Supporting Information

Simultaneously boosting electrical and thermal transport properties of $CuGaTe_2$ through XCl_2 (X = Cd, Zn) doping-driven band and defect engineering

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Calculation details

$Cu_{16}Ga_{16}Te_{32}+ZnCl_2= Cu_{16}Ga_{15}ZnTe_{32}+Ga+Cl+Cl$

$Cu_{16}Ga_{16}Te_{32}+ZnCl_2= Cu_{15}ZnGa_{16}Te_{31}Cl+Cl+Cu+Te$

$Cu_{16}Ga_{16}Te_{32}+ZnCl_2= Cu_{16}Ga_{15}ZnTe_{31}Cl+Cl+Ga+Te$

$Cu_{16}Ga_{16}Te_{32}+CdCl_2= Cu_{16}Ga_{15}CdTe_{32}+Ga+Cl+Cl$

$Cu_{16}Ga_{16}Te_{32}+CdCl_2= Cu_{15}CdGa_{16}Te_{31}Cl+Cl+Cu+Te$

$Cu_{16}Ga_{16}Te_{32}+CdCl_2= Cu_{16}Ga_{15}CdTe_{31}Cl+Cl+Ga+Te$

$E_{form1}=E_{total}(Cu_{16}Ga_{15}ZnTe_{32}) +E_{total}(Ga)+ E_{total}(Cl)+E_{total}(Cl)-E_{total}(Cu_{16}Ga_{16}Te_{32})-E_{total}(ZnCl_2) =-2.1$ eV

$E_{form1}= E_{total}(Cu_{15}ZnGa_{16}Te_{31}Cl) +E_{total}(Cl)+E_{total}(Cu)+E_{total}(Te)-E_{total}(Cu_{16}Ga_{16}Te_{32})-E_{total}(ZnCl_2) =-0.9$ eV

$E_{form3}= E_{total}(Cu_{16}Ga_{15}ZnTe_{31}Cl) +E_{total}(Cl)+E_{total}(Ga)+E_{total}(Te)-E_{total}(Cu_{16}Ga_{16}Te_{32})-E_{total}(ZnCl_2) =-1.5$ eV

$E_{form4}= E_{total}(Cu_{16}Ga_{15}CdTe_{32}) +E_{total}(Ga)+E_{total}(Cl)+E_{total}(Cl)-E_{total}(Cu_{16}Ga_{16}Te_{32})-E_{total}(CdCl_2) =-1.7$ eV

$E_{form5}= E_{total}(Cu_{15}ZnGa_{16}Te_{31}Cl) +E_{total}(Cl)+E_{total}(Cu)+E_{total}(Te)-E_{total}(Cu_{16}Ga_{16}Te_{32})-E_{total}(CdCl_2) =-0.6$ eV

$E_{form6}= E_{total}(Cu_{15}ZnGa_{16}Te_{31}Cl) +E_{total}(Cl)+E_{total}(Ga)+E_{total}(Te)-E_{total}(Cu_{16}Ga_{16}Te_{32})-E_{total}(CdCl_2) =-1.2$ eV

E_{form} represents formation energy and E_{total} stands for the total energy of each material.

$E_{form1} < E_{form3} < E_{form2}$

$E_{form4} < E_{form6} < E_{form5}$

Single parabolic band model: for the calculation the experimental value of:

$$S(\eta) = \frac{k_B}{e} \cdot \left[\frac{\left(r+\frac{5}{2}\right) \cdot F_{r+\frac{3}{2}}(\eta)}{\left(r+\frac{3}{2}\right) \cdot F_{r+\frac{1}{2}}(\eta)} - \eta \right]$$

$$n = \frac{1}{e \cdot R_H} = \frac{\left(2m^* \cdot k_B T\right)^{\frac{3}{2}}}{3\pi^2\hbar^3} \cdot \left| \frac{\left(r+\frac{3}{2}\right)^2 \cdot F^2_{r+\frac{1}{2}}(\eta)}{\left(2r+\frac{3}{2}\right) \cdot F_{2r+\frac{1}{2}}(\eta)} - \eta \right|$$

$$Fi(\eta) = \int_0^{\infty} \frac{x^i}{1+e^{x-\eta}} dx$$

In the equation, η represents the reduced Fermi level, k_B is the Boltzmann constant, e stands for the electron charge, r is the carrier scattering factor (r = -1/2 for acoustic phonon scattering), R_H represents the Hall coefficient, and ħ represents the reduced Planck constant.

Supporting Table

Table S1. Transverse (v_t), longitudinal (v_l), mean sound velocity (v_s) (m/s) of $Cu_{16}Ga_{16}Te_{32}$, $Cu_{16}Ga_{15}CdTe_{32}$ and $Cu_{16}Ga_{15}ZnTe_{32}$.

	v_t(m/s)	v_l(m/s)	v_s(m/s)
$Cu_{16}Ga_{16}Te_{32}$	1975	3795	2210
$Cu_{16}Ga_{15}CdTe_{32}$	1864	3728	2086
$Cu_{16}Ga_{15}ZnTe_{32}$	1900	3602	2050

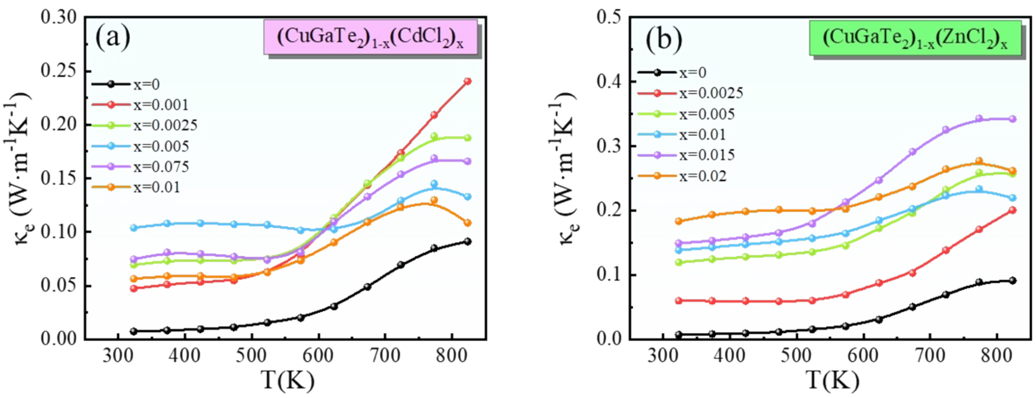

Figure S1. Temperature dependence of (a) Electronic thermal conductivity of $(CuGaTe_2)_{1-x}(CdCl_2)_x$ (x = 0, 0.001, 0.0025, 0.005, 0.0075, 0.01), (b) Electronic thermal conductivity of $(CuGaTe_2)_{1-x}(ZnCl_2)_x$ (x = 0, 0.0025, 0.005, 0.01, 0.015, 0.02)