

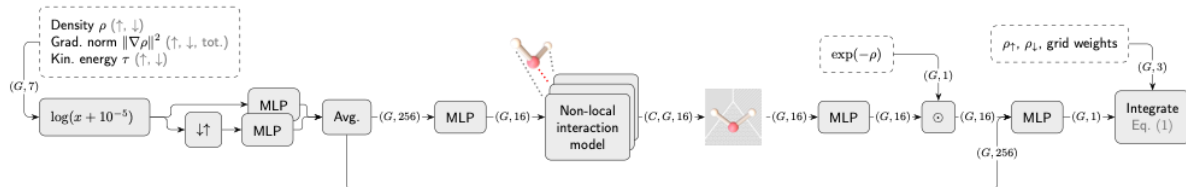


The not-so secret sauce

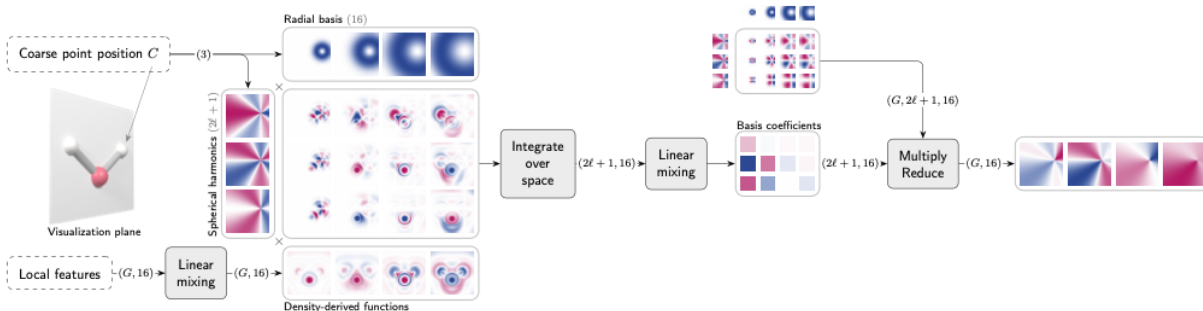
Equation to solve:

$$E_{XC}^{\theta}[\rho] = -\frac{3}{4} \left(\frac{6}{\pi}\right)^{\frac{1}{3}} \int \left(\rho^{(\uparrow)}(r)^{4/3} + \rho^{(\downarrow)}(r)^{4/3}\right) f_{\theta}[\mathbf{x}[\rho]](r) dr \quad (1)$$

- f_{θ} (enhancement factor) obtained through scheme below
- Use meta-GGA features: electronic density, gradient and kinetic energy density
- MLP = multilayer perceptron
- “G” refers to integration grid points while “C” to a coarse grid (atomic positions) \Rightarrow “coarse non-local interactions”



(a) Skala architecture overview



(b) Non-local interaction model

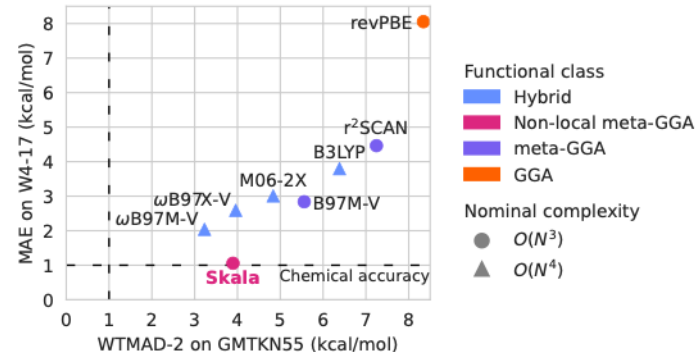
Goals

- Exploit Deep Learning (DL) to achieve chemical accuracy at low computational cost.
- Recover non-local interactions from semi-local features.

Training set

Dataset	Number of reactions		Avg. E [kcal/mol]	Elements	Description
	Full	Training			
MSR-ACC/					
TAE	80549	78650 (97.6%)	539.76	H, Li-F, Na-Cl	Total atomization energies
Conf	34021	33795 (99.3%)	1.68	H, Li-F, Na-Cl	Conformational energies
PA	10226	9961 (97.4%)	222.68	H, Li-F, Na-Cl	Proton affinities
IP	9962	9677 (97.1%)	164.82	H, Li-F, Na-Cl	Ionization potentials
Reactions	4964	3709 (74.7%)	40.63	H, C-O	Reaction paths
Atomic/					
TOT	16	16 (100.0%)	—	H-He, B-Ar	Atomic total energies
EA	11	11 (100.0%)	33.58	H, B-C, O-F, Na, Al-Cl	Atomic electron affinities
IP	43	43 (100.0%)	667.19	He, B-Ar	Atomic ionization potentials
W4-CC	14	14 (100.0%)	745.11	C	Total atomization energies of carbon clusters
NCIAtlas/					
D442x10	4420	4368 (98.8%)	1.38	H-He, B-Ne, P-Ar, Br-Kr, I-Xe	Dispersion interactions
R739x5	3695	3435 (93.0%)	1.09	H-He, B-Ne, P-Ar, Br-Kr, I-Xe	Repulsive contacts
HB300SPXx10	3000	2990 (99.7%)	3.18	H, C-F, P-Cl, Br, I	Hydrogen bonds
SH250x10	2500	2410 (96.4%)	3.99	H, C-F, P-Cl, As-Br, I	Sigma-hole contacts
Total	153421	149079 (97.2%)		H-Ar, As-Kr, I-Xe	

Performance



- Near chemical accuracy on TAEs (W4-17 dataset).
- Competitive for some thermodynamics (GMTKN55 dataset).