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Machine Learning Based Upscaling of Finite-Box Size Molecular Dynamics Diffusion Simulations

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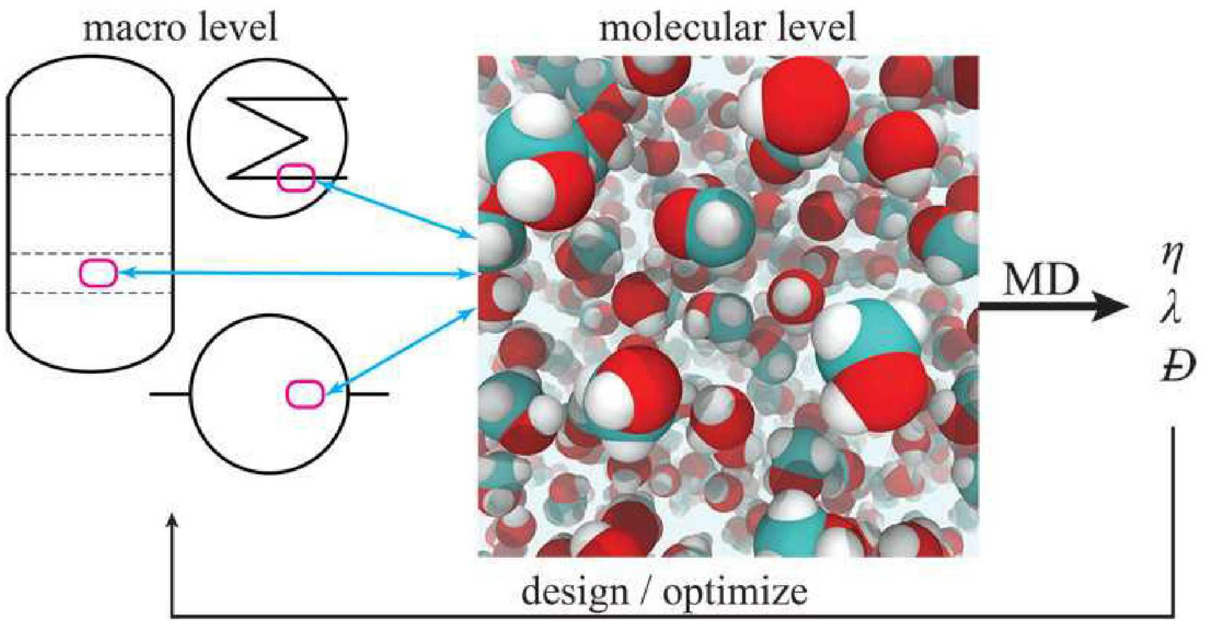
Molecular Dynamics Diffusion Simulations

- Diffusion experiments are slow, expensive, and often inconsistent
- Diffusion coefficients can be simulated using open-source MD packages
- MD uses Newton's equations of motion and an interatomic potential



S. H. Jamali *et al.*, OCTP: A Tool for On-the-Fly Calculation of Transport Properties of Fluids with the Order- n Algorithm in LAMMPS. *J Chem Inf Model* **59**, 1290-1294 (2019).

Self-Diffusion of Lennard-Jones Fluids

Full . Published Online: 15 July 2020 Accepted: June 2020

Machine learning prediction of self-diffusion in Lennard-Jones fluids

J. Chem. Phys. **153**, 034102 (2020); https://doi.org/10.1063/5.0011512

Joshua P. Allers[1], Jacob A. Harvey[2], Fernando H. Garzon[3,4], *and* Todd M. Alam[1,a)]

- Joshua Allers et al. showed that artificial neural networks (ANN) can be trained to predict self-diffusion of Lennard Jones (LJ) fluids
- ANN can replicate the MD results at a lower computational cost
- Expand: real fluids, diffusion in pores, multicomponent

J. Allers, J. Harvey, F. Garzon, T. Alam, Machine learning prediction of self-diffusion in Lennard-Jones fluids. *J Chem Phys* **153**, (2020).

Jamali 2018

JCTC Journal of Chemical Theory and Computation

Article

Cite This: *J. Chem. Theory Comput.* 2018, 14, 2667−2677

pubs.acs.org/JCTC

Finite-Size Effects of Binary Mutual Diffusion Coefficients from Molecular Dynamics

Seyed Hossein Jamali,[†] Ludger Wolff,[‡] Tim M. Becker,[†] André Bardow,[‡] Thijs J. H. Vlugt,[†] and Othonas A. Moultos*,[†]

- Jamali published data for over 1000 simulation of binary LJ fluids
- Self-diffusion and Maxwell-Stefan diffusion coefficients

S. H. Jamali *et al.*, Finite-Size Effects of Binary Mutual Diffusion Coefficients from Molecular Dynamics. *J Chem Theory Comput* **14**, 2667-2677 (2018).

MD Input Parameters

N = Number of particles

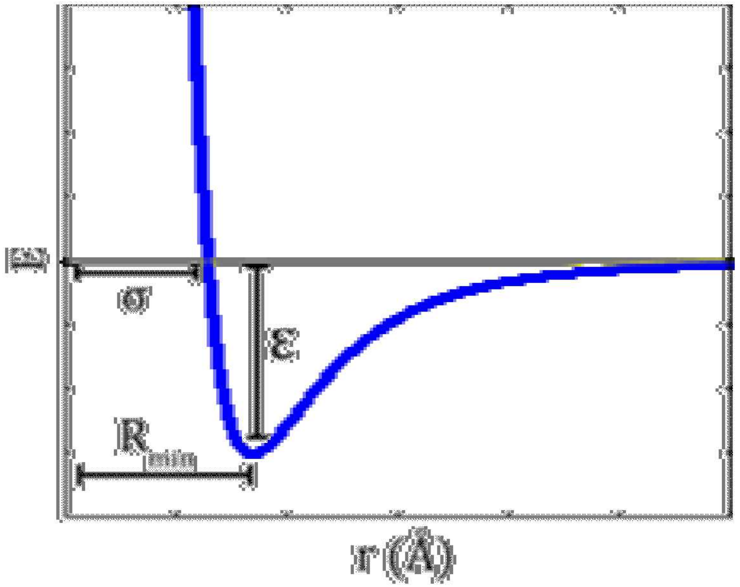
x = Mole fraction of Particle 1

ε = Well depth (strength of attractive forces)

σ = Van der Walls radius (particle size)

k = Non-ideal mixing parameter

Parameter	Values
N	500, 1000, 2000, 4000
x_1	0.1, 0.3, 0.5, 0.7 0.9
ε_2	1.0, 0.8, 0.6, 0.5
σ_2	1.0, 1.2, 1.4, 1.6
k_{12}	0.05, 0.0, -0.3, -0.6

