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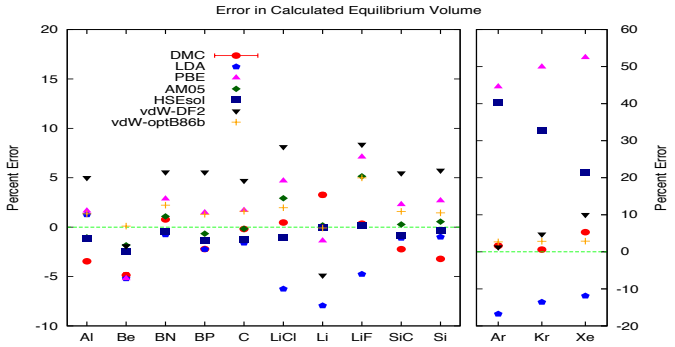

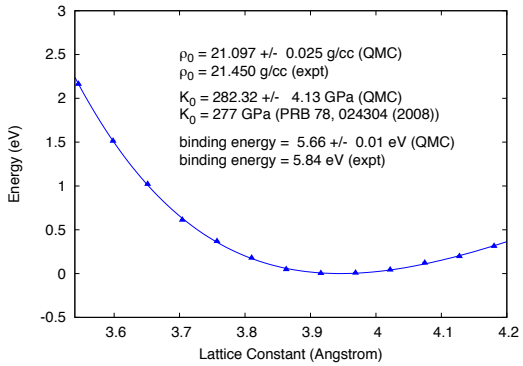

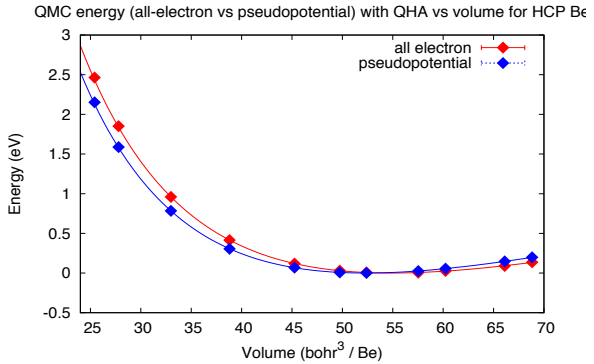

Advances in the application of diffusion Monte Carlo to solids

L. Shulenburger and T. R. Mattsson

Thursday, March 6, 2014, Session S27, 8:36 AM

