Applications of 1st-Principles Based Evolution Algorithms in Material Design and Discovery

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1st Workshop of HPC on Nano-scale Materials Research

Aug/13/2009, NCKU, Tainan, Taiwan

Outlines:

- ***What is 1st-Principles Simulations?***

  Why do we need Evolution Algoritm?

- ***Proton Order/Disorder transitions in Ice***

  a 70 year old problem in ice physics.

- ***ZnO based alloy***

  Alloy Synthesis on the Cloud ?

- ***Structure of nano-sized clusters***

Knowing the structure is the FIRST step toward understanding Material Properties

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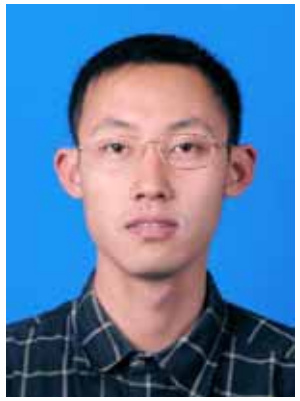


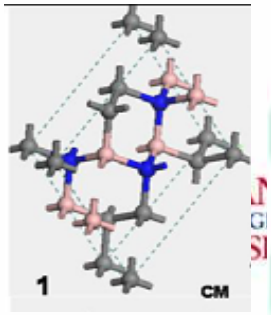
Wu Hongyu

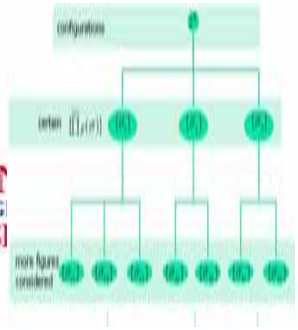
Nguyen Quoc Chinh

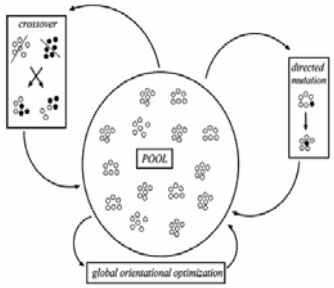
Bing Dan

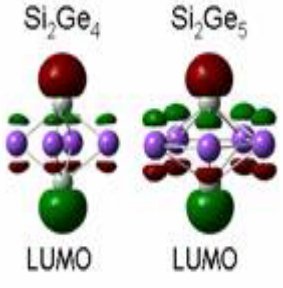
Zhang Jingyun

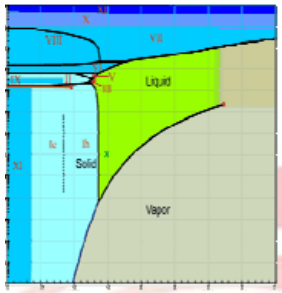
Lee Ching-Tao



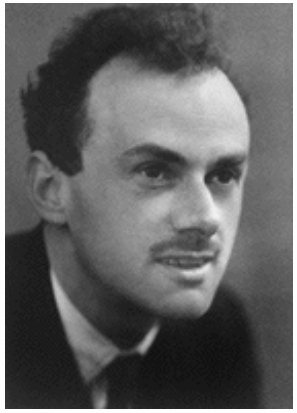






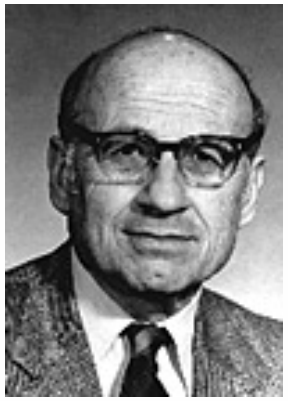


Basis of 1st-Principles Methods



Erwin Schrödinger Paul A. M. Dirac

Nobel Prize in Physics 1933



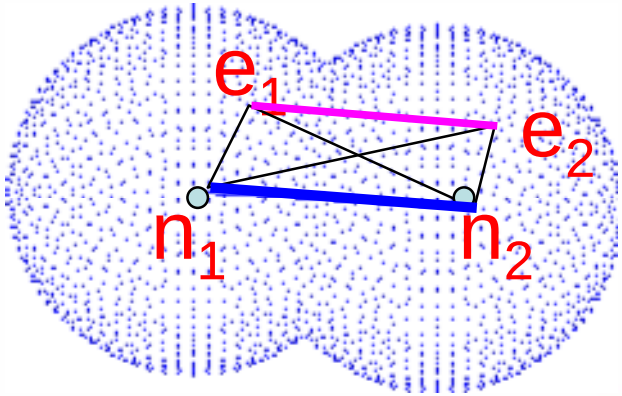
Walter Kohn John A. Pople

Nobel Prize in Chem. 1998

Dirac (1929): "The underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known."

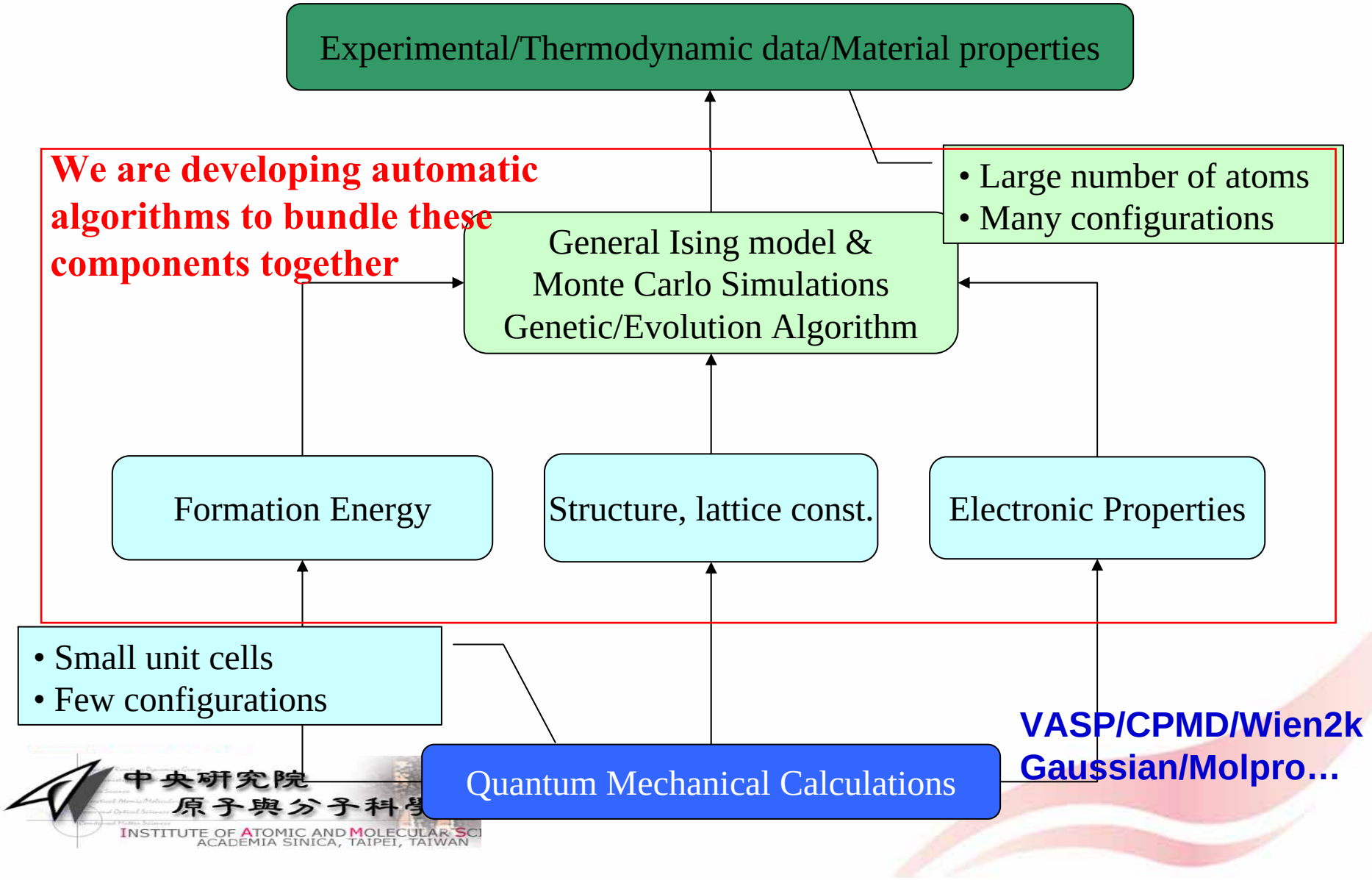
Schrödinger equation:

$$\mathbf{H}\psi = E\psi$$

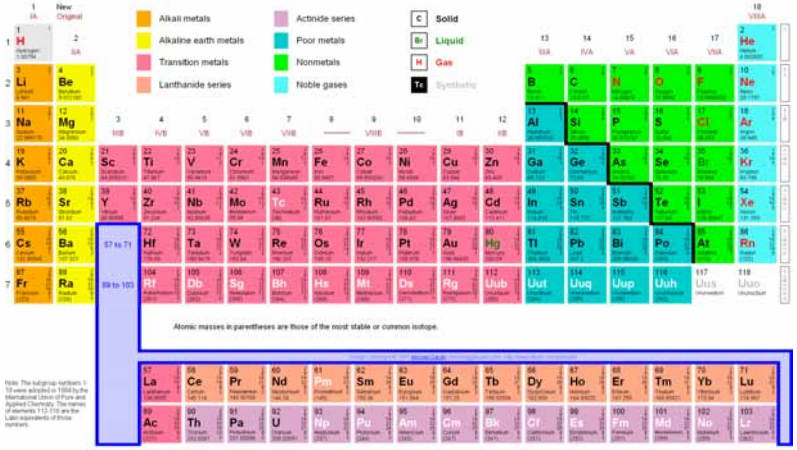


"for his development of DFT and comp. methods in quantum chemistry"

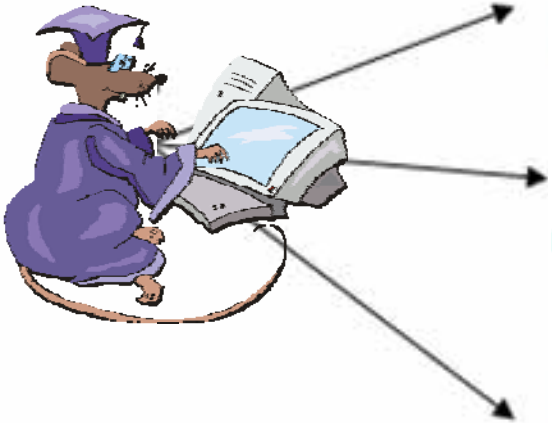
Multi-scale simulation from Ab Initio

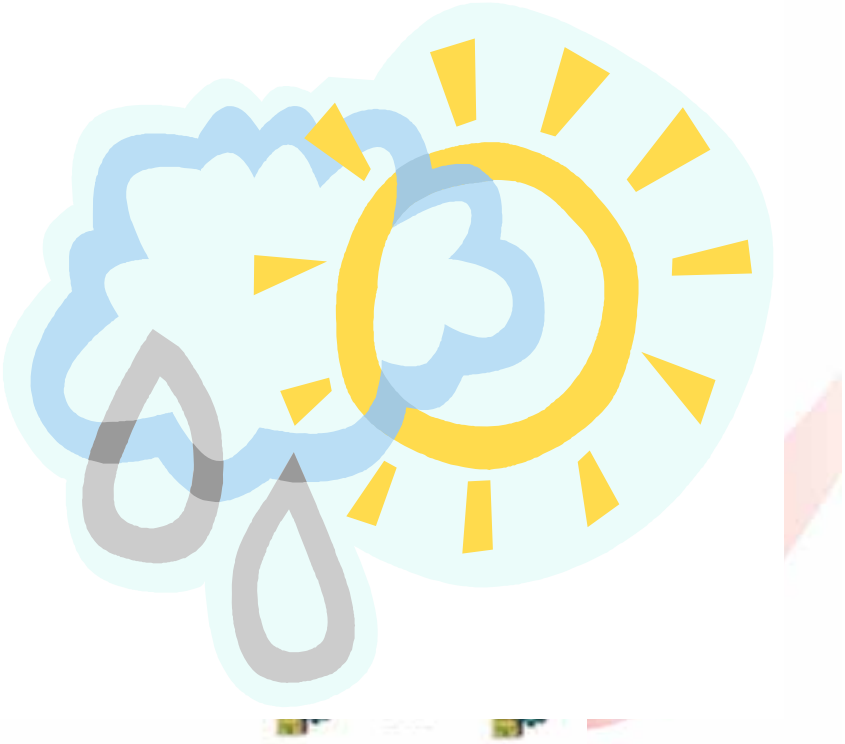


Material Synthesis on the CLOUD



GRID → CLOUD





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- *What is 1st-Principles Simulations:*
  Why Multi-scale and go CLOUD?

