Physics-enhanced neural networks for equation-of-state calculations

Max Born Symposium “Many particle systems under extreme conditions”

04.12.2023 // Timothy Callow

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Uniwersytet Wrocławski

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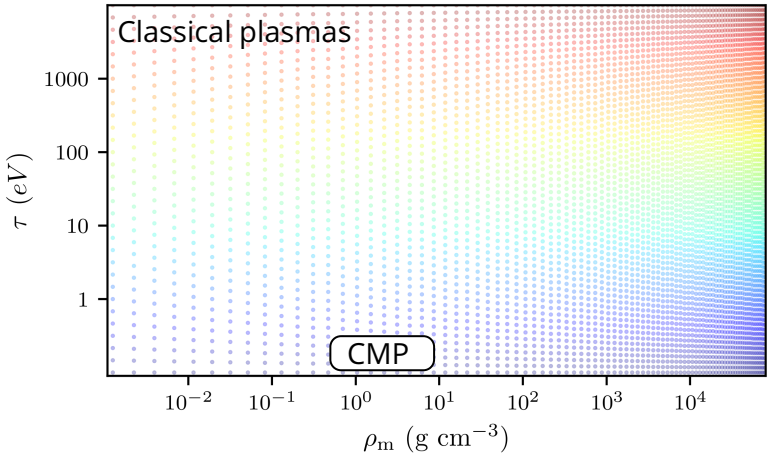
Contents

1. Motivation
2. Average-atom models
3. Average-atom EOS results
4. Neural network methodology
5. Neural network EOS results
6. Comparison with other approaches
7. Conclusions

Warm-dense matter and inertial confinement fusion



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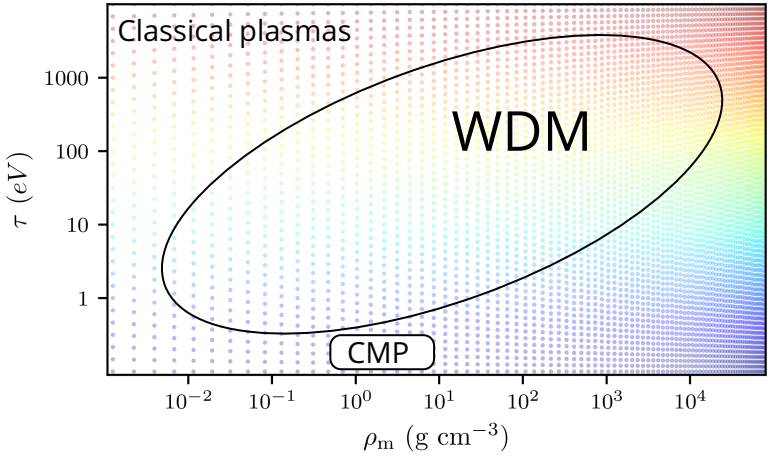

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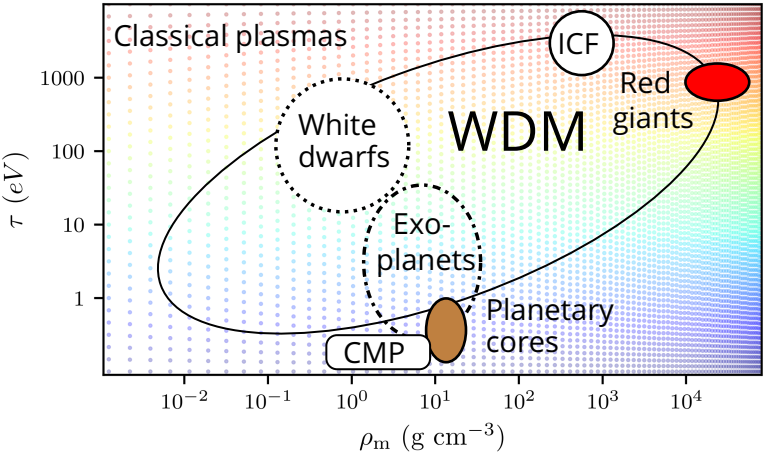

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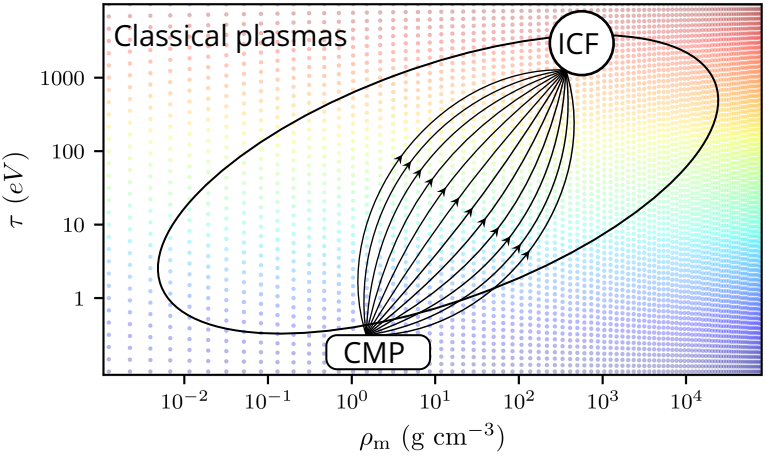
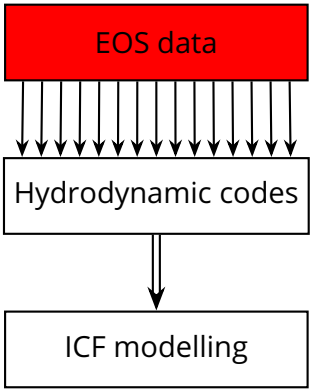


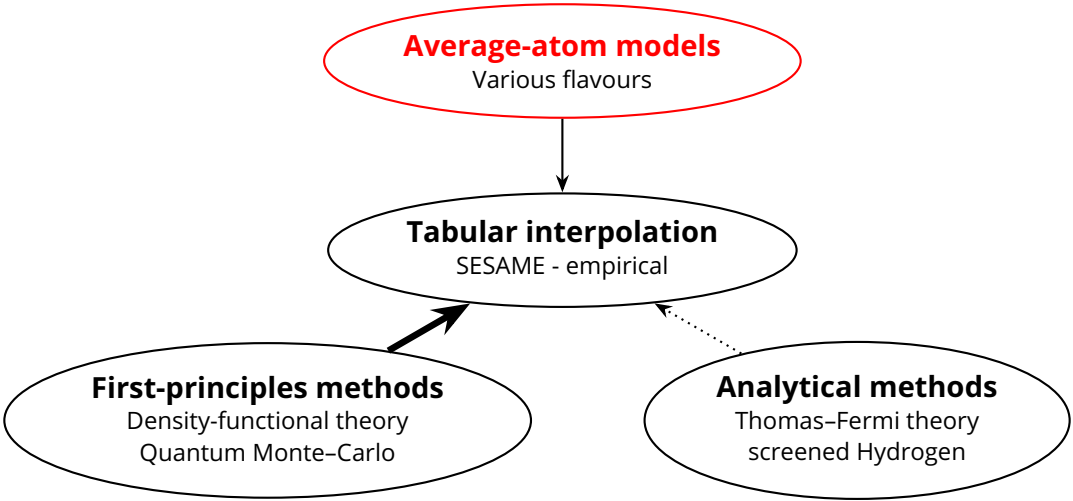
ICF Modelling



How do we provide EOS data (pressure and derivatives) to hydrodynamic codes

- **instantaneously**, and
- across a **wide range** of temperatures and densities?

