Machine Learning Assisted Prediction of Aperiodic Superlattice Structures with Ultralow Lattice Thermal Conductivity for NASA's Radioisotope Thermoelectric Generators

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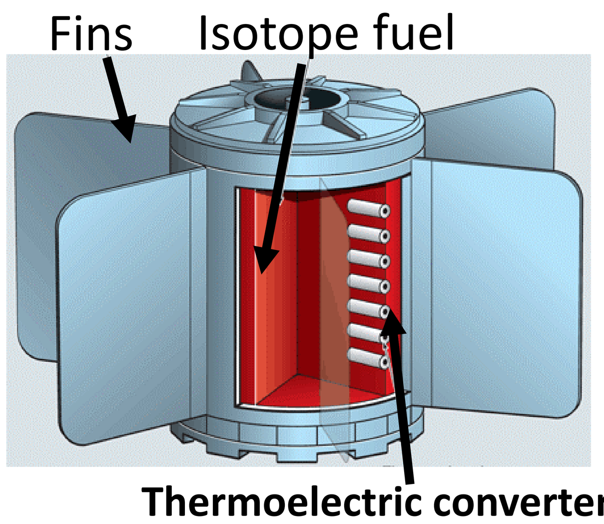
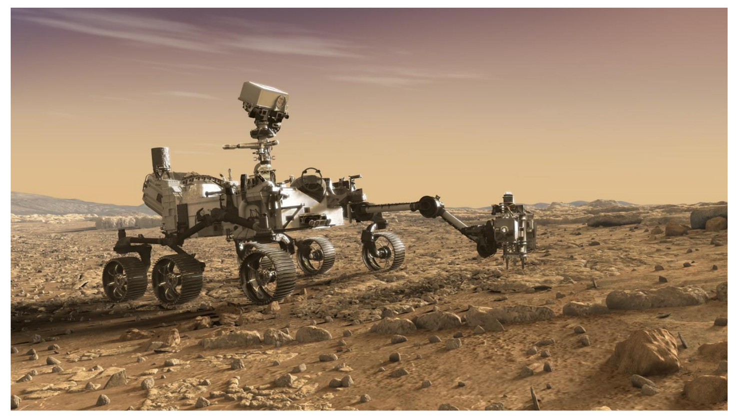
Why do we need efficient thermoelectric (TE) devices?



Radioisotope thermoelectric generator (RTG)



Mars rover

- **NASA Applications:** RTG is the primary power source of most of NASA's deep space missions. The poor efficiency of TE materials is one significant limiting factor for such RTGs.
- **Energy Recycling:** Around 70% of the consumed energy is wasted in the form of low-grade heat. Thermoelectric devices can generate usable electricity from the waste heat.
- **Cooler or Refrigerator:** Thermoelectric coolers are noise free, do not need working fluids, and occupy less space. Thermoelectric materials are used for cooling small-scale devices.

