Wide Ranging Ionic Transport Coefficients for High-Energy-Density Applications

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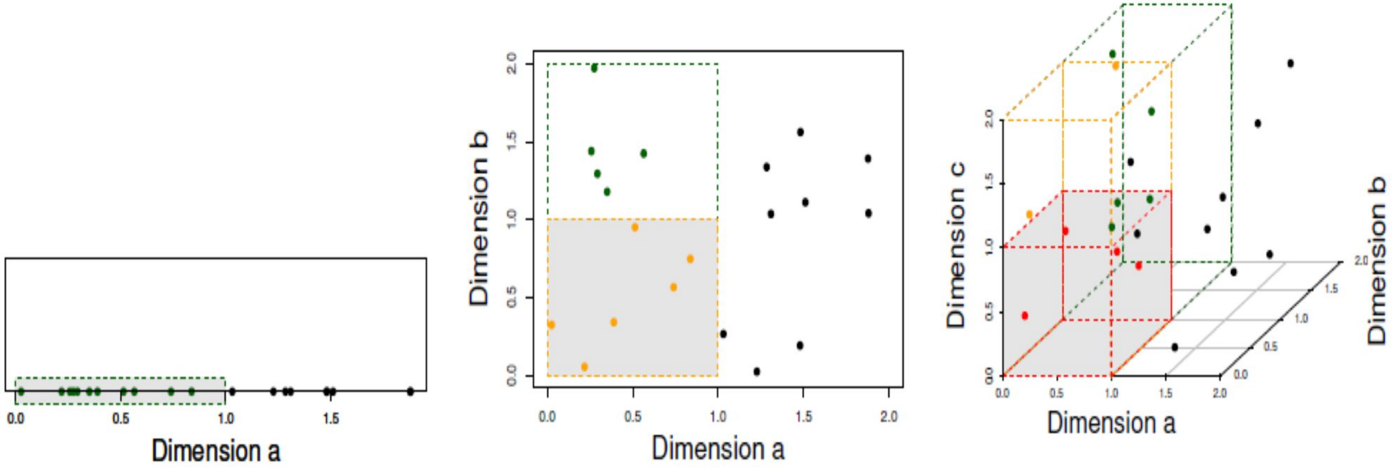
Goal: Generate Data with Data Science in Mind

1. Generate the most accurate data with minimal statistical noise
2. Use existing low- and high-fidelity data optimally
3. Identify where new data is needed

The Curse of Dimensionality Results in Sparse Datasets

Question: How does some output f depend on its inputs?

You have the resource to compute 20 points of data.



(a) (b) (c)

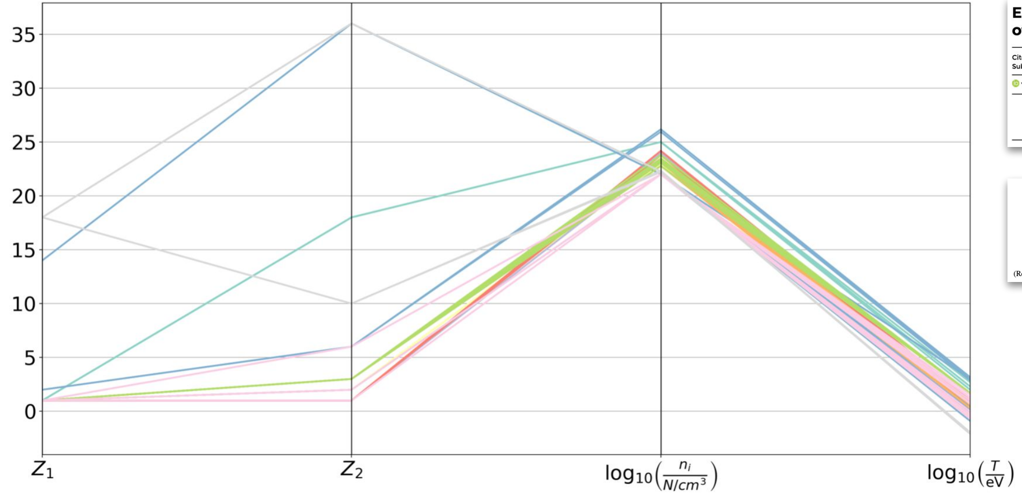
6 points in unit bin

4 points in unit bin

1. Rajput, Dharmveer Singh, Pramod Kumar Singh, and Mahua Bhattacharya. "IQRAM: a high dimensional data clustering technique." *International Journal of Knowledge Engineering and Data Mining* 2.2-3 (2012): 117-136.

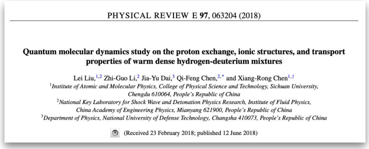
Curse of Dimensionality in Binary Mixture Simulations

- 9 articles were selected from the literature.
- A dataset of 99 interdiffusion coefficients for binary mixtures was created.



Each color denotes a different article.

86% of data contained simulations of H.


PHYSICAL REVIEW E **97**, 063204 (2018)

Quantum molecular dynamics study on the proton exchange, ionic structures, and transport properties of warm dense hydrogen-deuterium mixtures

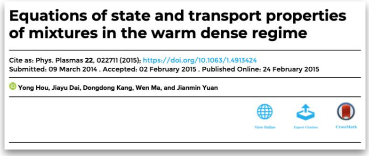
Lei Liu,[1,2] Zhi-Guo Li,[2] Jia-Yu Dai,[3] Qi-Feng Chen,[2,*] and Xiang-Rong Chen[1,†]
[1]*Institute of Atomic and Molecular Physics, College of Physical Science and Technology, Sichuan University, Chengdu 610064, People's Republic of China*
[2]*National Key Laboratory for Shock Wave and Detonation Physics Research, Institute of Fluid Physics, China Academy of Engineering Physics, Mianyang 621900, People's Republic of China*
[3]*Department of Physics, National University of Defense Technology, Changsha 410073, People's Republic of China*

(Received 23 February 2018; published 12 June 2018)


PHYSICAL REVIEW E **90**, 023104 (2014)

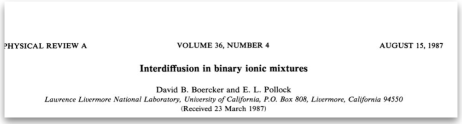
Diffusivity in asymmetric Yukawa ionic mixtures in dense plasmas

Tomorr Haxhimali,* Robert E. Rudd, William H. Cabot, and Frank R. Graziani
Lawrence Livermore National Laboratory, Livermore, California 94550, USA
(Received 28 May 2014; published 21 August 2014)


Equations of state and transport properties of mixtures in the warm dense regime

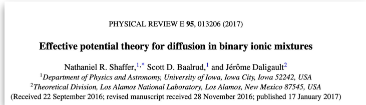
Cite as: Phys. Plasmas **22**, 022711 (2015); https://doi.org/10.1063/1.4913424
Submitted: 09 March 2014 . Accepted: 02 February 2015 . Published Online: 24 February 2015

Yong Hou, Jiayu Dai, Dongdong Kang, Wen Ma, and Jianmin Yuan


PHYSICAL REVIEW A VOLUME 36, NUMBER 4 AUGUST 15, 1987

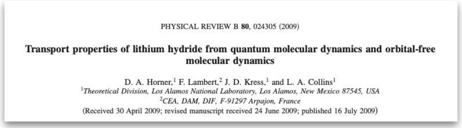
Interdiffusion in binary ionic mixtures

David B. Boercker and E. L. Pollock
Lawrence Livermore National Laboratory, University of California, P.O. Box 808, Livermore, California 94550
(Received 23 March 1987)


PHYSICAL REVIEW E **95**, 013206 (2017)

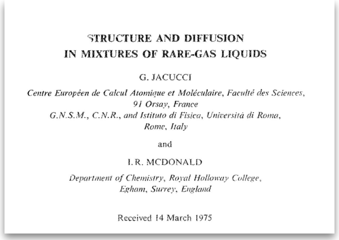
Effective potential theory for diffusion in binary ionic mixtures

