Fritz-Haber-Institut

NOMAD
NOVEL MATERIALS DISCOVERY

Exploring the materials space via regularized and symbolic regression (compressed sensing)

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Fritz-Haber-Institut der MPG, Berlin

ML for MS

ML4MS 2019
Young Researcher's Workshop on Machine Learning for Materials Science 2019
06-10 May 2019, Aalto University, Helsinki (FI)

Charts/maps of materials

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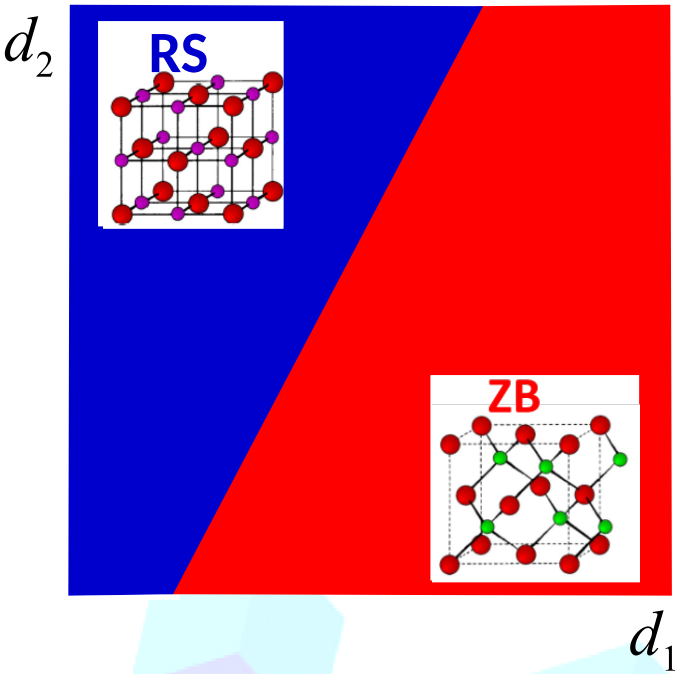

Octet
binaries

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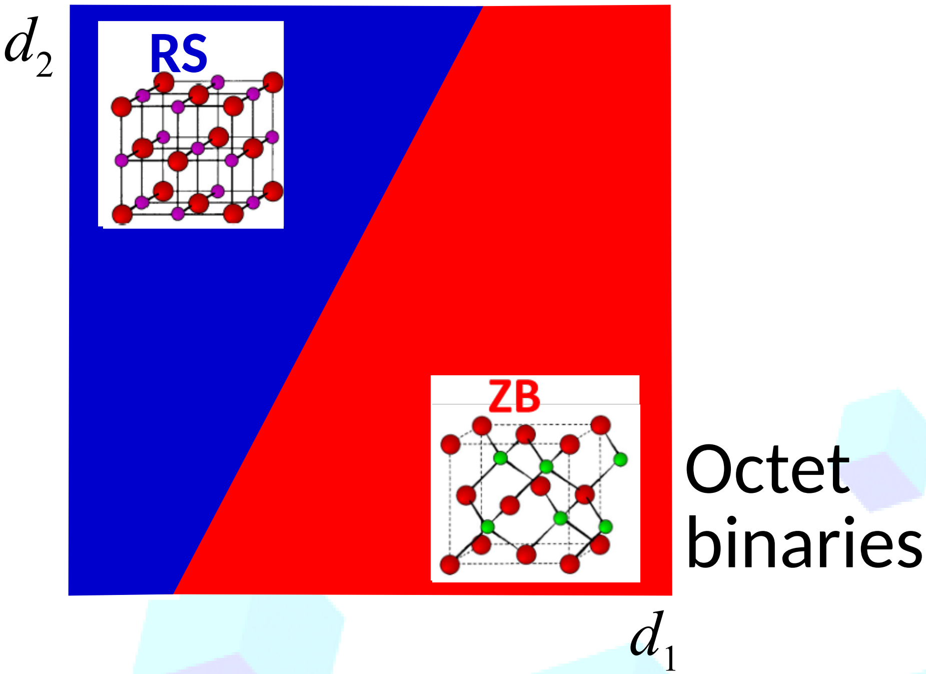

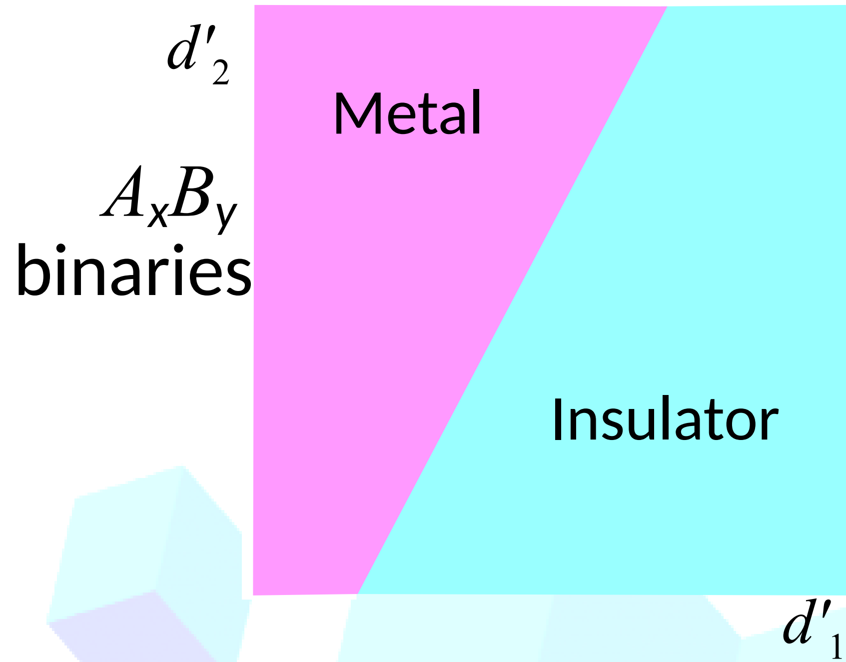

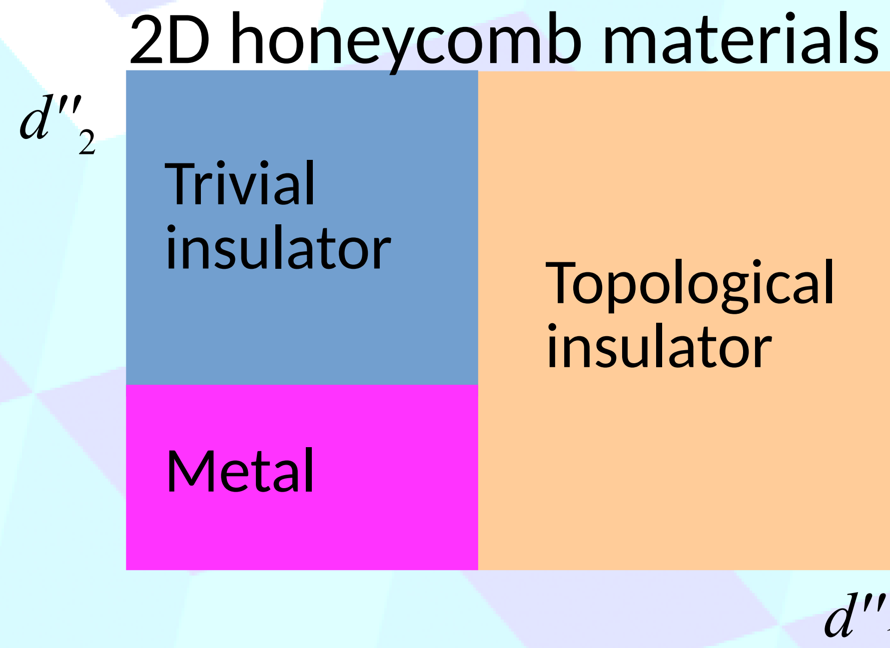

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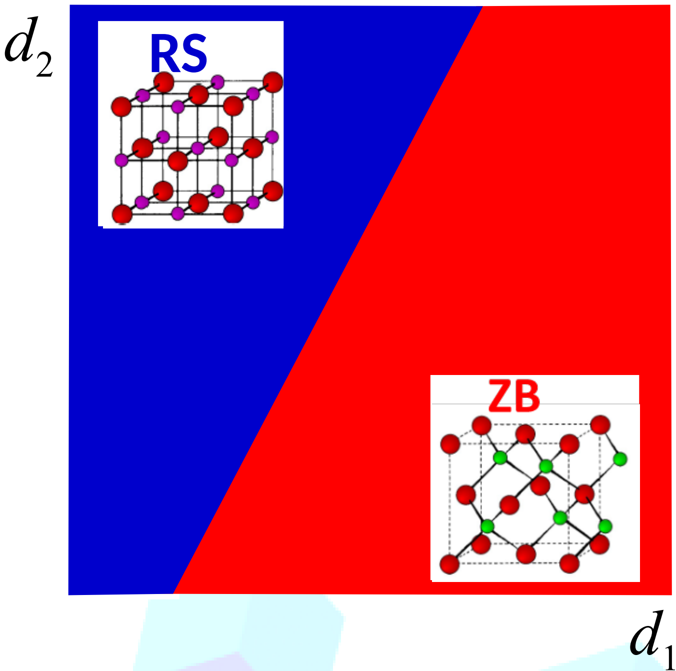
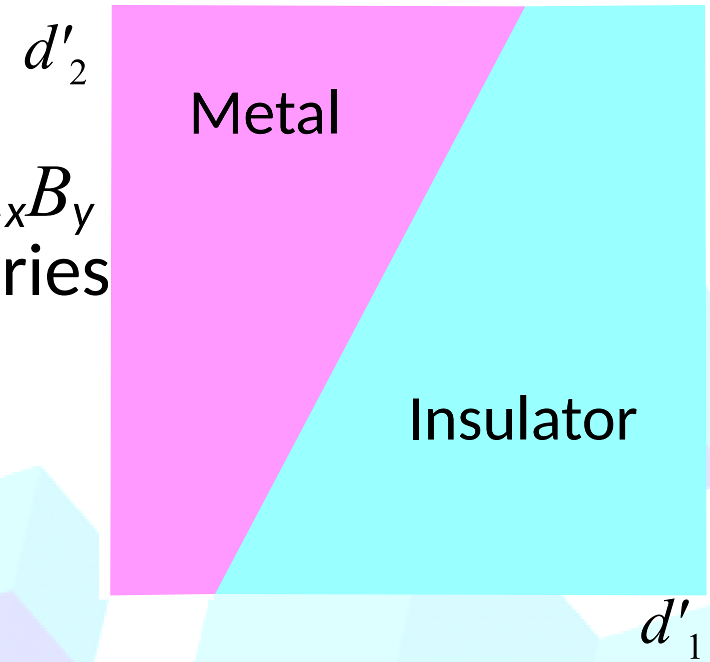
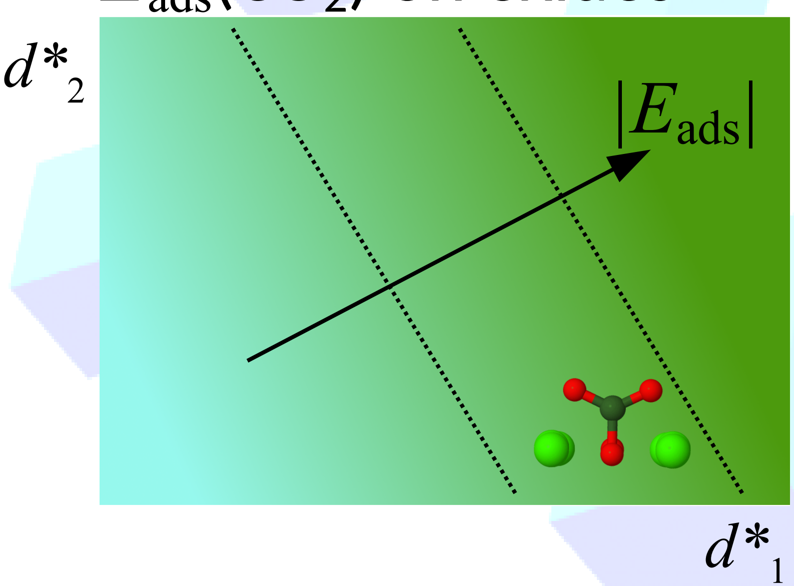


Octet binaries

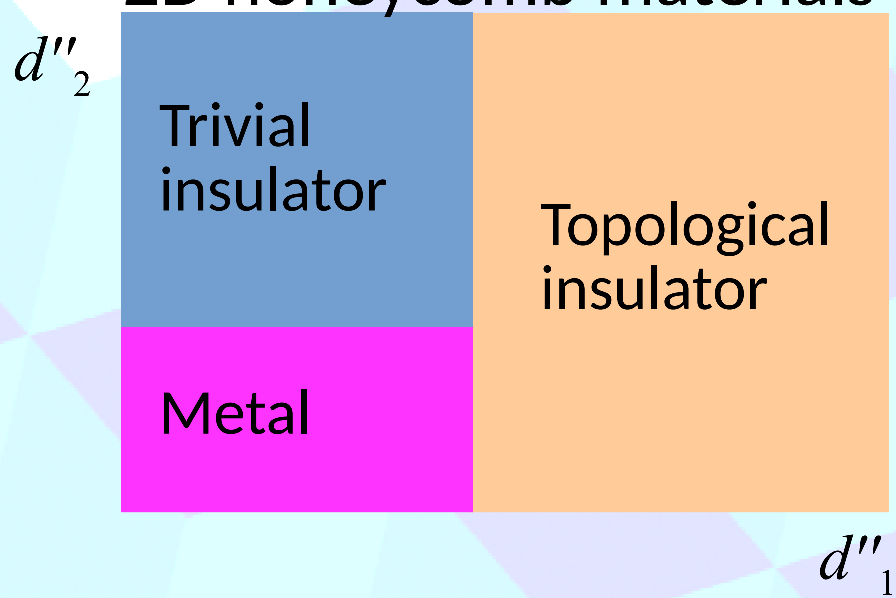
A_xB_y binaries



$E_{\mathrm{ads}}(CO_2)$ on oxides



2D honeycomb materials



Building maps of materials properties
A quantum many-body problem

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For instance, given a class of chemical compositions (e.g., via prototype formula, such as ABX_3):

- what is the most stable crystal structure of each material in the class?
- which materials are metals / topological insulators / superconductors ?
- which material has the highest melting point?
- which materials has a surface optimal for catalysing some chemical reaction?

The Big Picture

- Design of new materials:
  preparation, synthesis, and characterization is complex and costly

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- Data analytics tools will help to identify trends and anomalies in data and guide discovery of new materials

We have a dream

From the **periodic table of the elements**
to **charts of materials**

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From the **periodic table of the elements** to **charts of materials**

Reihen	Gruppe I. — R^2O	Gruppe II. — RO	Gruppe III. — R^2O^3	Gruppe IV. RH^4 RO^2	Gruppe V. RH^3 R^2O^5	Gruppe VI. RH^2 RO^3	Gruppe VII. RH R^2O^7	Gruppe VIII. — RO^4
1	H=1							
2	Li=7	Be=9.4	B=11	C=12	N=14	O=16	F=19	
3	Na=23	Mg=24	Al=27.3	Si=28	P=31	S=32	Cl=35.5	
4	K=39	Ca=40	—=44	Ti=48	V=51	Cr=52	Mn=55	Fe=56, Co=59, Ni=59, Cu=63.
5	(Cu=63)	Zn=65	—=68	—=72	As=75	Se=78	Br=80	
6	Rb=85	Sr=87	?Yt=88	Zr=90	Nb=94	Mo=96	—=100	Ru=104, Rh=104, Pd=106, Ag=108.
7	(Ag=108)	Cd=112	In=113	Sn=118	Sb=122	Te=125	J=127	
8	Cs=133	Ba=137	?Di=138	?Ce=140	—	—	—	— — — —
9	(—)	—	—	—	—	—	—	
10	—	—	?Er=178	?La=180	Ta=182	W=184	—	Os=195, Ir=197, Pt=198, Au=199.
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12	—	—	—	Th=231	—	U=240	—	— — — —

Mendeleev's **1871** periodic table

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Ga=69.7 Ge=72.6

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Suppose
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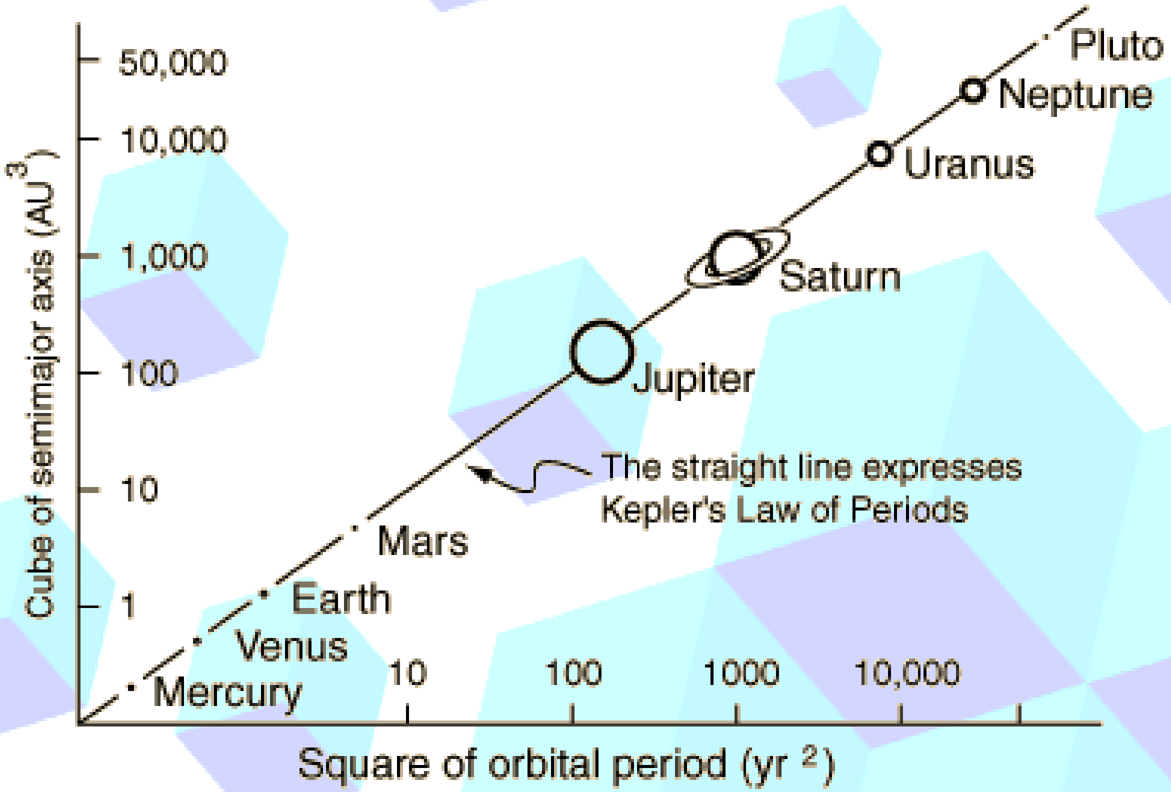

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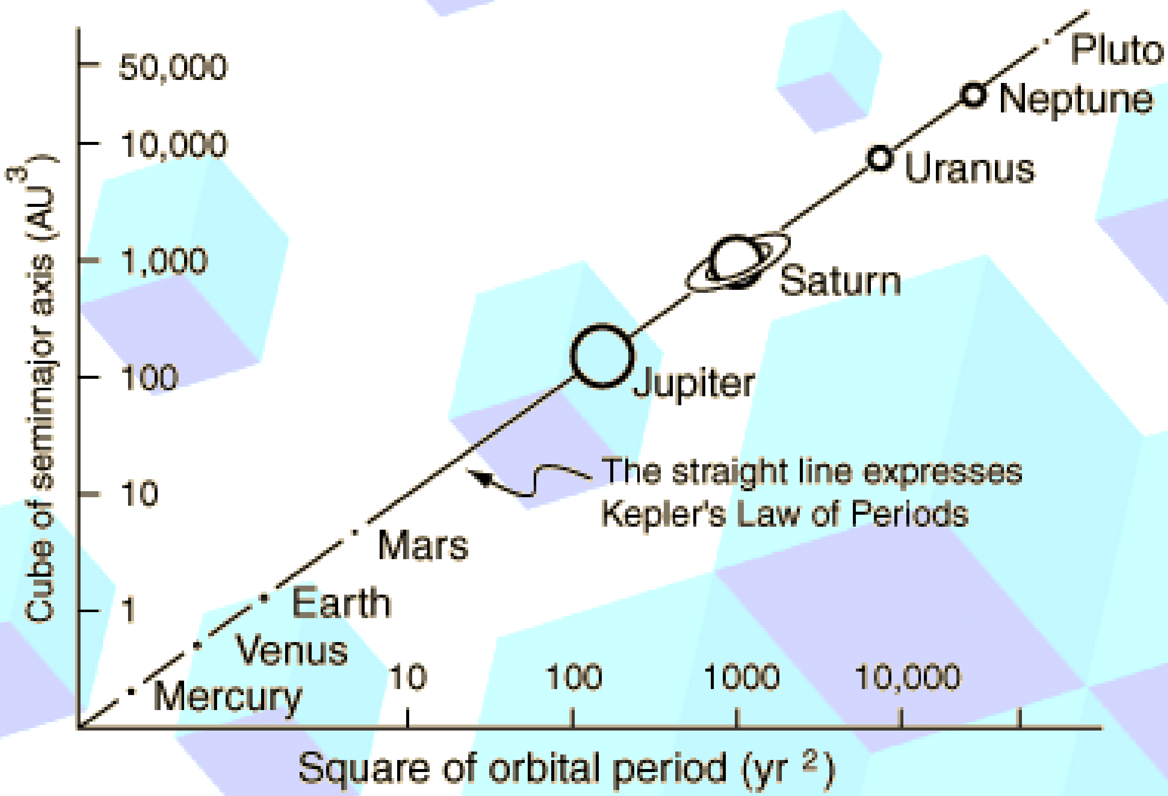

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Data
(collected by
Tycho Brahe)

↓

Statistical learning
(performed by
Johannes Kepler)

↓

Physical law
(assessed by
Isaac Newton)

Supervised (big-)data analysis: a flow chart

Training set
Calculate properties and functions
P_i, for many *materials*, i
E.g., Density-Functional Theory

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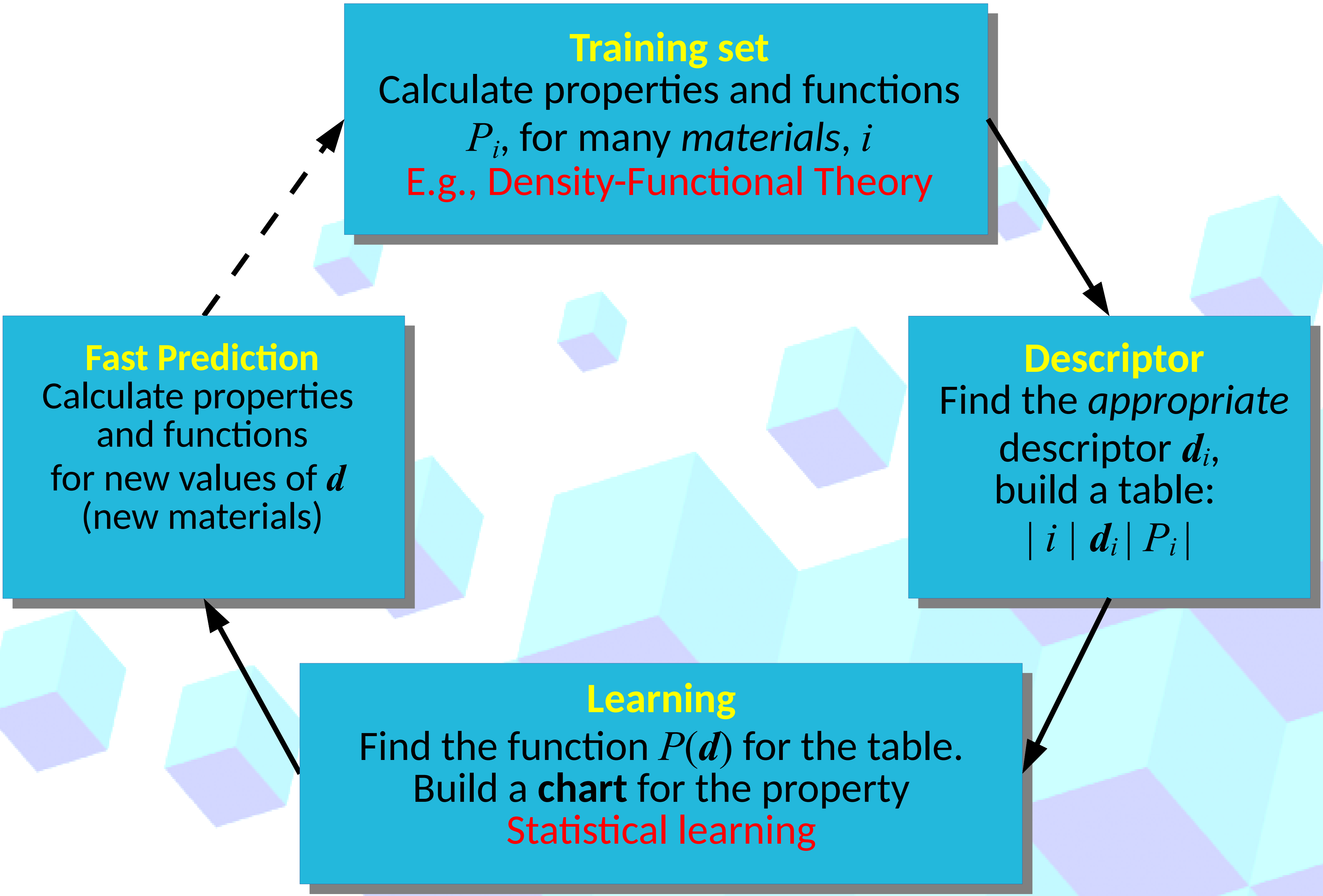

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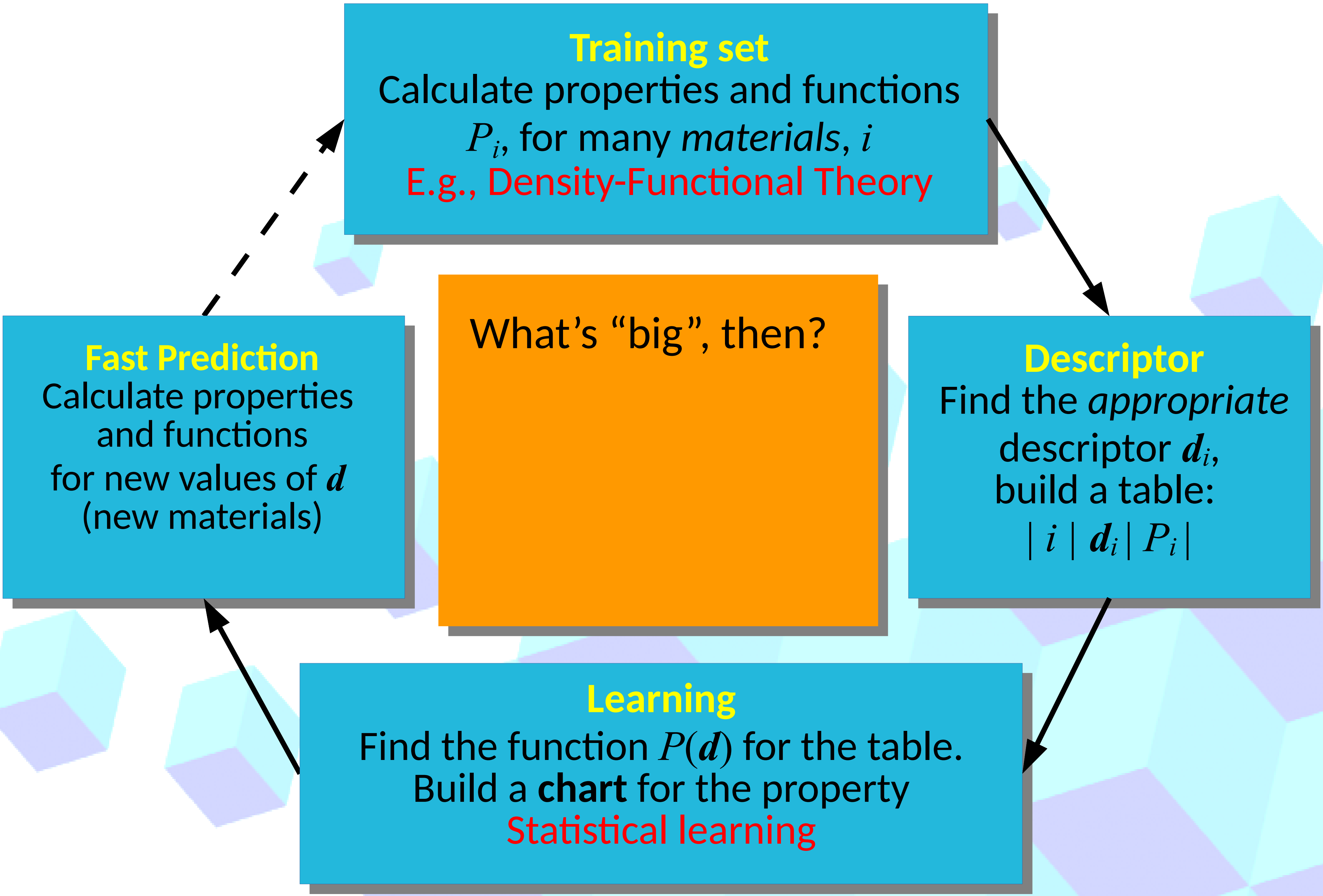

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What’s “big”, then?

- Volume
- Velocity
- Variety
- Veracity issue

Descriptor? Don't we know it from the start?

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$\{R_\mathrm{I}, Z_\mathrm{I}\} \rightarrow$ Hamiltonian

$\{R_\mathrm{I}\} \rightarrow$ Geometry
- translational, rotational, permutational invariant
- coarse graining $\{R_\mathrm{I}\}$?

