

## Supplementary information

### Towards the practical realization of high-performance Ag<sub>2</sub>Se-based thermoelectric coolers

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## I. Setting parameters for the finite element simulation of Ag<sub>2</sub>Se-based cooler

Table S1. Finite element simulation setting parameters.

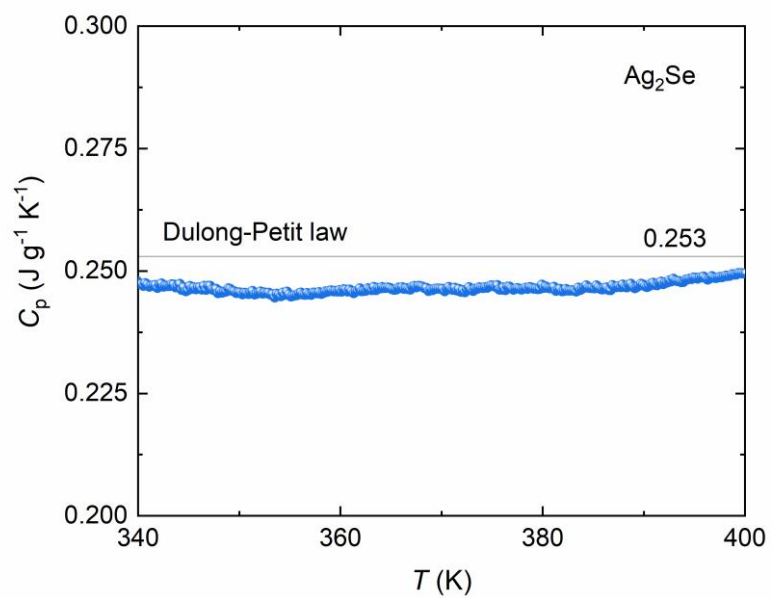
Parameter	Value
Cross-sectional area of the Ag <sub>2</sub> Se legs	$1.8 \times 1.8 \text{ mm}^2$
Height of the Ag <sub>2</sub> Se legs	2.5 mm
Cross-sectional area of the (Bi, Sb) <sub>2</sub> Te <sub>3</sub> legs	$2.0 \times 2.0 \text{ mm}^2$
Height of the (Bi, Sb) <sub>2</sub> Te <sub>3</sub> legs	2.5 mm
Thickness of copper electrode	0.065 mm
Thickness of Al <sub>2</sub> O <sub>3</sub> substrate	0.38 mm
The area of the Al <sub>2</sub> O <sub>3</sub> substrate of the cold side	$13.0 \times 13.0 \text{ mm}^2$
The area of the Al <sub>2</sub> O <sub>3</sub> substrate of the hot side	$13.0 \times 16.0 \text{ mm}^2$
Thermal conductivity of the substrate	$20 \text{ W m}^{-1} \text{ K}^{-1}$
Thermal conductivity of the thermal grease	$4.8 \text{ W m}^{-1} \text{ K}^{-1}$
Heat load	0.0-2.5 W
Hot-side temperature	300 K
Contact resistivity of Ag <sub>2</sub> Se legs	$5.0 \mu\Omega \text{ cm}^2$
Contact resistivity of (Bi, Sb) <sub>2</sub> Te <sub>3</sub> legs	$5.0 \mu\Omega \text{ cm}^2$

