The Many-Body Expansion Combined with Neural Networks.
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Fragmentation methods such as the many-body expansion (MBE) are a common strategy to model large systems by partitioning energies into a hierarchy of decreasingly significant contributions. The number of fragments required for chemical accuracy is still prohibitively expensive for ab-initio MBE to compete with force field approximations for applications beyond single-point energies. Alongside the MBE, empirical models of ab-initio potential energy surfaces have improved, especially non-linear models based on neural networks (NN) which can reproduce ab-initio potential energy surfaces rapidly and accurately. Although they are fast, NNs suffer from their own curse of dimensionality; they must be trained on a representative sample of chemical space. In this paper we examine the synergy of the MBE and NN’s, and explore their complementarity. The MBE offers a systematic way to treat systems of arbitrary size and intelligently sample chemical space. NN’s reduce, by a factor in excess of $10^6$ the computational overhead of the MBE and reproduce the accuracy of ab-initio calculations without specialized force fields. We show they are remarkably general, providing comparable accuracy with drastically different chemical embeddings. To assess this we test a new chemical embedding which can be inverted to predict molecules with desired properties.

Keywords: many-body expansion, neural networks, methanol

I. INTRODUCTION

The many-body expansion (MBE) lies at the heart of a multitude of computational methods being developed in the realm of ab-initio theory and force fields. In insulators the high-order terms of the MBE decay rapidly with distance, which makes this type of approximation useful for low-scaling, high-accuracy models of liquids, solids and biological molecules. However an ab-initio MBE is orders of magnitude more costly than a classical force field. The main limitation comes from the combinatorial growth of effort at each order.

In chemistry, neural networks are growing in popularity to predict molecular properties. However NN’s have their own limitations: their input must have a constant shape, moreover they must be trained on a representative number of samples, and chemical space grows exponentially with molecular size. This curse of dimensionality in the training set is the main barrier to the creation of a universal NN force field with very high accuracy. The purpose of this paper is to show that the MBE provides a very natural and accurate way to alleviate this curse of dimensionality while retaining the generality, accuracy and efficiency of a NN.

Force fields based on the many-body expansion are growing in popularity. Under the MBE scheme, the total energy of an system can be expanded as the sum of the many-body terms. High order terms are more costly calculations and the error of the MBE is often balanced with the error of the underlying model chemistry at third order. An electrostatically embedded MBE (EE-MBE) has also been proposed as a means to improve the accuracy. Others have suggested a many-body expansion scheme of overlapping-fragments as a way to improve upon the accuracy of the energies. Statistical models from machine learning are becoming popular chemical models. Examples include fitted potential energy surfaces with atom-centered symmetry functions and with permutation invariant polynomial. Permutationally invariant polynomials have been used to express the many-body energies of water clusters and water-methane clusters with great success. Also, machine learning has been used to predict properties, such as atomization energies, HOMO and LUMO eigenvalues, ionization potentials, force constants, dielectric constants, quantum transport coefficients and nuclear magnetic resonance parameters. To our knowledge, there are few works that combine neural networks with the MBE and those have focused on elemental solids. The closest work predicted the many-body energy of Si$_n$($n = 1, 2, \ldots, 7$) clusters. Bartók used machine learning techniques based on Bayesian inference to correct DFT one-body and two-body energies for water. In this paper we learn the many-body energies of condensed phase liquid methanol within $mE_h$ accuracy. We show that one can use the MBE for methanol clusters of a thousand molecules without significant computational expense on typical GPU workstations. We also present a novel chemical embedding, which has the advantage that it is invertible to ball-and-stick geometries, asses it as a descriptor to learn the MBE, and propose it as a useful tool for inverse-design.
Studies have shown that the MBE converges rapidly for van der Waals and water clusters.\cite{25,27,28,65} Convergence is relatively slow for metallic or covalent interactions, although schemes have been proposed to improve the accuracy of the MBE on covalent systems.\cite{65} We chose methanol for its strong hydrogen bonding, but nothing about this work is specialized or limited to systems of this size. RI-MP2 with the cc-pVTZ basis is used to calculate all of the training and testing data for the many-body energies. The integral precision and SCF convergence criteria were as tight as possible and BSSE using the K-mer centered basis set approach ($k = CBS$)\cite{28} was applied. Training and test geometries are drawn from an AMBER molecular dynamics trajectory\cite{68,69} of 108 methanol molecules at 330 K and ab-initio trajectory of three methanols at 500 K. The total data set include 844,800 samples one-body energies, 74,240 samples for two-body energies and 36,864 samples for three-body energies. 20% of the total data set is used for testing. All of the ab-initio calculations are done with the $Q - Chem$ package.\cite{68} Previous studies have shown that cumulative two-body energies and cumulative three-body energies converged at a cutoff of 10 Å for ($H_2O_2$).\cite{27,28} We also found that both the two-body and three-body energies negligibly different from limiting formulas at a cutoff of 10 Å as shown in Figure SI-1 so our dimers and trimers were generated with this cutoff of 10 Å. $Cuda \rightarrow Convne$\cite{69} was used to train and evaluate the neural network.