- ***Proton Order/Disorder transitions in Ice***
  a 70 year old problem in ice physics.

- *$Be_xZn_{1-x}O$?*
  Alloy Synthesis on the Cloud.

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Knowing the structure is the FIRST step toward understanding Material Properties

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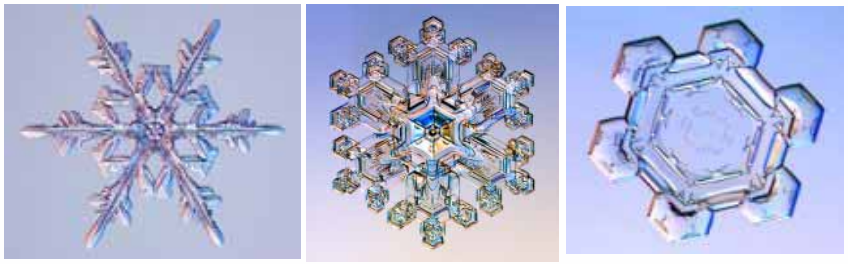
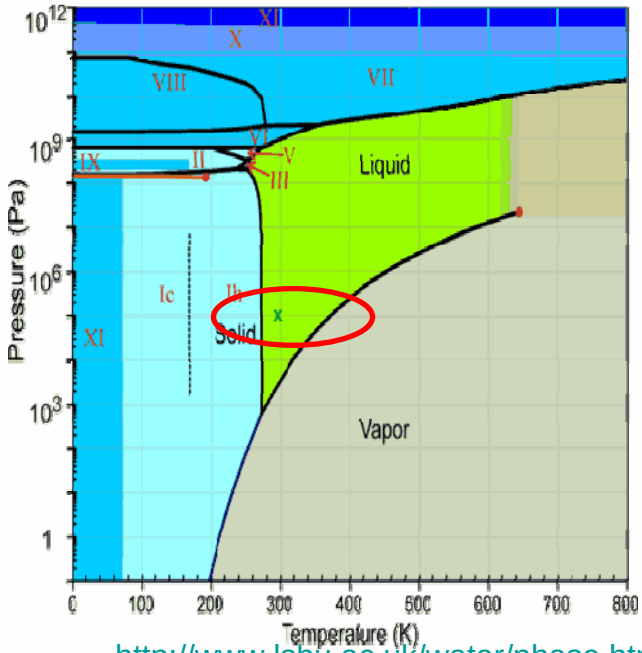
WHAT!! Ordinary Ice is NOT crystal.

- Snow flakes have beautiful ***hex-symmetry*** !!



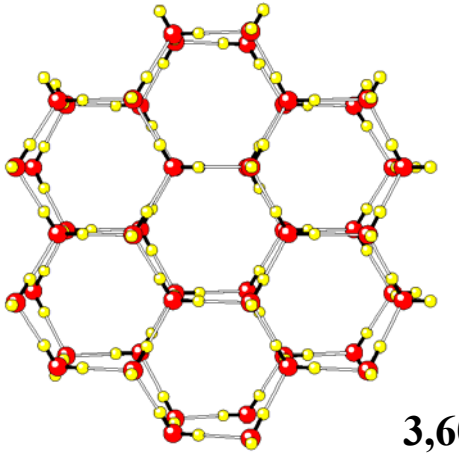
http://www.its.caltech.edu/~atomic/snowcrystals/



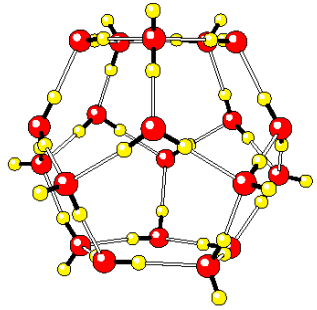
http://www.lsbu.ac.uk/water/phase.html

- A **crystal** is a solid in which the components are packed in a regularly ***ordered***, ***repeating*** pattern extending in all three spatial dimensions

- Ice has a ***disordered*** proton distribution.

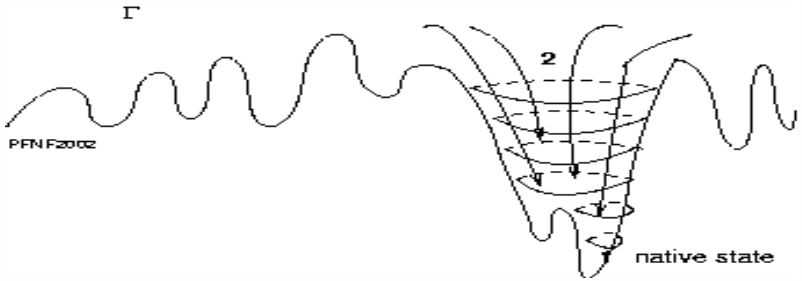


In this 48-water unit cell of ice-Ih, there are 2,404,144,962 H-bond isomers.



3,600,000 H-bond isomers.

Residual Entropy of ice-Ih

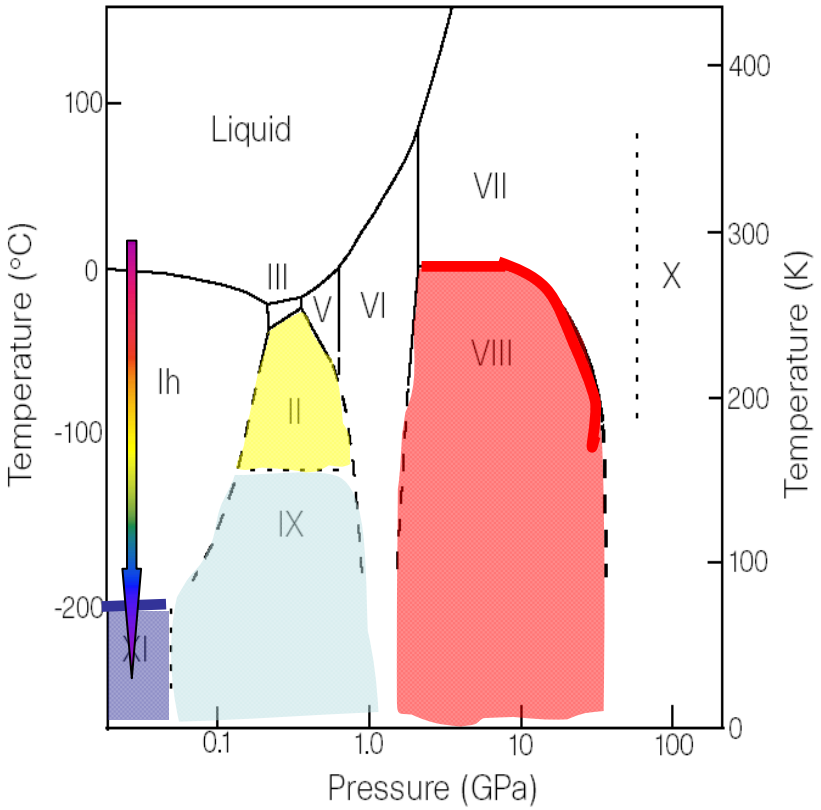


L.Pauling *JACS* **57**, 2680 (1935)

$$\Omega \propto (3/2)^N, S = k \ln \Omega = 3.37\ J/K$$

W.F. Giauque and J.W.Stout,
JACS **58**, 1144 (1936)

$$S(T \to 0) = 3.4 \pm 0.6\ J/K$$



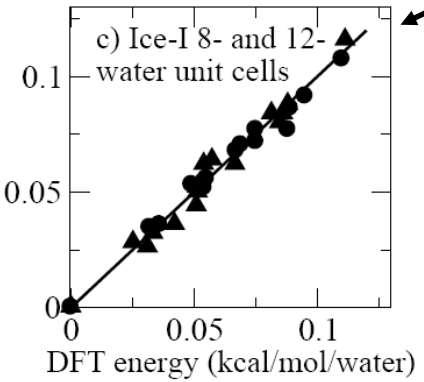
Nature ,vol 391, p.268

- Not just ice-Ih, many phases of ice are proton-disordered.
- Thermal properties of ice is essential to many important issues in physics, chemistry, and environment.
- Finding the proton-ordered ice, will help us better understand H-bonding.

NANYANG TECHNOLOGICAL UNIVERSITY

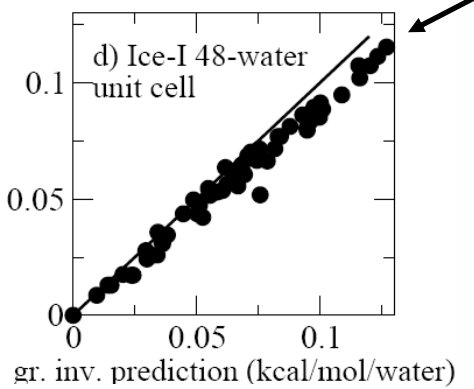
Proton Order/Disorder transition

Training graph invariants on small unit cells ***(Quantum Mech.)***

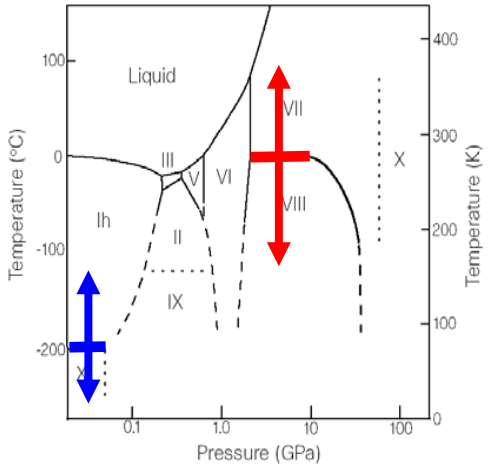


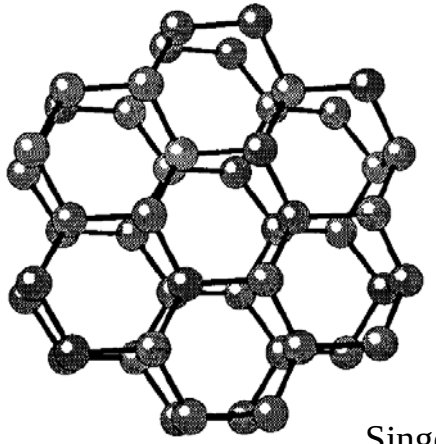
$$E(b_1, b_2, \ldots) = E_0 + \sum_r \alpha_r I_r + \sum_{rs} \alpha_{rs} I_{rs} + \ldots$$

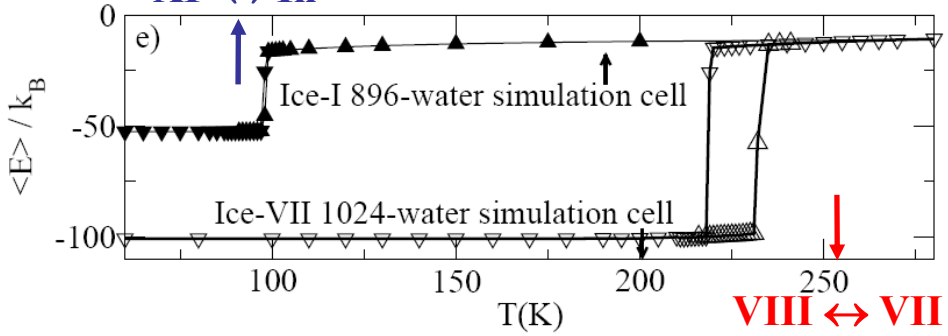
Testing the prediction on larger unit cells. ***(ab initio) – QM***



MC simulations on very large unit cells to simulate disorder/order transition. – ***SM***



