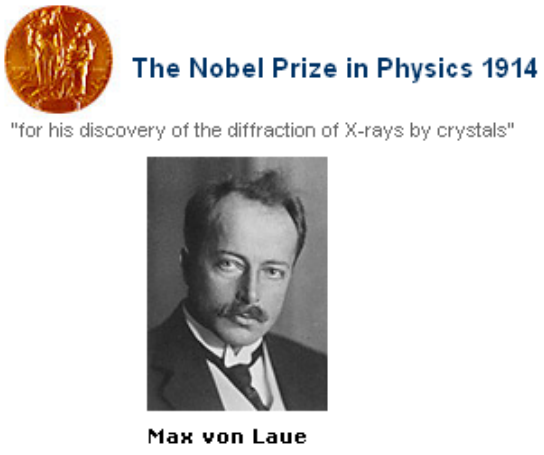
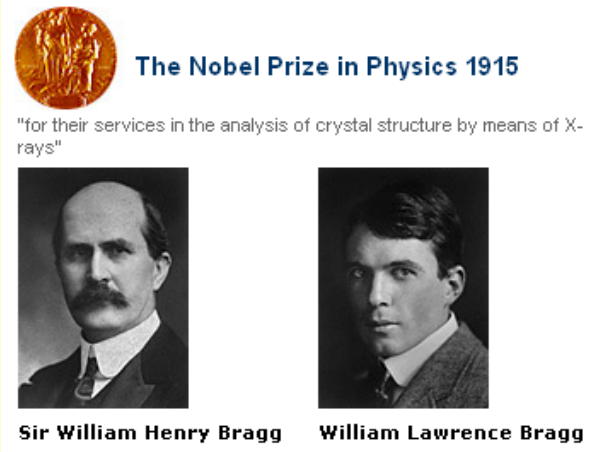
Artificial Intelligence Methods for Discovering Novel Materials and Exotic Chemistry

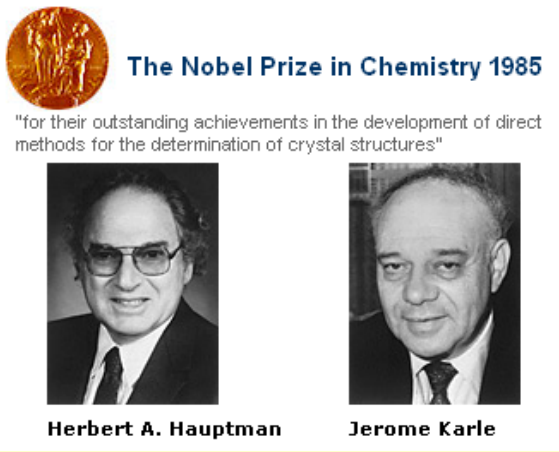
Artem R. Oganov

Skolkovo Institute of Science and Technology, Russia

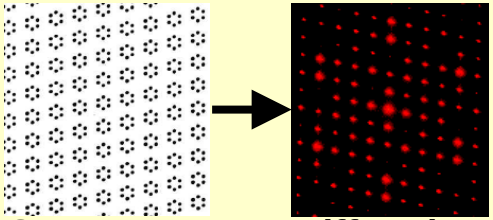
Crystal structure determines physical properties.
Crystal structure determination was a major breakthrough.





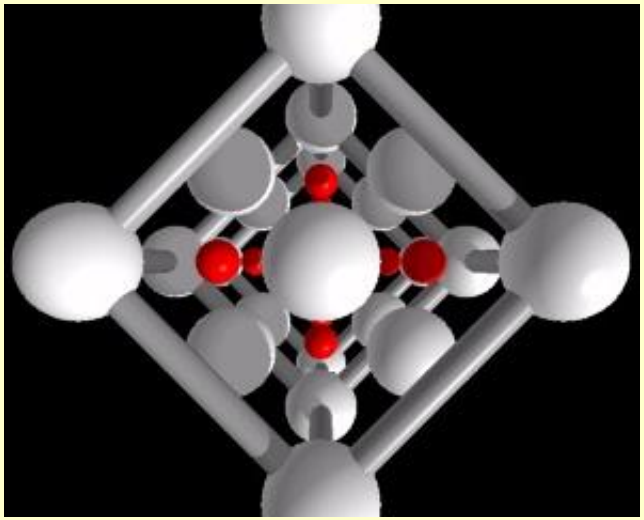


(from http://nobelprize.org)

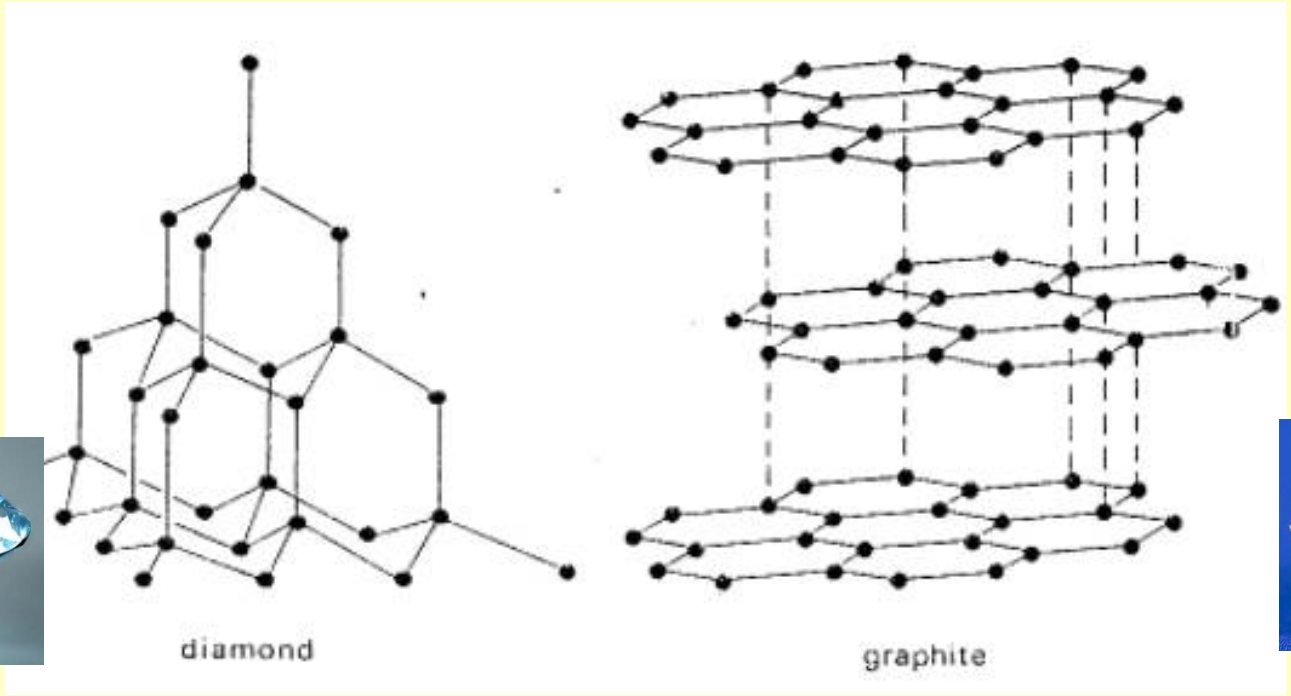


Zincblende ZnS.

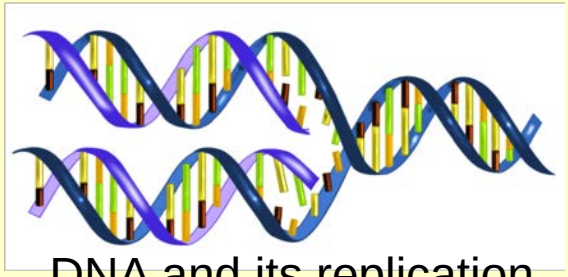
One of the first solved structures (1912-1913)



Structure determines properties of materials



Example: graphite and diamond have the same chemical composition, and their opposite properties are due to their different crystal structures.



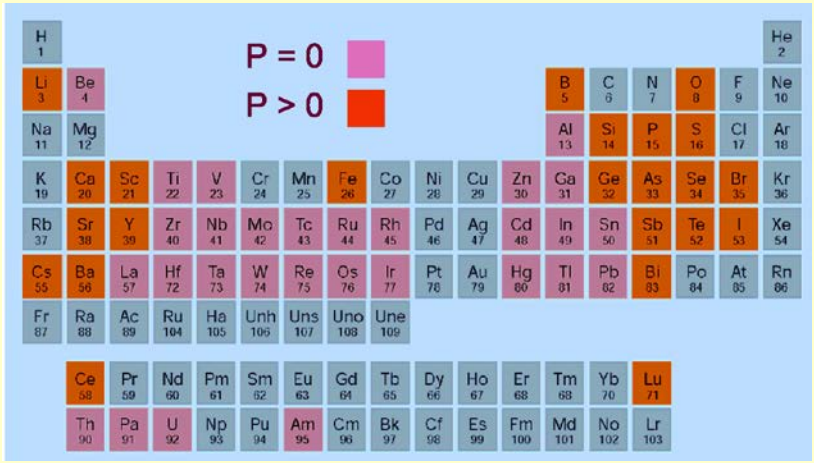
DNA and its replication

Functions of biomolecules are determined by their structure

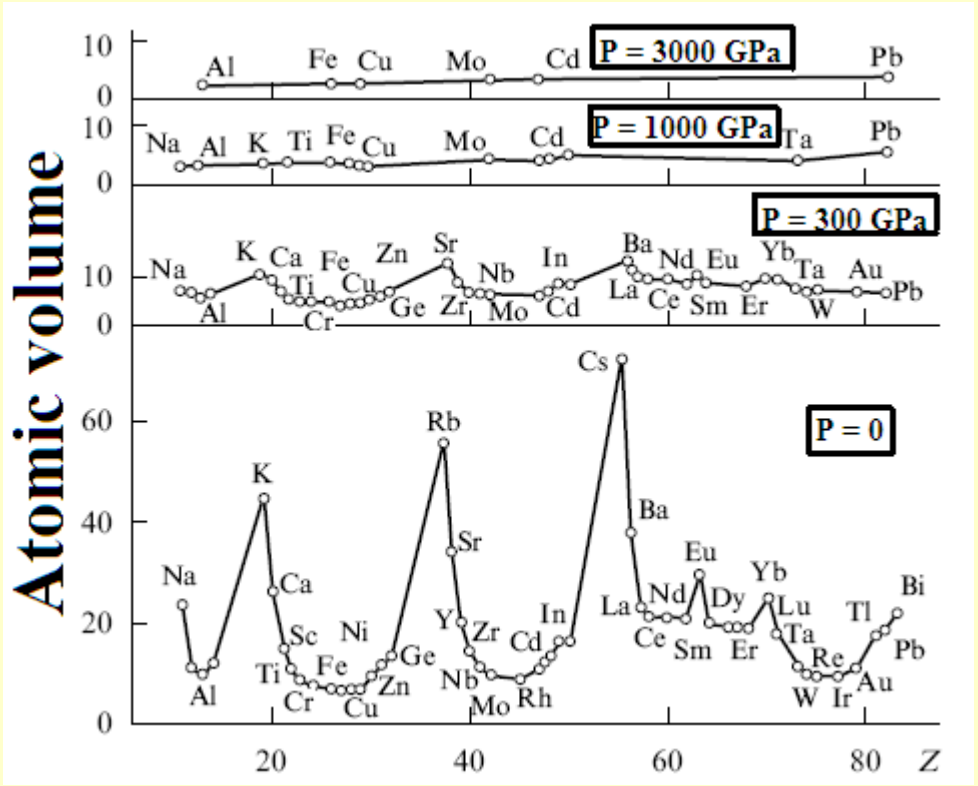
Structure of proteins

Under pressure, Periodic Law breaks down.

We understand the laws of nature only insofar as we understand their limitations

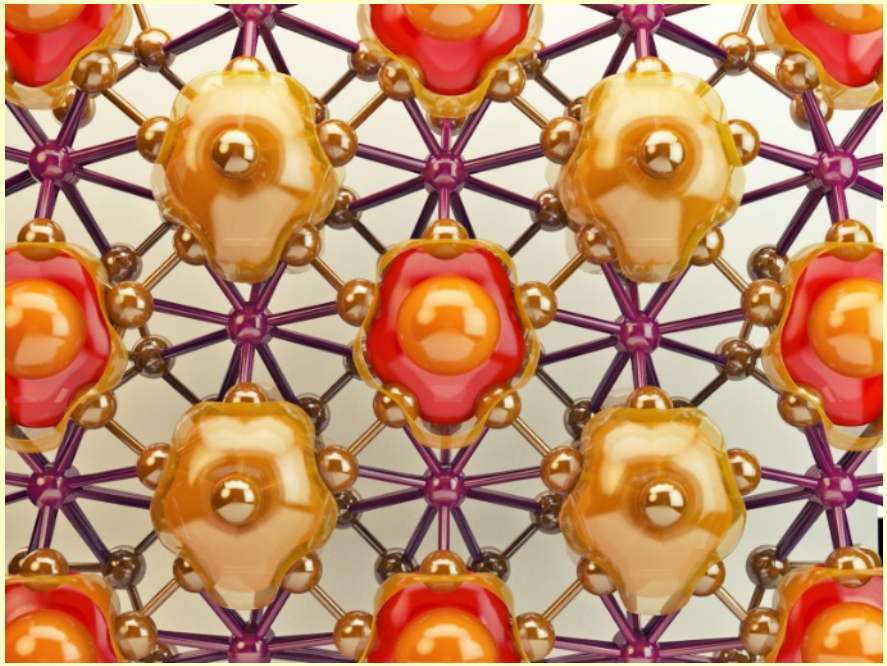


At 100 GPa oxygen becomes a superconductor!

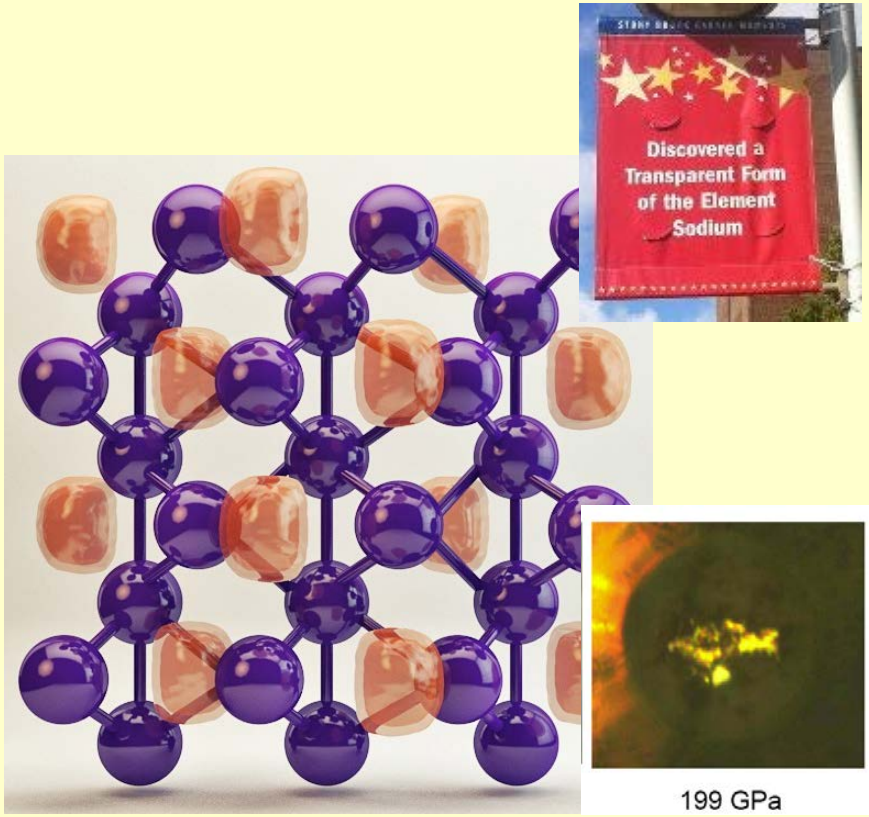


Periodic Law disappears at ultrahigh pressures (Al'tshuler, 1999)

New chemistry of the elements under pressure

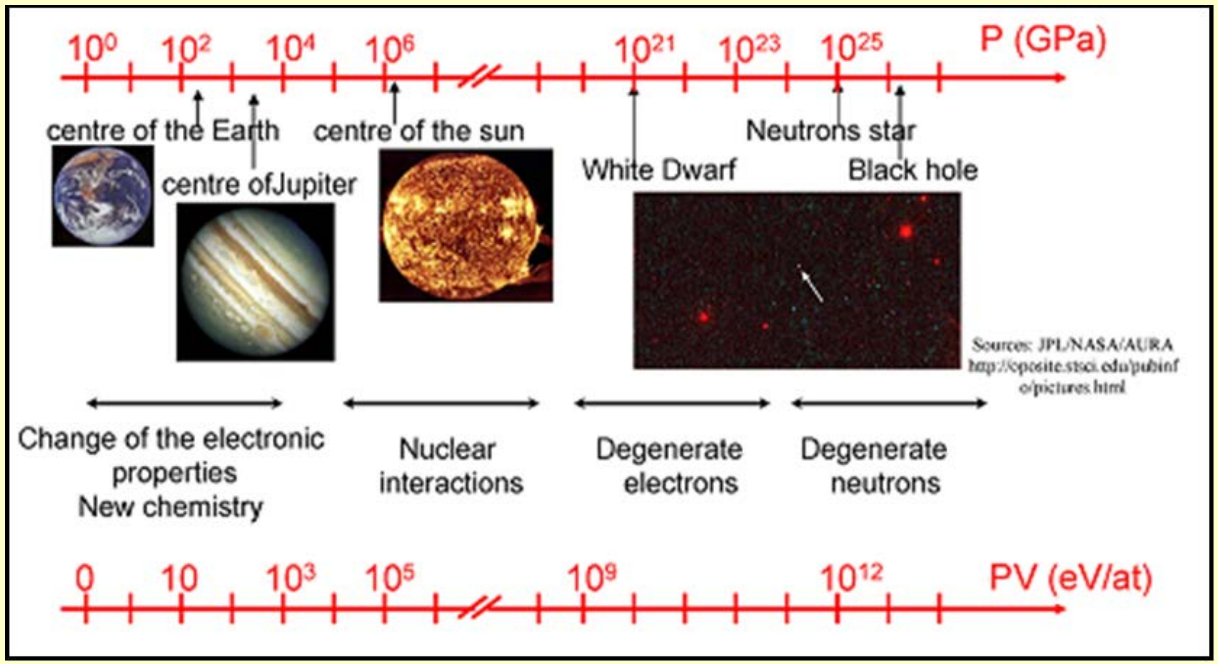


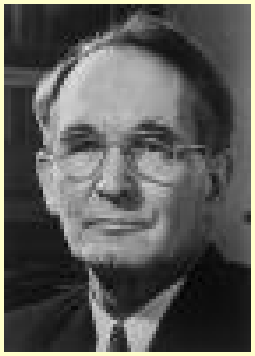
New superhard structure of boron
(Oganov et al., *Nature*, 2009)



High-pressure transparent
allotrope of sodium
(Ma & Oganov, *Nature*, 2009)

Most matter of the Universe is under pressure



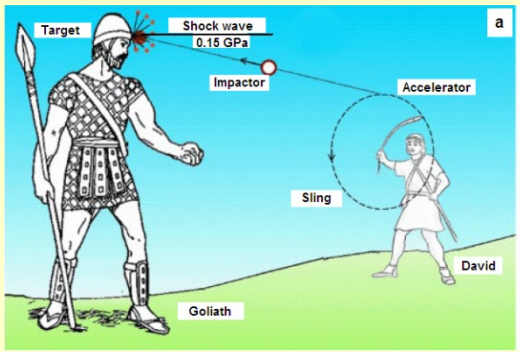


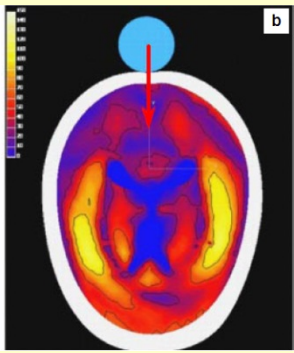
P.W. Bridgman

1946 Nobel laureate (Physics)

<u>Units:</u> 100 GPa = 1 Mbar =

200x





Briefly about big data

Big data have predictive power

We have:
~300,000 experimental (& >500,000 theoretical) inorganic crystal structures. For many of them, we have some physical properties (and for many, we don't).

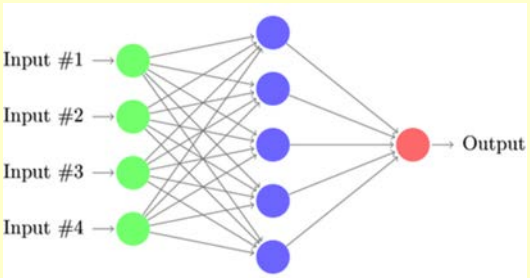
Many studied syntheses of compounds.

Many studied industrial production processes.

Big data analysis gives:
Fast predictions

Predictions where the full calculation would be too complicated.