Currently-used approaches



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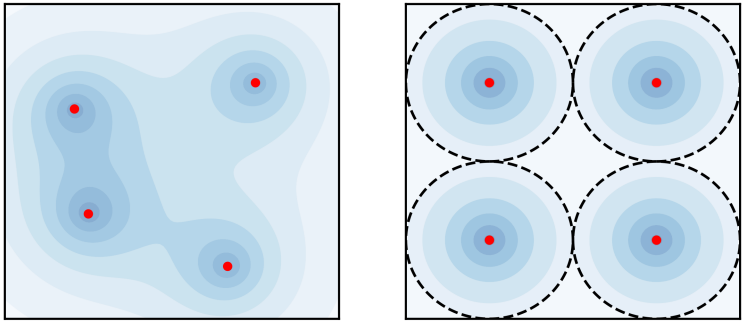
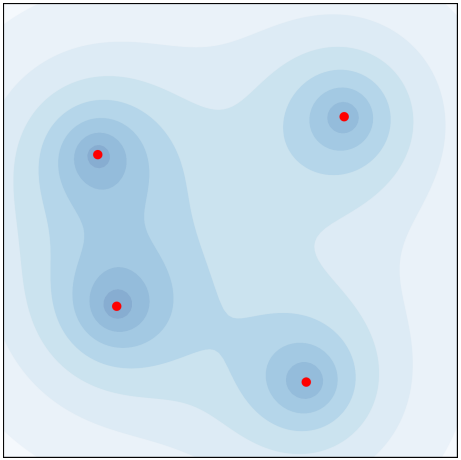
Average-atom models: concept



- We want to map the left hand picture (complex system) to the one on the right (simple system)

Voronoi decomposition of space



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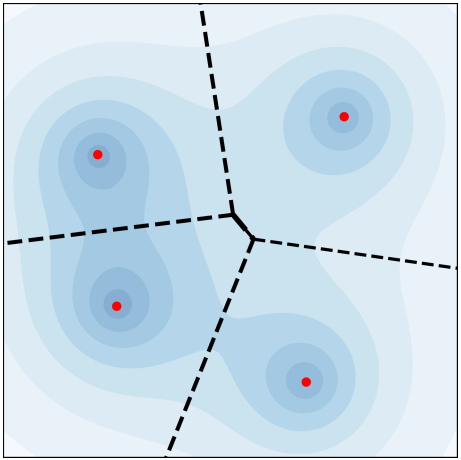
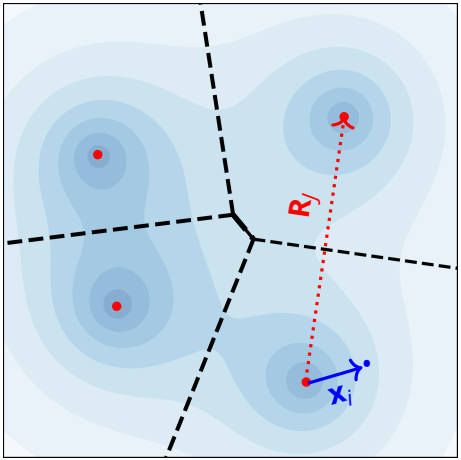


- Each electron is “assigned” to the nearest nucleus

Voronoi decomposition of space



- Each electron is “assigned” to the nearest nucleus
- A transformation to the new co-ordinate space defined by $\mathbf{R}_J, \mathbf{x}_{iJ}$ is made
- We shall expand in powers of $|\mathbf{y}_{iJ}| = \frac{|\mathbf{x}_i|}{|\mathbf{R}_J|} \leq \frac{1}{2}$

Expansion of coupling terms in Hamiltonian (i)

Under the assumption that the electron density in each cell is identical:

$$\hat{H} = N_\mathrm{n} \times \left\{ \hat{H}_\mathrm{el,at} + \frac{Z^2}{2} \sum_{J=2}^{N_\mathrm{n}} \frac{\hat{W}_J}{|\mathbf{R}_J|} \right\}$$

[1] Callow *et al.*, PRR **4**, 023055 (2022)

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- $\hat{H}_\mathrm{el,at}$ is a *single-atom* Hamiltonian
- $\hat{W}_J$ are *couplings* between nuclei and electrons in different cells
- Perturbative expansion of $\hat{W}_J$ terms: lowest order term $\hat{W}_J^{(0)} = 0$!
- So Hamiltonian becomes that of a single (average) atom to lowest order[1]

[1] Callow *et al.*, PRR **4**, 023055 (2022)

Boundary conditions

- We *implicitly* account for interactions between neighbouring atoms via boundary conditions
- We can impose smoothness of the electron density at the sphere's edge:

$$\left.\frac{\mathrm{d}n(r)}{\mathrm{d}r}\right|_{r=R_{\mathrm{WS}}} = 0$$

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  1. $0 = X_{nl}(R_{\mathrm{WS}}) \Rightarrow$ anti-bonding MO[2] ("Dirichlet")

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  2. $0 = \left.\frac{\mathrm{d}X_{nl}(r)}{\mathrm{d}r}\right|_{r=R_{\mathrm{WS}}} \Rightarrow$ bonding MO ("Neumann")

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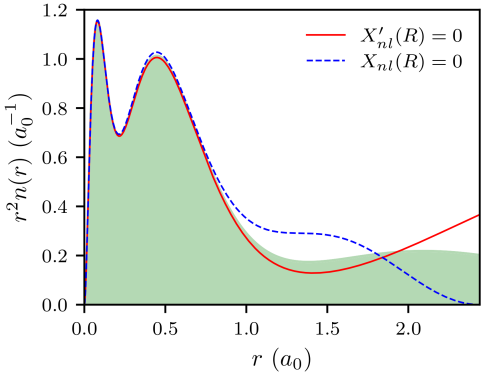
Boundary conditions: Band-structure model



Radial density distribution (Al, 5 g cm^{-3}) for the band-structure model

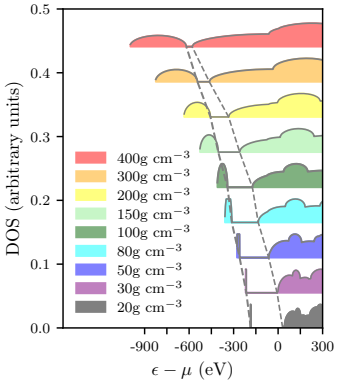
$$n(r) = 2\sum_k w_k \sum_{n,l} (2l+1) f_{knl} |X_{knl}(r)|^2$$

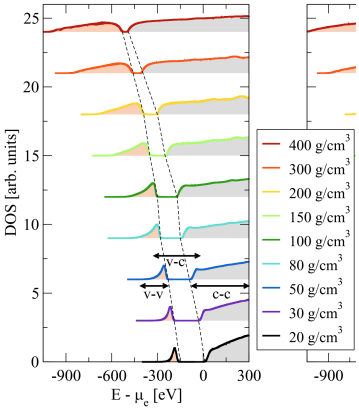
The Dirichlet and Neumann b.c.s define the *limiting wave-functions* of each band

Solve KS equations for *every* energy within the band

Weightings w_k from *Hubbard* DOS: Massacrier *et al.*, PRR **3** 023026 (2021)

DOS comparison between Massacrier AA and DFT-MD





DOS for Carbon at $T = 100$ eV. Left: AA model. Right: DFT-MD results from Bethkenhagen *et al* PRR **2**, 023260 (2020)

Pressure in average-atom models

- The total pressure is given as the sum of the ionic and electronic pressures
- Ionic pressure is computed from the *ideal gas law*:

$$P_{\text{ion}} = \frac{nRT}{V}$$

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- Various methods are available for the electronic pressure. We use the following four:

  1. Finite differences:

$$P_{\text{e}} = -\left.\frac{\partial F}{\partial V}\right|_{T}$$

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  2. Stress-tensor:

$$P_{\text{e}} = \frac{1}{3}\,\text{Tr}\left\{\frac{1}{2}\sum_k f_k\,\text{Re}\left(\frac{\partial\phi_k^*}{\partial x_i}\frac{\partial\phi_k}{\partial x_j} - \phi_k^*\frac{\partial^2\phi_k^*}{\partial x_i \partial x_j}\right)\right\}$$

Pressure in average-atom models

- The total pressure is given as the sum of the ionic and electronic pressures
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- Various methods are available for the electronic pressure. We use the following four:

  3. Virial theorem:

$$P_{\text{e}} = \frac{2T + E_{\text{en}} + W_{\text{xc}}}{3V}$$

Pressure in average-atom models

- The total pressure is given as the sum of the ionic and electronic pressures
- Ionic pressure is computed from the *ideal gas law*:

$$P_{\text{ion}} = \frac{nRT}{V}$$

- Various methods are available for the electronic pressure. We use the following four:

  4. Ideal approximation:

$$P_{\text{e}} = \frac{2^{3/2}}{3\pi^2} \int_{v_{\text{s}}(R_{\text{WS}})}^{\infty} \mathrm{d}\epsilon \, \frac{\epsilon^{3/2}}{1 + e^{(\epsilon-\mu)/T}} \, .$$

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Average-atom pressure results

- We test on the first-principles dataset by Militzer *et al*, calculated using DFT-MD and PIMC methods: PRE **103**, 013203 (2021)
- In total we compare 2181 data points, spanning 11 elements

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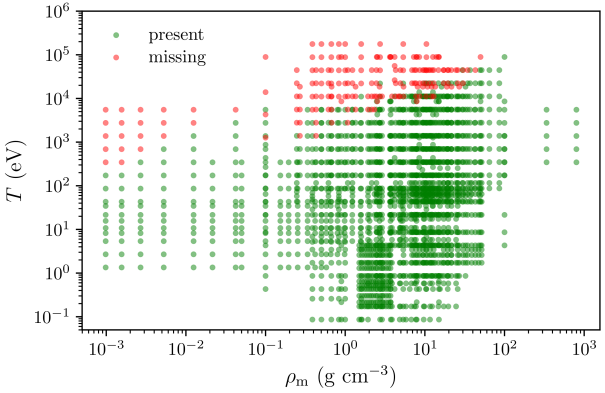
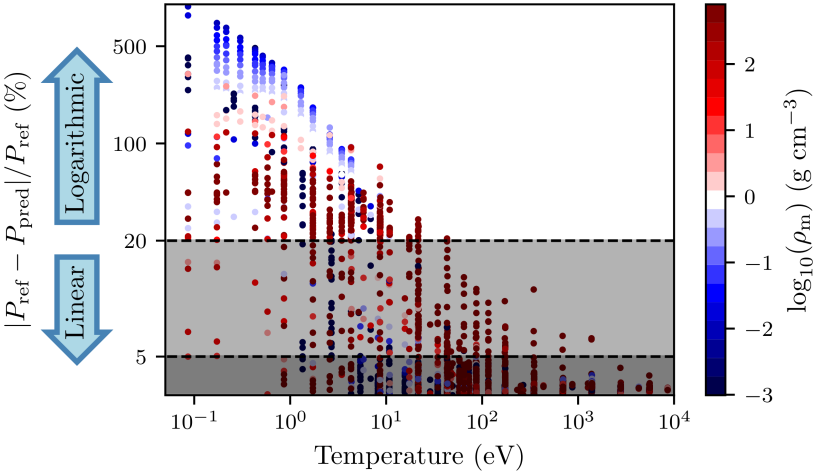
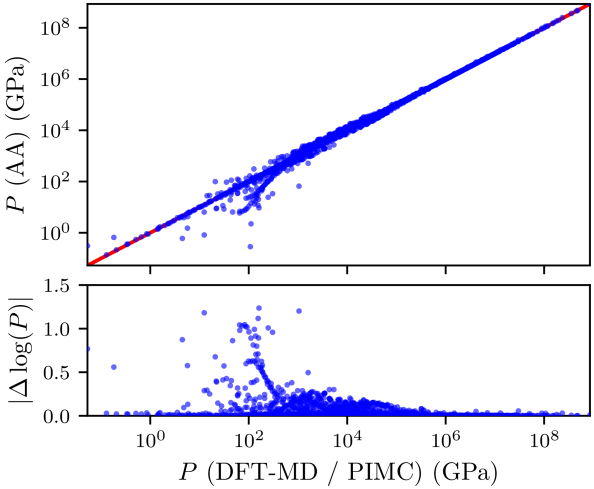


Average-atom pressure results (i)



Pressure results (ii)



Average-atom pressure results: summary

	All temperatures	$T > 10$ eV
MAPE[3]	33	
MALE[4]	8.7	
f_{20}[5]	75	
f_5[6]	63	

[3] Mean absolute percentage error
[4] Mean absolute log error $\times 100$
[5] Percentage of results with $< 20\%$ error
[6] Percentage of results with $< 5\%$ error

Average-atom pressure results: summary

	All temperatures	$T > 10$ eV
MAPE[7]	33	2.6
MALE[8]	8.7	1.1
f_{20}[9]	75	98
f_5[10]	63	87

[7] Mean absolute percentage error
[8] Mean absolute log error $\times$ 100
[9] Percentage of results with $< 20\%$ error
[10] Percentage of results with $< 5\%$ error

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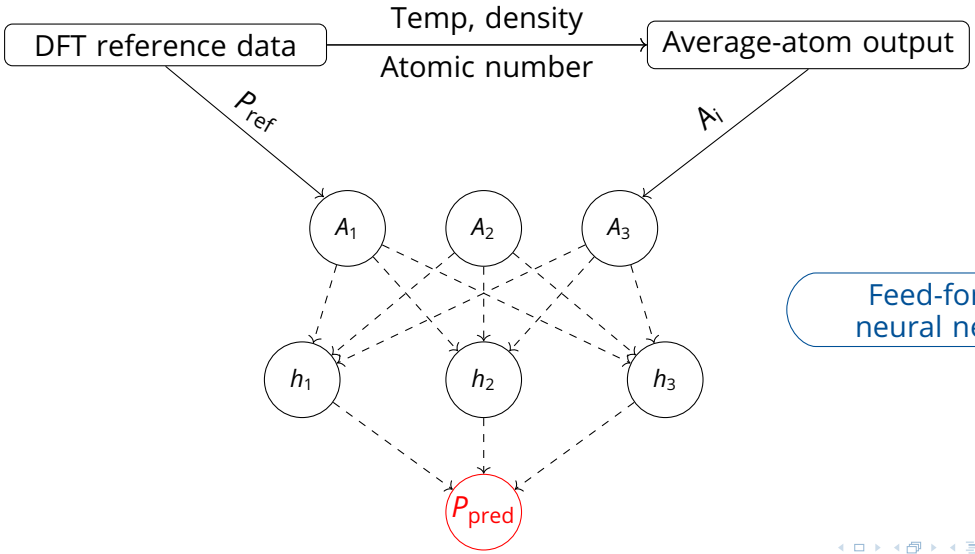
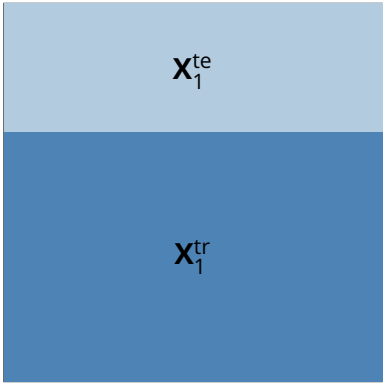
Neural network architecture



