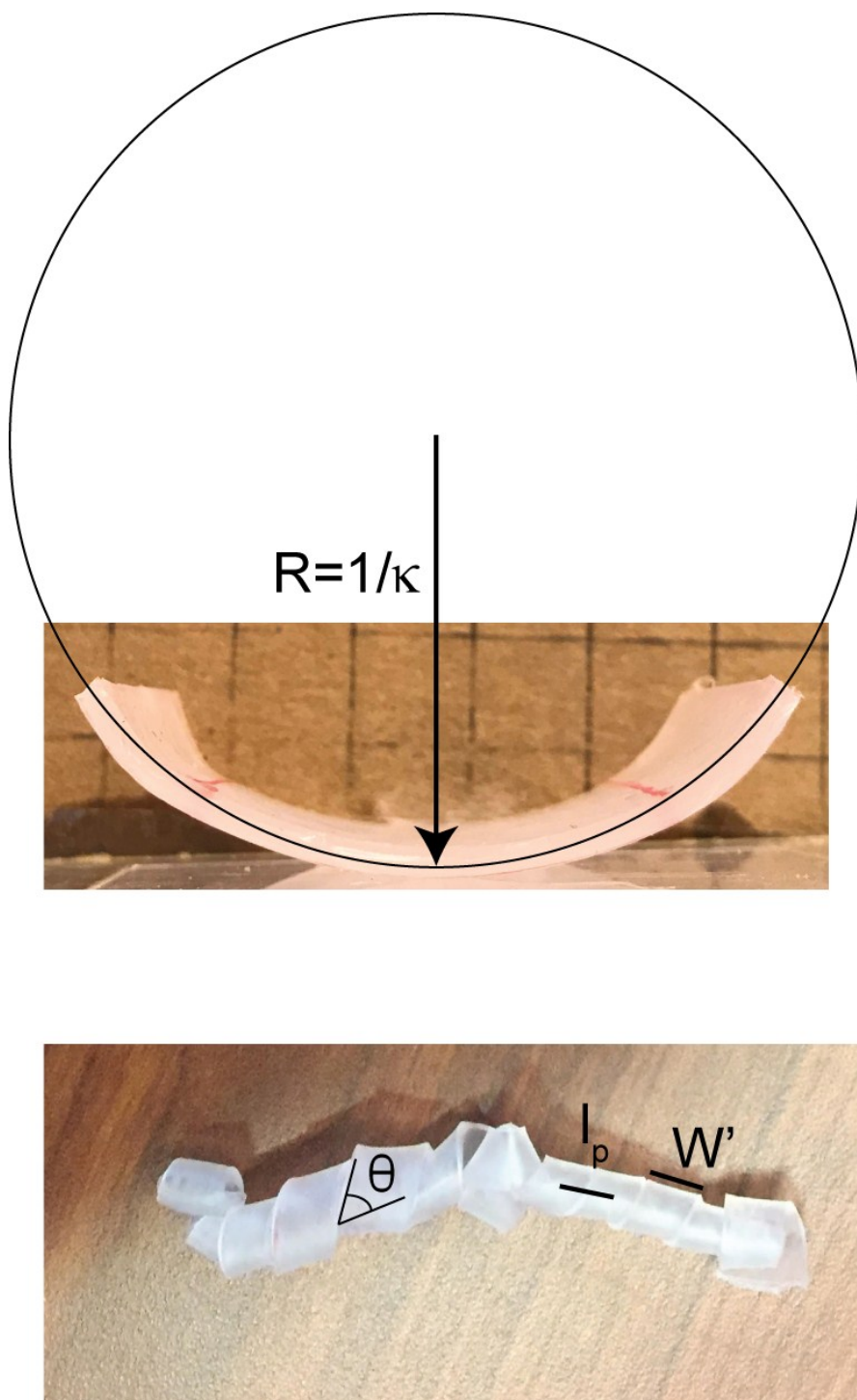
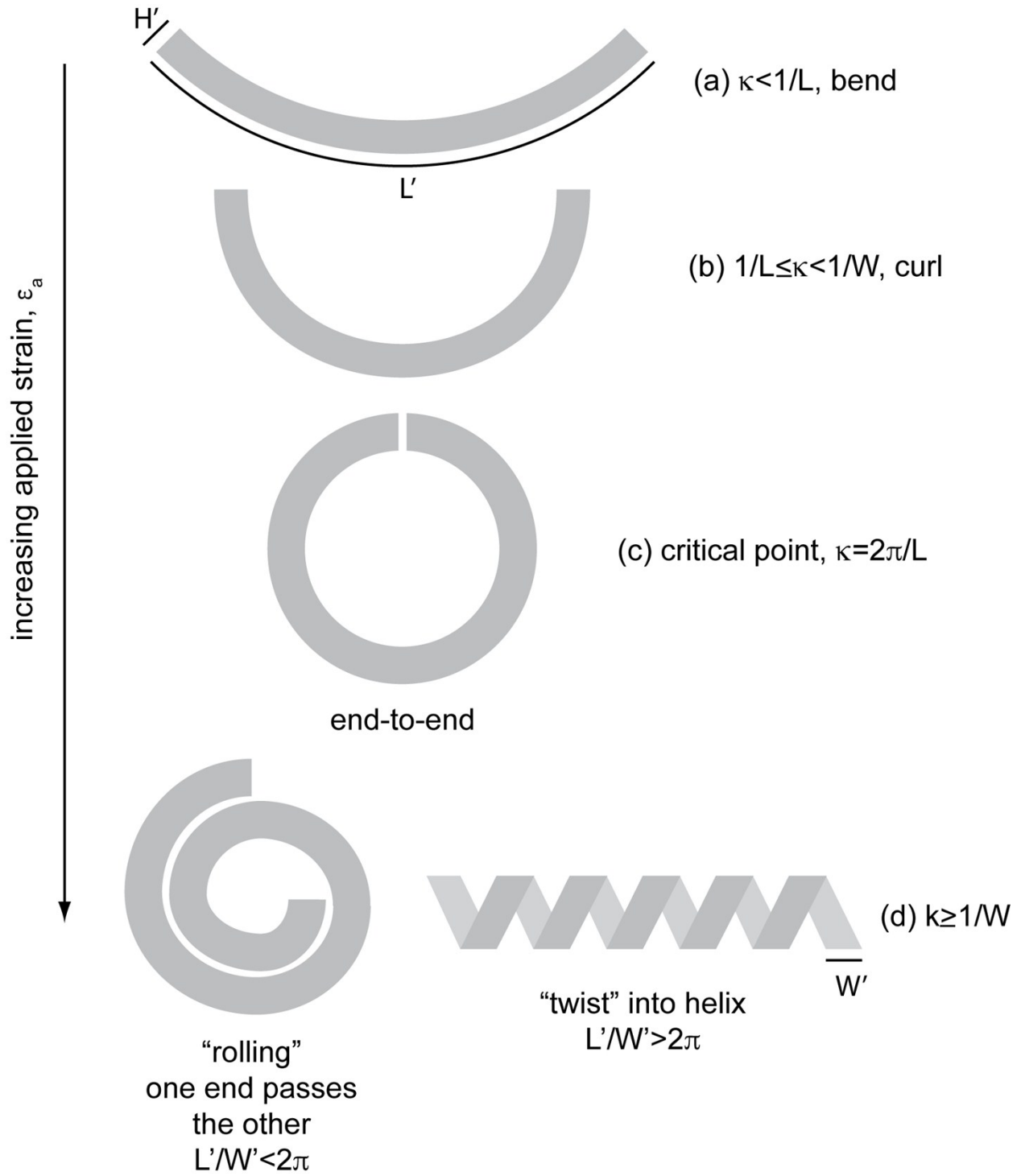