$\{Z_\mathrm{I}\} \rightarrow$ Chemistry

Regression: Mathematical formulation

Figure of merit to be optimized:

$$\underset{\boldsymbol{c}\in\mathbb{R}^M}{\operatorname{argmin}} \sum_{j=1}^{N} \left(P_j - \sum_{l=1}^{M} d_{j,l} c_l \right)^2 = \underset{\boldsymbol{c}\in\mathbb{R}^M}{\operatorname{argmin}} \| \boldsymbol{P} - \boldsymbol{D}\boldsymbol{c} \|_2^2$$

ℓ_2 norm

Ridge Regression: Mathematical formulation

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Regularization (prefer "lower complexity" in the solution)

$$\underset{\boldsymbol{c}\in\mathbb{R}^M}{\operatorname{argmin}} \|\boldsymbol{P} - \boldsymbol{D}\boldsymbol{c}\|_2^2 + \lambda \|\boldsymbol{c}\|_2^2$$

(Linear) ridge regression

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Explicit solver:

$$\boldsymbol{c} = \left(\boldsymbol{D}^\top \boldsymbol{D} + \lambda \boldsymbol{I} \right)^{-1} \boldsymbol{D}^\top \boldsymbol{P}$$

Alternative view, via Hilbert space representation theorem:

$$\boldsymbol{c} = \sum_j \alpha_j \boldsymbol{d}_j$$

Sum over data points!

Kernel Ridge Regression: Mathematical formulation

$$\operatorname*{argmin}_{\boldsymbol{c} \in \mathbb{R}^M} \|\boldsymbol{P} - \boldsymbol{D}\boldsymbol{c}\|_2^2 + \lambda \|\boldsymbol{c}\|_2^2 \quad \Rightarrow \quad \boldsymbol{c} = \left(\boldsymbol{D}^\top \boldsymbol{D} + \lambda \boldsymbol{I}\right)^{-1} \boldsymbol{D}^\top \boldsymbol{P}$$

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$$K_{ij} = \langle \boldsymbol{d}_i, \boldsymbol{d}_j \rangle$$

Linear kernel

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Non-linear kernel

$$\boldsymbol{c} = \sum_j \alpha_j \Phi(\boldsymbol{d}_j)$$

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$$K_{ij} = \langle \boldsymbol{d}_i, \boldsymbol{d}_j \rangle$$ **Linear kernel**

$$K_{ij} = (\langle \boldsymbol{d}_i, \boldsymbol{d}_j \rangle + b)^n$$ **Polynomial kernel**

$$K_{ij} = \exp\left(\frac{\|\boldsymbol{d}_i - \boldsymbol{d}_j\|^2}{2\sigma^2}\right)$$ **Gaussian (radial basis function) kernel**

$$K_{ij} = \exp\left(\frac{\|\boldsymbol{d}_i - \boldsymbol{d}_j\|}{\sigma}\right)$$ **Laplacian kernel**

Kernel Ridge Regression: Mathematical formulation

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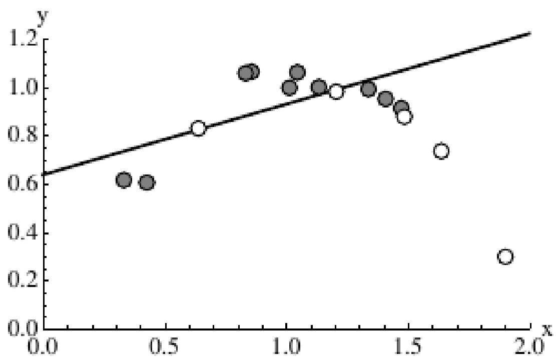
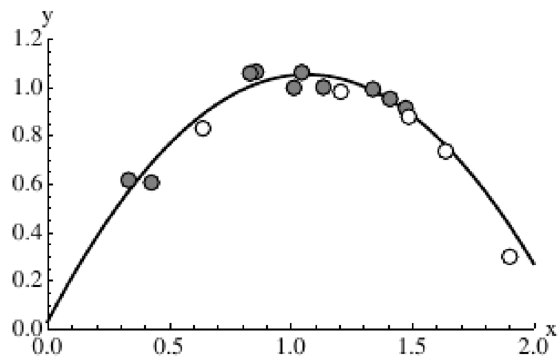
In all cases,
a kernel introduces a
similarity measure

Regularized regression in practice: beware of overfitting

Underfitting



Fitting



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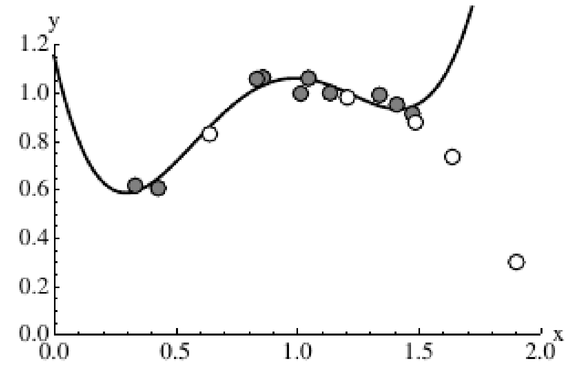

Training/ validation error

0.123 / 0.443

0.044 / 0.068

0.036 / 0.939

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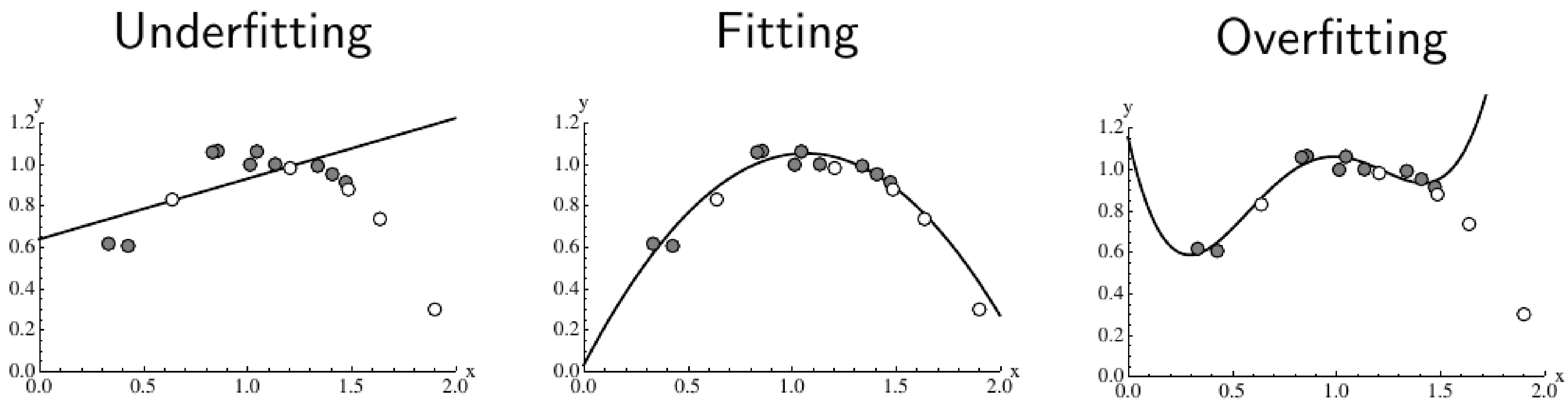

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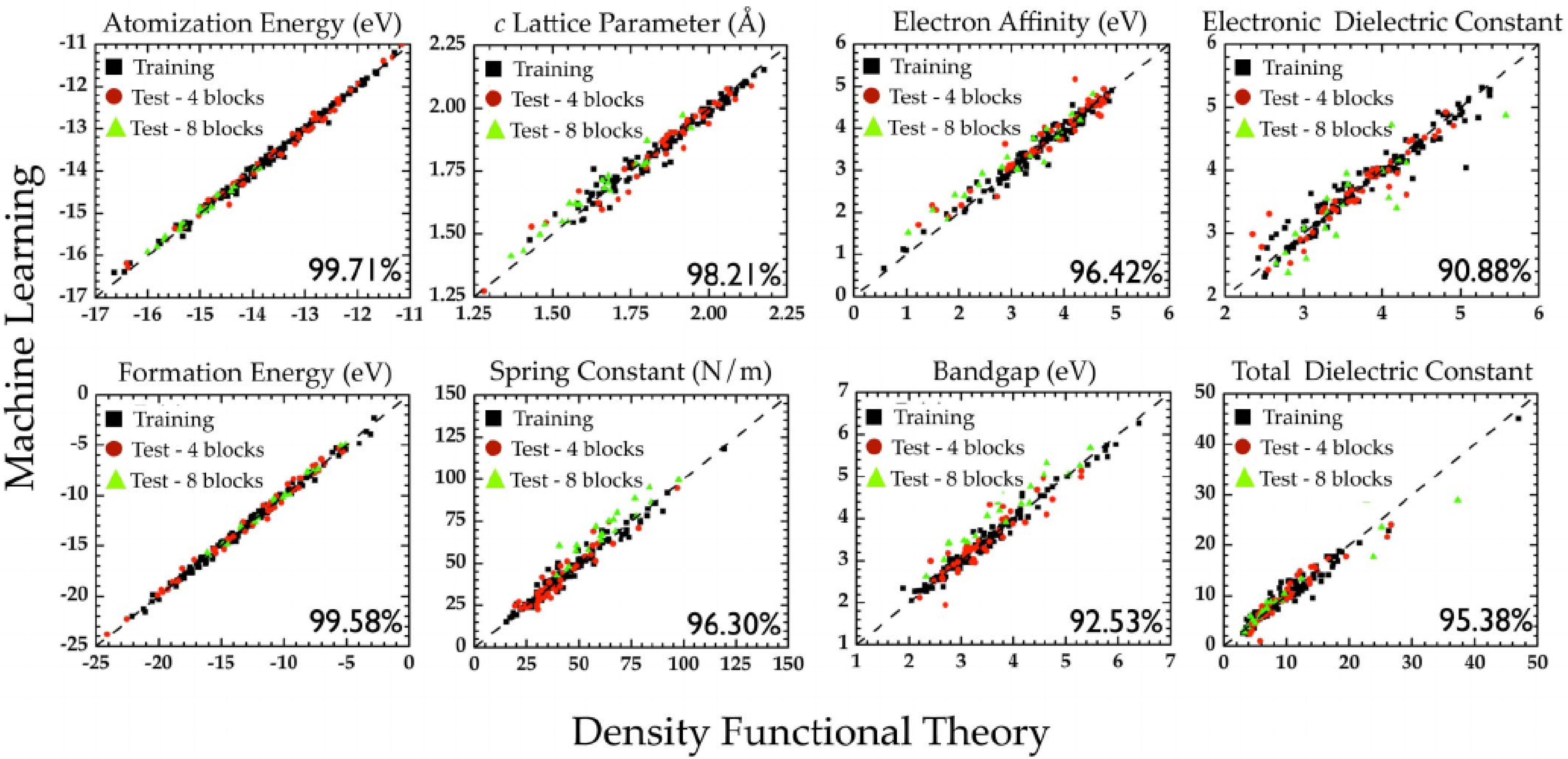
KRR success stories: 1D polymers "eugenetics"

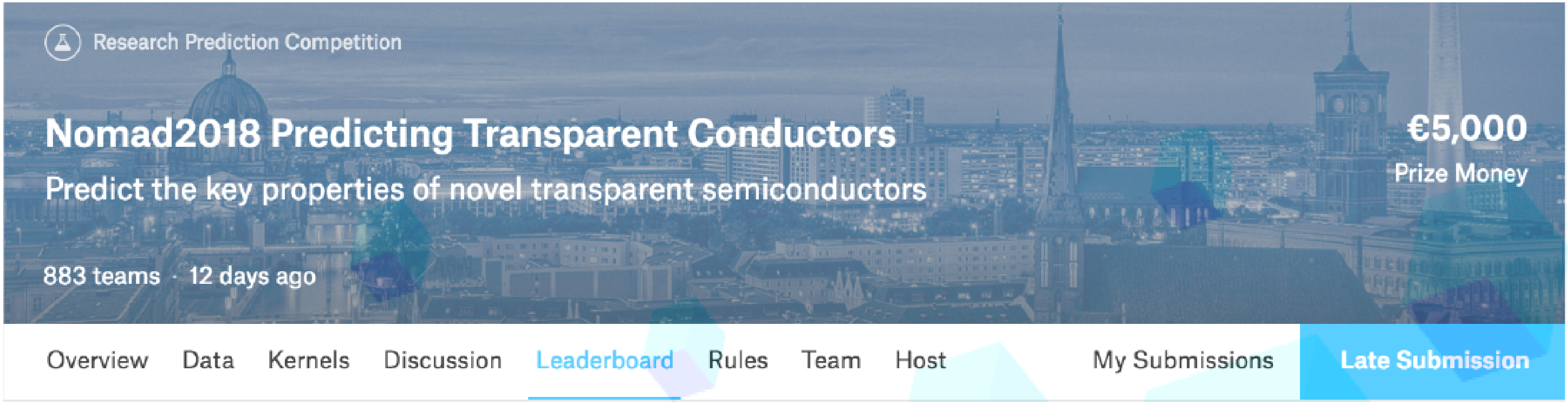
Data: 175 linear 4-blocks periodic polymers. 7 blocks: CH_2, SiF_2, $SiCl_2$, GeF_2, $GeCl_2$, SnF_2, $SnCl_2$,

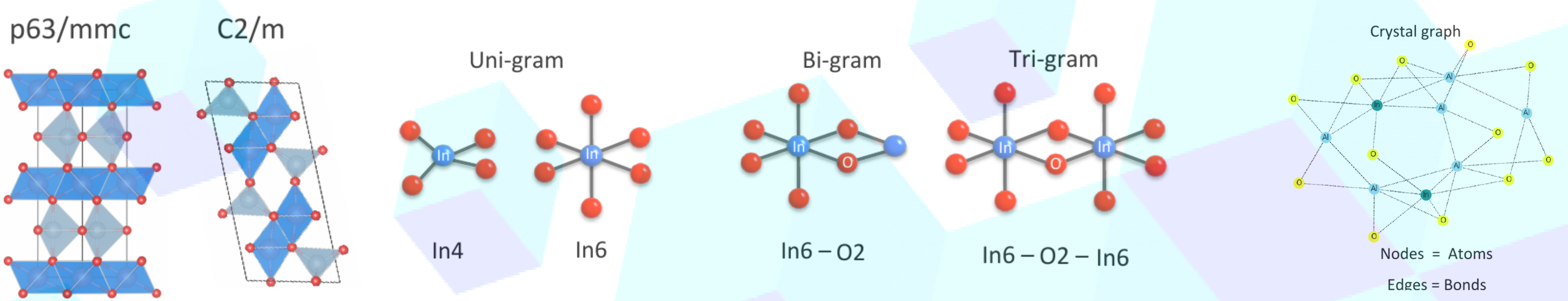
Descriptor: 20 dimensions [# building blocks of type *i*, of *ii* pairs, of *iii* triplets]



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

KRR success stories: *n*-grams for kaggle





Input features: count number of sequences of various lengths

Unigrams: 2 O3, 2 Ga4, 1 O2, 1 In5

Bigrams: 2 O3-Ga4, 2 Ga4-O2, 1 O3-In5

Sutton *et al.*, npj Comp. Materials, in press (2019), arXiv: 1812.00085

Compressed sensing: the quest for descriptors and predictive models

$$\arg\min_{\boldsymbol{c}} \left(\|\boldsymbol{P} - \boldsymbol{D}\boldsymbol{c}\|_2^2 + \lambda \|\boldsymbol{c}\|_0 \right)$$

Compressed-sensing-based model identification:
Shares concepts with

Regularized regression. But: Massive sparsification.

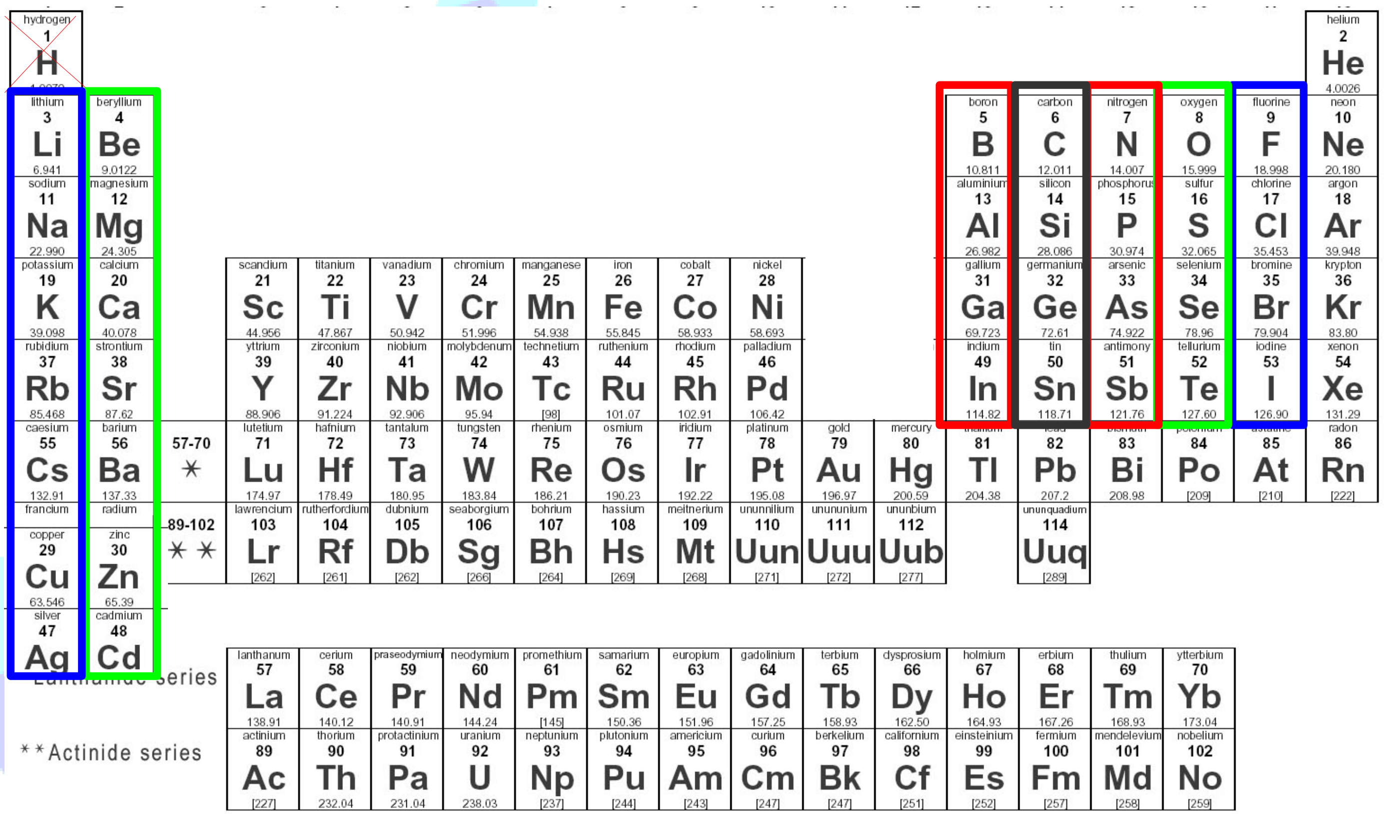
Dimensionality reduction. But supervised, and yielding sparse, "inspectable" descriptors

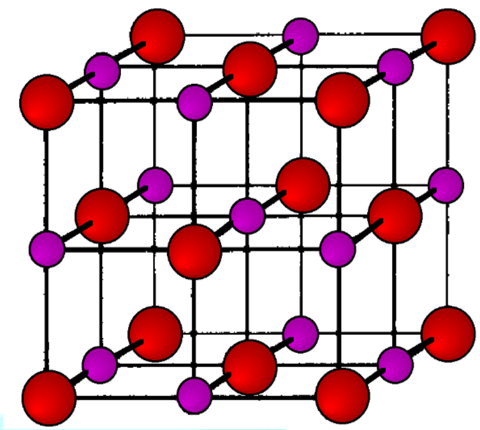
Feature/Basis-set selection/extraction. But: non-greedy solver.

Symbolic regression. But: deterministic solver.

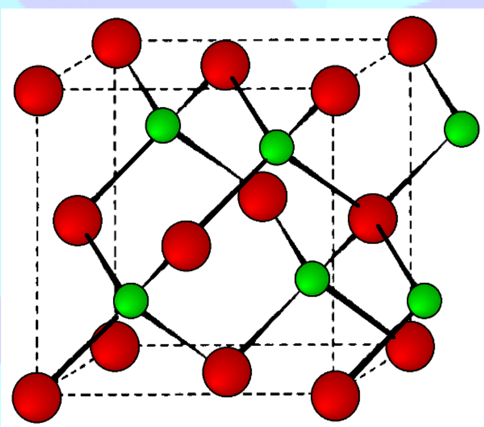
An example: predicting crystal structures from the composition

82 octet AB binary compounds





Rock salt



Zinc blende

An example: predicting crystal structures from the composition

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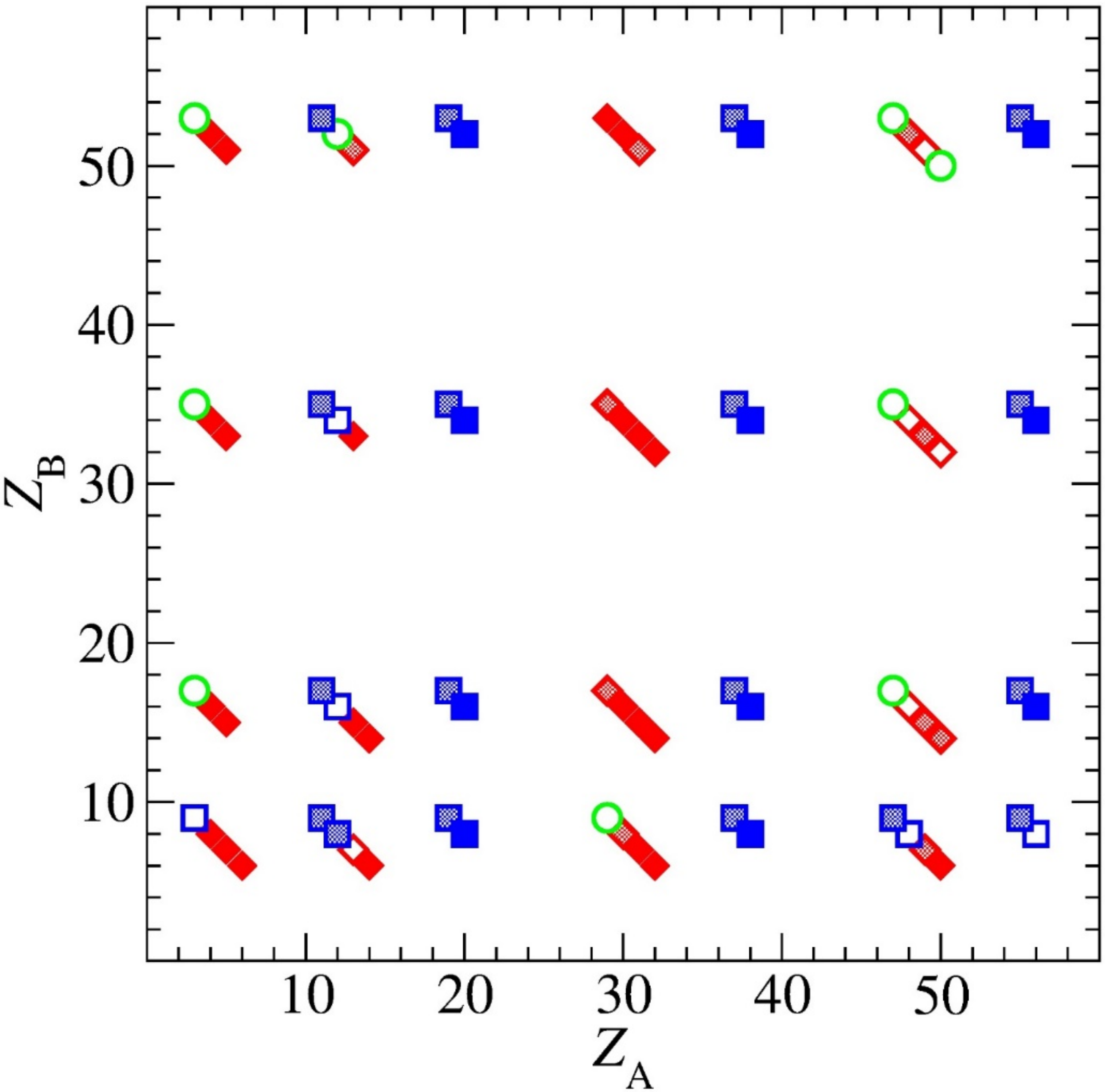
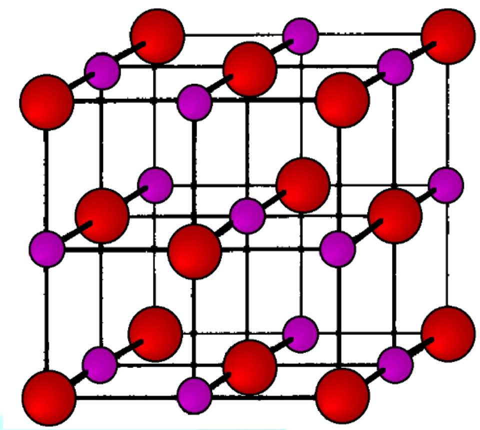
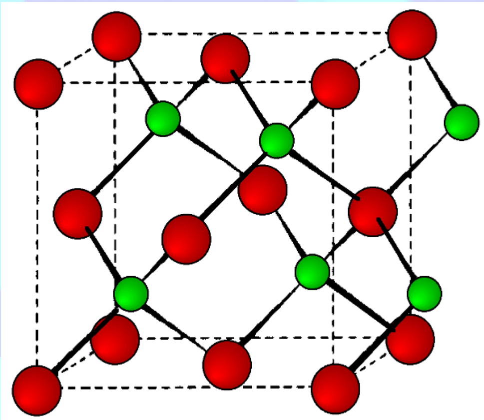




Rock salt

- Rock salt
- Rock salt/Zinc blende
- Zinc blende



Zinc blende

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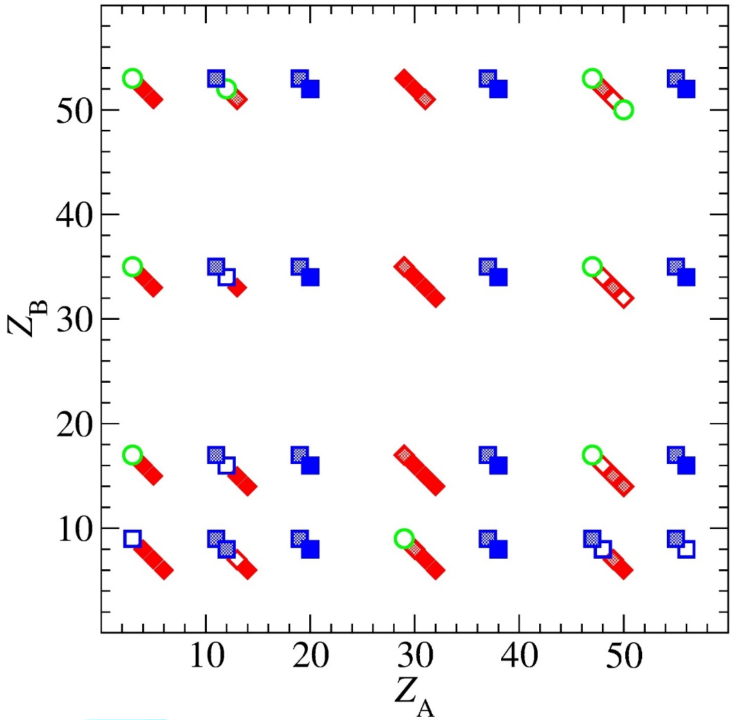

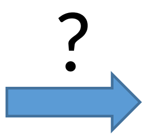

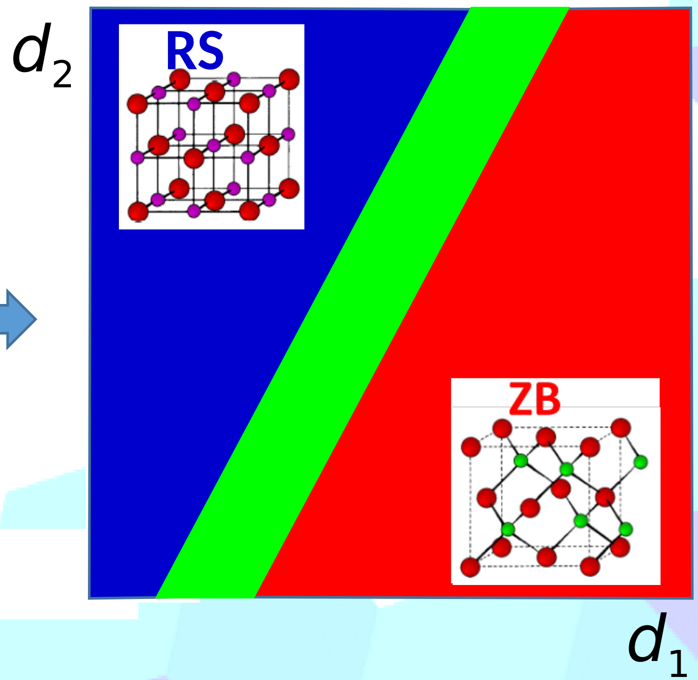

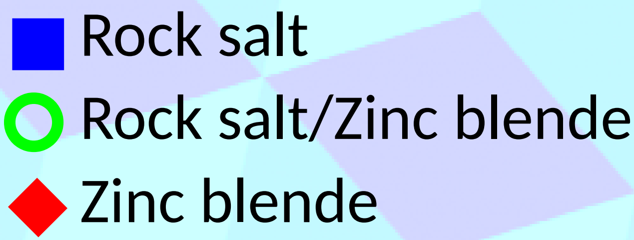

J. A. van Vechten, Phys. Rev. 182, 891 (1969).
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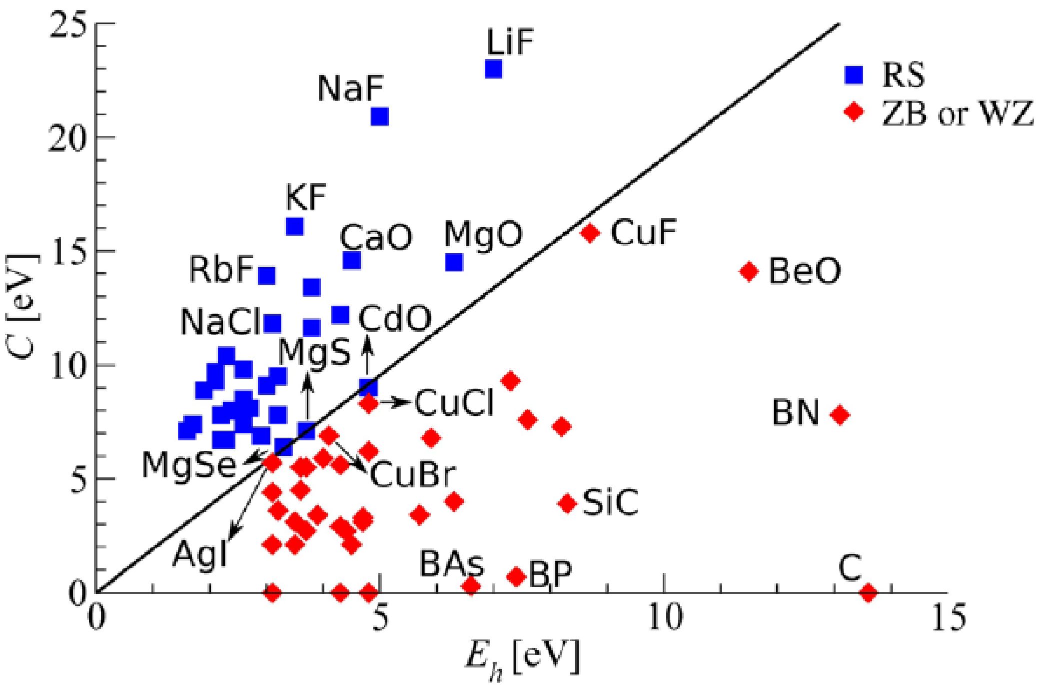

d_1

The descriptor proposed by Phillips and van Vechten in 1969-70 depends on:
- lattice parameter
- electrical conductivity

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Ansatz: atomic features

- HOMO
- LUMO
- Ionization Potential
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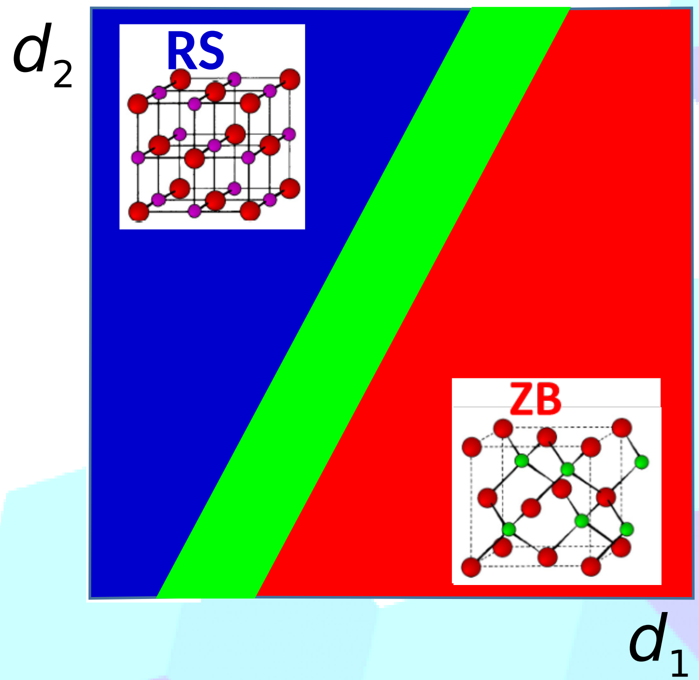
- Radius of valence *p* orbital
- Radius of valence *d* orbital
- … ?