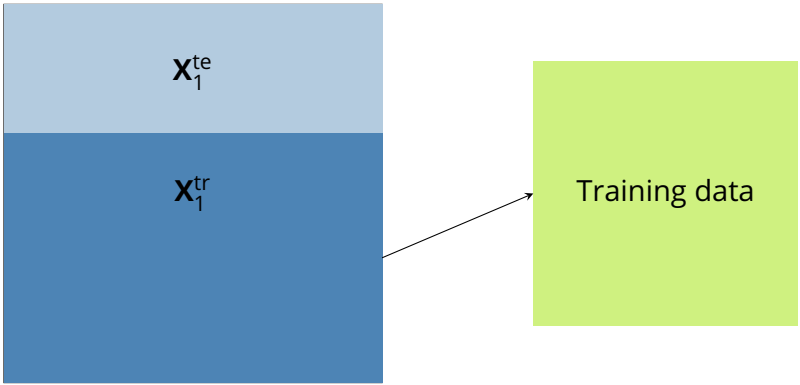
Feed-forward neural network

Neural network methodology

Neural network methodology



Neural network methodology



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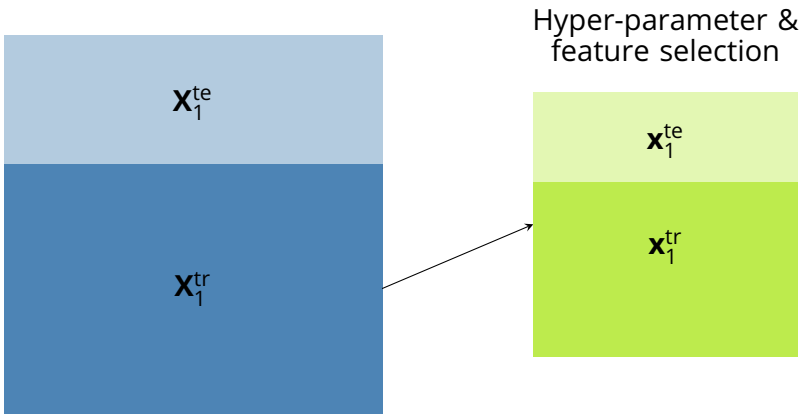

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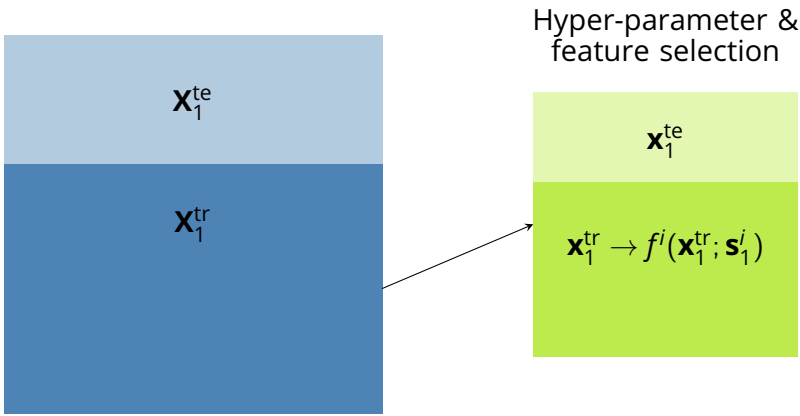

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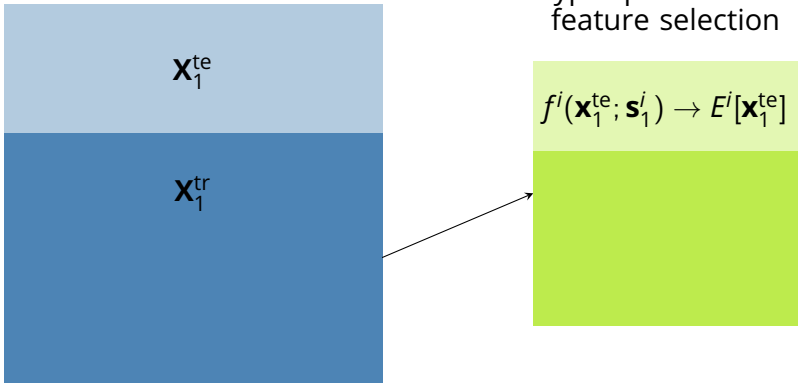

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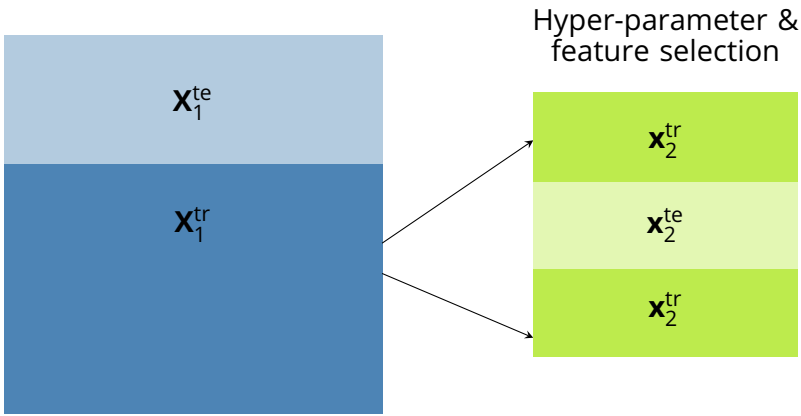

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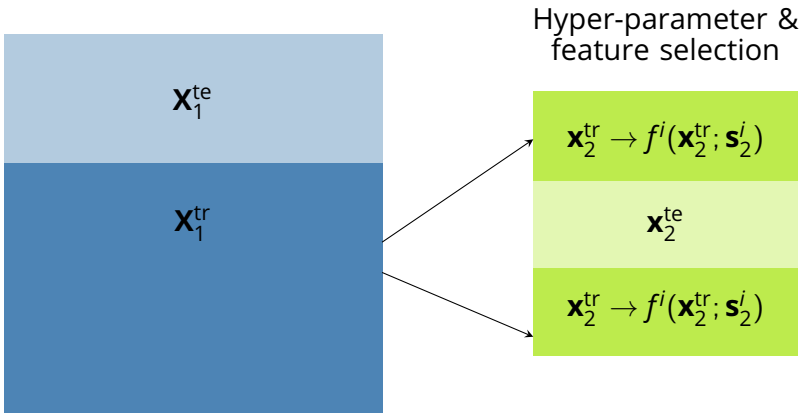

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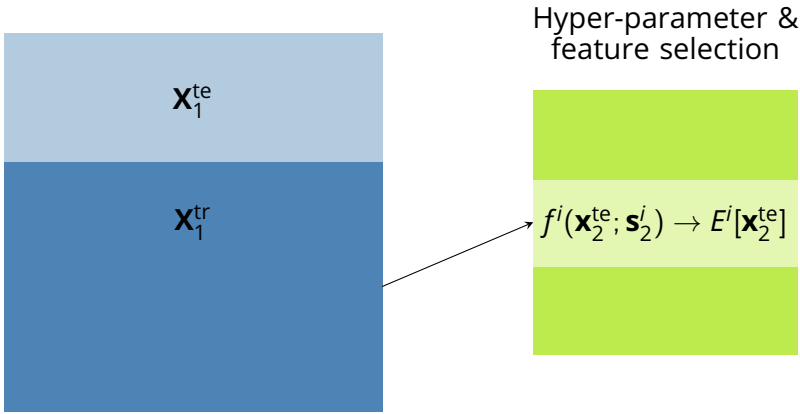

