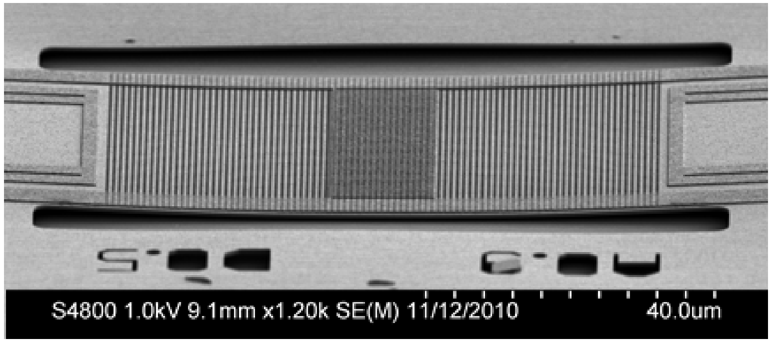

Metallic Inclusions in Phononic Crystals for Thermoelectric Applications

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U.S. DEPARTMENT OF ENERGY

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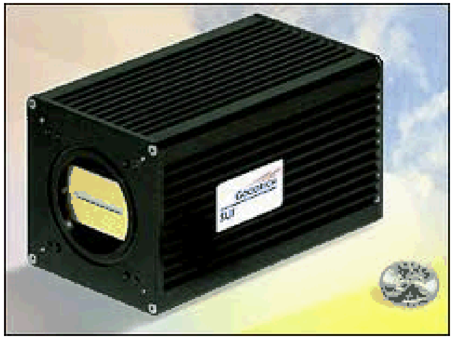
Motivation: Thermoelectrics

Motivation

- **Efficient Thermoelectrics:**
  - » Cooling
  - » Power generation
- **Applications:**
  - » Sensors in space applications
  - » Industrial waste heat recovery
  - » Energy scavenging ➔ solar thermophotovoltaics
  - » Integrated microprocessor cooling for high-performance computing



Objective: High-ZT

ZT – Thermoelectric Figure-of-Merit

The ZT-Problem:

$ZT = S^2\sigma/\kappa T$ ➔ need $S\uparrow, \sigma\uparrow, \kappa\downarrow$

- The S-σ Problem:
  - » S is related to the electronic **entropy** (disorder/scattering) in the system
  - » σ is a measure of how fast we can propagate a current➔ decreases with increasing disorder
    - » Mott Equation:

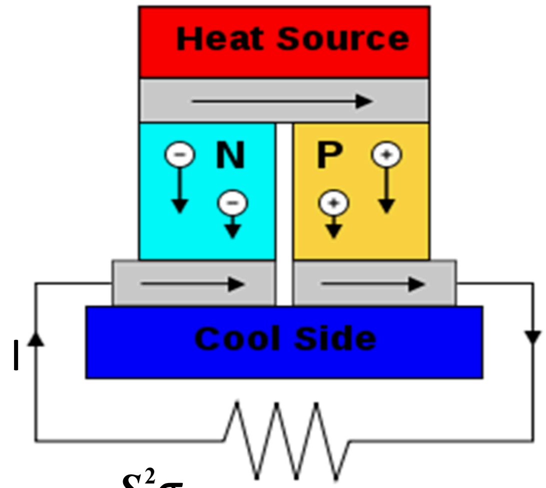
$$S = \frac{1}{\sigma}\frac{d\ln(\sigma(\varepsilon))}{d\varepsilon}\bigg|_{\varepsilon=\varepsilon_F}$$

➔ $S\uparrow \Leftrightarrow \sigma\downarrow$

- The κ-σ Problem:
  - » In semiconductors, "κ" is dominated by the phonon contribution
  - » κ reduced by increasing phonon scattering
    - » At the limit, this leads to an increase in electron scattering

➔ $\kappa\downarrow \Leftrightarrow \sigma\downarrow$

TE Generator



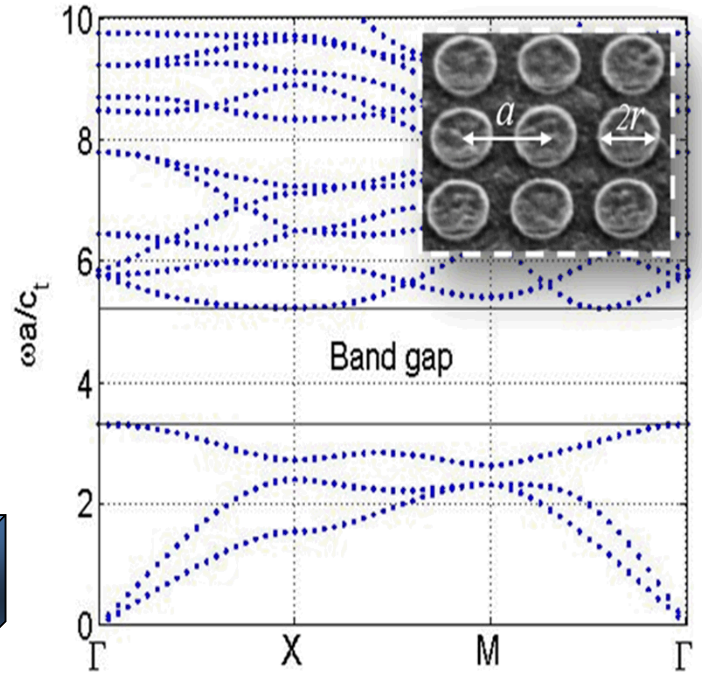
$$ZT = \frac{S^2\sigma}{\kappa}$$

S = Seebeck coefficient
σ = electrical conductivity
κ = thermal conductivity
T = temperature

$$\eta_{max} = \frac{T_H - T_C}{T_H}\frac{\sqrt{1+Z\bar{T}}-1}{\sqrt{1+Z\bar{T}}+\frac{T_C}{T_H}}$$

