Exceptional Thermoelectric Performance of a "Star-Like" SnSe Nanotube with Ultra-Low Thermal Conductivity and a High Power Factor

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I. Optimization of the tube structure

The initial structure of the (12, 0) nanotube is obtained by rolling twelve $[Sn_2Se_2]$ unit of SnSe layer along the zigzag direction (**Figure S1a**). Then the nanotube was firstly optimized with the Hellmann-Feynman force converge threshold of 0.01eV/Å. (**Figure S1b and c**). However, the calculated phonon spectrum shows that this zigzag (12, 0) SnSe nanotube presents large negative frequency at the Gamma point (**Figure S2**).
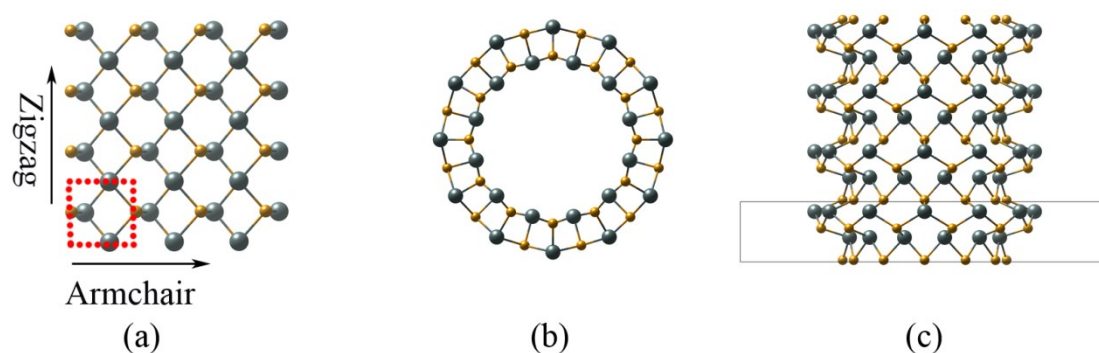

Figure S1. (a) Atomic structure of a 4×4 SnSe layer, dash red line is the unit cell. (b) Top view of zigzag (12, 0) SnSe tube. (c) Side view of the zigzag (12, 0) tube. The grey line is the unit cell. Se: brown, Sn: light cyan
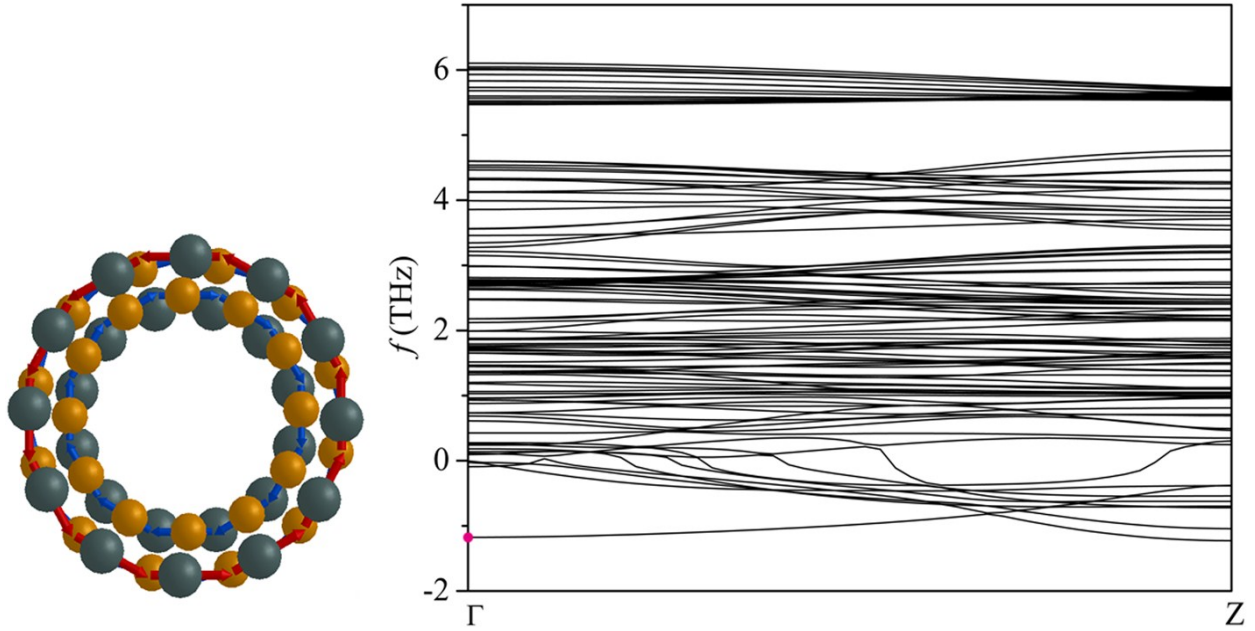
Figure S2. Calculated phonon spectrum for the zigzag SnSe (12, 0) tube (right) and the vibration mode of the −1.2 THz phonon branch at gamma point (left).

