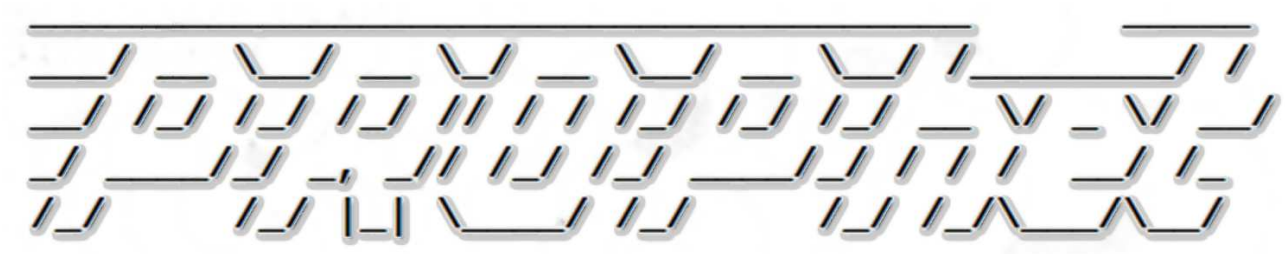

# Documentation

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## Contents

# Part I

# Introduction

PROPhet (short for PROPerty Prophet) uses machine learning techniques to find predictive connections between a chosen set of material or chemical properties and some desired target property. The idea is to facilitate the prediction of properties which are difficult or expensive to compute (e.g. optical or thermal properties) from those that are either already known (e.g. structure) or relatively easy to calculate (e.g. charge density). The user provides PROPhet with a set of training systems for which both the inputs and outputs are known, and PROPhet determines a mapping from input to output. The resulting model can be used to predict properties of new structures or systems not included in the fit.

One obvious use of PROPhet is to create atomistic potentials for various systems of interest. These potentials can be trained on relatively small systems directly from first principles data, without requiring any knowledge of the functional form. Once trained, they can be used to run molecular dynamics (MD) simulations on larger systems with a potential energy surface near the accuracy of DFT, but orders of magnitude faster. Using PROPhet potentials, one can compute thermal properties or long-time dynamical properties of complex systems. PROPhet comes with a built-in interface to the popular MD code LAMMPS, which allows PROPhet potentials to be used seamlessly in any LAMMPS run.

Another intended use of PROPhet is to facilitate the computation of difficult to compute properties (e.g. accurate bandgap, optical properties, response properties). Again, training can be conducted on small systems within the reach of first principles methods, and the resulting model used to predict the properties on large complex systems such as interfaces and heterostructures.

PROPhet is fully parallelized using MPI. Training examples are split over cores so that, in principle, one can use as many cores as there are training examples. PROPhet has been tested on up to 500 cores, and exhibits excellent scaling. There are also tons of tricks and algorithmic tweaks to make PROPhet as fast as possible.

# Part II

# Installation

Compilation of PROPhet uses the standard linux "configure $\rightarrow$ make $\rightarrow$ make install" chain. There are a few different options depending on your situation.

## MPI based compile

```
> ./configure [options]
> make
> make install
```

## Serial compile

```
> ./configure [options] –without-mpi
> make
> make install
```

## Linking with the LAMMPS[1] MD package

To use PROPhet potentials in LAMMPS, you must create a static library and link this into your LAMMPS install. The linking is done through the standard LAMMPS package system. See section 1.6 of this documentation for instructions on using PROPhet potentials in LAMMPS. Note that you will need the source package of LAMMPS to link with PROPhet.

In the following, we take PROPhet_DIR to be the src directory of your PROPhet installation and LAMMPS_DIR to be the src directory of your LAMMPS installation.

## Automatic Install

```
> ./configure [options] –enable-lammps=LAMMPS_DIR
> make
> make install
> relink your LAMMPS executable
```

**Note that this will alter the LAMMPS_DIR/Makefile.package file** to tell LAMMPS where to find the necessary PROPhet files. A backup of this file is saved to Makefile.package.backup to allow you to undo the changes. If you do not want this file to be overwritten, you may manually link PROPhet with your LAMMPS installation as described in the next section.

## Manually linking PROPhet to LAMMPS

If the automatic install of the previous section fails, there are a couple options. The PROPhet Makefile needs to have write access to the LAMMPS_DIR so that files can be copied and altered. If you do not have write access to this directory, you may need to either run "make install" as root, or change the permissions of LAMMPS_DIR.

Alternatively, you can manually install the PROPhet library into your LAMMPS installation. To do this you must:

1. copy the file "pair_nn.h" to LAMMPS_DIR (makes LAMMPS aware of PROPhet)
2. Edit LAMMPS_DIR/Makefile.package (tells LAMMPS where to find the necessary files)
   (a) add -IPROPhet_DIR to PKG_INC
   (b) add -LPROPhet_DIR to PKG_PATH
   (c) add -lPROPhet_lammps to PKG_LIB
3. (Re)-link LAMMPS

# Part III

# Quick start / tutorial

## 1 Example : Building a potential (structure $\rightarrow$ energy)

### 1.1 Introduction

One of the capabilities of the PROPhet code is to train a network using the atomic structure as input, following the approach of Behler[2–4]. In the special case that one has trained a network which maps structure to energy (a potential), the resulting fit can be used within the LAMMPS MD code by way of an interface provided with PROPhet. This interface allows near DFT quality potentials to be used in MD runs to compute a host of important material properties. This tutorial will guide the user through the creation, validation, and use of a potential.

In this tutorial you will learn how to perform the following:

- Setup a PROPhet Input file
- Train a network on Structure to Total Energy
- Validate the trained network
- Predict new structures
- Set up a LAMMPS run to use this potential

The data required for this tutorial are provided in the doc/tutorial/data directory. It should be noted that the potential we will create here is for instruction purposes only, and is not of production quality, as the input data and network quality are extremely limited. The data used for this tutorial, consisting of VASP total energy calculations for 300 structures of diamond-structured carbon, are stored in doc/tutorial/data.