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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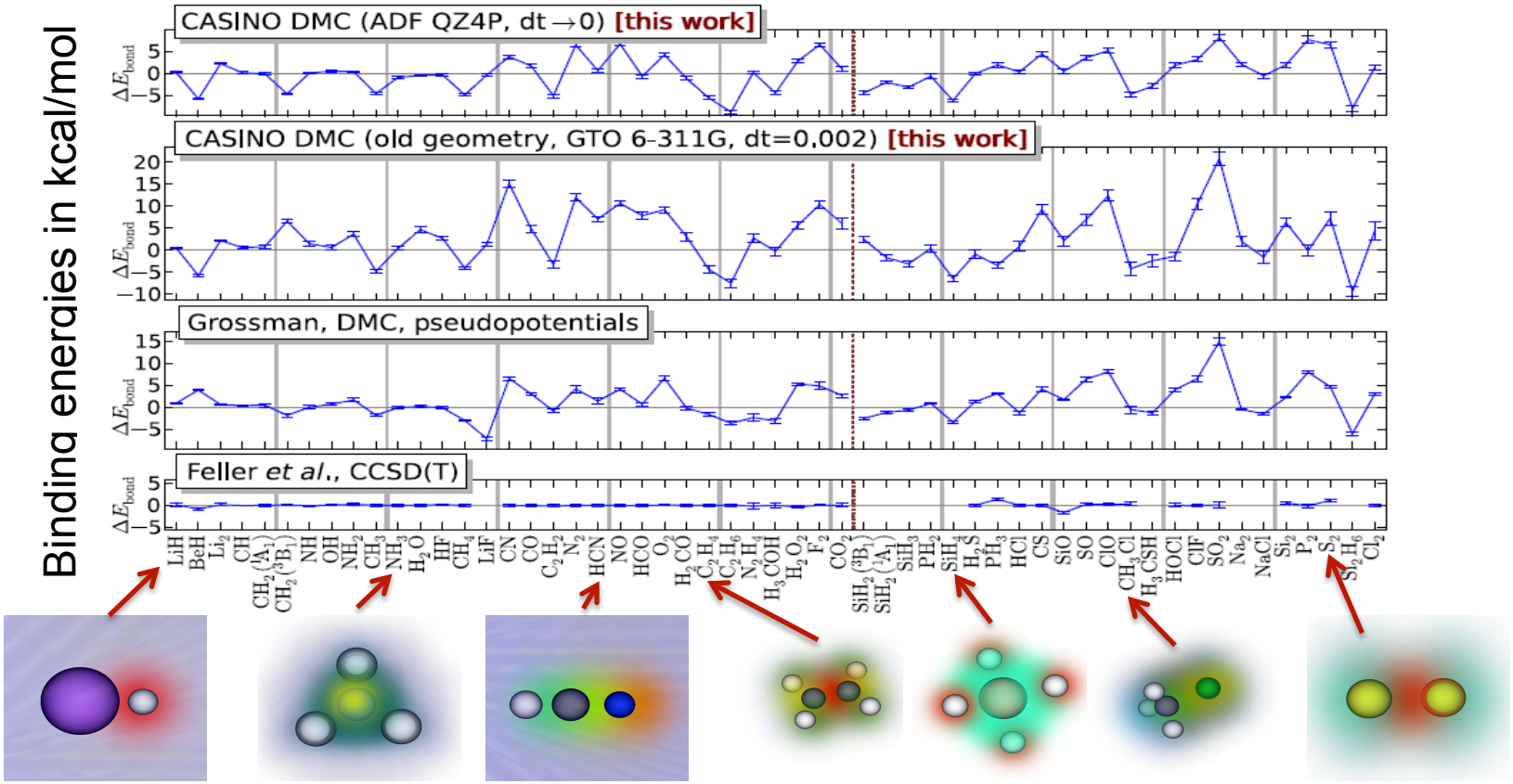
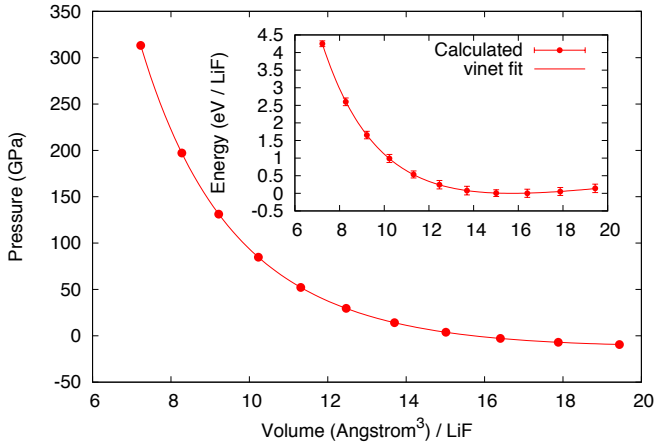
DMC has been extensively benchmarked for molecular systems



•from Nemec et al, JCP. **132**, 034111 (2010)