## 2. Sample density of Ag<sub>2</sub>Se materials with different sizes

**Table S2.** Sample density of Ag<sub>2</sub>Se materials with different sizes

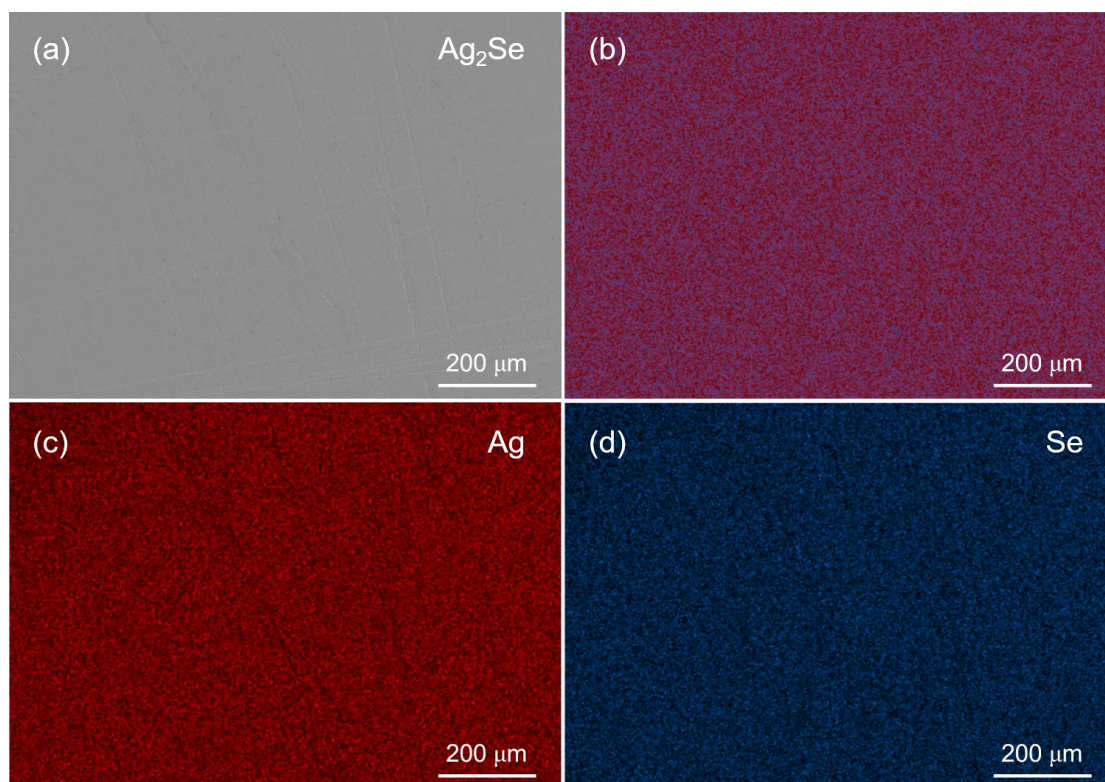
Sample diameter (mm)	Measured density (g cm <sup>-3</sup> )	Relative density (%)
10.0	8.05	98.17
12.7	8.07	98.41
20.0	8.03	97.93
25.4	8.06	98.29

### 3. Specific heat capacity of the $\text{Ag}_2\text{Se}$ sample



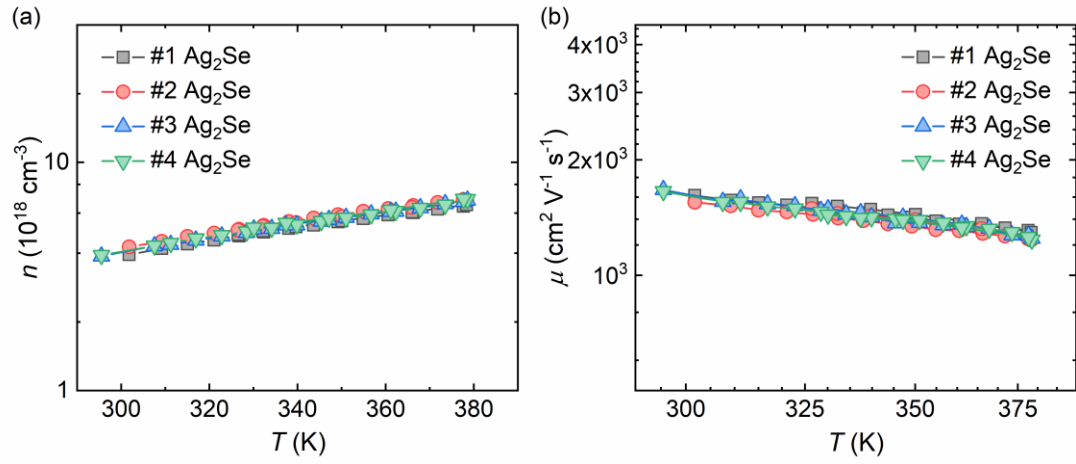
**Figure S1.** Specific heat capacity of the  $\text{Ag}_2\text{Se}$  sample.

