

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

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# 1 One dimension input NN (Figure 2)

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```
1 import autograd
2 import autograd.numpy as np
3 from autograd import hessian
4 import matplotlib.pyplot as plt
5 from autograd import grad
6 import autograd.numpy.random as npr
7 from scipy.stats.distributions import t
8 from scipy.optimize import minimize
9
10 # lennard jones potential
11 def func(x, e, s):
12     return 4 * e * (np.power(np.divide(s, x), 12) -
13                    np.power(np.divide(s, x), 6))
14
15 etrue = 10
16 strue = 0.34
17 numpts = 23
18
19 # xfit is for plotting
20 xfit = np.arange(0.34, 0.49, 0.001)
21 xfit = np.expand_dims(xfit, axis=1)
22
23 # weightsparser to help roll and unroll weights and biases.
24 class WeightsParser(object):
25     """A helper class to index into a parameter vector."""
26
27     def __init__(self):
28         self.idx_and_shapes = {}
29         self.N = 0
30
31     def add_weights(self, name, shape):
32         start = self.N
33         self.N += np.prod(shape)
34         self.idx_and_shapes[name] = (slice(start, self.N), shape)
35
36     def get(self, vect, name):
37         idxs, shape = self.idx_and_shapes[name]
38         return np.reshape(vect[idxs], shape)
39
40 # params is a 1-d vector of weights and biases
41 # parser is object that makes it easy to unroll params into matrices of
42 # weights and biases.
43 def init_random_params(scale, layer_sizes, rs=None):
44     if rs is None:
45         rs = npr.RandomState(2)
46     parser = WeightsParser()
47     for i, shape in enumerate(zip(layer_sizes[:-1], layer_sizes[1:])):
48         parser.add_weights(('weights', i), shape)
49         parser.add_weights(('biases', i), (1, shape[1]))
50     return rs.randn(parser.N), parser
51
52 # nn predict by unrolling w parser.
53 def nn_predict(params, inputs, nonlinearity=np.tanh):
54     cur_units = inputs
55     for layer in range(len(layer_sizes) - 1):
56         cur_W = parser.get(params, ('weights', layer))
57         cur_B = parser.get(params, ('biases', layer))
58         cur_units = np.dot(cur_units, cur_W) + cur_B
59         if layer < len(layer_sizes) - 2:
60             cur_units = nonlinearity(cur_units)
61     return cur_units
62
63 # objective with regularization to be used with scipy minimize
64 def objective12(params, X, r, alpha=0):
65     ypredict = nn_predict(params, X)
```

```

66     errs = r - ypredict
67     weights = params[idxs]
68     return np.sum(errs**2) + alpha * np.linalg.norm(weights)
69
70 layer_sizes = [1, 4, 1]
71 _, parser = init_random_params(1, layer_sizes)
72
73 # get the index of the weights, because only regularizing weights.
74 idxs = []
75 for layer in range(len(layer_sizes) - 1):
76     sliceidx, _ = parser.idxs_and_shapes[('weights', layer)]
77     idxs += [np.r_[sliceidx]]
78 idxs = np.array(idxs).flatten()
79
80 #sum-squared-errors
81 def sse(params, X, r):
82     ypredict = nn_predict(params, X)
83     errs = r - ypredict
84     return np.sum(errs**2)
85
86 #get inverse fisher information
87 def get_pcov(h):
88     eigs0 = np.linalg.eigvalsh(h)[0]
89     if (eigs0 < 0):
90         eps = max(1e-5, eigs0*-1.05)
91     else:
92         eps = 1e-5
93     j = np.linalg.pinv(h + eps * np.identity(h.shape[0]))
94     pcov1 = j * scaling
95     u, v = np.linalg.eigh(pcov1)
96     return v @ np.diag(np.maximum(u, 0)) @ v.T
97
98 #get standard errors of prediction, confidence
99 def getpredse(x, params):
100     gprime = autograd.elementwise_grad(nn_predict, 0)(params, x)
101     sesq = gprime @ pcov @ gprime
102     return np.sqrt(sesq), np.sqrt(sesq + scaling)
103
104 #get standard errors for a dataset
105 def get_se_dataset(xfit, params):
106     predsese = []
107     for i in xfit:
108         predsese += [getpredse(i, params)]
109     return np.array(predsese)
110
111 # to make plot
112 # data for panel 1.
113 numpts = 23
114 xa = np.linspace(0.35, 0.45, numpts)
115 np.random.seed(seed=0)
116
117 ya = func(xa, etrue, strue) + np.random.normal(scale=0.2, size=xa.shape)
118
119 Xa = np.expand_dims(xa, axis=1)
120 ra = np.expand_dims(ya, axis=1)
121
122 initial_guess, parser = init_random_params(1, layer_sizes)
123
124 sol = minimize(objective12, initial_guess, args=(Xa, ra, 0.01) )
125 paramsa = sol.x
126
127 h = hessian(sse, 0)(paramsa, Xa, ra)
128 numpts_a = Xa.shape[0]
129 scaling = sse(paramsa, Xa, ra)/numpts_a
130
131 pcov = get_pcov(h)
132
133 predsese_a = get_se_dataset(xfit, paramsa)

```


```

134
135 #data for panel 2.
136 x1 = np.linspace(0.35, 0.365, 7)
137 x2 = np.linspace(0.415, 0.45, 9)
138 xb = np.concatenate((x1,x2))
139 yb = func(xb, etrue, strue) + np.random.normal(scale=0.2, size=xb.shape)
140
141 Xb = np.expand_dims(xb, axis=1)
142 rb = np.expand_dims(yb, axis=1)
143
144 initial_guess, _ = init_random_params(1, layer_sizes)
145
146 sol = minimize(objectivel2, initial_guess, args=(Xb,rb,0.005) )
147 paramsb = sol.x
148
149 h = hessian(sse,0)(paramsb, Xb, rb)
150 numptsb = Xb.shape[0]
151 scaling = sse(paramsb, Xb, rb)/numptsb
152
153 pcov = get_pcov(h)
154
155 predsesb = get_se_dataset(xfit, paramsb)
156
157
158 #make a plot.
159 plt.clf()
160 fig, ax = plt.subplots(ncols=2, nrows = 1, sharex=False, sharey='row')
161 fig.set_size_inches(7,3)
162 tvala = t.ppf(0.975, numptsb)
163 tvalb = t.ppf(0.975, numptsb)
164
165 ypreda = nn_predict(paramsa, xfit).flatten()
166 ypredb = nn_predict(paramsb, xfit).flatten()
167
168 ax[0].set_title(' ')
169 ax[0].plot(Xa, ra, 'bo')
170 ax[0].plot(xfit, ypreda)
171 ax[0].plot(xfit, func(xfit, etrue, strue))
172 ax[0].plot(xfit, ypreda + predsesa[:,0] * tvala, '--r')
173 ax[0].plot(xfit, ypreda - predsesa[:,0] * tvala, '--r')
174 ax[0].set_xlabel('x')
175 ax[0].set_ylabel('y')
176
177 ax[1].set_title(' ')
178 ax[1].plot(Xb, rb, 'bo')
179 ax[1].plot(xfit, ypredb)
180 ax[1].plot(xfit, func(xfit, etrue, strue))
181 ax[1].plot(xfit, ypredb + predsesb[:,0] * tvalb, '--r')
182 ax[1].plot(xfit, ypredb - predsesb[:,0] * tvalb, '--r')
183 ax[1].set_xlabel('x')
184
185 ax[1].legend(['Data', 'NN', 'f(x)', '95% confidence'])
186
187 plt.subplots_adjust(wspace=0)
188 plt.tight_layout()
189
190 plt.savefig(f'subplot-2panel.png', dpi=200)
191 print(''''#+attr_org: :width 600
192 #+caption: Figure 2
193 [./subplot-2panel.png]''')

```

---

## 2 Training a SingleNN model

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

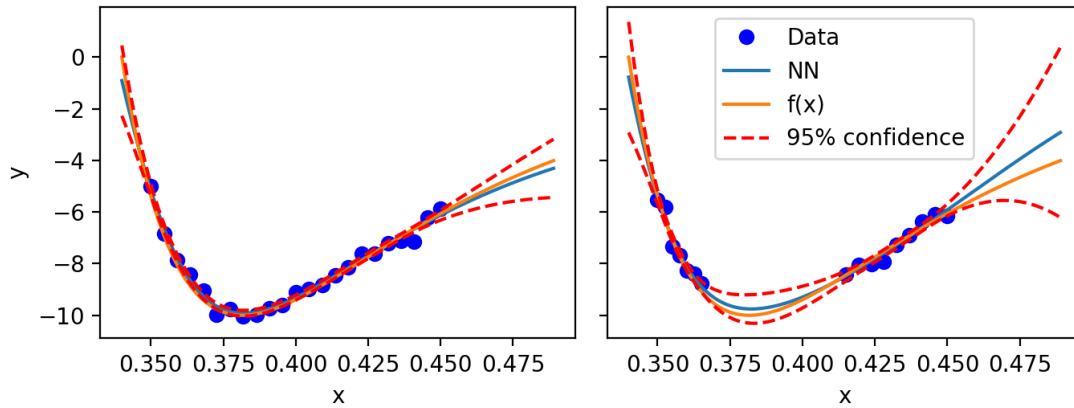


Figure 1: Figure 2

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".

---

```

1  import sys
2
3  sys.path.append("../SimpleNN")
4  sys.path.append("../")
5
6  import os
7  from ase.db import connect
8  import torch
9  from ContextManager import cd
10 from preprocess import train_test_split, train_val_split, get_scaling, CV
11 from preprocess import snn2sav
12 from NN import MultiLayerNet
13 from train import train, evaluate
14 from fp_calculator import set_sym, calculate_fp
15 import pickle
16
17 is_train = True
18 is_transfer = False
19 is_force = True
20
21 if is_train and is_transfer:
22     raise ValueError('train and transfer could not be true at the same time.')
23
24 #####
25 #Hyperparameters
26 #####
27 E_coeff = 100
28 if is_force:
29     F_coeff = 1
30 else:
31     F_coeff = 0
32
33 val_interval = 10
34 n_val_stop = 10
35 epoch = 3000

```

