SAND2008-5153P

Abstract

The overall goal of devising a methodology f[illegible] reliably performing *de novo* materials design will be briefly reviewed, as well as ways how to accomplish this task. Thereafter, I will give an overview of various practical issues which are of relevance to the two main-aspects of my research, (i) the variation of chemical structure and composition, and (ii) the associated response of multiple-scale structure property relationships. Efforts, results, and progress made during the first year as a Truman Fellow will be reported and critically discussed. Specifically, I will talk about external and internal collaborations initiated, scientific visits and hosted visitors, conferences attended, acquisitions made, and — most importantly — the scientific work performed so far [1-3]. Eventually, future plans and strategies will be outlined.

[1] O. A. von Lilienfeld and P. Schultz. Structure and Band Gaps of Ga-(V) Semiconductors: The Challenge of Ga Pseudopotentials. Phys. Rev. B, 77:115202, 2008.
[2] A. Tkatchenko and O. A. von Lilienfeld. Popular Kohn-Sham Density Functionals Strongly Over-estimate Many Body Interactions in van der Waals Systems. Phys. Rev. B, 78:045116, 2008.
[3] O. A. von Lilienfeld, et. al., 2008. In preparation.

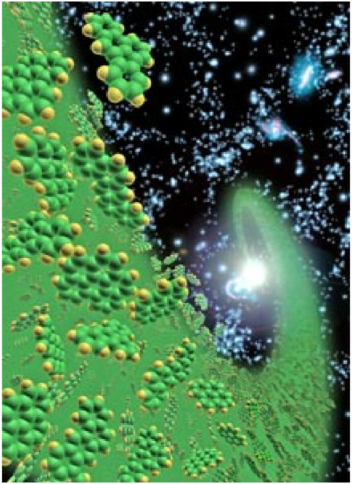
Multiscale schemes for the predictive description and virtual engineering of materials — a 1st year progress report

O. Anatole von Lilienfeld
Multiscale Dynamic Material Modeling Department (1435)
Sandia National Laboratories

Truman Fellow Colloquium, August 5, 2008

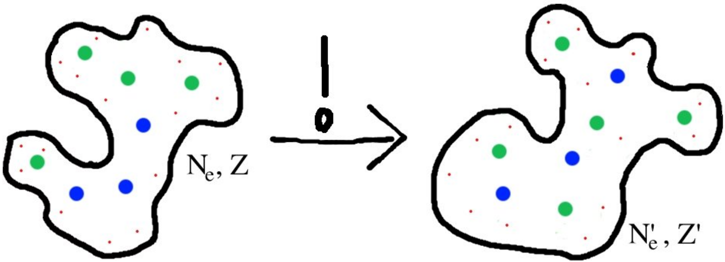
Sandia National Laboratories

Outline - going beyond description



http://www.rsc.org/

Materials design and the philosopher's stone?



- Render material's properties valuable through "alchemical" changes
- Properties of matter defined by composition

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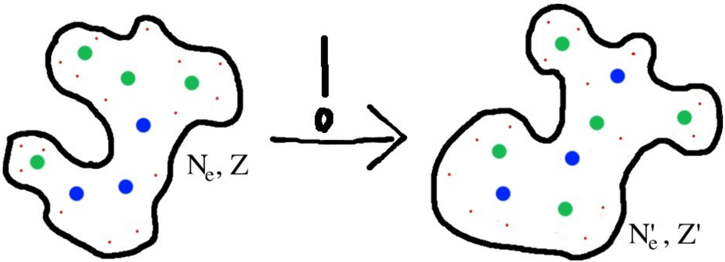

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→ tune properties by variation of matter, *i.e.* composition

≡ optimize properties in "chemical compound space"

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http://www.nystar.state.ny.us/

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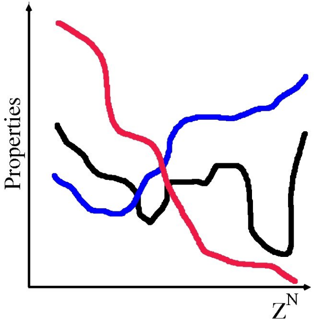

