NOMAD

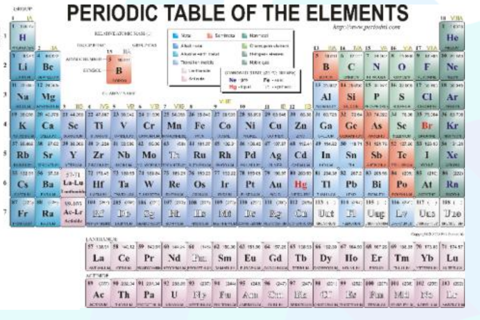
NOVEL MATERIALS DISCOVERY

Patterns, Correlations, and Causality in Big Data of Materials: Analytics for Novel Materials Discovery

From *the periodic table of the elements* to *a chart (a map) of materials*: Organize materials according to their properties and functions.

Dmitri Mendeleev (1834-1907)

- crystal-structure prediction
- figure of merit of thermoelectrics (as function of *T*)
- turn-over frequency of catalytic materials (as function of *T* and *p*)
- efficiency of photovoltaic systems
- etc.



(*) Work performed in collaboration with
Luca Ghiringhelli, Jan Vybiral, Claudia Draxl, et al.

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NOVEL MATERIALS DISCOVERY

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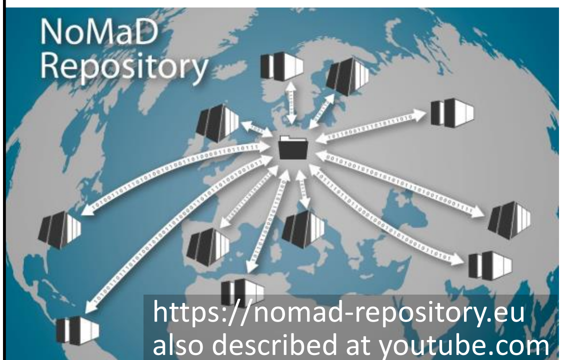

The NoMaD Repository accepts (and requests) in- and output files of all important codes. Currently, the NoMaD Repository contains **3,026,745** entries.

http://NOMAD-CoE.eu

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https://www.youtube.com/watch?v=L-nmRSH4NQM
http://v.youku.com/v_show/id_XMTM0NDA0NDIxMg==.html

NoMaD

The Novel Materials Discovery Repository

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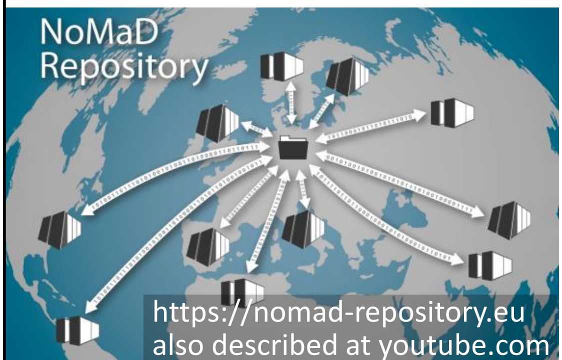
NOVEL MATERIALS DISCOVERY



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There are 30-40 important codes used in computational materials science.

Nomenclature, data representation, and file formats of the input and output files of these codes are different. The heterogeneity could hardly be worse.

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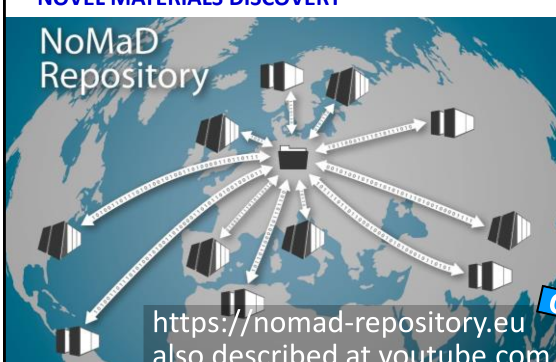

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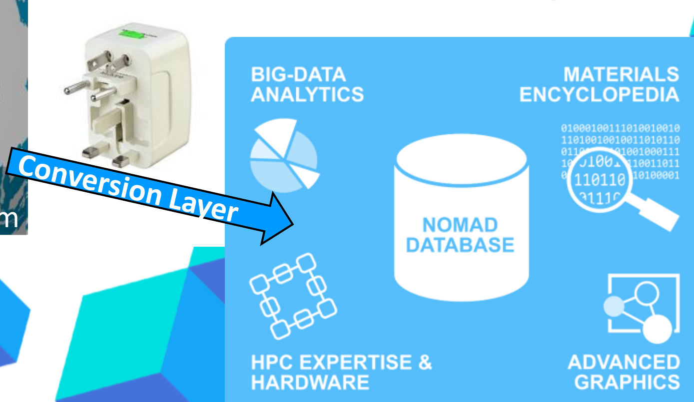

