Deep learning and the Schrödinger equation

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We have trained a deep (convolutional) neural network to predict the ground-state energy of an electron in four classes of confining two-dimensional electrostatic potentials. On randomly generated potentials, for which there is no analytic form for either the potential or the ground-state energy, the model was able to predict the ground-state energy to within chemical accuracy, with a median absolute error of 1.49 mHa. We also investigate the performance of the model in predicting other quantities such as the kinetic energy and the first excited-state energy.

Solving the electronic structure problem for molecules, materials, and interfaces is of fundamental importance to a large number of disciplines including physics, chemistry, and materials science. Since the early development of quantum mechanics, it has been noted, by Dirac among others, that “...approximate, practical methods of applying quantum mechanics should be developed, which can lead to an explanation of the main features of complex atomic systems without too much computation” [1]. Historically, this has meant invoking approximate forms of the underlying interactions (e.g. mean field, tight binding, etc.), or relying on phenomenological fits to a limited number of either experimental observations or theoretical results (e.g. force fields) [2–8]. The development of feature-based models is not new in the scientific literature. Indeed, prior even to the acceptance of the atomic hypothesis, van der Waals argued for an equation of state based on two physical features [9]. Machine learning (i.e. fitting parameters within a model) has been used in physics and chemistry since the dawn of the computer age. The term machine learning is new; the approach is not.

More recently, high-level ab initio calculations have been used to train artificial neural networks to fit high-dimensional interaction models [10–15], and to make informed predictions about material properties [16, 17]. These approaches have proven to be quite powerful, yielding models trained for specific atomic species or based upon hand-selected geometric features [18, 19]. Hand-selected features are arguably a significant limitation of such approaches, with the outcomes dependent upon the choice of input representation and the inclusion of all relevant features. This limitation is well known in the fields of handwriting recognition and image classification, where the performance of the traditional hand-selected feature approach has stagnated [21].

Such feature-based approaches are also being used in materials discovery [22–24] to assist materials scientists in efficiently targeting their search at promising material candidates. Unsupervised learning techniques have been used to identify phases in many-body atomic configurations [25]. In previous work, an artificial neural network was shown to interpolate the mapping of position to wavefunction for a specific electrostatic potential [26, 28], but the fit was not transferable, a limitation also present in other applications of artificial neural networks to partial differential equations [29, 30]. By transferable, we mean that a model trained on a particular form of partial differential equation will accurately and reliably predict results for examples of the same form (in our case, different confining potentials).

Machine learning can also be used to accelerate or bypass some of the heavy machinery of the ab initio method itself. In [31], the authors replaced the kinetic energy functional within density functional theory with a machine-learned one, and in [32] and [33], the authors “learned” the mappings from potential to electron density, and charge density to kinetic energy, respectively.

Here, we use a fundamentally different approach inspired by the successful application of deep convolutional neural networks to problems in computer vision [34–37] and computational games [38, 39]. Rather than seeking an appropriate input representation to capture the relevant physical attributes of a system, we train a highly flexible model on an enormous collection of ground-truth examples. In doing so, the deep neural network learns both the features (in weight space) and the mapping required to produce the desired output. This approach does not depend on the appropriate selection of input representations and features, and as such, we call this “featureless learning”. Such an approach may offer a more scalable and parallizable approach to large-scale electronic structure problems than existing methods can offer.

In this Letter, we demonstrate the success of a featureless machine learning approach, a convolutional deep
neural network, at learning the mapping between a confining electrostatic potential and quantities such as the ground state energy, kinetic energy, and first excited-state of a bound electron. The excellent performance of our model suggests deep learning as an important new direction for treating multi-electron systems in materials.

It is known that a sufficiently large artificial neural network can approximate any continuous mapping \[ f \], but the cost of optimizing such a network can be prohibitive. Convolutional neural networks make computation feasible by exploiting the spatial structure of input data \[ \mathcal{D} \], similar to how the neurons in the visual cortex function. When multiple convolutional layers are included, the network is called a deep convolutional neural network, forming a hierarchy of feature detection. This makes them particularly well suited to data rooted in physical origin, since many physical systems also display a structural hierarchy. Applications of such a network structure in the field of electronic structure, however, are few (although recent work focused on training against a geometric matrix representation looks particularly promising).

Developing a deep learning model involves both the design of the network architecture and the acquisition of training data. The latter is the most important aspect of a machine learning model, as it defines the transferability of the resulting model. We investigated four classes of potentials: simple harmonic oscillators (SHO), “infinite” wells (IW, i.e. “particle in a box”), double-well inverted Gaussians (DIG), and random potentials (RND). Each potential can be thought of as a grayscale image: a grid of floating-point numbers.