Predictions where there's no theory.

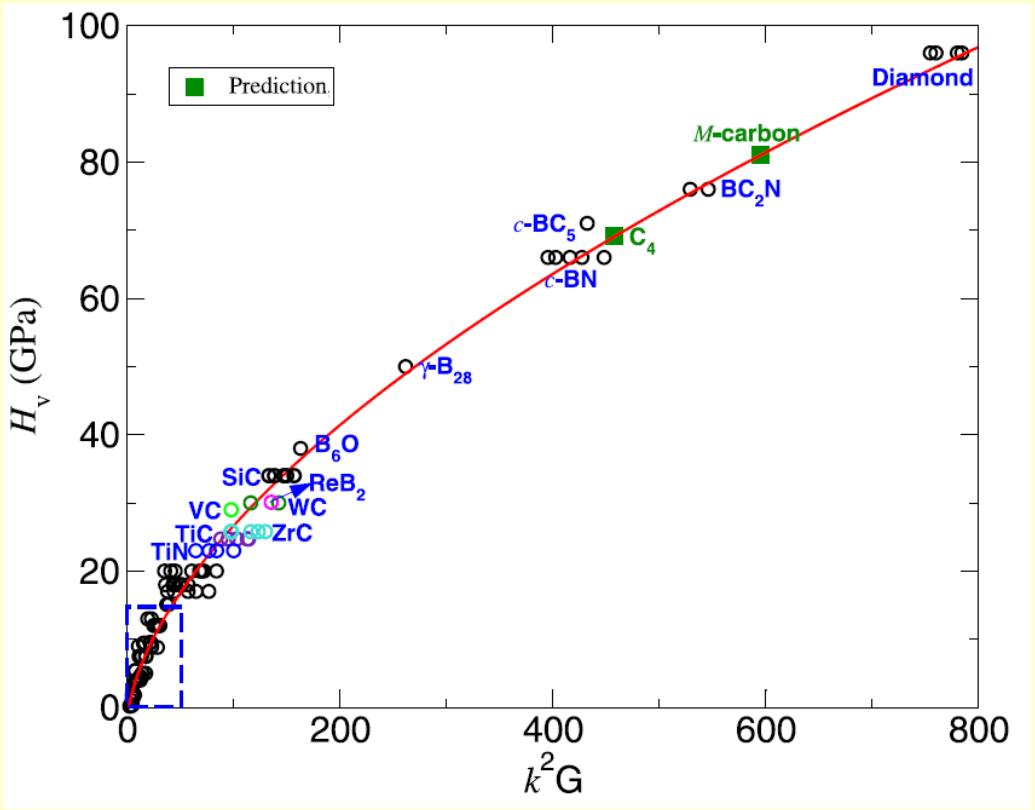


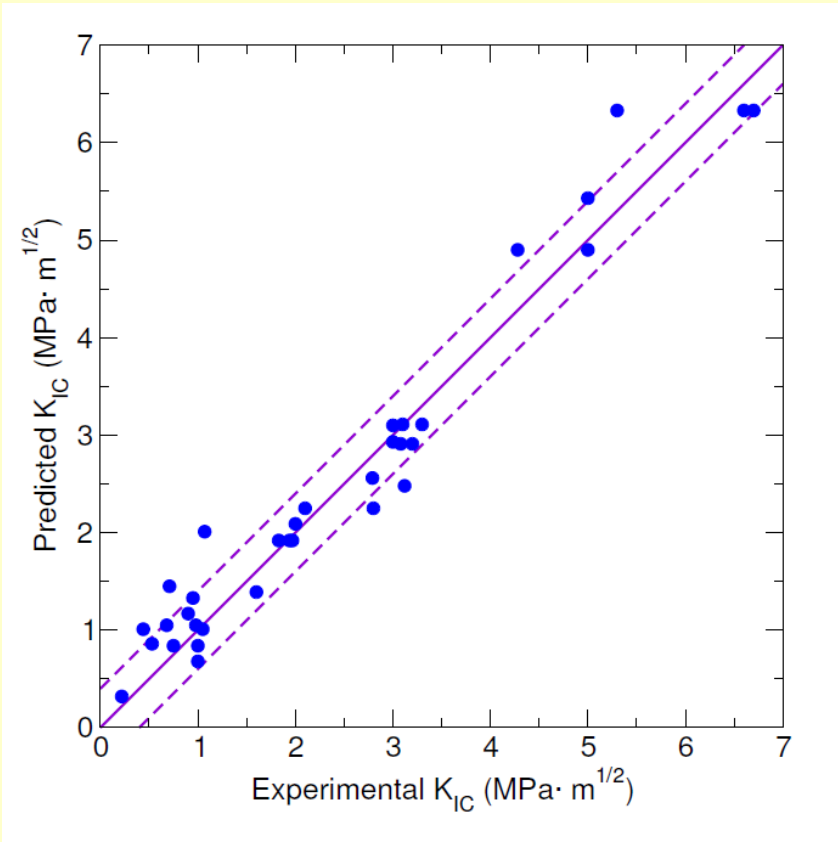
			Ti = 50	Zr = 90	? = 180	
			V = 51	Nb = 94	Ta = 182	
			Cr = 52	Mo = 96	W = 186	
			Mn = 55	Rh = 104,4	Pt = 197,4	
			Fe = 56	Ru = 104,4	Ir = 198	
		Ni =	Co = 59	Pd = 106,6	Os = 199	
H = 1			Cu = 63,4	Ag = 108	Hg = 200	
	Be = 9,4	Mg = 24	Zn = 65,2	Cd = 112		
	B = 11	Al = 27,4	? = 68	Ur = 116	Au = 197?	
	C = 12	Si = 28	? = 70	Sn = 118		
	N = 14	P = 31	As = 75	Sb = 122	Bi = 210?	
	O = 16	S = 32	Se = 79,4	Te = 128?		
	F = 19	Cl = 35,5	Br = 80	J = 127		
Li = 7	Na = 23	K = 39	Rb = 85,4	Cs = 133	Tl = 204	
		Ca = 40	Sr = 87,6	Ba = 137	Pb = 207	
		? = 45	Ce = 92			
		?Er = 56	La = 94			
		?Yt = 60	Di = 95			
		?In = 75,6	Th = 118?			

Predicting complex properties: hardness [Chen et al., 2011] and fracture toughness [Niu & Oganov, 2019]

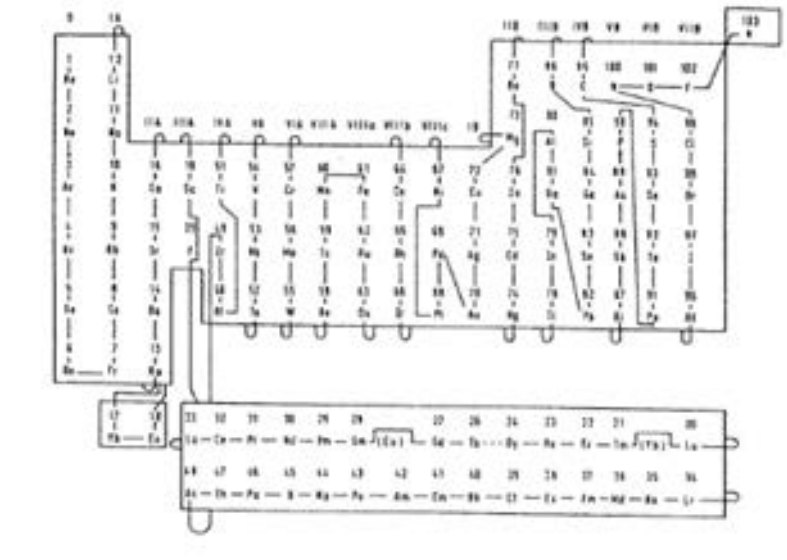
$$H_v = 2\left(k^2 G\right)^{0.585} - 3$$

$$K_{IC} = (1+\alpha)\cdot V^{1/6}\cdot G\cdot (B/G)^{1/2}$$





Mendeleev number (Pettifor, 1984). Prediction of stability, structure, and properties of materials



Mendeleev numbers of the elements

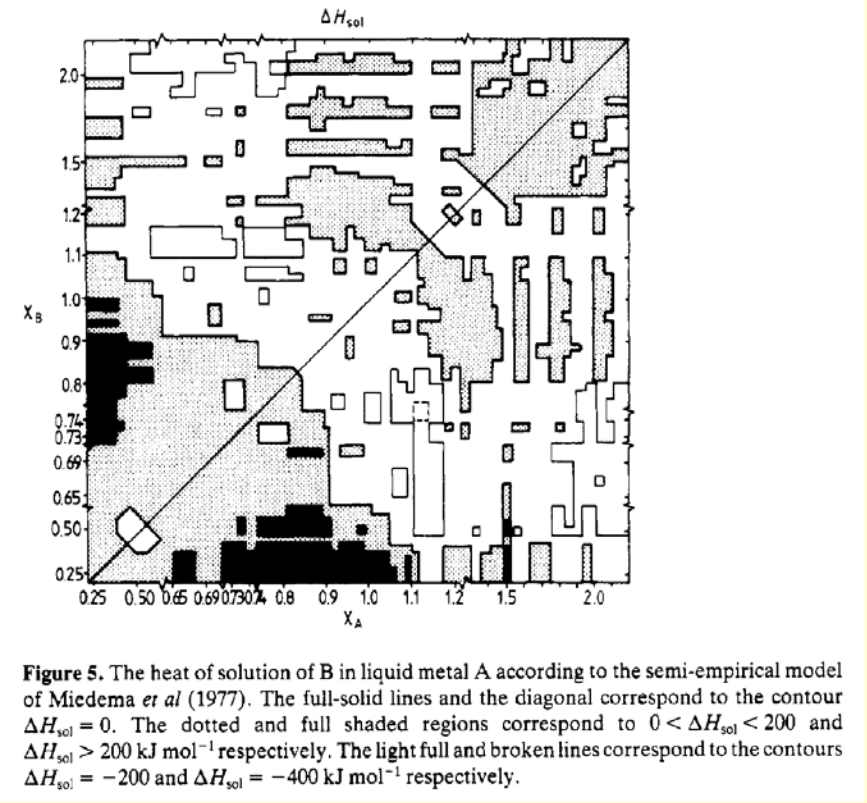
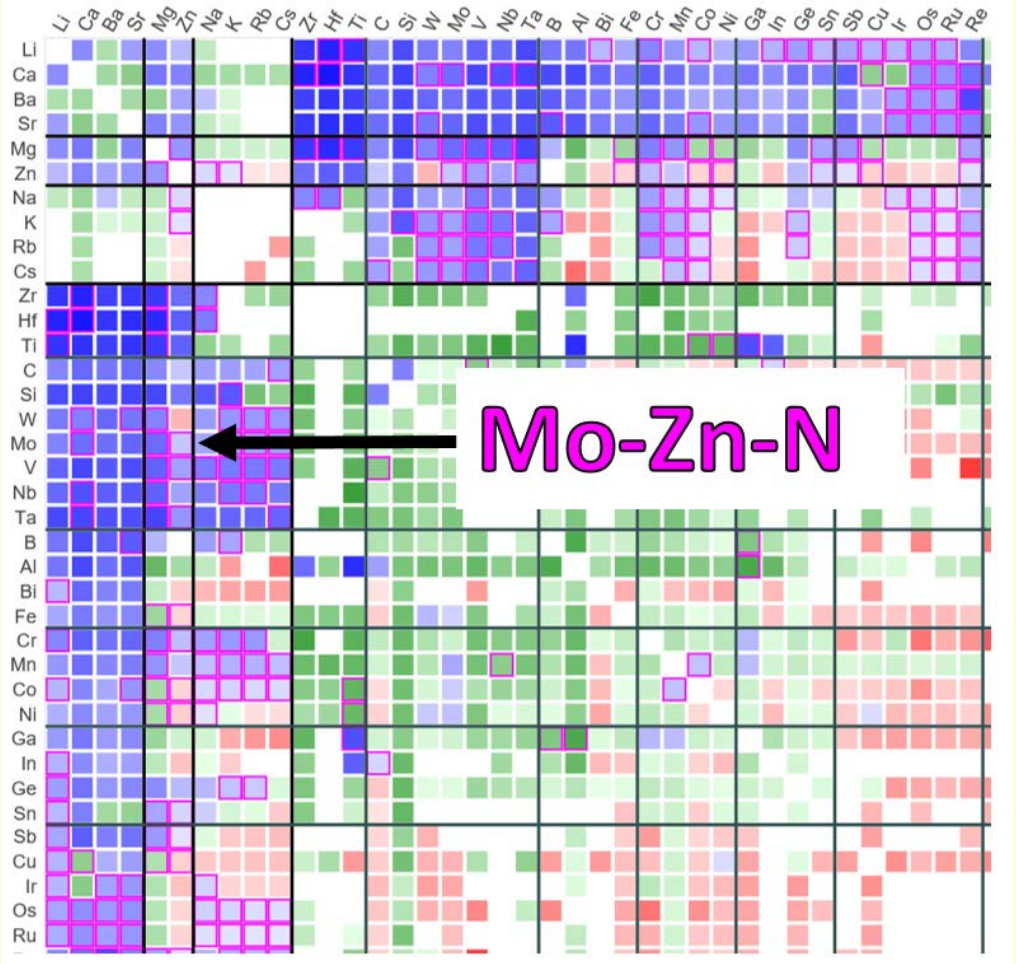


Figure 5. The heat of solution of B in liquid metal A according to the semi-empirical model of Miedema *et al* (1977). The full-solid lines and the diagonal correspond to the contour $\Delta H_{sol} = 0$. The dotted and full shaded regions correspond to $0 < \Delta H_{sol} < 200$ and $\Delta H_{sol} > 200$ kJ mol^{-1} respectively. The light full and broken lines correspond to the contours $\Delta H_{sol} = -200$ and $\Delta H_{sol} = -400$ kJ mol^{-1} respectively.

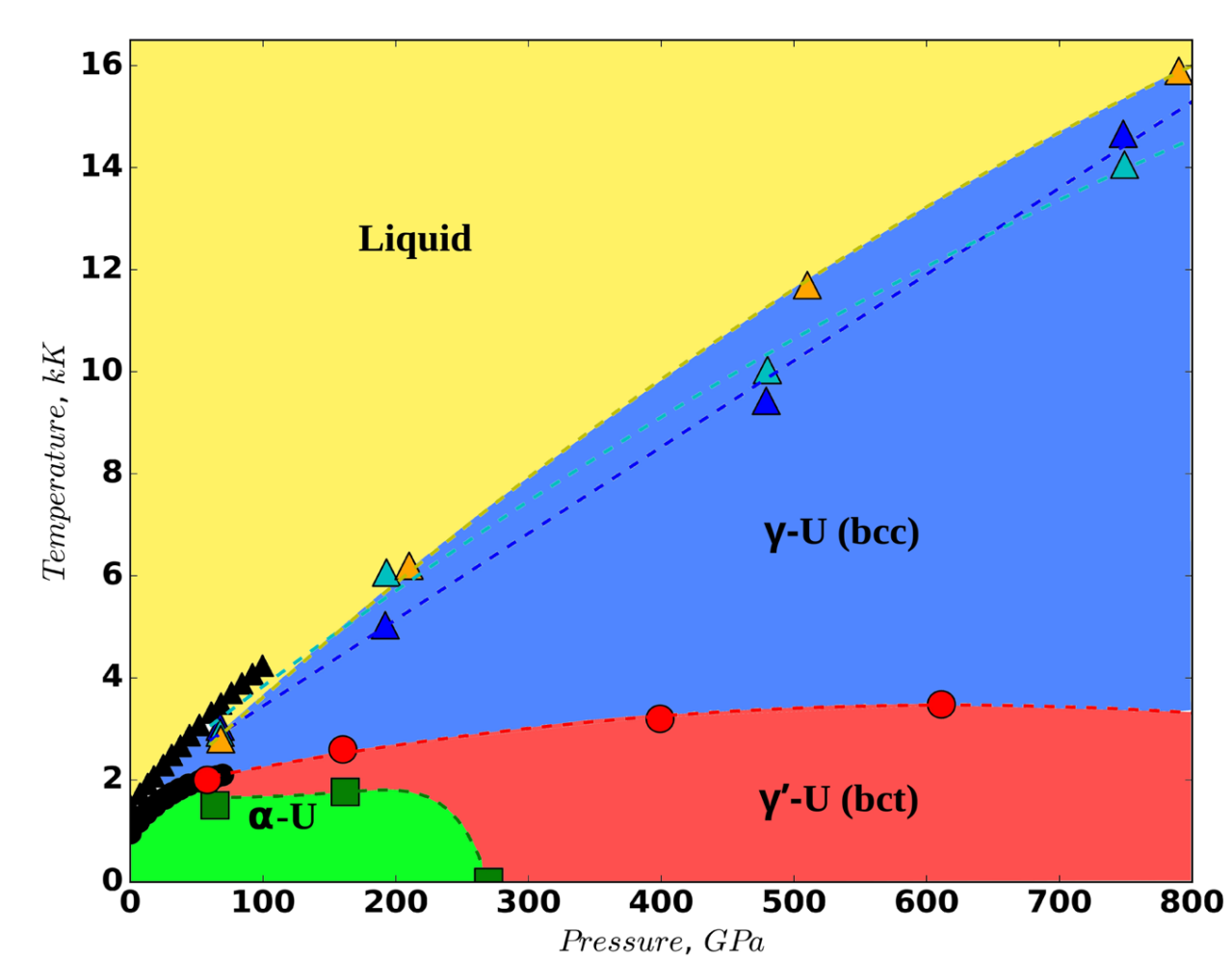
Enthalpies of formation of compounds

Example: search for new ternary nitrides (Sun, 2019)



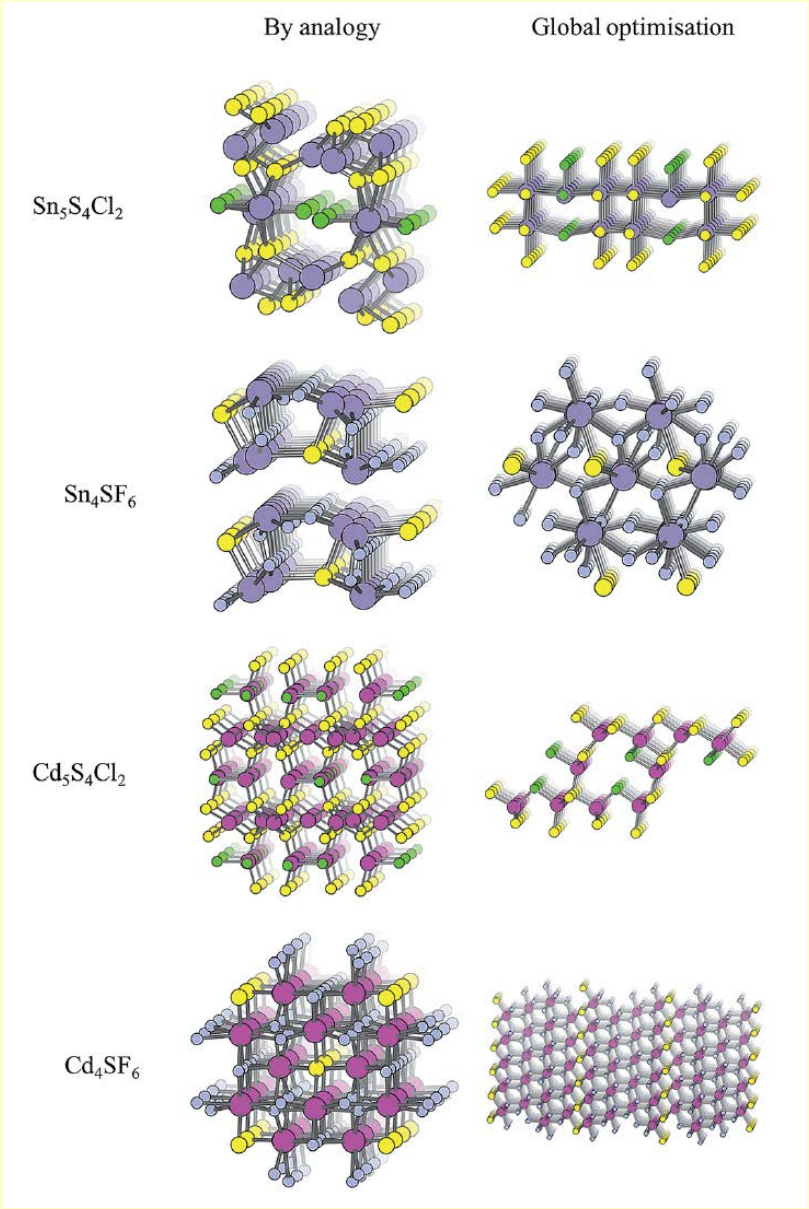
916 systems
246 contain stable nitrides
In 127 of them nitrides were not experimentally known

Machine learning can be used for accelerating simulations without loss of accuracy



Phase diagram of uranium obtained with machine learning (Kruglov & Oganov, submitted)

Data mining gives quick-n-reasonable answers, but beware!



