Lubricated friction at the nanoscale: insights from molecular dynamics simulations and machine learning

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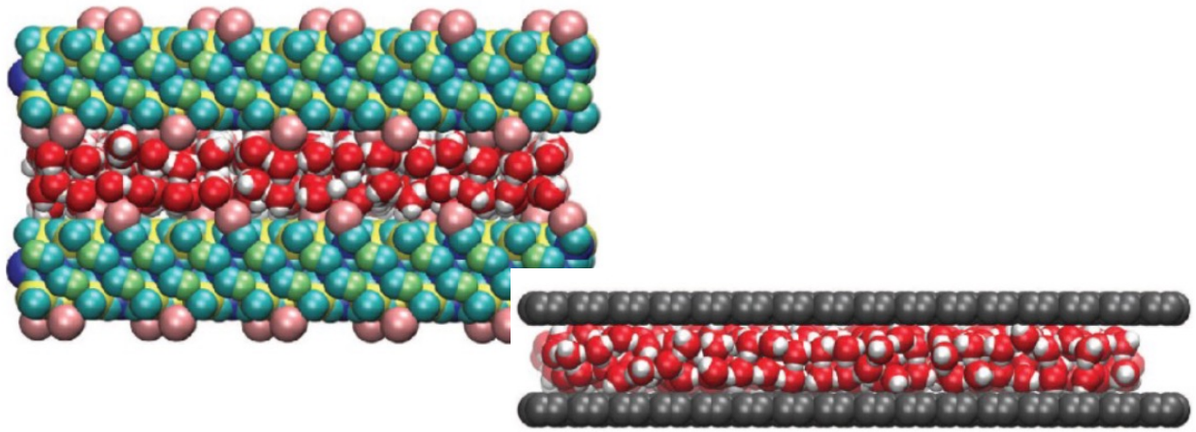
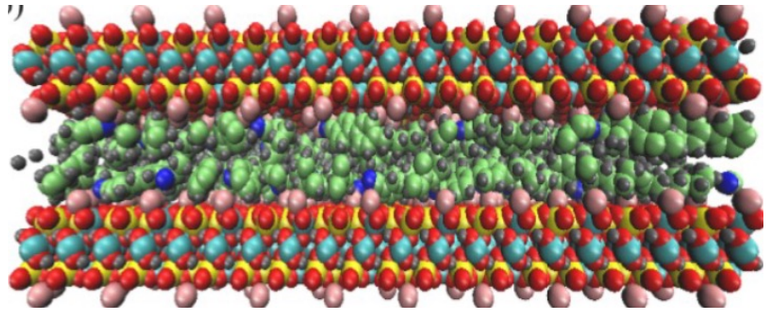
Outline: 3 topics on lubricated friction

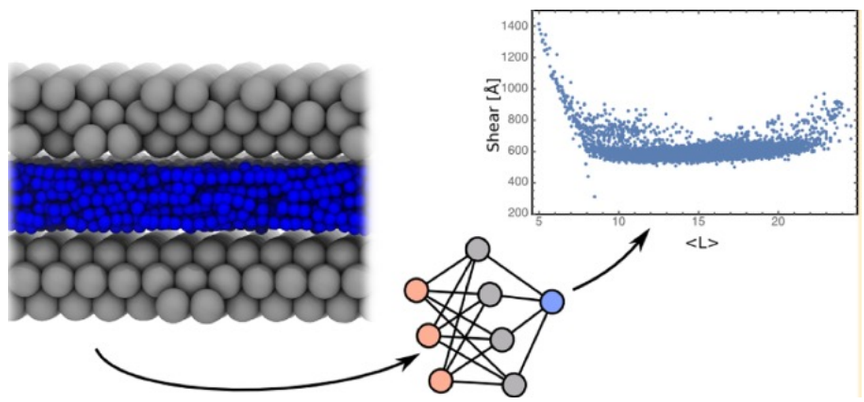
- Water confined by mica and graphene.

- Liquid crystal and hexane molecules confined by mica.

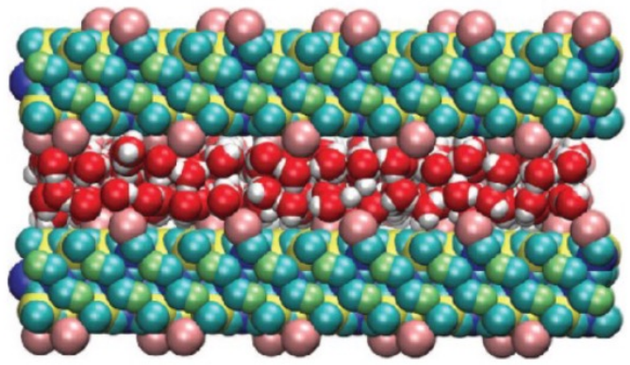
- Machine learning the relation between toy lubricant composition and frictional performance.