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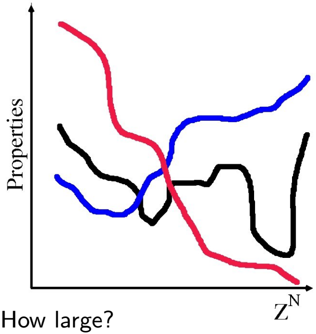

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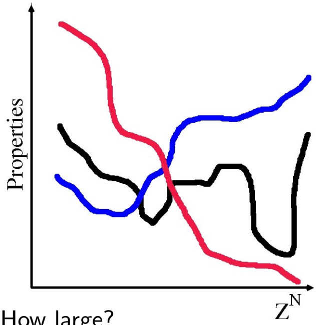

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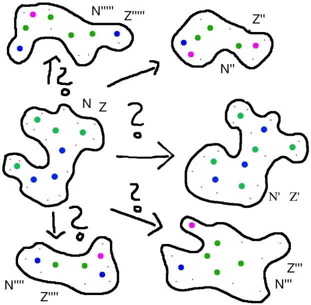

... but how similar?

Compound space — or rather set of compounds?

What is close, what is distant?

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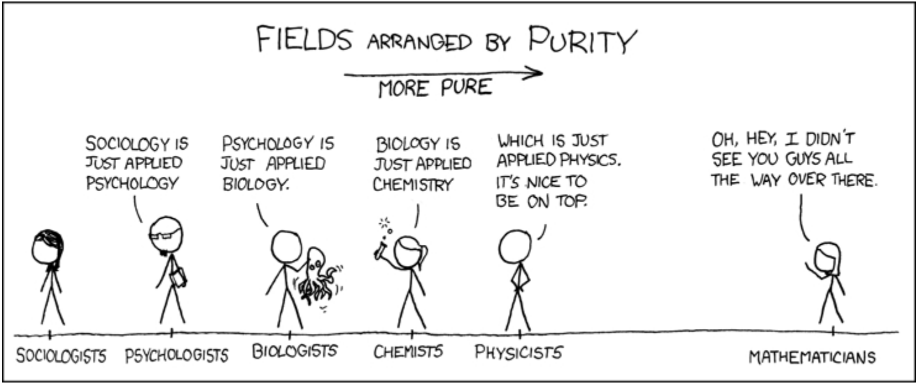
What is close, what is distant?

It depends ...



http://xkcd.com/

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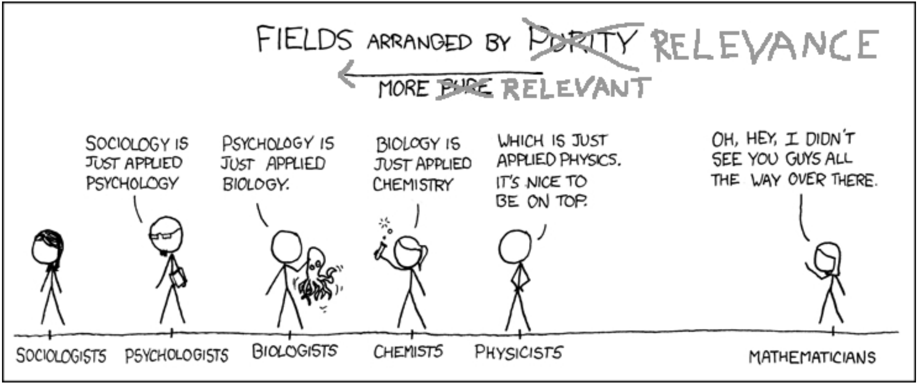

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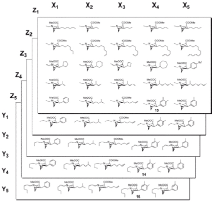
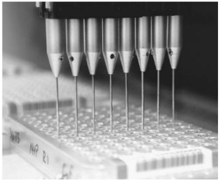
Combinatorial chemistry





