A122

異なる親和性でのPt/n-アルカン界面の界面熱抵抗と表面液体構造の相関

Surface Liquid Structure Correlation with Interface Thermal Resistance of Pt/*n*-alkane Interface at Different Affinities

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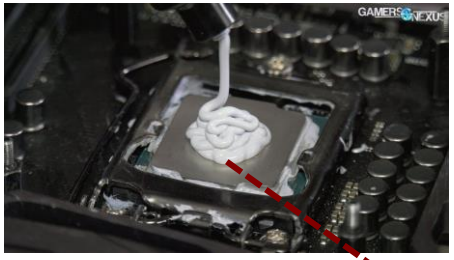
東北大学

Institute of Fluid Science Tohoku University IFS

Background



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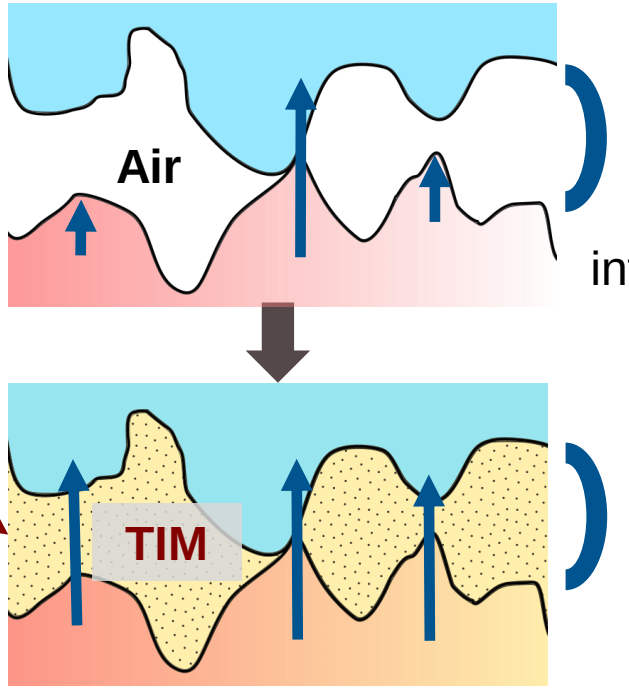
Thermal interface material (TIM) fills the air–gap



ITR is very high

interface thermal resistance

ITR is decreased

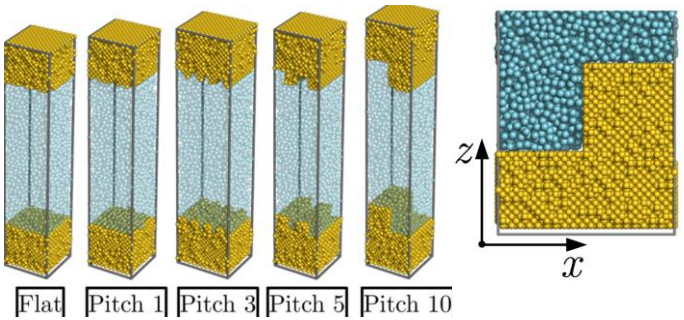
Billions transistors in one chip

- size of semiconductors ↓
- density of heat dissipation ↑
- significance of ITR ↑

→ Understanding thermal transport mechanism at the interface of solid-TIM at nano scale is essential

Existing Research on ITR of SLS System

- work of our group



Surblys D, et al. *J. Chem. Phys.* **150**, 114705 (2019)

Au-Ar-Au system

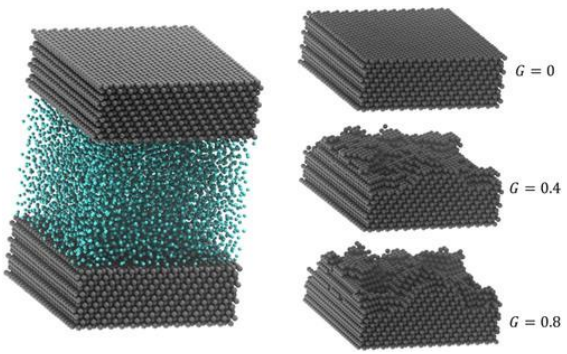
at low affinity: $ITR_{large\ groove} > ITR_{small\ groove}$

at high affinity: $ITR_{rough\ surface} < ITR_{flat\ surface}$

at super high affinity: highly ordered liquids increase ITR

Polyatomic SLS system should be paid more attention. Roughness is an essential parameter on ITR.

- work of others



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Ag-Ar-Ag system

- Same fractal dimension and depth of roughness have same ITR and liquid density profiles.
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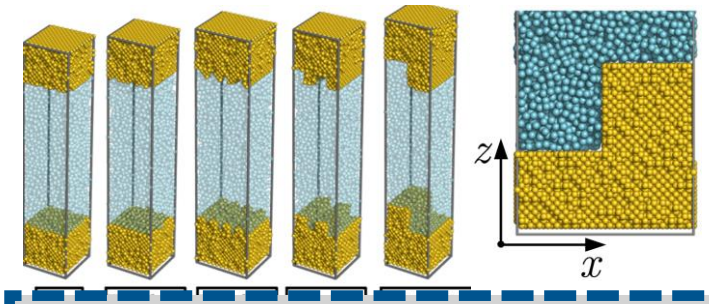
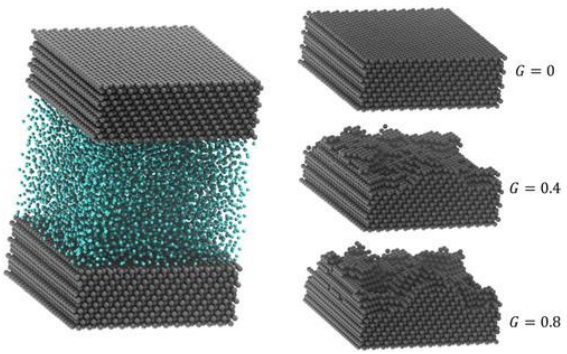


Au-Ar-Au system

Objective:
Investigating surface liquid structure correlation with interface thermal resistance of Pt/*n*-alkane (TIM) interfaces at different affinities

- work of others

Ag-Ar-Ag system



Frank M, et al. *J. Chem. Phys.* **151**, 134705 (2019)