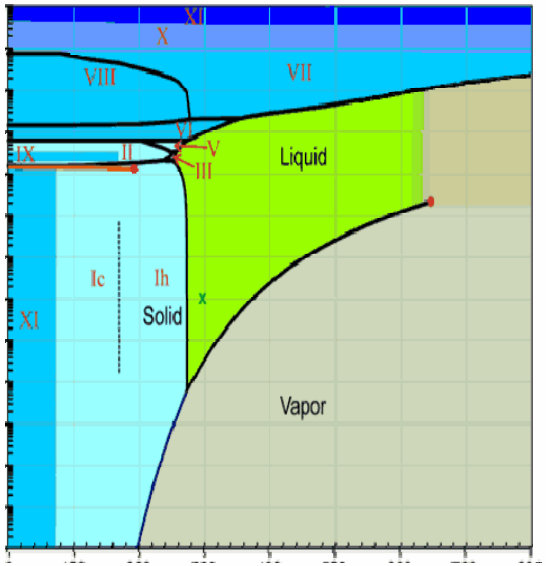




Singer, Kuo, *et al*, ***Phys. Rev. Lett. 94,*** 135702 (2005)]

What else about ice:

- *New Phases?*



- *Effects of Pressure?*

- *X-ray Absorption Spectra*

Zhang Jingyun

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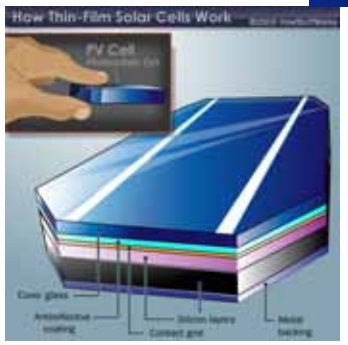
ZnO Alloys for opto-electronic applications

ZnO

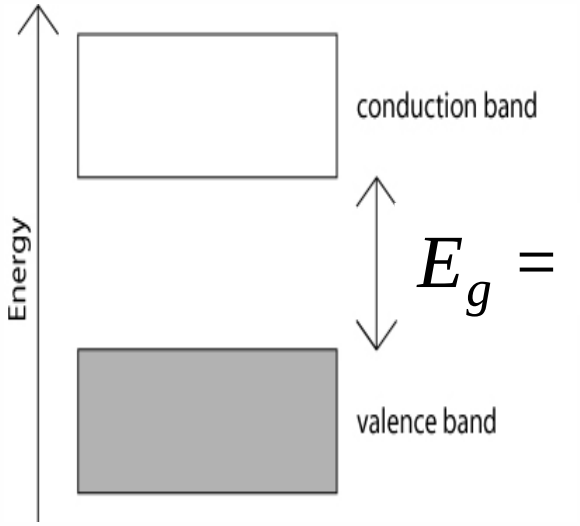
LED



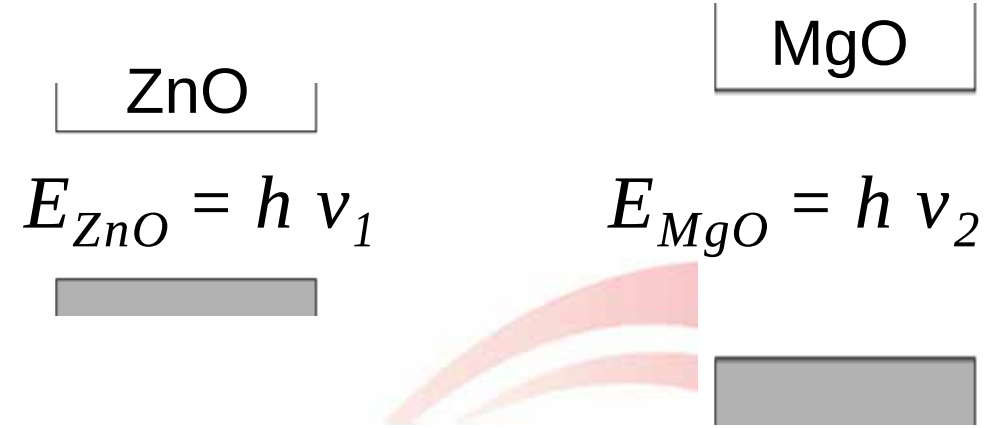
Solar Cells

Sun cream

- **Band Gaps and light**

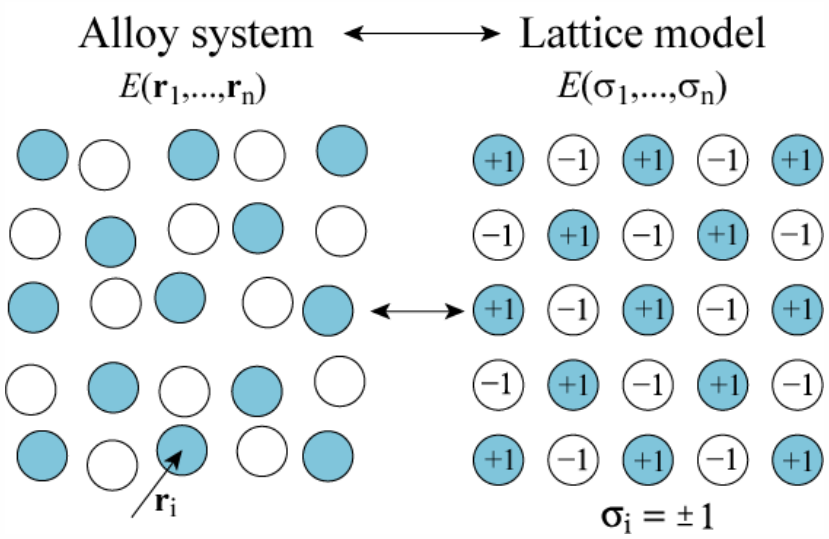


- **Tunable E_g**



Semiconductor Alloys – $Be_xZn_{1-x}O$

➢ Cluster Expansion Theory

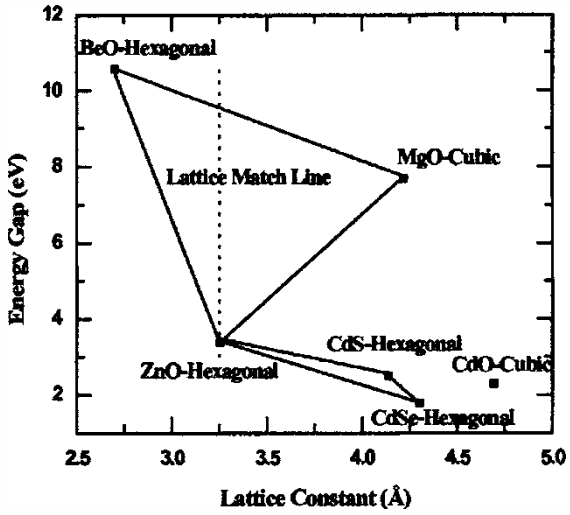


$$E(\sigma_1,...,\sigma_n) = \sum_{\{i,j\}} J_{ij}\,\sigma_i\sigma_j + \sum_{\{i,j,k\}} J_{ijk}\,\sigma_i\sigma_j\sigma_k + ...$$

How many J_{ij} are needed? 2^N

How many σ are needed? 2^N

➢ $Be_xZn_{1-x}O$

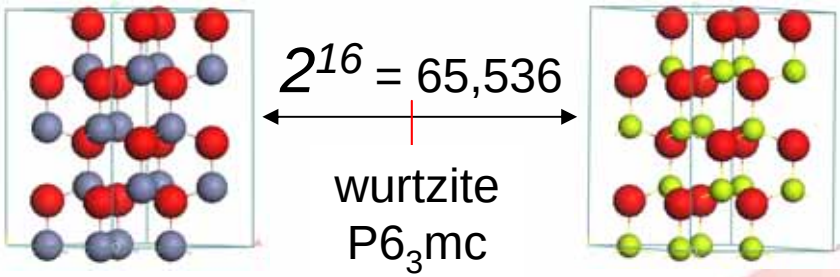


ZnO (*Eg*=3.4eV)

MgO (*Eg*=7.7eV)

BeO (*Eg*=10.6eV)

APL **88**, 052103 (2006)



2^{16} = 65,536

wurtzite $P6_3mc$

652 sym-distinc

ZnO

x=0

BeO

x=1

σ=(Zn, Zn, …)

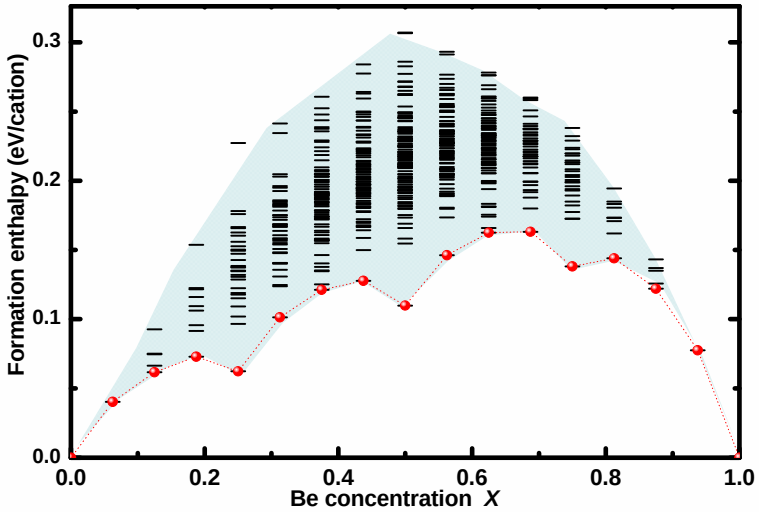
σ=(Be, Be, …)

$Be_xZn_{1-x}O$ Alloys

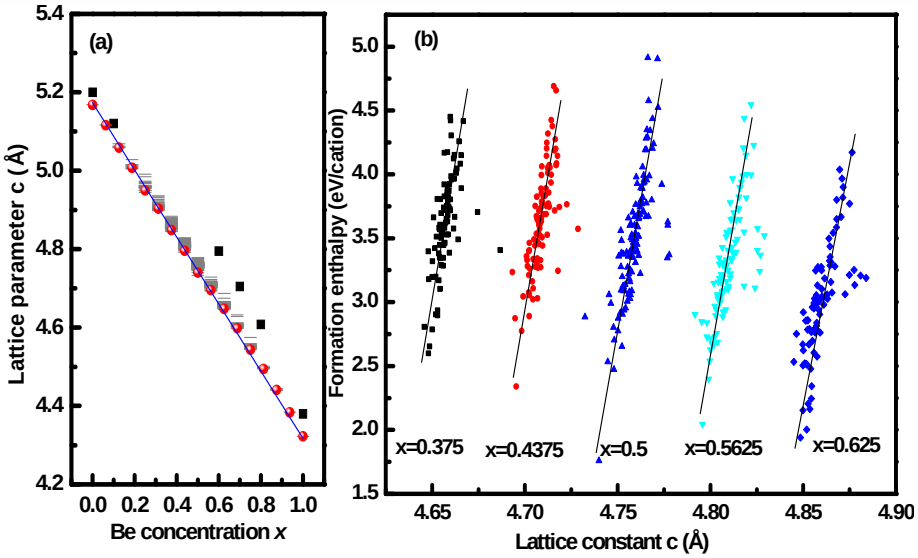
Fan, Zhu et al,***APL,* 91**, 121121 ('07).

