Supplementary information

Giant thermal conductivity and strain thermal response of nitrogen substituted diamane：A machine-learning-based prediction

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Details of MTPs:

MTPs is a type of local potentials. In this context, the total energy E of a sample containing N atoms is the sum of contributions Ω from the neighborhoods o_i of each i-th atom.

$$E \equiv E^{MTP} = \sum_{i=1}^{N} \Omega(o_i) \tag{1}$$

The neighborhood of a central atom is defined as a set.

$$o_i = \left(\{r_{i1}, z_i, z_1\} \ldots, \{r_{ij}, z_i, z_j\} \ldots, \{r_{iN_{\text{neigh}}}, z_i, z_{N_{\text{neigh}}}\}\right) \tag{2}$$

In this context, the j-th atom is referred to as a neighboring atom within a predefined cutoff radius R_{cut} from the i-th (central) atom. z_i and z_j denote the types of the central and neighboring atoms, respectively, r_{ij} represents the corresponding interatomic vector, and N_{neigh} is the number of atoms in the neighborhood. The contribution of each central atom and its associated neighborhood to the total system energy is expressed as:

$$\Omega(u_i) = \sum_{\alpha} \xi_{\alpha} B_{\alpha}(o_i) \tag{3}$$

B_α represents a basis function, where ξ_α is an MLIPs parameter. The basis functions are constructed based on all possible contractions of moment tensor descriptors, generating the following scalar:

$$M_{\mu,v}(r_i) = \sum_{j=1}^{N_{\text{nei}}} f_{\mu}\left(|r_{ij}|, z_i, z_j\right) r_{ij}^{\otimes v} \tag{3}$$

$f_{\mu}\left(|r_{ij}|, z_i, z_j\right)$ is the radial component of potential MLIP, which depends solely on the distance between atoms i and j and their atomic types. The radial component is

obtained by multiplying a set of radial basis functions $\varphi_\beta\left(\left|r_{ij}\right|\right)$ by a smoothing factor $\left(R_{\text{cut}}-\left|r_{ij}\right|\right)^2$.

$$f_\mu\left(\left|r_{ij}\right|,z_i,z_j\right)=\sum_\beta c^{(\beta)}_{\mu,z_i,z_j}\varphi_\beta\left(\left|r_{ij}\right|\right)\left(R_{\text{cut}}-\left|r_{ij}\right|\right)^2 \tag{4}$$

$c^{(\beta)}_{\mu,z_i,z_j}$ are the radial coefficients (parameters). The MTP parameters of ξ_α and $c^{(\beta)}_{\mu,z_i,z_j}$ are acquired by solving the minimization problem of:

$$\sum_{k=1}^{K}\left[w_e\left(E_k^{\text{AIMD}}-E_k^{\text{MTP}}\right)^2+w_f\sum_i^N\left|f_{k,i}^{\text{AIMD}}-f_{k,i}^{\text{MTP}}\right|^2+w_s\sum_{i,j=1}^{3}\left|\sigma_{k,ij}^{\text{AIMD}}-\sigma_{k,ij}^{\text{MTP}}\right|^2\right]\rightarrow\min \tag{5}$$

Linearized phonon BTE:

At the temperature *T*, the phonon distribution in the crystal obeys the Bose–Einstein distribution in thermodynamic equilibrium state, as follows:

$$f_\lambda^0 = \frac{1}{e^{\hbar\omega/k_BT} - 1} \tag{6}$$

A temperature gradient drives a phonon heat current by diverting phonon distribution from the equilibrium distribution, as follows:

$$f_\lambda = f_\lambda^0 - \frac{\partial f_\lambda^0}{\partial E_\lambda}\Psi_\lambda = f_\lambda^0 + f_\lambda^0\left(1 + f_\lambda^0\right)\Psi_\lambda \tag{7}$$

The resulting non-zero phonon heat flux **J** can be expressed as follows [1]:

$$\mathbf{J} = \frac{1}{NV}\sum_\lambda \hbar\omega_\lambda \mathbf{v}_\lambda f_\lambda = \frac{1}{k_BT^2NV}\sum_\lambda f_\lambda^0\left(1 + f_\lambda^0\right)\left(\hbar\omega_\lambda\right)^2 \mathbf{v}_\lambda\left(\mathbf{F}_\lambda \cdot \nabla T\right) \tag{8}$$

where λ includes the phonon branch index p and wave vector q; ω_λ and $\mathbf{v}_\lambda$ are the angular frequency and group velocity of phonon mode λ, respectively; $\mathbf{F}_\lambda$ is the mean free displacement of phonons; N is the number of q points in the first Brillouin zone; V is the volume of the unit cell; and T is the temperature.

