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Non-linear Thermal Resistance Trend with Increasing Bilayer Density

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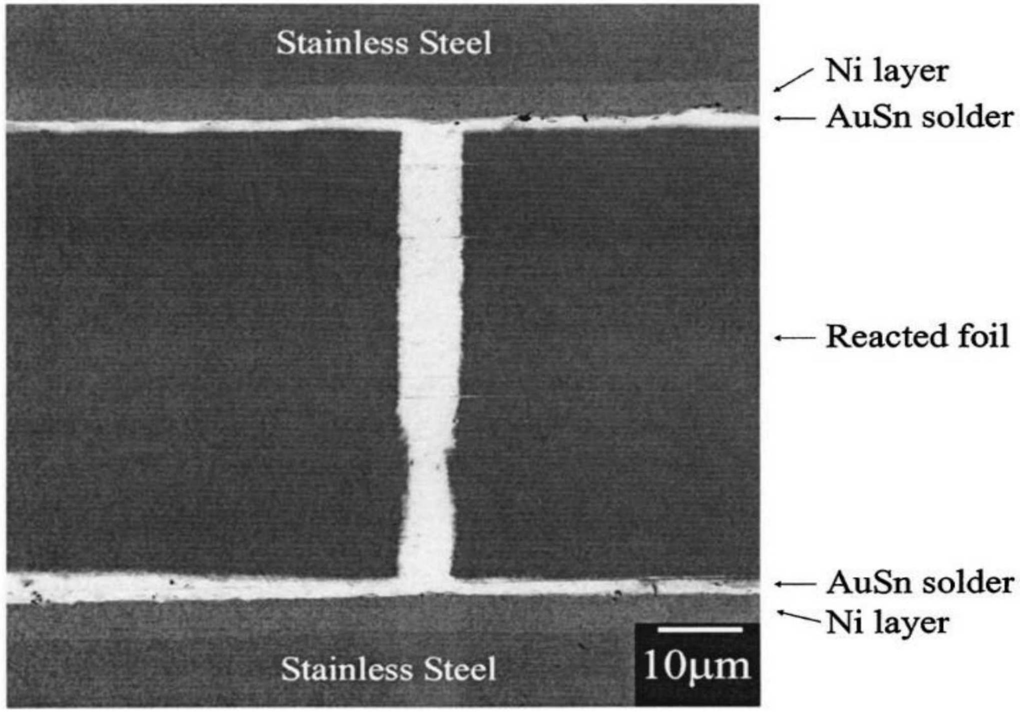
Reactive metal multilayers (RMM)



SEM micrograph of stainless steel components joined using reactive Al/Ni foils and AuSn solder [3]

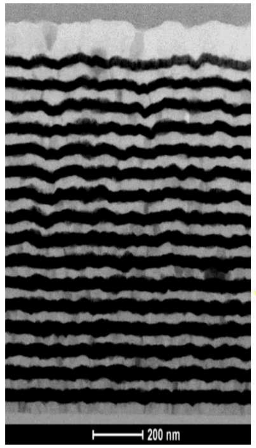
<u>Potential applications</u>

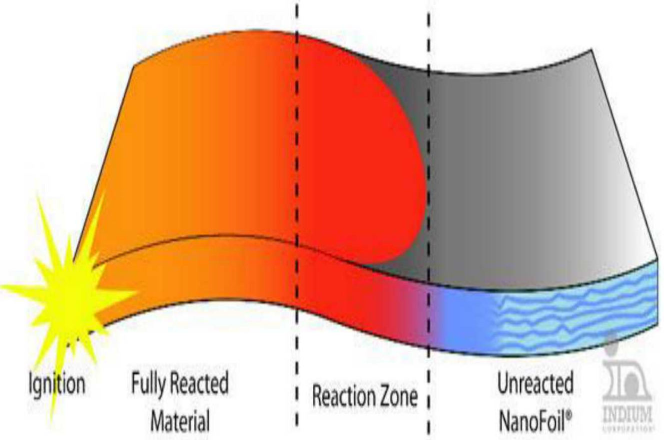
- Heat source for thermal battery [1]
- Lead-free soldering [2]
- Lead-free explosive primers [3]
- Metallic glass welding [4]

[5] EmpaChannel, https://www.youtube.com/watch?v=1ti4JJR3piQ

[1] J. L. Prentice United States Patent No. 4,158,084, 1979
[2] J. Wang et al., Applied Physics Letters, 2003
[3] K. T. Higa, Journal of Propulsion and Power, 2007
[4] A. J. Swiston et al Acta Materialia, 2005

