Supporting Information

Fabricating Multi-scale Controllable PEDOT:PSS arrays via Templated Freezing Assembly

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Contents

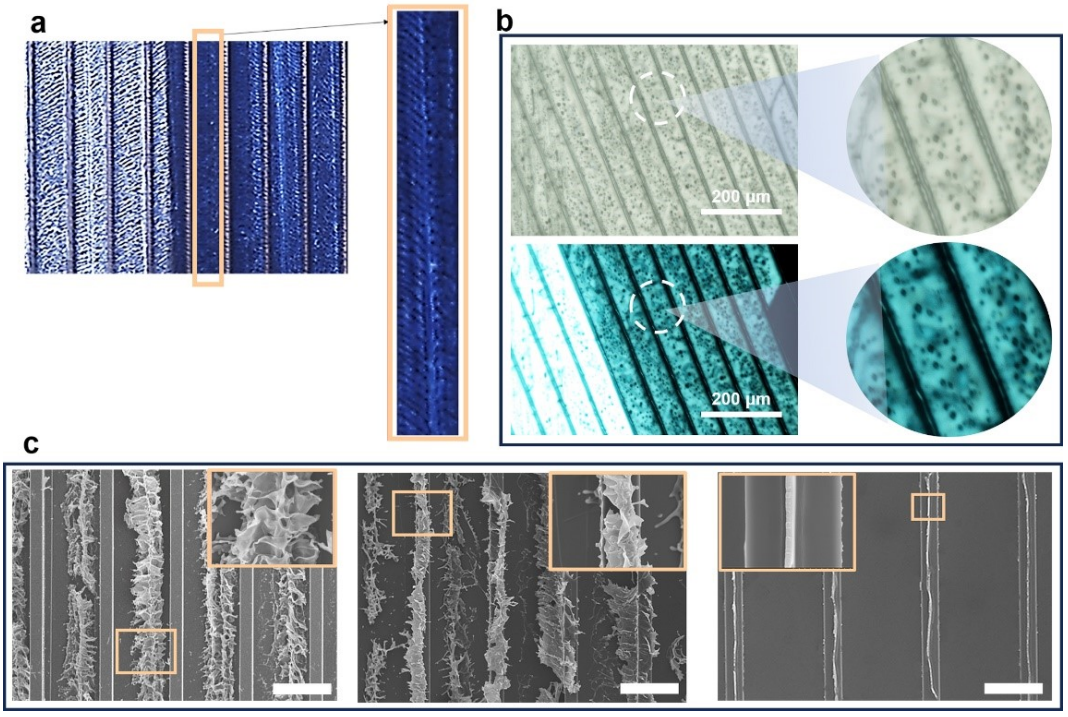

Fig. S1 The optical microscopy image of ice crystal grains. **(a)** The image of tiny ice crystal grains after freezing vertically. **(b)** The image of long-strip ice crystal on template, where the PEDOT:PSS fibers were in situ assembled between unidirectional ice crystal arrays, and obvious birefringence was observed in situ by polarized optical microscopy images. **(c)** SEM images of PEDOT:PSS migrating on the template surface during freezing assembly process. All scale bars were 40 μm.

The preparation of SEM images of PEDOT:PSS migrating on the template surface during freezing assembly process. First, vertical freezing of suspension along the direction of template on the pre-cooled (-40 °C) cryo-stage, the PEDOT:PSS was located in the middle of the template groove. Second, increasing the annealing temperature (range from -3 °C to -9 °C) the PEDOT:PSS was migrating to the template upper surface boundaries. Last, extending assembly time, the PEDOT:PSS was located on the template surface.

