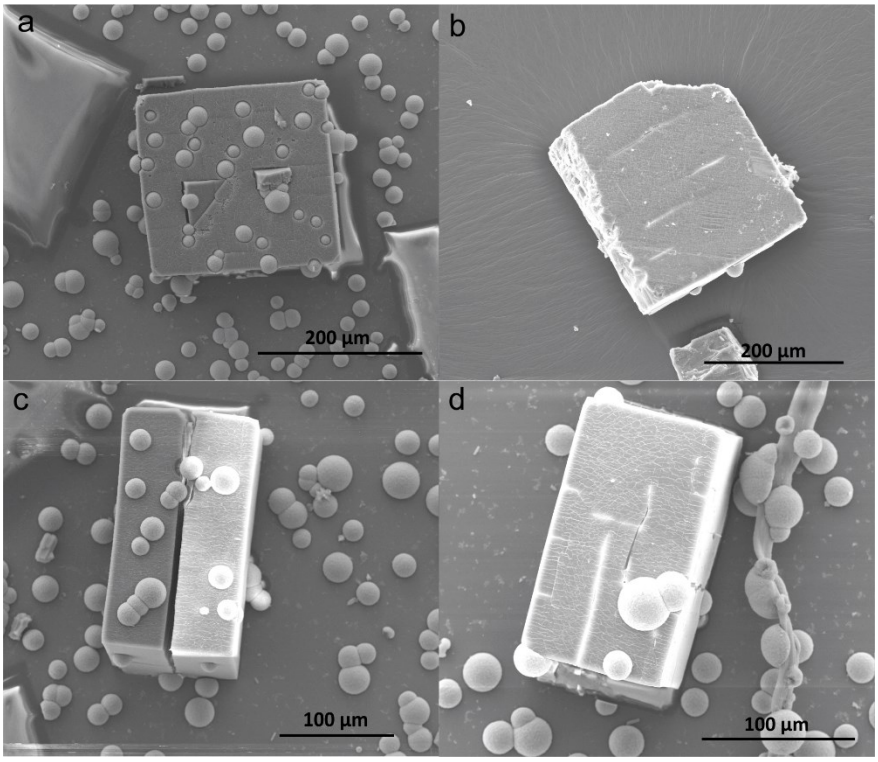

Figure S1. Scanning electron micrographs of crystal of **1** showing both the {001} (a, b) and {100} (c, d) facets.

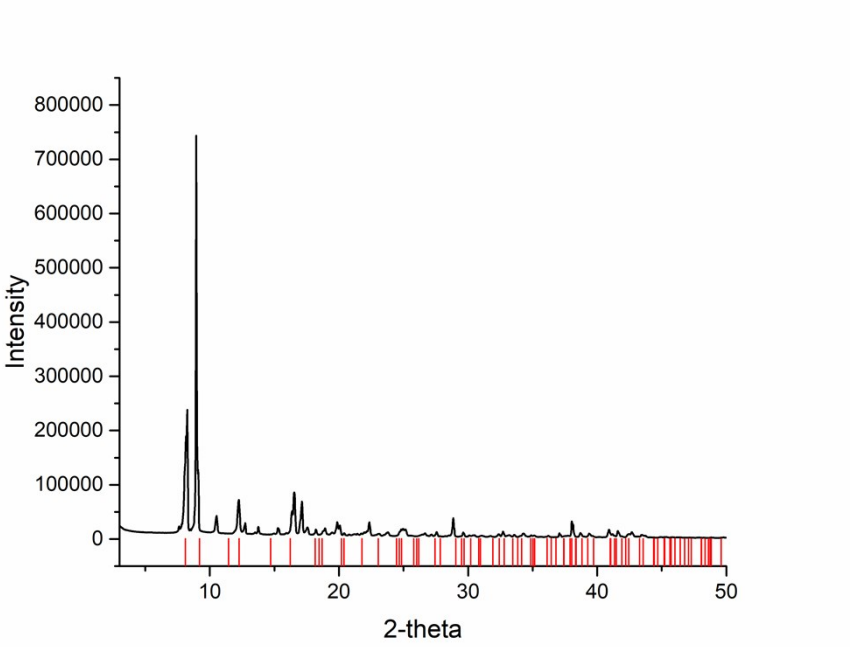

Figure S2. PXRD pattern of the crystalline product formed from the synthesis of **1**. The experimental diffraction pattern is shown as the black line, and the red tick marks represent the calculated reflection positions for **1** (tetragonal, P4/mmm, a = b = 10.9212(6) Å, c = 9.6108(7) Å, α = β = γ = 90°).[1]

