Efficient prediction of density functional theory Hamiltonian with graph neural network

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ABSTRACT: Despite the past decades have witnessed many successes of machine learning methods in predicting physical properties, the prediction of Hamiltonian and thus electronic properties is still out of satisfactions. Here based on the graph neural network architecture, we present an extendible neural network model to learn the Hamiltonian from ab initio data with only local structures as inputs. The local coordinates, encoded using the convolutional neural network and designed to reserve the Hermitian symmetry, is used to map hopping parameters onto local structures. We demonstrate the performance of our model using SiGe random alloys as an example. We show that, our neural network model, although trained using small-size systems, can predict Hamiltonian as well as electronic properties such as band structures and densities of states (DOS) for large-size systems within the ab initio accuracy, justifying its extendibility. In combination with the high efficiency of our model, i.e., it takes only seconds to get the Hamiltonian of a 1728-atom system, our work provides a general framework to efficiently and accurately predict the electronic properties, which provides new insights into computational physics and will accelerate the research for large-scale materials.

1. Introduction

Hamiltonian lies in the heart of solid physics as it determines the electronic properties. The eigenstates of a Hamiltonian can then be used to obtain other properties. For periodic systems, Hamiltonian needs to be solved in the reciprocal space, which gives information of band structures. The past half century has witnessed the development of solving Hamiltonian problems based on the density functional theory (DFT)1, which has achieved great success in understanding electronic properties. However, with the system size increasing, the cost of solving Hamiltonian using DFT increases dramatically, which has become one of the greatest challenges in the solid physics field.

Recently, the development of machine learning methods has shed light on solving Hamiltonian of large systems2-4. One inspiring and successful case is the neural network (NN) potential models5-7, which learn atomic energy and force information from the first-principles data and generate numerical interatomic potentials, thus enabling fast and accurate molecular dynamics simulations of large systems. In fact, attempts using machine learning models for property predictions have also been made by kind of forcibly relating the global atomic structures to physical properties8-9 using data from public database such as the Materials Project10. However, these models generally yield low accuracy and poor extendibility to predict electronic properties such as bandgaps, defects, and optical properties.

Mapping Hamiltonian onto some locality is crucial to design an extendable model11-12. Inspired by the tight binding method13, the Hamiltonian can be represented by hopping parameters connecting the localized basis functions. Using the numerical pseudo-atomic orbitals (PAOs) ϕ(r)
as basis\textsuperscript{14}, the hopping parameter between atoms \( i \) and \( j \) is defined by:

\[
h^{(i,j)}_{\alpha\beta} = \langle \phi_{\alpha}(r - \tau_i) | \hat{H} | \phi_{\beta}(r - \tau_j - R_n) \rangle
\]

(1)

where \( \alpha \) and \( \beta \) denote the orbitals, \( \tau \) is the atomic position and \( R_n \) is the lattice vector. Note that, hopping parameters have locality and can be determined by local structures because hopping can be negligible when atomic distance is larger than a certain cutoff radius. Therefore, the Hamiltonian matrix, which are constructed by hopping parameters, can be determined from local structures, and thus an extendable model for predicting Hamiltonian is theoretically feasible. In previous studies, Hegde and Bowen proposed a model for predicting Hamiltonians in a small basis set and studied the s-orbital Cu and sp\textsuperscript{3}-orbital diamond systems\textsuperscript{15}. Schütz \textit{et al.} developed the SchNOrb model for small individual molecules with data augmentation\textsuperscript{16}. Gu \textit{et al.} introduced an NN representation of tight binding Hamiltonians for one-dimensional carbon chains\textsuperscript{17}.

However, because the Hamiltonian matrix is covariant under rotations of coordinates, prediction of hopping parameters is more complicated than predicting measurable properties such as bandgaps which are independent of the choices of coordinate systems. Although the data augmentation technique can reflect rotations\textsuperscript{16}, it is obviously inefficient. To fix the rotation freedoms, Li \textit{et al.} recently introduced the local coordinate (LC)\textsuperscript{18} for each atom pair based on the local structures so that the hopping parameters can be trained in the LC and then transformed back to the global coordinate to construct the Hamiltonian matrix. Using the atomic-like basis functions, the rotational transformations can be well described by the Wigner-D matrix. Nevertheless, an efficient model for predicting Hamiltonian and thus electronic properties is still yet to be established.

