Supporting Information: Uncertainty Quantification in Machine Learning and Nonlinear Least Squares Regression Models

Ni Zhan and John R. Kitchin

July 22, 2021

1 One dimension input NN (Figure 2)

```
import autograd
import autograd.numpy as np
from autograd import hessian
import matplotlib.pyplot as plt
from autograd import grad
import autograd.numpy.random as npr
from scipy.stats.distributions import t
from scipy.optimize import minimize

# lennard jones potential
def func(x, e, s):
    return 4 * e * (np.power(np.divide(s, x), 12) -
                    np.power(np.divide(s, x), 6))

etrue = 10
strue = 0.34
numpts = 23

#xfit is for plotting
xfit = np.arange(0.34, 0.49, 0.001)
xfit = np.expand_dims(xfit, axis=1)

# weightsparser to help roll and unroll weights and biases.
class WeightsParser(object):
    """A helper class to index into a parameter vector."""

    def __init__(self):
        self.idxs_and_shapes = {}
        self.N = 0

    def add_weights(self, name, shape):
        start = self.N
        self.N += np.prod(shape)
        self.idxs_and_shapes[name] = (slice(start, self.N), shape)

    def get(self, vect, name):
        idxs, shape = self.idxs_and_shapes[name]
        return np.reshape(vect[idxs], shape)

# params is a 1-d vector of weights and biases
# parser is object that makes it easy to unroll params into matrices of
# weights and biases.
def init_random_params(scale, layer_sizes, rs=None):
    if rs is None:
        rs = npr.RandomState(2)
    parser = WeightsParser()
    for i, shape in enumerate(zip(layer_sizes[:-1], layer_sizes[1:])):
        parser.add_weights(('weights', i), shape)
        parser.add_weights(('biases', i), (1, shape[1]))
    return rs.randn(parser.N), parser

# nn predict by unrolling w parser.
def nn_predict(params, inputs, nonlinearity=np.tanh):
    cur_units = inputs
    for layer in range(len(layer_sizes) - 1):
        cur_W = parser.get(params, ('weights', layer))
        cur_B = parser.get(params, ('biases', layer))
        cur_units = np.dot(cur_units, cur_W) + cur_B
        if layer < len(layer_sizes) - 2:
            cur_units = nonlinearity(cur_units)
    return cur_units

#objective with regularization to be used with scipy minimize
def objectivel2(params, X, r, alpha=0):
    ypredict = nn_predict(params, X)
```

```
    errs = r - ypredict
    weights = params[idxs]
    return np.sum(errs**2) + alpha * np.linalg.norm(weights)

layer_sizes = [1, 4, 1]
_, parser = init_random_params(1, layer_sizes)

# get the index of the weights, because only regularizing weights.
idxs = []
for layer in range(len(layer_sizes) - 1):
    sliceidx, _ = parser.idxs_and_shapes[('weights', layer)]
    idxs += [np.r_[sliceidx]]
idxs = np.array(idxs).flatten()

#sum-squared-errors
def sse(params, X, r):
    ypredict = nn_predict(params, X)
    errs = r - ypredict
    return np.sum(errs**2)

#get inverse fisher information
def get_pcov(h):
    eigs0 = np.linalg.eigvalsh(h)[0]
    if (eigs0 <0):
        eps = max(1e-5, eigs0*-1.05)
    else:
        eps = 1e-5
    j = np.linalg.pinv(h + eps * np.identity(h.shape[0]))
    pcov1 = j * scaling
    u, v = np.linalg.eigh(pcov1)
    return v @ np.diag(np.maximum(u,0)) @ v.T

#get standard errors of prediction, confidence
def getpredse(x, params):
    gprime = autograd.elementwise_grad(nn_predict,0)(params, x)
    sesq = gprime @ pcov @ gprime
    return np.sqrt(sesq), np.sqrt(sesq + scaling)

#get standard errors for a dataset
def get_se_dataset(xfit, params):
    predses = []
    for i in xfit:
        predses += [getpredse(i, params)]
    return np.array(predses)

# to make plot
# data for panel 1.
numpts = 23
xa = np.linspace(0.35, 0.45, numpts)
np.random.seed(seed=0)

ya = func(xa, etrue, strue) + np.random.normal(scale=0.2, size=xa.shape)

Xa = np.expand_dims(xa, axis=1)
ra = np.expand_dims(ya, axis=1)

initial_guess, parser = init_random_params(1, layer_sizes)

sol = minimize(objectivel2, initial_guess, args=(Xa,ra,0.01) )
paramsa = sol.x

h = hessian(sse,0)(paramsa, Xa, ra)
numptsa = Xa.shape[0]
scaling = sse(paramsa, Xa, ra)/numptsa

pcov = get_pcov(h)

predsesa = get_se_dataset(xfit, paramsa)
```

```python

#data for panel 2.
x1 = np.linspace(0.35, 0.365, 7)
x2 = np.linspace(0.415, 0.45, 9)
xb = np.concatenate((x1,x2))
yb = func(xb, etrue, strue) + np.random.normal(scale=0.2, size=xb.shape)

Xb = np.expand_dims(xb, axis=1)
rb = np.expand_dims(yb, axis=1)

initial_guess, _ = init_random_params(1, layer_sizes)

sol = minimize(objectivel2, initial_guess, args=(Xb,rb,0.005) )
paramsb = sol.x

h = hessian(sse,0)(paramsb, Xb, rb)
numptsb = Xb.shape[0]
scaling = sse(paramsb, Xb, rb)/numptsb

pcov = get_pcov(h)

predsesb = get_se_dataset(xfit, paramsb)


#make a plot.
plt.clf()
fig, ax = plt.subplots(ncols =2, nrows = 1, sharex=False, sharey='row')
fig.set_size_inches(7,3)
tvala = t.ppf(0.975, numptsa)
tvalb = t.ppf(0.975, numptsb)

ypreda = nn_predict(paramsa, xfit).flatten()
ypredb = nn_predict(paramsb, xfit).flatten()

ax[0].set_title(' ')
ax[0].plot(Xa, ra, 'bo')
ax[0].plot(xfit, ypreda)
ax[0].plot(xfit, func(xfit, etrue, strue))
ax[0].plot(xfit, ypreda + predsesa[:,0] * tvala, '--r')
ax[0].plot(xfit, ypreda - predsesa[:,0] * tvala, '--r')
ax[0].set_xlabel('x')
ax[0].set_ylabel('y')

ax[1].set_title(' ')
ax[1].plot(Xb, rb, 'bo')
ax[1].plot(xfit, ypredb)
ax[1].plot(xfit, func(xfit, etrue, strue))
ax[1].plot(xfit, ypredb + predsesb[:,0] * tvalb, '--r')
ax[1].plot(xfit, ypredb - predsesb[:,0] * tvalb, '--r')
ax[1].set_xlabel('x')

ax[1].legend(['Data', 'NN', 'f(x)', '95% confidence'])

plt.subplots_adjust(wspace=0)
plt.tight_layout()

plt.savefig(f'subplot-2panel.png', dpi=200)
print('''#+attr_org: :width 600
#+caption: Figure 2
[[./subplot-2panel.png]]''')
```

2 Training a SingleNN model

The database file used for the first potential contained configurations with 3.934 Å lattice constant.

