

Supporting Information:

**Transparent and hybrid multilayer films with improved
thermoelectric performance by chalcogenide-interlayer-induced
transport enhancement**

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Supporting Information Contents:

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1. Experimental Methods

1.1. Fabrication of PEDOT-SnSeTe nanosheets

Solid-state reaction, hydrothermal lithium (Li) intercalation, and subsequent exfoliation were performed to fabricate the SnSeTe nanosheets, according to the previously reported experimental method with the stoichiometric ratio of Sn, Se, and Te (1:0.97:0.03).^{S1} The resulting SnSeTe nanosheets (0.5 g) and deionized water (100 mL) were added into a beaker. Dodecylbenzenesulfonic acid (DBSA) (0.032 g) was then added into the same beaker. A black colloidal solution was obtained upon stirring at 30 °C for 1 h. 3,4-ethylenedioxythiophene (EDOT) (0.047 g) was added into the beaker to prepare the PEDOT-SnSeTe nanosheet sample, which was further stirred at 303 K for 1 h. To carry out the polymerization of EDOT, another solution containing ammonium persulfate (0.182 g) and deionized water (10 mL) was added to the colloidal solution, which was stirred at 303 K for 24 h. It was then centrifuged at 8000 rpm for 30 min and redispersed in deionized water. After several centrifugation and redispersion cycles, the PEDOT-SnSeTe nanosheets were finally obtained.

1.2. Fabrication of multilayer films

The PEDOT-SnSeTe nanosheets (0.03 g) were dispersed in 30 mL of chloroform, and

then sonicated for 30 min. The PEDOT:PSS solution (PH 1000, Heraeus Clevios GmbH) was mixed with 5 vol.% of DMSO, filtered through a syringe filter (pore size of 0.45 μm), and further sonicated for 2 h. It was then dropped onto a square glass substrate ($2 \times 2 \text{ cm}^2$) and spin-coated at 2000 rpm for 120 s. The film was heated at 413 K for 10 min to remove the residual solvent, following which the solution of PEDOT-SnSeTe nanosheets was further spin-coated under the same conditions on the deposited PEDOT:PSS film, followed by another PEDOT:PSS spin-coating to obtain a PEDOT:PSS/PEDOT-SnSeTe nanosheets/PEDOT:PSS (PSP) multilayer, which was heated at 413 K for 10 min. Different spin speeds were employed for the film deposition yielding PSP- x multilayer films (where x indicates the spin-coating speed of the PSP multilayer film; $x = 1000, 2000, 3000, 4000$, and 5000 rpm).

1.3. Characterization

The binding energies of the materials were investigated by X-ray photoelectron spectroscopy (XPS, Thermo UK K-alpha) with a monochromated Al K_α radiation (1486.6 eV). The four-point probe method with disk-shaped compressed pellets was used to measure the σ values of the composites. A homemade device containing a pair of thermocouples and voltmeters was used to measure S . X-ray diffraction (XRD, New D8 Advance, Bruker AXS) was performed at 40 mA, 40 kV, and scan rate of $1^\circ \cdot \text{s}^{-1}$ with Cu K_α radiation ($\lambda = 0.154056 \text{ nm}$) to characterize the crystal structures of the materials. A thermogravimetric analysis (TGA, TGA-2050, TA Instruments) was carried out to analyze the thermal degradations of the samples under an N_2 atmosphere at a heating rate of $10 \text{ K} \cdot \text{min}^{-1}$. The morphologies and microstructures of the materials were determined using field-emission scanning electron microscopy (FE-SEM, SIGMA, Carl Zeiss) and field-emission transmission electron microscopy (FE-TEM, JEM-2100F, JEOL), respectively. The charge carrier concentration (n) was determined by a Hall

effect measurement using the Van der Pauw four-point probe configuration (HMS-3000, Ecopia) at 300 K. The carrier mobility (μ) was calculated using the measured σ and n values. Five samples of each product were fabricated to evaluate the reproducibility of the experiments, carried out at 300 K.

References

- S1. Ju, H.; Park, D.; Kim, J., Solution-Processable Flexible Thermoelectric Composite Films Based on Conductive Polymer/SnSe_{0.8}S_{0.2} Nanosheets/Carbon Nanotubes for Wearable Electronic Applications. *J. Mater. Chem. A* **2018**, *6*, 5627–5634.

2. Figures

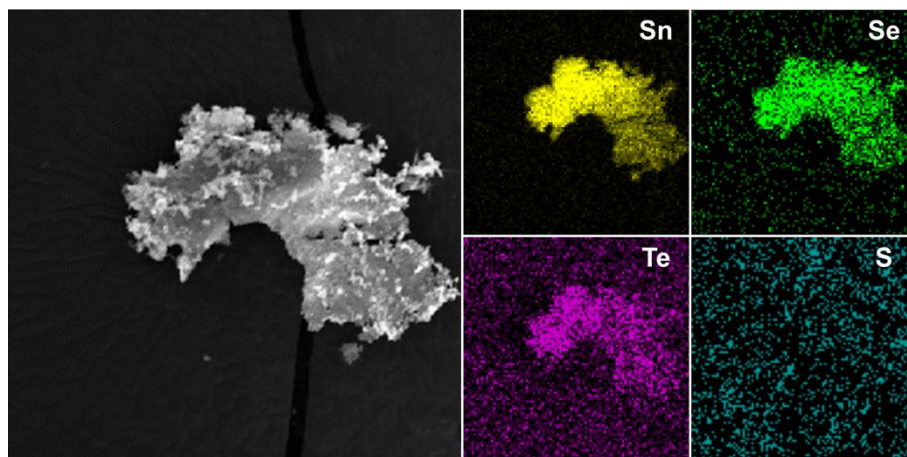


Figure S1. FE-SEM image and corresponding EDS maps of the SnSeTe nanosheets.

