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Electronic supplementary information (ESI)

Supporting Information for this work is available free of charge:

Cross-sectional scanning electron micrographs and polarized optical micrographs of the studied freestanding liquid crystal network, polyamide 6, poly(ethylene terephthalate) films and bilayers.

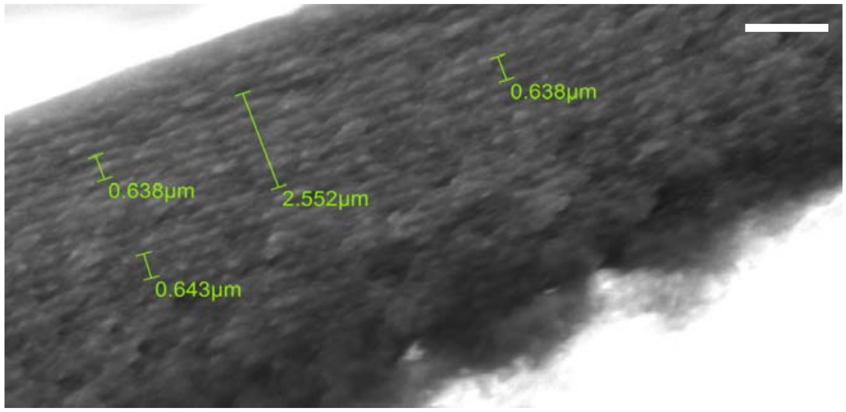


Figure S1. *A cross-sectional scanning electron micrograph of a freestanding chiral nematic poly(nOBA/C6M) film containing 3.13 wt.% of LC756. The film was cold fractured in liquid nitrogen to determine its helical twisting power. The scale bar equals 2 µm*

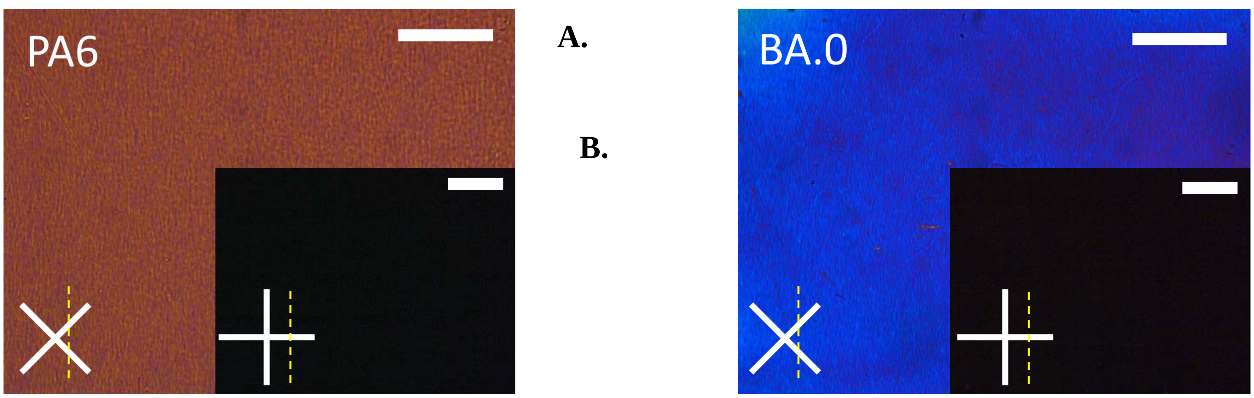


Figure S2. *Polarized optical micrographs of **A.** oriented PA6 and **B.** the LCN/PA6 bilayer actuator with a 0 ° in-plane twist angle (**BA.0**). The homogenous black state between crossed polarizers in the bilayer films is indicative for a self-aligned LCN coating. The yellow dashed line represents the orientation direction of PA6. The scale bars equal 100 µm.*