#### 4. Energy dispersive spectroscopy mapping of the $\text{Ag}_2\text{Se}$ sample



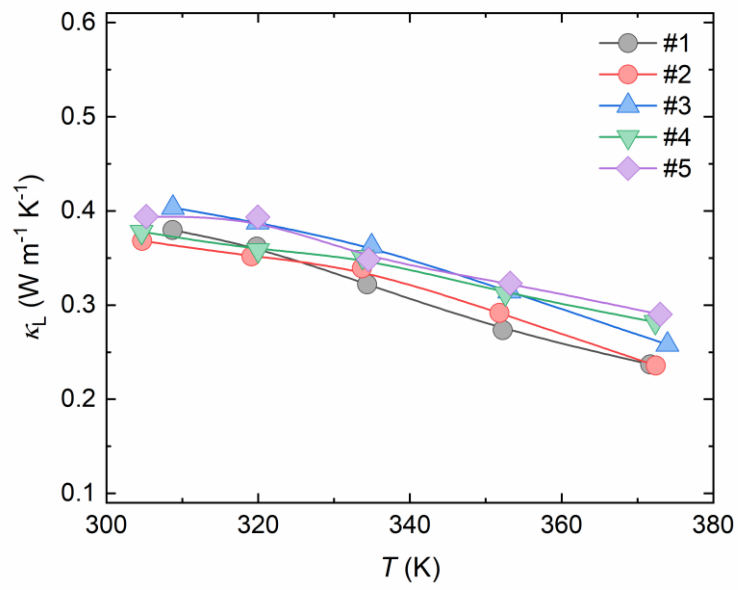
**Figure S2.** (a) Surface morphology, and (b-d) energy dispersive spectroscopy mapping of the  $\text{Ag}_2\text{Se}$  sample.

## 5. Hall carrier concentration and mobility of the $\text{Ag}_2\text{Se}$ sample



**Figure S3. Hall carrier concentration and mobility of the  $\text{Ag}_2\text{Se}$  sample.** (a) Carrier concentration. (b) Carrier mobility.

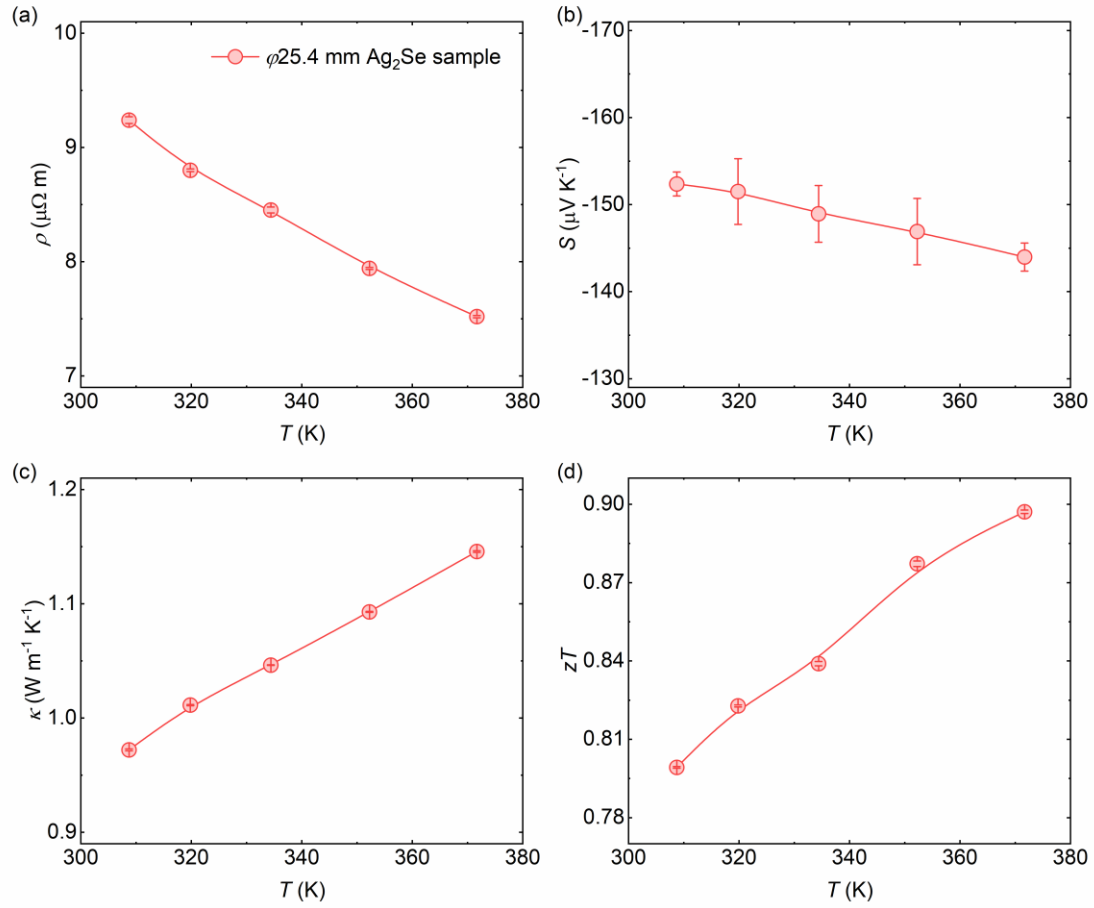
## 6. The lattice thermal conductivity of the Ag<sub>2</sub>Se sample



