Electronic Supplementary Information

## Molecularly-ordered hydrogels with controllable, anisotropic stimulus response

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Figure S1. Phase diagram of PDI-DA/PDI-OH content in water. Total chromonic concentration is kept constant at 20 wt%, and the ratio between the two chromonic molecules is varied.



Figure S2. (A) Tensile testing set up in the CellScale Microsquisher. The beam, clamps, and sample are contained in a bath. The beam is controlled by the actuator on one end and tracked by the camera on the opposite end. (B) Example image from the CellSquisher camera. The gap length is measured at the beginning of the test, and a tracking point on the top clamp is used to measure the instantaneous strain.



Figure S3. Phase diagrams of solutions with 20 wt% PDI-DA and varying weight percent of (A) hydroxyethyl acrylate, (B) n-isopropyl acrylamide, or (C) acrylic acid in water.

**Table S1**. Degree of Modulus Anisotropy in Chromonic Films of Varying Crosslink Density.

|                    | PDI-DA content (%) |      |      |      |
|--------------------|--------------------|------|------|------|
|                    | 5                  | 10   | 15   | 20   |
| Modulus Anisotropy | 1.09               | 1.43 | 1.86 | 1.97 |



Figure S4. Polarizing optical microscopy images of 15% DSCG solutions in 50  $\mu$ m capillaries with planar (A,B) and homeotropic (C,D) wall anchoring. (A) Doubly twisted alignment resulting from planar wall anchoring with two defects. (B) Handedness change of molecular twist is visualized by inserting a quarter wave plate (black arrow) at 45° to both polarizer and analyzer. (C) Homeotropic wall anchoring results in light extinction when the sample is parallel to either the polarizer or analyzer. (D) Twisted escaped radial configuration becomes evident when the capillary is at a non-zero angle to the polarizer. Scale bars represent 50  $\mu$ m.



Figure S5. Polarizing optical microscopy images of solutions containing polymerizable, pH-responsive solutions in 50 µm cylindrical capillaries with planar wall anchoring exhibiting doubly twisted alignment. (A) Three defects in alignment resulting from changes in twist handedness. (B) Handedness change of molecular twist is visualized by inserting a quarter wave plate (black arrow) at 45° to both polarizer and analyzer. Scale bars represent 50 µm.



Figure S6. (A) Fluorescence microscopy of a pH-responsive sample swollen in base and stained with fluorescein sodium salt showing the polymer microstructure. (B) Polarizing optical microscopy of pH-responsive samples polymerized in rectangular capillaries and swollen in base for 24 hours. (C) Polarizing optical microscopy with a quarter waveplate inserted (black arrow) to visualize the angle of alignment in samples at varying angles to the waveplate. Yellow coloring indicates alignment along the waveplate, and blue coloring indicates alignment perpendicular to the waveplate. Scale bars represent 200 µm.



Figure S7. (A) Axis-dependent dimensional change of pH-responsive chromonic gels in response to pH 4 or pH 9 solutions. (B) Axis-independent dimensional change of pH-responsive model gels in response to pH 4 or pH 9 solutions.