

## Supporting Information

# Effective Enhancement of Thermoelectric and Mechanical Properties of Germanium Telluride via Rhenium-doping

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## Hall Concentration and Mobility

**Table S1.** Hall concentration and Hall mobility of all samples.

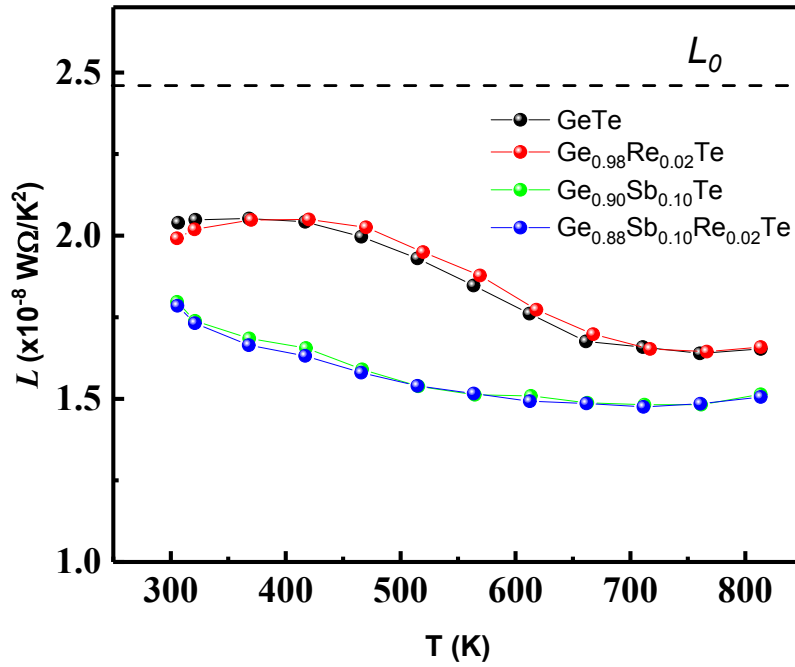
Composition	$n_H$ (cm <sup>-3</sup> )	$\mu_H$ (cm <sup>2</sup> /Vs)
GeTe	$7.5 \times 10^{20}$	64.9
Ge <sub>0.98</sub> Re <sub>0.02</sub> Te	$8.1 \times 10^{20}$	56.6
Ge <sub>0.90</sub> Sb <sub>0.10</sub> Te	$2.7 \times 10^{20}$	31.5
Ge <sub>0.88</sub> Sb <sub>0.10</sub> Re <sub>0.02</sub> Te	$2.6 \times 10^{20}$	29.6

**Table S2.** Physical properties used to model  $\kappa_L$  in GeTe based on various phonon scattering processes.

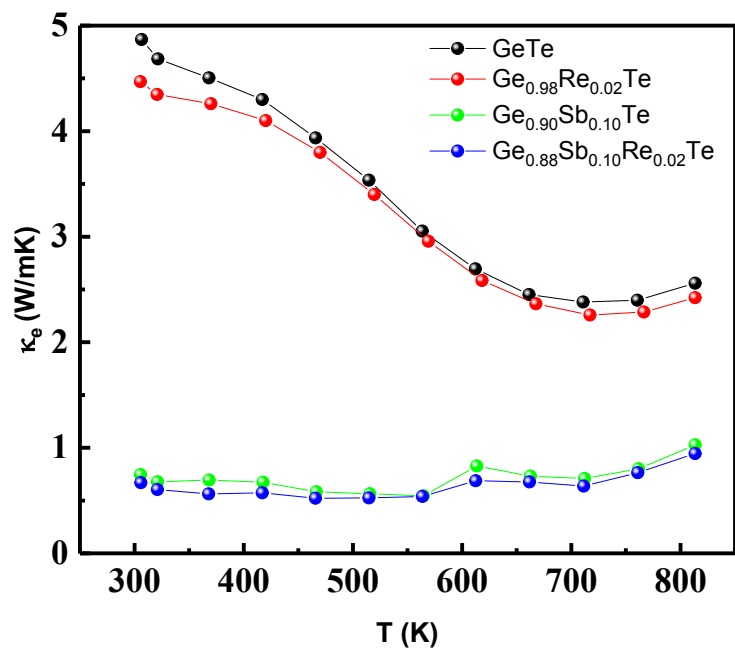
Parameters	Values
v <sub>L</sub> , m/s	3410
v <sub>T</sub> , m/s	1995
v <sub>m</sub> , m/s	2210
Atomic mass, kg	$1.66 \times 10^{25}$
Sample density, g/cm <sup>3</sup>	6.14
Debye T, K	122
$\gamma$	1.45
Poisson's ratio	0.24
Bulk modulus, GPa	39.9
Young's Modulus, GPa	62.2
Shear Modulus, GPa	25.5
Grain size, $\mu\text{m}$	10