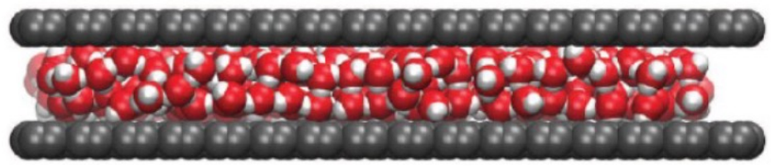






1. Water confined by mica and graphene

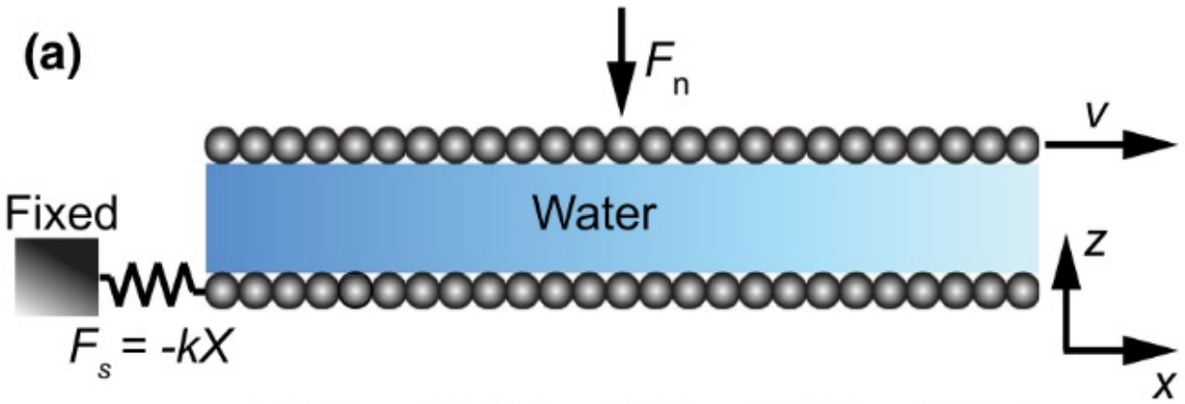


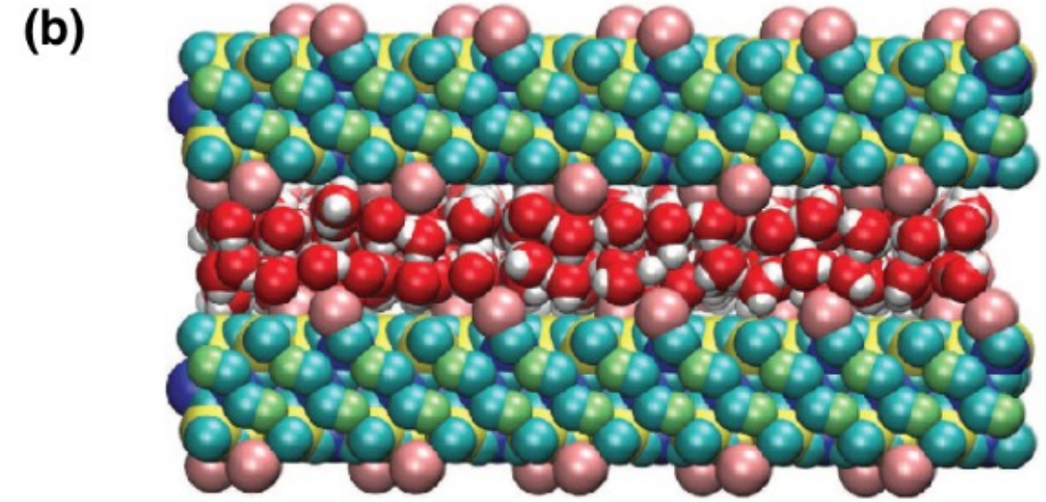


W. Chen, A. S. Foster, M. J. Alava, and LL,
Phys. Rev. Lett. 114, 095502 (2015).

Water confined by mica and graphene

- Rigid surfaces: Hydrophilic mica vs hydphobic graphene.
- Force fields from Heinz et al., Chem. Mater. (2005) for mica, and from Saito et al., Chem. Phys. Lett. (2001) for graphene.
- Flexible water molecules (SPC/Fw) in between.
- Langevin thermostat along y (no streaming bias), T = 295 K.
- Apply 1 atm pressure, sliding velocity 0.1 m/s.
- Simulations with LAMMPS.





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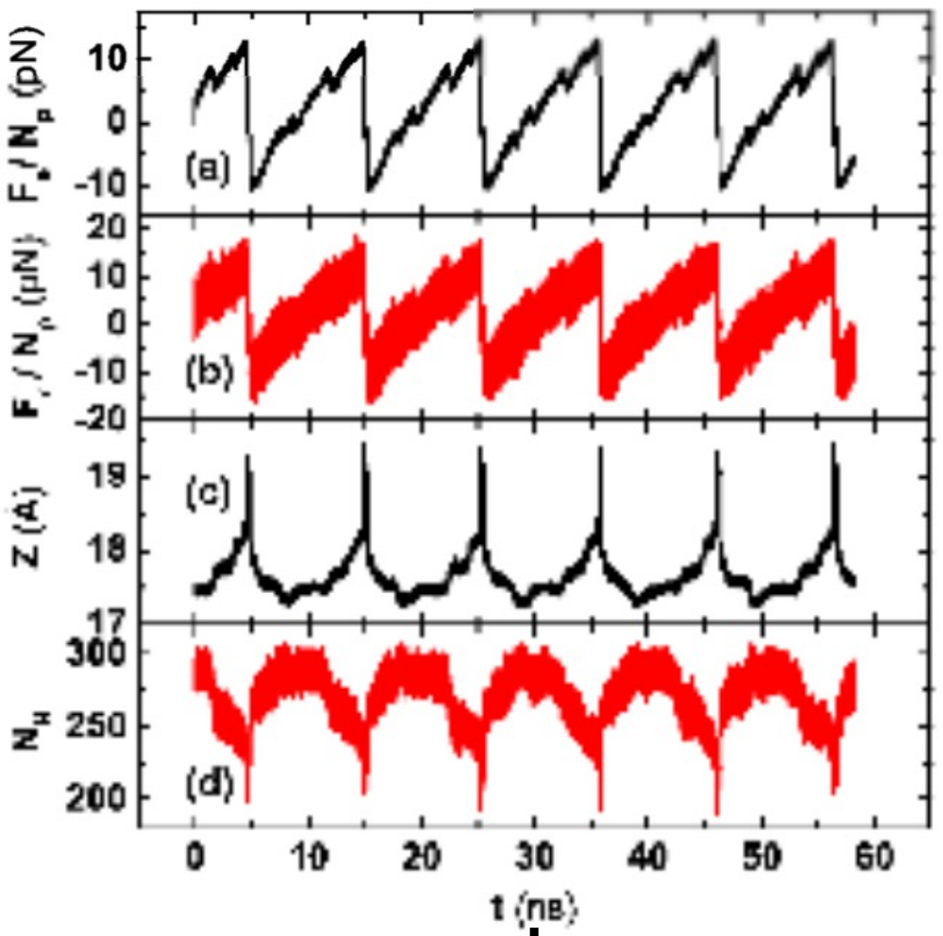
mica: $KAl_2(Al, Si_3)O_{10}(OH)_2$

Water confined by mica and graphene

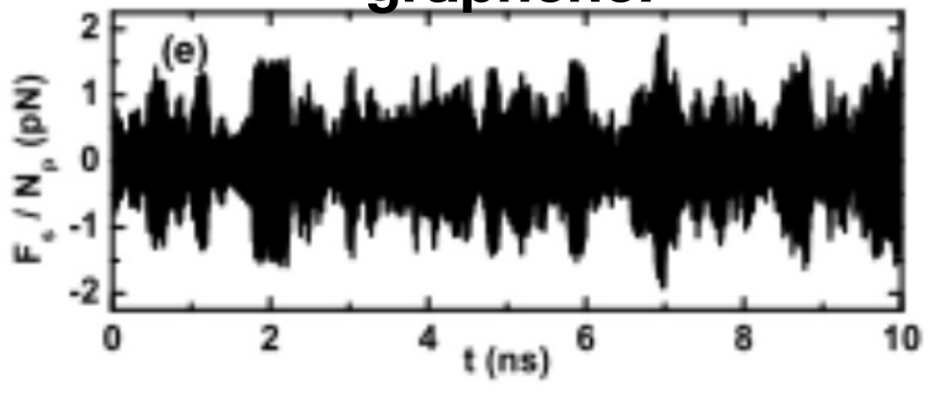
- Start by considering ”thin” layers of water.
- Stick-slip dynamics for the hydrophilic mica-confined system.
- Jumps of the top plate and broken hydrogen bonds between water and mica during slip events.
- No stick-slip in the hydrophobic graphene-confined system.

