Evolutionary Algorithms for Structure Discovery

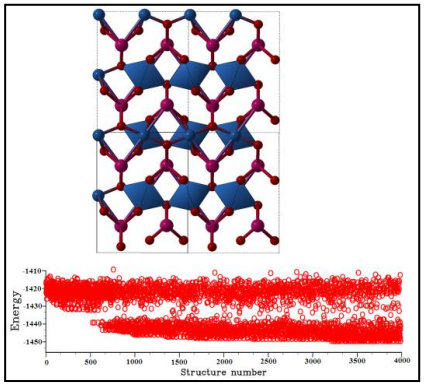
Qiang Zhu

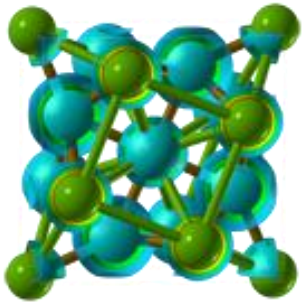
OUTLINE

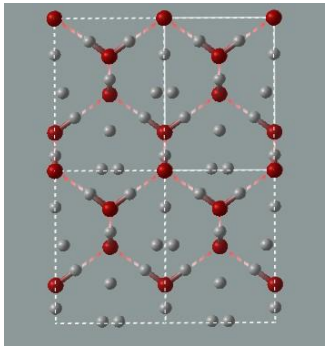
1. (Crystal) Structure Prediction

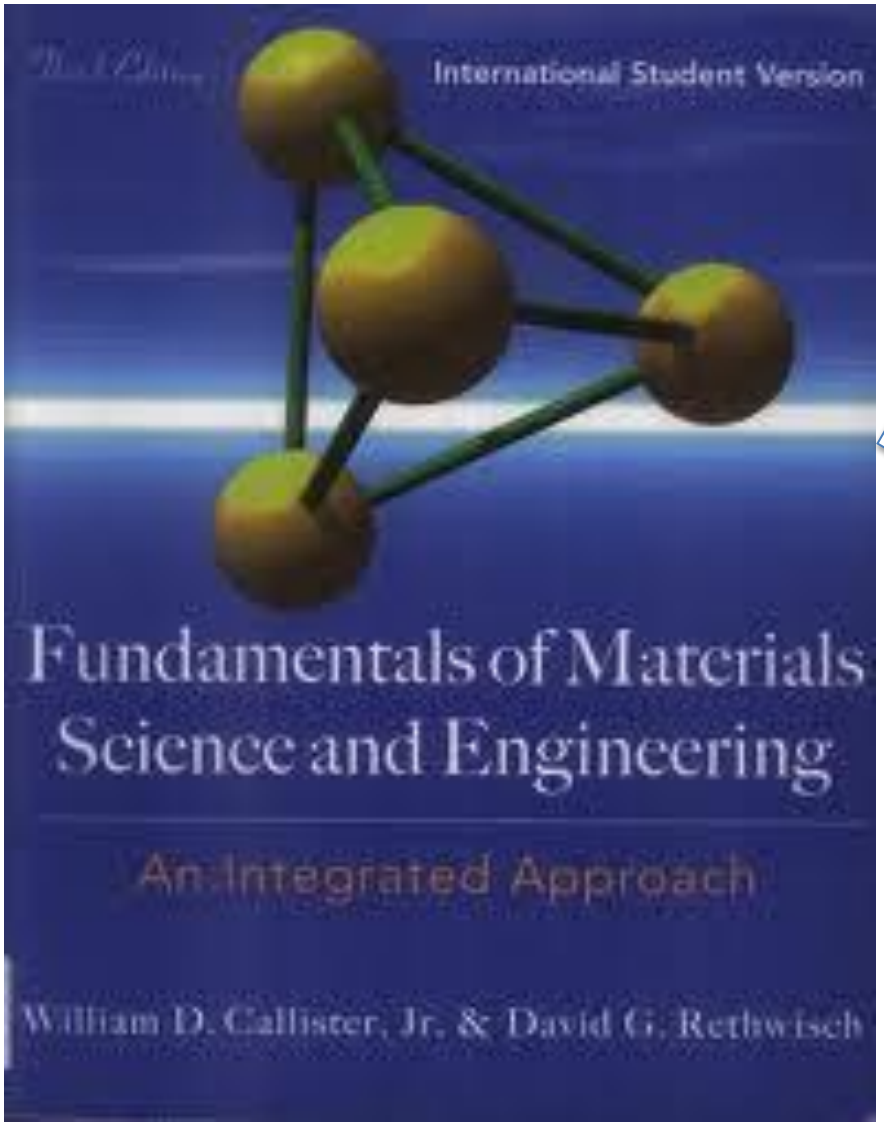
2.1: Variable composition Prediction	2.2 Constrained global optimization	2.3 Low dimensional systems

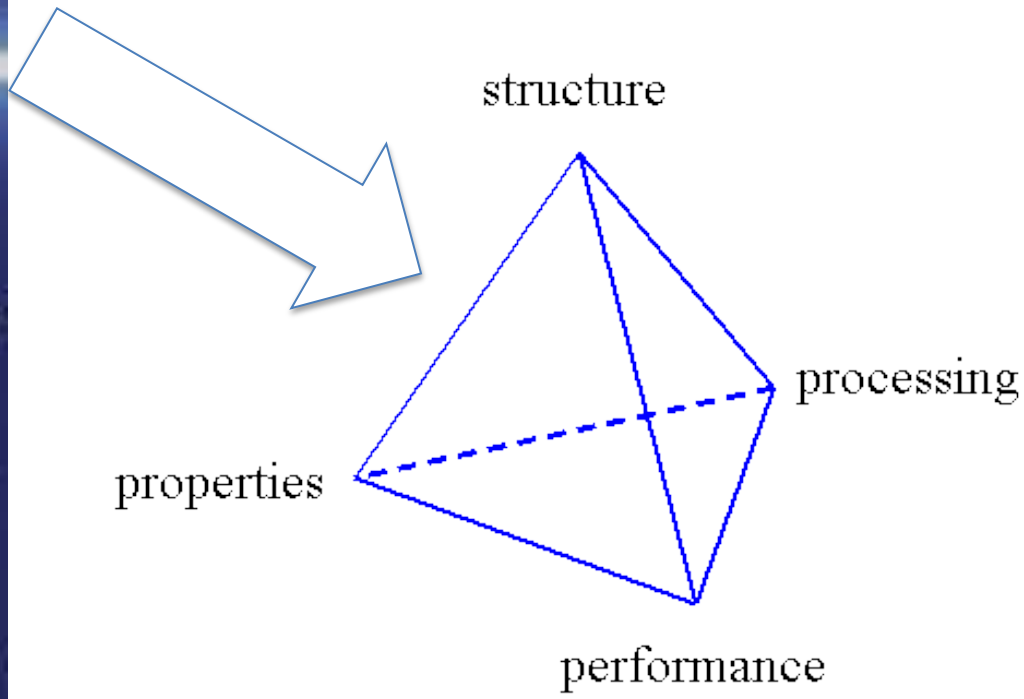
3. OUTLOOK: Materials Discovery & Design








International Student Version
Fundamentals of Materials Science and Engineering
An Integrated Approach
William D. Callister, Jr. & David G. Rethwisch


structure
processing
properties
performance

Crystal Structure is the basis for understanding materials properties and behavior

The Nobel Prize in Physics 1914

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

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

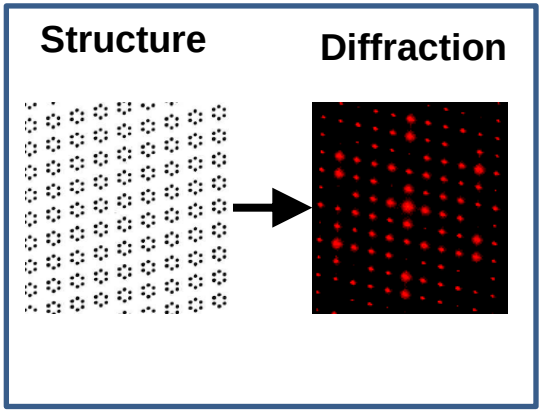
The Nobel Prize in Chemistry 1985

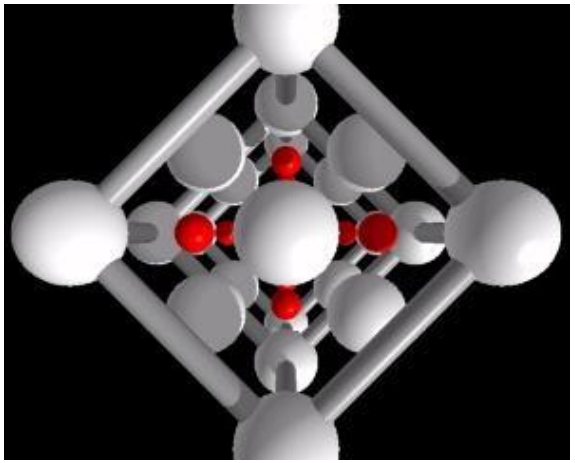
"for their outstanding achievements in the development of direct methods for the determination of crystal structures"

Herbert A. Hauptman

Jerome Karle

(from http://nobelprize.org)





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

Are Crystal Structures Predictable?

ANGELO GAVEZZOTTI*

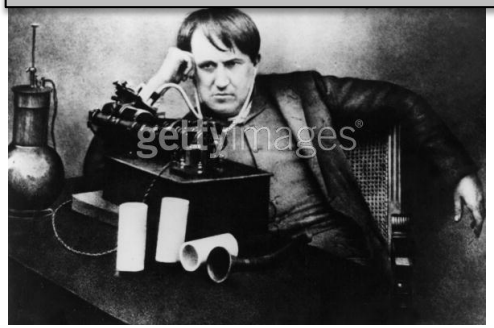
"No": by just writing down this concise statement, in what would be the first one-word paper in the chemical literature, one could safely summarize the present state of affairs

ONE of the continuing scandals in the physical sciences is that it remains in general impossible to predict the structure of even the simplest crystalline solids from a knowledge of their chemical composition. Who, for example, would guess that graphite, not diamond, is the thermodynamically stable allotrope of carbon at ordinary temperature and pressure? Solids such as crystalline water (ice) are still thought to lie beyond mortals' ken.

J. Maddox
(*Nature*, 1988)

To predict crystal structures?

"I have not failed (ten thousand times). I've just found 10000 ways that won't work"
----- Thomas. Edison



Phys. Rev. B26, 5668 (1982)

Theory of static structural properties, crystal stability, and phase transformations: Application to Si and Ge

M. T. Yin* and Marvin L. Cohen
Department of Physics, University of California, Berkeley, California 94720 and Materials and Molecular Research Division, Lawrence Berkeley Laboratory,

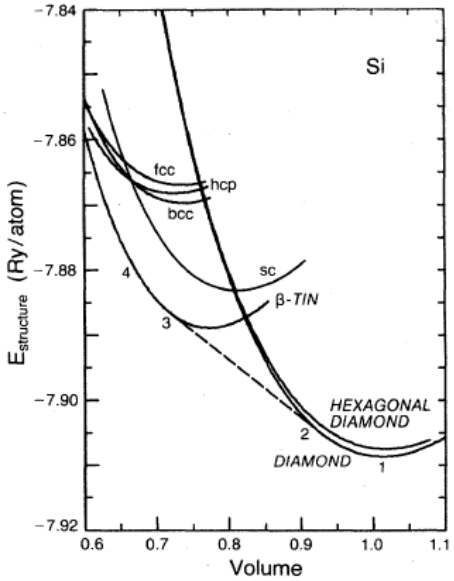
Generation

database/experiment

Energy Ranking

Ab-initio (DFT / DFT-D)

Search is missing!



Modern Crystal Structure Prediction (CSP)

Reliable

- Ranking
- Searching

Universal

Transferable

Economical

Computing time

Automatic

User friendly

CSP Breakthroughs (2000s)...

Inorganic crystals (Breakthroughs since 2003)

- **Metadynamics** (Martonak, 2003)
- **Data mining** (Curtarolo & Ceder, 2003)
- **Minima hopping** (Goedecker, 2004)
- **Evolutionary algorithms(EA) /PSO** (Oganov, 2006; Ma, 2010;)
- **Random sampling** (Pickard, 2006)
- **Genetic algorithms** (Ho, 1997, Ho & Wang, 2010)

RESEARCH NEWS

Crystal structure prediction – evolutionary or revolutionary crystallography?

S. L. Chaplot and K. R. Rao CURRENT SCIENCE, VOL. 91, NO. 11, 10 DECEMBER 2006

Overview of USPEX (Oganov *JCP.* 2006)

Organic crystals

Significant progress in predicting the crystal structures of small organic molecules – a report on the fourth blind test

USPEX

Universal Structure Predictor: Evolutionary Xtallography

- ***Ab initio* determination**

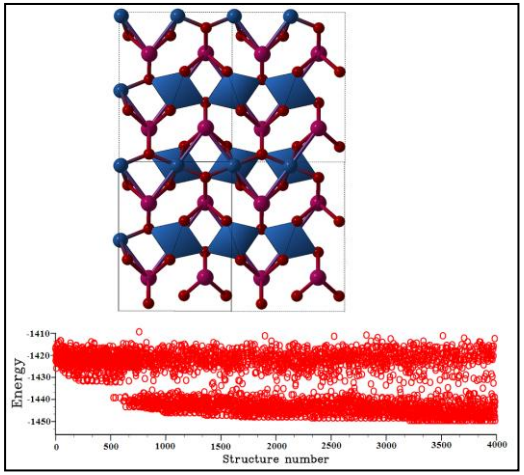
  Few INPUTs (Chemical formula + P/T condition)

- ***Evolutionary Algorithm***

  Self-improved searching process

Website: http://uspex.stonybrook.edu

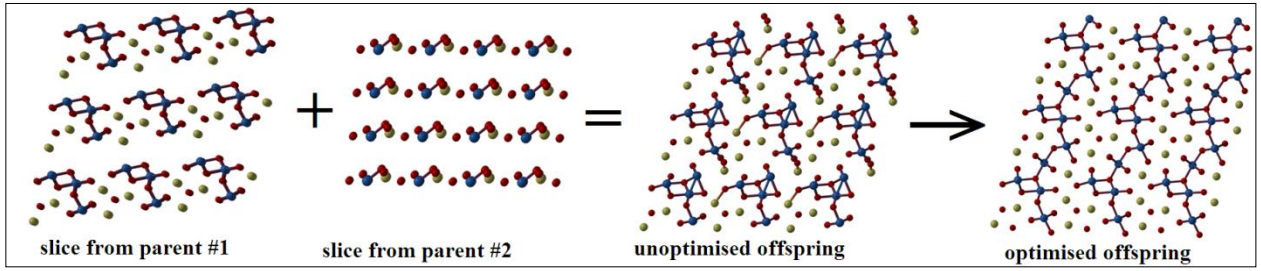
Oganov A.R., Lyakhov A.O., Valle M. (2011).
How evolutionary crystal structure prediction works - and why.
***Acc. Chem. Res.* 44, 227-237.**



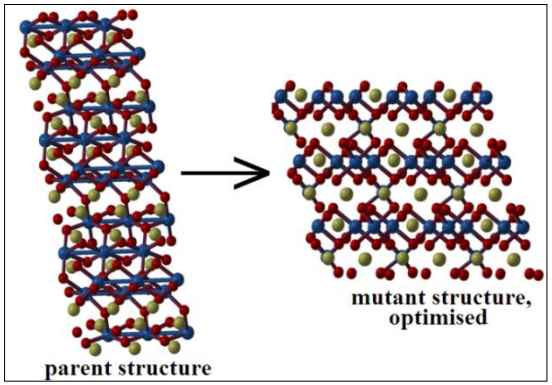
USPEX

1) **(Random) initial population:** from INPUTs : chemical formula
2) **Relaxation** at P/T conidtion : (free) energy (done by VASP, .etc)
3) **Selection**: lowest-energy structures as parents
4) **New population**: Standard variation operations

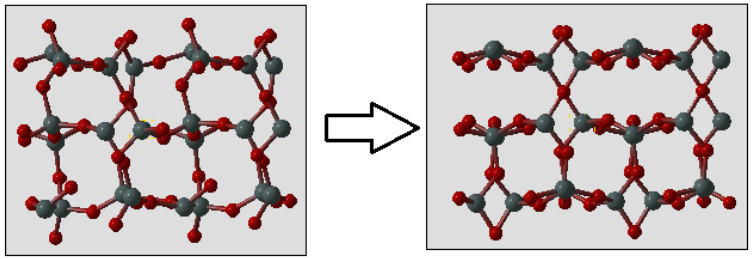
(1) Heredity



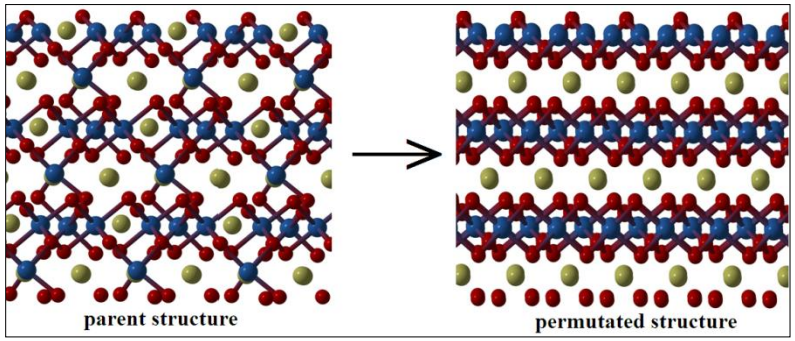
(2) Lattice mutation



(3) Softmutation



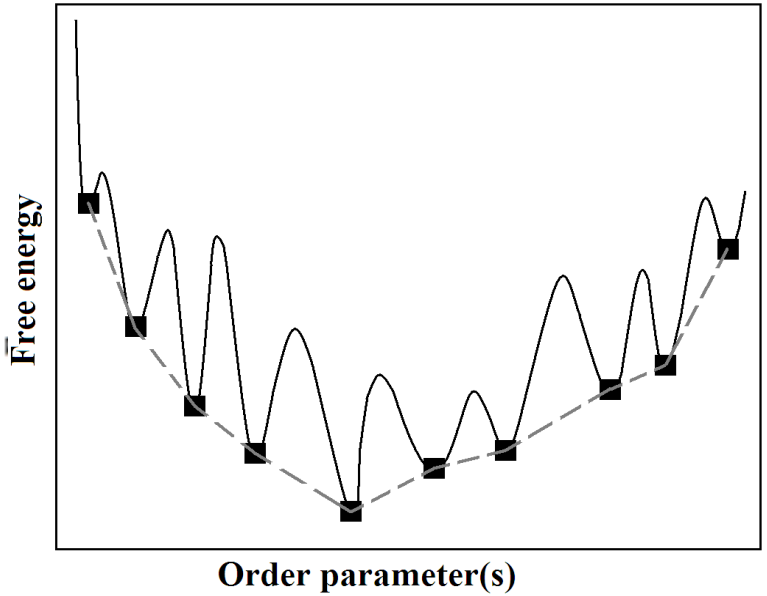
(4) Permutation



Modern CSP theory

CSP Problem:
Find global energy minimum

- NP hard Problem ~ exp(βd)
- Dimensionality $d = 3N + 3$.
- Very sensitive to small changes
- HUGE and 'noisy' landscape



CSP Solution:
Global optimization of local minima

- Reduced dimensionality ($d^* = 3N + 3 - \kappa$)
- Simplified curvature of energy landscape

CSP complexity

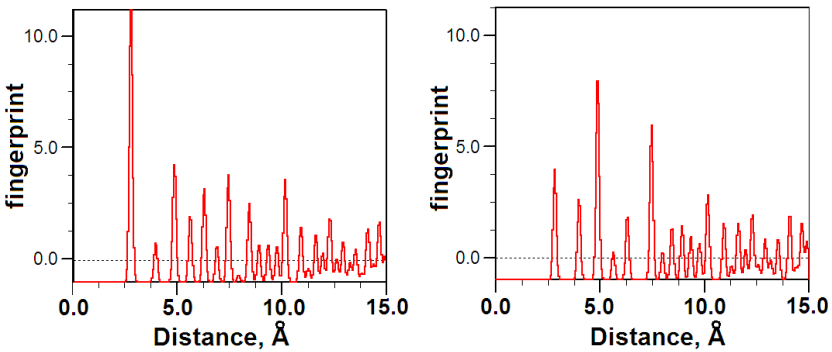
	d	d^*
Au_8Pd_4	39	10.9
$Mg_4N_4H_4$	39	32.5
$Mg_{16}O_{16}$	**99**	11.6

ARO et al., Acc. Chem. Res. 2011

Fingerprint Theory

Fingerprint function : a 1D-descriptor of the structure
Similar to diffraction spectrum, PCF, ...

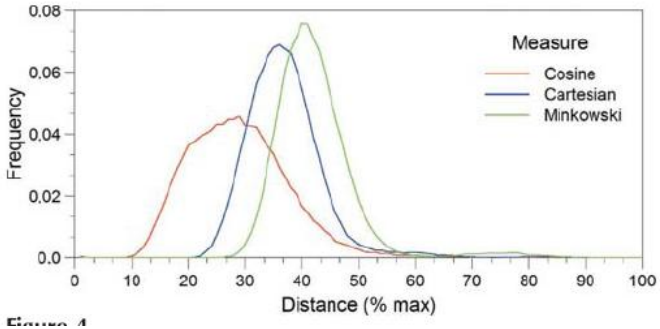
$$F_{AB}(R) = \sum_{A_i,\text{cell}} \sum_{B_j} \frac{\delta(R - R_{ij})}{4\pi R_{ij}^2 \frac{N_A N_B}{V} \Delta} - 1 = g_{AB}(R) - 1$$



Difference between 2 structures is given by "distance", e.g.:

$$\text{dist}(i, j) = \frac{1}{2}\left(1 - \frac{F_i \cdot F_j}{\|F_i\| \, \|F_j\|}\right).$$

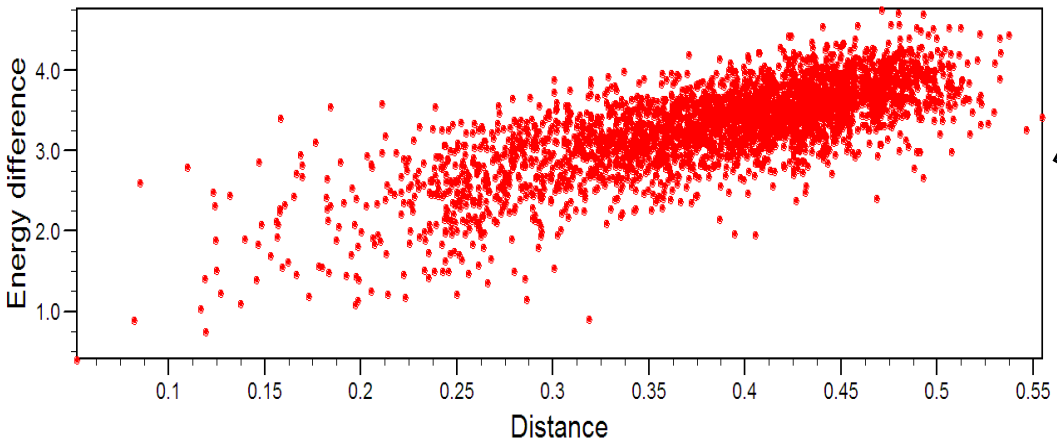
Cosine distance



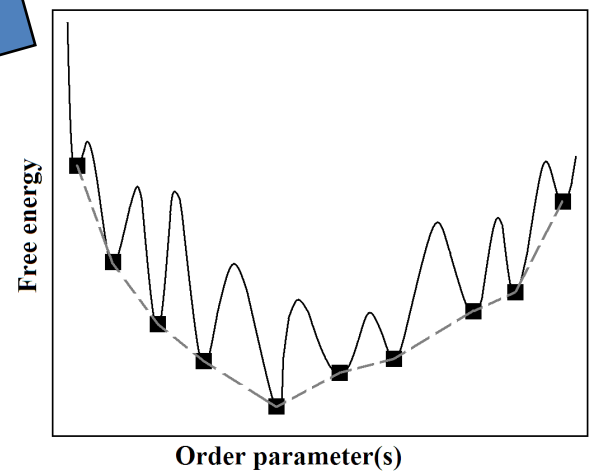
ARO & Valle, J. Chem. Phys., 2009

Simple systems

GaAs



Energy v.s. distance plot



1 D representation of funnel

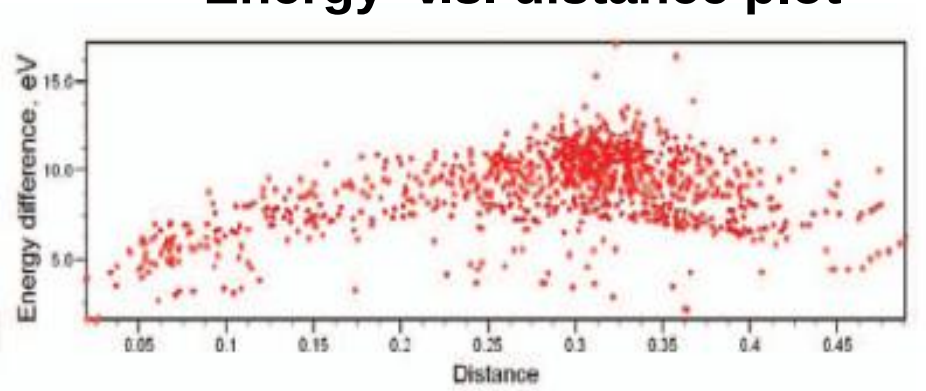
Sampling in discrete space, while optimization makes it continuous