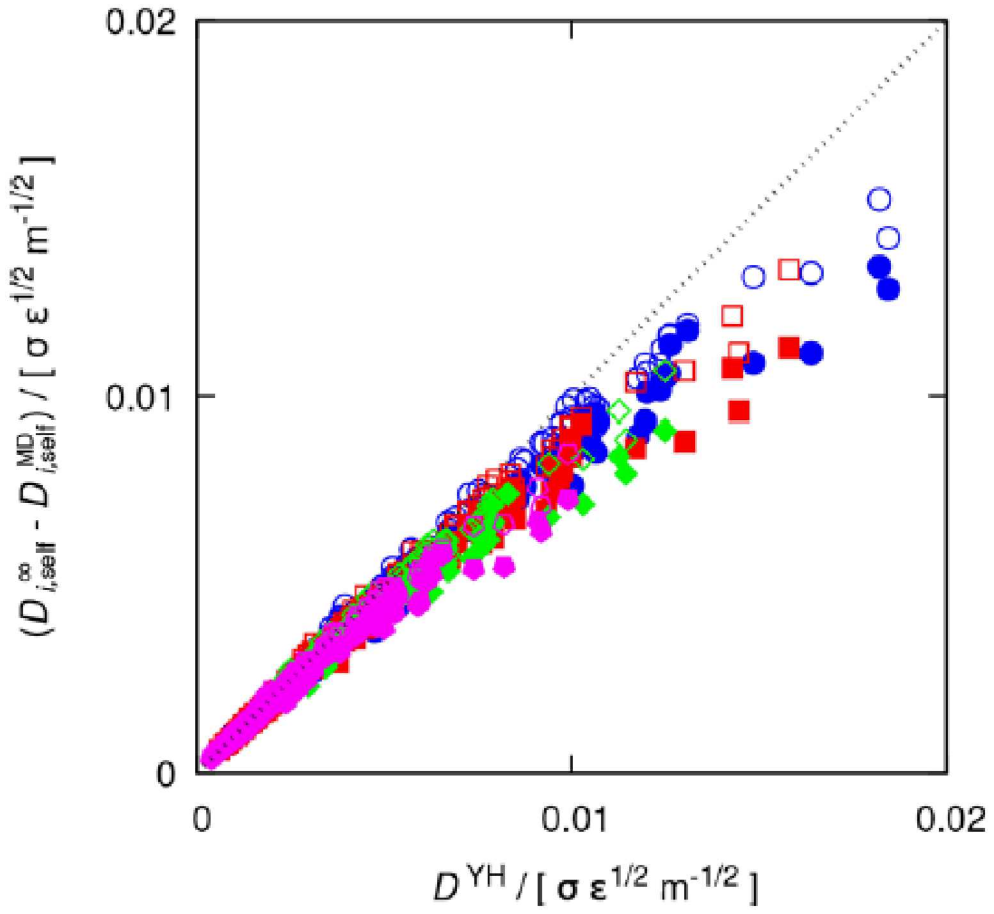


Self-Diffusion Finite-Box Effects

- MD diffusion simulations show finite-box size effects
- Yeh and Hummer (YH) correction used for self-diffusion of single component fluids

YH Correction: $D^{\infty} = D^{MD} + \frac{\xi k_B T}{6\pi\eta L}$

- Jamali used the YH correction for binary fluids

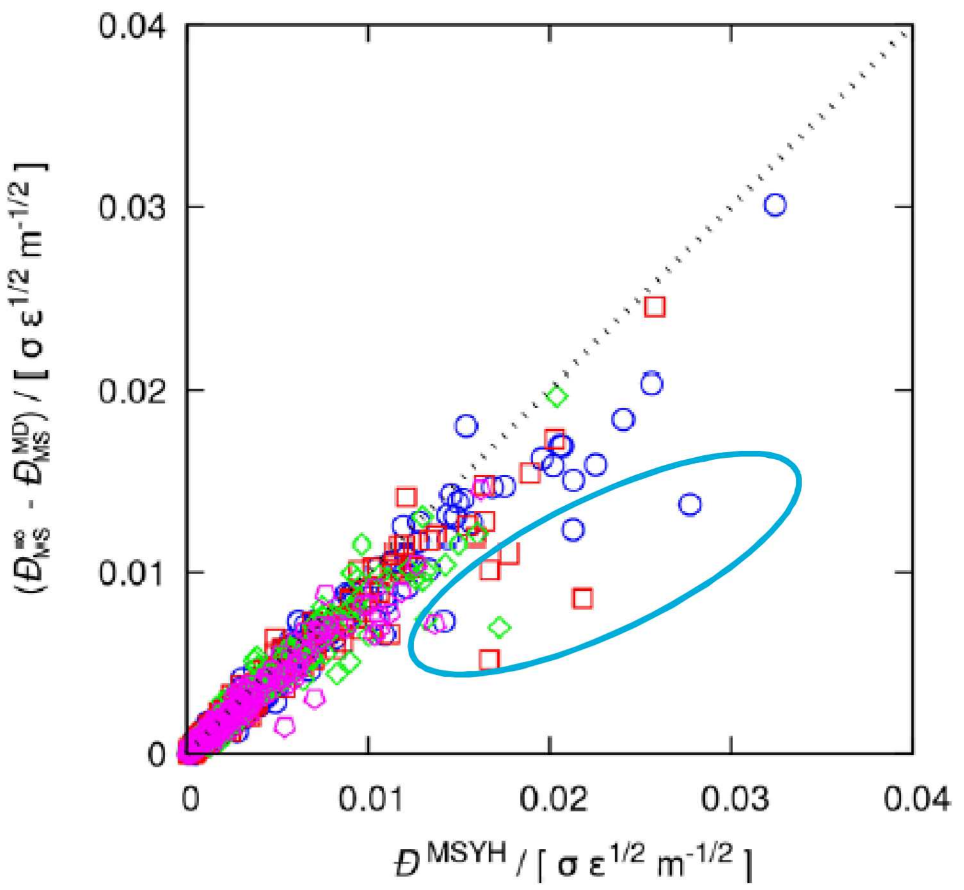


S. H. Jamali *et al.*, Finite-Size Effects of Binary Mutual Diffusion Coefficients from Molecular Dynamics. *J Chem Theory Comput* **14**, 2667-2677 (2018).