```

36
37 opt_method = 'lbfgs'
38
39
40 if opt_method == 'lbfgs':
41     history_size = 100
42     lr = 1
43     max_iter = 10
44     line_search_fn = 'strong_wolfe'
45
46
47 convergence = {'E_cov':0.0005, 'F_cov':0.005}
48
49 # min_max will scale fingerprints to (0,1)
50 fp_scale_method = 'min_max'
51 e_scale_method = 'min_max'
52
53
54 test_percent = 0.2
55 # Percentage from train+val
56 val_percent = 0.2
57
58 # Training model configuration
59 SEED = [2]
60 n_nodes = [11,11]
61 activations = [torch.nn.Sigmoid(), torch.nn.Sigmoid()]
62 lr = 1
63 hp = {'n_nodes': n_nodes, 'activations': activations, 'lr': lr}
64
65 #####
66 #Configuration
67 #####
68
69 if is_train:
70     # The Name of the training
71     Name = f'lattice39'
72     for seed in SEED:
73         if not os.path.exists(Name+f'-' + str(seed)):
74             os.makedirs(Name+f'-' + str(seed))
75
76     dbfile = f'data/lattice39.db'
77     db = connect(dbfile)
78
79     elements = ['Pd', 'Au']
80     nelem = len(elements)
81     # This is the energy of the metal in its ground state structure
82     #if you don't know the energy of the ground state structure,
83     # you can set it to None
84     element_energy = None
85     # Allen electronegativity
86     weights = [1.58, 1.92]
87
88
89     Gs = [22]
90     cutoff = 6.35
91     g2_etas = [0.00, 0.10713, 0.285686, 0.892769]
92     g2_Rses = [0.0]
93
94
95     sym_params = [Gs, cutoff, g2_etas, g2_Rses, elements, weights, element_energy]
96     params_set = set_sym(elements, Gs, cutoff,
97                          g2_etas=g2_etas, g2_Rses=g2_Rses,
98                          weights=weights)
99
100     N_sym = params_set[elements[0]]['num']
101
102 #####
103 #Training



```

```

104 #####
105
106 Name = f'lattice39'
107 if is_train:
108     for seed in SEED:
109         # This use the context manager to operate in the data directory
110         with cd(Name+f'_{seed}'):
111             pickle.dump(sym_params, open("sym_params.sav", "wb"))
112             logfile = open('log.txt', 'w+')
113             resultfile = open('result.txt', 'w+')
114
115             if os.path.exists('test.sav'):
116                 logfile.write('Did not calculate symfunctions.\n')
117             else:
118                 data_dict = snn2sav(db, Name, elements, params_set,
119                                     element_energy=element_energy)
120                 train_dict = train_test_split(data_dict, 1-test_percent, seed=seed)
121                 train_val_split(train_dict, 1-val_percent, seed=seed)
122
123                 logfile.flush()
124
125                 train_dict = torch.load('final_train.sav')
126                 val_dict = torch.load('final_val.sav')
127                 test_dict = torch.load('test.sav')
128                 scaling = get_scaling(train_dict, fp_scale_method, e_scale_method)
129
130
131                 n_nodes = hp['n_nodes']
132                 activations = hp['activations']
133                 lr = hp['lr']
134                 model = MultiLayerNet(N_sym, n_nodes, activations, nelelem, scaling=scaling)
135                 if opt_method == 'lbfgs':
136                     optimizer = torch.optim.LBFGS(model.parameters(), lr=lr,
137                                                    max_iter=max_iter, history_size=history_size,
138                                                    line_search_fn=line_search_fn)
139
140                 results = train(train_dict, val_dict,
141                                model,
142                                opt_method, optimizer,
143                                E_coeff, F_coeff,
144                                epoch, val_interval,
145                                n_val_stop,
146                                convergence, is_force,
147                                logfile)
148                 [loss, E_MAE, F_MAE, v_loss, v_E_MAE, v_F_MAE] = results
149
150                 test_results = evaluate(test_dict, E_coeff, F_coeff, is_force)
151                 [test_loss, test_E_MAE, test_F_MAE] = test_results
152                 resultfile.write(f'Hyperparameter: n_nodes = {n_nodes}, activations = {activations}, lr = {lr}\n')
153                 resultfile.write(f'loss = {loss}, E_MAE = {E_MAE}, F_MAE = {F_MAE}.\n')
154                 resultfile.write(f'v_loss = {v_loss}, v_E_MAE = {v_E_MAE}, v_F_MAE = {v_F_MAE}.\n')
155                 resultfile.write(f'test_loss = {test_loss}, test_E_MAE = {test_E_MAE}, test_F_MAE = {test_F_MAE}.\n')
156
157
158                 logfile.close()
159                 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".

---

```

1  import sys
2
3  sys.path.append("../SimpleNN")
4  sys.path.append("../")
5
6  import os
7  from ase.db import connect
8  from ContextManager import cd
9  from preprocess import train_test_split, train_val_split, get_scaling, CV
10 from preprocess import snn2sav
11 from fp_calculator import set_sym, calculate_fp
12
13
14 # min_max will scale fingerprints to (0,1)
15 fp_scale_method = 'min_max'
16 e_scale_method = 'min_max'
17
18
19 test_percent = 0.2
20 # Percentage from train+val
21 val_percent = 0.2
22
23 # Training model configuration
24 SEED = [2]
25
26 #####
27 #Split Predict-4.0 dataset
28 #####
29
30
31 Name = f'lattice40_pred'
32
33 for seed in SEED:
34     if not os.path.exists(Name+f'-{seed}'):
35         os.makedirs(Name+f'-{seed}')
36
37 dbfile = 'data/lattice40.db'
38 db = connect(dbfile)
39
40 elements = ['Pd', 'Au']
41 nelem = len(elements)
42
43 element_energy = None
44 weights = [1.58, 1.92]
45
46 Gs = [22]
47 cutoff = 6.35
48 g2_etas = [0.00, 0.10713, 0.285686, 0.892769]
49 g2_Rses = [0.0]
50
51
52 sym_params = [Gs, cutoff, g2_etas, g2_Rses, elements, weights, element_energy]
53 params_set = set_sym(elements, Gs, cutoff,
54                      g2_etas=g2_etas, g2_Rses=g2_Rses,
55                      weights=weights)
56 N_sym = params_set[elements[0]]['num']
57
58 with cd(Name+f'-{seed}'):
59     data_dict = snn2sav(db, Name, elements, params_set,
60                       element_energy=element_energy)
61
62     train_dict = train_test_split(data_dict, 1-0.2, seed=seed)
63     train_val_split(train_dict, 1-0.2, seed=seed)
64
65 #####
66 #Split Predict-4.1 dataset
67 #####

```



```

68
69 Name = f'lattice41_pred'
70
71 for seed in SEED:
72     if not os.path.exists(Name+f'--{seed}'):
73         os.makedirs(Name+f'--{seed}')
74
75 dbfile = 'data/lattice41.db'
76 db = connect(dbfile)
77
78 with cd(Name+f'--{seed}'):
79     data_dict = snn2sav(db, Name, elements, params_set,
80                       element_energy=element_energy)
81
82     train_dict = train_test_split(data_dict,1-0.2,seed=seed)
83     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: .

---

```

1  import torch
2  from uncert import evaluate_uncert
3  import numpy as np
4  import matplotlib.pyplot as plt
5  from scipy.stats.distributions import t
6  from Batch import batch_pad
7
8  #get inverse fisher information
9  def get_pcov(h):
10     eigs0 = np.linalg.eigvalsh(h)[0]
11     if (eigs0 < 0):
12         eps = max(1e-5, eigs0*-1.05)
13     else:
14         eps = 1e-5
15     j = np.linalg.pinv(h + eps*np.identity(h.shape[0]))
16     pcov1 = j*alpha
17     u, v = np.linalg.eigh(pcov1)
18     return v @ np.diag(np.maximum(u,0)) @ v.T
19
20
21 def flatten_gprime(agrad):
22     cnt = 0
23     for g in agrad:
24         g_vector = g.contiguous().view(-1) if cnt ==0 else torch.cat([g_vector, g.contiguous().view(-1)])
25         cnt = 1
26     return g_vector
27
28 #get uncertainties for a dataset
29 def get_uncerts(name, data_dict):
30     model = torch.load(name)
31     scaling = model.scaling
32     gmin = scaling['gmin']
33     gmax = scaling['gmax']
34     emin = scaling['emin']
35     emax = scaling['emax']
36
37     ids = np.array(list(data_dict.keys()))
38     batch_info = batch_pad(data_dict,ids)
39     b_fp = batch_info['b_fp']
40
41     b_e_mask = batch_info['b_e_mask']
42     b_fp.requires_grad = True
43     sb_fp = (b_fp - gmin) / (gmax - gmin)

```

```

44
45     N_atoms = batch_info['N_atoms'].view(-1)
46     b_e = batch_info['b_e'].view(-1)
47     b_f = batch_info['b_f']
48
49     Atomic_Es = model(sb_fp)
50     E_predict = torch.sum(Atomic_Es * b_e_mask, dim = [1,2])
51     E_predict = E_predict/N_atoms
52     E_predict = E_predict * (emax - emin) + emin
53
54     uncerts = []
55     for i, ei in enumerate(E_predict):
56         gprime = torch.autograd.grad(ei, model.parameters(), create_graph=True, retain_graph=True)
57         gprime = flatten_gprime(gprime).detach().numpy()
58         se = gprime @ pcov @ gprime
59         uncerts += [(np.sqrt(se), np.sqrt(se + rmse.item()*2), np.linalg.norm(gprime))]
60     uncerts = np.array(uncerts)
61     return uncerts
62
63
64 Name = 'lattice39-2'
65
66 #load datasets
67 train_dict = torch.load(f'{Name}/final_train.sav')
68 val_dict = torch.load(f'{Name}/final_val.sav')
69 test_dict = torch.load(f'{Name}/test.sav')
70
71 #get NN predictions, RMSE, hessian
72 pred_e, actual_e, rmse, h = evaluate_uncert(f'{Name}/best_model',train_dict, True)
73 h = h.detach().numpy()
74 pred_e_val, actual_e_val, rmse_val = evaluate_uncert(f'{Name}/best_model',val_dict, False)
75 pred_e_test, actual_e_test, rmse_test = evaluate_uncert(f'{Name}/best_model',test_dict, False)
76
77
78 ndata = pred_e.shape[0]
79 alpha = rmse.item()*2
80 pcov = get_pcov(h)
81
82 #get uncertainties
83 uncerts_val = get_uncerts(f'{Name}/best_model',val_dict)
84 uncerts_train = get_uncerts(f'{Name}/best_model',train_dict)
85 uncerts_test = get_uncerts(f'{Name}/best_model',test_dict)
86
87 #####
88 #Parity Plot
89 #####
90
91 plt.clf()
92
93 fig, ax = plt.subplots(ncols=2, nrows=1, sharex=False, sharey='row')
94
95 fig.set_size_inches(7, 3.5)
96
97 eline = np.linspace(np.min(np.concatenate((actual_e, actual_e_test))),
98                     np.max(np.concatenate((actual_e, actual_e_test))), 10)
99
100 ax[0].set_title(' ')
101 ax[0].plot(actual_e, pred_e, '.',color='tab:orange', alpha=1, label='Train')
102 ax[0].set_xlabel(' ')
103 ax[0].set_ylabel('NN Energy (eV/atom)')
104 ax[0].legend()
105 ax[0].plot(eline, eline,'k--',alpha=0.7)
106
107 ax[1].plot(eline, eline,'k--',alpha=0.7)
108 ax[1].plot(actual_e_val, pred_e_val, '.',color='g', alpha=0.9, label='Validation')
109 ax[1].plot(actual_e_test, pred_e_test, '.',color='y', alpha=0.8, label='Test')
110 ax[1].legend()
111

```