- **Heredity**
- **Mutation (small degree)**

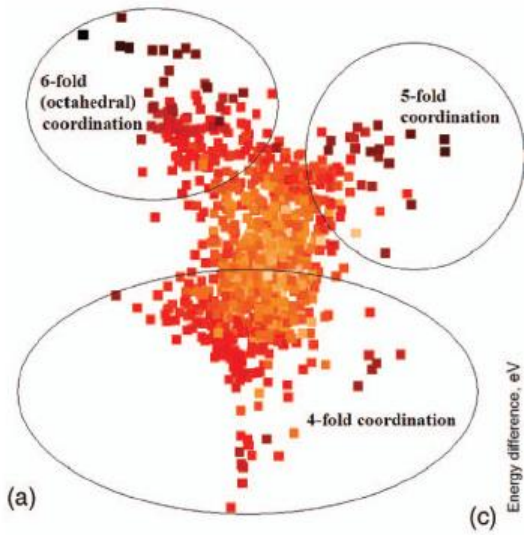
Complex systems

MgO

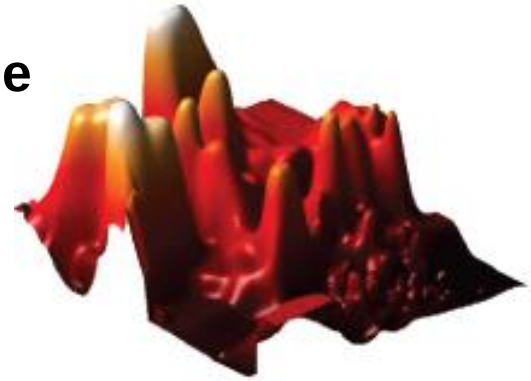
Energy v.s. distance plot



2D projection



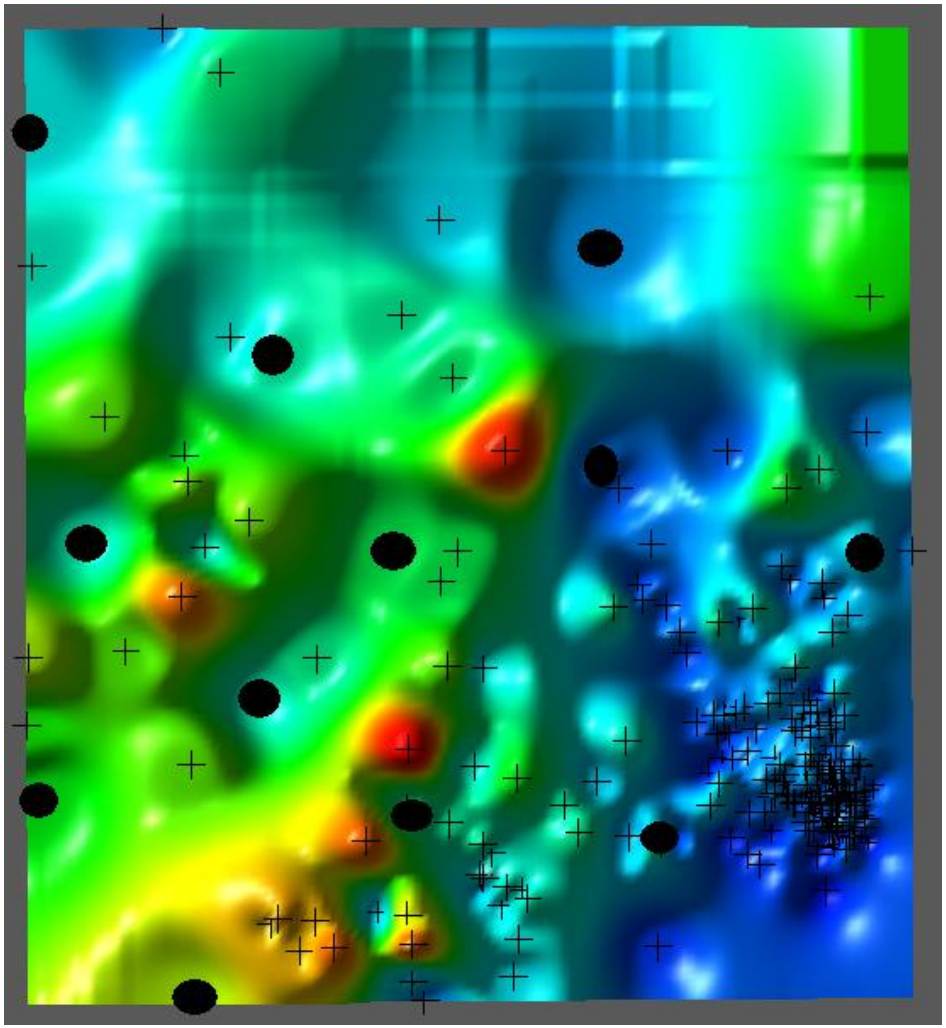
Energy landscape

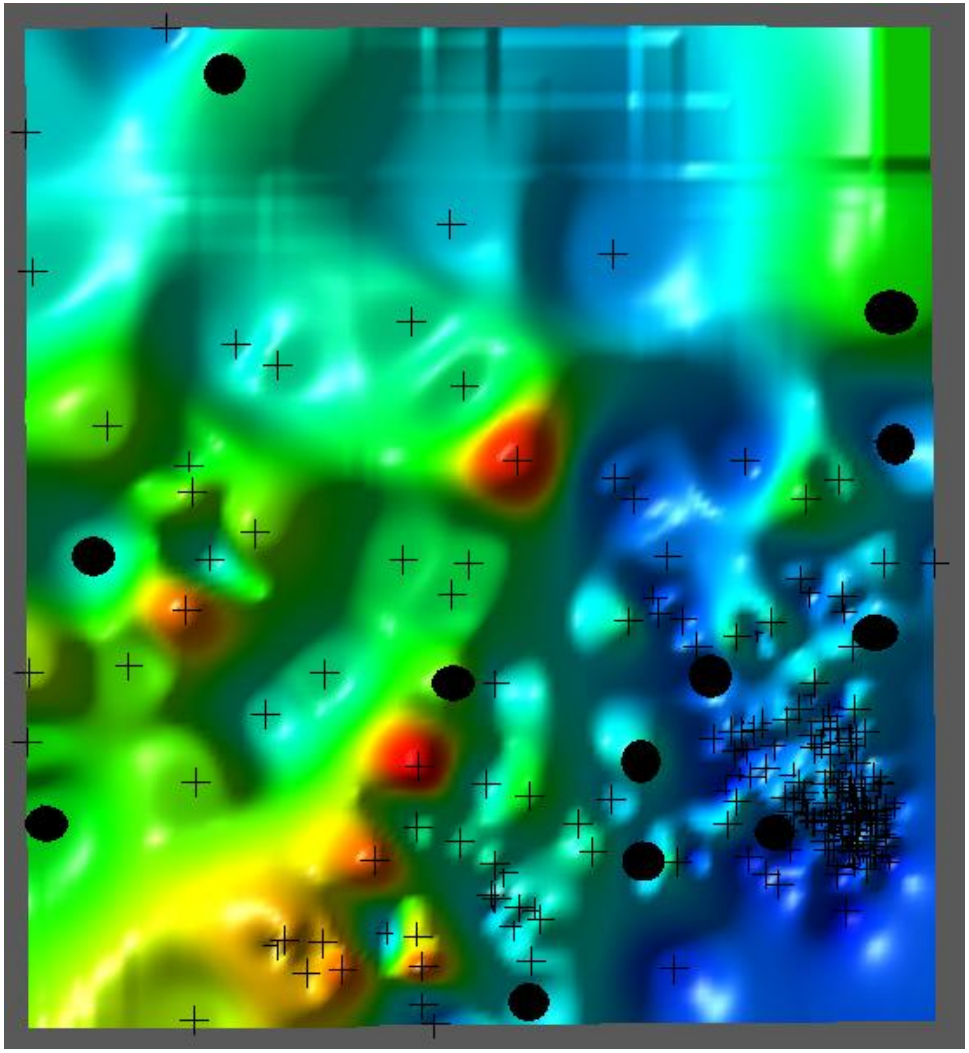


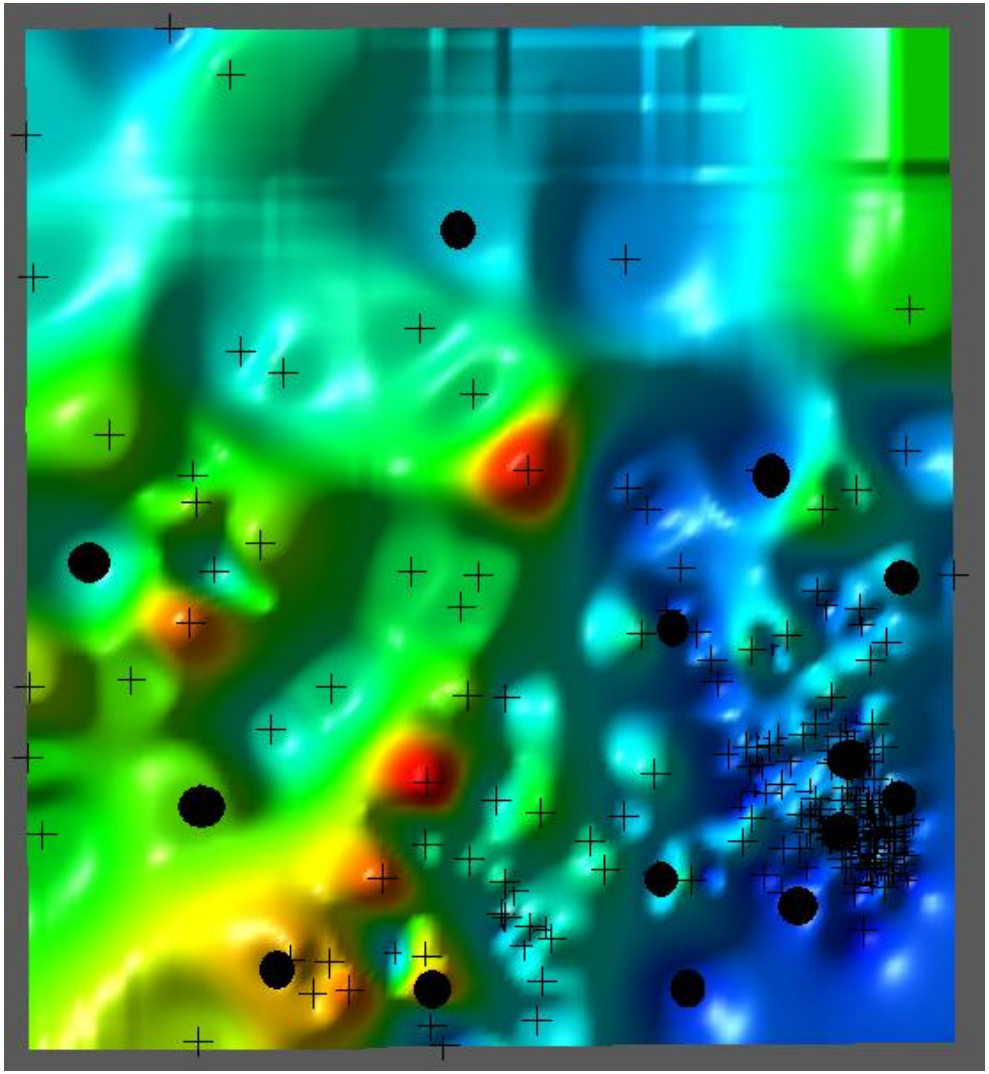
Structural Diversity is essential to explore the complex landscape!

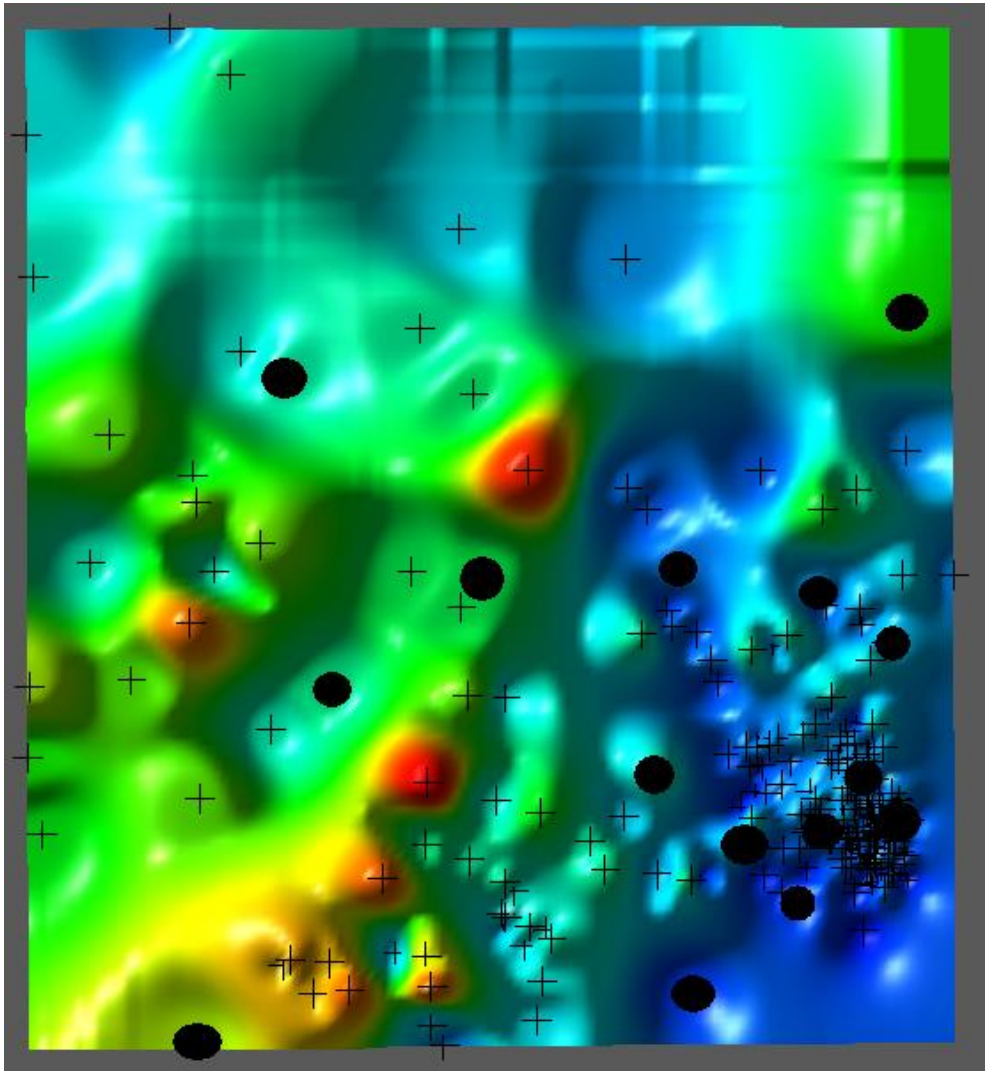
- **Random generation (new blood)**
- **Mutation (large degree)**

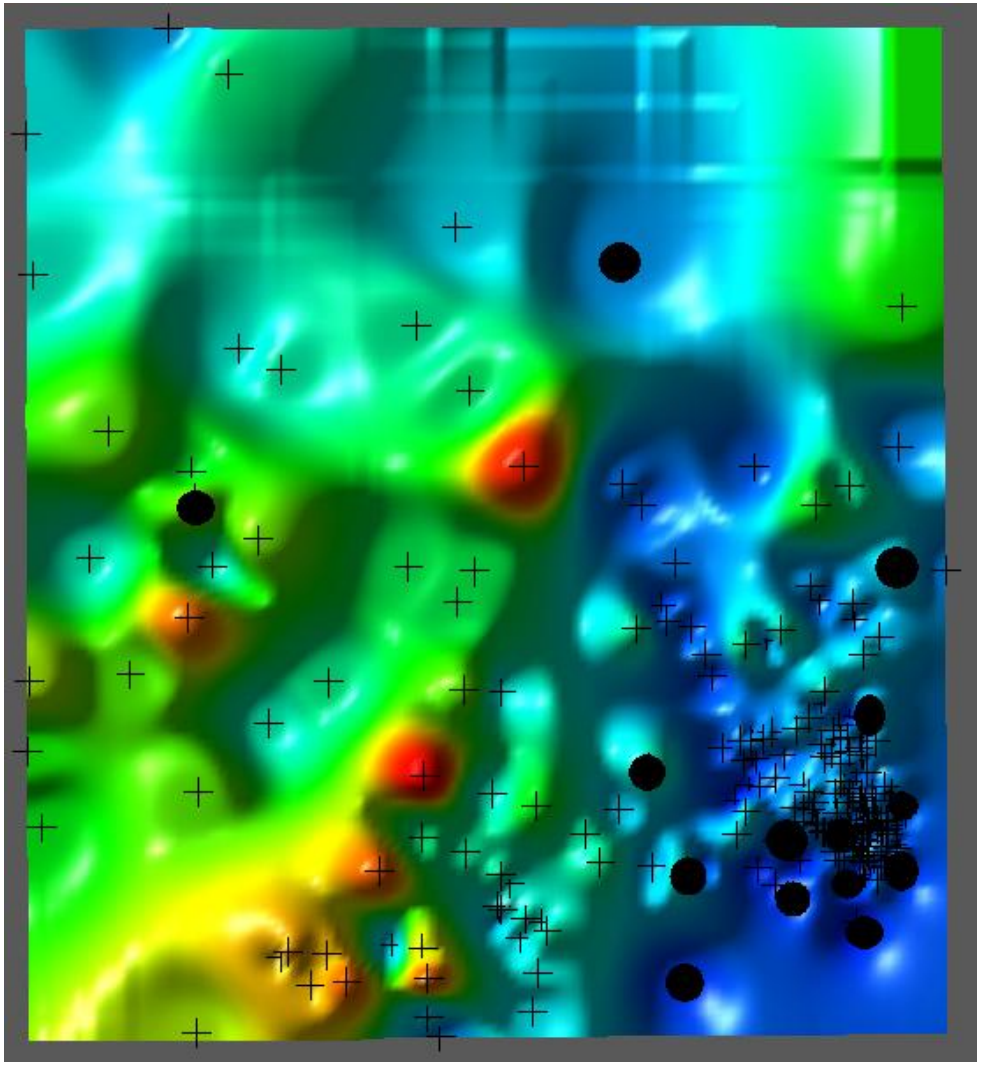
ARO & Valle, J. Chem. Phys. , 2009





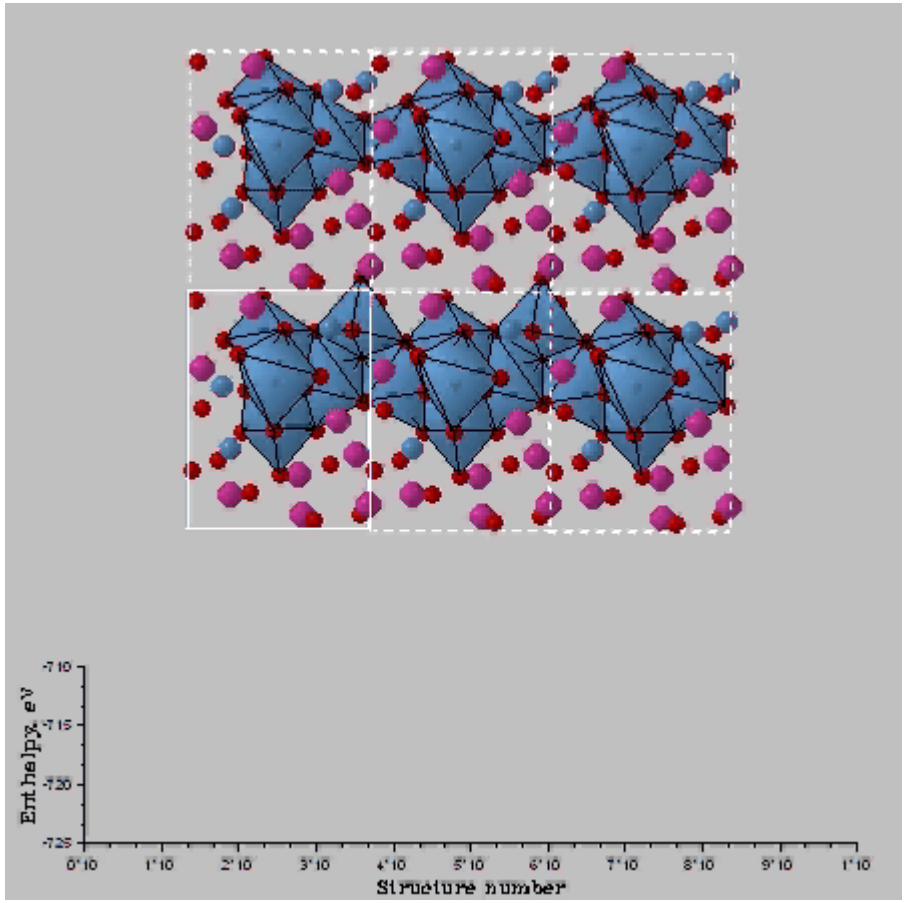




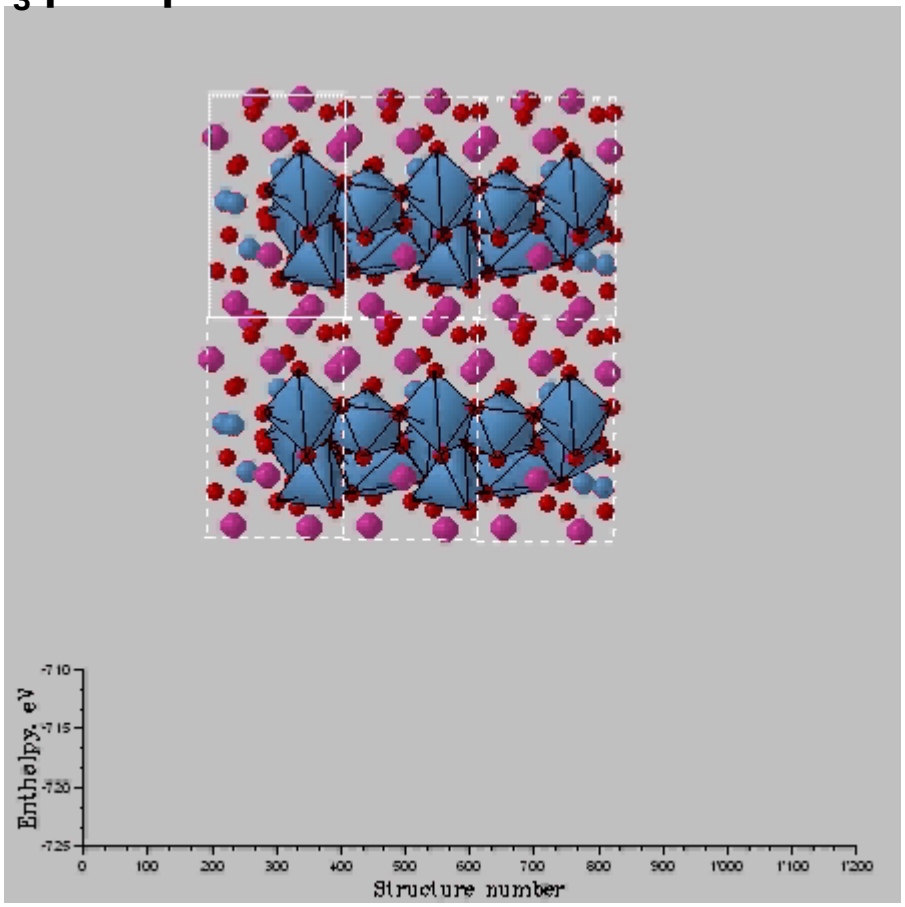


When searching efficiency matters!

40-atom cell of $MgSiO_3$ post-perovskite



Random



USPEX

Part II: Structure Prediction

I. Variable compositional prediction

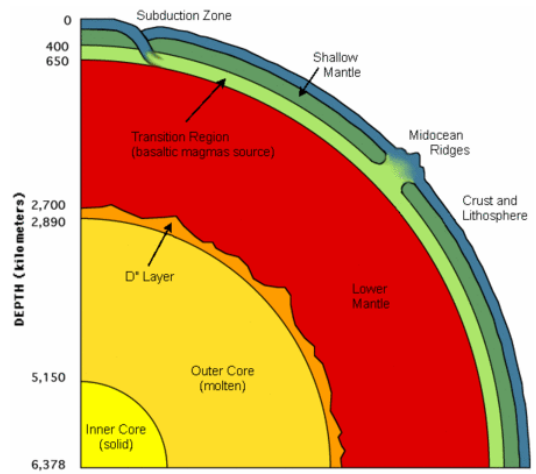
II. Constrained global optimization

III. Low dimensional systems

Missing Xenon Paradox

" our atmosphere contains far less xenon, relative to the lighter noble gases, than meteorites similar to the rocky material that formed the Earth,"

----- Anderson, E, .*Science*, 1977



Hypothesis: Xenon is stored in the Earth's mantle ?
(most likely with perovskite, oxides or silicates)

Sanloup et al, Science, 2005

Xenon Chemistry

Ambient condition

Xe^{II} — XeF_2, XeF^+ and $Xe_2F_3^+$ salts, XeF_3^-

Xe^{IV} — XeF_4, XeF_5^- salts, $XeOF_2$

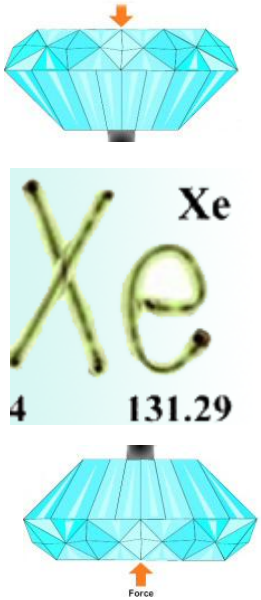
Xe^{VI} — XeF_6, XeF_5^+, XeF_7^- and $Xe_2F_{13}^-$ salts, XeO_3, $XeOF_2$

Xe^{VIII} — XeO_4, H_4XeO_6, XeO_6^{4-}salts

Xenon does exhibit multiple valence state

Grochala, Chem. Soc. Rev, 2007

Earth's mantle condition



Prediction on Xe-O system

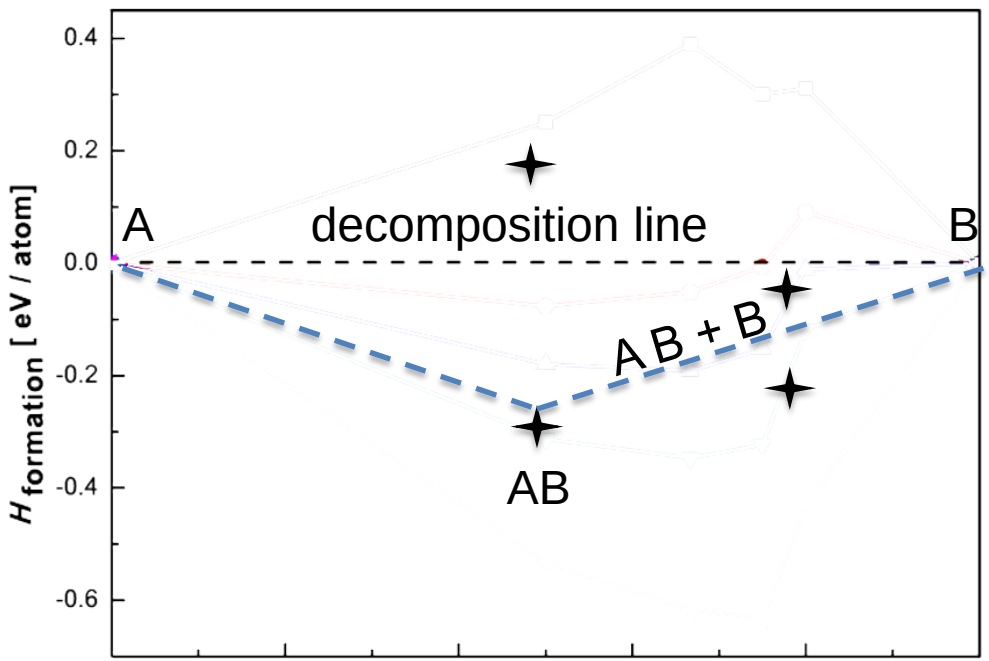
Pressures(GPa):