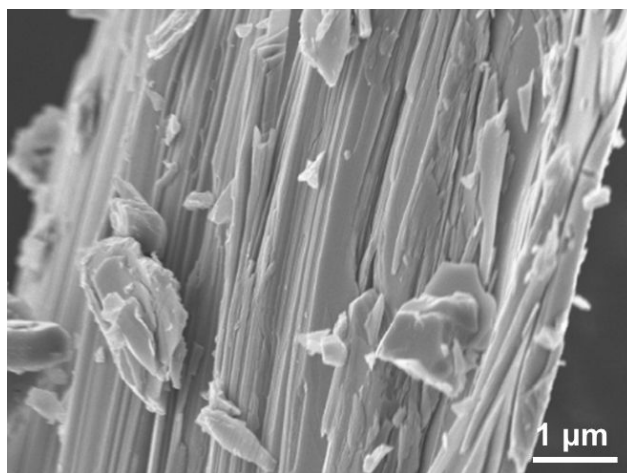


Figure S2. FE-SEM image of the SnSeTe bulk material, revealing the intrinsic layered structure.

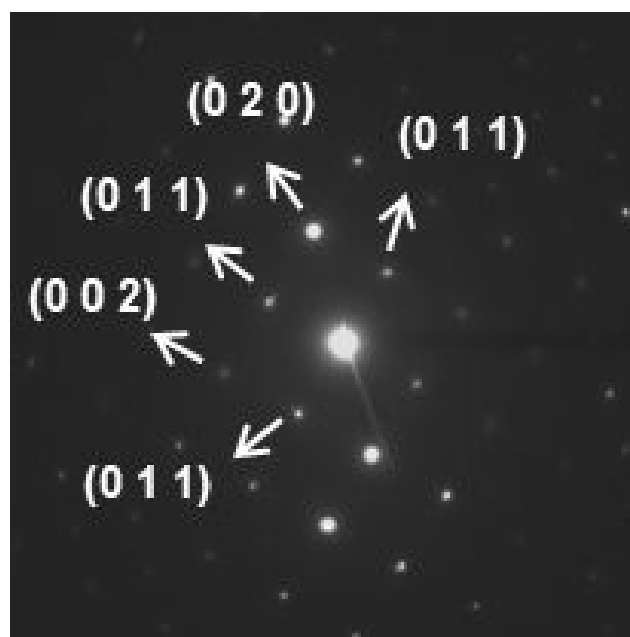


Figure S3. Selected-area electron diffraction pattern of the SnSeTe nanosheet.

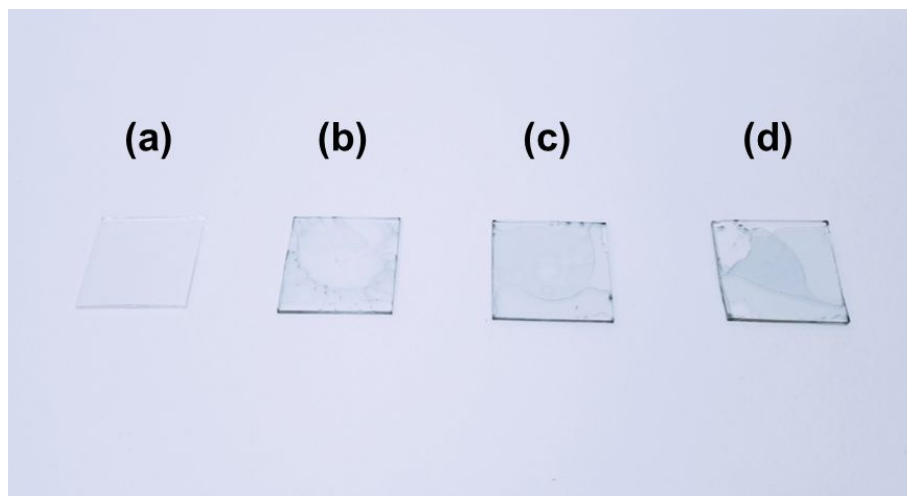


Figure S4. Digital photographs of the (a) glass substrate, (b) PEDOT:PSS layer, (c) PEDOT:PSS/PEDOT-SnSeTe nanosheet (PS) bilayer, and (d) PSP multilayer.

3. Tables

Films	n (cm ⁻³)	μ (cm ² /V·s)
PEDOT:PSS	7.869×10 ²²	0.051
PS bilayer	1.749×10 ²⁰	2.32
PSP multilayer	4.133×10 ²⁰	1.48

Table S1. Carrier concentration and mobility values of the PEDOT:PSS, PS bilayer, and PSP multilayer films.

RPM	n (cm ⁻³)	μ (cm ² /V·s)
1000	5.269×10 ²⁰	1.232
2000	4.133×10 ²⁰	1.48
3000	3.398×10 ²⁰	1.69
4000	2.551×10 ²⁰	2.08
5000	2.09×10 ²⁰	2.21

Table S2. Carrier transport properties of the PSP multilayer films obtained at different spin-coating speeds (rpm).