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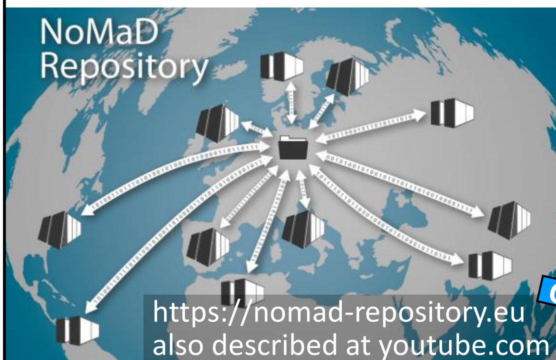

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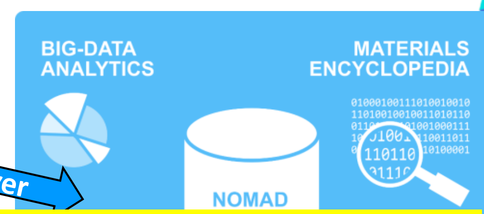
Conversion Layer



Why do we consider "all" electronic structure theory codes in our NOMAD repository and data base?

There are millions of data already. **Do science with the data! Select the studied systems more intelligently.**

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NOVEL MATERIALS DISCOVERY

The Big-Data Challenge
The Amount of Data Created Every Day Is Significant

Most Data Are Not Used And Even Thrown Away

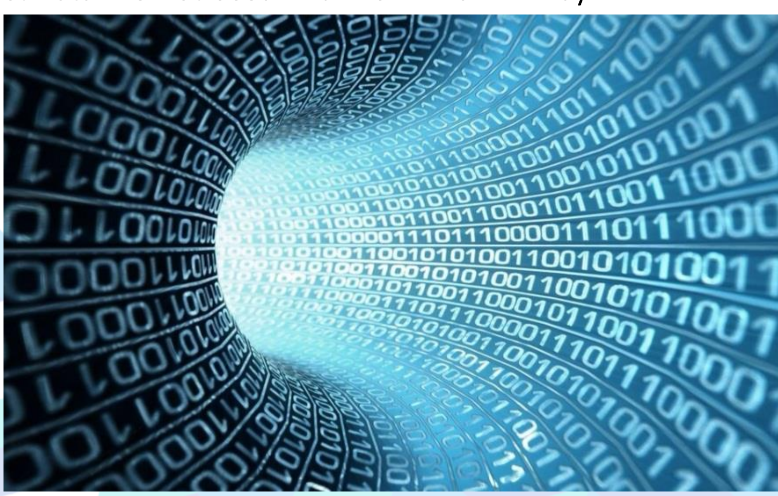
The Four V and an A !

Volume (amount of data),

Variety (heterogeneity of form and meaning of data),

Velocity at which data may change or new data arrive,

Veracity (uncertainty of quality).



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NOVEL MATERIALS DISCOVERY

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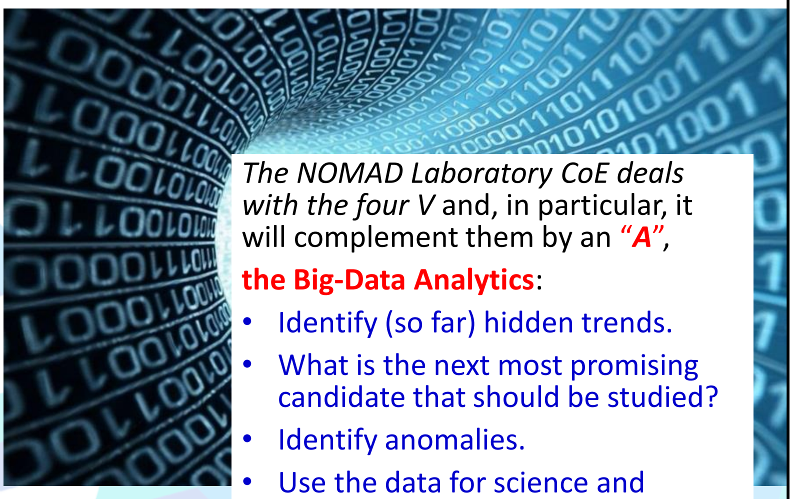
Query and read out what was stored;
high-throughput screening.



The NOMAD Laboratory CoE deals with the four V and, in particular, it will complement them by an *"A"*,
the Big-Data Analytics:

- Identify (so far) hidden trends.
- What is the next most promising candidate that should be studied?
- Identify anomalies.
- Use the data for science and engineering, for R & D.