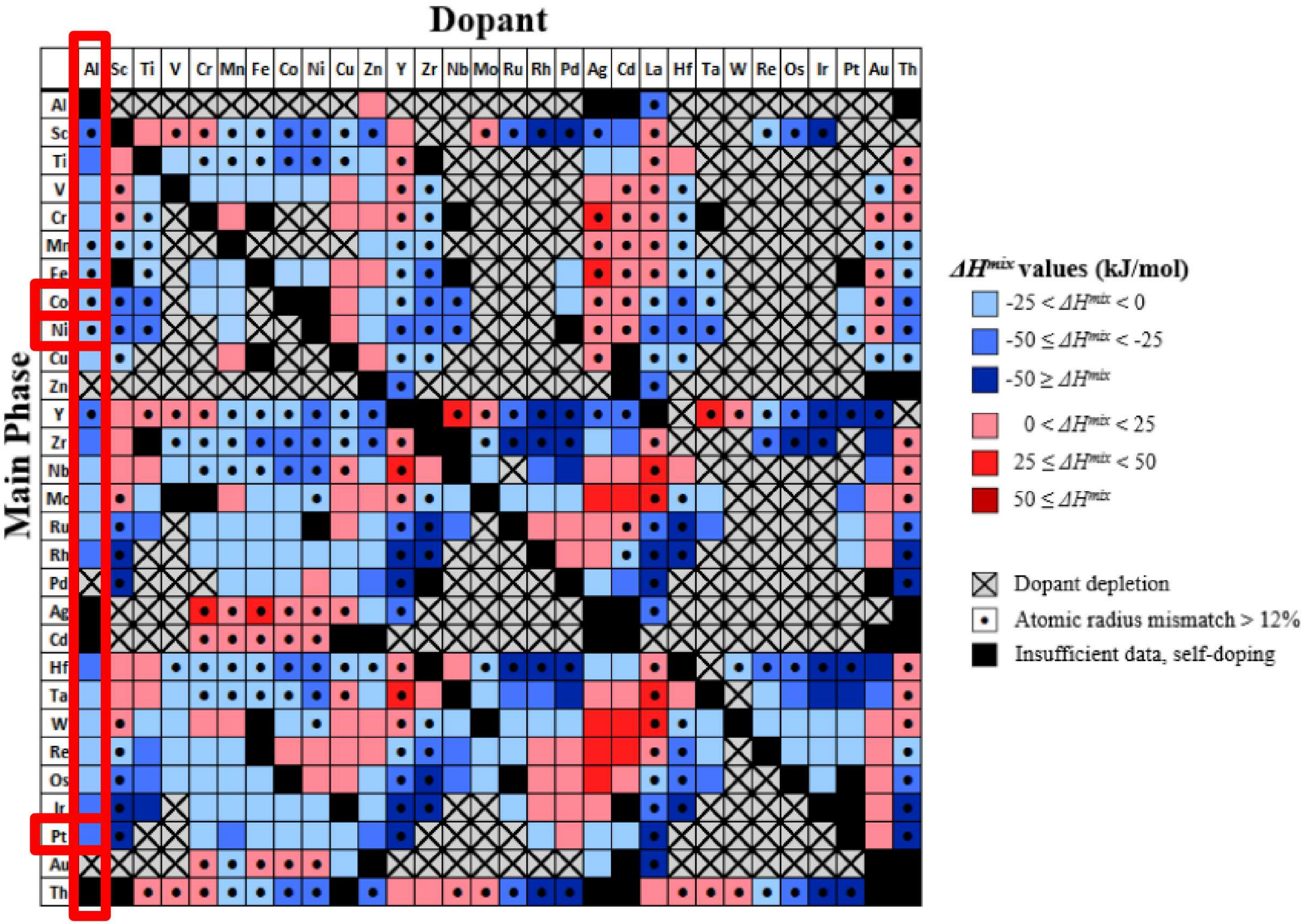
Material selection for mixing reactions





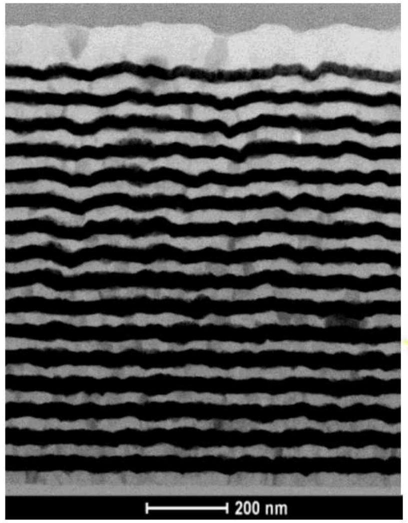
Material selection limitations:

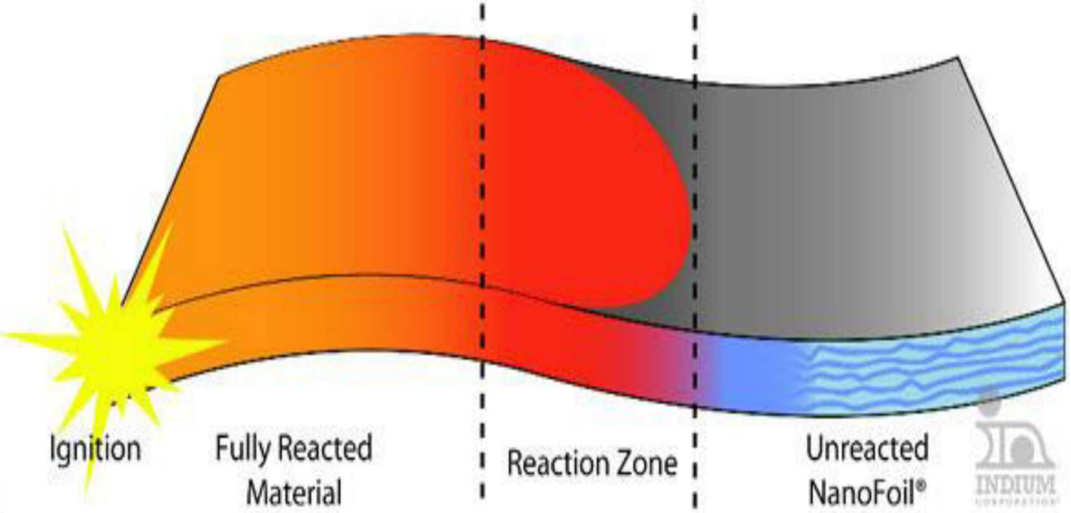
- Adiabatic rxn must exceed melting point of product
- Thermal conductivity must be mitigated
- Activation energy barriers