Nathaniel R. Shaffer,[1,*] Scott D. Baalrud,[1] and Jérôme Daligault[2]
[1]*Department of Physics and Astronomy, University of Iowa, Iowa City, Iowa 52242, USA*
[2]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
(Received 22 September 2016; revised manuscript received 28 November 2016; published 17 January 2017)


PHYSICAL REVIEW B **80**, 024305 (2009)

Transport properties of lithium hydride from quantum molecular dynamics and orbital-free molecular dynamics

D. A. Horner,[1] F. Lambert,[2] J. D. Kress,[1] and L. A. Collins[1]
[1]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[2]*CEA, DAM, DIF, F-91297 Arpajon, France*
(Received 30 April 2009; revised manuscript received 24 June 2009; published 16 July 2009)


STRUCTURE AND DIFFUSION IN MIXTURES OF RARE-GAS LIQUIDS

G. JACUCCI
Centre Européen de Calcul Atomique et Moléculaire, Faculté des Sciences, 91 Orsay, France
G.N.S.M., C.N.R., and Istituto di Fisica, Università di Roma, Rome, Italy

and

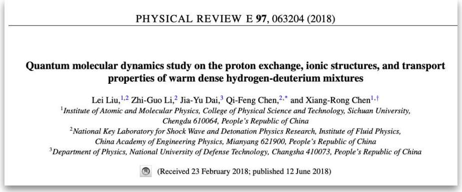
I. R. MCDONALD
Department of Chemistry, Royal Holloway College, Egham, Surrey, England

Received 14 March 1975


PHYSICAL REVIEW E **82**, 036404 (2010)

Viscosity and mutual diffusion of deuterium-tritium mixtures in the warm-dense-matter regime

J. D. Kress,[1] James S. Cohen,[1] D. A. Horner,[1] F. Lambert,[2] and L. A. Collins[1]
[1]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[2]*CEA, DAM, DIF, F-91297 Arpajon, France*
(Received 7 June 2010; published 14 September 2010)


PHYSICAL REVIEW E **97**, 063204 (2018)

Quantum molecular dynamics study on the proton exchange, ionic structures, and transport properties of warm dense hydrogen-deuterium mixtures

Lei Liu,[1,2] Zhi-Guo Li,[2] Jia-Yu Dai,[3] Qi-Feng Chen,[2,*] and Xiang-Rong Chen[1,†]
[1]*Institute of Atomic and Molecular Physics, College of Physical Science and Technology, Sichuan University, Chengdu 610064, People's Republic of China*
[2]*National Key Laboratory for Shock Wave and Detonation Physics Research, Institute of Fluid Physics, China Academy of Engineering Physics, Mianyang 621900, People's Republic of China*
[3]*Department of Physics, National University of Defense Technology, Changsha 410073, People's Republic of China*

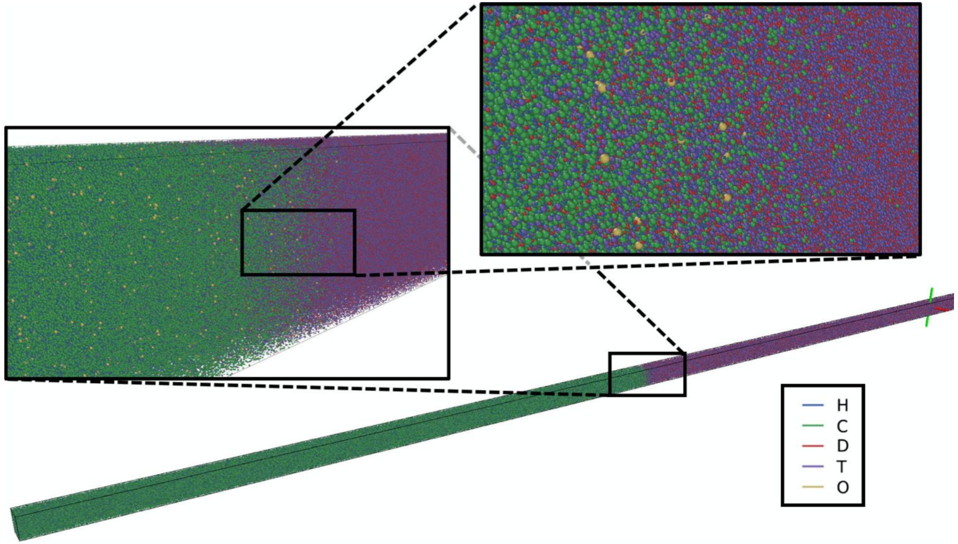
(Received 23 February 2018; published 12 June 2018)

Curse of Dimensionality in Multispecies Simulations

For a large-scale, non-homogeneous, molecular dynamics simulation of 5 species we have:

5 species + 5 density fields + 5 temperatures fields ≈ 15 dimensions

3 points in each dimension:14,348,907 data points



1. Stanton, L. G., J. N. Glosli, and M. S. Murillo. "Multiscale molecular dynamics model for heterogeneous charged systems." *Physical Review X* 8.2 (2018): 021044.

Generating Large Datasets Demands Substantial Resource

Few data points

Number of Data Points

Many data points

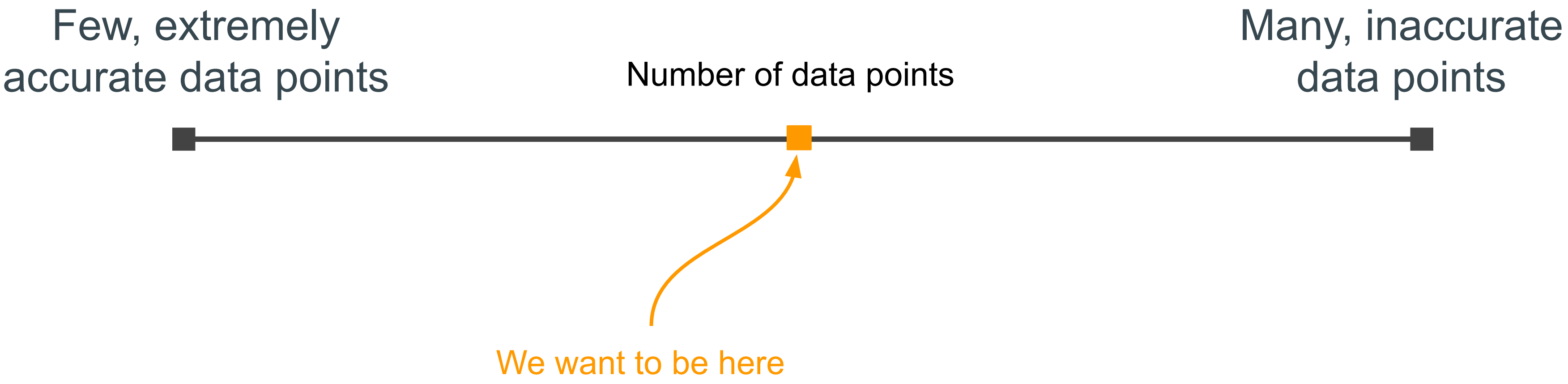
- Demands little memory
- Fast to access and create
- Curse of dimensionality

We want to be here

- Solves the curse of dimensionality
- Demands large resource to store and access
- Computationally expensive to generate

Resource Limitations Reduce Dataset Accuracy

Imagine that you have only 1 month to generate data from simulations, how should we proceed?



Goal: Generate Data with Data Science in Mind

1. Generate the most accurate data with minimal statistical noise
2. Use existing low- and high-fidelity data optimally
3. Identify where new data is needed

We aim to accomplish these goals in the setting of ionic transport coefficients.

Efficacy of the radial pair potential approximation for molecular dynamics simulations of dense plasmas F

Cite as: Phys. Plasmas **28**, 032706 (2021); https://doi.org/10.1063/5.0040062
Submitted: 09 December 2020 . Accepted: 05 February 2021 . Published Online: 11 March 2021

Lucas J. Stanek, Raymond C. Clay, M. W. C. Dharma-wardana, Mitchell A. Wood, Kristian R. C. Beckwith, and Michael S. Murillo

COLLECTIONS

F This paper was selected as Featured

What is the "best" force law to use for molecular dynamics simulation?