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NOVEL MATERIALS DISCOVERY

Science **351**, aad3000 (2016)

RESEARCH

RESEARCH ARTICLE

DFT METHODS

Reproducibility in density functional theory calculations of solids

Kurt Lejaeghere,[1*] Gustav Bihlmayer,[2] Torbjörn Björkman,[3,4] Peter Blaha,[5]

Science **351**, aad3000 (2016)

Kurt Lejaeghere,[1*] Gustav Bihlmayer,[2] Torbjörn Björkman,[3,4] Peter Blaha,[5] Stefan Blügel,[2] Volker Blum,[6] Damien Caliste,[7,8] Ivano E. Castelli,[9] Stewart J. Clark,[10] Andrea Dal Corso,[11] Stefano de Gironcoli,[11] Thierry Deutsch,[7,8] John Kay Dewhurst,[12] Igor Di Marco,[13] Claudia Draxl,[14,15] Marcin Dułak,[16] Olle Eriksson,[13] José A. Flores-Livas,[12] Kevin F. Garrity,[17] Luigi Genovese,[7,8] Paolo Giannozzi,[18] Matteo Giantomassi,[19] Stefan Goedecker,[20] Xavier Gonze,[19] Oscar Grånäs,[13,21] E. K. U. Gross,[12] Andris Gulans,[14,15] François Gygi,[22] D. R. Hamann,[23,24] Phil J. Hasnip,[25] N. A. W. Holzwarth,[26] Diana Iuşan,[13] Dominik B. Jochym,[27] François Jollet,[28] Daniel Jones,[29] Georg Kresse,[30] Klaus Koepernik,[31,32] Emine Küçükbenli,[9,11] Yaroslav O. Kvashnin,[13] Inka L. M. Locht,[13,33] Sven Lubeck,[14] Martijn Marsman,[30] Nicola Marzari,[9] Ulrike Nitzsche,[31] Lars Nordström,[13] Taisuke Ozaki,[34] Lorenzo Paulatto,[35] Chris J. Pickard,[36] Ward Poelmans,[1,37] Matt I. J. Probert,[25] Keith Refson,[38,39] Manuel Richter,[31,32] Gian-Marco Rignanese,[19] Santanu Saha,[20] Matthias Scheffler,[15,40] Martin Schlipf,[22] Karlheinz Schwarz,[5] Sangeeta Sharma,[12] Francesca Tavazza,[17] Patrik Thunström,[41] Alexandre Tkatchenko,[15,42] Marc Torrent,[28] David Vanderbilt,[23] Michiel J. van Setten,[19] Veronique Van Speybroeck,[1] John M. Wills,[43] Jonathan R. Yates,[29] Guo-Xu Zhang,[44] Stefaan Cottenier[1,45*]

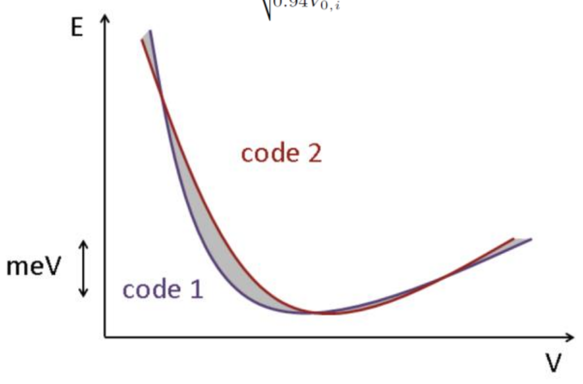
Science **351**, aad3000 (2016)

NOVEL MATERIALS DISCOVERY

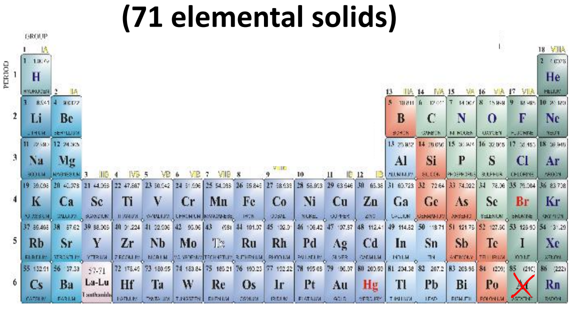
Δ-Value Project in Materials Science

Science 351, aad3000 (2016)

$$\Delta_i(a,b) = \sqrt{\int_{0.94V_{0,i}}^{1.06V_{0,i}} \frac{\left(E_{b,i}(V) - E_{a,i}(V)\right)^2}{0.12V_{0,i}} \, \mathrm{d}V}$$



An exhaustive test set (71 elemental solids)



NOVEL MATERIALS DISCOVERY

Comparing Solid State DFT Codes, Basis Sets, and Potentials (K. Lejaeghere et al.)

https://molmod.ugent.be/deltacodesdft
Science 351, aad3000 (2016)

