Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Unusual Strain-Dependent Thermal Conductivity Modulation of Silver Nanoflower-Polyurethane Fibers

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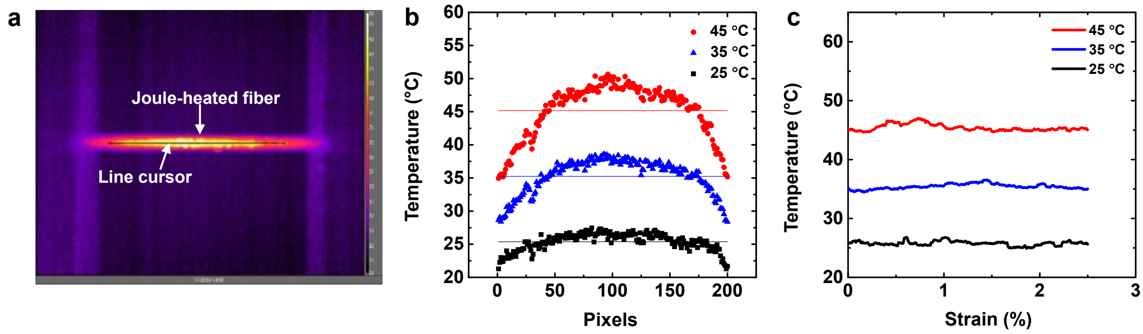

Figure S1. Temperature distribution of Joule-heated Ag-PU fibers during stretching (0-2.5% strain). (a) An infrared image of Joule-heated fiber. The mean temperature was 45 °C, which was obtained by averaging the temperatures along the line cursor at the center of the fiber. (b) Temperature profiles of Joule-heated fibers at different mean temperatures (25, 35, and 45 °C). (c) Real-time mean temperature monitoring of Joule-heated fibers during stretching (0-2.5% strain).

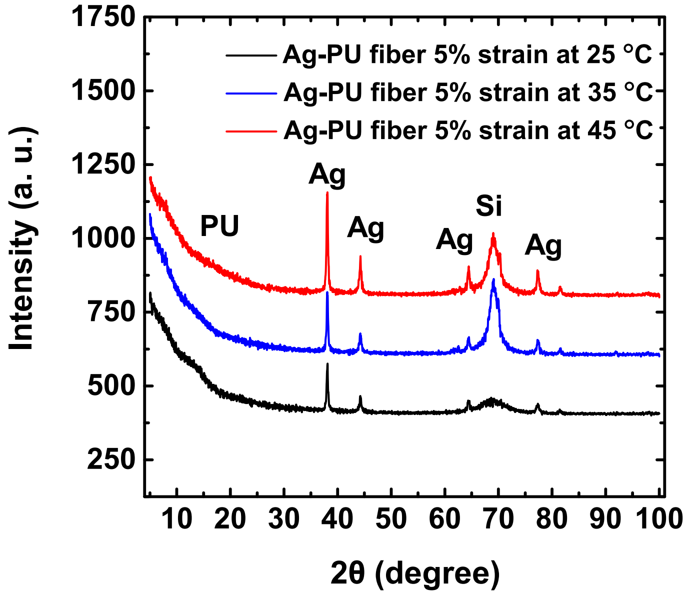

Figure S2. XRD patterns of stretched Ag-PU fibers (5% strain) at 25, 35, and 45 °C.

Steady-state 1-dimensional Fourier's conduction model

The thermal conductivity (κ) of an Ag-PU fiber was obtained using the steady-state 1-dimensional Fourier's conduction equation.[S1,S2] The Ag-PU fiber was regarded as a 1-dimensional object due to the small diameter and large length. The convective heat loss was neglected since the experiment was carried out in vacuum.

$$\kappa\frac{d^2T}{dx^2} + q = 0 \tag{S1}$$

The volumetric Joule heat generation is given by $q = UI/2A_cL$, where U is the voltage drop across the fiber, and I is the supplied current. A_c and L are the cross-sectional area and half length of the fiber.

