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Supporting Information (SI)

Orientation Control and Crystallization in a Soft Confined Phase Separated Block Copolymer

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SI 1: Crystallization Induced by AFM Tip

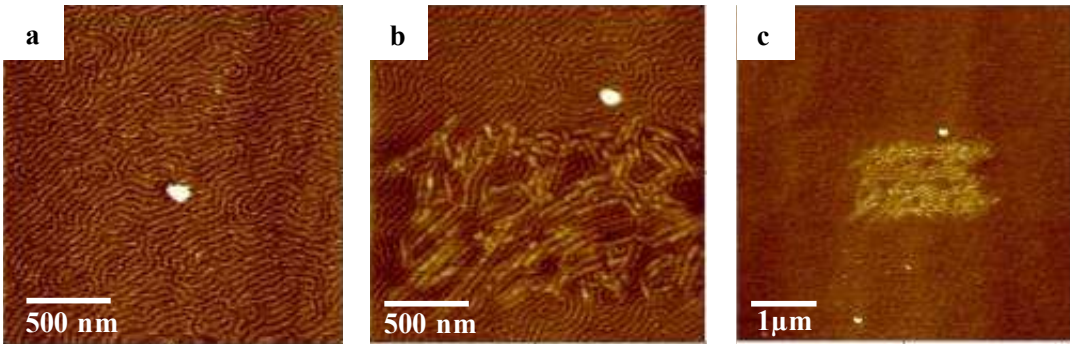


Figure S1: AFM phase images showing the crystallization induced by the AFM tip during the aligning of the cylinders at 110 °C: **(a)** before dropping the amplitude setpoint (i.e. increasing the tapping force) and **(b, c)** after that, when the crystallization was induced.

SI 2: Reducing the Number of Defects

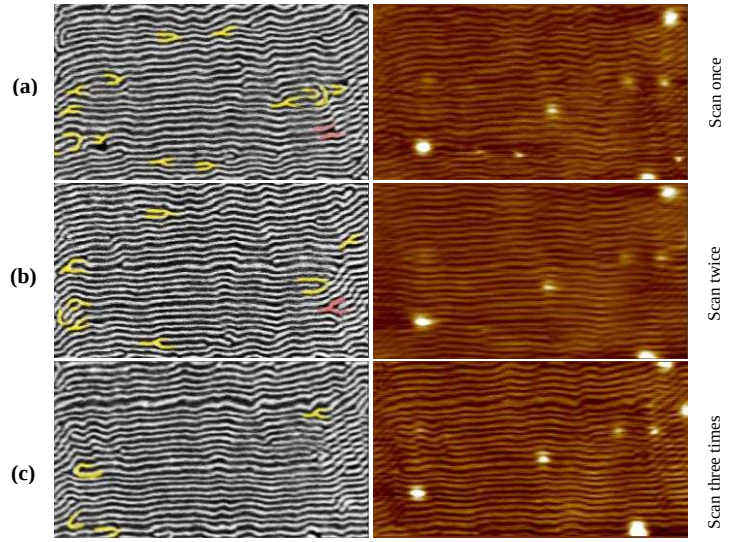


Figure S2: AFM phase (the left column) and height (the right column) images showing the effect of the number of orienting scans on the defects per unit area, **(a)-(c)** 1st, 2nd, and 3rd scan, respectively with relatively high tapping force (lower amplitude and lower $r_{sp} \approx 0.1$) at 115 ºC. The defects are highlighted to aid the reader to follow the reduction as a function of the number of scans. The region highlighted in red shows an example of producing a new defect with the same scanning conditions but where the overall number of defects is still reduced. The image size is 3×1.5 µm and the black to white scale 12º.