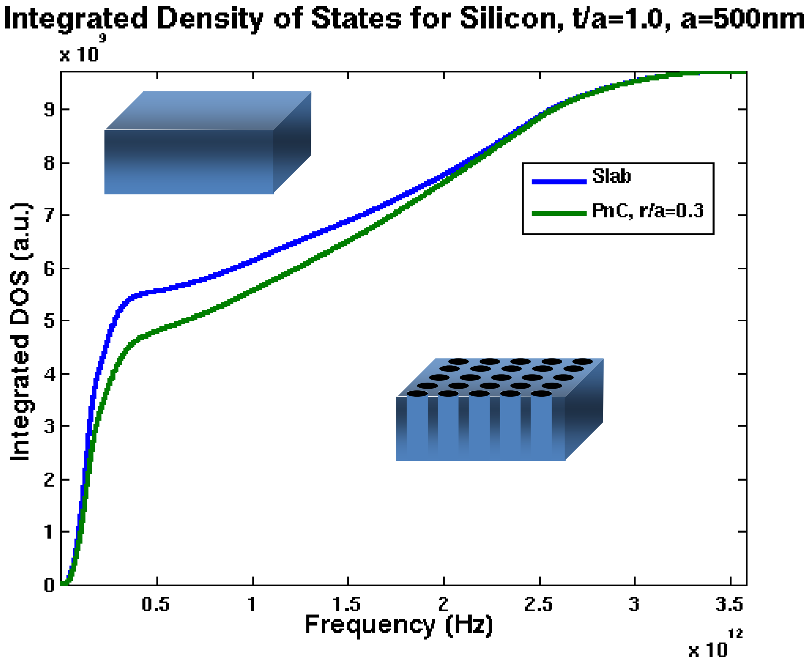
Phononic Crystals

Decoupling κ and σ

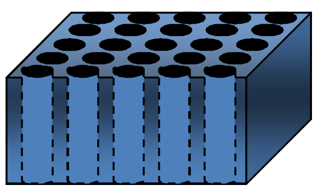


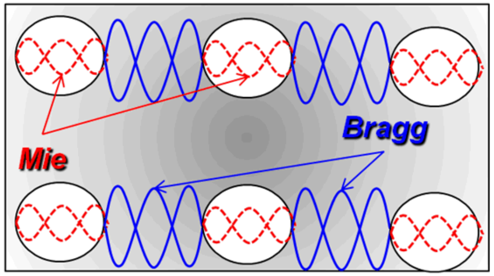
What is a phononic crystal (PnC)?

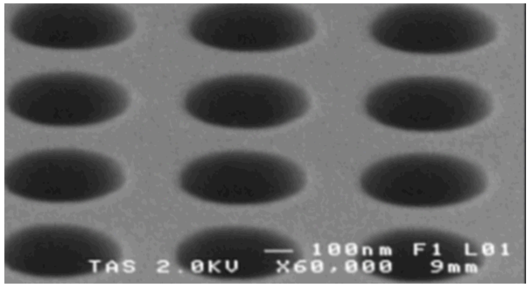
- Periodic arrangement of elastic scattering centers in a matrix material that exhibits both incoherent and Mie and Bragg resonant scattering
- Requires sufficient mechanical impedance mismatch