The boundary condition is as follows.[S1,S2] The temperature gradient (dT/dx) is zero at the center ($x = 0$) due to symmetry (Fig. 2c), and the temperature at the end ($x = L$) is T_{end} (Fig. 2c). The integration of Eq. S1 led to the quadratic equation.[S1,S2]

$$T_x = T_{end} + \frac{q}{2k}(L^2 - x^2) \tag{S2}$$

Finally, thermal conductivity was calculated using Eq. S3 where T_{center} was temperature at the center ($x = 0$).[S1,S2]

$$\kappa = \frac{L}{4A_c}\cdot\frac{UI}{(T_{center} - T_{end})} \tag{S3}$$

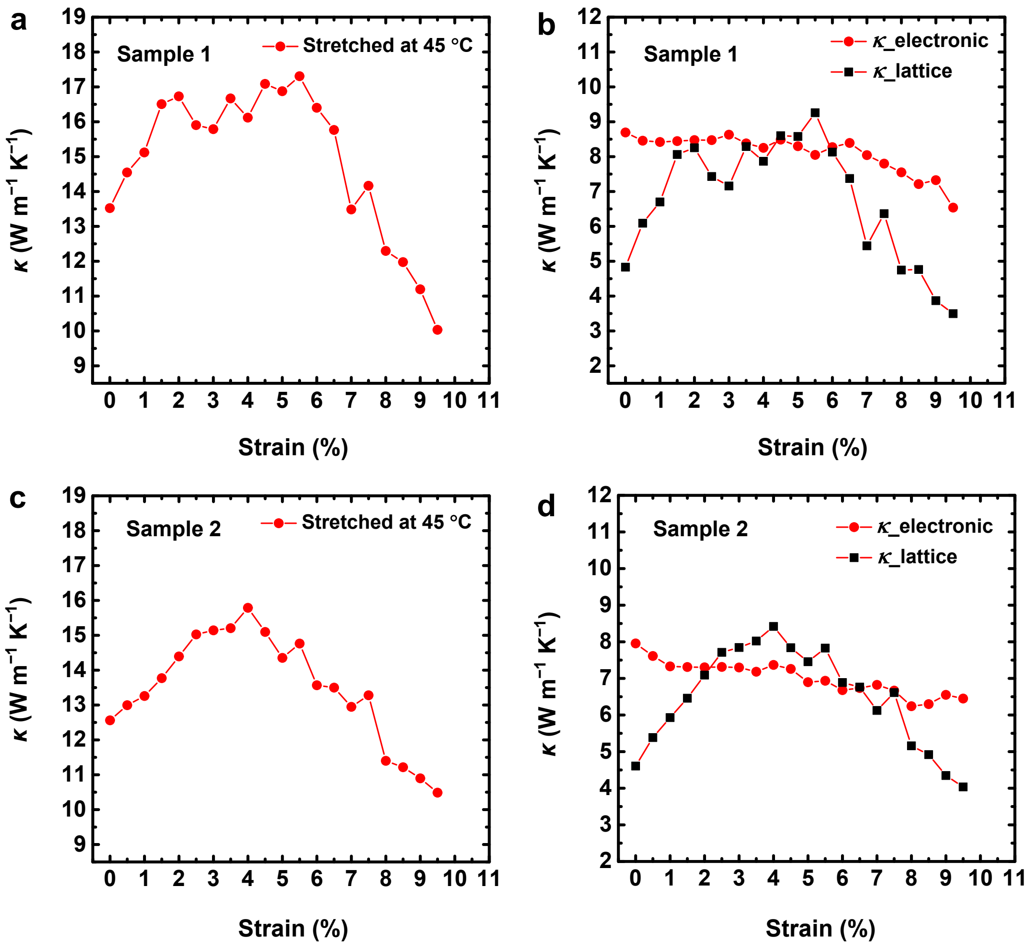

Figure S3. Control experiments were carried out using two new Ag-PU fibers, and the strain was increased from 0 to 9.5 % with an interval of 0.5 %. The fibers were strained at 45 °C. (a, b) The experimentally-measured total thermal conductivity and Wiedemann–Franz law analysis (electronic and lattice thermal conductivity) of sample 1. (c, d) The experimentally-measured total thermal conductivity and Wiedemann–Franz law analysis (electronic and lattice thermal conductivity) of sample 2.

