Machine learning the physical non-local exchange-correlation functional of
density-functional theory

Jonathan Schmidt, 1 Carlos L. Benavides-Riveros, 1,∗ and Miguel A. L. Marques1,†
1 Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, 06120 Halle (Saale), Germany
(Dated: August 20, 2019)

We train a neural network as the universal exchange-correlation functional of density-functional theory that simultaneously reproduces both the exact exchange-correlation energy and potential. This functional is extremely non-local, but retains the computational scaling of traditional local or semi-local approximations. It therefore holds the promise of solving some of the delocalization problems that plague density-functional theory, while maintaining the computational efficiency that characterizes the Kohn-Sham equations. Furthermore, by using automatic differentiation, a capability present in modern machine-learning frameworks, we impose the exact mathematical relation between the exchange-correlation energy and the potential, leading to a fully consistent method. We demonstrate the feasibility of our approach by looking at one-dimensional systems with two strongly-correlated electrons, where density-functional methods are known to fail, and investigate the behavior and performance of our functional by varying the degree of non-locality.

I. INTRODUCTION

Nowadays density functional theory (DFT) is the cornerstone of computational theoretical physics and quantum chemistry, as it provides the prevalent method for the calculation of the electronic structure of both solids and molecules. Based on the Hohenberg-Kohn theorems [1], DFT reformulates the quantum many-electron problem as a theory of the ground-state electronic density \( n(r) \). The success of DFT is to a large extent due to the existence of a system of non-interacting electrons (the Kohn-Sham system) that has the same ground-state density as the interacting electrons. This leads to the Kohn-Sham equations, a set of self-consistent equations for one-particle orbitals [2]. In such a formalism the ground-state (GS) energy can be expressed as:

\[
E_{\text{GS}} = \sum_i \epsilon_i + E_{\text{xc}}[n] - \int d^3 r \, v_{\text{xc}}(r)n(r) - E_H[n],
\]

where \( \epsilon_i \) are the eigenvalues of the Kohn-Sham Hamiltonian, \( v_{\text{xc}}(r) \) is the exchange-correlation potential, \( E_H[n] \) is the Hartree energy, and \( E_{\text{xc}}[n] \) is the exchange-correlation energy. The exchange-correlation potential is defined as the functional derivative of the universal exchange-correlation energy functional:

\[
v_{\text{xc}}(r) = \frac{\delta E_{\text{xc}}[n]}{\delta n(r)}. \tag{2}
\]

Due to the Hohenberg-Kohn existence theorems, if the exact exchange-correlation energy functional \( E_{\text{xc}}[n] \) is known, Kohn-Sham DFT then yields the exact ground-state energy and the exact ground-state electronic density.

Traditionally, “educated” formal expressions of the exchange-correlation energy functional have been proposed by a combination of theoretical insight, highly accurate Monte-Carlo [3] or quantum chemical simulations, or by fitting general expressions to experimental data. In general, functionals can be sorted according to Jacob’s ladder [4]: the lowest rung of the ladder is occupied by local-density approximations (LDA) that use solely single density points as inputs [5–7]. The second rung is occupied by generalized-gradient approximations (GGA) that include the gradient of the density [8, 9]. This is followed by the meta-GGAs [10] (that use the kinetic-energy density) and hybrid functionals [11–13] (that mix a fraction of non-local Fock exchange) on the subsequent rungs. Note that more than 500 of these functionals have been proposed in the past decades [14], although most of them with rather limited impact.

In spite of the success of DFT in dealing efficiently with electronic systems, it still suffers from stubborn quantitative and qualitative failures. For instance, barriers of chemical reactions, band gaps of materials, or molecular dissociation energies are usually underestimated [15]. Degenerate or near-degenerate states are also poorly described by DFT. While hybrid functionals can alleviate some of the problems of traditional semi-local functionals, they come at a greatly increased computational cost that limits severely the number and size of systems that can be researched. It is believed that many of these problems originate in the delocalization and static correlation errors which plague approximate functionals [16–18]. Roughly speaking, the delocalization error refers to the tendency of DFT functionals to spread out the electron density, while the static correlation arises from the difficulty of describing degenerated states with a single Slater determinant [19].