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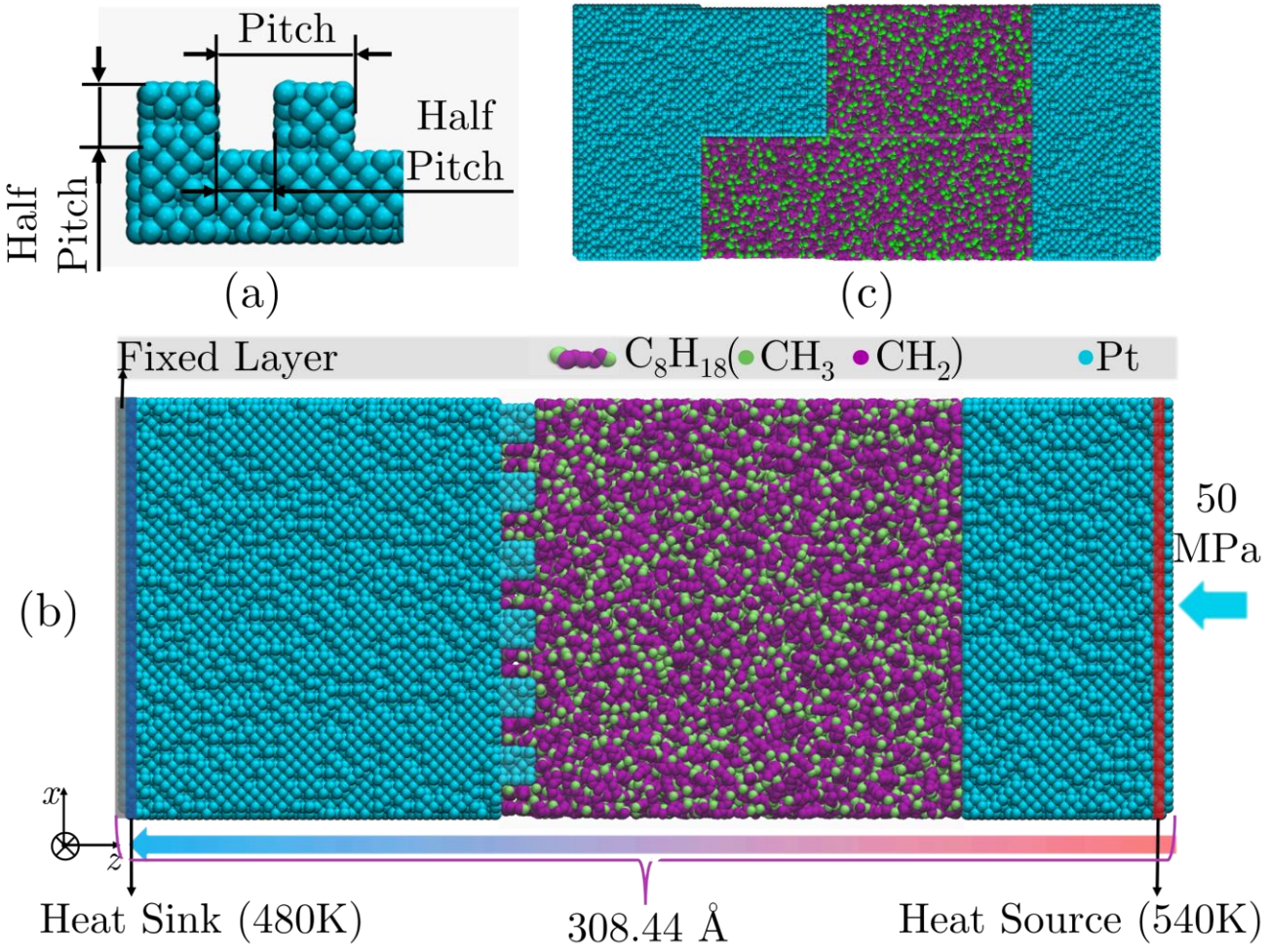
Simulation System



polymer chain-length:

- 8, 16, 24, 36

pitch length:

- 0
- 19.575005 Å
- 117.45003 Å

(a) definition of pitch
(b) small groove system
(c) large groove system

NEMD techniques were applied.

All The Systems:

L_x=L_y=117.45 Å

$N_{\text{united-atom}}$ = 43200

Potential

potential models

- Platinum solid: Morse potential (Pamuk H Ö, et al. Phys. Status Solidi A 37, 1976)
- *n*-alkane: NERD potential (Nath S K, et al. J. Chem. Phys. 108.23, 1998)
- The Lorentz-Berthelot combining rule is adopted to LJ interactions between sites of different types.

NERD potential

$$E_{\text{CH2-CH2/CH3}} = E_{\text{vdW}} + E_{\text{strch}} + E_{\text{bend}} + E_{\text{tor}}$$

$$E_{\text{vdW}} = \sum_{i<j} 4\varepsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{6}\right]$$

$$E_{\text{strch}} = \sum_{\text{strch}} \frac{1}{2} k_r (r_{ij} - r_0)^2$$

$$E_{\text{bend}} = \sum_{\text{bend}} \frac{1}{2} k_\theta (\theta_{ijk} - \theta_0)^2$$

$$E_{\text{tor}}(\phi) = \sum_{\text{tor}} E_1 + E_2\cos(\phi) + E_3\cos^2(\phi) + E_4\cos^3(\phi)$$

Morse potential

$$E_{\text{Pt-Pt}} = \sum_{i<j} D\left[e^{-2\alpha(r_{ij}-r_0)} - 2e^{-\alpha(r_{ij}-r_0)}\right]$$

