ML models for computed band gap of HOIPs

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This notebook is a part of [V. N. Tuoc, Nga. T. T. Nguyen, V. Sharma, and T. D. Huan, *Probabilistic deep learning approach for targeted hybrid organic-inorganic perovskites*, 2021], and will also be an example of matsML toolkit. Results obtained here can be found in this work.

The original (raw) dataset containing the computed band gap of 1,346 atomic structures predicted for 192 chemical compositions of hybrid organic-inorganic perovskites (HOIPs) is available at [C. Kim, T.D. Huan, S. Krishnan, and R. Ramprasad, Scientific Data 4, 170057 ('17); https://www.nature.com/articles/sdata201757]. Here, three fingerprinted versions of this dataset (S1, S2, and S3) will be fetched from http://www.matsml.org/ and learned to develop 5 ML models (M1, M2, M3, M4, and M5), which are based on Gaussian Process Regression, fully connected Neural Net, and Probability Neural Net. Computations performed using matsML toolkit, available at https://github.com/huantd/matsml.git.

Among 5 models developed, M5 demonstrates a reasonable way to handle the aleatoric uncertainty in deep learning of materials data. More details on this topic can be found in "*Probabilistic deep learning approach for targeted hybrid organic-inorganic perovskites*", the reference mentioned above.

1. Download data

Three (fingerprinted) datasets (S1, S2, and S3) used for the work will be obtained. In fact, S2 has 2 versions, one with selector and one not.

In [1]:

```
from matsml.data import Datasets

data=Datasets(S1='fp_hoips_S1_1dest',S2a='fp_hoips_S2a_2dest',
              S2b='fp_hoips_S2b_1dest',S3='fp_hoips_S3_4tfp')
data.load_dataset()
```

```
matsML, version 1.0.0
*****
Load requested dataset(s)
  Data saved in fp_hoips_S1_1dest.csv.gz
  Data saved in fp_hoips_S2a_2dest.csv.gz
  Data saved in fp_hoips_S2b_1dest.csv.gz
  Data saved in fp_hoips_S3_4tfp.csv.gz
```

2. Obtained datasets parameters

In [2]:

```
# data parameters for learning

n_trains=0.9              # 90% for training, 10% for test
sampling='random'         # method for train/test spliting
x_scaling='minmax'        # method for x scaling
y_scaling='minmax'        # method for y scaling

# Dict of data parameters
data1_params={'data_file':'fp_hoips_S1_1dest.csv.gz','id_col':['ID'],'y_cols':['Ymean'],'comment_cols':[],
    'y_scaling':y_scaling,'x_scaling':x_scaling,'sampling':sampling,'n_trains':n_trains}

data2a_params={'data_file':'fp_hoips_S2a_2dest.csv.gz','id_col':['ID'],'y_cols':['Ymean','Ystd'],
    'comment_cols':[],'y_scaling':y_scaling,'x_scaling':x_scaling,'sampling':sampling,'n_trains':n_trains}

data2b_params={'data_file':'fp_hoips_S2b_1dest.csv.gz','id_col':['ID'],'y_cols':['prop_value'],
    'comment_cols':['Ymean','Ystd','hid'],'y_scaling':y_scaling,'x_scaling':x_scaling,
    'sampling':sampling,'n_trains':n_trains}

data3_params={'data_file':'fp_hoips_S3_4tfp.csv.gz','id_col':['ID'],'y_cols':['Egap'],'comment_cols':[],
    'x_scaling':x_scaling,'y_scaling':'none', 'sampling':sampling,'n_trains':1.0}
```

