

## Supporting Information

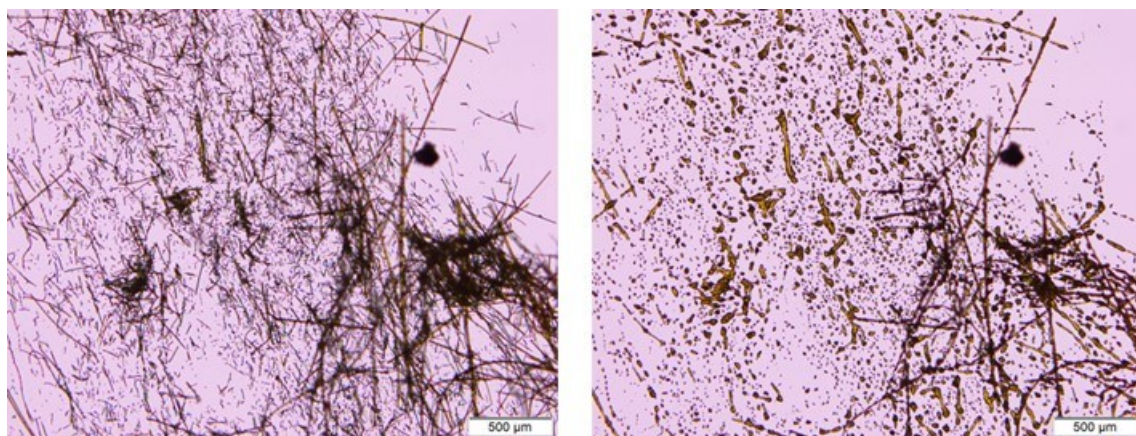
### Negative Phototactic Behaviour of Crystals on a Glass Surface

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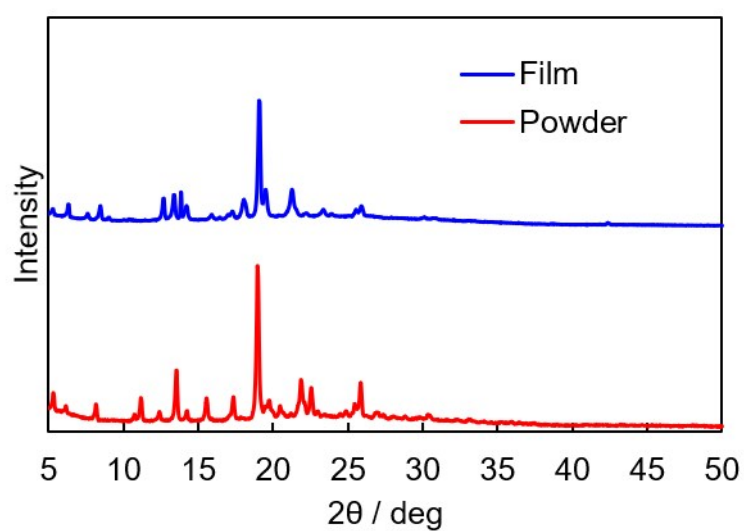
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#### Experimental Section

