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A new approach to potential fitting using neural networks

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Abstract

A methodology is presented for developing transferable empirical potential functions without following the usual procedure of postulating a functional form. Instead, a neural network (NN) is employed to learn the functional relationships of potential energy surfaces from the local geometric arrangement of atoms. The methodology is illustrated by training the NN model on tens of thousands of individual data points derived from the tight-binding (TB) method for a wide range of silicon systems including both small clusters and bulk structures. Comparisons of the potential's properties with experimental data, quantum methods and other Si potentials have been made.

The NN model successfully fitted energy variations of the different test cases as a function of bond distances, bond angles, lattice constants and elastic properties for both equilibrium and non-equilibrium small cluster and bulk structures. This indicates a robust and consistent methodology for fitting empirical potentials which can be applied to a wide range of materials independent of the type of bonding or their crystal structure.

Keywords: Neural network, tight-binding, empirical potentials
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I. Introduction

The problem of obtaining classical potential energy surfaces to describe accurately the properties of how particles interact is a long standing one and so far there is no consensus on the best approach to take despite the fact that such classical potentials are being ever more applied to a wide range of problems in materials science. For close nuclear separation it is generally accepted that the screened Coulomb (pair) potentials can provide a good description of the interactions with the ZBL [1] potential being most commonly used both in binary collision and Molecular Dynamics (MD) codes. Potentials to model bonding have less widespread acceptance and progress in obtaining good potential functions to model near equilibrium structures has been slow; most potential functions that are commonly used in computer simulations having their origins in the 1980's. Those that model the bonding interactions fall basically into three categories, the bond order [3, 4, 5], Stillinger-Weber [6] or Modified embedded atom type [7]. Generally these potentials assume a given functional form and then try to fit a wide variety of experimental and other data to the functional form chosen. The result is not entirely successful with for example the Tersoff potential producing the wrong melting temperature by a factor of 2 and the Stillinger-Weber potential wrongly predicting the energetics of small clusters.

At the last COSIRES meeting in Helsinki, Thijsse [2] described the most important problems with all the popular classical potentials used in the modelling of dynamic processes in Si. The main problem defined by Thijsse is that when dealing with dynamical processes at a surface and within the bulk of a material, parts of the configuration space are tested to which the potential has not been fitted. Thijsse made some improvements to the potential description and it was suggested that a modified embedded atom method [7] could provide the basis for a more satisfactory potential for modelling sputtering of Si surfaces. Potentials to describe sputtering have to be especially robust because they need to describe bulk and surface properties as well as the properties of small clusters that may be ejected during the sputtering process.

Another approach had also been suggested at the COSIRES meeting held at Pennsylvania State University [8, 9] through the use of neural networks (NN's) to fit the potential energy surface. In that case only the feasibility of the method was examined showing how the many-body term in the Tersoff potential could be fitted by a neural network whose inputs depended only on the local geometry of an Si atom. Some MD simulations were carried out which showed that there was a reduction in speed of the simulations to about 60% of

those using the Tersoff potential directly. NN's provide a useful and effective tool for modelling complex processes as they are able to extract the functional relationships between model inputs and outputs from the data without requiring explicit consideration of the actual data generation process.

In this paper we take the process one stage further by developing a NN formulation that is fitted to thousands of data point obtained from a single source, namely Tight Binding (TB) calculations. To enable the NN potential to be environment-dependent, we generated training data for a range of atomic configurations where the co-ordination numbers were different and also for non-equilibrium structures. We tested our model over a wide set of test cases for which data was not fitted during the training process, and, consequently, were unseen by the NN. The description of the model and its predictions are described in the following sections.

II. Development of the NN potential

A. The variables for the problem

Given a system consisting of a total of N_{atom} atoms and its initial configuration $\{\mathbf{R}\}$ of atomic coordinates, we use the neural network model to define the energy, E_i , of each atom i such that

$$E_i(\{\mathbf{R}\}) = Y(\mathbf{X}_i(\{\mathbf{R}\}), \mathbf{w}) \quad (1)$$

where $\mathbf{w}$ is the vector of the optimal parameters (weights and biases) of the NN model and $\mathbf{X}_i(\{\mathbf{R}\})$ is the set of vectors of input variables derived from $\{\mathbf{R}\}$ and Y is the composition of activation functions representing the model.