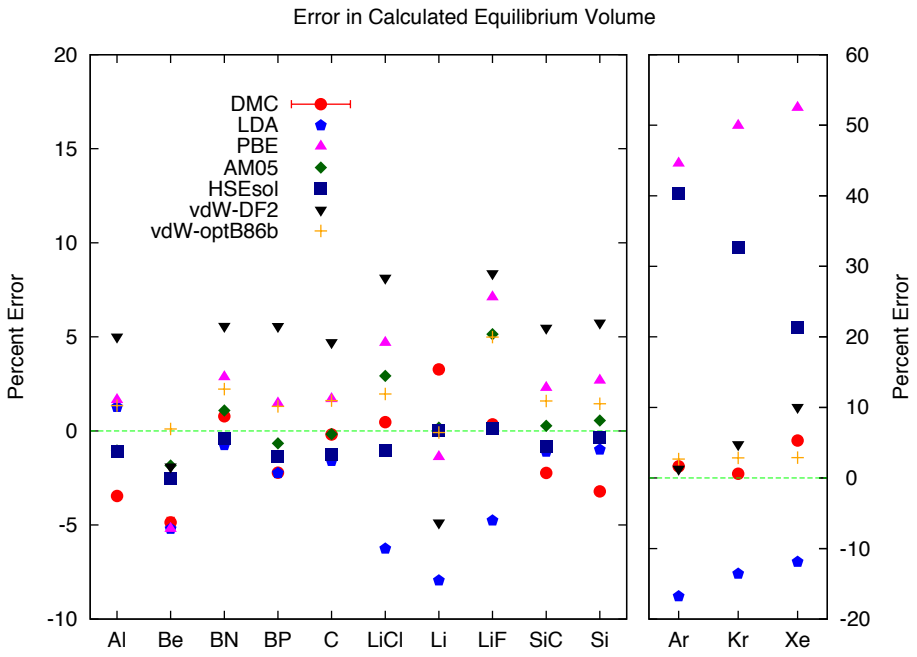
We conducted the first extensive benchmarks of DMC on condensed matter

- Test compares to easily measured experimental data
  - high pressure calculations to derive properties of ambient phase
- Previous calculations have required 1 year of time on NSF machines for a single solid
- Choice of systematic approximations can greatly affect results
- Calculations performed on Cielo



We conducted the first extensive benchmarks of DMC on condensed matter

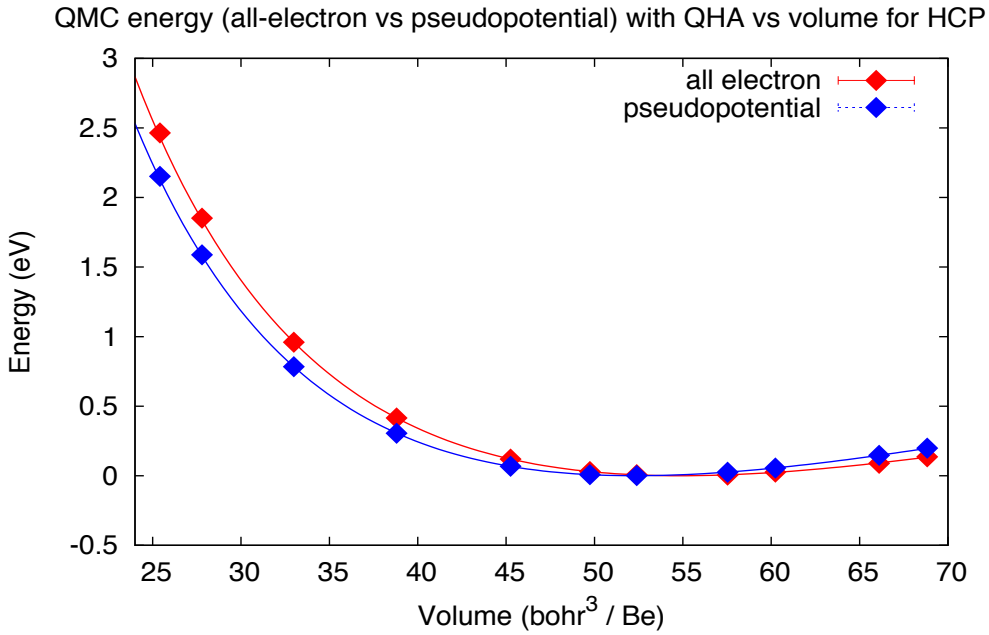
- **Fit Vinet form to E(V) and compare equilibrium volume (density) and bulk modulus (compressibility) to experiment**



- Materials span a factor of 10 in equilibrium volume
- Four types of bonding are included
  - Ionic
  - Covalent
  - Metallic
  - Van der Waals
- Lattice Constants within ~0.9%
- This provides a new baseline procedure for a QMC calculations
- PRB 88, 245117 (2013)

Mean error: -0.38 +/- 0.15
Mean absolute error: 2.28 +/- 0.15
RMS error: -0.697 +/- 0.066%
Mean absolute relative error: 1.79 +/- 0.07%