**Table S3.** Elemental composition of GeTe with various doping levels of Sb and Re obtained from XRF spectra.

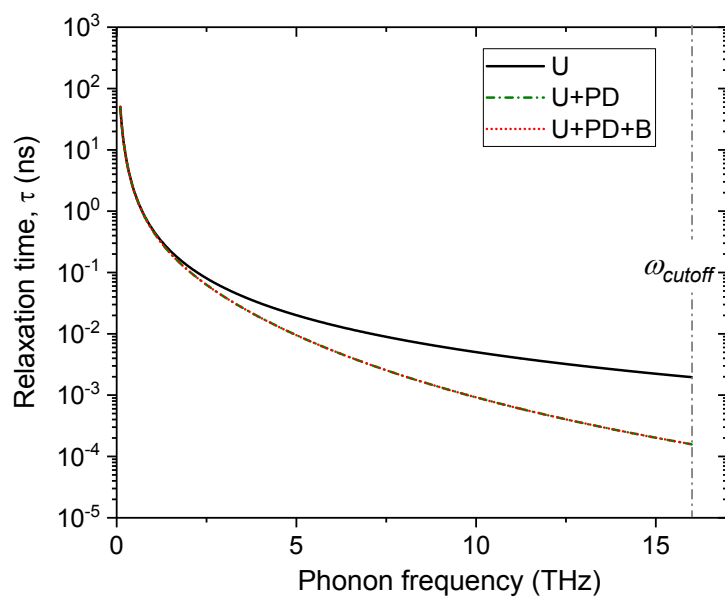
Compound	Ge (at%)	Re (at%)	Sb (at%)	Te (at%)
GeTe	46.6	-	-	53.4
Ge <sub>0.98</sub> Re <sub>0.02</sub> Te	46.7	0.9	-	52.4
Ge <sub>0.90</sub> Sb <sub>0.10</sub> Te	41.9	-	4.6	53.5
Ge <sub>0.88</sub> Sb <sub>0.10</sub> Re <sub>0.02</sub> Te	41.7	0.8	4.3	53.2



