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Effect of nodal surface on the geometry of antiferromagnetic iron oxide

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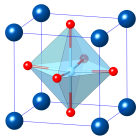
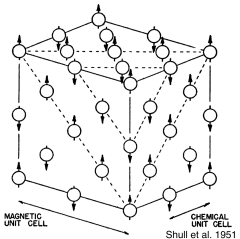
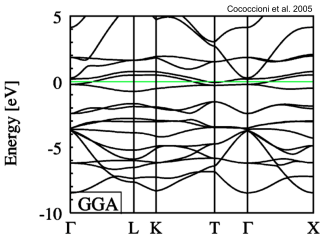
CPSFM | Center for Predictive Simulation of Functional Materials

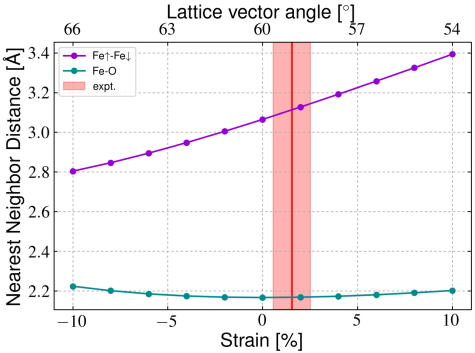


Thanks to the Center for Predictive Simulation of Functional Materials, DOE BES Computational Materials Sciences program. http://cpsfm.ornl.gov/

FeO is an interesting system for theory

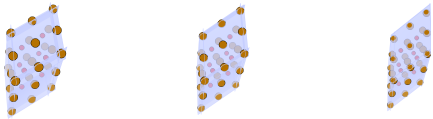






Groundstate properties:

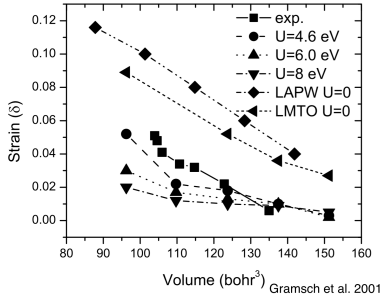
- Antiferromagnetic structure
- Rhombohedral lattice distortion
- Prototypical Mott insulator



Previous work on the ground state

Focus on rhombohedral lattice distortion:

- DFT/LAPW highlights that care is required[1]



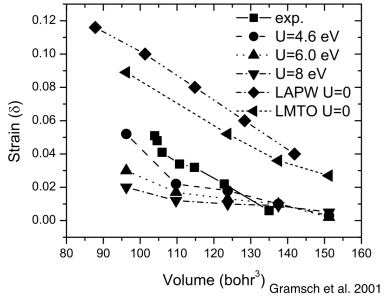
Gramsch et al. 2001

[1] Gramsch et al. (2003) 10.2138/am-2003-2-301

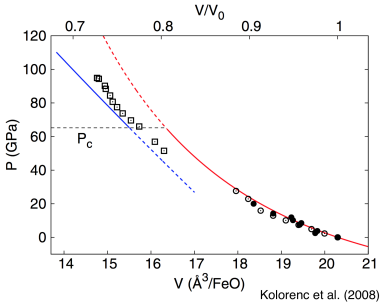
Previous work on the ground state

Focus on rhombohedral lattice distortion:

- DFT/LAPW highlights that care is required[1]
- QMC relatively unexplored, limited to EOS[2]



Gramsch et al. 2001



Kolorenc et al. (2008)

[1] Gramsch et al. (2003) 10.2138/am-2003-2-301

[2] Kolorenc et al. (2008) 10.1103/PhysRevLett.101.185502

How to do better than DFT

From the exact Born-Oppenheimer electronic hamiltonian:

$$\hat{H} = -\frac{1}{2}\sum_i \nabla_i^2 - \sum_{i,I} \frac{Z_I}{|\mathbf{r}_i - \mathbf{R}_I|} + \sum_{i<j} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$

DFT

$$\hat{H} = \sum_i h_i + V_{xc}$$

- Mapping onto $\hat{H}_{eff}$
- Non-interacting particles
- Parameterized V_{xc}
- Can solve exactly

QMC

- Stochastically sample $\hat{H}$
- Statistical (variational) estimate of E_0
- Input Ψ^T from, e.g. DFT
- Variational theorem holds
- Suffers from notorious "sign problem"

Can we reproduce rhombohedral lattice distortion in AFM B1 with QMC?