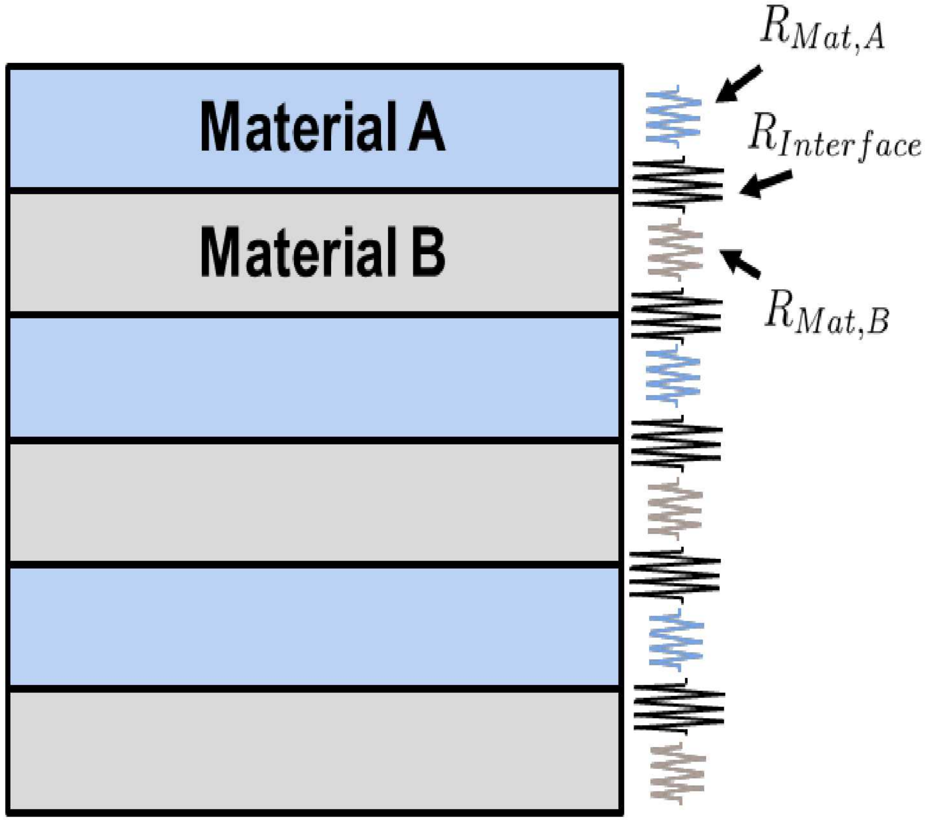


[6] Schuler & Rupert. *Acta Materiala*. 2017

Series lumped resistor model



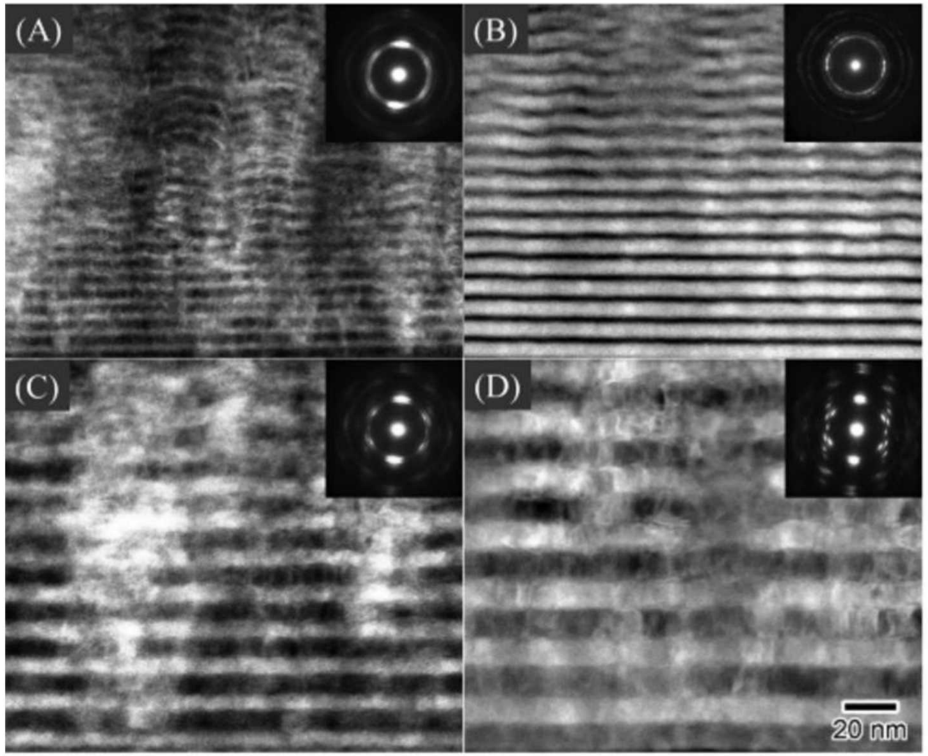


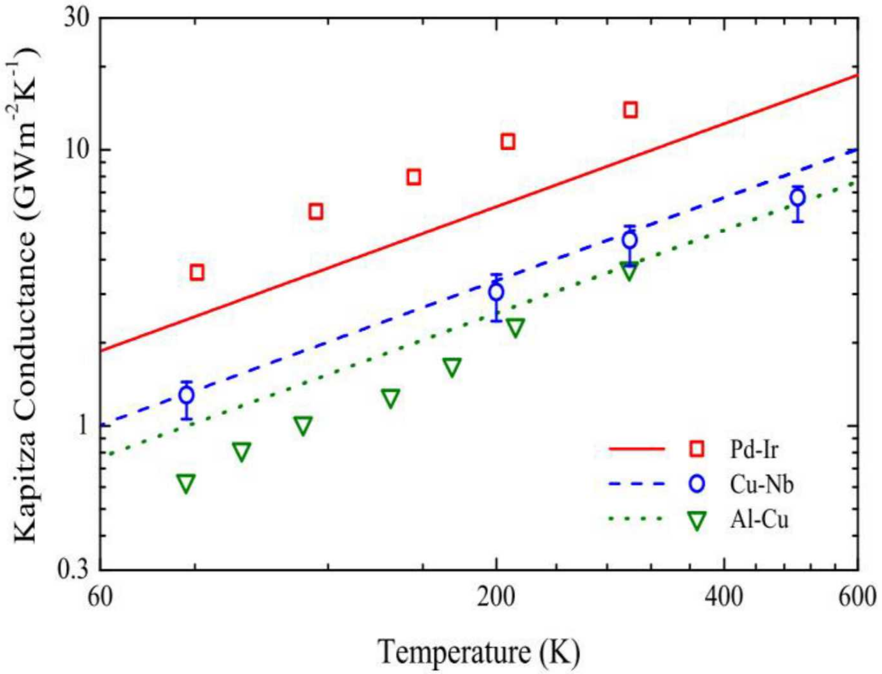


$$R_{Tot} = R_{Mat,A} + R_{Mat,B} + nR_{Interface}$$

$$\zeta_{1\to 2} = \frac{D(\varepsilon_{F,2})\nu_{F,2}}{D(\varepsilon_{F,2})\nu_{F,2} + D(\varepsilon_{F,1})\nu_{F,1}}$$

Electron diffuse mismatch model - EDMM



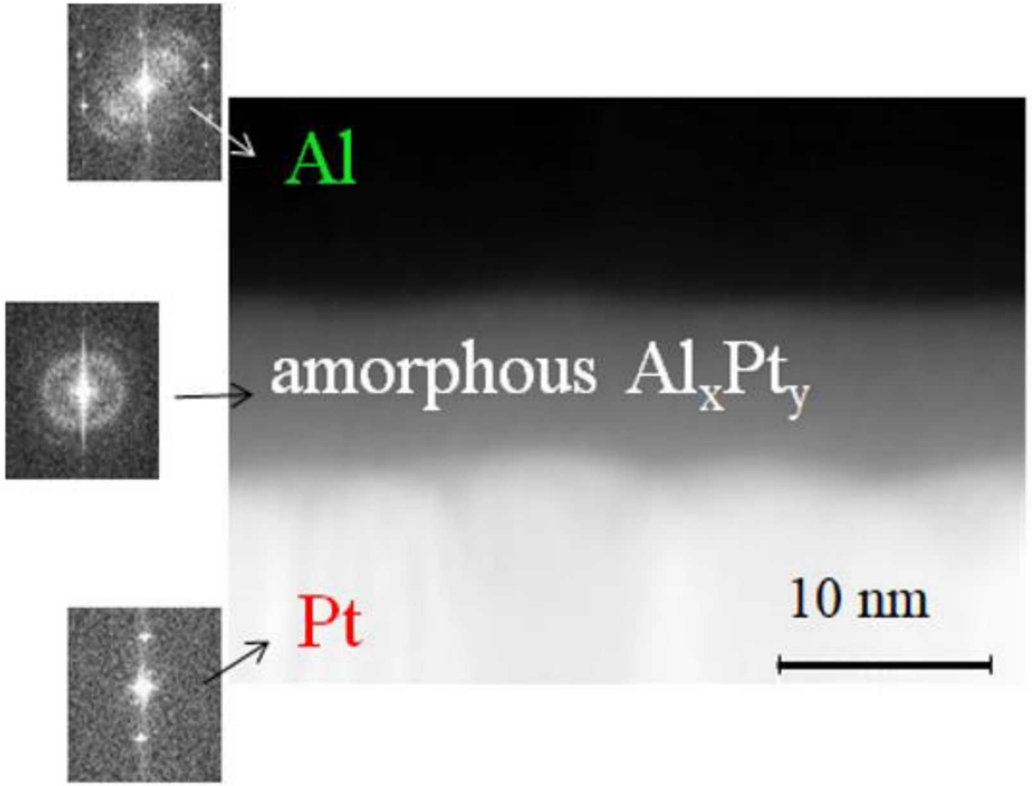


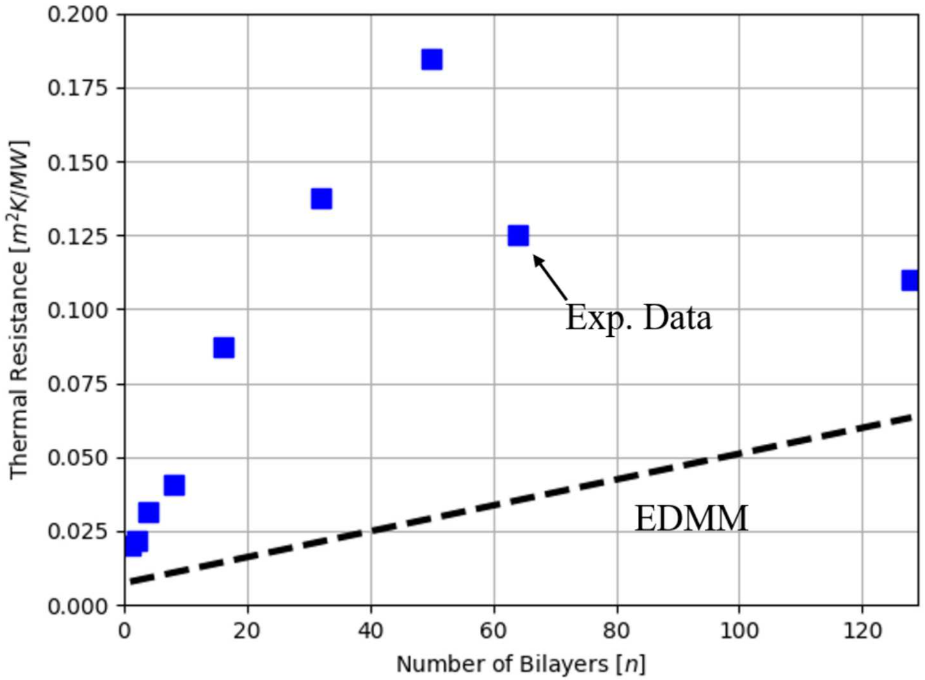
[Left] HAADF STEM images for Cu-Nb multilayers with periodicity lengths (thickness of Cu+Nb) (A) 5.4 ± 0.5 nm (B) 8.4 ± 1.1 nm (C) 12.5 ± 1.4 nm (D) 21.4 ± 2.0 nm [Right] Kapitza conductance data and EDMM calculations given by EDMM for Cu-Nb [6], Al-Cu [7], and Ir-Pd [8] versus temperature

