

Machine learning semilocal exchange-correlation functionals for Kohn-Sham density functional theory of the Hubbard model

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The Hubbard model provides a test bed to investigate the complex behavior arising from electron-electron interaction in strongly correlated systems and naturally emerges as the foundation model for lattice density functional theory (DFT). Similarly to conventional DFT, lattice DFT computes the ground-state energy of a given lattice model, by minimizing an energy functional of the on-site occupations. The energy then comprises a contribution that depends on the external potential and a universal contribution describing the kinetic and the electron-electron interaction energy. Here we use machine learning to construct a class of scalable “semilocal” exchange-correlation functionals with an arbitrary degree of nonlocality for the one-dimensional spinfull Hubbard model. Then, by functional derivative we construct an associated Kohn-Sham potential, that is used to solve the associated Kohn-Sham equations. This effectively forms a close workflow for machine-learning Kohn-Sham lattice DFT. After having investigated how the accuracy of the semilocal approximation depends on the degree of nonlocality, we use our Kohn-Sham scheme to compute the polarizability of linear chains, either homogeneous or disordered, approaching the thermodynamic limit.

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I. INTRODUCTION

Over the last few decades, density functional theory (DFT) has become the computational method of choice across several different fields, going from quantum chemistry to material science. Today, DFT serves as the primary tool for calculating the electronic structure of both solids and molecules, thanks to its foundation in the rigorously established Hohenberg-Kohn theorems [1], to the practical Kohn-Sham functional-minimization scheme [2], and to the numerous available efficient numerical implementations [3]. The Hohenberg-Kohn theorems establish the existence of a universal energy functional, $F[n]$, of the electron charge density, $n(\mathbf{r})$, functional that describes the kinetic and the electron-electron repulsion energy. This adds to the energy associated with the external potential to give a total-energy functional, which presents its minimum at the ground-state density. Thus, in principle, one can solve any ground-state electronic quantum-mechanics problem, by simply scanning through all the legitimate charge densities, in the search of the functional minimum. This conceptually simple strategy, known as orbital-free DFT [4], is however practically impossible.

Two main obstacles prevent the direct minimization of $F[n]$. First, the exact functional is unknown, so that an exact solution of the problem is not available. This is partially mitigated by the possibility to break the functional into three contributions, (i) the kinetic energy of a non-interacting system having the same density, $T_0[n]$, (ii) the classical Coulomb energy (Hartree energy) and (iii) the so-called exchange-correlation energy, $E_{xc}[n]$. This last term describes the nonclassical electron-electron interaction and the correction to the noninteracting kinetic energy arising from electron correlation. Importantly, there are many approximations to $E_{xc}[n]$ that allows one to perform accurate calculations. Hence, the DFT problem, at least in an approximated form, is solvable. Yet, orbital-free DFT remains challenging, since the exact density-functional form for the noninteracting kinetic energy is unknown and there are no accurate and universal approximations for it working across different external potentials.

As a consequence, practically any numerical implementation of DFT follows the Kohn-Sham (KS) procedure. This consists in mapping the interacting many-body problem onto a fictitious single-particle one where, by construction, the two problems share the same ground-state energy and density. The potential associated with the single-particle problem is constructed from $F[n]$ by functional derivative and it is itself density dependent. As such, the noninteracting kinetic energy is computed exactly, but the solution of the KS equations must be iterative. Although, in principle, the exact functional depends on the electron density everywhere in space, namely, it is nonlocal, the most common approximations to $E_{xc}[n]$ are local or semilocal. For instance, the local density approximation (LDA) assigns the exchange-correlation energy at a

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given point to that of the homogeneous electron gas computed for the density at that point [2]. More sophisticated and accurate approximations can then be constructed by expanding the functional dependence of the exchange-correlation energy to the density gradient and higher derivatives, following a systematic Jacob's ladder [5]. These work well across a broad range of electronic-structure types, but they usually fail for systems presenting strongly correlated electrons [6]. The systematic construction of nonlocal functionals that satisfy the known conditions of exact DFT is highly nontrivial and their deployment in calculations for real systems is extremely numerically demanding.

Some of these challenges motivated the construction of DFT for lattice models, namely, of lattice density functional theory (LDFT) [7,8]. To fix the idea, let us consider the one-dimensional (1D) single-orbital Hubbard model [9]. LDFT then translates the Hohenberg-Kohn theorems by replacing the electron density with the site occupation. One can then define a universal functional that depends on the entire occupation manifold, namely, on the average occupation of each site comprising the model. Although one should be careful to transfer DFT concepts to LDFT [10–13], the study of lattice models has a number of advantages. In particular, in LDFT the charge density has a simple single representation, some exact limiting solutions are known [14], and, for relatively small systems, exact numerical results can be obtained at a moderate computational cost. As such, LDFT can be seen as a simple playground to investigate some fundamental questions of DFT. For instance, by using the Bethe ansatz solution of the Hubbard model one can construct LDA functionals [15,16] and then investigate the response of strongly correlated systems to electric [17] and magnetic fields [18], their susceptibility [19], the formation of the Mott gap [20] and quantum transport [21].

An interesting aspect of any functional theory is given by the possibility of combining it with machine-learning (ML) techniques. In fact, within conventional DFT, ML appears ideal to construct numerical mappings between the electron density and the energy [22–29], between the external potential and the electron density [30–32], between the electron density and the ground-state wave function [33] or between the electron density and an observable [34]. There are also attempts to learn, through symbolic regression, analytic expressions of unknown functionals, such as in the case of the 1D noninteracting kinetic-energy density [35]. Moving to lattice models, we have shown that a numerically exact functional can be constructed for the spin-full Hubbard model in one dimension [36]. This depends on the site occupations of the entire system, namely, it is a fully nonlocal functional, and it satisfies both Hohenberg-Kohn theorems. Unfortunately, since the energy depends on the entire site occupation manifold, a functional trained on a N -site system cannot be used to predict the energy of systems of other size. We have then avoided such drawbacks by constructing semilocal ML functionals, this time for the spinless Hubbard model [37]. In both these two examples, the energy minimization is performed in an orbital-free-DFT fashion. A similar approach has been proposed for the study of Ising models [38]. It has also been shown that one can construct a potential-functional theory, mapping directly from the potential to the density [39].

To overcome the use of orbital-free-DFT for lattice models, so that functionals become fully transferrable between systems of different size, we have here taken an alternative approach. Namely, we demonstrate that a Kohn-Sham potential for the spin-full 1D Hubbard model can be obtained from the numerical ML energy functional by functional derivative. This is constructed semilocally so that it can be trained on relatively small systems and then used for much larger ones. In a nutshell the present work shows that Hohenberg-Kohn LDFT can be solved in a Kohn-Sham framework, where the kinetic energy for the noninteracting system is exactly defined regardless of the system size. With this at hand, we are able to investigate relevant quantities as they approach the thermodynamic limit, namely, for a large number of sites. In particular, we concentrate here on the charge density response of the one-dimensional Hubbard model in the metallic state and in the presence of disorder, for which we evaluate the polarizability.

The paper is structured as follows: In Sec. II we present our methodology, by setting up the problem, discussing how to generate the target dataset, and by explaining how we train the ML functional. Our presentation of the results begins in Sec. III A by demonstrating the performance of our KS equations and the accuracy of the constructed KS potential. This is followed by an analysis of the nonlocality of the functional in Sec. III B. We then proceed to look at some applications of the trained functional. In Sec. III C we apply it to solve systems with a filling factor outside the range used for the training. Then, we analyze the (deviation from) piecewise-linearity of the energy at fractional electron filling and the associated derivative discontinuity [see Sec. III D]. Finally, in Sec. III E we investigate the response of the charge density to an electric field and the ability of the model to solve systems with thousands of lattice sites. Conclusions are drawn in Sec. IV.

II. METHOD

A. Kohn-Sham LDFT setup for Hubbard model

Our analysis is based on the one-dimensional, single-orbital, spin-full Hubbard model [9], described by the following Hamiltonian,

$$\hat{H}_U = \hat{T} + \hat{U} + \sum_{i\sigma} v_i \hat{n}_{i\sigma}. \quad (1)$$

This comprises the kinetic energy,

$$\hat{T} = -t \sum_{i\sigma} (\hat{c}_{i\sigma}^\dagger \hat{c}_{i+1,\sigma} + \hat{c}_{i+1,\sigma}^\dagger \hat{c}_{i\sigma}), \quad (2)$$

the electron-electron repulsion,

$$\hat{U} = U \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}, \quad (3)$$

and the external potential, defined through the set of on-site energies $\{v_i\}$. Here $\hat{c}_{i\sigma}^\dagger$ ($\hat{c}_{i\sigma}$) is the fermionic creation (annihilation) operator for an electron at site i with spin $\sigma = \uparrow, \downarrow$, the $\hat{n}_{i\sigma} = \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma}$ are the occupation operators, while t and $U > 0$ are the electronic hopping integral and the Coulomb repulsion energy, respectively.