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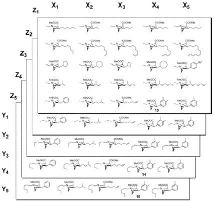
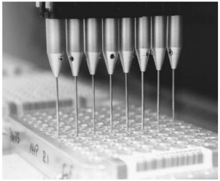
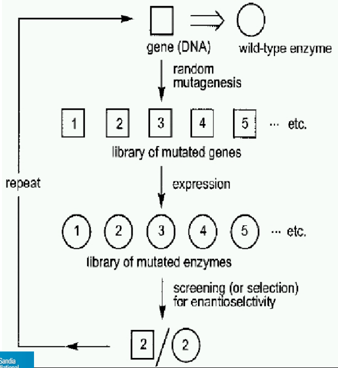




Directed evolution [*PNAS* **101** 5716 (2003)]



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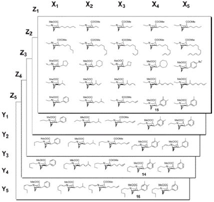

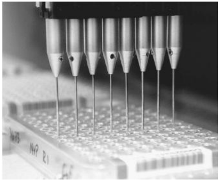

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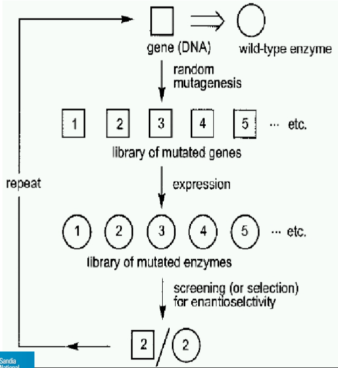
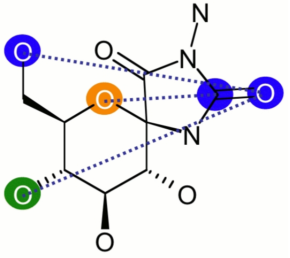


Structure property relationships

scoring functions/descriptors

⇐ Hammett equation (1937)



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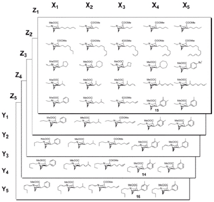

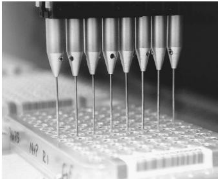

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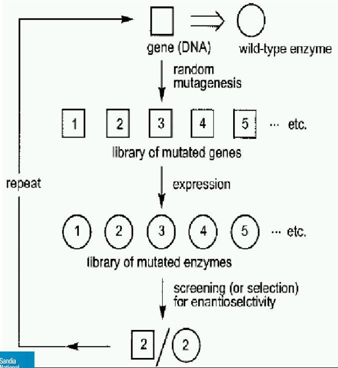

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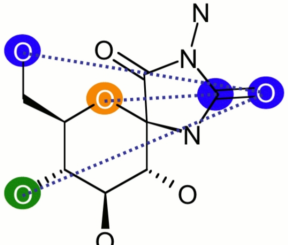
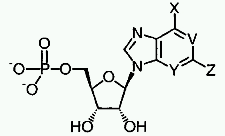


Rational compound design

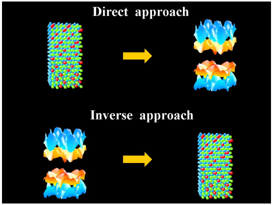
"atomistic screening"



Compounds	X	Y	Z	V
7	NH_2	N	SCH_3	N
8	NH_2	N	CH_3	N
9	NH_2	N	C_2H_6	N
10	NH_2	N	Cl	N

“inverse approaches” — chronological

Alex Zunger (RENL):



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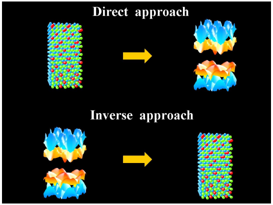