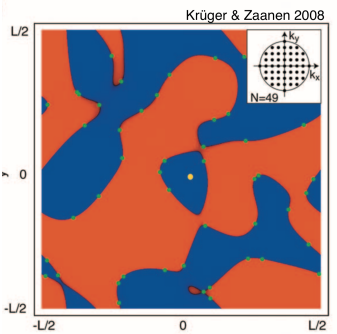
$$\Psi^{QMC}(\mathbf{r}) = D^{\uparrow}(\mathbf{r})D^{\downarrow}(\mathbf{r})e^{J(\mathbf{r})}$$

with

$$D \equiv \begin{vmatrix} \phi_1(\mathbf{r}_1) & \phi_1(\mathbf{r}_2) & \dots & \phi_1(\mathbf{r}_N) \\ \phi_2(\mathbf{r}_1) & \phi_2(\mathbf{r}_2) & \dots & \phi_2(\mathbf{r}_N) \\ \vdots & \vdots & \ddots & \vdots \\ \phi_N(\mathbf{r}_1) & \phi_N(\mathbf{r}_2) & \dots & \phi_N(\mathbf{r}_N) \end{vmatrix}$$

where

$\{\phi_i\}$ are generated from various flavors of DFT



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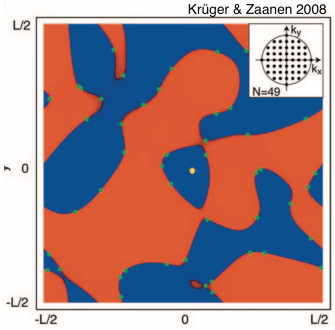
where

$\{\phi_i\}$ are generated from various flavors of DFT ← This is the hard part



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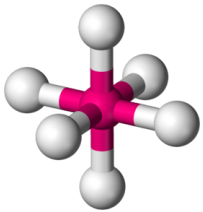
QMCPACK

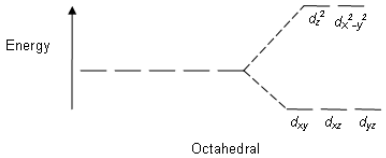
Forthcoming paper on arXiv: `https://arxiv.org/abs/1802.06922`

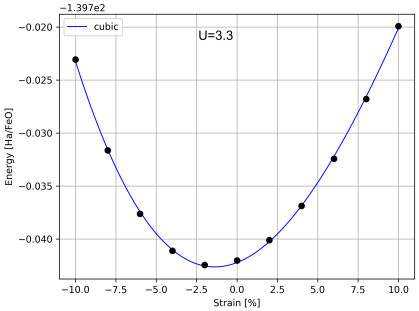
Wave function generation example: DFT+U

Summary of $+U$ calculations:

- d-matrix predicted equilibrium strain
- Not obvious (to me) which is best
- Metal/insulator both possible







spin config:

$$\begin{array}{ll} \uparrow & 1\,1\,1\,1\,1 \\ \downarrow & 0\,0\,0\,0\,1 \end{array}$$

Wave function generation example: DFT+U

Summary of $+U$ calculations:

- d-matrix predicted equilibrium strain
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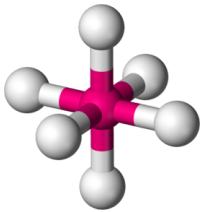
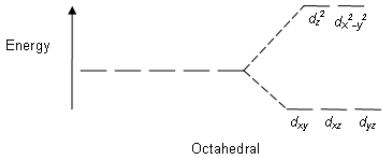
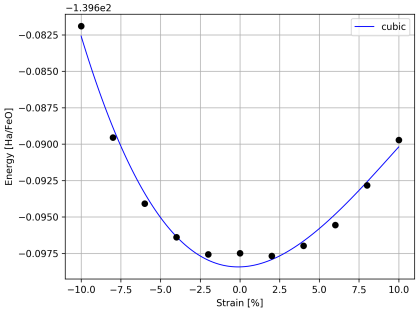






spin config:

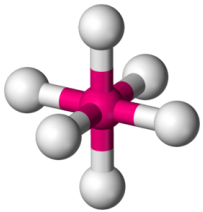
$$\uparrow \quad 1\,1\,1\,1\,1$$

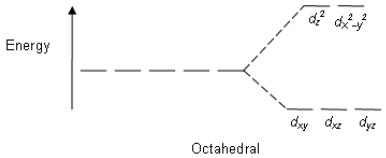
$$\downarrow \quad 0\,0\,0\,\frac{1}{2}\,\frac{1}{2}$$

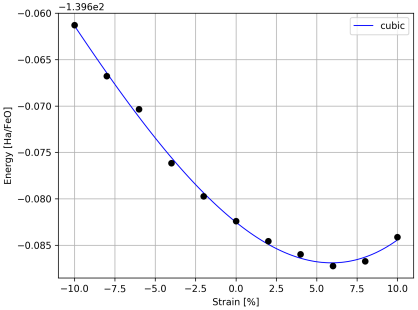
Wave function generation example: DFT+U

Summary of $+U$ calculations:

- d-matrix predicted equilibrium strain
- Not obvious (to me) which is best
- Metal/insulator both possible







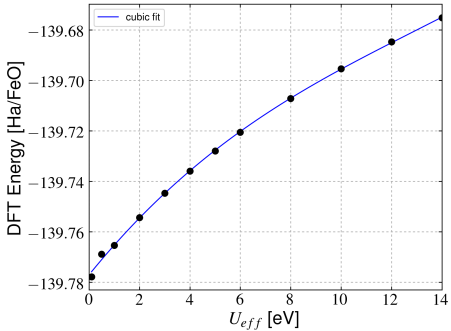
spin config:

$$\uparrow \quad 1\,1\,1\,1\,1$$

$$\downarrow \quad \frac{1}{2}\,0\,0\,0\,\frac{1}{2}$$

Effect of U_{eff} on trial wave function

DFT



As expected, DFT energy monotonically increases with U_{eff}

Effect of U_{eff} on trial wave function

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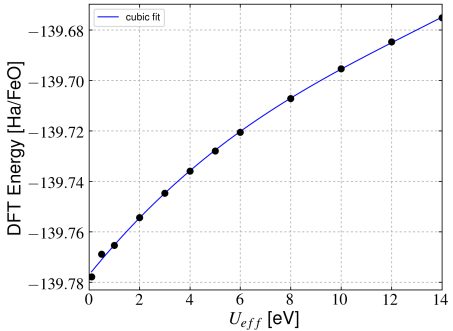
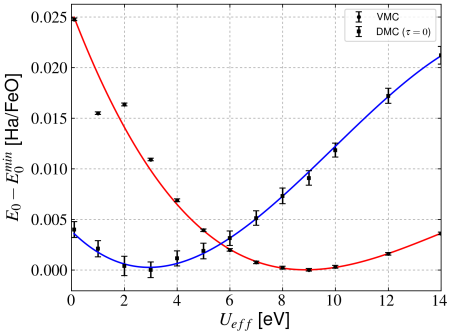
DFT



QMC - single twist



VMC and DMC predict optimal U_{eff} at a single twist

Effect of U_{eff} on trial wave function

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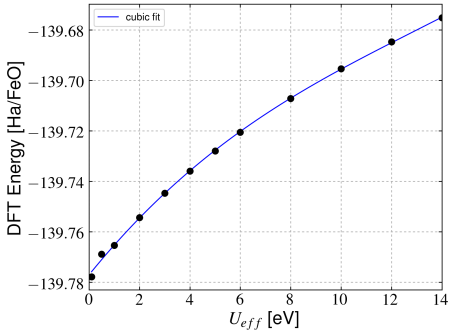
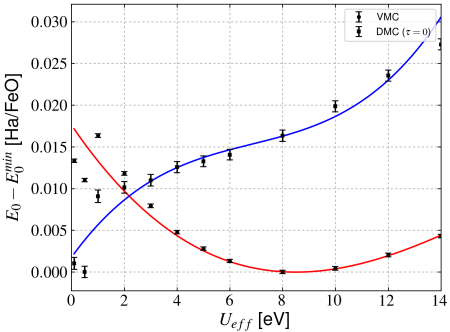
DFT