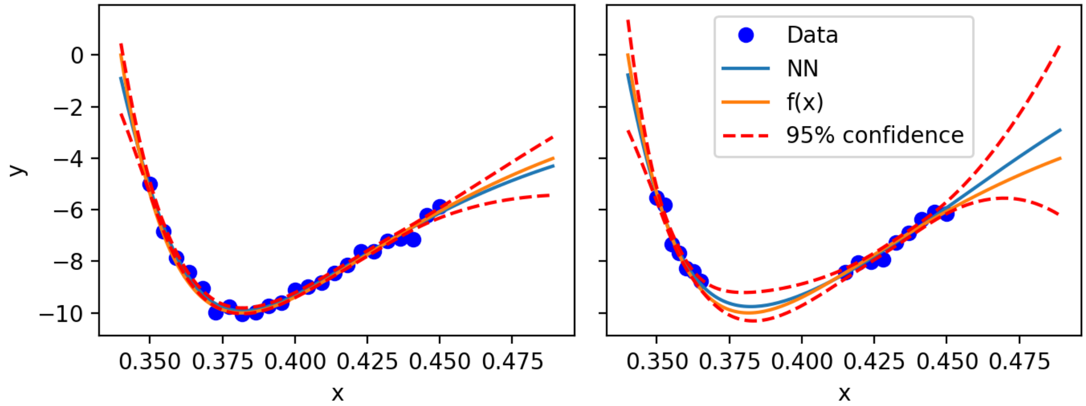

Figure 1: Figure 2

The following code uses singleNN code found here: https://github.com/lmj1029123/SingleNN, and mostly follows the github tutorial. The code splits the dataset, configures the singleNN, and trains the model. The code generates a directory folder "lattice39-2" with relevant files: splitted dataset files "final_train.sav", "final_val.sav", "test.sav"; model file "best_model".

```
import sys

sys.path.append("../SimpleNN")
sys.path.append("../")

import os
from ase.db import connect
import torch
from ContextManager import cd
from preprocess import train_test_split, train_val_split, get_scaling, CV
from preprocess import snn2sav
from NN import MultiLayerNet
from train import train, evaluate
from fp_calculator import set_sym, calculate_fp
import pickle

is_train = True
is_transfer = False
is_force = True

if is_train and is_transfer:
    raise ValueError('train and transfer could not be true at the same time.')

##################################################################################
#Hyperparameters
##################################################################################
E_coeff = 100
if is_force:
    F_coeff = 1
else:
    F_coeff = 0

val_interval = 10
n_val_stop = 10
epoch = 3000
```

```python

opt_method = 'lbfgs'


if opt_method == 'lbfgs':
    history_size = 100
    lr = 1
    max_iter = 10
    line_search_fn = 'strong_wolfe'


convergence = {'E_cov':0.0005,'F_cov':0.005}

# min_max will scale fingerprints to (0,1)
fp_scale_method = 'min_max'
e_scale_method = 'min_max'


test_percent = 0.2
# Pecentage from train+val
val_percent = 0.2

# Training model configuration
SEED = [2]
n_nodes = [11,11]
activations = [torch.nn.Sigmoid(), torch.nn.Sigmoid()]
lr = 1
hp = {'n_nodes': n_nodes, 'activations': activations, 'lr': lr}

####################################################################################
#Configuration
####################################################################################

if is_train:
    # The Name of the training
    Name = f'lattice39'
    for seed in SEED:
        if not os.path.exists(Name+f'-{seed}'):
            os.makedirs(Name+f'-{seed}')

    dbfile = f'data/lattice39.db'
    db = connect(dbfile)

    elements = ['Pd', 'Au']
    nelem = len(elements)
    # This is the energy of the metal in its ground state structure
    #if you don't know the energy of the ground state structure,
    # you can set it to None
    element_energy = None
    # Allen electronegativity
    weights =[1.58, 1.92]


    Gs = [22]
    cutoff = 6.35
    g2_etas = [0.00, 0.10713, 0.285686, 0.892769]
    g2_Rses = [0.0]


    sym_params = [Gs, cutoff, g2_etas, g2_Rses, elements, weights, element_energy]
    params_set = set_sym(elements, Gs, cutoff,
                         g2_etas=g2_etas, g2_Rses=g2_Rses,
                         weights=weights)

    N_sym = params_set[elements[0]]['num']

####################################################################################
#Training
```

```python
###############################################################################

Name = f'lattice39'
if is_train:
    for seed in SEED:
        # This use the context manager to operate in the data directory
        with cd(Name+f'-{seed}'):
            pickle.dump(sym_params, open("sym_params.sav", "wb"))
            logfile = open('log.txt','w+')
            resultfile = open('result.txt','w+')

            if os.path.exists('test.sav'):
                logfile.write('Did not calculate symfunctions.\n')
            else:
                data_dict = snn2sav(db, Name, elements, params_set,
                                    element_energy=element_energy)
                train_dict = train_test_split(data_dict,1-test_percent,seed=seed)
                train_val_split(train_dict,1-val_percent,seed=seed)

            logfile.flush()

            train_dict = torch.load('final_train.sav')
            val_dict = torch.load('final_val.sav')
            test_dict = torch.load('test.sav')
            scaling = get_scaling(train_dict, fp_scale_method, e_scale_method)


            n_nodes = hp['n_nodes']
            activations = hp['activations']
            lr = hp['lr']
            model = MultiLayerNet(N_sym, n_nodes, activations, nelem, scaling=scaling)
            if opt_method == 'lbfgs':
                optimizer = torch.optim.LBFGS(model.parameters(), lr=lr,
                                              max_iter=max_iter, history_size=history_size,
                                              line_search_fn=line_search_fn)

            results = train(train_dict, val_dict,
                            model,
                            opt_method, optimizer,
                            E_coeff, F_coeff,
                            epoch, val_interval,
                            n_val_stop,
                            convergence, is_force,
                            logfile)
            [loss, E_MAE, F_MAE, v_loss, v_E_MAE, v_F_MAE] = results

            test_results = evaluate(test_dict, E_coeff, F_coeff, is_force)
            [test_loss, test_E_MAE, test_F_MAE] =test_results
            resultfile.write(f'Hyperparameter: n_nodes = {n_nodes}, activations = {activations}, lr = {lr}\n')
            resultfile.write(f'loss = {loss}, E_MAE = {E_MAE}, F_MAE = {F_MAE}.\n')
            resultfile.write(f'v_loss = {v_loss}, v_E_MAE = {v_E_MAE}, v_F_MAE = {v_F_MAE}.\n')
            resultfile.write(f'test_loss = {test_loss}, test_E_MAE = {test_E_MAE}, test_F_MAE = {test_F_MAE}.\n')


            logfile.close()
            resultfile.close()
```

3 Preprocessing the predict-4.0 and 4.1 datasets

The database files containing configurations with 4.034 Å lattice constant: , and configurations with 4.134 Å lattice constant:

The following code splits the predict-4.0 and 4.1 datasets, generating directory folders "lattice40_pred-2" and "lattice41_pred-2" with relevant files: split dataset files "final_train.sav", "final_val.sav", "test.sav".

```python
import sys

sys.path.append("../SimpleNN")
sys.path.append("../")

import os
from ase.db import connect
from ContextManager import cd
from preprocess import train_test_split, train_val_split, get_scaling, CV
from preprocess import snn2sav
from fp_calculator import set_sym, calculate_fp


# min_max will scale fingerprints to (0,1)
fp_scale_method = 'min_max'
e_scale_method = 'min_max'


test_percent = 0.2
# Pecentage from train+val
val_percent = 0.2

# Training model configuration
SEED = [2]

################################################################################
#Split Predict-4.0 dataset
################################################################################


Name = f'lattice40_pred'

for seed in SEED:
    if not os.path.exists(Name+f'-{seed}'):
        os.makedirs(Name+f'-{seed}')

dbfile = 'data/lattice40.db'
db = connect(dbfile)

elements = ['Pd', 'Au']
nelem = len(elements)

element_energy = None
weights =[1.58, 1.92]

Gs = [22]
cutoff = 6.35
g2_etas = [0.00, 0.10713, 0.285686, 0.892769]
g2_Rses = [0.0]


sym_params = [Gs, cutoff, g2_etas, g2_Rses, elements, weights, element_energy]
params_set = set_sym(elements, Gs, cutoff,
                         g2_etas=g2_etas, g2_Rses=g2_Rses,
                          weights=weights)
N_sym = params_set[elements[0]]['num']

with cd(Name+f'-{seed}'):
    data_dict = snn2sav(db, Name, elements, params_set,
                                    element_energy=element_energy)

    train_dict = train_test_split(data_dict,1-0.2,seed=seed)
    train_val_split(train_dict,1-0.2,seed=seed)

################################################################################
#Split Predict-4.1 dataset
################################################################################
```

```
Name = f'lattice41_pred'

for seed in SEED:
    if not os.path.exists(Name+f'-{seed}'):
        os.makedirs(Name+f'-{seed}')

dbfile = 'data/lattice41.db'
db = connect(dbfile)

with cd(Name+f'-{seed}'):
    data_dict = snn2sav(db, Name, elements, params_set,
                                    element_energy=element_energy)

    train_dict = train_test_split(data_dict,1-0.2,seed=seed)
    train_val_split(train_dict,1-0.2,seed=seed)
```

4 Uncertainty and plots for first model

The following code imports functions from the python file: .

```
import torch
from uncert import evaluate_uncert
import numpy as np
import matplotlib.pyplot as plt
from scipy.stats.distributions import t
from Batch import batch_pad

#get inverse fisher information
def get_pcov(h):
    eigs0 = np.linalg.eigvalsh(h)[0]
    if (eigs0 <0):
        eps = max(1e-5, eigs0*-1.05)
    else:
        eps = 1e-5
    j = np.linalg.pinv(h + eps*np.identity(h.shape[0]))
    pcov1 = j*alpha
    u, v = np.linalg.eigh(pcov1)
    return v @ np.diag(np.maximum(u,0)) @ v.T


def flatten_gprime(agrad):
    cnt = 0
    for g in agrad:
        g_vector = g.contiguous().view(-1) if cnt ==0 else torch.cat([g_vector, g.contiguous().view(-1)])
        cnt = 1
    return g_vector

#get uncertainties for a dataset
def get_uncerts(name, data_dict):
    model = torch.load(name)
    scaling = model.scaling
    gmin = scaling['gmin']
    gmax = scaling['gmax']
    emin = scaling['emin']
    emax = scaling['emax']

    ids = np.array(list(data_dict.keys()))
    batch_info = batch_pad(data_dict,ids)
    b_fp = batch_info['b_fp']

    b_e_mask = batch_info['b_e_mask']
    b_fp.requires_grad = True
    sb_fp = (b_fp - gmin) / (gmax - gmin)
```

```python

    N_atoms = batch_info['N_atoms'].view(-1)
    b_e = batch_info['b_e'].view(-1)
    b_f = batch_info['b_f']

    Atomic_Es = model(sb_fp)
    E_predict = torch.sum(Atomic_Es * b_e_mask, dim = [1,2])
    E_predict = E_predict/N_atoms
    E_predict = E_predict * (emax - emin) + emin

    uncerts = []
    for i, ei in enumerate(E_predict):
        gprime = torch.autograd.grad(ei, model.parameters(), create_graph=True, retain_graph=True)
        gprime = flatten_gprime(gprime).detach().numpy()
        se = gprime @ pcov @ gprime
        uncerts += [(np.sqrt(se), np.sqrt(se + rmse.item()**2), np.linalg.norm(gprime))]
    uncerts = np.array(uncerts)
    return uncerts


Name = 'lattice39-2'

#load datasets
train_dict = torch.load(f'{Name}/final_train.sav')
val_dict = torch.load(f'{Name}/final_val.sav')
test_dict = torch.load(f'{Name}/test.sav')

#get NN predictions, RMSE, hessian
pred_e, actual_e, rmse, h = evaluate_uncert(f'{Name}/best_model',train_dict, True)
h = h.detach().numpy()
pred_e_val, actual_e_val, rmse_val = evaluate_uncert(f'{Name}/best_model',val_dict, False)
pred_e_test, actual_e_test, rmse_test = evaluate_uncert(f'{Name}/best_model',test_dict, False)


ndata = pred_e.shape[0]
alpha = rmse.item()**2
pcov = get_pcov(h)

#get uncertainties
uncerts_val = get_uncerts(f'{Name}/best_model',val_dict)
uncerts_train = get_uncerts(f'{Name}/best_model',train_dict)
uncerts_test = get_uncerts(f'{Name}/best_model',test_dict)

################################################################################
#Parity Plot
################################################################################

plt.clf()

fig, ax = plt.subplots(ncols =2, nrows=1, sharex=False, sharey='row')

fig.set_size_inches(7, 3.5)

eline = np.linspace(np.min(np.concatenate((actual_e, actual_e_test))),
                    np.max(np.concatenate((actual_e, actual_e_test))), 10)

ax[0].set_title(' ')
ax[0].plot(actual_e, pred_e, '.',color='tab:orange', alpha=1, label='Train')
ax[0].set_xlabel(' ')
ax[0].set_ylabel('NN Energy (eV/atom)')
ax[0].legend()
ax[0].plot(eline, eline,'k--',alpha=0.7)

ax[1].plot(eline, eline,'k--',alpha=0.7)
ax[1].plot(actual_e_val, pred_e_val, '.',color='g', alpha=0.9, label='Validation')
ax[1].plot(actual_e_test, pred_e_test, '.',color='y', alpha=0.8, label='Test')
ax[1].legend()

