

Introduction

We develop a **multiscale modeling** framework for graphene-copper metal-matrix composites to enable high-performance thermal materials. The model consists of:

- **Non-Equilibrium Molecular Dynamics (NEMD)** simulations to resolve and quantify nanoscale thermal behavior at graphene-copper interfaces.
- **Stochastic upscaling** to capture variability in graphene orientation and spacing within the powder microstructure and bridging nano to millimeter scale.
- **Homogenizing** mesoscale behavior into a continuum model to link nanoscale mechanisms with macroscopic thermal properties.

The outcome enables quantitative design for Additive Manufacturing (AM) of high-performance thermal materials.

Upscaling

Aim: Heat transfer simulations for design of nanocomposites from nanoscale effects to microscale distributions.

Approach:

- NEMD simulations to include the angle-dependent interface conductivity.
- Two-step upscaling: Stochastic Upscaling and Homogenization
- Imaging as input for orientation and distribution probabilities.

Challenge: Capture effects of interface interactions and anisotropy.

Results: Conceptual two-step upscaling approach for graded local structures as input for modeling to optimize part design with AM.

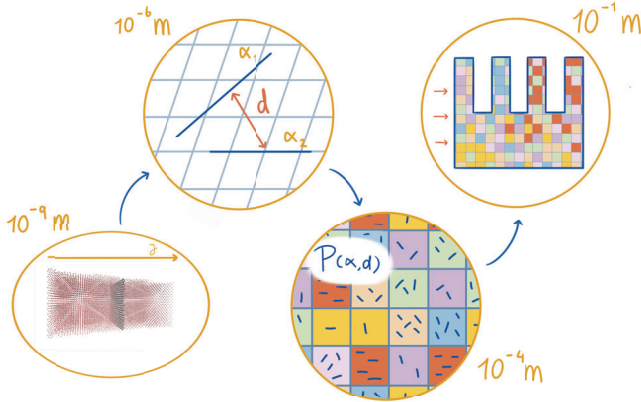


Figure 1. Conceptual sketch of upscaling approach from nanoscale to part properties. On micrometers scales the angle α between the interface and the temperature gradient determines local transport characteristics.

The Material system: Graphene-Copper

- **Enhancing thermal, mechanical and electrical properties** by the addition of a 2D phase.
- High in-plane thermal conductivity of graphene 2000 W/mK to enhance the conductivity of copper, 400 W/mK.
- Composite feedstock powder for **Additive Manufacturing** employing ball-milling.
- **Anisotropic** and multimaterial interfaces require **multiscale simulations**.
- Informed simulations by SEM **imaging** microstructures.

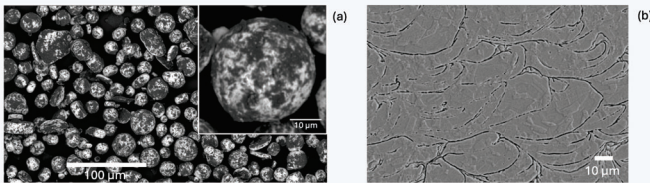


Figure 2. SEM images of Copper-Graphene composite powder (a) and consolidated material (b).

The Nanoscale simulation: NEMD

- **Non-Equilibrium Molecular Dynamics (NEMD)** simulations using the LAMMPS [1] software with force field potentials EAM, AIREBO, and LJ (with mixing rule) for C-C, Cu-Cu, and C-Cu interactions, resp.
- **Temperature gradient** across the domain, maintained with Langevin thermostats.
- **Interface conductance** is calculated after an equilibrium run (1 ns) with:

$$\kappa = \frac{1}{R_k} = \frac{J}{\Delta T} \quad (1)$$

with κ the interface conductance [W/m²K], R_k the Kapitza resistance [m² K/W], ΔT the temperature drop over the interface [K] and J the heat flux [W/m²].

- Linear fits of the temperature in copper regions are used to obtain ΔT and a reliability assessment. Simulation parameters are varied to assess the dependency of the outcome on the simulation setup [2].

Results

Validation NEMD

- Through-plane interface conductance of copper-graphene-copper of $\kappa = 629 \pm 18$ MW/m²K comparable to the literature case, Zhu et al. (2022) [3], of 640.2 MW/m²K.
- **Temperature gradient independence** of conductance in the range $\Delta T \in [50 - 200]$.

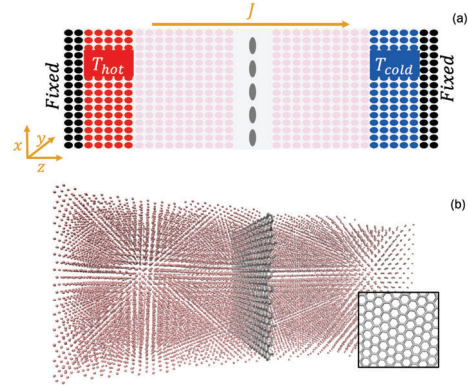


Figure 3. NEMD setup sketch (a) with statistically stationary hot and cold temperatures forcing heat transport across a graphene interface, and VMD visualization [4] of simulation (b).

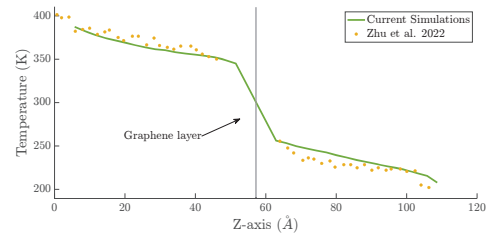


Figure 4. Average temperature profile along the z-axis of the NEMD simulation of $\alpha = 90^\circ$ of current simulations and a literature case.

Angular dependency

- Due to anisotropy of graphene, we investigate angular dependency of average thermal properties.
- Temperature and flux measured locally in a centered volume with high statistical uncertainty.
- Increase in conductivity upon alignment with heat transfer direction.
- Results can be decomposed with $\kappa_{||} \approx 14432$, $\kappa_{\perp} \approx 482$ [MW/m²K] for a finite sheet of 50 Å.

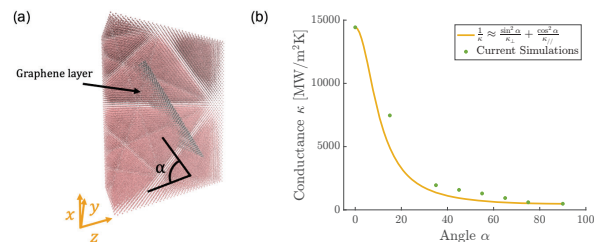


Figure 5. NEMD simulations for single graphene sheet with angle α w.r.t. the thermal gradient direction. Setup visualized with VMD (a) and interface conduction as a function of α (b) for simulation outcomes and the decomposition approximation.

Outlook

- Angular conductivity for varying finite sheet lengths and phonon mode analysis.
- Further completion of the conceptual **multiscale model**.
- Validation with **experimental data**.
- Inclusion **electron transport** calculations and electron-phonon interaction.
- **Uncertainty Quantification (UQ)** NEMD simulations as well as upscaling model for improved reliability.

References

- [1] Thompson, A.P., et al. *Comput. Phys. Commun.* (2022) **271** 108171
- [2] Chen, J. et al. *Rev. Mod. Phys.* (2022) **94** 025002
- [3] Zhu, J. et al. *Materials* (2022) **15** 7588
- [4] Humphrey, W., Dalke, A. and Schulten, K., *J. Molec. Graphics* (1996) **14** 33-38

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