Poster Session A

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A1

Special Quasirandom Structure Approach to Studying Perovskite Solid Solutions

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Perovskite compounds, ABO_3, that have novel, multiferroic properties have increasingly complex compositions, with elemental mixing on both the A and B cation sites. Although it is desirable to use theoretical methods to predict the structures and properties, these complex compounds pose a unique challenge to computational studies. Here we demonstrate using special quasirandom structures (SQS) to predict the propertied of perovskite solid-solutions.

Using a 40-atom SQS the local structure and bonding within $K_{0.5}Na_{0.5}NbO_3$ is studied. To validate the method, a $K_{0.5}Na_{0.5}NbO_3$ specimen is synthesized and its pair distribution function (PDF) is determined by neutron diffraction. A simulated neutron diffraction PDF is determined and compared to the experimental results. Also a 40-atom rock-salt supercell (RSS), which is commonly used for theoretical calculations, is examined. The SQS predicted and experimentally determined PDFs agree out to 12 Å. The RSS, which is commonly found in the literature, deviates from the experimental results at distances as small as 4 Å. This demonstrates that the SQS is significantly more accurate than the existing calculations based on RSS.

The SQS approach is further validated by comparing SQS computed results to those computed using large (8×8×8) supercells to approximate a true random system. Well-characterized core shell atomic potentials are used to represent $Ba_xSr_{1-x}TiO_3$. To demonstrate the impact of system selection on the dynamical properties of perovskite oxides the phonon density of states is predicted for SQSs of varying quality, short-ranged ordered systems, e.g., the RSS, and the true random system. Three compositions are selected for testing.

A2

Abstract Submitted
for the ES2015 Workshop

Sorting Category: (Theoretical)

Detection of trap states using pump-probe measurements and TD-DFT GRAHAM CLENDENNING, University of Ontario Institute of Technology, STEPHANIE CHOING, University of California, Berkeley, AARON FRANCIS, North Carolina State University, MICHAEL SCHUURMAN, National Research Council, Ottawa, Canada, ROGER SOMMER, North Carolina State University, WALTER WEARE, North Carolina State University, TANJA CUK, University of California, Berkeley, ISAAC TAMBLYN, University of Ontario Institute of Technology — In artificial photosynthetic devices, light harvesting molecules drive reactions which create fuel (e.g by splitting water). For optimal fuel generation efficiency a long charge transfer excited state lifetime is necessary. In this study, we considered the experimentally synthesized vanadium(V) oxo compound VOLF which has been observed to remain in the optically excited state for 438 ps. We used linear response time dependent density functional theory (TD-DFT) to compute and interpret static UV-Vis spectra and ultrafast transient absorption. To assess effects due to finite temperature, we used first principles molecular dynamics to generate multiple starting configurations in different solvent environments. Comparing with experiment, we find good agreement with the observed spectra. Furthermore, we find that the bright state is associated with charge transfer to the vanadium from surrounding ligands. Our results suggest transitions between low lying d-states cause relaxation to the ground state to be both dipole and vibronically forbidden. This phenomenon gives rise to extended excited state lifetimes.

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☐ Prefer Oral Session
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A3

The Role of Many-Body Dispersion Interactions in the Racemic and Enantiomeric forms of Crystal Alanine. FARREN CURTIS, Physics and Engineering Physics, Tulane University, New Orleans, LA, ALEXANDRE TKATCHENKO, Fritz Haber Institute of the Max Planck Society, Berlin, Germany, NOA MAROM, Physics and Engineering Physics, Tulane University, New Orleans, LA, Polymorphism in molecular crystals is a phenomenon where crystals of the same molecular composition can crystallize in more than one structure. Different polymorphs of the same crystal may be extremely close in energy but may possess vastly different physical properties. It is well-known that hydrogen bonding and van der Waals (vdW) interactions play a large role in the formation and stability of polymorphic molecular crystals but these interactions are absent from standard semi-local Density Functional Theory (DFT) functionals in first-principles calculations. Recently, there has been much progress on dispersion-corrected DFT methods that use various vdW and dispersion corrections on top of a standard DFT calculation. These methods have demonstrated unprecedented accuracy in calculating the the structures, relative stabilities, and even anisotropic elastic properties of molecular crystals [Angew. Chem. Int. Ed. 52, 101002 (2013), PRL 113, 055701 (2014)]. Pairwise dispersion methods, such as the Tkatchenko-Sheffler (TS) [PRL 102, 073005 (2009)] method, apply a pairwise correction to approximate the leading term in the vdW energy. Many-body methods such as MBD [Phys. Rev. Lett. 108, 236402 (2012)] or MBD@rsSCS [J. Chem. Phys. 140, 18A508 (2014)], although computationally expensive, provide even higher order corrections to the vdW energy. In this work we look at two polymorphs of the amino acid Alanine which forms either enantiomeric (L-alanine) or racemic (DL-alanine) crystals. These two polymorphs are a challenging case for dispersion methods because one needs a highly accurate description of the energetics in order to calculate their relative stability. We compare the results of PBE, PBE+TS, and PBE+MBD@rsSCSC for the calculation of the structure, relative stability, and certain elastic properties of these crystals.