PnC





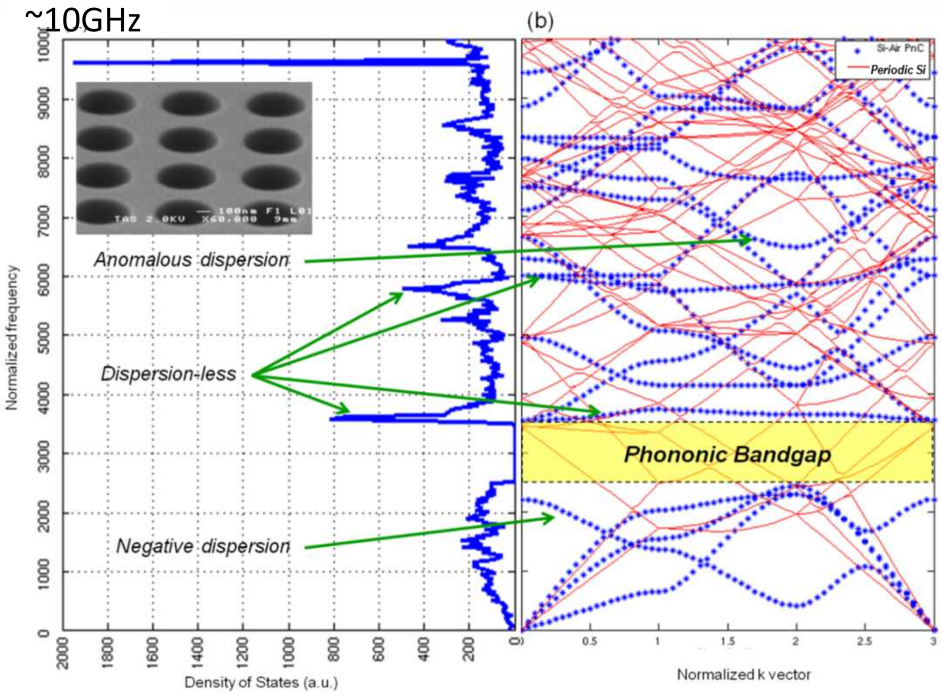


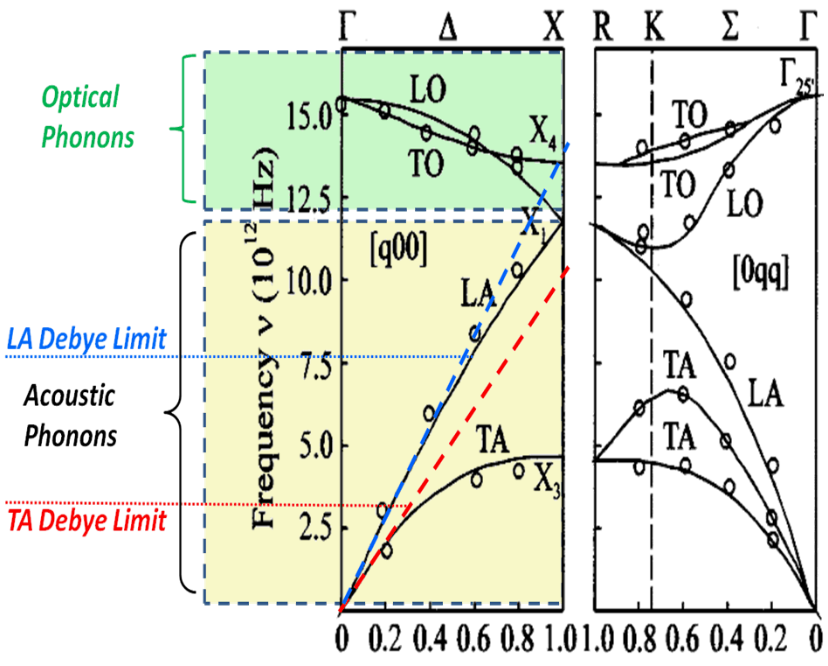
Phononic Crystals

Coherent Phonon Scattering

Thermal transport in phononic crystals

- Coherent (lattice) scattering in addition to material removal and incoherent (boundary) scattering
- Periodicity ➔ Real (as oppose to artificial) zone folding
- Flat Bands ➔ Reduced group velocity, modified density of states
- Negative Bands ➔ Backward propagation (backward scattering)

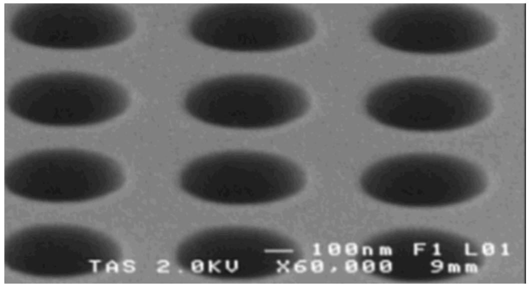




De-Coupling κ and σ in PhCs

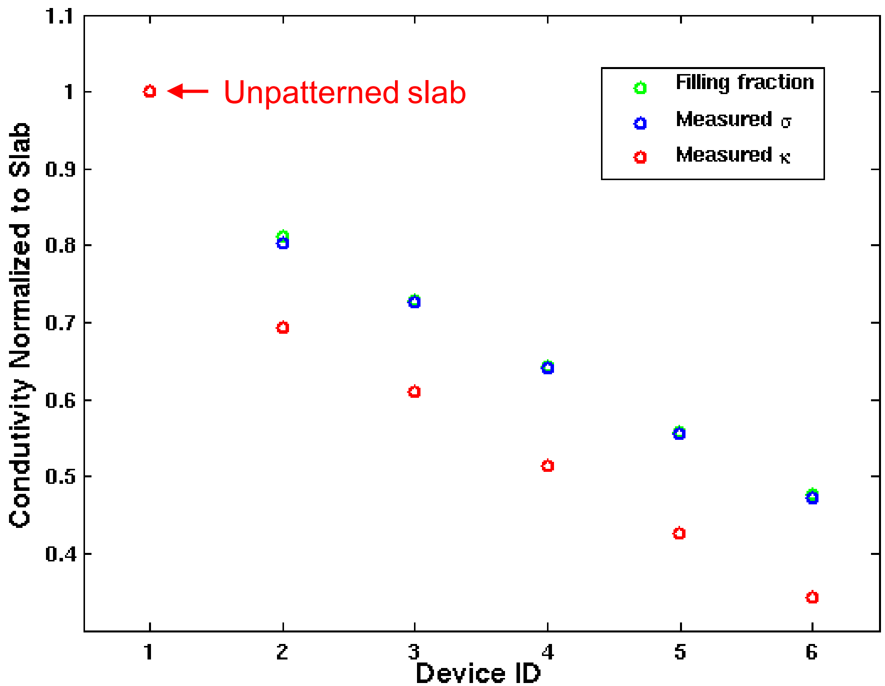
Si-Air PnCs

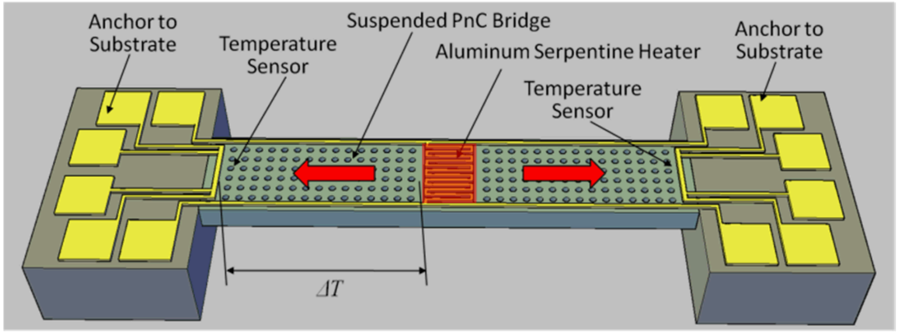
Si-W PnC

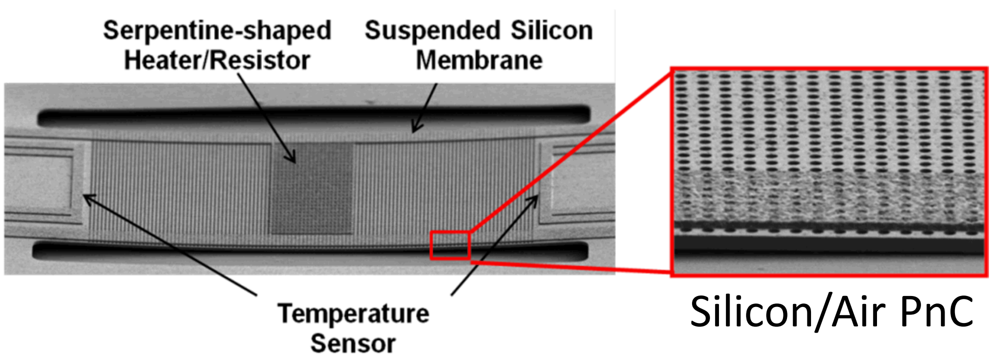


How do phononic crystals affect κ?

