Supporting Information

Enhancing Near-Room-Temperature GeTe Thermoelectrics through In/Pb Co-doping

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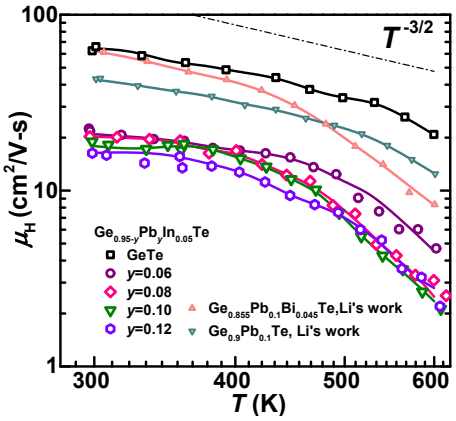

Figure S1 Temperature dependent hall mobility for $Ge_{0.95-y}Pb_yIn_{0.05}Te$ ($0 \le y \le 0.12$) samples in this work with comparison to literature work, indicating the predominant phonon scattering.

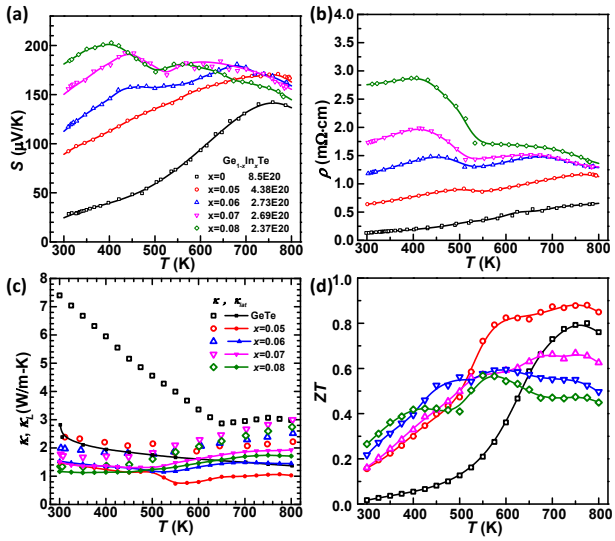

Figure S2 Temperature dependent Seebeck coefficient (a), resistivity (b), thermal conductivity and its lattice component (c) and zT value (d) for $Ge_{1-x}In_xTe$ ($0 \le x \le 0.08$) samples in this work.

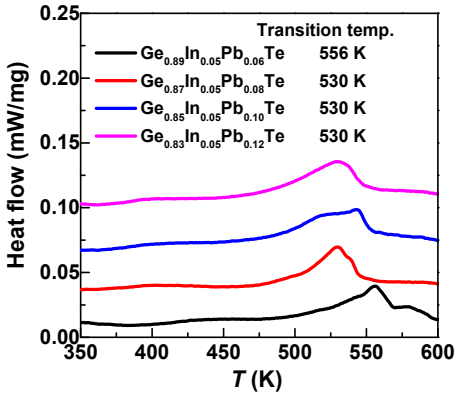

Figure S3 Heat flow as a function of temperature for $Ge_{0.95-y}In_{0.05}Pb_yIn_{0.05}Te$ ($0.06 \le y \le 0.012$) samples, indicating a phase transition above 500 K.

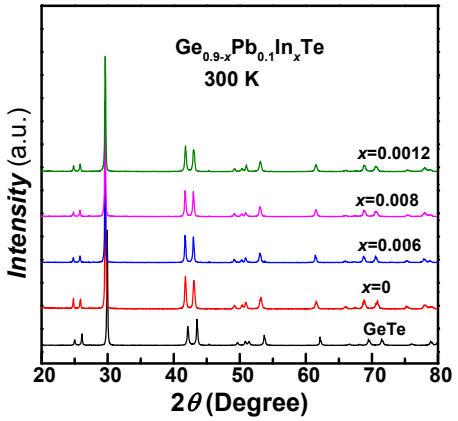

Figure S4 Room temperature powder X-ray diffraction (XRD) patterns for $Ge_{0.9-x}Pb_{0.1}In_xTe$ ($0 \le x \le 0.012$) samples.