## 1.2 Setting up the PROPhet input file

PROPhet uses a single required input file (by default called “input_file”), which holds all the parameters for the run. PROPhet will expect this file in the directory in which it runs. A different file name can be specified on the command line with the -in FILE flag. Throughout this tutorial, we will be using the term input_file as a generic reference to whatever file is specified for the main input. A fully annotated input_file is supplied with the distribution of PROPhet (in doc/input_file).

In this tutorial, we will create a potential mapping structure to total energy. Within the doc/tutorial directory are a number of files, 3 of which are called “input_train”, “input_validate”, and “input_predict”. These are the main PROPhet input files for each stage of the tutorial. It is generally not necessary to split different phases of the calculations into different input files, as only minor changes are needed depending on the run. The files are split here only for convenience.

If you examine the input_train file you will find the following:

input file

```
# Network details
# ————————————————————————
network_type = nn                    # Perform a fit using a neural
network hidden = 20                  # 1 hidden layer of 20 nodes
input = structure                    # Use the atomic configuration as input
output = energy                      # Use total energy as output
transfer = tanh                      # Use a tanh transfer funtion for all nodes

# Specify the geometry mapping functions
# ————————————————————————
Nradial = 5                          # Number of pairwise mapping functions
Nangular = 4                         # Number of 3-body mapping functions
Rcut = 4                             # Cutoff for the mapping functions

# Training details
# ————————————————————————-
algorithm = rprop                    # Use the default Rprop training algorithm
Niterations = 300                    # Stop training after 300 iterations
threshold = 0                        # Don't stop on the basis of the gradient
Nprint = 10                          # Print fitting information every 10 steps
#checkpoint_in = potential           # We're not restarting so no need to read a potential
checkpoint_out = potential           # Save the potential using the base name "potential"
Ncheckpoint = 100                    # Save the potential every 100 steps

# specify the data
# ————————————————————————
include training_data                # Data are specified in the "training_data" file
```

You will notice this input file is broken into four primary areas: "Network Details", "Geometry Mapping", "Training Details" and "Data Specification". Note that the grouping is for readability and is not required. With the exception of the “data” statement (described in data) the input file can be in any order. Also, the input file is case insensitive and whitespace is ignored.

The Network Details section defines the type of machine learning we will be using. Currently only neural networks (nn) are implemented. “hidden” specifies the topology of the network. We are using a single layer of 20 nodes here to make a quick fit. More layers can be specified with additional integers, each one giving the number of nodes in that layer (for example, “hidden = 35 35” would give 2 layers, each with 35 nodes). “input” and “output” define the properties that the network will be trained to, here we are inputting ’structure’ to the network and the output is ’energy.’ Finally, “transfer” is the type of transfer function in the nodes. We are using hyperbolic tangent functions (tanh).

The Geometry Mapping section defines the key values for generation of the mappings functions (see Section Mapping function specification and Reference[4] for details). In this case “Nradial” and “Nangular” define 5 and 4 functions, respectively, to be generated internally by the code. Care must generally be taken here, since, in multi-species systems, the number of radial and angular functions grows rapidly due to the interaction differences between the different species. As an example, if we were training $CO_2$ data, Nradial = 5 would actually generate 10 different 2-body functions per atom in the system. These would correspond to X-O and X-C interactions (X being the central atom type) for each of the 5 functions requested. In the case of Nangular = 5, this would swell to 15 as the X-C-C, X-C-O, and X-O-O interactions have to all be treated separately. Finally, “Rcut” is the cutoff radius (in \AA) for the mapping functions.

The Training Details section describes how the network is converged. The main point here to comment on is the “checkpoint_in” and “checkpoint_out” lines. When performing a training run with PROPhet, the file “checkpoint_out” will be written every “Ncheckpoint” iterations. This allows for the option to restart the calculation if it gets interrupted before either “Niterations” or “threshold” are met. If a restart (or a validation, to be describe later) is desired, un-commenting “checkpoint_in” allows for the network to be reread and restarted.

Finally, in the Data Specification section, we “include” a file specifying the training data

into the input_file. This is to increase readability as data specification is just a list of directory locations. While these locations can be specified directly in the input_file, it is easier to read if you place them in a separate file. The "include" flag in PROPhet works similarly to the same flag in C/C++, placing the contents of the included file at the spot where "include" was invoked. For example, this include file training_data looks like the following:

training_data

```
data code=vasp
./data/1
./data/2
./data/3
./data/4
   ⋮
```

The "code" flag specifies the code that was used to generate the data. Currently VASP, FHIAims, and Quantum Espresso (QE) can all be specified here. With this information, PROPhet can read many desired properties directly from the ab initio output files. It is worth noting that for qe, the energy is not stored in standardized output files. Therefore, PROPhet cannot read the energy directly from a QE run and this must be specified with the "user_input" mechanism (see The user_input file).

Now that a basic understanding of the input_file has been described (take a look a the fully annotated file in the distribution and Section 4 for other options), we are ready to train the network.

## 1.3 Training

Examine the input_train and the training_data files. The former contains the parameters for this training run and the latter contains the data we will use for training. You will notice that we are only including 250 structures in this training set. The other 50 are set aside

to check that the trained potential can predict new structures not included in the training. This will be performed in the Predict section.

To train the network, you simply specify the "-train" command line flag, as:

```
> mpirun PROPhet -in input_train -train
```

Note that "-train" is the default behavior, so the flag can actually be left off.