Code	Version	Basis	Electron treatment	Δ-value meV/atom
WIEN2k	13.1	LAPW/APW+lo	all-electron	0
FHI-aims	081213	tier2 numerical orbitals	all-electron (relativistic atomic_zora scalar)	0.2
Exciting	development version	LAPW+xlo	all-electron	0.2
Quantum ESPRESSO	5.1	plane waves	SSSP Accuracy (mixed NC/US/PAW potential lib.)	0.3
Elk	3.1.5	APW+lo	all-electron	0.3
VASP	5.2.12	plane waves	PAW 2015	0.4
FHI-aims	081213	tier2 numerical orbitals	all-electron (relativistic zora scalar 1e-12)	0.4
CASTEP	9.0	plane waves	OTFG CASTEP 9.0	0.5

NOVEL MATERIALS DISCOVERY

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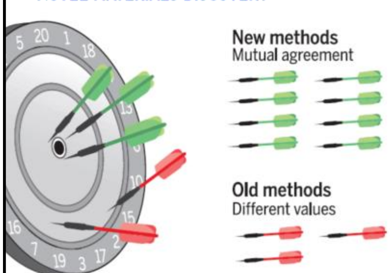
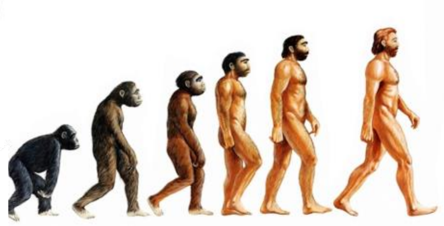
Science 351, aad3000 (2016)



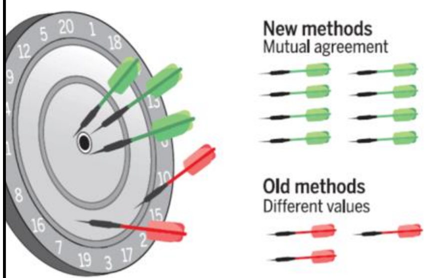


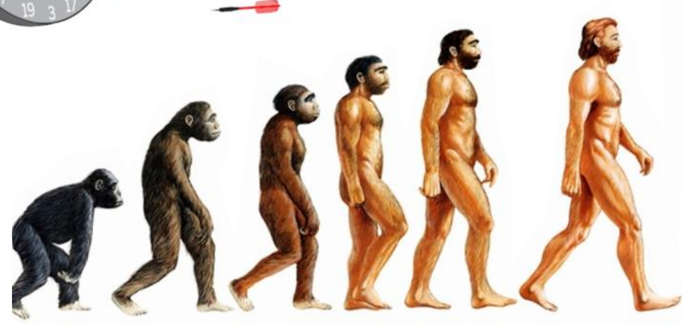
O_2-crystal

	exciting	FHI-AIMS	FLEUR	FPLO	RSPt	WIEN2k	VASP201	VASP2012/VASP
exciting		0.1	0.4	1.8	0.7	0.6	7.7	0.2
FHI-AIMS/tier2	0.1		0.3	1.7	0.6	0.5	7.6	0.1
FLEUR	0.4	0.3		1.4	0.3	0.3	7.2	0.2
FPLO/T+F+s	1.8	1.7	1.4		1.1	1.2	5.8	1.6
RSPt	0.7	0.6	0.3	1.1		0.1	6.9	0.5
WIEN2k/acc	0.6	0.5	0.3	1.2	0.1		7.1	0.4
VASP2011/VASP	7.7	7.6	7.2	5.8	6.9	7.1		7.5
VASP2012/VASP	0.2	0.1	0.2	1.6	0.5	0.4	7.5	

Comparing Solid State DFT Codes, Basis Sets, and Potentials (K. Lejaeghere et al.)

Science 351, aad3000 (2016)





	year	⟨Δ⟩ vs AE
JTH01/ABINIT	2013	1.1
JTH02/ABINIT	2014	0.6
Vdb/CASTEP	1998	6.5
OTFG7/CASTEP	2013	2.6
OTFG9/CASTEP	2015	0.7
GPAW06/GPAW	2010	3.6
GPAW09/GPAW	2012	1.6
PSlib031/QE	2013	1.7
PSlib100/QE	2013	1.0
VASP2007/VASP	2007	2.0
VASP2012/VASP	2012	0.8
VASPGW2015/VASP	2015	0.6

Comparing Solid State DFT Codes, Basis Sets, and Potentials (K. Lejaeghere et al.)

NOVEL MATERIALS DISCOVERY

Use a code and settings (pseudopotentials (if necessary), basis set, etc.) that survived the Delta test.

