N000141812724: Enhancing Thermal Transport at Solid-Solid Interfaces

Final Report

Reporting Period: 6/16/2018-10/31/2021

Submitter: Zhiting Tian

Major Goals

The objective of this proposal is to significantly advance the fundamental knowledge of interfacial thermal transport, enabling the rational design of thermal interfaces with ultralow thermal interface resistance for efficient cooling of electronics. The specific objectives are 1. Formulate a comprehensive atomistic Green's function (AGF) model: Detailed information of interface transmittance can unveil the underlying processes that occur at interfaces, which, together with phonon transport processes in bulk materials, will complete the picture of phonon transport. 2. Develop a new measurement approach: To validate the simulation results, we will introduce a new technique for interfacial thermal measurement -- transient thermal grating (TTG) spectroscopy, which probes thermal transport at the interfaces with high sensitivity.

Accomplishments

What were the major goals and objectives of the project? What was accomplished towards achieving these goals? How were the results disseminated to communities of interest?

The five major accomplishments over the three years are:

(1) We developed a rigorous anharmonic AGF formulation to include anharmonicity at interface region for 3D structures using first-principles force constants. The traditional atomistic Green's function (AGF) was formulated in the harmonic regime, preventing it from capturing the role of anharmonicity in interfacial thermal transport. Incorporating anharmonicity into the AGF had long been desired but remains challenging. And this was the very *first* time that the AGF was applied to a 3D anharmonic interface over the past two decades. More specifically, we developed a Fourier decomposition method for third-order tensors and introduced the three-phonon scattering at solid-solid interfaces via the many-body self-energy in the reciprocal space without approximation. Without any fitting parameters, we can obtain the thermal interface conductance using first-principles force constants as the only input. By establishing mass-mismatched silicon-/aluminum-based interfaces, we demonstrated how the anharmonicity at the interface activates phonons that were previously blocked in the harmonic regime, and hence the interface conductance would be enhanced. By depicting the interface heat flow over the phonon frequency range, we exhaustively revealed how the phonons from the high-frequency range scattered into phonons with a lower frequency to flow across the mass-mismatched interface. With the help of phonon density of states curves, we explained such conductance enhancement was driven by the phonon density of states overlapping percentage. Our 3D anharmonic AGF formulation can be employed for all types of 3D interfaces. It provides new insights into engineering interfacial thermal transport to achieve

ultrahigh thermal conductance. This work has been published in Physical Review B rapid communication. We also presented this work at ASME Summer Heat Transfer Conference in 2019.

(2) We made one breakthrough in observing phonon Anderson localization. Anderson localization is a ubiquitous wave effect and has been directly observed in light, ultracold atoms, and ultrasound. Phonon Anderson localization has been receiving increasing interest to achieve ultralow thermal conductivity. For high-thermal-conductivity applications, Anderson localization should be intentionally avoided. Either way, a fundamental understanding of phonon Anderson localization is needed. However, due to challenges to resolve and track the mode-specific transmission, the existence of phonon Anderson localization has only been inferred from the decay of overall thermal conductivity versus device length. Using normal mode decomposition within the AGF framework and carefully designing the systems, we were finally able to provide the *first direct* evidence of thermal phonon Anderson localization using the hallmark of Anderson localization -- the exponential decay of their mode-resolved transmissions. We unambiguously captured localized modes in different structures, even with an upward thermal conductivity trend. In other words, we demonstrate that the macroscopic criterion based on overall thermal conductivity is overly strict to identify the phonon Anderson localization. The mode-level analysis also uncovers a lot of otherwise-buried information, including mode-resolved localization length, onset frequency for the localization-dominant region, etc. Contrary to the commonly held spectral picture, the localization length at a given frequency has widespread, and there is not a clean frequency cutoff between localized and delocalized modes. Our proof of phonon Anderson localization, as well as the new knowledge enabled by the detailed mode-level analysis, are crucial to transform our understanding of thermal transport in highly disordered materials or interfaces and to enable a new paradigm to control heat flow for thermal applications where Anderson localization is strategically leveraged to minimize thermal conductivity or intentionally avoided to maximize thermal conductivity. We have one paper published on Physical Review B.