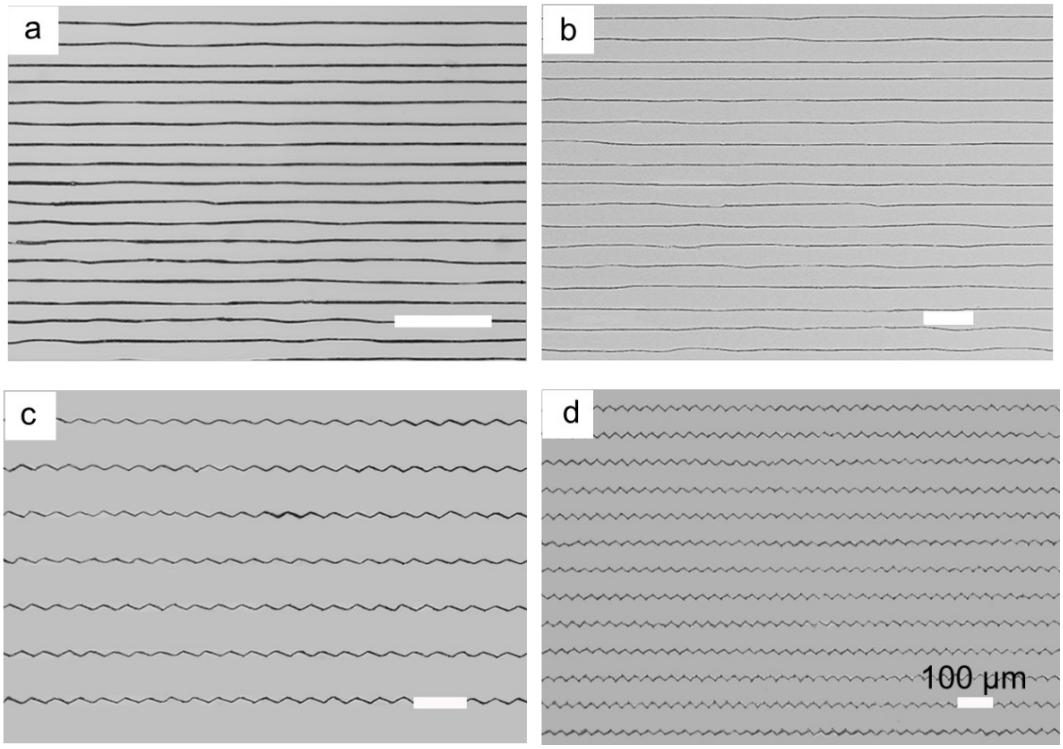

Fig. S2 Large-area PEDOT:PSS fibers array. **(a-b)** Linear nanofibers array (L = 20 μm and 50 μm). **(c-d)** Bending arrays (θ = 120°, L = 90 μm, 120 μm); (θ = 90°, L = 80 μm, 110 μm). All scale bars were 100 μm.

Preparation condition of PEDOT:PSS nanofiber. First, vertical freezing of suspension along the direction of template on the pre-cooled (-40 °C) cryo-stage. Second, increasing the annealing temperature. **(a-b)** L = 20 μm and 50 μm, annealing temperature range from -3 °C to -5 °C, extending assembly time more than 1 h. **(c-d)** Bending arrays (θ = 120°, L = 90 μm, 120 μm); (θ = 90°, L = 80 μm, 110 μm), annealing temperature was -3 °C, extending assembly time more than 1 h, the tiny ice crystal grains annealed into large single crystal, Last, removes single crystal ice the PEDOT:PSS fibers array was obtained.

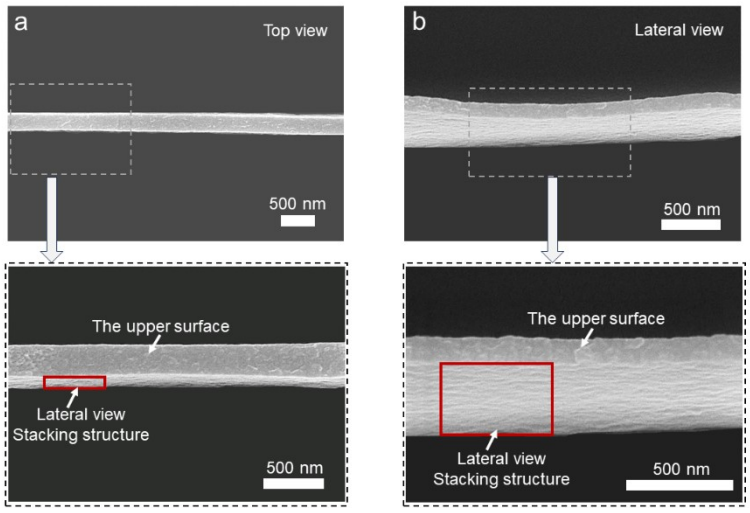

Fig. S3 SEM image of PEDOT:PSS nanowire microstructure. **(a)** The PEDOT:PSS nanowire was rotated 30° clockwise. **(b)** The PEDOT:PSS nanowire was rotated 60° clockwise. The lateral view indicate that the interior of the nanowire was layer-by-layer stacked structure.