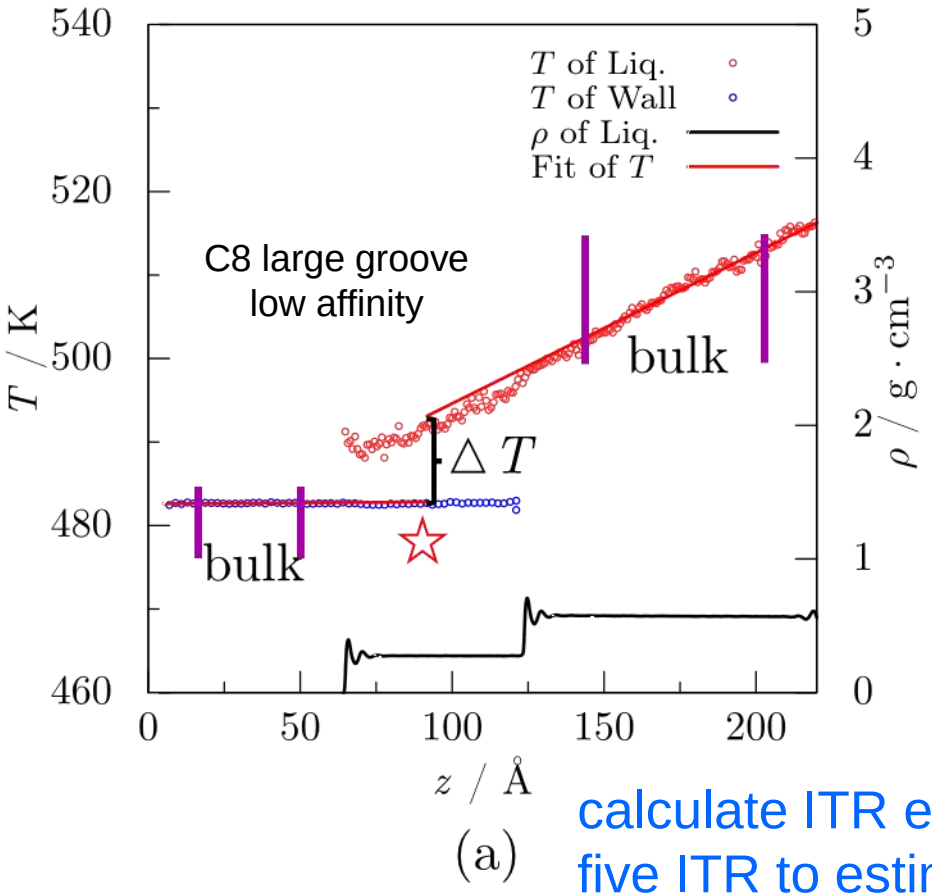
Affinity

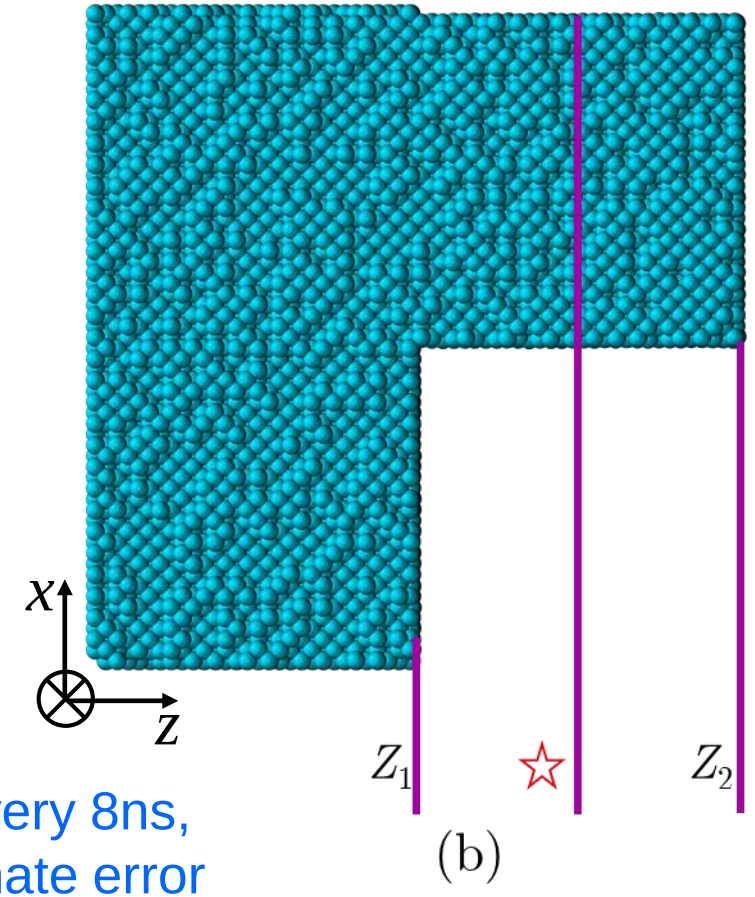
$$E_{\text{Pt-CH2/CH3}} = \sum_{\substack{i\in\text{Pt} \\ j\in\text{CH2/CH3}}} 4\eta\varepsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{6}\right]$$

0.1: low affinity
0.5: high affinity

ITR Calculation

- linear fitting of T
- interface position





calculate ITR every 8ns,
five ITR to estimate error

$$J_z = E_{\text{sink}} / S_{xy} t$$

Fourier's Law:

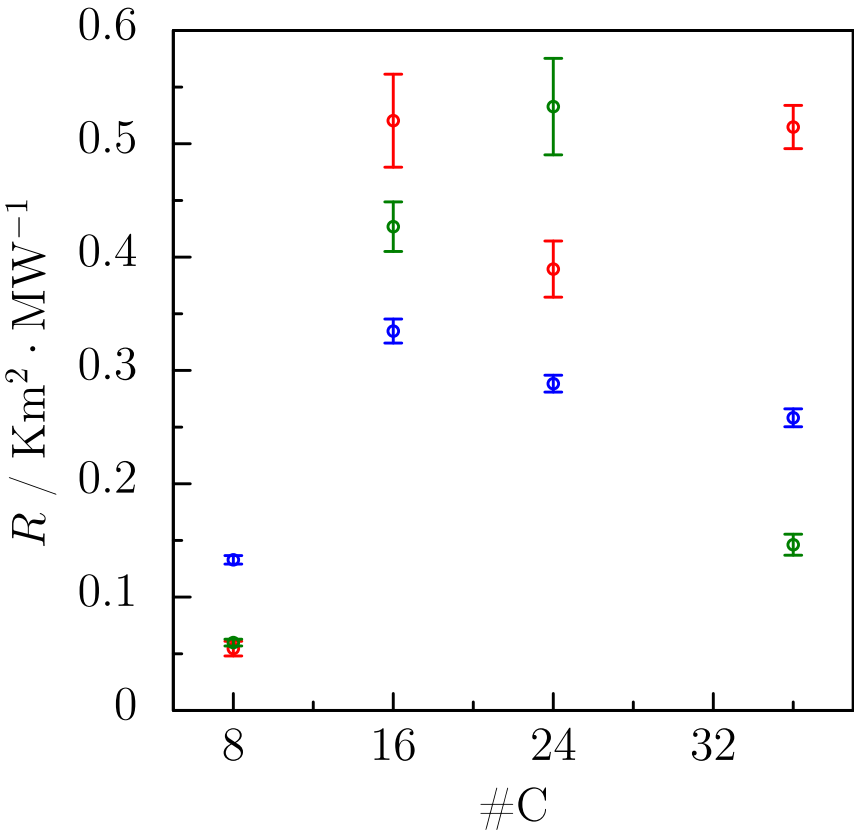
$$\text{ITR} = \Delta T / J_z$$

rough surface: midpoint of rightmost layer position of not grooved section, Z_1, and that of grooved section, Z_2

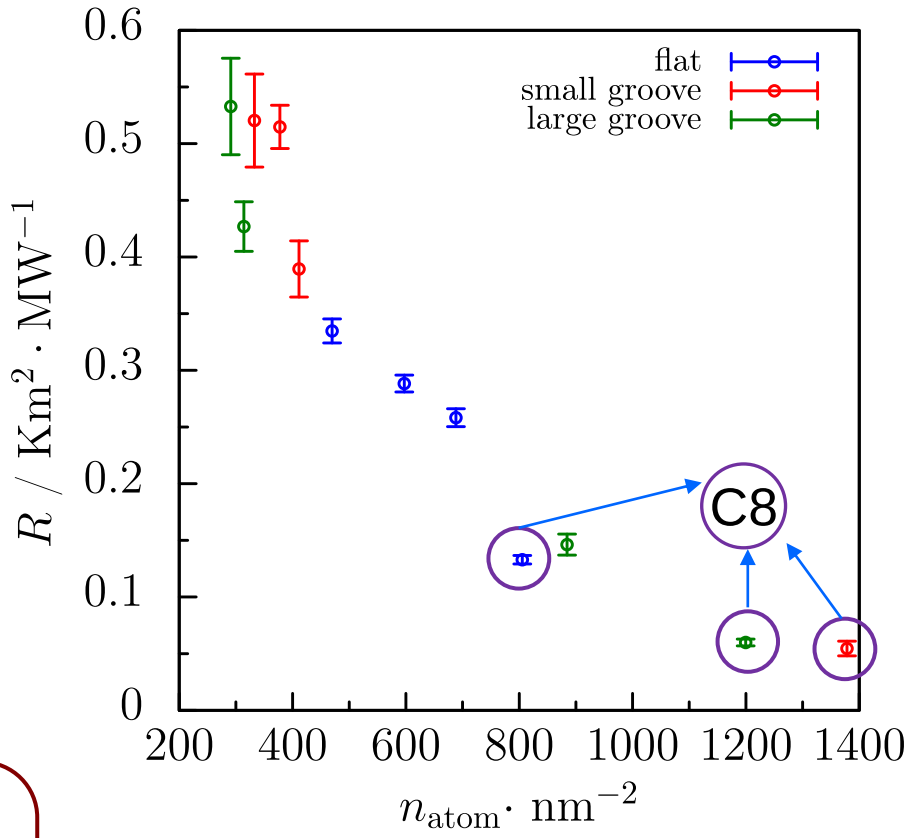
flat surface: middle position of rightmost layer and that of first density peak