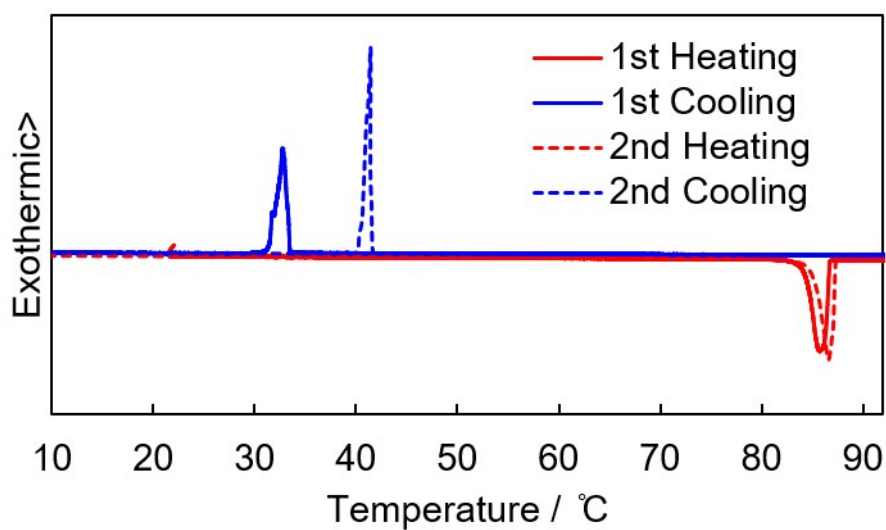
The 4-MAAB was purchased from Tokyo Chemical Industry Co., Ltd. Purification by silica gel column chromatography and recrystallization from hexane were carried out. Absorbance spectra for 4-MAAB solution and a thin film were measured using a Ocean Optics spectrometer (flame). A deuterium tungsten halogen light source (DH-2000-BAL) was used as the probe light. Differential scanning calorimetry (DSC) measurements were carried out using an SII Nanotechnology DSC6100. The 4-MAAB crystals were ground in a mortar and the powders were placed on a cover glass (Matsunami Glass Ind., Ltd, thickness: 0.12–0.17 mm). The glass substrate was put on a stage with a hole 2 cm in diameter in the middle, which enabled light irradiation from the back side of the substrate. The motion of the crystals was obtained using an OLYMPUS BXFM microscope. A Blue LED (CCS Inc., 465 nm: HLV2-22BL-3W, 405 nm: HLV-24VL405) was used for light irradiation. The light intensity was measured using a Newport 1917-R optical power meter with an 818-ST-UV photodetector. For white light irradiation, we used an LED (AITECSYSTEM, TSPA22x8-57W); the illuminance was approximately 800 klx. A xenon lamp (ASAHI SPECTRA, HAL-320) was used as a solar simulated light source; the illuminance was approximately 1500 klx. Photoinduced phase transition of the thin film of 4-MAAB was observed using a polarizing microscope (OLYMPUS BX51). The excitation light source was a high-pressure Hg lamp (USH-103OL); a fluorescence mirror unit (U-MNBV) was used for blue light irradiation. Powder X-ray diffraction experiments were performed by RIGAKU SMARTLAB with Cu K $\alpha$  radiation. For analysis of the motion, we defined the travel distance as the lateral distance moved by the rear edge of the crystal. In each experiment, the average velocity of 9~13 crystals was measured.



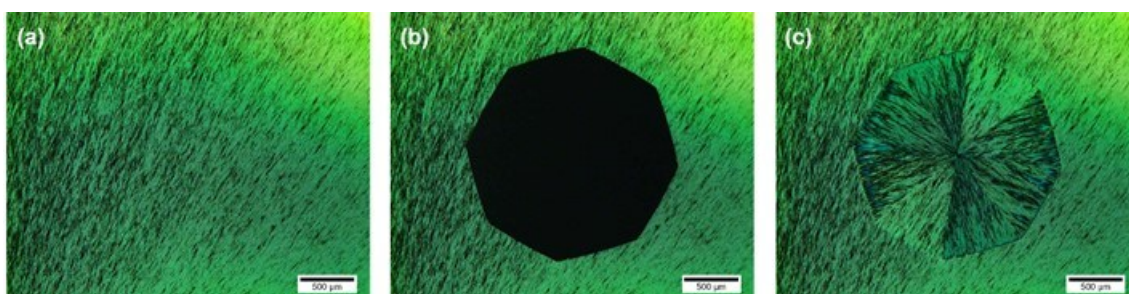
**Fig. S1** Photographs of 4-MAAB fibrous crystals (left) before and (right) after light irradiation at 465 nm. See Movie S1.



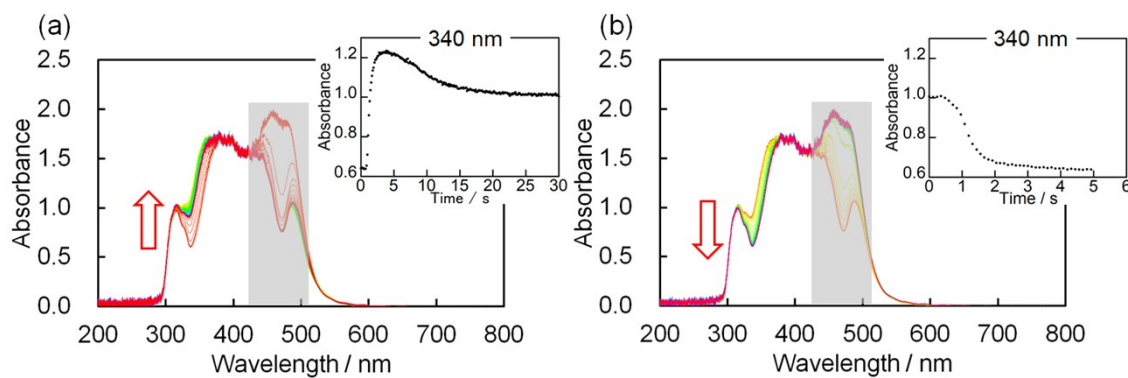
**Fig. S2** A powder XRD pattern of 4-MAAB crystals. "Film" was prepared by cooling the molten sample. "Powder" was prepared by grounding the fibrous crystals.