W. Chen, A. S. Foster, M. J. Alava, and LL, Phys. Rev. Lett. 114, 095502 (2015).

mica:



graphene:

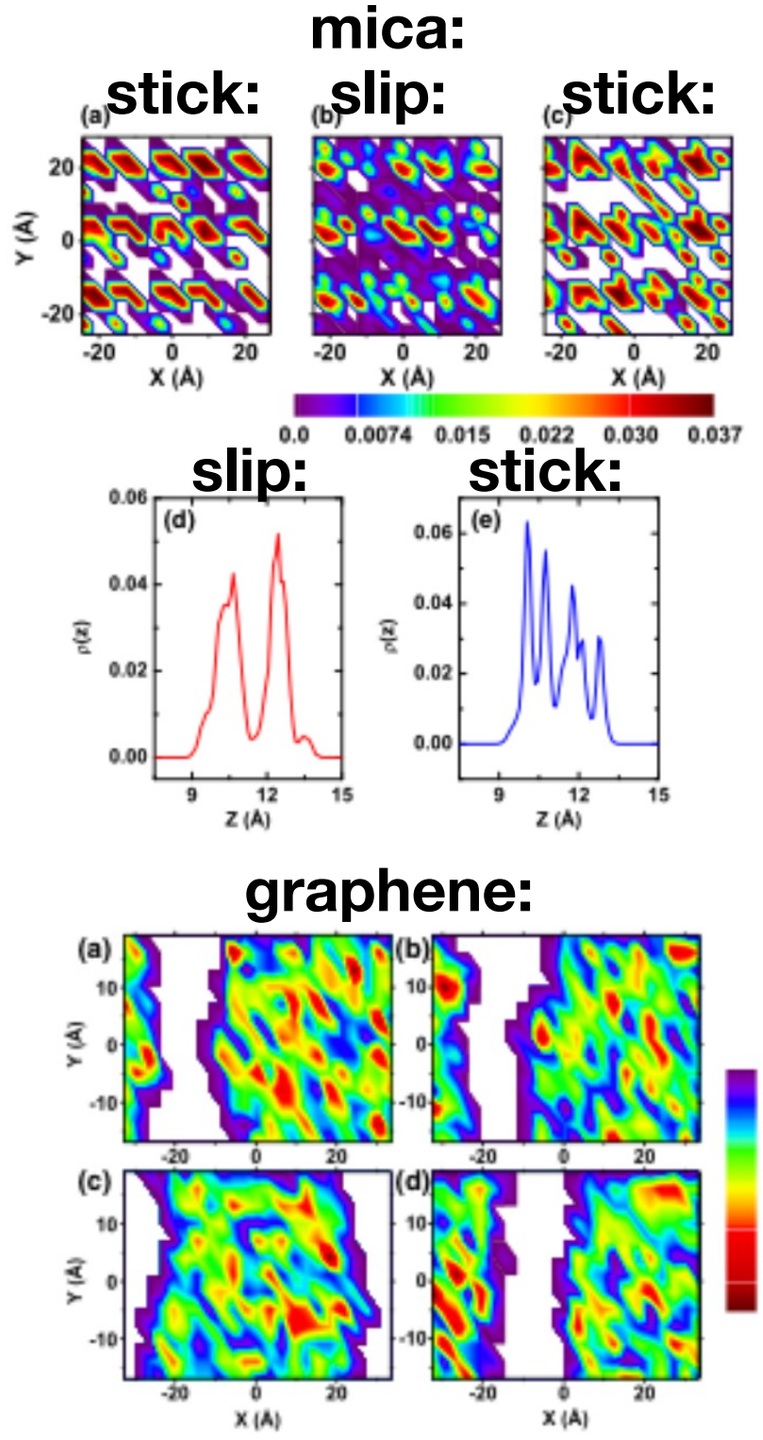


Water confined by mica: nanoscale ”capillary bridges”

- In the stick phase, water molecules condence around the potassium ions of mica.

- These nanoscale ”capillary bridges” break during the slip events.

- No such mechanism for graphene, and hence no stick slip.



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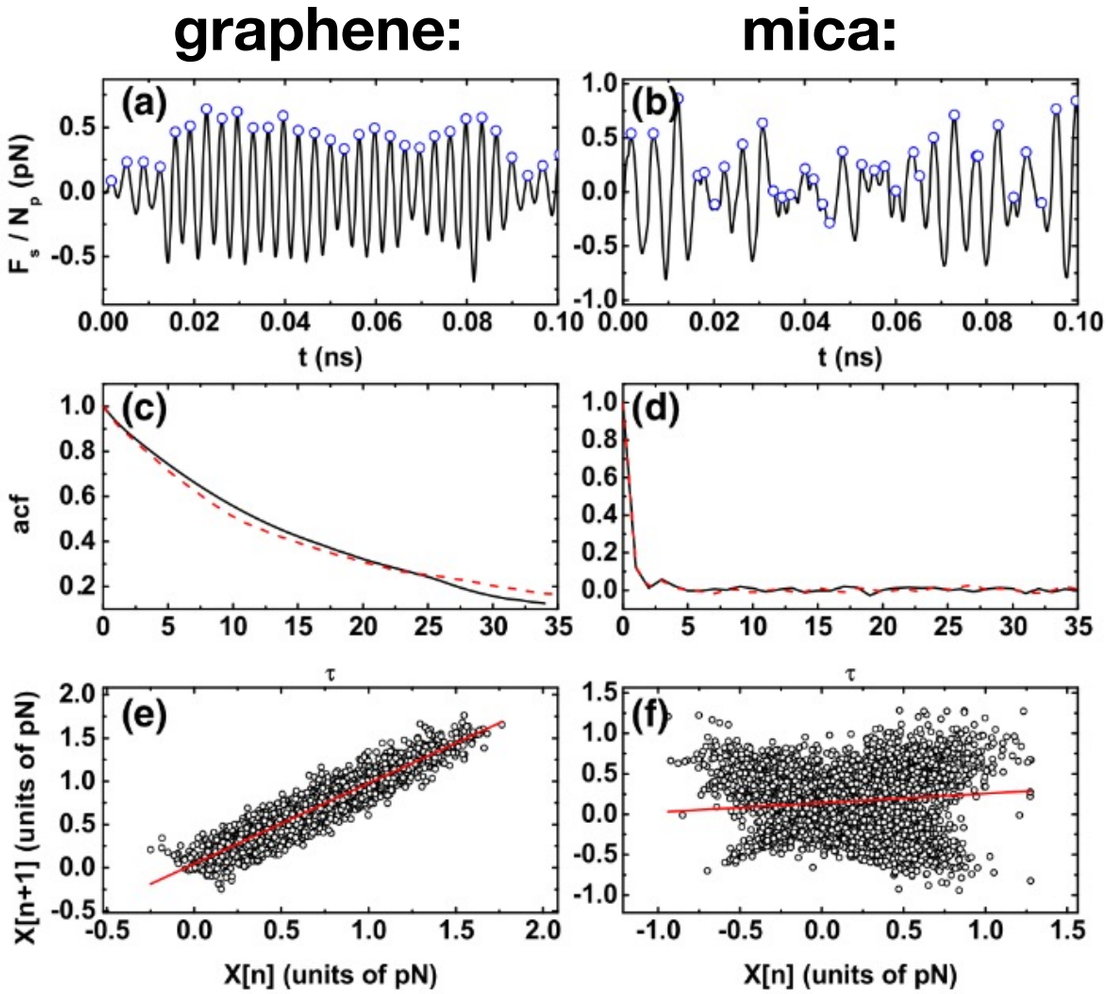
Thicker water layers: short time scale dynamics