$$R_{tot} = R_0 + \frac{n}{h_{1\to 2}}$$

[7] R. Cheaito et al., Applied Physics Letters, 2015
[8] C. Gundrum et al., Phys. Rev. B, 2005
[9] R. B. Wilson and D. G. Cahill, Phys. Rev. Lett., 2012

Nonlinear trends with increasing bilayer density





[Left] TEM image of a 10 nm Al/Pt interface with diffraction patterns quantifying crystallinity and amorphous regions. [Right] SNL experimental resistance of Al/Pt RMM compared with EDMM approximation

$$R_{Tot} = R_{Mat,A} + R_{Mat,B} + nR_{Interface}$$

[10] D. P. Adams, Thin Solid Films 2015.

First principles calculations - TranSIESTA

<u>Previous work modeling amorphous transport</u>

- Boltzmann transport equation [10]
    - High cost
    - Difficult to parameterize

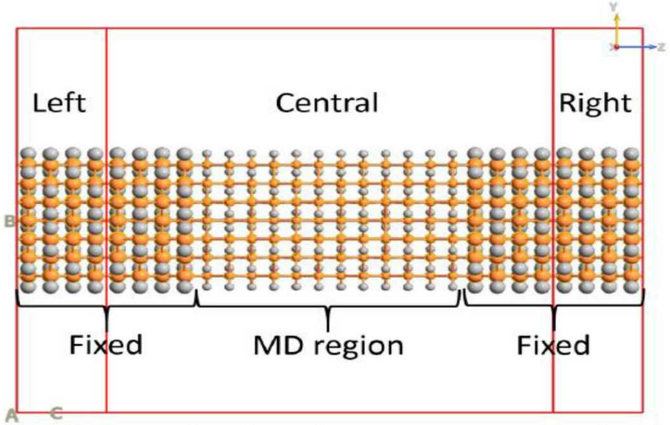
<u>Alternatively</u>

- Length dependent MD-Landauer approach [11]
- Electron-phonon scattering from averaged collective transmission

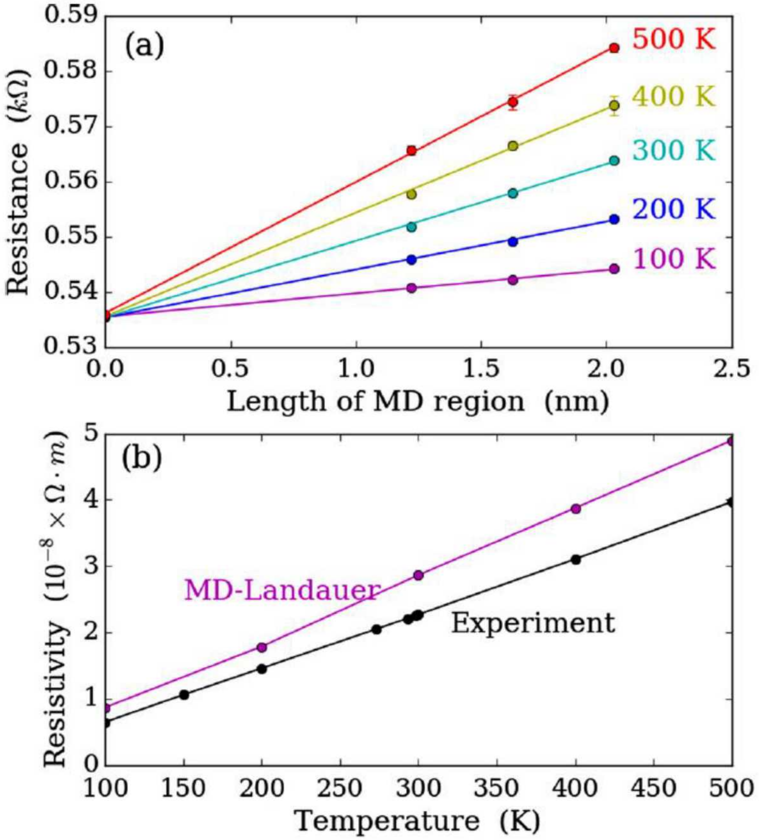
$$G = \frac{1}{R} = G_0 \int T(\varepsilon) \left(-\frac{\partial f}{\partial \varepsilon} \right) d\varepsilon$$

Electronic conductance

$$AR = \rho L$$



MD-Landauer device for transport calculations



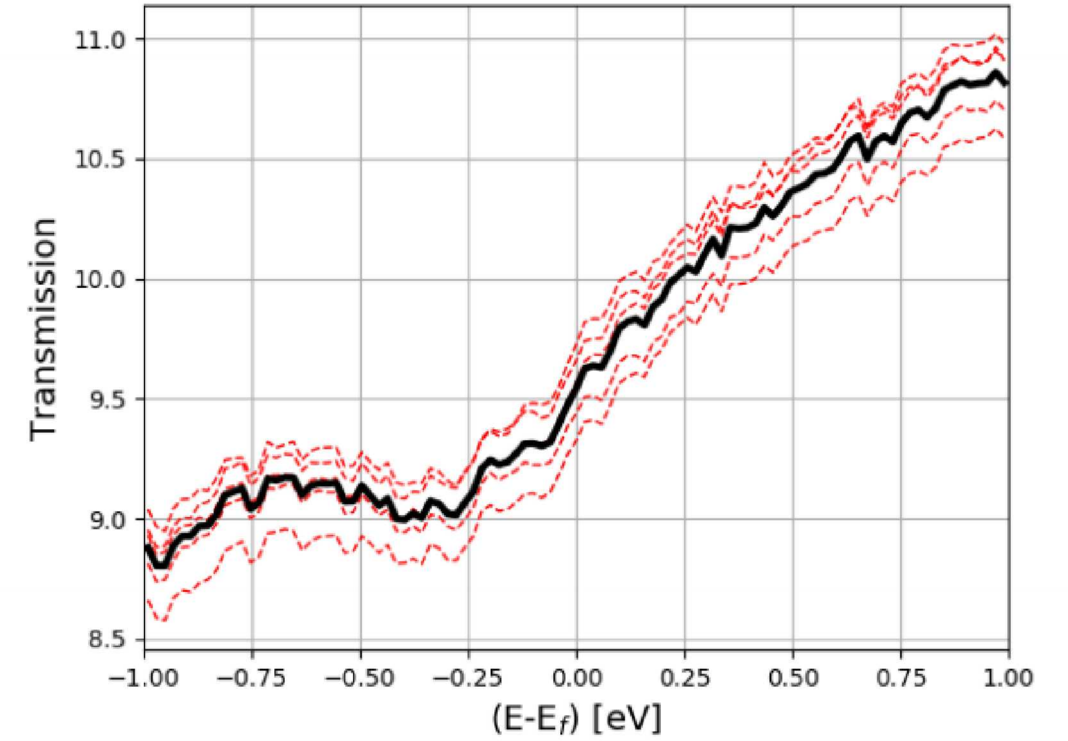
MD-Landauer calculations for bulk Au devices. Resistivity taken from slope of R vs. L.