Maxwell-Stefan Diffusion Finite-Box Effects

- Jamali adapted the YH correction to be used for Maxwell-Stefan Diffusion
- The correction factor overpredicts by >100% for some datapoints

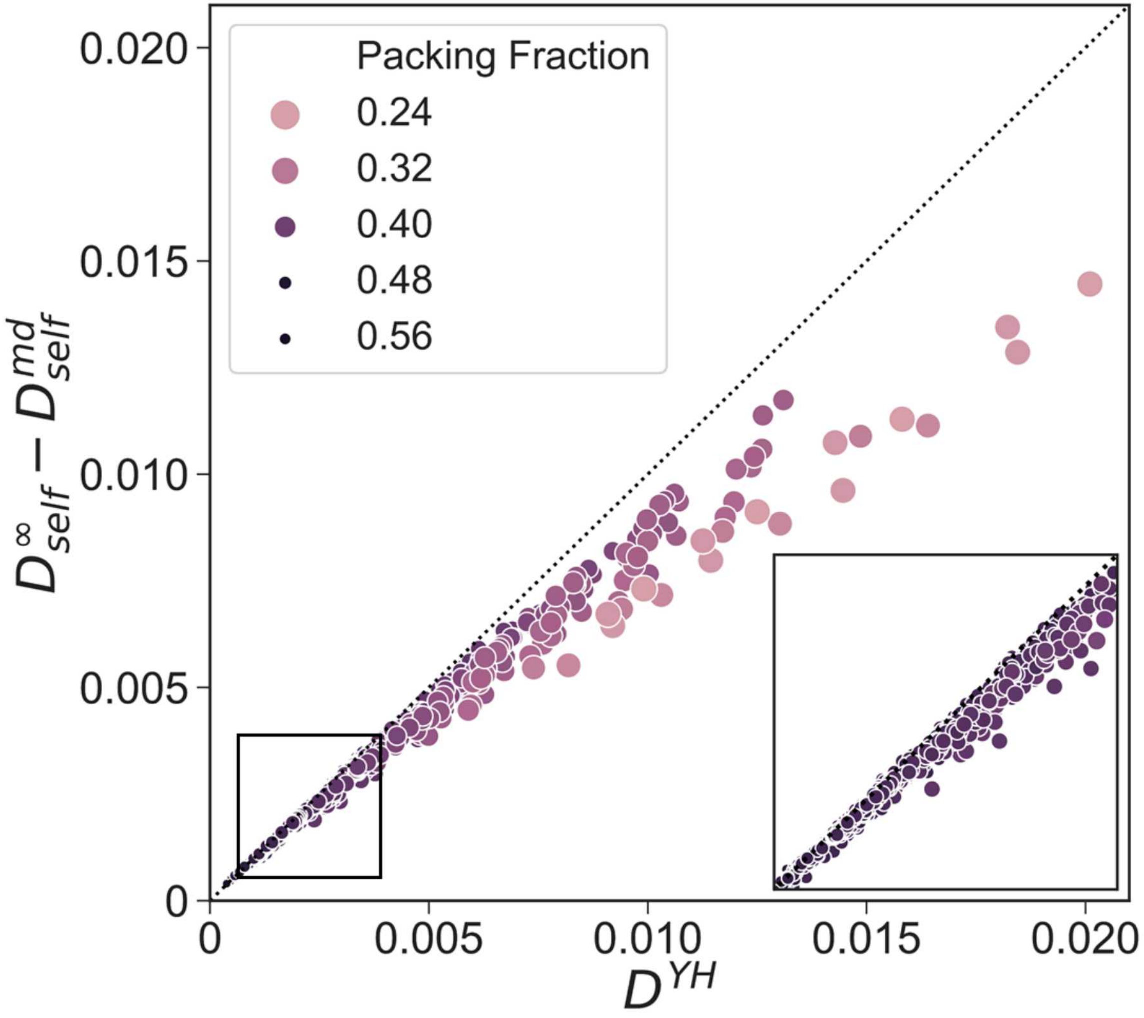
$$\text{YHMS: } D^{\infty} = D^{MD} + \frac{\xi k_B T}{6\pi\eta L\Gamma}$$

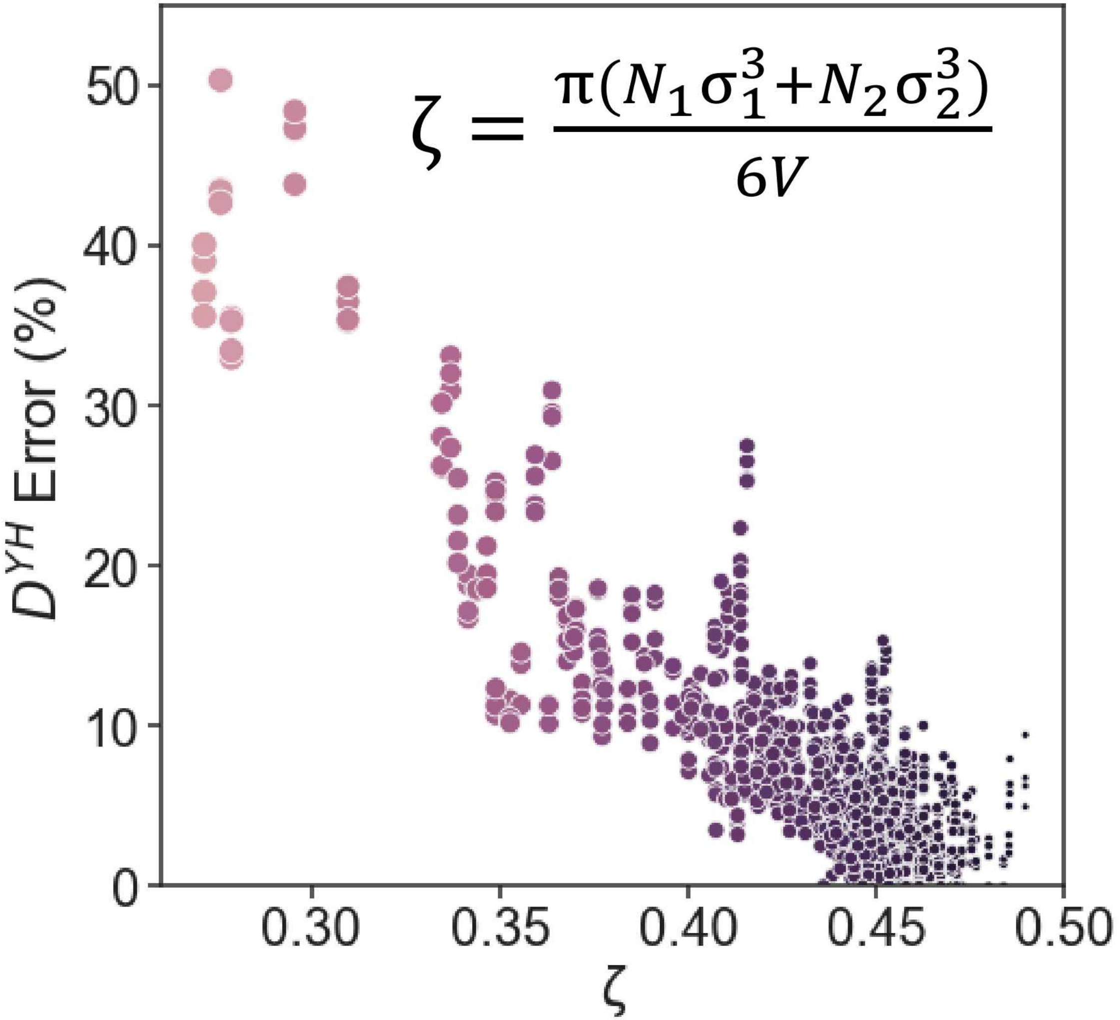


Can we use ML to create better finite-box size corrections?