The LDFT version of the Hohenberg-Kohn theorems [7,8] establishes that there exists a universal functional, $F_U[\{n_{i\sigma}\}]$,

of the spin-resolved site occupations, $n_{i\sigma} = \langle \hat{n}_{i\sigma} \rangle$, such that the total energy of the system, $E_U[\{n_{i\sigma}\}]$, can be written as

$$E_U[\{n_{i\sigma}\}] = F_U[\{n_{i\sigma}\}] + \sum_{i\sigma} n_{i\sigma} v_i, \quad (4)$$

where by definition,

$$F_U[\{n_{i\sigma}\}] = \langle \Psi[\{n_{i\sigma}\}] | \hat{T} + \hat{U} | \Psi[\{n_{i\sigma}\}] \rangle, \quad (5)$$

with $|\Psi[\{n_{i\sigma}\}]\rangle$ being the many-body wave function that yields the occupations $\{n_{i\sigma}\}$ and minimizes the expectation value of $\hat{T} + \hat{U}$. Note that in this context the concept of “universality” is somehow more restrictive than in *ab initio* DFT. In fact, as in *ab initio*, here a functional is universal when it does not depend of the external potential, namely, on the on-site energies. However, it does depend on both the kinetic and the Hubbard energy. Thus, one has a different “universal” functional for every different choice of kinetic energy (dimensionality and connectivity of the system and range of the hopping matrix) and of the Hubbard U parameter, with the $U = 0$ limit recovering the noninteracting tight-binding model. Then, in analogy with conventional DFT, one defines the exchange-correlation energy, which here is denoted with “Hxc,” since it includes also the classical Hartree contribution (from now on this will be referred to as the “Hartree-exchange-correlation” energy),

$$E_{\text{Hxc}}[\{n_{i\sigma}\}] = F_U[\{n_{i\sigma}\}] - F_{U=0}[\{n_{i\sigma}\}]. \quad (6)$$

With such definition at hand we can readily write down the associated KS equations as follows:

$$(\hat{T} + v_{\text{eff},i\sigma})\phi_m = \epsilon_m \phi_m, \quad (7)$$

where $v_{\text{eff},i\sigma} = v_{\text{Hxc},i\sigma} + v_i$ is the KS potential, while ϵ_m and ϕ_m are the KS energies and orbitals, respectively. Formally, the Hartree-exchange-correlation potential is obtained as functional derivative of the corresponding energy, $v_{\text{Hxc},i\sigma} = \partial E_{\text{Hxc}} / \partial n_{i\sigma}$, but in practice this is just a standard derivative, since E_{Hxc} is a function of the $2L$ -dimensional vector of site occupations, $\{n_{i\sigma}\}$, where L is the total number of sites.

Throughout this work, we consider the case of one-third filling, $N_e/L = 2/3$ (N_e is the total number of electrons), which is representative of the metallic phase. Thus, we stay away from half filling, corresponding to the Mott insulating phase, as this presents a much longer correlation length and it is usually much harder to converge. Furthermore, we look at finite-size chains with open boundary conditions and a net spin-zero state, namely, at the situation $N_\uparrow = N_\downarrow = N_e/2$. In this case, spin symmetry allows us to drop the spin index and we express all quantities in terms of the total occupation $n_i = n_{i\uparrow} + n_{i\downarrow}$.

B. Generation of the training and test sets

The construction of the ML Hartree-exchange-correlation energy consists in establishing a mapping between the ground-state site occupations, $\{n_{i\sigma}\}$, and E_{Hxc} . As such, for a given set of on-site energies $\{v_i\}$ one needs to compute the exact ground-state energy of the system. The value of E_{Hxc} is then obtained by subtracting the contribution from the external potential [see Eq. (1)] and the kinetic energy of the

noninteracting system [see Eq. (6)]. We use the density-matrix renormalization group (DMRG) [40] algorithm via TenPy [41] to solve the full many-body problem. Throughout this work, we fix the hopping parameter to $t = 1$, which sets the energy scale of the problem, and consider the case $U = 4$. The training set is constructed from data corresponding to lattice of different sizes, $L = 18, 21, 24$, and for each one of them we produce 17 000 data points.

To generate a dense and diverse training dataset, we use the following strategy: A data point here corresponds to a different choice of external potential, generated randomly within an interval $v_i^{(k)} \in [0, \lambda^{(k)}]$. In particular, for each lattice size, $n = 15\,000$ configurations are obtained from $\lambda^{(k)} = \sqrt{9k/(n-1)}$. The first data point $\lambda^{(k=0)} = 0$ gives us the homogeneous case, while the final point gives $\lambda^{(k=14999)} = 3$. The square root dependence on k is selected as it gives rise to a good distribution of densities because it pushes more of the data points further away from the homogeneous case. Then, we obtain a further $n = 2000$ configurations from $\lambda^{(k)} = 3 + 3k/(n-1)$, a choice that guarantees enough diversity in the training set. Finally, we also include in the training dataset potentials obtained by applying constant electric fields of different strength [see Sec. III E] over the homogeneous lattice $v_i = 0$. The DMRG data are here deemed as “exact,” although their accuracy is limited by the DMRG calculation itself. Poorly converged DMRG data are not included in the training set. The threshold of accuracy was set by requiring that the relative change in the energy is within 10^{-5} after the final step.

As far as the test set is concerned, this is also generated by DMRG for a larger variety of lattice sizes, $L = 24, 30, 36, 42, 48, 54, 60$, with $n = 1000$ random external potentials taken from $\lambda^{(k)} = \sqrt{9k/(n-1)}$. In this case, we do not reject any DMRG results, not to create bias in the test set, namely, we want to avoid biasing the test set toward some particular class of disorder configurations. Since one of the target quantities is the KS Hartree-exchange-correlation potential obtained as the functional derivative of the corresponding energy, we also need to generate test data for v_{Hxc} . For this quantity we follow a well-established reverse engineering strategy, sometimes known as the inverse-Kohn-Sham method [42]. This essentially consists of varying the effective potential until the targeted site occupations are obtained, namely, those computed with DMRG. In practice, we minimize the Euclidean distance $\|n[v_{\text{eff}}] - n_{\text{DMRG}}\|$ and the convergence is set by a threshold of 10^{-5} . The same criterion is adopted for each lattice size investigated. In our case, we use the robust Broyden-Fletcher-Goldfarb-Shanno (BFGS) algorithm [43–46], as implemented in SciPy [47], to descend onto the effective potential that reproduces the DMRG density. The workflow of the data generation is presented in Fig. 1. All of our data and some examples of our code are publicly available on GitHub [48].

C. Machine-learning the functional

We have mentioned before, that our functional is constructed semilocal. This actually means that E_{Hxc} is obtained as the sum of site-dependent Hartree-exchange-correlation

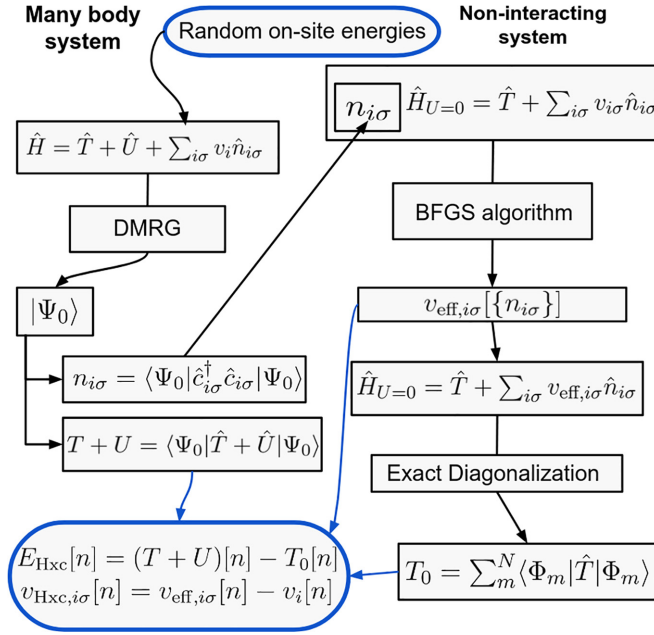


FIG. 1. Workflow of the generation of the “exact” data. On the left-hand side we describe the DMRG pipeline. By starting with some random on-site-energy configuration, we use the DMRG algorithm to find the ground-state wave function, density, and energy. The construction of the exact KS potential is depicted on the right-hand side. The DMRG density is supplied to the BFGS algorithm, along with the noninteracting Hamiltonian to find the effective potential that reproduces the ground-state density. Finally, $E_{\text{Hxc}}[n]$ and $v_{\text{Hxc},i\sigma}[n]$ are computed by subtraction (blue box).

energy densities e_{Hxc} , which in turn, depend on the occupation around that site within a given cutoff a , namely, we have

$$E_{\text{Hxc}}^{\text{ML}}[n] = \sum_i^{L+2a} e_{\text{Hxc}}^{\text{ML}}[\bar{n}_{i,a}], \quad (8)$$

where $\bar{n}_{i,a} = (n_{i-a}, n_{i-a+1}, \dots, n_i, \dots, n_{i+a-1}, n_{i+a})$, and a is positive integer, which we call the nonlocality parameter (cut-off radius). Note that the summation extends beyond indices associated with the actual lattice, since the nonlocality assigns nonvanishing $\bar{n}_{i,a}$ vectors even when the index i is outside the lattice boundaries. Then, the ML KS potential at site i for spin σ , $v_{\text{Hxc},i\sigma}^{\text{ML}}$, is computed by summing up the partial derivative with respect to $n_{i\sigma}$ of all the energy-density terms that contain $n_{i\sigma}$

$$v_{\text{Hxc},i\sigma}^{\text{ML}}[n] = \frac{\partial}{\partial n_{i\sigma}} E_{\text{Hxc}}^{\text{ML}}[n] = \frac{\partial}{\partial n_{i\sigma}} \sum_j e_{\text{Hxc}}^{\text{ML}}[\bar{n}_{j,a}]. \quad (9)$$

The ML Hartree-exchange-correlation energy is constructed as a fully connected neural network comprising five dense layers, each of 64 nodes, while the KS potential is obtained by the automatic differentiation as supplied by TensorFlow [49]. We choose the exponential linear unit as an activation function to ensure that the functional derivatives are continuous. In each case, we train for 5000 epochs with the Adam optimizer and a batch size of 32. One neural network is trained for each value of the nonlocality parameter explored

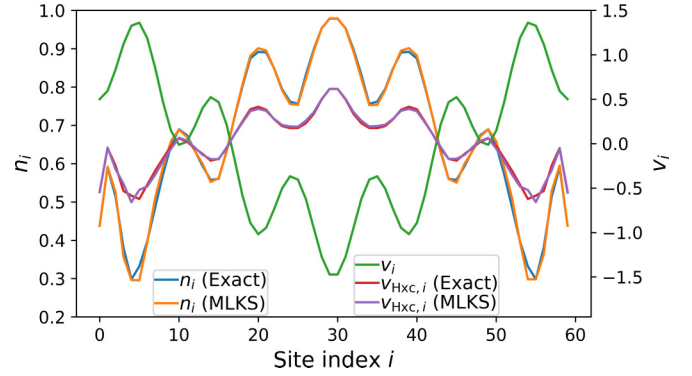


FIG. 2. The ground-state site occupations (left-hand side scale) and the Hartree-exchange-correlation potential (right-hand side scale) obtained from the MLKS scheme are plotted alongside the exact DMRG results (denoted as exact). For this visual comparison we consider a $L = 60$ system and an external potential (green line) of the form $v_i = \cos(2\pi i/L) - \frac{1}{2} \cos(12\pi i/L)$.

