Supporting Information

Lattice anharmonicity in charge compensated higher manganese silicide single crystals

Nagendra Singh Chauhan*, Ichiro Ono, and Yuzuru Miyazaki

Department of Applied Physics, Graduate School of Engineering, Tohoku University, Sendai, Miyagi, Japan - 980-8579

*Corresponding author: nagendra599@gmail.com; chauhan.nagendra.singh.b1@tohoku.ac.jp
TEL/FAX: +81-22-795-7970/7971

S1. Structural Parameter and Crystallographic details - To determine the lattice parameters and phase identification, a (3+1) dimensional superspace group approach was used in conjunction with Le Bail analysis on the JANA2006 software package.

Centrosymmetric super-space group: ***I*4$_1$/*amd*(00γ)00*ss* (No. 141)**

Table S1. Refined parameters and crystallographic details from Le Bail analysis of the powder XRD data for nominal $(Mn_{1-2x}V_xFe_x)Si_{1.74}$ samples at room temperature.

Chemical Composition	**GOF**	**Cell Parameters ($MnSi_\gamma$ Phase)**			
$(Mn_{1-2x}V_xFe_x)Si_{1.74}$	R_{wp}/R_e	$a = b$ (Å)	c_{Mn} (Å)	c_{Si} (Å)	$\gamma = c_{Mn}/c_{Si}$
$x = 0.01$	1.41	5.5277(1)	4.3684(1)	2.5160(1)	1.7378(1)
$x = 0.02$	1.40	5.5236(1)	4.3681(1)	2.5345(1)	1.7234(1)
$x = 0.03$	1.32	5.5260(1)	4.3697(1)	2.5343(1)	1.7242(1)
$x = 0.04$	1.36	5.5252(1)	4.3688(1)	2.5353(1)	1.7228(1)
$x = 0.05$	1.21	5.5234(1)	4.3664(1)	2.5372(1)	1.7218(1)
$x = 0.06$	1.23	5.5221(1)	4.3652(1)	2.5363(1)	1.7209(1)

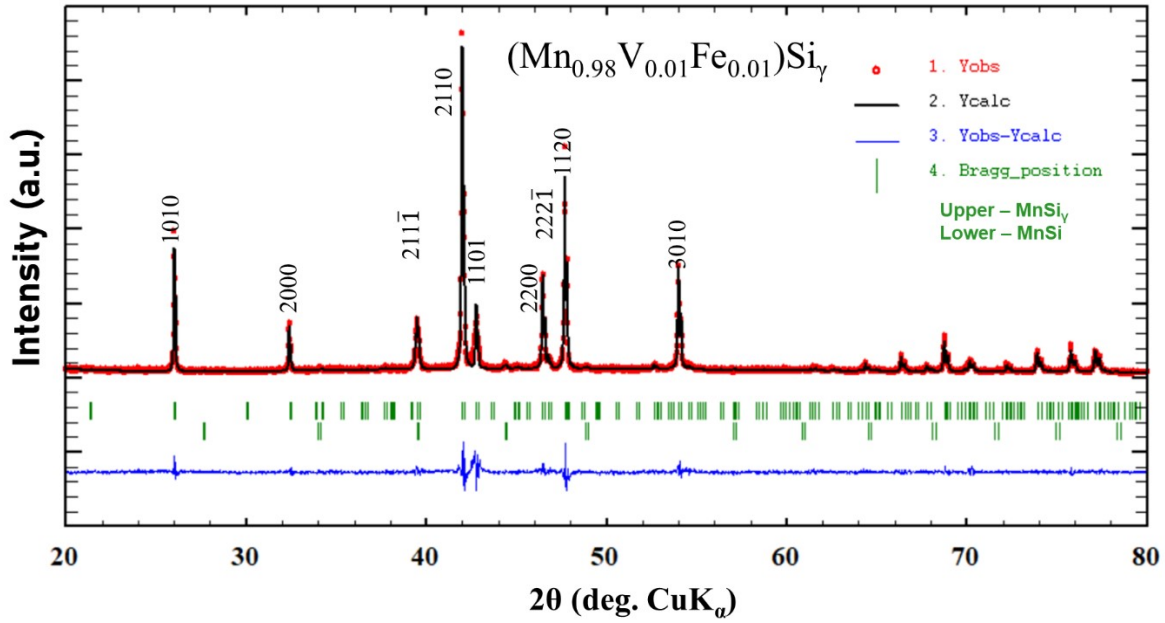

Figure S1. Le Bail analyis of XRD pattern of nominal composition $(Mn_{0.98}V_{0.01}Fe_{0.01})Si_{1.74}$ displaying Bragg positions.