S. H. Jamali *et al.*, Finite-Size Effects of Binary Mutual Diffusion Coefficients from Molecular Dynamics. *J Chem Theory Comput* **14**, 2667-2677 (2018).

Heyes 2007 Packing Fraction Empirical Relationship



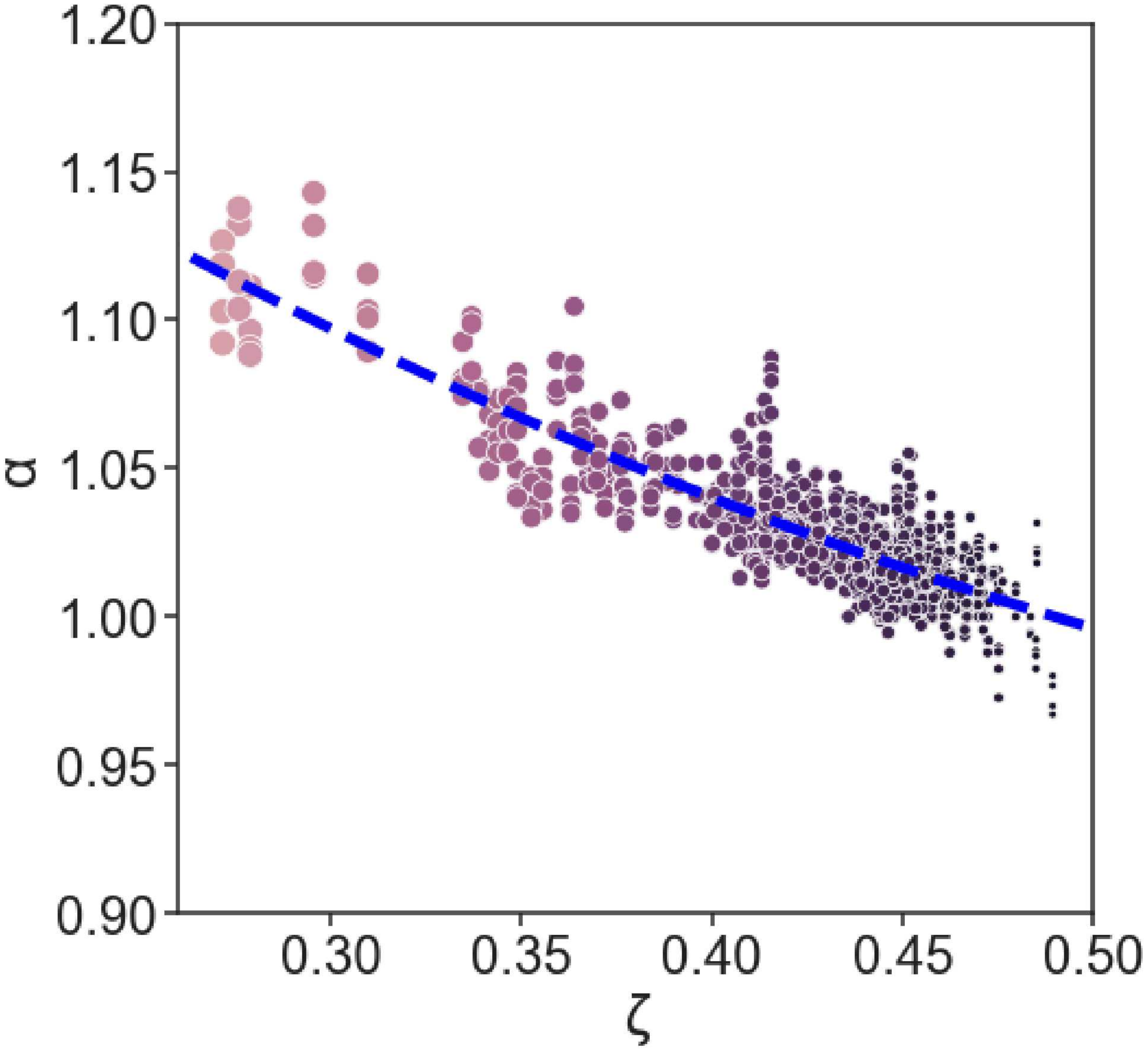


$$\zeta = \frac{\pi(N_1\sigma_1^3 + N_2\sigma_2^3)}{6V}$$

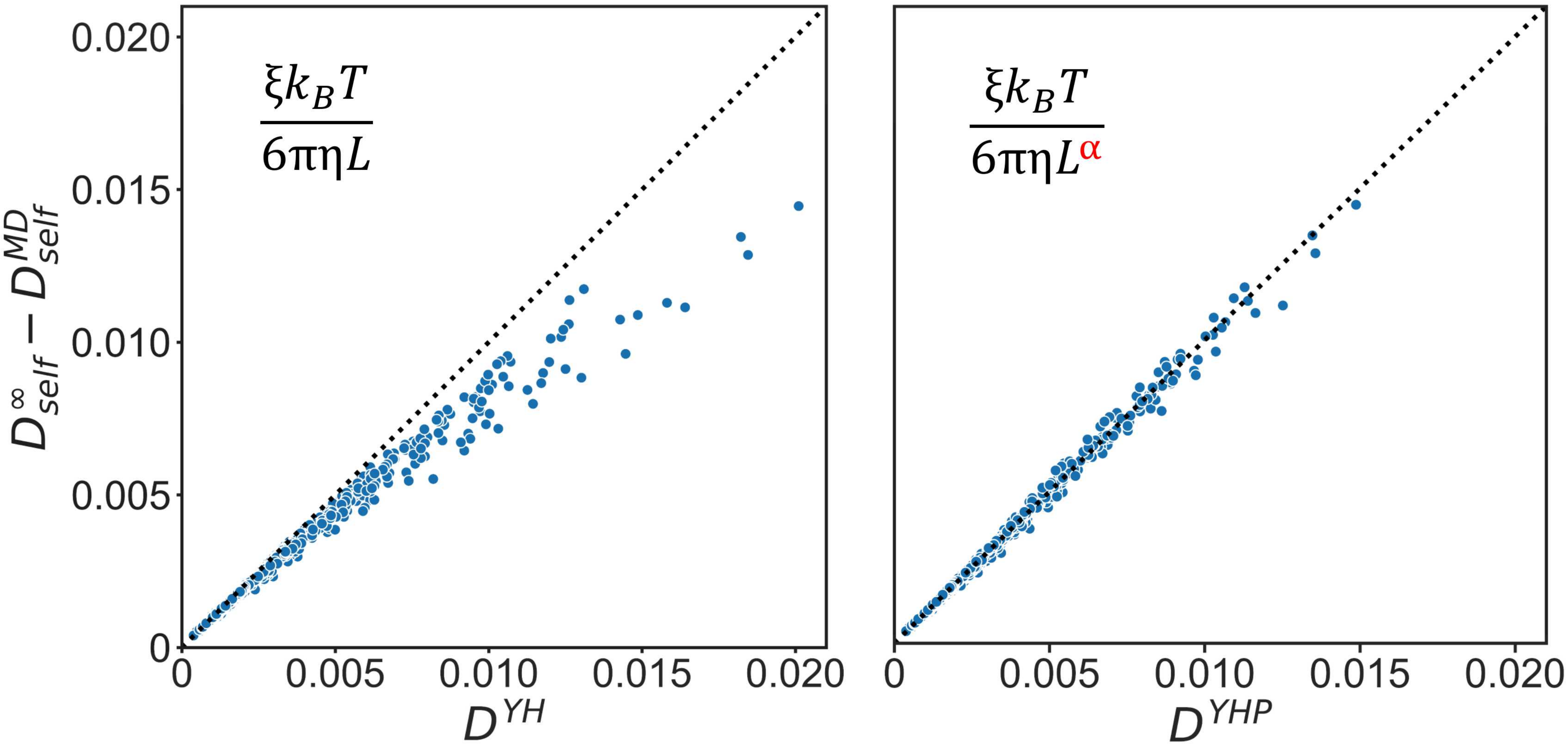
D. M. Heyes, M. J. Cass, J. G. Powles, W. A. Evans, Self-diffusion coefficient of the hard-sphere fluid: system size dependence and empirical correlations. *J Phys Chem B* **111**, 1455-1464 (2007).

Heyes 2007 Packing Fraction Empirical Relationship