$a = 1, 2, 3, 4$. The learning rate is decayed according to

$$\lambda(j) = \lambda_0 \left(\frac{1}{30} \right)^{j/5000} \quad (10)$$

where $\lambda_0 = 3 \times 10^{-4}$ and j is the epoch iteration. It is necessary to use zero-padding while computing the energy density near the boundary, as the nonlocal terms extend beyond it. In the context of LDFT, the use of zero padding can be understood as follows: Our finite system of L -sites with open boundary conditions can be considered to lie in the infinite chain with the on-site energies given by

$$v_i = \begin{cases} v_i & \text{if } 0 \leq i < L \\ +\infty & \text{otherwise.} \end{cases} \quad (11)$$

Corresponding to these on-site energies, the densities will be

$$n_{i\sigma} = \begin{cases} n_{i\sigma}[\{v_i\}] & \text{if } 0 \leq i < L \\ 0 & \text{otherwise.} \end{cases} \quad (12)$$

Data from larger systems can be understood to lie on the same infinite chain, the only difference being that some on-site energies at the boundary are brought down from infinity, allowing electrons to access those sites. Moreover, this compliments the fact that the $\hat{T} + \hat{U}$ component of the Hamiltonian is invariant, so that the same density-functional is valid for different numbers of lattice sites.

III. RESULTS

A. Solving the Kohn-Sham equations

Let us start our analysis by giving a simple demonstration of a successful implementation of the trained functional and how to use it. In Fig. 2 we present the site occupations and the converged KS Hartree-exchange-correlation potential for a system of $L = 60$ sites and an external potential of the form $v_i = \cos(2\pi i/L) - \frac{1}{2} \cos(12\pi i/L)$. The functional in this case was trained over $L = 18, 21, 24$ sites and here we compare the results from our ML KS (MLKS) scheme and the exact DMRG reference. The KS self-consistent cycle is performed with a simple linear-mixing scheme [50],

in which the input set of site occupations at the $(k+1)$ st iteration, n^{k+1} , is constructed as $n^{k+1} = (1-\lambda)n^k + \lambda n^k$, where n^k is the set of output occupations at iteration k . Here we set λ to decay from 0.03 to 0.005 over the first 2000 iterations and then to remain constant at 0.005 should more iterations be required, and the convergence criterion is $\|n^k - n^{k+1}\| < 10^{-5}$. As we can observe in Fig. 2, there is a good visual agreement between the MLKS-calculated quantities (occupations and potential) and the DMRG reference. In particular, v_{Hxc} appears to have the correct expected behavior, namely, it becomes more prominent in regions of high electron density, corresponding to minima of the external potential.

It is important to note that when assessing the accuracy of the KS scheme at convergence one has to consider that the error in the KS potential itself adds to that of the ground-state site occupations, which are determined by the potential through the self-consistent cycle. It is then interesting to disentangle such errors by computing a number of additional quantities. In particular, in Fig. 3 we compare both the Hartree-exchange-correlation KS potential and energy density computed at either the exact DMRG density [$e_{\text{Hxc}}^{\text{app}}[n_{\text{exact}}]$ and $v_{\text{Hxc}}^{\text{app}}[n_{\text{exact}}]$, in Figs. 3(a) and 3(d), respectively] or the KS occupations [$e_{\text{Hxc}}^{\text{app}}[n_{\text{KS}}]$ and $v_{\text{Hxc}}^{\text{app}}[n_{\text{KS}}]$, in panels 3(b) and 3(e), respectively], with the corresponding exact (DMRG) quantities. Here, “app” denotes the relevant approximate functional. Namely, our MLKS functional, or the fully numerical implementation of the Bethe-ansatz LDA (BALDA) [8,51–54]. The BALDA is perhaps the most developed non-ML approximation to the exchange-correlation functional for the Hubbard model. It can be used both in a orbital-free DFT mode or within a KS scheme. Here it will represent a point of comparison for our developed functional (note that also for the BALDA we evaluate the Hartree contribution to energy and potential together with the exchange-correlation one). For the results of Fig. 3 we consider a test system of 60 sites and the various parity plots are constructed over 1000 disorder configurations.

In the case of MLKS, it appears that the error accumulated over the KS self-consistent cycle is only minor. In fact, we find a mean absolute (MAE) error over the Hartree-exchange-correlation energy density of 2.2×10^{-4} , when this is computed at the exact DMRG occupations, an MAE that grows to only 3×10^{-4} with the converged KS occupations. Note that $e_{\text{Hxc}}^{\text{ML}}$ averages at around 0.3 for this choice of disorder and occupation, meaning that the relative error on the energy density is of the order of 0.1%. Interestingly, the results for the KS Hartree-exchange-correlation potential returns a situation where the MAE is lower, even if marginally when v_{Hxc} is computed at the KS occupations rather than at the exact ones. This suggests that the functional derivative defining the KS potential may slightly shift the ground-state occupations from their exact values. In any case, an overall assessment of the model constructed can be obtained from Figs. 3(c) and 3(f), where we show the parity plot for the total energy density, F_U/L , and the KS occupations, respectively. These give us relative MAEs of 0.07% (F_U/L) and 1% (n), pointing to an extremely accurate model, whose main error accumulates in the self-consistent occupations.

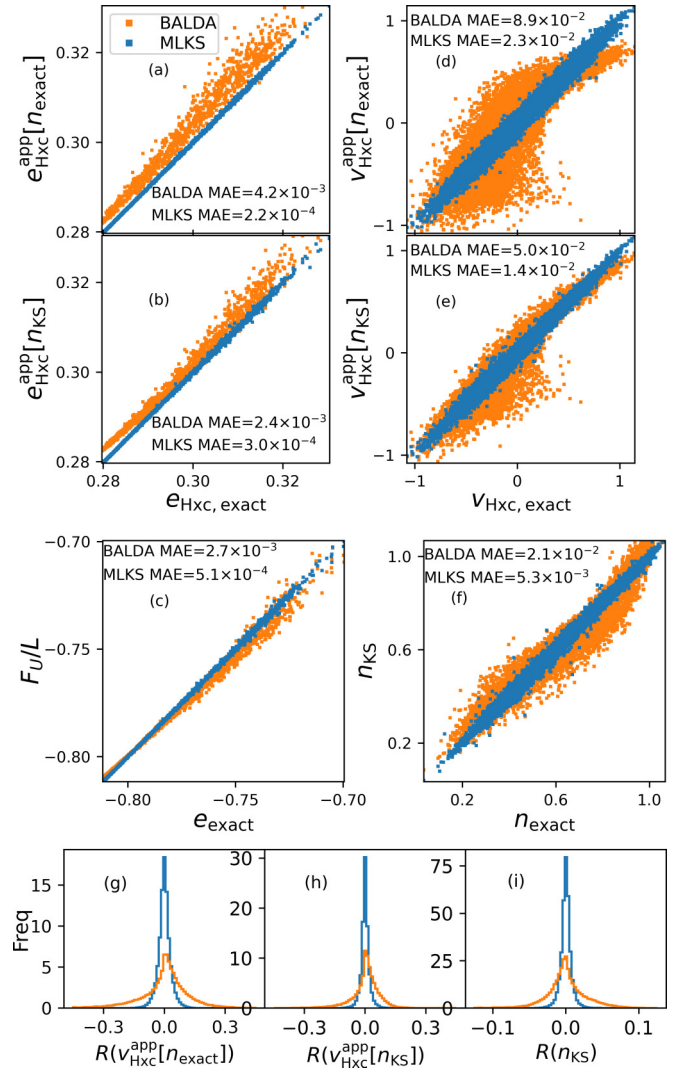


FIG. 3. Analysis of the accuracy of the MLKS and BALDA functionals. For a system of $L = 60$ sites, we present the parity plots of both the ML Hartree-exchange-correlation energy density, $e_{\text{Hxc}}^{\text{app}}$, and the KS Hartree-exchange-correlation potential, $v_{\text{Hxc}}^{\text{app}}$, computed either at the exact (DMRG) occupations [panels (a) and (d), respectively] or at the converged KS occupations, n_{KS} [panels (b) and (e), respectively]. The relevant approximate functional is denoted “app,” and corresponds to that indicated in the legend. In panels (c) and (f) we present the parity plots for the KS total energy density and occupations, respectively. Panels (g) and (h) display histograms of the residuals of $v_{\text{Hxc}}^{\text{app}}$ at the exact and converged KS densities, and panel (i) shows the densities. Here, $R(x)$ denotes the residual quantity. For instance $R(n_{\text{KS}}) = n_{\text{KS}} - n_{\text{exact}}$. Results are presented for the MLKS functional with a nonlocality parameter $a = 4$ and over a test set of 1000 configurations. In the various panels we also report the MAE.

