ADVANCED FUNCTIONAL MATERIALS

Supporting Information

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Light-Controlled Actuation, Transduction, and Modulation of Magnetic Strength in Polymer Nanocomposites

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Detailed explanations on the reversible magnetization behavior are added here supporting the findings shown in Figure 5. Assumptions refer to the Stoner-Wohlfarth theory,[1] as previously shown.[2,3] Measurements, in parallel orientation show remanent magnetization that increases slightly until the phase transition temperature upon which the remanent magnetization decreases about 14%. When the sample is re-cooled, the magnetization increases about 10% at the phase transition temperature, and finally shows reversible magnetization behavior with further cooling. The decrease in magnetization after the first cycle can be explained by both effects of entropy driven magnetization relaxation and a loss of orientation information by some particles during the reorientation process. It is interesting that similar behavior is not found for the LCE nanocomposite, when it was stretched to $\lambda = 2.0$ (Figure 6c), where higher alignment of the major magnetic axis is imprinted, and thus magnetization vector rotation is less favorable. The observed thermal hysteresis confirms the findings from the thermoelastic experiments (Figure 1). The same effect has been observed in temperature dependent X-ray studies (not presented here), and is associated to the polymesomorphism of the polymer, *i.e.* a symmetry break at 45 °C and the isotropization at 80 °C.[2,4] The initial remanent magnetization is weaker in the perpendicular direction than in the parallel direction, due to the decreased orientation of the preferred particle magnetization

direction.[2,3] Thus, during the experiment it decreases significantly about 16% due to randomization of the magnetization vector that aligns with the major particle axis. In order to distinguish this effect, in Figure 5, the non-stretched reference sample LCE is shown for comparison (shifted for overlapping), as it follows the same trend in the heating-cooling cycle. Based on this difference, the phase transition related change in magnetization is obvious also in this measurement: Upon heating, the remanent magnetization passes through a plateau during the expansion of the sample in the measuring direction, due to the increasing population of particles oriented along the measuring direction, as observed in the X-ray experiments. Upon cooling, with the phase transition, the remanent magnetization recovers its originally observed trend, which is obvious in comparison to the reference LCE nanocomposite.

Supporting Figures:

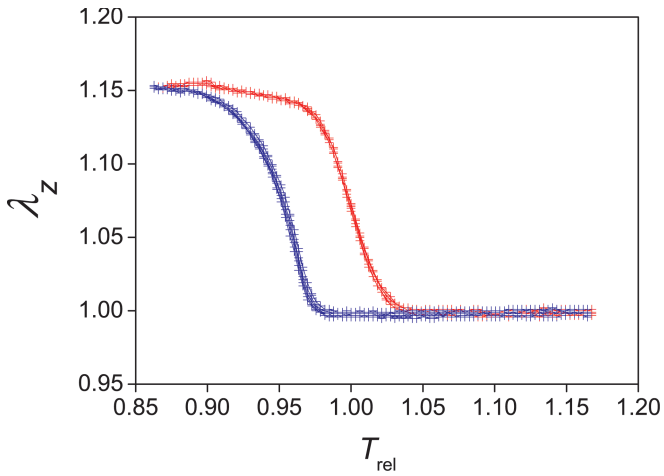

Figure S1. Thermoelastic hysteresis of the OLCE. The shape change during six subsequent heating (red)-cooling (blue) cycles is shown in function of the relative temperature, normalized to the clearing temperature $T_{rel} = T / T_{cl.}$.

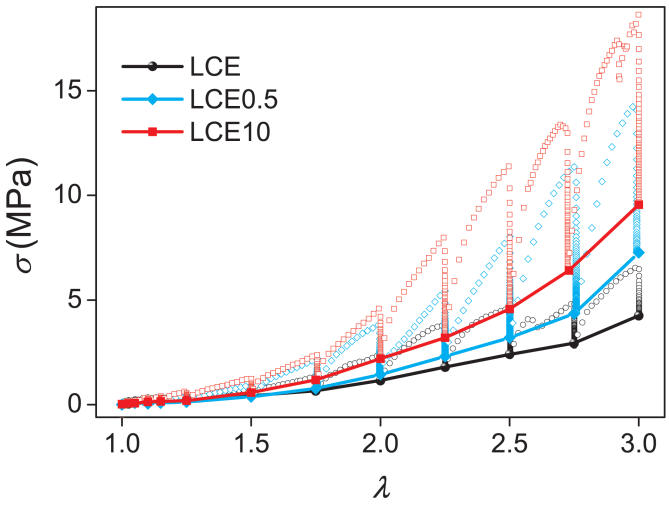

Figure S2. Stress-strain measurements on liquid-crystalline elastomers without NPs (LCE), and liquid-crystalline elastomers with 0.5 wt-% NPs (LCE0.5) and 10 wt-% NPs (LCE10).

[1] E.C. Stoner, E.P. Wohlfarth, *Philos. Trans. R. Soc. A* **1948** *240*, 599-624.

[2] J.M. Haberl, A. Sánchez-Ferrer, A.M. Mihut, H. Dietsch, A.M. Hirt, R. Mezzenga, *Nanoscale*, **2013** *5*, 5539–5548.

[3] J.M. Haberl, A. Sánchez-Ferrer, A.M. Mihut, H. Dietsch, A.M. Hirt, R. Mezzenga, *Adv. Mater.* **2013**, *25*, 1787-1791.

[4] A. Martínez-Gómez, E. Pérez, A. Bello, *Colloid Polym Sci* **2010**, *288*, 859-867.