Geometry-Dependent Adhesion in Transparent, Monodomain Liquid Crystal Elastomers

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Supplementary Information:

Polarized Optical Microscopy

The quality of the films is characterized by polarizing microscopy. The LCE films, placed between crossed polarizers can be seen in Figure S1: the isotropic case shows no light at any angle due to a lack of order in the system; the homeotropic case also shows no light at any angle, due to mesogenic alignment in- and out-of-the-page. The two planar alignment modes show a dark state when the director is aligned with one of the polarizers and a bright state at 45°.

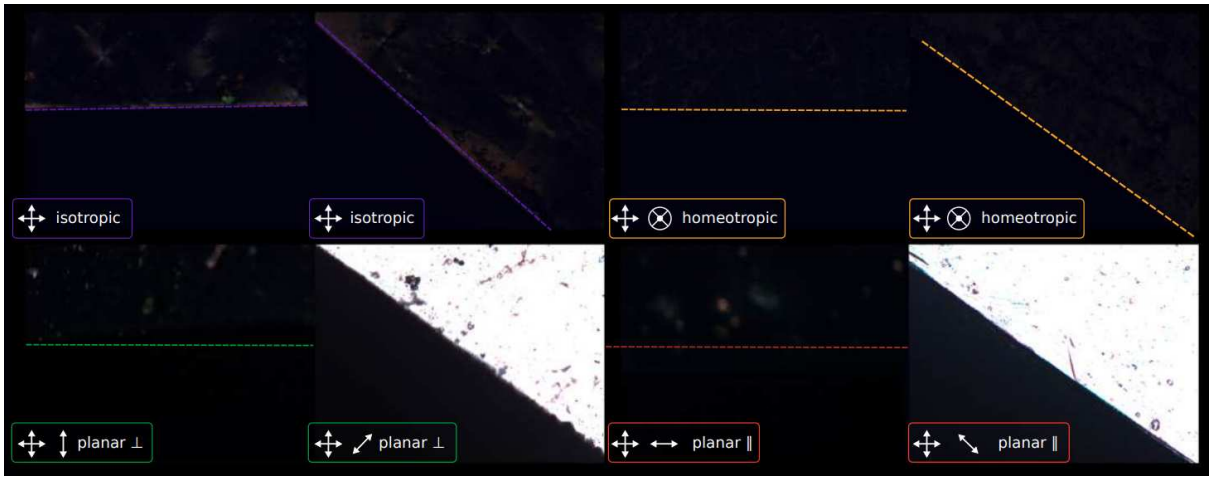

Figure S1. Polarizing microscopy images of LCE films between crossed polarizers. The excellent dark states of the homeotropic sample and the planar samples aligned with one polarizer confirm the high-quality liquid crystal alignment in the films. Each image shows a region ~1mm in size.

Debonding Force Measurements (Peel Test)

Peel measurements were performed at room temperature (20.5 °C). Due to the importance of surface effects on adhesion strength, the glass slides were thoroughly cleaned using sonication and isopropyl alcohol. In the float-glass production process, one side of a glass plane is in contact with tin and one with air, the air-side of the glass slides (determined *via* light scattering) was used for all peel test measurements. A weight was suspended from the strip of LCE which was adhered to the glass slide, maintaining a 90˚ peel angle. The mass of the weight was increased until steady-state peeling was achieved. No residue remained on the glass slide after the peel.

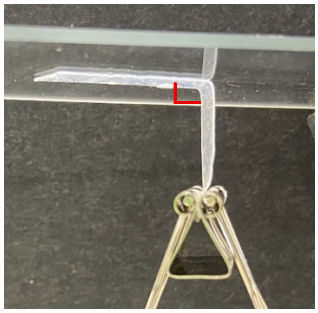

Figure S2. Photograph of the peel test in action. The peel angle of 90˚(indicated by the red lines) is a condition of the geometry where the slide is horizontal and the variable mass is vertical.

Relative Magnitudes of Terms Contributing to the Peel Energy

The adhesive energy per unit area, as shown in Eq. (1) in the main paper, consists of a 1st and 2nd order expansion term for the force per unit width found during the peel test. In the discussion, the second term was ignored due to its negligible impact on the result; the detailed measurement results are shown in Table S1. Due to the low magnitude of the force per width (F/w) measured in this peel test, the square of F/w, and thus the second term in Eq. (1) is insignificant compared with the measurement accuracy of the micrometer used.

Table S1. A comparison of the adhesive energy per unit area calculated using Equation 1 (main text) showing the magnitude of the first and second terms.

Alignment	**Θ (1st Term Only) [Jm^{-2}]**	**Θ (1st + 2nd Term) [Jm^{-2}]**
Planar Parallel	6.7 x10^{-5}	6.7 x10^{-5} + 1.9 x10^{-12}
Planar Perpendicular	4.3 x10^{-5}	4.3 x10^{-5} + 4.7 x10^{-12}
Homeotropic	1.5 x10^{-5}	1.5 x10^{-5} + 4.1 x10^{-12}
Isotropic	2.6 x10^{-5}	2.6 x10^{-5} + 5.4 x10^{-12}

Post-Peel Residue Test

Visual inspection (including at 100x magnification *via* microscopy) indicated that no residue was left on the glass slides after peeling the LCE films, suggesting adhesive rather than cohesive failure. To confirm the lack of residue associated with the LCEs, the system with highest adhesion (planar parallel) was considered and compared with other adhesive films. 1cm^2 squares of different materials were pressed onto glass slides under the same conditions as the peel test. The mass of the slide was recorded using a balance with 0.1mg accuracy before and after the adhesive had been applied and removed. Any increase in the mass is due to residue left behind by the tape, Table 3. The LCE failure mode left no discernable residue, indicating a purely adhesive system. This can be compared to three other tapes, 3M Kapton Tape, Selloltape Double-Sided, and an unbranded packing tape, which left behind 0.7, 0.9, and 0.5 mg of residue respectively. Each test was performed three times.

Table S2. Masses of glass slides before and after a peel test to determine the mass of residue left behind.

Tape	Slide Mass Before Adhesive ±0.0001 (g)	Slide Mass After Adhesive ±0.0001 (g)	Difference ±0.1 (mg)

LCE Planar Parallel	5.2896	5.2896	0.0
Kapton	5.2896	5.2903	0.7
Double-Sided Sellotape	5.2893	5.2902	0.9
Unbranded Packing Tape	5.2978	5.2983	0.5

Supplementary Data:

The data used in the article associated with this document can be accessed under the DOI https://doi.org/10.5518/1601 and contains data from: the peel testing, the dynamic shear testing, and the contact angle measuring.