Unlike the MLKS scheme, the BALDA results show some systematic error. Namely, e_{Hxc} appears to be over-approximated, while v_{Hxc} has some curvature relative to the parity line. Visually, the latter error [see Figs. 3(d) through 3(f)] is inflated by the high number of data points on the plot, 60 000, where the majority of the points lie relatively close to the parity line, although they are not visible. Therefore, for a better visualization, we plot a histogram of the residuals

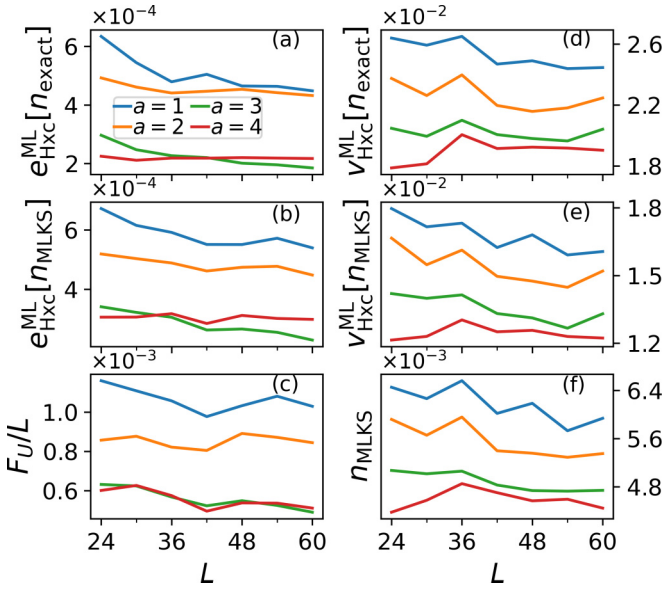


FIG. 4. The MAEs, computed over 1000 disorder configurations for each system, as a function of the system size, L . Data are presented for the ML Hartree-exchange-correlation energy density, $e_{\text{Hxc}}^{\text{ML}}$, and the KS Hartree-exchange-correlation potential, $v_{\text{Hxc}}^{\text{ML}}$, computed either at the exact (DMRG) occupations [panels (a) and (d), respectively] or at the converged KS occupations [panels (b) and (e), respectively]. In panels (c) and (f) we present the MAE for the ML KS total energy density and occupations, respectively. In all cases, we show results for different values of the nonlocality parameter a .

in Figs. 3(g) through 3(i). Similarly to the case of MLKS, but to a much greater extent, the BALDA results improve dramatically, when computed at the converged KS density instead of the exact one. In fact, the errors on e_{Hxc} and v_{Hxc} drop roughly by a factor of 2. Furthermore, in Fig. 3(c) the systematic over-approximation of e_{Hxc} appears to be canceled out by an under-approximation of the noninteracting kinetic energy, giving a e_{KS} in a satisfactory agreement with the exact data.

All considered, the MLKS functional appears to be in much better agreement with the exact functional than the well-known BALDA. The evidence for this is (i) the consistently lower values of MAE for each quantity, (ii) the absence of systematic errors in the parity plots, and (iii) the higher degree of consistency between the results calculated at the exact and KS densities. In Appendix, we repeat the same test, using the fully local $a = 0$ ML functional and discuss how that compares to the BALDA.

B. System size and nonlocality parameter

To have a better insight into the ML functional construction, it is useful to investigate its nonlocal properties. The effects of the functional built-in nonlocality are analyzed by looking at lattice sizes ranging from 24 to 60, and by computing the MAEs for different values of the nonlocality, $a = 1, 2, 3, 4$. The results ensemble-averaged over 1000 disorder configurations are summarized in Fig. 4, where we present the MAEs of $e_{\text{Hxc}}^{\text{ML}}[n_{\text{exact}}]$ and $e_{\text{Hxc}}^{\text{ML}}[n_{\text{MLKS}}]$ in Figs. 4(a) and 4(b), $v_{\text{Hxc}}^{\text{ML}}[n_{\text{exact}}]$ and $v_{\text{Hxc}}^{\text{ML}}[n_{\text{MLKS}}]$ in Figs. 4(d) and 4(e),

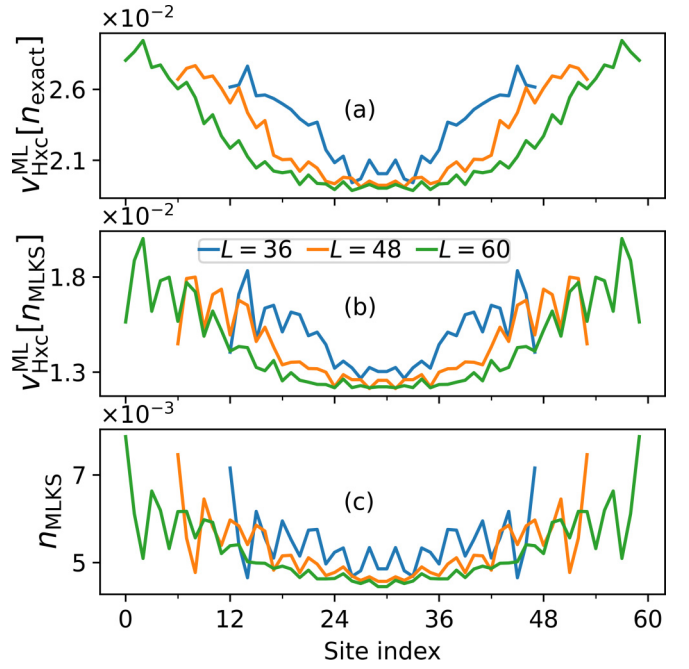


FIG. 5. Site-resolved MAE, computed over 1000 disorder configurations for three lattice sizes, $L = 36, 48, 60$ and nonlocality, $a = 4$. The (a) MAE for $v_{\text{Hxc}}^{\text{ML}}$ at exact site occupations, n_{exact} , and (b) at the MLKS ones, n_{MLKS} . In panel (c) we show the MAE of the MLKS site occupations (see text for details).

and again the MAEs of F_U/L and the KS occupations in Figs. 4(c) and 4(f). A number of trends are clearly visible.

First, one may notice that the functionals get progressively more accurate with increasing their nonlocality a , in particular when going from $a = 1$ to $a = 3$, after which the performance improvement seems to saturate. This is particularly true for the energy density $e_{\text{Hxc}}^{\text{ML}}$, while further improvements at $a = 4$ are found for the potential $v_{\text{Hxc}}^{\text{ML}}$. Such numerical result indicates that the nonlocal correlation of the Hubbard model at the chosen filling factor is well captured at $a \simeq 3$. It is also interesting to note that the smaller MAE for $v_{\text{Hxc}}^{\text{ML}}$ is computed at the MLKS occupations and not at the exact ones, $v_{\text{Hxc}}^{\text{ML}}[n_{\text{exact}}]$. This behavior was observed before for $a = 4$ and it is found here for all systems regardless of the nonlocality. The second noteworthy trend is that, in general, the MAE appears to decrease with increasing the lattice size. To investigate whether this is a boundary effect, we look into the site dependence of the MAEs as a function of the lattice size. In Fig. 5 we show the site-resolved MAE of the KS Hartree-exchange-correlation potential, computed at both the MLKS and exact occupations, and the final occupations, as a function of the lattice position. This is shown for three different lattices, $L = 36, 48$, and 60 , and the MAEs are averaged over 1000 different disorder configurations. We find that, on average, the MAE at the edges of the lattice is about 25% larger than in the middle, suggesting that the systematic trend in the MAE decrease with system size may be due to a decreasing edge-to-bulk ratio. In contrast, in the middle of the lattice, all quantities converge to a similar MAE, regardless of the lattice size. The loss of accuracy at the lattice edges may be attributed to an imbalance in the training data. In fact, the edges are characterized

by $\bar{n}_{i,a} = (n_{i-a}, n_{i-a+1}, \dots, n_i, \dots, n_{i+a-1}, n_{i+a})$ vectors presenting $n_j = 0$ for those sites lying outside the lattice. Vectors of this type, however, are less abundant in our training set. In fact, for a lattice of L sites and a nonlocal parameter a , there are $4a$ sites with $\bar{n}_{i,a}$ presenting at least one vanishing occupation and $L - 2a$ sites with all nonvanishing occupations.

In any case, our results return us a rather accurate Hartree-exchange-correlation functional, which can be used to derive a KS potential via functional derivative. Already with a modest degree of nonlocality, this allows us to compute energies and occupations close to those obtained with DMRG, with the accuracy improving as the system size grows.

C. Analysis of the functional at different filling factors

The universal DFT functional is defined for any total number of particles in the system. However, there is no guarantee that this property is transferred to approximated machine-learning formulations. Namely, although one can always extend a semilocal functional to filling factors outside the training range, it remains the question of whether the functional is actually accurate. This question naturally made us curious to test how well the trained functionals perform to filling factors away from $2/3$. By using both the DMRG and the MLKS scheme, we now map the density, energy and Hartree-exchange-correlation potential of 128 random configurations of a $L = 36$ lattice and different electron numbers, $N_e = 16, 18, \dots, 32$ (the training was performed for $N_e = 24$, corresponding to a filling factor of $2/3$). In this test, we use the same $\lambda^{(k)} = \sqrt{9k/(n-1)}$ distribution described in Sec. II B to generate the disorder configurations, and examine how the average error depends on the number of electrons.

The results are given in Fig. 6. Unsurprisingly, the error minimizes at $N_e/L = 2/3$, namely, at the same filling factor of the training set, where all energies densities shown in Figs. 6(a) through 6(c) take on minima of the order of 10^{-4} and have similar scaling away from this point. For instance, the converged Kohn-Sham energy density [Fig. 6(c), $a = 4$] has an MAE of 2×10^{-3} , 5×10^{-4} , 2×10^{-3} at $N_e/L = 1/2, 2/3$ and $5/6$, respectively. In the case of the Hartree-exchange-correlation potential and similarly for the occupation, the MAE varies more slowly against the number of electrons. In Fig. 6(f), the $a = 4$ curve has an MAE of 1×10^{-2} , 4×10^{-3} , 7×10^{-3} at $N_e/L = 1/2, 2/3, 5/6$, respectively. Finally, it is important to note that the accuracy of a given functional, over both energy density and potential, away from the training filling factor of $2/3$ does not seem to have a strong dependence on the degree of nonlocality. In fact, for some quantities and filling factors local functionals appear more accurate than nonlocal ones. This is, for instance, the case of the various energy densities at low filling factors ($< 2/3$), for which the $a = 1$ functional performs significantly better than any more nonlocal ones.