In this work, based on the graph neural network architecture and LC transformation, we develop an extendable model to predict the DFT Hamiltonian. The node and edge features are updated by message passing with attention mechanism. The LC information is encoded using the convolutional neural network and designed to reserve the Hermitian symmetry. The output of the model is the triangular portion of the hopping parameter matrix, and then all the hopping parameters are assembled into the Hamiltonian matrix. Using the SiGe random alloy systems as an example, we demonstrate that, our NN model, trained using small-size systems, can predict Hamiltonian as well as electronic properties such as band structures and densities of states (DOS) for large-size systems within the DFT accuracy but with very little cost, i.e., it takes only about 10 seconds to get the Hamiltonian of a 1728-atom system. In addition, the linear scaling with the number of atoms of our NN model makes it very promising for studying electronic properties of large-scale systems.

2. \textbf{Results}

\textbf{Architecture}

In our neural network model for predicting Hamiltonian, the nodes represent atoms and the edges represent bonds between atoms. The inputs include the atomic number \( \{Z_i\} \), the edge distances \( \{|r_{ij}|\} \), and the LC information \( \{|y_{ij}|\} \) which will be introduced in the following. The output consists of the upper triangular portion of each hopping matrix. The architecture of our neural network model is shown in Fig. 1. It consists of an embedding layer, an LC layer, several interaction layers, and an output layer.
Figure 1. The architecture of our neural network model. (a) The model consists of an embedding layer, an LC layer, five interaction layers, and an output layer. The output of LC layer is passed to each interaction layer. (b) Embedding layer. The atomic numbers are transformed to the initial node features through an embedding network. The interatomic distances are expanded by the Gaussian radial basis functions (RBF) to generate initial edge features. (c) LC layer with two filters for an example. (d) Interaction layer. The node features and edge features are updated. (e) Output layer.

Embedding layer

In the embedding layer as shown in Fig. 1b, the initial node features $v_i^{(0)} \in \mathbb{R}^N_i$ are calculated through an embedding network:

$$v_i^{(0)} = \text{Embedding} (Z_i), \quad (2)$$

and the initial edge features $e_{ij}^{(0)} \in \mathbb{R}^N_{ij}$ are the Gaussian radial basis functions\(^\text{19}\):

$$e_{ij}^{(0)} = \exp \left(-\frac{(|r_{ij}| - r_c)^2}{\sigma^2}\right), \quad (3)$$

where the centers $r_i$ are placed linearly between 0 and the cutoff radius $R_c$, and $\sigma^2$ is set to 0.0044 Å\(^2\).

LC layer

The LC layer is used to obtain the LC information for each edge $e_{ij}$ which is uniquely determined by the local structures of atoms $i$ and $j$. First, the local axis used to identify the LC for
the edge $ij$ is constructed with details in the “Methods” section. Then the LC information can be represented by the real spherical harmonics and distances. Here we use the edge $ij$ as an example to illustrate how LC information is obtained. First, the neighbors of atom $i$, denoted by $k$, are identified and sorted by distances $|r_{ik}|$. Then we calculate the real spherical harmonics of bond $ik$, denoted by $Y_{lm}(\theta_{ik}, \phi_{ik})$, where $\theta_{ik}$ and $\phi_{ik}$ are the polar and azimuthal angles, respectively. Each bond $ik$ can be represented by a list of real spherical harmonics ($0 \leq l \leq 4; -l \leq m \leq l$), yielding the orientational vector $y_{ik}^{ij} \in \mathbb{R}^{N \times N}$:

$$y_{ik}^{ij} = \frac{Y_{lm}(\theta_{ik}, \phi_{ik})}{1 + |r_{ik}|},$$

(4)

where $N_j$ is the number of neighbors of atom $i$ and $N_j = 25$ is the number of spherical harmonic functions. Similarly, for the atom $j$ we can obtain the orientational vector $y_{jk}^{ij} \in \mathbb{R}^{N \times N}$.