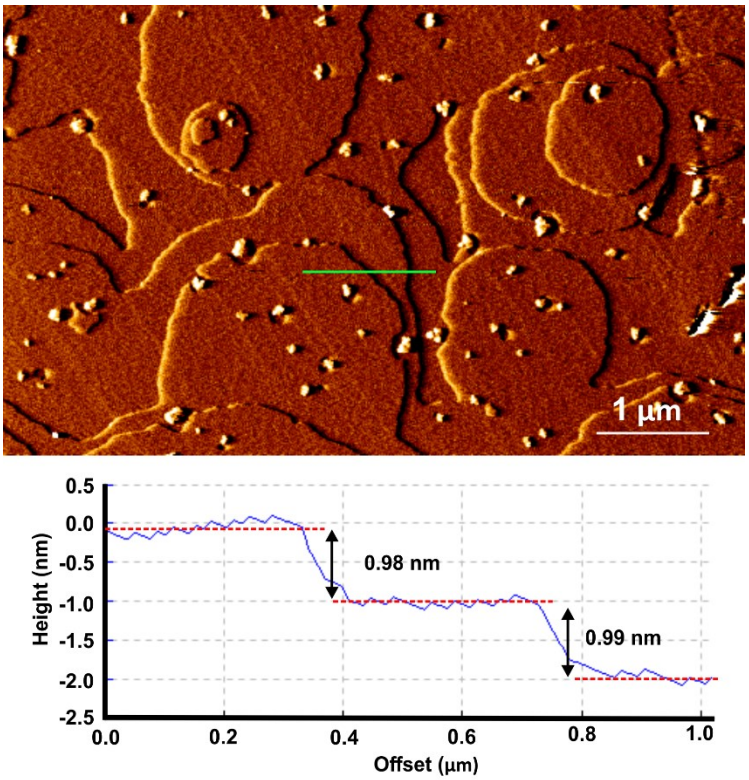

Figure S3. AFM deflection image of **1** with cross-section analysis (green line) showing the step heights that correspond to the d_{001}-lattice spacing. AFM image size is 6.5 x 4.0 µm^2.

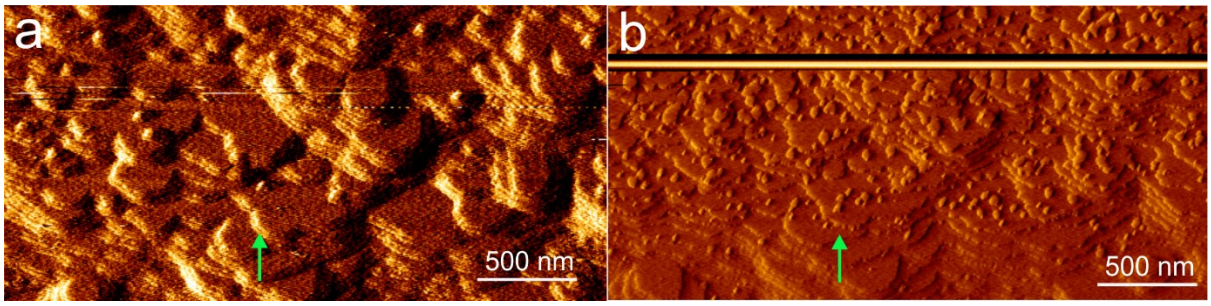

Figure S4. AFM deflection images of **1** under DMF (a) and after 6.4 min of growth solution **2** injection (b). Both AFM images cover the same area. The terrace shown with the green arrow is similar to the terrace shown in the green arrow in b. However, many additional 2D nuclei of **2** are present over the terraces in b. AFM images size are 3.0 x 1.5 μm^2.

Table S1. The values of the measured in-plane rotational epitaxy angles between the diagonals of several surface square growth layers, mounds and spirals and the edges of the core-shell **1@2** crystals.

Growth island	Angle (°)
1	6.03
2	5.09
3*	6.76
4	5.73
5	5.19
6	5.77
7	6.90
8	6.32
9	5.38
10	5.72
11	5.79
12	5.93
13	5.32
14	6.68
Average	5.9
Std. deviation	0.7

*Angle measured for the growth island highlighted in Figure 5a on the crystal shown in Figure 5d.

References

1 S. Furukawa, K. Hirai, K. Nakagawa, Y. Takashima, R. Matsuda, T. Tsuruoka, M. Kondo, R. Haruki, D. Tanaka, H. Sakamoto, S. Shimomura, O. Sakata, S. Kitagawa, Angew. Chem. Int. Ed. 2009, **48**, 1766.