SI 3: Crystal Morphology as a Function of Temperature

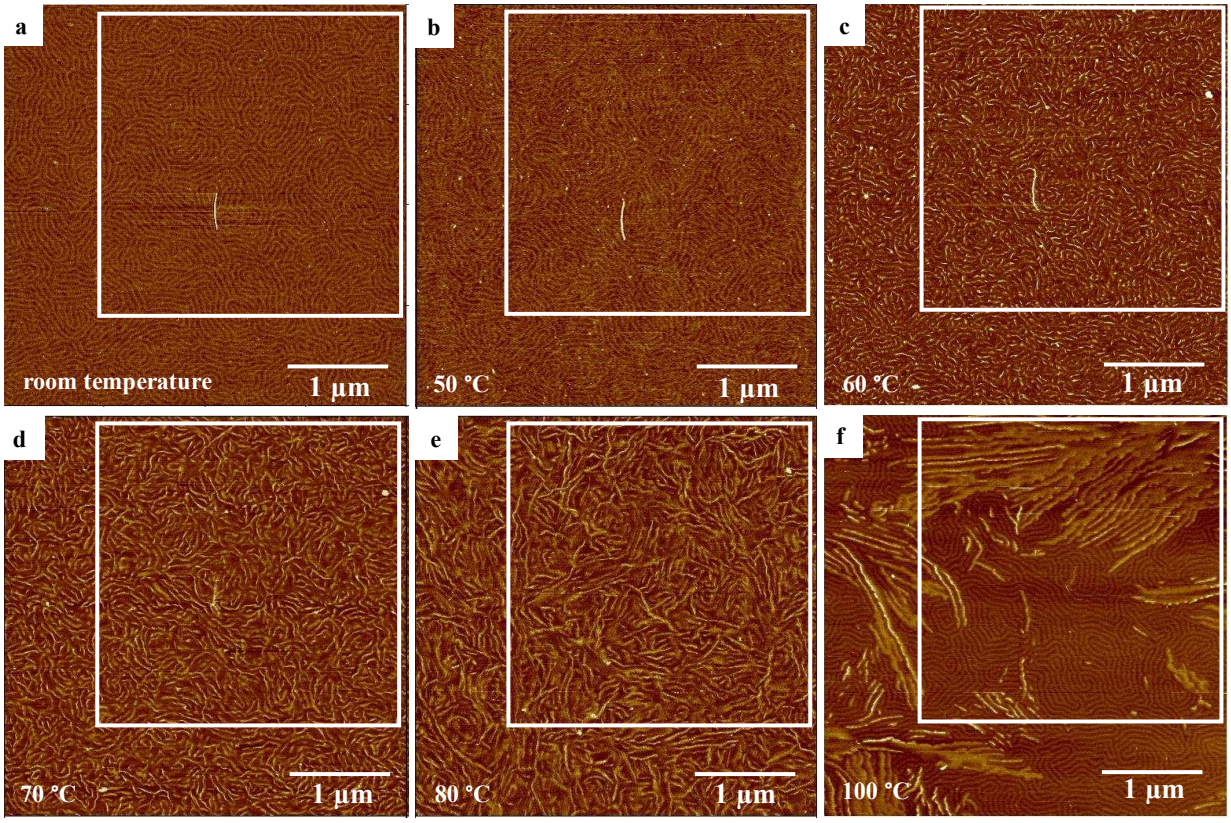


Figure S3: AFM phase images of crystallized E/MB after isothermal crystallization (the same area) at: (**a**) room temperature, (**b**) 50 °C, (**c**) 60 °C, (**d**) 70 °C, (**e**) 80 °C and (**f**) 100 °C (unoriented cylinders). The white squares illustrate the magnified area in **Figure 5** in the main text.

Another example of morphology formation from unoriented cylindrical microdomains (random) is shown in Figure S4.