Accordingly, the total energy, E_{total} , of the system is given by

$$E_{\text{total}}(\{\mathbf{R}\}) = \sum_i^{N_{\text{atom}}} E_i(\{\mathbf{R}\}) \quad (2)$$

The input variables are based on the geometry of the system. We predict E_i by considering inputs derived from each uniquely defined 4-atom chain $i - j - k - l$ between atoms i , j , k and l in the given sequence (see figure 1).

In the chain, adjacent atoms are nearest neighbours and $i \neq j \neq k \neq l$. In order to limit the large number of possible combinations of chains, atoms within a given first neighbour cut-off distance are only considered to be first neighbours if they are unscreened by intermediate atoms, see figure 2. In (a) atom j is regarded as a first neighbour

Figure 1: A 4-atom chain illustrating some input variables listed in table 1.

of atom i whereas in (b) atom k ‘screens’ j from i . This leads to a screening function S_{ij} between atoms i and j . The full details of S_{ij} and the screening procedure is given in [10], based on the idea put forward by Baskes [7].

Figure 2: The screening procedure. In (a) atom j is a first neighbour of atom i and in (b) atom j is not considered a first neighbour of atom i even with the same value of r_{ij} as in (a) because of the presence of atom k .

We have used 9 input variables to describe the structural properties of the systems. These variables are listed in table 1.

The uniqueness of the 4-atoms chain is accounted for by the inclusion of the torsional angle τ_{ijkl} in the training set. The first 6 variables in the list are related to the geometry of an $i - j - k - l$ chain as depicted in figure 1. The remaining 3 variables – N_i , N_j and N_{inputs} – provide information about the extent of screening present in forming different $i - j - k - l$ sequences. The variable N_{inputs} would be numerically equal to the number of input vectors for atom i in the network in the case where all i ’s neighbours were all unscreened. Its definition in terms of the screening functions allows N_{inputs} to be a continuous function so that new bonds are introduced smoothly. This together with the screening functions that multiply the weights of the connections between the nodes in the first and second hidden layers forces the output of the network, the energy per atom, to be a continuous function of the input variables.

C. The neural network model

variable	description
r_{ij}	length of bond $i - j$
r_{jk}	length of bond $j - k$
$\cos \theta_{ijk}$	cosine of angle between bonds $i - j$ and $j - k$
r_{kl}	length of bond $k - l$
$\cos \theta_{jkl}$	cosine of angle between bonds $j - k$ and $k - l$
$\cos \tau_{ijkl}$	cosine of torsion angle between bonds $i - j$ and $k - l$
$N_i = \sum_{i \neq j} S_{ij}$	sum of screening present over every bond $i - j$ formed per atom i
$N_j = \sum_{i \neq j, j \neq k} S_{jk}$	sum of screening present over every bond $j - k$ formed per atom j
$N_{inputs} = \sum_{i \neq j} S_{ij} [1 + \sum_{i \neq j \neq k} S_{jk} (1 + \sum_{i \neq j \neq k \neq l} S_{kl})]$	sum of screening present over every $i - j - k - l$ chain formed

Table 1: List of input variables used in training the NN model.

The neural network is developed in such a way that the input layer can vary with the number of 4-atom chains that can be formed. This is non-standard and was previously used by our group in the Tersoff potential fitting [9]. In addition the potential is constructed to be a continuous function of the input variables and invariant to the ordering of the inputs. Figure 3 illustrates the NN model with a feedforward connection. The output of the model is the total energy, E_i , of atom i and the input variables $x_p^{(n)}$, $p = 1, 2, \dots, 9$, are elements of the input vector $\mathbf{x}_i^{(n)}$ which is defined as

$$\mathbf{x}_i^{(n)} = \left(r_{ij}^{(n)}, r_{jk}^{(n)}, \cos \theta_{ijk}^{(n)}, r_{kl}^{(n)}, \cos \theta_{jkl}^{(n)}, \cos \tau_{ijkl}^{(n)}, N_i, N_j, N_{inputs} \right)^T \quad (3)$$

The index n runs over the N 4-atoms chains that are formed and the collection of the input vectors $\{\mathbf{x}_i^{(1)}, \mathbf{x}_i^{(2)}, \dots, \mathbf{x}_i^{(N)}\}$ forms the set $\mathbf{X}_i$ (equation (1)). It can be seen that the variables N_i , N_j and N_{inputs} are independent of n . The inputs only operate at full strength when there is no screening through multiplication by the screening factor which reduces their influence as the bonds become weaker. This is required for continuity.