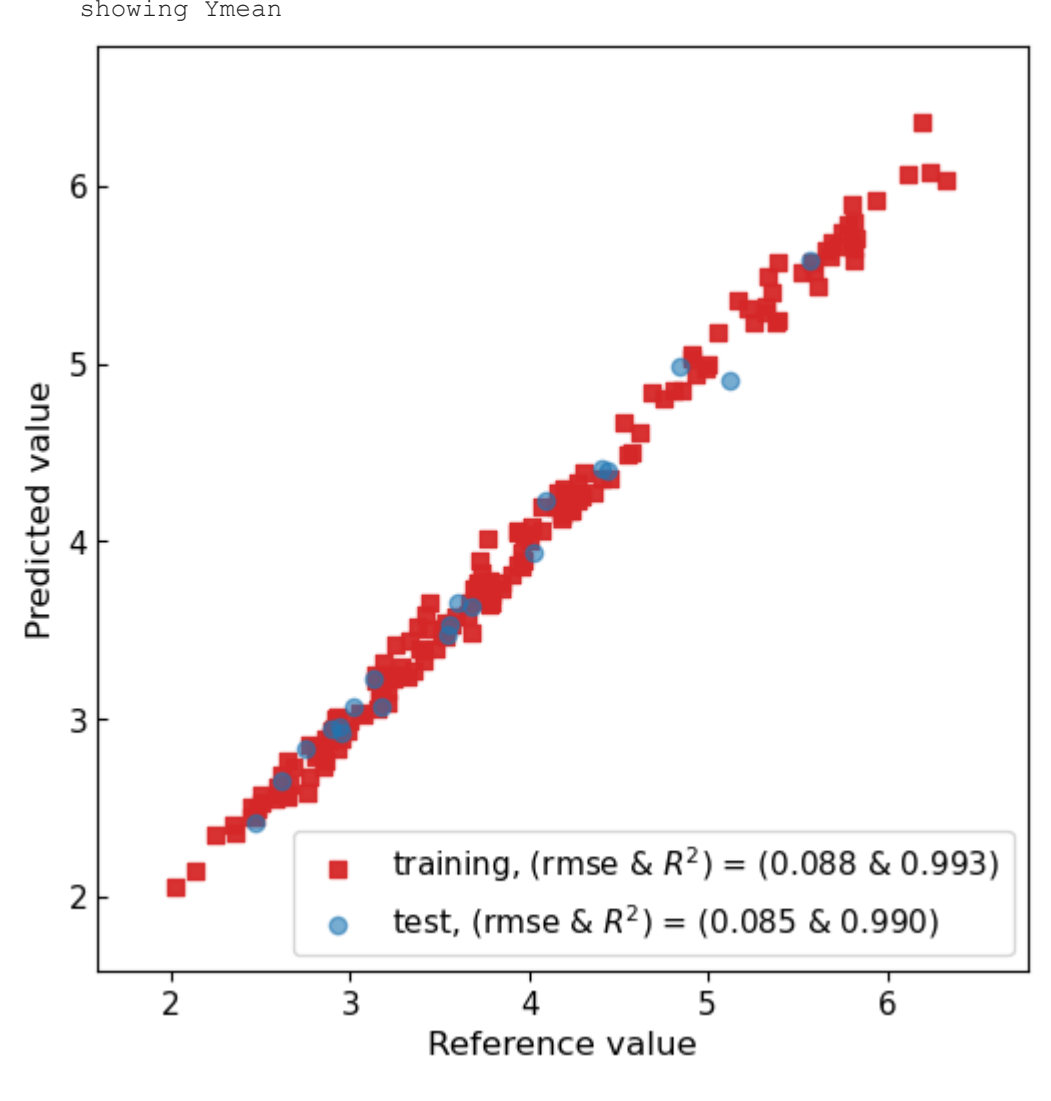
3. ML Models

3a. Model M1: GPR on S1

In [3]:

```
from matsml.models import GPR

# Model parameters
nfold_cv=5                        # Number of folds for cross validation
model_file='M1.pkl'               # Name of the model file to be created
verbosity=0
rmse_cv=False
n_restarts_optimizer=100

model_params={'nfold_cv':nfold_cv,'n_restarts_optimizer':n_restarts_optimizer,'model_file':model_file,
    'verbosity':verbosity,'rmse_cv':rmse_cv}

model=GPR(data_params=data1_params,model_params=model_params)
model.train()
model.plot(pdf_output=False)
```

```
Learning fingerprinted/featured data
  algorithm                     gaussian process regression w/ scikit-learn
  nfold_cv                      5
  optimizer                     fmin_l_bfgs_b
  n_restarts_optimizer          100
  rmse_cv                       False
Checking parameters
  all passed                    True
Read data
  data file                     fp_hoips_S1_1dest.csv.gz
  data size                     192
  training size                 172 (89.6 %)
  test size                     20 (10.4 %)
  x dimensionality              32
  y dimensionality              1
  y label(s)                    ['Ymean']
Scaling x                       minmax
  xscaler saved in              xscaler.pkl
Scaling y                       minmax
Prepare train/test sets         random
Training model w/ cross validation
  cv,rmse_train,rmse_test,rmse_opt: 0 0.018522 0.038449 0.038449
  cv,rmse_train,rmse_test,rmse_opt: 1 0.018840 0.051316 0.038449
  cv,rmse_train,rmse_test,rmse_opt: 2 0.021326 0.037287 0.037287
  cv,rmse_train,rmse_test,rmse_opt: 3 0.018741 0.028500 0.028500
  cv,rmse_train,rmse_test,rmse_opt: 4 0.019046 0.043592 0.028500
GPR model trained, now make predictions & invert scaling
  unscaling y: minmax
     rmse training     Ymean            0.088035
  unscaling y: minmax
     rmse test         Ymean            0.08475
Predictions made & saved in "training.csv" & "test.csv"
Plot results in "training.csv" & "test.csv"
  training, (rmse & R2) = (0.088 & 0.993)
  test, (rmse & R2) = (0.085 & 0.990)
  showing Ymean
```

