Thermal transfer at crystalline-amorphous interfaces: A Molecular dynamics study

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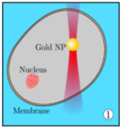
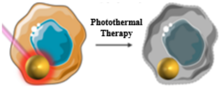
Gold nanoparticles? Bare & Coated



J. Lombard et al.; Phys. Rev. Res. (2021)



S.B Aboeleneen *et al.; Nano Convergence 9*, 37 (2022)

Figure 1: Biological applications for laser heated gold-nanoparticles

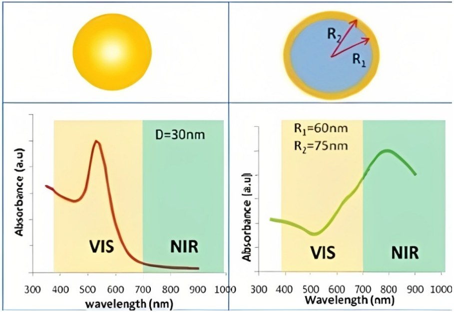
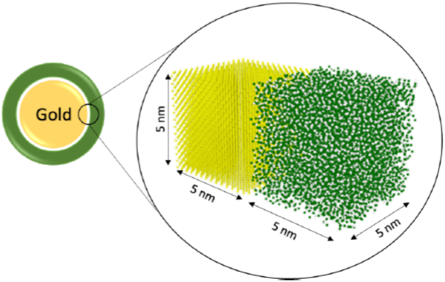


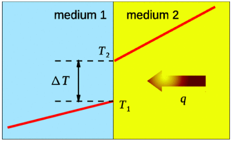
Figure 2: Optical properties of selected gold nanoparticles

Qin and Bischof;Chemical Society Reviews 41(3) (2012)

- **Core-Shell nanoparticles** can absorb radiation in the **near-infrared** region where tissues are **transparent**
- How **heat** is transferred between the **core shell nanoparticle** and **water**? What happens at the interface between the **core** and the **shell**?

Kapitza Resistance: Thermal Resistance at the Interface





- **First step:** Necessity to calculate the different **Interfacial thermal conductance** (ITC) and **resistance** (ITR)
- There are several methods for performing such calculations; one of the most adapted method is: **Molecular Dynamics**

SM Hatam-Lee et al.; Nanoscale and Microscale Thermophysical Engineering (2022)

S. Merabia and K. Termentzidis; Phys. Rev. B 86, 094303 (2012)

Phonons in Crystalline and Amorphous Interfaces

- **Crystalline** interfaces exhibit **significantly lower interfacial thermal conductance (ITC) values of up to 5 times** when compared to their **amorphous** counterparts

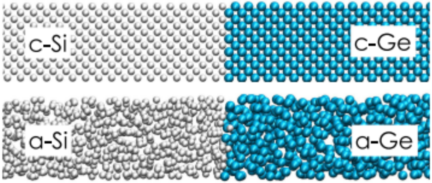


Figure 3: An example of the studied atomic structures of the silicon-germanium interfaces where high ITC is observed in the case of amorphous interfaces

Gordiz and Henry.; J. Appl. Phys. 121 (2): 025102 (2017)

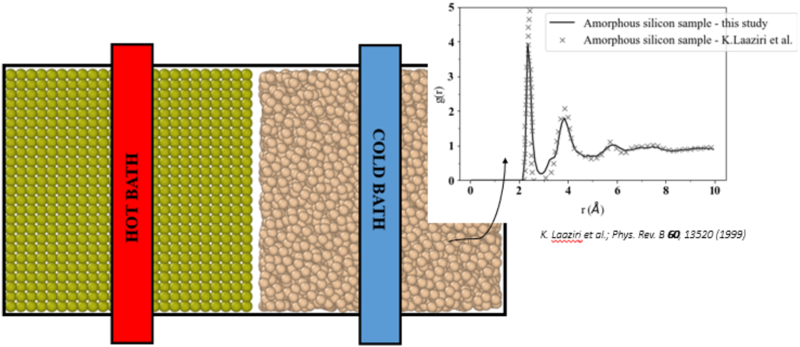
Giri et al; J. Appl. Phys. 118, 165303 (2015)

- Crystalline interfaces → **extended** phonon modes
- Amorphous interfaces → **nonpropagating** phonon modes

- What about **Crystalline - Amorphous** interfaces?

