Properties of minerals and other things from fundamental physics: From DFT to DMFT and QMC

R.E. COHEN

Geophysical Laboratory, Carnegie Institution of Washington, Washington, D.C. 20015

We now have a wide range of methods and community codes to study minerals, molecules, and fluids computationally, and to increase our understanding of Earth's contituents throughout the pressure, temperature, and composition. Challenges remain as many problems of interest are barely computationally tractable, and fundamental issues remain for iron- and other transitional metal bearing systems. For systems without transition metal ions, and for metallic transition metals like iron and its alloys, we can accurate compute most thermodynamic and transport properties of interest to geochemistry with sufficient accuracy for application. A range of techniques are available, varying in speed and accuracy. Kinetics and rheology remain challenges. For transition ion bearing systems are among the forefront problems in solid state physics.

I will talk about applications of density functional theory (DFT), dynamical mean field theory (DMFT) and quantum Monte Carlo (QMC), and will review examples from our work ranging from MgO, $MgSiO_3$, and Fe where density functional computations allow accurate predictions of thermal equations of state, elasticity, and thermal and chemical diffusivity, to transition metal oxides such as FeO which require many-body techniques suchas DMFT and QMC to obtain accurate results. Problems also sometimes arise in unexpected places. The local density approximation (LDA) predicts good properties for SiO_2 quartz and stishovite, but incorrectly gives stishovite as the ground state. The generalized gradient approximation (GGA) fixes this problem, but is less accurate for elastic properties. We have been applying quantum Monte Carlo (QMC) combined with phonons computed using density functional perturbation theory (DFPT) to silica, and obtain experimental accuracy in the thermal equation of state and phase transtions. For Feo we are using correlated methods based on LDA, such as LDA+U and DMFT, as well as QMC. DMFT has the advantage of giving the spectral density)electrnoic structure) as well as the total energy, but still contains approximations. QMC is an exact computation under the constraint of the many-body nodes which comes from a trial wave function.

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Iron in minerals under pressure

R.E. COHEN[1], L. SHULENBERGER[1], P. GANESH[1], J. OJWANG[1] AND S. SAVRASOV[2]

[1]Geophysical Laboratory, Carnegie Institution of Washington, Washington, D.C. 20015 USA

[2]University of California-Davis, Davis, CA, USA

We are using a variety of first-principles methods ranging from density functional theory [1, 2], LDA+U [3], dynamical mean field theory, and quantum Monte Carlo to understand the behavior of iron-bearing minerals under pressure. There are a number of possible phase transitions in iron-bearing minerals under pressure, including structural phase transitions, high-spin low-spin transitions [1, 2], magnetic ordering transitions (e.g. antiferromagnetic to ferromagnetic or paramagnetic) [4], and insulator to metal transitions. Some of these may occur together, such as a structural transition from an insulating to a metallic phase. We have been studying the behavior of pure FeO, iron-bearing perovskite, and iron-bearing post-perovskite, as well as other transition metal oxides in order to better understand the behavior of iron.

LDA+U predicts a low-symmetry, monoclinic, ground state for pure FeO, with a very high-pressure high-spin to low-spin transition. It predicts that FeO will remain high-spin through mantle pressures. Dynamical mean field theory (DMFT) within the simplest Hubbard I approximation predicts FeO to undergo a metallization transition under pressure, where essentially Fe^{2+} loses an electron, which becomes itinerant, similar to the behavior of EuO. We will use more advanced DMFT methods to see if this predicted behavior is robust.

LDA computations predict an antiferromagnetic to ferromagnetic transition in FeO under pressure [4]. Preliminary QMC computations are consistent with this prediction. The spin state of iron in perovskite and post-perovskite will also be discussed.

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