More recently, machine learning (ML) has revolutionized many fields of computational sciences, such as image or speech recognition [20, 21], and has found countless applications in material science [22–24]. Within DFT,
the application of ML techniques to the formulation of density functionals has already a long history [25]. In 2012, an ML approximation for the kinetic energy functional $T_{\text{k}}[n]$ was constructed for a system of noninteracting spinless fermions [26, 27]. In order to exploit the Hohenberg-Kohn density-potential map, an ML model was later trained to learn the fundamental relation of DFT between external potentials and electronic densities [28]. These works focused mainly on developing functionals for the total energy or the non-interacting kinetic energy to facilitate orbital-free DFT calculations. More recently, some works have addressed the problem of training the exchange-correlation potential [29–33]. However, this line of research has been limited by the fact that the exchange-correlation potential was not obtained from the exchange-correlation energy through the functional derivative of Eq. (2).

It is true that one can find in the literature a series of approximations to the exchange-correlation functionals that do not fulfill Eq. (2). For example, the Krieger-Lee-Lafraite approximation [34] breaks this connection in order to simplify the implementations of orbital functionals using the optimized effective method [35, 36]. Sometimes, it is also convenient to approximate directly the potential (e.g., in the van Leeuwen-Baerends GGA from 1994 [37] or the modified Becke-Johnson potential [38, 39]), leading again to expressions that do not obey Eq. (2). These so-called “stray” functionals [40] have found some important applications. For example, the modified Becke-Johnson is one of the most successful functionals to calculate electronic band-gaps [41]. Unfortunately, they are also found to break a series of exact theorems and conditions [40], severely limiting their universality and range of applicability. By and large, it is highly advantageous to develop consistent functionals that obey the important Eq. (2).

Modern ML frameworks, like pytorch [42] and tensorflow [43], allow for automatic differentiation with respect to any parameter. Recently, Nagai et al. used this functionality to train exchange-correlation potentials for molecules [44]. They trained neural networks through a Monte-Carlo updating scheme to reproduce accurate energies and densities of molecules. The functionals by Nagai and coauthors follow the traditional approaches of an LDA, GGA, meta-GGA, and add a related near-region approximation. Although a clear step forward, using traditional forms for the exchange-correlation functional is unlikely to lead to fundamentally better, disruptive approximations to the exchange-correlation functionals. New paradigms have to be sought in order to unleash the power of ML techniques to its full extent.

In this paper we use the auto-differentiation functionality to train neural-network exchange-correlation functionals through back propagation. The networks are trained to reproduce not only the correct exchange-correlation energy $E_{\text{xc}}$, but also the exchange-correlation potential $v_{\text{xc}}(r)$ consistently as its functional derivative with respect to the density. Consequently, the resulting functional allows for self-consistent calculations and can easily be integrated into existing Kohn-Sham DFT frameworks. Furthermore, these functionals can be made highly non-local by using the information of the density in a finite neighborhood as input to the neural network, allowing for far more non-locality than traditional LDA or GGA functionals, despite having the same computational scaling with system size. Therefore, this approach promises to alleviate the delocalization problems of DFT and to improve its accuracy without the computational expense of hybrid functionals. To demonstrate the feasibility of this approach, we developed an ML functional for the exchange-correlation energy and exchange-correlation potential based on exact results for two electrons in one-dimension (1D).

The paper is organized as follows. Sect. II presents the details of the dataset, training process, and neural networks. The exact dependence of the functional on the degree of locality and its behavior is discussed in Sec. III, as well as our results for the 1D homogeneous electron gas and the H₂ molecule along the dissociation path. Finally, in Sect. IV we discuss our conclusions and future research directions.

II. MACHINE LEARNING DETAILS

A. Data

The training data was produced by solving exactly the one-dimensional two-electron problem in the external potential generated by up to three different nuclei. Softening the Coulomb interaction,

$$\frac{1}{r} \rightarrow \frac{1}{\sqrt{1 + x^2}},$$

we obtain the 1D Hamiltonian driven by the interaction of the two electrons, namely,

$$H(x_1, x_2) = -\sum_{i=1}^{2} \left[ \frac{1}{2} \partial_x^2 + v(x_i) \right] + \frac{1}{\sqrt{1 + (x_1 - x_2)^2}},$$

where the external potential is given by the superposition of three potentials,

$$v(x) = \sum_{k=1}^{3} \frac{Z_k}{\sqrt{1 + (x - a_k)^2}}.$$  \hspace{1cm} (4)

The total charge of the nuclei $Z = \sum_k Z_k$ is equal to 2 or 3. Since the ground-state problem of the Hamiltonian $H(x_1, x_2)$ can be treated as a one-particle problem in two dimensions, the problem can be solved exactly.