```

112 plt.figtext(0.55,0.04,"DFT Energy (eV/atom)", va="center", ha="center", size=10.5)
113 plt.tight_layout()
114 plt.savefig('subplotparityslides-energy-only.png')
115 print(''#+attr_org: :width 600
116 #+caption: Figure 4
117 [./subplotparityslides-energy-only.png]')
118
119 #####
120 # Distribution of uncertainties
121 #####
122
123 plt.clf()
124 plt.hist(uncerts_train[:,0], label='Train', density=True, alpha=0.5, color='tab:orange')
125 plt.hist(uncerts_val[:,0], label='Validation', density=True, alpha=0.5, color='g')
126 plt.hist(uncerts_test[:,0], label='Test', density=True, alpha=0.5, color='y')
127 plt.legend()
128 plt.xlabel('Standard Error Confidence (eV/atom)')
129 plt.ylabel('Density')
130 plt.tight_layout()
131 plt.savefig('hist-uncerts-pot1.png')
132 print(''#+attr_org: :width 600
133 #+caption: Figure 5
134 [./hist-uncerts-pot1.png]')
135
136 #####
137 # Parity plot with 95% prediction interval
138 #####
139
140 plt.clf()
141 tval = t.ppf(0.975, ndata)
142 plt.errorbar(actual_e_test, pred_e_test, yerr=tval*uncerts_test[:,1], fmt='y_',
143             ecolr='m', label='Test, 95% prediction')
144
145 plt.xlabel('DFT Energy (eV/atom)')
146 plt.ylabel('NN Energy (eV/atom)')
147 plt.plot([np.min(actual_e_test), np.max(actual_e_test)],
148         [np.min(actual_e_test),
149          np.max(actual_e_test)], 'k--', alpha=0.7, linewidth=0.3)
150
151 plt.legend()
152 plt.tight_layout()
153 plt.savefig('parity-errorbar-test-pot1-prediction.png', dpi=200)
154 print(''#+attr_org: :width 600
155 #+caption: Figure 6
156 [./parity-errorbar-test-pot1-prediction.png]')
157
158 #####
159 #Inference on predict-4.0 and 4.1 dataset
160 #####
161
162 data_dict = torch.load(f'lattice40_pred-2/test.sav')
163 pred_e_40p, actual_e_40p, rmse_40p = evaluate_uncert(f'{Name}/best_model',data_dict, False)
164 uncerts_40p = get_uncerts(f'{Name}/best_model',data_dict)
165
166 data_dict = torch.load(f'lattice41_pred-2/test.sav')
167 pred_e_41p, actual_e_41p, rmse_41p = evaluate_uncert(f'{Name}/best_model',data_dict, False)
168 uncerts_41p = get_uncerts(f'{Name}/best_model',data_dict)
169
170 #make plot
171
172 plt.clf()
173 fig, ax = plt.subplots(ncols=2, nrows=1, sharex=False, sharey='row')
174 fig.set_size_inches(10, 4)
175 ax[0].set_title(' ')
176 ax[0].errorbar(actual_e_40p, pred_e_40p, yerr = tval * uncerts_40p[:,1], color='tab:pink',
177               fmt = '_', ecolr='r', label='Predict 4.0, 95% prediction')
178 ax[0].set_xlabel(' ')
179 ax[0].set_ylabel('NN Energy (eV/atom)')

```

```

180 ax[0].legend()
181 eline = np.linspace(np.min(np.concatenate((actual_e_40p, actual_e_41p))),
182                     np.max(np.concatenate((actual_e_40p, actual_e_41p))), 10)
183 ax[0].plot(eline, eline, 'k--', alpha=0.8, linewidth=0.5)
184
185 ax[1].errorbar(actual_e_41p, pred_e_41p, yerr = tval * uncerts_41p[:,1],
186               fmt = 'b_', ecolor='c', label='Predict 4.1, 95% prediction')
187 ax[1].legend()
188 ax[1].plot(eline, eline, 'k--', alpha=0.7, linewidth=0.5)
189
190 plt.figtext(0.55,0.03,"DFT Energy (eV/atom)", va="center", ha="center", size=10.5)
191
192 plt.tight_layout()
193 plt.savefig('subplot-parity-40-41-pot-prediction.png', dpi=200)
194 print(''+attr_org: :width 600
195      #+caption: Figure 7
196      [./subplot-parity-40-41-pot-prediction.png]))
197
198 #####
199 #Uncertainty vs True Error Scatterplot
200 #####
201
202
203 def scatter_hist(x, y, ax, ax_histx, ax_histy, label, color=None):
204     # no labels
205     ax_histx.tick_params(axis="x", labelbottom=False)
206     ax_histy.tick_params(axis="y", labelleft=False)
207
208     # the scatter plot:
209     ax.scatter(x, y, alpha=0.5, label=label, color=color)
210
211     # now determine nice limits by hand:
212     binwidth = 0.0001
213     xymax = max(np.max(np.abs(x)), np.max(np.abs(y)))
214     lim = (int(xymax/binwidth)+1)*binwidth
215
216     #bins = np.arange(0, lim + binwidth, binwidth)
217     ax_histx.hist(x, alpha=0.5, color=color, density=True)
218     ax_histy.hist(y, orientation='horizontal', alpha=0.5, color=color, density=True)
219
220 fig = plt.figure(figsize=(8, 8))
221 # Add a gridspec with two rows and two columns and a ratio of 2 to 7 between
222 # the size of the marginal axes and the main axes in both directions.
223 # Also adjust the subplot parameters for a square plot.
224 gs = fig.add_gridspec(2, 2, width_ratios=(7, 2), height_ratios=(2, 7),
225                       left=0.11, right=0.98, bottom=0.07, top=0.97, wspace=0.05, hspace=0.05)
226
227 ax = fig.add_subplot(gs[1, 0])
228 ax_histx = fig.add_subplot(gs[0, 0], sharex=ax)
229 ax_histy = fig.add_subplot(gs[1, 1], sharey=ax)
230
231 # use the previously defined function
232
233 scatter_hist(np.absolute(actual_e_test-pred_e_test), uncerts_test[:,0],
234             ax, ax_histx, ax_histy, 'Test', 'y')
235 scatter_hist(np.absolute(actual_e_40p-pred_e_40p), uncerts_40p[:,0],
236             ax, ax_histx, ax_histy, 'Predict 4.0', 'tab:pink')
237 scatter_hist(np.absolute(pred_e_41p-actual_e_41p), uncerts_41p[:,0],
238             ax, ax_histx, ax_histy, 'Predict 4.1')
239
240
241 ax.set_xlabel('Absolute Error Energy (eV/atom)')
242 ax.set_ylabel('Standard Error Confidence (eV/atom)')
243 ax.legend()
244 plt.savefig('uncert-v-error-w-hist-pot1-origw-test.png', dpi=200, bbox_inches='tight')
245 print(''+attr_org: :width 600
246      #+caption: Figure 9
247      [./uncert-v-error-w-hist-pot1-origw-test.png]))

```

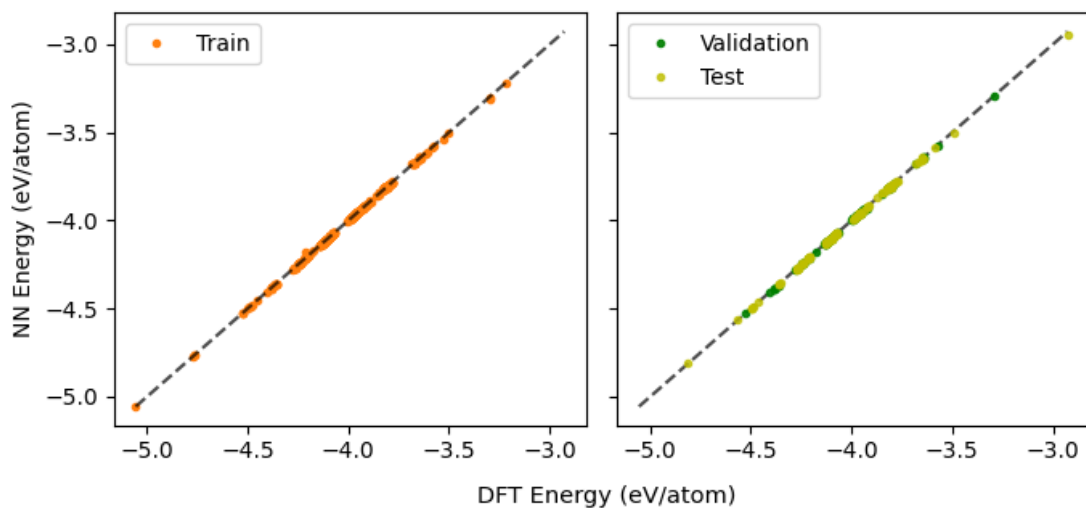


Figure 2: Figure 4

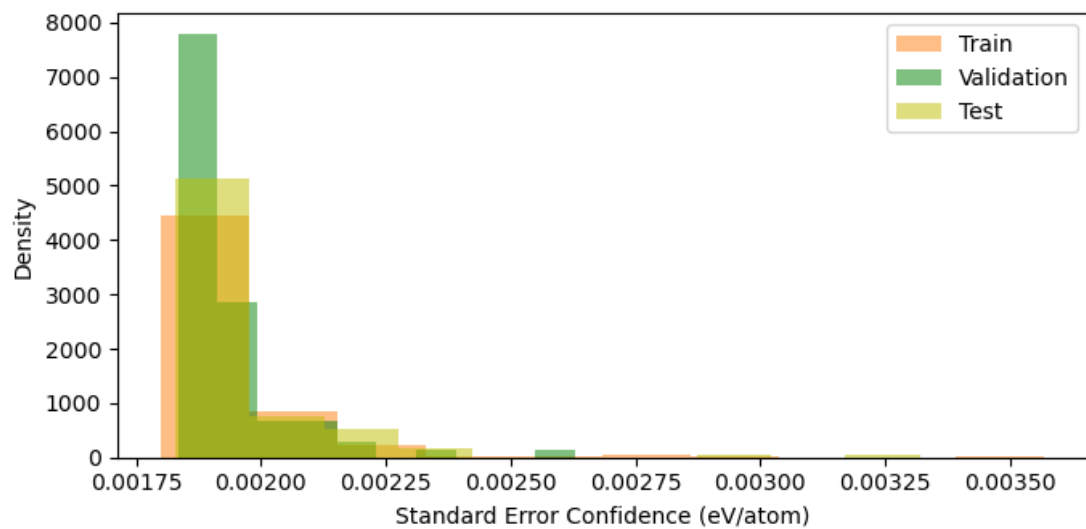


Figure 3: Figure 5

## 5 Fingerprints