The success of the model's performance for different filling factors is likely due to the inhomogeneous nature of the training data, combined with the semilocal architecture of the functional. This gives the model exposure to density regions with occupations both higher and lower than $2/3$ filling. To

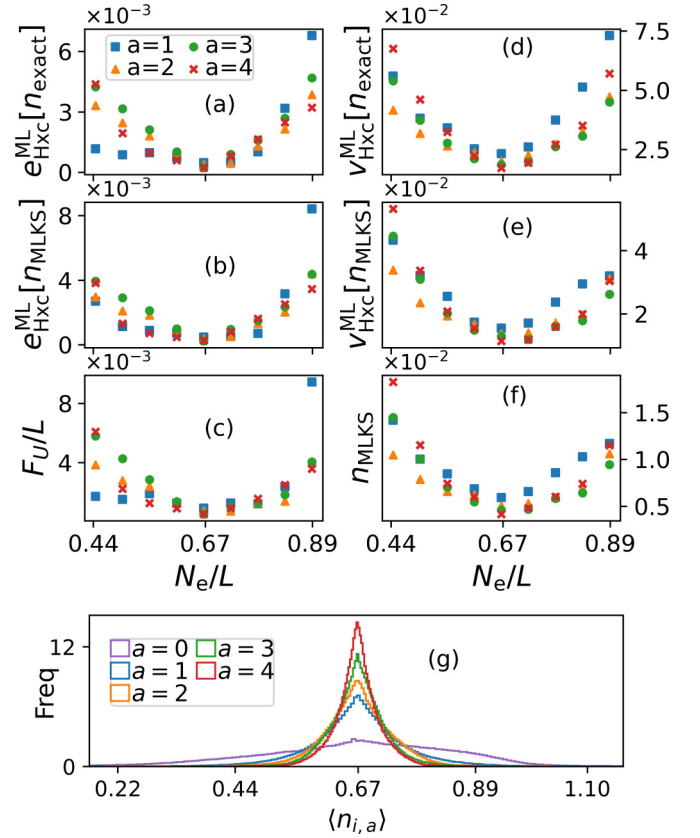


FIG. 6. MAE obtained over 128 disordered configurations for the Hartree-exchange-correlation energy density and the KS potential of a system of $L = 36$ sites and different number of electrons, N_e . The model has been trained at $N_e/L = 2/3$, where the highest accuracy is reached. The various quantities have been defined in the main text. In panel (g) we show the normalized distributions of the average local occupations, $\langle n_{i,a} \rangle$, computed over the entire training set.

understand better this point, in panel (g) of Fig. 6 we plot the distribution of the average local occupation. This is defined as $\langle n_{i,a} \rangle = \sum_{j=i-a}^{i+a} n_j / (2a + 1)$, namely, it is the average filling factor of the portion of the local occupations that determine the Kohn-Sham potential, a quantity that depends on the degree of nonlocality, a . As expected, all distributions are centered around the training filling factor of $2/3$, but their width gets progressively narrower as the functional becomes more nonlocal. This means that during the training the more nonlocal functionals experience a narrower distribution of local filling factors. As a result, although nonlocal functionals are more accurate at the global filling factor of $2/3$, their performance degrades faster away from it.

It can be thus expected that the functional accuracy will improve as the system size is increased, provided that only a few electrons are added, since $(N_e \pm \delta N_e)/L \approx N_e/L$ for large L (namely for variations close to the training range, around the maximum of the $\langle n_{i,a} \rangle$ distributions). The ability of the model to work away from the training filling factor is a useful property because it allows one to perform tests, which require N_e to be varied. For instance, one could compute the band gap, or the inverse compressibility [55,56]. In particular, in the next Sec. III D, we test whether the KS energy is piecewise linear.

D. Piecewise linearity of the Kohn-Sham energy

Conventional DFT can be extended to fractional numbers of electrons by using a zero-temperature ensemble of integer electron states. Within this framework, it can be shown that the exact DFT total energy computed at integer electron-number values connects linearly [57], namely, the $E - N_e$ curve is piecewise linear. This property formally writes

$$E(N) = (1 - x)E(N_0) + xE(N_0 + 1), \quad (13)$$

where $N = N_0 + x$, N_0 is an integer and x is taken in the interval $[0, 1]$. The piecewise linearity, however, is not satisfied by semilocal density functionals [58–65]. For instance, in the local density approximation such curve is approximately parabolic around the neutrality point. The shortfall is widely attributed to the “delocalization error” or the related “self-interaction error,” which gives rise to a set of hallmark DFT deficiencies: an inaccurate description of charge transfer and charge distribution in separated systems, a systematic underestimation of the band gaps, and generally over-delocalized orbitals [66].

If Eq. (13) holds, then the highest occupied Kohn-Sham orbital at $N = N_0$, $\epsilon_H^{N_0}$, is related to the slope of $E(N)$ as

$$\epsilon_H^{N_0} = \frac{\partial E}{\partial N} = E(N_0) - E(N_0 - 1), \quad (14)$$

so that $-\epsilon_H^{N_0}$ equals the ionization potential, IP . The same argument can then be applied to the case of $N_0 + 1$ electrons, where one finds the electron affinity, EA , of the N_0 electron system. Thus, one can conclude that the fundamental gap of a molecule containing N_0 electrons is simply

$$\Delta^{N_0} = IP - EA = \lim_{x \rightarrow 0} \left\{ \left. \frac{\partial E}{\partial N} \right|_{N_0+x} - \left. \frac{\partial E}{\partial N} \right|_{N_0-x} \right\}. \quad (15)$$

In general, Δ^{N_0} is not the difference between the lowest-unoccupied and the highest-occupied Kohn-Sham orbital (the Kohn-Sham gap), $\Delta_{KS}^{N_0} = \epsilon_L^{N_0} - \epsilon_H^{N_0}$, a fact attributed to the lack of derivative discontinuity of the exchange-correlation energy.

We now test both the MLKS functional (we focus here only on the $a = 4$ case) and the BALDA against this behavior for a homogeneous $L = 36$ lattice, which can be viewed as a large molecule. In the upper panel of Fig. 7 we present the total energy as a function of the number of electrons, while the lower panel shows its derivative. The exact limit is established by the DMRG results. In this case we have access only to integer values of the electron number and the energy, by construction, is piecewise linear at any integer.

In contrast, the total energy of both the MLKS functional and the BALDA changes slope only at even electron numbers, while in between those the $E(N_e)$ curve is convex. This is because the MLKS functional has been trained only for the $N_\uparrow = N_\downarrow$ case and we do not have access to broken-symmetry solutions. As a consequence, the Kohn-Sham energy levels remain always spin degenerate, so that while varying N_e , the highest occupied Kohn-Sham orbital is continuously filled until it accommodates two electrons under the constrain $N_\uparrow = N_\downarrow$. This gives rise to a linear dependence of the energy derivative in between two even integers (see lower panel of Fig. 7).

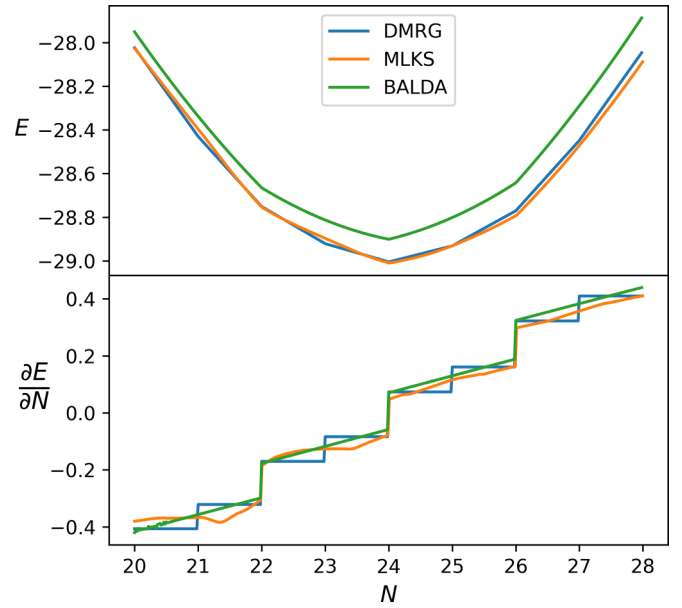


FIG. 7. Total energy against the number of electrons (top panel) and its derivative (bottom panel) for different functionals. Results are presented for a homogeneous $L = 36$ lattice. The blue curves (DMRG) are computed by DMRG at integer electron numbers and interpolated linearly using Eq. (13). The BALDA and MLKS curves are computed by solving the Kohn-Sham equations at fractional occupation. In the case of MLKS we have used the functional with $a = 4$.

Such dependence is only partially satisfied for $N_0 < 24$, probably due to some numerical error.

Note that, certainly among the training data there are several configurations where the local occupations (those defining the Kohn-Sham potential) add to an odd number of electrons. However, in order to recover a discontinuity in the energy derivative at odd integers, one should include in the training set broken-symmetry solutions for an odd total number of particles. These are the only configurations, for which the frontier Kohn-Sham orbitals result spin polarized. A functional with two spin channels could then be constructed and a spin-dependent potential v_i^σ can be computed from it by functional derivative. This will allow for the solution of a set of coupled Kohn-Sham equations, resulting in the occupation of a new Kohn-Sham orbital at odd integer number of electrons. Note also that in standard DFT the exact functional should satisfy the flat-plane condition with respect to the magnetization [67,68], so that one, in principle, does not need a broken-symmetry solution to satisfy the derivative discontinuity at odd electron count. This conditions, however, is not enforced in our functional and not incorporated in the training set. The linearity of $\partial E/\partial N$ between even integers implies that in that interval the Kohn-Sham energy is parabolic with the electron filling. This means that the Hartree contribution to the total energy, $E_H = \frac{U}{4} \sum_i n_i^2$, is not compensated by the curvature of the exchange-correlation energy (here we refer to the conventional definition of exchange-correlation energy, since the Hartree-exchange-correlation energy of the MLKS functional contains the Hartree term). The persistence of this error through the BALDA and the MLKS functional is thus

TABLE I. System gap computed at even-integer electron numbers N_0 . In the table we report the DMRG gap, $\Delta_{\text{DMRG}}^{N_0}$, the LDFT gap computed from Eq. (15), $\Delta_{\text{app}}^{N_0}$, and the Kohn-Sham gap, $\Delta_{\text{KS}}^{N_0}$. In addition we report the corresponding percentage errors, $\delta_{\text{app}}^{N_0}$ and $\delta_{\text{KS}}^{N_0}$, respectively. Data correspond to those presented in Fig. 7 for a homogeneous $L = 36$ lattice.

