Electronic Supplementary Material (ESI) for Soft Matter.

Electronic Supplementary Information

In-situ Characterization of Strain-Induced Crystallization of Natural Rubber by Synchrotron Radiation Wide-Angle X-ray Diffraction: Construction of Crystal Network at Low Temperatures

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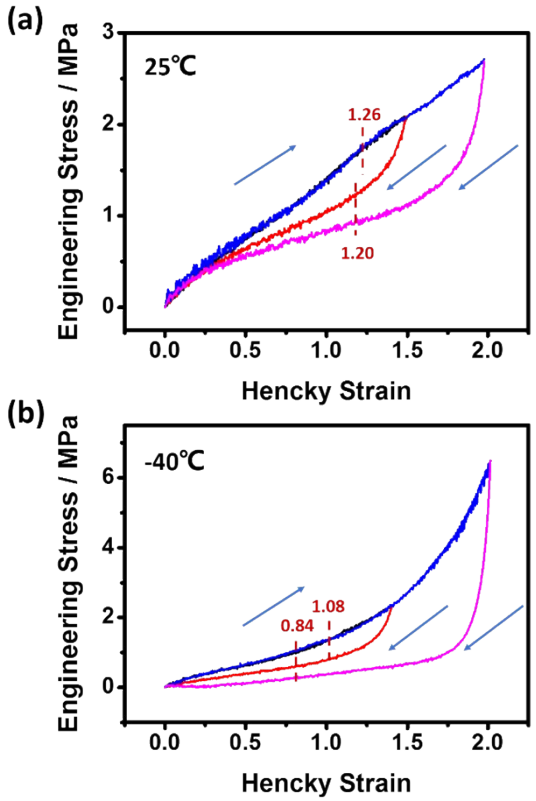

Fig. S1 The stress-strain curves of NR at (a) 25 °C and (b) -40 °C.