Isolate pseudopotential errors by performing all electron DMC calculations

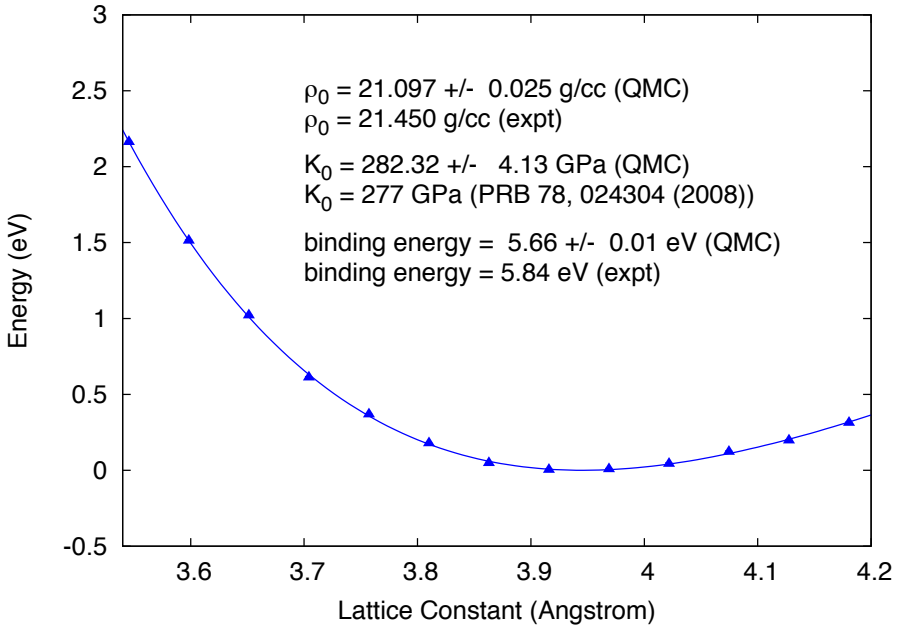


- Previous study concluded that dominant error came from pseudopotentials
- Study ambient phase of beryllium
- Replace pseudopotential with coulomb potential for QMC
- All properties of HCP (ambient) phase agree with experiment

HCP Equilibrium Parameters			
	QMC	All Electron QMC	Exp
c/a	1.569 +/- 0.004	1.569 +/- 0.004	1.568
V_0 (angstrom^3)	7.746 +/- 0.078	8.123 +/- 0.006	8.117
Bulk Modulus (GPa)	124 +/- 2	115.7 +/- 1.5	116.8

Revise pseudopotential generation scheme and apply to heavier elements

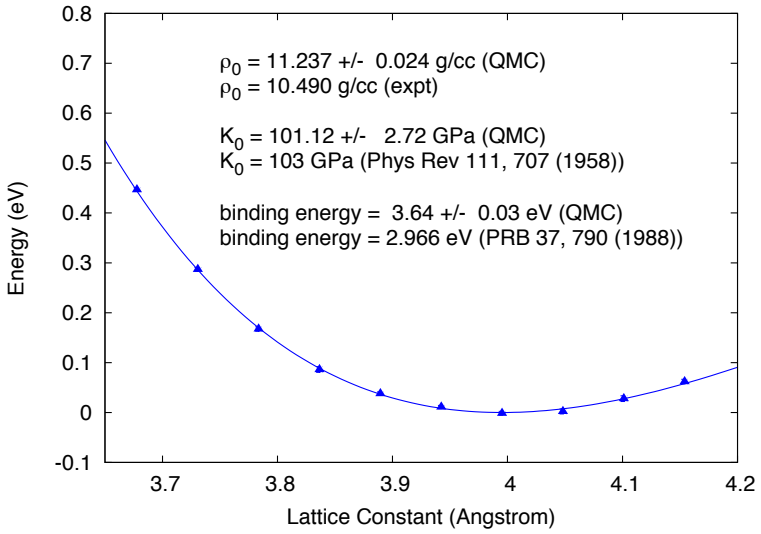
- Still require DFT based pseudopotentials to accurately reproduce all electron results
- Attempt to reduce size of locality error by making nonlocal channels similar to local
- Preserve Kleinman-Bylander form for DFT, but allow change of local channel for DMC
- Choose core-valence separation based on separation in energy