Davies & Oganov (2018): 4 interesting semiconductors, and for each data mining gave a wrong structure:
-for $Sn_5S_4Cl_2$ by 24.7 meV/atom,
-for Sn_4SF_6 by 5.1 meV/atom,
-for Cd_4SF_6 by 0.2 meV/atom,
-for $Cd_5S_4Cl_2$ by 33.3 meV/atom.

Crystal Structure Prediction: Impossible is Possible

-Crystal structures and stable stoichiometries can be predicted
-New tool to explore matter at extreme conditions & discover novel materials
-Evolutionary crystal structure prediction, Maxwell's convex hull construction

Until recently, experiment was the only source of crystal structures

The Nobel Prize in Physics 1914

"for his discovery of the diffraction of X-rays by crystals"

Max von Laue

The Nobel Prize in Physics 1915

"for their services in the analysis of crystal structure by means of X-rays"

Sir William Henry Bragg

William Lawrence Bragg

The Nobel Prize in Chemistry 1985

"for their outstanding achievements in the development of direct methods for the determination of crystal structures"

Herbert A. Hauptman

Jerome Karle

(from http://nobelprize.org)

Acc. Chem. Res. **1994,** *27,* 309−314

Are Crystal Structures Predictable?

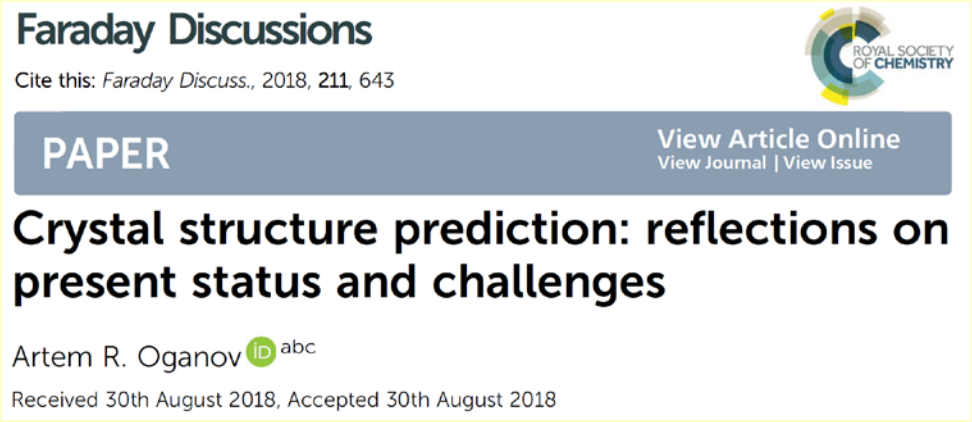
ANGELO GAVEZZOTTI*

"No": by just writing down this concise statement, in what would be the first one-word paper in the chemical literature, one could safely summarize the present state of affairs

$$C = \frac{1}{(V/\delta^3)} \frac{(V/\delta^3)!}{[(V/\delta^3) - N]!N!}$$

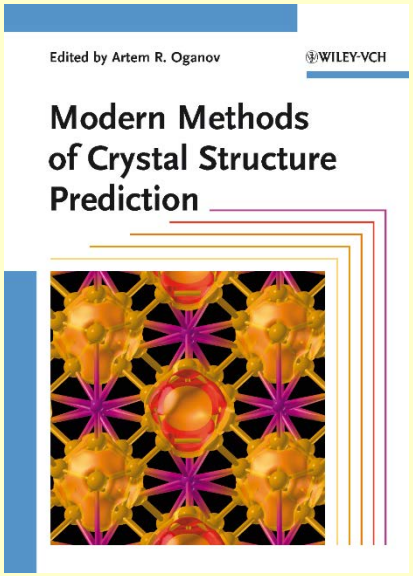
- Crystal structure prediction is an NP-hard problem. Intractable?

Introduction(s) to crystal structure prediction

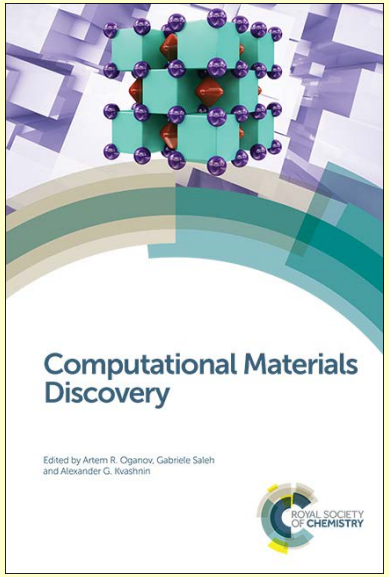


***Faraday Discussions* (2018)**

***Nature Reviews Materials* (2019)**

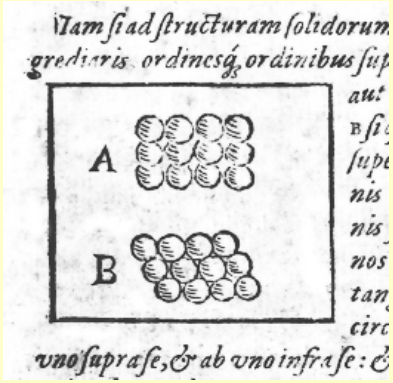


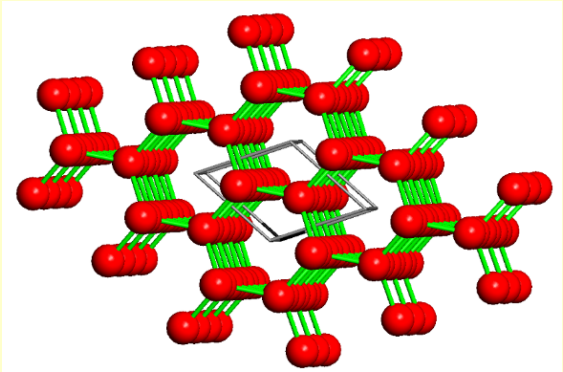
2011



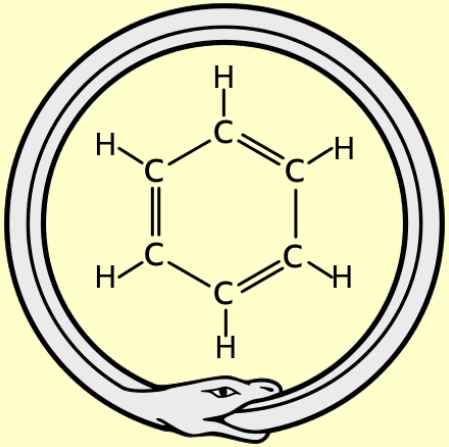
2018

…Kepler's, Kekule's and Barlow's "predictions" we will rather call strikes of intuition

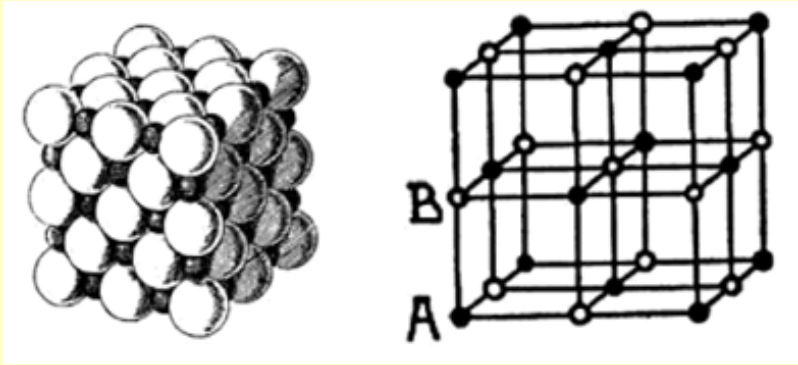




Kepler's (1611) and modern models of the structure of ice



Kekule's vision of the structure of benzene (1857)



Barlow's (1897) and Bragg's (1913) models of the structure of NaCl

The USPEX project
(Universal Structure Predictor: Evolutionary Xtallography)

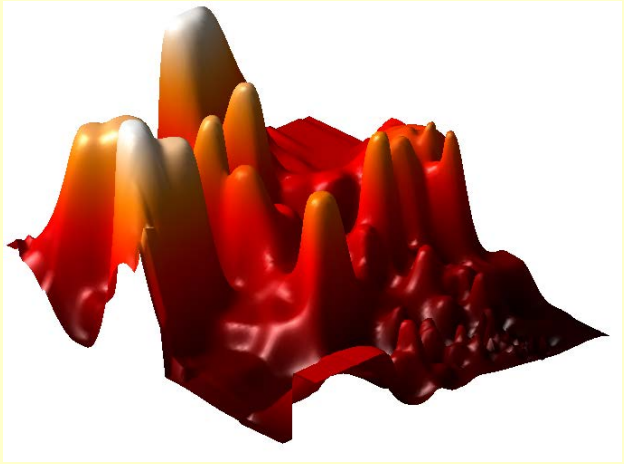
http://uspex-team.org

[Oganov A.R., Glass C.W., *J.Chem.Phys.* 124, 244704 (2006)]

- **Combination of evolutionary algorithm and quantum-mechanical calculations.**
- **>5000 users.**

- **Solves «intractable» problem of structure prediction**
  -3D, 2D, 1D, 0D –systems,
  -prediction of phase transition mechanisms.

- **Interfaced with: VASP, Quantum Espresso, CASTEP, FHI-aims, ABINIT, Siesta, Gaussian, ORCA, ATK, DFTB, MOPAC, GULP, LAMMPS, Tinker, DMACRYS**



Energy landscape of Au_8Pd

W. Kohn

J. P. Perdew

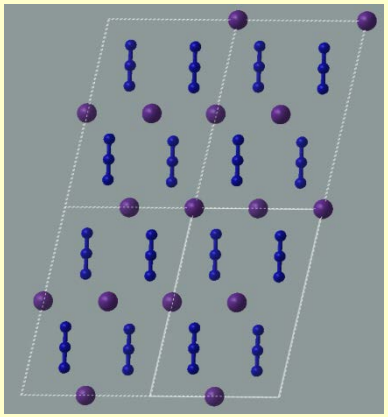
$$(-\frac{\nabla^2}{2} + v_{e-n}[\rho(\mathbf{r})] + v_H[\rho(\mathbf{r})] + v_{xc}[\rho(\mathbf{r})])\phi_i(\mathbf{r}) = \varepsilon_i\phi_i(\mathbf{r})$$

$$E_{GGA,\mathrm{xc}} = \int \mathrm{d}\mathbf{r}\, F_{\mathrm{xc}}(\rho, \frac{|\nabla\rho|}{2k_{\mathrm{F}}\rho(\mathbf{r})})\rho(\mathbf{r})e_{\mathrm{x}}[\rho(\mathbf{r})]$$

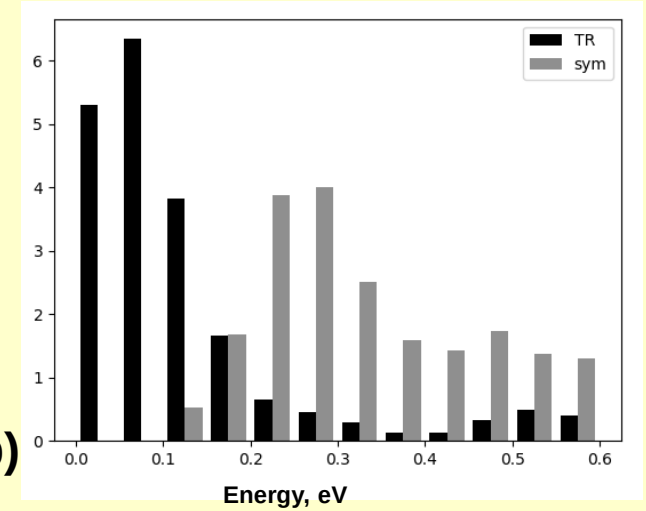
New methodological development: topological structure generator (Bushlanov, Blatov, Oganov, 2019)

Speedup ~3 times

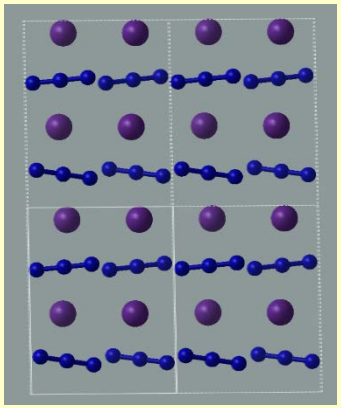
(a)



(b)



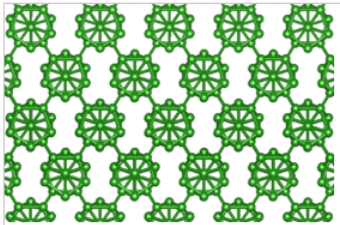
(c)



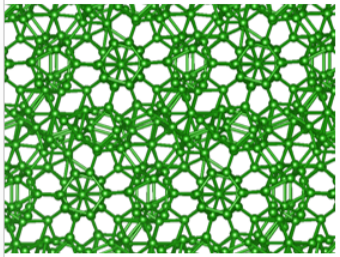
Example of KN_3: (a) topological structure, (c) random symmetric structure, (c) energy distribution of topological (TR) and random symmetric structures

Statistics (100 runs) of USPEX performance on $MgAl_2O_4$ (28 atoms/cell) at 100 GPa

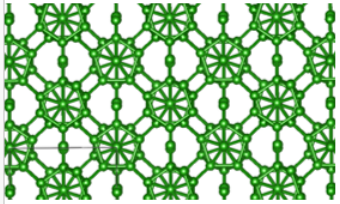
	Old USPEX	On-the-fly adaptation	Adaptation +topology	GPSO CALYPSO	SABC CALYPSO
<#structures>	1307	1069	368	1443	611
Success rate	100%	100%	100%	86%	100%



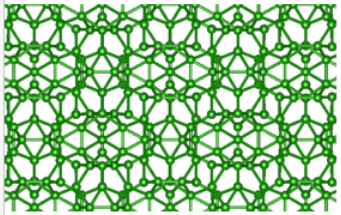
α-boron
E^{DFT} = –6.706 eV/atom
Atoms: 12,
Space group: *R*-3*m*,
Core-hours: 10^3 AL-MTP vs. $3 \cdot 10^3$ DFT
$|E^{DFT} - E^{MTP}|$ = 28.6 meV/atom



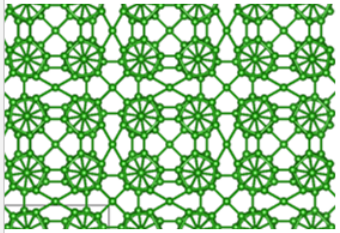
β-boron approximant
E^{DFT} = –6.704 eV/atom,
Atoms: 106,
Space group: *P*1,
Core-hours: $7 \cdot 10^3$ AL-MTP vs. $6.6 \cdot 10^7$ DFT
$|E^{DFT} - E^{MTP}|$ = 10.1 meV/atom



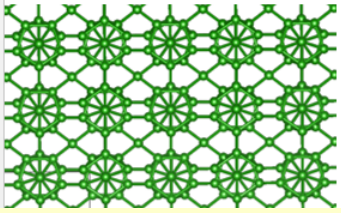
γ-boron
E^{DFT} = –6.678 eV/atom
Atoms: 28,
Space group: *Pnnm*,
Core-hours: $2 \cdot 10^3$ AL-MTP vs. $2.5 \cdot 10^4$ DFT
$|E^{DFT} - E^{MTP}|$ = 58.1 meV/atom



E^{DFT} = –6.667 eV/atom,
Atoms: 54,
Space group: *Im*-3,
Core-hours: $3 \cdot 10^3$ AL-MTP vs. $3.5 \cdot 10^5$ DFT
$|E^{DFT} - E^{MTP}|$ = 7.3 meV/atom



E^{DFT} = –6.667 eV/atom,
Atoms: 52,
Space group: *P*-42*m*,
Core-hours: $3 \cdot 10^3$ AL-MTP vs. $3.2 \cdot 10^5$ DFT
$|E^{DFT} - E^{MTP}|$ = 37.3 meV/atom



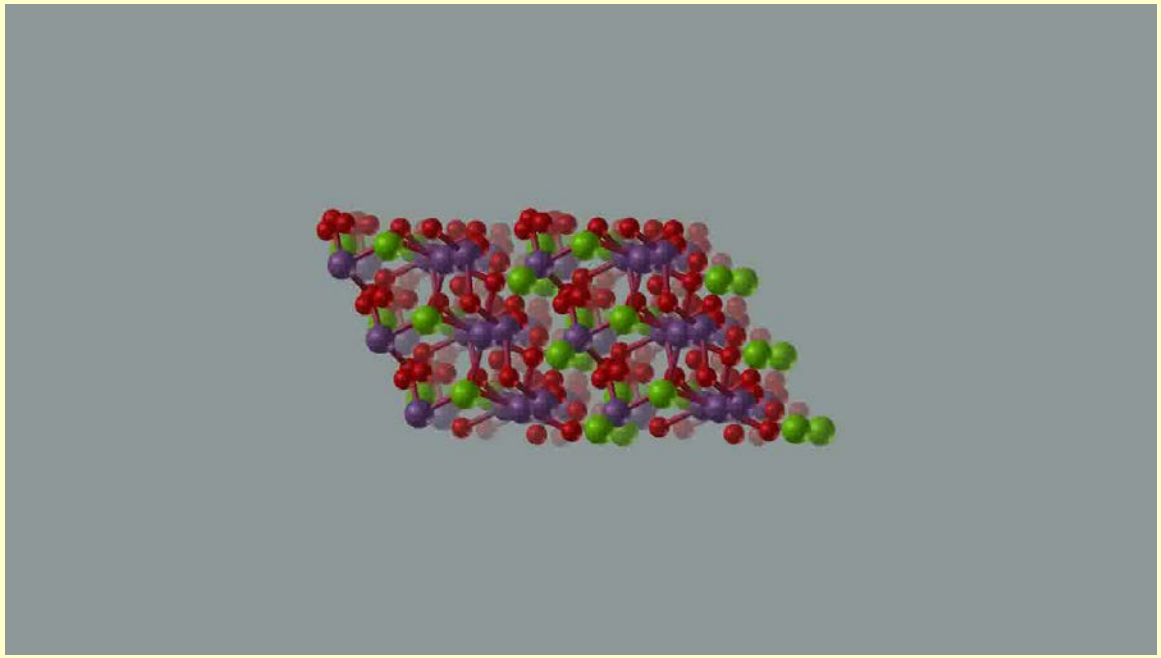
E^{DFT} = –6.665 eV/atom,
Atoms: 26,
Space group: *Cccm*,
Core-hours: $2 \cdot 10^3$ AL-MTP vs. $2.1 \cdot 10^4$ DFT
$|E^{DFT} - E^{MTP}|$ = 13.6 meV/atom

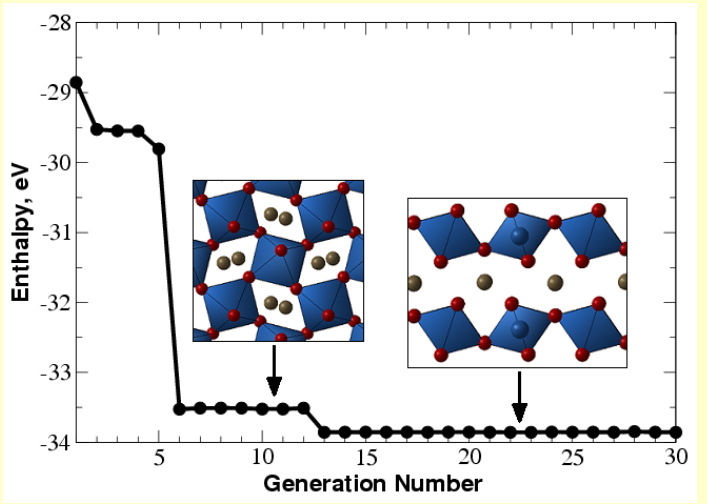
Handling complexity with machine learning: boron allotropes

(E.Podryabinkin, E. Tikhonov, A. Shapeev, A.R. Oganov, PRB, 2019)

- **ML potential with active learning (Shapeev, 2018). 800 parameters.**
- **MAE = 11 meV/atom.**
- **Reproduced α-, β-, γ-, T52 phases of boron.**
- **Predicted low-energy metastable cubic cI54 phase.**
- **Speedup by 100-10,000 times.**

Test: $MgSiO_3$ at 120 GPa





<u>120 GPa:</u> post-perovskite is stable

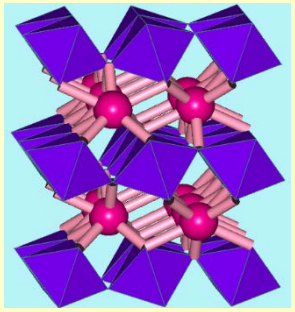
[Oganov & Glass, J.Chem.Phys. 2006]

