

Electronic Supplementary Information (ESI)

for

**Organic Ionic Electro-Optic Crystals Grown by Specific
Interactions on Templates for THz Wave Photonics**

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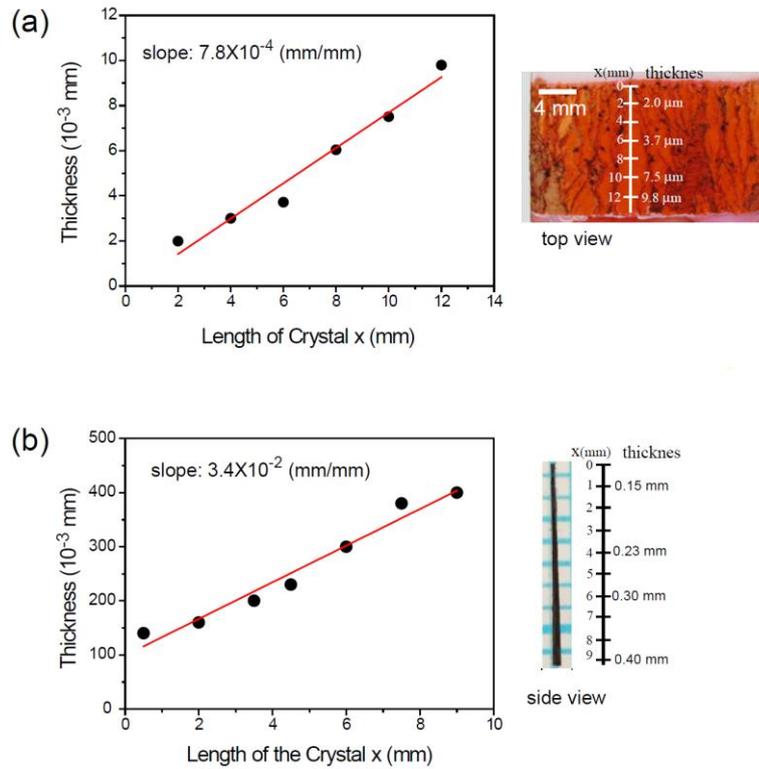


Figure S1. The crystal thickness of as-grown DSTMS crystals with (a) 22 % of the saturated concentration at 30 °C, for which the evaporation temperature is 30 °C and (b) 80 % of the saturated concentration at 40 °C, for which the evaporation temperature is 40 °C. The photograph of Figure S1b shows the side view of the crystal shown in Figure 2b. The slope of the thickness variation with respect to the length of the crystal is 7.8×10^{-4} and 3.4×10^{-2} mm-thickness/mm-length for Figure S1a and S1b, respectively.

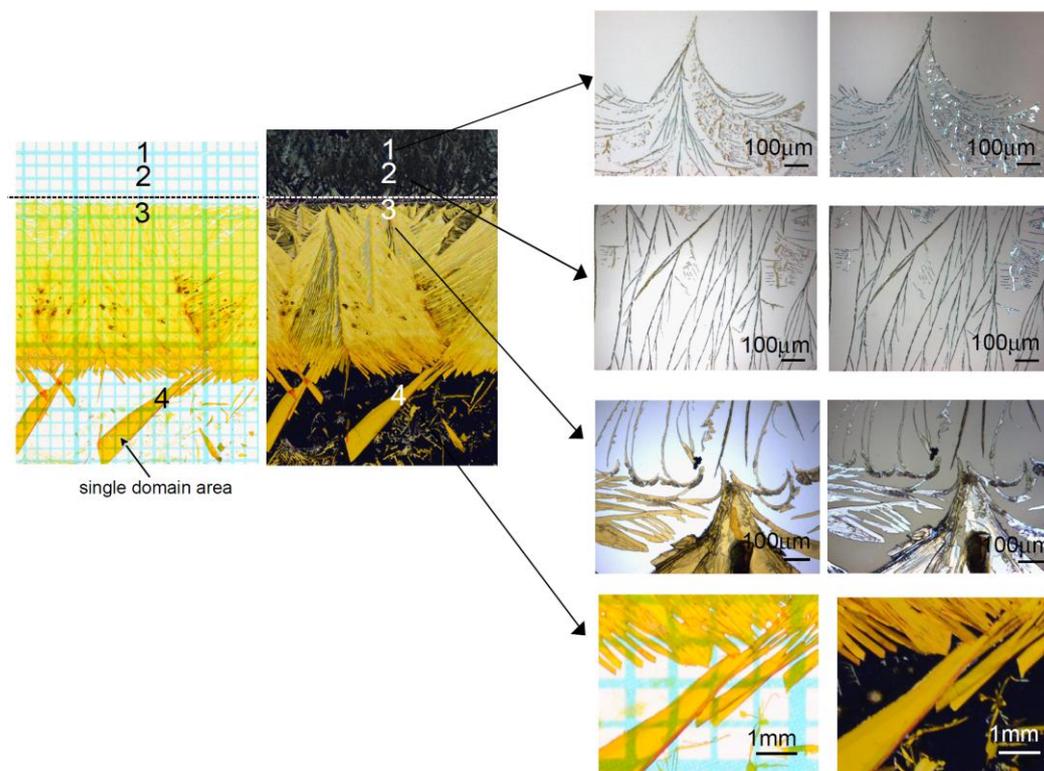


Figure S2. Investigation of the nucleation and the growth mechanism of HMQ-T: Photographs of as-grown HMQ-T crystals with 80 % of the saturated concentration at 40 °C for which the evaporation temperature is 40 °C. The dotted line presents the top contact line with solution. Right images are photographs taken between crossed polarizers. The upper two positions (1 and 2) are in the thin layer in Figure 1.