**Figure S4.** The lattice thermal conductivity of the Ag<sub>2</sub>Se sample.

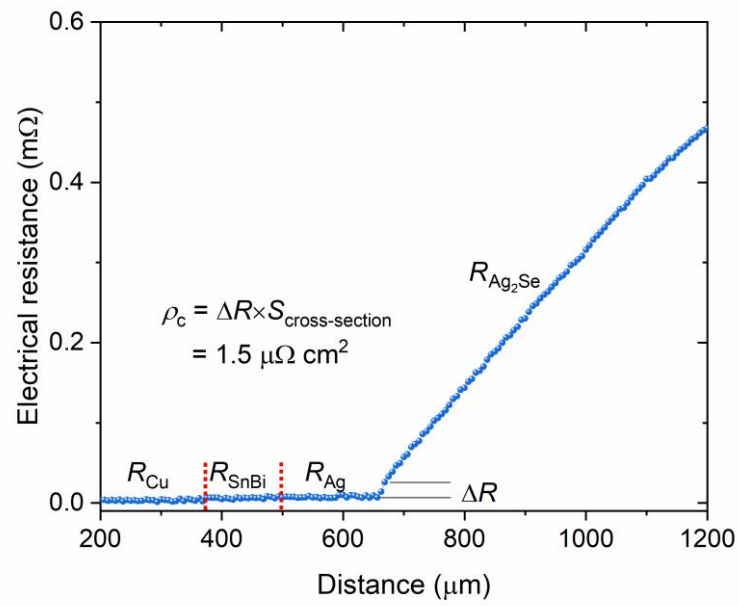


## 7. Thermoelectric properties of the $\text{Ag}_2\text{Se}$ samples



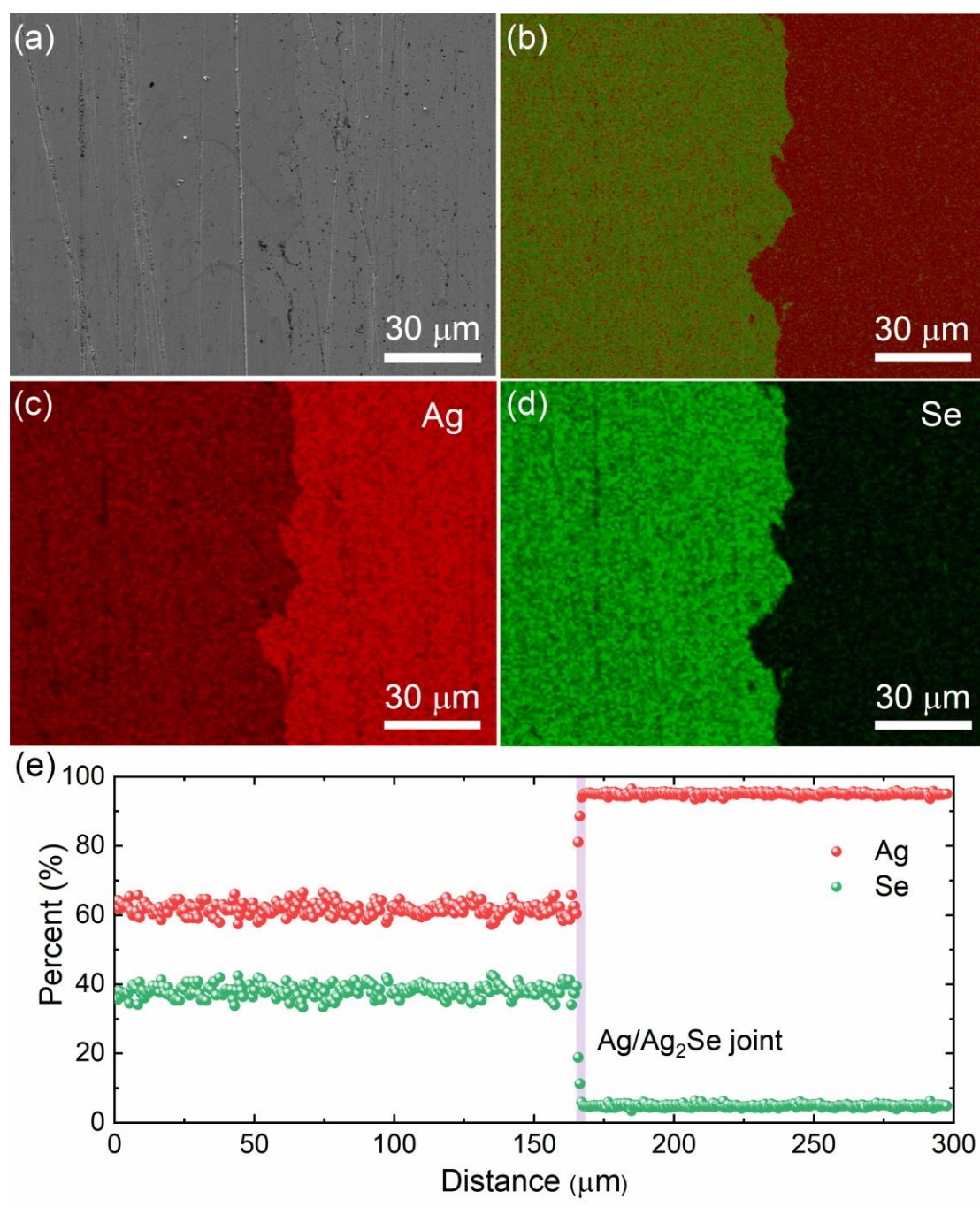
**Figure S5. Thermoelectric properties of the  $\text{Ag}_2\text{Se}$  samples.** Temperature-dependent (a) electrical resistivity, (b) Seebeck coefficient, (c) thermal conductivity, and (d)  $zT$  values.