Towards better thermoelectric materials

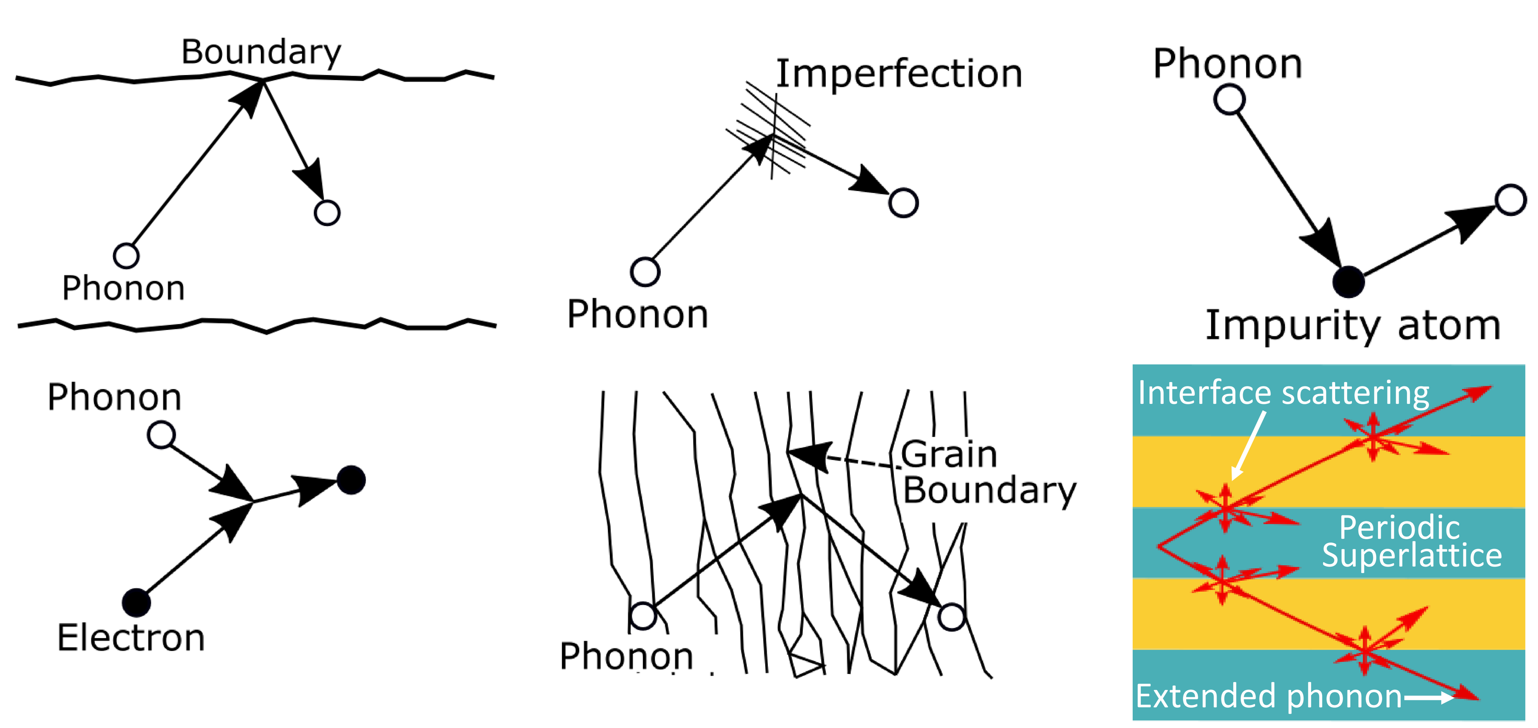
- Figure of merit (ZT) of thermoelectric materials

$$ZT = \frac{\sigma S^2 T}{\kappa_e + \kappa_L}$$

Electronic thermal conductivity (κ_e); Lattice thermal conductivity (κ_L)

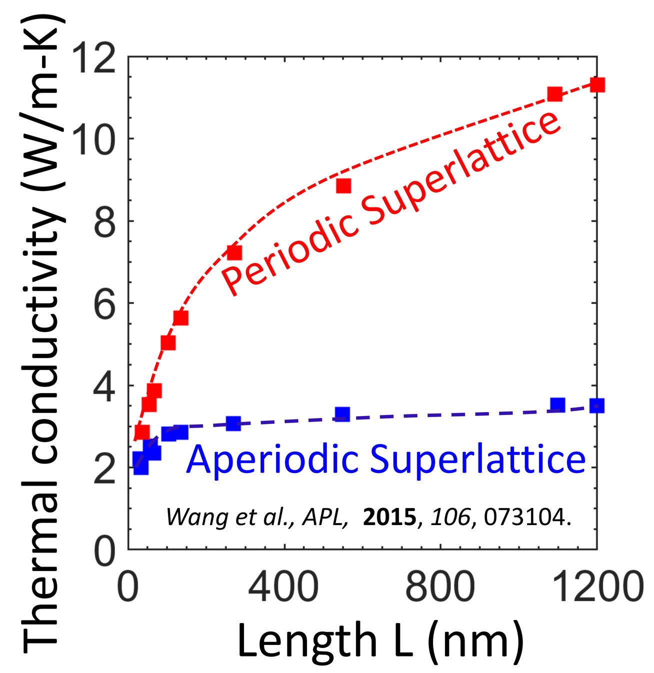
σ = Electrical conductivity
S = Seebeck coefficient
T = Temperature
κ = Thermal conductivity