- Considering thicker water layers leads to absence of stick-slip for both confining surfaces.

- The time-dependent amplitude of the friction force oscillations may be modeled as an Ornstein-Uhlenbeck process.

- Distinct signatures of mica and graphene observable in the fluctuations.

- Mica: strength of W does not depend on film thickness above ~1.8 nm.



$$f = 1/(2\pi)\sqrt{k/M}$$

$$X(n+1) - X(n) = -(1-\alpha)X(n) + W$$

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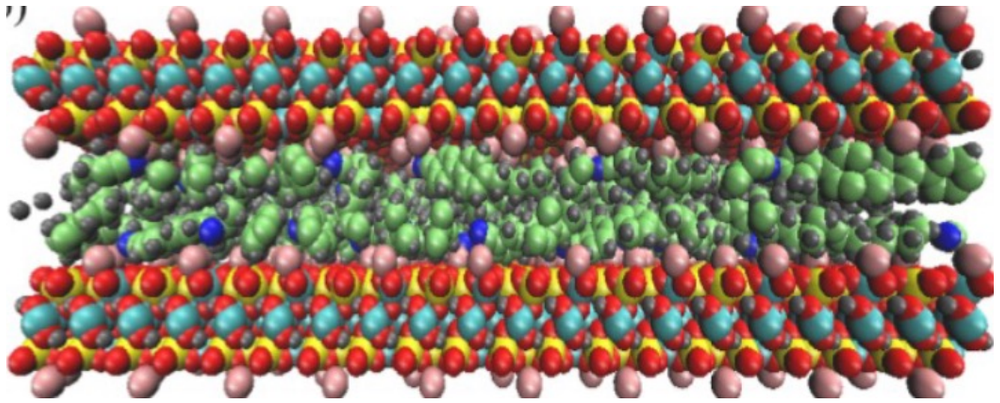
2. Liquid crystal (6CB) and hexane confined by mica



P. Kumar Jana, W. Chen, M. J. Alava, and LL, to be submitted.

Liquid crystal (6CB) and hexane confined by mica

- Rigid mica surfaces.
- As the arrangement of potassiums on mica is not known, consider 3 different cases.
- Flexible 6CB and hexane molecules, force fields from Adam et al., Phys. Rev. E (1997) and Cheung et al., Phys. Rev. E (2002).
- Langevin thermostat along y (no streaming bias), T = 298 K.
- Apply 1 atm pressure, sliding velocity 0.1 m/s.
- Simulations with LAMMPS.