We sampled 20 000 systems and calculated their exact ground-state energy and ground-state electronic density. We used a grid spacing of 0.1 a.u., and a box size of 20 a.u., leading to a grid with 201 points. The nuclei positions $a_i$ in Eq. (4) were normally distributed with zero mean and variance of 4 a.u. We then solved
the corresponding inverse Kohn-Sham problem in OCTOPUS [45] to find the exact exchange-correlation energy and potential. Since the inversion is known to be numerically unstable [46], we removed outliers that result from these instabilities. We used up to 12 800 of these systems for training, 6 400 for validation during the training, and 2 000 systems for the test set. Furthermore, training was considerably improved when removing outliers with $E_{\text{xc}} \succ -0.55$ a.u. from the training set. No outliers were removed from the test set to allow for a completely unbiased evaluation of the functionals.

In general, one would have to double the data by mirroring the systems to learn the correct symmetry. However, in this specific case one can simply build the symmetry directly into the neural network functional, as explained in the next subsection.

B. Topology of the neural network

Our ML functional scans the density of the total system, as illustrated in Fig. 1. The density in a neighborhood of the test point is used as the input for the neural network, that then outputs a local exchange-correlation energy density. Specifically, the network takes a certain number of density points as input, which we call $\kappa$, the kernel size. This is the degree of locality of the ML functional. At the borders of the system the density is padded with $\kappa-1=5$ zeros. Starting at one of the borders, the network calculates the local exchange-correlation energy for $\kappa$ points. In the next step, the input of the network is moved by one grid point and it is evaluated again.

![FIG. 1. Structure of the ML functional in 1D with degree of locality equal to $\kappa=6$ (see text). At the borders, the density is padded with $\kappa-1=5$ zeros. Starting at one of the borders, the network calculates the local exchange-correlation energy for $\kappa$ points. In the next step, the input of the network is moved by one grid point and it is evaluated again.](image)

The training for the exchange-correlation energy converges quite fast after a few hundred epochs. The convergence of the potential can take thousands of epochs depending on the training set and batch size. At each training step the model was saved if it improved the validation error for the potential. The model with the low-
TABLE I. Mean absolute errors (MAE) for the total energy in self-consistent calculations for various kernel sizes of our ML-DFT functional, relative to the error of the one-dimensional LDA of Ref. 52. For reference, the mean absolute error of the LDA of Ref. 52 is $1.4 \times 10^{-2}$ a.u.

| Kernel-size | MAE(ML)/MAE(LDA)[%] |
|-------------|----------------------|
| LDA         | 100                  |
| 1           | 38.1                 |
| 15          | 21.8                 |
| 30          | 8.2                  |
| 60          | 8.2                  |
| 120         | 7.1                  |
| 180         | 6.5                  |

The results for the predicted total energies of the test set (relative to the energy of $H_2$ at its equilibrium distance) are presented in Table I. Our ML-LDA (i.e., the functional with kernel size $\kappa = 1$) already performs better than the traditional LDA. As the ML-LDA was trained to reproduce the exchange-correlation energy of heterogeneous systems while traditional LDAs are “trained” for constant densities the difference in performance is not surprising. Increasing the kernel size leads to a monotonical decrease of the error, and improves the results by more than a factor of six for the larger sizes. The optimal kernel size is, in our opinion, around 30 (i.e., 3 a.u.), as larger kernels do not provide a significant advantage. Furthermore, some of the functionals with larger kernel sizes also demonstrate unphysical behavior (see Sec. III B).

Ultimately, the more non-local the functional is, the higher the complexity and the larger the number of parameters. This reason, together with the need to represent more long-range interactions that are based on different physical principles (such as van der Waals interaction), makes the training considerably more difficult. One approach to circumvent this problem is to keep the non-locality limited to ranges on the scale of molecular bonds. This allows for simpler training and still includes most of the non-locality that is required for the exchange-correlation energy. Another possibility would be to enlarge the non-locality by increasing the architectural complexity of the functional.