Choosing the chemical embedding for the system as the input to the NN has a great effect on performance. Many different chemical descriptors have been proposed, including the Coulomb matrix,\cite{70,71} symmetry functions,\cite{72,73} bispectrum,\cite{74,75} permutation invariant polynomials,\cite{76,77} metric fingerprint,\cite{78,79} and the radial distribution Fourier series\cite{80} which is based on the electronic density and is similar to a descriptor our group has used in the past for learning kinetic functionals.\cite{81} Systematic comparison of different descriptors is beyond the scope of this paper and we choose the Coulomb matrix (CM) as our input for neural networks for its simplicity and we show that it is capable of the task. The CM, however, is not permutationally invariant, therefore, in this study we augmented our training data with all the permutations of hydrogen atoms on carbon and all the permutations of methanol molecules in the dimer and trimer to learn the permutation invariance. Similar data augmentation techniques have been widely used in image recognition to achieve translation and rotation invariance.\cite{82,83} As shown in the Figure SI-2 and Figure SI-3, the permutation invariance is learned with satisfying accuracy. The permutation invariance can also be avoided by averaging the result of all the possible permutations.

We experimented with a novel chemical embedding, which we call the depth map (D-map). The purpose of this descriptor is not to improve over the accuracy of the CM, but rather to have an input which provides reasonable energies and inverts directly to molecular geometries. If networks could accurately learn from an invertible input, they could also become useful tools for molecular inverse-design. Similar types of NN inputs have been used in the area of 3D detection and object recognition.\cite{84,85} An example D-map can be seen in Figure 1. It is simply a depth of field image of a ball-and-stick structure. A simple routine to calculate one is given in the supplement. Given the usefulness of this input in molecular design we were curious how well it could be used to predict energies, and will compare it to the CM in the results. Generative Adversarial Networks (GANs) have since been studied extensively for their ability to hallucinate authentic looking images.\cite{86,87} We trained a GAN on the D-map to produce hallucinated images of methanols, and discuss the utility this provides.

### III. RESULTS

Figure 2 shows the comparison of the one-body, two-body and three-body energies calculated using MP2 and our neural network for all the test samples. The neural networks give close agreement with MP2 such that errors in the underlying model chemistry would be the limiting factor of MBE-NN. The Mean Absolute Error (MAE) and Mean Signed Error (MSE) of the energy of each many-body terms over a test set.
TABLE I. The MAE and MSE (microhartree) of one-body energy, two-body energy, three-body energy with Coulomb matrix input and three-body energy with depth map input predicted by neural network. We calculate a rate of error as MAE(microhartree)/wall-hours of RI-MP2 in Q-Chem required to generate the training data.

| Error  | 1-body | 2-body | 3-body (CM) | 3-body (DM) |
|--------|--------|--------|-------------|-------------|
| MSE    | 0.24   | 0.90   | -1.16       | -3.96       |
| MAE    | 5.99   | 15.6   | 20.0        | 39.0        |
| Rate   | 0.005  | 0.008  | 0.005       | 0.009       |

The MSE is balanced in the sense that each order of the expansion adds comparable microhartree errors to a total energy, as we will discuss in results which follow.

The higher MAE of the higher-body terms is a predictable consequence of our design principle that the ratio of total error to wall-hours should be kept roughly constant at each order of expansion. The three-body energy is a surface of much higher dimension than the two body energy, and this causes difficulty in three ways: a need for more training data, a larger number of invariances, and network capacity. However, we use much less three body data, because it is more expensive and less significant. The histogram of errors of all the many-body terms are also shown in Figure 2 and appears uncorrelated which is supported by our later observation that the error per-methanol is essentially insensitive to system size.

