

Supplementary Materials

Designable transverse thermoelectricity through interlayer thermal coupling for conformal heat harvesting

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This supporting file includes:

- Supplementary Note 1 to 8
- Supplementary Figures 1 to 24
- Supplementary Tables 1 and 3

The sequence is the same as they are mentioned in the main text.

Supplementary Note 1: Bilayer thermal coupling model and analytical solution

First, we consider the following three-dimensional model in which two rectangular bars with dimensions L along the x -axis and a along the y -axis are stacked along the z -axis and separated by a coupling layer, and a heat source and a cold reservoir are applied along the x -axis, enabling a temperature difference ΔT on the bars. The upper layer (Layer 1) of thickness d_1 is composed of an isotropic material, while the lower layer (Layer 2) of thickness d_2 is composed of an anisotropic material with off-diagonal terms in its thermal conductivity tensor. With the assistance of the coupling layer, the thermal field of upper-layer bar can be synergistically manipulated by tailoring the lower layer thermal field. According to Fourier's law, the heat flux density vector is given by^[1,2]:

$$\mathbf{q} = -\kappa \nabla T \quad (\text{S1})$$

where κ is the thermal conductivity tensor. The local energy conservation law in the transient case reads:

$$\rho c \frac{\partial T}{\partial t} + \nabla \cdot \mathbf{q} = 0 \quad (\text{S2})$$

where ρ denotes the mass density and c represents the specific heat capacity. Under steady-state conditions ($\partial T / \partial t = 0$), the energy flux yields $\nabla \cdot \mathbf{q} = 0$, leading to the steady-state heat conduction equation in a solid:

$$\nabla \cdot (\kappa \nabla T) = 0 \quad (\text{S3})$$

Returning to the bilayer thermal coupling system Supplementary Figure 4, we denote the temperature fields in the upper and lower layers as T_1 and T_2 , respectively. Since the two layers are connected by a thin coupling layer of thickness d_c , heat exchange takes place across their interface. The interlayer heat flux can be written as:

$$\mathbf{q}_{int} = \frac{\kappa_c}{d_c} (T_1 - T_2) \quad (\text{S4})$$

where κ_c is the thermal conductivity of the coupling layer. Under thin-layer conditions, thermal evolution along the z -direction within each layer can be neglected (simplifying the three-dimensional thermal field to a two-dimensional distribution), the governing equations for the bilayer system are

$$\begin{cases} \nabla \cdot (d_1 \kappa_1 \nabla T_1) + \frac{\kappa_c}{d_c} (T_2 - T_1) = 0 \\ \nabla \cdot (d_2 \kappa_2 \nabla T_2) + \frac{\kappa_c}{d_c} (T_1 - T_2) = 0 \end{cases} \quad (\text{S5})$$

where κ_1 and κ_2 are the thermal conductivity tensors of the upper and lower layers. Herein, a semi-analytical solution is considered with the boundary condition (Heat-source T_h and cold-reservoir T_c are respectively applied at $x=-L/2$ and $x=L/2$). Then, we maintain the thermal condition of the lower layer and tailor the temperature field with a rotation angle θ . The simplified balance equation for the upper layer (isotropic with scalar conductivity k) reads:

$$d_1 \kappa_1 \nabla^2 T_1 + \frac{\kappa_c}{d_c} \left(T_0 - \frac{T_h - T_c}{L} (x \cos \theta + y \sin \theta) - T_1 \right) = 0 \quad (S6)$$

Here, $T_0 = \frac{T_h + T_c}{2}$ is the temperature along the central line. Subject to the Neumann boundary condition ($\partial T / \partial y = 0$ at $x = \pm a/2$), we introduce the following dimensionless parameters: the normalized coordinates $\xi = x/L$, $\eta = y/a$ and the dimensionless temperature $\tau = (T_1 - T_0) / (T_h - T_c)$. Accordingly, Equation S6 reduces to a steady-state inhomogeneous Helmholtz-type equation^[3,4]:

$$\frac{\partial^2 \tau}{\partial \xi^2} + \frac{1}{\beta^2} \frac{\partial^2 \tau}{\partial \eta^2} - \Gamma \tau = \Gamma (\xi \cos \theta + \beta \eta \sin \theta) \quad (S7)$$

where parameter $\Gamma = \kappa_c L^2 / \kappa_1 d_1 d_c$ represents the coupling strength and $\beta = a/L$ is the width-to-length ratio. To facilitate the solution, we further decompose the dimensionless temperature $\tau(\xi, \eta)$ into a linear background term $-\xi$ (which exactly satisfies the lateral Neumann boundary conditions and the imposed temperature gradient) and a correction term $\psi(\xi, \eta)$ that accounts for the temperature twist^[5]. Substituting $\tau(\xi, \eta) = -\xi + \psi(\xi, \eta)$ into Equation S7 yields the governing equation for ψ :

$$\frac{\partial^2 \psi}{\partial \xi^2} + \frac{1}{\beta^2} \frac{\partial^2 \psi}{\partial \eta^2} - \Gamma \psi = \Gamma [\xi (\cos \theta - 1) + \beta \eta \sin \theta] \quad (S8)$$

With homogeneous boundary conditions $\psi(\pm \frac{1}{2}, \eta) = 0$ and $\partial \psi / \partial \eta|_{\eta=\pm 1/2} = 0$, the method of separation of variables leads to the eigenfunctions $\psi_n(\eta) = \cos \left[n\pi \left(\eta + \frac{1}{2} \right) \right]$ for $n=0, 1, 2, \dots$. Thus, the solution can be expanded as:

$$\psi(\xi, \eta) = \sum_{n=0}^{\infty} \Phi_n(\xi) \psi_n(\eta) \quad (S9)$$

Substituting the series expansion into Equation S8 and exploiting the orthogonality of eigenfunctions, the modal ordinary differential equations governing the expansion coefficients are obtained as:

$$\Phi_n'' - \lambda_n^2 \Phi_n = R_n(\xi) \quad (S10)$$

where $\lambda_n = \sqrt{\Gamma + (n\pi/\beta)^2}$ is the n th eigenvalue obtained from the separation of variables in the η -direction and the modal source terms $R_n(\xi)$ are the expansion coefficients of the right-hand side of Equation S8, as $R(\xi, \eta) = \Gamma [\xi (\cos \theta - 1) + \beta \eta \sin \theta]$. Beginning with the expansion

$R(\xi, \eta) = \sum_{m=0}^{\infty} R_m(\xi) \psi_m(\eta)$, we multiply both sides by the n th eigenfunction $\psi_n(\eta)$ and integrate over $\eta \in [-1/2, 1/2]$ to obtain $\int_{-1/2}^{1/2} R(\xi, \eta) \psi_n(\eta) d\eta = \int_{-1/2}^{1/2} \left[\sum_{m=0}^{\infty} R_m(\xi) \psi_m(\eta) \right] \psi_n(\eta) d\eta$. Assuming uniform convergence of the series, we interchange the summation and integration, yielding $\int_{-1/2}^{1/2} \left[\sum_{m=0}^{\infty} R_m(\xi) \psi_m(\eta) \right] \psi_n(\eta) d\eta = \sum_{m=0}^{\infty} R_m(\xi) \int_{-1/2}^{1/2} \psi_m(\eta) \psi_n(\eta) d\eta$. According to the orthogonality conditions:

$$\int_{-1/2}^{1/2} \psi_m(\eta) \psi_n(\eta) d\eta = \begin{cases} 1, & m=n=0 \\ \frac{1}{2}, & m=n \neq 0 \\ 0, & m \neq n \end{cases} \quad (\text{S11})$$

The integral vanishes for all $m \neq n$, causing the infinite series to collapse to a single term:

$$\int_{-1/2}^{1/2} R(\xi, \eta) \psi_n(\eta) d\eta = R_n(\xi) \cdot \begin{cases} 1, & n=0 \\ \frac{1}{2}, & n \geq 1 \end{cases} \quad (\text{S12})$$

Thus, the expansion coefficients are obtained as:

$$R_n(\xi) = \begin{cases} \int_{-1/2}^{1/2} R(\xi, \eta) d\eta, & n=0 \\ 2 \int_{-1/2}^{1/2} R(\xi, \eta) \psi_n(\eta) d\eta, & n \geq 1 \end{cases} \quad (\text{S13})$$

Yielding $R_0(\xi) = \Gamma \xi (\cos \theta - 1)$ for the zeroth mode, $R_n(\xi) = -4\Gamma \beta \sin \theta / (n\pi)^2$ for odd modes ($n=1, 3, 5, \dots$), and $R_n=0$ for even modes ($n=2, 4, 6, \dots$). We now solve the Equation S10 with the boundary conditions $\Phi_n(\pm 1/2) = 0$. For the zeroth mode, the governing equation is

$\Phi_0'' - \Gamma \Phi_0 = \Gamma \xi (\cos \theta - 1)$, and the general solution can be written as:

$\Phi_0(\xi) = -(\cos \theta - 1)\xi + C_1 \cosh(\sqrt{\Gamma}\xi) + C_2 \sinh(\sqrt{\Gamma}\xi)$. Applying the boundary conditions:

$$\begin{cases} \Phi_0\left(\frac{1}{2}\right) = -(\cos \theta - 1)\frac{1}{2} + C_1 \cosh\left(\frac{\sqrt{\Gamma}}{2}\right) + C_2 \sinh\left(\frac{\sqrt{\Gamma}}{2}\right) = 0 \\ \Phi_0\left(-\frac{1}{2}\right) = (\cos \theta - 1)\frac{1}{2} + C_1 \cosh\left(\frac{\sqrt{\Gamma}}{2}\right) - C_2 \sinh\left(\frac{\sqrt{\Gamma}}{2}\right) = 0 \end{cases} \quad (\text{S14})$$

The zeroth-order correction term is thus obtained as:

$$\Phi_0(\xi) = (1 - \cos \theta) \left[\xi - \frac{\sinh(\sqrt{\Gamma}\xi)}{2 \sinh\left(\frac{\sqrt{\Gamma}}{2}\right)} \right] \quad (\text{S15})$$

And for the odd modes, the Equation S10 becomes $\Phi_n'' - \lambda_n^2 \Phi_n = -4\Gamma \beta \sin \theta / (n\pi)^2$, so the general

solution takes the form: $\Phi_n(\xi) = -\frac{R_n}{\lambda_n^2} + C_1 \cosh(\lambda_n \xi) + C_2 \sinh(\lambda_n \xi)$. Applying the boundary

conditions:

$$\begin{cases} \Phi_n\left(\frac{1}{2}\right) = -\frac{R_n}{\lambda_n^2} + C_1 \cosh\left(\frac{\lambda_n}{2}\right) + C_2 \sinh\left(\frac{\lambda_n}{2}\right) = 0 \\ \Phi_n\left(-\frac{1}{2}\right) = -\frac{R_n}{\lambda_n^2} + C_1 \cosh\left(\frac{\lambda_n}{2}\right) - C_2 \sinh\left(\frac{\lambda_n}{2}\right) = 0 \end{cases} \quad (\text{S16})$$

Solving the system yields:

$$\Phi_n(\zeta) = \frac{4\Gamma\beta \sin \theta}{(\pi)^2 \lambda_n^2} \left[1 - \frac{\cosh(\lambda_n \zeta)}{\cosh\left(\frac{\lambda_n}{2}\right)} \right] \quad (\text{S17})$$

For even modes, since $R_n=0$ and the boundary conditions are homogeneous, $\Phi_n=0$. Assembling the contributions in Equation S15 and S17, the complete dimensionless temperature distribution is obtained as:

$$\tau(\zeta, \eta) = -\zeta \cos \theta - \frac{1 - \cos \theta}{2} \frac{\sinh(\sqrt{\Gamma} \zeta)}{\sinh\left(\frac{\sqrt{\Gamma}}{2}\right)} + \sum_{n=1,3,5,\dots}^{\infty} \frac{4\Gamma\beta \sin \theta}{(\pi)^2 \lambda_n^2} \left[1 - \frac{\cosh(\lambda_n \zeta)}{\cosh\left(\frac{\lambda_n}{2}\right)} \right] \cos \left[n\pi \left(\eta + \frac{1}{2} \right) \right] \quad (\text{S18})$$

Supplementary Note 2: Effective transverse thermoelectric transport coefficients

The upper thermoelectric layer in the bilayer coupling system has geometric dimensions: length L along the x -direction (the principal direction of the applied thermal bias), width a along the y -direction (the transverse electrode spacing), and thickness d_1 . Under the interlayer-coupled oblique thermal excitation, the temperature field in the upper layer exhibits both a longitudinal temperature difference ΔT_x and an induced transverse temperature difference ΔT_y across the width a .

Within the linear Onsager transport framework, the coupled system can be described as an effective two-dimensional thermoelectric medium with tensorial transport coefficients. The constitutive relations for the electric field E and heat flux q in the upper layer are expressed in matrix form as:

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix} = \rho \begin{pmatrix} J_x \\ J_y \end{pmatrix} + \mathbf{S}^* \begin{pmatrix} \nabla_x T \\ \nabla_y T \end{pmatrix} \quad (\text{S19})$$

where $\mathbf{J} = (J_x, J_y)^T$ is the electric current density, $\rho = \rho \mathbf{I}$ is the isotropic electrical resistivity tensor with $\rho = 1/\sigma$ (σ being the electrical conductivity), and $\mathbf{S}^*$ is the effective Seebeck tensor that captures the emergent anisotropy arising from the oblique thermal boundary condition.