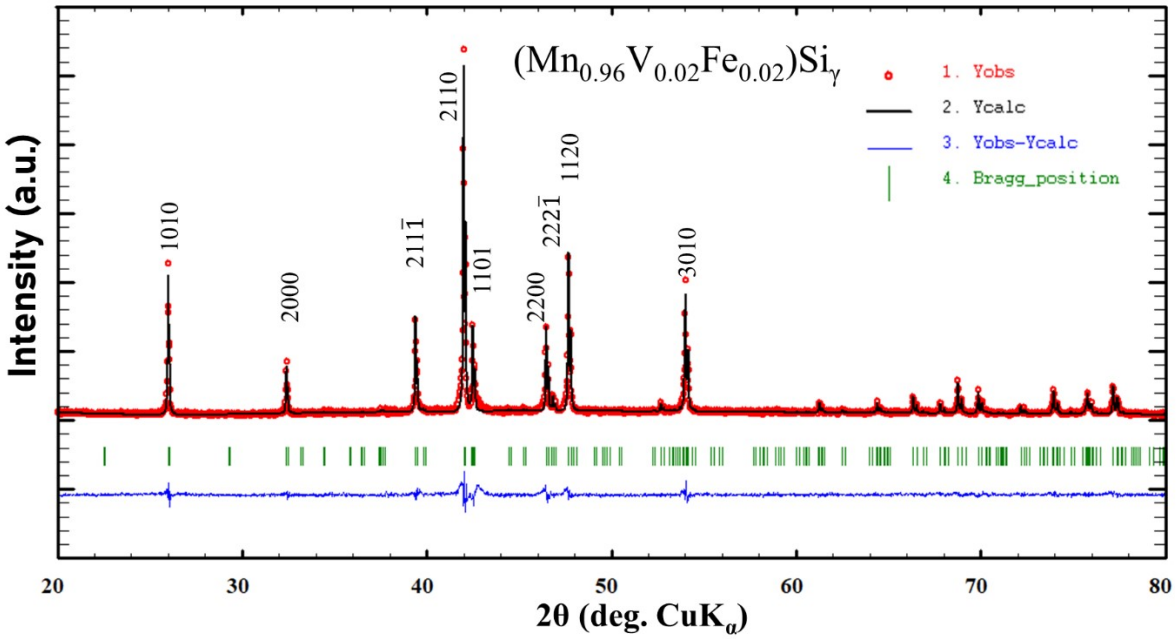

Figure S2. Le Bail analyis of XRD pattern of nominal composition $(Mn_{0.96}V_{0.02}Fe_{0.02})Si_{1.74}$ displaying Bragg positions.