According to Fourier's law $\mathbf{J}^\alpha = -\sum_\beta \kappa^{\alpha\beta}(\nabla T)^\beta$, the coefficient of thermal conductivity can be obtained as follows:

$$\kappa^{\alpha\beta} = \frac{1}{k_BT^2NV}\sum_\lambda f_\lambda^0\left(1 + f_\lambda^0\right)\left(\hbar\omega_\lambda\right)^2 v_\lambda^\alpha F_\lambda^\beta \tag{9}$$

where $\kappa^{\alpha\beta}$ represents κ corresponding to the heat flow generated in the α direction by the temperature gradient in the β direction.

The linearized BTE $\mathbf{F}_\lambda$ can then be written as follows [2]:

$$\mathbf{F}_\lambda = \tau_\lambda^0\left(\mathbf{v}_\lambda + \mathbf{\Delta}_\lambda\right) \tag{10}$$

where $\mathbf{\Delta}_\lambda$ represents the effective change in velocity after scattering and τ_λ^0 is the lifetime of mode λ, the inverse of which is the scattering rate calculated by Matthiessen's rule [3]. The phonon–phonon (anharmonic) and phonon–isotope scatterings with the natural isotopic distribution of diamond are considered. Total

scattering rates $1/\tau_\lambda^0$ are determined as follows:

$$\frac{1}{\tau_\lambda^0}=\frac{1}{\tau_\lambda^{\text{anh}}}+\frac{1}{\tau_\lambda^{\text{iso}}} \tag{11}$$

The phonon–phonon scattering rates require the identification of a set of three-phonon scattering processes that satisfy phonon energy and momentum conservation conditions, as follows [4]:

$$\omega_\lambda \pm \omega_{\lambda'}=\omega_{\lambda''} \text{ and } \mathbf{q}\pm\mathbf{q}'=\mathbf{q}''+\mathbf{K} \tag{12}$$

Where λ, λ', and λ'' represent the three phonons involved; K is the reciprocal lattice vector. K = 0 characterizes momentum-conserving normal processes and K ≠ 0 corresponds to resistive umklapp processes [5]. However, the intrinsic three-phonon scattering rates are treated similarly ("+" represents absorption processions and "–" represents emission processions), as follows:

$$\Gamma^{+}_{\lambda\lambda'\lambda''}=\frac{\hbar\pi}{4}\frac{f_0'-f_0''}{\omega_\lambda\omega_{\lambda'}\omega_{\lambda''}}\left|V^{+}_{\lambda\lambda'\lambda''}\right|^2\delta\left(\omega_\lambda+\omega_{\lambda'}-\omega_{\lambda''}\right) \tag{13}$$

$$\Gamma^{-}_{\lambda\lambda'\lambda''}=\frac{\hbar\pi}{4}\frac{f_0'+f_0''+1}{\omega_\lambda\omega_{\lambda'}\omega_{\lambda''}}\left|V^{-}_{\lambda\lambda'\lambda''}\right|^2\delta\left(\omega_\lambda-\omega_{\lambda'}-\omega_{\lambda''}\right) \tag{14}$$

The Dirac distribution $\delta\left(\omega_\lambda \pm \omega_{\lambda'}-\omega_{\lambda''}\right)$ enforces the conservation of energy in the absorption and emission processes. The scattering matrix elements $V^{\pm}_{\lambda\lambda'\lambda''}$ can be calculated by third-order IFCs, as follows [6]:

$$V^{\pm}_{\lambda\lambda'\lambda''}=\sum_{i\in u.c.}\sum_{j,k}\sum_{\alpha\beta\gamma}\Phi^{\alpha\beta\gamma}_{ijk}\frac{e^{\alpha}_{\lambda}(i)e^{\beta}_{p',\pm\mathbf{q}'}(j)e^{\gamma}_{p'',-\mathbf{q}''}(k)}{\sqrt{M_iM_jM_k}} \tag{15}$$

where $\Phi^{\alpha\beta\gamma}_{ijk}$ is the anharmonic IFCs matrix. It refers to movement of the two atoms in multiple directions in the supercell, followed by calculation of the Hellmann–Feynman (H–F) force after displacement.