Results at low affinity

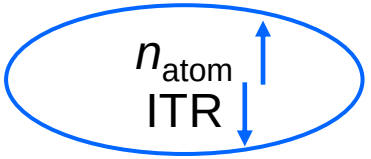
- ITR at different roughness scales



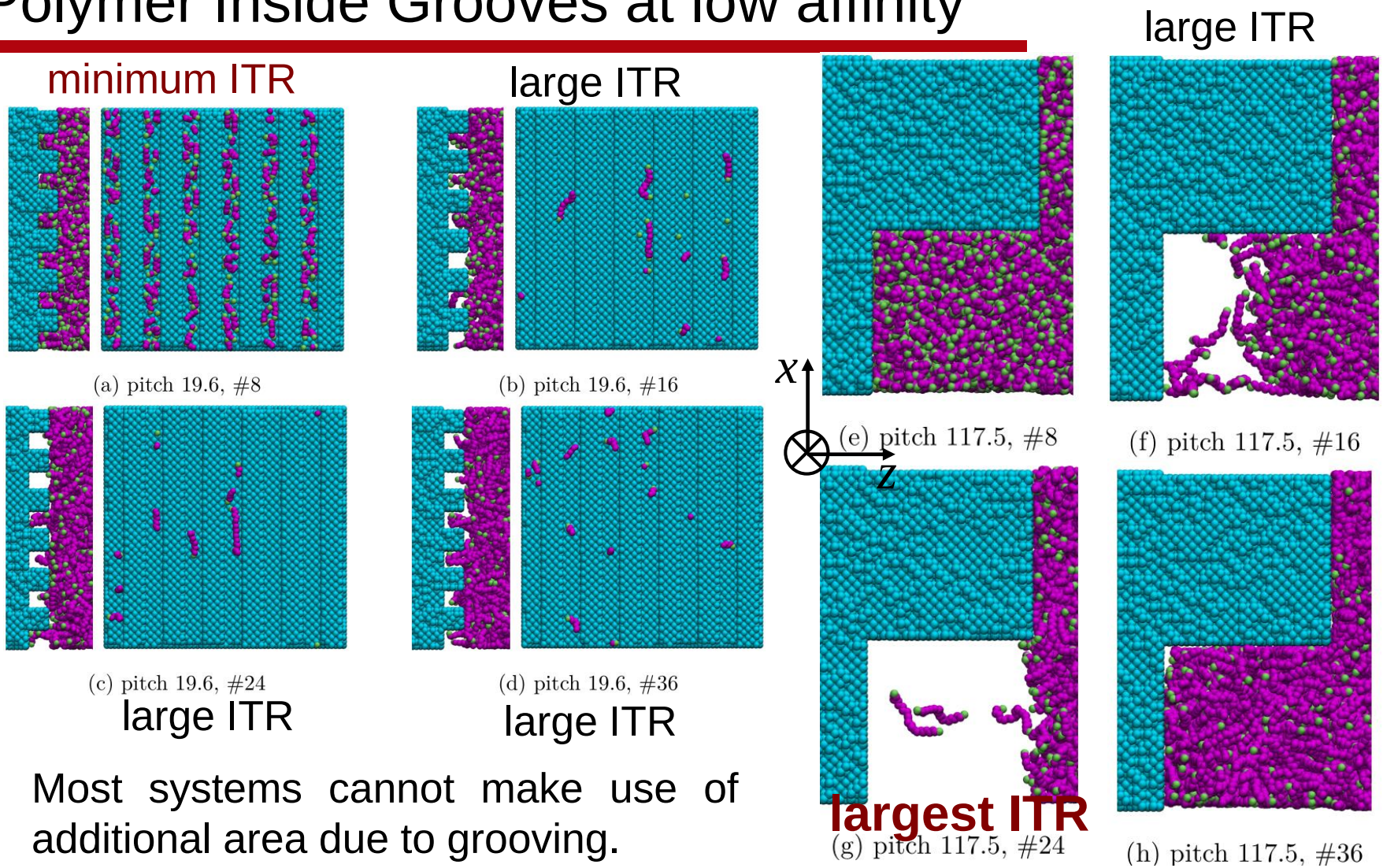
- adsorbed atoms per cross-section area



- ITR varies with chain-length
- shortest chain performed best interface thermal transport ability
- Roughness is not beneficial for most systems



Polymer Inside Grooves at low affinity



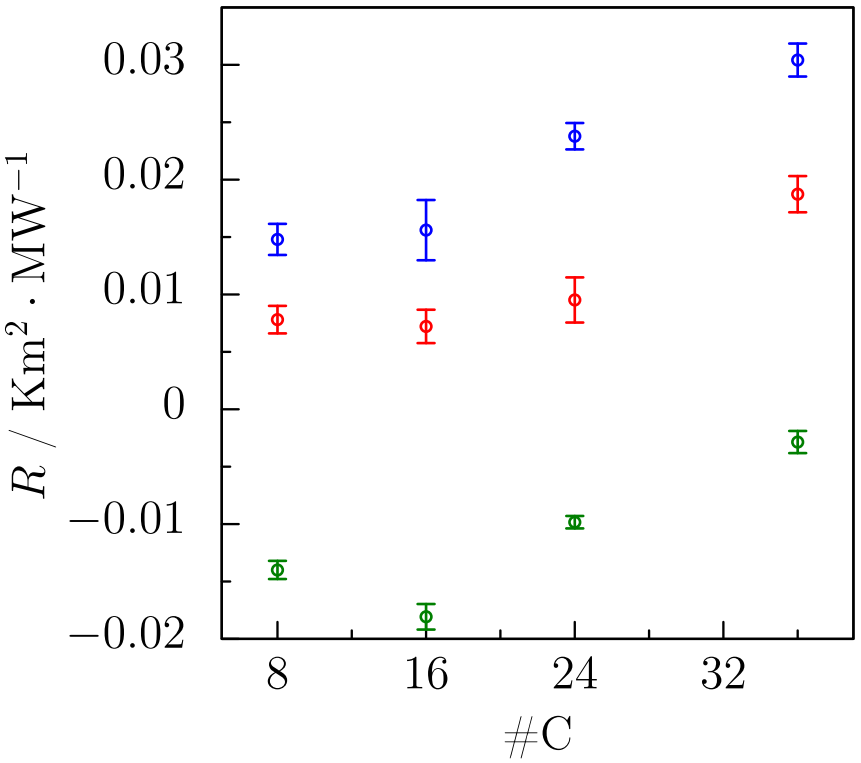
- Most systems cannot make use of additional area due to grooving.

- C8-small groove & C8-large groove & C36-large groove can make use of additional surface area.

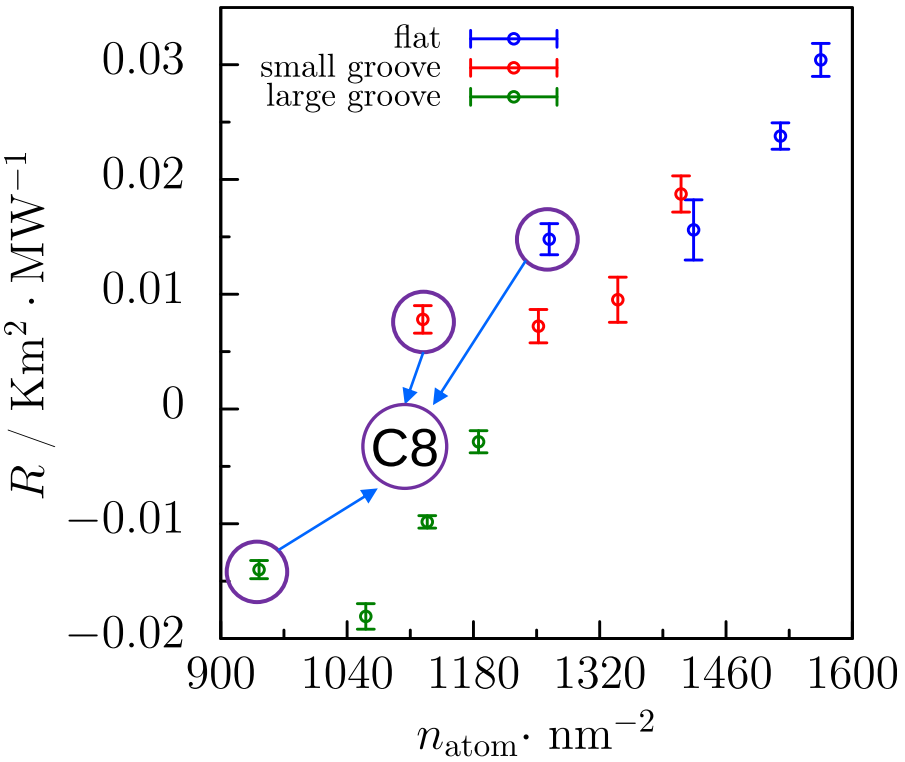
previous Au-Ar-Au work: all grooves were at Cassie state at low affinity