➢ Formation Enthalpy:

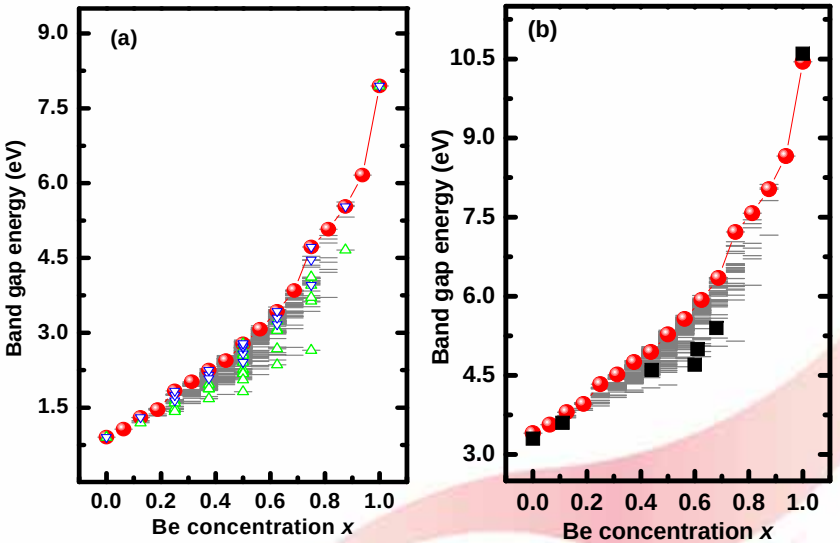
$$\Delta H_f[\sigma,x] = E[\sigma, Be_xZn_{1-x}O] - xE[BeO] - (1-x)E[ZnO]$$



➢ Lattice constants



➢ Band Gaps



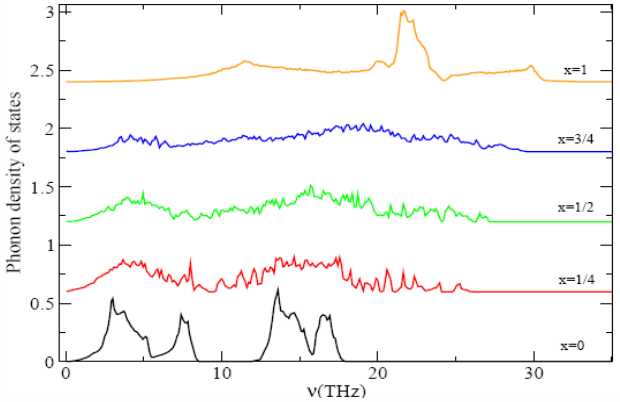
△ [100] SL ▽ [001] SL ■ Exp

XF Fan, .., and Jer-Lai Kuo,
App. Phys. Lett. ***91, 121121 (2007)***

Effect of Lattice Vibration

➢ *Phonon DOS*

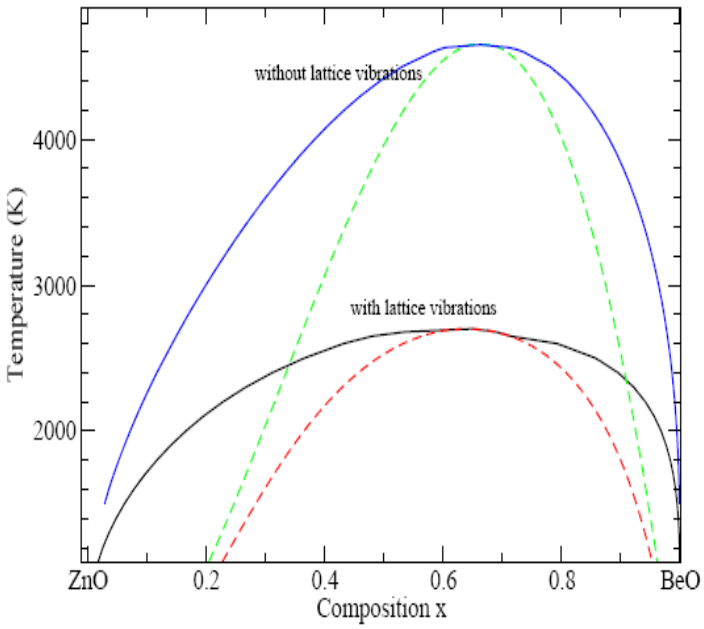
- PWSCF: linear response
  good for crystal.
- PHONE: calculate DOS



➢ *Effect of Lattice Vibration*

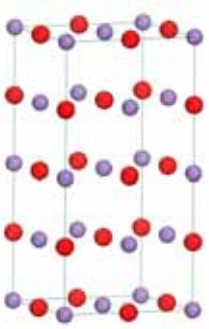
Approximations made:

- Bragg-Williams app.
- Neglect short range order
- Harmonic app.
- Small super-cell size

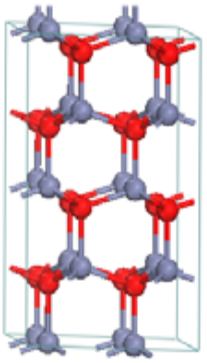


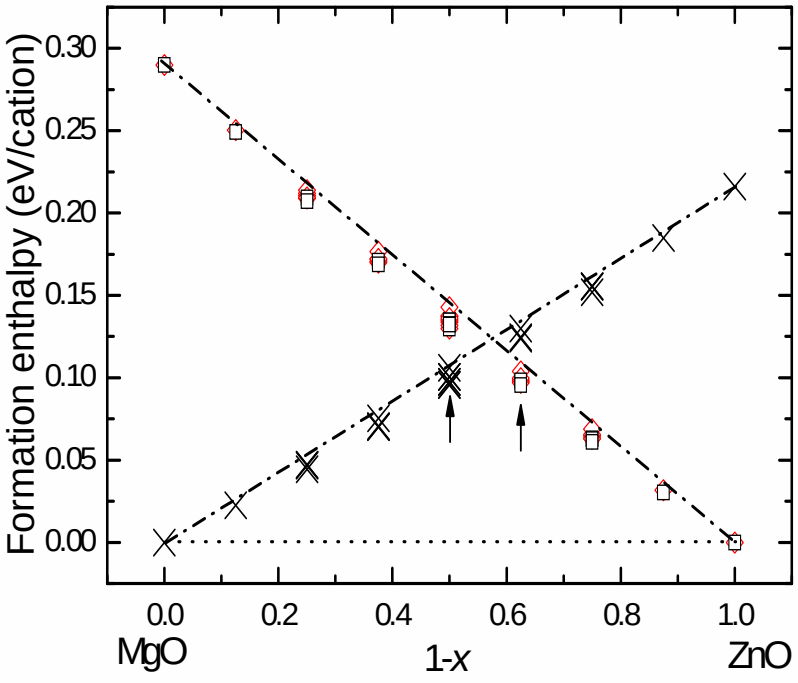
CK Gan, XF Fan, and Jer-Lai Kuo,
Comp. Mat. Sci (in press)

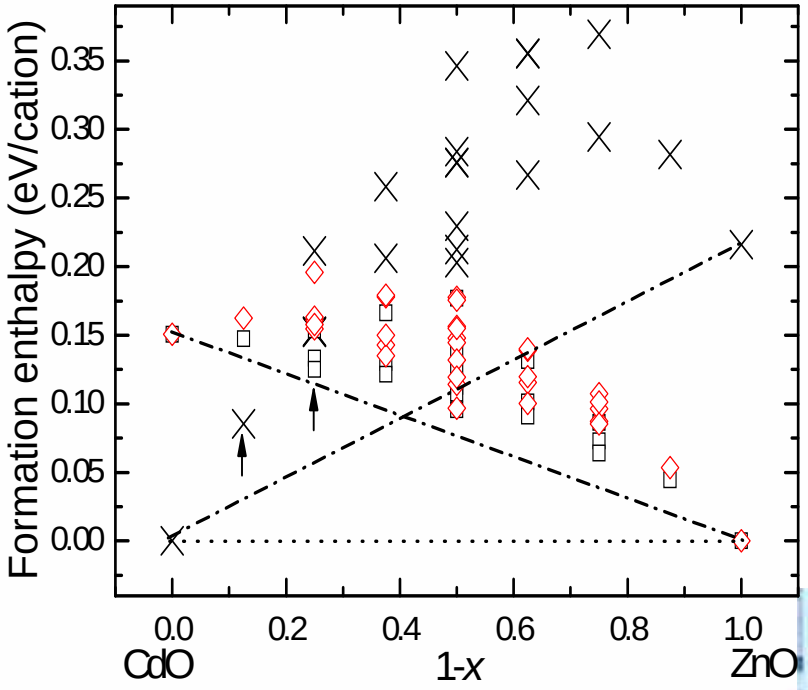
Non-isostructural alloys



- $Mg_xZn_{1-x}O$, $Cd_xZn_{1-x}O$,.... 66







X. F. Fan, HD Sun, Z.X. Shen, and Jer-Lai Kuo, *J. Phys. Cond. Matt.* ***20, 235221 (2008)***

Material Discovery on the CLOUD

- ***1st-Principle Calculations to replace wet-lab based methods?***

- ***Improve efficiency***

2 month (2007) with in-house PC clusters

2 days (2008) with in-house PC clusters

2 hrs (2009) with large HPC?

- ***Material Design***

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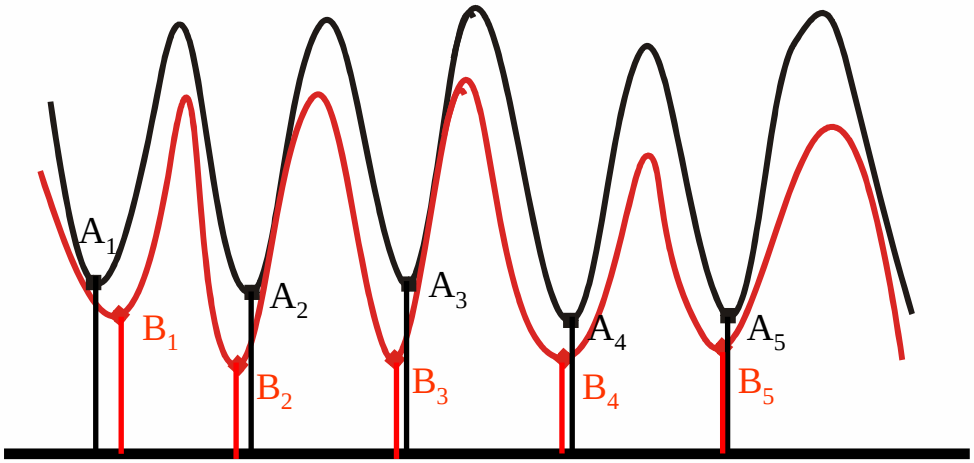
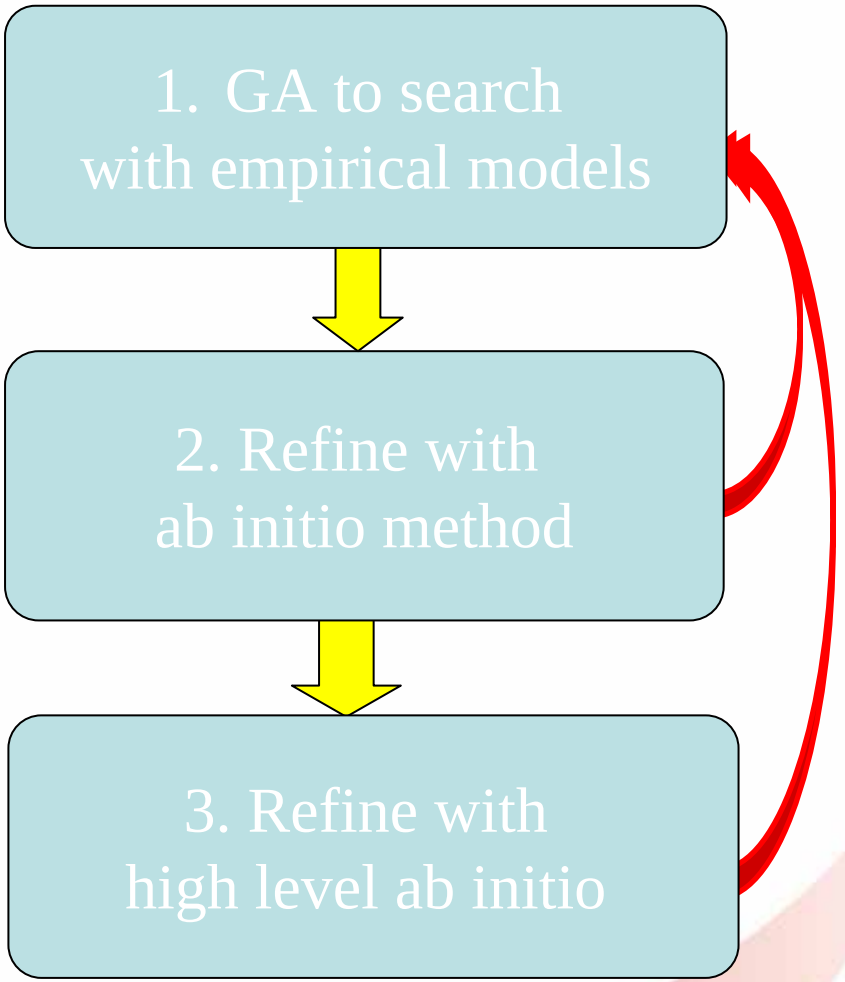
Basic scheme of the Multi-scale Method

- Use empirical models to quickly explore the PES of water
- Ab initio results can be used in return to reparameterized models