Finally, we obtain the anharmonic scattering rates $1/\tau_\lambda^{\text{anh}}$ according to the following equation:

$$\frac{1}{\tau_{\lambda}^{\text{anh}}}=\frac{1}{N}\left(\sum_{\lambda^{'}\lambda^{''}}^{+}\Gamma_{\lambda\lambda^{'}\lambda^{''}}^{+}+\sum_{\lambda^{'}\lambda^{''}}^{-}\frac{1}{2}\Gamma_{\lambda\lambda^{'}\lambda^{''}}^{-}\right) \tag{16}$$

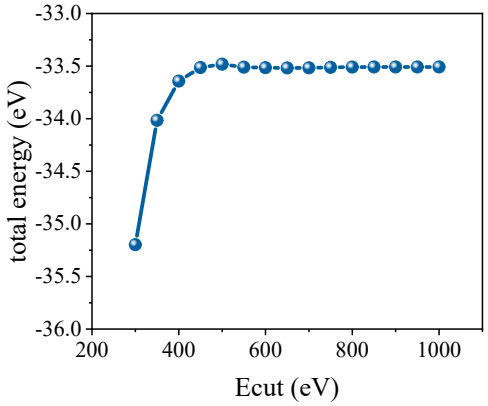
In addition to intrinsic three-phonon scattering, elastic scattering of phonons introduced by isotope impurities is present, as follows [7, 8]:

$$\frac{1}{\tau_{\lambda}^{\text{iso}}}=\frac{1}{N}\sum_{\lambda^{'}}\Gamma_{\lambda\lambda^{'}}=\frac{1}{N}\sum_{\lambda^{'}}\frac{\pi\omega^{2}}{2}\sum_{i\in\text{u.c.}}g(i)\left|\mathbf{e}_{\lambda}^{*}(i)\cdot\mathbf{e}_{\lambda^{'}}(i)\right|^{2}\delta\left(\omega_{\lambda}-\omega_{\lambda^{'}}\right) \tag{17}$$

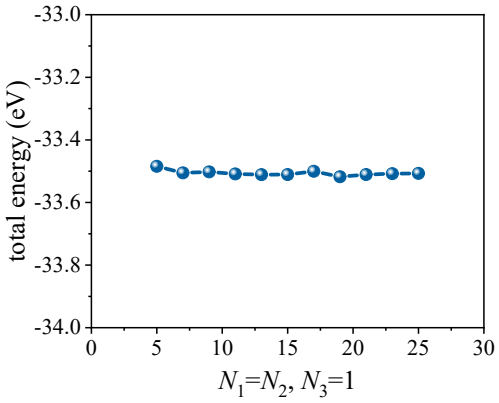
where $g(i)=\sum_{S}f_{S}(i)[1-M_{S}(i)/\overline{M}(i)]^{2}$ is the Pearson deviation coefficient of the masses $M_{S}(i)$ of isotopes *s* of atom *i*.

Convergence tests:

Structural optimization parameter:

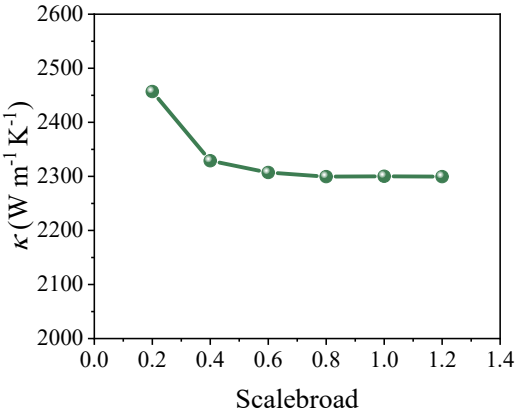


(a) Ecut

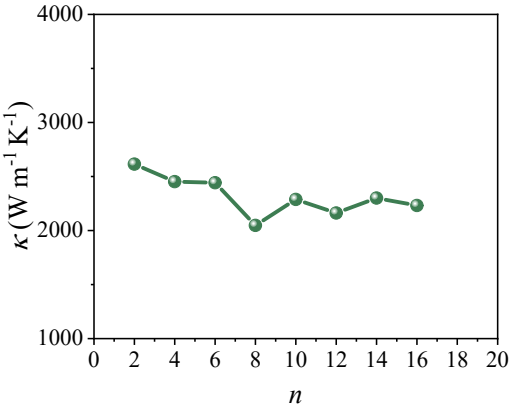


(b) *k*-points

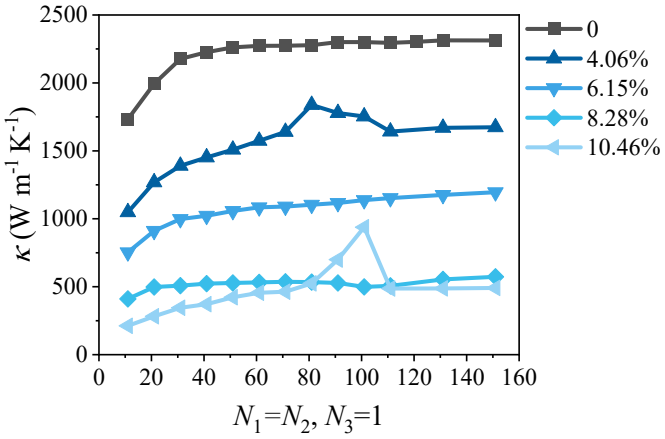
Thermal conductivity:



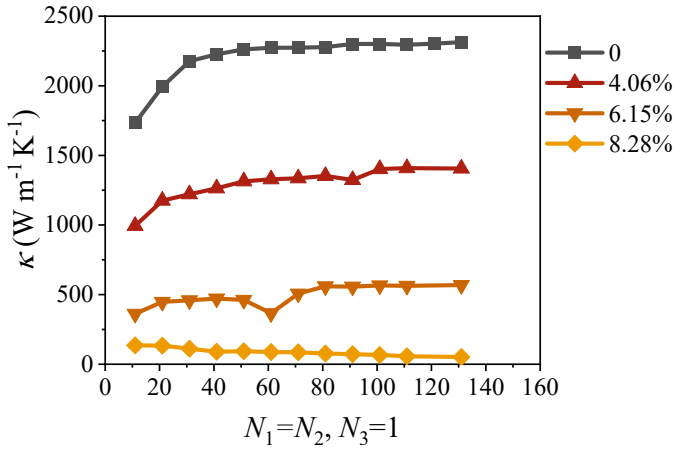
(c) scalebroad



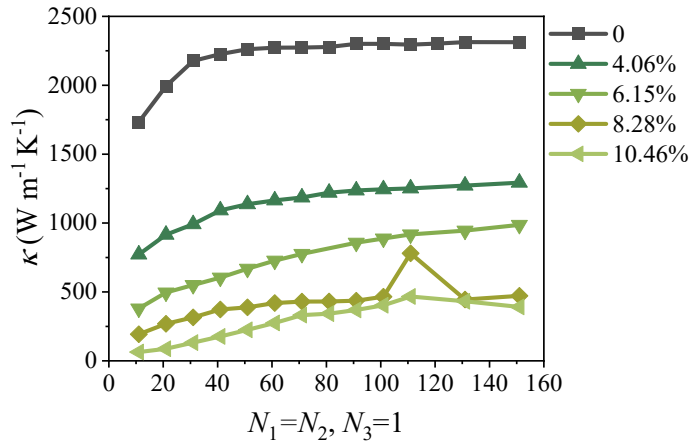
(d) critical atomic number *n* of force cutoff



(e) *q*-points (zigzag strain)



(f) q-points (armchair strain)



(g) q-points (biaxial strain)

Fig. S1. Convergence test

Structural parameter:

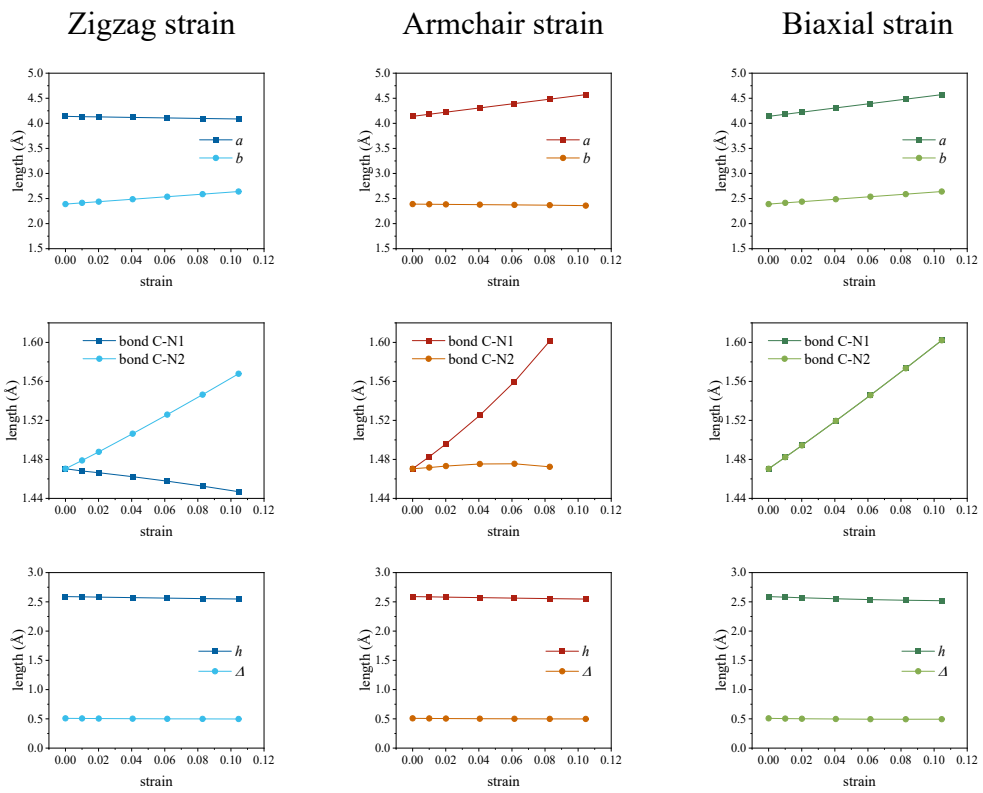


Fig. S2. The structural parameters of NCCN during the strain process

Thermal conductivity:

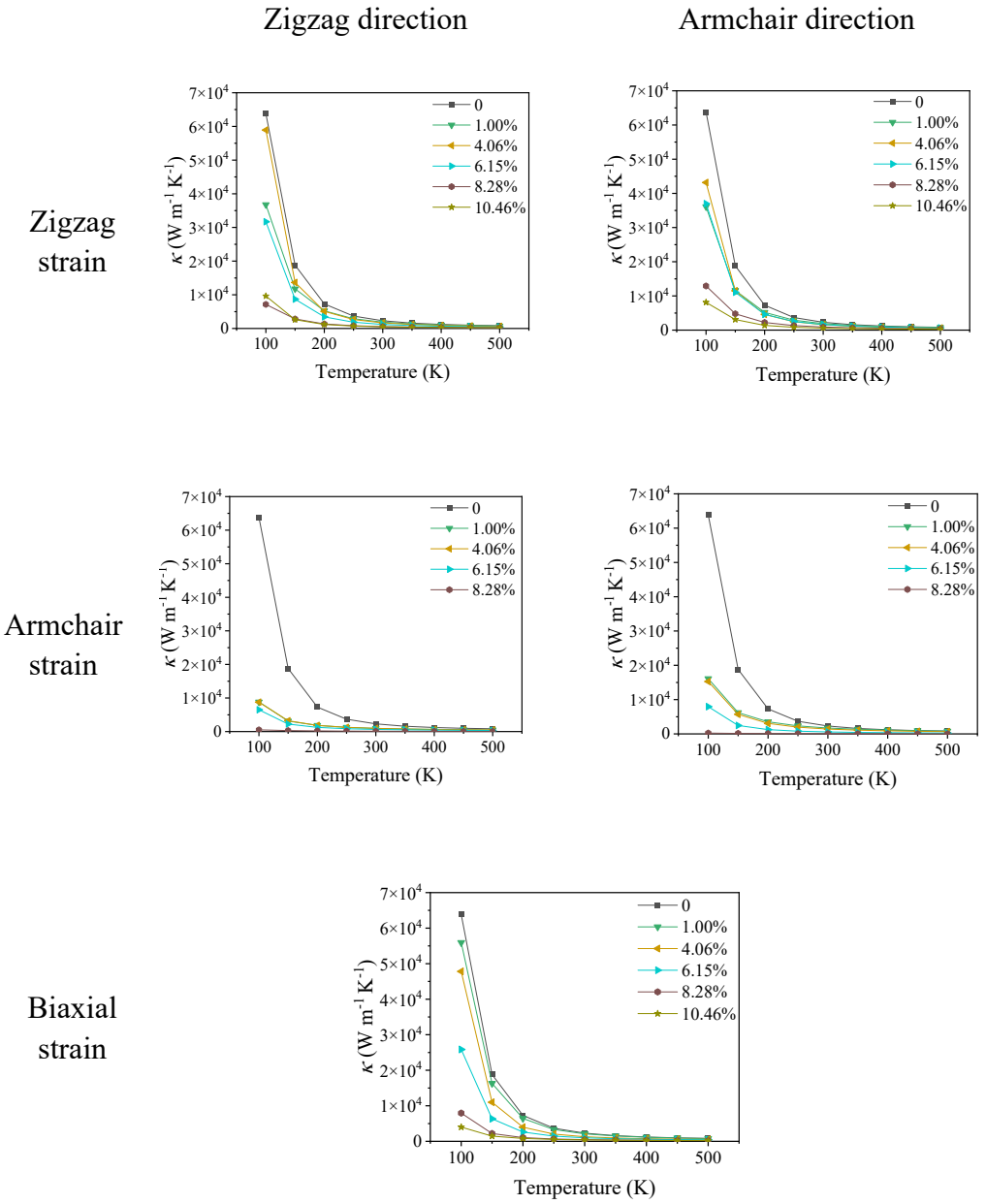
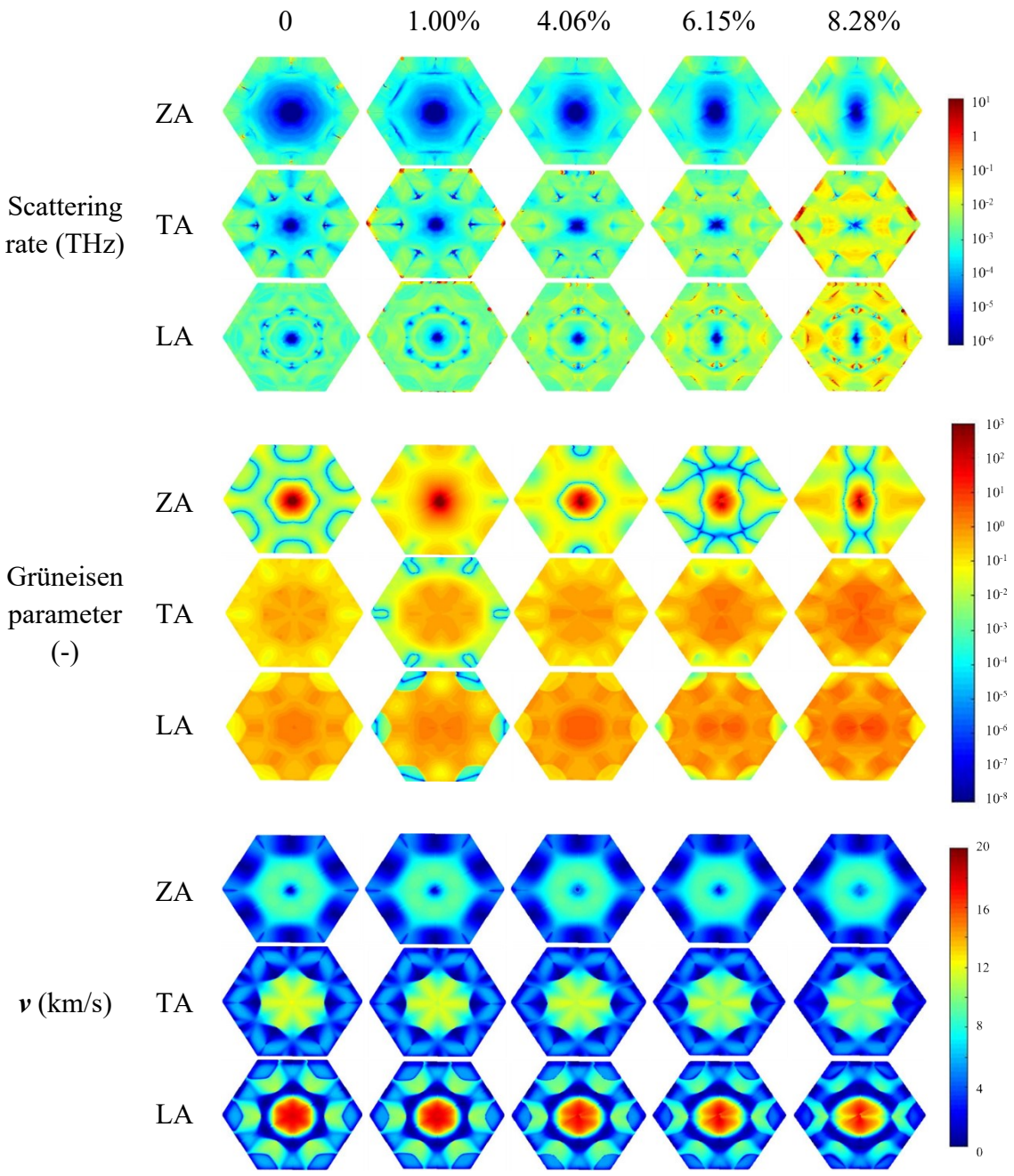