PROPhet will run for 300 iterations (as is set in the input_file). (Again, note that a production network will need to be trained with many more data points and iterations.) The current error of the network is given in the second column of the output. The units of this quantity are the same as the units of the target values you provided to PROPhet, in this case, since we are using VASP data, the units are eV. At the completion of the run you should see a new file called potential_C, which holds the parameters and details of the current fit. The name of this file is specified in the "checkpoint_out" parameter in the input_file. Note that the method PROPhet uses to fit structures creates a separate network for each atomic species present. Thus, the name specified in "checkpoint_out" is the base name of the files, and PROPhet will automatically append the atomic symbol to this name, creating a separate file for each atom type and storing the appropriate parameters in each. Here we have only 1 type, so PROPhet has created only 1 checkpoint file. The checkpoint file (which is written every Ncheckpoint training steps) is used to restart an interrupted run, or to use the network for predictions/MD after it has been created. Section 6 discusses the format of this file.

We now have a PROPhet potential, ready to be tested and used.

## 1.4 Validate

The first step in making sure that you have a working network is to validate it. A validation simply 'fires' the network with a given set of inputs and performs a minimal analysis. This helps verify that the network is a faithful representation of the data it was trained on. PROPhet will generate some simple measures of the fidelity of the fit.

Take a look at input_validate and training_data files. The only difference between the input_train and input_validate files is that the "checkpoint_in" line is un-commented in the

latter. When you run with a “-validate” flag PROPhet will read the already trained network from the file specified in “checkpoint_in” and then ’fire’ it against the data you provide it. As with the “checkpoint_out” flag, “checkpoint_in” should specify only the base name of the potential, and PROPhet will automatically handle the atomic species suffixes it added.

To run this manually simply add the “-validate” command line flag as:

```
> mpirun PROPhet -in input_validate -validate
```

This will fire the network with the 250 structures that the network was trained with. PROPhet will print the predicted and known energies of each system, along with a simple analysis of the fit. In this case, the end of the output should look something like:

STDOUT

```
Minimum Error = 0.00137437
Maximum Error = 1.55498
RMS Error = 0.543206
```

Notice that the RMS error is identical to the fitting error that we finished with during our training run. The minimum and maximum errors fix the bounds of the distribution. Below these statistics are the results of an (approximate) Kolmogorov-Smirnov test. This is a (very rough) measure of how likely the set of target values (the ones provided as the “correct” answers) and the network outputs are to coming from the same overall distribution. The test gives a p-value for accepting the hypothesis that the sets come from the same distribution. In this case, the results of test show:

STDOUT

```
– Approximate Kolmogorov-Smirnov test –
D = 0.032
p value = 0.999543
```

**This test is meant only as a guide to compare different fits, and the results should not be taken to be quantitative measures of fitness.**

## 1.5 Predict

The power of PROPhet is the ability to predict properties of interest for systems that were not used in fitting. By setting aside the 50 structures in the beginning of the training, we can now test the network on those 50 structures. In other words, we want to 'fire' the network while feeding in these unknown structures.

Take a look at input_predict and training_data. You will notice that input_predict is identical to that in the validation example except for the data included. In "predict_data" you will only find 50 data points, none of which were included in the fit. To test the predictive power of the model, you can run with the same "-validate" flag as the previous section on data not included in the fit.

```
> mpirun PROPhet -in input_predict -validate
```

When this completes, you should find something like the following output:

STDOUT

```
Minimum Error = -0.0146453
Maximum Error = -1.38611
RMS Error = 0.566501
```

Note that the "-validate" flag only works if we have known target values for the property we're predicting, that is, if we've already done the ab initio calculations to compare to. To fully predict the property, you can use the "-run" command line flag instead of "-validate". This will obviously not produce the error statistics, but will instead just print out the predicted energies for each structure.

## 1.6 LAMMPS

Once you have a potential, you probably want to do something with it. PROPhet includes an interface to the popular MD code LAMMPS[5], so you can use PROPhet potentials in sophisticated MD runs. Using PROPhet potentials within LAMMPS is fairly straightforward. (Note that you will need to use a LAMMPS executable that has had PROPhet linked in. This process is described in section II. If you run the following without a properly compiled LAMMPS executable, the code will simply die at the potential setting step.) The key lines of a LAMMPS input file are as follows:

LAMMPS input file

```
pair_style nn Rcut
pair_coeff 1 potential_C
```

☜ make sure Rcut is set to the same value that it was when the potential was trained

where the first line tells LAMMPS to use a PROPhet potential (nn) with a particular cutoff radius (Rcut), and the second line tells LAMMPS in which file the parameters for atom type 1 are to be found. If your system has multiple atomic species, you will need a separate "pair_coeff" line for each type. Note that, while a PROPhet potential is not strictly a pairwise potential, it uses the LAMMPS "pair" class since it depends only on the coordinates of neighboring atoms.

**There are 2 critical things to keep in mind when using PROPhet potentials in LAMMPS:**

- ✴ The Rcut specified must be the same as was used in training. Otherwise, the difference in the number of neighbors will cause the run to be unstable, as the mapping functions will be taking on systematically different values from those they saw during training.
- ✴ The units command used in LAMMPS should correspond to the units used in training. PROPhet does not have internal units, but simply fits the structure (in whatever units you gave it) to the energy (in whatever units you gave it). The units will gener-

ally depend on which code generated the data, unless units were converted explicitly before training.

## 1.7 Further information

This is the basic flow of using PROPhet to fit system properties. This tutorial has given an example of fitting atomic structure to the total energy, but there are many other properties that PROPhet can extract automatically from data files (see section 4.3 for a list). In addition, since the property of interest may not be in this list, PROPhet provides a mechanism by which the user can provide their own property data. This mechanism is discussed in section 3. Note that, because of a complication in the way Quantum Espresso stores information in its files, PROPhet cannot automatically extract the total energy from a QE run. If you want to use the energy from a QE run in a PROPhet fit, you must use this mechanism.

# Part IV

# Reference guide

This section is intended to be a reference for the input and command line parameters needed to run PROPhet effectively.