I. Effective Anisotropic Seebeck Tensor, Power Factor and Figure of Merit

Under open-circuit conditions ($J_x = J_y = 0$), the Seebeck effect generates an electric field parallel to the local temperature gradient, $\mathbf{E} = S \nabla T$, where S is the intrinsic Seebeck tensor of the thermoelectric material (the voltage produced per unit temperature difference). The transverse electric field follows $E_y = S(\partial T / \partial y)$. Integrating this field over the width a yields the open-circuit transverse voltage:

$$V_y = \int_0^a E_y dy = S \Delta T_y \quad (\text{S20})$$

The device-level effective transverse Seebeck tensor is defined as the ratio of the measured transverse voltage to the applied longitudinal temperature difference:

$$S_{yx}^* = \frac{V_y}{\Delta T_x} = S \frac{\Delta T_y}{\Delta T_x} \quad (\text{S21})$$

This relation explicitly separates the material intrinsic response (S) from the geometric thermal projection ($\Delta T_y / \Delta T_x$). The effective Seebeck tensor therefore takes the form:

$$S^* = \begin{pmatrix} S & 0 \\ S \frac{\Delta T_y}{\Delta T_x} & S \end{pmatrix} \quad (\text{S22})$$

The off-diagonal component S_{yx}^* is not an intrinsic material property, but a geometric consequence of the interlayer thermal coupling, and can be continuously tuned by adjusting the coupling strength without modifying the thermoelectric material itself.

The effective transverse power factor is constructed from the off-diagonal Seebeck component and the electrical conductivity:

$$PF_{yx}^* = (S_{yx}^*)^2 \sigma = S^2 \sigma \left(\frac{\Delta T_y}{\Delta T_x} \right)^2 = PF_0 \left(\frac{\Delta T_y}{\Delta T_x} \right)^2 \quad (\text{S23})$$

Where $PF_0 = S^2 \sigma$ is the intrinsic power factor of the upper-layer material. The effective transverse figure of merit follows as:

$$ZT_{yx}^* = \frac{PF_{yx}^* T}{\kappa_1} = \frac{S^2 \sigma T}{\kappa_1} \left(\frac{\Delta T_y}{\Delta T_x} \right)^2 = ZT_0 \left(\frac{\Delta T_y}{\Delta T_x} \right)^2 \quad (\text{S24})$$

where κ_1 is the thermal conductivity of the upper layer, T is the average operating temperature, and $ZT_0 = S^2 \sigma T / \kappa_1$ is the intrinsic thermoelectric figure of merit. This result reveals that the cooperative bilayer coupling endows the isotropic material with an extrinsic, geometry-tunable thermoelectric response that scales quadratically with the transverse-to-longitudinal temperature ratio.

II. Maximum Output Power and Conversion Efficiency

The transverse electrical resistance for current flowing along the y -direction through a cross-sectional area $L \times d_1$ is determined by the resistivity ρ and the current-path geometry:

$$R_y = \rho \frac{a}{L d_1} \quad (\text{S25})$$

Under matched-load conditions ($R_L = R_y$), the maximum transverse output power is:

$$P_{\max} = \frac{V_y^2}{4R_y} = \frac{S^2 \sigma a d_1}{4L} \Delta T_x^2 \left(\frac{\Delta T_y}{\Delta T_x} \right)^2 \quad (\text{S26})$$

Experimentally, $P_{\max}$ is determined by measuring the open-circuit voltage V_{oc} and the device internal resistance R under isothermal conditions, yielding $P_{\max} = V_{oc}^2 / 4R$.

The thermoelectric conversion efficiency is defined as the ratio of the maximum electrical output power to the input thermal power. The input thermal power is the longitudinal heat flux through the upper layer:

$$Q_{\text{in}} = q_x \cdot (a d_1) = \kappa_1 \frac{\Delta T_x}{L} a d_1 \quad (\text{S27})$$

The direct heat-balance efficiency is therefore:

$$\eta = \frac{P_{\max}}{Q_{\text{in}}} = \frac{S^2 \sigma}{4 \kappa_1} \Delta T_x \left(\frac{\Delta T_y}{\Delta T_x} \right)^2 = \frac{ZT_0}{4} \frac{\Delta T_x}{T} \left(\frac{\Delta T_y}{\Delta T_x} \right)^2 = \frac{ZT_{yx}^*}{4} \frac{\Delta T_x}{T} \quad (\text{S28})$$

This expression elucidates the linear scaling with the temperature span ΔT_x and the quadratic dependence on the transverse thermal ratio. For a more precise evaluation over arbitrary temperature spans, the effective transverse figure of merit ZT_{yx}^* can be substituted into the standard Ioffe–Goldsmid relation:

$$\eta_{\max} = \eta_{\text{Carnot}} \cdot \frac{\sqrt{1 + ZT_{yx}^*} - 1}{\sqrt{1 + ZT_{yx}^* + T_c/T_h}} \quad (\text{S29})$$

where $\eta_{\text{Carnot}} = (T_h - T_c)/T_h$ is the Carnot efficiency. Together, these expressions provide a complete analytical description of the interlayer-coupled transverse thermoelectric performance, from the emergent effective Seebeck tensor to the maximum output power and conversion efficiency.

Supplementary Note 3: Equivalent off-diagonal thermal conductivity

In the bilayer thermal coupling system, the upper layer is intrinsically isotropic with scalar thermal conductivity κ_1 . Under the interlayer-coupled oblique thermal excitation, the heat flux in the upper layer is no longer parallel to the temperature gradient. This emergent anisotropic transport can be described by an effective thermal conductivity tensor κ^* . Following the standard characterization procedure, the effective off-diagonal component is determined by applying a longitudinal temperature gradient and measuring the induced transverse heat flux:

$$\kappa_{yx}^* = \frac{q_y}{\left(\partial T / \partial x\right)_x} \quad (\text{S30})$$

Within the linear Onsager framework, the transverse heat flux is driven by the transverse temperature gradient, yielding:

$$\frac{\kappa_{yx}^*}{\kappa_1} = \frac{\Delta T_y}{\Delta T_x} \quad (\text{S31})$$

This relation explicitly reveals that the equivalent off-diagonal thermal conductivity of the intrinsically isotropic upper layer scales linearly with the transverse-to-longitudinal temperature ratio, which is purely governed by the interlayer geometric thermal coupling.

Supplementary Note 4: Coordinate transformation for boundary distortion elimination

To simplify the analysis, we consider a two-dimensional steady-state heat conduction problem in an anisotropic medium characterized by an off-diagonal thermal conductivity tensor $\hat{\kappa} = \begin{pmatrix} \kappa_{xx} & \kappa_{xy} \\ \kappa_{yx} & \kappa_{yy} \end{pmatrix}$.

The temperature field $T(x,y)$ satisfies the governing equation^[2,6]:

$$\kappa_{xx} \frac{\partial^2 T}{\partial x^2} + 2\kappa_{xy} \frac{\partial^2 T}{\partial x \partial y} + \kappa_{yy} \frac{\partial^2 T}{\partial y^2} = 0 \quad (\text{S32})$$

In a finite rectangular domain with adiabatic lateral boundaries, the intrinsic oblique thermal field becomes incompatible with rigid vertical boundary constraints, resulting in thermal field distortion near the edges and precluding a simple analytical solution^[7,8].

Consider the scenario where a uniform heat flux flows parallel to the x -direction. In the bulk region with uniform $\hat{\kappa}$, the isothermal lines naturally tilt with an angle θ relative to the y -axis, determined by the tensor components as $\tan\theta = \frac{\kappa_{xy}}{\kappa_{yy}}$. To mitigate edge distortion while maintaining the oblique thermal field, a linear coordinate transformation is adopted to compress lateral trapezoidal domains along the heat flow direction into rectangular boundary regions^[9].

As illustrated in Supplementary Figure 8, we first construct a virtual parallelogram space with uniform background thermal conductivity $\hat{\kappa}$, where the inclined sides form an angle θ with the y -axis direction. A rectangular domain of dimensions $L \times a$ is extracted at the center of this virtual space. This rectangular region remains invariant under the transformation, serving as the physical bulk material. The two lateral regions outside this central rectangle are then linearly mapped into rectangular boundary regions adjacent to the central domain.

Due to spatial symmetry, we focus here on the left boundary region I. The virtual trapezoidal region A'B'CD is mapped to the physical rectangular region ABCD, where the interface CD (at $x = -\frac{L}{2}$) remains fixed during the transformation. The coordinates in the virtual space are defined as:

$A' \left(-\frac{L}{2} - \delta, \frac{a}{2} \right), B' \left(-\frac{L}{2} - \delta - \tan\theta \cdot a, -\frac{a}{2} \right), C \left(-\frac{L}{2}, -\frac{a}{2} \right), D \left(-\frac{L}{2}, \frac{a}{2} \right)$ After transformation, the physical boundary region occupies the region $x \in \left[-\frac{L}{2} - l, -\frac{L}{2} \right], y \in \left[-\frac{a}{2}, \frac{a}{2} \right]$. The coordinate transformation from the virtual space (x', y') to the physical space (x, y) is given by:

$$\begin{cases} x = -\frac{L}{2} + \frac{\delta \left(x' + \frac{L}{2} \right)}{\delta + \left(\frac{a}{2} - y' \right) \tan\theta} \\ y = y' \end{cases} \quad (\text{S33})$$

where the local width function of the virtual trapezoid is $w(y) = \delta + \left(\frac{a}{2} - y\right) \tan \theta$. The Jacobian matrix

$\mathbf{J} = \frac{\partial(x,y)}{\partial(x',y')}$ of this transformation is:

$$\mathbf{J} = \begin{pmatrix} \frac{\delta}{w(y)} & \frac{\left(x + \frac{L}{2}\right) \tan \theta}{w(y)} \\ 0 & 1 \end{pmatrix} \quad (\text{S34})$$

According to the transformation thermodynamics, the effective thermal conductivity tensor in the physical boundary region is obtained as^[10,11]:

$$\tilde{\kappa}(x,y) = \frac{\mathbf{J} \hat{\kappa} \mathbf{J}^T}{\det(\mathbf{J})} \quad (\text{S35})$$

This spatially varying conductivity tensor compensates for the boundary geometry mismatch, ensuring that the oblique thermal field in the bulk region transitions smoothly to vertical isotherms at the lateral boundaries.

The central active region was designed with dimensions $L \times a = 15 \text{ mm} \times 15 \text{ mm}$. For a target deflection of 30° , the thermal conductivity tensor components satisfy $\kappa_{xy}/\kappa_{yy} = \sqrt{3}$, which can be realized through an anisotropic multilayered composite with appropriately oriented constituent layers. For the 60° configuration, the ratio $\kappa_{xy}/\kappa_{yy} = \sqrt{3}/3$ is achieved by adjusting the lamination orientation and volume fraction of the layered structure. The detailed design method for these tensor components and their mapping to specific microstructural geometries will be provided in Supplementary Note 6. Based on the coordinate transformation framework, gradient-index boundary layers are introduced at the lateral peripheries of the central region. The physical boundary architecture extends $l = 7.5 \text{ mm}$ beyond the central domain for all configurations. To ensure that the same applied longitudinal temperature gradient produces consistent heat flux densities and comparable thermal boundary conditions across different angular configurations, the virtual spaces are designed with a uniform x -direction projected distance at the midline ($y=0$). Taking the 60° configuration as the reference ($\delta=0$), the 30° configuration requires an additional virtual transition thickness $\delta = 8.66 \text{ mm}$ to compensate for its shallower inclination, thereby matching the x -direction projection of the 60° case. Within this transition region, the thermal conductivity tensor components vary spatially according to the affine transformation mapping, ensuring a smooth evolution from the anisotropic central tensor to the isotropic outer boundaries. This geometric invariance guarantees that the thermal driving force remains uniform across different angular configurations, enabling fair comparison of interlayer coupling effects. Supplementary Figure 9 illustrates the calculated spatial

distributions of κ_{xx} , κ_{xy} and κ_{yy} across the boundary transition zones, demonstrating the continuous variation required for impedance matching.

Finite element simulations were conducted using COMSOL Multiphysics. The lateral boundaries were prescribed as high temperature (353 K) and low temperature (293 K), while all remaining boundaries were specified as adiabatic. Supplementary Figure 10 presents the thermal field distributions for both configurations. Supplementary Figure 10a and 10c display the temperature contours and heat flux trajectories within the central region for the 30° and 60° cases, respectively. The isotherms maintain uniform oblique orientations throughout the central domain without the warping typically associated with rectangular off-diagonal heat conduction, indicating that the transformation-based boundary regions successfully decouple the field generation from geometric constraints.

Supplementary Figure 10b and 10d present quantitative line scans along the horizontal centerline of the structure. The temperature gradient deflection angle transitions smoothly from 0° at the outer boundaries to the target values within the central region, with the angular variation confined to the boundary transition zones. Concurrently, the horizontal heat flux density q_x remains uniform across the entire domain, confirming that the boundary design eliminates distortions without introducing additional thermal resistance perpendicular to the flow.