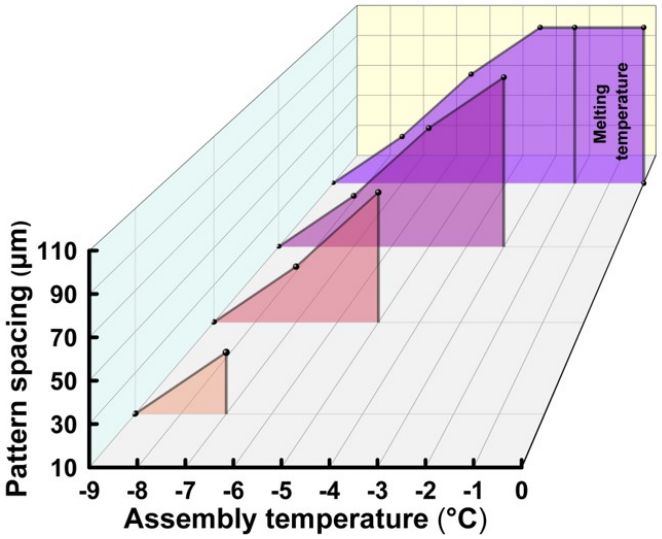

Fig. S4 The phase diagram shows the correlation of assembly temperature with pattern spacing.

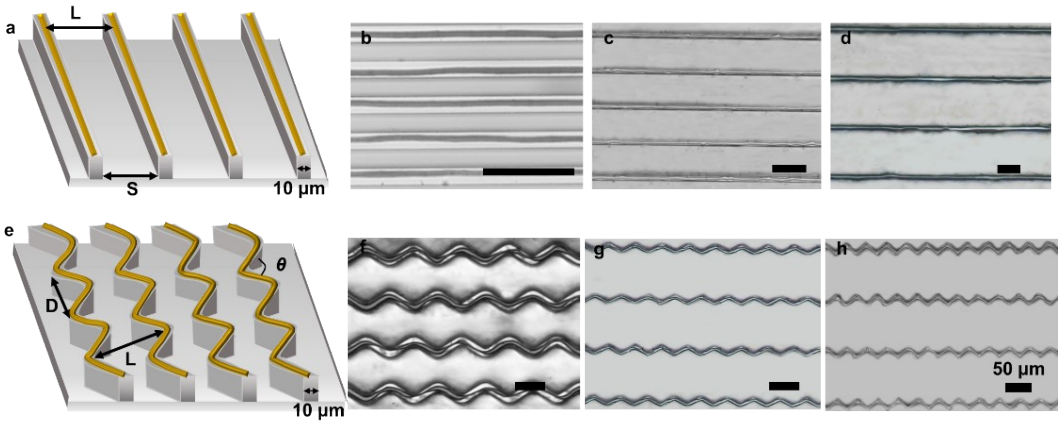

Fig. S5 Optical microscopy images of PEDOT:PSS fibers assembled on upper surface of template. **(a)** Scheme of PEDOT:PSS fibers on linear patterned template. **(b-d)** PEDOT:PSS fibers were assembled on upper surface of template, with the line interval (L) of 20 μm, 50 μm, and 110 μm respectively. **(e)** Scheme of PEDOT:PSS fibers on bending patterned template. **(f-h)** Various bending nanowires on bending template. The $\theta = 120^{o}$ for (f) and (g), $\theta = 90^{o}$ for (h). All scale bars were 50 μm.

