Predicting Perovskite Formability Using Machine Learning

CHEM404 Project Presentation

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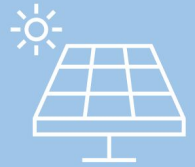
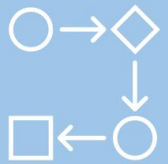
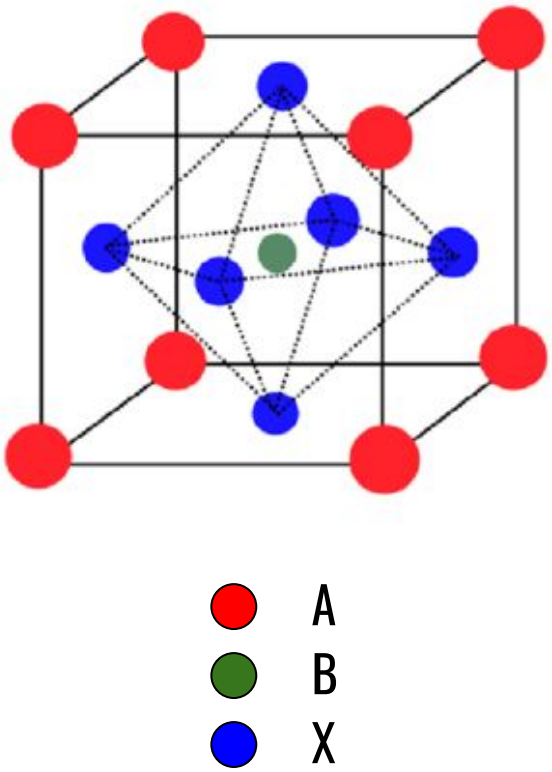
Outline

1. Introduction to Perovskites
2. Machine Learning
3. Methodology
4. Results
5. Conclusions







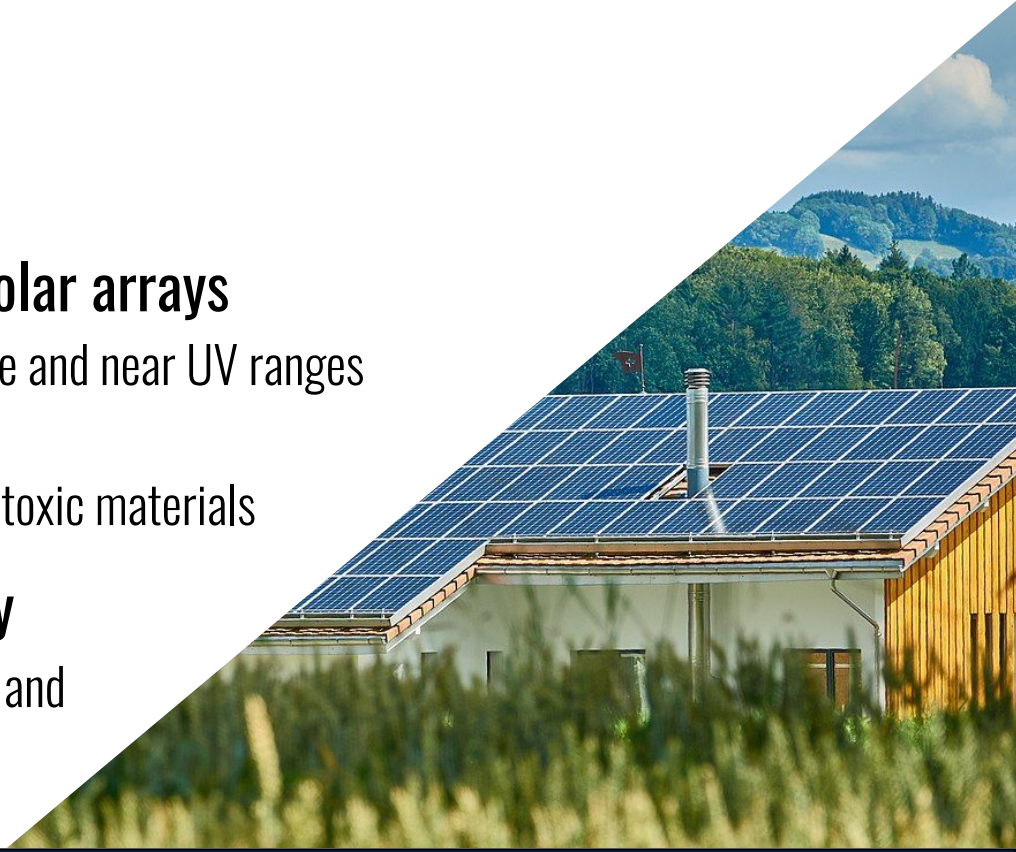
The Perovskite Structure

- Strictly a mineral of formula $CaTiO_3$
- Now used to refer to crystalline structures of ABX_3 of the same crystal structure
  - A = Large metal cation
  - B = Small metal cation
  - X = Electronegative anion (F, Cl, Br, I, O)

Tilley, R. J. D. *Perovskites: Structure-Property Relationships*; John Wiley & Sons: West Sussex, United Kingdom, **2016**.

Perovskite Applications

- Cheaper, safer, thinner, lighter solar arrays
  - Good absorbance coefficient in visible and near UV ranges
  - Long diffusion distances for excitons
  - Possibility to be made of cheap, non-toxic materials
- >20% solar conversion efficiency
  - GaAs is more efficient but expensive and potentially toxic



Chen, Y.; Zhang, L.; Zhang.; Gao, H.; Yan, H. *RSC Adv.*, **2018**, *8*, 10489-10508.

Stranks, S.D.; Eperon, G. E.; Grancini, G.; Menalaou, C.; Alcocer, M. J. P.; Leijtens, T.; Herz, L. M.; Petrozza, A.; Snaith, H. J. *Science*, **2013**, *342*, 341-344.

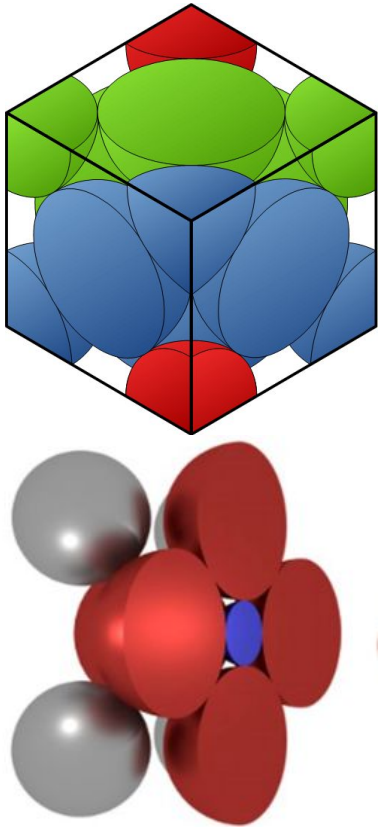
De Wolf, S.; Holovksy, J.; Moon, S. J.; Löper, P.; Niesen, B.; Ledinsky, M.; Haug, F. J.; Yum, J. H.; Ballif, C. *d*

Crystal Structure

- Not all structures of formula ABX_3 form perovskites
  - Dependent on spatial parameters: radii of ions
- Tolerance, τ, defined as:

$$\tau = \frac{r_A + r_X}{\sqrt{2}(r_B + r_X)}$$

- τ = 1 defines ideal sphere packing of cubic perovskite
  - Formability probable in range $0.813 < \tau < 1.107$
- Octahedral, o_f, factor defined as: $o_f = \frac{r_B}{r_X}$



Assadi, M. K.; Bakhoda, S.; Saidur, R.; Hanaei, H.; *Renew. Sust. Energ. Rev.*, **2017**, *81*, 2812-2822.