E(Rock salt) – *E*(Zinc blende)



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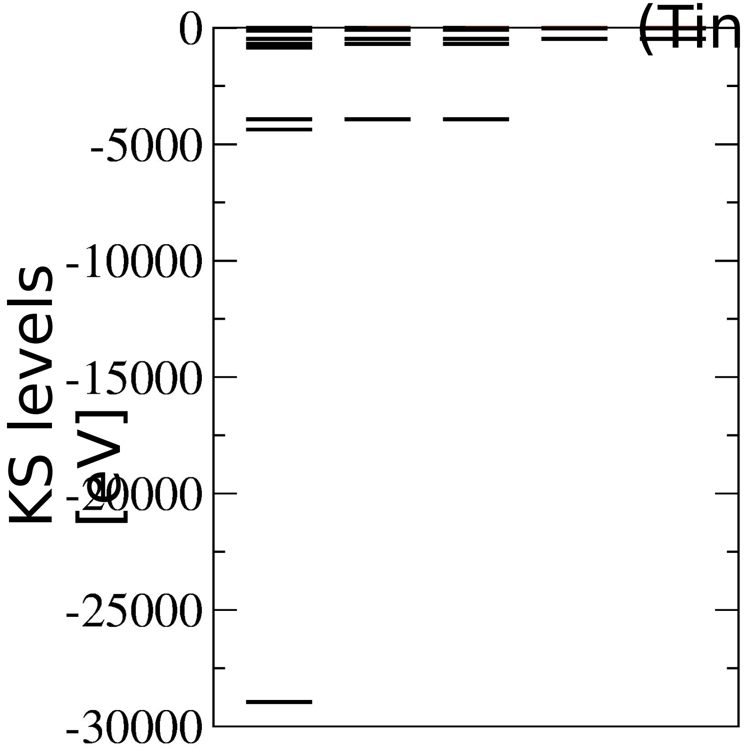
Primary (atomic) features

example: Sn (Tin)



Primary (atomic) features

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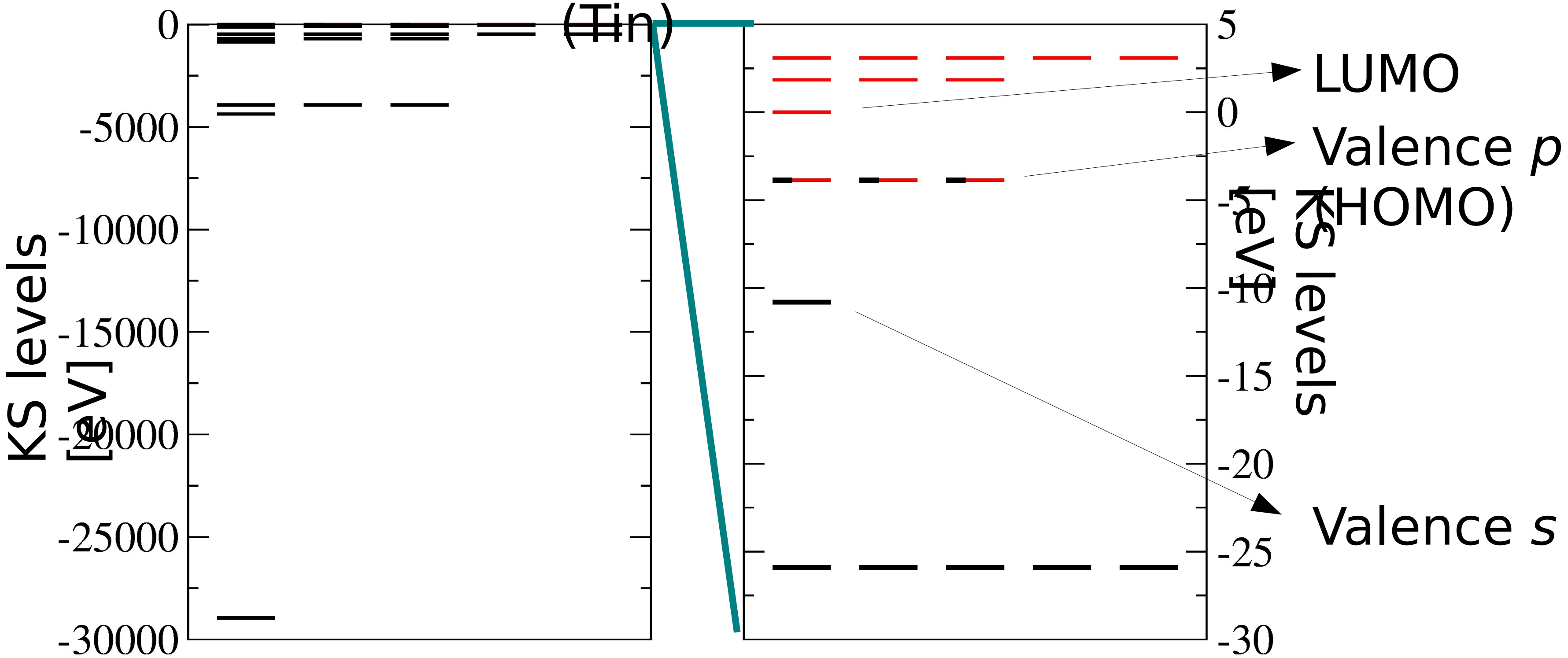
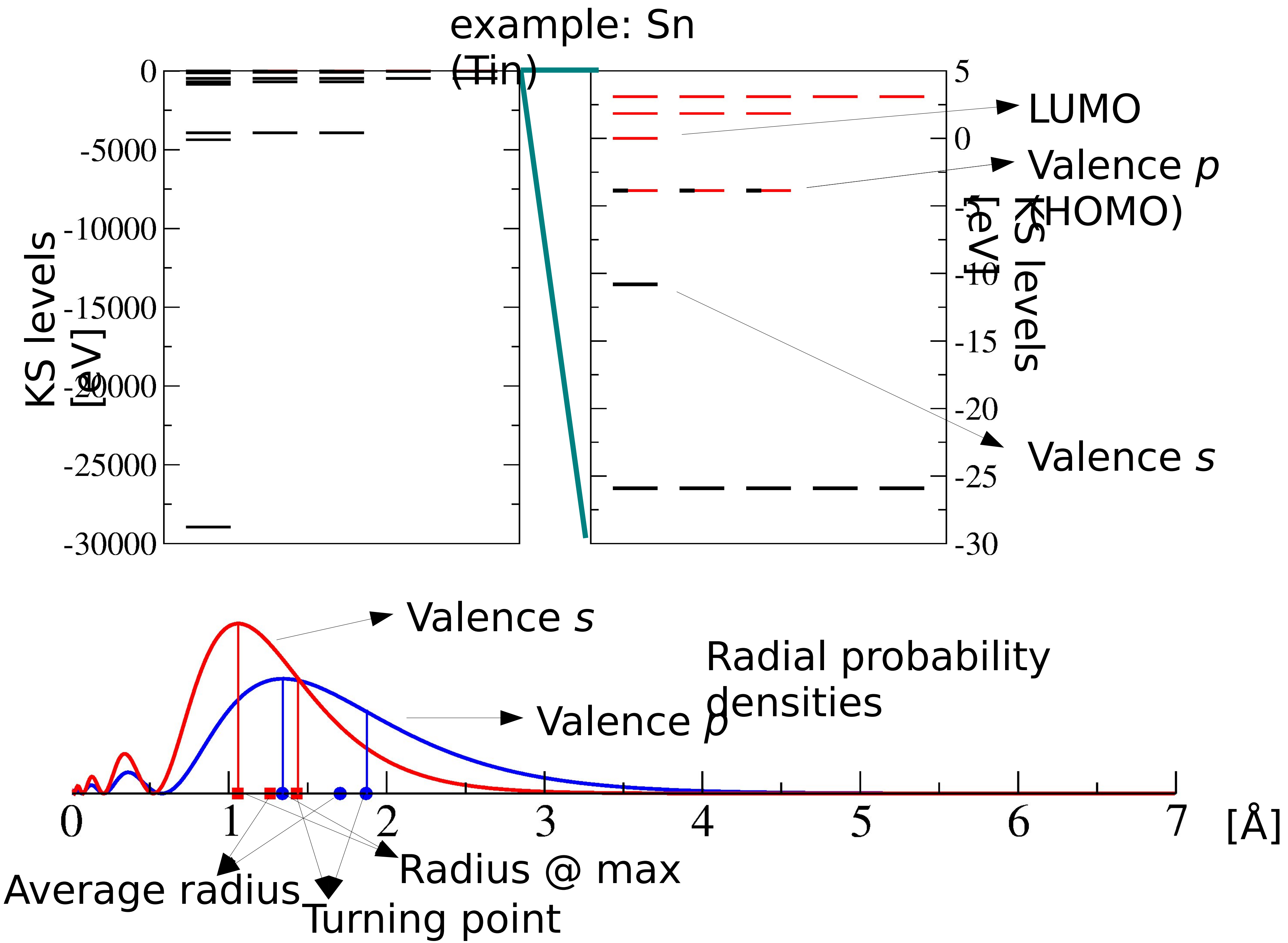


Primary (atomic) features



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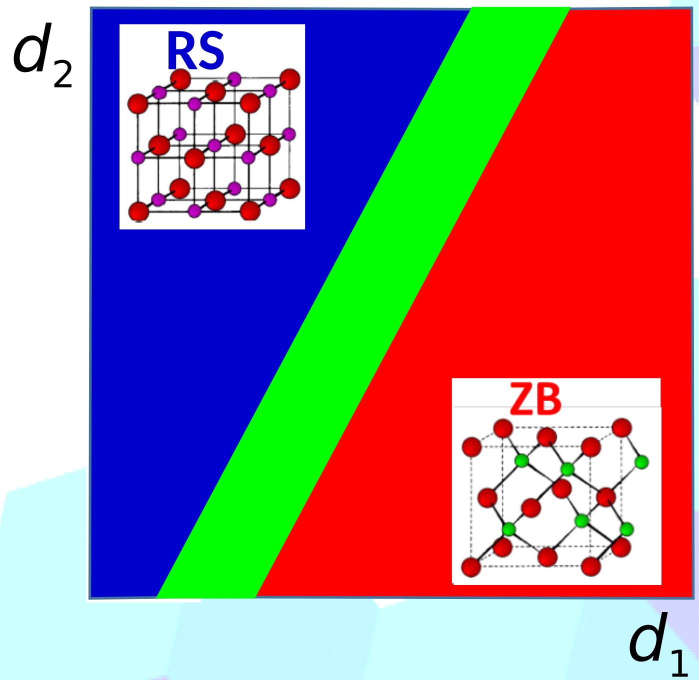

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Y. Saad, D. Gao, T. Ngo, S. Bobbitt, J. R. Chelikowsky, and W. Andreoni, Phys. Rev. B 85, 104104 (2012).

Compressed sensing

Aim: finding descriptors and learning predictive models
Ansatz:
$\boldsymbol{P} = c_1\boldsymbol{d}_1 + c_2\boldsymbol{d}_2 + \ldots c_n\boldsymbol{d}_n$
Where
$\boldsymbol{P}$ is the property of interest
$\boldsymbol{d}_1, \ldots \boldsymbol{d}_n$ are candidate features, i.e., nonlinear functions of primary features (EA, IP, …)
$c_1, \ldots c_\mathrm{n}$ are unknown coefficients, with the extra constraint that these (nonzero) coefficients should be as few as possible.

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With a foreword on
dimensionality reduction

Linear dimensionality reduction: Principal components

Pearson, K. "On Lines and Planes of Closest Fit to Systems of Points in Space". Philosophical Magazine 2, 559 (1901)

Linear dimensionality reduction: Principal components

Pearson, K. "On Lines and Planes of Closest Fit to Systems of Points in Space". Philosophical Magazine 2, 559 (1901)

Orthonormal transformation of coordinates, converting a set of (possibly) linearly correlated coordinates into a new set of linearly uncorrelated (called principal or normal) components, such that the first component has the largest variance and each subsequent has the largest variance constrained to being orthogonal to all the preceding components

Linear dimensionality reduction: Principal components

Ansatz: atomic features

- Valence number Z_v
- Energy of valence *s* orbital E_s
- Energy of valence *p* orbital E_p
- Radius of valence *s* orbital r_s
- Radius of valence *p* orbital r_p

r_s, r_p, $E_s/\sqrt{Z_v}$, $E_p/\sqrt{z_v}$,
for A and B atoms

est

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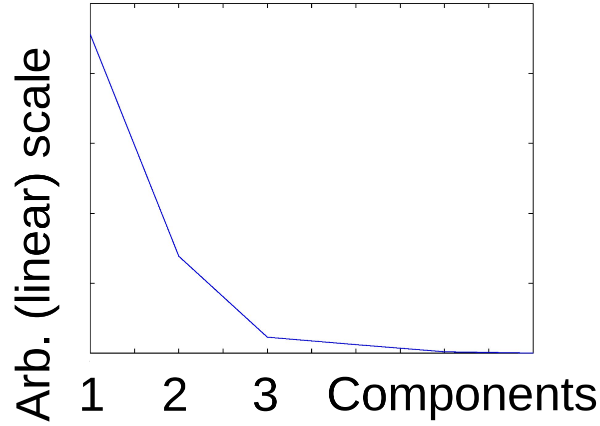

Saad, …, Chelikowsky, and Andreoni, PRB 85, 104104 (2012)

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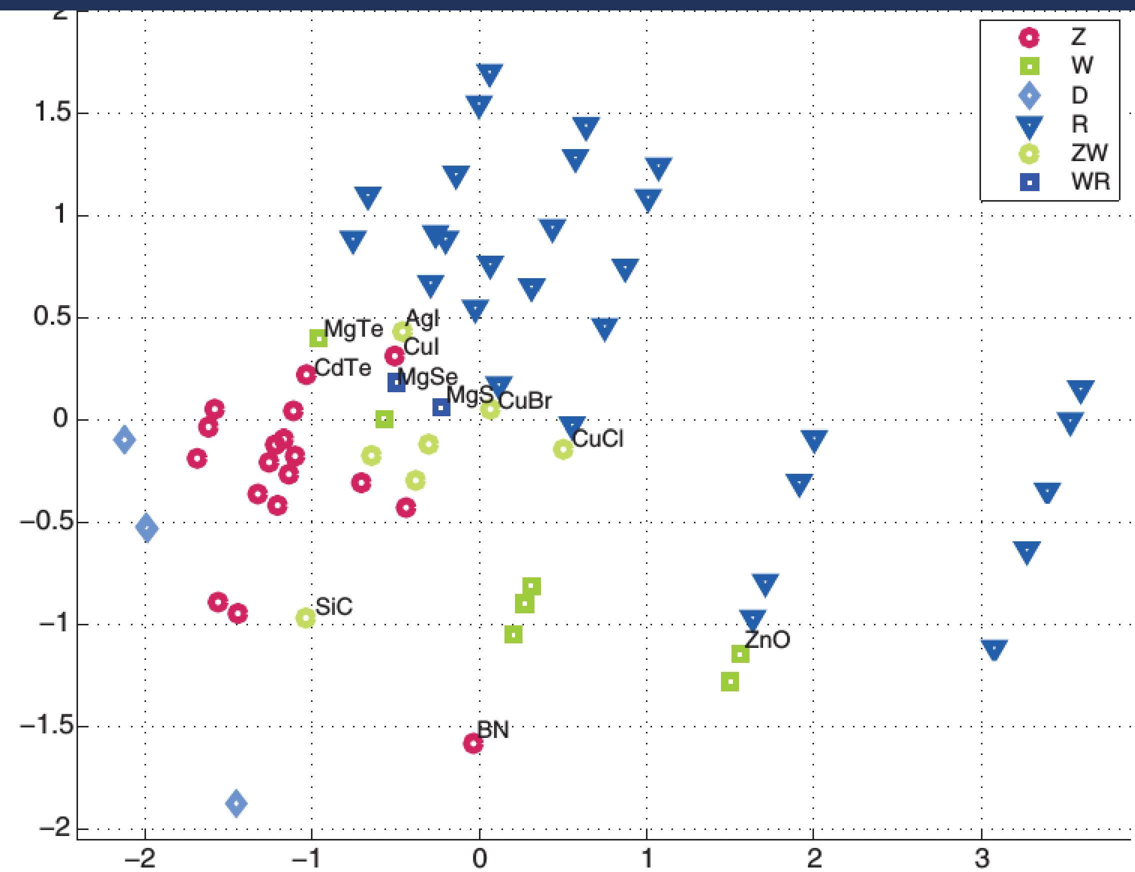

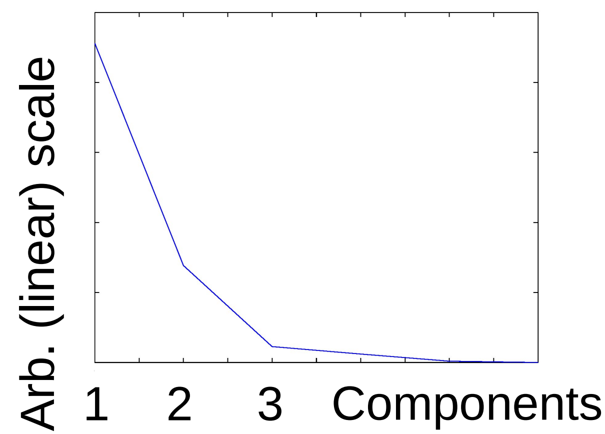

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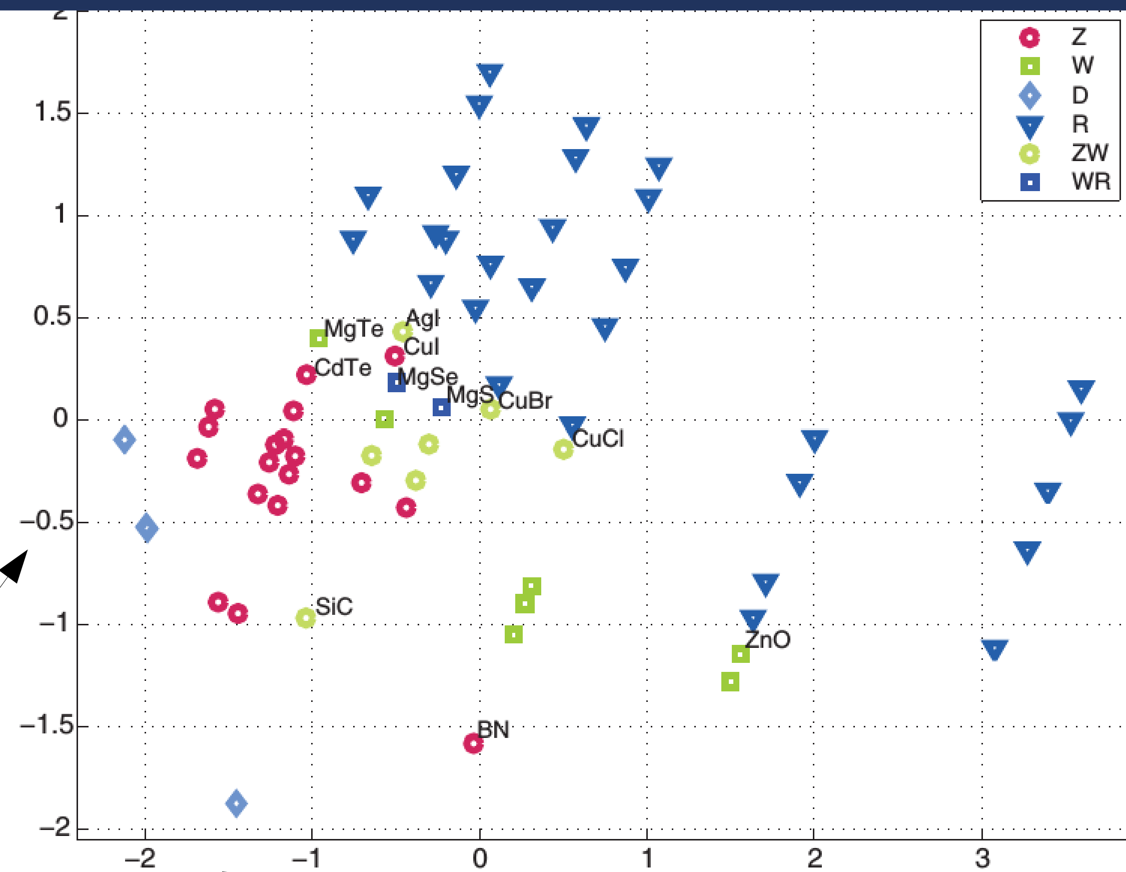
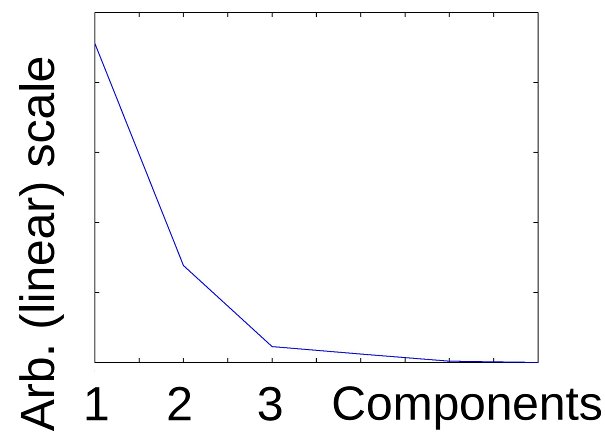


What's on the axes?

Linear combination of (possibly all) the initial dimensions



Saad, …, Chelikowsky, and Andreoni, PRB 85, 104104 (2012)

Compressed sensing: the quest for descriptors and predictive models

82 octet AB binary compounds

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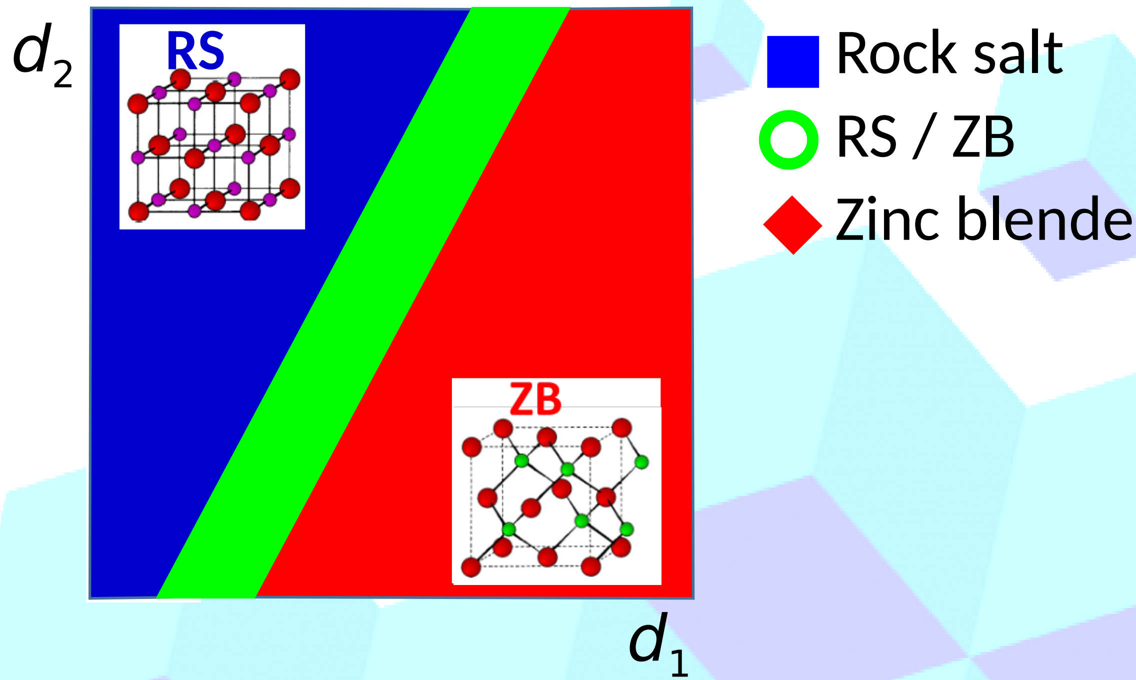
- HOMO
- LUMO
- Ionization Potential
- Electron Affinity
- Radius of valence *s* orbital
- Radius of valence *p* orbital
- Radius of valence *d* orbital
- Thousands to billions of non-linear functions of the above



$$\boldsymbol{P} = c_1\boldsymbol{d}_1 + c_2\boldsymbol{d}_2 + \dots c_n\boldsymbol{d}_n$$