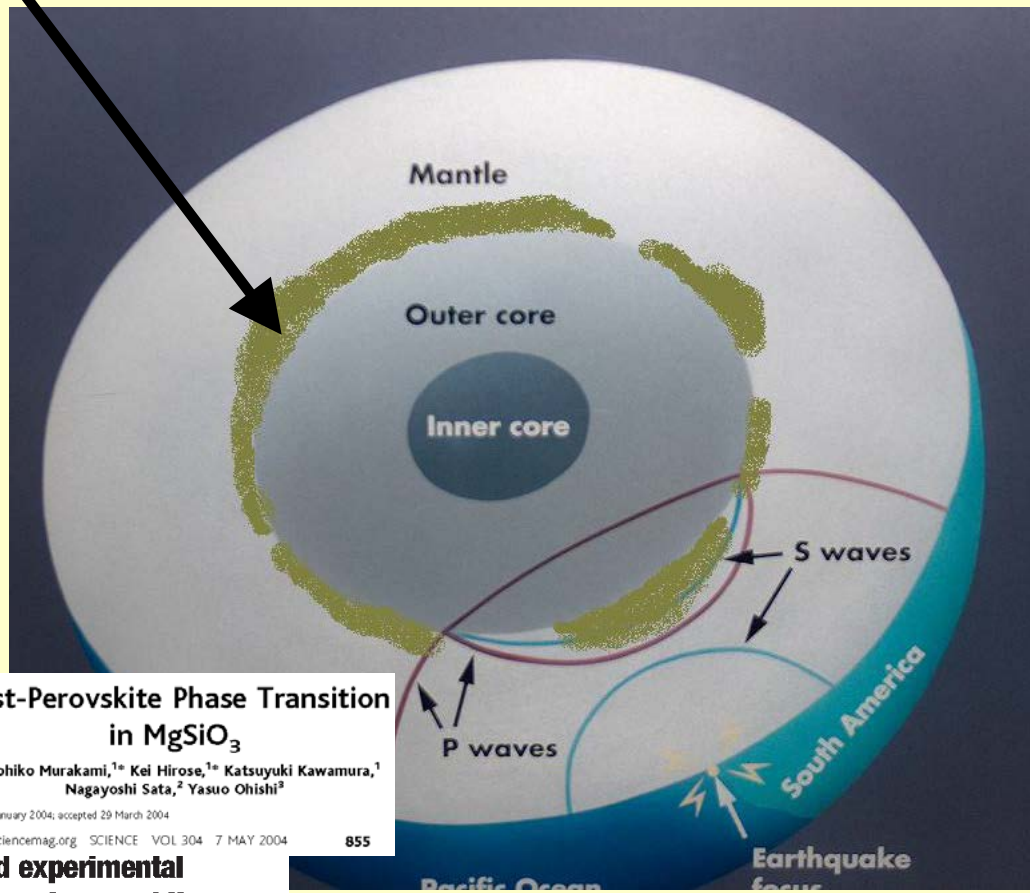
Properties of D” layer (2700-2890 km) were explained by $MgSiO_3$ post-perovskite in 2004

D” – root of hot spots

$MgSiO_3$ makes ~75 vol.% of lower mantle

Anomalies of D”:
seismic discontinuity,
anisotropy





Post-Perovskite Phase Transition in $MgSiO_3$

Motohiko Murakami,[1*] Kei Hirose,[1*] Katsuyuki Kawamura,[1] Nagayoshi Sata,[2] Yasuo Ohishi[3]

22 January 2004; accepted 29 March 2004

Theoretical and experimental evidence for a post-perovskite phase of $MgSiO_3$ in Earth's D'' layer

Artem R. Oganov[1] & Shigeaki Ono[2]

NATURE | VOL 430 | 22 JULY 2004 | www.nature.com/nature
Received 24 March; accepted 27 May 2004; doi:10.1038/nature02701.

Is there a post-post-perovskite?

Mg-Si-O (super)Earths

No: $MgSiO_3$ post-perovskite decomposes inside super-Earths, expect complex structure of super-Earths:

$MgSiO_3 \rightarrow Mg_2SiO_4 + MgSi_2O_5 \rightarrow Mg_2SiO_4 + SiO_2 \rightarrow MgO + SiO_2$ (T < 6500 K)

$MgSiO_3 \rightarrow Mg_2SiO_4 + MgSi_2O_5 \rightarrow MgSi_2O_5 + MgO \rightarrow MgO + SiO_2$ (T > 6500 K)

[Niu & Oganov, *Sci. Rep.* 2015]

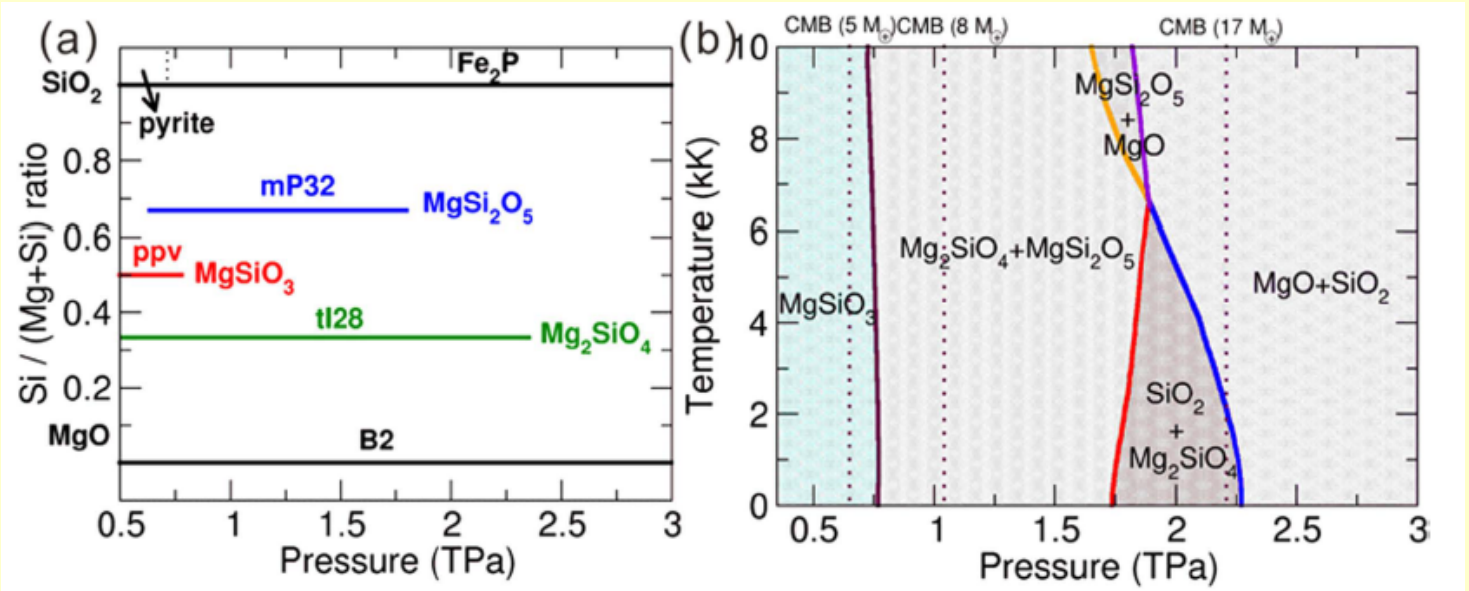
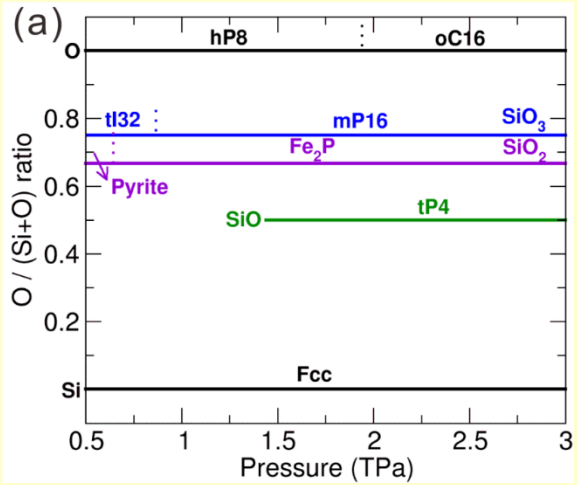
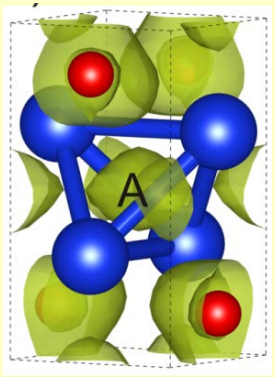


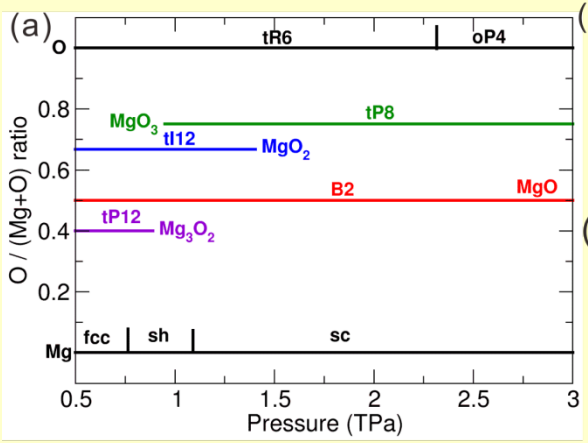
Figure 4. (a) Pressure-composition phase diagram of the pseudo-binary MgO-SiO_2 system. (b) *P-T* phase diagram of $MgSiO_3$. The core-mantle boundary (CMB) pressures of super-Earths and mega-Earths with 5, 8 and 17 $M_\oplus$ are also plotted by vertical dashed lines.

Do we really know what's inside planets? "Forbidden" MgO_2, Mg_3O_2, SiO_3, AlO_2 etc. are stable at planetary pressures





Si-O phase diagram and structure of SiO
(Niu & Oganov, *Sci. Rep.* 2015)

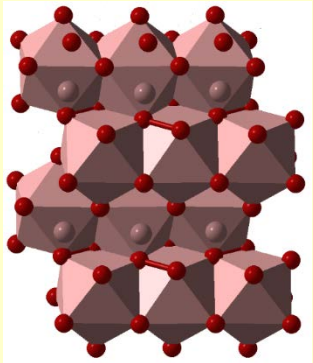


Mg-O phase diagram and structure of MgO_3
(Niu & Oganov, *Sci. Rep.* 2015;
Zhu & Oganov, *PCCP* 2012)

Al-O system:

Stable aluminum "oxide peroxides":
$Al_4O_7 = Al_4O_5[O_2]$, stable at 330-443 GPa
$AlO_2 = Al_2O_2[O_2]$, stable at >332 GPa

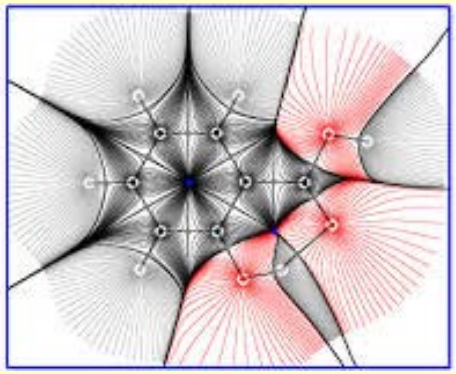
(Liu, Oganov, Kresse, *Sci. Rep.* 2015)



Structure of AlO_2

Predicting Unusual Compounds

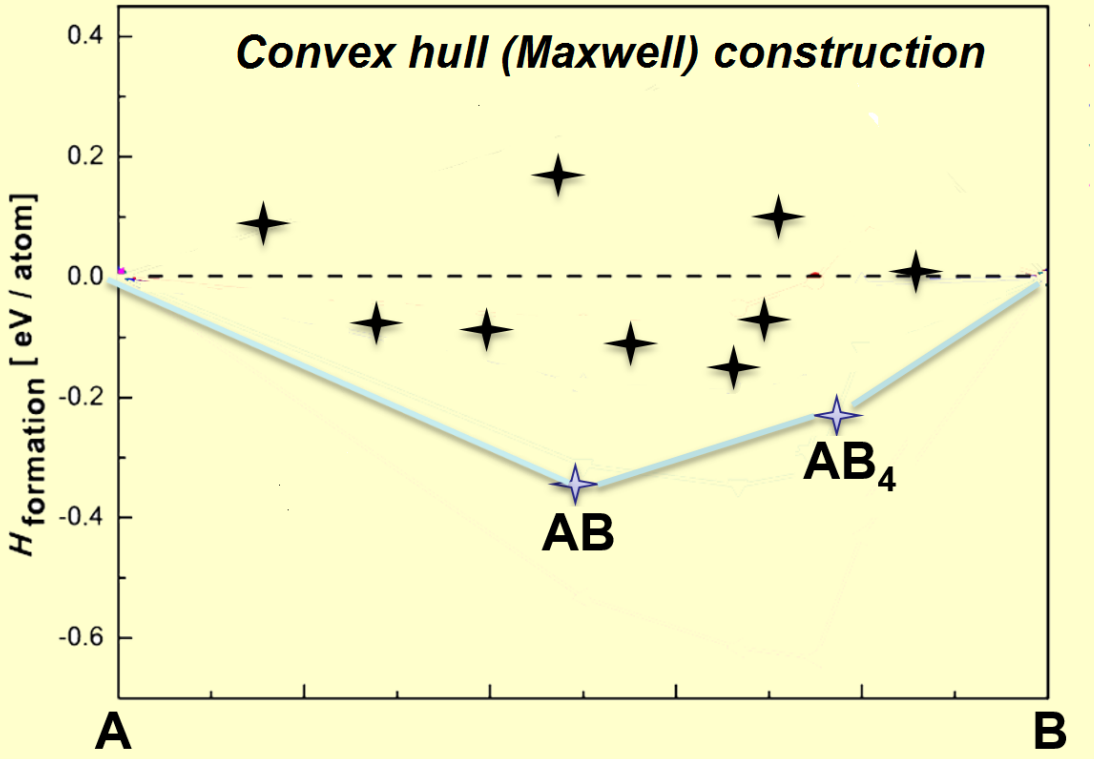
-"Forbidden" stoichiometries become stable: Na_3Cl, etc.
-Unusual valence/oxidation states appear: Cl^{2-}, Cs^{5+}, etc.
-He becomes chemically active.
-Unusual properties: e.g., room-temperature superconductivity.

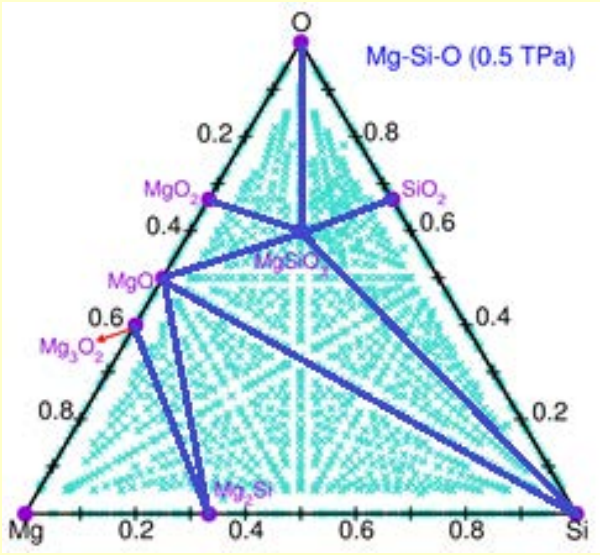


Bader analysis:
-Reasonable atomic charges (close to Pauling)

USPEX can automatically find all stable compounds in a multicomponent system.

Thermodynamic stability in variable-composition systems





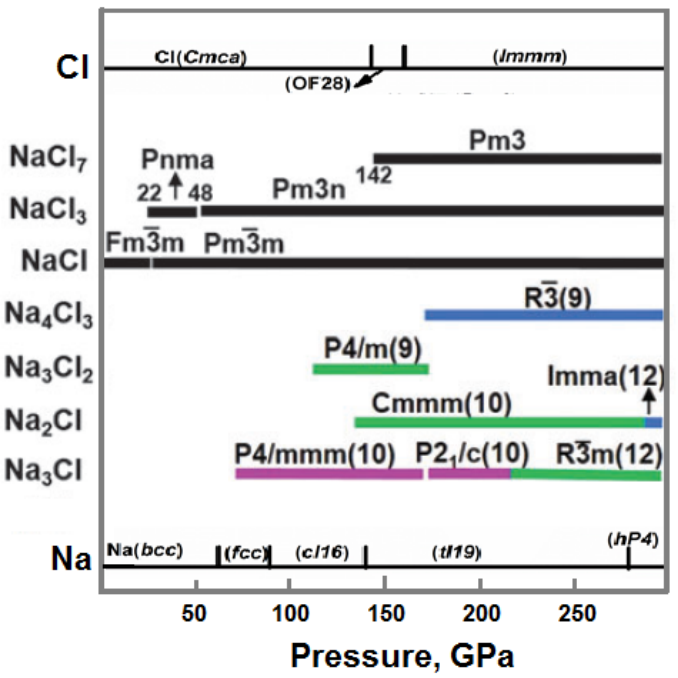
3-component convex hull: Mg-Si-O system at 500 GPa (Niu & Oganov, *Sci. Rep.* 2015)

Stable structure must be below all the possible decomposition lines !!

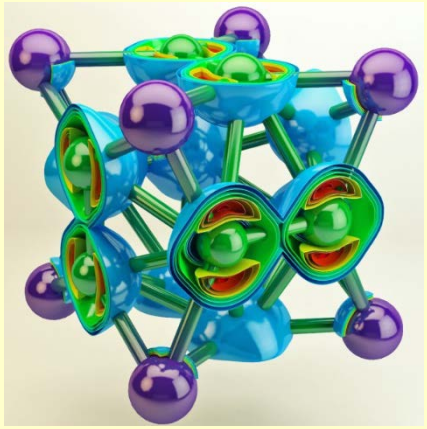
Unexpected chemistry of sodium chloride(s) :

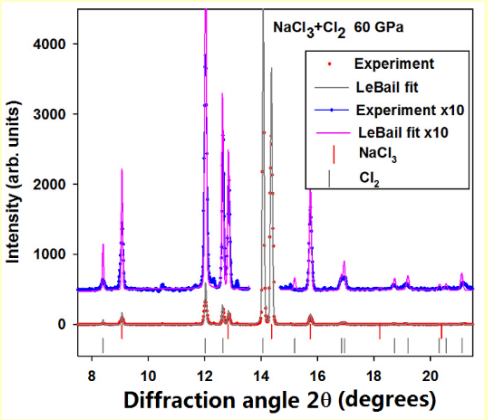
Na-Cl

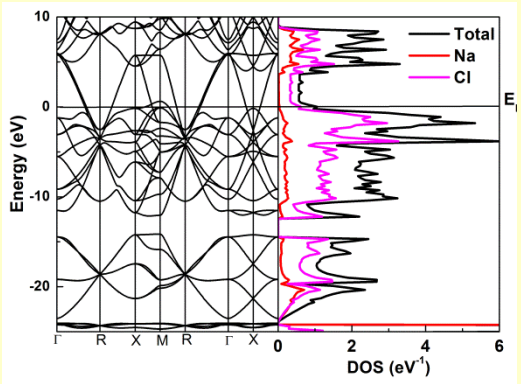
Na_3Cl, Na_2Cl, Na_3Cl_2, NaCl, $NaCl_3$, $NaCl_7$ are stable under pressure. Why? (Zhang, Oganov, Goncharov, *Science*, 2013).

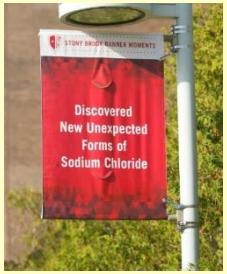


Stability fields of sodium chlorides









$NaCl_3$: atomic and electronic structure, and experimental XRD pattern

Chemical anomalies:

-Divalent Cl in Na_2Cl!

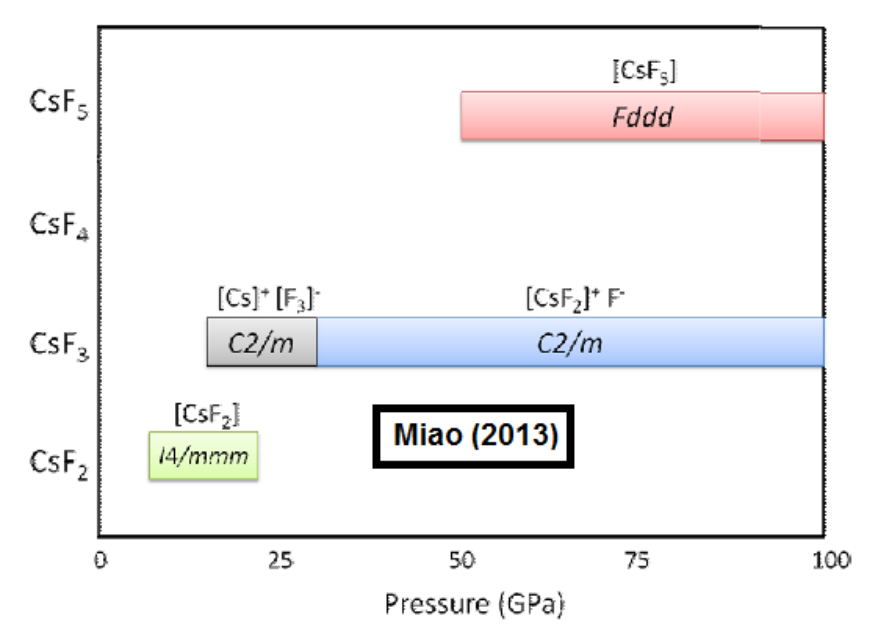
-Coexistence of metallic and ionic blocks in Na_3Cl!