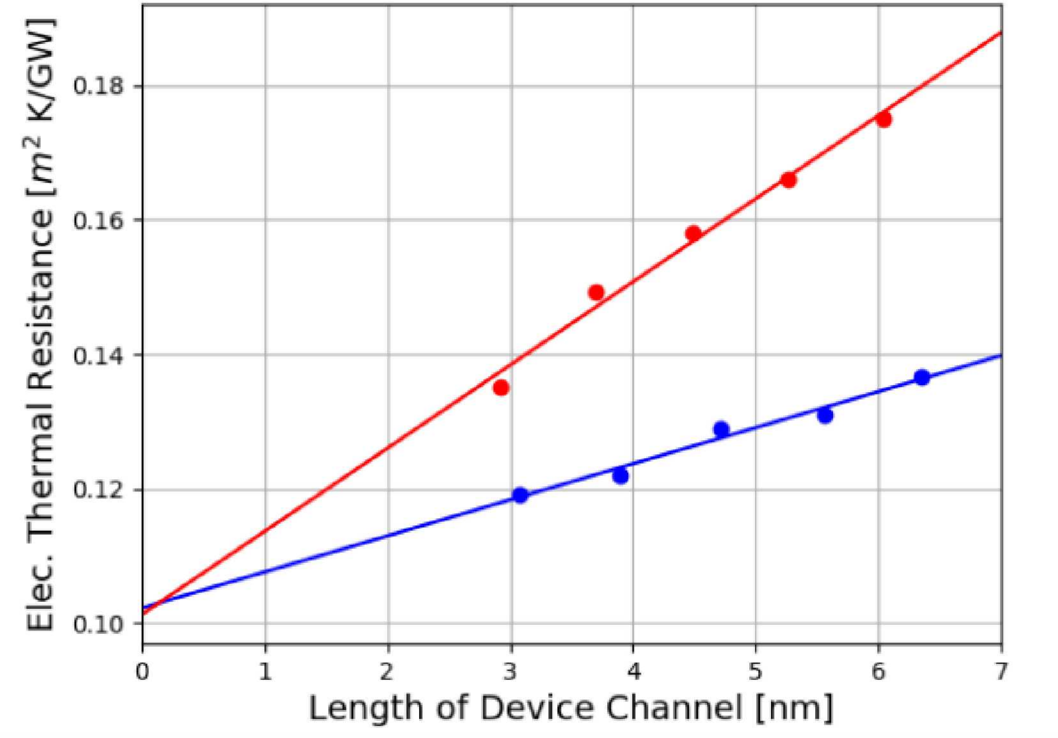
[10] C. H. Park et al., Nano Letters, 2014

[11] T. Markussen et al., Physical Review B, 2017

First principles calculations: MD-Landauer

$$G = \frac{1}{R} = G_0 \int T(\varepsilon) \left(-\frac{\partial f}{\partial \varepsilon} \right) d\varepsilon$$

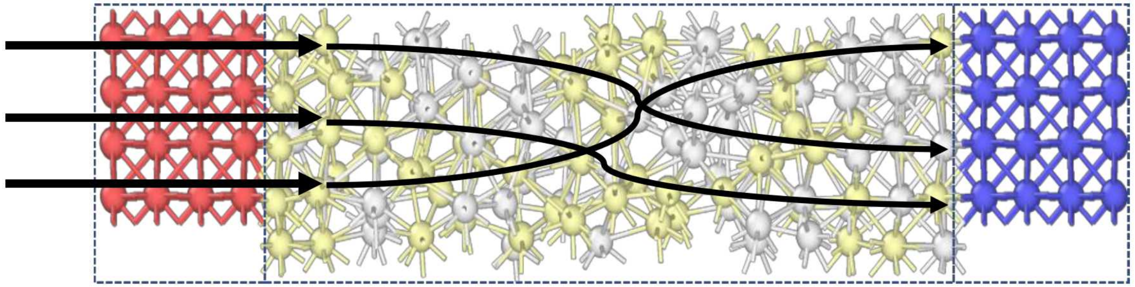


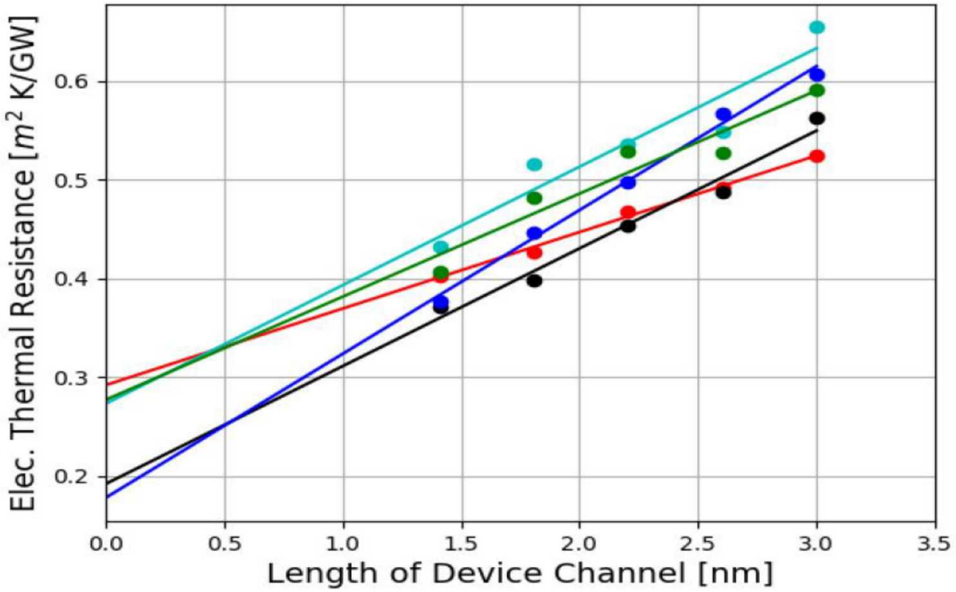


[Left] Crystalline Al transmission spectra at 300K. N = 5.