N_0	DMRG	MLKS				BALDA			
	$\Delta_{\text{DMRG}}^{N_0}$	$\Delta_{\text{app}}^{N_0}$	$\delta_{\text{app}}^{N_0} (\%)$	$\Delta_{\text{KS}}^{N_0}$	$\delta_{\text{KS}}^{N_0} (\%)$	$\Delta_{\text{app}}^{N_0}$	$\delta_{\text{app}}^{N_0} (\%)$	$\Delta_{\text{KS}}^{N_0}$	$\delta_{\text{KS}}^{N_0} (\%)$
22	0.151	0.121	20.3	0.127	16.1	0.122	19.1	0.127	15.8
24	0.157	0.127	19.4	0.135	13.7	0.130	17.4	0.138	12.0
26	0.162	0.135	16.3	0.144	10.8	0.136	15.5	0.142	12.0
28	0.164	0.141	13.9	0.148	10.1	0.142	13.7	0.149	9.1

inherent to (semi-) local functionals, as is the case in *ab initio* DFT.

To conclude this section we verify the accuracy of the constructed functional at describing the system gap, as defined in Eq. (15). In this case we have access to the exact DMRG result, $\Delta_{\text{DMRG}}^{N_0}$, and to the LDFT values, $\Delta_{\text{app}}^{N_0}$. The comparison is carried out only at even integers where the LDFT gaps are defined, with the results presented in Table I. In particular, together with $\Delta_{\text{app}}^{N_0}$ we also report the computed Kohn-Sham gap, $\Delta_{\text{KS}}^{N_0}$, and the corresponding percentage errors, $\delta_{\alpha}^{N_0} (\Delta_{\text{DMRG}}^{N_0} - \Delta_{\alpha}^{N_0}) / \Delta_{\text{DMRG}}^{N_0}$, with $\alpha = \text{app}$ and KS.

From the table it emerges clear that Δ^{N_0} computed at the LDFT level remains systematically smaller than its corresponding exact DMRG result. In particular, we find little difference between the BALDA and our MLKS functional, both returning an underestimation of the gaps in between 13% and 20%. Importantly, the Kohn-Sham gap, $\Delta_{\text{KS}}^{N_0}$, is quite close to $\Delta_{\text{app}}^{N_0}$, so that the exchange and correlation energy has no derivative discontinuity, both in the case of MLKS and the BALDA. Note that this has not to be confused with the discontinuity that the BALDA presents at half filling [15], where the functional fits the Mott transition of the uniform infinite 1D Hubbard model. In that case the local KS exchange-correlation potential jumps discontinuously when the local occupation passes $n_i = 1$, but this does not correspond to a global derivative discontinuity, unless one looks at a single-site problem.

E. Response to an electric field

To demonstrate the use of our ML KS scheme we explore the response of the one-dimensional Hubbard model to an external, constant electric field $\mathcal{E}$. The response is quantified by the polarizability, α , defined as the derivative of the induced electrical dipole moment, $P(\mathcal{E})$, with respect to the electric field, in the limit $\mathcal{E} \rightarrow 0$. The presence of the electric field adds a linear on-site potential $v_{\mathcal{E}i} = e\mathcal{E}(i - \bar{x})$, which introduces a new term to the Hamiltonian,

$$\hat{H}_{\mathcal{E}} = \sum_{\sigma} \sum_{i=1}^L v_{\mathcal{E}i} \hat{c}_{i\sigma}^{\dagger} \hat{c}_{i\sigma} = e\mathcal{E} \sum_{\sigma} \sum_{i=1}^L (i - \bar{x}) \hat{c}_{i\sigma}^{\dagger} \hat{c}_{i\sigma}, \quad (16)$$

where $\bar{x}$ is the central position in the lattice. The dipole moment can then be computed as the expectation value of the

dipole operator,

$$P(\mathcal{E}) = e \langle \Psi_0(\mathcal{E}) | \sum_{\sigma} \sum_{i=1}^L (i - \bar{x}) \hat{c}_{i\sigma}^{\dagger} \hat{c}_{i\sigma} | \Psi_0(\mathcal{E}) \rangle, \quad (17)$$

where $|\Psi_0(\mathcal{E})\rangle$ is the many-body ground-state wave-function for the given electric field $\mathcal{E}$. The same procedure can then be followed by using our ML KS scheme, by solving the self-consistent KS equations in the presence of the electric field term of Eq. (16), and then by computing

$$P(\mathcal{E})^{\text{KS}} = e \sum_{\sigma} \sum_{i=1}^L (i - \bar{x}) n_{i\sigma}^{\text{MLKS}}. \quad (18)$$

The polarizability is then obtained as the numerical derivative of $P(\mathcal{E})$ at $\mathcal{E} = 0$,

$$\alpha = \left. \frac{dP(\mathcal{E})}{d\mathcal{E}} \right|_{\mathcal{E}=0}. \quad (19)$$

1. Polarizability scaling

Our computed polarizability α as a function of the system length L is presented in Fig. 8 for homogeneous lattices of size $L = 18, 21, 24, \dots, 72$. In the figure we present the exact DMRG results together with the errors of our ML KS scheme

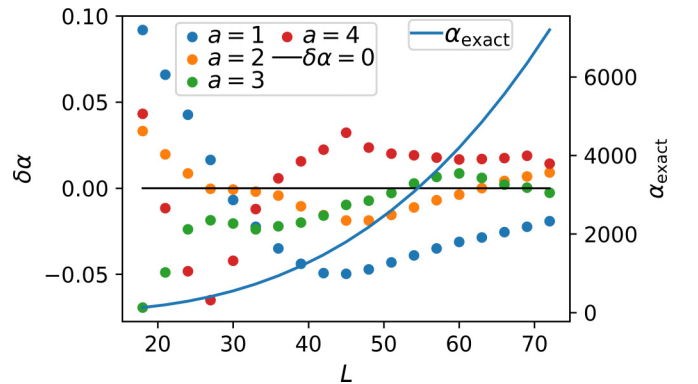


FIG. 8. Polarizability α of the one-dimensional (1D) Hubbard model at one-third-filling, plotted against the number of sites, L , of a linear open-ended chain. Here α_{exact} is computed with DMRG (blue line, right-hand-side scale), while the symbols show the fractional error $\delta\alpha = (\alpha_{\text{MLKS}} - \alpha_{\text{exact}}) / \alpha_{\text{exact}}$ of the MLKS-calculated polarizability scheme relative to the exact results (left-hand-side y axis). The calculation is repeated for functionals having different nonlocality parameter a .

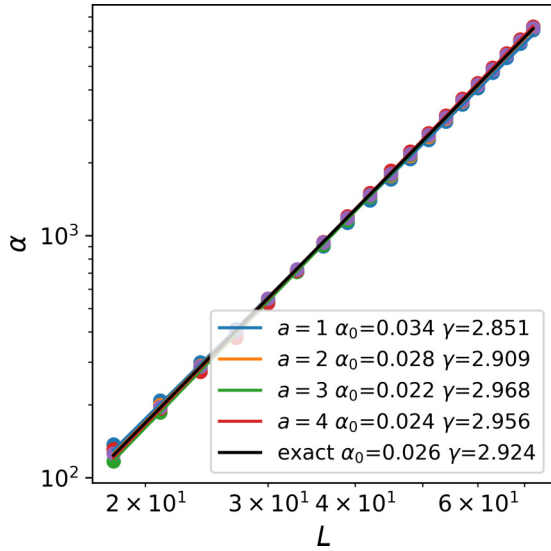


FIG. 9. Polarizability α versus lattice size L plotted on a log-log scale. The legend distinguishes between the various MLKS functionals and the exact results. Also given in the legend are the corresponding values found from fitting the function $\alpha(U; L) = \alpha_0 L^\gamma$.

for models constructed with different nonlocality parameters. Clearly, there is an excellent agreement between the DMRG and MLKS results across the entire range of lengths L . The agreement is particularly good for long chains and nonlocal functionals and remains always below 5%, except for the small lattices and the $a = 1$ case. This reflects the already noted improvement of the Hartree-exchange-correlation functional for large systems arising from the reduced relevance of the edge occupations. Furthermore, it is clear that the nonlocality improves the quality of the ML KS results, although there is little quantitative difference between different degrees of nonlocality.

The polarizability as a function of system size follows a power-law [17] behavior

$$\alpha(U; L) = \alpha_0 L^\gamma.$$

A numerical fit to the above scaling law [see Fig. 9] yields us exponents $\gamma = 2.851, 2.909, 2.968, 2.956$ for nonlocality $a = 1, 2, 3$, and 4, respectively. This is very close to $\gamma = 3$ expected for free electrons in 1D [69] and in line with results obtained with Bethe-ansatz LDFT results [17].