Results at high affinity

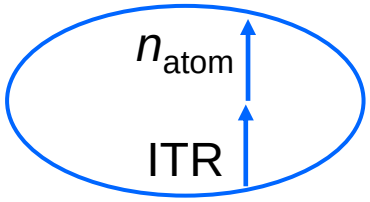
- ITR at different roughness scales



- adsorbed atoms per apparent area



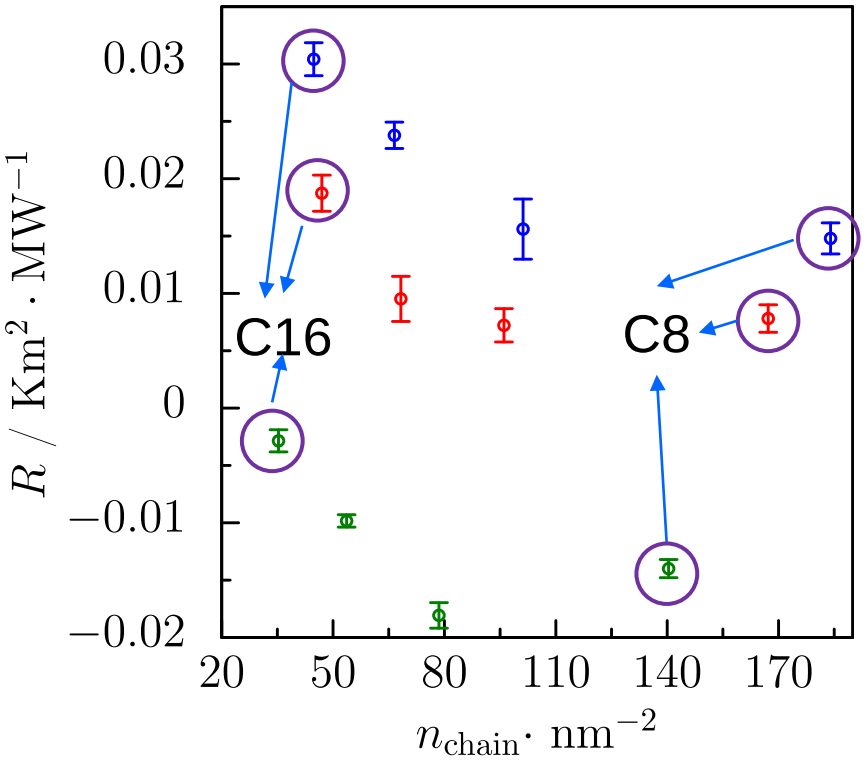
- ITR increases with chain-length overall
- $ITR_{large\ groove} < ITR_{small\ groove} < ITR_{flat}$
- ITR_{C8} close to ITR_{C16}

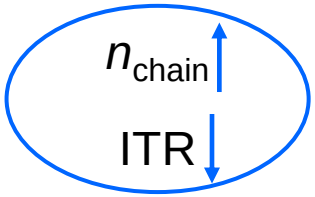


Unexpected result

Results at η of 0.5

- adsorbed chains per apparent area





Transport term

$$J_{\mathrm{tot}} = J_{\mathrm{kin}} + J_{\mathrm{pot}}$$

$$+J_{\mathrm{intramolecular}} + J_{\mathrm{intermolecular}}$$

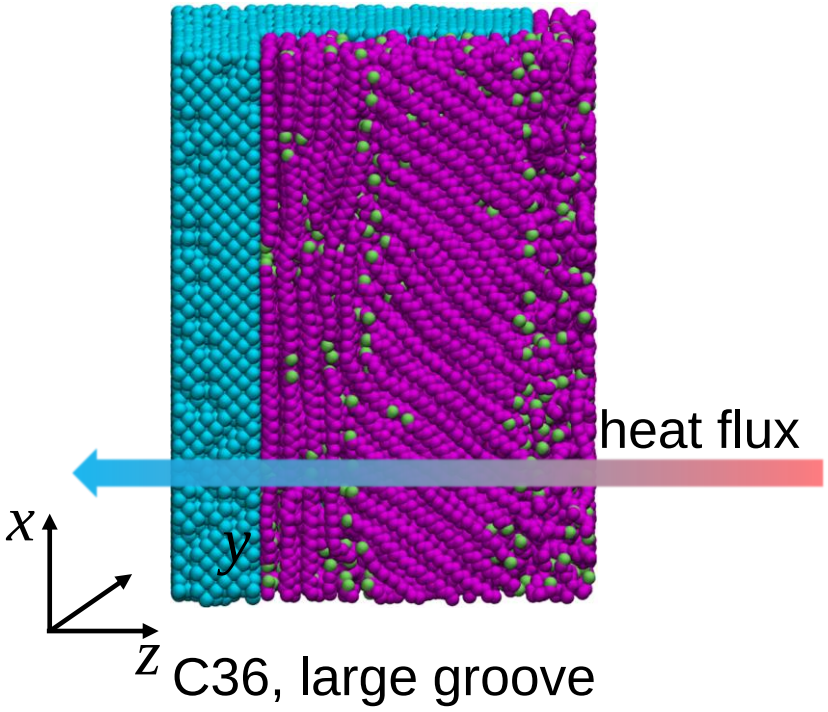
Interaction term

more adsorbed chains may produce more heat paths.

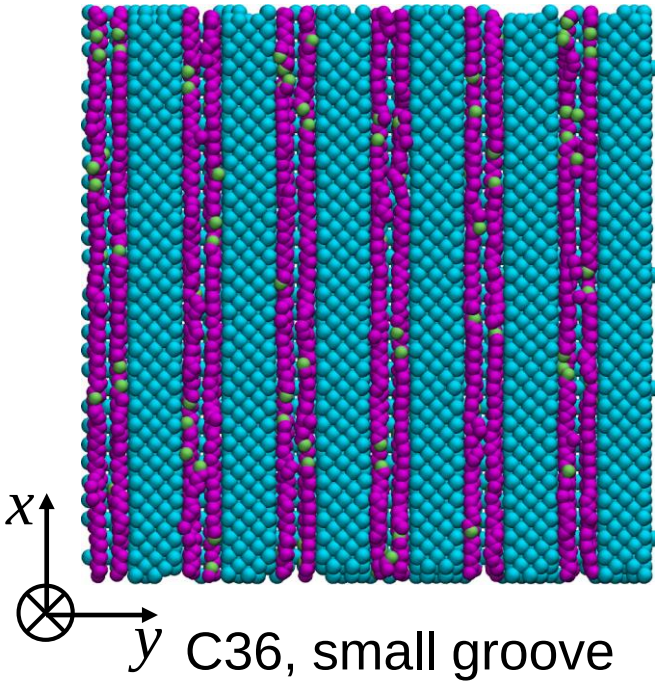
- Intramolecular interaction contribution dominantes as chain-length over 16.

Ohara T, et al. *J. Chem. Phys.* **135**, 034507 (2011)
Matsubara H, et al. *J. Chem. Phys.* **142**, 164509 (2015)

Polymer Inside Grooves at high affinity



C36, large groove



C36, small groove

chains is constrained vertical to heat flux

In case of large groove, *x*-*z* interfaces can adsorb chains at direction not vertical to overall heat flux direction.

$ITR_{large\ groove} < ITR_{small\ groove}$

Orientation of chains effects heat conduction.

Nakano T, et al. *J. Chem. Phys.* **133**, 154705 (2010)

Additional area due to grooving can be made full use of .

