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Assessing the Limitations of Transparent Conducting Oxides as Thermoelectrics — Supplementary Information

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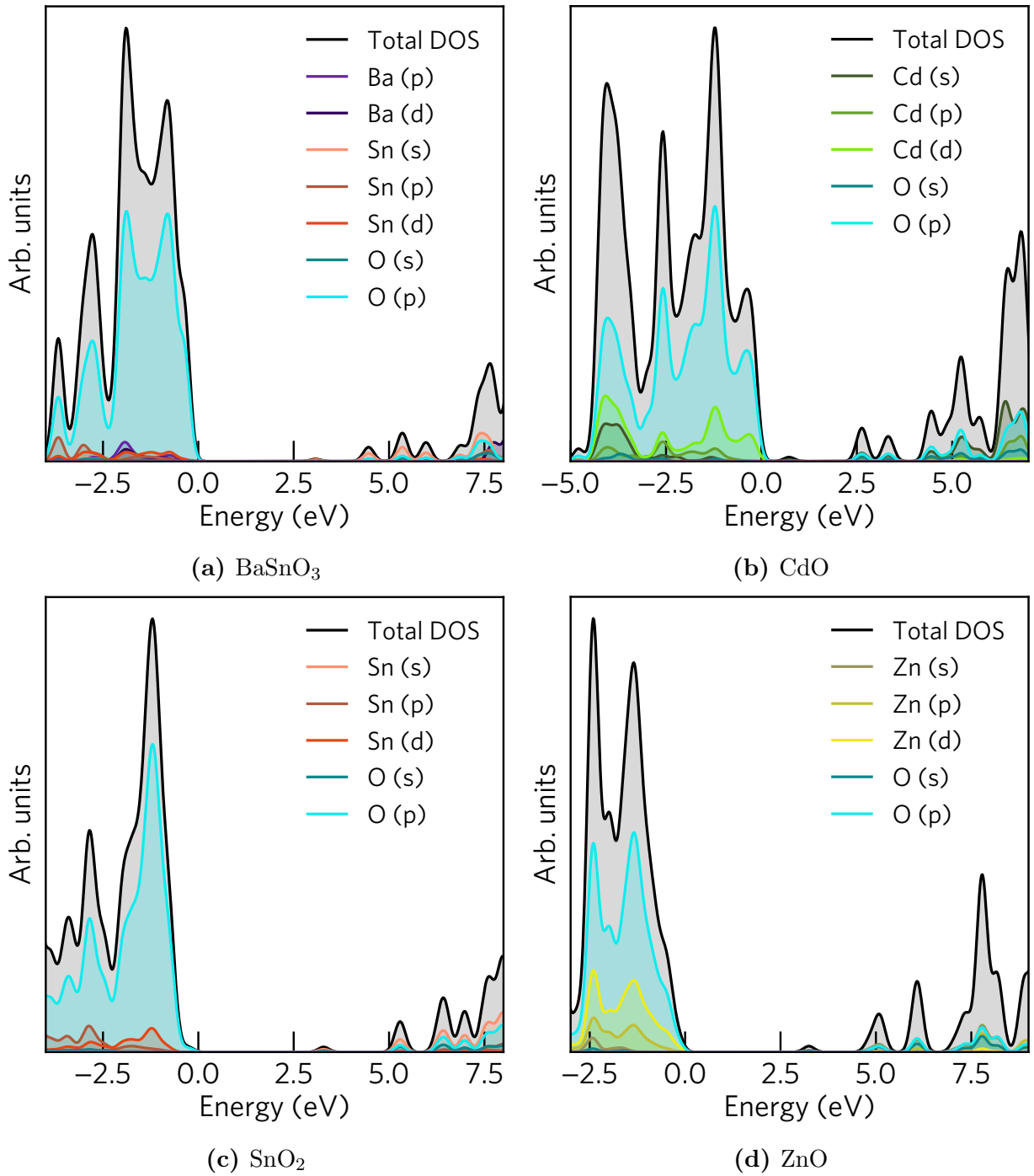
Lattice Parameters

Table S1: Structural parameters for the conventional cells of $BaSnO_3$, CdO, SnO_2 and ZnO relaxed via PBEsol. Values in brackets compare to experiment.[1–4]

Material	a (Å)	b (Å)	c (Å)	α (°)	β (°)	γ (°)
$BaSnO_3$	4.13 (+0.5 %)	4.13 (+0.5 %)	4.13 (+0.5 %)	90	90	90
CdO	4.71 (+0.2 %)	4.71 (+0.2 %)	4.71 (+0.2 %)	90	90	90
SnO_2	4.77 (+0.7 %)	4.77 (+0.7 %)	3.22 (+0.8 %)	90	90	90
ZnO	3.24 (-0.3 %)	3.24 (-0.3 %)	5.22 (+0.2 %)	90	90	120

Electronic Properties



(a) $BaSnO_3$ **(b)** CdO

(c) SnO_2 **(d)** ZnO

Figure S1: The densities of states of (a) $BaSnO_3$, (b) CdO, (c) SnO_2 and (d) ZnO, plotted with sumo.[5] They are normalised so the valence band maximum is at 0 eV.

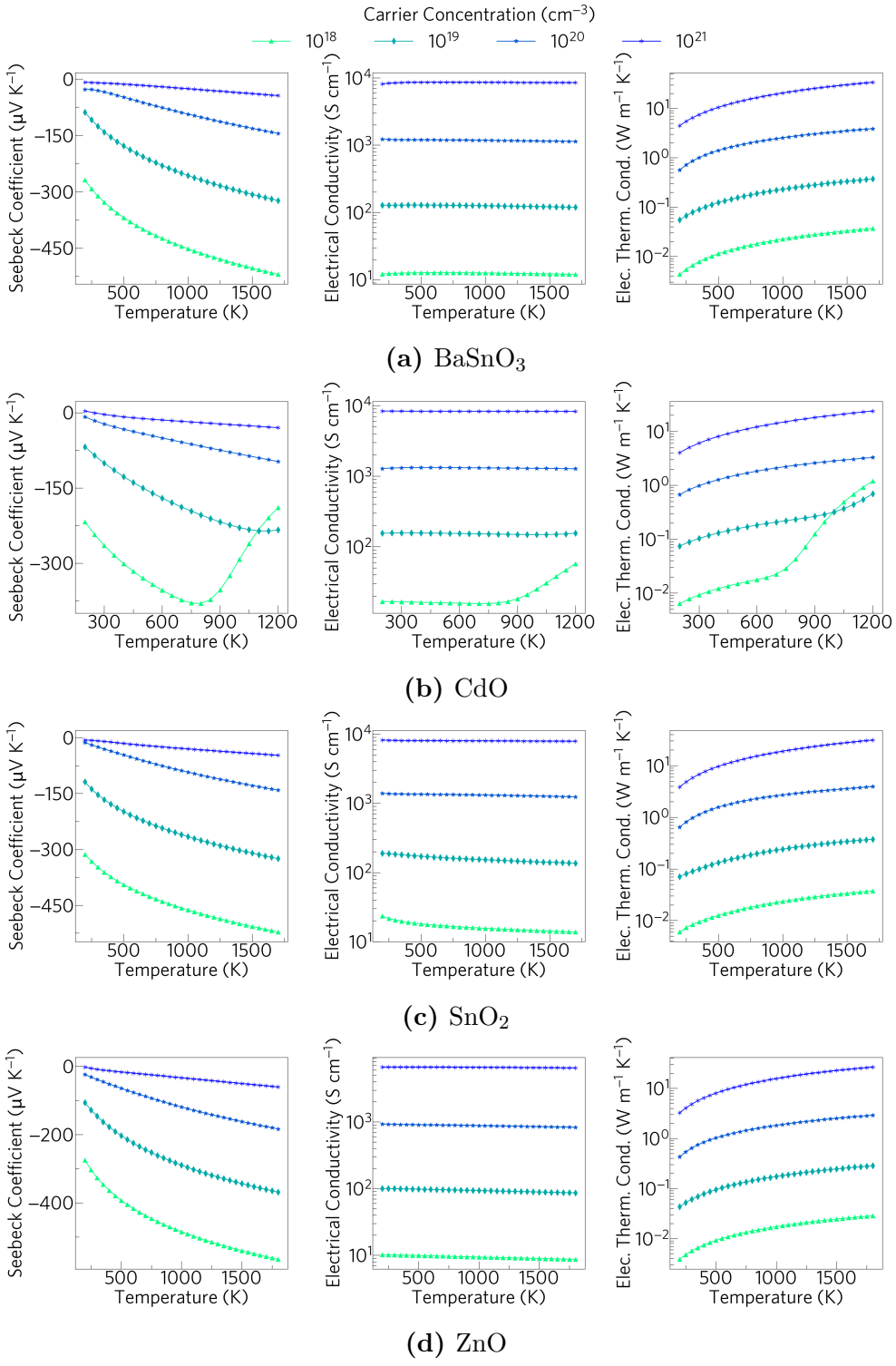
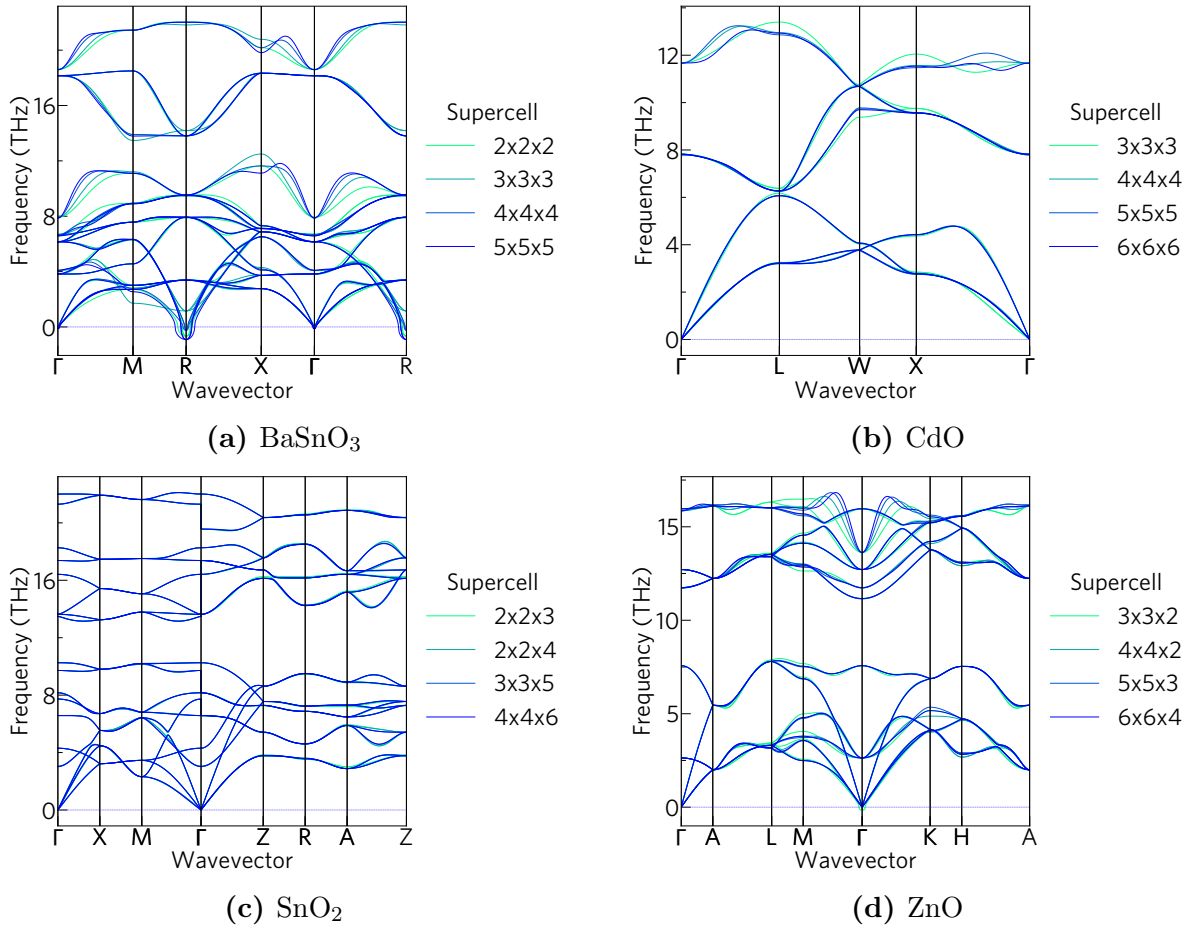


Figure S2: Plots of Seebeck coefficient, electrical conductivity, and electronic thermal conductivity, against temperature at various n-type carrier concentrations for (a) $BaSnO_3$, (b) CdO, (c) SnO_2 and (d) ZnO. The values have been averaged across crystal orientation for SnO_2 and ZnO as real devices would need to be nanostructured.

Phonon Dispersions



(a) $BaSnO_3$ **(b)** CdO

(c) SnO_2 **(d)** ZnO

Figure S3: The convergence of the phonon band structure with supercell size, calculated with Phonopy,[6] including the non-analytical correction.[7]

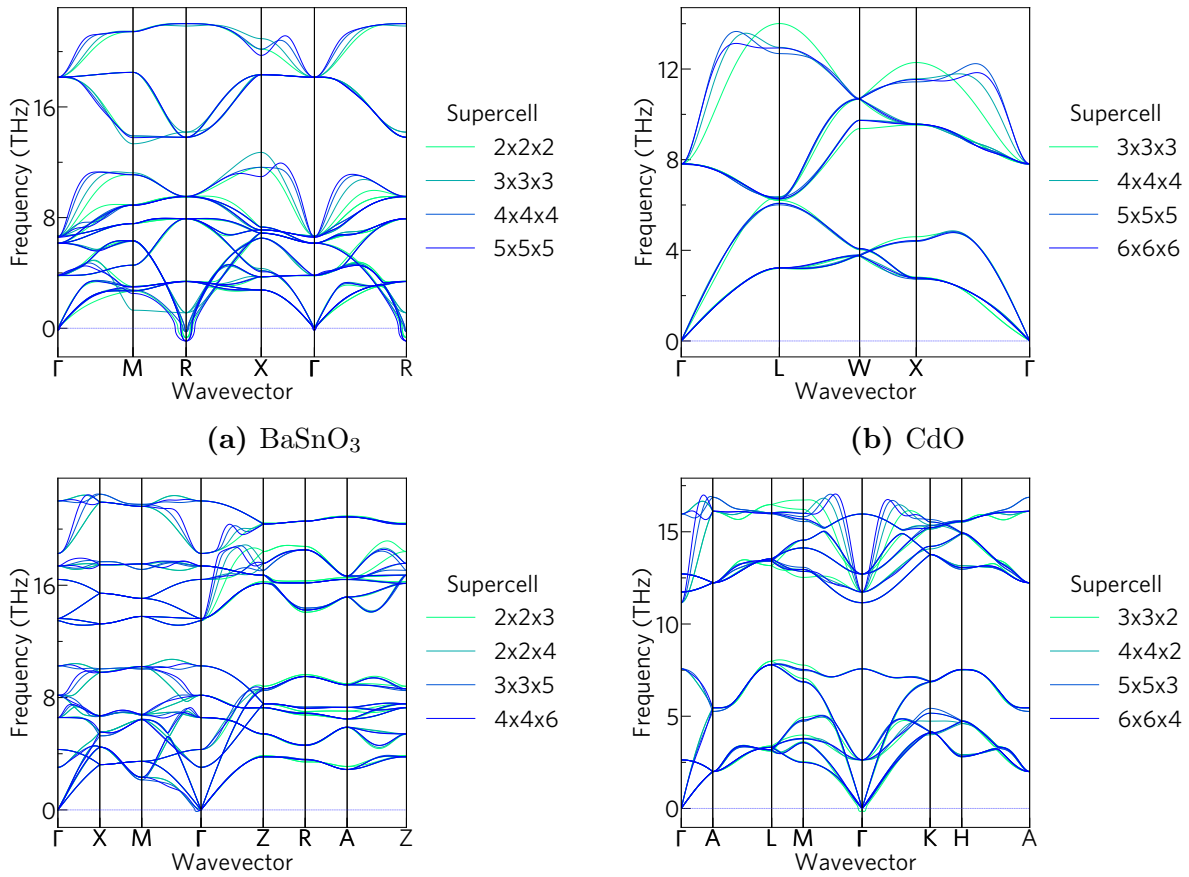
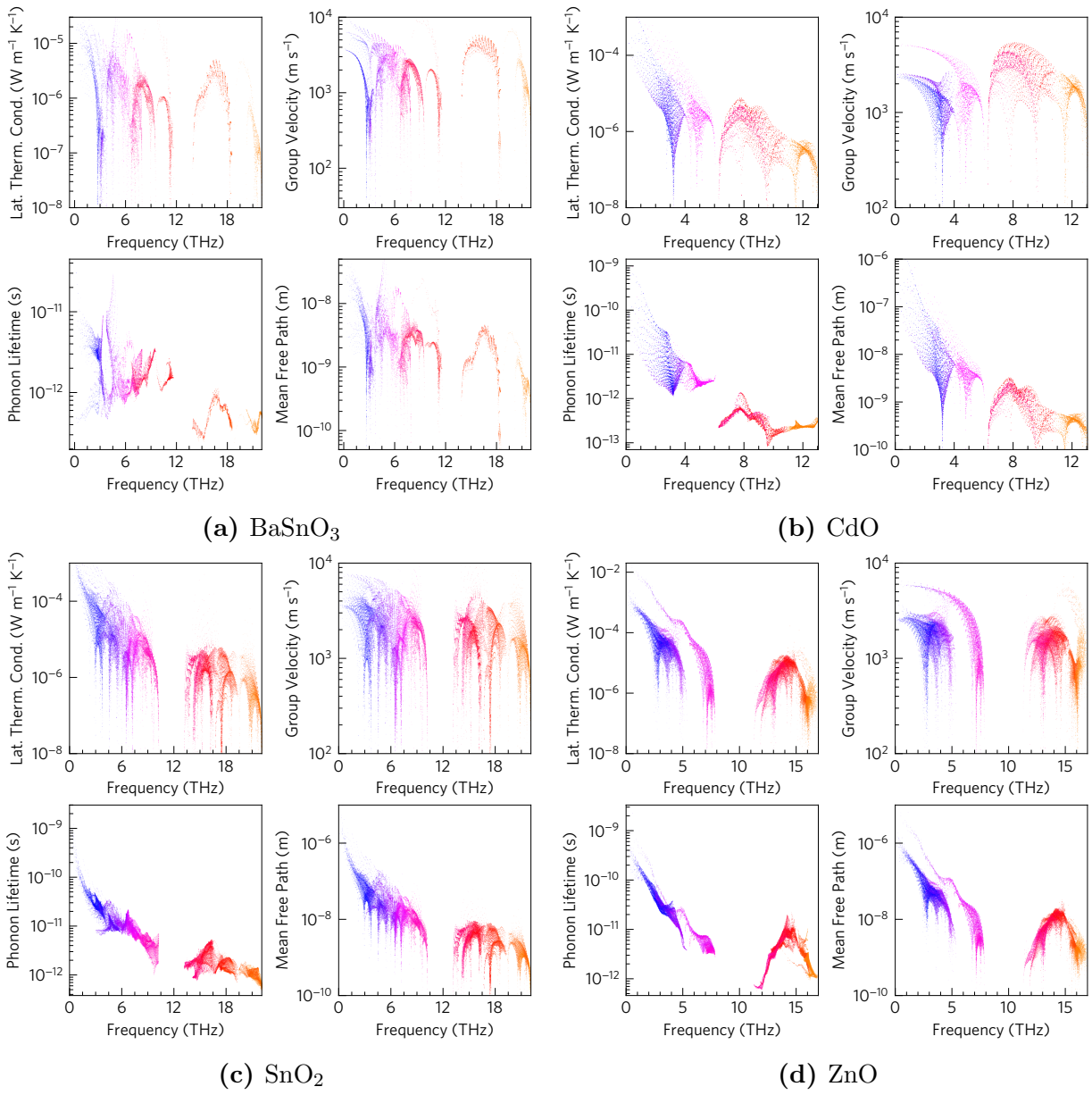


Figure S4: The convergence of the phonon band structure with supercell size, as in Figure S3, but without the non-analytical correction.[6,7]

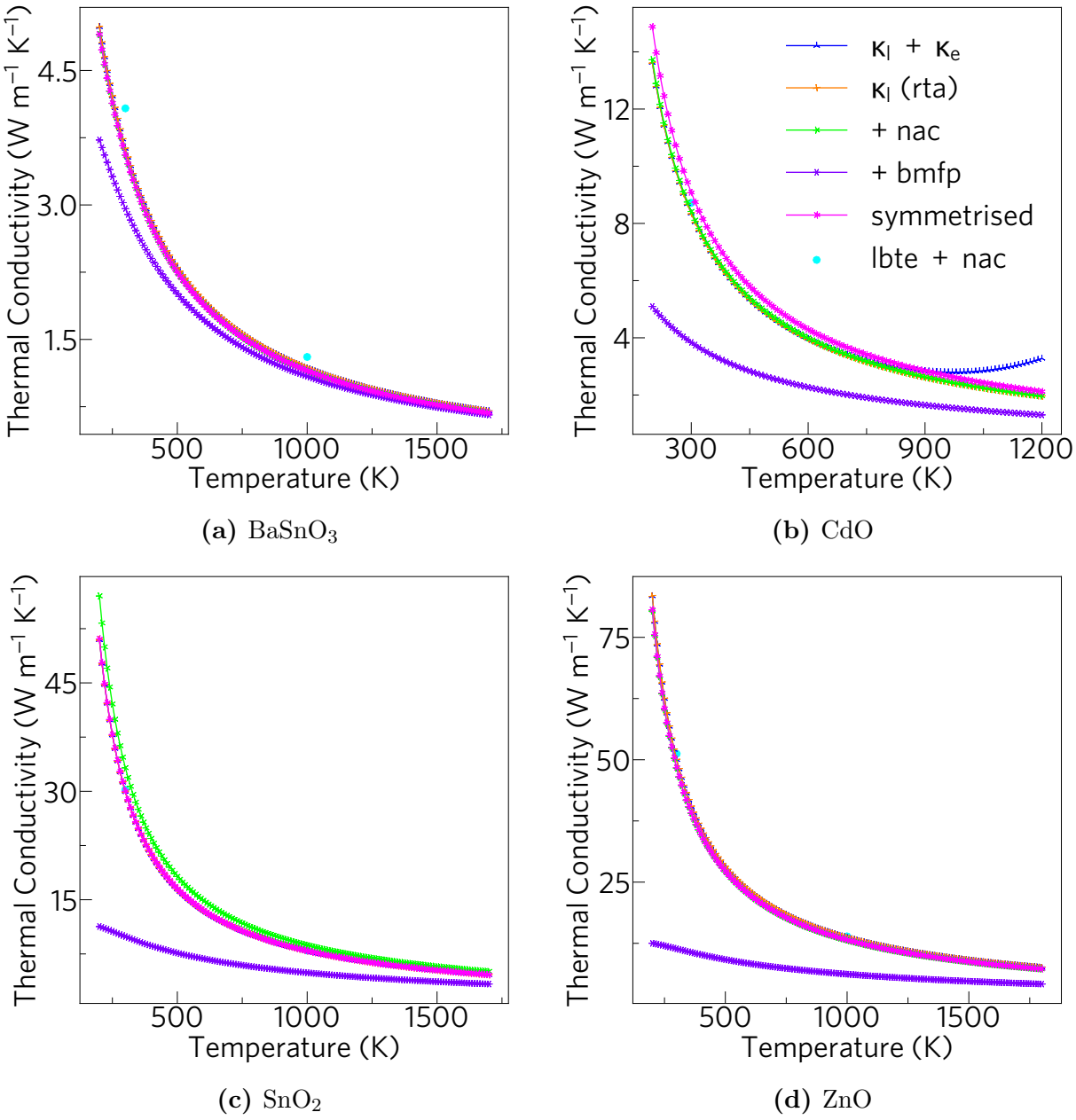
Phono3py Data



(a) $BaSnO_3$ **(b)** CdO

(c) SnO_2 **(d)** ZnO

Figure S5: Clockwise from top left per subfigure, the lattice thermal conductivity, group velocity, mean free path and lifetime for each mode at each q-point, coloured by band, for (a) $BaSnO_3$, (b) CdO, (c) SnO_2 and (d) ZnO. Data calculated with Phono3py.[8]



(a) $BaSnO_3$ **(b)** CdO

(c) SnO_2 **(d)** ZnO

Figure S6: Comparison of thermal conductivities of (a) $BaSnO_3$, (b) CdO, (c) SnO_2 and (d) ZnO via different methods. The overall thermal conductivities are shown (blue), followed by the lattice thermal conductivities calculated with the relaxation time approximation (RTA, orange), adding the non-analytic correction (NAC, lime), limiting the phonon mean free path to 20nm using Phono3py's boundary mean free path (BMFP) method (purple), symmetrising the force constants (pink) and fully solving the linearised Boltzmann transport equations (LBTE) without the RTA (cyan). All lattice thermal conductivities calculated with Phono3py,[8] and electronic contributions with BoltzTraP.[9]

$BaSnO_3$ Antisite Disorder

Table S2: Energies, E, formation energies relative to the pure sample, E_f, defect concentrations at 300, 1000 and 1800 K, f, and the mass variance parameter of $\mathrm{Ba_{Sn}}$ and $\mathrm{Sn_{Ba}}$ at 1800 K, g, for $3 \times 3 \times 3$ supercells of $BaSnO_3$ in the pure form, and with antisite defects. In each case a Sn has been put on the Ba site at $\left(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}\right)$; near refers to where the Ba is put on the Sn site at $\left(\frac{2}{3}, \frac{2}{3}, \frac{2}{3}\right)$, in which case the Sn relaxed $(0.473, 0.473, 0.473)$ and the Ba to $(0.647, 0.647, 0.647)$. Far refers to where the Ba was placed on the Sn site at $(0, 0, 0)$, in which case no lattice distortion occured. The defect concentrations are calculated as:

$$f = \frac{N}{N_0} = e^{-\frac{E_\mathrm{f}}{k_\mathrm{B} T}}$$

where k_B is the Boltztmann constant and T is the thermodynamic temperature. The mass variance parameter is calculated via the method used in Phono3py, based on the method of Shin-ichiro Tamura:[10]

$$g = \sum_i f_i \left(1 - \frac{m_i}{\overline{m}}\right)^2$$

where m is the mass of the atom i and $\overline{m}$ is the average mass on the site. For a pure sample this would result in a mass variance parameter of 0, and even at 1800 K the value of those calculated here is only negligbly higher than this. The formation energies are consistent with previous calculations[11,12] and these results indicate that antisite defects will not be prevalent in $BaSnO_3$

System	E (eV)	E_f (eV)	f (300 K)	f (1000 K)	f (1800 K)	$g_{\mathrm{Ba_{Sn}}}$	$g_{\mathrm{Sn_{Ba}}}$
Pure	-900	0	1	1	1	0	0
Near	-891	4.45	1.70×10^{-75}	3.71×10^{-23}	3.46×10^{-13}	8.51×10^{-15}	6.36×10^{-15}
Far	-890	4.89	7.71×10^{-83}	2.32×10^{-25}	2.06×10^{-14}	5.07×10^{-16}	3.79×10^{-16}

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