Although the scaling of the networks to realistic three-dimensional systems is non-trivial, we expect that both the number of trainable parameters and the density points in the training set will grow cubically when transitioning to three-dimensional systems. In this sense, we expect a similar demand for training data as in 1D. Furthermore, realistic systems are usually far larger and therefore provide more “local” training samples per system for the neural network. Recent research by Nagai and coauthors points in the same direction [44]. Indeed, they only required a few sample molecules and used far more parameters to learn a much more local (and in this sense simpler) functional than the ones used here.
B. H₂ dissociation

In the previous subsection we showed that our ML functionals are able to accurately reproduce the test samples. Now, we go a step further and test how our functionals perform self-consistently in a couple of paradigmatic cases. In Fig. 2 we present our calculations for the H₂ molecule in 1D along the dissociation path with functionals of varying non-locality in comparison to the exact result. The curves in Fig. 2 are shifted to have the same equilibrium energies. As is well-known, the traditional LDA completely fails to produce the correct dissociation limit [52]. The same behaviour is observed for the ML LDA (with a kernel size of 1). Remarkably, using increasingly more non-local functionals, we can reduce the relative energy error to 3.2% of the LDA error.

It is obvious that even the functional with a kernel size of 30 will start failing above a certain distance. This is a conceptual problem of local KS-DFT and can only be alleviated and not eliminated in our approach. It can already be considered a success that our functionals are able to reproduce the dissociation curve reasonably well far beyond their own degree of non-locality. Yet it has to be noted that not all functionals performed that well. Some of the functionals with larger kernels failed to reproduce a physical behavior with respect to the dissociation distance and produced multiple local minima and maxima. Despite these problems, they still return the correct equilibrium distance and on average far better energies than the LDA. This unphysical behavior of some functionals just stresses the fact that a rigorous validation on a multitude of different systems will be essential to arrive at a working functional. It has to be noted that a larger training set and a longer training time was far more beneficial for this validation than for example the average error. As systems similar to the dissociated molecule are most likely outliers of the training data this is not surprising. Note that the need for more training data can however be avoided by active learning and a thoughtful construction of the training set.

C. Homogeneous-electron gas

In this section we study the homogeneous-electron gas, a model system that is used in the construction of the majority of exchange-correlation functionals. We can simulate this system with our neural networks by providing them with a constant electronic density as input. The results can then be compared to the numerically exact values for the energy density as obtained, for example, from quantum Monte-Carlo simulations [52]. Our results are depicted in Fig. 3 as a function of the Wigner-Seitz radius \( r_s = 1/2n \).

Some difficulties for our neural-network functionals are evident. First, the functionals were trained only for systems with a specific size while the homogeneous electron gas is, in fact, an infinite periodic system. Second, as the histogram in Fig. 3 illustrates, the training data does not contain almost any samples with high densities \( r_s < 1 \). Naturally, the availability of training data similar to the homogeneous electron gas is even more important for the more non-local functionals as they take into account larger regions of space.

The first challenge results in the fact that the non-zero biases in each layer cause the neural networks to output a non-zero value for zero density. When training for different system sizes there are several ways to avoid this failure. First one could solve the problem by adding systems padded with different amounts of zeros at the border to force the neural network to learn the correct relationship. As a second possibility, one could force all biases of all layers to zero, however, this would severely limit the expressibility of the networks. To circumvent this problem, and in order to compare the behavior of the energy with respect to the Wigner-Seitz radius, we shifted the curves in Fig. 3 to yield zero energy for zero density.

Despite the small amount of training data at high density, functionals with larger kernel sizes still generalize on average far better to the homogeneous electron gas. While it is not obvious whether this will remain true in three dimensions, it is nevertheless promising that the extra non-local information in the larger kernels might help the functionals to be generalized. Constant densities will be an essential feature of a functional for solid state physics. Fortunately, exact training data in the form of quantum Monte-Carlo calculations already exists for this
IV. CONCLUSIONS AND OUTLOOK

We have demonstrated the viability of learning an exchange-correlation potential, via the differentiation of the exchange-correlation energy in a physically consistent manner. This procedure allows for standard self-consistent Kohn-Sham calculations. From the presented data, it is evident that neural-network functionals trained on the exchange-correlation potential and energy have the potential to be far more precise than previous local DFT functionals. Increasing the non-locality of the functional allows for an extremely precise treatment of the electronic interaction on the scale of at least a few atomic units and, to a certain extent, even solve long-standing problems in DFT like e.g. molecular dissociation.