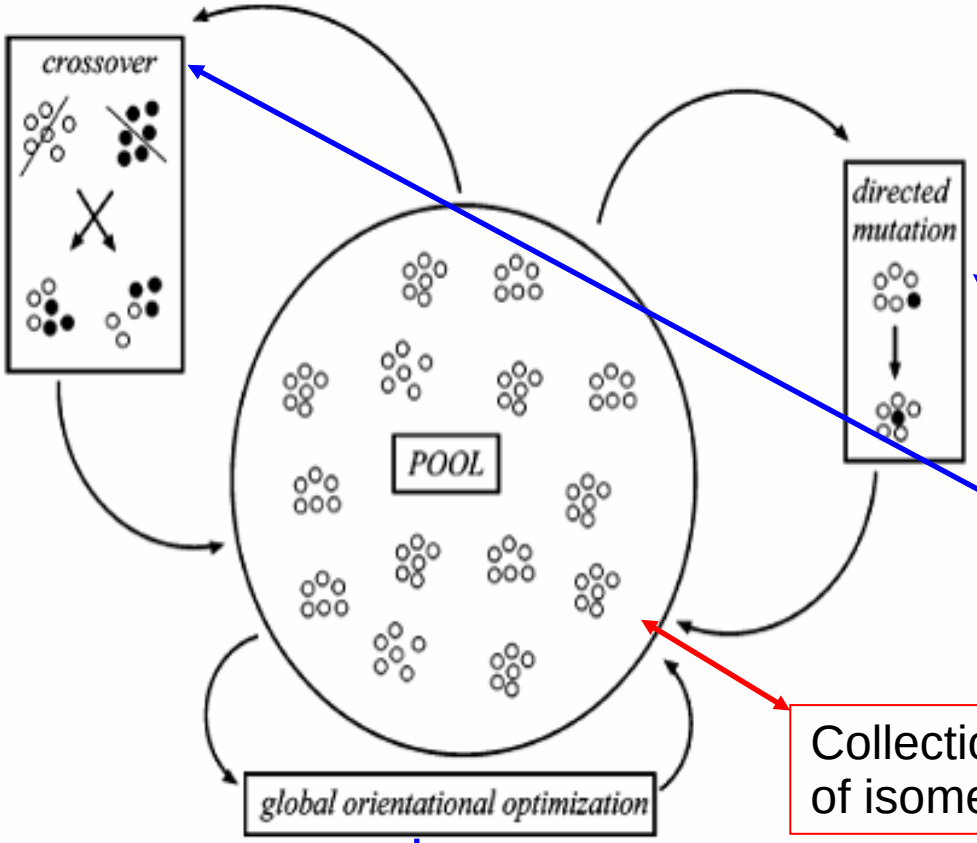
➔A replaced by (➔B➔A)

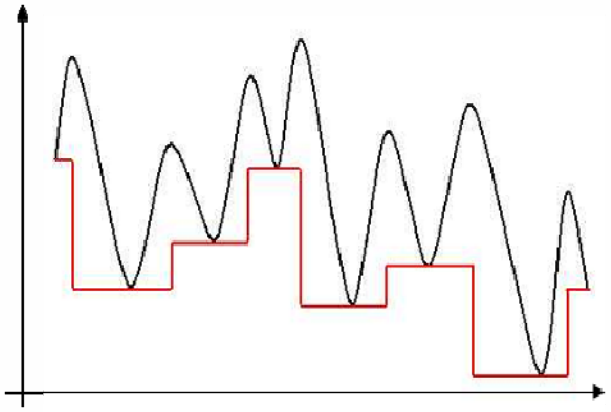


Nguyen QC.. Jer-Lai Kuo,
J. Phys. Chem. ***A 112, 6257 (2008)***

Exploring the Energy Landscape of Clusters using Genetic Algorithms

Asynchronized GA: by Bernd Hartke





Genetic Algorithm

Mutation & Crossover : works directly on coordinates of molecular clusters

Collection of isomers

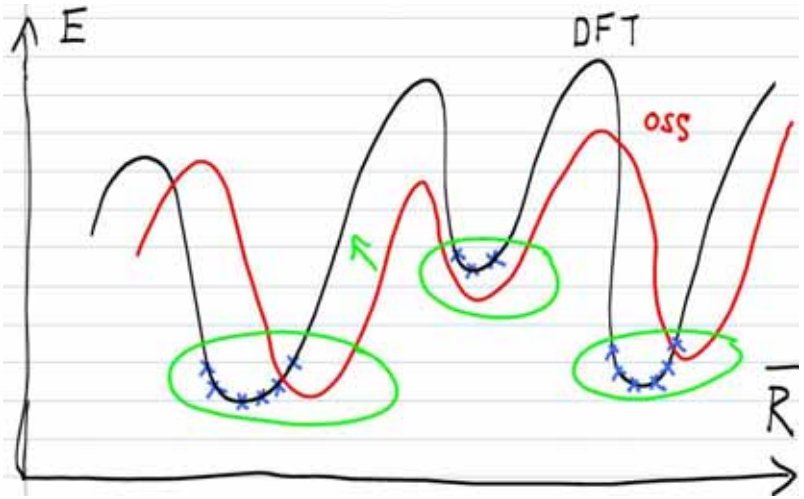
A collector was implemented to gather new isomer found by GA

suitable for GRID comp.

Soh, Nguyen ...Ong, Kuo
IEEE/ACM trans…

Parameterizing empirical models

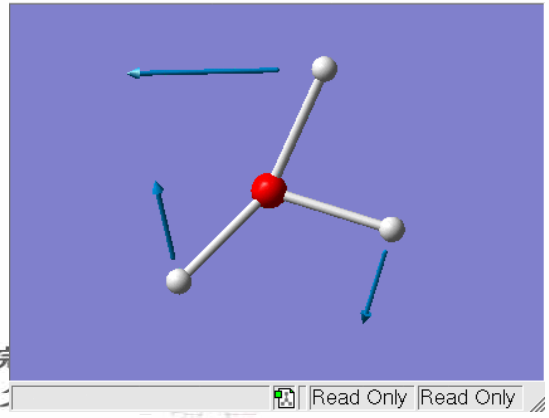
- Levenberg-Marquardt nonlinear least square algorithm + Genetic Algorithm

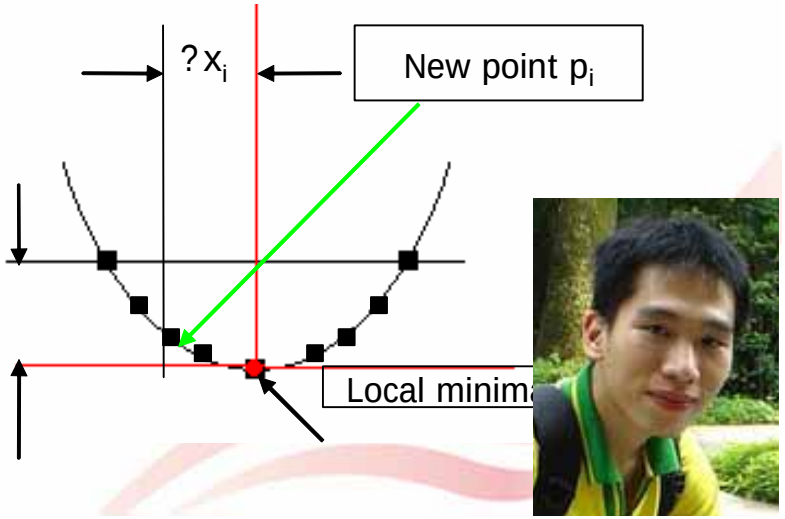


- Objective function

$$F(p)=\sqrt{\frac{1}{M}\sum_{k=1}^{M}(E_{OSS2}^{k}(p)-E_{DFT}^{k})^{2}}$$

p: parameter, M: # of data points,
E_{Oss2}^{k}, E_{DFT}^{k}: OSS2 and DFT binding energy of configuration k



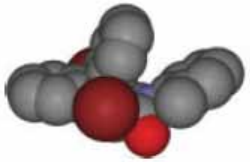


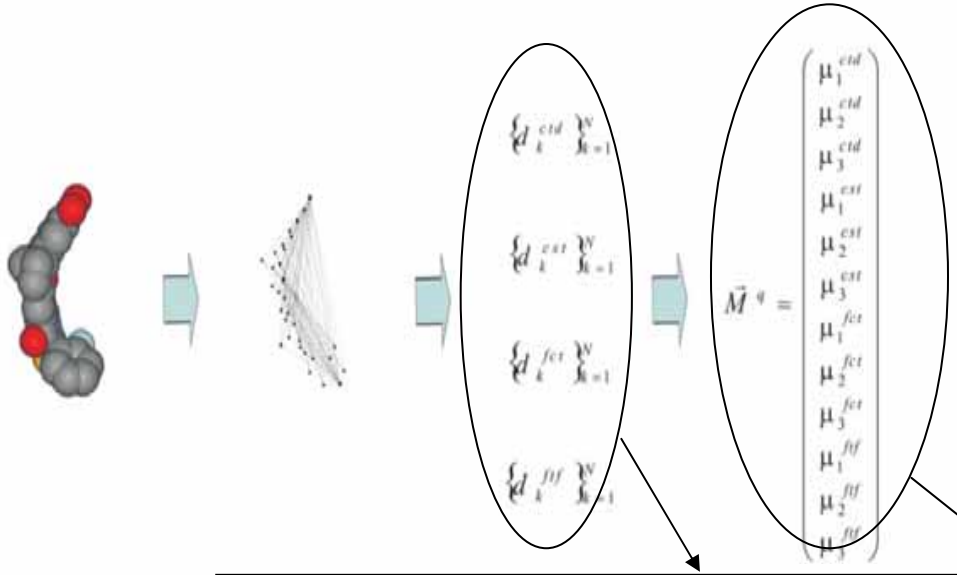
How to distinguish diff. isomers?

"Ultra fast shape recognition"

Ballester and Richards, J. Comp. Chem. **28**, 1711 (2007)

- Similarity index ranges from 0 to 1:
  - 0 : totally different
  - 1 : exactly the same
- Differ by just 1 atom, SI= 0.966





$\{d_k^{ctd}\}$: to molecular centroid (ctd)

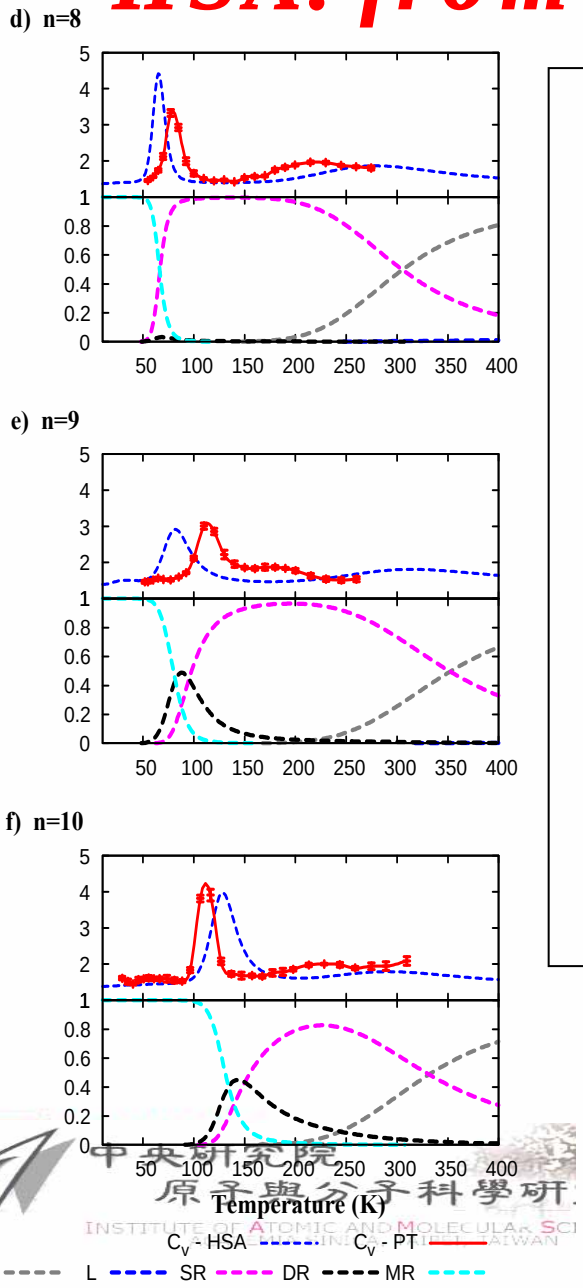
$\{d_k^{cst}\}$: to the closest atom (cst) of ctd

{dkfct}: to the furthest atom (fct) of ctd

{dkftf}: to the furthest atom (ftf) of fct

To make them independent to the size of cluster, the first, second and third moments of each set are used.