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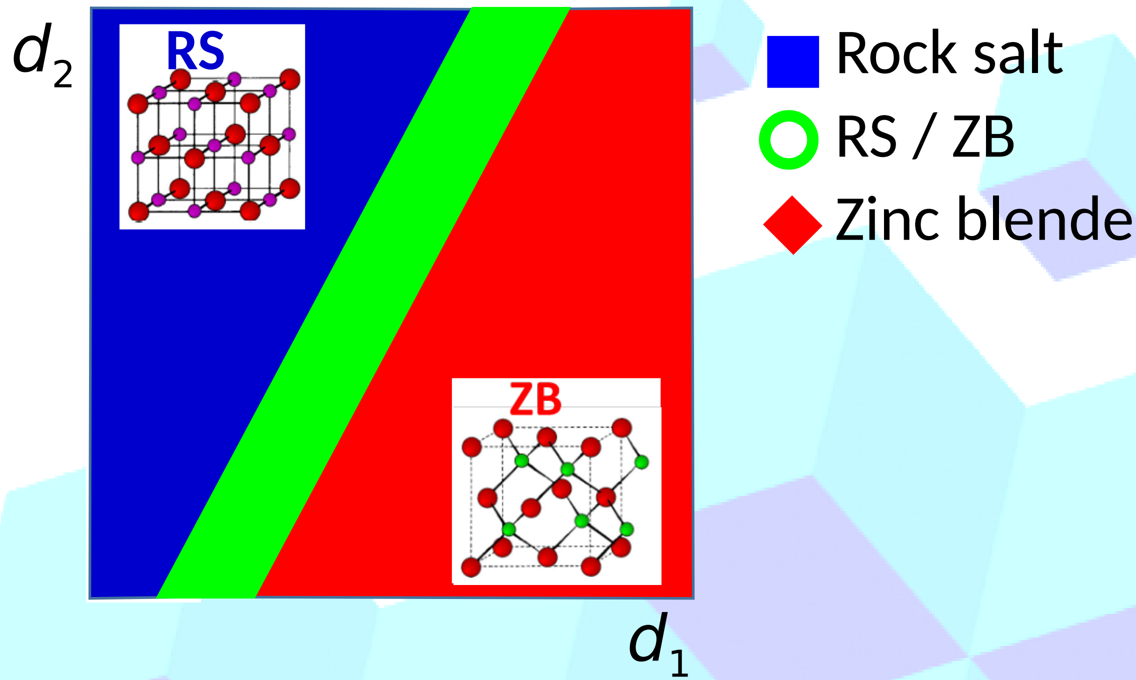
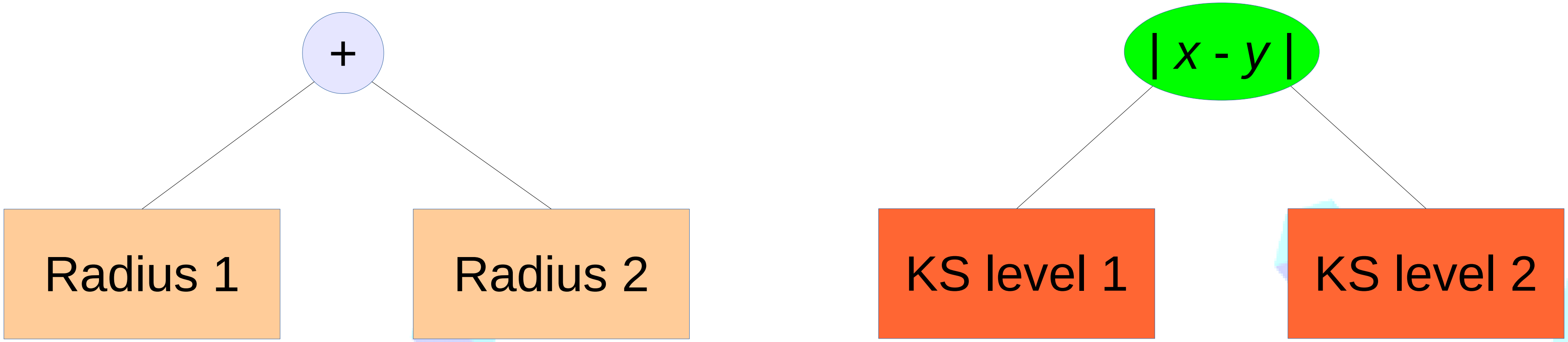


Symbolic Regression

$\boldsymbol{P} = c_1\boldsymbol{d}_1 + c_2\boldsymbol{d}_2 + \ldots\, c_n\boldsymbol{d}_n$

E(Rock salt) – *E*(Zinc blende)

Systematic construction of the feature space



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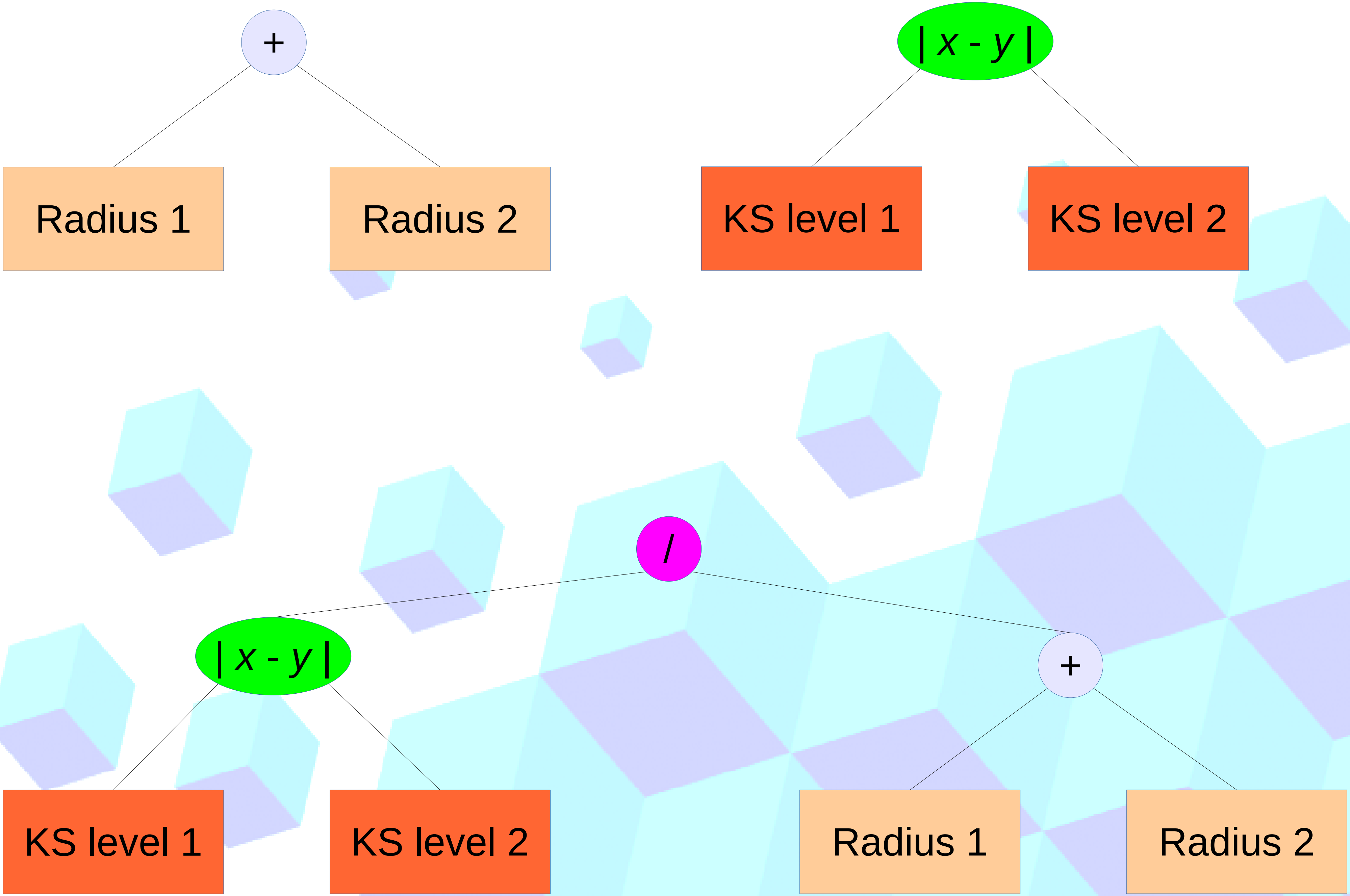
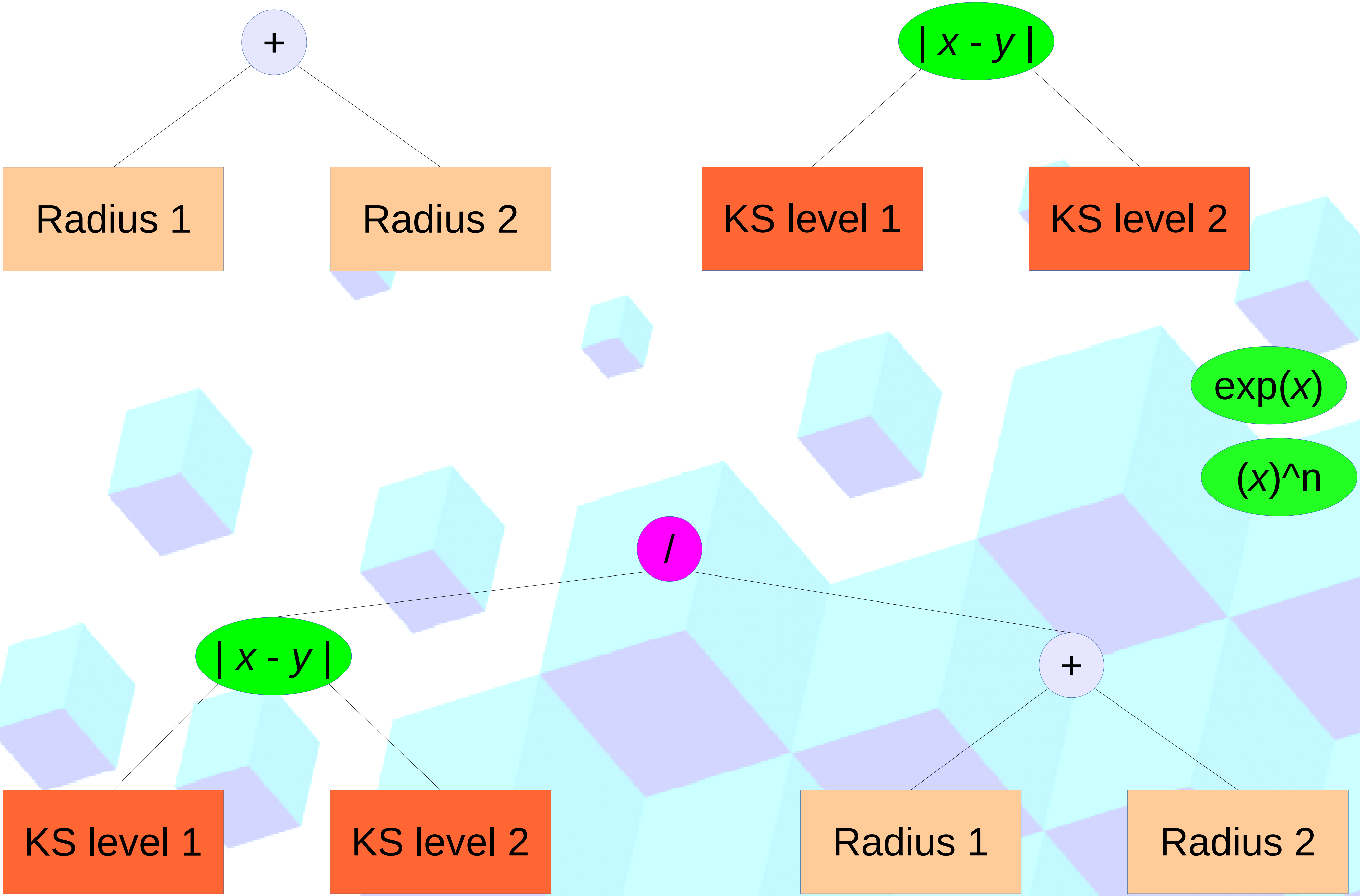


Systematic construction of the feature space



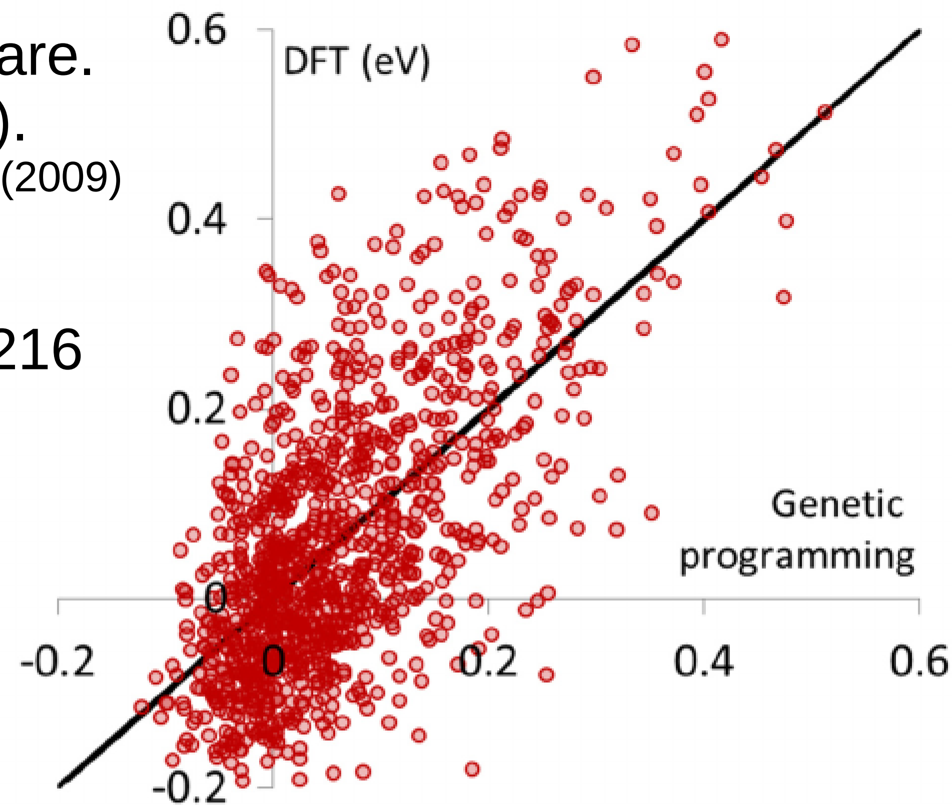
Systematic construction of the feature space: EUREQA

EUREQA: genetic programming software.
Global optimization (genetic algorithm).
Schmidt M., Lipson H., Science, Vol. 324, No. 5923, (2009)

T. Müller et al. PRB **89** 115202 (2014):
Data: ~1000 amorphous structures of 216 Si atoms (saturated)

Property: hole trap depth

$$\frac{\min(1.66355,a)\max(5.37551,c)-f-bd}{g} - h\max(3.42929,e),$$



Descriptor (candidates: 242)

a The largest distance between a H atom and its nearest Si neighbor
b The shortest distance between a Si atom and its sixth-nearest Si neighbor
c The maximum bond valence sum on a Si atom
d The smallest value for the fifth-smallest relative bond length around a Si atom
e The fourth-shortest distance between a Si atom and its eighth-nearest neighbor
f The second-shortest distance between a Si atom and its fifth-nearest neighbor
g The third-shortest distance between a Si atom and its sixth-nearest neighbor
h The H-Si nearest-neighbor distance for the hydrogen atom with the fourth-smallest difference between the distances to the two Si atoms nearest to a H atom

Building block
Constant value
Input variable
Addition
Subtraction
Multiplication
Division
Negation
Exponential
Natural logarithm
Power
Square root
Logistic function
Minimum
Maximum
Absolute value

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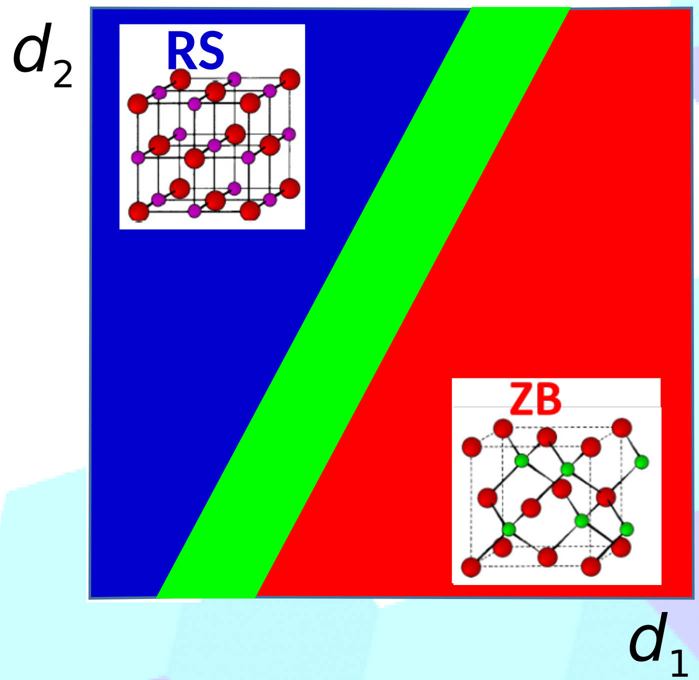

- Rock salt
- RS / ZB
- Zinc blende

$$\boldsymbol{P} = c_1\boldsymbol{d}_1 + c_2\boldsymbol{d}_2 + \ldots c_n\boldsymbol{d}_n$$

$$\underset{\boldsymbol{c}\in\mathbb{R}^M}{\operatorname{argmin}} \|\boldsymbol{P} - \boldsymbol{D}\boldsymbol{c}\|_2^2 + \lambda\|\boldsymbol{c}\|_0$$

E(Rock salt) – *E*(Zinc blende)

Compressed sensing: the quest for descriptors and predictive models

Ideal method: regression with ℓ_0 regularization

$$\underset{\boldsymbol{c}\in\mathbb{R}^M}{\operatorname{argmin}}\left(\|\boldsymbol{P}-\boldsymbol{D}\boldsymbol{c}\|_2^2+\lambda\|\boldsymbol{c}\|_0\right)$$

Optimal solution
Non-polinomial complexity
Small # columns in $\boldsymbol{D}$

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Optimal solution
Non-polinomial complexity
Small # columns in ***D***

$\|\boldsymbol{c}\|_0$	# of nonzero elements of ***c***
$\|\boldsymbol{c}\|_2$	Euclidean. Square root of sum of squares of the elements of ***c***)

Compressed sensing: the quest for descriptors and predictive models

Ideal method: regression with ℓ_0 regularization

$$\underset{c \in \mathbb{R}^M}{\operatorname{argmin}} \left(\|P - Dc\|_2^2 + \lambda \|c\|_0 \right)$$

Optimal solution
Non-polinomial complexity
Small # columns in ***D***

$\|c\|_0$	# of nonzero elements of ***c***
$\|c\|_2$	Euclidean. Square root of sum of squares of the elements of ***c***)

For matrices D with uncorrelated columns: LASSO

$$\underset{c \in \mathbb{R}^M}{\operatorname{argmin}} \|P - Dc\|_2^2 + \lambda \|c\|_1$$

(Possibly) optimal solution
Convex optimization
Moderate # columns in ***D***

$\|c\|_1$	"Manhattan". Sum of absolute values of the elements of ***c***

Compressed sensing in materials science

PRL 113, 185501 (2014) PHYSICAL REVIEW LETTERS week ending 31 OCTOBER 2014

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

Fei Zhou (周非)
Physical and Life Sciences Directorate, Lawrence Livermore National Laboratory, Livermore, California 94550, USA

Weston Nielson, Yi Xia, and Vidvuds Ozoliņš
Department of Materials Science and Engineering, University of California, Los Angeles, California 90095-1595, USA

(Received 22 April 2014; published 27 October 2014)

PNAS

Compressed modes for variational problems in mathematics and physics

Vidvuds Ozoliņš[a,1], Rongjie Lai[b,1], Russel Caflisch[c,1], and Stanley Osher[c,1,2]

Departments of [a]Materials Science and Engineering, and [c]Mathematics, University of California, Los Angeles, CA 90095-1555; and [b]Department of Mathematics, University of California, Irvine, CA 92697-3875

Contributed by Stanley Osher, October 8, 2013 (sent for review September 3, 2013)

PHYSICAL REVIEW B **87**, 035125 (2013)

Compressive sensing as a paradigm for building physics models

Lance J. Nelson and Gus L. W. Hart
Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84602, USA

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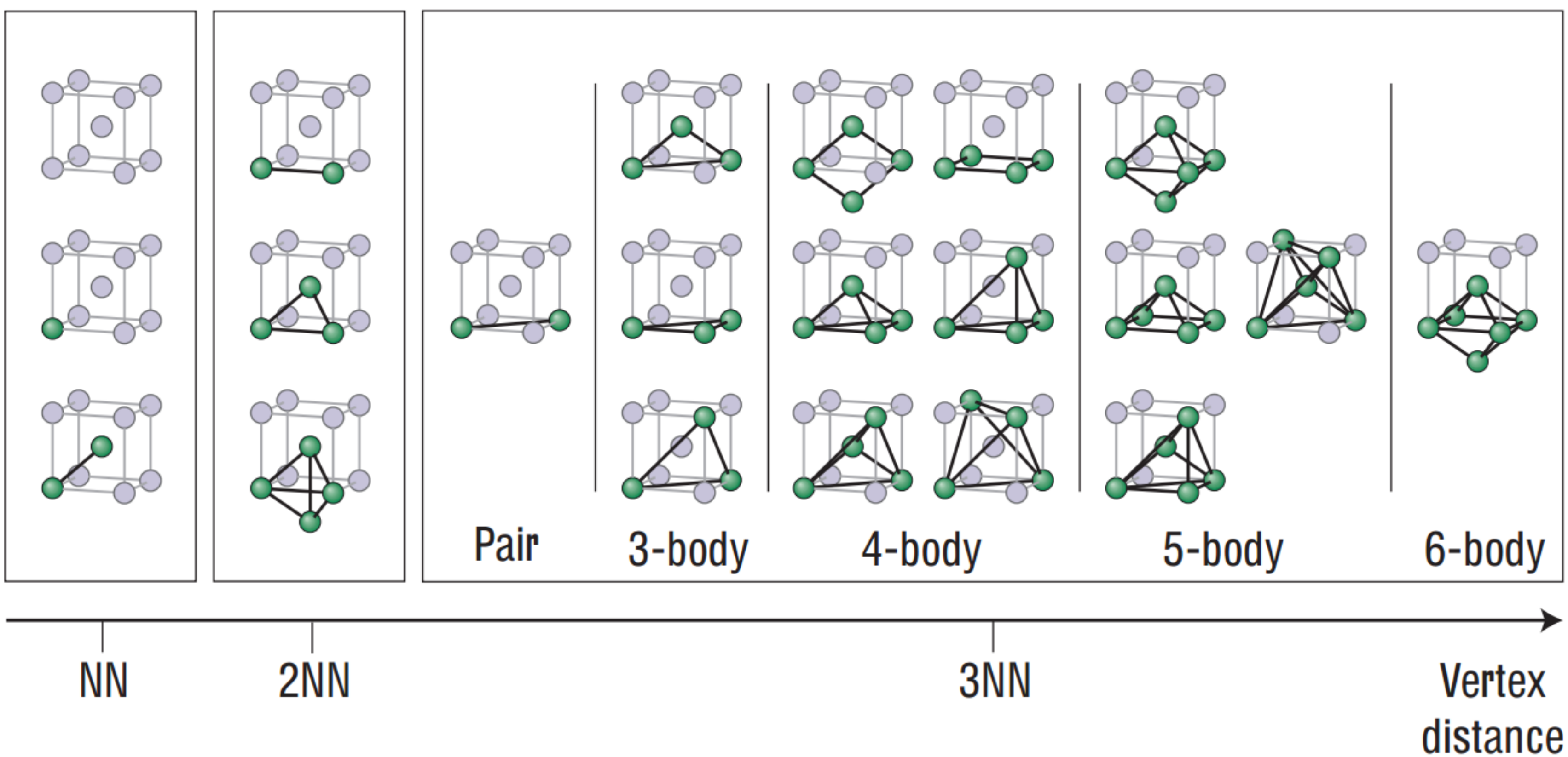
$$E(\sigma) = E_0 + \sum_f \bar{\Pi}_f(\sigma) J_f$$



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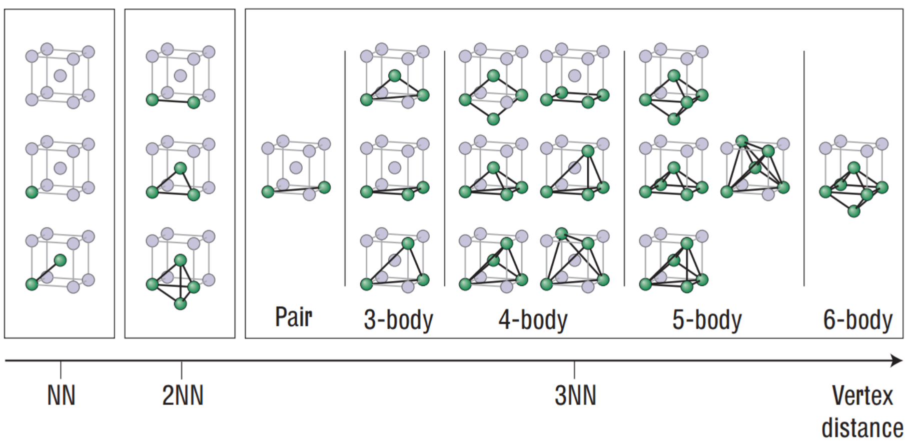
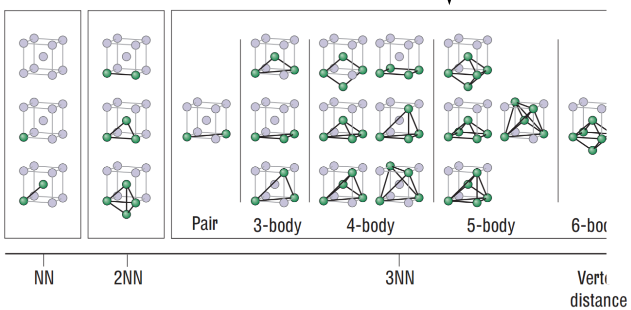


$$\min_u \mu \|\vec{u}\|_1 + \frac{1}{2}\|\mathbb{A}\vec{u} - \vec{f}\|^2$$

Bregman Iteration

$$\boxed{\begin{aligned} \vec{f}^{k+1} &= \vec{f} + (\vec{f}^k - \mathbb{A}\vec{u}^k), \\ \vec{u}^{k+1} &= \arg\min_u \mu\|\vec{u}\|_1 + \frac{1}{2}\|\mathbb{A}\vec{u} - \vec{f}^{k+1}\|^2, \end{aligned}}$$

$$E(\sigma) = E_0 + \sum_f \bar{\Pi}_f(\sigma) J_f$$



PHYSICAL REVIEW B **87**, 035125 (2013)

Compressive sensing as a paradigm for building physics models

Lance J. Nelson and Gus L. W. Hart
Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84602, USA

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(Received 26 June 2012; revised manuscript received 26 September 2012; published 18 January 2013)

$$\min_u \mu \|\vec{u}\|_1 + \tfrac{1}{2}\|\mathbb{A}\vec{u} - \vec{f}\|^2$$

Bregman Iteration

$$\vec{u} = \arg\min_{u,d} \|\vec{d}\|_1 + \tfrac{1}{2}\|\mathbb{A}\vec{u} - \vec{f}\|^2 + \tfrac{\lambda}{2}\|\vec{d} - \mu\vec{u}\|^2$$

$$\boxed{\begin{aligned} \vec{f}^{k+1} &= \vec{f} + (\vec{f}^k - \mathbb{A}\vec{u}^k), \\ \vec{u}^{k+1} &= \arg\min_u \mu\|\vec{u}\|_1 + \tfrac{1}{2}\|\mathbb{A}\vec{u} - \vec{f}^{k+1}\|^2, \end{aligned}}$$

Split Bregman Iteration

$$\boxed{\begin{aligned} \vec{u}^{k+1} &= \arg\min_u \tfrac{1}{2}\|\mathbb{A}\vec{u} - \vec{f}\|^2 + \tfrac{\lambda}{2}\|\vec{d}^k - \mu\vec{u} - \vec{b}^k\|^2, \\ \vec{d}^{k+1} &= \arg\min_d \|\vec{d}\|_1 + \tfrac{\lambda}{2}\|\vec{d} - \mu\vec{u}^{k+1} - \vec{b}^k\|^2, \\ \vec{b}^{k+1} &= \vec{b}^k + \mu\vec{u}^{k+1} - \vec{d}^{k+1}, \end{aligned}}$$

Compressed modes for variational problems in mathematics and physics

Vidvuds Ozoliņš[a,1], Rongjie Lai[b,1], Russel Caflisch[c,1], and Stanley Osher[c,1,2]

Departments of [a]Materials Science and Engineering, and [c]Mathematics, University of California, Los Angeles, CA 90095-1555; and [b]Department of Mathematics, University of California, Irvine, CA 92697-3875

Contributed by Stanley Osher, October 8, 2013 (sent for review September 3, 2013)

$$E_0 = \min_{\Phi_N} \sum_{j=1}^{N} \left\langle \phi_j, \hat{H}\phi_j \right\rangle \quad \text{s.t.} \quad \left\langle \phi_j, \phi_k \right\rangle = \delta_{jk}.$$

$$W_j(\mathbf{x}) = \sum_k U_{jk}\phi_k(\mathbf{x})$$

$$\left\langle \Delta \mathbf{x}_j^2 \right\rangle = \left\langle W_j, \left(\mathbf{x} - \langle \mathbf{x}_j \rangle\right)^2 W_j \right\rangle \qquad \langle \mathbf{x}_j \rangle = \langle W_j, \mathbf{x} W_j \rangle$$

Parameter free maximally localised Wannier functions?

$$E = \min_{\Psi_N} \sum_{j=1}^{N} \left(\frac{1}{\mu} \left|\psi_j\right|_1 + \left\langle \psi_j, \hat{H}\psi_j \right\rangle \right) \quad \text{s.t.} \quad \left\langle \psi_j, \psi_k \right\rangle = \delta_{jk}$$

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

Fei Zhou (周非)
Physical and Life Sciences Directorate, Lawrence Livermore National Laboratory, Livermore, California 94550, USA

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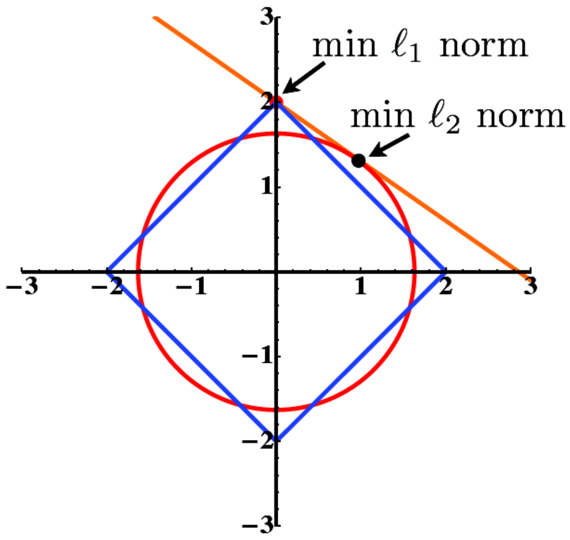
Department of Materials Science and Engineering, University of California, Los Angeles, California 90095-1595, USA
(Received 22 April 2014; published 27 October 2014)

$$V = V_0 + \Phi_{\mathbf{a}} u_{\mathbf{a}} + \frac{\Phi_{\mathbf{ab}}}{2} u_{\mathbf{a}} u_{\mathbf{b}} + \frac{\Phi_{\mathbf{abc}}}{3!} u_{\mathbf{a}} u_{\mathbf{b}} u_{\mathbf{c}} + \cdots,$$

$$\Phi_{\mathbf{ab}} \equiv \Phi_{ij}(ab) = \partial^2 V / \partial u_{\mathbf{a}} \partial u_{\mathbf{b}}$$
$$\Phi_{\mathbf{abc}} \equiv \Phi_{ijk}(abc) = \partial^3 V / \partial u_{\mathbf{a}} \partial u_{\mathbf{b}} \partial u_{\mathbf{c}}$$

$$\begin{aligned} \Phi^{\mathrm{CS}} &= \arg\min_{\Phi} \|\Phi\|_1 + \frac{\mu}{2} \|\mathbf{F} - \mathbb{A}\Phi\|_2^2 \\ &= \arg\min_{\Phi} \sum_I |\Phi_I| + \frac{\mu}{2} \sum_{ai} (F_{ai} - A_{ai,J}\Phi_J)^2 \end{aligned}$$