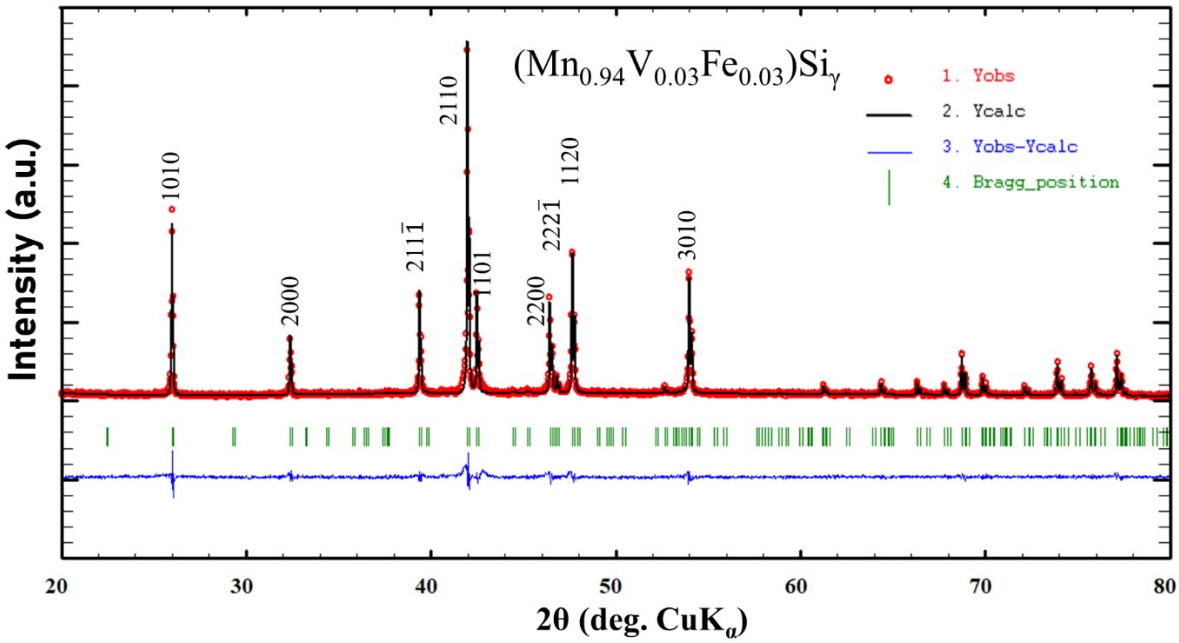

Figure S3. Le Bail analyis of XRD pattern of nominal composition $(Mn_{0.94}V_{0.03}Fe_{0.03})Si_{1.74}$ displaying Bragg positions.

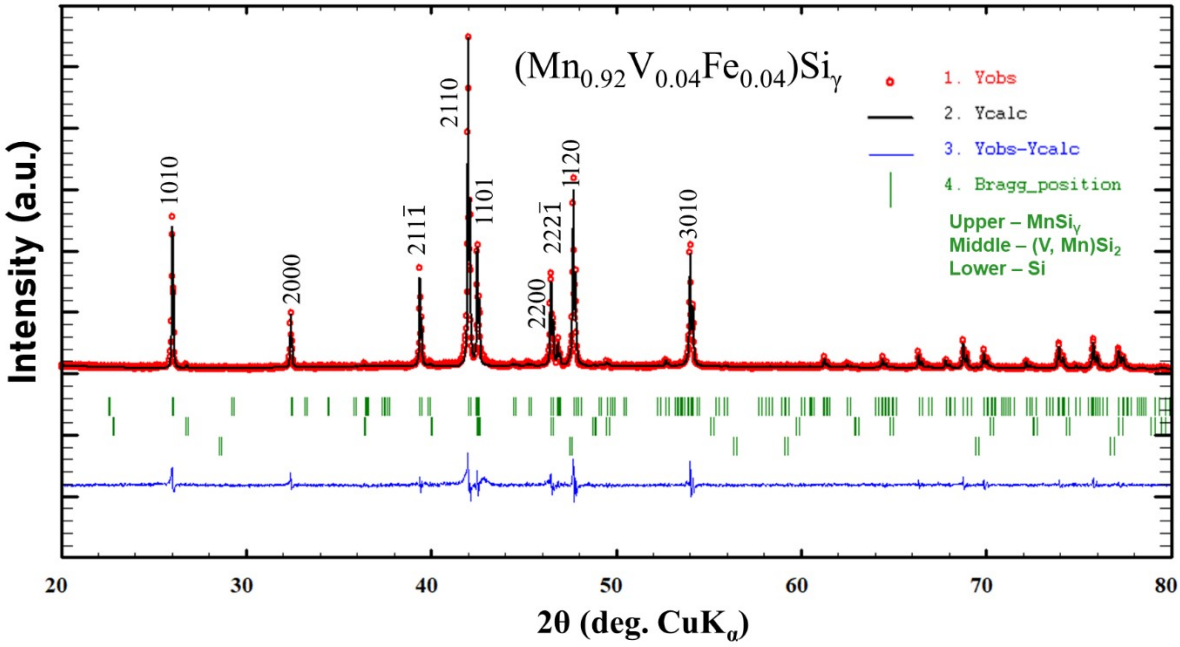

Figure S4. Le Bail analyis of XRD pattern of nominal composition $(Mn_{0.92}V_{0.04}Fe_{0.04})Si_{1.74}$ displaying Bragg positions.