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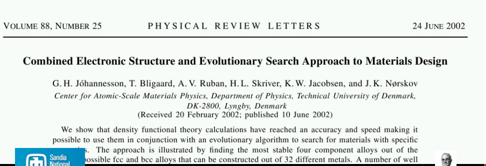

VOLUME 88, NUMBER 25 PHYSICAL REVIEW LETTERS 24 JUNE 2002

Combined Electronic Structure and Evolutionary Search Approach to Materials Design

G. H. Jóhannesson, T. Bligaard, A. V. Ruban, H. L. Skriver, K. W. Jacobsen, and J. K. Nørskov
Center for Atomic-Scale Materials Physics, Department of Physics, Technical University of Denmark, DK-2800, Lyngby, Denmark
(Received 20 February 2002; published 10 June 2002)

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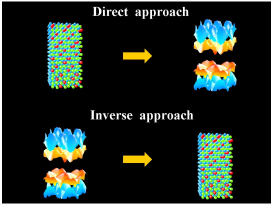

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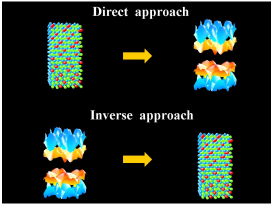

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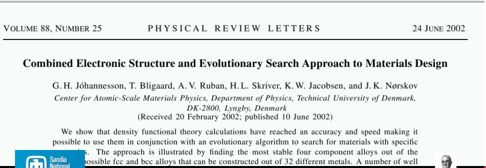
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David Beratan & Weitao Yang (Duke): 'Interpolated potentials' → molecular polarizabilities *JACS* (2006)

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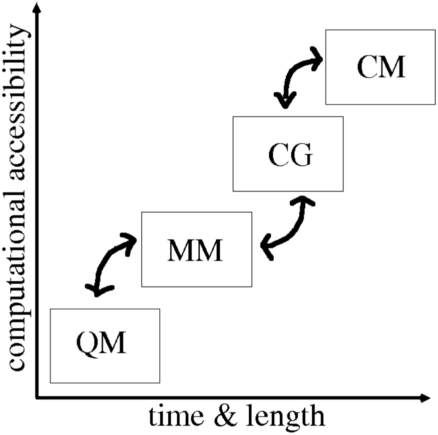
4. test, assess, and apply to materials design, and subsequently forward to synthetic groups
   $\longrightarrow$ Holy Grail (verbatim) FY10

Why accuracy? — error control of structure-property map

map



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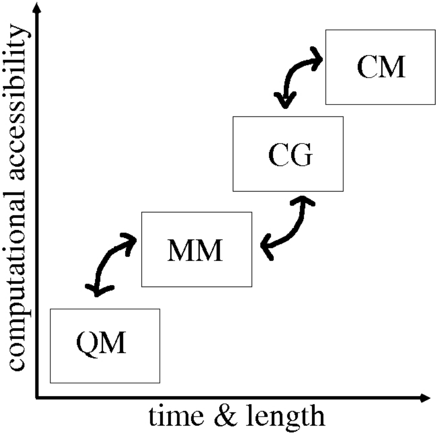
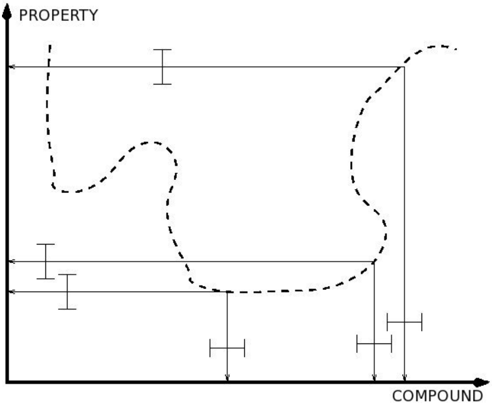


error control



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- subsystem functionals by Armiento & Mattsson (SNL) *PRB* 2002

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Today: intermolecular interactions (bulk) and Fermi level (band-structure)