2. Response of the exchange-correlation potential

In general, in conventional DFT the local and semilocal functionals fail in predicting accurate linear polarizabilities, a feature associated with the incorrect response of the exchange-correlation potential to the electric field. This failure ultimately boils down to the self-interaction error [70,71]. Thus, it is important to investigate the response of our ML KS potential to the external field. The exercise can be simply done by plotting the potential response, namely, the difference between the potential computed in the presence of a field and without it, $\Delta v = v^\mathcal{E} - v^{\mathcal{E}=0}$, as a function of the site index. At this point, it is useful to separate the classical and nonclassical contributions to the

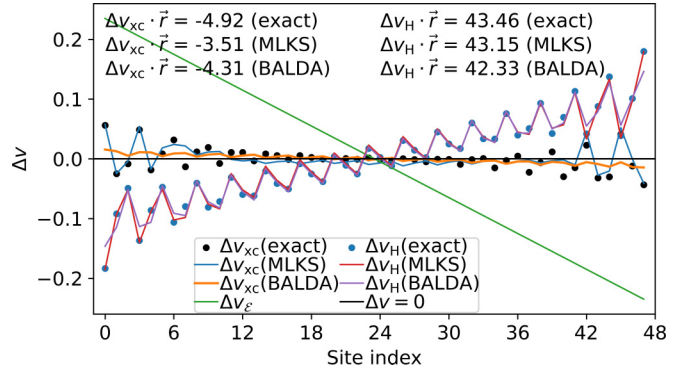


FIG. 10. Response of the Hartree and exchange-correlation potential to the external electrostatic potential, plotted against the site index for a homogeneous $L = 48$ lattice. For the MLKS calculations we use the $a = 3$ functional. A measure of the response can be obtained from $\Delta v \cdot \vec{r}$ (see text), whose values are reported in the legends.

Hartree-exchange-correlation potential, namely, to subtract from the v_{Hxc} previously defined the Hartree component, which is here written as $(v_{\text{H}})_i = U n_i / 2$. This defines the exchange-correlation potential $v_{\text{xc}} = v_{\text{Hxc}} - v_{\text{H}}$.

In Fig. 10 we then plot Δv_{H} , Δv_{xc} , and Δv_{E} , where v_{E} is the potential associated with the electric field [see Eq. (16)]. Results are presented for a system of $L = 48$ sites and for the ML functional with $a = 3$, which has returned the most accurate polarizability. For completeness we also include results obtained within the BALDA. As usual, we compare the DMRG results (dots) against those obtained with MLKS, showing once again an excellent agreement. As expected the Hartree potential responds against the external one. In contrast, Δv_{xc} has the same slope of Δv_{E} . This behavior is opposite to what found for *ab initio* DFT, but it is consistent with previous Bethe-ansatz LDFT results [17], as confirmed numerically here as well. To quantify the potential response, we compute for each of the potentials the product, $\sum_i \Delta v_i \cdot (i - \bar{x})$, which we refer to as $\Delta v \cdot \vec{r}$ and describes the slope of the response. With this definition at hand, we conclude that the Hartree response is about tenfold larger than the exchange-correlation ones. It is also worth noting that the results obtained with our ML functional are close to those obtain with the BALDA, indicating that at least for this quantity the nonlocality does not seem to play a key role.

3. Polarizability for disordered systems as they approach the thermodynamic limit

One of the major advantages of an MLKS scheme is that it can be applied to any system size and distribution of on-site energies (within the range of the training data). As such, it is ideal to investigate the effect of disorder as the system approaches the thermodynamic limit, an exercise performed here for the polarizability. Our analysis starts with systems comprising 21 and 42 sites, which are accessible by DMRG. For both system sizes, we generate $n = 2000$ random disorder configurations from $v_i^{(k)} \in [-\lambda^{(k)}, \lambda^{(k)}]$ where $\lambda^{(k)} = 3k/2(n-1)$. Then, we compute and plot the polarizability against a one-dimensional measure of the

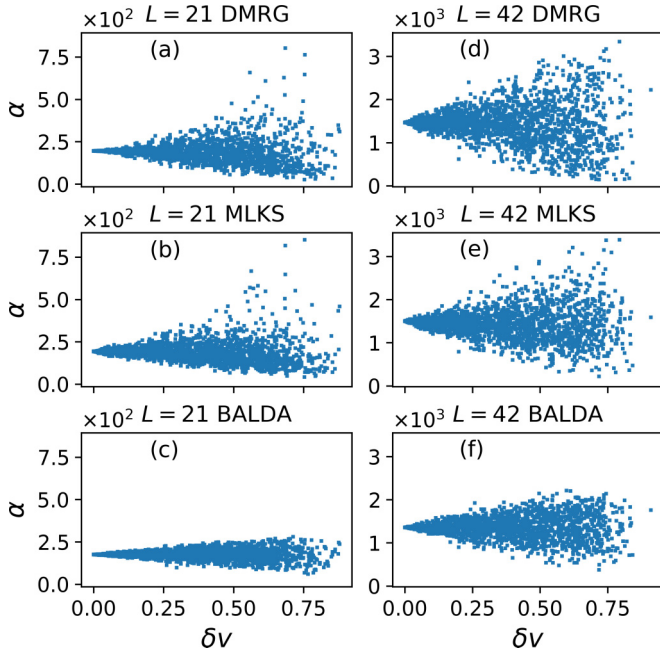


FIG. 11. The polarizability α against disorder strength $\delta v = \frac{1}{L} \sum_i^L |v_i|$ computed with DMRG (top row), MLKS (middle row), and BALDA (bottom row), for systems of $L = 21$ lattice sites (left column) and $L = 42$ (right column). In each subplot we show data for 2000 disorder configurations. In the case of MLKS, we use the model trained with nonlocality $a = 4$.

disorder, namely, the absolute variance $\delta v = \frac{1}{L} \sum_i^L |v_i|$ (note that each disorder configuration has zero mean).

The results are shown in the Fig. 11, where we present the polarizability for all 2000 configurations, computed with DMRG and with the KS schemes, either using our ML functional ($a = 4$) or the BALDA. Clearly, our MLKS-computed polarizabilities are in close agreement with the exact DMRG results across the entire range of disorder investigated. In contrast, the BALDA provides good polarizabilities for weak disorder but tends to drastically underestimate the spread of the distribution for high disorder. This is the situation where the site occupations may distribute over a larger range. It is also clear that the distributions of α as a function of δv are different for systems of different size, namely, they are less diffused for long chains. We return to this point when analyzing the limit of large chains.

For our large-scale investigation, we examine systems containing respectively $L = 21, 42, 84, 168, 336, 672$, and 1344 sites. In this case we consider only four different levels of disorder, namely, $\delta v = 0.05, 0.1, 0.2, 0.4$. For each L and δv we generate 200 random disorder configurations and rescale the on-site energies so to return the required δv value. The polarizability is then computed according to Eq. (19) with a finite difference of $\mathcal{E} = 1/2L$. For this scaling exercise we monitor the relative change in the average polarizability with respect to that of the homogeneous system, α_0 , namely,

$$R_{\text{avg}}(\delta v, L) = \frac{\alpha_{\text{avg}}(\delta v, L) - \alpha_0}{\alpha_0}, \quad (20)$$

while the distribution of the data is analyzed by computing the 2.5th and 97.5th percentiles.

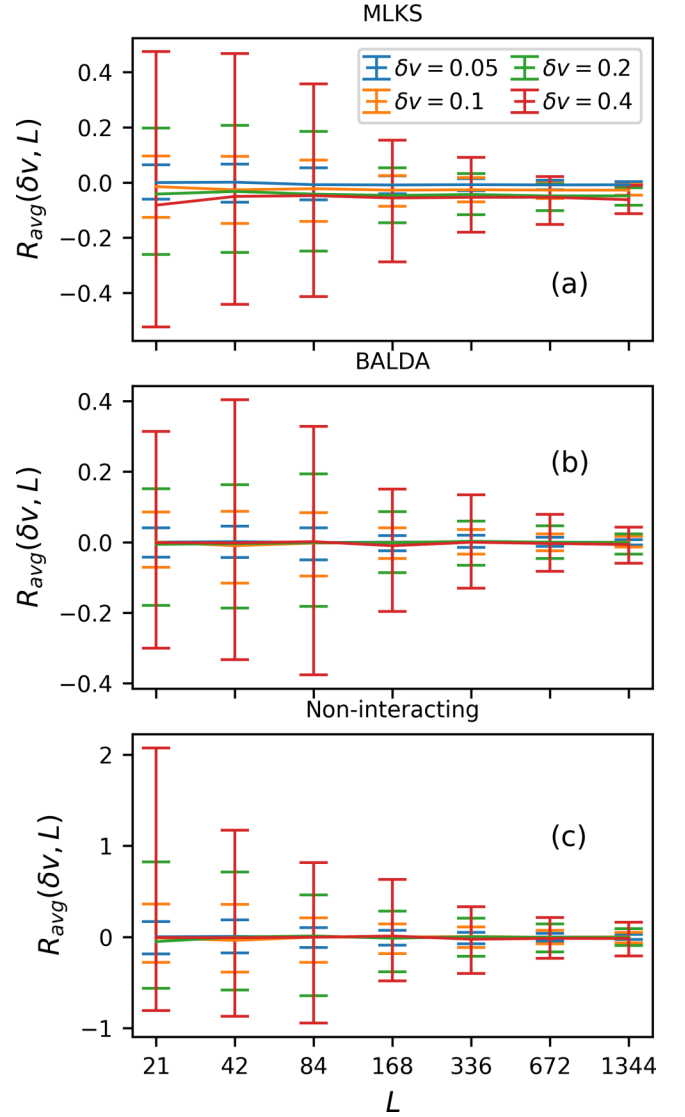


FIG. 12. Relative change in the averaged polarizability, $R_{\text{avg}}(\delta v, L)$ as a function of the lattice size L , for different levels of disorder δv . The lower (upper) error bars give the 2.5th (97.5th) percentile of the distribution. Panels (a), (b), and (c) show the results from MLKS, BALDA, and the noninteracting tight-binding model, respectively. There are 200 data points for each $(L, \delta v)$ pair. For the MLKS scheme, we use the model trained with nonlocality $a = 4$.

For these system sizes we no longer have access to DMRG results, so that we limit the comparison of our MLKS results to those obtained with BALDA and to the noninteracting case. In Fig. 12 we present $R_{\text{avg}}(\delta v, L)$ against length for different levels of disorder and the different models used. A few key points must be noticed. First, in all cases the average polarizability remains close to that computed for the homogeneous case at the same level of theory. In the noninteracting case, at low disorder, the individual polarizabilities computed for different configurations distribute symmetrically about the average, a symmetry that breaks down for large δv . Overall then, the spread of $R_{\text{avg}}(\delta v, L)$ values reduces monotonically with the system size, suggesting that in the thermodynamic limit (infinite L) one should expect the polarizability to be that of the homogeneous case.