### 8. Contact resistivity of the soldered Ag/Ag<sub>2</sub>Se joint



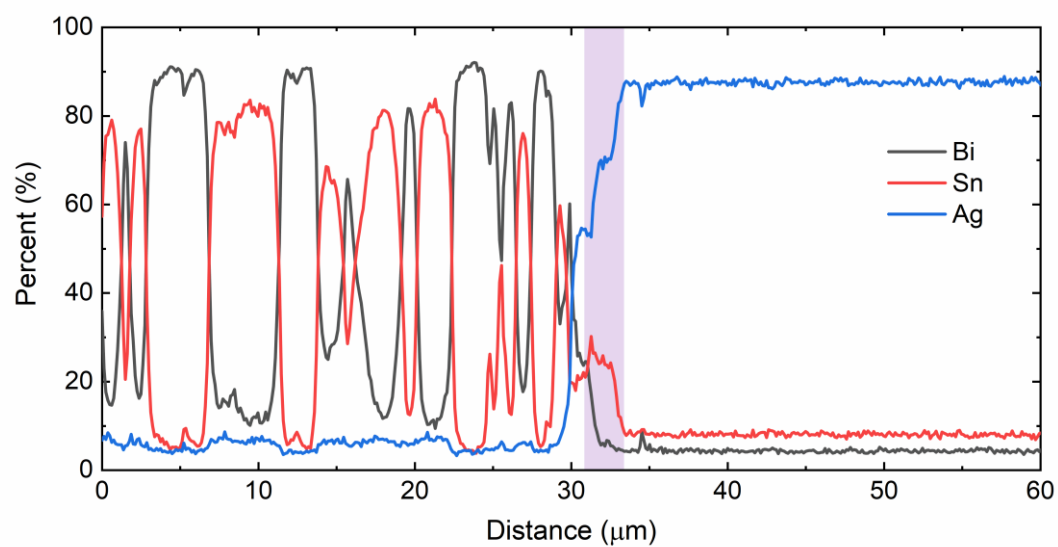
**Figure S6.** Contact resistivity of the soldered Ag/Ag<sub>2</sub>Se joint.

## 9. Energy dispersive spectroscopy and linear scanning of the Ag/Ag<sub>2</sub>Se interface



**Figure S7.** (a) The Ag/Ag<sub>2</sub>Se interface and (b-d) corresponding EDS mapping for the interface. (e) Linear EDS scanning across the Ag/Ag<sub>2</sub>Se interface.