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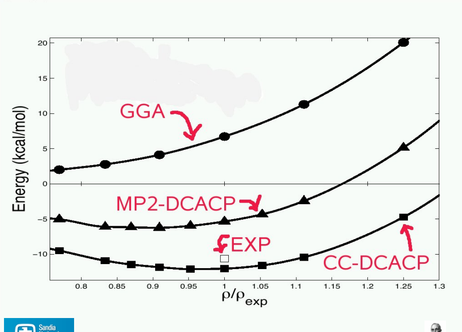
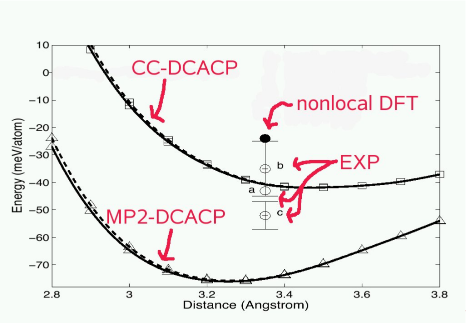
BUT DFT & intermolecular energies problematic:

Benzene crystal



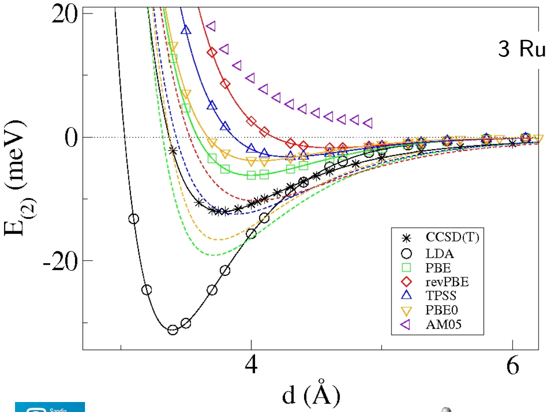
Three graphene sheets



London Dispersion and DFT

Heitler, Eisenschitz, and London, *Z f Phys* (1927, 1930)

$$E[A,B] \;=\; \underbrace{-\frac{C_{6A}C_{6B}}{d_{AB}^{6}}}_{\text{slowest}} \underbrace{-\frac{C_{8A}C_{8B}}{d_{AB}^{8}}-\frac{C_{10A}C_{10B}}{d_{AB}^{10}}+\ldots}_{\text{negligible?}}$$



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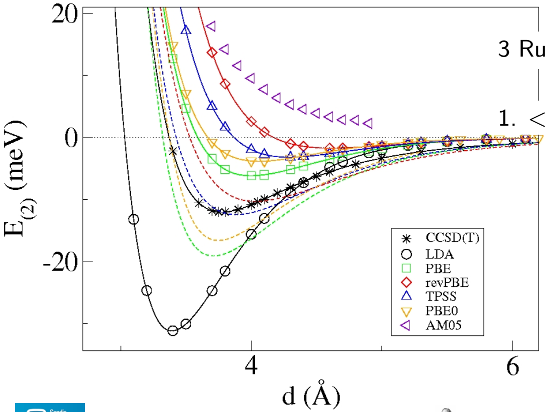

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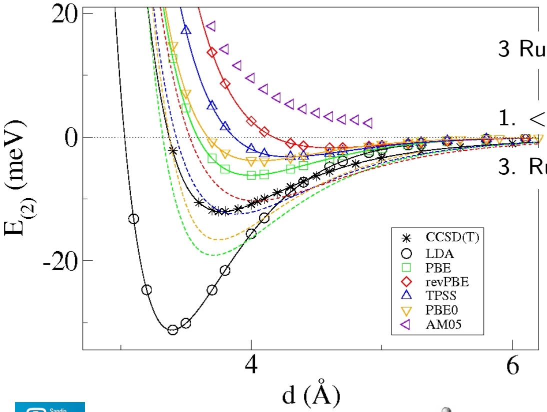

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3. Rung + C_6 deviate $\lesssim 3$ meV
→ sufficient for bulk, such as molecular crystals . . .?