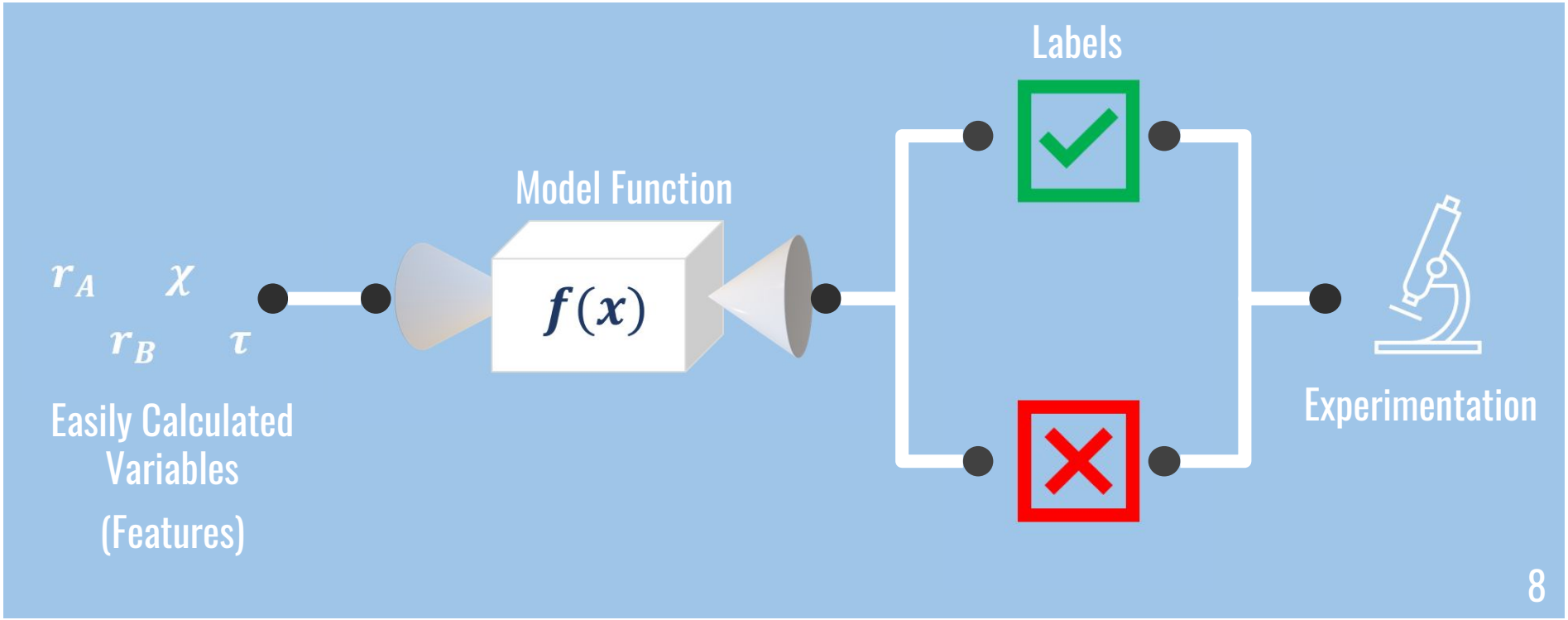
What combinations of A, B, X, will form a perovskite crystal structure?

Classification in Machine Learning

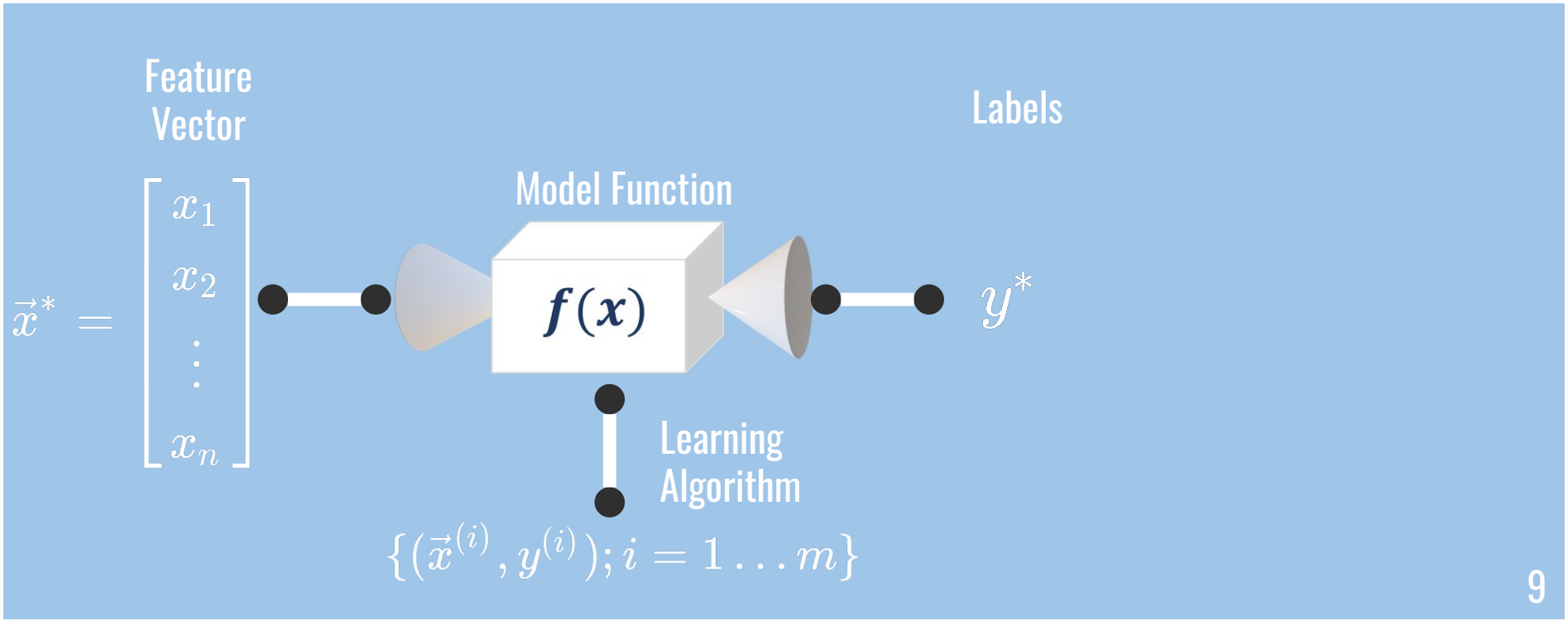
- In machine learning, classification refers to a predictive modeling problem of predicting the class of given data points.
- In mathematical language, the task is to approximate a model function f(**x**) that maps the input variables **x** (features) to discrete output variables y (labels).
- Model parameters of ML govern the position of decision boundaries, and optimal parameters are selected by evaluating the model performance (or prediction accuracy) on unseen data.

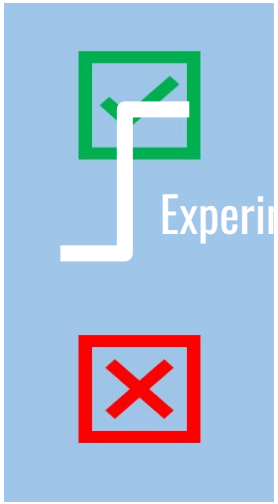
Vanderplas, J. T. *Python data science handbook: essential tools for working with data*; O'Reilly: Sebastopol, **2017**.

Workflow


Labels
Model Function
r_A χ
r_B τ
$f(x)$
Easily Calculated Variables
(Features)
Experimentation