HSA: from isomers to thermal prop.

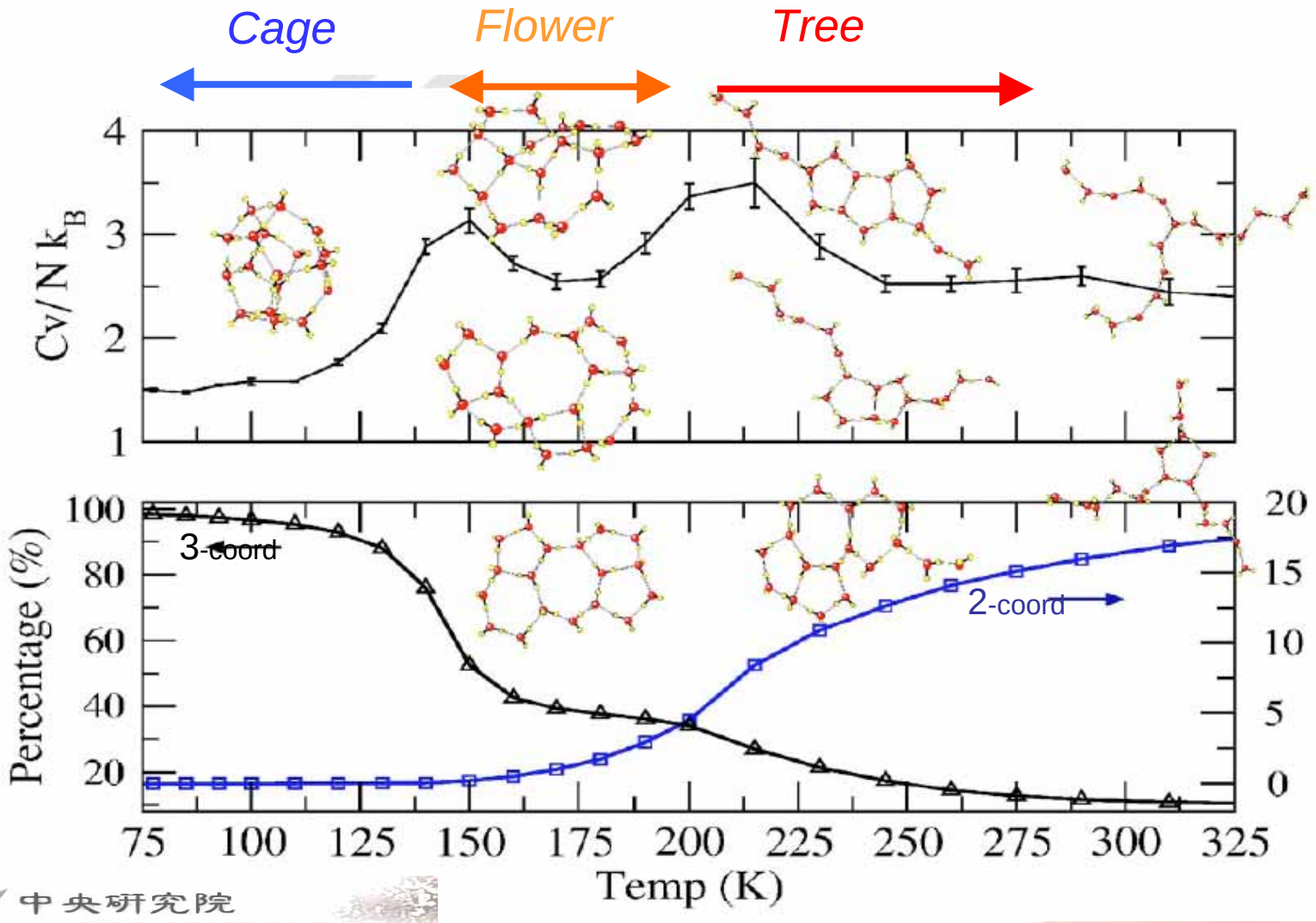


Harmonic superposition approximation

- The canonical partition function Z (β) can be approximated as the summation of harmonic contributions of all collected local minima.
- $$Z(\beta) = \sum_a n_a Z_a(\beta)$$
- The finite temperature behavior (heat capacity curves, structural transitions, canonical probabilities .i.e.) of Wn+ clusters can be derived afterward.

Nguyen, Ong, and Kuo,
"A multi-scale approach to study thermal behavior of protonated water clusters $H^+(H_2O)_n$",
J. Chem. Theory and Comp.)

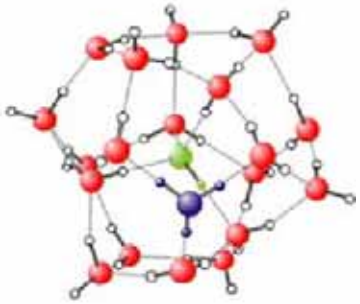
Two-stage melting of $H^+(H_2O)_{18}$

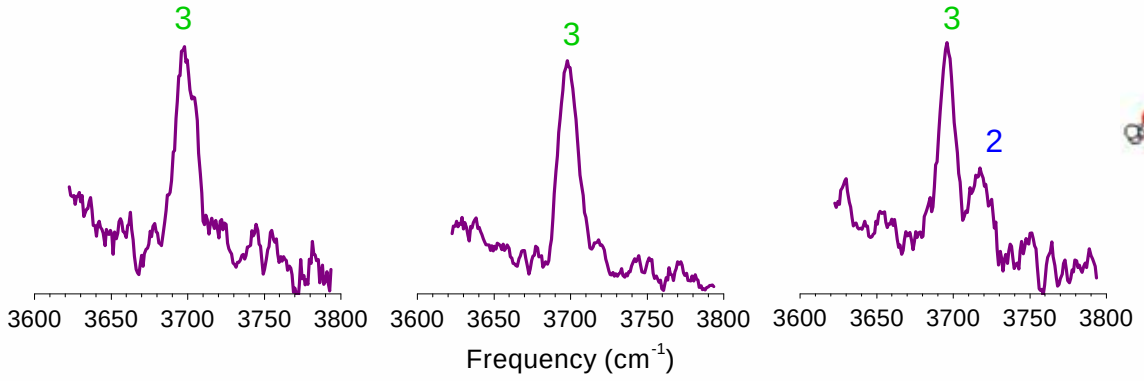


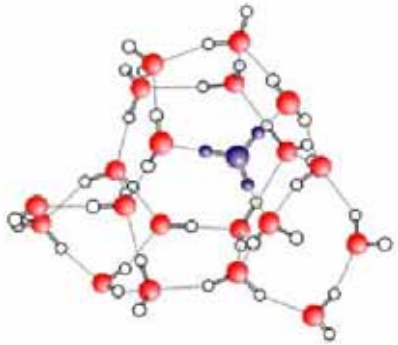
Kuo and Klein *J. Chem. Phys.* 112, 24516 (2005).

Melting of $H^+(H_2O)_{21}$ cage (135~155K)

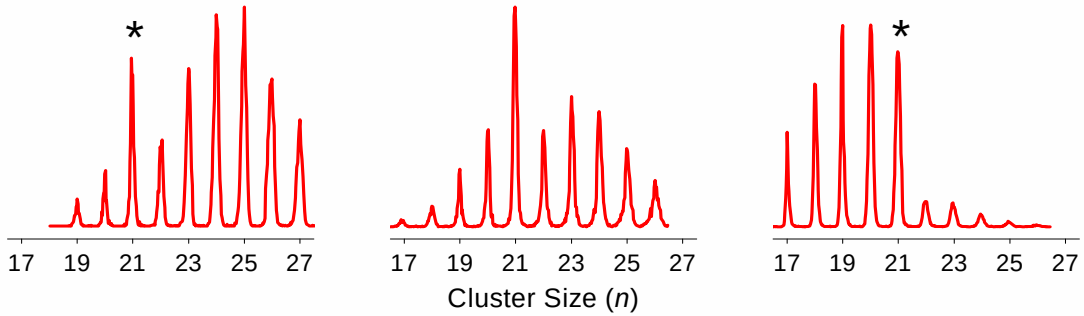
(Temp) cold ---------- warm

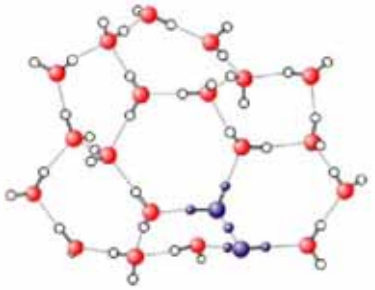




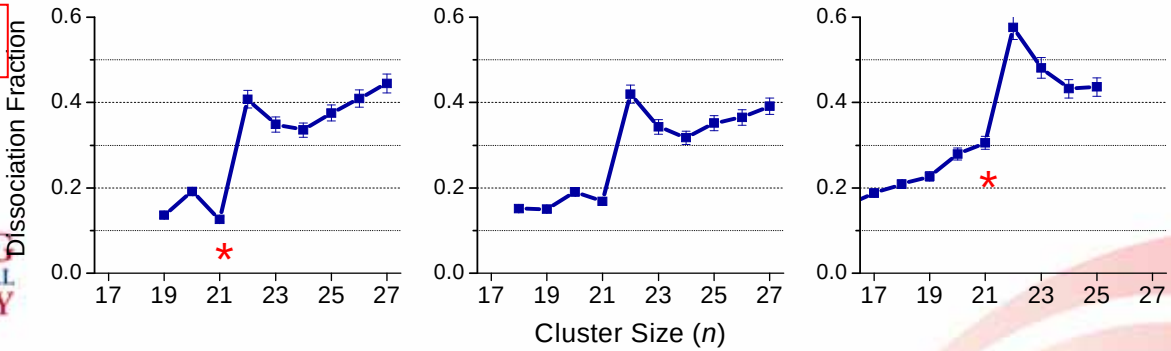


Mass spectra





Dissociation



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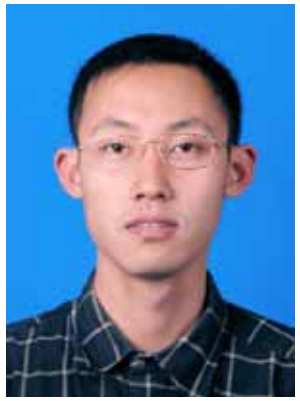
$$ =

Fan XiaoFeng



Wu Hongyu

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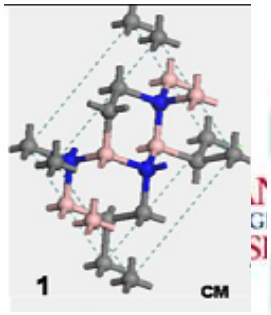
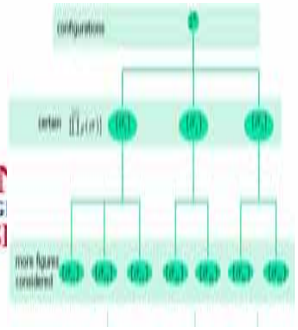
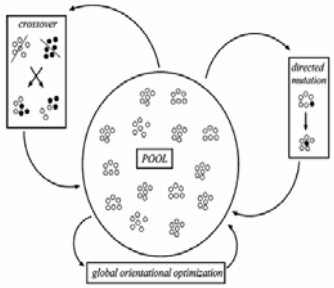
Bing Dan