```

1 import torch
2 from uncert import get_fps
3 import matplotlib.pyplot as plt
4
5 Name = 'lattice39-2'
6 train_dict = torch.load(f'{Name}/final_train.sav')
7 fp_train, e_mask_train = get_fps(f'{Name}/best_model', train_dict)

```

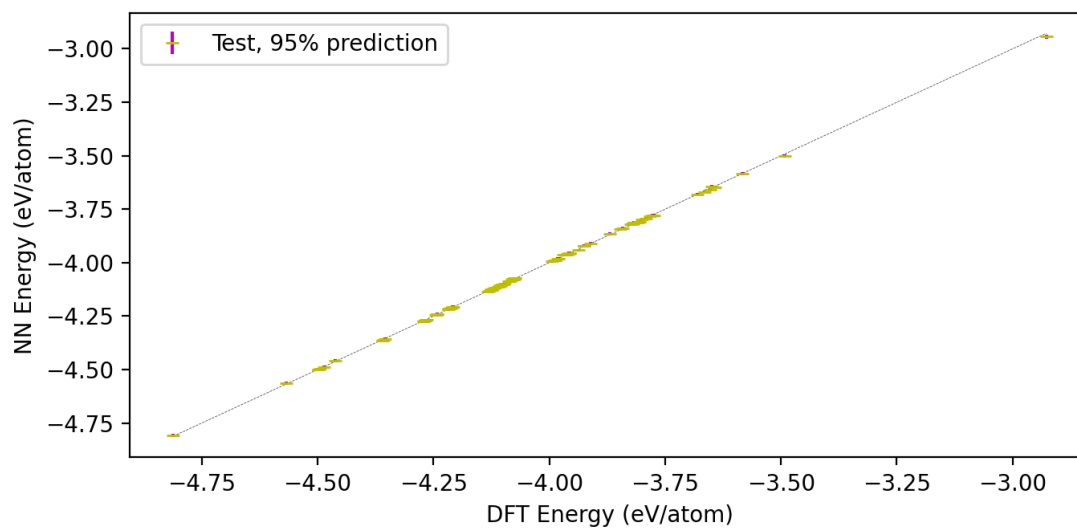


Figure 4: Figure 6

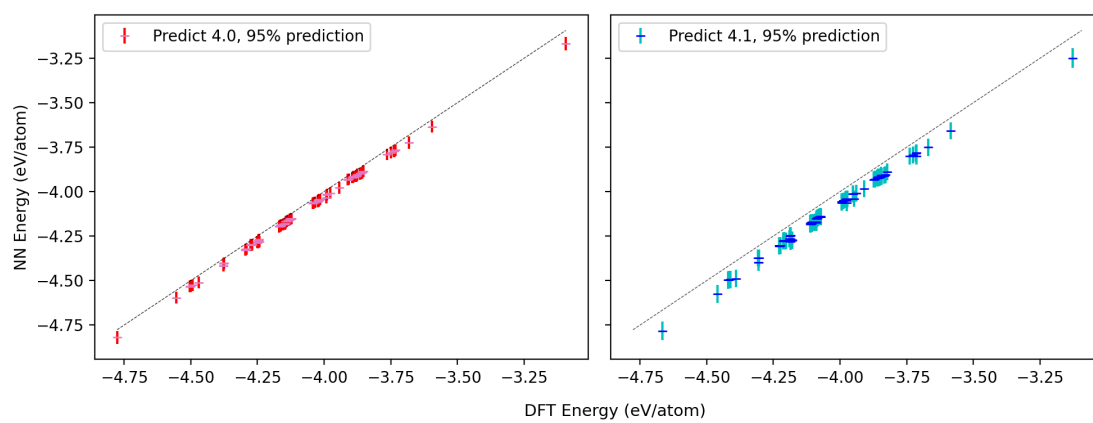


Figure 5: Figure 7

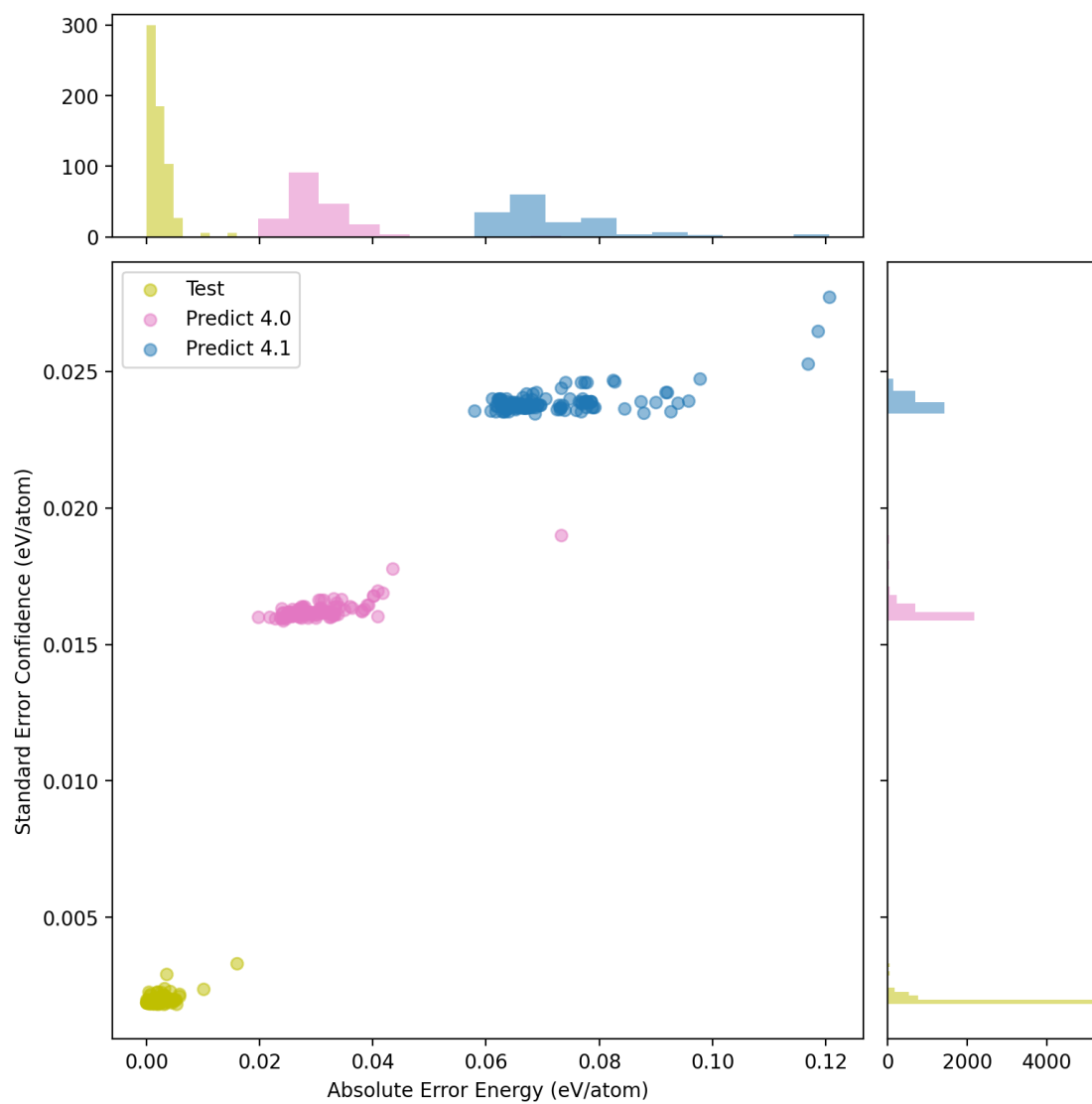


Figure 6: Figure 9

```

8
9 data_dict = torch.load(f'lattice40_pred-2/test.sav')
10 fp_40, e_mask_40 = get_fps(f'{Name}/best_model', data_dict)
11
12 data_dict = torch.load(f'lattice41_pred-2/test.sav')
13 fp_41, e_mask_41 = get_fps(f'{Name}/best_model', data_dict)
14
15 for i in range(2):
16     for j in range(4):
17         plt.clf()
18         plt.hist(fp_train[e_mask_train[:, :, i]==1][:, j], alpha=0.5, density=True, label='Train', color='y')
19         plt.hist(fp_40[e_mask_40[:, :, i]==1][:, j], alpha=0.5, density=True, label='Predict 4.0', color='tab:pink')
20
21         plt.hist(fp_41[e_mask_41[:, :, i]==1][:, j], alpha=0.5, density=True, label='Predict 4.1')
22         plt.xlabel('Fingerprint Value')
23         plt.ylabel('Density')
24         plt.legend()
25         plt.tight_layout()
26         plt.savefig(f'fps-hist-el{i}-fp{j}.png')
27
28     print(f'##attr_org: :width 600
29 #+caption: fps-hist-el{i}-fp{j}
30 [./fps-hist-el{i}-fp{j}.png]')

```

---

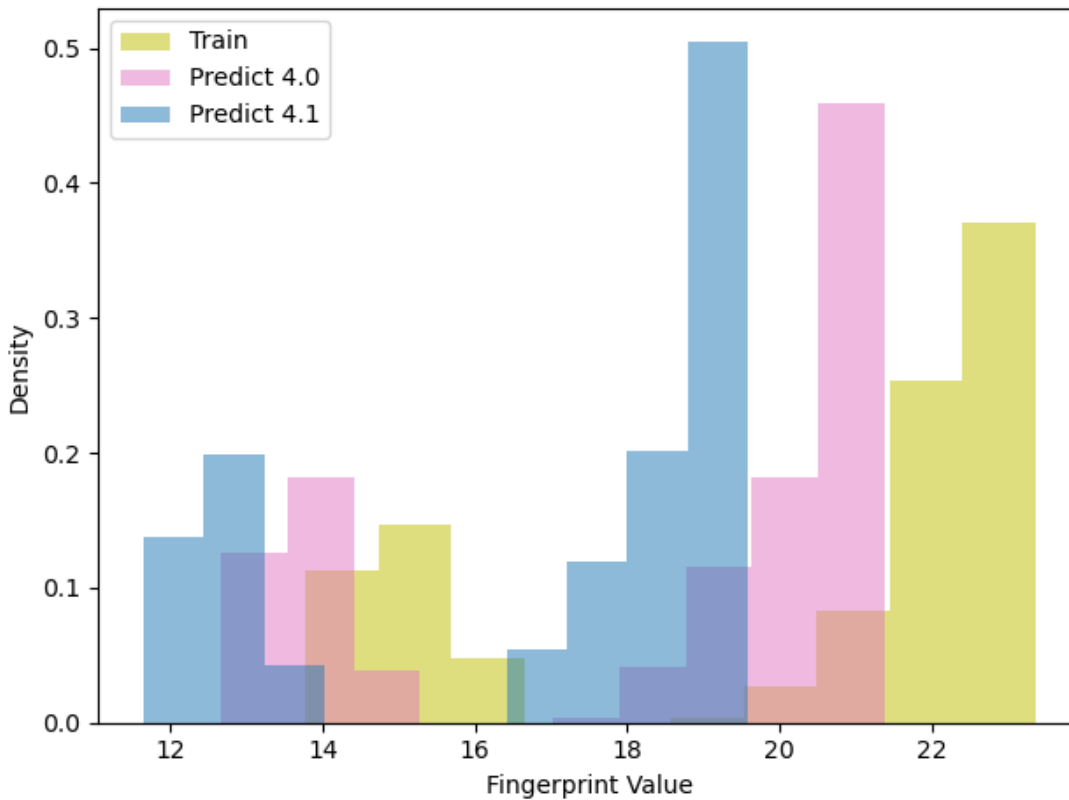


Figure 7: fps-hist-el0-fp0

## 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



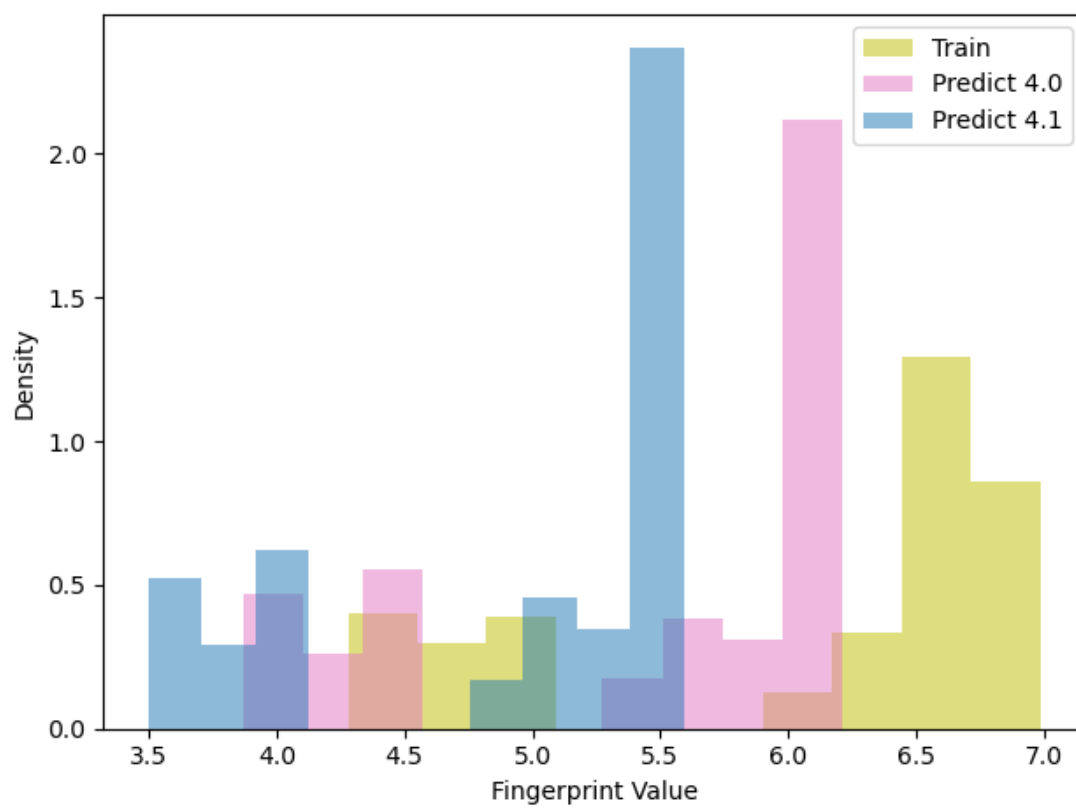


Figure 8: fps-hist-el0-fp1

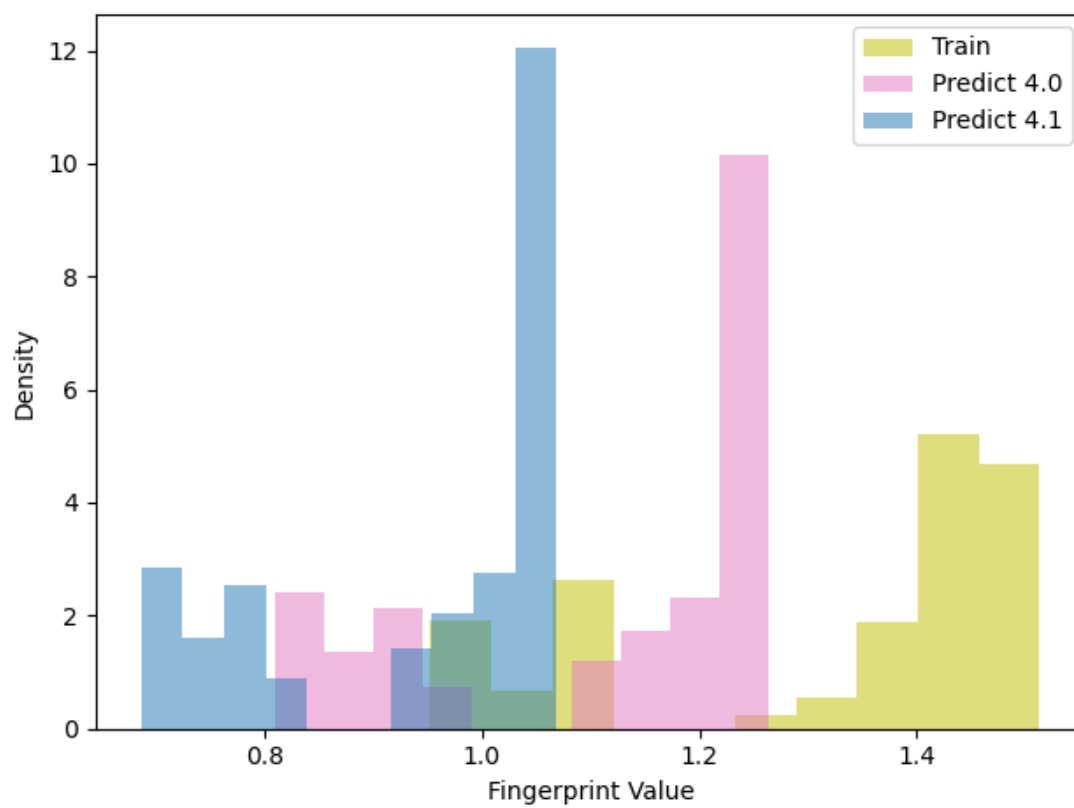


Figure 9: fps-hist-el0-fp2

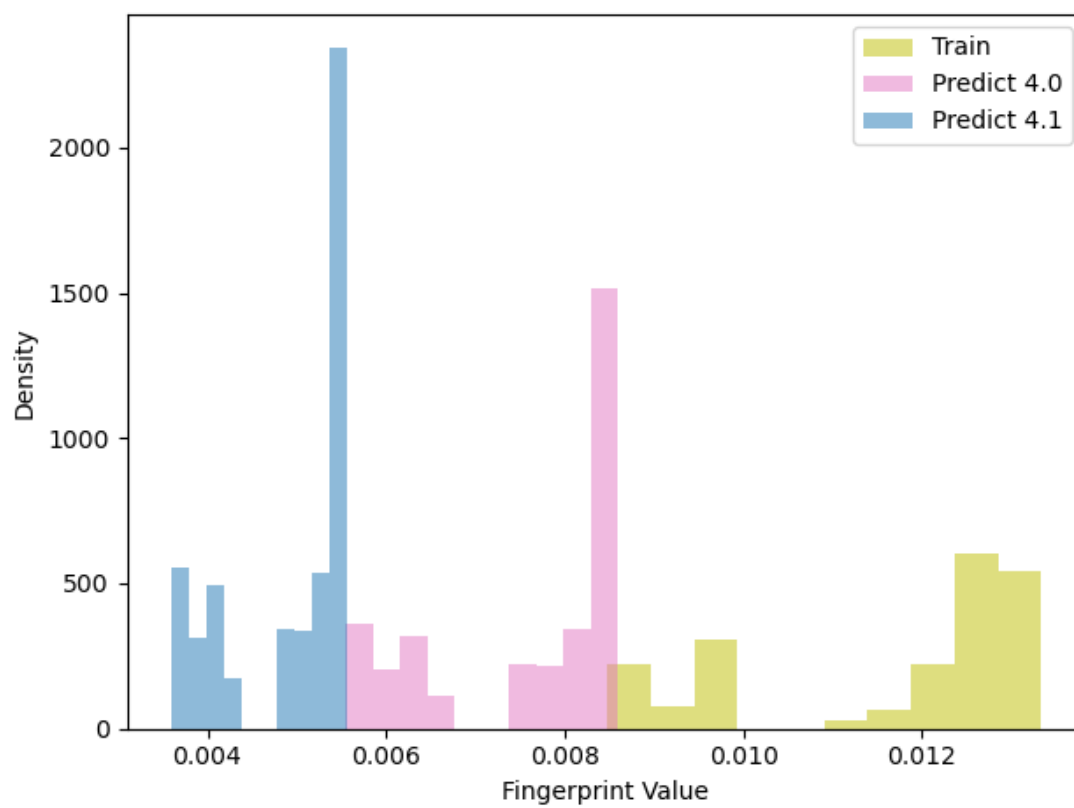


Figure 10: fps-hist-el0-fp3

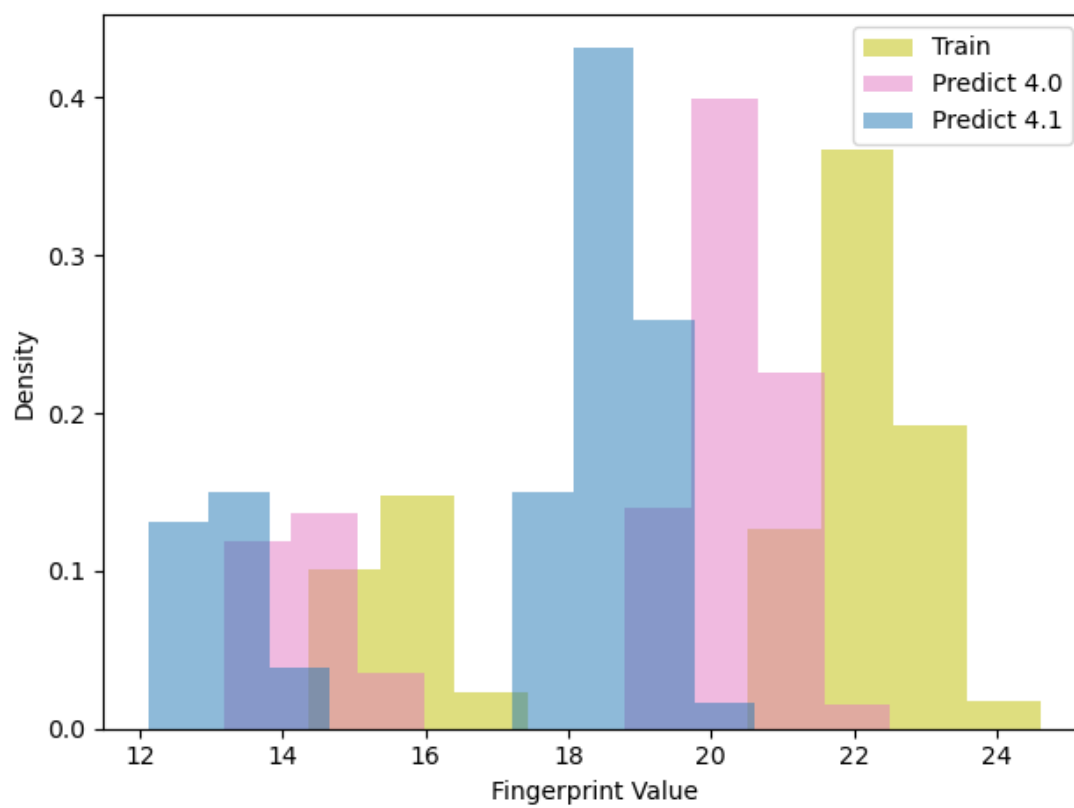


Figure 11: fps-hist-el1-fp0

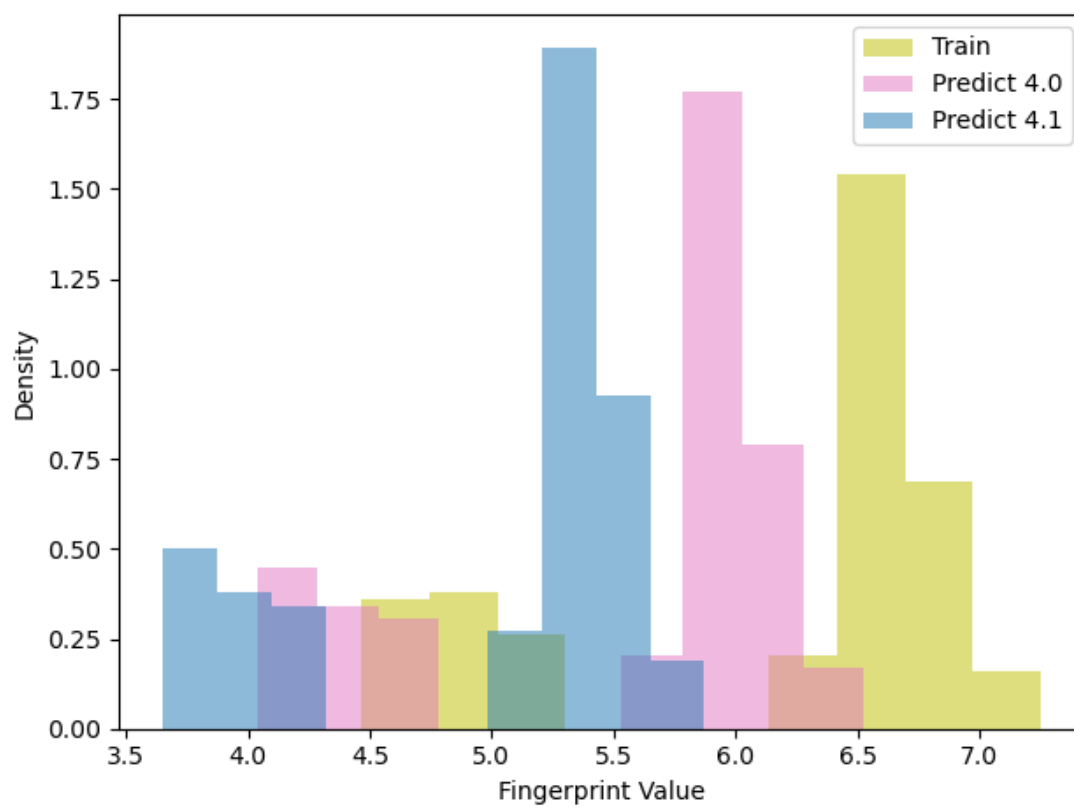


Figure 12: fps-hist-el1-fp1

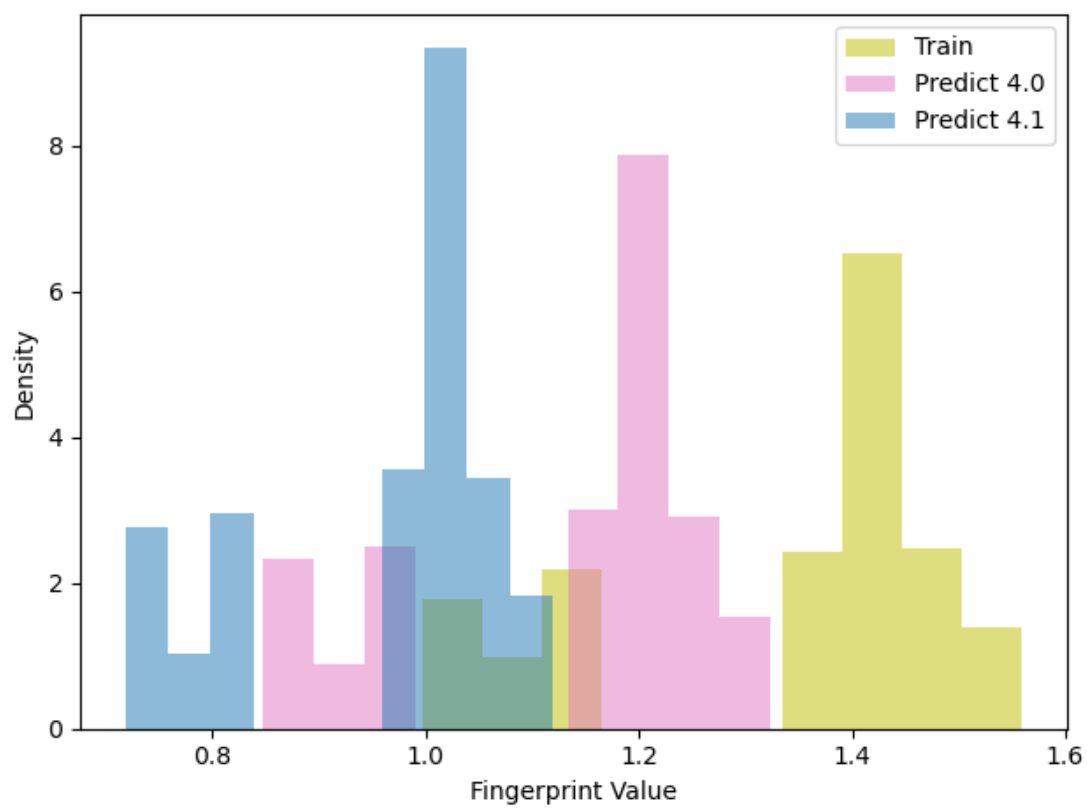


Figure 13: fps-hist-ell-fp2

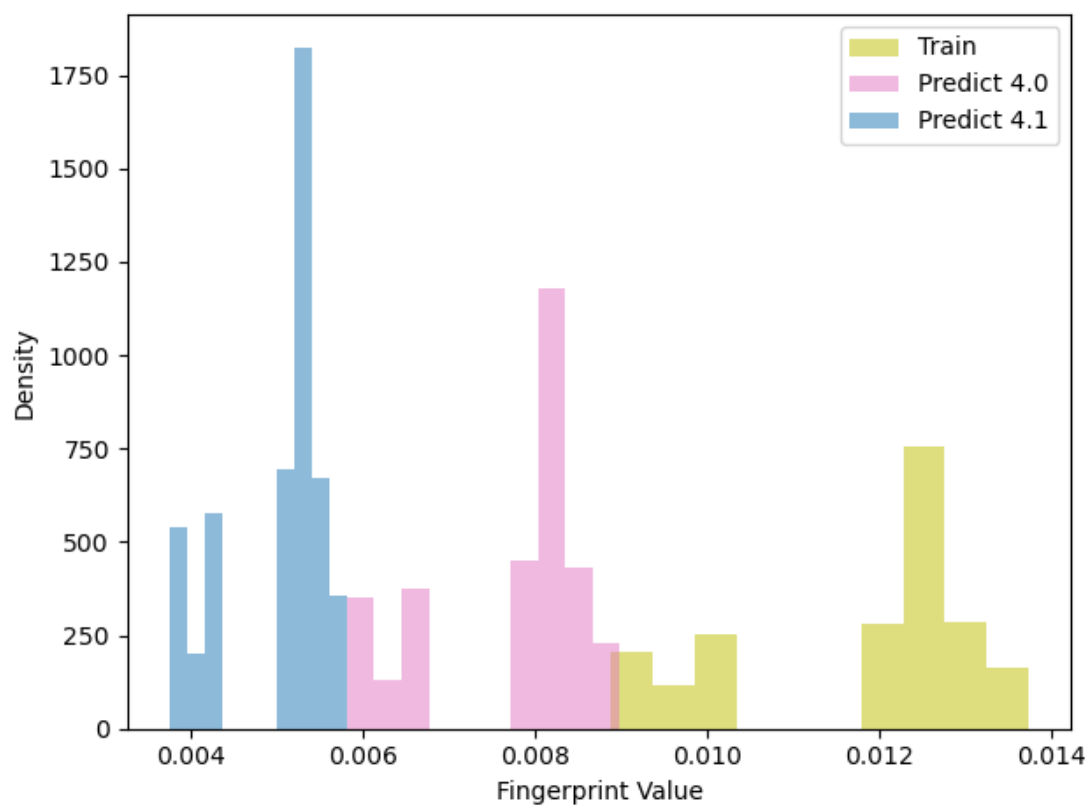


Figure 14: fps-hist-ell-fp3

"lattice39-40-41-2" with relevant files: concatenated dataset files "final\_train.sav", "final\_val.sav", "test.sav"; model file "best\_model".

---

```
1 import sys
2
3 sys.path.append("../SimpleNN")
4 sys.path.append("../")
5
6 import os
7 from ase.db import connect
8 import torch
9 from ContextManager import cd
10 from preprocess import train_test_split, train_val_split, get_scaling, CV
11 from preprocess import snn2sav
12 from NN import MultiLayerNet
13 from train import train, evaluate
14 from fp_calculator import set_sym, calculate_fp
15 import pickle
16
17 is_train = True
18 is_transfer = False
19 is_force = True
20
21 if is_train and is_transfer:
22     raise ValueError('train and transfer could not be true at the same time.')
23
24 #####
25 #Hyperparameters
26 #####
27 E_coeff = 100
28 if is_force:
29     F_coeff = 1
30 else:
31     F_coeff = 0
32
33 val_interval = 10
34 n_val_stop = 10
35 epoch = 3000
36
37 opt_method = 'lbfgs'
38
39
40 if opt_method == 'lbfgs':
41     history_size = 100
42     lr = 1
43     max_iter = 10
44     line_search_fn = 'strong_wolfe'
45
46
47 convergence = {'E_cov':0.0005, 'F_cov':0.005}
48
49 # min_max will scale fingerprints to (0,1)
50 fp_scale_method = 'min_max'
51 e_scale_method = 'min_max'
52
53
54 test_percent = 0.2
55 # Percentage from train+val
56 val_percent = 0.2
57
58 # Training model configuration
59 SEED = [2]
60 n_nodes = [11,11]
61 activations = [torch.nn.Sigmoid(), torch.nn.Sigmoid()]
62
63 lr = 1
64 hp = {'n_nodes': n_nodes, 'activations': activations, 'lr': lr}
```



```

65
66 #####
67 #Configuration
68 #####
69
70 elements = ['Pd', 'Au']
71 nelelem = len(elements)
72
73 element_energy = None
74 weights = [1.58, 1.92]
75
76 Gs = [22]
77 cutoff = 6.35
78 g2_etas = [0.00, 0.10713, 0.285686, 0.892769]
79 g2_Rses = [0.0]
80
81 sym_params = [Gs, cutoff, g2_etas, g2_Rses, elements, weights, element_energy]
82 params_set = set_sym(elements, Gs, cutoff,
83                      g2_etas=g2_etas, g2_Rses=g2_Rses,
84                      weights=weights)
85 N_sym = params_set[elements[0]]['num']
86
87 #####
88 #Training
89 #####
90
91 Name = 'lattice39-40-41'
92
93 if is_train:
94     for seed in SEED:
95         # This use the context manager to operate in the data directory
96
97         if not os.path.exists(Name+f'--{seed}'):
98             os.makedirs(Name+f'--{seed}')
99
100         with cd(Name+f'--{seed}'):
101             pickle.dump(sym_params, open("sym_params.sav", "wb"))
102             logfile = open('log.txt', 'w+')
103             resultfile = open('result.txt', 'w+')
104
105             if os.path.exists('test.sav'):
106                 logfile.write('Did not calculate symfunctions.\n')
107             else:
108                 #this part is to concatenate the train-data subsets together.
109                 train_dict1 = torch.load('../lattice39-2/final_train.sav')
110                 train_dict2 = torch.load('../lattice40_pred-2/final_train.sav')
111                 train_dict3 = torch.load('../lattice41_pred-2/final_train.sav')
112                 train_dict = dict(train_dict1)
113                 new_dict = {k+1000: v for k, v in train_dict2.items()}
114                 train_dict.update(new_dict)
115                 new_dict = {k+2000: v for k, v in train_dict3.items()}
116                 train_dict.update(new_dict)
117
118                 val_dict1 = torch.load('../lattice39-2/final_val.sav')
119                 val_dict2 = torch.load('../lattice40_pred-2/final_val.sav')
120                 val_dict3 = torch.load('../lattice41_pred-2/final_val.sav')
121                 val_dict = dict(val_dict1)
122                 new_dict = {k+1000: v for k, v in val_dict2.items()}
123                 val_dict.update(new_dict)
124                 new_dict = {k+2000: v for k, v in val_dict3.items()}
125                 val_dict.update(new_dict)
126
127                 test_dict1 = torch.load('../lattice39-2/test.sav')
128                 test_dict2 = torch.load('../lattice40_pred-2/test.sav')
129                 test_dict3 = torch.load('../lattice41_pred-2/test.sav')
130                 test_dict = dict(test_dict1)
131                 new_dict = {k+1000: v for k, v in test_dict2.items()}

```

```

133         test_dict.update(new_dict)
134         new_dict = {k+2000: v for k, v in test_dict3.items()}
135         test_dict.update(new_dict)
136
137
138
139         torch.save(train_dict, 'final_train.sav')
140         torch.save(val_dict, 'final_val.sav')
141         torch.save(test_dict, 'test.sav')
142
143         scaling = get_scaling(train_dict, fp_scale_method, e_scale_method)
144
145         n_nodes = hp['n_nodes']
146         activations = hp['activations']
147         lr = hp['lr']
148         #model = torch.load('../lattice39-2/best_model')
149         model = MultiLayerNet(N_sym, n_nodes, activations, nelelem, scaling=scaling)
150         if opt_method == 'lbfgs':
151             optimizer = torch.optim.LBFGS(model.parameters(), lr=lr,
152                                           max_iter=max_iter, history_size=history_size,
153                                           line_search_fn=line_search_fn)
154
155         results = train(train_dict, val_dict,
156                       model,
157                       opt_method, optimizer,
158                       E_coeff, F_coeff,
159                       epoch, val_interval,
160                       n_val_stop,
161                       convergence, is_force,
162                       logfile)
163         [loss, E_MAE, F_MAE, v_loss, v_E_MAE, v_F_MAE] = results
164
165         test_results = evaluate(test_dict, E_coeff, F_coeff, is_force)
166         [test_loss, test_E_MAE, test_F_MAE] = test_results
167         resultfile.write(f'Hyperparameter: n_nodes = {n_nodes}, activations = {activations}, lr = {lr}\n')
168         resultfile.write(f'loss = {loss}, E_MAE = {E_MAE}, F_MAE = {F_MAE}.\n')
169         resultfile.write(f'v_loss = {v_loss}, v_E_MAE = {v_E_MAE}, v_F_MAE = {v_F_MAE}.\n')
170         resultfile.write(f'test_loss = {test_loss}, test_E_MAE = {test_E_MAE}, test_F_MAE = {test_F_MAE}.\n')
171
172
173         logfile.close()
174         resultfile.close()

```

---

## 7 Uncertainty for retrained model

---

```

1  import torch
2  from uncert import evaluate_uncert
3  import numpy as np
4  import matplotlib.pyplot as plt
5  from scipy.stats.distributions import t
6  from Batch import batch_pad
7
8  #get inverse fisher information
9  def get_pcov(h):
10     eigs0 = np.linalg.eigvalsh(h)[0]
11     if (eigs0 < 0):
12         eps = max(1e-5, eigs0*-1.05)
13     else:
14         eps = 1e-5
15     j = np.linalg.pinv(h + eps*np.identity(h.shape[0]))
16     pcov1 = j*alpha
17     u, v = np.linalg.eigh(pcov1)
18     return v @ np.diag(np.maximum(u,0)) @ v.T
19
20
21 def flatten_gprime(agrad):

```

```

22     cnt = 0
23     for g in agrad:
24         g_vector = g.contiguous().view(-1) if cnt ==0 else torch.cat([g_vector, g.contiguous().view(-1)])
25         cnt = 1
26     return g_vector
27
28 #get uncertainties for a dataset
29 def get_uncerts(name, data_dict):
30     model = torch.load(name)
31     scaling = model.scaling
32     gmin = scaling['gmin']
33     gmax = scaling['gmax']
34     emin = scaling['emin']
35     emax = scaling['emax']
36
37     ids = np.array(list(data_dict.keys()))
38     batch_info = batch_pad(data_dict,ids)
39     b_fp = batch_info['b_fp']
40
41     b_e_mask = batch_info['b_e_mask']
42     b_fp.requires_grad = True
43     sb_fp = (b_fp - gmin) / (gmax - gmin)
44
45     N_atoms = batch_info['N_atoms'].view(-1)
46     b_e = batch_info['b_e'].view(-1)
47     b_f = batch_info['b_f']
48
49     Atomic_Es = model(sb_fp)
50     E_predict = torch.sum(Atomic_Es * b_e_mask, dim = [1,2])
51     E_predict = E_predict/N_atoms
52     E_predict = E_predict * (emax - emin) + emin
53
54     uncerts = []
55     for i, ei in enumerate(E_predict):
56         gprime = torch.autograd.grad(ei, model.parameters(), create_graph=True, retain_graph=True)
57         gprime = flatten_gprime(gprime).detach().numpy()
58         se = gprime @ pcov @ gprime
59         uncerts += [(np.sqrt(se), np.sqrt(se + rmse.item()*2), np.linalg.norm(gprime))]
60     uncerts = np.array(uncerts)
61     return uncerts
62
63 Name = 'lattice39-40-41-2'
64
65 train_dict = torch.load(f'{Name}/final_train.sav')
66 val_dict = torch.load(f'{Name}/final_val.sav')
67 test_dict = torch.load(f'{Name}/test.sav')
68
69 pred_e, actual_e, rmse, h = evaluate_uncert(f'{Name}/best_model',train_dict, True)
70 h = h.detach().numpy()
71 pred_e_val, actual_e_val, rmse_val = evaluate_uncert(f'{Name}/best_model',val_dict, False)
72 pred_e_test, actual_e_test, rmse_test = evaluate_uncert(f'{Name}/best_model',test_dict, False)
73
74 ndata = pred_e.shape[0]
75 alpha = rmse.item()*2
76 pcov = get_pcov(h)
77
78 uncerts_val = get_uncerts(f'{Name}/best_model',val_dict)
79 uncerts_train = get_uncerts(f'{Name}/best_model',train_dict)
80 uncerts_test = get_uncerts(f'{Name}/best_model',test_dict)
81
82
83 #####
84 #Parity plot after retraining
85 #####
86
87 data_dict = torch.load(f'lattice40_pred-2/test.sav')
88 pred_e_40p, actual_e_40p, rmse_40p = evaluate_uncert(f'{Name}/best_model',data_dict, False)
89 uncerts_40p = get_uncerts(f'{Name}/best_model',data_dict)

```