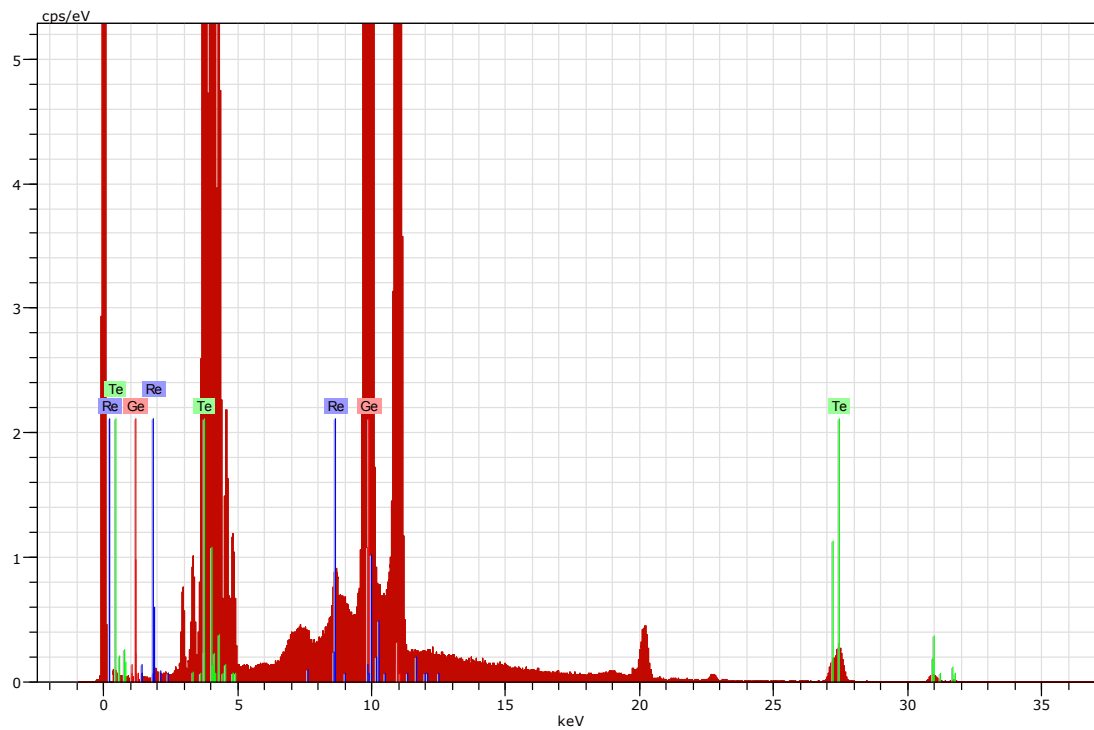
**Figure S1.** The Lorenz number from the Single Parabolic Band for all samples



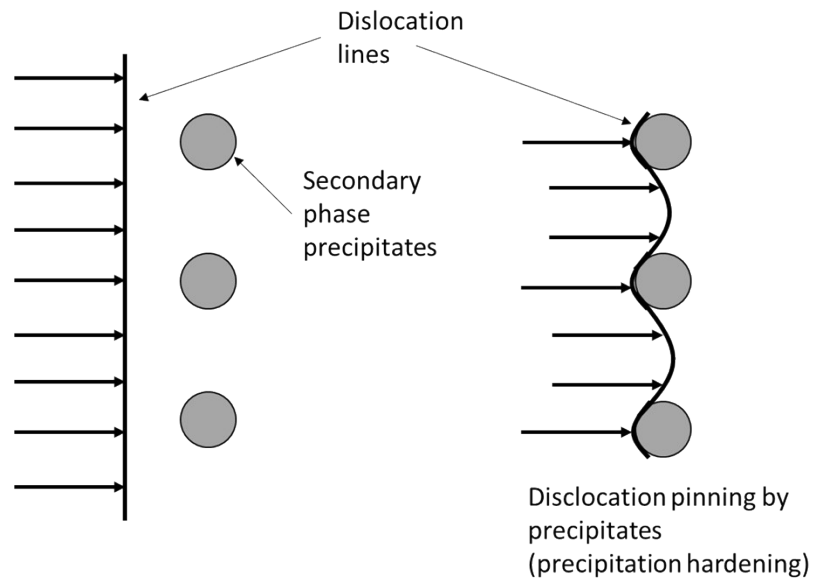
**Figure S2.** Electronic thermal conductivity for all samples



**Figure S3.** Phonon relaxation time as a function of frequency for various scattering processes.



**Figure S4.** X-ray fluorescence spectra for  $\text{Ge}_{0.98}\text{Re}_{0.02}\text{Te}$



**Figure S5.** Illustration of precipitation hardening mechanisms, resulting in hardness enhancements.<sup>1</sup>

## Jonker analysis

In order to obtain Jonker's plot in Figure 2a, the reduced Fermi level,  $\eta$  was extracted from Seebeck value:

$$S = \pm \frac{k_B}{e} \left[ \eta - \frac{\left(r + \frac{5}{2}\right) F_{r+1.5}(\eta)}{\left(r + \frac{3}{2}\right) F_{r+0.5}(\eta)} \right] \quad (1)$$