5, 50, 100, 120, 150, 180, 200 , 220

Stoichiometry:

XeO, XeO_2, XeO_3, XeO_4

Thermodynamical Stability

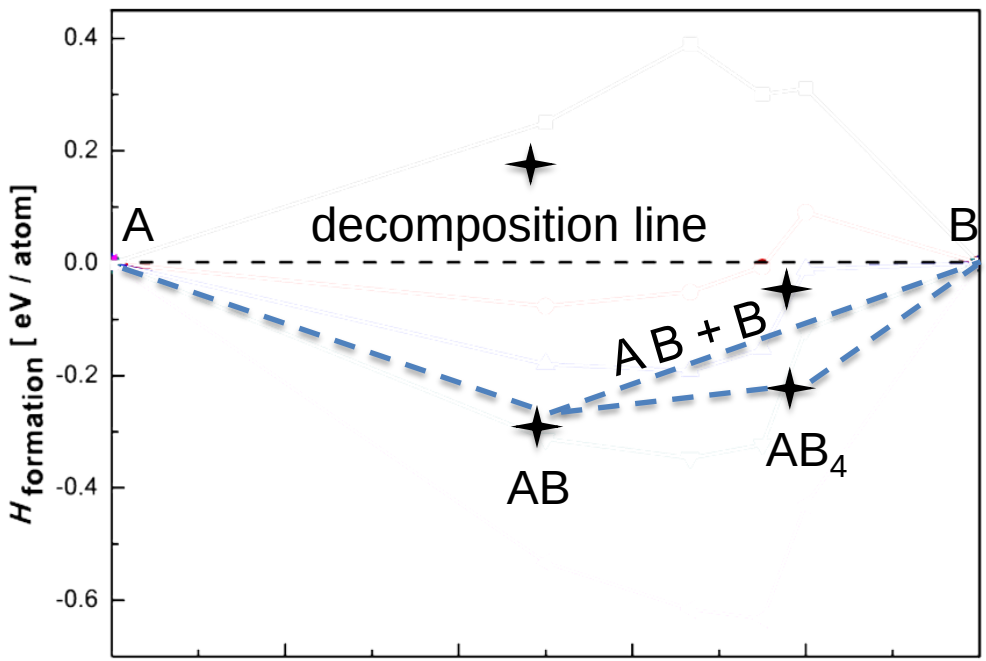


$$A + B \longrightarrow AB$$

$[E_{AB} - (E_A + E_B)]>0$ AB decompose

$[E_{AB} - (E_A + E_B)]<0$ AB is stable

Thermodynamical Stability



$$A + B \longrightarrow AB$$

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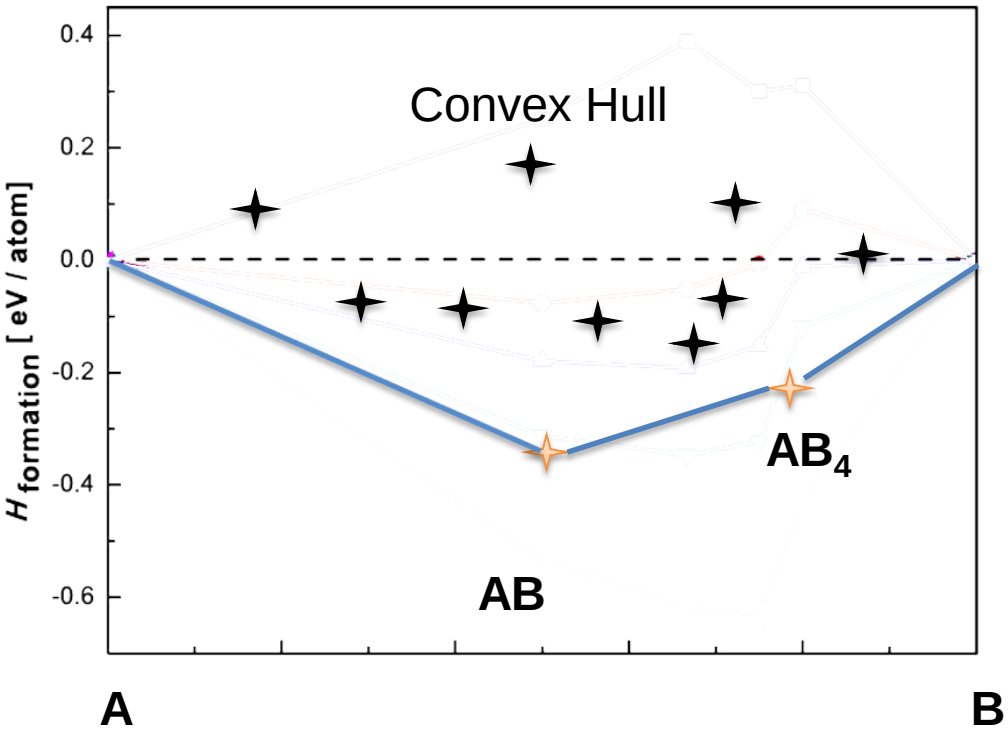
$[E_{AB} - (E_A + E_B)]<0$ AB is stable

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

Thermodynamical Stability



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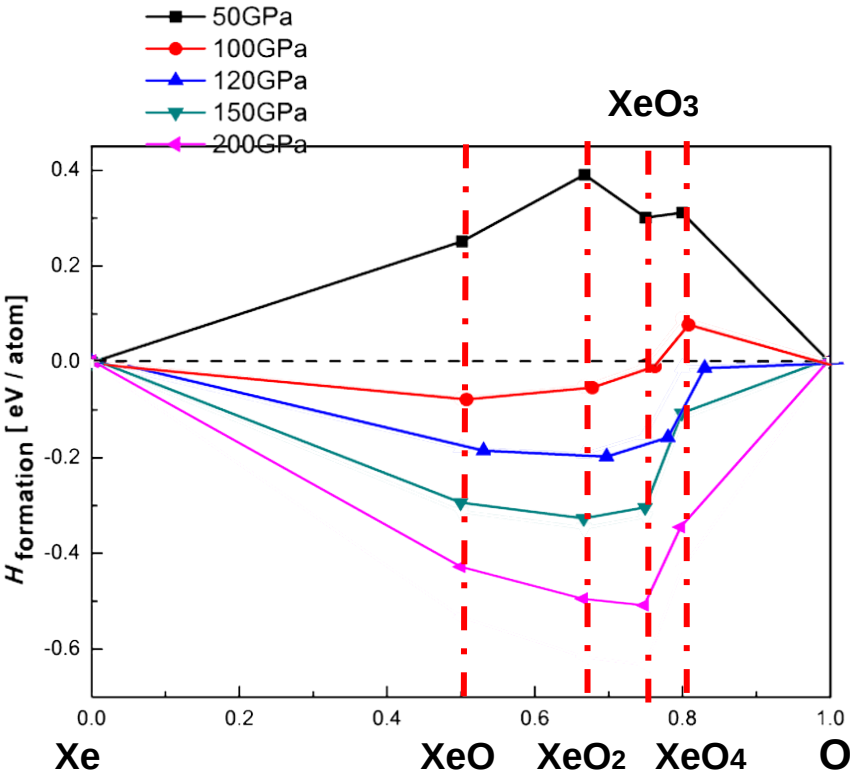
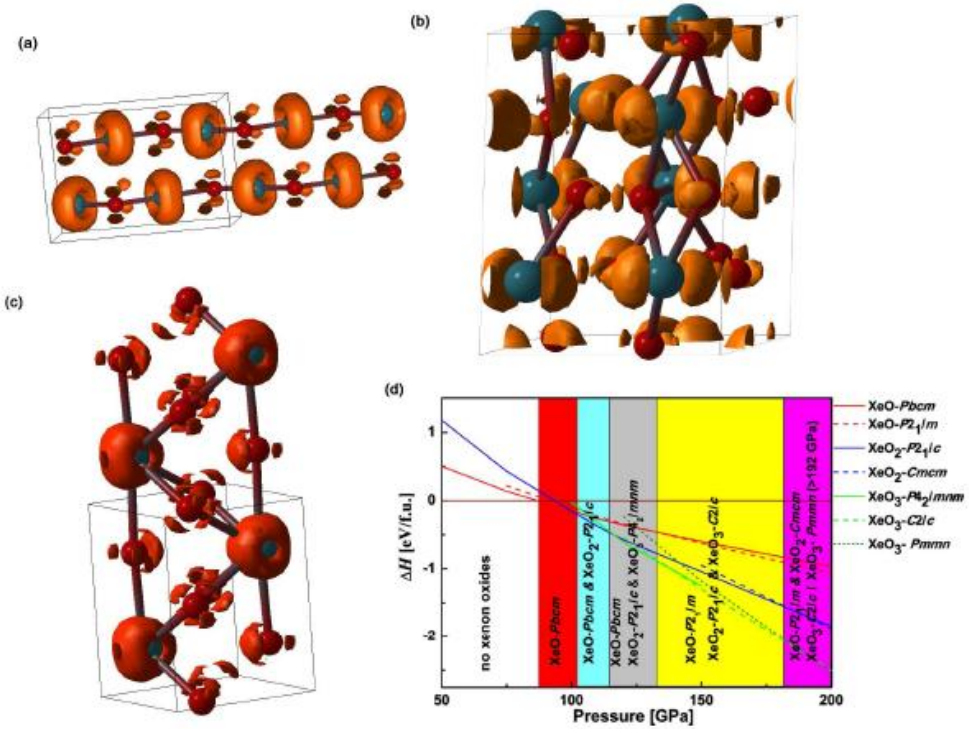
Stable structure must be below all the possible decomposition lines !!

Xenon oxides at high pressure





Zhu, et al, Nature Chemistry, 2013

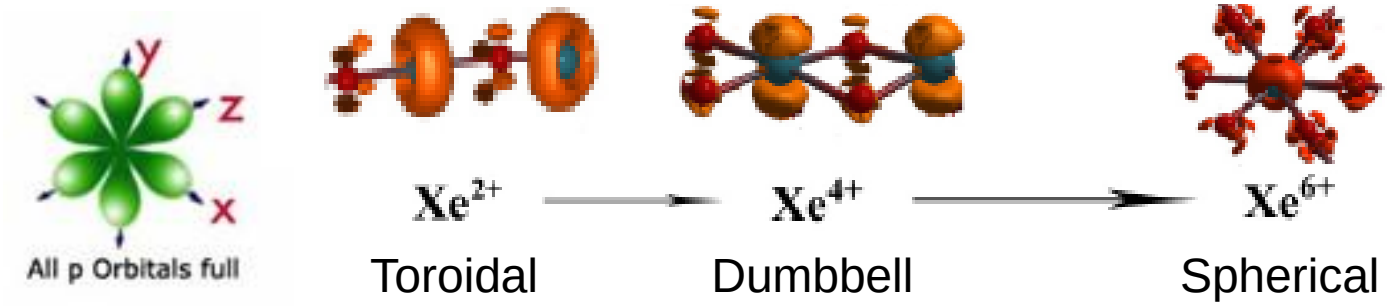
nature chemistry

ARTICLES

PUBLISHED ONLINE: XX XX 2012 | DOI: 10.1038/NCHEM.1497

Stability of xenon oxides at high pressures

Qiang Zhu[1]*, Daniel Y. Jung[2], Artem R. Oganov[1,3]*, Colin W. Glass[4], Carlo Gatti[5] and Andriy O. Lyakhov[1]

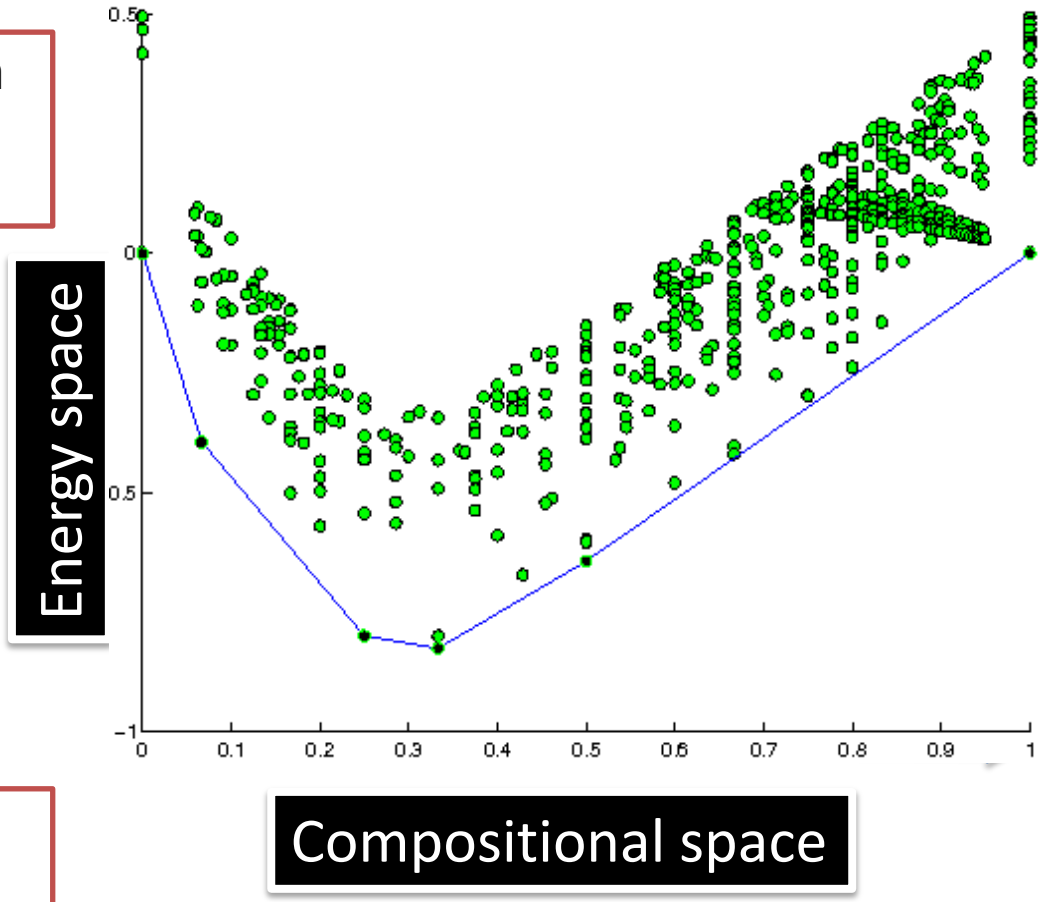


Rule:

New stoichiometric compounds might exist under high pressures ! -- can we predict them automatically?

To *predict* all stable compositions and structures simultaneously?

Q1: How to evaluate the quality of each structure with different compositions?
A1: Optimization target: convex hull

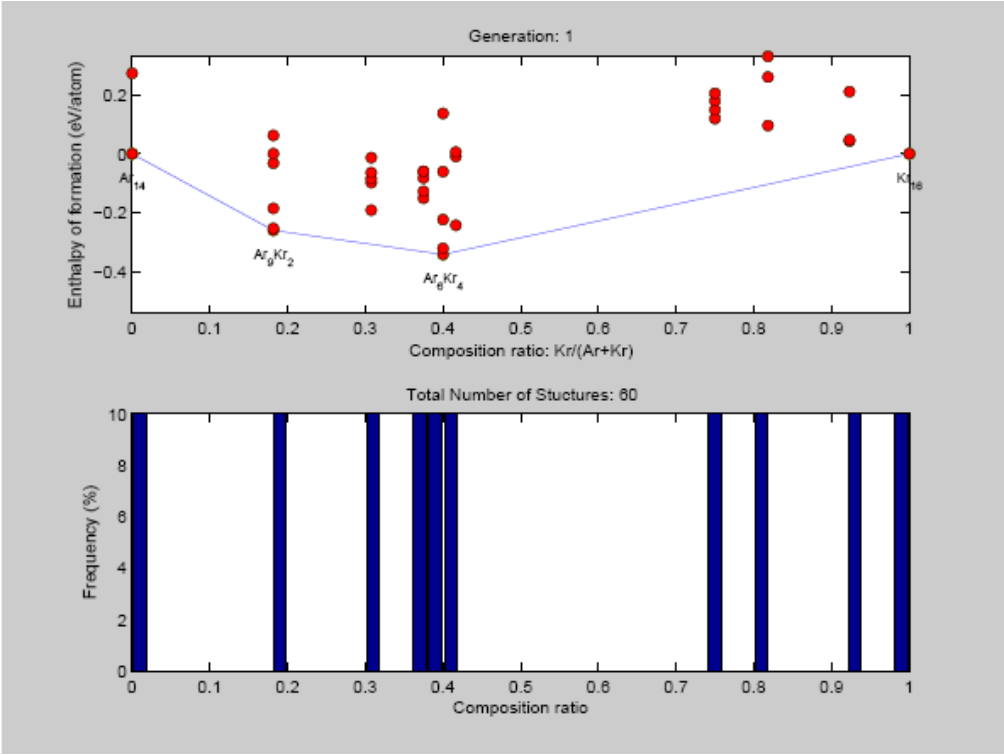


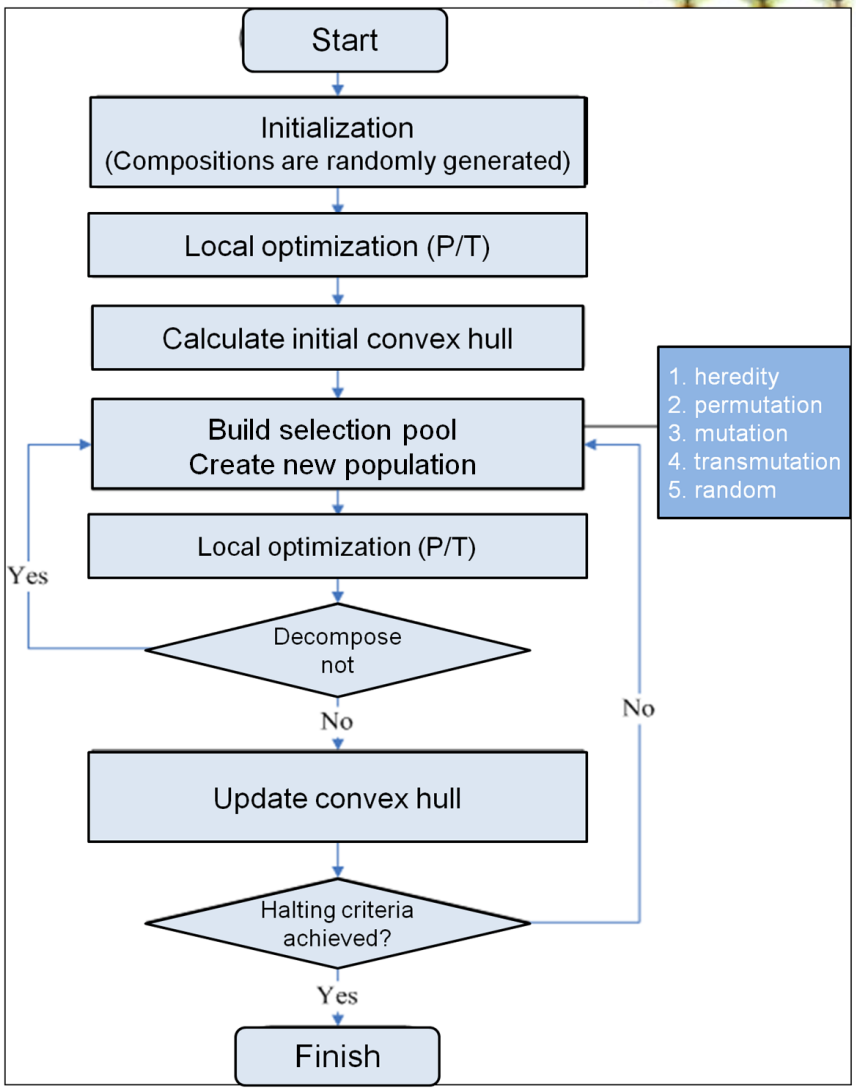
Q2: Allow the compositional variation?
A3: Smart variation operators