## 2 command line flags

Most control parameters should be specified in the input file. Command line flags are used to tell PROPhet where to find the input file, and what to do with what it finds there. For easy reference, Table 1 lists the possible command line flags. If no command line flags are specified, PROPhet will look for a file called input_file and perform training on what it finds there. The name of the input file can be specified with the "-in" flag. Actions other than training can be performed with the other flags shown in Table 1.

Table 1: Command line flags accepted by PROPhet

| Flag | argument | description | default |
|---|---|---|---|
| -in | filename | sets the input file to read | input_file |
| -train | — | train on the input set (default) | ☑ |
| -run | — | predict the input set | ☐ |
| -validate | — | analyze the current fit | ☐ |

The three flags "-train", "-run", and "-validate" are action flags that tell PROPhet what task to perform. It does not make sense to specify more than one of these flags.

### 2.1 -train

This flag, which is the default, instructs PROPhet to perform a fit given the data in the input file.

## 2.2 -run

This flag instructs PROPhet to perform predictions on the data using a pre-existing fit. Note that that “checkpoint_in”, which specifies the existing network, must be specified in the input file (see Section 4).

## 2.3 -validate

This flag tells PROPhet to perform some analysis of a pre-existing fit. Generally, this should be done with the fitting data after the fit has been performed. PROPhet will print out all the network outputs and their associated targets, as well as some statistics regarding the fit. Included is an approximate Kolmogorov-Smirnov statistic, which indicates the likelihood that two distributions are the same. In this case, the two distributions are the set of network outputs and the set of targets. If these are deemed significantly different, it is likely the fit needs to be improved. This statistic is advantageous as it makes no assumptions about the form of the distributions, and is robust to shape and scale changes.

# 3 Specifying data

The most important part of using PROPhet is specifying the data set for training or predictions (we will call these the training examples). For many properties, PROPhet can extract the data automatically from the output files of several codes (currently VASP, Quantum Espresso, and FHI-aims). For VASP and Quantum Espresso, the data are extracted from the xml files they write, while the data are pulled from a plain text output file for FHI-Aims.

## Organizing the actual data

There should be a separate base directory for each different training example (generally a training example will correspond to a particular structure). If multiple properties are being used as inputs/output, they can come from a single run or several different runs. If the

runs are separate, the data for each property should be stored in subdirectories of the base directory for that training example. These can be named as desired, we will tell PROPhet how to find the data next.

## Specifying data to PROPhet

At its simplest, specifying data just requires one to provide a list of the base directories for the training examples, each on a separate line, as in

input file

```
absolute_path_to_training_example_1
absolute_path_to_training_example_2
⋮
absolute_path_to_training_example_N
```

This list can be placed directly in the input file or in a separate file which is inserted into the input file with an "include" statement (see Section 4.22). The code that generated the data, along with the directories associated with different properties are set by the "data" input file parameter. This parameter affects all examples that come after it (including those in any files inserted using the "include" statement) until the next "data" statement. The "data" statement should specify which code is being used either with "code = vasp" or "code = qe". The subdirectories corresponding to each property are specified within the "data" statement by "property=subdirectory" statements, where "property" is one of the set given in Section 4.3 and "subdirectory" is the *relative* path from the base directory for that training example to the directory containing the property of interest. Only calculations of properties residing in directories other than the base directory for each training example need to be specified in this way.

As an example, suppose you are using VASP, and have calculated the charge density on a fine grid, and the total energy using a coarser grid. You might store the energy for training example I in /home/mydir/data/I, and the charge density in /home/mydir/data/I/Chg_density.

This situation can be specified with

input file

```
data code=vasp density=Chg_density
/home/mydir/data/1
/home/mydir/data/2
        ⋮
```

The modifier telling PROPhet that the charge density is in a subdirectory Chg_density, applies to all directories that follow until the next data statement. So if, for example, you only want to use the charge density on the fine grid for the first 10 examples, you can say

input file

```
data code=vasp density=Chg_density
/home/mydir/data/1
/home/mydir/data/2
        ⋮
/home/mydir/data/10
data code=vasp
/home/mydir/data/11
/home/mydir/data/12
        ⋮
```

## Per-code caveats

Due to differences in how the first-principles codes store information, there are some things to keep in mind depending on which first principles code you're generating data

with.

**VASP** VASP writes an xml file (vasprun.xml) to the directory in which the run is conducted, so this is the directory that should be given to PROPhet.

**QE** Quantum Espresso creates a subdirectory (by default called pwscf.save) in which it stores the xml and charge density files. This subdirectory should be used as the base directory for specifying QE data to PROPhet. Also, the total energy is not written to the QE xml file. If you want to use energy computed with QE with a network you must either use the user_input file (see Section 3) or put the energy into the xml file yourself by adding the line <TOTAL_ENERGY> put_QE_energy_here </TOTAL_ENERGY> to the xml file of each training example.

**FHI-Aims** FHI-Aims does not write standard files during runtime. Instead, it prints its data to STDOUT, which must be redirected to a file. Rather than require the user to name this file a certain ways, PROPhet requires that the name of the file instead of the directory in which it resides as is the case for VASP and QE data.

## The user_input file

It is possible to use PROPhet with properties not handled by the built-in interface. These can be given to PROPhet via a special file called “user_input”. Lines in this file correspond to values you want PROPhet to use for training. Each line corresponds to a single property, which can be either a scalar or a vector. As with built-in properties, these user-defined properties are given to PROPhet in “input” and “output” statements in the input file using the parameter

user#

where # is an integer giving the line number of that property (starting from 1) in the user_input file.