Approximate Force Laws

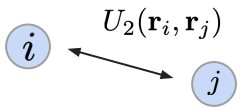
$$U_{tot} = U_N(\mathbf{r}_1, \mathbf{r}_2, \cdots, \mathbf{r}_N)$$

Potential energy surface

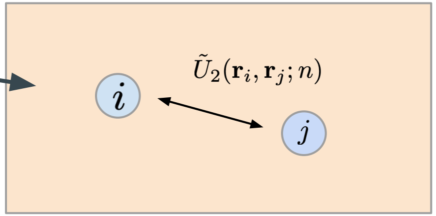
$$U_{tot} = \underbrace{\sum_i^N U_1(\mathbf{r}_i) + \sum_{i,j}^N U_2(\mathbf{r}_i, \mathbf{r}_j)}_{\text{Two-body potential}} + \sum_{i,j,k}^N U_3(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) + \cdots$$

$$U_{tot} \approx \sum_{i,j}^N \tilde{U}_2(\mathbf{r}_i, \mathbf{r}_j; n)$$

"Effective" pair-interaction potential



What is the best way to determine the "effectiveness"?



Force Laws Vary in Fidelity and Computation Cost

High Computation Cost

Low Computation Cost

Kohn-Sham Density Functional Theory MD (KSMD) — *N*-body

Spectral Neighborhood Analysis Potential (SNAP) — quad-interaction potential

Force-Matched Pair Potential (FM)

Neutral Pseudo Atom Pair Potential (NPA)

Thomas-Fermi Yukawa Pair Potential (TFY)

pair-potential

Few Assumptions Made

Many Assumptions Made

Effective Pair Interaction Potentials from Perturbation Theory

Mean-ionization

Coulomb interaction

Electron-ion pseudo potential

Susceptibility

$$u(k) = \langle Z \rangle^2 u_C(k) + |u_{ei}(k)|^2 \chi(k)$$

NPA model

$$u_{ei} \approx \frac{-4\pi \langle Z \rangle e^2}{k^2}$$

$$\chi(k) \approx \chi_{TF}(k \to 0)$$

Calculates $u(k)$ by assuming:

- The mean ionization and pseudopotential are computed from a Kohn-Sham Mermin approach.
- The susceptibility is given by the Lindhard function with local field corrections.

$$u^{TFY}(r) = \frac{\langle Z \rangle^2 e^2}{r} e^{-r/\lambda_{TF}}$$

Thomas-Fermi Yukawa (TFY) effective pair potential

Two parameter, short range, pair interactions

1.J. Porter, N. Ashcroft, and G. Chester, "Pair potentials for simple metallic systems: Beyond linear response," Phys. Rev. B 81, 224113 (2010).

Force Laws Vary in Fidelity and Computation Cost

Kohn-Sham Density Functional Theory MD (KSMD)	*N*-body
Spectral Neighborhood Analysis Potential (SNAP)	quad-interaction potential
Force-Matched Pair Potential (FM)	pair-potential
Neutral Pseudo Atom Pair Potential (NPA)	
Thomas-Fermi Yukawa Pair Potential (TFY)	

Force-Matched Pair Potential: Minimizing a Loss Function

$$L(\zeta) = \sum_{k=1}^{K} w_k \left(F_k(\zeta) - F_k^0\right)^2$$

Parameters or "shape" of the pair potential

Force law we wish to determine

Reference force data

$$F^{yuk}(r;\zeta) = -\nabla\left(\frac{\zeta_1}{r} e^{-r\zeta_2}\right)$$

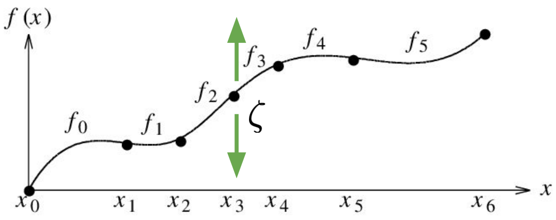


Figure 1. Cubic spline.

1: Ercolessi and Adams, "Interatomic potentials from first-principles calculations: the force-matching method" *EPL (Europhysics Letters)* 26, 583 (1994).

2: Brommer and Gähler, "Potfit: effective potentials from ab initio data", *Modelling and Simulation in Materials Science and Engineering* 15, 295 (2007).

3: Wolberg, G. and Itzik Alfy. "Monotonic cubic spline interpolation." *1999 Proceedings Computer Graphics International* (1999): 188-195.

Force Laws Vary in Fidelity and Computation Cost

Kohn-Sham Density Functional Theory MD (KSMD) — *N*-body

Spectral Neighborhood Analysis Potential (SNAP) — quad-interaction potential

Force-Matched Pair Potential (FM)

Neutral Pseudo Atom Pair Potential (NPA)

Thomas-Fermi Yukawa Pair Potential (TFY)

pair-potential

Four-body interactions via SNAP

SNAP regresses against KSMD data but computes an effective quad-interaction potential.

The SNAP will be selectively employed to assess importance of interactions beyond the pair-potential.

1. A. Thompson, L. Swiler, C. Trott, S. Foiles, and G. Tucker, “Spectral neighbor analysis method for automated generation of quantum-accurate interatomic potentials,” J. Comput. Phys. 285, 316 (2015).

Force Laws Vary in Fidelity and Computation Cost

Kohn-Sham Density Functional Theory MD (KSMD)	*N*-body
Spectral Neighborhood Analysis Potential (SNAP)	quad-interaction potential
Force-Matched Pair Potential (FM)	pair-potential
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N-body interactions via Kohn-Sham DFT MD

$$n_e(\mathbf{r}) = \sum_i f_i(T)|\phi_i(\mathbf{r})|^2$$

$$\left(-\frac{1}{2}\nabla^2 + v_{eff}(\mathbf{r})\right)\phi_i(\mathbf{r}) = \epsilon_i\phi_i(\mathbf{r})$$

Schrodinger's equation for orbitals

$$v_{eff}(\mathbf{r}) = V_{ext}(\mathbf{r}) + \int d\mathbf{r}' \left[\frac{n_e(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + \frac{\delta E_{xc}[n_e]}{\delta n_e(\mathbf{r})}\right]$$

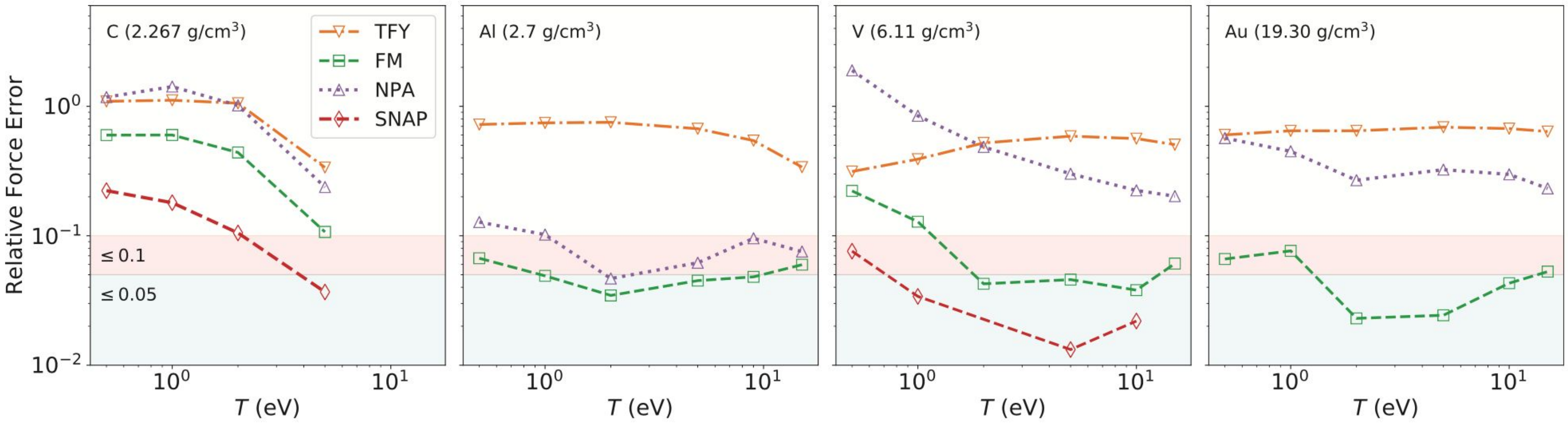
Effective potential using the PBE functional and PAW pseudopotential.