```

90
91 data_dict = torch.load(f'lattice41_pred-2/test.sav')
92 pred_e_41p, actual_e_41p, rmse_41p = evaluate_uncert(f'{Name}/best_model', data_dict, False)
93 uncerts_41p = get_uncerts(f'{Name}/best_model', data_dict)
94
95 tval = t.ppf(0.975, ndata)
96 plt.clf()
97
98 fig, ax = plt.subplots(ncols=2, nrows=1, sharex=False, sharey='row')
99 fig.set_size_inches(10, 4)
100 ax[0].set_title(' ')
101 ax[0].errorbar(actual_e, pred_e, yerr = tval * uncerts_train[:,1], fmt = 'y_', ecolor='m',
102               label='Train, 95% prediction')
103 ax[0].set_xlabel(' ')
104 ax[0].set_ylabel('NN Energy (eV/atom)')
105 ax[0].legend()
106 eline = np.linspace(np.min(actual_e), np.max(actual_e_40p), 10)
107 ax[0].plot(eline, eline, 'k--', alpha=0.7, linewidth=0.3)
108
109 ax[1].errorbar(actual_e_40p, pred_e_40p, yerr = tval * uncerts_40p[:,1], color='tab:pink',
110               fmt = '_', ecolor='b', label='Predict 4.0, 4.1, 95% prediction')
111 ax[1].errorbar(actual_e_41p, pred_e_41p, yerr = tval * uncerts_41p[:,1], color='tab:pink',
112               fmt = '_', ecolor='b', label='')
113 ax[1].legend()
114 ax[1].plot(eline, eline, 'k--', alpha=0.7, linewidth=0.3)
115
116 plt.figtext(0.55, 0.03, "DFT Energy (eV/atom)", va="center", ha="center", size=10.5)
117
118 plt.tight_layout()
119 plt.savefig('subplot-parity-40-41-pot2-pred-v2.png', dpi=200)
120 print('#+attr_org: :width 600
121 #+caption: Figure 10
122 [./subplot-parity-40-41-pot2-pred-v2.png]]
123 ''')
124
125 #####
126 #Uncertainty vs True Error Scatterplot
127 #####
128
129 def scatter_hist(x, y, ax, ax_histx, ax_histy, label, color=None):
130     # no labels
131     ax_histx.tick_params(axis="x", labelbottom=False)
132     ax_histy.tick_params(axis="y", labelleft=False)
133
134     # the scatter plot:
135     ax.scatter(x, y, alpha=0.5, label=label, color=color)
136
137     # now determine nice limits by hand:
138     binwidth = 0.0001
139     xymax = max(np.max(np.abs(x)), np.max(np.abs(y)))
140     lim = (int(xymax/binwidth)+1)*binwidth
141
142     #bins = np.arange(0, lim + binwidth, binwidth)
143     ax_histx.hist(x, alpha=0.5, color=color, density=True)
144     ax_histy.hist(y, orientation='horizontal', alpha=0.5, color=color, density=True)
145
146 fig = plt.figure(figsize=(8, 8))
147 # Add a gridspec with two rows and two columns and a ratio of 2 to 7 between
148 # the size of the marginal axes and the main axes in both directions.
149 # Also adjust the subplot parameters for a square plot.
150 gs = fig.add_gridspec(2, 2, width_ratios=(7, 2), height_ratios=(2, 7),
151                       left=0.11, right=0.98, bottom=0.07, top=0.97, wspace=0.05, hspace=0.05)
152
153 ax = fig.add_subplot(gs[1, 0])
154 ax_histx = fig.add_subplot(gs[0, 0], sharex=ax)
155 ax_histy = fig.add_subplot(gs[1, 1], sharey=ax)
156
157 # use the previously defined function

```

```

158 scatter_hist(np.absolute(actual_e-pred_e), uncerts_train[:,0], ax, ax_histx, ax_histy,
159              'Train', 'y')
160 scatter_hist(np.absolute(actual_e_40p-pred_e_40p), uncerts_40p[:,0], ax, ax_histx, ax_histy,
161              'Predict 4.0', 'tab:pink')
162 scatter_hist(np.absolute(pred_e_41p-actual_e_41p), uncerts_41p[:,0], ax, ax_histx, ax_histy,
163              'Predict 4.1')
164
165 ax.set_xlabel('Absolute Error Energy (eV/atom)')
166 ax.set_ylabel('Standard Error Confidence (eV/atom)')
167 ax.legend()
168 plt.savefig('uncert-v-error-w-hist-ret40-41-orig.png', dpi=200, bbox_inches='tight')
169 print('')
170 #+attr_org: :width 600
171 #+caption: Figure 11
172 [[./uncert-v-error-w-hist-ret40-41-orig.png]]
173 '''

```

---

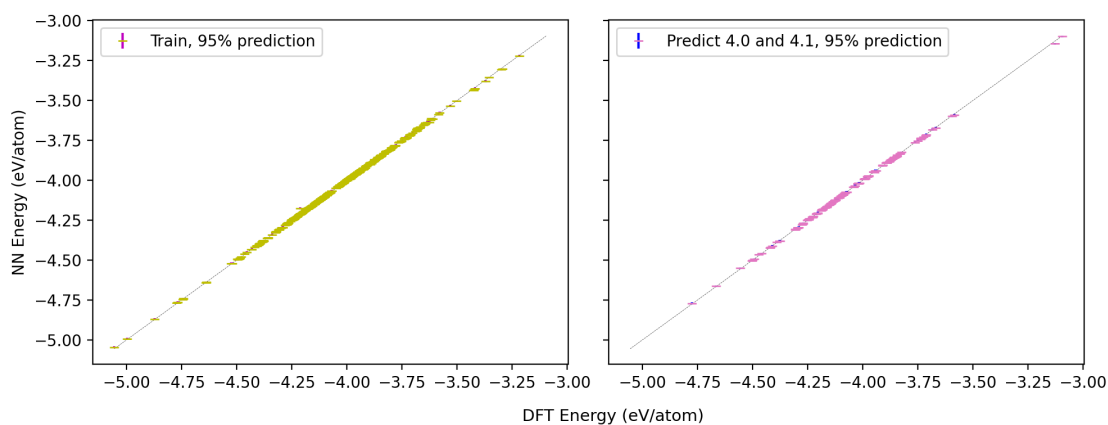


Figure 15: Figure 10

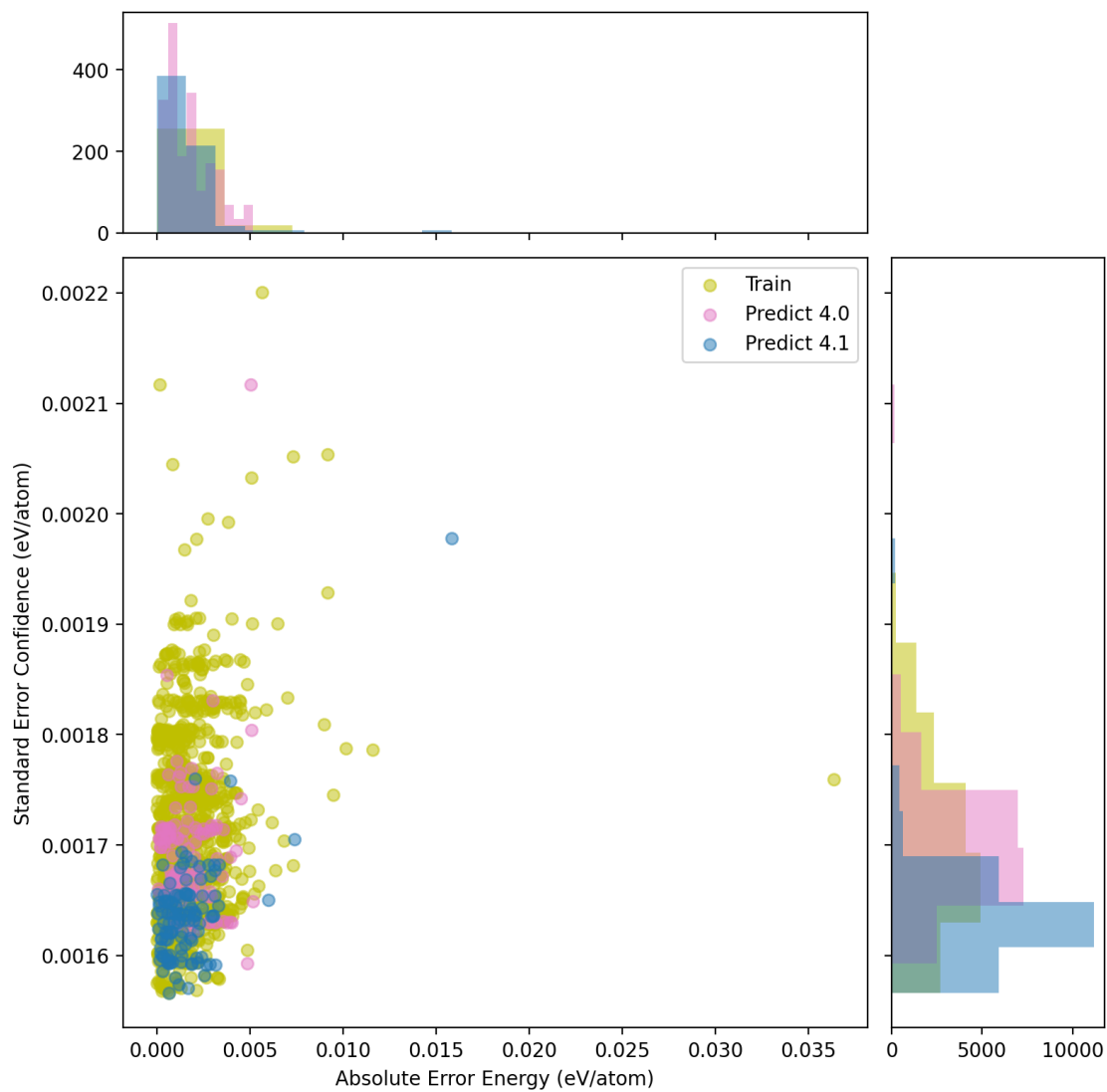


Figure 16: Figure 11