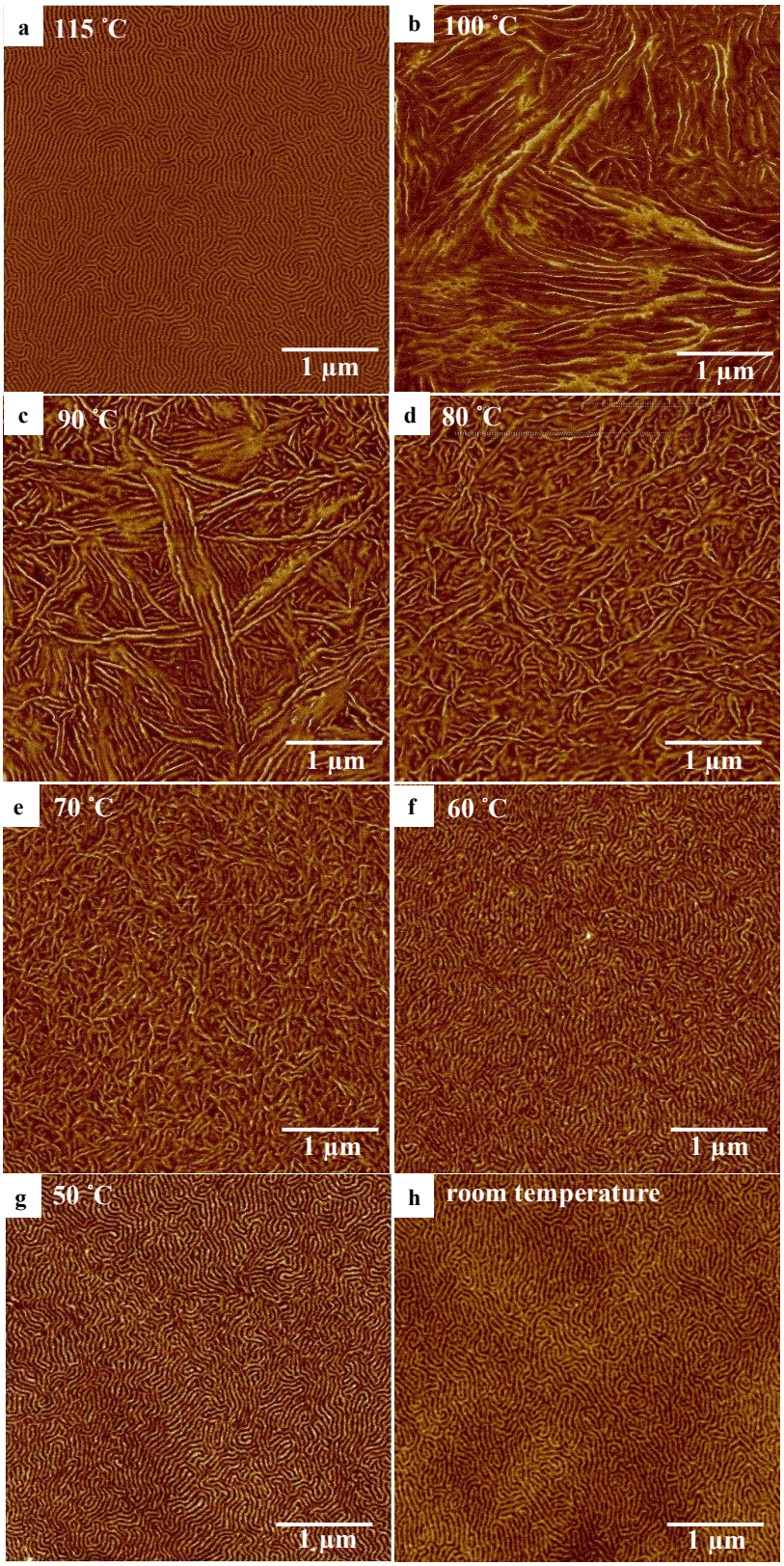


Figure S4: AFM phase image of (**a**) melt phase E/MB at 115 °C and (**b-h**) crystallized E/MB after isothermal crystallization at (**b**) 100 °C, (**c**) 90 °C, (**d**) 80 °C, (**e**) 70 °C, (**f**) 60 °C, (**g**) 50 °C and (**h**) room temperature (unoriented cylinders).