Supplementary Note 5: Finite-element discretization and mesh convergence

While the continuous coordinate transformation framework presented enables theoretically perfect thermal field manipulation, the spatially varying thermal conductivity tensor $\kappa(x,y)$ derived from continuous mapping poses significant challenges for practical fabrication. To bridge this gap, we adopt the finite element method (FEM), an efficient and reliable numerical approach for solving complex physical field problems. This method discretizes the continuously varying transformation domain into a set of small, uniform elements, enabling precise control of the heat flux and temperature field within the defined region^[9,12,13].

The FEM approach discretizes the target transformation domain into j quadrilateral finite elements, where within each element the thermal conductivity tensor is treated as constant. For an arbitrary quadrilateral element mapping from the virtual space (x',y') to the physical space (x,y) , the transformation is characterized by the discrete Jacobian matrix evaluated at the element centroid or edge midpoints.

As illustrated in Supplementary Figure 11, the four edge midpoints of element $s'_i(x'_i,y'_i)$ in virtual space and $s_i(x_i,y_i)$ in physical space ($i = 1,2,3,4$). The discrete Jacobian approximating the local deformation gradient is constructed as:

$$J_j = \begin{pmatrix} \frac{\partial x}{\partial x'} & \frac{\partial x}{\partial y'} \\ \frac{\partial y}{\partial x'} & \frac{\partial y}{\partial y'} \end{pmatrix} = \begin{pmatrix} \frac{dx}{dx'} & \frac{dx}{dy'} \\ \frac{dy}{dx'} & \frac{dy}{dy'} \end{pmatrix} = \begin{pmatrix} \frac{x_2-x_4}{x'_2-x'_4} & \frac{x_3-x_1}{y'_3-y'_1} \\ \frac{y_2-y_4}{x'_2-x'_4} & \frac{y_3-y_1}{y'_3-y'_1} \end{pmatrix} \quad (\text{S36})$$

where J_j denotes the Jacobian matrix of the j -th finite element. The transformed discrete thermal conductivity is given by:

$$[\tilde{\kappa}]_j = \frac{J_j \hat{\kappa} J_j^T}{\det(J_j)} \quad (\text{S37})$$

where $\hat{\kappa}$ represents the uniform thermal conductivity tensor in the virtual space.

The boundary regions for 30° and 60° oblique field configurations are discretized using progressively refined meshes with element densities of 1×2, 2×4, 4×8, and 10×20. Supplementary Figures 12 and 13 present the calculated spatial distributions of thermal conductivity tensor components for both configurations. Then, we numerically investigate the relationship between finite element density and the performance of thermal metamaterials. Supplementary Figure 14 presents the effect of different mesh densities on boundary manipulation for temperature gradient deflections of 30° and 60°, respectively.

Examination of the temperature field distributions and analysis along the central axis reveal that as the finite element density decreases, minor deviations emerge in the boundary regions, yet the central active region consistently exhibits uniform thermal gradient deflection matching the target angles. This robustness arises because the carefully designed central thermal conductivity tensor $\hat{\kappa}$ dominates the overall heat transport behavior, while the boundary layers function primarily as impedance-matching transition zones where moderate discretization errors are tolerated.

Supplementary Note 6: Effective medium theory for anisotropic thermal conductivity

While the continuous spatial variation of this tensor within the boundary transition regions is theoretically ideal for eliminating field distortions, practical fabrication demands a discrete, piecewise-homogeneous approximation. As outlined in Supplementary Note 5, the FEM discretization strategy evaluates the continuously varying tensor at the geometric center of each element and treats it as a uniform effective medium. To further simplify manufacturing, the entire boundary transition zone is herein treated as a single effective element, adopting the tensor evaluated at its geometric center as the homogeneous property for the whole region.

The target thermal conductivity tensor for each functional region is represented as a symmetric second-order tensor $\widetilde{\kappa}_{i,j} = \begin{pmatrix} \kappa_{xx} & \kappa_{xy} \\ \kappa_{xy} & \kappa_{yy} \end{pmatrix}$. The anisotropic thermal conductivity tensor with non-zero off-diagonal components can be physically realized through periodically laminated composites comprising two isotropic constituent materials, denoted A and B, with thermal conductivities κ_A and κ_B , respectively. Consider an alternating bilayer stack oriented parallel to the x -axis. The effective thermal conductivity in the principal directions follows the parallel and series rules:

$$\begin{aligned} \kappa_{//} &= f_A \kappa_A + f_B \kappa_B \\ \kappa_{\perp} &= (f_A / \kappa_A + f_B / \kappa_B)^{-1} \end{aligned} \quad (\text{S38})$$

where f_A and f_B are the volume fractions of materials A and B ($f_A + f_B = 1$). This yields an orthotropic effective tensor in the local laminate coordinates:

$$\widehat{\kappa}_{i,j} = \begin{bmatrix} \kappa_{//} & 0 \\ 0 & \kappa_{\perp} \end{bmatrix} \quad (\text{S39})$$

To obtain the required off-diagonal components, the laminated stack is rotated by an angle α about the z -axis. The coordinate transformation is governed by the rotation matrix:

$$\mathbf{J}_{\alpha} = \begin{pmatrix} \cos \alpha & \sin \alpha \\ -\sin \alpha & \cos \alpha \end{pmatrix} \quad (\text{S40})$$

Applying the tensor transformation rule yields the effective thermal conductivity in the global coordinate system:

$$\widetilde{\kappa}_{i,j} = \frac{\mathbf{J}_{\alpha} \widehat{\kappa}_{i,j} \mathbf{J}_{\alpha}^T}{\det(\mathbf{J}_{\alpha})} = \begin{pmatrix} \kappa_{//} \cos^2 \alpha + \kappa_{\perp} \sin^2 \alpha & (\kappa_{//} - \kappa_{\perp}) \sin \alpha \cos \alpha \\ (\kappa_{//} - \kappa_{\perp}) \sin \alpha \cos \alpha & \kappa_{//} \sin^2 \alpha + \kappa_{\perp} \cos^2 \alpha \end{pmatrix} \quad (\text{S41})$$

Thus, by appropriately selecting the constituent conductivities (κ_A, κ_B), the volume fraction (f_A, f_B), and the lamination angle α , any target thermal conductivity tensor satisfying positive-definiteness can be physically emulated.

The microstructural design proceeds in two stages. First, the central active region is configured to establish a uniform oblique thermal field with deflection angle θ , requiring the tensor components to satisfy $\kappa_{xy} / \kappa_{yy} = \tan \theta$. Second, to simplify the design and fabrication workflow, the entire

boundary transition region is treated as a single homogeneous effective element, adopting the thermal conductivity tensor evaluated at its geometric center as the uniform property for the whole zone. Both the central and boundary regions employ an identical high-conductivity skeleton material (material A), and only the local volume fraction and lamination orientation are spatially adjusted to match the respective target tensor components. The calculated constituent parameters for the central and boundary regions at various deflection angles are summarized in Supplementary Table 1.

Supplementary Note 7: Influence of interfacial contact resistance, convection, and radiative heat loss

To quantitatively evaluate the effects of interfacial contact resistance, convective heat transfer, and thermal radiation on interlayer thermal coupling and thermoelectric performance under experimental conditions, systematic finite-element simulations were performed. Under typical laboratory static testing conditions, the natural convection coefficient was set to $h=5 \text{ W}/(\text{m}^2 \cdot \text{K})$, the surface emissivity of the cut bulk Bi_2Te_3 thermoelectric plate was taken as $\varepsilon=0.4$, and the ambient temperature was 298 K. Supplementary Figure 16a compares the temperature contours considering only the contact thermal resistance $R_s=1 \times 10^{-3} \text{ m}^2 \cdot \text{K}/\text{W}$ (left column) with those simultaneously accounting for contact resistance, convection, and radiation (right column). Supplementary Figures 16b and 16c present the temperature profiles along the central y-axis of the upper layer for deflection angles of $\theta = 60^\circ$ and 30° , respectively, comparing the ideal case, the case with only contact resistance, and the case with all three loss mechanisms. The results show that introducing convection and radiation yields temperature distributions nearly identical to the case with only contact resistance, with a maximum deviation below 0.3 K, indicating that both effects are negligible for interlayer thermal coupling under laboratory conditions.

Contact thermal resistance is the primary source of deviation between experiment and theory. As described in the main text, the dimensionless coupling parameter Γ factorizes into a geometric term and an interfacial thermal conductance: $\Gamma=(L^2/\kappa_1 d_1) \cdot (\kappa_c/d_c)$. In practice, an additional areal-specific thermal contact resistance R_s is present in series with the coupling layer. The total interfacial thermal resistance per unit area is therefore $R_{\text{tot}}=d_c/\kappa_c+R_s$, and the effective coupling parameter reads

$$\Gamma_{\text{eff}} = \frac{L^2}{\kappa_1 d_1} \cdot \frac{1}{\frac{d_c}{\kappa_c} + R_s} \quad (\text{S42})$$

Substituting $d_c/\kappa_c=L^2/(\Gamma \kappa_1 d_1)$ into the denominator yields

$$\Gamma_{\text{eff}} = \frac{\Gamma}{1 + \frac{\Gamma R_s \kappa_1 d_1}{L^2}} \quad (\text{S43})$$

Supplementary Figure 16d shows the variation of Γ_{eff} with the ideal Γ for different values of R_s ($1 \times 10^{-4}, 1 \times 10^{-3}, 5 \times 10^{-3} \text{ m}^2 \cdot \text{K}/\text{W}$). Larger R_s leads to greater deviation of Γ_{eff} from the ideal value; for a fixed R_s , larger ideal Γ results in more pronounced suppression of the effective coupling by contact resistance. For example, when $R_s=1 \times 10^{-3} \text{ m}^2 \cdot \text{K}/\text{W}$, the effective coupling parameter for $\Gamma=100$ is reduced to only about 40. Based on the corrected Γ_{eff} , the transverse

thermoelectric performance was further evaluated: Supplementary Figures 16e and 16f present the power factor and output power density as functions of R_s , respectively. At $\theta=60^\circ$, the performance is more sensitive to R_s because larger deflection angles require stronger thermal coupling to fully transmit the oblique thermal field to the upper layer. When R_s increases from 0 to $5 \times 10^{-3} \text{ m}^2 \cdot \text{K/W}$, the power density of the 60° device decreases by approximately 40%, whereas the 30° device exhibits a smaller decline. This quantitative result explains the primary reason why experimental values fall below ideal predictions, and indicates that interfacial thermal resistance optimization is key to improving device performance.

Supplementary Note 8: Systematic error analysis

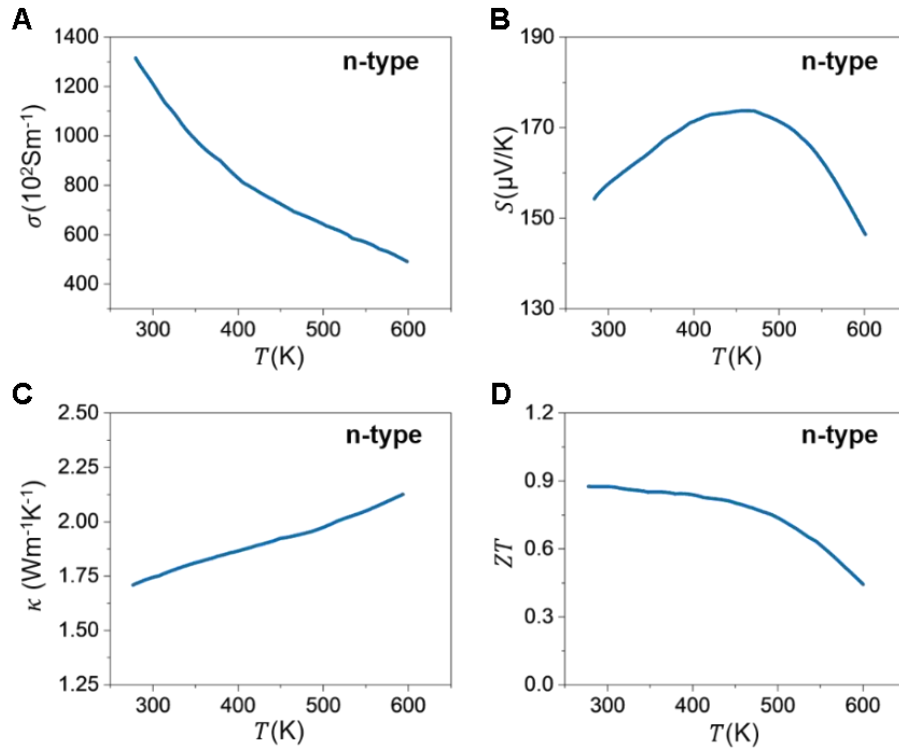
To quantitatively separate the contributions of boundary-layer design and interfacial/coupling-layer losses on the effective transverse Seebeck response, we define the experimental-to-simulated ratio as the total error function:

$$f(\theta, \Gamma) = \frac{\left(S_{yx}^*/S\right)_{exp}}{\left(S_{yx}^*/S\right)_{sim}} \quad (\text{S44})$$