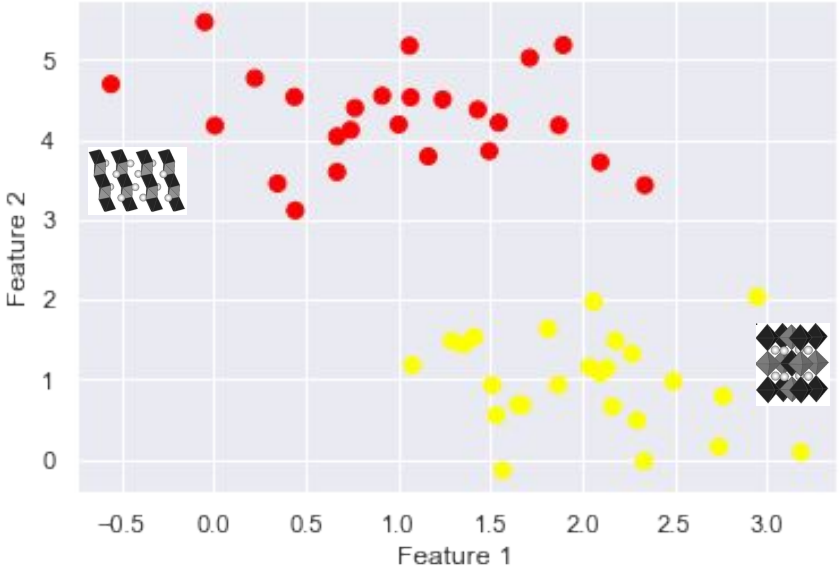
Workflow

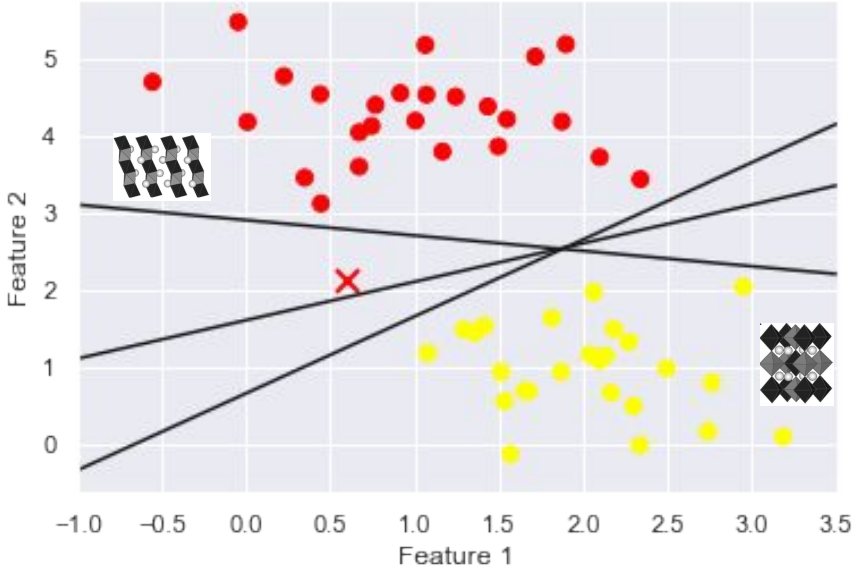

Feature Vector
Labels
Model Function
$\vec{x}^* = \begin{bmatrix} x_1 \\ x_2 \\ \vdots \\ x_n \end{bmatrix}$
$f(x)$
y^*
Learning Algorithm
$\{(\vec{x}^{(i)}, y^{(i)}); i = 1 \ldots m\}$



Vanderplas, J. T. *Python data science handbook: essential tools for working with data*; O'Reilly: Sebastopol, **2017**.

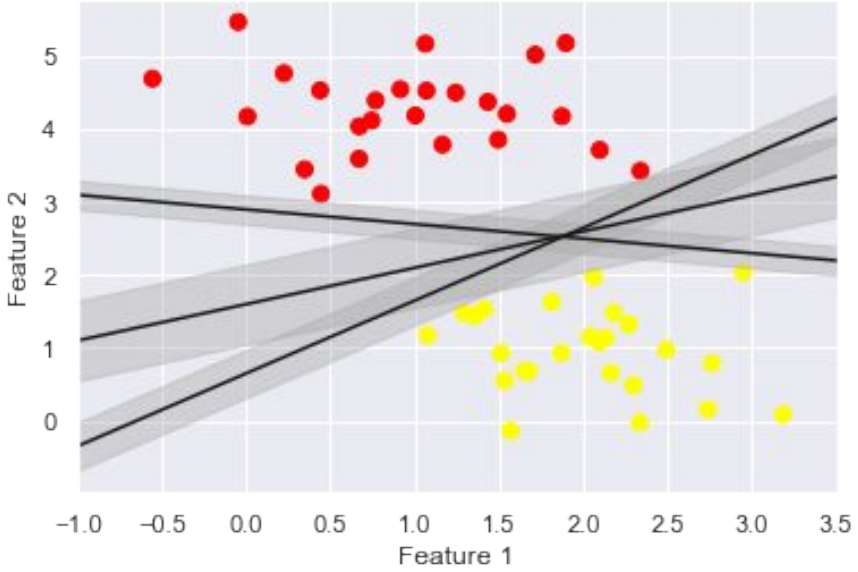
Support Vector Machine (SVM)

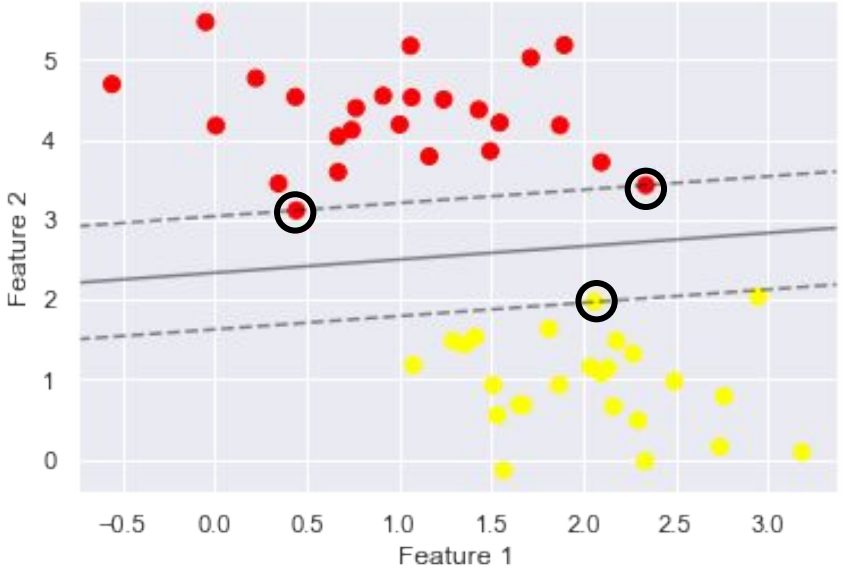




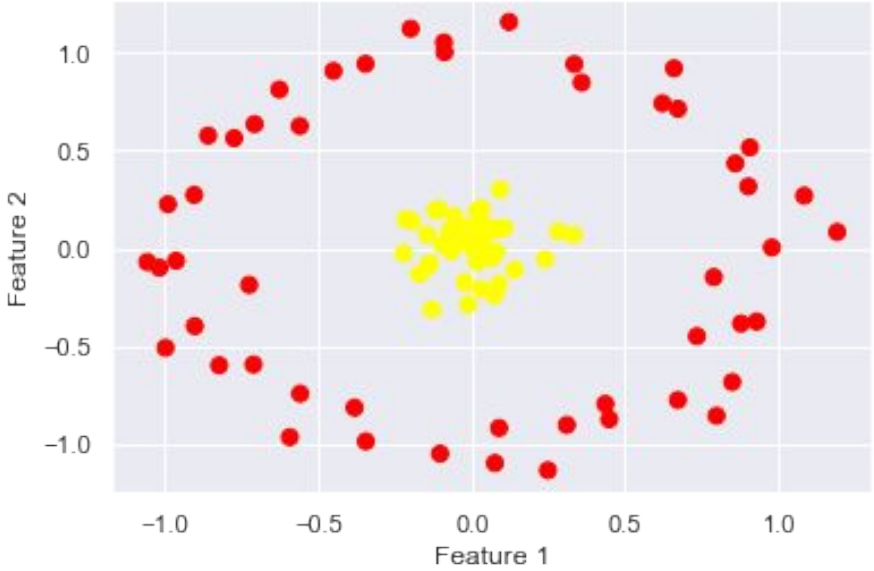
Vanderplas, J. T. *Python data science handbook: essential tools for working with data*; O'Reilly: Sebastopol, **2017**.