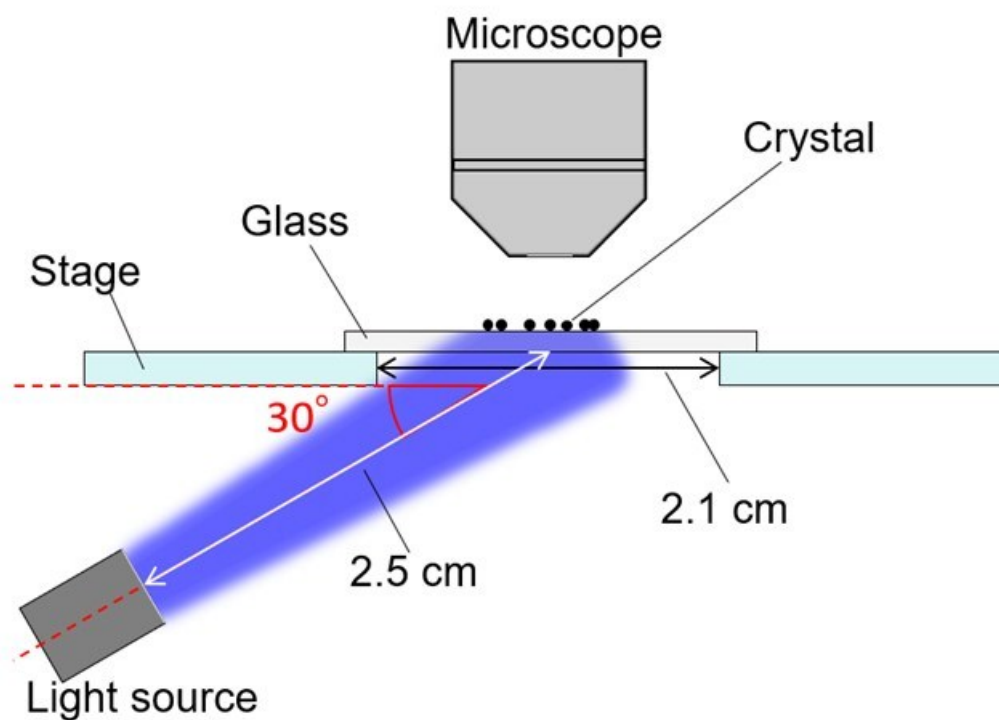
**Fig. S3** Differential scanning calorimetry (DSC) thermograms of 4-MAAB in the heating and cooling cycles at the scan rate of  $0.5 \text{ K min}^{-1}$ .



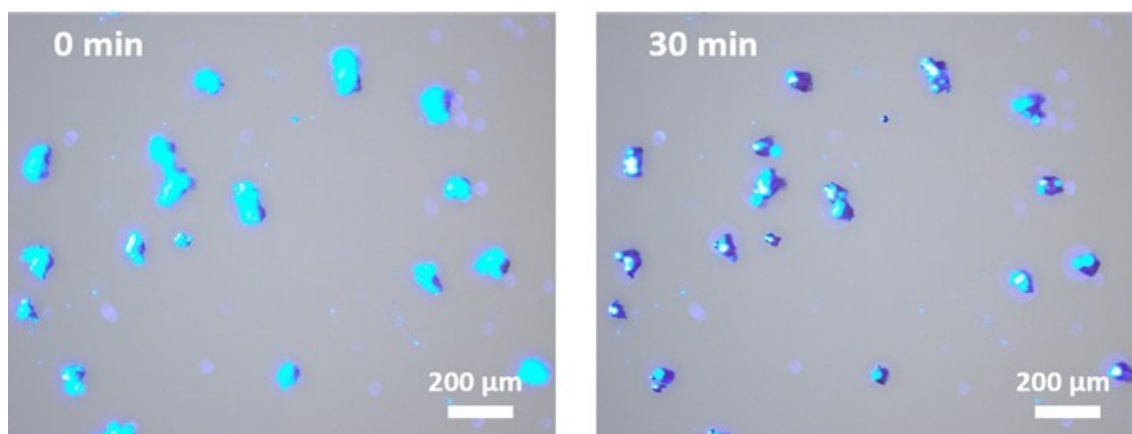
**Fig. S4** Polarizing optical micrographs of a thin film of 4-MAAB crystals (a) before, (b) just after and (c) 200 s after irradiation. See Movie S2.