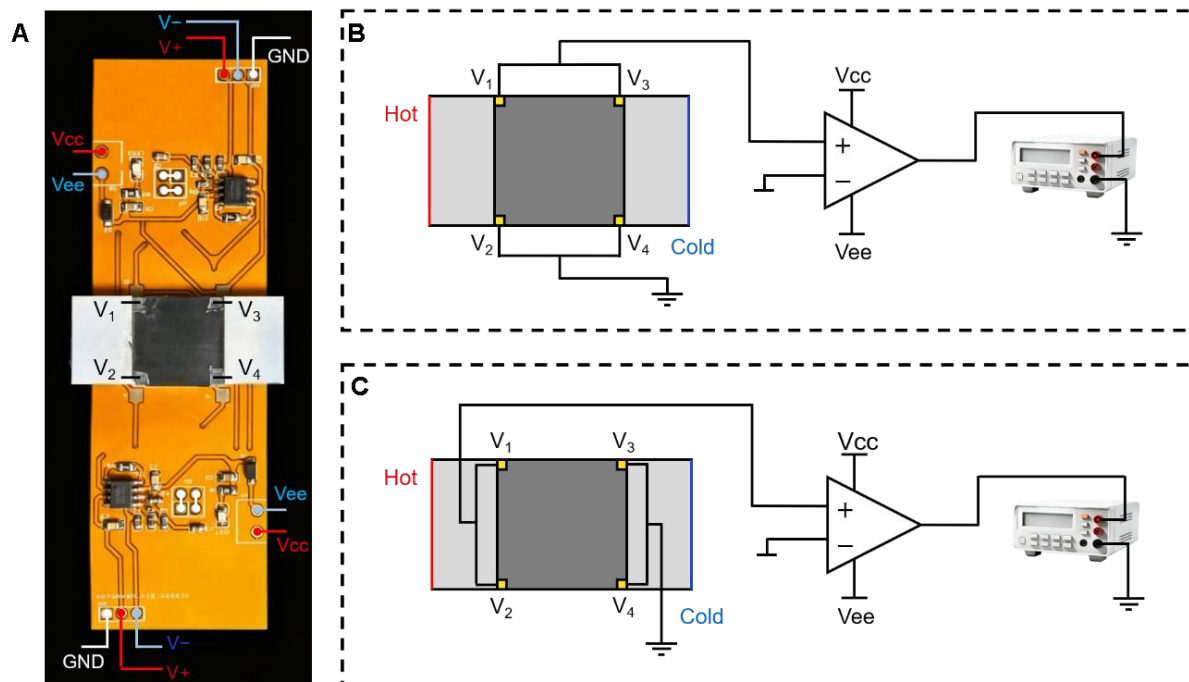
Because the boundary transition zones (angle-dependent) and the vertical coupling interface (Γ - dependent) are spatially orthogonal in geometry, the total attenuation can be empirically factorized as $f(\theta, \Gamma) = f_1(\theta) \times f_2(\Gamma)$, where $f_2(\Gamma)$ captures the coupling-layer contribution including interfacial contact thermal resistance, and $f_1(\theta)$ captures the boundary-layer contribution arising from fabrication imperfections in the gradient-index transition zones.

Supplementary Figure 20a and 20b present the simulated and experimental normalized off-diagonal Seebeck coefficients S_{yx}^*/S across the full parameter space of θ and Γ , respectively. The simulated results serve as the ideal reference, while the experimental data exhibit a consistent attenuation pattern relative to the theoretical predictions. Supplementary Figure 20c shows that, for a fixed deflection angle, the experimental S_{yx}^*/S increases with Γ and plateaus in the moderate-to-strong coupling regime ($\Gamma \geq 25$), where the experimental-to-simulated ratio stabilizes with a residual deviation below approximately 10%. This saturation indicates that interfacial contact thermal resistance—acting in series with the coupling layer—constitutes the baseline systematic loss that cannot be eliminated by merely increasing the coupling strength. Supplementary Figure 20d further reveals that S_{yx}^*/S depends strongly on θ . In the low-angle regime ($\theta \leq 30^\circ$), the experimental deviation from simulation is less than 10%, indicating that interfacial thermal resistance dominates the loss channel. As the deflection angle increases toward 45° and 60° , the heat-flux deflection ratio and the degree of thermal twisting grow substantially, amplifying the influence of the boundary transition zones on the overall thermal field and, consequently, on the transverse Seebeck response. While the increased deviation at large angles could, in principle, reflect the heightened sensitivity of transverse heat flux to interfacial contact resistance, the non-monotonic scatter of the residual factor suggests that the boundary-layer structure also contributes to the angle-dependent variations. Thus, the two loss channels operate with different weightings across the angular range: the interfacial contact resistance sets the baseline attenuation, while the boundary-layer influence becomes increasingly pronounced under large-angle thermal twisting.

Optimization should therefore address both channels simultaneously. For the interfacial layer, contact thermal resistance can be reduced by polishing the thermoelectric layer surface to improve flatness and applying uniform bonding pressure via vacuum lamination. For the boundary layer, lamination alignment precision should be tightened to minimize gap-induced thermal leakage at discrete block interfaces.



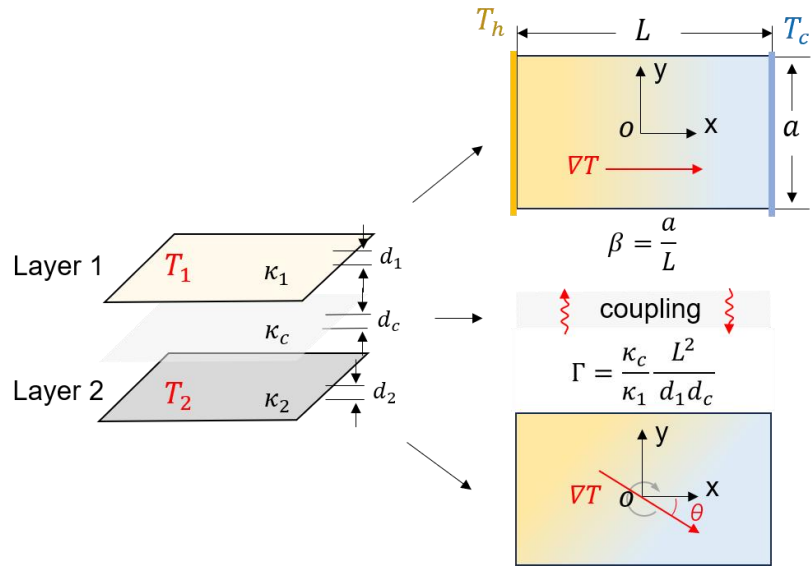
Supplementary Figure 1. Material properties defined in COMSOL Multiphysics simulations for the thermoelectric layer. (A) Electrical conductivity, (B) Seebeck coefficient, (C) Thermal conductivity, and (D) Figure of merit ZT .



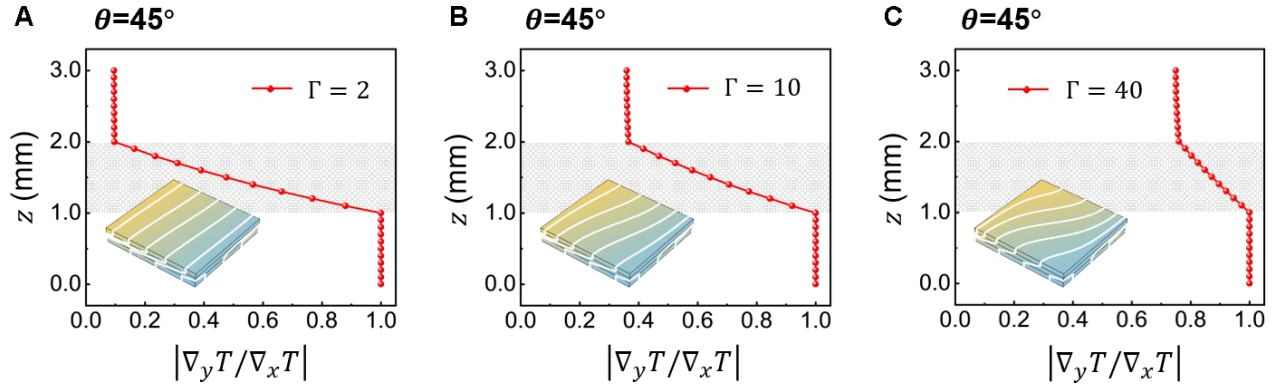
Supplementary Figure 2. Photographs and circuit schematics of the thermoelectric voltage measurement system. (A) Optical photograph of the FPC test board integrated with the thermoelectric chip and 100× pre-channel amplifier circuit. (B), (C) Circuit wiring diagrams for transverse and longitudinal thermoelectric voltage acquisition, respectively, of the hot-side and cold-side electrodes.



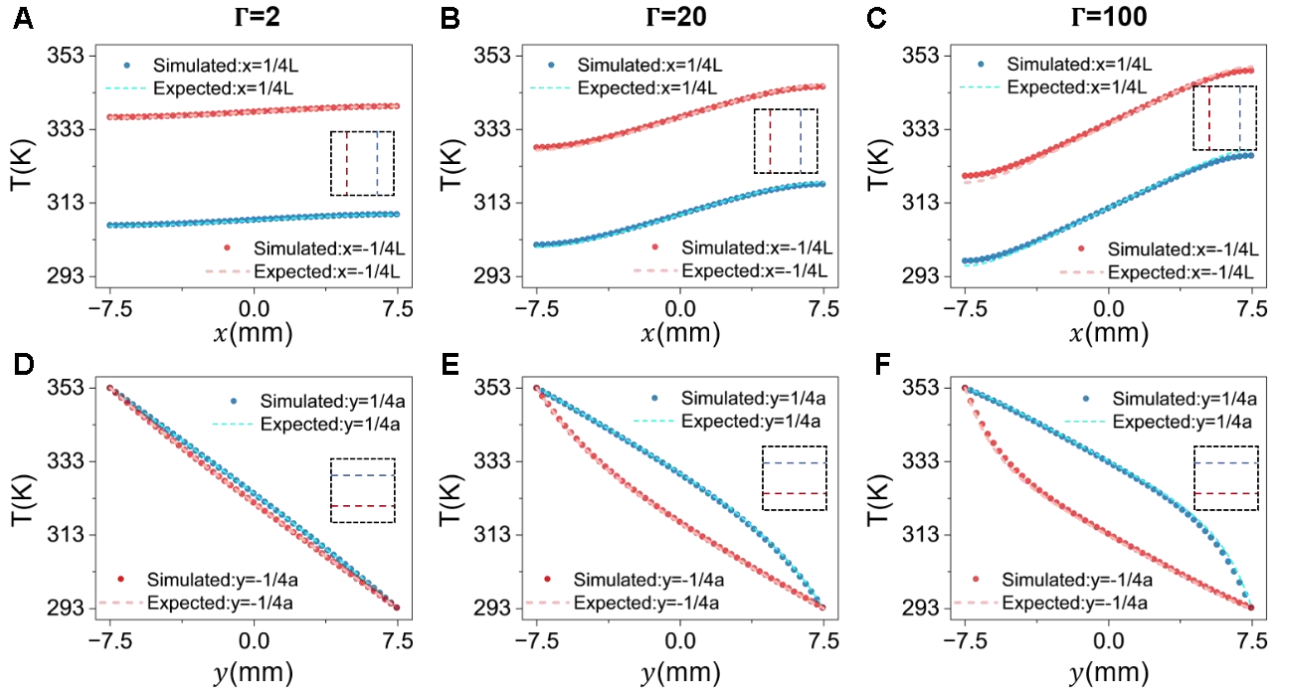
Supplementary Figure 3. Optical image of the experimental setup for characterizing the transverse thermoelectric performance of the flexible device.



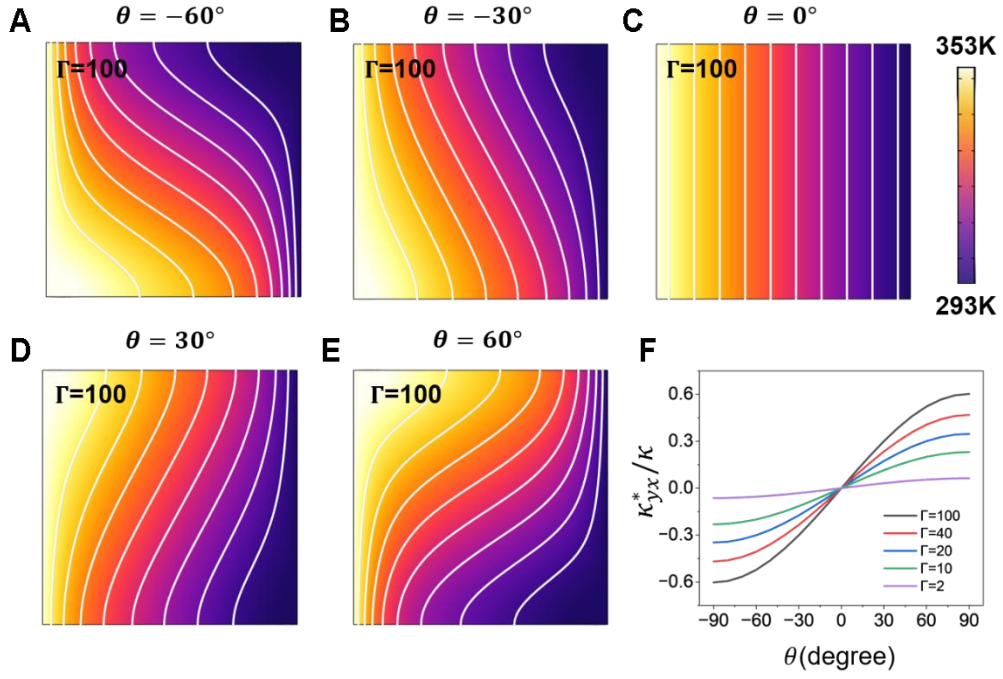
Supplementary Figure 4. Schematic diagram of thermal coupling between two stacked rectangular bars.