To extract the LC features $s_{ij}$ from the orientational vectors $y_{ik}^{ij}$ and $y_{jk}^{ij}$, we utilize the convolutional neural network\textsuperscript{20-21} architecture as shown in figure 1(c). First, we restrict $N_i = N_j$ so that the two orientational vectors $y_{ik}^{ij}$ and $y_{jk}^{ij}$ can be concatenated as the input of the LC layer $S_{ij}^{(0)} \in \mathbb{R}^{N \times N \times 2}$. Then a convolutional operator with a filter $w \in \mathbb{R}^{h \times 2}$ is applied to a window of size $h$ to produce a new feature. Performing the convolution operator on the input $S_{ij}^{(0)}$ generates a new feature map $c \in \mathbb{R}^{N \times h \times 1}$. Then max pooling is performed to extract a scalar from each feature map. We have used three window sizes: 2, 3 and 4, each of which has 64 filters. Thus, for each LC, 192 new features are extracted by the convolutional operator, and then these features are passed to a dense layer to generate outputs $s_{ij} \in \mathbb{R}^{N \times 1}$.

Another important thing is that, in order to keep the Hermitian property of the Hamiltonian, the LC has to be determined by two different protocols as described in “Methods” section. Consequently, we train two models with the same architecture to construct the full Hamiltonian matrix and each model outputs the upper triangular matrix elements.

**Interaction layer**

After the processing of the embedding layer and the LC layer, information is passed to the multiple stacked interaction layers. In the interaction layer, the attention mechanism\textsuperscript{22-23} and skip connection are used for updating node features. The LC features are concatenated to the corresponding edge features. As shown in figure 1(d), the node features $v_i \in \mathbb{R}^{N}$ are updated as follows:
\[ v_i^{(l)} = \sum_{k \in \mathcal{N}(i)} \alpha_{i,k} \Phi v_k^{(l-1)} + \nu_i^{(l-1)}, \]

where \( k \) denotes the neighbor of \( i \), \( l \) refers to the \( l \)-th interaction layer, and \( \Phi \) denotes a dense layer. The attention coefficient \( \alpha_{i,k} \) are computed as:

\[ \alpha_{i,k} = \frac{\exp\left(a^T \text{LeakyReLU}(\Phi[v_i \parallel v_k \parallel e_{ik} \parallel s_k])\right)}{\sum_{k \in \mathcal{N}(i)} \exp\left(a^T \text{LeakyReLU}(\Phi[v_i \parallel v_k \parallel e_{ik} \parallel s_k])\right)}, \]

where \( a^T \) is a learnable weight variable and \( \parallel \) denotes concatenation. After the node features are updated, the edge features are updated as follows:

\[ e_{ij}^{(l)} = \Phi\left[h\text{-swish}\left[\Phi(v_i^{(l)} \parallel v_j^{(l)} \parallel e_{ij}^{(l-1)} \parallel s_{ij})\right]\right], \]

where \( h\text{-swish} \) is the hard version of the swish activation function:

\[ h\text{-swish}[x] = x \frac{\text{ReLU}(x + 3)}{6}. \]

\section*{Output layer}

The output layer consists of two stacked dense layers with \( h\text{-swish} \):

\[ h_{ij} = \Phi\left[h\text{-swish}\left[\Phi(v_i \parallel v_j \parallel e_{ij} \parallel s_{ij})\right]\right], \]

where \( h_{ij} \in \mathbb{R}^{N_i} \) is the upper triangular portion of the hopping matrix of \( ij \). Finally, the \( h_{ij} \) of all edges are assembled into the Hamiltonian matrix.