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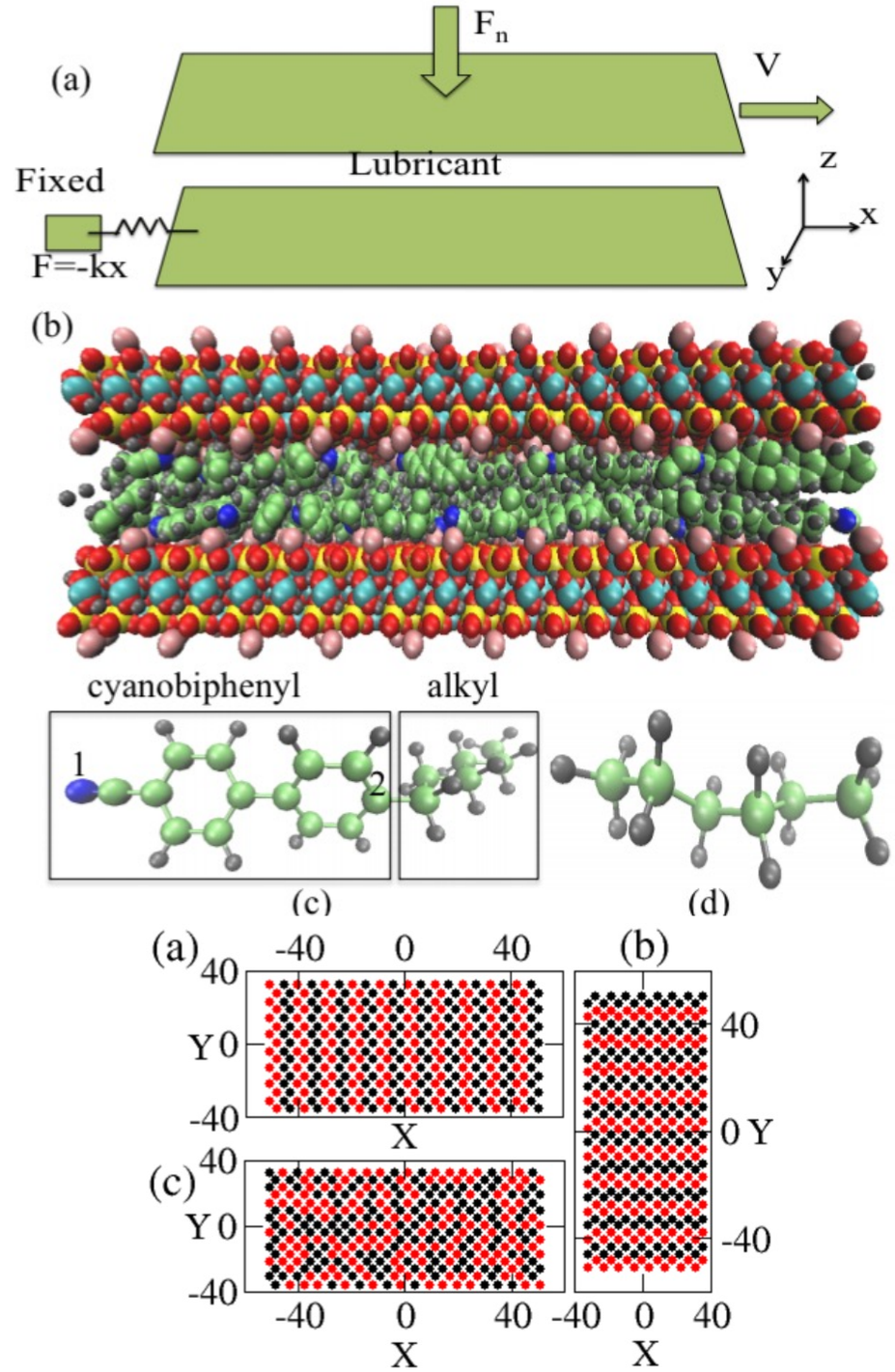
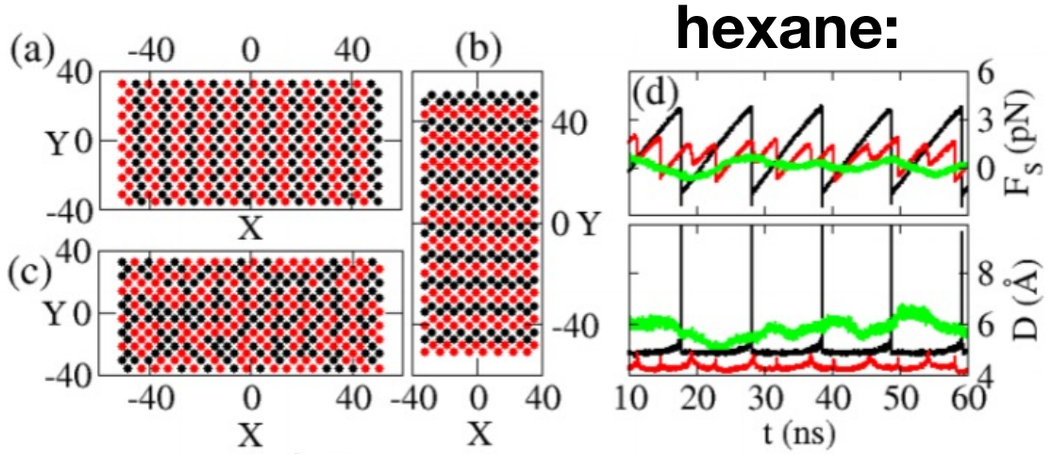


Monolayers of 6CB and hexane: stick-slip

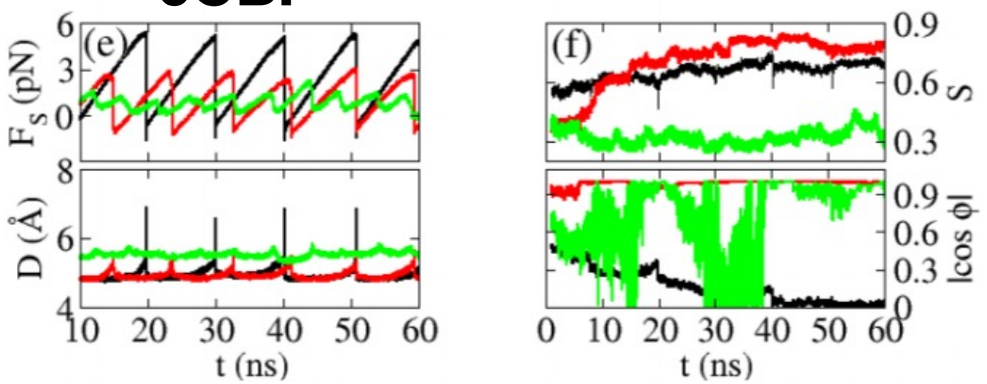
- Both 6CB and hexane exhibit stick-slip.
- Stick-slip magnitude controlled by the arrangement of the mica potassiums: grooves parallel/ perpendicular to sliding, or randomly positioned ions.
- Competing ordering mechanisms lead to variations in the nematic order parameter.

P. Kumar Jana, W. Chen, M. J. Alava, and LL, to be submitted.

hexane:



6CB:



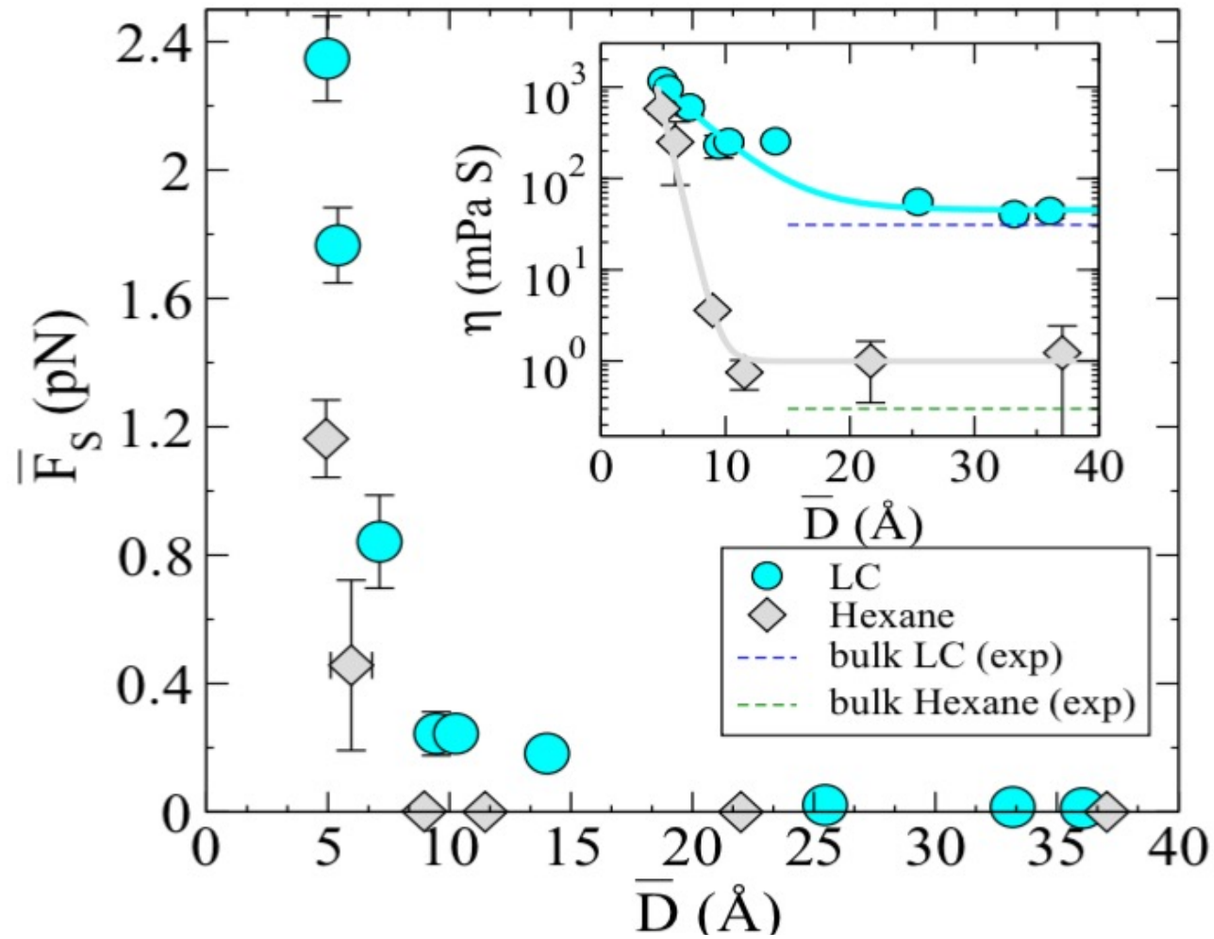
$$Q_{\alpha,\beta} = \frac{1}{N}\sum_{i}^{N}\left(\frac{3}{2}u_{\alpha}^{i}u_{\beta}^{i} - \frac{1}{2}\delta_{\alpha,\beta}\right)$$