## 10. Linear energy dispersive spectroscopy scanning of the Ag/SnBi interface



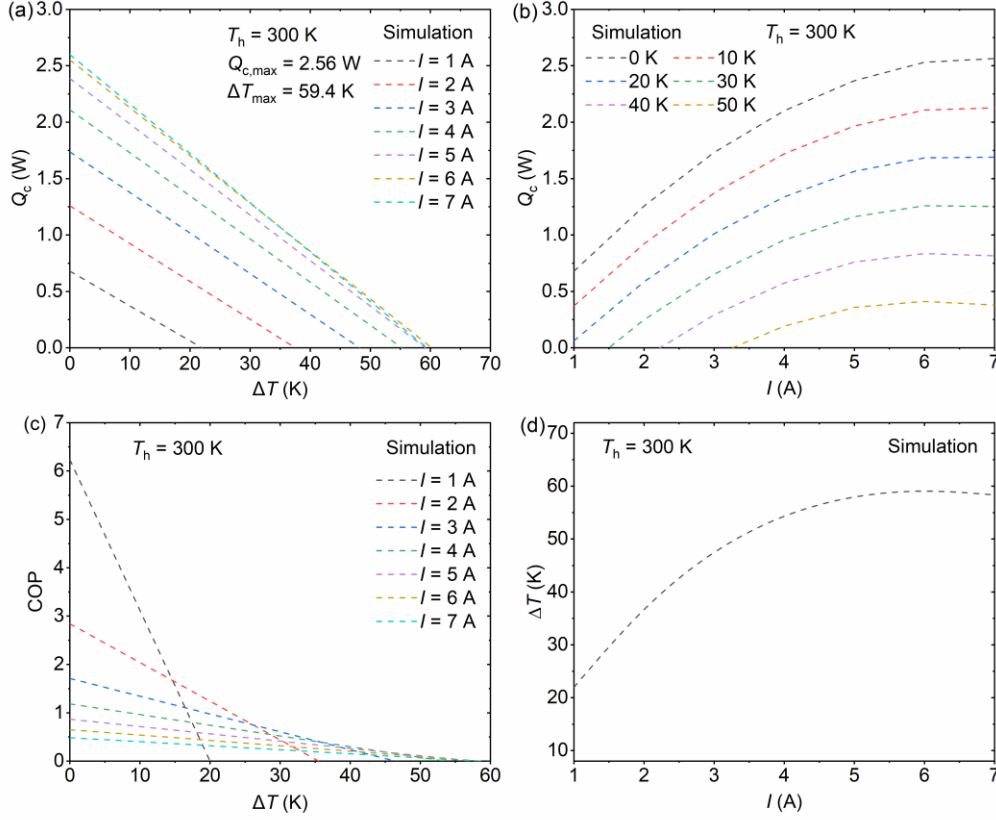
**Figure S8.** Linear EDS scanning across the Ag/SnBi solder interface.

# 11. Comparison of thermoelectric cooling performance for Ag<sub>2</sub>Se, Bi<sub>2</sub>Te<sub>3</sub>, and Mg<sub>3</sub>(Sb, Bi)<sub>2</sub>-based devices at the hot-side temperature of 300 K

**Table S3.** Comparison of thermoelectric cooling performance for Ag<sub>2</sub>Se, Bi<sub>2</sub>Te<sub>3</sub>, and Mg<sub>3</sub>(Sb, Bi)<sub>2</sub>-based devices at the hot-side temperature of 300 K

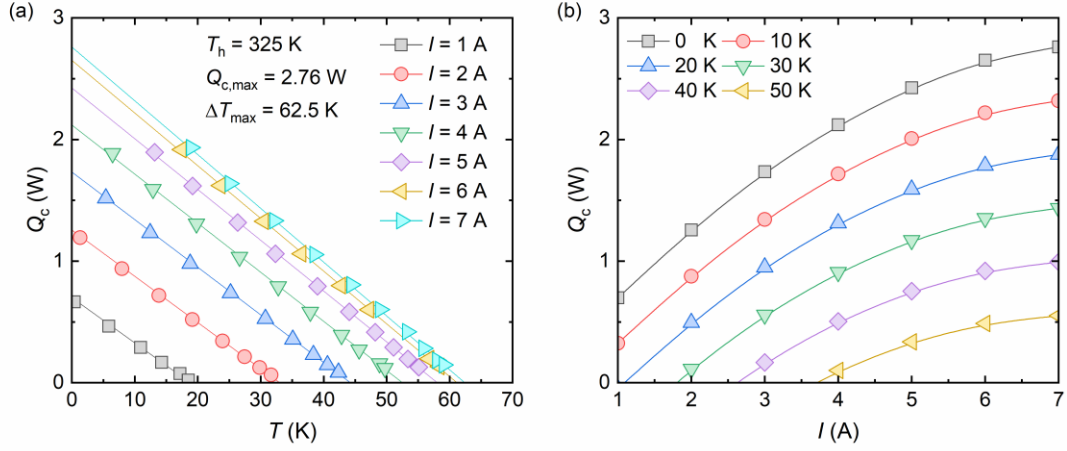
n-type leg	p-type leg	Cooling temperature difference (K)	Cooling power density (W cm <sup>-2</sup> )	Reference
Ag <sub>2</sub> Se	Bi <sub>2</sub> Te <sub>3</sub> alloys	55.4	1.5	This work
Ag <sub>2</sub> Se	Bi <sub>2</sub> Te <sub>3</sub> alloys	57.7	1.5	Jiang <i>et al.</i> <sup>[1]</sup>
Ag <sub>2</sub> Se	Bi <sub>2</sub> Te <sub>3</sub> alloys	56.0	1.4	Liu <i>et al.</i> <sup>[2]</sup>
Ag <sub>2</sub> Se	MgAgSb	52.0	0.8	Zhao <i>et al.</i> <sup>[3]</sup>
Bi <sub>2</sub> Te <sub>3</sub>	Bi <sub>2</sub> Te <sub>3</sub> alloys	70.1	1.6	Sun <i>et al.</i> <sup>[4]</sup>
Bi <sub>2</sub> Te <sub>3</sub>	Bi <sub>2</sub> Te <sub>3</sub> alloys	73.9	2.2	Zhao <i>et al.</i> <sup>[5]</sup>
Mg <sub>3</sub> (Sb, Bi) <sub>2</sub>	Bi <sub>2</sub> Te <sub>3</sub> alloys	69.0	1.3	Ma <i>et al.</i> <sup>[6]</sup>
Mg <sub>3</sub> (Sb, Bi) <sub>2</sub>	MgAgSb	52.0	0.8	Xie <i>et al.</i> <sup>[7]</sup>
Mg <sub>3</sub> (Sb, Bi) <sub>2</sub>	MgAgSb	61.0	-	Zhang <i>et al.</i> <sup>[8]</sup>

## 12. The simulated cooling performance of the 7 pairs $\text{Ag}_2\text{Se}/(\text{Bi}, \text{Sb})_2\text{Te}_3$ thermoelectric cooler

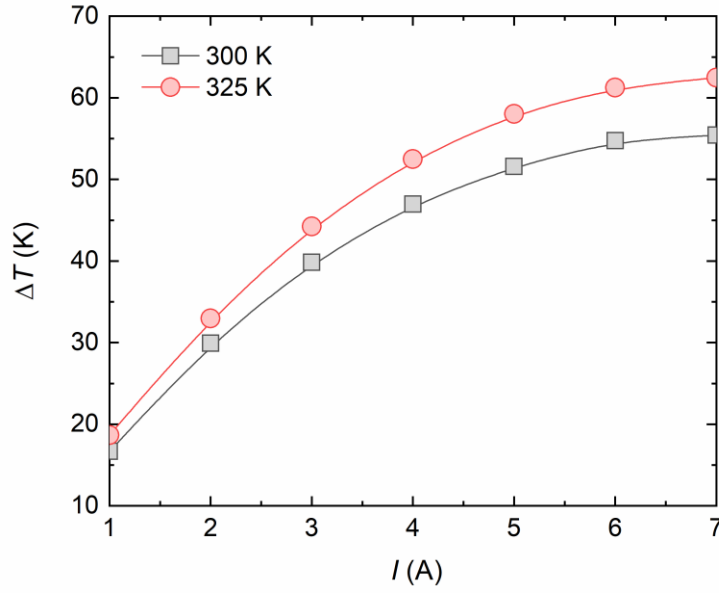


**Figure S9. The simulated cooling performance of the 7 pairs  $\text{Ag}_2\text{Se}/(\text{Bi}, \text{Sb})_2\text{Te}_3$  thermoelectric cooler.** Cooling power as a function of (a) the temperature difference at different electrical currents and (b) electrical current at different temperature differences. (c) The coefficient of performance as a function of the temperature difference at different electrical currents. (d) The maximum cooling temperature difference at different electrical currents.