For example, suppose you wanted a network to fit the 9 elements of the dielectric tensor and the total energy to the fermi level, you could do this by specifying

input file

```
input = user1 energy
output = user2
```

You would then create a file in the directory tree of each training example with 2 lines, the first holding the 9 dielectric tensor values (separated by whitespace) and the second with the fermi level. If the file is placed in a directory other than the base directory, this can be specified as usual within a data statement as

input file

```
data user=relative_path_to_user_input_file ...
```

# 4 The input file

The input file of PROPhet is free format; whitespace is ignored and the file is not case sensitive (with the exception of directory/file names). Comments can be started with the "#" character. Parameters are specified by parameter=value pairs, and can be given in any order, with the exception of data blocks (see section 4.21).

Table 2: Reference table of input file parameters. Required parameters are marked with "»".

| | Parameter | Description | input type | Default |
|---|---|---|---|---|
| | **Network construction parameters** | | | |
| » | network_type | type of machine learning network | string | nn |
| » | hidden | topology of hidden layers | vector of ints | – |
| » | input | desired input properties | vector of strings | – |
| » | output | the target property | vector of strings | – |
| | transfer | hidden nodes use this transfer function | string | tanh |
| | **Parameters governing training** | | | |
| | algorithm | training algorithm to use | string | rprop |
| | threshold | stop training when gradient drops below threshold | real | 1e-6 |
| | Niterations | maximum number of iterations | int | $\infty$ |
| | Nprint | print progress every Nprint timesteps | int | 100 |
| | Ncheckpoint | save network every Ncheckpoint iterations | int | 100 |
| | checkpoint_in | base name of the checkpoint file to read | string | – |
| | checkpoint_out | base name of the checkpoint file to write | string | – |
| | regularization | add sum of squared parameters to error function | real | 0 |
| | precondition_output | turn on/off output preconditioning | bool | false |
| | sd_momentum | magnitude of momentum term in steepest descent | real | 0 |
| | line_min_epsilon | threshold for BFGS line minimization | real | 0.1*threshold |
| | **Parameters for potentials** | | | |
| | Rcut | radial cutoff distance | real | 6 |
| | Nradial | number of radial functions to use | int | 8 |
| | Nangular | number of angular functions to use | int | 8 |
| | G1 | Specified functions of type G1 | vector of reals | – |
| | G2 | Specified functions of type G2 | vector of reals | – |
| | G3 | Specified functions of type G3 | vector of reals | – |
| | G4 | Specified functions of type G4 | vector of reals | – |
| | **Data specification block** | | | |
| » | data | data specifications follow | vector of strings | – |
| | **MISC** | | | |
| | include | Recursively include other input files | string | – |
| | downsample | Reduction factor for sampling a density | int | 1 |

## 4.1 network_type

**value** string

**description** This determines the underlying machine learning method to use. Currently only neural_networks (nn) are implemented.

## 4.2 hidden

**value** vector of ints

**description** This determines the network topology by specifying the hidden nodes. There should be 1 integer for each layer, specifying the number of nodes in that layer. For example, a network with 2 hidden layers of 10 and 15 nodes would be specified by "hidden = 10 15". Note that input and output layers are not specified here, but are determined automatically by PROPhet.

## 4.3 input

**value** vector of strings

**description** These are the system properties to use as input to the network. The current built-in possibilities are

- ✶energy
- ✶density
- ✶energy
- ✶volume
- ✶dft_gap
- ✶gw_gap
- ✶user# (see The user_input file)

PROPhet will automatically extract these properties from the output files of their runs. With the exception of "structure", which can currently only be used alone, any combination of these properties can be specified for input.

## 4.4 output

**value** string

**description** Same as input, except that currently only a single output can be specified. Also, it is not possible to use “structure” as an output.

## 4.5 transfer

**value** string

**description** The non-linear transfer function to be used by hidden nodes (the output node is always linear). Current options are

- tanh
- tanh_spline

Specifying the transfer function in the input file sets **all** hidden nodes with this function. The transfer function can be set on a per-node basis by editing the checkpoint file (see Section 6)

## 4.6 algorithm

**value** string

**description** This sets the training algorithm to use. Current options are

- rprop
- steepest_descent
- bfgs

Rprop (which is the default) specifies a flavor of the resilient backpropagation algorithm and is an excellent choice in the initial stages of training when one starts far from the minimum.

Steepest descent is steepest descent with an adaptive step size. One can couple this with the “sd_momentum” parameter to use a momentum term that includes a portion (set by the magnitude of sd_momentum) of the previous step into the current step.

BFGS invokes the limited memory BFGS algorithm, and is a good choice when close to the minimum. This option performs a line minimization using Brent’s method at each

step. Because of this, care should be taken that one has reached a convex and relatively smooth region of parameter space (i.e. generally don't use this in the initial stages of training). Also, the line minimization can require many sub-steps, so each step can take significantly longer than either Rprop or steepest_descent, but more progress should be made on each step.

## 4.7 threshold

**value** real

**description** This is one way to set a stopping criterion for training. Training will stop when the magnitude of the gradient (the last column of STDOUT) drops below this value.

## 4.8 Niterations

**value** int

**description** This is one way to set a stopping criterion for training. Training will stop when the number of iterations reaches Niterations.

## 4.9 Nprint

**value** int

**description** Information will be printed to STDOUT every Nprint iterations. Note that printing too frequently can sometimes slow down a run.

## 4.10 Ncheckpoint

**value** int

**description** The checkpoint files will be written to "checkpoint_out" every Ncheckpoint iterations.

## 4.11 checkpoint_in

**value** string

**description** This sets the name of the checkpoint file from which PROPhet will read. Note that if checkpoint_in is specified, the file must exist or PROPhet will complain. checkpoint_in must be specified for any task that requires knowledge of a pre-existing network (i.e. continuing an already started training run, predicting new examples using -run, or validating a fit using -validate).

NOTE: In the case that one is using input = structure, checkpoint_in specifies the base name of the files. PROPhet will add _ELEMENT (where ELEMENT is an atomic symbol) to this name and read from a separate file for each atomic species.

