Jacob's Elevator - Neural Network Exchange-Correlation Functionals

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Kohn-Sham Density Functional Theory

Hohenberg-Kohn theorem:

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$$\left(-\frac{1}{2}\boldsymbol{\nabla}^2 + v_{\mathrm{KS}}[n](r) \right)\phi_j = \epsilon_j \phi_j(r)$$

$$v_{\mathrm{KS}}[n](r) = v_{ext}(r) + v_H[n](r) + v_{xc}[n](r)$$

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$$E_v[n_{gs}] = T_{\mathrm{KS}}[n] + V[n] + U_H[n] + E_{xc}[n]$$

$$v_{xc}(r) = \frac{\delta E_{xc}}{\delta n(r)}$$

How to write E_{xc} and v_{xc}???

Jacobs Ladder

Heaven of chemical accuracy

Hybrid

Meta-GGA

GGA

LDA

Hartree World

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Machine Learning Approaches for DFT

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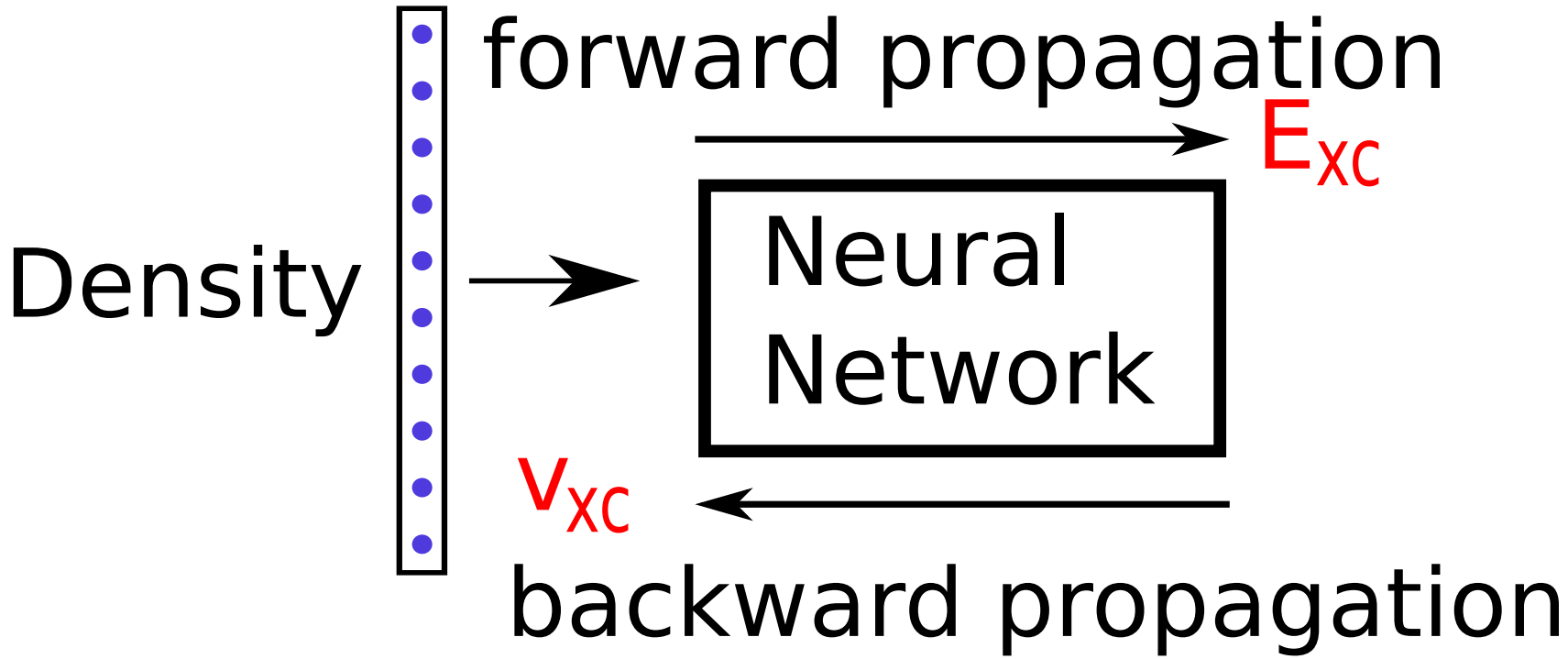
Previous approaches:

- Learning the exchange-correlation potential *Tozer et al. J.Chem.Phys. 1996*, *Nagai et al. J.Chem.Phys. 2018*
- Learning the non-interacting kinetic energy functional *Snyder et al. J.Chem.Phys 2013*, *Brockherde et al. Nat.Commun. 2017*

Challenges:

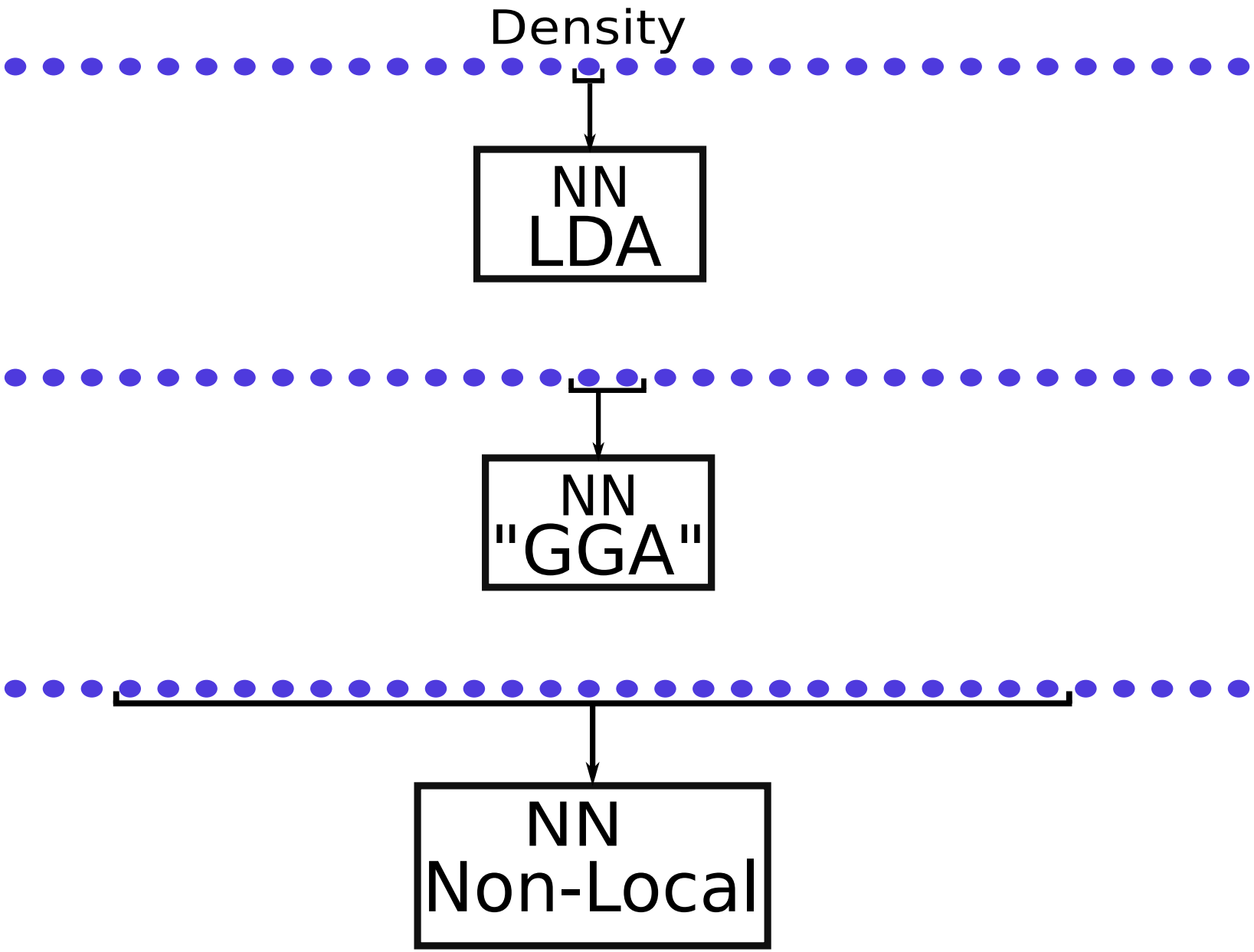
- Availability of the functional derivative (*Nagai et al. Arxiv 2019*)
- Notoriously difficult to develop accurate kinetic energy functional