We implemented a standard finite-difference method to solve the eigenvalue problem

\[
\hat{H} \psi = (\hat{T} + \hat{V}) \psi = \varepsilon \psi
\]

for each potential \( V \) we created. The potentials were generated with a dynamic range and length scale suitable to produce ground-state energies within a physically relevant range. With the random potentials, special care was taken to ensure that some training examples produced non-trivial wavefunctions (Fig. 2). Atomic units
are used, such that $\hbar = m_e = 1$. The potentials are represented on a square domain from $-20$ to $20$ a.u., discretized on a $256 \times 256$ grid. As the simple harmonic oscillator potentials have an analytic solution, we used this as reference with which to validate the accuracy of the solver. The median absolute error between the analytic and the calculated energies for all simple harmonic oscillator potentials was $0.12$ mHa. We discuss the generation of all potentials further in the Supplementary Information.

The simple harmonic oscillator presents the simplest case for a convolutional neural network as there is an analytic solution dependent on two simple parameters ($k_x$ and $k_y$) which uniquely define the ground-state energy of a single electron ($\varepsilon_0 = \frac{1}{2} (k_x^2 + k_y^2)$). Furthermore, these parameters represent a very physical and visible quantity: the curvature of the potential in the two primary axes. Although these parameters are not provided to the neural network explicitly, the fact that a simple mapping exists means that the convolutional neural network need only learn it to accurately predict energies.

A similar situation exists for the infinite well. Like the simple harmonic oscillator, the ground state energy depends only on the width of the well in the two dimensions ($\varepsilon_0 = \frac{1}{2} \pi^2 \hbar^2 (L_x^2 + L_y^2)$). It would be no surprise if even a modest network architecture is able to accurately “discover” this mapping. An untrained human, given a ruler, sufficient examples, and an abundance of time would likely succeed in determining this mapping.

The double-well inverted Gaussian dataset is more complex in two respects. First, the potential, generated by summing a pair of 2D-Gaussians, depends on significantly more parameters: the depth, width, and aspect ratio of each Gaussian, as well as the relative positions of the wells will impact the ground state energy. Furthermore, there is no known analytical solution for a single electron in a potential well of this nature. There is, however, a concise function which describes the underlying potential, and while this is not directly accessible to the convolutional neural network, one must wonder if the existence of such simplifies the task of the convolutional neural network. Gaussian confining potentials appear in works relating to quantum dots \[49, 50\].

The random dataset presents the ultimate challenge. Each random potential is generated by a multi-step process with randomness introduced at numerous steps along the way. There is no closed-form equation to represent the potentials, and certainly not the eigenenergies. A convolutional neural network tasked with learning the solution to the Schrödinger equation through these examples would have to base its predictions on many individual features, truly “learning” the mapping of potential to energy. One might question our omission of the Coulomb potential as an additional canonical example. The singular nature of the Coulomb potential is difficult to represent within a finite dynamic range, and, more importantly, the electronic structure methods that we would ultimately seek to reproduce already have frameworks in place to deal with these singularities (e.g. pseudopotentials).

We chose to use a simple, yet deep neural network architecture (shown in Fig. 1) composed of a number of repeated units of convolutional layers, with sizes chosen for a balance of speed and accuracy (inset of Fig. 3). We used the AdaDelta \[51\] optimization scheme to minimize the loss function (Fig. 3), the mean-squared error between the true energy and the convolutional neural network prediction. A detailed explanation of how we arrived at this network and the training process is provided in the Supplementary Information. We have used a custom-built multi-GPU (graphical processing unit) implementation of TensorFlow \[52\] to train the neural network. Unless otherwise specified, all training datasets consisted of 200,000 training examples and training was run for 1000 epochs. All reported errors are based on evaluating the trained model on validation datasets consisting of 50,000 potentials not accessible to the network during the training process.

Fig. 4(a-d) displays the results for the simple harmonic oscillator, infinite well, double-well inverted Gaussian, and random potentials. The simple harmonic oscillator, being one of the simplest potentials, performed extremely well. The trained model was able to predict the ground state energies with a median absolute error (MAE) of $1.51$ mHa.

The infinite well potentials performed moderately well with a MAE of $5.04$ mHa. This is notably poorer than the simple harmonic oscillator potentials, despite their similarity in being analytically dependent upon two simple parameters. This is likely due to the sharp discontinuity associated with the infinite well potentials, combined...
with the sparsity of information present in the binary-valued potentials.