Code	Version	Basis	Electron treatment	Δ-value meV/atom
WIEN2k	13.1	LAPW/APW+lo	all-elec	0
FHI-aims	081213	tier2 numerical orbitals		0.2
Exciting	development version	LAPW+xlo		0.2
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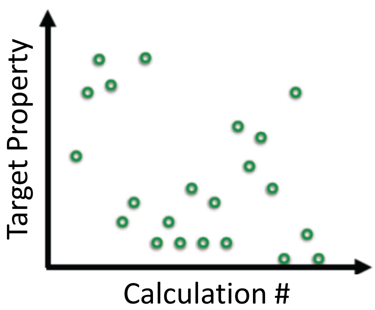
FHI-aims	08	r2 numerical orbitals	all-electron (relativistic zora scalar 1e-12)	0.4
CASTEP	9.0	plane waves	OTFG CASTEP 9.0	0.5

A Delta value of < 0.5 is considered "perfect".
Most codes/pseudopotentials (now) perform with Δ-values between 1 and 2. Δ can be as high as 10.

Find Structure in Big Data That Is *A Priori* “Not Visible”
Data Fitting, Statistical Learning, Machine Learning

Arrange/organize materials with respect to a property and a *set of simple descriptive parameters (a descriptor).*



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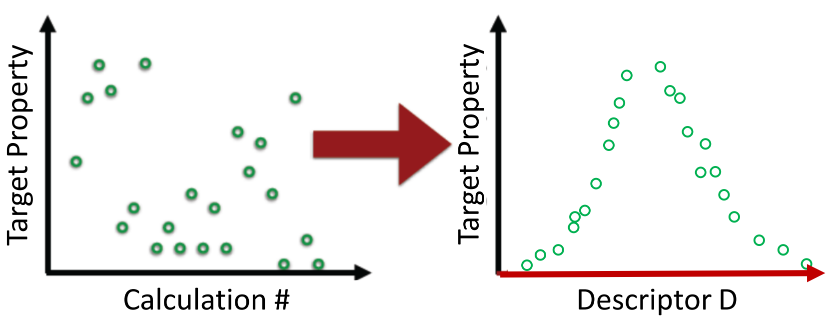

The descriptor can be designed: Rupp, von Lilienfeld, Behler, Csanyi, Seko, Tsuda, …

The descriptor can be selected out of a large set of candidates: Ozolins, Ghiringhelli.

Find Structure in Big Data That Is *A Priori* "Not Visible"
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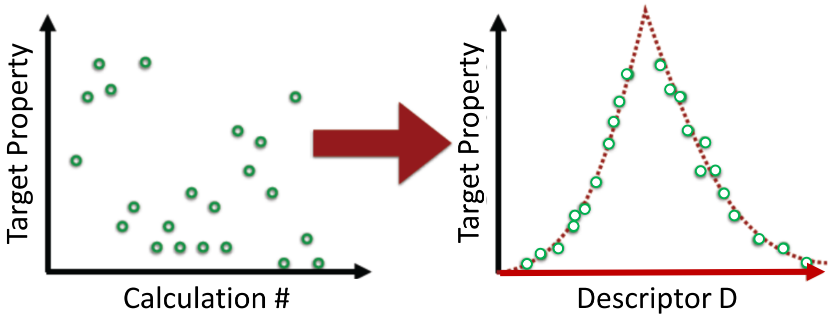

The descriptor can be designed: Rupp, von Lilienfeld, Behler, Csanyi, Seko, Tsuda, …

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More data means a better representation. Will we ever have enough data?

Big-Data-Driven Science vs. Model-Driven Science

NOVEL MATERIALS DISCOVERY

Traditional approach in the empirical sciences (e.g. physics, chemistry):

- Study a few systems
- Build a model,
- Improve the model when needed

(e.g. strength of transition metals Ti, … Fe, … Cu: d-band occupation, etc.).

The new option offered by Big-Data Analytics (and big-data-driven science):

- Find structure in big data that is probably invisible by standard tools.
- Offer many (thousands) of optional models and
- employ applied mathematics/information theory to find out which model is best (e.g. compressed sensing, statistical learning).

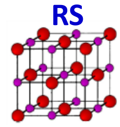
NOVEL MATERIALS DISCOVERY

Proof of Concept: Descriptor for the Classification “Zincblende/Wurtzite or Rocksalt?”

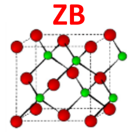
Crystal-structure prediction was and is one of the most important, basic challenges in materials science and engineering.

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?”**

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.

Complexity: $T_s[n]$ and E_{xc}.