Molecular crystal structure prediction

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'Major Advance in crystal structure prediction' — CSPBT 2007

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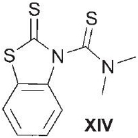
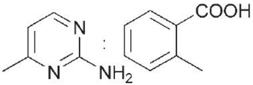
XII

XIII



XIV



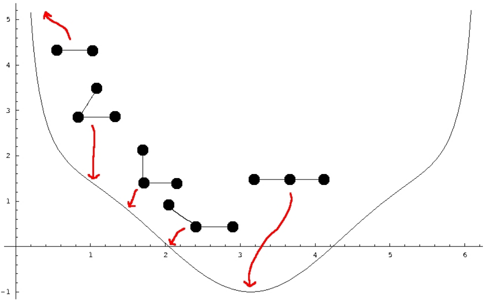
XV

Compd	Hybrid rank[a]	TMFF rank[b]	Hybrid ΔE [kcal mol^{-1} atom^{-1}]
XII	1	1, 37, 38, 69	0.000
	2	7	0.036
	3	13	0.059
XIII	1	2	0.000
	2	4	0.027
	3	3	0.029
XIV	1	1, 8	0.000
	2	9	0.019
	3	19, 25	0.042
XV	1	37	0.000
	2	17	0.015

MBC are significant for bulk — role of 3-body?

Axilrod and Teller *J Chem Phys* (1943)

$$E_{(3)}[A,B,C] = \frac{3\, C_{9A} C_{9B} C_{9C} \times (\cos[\gamma_{AB}]\cos[\gamma_{AC}]\cos[\gamma_{BC}] + 1)}{d^3_{AB} d^3_{AC} d^3_{BC}}$$



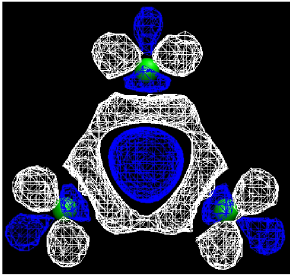
some interesting situations:

1. superimposed ($\gamma \rightarrow 0$): $E \rightarrow +\infty$
2. equilateral triangle ($\gamma = \pi/3$): $E \approx \frac{11}{8} \frac{C_{9A} C_{9B} C_{9C}}{3\, d^3_{AB}}$
3. right triangle ($\gamma = \pi/2$): $E \approx \frac{C_{9A} C_{9B} C_{9C}}{d^3_{AB} d^3_{AC} d^3_{BC}}$
4. linear ($\gamma = \pi$): $E \approx -\frac{C_{9A} C_{9B} C_{9C}}{d^3_{AB} d^3_{AC} d^3_{BC}}$

MBC are crucial for bulk — 3-body & DFT?

$$E_{(3)} = E^{trim} - 3E_{(1)} - 3E_{(2)} = E^{trim} - 3E^{dim} + 3E_{(1)}$$

Ar_3 in the equilateral triangle



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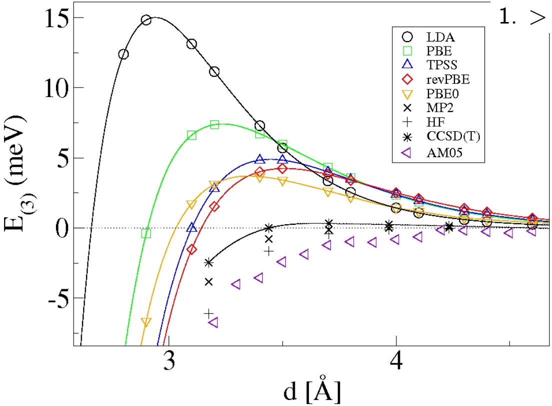
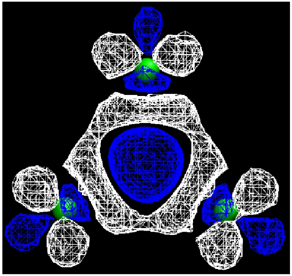