Thicker lubricant layers: towards bulk viscosity

- Decreasing friction force and dynamic viscosity with increasing film thickness D.

- Exponential fits to the dynamic viscosities lead to decay lengths of 0.7 and 3.4 Å for hexane and mica, respectively.

- Both systems appear to approach the literature values of their bulk viscosities for large D: V=0.1 m/s slow enough to avoid large rate effects.

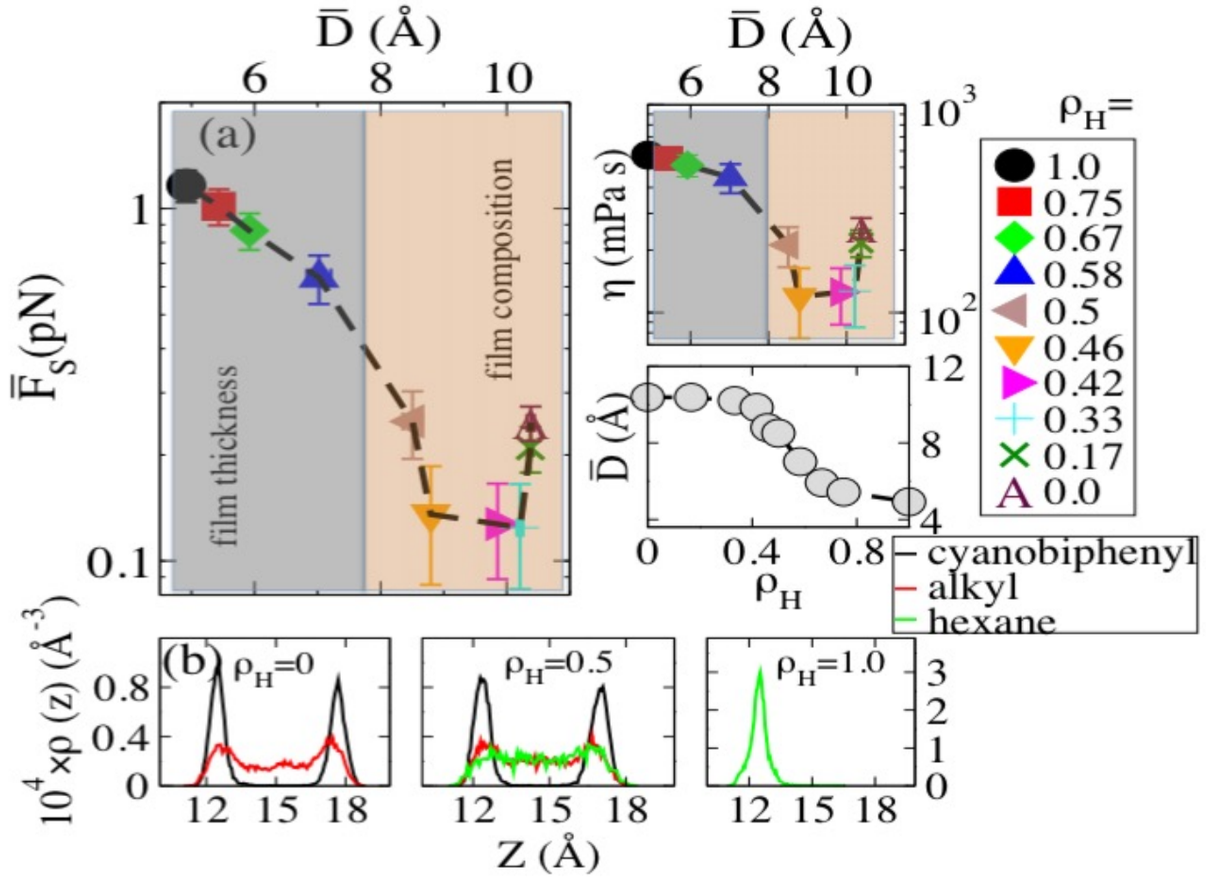


$$\bar{F}_s N_p = \eta A (V/\bar{D})$$

P. Kumar Jana, W. Chen, M. J. Alava, and LL, to be submitted.

Mixtures of 6CB and hexane: nonmonotonic behavior

- Fix the total number of molecules to 144, and vary the fraction of hexane.
- 6CB is the bigger molecule; D increases with decreasing hexane concentration.
- Two regimes: for large hexane concentration, friction decreases with D, while for systems with mostly 6CB, friction increases with D.
- The ”sticky” nature of 6CB dominates over the decrease of friction due to increasing D.