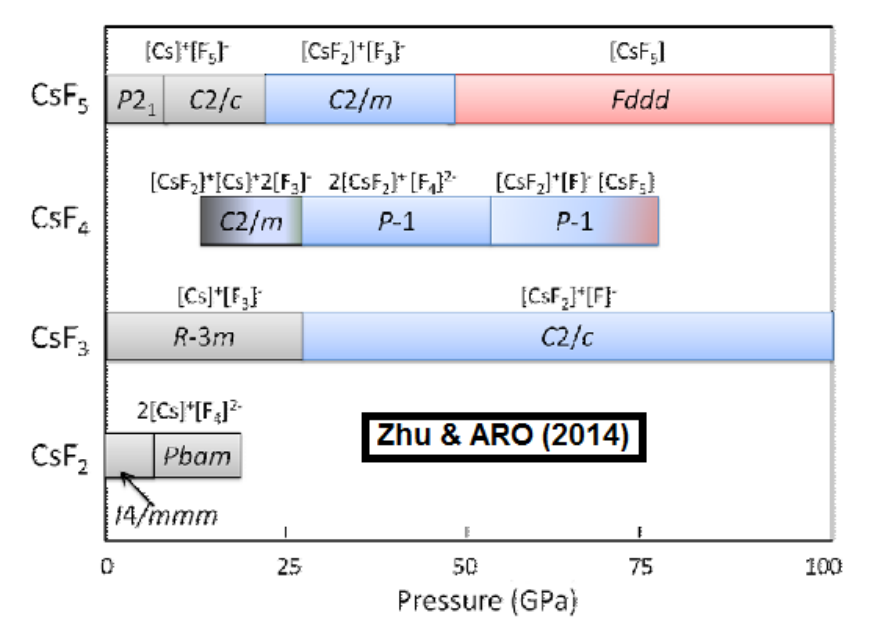
-Positively charged Cl in $NaCl_7$!

[Zhang, Oganov, et al., *Science* (2013)]
[Saleh & Oganov, *PCCP* (2015)]

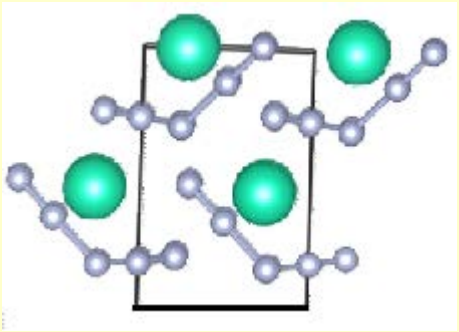
“Forbidden” compounds can be practically useful

Cs-F





Phase diagram of the Cs-F - Miao (2013) and corrected (Zhu & ARO, 2014)



Structure of CsF_5, at 1 atm

- **Miao (2013): predicted (incorrectly) novel compounds using CALYPSO. Zhu & Oganov (2015) – new phase diagram using USPEX.**
- **CsF_2, CsF_3, CsF_5 are stable at 1 atm and can be used for fluorine storage. Decomposition temperatures ~250-400 K. US patent (2013).**
- **At >40 GPa – Cs(V) in *Fddd-CsF_5*.**

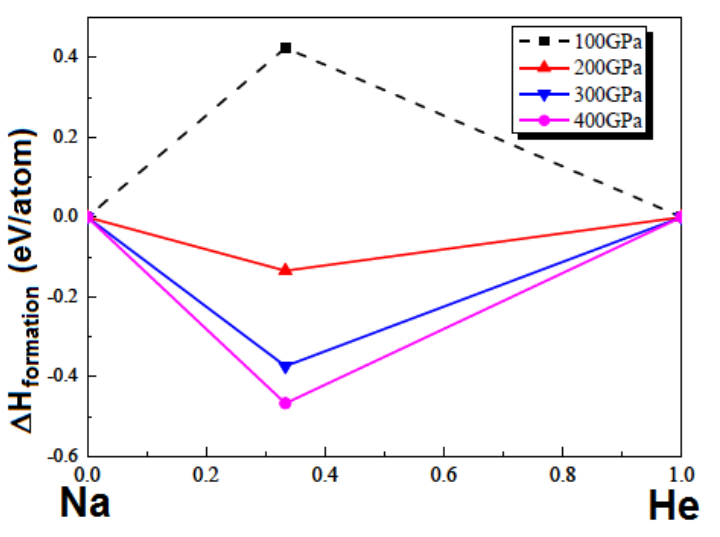
[Zhu & Oganov, *Sci. Rep.* (2015)]

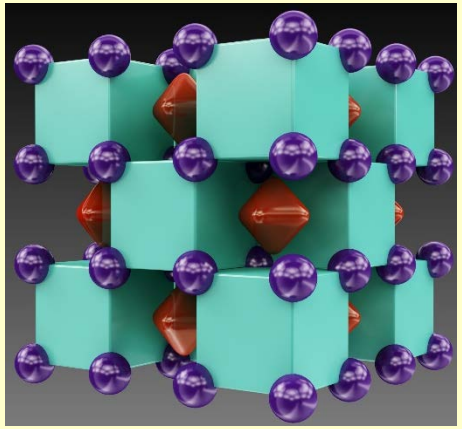
Helium chemistry? Yes!

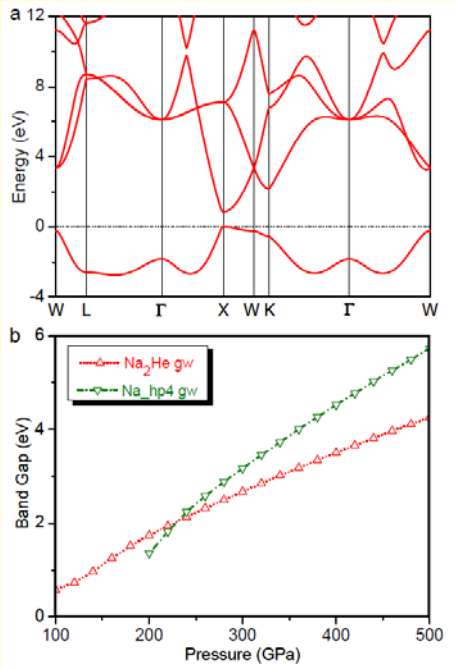
(Dong, Oganov, Goncharov, *Nature Chemistry* 2017)

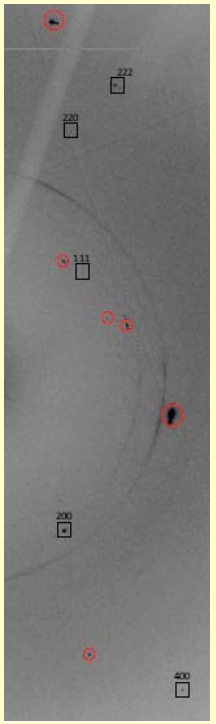
Na-He

- Helium is the 2nd most abundant element in the Universe (24 wt.%).
- Helium: ionization potential = 24.39 eV (record!)
  electron affinity = 0.08 eV
- No stable compounds are known at normal conditions. Under pressure: van der Waals compound $NeHe_2$ (Loubeyre et al., 1993).





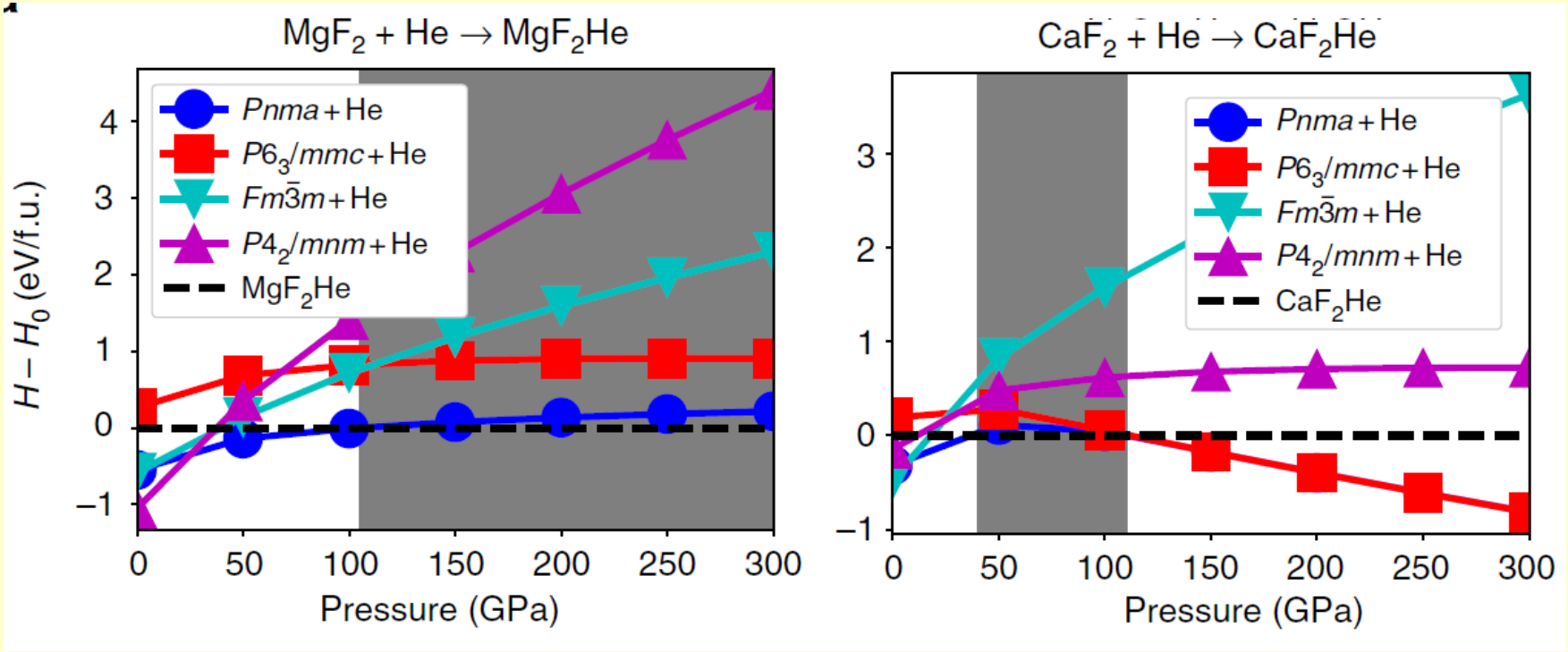




1. Na_2He is stable at >120 GPa, at least up to 1000 GPa.
2. Stabilized by an acceptor of an electron pair on the "2e" site. Na_2HeO – stable already at 14 GPa.

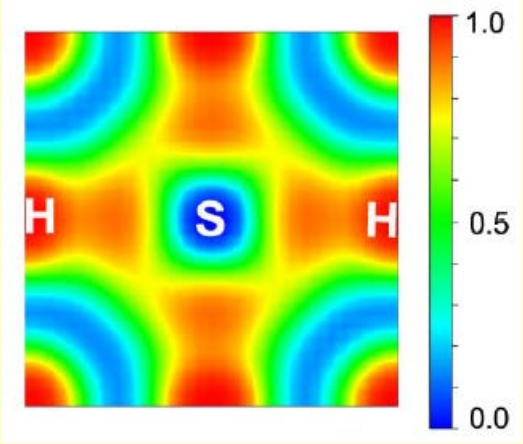
Noble gases are not inert under pressure (Liu et al., *Nature Comm.* 2018)

- **Helium reacts with Na, Na_2O, H_2O, SiO_2, MgF_2, CaF_2, …**
- **Noble gases can be retained in Earth's mantle (and core?).**



Recent record of high-Tc superconductivity: 203 Kelvin (Duan et al., *Sci. Rep.* 4, 6968 (2014))

H-S



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SUBJECT AREAS:
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Correspondence and requests for materials should be addressed to T.C. (cuitian@jlu.edu.cn)

Pressure-induced metallization of dense $(H_2S)_2H_2$ with high-T_c superconductivity

Defang Duan[1,2], Yunxian Liu[1], Fubo Tian[1], Da Li[1], Xiaoli Huang[1], Zhonglong Zhao[1], Hongyu Yu[1], Bingbing Liu[1], Wenjing Tian[2] & Tian Cui[1]

[1]State Key Laboratory of Superhard Materials, College of physics, Jilin University, Changchun, 130012, P. R. China, [2]State Key Laboratory of Supramolecular Structure and Materials, Jilin University, Changchun, 130012, P. R. China.

The high pressure structures, metallization, and superconductivity of recently synthesized H_2-containing compounds $(H_2S)_2H_2$ are elucidated by *ab initio* calculations. The ordered crystal structure with *P*1 symmetry is determined, supported by the good agreement between theoretical and experimental X-ray diffraction data, equation of states, and Raman spectra. The *Cccm* structure is favorable with partial hydrogen bond symmetrization above 37 GPa. Upon further compression, H_2 molecules disappear and two intriguing metallic structures with *R*3*m* and *Im*-3*m* symmetries are reconstructive above 111 and 180 GPa, respectively. The predicted metallization pressure is 111 GPa, which is approximately one-third of the currently suggested metallization pressure of bulk molecular hydrogen. Application of the Allen-Dynes-modified McMillan equation for the *Im*-3*m* structure yields high T_c values of 191 K to 204 K at 200 GPa, which is among the highest values reported for H_2-rich van der Waals compounds and MH_3 type hydride thus far.

Conventional superconductivity at 203 kelvin at high pressures in the sulfur hydride system

A. P. Drozdov, M. I. Eremets, I. A. Troyan, V. Ksenofontov & S. I. Shylin

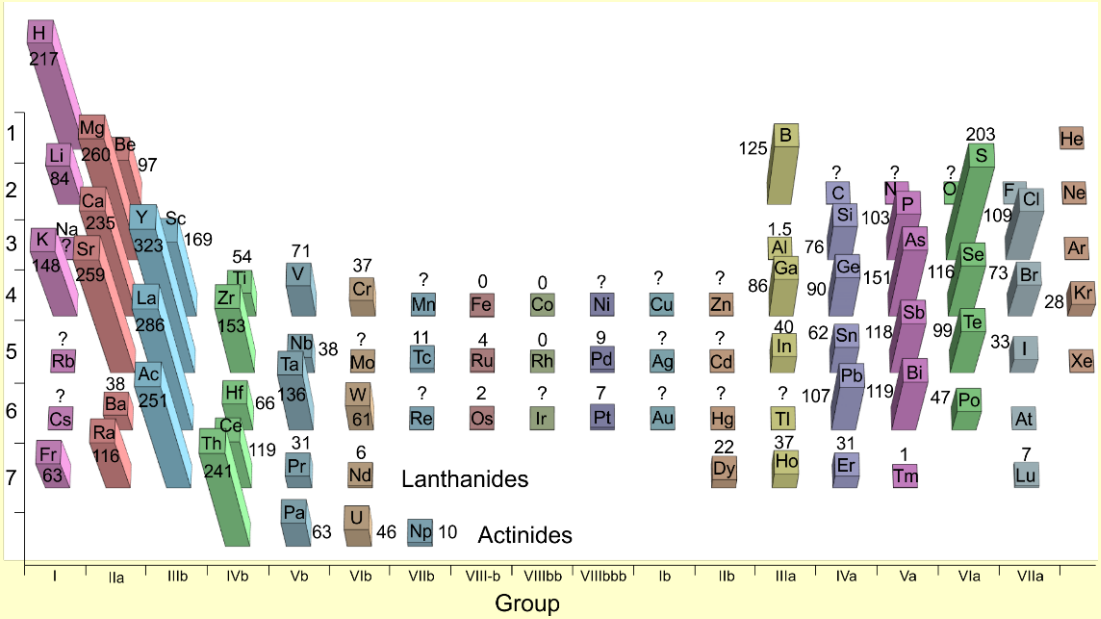
Nature (2015) | doi:10.1038/nature14964
Received 25 June 2015 | Accepted 22 July 2015 | Published online 17 August 2015

A superconductor is a material that can conduct electricity without resistance below a superconducting transition temperature, T_c. The highest T_c that has been achieved to date is in the copper oxide system[1]: 133 kelvin at ambient pressure[2] and 164 kelvin at high pressures[3]. As the nature of superconductivity in these materials is still not fully understood (they are not conventional superconductors), the prospects for achieving still higher transition temperatures by this route are not clear. In contrast, the Bardeen–Cooper–Schrieffer theory of conventional superconductivity gives a guide for achieving high T_c with no theoretical upper bound—all that is needed is a favourable combination of high-frequency phonons, strong electron–phonon coupling, and a high density of states[4]. These conditions can in principle be fulfilled for metallic hydrogen and covalent compounds dominated by hydrogen[5, 6], as hydrogen atoms provide the necessary high-frequency phonon modes as well as the strong electron–phonon coupling. Numerous calculations support this idea and have predicted transition temperatures in the range 50–235 kelvin for many hydrides[7], but only a moderate T_c of 17 kelvin has been observed experimentally[8]. Here we investigate sulfur hydride[9], where a T_c of 80 kelvin has been predicted[10]. We find that this system transforms to a metal at a pressure of approximately 90 gigapascals. On cooling, we see signatures of superconductivity: a sharp drop of the resistivity to zero and a decrease of the transition temperature with magnetic field, with magnetic susceptibility measurements confirming a T_c of 203 kelvin. Moreover, a pronounced isotope shift of T_c in sulfur deuteride is suggestive of an electron–phonon mechanism of superconductivity that is consistent with the Bardeen–Cooper–Schrieffer scenario. We argue that the phase responsible for high-T_c superconductivity in this system is likely to be H_3S, formed from H_2S by decomposition under pressure. These findings raise hope for the prospects for achieving room-temperature superconductivity in other hydrogen-based materials.

- **Old record Tc=135 K (Schilling, 1993) is broken: theorists (T. Cui, 2014) predicted new compound H_3S with Tc~200 K.**
- **Confirmed by A. Drozdov et al. (*Nature* 525, 73 (2015)).**

Superconductivity is linked with Mendeleev's Table

[Semenok & Oganov, JPCL, 2018]

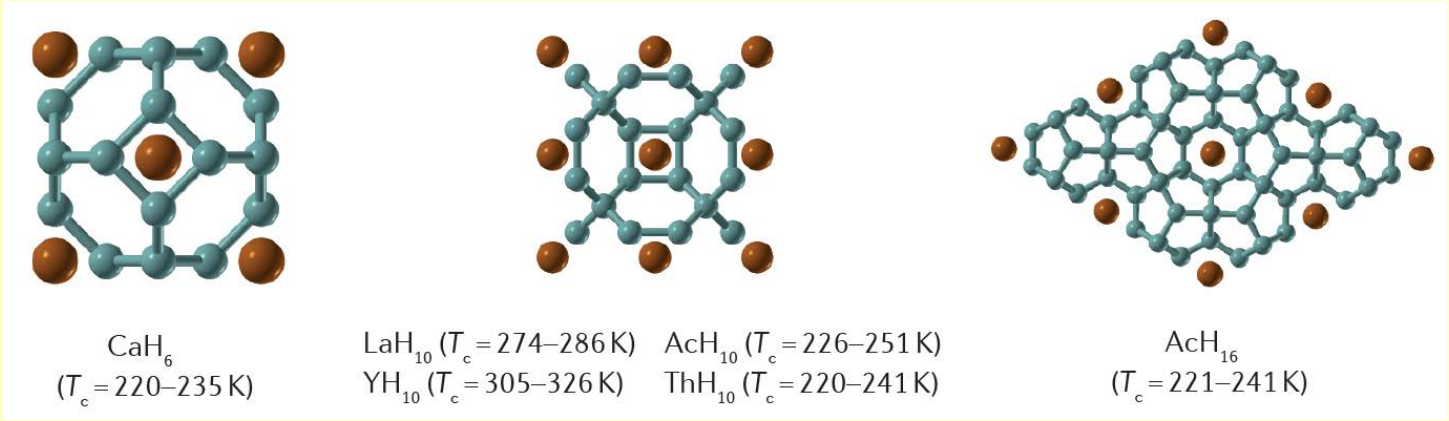


LaH_{10}: record Tc (260 K @ 190 GPa) (Somayazulu et al., 2019).

Test of idea: Th and Ac hydrides have high-Tc superconductivity.

ThH_{10}: Tc=241 K at 100 GPa (Kvashnin & Oganov, 2018).