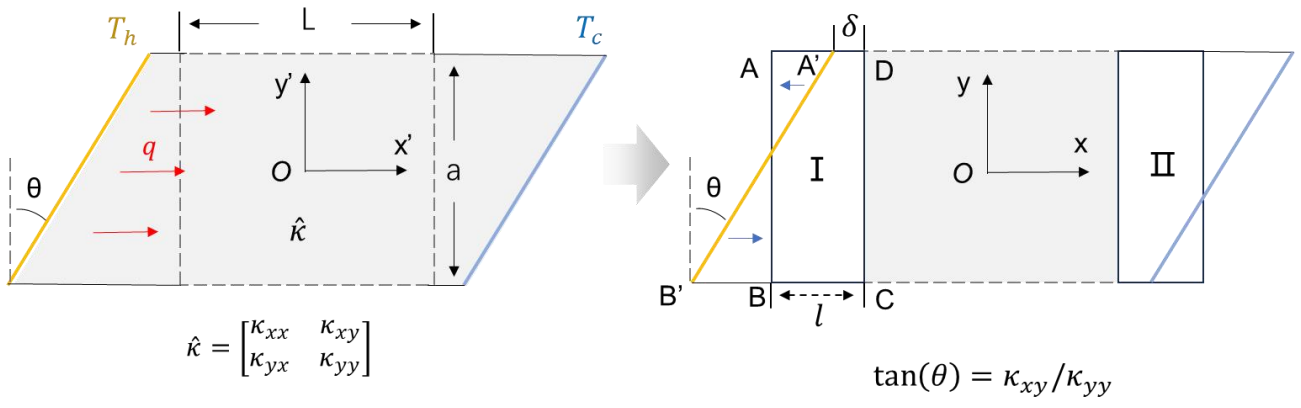
Supplementary Figure 5. Vertical profiles of the temperature gradient deflection ratio $|\nabla_y T / \nabla_x T|$ along the thickness coordinate z at fixed thermal layer deflection angle $\theta=45^\circ$ for three coupling parameters: (A) $\Gamma=2$, (B) $\Gamma=10$, (C) $\Gamma=40$. The gray shaded region corresponds to the intermediate coupling interlayer. The value of $|\nabla_y T / \nabla_x T|$ is nearly uniform within both the upper thermoelectric layer and the lower thermal layer, which supports the adoption of a two-dimensional coupled approximation for subsequent theoretical derivations.



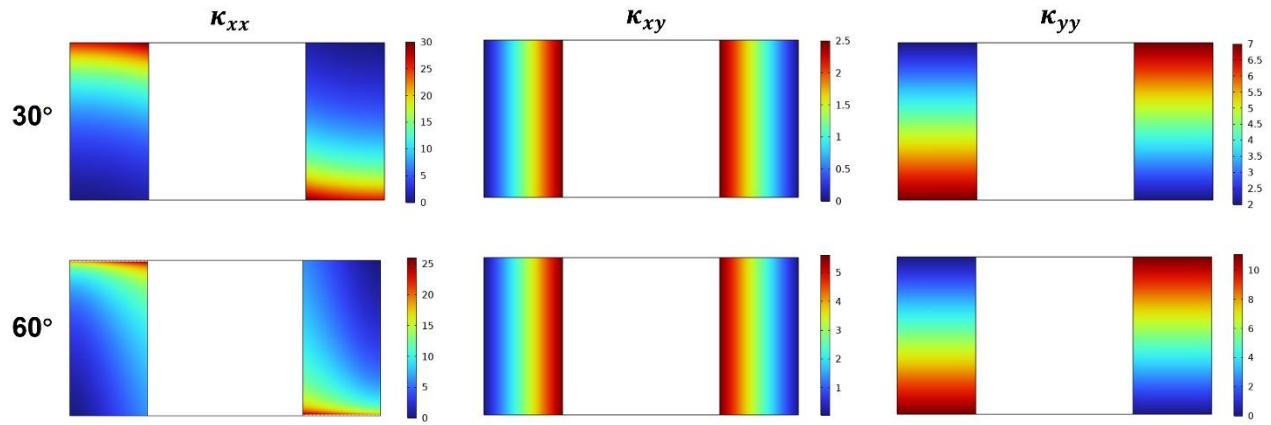
Supplementary Figure 6. Comparison between theoretical predictions and finite-element simulations at the representative cut lines. (Top row) Transverse temperature profiles along the lines at $x = \pm 1/4L$. (Bottom row) Longitudinal temperature profiles along the lines at $y = \pm 1/4a$. From left to right, the columns correspond to the interlayer coupling strengths $\Gamma = 2, 20$, and 100 , respectively. Dashed lines denote theoretical expectations, while symbols denote finite-element simulation results. In the top row, blue and red symbols correspond to $x = 1/4L$ and $x = -1/4L$, respectively; in the bottom row, they correspond to $y = 1/4a$ and $y = -1/4a$, respectively.



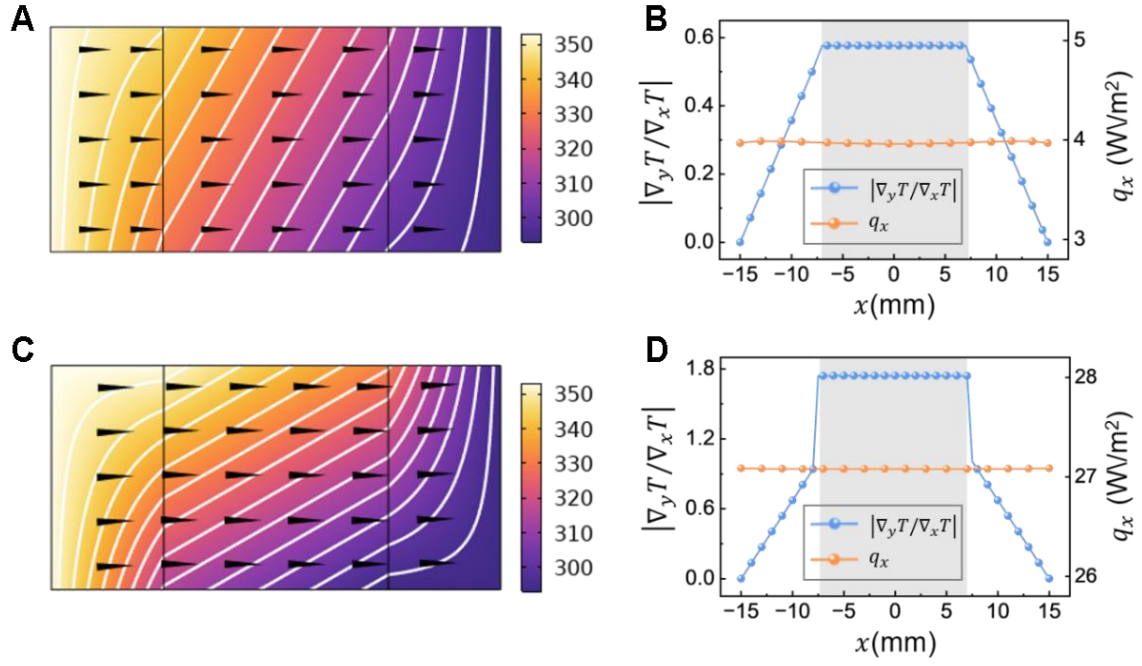
Supplementary Figure 7. Thermal field distributions and effective off-diagonal thermal conductivity induced by interlayer thermal coupling under varied thermal layer deflection angles at fixed coupling parameter $\Gamma=100$. (A–E) Simulated temperature contours and isotherms for deflection angles $\theta=-60^\circ, -30^\circ, 0^\circ, 30^\circ, 60^\circ$. (F) Normalized effective off-diagonal thermal conductivity κ_{yx}^*/κ plotted against deflection angle θ for multiple coupling parameters Γ .



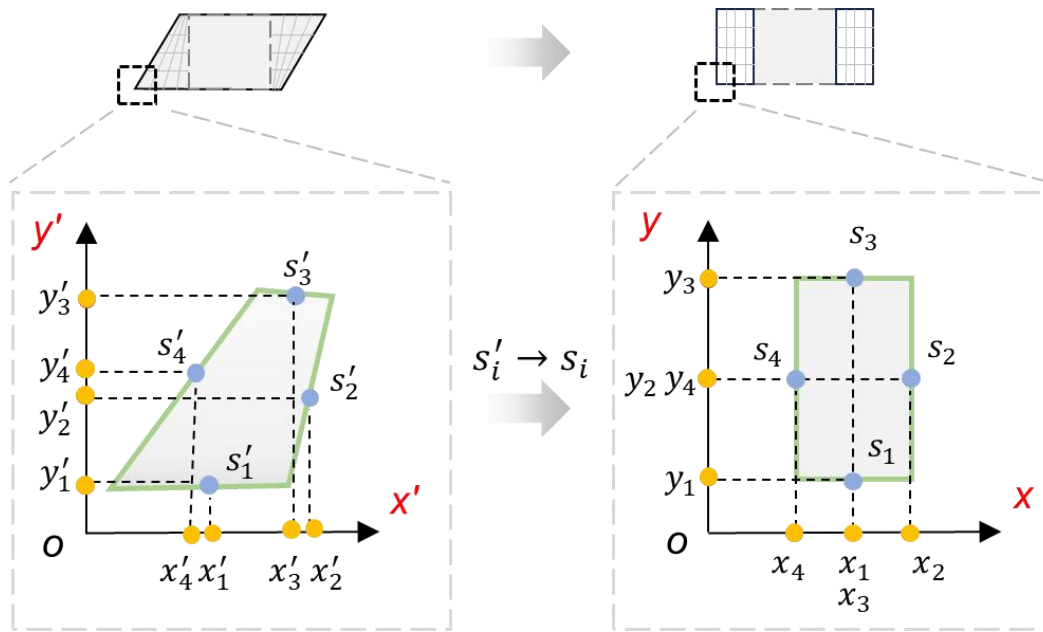
Supplementary Figure 8. Schematic diagram of the boundary region design principle.



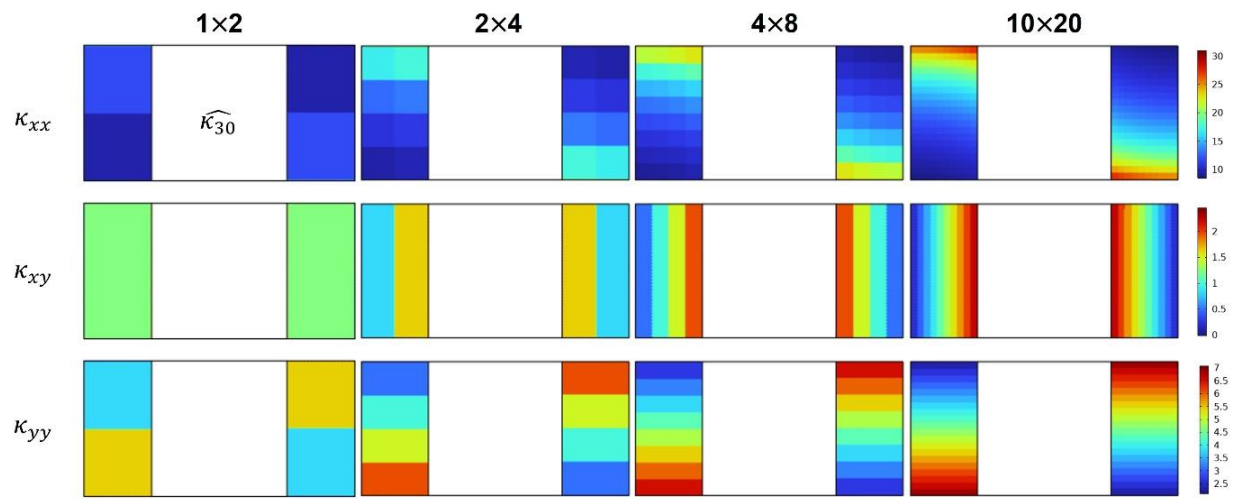
Supplementary Figure 9. Thermal conductivity tensor component distributions in the boundary regions for 30° and 60° thermal field deflections.