Compressed sensing: the quest for descriptors and predictive models

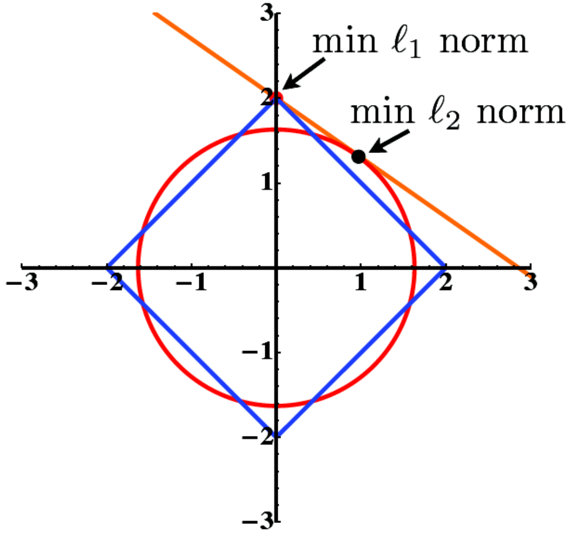


$$\underset{\boldsymbol{c}\in\mathbb{R}^M}{\operatorname{argmin}} \|\boldsymbol{P} - \boldsymbol{D}\boldsymbol{c}\|_2^2 + \lambda\|\boldsymbol{c}\|_1$$

(Possibly) optimal solution
Convex optimization
Moderate # columns in ***D***

$\|\boldsymbol{c}\|_1$ "Manhattan". Sum of absolute values of the elements of ***c***

Compressed sensing: the quest for descriptors and predictive models



When there are high correlations, LASSO+ℓ_0 (LMG *et al*. PRL 2015):
- use LASSO with lambda in order to "switch on" few tens features (say 30-50)
- perform ℓ_0 regularization, i.e., for 1,2,3D solution, enumerate all 1- 2- 3-tuples and find the best fitting tuple.

$$\underset{\boldsymbol{c}\in\mathbb{R}^M}{\mathrm{argmin}} \|\boldsymbol{P} - \boldsymbol{D}\boldsymbol{c}\|_2^2 + \lambda\|\boldsymbol{c}\|_1$$

(Possibly) optimal solution
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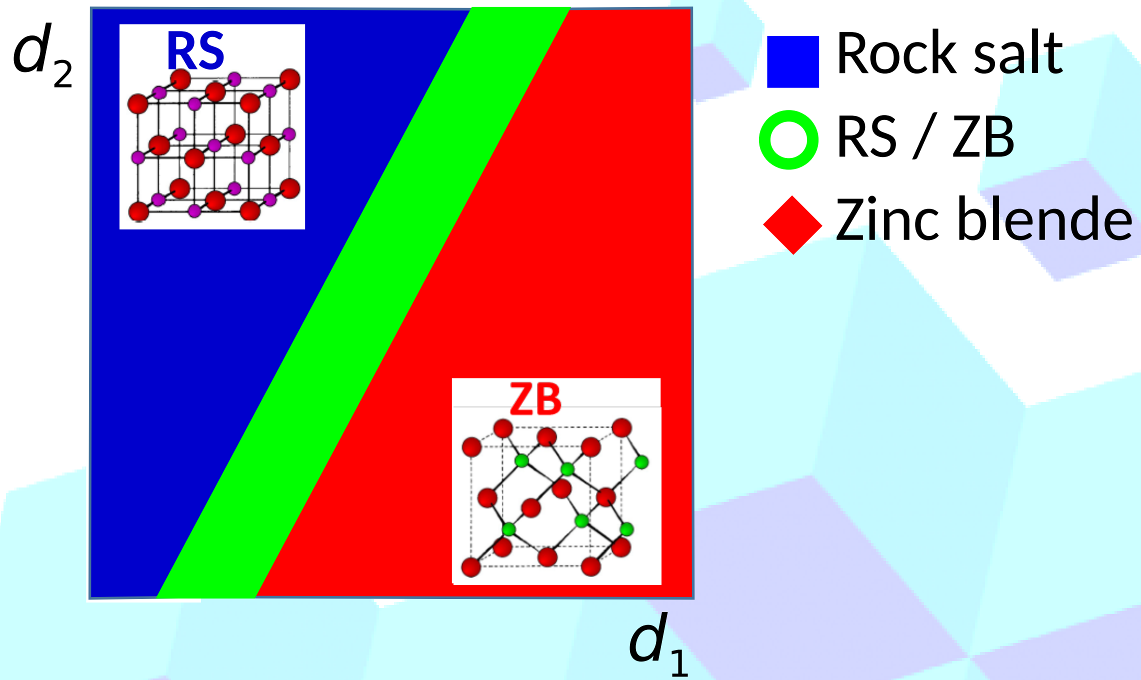
Compressed sensing: the quest for descriptors and predictive models

82 octet AB binary compounds

Ansatz: atomic features

- HOMO
- LUMO
- Ionization Potential
- Electron Affinity
- Radius of valence *s* orbital
- Radius of valence *p* orbital
- Radius of valence *d* orbital
- Billions of non-linear functions of the above



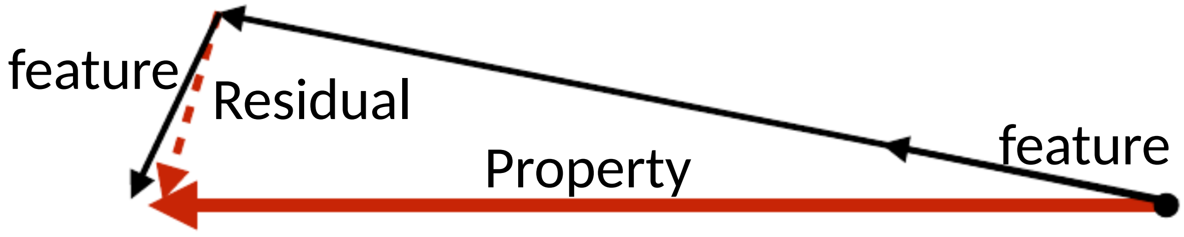
$$\boldsymbol{P} = c_1\boldsymbol{d}_1 + c_2\boldsymbol{d}_2 + \dots c_n\boldsymbol{d}_n$$

$$\underset{\boldsymbol{c}\in\mathbb{R}^M}{\operatorname{argmin}} \|\boldsymbol{P} - \boldsymbol{D}\boldsymbol{c}\|_2^2 + \lambda\|\boldsymbol{c}\|_0$$

E(Rock salt) – *E*(Zinc blende)

Compressed sensing: the quest for descriptors and predictive models

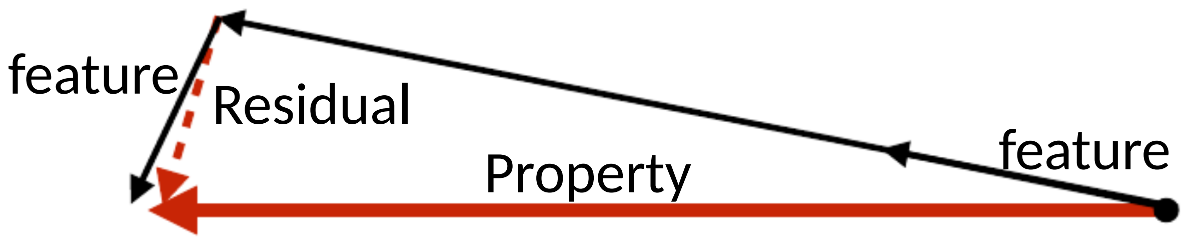
From orthogonal matching pursuit ….



Local-optimum solution
Huge # columns in D

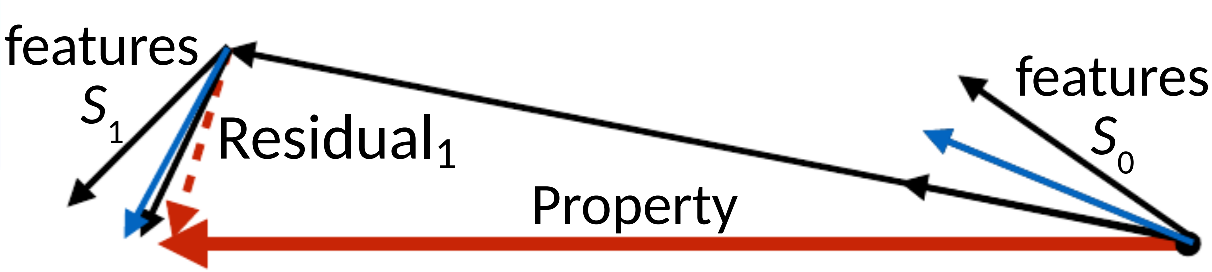
Compressed sensing: the quest for descriptors and predictive models

From orthogonal matching pursuit ....



Local-optimum solution
Huge # columns in D

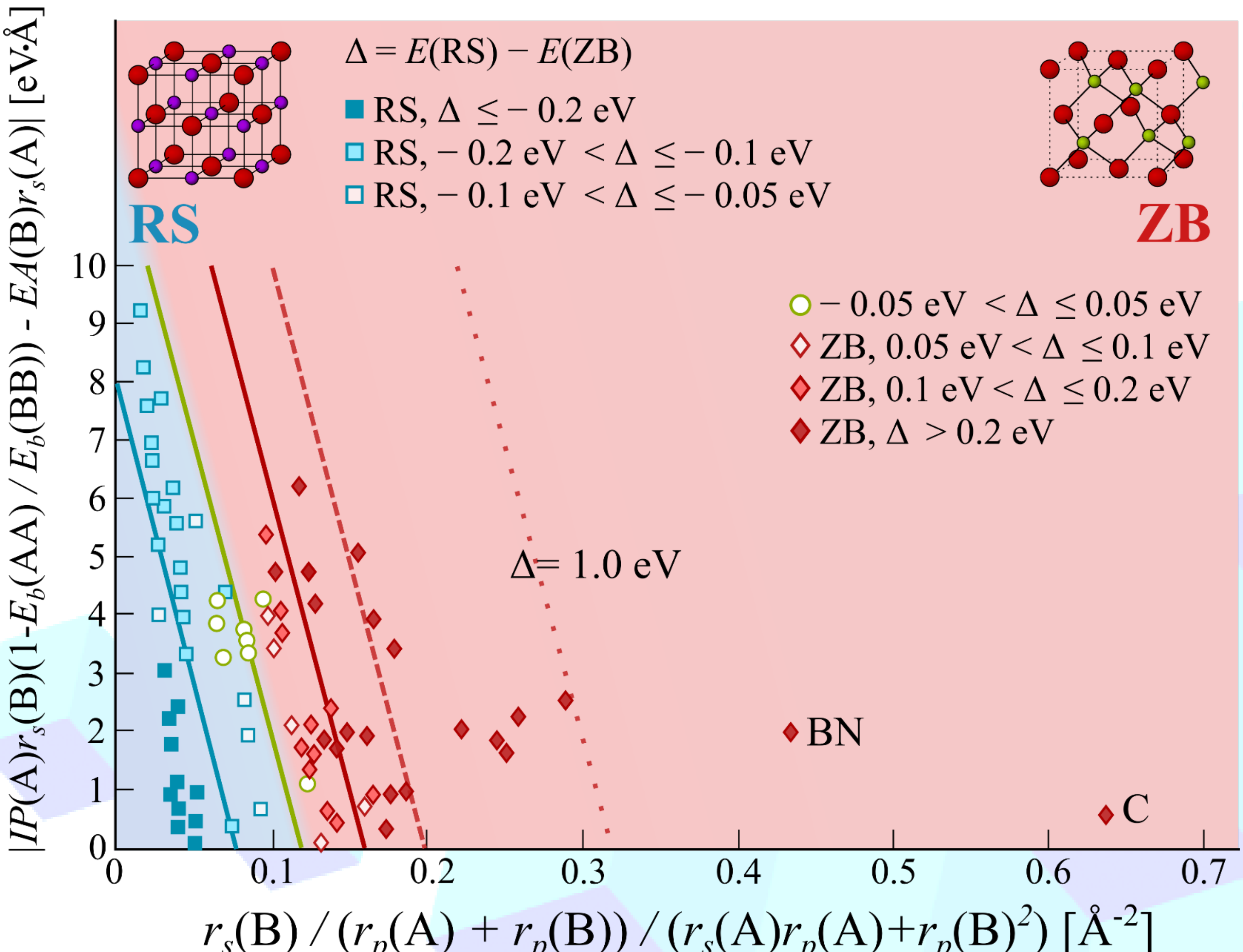
... to Sure Independence Screening + Sparsifying Operator (SISSO)



Proxy of
global-optimum solution
Huge # columns in D

R. Ouyang *et al.* PRM **2**, 083802 (2018), published 7 August 2018)

Compressed sensing: the quest for descriptors and predictive models



Structure map with SISSO, starting from 7 atomic + 6 dimer features
Feature space: 10^{11} features

R. Ouyang *et al.* PRM (2018)

One descriptor to rule them all: Multi-task learning

$$\{P^{(1)}, P^{(2)}, \ldots, P^{N^{\mathrm{T}}}\}$$

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$$\{P^{(1)}, P^{(2)}, \ldots, P^{N^{\mathrm{T}}}\} \Rightarrow P^k = \boldsymbol{d} \cdot \boldsymbol{c}^k$$

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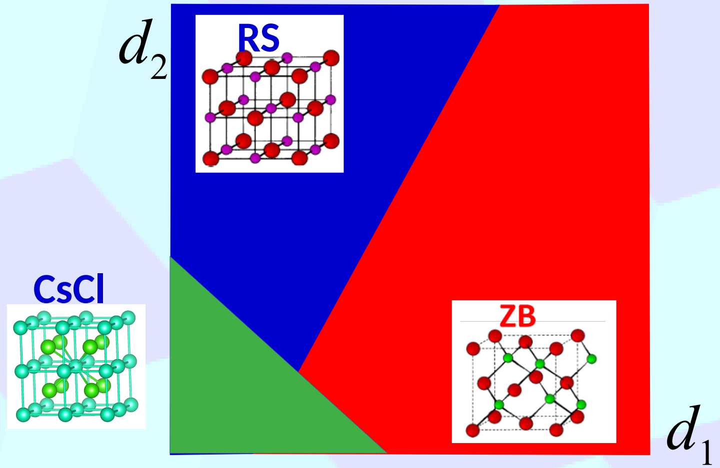
Application: multi-phase stability diagram
Properties: crystal-structure formation energies



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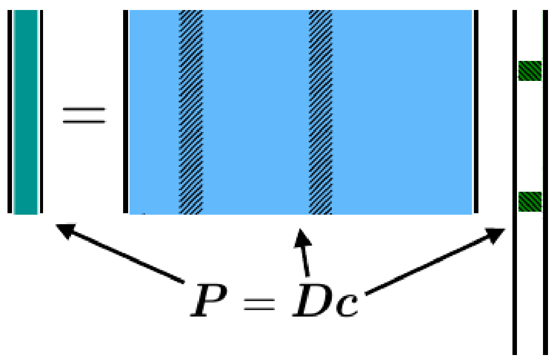
ST-SISSO



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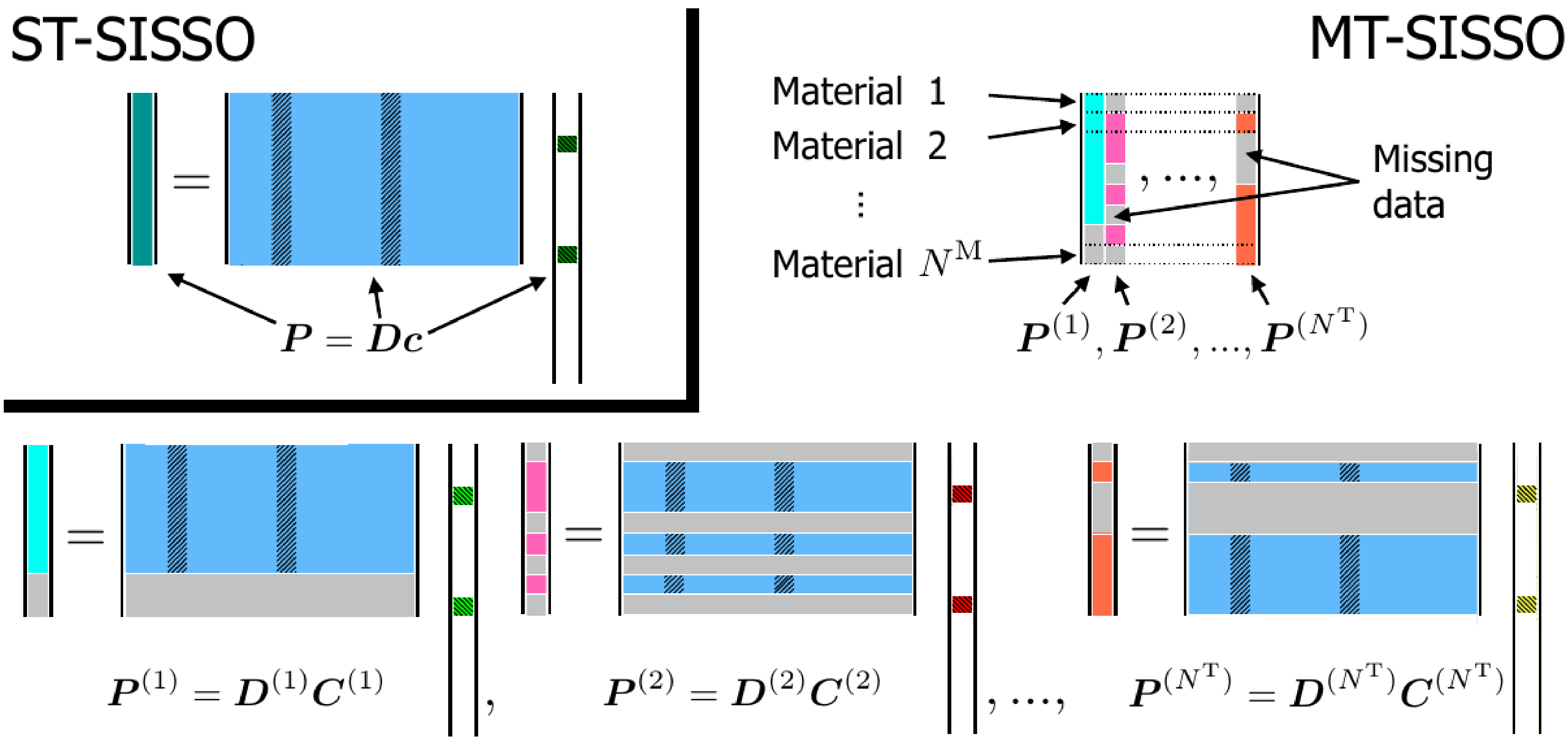

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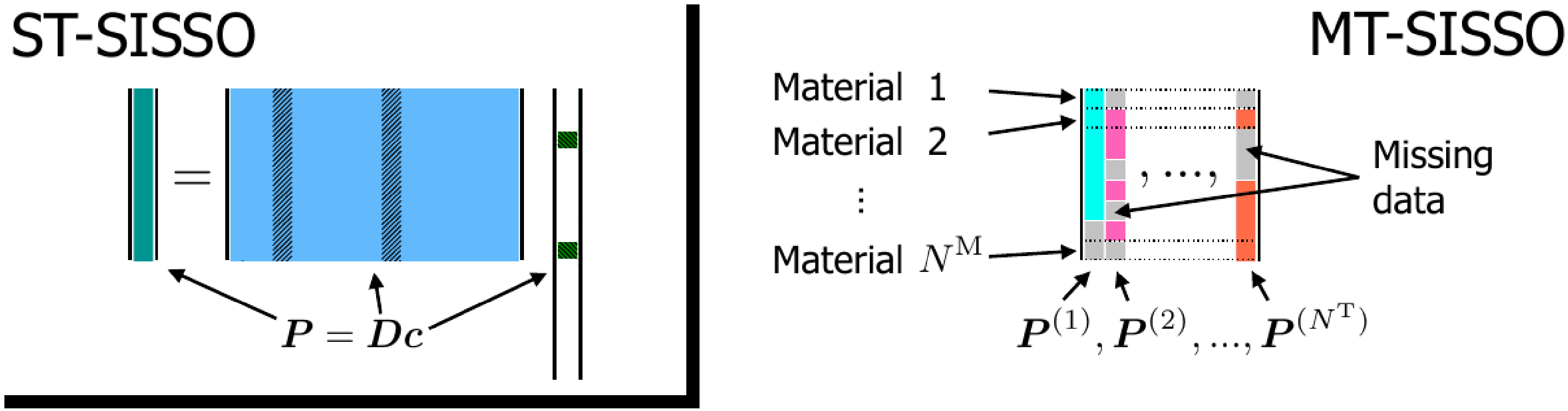

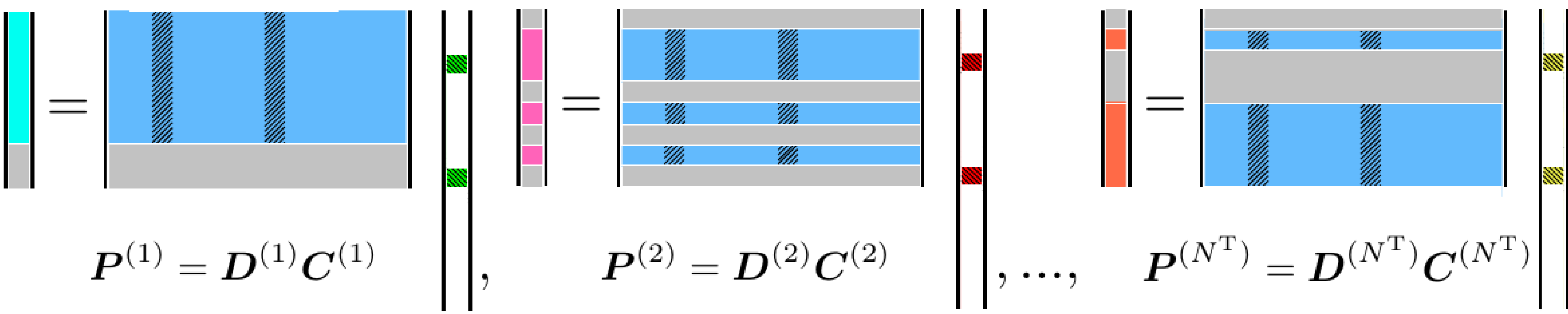

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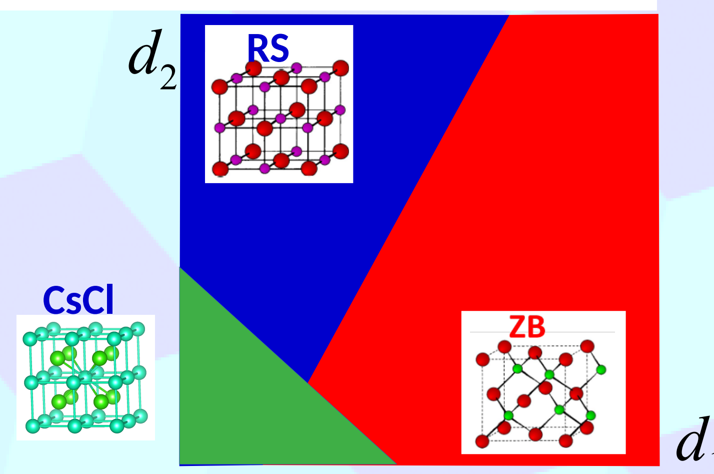

One descriptor to rule them all: Multi-task learning

$$\{P^{(1)}, P^{(2)}, \ldots, P^{N^{\mathrm{T}}}\} \longrightarrow P^k = \boldsymbol{d} \cdot \boldsymbol{c}^k$$

$$\arg\min_{\boldsymbol{c}} \left(\|\boldsymbol{P} - \boldsymbol{D}\boldsymbol{c}\|_2^2 + \lambda \|\boldsymbol{c}\|_0 \right)$$

$$\arg\min_{\boldsymbol{C}} \sum_{k=1}^{N^{\mathrm{T}}} \frac{1}{N_k^{\mathrm{M}}} \left\| \boldsymbol{P}^k - \boldsymbol{D}^k \boldsymbol{C}^k \right\|_2^2 + \lambda \left\| \boldsymbol{C} \right\|_0$$

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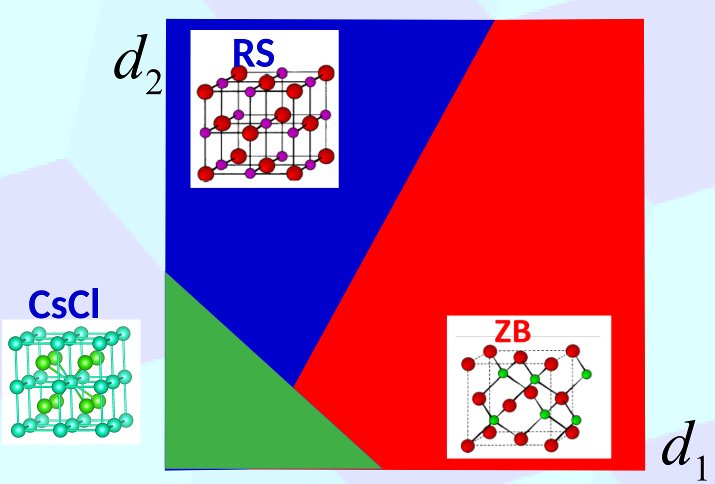
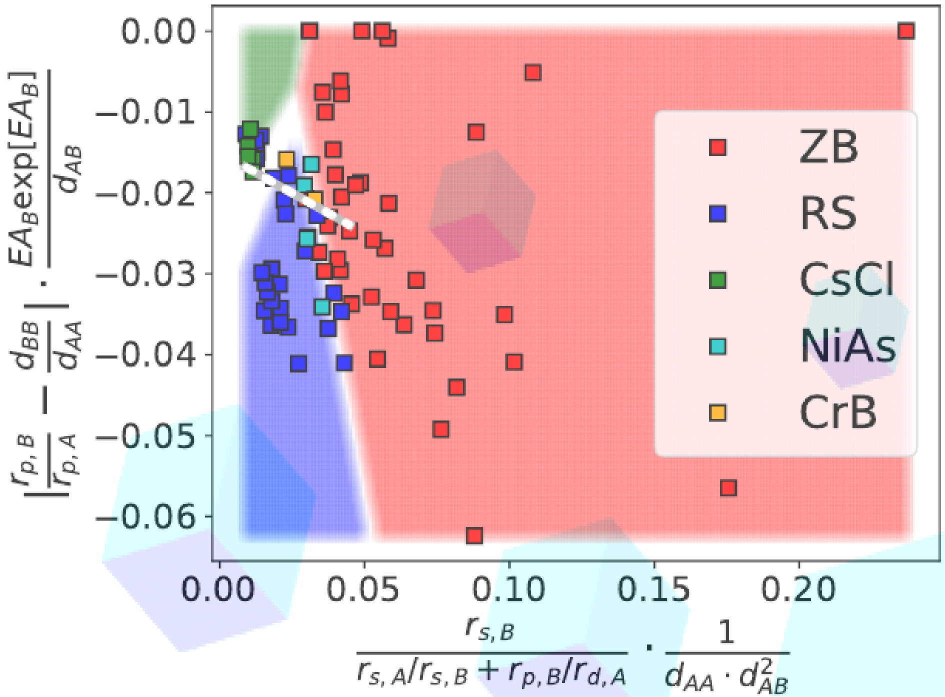
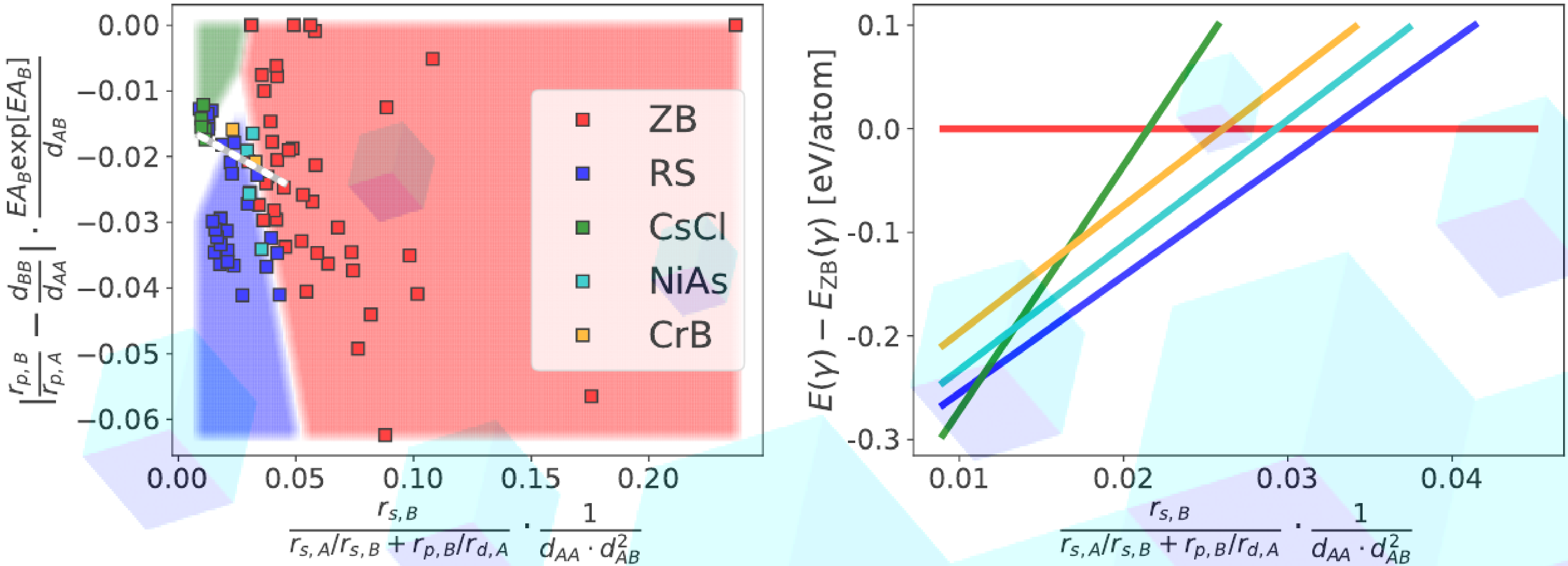


One descriptor to rule them all: Multi-task SISSO Energy differences among 5 crystal structures.