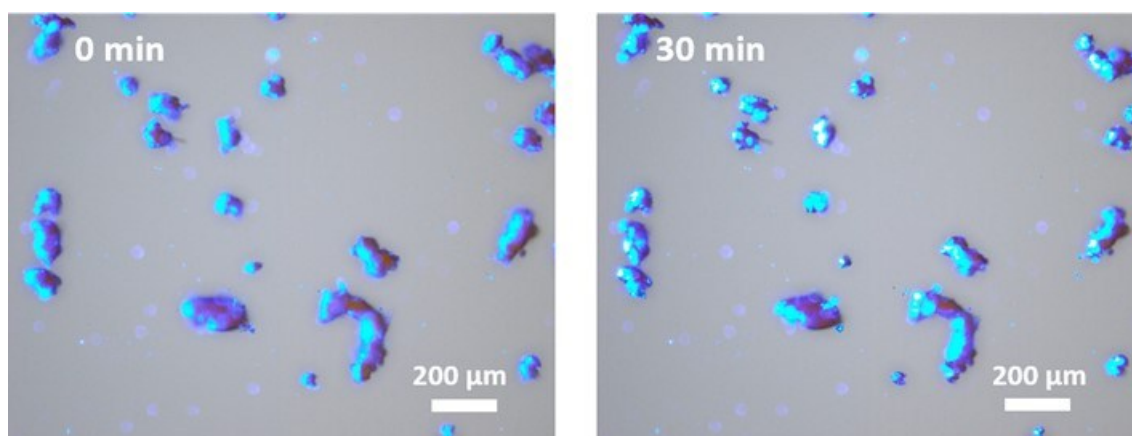
**Fig. S5** Absorption spectra of 4-MAAB as a film (a) during or (b) immediately after ceasing photoirradiation. Each absorption in the grey shaded area contains reflection of the excitation light. The insets show the temporal changes in absorption at 340 nm. A spectrum was recorded every 0.1 s.



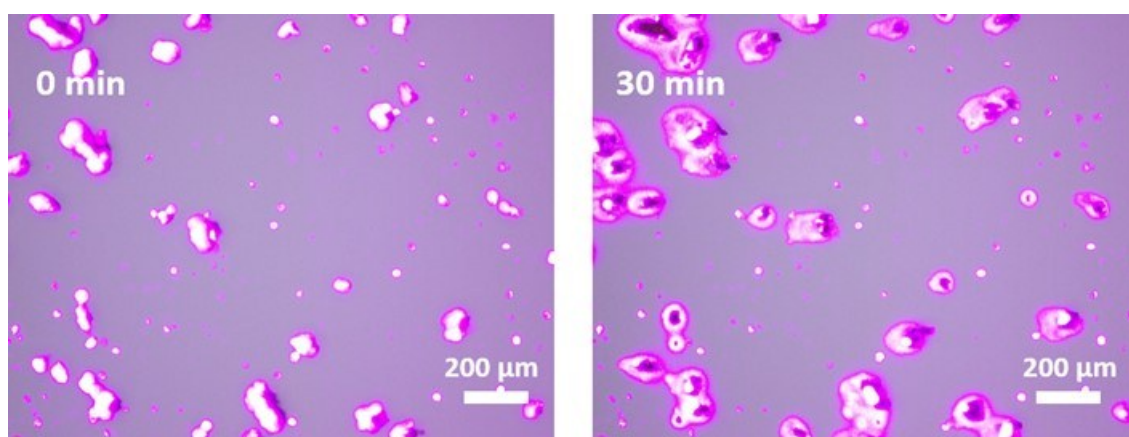
**Fig. S6** Schematic illustration of the experimental set-up.