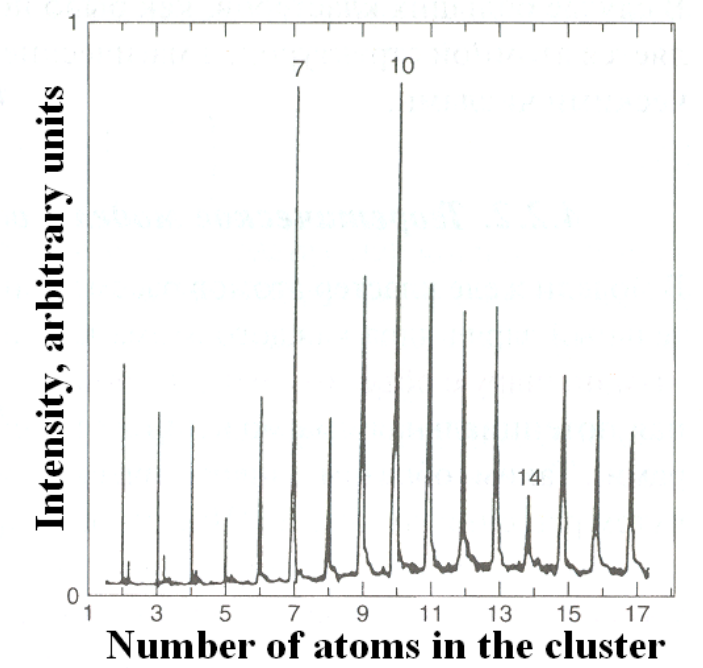
Distribution of Tc for hydrides



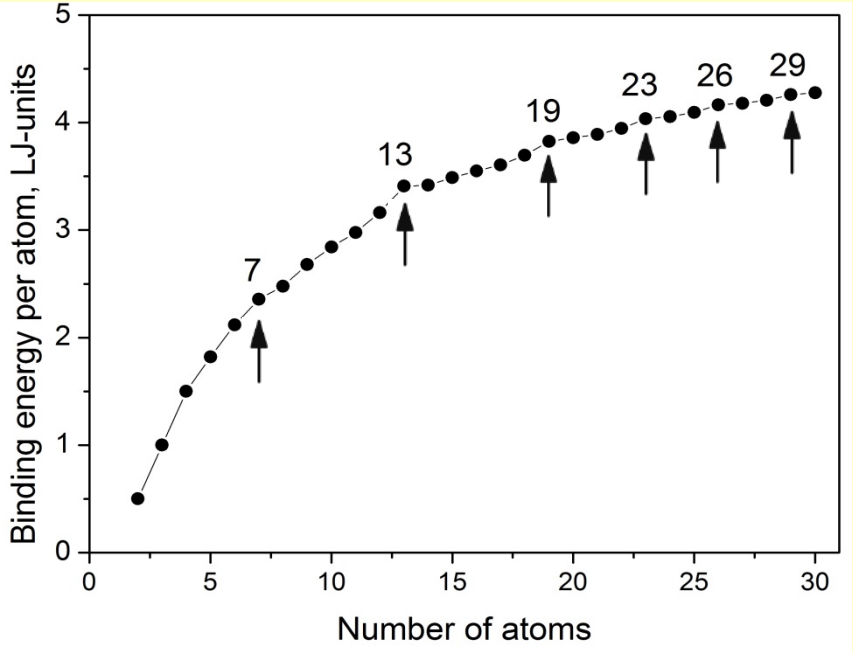
Predicting Stable Nanoclusters

-Only conditional stability. Magic clusters. Similar to atomic nuclei.
-Unusual stable compositions are typical.
-Explanation of carcinogenicity of oxide dust?

Stability of nanoparticles: conditional

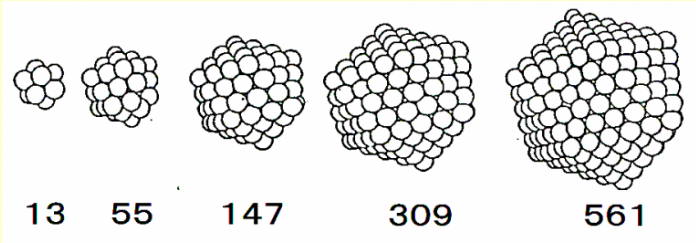


Mass spectrum of Pb_n clusters (Poole & Owens, 2003)



Lennard-Jones clusters

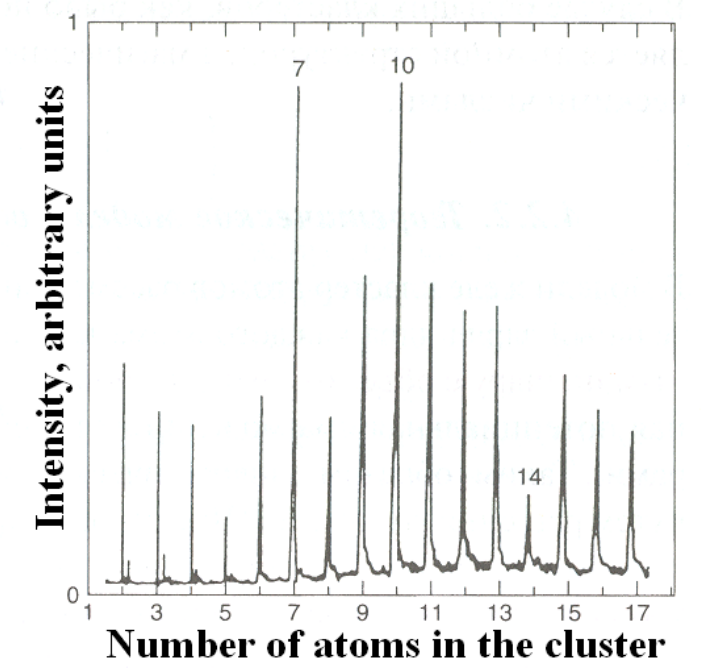
↑ – “magic” clusters



Stability grows with cluster size. We define stability relative to neighboring compositions. Especially stable clusters have filled electronic and/or structural shells.

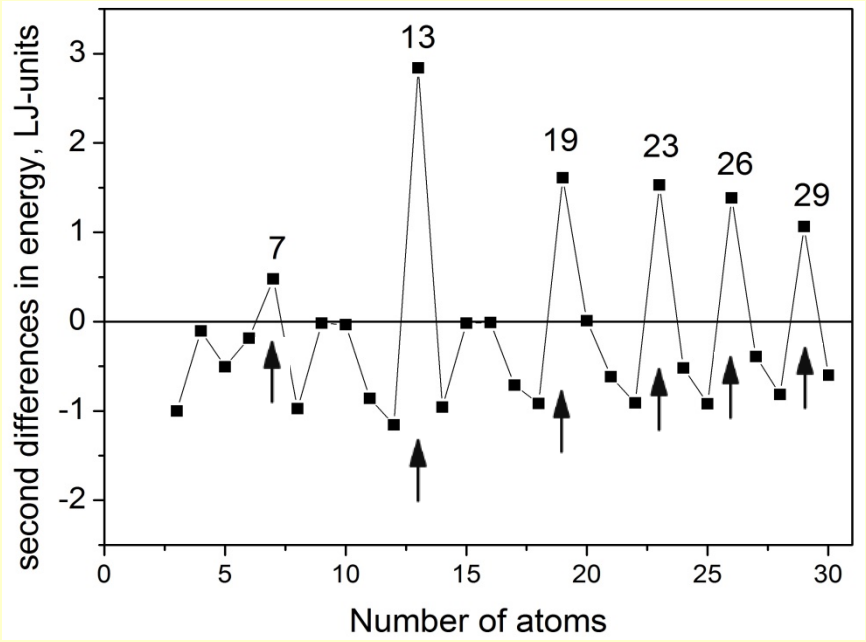
Stability of clusters

Real system: Pb clusters



Mass-spectrum of Pb_n clusters (from Poole & Owens, 2003)

Model system: Lennard-Jones clusters



– magic clusters.

Criterion of local stability (magic clusters):

$$\Delta^2 \mathrm{E} = \mathrm{E}(n+1) + \mathrm{E}(n-1) - 2\mathrm{E}(n) > 0$$

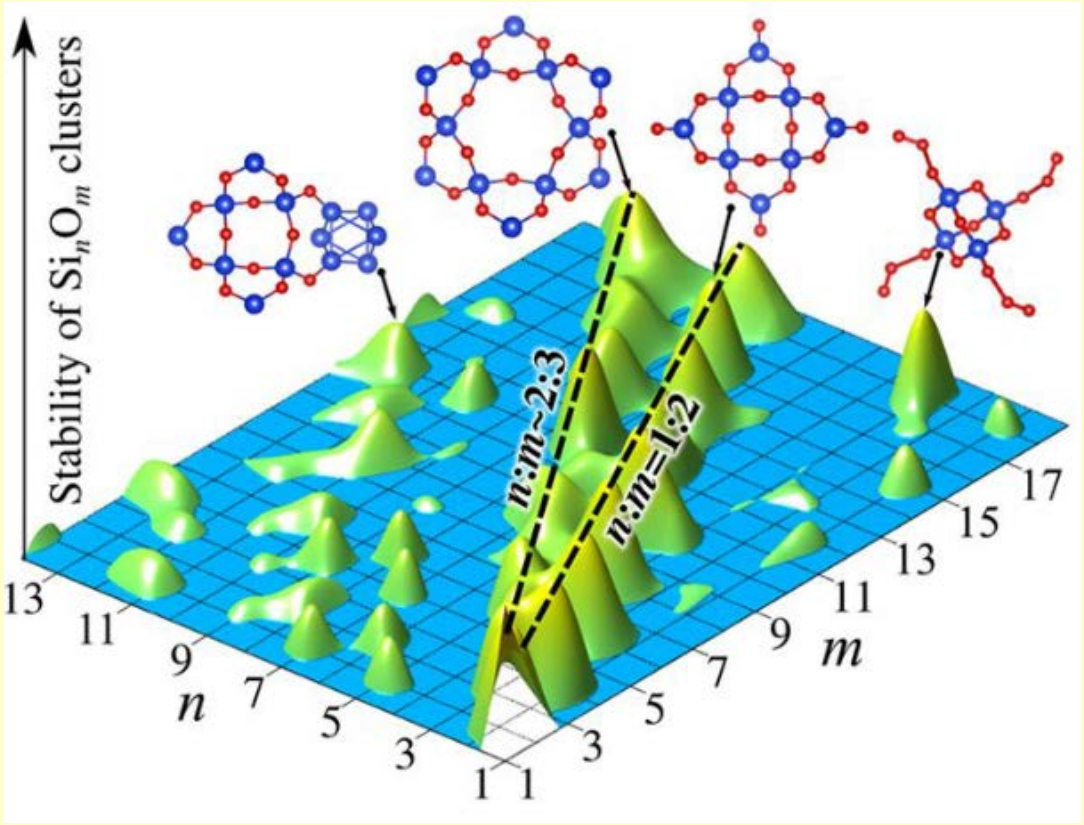
For binary clusters (A_mB_n):

$$\Delta_x^2 \mathrm{E} = \mathrm{E}(m,n+1) + \mathrm{E}(m,n-1) - 2\mathrm{E}(m,n) > 0$$

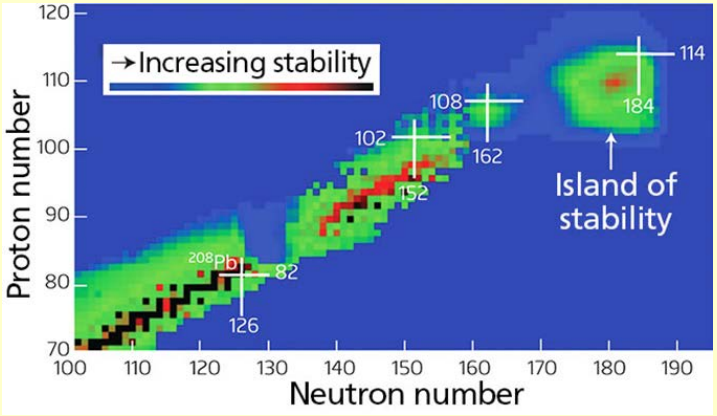
$$\Delta_y^2 \mathrm{E} = \mathrm{E}(m+1,n) + \mathrm{E}(m-1,n) - 2\mathrm{E}(m,n) > 0$$

Map of stability of Si-O clusters
[Lepeshkin & Oganov, *J. Phys. Chem. Lett.* 2019]

Si-O



Ridges of stability: SiO_2, Si_2O_3
Islands of stability: e.g., Si_4O_{18}

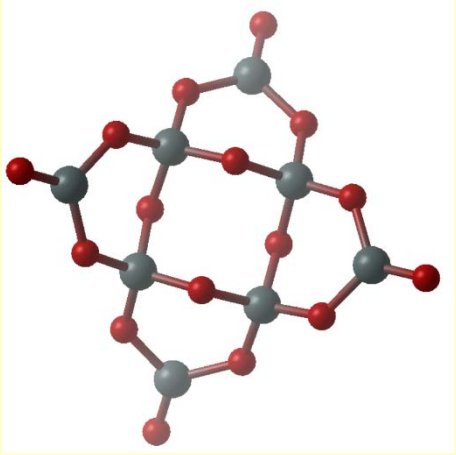


«Magic» nuclei: with filled proton or neutron shells (2, 8, 20, 28, 50, 82, 126 p or n)
($1s^2/2p^6/3d^{10}2s^2/4f^8/4f^63p^65g^{10}/5g^84d^{10}3s^26h^{12}$)

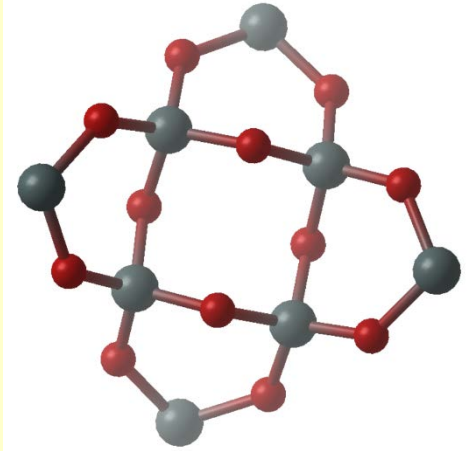
Magic numbers of electrons = 2, 10, 18, 36, 54, 86, 118)

Analogy with magic atomic nuclei

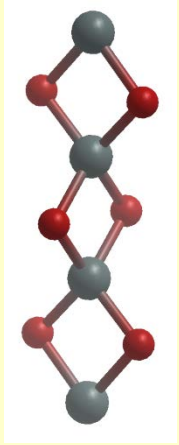
Si-O



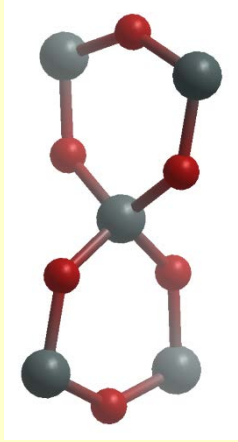
Si_8O_{16}



Si_8O_{12}

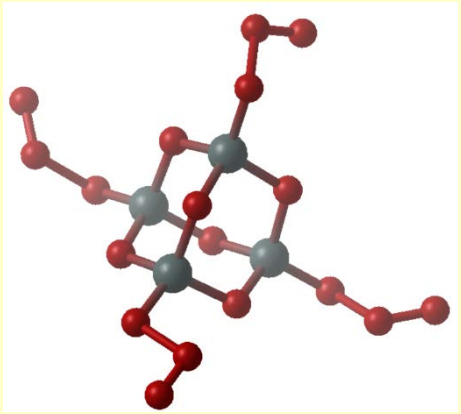


Si_4O_6

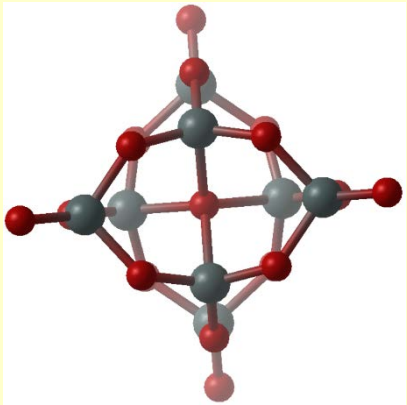


Si_5O_6

Magic clusters. Non-magnetic

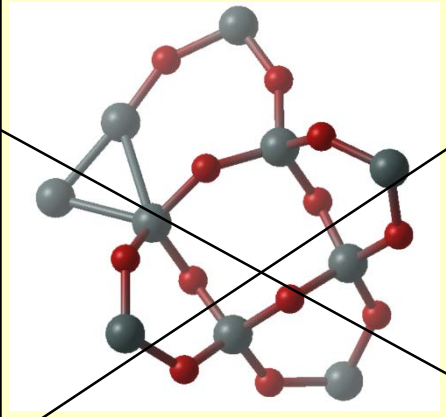


Si_4O_{18}



Si_8O_{17}

Magic magnetic(!) clusters. Excess of O

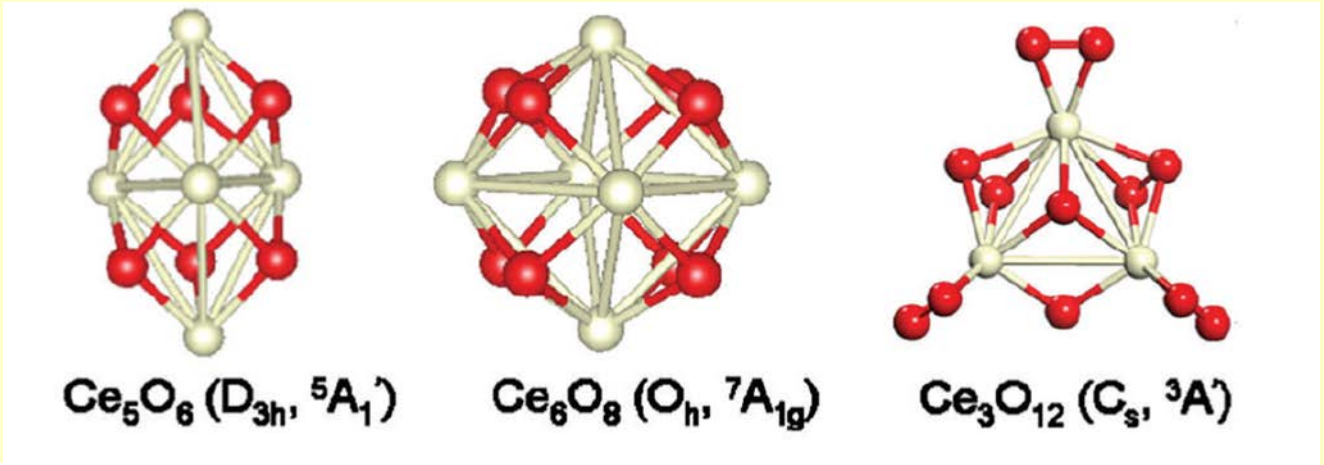


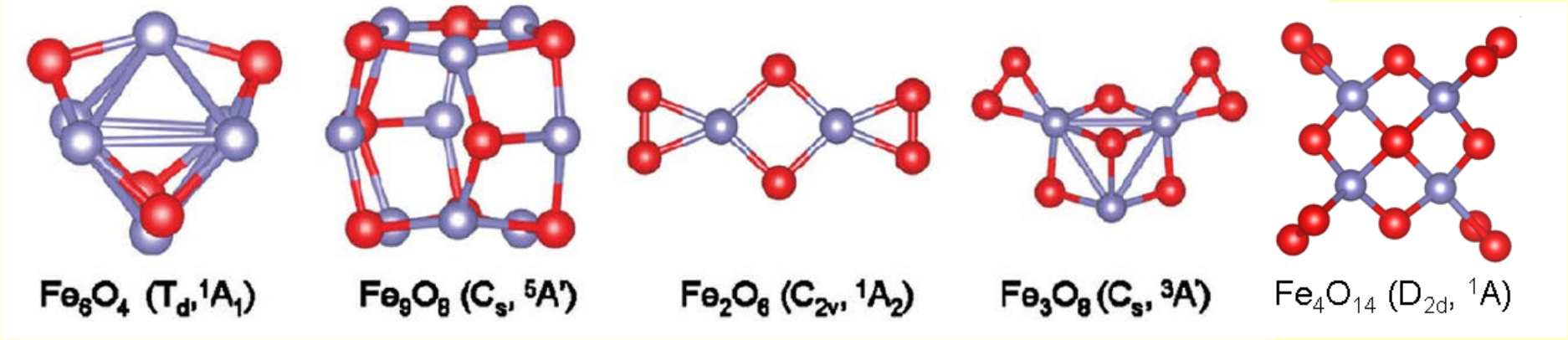
$Si_{10}O_{12}$

Unstable

Unusual compositions of transition metal oxide clusters

[Yu & Oganov, *Phys. Chem. Chem. Phys.*, 2018]





Do crystals grow from such particles?

Predicting Optimal Materials

-Superior thermoelectrics: possible!
-New superhard materials: WB_5 etc.

-Pareto optimization of properties & stability.
-Mendelevian search for exploring chemical space.