Algorithm

A_xB_y

Lennard-Jones system



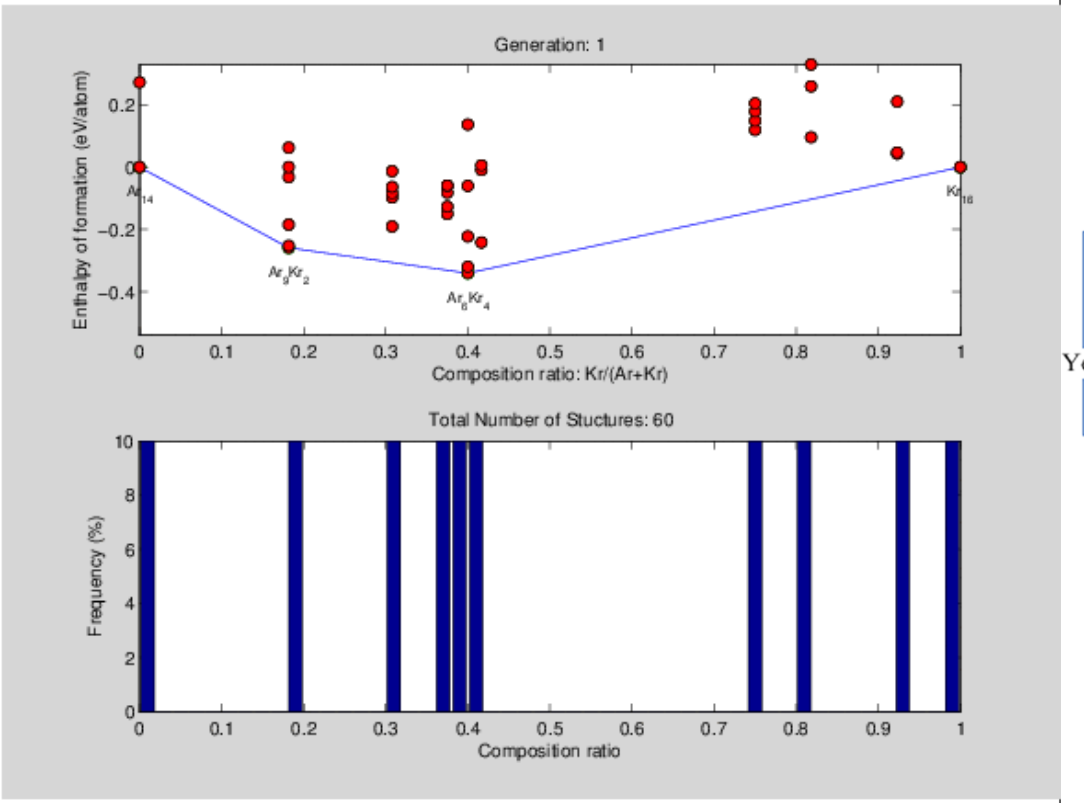


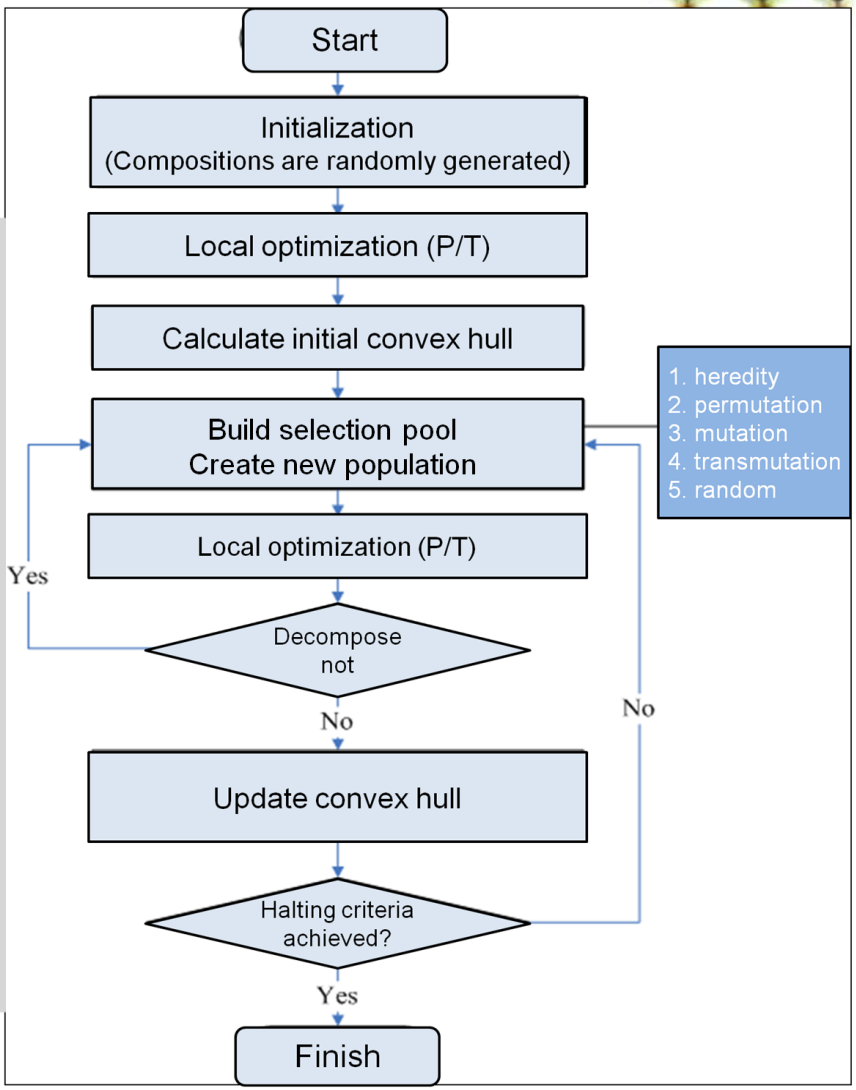
Zhu, et al, Springer Series, 2013

Algorithm

A_xB_y

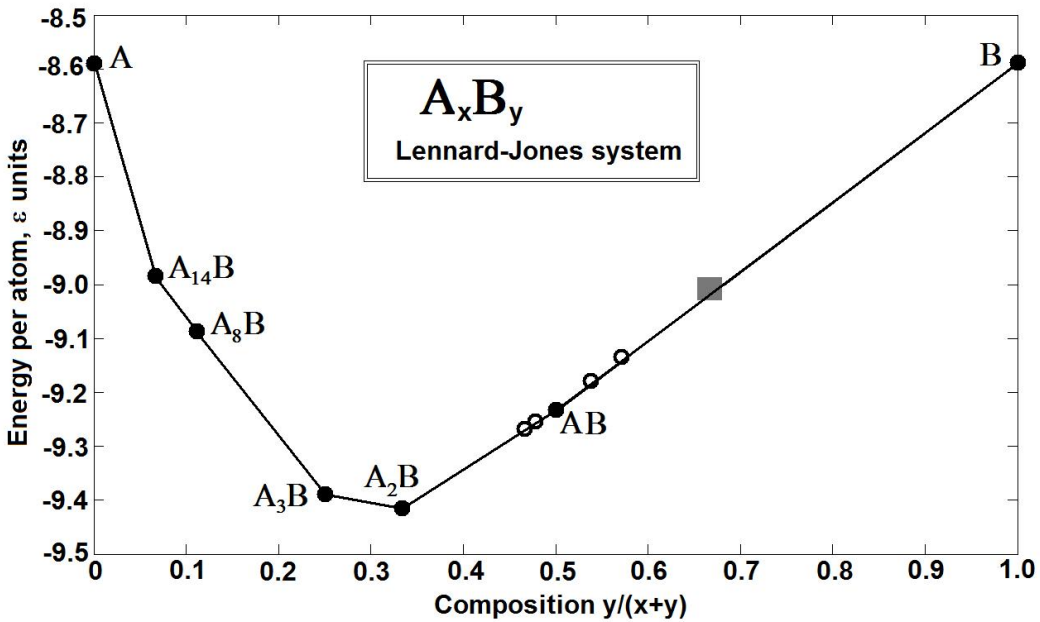
Lennard-Jones system

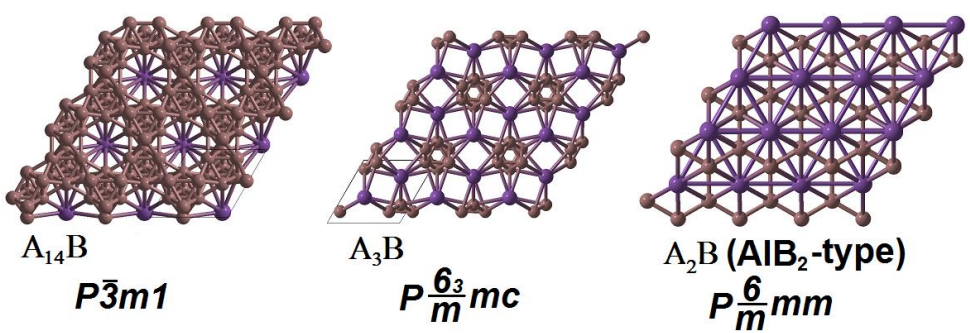


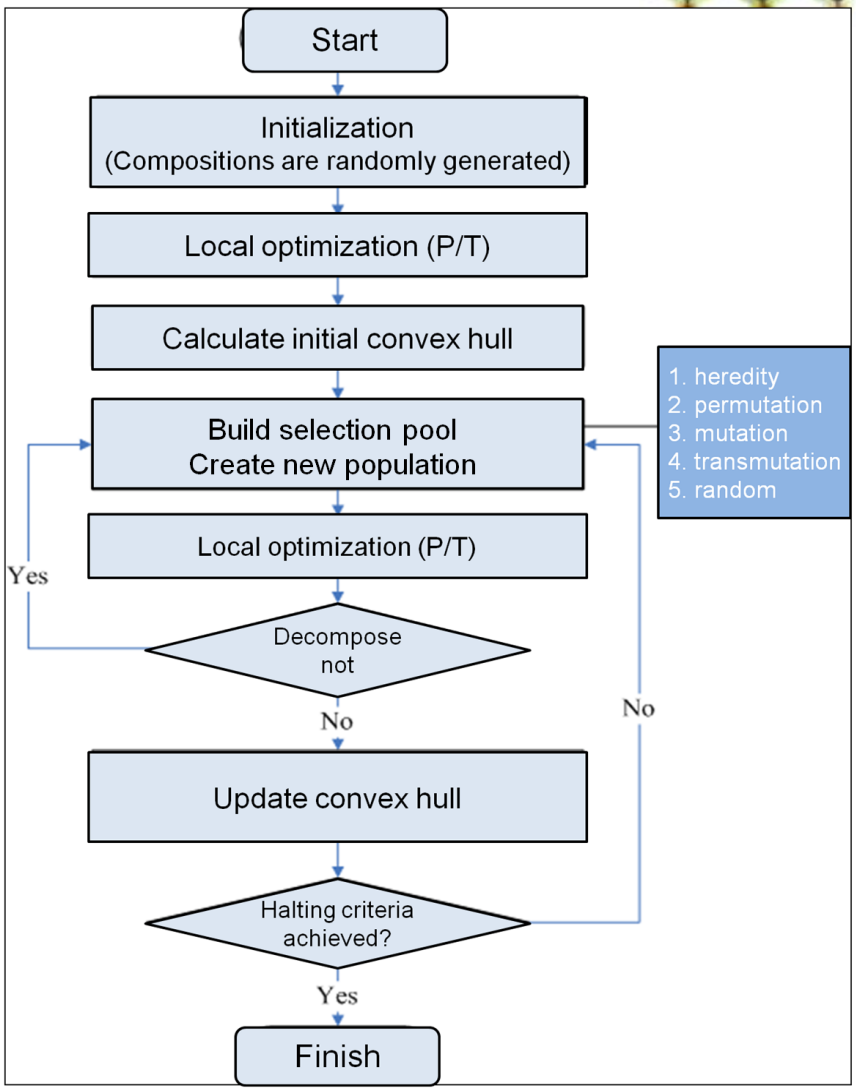


Zhu, et al, Springer Series, 2013

Algorithm



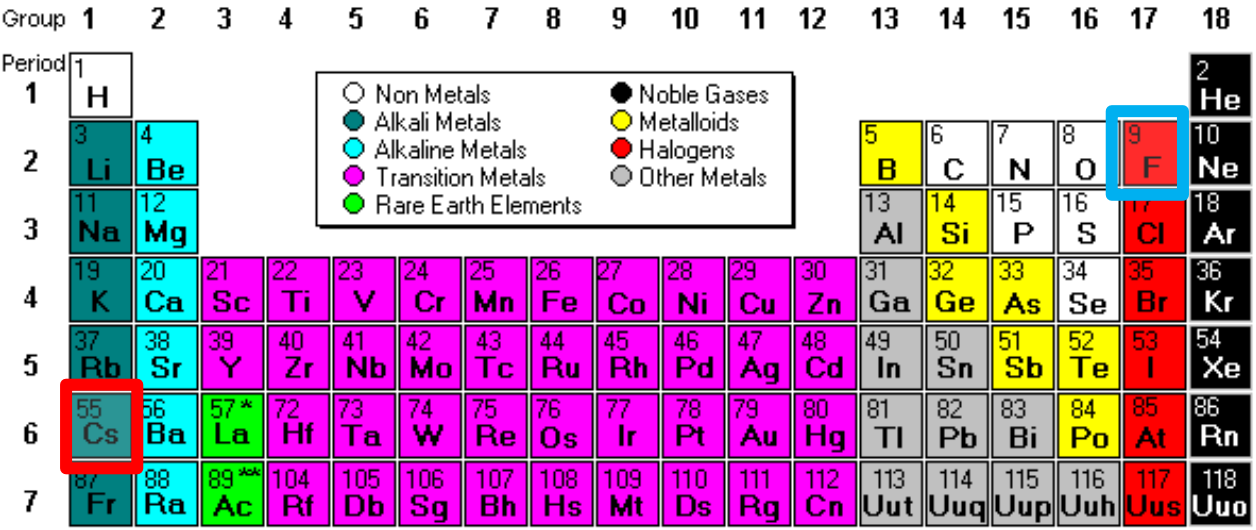


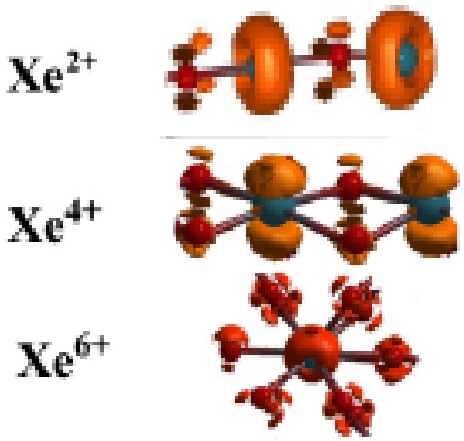


Zhu, et al, Springer Series, 2013

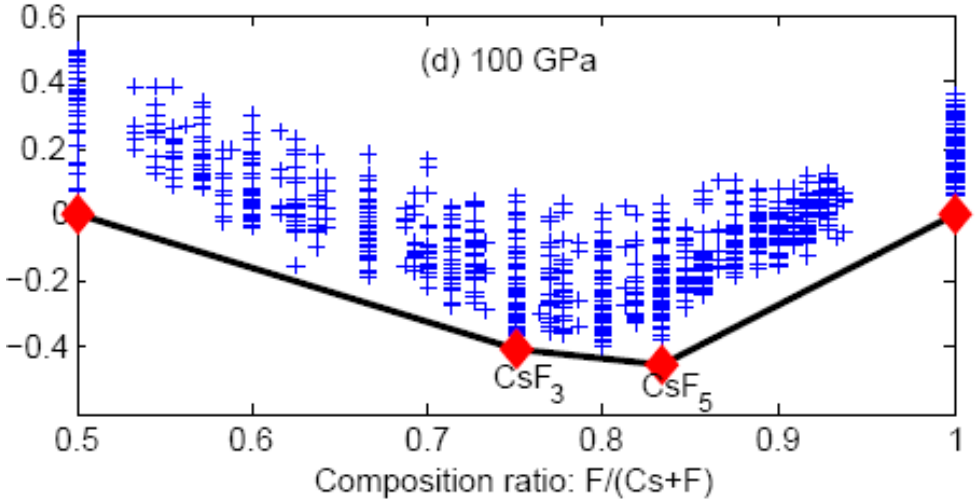
Applications

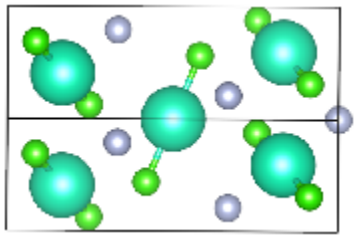
Cs-F



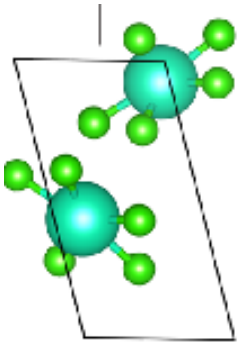


Can Cs^+ be oxidized further?





$[CsF_2]$ F



CsF_5

arXiv:1408.5551

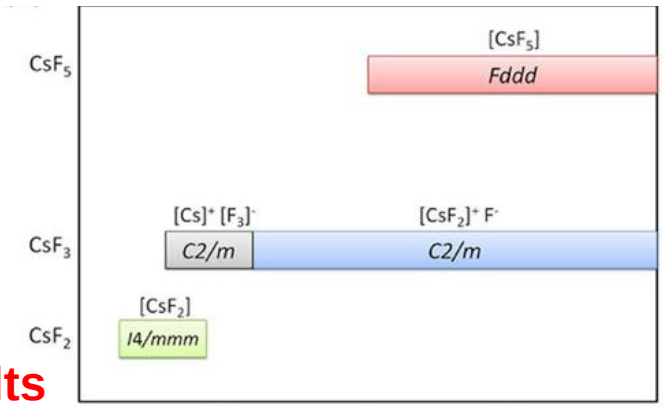
Applications

Cs-F

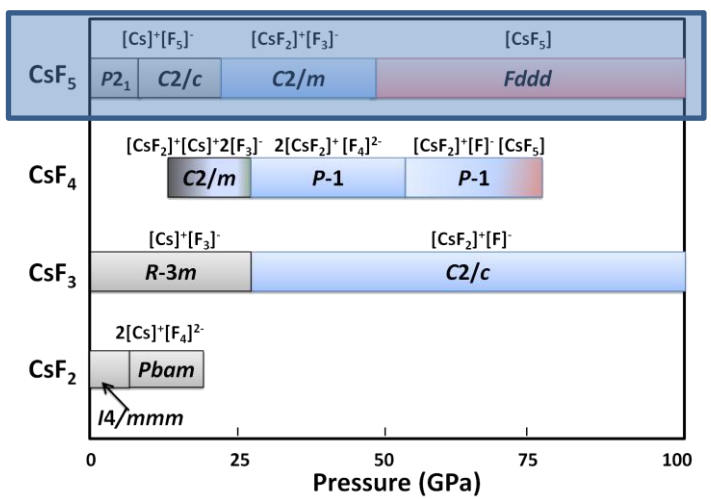
ARTICLES
PUBLISHED ONLINE: 23 SEPTEMBER 2013 | DOI: 10.1038/NCHEM.1754
nature chemistry

Caesium in high oxidation states and as a *p*-block element

Mao-sheng Miao*

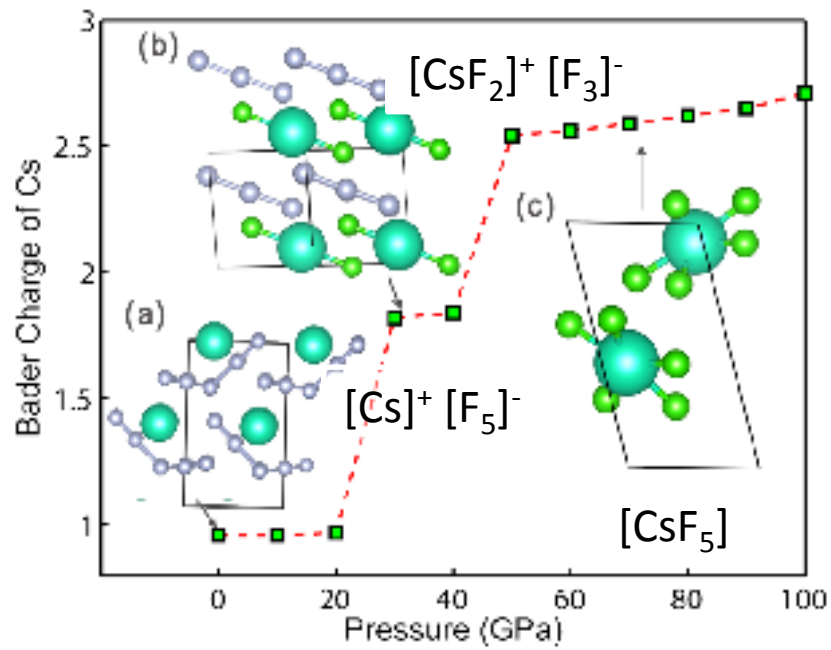


Our results



Can Cs be oxidized further?
Yes. Cs^{+}, Cs^{3+}, Cs^{5+}

But, polyfluoride anions also contributes!
$[F]^{-}$ $[F_3]^{-}$ $[F_5]^{-}$



arXiv:1408.5551

Applications

PAPER

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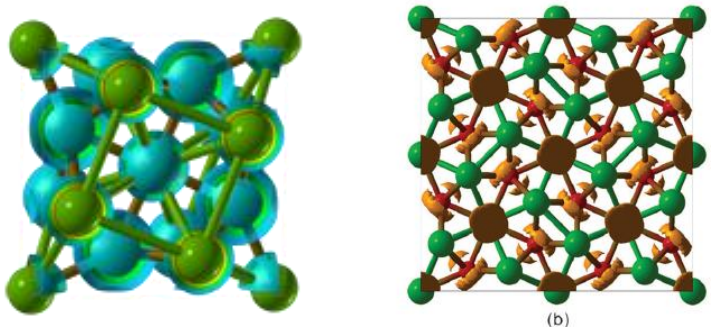
View Journal | View Issue

Novel stable compounds in the Mg–O system under high pressure†

Cite this: *Phys. Chem. Chem. Phys.*, 2013, **15**, 7696

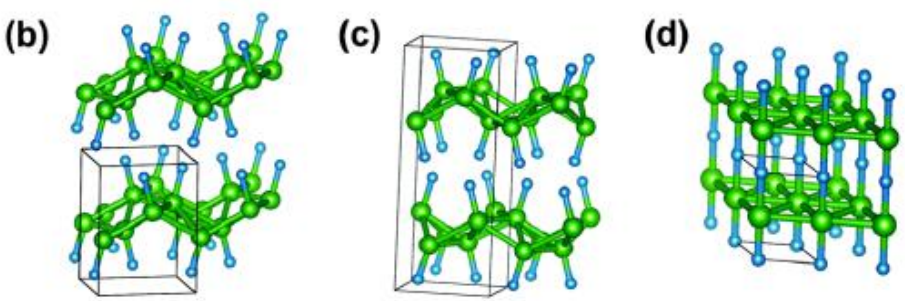
Qiang Zhu,*[a] Artem R. Oganov[ab] and Andriy O. Lyakhov[a]



0, 165504 (2013) PHYSICAL REVIEW LETTERS week 19 APl

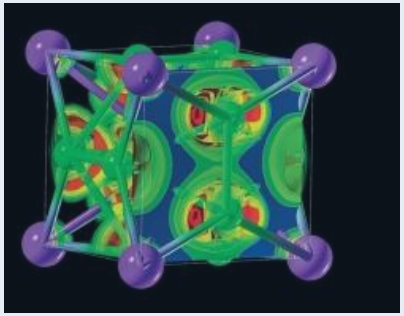
Pressure-Induced Stabilization and Insulator-Superconductor Transition of BH

Chao-Hao Hu,[1,2,*] Artem R. Oganov,[2,3] Qiang Zhu,[2] Guang-Rui Qian,[2] Gilles Frapper,[4] Andriy O. Lyakhov,[2] and Huai-Ying Zhou[1]



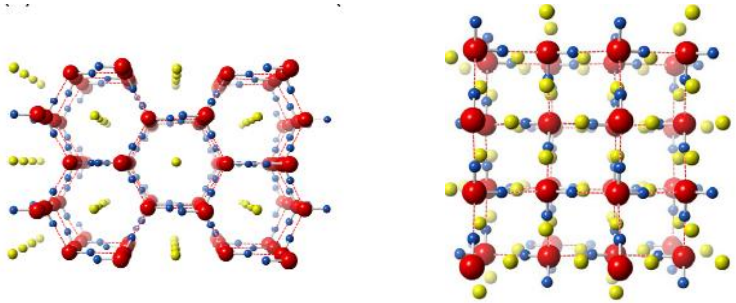
Unexpected Stable Stoichiometries of Sodium Chlorides

Weiwei Zhang,[1,2*†] Artem R. Oganov,[2,3,4*†] Alexander F. Goncharov,[5,6] Qiang Zhu,[2] Salah Eddine Boulfelfel,[2] Andriy O. Lyakhov,[2] Elissaios Stavrou,[5] Maddury Somayazulu,[5] Vitali B. Prakapenka,[7] Zuzana Konôpková[8]

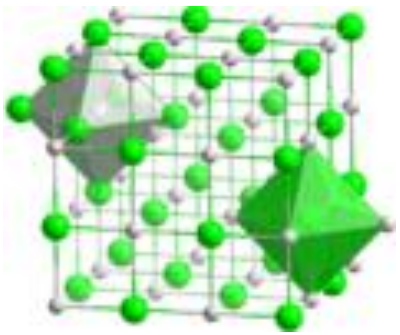


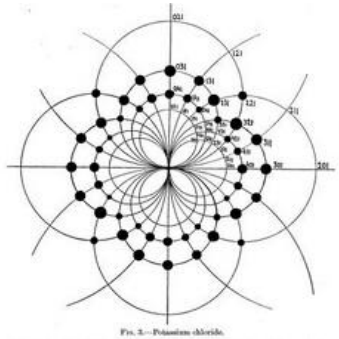
Novel Hydrogen Hydrate Structures under Pressure

Guang-Rui Qian[1], Andriy O. Lyakhov[1], Qiang Zhu[1], Artem R. Oganov[2,3,4] & Xiao Dong[1,5]

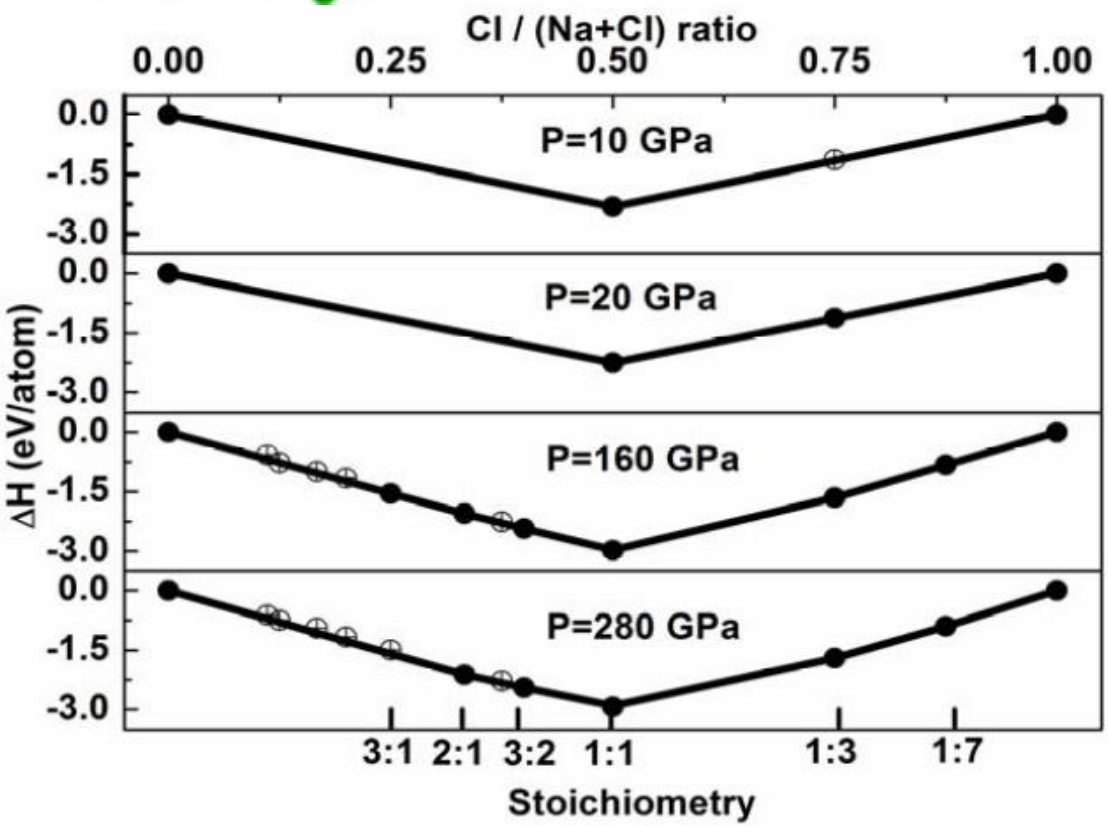


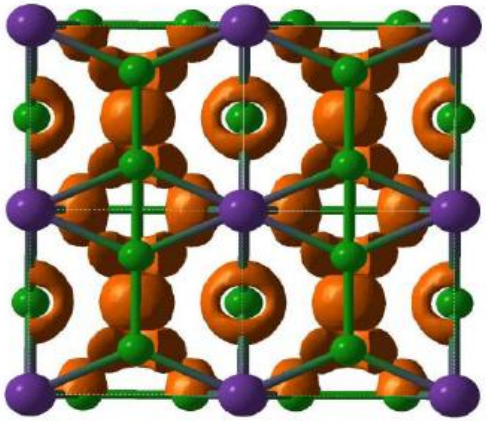
Applications





Diffraction of NaCl by Brag, **1913**





$NaCl_3$, stable from ~20 GPa

Zhang, et al, Science, 2013

I. Variable compositional prediction

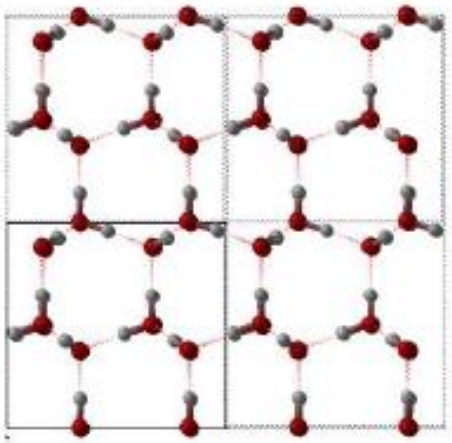
II. Constrained global optimization

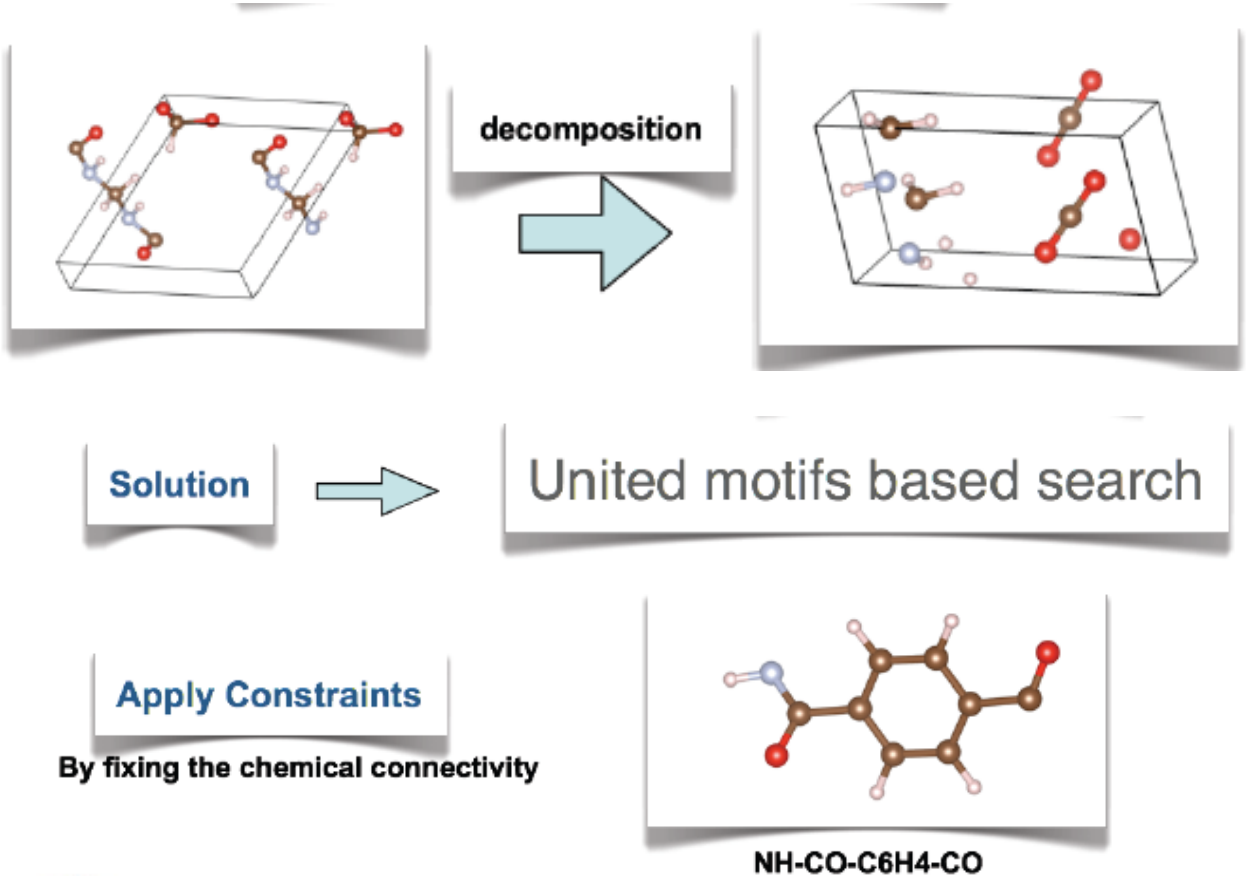
III. Low dimensional systems

Predicting crystals from motifs

Crystal structures are based on the packing of well-defined motifs

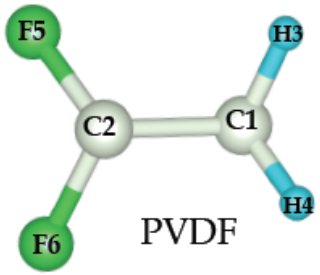
- Molecular crystals
- Polymers
- Inorganic crystals