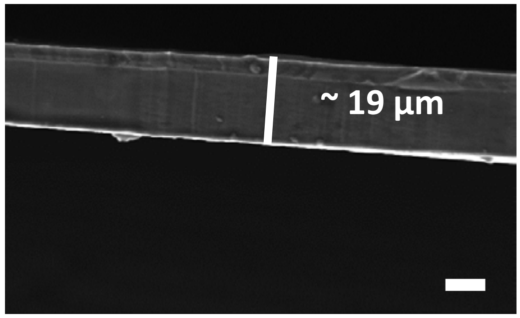
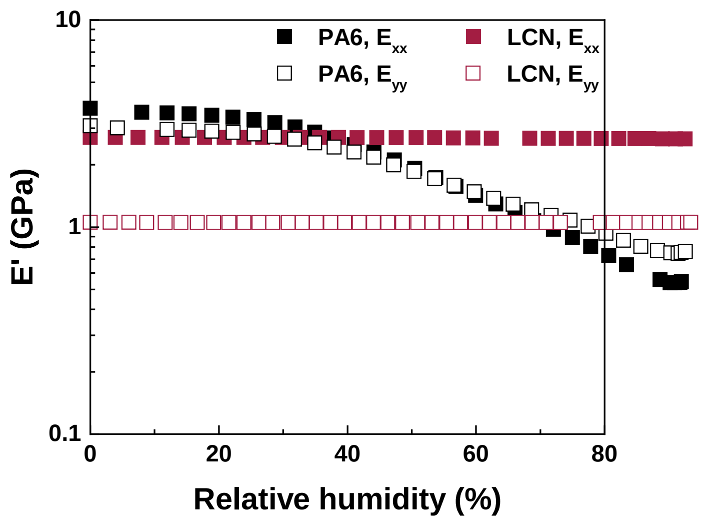
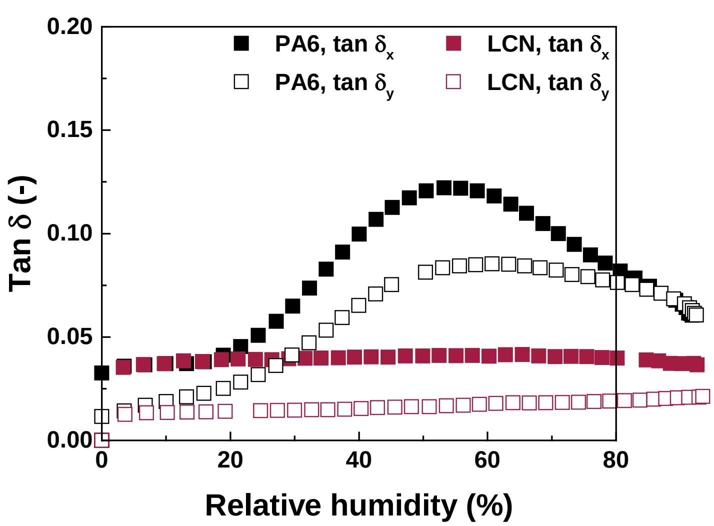


Figure S3. *A cross-sectional scanning electron micrograph of a LCN/PA6 bilayer actuator. The commercial PA6 film is 15 micrometers thick and topped by a spray-applied 4 micrometer LCN coating. The scale bar equals 10 µm.*

A.



B.



C.

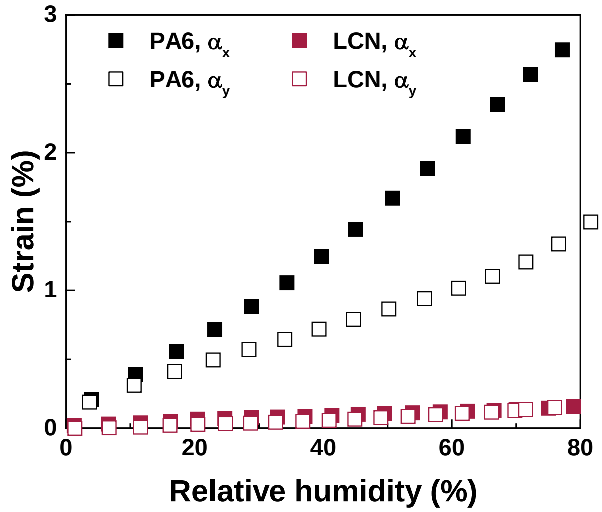
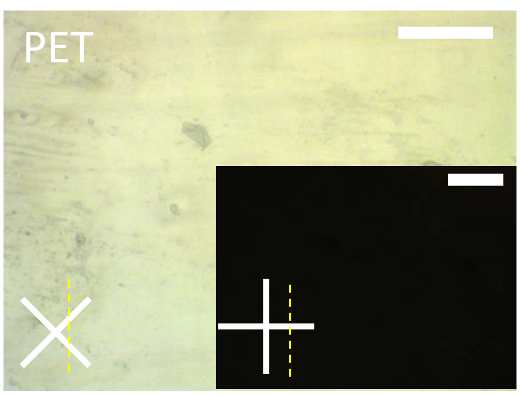