NOVEL MATERIALS DISCOVERY

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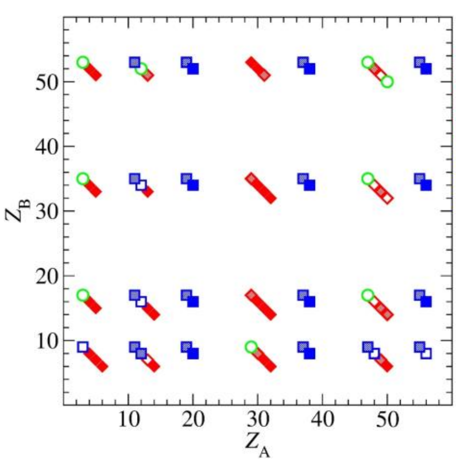
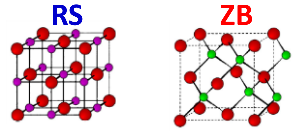


RS **ZB**



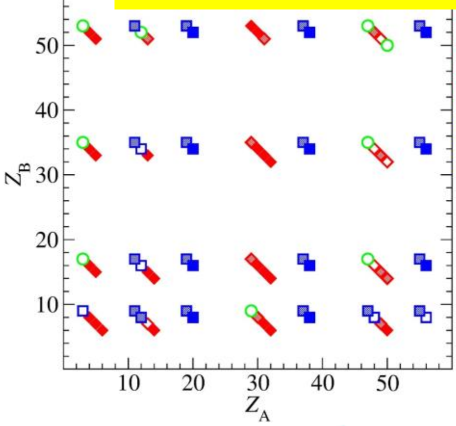
$\Delta = E(\mathrm{RS}) - E(\mathrm{ZB})$

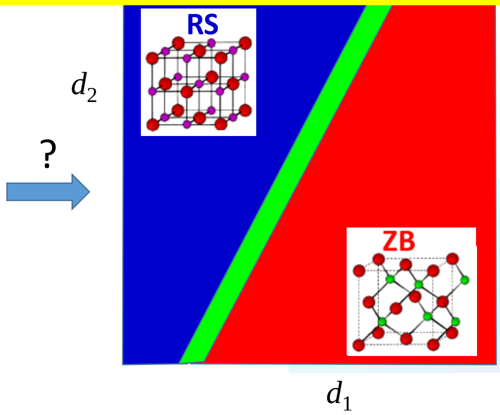
- ZB, $\Delta > 0.2$ eV
- ZB, 0.1 eV $< \Delta \le 0.2$ eV
- ZB, 0.05 eV $< \Delta \le 0.1$ eV
- -0.05 eV $< \Delta \le 0.05$ eV
- RS, -0.1 eV $< \Delta \le -0.05$ eV
- RS, -0.2 eV $< \Delta \le -0.1$ eV
- RS, $\Delta \le -0.2$ eV

Proof of Concept: Descriptor for the Classification "Zincblende/Wurtzite or Rocksalt?"

We need to arrange the data such that statistical learning is efficient. We need a good set of descriptive parameters.

How to find d_1, d_2?
In reality the representation will be higher than 2-dimensional.

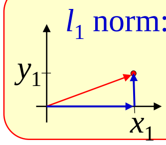




J. A. van Vechten, Phys. Rev. 182, 891 (1969).
J. C. Phillips, Rev. Mod. Phys. 42, 317 (1970).
A. Zunger, Phys. Rev. B 22, 5839 (1980).
D. G. Pettifor, Solid State Commun. 51, 31 (1984).
Y. Saad, D. Gao, T. Ngo, S. Bobbitt, J. R. Chelikowsky, and W. Andreoni, Phys. Rev. B 85, 104104 (2012).

Statistical Learning (Machine Learning), Compressed Sensing

NOVEL MATERIALS DISCOVERY



l_1 norm: $|x_1| + |y_1|$ Manhattan (taxi cab) distance

l_2 norm: sqrt($x_1^2 + y_1^2$)

kernel ridge regression

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

minimize

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

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

linear

R. Tibshirani, J. Royal Statist. Soc. B 58, 267 (1996)

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

$$\sum_{i=1}^{N}(P(\boldsymbol{d}_i) - P_i)^2 \; + \; \lambda\|\boldsymbol{c}\|_1$$

$$\|\boldsymbol{c}\|_1 = \sum_{\alpha=1}^{M}|c_\alpha|$$

least absolute shrinkage and selection operator (LASSO) for feature selection

NOMAD 1) Primary Features, 2) Feature Space, 3) Descriptors

1)

ID	Description — **free atoms**	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

ID	Description — **free dimers**	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