Applying Constraints

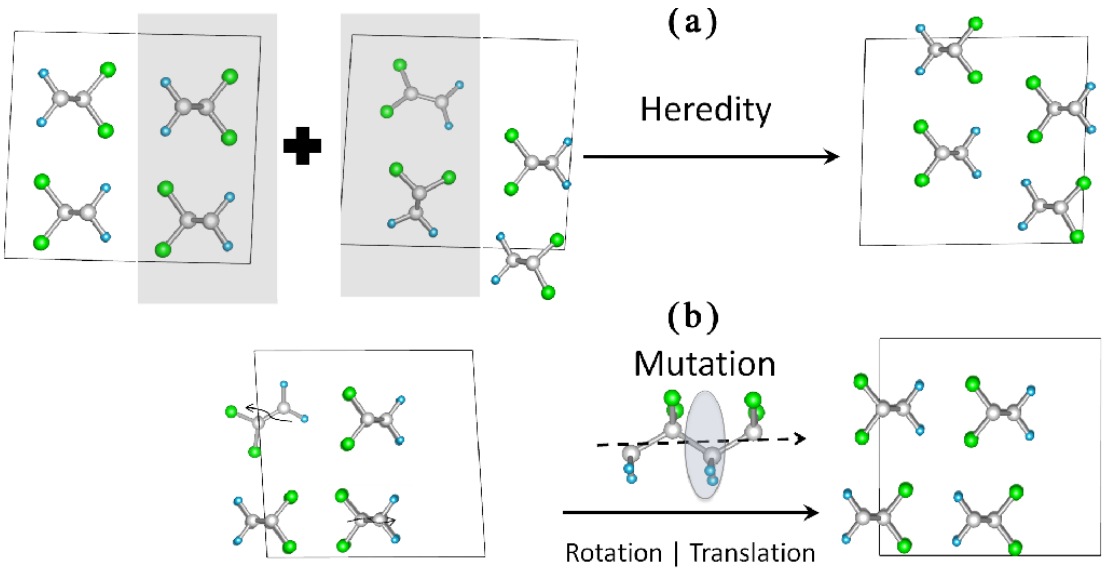
C	4.8172	5.1613	1.3406	0	0	0
C	4.0860	4.7547	0.0594	1	0	0
H	5.8261	4.7100	1.3406	1	2	0
H	4.9782	6.2554	1.3387	1	2	3
F	3.0027	5.5565	0.0590	2	1	3
F	3.9390	3.4763	0.0617	2	1	3
	Cartesian coordinate			*i*	*j*	*k*



	R_{j0}	A_{ji0}	T_{kji0}
C	*	*	*
C	1.5111	*	*
H	1.0899	1.9075	*
H	1.0994	1.8951	-2.0315
F	1.3940	1.8797	3.1124
F	1.3874	1.9366	-1.1880

Z-matrix

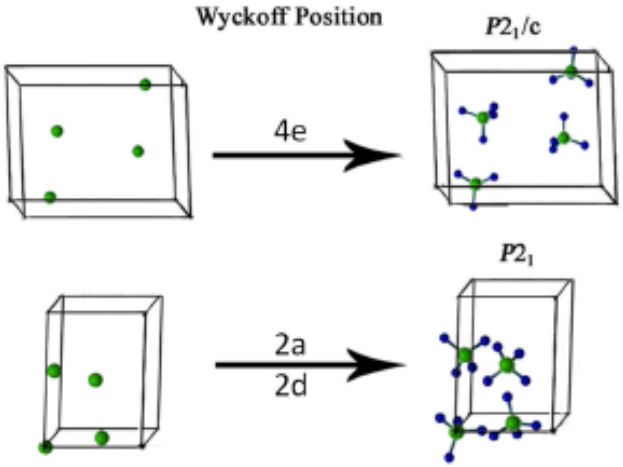
Internal coordinates



1. Build the Wyckoff positions
2. Track the symmetry operations (point group + translation)
3. Construct the whole molecule

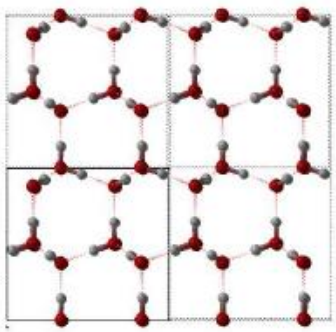
Space group: 14

'oint group	Translation
2/m	
(x, y, z)	(0, 0, 0)
(-x,y,-z)	(0,1/2,1/2)
(-x,-y,-z)	(0, 0, 0)
(x,-y, z)	(0,1/2,1/2)

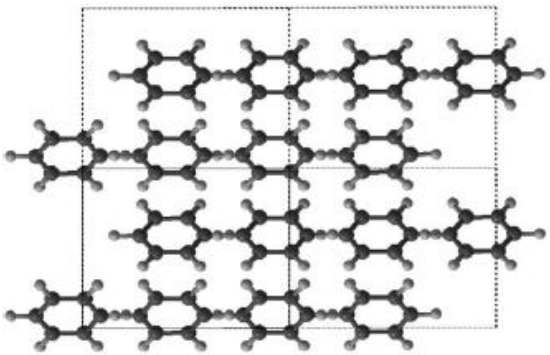


Applications: Molecular Crystals

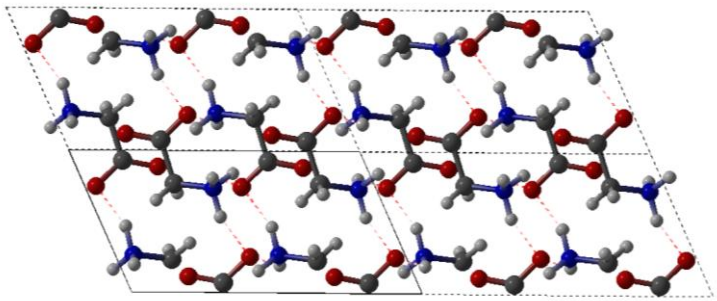
H_2O



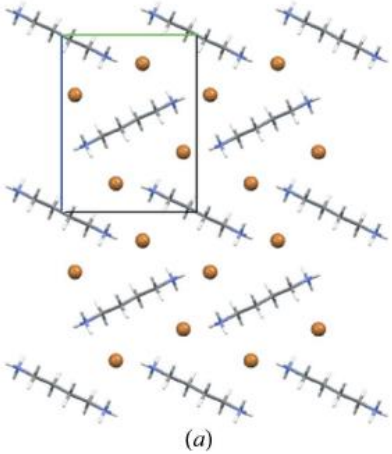
benzene



glycine

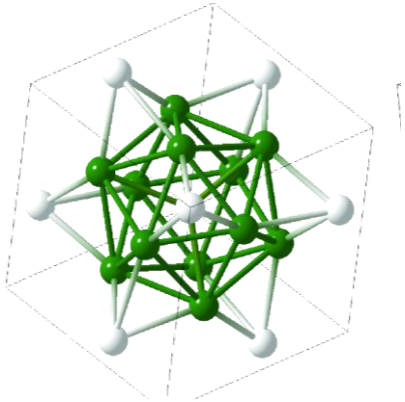


Butane-1,4-diammonium dibromide

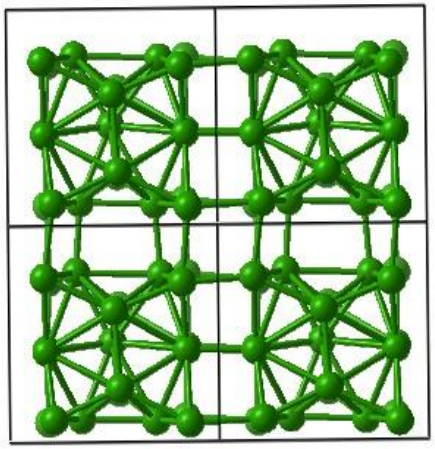


Zhu, et al, Acta Cryst B, 2012

Long term challenge
high pressure phase of Methane



(111) view



2*2*2 (100) view

Applications: Inorganic Crystals

PRL **109**, 245503 (2012) PHYSICAL REVIEW LETTERS week ending 14 DECEMBER 2012

First-Principles Determination of the Structure of Magnesium Borohydride

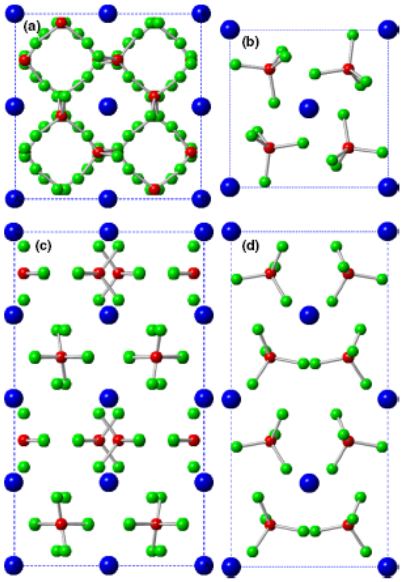
Xiang-Feng Zhou,[1,2,*] Artem R. Oganov,[2,3] Guang-Rui Qian,[2] and Qiang Zhu[2]

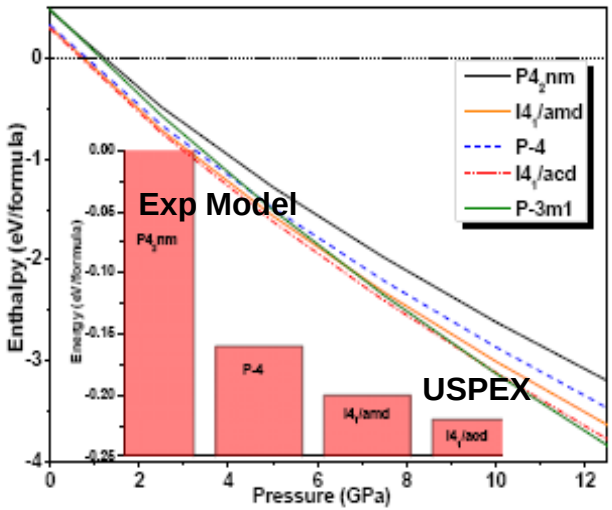
[1]*School of Physics and Key Laboratory of Weak-Light Nonlinear Photonics, Nankai University, Tianjin 300071, China*

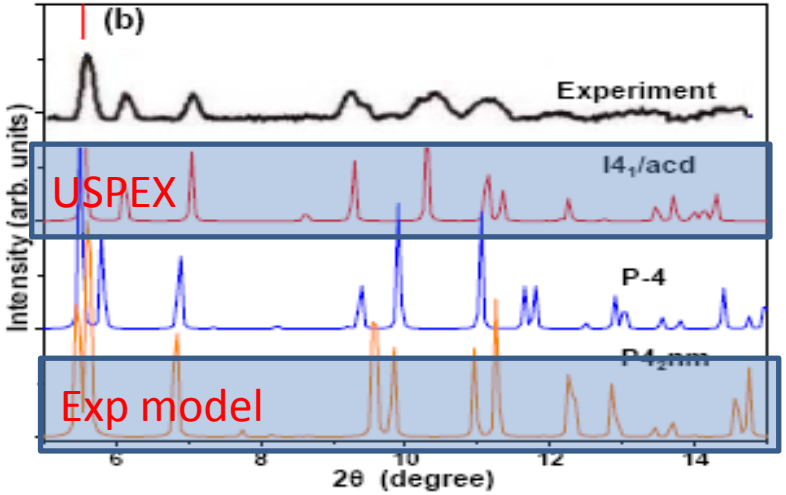
[2]*Department of Geosciences, Department of Physics and Astronomy, Stony Brook University, Stony Brook, New York 11794, USA*

[3]*Geology Department, Moscow State University, Moscow 119992, Russia*

(Received 2 October 2012; published 10 December 2012)

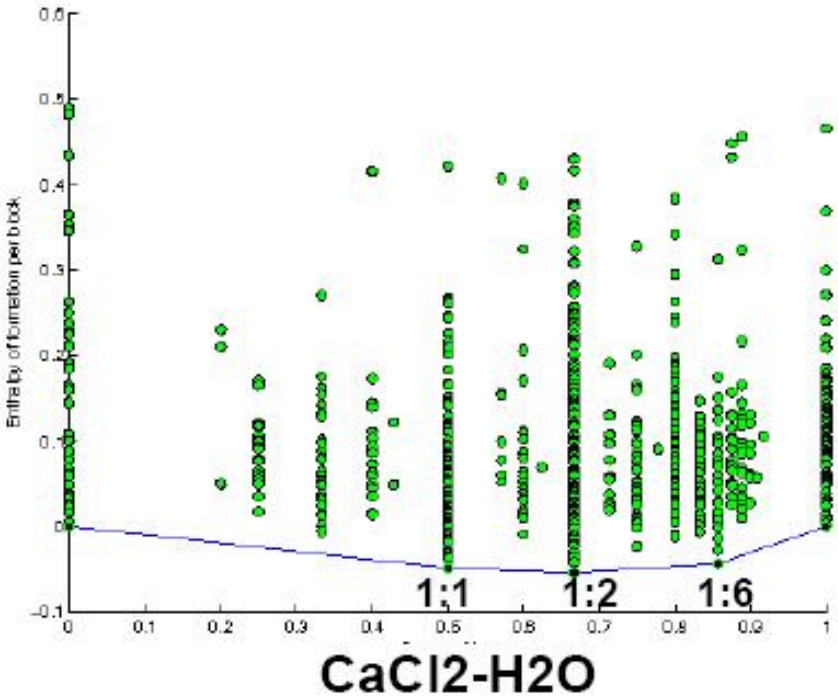


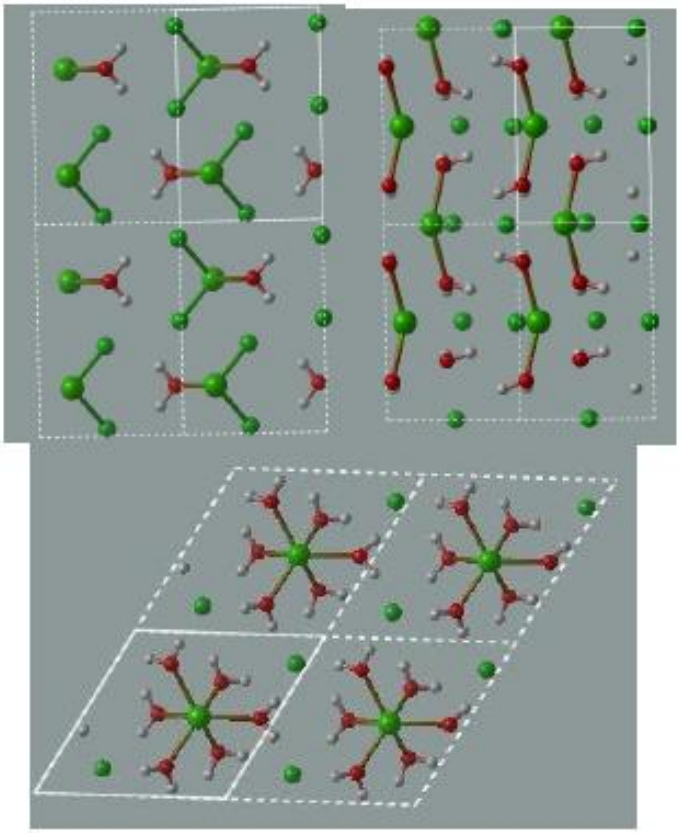




Applications: Hydrates/Co-crystals

block 1: Ca + 2Cl
block 2 : H_2O





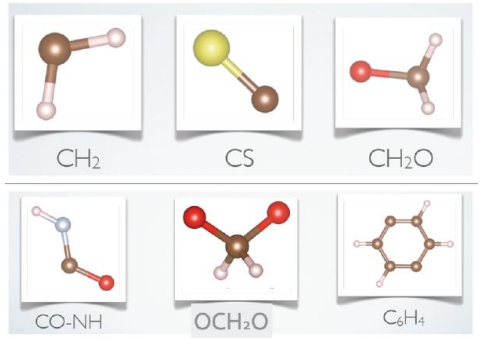
Variable composition + Molecular crystal structure prediction

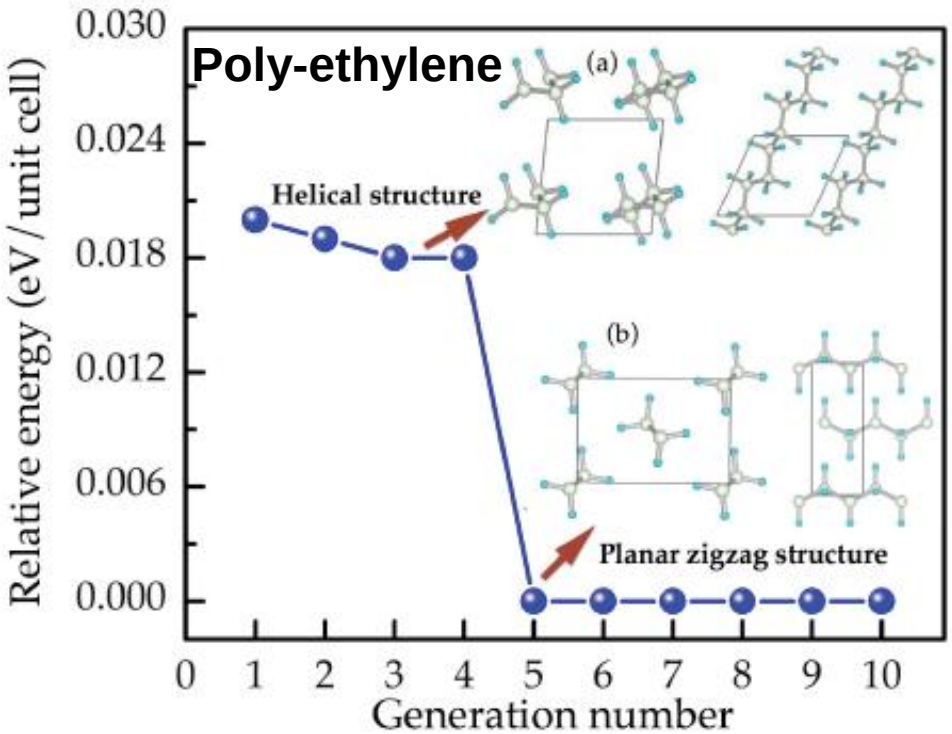
Zhu, unpublished

Applications: Polymers

Motif-based search

- Bond connectivity





Polymer	Method	a(Å)	b(Å)	c(Å)	β(°)	Density (g/cm^3)
PE	Expt.[34]	7.12	4.85	2.55		0.997
$[-CH_2-]_n$	PBE[31]	8.20	5.60	2.55		0.796
($Pnma$)	PBE-TS[31]	7.01	4.76	2.56		1.091
	PBE-TS*	7.02	4.76	2.56		1.091
PA	Expt.[34]	4.24	7.32	2.46	91-94	1.130
$[-C_2H_2-]_n$	PBE[31]	5.00	7.74	2.46	90.3	0.908
($P2_1/n$)	PBE-TS[31]	4.01	7.19	2.46	90.3	1.22
	PBE-TS*	4.00	7.22	2.46	90.6	1.191
PGA	Expt.[34]	5.22	6.19	7.02		1.700
$[-C_2H_2O_2-]_n$	PBE[31]	5.07	5.58	6.96		1.958
($Pcmn$)	PBE-TS[31]	5.09	6.11	7.03		1.763
	PBE-TS*	5.13	6.09	7.01		1.79
PVC	Expt.[34]	10.24	5.24	5.08		1.523
$[-CH_2-CHCl-]_n$	PBE[31]	10.45	5.50	5.05		1.430
($Pbcm$)	PBE-TS[31]	10.11	5.15	5.08		1.540
	PBE-TS*	10.14	5.16	5.08		1.530
POM	Expt.[34,38]	4.77	7.65	3.56		0.922
$[-CH_2-O-]_n$	PBE[31]	5.40	8.37	3.63		0.730
($P2_12_12_1$)	PBE-TS[31]	4.59	7.72	3.57		0.947
	PBE-TS*	4.55	7.75	3.59		0.95
PPO	Expt.[34]	8.07	5.54	9.72		1.408
$[-C_6H_4O-]_n$	PBE[31]	8.42	5.88	9.85		1.254
($Pbcn$)	PBE-TS[31]	8.04	5.37	9.75		1.453
	PBE-TS*	8.02	5.36	9.75		1.491
PPS	Expt.[34]	8.67	5.61	10.26		1.440
$[-C_6H_4S-]_n$	PBE[31]	8.85	5.73	10.26		1.381
($Pbcn$)	PBE-TS[31]	8.48	5.54	10.25		1.492
	PBE-TS*	8.48	5.53	10.26		1.491

* This work.