(3) Quantum thermal rectification at 3D interfaces. A thermal rectifier gives direction-dependent heat flow. In analog to the electronic diodes, heat flows preferentially in one direction in a thermal rectifier. Different mechanisms have been proposed for thermal rectification effects in the classical regime. Using our rigorous 3D anharmonic AGF formulation developed in this project, we discovered a new thermal rectification mechanism in the quantum regime for 3D solid interfaces, which is a universal phenomenon that can be applied to different types of interfaces. Our detailed atomic calculations revealed that the anharmonic scattering across the interface acts on the asymmetric phonon population on both sides of the interface and generates the necessary spatial asymmetry and nonlinearity to achieve thermal rectification. Furthermore, we explored how such thermal rectification ratio would change for different types of interfaces and anharmonicity strength. We obtained general guidance for identifying interfaces with a strong thermal rectification effect and the underlying physical mechanisms for such rectification effect. The manuscript was about to be submitted.

(4) Directly probe thermal transport across interfaces using transient thermal grating. We utilized our thermal transient grating (TTG) systems with the reflection geometry to develop a new interface thermal transport measurement technique. We tested the system we developed with the

Cu/AuSn/Cu interfaces prepared by the United States Navy Academy. We obtain very promising results for different samples. As we swept our laser beam from the left lead to the right lead, we observed the thermal diffusivity matched with the literature value of Cu at the left lead and then drops across the interface range and then increases back for the other lead. We illustrated the effectiveness of our TTG-based interface thermal transport measurement technique. The manuscript is being prepared.

(5) Machine-learning-assisted interface design for guaranteed thermal conductance enhancement. Enhancing thermal interface conductance plays a crucial role in microelectronics cooling because the continued reduction of the device size and increased power intensity demand more efficient heat dissipation. Although light atom doping and interface roughness were reported to slightly enhance thermal interface conductance in certain cases, they could also reduce the thermal interface conductance depending on the interface configurations. By leveraging Bayesian optimization, we provide the practical guidelines to achieve guaranteed enhancement of thermal interface conductance via a proper selection of dopant mass and/or concentration, regardless of interfacial configurations. For perfect interfaces, dopants whose masses are in between the leads can always give improved thermal interface conductance. For rough interfaces, the maximum conductance is always larger than the perfect interface, but the minimum conductance is generally smaller. One needs to properly control the dopant concentration on a rough interface to assure the enhancement. We demonstrate that the data-driven approach could be a helpful tool for interface design with desired thermal interface conductance. We have one manuscript under review on this work.

In addition, we have completed three collaborative works with the MURI team: (1) Al/sapphire interface. By incorporating first-principles force constants of the leads and interface region into AGF, our calculated thermal conductance as a function of temperature agrees well with experimental data. We published two joint papers on Communications Physics and Physical Review B. (2) Ru/sapphire interface. By incorporating first-principles force constants of the leads and interface region into AGF, we calculated the vibrational density of states and thermal conductance as a function of temperature agrees well with experimental data. (3) Ge/GaAs interface. By incorporating first-principles force constants of the leads and interface region into AGF, our calculated 4-probe thermal conductance matched with their lower limit of interface thermal conductance at 300 K. One joint manuscript is in preparation.

Besides, we have accomplished several other projects: (1) We studied thermal transport across grain boundaries. We found that the perfect grain boundaries do not significantly reduce the thermal conductance, while defective grain boundaries can dramatically reduce the thermal conductance. We published one paper in Frontiers in Physics in 2019. (2) Boron suboxides (B6O) were identified to be a superhard material, and their stiff bonds are expected to give large thermal conductivity. But their complex crystal structures suggest otherwise. Using first-principles calculations, we show that both α- and β-B6O have unusually high lattice thermal conductivities of 284.9 and 207.1 W/(m K), respectively, at room temperature, despite their complex structures. Our detailed phonon analysis attributed the dominant factor of its large thermal conductivity to its strong bond strength. Their large group velocities result from the strong bonding and light atomic

