# Advanced Materials Modeling:

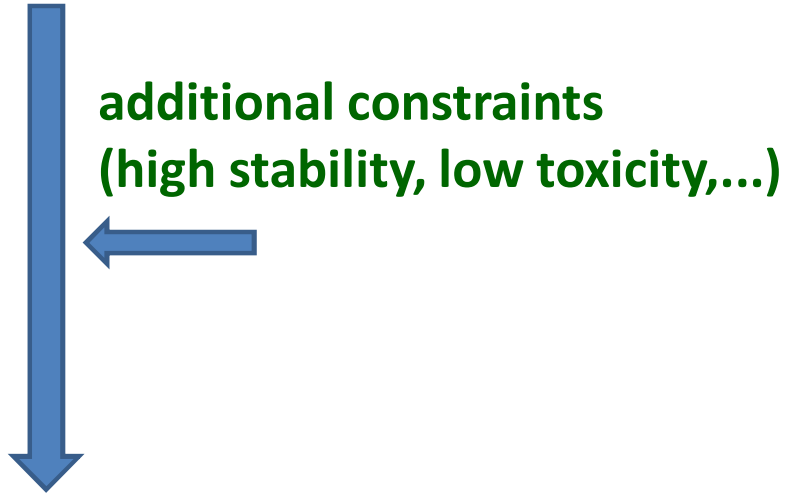
# Exploring Materials Space

*Center for Energy Science and Technology (CEST)*
*Skolkovo Institute of Science and Technology*
*Moscow, Russia*

# High-throughput computational materials design

## Top-down design:

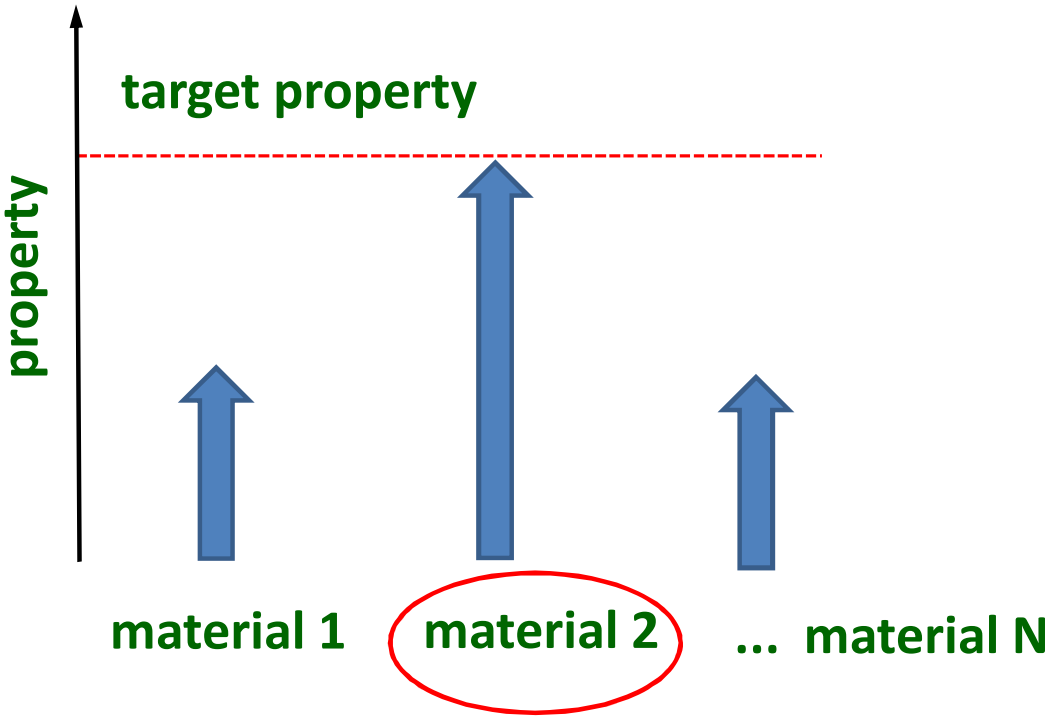
target property (high activity and selectivity of a catalyst)




synthesis recipe

not clear how to achieve this!

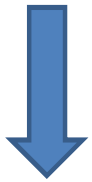
## Bottom-up design:




high-throughput screening

# The key issue: Complexity

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

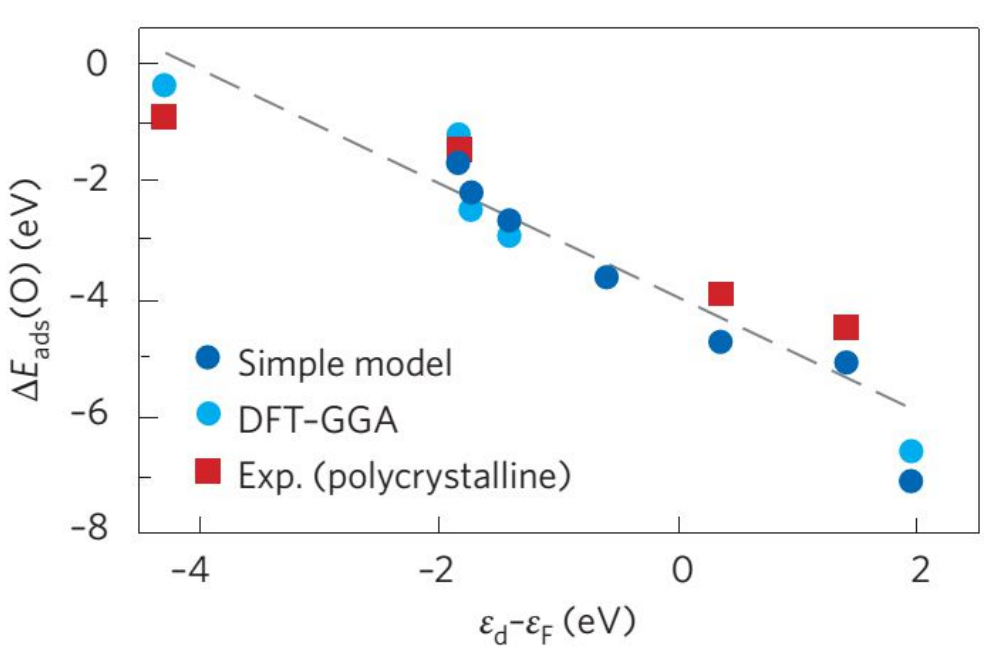


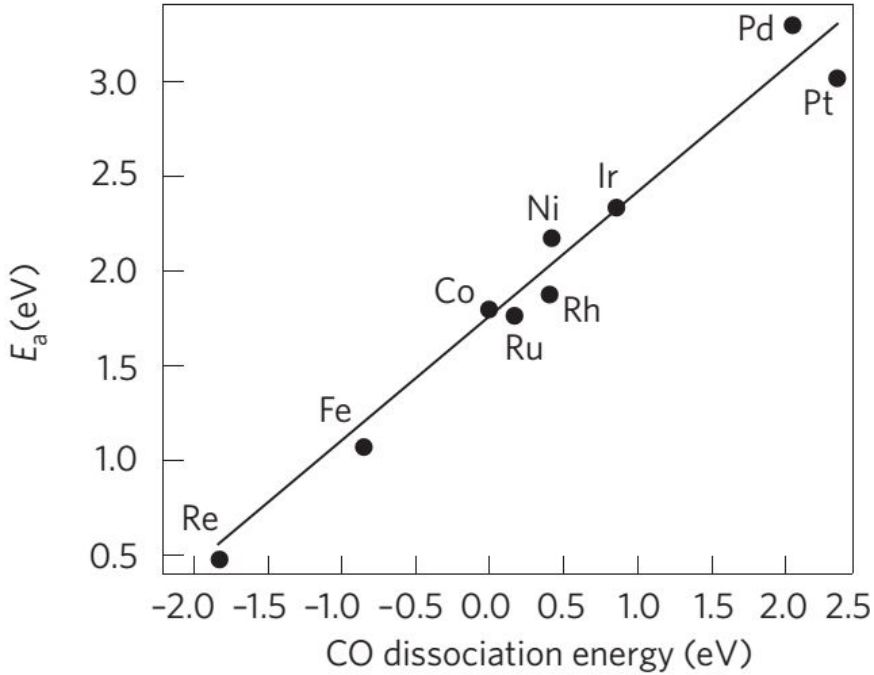
1) **Many-body problem ($3(n+N)$-dimensional)**

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

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

# Descriptors



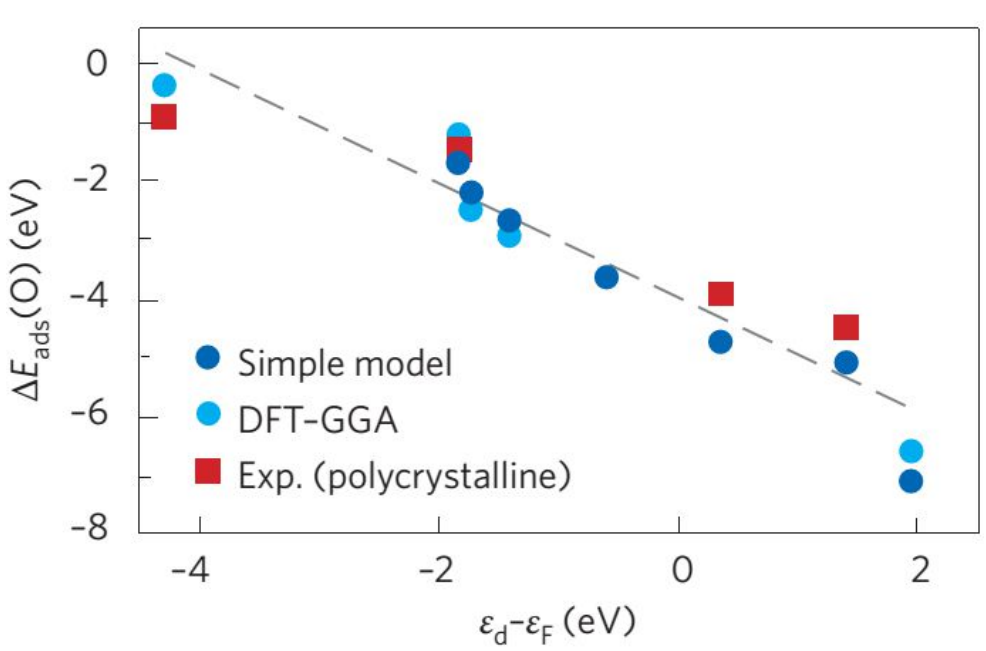



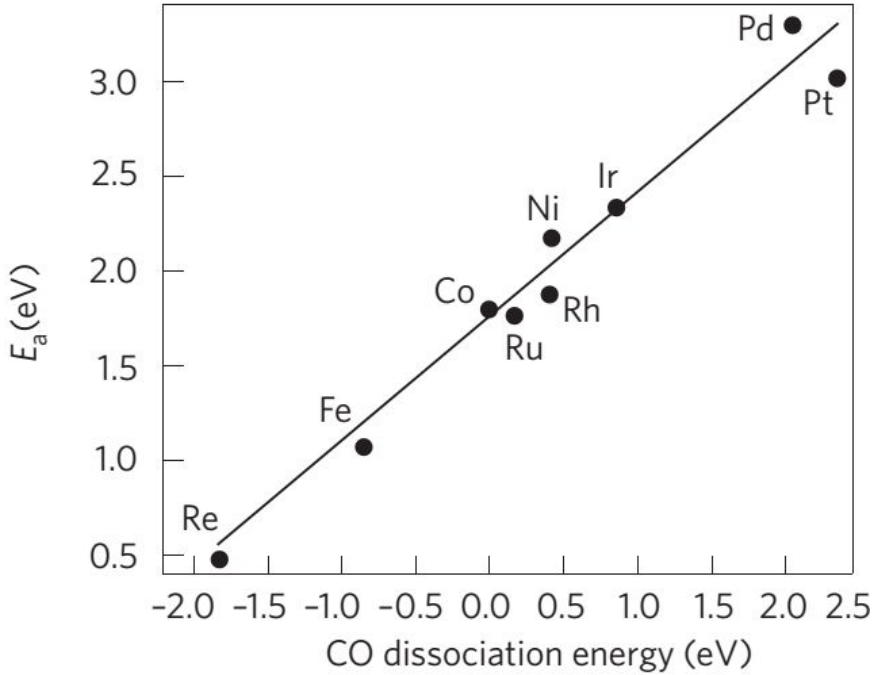

**Simple(r) properties (bulk d-band center position and CO dissociation energy) are correlated to more complex properties (adsorption energy and reaction barrier)**

**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



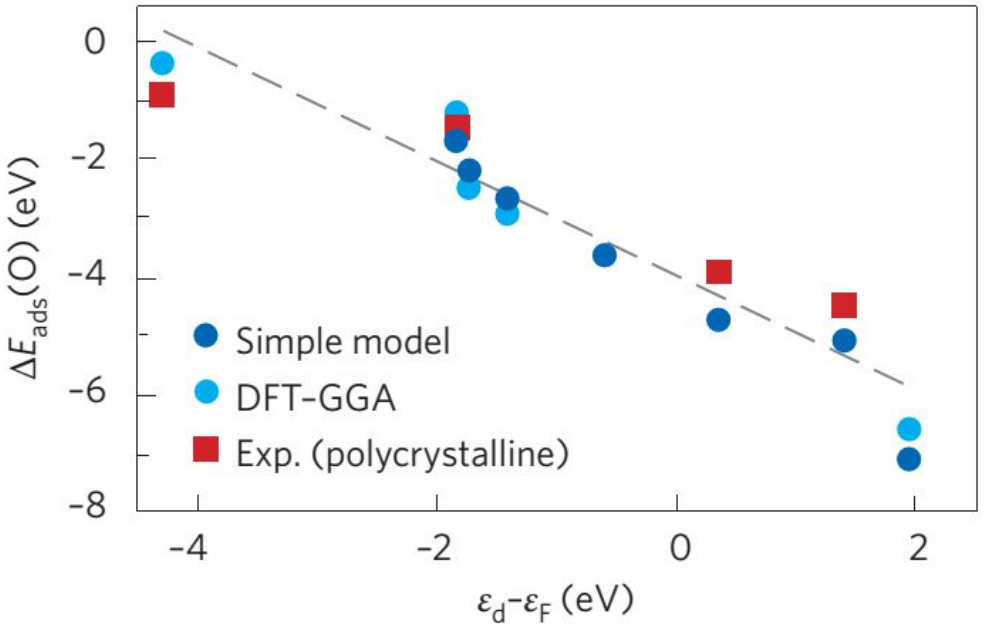



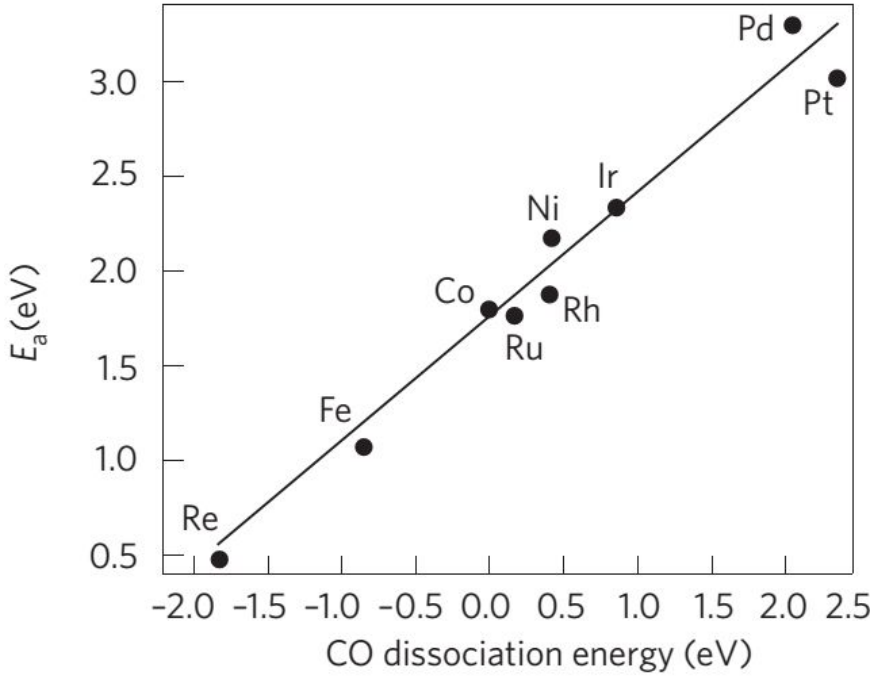

**A simple physical model (Newns-Anderson) motivates the *d*-band center descriptor**

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

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

# Descriptors



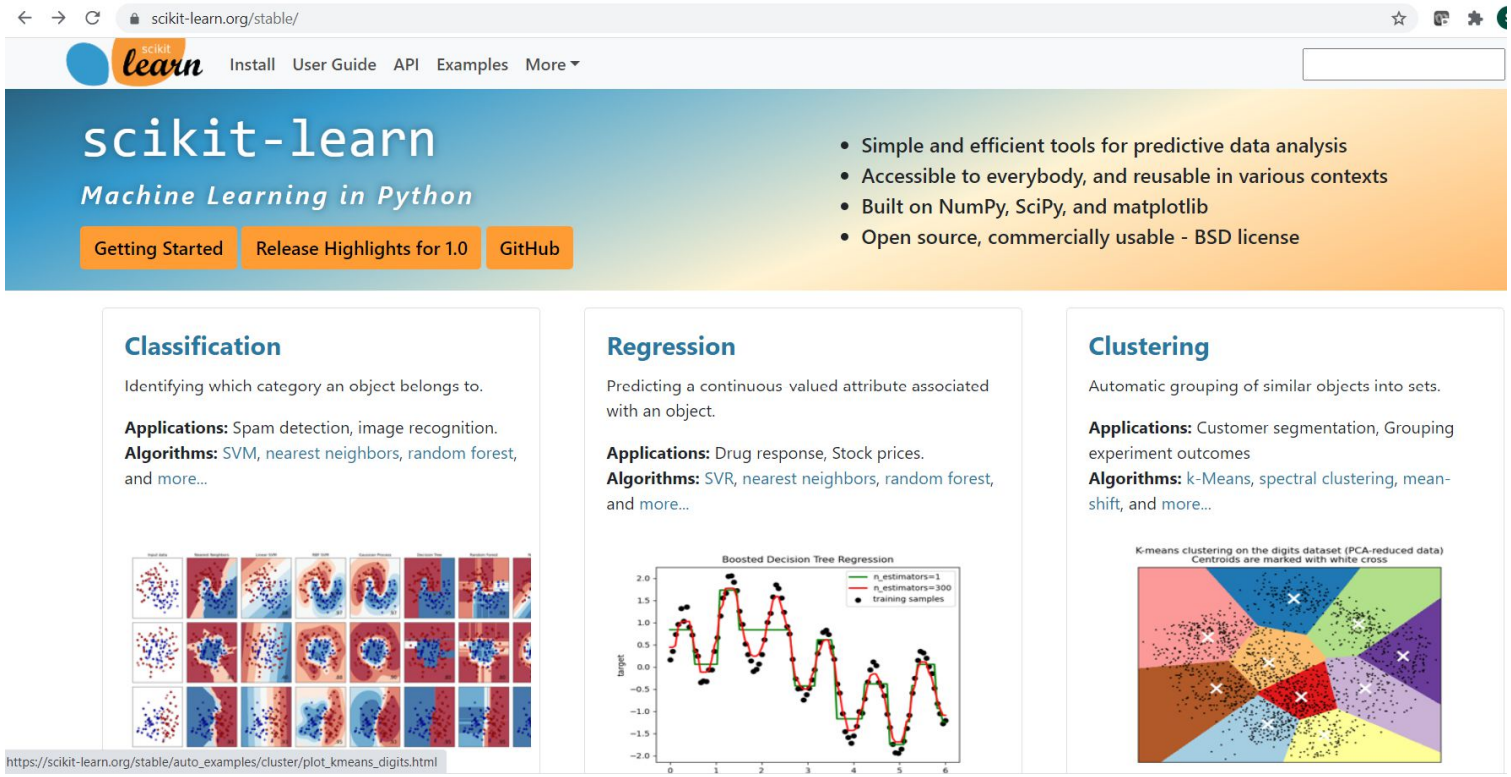

**A simple physical model (Newns-Anderson) motivates the *d*-band center descriptor**

**Find descriptor from DATA!**

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

# Artificial intelligence (including machine learning)

- Neural networks
- Bayesian inference
- Clustering
- Kernel ridge regression
- Symbolic regression
- Decision trees
- Data mining

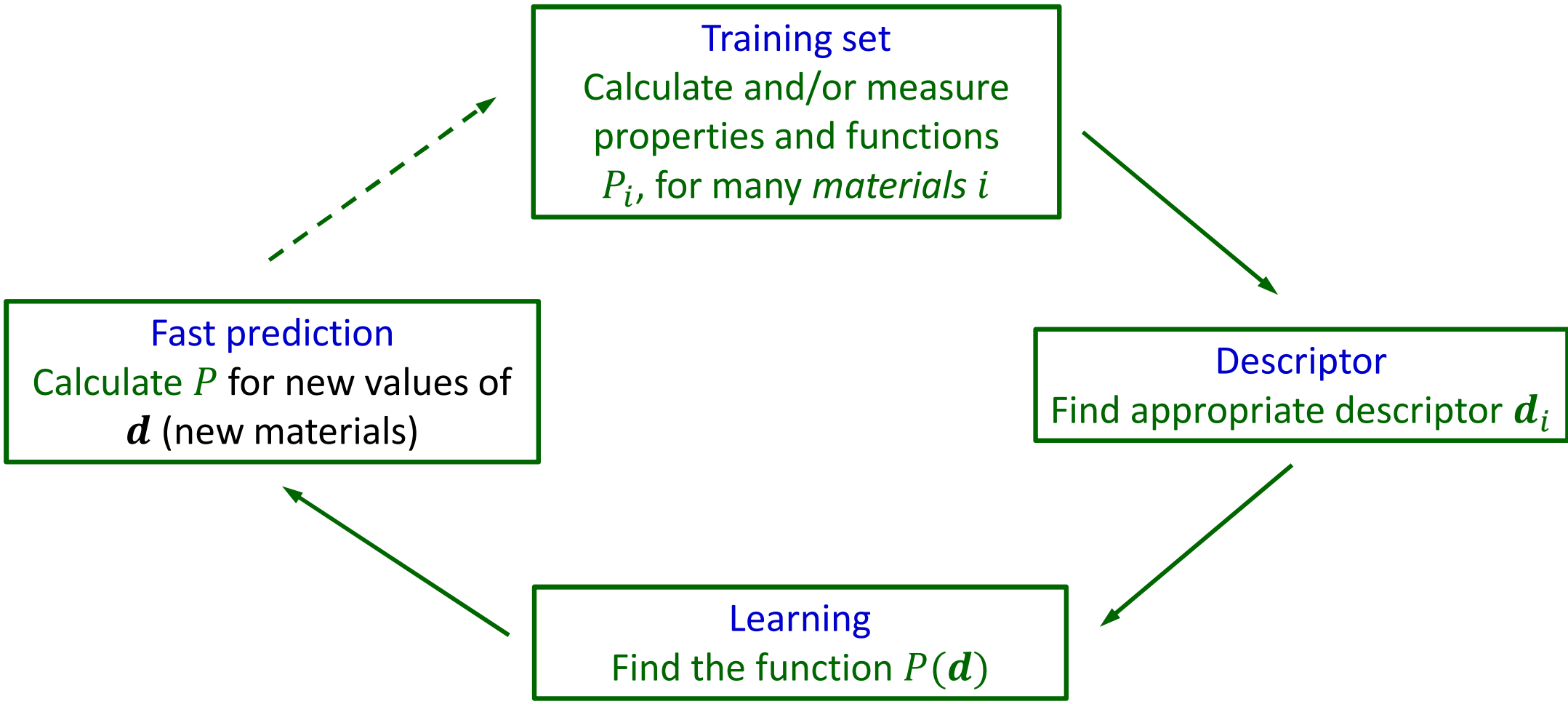



**We develop: Compressed-sensing based symbolic regression**

**Subgroup discovery (data mining)**

**Can work with small data sets, give physically interpretable results**

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

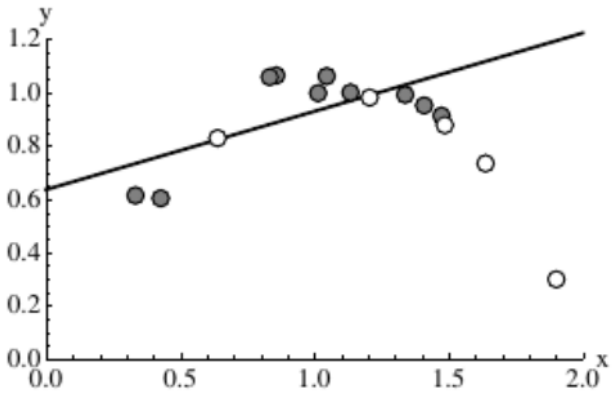
minimize

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

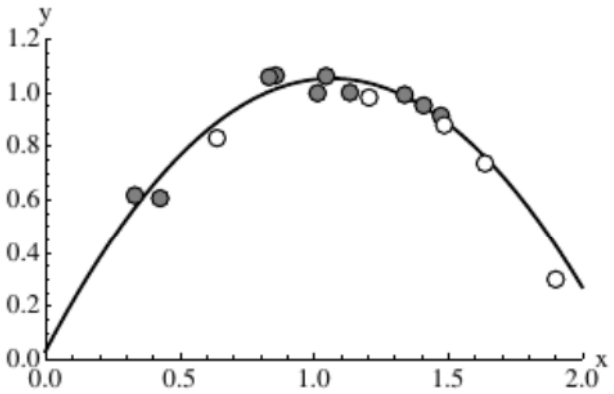
# Regression: Importance of regularization

● training ○ validation

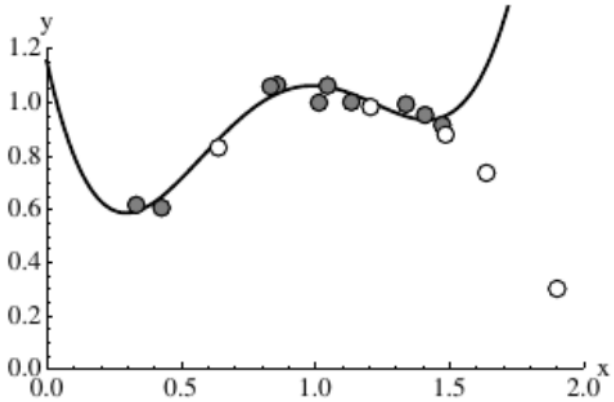
Underfitting




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 |
| --- | --- |
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| 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

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

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

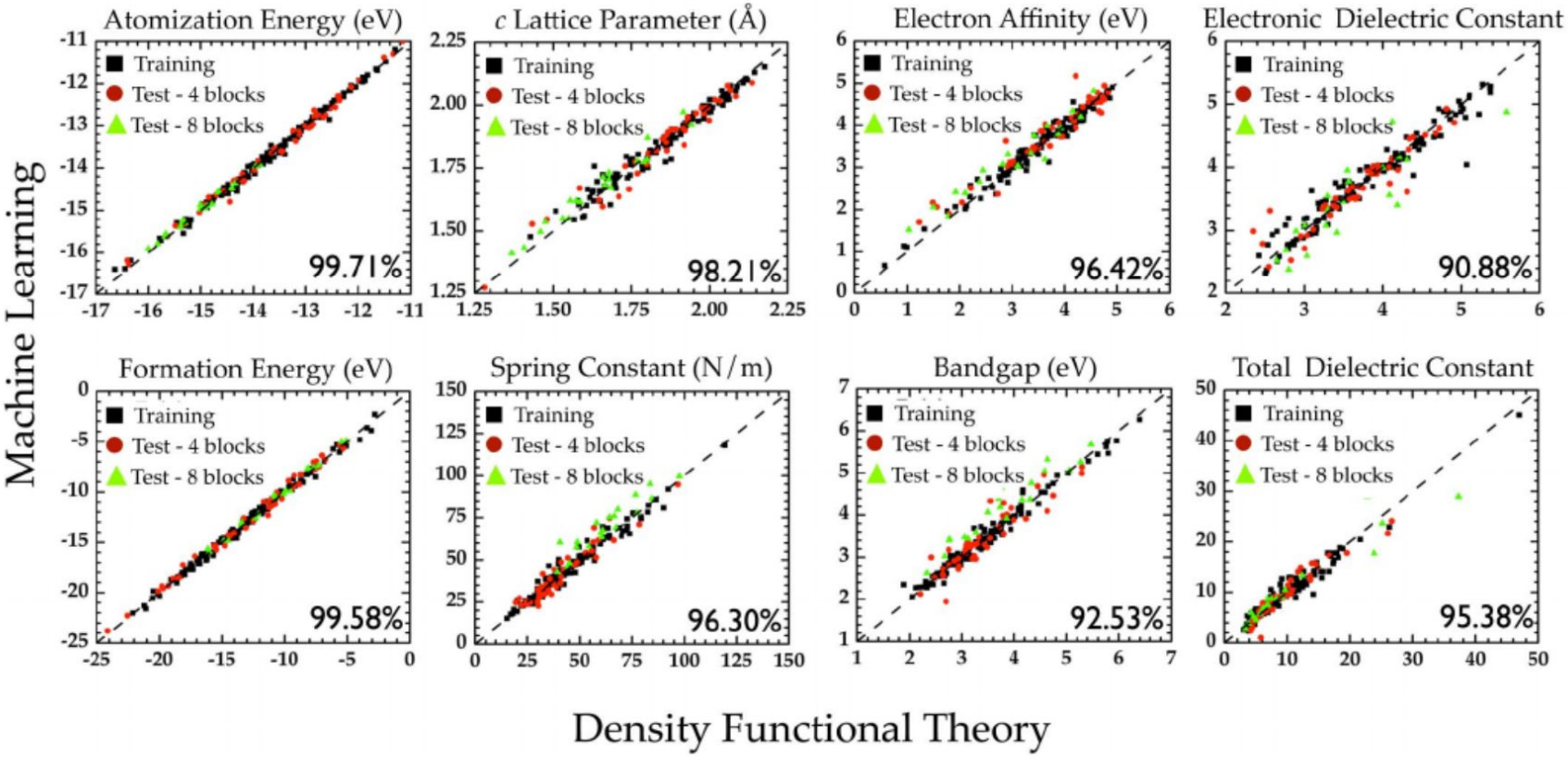
$$\lambda \|\boldsymbol{c}\|_0$$

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

# 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

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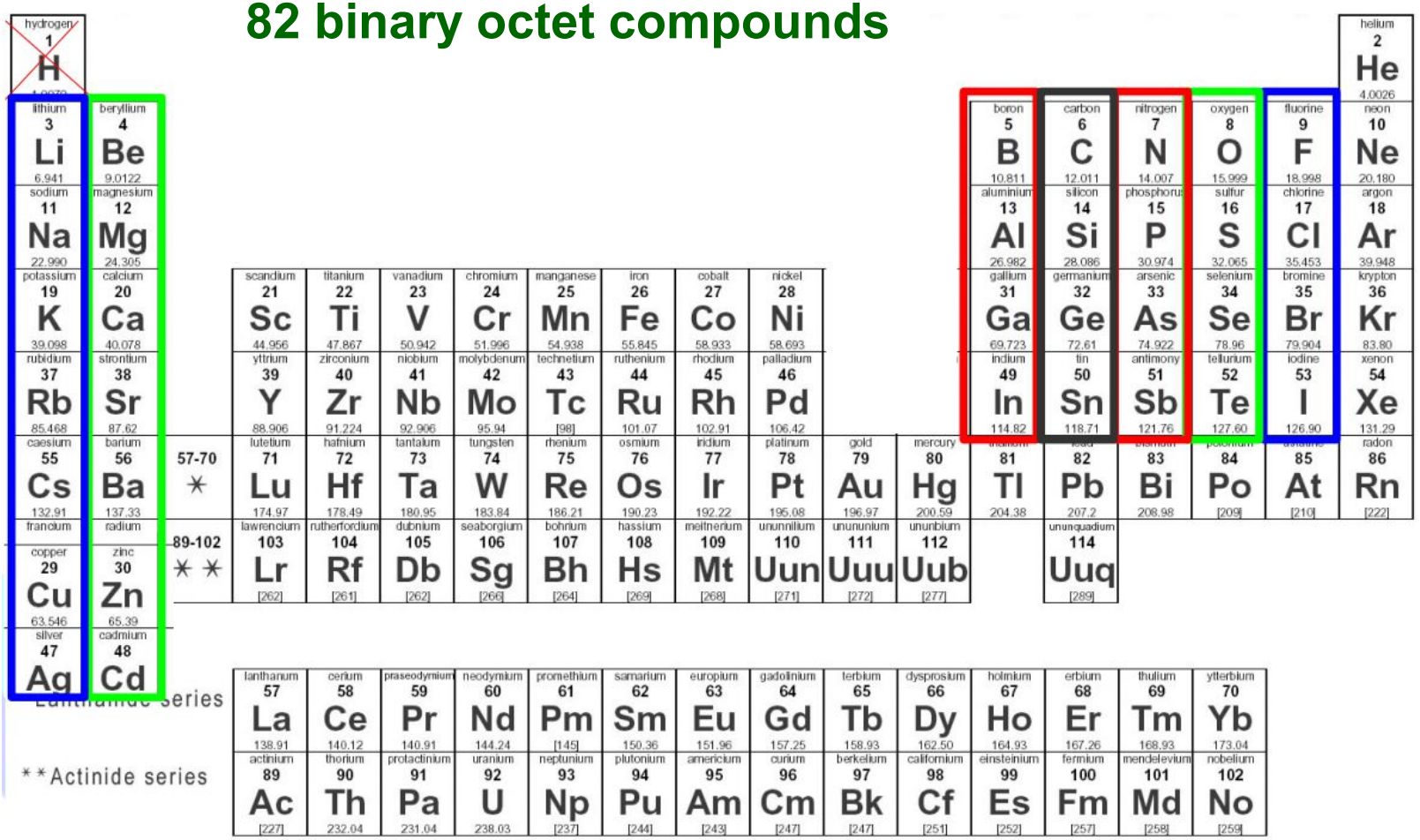
**Idea: calculate many *physically motivated* quantities (features), and use these features as a basis for the physical model under compactness constraints**

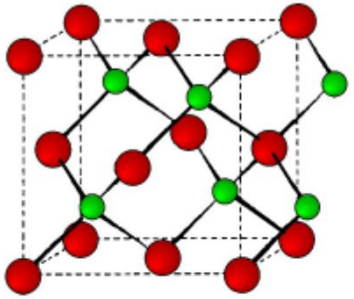
L. M. Ghiringhelli, J. Vybiral, S. V. Levchenko, C. Draxl, and M. Scheffler, Phys. Rev. Lett. **114**, 105503 (2015)

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

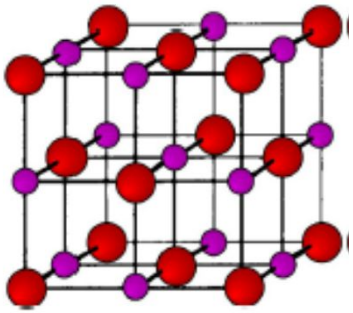
## 82 binary octet compounds




ZB



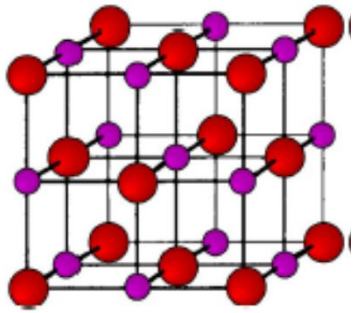
RS



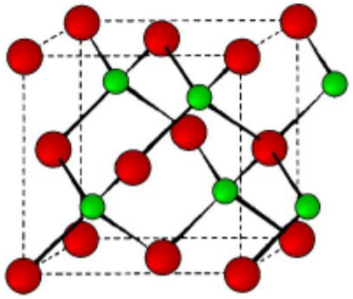
# Proof of Concept: Descriptor for the Classification "Zincblende/Wurtzite (ZB/W) or Rocksalt (RS)?"

**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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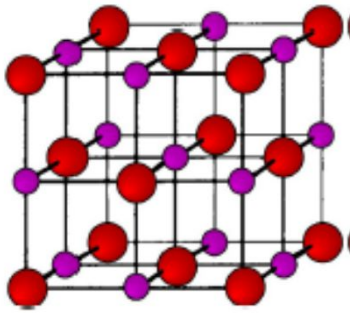
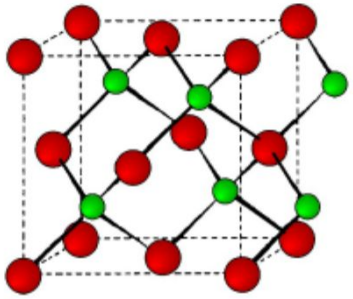


ZB



**J. A. van Vechten**, Phys. Rev. 182, 891 (1969). **J. C. Phillips**, Rev. Mod. Phys. 42, 317 (1970).
**J. John and A.N. Bloch**, Phys. Rev. Let. 33, 1095 (1974) **J. R. Chelikowsky and J. C. Phillips**, Phys. Rev. B 33, 2453 (1978)
**A. Zunger**, Phys. Rev. B 22, 5839 (1980).
**D. G. Petifor**, Solid State Commun. 51, 31 (1984). **Y. Saad, D. Gao, T. Ngo, S. Bobbit, J. R. Chelikowsky, and W. Andreoni**, Phys. Rev. B 85, 104104 (2012).

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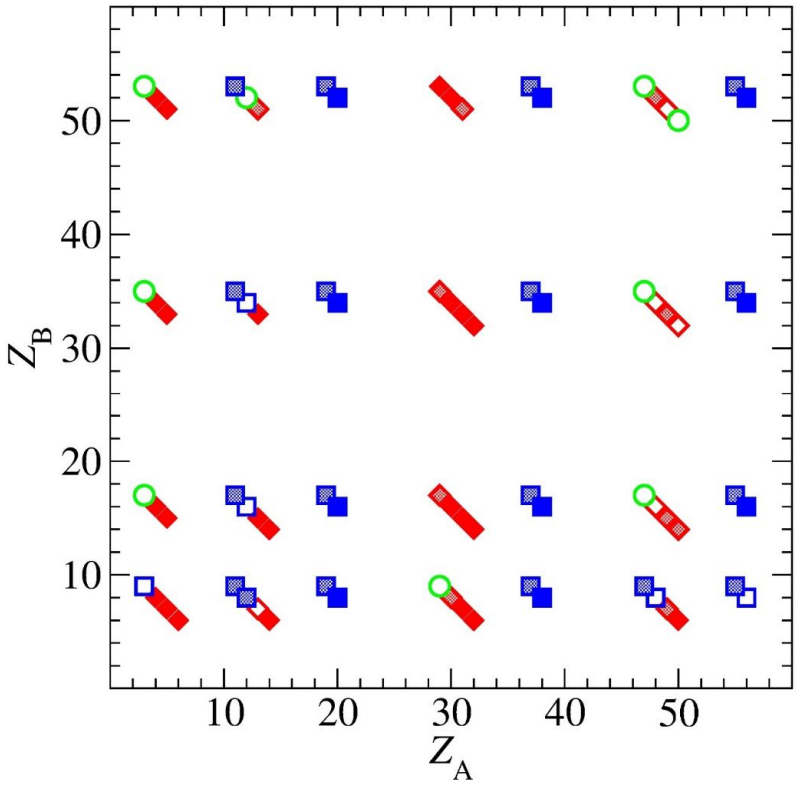
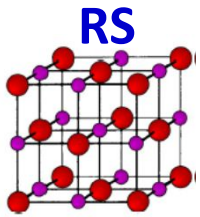
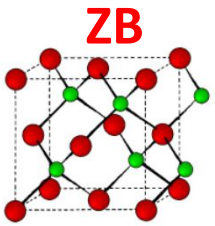
Can we predict not yet calculated structures from $Z_A$ and $Z_B$? **Can we create a map: "The *ZB/W* community lives here and the *RS* community there?"**




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

- 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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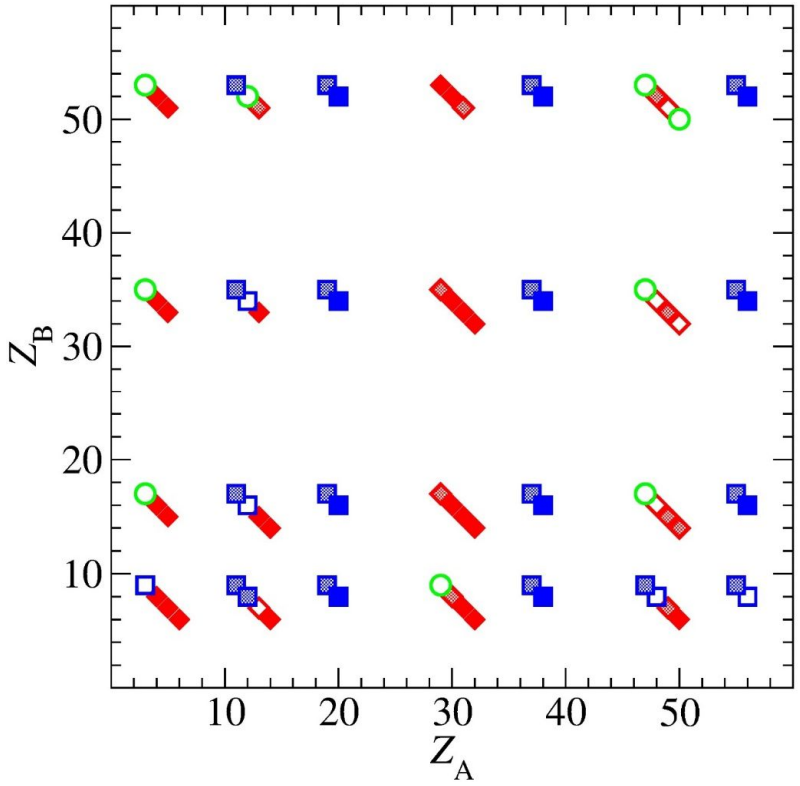
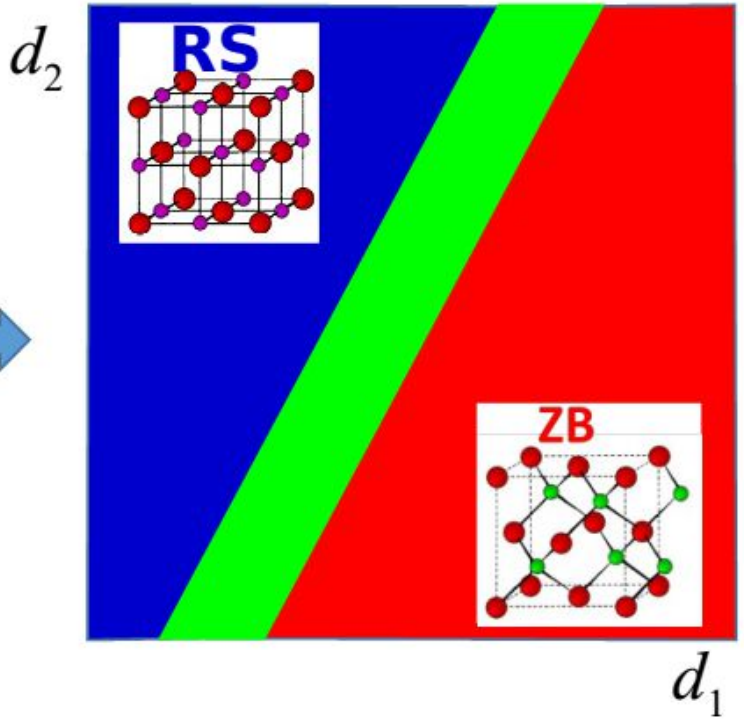
No complexity reduction → need a better basis







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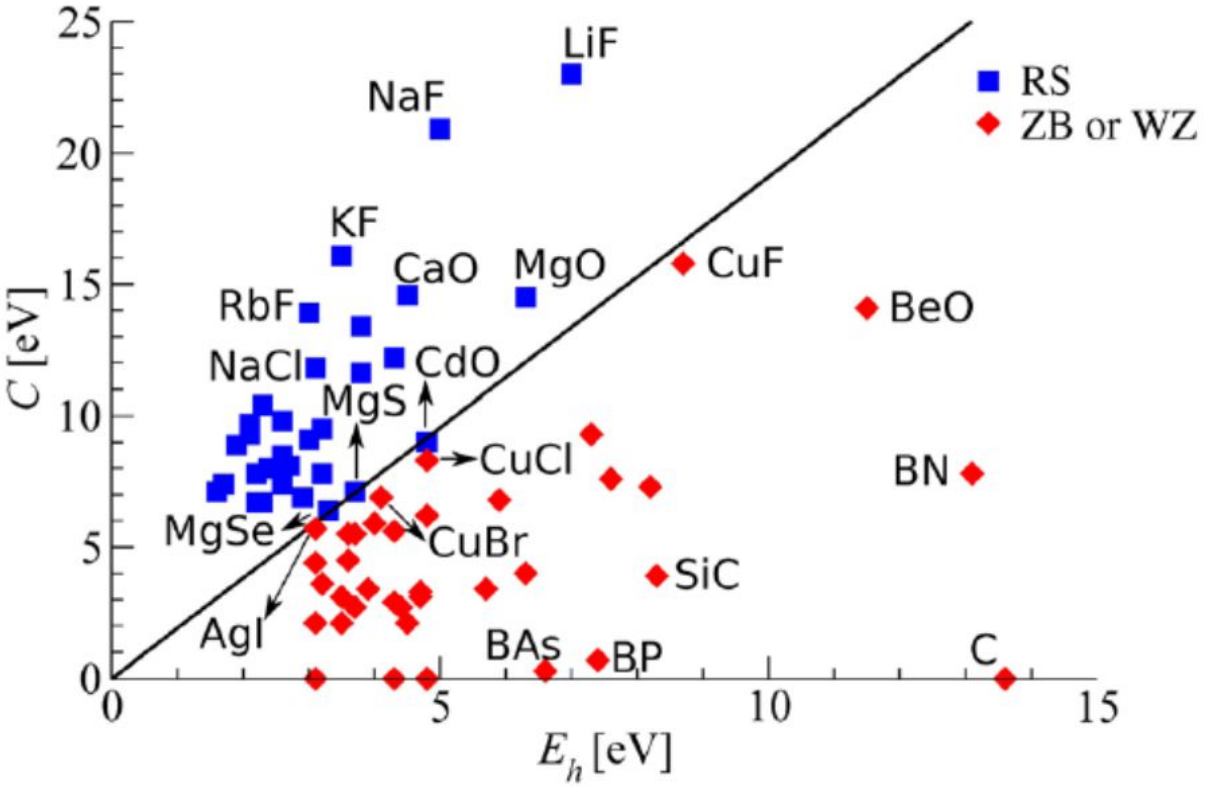


**descriptor can be determined spectroscopically - properties of the solid**

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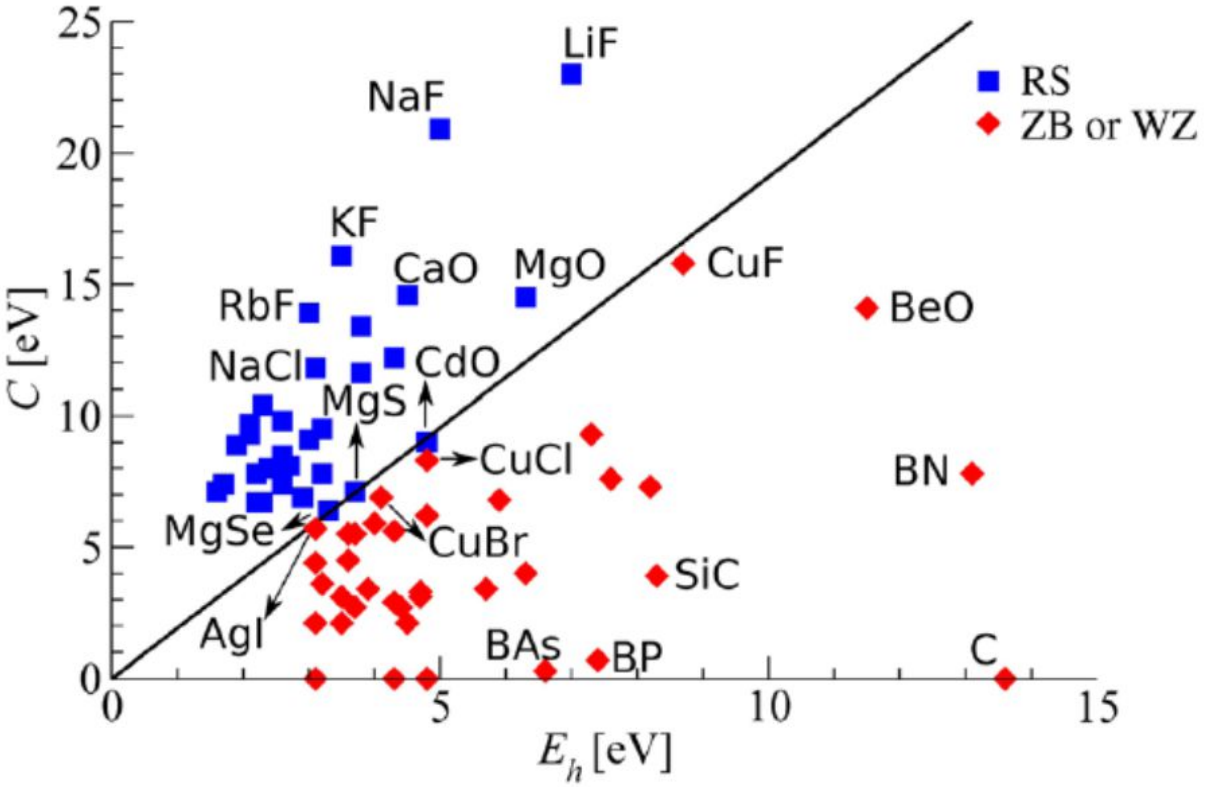



**descriptor can be determined spectroscopically - properties of the solid**

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

# Primary features and feature space

free atoms

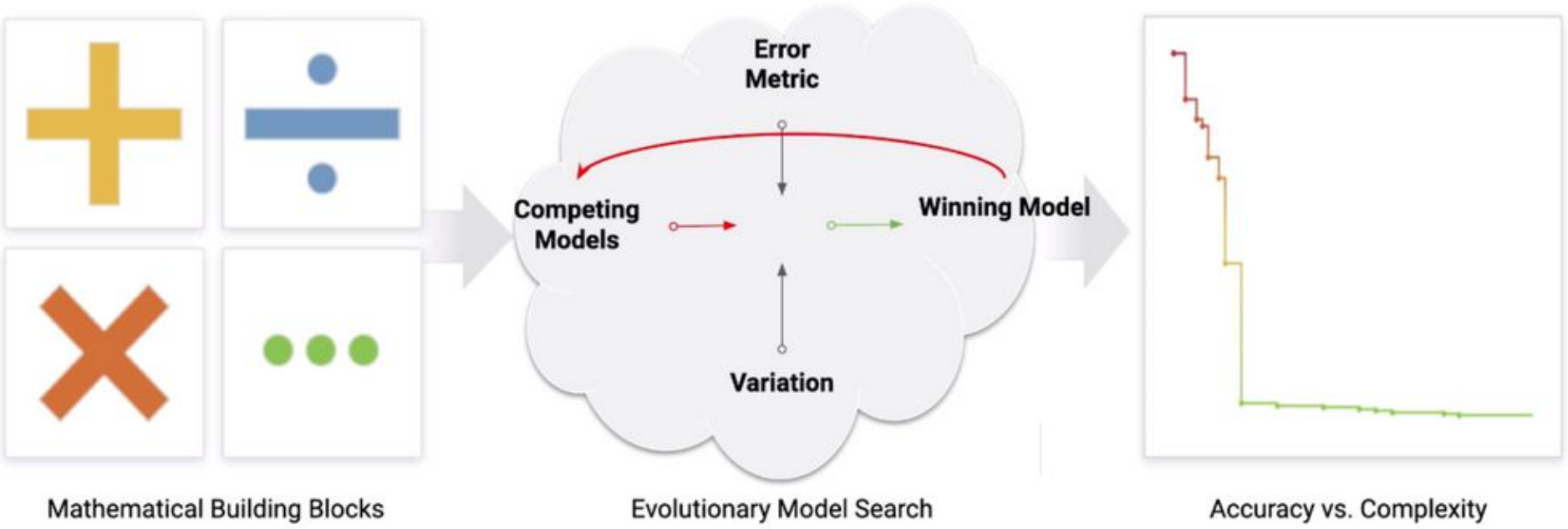
| ID | Description | Symbols | # |
|---|---|---|---|
| $A1$ | Ionization Potential (IP) and Electron Affinity (EA) | IP(A) EA(A) IP(B) EA(B) [1] | 4 |
| $A2$ | Highest occupied (H) and lowest unoccupied (L) Kohn-Sham levels | H(A) L(A) H(B) L(B) | 4 |
| $A3$ | Radius at the max. value of $s$, $p$, and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

| 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)

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

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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)

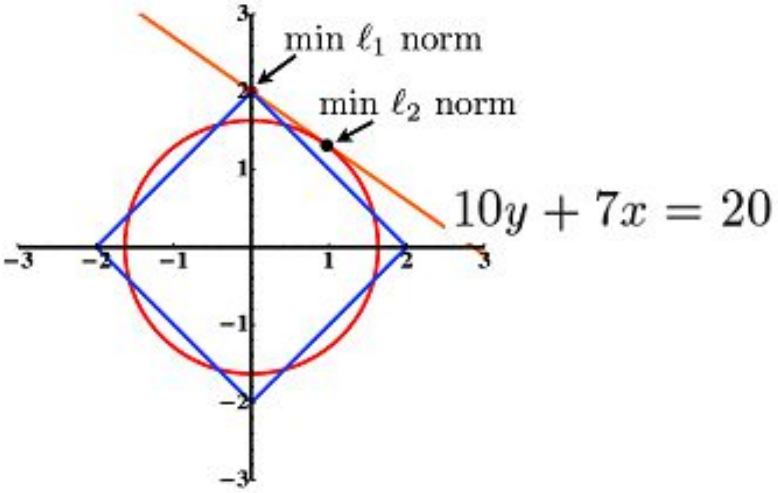
$d_{j,l}$ -- value of feature $l$ related to material $j$ (e.g., $|r_s(A_j) - r_p(B_j)|$) (a basis function in materials space)

$c_l$ -- 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 $c_l$?

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




$\|c\|_1 = \sum_l |c_l|$ -- 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

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

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

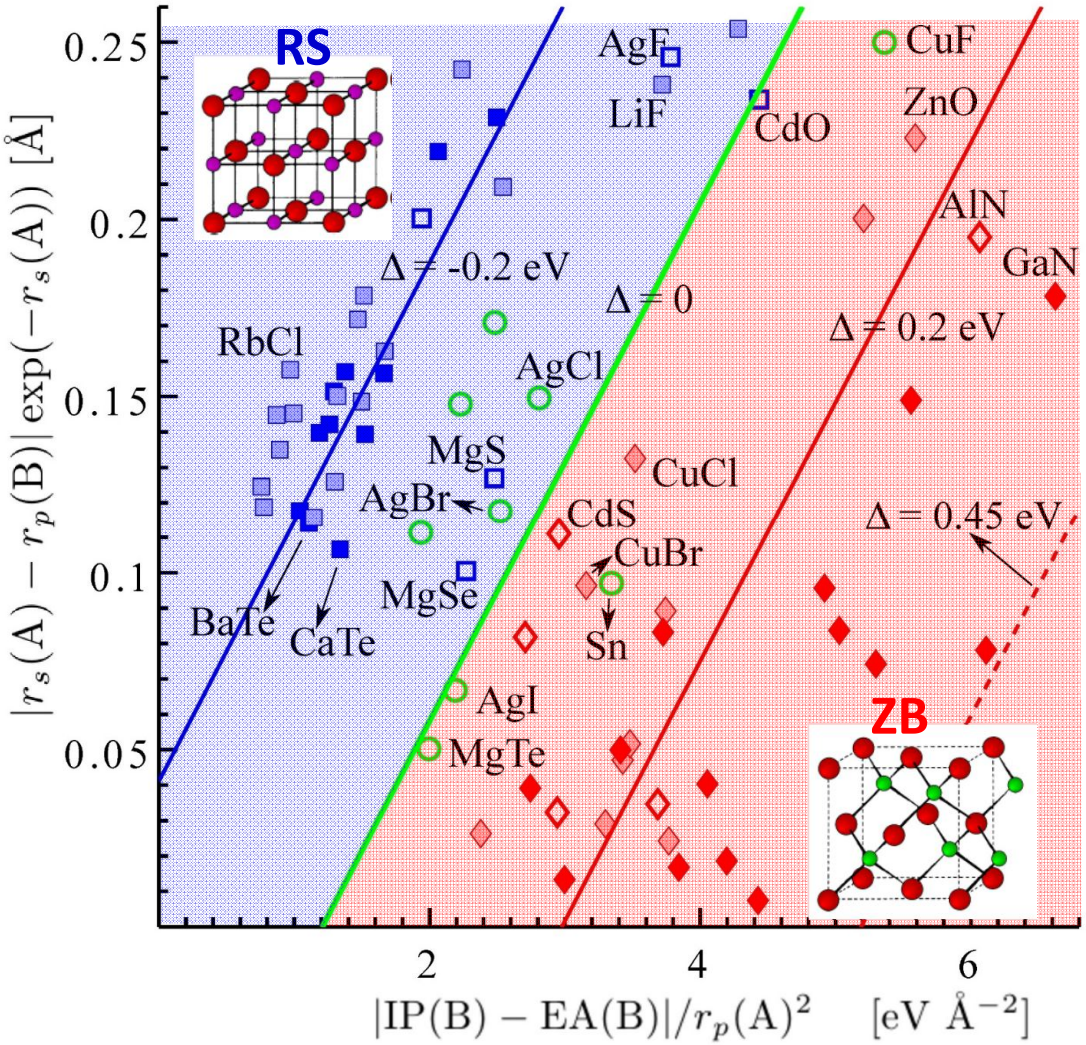
$$\Delta E = 0.113\frac{\mathrm{EA(B)}-\mathrm{IP(B)}}{r_p(\mathrm{A})^2} + 1.542\frac{|r_s(\mathrm{A})-r_p(\mathrm{B})|}{\exp(r_s(\mathrm{A}))} - 0.137 \qquad \text{2D}$$

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

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

**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).

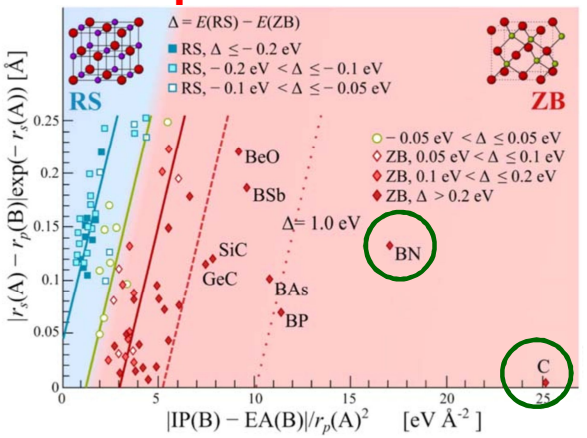
L.M. Ghiringhelli, J. Vybiral, S.V. Levchenko, C. Draxl, and M. Scheffler,
Phys. Rev. Lett. **114**, 105503 (2015).

# 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**

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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**

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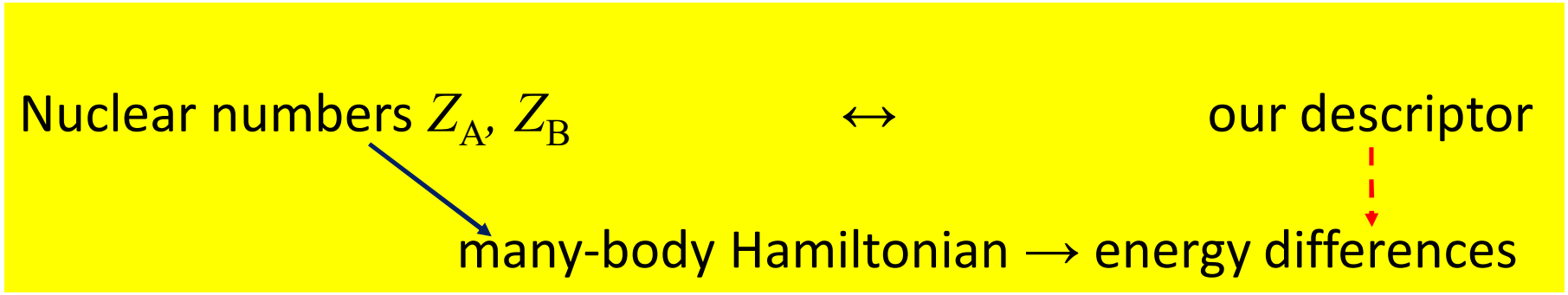
Gaussian-kernel ridge regression

LASSO

# Drawing causal inference from data




a mapping exists, even a physical intuition exists, but $\Delta E$ 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

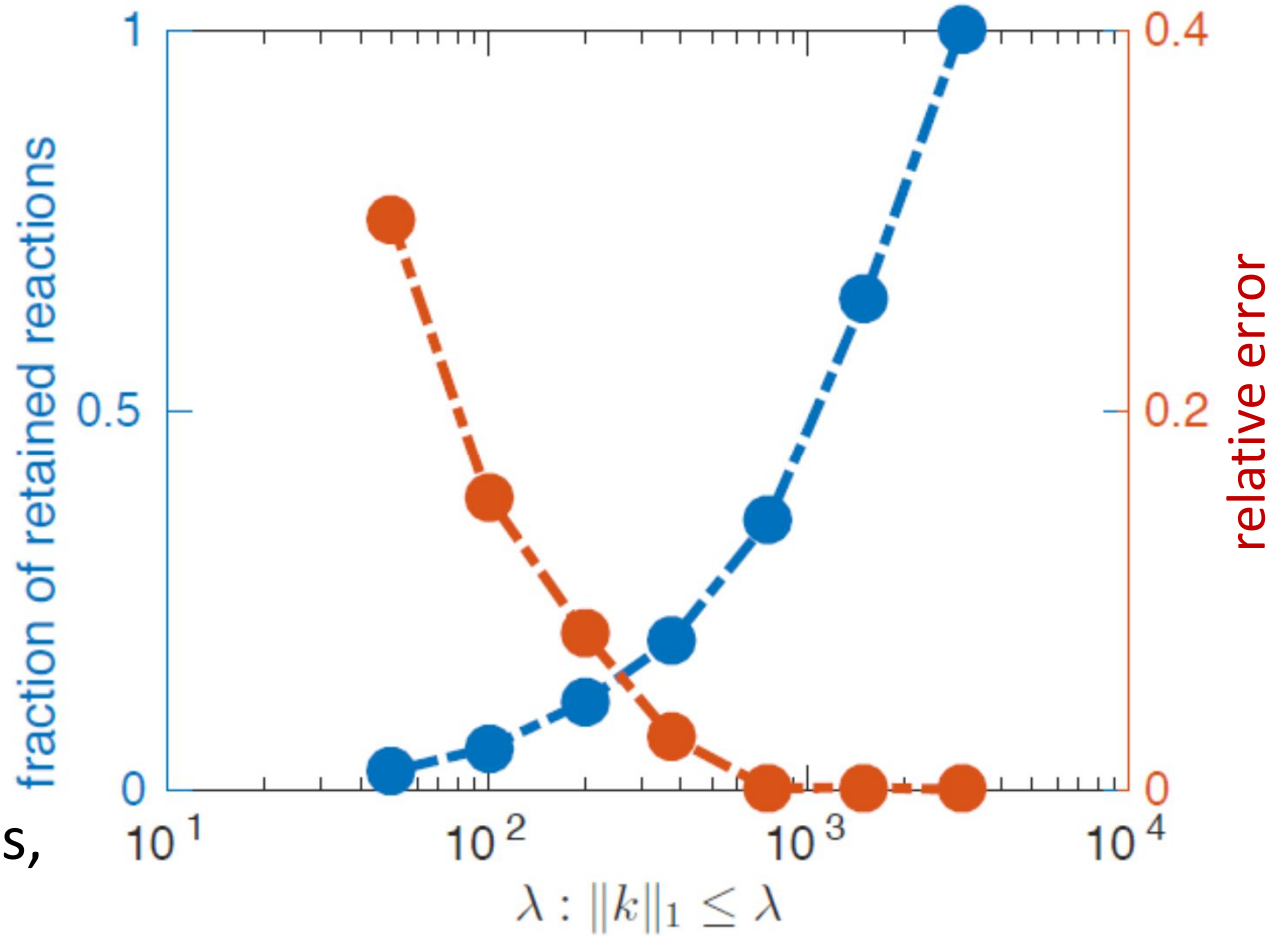
# $CH_4$ chemical decomposition under shock-compression conditions (high *T* and *p*)

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)

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

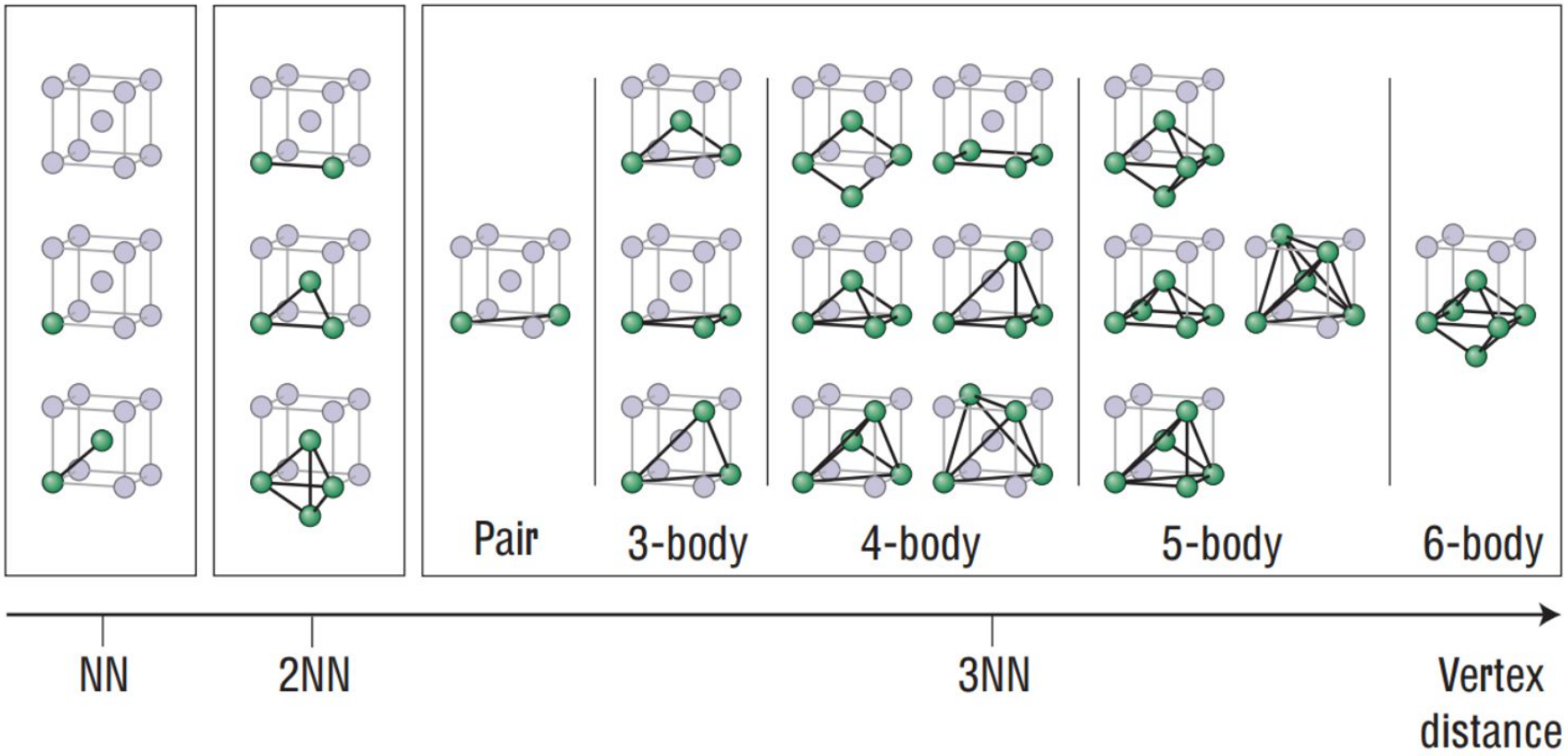
$$A_{aJ} = \begin{bmatrix} -1 & u_b^1 & -\frac{1}{2}u_b^1u_c^1 & \cdots \\ \vdots & \vdots & \vdots & \\ -1 & u_b^L & -\frac{1}{2}u_b^Lu_c^L & \cdots \end{bmatrix}$$

→ 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)**

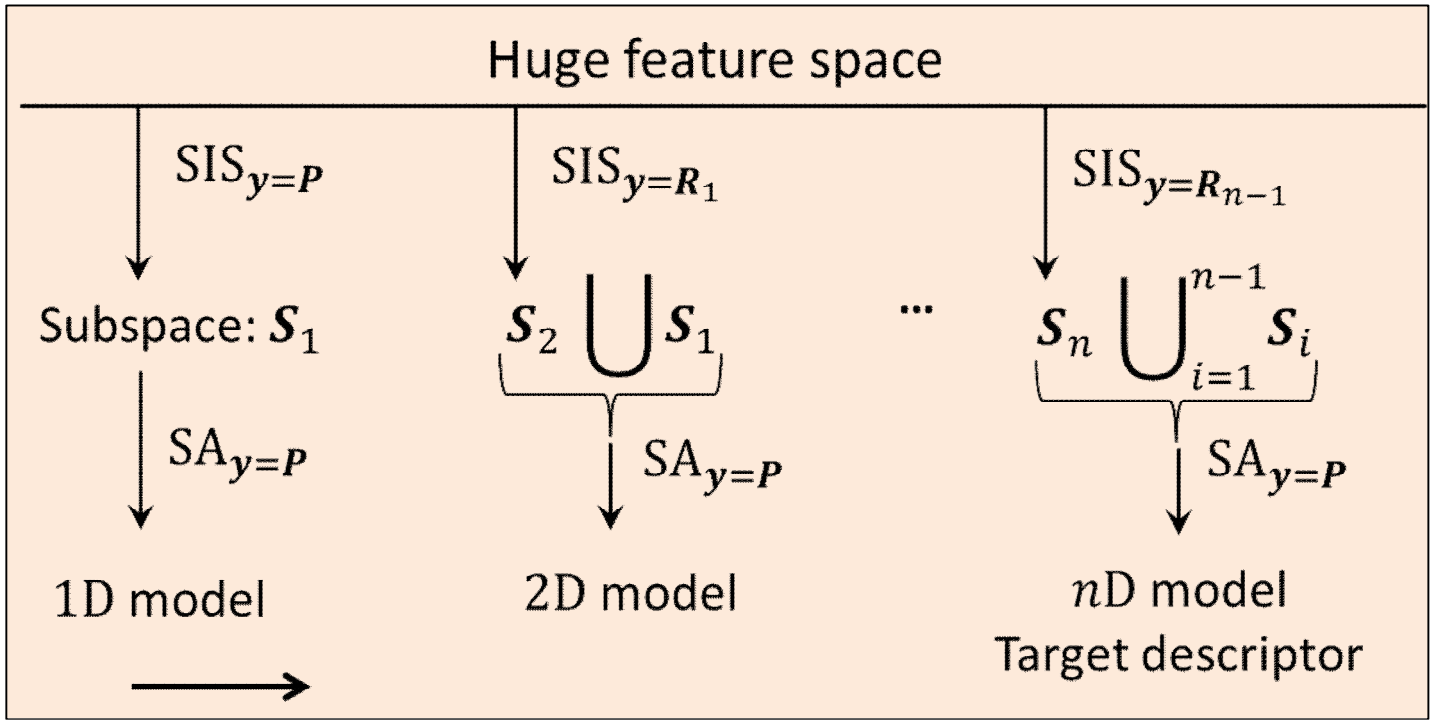
1. Systematically construct a huge feature space ($10^{11}$) from primary features: $\hat{R} = \{+, -, \cdot, ^{-1}, ^2, ^3, \sqrt{\ }, exp, log, |-|\}$(use physically meaningful combinations!)
2. Select top ranked features using *Sure Independence Screening (SIS)*[1] (correlation learning). Select $n$ 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)

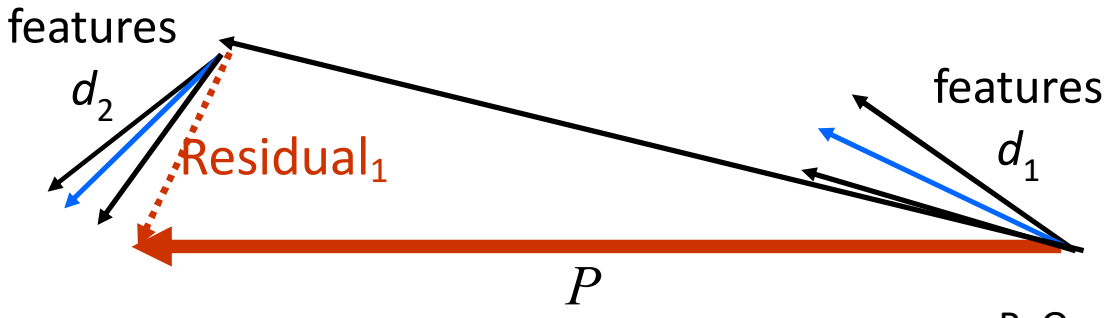
# SISSO: Iterative residual fitting




$\boldsymbol{y}$: response vector

$\boldsymbol{P}$: target material property

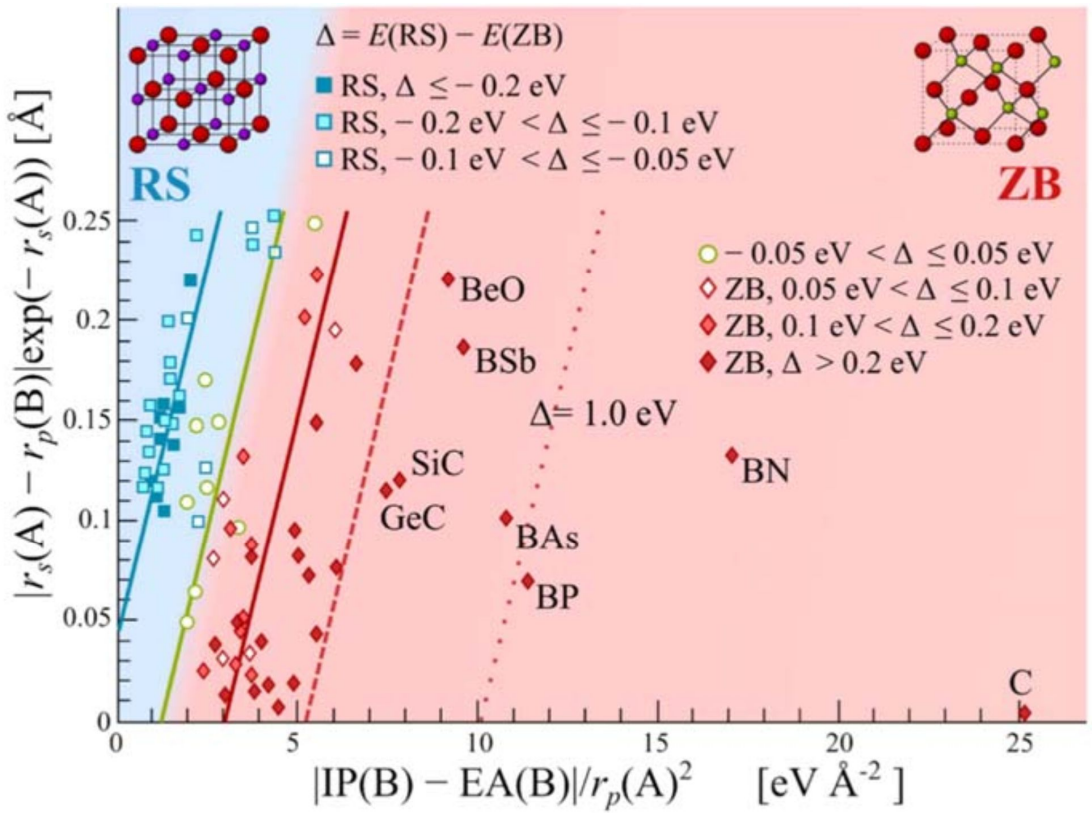
Residual: $R = P - \sum_i c_i d_i$



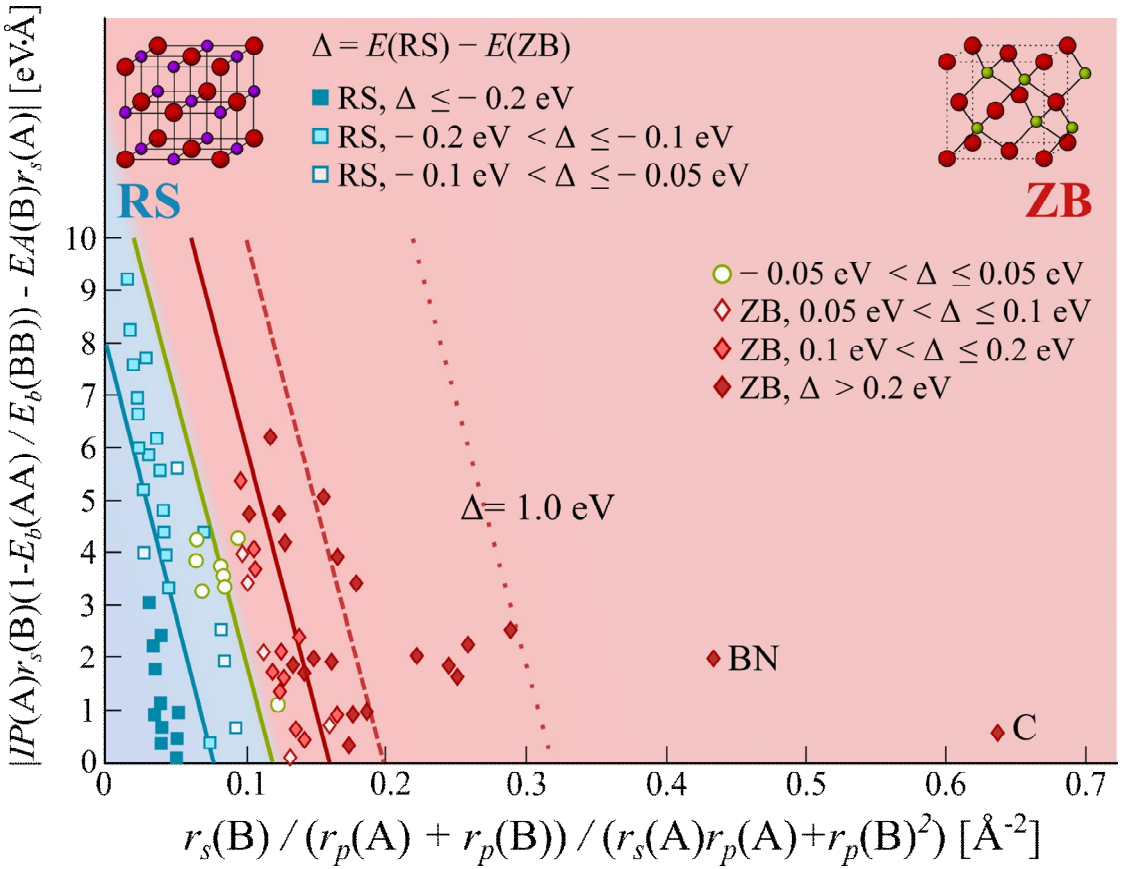

R. Ouyang, *et al.*, Physical Review Materials 2, 083802 (2018)

# SISSO: Performance

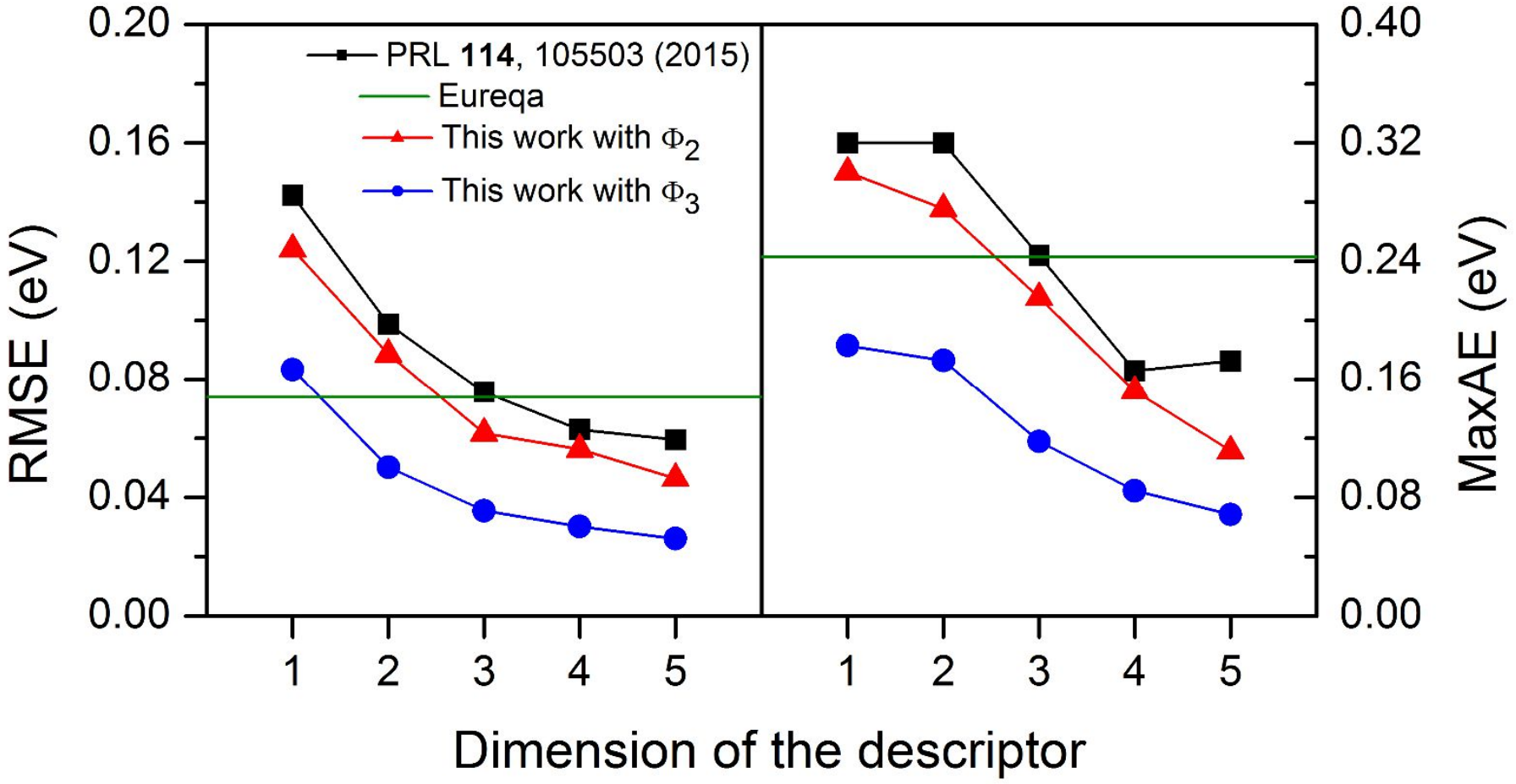
## LASSO(+$l_0$)




## SISSO



# SISSO: Performance



# SISSO: Multitask and categorical

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

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

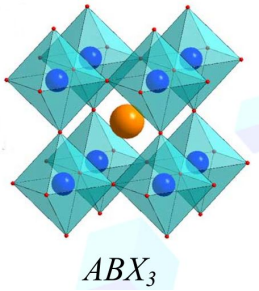
**Categorical (can be also multitask): Property - material belongs to a given class (yes/no)**

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

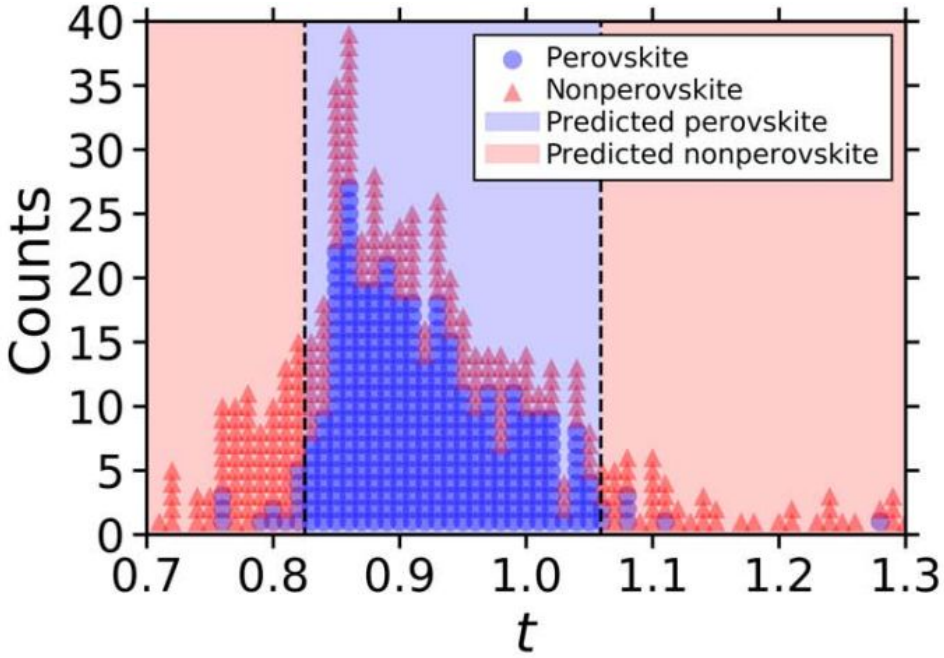
**number of data in the overlap region between domains of different classes in $d$-space**

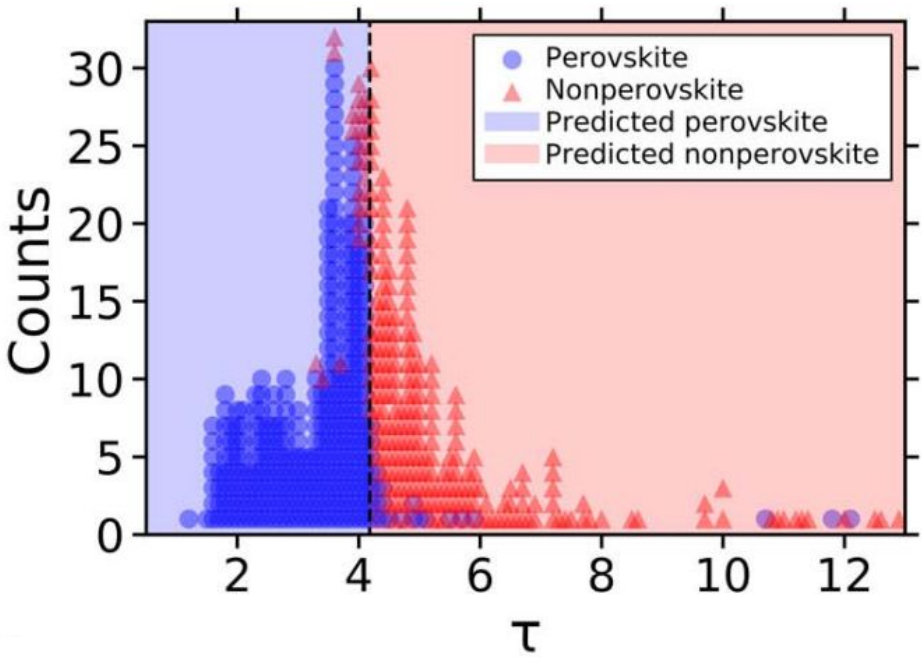
R. Ouyang, *et al*., J. Phys.: Mater. 2, 024002 (2019)

# 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%**

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

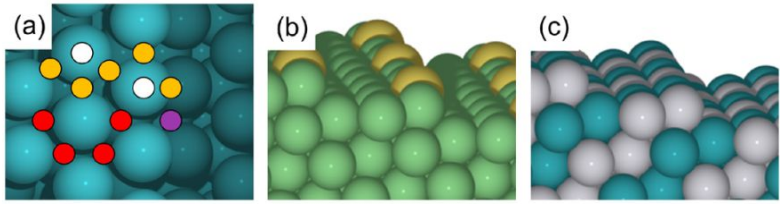
oxidation state

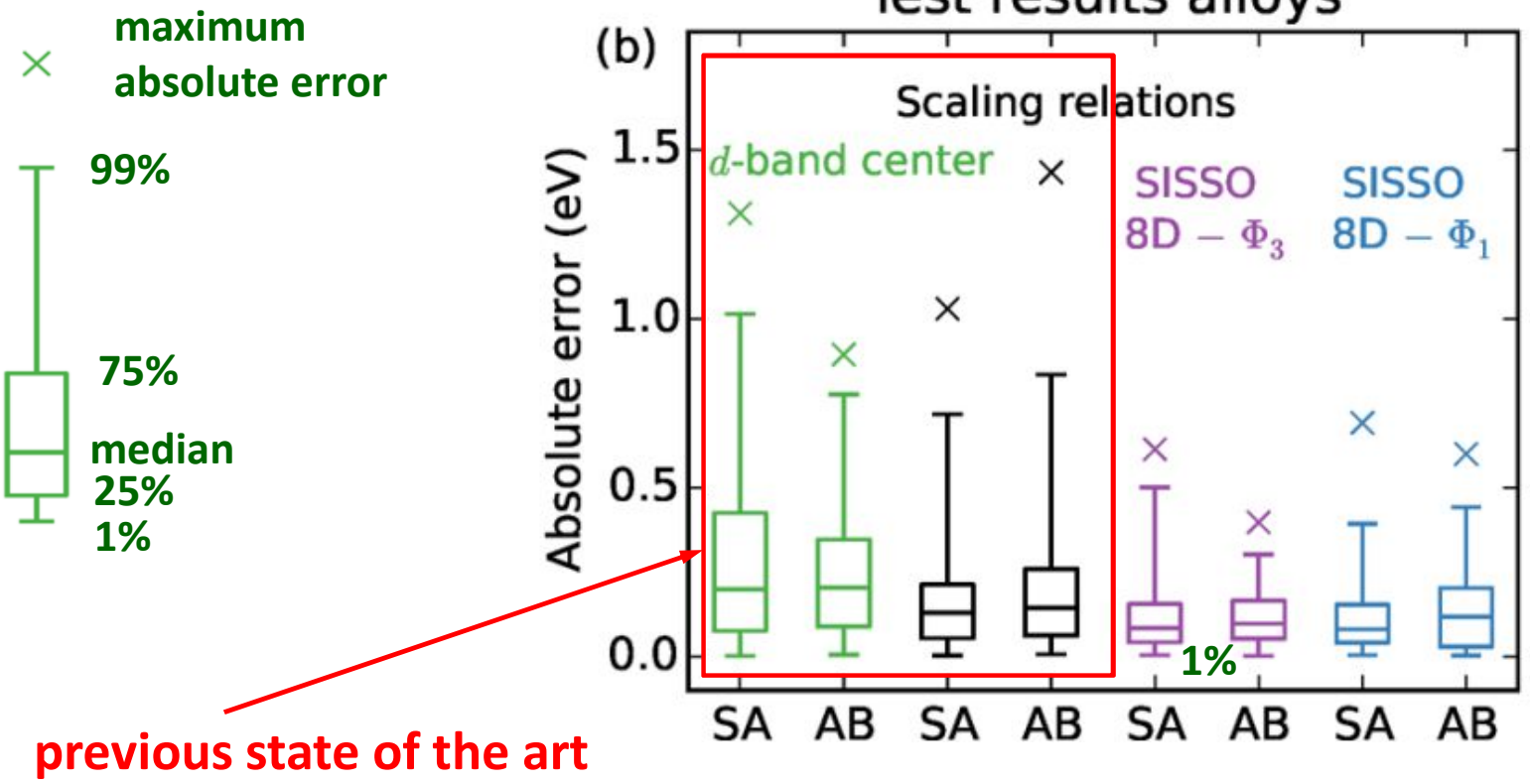
C. Bartel *et al.*, Sci. Adv. *5, eaav0693* (2019)

# SISSO: Examples

- Adsorption of molecules on metal surfaces

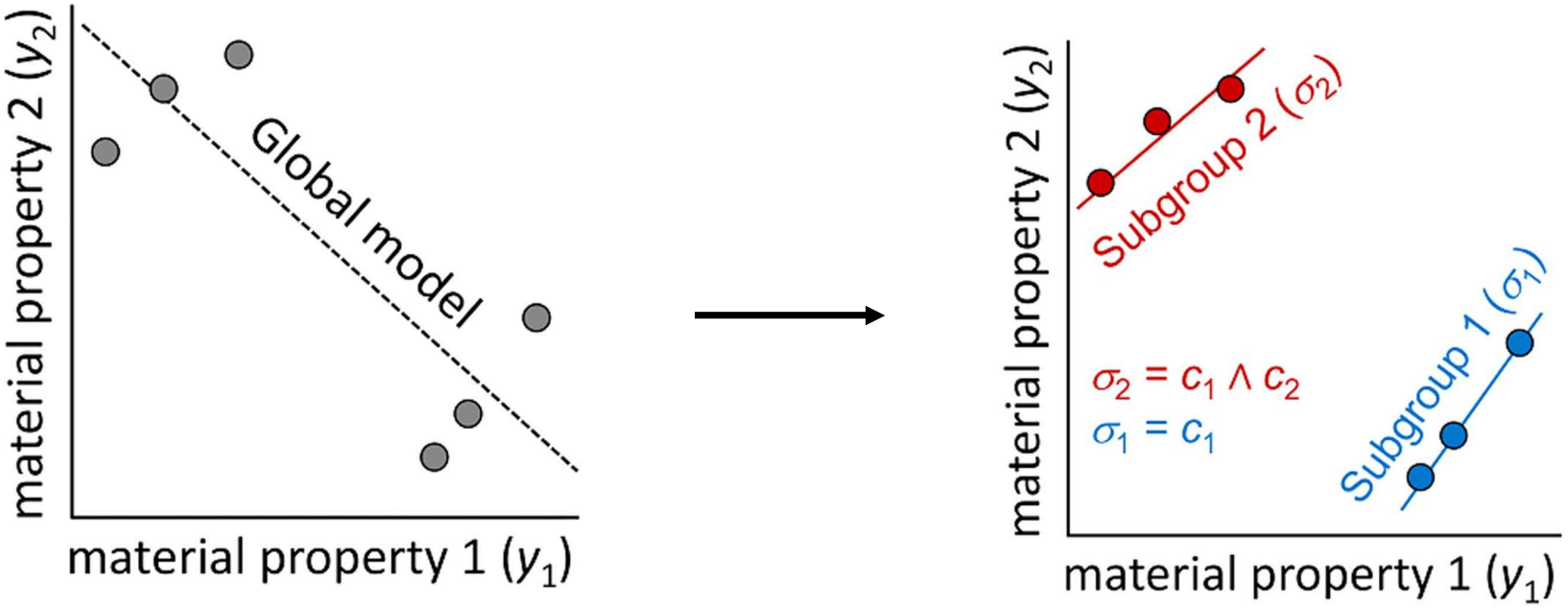
Adsorption of C, CH, CO, H, O, OH)




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

# Data mining: Subgroup discovery




**Subgroups are defined by selectors $\sigma$ 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***

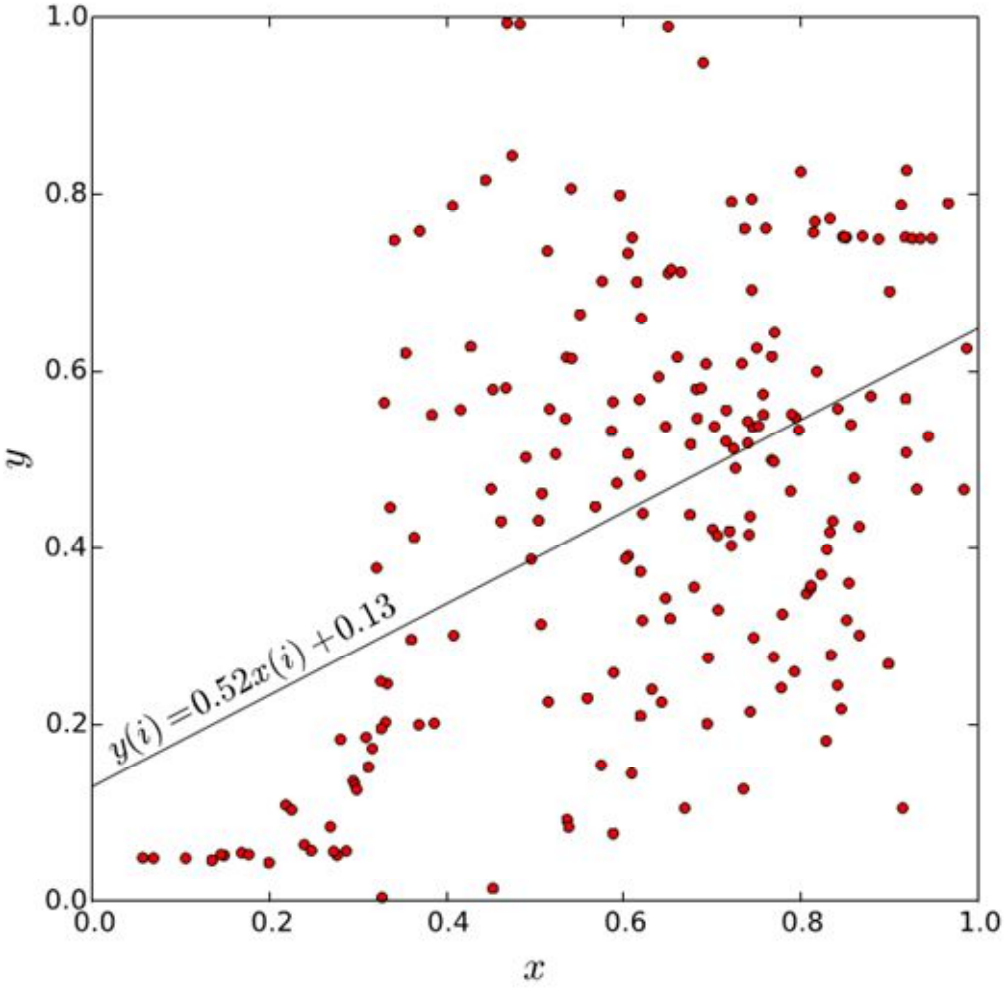
$$f = N_{subgroup}/N_{all} \times |mean_{subgroup} - mean_{all}| \times (1 - variance_{subgroup}/variance_{all})$$

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

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

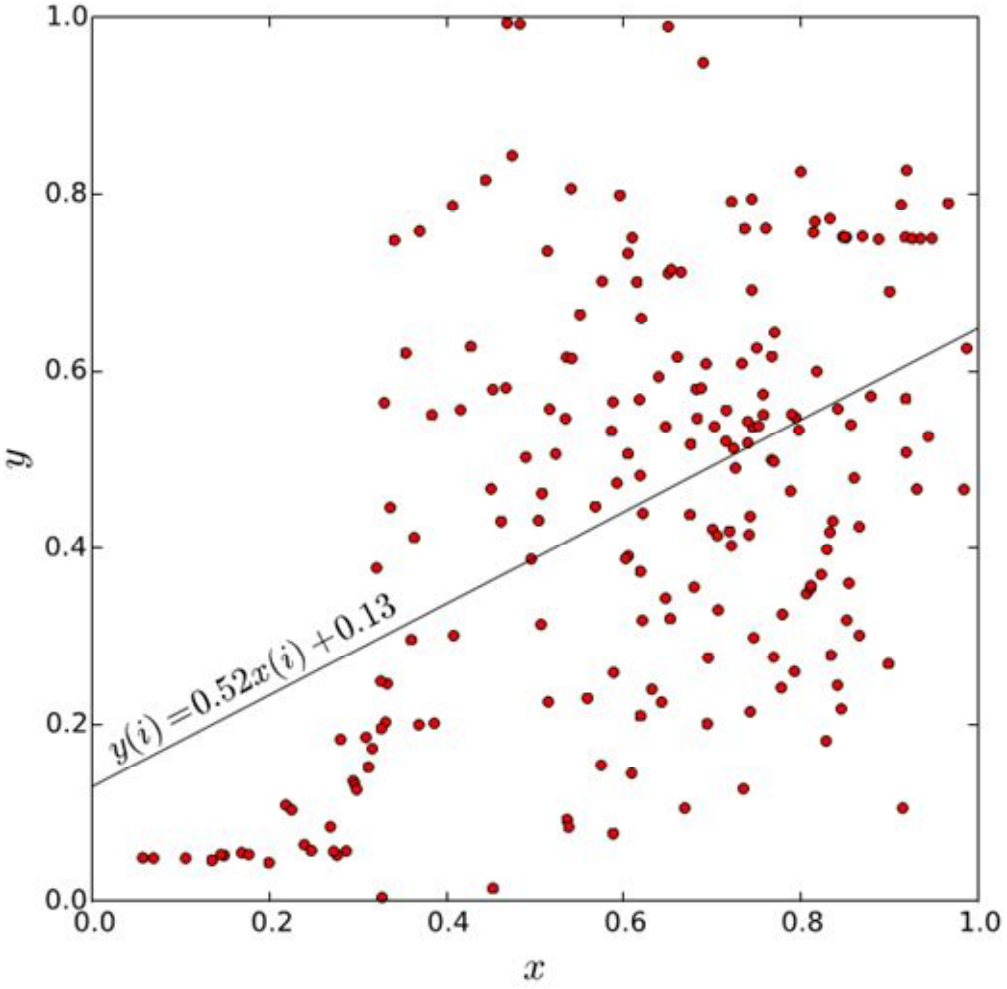
W. Klösgen, Advances in Knowledge Discovery and Data Mining. Palo Alto, CA: AAAI Press; 1996, 249

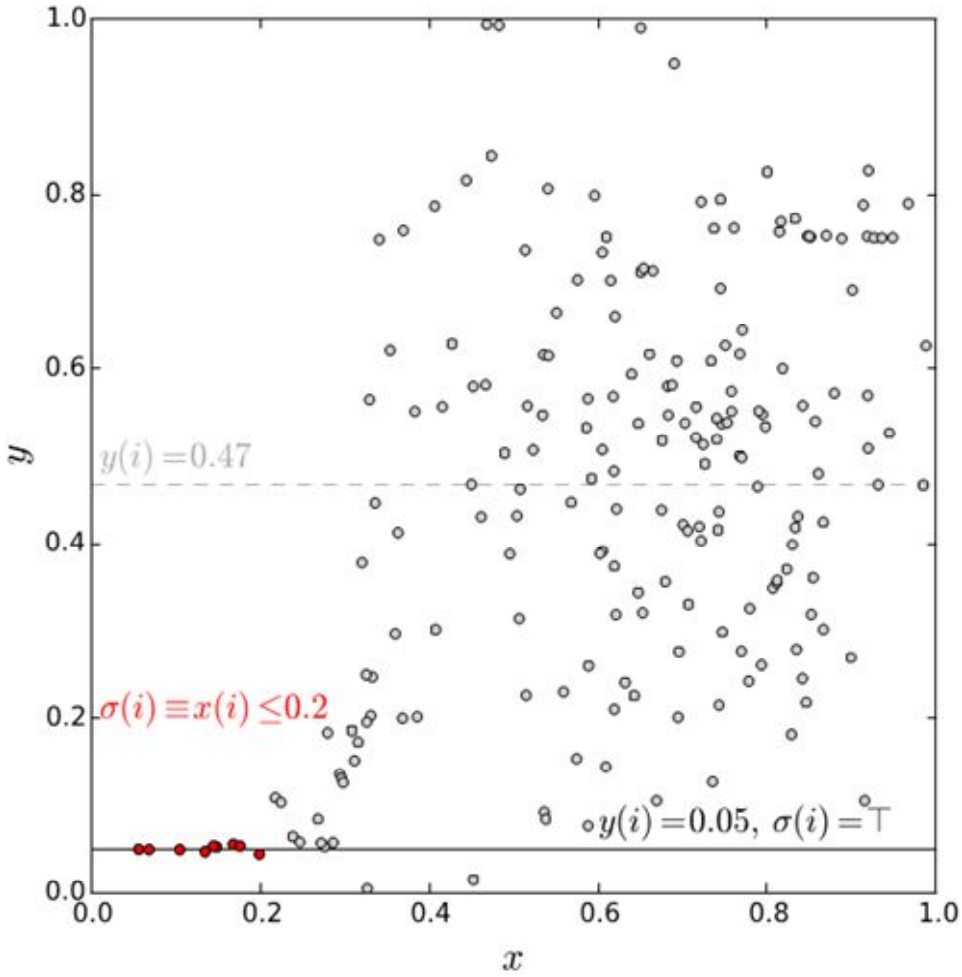
# Data mining: Subgroup discovery




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

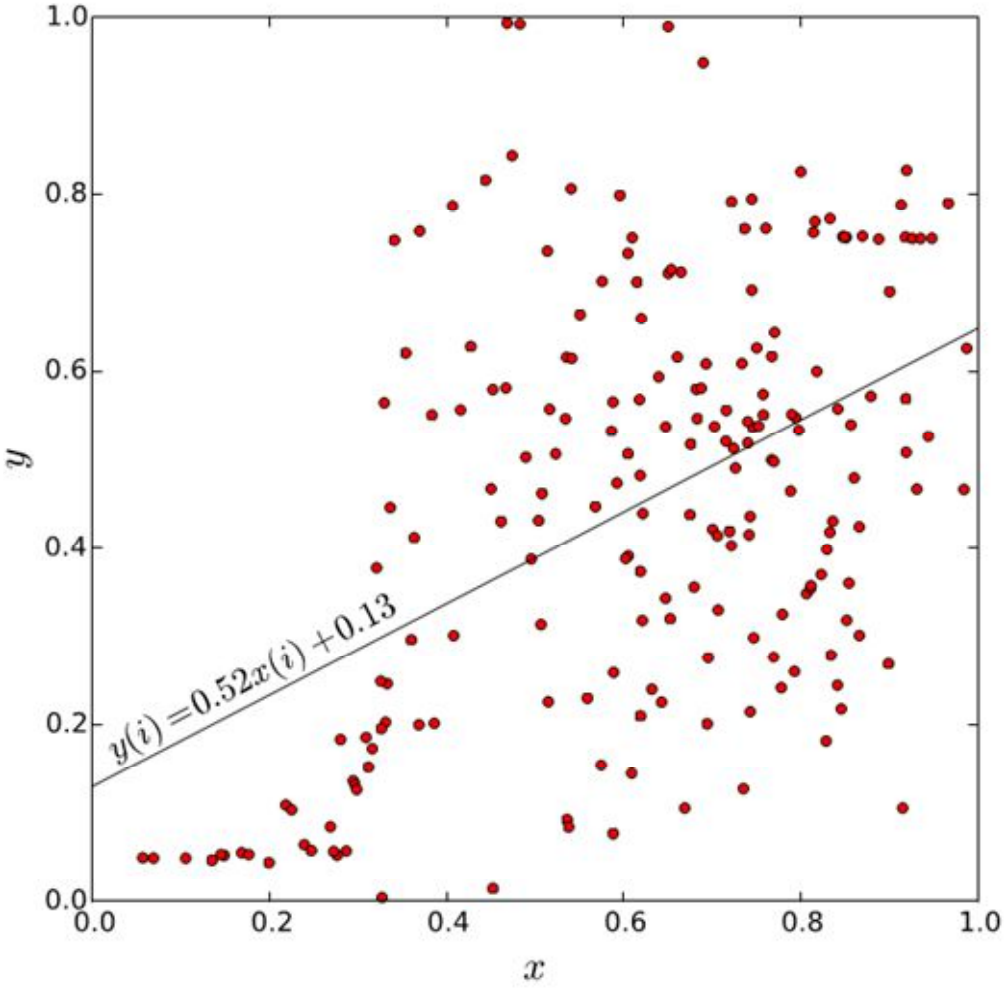
# Data mining: Subgroup discovery

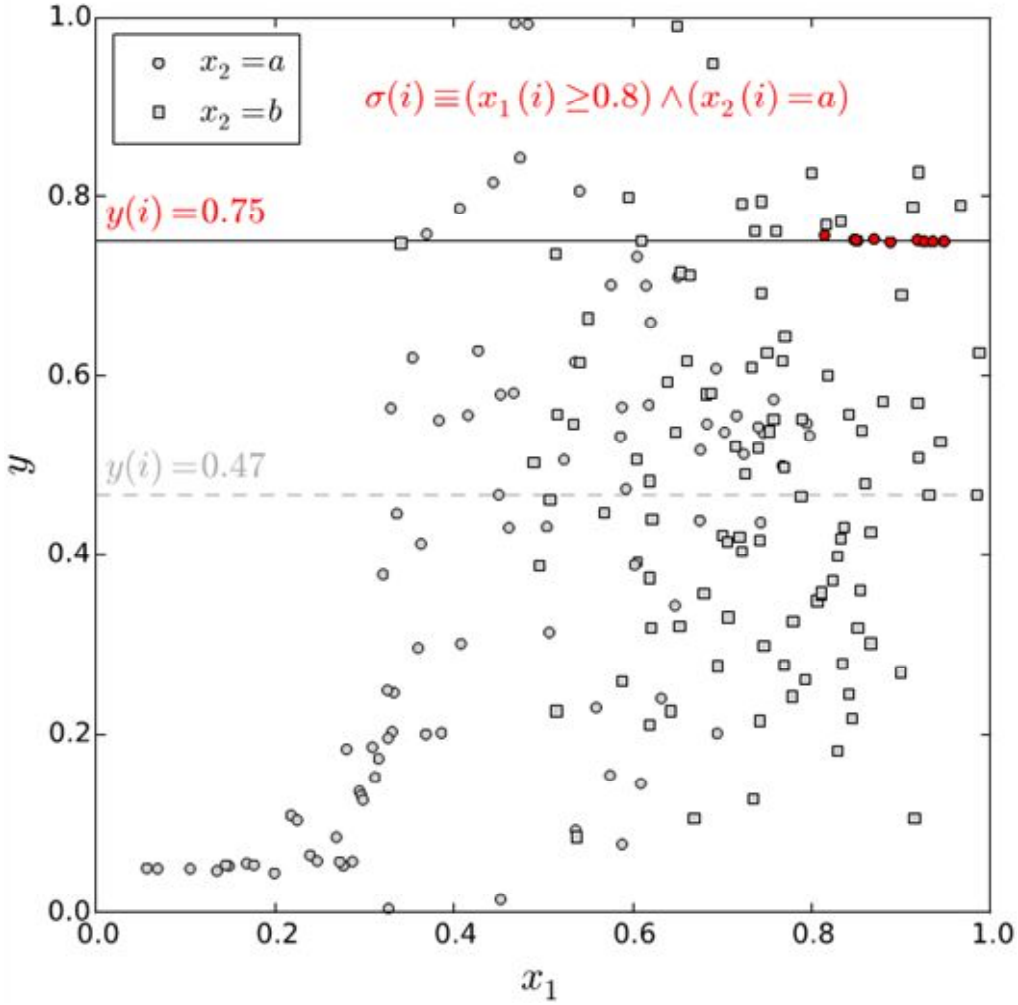



M. Boley *et al*., Data Min. Knowl. Disc. 31, 1391 (2017); B. Goldsmith *et al*., New J. Phys. 19, 013031 (2017)

# Data mining: Subgroup discovery



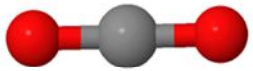



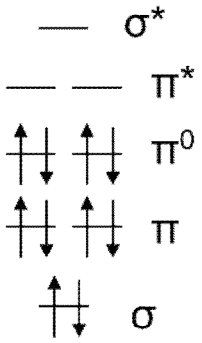

M. Boley *et al*., Data Min. Knowl. Disc. 31, 1391 (2017); B. Goldsmith *et al*., New J. Phys. 19, 013031 (2017)

# Subgroup discovery: $CO_2$ activation by adsorption

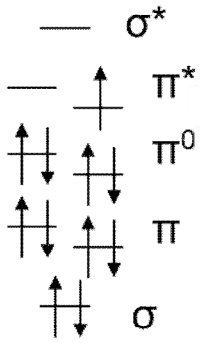
$CO_2$

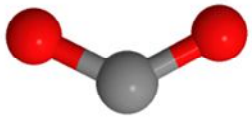
C-O bond elongation, O-C-O bending angle → indicators of activation



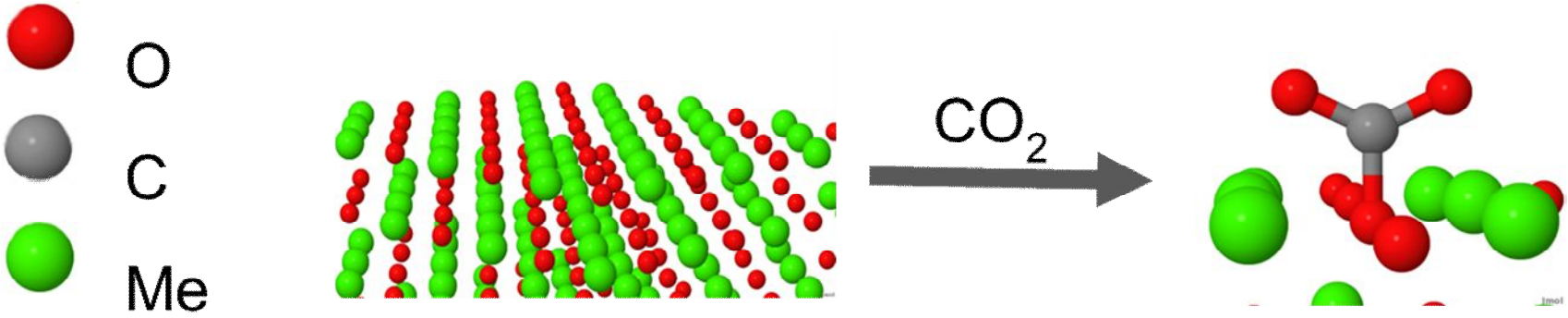



charge transfer



# Subgroup discovery: $CO_2$ activation by adsorption




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

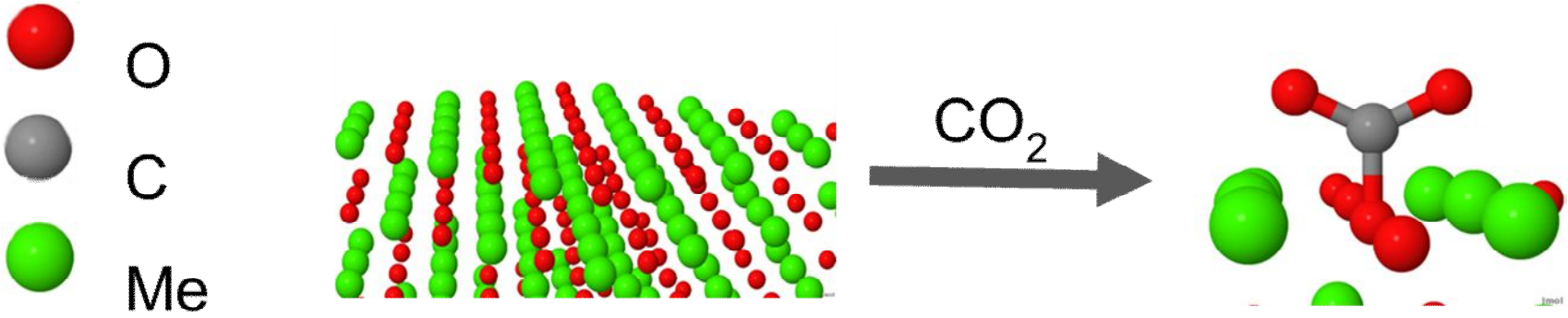
Sabatier reaction:
$CO_2 + 4H_2 = CH_4 + 2H_2O$

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?**

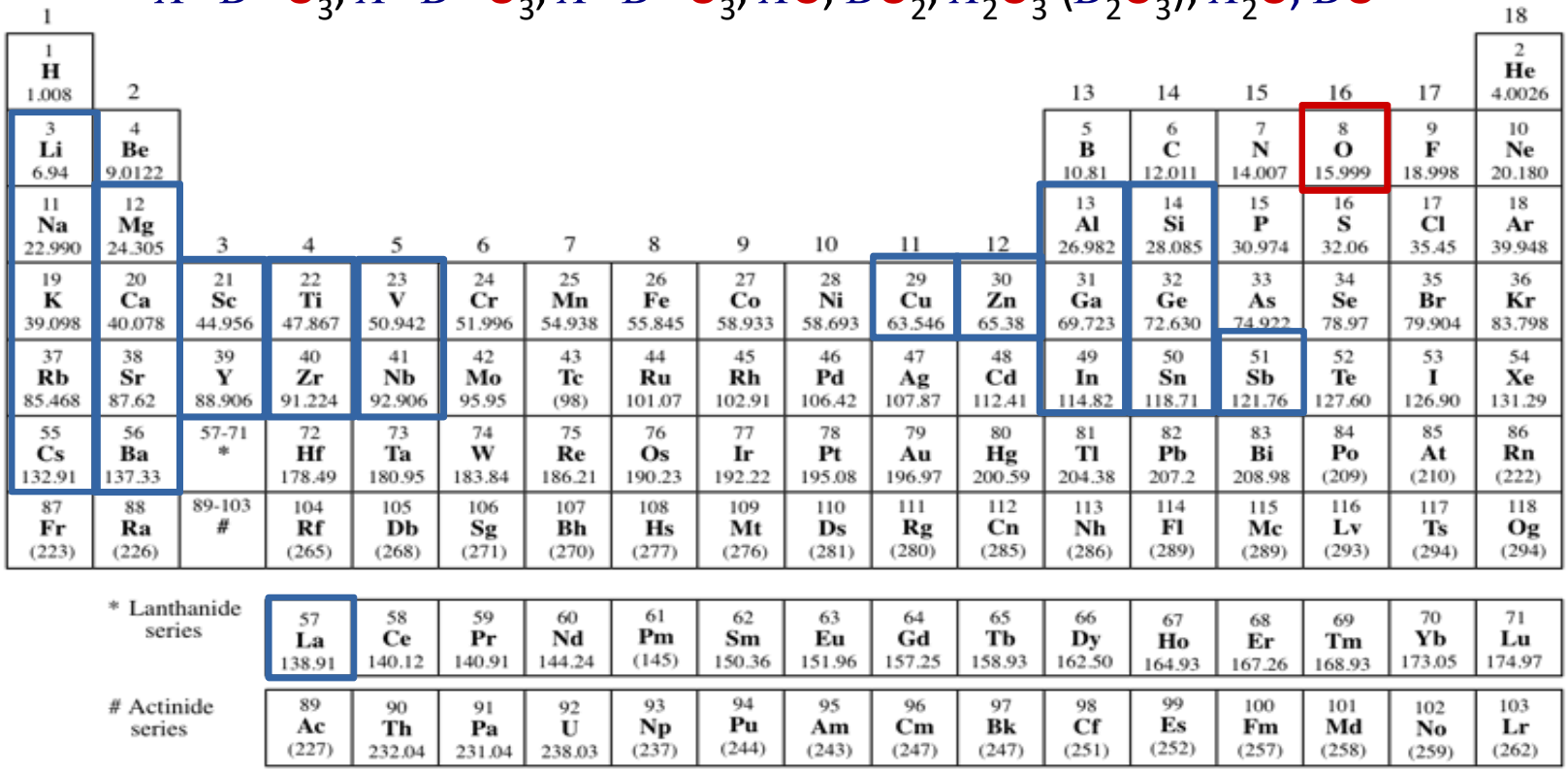
**Use subgroup discovery to find materials that optimize activation indicators**

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

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

# Subgroup discovery: $CO_2$ activation by adsorption

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

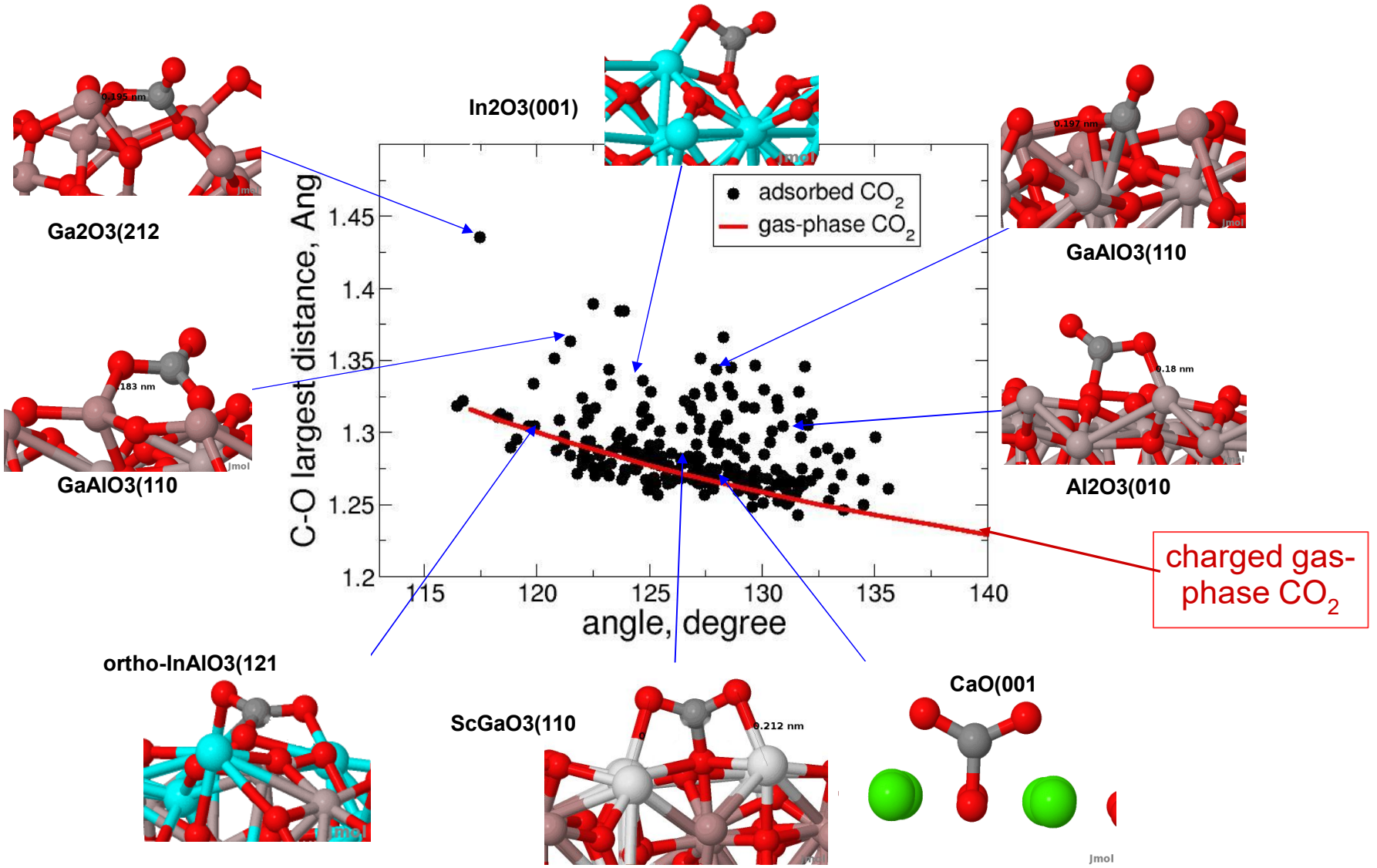



**71** oxide materials

**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

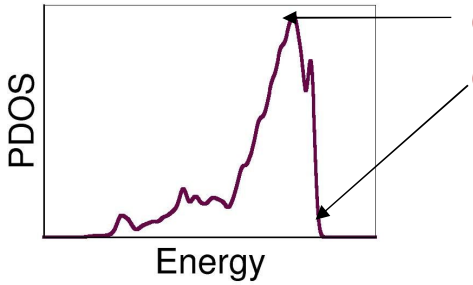
**Site-specific features:**

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

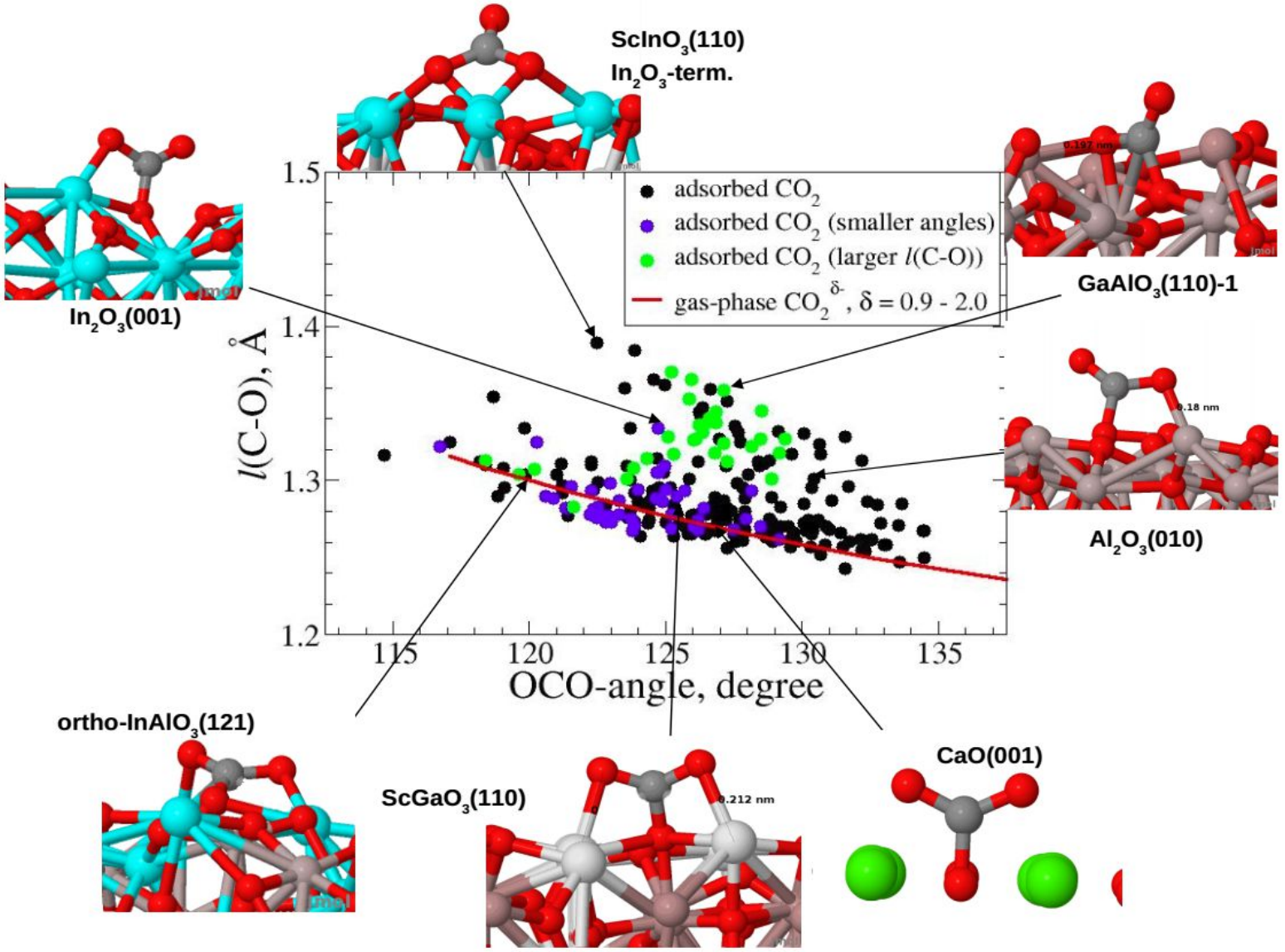
features of
O 2*p*-PDOS



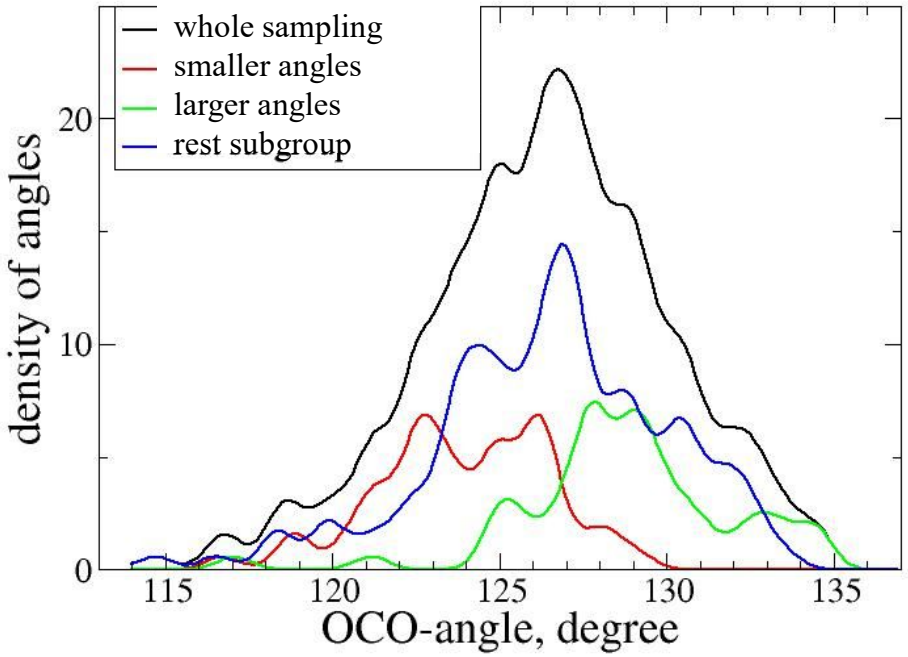

energy of maximum
energy of top

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

# Subgroup discovery: Adsorbed CO2 properties

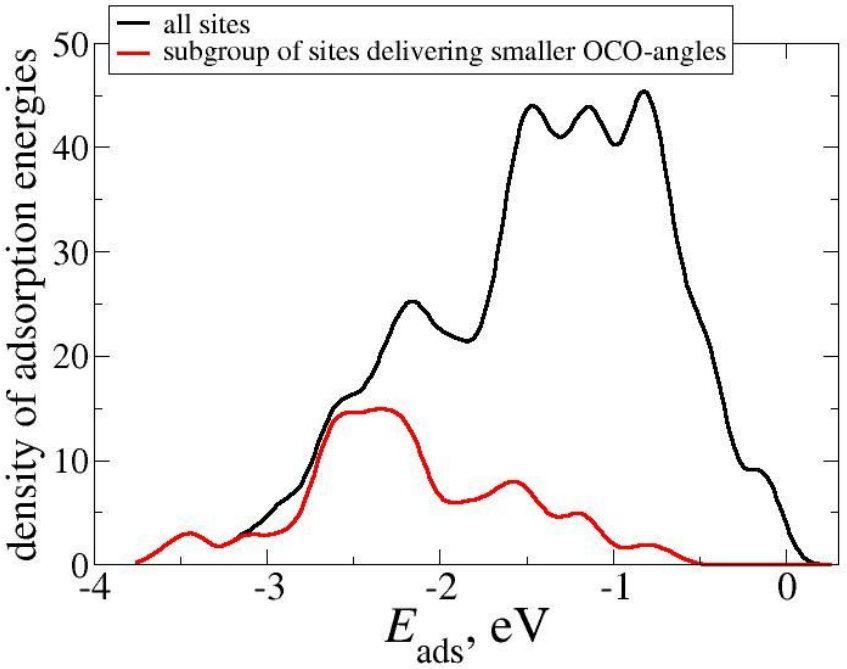


# Subgroup discovery: Analysis of the OCO angle



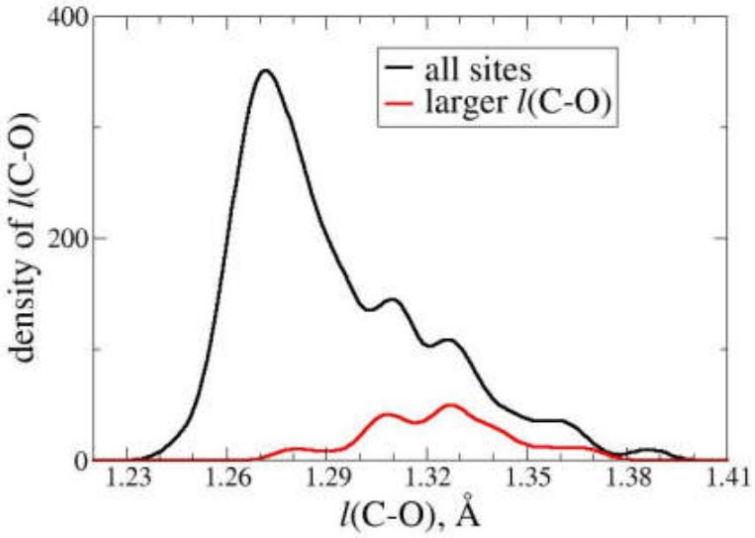

**sites delivering smaller angles (59 adsorption sites):**

**(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)**



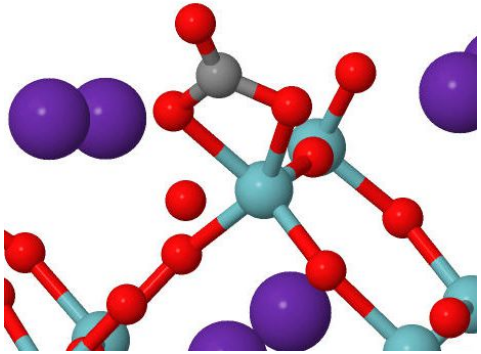

**Most of the site delivering smaller OCO angles are on ionic (basic) materials**

# 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)**



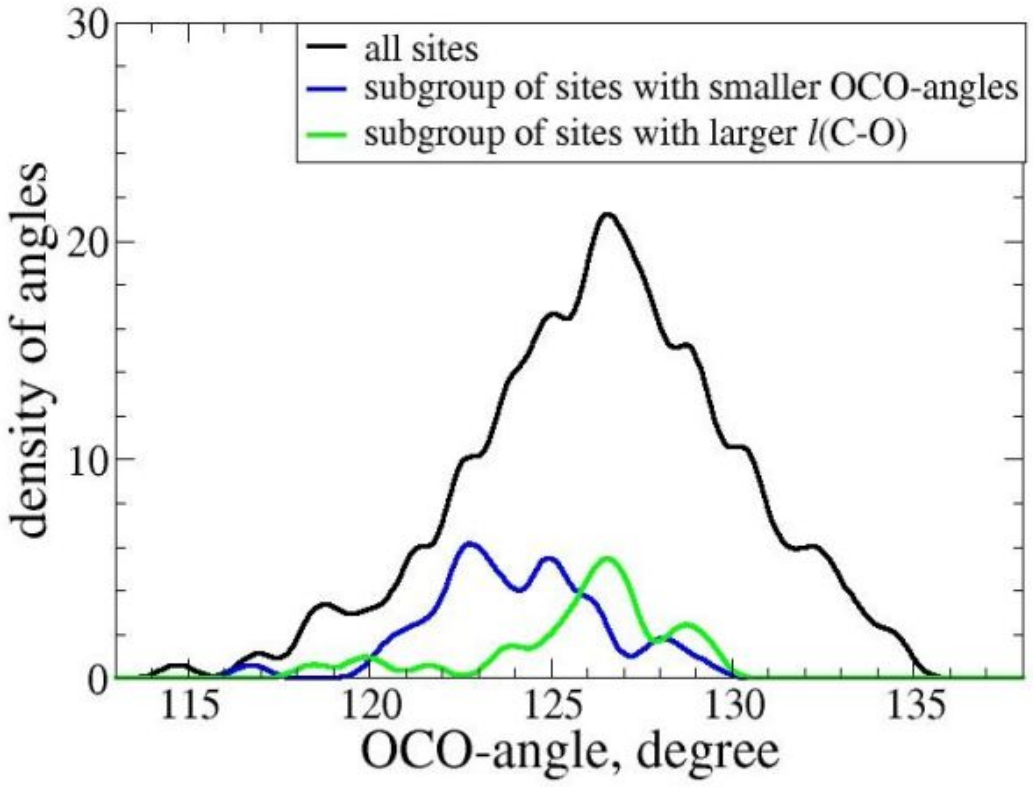
$LaGaO_3$ – cathode material in high-temperature electrochemical $CO_2$ reduction;

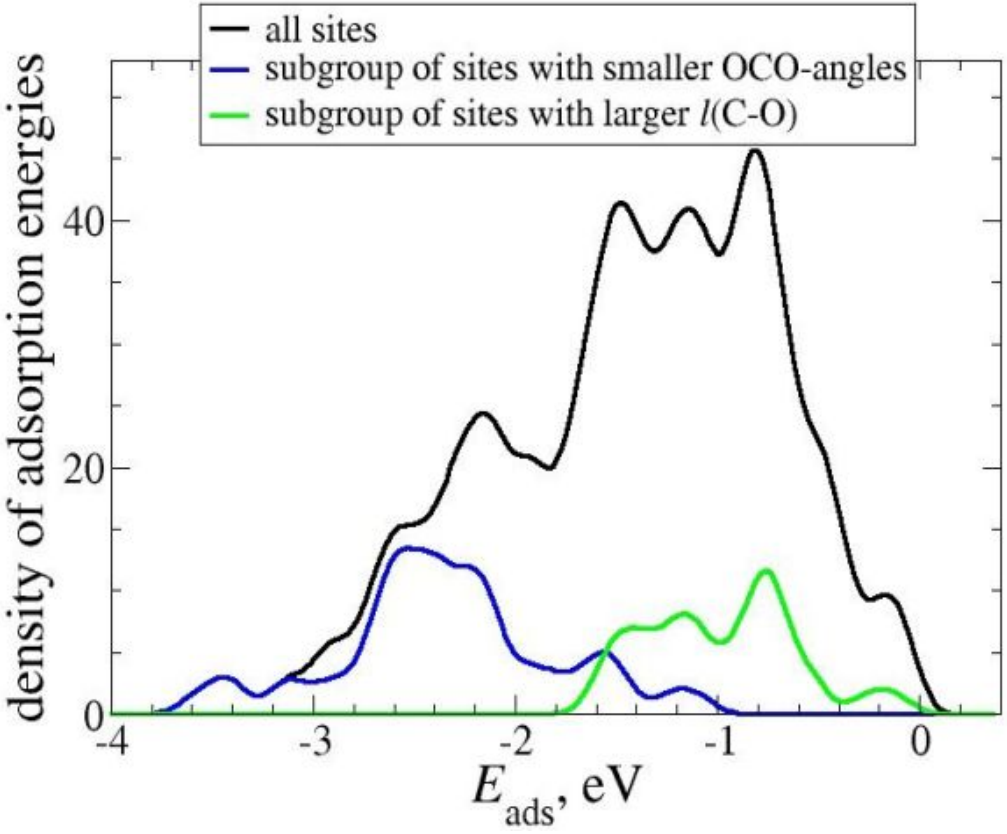
$KNbO_3$ – photocatalytic reduction of $CO_2$ into $CH_4$;

$NaNbO_3$ – 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



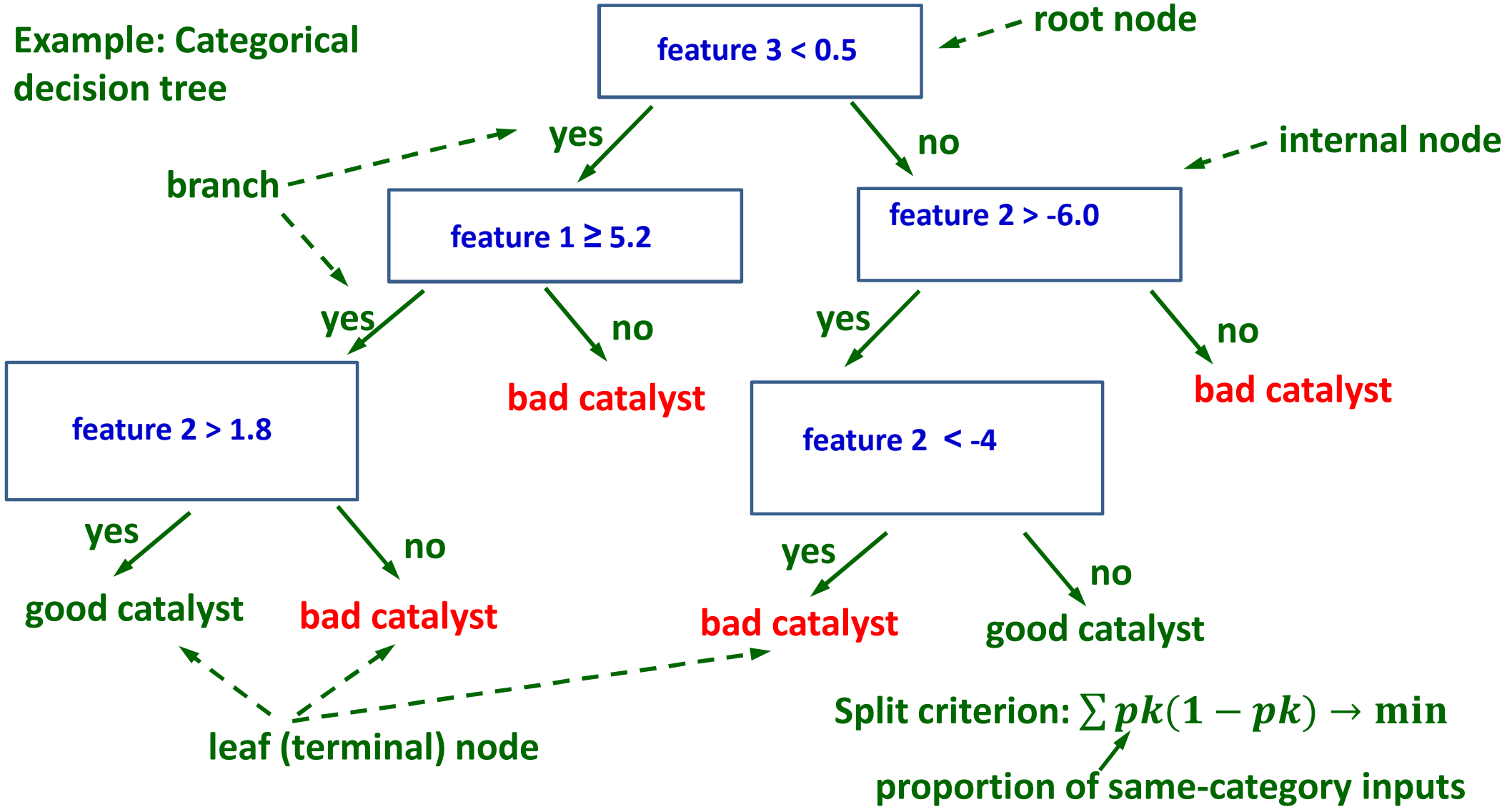

**Longer C-O implies smaller OCO angles, but not too small → no catalyst poisoning**

# SISSO and SGD software

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

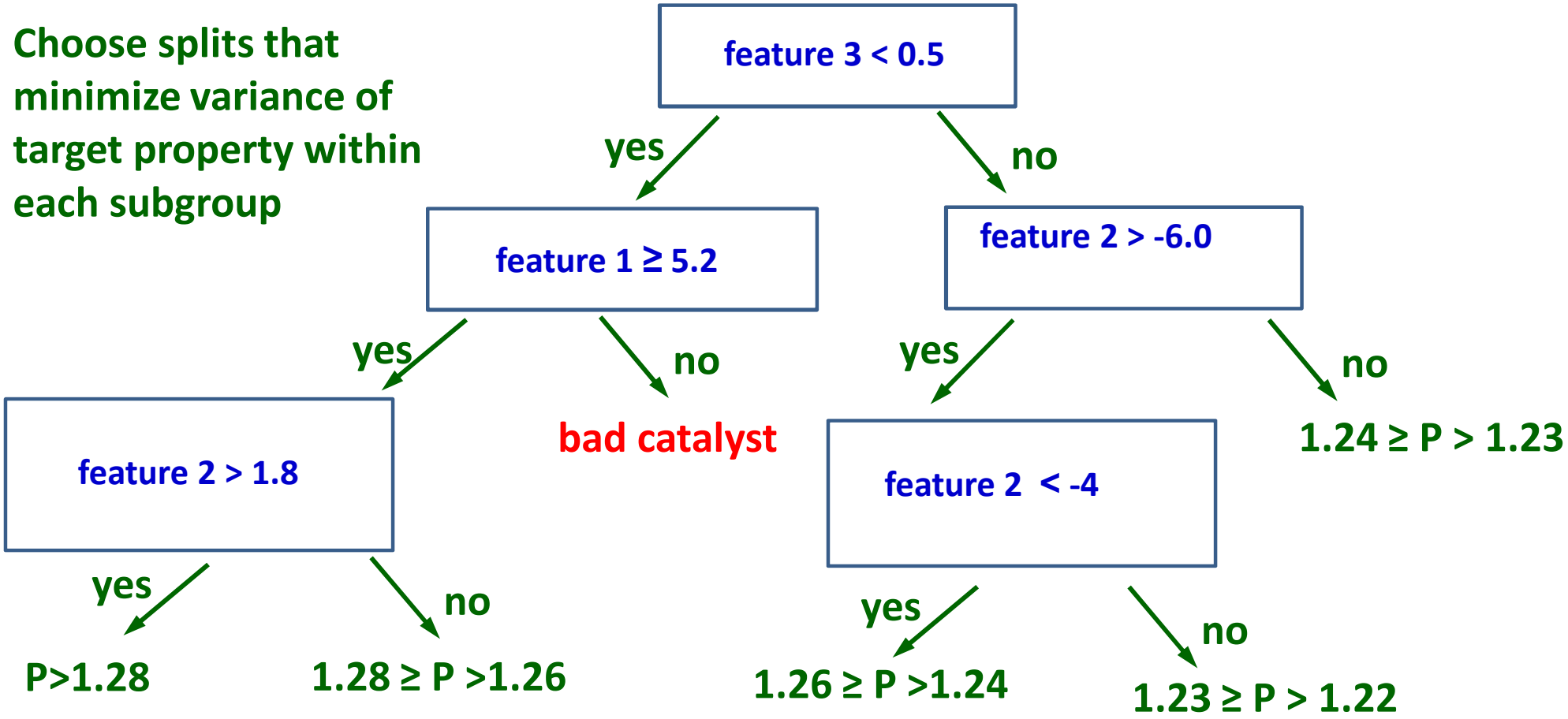
**Subgroup discovery:** https://bitbucket.org/realKD/creedo/wiki/Home

# 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**

THE JOURNAL OF PHYSICAL CHEMISTRY C

Article

Cite This: J. Phys. Chem. C 2019, 123, 2321−2328

pubs.acs.org/JPCC

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

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

JACS

JOURNAL OF THE AMERICAN CHEMICAL SOCIETY

Article

Cite This: J. Am. Chem. Soc. 2018, 140, 4678−4683

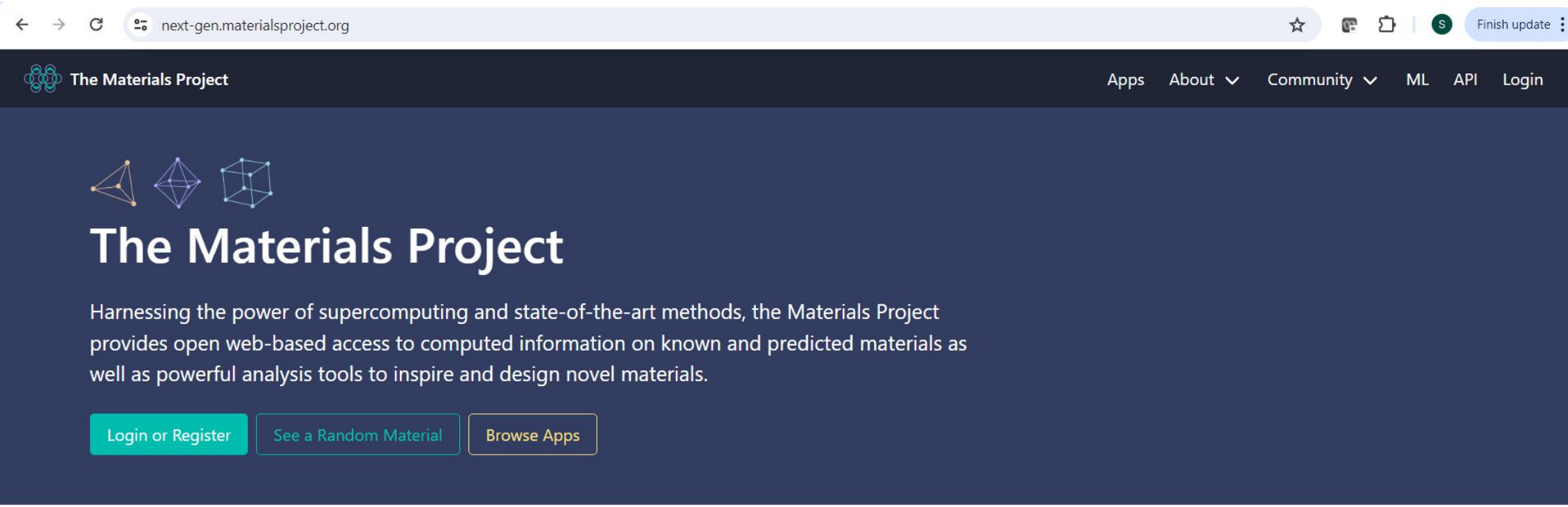
pubs.acs.org/JACS

## 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*,[†]

# Computational databases

**General idea: Create infrastructure for storing, querying, and analyzing computational materials science data**




**Leaders: Kristin Persson (Lawrence Berkeley National Laboratory), Gerbrand Ceder (University of California at Berkeley)**

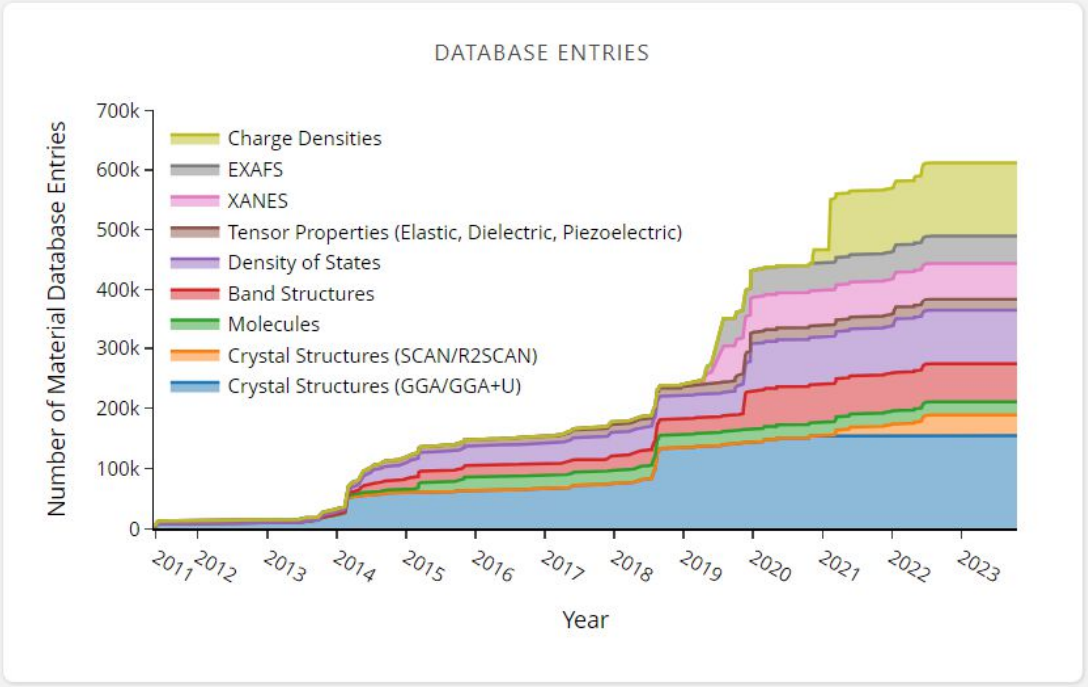
**Structures are mostly from ICSD database (https://icsd.products.fiz-karlsruhe.de/)**

**Motto: We are in full control of the calculations and data. You can contribute, but first discuss with us**

# Materials Project: Features

## The Materials Project by the numbers

| | |
|---|---|
| MATERIALS<br>154,718 | REGISTERED USERS<br>460,000+ |
| INTERCALATION ELECTRODES<br>4,351 | CITATIONS<br>42,000+ |
| MOLECULES<br>172,874 | CPU HOURS/YEAR<br>100 million |




**All calculations are performed with GGA or GGA+*U***

**Typical data: relaxed crystal structure, band structure, DOS, energy from the convex hull, elastic properties, X-ray absorption and diffraction spectra, piezoelectric tensors,**

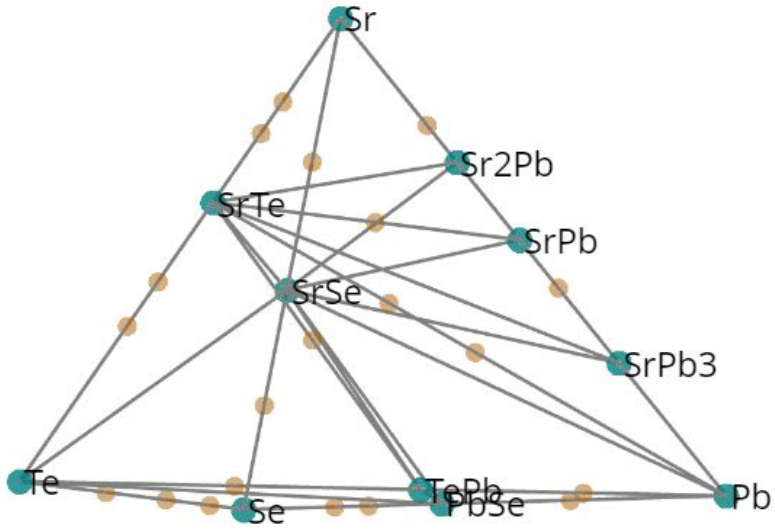
oqmd.org

# OQMD

The Open Quantum Materials Database

Home Materials Analysis Documentation API Download

The OQMD is a database of DFT calculated thermodynamic and structural properties of **1,226,781** materials, created in Chris Wolverton's group at Northwestern University.




## Shortcuts

**Search**
Material Compositions

**Query**
Materials Data

**Create**
Phase Diagrams

**Determine**
Ground State Compositions (GCLP)

**Visualize**
Crystal Structures

**RESTful API**
(OPTIMADE)

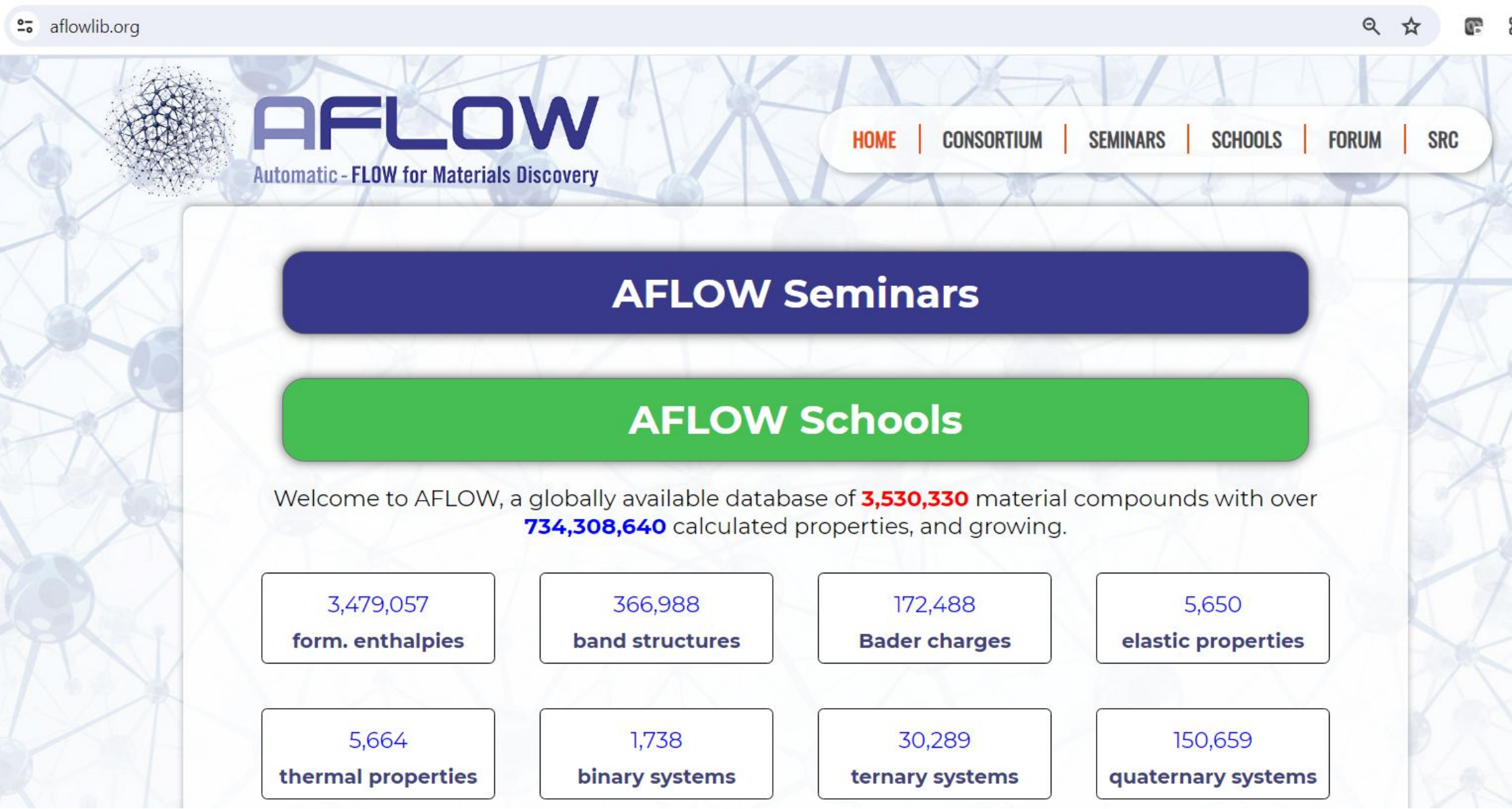
# The Open Quantum Materials Database: Features

**Motto: We do all the calculations**

**All calculations are performed with GGA or GGA+*U***

**Structures include hypothetical materials (not known experimentally)**

**Typical data: Formation and decomposition energies**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form. enthalpies
366,988
band structures
172,488
Bader charges
5,650
elastic properties
5,664
thermal properties
1,738
binary systems
30,289
ternary systems
150,659
quaternary systems

# Automatic FLOW library: Features

**Leader: Stefano Curtarolo (Duke University)**

**Motto: We do all the calculations**

**Calculations perfomed with GGA, GGA+*U*, ACBN0 (pseudo-hybrid)**

**Typical data: Relaxed geometries, electronic and phonon band structures, magnetic properties, thermodynamic properties**

**Provides tools for performing high-throughput calculations**

nomad-lab.eu/nomad-lab/

NOMAD

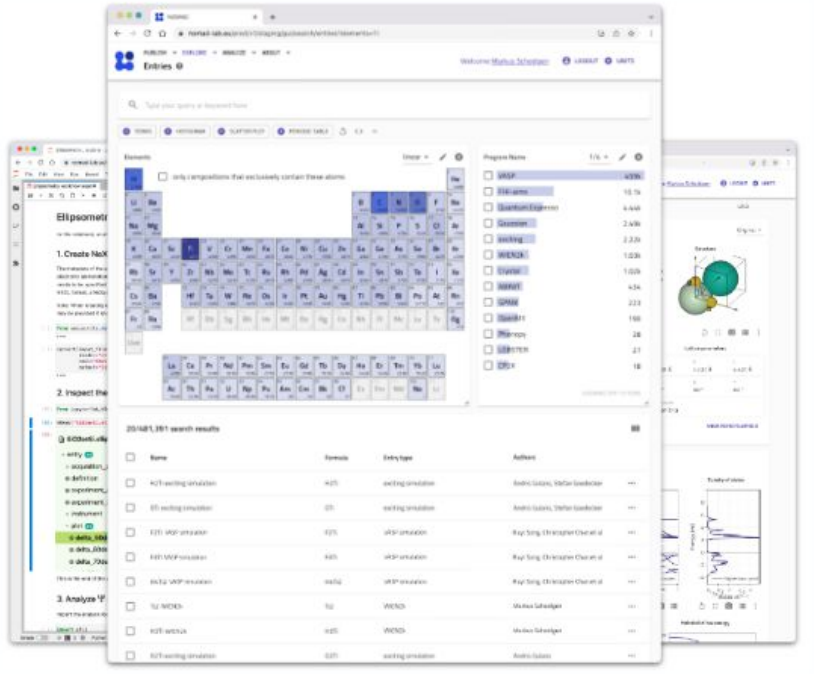
SOLUTIONS LEARN GET INVOLVED ABOUT OPEN NOMAD

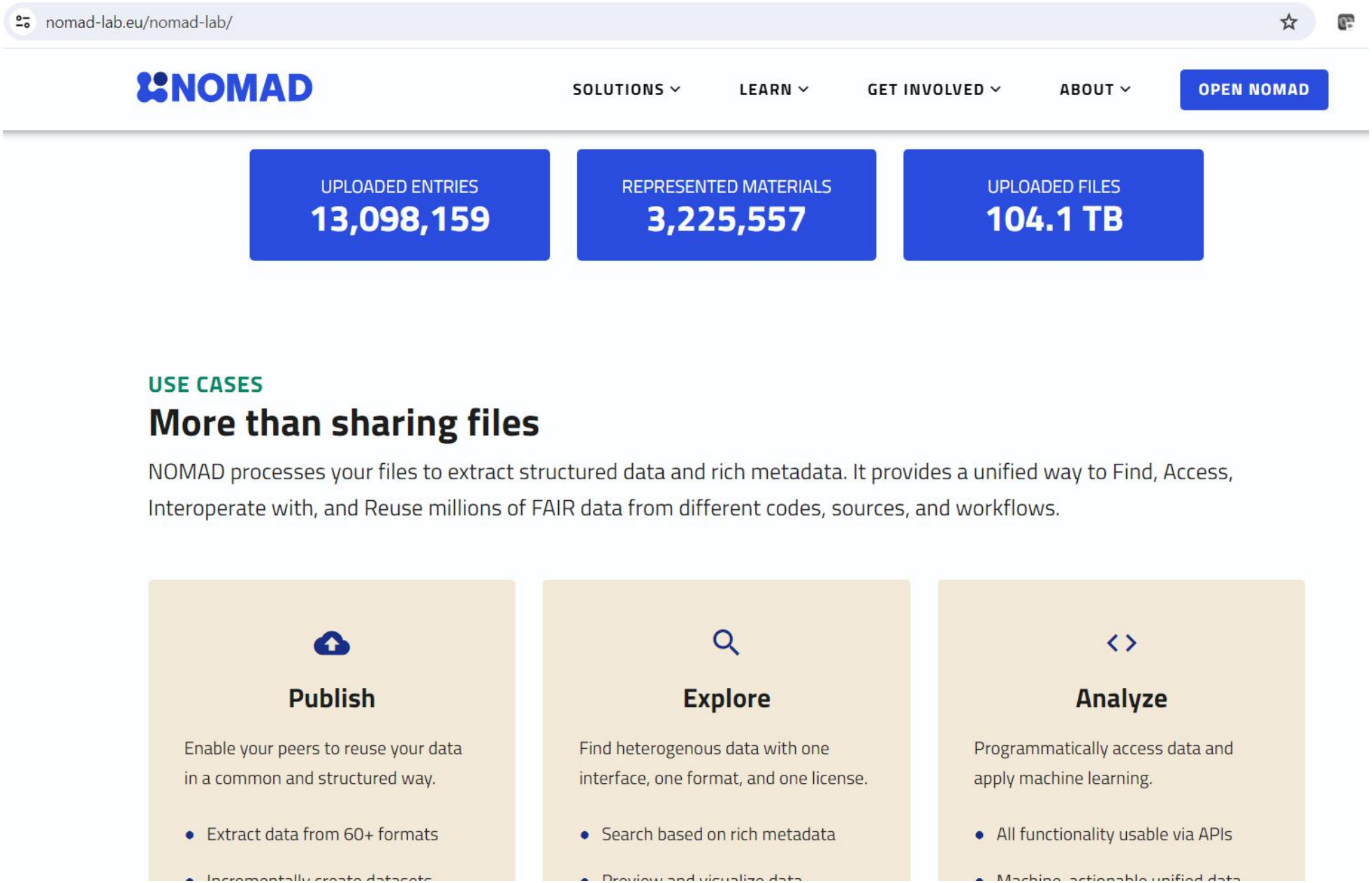
# NOMAD

## Materials science data managed and shared

NOMAD lets you manage and share your materials science data in a way that makes it truly useful to you, your group, and the community. **Free and open source.**

Open NOMAD →

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Analyze
Programmatically access data and apply machine learning.
All functionality usable via APIs

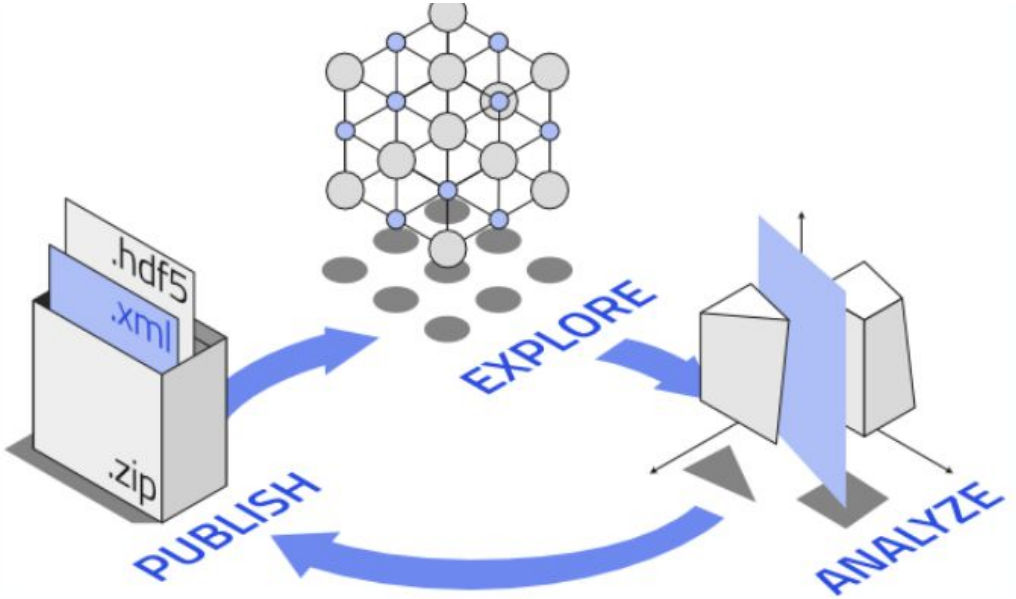
# The NOMAD (Novel Materials Discovery) Laboratory

## Part of FAIRmat consortium for condensed matter and the chemical physics of solids

**NOMAD**

**Share data, not files.**

NOMAD lets you extract and publish structured data with rich metadata. It provides a unified way to Find, Access, Interoperate with, and Reuse millions of FAIR data from different codes and sources.




**Contains raw data (input and output) uploaded by users**

# The NOMAD (Novel Materials Discovery) Laboratory

## Part of FAIRmat consortium for condensed matter and the chemical physics of solids

Leaders: Claudia Draxl, Matthias Scheffler (Humboldt University, Berlin)

Motto: We will store your data, you decide if it is open access or restricted.
Even inaccurate calculations can be used for learning, provided all metadata (code version, method, basis set, etc.) are known

Includes data from AFLOW, OQMD, Materials Project

Automatic parsing of inputs and outputs from all major electronic-structure packages

Common format (metadata) for data from different electronic-structure packages

Parsable data: Total energies, geometry optimization, molecular dynamics, thermodynamic properties

aiida.net

AiiDA

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Search Ctrl + K

# Automated workflows for computational science.

Download

Provenance Tracking

Plugin Framework

HPC Interface

Open Source

## Events

**Mon 28th Aug - Fri 1st Sep 2023**
*AiiDA intro and demo at the Adv. Quantum ESPRESSO school*
*Pavia (IT)*

**Fri 20th May 2022**
*AiiDA introductory tutorial*
*University of Antwerp (BE)*

**Mon 6th - Fri 10th Dec 2021**
*Fifth annual AiiDA coding week*
*Leysin (CH)*

**Fri 28th May 20**
*AiiDA tutorial at*
*on Advanced Mat*
*Molecular Modell*
*Quantum ESPRES*

**Tue 4th - Fri 7th Oct 2022**
*AiiDA online demonstration and virtual tutorial*
*Online*

**Mon 16th - Fri 20th May 2022**
*Wannier 2022 Summer School*
*Trieste (IT)*

**Mon 5th - Fri 9th Jul 2021**
*Tutorial on running and writing workflows with AiiDA*
*Online*

# Automated Interactive Infrastructure and Database for Computational Science (AiiDA)

> AiiDA is an open-source Python infrastructure to help researchers with automating, managing, persisting, sharing and reproducing the complex workflows associated with modern computational science and all associated data.

**Leader: Nicola Marzari (EPFL, Switzerland)**

**Motto: Build your own database with our tools**

**Provides tools for performing high-throughput calculations**