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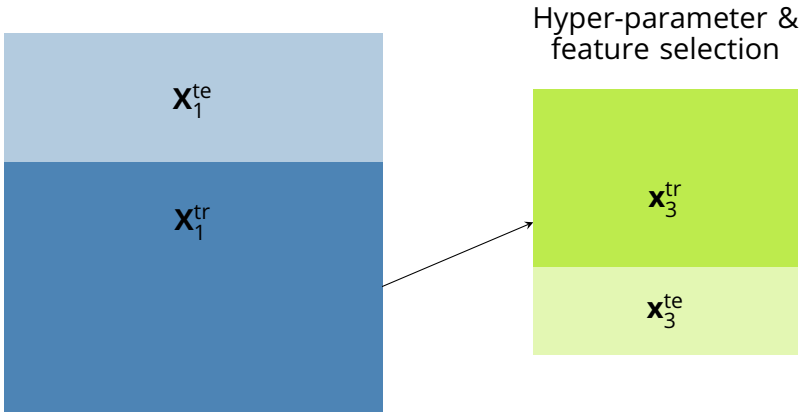

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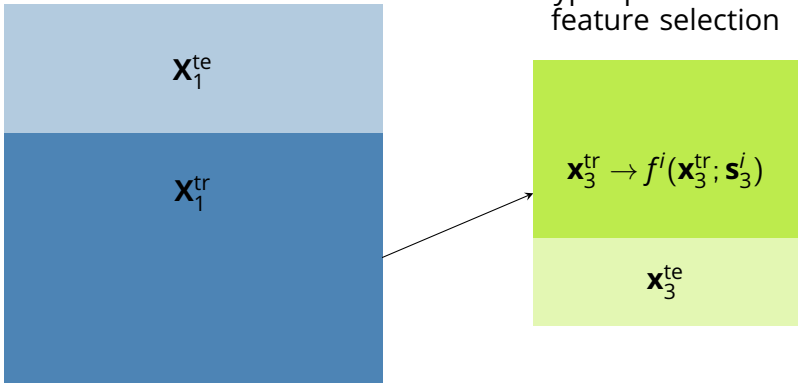

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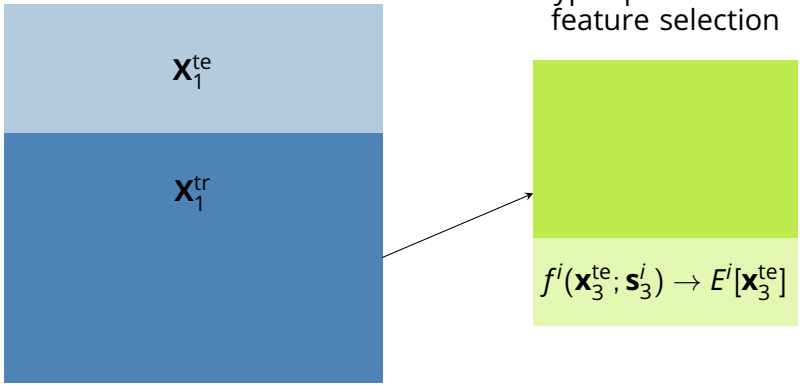

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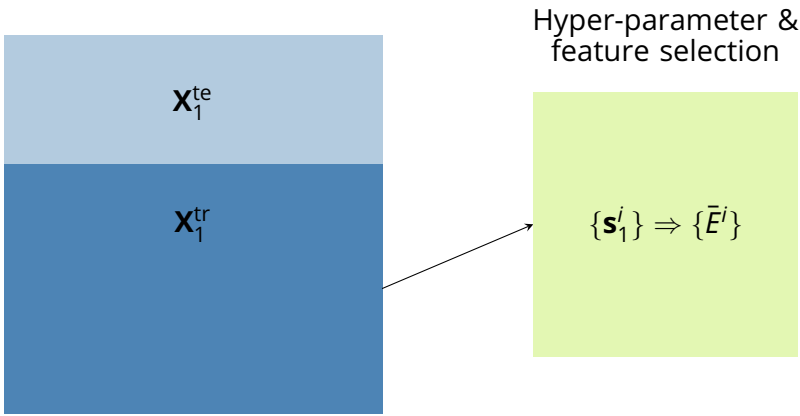

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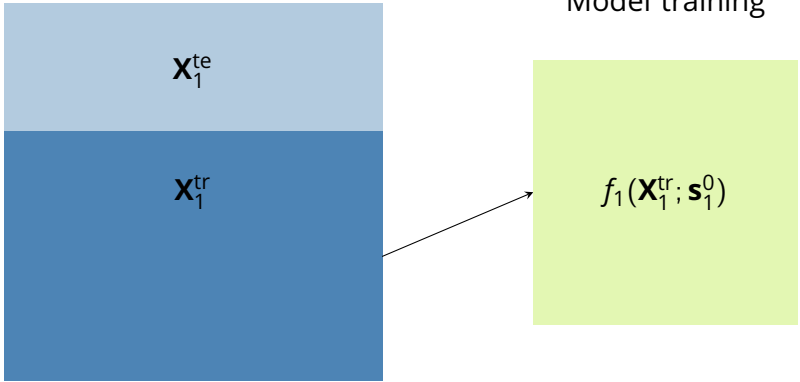

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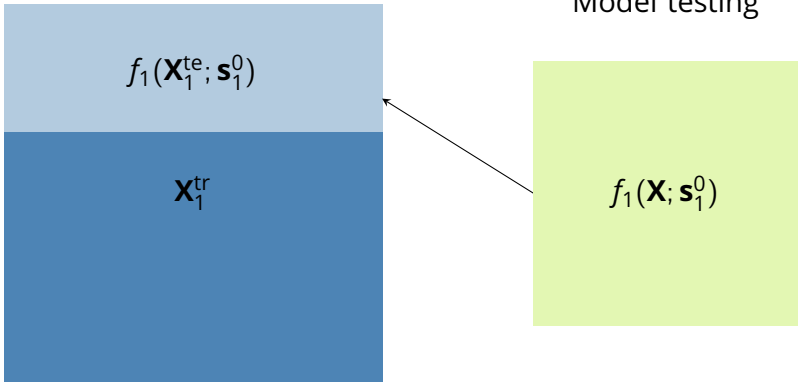

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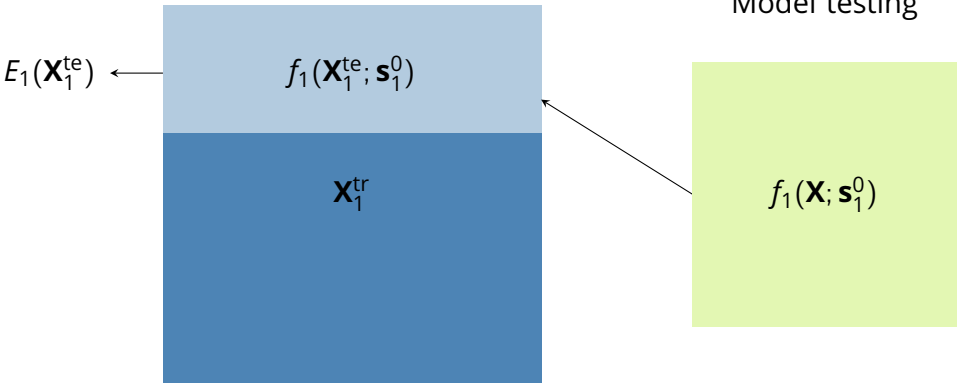