First Approach



Calculate v_{xc} through autograd functionality of neural network frameworks

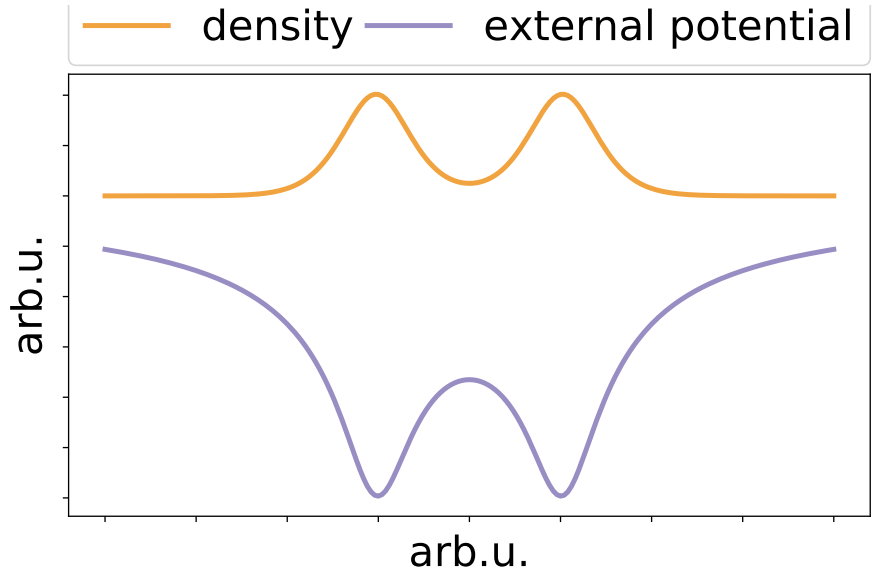
Locality



Data

- Exact calculations with 2 electrons in randomized 1D-external potentials
- Inversion of the Kohn-Sham system leads to E_{xc} and v_{xc} which is used for training

$$v_{ext} = -\frac{Z_1}{(\alpha_1 + (r - R_1)^2)^{\frac{1}{2}}} - \frac{Z_2}{(\alpha_2 + (r - R_2)^2)^{\frac{1}{2}}} - \frac{Z_3}{(\alpha_3 + (r - R_3)^2)^{\frac{1}{2}}}$$