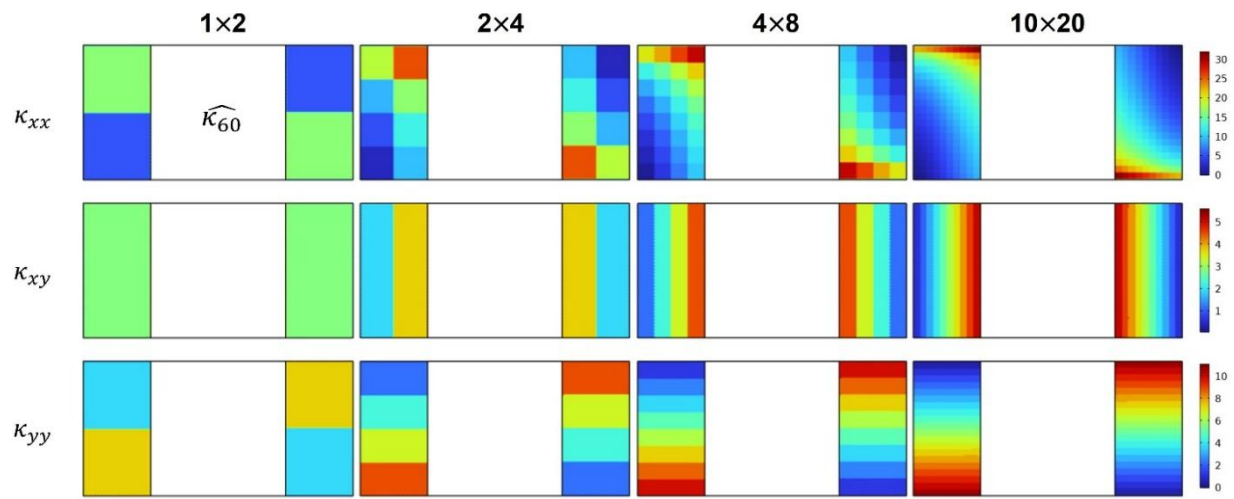
Supplementary Figure 10. Simulation results for thermal metamaterials with integrated boundary regions. Temperature field distribution (A), x -axis profiles of deflection angle and heat flux (B) for the 30° configuration. (C, D) Corresponding results for the 60° configuration.



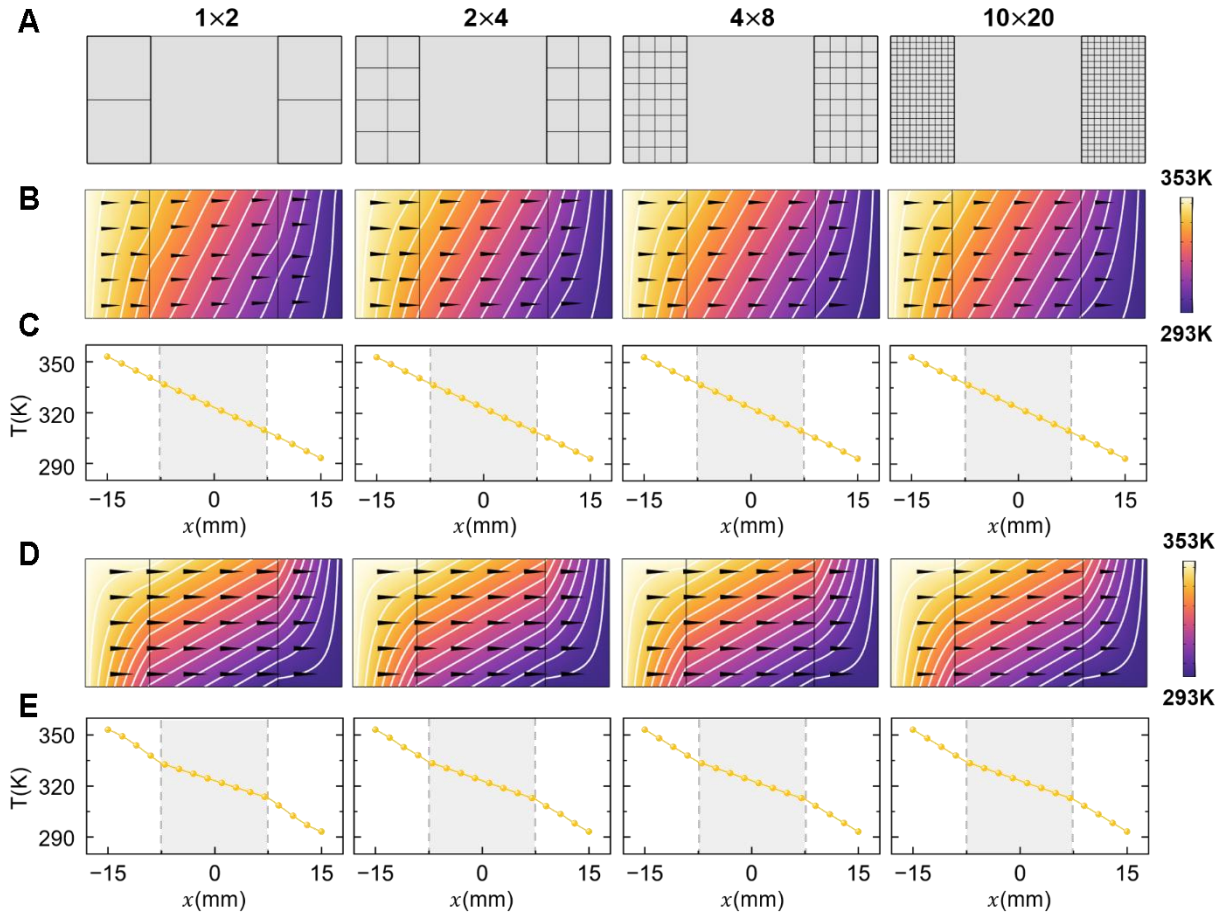
Supplementary Figure 11. Schematic of finite-element coordinate transformation.



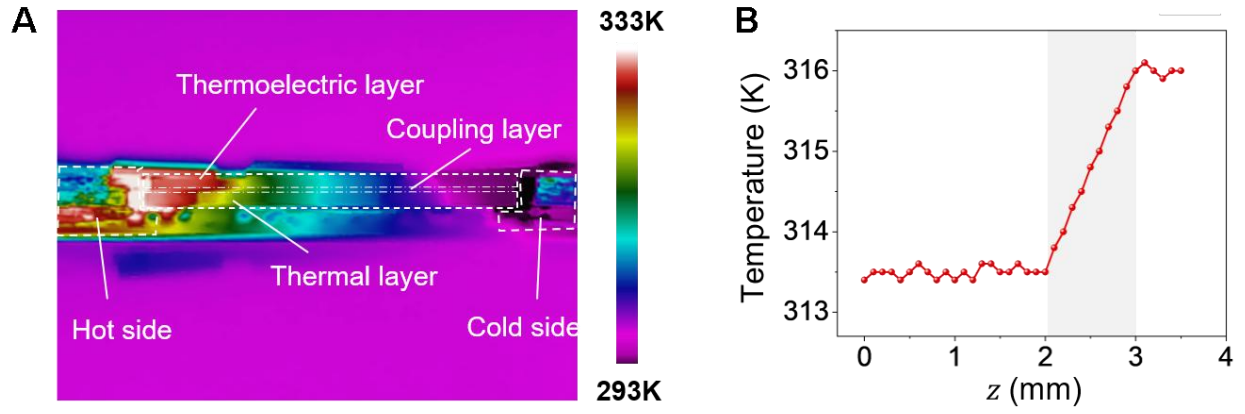
Supplementary Figure 12. Thermal conductivity tensor component distributions in the boundary region for the 30° configuration under varying finite element densities.



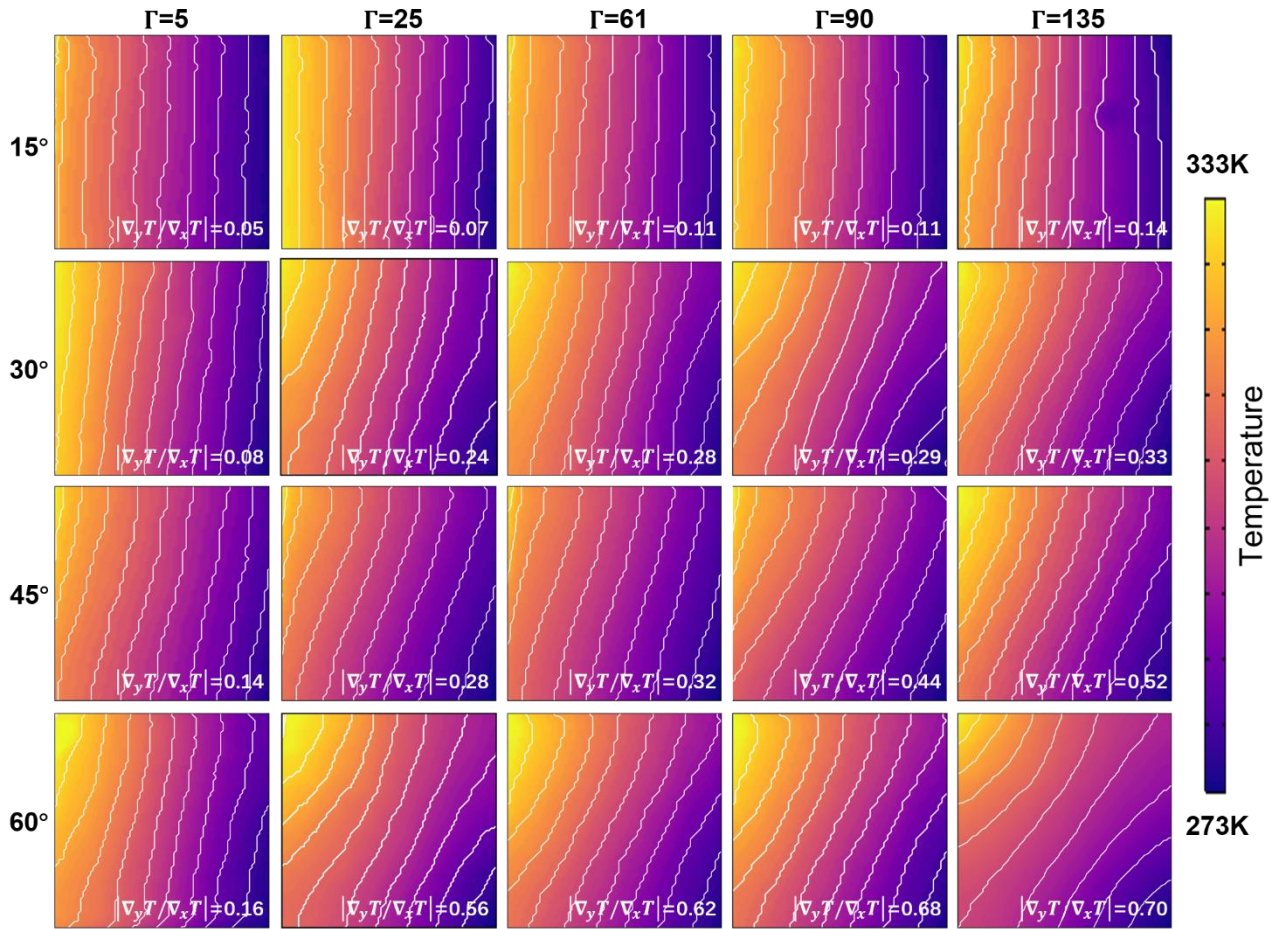
Supplementary Figure 13. Corresponding thermal conductivity distributions for the 60° configuration with progressively refined mesh densities.