The NN shown in figure 3 was arrived at through an evolution process that tested many different types of neural networks with varying numbers of hidden layers and methods for connecting the weights between the layers. This NN model turned out to be the most efficient in fitting all the data supplied to it. For a description of all the various models tested see [10].

Figure 3: The NN model used to fit potential energy surfaces. The x 's are the input variables, $S^{(n)}$ are screening factors, w^{HI} 's are the weights connecting the first hidden layer to the input layer, w^{HH} are weights connecting the two hidden layers and w^{OH} are weights between the second hidden layer and the output layer. The y 's represent the outputs of each neuron. Each neuron also has an associated bias, b_i^α , $\alpha = A, B, C$.

Each set n of input variables has a one-to-one correspondence with a set of neurons in the first hidden layer containing a fixed number, Q , of neurons. Every weight w_{qp}^{HI} between the two sets of nodes is multiplied by the factor $S^{(n)}$ which is the extent of screening in the bond $i - j$, *i.e.*, $S^{(n)} = S_{ij}^{(n)}$.

To meet the requirement of the potential to be invariant to any ordering of input data, the weights and the biases between variable sets of neurons are shared. Thus, for connections between the input and the first hidden layer we have

$$w_{qp}^{HI(1)} = w_{qp}^{HI(2)} = \dots = w_{qp}^{HI(N)} = w_{qp}^{HI} \quad (4)$$

and, for those between the two hidden layers we have

$$w_{rq}^{HH(1)} = w_{rq}^{HH(2)} = w_{rq}^{HH(N)} = w_{rq}^{HH}, \text{ if } r = q \quad (5)$$

for $p = 0, 1, \dots, P$, and $q = 1, 2, \dots, Q$ and $r = 1, 2, \dots, Q$.

III. Fitting Process

A. The tight-binding formalism

The training data for the NN potential was generated using the Frauenheim non-orthogonal TB method [11] from PLATO [12]. Here we outline the representation of the total energy of the TB model in terms of the onsite and bond energies. The total energy, E_{total} , of a system of N_{atom} atoms is written as a sum of the band-structure energy, E_{bs} , and the repulsive potential $\Phi(r_{ij})$, *i.e.*,

$$E_{\text{total}} = E_{bs} + \frac{1}{2} \sum_{j \neq i}^{N_{\text{atom}}} \Phi(r_{ij}) \quad (6)$$

The total energy in equation (6) can be rewritten as

$$E_{\text{total}} = \sum_i^{N_{\text{atom}}} \left(O_i + \frac{1}{2} \sum_{j \neq i}^{N_{\text{atom}}} B_{ij} \right) = \sum_i^{N_{\text{atom}}} E_i(\{\mathbf{R}\}) \quad (7)$$

where O_i is the onsite energy of atom i and is given by

$$O_i = \sum_{i\alpha\beta} \rho_{i\alpha i\beta} H_{i\alpha i\beta} \quad (8)$$

and B_{ij} is the bond energy between atoms i and j , and is given by

$$B_{ij} = \sum_{\substack{i\alpha j\beta \\ i \neq j}} \rho_{i\alpha j\beta} H_{i\alpha j\beta} + \Phi(r_{ij}) \quad (9)$$

In equations (8) and (9), α and β span all the basis orbitals positioned on a given atom. For example, in the case of Si, a choice of minimal orbital basis can be the valence s , p_x , p_y and p_z orbitals. $\rho_{i\alpha j\beta}$ and $H_{i\alpha j\beta}$ are the elements of the density and the Hamiltonian matrices respectively.

B. Generating the data set

In order to ensure that each set of input vector $\mathbf{x}_i^{(n)}$ (equation (3)) has a fixed dimension, we had to account for the non-existence of bonds in forming the sequence $i - j - k - l$. An absent bond was modelled by an imaginary interaction at a distance r_{max} , the potential cut-off away. The associated missing bond angles were taken to be 180° .