```

```
plt.figtext(0.55,0.04,"DFT Energy (eV/atom)", va="center", ha="center", size=10.5)
plt.tight_layout()
plt.savefig('subplotparityslides-energy-only.png')
print('''#+attr_org: :width 600
#+caption: Figure 4
[[./subplotparityslides-energy-only.png]]''')

#####################################################################################
# Distribution of uncertainties
#####################################################################################

plt.clf()
plt.hist(uncerts_train[:,0], label='Train', density=True, alpha=0.5, color='tab:orange')
plt.hist(uncerts_val[:,0], label='Validation', density='True', alpha=0.5, color='g')
plt.hist(uncerts_test[:,0], label='Test', density='True', alpha=0.5, color='y')
plt.legend()
plt.xlabel('Standard Error Confidence (eV/atom)')
plt.ylabel('Density')
plt.tight_layout()
plt.savefig('hist-uncerts-pot1.png')
print('''#+attr_org: :width 600
#+caption: Figure 5
[[./hist-uncerts-pot1.png]]''')


#####################################################################################
# Parity plot with 95% prediction interval
#####################################################################################

plt.clf()
tval = t.ppf(0.975, ndata)
plt.errorbar(actual_e_test, pred_e_test, yerr=tval*uncerts_test[:,1], fmt='y_',
             ecolor='m', label='Test, 95% prediction')

plt.xlabel('DFT Energy (eV/atom)')
plt.ylabel('NN Energy (eV/atom)')
plt.plot([np.min(actual_e_test), np.max(actual_e_test)],
         [np.min(actual_e_test),
          np.max(actual_e_test)],'k--', alpha=0.7, linewidth=0.3)
plt.legend()
plt.tight_layout()
plt.savefig('parity-errorbar-test-pot1-prediction.png', dpi=200)
print('''#+attr_org: :width 600
#+caption: Figure 6
[[./parity-errorbar-test-pot1-prediction.png]]''')

#####################################################################################
#Inference on predict-4.0 and 4.1 dataset
#####################################################################################

data_dict = torch.load(f'lattice40_pred-2/test.sav')
pred_e_40p, actual_e_40p, rmse_40p = evaluate_uncert(f'{Name}/best_model',data_dict, False)
uncerts_40p = get_uncerts(f'{Name}/best_model',data_dict)

data_dict = torch.load(f'lattice41_pred-2/test.sav')
pred_e_41p, actual_e_41p, rmse_41p = evaluate_uncert(f'{Name}/best_model',data_dict, False)
uncerts_41p = get_uncerts(f'{Name}/best_model',data_dict)

#make plot

plt.clf()
fig, ax = plt.subplots(ncols =2, nrows=1, sharex=False, sharey='row')
fig.set_size_inches(10, 4)
ax[0].set_title(' ')
ax[0].errorbar(actual_e_40p, pred_e_40p, yerr = tval * uncerts_40p[:,1], color='tab:pink',
               fmt = '_', ecolor='r', label='Predict 4.0, 95% prediction')
ax[0].set_xlabel(' ')
ax[0].set_ylabel('NN Energy (eV/atom)')
```

```python
ax[0].legend()
eline = np.linspace(np.min(np.concatenate((actual_e_40p, actual_e_41p))),
                    np.max(np.concatenate((actual_e_40p, actual_e_41p))), 10)
ax[0].plot(eline, eline,'k--', alpha=0.8, linewidth=0.5)

ax[1].errorbar(actual_e_41p, pred_e_41p, yerr =  tval * uncerts_41p[:,1],
               fmt = 'b_', ecolor='c', label='Predict 4.1, 95% prediction')
ax[1].legend()
ax[1].plot(eline, eline,'k--', alpha=0.7, linewidth=0.5)

plt.figtext(0.55,0.03,"DFT Energy (eV/atom)", va="center", ha="center", size=10.5)

plt.tight_layout()
plt.savefig('subplot-parity-40-41-pot-prediction.png', dpi=200)
print('''#+attr_org: :width 600
#+caption: Figure 7
[[./subplot-parity-40-41-pot-prediction.png]]''')

##################################################################################
#Uncertainty vs True Error Scatterplot
##################################################################################


def scatter_hist(x, y, ax, ax_histx, ax_histy, label, color=None):
    # no labels
    ax_histx.tick_params(axis="x", labelbottom=False)
    ax_histy.tick_params(axis="y", labelleft=False)

    # the scatter plot:
    ax.scatter(x, y, alpha=0.5, label=label, color=color)

    # now determine nice limits by hand:
    binwidth = 0.0001
    xymax = max(np.max(np.abs(x)), np.max(np.abs(y)))
    lim = (int(xymax/binwidth)+1)*binwidth

    #bins = np.arange(0, lim + binwidth, binwidth)
    ax_histx.hist(x,  alpha=0.5, color=color, density=True)
    ax_histy.hist(y,  orientation='horizontal', alpha=0.5, color=color, density=True)

fig = plt.figure(figsize=(8, 8))
# Add a gridspec with two rows and two columns and a ratio of 2 to 7 between
# the size of the marginal axes and the main axes in both directions.
# Also adjust the subplot parameters for a square plot.
gs = fig.add_gridspec(2, 2,  width_ratios=(7, 2), height_ratios=(2, 7),
                      left=0.11, right=0.98, bottom=0.07, top=0.97, wspace=0.05, hspace=0.05)

ax = fig.add_subplot(gs[1, 0])
ax_histx = fig.add_subplot(gs[0, 0], sharex=ax)
ax_histy = fig.add_subplot(gs[1, 1], sharey=ax)

# use the previously defined function

scatter_hist(np.absolute(actual_e_test-pred_e_test),  uncerts_test[:,0],
             ax, ax_histx, ax_histy, 'Test', 'y')
scatter_hist(np.absolute(actual_e_40p-pred_e_40p), uncerts_40p[:,0],
             ax, ax_histx, ax_histy, 'Predict 4.0', 'tab:pink')
scatter_hist(np.absolute(pred_e_41p-actual_e_41p), uncerts_41p[:,0],
             ax, ax_histx, ax_histy, 'Predict 4.1')


ax.set_xlabel('Absolute Error Energy (eV/atom)')
ax.set_ylabel('Standard Error Confidence (eV/atom)')
ax.legend()
plt.savefig('uncert-v-error-w-hist-pot1-origw-test.png', dpi=200,bbox_inches='tight')
print('''#+attr_org: :width 600
#+caption: Figure 9
[[./uncert-v-error-w-hist-pot1-origw-test.png]]''')
```

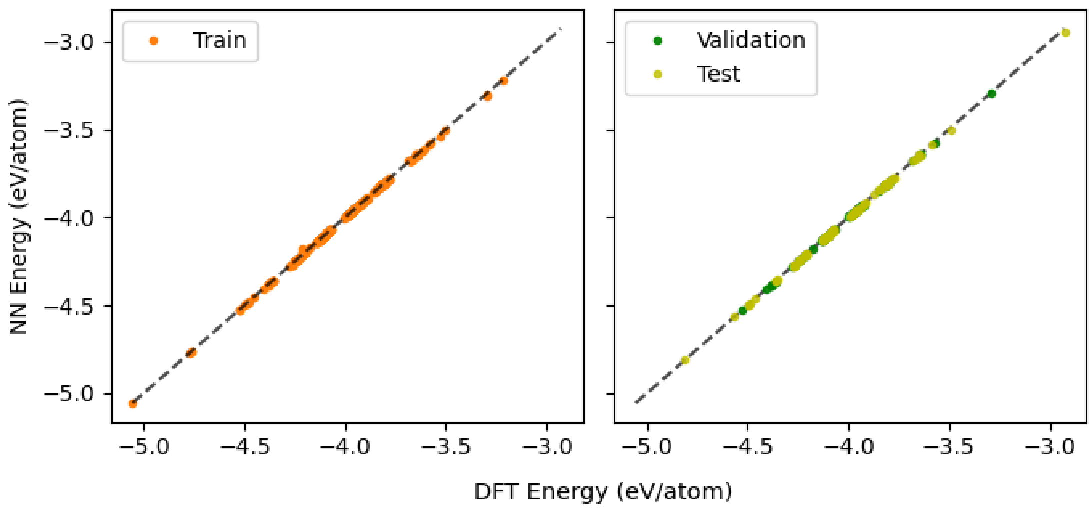

Figure 2: Figure 4

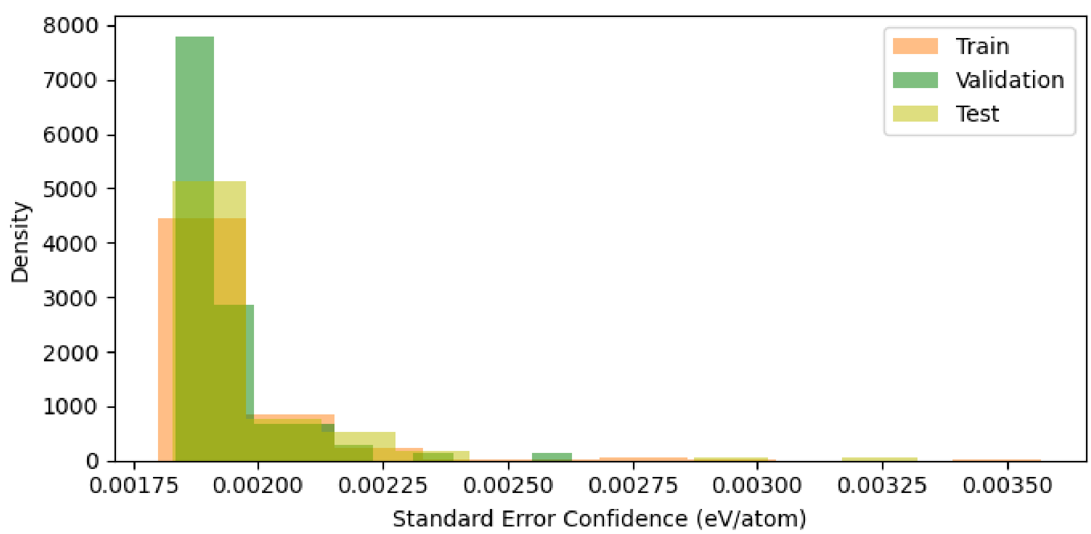

Figure 3: Figure 5

5 Fingerprints

```
import torch
from uncert import get_fps
import matplotlib.pyplot as plt

Name = 'lattice39-2'
train_dict = torch.load(f'{Name}/final_train.sav')
fp_train, e_mask_train = get_fps(f'{Name}/best_model', train_dict)
```