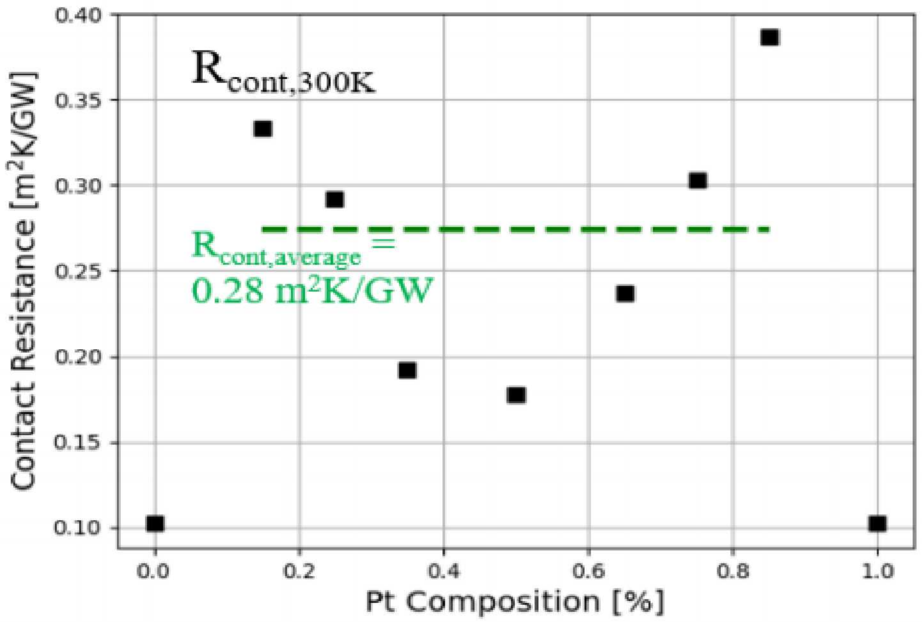
[Right] Length dependent resistance for Al (blue) and Pt (red) at 300 K. Calculated thermal conductivity of Al = 185 W m−1 K−1 and Pt = 76 W m−1 K−1

First principles calculations: MD-Landauer



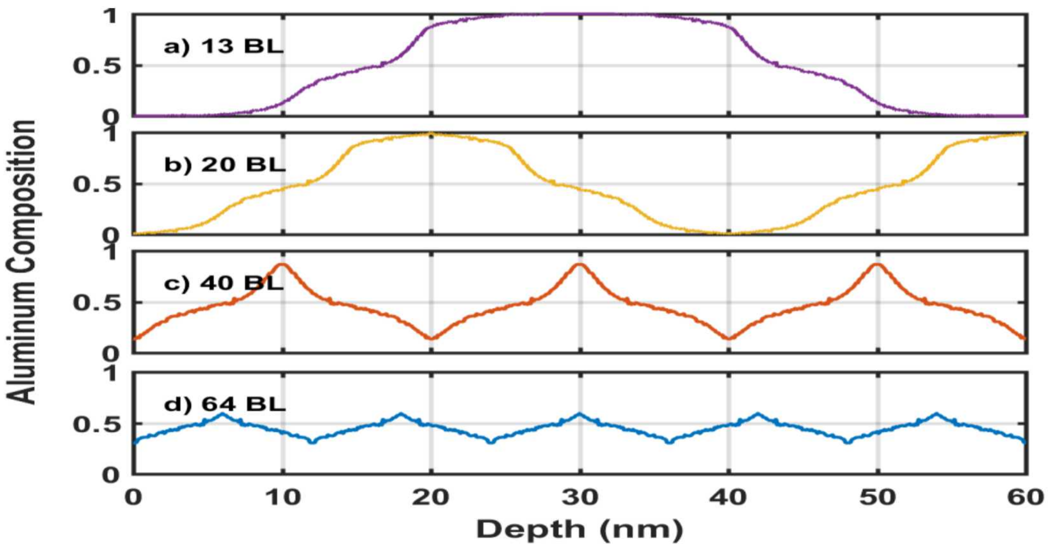


Length dependent device thermal resistance for Al/Pt alloy at 300K. 25% Pt = red, 35% Pt = black, 50 % Pt = blue, 65%, Pt = cyan, 75% Pt = green

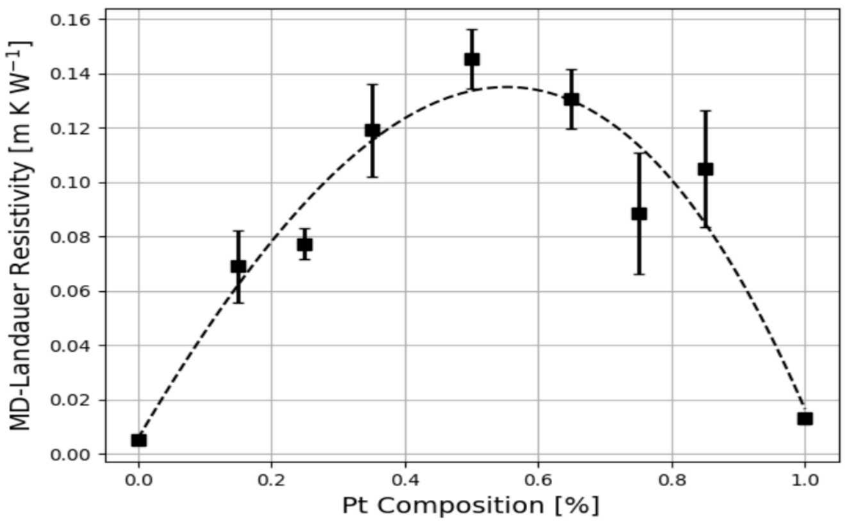


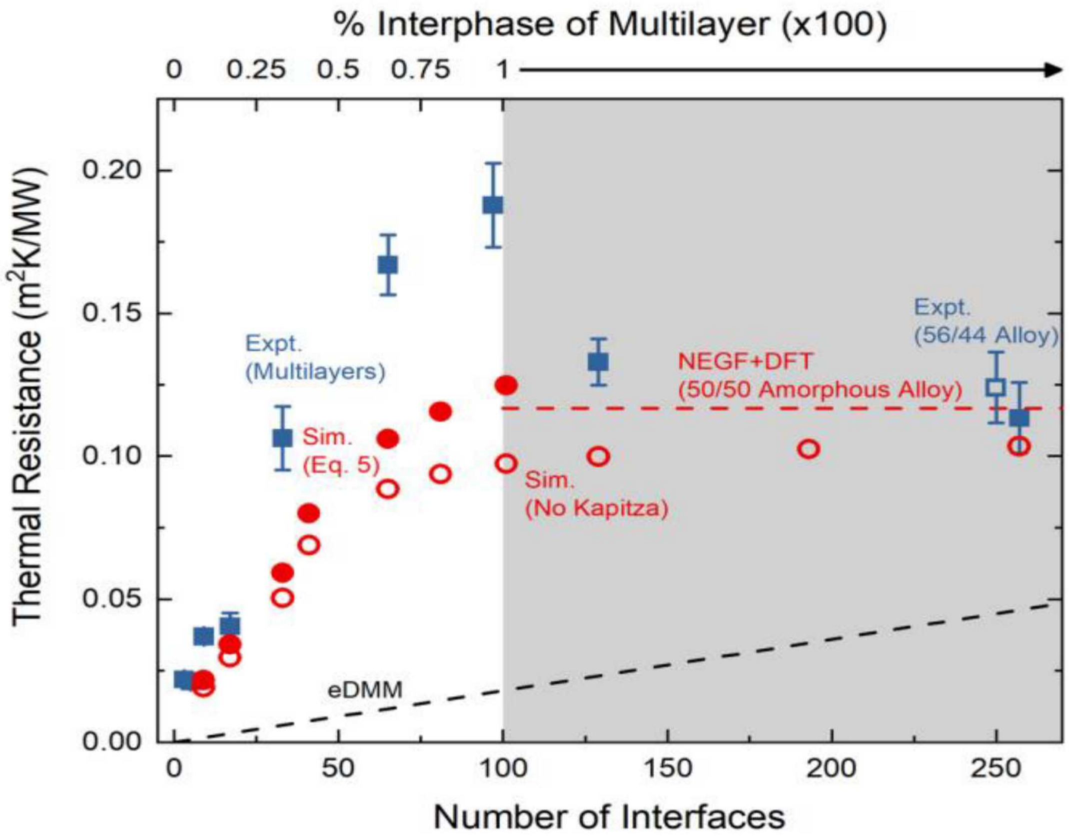
Compositional dependence of contact resistance. End points are fully crystalline Al/Pt devices with identical atom type leads.