Ouyang, Ahmetcik, Carbogno, Scheffler & LMG, J. Phys. Mater. 2, 024002 (2019)

One descriptor to rule them all: Multi-task SISSO
Energy differences among 5 crystal structures.



Ouyang, Ahmetcik, Carbogno, Scheffler & LMG, J. Phys. Mater. 2, 024002 (2019)

A general scheme about training, cross-validation, test

Training: input (features, descriptor) + labels (values target property)
→ yields one model which minimizes a cost function (incl. regularization)

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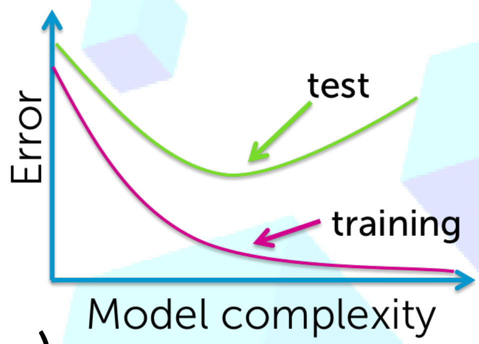

Cross-validation: used to tune model-complexity
- perform training *n* times on different split of data. Training + test/validation sets

→ yields one model that minimizes the test (validation) error

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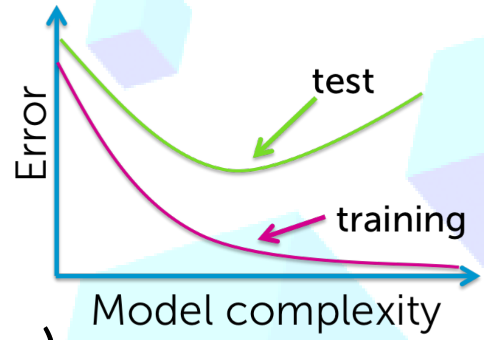

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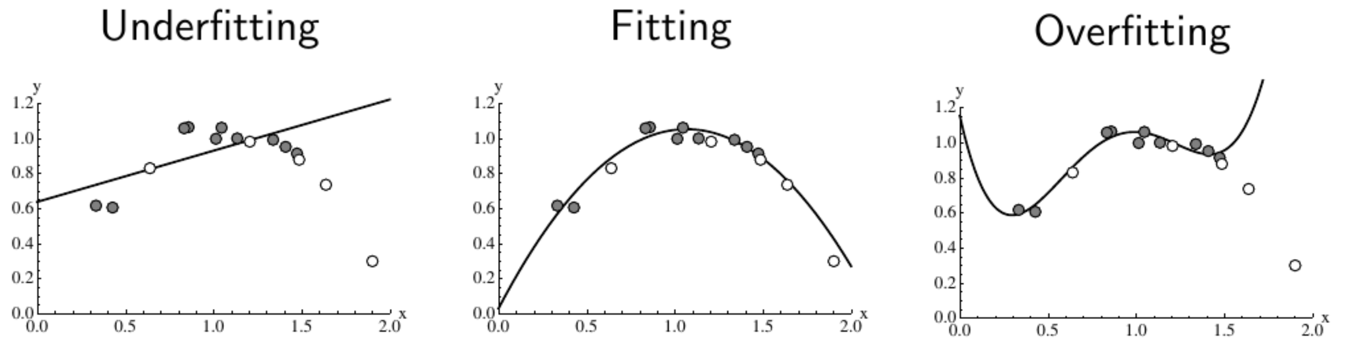

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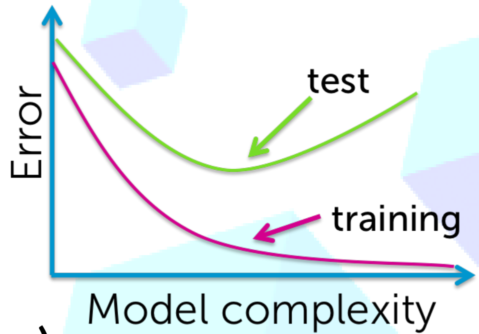

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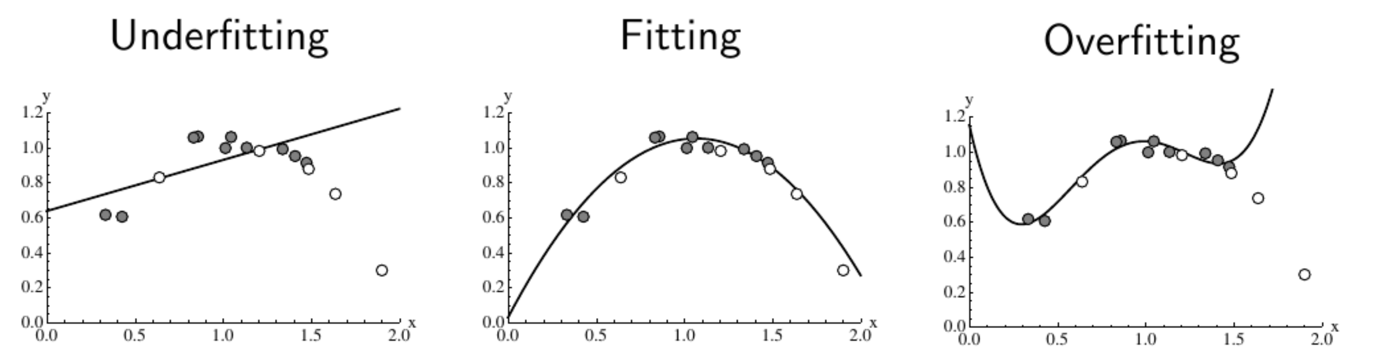

Test: evaluation of the performance of the model on data never used for training (i.e., the whole cross-validation procedure), aka left-out set

Data-driven model complexity

- In compressed sensing
  the “hyperparameters” are
  - the level of sparsity
  (optimal dimensionality of the
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  - the size of the feature space

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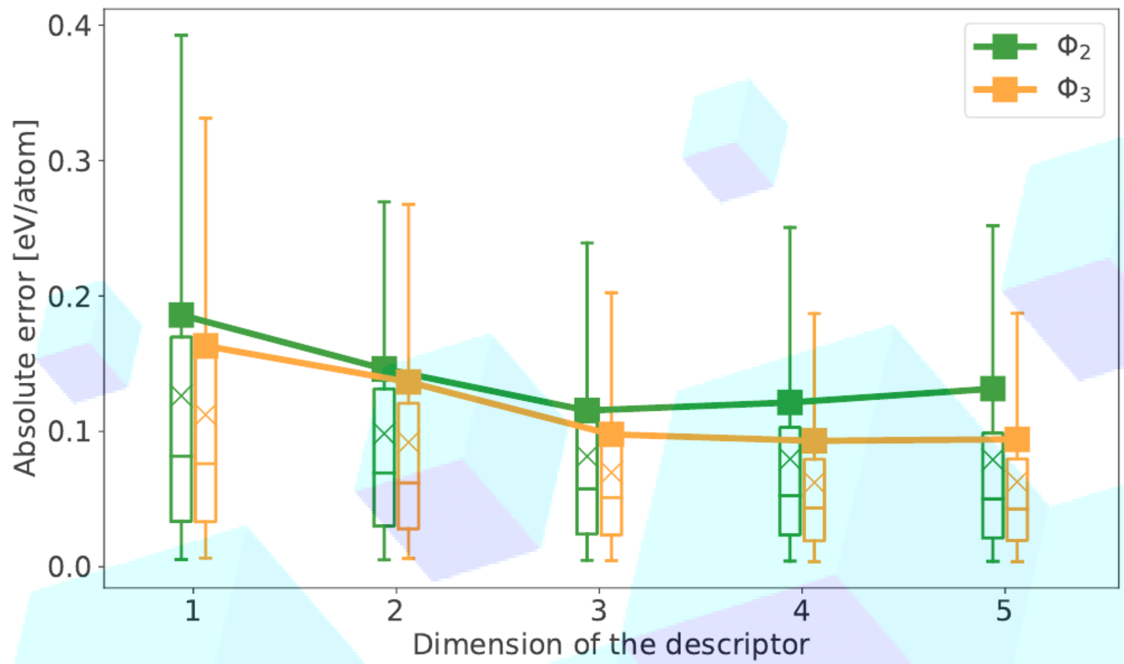
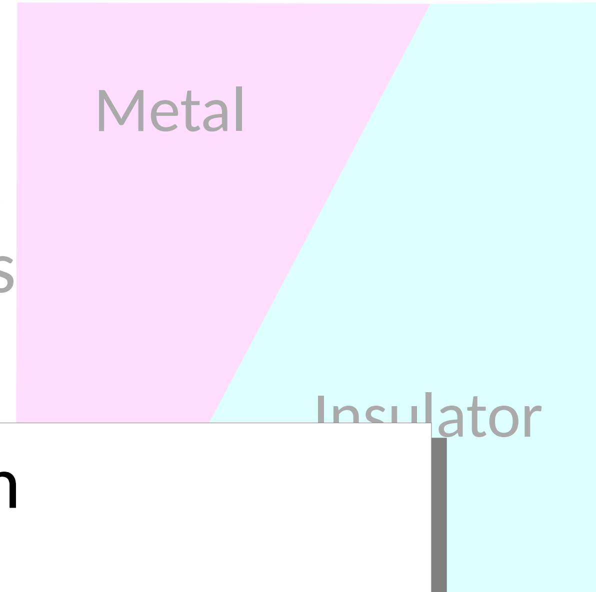


Ouyang, Ahmetcik, Carbogno, Scheffler & LMG, J. Phys. Mater. 2, 024002 (2019)

Charts/maps of materials

$$\underset{\boldsymbol{c}\in\mathbb{R}^M}{\operatorname{argmin}}\left(\underbrace{\|\boldsymbol{P}-\boldsymbol{D}\boldsymbol{c}\|_2^2}\ +\ \lambda\|\boldsymbol{c}\|_0\right)$$

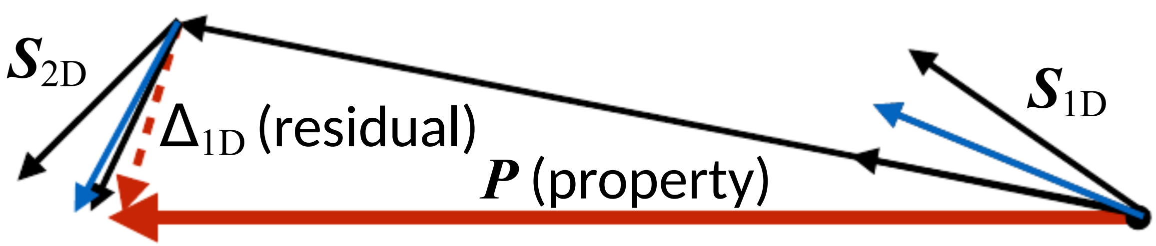
New cost function to be minimized: overlap of *convex* domains



1. # points in the *convex* overlap domain
2. Area of the domain overlap
3. Distance between domains

Good also for multi-categorical problems
(see A. F. Bialon *et al*., Chem. Mater. **28**, 2550 (2016))

Iterative generation of feature subspaces



SISO: metal/nonmetal classification of binary materials

Challenge:

Given the formula A_xB_y of a binary material AND its crystal structure, **is it a metal or a nonmetal**?

Dataset:

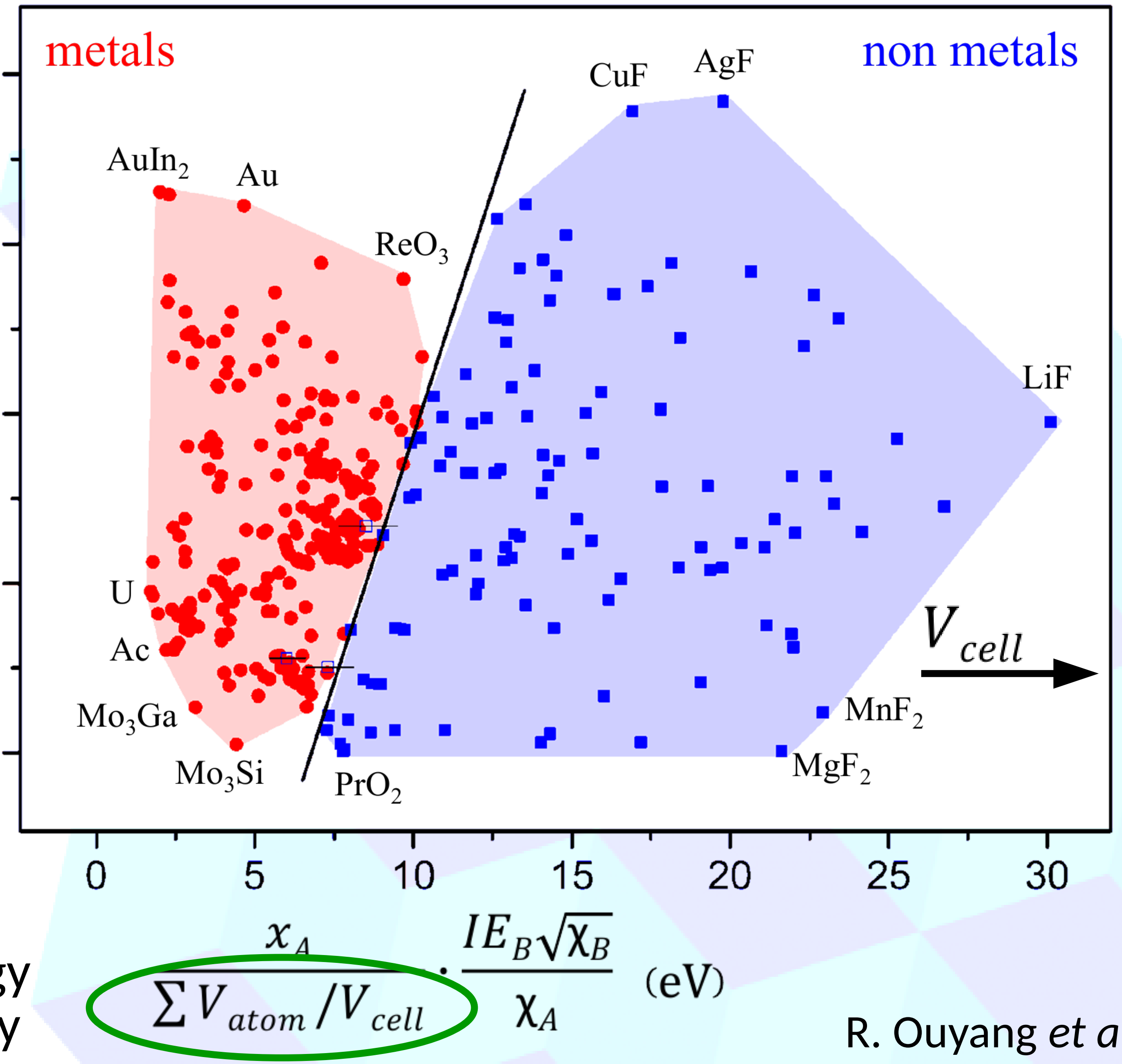
~300 materials from *Springer Materials*

- B is a p-block element, A any element
- 3D materials (i.e., not layered)
- At least one 1st neighbor of A(B) is B(A)
  (i.e., no materials containing "clusters" of A and/or B)

Classification AND primary features from *experiments*:

- ionization energy, electron affinity,
- (Pauling) electronegativity,
- covalent radius,
- valence, atomic fraction,
- AB interatomic distance,
- cell volume normalized by the sum of atomic volumes

SISO: metal/nonmetal classification of binary materials



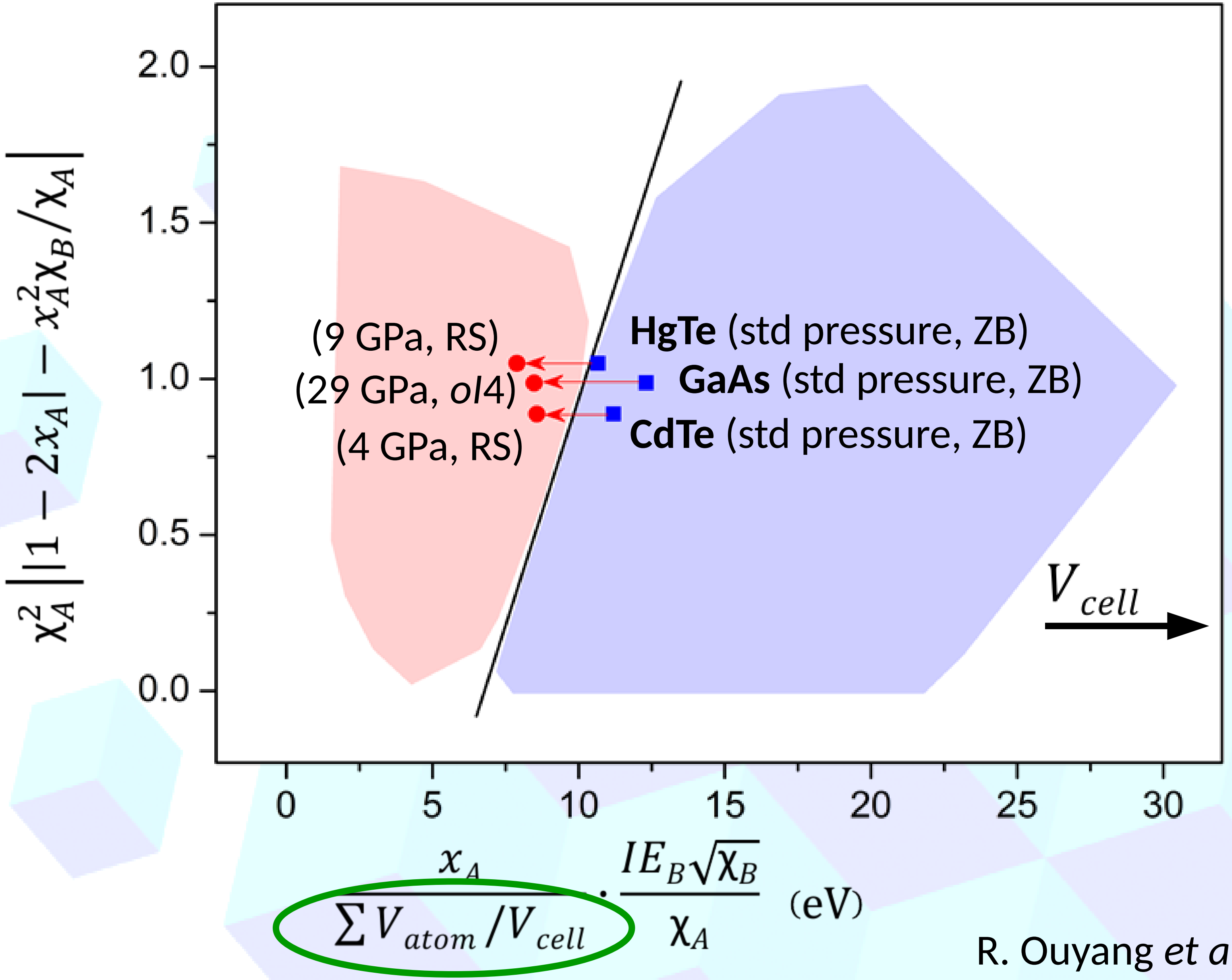
x Atomic fraction

IE Ionization energy

χ Electronegativity

R. Ouyang *et al.* PRM (2018)

SISSO: metal/nonmetal classification of binary materials



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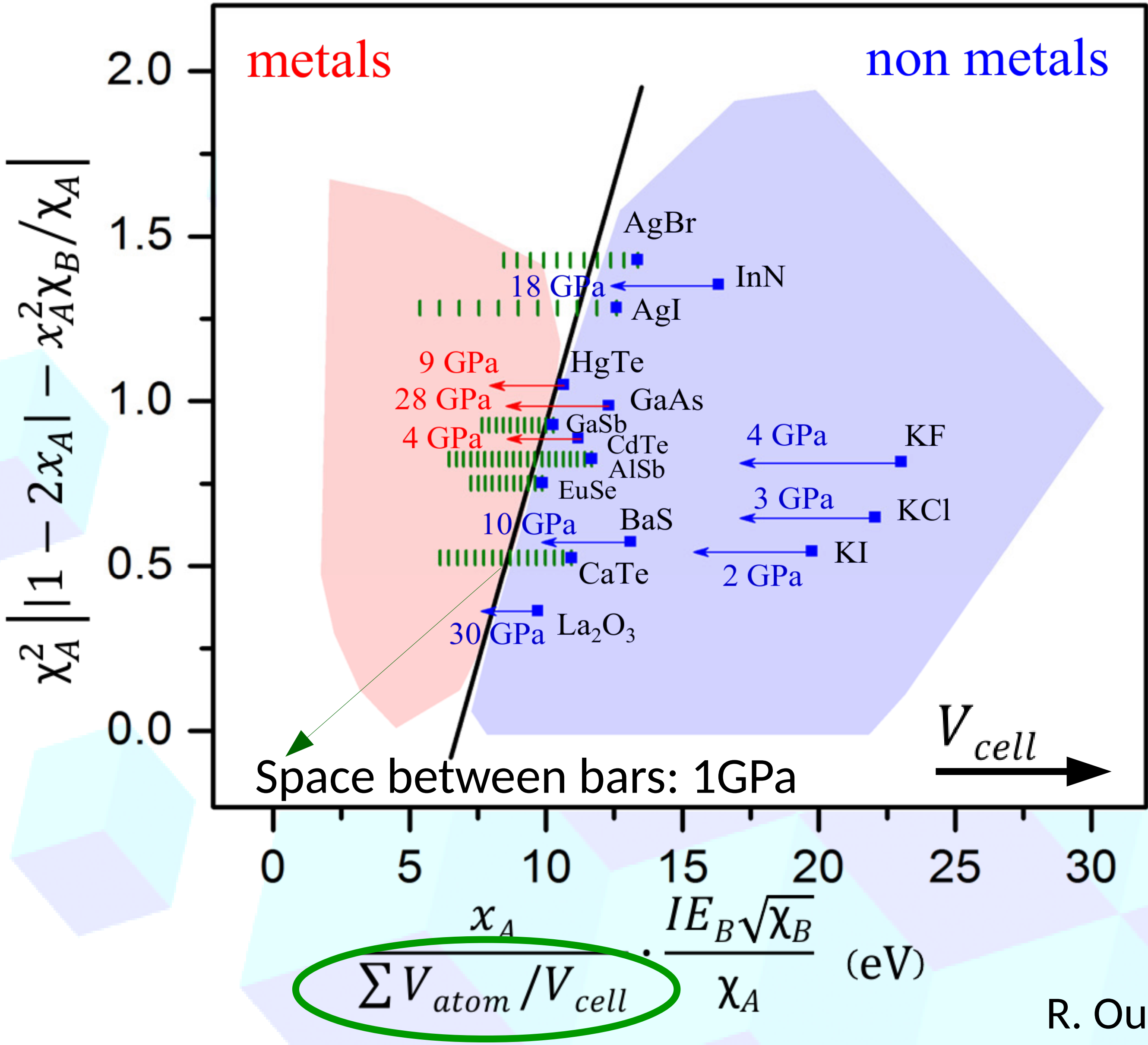
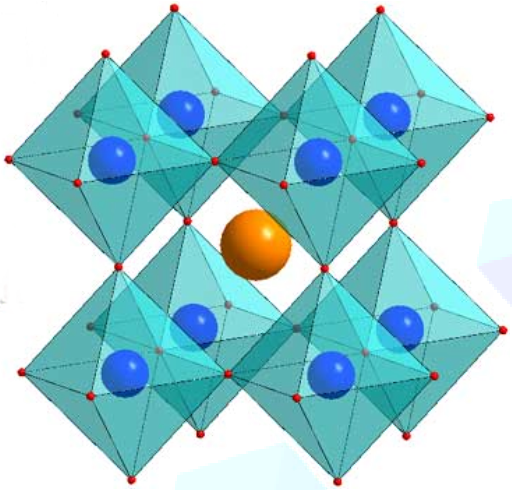


R. Ouyang *et al.* PRM (2018)

Perovskites' stability: an improved Goldschmidt Tolerance Factor

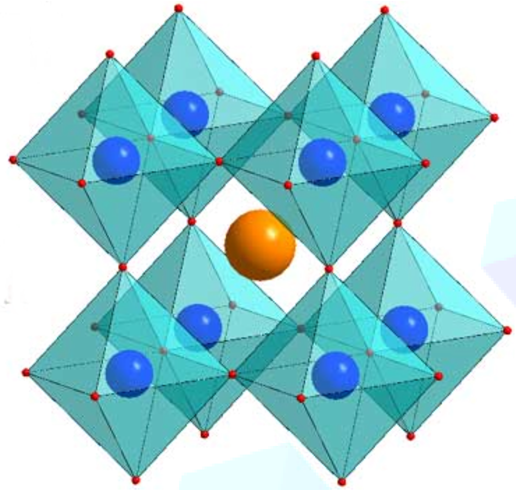


ABX$_3$

$$t = \frac{r_A + r_X}{\sqrt{2}(r_B + r_X)}$$ → Ionic radius

Goldschmidt* stable perovskites: 0.825 < ***t*** < 1.059, accuracy 79%

Perovskites' stability: an improved Goldschmidt Tolerance Factor



ABX$_3$

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

Ionic radius

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

Oxidation state

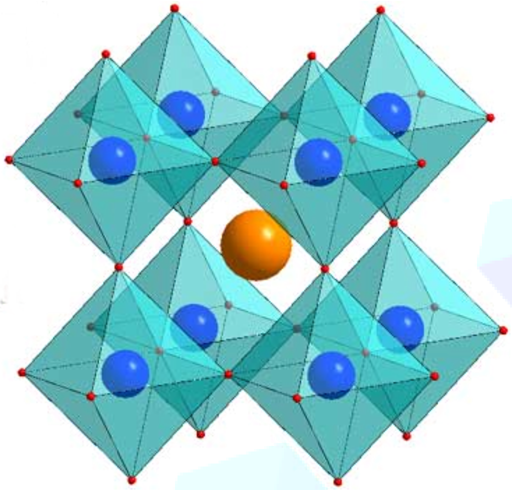
1 / μ = Octahedral factor

Goldschmidt* stable perovskites: 0.825 < t < 1.059, accuracy 79%

Our stable perovskites: τ < 4.18, accuracy 92%

Bartel, Sutton, Goldsmith, Ouyang, Musgrave, LMG &Scheffler, Sci. Adv. *5, eaav0693 (2019)*

Perovskites' stability: an improved Goldschmidt Tolerance Factor



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Goldschmidt* stable perovskites: 0.825 < t < 1.059, accuracy 79%

Our stable perovskites: τ < 4.18, accuracy 92%

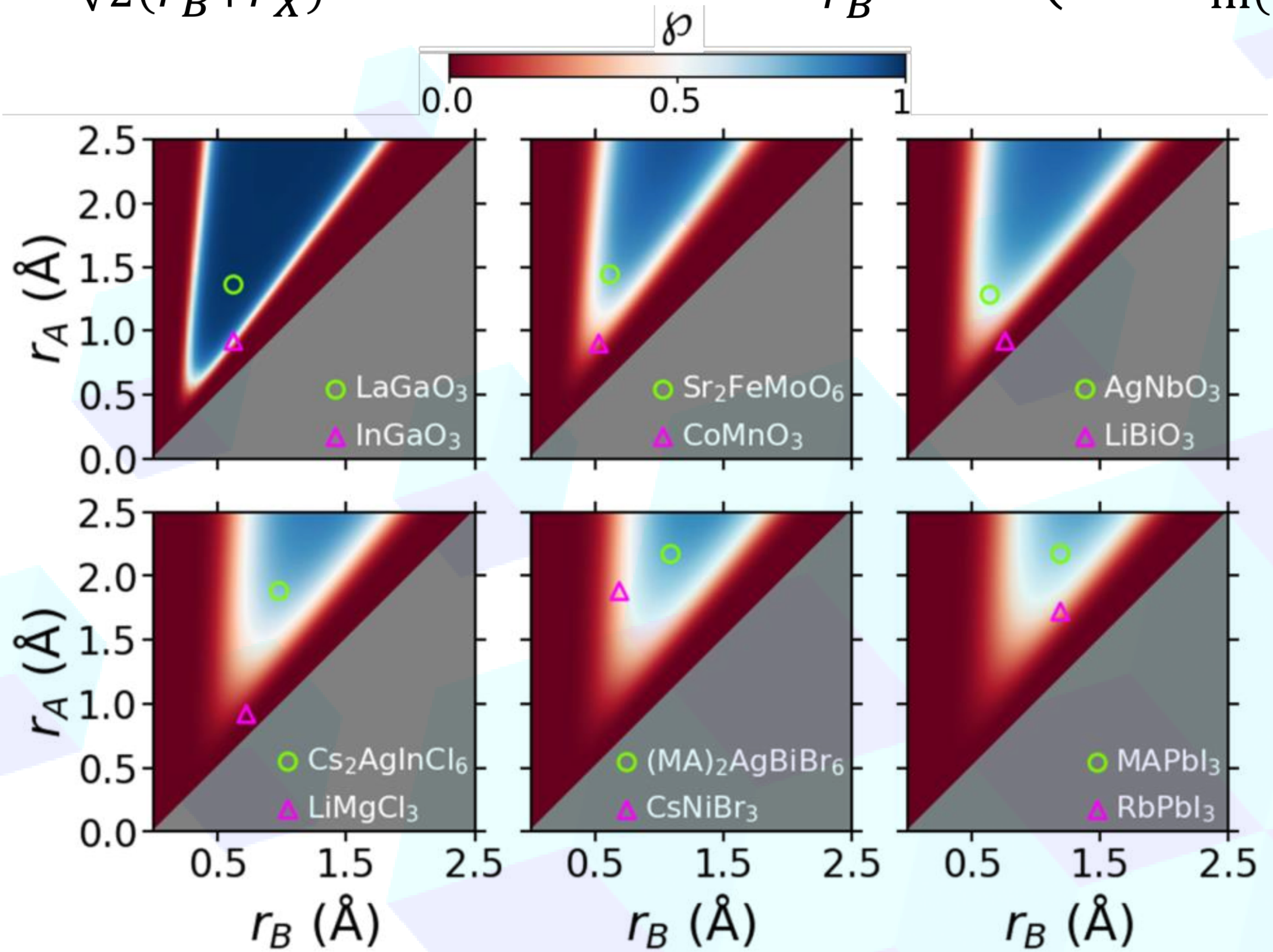
τ < 3.31 or τ > 5.92, 99% accuracy (1/3 of the training data)