Force Laws Vary in Fidelity and Computation Cost

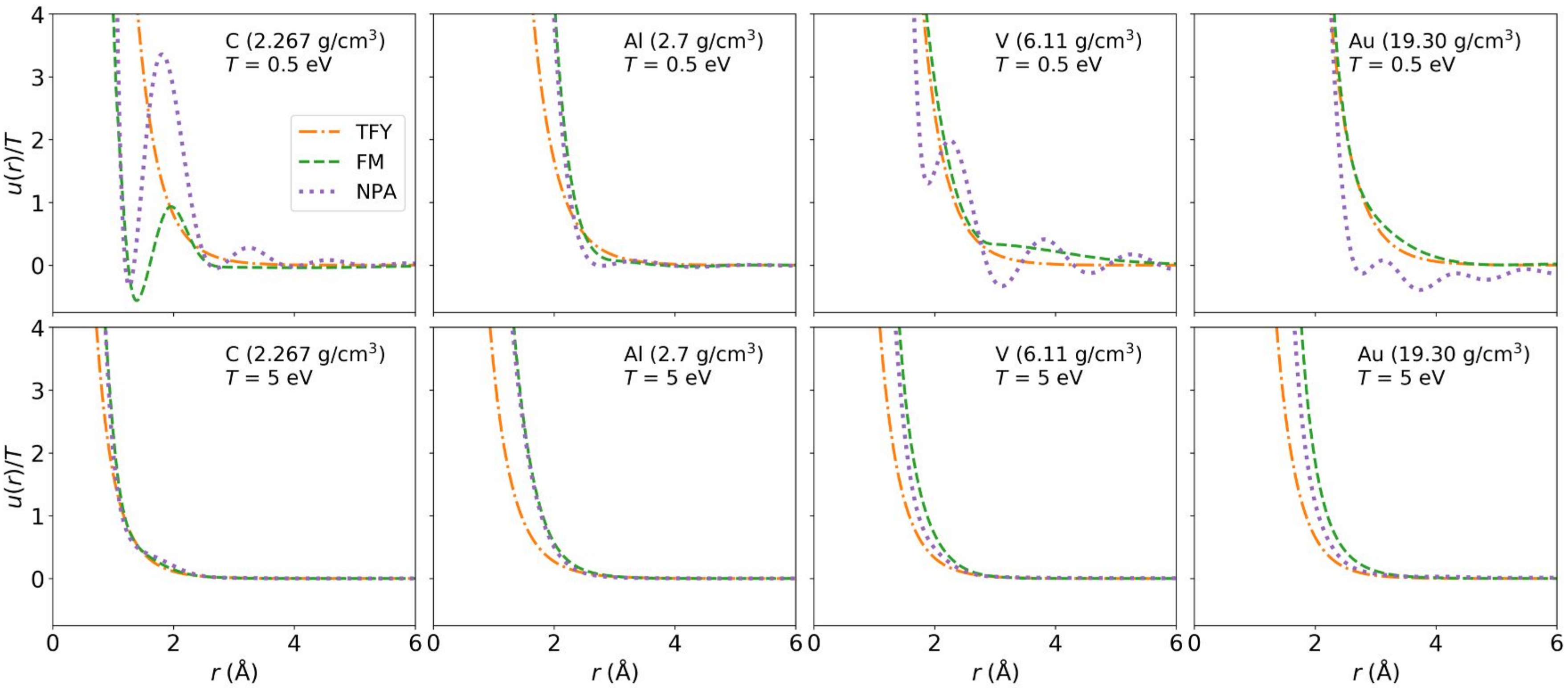
Performed MD for 7 elements (insulators, noble gas, metals) and compared results

- Kohn-Sham Density Functional Theory MD (KSMD)
- Spectral Neighborhood Analysis Potential (SNAP)
- Force-Matched Pair Potential (FM)
- Neutral Pseudo Atom Pair Potential (NPA)
- Thomas-Fermi Yukawa Pair Potential (TFY)

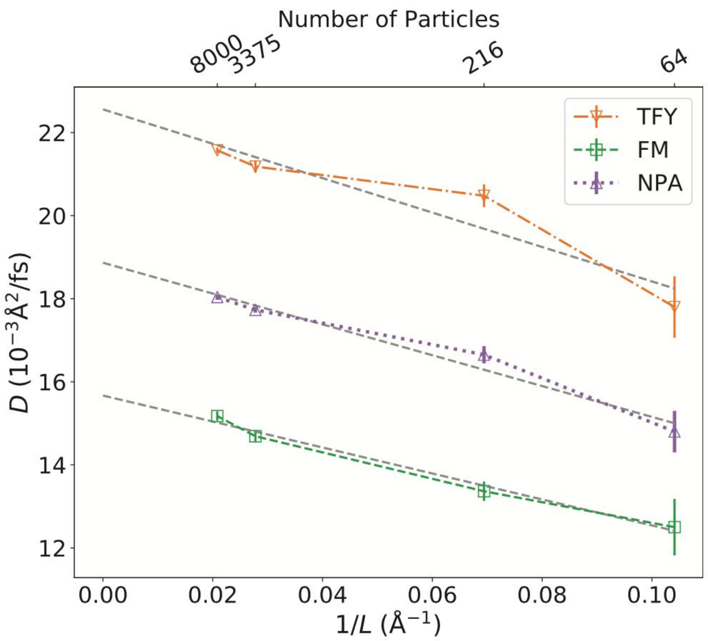
Force Error Analysis: Comparing with KSMD



Representation of Pair Interaction Potentials



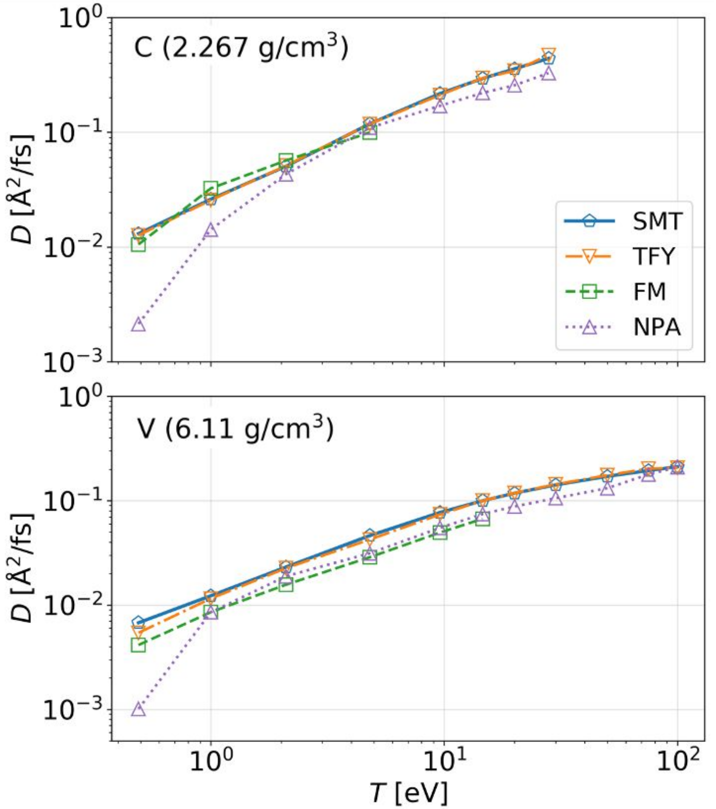
Finite-Size Effects Cause Significant Statistical Errors



Relative Model Error: ~35%

Error from finite-size effects: ~20%

Analytic Models Avoid Finite Size Effects



Stanton-Murillo Transport (SMT) Model

PHYSICAL REVIEW E **93**, 043203 (2016)