Capturing the roll-over effect

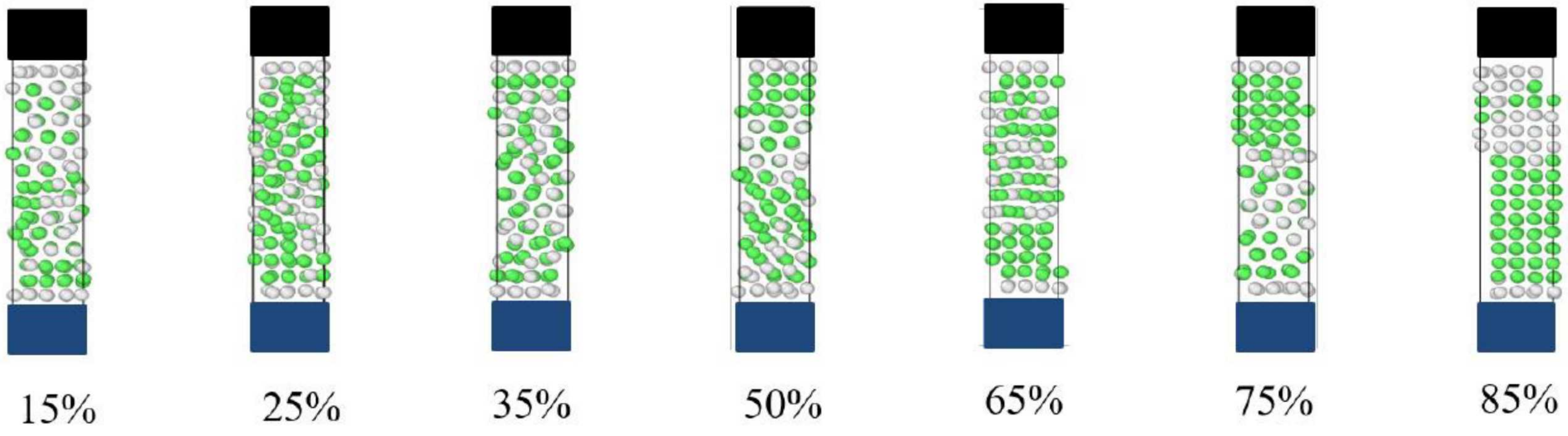


$$R_{BD,NEGF+DFT} = \sum_{i}^{n} \frac{d_i}{\kappa(x_i)} + R_{Kapitza}$$

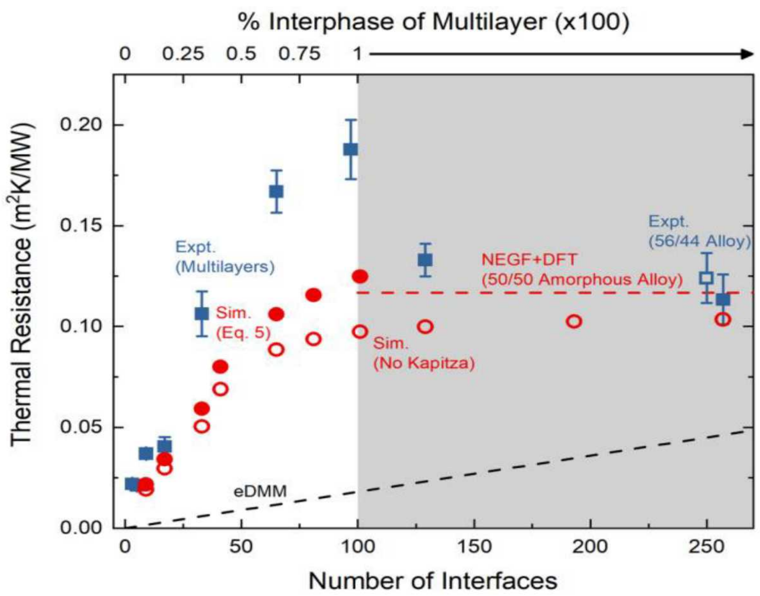


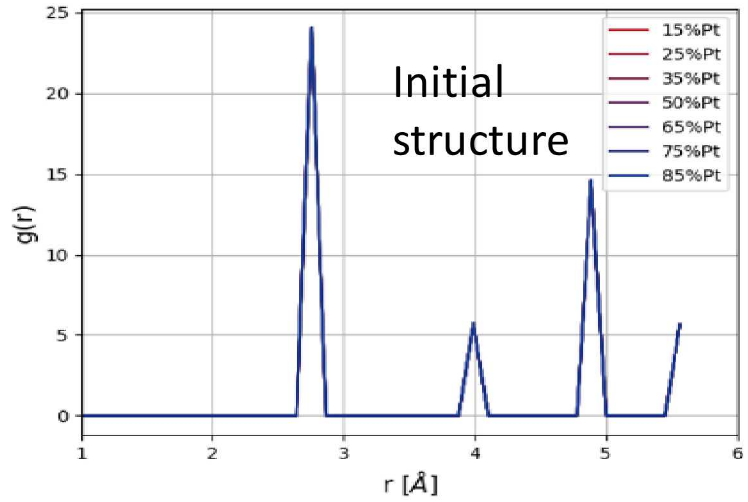


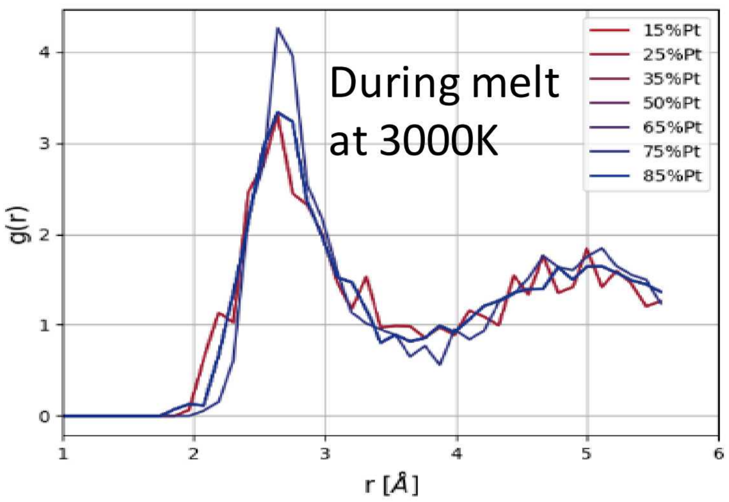
How amorphous is amorphous? Explanations of underestimation