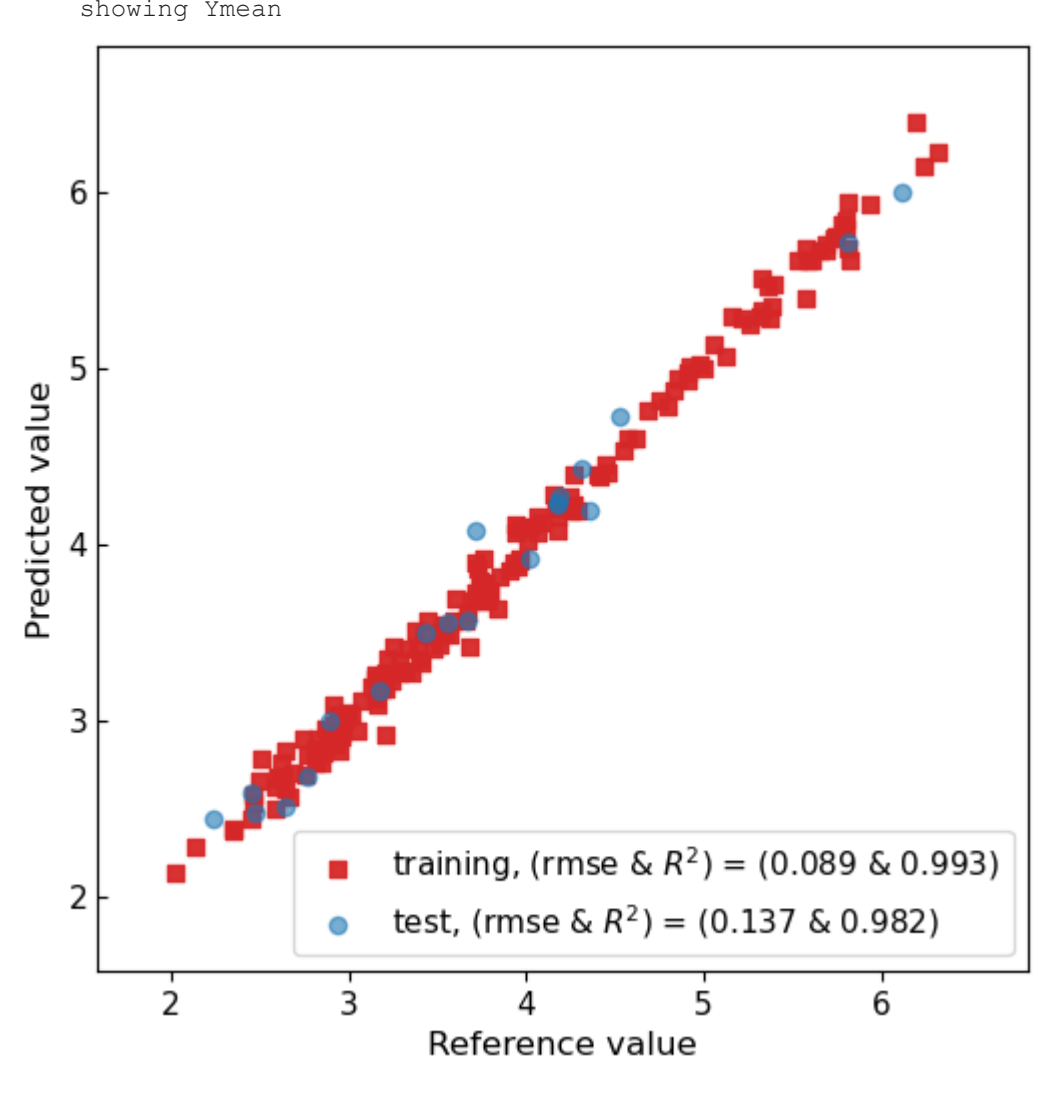


3b. Model M2: FCNN on S1

In [4]:

```
from matsml.models import FCNN

# Model parameters
layers=[5]                    # list of nodes in hidden layers
epochs=2000                   # Epochs
nfold_cv=5                    # Number of folds for cross validation
use_bias=True                 # Use bias term or not
model_file='M2.pkl'           # Name of the model file to be created
verbosity=0                   # Verbosity, 0 or 1
batch_size=32                 # Default = 32
loss='mse'
activ_funct='selu'            # Options: "tanh", "relu", and more
optimizer='nadam'             # Options: "Nadam", "Adam", and more

# Dict of model parameters
model_params={'layers':layers,'activ_funct':activ_funct,'epochs':epochs,'nfold_cv':nfold_cv,
    'optimizer':optimizer,'use_bias':use_bias,'model_file':model_file,'loss':loss,
    'batch_size':batch_size,'verbosity':verbosity,'rmse_cv':False}

model=FCNN(data_params=data1_params,model_params=model_params)
model.train()
model.plot(pdf_output=False)
```

```
Learning fingerprinted/featured data
  algorithm                     fully connected NeuralNet w/ TensorFlow
  layers                        [5]
  activ_funct                   selu
  epochs                        2000
  optimizer                     nadam
  nfold_cv                      5
Checking parameters
  all passed                    True
Read data
  data file                     fp_hoips_S1_1dest.csv.gz
  data size                     192
  training size                 172 (89.6 %)
  test size                     20 (10.4 %)
  x dimensionality              32
  y dimensionality              1
  y label(s)                    ['Ymean']
Scaling x                       minmax
  xscaler saved in              xscaler.pkl
Scaling y                       minmax
Prepare train/test sets         random
Building model                  FCNN
Training model w/ cross validation
  cv,rmse_train,rmse_test,rmse_opt: 0 0.025405 0.033886 0.033886
  cv,rmse_train,rmse_test,rmse_opt: 1 0.018399 0.043445 0.033886
  cv,rmse_train,rmse_test,rmse_opt: 2 0.019758 0.084789 0.033886
  cv,rmse_train,rmse_test,rmse_opt: 3 0.020367 0.028783 0.028783
  cv,rmse_train,rmse_test,rmse_opt: 4 0.018889 0.026327 0.026327
  Optimal ncv:  4 ; optimal NET saved
FCNN trained, now make predictions & invert scaling
  unscaling y: minmax
     rmse training     Ymean            0.088506
  unscaling y: minmax
     rmse test         Ymean            0.137268
Predictions made & saved in "training.csv" & "test.csv"
Plot results in "training.csv" & "test.csv"
  training, (rmse & R2) = (0.089 & 0.993)
  test, (rmse & R2) = (0.137 & 0.982)
  showing Ymean
```

