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Effects of pressure on the magnetic properties of FeO: A diffusion Monte Carlo study

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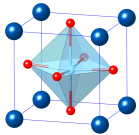
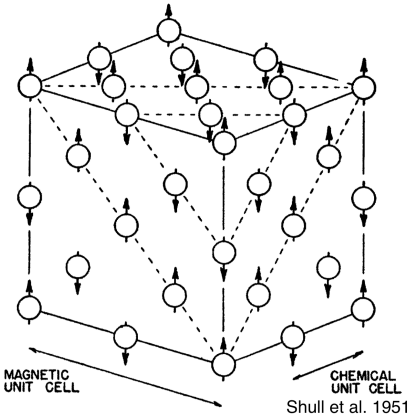
SAND 600376.

CPSFM | Center for Predictive Simulation of Functional Materials



Thanks to the Center for Predictive Simulation of Functional Materials, DOE BES Computational Materials Sciences program. http://cpsfm.ornl.gov/

Antiferromagnetic B1 FeO



Shull et al. 1951

FM ordering of Fe on [111].

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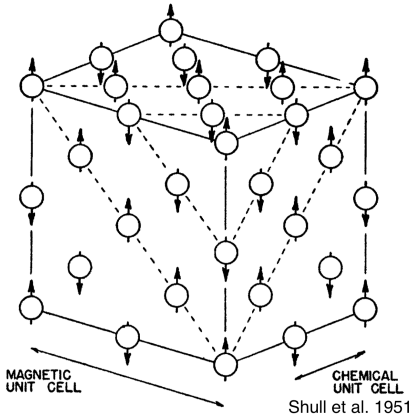

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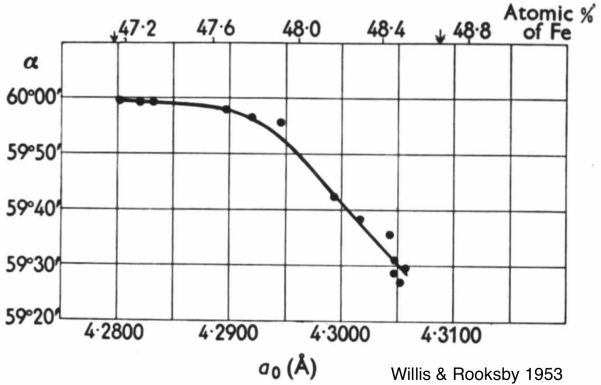

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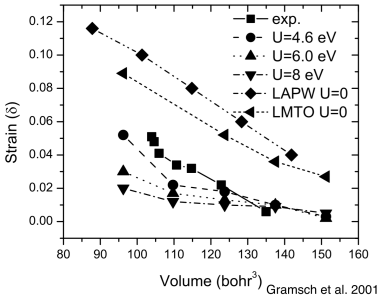
FM ordering of Fe on [111].

Strain depends on composition.

Previous work and current research goals

Previous DFT investigated strain, but limited XC functionals[†].



Gramsch et al. 2001

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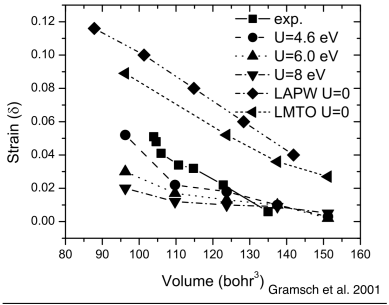

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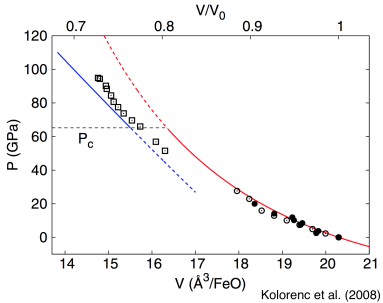

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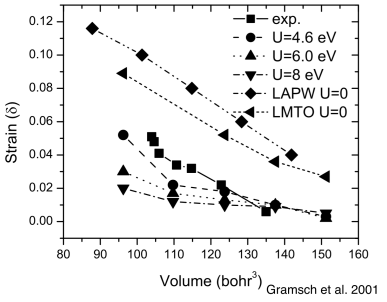
Revisit FeO using DFT + QMC and look at equilibrium strain.



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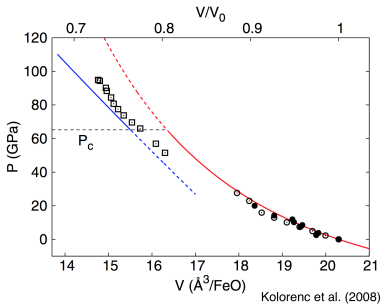

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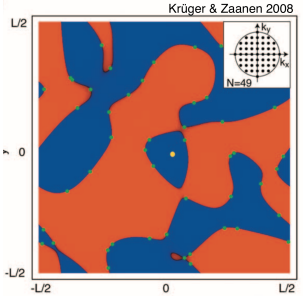
Computational approach

Pseudopotentials Hard NCPP's[1] suitable for QMC calculations.