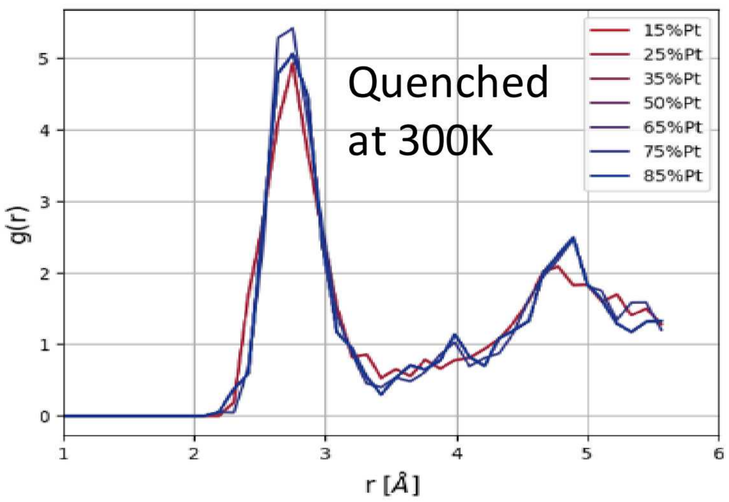


Pt content









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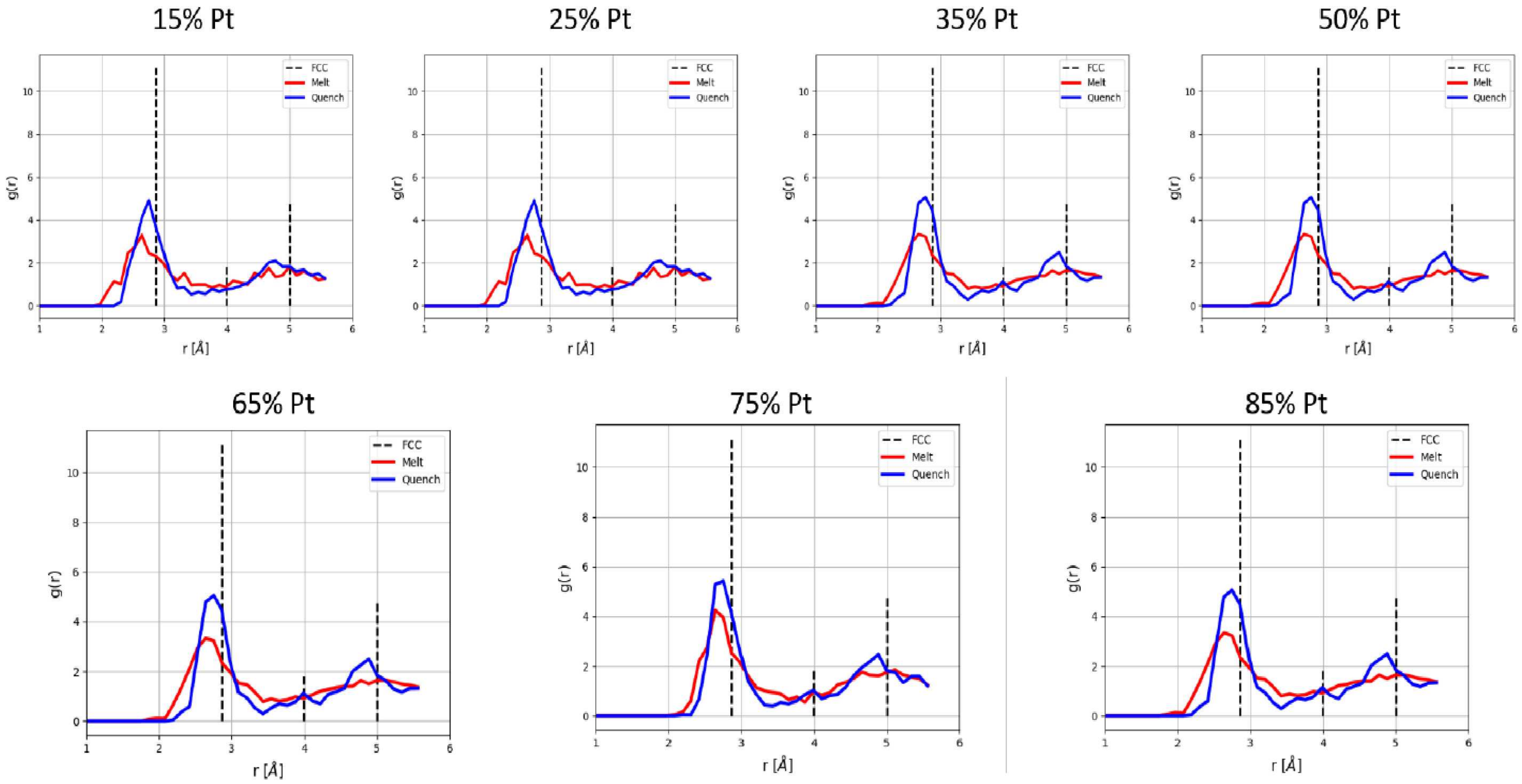

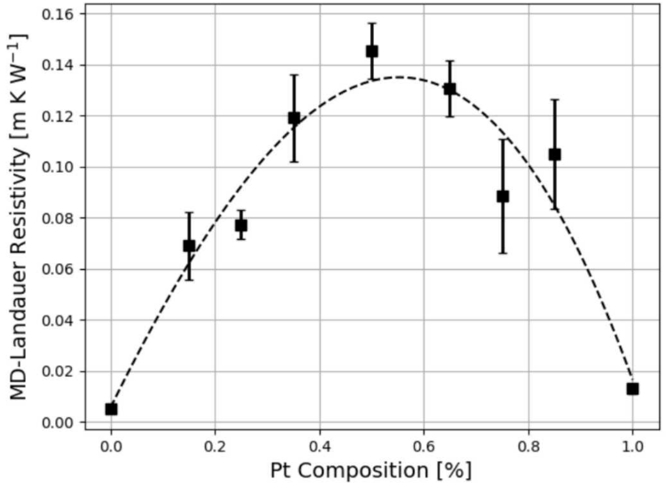

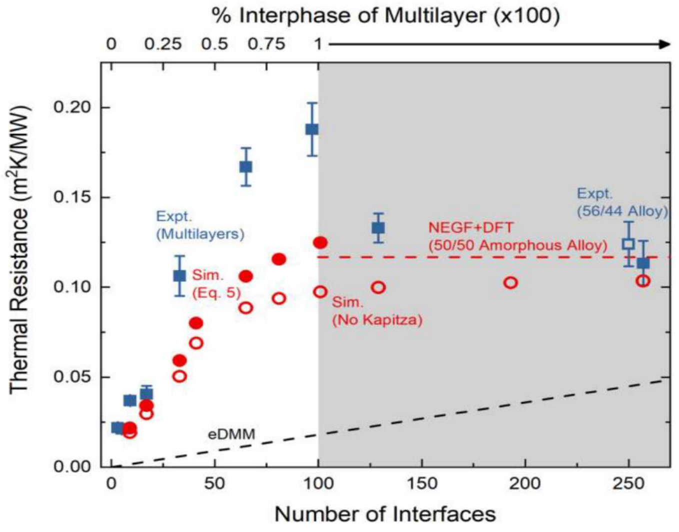

Conclusions and Remarks

- Al/Pt multilayers
  - Excellent heat of mixing
  - Amorphous layer acts at 3rd resistor in series model
  - Critical thickness for maximum resistance
- MD-DFT modeling
  - Accurately predict thermal transport properties of individual amorphous alloy compositions
  - Successfully capture roll-over effects through addition of contact resistance for compositions
  - Complexity of gradients, crystalline phases, and interacting effects not yet captured in model

Acknowledgements

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- Purdue RCAC Computing resources

- Materials Engineering department at Purdue

- Strachan research group (current and prior members)