mass, while their large phonon lifetimes can be explained by small anharmonicity and limited scattering phase space. Our results show that materials with complex unit cells like a- and b-B6O can still have high thermal conductivity. The combination of large thermal conductivity and excellent physical hardness makes B6O a promising material for lightweight, multifunctional thermal management applications. This work has been published in Applied Physics Letters and featured as a front cover. (3) We investigated the role of van der Waals (vdW) interactions on thermal transport using molecular dynamics simulations and highlighted the importance of vdW interactions to interfacial thermal transport. We published one paper in Carbon 2019. (4) Graphamid is a two-dimensional (2D) polymer. It was predicted to have superior mechanical properties owing to its strong hydrogen bonds. The knowledge of thermal and electrical transport properties in graphamid, which is critical to its future multifunctional applications, remains unknown. Using molecular dynamics simulations and first-principles calculations, we demonstrate that hydrogen bonds drastically enhance out-of-plane thermal and electrical transport in graphamid. The fundamental knowledge from this study can potentially advance the development of hydrogen-bond-enhanced thermal interface transport. This work has been published in ACS Applied Nano Materials 2020.

What opportunities for training and professional development did the project provide?

1. We wrote a book chapter on "Introduction to atomistic Green's function approach: application to nanoscale phonon transport" and published it in "Nanoscale Energy Transport: Emerging phenomena, methods, and applications".

2. Student Jinghang Dai gave a conference talk on the anharmonic AGF method. J. Dai, R. Hu, Z.T. Tian, “Nanoscale Thermal Transport Across 3-D Solid-Solid Interface Through Anharmonic Green’s Function Approach”, 2019 ASME Summer Heat Transfer Conference, Bellevue, WA, HT2019-3581

What honors or awards were received under this project in this reporting period?

1. PI Tian:

• Air Force Research Lab Summer Faculty Fellowship, 2021
• ASME Fellow, 2019
• President’s Council of Cornell Women (PCCW) Affinito-Stewart Award, 2019
• ACS Polymeric Materials Science and Engineering (PMSE) Young Investigator Award, 2019

2. Students:

• Jinghang Dai, KIC Knight Family Foundation Graduate Fellowship in NanoScience and Technology
• Chen Li, Cornell Graduate School Conference Travel Grant.

Technology Transfer

Nothing to Report.

Participants

Name	Role	Person Months
Chen, Li	Graduate Student (research assistant)	30
Dai, Jinghang	Graduate Student (research assistant)	36
Hu, Renjiu	Graduate Student (research assistant)	24
Ma, Hao	Graduate Student (research assistant)	12
Meyer, Talisi	Graduate Student (research assistant)	6
Medina, Mario	Graduate Student	3
Christiansen-Salameh, Joyce	Graduate Student	3

Products

Journal Articles:

1. H. Ma, H. Babaei, and Z.T. Tian. "The Importance of van der Waals Interactions to Thermal Transport in Graphene-C60 Heterostructures", Carbon 148, 196-203 (2019)

https://doi.org/10.1016/j.carbon.2019.03.076

2. C. Li, and ZT Tian, "Phonon Transmission across Silicon Grain Boundaries by Atomistic Green's Function Method", Frontiers in Physics: Computational Physics 7(3), 1-6 (2019)

https://doi.org/10.3389/fphy.2019.00003

3. J. Dai, and ZT Tian, "Rigorous Formalism of Anharmonic Atomistic Green's Function for Three-dimensional Interfaces", Physical Review B: Rapid Communications 101, 041301(R) (2020)

https://doi.org/10.1103/PhysRevB.101.041301

4. Z. Cheng, Y.R. Koh, H. Ahmad, R. Hu, J. Shi, M. Liao, Y. Wang, T. Bai, R. Li, E. Lee, E. Clinton, C. Matthews, Z. Engel, L. Yates, T. Luo, M. Goorsky, A. Doolittle, Z.T. Tian, P. Hopkins, S. Graham, "Thermal Conductance across Harmonic-Matched Epitaxial Al-Sapphire Heterointerfaces", Communications Physics, 3, 115 (2020)