\section*{Evaluation}

With the above network for learning Hamiltonian in hand, we then turn to real systems to demonstrate the feasibility of learning electronic properties using the NN methods. Here we use the three-dimensional SiGe random alloys as an example. Our goal is to map the local atomic structures to the hopping parameters \( h_{\alpha_i,j}^{(K)} \) using our developed NN models. The \( s_2p_2d_1 \) PAOs are used as the basis functions and the hopping parameters for each pair \( ij \) are represented by a \( 13 \times 13 \) matrix. The training set contains \( 66 \times 2 \times 2 \times 2 \) supercells and \( 1000 \times 3 \times 3 \times 3 \) supercells, in which equivalent structures have been filtered. Details for data generation are provided in the “Methods” section. Then randomly generated structures are used for the testing set. Note that, the testing structures can have much larger sizes than the training structures and the largest testing structure we used is a \( 6 \times 6 \times 6 \) supercell consisting of 1728 atoms and 25,406,784 hopping parameters. All the electronic structures are calculated using the DFT methods (see the Methods section). After training the models, the band structures and density of states (DOS) are calculated using the learned Hamiltonian.

The mean absolute error (MAE) between the DFT calculated and learned hopping parameters is used to evaluate our NN model. For the training set, the MAEs of the two models are 0.50 meV and 0.52 meV, respectively. For the testing set, the MAEs range from 0.5 to 0.8 meV as shown in Fig. 2(a). The comparisons between the predicted hopping parameters and the DFT results for the
testing structures are also shown in Fig. 2b. As can be seen, our models give very good predictions of hopping parameters.

The predictive power of our NN Hamiltonian model is further demonstrated by calculating the physical properties using the predicted hopping parameters. The band structure and DOS of a 2×2 supercell in the testing set are shown in figure 3(a). The results obtained by the NN Hamiltonian and by the DFT calculation agree with each other perfectly. For larger supercells, the band structures are not shown due to the heavy band folding and we just give the DOS. Figure 3(b) shows the DOSs of the 3×3, 4×4, 5×5 and 6×6 supercells. The NN predicted results match very well with the DFT calculations.

Figure 2. Accuracy evaluation of our model. (a) MAE for testing set. (b) Element-wise error distributions of 3×3, 4×4, 5×5 and 6×6 supercells.

Figure 3. Comparison of band structures and DOSs obtained using DFT (red) and our model (blue). (a) Band structure and DOS of the 2×2 supercell. (b) DOSs of 3×3, 4×4, 5×5 and 6×6 supercells.

3. Discussion

The major advantage of the NN model over the DFT calculation is the computational efficiency. The computational cost of our NN model scales linearly with the number of atoms, compared with the cubic scaling of the DFT calculation. The inference times of our NN model are summarized in Table 1. For the 6×6 supercell with 1728 atoms, our model only costs 10.5 seconds to predict the system Hamiltonian. For comparison, the computational time for the DFT self-consistent calculation
is about 4800 seconds using 96 CPU cores, depends on the iteration steps. In this work, our model is trained using one GeForce RTX 3090 card, and the DFT calculations are performed using the Intel Xeon Platinum 9242 CPU.

Table 1. The inference time of our NN model.

| Supercell | 222  | 333  | 444  | 555  | 666  |
|-----------|------|------|------|------|------|
| Number of atoms | 64   | 216  | 512  | 1000 | 1728 |
| Inference Time (s) | 0.6  | 1.3  | 3.1  | 6.3  | 10.5 |

With the DFT accuracy and very high computational efficiency, our NN Hamiltonian model can be applied to study electronic properties of very large systems, such as high-entropy alloys\textsuperscript{25} or defect structures\textsuperscript{26}. Besides, our model is also expected to accelerate inverse design of materials with targeted electronic properties\textsuperscript{27-28} such as band gaps, where the DFT calculations are performed more than thousands times for structure relaxation and property evaluation. The structure relaxations can be efficiently performed using the NN potential models\textsuperscript{29}, and our NN Hamiltonian model can be used to evaluate the electric properties to avoid the computationally expensive DFT calculations.

