Optimization of Electronic Transport in Defected Semiconductor Superlattices

Vitaly Proshchenko & Sanghamitra Neogi

Ann and H.J. Smead Aerospace Engineering Sciencies, University of Colorado Boulder, Boulder, Colorado 80309

Motivation

Thermoelectric materials:
Promising candidates for energy harvesting

- Quiet operation
- No moving parts
- Small size and weight
- Environmentally friendly

$$zT = \frac{S^2\sigma}{\kappa_l + \kappa_e}$$

Phonon scattering
- SL interfaces [1]
- Interstitial defects [2]

Thermal conductivity $\kappa_l + \kappa_e$ ↓ — Power factor $S^2\sigma$ ↑ ⇒ ?

Stratagies to optimize electronic transport in:

- Multilayered systems under strain
- Bulk systems with defects
- Defected multilayered systems

Methods

Boltzmann Transport Equations

$$\mathcal{L}^{(a)} = \frac{e^2}{V}\int_{BZ}\frac{d^3k}{4\pi^3}\left[\tau(\mathbf{k})\nu(\mathbf{k})\nu(\mathbf{k})(\epsilon_\mathbf{k}-\mu(T))^a\left(-\frac{\partial f_\mu}{\partial\epsilon_\mathbf{k}}\right)\right]$$

$\sigma = \mathcal{L}^0$, electrical conductivity

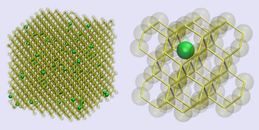
$S = \frac{1}{eT}\mathcal{L}^1/\mathcal{L}^0$ Seebeck coefficient

$\kappa_e = \frac{1}{e^2T}\left(\mathcal{L}^2 - (\mathcal{L}^1)^2/\mathcal{L}^0\right)$ electronic thermal conductivity

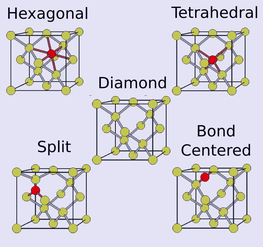
DFT (QuantumEspresso)
- structure relaxation
- band-energies on a fine grid (PBE functional)

Systems

Bulk systems with defects



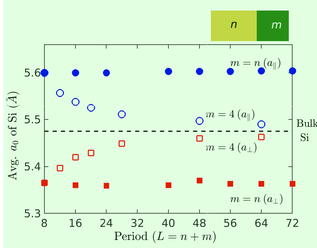
Representative configurations of bulk Si with Ge interstitials



Energetically stable interstitial sites in the diamond lattice

Symmetrically strained SL

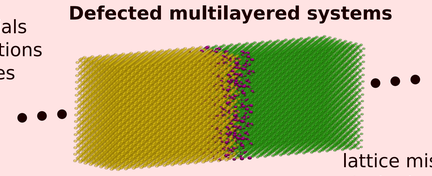
Representative configuration of ideal Si/Ge superlattice



Average lattice constants of confined Si in symetrically strained Si/Ge superlattices