## 4.12 checkpoint_out

**value** string

**description** This sets the name of the checkpoint file to which PROPhet will write. This file need not exist, but will be overwritten if it does exist.

NOTE: In the case that one is using input = structure, checkpoint_out specifies the base name of the files. PROPhet will add _ELEMENT (where ELEMENT is an atomic symbol) to this name and write to a separate file for each atomic species.

## 4.13 regularization

**value** real

**description** This turns on L2 preconditioning of the network during training. L2 preconditioning puts constraints on the magnitude of network parameters by adding the sum of the squared magnitudes of the parameters to the error definition. That is, the error is written as

## 4.14 precondition_output

**value** boolean (1/0 or true/false)

**description** Instructs PROPhet whether to use output preconditioning, which scales and shifts the output so that their mean is 0 and variance is 1. This can speed up training in cases where the outputs of all training examples and expected predictions are similar, but can cause problems if e.g. one is making a potential with significantly different supercell sizes. Use this option with caution.

## 4.15 sd_momentum

**value** real

**description** This sets a momentum for the steepest descent algorithm. With momentum turned on, a portion of the previous step direction will be added to the current step direction. This can help avoid local minima in certain circumstances. This value sets a relative weight for the old step relative to the new, with 0 being no momentum and 1 being equally weighted with the current step.

## 4.16 line_min_epsilon

**value** real

**description** When the BFGS algorithm is used, a full line minimization is done at each step. The default tolerance for this minimization is 0.1*threshold, that is, the line minimization stops when the magnitude of the gradient along the line drops below 0.1*threshold. If threshold is set too low, for example if it is set to 0 to avoid stopping based on gradient magnitude, this can obviously cause problems. Generally, PROPhet will complain bitterly about line minimization not reaching the specified tolerance. This parameter allows the user to set a different line minimization tolerance. The line minimization will stop when the magnitude of the gradient projected along the search direction drops below line_min_epsilon.

## 4.17 Rcut

**value** real

**description** This sets the cutoff radius for inter-atomic interactions in a potential. Atoms beyond this distance will not be included in the sums over atomic interactions. This parameter must be chosen somewhat carefully for periodic systems. If Rcut is larger than the lattice constant, it will include redundant atoms which don't add information. If a potential trained in this manner is used to predict systems with a larger unit cell size, problems can occur since the atoms outside the original cell will generally have positions different than they had in any input example. This should not be a problem if enough data are included for which the lattice constant is larger than Rcut.

## 4.18 Nradial

**value** int

**description** This tells PROPhet to use *Nradial* radial functions of type G2 (see Section 4.20.2). These are chosen using equally spaced Rs from 0 to Rcut, and are given widths such that one function drops to 1e-5 exactly at the center of the function 2 over from it.

## 4.19 Nangular

**value** int

**description** This tells PROPhet to use *Nangular* angular functions of type G4 (see Section 4.20.4). These are chosen with equally spaced $\xi$.

## 4.20 Mapping function specification

Nradial and Nangular (or their defaults) can be used to automatically select mapping functions for a potential. The parameters G1, G2, G3, and G4 described here can be used to manually define all functions. For a full description of the method used here see[2,3]

For all G-functions, the radial cutoff function $f_c(R)$ is defined as either:
$f_c(R) = \frac{1}{2}\left(\cos(\frac{\pi R}{R_{cut}}) + 1\right)$ cutoff_type = 0
or
$f_c(R) = \left(\tanh(1 - \frac{R}{R_{cut}}\right)^3$ cutoff_type = 1
where Rcut is the cutoff radius defined in Section 4.17, and $R$ is the distance between two atoms.

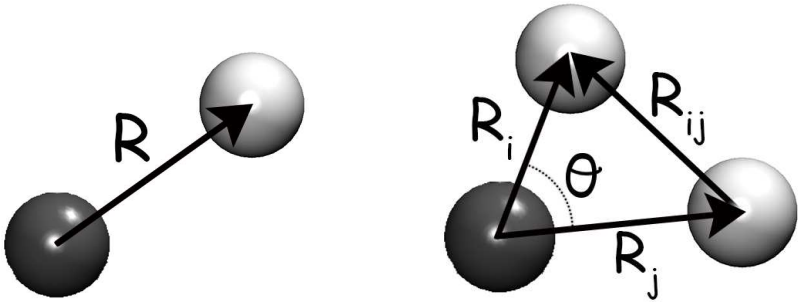



Figure 1: Schematic diagram showing definition of coordinates for (left) a pairwise interaction for G1 and G2 and (right) a 3-body G3 and G4 interaction.

#### 4.20.1 G1

**value** vector of reals ( $R_{cut}$ cutoff_type )
**description** Pairwise function given by
$G_1 = f_c(R)$

#### 4.20.2 G2

**value** vector of reals ( $R_{cut}$ cutoff_type $\eta$ $R_s$ )
**description** Pairwise function given by
$G_2 = e^{\left(-\eta(R-R_s)^2\right)} f_c(R)$

#### 4.20.3 G3

**value** vector of reals ( $R_{cut}$ cutoff_type $\eta$ $\xi$ $\lambda$ )

**description** 3-body function given by
$G_3 = 2^{(1-\xi)} \left(1 + \lambda \cos(\theta)\right)^{\xi} e^{\left(-\eta\left(R_i^2+R_j^2+R_{ij}^2\right)\right)} f_c(R_i) f_c(R_j) f_c(R_{ij})$
(see Figure **??** for coordinate definitions).