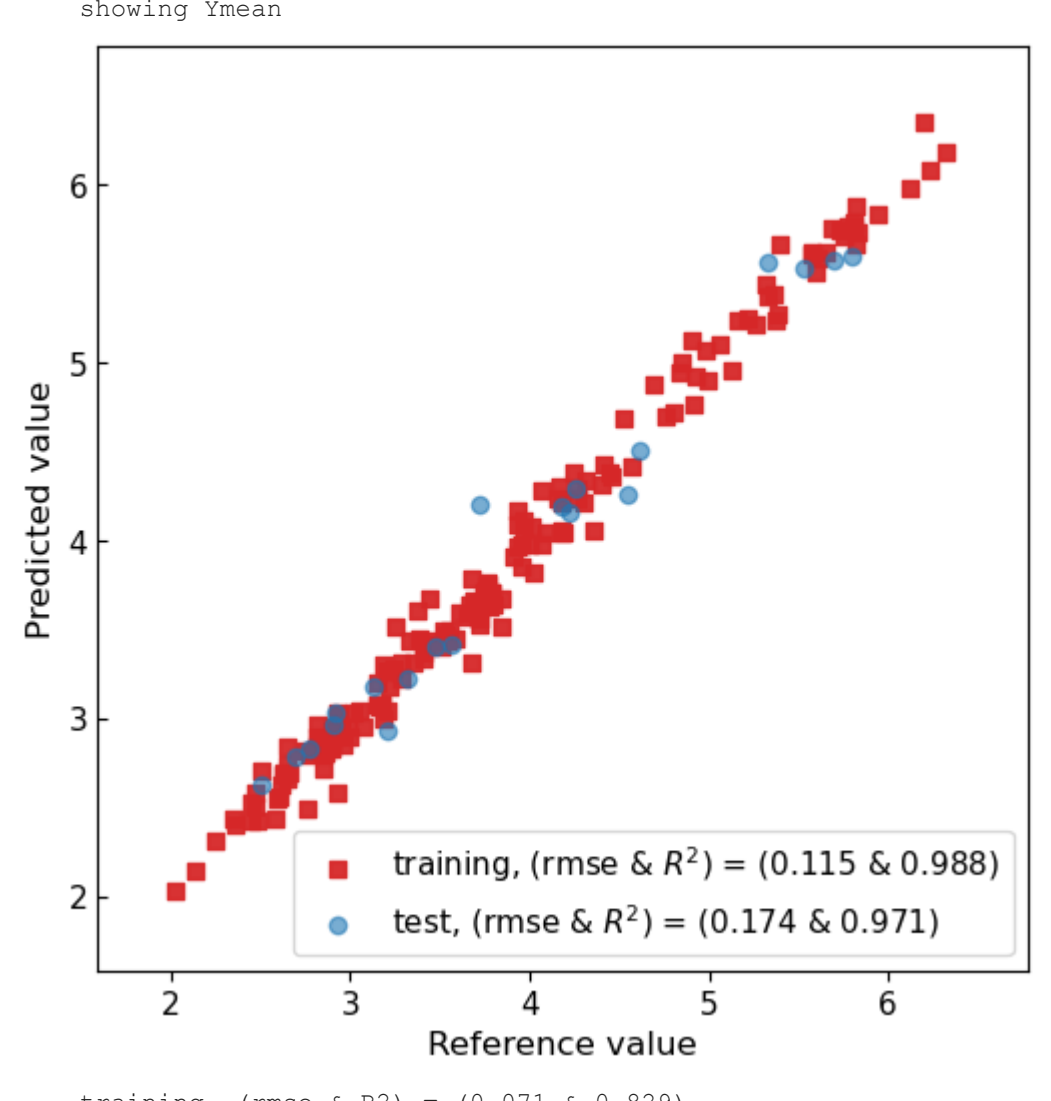


3c. Model M3: FCNN on S2a

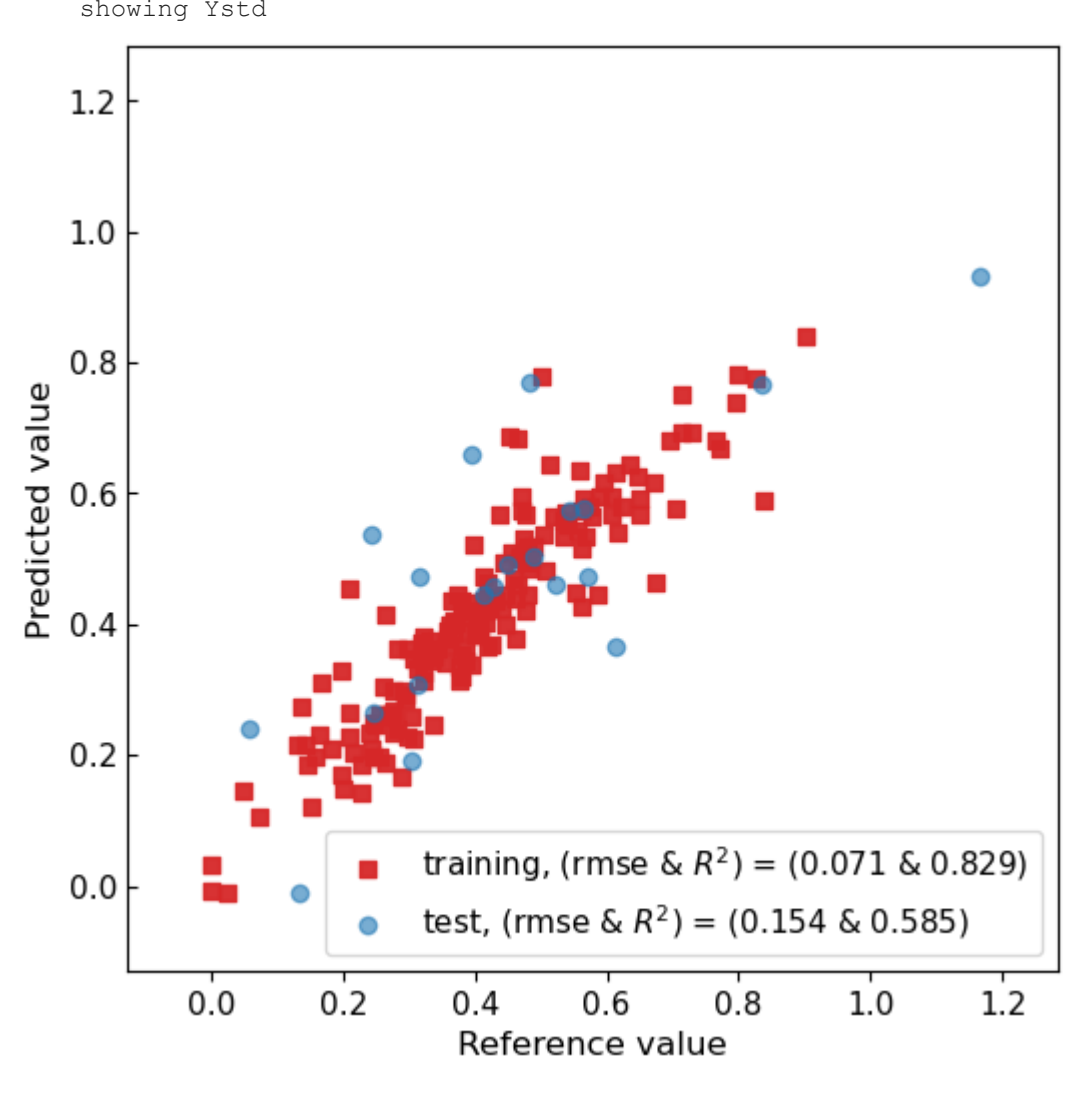
In [5]:

```
# Model parameters
layers=[5,5]
epochs=10000
nfold_cv=5
use_bias=True
model_file='M3.pkl'
loss='mse'
verbosity=0
batch_size=32
activ_funct='elu'
optimizer = 'nadam'

model_params={'layers':layers,'activ_funct':activ_funct,'epochs':epochs,'nfold_cv':nfold_cv,
    'optimizer':optimizer,'use_bias':use_bias,'model_file':model_file,'loss':loss,
    'batch_size':batch_size,'verbosity':verbosity,'rmse_cv':False}

model=FCNN(data_params=data2a_params,model_params=model_params)
model.train()
model.plot(pdf_output=False)
```

```
Learning fingerprinted/featured data
  algorithm                     fully connected NeuralNet w/ TensorFlow
  layers                        [5, 5]
  activ_funct                   elu
  epochs                        10000
  optimizer                     nadam
  nfold_cv                      5
Checking parameters
  all passed                    True
Read data
  data file                     fp_hoips_S2a_2dest.csv.gz
  data size                     192
  training size                 172 (89.6 %)
  test size                     20 (10.4 %)
  x dimensionality              31
  y dimensionality              2