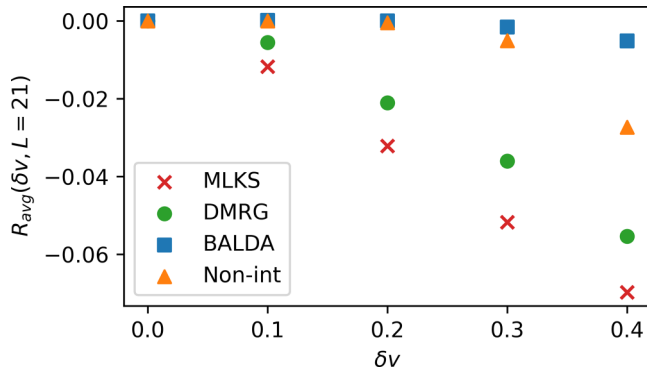


FIG. 13. R_{avg} against disorder strength $\delta v = 0.1, 0.2, 0.3, 0.4$ for a chain containing 21 sites, computed at different levels of theory. For each δv we average over $n = 16$ disorder configurations for MLKS and zero for DMRG, and over $n = 51$ for BALDA and $n = 200$ for the noninteracting case. This returns approximately the same variance.

When Coulomb repulsion is switched on (finite U) both our MLKS and the BALDA scheme return us a polarizability spread significantly smaller than that of the noninteracting case. This is not surprising since the Coulomb repulsion is more pronounced in regions of high density, so that for the same on-site energy random arrangement one expects the interacting problem to yield a more homogeneous charge density. In turn, this reduces the deviation of the computed polarizability from their average. This effect can be quantified by looking at the difference between the 97.5th and the 2.5th percentile of the distribution for the largest chain, $L = 1,344$, and strongest disorder, $\delta v = 0.4$. This is 0.1 for both MLKS and BALDA and 0.37 for the noninteracting case.

Finally, we wish to take a closer look at the dependence of the average polarizability as a function of the disorder strength. As noted before, this eventually approaches the homogeneous-chain limit, but for finite chains a small dependence is noticed. As this is difficult to extract from the plots in Fig. 12 due to the scale, in Fig. 13 we present the deviation of the average α from the homogeneous case as a function of δv for a 21-site-long chain and the different models. In constructing the graph we have averaged over a different number of samples, so that the variance of the different methods is approximately similar (the average is equally statistically significant). Clearly the deviation from the homogeneous limit, as expected, increases with the disorder strength. Such an increase is underestimated by both BALDA and by the noninteracting tight-binding model, but it is well captured by our MLKS scheme, which remains close to the DMRG results at all δv .

IV. CONCLUSION AND OUTLOOK

We have presented a method for the construction of an ML energy functional for the one-dimensional Hubbard model, which can define a Kohn-Sham potential by functional derivative. This enables us to use Kohn-Sham DFT for a lattice model, with an accuracy comparable to that of DMRG, which is used to generate the training set. In particular, we construct a class of functionals with different degree of nonlocality.

These can be trained on systems with moderate size and deployed for much larger ones. In general, we find that the accuracy of the functional improves with the degree of nonlocality and that most of the error concentrates on lattice sites at the edge of the linear molecules. These present charge distribution unevenly represented in the training set, so that the error has to be considered numerical. All constructed functionals perform significantly better than the Bethe-Ansatz local density approximation (BALDA), often used for applying DFT to the Hubbard model.

Since the computational costs of our MLKS functionals are similar to that of the BALDA and scale with the number of sites, the method can be used to investigate large systems and/or situations where ensemble averages are important. For this reason, we looked at the electrical polarizability of both homogeneous and disordered linear chain as a function of the chain length and eventually the disorder strength. We found that MLKS can return polarizability values in close agreement to DMRG results and, in the case of disorder, a very similar spread in their distributions. This is not the case of the BALDA, which tends to underestimate the spreads. In any case, we find that the polarizability of disordered systems converges to that of the homogeneous case as the system size is increased and that the convergence is faster for the interacting case.

The method introduced here, despite allowing one to use the same functional for systems of different size, still presents a number of limitations. First, the functionals are bound by the Hohenberg-Kohn theorems, so that they are defined for a specific value of U/t and for a specific form of the kinetic energy (dimensionality and range of the interaction). For this reason different Coulomb strengths and different type of kinetic energy will require a new parametrization. In principle, one could train a single model to work with a range of model parameters $\{U_i, t_{ij}\}$, which effectively would act as a “collection” of functionals. Whether the model will be able to interpolate across different parameters remains to be seen, since there is no theorem to determine that such interpolation is possible. Certainly, the good success of the BALDA [8,51–54] suggests that such avenue is likely to be pursuable, at least within some range of U/t values. Alternatively, one can move to density-matrix functional theory, where the fundamental quantity becomes the single-particle density matrix and not the site occupations. In this case, the quantity to determine scales as the square of the atomic sites (or the square of number of atomic orbitals), but one has access to a more general definition of kinetic energy [72]. This, in principle, can allow us to explore different regimes of correlation and perhaps move across phase transitions.

A second limitation is that here the models have been trained specifically for the 2/3 filling case. Although we have shown that the functionals are transferable to different, although close, filling factors, it is reasonable to expect that our semilocal approximation will break down as the model correlation length diverges. The Mott-insulator phase transition of the one-dimensional Hubbard model at half filling is one such case. Despite the limitations, it appears that our MLKS scheme can still be useful to explore similar models of relevance either at fundamental level or for possible applications in materials science. For instance, the extension to

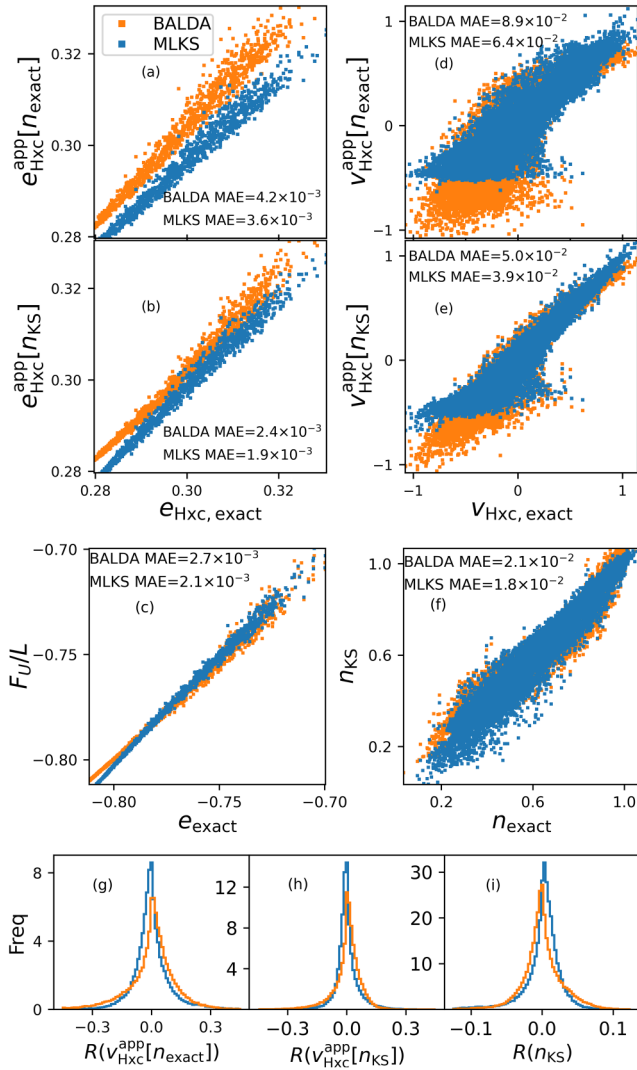


FIG. 14. Analysis of the accuracy of the KS ML functional. For a system $L = 60$ we present the parity plot of both the ML Hartree-exchange-correlation energy density, $e_{\text{Hxc}}^{\text{ML}}$, and the KS Hartree-exchange-correlation potential, $v_{\text{Hxc}}^{\text{ML}}$, computed either at the exact (DMRG) occupations [panels (a) and (d), respectively] or at the converged KS occupations [panels (b) and (e), respectively]. In panels (c) and (f) we present the parity plot for the ML KS total energy density and occupations, respectively. Results are presented for the functional with a nonlocality parameter $a = 0$ and over a test set of 1000 configurations.

higher dimensions is in reach and only limited by our ability to generate exact results for the training. Also intriguing is the possibility to extend our construction to the canonical ensemble [37] and to bosonic degrees of freedom [73]. One can then investigate phenomena such as superconductivity or polaron formation and transport.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [48].

APPENDIX: MACHINE-LEARNING THE LOCAL FUNCTIONAL

In Sec. III A we have examined the parity plots of both the $a = 4$ functional as well as the BALDA. It is interesting to see how well the ML fully local, $a = 0$, functional compares to the BALDA. Thus, we perform the same test as in Sec. III A, however with $a = 0$ instead of $a = 4$. The results are given in Fig. 14. It is interesting to see that when the ML model is reduced from a semilocal functional to a fully local one, it acquires some of the same systematic errors that are displayed by the BALDA. The $a = 0$ MLKS returns lower MAE's for each of the six quantities [Figs. 14(a) through 14(f)]. This can be understood as follows: The BALDA is a functional that is exact in the case of the infinite homogeneous one-dimensional (1D) chain for any given value of the filling, whereas the $a = 0$ is a local model that relates an input density to an energy, that is optimized to be most accurate for the systems in the training set (2/3 filling). In fact, any discrepancy between the BALDA and the MLKS $a = 0$ functional implies that the latter does not recover the correct infinite limit of the homogeneous electro density.

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