- Modification of phonon dispersion "$\omega(k)$", which is related to the phonon group velocity and influences the phonon scattering lifetime
- Incoherent scattering due to the interface at each inclusion
- Redistribution of the phonon density of states as compared with bulk







Silicon/Air PnC

PnC Thermal Conductivity

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Callaway-Holland Model

How do phononic crystals affect κ?

- Modification of phonon dispersion "$\omega(k)$", which is related to the phonon group velocity and influences the phonon scattering lifetime
- Incoherent scattering due to the interface at each inclusion
- Redistribution of the phonon density of states as compared with bulk

$\omega(k)$ is the phonon dispersion
$\upsilon(k) = \partial\omega(k)/\partial k$ is the phonon group velocity
$\tau(k)$ is the phonon scattering lifetime
k is the wavevector
j = 1, 2, 3 (1 longitudinal and 2 transverse modes)
L is the limiting dimension (smallest feature size)

Thermal conductivity given by Callaway-Holland model:

$$\kappa = \frac{1}{6\pi^2}\sum_j \int_k \frac{\hbar^2\omega_j^2(k)}{k_B T^2} \frac{\exp\left[\frac{\hbar\omega_j(k)}{k_B T}\right]}{\left(\exp\left[\frac{\hbar\omega_j(k)}{k_B T}\right]-1\right)^2} \upsilon_j^2(k)\tau_j(k)k^2 dk$$

$$\frac{1}{\tau_j(k)} = \frac{1}{\tau_{Umklapp,j}} + \frac{1}{\tau_{impurity,j}} + \frac{1}{\tau_{boundary,j}}$$

$$\frac{1}{\tau_{Umklapp,j}} = BT\omega_j^2(k)\exp\left[\frac{C}{T}\right]$$

$$\frac{1}{\tau_{impurity,j}} = D\omega_j^4(k)$$

$$\frac{1}{\tau_{boundary,j}} = \frac{L}{\upsilon_j(k)}$$

where *B*, *C*, *D*, and *E* are constants determined by fitting κ to experimental data

To modify κ, we need to change:

a. **Dispersion "$\omega(k)$"**
b. **Minimum feature size "*L*"**

PnC Thermal Conductivity

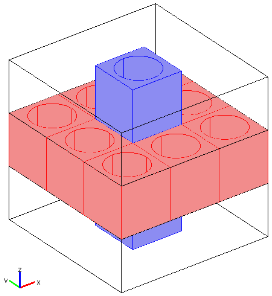
Hybrid LD + PWE Approach

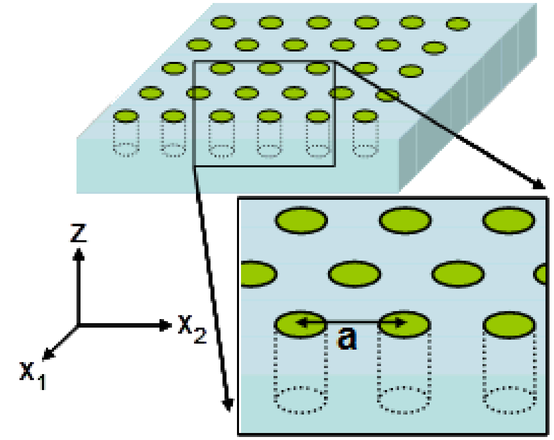
Plane-wave expansion (PWE) combined with lattice dynamics

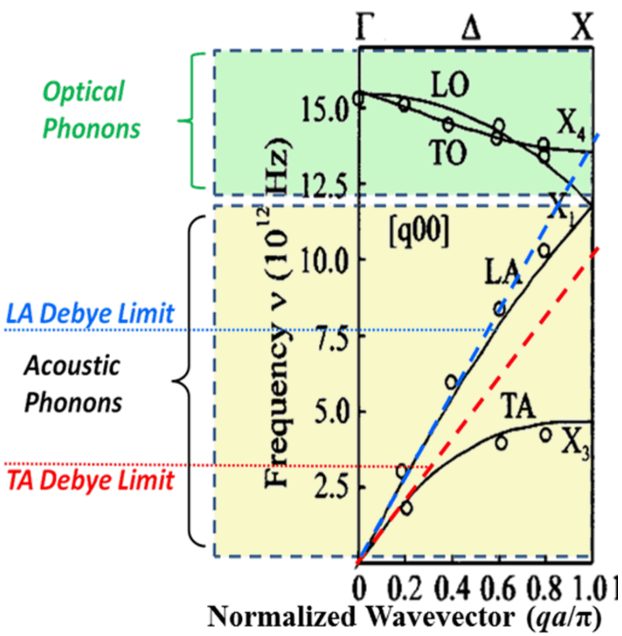
- » Continuum mechanics-based PWE theory breaks down at nanometer length scales
- » Lattice dynamics/molecular dynamics modeling becomes computationally intensive at the same limit
- » Simulation tool developed for calculating *κ* that supplements continuum mechanics with corrections from lattice dynamics

PWE dispersion:

- » Quantitatively valid in the Debye-limit portion of the dispersion
- » Qualitatively valid in the linear/slow portion of the material dispersion
- » Must calculate >45000 bands for room-temperature *κ* in Si