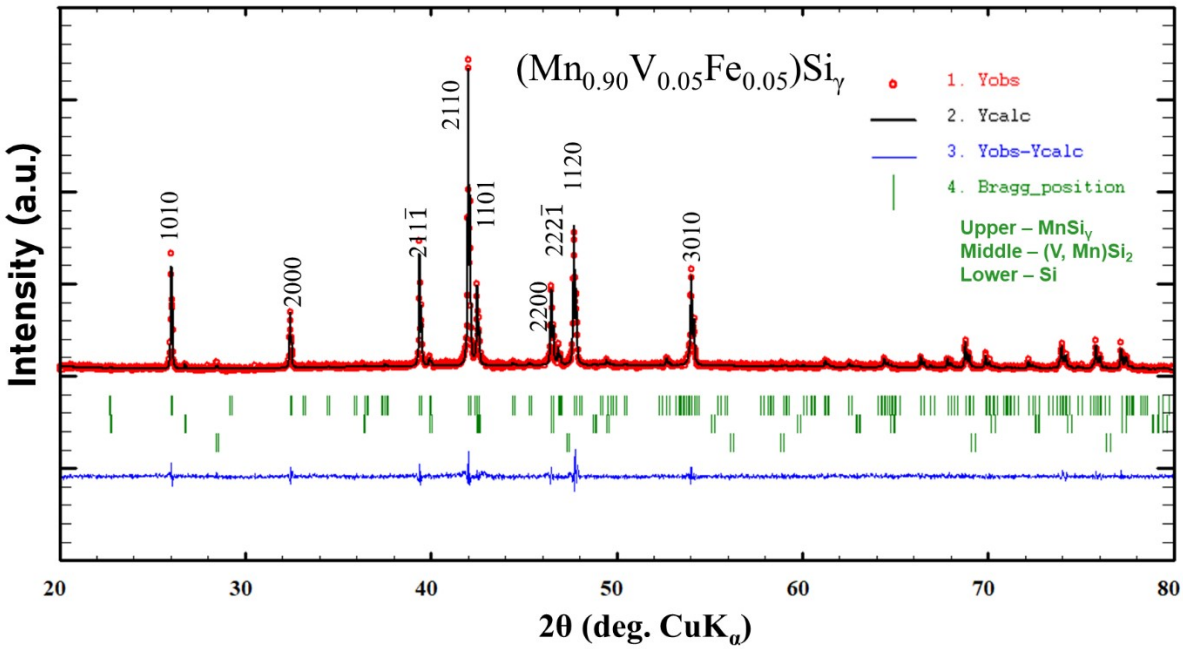

Figure S5. Le Bail analyis of XRD pattern of nominal composition $(Mn_{0.90}V_{0.05}Fe_{0.05})Si_{1.74}$ displaying Bragg positions.

