

# **Thermo-mechanical Property Coupling at Molecularly Tailored Metal-Ceramic Interfaces**

By

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# Abstract

The ability to manipulate interfacial heat transport is highly desired for the thermal management of a number of nanoscale devices and technologies such as nanoelectronics, nanocomposites, energy generation devices, and solid-state lighting. Interfaces become *the* limiting factor to heat transport when feature sizes are on the order of phonon mean free paths. Because thermal management is a limiting factor in the performance and reliability of such devices, a robust method to rationally tune interfacial heat transport without changing the interfacial materials themselves is of great interest.

This thesis investigates a method to tune interfacial thermal conductance,  $G_{\text{int}}$ , at metal-ceramic interfaces by altering the interfacial bond strength through chemical functionalization. A four-fold enhancement in  $G_{\text{int}}$  is realized through strengthened bonding with a nanomolecular layer, and the unique structure of this layer allows its insertion at the interface without encumbering heat flow. The nanomolecular layer's bifunctional chemistry allows fine control over bonding with both of the materials at the interface and therefore a further ability to tune  $G_{\text{int}}$ , with application to multiple materials systems and nanomolecular layers. These results are verified by molecular dynamics simulations which accurately predict the observed interfacial conductance enhancement, demonstrate the importance of bonding between the nanomolecular layer and both interfaces, and indi-

cate that a high phonon density of states in the nanomolecular layer mediates interfacial heat transfer.

Reducing interfacial bond strength accordingly decreases interfacial thermal conductance and expands the range of  $G_{\text{int}}$  tunability to over an order of magnitude, i.e.  $30 \text{ MWm}^{-2}\text{K}^{-1} \leq G_{\text{int}} \leq 430 \text{ MWm}^{-2}\text{K}^{-1}$ . Single layer graphene limits bonding at the interface to van der Waals interactions and produces the lowest values of  $G_{\text{int}}$ . In this regime, poor bonding has such a strong impact on interfacial thermal conductance that  $G_{\text{int}}$  is independent of the type of materials used at the interface. Changes in the strength of van der Waals bonding through chemical functionalization of the substrate or oxidation of the graphene layer produce noticeable changes in interfacial chemistry but do not meaningfully impact the interfacial thermal conductance or interfacial toughness. These results indicate a pathway toward achieving greater control of interfacial thermal transport in a variety of materials systems, with wide application in nanostructured materials and devices.