For every Seebeck value,  $\eta$  corresponding to acoustic phonon scattering ( $r = -0.5$ ) and ionized impurity scattering ( $r = 1.5$ ) can be calculated. Subsequently, the corresponding  $\eta$  values can be used to obtain electrical conductivities  $\sigma$  for particular transport coefficient value,  $\sigma_{E_0}$ :

$$\sigma = \sigma_{E_0} \ln(1 + e^\eta) \quad (2)$$

The results were then plotted as shown in Figure 2a.  $\sigma_{E_0}$  can then be estimated by looking at the position and trend of the experimental data from each sample.

## Quality factor $B$ , Transport Coefficients $\sigma_{E_0}$ , and Weighted Mobility $\mu_w$

To understand the nature of transport in more detail, we compute the  $\sigma_{E_0}$  from electrical conductivity  $\sigma$ , which can be expressed as:

$$\sigma = \sigma_{E_0} \ln(1 + e^\eta) \quad (3)$$

Essentially,  $\sigma_{E_0}$  is a convenient expression of electrical conductivity that is independent of carrier concentration. This is especially useful in our case since the carrier concentration values obtained via Hall measurements may not be accurate due to the non-linearity of the Hall voltage versus magnetic field. (i.e. the Hall carrier concentration is typically taken as the linear slope of Hall voltage versus magnetic field, non-linearity in Hall voltage versus magnetic field makes data interpretation inaccurate). Large  $\sigma_{E_0}$  can be associated with good crystalline quality and vice versa. Furthermore, the carrier mobility-equivalent for  $\sigma_{E_0}$  can be expressed as weighted-mobility  $\mu_w$ . The relation between  $\sigma_{E_0}$  and  $\mu_w$  can be expressed as:

$$\sigma_{E_0} = \frac{e(2m_e k_B T)^{3/2}}{3\pi^2 \hbar^3} \mu_W \quad (4)$$

$$\mu_W = \mu_0 \left( \frac{m_{DOS}^*}{m_e} \right)^{3/2} \quad (5)$$

The main advantage of using weighted-mobility over inaccurately determined Hall mobility lies in the fact that weighted-mobility takes into account the  $m_{DOS}^*$  (density of states effective mass). Since the density of states effective mass provides a direct correlation to the Seebeck coefficient, the inverse correlation between electrical conductivity and Seebeck coefficient can be clearly accounted for by looking at the weighted mobility. Hence, it can be used as a robust indication of the thermoelectric power factor. It is important to note that while weighted mobility share some similarities with Hall mobility, their magnitude generally differs, especially for compounds with high band-degeneracy (high  $m_{DOS}^*$ ). This comes from the fact that weighted mobility has a  $m_{DOS}^{*3/2}$  dependence whereas Hall mobility only depends on  $\mu_0$  (intrinsic mobility) as well as the reduced Fermi level and scattering mechanism.

Lastly, the quality factor B can be evaluated from  $\sigma_{E_0}$  based on the following:

$$B = \left( \frac{k_B}{e} \right)^2 \frac{T}{k_L} \sigma_{E_0} \quad (6)$$

It is evident from equation 5 that in order to enhance the quality factor,  $\sigma_{E_0}$  must be enhanced, either via band convergence, resonant doping, energy filtering, or deformation potential engineering to increase  $m_{DOS}^*$ . Alternatively,  $k_L$  can be reduced via point defects, strain, dislocation, or stacking faults.

## Lorenz Number

The Lorenz number used in this work is calculated from the semiclassical Boltzmann Transport Equations under single parabolic band assumption:

$$L = \left( \frac{k_B}{e} \right)^2 \left[ \frac{\left( r + \frac{7}{2} \right) F_{r+1.5}(\eta)}{\left( r + \frac{3}{2} \right) F_{r+0.5}(\eta)} - \frac{\left( r + \frac{5}{2} \right) F_{r+1.5}(\eta)}{\left( r + \frac{3}{2} \right) F_{r+0.5}(\eta)} \right]^2 \quad (7)$$

Where  $r$  represents the carrier scattering exponent, set at -0.5 for acoustic phonon scattering.