1. > 2. > 3. ≈ X ≈ EMP > REF > n-GGA



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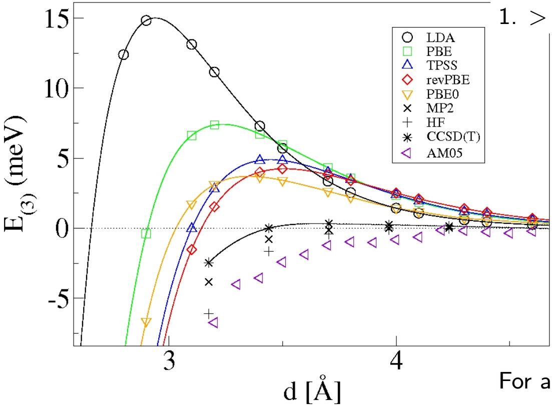

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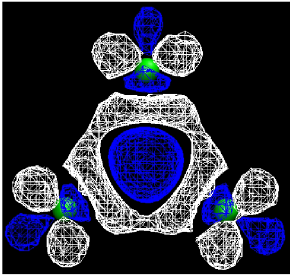

For all but n-GGA C_9 would worsen things

… and bulk?

Bulk's MBC & DFT?

$$E_{MBC} = E_{coh} - E^{(2)}_{coh}, \qquad E^{(2)}_{coh} = \frac{1}{N}\sum_{B} E_{(2)}(d_{AB})$$

A Tkatchenko and OAvL *PRB* **78** 045116 (2008)

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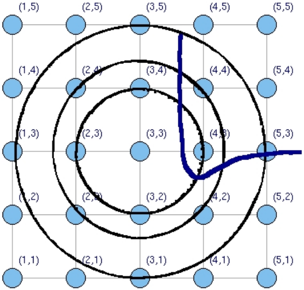

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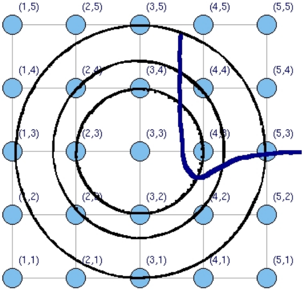

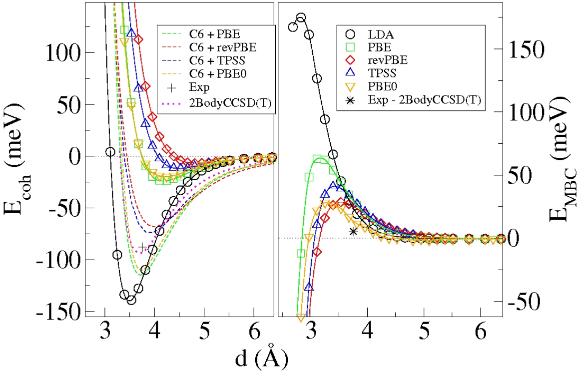

A Tkatchenko and OAvL *PRB* **78** 045116 (2008)

Excited states are related to band-gap

Effect of functional and pseudopotentials on band-gap

Within DFT, pseudopotentials can account for

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- self-interaction errors (Baumeier et al. *PRB* (2006)
- exact exchange (OAvL et al. *JCP* (2005))
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Can one improve upon band-gap?

YES (Christensen *PRB* (1984), Segev et al. *PRB* (2007))

GaN, GaP, GaAs — industrially relevant semi-conductors whose band-gap is crucial to determine effects of defects