The phonon vector of this negative vibration mode shows that the neighbouring two SnSe rings rotate towards opposite direction (**Figure S2**). Based on this information, we rotated one of the SnSe ring with appropriate angle around the center of the ring. We then further optimize this structure with more rigid force converge threshold of 10^{-4} eV/Å. The new 'star' like (12, 0) SnSe nanotube (SNT) is obtained and its atomic coordination is listed in **Table S1**. This SNT is rather stable without any negative frequency as discussed in the main text.

Table S1: Fractional coordination of the "star-like" SnSe (12, 0) nanotube. Lattice constant: a=b=27.007451294, c=6.169519453.

Element	x	y	z
Se	0.7743489991918372	0.6581860188456339	0.2066922856282074
Se	0.7482409847282608	0.5673325424139219	0.7073095517835499
Se	0.2256510008081695	0.3418139811543656	0.2066922856282074
Se	0.2517590152717393	0.4326674575860784	0.7073095517835499
Se	0.3418139811543656	0.7743489991918372	0.2066922856282074

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Se	0.5673325424139219	0.7482409847282608	0.7073095517835499
Se	0.8153246712804810	0.5000000000000000	0.2082402420171416
Se	0.1846753287195188	0.5000000000000000	0.2082402420171416
Se	0.5000000000000000	0.8153246712804810	0.2082402420171416
Se	0.5000000000000000	0.1846753287195188	0.2082402420171416
Se	0.6845355469838476	0.6845355469838476	0.7114978658009643
Se	0.3154644530161527	0.3154644530161527	0.7114978658009643
Se	0.3154644530161527	0.6845355469838476	0.7114978658009643
Se	0.6845355469838476	0.3154644530161527	0.7114978658009643
Sn	0.7351533588451987	0.5637355823049259	0.1686913125575043
Sn	0.7831422682017040	0.6622650214421075	0.7361394729150686
Sn	0.2648466411548010	0.4362644176950744	0.1686913125575043
Sn	0.2168577317982961	0.3377349785578926	0.7361394729150686
Sn	0.4362644176950744	0.7351533588451987	0.1686913125575043
Sn	0.3377349785578926	0.7831422682017040	0.7361394729150686
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Sn	0.6622650214421075	0.2168577317982961	0.7361394729150686
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Sn	0.5637355823049259	0.7351533588451987	0.1686913125575043
Sn	0.6622650214421075	0.7831422682017040	0.7361394729150686
Sn	0.8237111384292854	0.5000000000000000	0.6707821199884521
Sn	0.1762888615707079	0.5000000000000000	0.6707821199884521
Sn	0.5000000000000000	0.8237111384292854	0.6707821199884521
Sn	0.5000000000000000	0.1762888615707079	0.6707821199884521
Sn	0.6732191235206533	0.6732191235206533	0.2617140479558535
Sn	0.3267808764793468	0.3267808764793468	0.2617140479558535
Sn	0.3267808764793468	0.6732191235206538	0.2617140479558535
Sn	0.6732191235206533	0.3267808764793463	0.2617140479558535

II. Boltzmann transport theory

The semi-classic Boltzmann transport theory describes the electrical conductivity tensor $\sigma_{\alpha\beta}(T,\mu)$ and electric thermal conductivity $\kappa_{\alpha\beta}^{e}$ at a non-zero electric current as following:[1]

$$\sigma_{\alpha\beta}(T,\mu) = \frac{1}{\Omega} \int \sigma_{\alpha\beta}(\varepsilon) \left[-\frac{\partial f}{\partial \varepsilon} \right] d\varepsilon \tag{S1}$$

$$\kappa_{\alpha\beta}^{e}(T,\mu) = \frac{1}{e^{2}T\Omega} \int \sigma_{\alpha\beta}(\varepsilon)(\varepsilon - \mu)^{2} \left[-\frac{\partial f}{\partial E} \right] d\varepsilon \tag{S2}$$

where Ω, ε, f and e are reciprocal space volume, carry energy, Fermi distribution function and electron charge, respectively. The electrical conductivity tensor $\sigma_{\alpha\beta}(\varepsilon)$ can be expressed as:

$$\sigma_{\alpha\beta}(\varepsilon) = \frac{e^{2}\tau}{N} \sum_{i,k} v_{\alpha} v_{\beta} \frac{\delta(\varepsilon - \varepsilon_{i,k})}{d\varepsilon} \tag{S3}$$

where τ, v_{α} and k, are the electron relaxation time, group velocity and the wave-vector.