  y label(s)                    ['Ymean', 'Ystd']
Scaling x                       minmax
  xscaler saved in              xscaler.pkl
Scaling y                       minmax
Prepare train/test sets         random
Building model                  FCNN
Training model w/ cross validation
  cv,rmse_train,rmse_test,rmse_opt: 0 0.050190 0.086481 0.086481
  cv,rmse_train,rmse_test,rmse_opt: 1 0.042709 0.078880 0.078880
  cv,rmse_train,rmse_test,rmse_opt: 2 0.034806 0.087286 0.078880
  cv,rmse_train,rmse_test,rmse_opt: 3 0.034643 0.078801 0.078801
  cv,rmse_train,rmse_test,rmse_opt: 4 0.033208 0.087348 0.078801
  Optimal ncv:  3 ; optimal NET saved
FCNN trained, now make predictions & invert scaling
  unscaling y: minmax
     rmse training     Ymean            0.114854
     rmse training     Ystd             0.070648
  unscaling y: minmax
     rmse test         Ymean            0.173631
     rmse test         Ystd             0.153502
Predictions made & saved in "training.csv" & "test.csv"
Plot results in "training.csv" & "test.csv"
  training, (rmse & R2) = (0.115 & 0.988)
  test, (rmse & R2) = (0.174 & 0.971)
  showing Ymean
```




```
  training, (rmse & R2) = (0.071 & 0.829)
  test, (rmse & R2) = (0.154 & 0.585)
  showing Ystd
```