- Add an empirically fit parameter based on packing fraction
- Heyes suggests: $D^{\infty} = D^{MD} + \frac{\xi k_B T}{6\pi\eta L^{\alpha}}$
- $\alpha(\zeta) = a_1 + a_2\,\zeta + a_3\,\zeta\,\ldots$
- There is no physical argument for this relationship

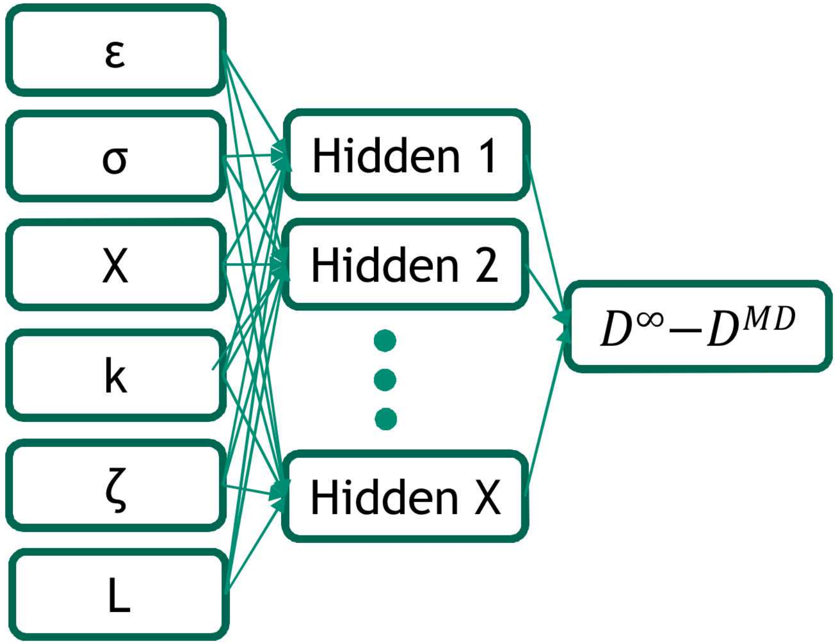


Heyes 2007 Packing Fraction Empirical Relationship



ANN SD Correction Prediction Option 1

Train ANN to directly predict correction factor: $D^{\infty} - D^{MD}$



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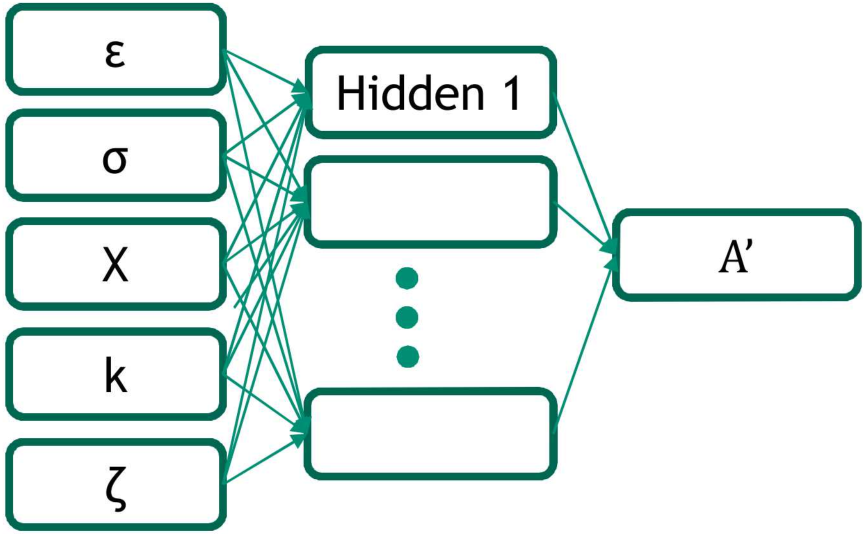
ANN SD Correction Prediction Option 2