ESPRESSO SCF calculations for B1 AFM FeO strained primitive cells with different XC functionals.

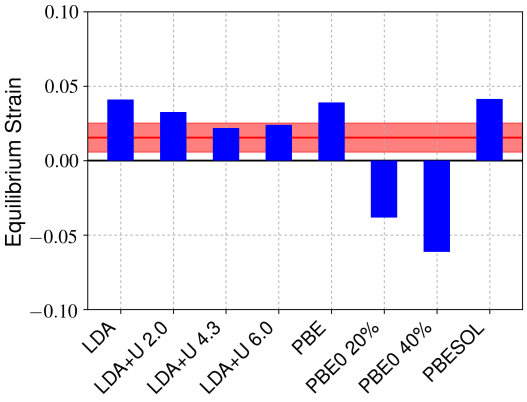
QMCPACK VMC optimization and FNDMC using DFT trial wave functions.

$$\Psi^{QMC} = \begin{vmatrix} \phi_1(\mathbf{r}_1) & \phi_1(\mathbf{r}_2) & \dots & \phi_1(\mathbf{r}_N) \\ \phi_2(\mathbf{r}_1) & \phi_2(\mathbf{r}_2) & \dots & \phi_2(\mathbf{r}_N) \\ \vdots & \vdots & \ddots & \vdots \\ \phi_N(\mathbf{r}_1) & \phi_N(\mathbf{r}_2) & \dots & \phi_N(\mathbf{r}_N) \end{vmatrix} e^{J(\{\mathbf{r},\mathbf{R}\})}$$

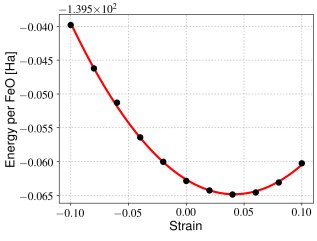


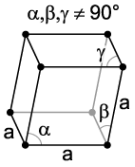
[1] Krogel et al. 2016 PRB. 10.1103/PhysRevB.93.075143

DFT results



$$\epsilon = 1 - \alpha/60$$

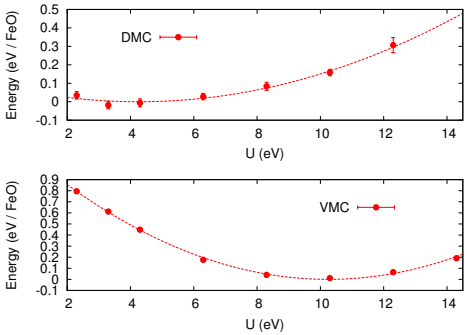




$E^{cut} = 150$ Ha and 5x5x5 k-point mesh.

Expt. data from Willis & Rooksby 1953, Fjellvåg et al. 1996, Battle & Cheetham 1976.

AFM FeO nodal surface is very sensitive



DMC and VMC yield different U's (Benali et al. 2016 10.1039/c6cp02067d).

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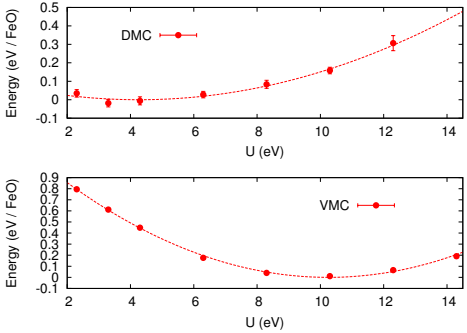
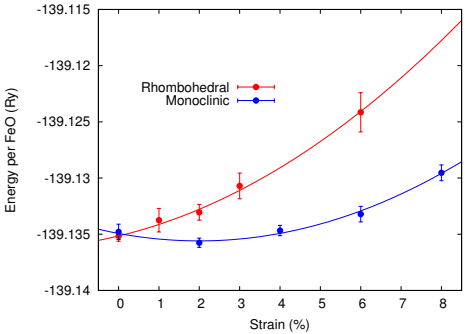
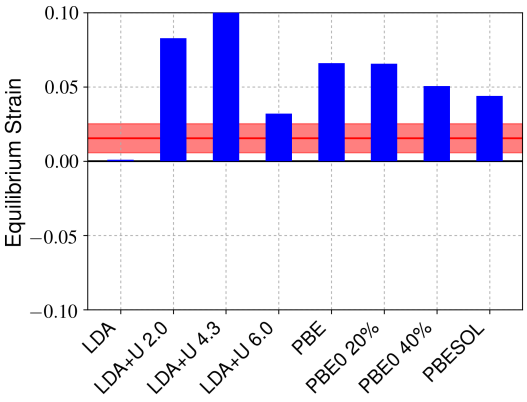




DMC and VMC yield different U's (Benali et al. 2016 10.1039/c6cp02067d).