Figure S4. *Dynamic mechanical analysis of oriented 15 µm PA6 and 20 µm LCN films.* ***A.*** *Storage moduli (E') of polyamide 6 (PA6, black symbols) and poly(nOBA/C6M) (LCN, red symbols) measured parallel (filled symbols) and perpendicular (open symbols) to the alignment director plotted as a function of relative humidity.* ***B.*** *The anisotropic tan δ values of both PA6 and LCN films.* ***C.*** *Coefficients of humidity expansion parallel (filled symbols) and perpendicular (open symbols) to the LC alignment director given as a strain (ε).*

A.



B.

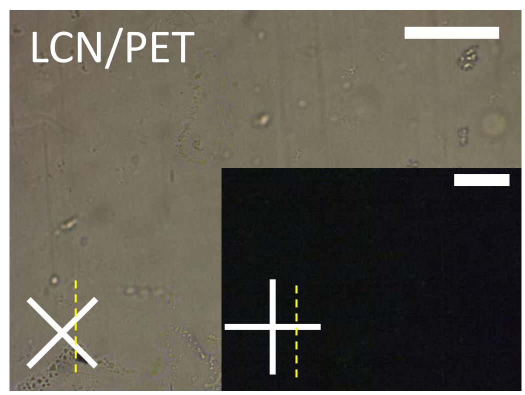


Figure S5. *Polarized optical micrographs of* ***A.*** *oriented poly(ethylene terephthalate) (PET) and* ***B.*** *the LCN/PET bilayer actuator with a 0 ° in-plane twist angle (****BA.0****). The homogenous black state between crossed polarizers in the bilayer films is indicative for a self-aligned LCN coating. The yellow dashed line represents the orientation direction of PET. The scale bars equal 100 µm.*

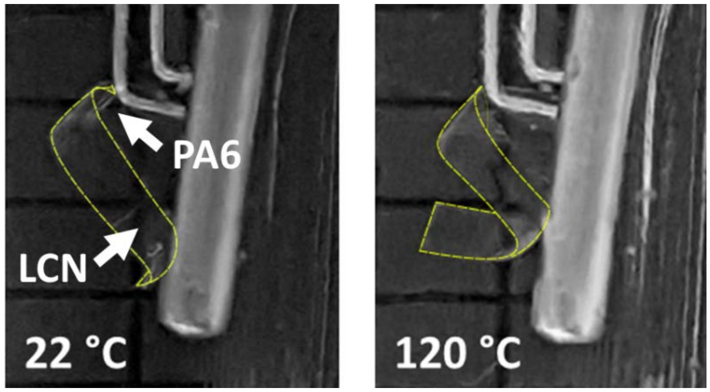


Figure S6. *Preliminary results on a pre-dried PA6 with an alternative spray-applied chiral nematic liquid crystal coating that thermally actuates. The images shown are taken at 22 °C and 120 °C in a silicon oil bath.*

Finite Element Method

The numerical model was composed in MSC. Marc Mentat® (v.2014.0.0) assuming a 2-D geometry of a flat ribbon (20 x 3 x 0.019 mm^3). The high aspect ratio (length: thickness) ribbon was subdivided in 40 x 6 solid-shell elements to reduce computational times. A quadratic thin-shell element (Element Type 139) was utilized with all displacements and rotations as degrees of freedom (DOF).

Bilayer actuators were simulated as a composite consisting of a 15 µm uniaxial PA6 substrate and a 4 µm LCN coating. Molecular orientation (twisted alignment) was embedded in the LCN coating by treating it as a multi-layer composite (15 layers in total) with a linearly increasing in-plane twist angle over the thickness. The boundary conditions at the clamping position is chosen as a 6-DOF fixed condition at the short end of the ribbon, thereby imposing zero translation/rotation of the 6 nodes at one end. The numerical solver further assumes large-strain behavior, and increases the input value of RH % linearly in time.