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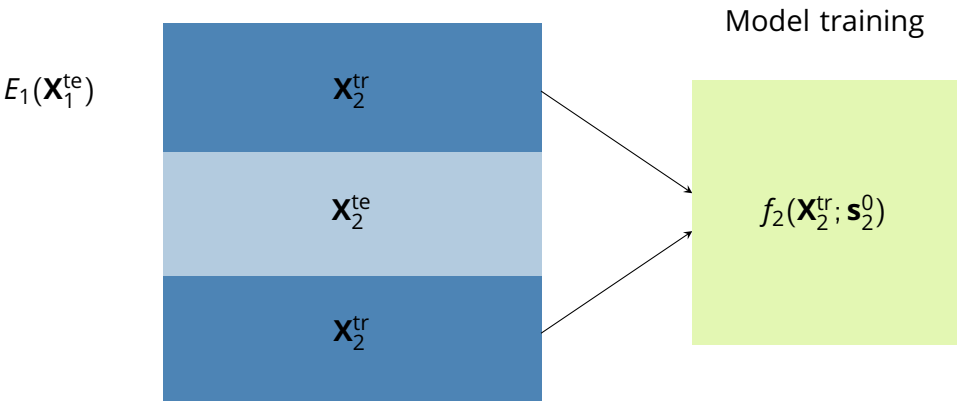

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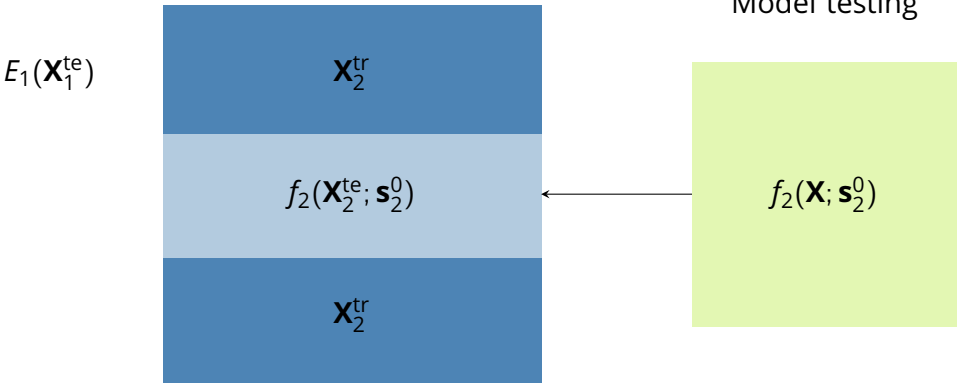

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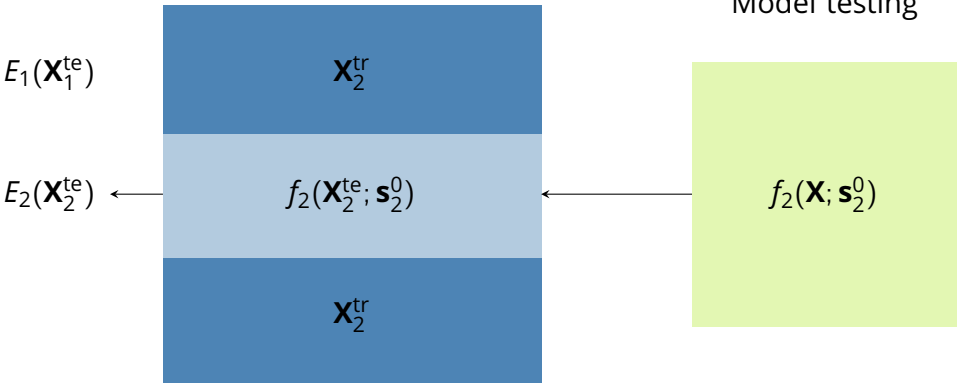

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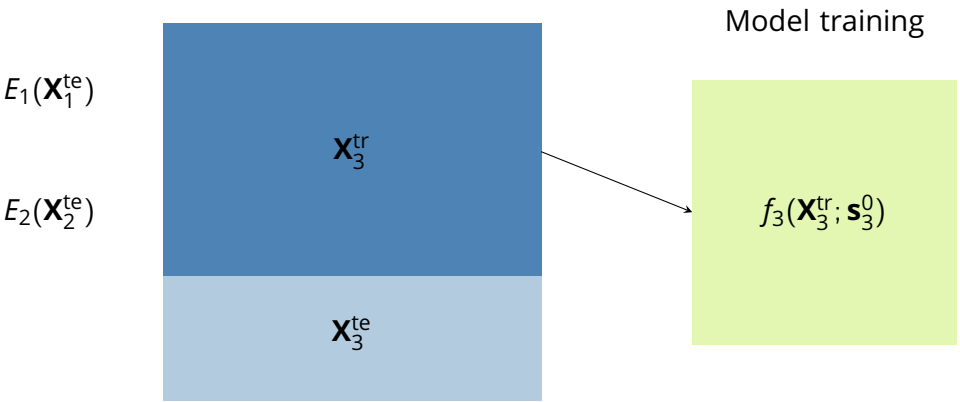

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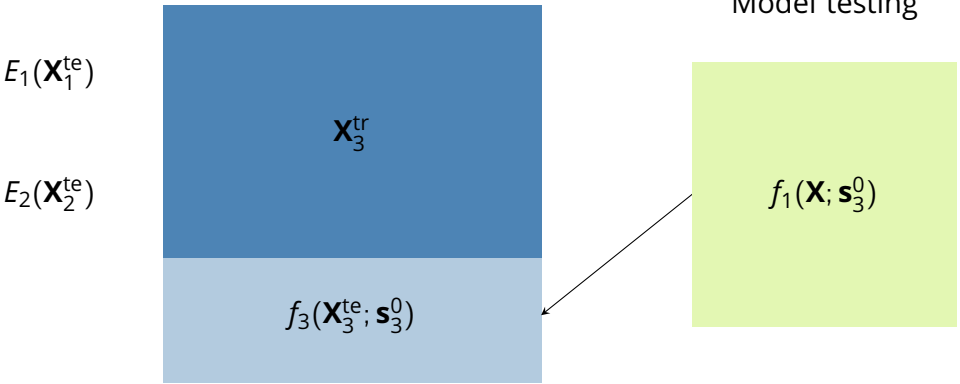

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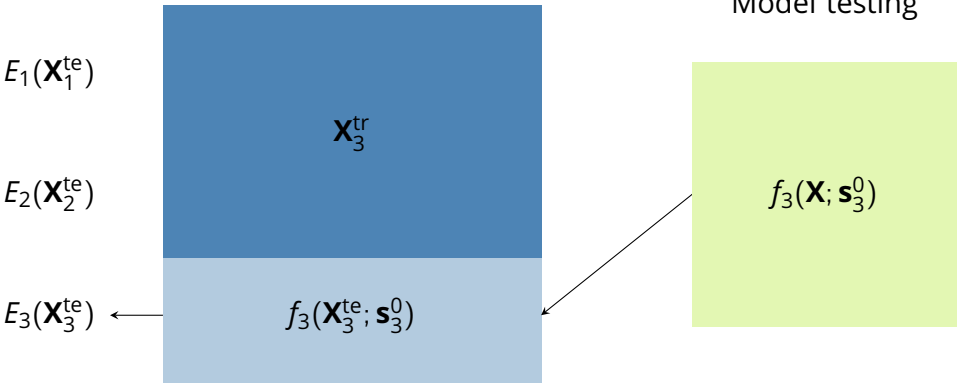