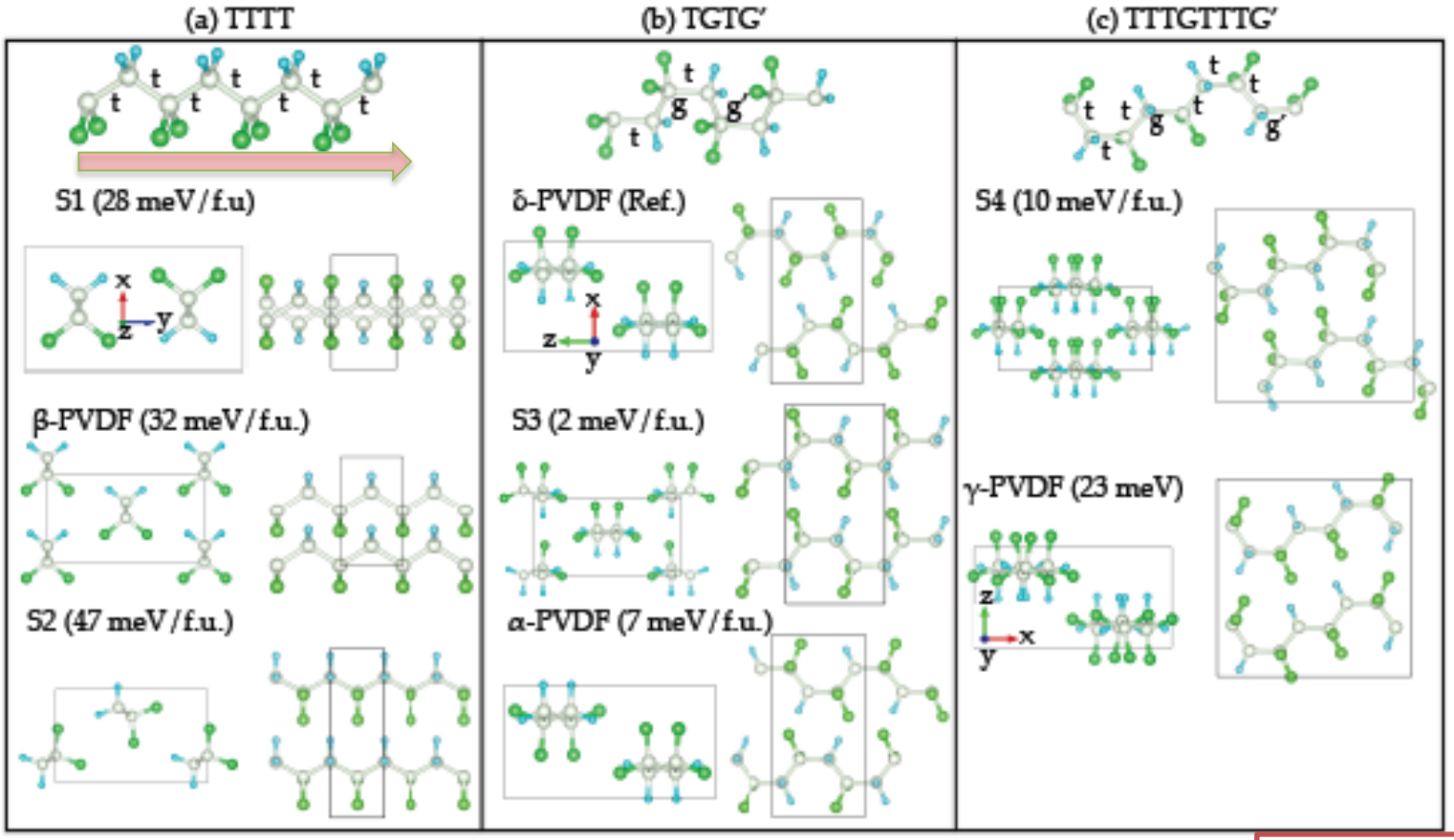
Zhu, J Chem. Phys., 2014

Applications: Polymers

Motif-based search

- ❖ Bond connectivity
- ❖ Orientation

PVDF



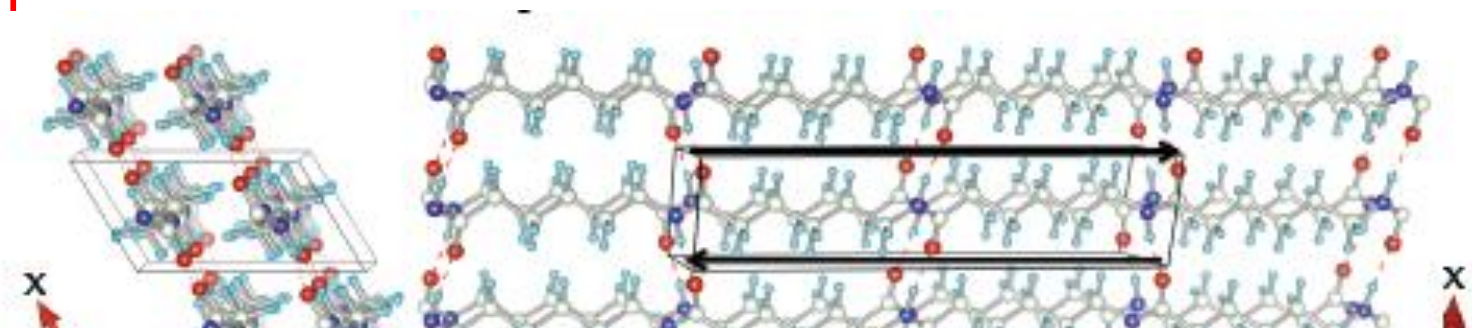
Zhu, J Chem. Phys., 2014

Applications: Polymers

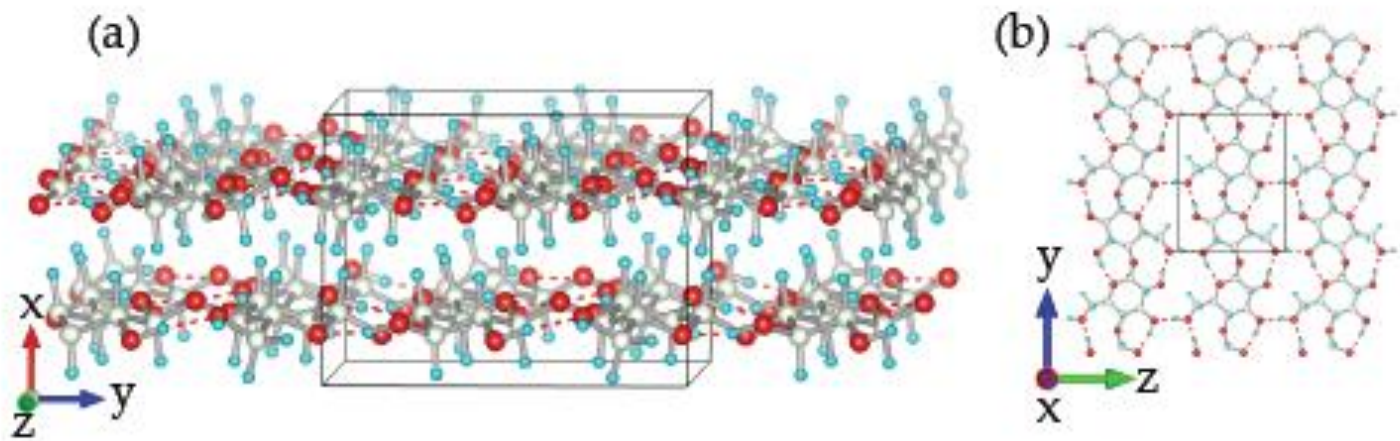
Motif-based search

- ❖ Bond connectivity
- ❖ Orientation

Nylon-6



Cellulose



Zhu, J Chem. Phys., 2014

Applications: Polymers

ARTICLE

Received 20 May 2014 | Accepted 29 Jul 2014 | Published 17 Sep 2014

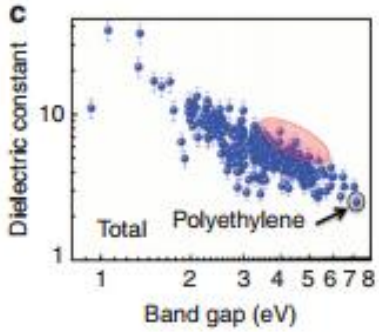
DOI: 10.1038/ncomms5845

Rational design of all organic polymer dielectrics

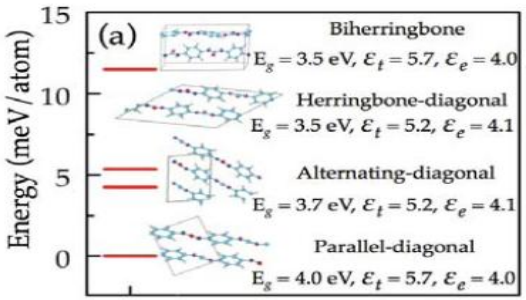
Vinit Sharma[1,2], Chenchen Wang[1,2], Robert G. Lorenzini[2,3], Rui Ma[2,3], Qiang Zhu[4], Daniel W. Sinkovits[5],

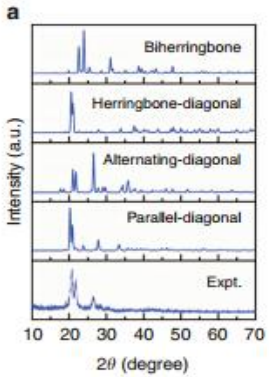
1, Chemical Exploration

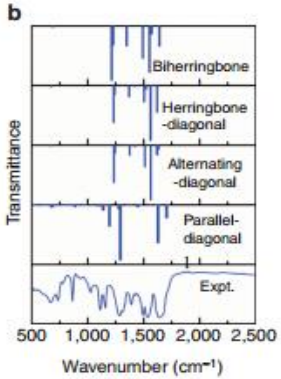
System repeat unit
NH-CO-NH-C_6H_4[56]
CO-NH-CO-C_6H_4
NH-CS-NH-C_6H_4[56]
NH-C_6H_4-C_6H_4-C_6H_4
CO-C_6H_4-CO-O
C_6H_4-C_6H_4-C_6H_4-O
CH_2-C_6H_4-C_6H_4-O
CH_2-CO-C_6H_4-O[57]
CH_2-C_6H_6-CO-O
CH_2-NH-CO-NH[56]
CH_2-NH-CS-NH[56]
CH_2-C_6H_4-CH_2-O

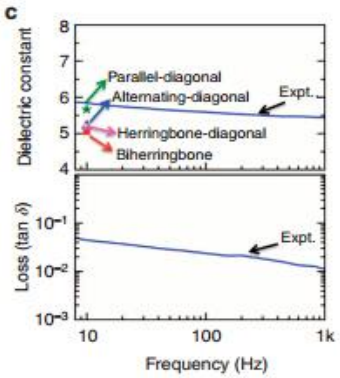


2, Structure (Property) Prediction:









3, Synthesis &Characterization

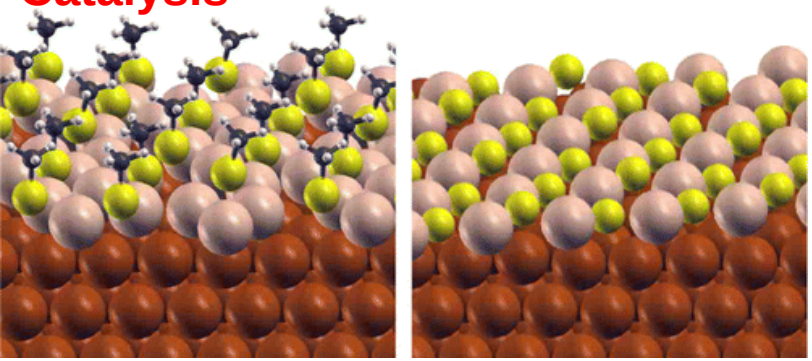
I. Variable compositional prediction

II. Constrained global optimization

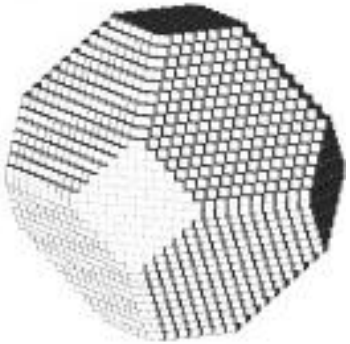
III. Low dimensional systems

Low dimensional Systems: Surfaces

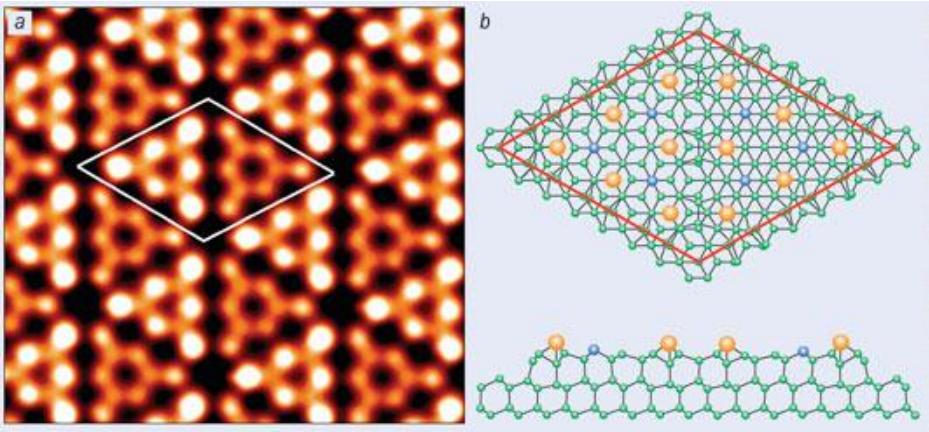
Catalysis



Morphology



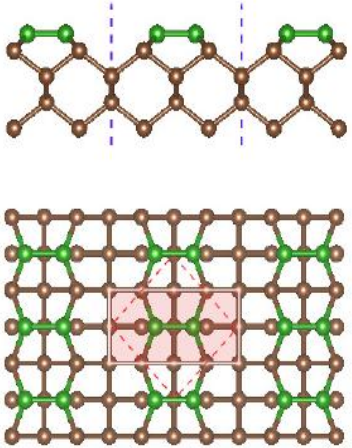
Experiment: High resolution STM



Si(111)-7x7 reconstruction

Wang, PRB, 2004

Prediction by Evolutionary algorithm

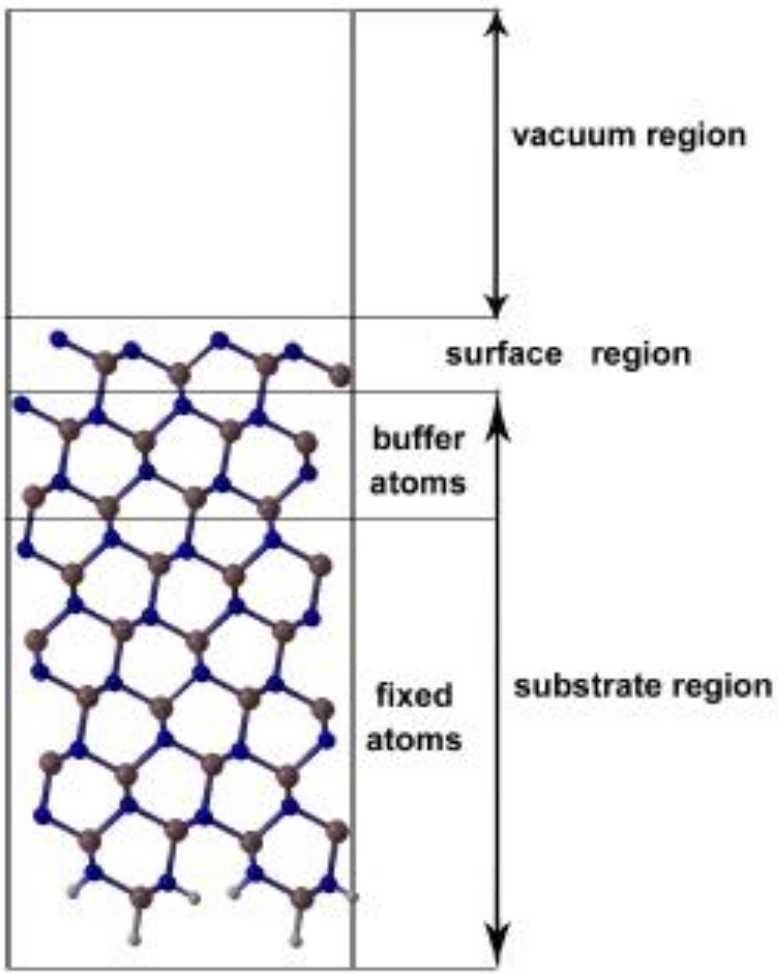


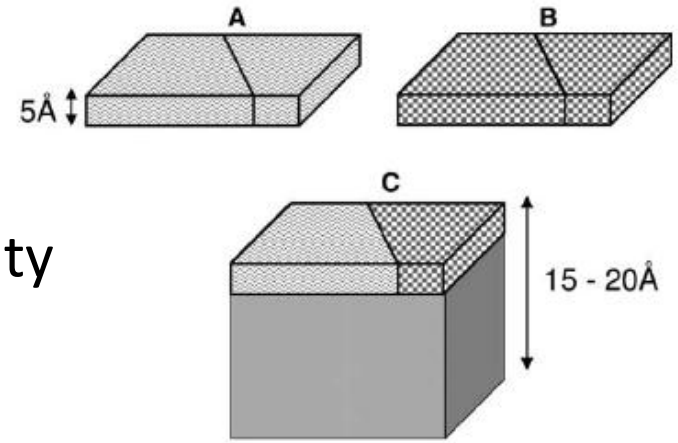
Diamond (100) -2x1

Frauenheim, PRB, 1993

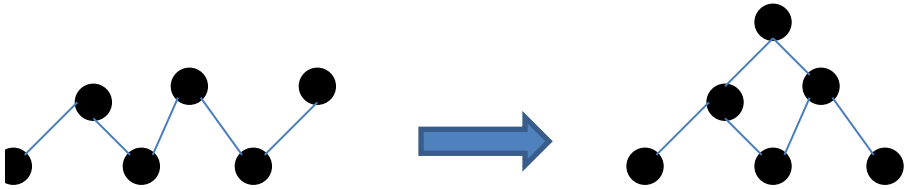
Zhu, Phys. Rev. B, 2013

EA in Predicting Surfaces

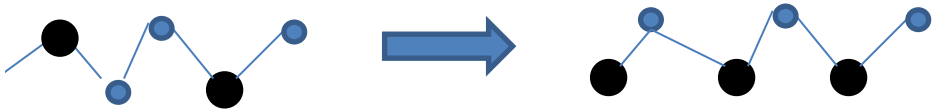




1. Heredity



2. Mutation

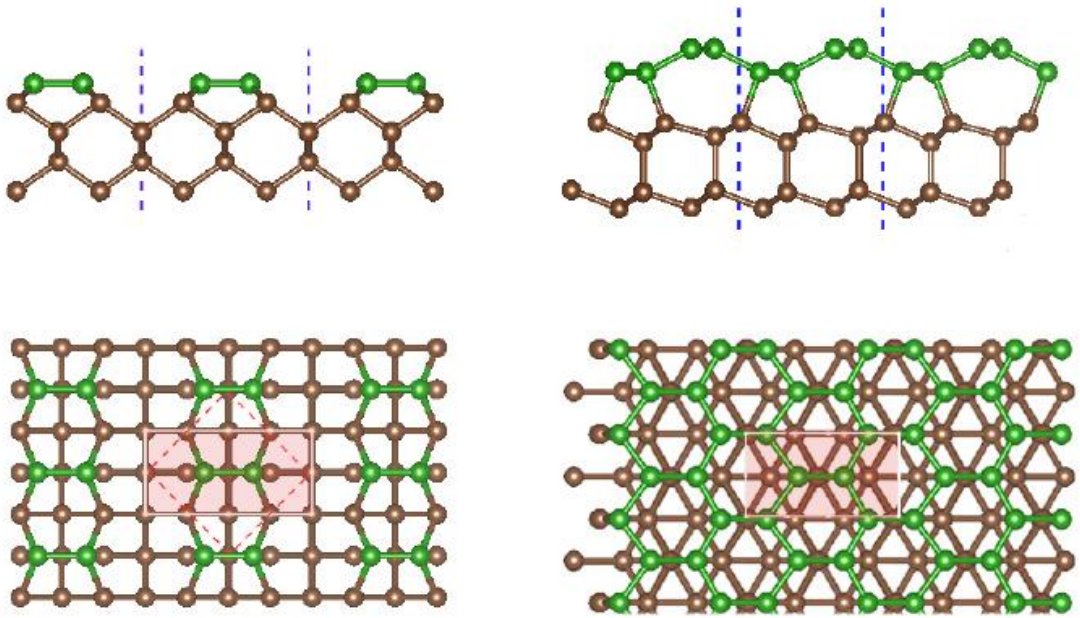


3. Permutation

Main features

Simple input information

- surface atom
- substrate



Diamond (100) 2*1 **Diamond (111) 2*1**

Main features

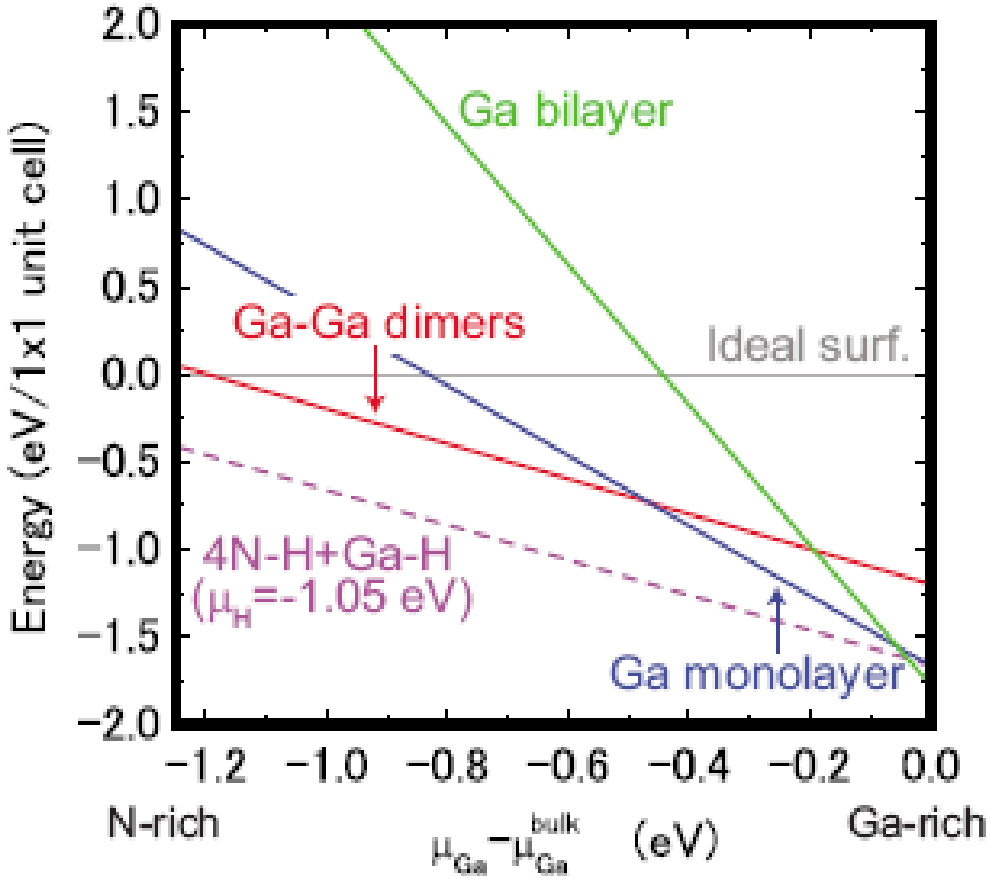
Simple input information

- surface atom
- substrate

More intelligent ?

- Variable number of surface atoms
- Variable reconstruction cell (from 1*1 to N*N)

Predicting Reconstructions with Variable Stoichiometry



A typical phase diagram of GaN (1011) surfaces. Akiyama, Phys. Rev. B, 2010

Surface stability is function of chemical potential

Surfaces with variable stoichiometry

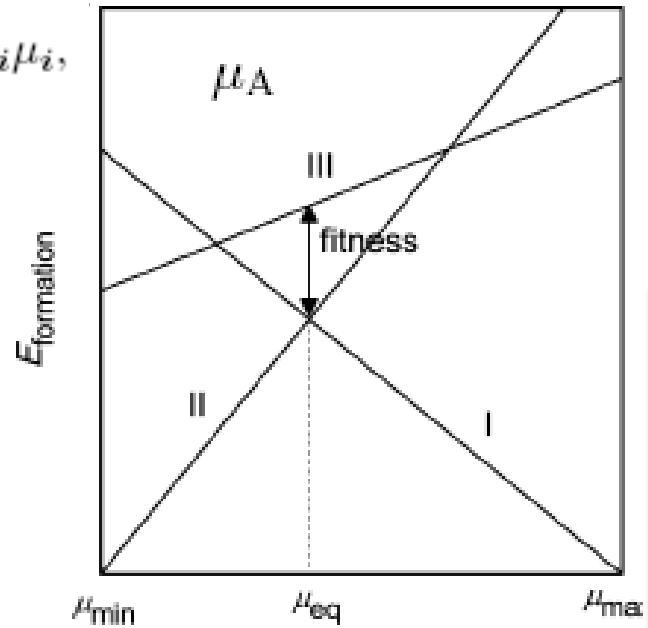
Surface energy

$$E_{\text{formation}} = E_{\text{total}} - E_{\text{ref}} - \sum_i n_i \mu_i,$$

$$\mu_{\text{A}} + \mu_{\text{B}} = E_{\text{AB}}.$$

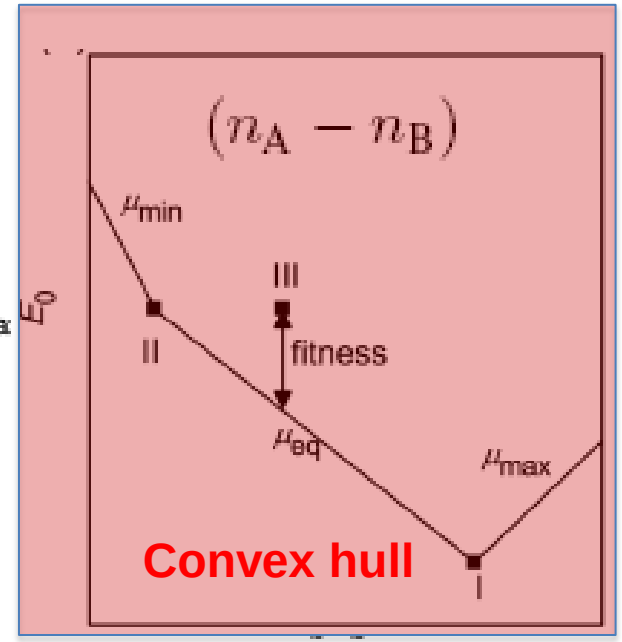
$$\mu_{\text{A}} \leq E_{\text{A}}.$$

$$\mu_{\text{B}} \leq E_{\text{B}}.$$



$$E_{\text{formation}} = E_{\text{tot}} - E_{\text{ref}} - n_{\text{B}} E_{\text{AB}} - \mu_{\text{A}}(n_{\text{A}} - n_{\text{B}})$$

$$E_{\text{AB}} - E_{\text{B}} \leq \mu_{\text{A}} \leq E_{\text{A}}$$



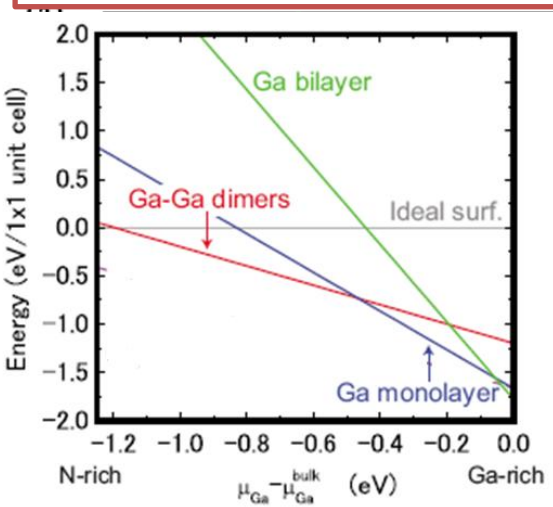
Zhu et al, Phys. Rev. B, 2013

Surfaces with variable stoichiometry

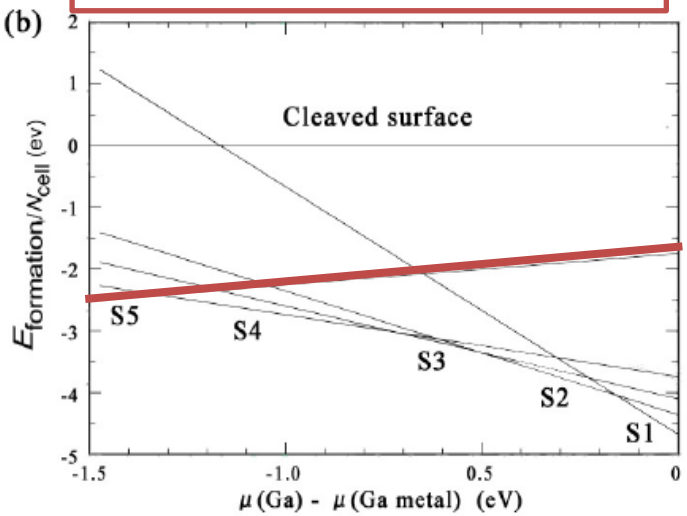
GaN: (10$\bar{1}$1)

- Variable cell
- Variable stoichiometry

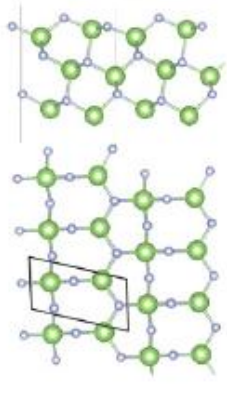
Chemical Intuition
Akiyama, Phys. Rev. B, 2010



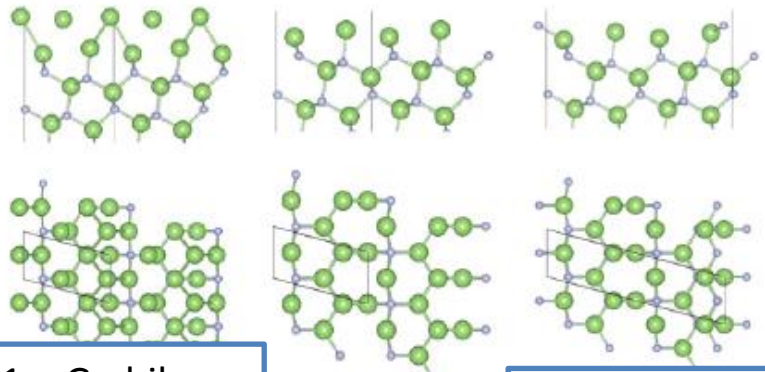
Automated Search
Zhu et al, Phys. Rev. B, 2013