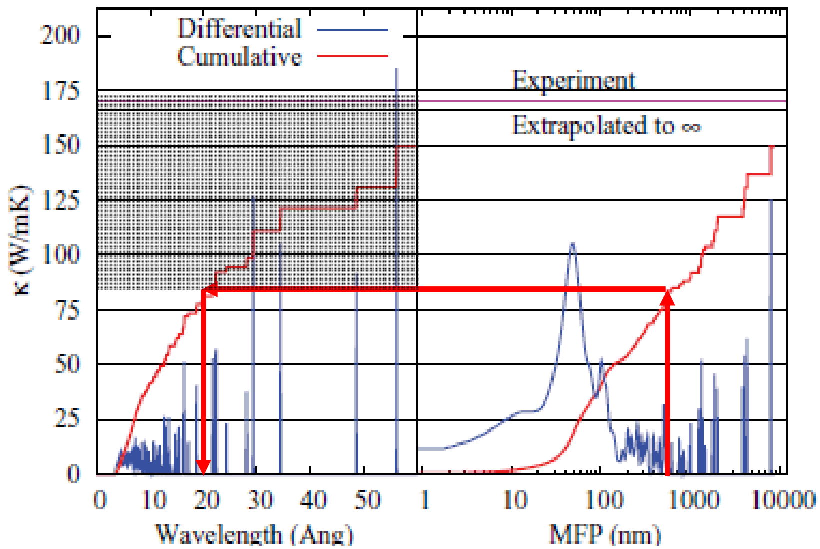
PnC Thermal Conductivity

Threshold Mean Free Path

Prediction of κ using our hybrid model

- Dispersion data of PnCs performed up to ~2THz using plane-wave expansion (PWE) used directly to calculate κ
  - Threshold MFP (TMFP) uses phonon MPF data to distinguish the phonon population as coherent or incoherent
  - Phonons with a MFP of **2µm and longer** experience coherent PnC scattering

➔ Si MFPs span from 1nm to 100µm at T_{room}

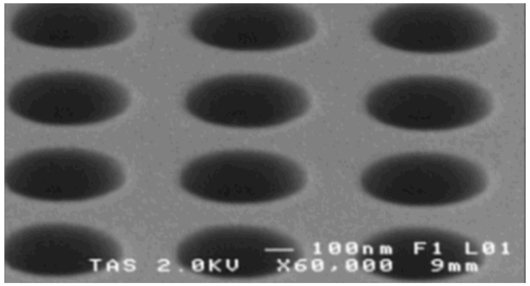


Gang Chen, *et. al.*, *PRB* **84**, 085204 (2011)

PnC Thermal Conductivity

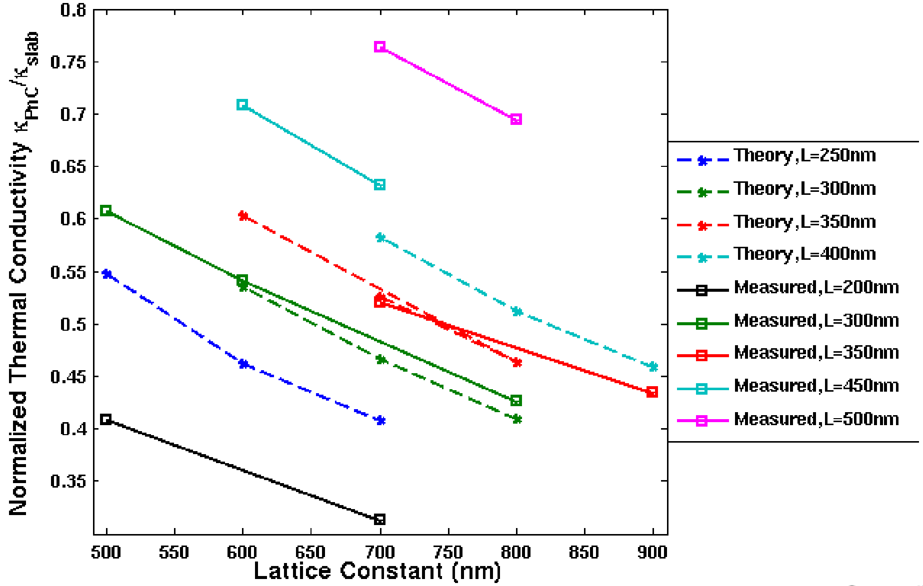
Verification of Hybrid Model

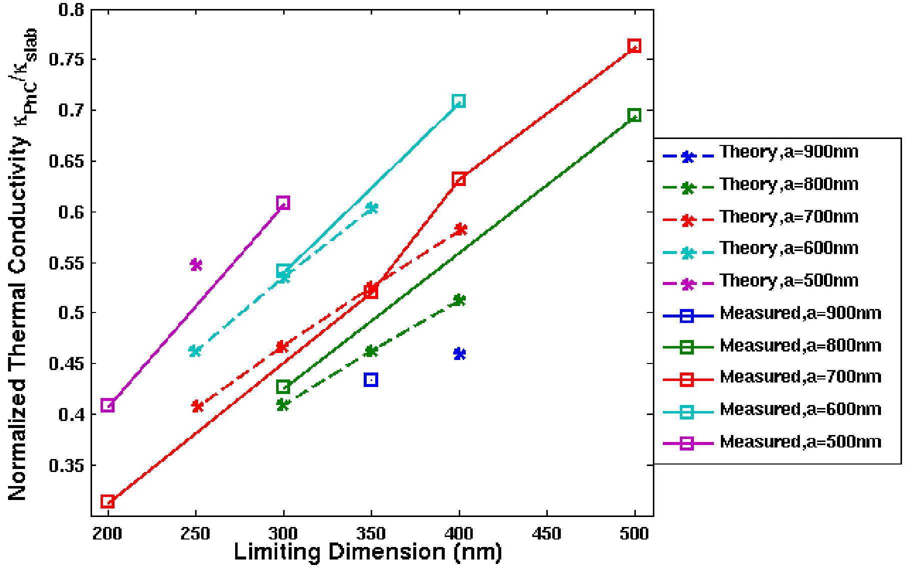
Si-W PnC

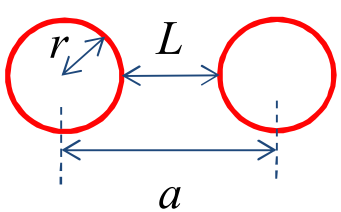


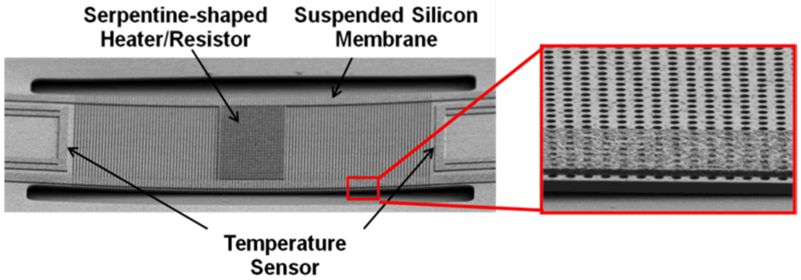
Coherent vs. Incoherent Phonon Scattering in Si/Air PnCs:

- Theoretical predictions of κ reduction match well with measured data
  - Fabricated parameters varied slightly from what was specified
  - TMFP = 2µm was assumed for all PnCs





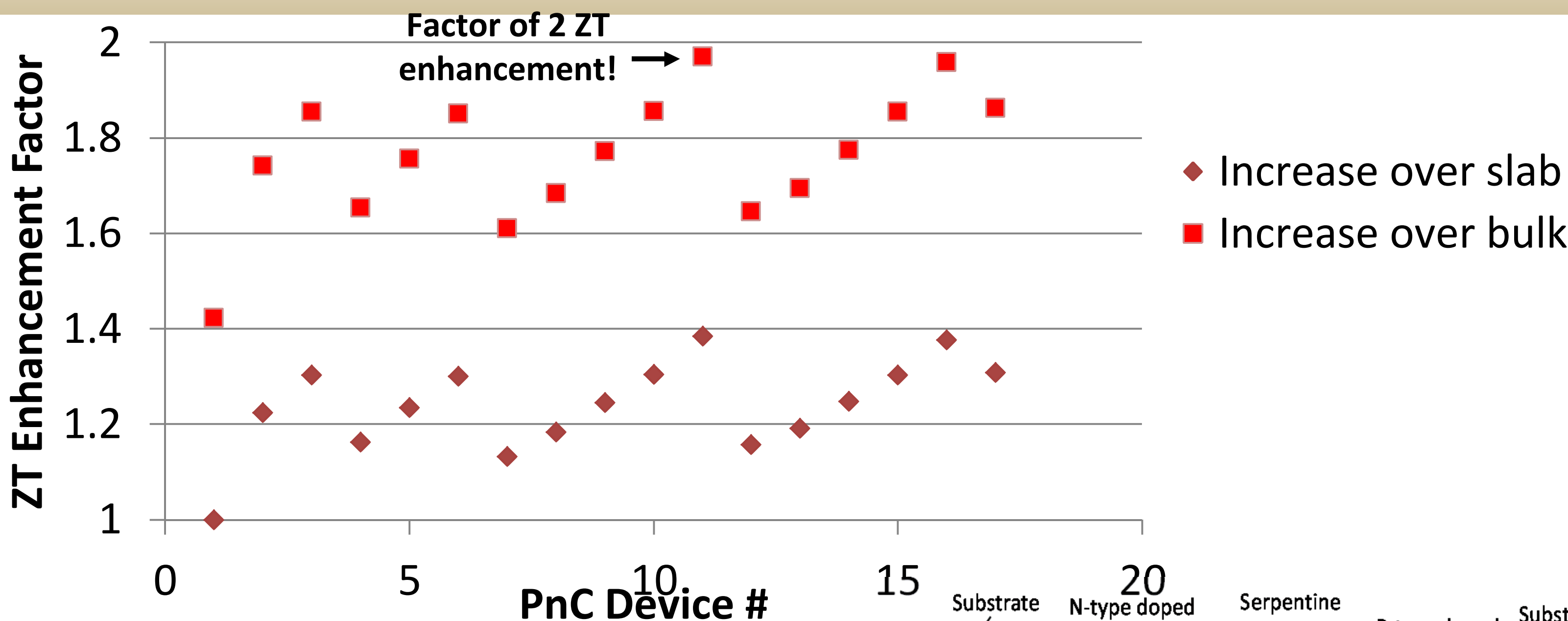


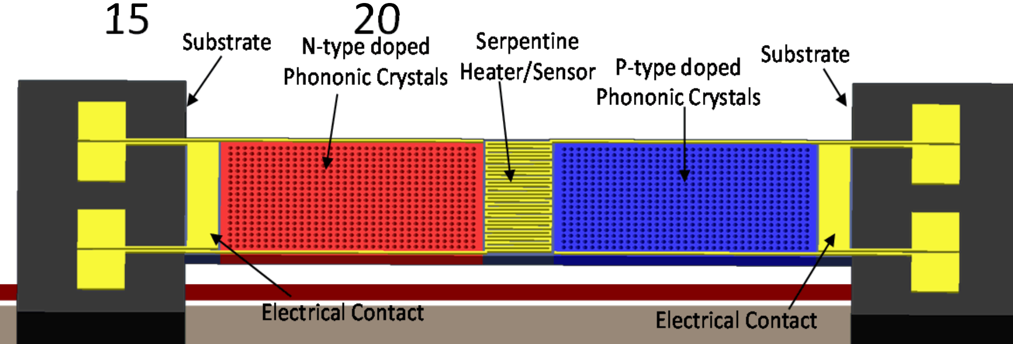


ZT Prediction

Overall System Performance

- » Enhancement in ZT by a factor of 2x versus bulk Si
  - » 1.4x Enhancement as compared to an unpatterned slab
- » Applicable to any material system
  - » ZT must be limited by high κ, and have a κ that is dominated by phonon transport (i.e. most semiconductors)





Concurrent Engineering of σ and κ

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Si-Metal PnCs

Concurrent σ Enhancement and κ Reduction:

» Si-metal PnCs to reduce κ with simultaneous σ enhancement

» Is σ actually enhanced, and is κ still reduced without the porosity factor?