In conclusion, based on the graph neural network architecture and using the local atomic structure information, we have developed an extendable NN model to predict the DFT Hamiltonian. We have designed the LC for each edge to fix the rotational freedom of the Hamiltonian matrix and reserve the Hermitian property of Hamiltonian to improve the efficiency. By employing SiGe random alloys as an example, we have demonstrated the high accuracy, extendibility, and efficiency of our model in predicting the Hamiltonian and electronic properties. Our work thus opens doors for studying the electronic properties of large-scale systems using machine learning methods.

4. Methods

Data generation

The SiGe random alloys are obtained by randomly occupying the zinc-blende lattice sites with the Si or Ge atoms. The number of possible combinations in a supercell with N sites is given by the combinatorial number $C(N, N/2)$, which could be incredibly large as the total atom number increases. Note that, most of the structures are identical under certain symmetry operations. Therefore, we develop a scheme to filter the identical structures.

Figure 4. Illustration of the two local structures of atom 0. The empty and full circles denote elemental type A and B, respectively.
For each atom, we construct its unique local structure information by its neighbors in the following way. As shown in Fig. 4, we first calculate the dot product of \( r_{A,0} \) and \( r_{B,0} \) (atomic positions with respect to the target atom 0), and then obtain a list of dot products for the target atom:

\[
\nu(0) = \{ r_{10} \cdot r_{30}, r_{10} \cdot r_{00}, r_{10} \cdot r_{40} \}.
\]

Then we sort the elements in the list \( \nu(0) \) in an ascending order to represent the local structure information. It is easy to check that the two local structures in figure 4 are different.

We calculate the local structure encodings by equation (10) for all atoms, remove duplicated ones and add the encodings into a database. When a new structure is generated, we compare the new encodings with those in database. If new local structures are found, then the generated structure is added into the training set and the database is updated accordingly. New structures are generated repeatedly until no more new local structures are found. In this work, we collect 66 samples of \( 2 \times 2 \times 2 \) supercells and 1000 samples of \( 3 \times 3 \times 3 \) supercells for training. The samples in the testing set are randomly generated with supercell sizes ranging from \( 2 \times 2 \times 2 \) to \( 6 \times 6 \times 6 \).

**DFT calculation**

Before calculating the hopping parameters, all the structures are relaxed with fixed cell using the Vienna ab initio Simulation Package\(^{30}\). The projector augmented wave (PAW)\(^{31}\) type pseudopotential and the local spin density approximation (LSDA) of Ceperley-Alder (CA)\(^{32}\) is used for the exchange-correlation functional.

Then we employ the numerical pseudo-atomic orbitals (PAOs) as implemented in OpenMX software\(^{33}\) to perform DFT calculations for the hopping parameters. The PAO is given by a product of a radial function \( R \) and a real spherical harmonic function \( Y \) as:

\[
\phi(r) = R(r)Y(\theta, \varphi),
\]

where \( R \) is defined numerically and finite within a cutoff radius. The Hamiltonian and the overlap matrices are given by:

\[
H_{\alpha,\beta}^{(k)} = \sum_n e^{R_n \cdot k} \langle \phi_{\alpha} (r - \tau_j) | \hat{H} | \phi_{\beta} (r - \tau_j - R_n) \rangle,
\]

\[
S_{\alpha,\beta}^{(k)} = \sum_n e^{R_n \cdot k} \langle \phi_{\alpha} (r - \tau_j) | \phi_{\beta} (r - \tau_j - R_n) \rangle,
\]

where \( i \) and \( \alpha \) denote atom and orbital, respectively, \( \tau \) is the atomic position and \( R_n \) is the lattice vector. According to the principle of locality, the hopping parameters \( h_{\alpha,\beta}^{(R_n)} \) are determined by the local structures of atoms \( i \) and \( j \) within certain cutoff radius. The overlap parameters \( s_{\alpha,\beta}^{(R_n)} \) are calculated by the PAOs without DFT calculations. Then the eigenvalues \( \nu_{\mu k} \) as well as
eigenstates \( E_{\mu k} \) of a system is obtained by solving the generalized eigenvalue problem:

\[
H^{(k)} \psi_{\mu k} = E_{\mu k} S^{(k)} \psi_{\mu k}.
\]  

(14)

The Si7.0-s2p2d1 and Ge7.0-s2p2d1 PAOs are used as the basis functions and the cut-off radius is set as 7.0 Bohr. For the supercells up to \(4 \times 4 \times 4\), the k-points are generated so that the spacing between k-points are smaller than 0.16 Å\(^{-1}\). For the \(5 \times 5 \times 5\) and larger supercells, single gamma point is used.