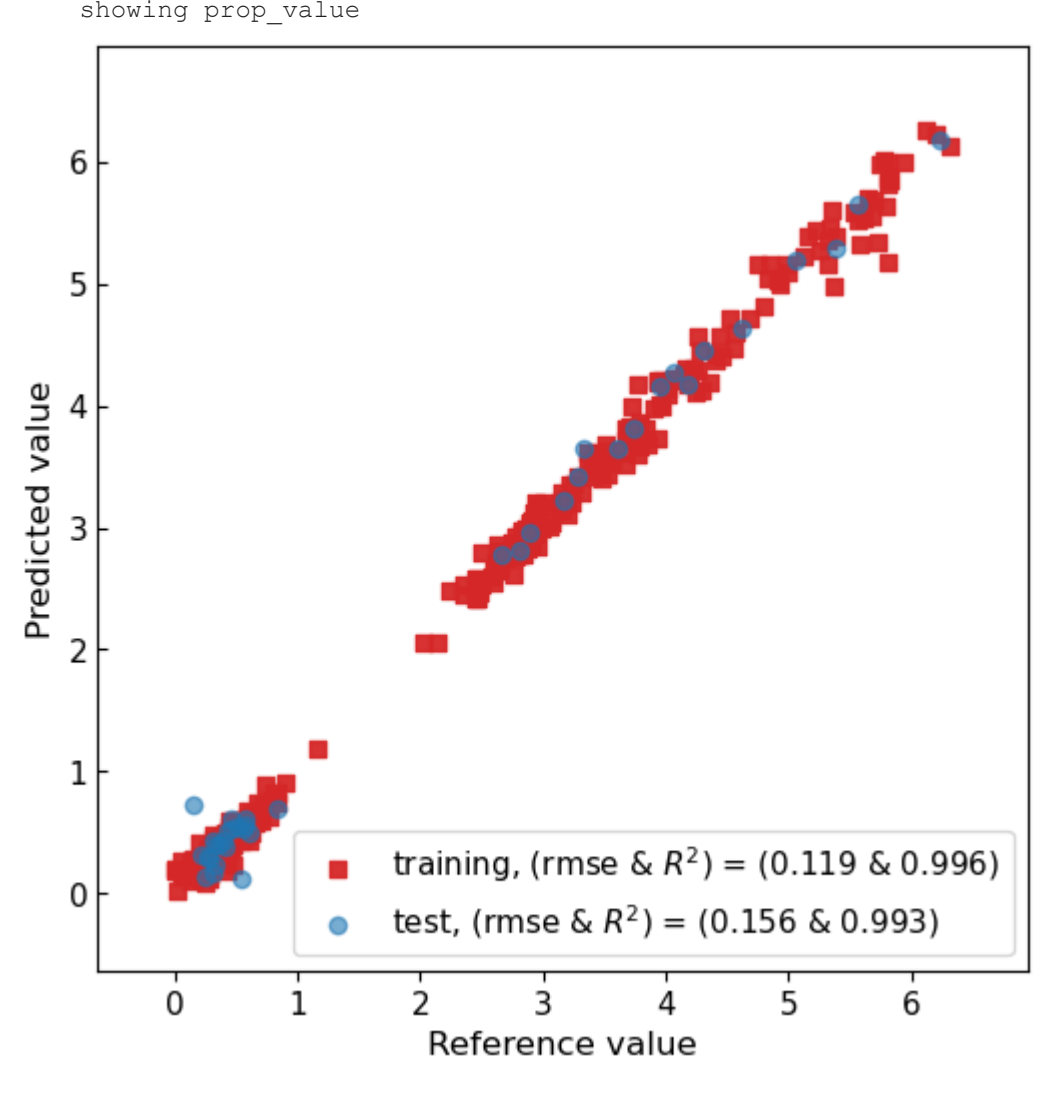


3d. Model M4: FCNN on S2b

In [6]:

```
# Model parameters
layers=[4,4]
epochs=5000
nfold_cv=5
use_bias=True
model_file='M4.pkl'
loss='mse'
verbosity=0
batch_size=32
activ_funct='tanh'
optimizer='nadam'

model_params={'layers':layers,'activ_funct':activ_funct,'epochs':epochs,'nfold_cv':nfold_cv,
    'optimizer':optimizer,'use_bias':use_bias,'model_file':model_file,'loss':loss,
    'batch_size':batch_size,'verbosity':verbosity,'rmse_cv':False}

model=FCNN(data_params=data2b_params,model_params=model_params)
model.train()
model.plot(pdf_output=False)
```

```
Learning fingerprinted/featured data
  algorithm                     fully connected NeuralNet w/ TensorFlow
  layers                        [4, 4]
  activ_funct                   tanh
  epochs                        5000
  optimizer                     nadam
  nfold_cv                      5
Checking parameters
  all passed                    True
Read data
  data file                     fp_hoips_S2b_1dest.csv.gz
  data size                     384
  training size                 345 (89.8 %)
  test size                     39 (10.2 %)
  x dimensionality              53
  y dimensionality              1
  y label(s)                    ['prop_value']
Scaling x                       minmax
  xscaler saved in              xscaler.pkl
Scaling y                       minmax
Prepare train/test sets         random
Building model                  FCNN
Training model w/ cross validation
  cv,rmse_train,rmse_test,rmse_opt: 0 0.045158 0.112265 0.112265
  cv,rmse_train,rmse_test,rmse_opt: 1 0.048173 0.090974 0.090974
  cv,rmse_train,rmse_test,rmse_opt: 2 0.047811 0.080596 0.080596
  cv,rmse_train,rmse_test,rmse_opt: 3 0.050388 0.059932 0.059932
  cv,rmse_train,rmse_test,rmse_opt: 4 0.045543 0.106781 0.059932
  Optimal ncv:  3 ; optimal NET saved
FCNN trained, now make predictions & invert scaling
  unscaling y: minmax
    rmse training     selector1 prop_value       0.148816
    rmse training     selector2 prop_value       0.07692
  unscaling y: minmax
    rmse test         selector1 prop_value       0.130914
    rmse test         selector2 prop_value       0.172489
Predictions made & saved in "training.csv" & "test.csv"
Plot results in "training.csv" & "test.csv"
  training, (rmse & R2) = (0.119 & 0.996)
  test, (rmse & R2) = (0.156 & 0.993)
  showing prop_value
```