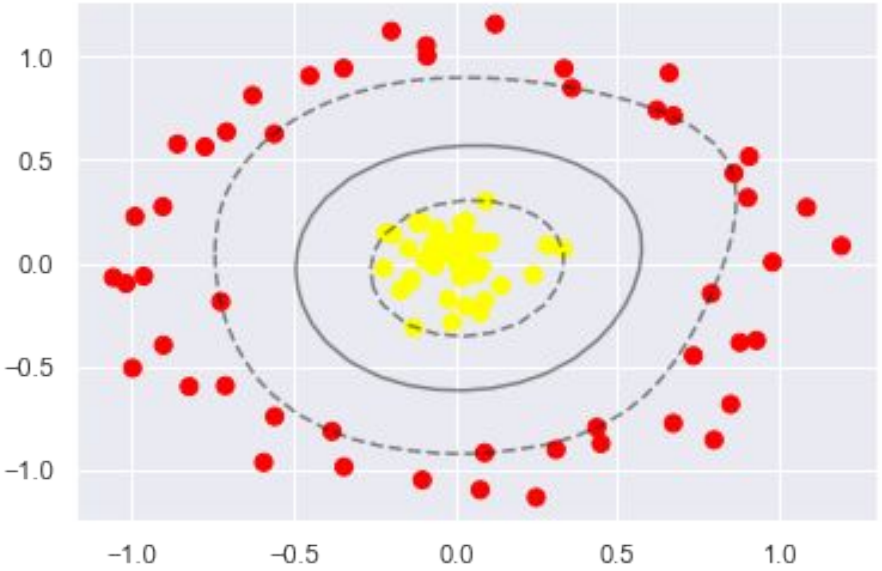
Support Vector Machine (SVM) - Linear



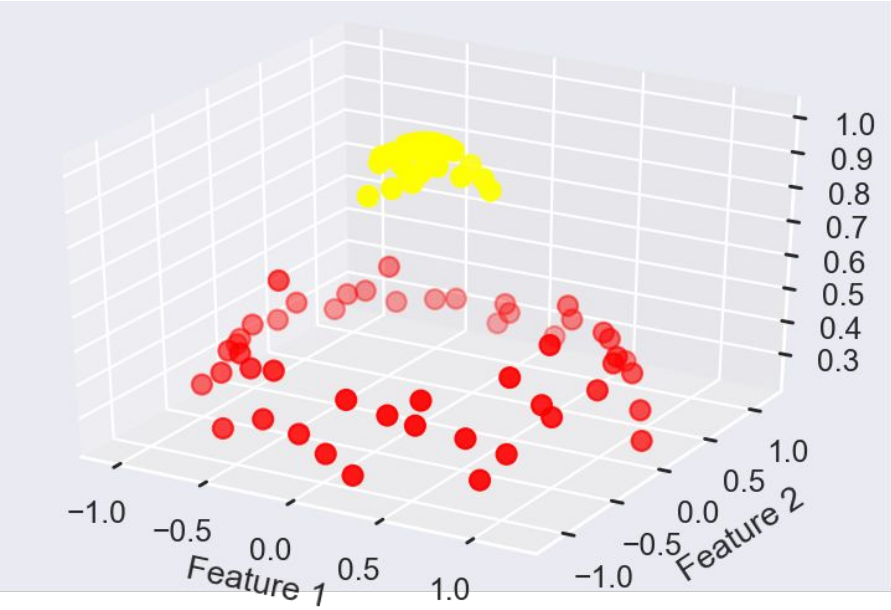


Support Vector Machine (SVM) - Non-Linear





Support Vector Machine (SVM) - Kernel Trick



Radial Basis Function (RBF):

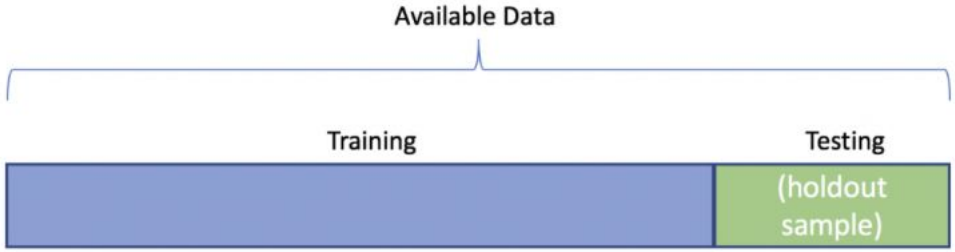
$$k(\mathbf{x}_i, \mathbf{x}_j) = \exp\left(-\gamma|\mathbf{x}_i - \mathbf{x}_j|^2\right)$$

$\mathbf{x}_i$ --- Feature vector

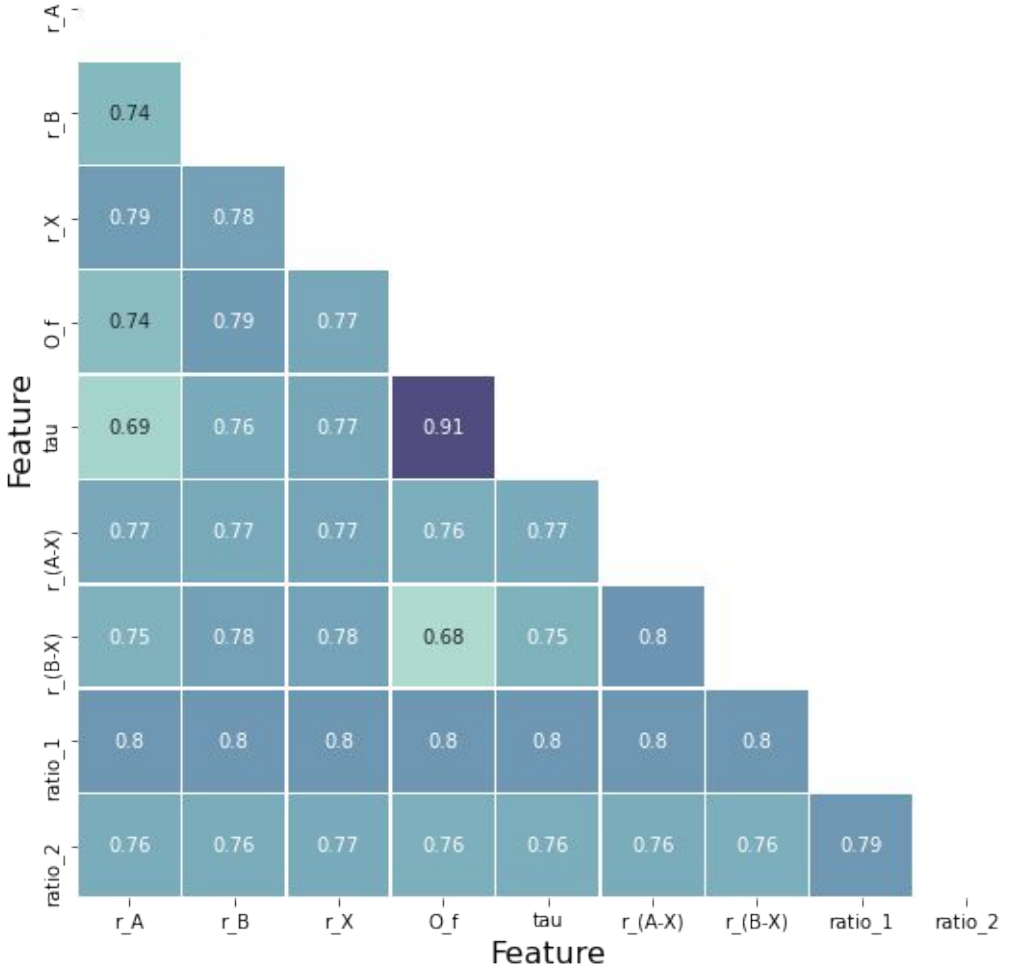
$$\gamma = \frac{1}{2\sigma^2}$$

Training Sets vs Test Sets

- Data
  - Contain the parameters and results given by experiments
- Training Set
  - Used to construct and optimize the prediction model
- Test Set
  - Used to evaluate the accuracy of the model



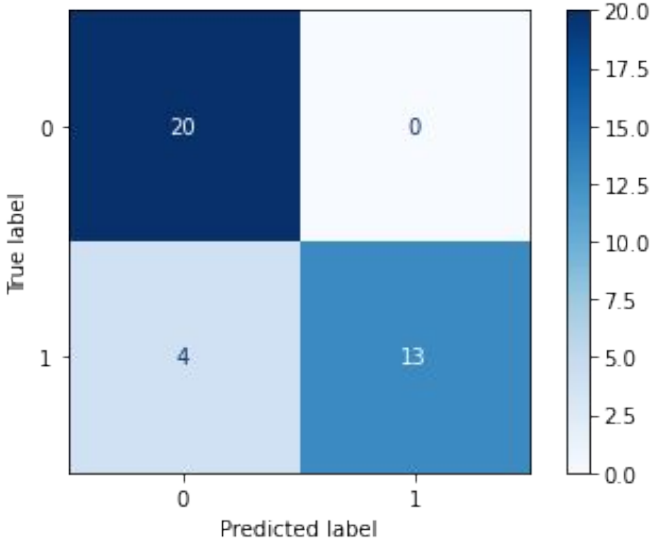
https://algotrading101.com/learn/train-test-split/