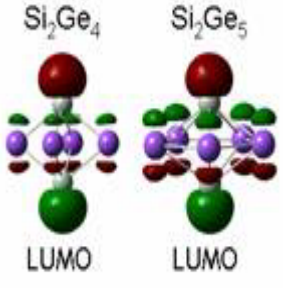
Zhang Jingyun

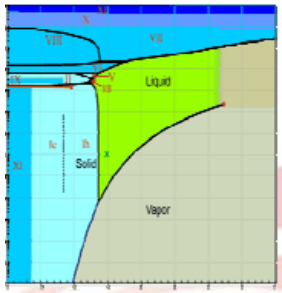
Lee Ching-Tao



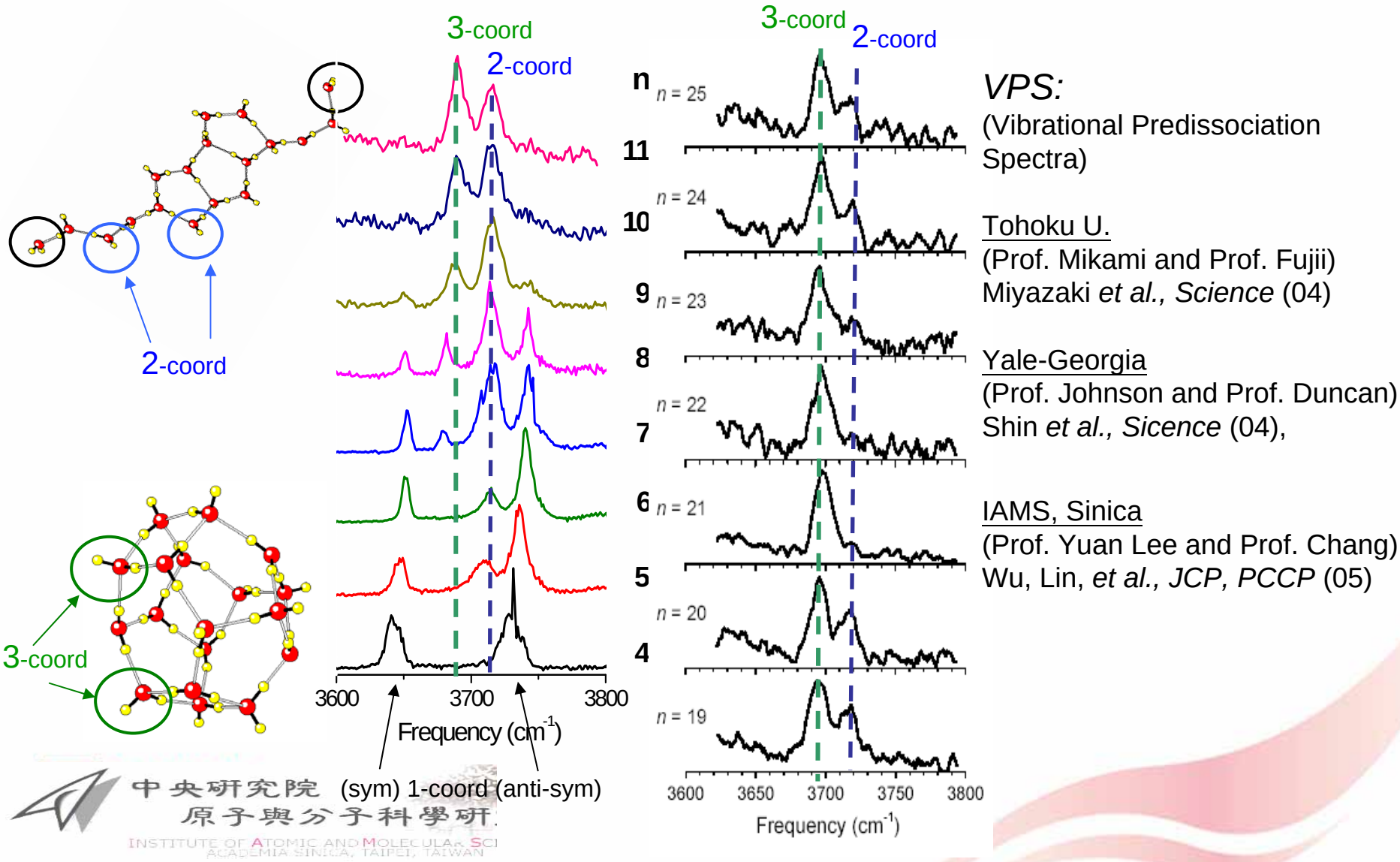








Structural info from VPS



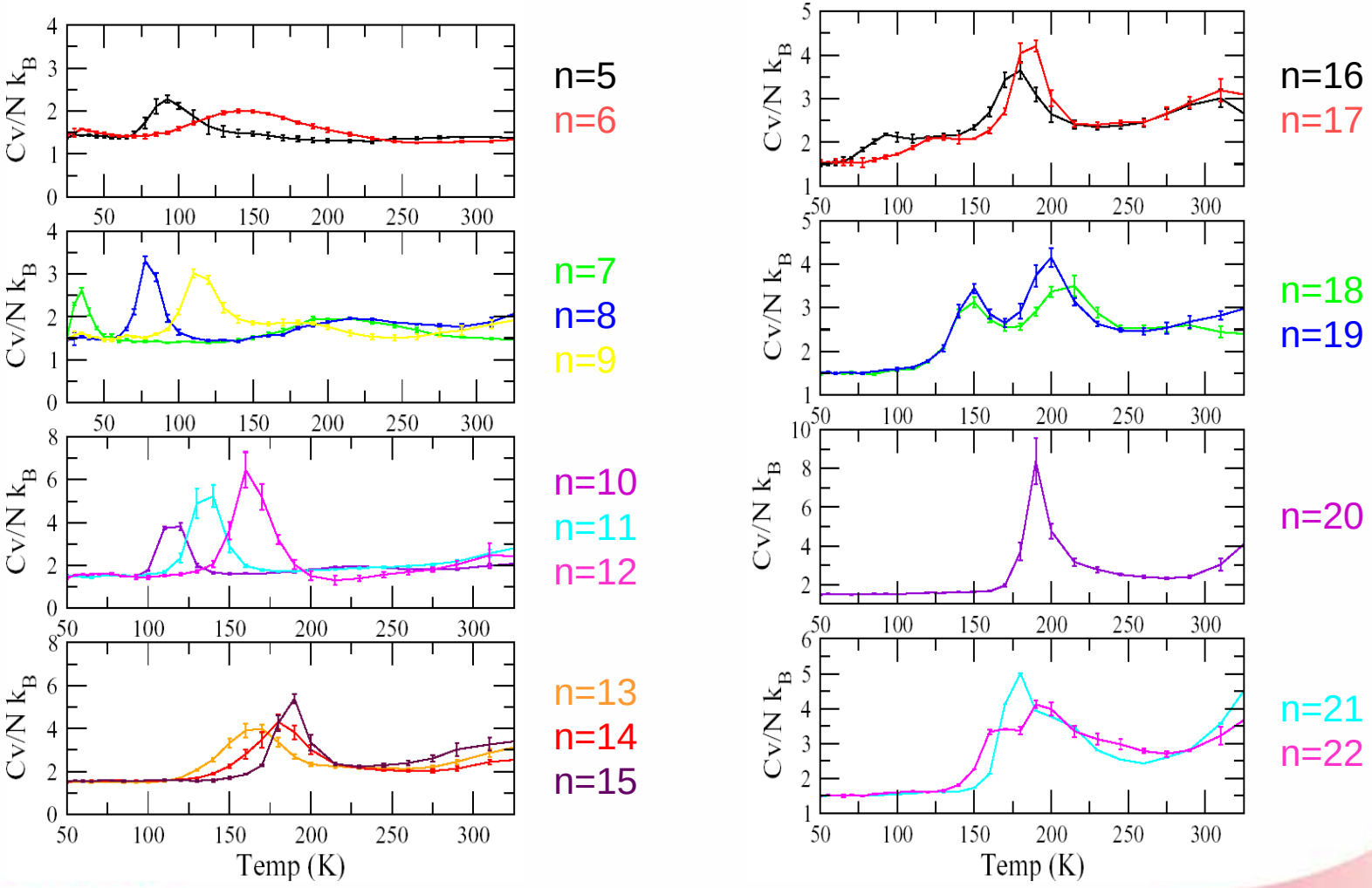
VPS:
(Vibrational Predissociation Spectra)

Tohoku U.
(Prof. Mikami and Prof. Fujii)
Miyazaki *et al., Science* (04)

Yale-Georgia
(Prof. Johnson and Prof. Duncan)
Shin *et al., Sicence* (04),

IAMS, Sinica
(Prof. Yuan Lee and Prof. Chang)
Wu, Lin, *et al., JCP, PCCP* (05)

Thermal behavior of $H^+(H_2O)_n$



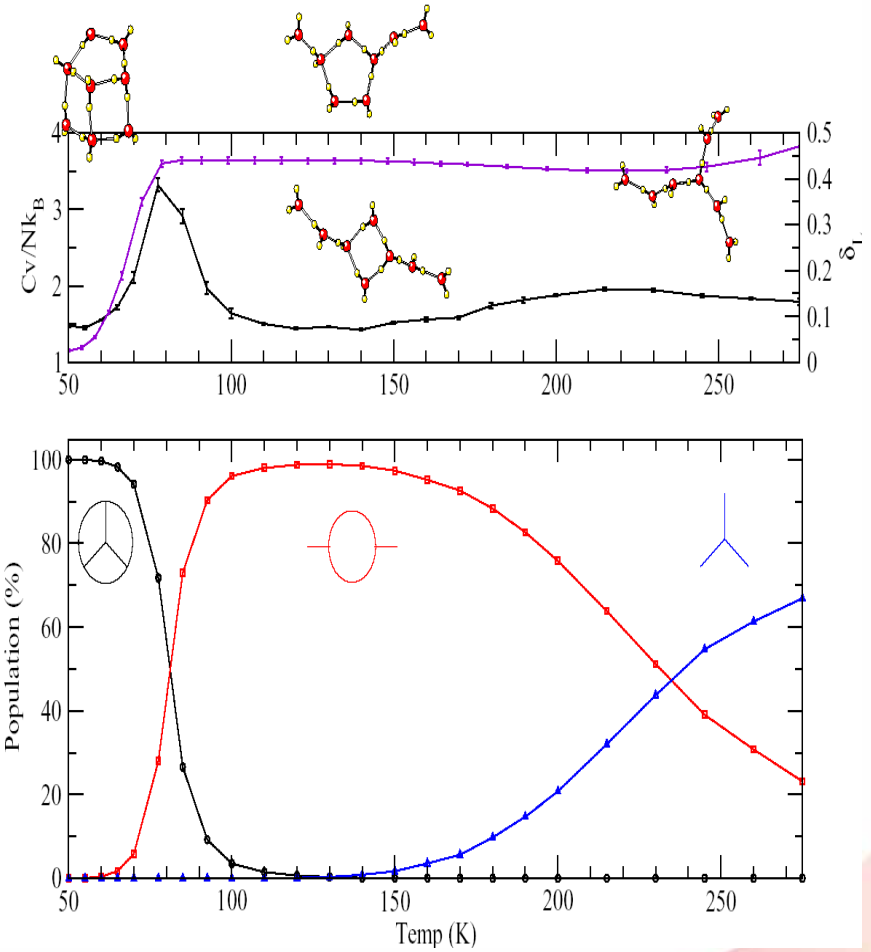
!! Peaks in Cv are associated to structural changes. !!

of Local minimum and why is statistical average important!