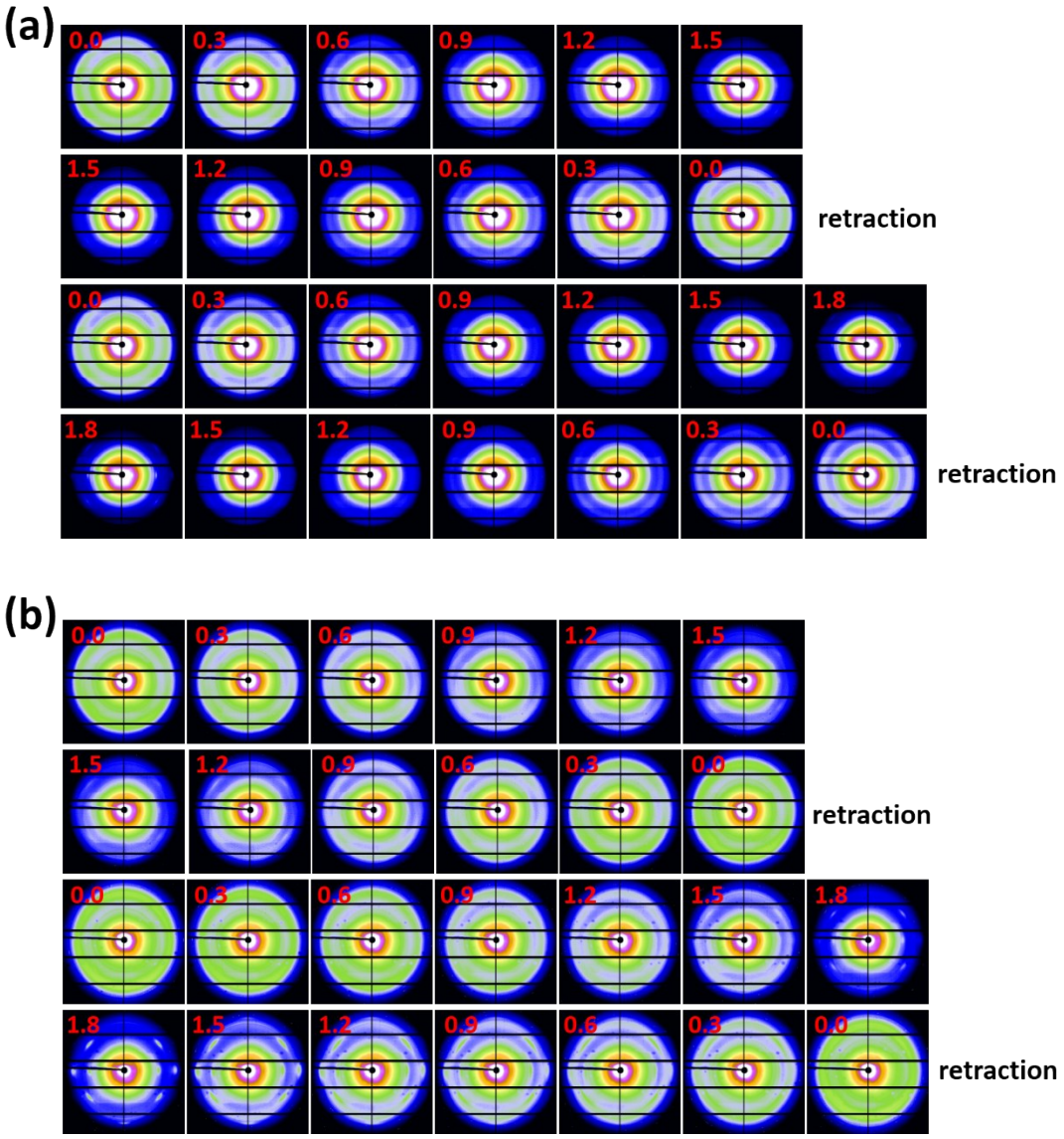

Fig. S2 Sequential evolution of 2D SR-WAXD patterns during deformation-retraction at (a) 25 °C and (b) -40 °C. The tensile direction is vertical and the corresponding Hencky strain values ε are shown at the top left corner of each diffraction pattern.

The strain rate was $0.004s^{-1}$ and the SR-WAXD pattern collecting time was 15s. We choose 25 °C and -40 °C as two typical temperatures in Zone I and Zone II, respectively. The value of ε we choose are 1.5 (region II) and 2.0 (region III)

When retracting at $\varepsilon = 1.5$, the NR crystal disappears at $\varepsilon = 1.26$ at 25 °C and 0.84 at -40 °C. Compared with the onset strain $\varepsilon_{onset} = 1.26$ and 1.08, respectively. The retracting process at $\varepsilon = 2.0$ at 25 °C is similar to that at $\varepsilon = 1.5$. However, the retracting process at $\varepsilon = 2.0$ at -40 °C is totally different. The stress reduces sharply, and the NR

crystal exists during the whole retraction. It suggests that the crystal network nearly have no elasticity in that this network is hard to disappear even there exists no external force.

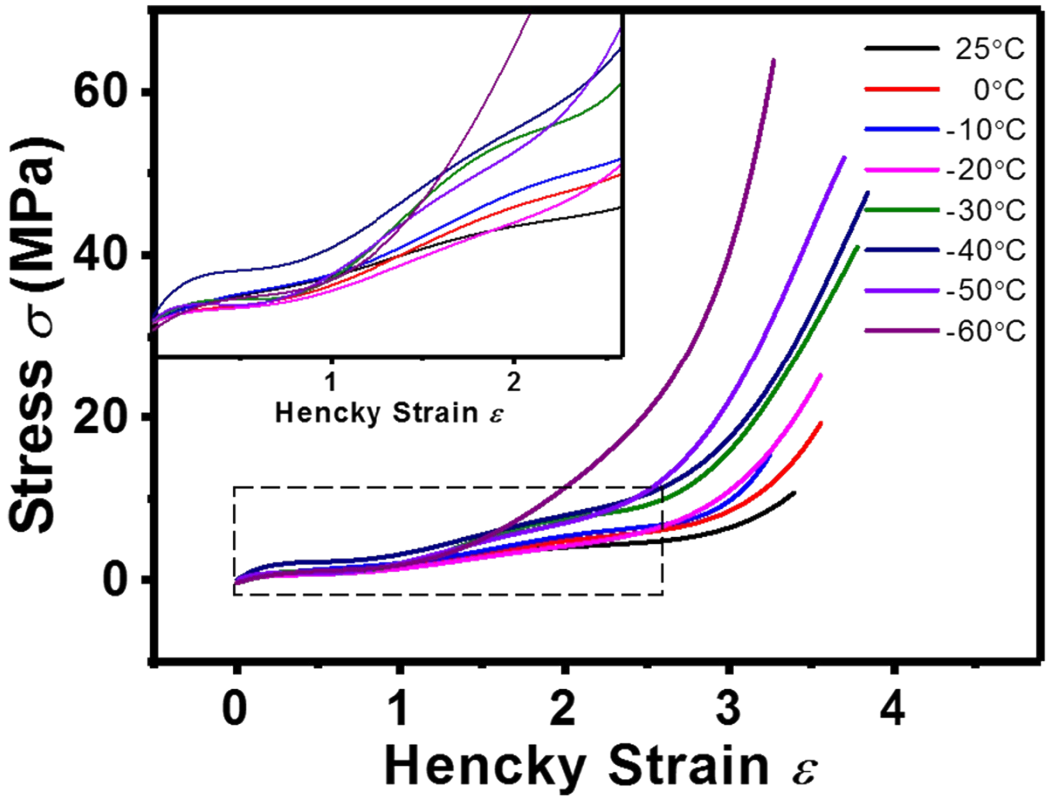

Fig. S3 The stress-strain curves of NR at various temperatures (stress represents the engineering stress). Magnified **Fig. 4**.