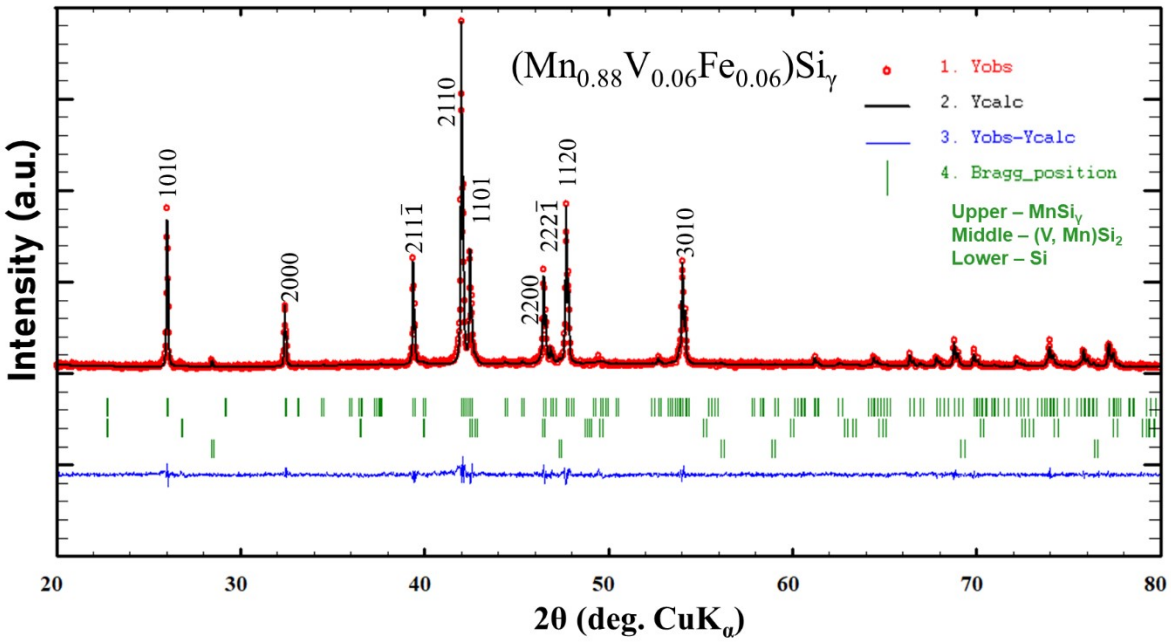

Figure S6. Le Bail analyis of XRD pattern of nominal composition $(Mn_{0.88}V_{0.06}Fe_{0.06})Si_{1.74}$ displaying Bragg positions.