Loss-Function

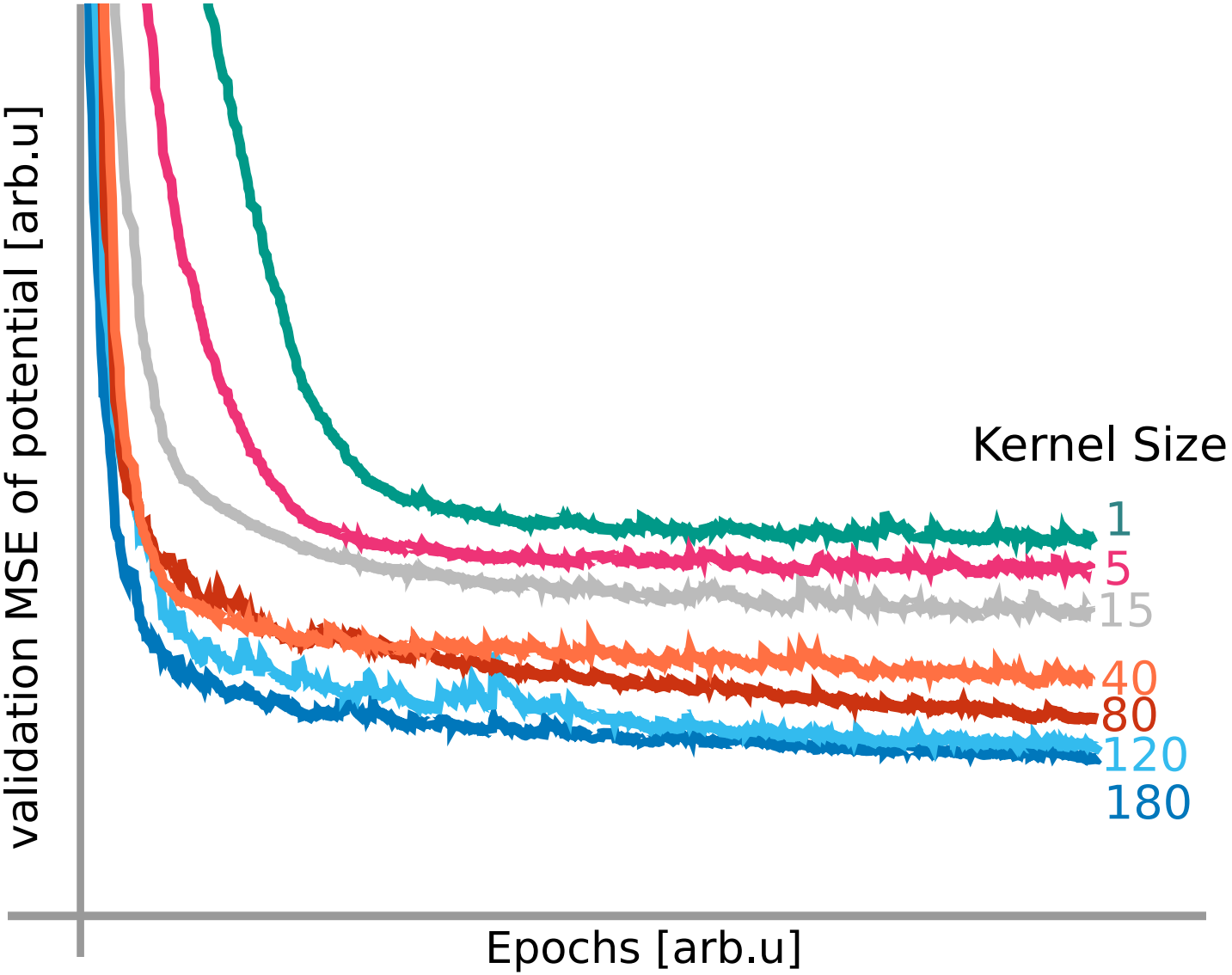
Goal achieve optimal total energy:

$$E_{total} = \sum E_i + E_{xc} - \int v_{xc}(r) n(r) dr - E_H$$

Loss function:

$$L(\theta, n_i) = \alpha \mathrm{MSE}(E_{xc}) + \beta MSE(v_{xc}) + \gamma \mathrm{MSE}(\frac{\mathrm{d}v_{xc}(r)}{\mathrm{dr}}) +$$
$$\delta \mathrm{MSE}(E_{xc} - \int v_{xc}(r) n(r) \mathrm{d}r)$$