SI 4: Following Crystallization in Pre-Oriented Domains in Situ

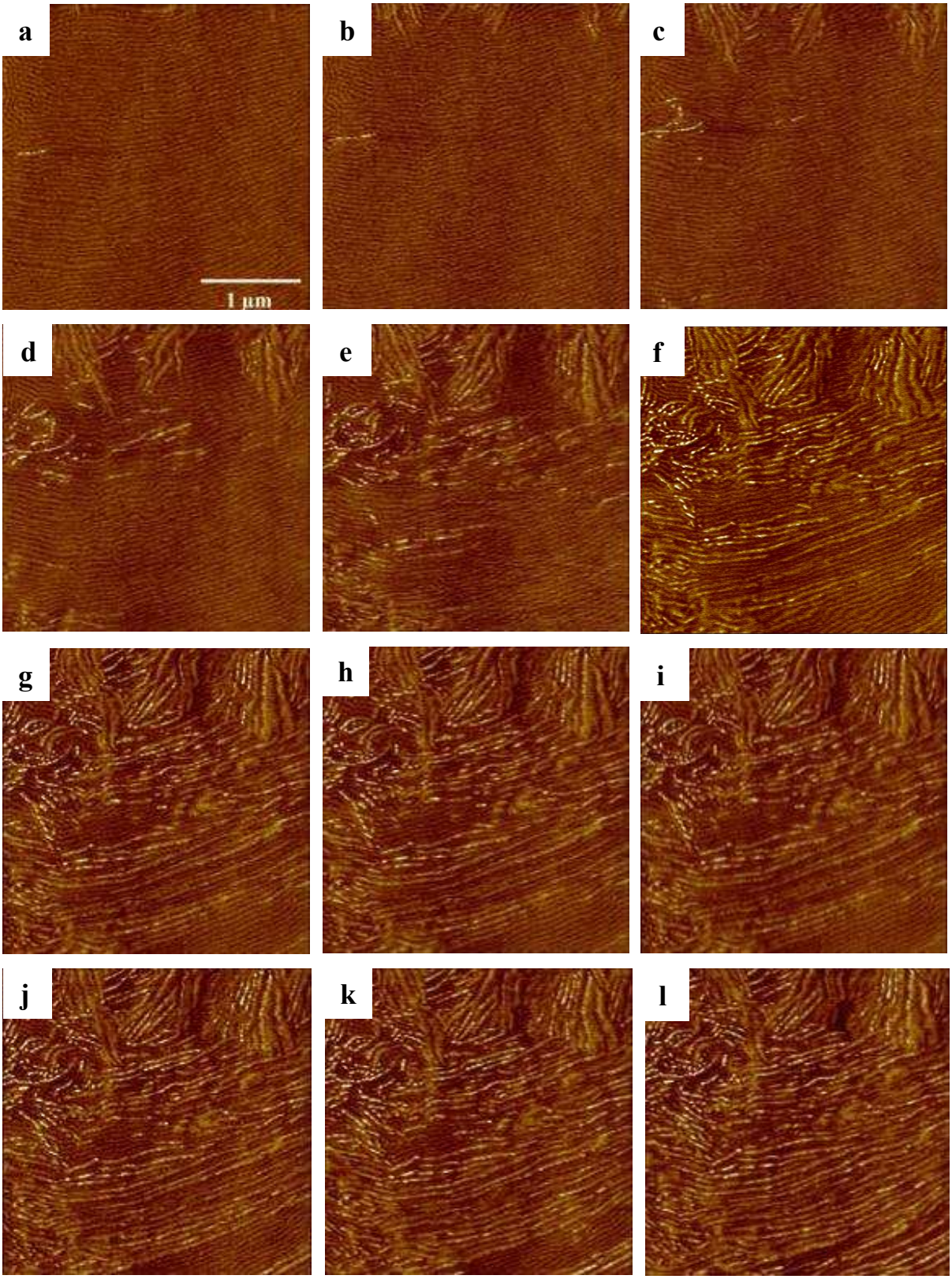


Figure S5: AFM phase images showing the crystallization in E/MB from the aligned melt structure at 99 °C, taken at **(a)** 0 s, **(b)** 149 s, **(c)** 298 s, **(d)** 596 s, **(e)** 894 s, **(f)** 1192 s, **(g)** 1341 s, **(h)** 1490 s, **(i)** 1639 s, **(j)** 1788 s, **(k)** 2086 and **(l)** 2980 s. Dark to bright represents a variation in phase of 7°.