#### 4.20.4 G4

**value** vector of reals ($R_{cut}$ cutoff_type $\eta$ $\xi$ $\lambda$ )
**description** 3-body function given by
$G_4 = 2^{(1-\xi)} \left(1 + \lambda \cos(\theta)\right) e^{\left(-\left(R_i^2+R_j^2\right)\right)} f_c(R_i) f_c(R_j)$
(see Figure **??** for coordinate definitions)

## 4.21 data

**value** vector of strings (format parameter=value)
**description** While data in PROPhet are specified as a series of input lines containing base directory/file names, this critical parameter modifies how these data are handled. The modification affects all input lines that follow until the next data statement. The most important parameter here is “code” which tells PROPhet which code (currently VASP, QE, or FHIAims) was used to generate the data. PROPhet will extract the necessary properties directly from the output files. Note that any code can be used with PROPhet if the data can be massaged into the format of one of these codes. The remaining parameters modify the directory (relative to the base) for each property specified in “input”. This feature allows different properties to be computed in separate runs. For example, to do a network fitting the DFT band gap (computed in the base directory) to the band gap computed with the GW method (computed in a subdirectory called GW), both computed in VASP, the data line should read “data code=vasp gw_gap=GW”. Note that if all properties are to be taken from a single run, only the “code” parameter needs to be specified.

## 4.22 include

**value** string (filename)

**description** This powerful parameter allows the user to recursively insert input files. The include parameter in PROPhet works similarly to the way it works in the C/C++ language. The file named in "value" is essentially pasted to this location in the input file. This command can be stacked, i.e. a file *included* can itself contain include directives. This parameter can be used to easily switch data sets for training and testing. The different sets can be specified in different files which are selected with the appropriate "include" statement. It can also be used to switch training schemes for the same data set.

## 4.23 downsample

**value** int

**description** When providing a charge density to PROPhet, it is sometimes useful to speed calculations by sampling the density on a coarser grid than the one provided. Setting "downsample" resamples the density by averaging over blocks of size $N \times N \times N$. Note that it is currently only possible to use this flag with densities from VASP, and the blocks must be cubic (i.e. you can't specify different step sizes per direction).

# 5 Output

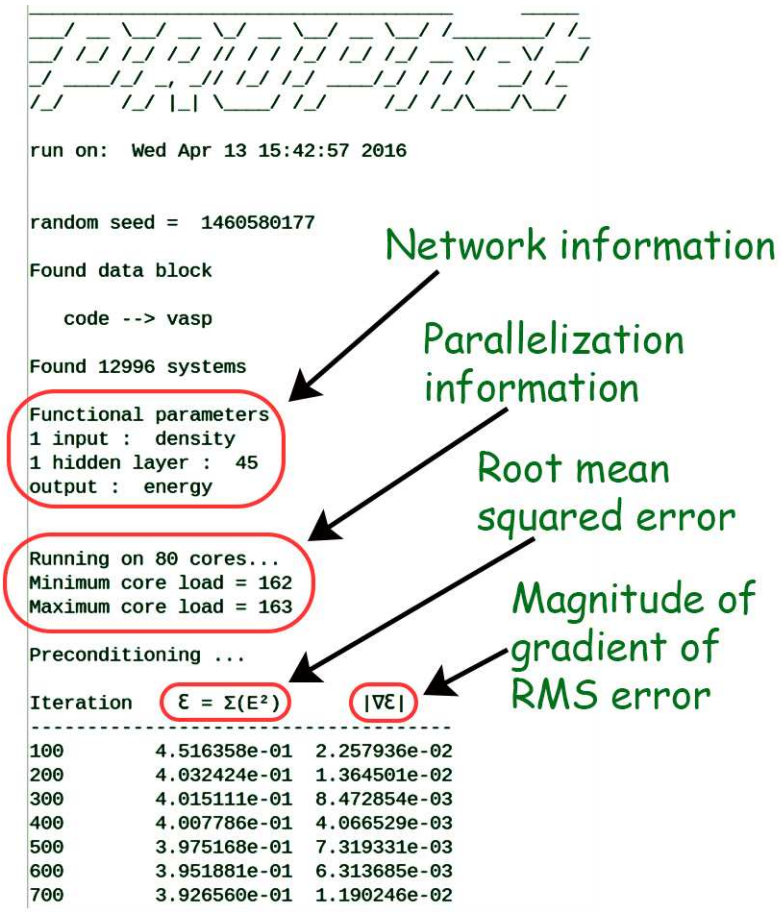



Figure 2: Standard output of PROPhet.

# 6 The checkpoint file

Network states are saved in checkpoint files specified by the “checkpoint_out” input parameter. The format of this file depends on the precise run being done. Figure 3 shows the layout of the header information of the checkpoint file.

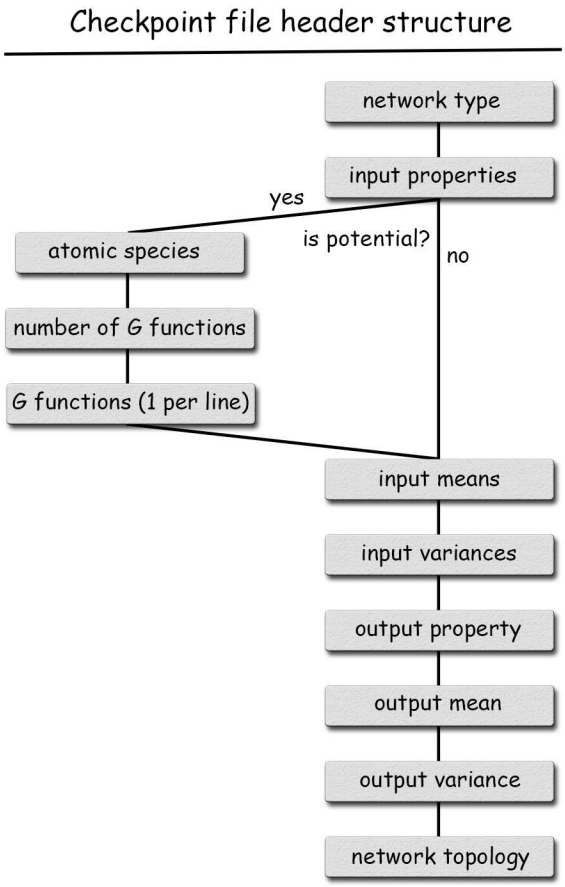
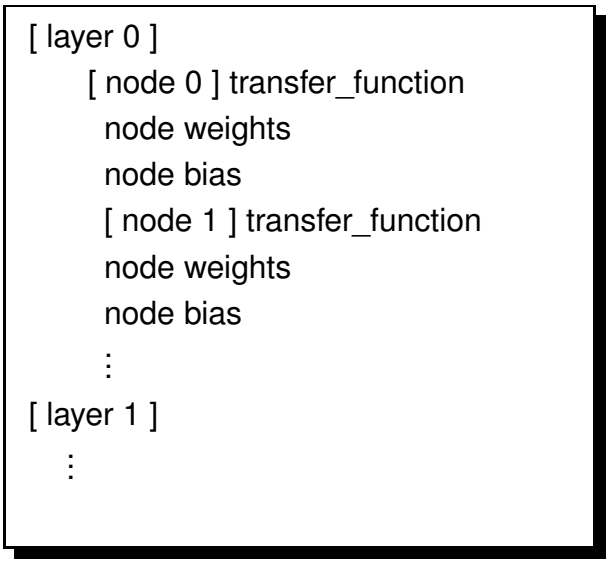