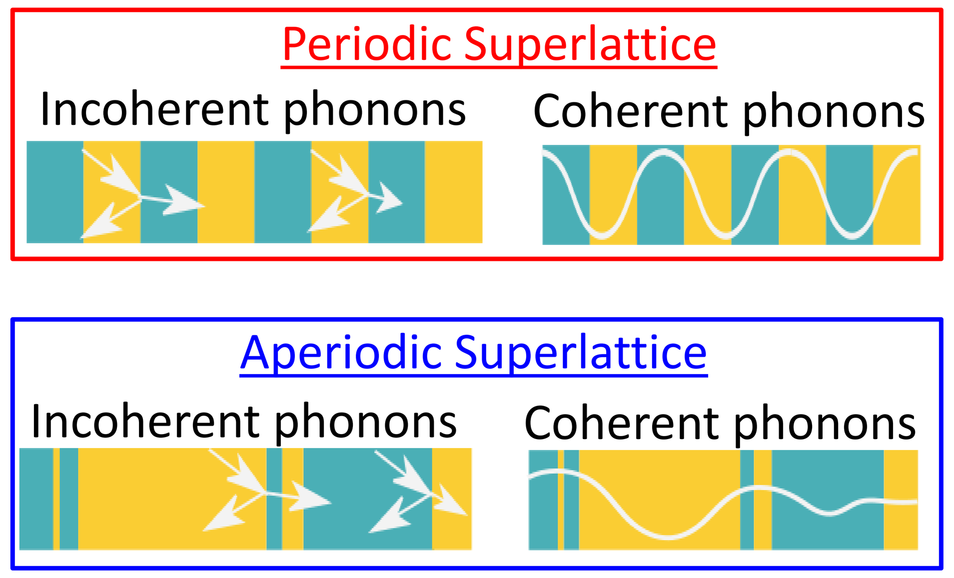
- Minimizing the lattice thermal conductivity while retaining good electrical conduction is essential for developing high-ZT thermoelectric materials.
- Several scattering strategies are used for reducing κ_L................



- These scattering mechanisms are well studied and the outcome tends to be saturated. We need new strategies to further reduce κ_L.

Aperiodic superlattice structures exhibit significantly lower thermal conductivity than periodic superlattice



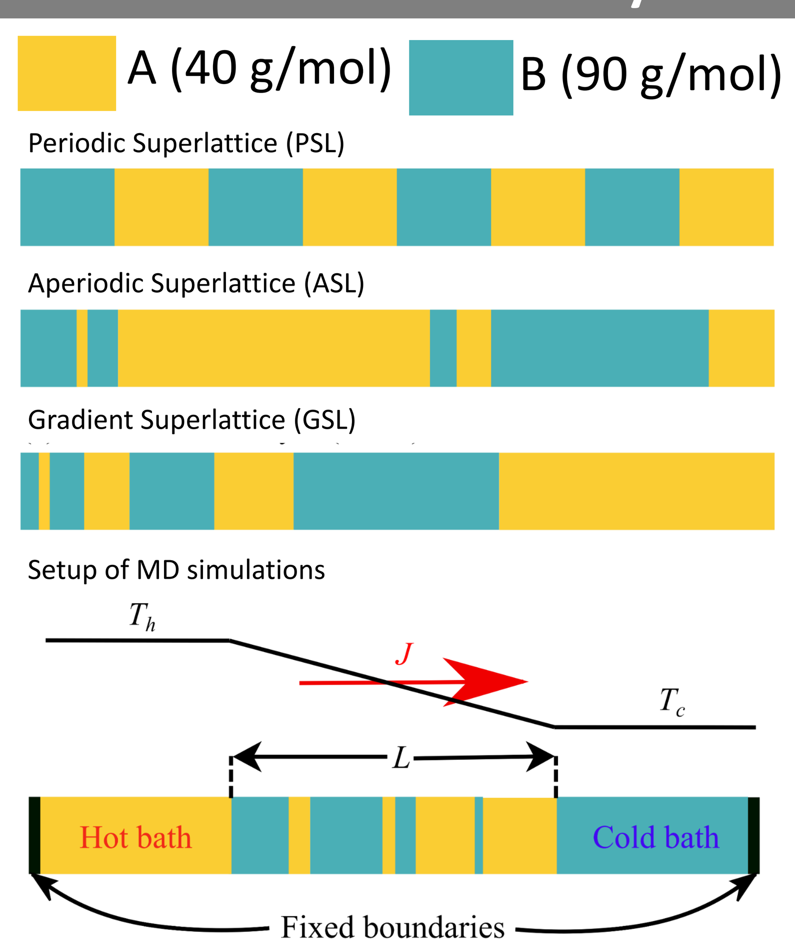


Aperiodic Superlattice (ASLs) localize the coherent phonon transport and therefore, exhibit lower thermal conductivity than superlattice structure

Objective of this work

- Correlate the randomness and thermal conductivity of ASLs
- Achieve optimized ASLs through machine learning
- Further reduction of ASL thermal conductivity through rational doping

Molecular dynamics (MD) simulations



- Lennard-Jones potential

$$\Phi_{ij} = 4\varepsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right]$$