τ < 3.31 or τ > 12.08, 100% accuracy (1/4 of the training data)

Bartel, Sutton, Goldsmith, Ouyang, Musgrave, LMG &Scheffler, Sci. Adv. *5, eaav0693 (2019)*

Improved Goldschmidt Tolerance Factor: Materials design

$$t = \frac{r_A + r_X}{\sqrt{2}(r_B + r_X)} \longrightarrow \tau = \frac{r_X}{r_B} - n_A\left(n_A - \frac{r_A/r_B}{\ln(r_A/r_B)}\right)$$



n_A

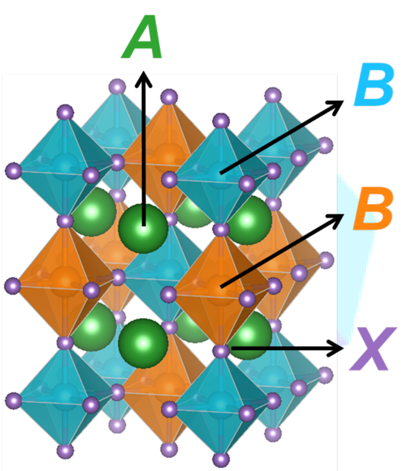
$X = O^{2-}$

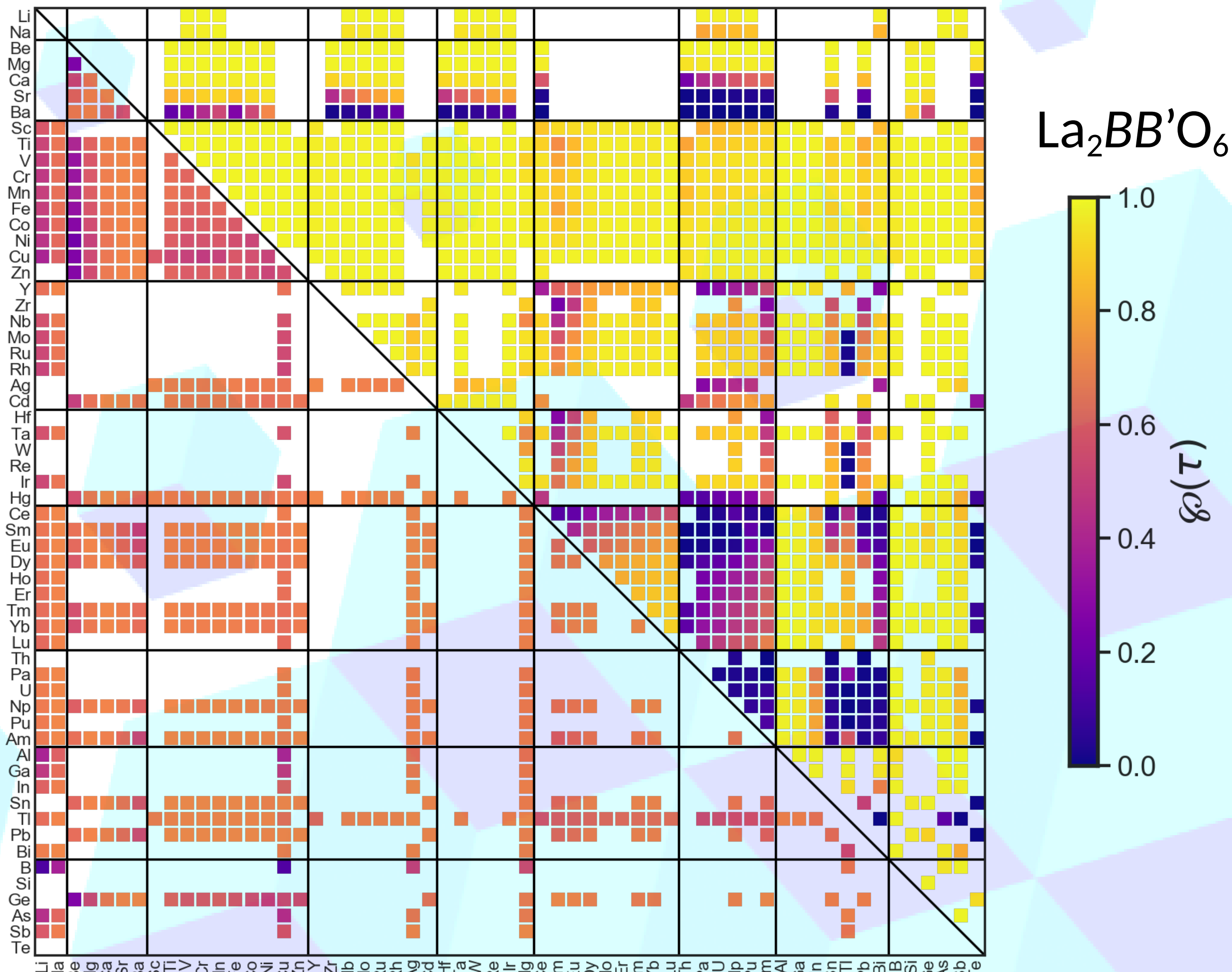
$n_A = 1^+$

X

Improved Goldschmidt Tolerance Factor: Extension of the materials space

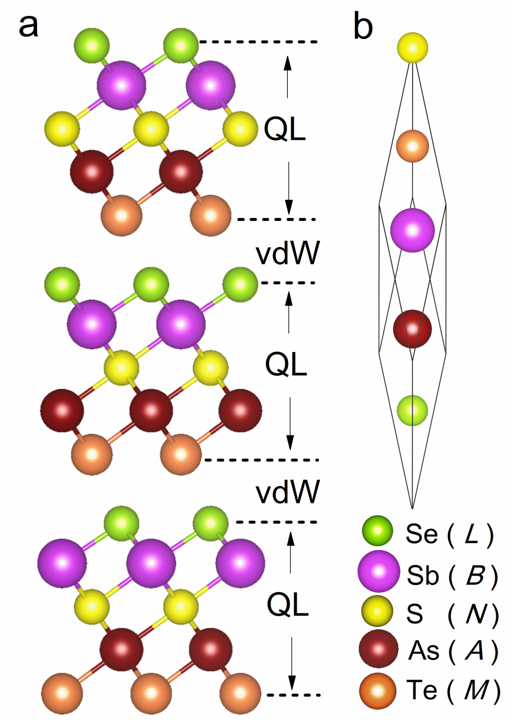
$$t = \frac{r_A + r_X}{\sqrt{2}(r_B + r_X)} \longrightarrow \tau = \frac{r_X}{r_B} - n_A\left(n_A - \frac{r_A/r_B}{\ln(r_A/r_B)}\right)$$





SISSO: predicting new tetradymite topological insulators

Prototype formula:
AB-LNM
AB = {As,Sb,Bi}
LNM = {S, Se,Te}



Cao, Liu, Ouyang, LMG, Zhou, Scheffler Zhang, Carbogno, in preparation (2019)

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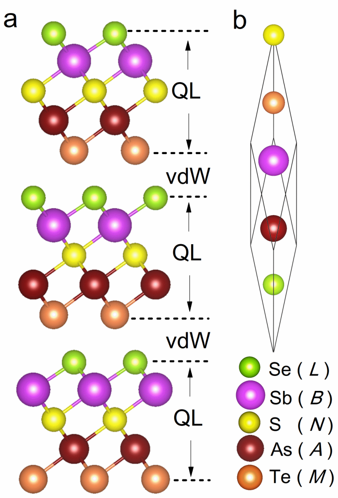

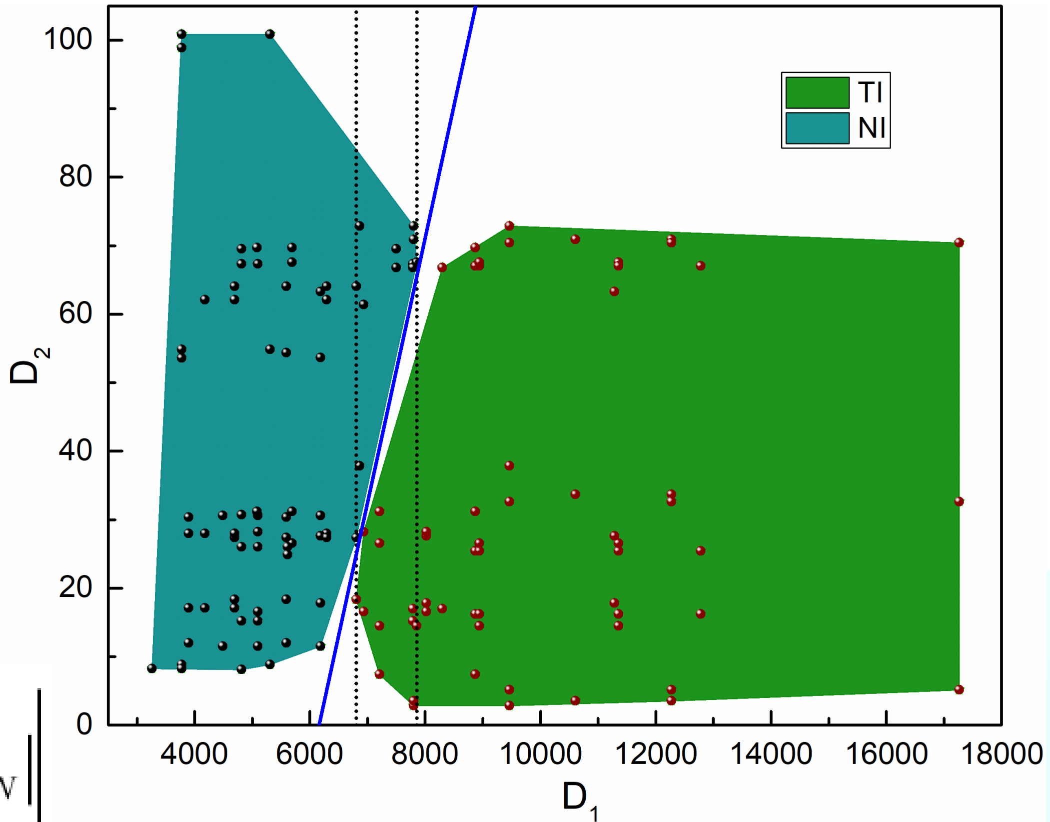

$$D_1 = (Z_A + Z_B) \cdot (Z_L + Z_M) - |Z_A Z_M - Z_B Z_L|$$

$$D_2 = \left| \frac{(\chi_M + \chi_N) \cdot Z_E}{\chi_A} - (Z_M + Z_N) - |Z_M - Z_N| \right|$$

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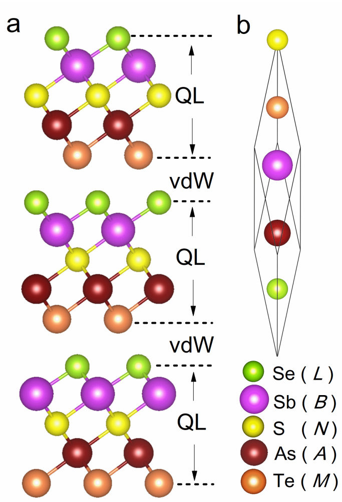

$$As_xSb_yBi_{2-x-y}S_aSe_bTe_{3-a-b}$$

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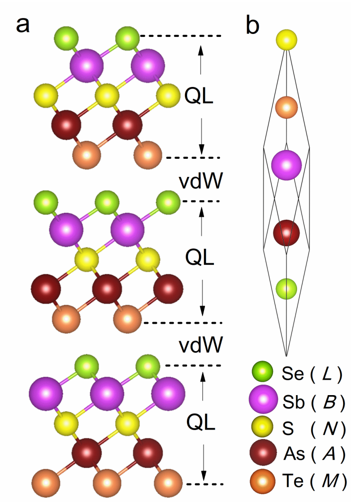

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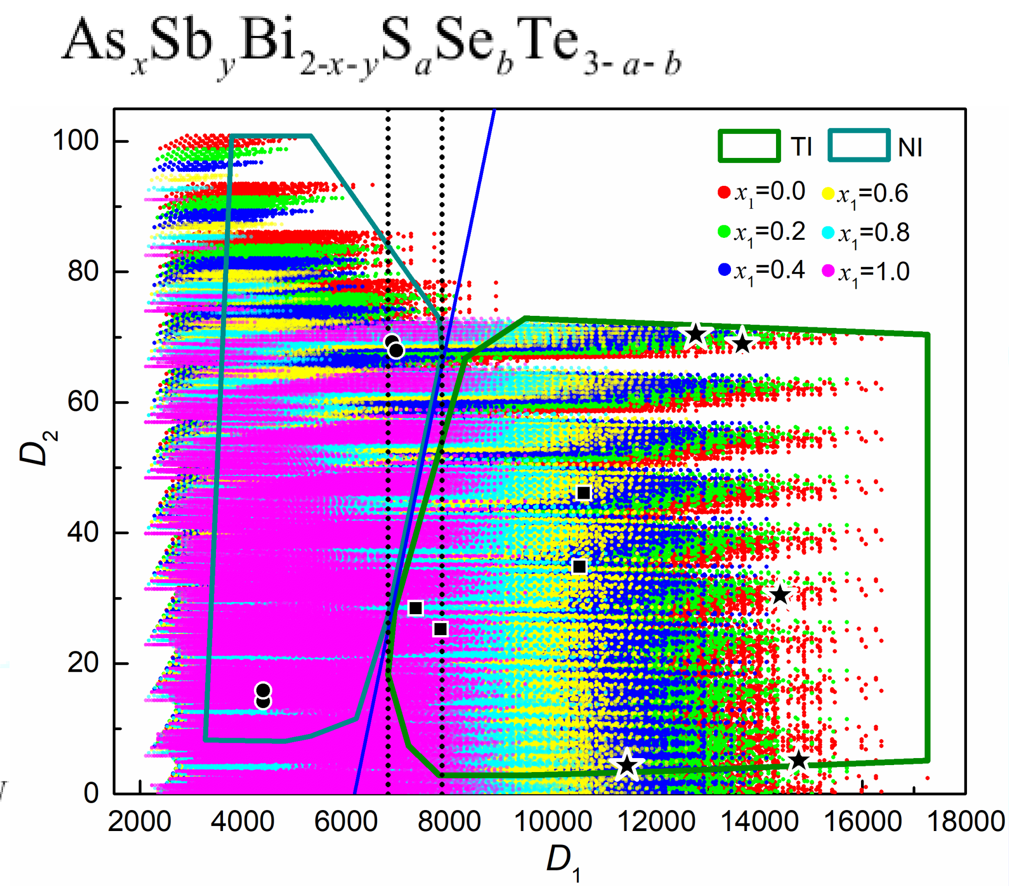

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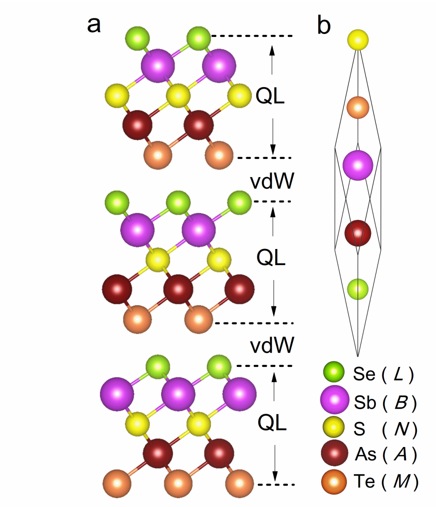

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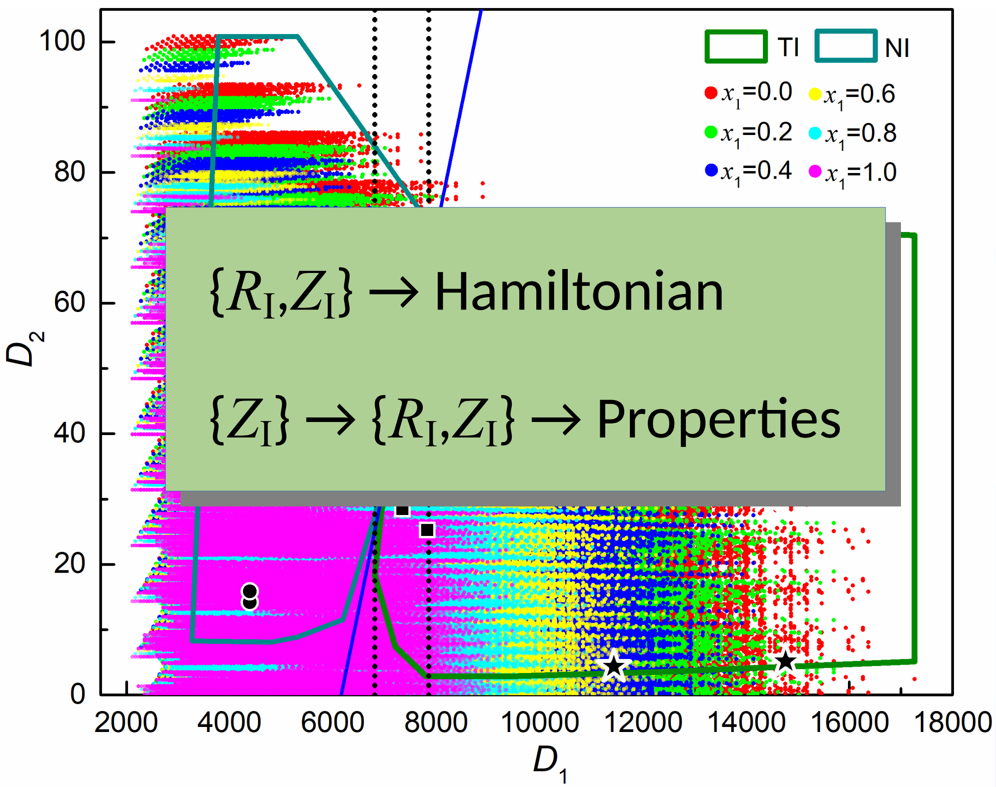


$\{R_\mathrm{I}, Z_\mathrm{I}\} \rightarrow$ Hamiltonian

$\{Z_\mathrm{I}\} \rightarrow \{R_\mathrm{I}, Z_\mathrm{I}\} \rightarrow$ Properties

Cao, Liu, Ouyang, LMG, Zhou, Scheffler Zhang, Carbogno, in preparation (2019)

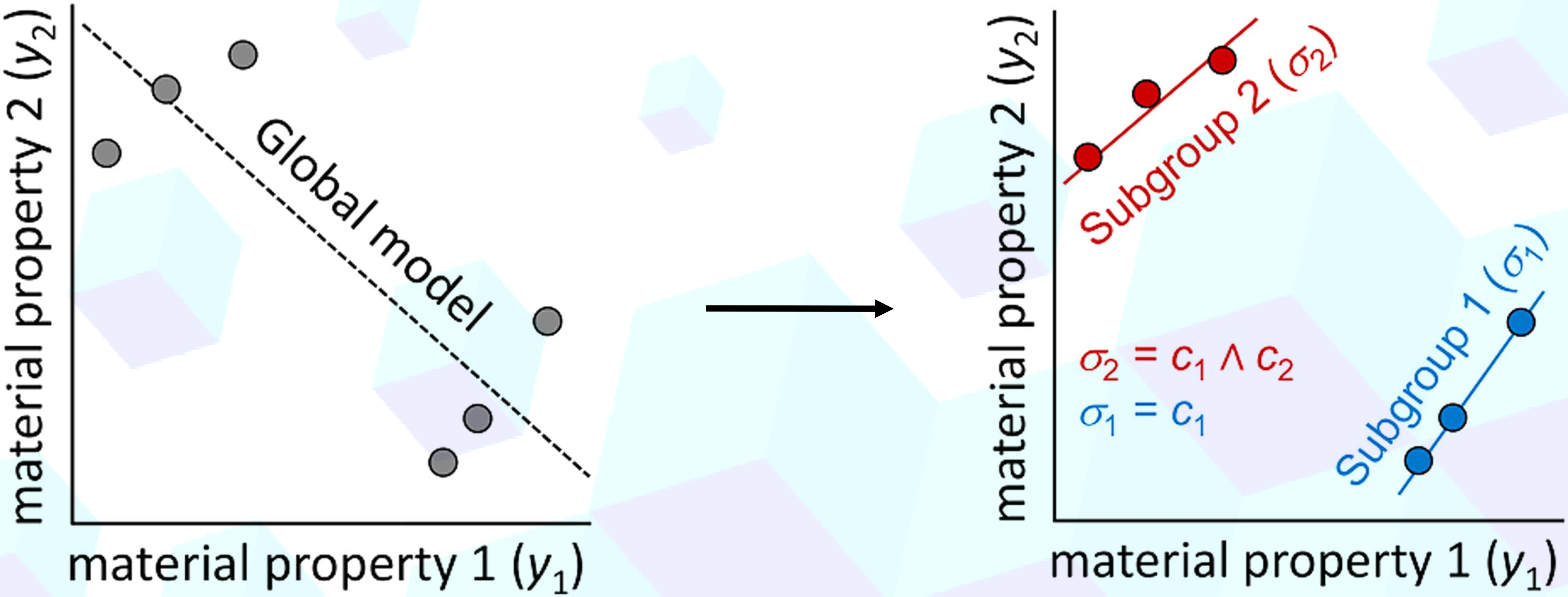
Compressed-sensing-based model identification (SISSO, and beyond): The context

$$\arg\min_{\boldsymbol{c}} \left(\|\boldsymbol{P} - \boldsymbol{D}\boldsymbol{c}\|_2^2 + \lambda \|\boldsymbol{c}\|_0 \right)$$

Compressed-sensing-based model identification:
Shares concepts with

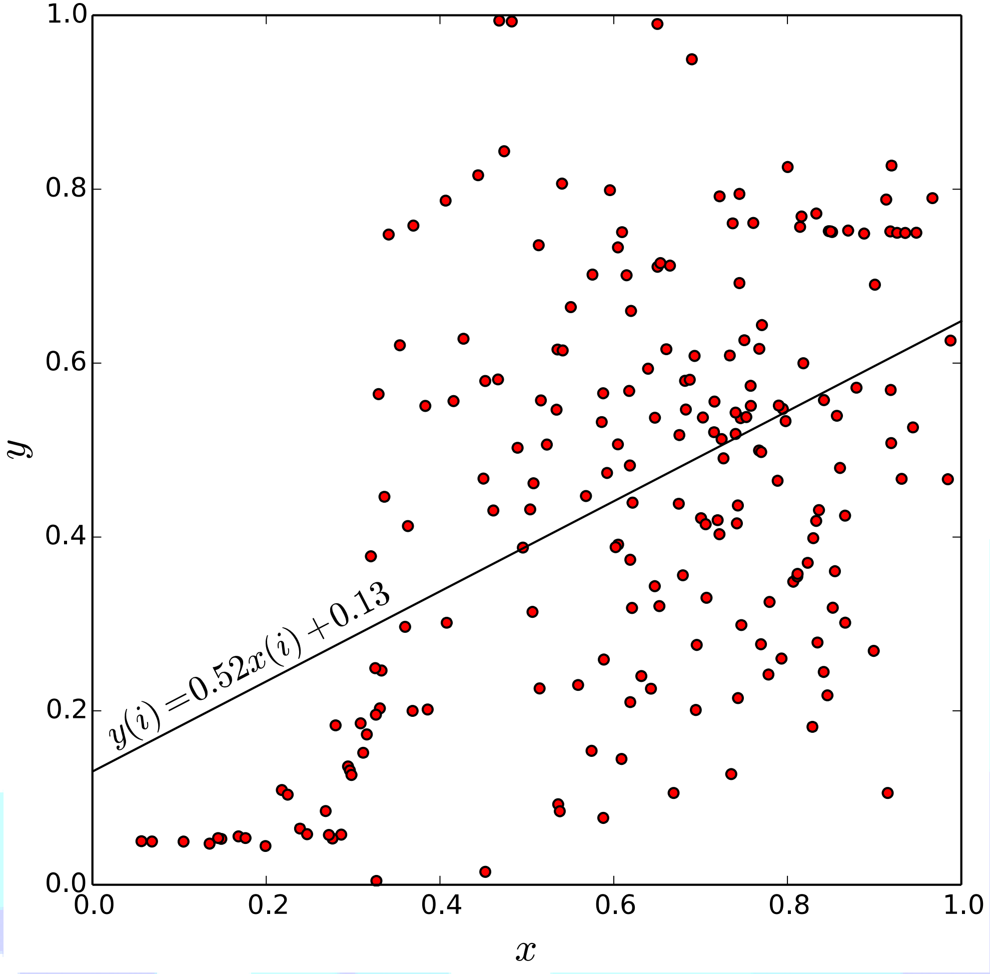
- Regularized regression. But: Massive sparsification.
- Dimensionality reduction. But <u>supervised</u>, and yielding sparse, “inspectable” descriptors
- Feature/Basis-set selection/extraction. But: non-greedy solver.
- Symbolic regression. But: deterministic solver.

Subgroup discovery



Boley, Goldsmith, LMG & Vreeken, Data Min. Knowl. Disc. **31**, 1391 (2017)
Goldsmith, Boley, Vreeken, Scheffler & LMG, New J. Phys. **19**, 013031 (2017)

Subgroup discovery: finding descriptive statements about outstanding groups



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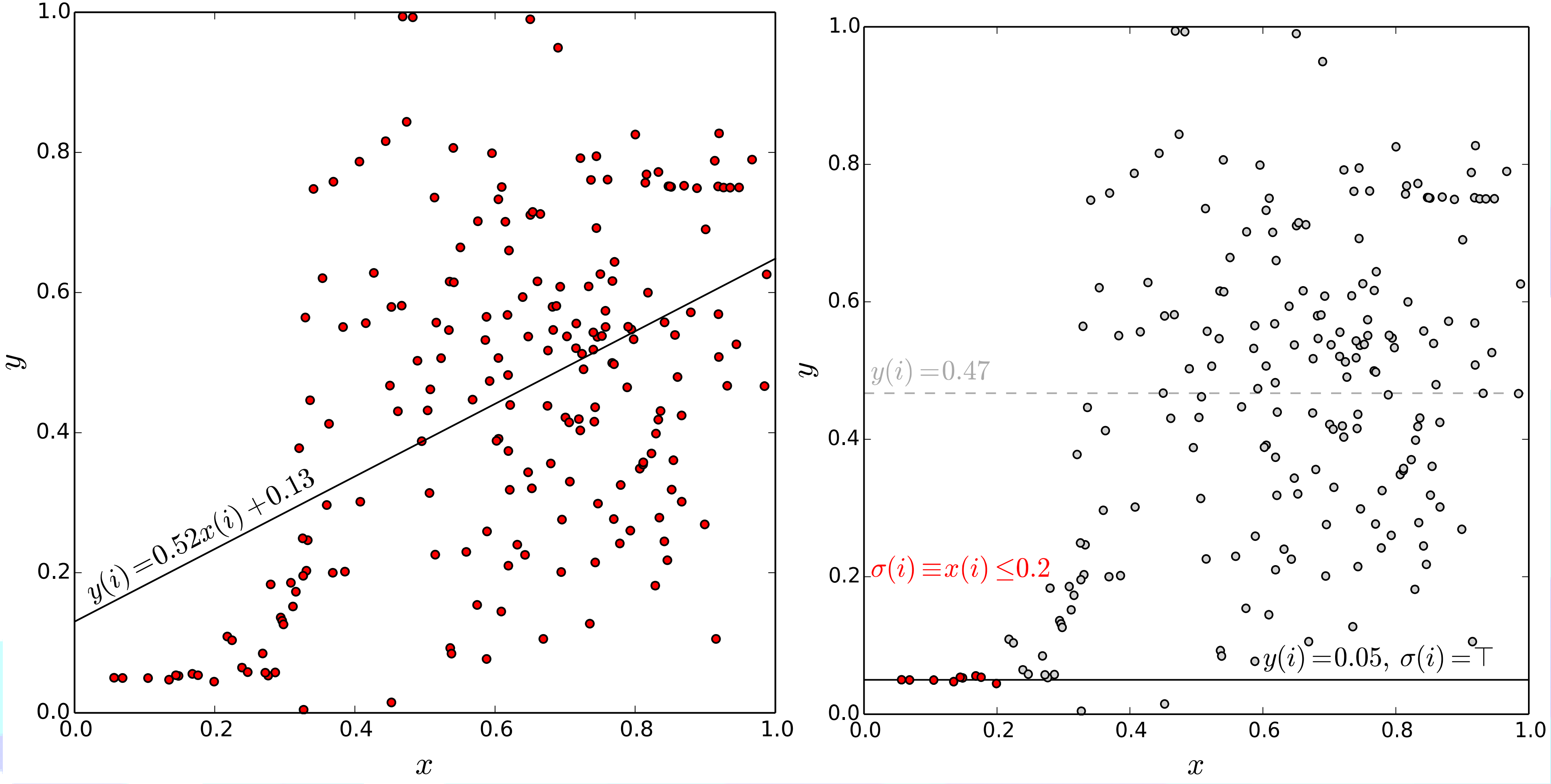

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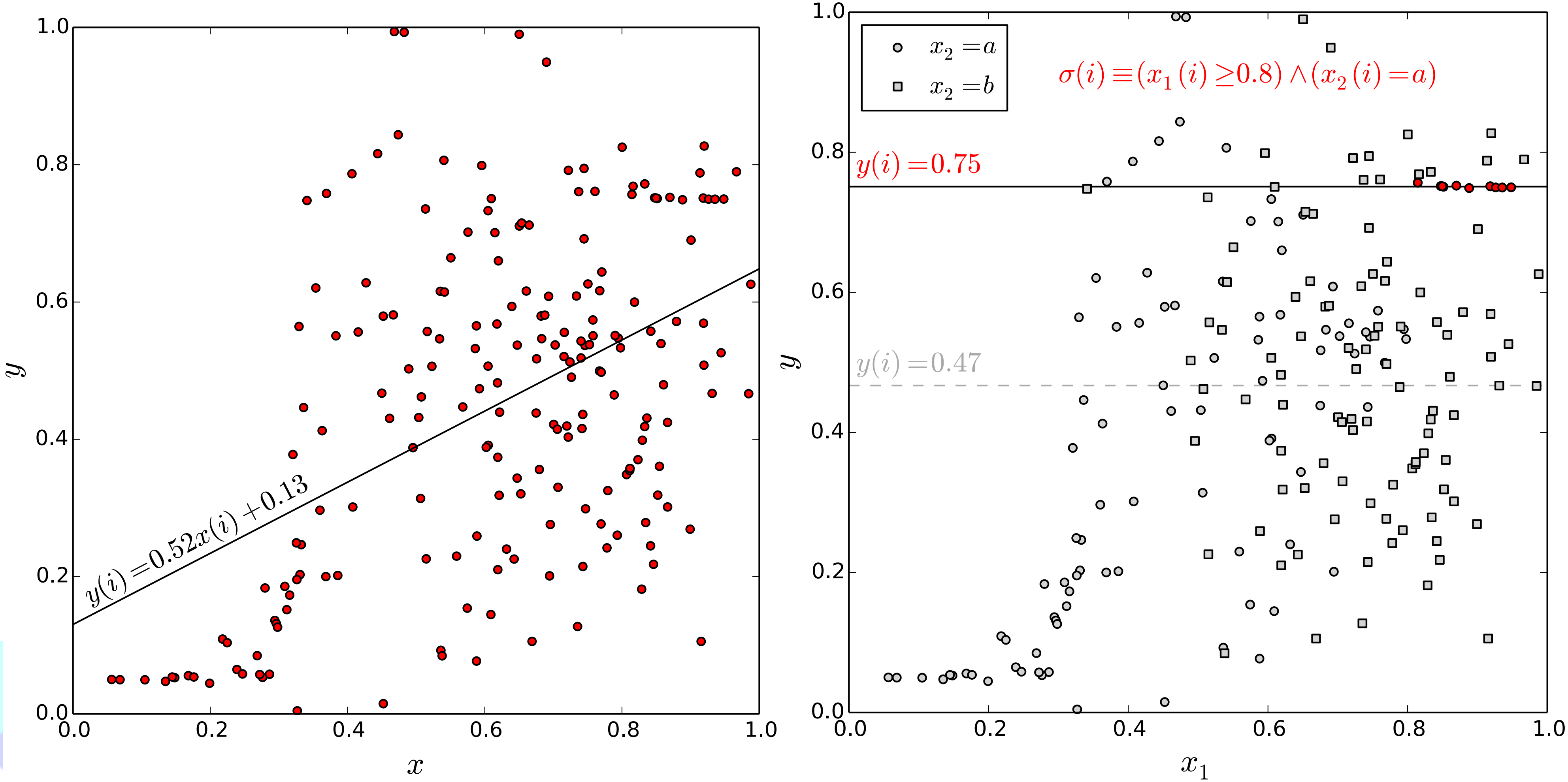
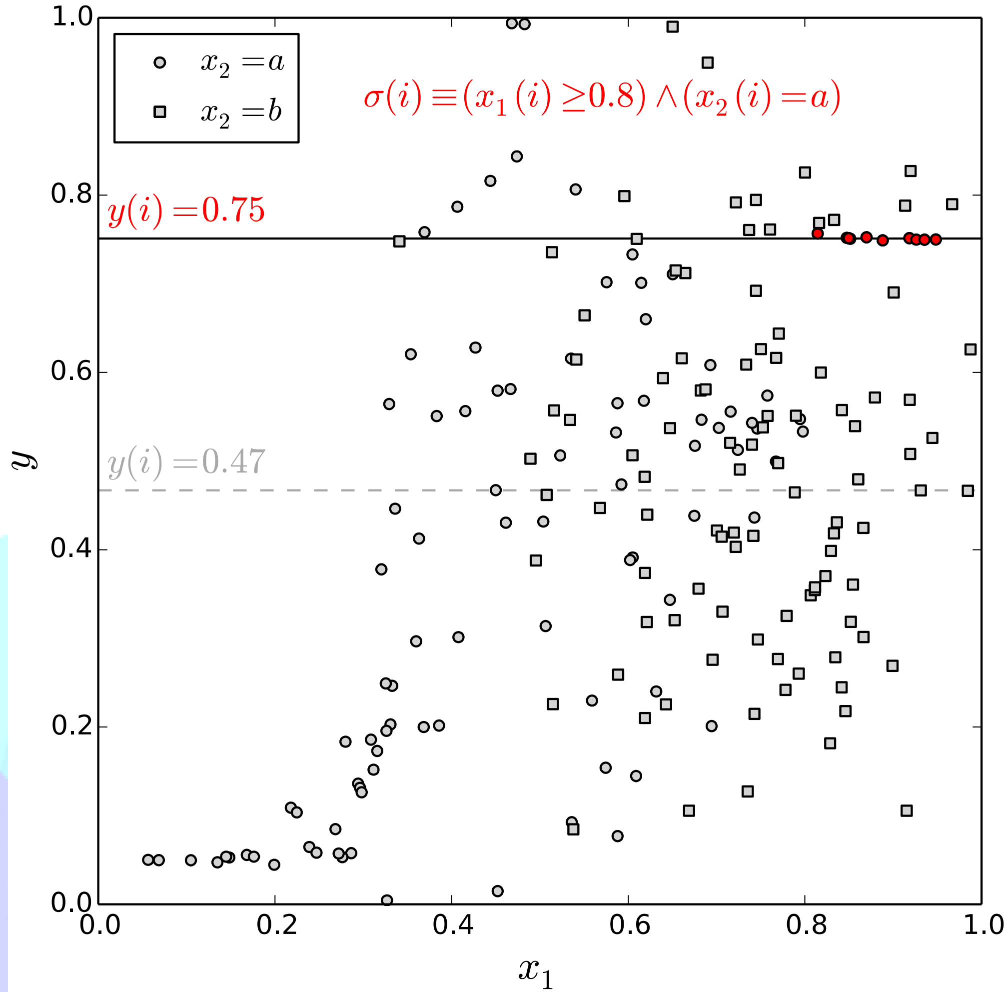


Subgroup discovery: finding descriptive statements about outstanding groups

Ingredients:
Population $P = \{1, \ldots, n\}$
Target Variable y: $P \rightarrow Y$
Description variables x_j: $P \rightarrow X_j$
Basic propositions $\Pi = \{\pi_1, \ldots, \pi_k\}$
Objective functions: f
{All possible subgroups of P} $\rightarrow \mathbb{R}$



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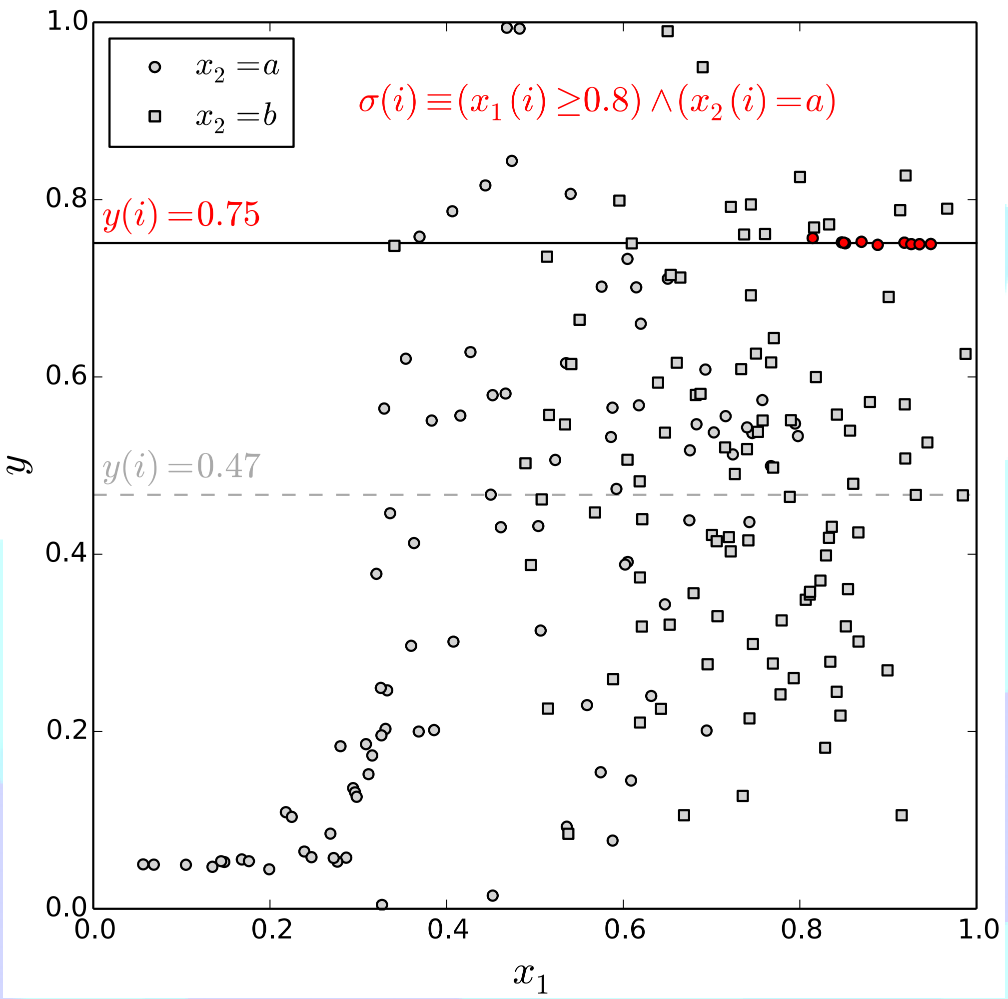
$\{$All possible subgroups of $P\} \rightarrow \mathbb{R}$

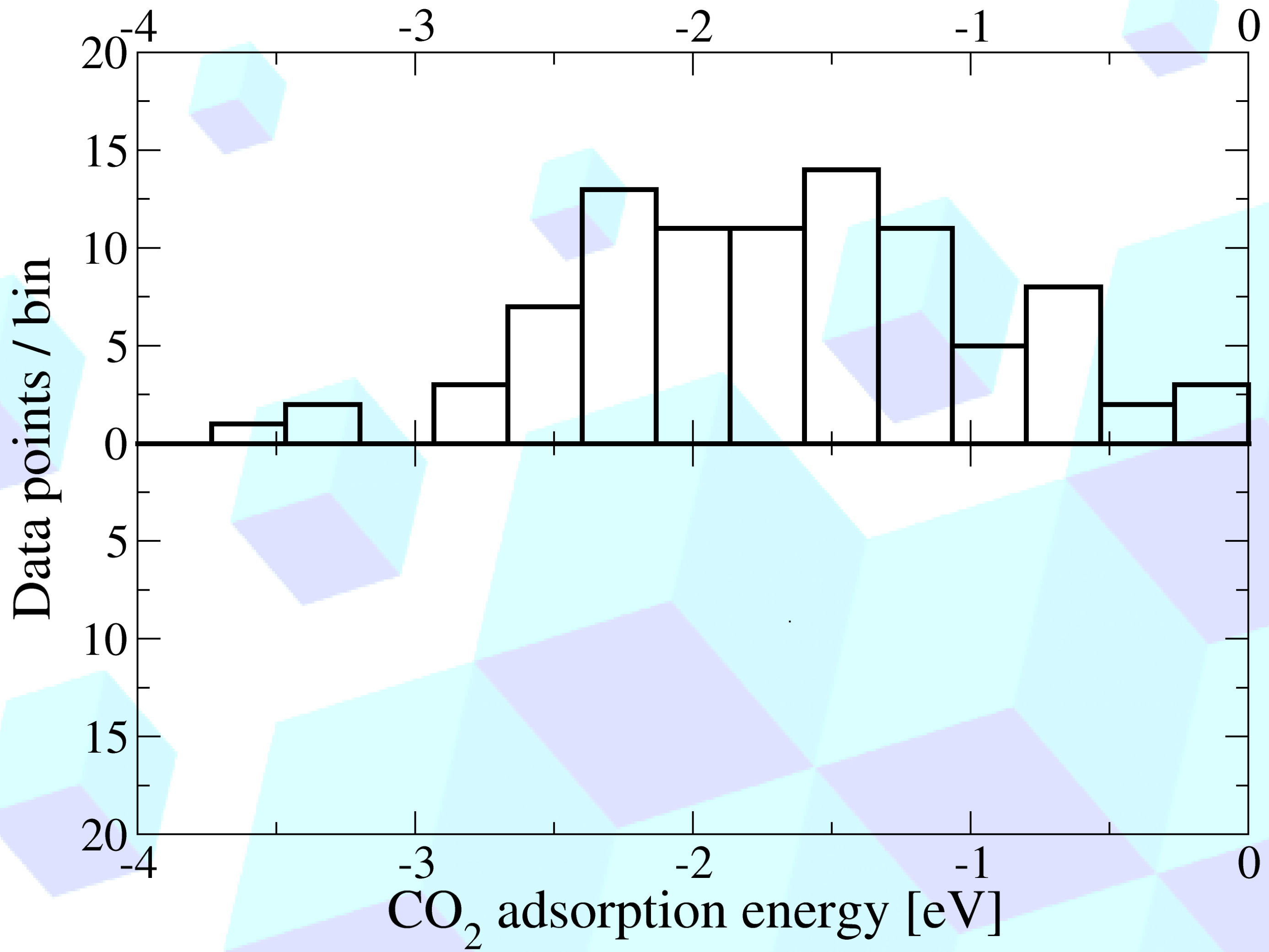
Task:
Finding $\sigma(i) = \pi_1(i) \wedge \dots \wedge \pi_m(i)$
For which $f(P) = \max$

Typical form of f:
"Size of subgroup" × "Reduction of variance of Y compared to the whole population"



Subgroup discovery in practice

Distribution of adsorption energies of CO_2
on different surfaces of several metal-oxides



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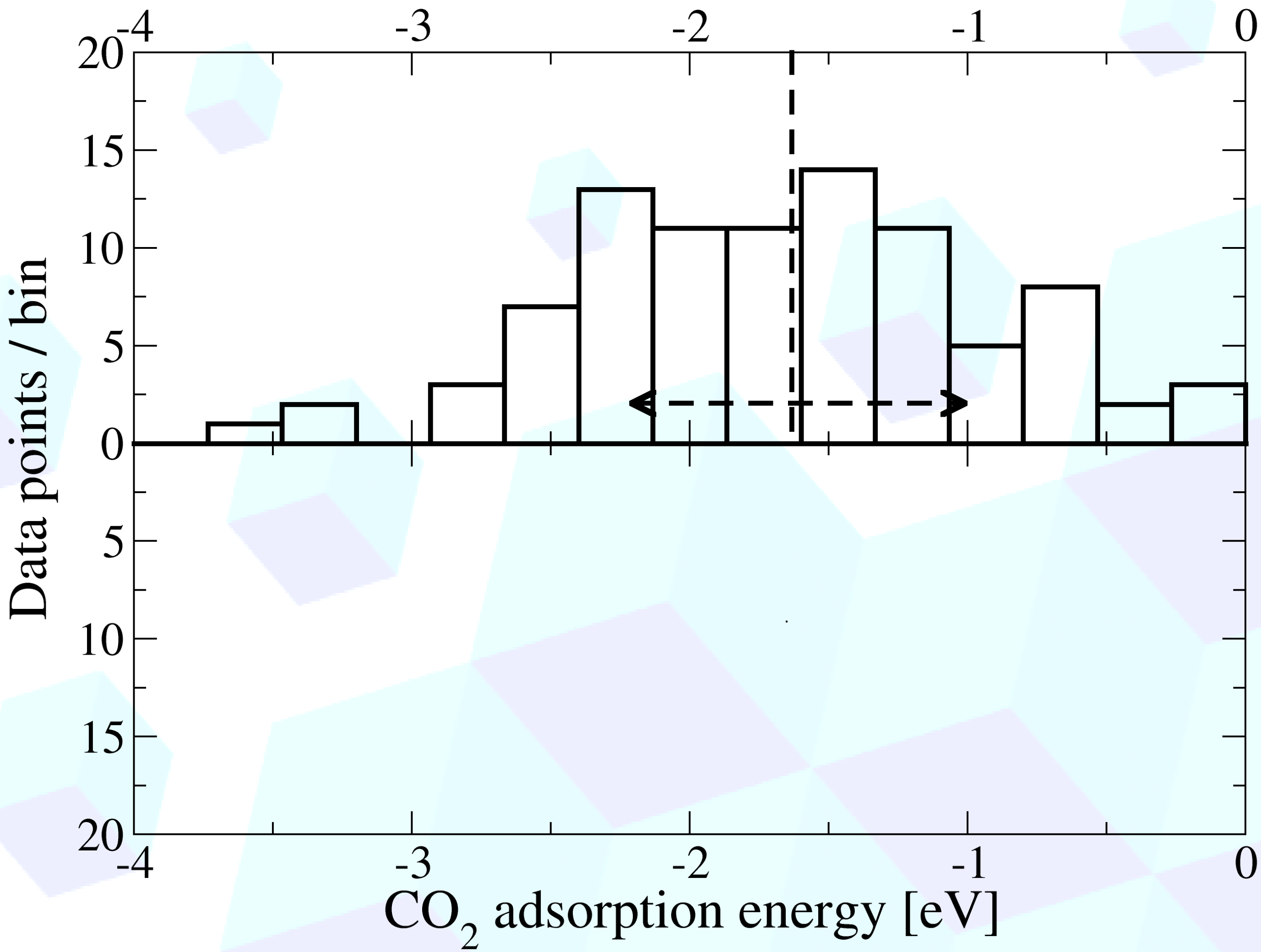

Subgroup discovery in practice

$$\underset{SG \subset P}{\operatorname{argmax}}\, U = \frac{\#SG}{\#P}\left(1 - \frac{\operatorname{mad}(SG)}{\operatorname{mad}(P)}\right)\left|\operatorname{med}(SG) - \operatorname{med}(P)\right|$$

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Size of full set P

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Mean absolute deviation
from the median
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Size of full set P

Median of the distribution

SG is *described* by a *selector*,
a conjunction of statements (s_1 ^ s_2 ^ ...)
about a list of given features e.g.,
s_1 = surface energy larger than ... ,
s_2 = *p*-band center of surface O less than ...

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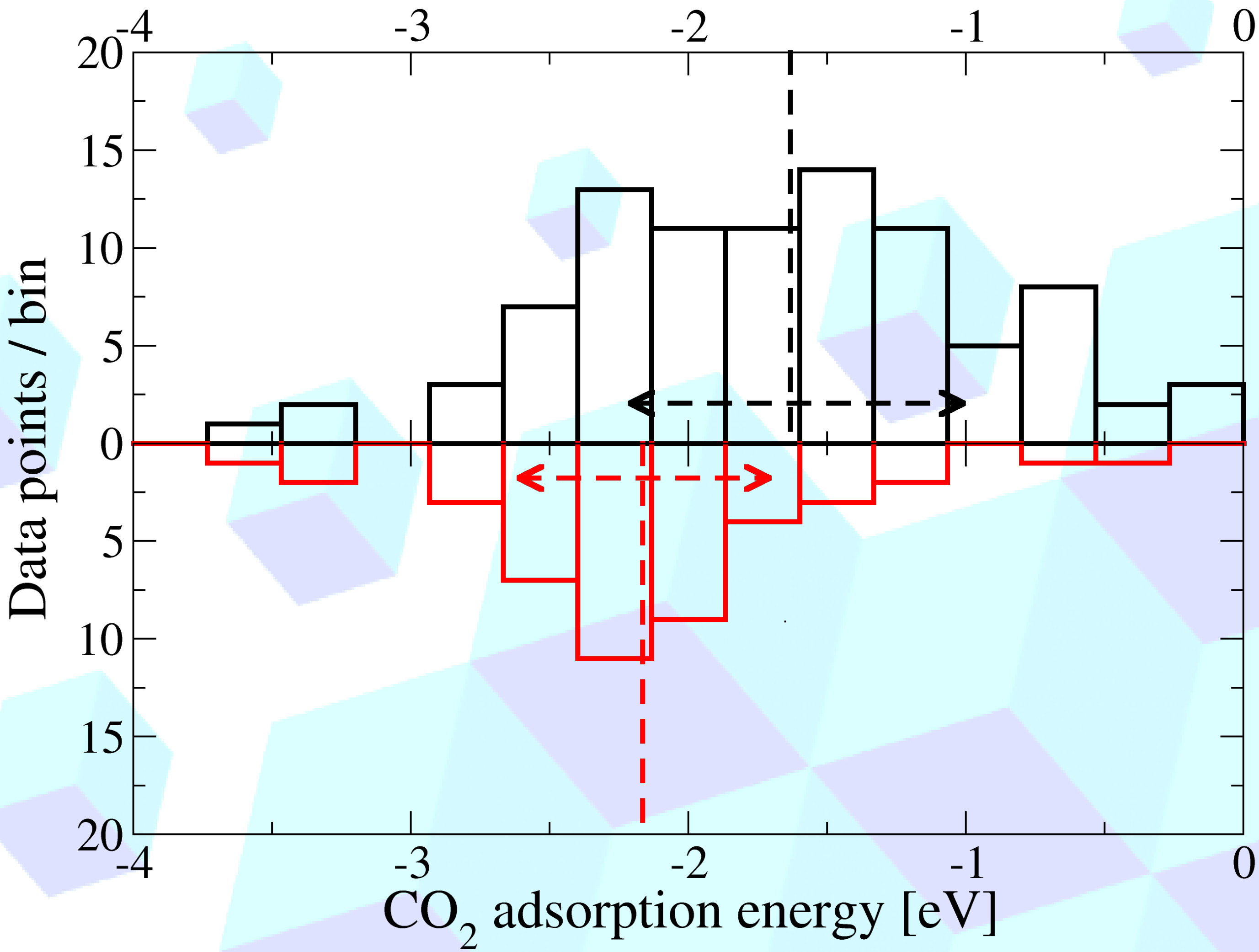

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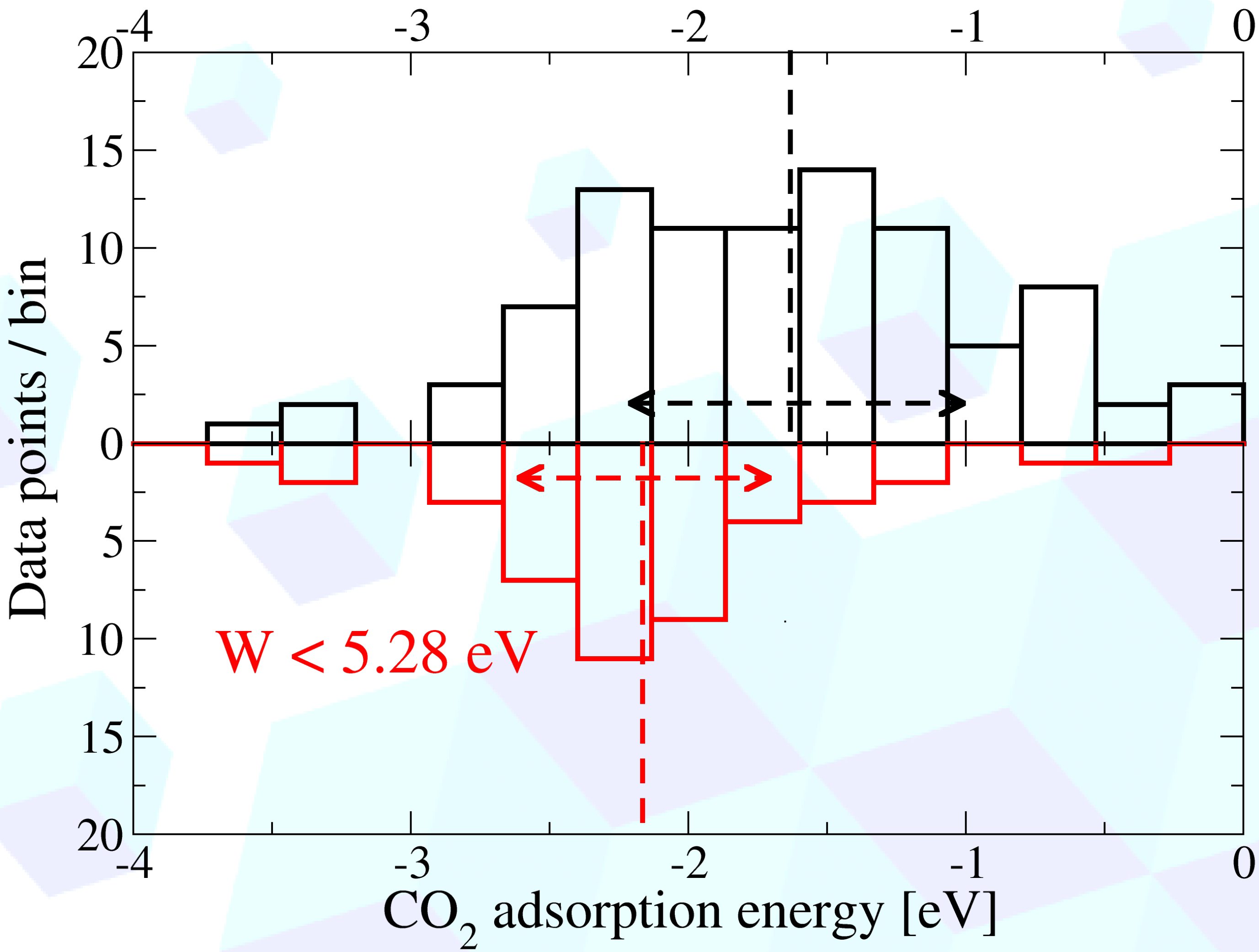

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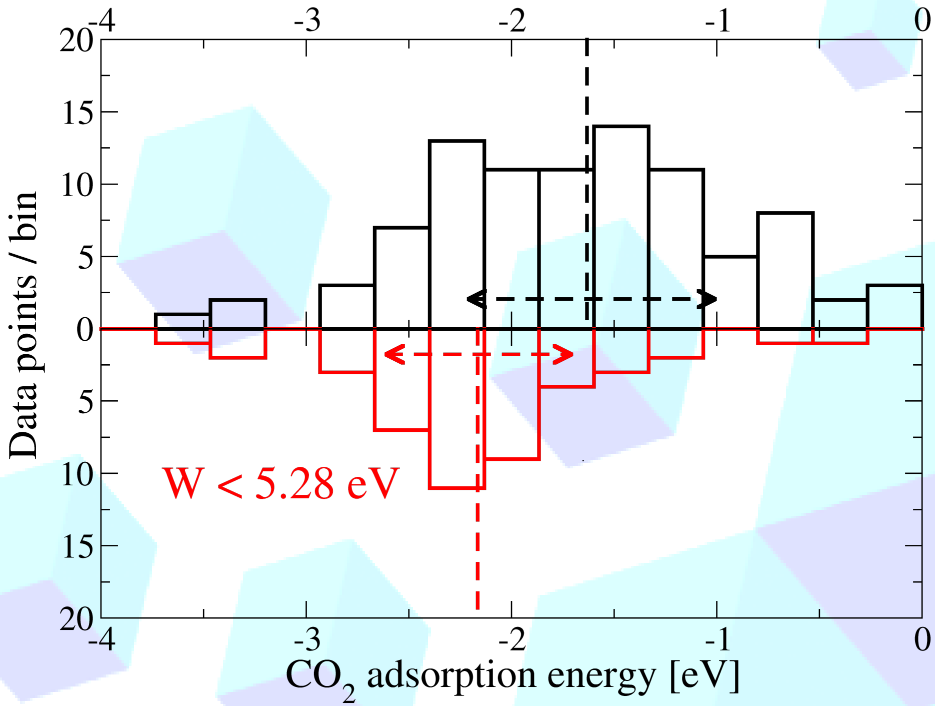

The (SISSO) model for the discovered subgroup
- is more accurate than the global model
- has a different descriptor due to different physics.

Small work function:
Surfaces with dominantly ionic character

Acknowledgements

Compressed sensing, SISSO, and metal/insulator proof of concepts
Jan Vybiral, Runhai Ouyang, Emre Ahmetcik, Stefano Curtarolo, Sergey Levchenko, Claudia Draxl

Application of SISSO to perovskites
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Application of SISSO to topological insulators
Guohua Cao, Runhai Ouyang, Zizhen Zhou, Huijun Liu, Christian Carbogno, Zhenyu Zhangave

Transparent conducting oxide: NOMAD-kaggle competition
Christopher Sutton, Angelo Ziletti,
Claudia Draxl, Daan Frenkel, Kristian Thygesen, Samuel Kaski, Bernhard Schölkopf

Subgroup Discovery and application to CO_2 adsorption
Mario Boley, Jilles Vreeken, Aleksei Mazheika, Sergey Levchenko

All the above
Matthias Scheffler

NOMAD
NOVEL MATERIALS DISCOVERY

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Tutorial (jupyter notebook)
On symbolic + regularized regression (from linear regression to SISSO)

Ask me (luca@fhi-berlin.mpg.de)
for user ID and password

All the above
Matthias Scheffler

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