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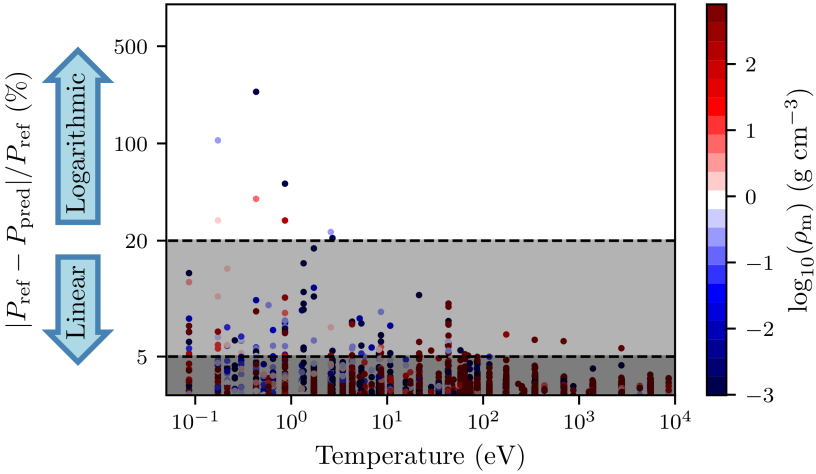
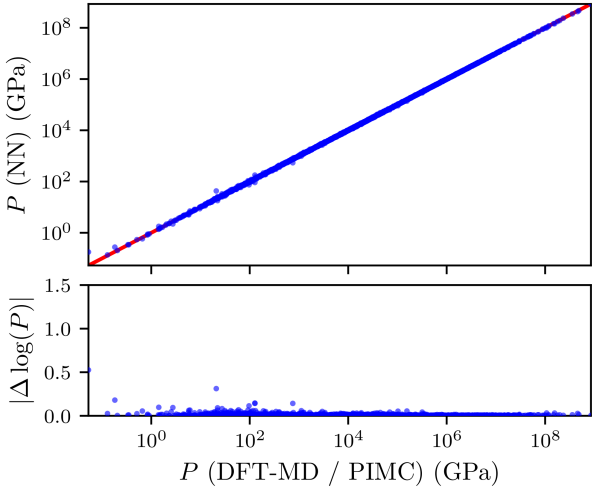
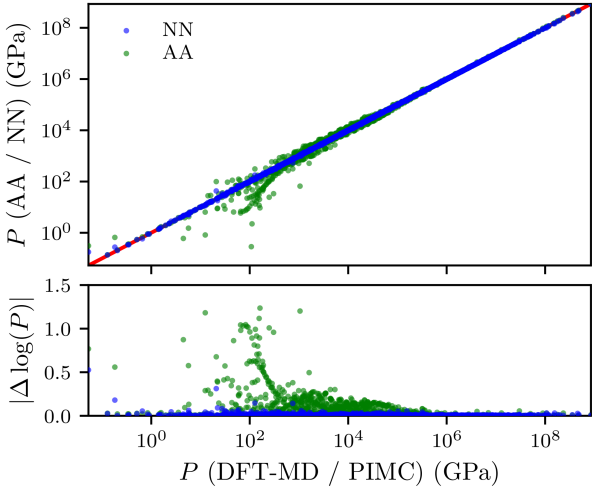


Neural network pressure results (ii)



Neural network vs average-atom pressure results



Average-atom vs neural network pressure results: summary

	All temps, AA	All temps, NN	$T > 10$ eV, AA	$T > 10$ eV, AA
MAPE	33			
MALE	8.7			
f_{20}	75			
f_5	63			

Average-atom vs neural network pressure results: summary

	All temps, AA	All temps, NN	$T > 10$ eV, AA	$T > 10$ eV, AA
MAPE	33	1.8		
MALE	8.7	0.77		
f_{20}	75	99.6		
f_5	63	94		

Average-atom vs neural network pressure results: summary

	All temps,AA	All temps, NN	$T > 10$ eV, AA	$T > 10$ eV, AA
MAPE	33	1.8	2.6	
MALE	8.7	0.77	1.1	
f_{20}	75	99.6	98	
f_5	63	94	87	

Average-atom vs neural network pressure results: summary

	All temps, AA	All temps, NN	$T > 10$ eV, AA	$T > 10$ eV, AA
MAPE	33	1.8	2.6	1.2
MALE	8.7	0.77	1.1	0.51
f_{20}	75	99.6	98	100
f_5	63	94	87	98

Performance on out-of-distribution data

- How does the neural network model perform (compared to the average-atom model) on **unseen** elements?
- We test on the DFT-MD dataset of Ding and Hu, PoP **24**(6):062702, 2017, which contains Beryllium only, not present in the FPEOS dataset

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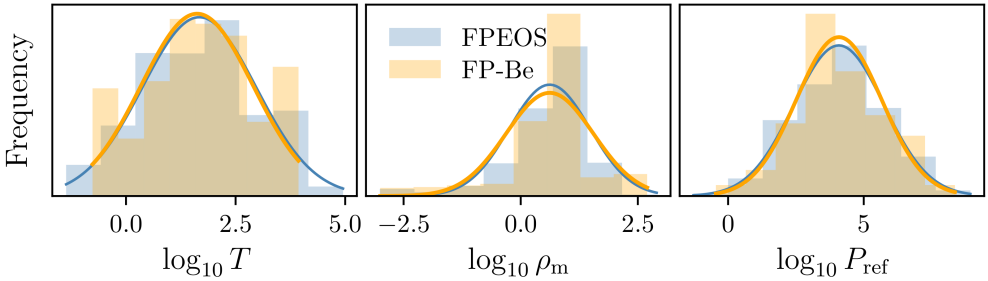
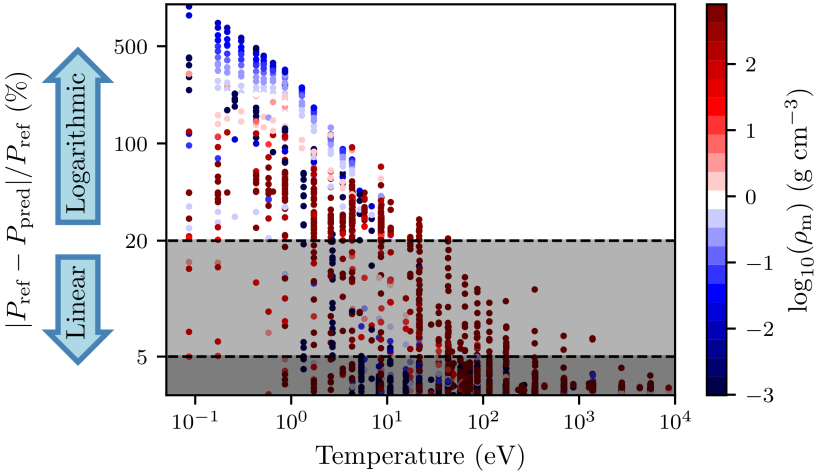
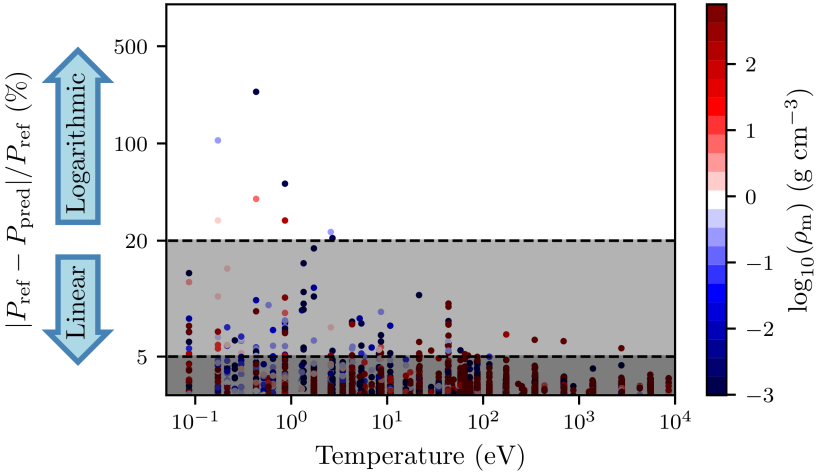