Excited states are related to band-gap

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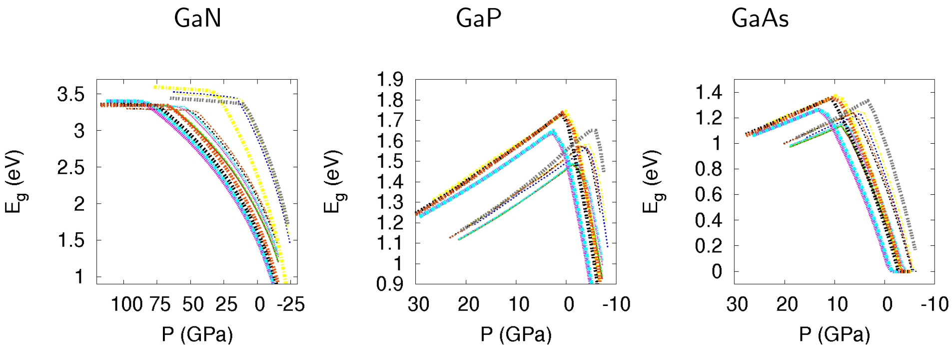

OAvL and P Schultz *PRB* **77** 115202 (2008)

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xi Alchemical design of molecular conductors (I-Chun Lin and Koichi Yamashita, Tokyo University)

Conferences, visits, visitors

- Long Range Interactions in Nanoscale Science, October 2007, Annapolis, sponsored by the Council for the Division of Materials Sciences and Engineering (DMS&E). DMS&E is one of the research divisions in the Office of Basic Energy Sciences of the U.S. Department of Energy (DOE)
- 3 weeks visit group of Prof. Artem Oganov, ETH Zürich (crystal structure prediction), Nov 2007
- 3 weeks visit of Prof. Scheffler's theory department at FHI Berlin (3-body study with Alexandre), Dec 2007
- hosted 8 weeks visit of Matteo Guglielmi, (Jan & Feb 2008)
- American Physical Society meeting, March 2008, New Orleans
- Institute of Pure and Applied Mathematics, UCLA, reunion of Fall 2005 long-term program 'Bridging Time and Length Scales in Materials Science and Bio-Physics', CA, Jun 2008
- 1 week visit Prof. Koichi Yamashita at Tokyo University (molecular conductor), Jun 2008
- Meeting of the World-Association of Theoretically Oriented Chemists 2008 (WATOC08), Sydney AU, Sep 2008
- Various stays with Prof. Mark Tuckerman's group at NYU

¿ $$$? — acquisitions, contracts, students, metrics

- asgard — Desktop, quad-core Intel, 3 TB hard drive, 1 GB memory, linux, SRN
- frigg — Rack Compute node, 4×quad-core Intel, rack compute node, 0.5 TB hard drive, 128 GB memory, linux, SON
- TURBOMOLE — 'one of the fastest and most stable codes available for standard quantum chemical applications'
- postdoctoral researcher at NYU — 3 months of salary
- three PhD-students hosted at SNL — each for 3 months
- travel & collaborations
- http://www.cs.sandia.gov/~oavonli
- OAvL and P Schultz *PRB* **77** 115202 (2008), A Tkatchenko and OAvL *PRB* **78** 045116 (2008)

Overview

	goals	systems	SNL	needs
1^{st} Yr	accurate DFT	crystals	*ab initio*	collaborations, `software`
2^{nd} Yr	multiscaling & GCE	T_m, reactions	realistic reactive	more man-power
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⇒ opportunity to assemble pieces to accomplish major breakthrough

Acknowledgments

Mentor Ann Mattsson

Manager John Aidun

SNL Bob Bradshaw, Peter Feibelman, Harold Hjalmarson, Kevin Leung, Thomas Mattsson, Richard Muller, Steven Plimpton, Susan Rempe, Peter Schultz, John Shelnut, Nathan Siegel, Aidan Thompson

1435 Multiscale Dynamic Material Modeling Department

CSRI Deanna Ceballos (SMLS) and Vonda Coleman (OAA); Bill Goldman and Roger Retal (CSUs)

$$$ US-NSF (IPAM workshop at UCLA), DOE (LRI workshop & Solar Energy), Tokyo University (visit),

LDRD: Truman Fellowship from Sandia National Laboratories

Yolanda Moreno (LDRD); Wendy Cieslak (VP 1000); Ilke Arslan, Whitney Colella, Darin Desilets, Jacques Loui, Gregory Nielson, David Scrymgeour (fellow Fellows)

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