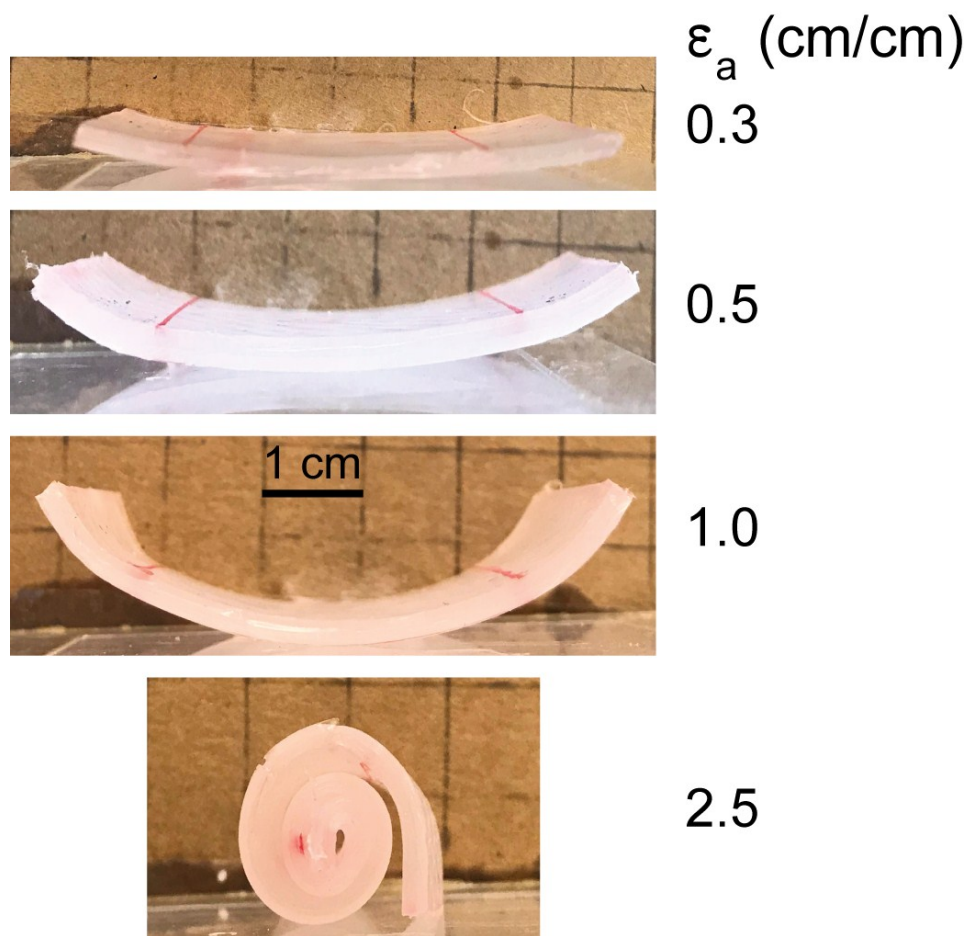
Electronic Supplemental Information (ESI)



