Supporting Information

Discordant Nature of Cd in GeTe Enhances Phonon Scattering and Improves Band Convergence for High Thermoelectric Performance

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Figure S1. Heat flow from DSC measurements showing the phase transition temperature of Ge$_{1-x}$Cd$_x$Te ($x = 0-0.05$).
Scattering parameter calculations: Callaway suggests that the impurity scattering parameter ($\Gamma$) can be calculated by fitting the lattice thermal conductivity of a disordered compound ($\kappa_L$) and the lattice thermal conductivity of an ordered pure GeTe ($\kappa^p_L$) compound through Eq. 1:

$$\frac{\kappa_L}{\kappa^p_L} = \tan^{-1} u, \quad u^2 = \frac{\pi^2 \Theta_D \Omega}{v^2} \kappa^p_L \Gamma,$$

(1)

where $u$ is the disorder scaling parameter, $\Theta_D$ is the Debye temperature (244 K for pure GeTe), $\Omega$ is the average atomic volume, $\hbar$ is the Planck’s constant, and $v$ is the average sound velocity (2452 m/s for pure GeTe). In the model of Slack$^1$ and Abeles$^2$, considering the disorder scattering parameter as a combined value of disorder from both mass and strain field fluctuations allows to express $\Gamma$ as: $\Gamma = \Gamma_M + \Gamma_S$, where $\Gamma_M$ and $\Gamma_S$ are the mass and strain fluctuation parameters, respectively, given by...
Here $n$ is the number of different atoms in the lattice ($n = 2$ in GeTe) and $c_i$ is the degeneracy of the atomic occupancy ($c_1 = c_2 = 1$), $\bar{M}_i$ and $\bar{r}_i$ are the average atomic mass and radius on the $i^{th}$ sublattice, respectively, $\bar{M}$ is the average relative atomic mass of the compound, $f_i^k$ is the fractional occupation of the $k^{th}$ atoms on the $i^{th}$ sublattice, $\varepsilon_1$ is the phenomenological adjustable parameter, $M_i^k$ and $r_i^k$ are the atomic mass and radius, respectively, expressed as:

\begin{align}
M_i^k &= \sum_k f_i^k M_i^k \\
\bar{M} &= \frac{\sum_{i=1}^n c_i \bar{M}_i}{\sum_{i=1}^n c_i} \\
r_i^k &= \sum_k f_i^k r_i^k \\
M_i^k &= \sum_{k} f_i^k M_i^k \\
\bar{r}_i &= \frac{\sum_{i=1}^n c_i \bar{r}_i}{\sum_{i=1}^n c_i}
\end{align}
Following the above expressions, a simplified expression for the impurity scattering parameter \( \Gamma \) is derived and can be written as:

\[
\Gamma = \frac{1}{4} \left( \frac{\overline{M}}{M} \right)^2 \chi (1 - x) \left[ \left( \frac{M_1 - M_2}{\overline{M}} \right)^2 + \epsilon_1 \left( \frac{r_1 - r_2}{\overline{r}_1} \right)^2 \right].
\]

(7)

Figure S3. Temperature dependent thermoelectric transport properties for Ge\(_{1-y}\)Sb\(_{y}\)Te (\( y = 0\)–0.10): (a) electrical conductivity, (b) Seebeck coefficient, (c) power factor, (d) total thermal conductivity, (e) lattice thermal conductivity and (f) figure of merit, ZT.

**Electronic thermal conductivity and Lorenz number of Ge\(_{0.97-y}\)Cd\(_{0.03}\)Sb\(_{y}\)Te:** The electronic thermal conductivity is calculated from the Wiedemann-Franz law, \( \kappa_e = L \sigma T \), where \( L \) is the Lorenz number. \( L \) was calculated using the chemical potential, estimated by fitting the
experimental Seebeck coefficient. $\sigma$ is the measured electrical conductivity and $T$ is the absolute temperature.

Figure S4. (a) Temperature-dependent electronic thermal conductivity of the Ge$_{0.97-y}$Cd$_{0.03}$Sb$_y$Te ($y = 0$-0.10) sample and (b) Lorenz number as a function of temperature of Ge$_{0.97-y}$Cd$_{0.03}$Sb$_y$Te ($y = 0$-0.10).

References