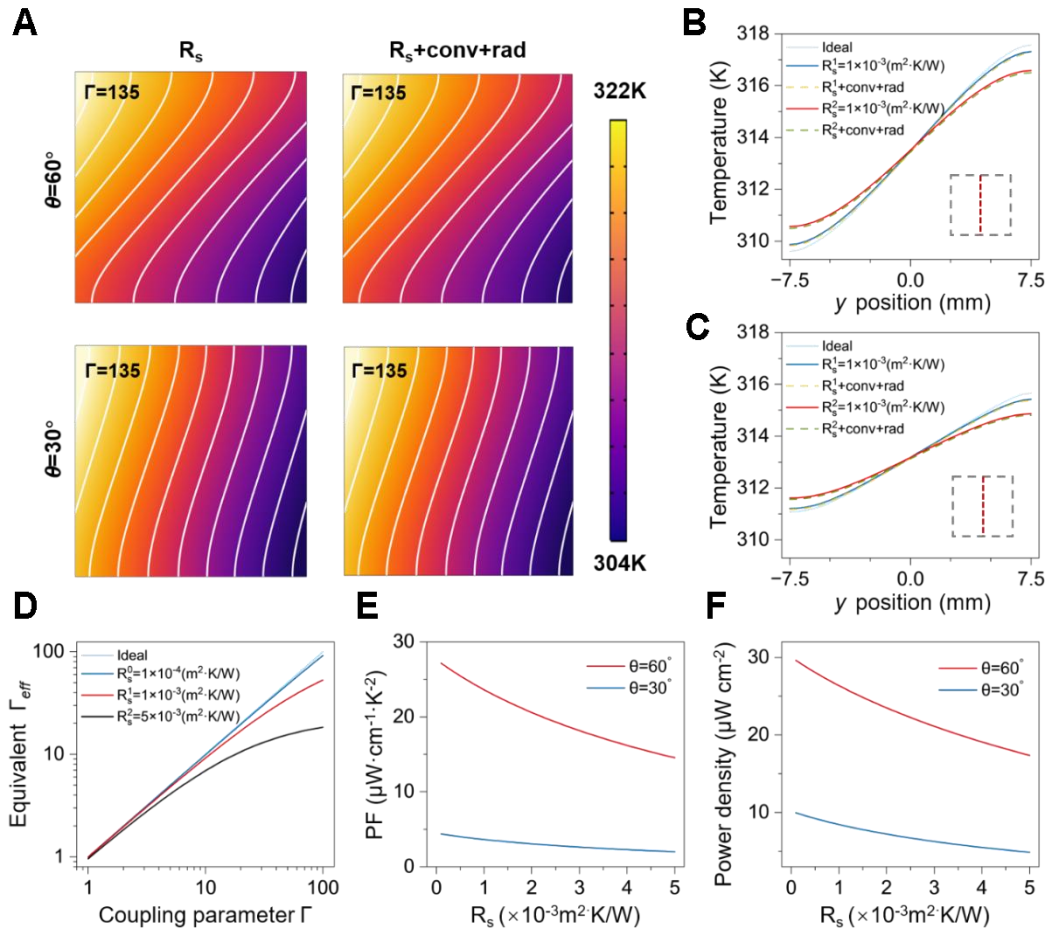
Supplementary Figure 14. Comparative finite element simulations under varying mesh densities. (A) Discretization schemes with different element densities. (B) Thermal field distributions and (C) temperature profiles along the x -axis for the 30° configuration. (D), (E) Corresponding thermal fields and line-scan temperature distributions for the 60° configuration.



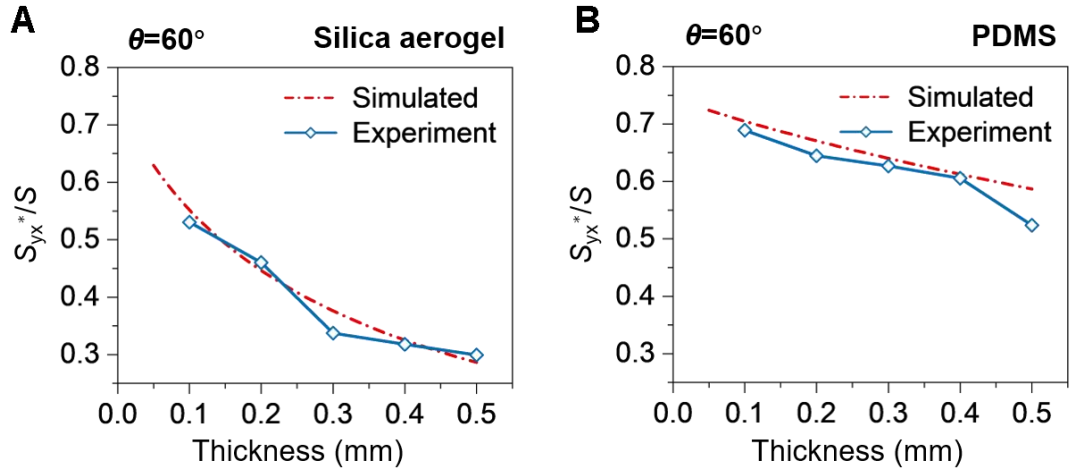
Supplementary Figure 15. Experimental validation of through-thickness temperature uniformity. (A) Side-view infrared thermograph of the bilayer device under $\Gamma=135$ and $\theta=60^\circ$, showing the thermoelectric layer, coupling layer, and thermal layer. (B) Temperature profile along the z-axis (thickness direction). The gray shaded region corresponds to the coupling layer. The nearly flat temperature profiles within the thermoelectric and thermal layers confirm that the through-thickness temperature gradient is negligible, validating the two-dimensional approximation adopted in the theoretical model.



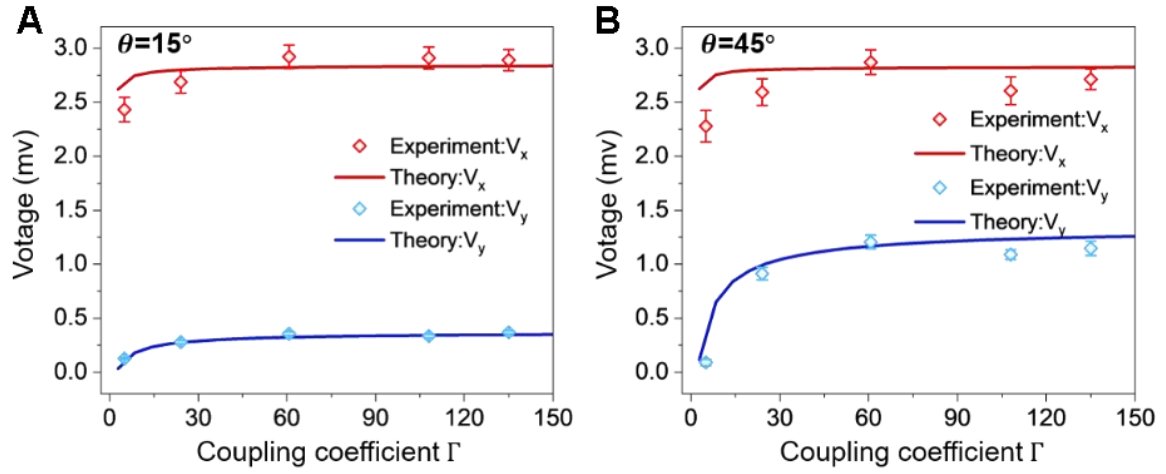
Supplementary Figure 16. Measured temperature contour distributions of the upper thermoelectric layer at four thermal layer deflection angles (15°, 30°, 45°, 60°) and five interlayer coupling parameters ($\Gamma=5, 25, 61, 90, 135$). White solid lines represent isotherms, and the value $|\nabla_y T / \nabla_x T|$ at the bottom-right of each subplot quantifies the thermal field deflection magnitude.



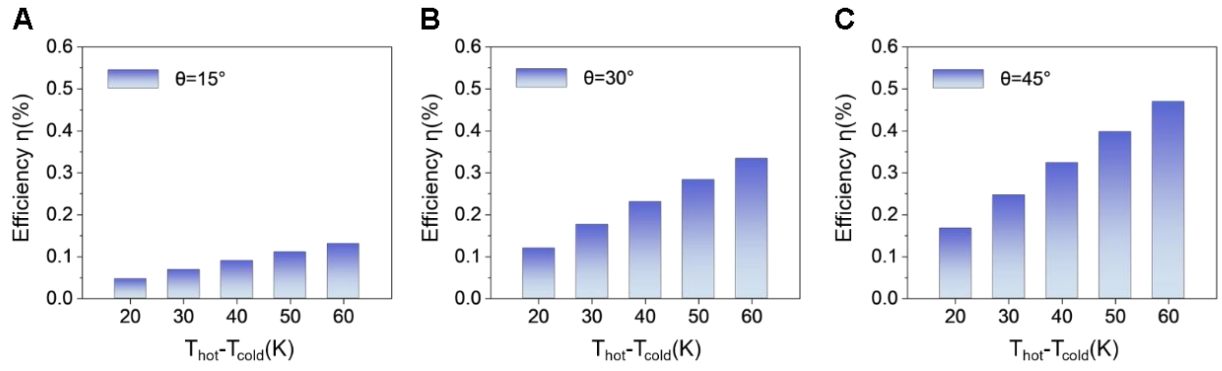
Supplementary Figure 17. Numerical simulation results accounting for the effects of interlayer thermal contact resistance, convection, and radiative heat loss. (A) Temperature distributions in the thermoelectric layer under $\Gamma = 135$ with deflection angles of 30° and 60° for the thermal conductive layer. The left column corresponds to the case considering only contact thermal resistance ($R_s = 1 \times 10^{-3} \text{ m}^2 \cdot \text{K/W}$), while the right column includes all three loss mechanisms, i.e., contact resistance, convection ($h = 5 \text{ W}/(\text{m}^2 \cdot \text{K})$) and radiation (emissivity $\varepsilon = 0.4$, ambient temperature $T_{\text{amb}} = 298 \text{ K}$) under laboratory conditions. (B) and (C) show the temperature profiles along the y-axis of the thermoelectric layer for deflection angles of 60° (B) and 30° (C), respectively, comparing the ideal case (without losses) with cases considering different loss combinations. The results indicate that the influences of convection and radiative heat loss are negligible. (D) Relationship between the effective coupling parameter Γ_{eff} and the ideal Γ value when contact thermal resistance is taken into account. (E) Power factor (PF) as a function of contact thermal resistance R_s , with the red curve for $\theta = 60^\circ$ and the blue curve for $\theta = 30^\circ$. (F) Output power density as a function of contact thermal resistance R_s , with the red curve for $\theta = 60^\circ$ and the blue curve for $\theta = 30^\circ$.



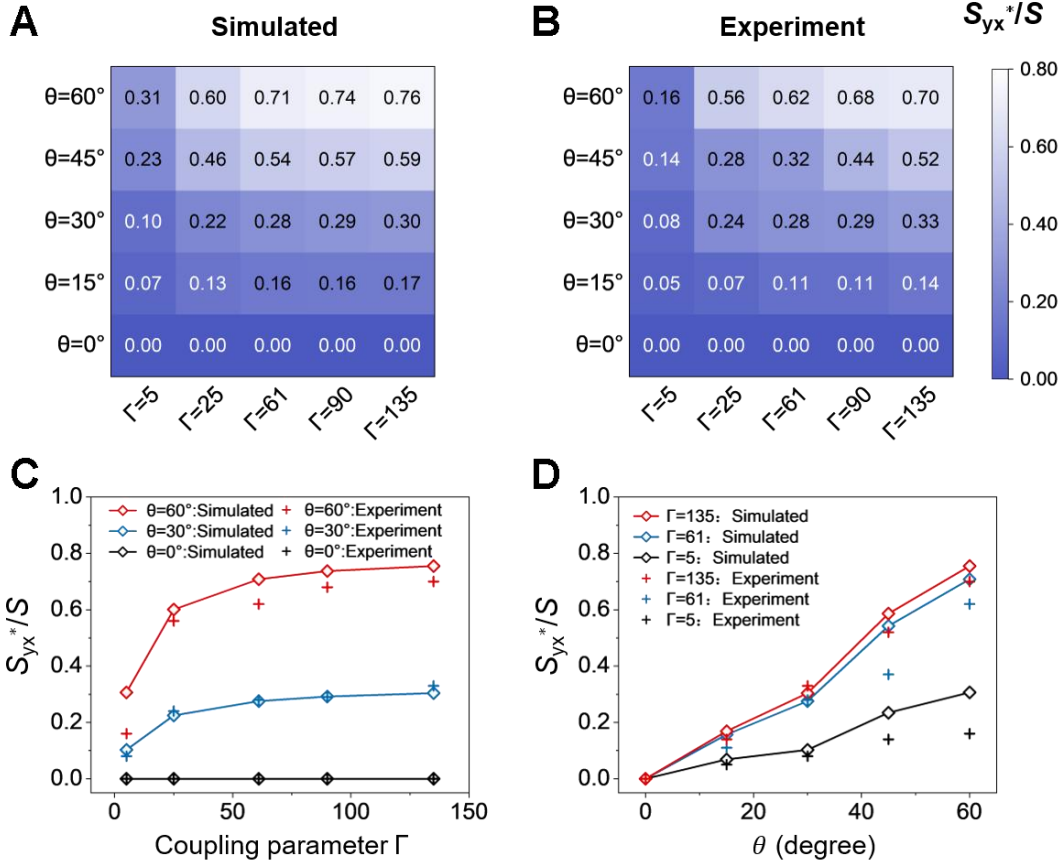
Supplementary Figure 18. Effective Seebeck coefficient as a function of coupling layer thickness for different materials. (A) Silica aerogel and (B) PDMS coupling films of varying thickness were integrated into the bilayer device with a fixed bottom-layer deflection angle of $\theta=60^\circ$ and an applied temperature difference of $\Delta T=40$ K. Dashed lines and dots represent the simulated and experimental results of the normalized off-diagonal Seebeck coefficient (S_{yx}^*/S), respectively.



Supplementary Figure 19. (A) Experimental open-circuit voltages and numerical predictions for longitudinal (V_x , red) and transverse (V_y , blue) configurations at $\theta=15^\circ$ under an applied temperature difference of 40 K. Error bars denote the standard deviation (SD) of five repeated measurements ($n = 5$). (B) Corresponding experimental and theoretical voltages for $\theta=45^\circ$.