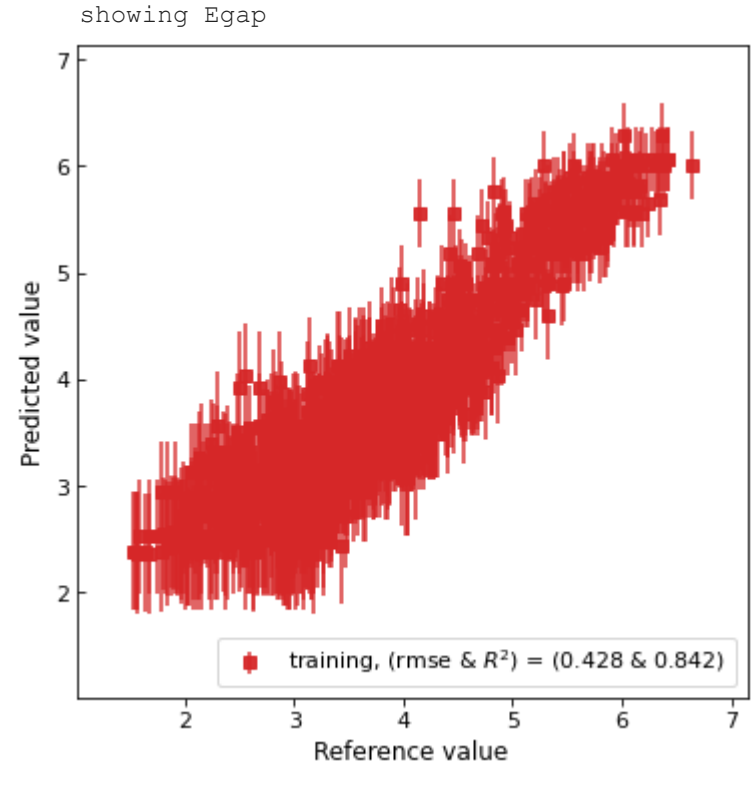


3e. Model M5: Probabilistic Neural Net on S3

In [9]:

```
from matsml.models import PrFCNN

layers=[5]
epochs=200
nfold_cv=5
use_bias=True
model_file='M5.pkl'
loss='mse'
verbosity=0
batch_size=32
activ_funct='selu'
optimizer='nadam'

model_params={'layers':layers,'activ_funct':activ_funct,'epochs':epochs,'nfold_cv':nfold_cv,
    'optimizer':optimizer,'use_bias':use_bias,'model_file':model_file,'loss':loss,'batch_size':batch_size,
    'verbosity':verbosity,'rmse_cv':False}

model=PrFCNN(data_params=data3_params,model_params=model_params)
model.train()
model.plot(pdf_output=False)
```

```
Learning fingerprinted/featured data
  algorithm                     Probabilistic NeuralNet w/ TensorFlow-Probability
  layers                        [5]
  activ_funct                   selu
  epochs                        200
  optimizer                     nadam
  nfold_cv                      5
Checking parameters
  all passed                    True
Read data
  data file                     fp_hoips_S3_4tfp.csv.gz
  data size                     1346
  training size                 1346 (100.0 %)
  test size                     0 (0.0 %)
  x dimensionality              221
  y dimensionality              1
  y label(s)                    ['Egap']
Scaling x                       minmax
  xscaler saved in              xscaler.pkl
Scaling y                       none
Prepare train/test sets         random
Building model                  PrFCNN
Training PrFCNN w/ cross validation
  cv,rmse_train,rmse_test,rmse_opt: 0 0.620635 0.582243 0.582243
  cv,rmse_train,rmse_test,rmse_opt: 1 0.603726 0.645952 0.582243
  cv,rmse_train,rmse_test,rmse_opt: 2 0.580510 0.597936 0.582243
  cv,rmse_train,rmse_test,rmse_opt: 3 0.599846 0.621374 0.582243
  cv,rmse_train,rmse_test,rmse_opt: 4 0.622166 0.654577 0.582243
  Optimal ncv:  0
PrFCNN trained, now make predictions & invert scaling
  unscaling y: none
     rmse training     Egap            0.428167
Predictions made & saved in "training.csv"

Plot results in "training.csv" & "test.csv"
  showing Egap
```



In [10]:

```
import io, requests
import matplotlib.pyplot as plt
import pandas as pd
import numpy as np
%matplotlib inline

# read the trained data
pred=pd.read_csv('training.csv')

# Trained data provide ID of 1346 cases, but we need the name of the organic cations A,
# cation B, and anions X also. They can be obtained here
sum_url='http://www.matsml.org/data/hoips2017_comp.csv'
mapping=pd.read_csv(io.StringIO(requests.get(sum_url).content.decode('utf-8')))

# We will plot 16 compositions ASnI3 made of 16 cations A and Sn for B and I for X.
organics=['Acetamidinium','Ammonium','Azetidinium','Butylammonium','Dimethylammonium','Ethylammonium',
    'Formamidinium','Guanidinium','Hydrazinium','Hydroxylammonium','Imidazolium','Isopropylammonium',
    'Methylammonium','Propylammonium','Tetramethylammonium','Trimethylammonium']
cations = ['Sn']
anions = ['I']

comps = [(organic,cation,anion) for organic in organics for cation in cations\
    for anion in anions]

# 2 DataFrame will be extracted from pred for ploting
bandgap_strs=pd.DataFrame(columns=['cid','id','organic_cat','bandgap'])
bandgap_comp=pd.DataFrame(columns=['cid','mean_comput','std_comput','mean_pred','std_pred'])

# For each of 16 compositions, extract needed data and store in "bandgap_strs" and "bandgap_comp"
for cid in range(len(comps)):
    comp = comps[cid]
    sel_rows = mapping[(mapping['organic']==comp[0]) & \
            (mapping['cation']==comp[1]) &(mapping['anion']==comp[2])]
    sel_ids = list(sel_rows['ID'])
    sel_pred = pred[pred['ID'].isin(sel_ids)]
    sel_pred.reset_index(drop=True, inplace = True)
    bandgap_comp.loc[len(bandgap_comp)]=[cid,np.mean(sel_pred['Egap']), \
            np.std(sel_pred['Egap']),sel_pred.at[0,'md_Egap'], sel_pred.at[0,'md_Egap_err']]
    for idx,eg in zip(list(sel_pred['ID']), list(sel_pred['Egap'])):
        bandgap_strs.loc[len(bandgap_strs)]=[cid,idx,comp[0],eg]


# Make figure
fig,ax = plt.subplots(figsize = (8,6),frameon=True)
plt.subplots_adjust(left=0.12, bottom=0.21, right=0.98, top=0.98, wspace=0,
        hspace=0)

plt.rcParams["font.size"] = 16

plt.box(True)
plt.tick_params(axis='x',which='both',bottom=True,top=True,labelbottom=True)
plt.tick_params(axis='y',which='both',right=True,left=True,direction='in',labelleft=True,length=5)

ax.set_ylim([0.0,4.0])

plt.tick_params(axis='x',which='both',direction='in',labelsize=12, top=True)
plt.tick_params(axis='y',which='both',direction='in',labelsize=12, right=True)
ax.set_xticks(np.arange(0, 16, 1))
ax.set_xticklabels(organics, rotation=35, ha='right')
plt.ylabel(r"$E_{\rm g}$ (eV)",color="black",fontsize=18)

ax.scatter(bandgap_strs['cid'], bandgap_strs['bandgap'],color='royalblue',
        alpha = 0.75, zorder=-3, label='computed data')

ax.errorbar(bandgap_comp['cid'],bandgap_comp['mean_comput'], yerr=bandgap_comp['std_comput'], capsize=3,
        color = 'darkgoldenrod', alpha = 0.5, zorder=-3, markersize = 5,
        fmt='s', label=r'computed $E_{\rm g}^{\rm mean} \pm E_{\rm g}^{\rm std}$')

ax.plot(bandgap_comp['cid'], bandgap_comp['mean_pred'],color='tab:red',linewidth = 2,
        label = r'predicted $E_{\rm g}^{\rm mean}$')

ax.fill_between(bandgap_comp['cid'], bandgap_comp['mean_pred'] - 2* bandgap_comp['std_pred'],
        bandgap_comp['mean_pred'] + 2* bandgap_comp['std_pred'], color = '#2ca02c', alpha = 0.15,
        label = r'predicted $E_{\rm g}^{\rm mean} \pm 2E_{\rm g}^{\rm std}$')

handles, labels = ax.get_legend_handles_labels()
labels, handles = zip(*sorted(zip(labels, handles), key=lambda t: t[0]))
ax.legend(handles, labels, loc="lower right", fontsize = 13)
```