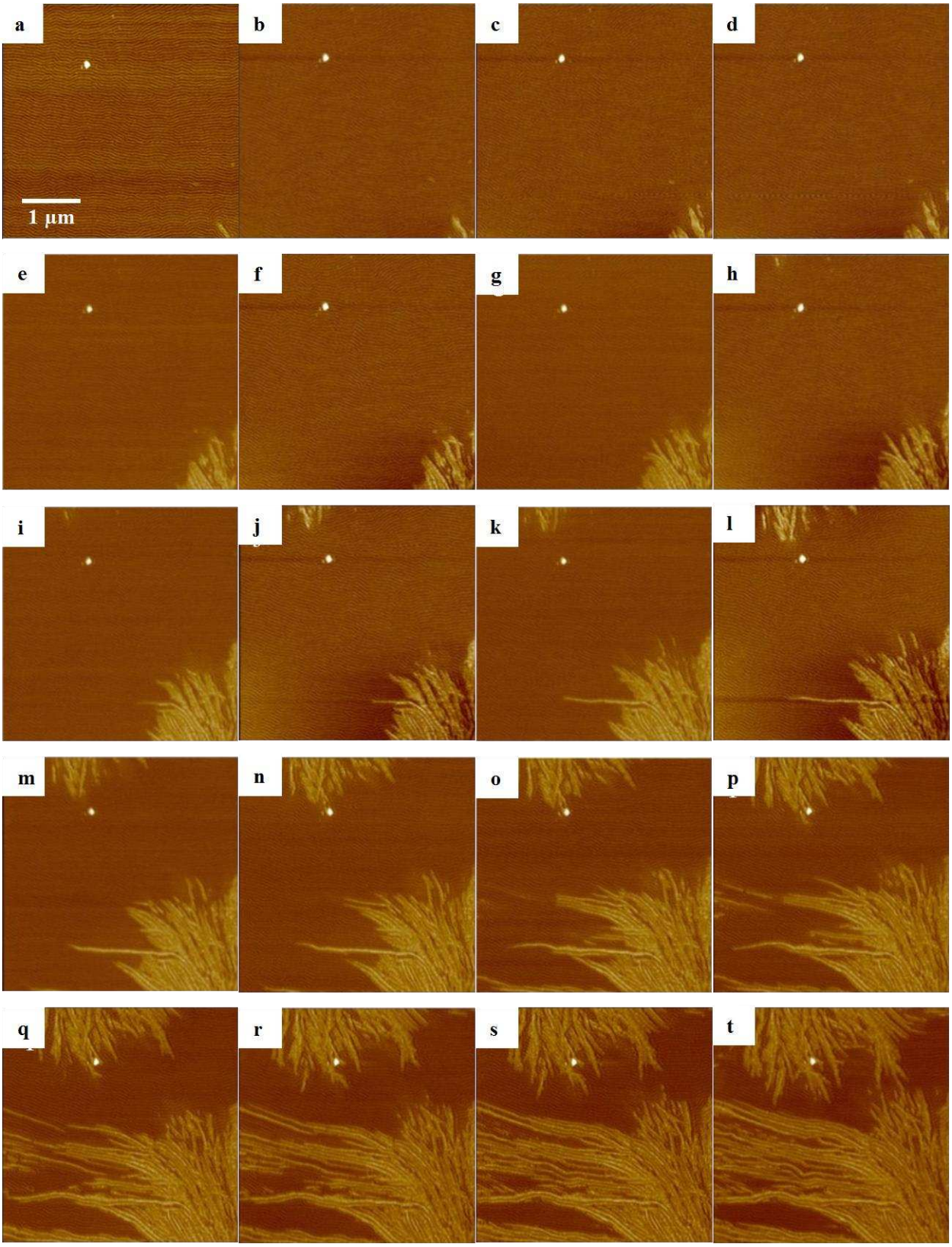
SI 5: Growth Rates & Ratio of Parallel and Perpendicular Crystals

The measurement of the difference in the growth rates of the crystals growing parallel to the cylinders and perpendicular to the cylinders is summarized in the below table (See the corresponding graphs of these values in the main text)**.**

Table S1: The ratios and overall average growth rates of a number of crystals growing along the cylinder axis and the growth rate perpendicular to the cylinder axis at temperatures of 97 °C–100 °C.