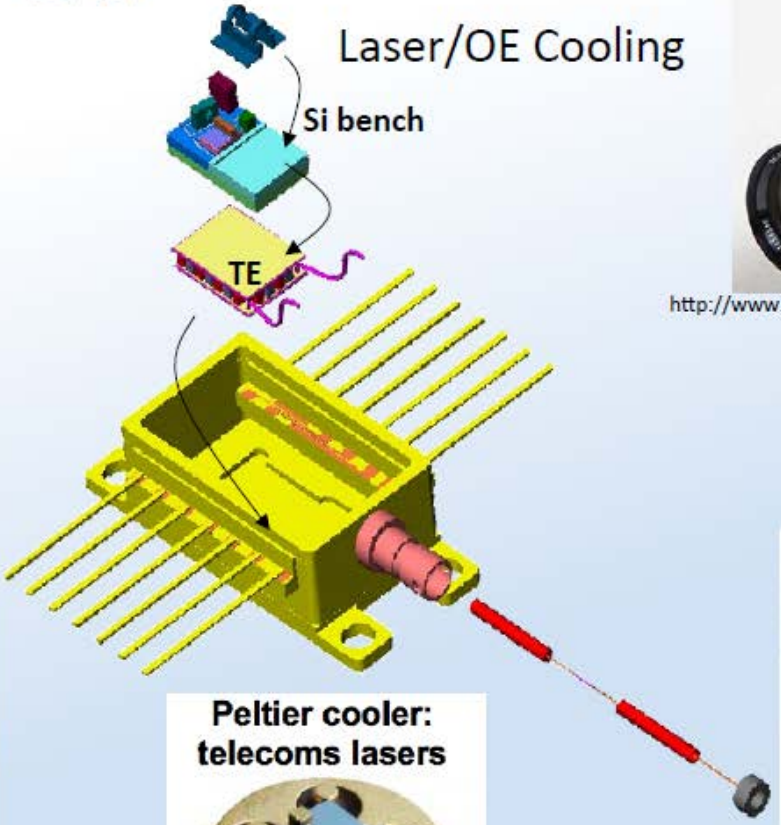
Towards materials design: example of thermoelectrics

Applications

Water/Beer/Wine Cooler

www.kingsbottle.com

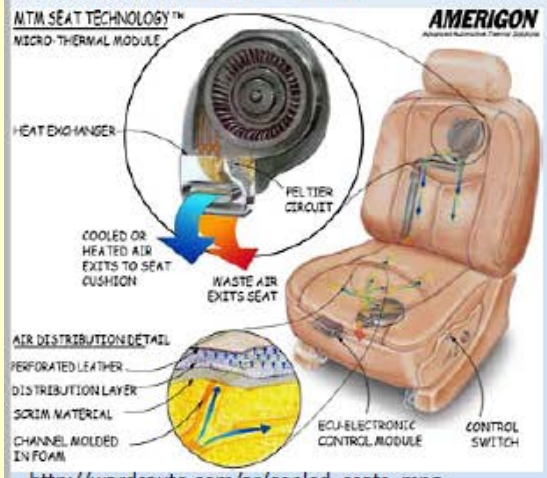
Laser/OE Cooling



Cryogenic IR Night Vision

http://www.x20.org/products/pv400-used-night-vision-scope/

Cooled Car Seat

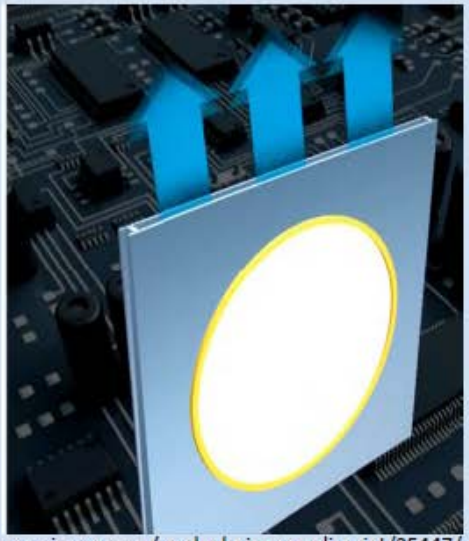


http://wardsauto.com/ar/cooled_seats_mpg

Peltier cooler: telecoms lasers

Copyright Micropelt

Electronic Cooling

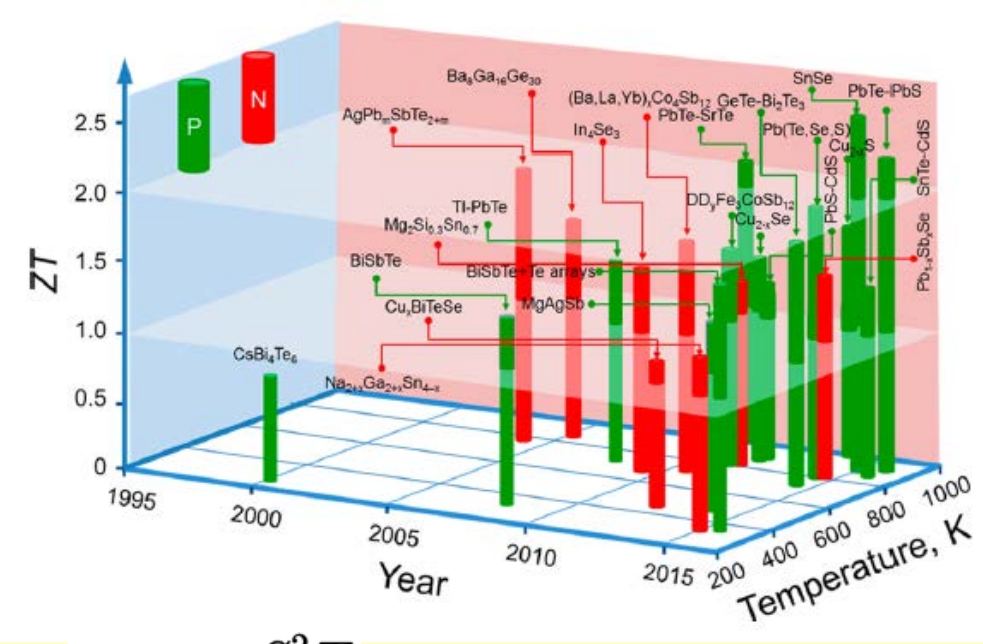


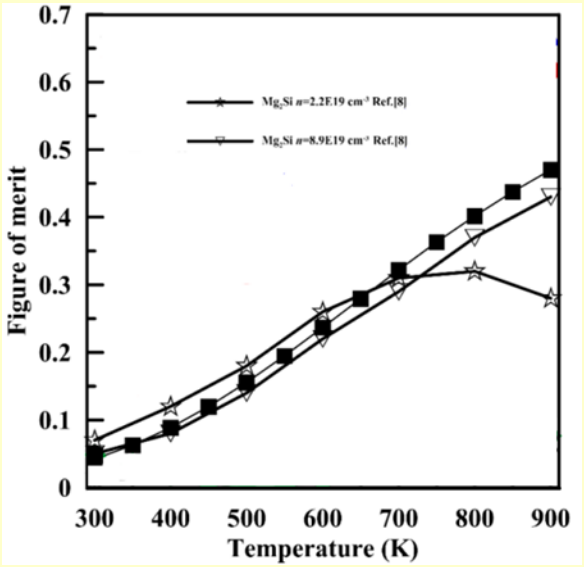
http://www.gizmag.com/ge-dual-piezo-cooling-jet/25447/

How to improve efficiency of thermoelectric devices?

"One shouldn't work on semiconductors, that is a filthy mess; who knows whether any semiconductors exist"

-W. Pauli, letter to R. Peierls (1931)





[Fan & Oganov (2018)]

$$ZT = \frac{\sigma S^2 T}{\kappa}$$

$$S = \frac{8\pi^2 k_B^2}{3eh^2} m^* T \left(\frac{\pi}{3n}\right)^{2/3}$$

$$\eta = \frac{\Delta T}{T_H} \frac{\sqrt{1+zT}-1}{\sqrt{1+zT}+\frac{T_C}{T_H}}$$

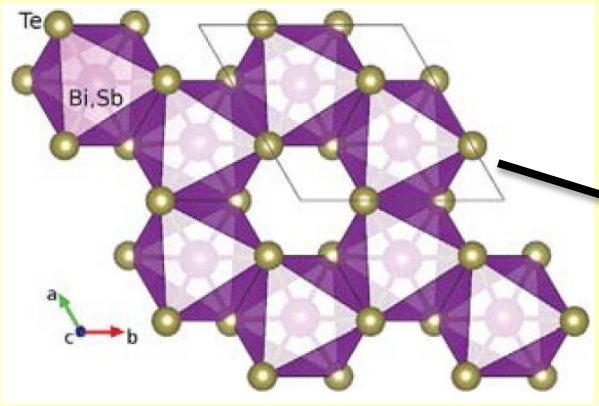
- efficiency

Multiobjective (Pareto) optimization finds a new thermoelectric polymorph of Bi_2Te_3

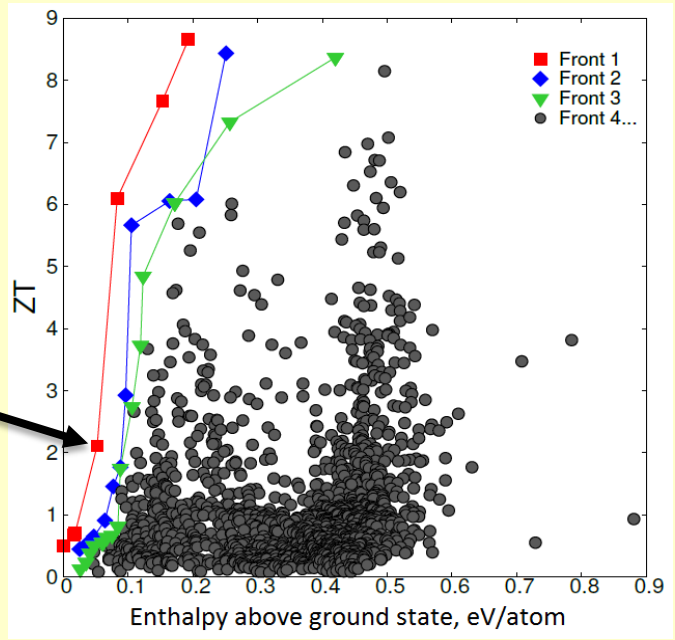
Computer Physics Communications 222 (2018) 152–157

Efficient technique for computational design of thermoelectric materials

Maribel Núñez-Valdez , Zahed Allahyari , Tao Fan , Artem R. Oganov



Predicted *P*6_3*cm* structure of Bi_2Te_3



Pareto optimization of ZT and stability in the Bi-Te system

Similar conclusions from data mining

Journal of
Materials Chemistry C

PAPER

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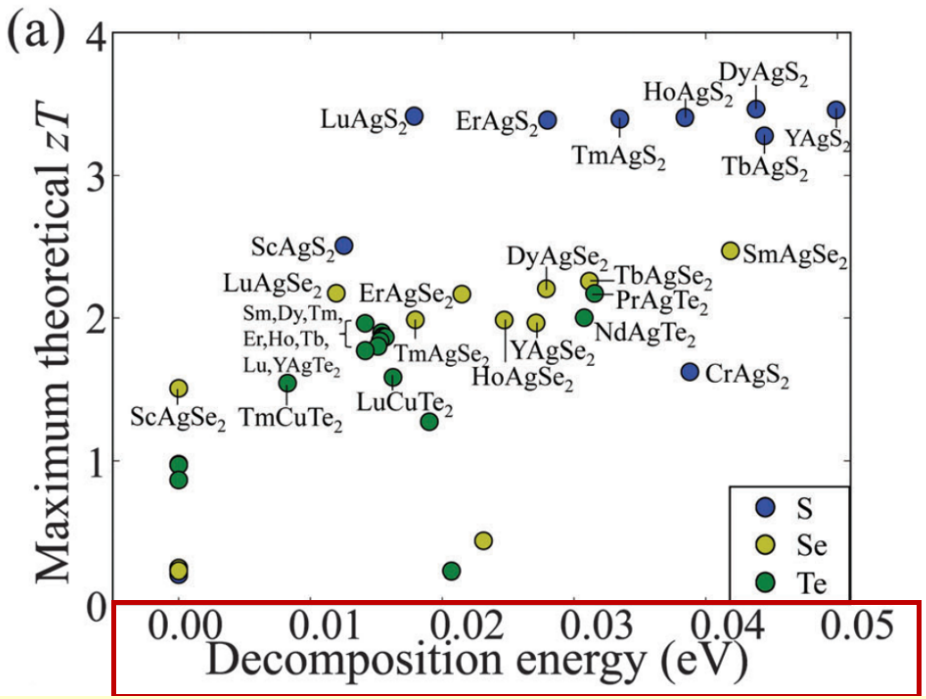
View Journal

Computational and experimental investigation of $TmAgTe_2$ and XYZ_2 compounds, a new group of thermoelectric materials identified by first-principles high-throughput screening†

Cite this: DOI: 10.1039/c5tc01440a

Hong Zhu,‡[a] Geoffroy Hautier,[b] Umut Aydemir,[c] Zachary M. Gibbs,[d] Guodong Li,[c] Saurabh Bajaj,[c] Jan-Hendrik Pöhls,[e] Danny Broberg,[f] Wei Chen,[g] Anubhav Jain,[f] Mary Anne White,[d] Mark Asta,[e] G. Jeffrey Snyder,[c] Kristin Persson[f] and Gerbrand Ceder*[afg]

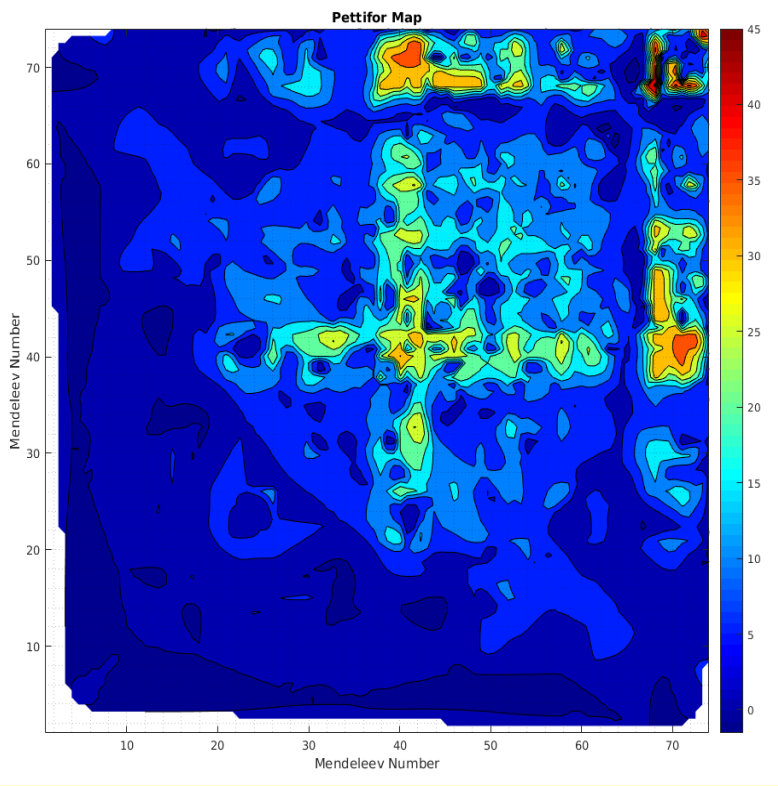


We can simultaneously optimize composition, structure, stability and other properties for a given chemical system.

Now, let’s predict the best material(s) among all possible chemical systems!

Mendelevian Search – breakthrough method for discovering best materials among all possible compounds

[Allahyari & Oganov, 2018]



- **118 elements**
- **7021 binary systems**
- **273937 ternaries**
- **In each system - ∞ possible structures**

Mendeleev Number – a way to arrange elements and compounds by properties

[Pettifor, 1984; Allahyari & Oganov, 2018]

Mendeleev Number	Atom	Mendeleev Number	Atom	Mendeleev Number	Atom
1	Fr	32	Tl	62	Po
2	Cs	33	U	63	Fe
3	Rb	34	Pa	64	Cu
4	K	35	Zr	65	Co
5	Ra	36	Pu	66	As
6	Ba	37	Np	67	Ni
7	Sm	38	Nb	68	Kr
8	Gd	39	Ta	69	Mo
9	Eu	40	In	70	I
10	Sr	41	Pb	71	Pd
11	Tm	42	Cd	72	Ir
12	Pm	43	Xe	73	Os
13	Ca	44	Ti	74	P
14	Na	45	Al	75	Ru
15	Ac	46	Bi	76	Pt
16	La	47	Sn	77	At
17	Yb	48	Hg	78	Rh
18	Tb	49	Zn	79	W
19	Y	50	Ga	80	Rn
20	Dy	51	V	81	Se
21	Ho	52	Mn	82	B
22	Ce	53	Sb	83	Au
23	Er	54	Te	84	S
24	Li	55	Cr	85	Br
25	Th	56	Ag	86	H
26	Lu	57	Be	87	C
27	Pr	58	Ge	88	Cl
28	Nd	59	Re	89	N
29	Mg	60	Si	90	O
30	Sc	61	Tc	91	F
31	Hf				

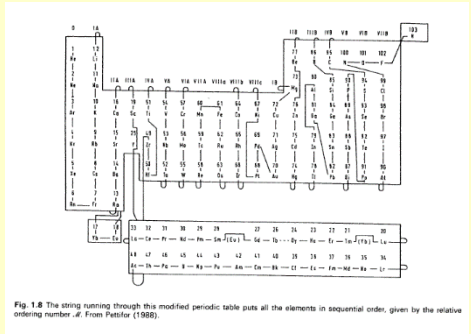
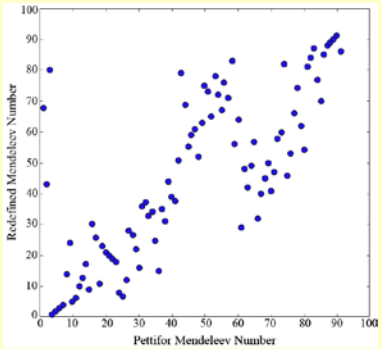
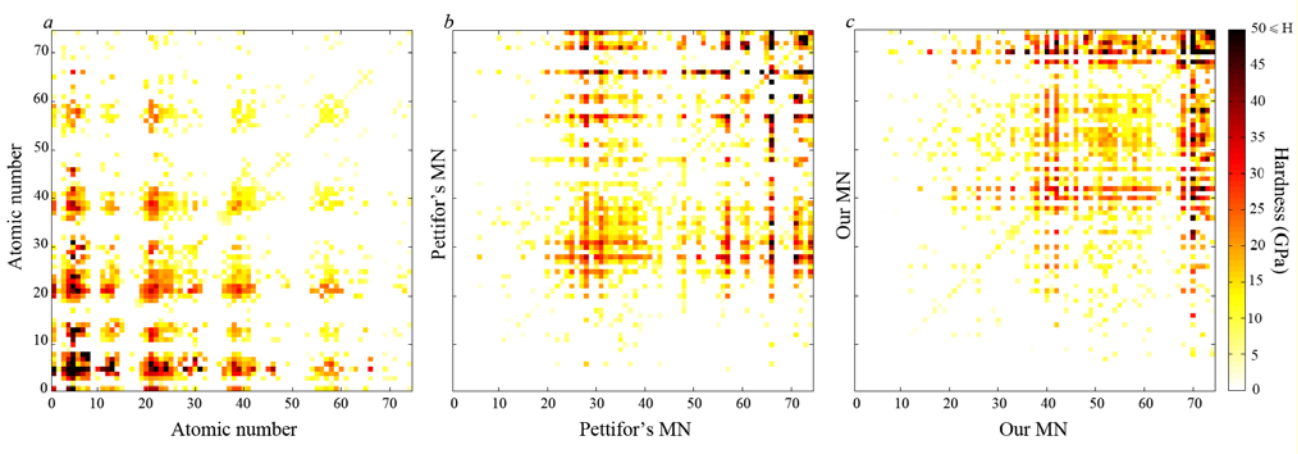


Fig. 1.8 The string running through this modified periodic table puts all the elements in sequential order, given by the relative ordering number, A'. From Pettifor (1988).