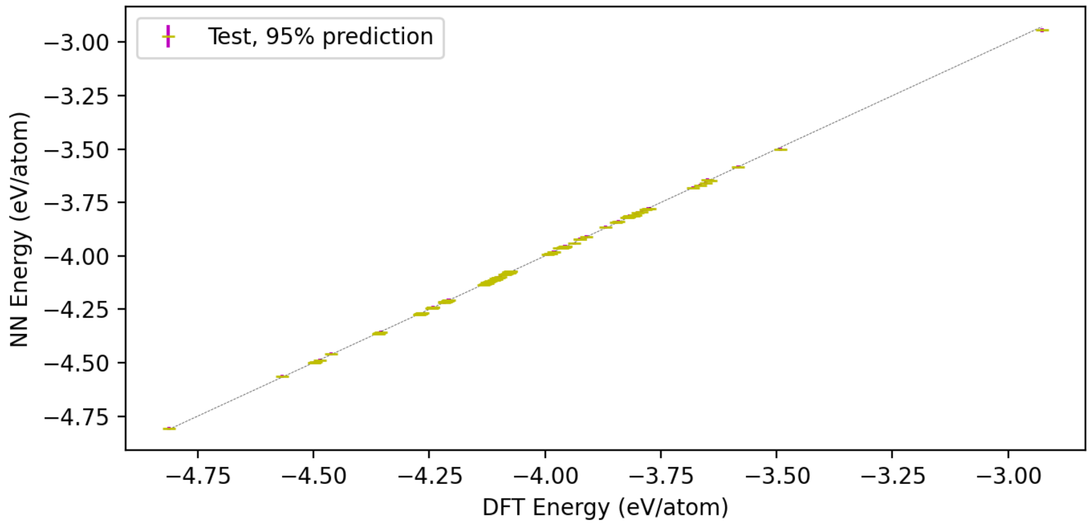

Figure 4: Figure 6

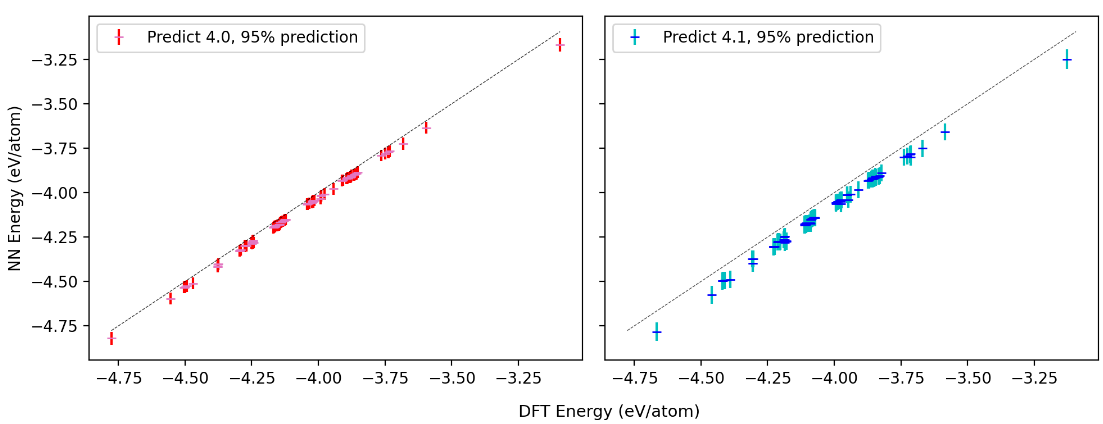

Figure 5: Figure 7

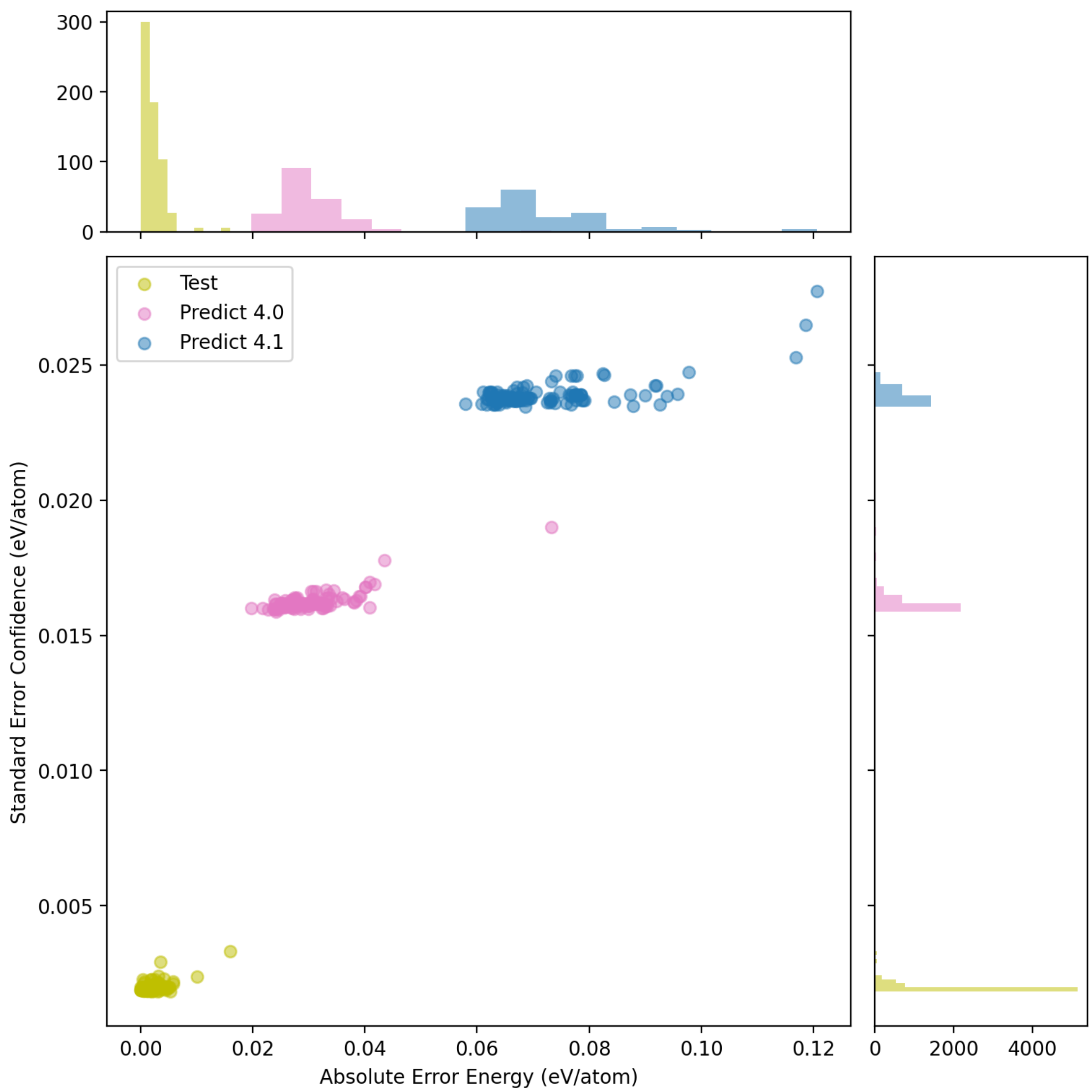

Figure 6: Figure 9

```

data_dict = torch.load(f'lattice40_pred-2/test.sav')
fp_40, e_mask_40 = get_fps(f'{Name}/best_model', data_dict)

data_dict = torch.load(f'lattice41_pred-2/test.sav')
fp_41, e_mask_41 = get_fps(f'{Name}/best_model', data_dict)

for i in range(2):
    for j in range(4):
        plt.clf()
        plt.hist(fp_train[e_mask_train[:,:,i]==1][:,j],alpha=0.5, density=True,label='Train', color='y')
        plt.hist(fp_40[e_mask_40[:,:,i]==1][:,j],alpha=0.5, density=True,label='Predict 4.0', color='tab:pink')

        plt.hist(fp_41[e_mask_41[:,:,i]==1][:,j],alpha=0.5, density=True,label='Predict 4.1')
        plt.xlabel('Fingerprint Value')
        plt.ylabel('Density')
        plt.legend()
        plt.tight_layout()
        plt.savefig(f'fps-hist-el{i}-fp{j}.png')

        print(f'''#+attr_org: :width 600
#+caption: fps-hist-el{i}-fp{j}
[[./fps-hist-el{i}-fp{j}.png]]''')
```