Equilibrium DMC strain sensitive to nodal surface.

Preliminary FNDMC results



✓ Correct sign of strain for all XC's.

Expt. data from Willis & Rooksby 1953, Fjellvåg et al. 1996, Battle & Cheetham 1976.

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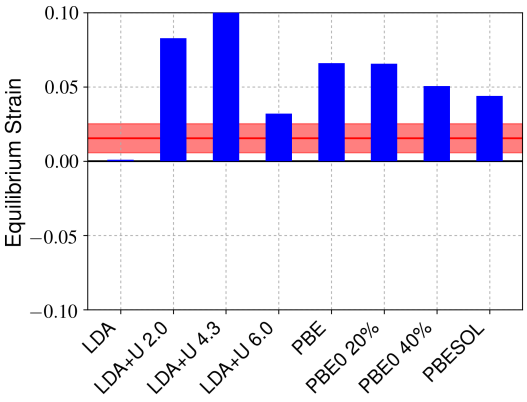

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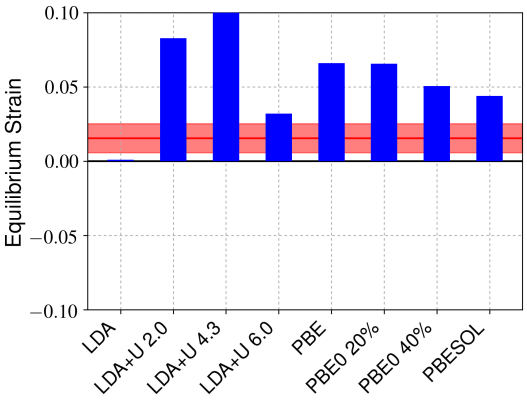

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"Best" LDA, LDA +U 6.0, PBESOL

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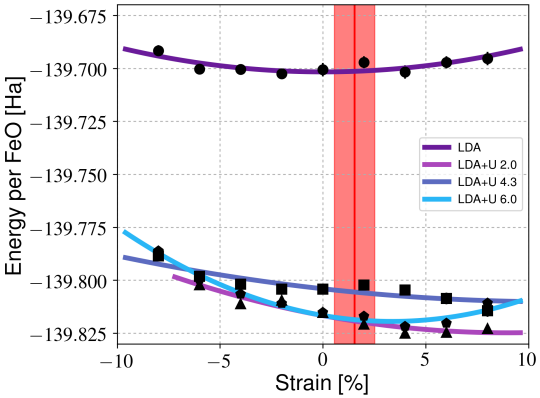
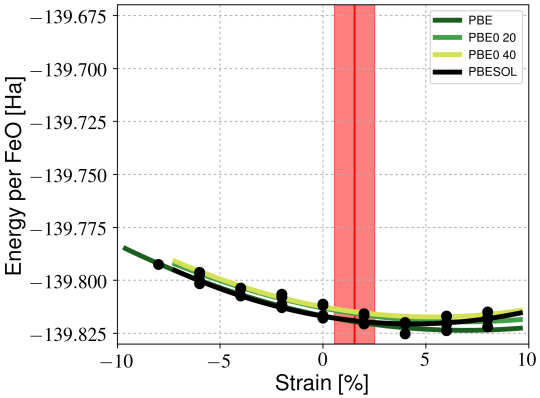
Preliminary FNDMC Results





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Progress thus far

At the DFT level, LDA +U seems to do well on strain.

DMC favors higher U than DFT.

DMC correctly predicts the sign of the strain for all XC's tested.

At the DMC level, LDA +U 6.0, PBE, and PBESOL lowest energy.

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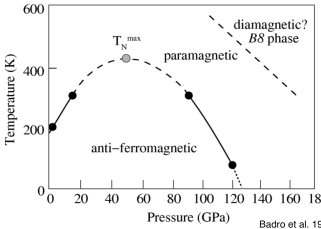
Ongoing Research Questions:

Estimate band gap under ambient conditions w/ DMC.

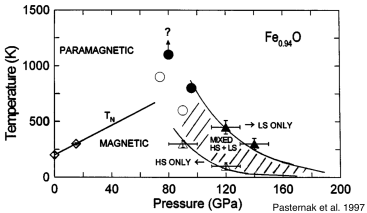
Investigate strain evolution at high pressure.