Pettifor's construction



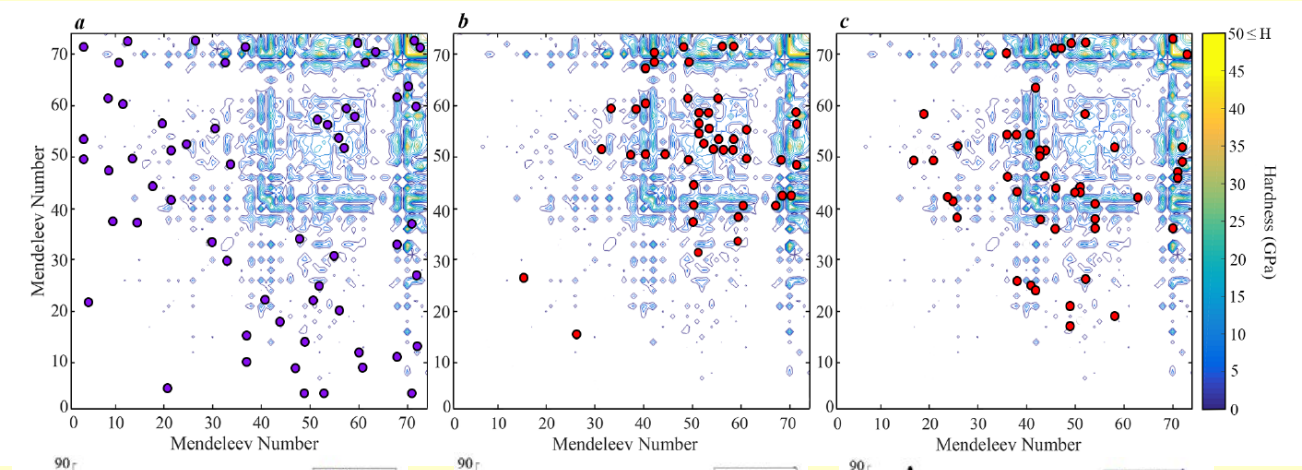
Comparison with Pettifor's numbers

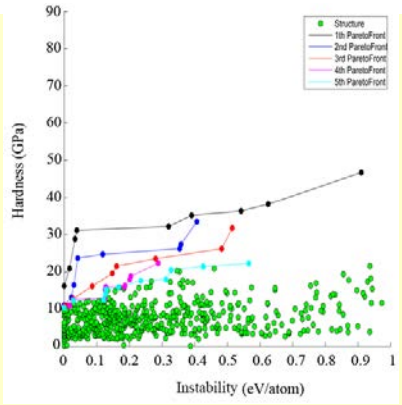


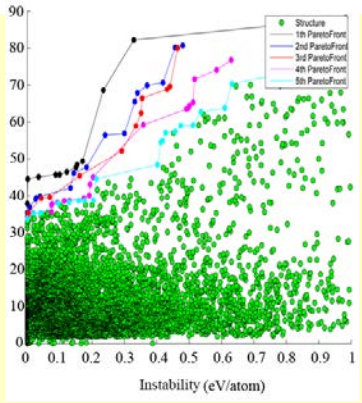
Grouping of hardness by (a) sequential number, (b) Pettifor's Mendeleev number, (c) our Mendeleev number

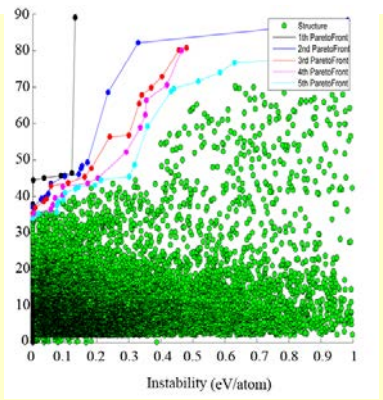
Mendelevian search for the hardest possible material: diamond and lonsdaleite are found!

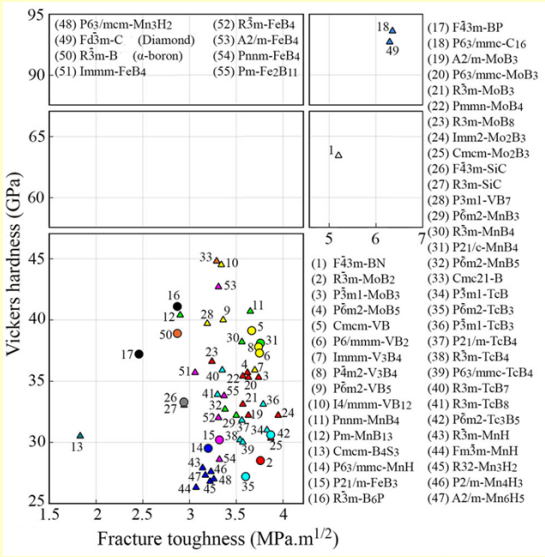
1st generation **5th generation** **10th generation**





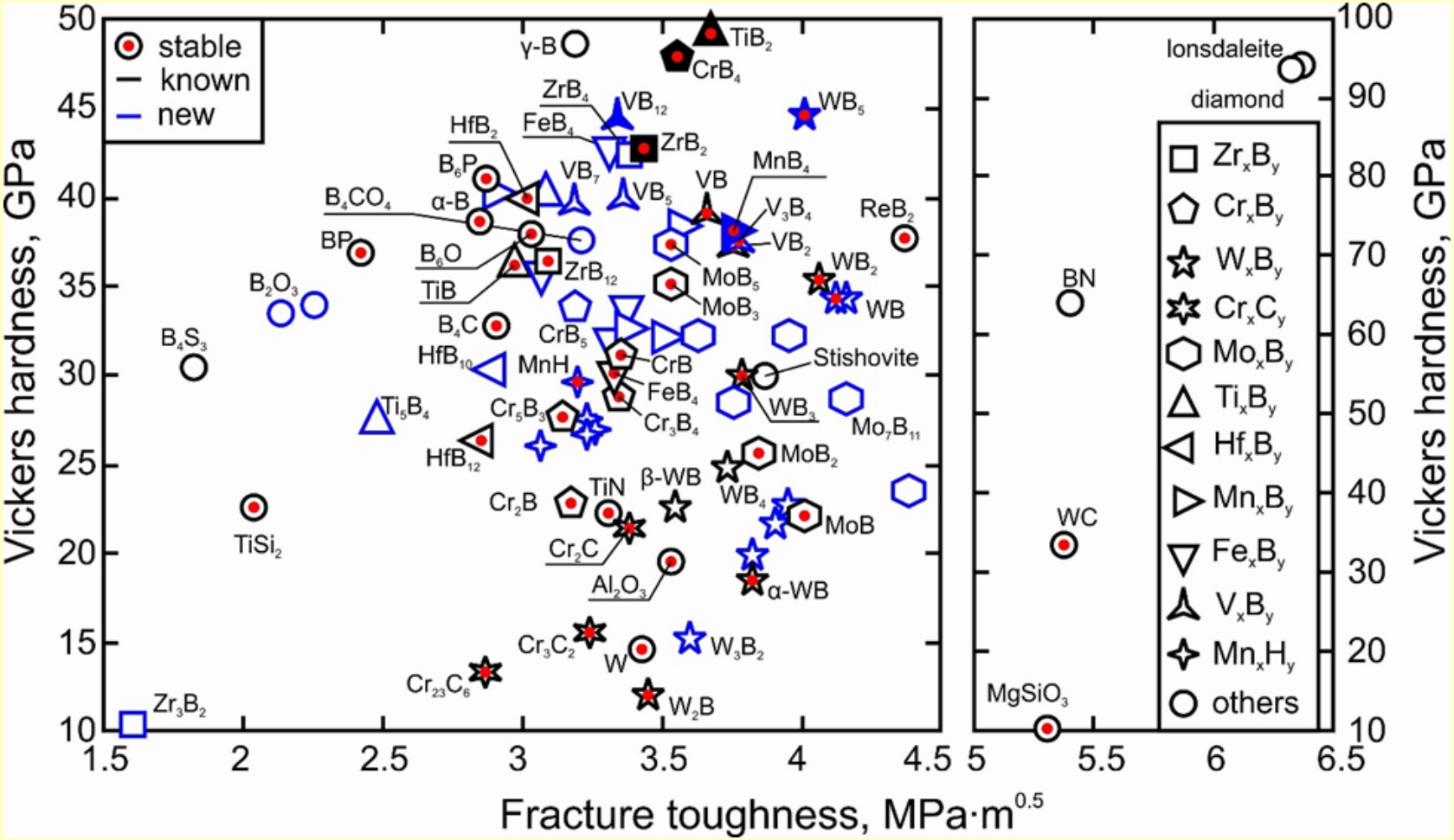






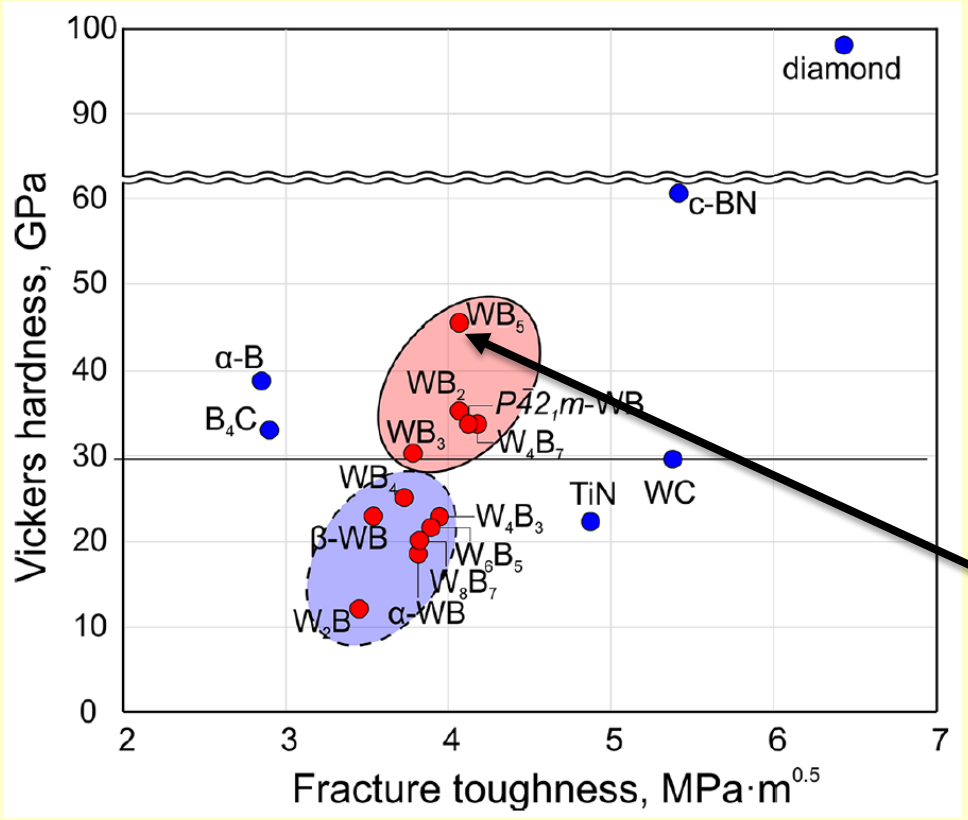
“Treasure map” of superhard materials

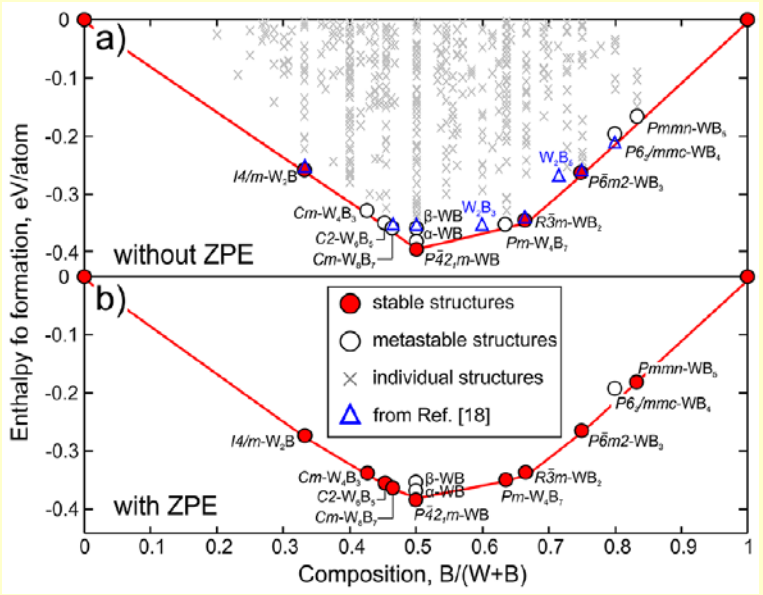
[Kvashnin, Allahyari, Oganov, 2019]

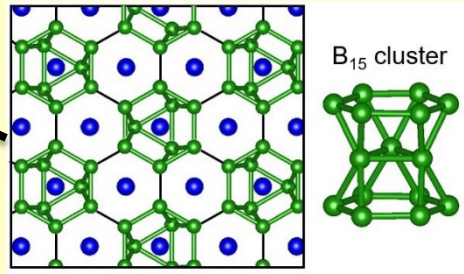


WB_5: new supermaterial

[Kvashnin & Oganov, J. Phys. Chem. Lett., 2018]





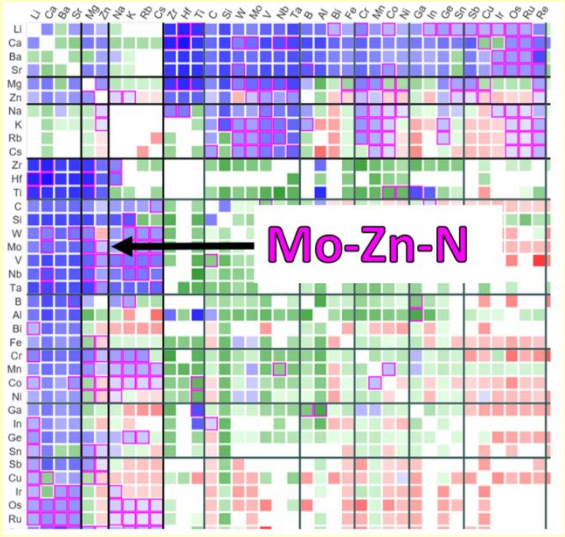


New material WB_5

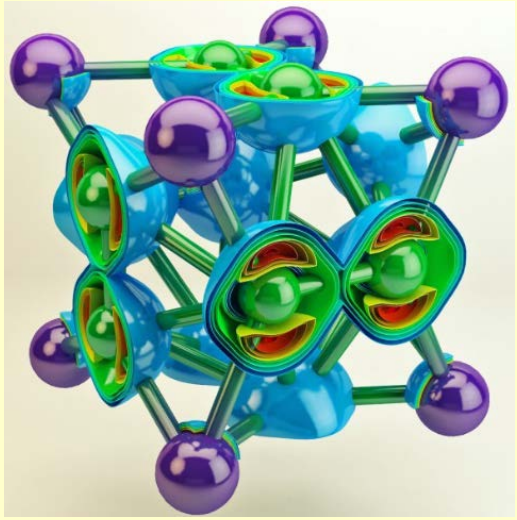
Tungsten carbide WC - standard

Synthesized by V. Filonenko

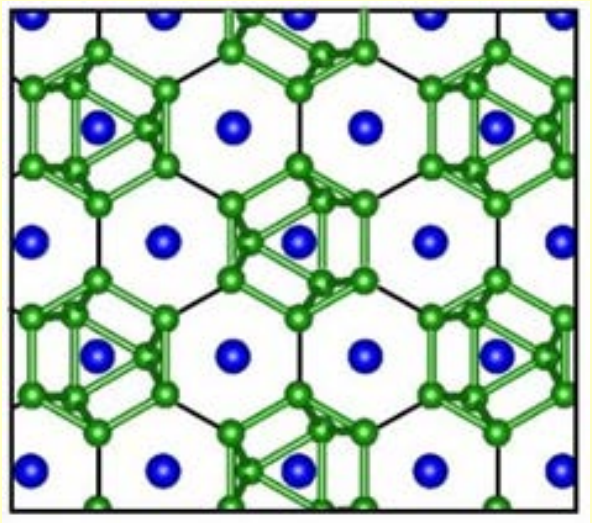
Advanced algorithms predict new supermaterials and help us understand nature



Power and limitations of machine learning



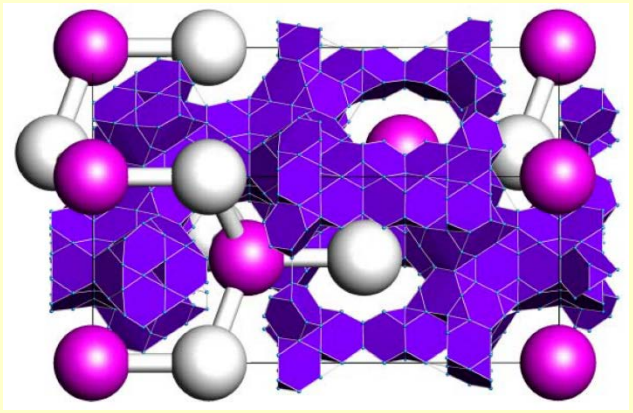
New unusual compounds
& almost room-T superconductivity

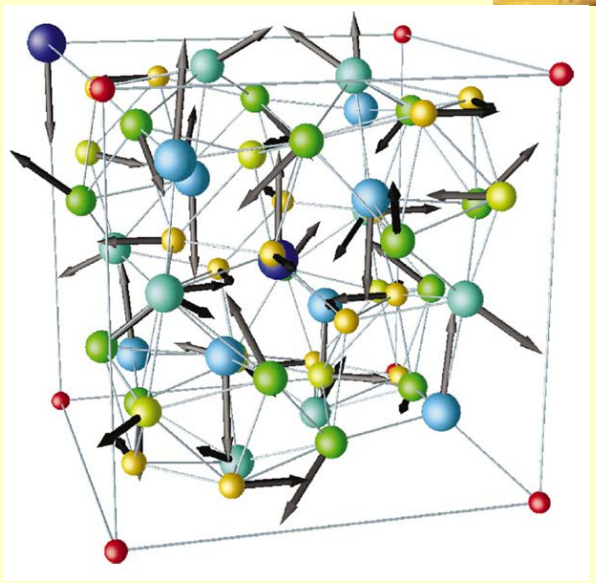


New superhard materials

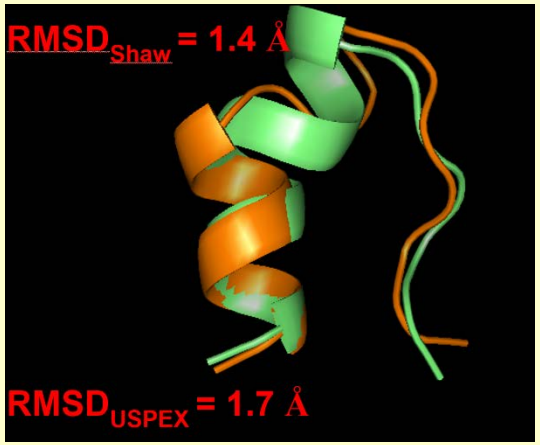
Limitations/challenges of structure prediction methods

Complex metallic alloys: e.g., Samson phase, β-Mg_2Al_3: cF1168
(M. Feuerbacher et al., *Z. Kristallogr.* 222, 259 (2007)).



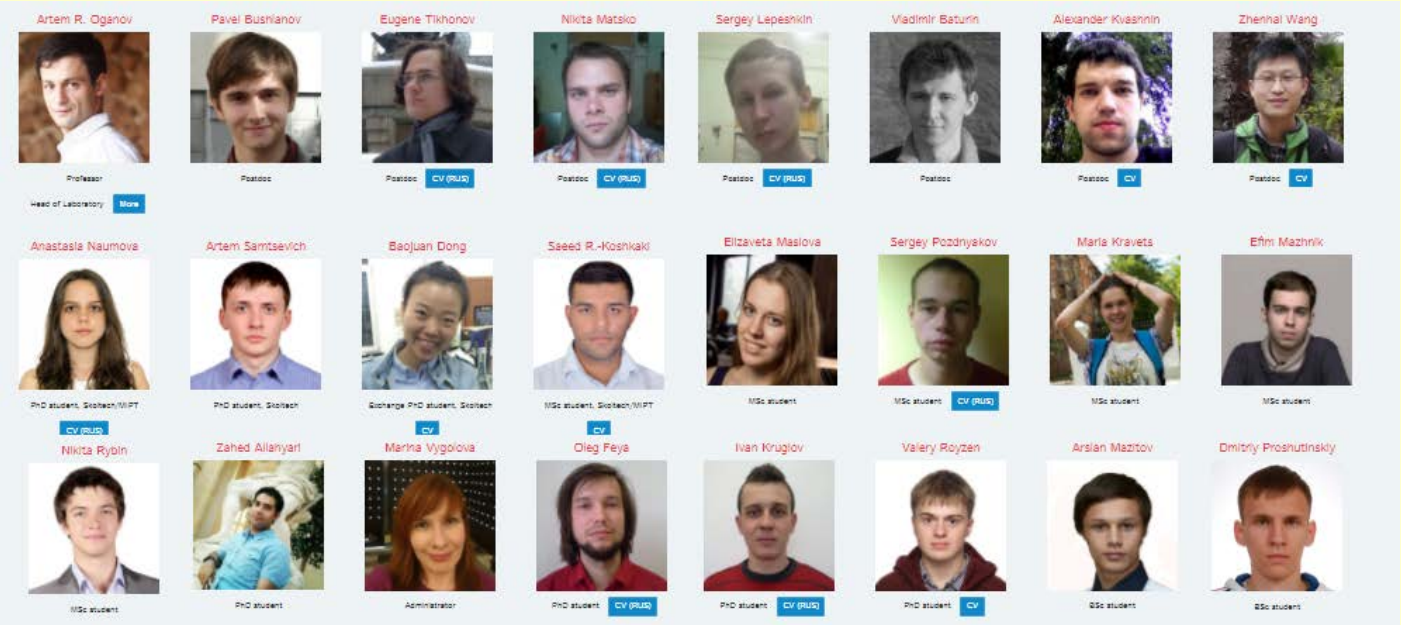


Non-collinear magnets: e.g., α-Mn
(D. Hobbs, J. Hafner, D. Spisak,
PRB **68**, 014407 (2003).



Large proteins: are they thermodynamically controlled?

The team. Where great minds do NOT think alike



Q. Zhu

X. Dong

A. Goncharov

V.A. Blatov

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