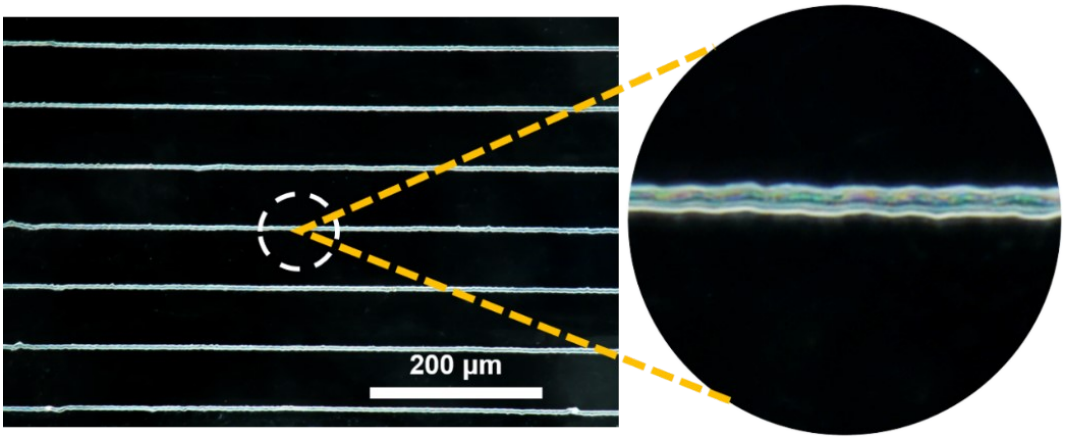

Fig. S6 The dark field image of bending PEDOT:PSS arrays with small angle indicated the freezing assembly strategy can achieve fine control on the micro-morphology of PEDOT:PSS fibers.

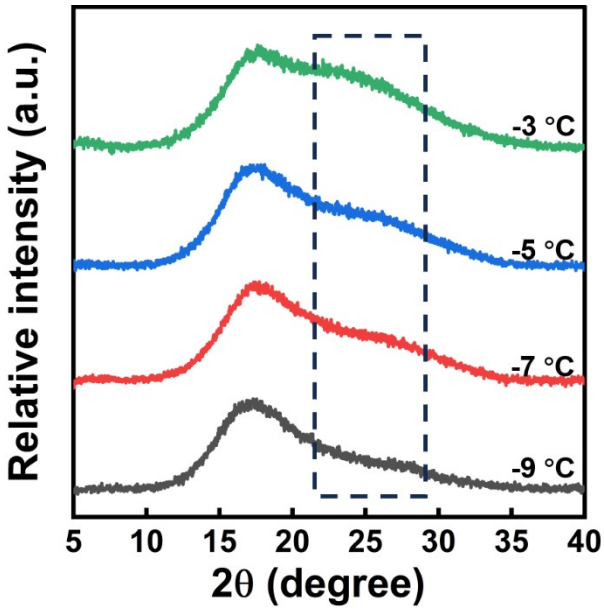

Fig. S7 XRD diffraction pattern of PEDOT:PSS assembled at different temperature (-3 ºC, -5 ºC, -7 ºC, -9 ºC) annealing for 48h. The higher assembly temperature results in stronger intensity at 2θ =25.6°.