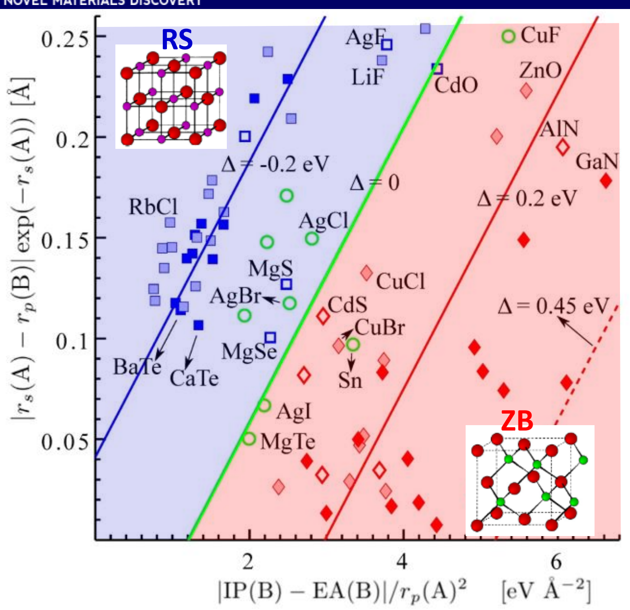
2) We start with 23 primary features and build > 10,000 non linear combinations

3) LASSO finds the descriptors: $\frac{IP(B) - EA(B)}{r_p(A)^2}$, $\frac{|r_s(A) - r_p(B)|}{\exp(r_s(A))}$, $\frac{|r_p(B) - r_s(B)|}{\exp(r_d(A) + r_s(B))}$

NOMAD "The Map" -- Compressed Sensing -- LASSO, 2-Dim. Descriptor

NOVEL MATERIALS DISCOVERY

L.M. Ghiringhelli, J. Vybiral, S.V. Levchenko, C. Draxl, M.S., PRL ***114****, 105503 (2015).*



$\Delta = E(RS) - E(ZB)$
- ZB, $\Delta > 0.2$ eV
- ZB, 0.1 eV $< \Delta \le 0.2$ eV
- ZB, 0.05 eV $< \Delta \le 0.1$ eV
- -0.05 eV $< \Delta \le 0.05$ eV
- RS, -0.1 eV $< \Delta \le -0.05$ eV
- RS, -0.2 eV $< \Delta \le -0.1$ eV
- RS, $\Delta \le -0.2$ eV

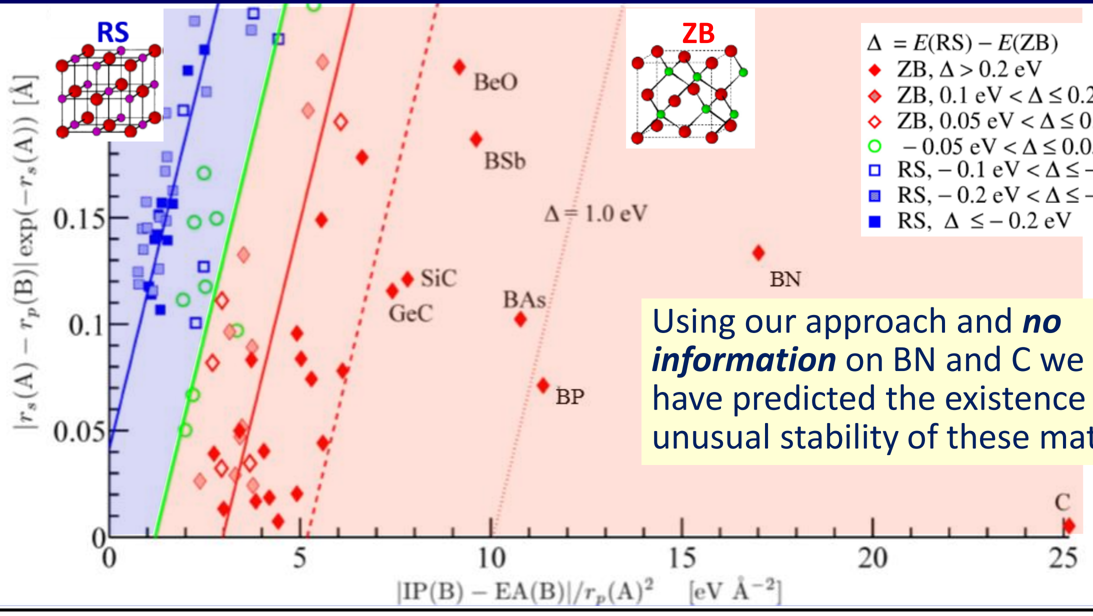
$$P(\mathbf{d}) = \mathbf{d}\mathbf{c}$$

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

NOVEL MATERIALS DISCOVERY

"The Map" -- Compressed Sensing -- LASSO, 2-Dim. Descriptor

L.M. Ghiringhelli, J. Vybiral, S.V. Levchenko, C. Draxl, M.S., PRL ***114****, 105503 (2015).*



Using our approach and ***no information*** on BN and C we would have predicted the existence and unusual stability of these materials.