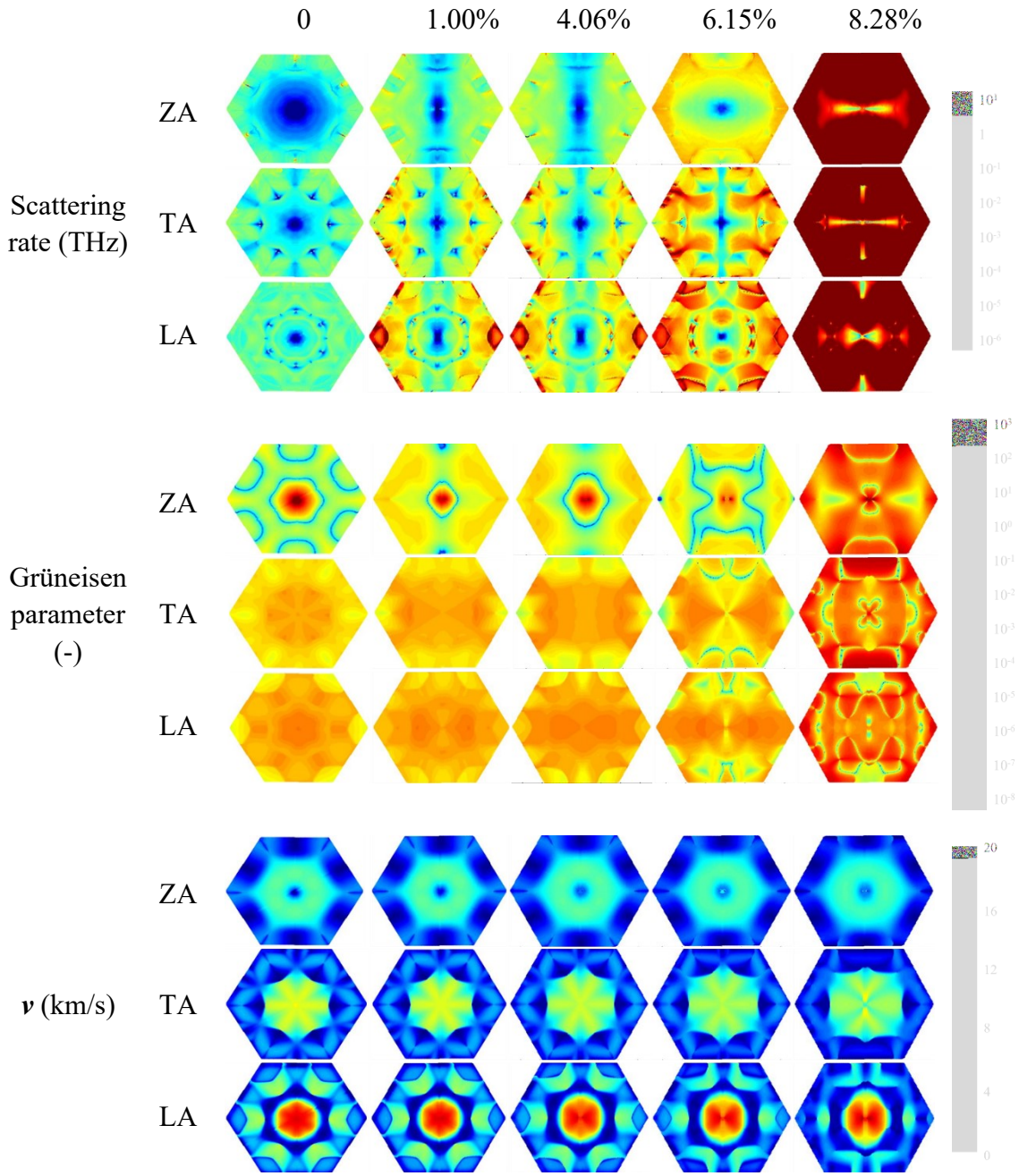