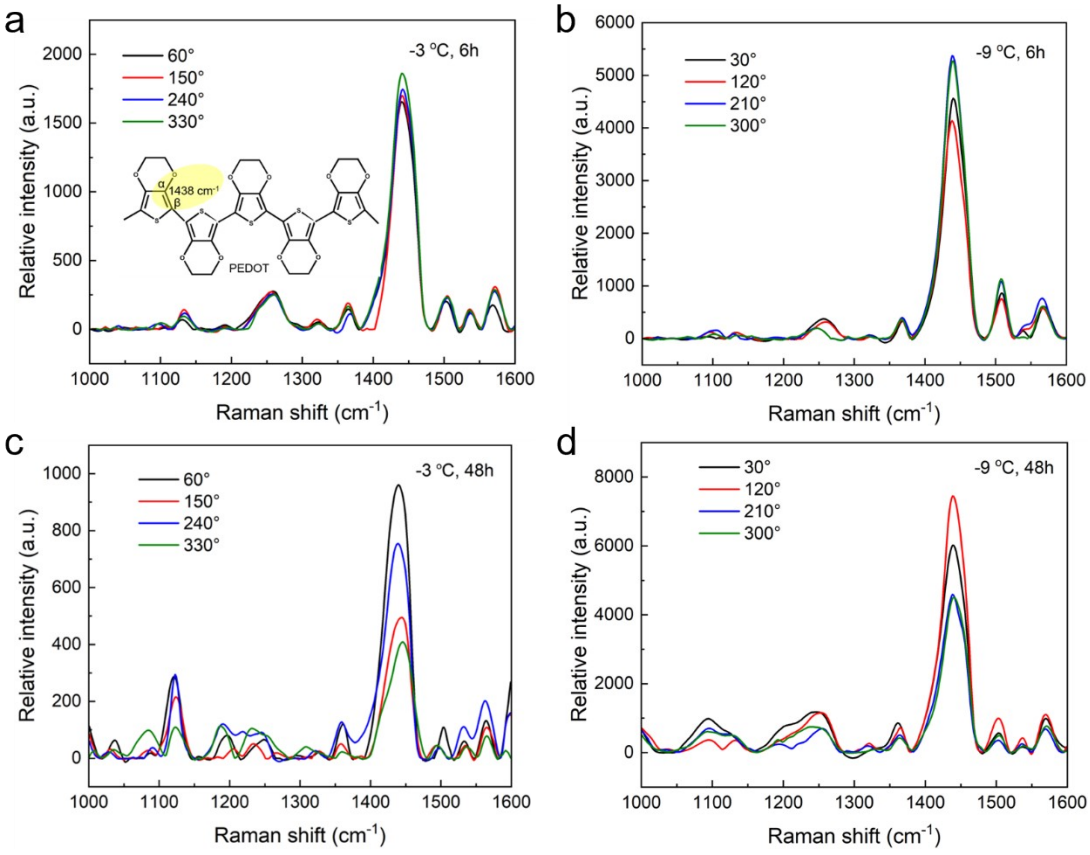

Fig. S8 **(a-b)** Raman spectra of PEDOT:PSS nanofibers assembled at -3 °C and -9 °C for 6 h in different polarized degrees. The peak of 1438 cm^{-1} was attributed to double-bond stretching vibration of $C_\alpha = C_\beta$(-O). **(c-d)** Raman spectra of PEDOT:PSS annealed at -3 °C and -9 °C for 48 h at diverse polarized degrees.

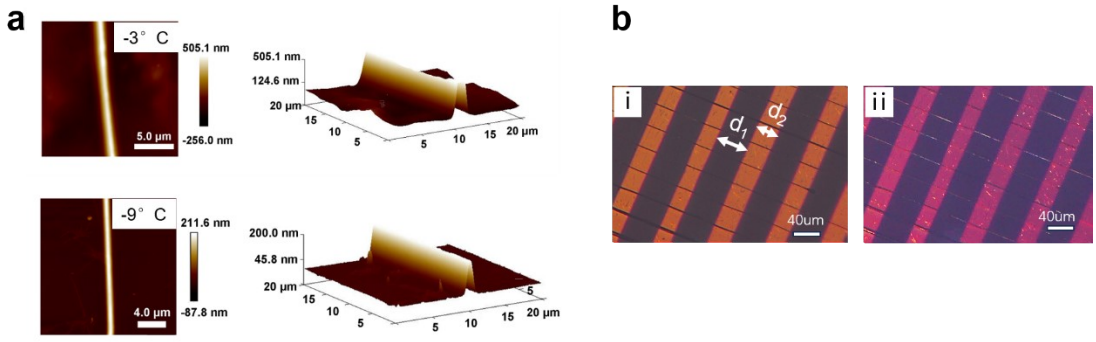

Fig. S9 Morphological characterization of PEDOT:PSS nanowires. **(a)** AFM image of PEDOT:PSS nanowires assembled at -3 °C and -9 °C, respectively. **(b)** Optical microscopy image of PEDOT:PSS pattern arrays coated with Au electrode (i) (d_1 was the distance between two adjacent Au electrodes; d_2 was width of Au electrode) and in situ polarized optical microscopy image of PEDOT:PSS pattern arrays (ii) .

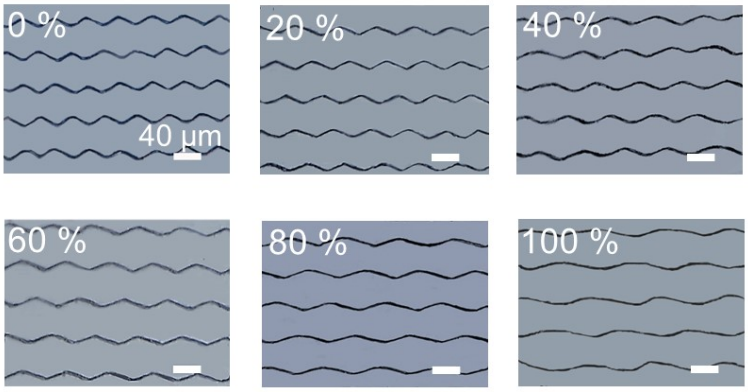

Fig. S10 PDMS film with PEDOT:PSS bend pattern in different strain (0%, 20%, 40%, 60%, 80%,100%). All scale bars were 40 μm.