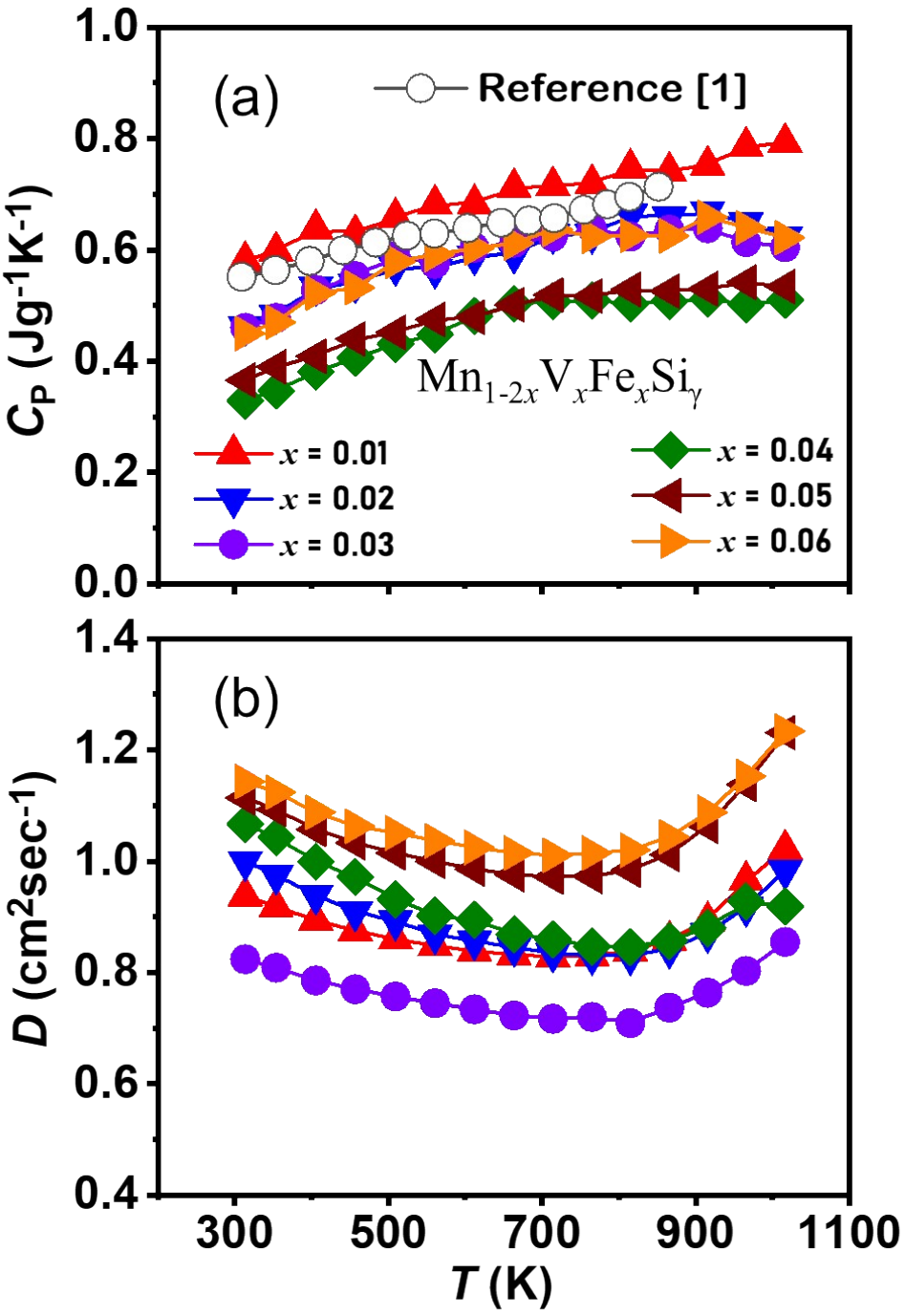

Figure S7. Temperature dependent (a) specific heat capacity (b) thermal diffusivity for nominal $(Mn_{1-2x}V_xFe_x)Si_{1.74}$ alloys. Specific heat reported previously by Chen et al.[1] shown for comparison.

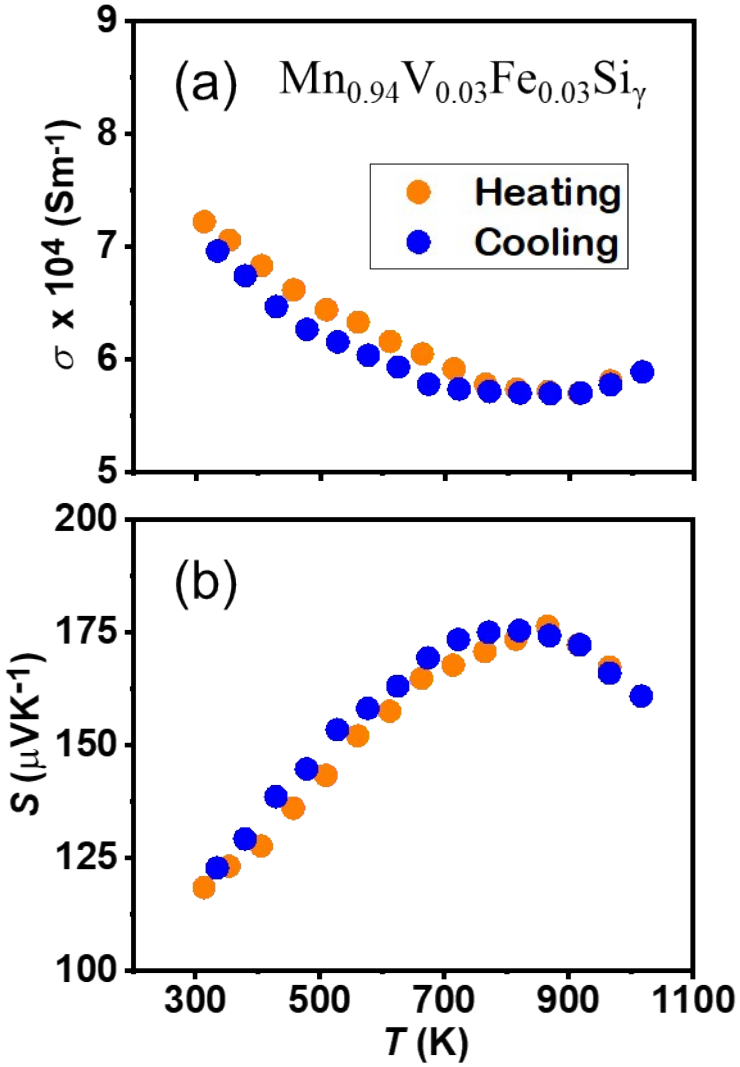

Figure S8. (a) Electrical conductivity and (b) Seebeck coefficient over heating and cooling cycle for representative $(Mn_{0.94}V_{0.03}Fe_{0.03})Si_{1.74}$ single crystal.