Figure 3: Schematic of the checkpoint file header structure. Each box corresponds to a single line in the file.

Following the header are the parameters of the network. They are organized by layer then node. Next to the start of each node is the transfer function for that node. These can be changed on a per node basis after network construction. The layout follows the pattern

checkpoint file




# Part V

# Theory

This part is intended for a brief introduction to the major approaches that PROPhet takes.

## 7 Charge density

The Hohenberg-Kohn theorems state that the ground state charge density of an atomic system forms a fundamental variable from which all relevant properties of the system can be determined. They do not necessarily tell us how to determine them, however. In the case of the total energy, it is known that there exists a density functional which can produce the energy from the density. The exact functional is not known, but reasonable approximations to it can be found. These approximations often take the form of an integral

over some function of the charge density (LDA) or the density and its gradient (GGA). In principle procedure could hold for approximations of properties besides energy, except that there is no intuitive form for the function.

This is precisely the situation in which machine learning techniques excel. The entire purpose of machine learning approaches such as neural networks, is to find mappings between inputs and outputs when no functional form can be found a priori. The trick with using machine learning here, is to determine how to give the network a charge density.

The idea is to use an approach similar to that taken to form LDA, that is, to write the functional for some property $A$ as,

$$A = \int f\left(\rho\right)\, d\Omega\ .$$

The function $f(\rho)$ is an unknown function that we seek to find via machine learning. To perform this learning task, PROPhet performs the integral explicitly for each charge density. For every point in the density the current network is fired and the results are summed after scaling by an appropriate $d\Omega$ (as determined by the sampling of the charge density). The resulting integral is compared to the target value for each training example and the error computed. Once all training examples have been computed, the gradient of the error with respect to network parameters is computed in the usual way.

# 8 Structure

Using the atomic structure (conformation) as an input requires a means by which to map atomic coordinates to input for a network. The network input should reflect the fact that exchange of two indistinguishable particles does not change the system's properties and also any relevant spatial symmetry. One must also ensure that the input vector to the network always has the same size and each of its elements always means the same thing. These restrictions actually rule out many intuitive methods to give the atomic structure to a network.

PROPhet performs the mapping of atomic coordinates to network inputs using the

method of Behler and Parrinello[2–4]. In this approach, each atom in the system has a separate network associated with it. This differs from most previous approaches that used 1 network for the entire system. The networks for all atoms of a given type (e.g. Au) are identical, but generally different from the networks for other types. To determine the value of a system property (e.g. energy) the networks are summed over, with each one producing the contribution to the energy from its individual atom. This summing over atomic contributions automatically satisfies the exchange symmetry mentioned above, since the sum is independent of the order in which it is taken.

The input to the network for each atom is a set of numbers which together form a descriptor of the chemical environment in which that atom resides. The numbers are found by evaluating special mapping functions. Different choices can be made for these mapping functions so long as they can be normalized and reflect necessary symmetries. In PROPhet, the mapping functions are given by the G1, G2, G3, and G4 functions discussed in Section Mapping function specification. Each of these functions takes the internal coordinates corresponding to a particular interaction and produces a number. The sum of these numbers over all interactions for a particular mapping function forms the value that is given to the network for that function. The summing over interactions is important, since the network must always be given the same number of inputs, but coordination numbers can change for different structures (during and MD simulation for example).

The interactions considered for each atom are limited to those inside some chosen cutoff radius. The use of a cutoff radius not only speeds computation, but allows the networks trained on a system of a given size to be used for systems with a different size, so long as there are roughly similar numbers of atoms inside the cutoff radii of both systems. This is a substantial advantage over methods that use a single network for the entire system, since these generally must be trained and used on a single system with the same number and types of atoms. This is especially advantageous since ab initio calculations are generally expensive and generally must be limited to relatively small systems. With PROPhet, you can train on small systems that are accessible to ab initio approaches, but run predictions (or MD simulations in the case of fitting potentials) on systems of more or less arbitrary size.

A full description of the method is outside the scope of this documentation, but we refer the interested reader to the literature[2,3].

# References

(1) Plimpton, S. *J. Comp. Phys.* **1995**, *117*, 1–19.

(2) Behler, J.; Parrinello, M. *Phys. Rev. Lett.* **2007**, *98*, 146401.

(3) Behler, J. *Journal of Physics: Condensed Matter* **2014**, *26*, 183001.

(4) Behler, J. *International Journal of Quantum Chemistry* 1032.

(5) LAMMPS website. http://lammps.sandia.gov/.