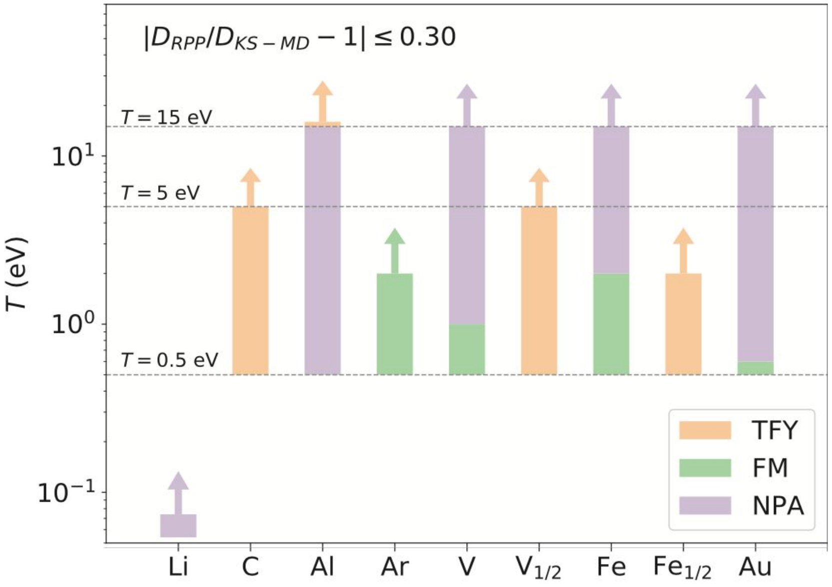
Ionic transport in high-energy-density matter

Liam G. Stanton[1,*] and Michael S. Murillo[2,*]
[1] *Center for Applied Scientific Computing, Lawrence Livermore National Laboratory, Livermore, California 94550, USA*
[2] *Computational Physics and Methods Group, MS D413, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
(Received 16 November 2015; published 8 April 2016)

Temperature Dependent Efficacy Boundary of Pair Potentials

Dataset:

- 7 elements (Z = 3 to 79)
- Solid and half-density cases

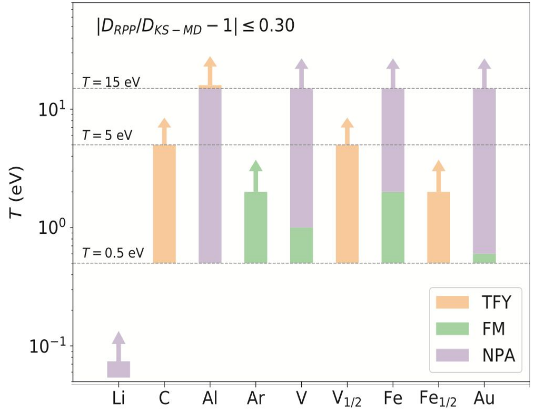


Provides a rule for creating the largest, most accurate dataset.

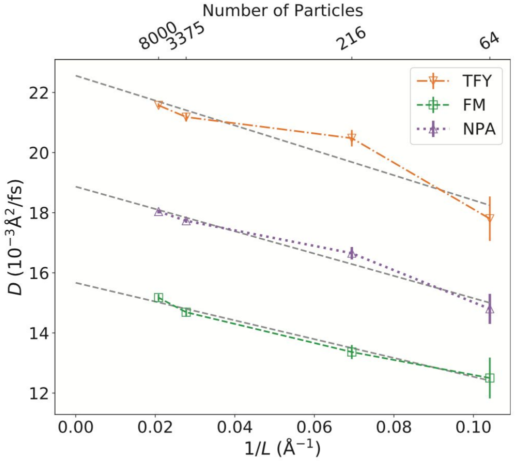
Goal: Generate Data with Data Science in Mind

1. Generate the most accurate data with minimal statistical noise
2. Use existing low- and high-fidelity data optimally
3. Identify where new data is needed

What model should we pick?



How can we increase the accuracy of our data?



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Multi-fidelity Interpolation of Sparse High-fidelity Data Available in Disparate Physical Regimes

Lucas J. Stanek,[1,*] Shaunak D. Bopardikar,[2,†] and Michael S. Murillo[1,‡]

[1] *Department of Computational Mathematics, Science and Engineering, Michigan State University, MI, USA*
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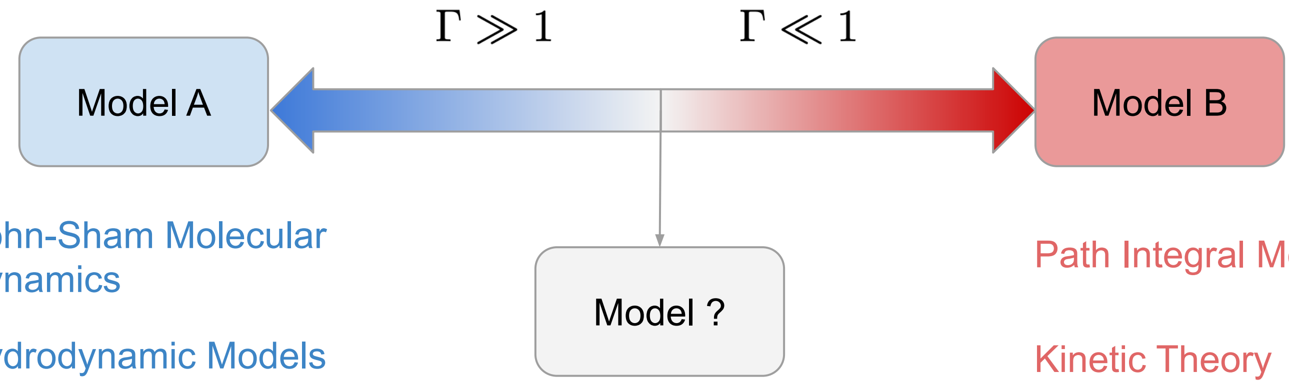
How can we use machine learning to extend the range of existing high-fidelity data?

Physical Models in Disparate Regimes

Γ Coulomb Coupling Parameter



Kohn-Sham Molecular Dynamics

Hydrodynamic Models

⋮

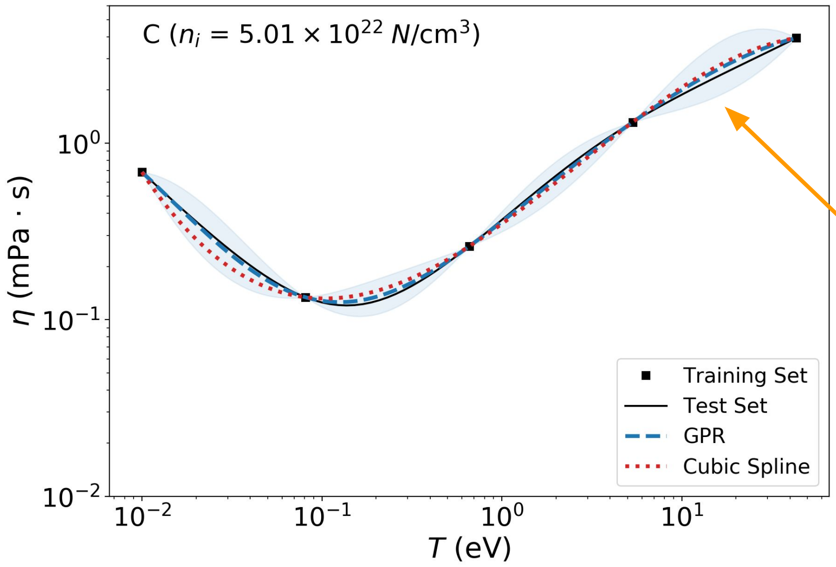
An interpolation problem

Path Integral Monte Carlo

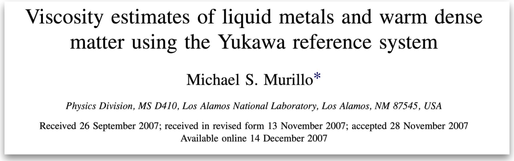
Kinetic Theory

⋮

Interpolation with Gaussian Process Regression (GPR)



Test and training data generated from the Yukawa viscosity model (YVM)



Viscosity estimates of liquid metals and warm dense matter using the Yukawa reference system