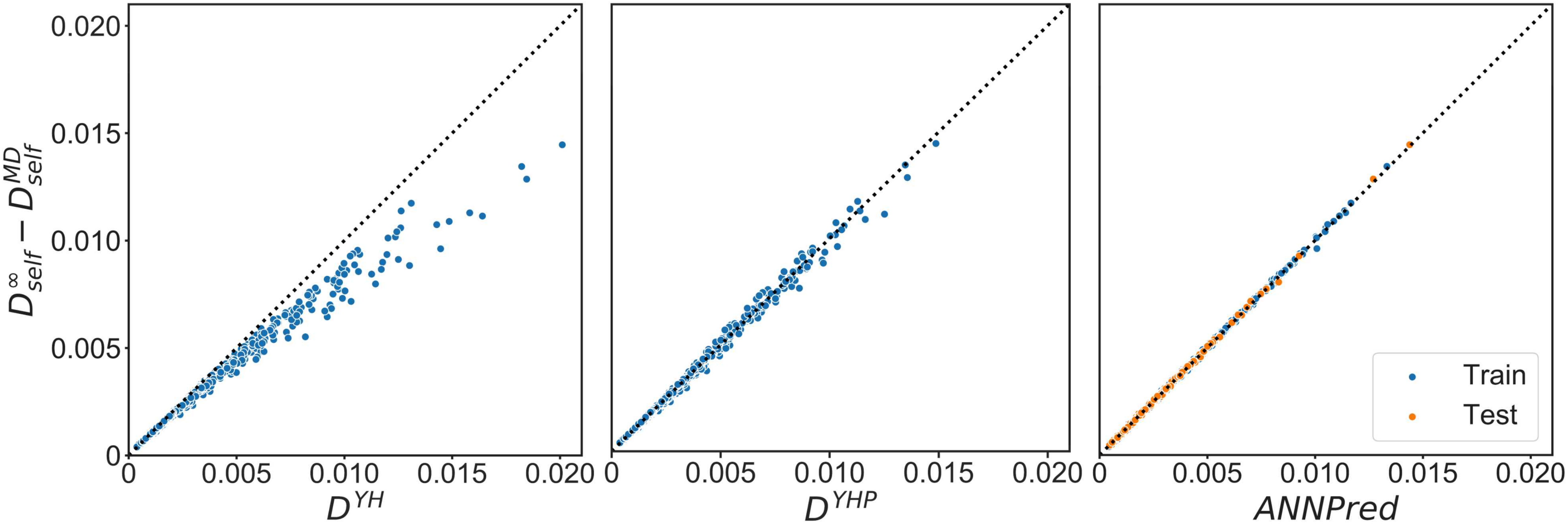
- Train ANN to predict A'
- Divide A' by L to get $D^{\infty} - D^{MD}$
- Preserving the L^{-1} scaling



$$D^{\infty} - D^{MD} = \frac{\xi k_B T}{6\pi\eta L}$$

$$D^{\infty} - D^{MD} = A'(\zeta) \times L^{-1}$$

ANN Correction Prediction

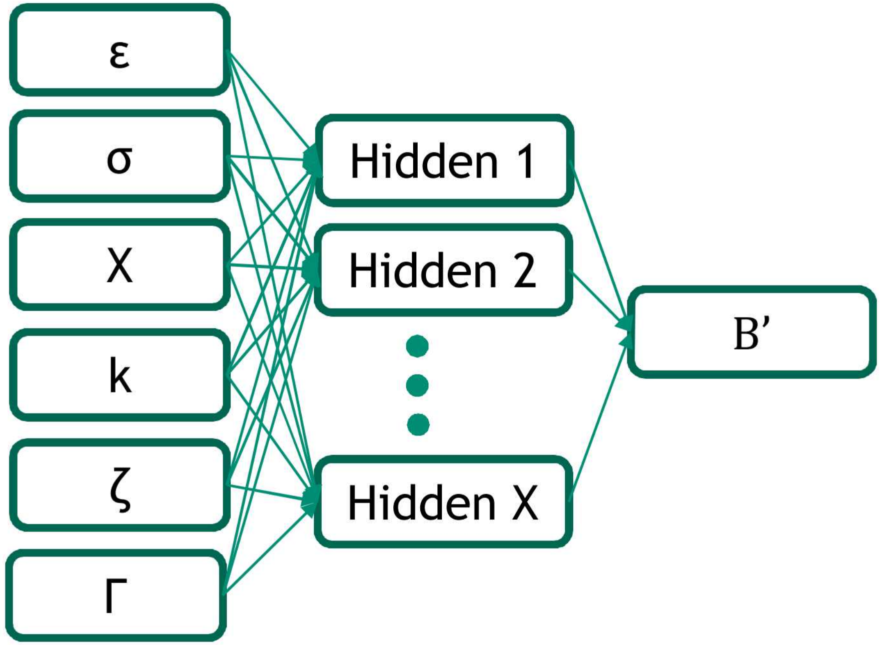


~25x reduction in MSE

~175x reduction in MSE

ANN MS Correction Prediction Option 2

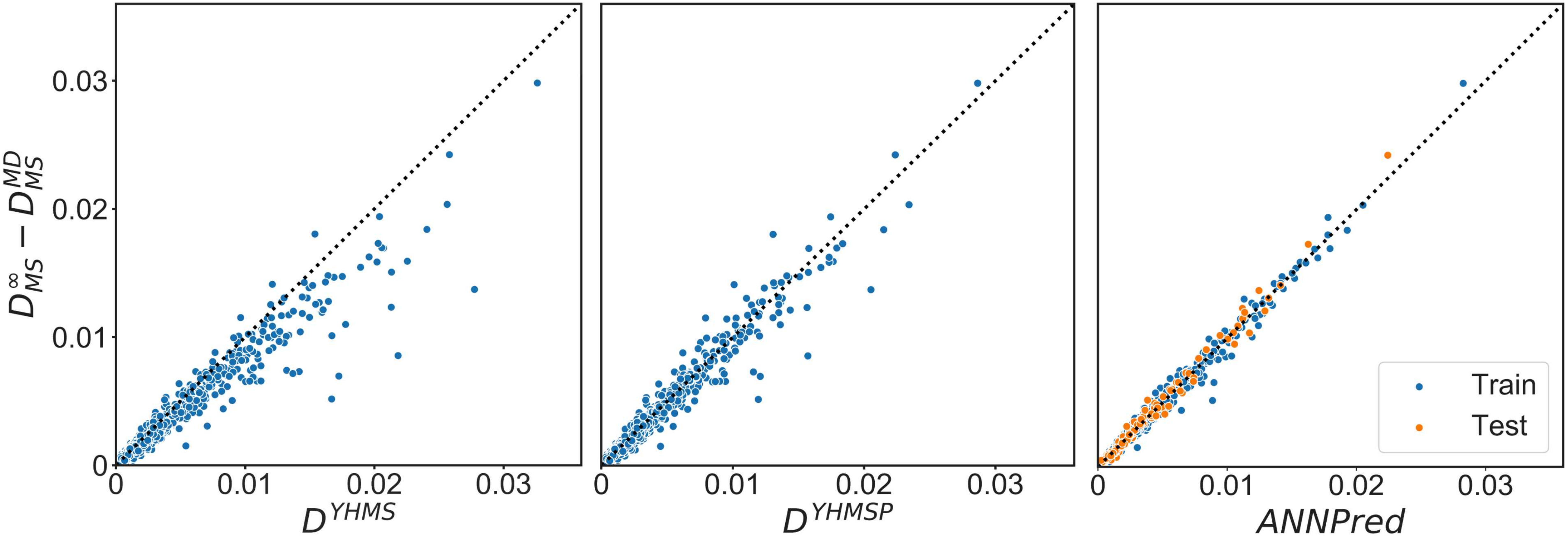
- Γ is a measure of interaction between species in a multicomponent mixture