Fig. S3. Change of thermal conductivity with temperature

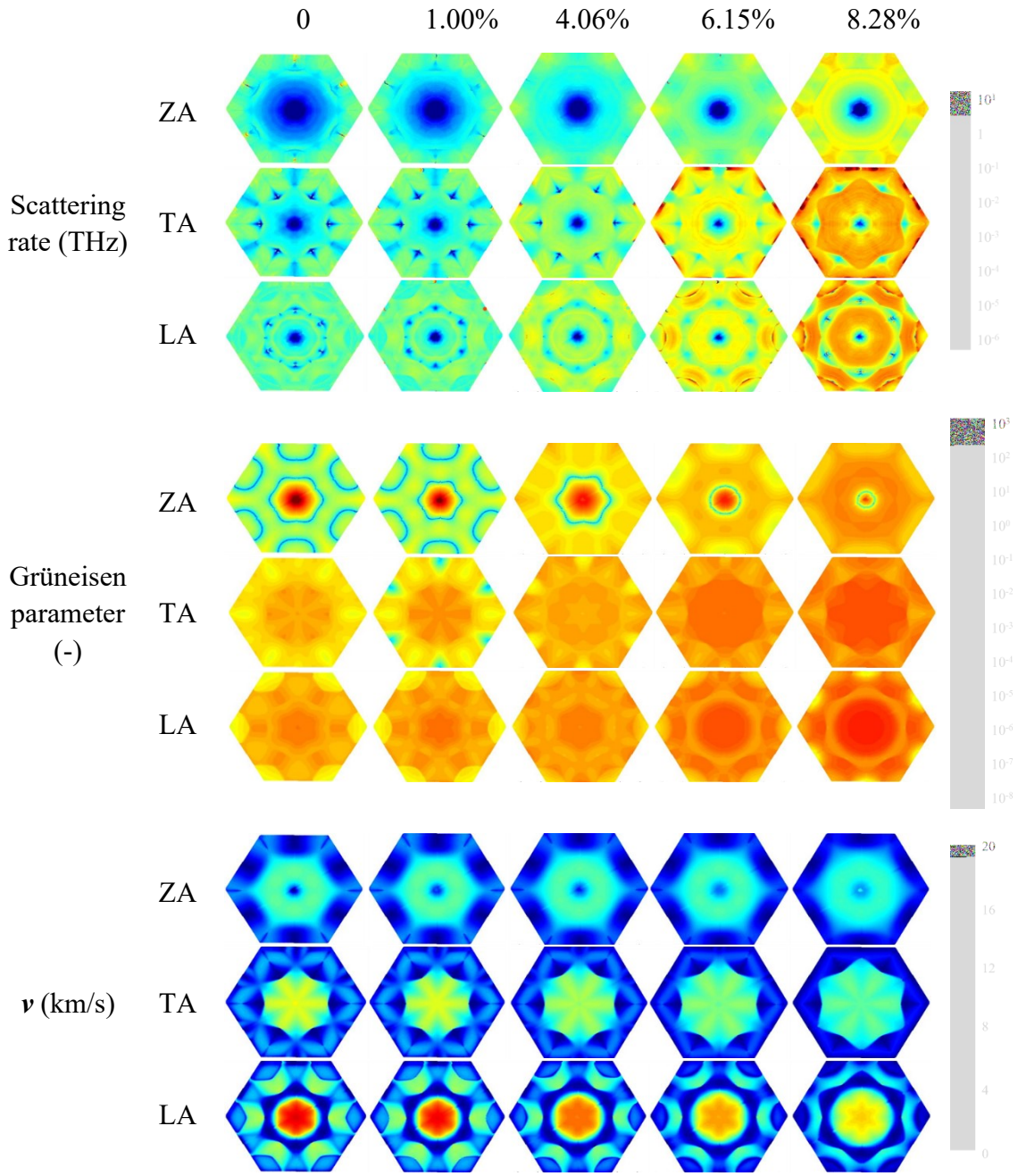
Phonon information:



(a) Zigzag strain



(b) Armchair strain



(c) Biaxial strain

Fig. S4. Distribution of phonon information (scattering rate, Grüneisen parameter, group velocity) in the first Brillouin zone for NCCN. Grüneisen parameters representing the anharmonic effect of phonons.

References

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