Michael S. Murillo*

Physics Division, MS D410, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

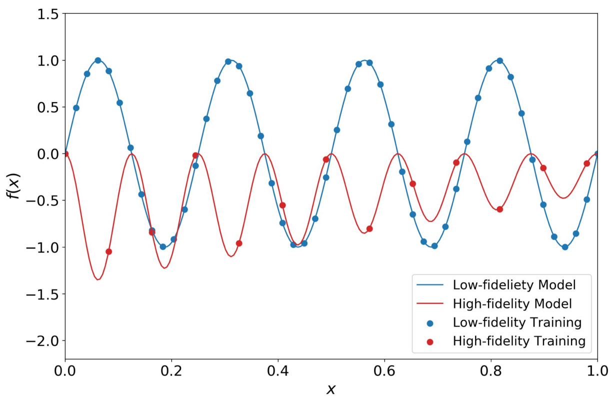
Received 26 September 2007; received in revised form 13 November 2007; accepted 28 November 2007
Available online 14 December 2007

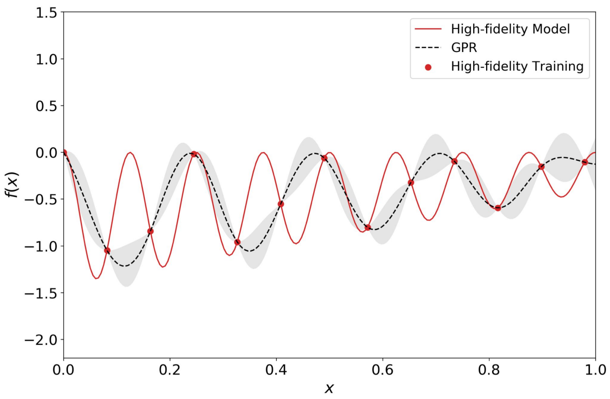
- GPR produces an uncertainty band.
- The most uncertain location is where the next data point should be calculated.

Informing High-fidelity Fits with Low-fidelity Data

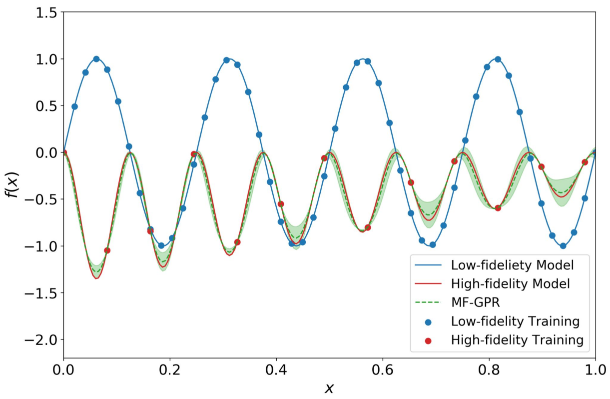
Model A: high-fidelity (HF) - expensive to generate data, very accurate (13 points)

Model B: low-fidelity (LF) - cheap to generate data, low accuracy (50 points)





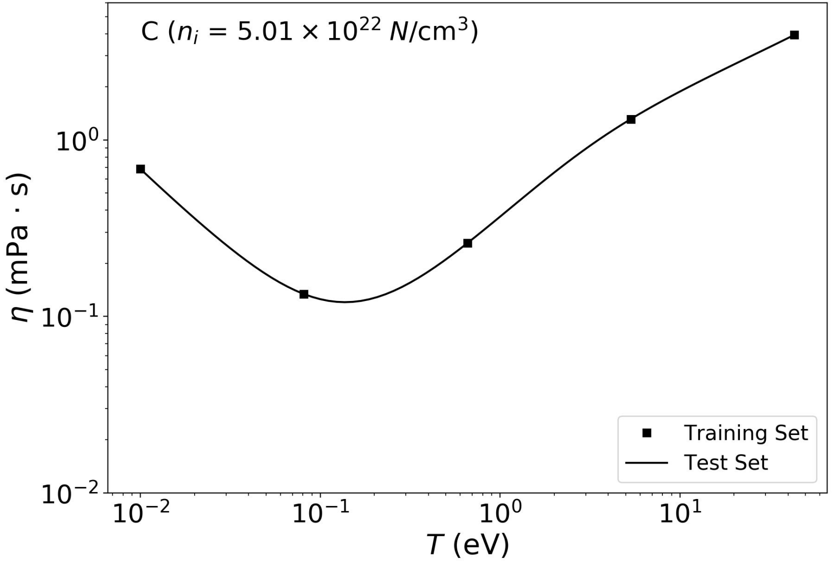
GPR



Multi-fidelity GPR "MF-GPR"

1.Cutajar, et al. *arXiv preprint arXiv:1903.07320* (2019)
2.Paleyes et al. Emulation of physical processes with Emukit (2019)

MF-GPR with Transport Coefficient Data



High-fidelity test and training data generated from the Yukawa viscosity model (YVM)

Viscosity estimates of liquid metals and warm dense matter using the Yukawa reference system

Michael S. Murillo*

Physics Division, MS D410, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

Received 26 September 2007; received in revised form 13 November 2007; accepted 28 November 2007
Available online 14 December 2007

Low-fidelity training data generated from the SMT model

PHYSICAL REVIEW E **93**, 043203 (2016)

Ionic transport in high-energy-density matter

Liam G. Stanton[1,*] and Michael S. Murillo[2,*]
[1]*Center for Applied Scientific Computing, Lawrence Livermore National Laboratory, Livermore, California 94550, USA*
[2]*Computational Physics and Methods Group, MS D413, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
(Received 16 November 2015; published 8 April 2016)

Gaussian Process Regression: a Kernel Method

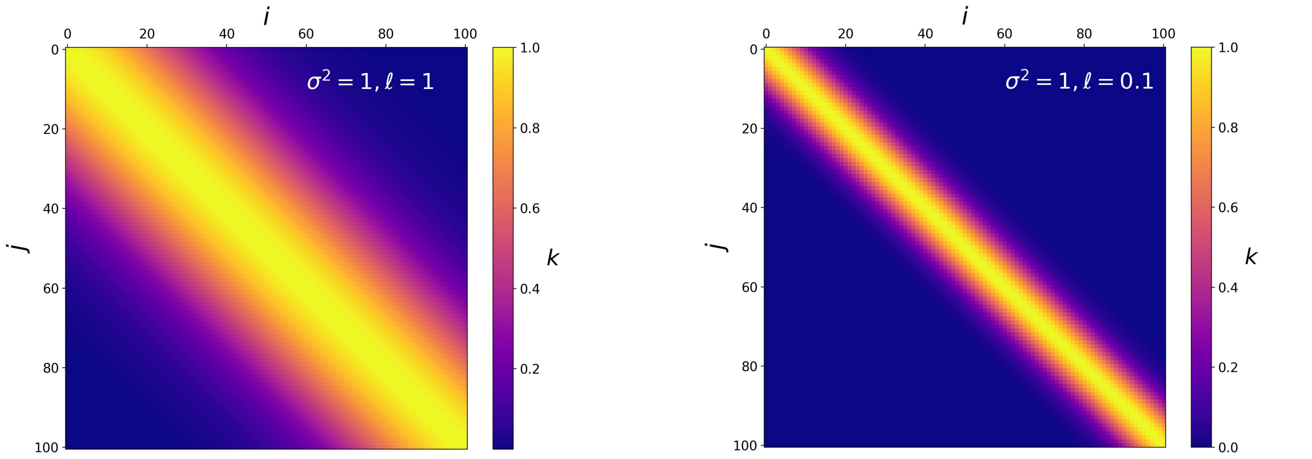
Squared exponential kernel

$$k(x_i, x_j) = \sigma^2 \exp\left(-\frac{1}{2\ell}\|x_i - x_j\|^2\right) \longrightarrow K(X, X)$$