For simplicity, we trained a neural network to the one-dimensional two-electron problem in the external potential generated by up to to three nuclei. Training three-dimensional systems will have to be accomplished by using data obtained with coupled-cluster, full configuration-interaction, or quantum Monte Carlo. While sufficient data to train a universal functional still has to be created, exchange-correlation energies and potentials for a few small molecules already exists and can provide a good starting point. The density representation on a grid is unfortunately not feasible for more general systems, as grid sizes and forms will vary. However, we think this can easily be circumvented by representing the density locally in some basis sets (e.g., Gaussians).

Finally, there is already a long history within DFT in the development of empirical functionals \([31, 32, 53–55]\). The machine learning paradigm allows us to drastically increase the amount of data used for the training and the complexity of these functionals. Including known exact conditions of the exchange-correlation functional in the learning process as constraints in the minimization will still be helpful \([56]\) and provide further conditions for validation. Furthermore, as the functionals will have to work in practically every density environment, the importance of an extremely in-depth validation cannot be overstated and will be essential to arrive at a widely used functional.

ACKNOWLEDGMENTS

We acknowledge partial support from the German DFG through project MA-6786/1.

[1] P. Hohenberg and W. Kohn, “Inhomogeneous electron gas,” Phys. Rev. 136, B864–B871 (1964).
[2] W. Kohn and L. J. Sham, “Self-consistent equations including exchange and correlation effects,” Phys. Rev. 140, A1133–A1138 (1965).
[3] D. M. Ceperley and B. J. Alder, “Ground state of the electron gas by a stochastic method,” Phys. Rev. Lett. 45, 566 (1980).
[4] J. P. Perdew and K. Schmidt, “Jacob’s ladder of density functional approximations for the exchange-correlation energy,” AIP Conf. Proc. 577, 1 (2001).
[5] S. H. Vosko, L. Wilk, and M. Nusair, “Accurate spin-dependent electron liquid correlation energies for local spin density calculations: a critical analysis,” Can. J. Phys. 58, 1200 (1980).
[6] L. A. Cole and J. P. Perdew, “Calculated electron affinities of the elements,” Phys. Rev. A 25, 1265 (1982).
[7] J. P. Perdew and Y. Wang, “Accurate and simple analytical representation of the electron-gas correlation energy,” Phys. Rev. B 45, 13244 (1992).
[8] J. P. Perdew, K. Burke, and M. Ernzerhof, “Generalized gradient approximation made simple,” Phys. Rev. Lett. 77, 3865 (1996).
[9] J. P. Perdew, K. Burke, and Y. Wang, “Generalized gradient approximation for the exchange-correlation hole of a many-electron system,” Phys. Rev. B 54, 16533 (1996).
[10] J. Sun, R. C. Remsing, Y. Zhang, Z. Sun, A. Ruzsinszky, H. Peng, Z. Yang, A. Paul, U. Waghmare, X. Wu, M. L. Klein, and J. P. Perdew, “Accurate first-principles structures and energies of diversely bonded systems from an efficient density functional,” Nat. Chem. 8, 831 (2016).
[11] A. D. Becke, “A new mixing of Hartree–Fock and local density-functional theories,” J. Chem. Phys. 98, 1372 (1993).
[12] J. P. Perdew, M. Ernzerhof, and K. Burke, “Rationale for mixing exact exchange with density functional approximations,” J. Chem. Phys. 105, 9982 (1996).
[13] C. Adamo and V. Barone, “Toward reliable density functional methods without adjustable parameters: The PBE0 model,” J. Chem. Phys. 110, 6158 (1999).
[14] A. P. E. Nordlander and S. A. Rice, “An overview of density functional methods,” SoftwareX 1, 1 (2016).
power polynomial kernel PCA,” in Advances in Neural Networks (Springer Berlin Heidelberg, 2010) pp. 144–151.