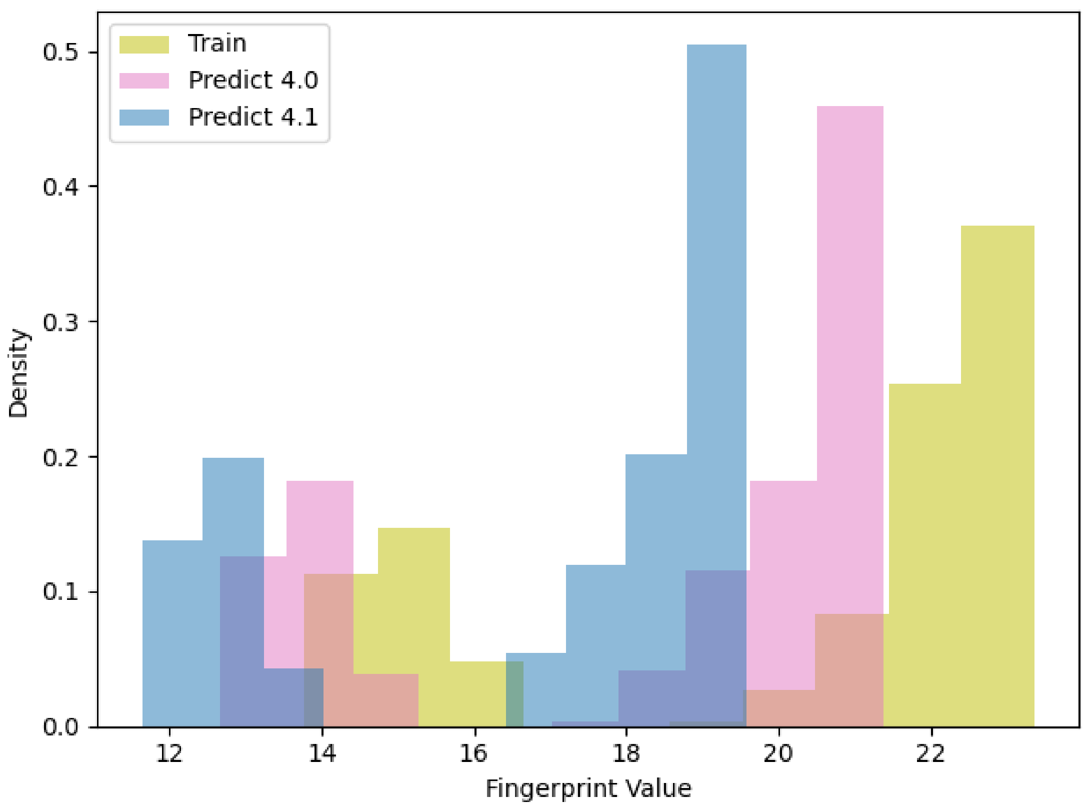

Figure 7: fps-hist-el0-fp0

6 Model retraining

The following code concatenates the original training-data with training portion of the predict-4.0 and 4.1 datasets. The code trains the potential and generates a directory folder