The value of r_{max} was chosen as 4.140 Å which is the same cutoff radius employed in the TB. Furthermore, for a given system, if two or

more atoms have the same local geometry, then the information for only one of these atoms was considered.

Once the data set was generated it was divided into the training, validation and test sets. The validation set was used to prevent overfitting of the model by allowing early stopping. Furthermore, the training and the validation sets contained cases of the different systems in almost equal proportion. This allowed the NN to give equal weightings to the different configuration spaces. A total of 23,050 data points were fitted distributed between the dimer, linear trimer, angled trimer, tetramer, diamond, BC8, ST12, BTC5 AND β -tin structures and another 2050 used for validation and test purposes.

To find the optimal network parameters the cost function SSE was minimised.

$$SSE = \frac{1}{m} \sum_{m=1} (t^{(m)} - y^{(m)})^2 \quad (10)$$

where $t^{(m)}$ represents the desired energy and $y^{(m)}$ represents the NN output. Initially random sets of parameters were assumed and during training with not all choices minimising SSE sufficiently. The parameters (weights) of the network were updated by the equation:

$$\Delta \mathbf{w} = -(\mathbf{H} + \mu \mathbf{I})^{-1} \mathbf{g} \quad (11)$$

where, $\mathbf{w}$ is the vector of the parameters of the model, $\mathbf{I}$ is the identity matrix, μ is an adaptive parameter, $\mathbf{g}$ is the gradient vector of SSE with respect to the parameters and $\mathbf{H}$ is the Hessian matrix. μ is increased by a factor μ^+ if SSE increases and is decreased by a factor μ^- if it decreases. Initially μ was taken to be 0.1. μ^+ and μ^- were set to 2.0 and 0.5 respectively. It was found that reasonable accuracy could be achieved by using 11 hidden nodes in each of the two layers, together with two sets of 11 biases. The full parameters for the model are given in [10]. Each neuron in the model was associated with the same activation function given by the standard sigmoidal function $\varphi(x) = \frac{1}{1+e^{-x}}$. All input variables were linearly transformed to the interval $[0.1, 0.9]$ by using their respective minimum (x_{min}) and maximum (x_{max}) values from the data set.

IV. Results and Discussion

The main objective was to fit the NN potential as exactly as possible to the tight binding data and any inaccuracies of the tight binding model will naturally be transmitted through the fitting process. However in addition to the direct comparison with the TB data, we have also compared experimental data, results from density functional theory

(DFT) and the potentials by Tersoff (T2,T3) and Stillinger-Weber (SW).

1. Bulk properties

The TB method computes the equilibrium cohesive energy of silicon, E_0 , to be -4.767 eV as compared to -4.629 eV in experiment. The NN potential underestimates E_0 by about 0.4% compared to the TB data. Some bulk properties of silicon are compared in table 2 where a_0 is the equilibrium lattice constant, C' is the distortion constant with its usual meaning, B is the bulk modulus and C_{11} , C_{12} and C_{44} are the elastic constants.

	NN	TB	Expt [13]	DFT [14, 15]	SW [6]	T2 [5]	T3 [9]
E_0 (eV/atom)	-4.762	-4.767	-4.63	-4.67	-4.630	-4.630	-4.630
a_0 (Å)	5.417	5.422	5.429	5.451	5.431	5.431	5.432
B (GPa)	110.1	110.1	99.0	93.0	108.3	98.0	98.0
C' (GPa)	52.5	52.4	51.25	49.0	40.0	17.95	33.5
C_{11} (GPa)	180.1	179.9	167.5	159.0	161.6	121.7	142.5
C_{12} (GPa)	75.1	75.1	65.0	61.0	81.6	85.8	75.4
C_{44} (GPa)	89.3		80.1	85.0	60.3	10.3	69.0

Table 2: A comparison of bulk silicon properties in its equilibrium diamond structure from experiment, the Frauenheim TB method, the NN potential, density functional theory (DFT), Stillinger and Weber (SW) potential and two Tersoff potentials (T2,T3).

The constant C' was overestimated by only about 2% from the experimental value as compared to percentage underestimation of 22% and 34% for the SW and T3 potentials respectively. C_{44} is also much more accurately predicted. The mean relative error obtained from the NN potential with respect to the training data set was 0.8% as compared to 5.5% with respect to experimental values. The mean relative error is the relative error averaged over all monitored properties.

2. Small Si clusters

Figure 4 shows the neural network predictions of the energies for the dimers (figure 4(a)), linear trimers as a function of r_{ij} (figures 4(b) and (c)), linear trimers as a function of r_{jk} (figures 4(d) and (e)) and angled trimers as a function of θ_{123} (figures 4(f)–(h)). The network potential accurately reproduced the dimer energy curve with a root mean square error (RMSE) of 0.038 eV. The calculated properties of

Figure 4: Predictions of E_i for the system of (a) dimers, (b)-(e) linear and (f)-(h) angled trimers using the unweighted limit-screened model. The continuous curves represent the target values from the TB method while the dashed curves represent the neural network predictions.

the equilibrium dimer energy D_e and bond length r_e were 4.397 eV and 1.974 Å. These compared favourably with the TB values. The equilibrium bonding distance r_0 of silicon dimers obtained from the NN potential was underestimated by about 13% compared to the experimental value. This deviation is the same as that in the original training data from the TB model.

An encouraging aspect of the trimer results is that the NN found the isosceles triangle structure with an apex angle of 78.4° as the minimum energy configuration for Si_3 . This is in agreement with ab initio results but different from the empirical potentials which all predict symmetric Si_3 structures and occurs because the tight binding trimer data to which the neural network is fitted also predicts this structure. The binding energy compared to ab initio calculations was however 2 eV lower. Results for 4-atom Si clusters are not given here but these were also encouraging with a small RMSE, albeit larger than that for the trimer cases, for both the angular and bond-length dependency.

3. Predictions from the Network

Although the network was successfully trained and validated, the success of the final model can only be judged by testing the model against unfitted data. For the bulk diamond structure, the potential was not fitted to the shear constant C_{44} . The value of 89.3 GPa shown in table 2 compares reasonably to the experimental value and is a big improvement on all the empirical potential predictions. The performance of the potential was also tested by application to two separate problems. The energy of the bulk Si clathrates Si(34) and Si(46) and point defects in diamond were calculated. Generally the network overestimated energy differences compared to ab initio calculations. The energy difference ΔE per atom from the diamond structure was 0.097 eV for the Si(34) structure compared to 0.055 eV from the ab initio calculations and 0.143 eV compared to 0.069 eV for the Si(46) structure. For the relaxed vacancy structure in bulk diamond the NN gives an energy of 4.78 eV. Ab initio results vary between 3 and 4 eV. For the tetrahedral interstitial the value is 7.55 eV compared to values between 5 and 6 eV and for the bond-centred interstitial 6.92 eV compared to ab initio values between 4 and 5. This is not especially good agreement, but of a similar accuracy to the predictions of the tight binding and empirical potentials.

Figure 5: Predictions of E_i for the bulk systems: (a) diamond, (b) BC8, (c) ST12, (d) BCT5, (e) β -tin and (f) and (g) distorted diamond lattice using the unweighted limit-screened model. The continuous curves represent the target values from the TB method while the dashed curves represent the neural network predictions.

V. Conclusion

We have developed a new methodology based on a NN formulation with variable inputs, for fitting classical potential energy surfaces to quantum mechanical data. The method has been illustrated by application to data generated from Frauenheim tight binding calculations but can be similarly applied to a full DFT approach. The potential has been constructed to be a continuous function of the input parameters and formulae for the derivatives (i.e forces) have been constructed. This should mean easy incorporation into an MD code running at a fraction of the computational cost of a direct tight binding calculation. The methodology has shown that it is possible to predict the properties of bulk Si and small Si clusters within the same framework, an important problem for computer simulations of sputtering. Differences in the predictions of the model between experiment and DFT calculations are reflective of the inaccuracy of the TB approach, to which the NN is trained, rather than a fundamental problem with the methodology. The advantage of the methodology presented here is that no functional form is assumed and so it is not specifically limited to covalent materials. All that is required as inputs is the local geometry surrounding an atom.

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