$(H_2O)_n$

n	GA TTM2F	re-opt B6	SHS*
4	14	10	
5	33	23	
6	154	95	
7	639	406	
8	2331	1429	(166)*

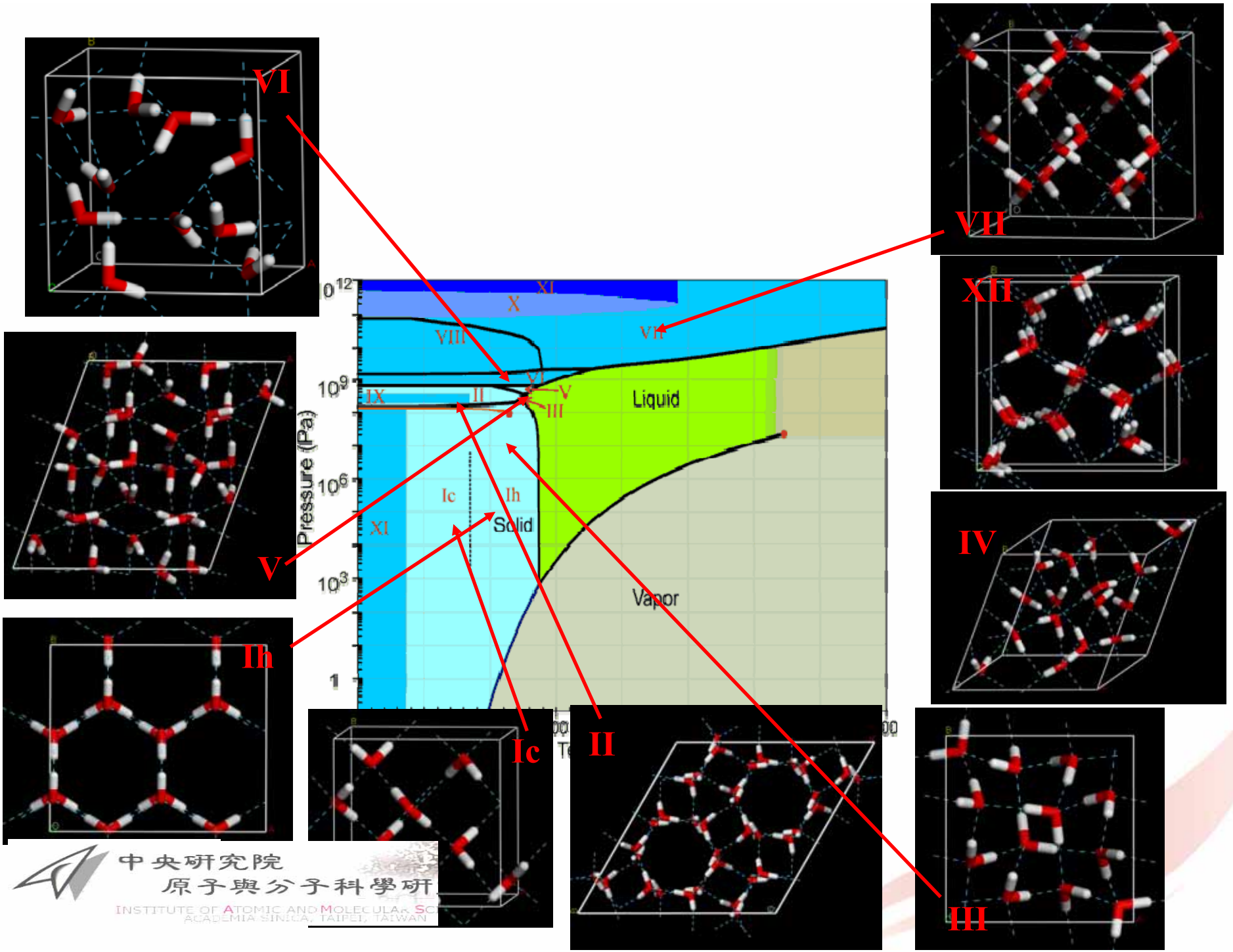
(*) K. Ohno, J. Phys. Chem. A, 111, 10732 (2007)



Dominating structures are the minimum of Free Energy!!

Development of Water Models

- OSS2 potential
  - Designed for $H^+(H_2O)_n$
  - $V_{total} = V_{charge\ interaction} + V_{polarizaton\ interaction} + V_{O\text{-}H\ interaction} + V_{O\text{-}O\ interaction} + V_{three\text{-}body\ interaction}$
  - Polarizable
  - No artificial charge
  - Dissociable ➔ Important in ionic chemistry
  - Overlap of electron clouds modelled by some screening functions



$Be_xZn_{1-x}O$ Alloys : Phase Diagram

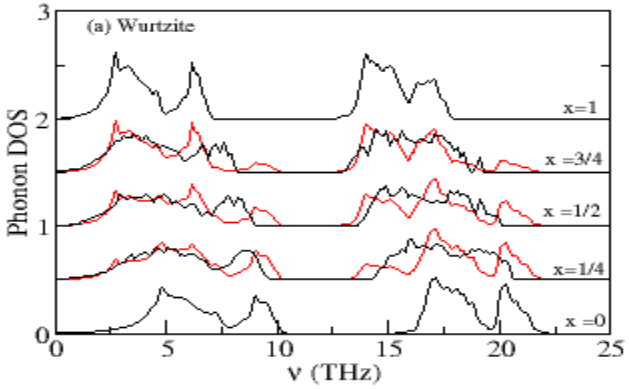
➢ Composite-T Phase Diagram

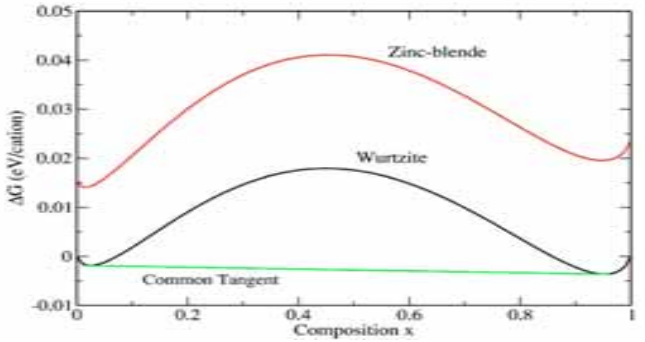
$$\Delta G = \Delta H_f - T\,\Delta S_{conf} + \Delta G_{vib}$$

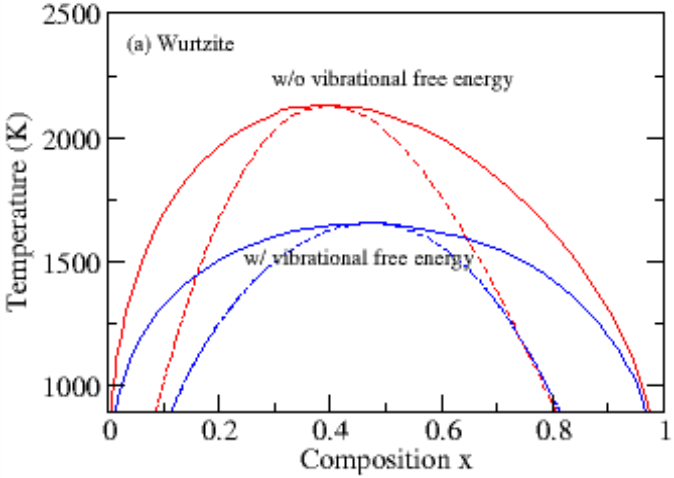
- ΔH_f (formation enthalpy)
- ΔS_{config} : Bragg-Williams app. for random alloys
- ΔG_{vib} : including ZPE

➢ Effect of Lattice Vibration

- PWSCF: linear response good for crystal.
- SQS: (special quasi-random structure)* to represent random alloy
- PHONE: calculate DOS







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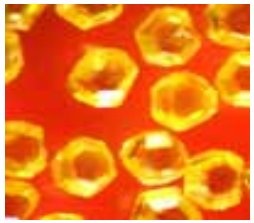
* Zunger, Wei, Ferreira and Bernard, PRL 65 (1990) 353

cubic– BC_xN: 2^{nd} hardest material ??



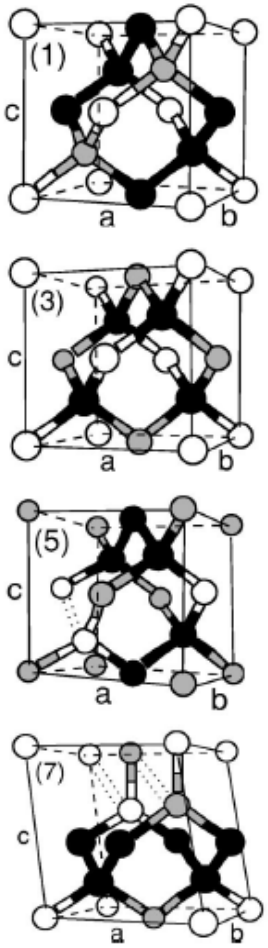
diamond

\+

c-BN

=??

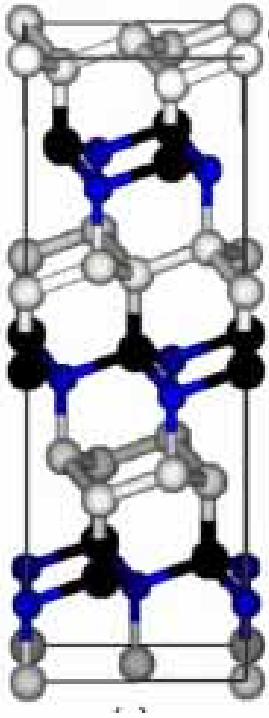
Sun, …Louie, **PRB, 64**, 094108 (2001)



420 alloy config.
(8-atom cubic cell)

↓

7 sym. dist. config.
(8-atom cubic cell)

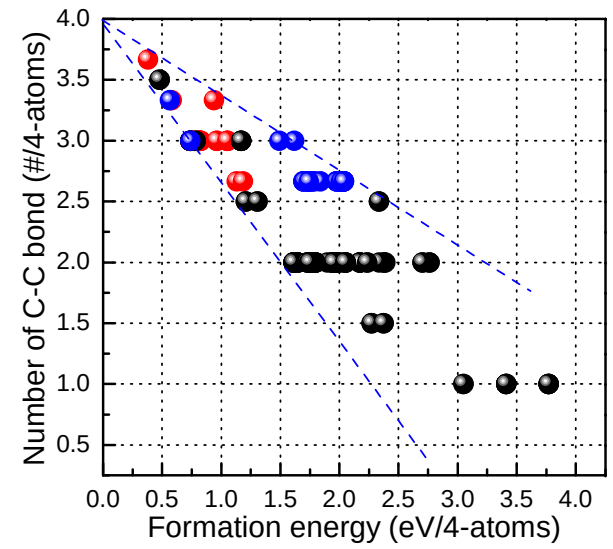


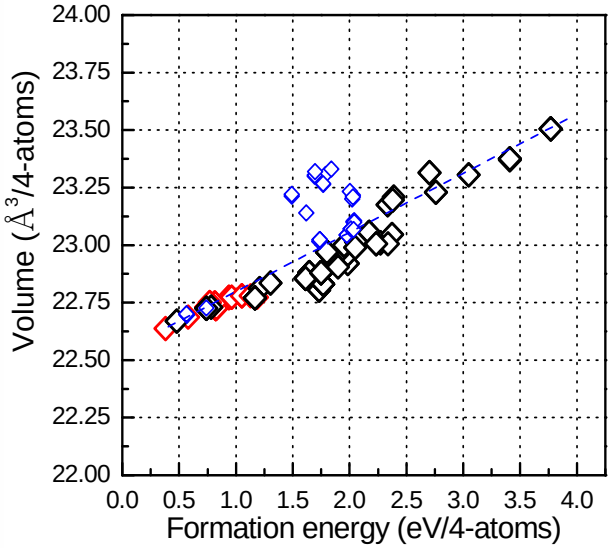
Chen, Gong, and Wei,
PRL. 98, 015502 (2007)

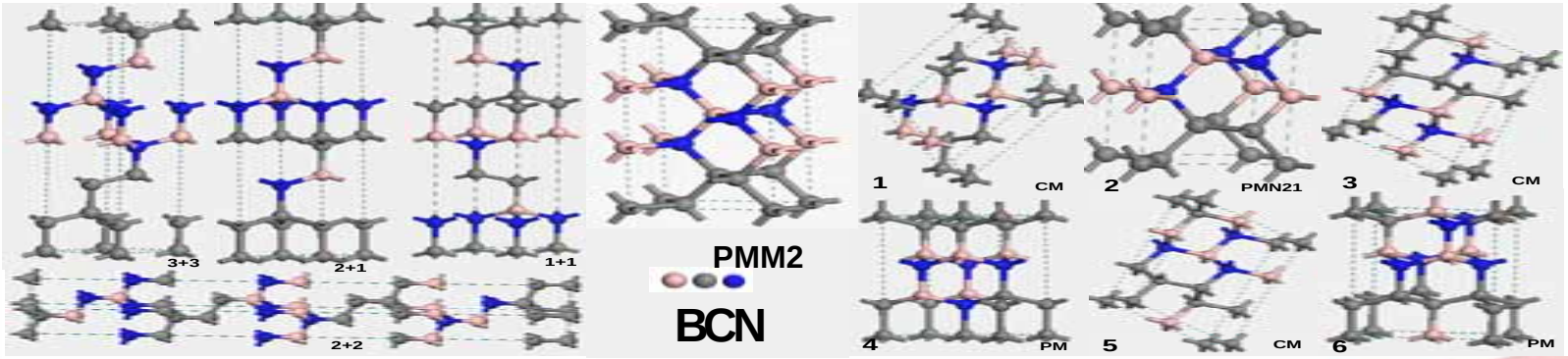
* (111) super-lattices config. are more stable

* Theyare also harder than *c*-BN

cubic– BC_xN: Bond Counting Rules







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