A. France-Lanord et al; J. Phys.: Condens. Matter 26 355801 (2014)

ITC by Non Equilibrium Molecular Dynamics (NEMD)



K. Laaziri et al.; Phys. Rev. B ***60***, *13520 (1999)*

- Interfacial thermal conductance (**G**) calculated as:

$$G = \frac{q}{\Delta T \times A}$$

- **q** is the heat flux
- **ΔT** is the temperature jump at the interface
- **A** is the cross-section area

ITC by NEMD (Cont'd): Example

- Interfacial thermal conductance calculated as:

$$G = \frac{q}{\Delta T \times A}$$

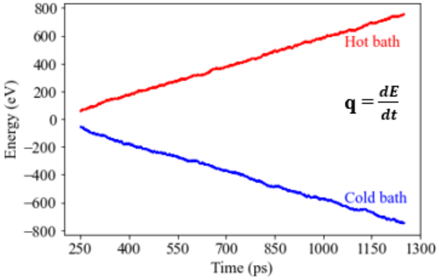


Figure 4: The time-dependent cumulative energy variation of the hot and cold baths

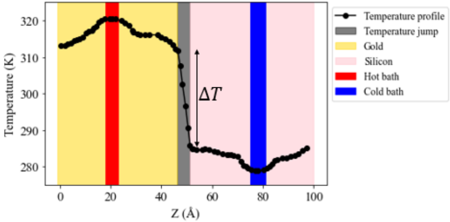


Figure 5: System temperature profile showing the temperature jump at the interface

ITC by NEMD (Cont'd): Results

	ITC (MW/m^2K)	ITR (m^2K/GW)
Gold-Crystalline Silicon	56.61 ± 08	17.85
Gold-Amorphous Silicon	152.17 ± 13	6.57
Gold-Crystalline Silica	102.05 ± 12	9.80
Gold-Amorphous Silica	170.28 ± 14	5.88

- Calculated ITC at the interface of the gold/crystalline-silicon system **agrees** with experimental measurements

  R. Cheaito et al.;Phys. Rev. B 91, 035432 (2015)

- The **interfacial thermal conductance** is ≈ **3 times** higher for **amorphous** silicon/gold interfaces as compared with **crystalline** silicon/gold interfaces

ITC by NEMD (Cont'd): Temperature and Roughness effect

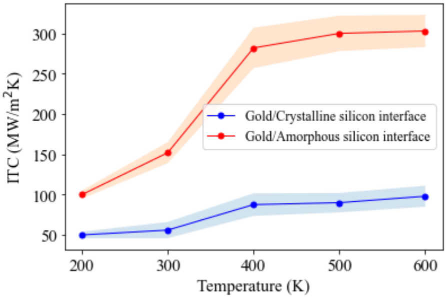


Figure 6: ITC at the gold/crystalline - amorphous silicon interfaces as a function of the system temperature

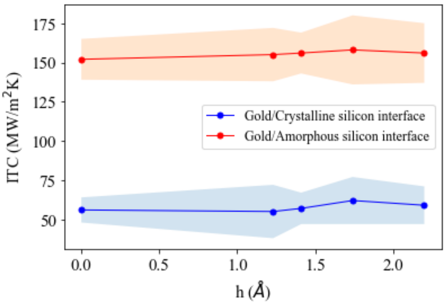


Figure 7: ITC at the gold/crystalline - amorphous silicon interfaces as a function of the height distribution h

ITC by Diffuse Mismatch Model (DMM)

- Phonon–Phonon ITC is given by the **Landauer's formulation:**

Swartz and Pohl; Rev. Mod. Phys. 61, 605 (1989)

$$G_p = \frac{1}{2}\sum_p \int_0^{\omega^c_{Au,p}} v_{Au,p}\hbar\omega D_{Au,p}(\omega)\frac{\partial f_{eq}(\omega, T)}{\partial T}\int_0^{\pi/2}\alpha_p(\omega, \cos\theta)\cos\theta d(\cos\theta)d\omega$$