Locality

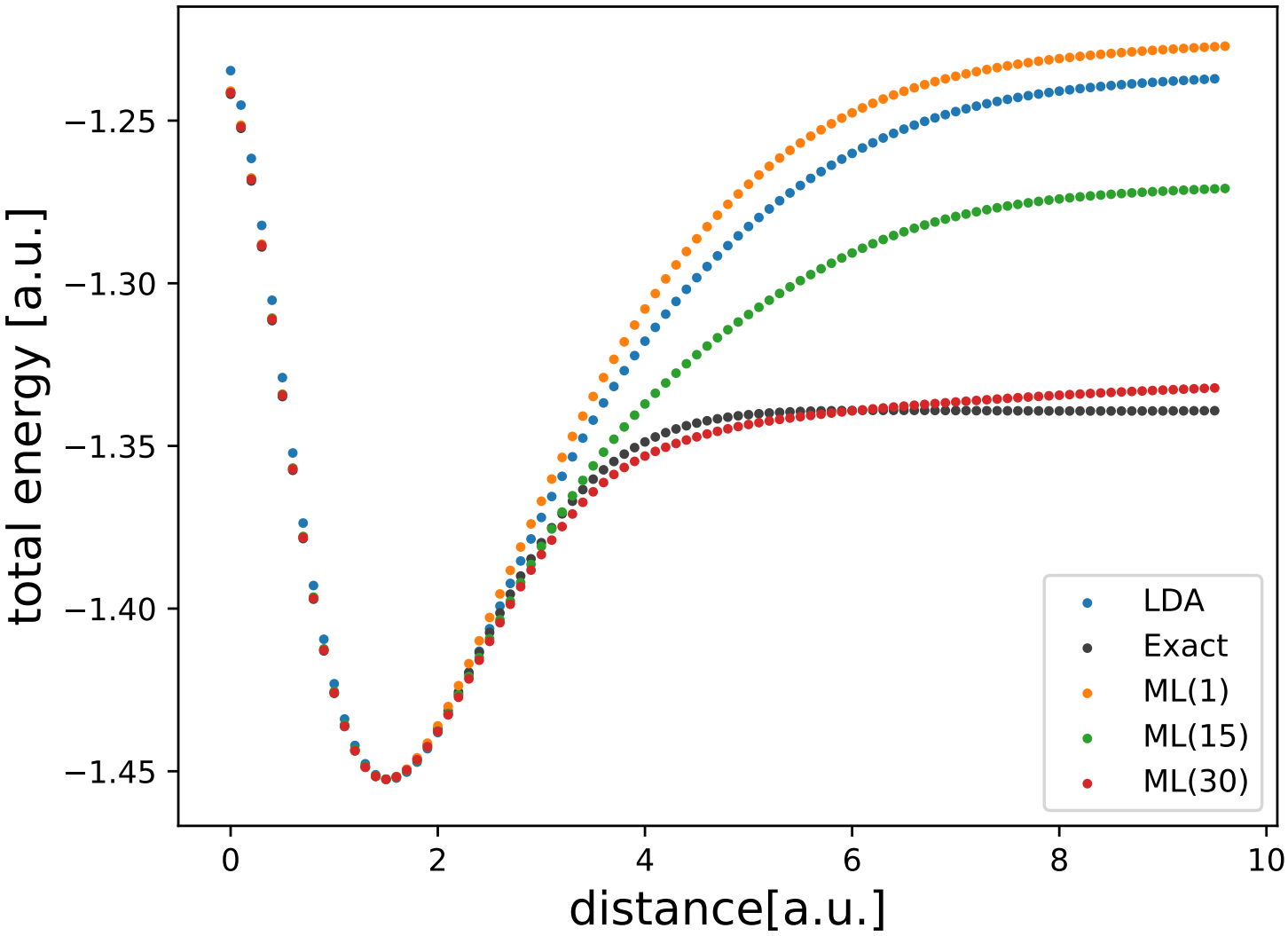

validation MSE of potential [arb.u]
Epochs [arb.u]
Kernel Size
1
5
15
40
80
120
180

