

## Electronic Supplementary Information (ESI)

for

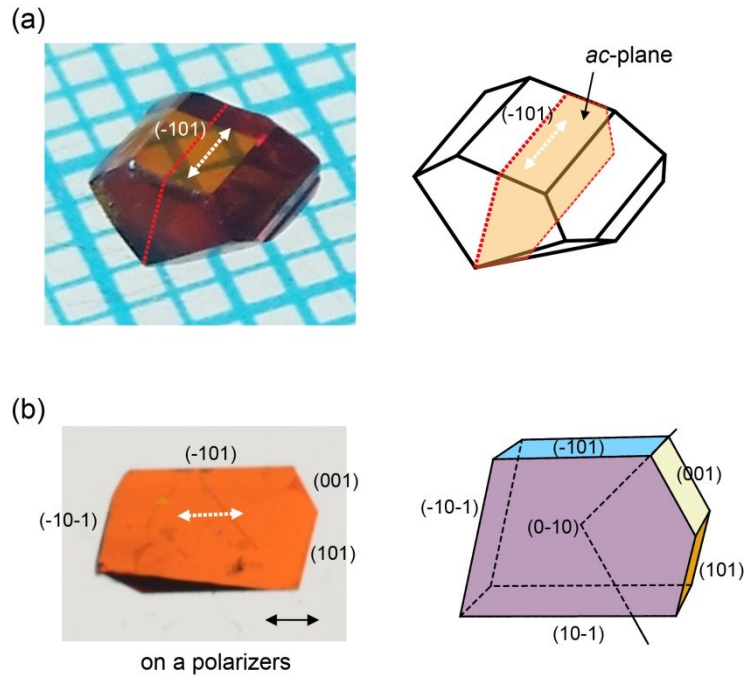
# **Electro-Optic Crystals Grown in Confined Geometry with Optimal Crystal Characteristics for THz Photonic Applications<sup>†</sup>**

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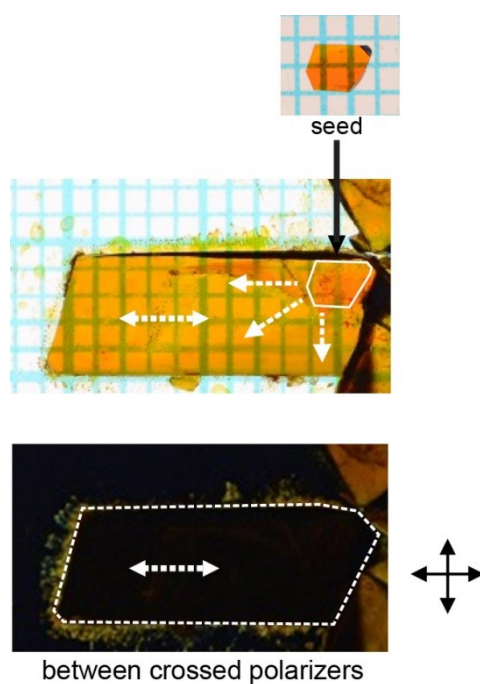
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**Fig. S1** (a) Photograph of HMQ-TMS crystal grown by conventional slow cooling method in methanol, which exhibit hexagonally-shaped block morphology. (b) Photograph of *ac*-plane-cut HMQ-TMS crystal prepared by cutting a bulk single crystal with its facet diagram. The dotted and the solid double arrows are parallel to the polar axis of HMQ-TMS crystals and to the light-polarization direction after the polarizer, respectively.



**Fig. S2** Photographs of HMQ-TMS crystals grown in 0.5 mm-gap confined geometry, in which 0.39 mm-thick HMQ-TMS seed with an area of 3.0 mm<sup>2</sup> grows to crystal with a larger area of 57.0 mm<sup>2</sup>. The dotted arrows present main crystals growth directions. The dotted and the solid double arrows are parallel to the polar axis of HMQ-TMS crystals and to the polarizer directions, respectively.

## **X-ray diffraction reflection patterns of HMQ-TMS crystals**

Fig. 4b shows X-ray diffraction reflection patterns of HMQ-TMS crystals grown in confined geometry. In high diffraction angle region ( $2\theta \approx 44-45^\circ$ ), a small broad peak appears. In this diffraction angle region ( $2\theta \approx 44-45^\circ$ ), various diffraction peak can be appear;  $2\theta = 44.038$  degree corresponding to (331) plane,  $44.216$  degree corresponding to (-226) plane,  $44.572$  degree corresponding to (-5-14) plane,  $44.673$  degree corresponding to (501) plane and  $44.888$  degree corresponding to (233) plane, which are calculated from single crystal structure data of HMQ-TMS crystals in Ref. 11.