https://doi.org/10.1038/s42005-020-0383-6

5. Y. R. Koh , J. Shi, B. Wang, R. Hu, H. Ahmad, S. Kerdsongpanya, E. Milosevic, W. A. Doolittle, D. Gall, Z. T. Tian, S. Graham, and P. E. Hopkins, "Thermal Boundary Conductance Across Epitaxial Metal/Sapphire Interfaces", Physical Review B 102, 205304 (2020)

https://doi.org/10.1103/PhysRevB.102.205304

6. H. Ma, C. Li, and ZT Tian, "Hydrogen Bonds Significantly Enhance Out-of-Plane Thermal and Electrical Transport in Graphamid", ACS Applied Nano Materials 3, 11090-11097 (2020)

10.1021/acsanm.0c02261

7. J. Dai, and ZT Tian, "Large Thermal Conductivity of Boron Suboxides Despite Complex Structures", Applied Physics Letters 118, 041901 (2021) (Featured as the front cover)

https://doi.org/10.1063/5.0034906

8. R. Hu, and ZT Tian, "Direct Observation of Phonon Anderson Localization in Si/Ge Aperiodic Superlattices", Physical Review B 103, 045304 (2021)

https://doi.org/10.1103/PhysRevB.103.045304

9. R. J. Warzoha, A. A. Wilson, B. F. Donovan, N. Donmezer, A. Giri , P. E. Hopkins, S. Choi, D. Pahinkar, J. Shi, S. Graham, and ZT Tian, "Applications and Impacts of Nanoscale Thermal Transport in Electronics Packaging", Journal of Electronic Packaging 143(2), 020804 (2021)

https://doi.org/10.1115/1.4049293

Book Chapters:

R. Hu, J. Dai, and ZT Tian, "Introduction to Atomistic Green's Function Approach: Application to Nanoscale Phonon Transport", in Nanoscale Energy Transport: Emerging phenomena, methods and applications by IOP Publishing, Online ISBN: 978-0-7503-1738-2 (2020) (Invited)

REPORT DOCUMENTATION PAGE

PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ORGANIZATION.

1. REPORT DATE	2. REPORT TYPE	3. DATES COVERED	
		START DATE	END DATE
28-01-2022	Final	16-06-2018	31-10-2021

4. TITLE AND SUBTITLE

Enhancing Thermal Transport at Solid-Solid Interfaces

5a. CONTRACT NUMBER	5b. GRANT NUMBER	5c. PROGRAM ELEMENT NUMBER
	N000141812724	
5d. PROJECT NUMBER	**5e. TASK NUMBER**	**5f. WORK UNIT NUMBER**

6. AUTHOR(S)

Tian, Zhiting

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)	8. PERFORMING ORGANIZATION REPORT NUMBER
Cornell University 124 Hoy Rd Ithaca, NY 14853	

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)	11. SPONSOR/MONITOR'S REPORT NUMBER(S)
Office of Naval Research 875 N. Randolph Street Arlington VA 22203-1995	ONR	

12. DISTRIBUTION/AVAILABILITY STATEMENT

Approved for Public Release; Distribution is Unlimited.

13. SUPPLEMENTARY NOTES

14. ABSTRACT

A major challenge in achieving high performance electronics is their thermal management, which is aggravated by poor thermal transport across solid-solid interfaces along the heat dissipation pathway. Developing energy-efficient heat removal technologies is critical to the performance, reliability, and lifetime of electronic devices. Fundamental understanding of interfacial thermal transport at solid-solid interfaces is essential to guiding the rational design of thermal interfaces with ultralow thermal interface resistance, which will mitigate the heat load and enable revolutionary advances in next-generation electronic devices. In this project, we integrated advanced atomistic-level simulation with powerful transient spectroscopy to fundamentally understand and enhance thermal transport at solid-solid interfaces. More specifically, we developed a rigorous formalism to calculate phonon transmission from first-principles with inclusion of anharmonicity. We also developed a new measurement technique to directly probe thermal transport processes directly at the interfaces. Experimental and computational modeling will combine to uncover interfacial thermal transport processes in great detail and guide, facilitating the design of next-generation electronics for DoD applications.