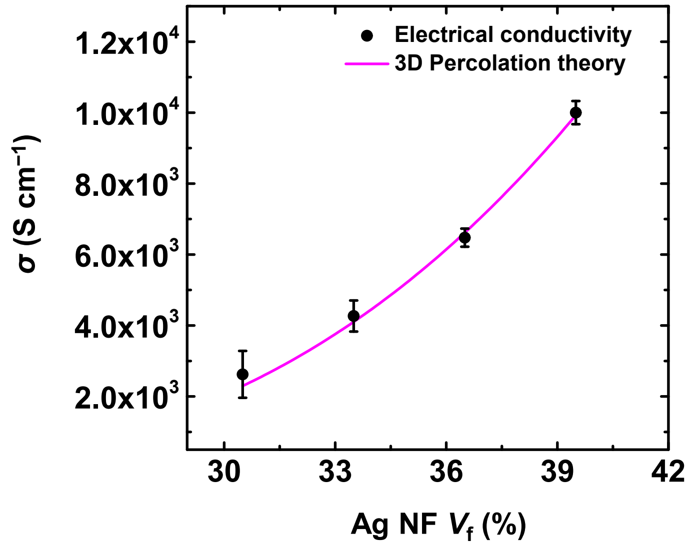

Figure S4. The effect of Ag NF concentration on the electrical conductivity of Ag-PU fibers. The 3D percolation theory prediction is also shown.

The electrical conductivity of polymer composites can be estimated using the power law relationship.[S3–S5]

$$\sigma = \sigma_o\left(V_f - V_c\right)^s \qquad \text{(S4)}$$

where σ is the electrical conductivity of Ag-PU fiber, σ_o is the conductivity of bulk silver, V_f is volumetric fraction of Ag NFs, V_c is the volumetric fraction at percolation threshold, and s is the fitting exponent. V_c (0.18) and s (2.7) were obtained by fitting the experimental data with the theory (Fig. S4). The volume of Ag NFs is fixed in the PU matrix, whereas the volume of matrix changes upon increasing strain (ϵ).[S5] This lead to a change in V_f which can be mathematically described using Poisson's ratio (v).[S5]

$$v = \frac{\left(D_1 - D_2\right)/D_1}{\left(L_2 - L_1\right)/L_1} = \frac{D_1 - D_2}{D_1\epsilon} \qquad \text{(S5)}$$

$$D_2 = D_1 - v\epsilon D_1 = D_1(1 - v\epsilon) \qquad \text{(S6)}$$

$$L_2 = L_1 + L_1\epsilon \tag{S7}$$

$$V_{matrix} = \pi\left(\frac{D_2}{2}\right)^2 L_2 = \pi\left(\frac{D_1}{2}\right)^2 L_1(1 - v\epsilon)^2(1 + \epsilon) \tag{S8}$$

where L_1 and D_1 are the initial length and diameter of the fiber, and L_2 and D_2 are the length and diameter of the fiber after stretching. Finally, V_f was calculated using Eq. S9,[S5] with 39.5 vol% of Ag NFs.

$$V_f = \frac{V_{silver}}{V_{matrix}} = \frac{0.395}{(1 - v\epsilon)^2(1 + \epsilon)} \tag{S9}$$

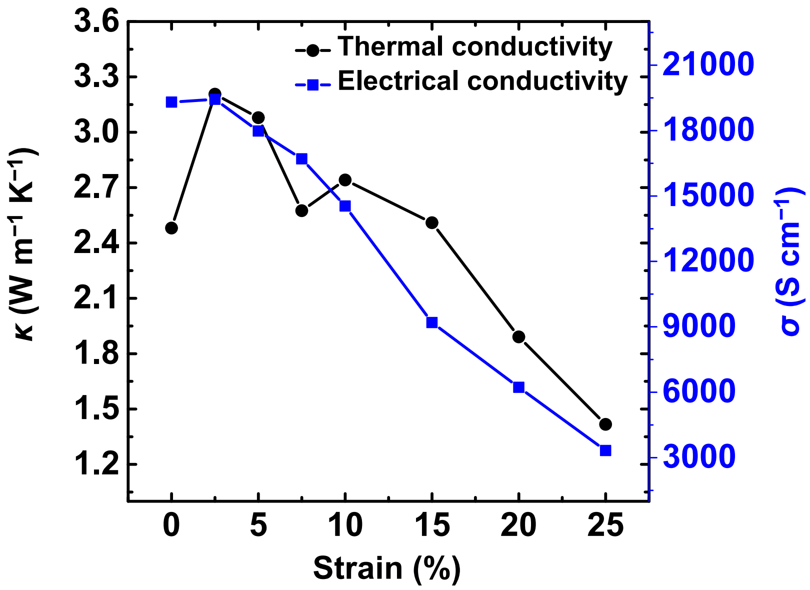

Figure S5. Strain-dependent thermal and electrical conductivity of the spherical Ag nanoparticle-polyurethane fiber. The mean temperature during stretching was 45 °C.

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