Data analytics approaches to materials design: Critical role of a descriptor

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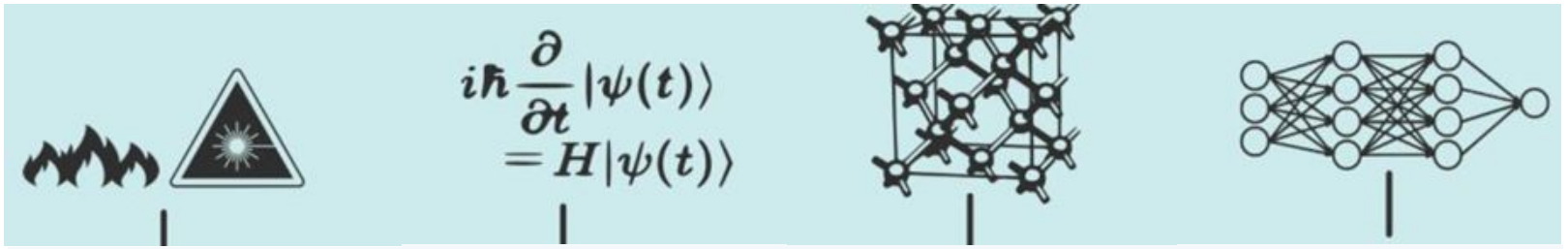
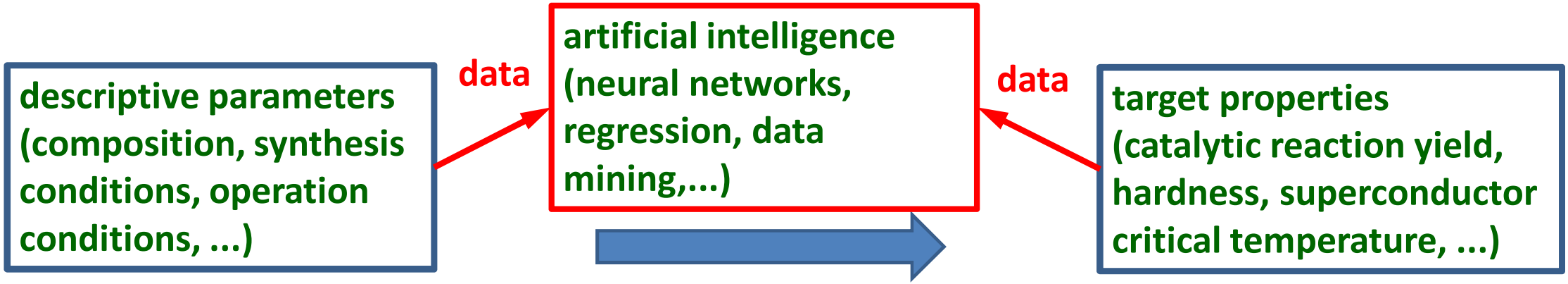
Moscow, Russia

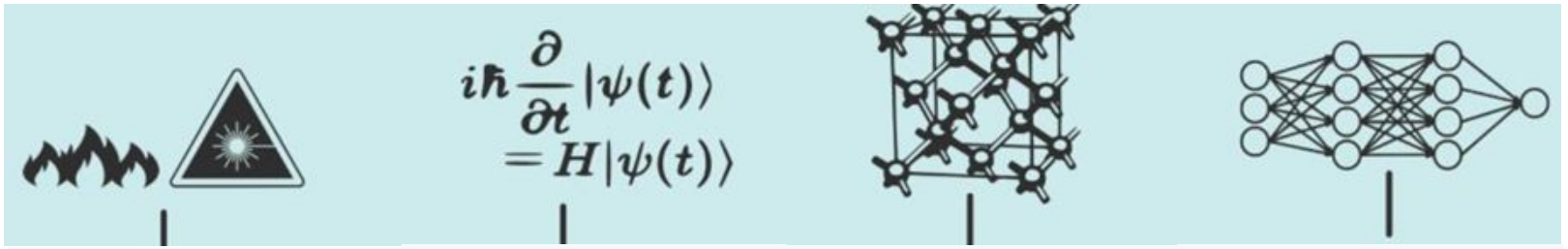
Research paradigm shift



Empirical science	Theoretical Science	Computational science	Data science
Experiments	Laws of classical mechanics, electrodynamics, etc.	Monte Carlo; molecular dynamics; density-functional theory and beyond	Detection of patterns and anomalies in Big Data; artificial intelligence; etc.



Research paradigm shift



Empirical science	Theoretical Science	Computational science	Data science
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descriptive parameters (composition, synthesis conditions, operation conditions, ...)

data

artificial intelligence scikit-learn (https://scikit-learn.org/stable/)

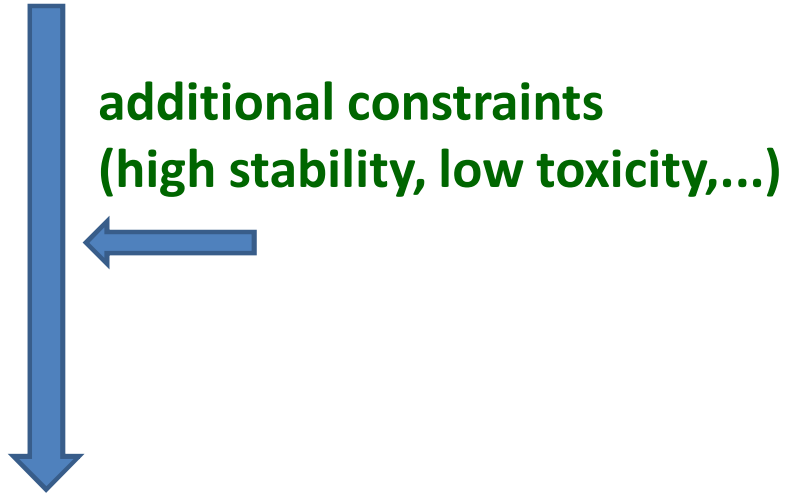
data

target properties (catalytic reaction yield, hardness, superconductor critical temperature, ...)

High-throughput computational materials design

Top-down design:

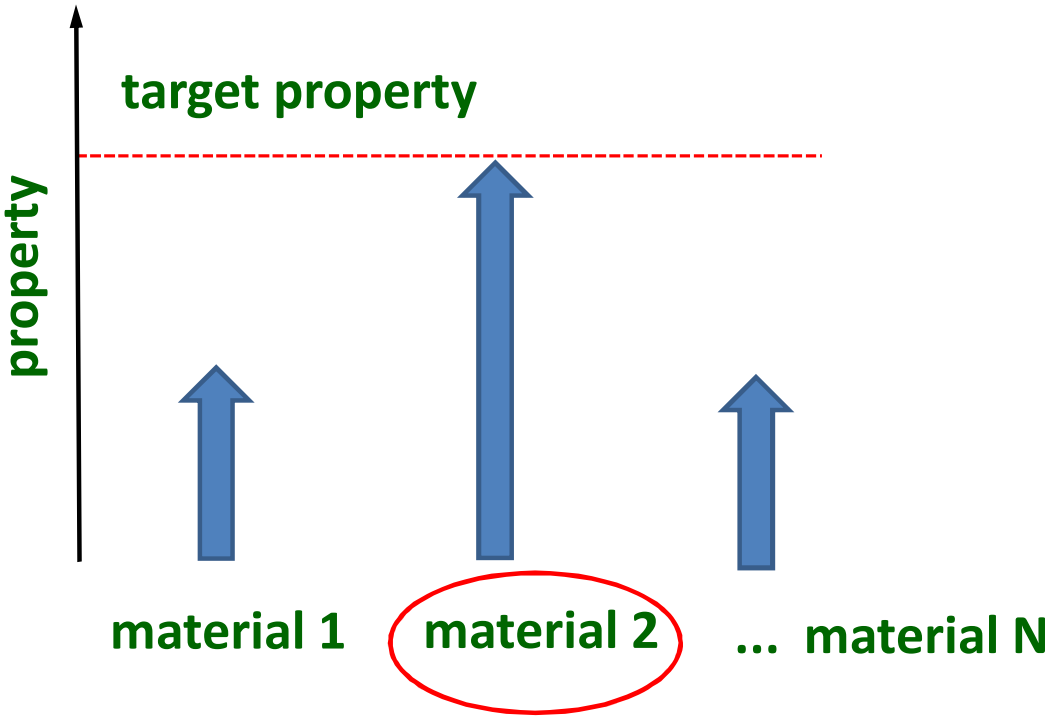
target property (high activity and selectivity of a catalyst)



synthesis recipe

not clear how to achieve this!

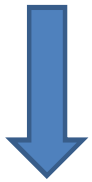
Bottom-up design:



high-throughput screening

The key issue: Complexity

$$i\frac{\partial\Psi(\boldsymbol{x}_1,\boldsymbol{x}_2,\ldots,\boldsymbol{x}_n,\boldsymbol{R}_1,\boldsymbol{R}_2,\ldots,\boldsymbol{R}_N,t)}{\partial t}=\hat{H}(t)\Psi(\boldsymbol{x}_1,\boldsymbol{x}_2,\ldots,\boldsymbol{x}_n,\boldsymbol{R}_1,\boldsymbol{R}_2,\ldots,\boldsymbol{R}_N,t)$$

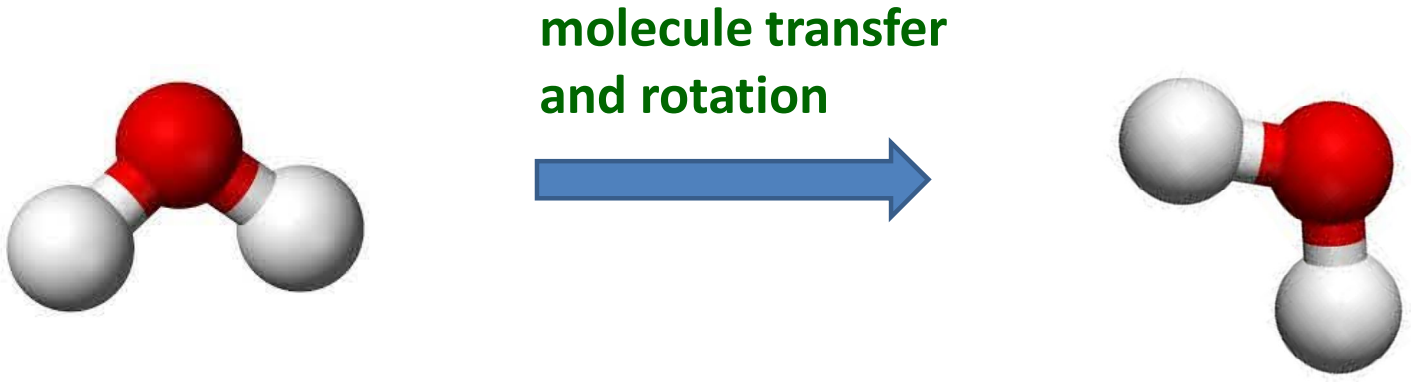


1) **Many-body problem ($3(n+N)$-dimensional)**

2) **Multiscale problem (tens orders of magnitude in time and space)**

However, there is hope that the complexity can be treated *incrementally*

Including science in descriptors



structure descriptor: Cartesian coordinates → changes, but properties do not change!

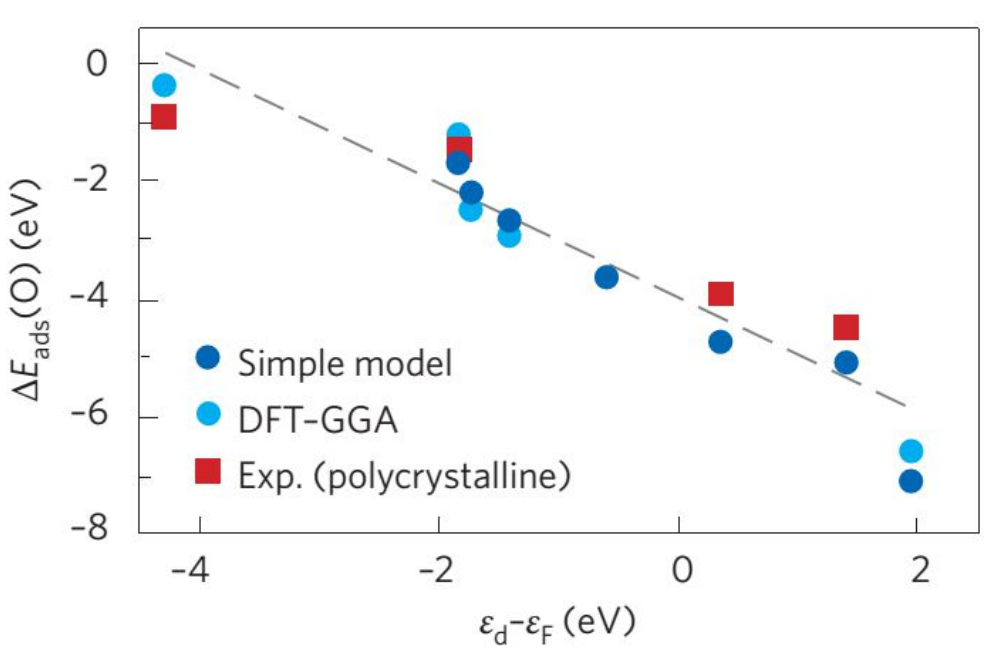
descriptive parameters (composition, synthesis conditions, operation conditions)

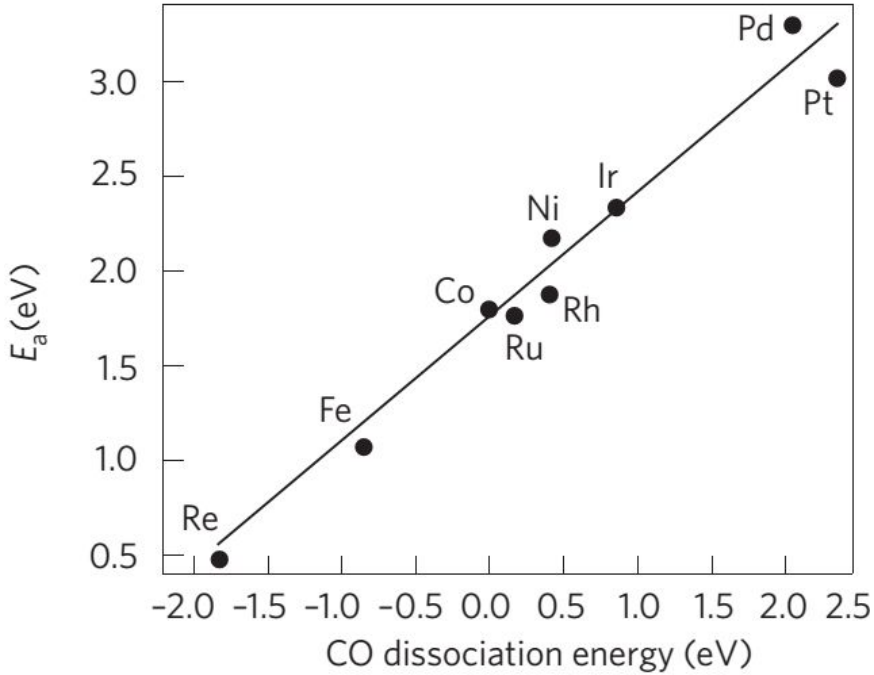
→

artificial intelligence (neural networks, regression, data mining,...)

machine will learn symmetries, not (other) physics -- much more data will be needed for an accurate model

Descriptors



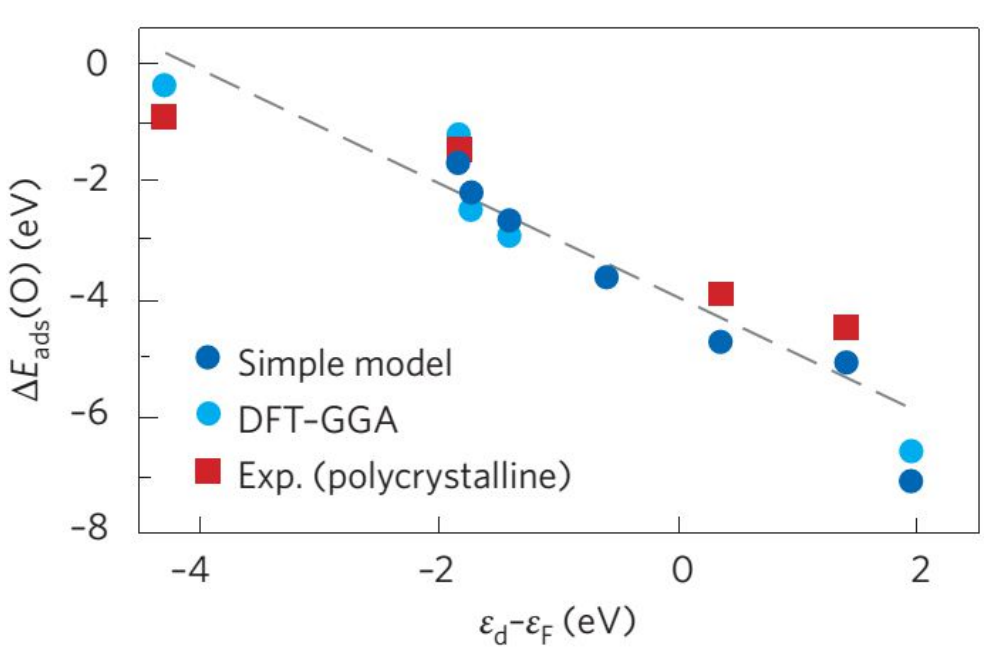


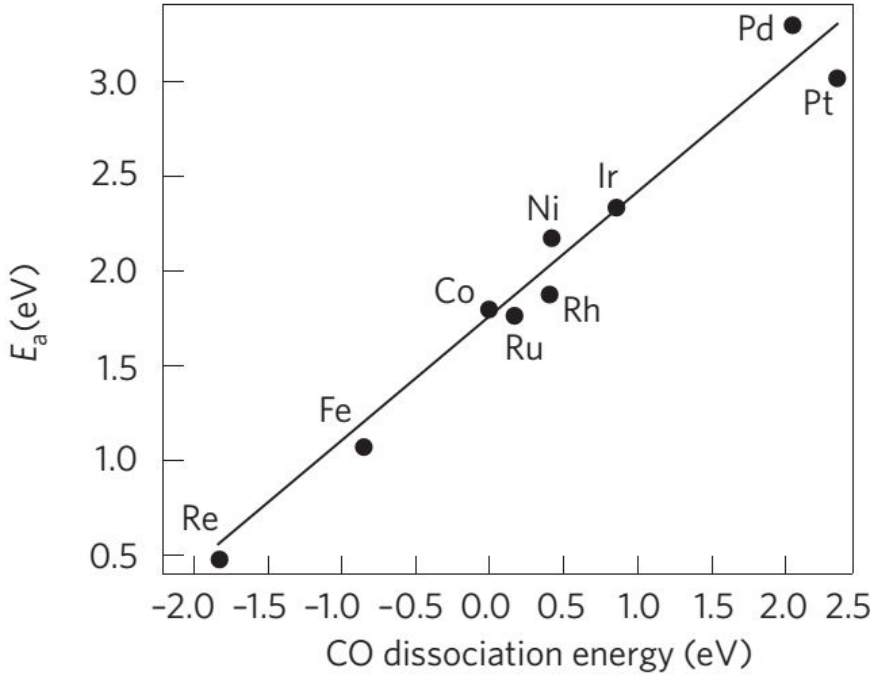
Simple(r) properties (bulk d-band center position and CO dissociation energy) are correlated to more complex properties (adsorption energy and reaction barrier)

The simpler quantities are called *descriptive parameters* (a *descriptor*)

J. K. Nørskov, T. Bligaard, J. Rossmeisl and C. H. Christensen, Nature Chemistry **1**, 37 (2009)

Descriptors





A simple physical model (Newns-Anderson) motivates the *d*-band center descriptor

What if we don't know such a model, or we need a more accurate and more widely applicable model?

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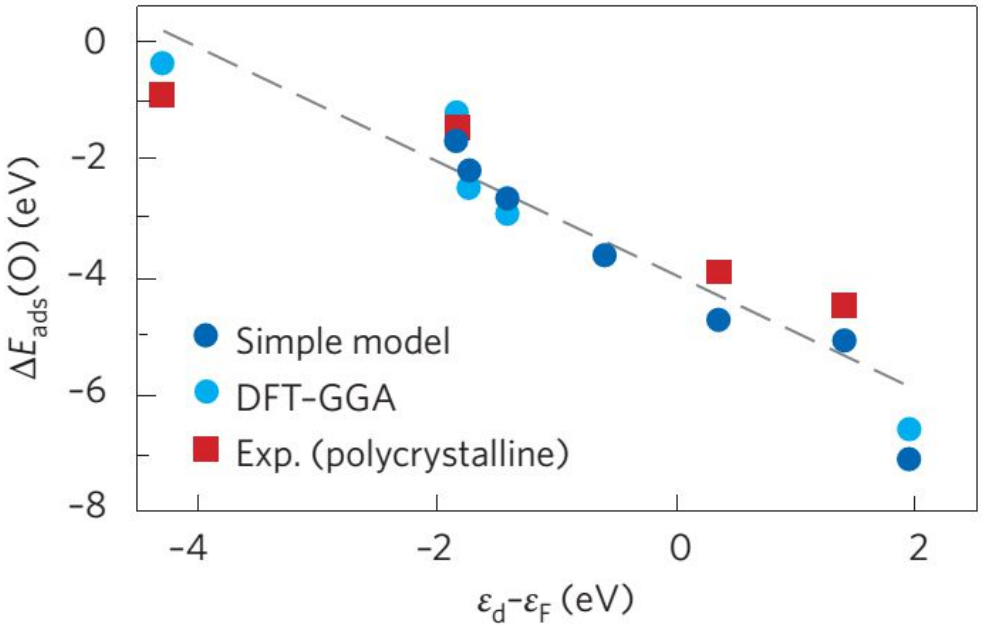

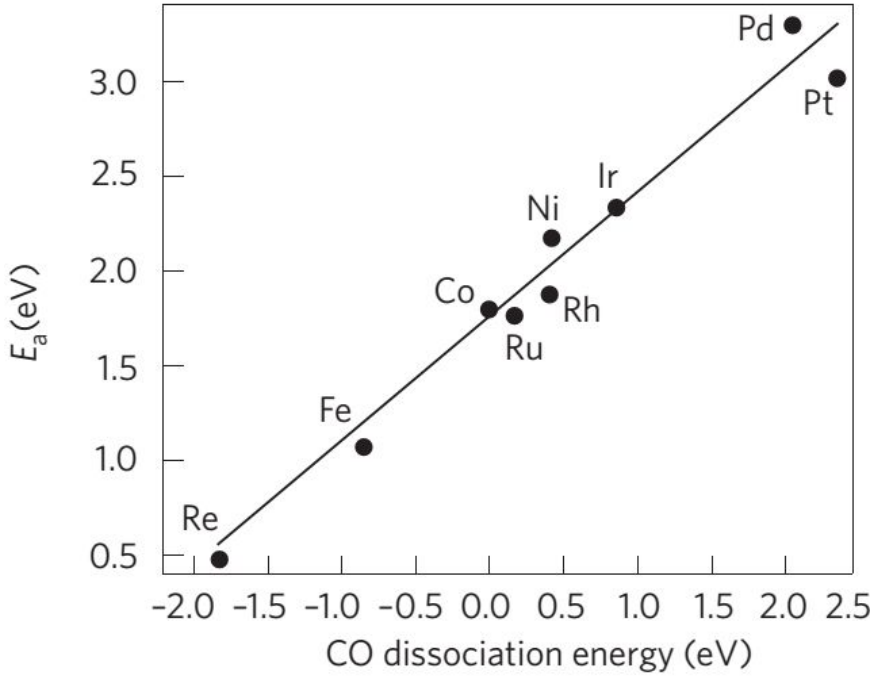

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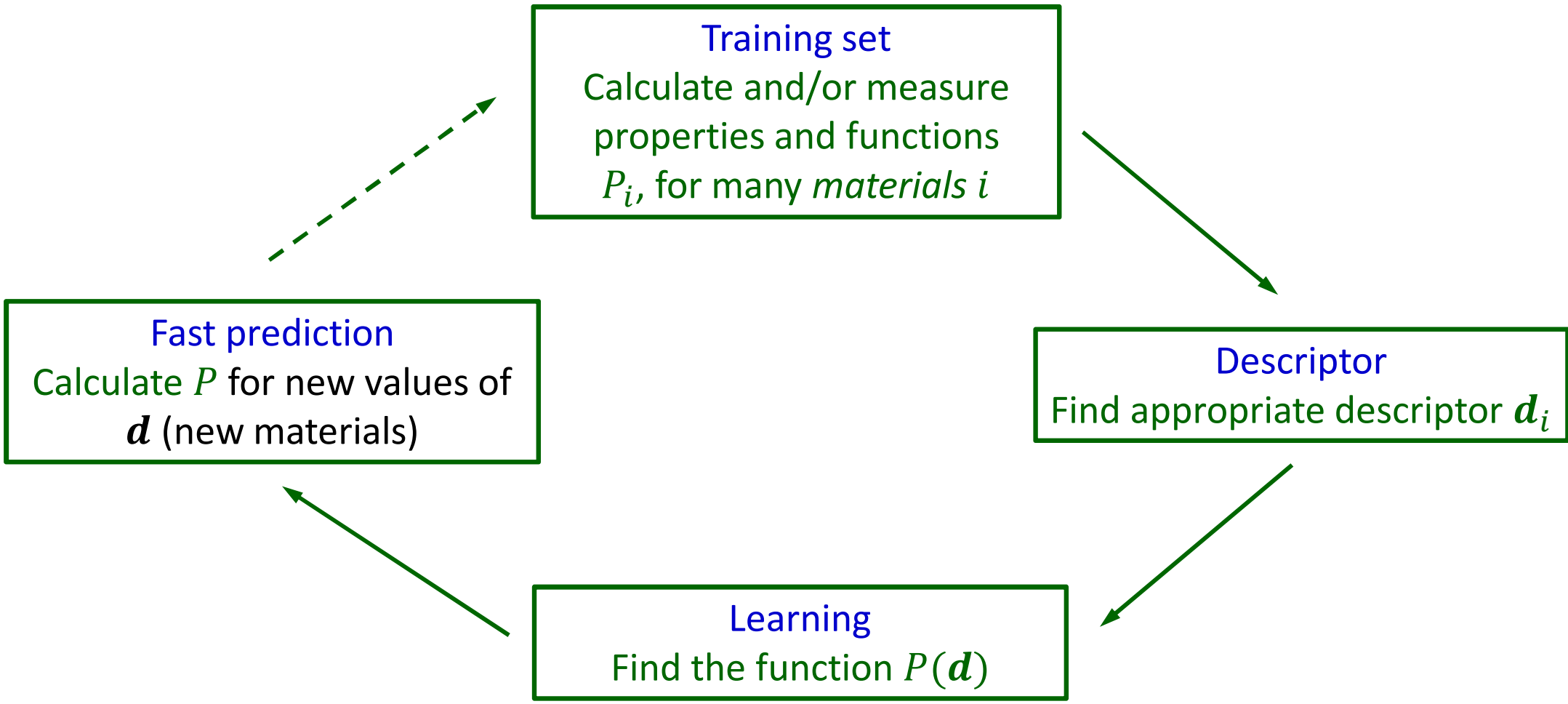
Find descriptor from DATA!

J. K. Nørskov, T. Bligaard, J. Rossmeisl and C. H. Christensen, Nature Chemistry **1**, 37 (2009)

Supervised data analysis



Descriptors

1) A descriptor d_i uniquely characterizes the material i as well as property-relevant elementary processes

2) The determination of the descriptor must not involve calculations as intensive as those needed for the evaluation of the property to be predicted

Target property model: Kernel ridge regression versus feature selection

Regression models: Basis set expansion in materials space

kernel ridge regression	linear
$P(\boldsymbol{d}) = \sum_{i=1}^{N} c_i \exp\left(-\|\boldsymbol{d}_i - \boldsymbol{d}\|_2^2 / 2\sigma^2\right)$	$P(\boldsymbol{d}) = \boldsymbol{d}\boldsymbol{c}$

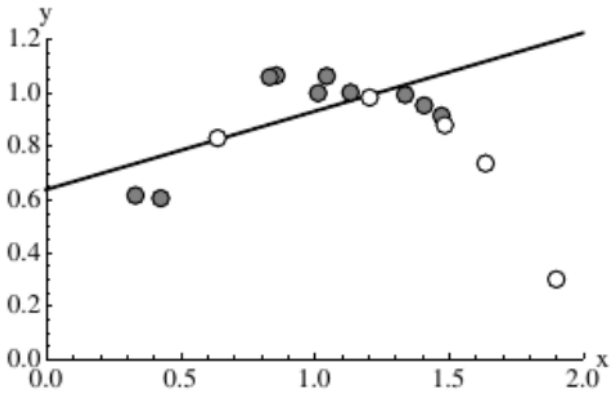
minimize

$$\|\boldsymbol{d}_i - \boldsymbol{d}_j\|_2^2 = \sum_{\alpha=1}^{\Omega} (d_{i,\alpha} - d_{j,\alpha})^2$$

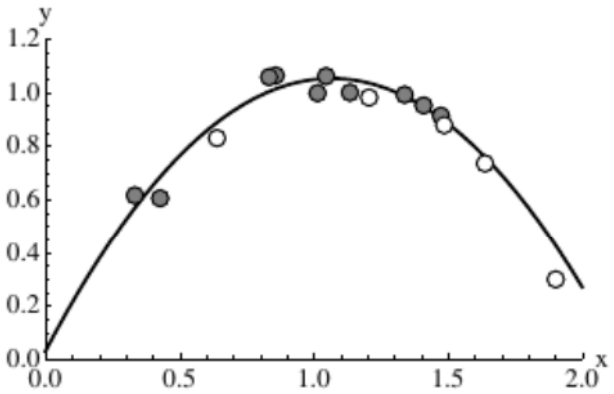
Regression: Importance of regularization

● training ○ validation

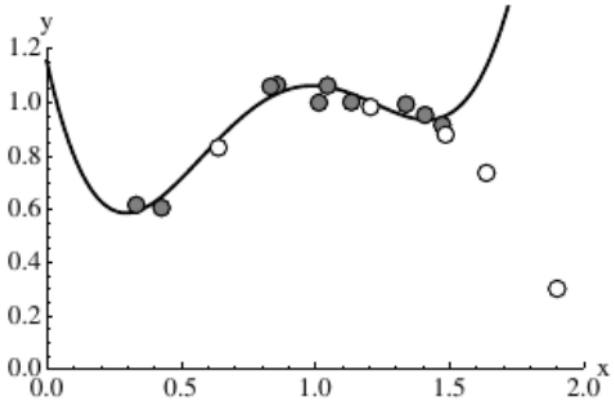
Underfitting



Fitting



Overfitting



Training/ validation error

0.123 / 0.443

0.044 / 0.068

0.036 / 0.939

$$\min_{c} \sum_i (P(d_i, c) - P_i)^2 + \lambda f(c), \min_{\lambda}(\text{validation error}) \rightarrow \lambda$$

Target property model: Kernel ridge regression versus feature selection

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kernel ridge regression	linear
$P(\boldsymbol{d}) = \sum_{i=1}^{N} c_i \exp\left(-\|\boldsymbol{d}_i - \boldsymbol{d}\|_2^2 / 2\sigma^2\right)$	$P(\boldsymbol{d}) = \boldsymbol{d}\boldsymbol{c}$
minimize	
$\sum_{i=1}^{N} (P(\boldsymbol{d}_i) - P_i)^2 \quad +$	$\sum_{i=1}^{N} (P(\boldsymbol{d}_i) - P_i)^2 \; +$
$\lambda \sum_{i,j=1}^{N,N} c_i c_j \exp\left(-\|\boldsymbol{d}_i - \boldsymbol{d}_j\|_2^2 / 2\sigma^2\right)$	$\lambda \|\boldsymbol{c}\|_0$
$\|\boldsymbol{d}_i - \boldsymbol{d}_j\|_2^2 = \sum_{\alpha=1}^{\Omega} (d_{i,\alpha} - d_{j,\alpha})^2$	

Target property model: Kernel ridge regression versus feature selection

kernel (Gaussian, Laplacian, linear $(d_i \cdot d_j)$)

kernel ridge regression

$$P(\boldsymbol{d}) = \sum_{i=1}^{N} c_i \exp\left(-\|\boldsymbol{d}_i - \boldsymbol{d}\|_2^2 / 2\sigma^2\right)$$

$$\sum_{i=1}^{N} (P(\boldsymbol{d}_i) - P_i)^2 \quad +$$

$$\lambda \sum_{i,j=1}^{N,N} c_i c_j \exp\left(-\|\boldsymbol{d}_i - \boldsymbol{d}_j\|_2^2 / 2\sigma^2\right)$$

penalty on similar data points

minimize

linear

$$P(\boldsymbol{d}) = \boldsymbol{d}\boldsymbol{c}$$

$$\sum_{i=1}^{N} (P(\boldsymbol{d}_i) - P_i)^2 \quad +$$

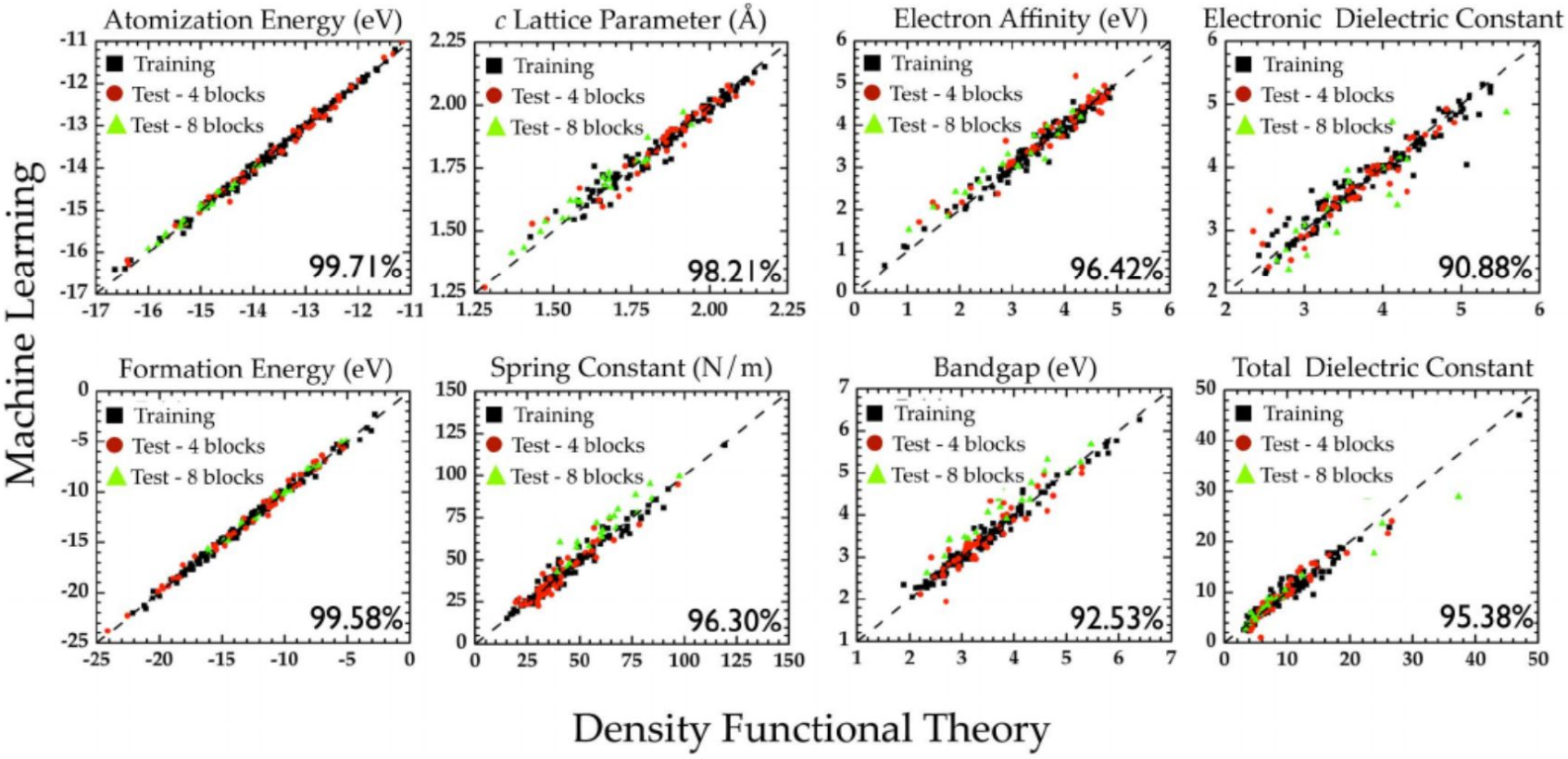
$$\lambda \|\boldsymbol{c}\|_0$$

penalty on the number of non-zero coefficients $\|\boldsymbol{c}\|_0$

(Gaussian) kernel ridge regression example

Data: 175 linear 4-blocks periodic polymers. 7 blocks: CH_2, SiF_2, $SiCl_2$, GeF_2, $GeCl_2$, SnF_2, $SnCl_2$,

Descriptor: 20 dimensions [# building blocks of type *i*, of *ii* pairs, of *iii* triplets]



Pilania, Wang, …, and Ramprasad, Scientific Reports 3, 2810 (2013). DOI: 10.1038/srep02810

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2) The determination of the descriptor must not involve calculations as intensive as those needed for the evaluation of the property to be predicted

3) The dimension Ω of the descriptor should be as low as possible (for a certain accuracy request)

Choose a physically motivated basis set!

L. M. Ghiringhelli, J. Vybiral, S. V. Levchenko, C. Draxl, and M. Scheffler, Phys. Rev. Lett. **114**, 105503 (2015)

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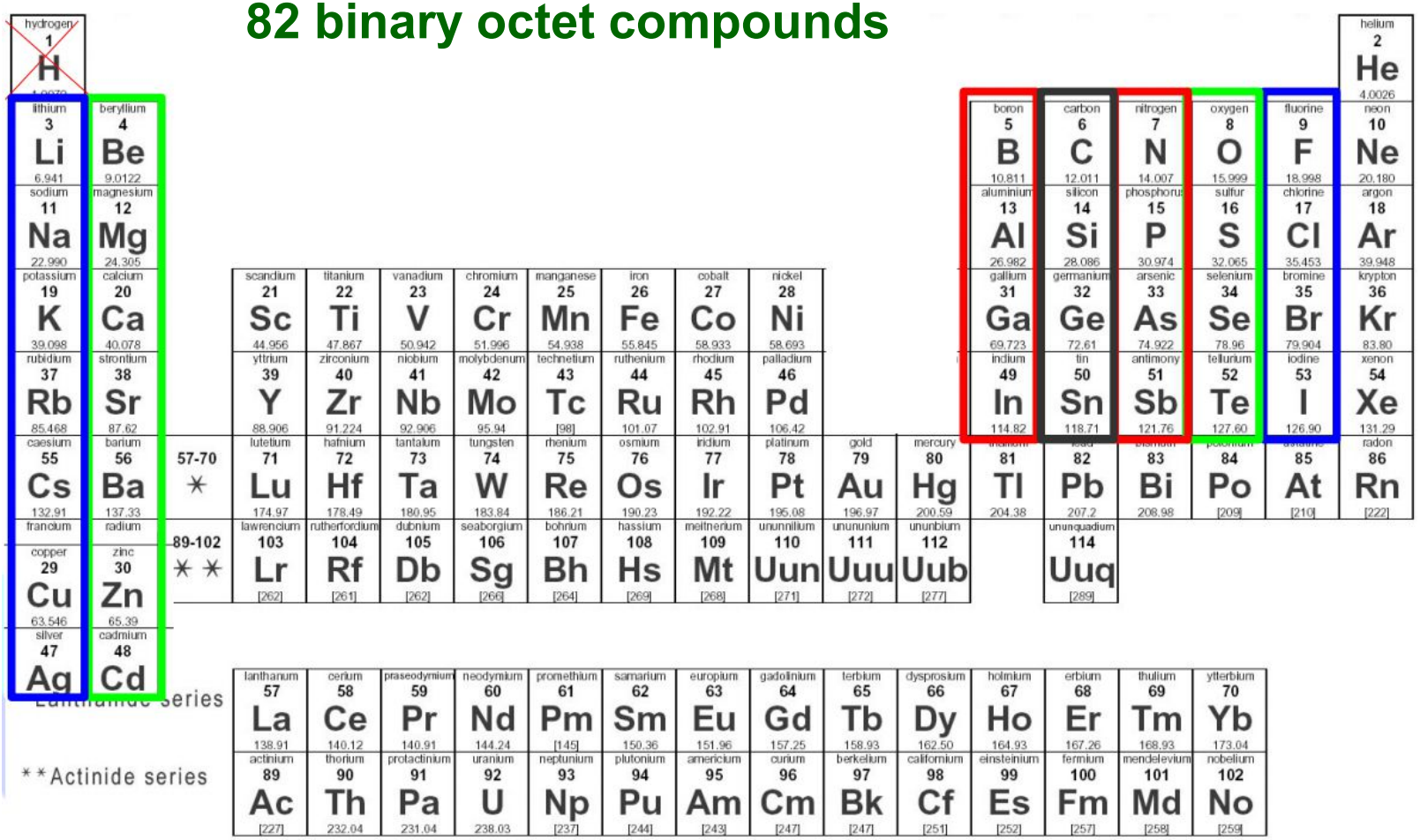
Idea: calculate many *physically motivated* quantities (features), and use these features as a basis for the physical model under compactness constraints

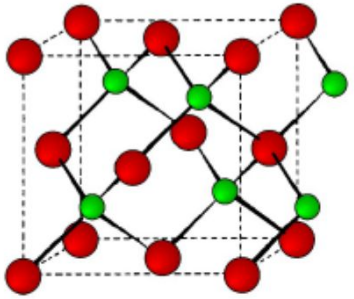
L. M. Ghiringhelli, J. Vybiral, S. V. Levchenko, C. Draxl, and M. Scheffler, Phys. Rev. Lett. **114**, 105503 (2015)

Proof of Concept: Descriptor for the Classification "Zincblende/Wurtzite (ZB/W) or Rocksalt (RS)?"

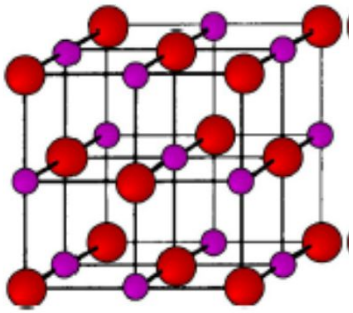
82 binary octet compounds



ZB



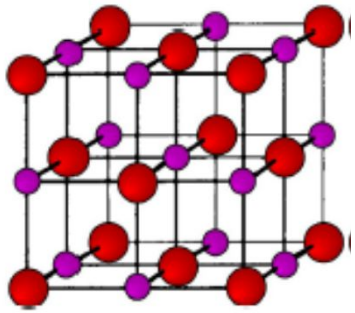
RS



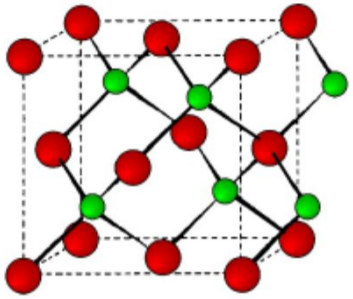
Proof of Concept: Descriptor for the Classification "Zincblende/Wurtzite (ZB/W) or Rocksalt (RS)?"

Crystal-structure prediction was and is one of the most important, basic challenges of materials science and engineering.

RS



ZB



Energy differences between different structures are very small.

For Si: 0.01% of the energy of a Si atom, or 0.1% of the 4 valence electrons.

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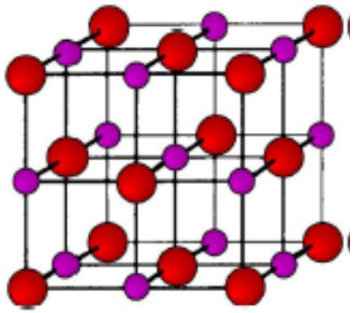
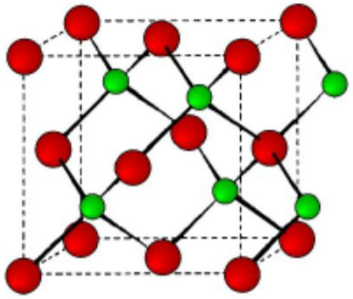
RS



ZB



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A. Zunger, Phys. Rev. B 22, 5839 (1980).
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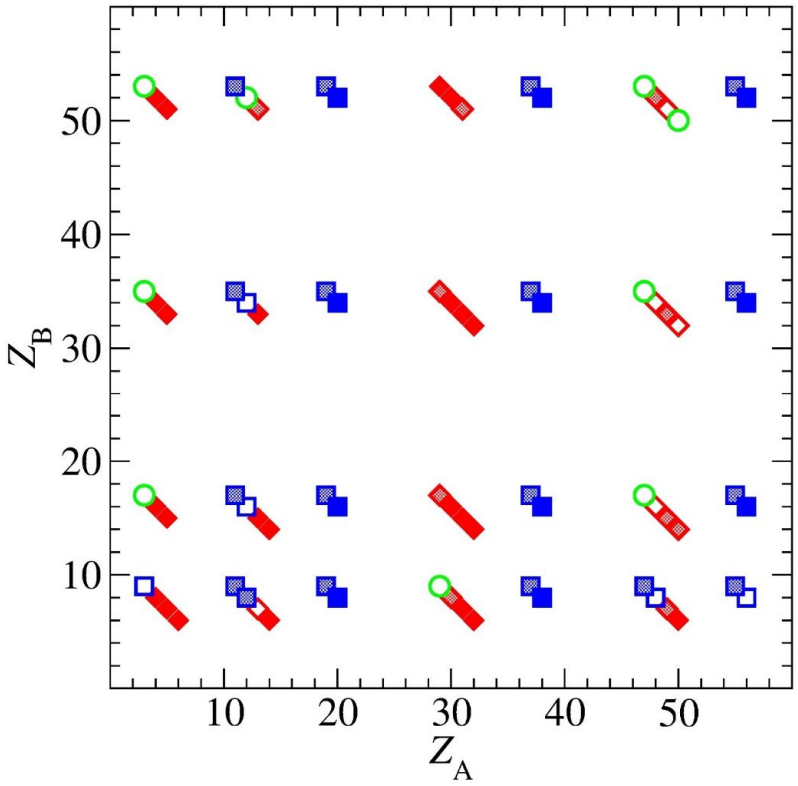
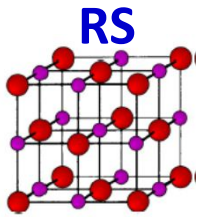
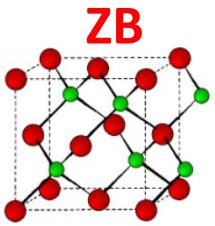
Can we predict not yet calculated structures from Z_A and Z_B? **Can we create a map: "The *ZB/W* community lives here and the *RS* community there?"**







$\Delta = E(\mathrm{RS}) - E(\mathrm{ZB})$

- ZB, $\Delta > 0.2$ eV
- ZB, $0.1\ \mathrm{eV} < \Delta \leq 0.2\ \mathrm{eV}$
- ZB, $0.05\ \mathrm{eV} < \Delta \leq 0.1\ \mathrm{eV}$
- $-0.05\ \mathrm{eV} < \Delta \leq 0.05\ \mathrm{eV}$
- RS, $-0.1\ \mathrm{eV} < \Delta \leq -0.05\ \mathrm{eV}$
- RS, $-0.2\ \mathrm{eV} < \Delta \leq -0.1\ \mathrm{eV}$
- RS, $\Delta \leq -0.2\ \mathrm{eV}$

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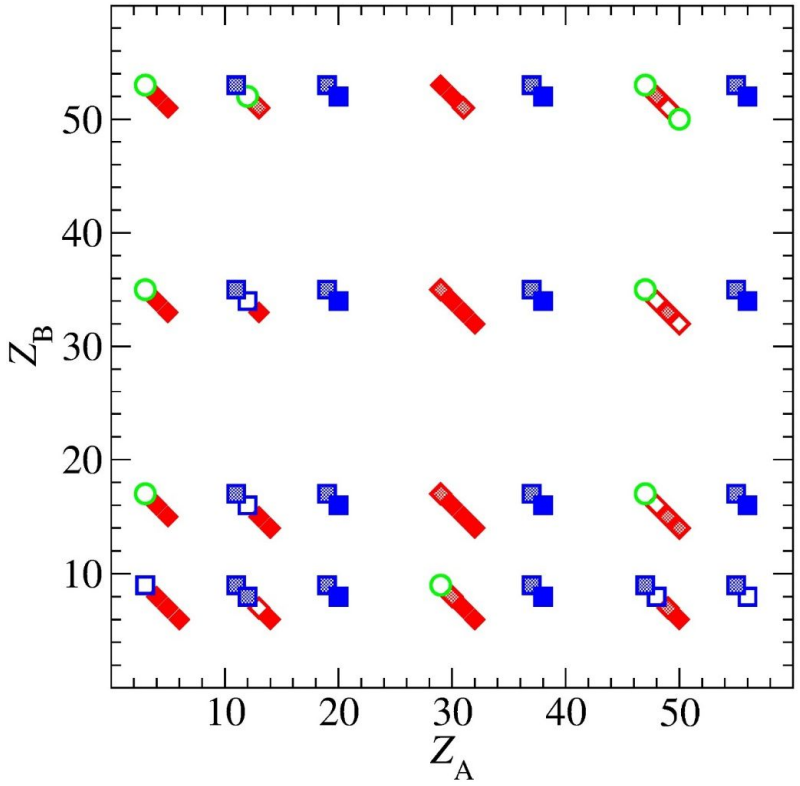
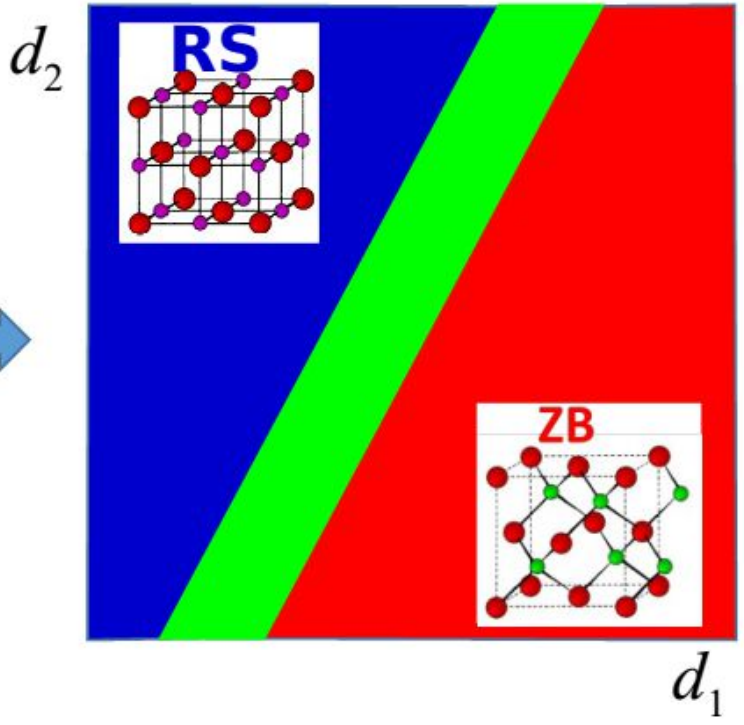
No complexity reduction → need a better basis





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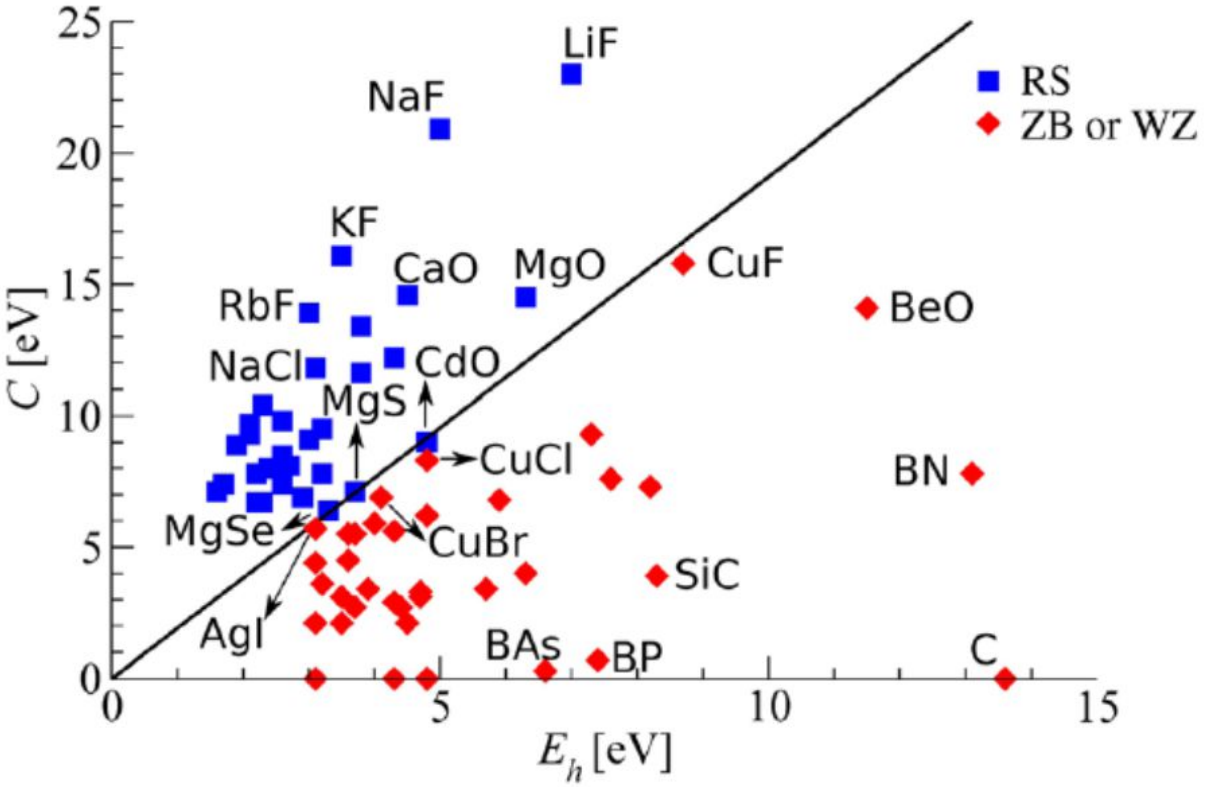

descriptor can be determined spectroscopically - properties of the solid

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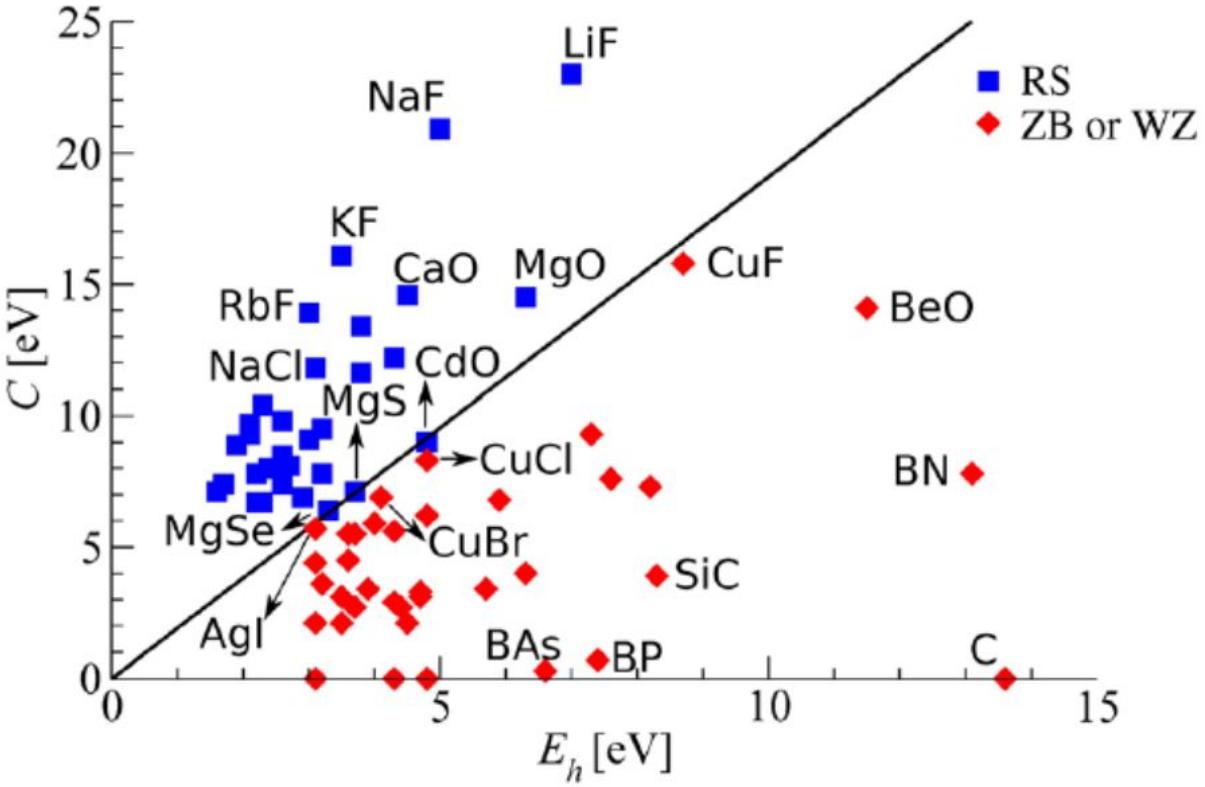


descriptor can be determined spectroscopically - properties of the solid

Can we create a map based on calculations simpler than bulk?

Primary features and feature space

free atoms

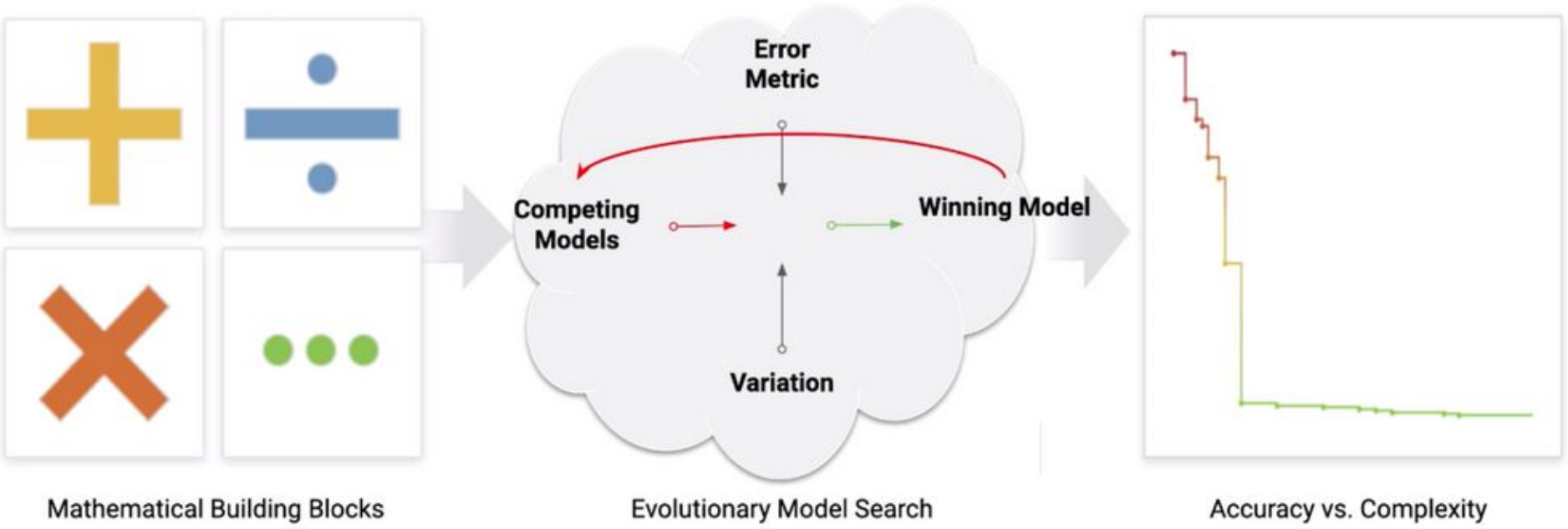
ID	Description	Symbols	#
$A1$	Ionization Potential (IP) and Electron Affinity (EA)	IP(A) EA(A) IP(B) EA(B) [1]	4
$A2$	Highest occupied (H) and lowest unoccupied (L) Kohn-Sham levels	H(A) L(A) H(B) L(B)	4
$A3$	Radius at the max. value of s, p, and d valence radial radial probability density	r_s(A) r_p(A) r_d(A) r_s(B) r_p(B) r_d(B)	6

free dimers

ID	Description	Symbols	#
$A4$	Binding energy	E_b(AA) E_b(BB) E_b(AB)	3
$A5$	HOMO-LUMO KS gap	HL(AA) HL(BB) HL(AB)	3
$A6$	Equilibrium distance	d(AA) d(BB) d(AB)	3

How to find the best model for our target property (energy difference between different crystal structures)?

Symbolic regression: Eureqa



Uses evolutionary algorithm to find the best formula describing target property

Assumes "gene" structure of the formula → bias

May result in an unnecessarily complex model

https://community.datarobot.com/t5/resources/introduction-to-eureqa/ta-p/2409

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free atoms

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free dimers

ID	Description	Symbols	#
$A4$	Binding energy	E_b(AA) E_b(BB) E_b(AB)	3
$A5$	HOMO-LUMO KS gap	HL(AA) HL(BB) HL(AB)	3
$A6$	Equilibrium distance	d(AA) d(BB) d(AB)	3

ID	description	prototype formula	#
$B1$	absolute differences and sums of $A1$	$\lvert\text{IP(A)} \pm \text{IP(B)}\rvert$	12
$B2$	absolute differences and sums of $A2$	$\lvert\text{L(B)} \pm \text{H(A)}\rvert$	12
$B3$	absolute differences and sums of $A3$	$\lvert r_p(\text{A}) \pm r_s(\text{A})\rvert$	30
$C3$	squares of $A3$ and $B3$ (only sums)	$r_s(\text{A})^2, (r_p(\text{A}) + r_s(\text{A}))^2$	21
$D3$	exponentials of $A3$ and $B3$ (only sums)	$\exp(r_s(\text{A})), \exp(r_p(\text{A}) \pm r_s(\text{A}))$	21
$E3$	exponentials of squared $A3$ and $B3$ (only sums)	$\exp(r_s(\text{A})^2), \exp(r_p(\text{A}) \pm r_s(\text{A})^2)$	21

We start with 23 primary features and build > 10,000 non-linear combinations

Mathematical formulation of the problem

P_j -- property value ($E_{ZB} - E_{RS}$) for material j (a function in materials space)

$d_{j,l}$ -- value of feature l related to material j (e.g., $\left|r_s(A_j) - r_p(B_j)\right|$) (a basis function in materials space)

c_l -- coefficient of the expansion of the property function in terms of basis functions:

$$P_j = \sum_l d_{j,l} c_l$$

How to find c_l?

$$\sum_j \left(P_j - \sum_l d_{j,l} c_l \right)^2 + \lambda \|\boldsymbol{c}\|_n \rightarrow \operatorname{argmin}(\boldsymbol{c})$$

regularization term to explore and ensure compactness of the expansion (reduce complexity)

Mathematical formulation of the problem

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$\|c\|_0$ -- number of non-zero coefficients → NP hard! (need to try all combinations)

Mathematical formulation of the problem

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$\|c\|_0$ -- number of non-zero coefficients → NP hard! (need to try all combinations)

$\|c\|_2 = \sum_l |c_l|^2$ -- ridge regression → not most compact!

$\|c\|_1 = \sum_l |c_l|$ -- LASSO (Least Absolute Shrinkage and Selection Operator) → convex problem, equivalent to the NP-hard if features (columns of d) are uncorrelated

Compressed (compressive?) sensing

Raw: 15MB

JPEG: 150KB

Expand in a basis (wavelets) → use LASSO to select most important basis functions → store compressed image

Mathematical formulation of the problem

P_j -- property value ($E_{ZB} - E_{RS}$) for material j (a function in materials space)

$d_{j,l}$ -- value of feature l related to material j (e.g., $|r_s(A_j) - r_p(B_j)|$) (a basis function in materials space)

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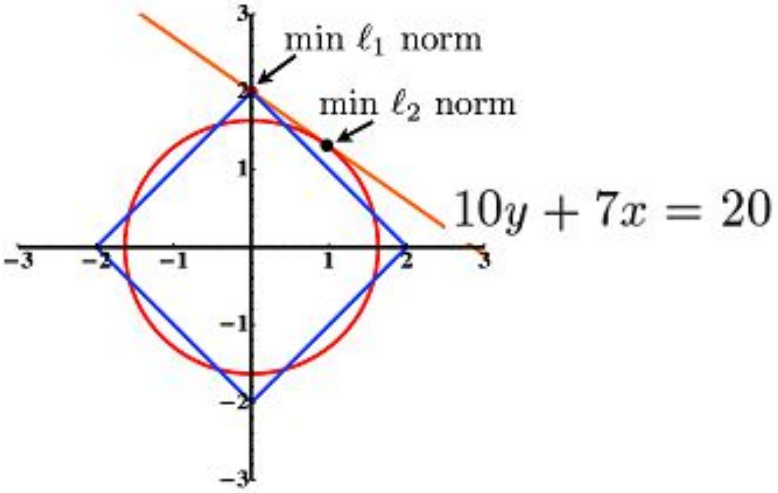
How to find c_l?

$$\sum_j \left(P_j - \sum_l d_{j,l} c_l \right)^2 + \lambda \|\boldsymbol{c}\|_n \rightarrow \text{argmin}(\boldsymbol{c})$$



$\|c\|_1 = \sum_l |c_l|$ -- LASSO (Least Absolute Shrinkage and Selection Operator) → convex problem, equivalent to the NP-hard if features (columns of D) are uncorrelated (no linear dependence in the basis set)

The descriptors selected with LASSO

$$\frac{\mathrm{IP(B)}-\mathrm{EA(B)}}{r_p(\mathrm{A})^2}\ \text{1D},\quad \frac{|r_s(\mathrm{A})-r_p(\mathrm{B})|}{\exp(r_s(\mathrm{A}))}\ \text{2D},\quad \frac{|r_p(\mathrm{B})-r_s(\mathrm{B})|}{\exp(r_d(\mathrm{A}))}\ \text{3D}$$

$$\Delta E = 0.117\frac{\mathrm{EA(B)}-\mathrm{IP(B)}}{r_p(\mathrm{A})^2} - 0.342 \qquad \text{1D}$$

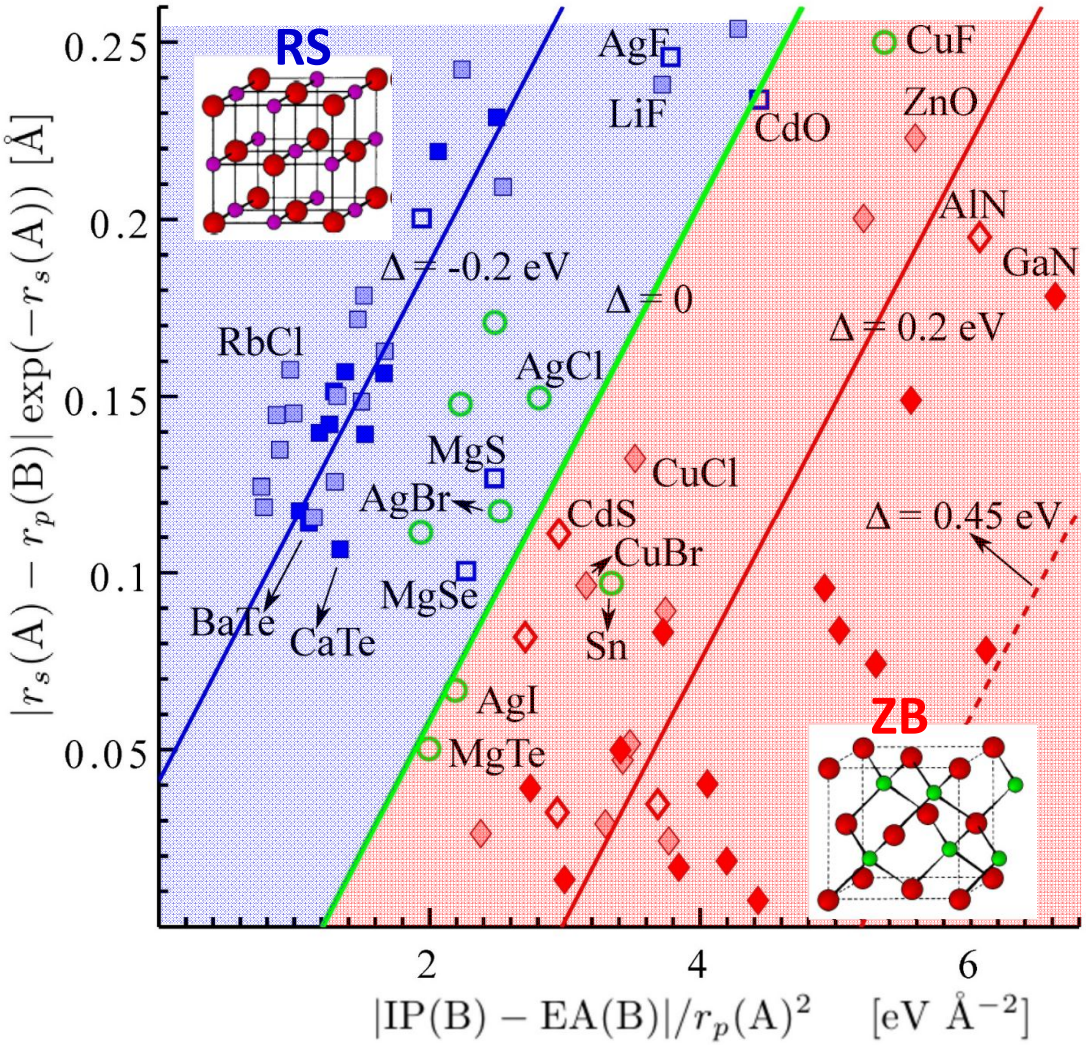
$$\Delta E = 0.113\frac{\mathrm{EA(B)}-\mathrm{IP(B)}}{r_p(\mathrm{A})^2} + 1.542\frac{|r_s(\mathrm{A})-r_p(\mathrm{B})|}{\exp(r_s(\mathrm{A}))} - 0.137 \qquad \text{2D}$$

$$\Delta E = 0.108\frac{\mathrm{EA(B)}-\mathrm{IP(B)}}{r_p(\mathrm{A})^2} + 1.790\frac{|r_s(\mathrm{A})-r_p(\mathrm{B})|}{\exp(r_s(\mathrm{A}))} + \qquad \text{3D}$$

$$+ 3.766\frac{|r_p(\mathrm{B})-r_s(\mathrm{B})|}{\exp(r_d(\mathrm{A}))} - 0.0267$$

Same features are selected for higher-dimensional descriptors, but this does not have to be the case

“The Map” -- compressed sensing -- LASSO, 2D descriptor



$\Delta = E(\mathrm{RS}) - E(\mathrm{ZB})$
- ZB, $\Delta > 0.2$ eV
- ZB, $0.1\ \mathrm{eV} < \Delta \leq 0.2$ eV
- ZB, $0.05\ \mathrm{eV} < \Delta \leq 0.1$ eV
- $-0.05\ \mathrm{eV} < \Delta \leq 0.05$ eV
- RS, $-0.1\ \mathrm{eV} < \Delta \leq -0.05$ eV
- RS, $-0.2\ \mathrm{eV} < \Delta \leq -0.1$ eV
- RS, $\Delta \leq -0.2$ eV

$$P(j) = \boldsymbol{d}(j)\boldsymbol{c}$$

The complexity and science is in the descriptor (identified from >10,000 features).

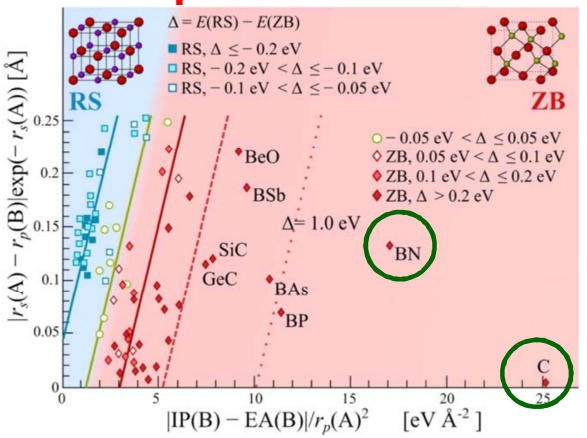
L.M. Ghiringhelli, J. Vybiral, S.V. Levchenko, C. Draxl, and M. Scheffler,
Phys. Rev. Lett. **114**, 105503 (2015).

Predictive power of the model

Hadn't we known about diamond … we'd have predicted it!

When both carbon diamond and BN are excluded from training:

	ΔE(LDA)	ΔE(predicted)
C	-2.64 eV	-1.44 eV
BN	-1.71 eV	-1.37 eV



Hadn't we known about any carbon-containing binary … we'd have predicted carbon chemistry (from atomic features)

If all C containing binaries (C, SiC, GeC, and SnC) are excluded from training, i.e. no explicit information on C is given to the model:

	ΔE(LDA)	ΔE(predicted)
C	-2.64 eV	-1.37 eV
SiC	-0.67 eV	-0.48 eV
GeC	-0.81 eV	-0.46 eV
SnC	-0.45 eV	-0.23 eV

Predictive power of the model

Mean absolute error (MAE), and maximum absolute error (MaxAE), in eV, (first two lines) and for a leave-10%-out cross validation (CV), averaged over 150 random selections of the training set (last two lines). For (Z_A*, Z_B*), each atom is identified by a string of three random numbers.

Descriptor	Z_A, Z_B	Z_A*, Z_B*	1D	2D	3D	5D
MAE	$1*10^{-4}$	$3*10^{-3}$	0.12	0.08	0.07	0.05
MaxAE	$8*10^{-4}$	0.03	0.32	0.32	0.24	0.20
MAE, CV	0.13	0.14	0.12	0.09	0.07	0.05
MaxAE, CV	0.43	0.42	0.27	0.18	0.16	0.12

Gaussian-kernel ridge regression

LASSO

Predictive power of the model

Mean absolute error (MAE), and maximum absolute error (MaxAE), in eV, (first two lines) and for a leave-10%-out cross validation (CV), averaged over 150 random selections of the training set (last two lines). For (Z_A*, Z_B*), each atom is identified by a string of three random numbers.

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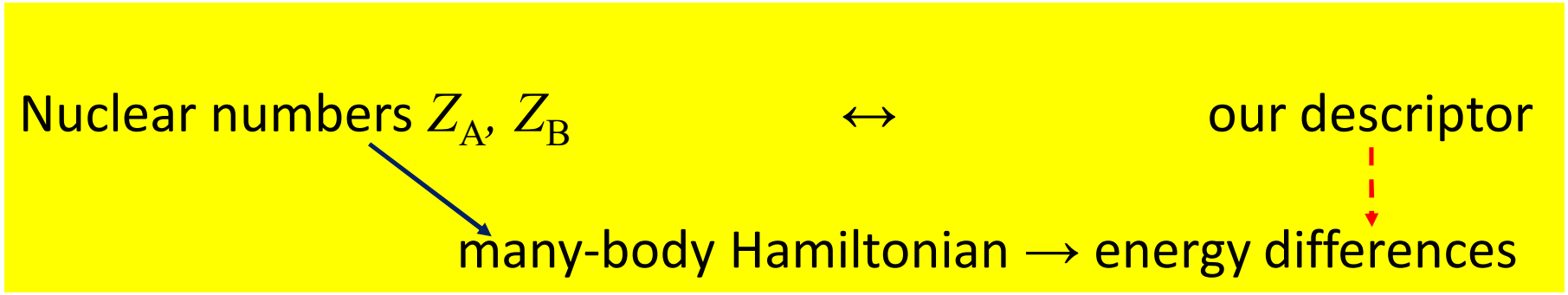
Gaussian-kernel ridge regression

LASSO

Drawing causal inference from data



a mapping exists, even a physical intuition exists, but ΔE does not listen directly to the descriptor (intricate causality)

$$P(j) = \boldsymbol{d}(j)\boldsymbol{c}$$

There are two aspects:

1) **practical aspect -- we benefit from knowing $d \rightarrow P$ mapping for any convenient $d(j)$ (analogy: plane waves)**
2) **physical aspect (understanding) -- we can reduce the complexity of the model and at the same time increase its applicability domain by a clever choice of $d(j)$ (analogy: atomic orbitals and molecular-orbital picture)**

We greatly benefit from $d(j)$ providing a framework for a rational analysis

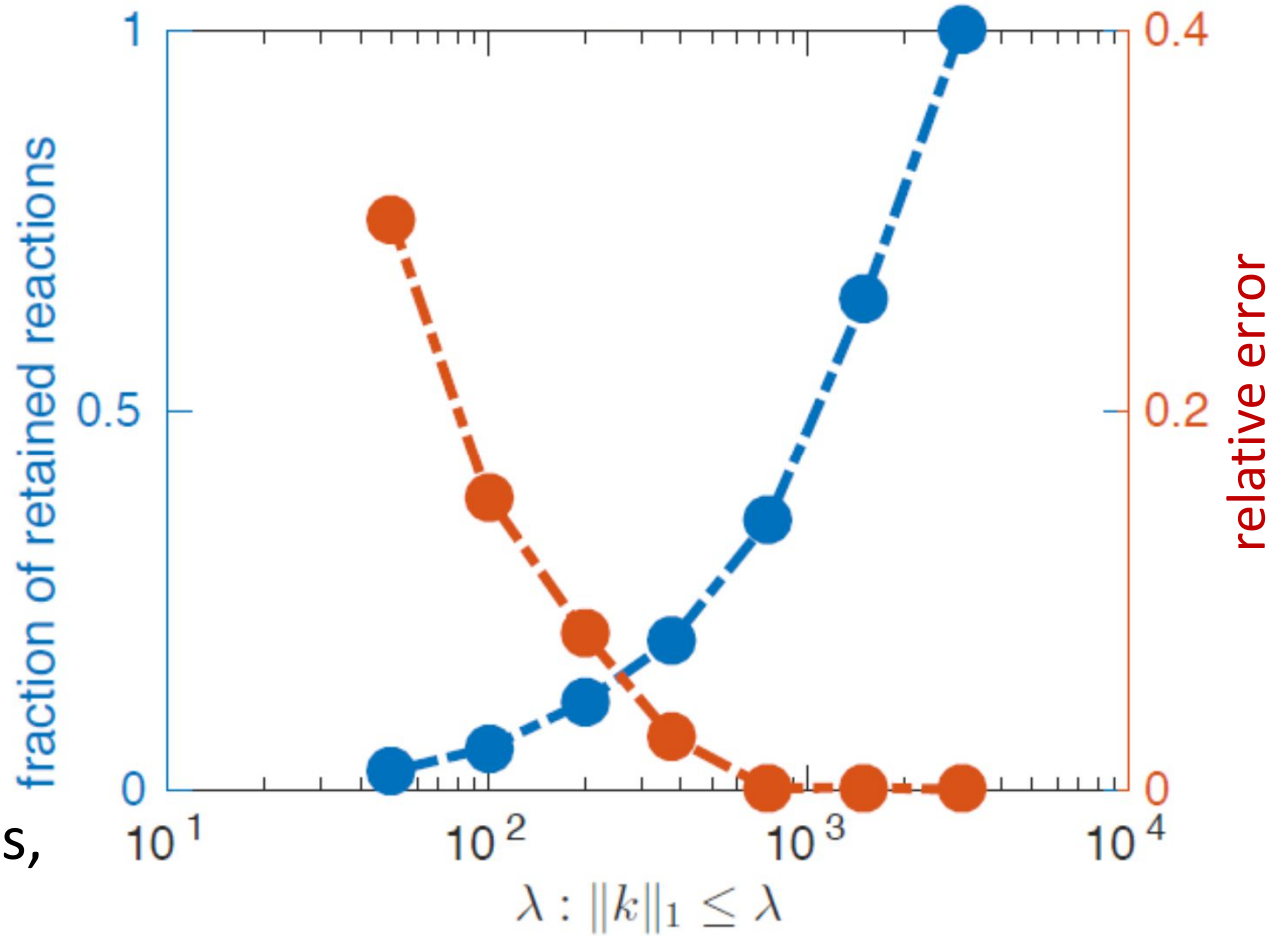
CH_4 chemical decomposition under shock-compression conditions (high *T* and *p*)

Yang, Q., Sing-Long, C. A., Reed, E. J., MRS Advances 1 (2016)

Methane at T = 3,300 K, p = 40.53 GPa: MD simulations (using a force-field description) find 2,613 different chemical reactions. Using compressed sensing it is shown that only 11% of them are relevant.

$$\min_{\hat{k}} \left\| A\hat{\boldsymbol{k}} - b \right\|_2$$
$$\text{subject to } \hat{\boldsymbol{k}} \geq 0, \left\| \hat{\boldsymbol{k}} \right\|_1 \leq \lambda$$

The A matrix has 2,613 columns, 2,395,918,510 rows



Lattice Anharmonicity and Thermal Conductivity from Compressive Sensing of First-Principles Calculations

$$\boldsymbol{F}_a = -\Phi_a - \Phi_{ab}\boldsymbol{u}_b - \frac{1}{2}\Phi_{abc}\boldsymbol{u}_b\boldsymbol{u}_c - \cdots$$

force on atom a (training data)

force constant tensor $\partial^2 E/\partial u_a \partial u_b$ (unknown)

displacement of atom c (training data)

$$\min_{\Phi}\left(\lambda\sum_I|\Phi_I| + \sum_a\left(F_a - A_{aJ}\Phi_J\right)^2\right) \rightarrow \Phi$$

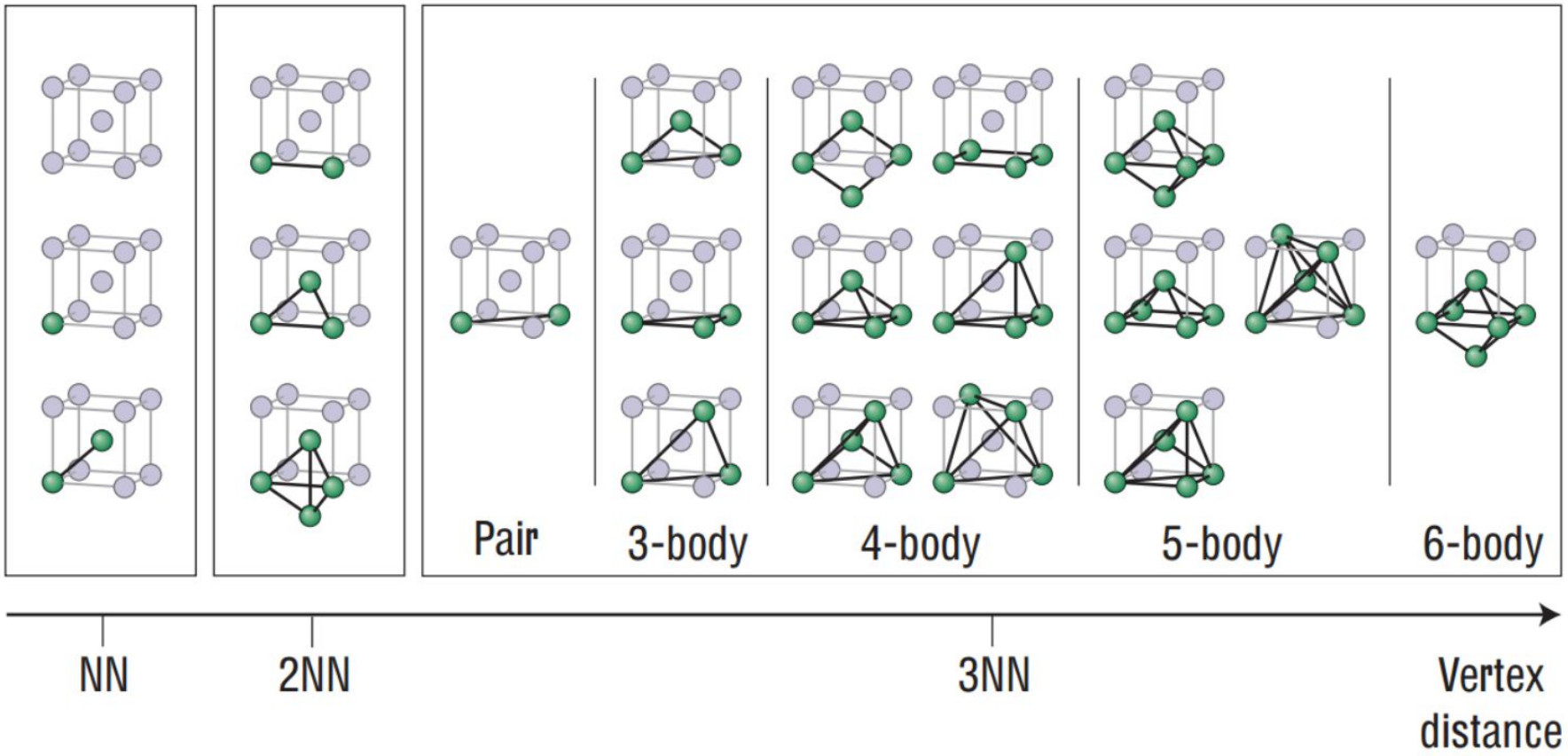
$$A_{aJ} = \begin{bmatrix} -1 & u_b^1 & -\frac{1}{2}u_b^1u_c^1 & \cdots \\ \vdots & \vdots & \vdots & \\ -1 & u_b^L & -\frac{1}{2}u_b^Lu_c^L & \cdots \end{bmatrix}$$

→ predictive model for anharmonic lattice dynamics

F. Zhou, W. Nielson, Y. Xia, and Vidvuds Ozoliņš, Phys. Rev. Lett. 113, 185501 (2014)

Compressive Sensing for Cluster Expansion

$$E(\sigma) = E_0 + \sum_f \Pi_f(\sigma) J_f \qquad \min_{J_f} \left(\lambda \sum_f |J_f| + \sum_i (E^{DFT}(\sigma_i) - E^{CE}(\sigma_i))^2 \right) \rightarrow J_f$$



L. J. Nelson, G. L. W. Hart, F. Zhou, and V. Ozoliņš, Phys. Rev. B 87, 035125 (2013)

Enabling Feature Spaces with Billions of Elements by Sure Independence Screening

$\|c\|_1 = \sum_l |c_l|$ **-- LASSO → convex problem, equivalent to the NP-hard if features are uncorrelated → not the case when many features are generated → Sure Independence Screening plus Selection Operator (SISSO)**

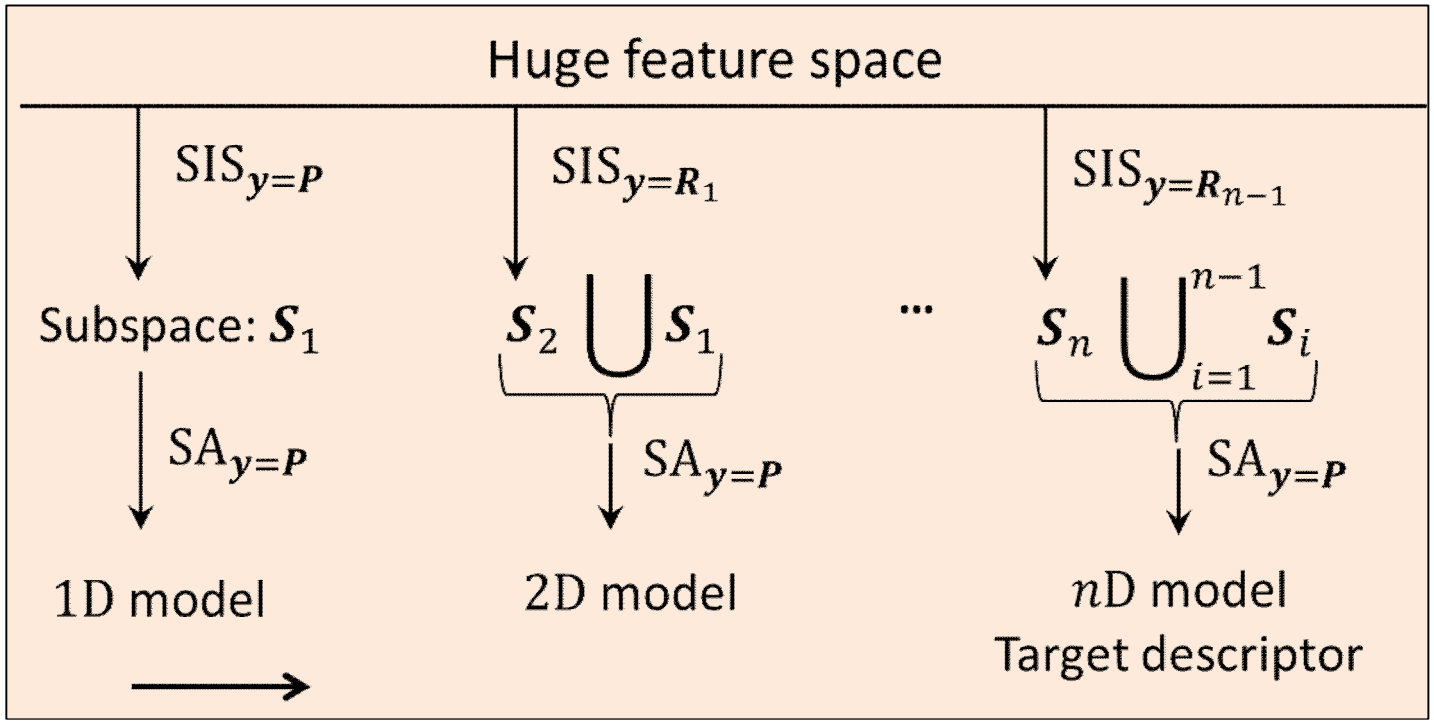
1. Systematically construct a huge feature space (10^{11}) from primary features: $\hat{R} = \{+, -, \cdot, ^{-1}, ^2, ^3, \sqrt{\ }, exp, log, |-|\}$(use physically meaningful combinations!)
2. Select top ranked features using *Sure Independence Screening (SIS)*[1] (correlation learning). Select n features corresponding to the n largest projection on the target property, i.e. largest components of the vector ($\boldsymbol{D}^T \boldsymbol{y}$)

   $\boldsymbol{y}$: vector with the target property (e.g., rock salt-zincblende energy differences; 82 elements)

   $\boldsymbol{D}$: matrix of the feature space (e.g., 82 x 100 billion elements)
3. Apply a sparsifying operator (l_0 regularization) to the selected features to determine 1D, 2D,... descriptors

R. Ouyang, *et al.*, Physical Review Materials 2, 083802 (2018)

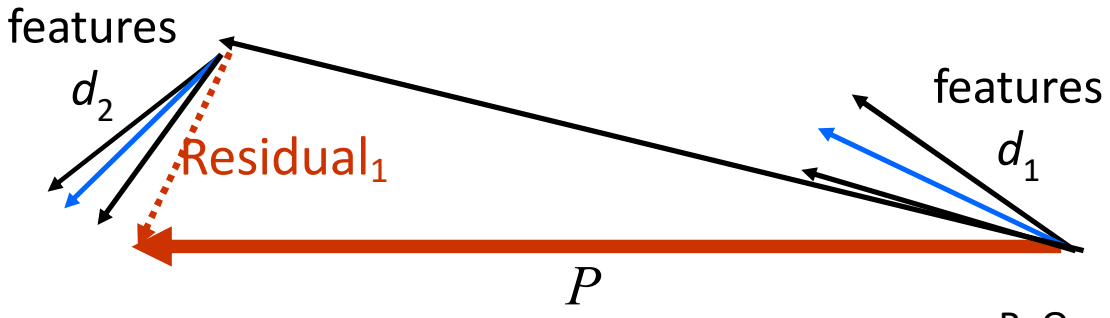
SISSO: Iterative residual fitting



$\boldsymbol{y}$: response vector

$\boldsymbol{P}$: target material property

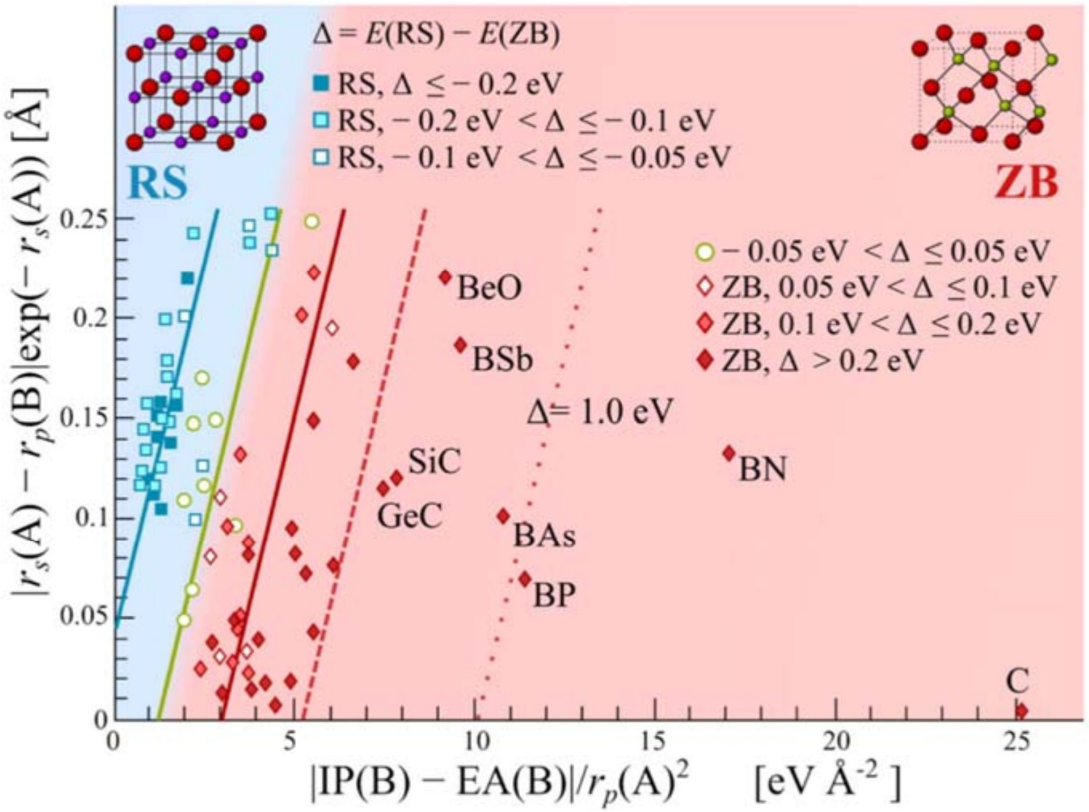
Residual: $R = P - \sum_i c_i d_i$



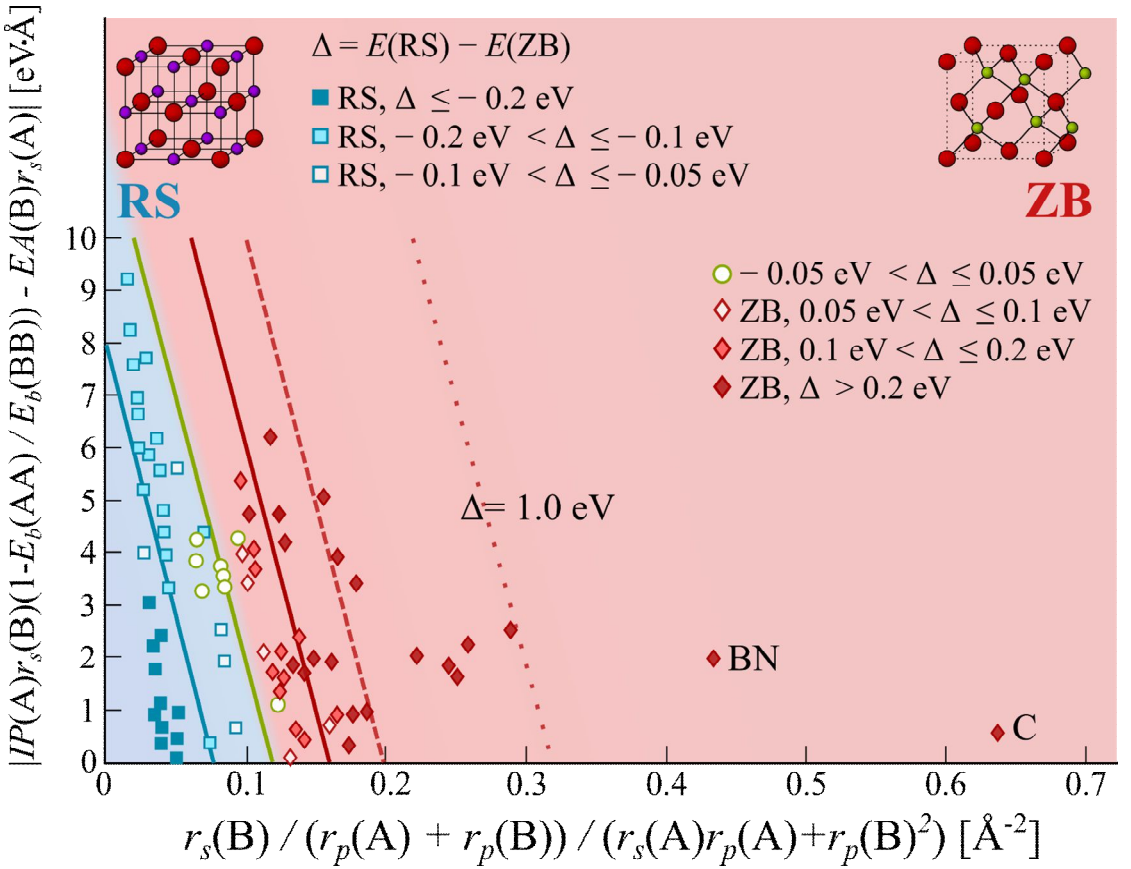
R. Ouyang, *et al.*, Physical Review Materials 2, 083802 (2018)

SISSO: Performance

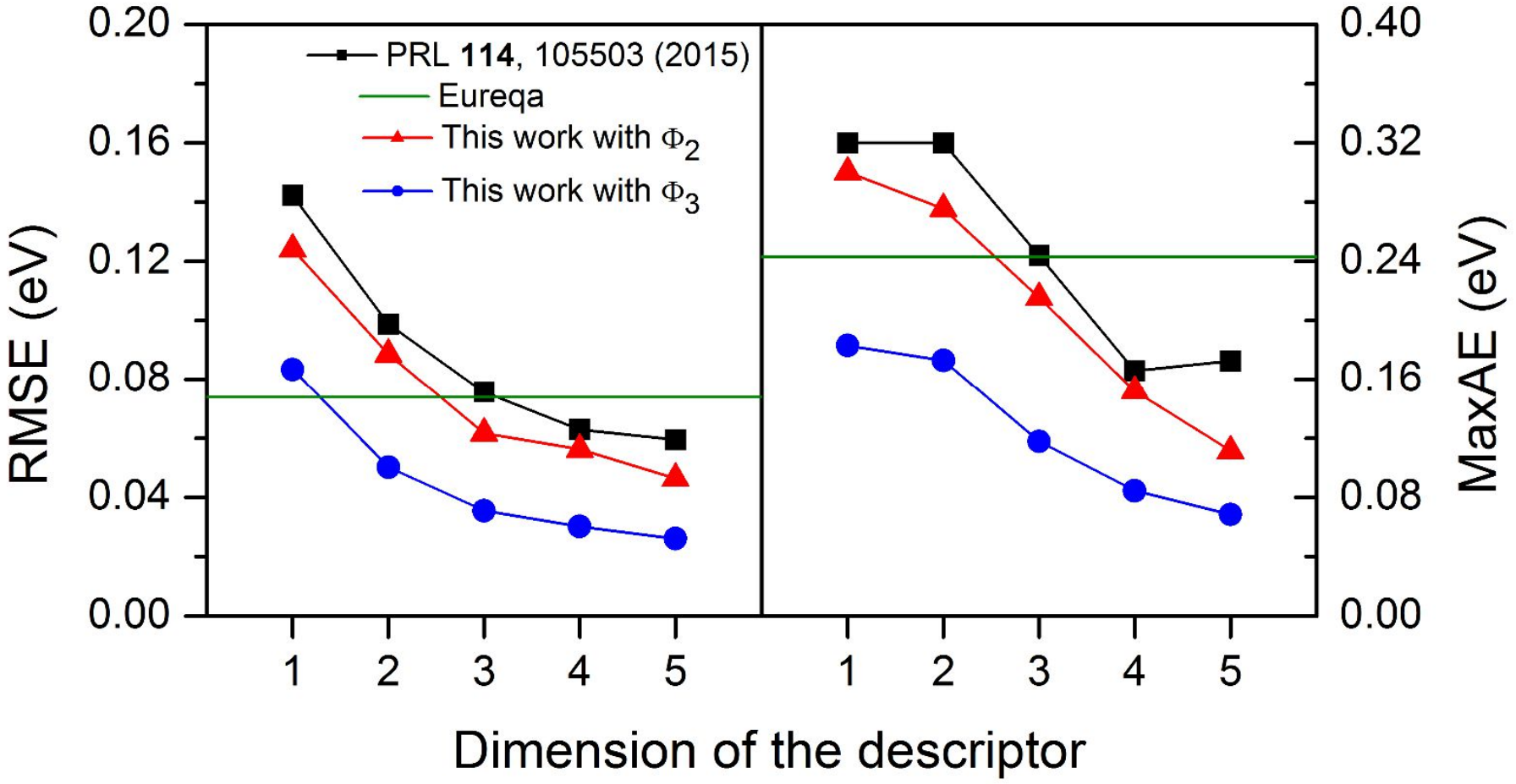
LASSO(+l_0)



SISSO



SISSO: Performance



SISSO: Multitask and categorical

Multitask: Construct simultaneously SISSO models for several properties with the same descriptor

$$\min_{\boldsymbol{c}} \left(\lambda \left\| c_i^k \right\|_0 + \sum_k \frac{1}{N_{\text{samples}}^k} \sum_{\substack{\text{samples} \\ \text{in } k}} \left(P^k - \boldsymbol{d}\boldsymbol{c}^k \right)^2 \right) \rightarrow \boldsymbol{c}$$

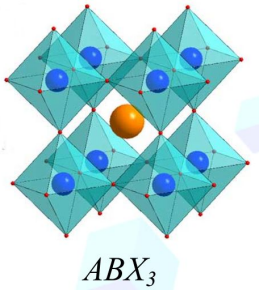
Categorical (can be also multitask): Property - material belongs to a given class (yes/no)

$$\min_{\boldsymbol{c}} \left(\lambda \left\| c_i^k \right\|_0 + \sum_{I=1}^{N_{\text{classes}}} \sum_{J \neq I} O_{IJ}(\boldsymbol{d}, \boldsymbol{c}) \right) \rightarrow \boldsymbol{c}$$

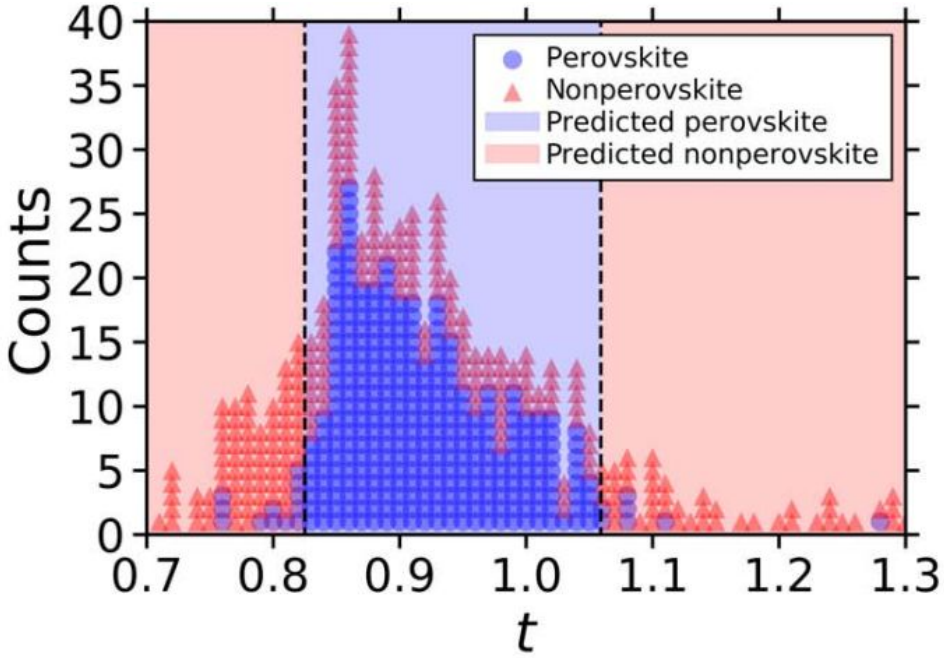
number of data in the overlap region between domains of different classes in d-space

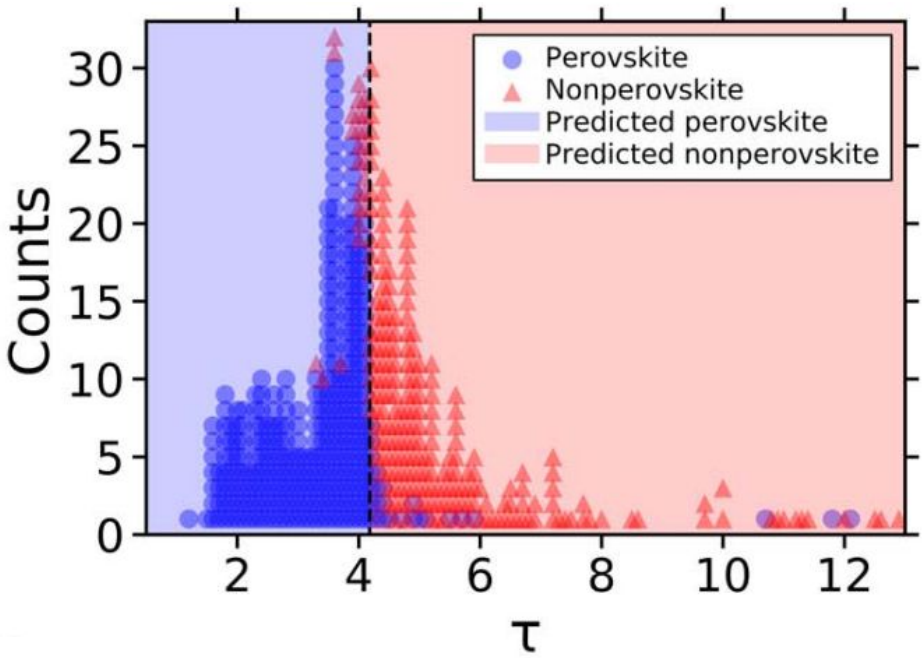
R. Ouyang, *et al*., J. Phys.: Mater. 2, 024002 (2019)

SISSO: Examples



- **Perovskite phase stability (improved tolerance factor)**





Goldschmidt factor: accuracy 79%

$$0.825 < \frac{r_A + r_X}{\sqrt{2}(r_B + r_X)} < 1.059$$

ionic radii

New factor: accuracy 92%

$$\frac{r_X}{r_B} - n_A\left(n_A - \frac{r_A/r_B}{\ln(r_A/r_B)}\right) < 4.18$$

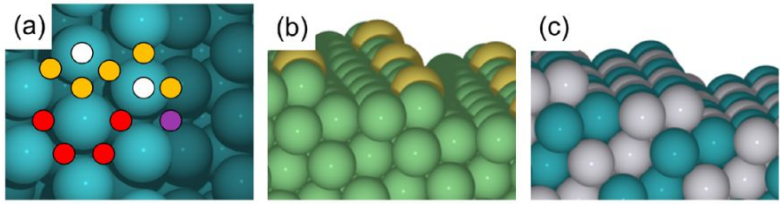
oxidation state

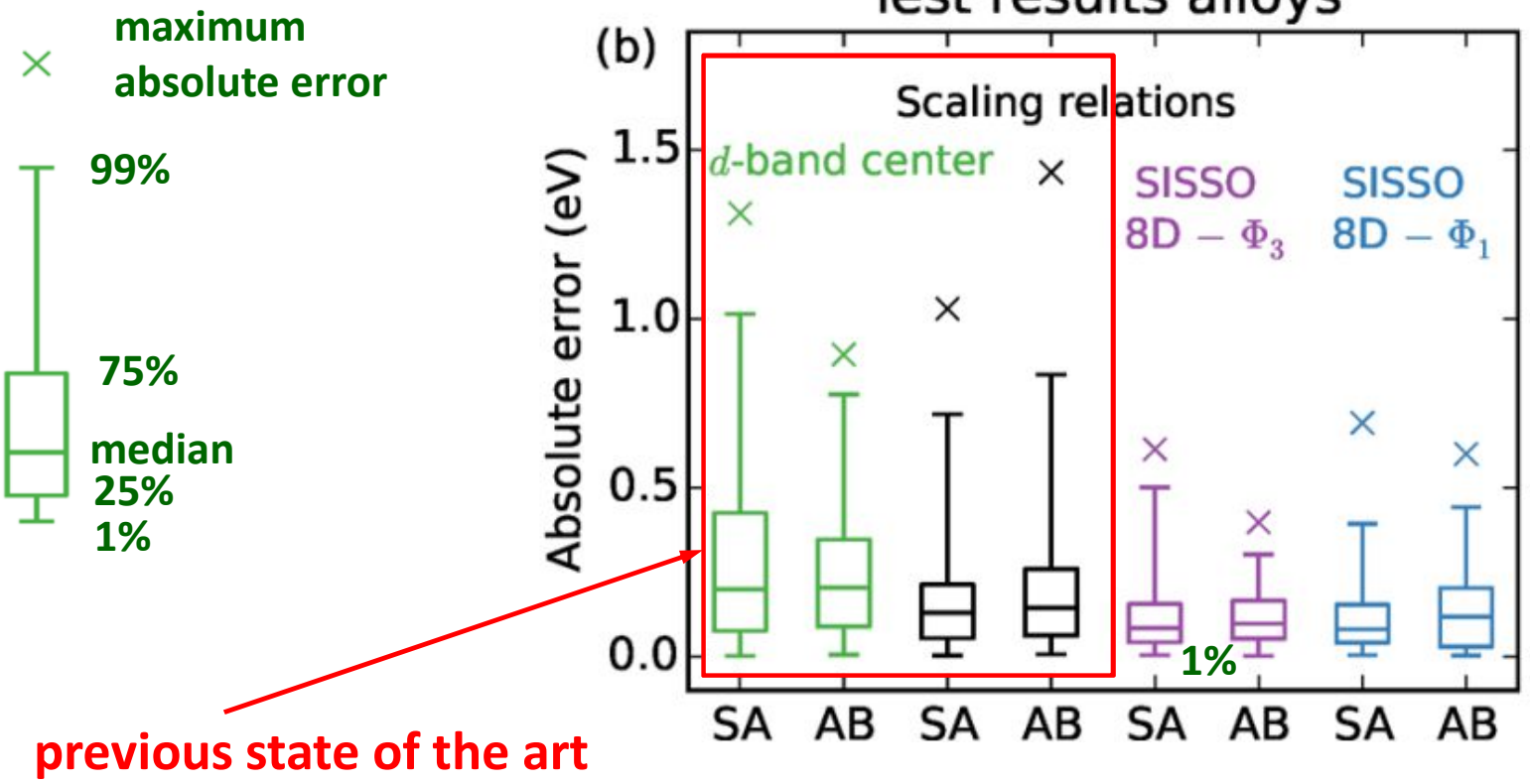
C. Bartel *et al.*, Sci. Adv. *5, eaav0693* (2019)

SISSO: Examples

- Adsorption of molecules on metal surfaces

Adsorption of C, CH, CO, H, O, OH)

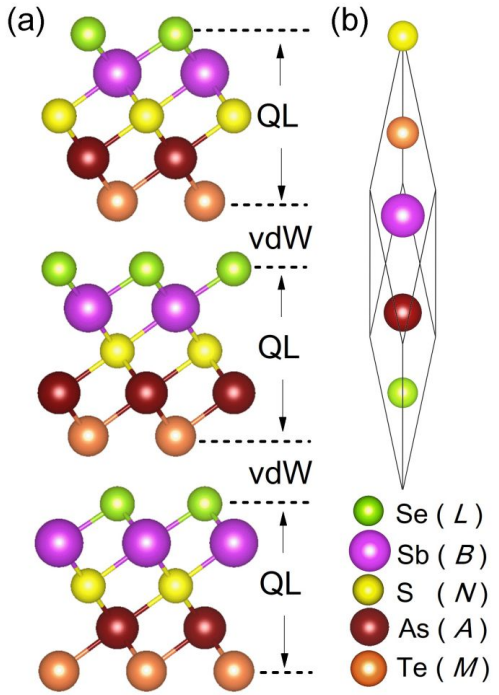


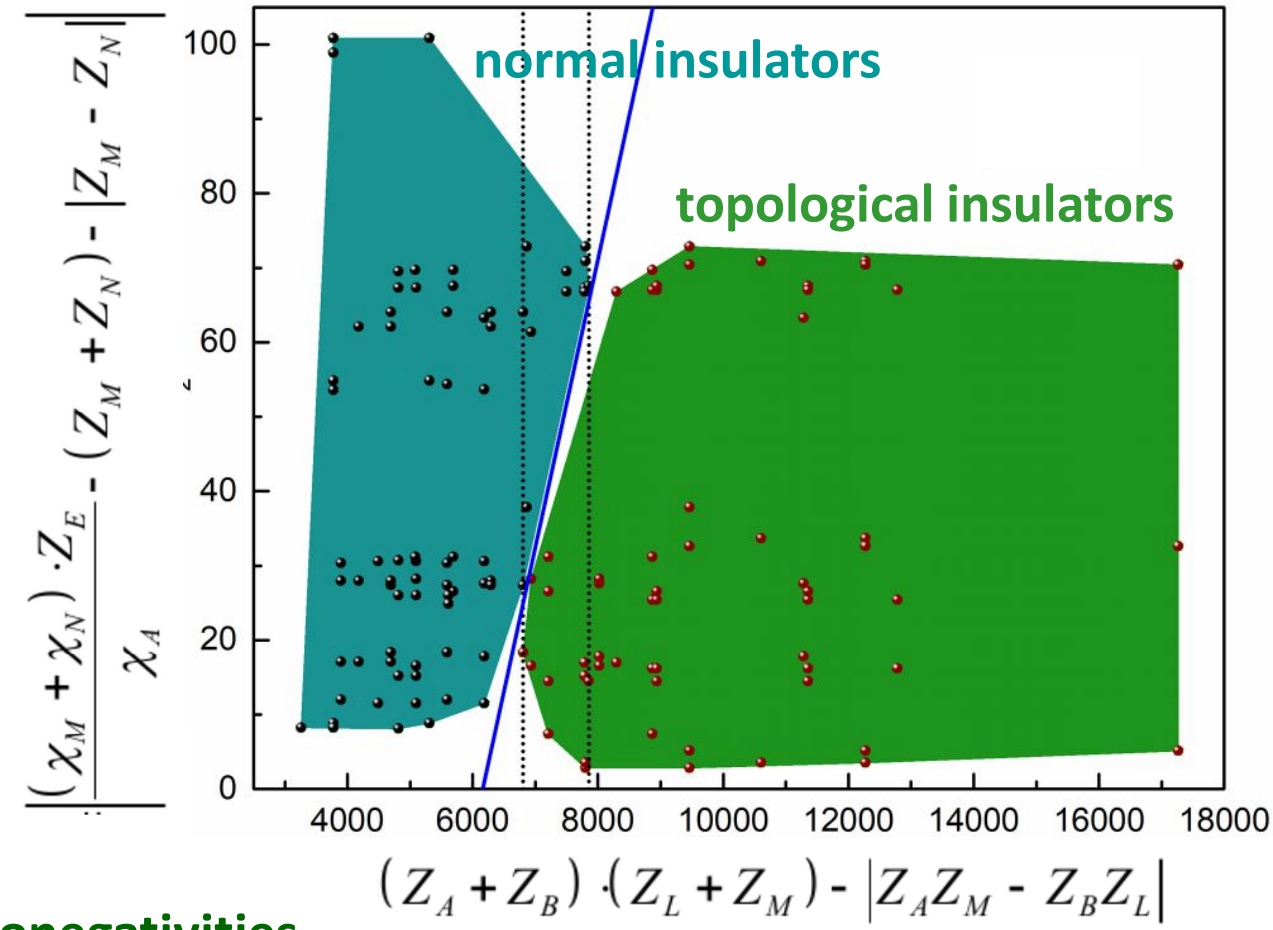


M. Andersen *et al*., ACS Catal. 9, 2752 (2019)

SISSO: Examples

- **Design of topological insulators (materials for spintronics, catalysis, thermoelectricity)**

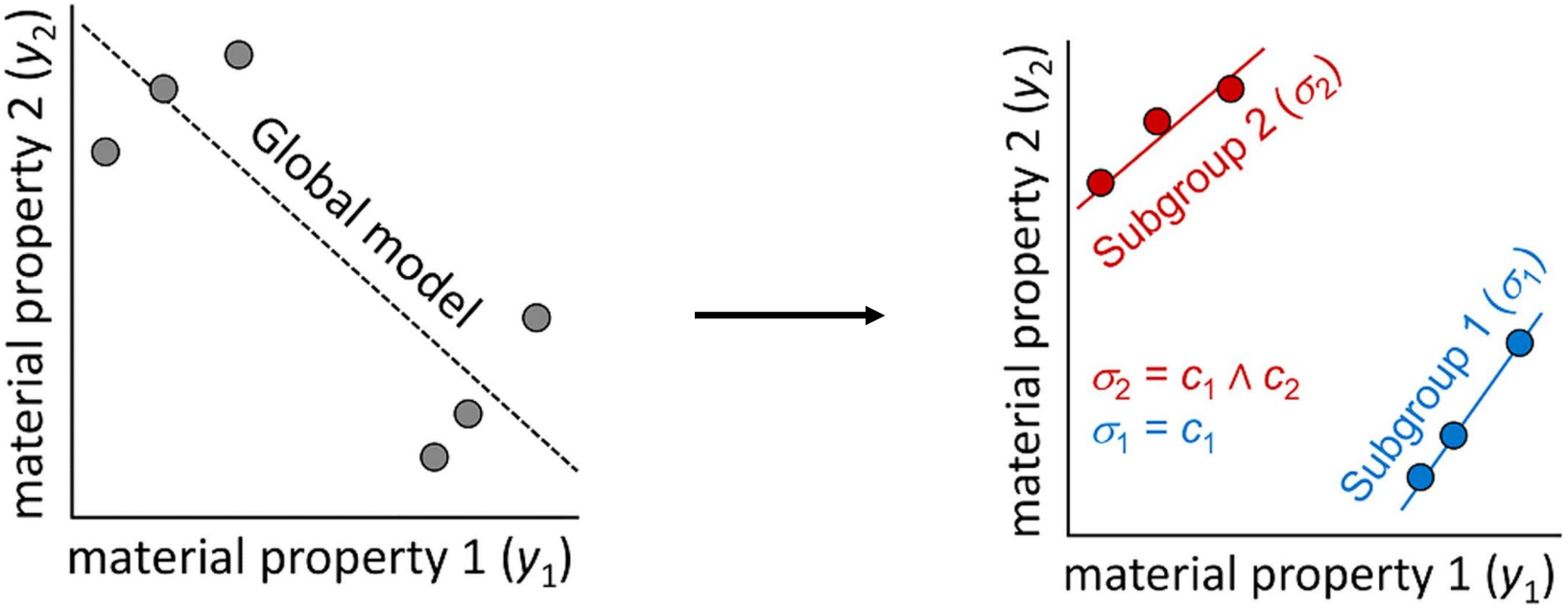




Z - atomic numbers, χ - electronegativities

G. Cao *et al.*, arXiv:1808.04733

Data mining: Subgroup discovery



Subgroups are defined by selectors σ expressed as "AND" combinations of statements like "band gap < 2 eV", "atom radius > 1.4 Å", etc.

SGD algorithm: find subgroups that maximize *quality function*

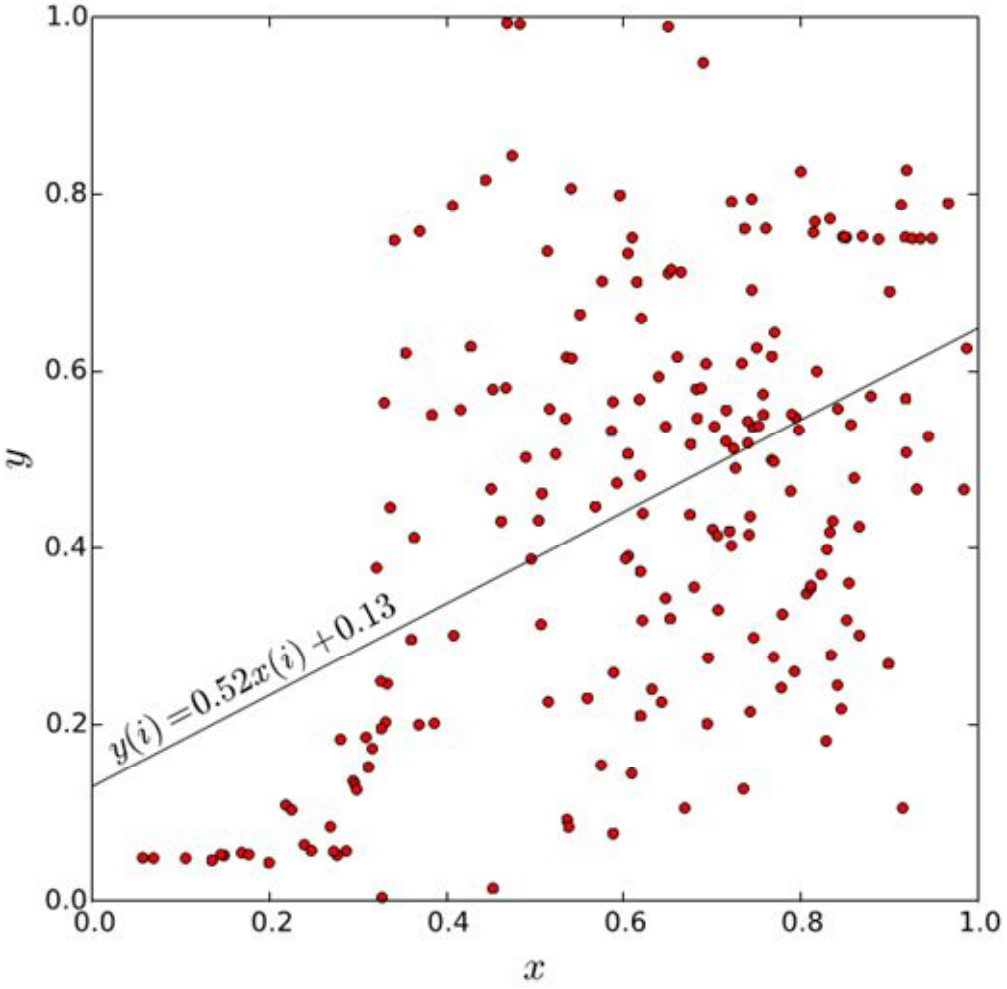
$$f = N_{subgroup}/N_{all} \times |mean_{subgroup} - mean_{all}| \times (1 - variance_{subgroup}/variance_{all})$$

Numerical separators ("band gap < 2 eV") from k-means clustering (unsupervised learning)

Search for subgroups: Monte Carlo or branch-and-bound algorithm

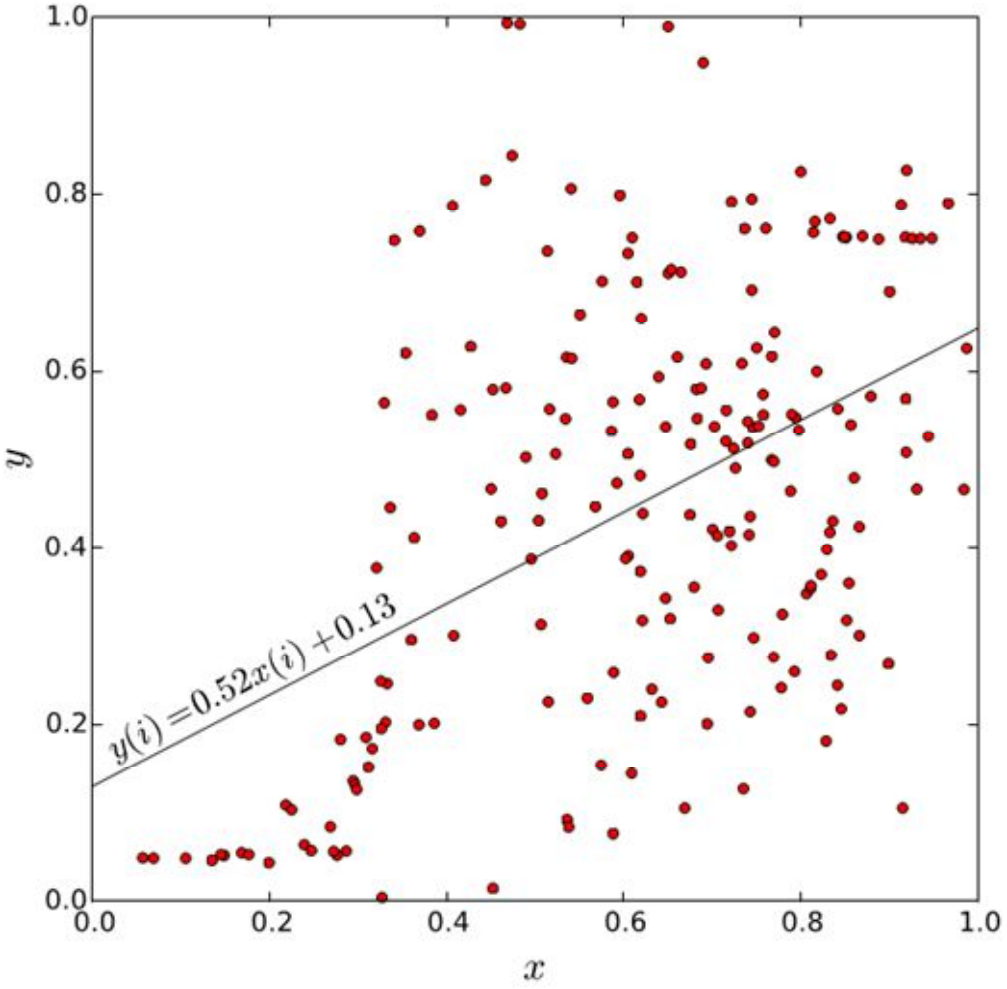
W. Klösgen, Advances in Knowledge Discovery and Data Mining. Palo Alto, CA: AAAI Press; 1996, 249

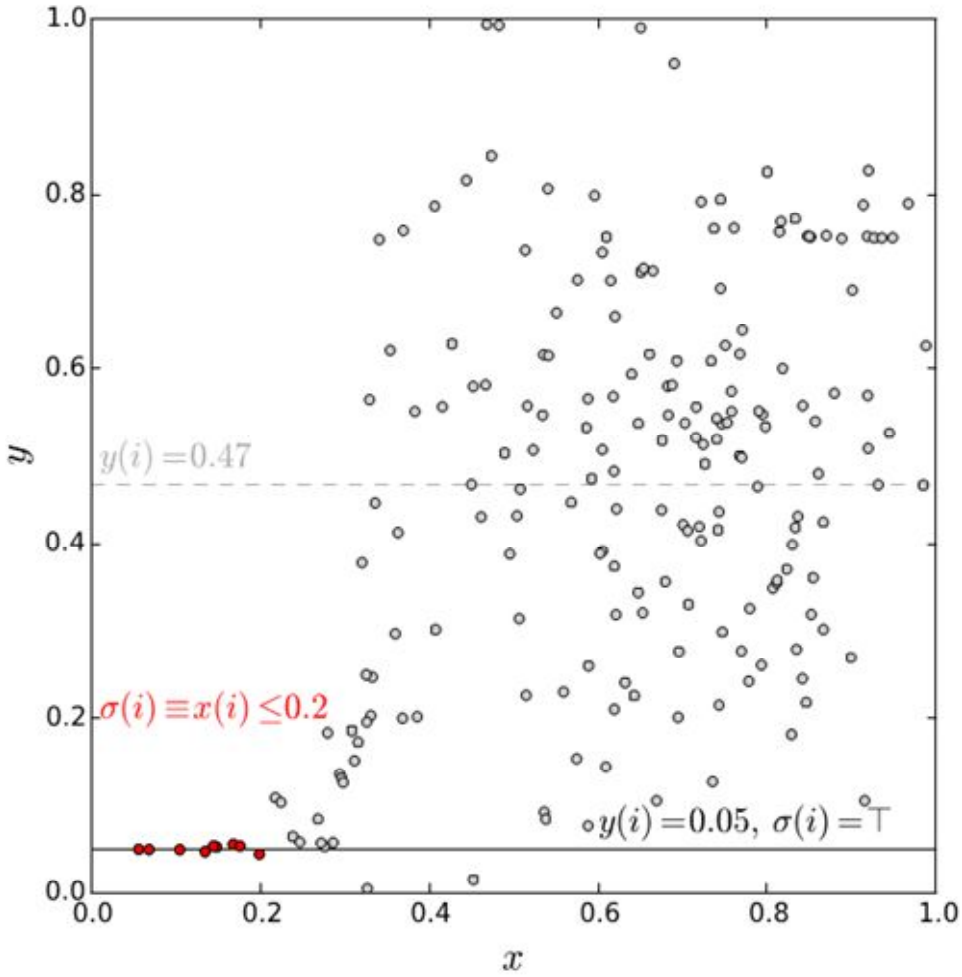
Data mining: Subgroup discovery



M. Boley *et al*., Data Min. Knowl. Disc. 31, 1391 (2017); B. Goldsmith *et al*., New J. Phys. 19, 013031 (2017)

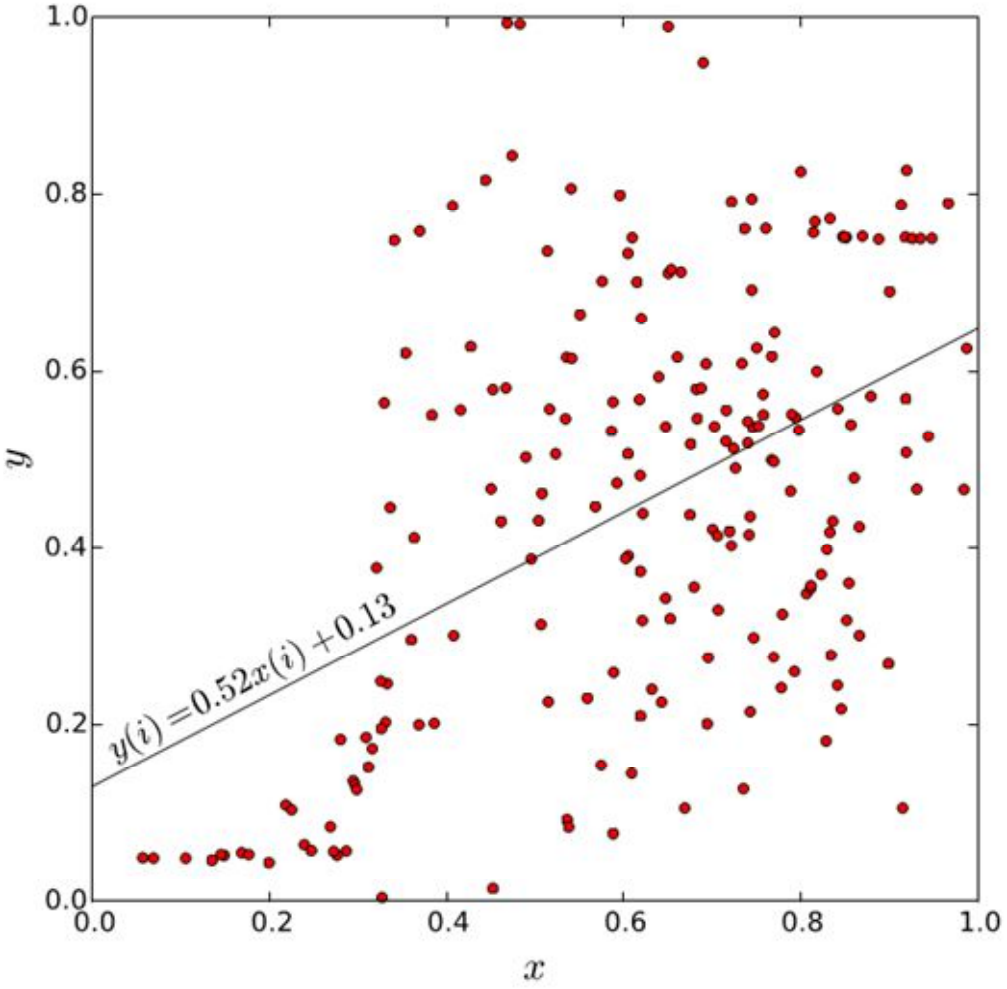
Data mining: Subgroup discovery

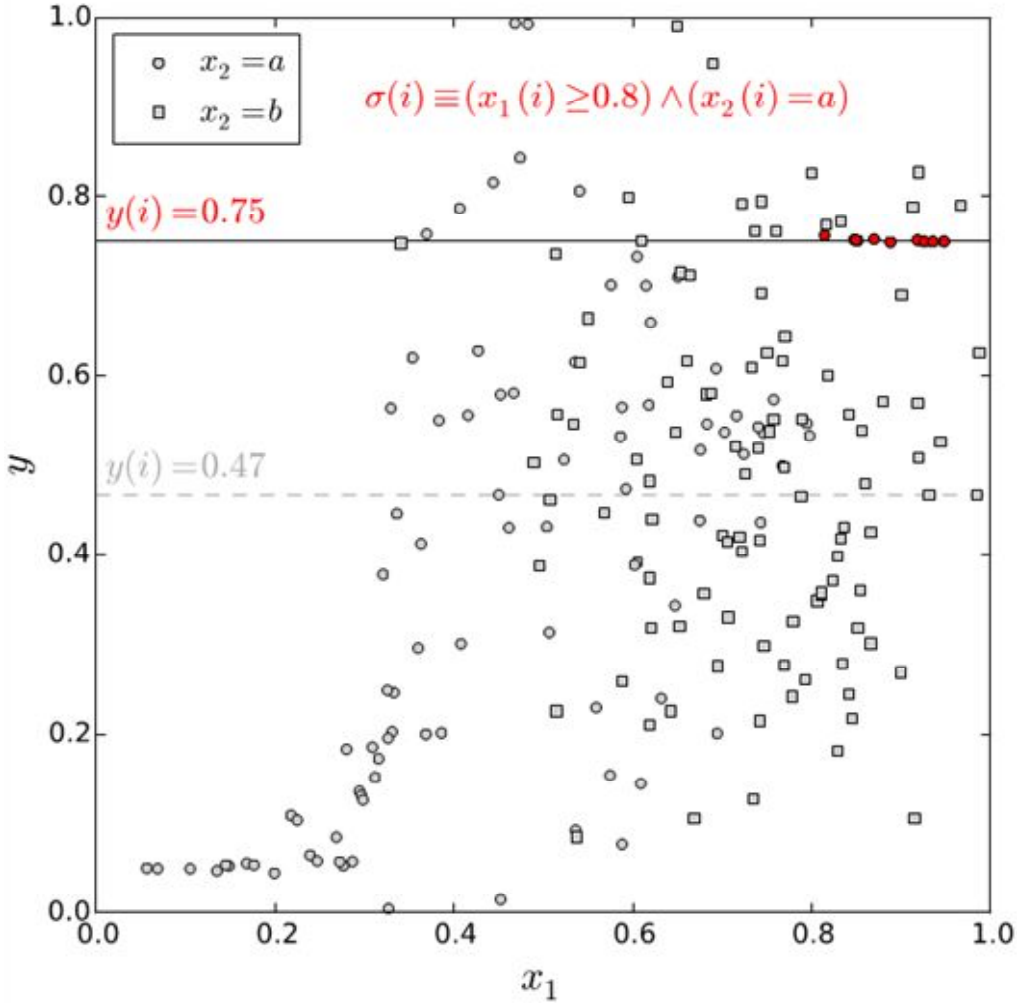




M. Boley *et al*., Data Min. Knowl. Disc. 31, 1391 (2017); B. Goldsmith *et al*., New J. Phys. 19, 013031 (2017)

Data mining: Subgroup discovery



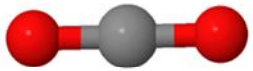


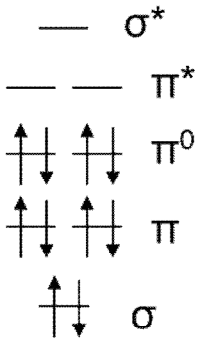
M. Boley *et al*., Data Min. Knowl. Disc. 31, 1391 (2017); B. Goldsmith *et al*., New J. Phys. 19, 013031 (2017)

Subgroup discovery: CO_2 activation by adsorption

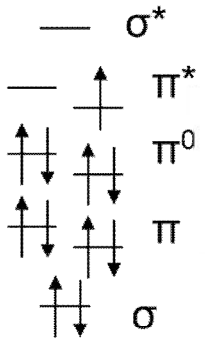
CO_2

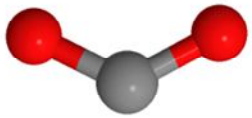
C-O bond elongation, O-C-O bending angle → indicators of activation



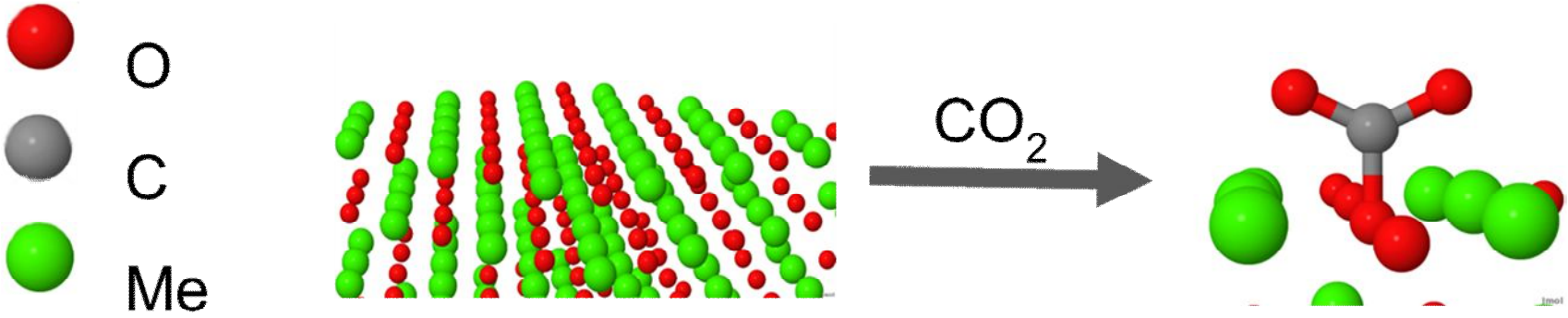


charge transfer





Subgroup discovery: CO_2 activation by adsorption



dry reforming of methane:
$CO_2 + CH_4 = 2H_2 + 2CO$

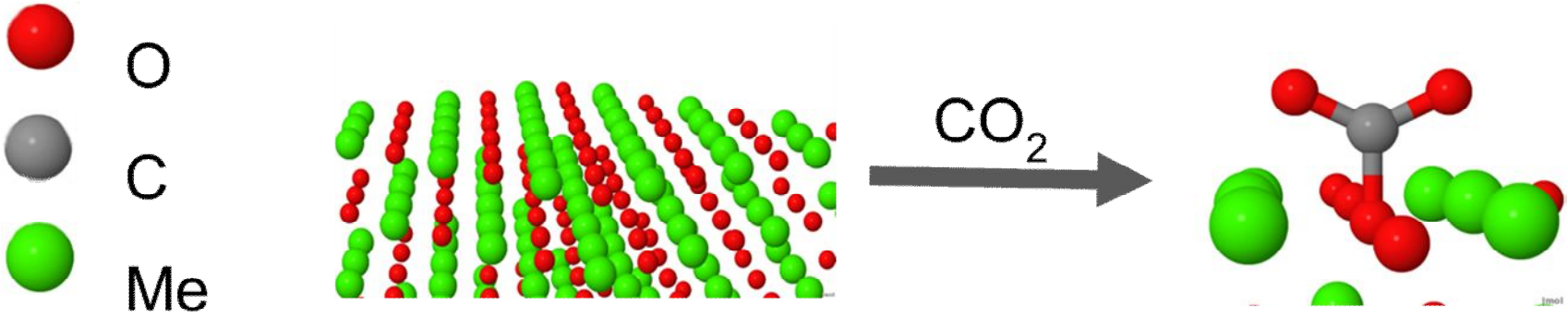
Sabatier reaction:
$CO_2 + 4H_2 = CH_4 + 2H_2O$

partial hydrogenation:
$CO_2 + 3H_2 = CH_3OH + H_2O$

Oxides:

- stable (structurally and compositionally) under increased temperatures;
- more resistant for poisoning;
- activation is frequently observed

Subgroup discovery: CO_2 activation by adsorption



C-O bond elongation, O-C-O bending angle → indicators of activation →

Which surface properties lead to desired indicators?

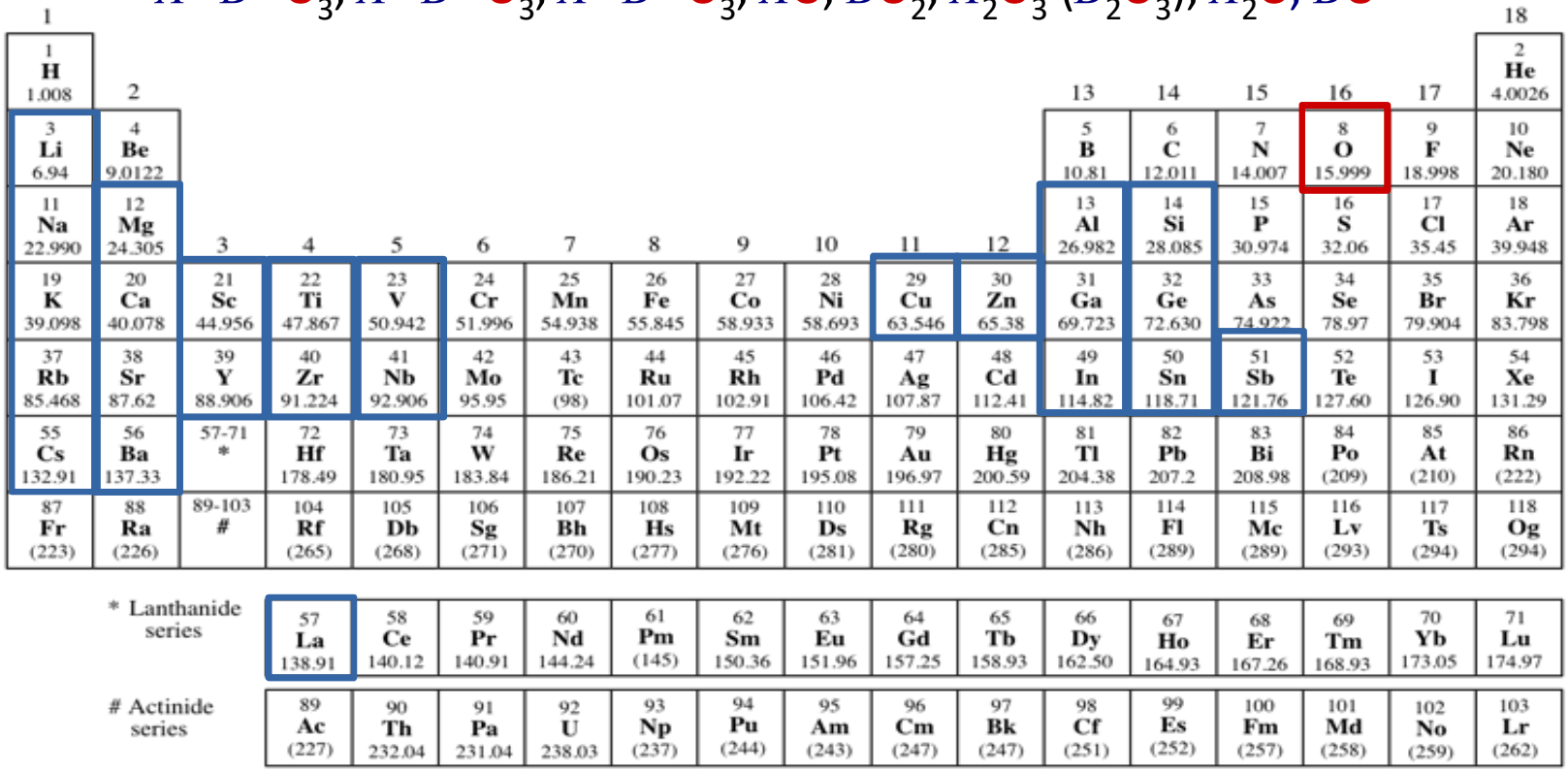
Use subgroup discovery to find materials that optimize activation indicators

$$f = N_{subgroup}/N_{all} \times (mean_{subgroup} - mean_{all}) \times (1 - variance_{subgroup}/variance_{all})$$

Maximize C-O bond length or O-C-O bending

Subgroup discovery: CO_2 activation by adsorption

$A^{2+}B^{4+}O_3$, $A^{3+}B^{3+}O_3$, $A^{1+}B^{5+}O_3$, AO, BO_2, A_2O_3 (B_2O_3), A_2O, BO

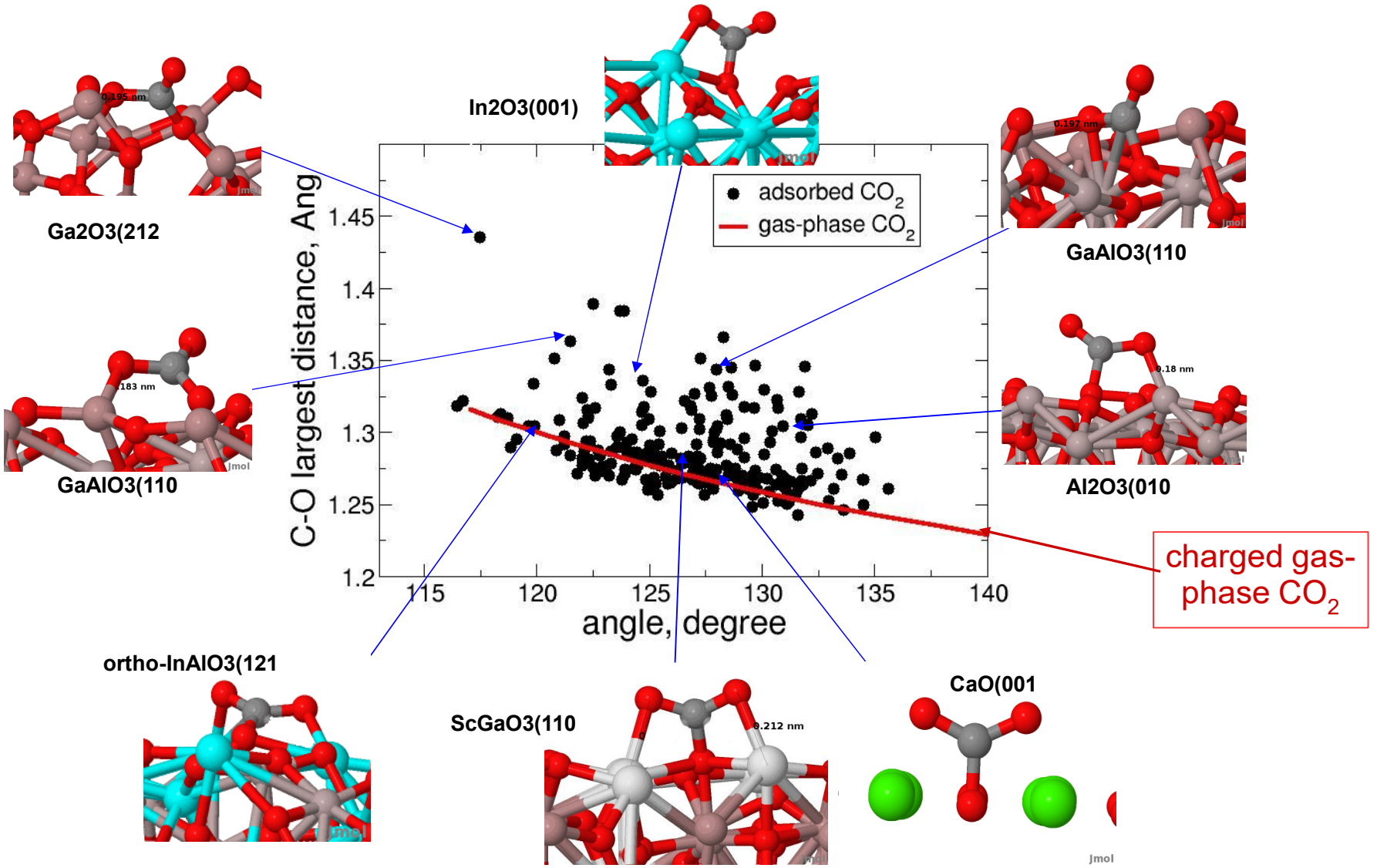


71 oxide materials

141 surfaces with Miller indexes ≤ 2

270 adsorption sites

Subgroup discovery: CO_2 activation by adsorption



Primary features

Atom:

electron affinity ionization potential electronegativity

$r_{l(\mathrm{HOMO})}$, $r_{l\text{-}1}$, r_{l+1} atomic numbers

Material:

work function band gap Cbm surface form. energy

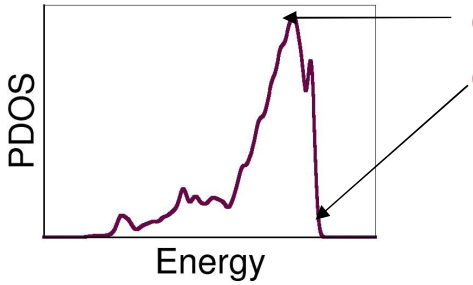
Site-specific features:

electrostatic potential Hirshfeld charge bond-valence of O

coordination number of O vdW C_6-coefficient polarizability

distances to 1st, 2nd, 3d nearest cations local-structure parameters

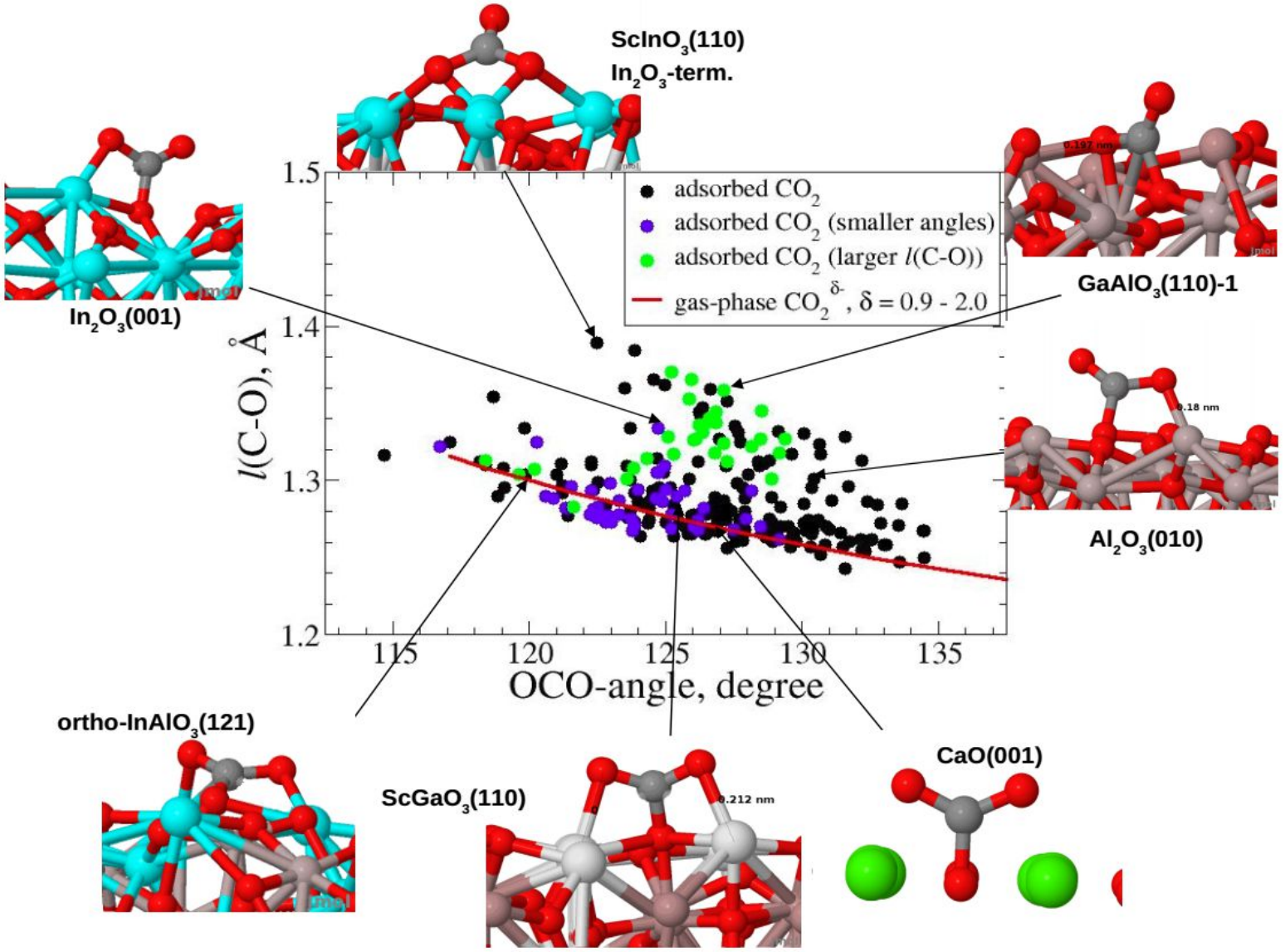
features of
O 2*p*-PDOS



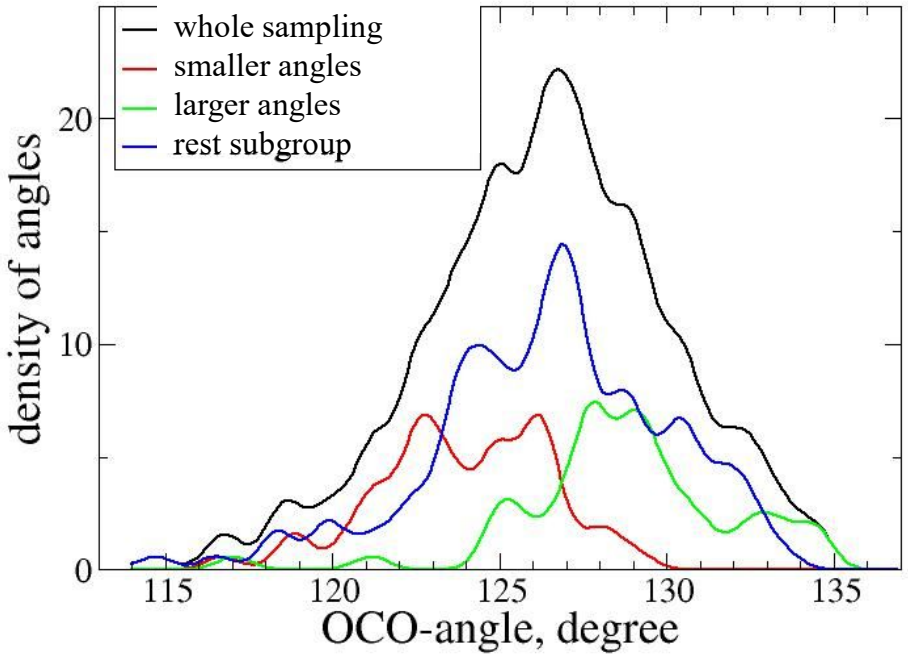
energy of maximum
energy of top

1st, 2nd, 3d, 4th moments
DOS moments: center, width, skewness, kurtosis

Subgroup discovery: Adsorbed CO2 properties

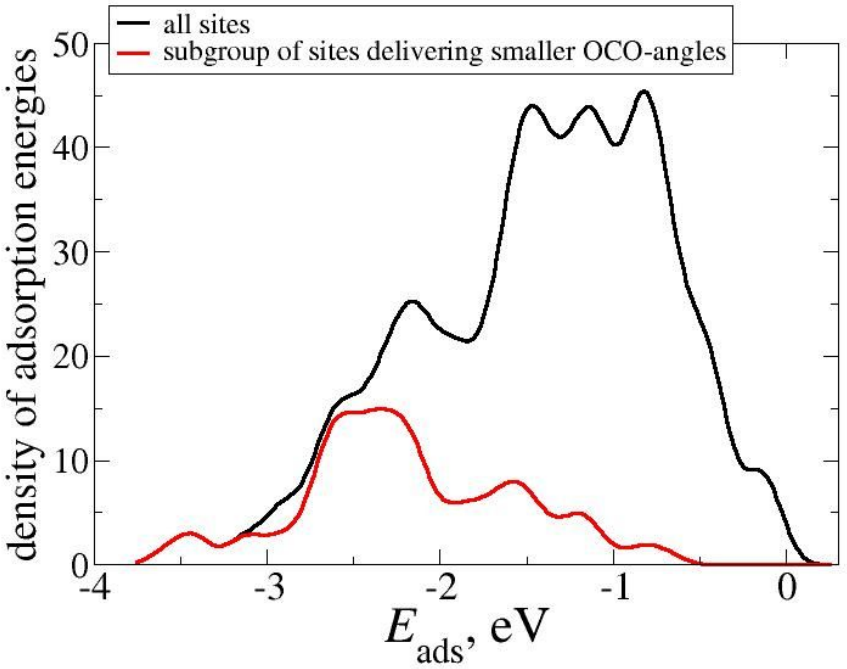


Subgroup discovery: Analysis of the OCO angle



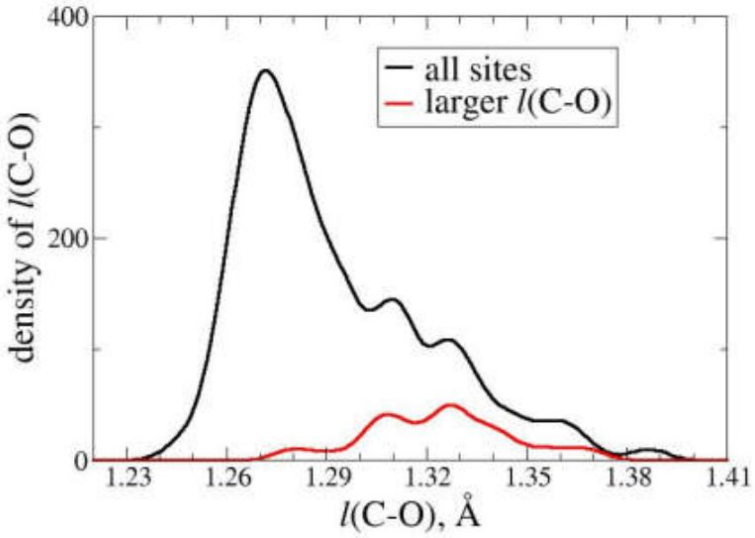
sites delivering smaller angles (59 adsorption sites):

(energy of O 2*p* band maximum > -6.0 eV) AND
(distance from O-site to first nearest cation > 1.8 Å) AND
(distance from O-site to second nearest cation > 2.1 Å)



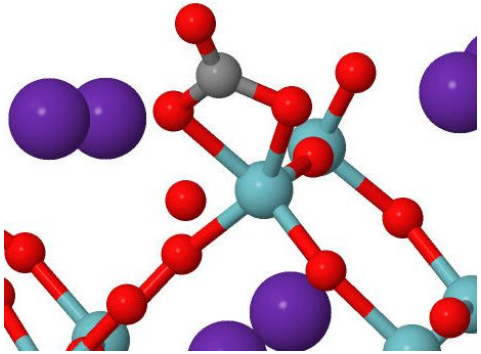
Most of the site delivering smaller OCO angles are on ionic (basic) materials

Subgroup discovery: Analysis of the C-O bond length



sites delivering larger *l*(CO) (33 sites):

(cation charge < 0.5e) AND
(work function ≥ 5.2 eV) AND
(distance from O site to second nearest cation ≥ 2.14 Å)



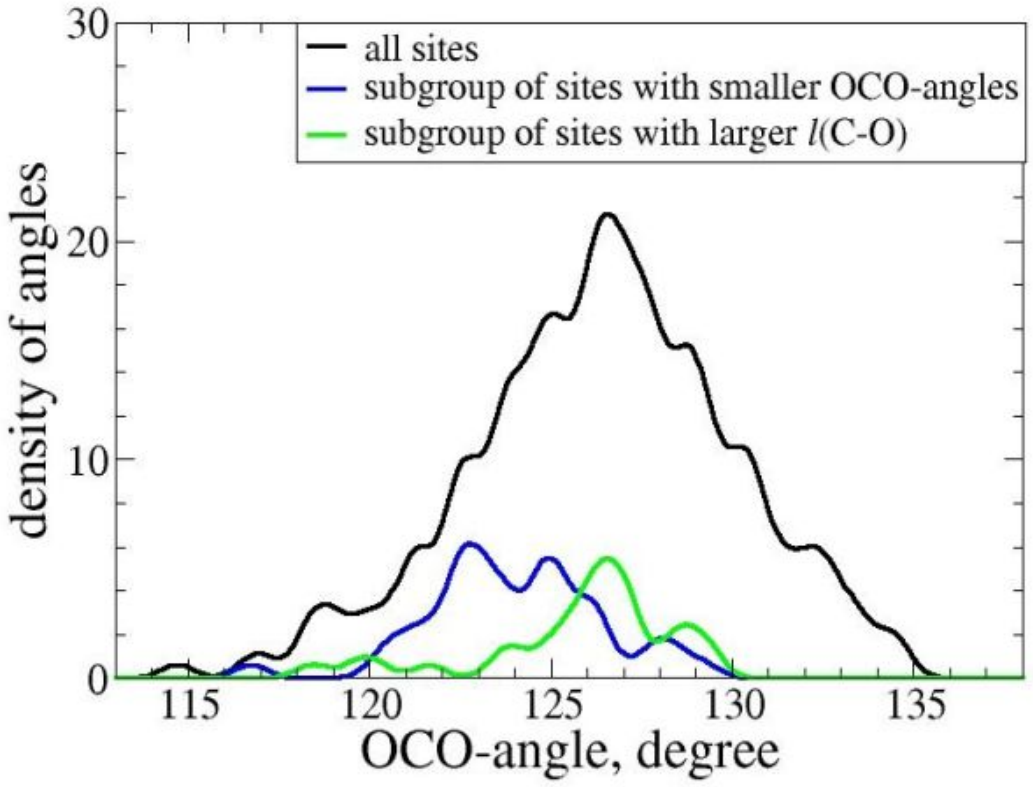
$LaGaO_3$ – cathode material in high-temperature electrochemical CO_2 reduction;

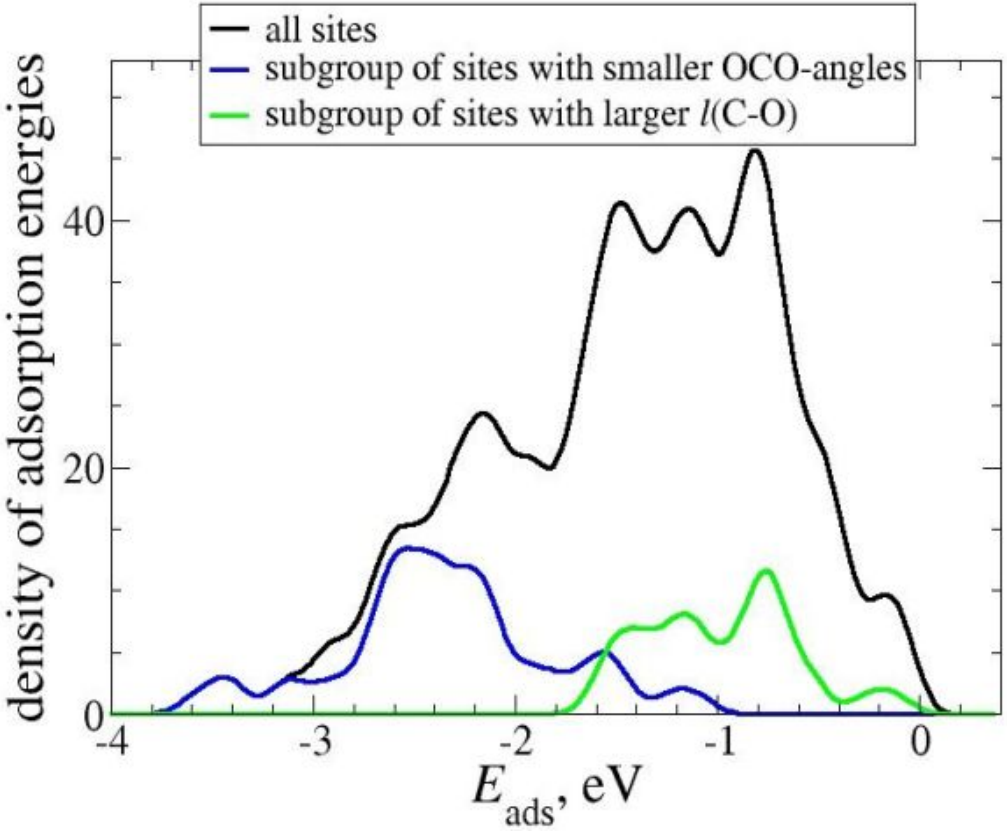
$KNbO_3$ – photocatalytic reduction of CO_2 into CH_4;

$NaNbO_3$ – photocatalyst for CO_2 reduction with ~70% of CO selectivity;

$NaSbO_3$ – material for CO_2 capture and storage (CCS)

Subgroup discovery: Alternative mechanisms of CO_2 activation



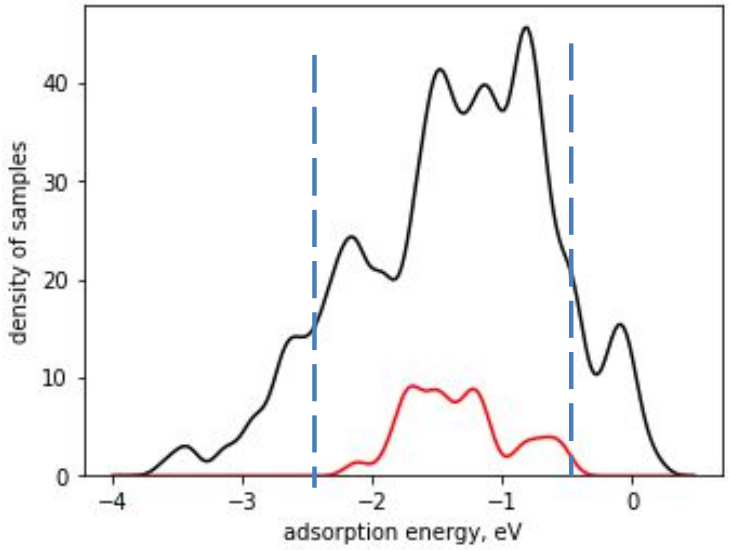


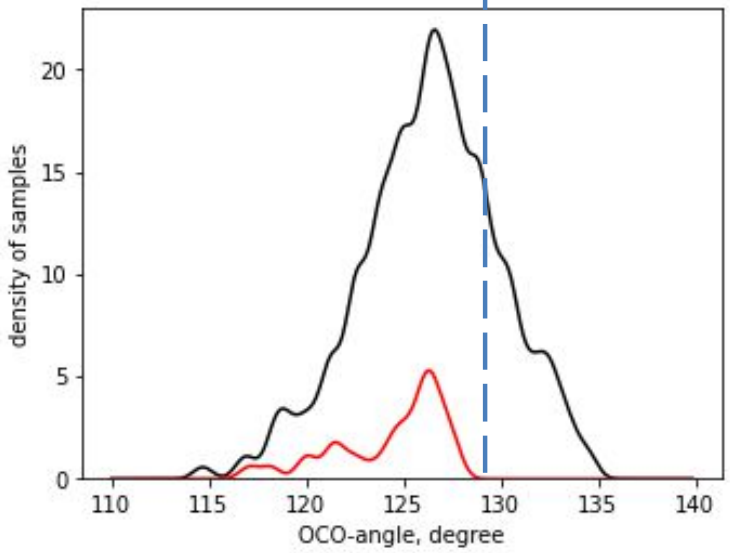
Longer C-O implies smaller OCO angles, but not too small → no catalyst poisoning

Subgroup discovery: A different approach

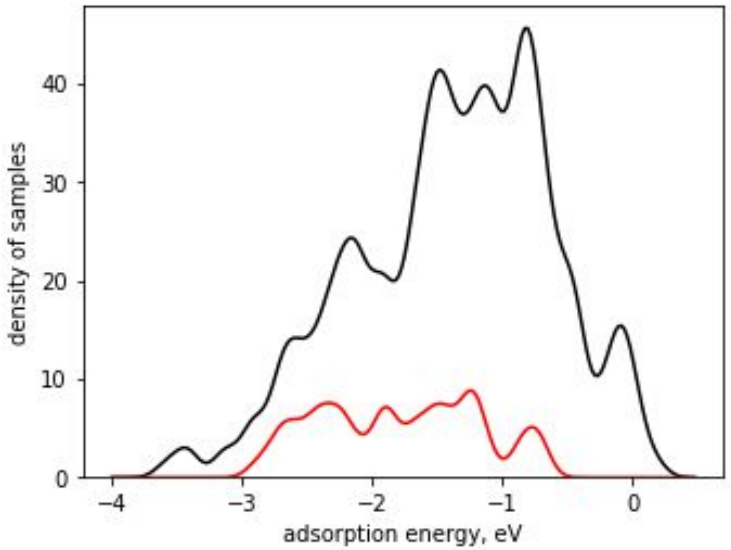
$$f = \left(\frac{N_{\text{subgroup}}}{N_{\text{all}}}\right)\left(\frac{mean_{\text{subgroup}} - mean_{\text{all}}}{min/max_{\text{all}} - mean_{\text{all}}}\right)\left(1 - \frac{variance_{\text{subgroup}}}{variance_{\text{all}}}\right)$$

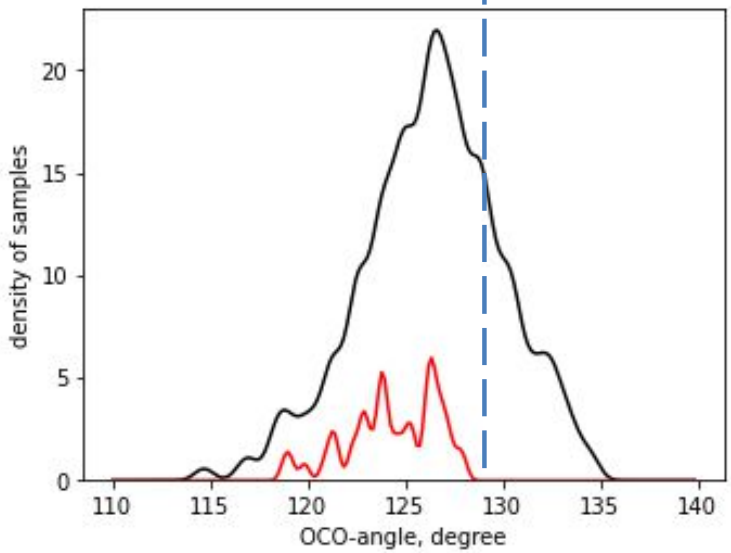
$$f = \left(\frac{N_{\text{subgroup}}}{N_{\text{all}}}\right)\left(\frac{min/max_{\text{subgroup}} - target_{\text{gas}}}{min/max_{\text{all}} - target_{\text{gas}}}\right)[\Theta(OCO < a)][\text{information gain}(E_{\text{ads}} \in (-2.5, -0.5))]$$



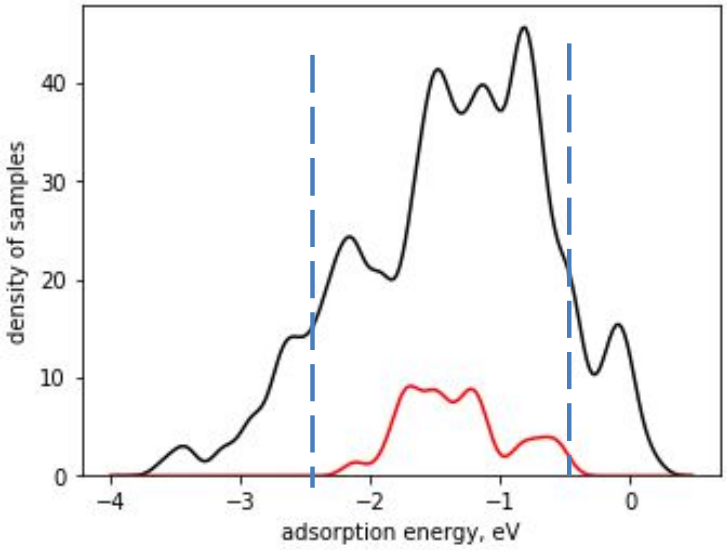


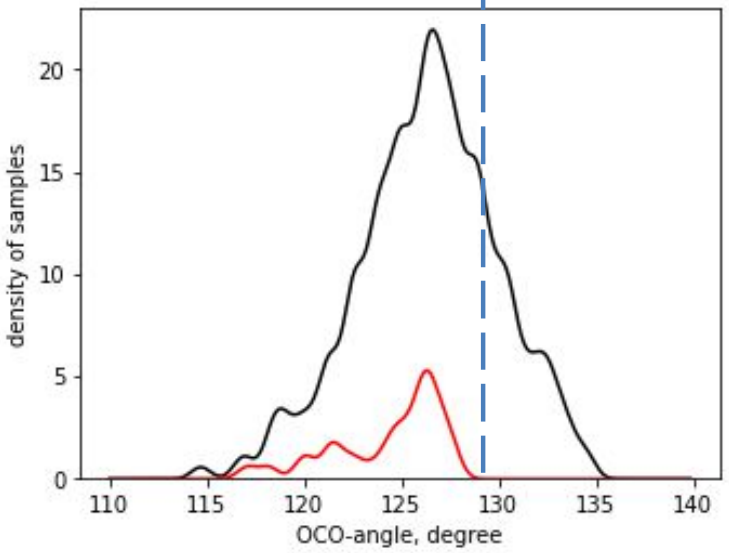
Subgroup discovery: A different approach





large Hirshfeld charge on surface O, lower coordination for smaller angles





with adsorption energy constraint:

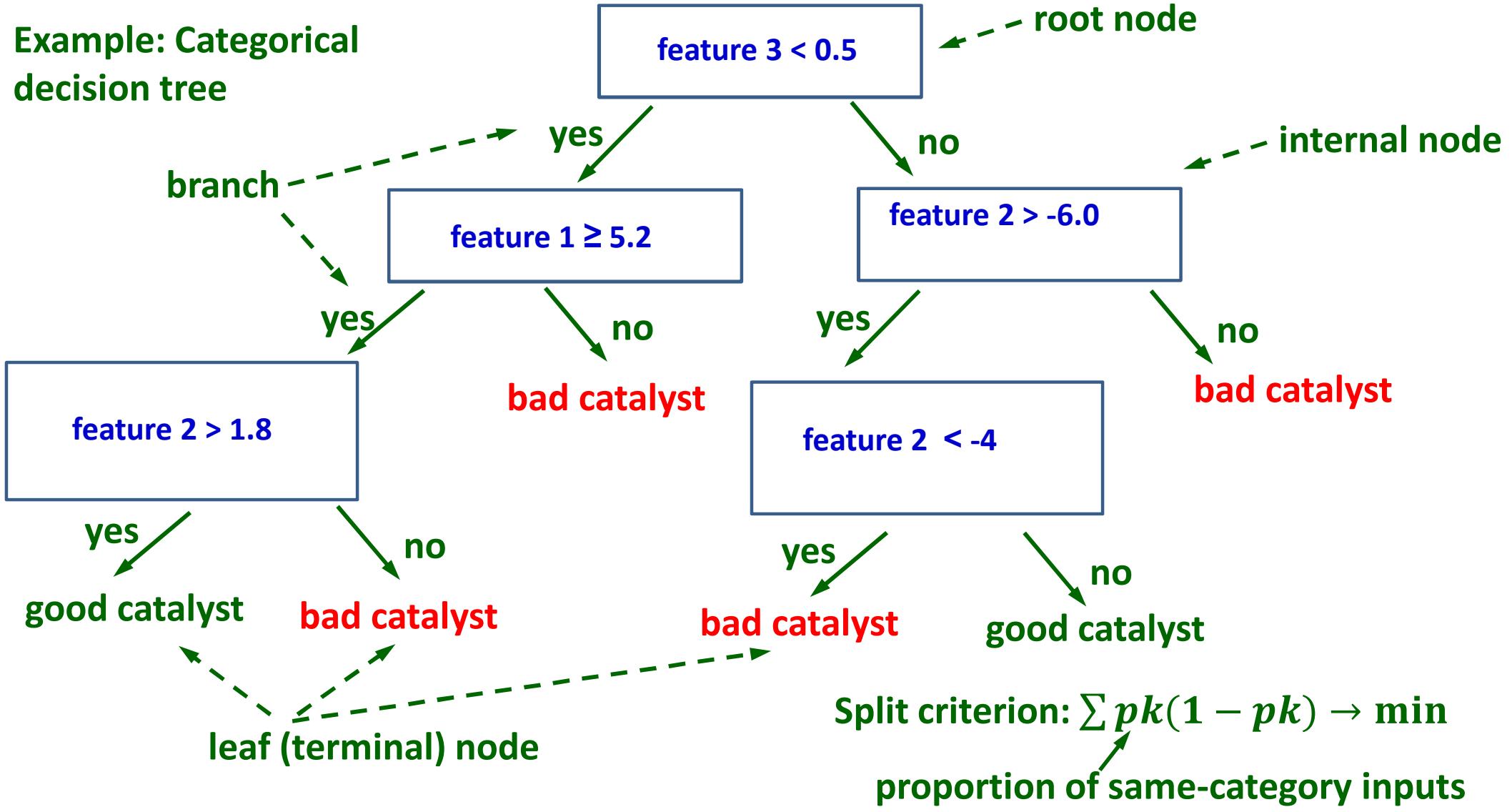
smaller charge on surface O, delocalized electron density, binding of O in CO_2 with surface cations

SISSO and SGD software

SISSO: https://github.com/rouyang2017/SISSO

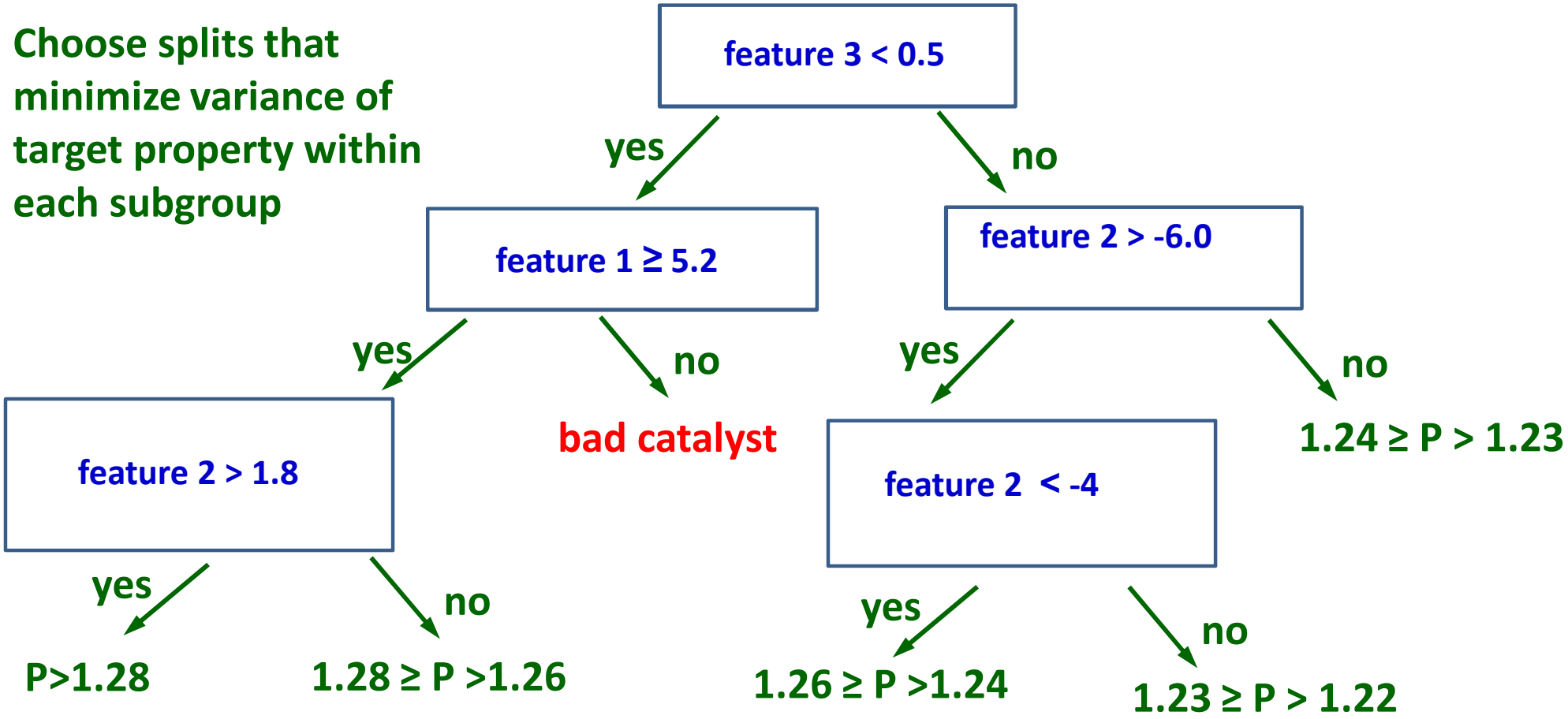
Subgroup discovery: http://www.realkd.org/

Decision trees



Decision tree regression

Choose splits that minimize variance of target property within each subgroup



Split criterion: $\sum(\text{target property} - \langle\text{target property}\rangle)^2 \rightarrow \min$ within each subgroup

Decision tree properties

- Simple to understand and interpret
- Global (important difference to subgroup discovery, which finds *locally unique* groups)
- Easy to overfit (can use LASSO-type penalty to solve this problem)
- Small change in data can lead to large change in the tree
- Relatively inaccurate

Random forest®

1) Perform tree regression or classification on several randomly selected subsets of data

2) In each tree, at each split choose randomly a fixed number of features, for which the best split is determined

3) Average predictions from the obtained trees

Properties:

- More accurate than a single tree (“each tree keeps other trees from making mistakes)
- Interpretability of the model is lost
- Can be use to select primary features for other approaches such as SISSO

Random forest®

Interesting application: Identify most important surface structural features that determine surface stability

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Automatic Prediction of Surface Phase Diagrams Using Ab Initio Grand Canonical Monte Carlo

Robert B. Wexler,[†] Tian Qiu,[†] and Andrew M. Rappe*

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Chemical Pressure-Driven Enhancement of the Hydrogen Evolving Activity of Ni_2P from Nonmetal Surface Doping Interpreted via Machine Learning

Robert B. Wexler,[†] John Mark P. Martirez,[‡] and Andrew M. Rappe*,[†]