[21] A. Waibel and K.-F. Lee, eds., Readings in Speech Recognition (Morgan Kaufmann, 1990).

[22] K. T. Butler, D. W. Davies, H. Cartwright, O. Isayev, and A. Walsh, “Machine learning for molecular and materials science,” Nature 559, 547 (2018).

[23] J. Schmidt, M. R. G. Marques, S. Botti, and M. A. L. Marques, “Recent advances and applications of machine learning in solid-state materials science,” npj Comput. Mat. 5 (2019).

[24] L. M. Ghiringhelli, J. Vybiral, S. V. Levchenko, C. Draxl, and M. Scheffler, “Big data of materials science: Critical role of the descriptor,” Phys. Rev. Lett. 114, 105503 (2015).

[25] D. J. Tozer, V. E. Ingamells, and N. C. Handy, “Exchange-correlation potentials,” J. Chem. Phys. 105, 9200 (1996).

[26] J. C. Snyder, M. Rupp, K. Hansen, K-R. Müller, and K. Burke, “Finding density functionals with machine learning,” Phys. Rev. Lett. 108, 253002 (2012).

[27] J. C. Snyder, M. Rupp, K. Hansen, L. Blooston, K-R. Müller, and K. Burke, “Orbital-free bond breaking via machine learning,” J. Chem. Phys. 139, 224104 (2013).

[28] F. Brockherde, L. Vogt, L. Li, M. E. Tuckerman, K. Burke, and K.-R. Müller, “Bypassing the Kohn-Sham equations with machine learning,” Nat. Commun. 8, 872 (2017).

[29] Q. Liu, J. C. Wang, P. L. Du, L. H. Hu, X. Zheng, and G. Chen, “Improving the performance of long-range-corrected exchange-correlation functional with an embedded neural network,” J. Phys. Chem. A 121, 7273 (2017).

[30] R. Nagal, R. Akashi, S. Sasaki, and S. Tsumeyuki, “Neural-network Kohn-Sham exchange-correlation potential and its out-of-training transferability,” J. Chem. Phys. 148, 241737 (2018).

[31] K. T. Lundgaard, J. Wellendorff, J. Voss, K. W. Jacobsen, and T. Bliagaard, “mBEEF-vdW: Robust fitting of error estimation density functionals,” Phys. Rev. B 93, 235162 (2016).

[32] J. Wellendorff, K. T. Lundgaard, A. Møgelhøj, V. Petzold, D. D. Landis, J. K. Norskov, T. Bliagaard, and K. W. Jacobsen, “Density functionals for surface science: Exchange-correlation model development with bayesian error estimation,” Phys. Rev. B 85, 235149 (2012).

[33] L. Li, T. E. Baker, S. R. White, and K. Burke, “Pure density functional for strong correlation and the thermodynamic limit from machine learning,” Phys. Rev. B 94, 245129 (2016).

[34] J. B. Krieger, Y. Li, and G. J. Iafrate, “Derivation and application of an accurate Kohn-Sham potential with integer discontinuity,” Phys. Lett. A 146, 256 (1990).

[35] R. T. Sharp and G. K. Horton, “A variational approach to the unipotential many-electron problem,” Phys. Rev. 90, 317–317 (1953).

[36] J. D. Talman and W. F. Shadwick, “Optimized effective atomic central potential,” Phys. Rev. A 14, 36–40 (1976).

[37] R. van Leeuwen and E. J. Baerends, “Exchange-correlation potential with correct asymptotic behavior,” Phys. Rev. A 49, 2421 (1994).

[38] A. D. Becke and E. R. Johnson, “A simple effective potential for exchange,” J. Chem. Phys. 124, 221101 (2006).

[39] F. Tran and P. Blaha, “Accurate Band Gaps of Semiconductors and Insulators with a Semilocal Exchange-Correlation Potential,” Phys. Rev. Lett. 102 (2009).

[40] A. P. Gaiduk and V. N. Staroverov, “How to tell when a model Kohn–Sham potential is not a functional derivative,” J. Chem. Phys. 131, 044107 (2009).

[41] P. Borlido, T. Aull, A. W. Huran, F. Tran, M. A. L. Marques, and S. Botti, “Large-Scale Benchmark of Exchange–Correlation Functionals for the Determination of Electronic Band Gaps of Solids,” J. Chem. Theory Comput. 0, null (2019).

[42] A. Paszke, S. Gross, S. Chintalal, G. Chanen, E. Yang, Z. DeVito, Z. Lin, A. Desmaison, L. Antiga, and A. Lerer, “Automatic differentiation in pytorch,” in NIPS 2017 Autodiff Workshop: The Future of Gradient-based Machine Learning Software and Techniques (2017).

[43] M. Abadi, A. Agarwal, P. Barham, E. Brevdo, Z. Chen, C. Citro, G. S. Corrado, A. Davis, J. Dean, M. Devin, S. Ghemawat, I. Goodfellow, A. Harp, G. Irving, M. Isard, Y. Jia, R. Jozefowicz, L. Kaiser, M. Kudlur, J. Levenberg, D. Mané, R. Monga, S. Moore, D. Murray, C. Olah, M. Schuster, J. Shlens, B. Steiner, I. Sutskever, K. Talwar, P. Tucker, V. Vanhoucke, V. Vasudevan, F. Viégas, O. Vinyals, P. Warden, M. Wattenberg, M. Wicke, Y. Yu, and X. Zheng, “TensorFlow: Large-scale machine learning on heterogeneous systems,” https://tensorflow.org/ (2015).

[44] R. Nagai, R. Akashi, and O. Sugino, “Completing density functional theory by machine-learning hidden messages from molecules,” arXiv:1903.00238 (2019).

[45] X. Andrade, D. Strubbe, U. De Giovannini, A. H. Larsen, M. J. T. Oliveira, J. Alberdi-Rodriguez, A. Varas, I. Theophilou, N. Helbig, M. J. Verstraete, L. Stella, F. Nogueira, A. Aspuru-Guzik, A. Castro, M. A. L. Marques, and A. Rubio, “Real-space grids and the Octopus code as tools for the development of new simulation approaches for electronic systems,” Phys. Chem. Chem. Phys. 17, 31371 (2015).

[46] D. S. Jensen and A. Wasserman, “Numerical methods for the inverse problem of density functional theory,” Int. J. Quantum Chem. 118, e25425 (2018).

[47] D.-A. Clevert, T. Unterthiner, and S. Hochreiter, “Fast and accurate deep network learning by exponential linear units (elus),” arXiv:1511.07289 (2015).

[48] “Ignite,” https://github.com/pytorch/ignite (2018).

[49] D. P. Kingma and J. Ba, “Adam: A method for stochastic optimization,” arXiv:1412.6980 (2014).

[50] D. Masters and C. Luschi, “Revisiting small batch training for deep neural networks,” arXiv:1804.07612 (2018).

[51] I. Goodfellow, Y. Bengio, and A. Courville, Deep Learning (MIT Press, 2016) http://www.deeplearningbook.org.

[52] N. Helbig, J. I. Fuku, M. Casula, M. J. Verstraete, M. A. L. Marques, I. V. Tokatly, and A. Rubio, “Density functional theory beyond the linear regime: Validating an adiabatic local density approximation,” Phys. Rev. A 83, 032503 (2011).

[53] H. S. Yu, X. He, and D. G. Truhlar, “MN15-L: A New Local Exchange-Correlation Functional for Kohn-Sham Density Functional Theory with Broad Accuracy for Atoms, Molecules, and Solids,” J. Chem. Theory Comput. 12, 1280 (2016).

[54] H. S. Yu, X. He, S. L. Li, and D. G. Truhlar, “MN15: A Kohn–Sham global-hybrid exchange–correlation density
functional with broad accuracy for multi-reference and single-reference systems and noncovalent interactions,” Chem. Sci. 7, 5032–5051 (2016).

[55] N. Mardirossian and M. Head-Gordon, “ωB97M-V: A combinatorially optimized, range-separated hybrid, meta-GGA density functional with VV10 nonlocal correlation,” J. Chem. Phys. 144, 214110 (2016).

[56] J. Hollingsworth, T. E. Baker, and K. Burke, “Can exact conditions improve machine-learned density functionals?” J. Chem. Phys. 148, 241743 (2018).