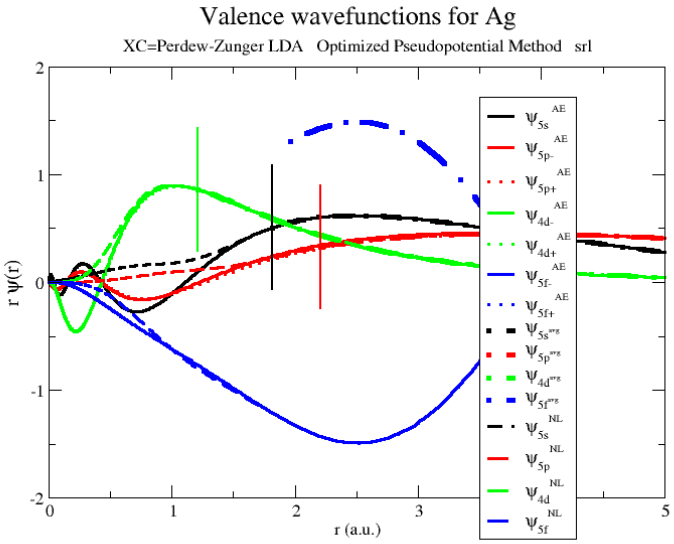
Application to FCC platinum yields encouraging results for ambient density, bulk modulus and cohesive energy

Unfortunately this method does not appear to be a silver bullet

- Elastic properties well reproduced
- Ambient density off by ~7%
- Variance of energy and timestep error are small
  - Wavefunction appears to be well matched to pseudopotential



- Consider strong spatial overlap of 4d with 5s and 5p wavefunctions



DMC is promising for condensed matter, but pseudopotentials remain a large source of uncertainty

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- Results of systematic study on solids show good but not perfect accuracy for solids of light elements
  - PRB 88, 245117 (2013)
- All electron calculations suggest single Slater-Jastrow form is not limiting for many solids
- Application to heavier elements meets with mixed success

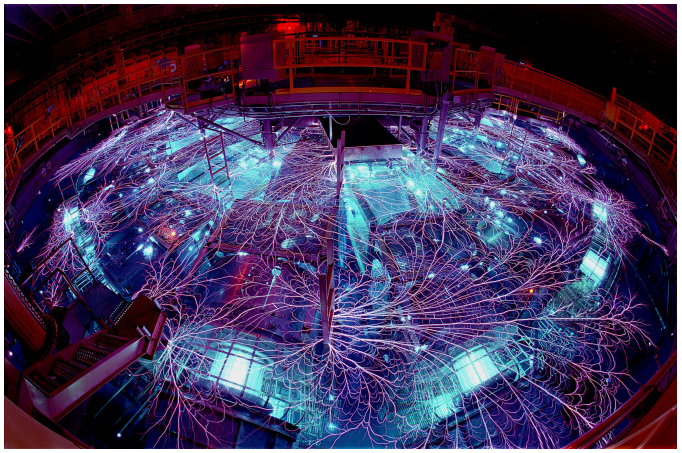
- Study new pseudopotential generation procedures
  - Many body approach of Trail and Needs JCP 139, 014101 (2013)
- Study many body tests of pseudopotentials
  - Excitation energies of atoms (need better wavefunctions?)
  - Bond lengths of small molecules

Acknowledgments

- Jeongnim Kim
- Kyle Cochrane
- Mike Desjarlais
- DOE BES
- Sandia high performance computing and ACES

Postdoctoral opportunity

- High energy density plasma physics department
- Use electronic structure techniques to predict properties of matter under extreme conditions
  - Density Functional Theory
  - Quantum Monte Carlo
- Strong possibilities for collaboration with experimentalists using the Z machine
- Excellent access to some of the largest computational resources in the world
- Competitive Salary
- www.sandia.gov/careers/students_postdocs/postdocs.html
  - Job ID: 645510
  - Search 1641



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