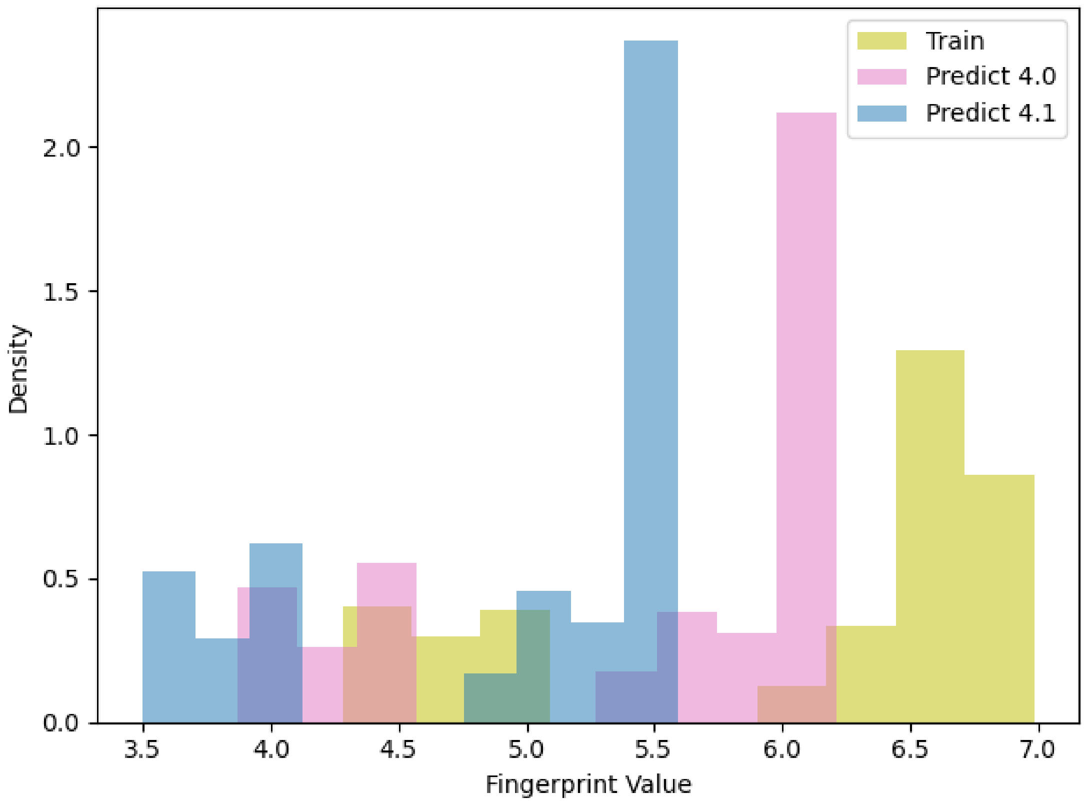

Figure 8: fps-hist-el0-fp1

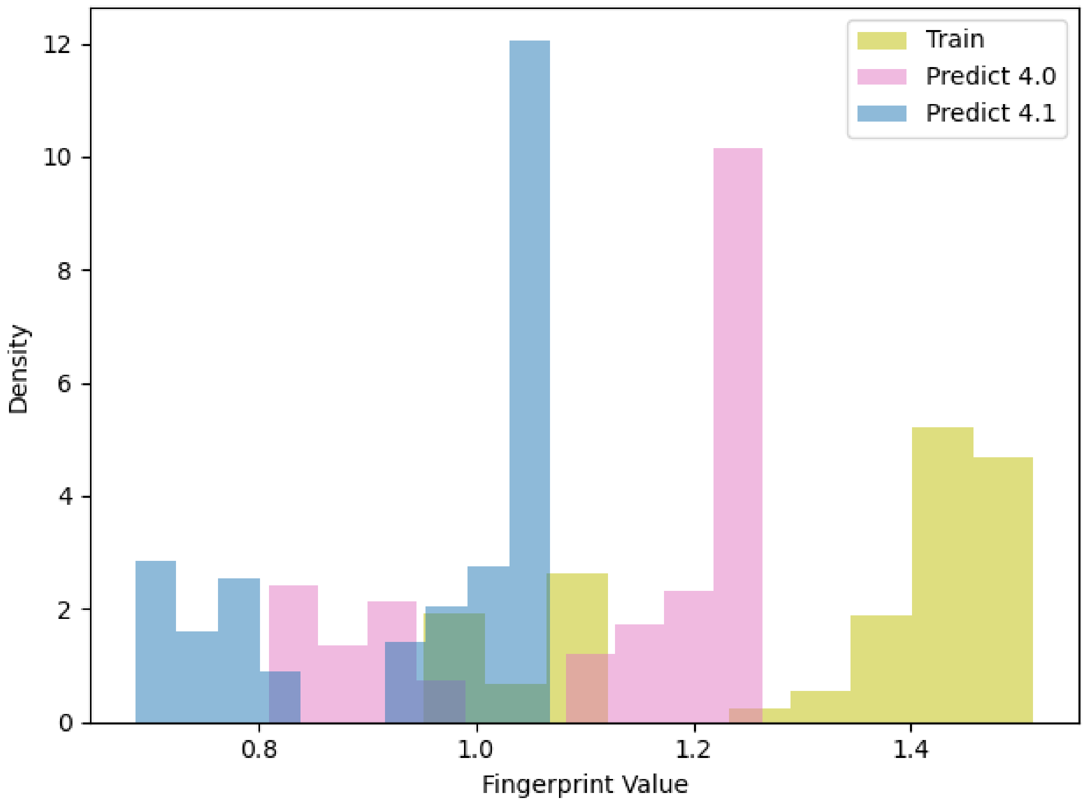

Figure 9: fps-hist-el0-fp2

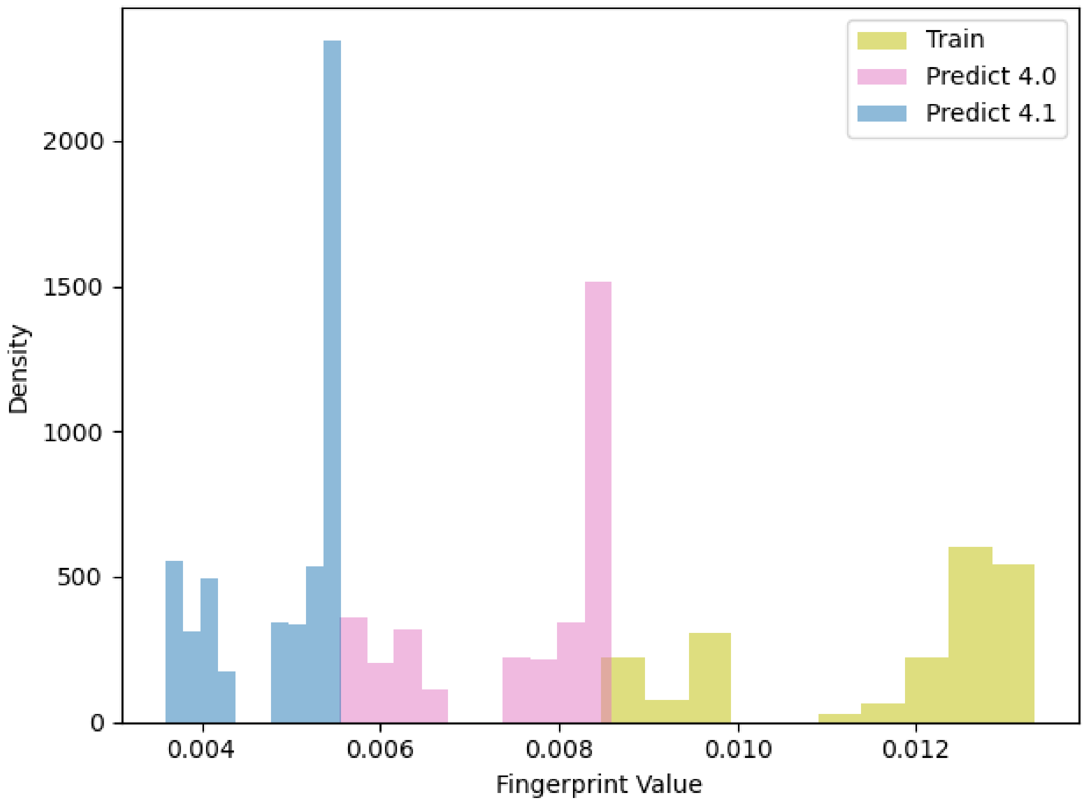

Figure 10: fps-hist-el0-fp3

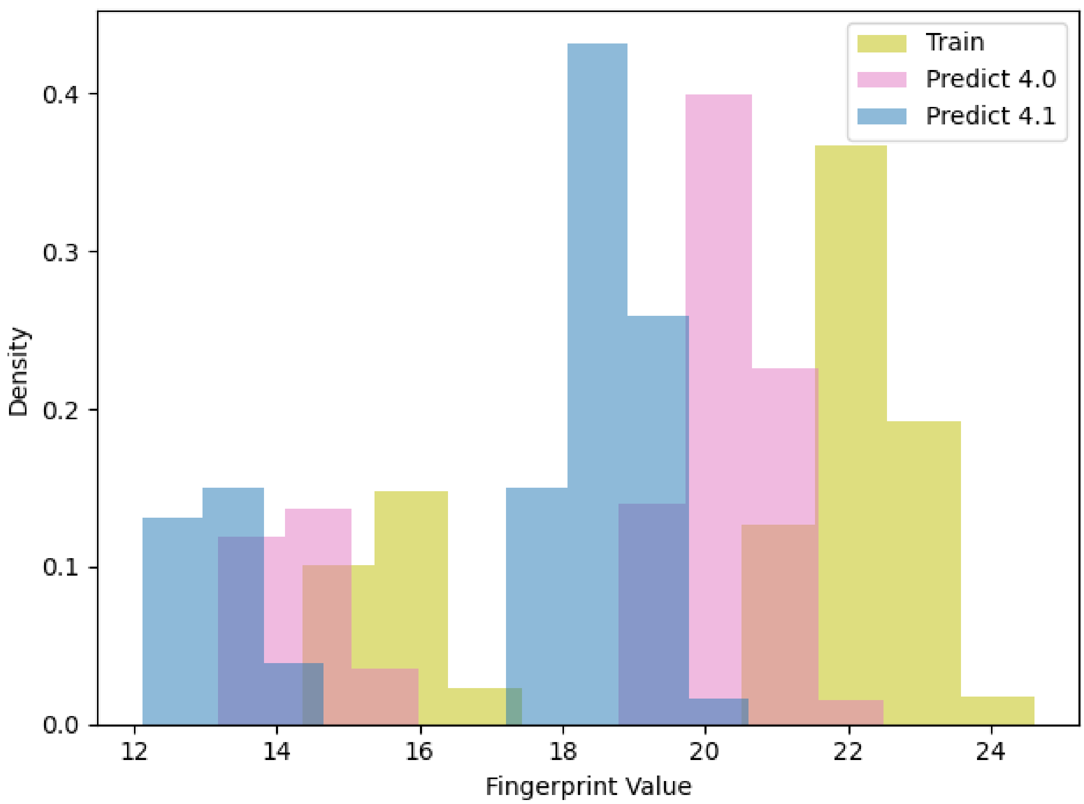

Figure 11: fps-hist-el1-fp0

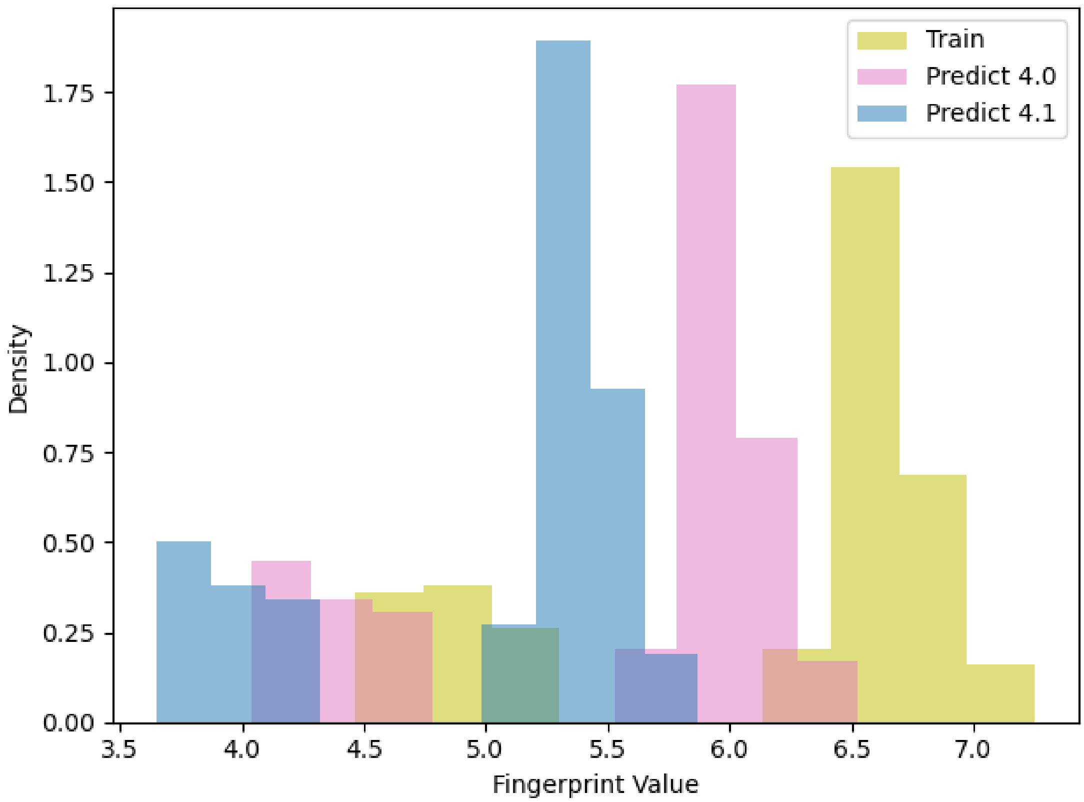

Figure 12: fps-hist-el1-fp1

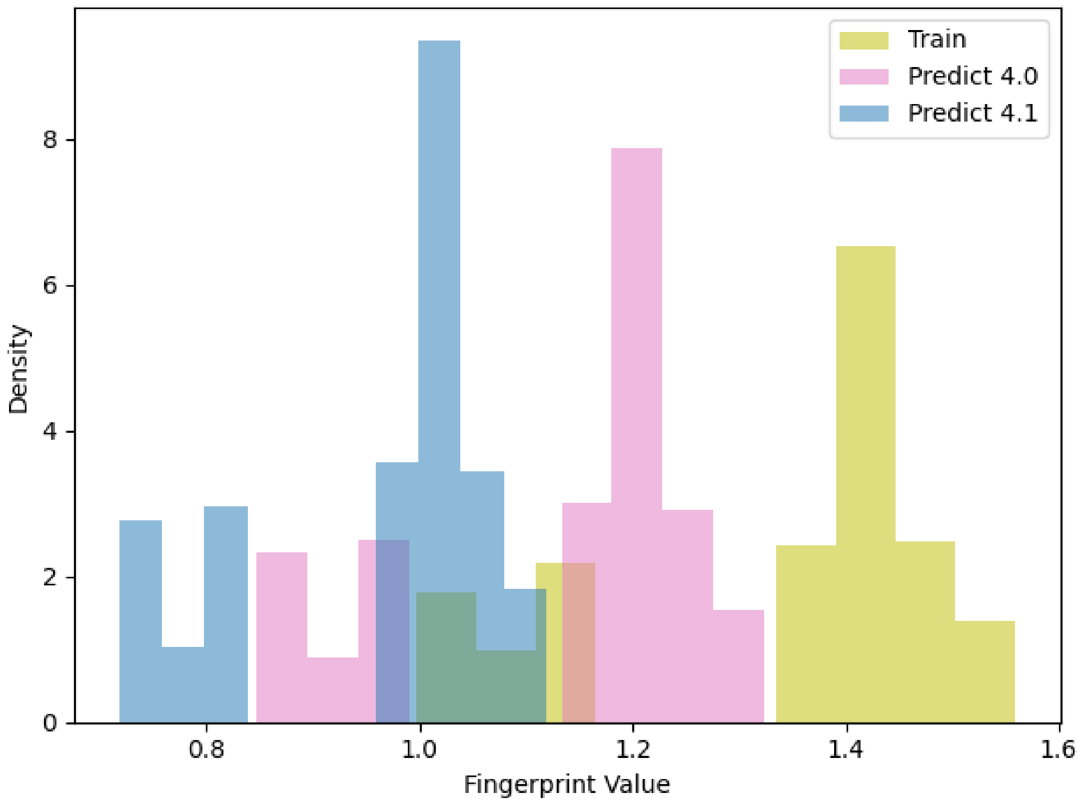

Figure 13: fps-hist-el1-fp2

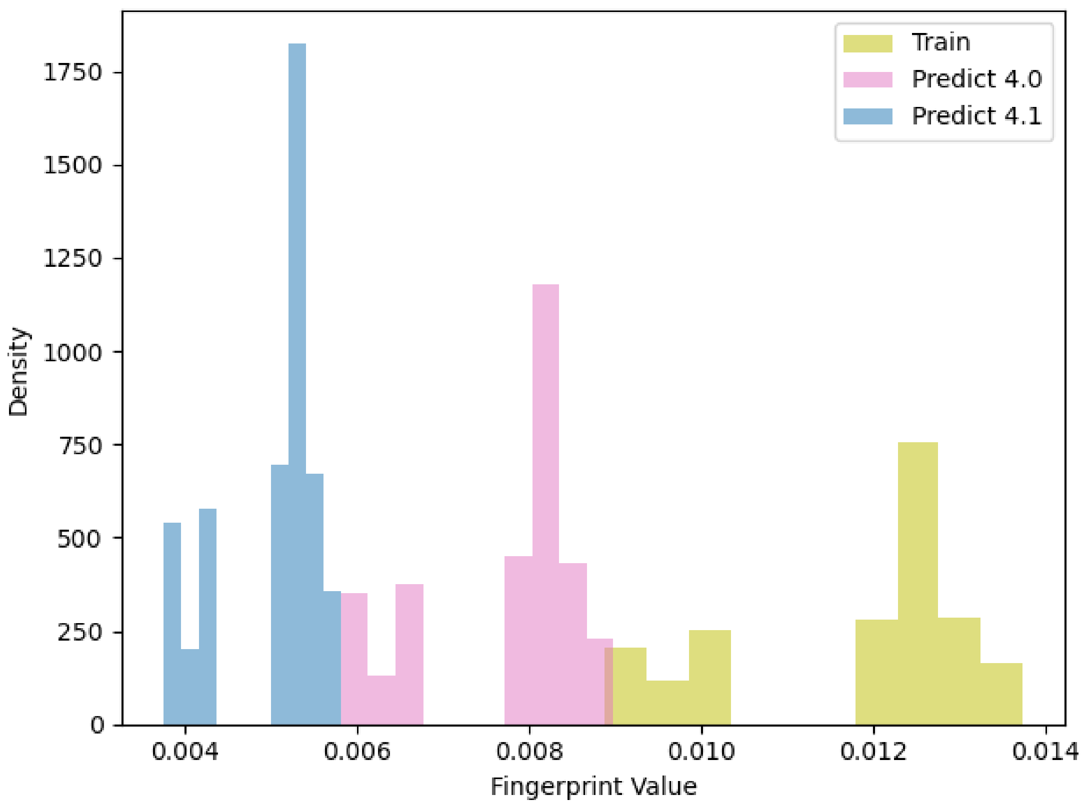

Figure 14: fps-hist-el1-fp3

"lattice39-40-41-2" with relevant files: concatenated dataset files "final_train.sav", "final_val.sav", "test.sav"; model file "best_model".

```python
import sys

sys.path.append("../SimpleNN")
sys.path.append("../")

import os
from ase.db import connect
import torch
from ContextManager import cd
from preprocess import train_test_split, train_val_split, get_scaling, CV
from preprocess import snn2sav
from NN import MultiLayerNet
from train import train, evaluate
from fp_calculator import set_sym, calculate_fp
import pickle

is_train = True
is_transfer = False
is_force = True

if is_train and is_transfer:
    raise ValueError('train and transfer could not be true at the same time.')

##################################################################################
#Hyperparameters
##################################################################################
E_coeff = 100
if is_force:
    F_coeff = 1
else:
    F_coeff = 0

val_interval = 10
n_val_stop = 10
epoch = 3000

opt_method = 'lbfgs'


if opt_method == 'lbfgs':
    history_size = 100
    lr = 1
    max_iter = 10
    line_search_fn = 'strong_wolfe'


convergence = {'E_cov':0.0005,'F_cov':0.005}

# min_max will scale fingerprints to (0,1)
fp_scale_method = 'min_max'
e_scale_method = 'min_max'


test_percent = 0.2
# Pecentage from train+val
val_percent = 0.2

# Training model configuration
SEED = [2]
n_nodes = [11,11]
activations = [torch.nn.Sigmoid(), torch.nn.Sigmoid()]

lr = 1
hp = {'n_nodes': n_nodes, 'activations': activations, 'lr': lr}
```

```python

################################################################################
#Configuration
################################################################################

elements = ['Pd', 'Au']
nelem = len(elements)

element_energy = None
weights =[1.58, 1.92]

Gs = [22]
cutoff = 6.35
g2_etas = [0.00, 0.10713, 0.285686, 0.892769]
g2_Rses = [0.0]

sym_params = [Gs, cutoff, g2_etas, g2_Rses, elements, weights, element_energy]
params_set = set_sym(elements, Gs, cutoff,
                         g2_etas=g2_etas, g2_Rses=g2_Rses,
                          weights=weights)
N_sym = params_set[elements[0]]['num']

################################################################################
#Training
################################################################################

Name = 'lattice39-40-41'

if is_train:
    for seed in SEED:
        # This use the context manager to operate in the data directory

        if not os.path.exists(Name+f'-{seed}'):
            os.makedirs(Name+f'-{seed}')

        with cd(Name+f'-{seed}'):
            pickle.dump(sym_params, open("sym_params.sav", "wb"))
            logfile = open('log.txt','w+')
            resultfile = open('result.txt','w+')

            if os.path.exists('test.sav'):
                logfile.write('Did not calculate symfunctions.\n')
            else:
                #this part is to concatenate the train-data subsets together.
                train_dict1 = torch.load('../lattice39-2/final_train.sav')
                train_dict2 = torch.load('../lattice40_pred-2/final_train.sav')
                train_dict3 = torch.load('../lattice41_pred-2/final_train.sav')
                train_dict = dict(train_dict1)
                new_dict = {k+1000: v for k, v in train_dict2.items()}
                train_dict.update(new_dict)
                new_dict = {k+2000: v for k, v in train_dict3.items()}
                train_dict.update(new_dict)

                val_dict1 = torch.load('../lattice39-2/final_val.sav')
                val_dict2 = torch.load('../lattice40_pred-2/final_val.sav')
                val_dict3 = torch.load('../lattice41_pred-2/final_val.sav')
                val_dict = dict(val_dict1)
                new_dict = {k+1000: v for k, v in val_dict2.items()}
                val_dict.update(new_dict)
                new_dict = {k+2000: v for k, v in val_dict3.items()}
                val_dict.update(new_dict)


                test_dict1 = torch.load('../lattice39-2/test.sav')
                test_dict2 = torch.load('../lattice40_pred-2/test.sav')
                test_dict3 = torch.load('../lattice41_pred-2/test.sav')
                test_dict = dict(test_dict1)
                new_dict = {k+1000: v for k, v in test_dict2.items()}
```

```
                test_dict.update(new_dict)
                new_dict = {k+2000: v for k, v in test_dict3.items()}
                test_dict.update(new_dict)



                torch.save(train_dict,'final_train.sav')
                torch.save(val_dict, 'final_val.sav')
                torch.save(test_dict, 'test.sav')

            scaling = get_scaling(train_dict, fp_scale_method, e_scale_method)

            n_nodes = hp['n_nodes']
            activations = hp['activations']
            lr = hp['lr']
            #model = torch.load('../lattice39-2/best_model')
            model = MultiLayerNet(N_sym, n_nodes, activations, nelem, scaling=scaling)
            if opt_method == 'lbfgs':
                optimizer = torch.optim.LBFGS(model.parameters(), lr=lr,
                                              max_iter=max_iter, history_size=history_size,
                                              line_search_fn=line_search_fn)

            results = train(train_dict, val_dict,
                            model,
                            opt_method, optimizer,
                            E_coeff, F_coeff,
                            epoch, val_interval,
                            n_val_stop,
                            convergence, is_force,
                            logfile)
            [loss, E_MAE, F_MAE, v_loss, v_E_MAE, v_F_MAE] = results

            test_results = evaluate(test_dict, E_coeff, F_coeff, is_force)
            [test_loss, test_E_MAE, test_F_MAE] =test_results
            resultfile.write(f'Hyperparameter: n_nodes = {n_nodes}, activations = {activations}, lr = {lr}\n')
            resultfile.write(f'loss = {loss}, E_MAE = {E_MAE}, F_MAE = {F_MAE}.\n')
            resultfile.write(f'v_loss = {v_loss}, v_E_MAE = {v_E_MAE}, v_F_MAE = {v_F_MAE}.\n')
            resultfile.write(f'test_loss = {test_loss}, test_E_MAE = {test_E_MAE}, test_F_MAE = {test_F_MAE}.\n')


            logfile.close()
            resultfile.close()
```