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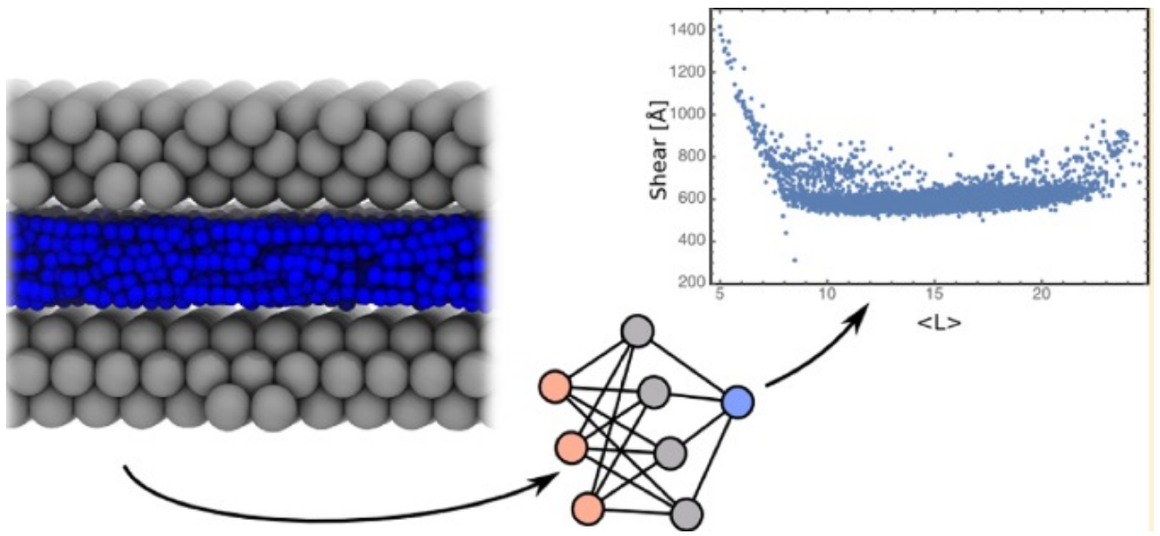
3. Machine learning the relation between lubricant composition and friction



M. Zaidan, F. Federici, LL, and A. S. Foster, J. Chem. Theory Comput. 13, 3 (2017).

Create a database: 8000 MD simulations of random lubricants

- No sufficiently large database available: create one!

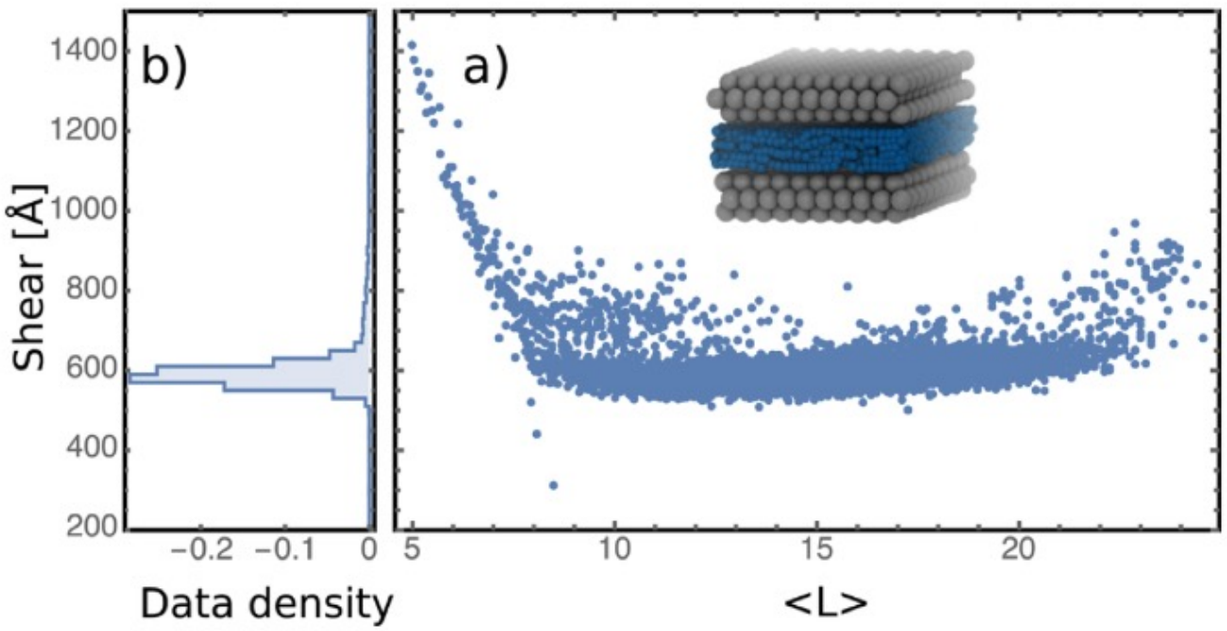
- Toy model: confining surfaces slabs of FCC lattice, flexible ”polymer” chains with chain lengths (max 25) picked randomly from random distributions.

- Chains of particles connected by springs, chain particles interact via the Lennard-Jones potential, chain-surface interactions are modeled by the Morse potential.

- Constant T (Langevin), constant load.

- Apply a constant shear force, measure the sliding distance over a fixed time (large shear = good lubricant).

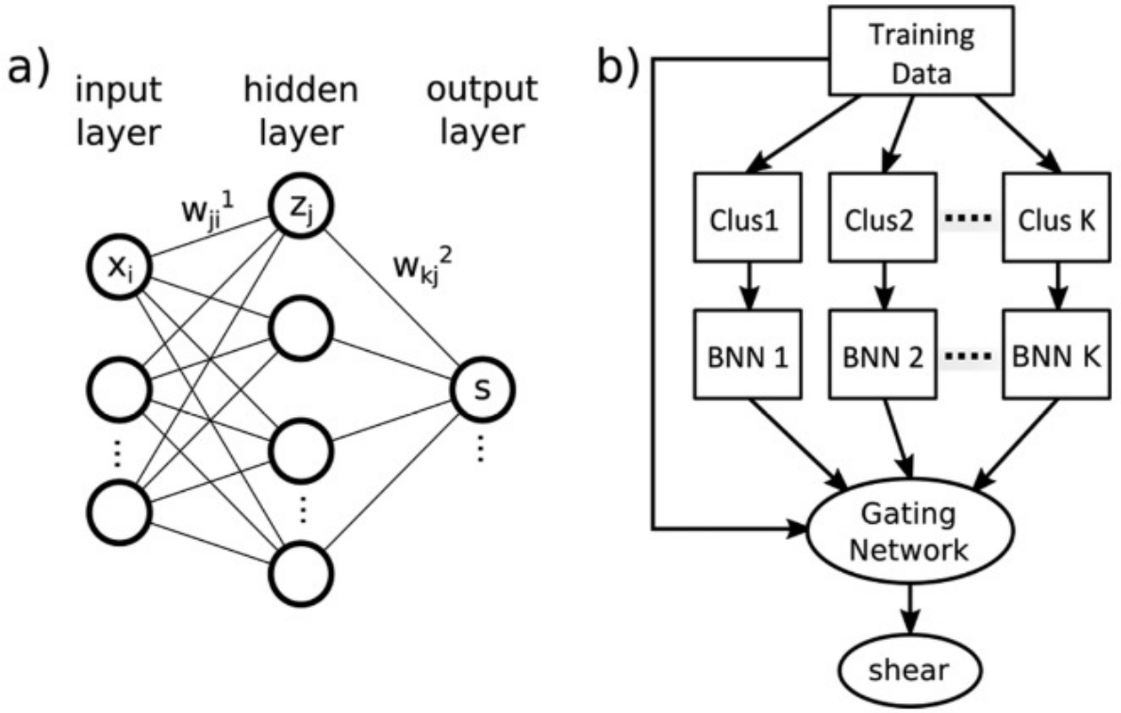
- One run takes a few hours on a GPU: a significant computational effort.



