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## **Journal Name**

## ARTICLE

## **Electronic Supplementary Information**

Synergetic optimization of electronic and thermal transport for high-performance thermoelectric GeSe-AgSbTe<sub>2</sub> alloy

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Fig. S1 (a) HRTEM image and (b) SAED pattern of  $GeSeAg_{0.2}Sb_{0.2}Te_{0.4}$ .



Fig. S2 SEM images of pure GeSe (a) and GeSe-20%AgSbTe<sub>2</sub> (b).



**Fig. S3** Thermoelectric properties of GeSe-15%AgSbTe<sub>2</sub> along the directions perpendicular (IP) and parallel (CP) to the pressing direction.







Fig. S5 Comparison of power factor (a) and thermal conductivity ( $\kappa_{tot}$ ,  $\kappa_{lat}$ ) (b) of GeSeAg<sub>0.2</sub>Sb<sub>0.2</sub>Te<sub>0.4</sub> and GeSeAg<sub>0.2</sub>Sb<sub>0.2</sub>Se<sub>0.4</sub>.





Fig. S6 Power factor of  $GeSeAg_{0.2}Sb_{0.2}Te_{0.4}$  during two-cycle heating-cooling processes.



Fig. S7 Temperature dependence of heat capacity (a) and  $^{f}$  thermal diffusion coefficients (b) of GeSe-xAgSbTe<sub>2</sub>.

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