- $\varepsilon = 0.1664$ eV and $\sigma = 0.34$ nm
- Conceptual materials for computational efficiency
- LAMMPS package
- Non-equilibrium molecular dynamics simulations
- κ_L is calculated as

$$\kappa_L = \frac{JL}{A_c(T_h - T_c)}$$

Quantification of randomness in aperiodic superlattice (ASL)

- We identified a few disorder parameters to correlate the randomness with κ_L of ASLs obtained through molecular dynamics simulations
- Thickness based randomization index

$$R_d = \sqrt{\frac{\sum_{i=2}^{N}\left[(d_{A,i} - d_{A,i-1})^2 + (d_{B,i} - d_{B,i-1})^2\right]}{N}}$$

- Period based randomization index

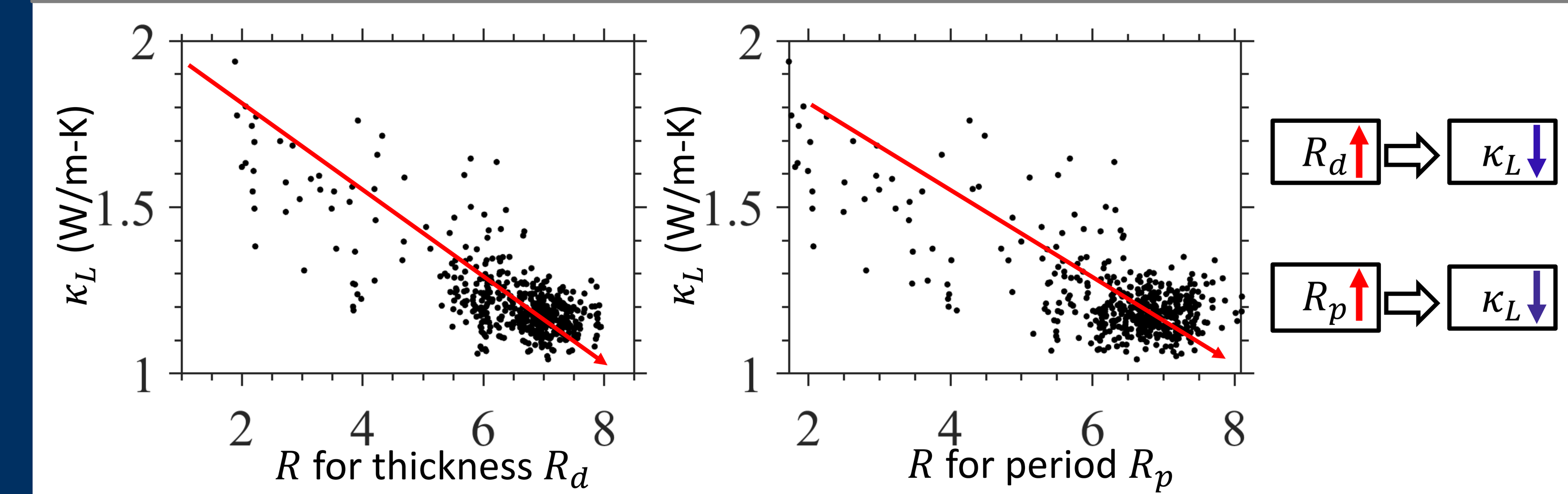
$$R_p = \sqrt{\frac{\sum_{i=2}^{N}\left[(d_{A,i} + d_{B,i})^2 + (d_{A,i-1} + d_{B,i-1})^2\right]}{N}}$$

- Standard deviation

$$\delta = \sqrt{\frac{\sum_{i=1}^{N}\left[(d_{A,i} - d)^2 + (d_{B,i} - d)^2\right]}{2N}}$$

Higher R_d, R_p, δ should result in lower κ_L

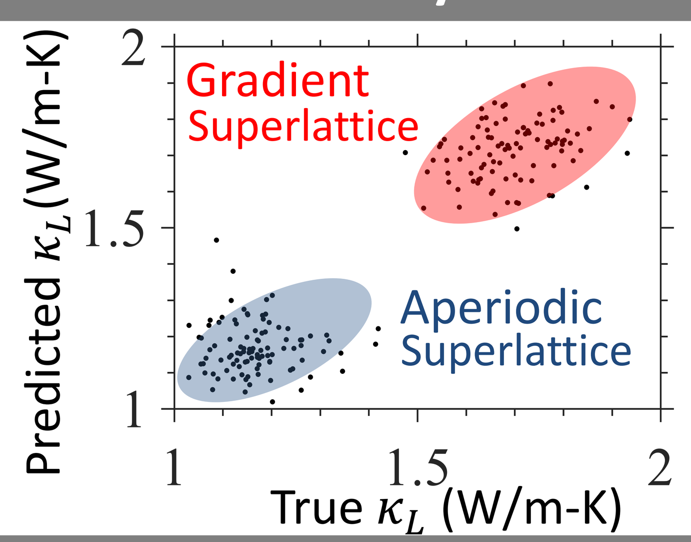
Randomness index and lattice thermal conductivity (κ_L) relation