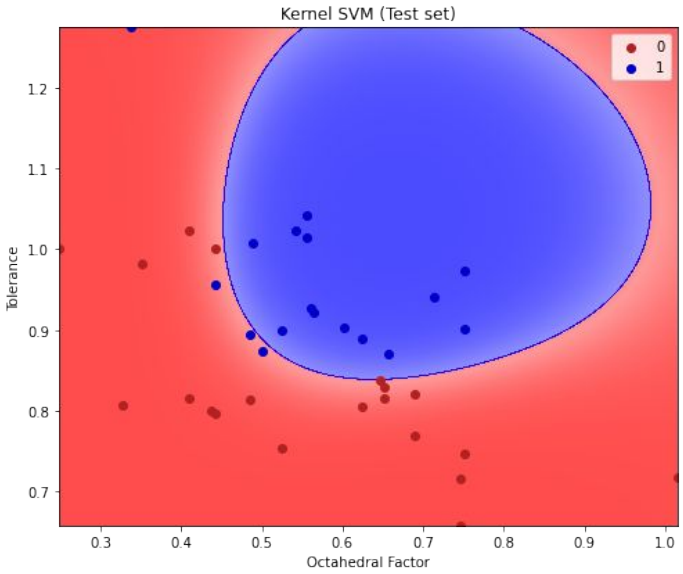
Feature Combination & Accuracy

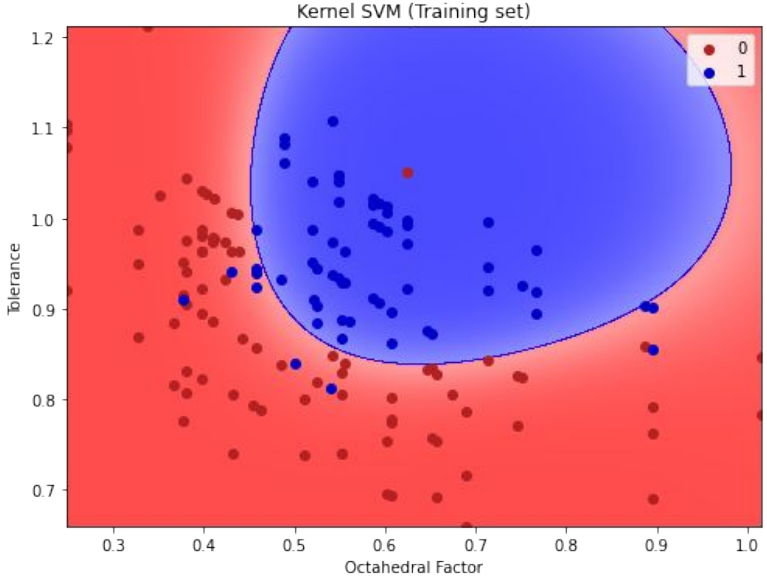
Confusion Matrix of

- Diagonal quadrants: number of correctly classified data
- Off-diagonal quadrants: number of incorrectly classified data
- Classifies all non-Perovskite compounds correctly
- Classified 13/17 Perovskite compounds correctly

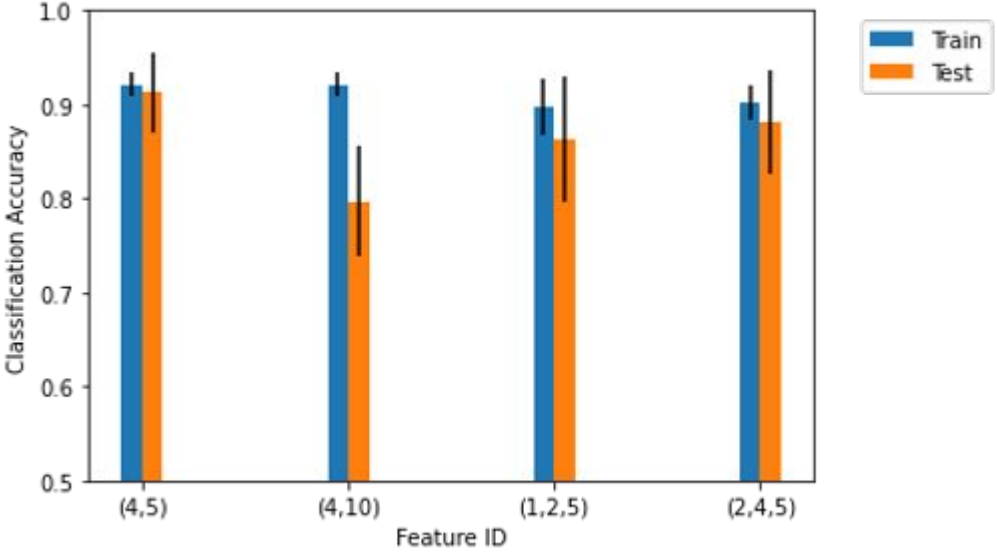


Contour of Tolerance & Octahedral Factor





Best Performing Features



1: Ionic Radii of Large Metal (r_A)
2: Ionic Radii of Small Metal (r_B)
4: Tolerance (τ)
5: Octahedral Factor ($O_h = r_B/r_X$)
10: Ratio of the sum of the s and p orbital radii r^{s+p}_A / r^{s+p}_X

Misclassifications

Most of the misclassifications are perovskite classified as non-perovskite

- Occur at the boundary
  - Predicted probabilities close to 50%
  - Non-perovskite can be synthesized in a long-lived metastable perovskite phase through non-equilibrium high pressure synthesis route.

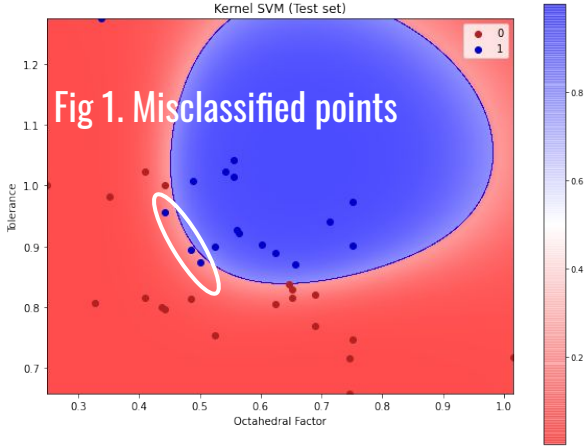
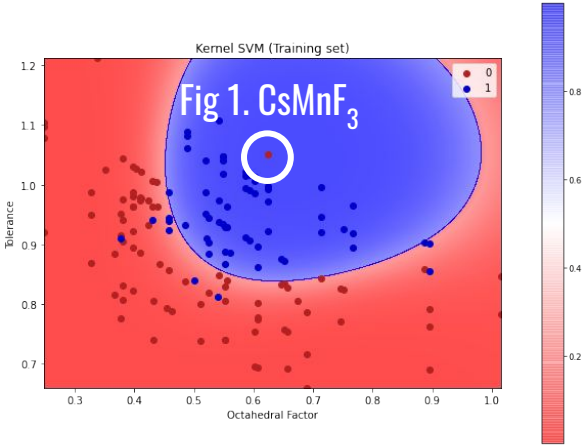


Fig 1. Misclassified points

Pilania G., Balachandran P., Kim C. and Lookman T. *Frontiers in material*, **2016**, Finding new perovskite halides via machine learning.3,19

Misclassifications

- Non-boundary misclassifications
  - $CsMnF_3$ non-perovskite as perovskite
  - Hexagonal antiferromagnetic structure of $CsMnF_3$ can be easily transformed to cubic perovskite at high pressure



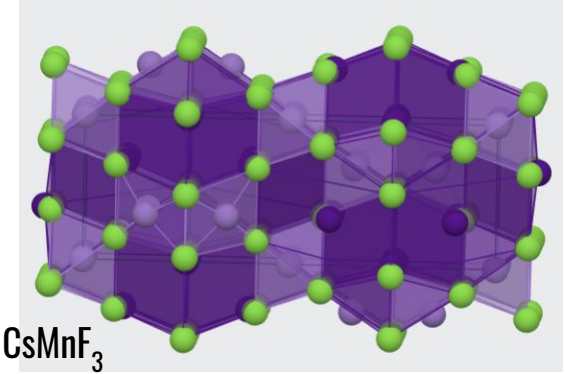


Fig.2 $CsMnF_3$

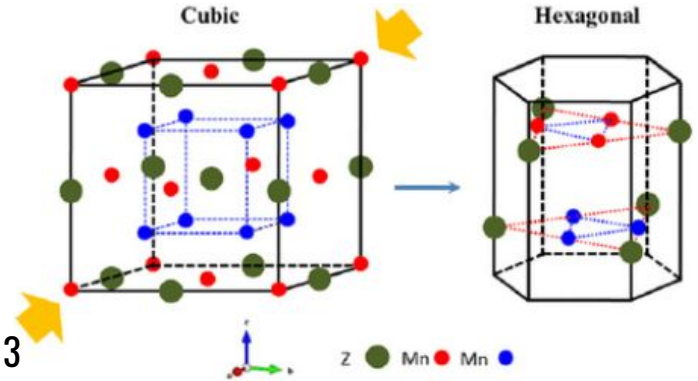


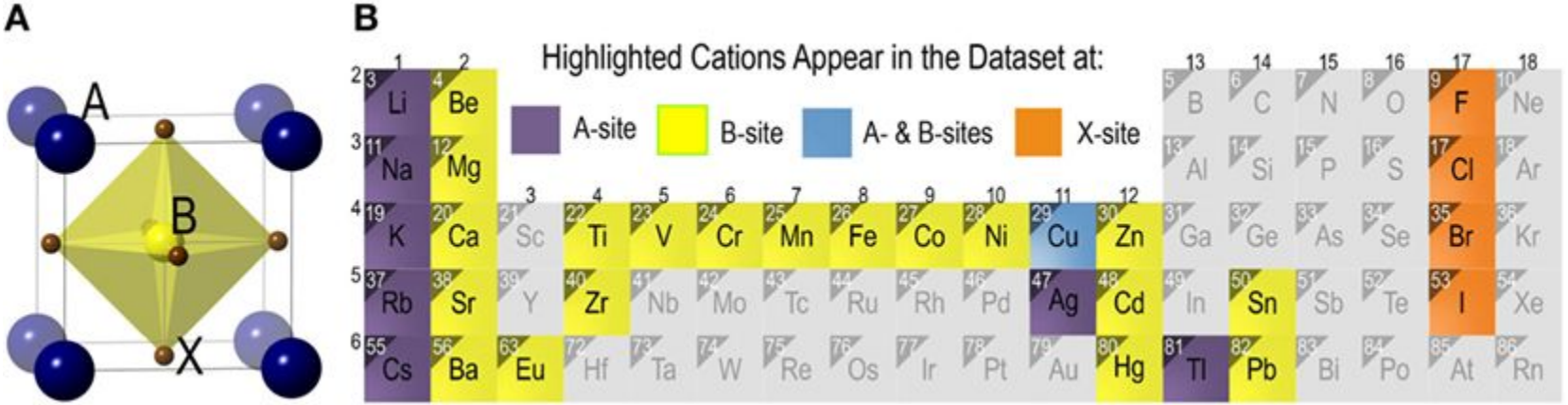
Fig. 3

Kafalas, J. and Longo J. *Solid state Chem* **1972** High pressure synthesis of (ABX3) (AX) n compounds. 4, 55

Zhang D., Yan B., Wu S., Kubler J., Kreiner G.,Parkin S and Felser C. *Journal of physics: Condensed matter*, **2013** First-principles study of the structural stability of cubic, tetragonal and hexagonal phases in Mn3z (Z = Ga, Sn and Ge) Heusler compounds, 25,206006

Implication

- Small dataset
  - 640 possible compositions
  - 185 know data
    - 8 A-site, 20 B-site, 4 X-site



Pilania G., Balachandran P., Kim C. and Lookman T. *Frontiers in material*, **2016**, Finding new perovskite halides via machine learning.3,19

New tolerance, τ, factor

$$\tau = \frac{r_X}{r_B} - n_A\left(n_A - \frac{r_A/r_B}{\ln(r_A/r_B)}\right)$$

n_A : oxidation state of A
R_A : ionic radius of ion A
R_B : ionic radius of ion B
- $r_A > r_B$ by definition

Bartel C., Sutton C., et al. *Materials science* **2019** New tolerance factor to predict the stability of perovskite oxides and halides. 5,2, eaav0693

New tolerance, τ, factor

$$\tau = \frac{r_X}{r_B} - n_A\left(n_A - \frac{r_A/r_B}{\ln(r_A/r_B)}\right)$$

Why it's better?

- Better sum of ionic radii estimates the interatomic bond distances for the structure
- Refined input radii and increased the dimensionality of the descriptor
- Monotonic dependency

Bartel C., Sutton C., et al. *Materials science* **2019** New tolerance factor to predict the stability of perovskite oxides and halides. 5,2, eaav0693

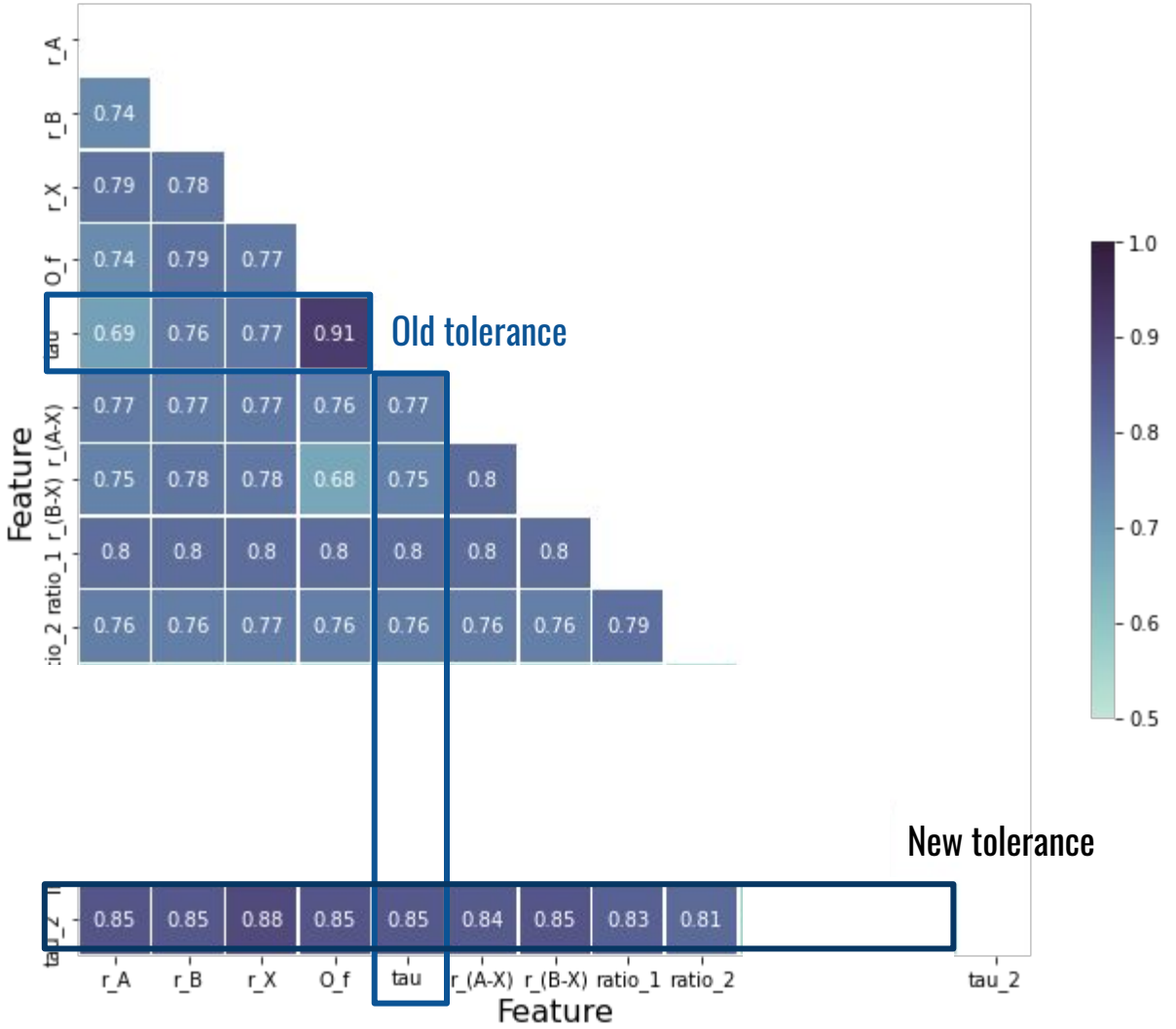
New tolerance, τ, factor

$$\tau = \frac{r_X}{r_B} - n_A\left(n_A - \frac{r_A/r_B}{\ln(r_A/r_B)}\right)$$

How it's better?

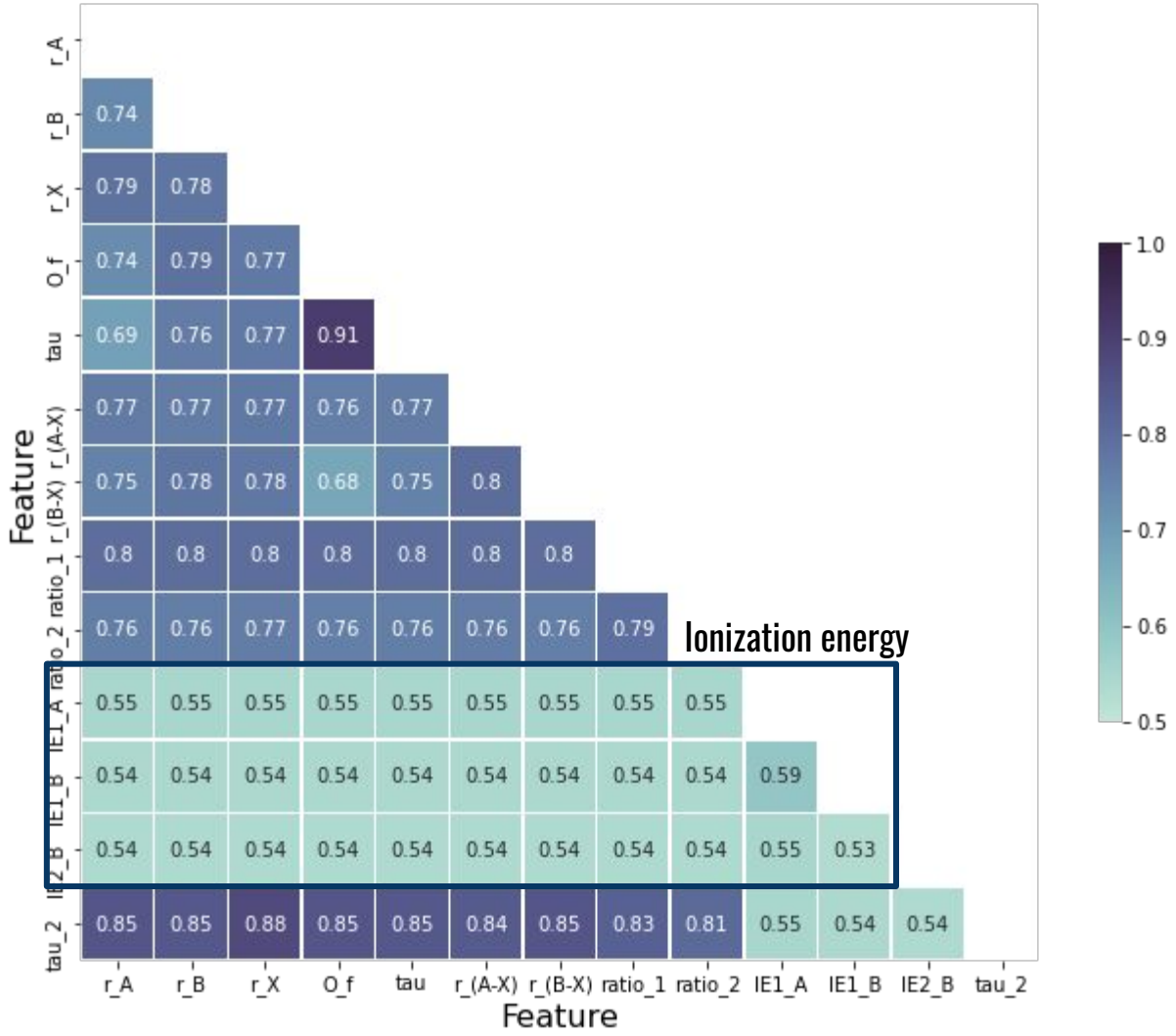
- Reduce false-positive prediction occurrence
- Increase the overall classification accuracy

Bartel C., Sutton C., et al. *Materials science* **2019** New tolerance factor to predict the stability of perovskite oxides and halides. 5,2, eaav0693



New Feature Combination & Accuracy

New tolerance



New Feature Combination & Accuracy

Ionization energy

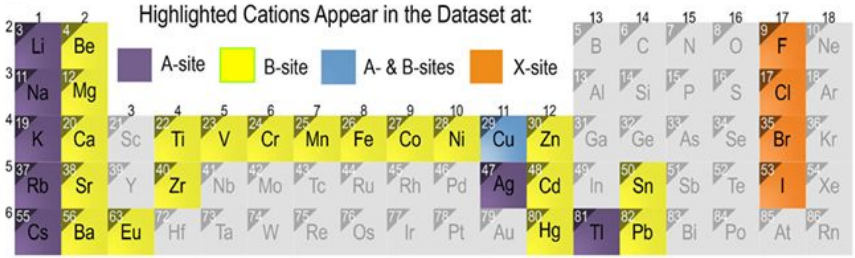
Limitations

- Data are highly repetitive
  - Only 4 different X-sites and a few A-sites and B-sites
- Data are similar
  - Close in periodic table, has similar structure and properties

No	System	Formabili	Assignme	1. Ra	2. Rb	3. Rx	4. t	5. of	miu	Ax	Bx	Xx	E(A-X)	8.(Ra
1	LiF–MgF2	No	0	1.13	0.72	1.33	0.849	0.54135	0.541	0.9	1.31	3.78	-2.88	-2.0
2	LiF–ZnF2	No	0	1.13	0.74	1.33	0.84	0.55639	0.556	0.9	1.44	3.78	-2.88	-1.9
3	LiF–MnF2	No	0	1.13	0.83	1.33	0.805	0.62406	0.624	0.9	2.04	3.78	-2.88	-1.4
4	LiF–CaF2	No	0	1.13	1	1.33	0.747	0.75188	0.752	0.9	1.17	3.78	-2.88	-2.2
5	LiF–PbF2	No	0	1.13	1.19	1.33	0.69	0.89474	0.895	0.9	1.92	3.78	-2.88	-1.5
6	NaF–NiF2	Yes	1	1.39	0.69	1.33	0.952	0.5188	0.519	0.89	1.76	3.78	-2.89	-2.1
7	NaF–MgF2	Yes	1	1.39	0.72	1.33	0.938	0.54135	0.541	0.89	1.31	3.78	-2.89	-2.5
8	NaF–CuF2	Yes	1	1.39	0.73	1.33	0.934	0.54887	0.549	0.89	1.08	3.78	-2.89	-2.8
9	NaF–ZnF2	Yes	1	1.39	0.74	1.33	0.929	0.55639	0.556	0.89	1.44	3.78	-2.89	-2.4
10	NaF–CoF2	Yes	1	1.39	0.745	1.33	0.927	0.56015	0.56	0.89	1.72	3.78	-2.89	-2.1
11	NaF–FeF2	Yes	1	1.39	0.78	1.33	0.912	0.58647	0.586	0.89	1.67	3.78	-2.89	-2.2
12	NaF–VF2	Yes	1	1.39	0.79	1.33	0.907	0.59398	0.594	0.89	2.22	3.78	-2.89	-1.6
13	NaF–CrF2	Yes	1	1.39	0.8	1.33	0.903	0.6015	0.602	0.89	2	3.78	-2.89	-1.8
14	NaF–MnF2	Yes	1	1.39	0.83	1.33	0.89	0.62406	0.624	0.89	2.04	3.78	-2.89	-1.8
15	NaF–CdF2	No	0	1.39	0.95	1.33	0.844	0.71429	0.714	0.89	1.4	3.78	-2.89	-2.4
16	NaF–CaF2	No	0	1.39	1	1.33	0.825	0.75188	0.752	0.89	1.17	3.78	-2.89	-2.7
17	NaF–PbF2	No	0	1.39	1.19	1.33	0.763	0.89474	0.895	0.89	1.92	3.78	-2.89	-1.9
18	NaF–BaF2	No	0	1.39	1.35	1.33	0.718	1.01504	1.01	0.89	1.08	3.78	-2.89	-2.8
19	AgF–CoF2	Yes	1	1.49	0.65	1.33	1.007	0.48872	0.489	1.07	1.72	3.78	-2.71	-2.3
20	AgF–NiF2	Yes	1	1.49	0.69	1.33	0.987	0.5188	0.519	1.07	1.76	3.78	-2.71	-2
21	AgF–MgF2	Yes	1	1.49	0.72	1.33	0.973	0.54135	0.541	1.07	1.31	3.78	-2.71	-2.7
22	AgF–ZnF2	Yes	1	1.49	0.74	1.33	0.963	0.55639	0.556	1.07	1.44	3.78	-2.71	-2.6
23	AgF–MnF2	Yes	1	1.49	0.83	1.33	0.923	0.62406	0.624	1.07	2.04	3.78	-2.71	-1.9
24	AgF–PbF2	No	0	1.49	1.19	1.33	0.791	0.89474	0.895	1.07	1.92	3.78	-2.71	-2.0
25	KF–CoF2	Yes	1	1.64	0.65	1.33	1.061	0.48872	0.489	0.8	1.72	3.78	-2.98	-2.5
26	KF–NiF2	Yes	1	1.64	0.69	1.33	1.04	0.5188	0.519	0.8	1.76	3.78	-2.98	-2.4
27	KF–MgF2	Yes	1	1.64	0.72	1.33	1.024	0.54135	0.541	0.8	1.31	3.78	-2.98	-3.0
28	KF–CuF2	Yes	1	1.64	0.73	1.33	1.019	0.54887	0.549	0.8	1.08	3.78	-2.98	-3.3
29	KF–ZnF2	Yes	1	1.64	0.74	1.33	1.015	0.55639	0.556	0.8	1.44	3.78	-2.98	-2.8
30	KF–FeF2	Yes	1	1.64	0.78	1.33	0.995	0.58647	0.586	0.8	1.67	3.78	-2.98	-2.6
31	KF–VF2	Yes	1	1.64	0.79	1.33	0.991	0.59398	0.594	0.8	2.22	3.78	-2.98	-1.9
32	KF–CrF2	Yes	1	1.64	0.8	1.33	0.986	0.6015	0.602	0.8	2	3.78	-2.98	-2.1
33	KF–MnF2	Yes	1	1.64	0.83	1.33	0.972	0.62406	0.624	0.8	2.04	3.78	-2.98	-2.1
34	KF–CdF2	Yes	1	1.64	0.95	1.33	0.921	0.71429	0.714	0.8	1.4	3.78	-2.98	-2.9
35	KF–CaF2	Yes	1	1.64	1	1.33	0.901	0.75188	0.752	0.8	1.17	3.78	-2.98	-3.2
36	KF–HgF2	Yes	1	1.64	1.02	1.33	0.894	0.76692	0.767	0.8	1.49	3.78	-2.98	-2.8
37	KF–BaF2	No	0	1.64	1.35	1.33	0.784	1.01504	1.015	0.8	1.08	3.78	-2.98	-3.3

Limitations

- Data are highly repetitive
  - Only 4 different X-sites and a few A-sites and B-sites
- Data are similar
  - Close in periodic table, has similar structure and properties

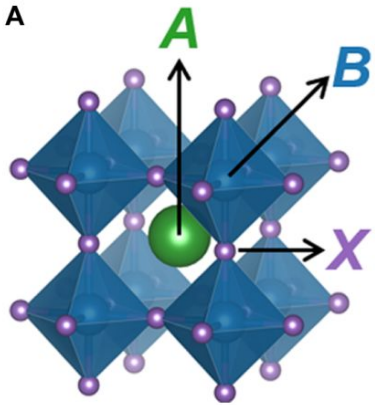


No	System	Formabili	Assignme	1. Ra	2. Rb	3. Rx	4. t	5. of	miu	Ax	Bx	Xx	E(A-X)	8.(Ra
1	LiF–MgF2	No	0	1.13	0.72	1.33	0.849	0.54135	0.541	0.9	1.31	3.78	-2.88	-2.0
2	LiF–ZnF2	No	0	1.13	0.74	1.33	0.84	0.55639	0.556	0.9	1.44	3.78	-2.88	-1.9
3	LiF–MnF2	No	0	1.13	0.83	1.33	0.805	0.62406	0.624	0.9	2.04	3.78	-2.88	-1.4
4	LiF–CaF2	No	0	1.13	1	1.33	0.747	0.75188	0.752	0.9	1.17	3.78	-2.88	-2.2
5	LiF–PbF2	No	0	1.13	1.19	1.33	0.69	0.89474	0.895	0.9	1.92	3.78	-2.88	-1.5
6	NaF–NiF2	Yes	1	1.39	0.69	1.33	0.952	0.5188	0.519	0.89	1.76	3.78	-2.89	-2.1
7	NaF–MgF2	Yes	1	1.39	0.72	1.33	0.938	0.54135	0.541	0.89	1.31	3.78	-2.89	-2.5
8	NaF–CuF2	Yes	1	1.39	0.73	1.33	0.934	0.54887	0.549	0.89	1.08	3.78	-2.89	-2.8
9	NaF–ZnF2	Yes	1	1.39	0.74	1.33	0.929	0.55639	0.556	0.89	1.44	3.78	-2.89	-2.4
10	NaF–CoF2	Yes	1	1.39	0.745	1.33	0.927	0.56015	0.56	0.89	1.72	3.78	-2.89	-2.1
11	NaF–FeF2	Yes	1	1.39	0.78	1.33	0.912	0.58647	0.586	0.89	1.67	3.78	-2.89	-2.2
12	NaF–VF2	Yes	1	1.39	0.79	1.33	0.907	0.59398	0.594	0.89	2.22	3.78	-2.89	-1.6
13	NaF–CrF2	Yes	1	1.39	0.8	1.33	0.903	0.6015	0.602	0.89	2	3.78	-2.89	-1.8
14	NaF–MnF2	Yes	1	1.39	0.83	1.33	0.89	0.62406	0.624	0.89	2.04	3.78	-2.89	-1.8
15	NaF–CdF2	No	0	1.39	0.95	1.33	0.844	0.71429	0.714	0.89	1.4	3.78	-2.89	-2.4
16	NaF–CaF2	No	0	1.39	1	1.33	0.825	0.75188	0.752	0.89	1.17	3.78	-2.89	-2.7
17	NaF–PbF2	No	0	1.39	1.19	1.33	0.763	0.89474	0.895	0.89	1.92	3.78	-2.89	-1.9
18	NaF–BaF2	No	0	1.39	1.35	1.33	0.718	1.01504	1.01	0.89	1.08	3.78	-2.89	-2.8
19	AgF–CoF2	Yes	1	1.49	0.65	1.33	1.007	0.48872	0.489	1.07	1.72	3.78	-2.71	-2.3
20	AgF–NiF2	Yes	1	1.49	0.69	1.33	0.987	0.5188	0.519	1.07	1.76	3.78	-2.71	-2
21	AgF–MgF2	Yes	1	1.49	0.72	1.33	0.973	0.54135	0.541	1.07	1.31	3.78	-2.71	-2.7
22	AgF–ZnF2	Yes	1	1.49	0.74	1.33	0.963	0.55639	0.556	1.07	1.44	3.78	-2.71	-2.6
23	AgF–MnF2	Yes	1	1.49	0.83	1.33	0.923	0.62406	0.624	1.07	2.04	3.78	-2.71	-1.9
24	AgF–PbF2	No	0	1.49	1.19	1.33	0.791	0.89474	0.895	1.07	1.92	3.78	-2.71	-2.0
25	KF–CoF2	Yes	1	1.64	0.65	1.33	1.061	0.48872	0.489	0.8	1.72	3.78	-2.98	-2.5
26	KF–NiF2	Yes	1	1.64	0.69	1.33	1.04	0.5188	0.519	0.8	1.76	3.78	-2.98	-2.4
27	KF–MgF2	Yes	1	1.64	0.72	1.33	1.024	0.54135	0.541	0.8	1.31	3.78	-2.98	-3.0
28	KF–CuF2	Yes	1	1.64	0.73	1.33	1.019	0.54887	0.549	0.8	1.08	3.78	-2.98	-3.3
29	KF–ZnF2	Yes	1	1.64	0.74	1.33	1.015	0.55639	0.556	0.8	1.44	3.78	-2.98	-2.8
30	KF–FeF2	Yes	1	1.64	0.78	1.33	0.995	0.58647	0.586	0.8	1.67	3.78	-2.98	-2.6
31	KF–VF2	Yes	1	1.64	0.79	1.33	0.991	0.59398	0.594	0.8	2.22	3.78	-2.98	-1.9
32	KF–CrF2	Yes	1	1.64	0.8	1.33	0.986	0.6015	0.602	0.8	2	3.78	-2.98	-2.1
33	KF–MnF2	Yes	1	1.64	0.83	1.33	0.972	0.62406	0.624	0.8	2.04	3.78	-2.98	-2.1
34	KF–CdF2	Yes	1	1.64	0.95	1.33	0.921	0.71429	0.714	0.8	1.4	3.78	-2.98	-2.9
35	KF–CaF2	Yes	1	1.64	1	1.33	0.901	0.75188	0.752	0.8	1.17	3.78	-2.98	-3.2
36	KF–HgF2	Yes	1	1.64	1.02	1.33	0.894	0.76692	0.767	0.8	1.49	3.78	-2.98	-2.8
37	KF–BaF2	No	0	1.64	1.35	1.33	0.784	1.01504	1.015	0.8	1.08	3.78	-2.98	-3.3

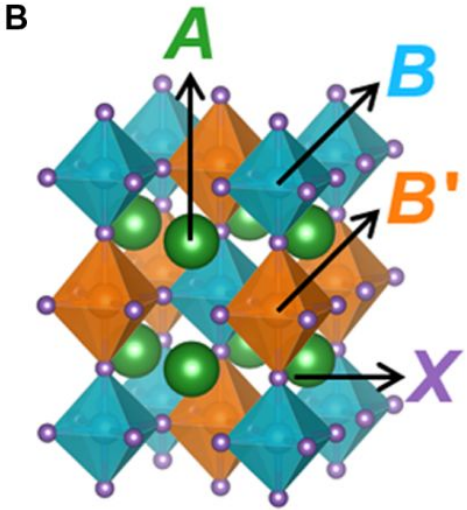
Conclusion

- Best performed features
  - Tolerance
  - Octahedral factor

- Future study
  - Double perovskite



perovskite



Double perovskite

Bartel C., Sutton C., et al. *Materials science* **2019** New tolerance factor to predict the stability of perovskite oxides and halides. 5,2, eaav0693

Thank you.

Questions?