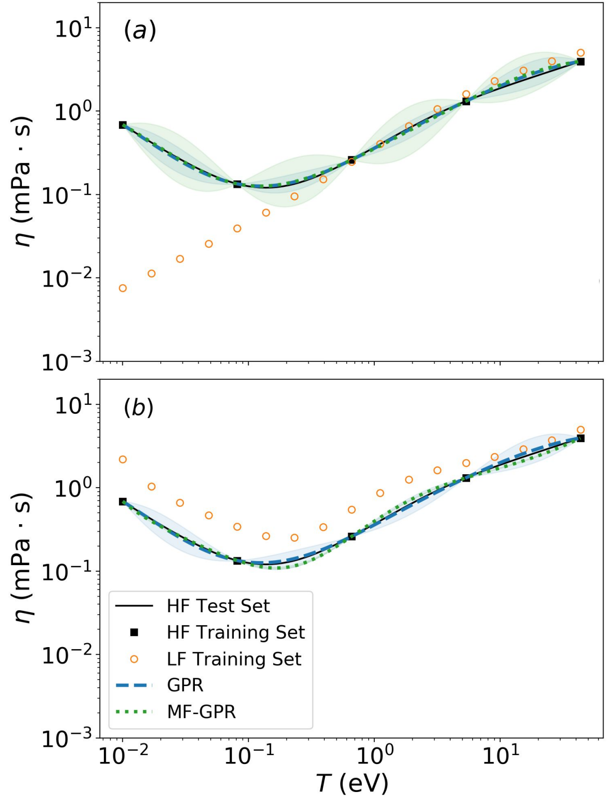
A measure of similarity between data points

σ^2 and ℓ are optimizable hyperparameters

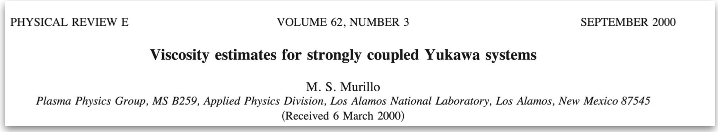
"Kernel matrix"



What LF Models Should we Choose?



(a) LF Model: SMT
(b) LF Model: Formulated using the Gibbs-Bogolyubov Inequality (YGBI)


PHYSICAL REVIEW E VOLUME 62, NUMBER 3 SEPTEMBER 2000

Viscosity estimates for strongly coupled Yukawa systems

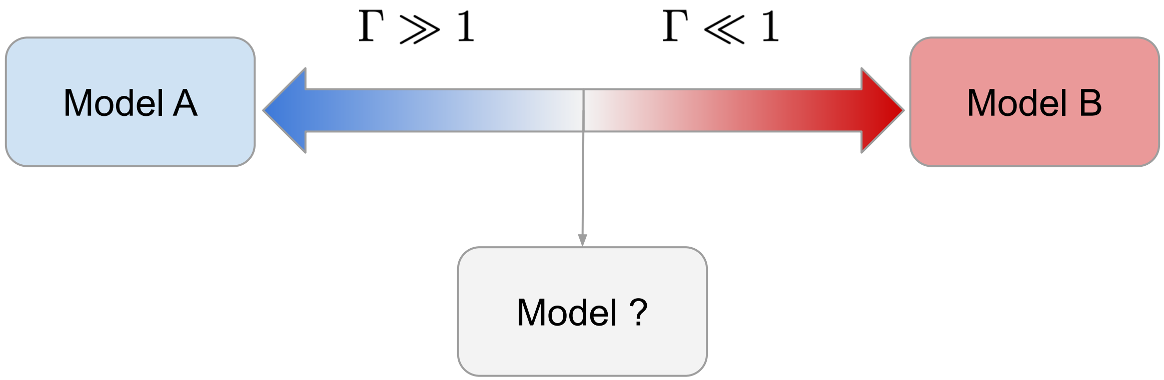
M. S. Murillo
Plasma Physics Group, MS B259, Applied Physics Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545
(Received 6 March 2000)

ρ measures the correlation between the low- and high-fidelity models.

The kernel matrix ρ reveals effectiveness of a LF model in MF-GPR.

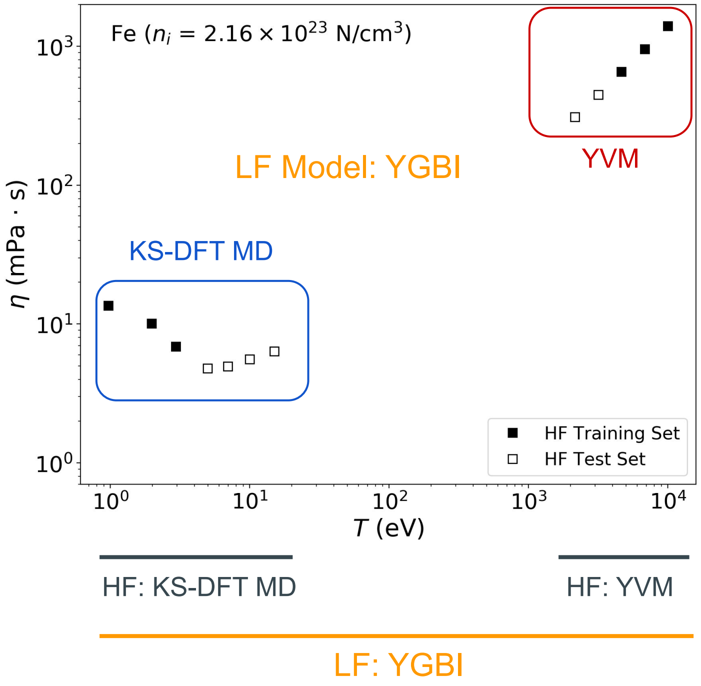
Physical Models are Often Asymptotically Accurate

Γ Coulomb Coupling Parameter

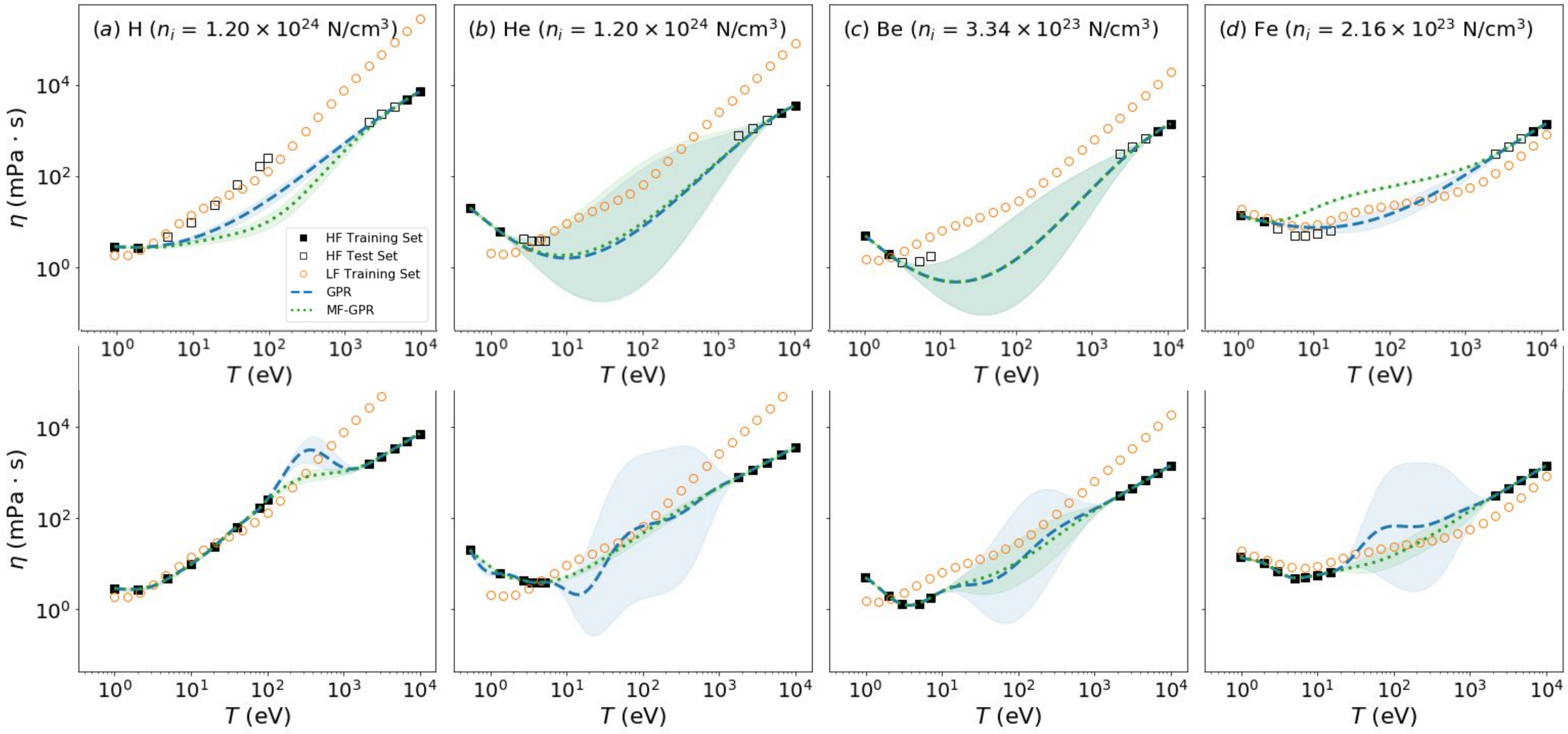


Can we use MF-GPR to help bridge the gap?