III. Convergence of lattice thermal conductivity

The convergence of the lattice thermal conductivity of the SNT was tested to ensure the accuracy of the calculation results. The variation of lattice thermal conductivity with respect to the force cutoff employed in third-order calculations are shown in **Figure S3**, which indicates that the lattice thermal conductivity of SNT converged well within the employed cutoff.
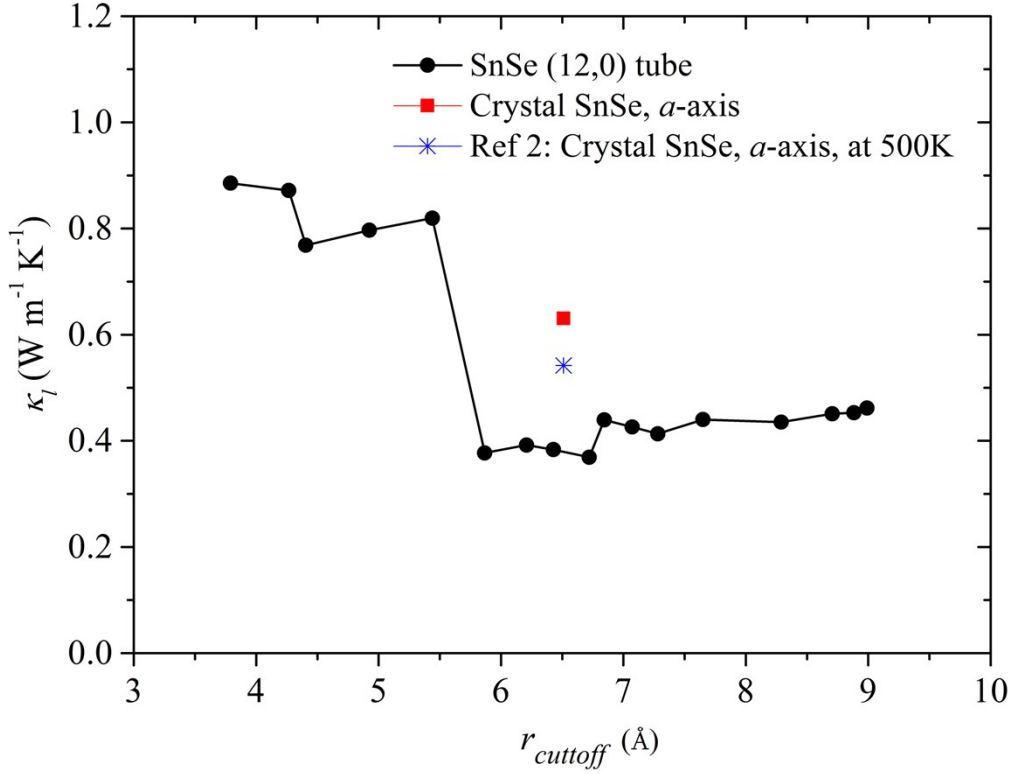
Figure S3. Convergence of the lattice thermal conductivity of the SNT at T = 300 K with respect to the force cutoff employed in third-order calculations. Points represent actual calculations; lines are provided only as a guide to the eye.

IV. Efficiency of a thermoelectric material

The power generation efficiency of a thermoelectric converter is defined as:[3]

$$\eta_p = \frac{\sqrt{1 + ZT_{av}} - 1}{\sqrt{1 + ZT_{av}} + \frac{T_c}{T_H}} \cdot \frac{T_h - T_c}{T_h}$$

(S4)

where T_h and T_c are the temperature at the hot and the cold ends, ZT_{av} is the average value of both the n-type and the p-type legs. If ZT_{av} approaches 4.0, the power generation efficiency η_p is about 27% at a typical value of T_c = 300 K and the hot and cold end temperature difference $T_h - T_c$ = 400 K.

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