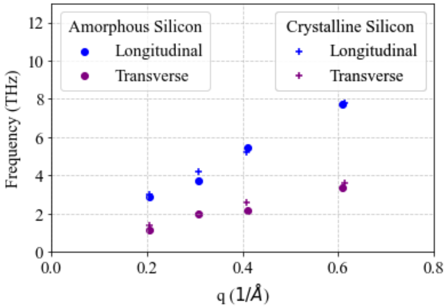


Figure 8: Acoustic dispersion obtained for both silicon structures

ITC (MW/m^2K)	NEMD	DMM
Gold-Crystalline Silicon	56.61	82.7
Gold-Amorphous Silicon	152.17	110

- **Huge differences** in between the results of the two approaches
- **DMM can't** reliably predict results

Phonon Modal Participation Ratio

- The **phonon modal participation ratio** is calculated as:

Donadio. D et al.; Phys. Rev. Lett. 102, 195901 (2009)

$$P(\mathbf{k},\nu) = \frac{1}{N_b \sum_1^{N_b} \left(\sum_\alpha^3 e_{i,\alpha}^*(\mathbf{k},\nu) e_{i,\alpha}(\mathbf{k},\nu) \right)^2}$$

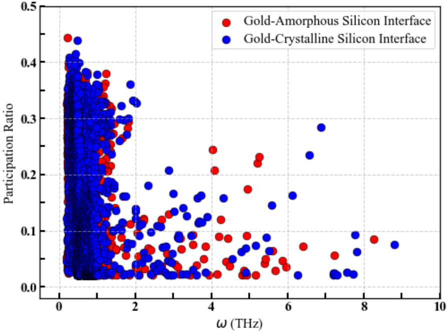


Figure 9: Phonon modal participation ratio at both interfaces

- $P(\mathbf{k},\nu) \leq 0.5 \rightarrow$ localized phonon mode
- $P(\mathbf{k},\nu) > 0.5 \rightarrow$ extended phonon mode

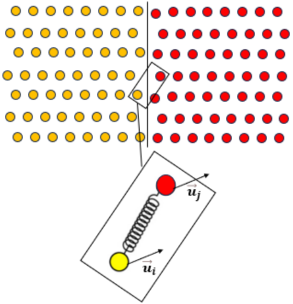
- Phonon localization **does not explain** the difference

How to Explain this Threefold Increase?

- **Calculated average** spring stiffness acting on a gold atom is about **30% greater** at the crystalline-amorphous silicon interfaces

- **Higher cross-correlation coefficient** between **atomic vibrational displacements** at the crystalline-amorphous silicon interfaces which directly influence ITC

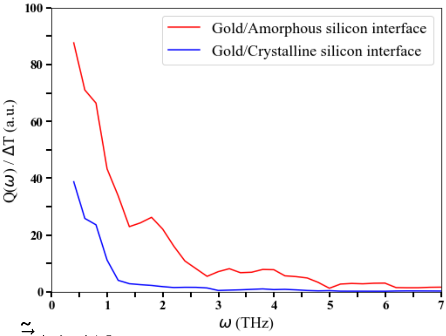
X. Li et al.; Nanoscale,15, 8603-8610 (2023)



- **How can we combine these two quantities?**

Frequency dependent Spring stiffness-atomic displacement Spectrum

- **Q(ω)** encodes the combined information on the **Spring stiffness** and the **atomic displacements cross-correlation**



$$Q(\omega) = \frac{-i\omega}{2}[\sum_{i \in Au} \sum_{j \in Si} \overleftrightarrow{K}_{ij} \langle \tilde{\vec{\Delta}}_{ij}(\omega) \cdot \tilde{\vec{\Delta}}^{*}_{ij}(\omega) \rangle]$$

Where: $\tilde{\vec{\Delta}}_{ij}(\omega) = \tilde{\vec{u}}_i(\omega) - \tilde{\vec{u}}_j(\omega)$; with: $\tilde{\vec{u}}_i(\omega) = \int_0^{t_{max}} \vec{u}_i(\tau) e^{i\omega\tau} \, d\tau$

- **Low frequencies are involved!**

Conclusion

- Characterize the **interfacial thermal resistance** at the different interfaces
- Report a **threefold increase** of interfacial thermal conductance for gold-amorphous interfaces
- Interpret this result by **enhanced bonding** and **atomic displacements cross correlations of low frequency phonon modes** at the interfaces

Thank You!