The model trained on the double-well inverted Gaussian potentials performed moderately well with a MAE of 2.70 mHa and the random potentials performed quite well with a MAE of 2.13 mHa. We noticed, however, that the loss was not completely converged at 1000 epochs, so we provided an additional 200,000 training examples to the network and allowed it to train for an additional 1000 epochs. With this added training, the model performed exceptionally well, with a MAE of 1.49 mHa, below the threshold of chemical accuracy (1 kcal/mol, 1.6 mHa). In Fig. 4(d), it is evident that the model performs more poorly at high energies, a result of the relative absence of high-energy training examples in the dataset. Given the great diversity in this latter set of potentials, it is impressive that the convolutional neural network was able to learn how to predict the energy with such a high degree of accuracy.

Now that we have a trained model that performs well on the random test set, we investigated its transferability to another class of potentials. The model trained on the random dataset is able to predict the ground-state energy of the double-well inverted Gaussian potentials with a MAE of 2.94 mHa. We can see in Fig. 5(c) that the model fails at high energies, an expected result given that the model was not exposed to many examples in this energy regime during training on the overall lower-energy random dataset. This moderately good performance is not entirely surprising; the production of the random potentials includes an element of Gaussian blurring, so the neural network would have been exposed to features similar to what it would see in the double-well inverted Gaussian dataset. However, this moderate performance is testament to the transferability of convolutional neural network models. Furthermore, we trained a model on an equal mixture of all four classes of potentials. It performs moderately with a MAE of 5.90 mHa. This error could be reduced through further tuning of the network architecture allowing it to better capture the higher variation in the dataset.

The total energy is just one of the many quantities associated with these one-electron systems. To demonstrate the applicability of deep neural network to other quantities, we trained a model on the first excited-state energy $\epsilon_1$ of the double-well inverted Gaussian potentials. The model achieved a MAE of 10.93 mHa. We now have two models capable of predicting the ground-state, and first excited-state energies separately, demonstrating that a neural network can learn quantities other than the ground-state energy.

The ground-state and first excited-state are both eigenvalues of the Hamiltonian. Therefore, we investigated the training of a model on the expectation value of the kinetic energy, $\langle \hat{T} \rangle = \langle \psi_0 | \hat{T} | \psi_0 \rangle$, under the ground state wavefunction $\psi_0$ that we computed numerically for the random potentials. Since $\hat{H}$ and $\hat{T}$ do not commute, the prediction of $\langle \hat{T} \rangle$ can no longer be summarized as an eigenvalue problem. The trained model predicts the kinetic energy value with a MAE of 2.98 mHa. While the spread of testing examples in Fig. 5(a) suggests the model performs more poorly, the absolute error is still small.
We note that many other machine learning algorithms exist and have traditionally seen great success, such as kernel ridge regression [18, 20, 32, 53, 55] and random forests [18, 56]. Like these algorithms, convolutional deep neural networks have the ability to “learn” relevant features and form a non-linear input-to-output mapping without prior formulation of an input representation [17, 77]. In our tests, these methods perform more poorly and scale such that a large number of training examples is infeasible. We have included a comparison of these alternative machine learning methods in the Supplementary Information, justifying our decision of using a deep convolutional neural network. One notable limitation of our approach is that the efficient training and evaluation of the deep neural network requires uniformity in the input size. Future work will focus on an approach that would allow transferability to variable input sizes.

In summary, convolutional deep neural networks are likely particularly well suited for electronic structure calculations as they are designed for data which has a spatial encoding of information. As the number of electrons in a system increases, the computational complexity grows polynomially. Accurate electronic structure methods (e.g. coupled cluster) exhibit a scaling with respect to the number of particles of $N^7$ and even the popular Kohn-Sham formalism of density functional theory scales as $N^3$ [58, 59]. The evaluation of a convolutional neural network exhibits no such scaling, and while the training process for more complicated systems would be more expensive, this is a one-time cost.

In this work, we have taken a simple problem (one electron in a confining potential), and demonstrated that a convolutional neural network can automatically extract features and learn the mapping between $V(r)$ and the ground-state energy $\varepsilon_0$ as well as the kinetic energy $(T)$, and first excited-state energy $\varepsilon_1$. Although our focus here has been on a particular type of problem, namely an electron in a confining 2D well, the concepts here are directly applicable to many problems in physics and engineering. Ultimately, we have demonstrated the ability of a deep neural network to learn, through example alone, how to rapidly approximate the solution to a set of partial differential equations. A generalizable, transferable deep learning approach to solving partial differential equations would impact all fields of theoretical physics and mathematics.

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