NOVEL MATERIALS DISCOVERY

Statistical Learning (Machine Learning): Descriptor

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

NOMAD
NOVEL MATERIALS DISCOVERY

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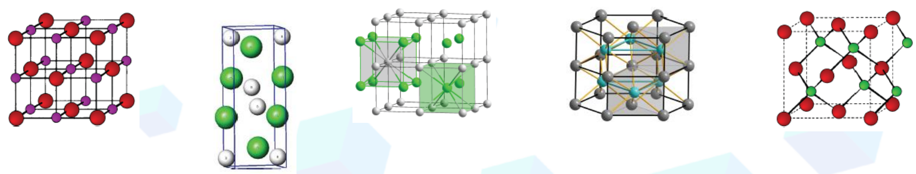
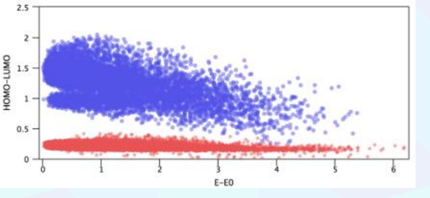
Other Statistical Learning Projects

- Metastabilities of binary compounds, at first considering 5 structures:

Rocksalt CrB CsCl NiAs Zincblende



- Determine the best, correlation-consistent basis functions from a pool of 10,000 Gaussians
- Subgroup discovery algorithms: find structure in big data and analyze what is behind.



Drawing Causal Inference from Big Data (Scientific Insight) -- can we trust a prediction?

Correlation between ***d*** and ***P*** , i.e. ***P*** is a function of ***d***, ***P***(***d***), reflects causal inference
if it is based on sufficient information(*)

Judea Pearl

There are four possibilities (types of causality) behind ***P***(***d***):

1. $d \rightarrow P$: ***P*** "listens" to ***d***
2. $A \rightarrow d$ and $A \rightarrow P$: There is no direct connection between ***d*** and ***P***, but ***d*** and ***P*** both "listen" to a third "actuator"
3. $P \rightarrow d$: ***d*** "listens" to ***P***
4. There is no direct connection between ***d*** and ***P***, but they have a common effect that listens to both and screams: "I occurred" (Berkson bias; Judea Pearl)

(*) Construct ***d*** with scientific knowledge (prejudice?), or use "big data" for $\{P_i\}$.

Drawing Causal Inference from Big Data (Scientific Insight) -- can we trust a prediction?

NOVEL MATERIALS DISCOVERY

LASSO has provided us with an equation for the quantitative energy difference:

$$\Delta E = 0.108 \frac{\mathrm{EA(B)} - \mathrm{IP(B)}}{r_p(\mathrm{A})^2} + 1.790 \frac{|r_s(\mathrm{A}) - r_p(\mathrm{B})|}{\exp(r_s(\mathrm{A}))} + 3.766 \frac{|r_p(\mathrm{B}) - r_s(\mathrm{B})|}{\exp(r_d(\mathrm{A}))} - 0.0267$$

This is an equation, not a scientific law:

Case #2:

Nuclear numbers $Z_{\mathrm{A}}, Z_{\mathrm{B}}$ ⟷ our descriptor

many-body Hamiltonian ⟶ energy differences

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

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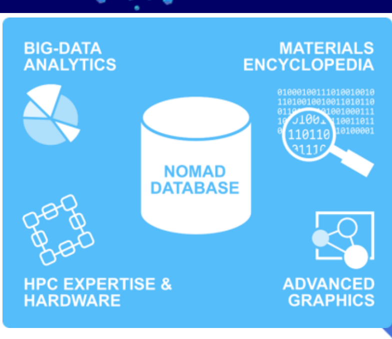
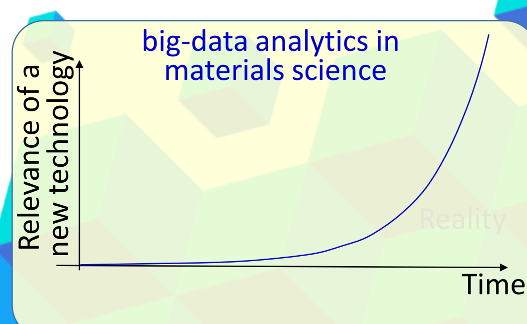


NOMAD develops a *Materials Encyclopedia* and *Big-Data Analytics* and *Advanced Graphics Tools* for materials science and engineering.



The amount of different materials is huge. However, the number of materials that exhibit a certain function, is rather small, i.e. **the space of chemical compounds is sparsely populated**.

We need to develop *domain-specific* compressed-sensing and machine-learning tools.

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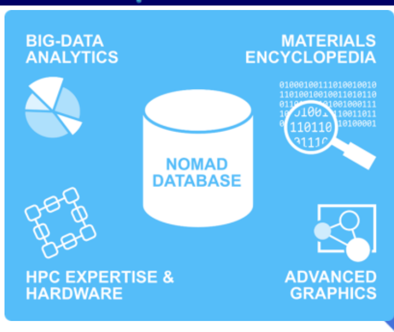

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