15. SUBJECT TERMS

Thermal interface conductance, Atomistic Green's function, Transient thermal grating

PREVIOUS EDITION IS OBSOLETE.

STANDARD FORM 298 (REV. 5/2020)

Prescribed by ANSI Std. Z39.18

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES
a. REPORT	b. ABSTRACT	C. THIS PAGE		
U	U	U	UU	6

19a. NAME OF RESPONSIBLE PERSON	19b. PHONE NUMBER *(Include area code)*
Zhiting Tian	607-255-0733

INSTRUCTIONS FOR COMPLETING SF 298

1. REPORT DATE.
Full publication date, including day, month, if available. Must cite at least the year and be Year 2000 compliant, e.g. 30-06-1998; xx-06-1998; xx-xx-1998.

2. REPORT TYPE.
State the type of report, such as final, technical, interim, memorandum, master's thesis, progress, quarterly, research, special, group study, etc.

3. DATES COVERED.
Indicate the time during which the work was performed and the report was written.

4. TITLE.
Enter title and subtitle with volume number and part number, if applicable. On classified documents, enter the title classification in parentheses.

5a. CONTRACT NUMBER.
Enter all contract numbers as they appear in the report, e.g. F33615-86-C-5169.

5b. GRANT NUMBER.
Enter all grant numbers as they appear in the report, e.g. AFOSR-82-1234.

5c. PROGRAM ELEMENT NUMBER.
Enter all program element numbers as they appear in the report, e.g. 61101A.

5d. PROJECT NUMBER.
Enter all project numbers as they appear in the report, e.g. 1F665702D1257; ILIR.

5e. TASK NUMBER. Enter all task numbers as they appear in the report, e.g. 05; RF0330201; T4112.

5f. WORK UNIT NUMBER.
Enter all work unit numbers as they appear in the report, e.g. 001; AFAPL30480105.

6. AUTHOR(S). Enter name(s) of person(s) responsible for writing the report, performing the research, or credited with the content of the report. The form of entry is the last name, first name, middle initial, and additional qualifiers separated by commas, e.g. Smith, Richard, J, Jr.

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES). Self-explanatory.

8. PERFORMING ORGANIZATION REPORT NUMBER.
Enter all unique alphanumeric report numbers assigned by the performing organization, e.g. BRL-1234; AFWL-TR-85-4017-Vol-21-PT-2.

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES). Enter the name and address of the organization(s) financially responsible for and monitoring the work.

10. SPONSOR/MONITOR'S ACRONYM(S). Enter, if available, e.g. BRL, ARDEC, NADC.

11. SPONSOR/MONITOR'S REPORT NUMBER(S). Enter report number as assigned by the sponsoring/monitoring agency, if available, e.g. BRL-TR-829; -215.

12. DISTRIBUTION/AVAILABILITY STATEMENT. Use agency-mandated availability statements to indicate the public availability or distribution limitations of the report. If additional limitations/ restrictions or special markings are indicated, follow agency authorization procedures, e.g. RD/ FRD, PROPIN, ITAR, etc. Include copyright information.

13. SUPPLEMENTARY NOTES. Enter information not included elsewhere such as: prepared in cooperation with; translation of; report supersedes; old edition number, etc.

14. ABSTRACT. A brief (approximately 200 words) factual summary of the most significant information.

15. SUBJECT TERMS. Key words or phrases identifying major concepts in the report.

16. SECURITY CLASSIFICATION. Enter security classification in accordance with security classification regulations, e.g. U, C, S, etc. If this form contains classified information, stamp classification level on the top and bottom of this page.

17. LIMITATION OF ABSTRACT. This block must be completed to assign a distribution limitation to the abstract. Enter UU (Unclassified Unlimited) or SAR (Same as Report). An entry in this block is necessary if the abstract is to be limited.