Look at the phase transition to iB8/B8.

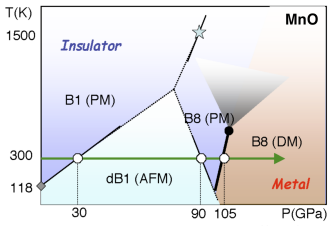
Predict MIT pressure.



Badro et al. 1999



Pasternak et al. 1997



Yoo et al. 2005

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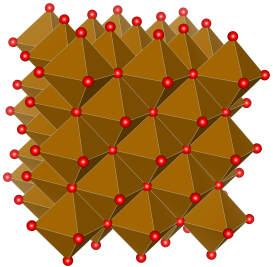
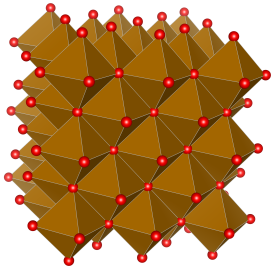
Thank you!

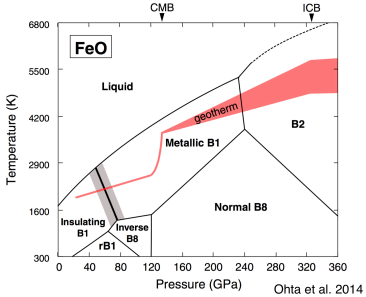
An important material with interesting properties



Rich phase diagram: electronic, magnetic, structural transitions.

An important material with interesting properties





Ohta et al. 2014

Rich phase diagram: electronic, magnetic, structural transitions.

Prototypical Mott insulator. LDA predicts metallic state.

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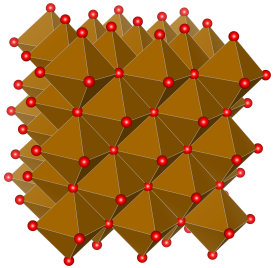

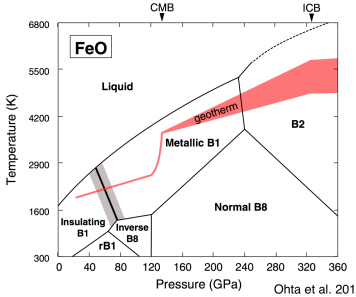

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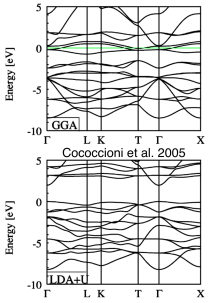

Cococcioni et al. 2005

Rich phase diagram: electronic, magnetic, structural transitions.

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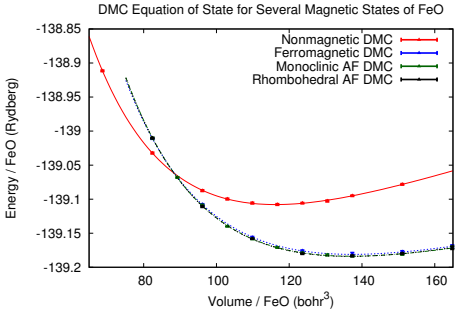
Natural samples are non-stoichiometric - $Fe_{1-x}O$, $x \approx 0.01 - 0.1$.

Comparing equations of state



EOS Parameter Comparison*:

Study	K_0 [GPa]	K_0' -	a_0 [Å]
Unstrained QMC	179(11)	4.8(5)	4.342(10)
Strained QMC	165(6)	4.7(3)	4.343(8)
Kolorenc QMC[1]	170(10)	5.3(7)	4.324(6)
Isaak LDA[2]	173	4.2	4.136
Fisher expt.[3]	149(1)	3.60(4)	4.334
McCammon expt.[4]	152	4.92	4.334

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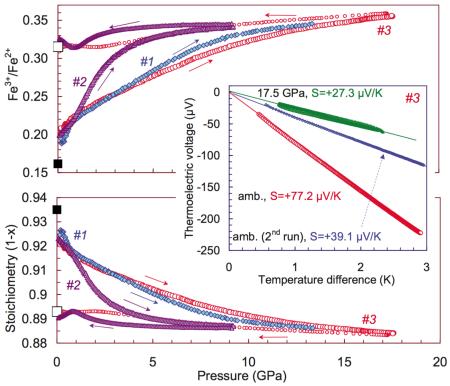

Ovsyannikov et al. 2010

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*Comparing EOS parameters is tricky! Experiments are non-stoichiometric, and everyone uses a different functional form. Additionally, there is some evidence that wüstite becomes more nonstoichiometric under pressure!