Defected multilayered systems

interstitials
substitutions
vacancies

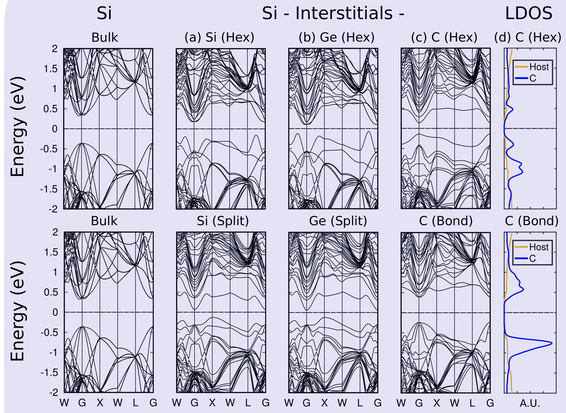


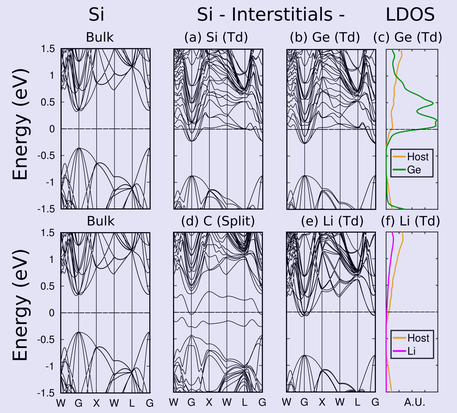
Si/Ge superlattice with defects

lattice mismatch strain due to defects substrate strain

Results & Discussion

Bulk systems with defects

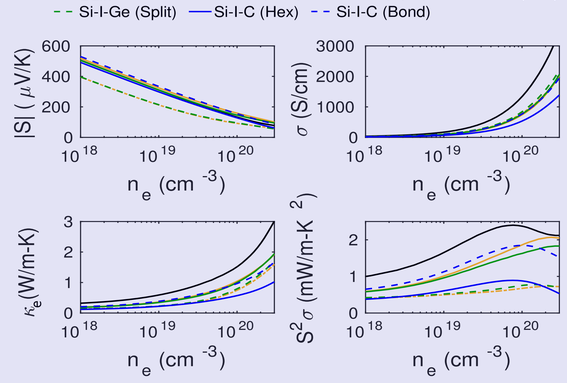




Band structure:

Additional deep and/or shallow energy levels

Same guest atoms placed in different positions lead to new band structures



LDOS:

Guest atoms create additional energy levels

All transport coefficients are altered due to defects

Hex Si and Ge interstitials → highest power factor

Band structure:

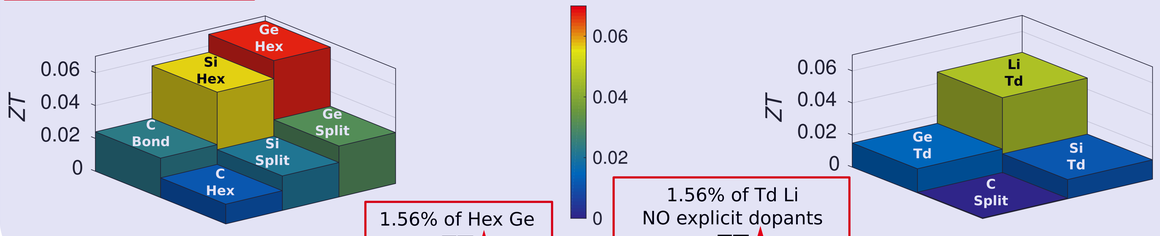
Additional deep and/or shallow energy levels

Fermi level is shifted to conduction zone → hightly n-doped samples

LDOS:

Tetrahedral Li do not contribute to formation of new levels but increase carrier concentration → band structure is almost unchanged