➔ Measure thermal and electrical conductivity of existing Si-W samples

- Effect on σ :

➢ Increased doping:

» Decreases the interface potential width ➔ electron tunneling

➢ Ohmic contact at interface:

$$\sigma_{PnC+W} \approx \sigma_{Si} \frac{1+ff_W}{1-ff_W}$$

- Effect on κ :

➢ Solid-solid PnC rather than solid-air:

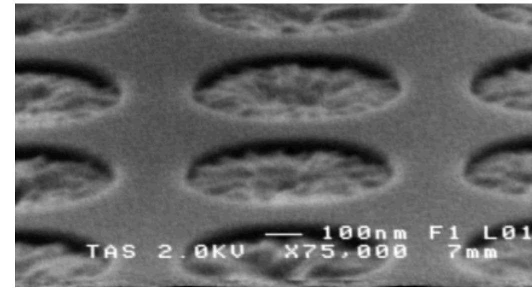
» Mechanical impedance mismatch is still very high

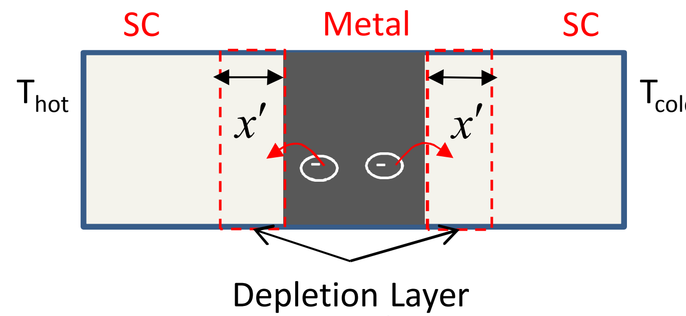
» Scatterer thermal conductivity is no longer negligible

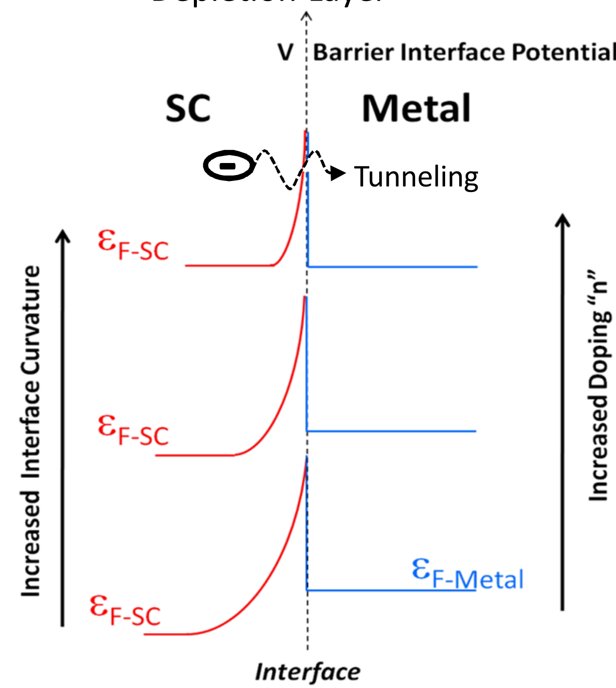
» Boundary thermal resistance must now be considered

$$\kappa_{PnC+W} \approx \kappa_{Si} \frac{C-ff_W}{C+ff_W}, C=1+2*\frac{\kappa_W}{\kappa_{Si}}$$

Si-W PnC







Concurrent Engineering of σ and κ

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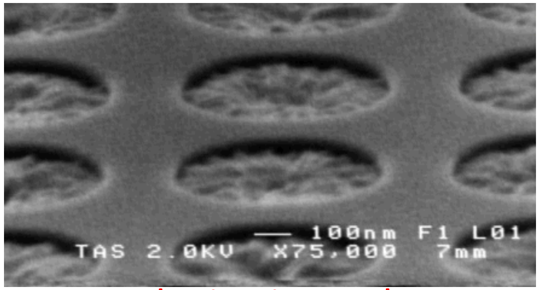
Theoretical Prediction

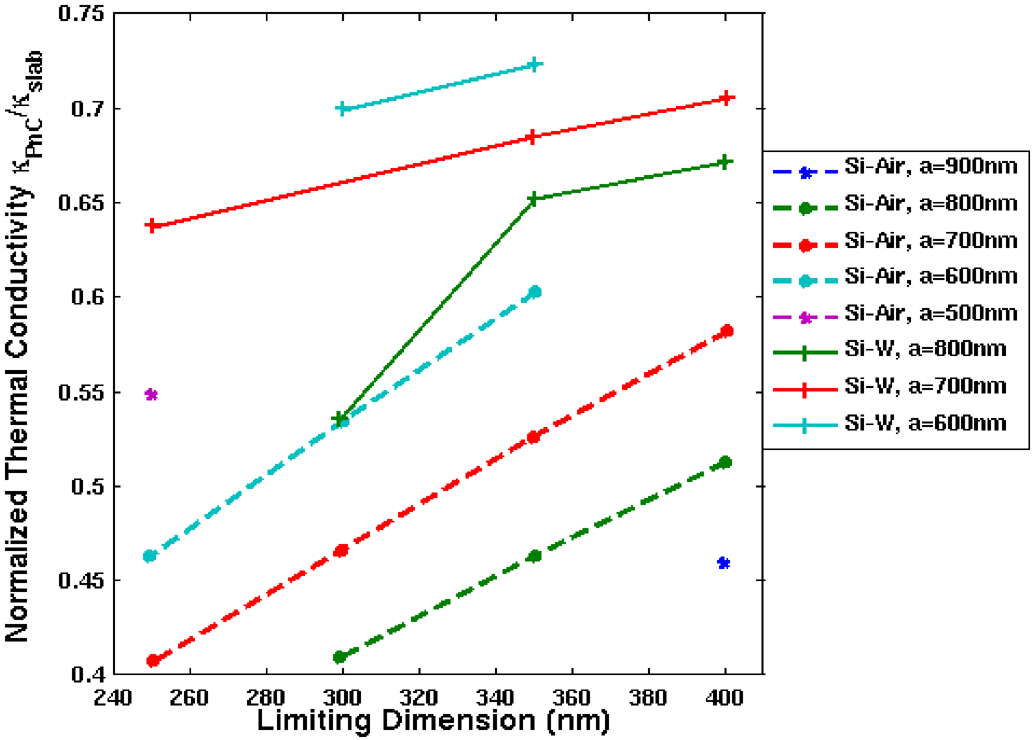
Si-W PnC



κ and σ in Si-metal PnC

Concurrent σ Enhancement and κ Reduction:

- Compared with Si-air, Si-W PnCs are predicted to have:
  - ~15% less thermal conductivity reduction, but
  - 1.7x – 3.8x enhancement in electrical conductivity

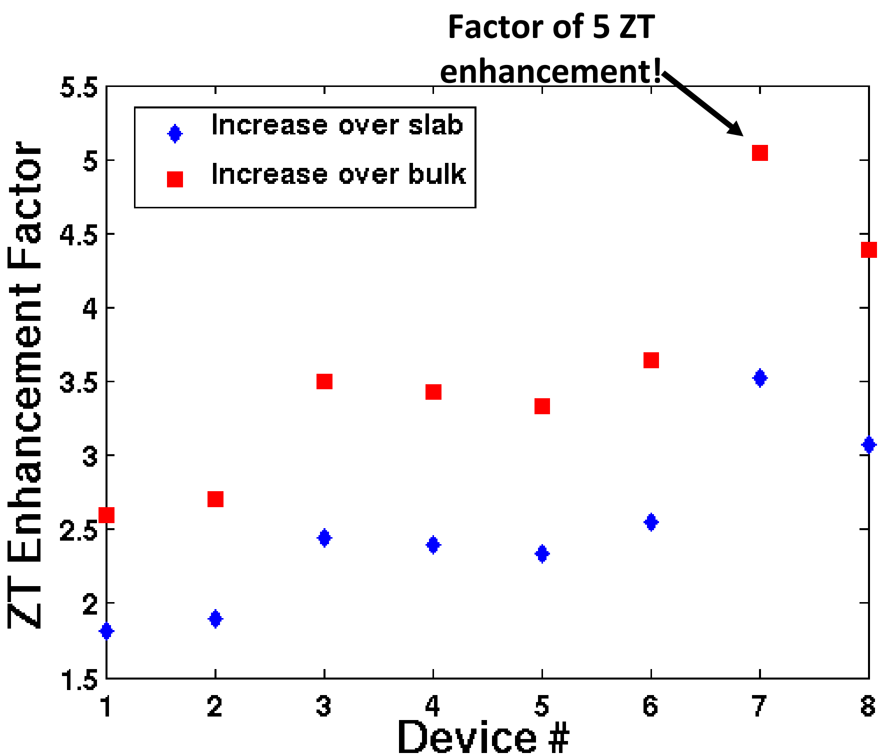


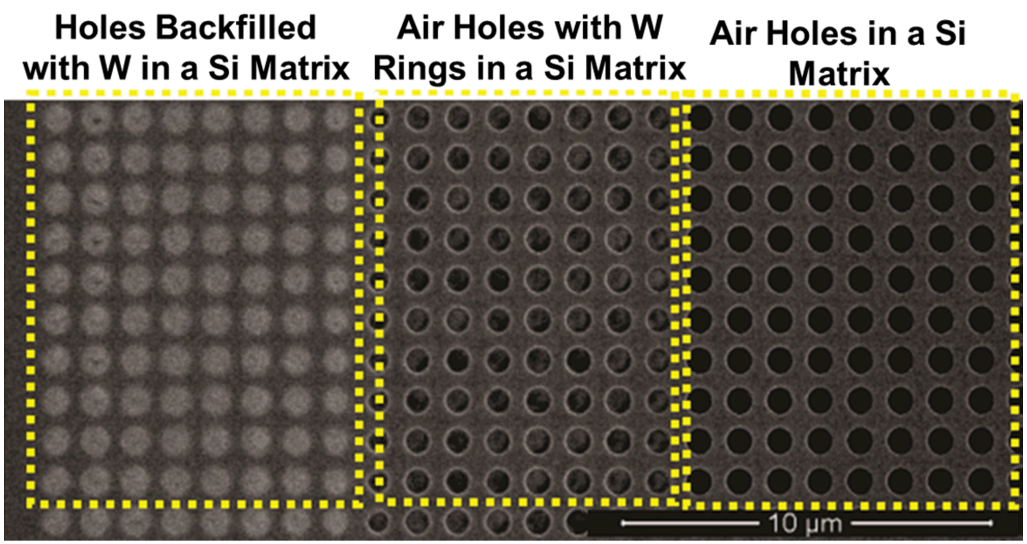
ZT Prediction

Si-W PnC Performance

Concurrent σ Enhancement and κ Reduction:

- Assuming Seebeck coefficient remains constant → 3.5x ZT enhancement compared to an unpatterned slab (~5x compared to bulk)
- Si-metal-air PnC may enhance ZT even more by reducing κ further





Conclusions

- We have shown the first experimental observation of decoupling of electrical and thermal conductivity in PnCs, and theoretical concurrent engineering of the conductivities in a Si-metal PnC for a factor of 5 enhancement of ZT.
- We also introduced the concept of a threshold mean free path in conjunction with a hybrid model, representing one of the most accurate models for thermal transport in periodic micro-scale devices.
- Our approach enables us to use the thermal conductivity as a macroscopic metric for inferring the average phonon coherence length in our PnC samples.
- This work may hold the key to unprecedented thermoelectric performance; using PnC topologies to further suppress phonon transport of microscale porous samples while enhancing electron transport.