$ITR_{rough} < ITR_{flat}$

Conclusion

- ITR is effected by the chain-length of polymer TIM: at low affinity, the rule of correlation is hard to summarize; at high affinity, ITR increases with chain-length overall.
- Shortest chain length performed best interface thermal transfer ability at all roughness scales.
- At low affinity: roughness is not beneficial as the additional area cannot be made full use of. In general, more adsorbed atoms result into smaller ITR.
- At high affinity: ITR increases with chain-length overall. Adsorbed chains rather than atoms should be cared. More adsorbed chains can cause smaller ITR which may produce more heat paths. Large groove systems own surfaces adsorbing chains orienting at directions not vertical to overall heat flux. $ITR_{large\ groove} < ITR_{small\ groove} < ITR_{flat}$.

This work was supported by JST CREST Grant Number JPMJCR17I2

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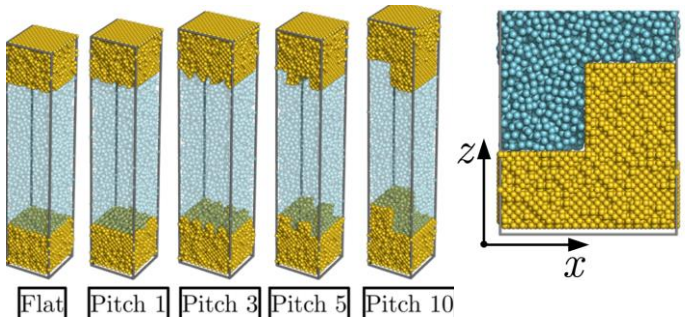

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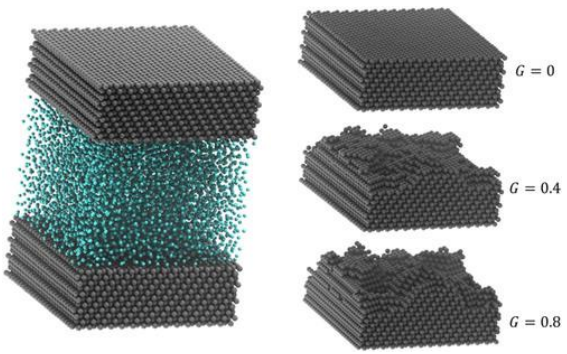

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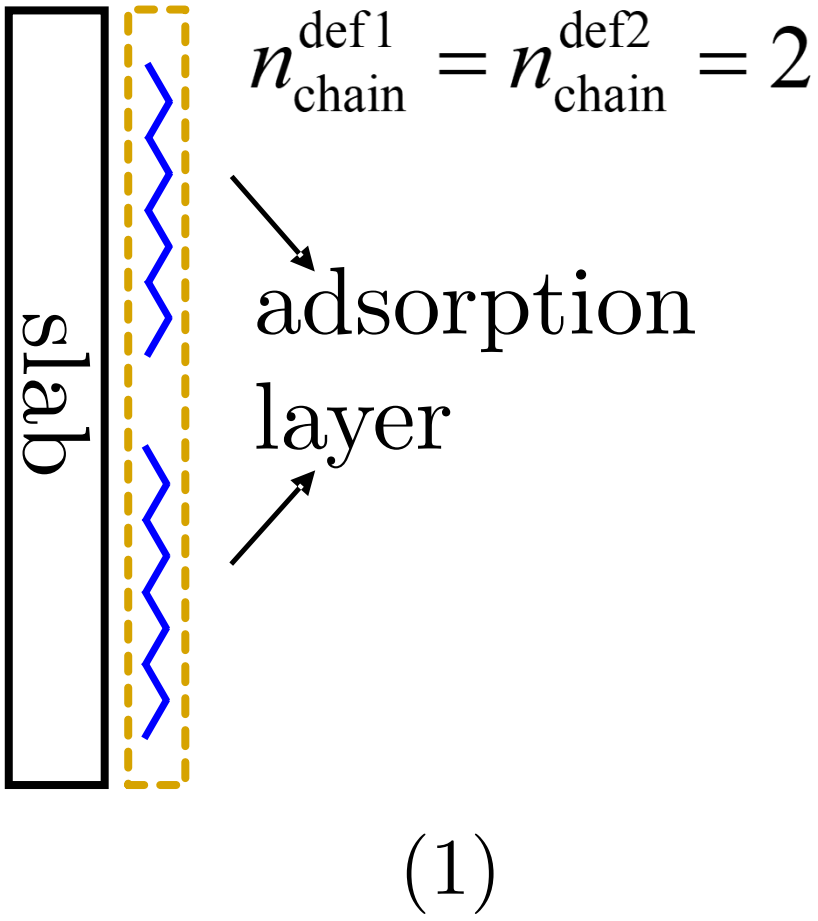
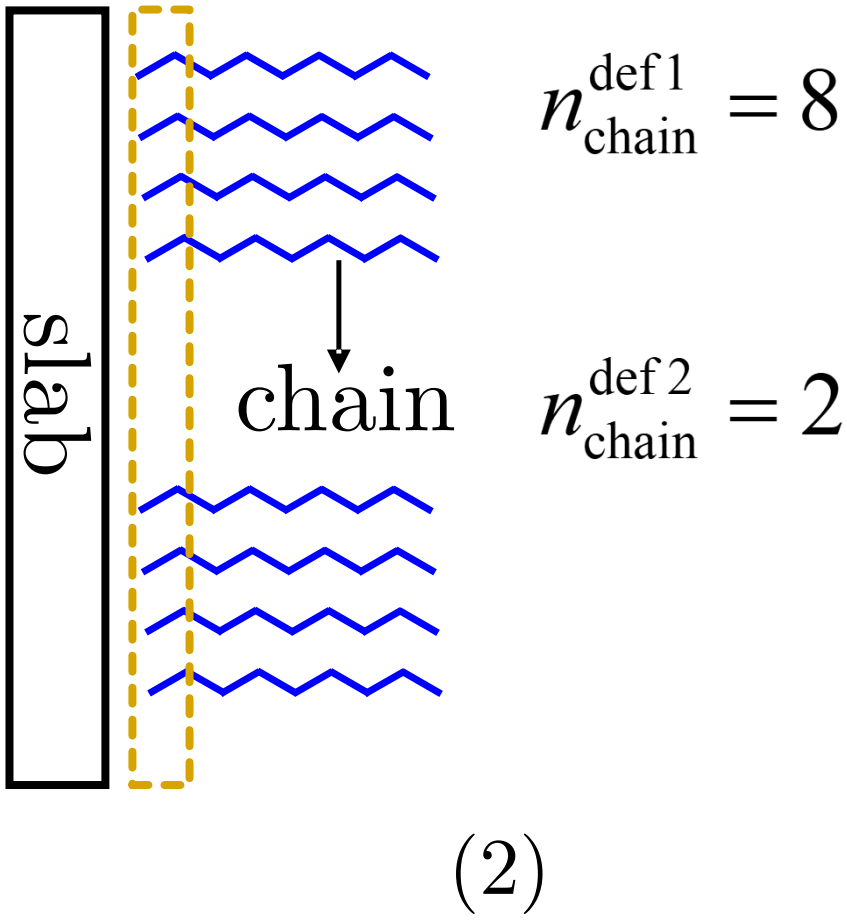

slab
$n_{\text{chain}}^{\text{def1}} = n_{\text{chain}}^{\text{def2}} = 2$
adsorption
layer

(1)