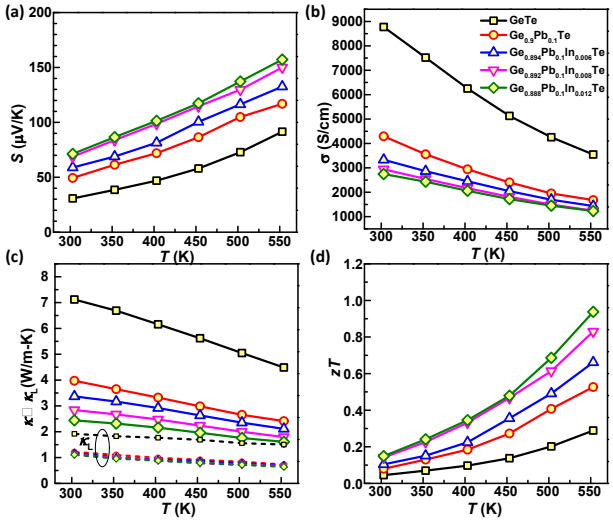

Figure S5 Temperature dependent Seebeck coefficient (a), resistivity (b), thermal conductivity and its lattice component (c) and zT value (d) for $Pb_{0.1}Ge_{0.9-x}In_xTe$ ($0 \le x \le 0.012$) samples in this work.

Debye-Callaway model:

For a material system with point defect and Umklapp scattering only, the relationship between the lattice thermal conductivity for pure ($\kappa_{L,pure}$) and alloy ($\kappa_{L,alloy}$) materials can be simplified as follows[1-4]:

$$\frac{\kappa_{L,alloy}}{\kappa_{L,pure}} = \frac{\arctan(u)}{u}, u^2 = \frac{\pi^2 \theta_D \Omega}{h v^2} \kappa_{L,pure} \Gamma_{exp}$$

Where, Ω is the volume per atom, h is the Plank constant, and θ_D=195 K is the Debye temperature and v=1967 m/s is the speed of sound for GeTe, Γ_{exp} is the disorder parameter including the mass (Mi/M) and strain ($\Delta\delta_i/\delta$) components via:

$$\Gamma_{exp} = \sum_i x_i \left[\left(\Delta M_i / M \right)^2 + \varepsilon \left(\Delta \delta_i / \delta \right)^2 \right]$$

$$\Delta M_i / M = \frac{M_i - M}{M}, M = \sum_i x_i M_i$$

$$\Delta \delta_i / \delta = \left(\frac{\delta_i - \delta}{\delta} \right), \delta = \sum_i x_i \delta_i$$

In the above equations, x_i, M_i and δ_i are the concentration, mass, and ionic radius of atoms of type i. ε=2(W+6.4γ)^2 is determined by the Gruneisen parameter γ=1.45 and W=4[5], the ratio between the relative change of bulk modulus and the strain, estimated for IV-VI semiconductors.

Reference:

(1) Abeles, B. Lattice Thermal Conductivity of Disordered Semiconductor Alloys at High Temperatures. *Phys Rev* **1963,** *131* (5), 1906-1911, DOI: 10.1103/PhysRev.131.1906.

(2) Callaway, J.; Vonbaeyer, H. C. Effect of Point Imperfections on Lattice Thermal Conductivity. *Phys Rev* **1960,** *120* (4), 1149-1154.

(3) Klemens, P. G. The Scattering of Low-Frequency Lattice Waves by Static Imperfections. *Proceedings of the Physical Society* **1955,** *A68* (12), 1113-1128.

(4) Klemens, P. G. Thermal Resistance due to Point Defects at High Temperatures. *Phys Rev* **1960,** *119* (2), 507-509, DOI: 10.1103/PhysRev.119.507.

(5) Wang, H.; Wang, J.; Cao, X.; Snyder, G. J. Thermoelectric alloys between PbSe and PbS with effective thermal conductivity reduction and high figure of merit. *Journal of Materials Chemistry A* **2014,** *2* (9), 3169, DOI: 10.1039/c3ta14929c.