Cleaved surface



S2: Ga-monolayer

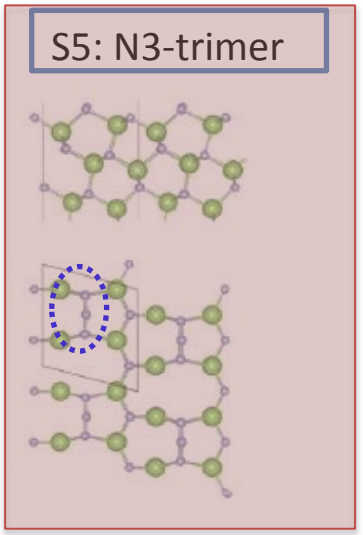


S1: Ga-bilayer

S4: 0.5 N-Vacancy

S3: 1N-Vacancy

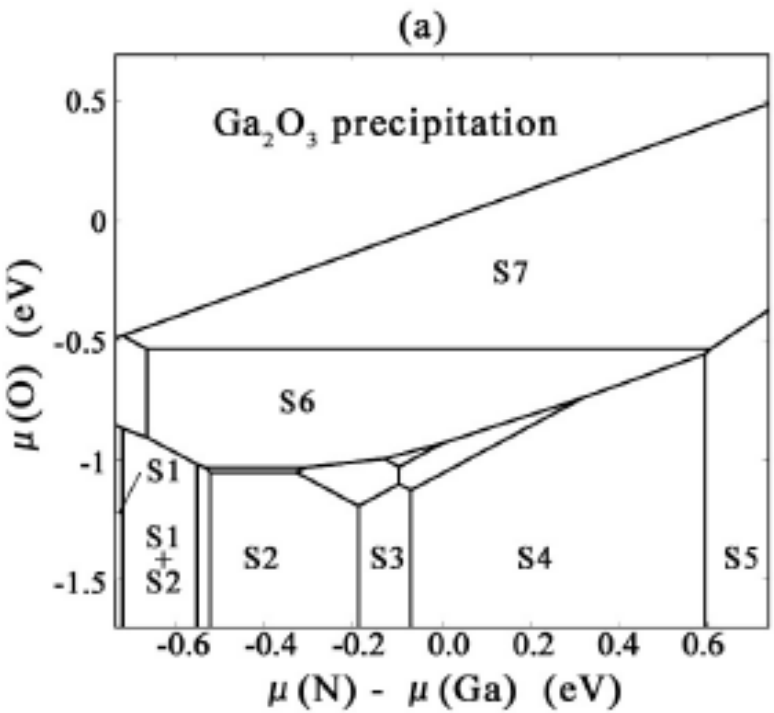
S5: N3-trimer

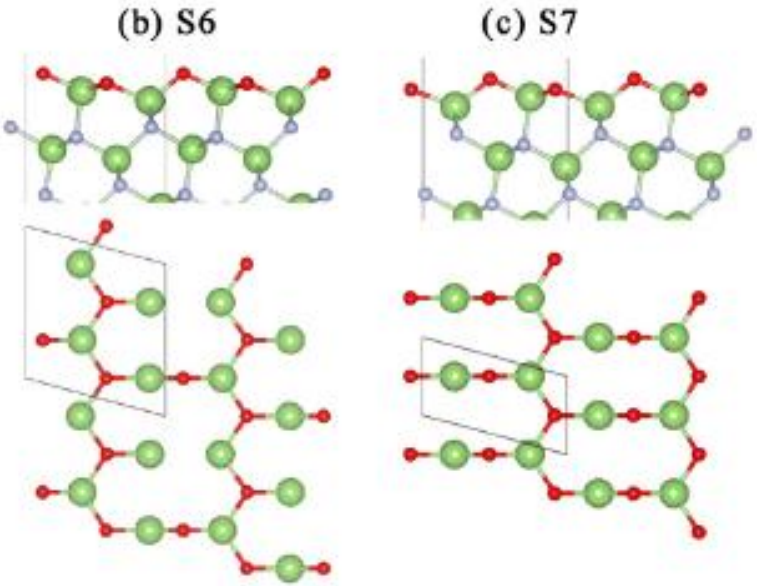


Surfaces with variable stoichiometry

GaN-O: (10$\bar{1}$1)

- Variable cell
- Variable stoichiometry





Zhu, Phys. Rev. B, 2013

USPEX: Computational Materials Design

Structure Prediction

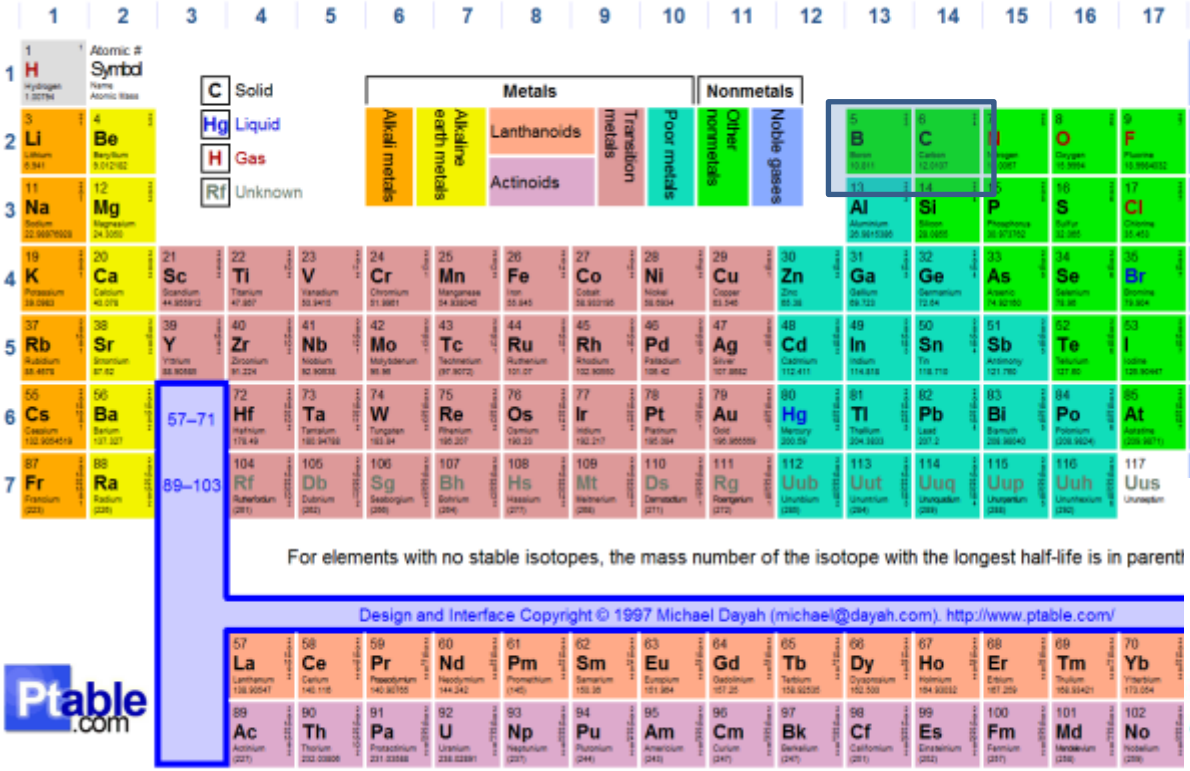
System

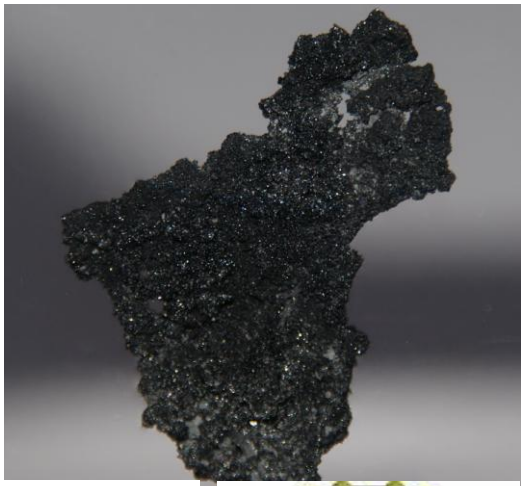
- **Dimension**
  - 0: Nano-particle;
  - 1: polymers;
  - 2: surfaces/2D crystals;
  - 3: Bulk
- **Stoichiometry**

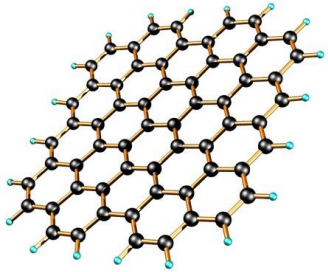
  0: fixed; 1: variable
- **Building block**

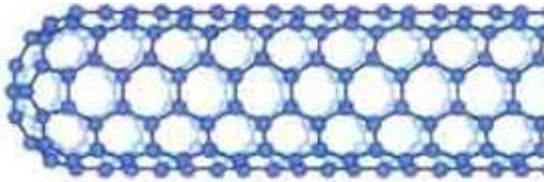
  0: atom; 1: molecule

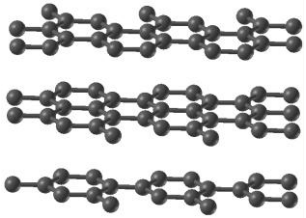
What can USPEX do?

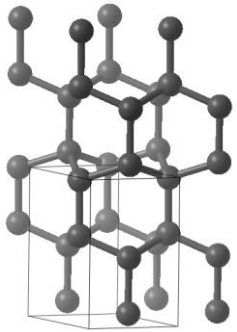


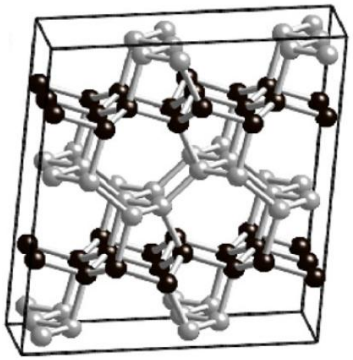












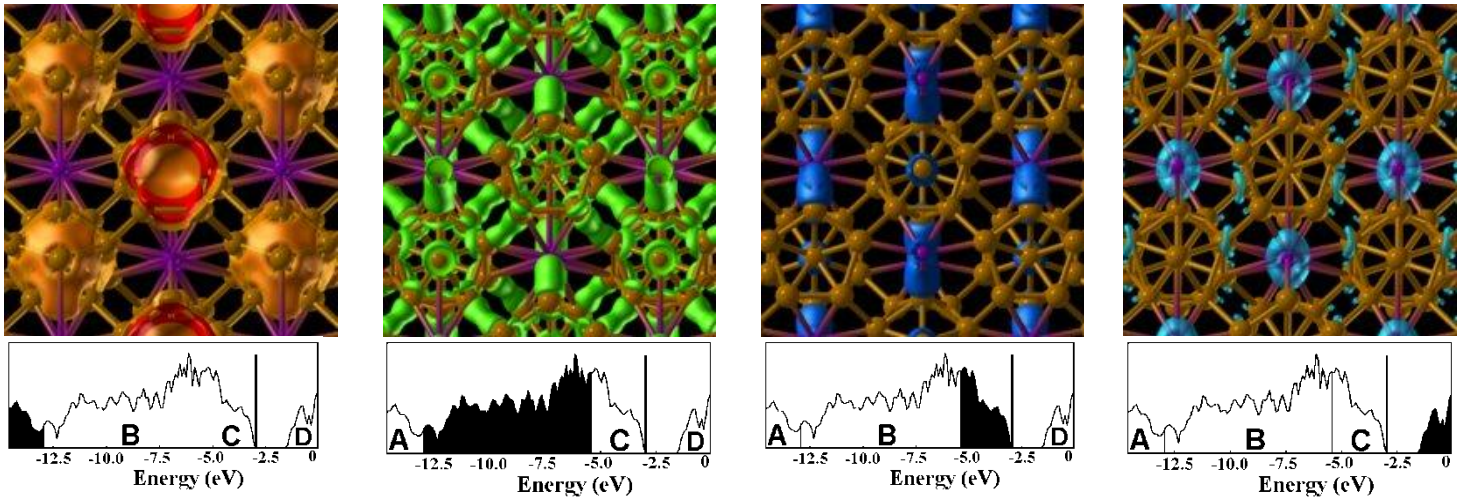
Vol 457 | 12 February 2009 | doi:10.1038/nature07736 nature

B: Crystal

LETTERS

Ionic high-pressure form of elemental boron

Artem R. Oganov[1,2]†, Jiuhua Chen[3,4], Carlo Gatti[5], Yanzhang Ma[6], Yanming Ma[1,7], Colin W. Glass[1], Zhenxian Liu[8], Tony Yu[3], Oleksandr O. Kurakevych[9] & Vladimir L. Solozhenko[9]



Charge separation between B_{12}-icosahedra and B_2-pairs is clear from DOS (B12)(B2)

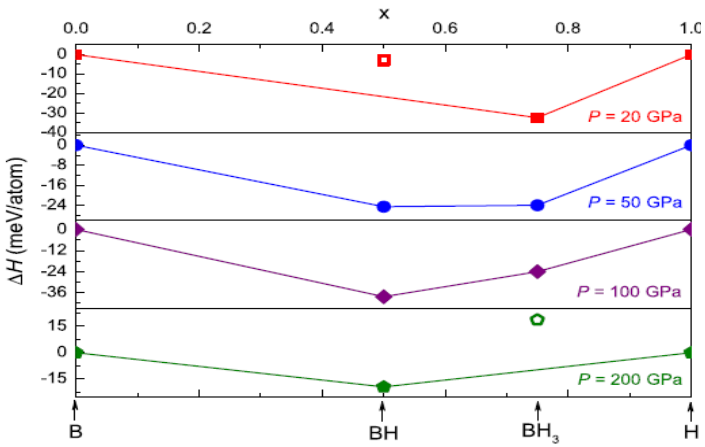
B: Compounds

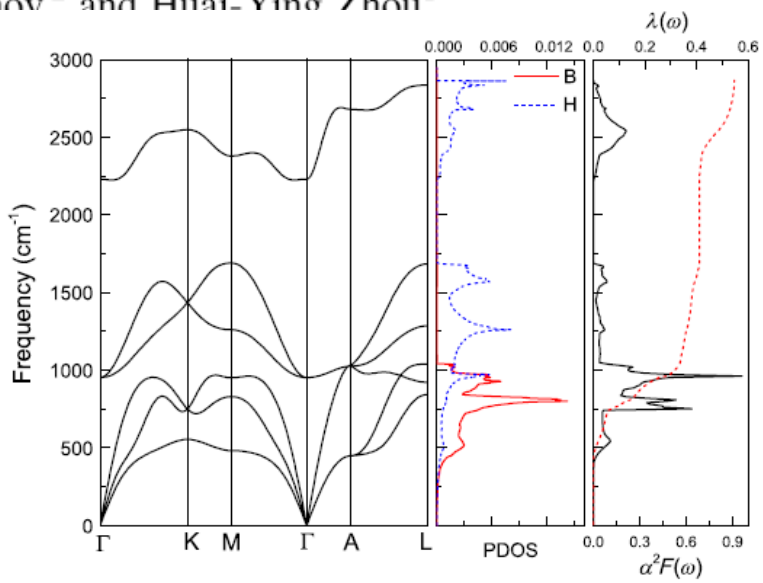
, 165504 (2013) PHYSICAL REVIEW LETTERS 19

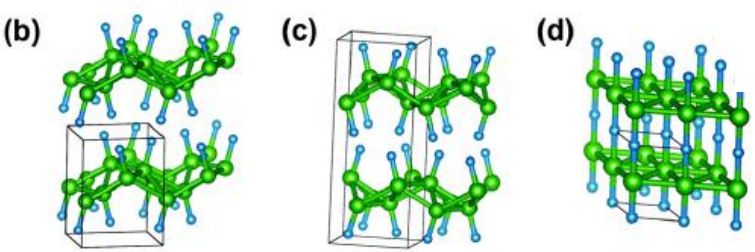
B-H

Pressure-Induced Stabilization and Insulator-Superconductor Transition of BH

Chao-Hao Hu,[1,2,*] Artem R. Oganov,[2,3] Qiang Zhu,[2] Guang-Rui Qian,[2] Gilles Frapper,[4] Andriy O. Lyakhov[2] and Huai-Ying Zhou[1]





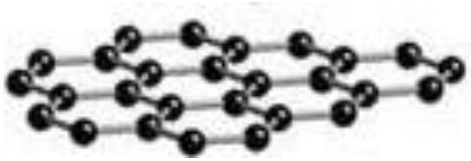


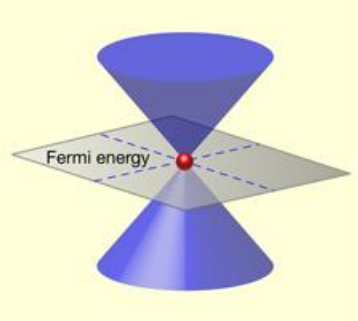
Boron compounds are well know for

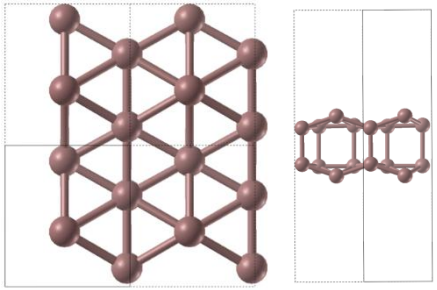
- **Superconductivity,**
- **Thermal ellectrics**
- **Mechanical properties**

B: 2D Crystal

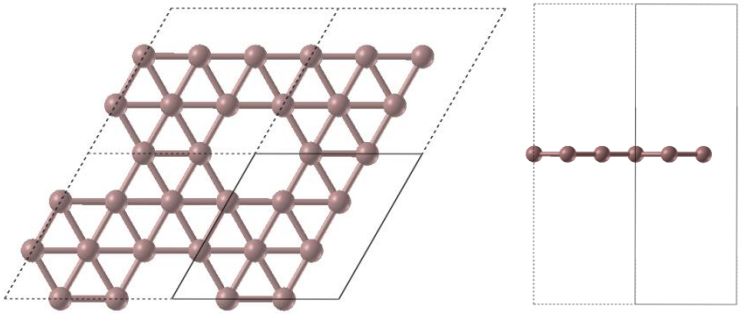
2007: Tang et al. (PRL) construct alpha-sheet.
2011: Luo et al. (JACS) confirm alpha-sheet.
2013: using USPEX, we find two MUCH more stable 2D-structures.



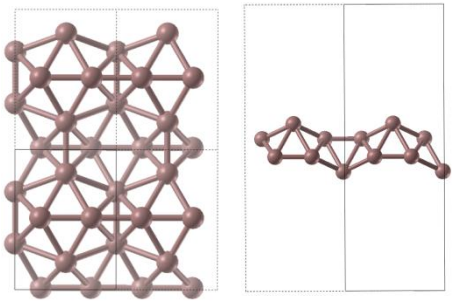




Structure 1,
60 meV/atom better



alpha-sheet



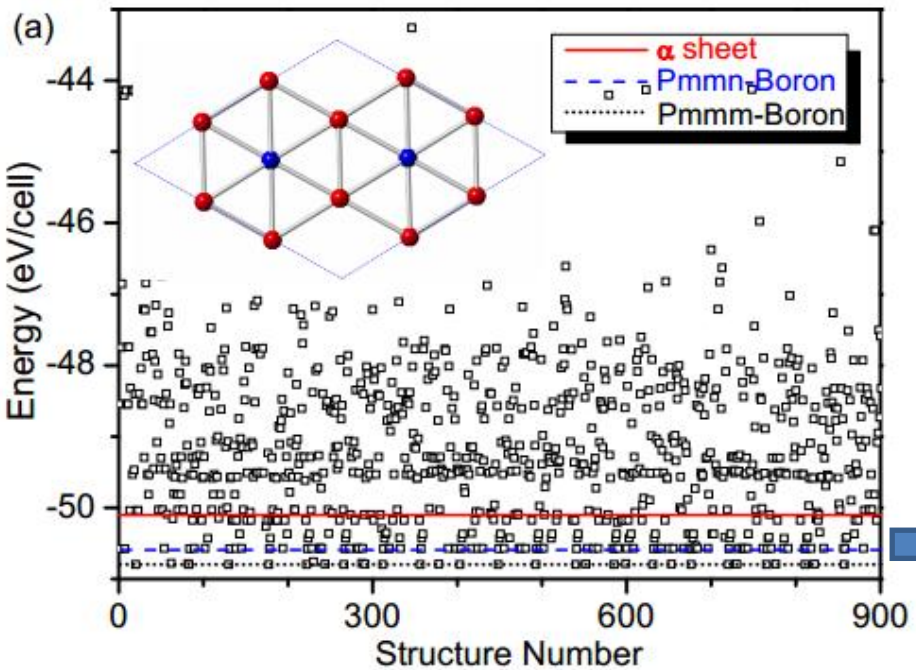
Structure 2,
80 meV/atom better

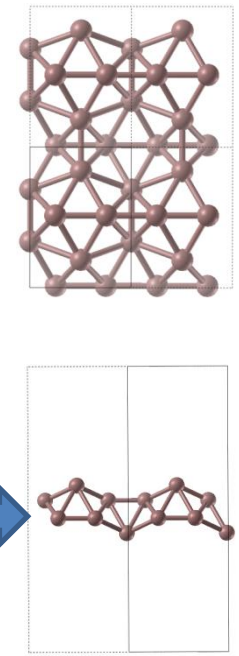
B: 2D Crystal

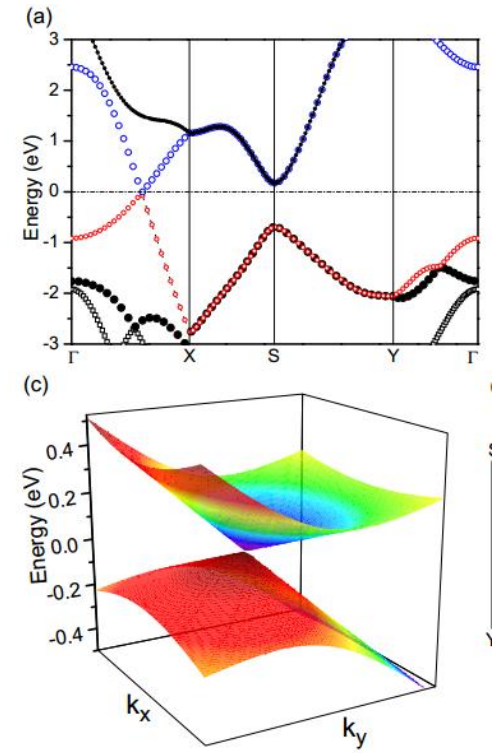
PRL **112,** 085502 (2014) PHYSICAL REVIEW LETTERS week ending 28 FEBRUARY 2014

Semimetallic Two-Dimensional Boron Allotrope with Massless Dirac Fermions

Xiang-Feng Zhou,[1,2,*] Xiao Dong,[1,2] Artem R. Oganov,[2,3,4] Qiang Zhu,[2] Yongjun Tian,[5] and Hui-Tian Wang[1,6]







B: Surface

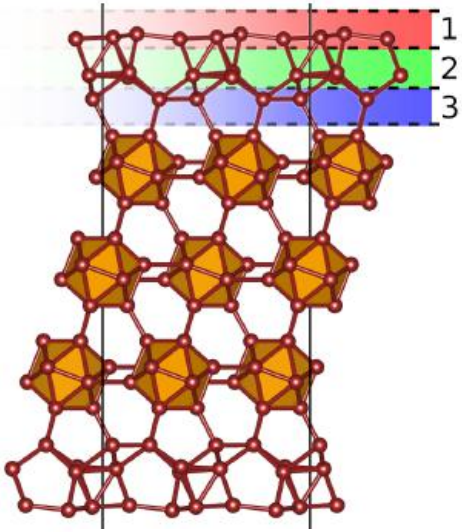
PRL **111,** 136101 (2013) PHYSICAL REVIEW LETTERS week ending 27 SEPTEMBER 2013

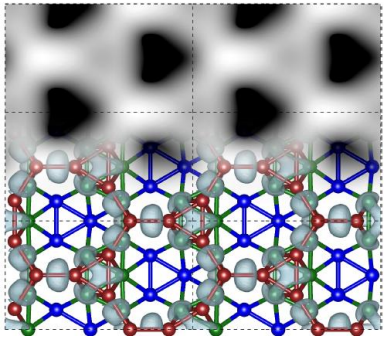
Conducting Boron Sheets Formed by the Reconstruction of the α-Boron (111) Surface

Maximilian Amsler,[1] Silvana Botti,[2] Miguel A. L. Marques,[2] and Stefan Goedecker[1,*]

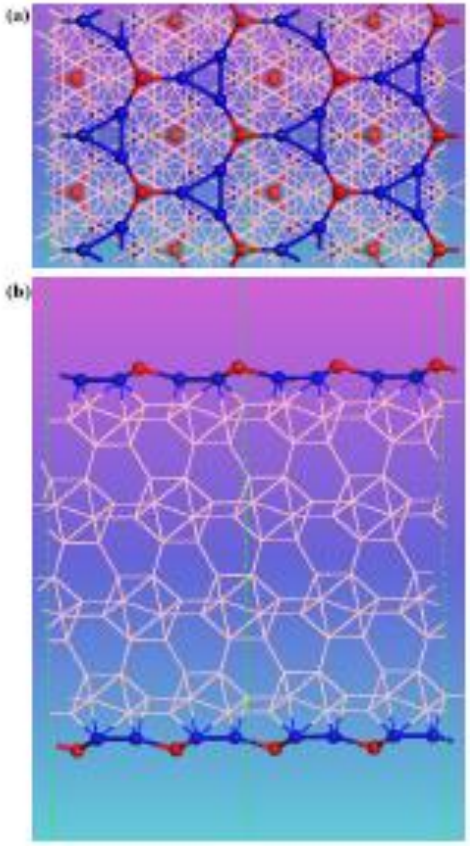
[1]*Department of Physics, Universität Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland*

[2]*Institut Lumière Matière, UMR5306 Université Lyon 1-CNRS, Université de Lyon, F-69622 Villeurbanne Cedex, France*





Minima Hopping



USPEX

TABLE I. The surface energies of the unreconstructed (111)-I, reconstructed (111)-$I_{R,(a)}$, and (111)-$I_{R,(z)}$ structures by using different functionals, in units of meV/Å^2.