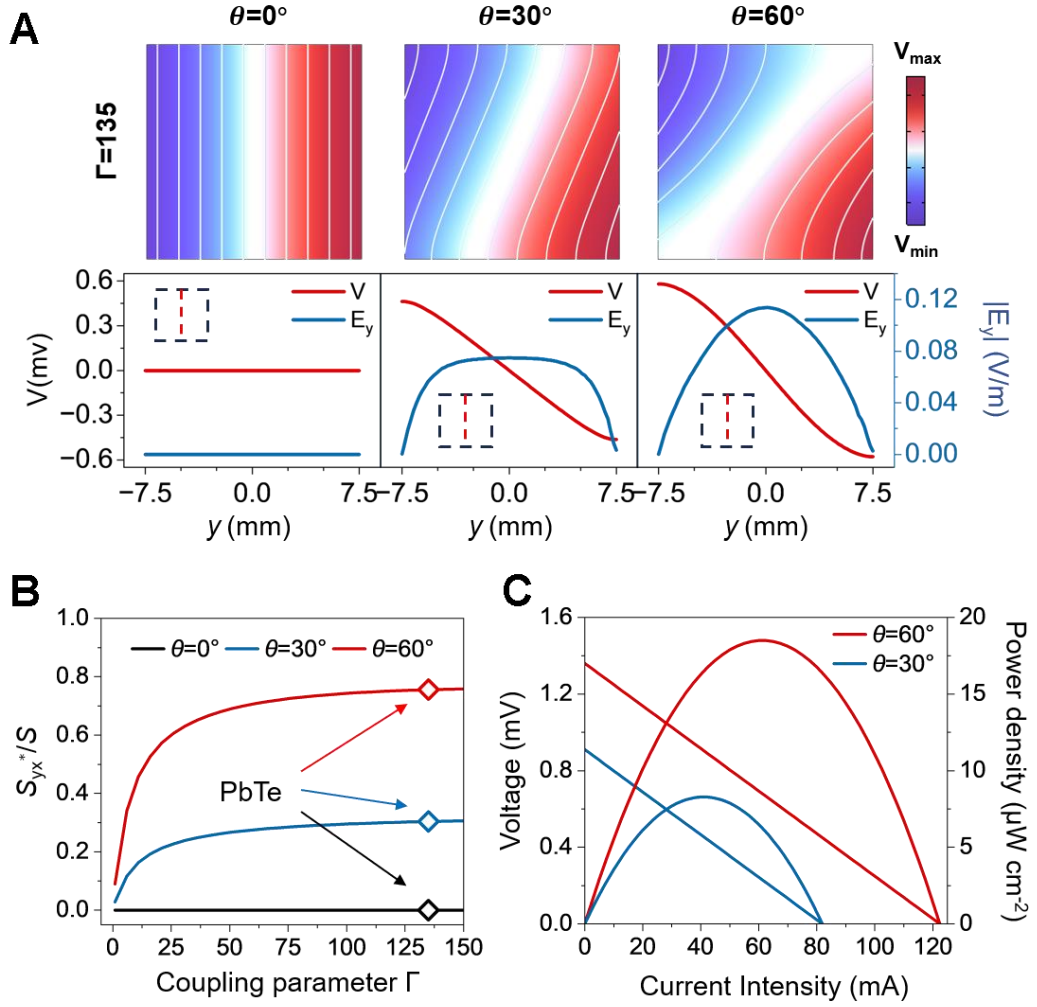


Supplementary Figure 20. Bar charts of the transverse thermoelectric conversion efficiency η measured with thermal layer deflection angles of 15° (A), 30° (B), and 45° (C).

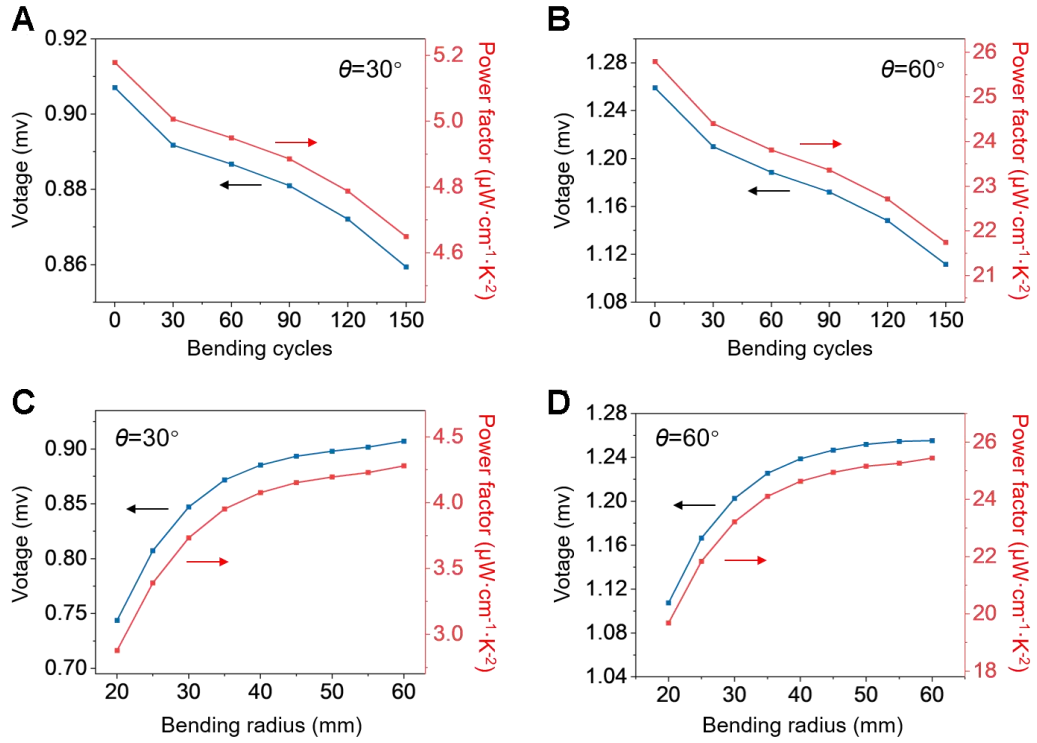


Supplementary Figure 21. Systematic error analysis of interlayer thermal coupling. (A, B)

Simulated (A) and experimental (B) normalized off-diagonal Seebeck coefficient S_{yx}^*/S as functions of the lower-layer deflection angle θ and the dimensionless coupling parameter Γ . (C) S_{yx}^*/S versus Γ for $\theta=0^\circ, 30^\circ$ and 60° . (D) S_{yx}^*/S versus θ for $\Gamma=5, 61$ and 135 . Cross markers denote experimental data; open diamonds with solid lines denote simulated results.

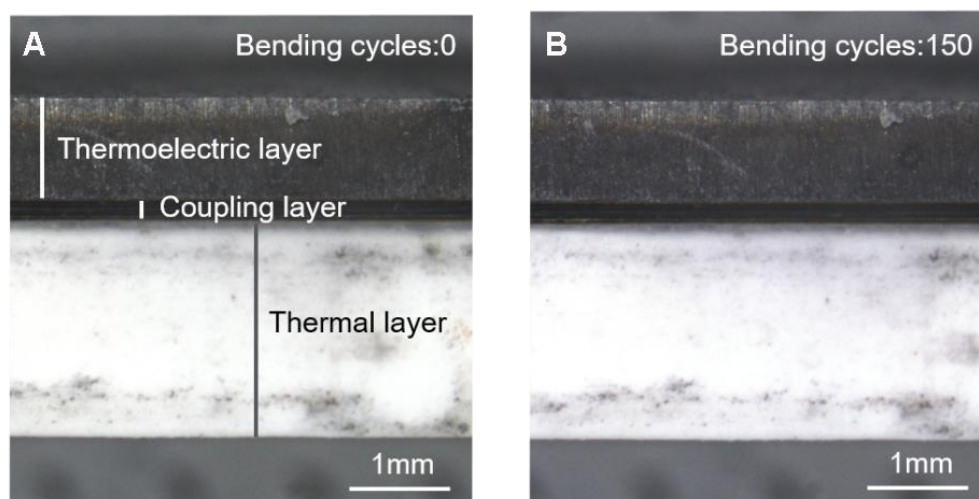


Supplementary Figure 22. Finite-element simulation results for the material-agnostic verification in PbTe thermoelectric material. (A) Simulated electric potential contours (upper panels) and corresponding vertical-center-axis electric field profiles (lower panels) of the thermoelectric layer under different deflection angles. The simulations are performed at $\Gamma = 135$ with deflection angles $\theta = 0^\circ$, 30° , and 60° . The red and blue lines denote the electric potential V and the transverse electric field E_y , respectively. (B) Ratio of the off-diagonal effective Seebeck tensor component induced by interlayer thermal coupling to the intrinsic Seebeck coefficient, S_{yx}^*/S , as a function of the coupling parameter Γ . The curve represents the theoretical results based on Bi_2Te_3 from Figure 4D, while the three discrete data points are obtained from the PbTe simulations in (A) for $\theta = 0^\circ$, 30° , and 60° , respectively. The excellent agreement between the PbTe data points and the Bi_2Te_3 theoretical curve demonstrates that the transverse thermoelectric response is material-agnostic. (C) Current–voltage characteristics and corresponding power density curves for the flexible devices with deflection angles $\theta = 30^\circ$ (blue) and 60° (red).



Supplementary Figure 23. Mechanical durability and flexibility characterization. (A, B)

Transverse open-circuit voltage and power factor of the flexible device with $\theta=30^\circ$ (A) and $\theta=60^\circ$ (B) as a function of bending cycles at a fixed bending radius of 50 mm. (C, D) Corresponding transverse open-circuit voltage and power factor for $\theta=30^\circ$ (C) and $\theta=60^\circ$ (D) measured at various static bending radii. All tests were conducted under an applied temperature difference of $\Delta T = 40$ K.



Supplementary Figure 24. Cross-sectional optical micrographs of the flexible bilayer device (A) as-fabricated and (B) after 150 bending cycles at a radius of 50 mm. The three-layer architecture (thermoelectric layer, coupling layer, and thermal layer) remains well-defined, with continuous and intimate interfacial contact preserved after cyclic bending. No delamination or macroscopic interfacial defects are observed. Scale bar, 1 mm.

Supplementary Table 1. Thin films with different thermal resistances used in experiments and their corresponding coupling parameters

Material	Thermal conductivity(W/(m·K))	Thickness(mm)	Dimensionless coupling strength Γ
Polyimide (PI) film	0.3	0.25	135
Polytetrafluoroethylene (PTFE) film	0.24	0.3	90
Polydimethylsiloxane (PDMS) film	0.27	0.5	61
Silica aerogel thermal insulation film	0.022	0.1	25
Silica aerogel thermal insulation film	0.022	0.5	5

Supplementary Table 2. Calculation results of thermal layer designs with various deflection angles based on effective medium theory

	Central region					Boundary region				
θ	$\kappa_{Central,A}$	$f_{Central,A}$	$\kappa_{Central,B}$	$f_{Central,B}$	α	$\kappa_{Boundary,A}$	$f_{Boundary,A}$	$\kappa_{Boundary,B}$	$f_{Boundary,B}$	α
0°	30	0.5	-	-	0°	30	0.29	-	-	0°
15°	30	0.5	1	0.5	2.3°	30	0.37	2.5	0.63	14.3°
30°	30	0.5	2	0.5	11.9°	30	0.53	1.4	0.47	15.8°
45°	30	0.5	1	0.5	10.2°	30	0.37	1	0.63	34.7°
60°	30	0.5	0.1	0.5	1.5°	30	0.4	0.3	0.6	36°

Supplementary Table 3. Comparison of performance and characteristics among different transverse thermoelectric strategies

Category	Research	External conditions	Effective Seebeck coefficient $ S_{yx}^* $ ($\mu\text{V}\cdot\text{K}^{-1}$)	Effective power factor ¹ $\text{PF}(\mu\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-2})$	Heat-flux deflection angle	Extensibility ratio ²
Coupling thermoelectric	This work	None	165.9	2751	0°~90°	~300
Longitudinal TE	Commercial Bi_2Te_3	None	~220	~3500	-	-
Artificially tilted multilayer (ATML)	[14]	0.8 T	57.1	1580	$30\pm 1^\circ$	7.5
	[15]	None	120.38	7150	$10^\circ/20^\circ/45^\circ$	3.2
	[16]	None	420	6800	$11^\circ/21^\circ/45^\circ$	3.0 ~ 4.0
	[17]	None	171.25	4000	16°	3.33
Ordinary Nernst Effect (ONE)	[18]	0.8 T	7.5	47.8	-	8.0
	[19]	1.2T	15.2 ± 0.4	1.21	-	498
	[20]	1T	4.5	0.5~1	-	26.7
	[21]	1T	6.5	30	-	12.5
Topological thermoelectric	[22]	0.7T, 80K	275	37000	-	1.8-2.3
	[23]	9T, 37.7K	1045	3.1×10^5	-	5~10
	[24]	5 T, 35 K	600	85000	-	5~10
	[25]	14T, 14 K	617	20393	-	3.3~5.0

¹The effective power factor of this work is measured under an applied temperature difference of $\Delta T=40$ K and a thermal layer deflection angle of 60° . The high power factors reported for topological thermoelectric materials are achieved under extreme conditions (cryogenic temperatures and strong magnetic fields).

²The extensibility ratio is defined as the in-plane characteristic dimension divided by the total thickness, characterizing the device thinness and scalability.

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