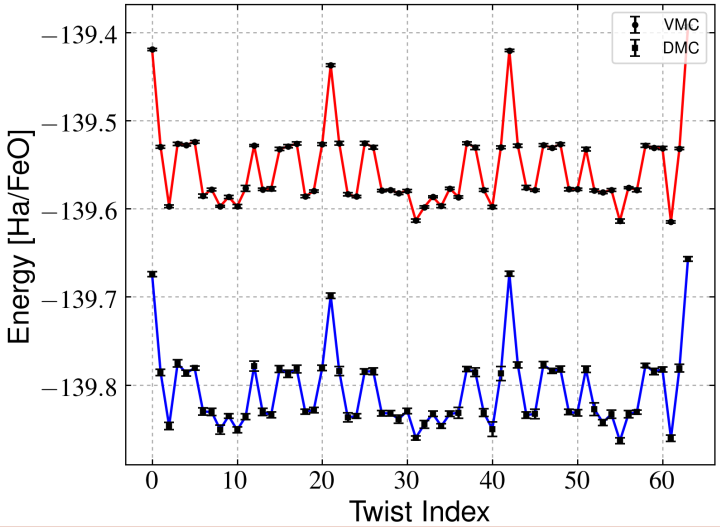


QMC - twist averaged

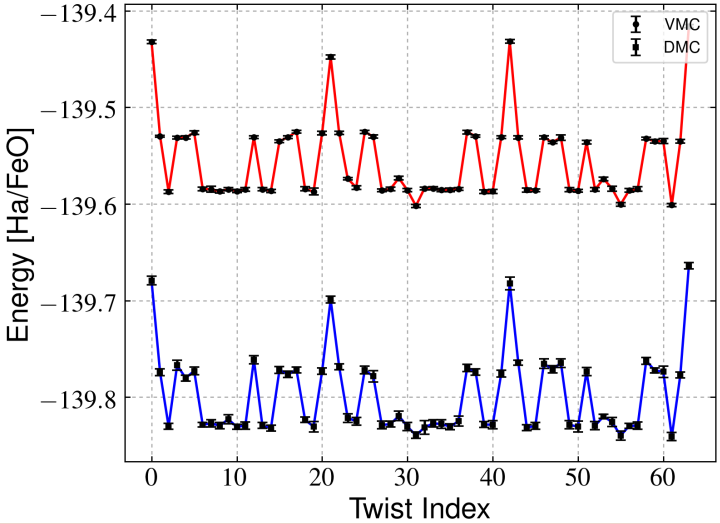


DMC energy also monotonically increases with U_{eff} after twist-averaging

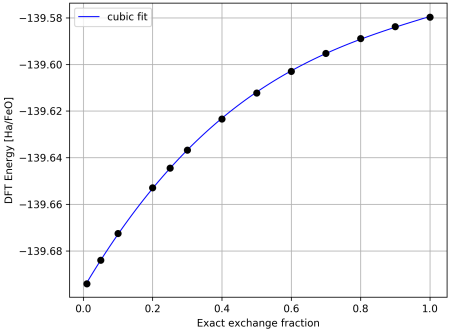
why is $+U$ higher?

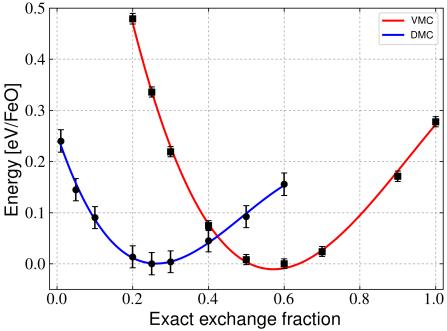


why is $+U$ higher?



Effect of hybrid exchange on trial wave function

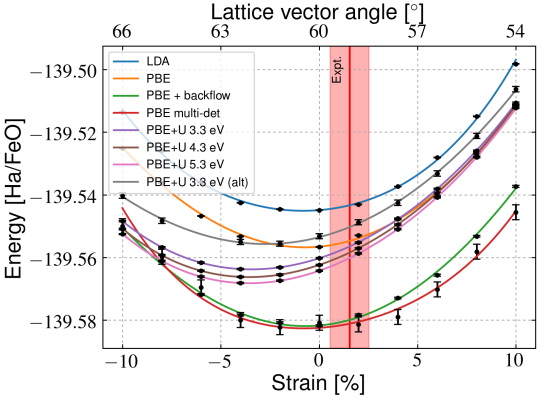




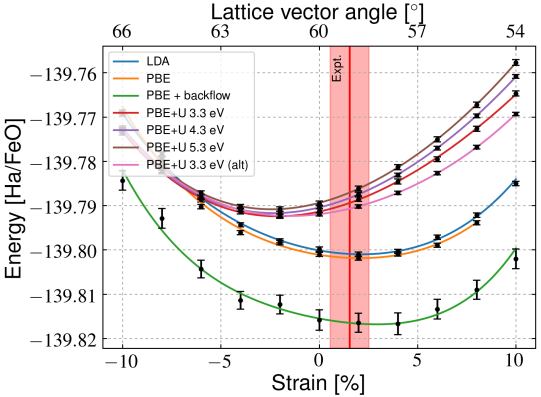
Results in progress:

DMC suggests exx approx. 4 mHa/FeO lower energy than $+U$

VMC results


Lattice vector angle [°]
66
63
60
57
54
LDA
PBE
PBE + backflow
PBE multi-det
PBE+U 3.3 eV
PBE+U 4.3 eV
PBE+U 5.3 eV
PBE+U 3.3 eV (alt)
Expt.
−139.50
−139.52
−139.54
−139.56
−139.58
Energy [Ha/FeO]
−10
−5
0
5
10
Strain [%]

DMC results


Lattice vector angle [°]
66
63
60
57
54
LDA
PBE
PBE + backflow
PBE+U 3.3 eV
PBE+U 4.3 eV
PBE+U 5.3 eV
PBE+U 3.3 eV (alt)
Expt.
Energy [Ha/FeO]
−139.76
−139.77
−139.78
−139.79
−139.80
−139.81
−139.82
−10
−5
0
5
10
Strain [%]

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Thank you!