Average-atom results for FP-Be dataset



Neural network results for FP-Be dataset



Average-atom vs neural network pressure results: summary

	All temps, AA	All temps, NN	$T > 10$ eV, AA	$T > 10$ eV, NN
MAPE	18			
MALE	5.4			
f_{20}	79			
f_5	59			

Average-atom vs neural network pressure results: summary

	All temps, AA	All temps, NN	$T > 10$ eV, AA	$T > 10$ eV, NN
MAPE	18	4.5		
MALE	5.4	1.9		
f_{20}	79	99		
f_5	59	70		

Average-atom vs neural network pressure results: summary

	All temps,AA	All temps, NN	$T > 10$ eV, AA	$T > 10$ eV, NN
MAPE	18	4.5	2.8	
MALE	5.4	1.9	1.2	
f_{20}	79	99	100	
f_5	59	70	80	

Average-atom vs neural network pressure results: summary

	All temps, AA	All temps, NN	$T > 10$ eV, AA	$T > 10$ eV, NN
MAPE	18	4.5	2.8	3.4
MALE	5.4	1.9	1.2	1.5
f_{20}	79	99	100	100
f_5	59	70	80	75

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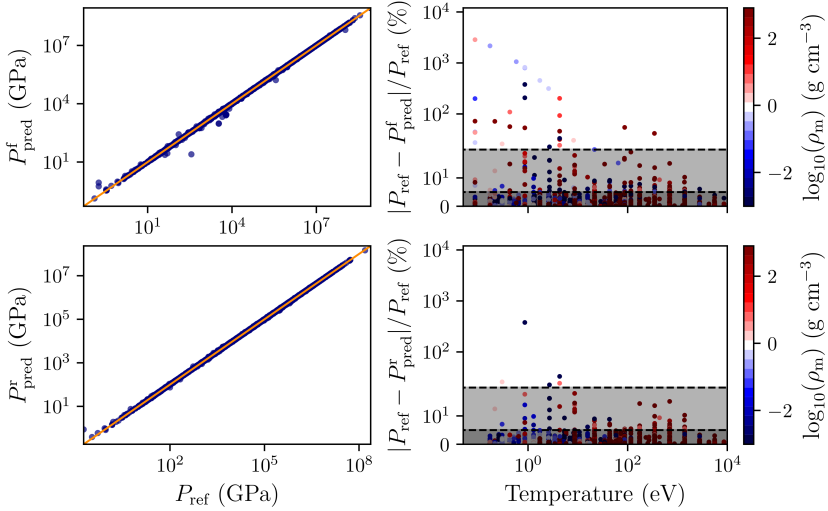
Comparison with direct interpolation of FPEOS data



Comparison with direct interpolation of FPEOS data

	FPEOS (1)	FPEOS (2)	NN
MAPE	1.6		
MALE	0.64		
f_{20}	99.7		
f_5	95		

Comparison with direct interpolation of FPEOS data

	FPEOS (1)	FPEOS (2)	NN
MAPE	1.6	[1300]	
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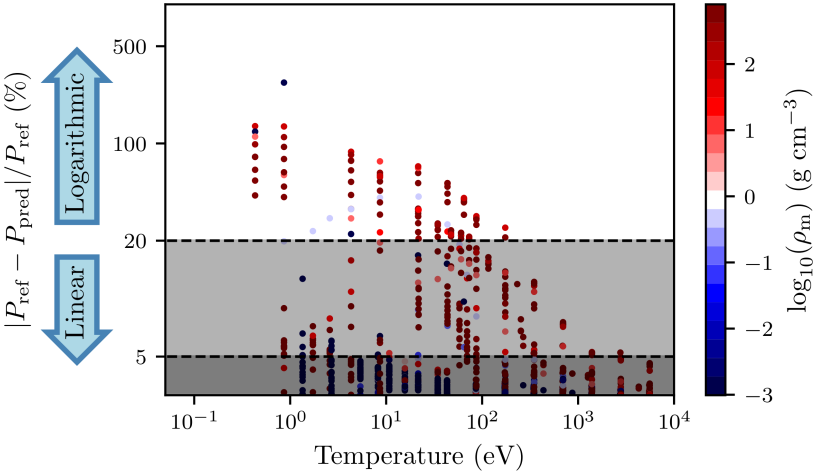
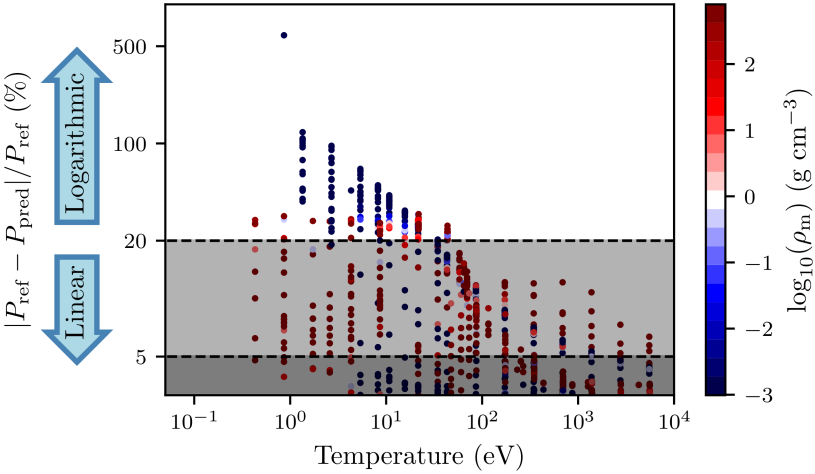
SESAME vs FPEOS



QEOS (analytical method) vs FPEOS



Comparison of AA and NN results with SESAME and QEOS

	SESAME[11]	QEOS	AA	NN
MAPE	8.9			
MALE	3.3			
f_{20}	88			
f_5	67			

[11] Thanks to Katharina Falk (HZDR) for access to SESAME code

Comparison of AA and NN results with SESAME and QEOS

	SESAME[11]	QEOS	AA	NN
MAPE	8.9	15		
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Comparison of AA and NN results with SESAME and QEOS

	SESAME[11]	QEOS	AA	NN
MAPE	8.9	15	7.6	
MALE	3.3	5.4	2.8	
f_{20}	88	77	90	
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N.B. AA and NN results have changed since only H, C, Al and Si elements now present!

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3. Average-atom codes are sufficiently accurate above 10 eV, but cannot be relied upon below that[12]
4. Our average-atom-informed neural-network approach is highly accurate across a wide range of temperatures
5. This approach has advantages over many current state-of-the-art alternatives (empirical, analytical, or interpolation of first-principles data)

[12] At present our AA code is not fast enough for inline hydro calculations, but we have solutions for this

References and thanks

Thanks to my collaborators:

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