7 Uncertainty for retrained model

```
import torch
from uncert import evaluate_uncert
import numpy as np
import matplotlib.pyplot as plt
from scipy.stats.distributions import t
from Batch import batch_pad

#get inverse fisher information
def get_pcov(h):
    eigs0 = np.linalg.eigvalsh(h)[0]
    if (eigs0 <0):
        eps = max(1e-5, eigs0*-1.05)
    else:
        eps = 1e-5
    j = np.linalg.pinv(h + eps*np.identity(h.shape[0]))
    pcov1 = j*alpha
    u, v = np.linalg.eigh(pcov1)
    return v @ np.diag(np.maximum(u,0)) @ v.T


def flatten_gprime(agrad):
```

```
    cnt = 0
    for g in agrad:
        g_vector = g.contiguous().view(-1) if cnt ==0 else torch.cat([g_vector, g.contiguous().view(-1)])
        cnt = 1
    return g_vector

#get uncertainties for a dataset
def get_uncerts(name, data_dict):
    model = torch.load(name)
    scaling = model.scaling
    gmin = scaling['gmin']
    gmax = scaling['gmax']
    emin = scaling['emin']
    emax = scaling['emax']

    ids = np.array(list(data_dict.keys()))
    batch_info = batch_pad(data_dict,ids)
    b_fp = batch_info['b_fp']

    b_e_mask = batch_info['b_e_mask']
    b_fp.requires_grad = True
    sb_fp = (b_fp - gmin) / (gmax - gmin)

    N_atoms = batch_info['N_atoms'].view(-1)
    b_e = batch_info['b_e'].view(-1)
    b_f = batch_info['b_f']

    Atomic_Es = model(sb_fp)
    E_predict = torch.sum(Atomic_Es * b_e_mask, dim = [1,2])
    E_predict = E_predict/N_atoms
    E_predict = E_predict * (emax - emin) + emin

    uncerts = []
    for i, ei in enumerate(E_predict):
        gprime = torch.autograd.grad(ei, model.parameters(), create_graph=True, retain_graph=True)
        gprime = flatten_gprime(gprime).detach().numpy()
        se = gprime @ pcov @ gprime
        uncerts += [(np.sqrt(se), np.sqrt(se + rmse.item()**2), np.linalg.norm(gprime))]
    uncerts = np.array(uncerts)
    return uncerts

Name = 'lattice39-40-41-2'

train_dict = torch.load(f'{Name}/final_train.sav')
val_dict = torch.load(f'{Name}/final_val.sav')
test_dict = torch.load(f'{Name}/test.sav')

pred_e, actual_e, rmse, h = evaluate_uncert(f'{Name}/best_model',train_dict, True)
h = h.detach().numpy()
pred_e_val, actual_e_val, rmse_val = evaluate_uncert(f'{Name}/best_model',val_dict, False)
pred_e_test, actual_e_test, rmse_test = evaluate_uncert(f'{Name}/best_model',test_dict, False)

ndata = pred_e.shape[0]
alpha = rmse.item()**2
pcov = get_pcov(h)

uncerts_val = get_uncerts(f'{Name}/best_model',val_dict)
uncerts_train = get_uncerts(f'{Name}/best_model',train_dict)
uncerts_test = get_uncerts(f'{Name}/best_model',test_dict)


####################################################################################
#Parity plot after retraining
####################################################################################

data_dict = torch.load(f'lattice40_pred-2/test.sav')
pred_e_40p, actual_e_40p, rmse_40p = evaluate_uncert(f'{Name}/best_model',data_dict, False)
uncerts_40p = get_uncerts(f'{Name}/best_model',data_dict)
```

```python

data_dict = torch.load(f'lattice41_pred-2/test.sav')
pred_e_41p, actual_e_41p, rmse_41p = evaluate_uncert(f'{Name}/best_model',data_dict, False)
uncerts_41p = get_uncerts(f'{Name}/best_model',data_dict)

tval = t.ppf(0.975, ndata)
plt.clf()

fig, ax = plt.subplots(ncols =2, nrows=1, sharex=False, sharey='row')
fig.set_size_inches(10, 4)
ax[0].set_title(' ')
ax[0].errorbar(actual_e, pred_e, yerr = tval * uncerts_train[:,1], fmt = 'y_', ecolor='m',
               label='Train, 95% prediction')
ax[0].set_xlabel(' ')
ax[0].set_ylabel('NN Energy (eV/atom)')
ax[0].legend()
eline = np.linspace(np.min(actual_e), np.max(actual_e_40p), 10)
ax[0].plot(eline, eline,'k--', alpha=0.7, linewidth=0.3)

ax[1].errorbar(actual_e_40p, pred_e_40p, yerr = tval * uncerts_40p[:,1], color='tab:pink',
               fmt = '_', ecolor='b', label='Predict 4.0, 4.1, 95% prediction')
ax[1].errorbar(actual_e_41p, pred_e_41p, yerr =  tval * uncerts_41p[:,1], color='tab:pink',
               fmt = '_', ecolor='b', label='')
ax[1].legend()
ax[1].plot(eline, eline,'k--', alpha=0.7, linewidth=0.3)

plt.figtext(0.55,0.03,"DFT Energy (eV/atom)", va="center", ha="center", size=10.5)

plt.tight_layout()
plt.savefig('subplot-parity-40-41-pot2-pred-v2.png', dpi=200)
print('''#+attr_org: :width 600
#+caption: Figure 10
[[./subplot-parity-40-41-pot2-pred-v2.png]]
''')

##################################################################################
#Uncertainty vs True Error Scatterplot
##################################################################################

def scatter_hist(x, y, ax, ax_histx, ax_histy, label, color=None):
    # no labels
    ax_histx.tick_params(axis="x", labelbottom=False)
    ax_histy.tick_params(axis="y", labelleft=False)

    # the scatter plot:
    ax.scatter(x, y, alpha=0.5, label=label, color=color)

    # now determine nice limits by hand:
    binwidth = 0.0001
    xymax = max(np.max(np.abs(x)), np.max(np.abs(y)))
    lim = (int(xymax/binwidth)+1)*binwidth

    #bins = np.arange(0, lim + binwidth, binwidth)
    ax_histx.hist(x,  alpha=0.5, color=color, density=True)
    ax_histy.hist(y,  orientation='horizontal', alpha=0.5, color=color, density=True)

fig = plt.figure(figsize=(8, 8))
# Add a gridspec with two rows and two columns and a ratio of 2 to 7 between
# the size of the marginal axes and the main axes in both directions.
# Also adjust the subplot parameters for a square plot.
gs = fig.add_gridspec(2, 2,  width_ratios=(7, 2), height_ratios=(2, 7),
                      left=0.11, right=0.98, bottom=0.07, top=0.97, wspace=0.05, hspace=0.05)

ax = fig.add_subplot(gs[1, 0])
ax_histx = fig.add_subplot(gs[0, 0], sharex=ax)
ax_histy = fig.add_subplot(gs[1, 1], sharey=ax)

# use the previously defined function
```

```python
scatter_hist(np.absolute(actual_e-pred_e),  uncerts_train[:,0], ax, ax_histx, ax_histy,
             'Train', 'y')
scatter_hist(np.absolute(actual_e_40p-pred_e_40p), uncerts_40p[:,0], ax, ax_histx, ax_histy,
             'Predict 4.0', 'tab:pink')
scatter_hist(np.absolute(pred_e_41p-actual_e_41p), uncerts_41p[:,0], ax, ax_histx, ax_histy,
             'Predict 4.1')

ax.set_xlabel('Absolute Error Energy (eV/atom)')
ax.set_ylabel('Standard Error Confidence (eV/atom)')
ax.legend()
plt.savefig('uncert-v-error-w-hist-ret40-41-orig.png', dpi=200,bbox_inches='tight')
print('''
#+attr_org: :width 600
#+caption: Figure 11
[[./uncert-v-error-w-hist-ret40-41-orig.png]]
''')
```

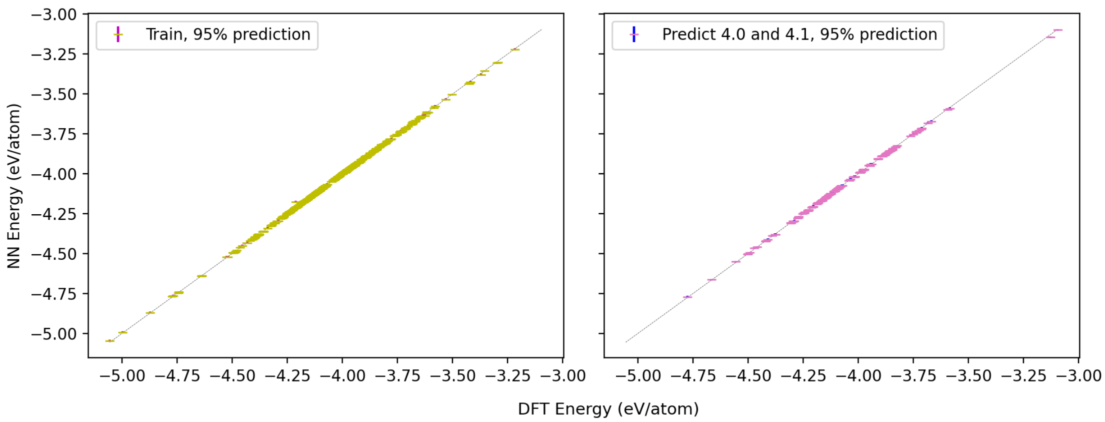

Figure 15: Figure 10

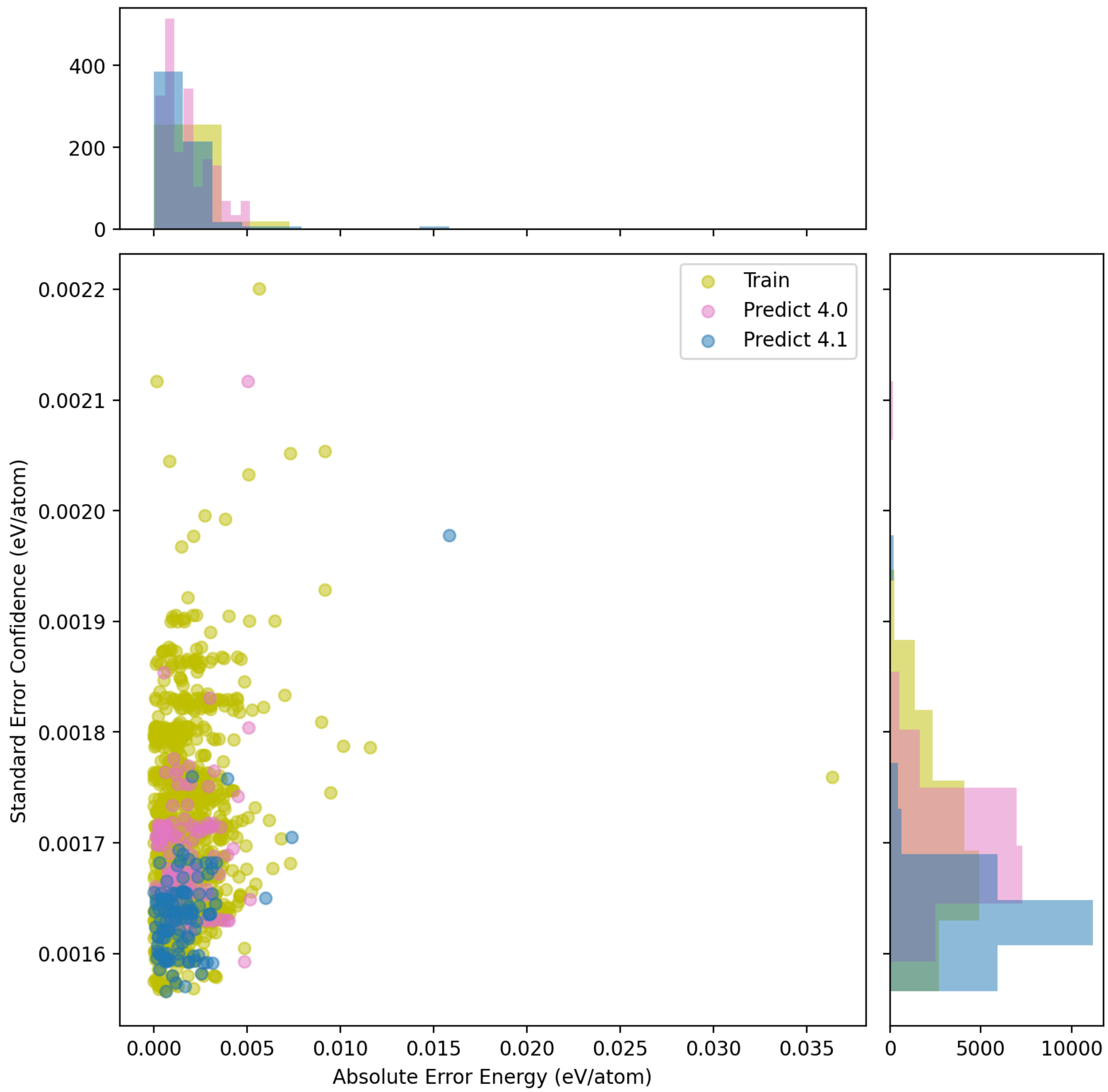

Figure 16: Figure 11