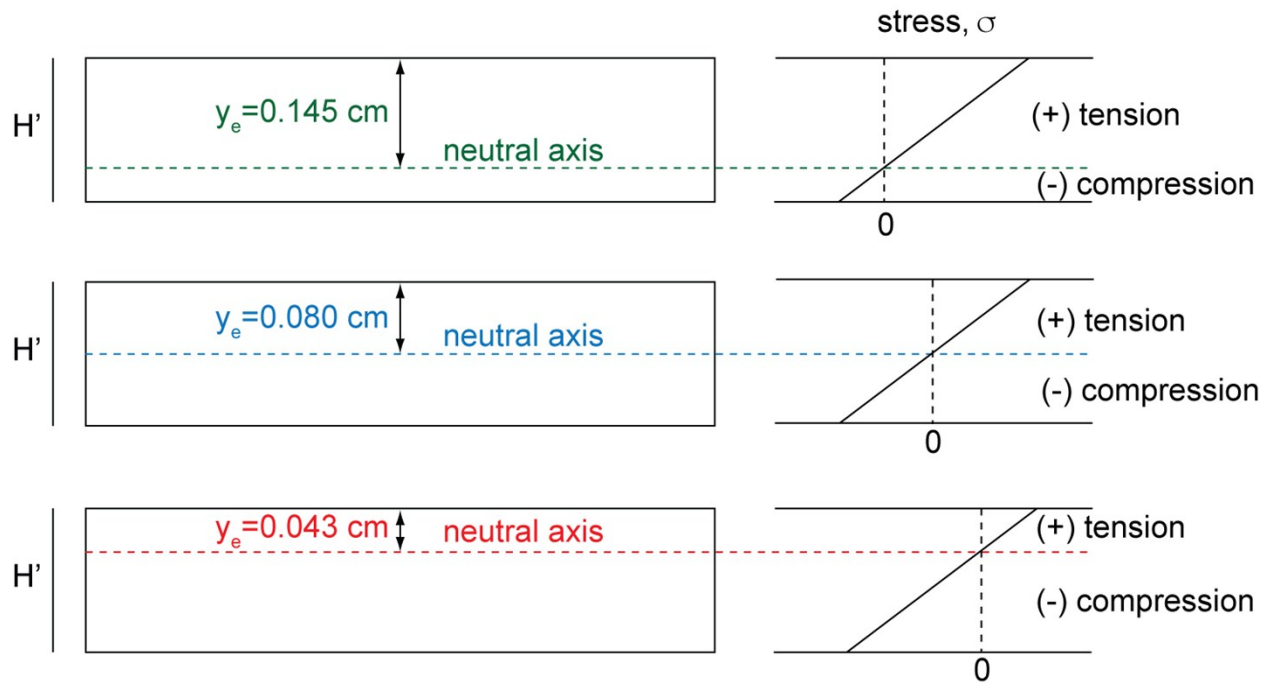
**Figure S1.** Definition of radius of curvature,  $R$ , curvature,  $\kappa$ , helical pitch,  $l_p$ , new resting width,  $W'$ , and helical pitch angle,  $\theta$ . Top:  $L/W=1$ ,  $W=3$  cm EG8842:EG8450 bilayer. Bottom:  $L/W=10$ ,  $W=1$  cm EG8842:EG8450 bilayer.



**Figure S2.** Schematic of the observed bilayer behavior showing (a) bending at low applied strain when  $\kappa < 1/L$ , (b) curling at moderate applied strain when  $1/L \leq \kappa < 1/W$ . (c) A critical point is reached at  $\kappa = 2\pi/L$  when the two ends of the bilayer touch. When the new resting aspect ratio  $L'/W' < 2\pi$  the bilayer rolls (d, left hand side). When  $L'/W' > 2\pi$ , the bilayer twists into a helix (d, right hand side).



**Figure S3.**  $L/W=1$ ,  $W=3$  cm bilayer strips bend at applied strains up to  $\epsilon_a=1$ . At  $\epsilon_a=2.5$  the bilayer strips roll into a tight cylinder as the ends pass one another but do not transition to twisting and  $L'/W'<2\pi$  but  $\kappa>1/W'$ .



**Figure S4.** The neutral axis is empirically observed to shift in the metastable region for the thermoplastic elastomer bilayers. Neutral axis shifts are shown quantitatively in Figure 6.