Structure-thermoelectric property map of bulk with interstitial defects

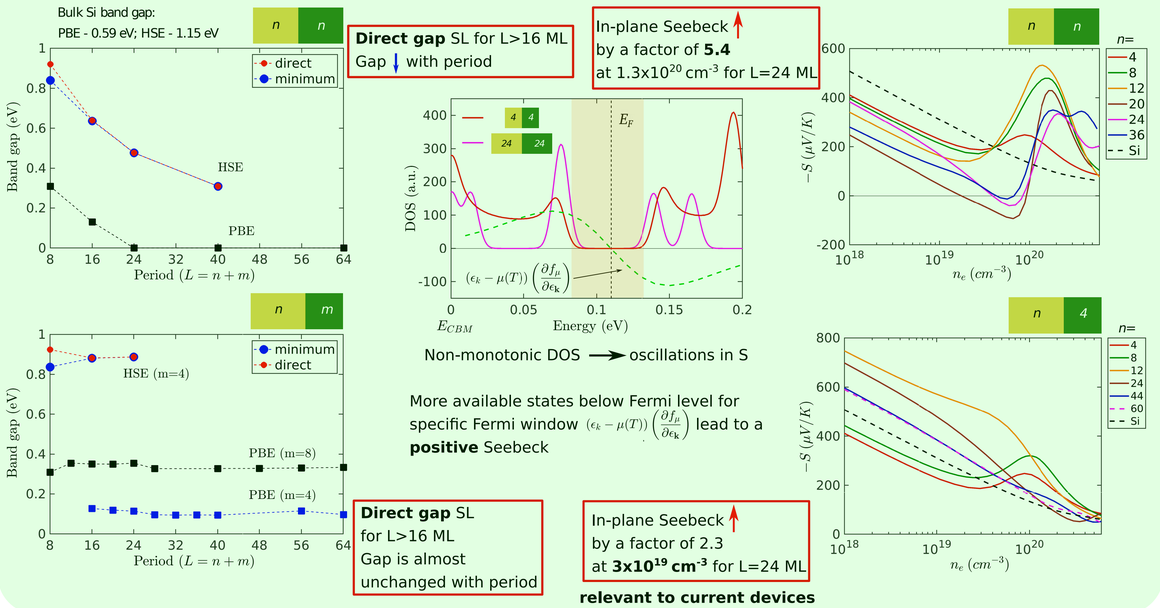


Bulk Si: ~0.004

1.56% of Hex Ge **ZT** ↑ by a factor of **17**

1.56% of Td Li NO explicit dopants **ZT** ↑ by a factor of **12**

Symmetrically strained SL



Direct gap SL for L>16 ML
Gap ↓ with period

In-plane Seebeck ↑ by a factor of **5.4** at 1.3x10^{20} cm^{-3} for L=24 ML

Non-monotonic DOS → oscillations in S

More available states below Fermi level for specific Fermi window $(\epsilon_k - \mu(T))\left(\frac{\partial f_\mu}{\partial \epsilon_k}\right)$ lead to a **positive** Seebeck

Direct gap SL for L>16 ML
Gap is almost unchanged with period

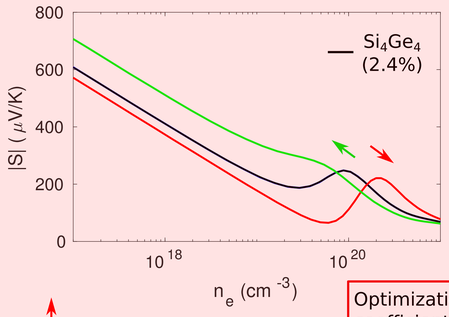
In-plane Seebeck ↑ by a factor of 2.3 at **3x10^{19} cm^{-3}** for L=24 ML

relevant to current devices

Ongoing work

Defected multilayered systems

Interstitial defects introduce:
- additional energy levels
- strain

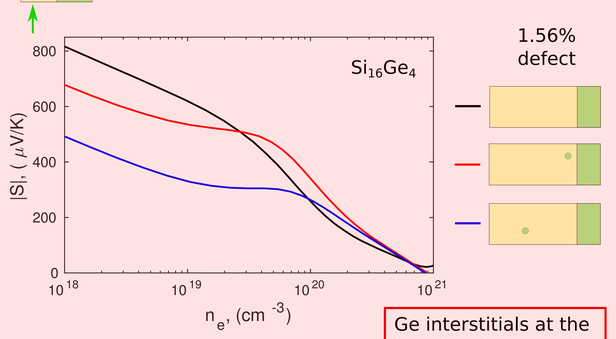


Ideal SL
3.0%
2.0%

Optimization of the Seebeck coefficient with strain

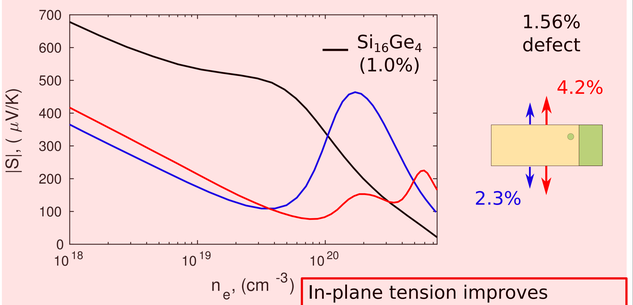
→ S ↑ at **higher** n_e

→ S ↑ at **lower** n_e



1.56% defect

Ge interstitials at the interface improve the Seebeck coefficient



1.56% defect
4.2%
2.3%

In-plane tension improves the Seebeck coefficient in defected $Si_{16}Ge_4$ SL by a factor of **2.4** at 2x10^{20} cm^{-3}

Conclusions

- 1.56% of Ge interstitial defects placed in hexagonal sites of bulk Si provide the best improvement of ZT by a factor of 17
- Interstitials offer a viable strategy to design n-type semiconductors without explicit dopants
- Seebeck coefficient becomes tunable with composition and period of SL
- Strain introduced by engineered defects has potential to to improve the Seebeck coefficient of multilayered systems

[1] S.-M. Lee, D. G. Cahill et.al., Appl. Phys. Lett. 70, 2957 (1997)
[2] P. Chen, N. A. Katcho et. al., Phys. Rev. Lett. 111, 115901 (2013)

Acknowledgements: The project is funded by the Defense Advanced Research Projects Agency (Defense Sciences Office) [Agreement No.: HR0011-16-2-0043].
This work used the Extreme Science and Engineering Discovery Environment (XSEDE), which is supported by National Science Foundation grant number ACI-1548562.
This work utilized the RMACC Summit supercomputer, which is supported by the National Science Foundation (awards ACI-1532235 and ACI-1532236), the University of Colorado Boulder, and Colorado State University.