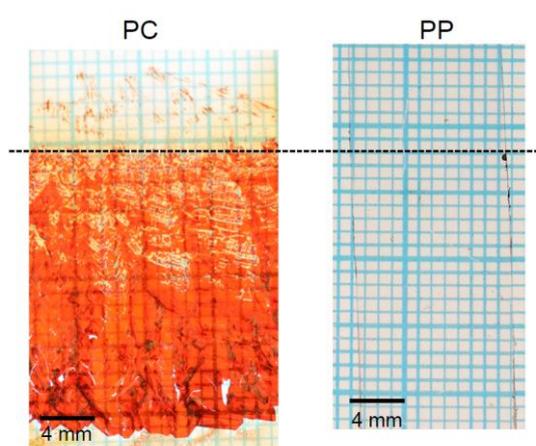


Figure S3. Photographs of as-grown DSTMS crystals with (a) 15 % of the saturated concentration at 30 °C on polycarbonate (PC) substrate having a polar surface, which is however less polar than a glass substrate, and (b) 30 % of the saturated concentration at 30 °C on polypropylene (PP) substrate having a non-polar surface. The evaporation temperature is 30 °C. The dotted line presents the top contact line with solution.

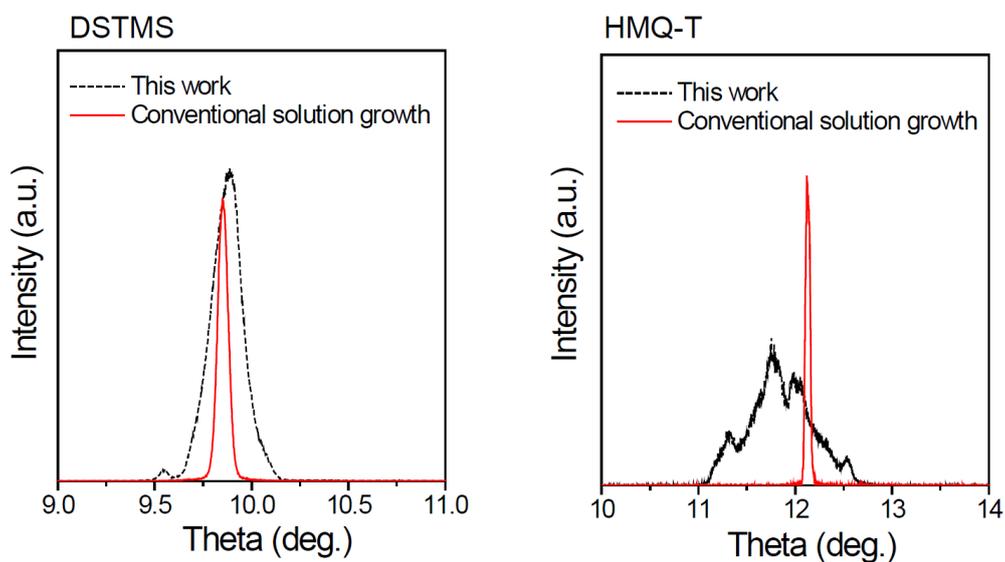


Figure S4. High resolution X-ray diffraction patterns of DSTMS and HMQ-T crystals grown on glass substrate, with 80 % of the saturated concentration at 40 °C for which the evaporation temperature is 40 °C. For comparison, DSTMS and HMQ-T crystals grown by conventional solution growth methods are also measured.

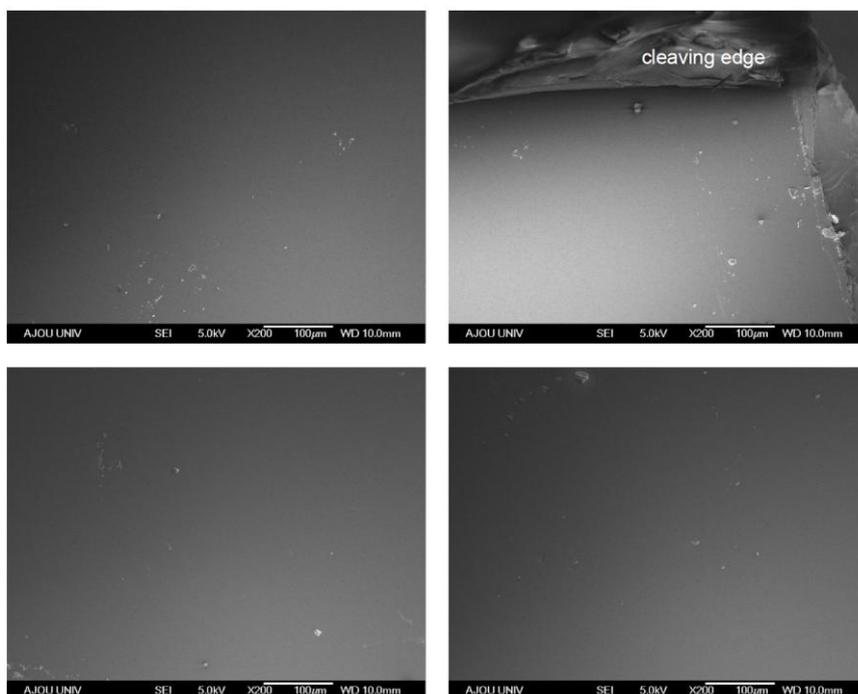


Figure S5. Scanning electron microscope (SEM) images of the highly flat side (i.e. the glass-template side) of DSTMS crystals, grown with 80 % of the saturated concentration at 40 °C.