M. Zaidan, F. Federici, LL, and A. S. Foster, J. Chem. Theory Comput. 13, 3 (2017).

Machine learning model: mixture of clustered Bayesian neural networks

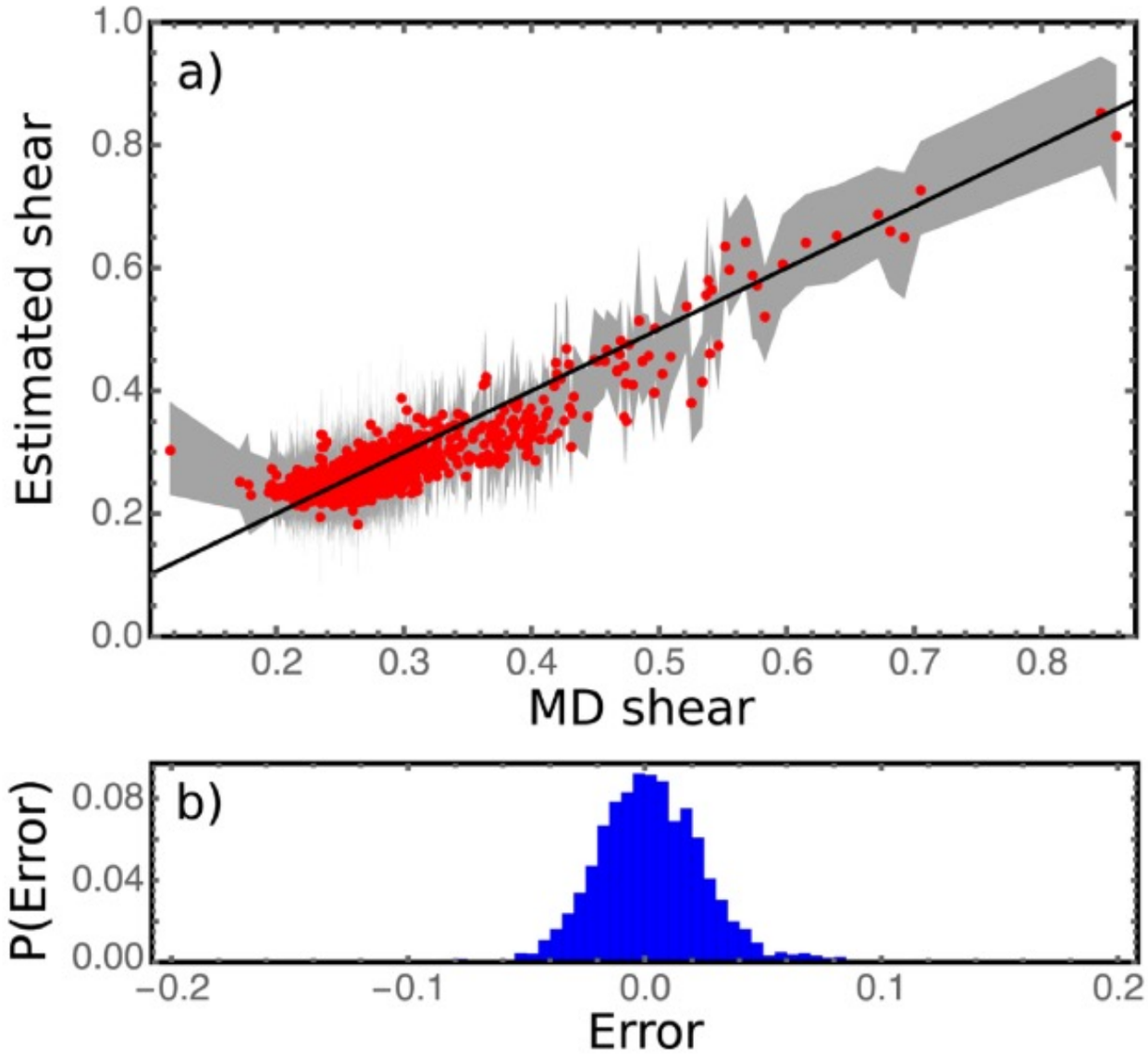
- Neural network: a mapping from the 25 dimensional input vector (”descriptor”) to a single number (”shear”).
- Use a training set (~70% of the data) to adjust the weights of the network.
- Test using the remaining ~30% of data
- Here, apply k-means clustering to divide the data into clusters, and train an expert network for each.
- Combine the outputs using a gating network.
- Better performance than using a single network.



M. Zaidan, F. Federici, LL, and A. S. Foster, J. Chem. Theory Comput. 13, 3 (2017).

Does it work?

- Yes, pretty well.
- Regression plot of estimated shear vs MD shear, considering the validation set.
- Most predicted shear values are less than 5% off.
- Looks promising: can we replace MD (which takes hours/run) by evaluation of the ML model, taking a fraction of a second?



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Some limitations…

- Try the following: feed the ML model a very large number of random chain length distributions, pick the best lubricants, check with MD (”lubricant optimisation”).

- It turns out that the model is not very good at coping with data that is has not seen before.

- The best lubricants according to the ML tend to be better than average, but not as good as predicted.

- Large fluctuations between the predicted and actual shear from sample to sample.

- Limited usefulness for screening new lubricants.

Conclusions

- Composition of the confining surfaces (mica vs graphene) controls the nature of water-lubricated friction at the nanoscale (stick-slip or not, etc.).

- Positions of the K ions on mica are important for properties of monolayer LC lubrication.

- Tuning the mixture of 6CB and hexane allows some degree of friction control.

- Neural network model able to learn the relation between toy lubricant composition and friction (but does not generalise very well to configurations it has not seen before)

Thank you!