The three-body network trained on the D-map also provides reasonable energies. Comparing the three-body energy plots for the CM and the D-map, in most cases the D-map appears to do nearly as well as the CM; however, for a handful of cases the D-map makes significantly poorer predictions. We note this tends to happen when all or part of an oxygen atom becomes eclipsed by the methyl group. Furthermore, we also note the D-map tends to make poorer predictions for energies which are near zero. The distribution of errors remains normally distributed. The D-map does provide a few advantages over the CM: It provides a low dimensional encoding of the space of methanol geometries. It also has constant shape regardless of chemical input, and suffers from fewer problems with invariances.

As expected the CM makes more accurate predictions than the D-map; however, our aim with the D-map was to provide a chemical embedding which can easily be mapped back to the original geometry of the system. For example this can be used to predict molecules, which maximize a desired property directly without searching chemical space. To this end we then trained a GAN, based on Radford et al. using the D-map by separating element types into separate color channels, to produce hallucinated images of methanol trimers. An example hallucinated D-map can be seen in the bottom panel of Figure 1. The network maps a random z-vector back to a D-map. By varying one element of the z-vector, we were able to control image generation to tune properties. The examples in Figures 1 are from the same z-vector with one varied element $z_i$, which rotates one of the methanol end over end. It is easy to imagine extending this to other properties and using a GAN for inverse-design. For the remainder of this paper we employ the Coulomb embedding for the MBE.

Table II gives the relative energies (mE\textsubscript{h}) of three minimal energy geometries of a methanol trimer.

| Geometry | MP2 | MBE-NN | HF* | B3LYP* |
|----------|-----|--------|-----|--------|
| chair    | 0   | 0      | 0   | 0      |
| bowl     | 1.50| 1.40   | 1.75| 2.02   |
| chain    | 4.54| 4.10   | 2.56| 5.29   |

\textsuperscript{a} MP2, HF and B3LYP energies are extrapolated to a complete basis.
cases in this region are sampled by the MD trajectories.

The relative energies of five random (MeOH)_{20} clusters are shown in Figure 4 to assess the errors due to NN and MBE. Compared with the real MP2 energies, the MAE of the MBE using MP2 (MP2-MBE) is 0.12 mE_h per molecule, and with NN-MBE 0.10 mE_h. Remarkably there is no degradation of accuracy involved in using NN-MBE, despite massive speedup. Instead the method is limited by the quality of the model chemistry it is built on and the accuracy of the MBE itself.

Proper treatment of solvent effects is crucial for describing most chemical processes. The top panel of Figure 5 shows the energy change of breaking a hydrogen bond between two methanols when the solvation shell is not included. MP2-MBE predicts the energy change to be 5.6 kcal/mol and NN-MBE gives 5.8 kcal/mol. When the solvation shell (with a radius of 10 Å) is included, as shown in the bottom panel, the energy change dramatically increased to 13.3 kcal/mol, which shows the large influence of solvent effects. The NN-MBE predicts the energy change with solvation shell to be 14.6 kcal/mol, 1.3 kcal/mol larger than the MP2-MBE result. Considering the speed up of the NN-MBE, discussed below, and its accuracy, the scheme shows promise for condensed phase phenomena.

We also investigated the error of NN-MBE as a function of system size. Figure 6 shows the error per molecule and the error per cluster of the NN-MBE (with respect to MP2-MBE) with an increasing number of molecules in the cluster. The error per cluster stays within the range of 3 mE_h and the error per molecule reaches a maximum at 70 units and shows signs of sub-extensive behavior. Figure SI-5 provides the total wall time comparison of the NN-MBE and MP2-MBE, showing that the NN-MBE offers a speed up of more than two million relative to MP2-MBE without any type of optimization.

IV. DISCUSSION AND CONCLUSIONS

We have shown that a NN-MBE can be used to calculate the energy of methanol clusters with a speed up in the millions with respect to the MP2-MBE. The error of the NN-MBE is within mE_h/s, which is similar to the error of MP2-MBE with expansion up to three-body terms. The histogram of the errors of the NN-MBE display Gaussian shape, which makes the error per molecule decrease with the increase of system size. The satisfying accuracy and huge speed up enable the NN-MBE to treat large system with ab-inito accuracy, which would otherwise be impossible, such as treating solvation shell effects in ab-inito calculations. The Coulomb matrix is not in-
variant to permutations and even though we have shown that permutation invariance can be learned by augmenting the training samples with all of the possible permutations, it is still not perfectly invariant. Our current study focused on methanol and this scheme can easily be generalized to other systems. We introduced a new descriptor, the D-map, which is invertible with the geometry of a system. The D-map was able to predict the three-body energies reasonably well, and provides several advantages of its own. We then showed that a generative adversarial network could be trained on the D-map to provide hallucinated images, which are tunable and should be useful for inverse molecular design.

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