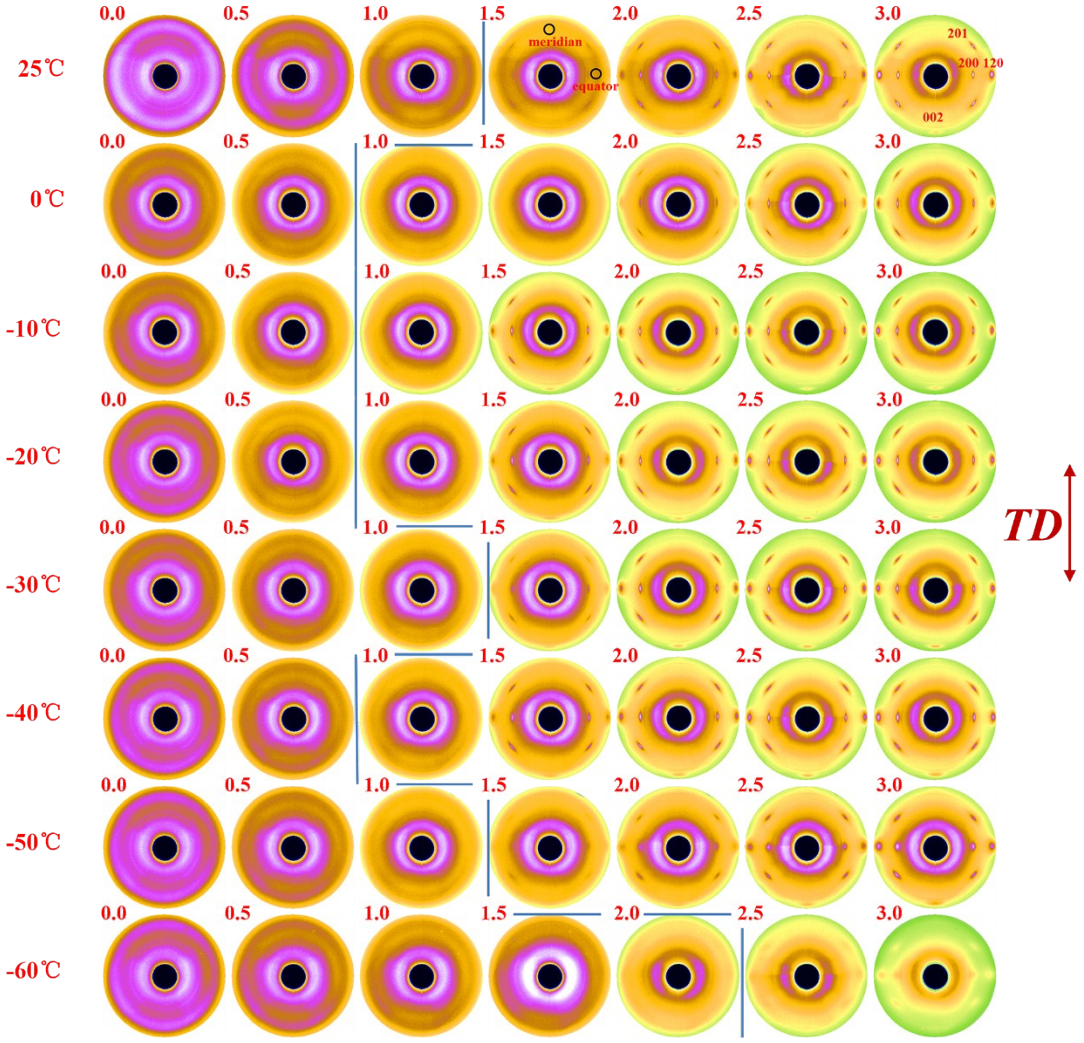

Fig. S4 Sequential evolution of 2D SR-WAXD patterns during uniaxial stretching at different temperatures. The tensile direction is vertical as shown by the double arrow, and the corresponding Hencky strain values ε are shown at the top left corner of each diffraction pattern. Magnified **Fig. 6**.