Out[10]:

```
<matplotlib.legend.Legend at 0x7f7d9875b970>
```

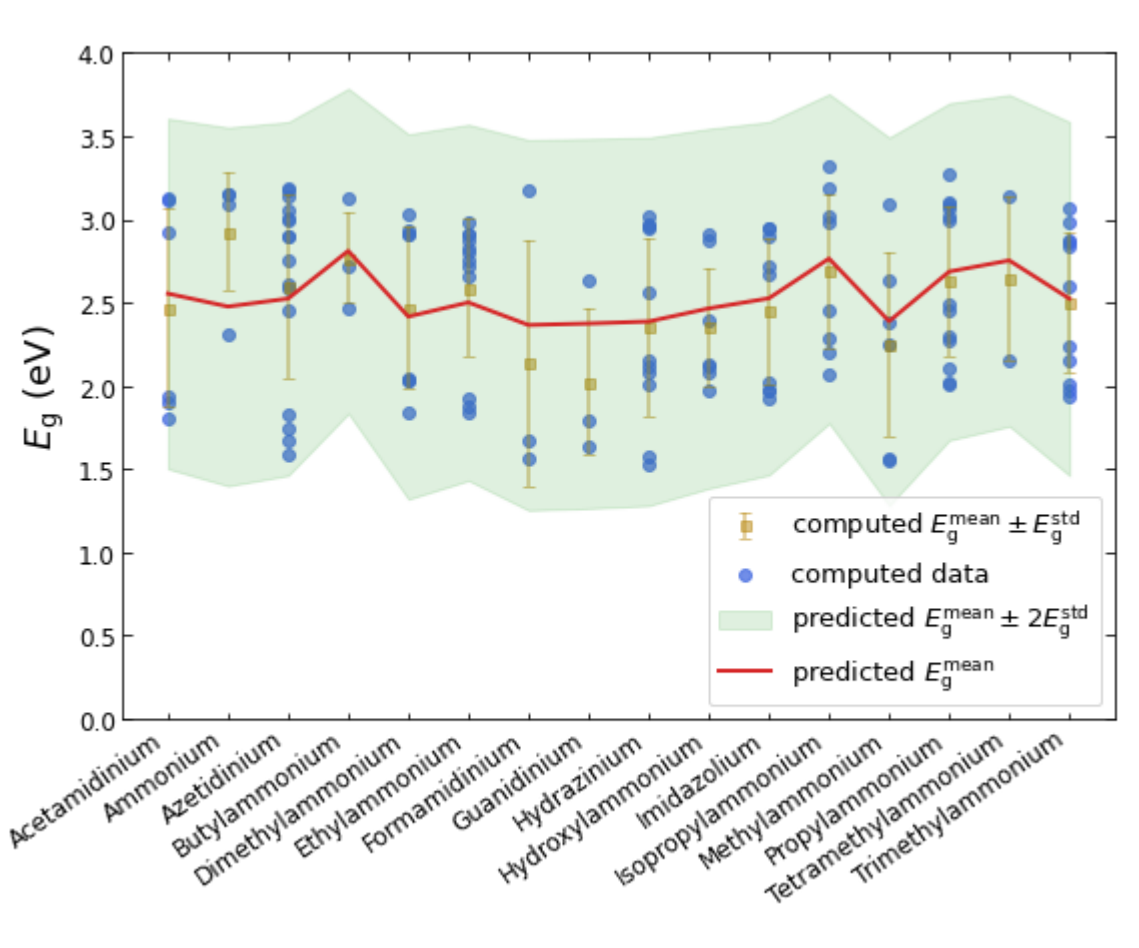


Fig. 1. Electronic band gap E_g (circles) computed for the predicted atomic structures of $ASnI_3$, 16 HOIP formulas corresponding to 16 organic cations A. For each formula, the mean and standard deviation of E_g, i.e., E_g^{mean} and E_g^{std}, are given by dark golden squares and associated errorbars. Predicted E_g^{mean} is given in red while the shaded area indicates the 95-percent confidence interval ($E_g^{mean} \pm 2E_g^{std}$) of the predictions using the probabilistic model developed in this work

In [ ]: