Structural phase transition of two dimensional single-layer SnTe from artificial neural network

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As machine learning becomes increasingly important in engineering and science, it is inevitable that its techniques will be applied to the investigation of materials, and in particular the structural phase transitions common in ferroelectric materials. Here, we build and train an artificial neural network to accurately predict the energy change associated with atom displacements and use the trained artificial neural network in Monte-Carlo simulations on ferroelectric materials to understand their phase transitions. We apply this approach to two dimensional single-layer SnTe and shows that it can indeed be used to simulate the phase transitions and predict the transition temperature. The artificial neural network, when viewed as a universal mathematical structure, can be readily transferred to the investigation of other ferroelectric materials when training data generated with ab initio methods are available.

Ferroelectric materials, which have spontaneous polarizations that can be reversed by an external electric field, constitute a group of functional materials that are important for many applications, e.g., ultrastable switches, phased-array radar, and dynamic random access memories. It is reported that SnTe thin films with a thickness of 1-unit cell (UC, with two layers of atoms) can have stable spontaneous polarization up to 270K, and 2-UC to 4-UC SnTe films also have strong ferroelectric properties at room temperature. The ferroelectricity of this two-dimensional material makes it a good candidate for applications in promising devices such as high-density memory, nano-sensors. In order to fully understand a ferroelectric material (e.g., SnTe), one needs to know if it indeed experiences a structural phase transition as the temperature decreases, and if so what the transition temperature is. It is also important to know the relation between the transition temperature Tc and other factors, such as strain and the number of layers. Since accurately predicting the phase transition temperature requires large systems, it is usually not possible with a pure ab initio approach, i.e., the density functional theory (DFT). In order to overcome the limitation of DFT with its huge computational cost for large systems, one need to propose empirical formulas to fit the inter-atomic potential energy (or forces) to DFT results with small systems, which is then applied to large systems. For more complex systems such as perovskites, the effective Hamiltonian approach has been a popular method to investigate their static and dynamic properties. For such first-principles-based methods to work, proper empirical formulas or the construction of the effective Hamiltonian (along with the specification of the local mode and the calculation of the coefficients of the effective Hamiltonian), neither of which is trivial if the aim is to achieve a general and transferable approach for different systems.

Recent advances in machine learning (ML) offers an alternative approach for the construction of the potential-energy surface (PES) by fitting large data sets from electronic structure calculations with DFT. The ML potential, when viewed as an universal mathematical structure, has the advantage that it can be used for very different systems with minimal modification. With such an approach, there is no need to reconstruct the formulas or the effective Hamiltonian for different systems, while the accuracy of the PES remains satisfactory. It is promising that the use of ML techniques will combine the advantages of both approaches, i.e., the accuracy of DFT and the efficiency of explicit formulas in obtaining the PES. While the ML approach has been applied to many interesting systems, the structural phase transitions induced by atom displacements, which is important in ferroelectric materials, have not been dealt with adequately. In this paper, it is our goal to develop a framework, which is as general as possible, to treat ferroelectric systems, especially their phase transitions. We note that such structural phase changes usually involve atom displacements within unit cells in crystalline phases, unlike the researches focusing on finding novel structures in liquid and amorphous phases.

In order to construct this framework and show its efficacy, we use the two dimensional (2D) single-layer SnTe system to ground this approach. It is known that SnTe bulk can have a structural phase transition and the SnTe thin films (of 2 layers of atoms) can have a rather high transition temperature (270 K), however, it is unknown if a single-layer SnTe can still have a phase transition when all the atoms are confined to 2D. To address this question, we adopt the ML approach, build artificial neural networks (ANN) suitable for the system with two different types of atoms, and take DFT results as training data for the supervised learning of the ANNs. By considering up to the second nearest neighbors, we have successfully constructed the PES (with respect to the displacement of atoms) and employed it in Monte-Carlo (MC) simulations, which demonstrate a structural phase transition indeed occurs at ~250 K. The approach and the programs we have developed are universal enough that they can be used to investigate different crystals and shed light on their structural phase transitions.

The key ingredient in this approach is to build and train an ANN that can efficiently and accurately predict the energy of a given atomic configuration. A single-layer SnTe has the simple structure as shown in Fig. 1(a) where Sn and Te atoms alternate with each other along both the (100) and (010) directions. To fulfill this mission, the ANN has the topology shown in Fig. 1(b), which is similar to those used by Behler and Parrinello. More specifically, the total energy E of the system is the sum of atomic contributions Ei,
where $E_i$ is the energy imposed on the $i$th atom by its neighboring environment, which will be determined by the ANN we build. It is realized that the potential energy between atoms decays rapidly with distance (which is termed the “nearsightedness”\cite{12,23}), therefore a cutoff function is usually used to limit the interaction between atoms to an appropriate range. For SnTe (and ferroelectric crystals in general), we will limit the interaction up to certain nearest neighbors. In this work, we have found that, limiting to the second nearest neighbors will produce satisfactory results. The input to each ANN in Fig. 1(b) is determined by the coordinates of the $i$th atom and its eight neighboring atoms, which are encircled in Fig. 1(a) where two situations (Sn in the center and Te in the center) are indicated. For the 2D structure, we only consider atom displacements inside the plane, resulting in the input of an a vector with 18 elements for each atom.

In order to properly and adequately represent the local chemical environment around an atom, we find that it is imperative to use atom-centered symmetry functions $G$ as descriptors, which are a series of symmetric functions that derive from the atom position\cite{23}. As indicated in Ref. 12, the number of symmetric functions describing a given structure should be greater than the degrees of freedom of the described system to ensure that all information is fully recorded. In Fig. 1(b), the column named “atomic NNs” contains identical ANNs that takes the chemical environment of an atom, which are encoded in $G$ as shown by the column named “Symmetry functions,” as input and outputs the energy, $E_i$, as indicated by the column named “atom energies”. Finally, $E_i$ is summed to give the total energy $E$.

The core components of the whole structure are the ANNs as shown schematically in Fig. 1(c), which can be built with much freedom. For instance, we can use a simple neural network with back-propagation\cite{24} or some deep neural networks\cite{24}. Here, given the relatively simple chemical environment, we have constructed an ANN with two hidden layers, each layer containing 40 nodes. In addition, since there are two types of atoms (Sn and Te) in the system, two separate ANNs, which have the same structure but different weights inside its nodes, were established to calculate the two types of energies, $E_{Sn}$ and $E_{Te}$ imposing on Sn and Te atoms, respectively.

We use supervised learning as implemented in PyTorch\cite{25} to train the proposed ANNs, where the DFT calculations are employed to obtain the training data set. Based on the 2D single-layer SnTe (the lattice constant $a_0 = 6.1836\text{Å}$, with a vacuum layer of $4a_0$ set in the $z$ direction.) shown in Fig. 1(a), \textit{ab initio} molecular dynamics simulation with GPAW\cite{26} is used to simulate a $2 \times 2$ system (16 atoms) from 500 K to 0 K in order to generate training samples. In these simulations, GPAW uses plane-waves with a cutoff energy of 750 eV, a $2 \times 2 \times 1$ Brillouin-zone sampling grid\cite{27} and the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional\cite{28}. A total of about 4000 configurations are calculated with GPAW,
of which 3900 was used to optimize the ANN, and 100 was used as a preliminary test of the predictive ability of the ANN. For each of the configuration, its energy is calculated and the energy of the original configuration, where none of the atoms is displaced, is used as the energy reference, \( E_0 \). Figure 2 compares the values predicted by the ANN and by GPAW, which shows a good agreement where with the maximum difference is within 6 meV per atom. We note that the energies of the chosen configurations used in Fig. 2 are all higher than \( E_0 \). The prediction by the ANNs for configurations with lower energies are even better as we discuss below (see Fig. 3).

To further verify the accuracy of the ANN, we have also generated special configurations where all the Te atoms are synchronously displaced along a particular direction to a distance \( d \) as shown in Fig. 3(a). We use the trained ANN to predict the energies of these configurations and generate the PES of SnTe as shown in Fig. 3(b). It is interesting to see that the ANN has generated a smooth PES with multiple local energy minimums. To quantitatively check the predictions of the ANN, we have sampled along the \( \langle 110 \rangle \) and the \( \langle 100 \rangle \) directions and compared the energy predicted by the ANN to those calculated with GPAW as shown in Figs. 3(c) and (d). For the region of interest (\( d \leq 0.4 \) Å), the accuracy provided by the ANN is stunning. Moreover, it is worth noting that the ANN has successfully reproduced the double-well potential which is a critical indication of possible structural phase transitions.¹⁵ This feat is remarkable when we recall that the ANN has a universal internal structure and is trained with configurations having essentially random atom displacements. While Figs. 3(c) and (d) show that the ANN can fit the PES very well along \( \langle 100 \rangle \) and the \( \langle 110 \rangle \) directions, one needs to keep in mind that the power of the ANN lies in their ability to predict the energy of any configuration, and Figs. 3(b–d) just show some special cases.

Having constructed and trained the ANN that can accurately generate the PES, we can now employ it in MC simulations as an energy calculator to investigate the structural phase transition of SnTe. More specifically, in MC when an atom is displaced, we use the ANN to predict how much energy change will arise. For the MC simulations, we set up a 2D \( 6 \times 6 \) supercell (144 atoms), and gradually cool down the system from 600 K to 20 K with a step of 20 K. At each temperature, we sweep the systems 80,000 times, all atoms are displaced randomly and the move may or may not be accepted depending on the incurred change of energy. We note that in order to mimic the epitaxially grown SnTe structure, the Sn atom ion the corner of each unit cell (there are two Sn atoms in each unit cell) is kept at its original position (i.e., no attempt to displace it is made) so that the crystal lattice is maintained. The MC simulation uses our home brewed PyMC², which is a modular Monte-Carlo simulation program specially designed for crystals.¹⁰

Figure 4 shows the temperature evolution of the average relative displacement \( \Delta d \), which is determined by the displacements of Sn and Te of the same unit cell, i.e.,

\[
\Delta d = \frac{1}{N} \sum_{i=1}^{N} \langle |d_{Te} - d_{Sn}| \rangle ,
\]

where \( \langle \ldots \rangle \) indicates the supercell average, and \( N \) is the number of MC sweeps chosen to use for the final average. As we can see from Fig. 4(a) a phase transition occurs at around 250 K, where the average displacement starts to steadily increase with the decreasing temperature, gradually reaching a displacement of about 0.26 Å along the \( \langle 110 \rangle \) direction, which is consistent with the energy minimum at \( d = 0.26 \) Å as shown in Fig. 3(c). Figure 4(a) shows that the displacement of Te is large while Sn remains mostly still as the corner Sn atoms are used to fix the crystal lattice. Our simulation results indicate that the epitaxially strained 2D single-layer SnTe has a comparable phase transition temperature as the 1UC SnTe that are previously investigated both experimentally and theoretically.¹¹

In summary, we have used ML techniques to build ANNs that are employed in MC simulations to investigate the structural phase transitions of ferroelectric single-layer SnTe,
which is still a very challenging task for pure ab initio computation. Unlike other approaches, no concrete model or formulas are necessary approximate the PES, which often requires a priori knowledge or a good understanding of the given system. The ML approach also removed the difficulty in determining the coefficients appearing in the equations used to describe the PES of a given model, essentially achieving a model-less approach. The ANNs work as a special universal mathematical structure and are capable for various systems as long as the training data from ab initio computation are available. Such virtues make this approach very general and flexible to investigate the ferroelectric phase transitions.

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