Surface	(111)-I	(111)-$I_{R,(a)}$	(111)-$I_{R,(z)}$	Reference
PBE	219.29	170.64	128.23	This work
	218.80	170.61		26
HSE	248.43	197.39	136.79	This work
	247.50	196.31		26

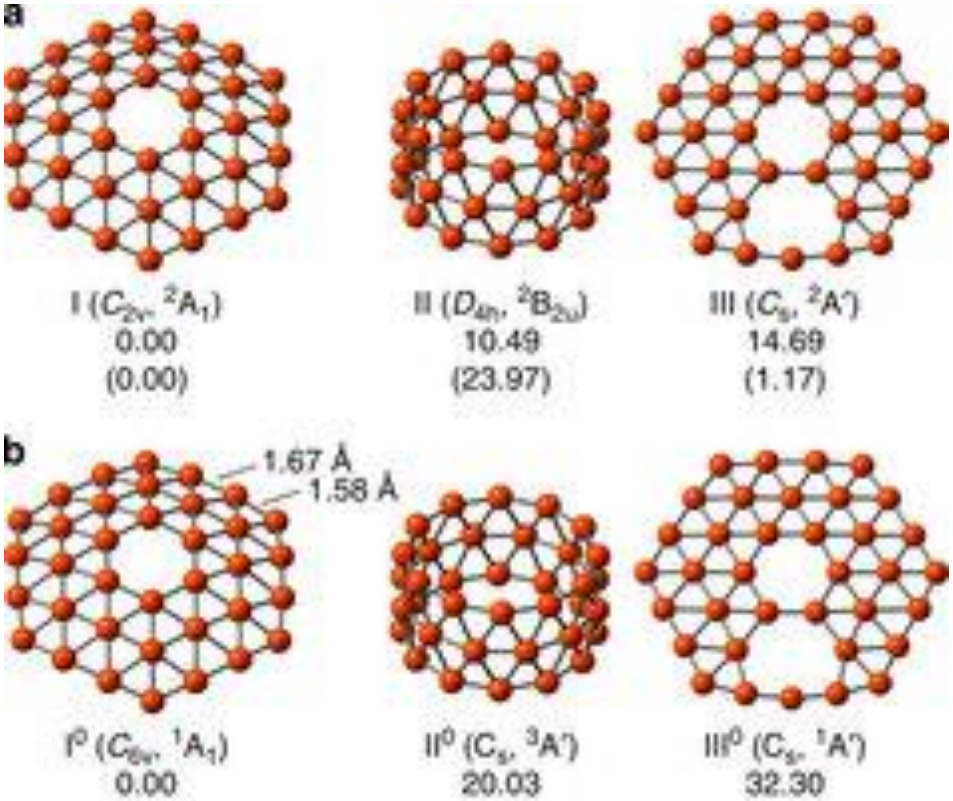
B: Nano clusters

NATURE COMMUNICATIONS | ARTICLE

Planar hexagonal B_{36} as a potential basis for extended single-atom layer boron sheets

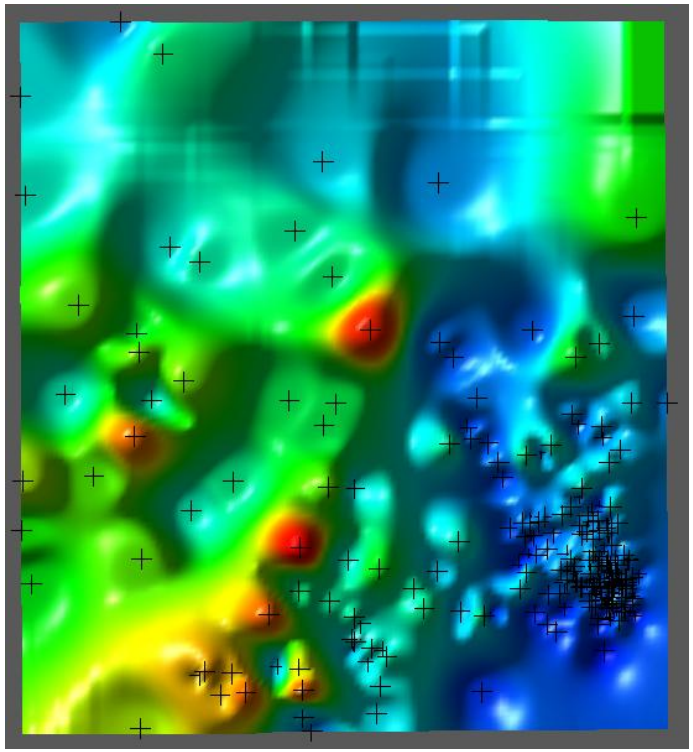
Zachary A. Piazza, Han-Shi Hu, Wei-Li Li, Ya-Fan Zhao, Jun Li & Lai-Sheng Wang

Affiliations | Contributions | Corresponding authors



OUTLOOK
Computational Materials Design

Searching for materials with target properties

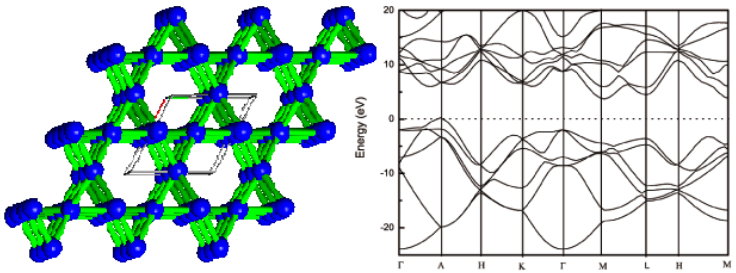


Hybrid optimization:

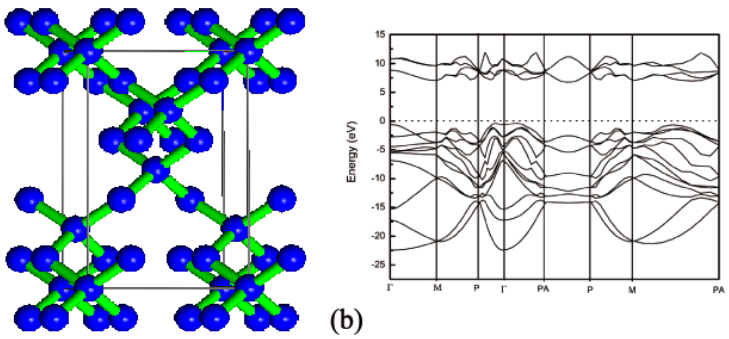
Global	**Properties: (hardness, bandgap, thermal electricity, high/low K)**
Local	Energy

Property landscape instead of energy landscape

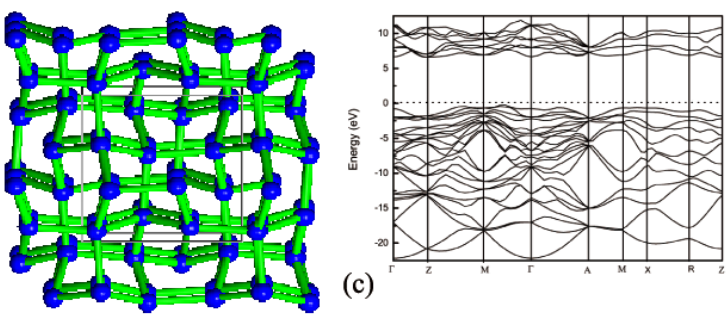
Superdense carbon allotropes



(a)



(b)



(c)

(1) Denser than diamond
(2) sp^3 hybridization
(3) superhard
(4) huge refractive indices (up to 2.8!)
(5) strong dispersion of light
(6) tP12 is the widest-gap form of carbon (7.3 eV)

Density

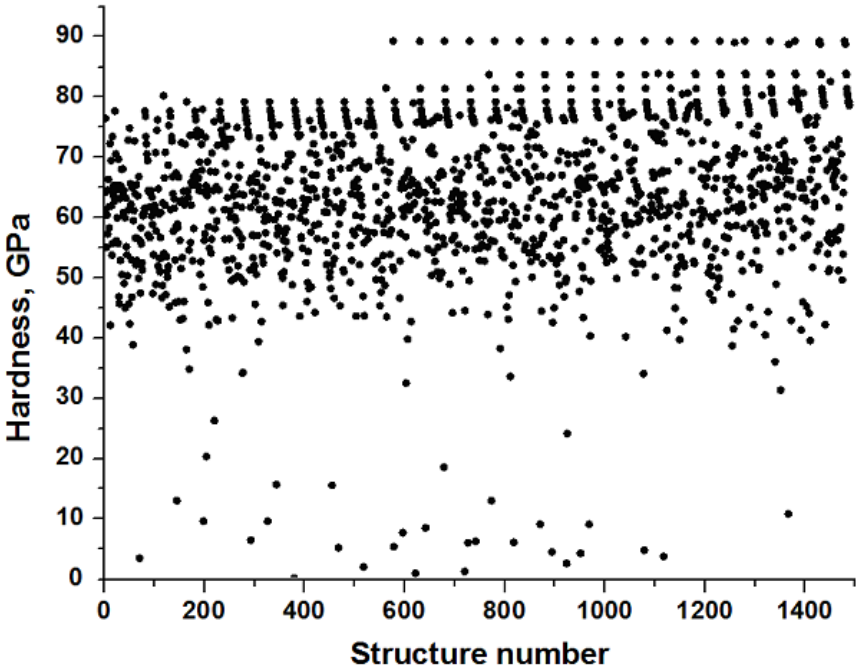
TABLE II. Energy relative to diamond (δE), volume (V), bulk modulus (B_0), average bond length (d), hardness(H), static dielectric constant ϵ_s for the investigated structures. Experimental data are in parentheses

Allotropes	δE eV/atom	V Å^3/atom	B_0 GPa	d Å	H GPa	Band gap eV	ϵ_s
Diamond	0	5.70 (5.68)	431.1 (446)	1.545 (1.54)	94.3 (96±5)	5.4 (5.5)	5.57 (5.68)
Lonsdaleite	0.024	5.71	437.3	1.548	93.2	5.0	5.52
M-carbon	0.159	5.97	392.7	1.551	89.8	5.0	5.52
bct-4	0.196	6.01	411.4	1.549	91.1	3.8	5.42
bc8	0.697	5.60	389.6	1.588	88.8	3.5	6.67
*h*P3	1.113	5.49	432.7	1.603	87.6	3.0	7.73
*t*I12	1.140	5.48	425.0	1.603	87.2	5.5	7.87
*t*P12	0.883	5.64	396.0	1.583	88.3	7.3	7.08

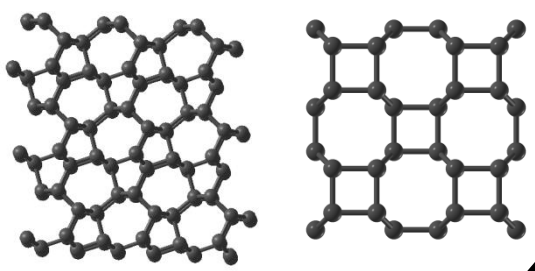
Zhu, et al., PRB, 2011

Superhard carbon allotropes

Hardness



Simulation for carbon, 16 atoms/cell



All of the hardest structures are sp^3-hybridized

Structure	Knoop hardness, GPa	Enthalpy, eV/atom
Diamond	89.7	0.000
Lonsdaleite	89.1	0.026
C2/m	84.3	0.163
I4/mmm	84.0	0.198
Cmcm	83.5	0.282
P2/m	83.4	0.166
$I2_12_12_1$	82.9	0.784
Fmmm	82.2	0.322
Cmcm	82.0	0.224
$P6_522$	81.3	0.111

Lyakhov, et al., PRB, 2011

High Energy Density Materials

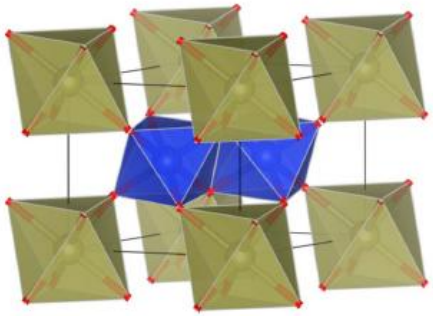
$$E = \frac{1}{2} C U^2,$$

Dielectric Const

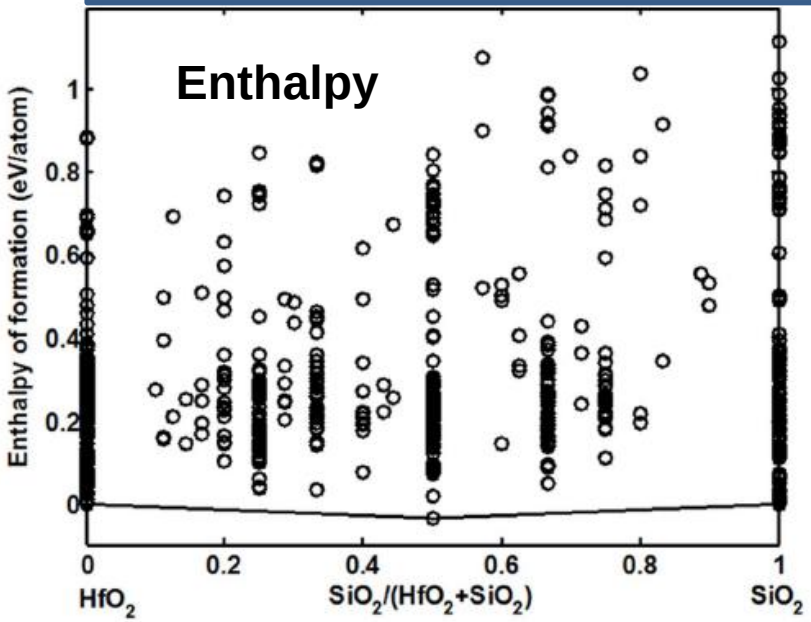
Band gap

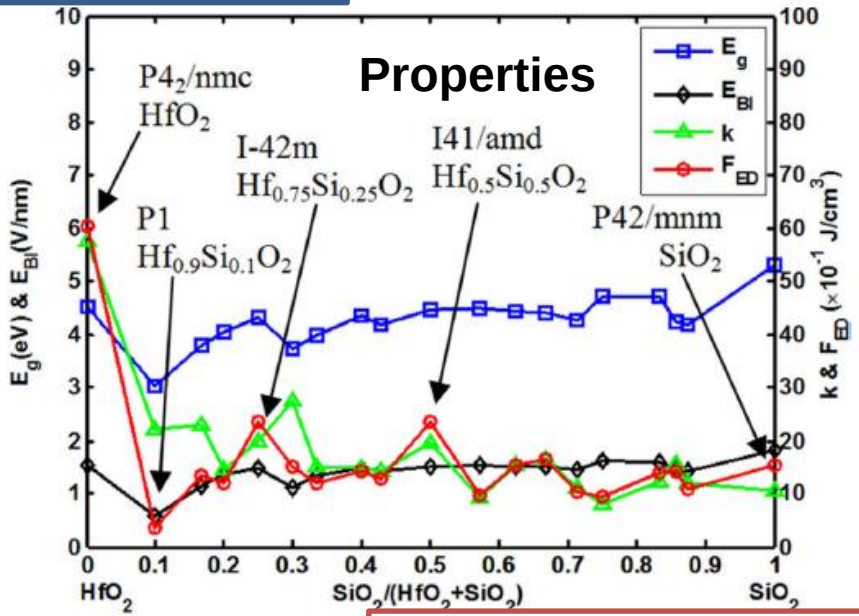
$$C = \varepsilon_0 k A / d \qquad E_{BI} = 1.36 \times (E_g / E_{gc})^{\alpha}$$

$$\max F_{ED} = \frac{1}{2} \varepsilon_0 k E_{BI}^2 = 8.1882\ \mathrm{J\ cm^{-3}} \times k (E_g / E_{gc})^{2\alpha}.$$



(a) $P\bar{3}1m$ ($Hf_{0.33}Si_{0.67}O_2$)





Zeng, et al., Acta Cryst, C, 2014

USPEX: Computational Materials Design

Structure Prediction

System

- **Dimension**
  - 0: Nano-particle;
  - 1: polymers;
  - 2: surfaces/2D crystals;
  - 3: Bulk
- **Stoichiometry**

  0: fixed; 1: variable
- **Building block**

  0: atom; 1: molecule

Target

- Density
- Hardness
- Dielectric constants
- Band gap
- Magnetic moment
- Super capacitor

MATERIALS PROJECT

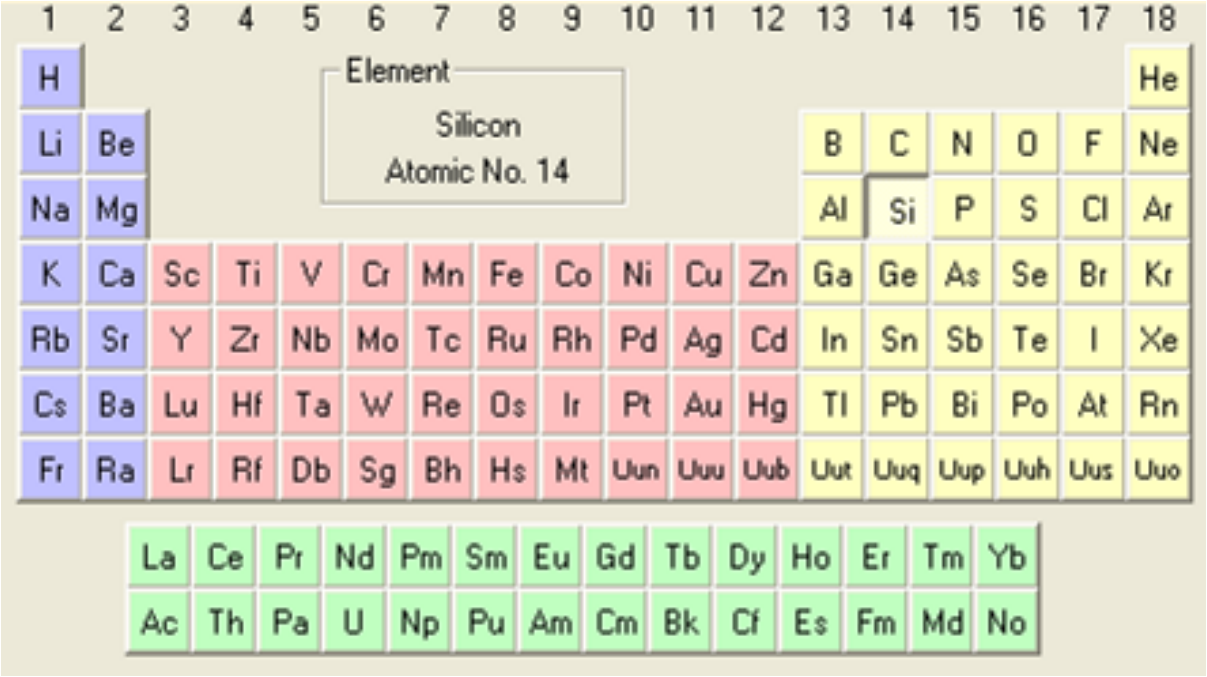
A Materials Genome Approach

Accelerating materials discovery through advanced scientific computing and innovative design tools.

Structural Search
Big data
Machine Learning

Chemical Space

Target Property



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Target

- Density
- Hardness
- Dielectric constants
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- Magnetic moment
- Super capacitor

Synthesis Route

Energy landscape

↓

Generalized Evolutionary Metadynamics

↓

Variable Cell-NEB & Transition Path Sampling

Features of the USPEX code:

Algorithm:

- Evolutionary optimization (USPEX algrorithm). Options to use random sampling, minima-hopping-like, particle-swarm optimization, metadynamics-like algorithms.
- Initialization using fully random, symmetric random structures, or user-fed structures (seeding).
- Fingerprint niching technique, local order parameter (ARO&Valle, 2009). Graph theory and Brown's model for analyzing topology. New variation operators (Lyakhov & ARO, 2010).

Types of runs:

- Global optimization of either the energy or properties (density, hardness, band gap, etc.)
- Fixed-cell or variable-cell, fixed-composition or variable-composition runs are possible.
- For molecular crystals, can operate with ready-made molecules.

Software aspects:

- Interfaced with VASP, SIESTA, CP2k, QuantumEspresso, DMACRYS, GULP.
- Excellent parallel scaling on up to $>10^4$ CPUs.

Analysis:

- Automatic detection of space groups.
- Benefits from powerful analysis and visualization code STM4.

Distribution:

- **USPEX code is freely available at: http://uspex.stonybrook.edu**
- Code distributed with a 50-page manual and ~21 tutorials/tests.
- USPEX Users and Developers Community (>1400 people) **> 2000**

Past workshops

2011 June, Poitiers, France
7 days
2.5 Days DFT
3.5 Days in USPEX
Lectures + tutorials
1 Day excursion

2011 August, Xi'an, China
3.5 days (Lectures + tutorials)
1 day social program

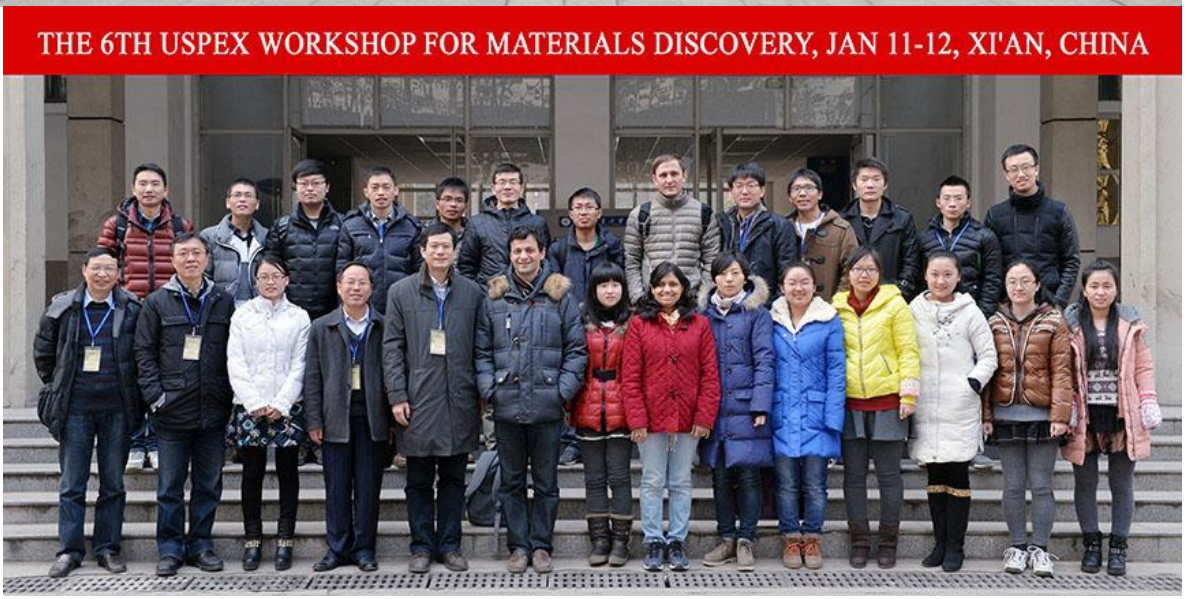
Past workshops

2012 October, Lausanne, Switzerland
4.5 days (Lectures + tutorials)

2012 December, Stony Brook, US
4.5 days (Lectures + tutorials)

Past workshops

2013, Guilin, China, 4.5 days



2014, Xi'an, China, 2.5 days
(for experienced users)

7th workshop in IUCr congress

Half-day workshop,
2 hours lecture
2 hours tutorials

Google uspex

Oganov's Lab

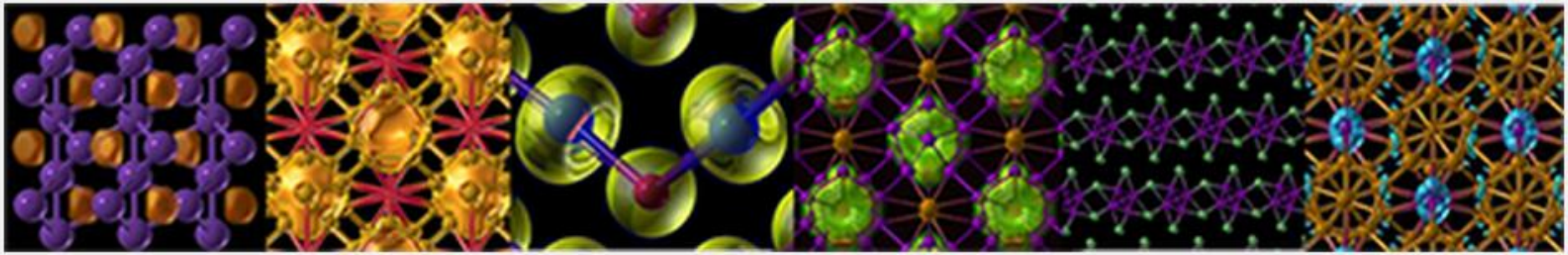
COMPUTATIONAL MATERIALS DISCOVERY LABORATORY

Home Laboratory USPEX A. R. Oganov New Materials New Minerals

USPEX

Overview Downloads Release Notes Publications Workshops Tools and Utilities Documentation Forum

Universal Structure Predictor: Evolutionary Xtallography



What is USPEX?

USPEX (Universal Structure Predictor: Evolutionary Xtalloraphy...and in Russian "uspekh" means "success" - owing to the high success rate and many useful results produced by this method!) is a method developed jointly by Artem R. Oganov, Andriy O. Lyakhov, Colin W. Glass, Qiang Zhu, and others implemented in the same-name code. The problem of crystal structure prediction is very old and does, in fact, constitute the central problem of theoretical crystal chemistry. In 1988 John Maddox wrote that:

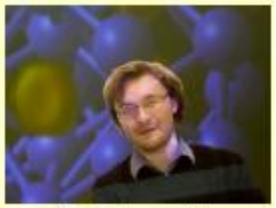
"One of the continuing scandals in the physical sciences is that it remains in general impossible to predict the structure of even the simplest crystalline solids from a knowledge of their chemical composition solids such as crystalline water (ice) are still thought to lie beyond mortals' ken".

USPEX method/code enables crystal structure prediction at arbitrary P-T conditions, given just the chemical composition of the material. Today, USPEX is used by over 1600 researchers worldwide. The popularity of USPEX is due to its extremely high efficiency and reliability. The First Blind Test of Inorganic Crystal Structure Prediction shows that USPEX outperforms other methods in terms of efficiency and reliability. In addition to crystal structure prediction, USPEX can work in other

Acknowledgement

Developers

A.R. Oganov


A.O. Lyakhov

C.W. Glass

Q. Zhu

G.R. Qian

M. Rakitin

H.T. Stokes

Q.F. Zeng

M. Valle

Funds

intel

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WHERE DISCOVERIES BEGIN

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Rapidly growing user community….

3 laboratories: US (Stony Brook), Russia (Moscow), China (Xi'an).

Thanks for your attentions!