### Simplified Debye-Callaway model for lattice thermal conductivity

In order to model the lattice thermal conductivity, Debye frequency was first determined from:

$$\omega_{max} = \omega_D = \left(\frac{6\pi^2}{V}\right)^{1/3} v_m \quad (8)$$

Where V is the atomic volume and  $v_m$  was obtained from equation (4). The acoustic branch maximum frequency can be expressed as:

$$\omega_a = \frac{\omega_D}{N^{1/3}} \quad (9)$$

Where N is the number of atoms per unit cell.

The Debye temperature  $\theta_D$  can then be expressed as:

$$\hbar\omega_D = k_B\theta_D \quad (10)$$

Subsequently, the phonon relaxation time  $\tau(\omega)$  can be calculated by accounting for contribution from Umklapp, grain boundaries, and point defects as following:

$$\tau_U^{-1} = \frac{2}{(6\pi^2)^{1/3}} \frac{k_B V^{1/3} \gamma^2 \omega^2 T}{\bar{M} v^3} \quad (11)$$

$$\tau_B^{-1} = \frac{v}{d} \quad (12)$$

$$\tau_{PD}^{-1} = \frac{V\omega^4}{4\pi v^3} \left( \sum_i f_i \left(1 - \frac{m_i}{\bar{m}}\right)^2 + \sum_i f_i \left(1 - \frac{r_i}{\bar{r}}\right)^2 \right) \quad (13)$$

In our case, the spectral heat capacity  $C_s(\omega)$  can be expressed as:

$$C_s(\omega) = \frac{3k_B\omega^2}{2\pi^2 v^3} \quad (14)$$

By assuming constant group velocity (sound velocity), we can express the spectral thermal conductivity  $\kappa_s(\omega)$  as:

$$\kappa_s(\omega) = C_s(\omega) v^2 \tau(\omega) \quad (15)$$

Finally, the lattice thermal conductivity can be obtained by integrating the spectral thermal conductivity over the entire frequency range up to  $\omega_a$ :

$$\kappa_L = \frac{1}{3} \int_0^{\omega_{max}} C_s(\omega) v^2 \tau(\omega) d\omega \quad (16)$$



In our experiments, we obtained both **elastic constant ( $E$ )** from nanoindentation as well as **longitudinal sound velocity ( $v_L$ )** from ultrasonic measurements. In order to obtain other elastic properties, we made use of the following equations:

$$B = \frac{E}{3(1 - 2\nu_p)} \quad (17)$$

where  $B$  = Bulk modulus;  $\nu_p$  = Poisson ratio

Both sides of the above equation can be expressed in terms of  $v_L$  and  $v_T$  (longitudinal and transverse sound velocity, respectively) as follows:

$$B = \rho \left( v_L^2 - \frac{4}{3}v_T^2 \right) \quad (18)$$

$$\nu_p = \frac{1 - 2\left(\frac{v_T}{v_L}\right)^2}{2 - 2\left(\frac{v_T}{v_L}\right)^2} \quad (19)$$

where  $\rho$  = density

The transverse sound velocity  $v_T$  can then be calculated by substituting equation (2) and (3) into equation (1) and solving for  $v_T$ . Subsequently, the average sound velocity,  $v_m$  can be determined via:

$$v_m = \left( \frac{1}{3} \left[ \frac{1}{v_L^3} + \frac{2}{v_T^3} \right] \right)^{-\frac{1}{3}} \quad (20)$$

Shear modulus  $\mu$  can be obtained from:

$$\mu = \rho v_T^2 \quad (21)$$

In addition, after obtaining the poisson ratio  $\nu_p$  from equation (3), the Gruneisen parameter  $\gamma$  can be determined by:

$$\gamma = \frac{3}{2} \left( \frac{1 + \nu_p}{2 - 3\nu_p} \right) \quad (22)$$

## References

1. R. R. Ambriz and D. Jaramillo, *Light Metal Alloys Applications*, 2014, 35-59.