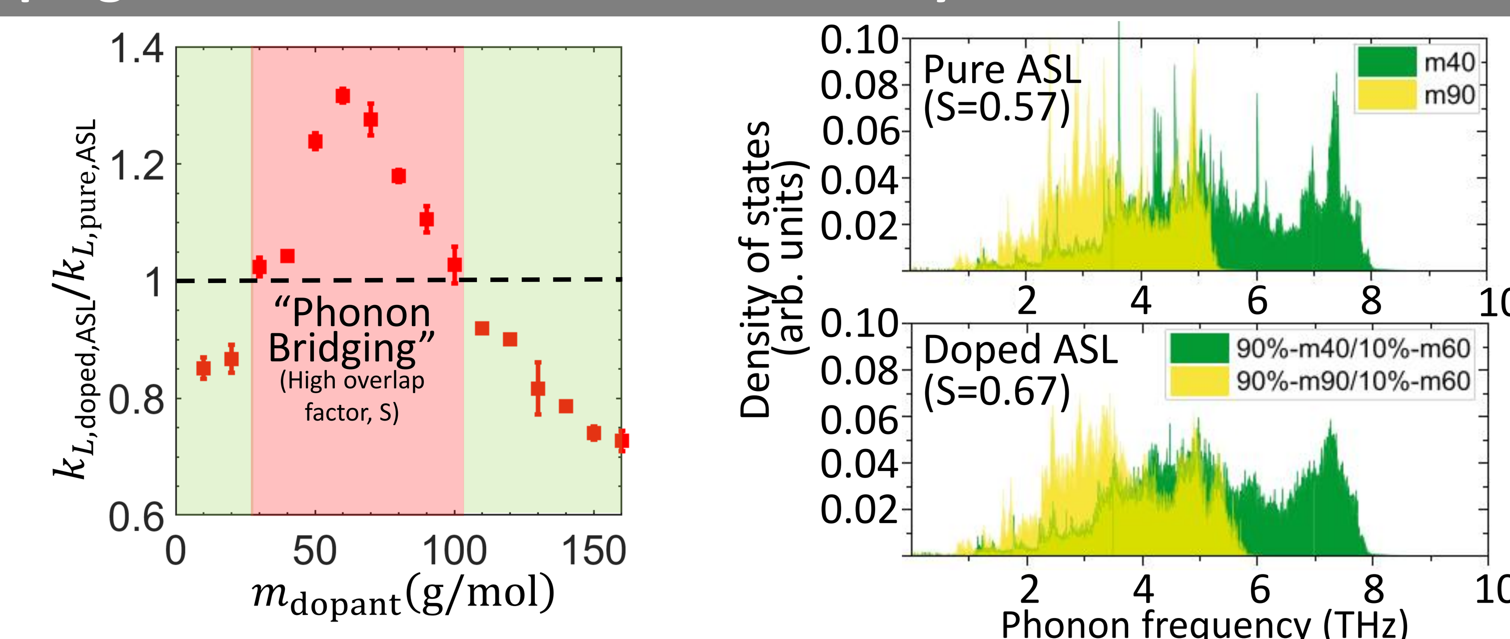
- We still could not find the best ASL configuration. Machine learning can aid us for the same.

Machine learning prediction of thermal conductivity

- Neural network
- Training data (3200 structures)
- Testing data (200 structures)
- Python packages
- Features: Thickness sequence, R_p, R_d, and δ.
- Machine learning model predicted k_L are compared with true-k_L



Doping reduces the thermal conductivity of ASLs even further



- Recommendations: $m_{\text{dopant}} \gg m_{\text{A,B(ASL)}}$ or $m_{\text{dopant}} \ll m_{\text{A,B (ASL)}}$

Conclusions

- We have identified several key features for machine learning to predict thermal conductivity of multilayer structures.
- We have demonstrated that rational doping can reduce the thermal conductivity of aperiodic superlattice structures even further

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References

1) Chakraborty, P; Liu, Y; Ma, T; Guo, X; Cao, L; Hu, R; Wang, Y. Quenching thermal transport in aperiodic superlattice: a moleucular dynamics and machine learning study. Applied Materials and Interfaces 2020.