$$D^{\infty} - D^{MD} = \frac{\xi k_B T}{6\pi\eta L\Gamma}$$

$$D^{\infty} - D^{MD} = B'(\zeta) \times L^{-1}$$

ANN Correction Prediction MS

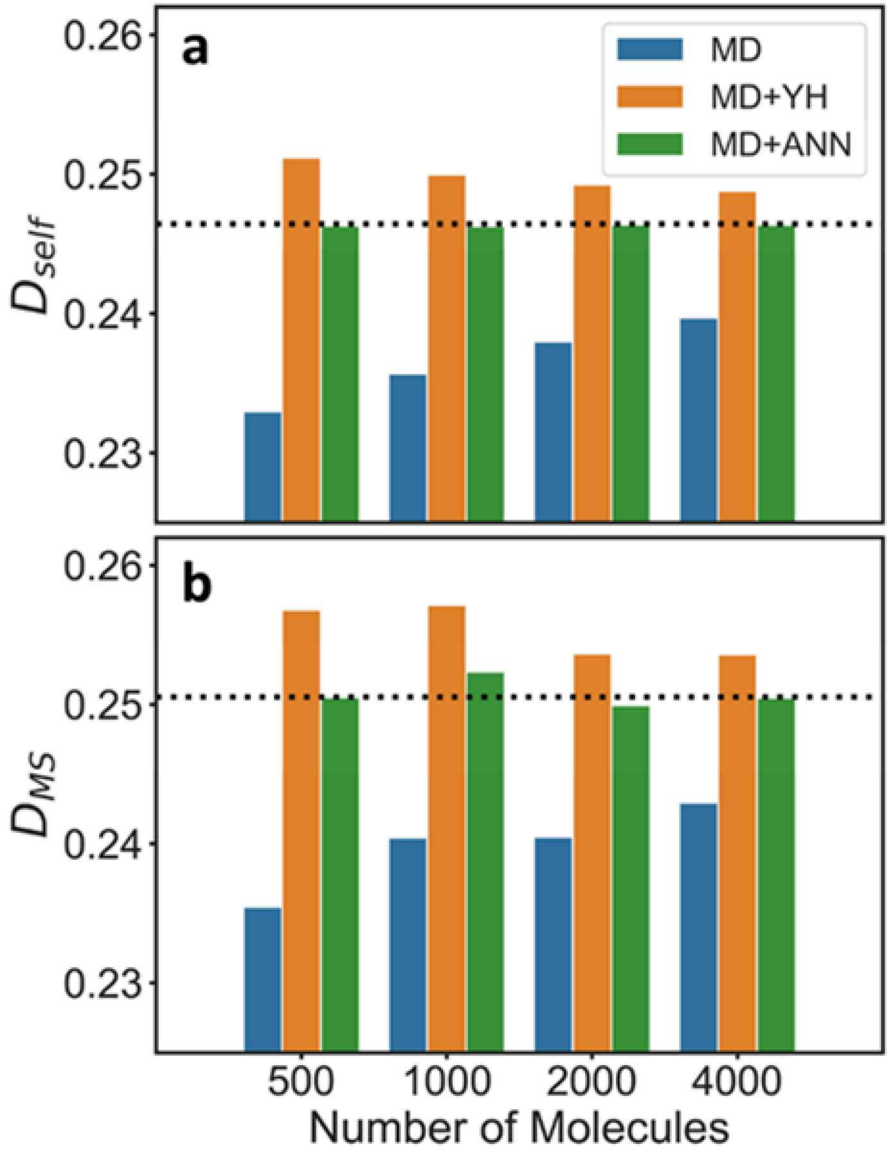


~3x reduction in MSE

~16x reduction in MSE

Diffusion Coefficient Predictions

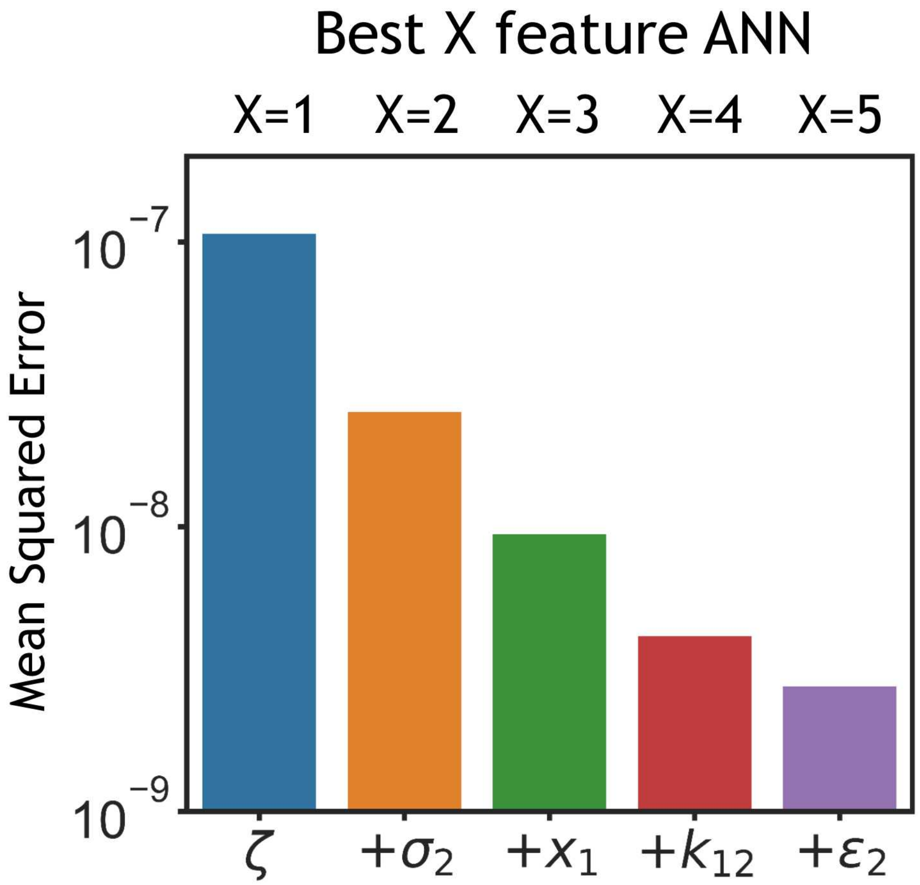
- MD alone under predicts the diffusion coefficient
- Adding the YH correction always overpredicts for the binary LJ fluids
- ANN performs consistently