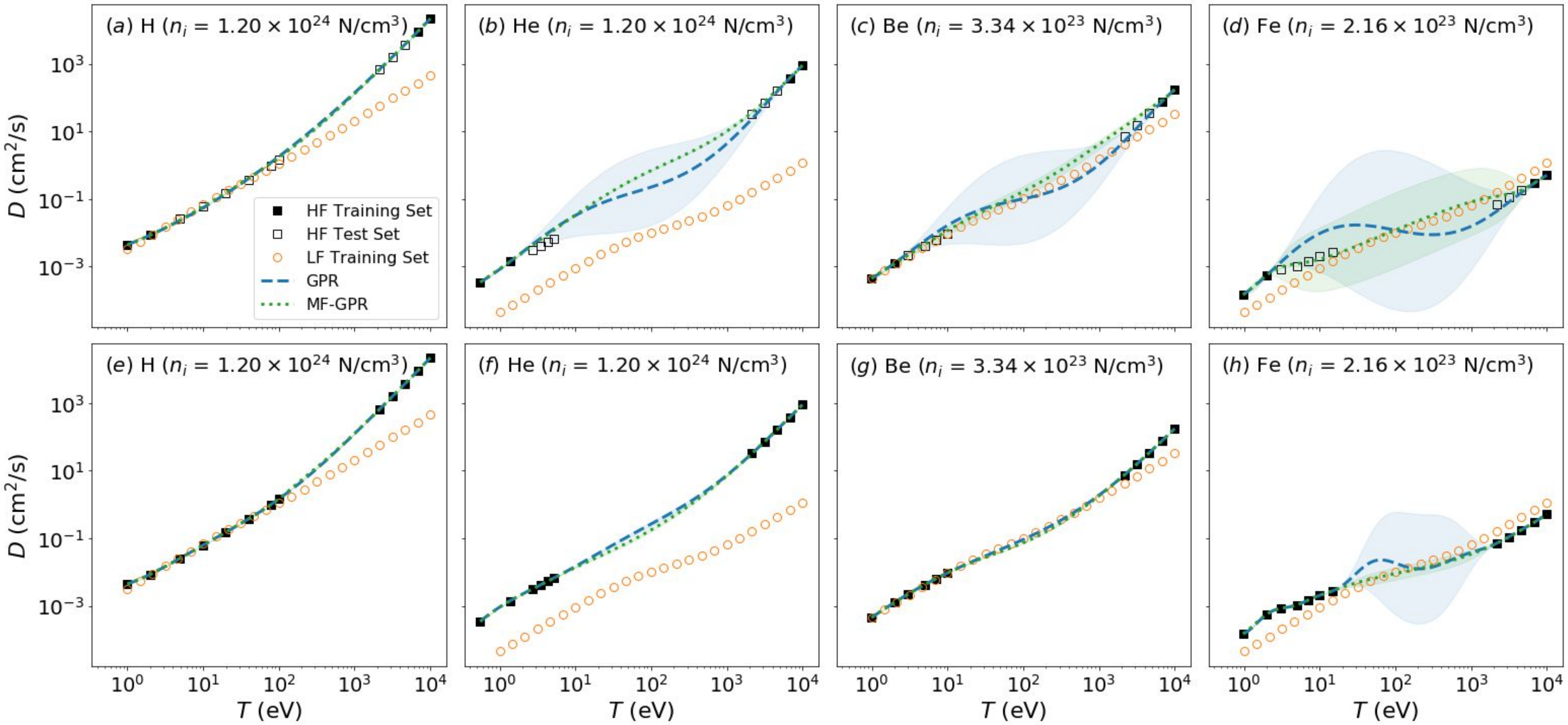
Interpolating Disparate Data with MF-GPR



Interpolating Disparate Viscosity Data with MF-GPR

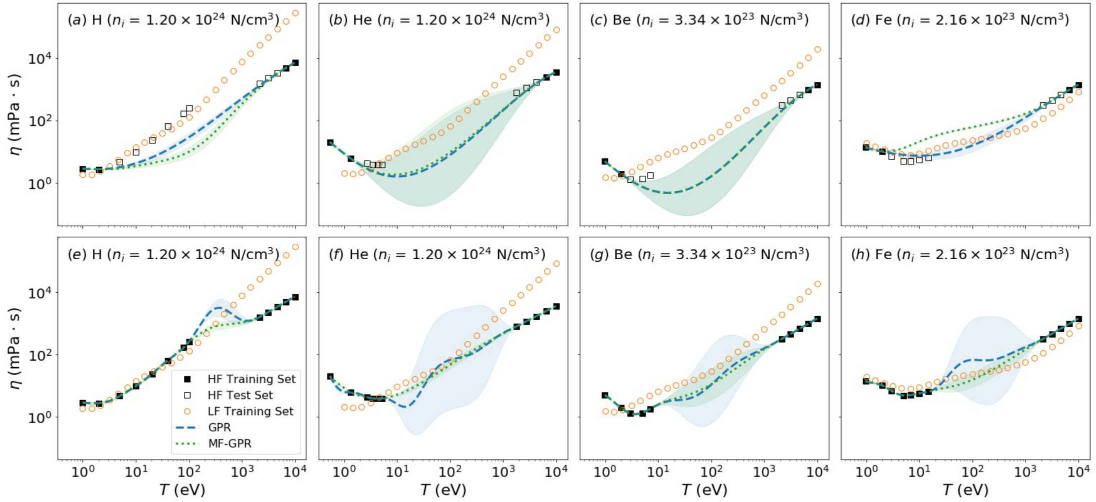


Interpolating Disparate Self-Diffusion Data with MF-GPR



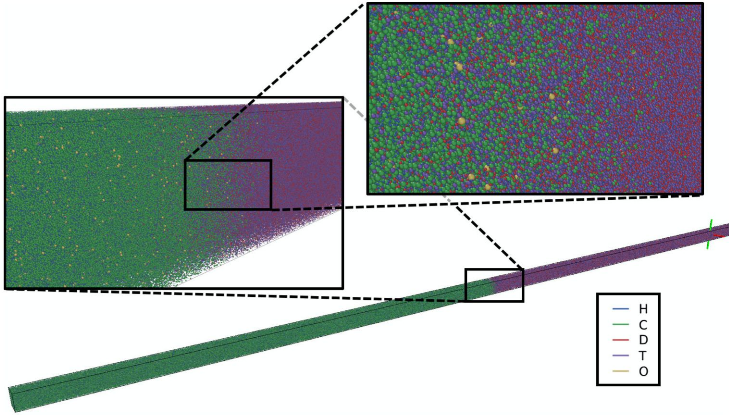
Goal: Generate Data with Data Science in Mind

1. Generate the most accurate data with minimal statistical noise
2. Use existing low- and high-fidelity data optimally
3. Identify where new data is needed



Outlook

- Large-scale, heterogeneous simulations are becoming more common and much less is known about such environments.
- Data science can guide simulations, experimental design, and data collection.
- We need computationally efficient, accurate models to effectively sample these large dimensional spaces.



Contributions

- Ruled out the need for 3-body potentials above temperatures of few eV.
- Validated transport models and provided a rule for generating large datasets of transport coefficients.
- Predicted transport coefficients across disparate regimes with sparse data, outperforming simpler techniques.
- Provided an avenue for understanding the applicability of LF models in a MF-GPR setting.