S2. Detailed calculation of disorder scattering parameter (Γ). To analyse the κ_L reduction in MnSi alloys, the disorder scattering parameter Γ with mass (Γ_m) and strain (Γ_s) fluctuation component, was evaluated using the model of Slack[2] and Abeles[3] as follows by taking into consideration the fractional occupation at Mn-sublattice by V and Ru as co-dopant.

$$\Gamma = \Gamma_m + \Gamma_s$$

$$\Gamma_m = \Gamma_m^V + \Gamma_m^{Fe}$$

$$\Gamma_s = \Gamma_s^V + \Gamma_s^{Fe}$$

$$\Gamma_m^V = \frac{1}{3}\left(\frac{\bar{M}_V}{\bar{\bar{M}}}\right)x(1-x)\left(\frac{M_V - M_{Mn}}{\bar{M}_V}\right)^2$$

$$\Gamma_m^{Fe} = \frac{1}{3}\left(\frac{\bar{M}_{Fe}}{\bar{\bar{M}}}\right)y(1-y)\left(\frac{M_{Fe} - M_{Mn}}{\bar{M}_{Fe}}\right)^2$$

$$\Gamma_s^V = \frac{1}{3}\left(\frac{\bar{M}}{\bar{\bar{M}}}\right)x(1-x)\varepsilon\left(\frac{r_V - r_{Mn}}{\bar{r}}\right)^2$$

$$\Gamma_s^{Fe} = \frac{1}{3}\left(\frac{\bar{M}}{\bar{\bar{M}}}\right)y(1-y)\varepsilon\left(\frac{r_V - r_{Mn}}{\bar{r}}\right)^2$$

$$\bar{r}_V = r_V x + r_{Mn}(1-x)$$

$$\bar{r}_{Fe} = r_{Fe} y + r_{Mn}(1-y)$$

$$\bar{M}_V = M_V x + M_{Mn}(1-x)$$

$$\bar{M}_{Fe} = M_{Fe} y + M_{Mn}(1-y)$$

$$\bar{\bar{M}}_V = \frac{1}{\gamma+1}(\bar{M}_V + \gamma M_{Si})$$

$$\bar{\bar{M}}_{Fe} = \frac{1}{\gamma+1}(\bar{M}_{Fe} + \gamma M_{Si})$$

where, M_{Mn}, M_V, M_{Fe}, and M_{Si} are the atomic weight of Mn, V, Fe, and Si, respectively; r_{Mn}, r_V, and r_{Fe} are the atomic radius of Mn, V, and Fe, respectively; *x* is the content of V, and *y* is the content of Fe; and ε is regarded as a phenomenological adjustable parameter calculated using the Grüneisen parameter (γ) and Poisson ratio (υ_P).[1,4]

(1) Chen, X.; Girard, S. N.; Meng, F.; Lara-Curzio, E.; Jin, S.; Goodenough, J. B.; Zhou, J.; Shi, L. Approaching the Minimum Thermal Conductivity in Rhenium-Substituted Higher Manganese Silicides. *Adv. Energy Mater.* **2014**, *4* (14), 1400452. https://doi.org/10.1002/aenm.201400452.

(2) Slack, G. A. Effect of Isotopes on Low-Temperature Thermal Conductivity. *Phys. Rev.* **1957**, *105* (3), 829–831. https://doi.org/10.1103/PhysRev.105.829.

(3) Abeles, B. Lattice Thermal Conductivity of Disordered Semiconductor Alloys at High Temperatures. *Phys. Rev.* **1963**, *131* (5), 1906–1911. https://doi.org/10.1103/PhysRev.131.1906.
(4) Sanditov, D. S.; Belomestnykh, V. N. Relation between the Parameters of the Elasticity Theory and Averaged Bulk Modulus of Solids. *Technical Physics* **2011**, *56* (11). https://doi.org/10.1134/S106378421111020X.