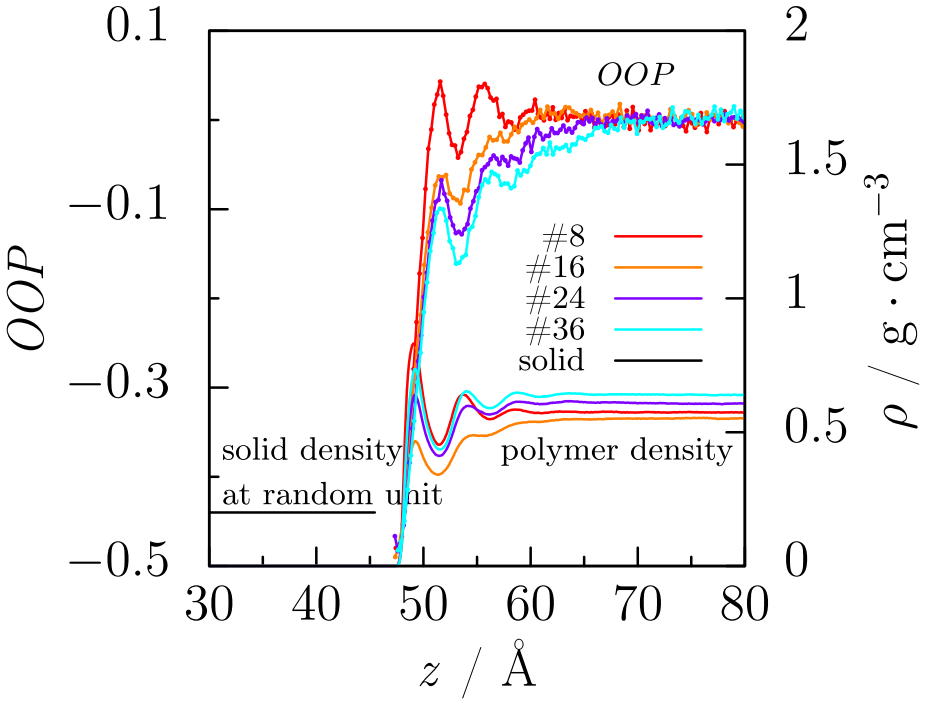

slab
chain
$n_{\text{chain}}^{\text{def1}} = 8$
$n_{\text{chain}}^{\text{def2}} = 2$

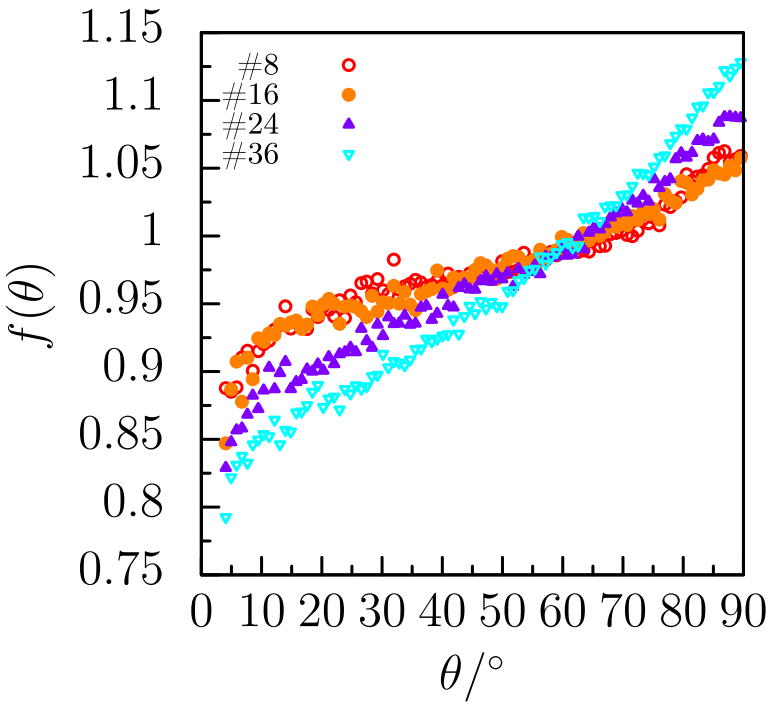
(2)

Liquid Structure at η of 0.1

- orientation order parameter
- orientation angle probability

for flat surface systems





$$OOP = \frac{1}{2}\left\langle 3\cos^2\theta^{z}_{s_i - s_{i+2}} - 1 \right\rangle$$

OOP = 1: vertical to cross-section
OOP = -0.5: parallel to cross-section

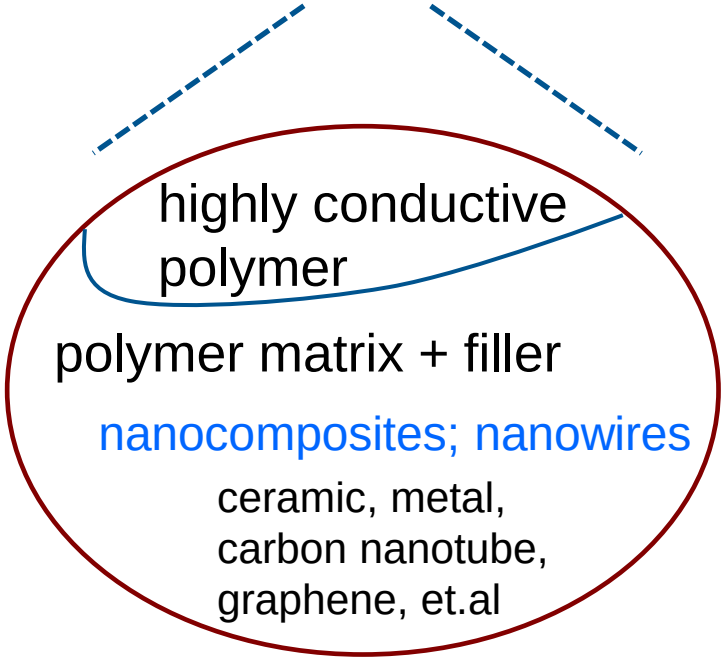
No apparent correlation between *OOP* and ITR was found.

Background

- TIM categories

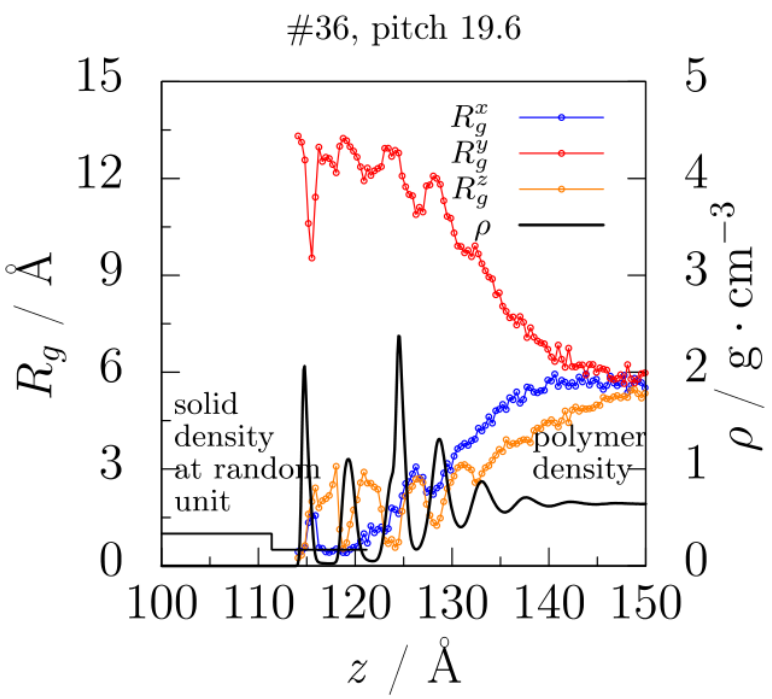
Type	Account
phase change TIMs	4%
metallic TIMs	9%
polymer-based TIMs	87%