### 13. Thermoelectric cooling performance of the Ag<sub>2</sub>Se-based cooler at the hot-side temperature of 325 K

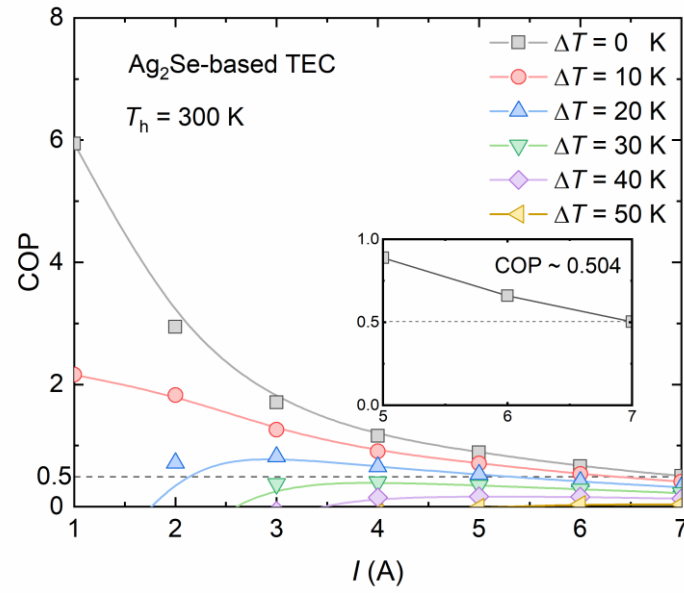


**Figure S10.** Cooling power as a function of (a) cooling temperature difference at different electrical currents, and (b) electrical current at different cooling temperature differences.



**Figure S11.** The relationship between cooling temperature difference and electrical current at the hot-side temperature of 300 and 325 K.

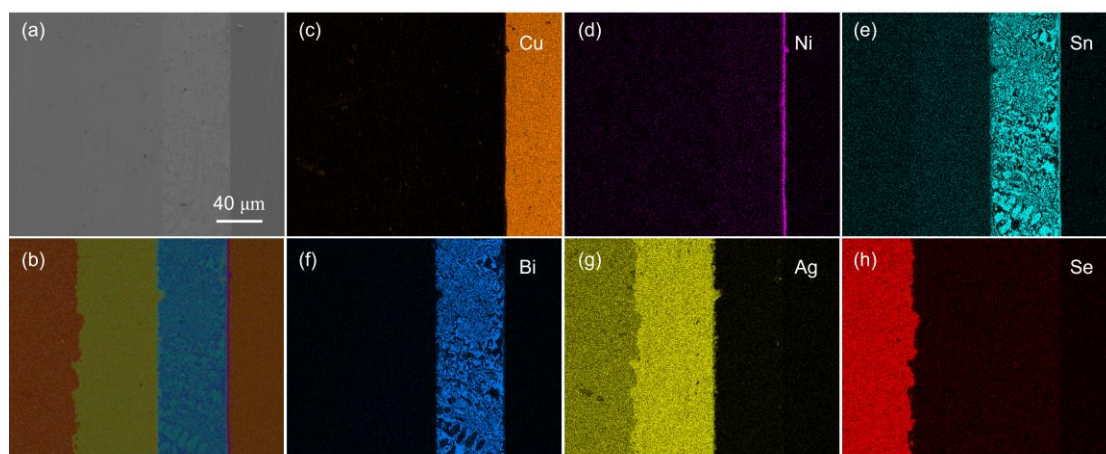
**14. Coefficient of performance as a function of the electrical current of the  $\text{Ag}_2\text{Se}$ -based thermoelectric cooler**



**Figure S12.** Coefficient of performance as a function of the electrical current of the  $\text{Ag}_2\text{Se}$ -based thermoelectric cooler at the hot-side temperature of 300 K.



## 15. Interfacial properties of the $\text{Ag}_2\text{Se}$ -based thermoelectric cooler after cycling



**Figure S13.** (a) Interfacial morphology, and (b-h) energy dispersive spectroscopy mapping of the interface of the  $\text{Ag}_2\text{Se}$ -based thermoelectric cooler.

## References:

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