Crystallization temperature (°C)	**Average growth rate (nms^{-1})**		**Ratio of growth rates**
	Parallel to cylinders	Perpendicular to cylinders	
100	0.916	0.19	4.8
99	1.46	0.28	5.2
98	2.1	0.35	6
97	3.4	0.425	8

SI 6: The Transition from ‘Breakout’ to ‘Templated’ Growth



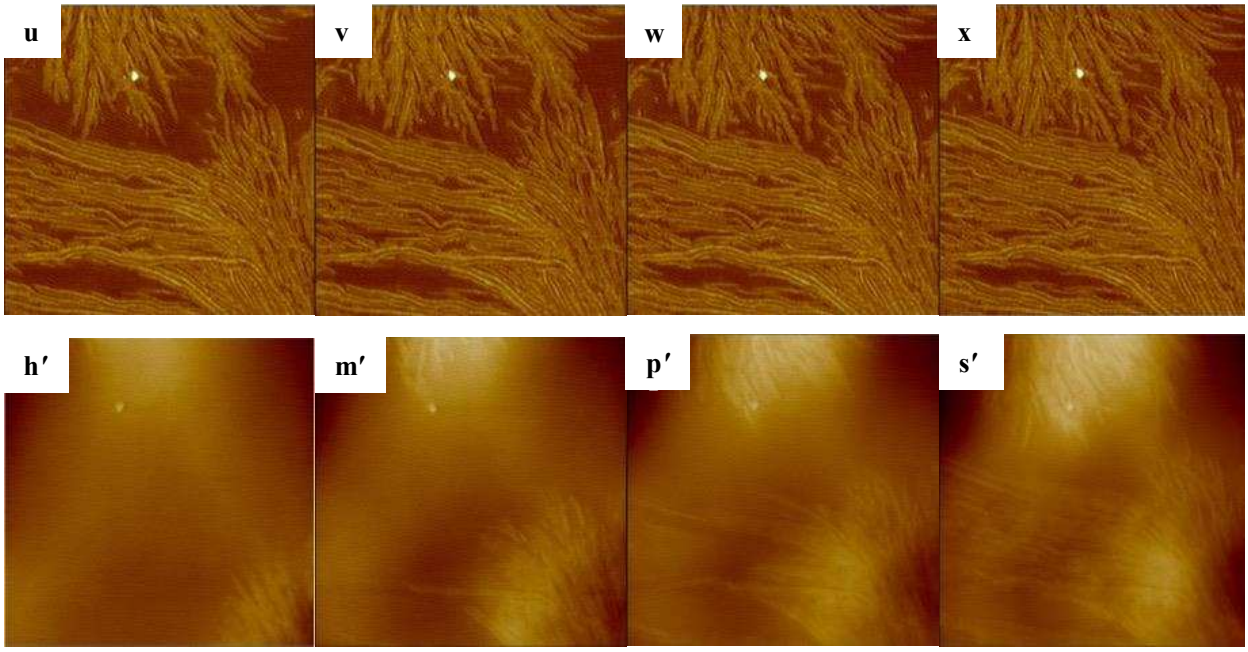


Figure S6: A full series of AFM phase images showing E/MB crystallization at 101 °C. The last bottom row (images **h′**, **m′**, **p′**, and **s′**) are the corresponding height images of **h**, **m**, **p**, and **s** image. Each image was captured in 128 s. Dark-bright represents a variation in phase of 25^{o} and the height scale is 120 nm.