**Local coordinate (LC)**

For the onsite hopping \((i=j)\), the LC is determined by the nearest neighbor \(n_1\) and the second nearest neighbor \(n_2\) of atom \(i\). The x-axis is defined along the vector \(\mathbf{e}_1 = \mathbf{r}_{n_1} - \mathbf{r}_i\), the y-axis is defined along the vector \(\mathbf{e}_2 = \mathbf{e}_1 \times (\mathbf{r}_{n_2} - \mathbf{r}_i)\), and the z-axis is defined along the vector \(\mathbf{e}_3 = \mathbf{e}_1 \times \mathbf{e}_2\).

For the offsite hopping \((i \neq j)\), we note that the Hamiltonian operator is Hermitian, i.e., \(h_{\alpha \beta; \mu \nu} = h^*_{\beta \alpha; \mu \nu}\). With such constraint, only the upper triangular portion of each hopping matrix are needed to construct the Hamiltonian matrix. To utilize the Hermitian property, the hopping matrices \(h_{ij}\) and \(h_{ji}\) should be defined on the same LC. We calculate the distance from the nearest neighbor to the target atom \(d_{\text{min}}\). If \(d_{\text{min}}(i) < d_{\text{min}}(j)\), then the x-axis is defined along \(\mathbf{e}_1 = \mathbf{r}_j - \mathbf{r}_i\), and the y-axis is defined along \(\mathbf{e}_2 = \mathbf{e}_1 \times (\mathbf{r}_{n_1} - \mathbf{r}_i)\), where \(n_1\) denotes the nearest neighbor of atom \(i\). If \(d_{\text{min}}(i) > d_{\text{min}}(j)\), then the x-axis is defined along \(\mathbf{e}_1 = \mathbf{r}_i - \mathbf{r}_j\), and the y-axis is defined similarly by the nearest neighbor of atom \(j\). Some special cases should be treated separately, and the details are provided in the supplementary information.

The \(h_{ij}\) and \(h_{ji}\) are defined on the same LC and they also share the same input information. As a result, one model is not sufficient. According to the distances of the nearest neighbors, the LCs for offsite hopping are defined by two different ways, and the hopping parameters are also divided into two groups. The two groups are trained separately to obtain two models. For the sake of efficiency, the onsite hopping belongs to one of the offsite groups, and therefore we do not have to train a third model. Finally, the Hamiltonian matrix is constructed by combining the outputs of the two models.

**Training details**

The model is trained by minimizing the difference between the predicted data \(h_{ij}\) and the DFT calculated data \(\hat{h}_{ij}\). We note that the Hamiltonian data are highly imbalanced, that is to say, most
targets are close to zero while a few ones are large than 10 eV. Therefore, a regression version of the Focal loss\(^{34-35}\) is used:

\[
L(y, \hat{y}) = \frac{1}{n} \sum_{i=1}^{n} \left( \frac{1}{1 + e^{-\beta(y_i - \hat{y}_i)}} \right)^\gamma |y_i - \hat{y}_i| \tag{15}
\]

where \(\beta = 0.25\) and \(\gamma = 2.0\) are adjustable hyper-parameters.

Our model is developed with Pytorch-Geometric\(^{36}\) Python library. Adam optimizer was used with initial learning rate of 0.001. The reduce on plateau strategy is used to schedule learning rate, and the lower bound of the learning rate is 1e-5. The hyper-parameters are provided in the supplementary information.

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