ANN Feature Importance

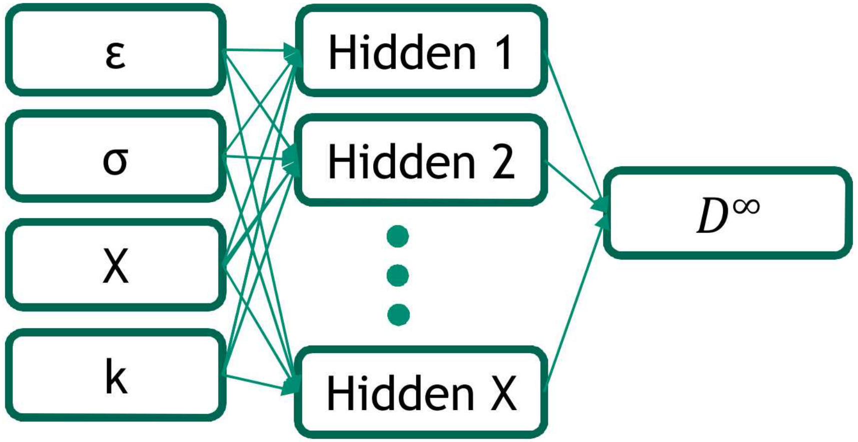
$$\text{ANN: } D^{\infty} - D^{MD} = A'(\zeta) \times L^{-1}$$

- Iteratively added features to the ANN to determine relative importance
- The error drops for every feature added
- For binary LJ fluids, A′ is a function of the LJ parameters



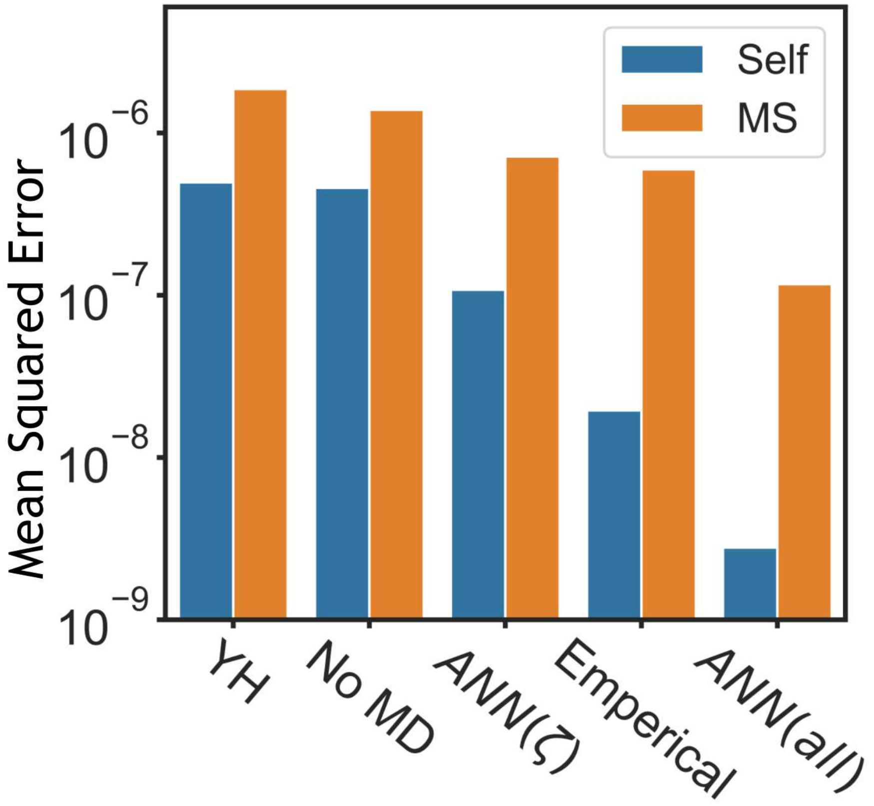
Replace MD Altogether?

- Directly predict D^{∞}, as opposed to calculating a value correction to the MD diffusion coefficient
- Limited to LJ parameters – no MD simulated variables like packing fraction
- Better prediction than YH corrected MD simulations
- Worse than ANN corrected MD simulations



Combination of molecular dynamics simulation and machine learning produces the best results

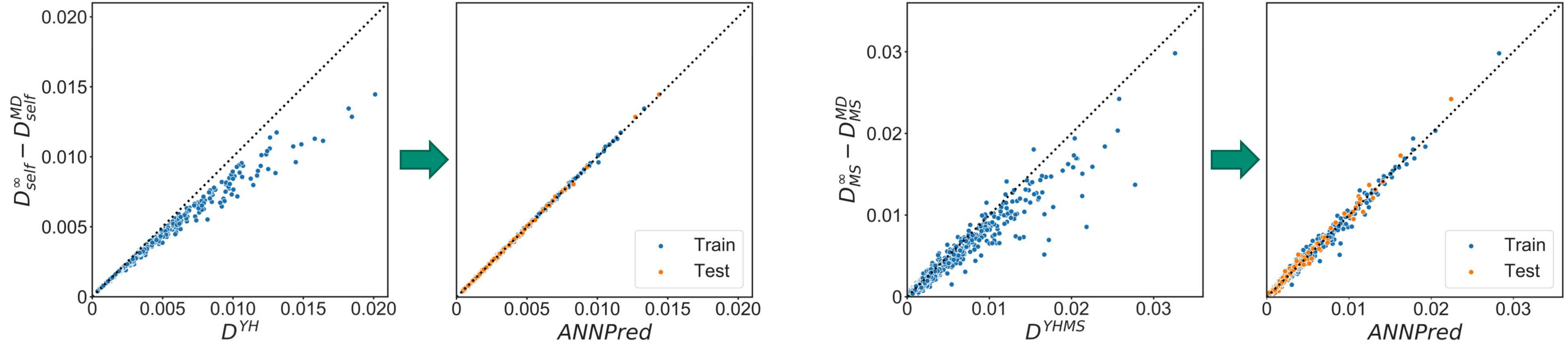
Overall Performance



Error reduction of over an order of magnitude for MS and over two orders of magnitude for SD

Summary

- We used machine learning, not to replicate the current state of the art finite-box size correction, but to improve it!
- The YH correction for single component mixtures is not sufficient for binary LJ mixtures
- A′/B′ are functions of all the LJ parameters $$D^{\infty} - D^{MD} = A'(\zeta) \times L^{-1}$$
- The L^{-1} dependence is important



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