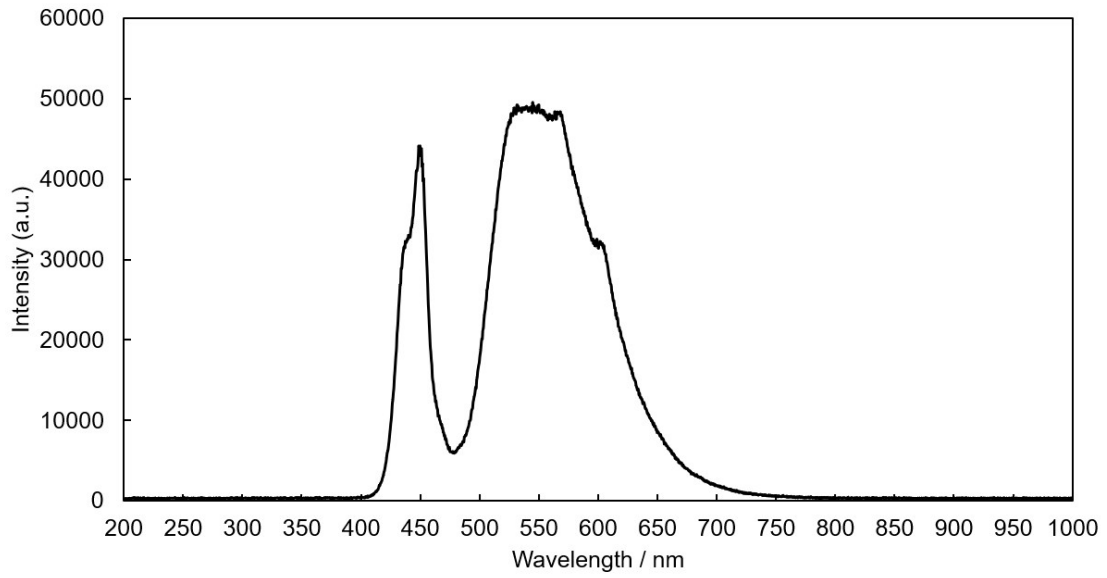
**Fig. S7** Micrographs of 4-MAAB crystals (left) 0 min and (right) 30 min after irradiation from the front side of the substrate. See Movie S4.



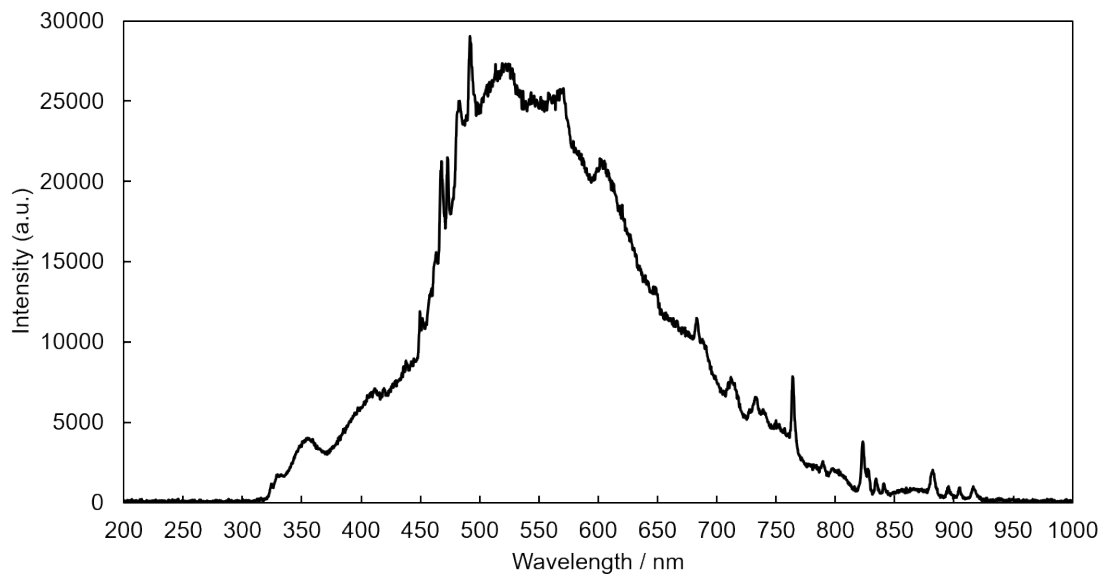
**Fig. S8** Micrographs of 4-MAAB crystals (left) 0 min and (right) 30 min after low-intensity irradiation ( $50 \text{ mW cm}^{-2}$ ) at 465 nm.



**Fig. S9** Micrographs of 4-MAAB crystals (left) 0min and (right) 30 min after irradiation at 405 nm. See Movie S5.



**Fig. S10** A luminescence spectrum of white LED light source.



**Fig. S11** A luminescence spectrum of xenon light source.