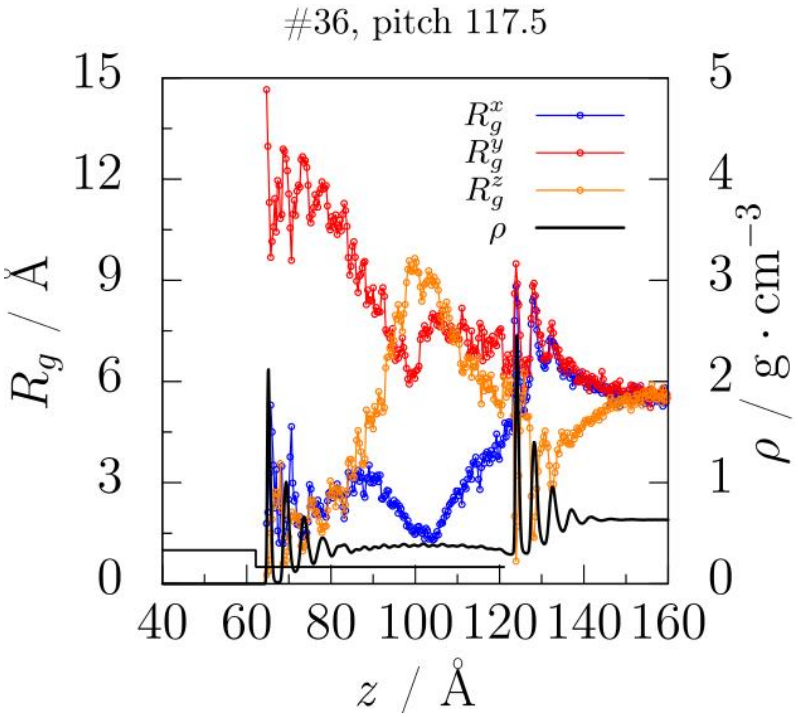
data source: BCC research



- merits of polymer-based TIM
  - ✓ mechanical properties
  - ✓ electronic properties
  - ✓ chemical properties
  - ✓ design flexibility
  - ✓ low price
  - ✓ conformability

Rog to explain pitch size setting

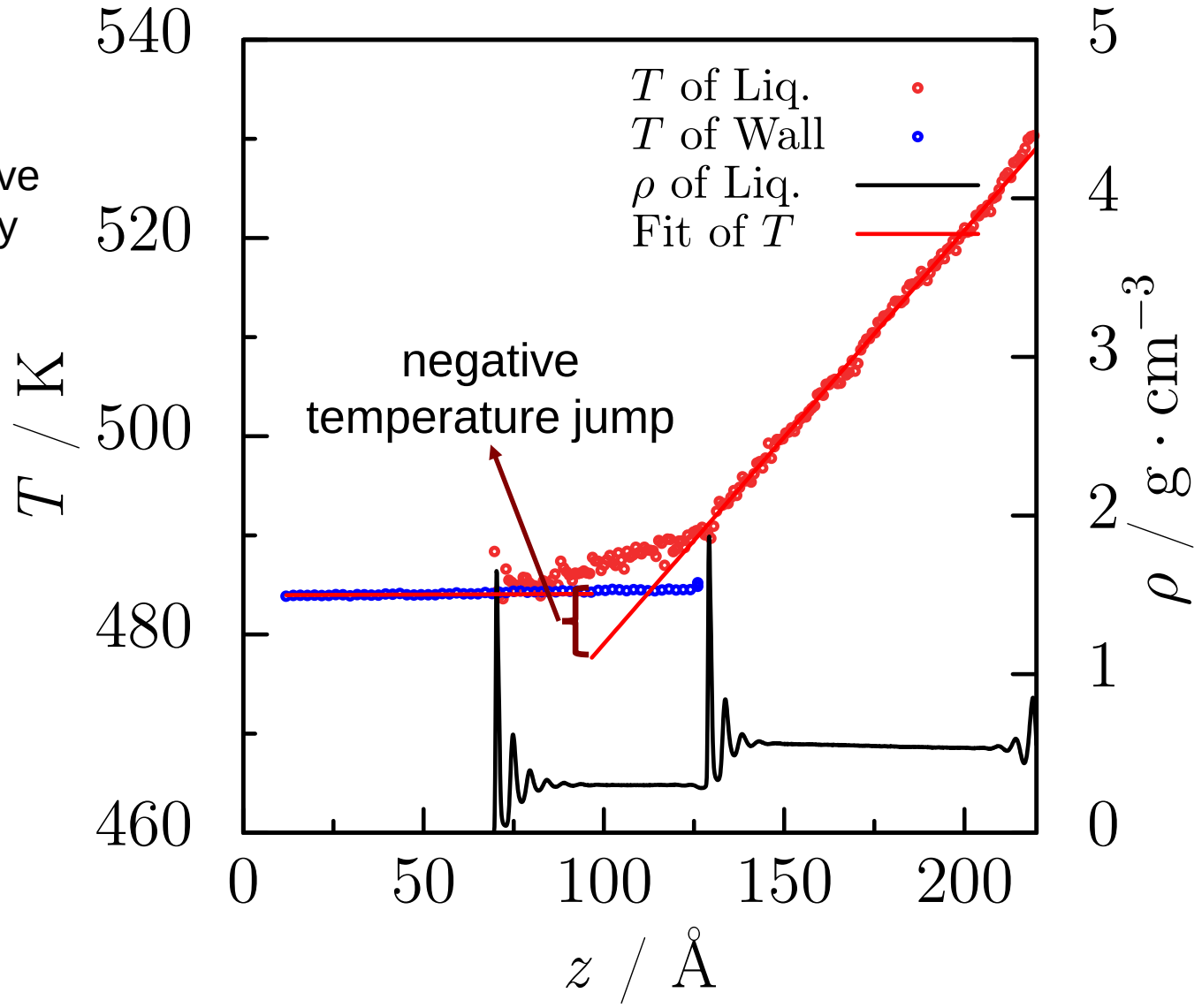




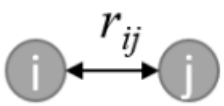
Negative ITR

- linear fitting of *T*

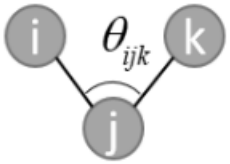
C16
large groove
high affinity



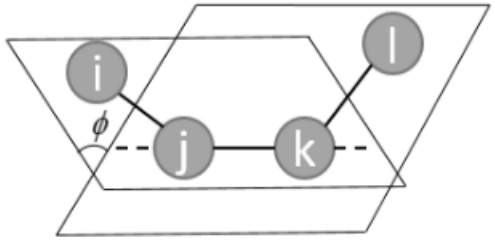
Index of vdW interaction



Stretching

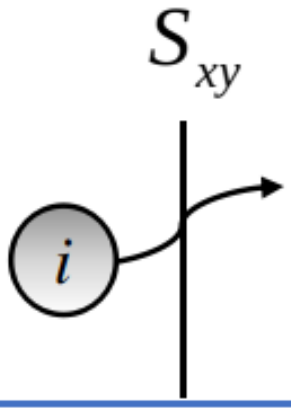


Bending

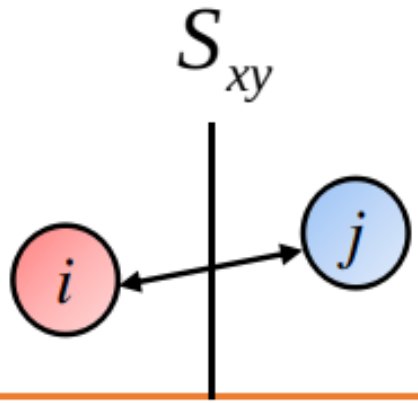


Torsion angle

Intramolecular interaction



Molecular transport



Intermolecular interaction

Cross-plane, apparent area explanation

cross- section: x-y plan

$S_{\text{cross-section}} = l_x * l_y$

$l_x = 2 * l_{x1}$

$S_{\text{apparent}} = 2*(l_{x1} + l_{z1}) * l_y$