Locality

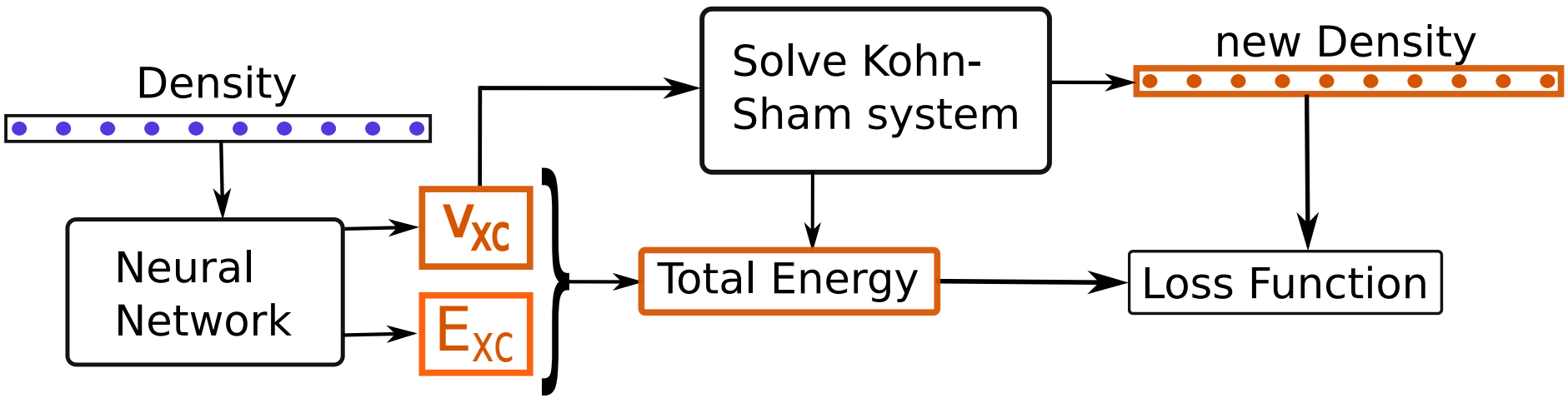
Relative energy errors for self consistent KS-calculations:

Kernel	MAE [a.u. 10^{-2}]	MAE(LDA)/MAE(ML)
DFT LDA	1.35	1.0
1	0.81	0.6
2	0.79	0.59
15	0.41	0.30
30	0.21	0.16
40	0.34	0.25

1D-H2-Dissociation



Self-consistent Training



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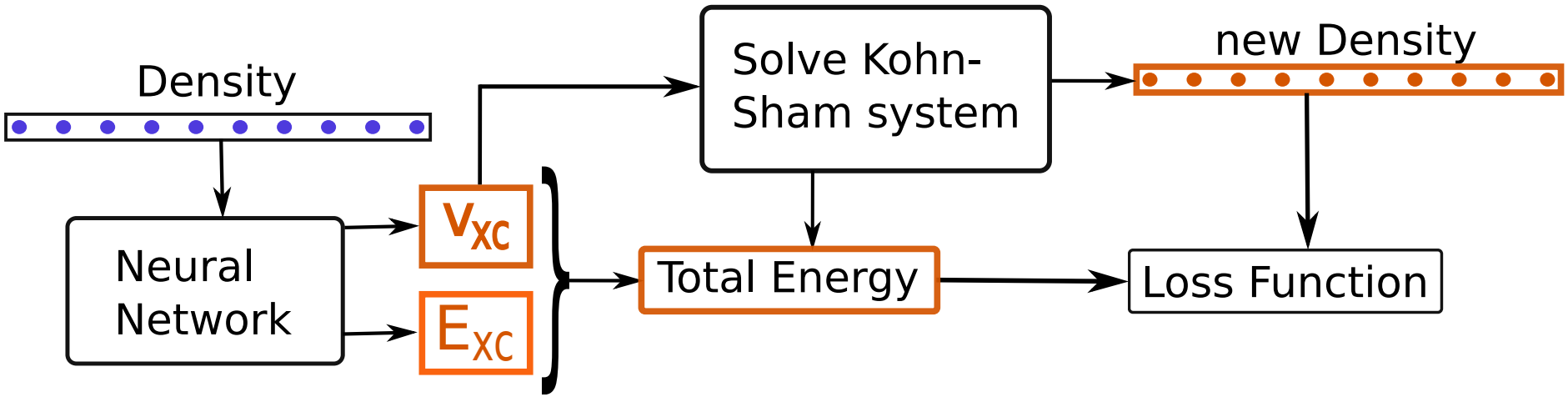

- Advantage: No inversion needed
- Disadvantage: More challenging to train

Summary

1. Modern machine learning frameworks allow the simple training of functionals and their derivatives
2. Neural networks facilitate the development of strongly non-local exchange-correlation functionals

Outlook:

- Development of 3D functionals for molecules and solids

Acknowledgement:
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