A4

Abstract Submitted for the
ES2015 Workshop
Developments in electronic structure theory
and excited states beyond ground DFT

Density functional theory study of perovskite oxide interfaces: Effects of exchange-correlation functional and thin film thickness on geometry, interface electronic structure, and magnetic order KEVIN J. MAY, ALEXIE M. KOLPAK, Massachustts Institute of Technology —Perovskite oxides have attracted significant attention in many diverse research fields such as electrocatalysis, oxide electronics and physics, owing to the variety of electronic and magnetic properties attainable in this family of materials. Thin films are of interest since their properties can differ drastically from those of the bulk[1]. Density functional theory drastically underestimates the band gap of many materials, and often inadequately describes the electron-electron interactions in strongly correlated materials. Previous work has shown that the choice of exchange-correlation (E_{xc}) functional can change the calculated magnetic ground state and structural properties of the bulk oxide. Moreover, the optimal E_{xc} can differ among different compounds[2,3]. This work aims to describe the structural, electronic and magnetic properties of $LaMO_3$ (M = V, Cr, Mn, Fe, Co, Ni) thin films on a $SrTiO_3$ substrate, as a function of the film thickness (1-4 unit cells) at a preliminary GGA level. Changes in the predicted magnetic ground state, octahedral rotation, and electronic reconstruction (such as two-dimensional electron gases) at the interface will be compared among different film thicknesses and reference bulk calculations.

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2. He, J. & Franchini, C. *Phys. Rev. B* **86,** 235117 (2012).
3. Yang, Z., Huang, Z., Ye, L. & Xie, X. *Phys. Rev. B* **60,** 15674–15682 (1999).

A first-principles-based study of thermal conductivity in $PbTiO_3$

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$PbTiO_3$ is a well studied ferroelectric perovskite. This work is about its lattice thermal conductivity, κ_L. We can expect κ_L to be low because of the presence of Pb, a heavy element. The structural instability associated with the transition from the paraelectric (PE) to the ferroelectric (FE) phase of $PbTiO_3$ may additionally contribute to lowering κ_L. In this work I use a Boltzmann transport equation solver for phonons, ShengBTE[1], to estimate κ_L of lead titanate. A key ingredient to this program is the force constant matrix of the material under study, obtained using harmonic approximation of the lattice vibrations. Density functional theory-based methods (at zero temperature) is used to calculate the above force constant. However, the cubic phase of $PbTiO_3$ shows a number of soft modes indicating its structural instability associated with the PE/FE phase transition. The presence of soft modes renders the above machinery unusable for the cubic phase. While we cannot calculate κ_L for the cubic phase, we can study the tetragonal phase. All the soft modes harden in the tetragonal phase, and harmonic approximation is generally valid again. More importantly, we can controllably soften the phonon branches in this structure by applying strain along the z direction. This exercise, albeit artificial, can tell us how the interaction among the acoustic and optic branches of phonons influences κ_L of $PbTiO_3$. Encouraged by the preliminary results indicating a low κ_L, I also look at the thermoelectric behavior of $PbTiO_3$. If it is possible to enhance the electrical conductivity of $PbTiO_3$ through doping, we could expect very good thermoelectric properties.

[1] Li W., Carrete J., Katcho N. A., Mingo N., Comp. Phys. Comm. **185**, 1747-1758 (2014).

Stability, Energetics, and Magnetic States of Cobalt Adatoms on Graphene†

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Research in the adsorption of transition metal adatoms on graphene has grown rapidly because of their possible use in spintronic applications. We investigate the stability and electronic properties of a single Co atom on graphene [1] with near-exact auxiliary-field quantum Monte Carlo (AFQMC) calculations [2]. We performed exact calculations on several model systems to benchmark the accuracy of various density functional theory methods. A frozen-orbital embedding scheme was combined with AFQMC to increase the reach in system sizes. Several energy minima are found as a function of the distance h between Co and graphene. Energetics only permit the Co atom to occupy the top site at $h = 2.2$ Å in a high-spin $3d^8 4s^1$ state, and the van der Waals region at $h = 3.3$ Å in a high-spin $3d^7 4s^2$ state. The findings provide an explanation for recent experimental observations with Co on free-standing graphene [3].

† This research work is supported by DOE, NSF, and ONR. Computing is provided by DOE (INCITE), NCSA Blue Waters, and W&M HPC facility.

[1] Virgus, Purwanto, Krakauer, and Zhang, Phys. Rev. Lett. **113**, 175502 (2014)

[2] Zhang and Krakauer, Phys. Rev. Lett. **90**, 136401 (2003)

[3] Eelbo, et al., Phys. Rev. B **87**, 205443 (2013)

Dirac line nodes in centrosymmetric crystals. YOUNGKUK KIM, The Makineni Theoretical Laboratories, Department of Chemistry, University of Pennsylvania, BENJAMIN J. WIEDER, CHARLES KANE, Department of Physics and Astronomy, University of Pennsylvania, ANDREW RAPPE, The Makineni Theoretical Laboratories, Department of Chemistry, University of Pennsylvania — Dirac line nodes (DLNs) are one–dimensional nodal lines of electronic energy bands with linear dispersion away from the line nodes. We propose new Z_2 topological characterization of semimetals hosting bulk DLNs, protected by inversion and time–reversal symmetries with vanishing spin–orbit interaction. aZ_2 topological invariants are introduced based on parity eigenvalues at the parity–invariant points in reciprocal space, dictating the presence of bulk DLNs and two-dimensional (2D) nearly-flat surface states. Using first-principles calculations, we predict that DLNs occur in Cu_3N near the Fermi energy when doped by transition metal atoms, such as Zn and Pd. The 2D surface states are demonstrated to emerge in the projected interior of the DLNs, and the effects of spin–orbit interactions and symmetry-breaking are briefly discussed.

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Ab initio study of a monolayer ZrO_2 epitaxial on Si

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Thin films of crystalline metal oxides grown on semiconductors have been of great scientific and technological interest because of their possible applications in electronic devices. One significant research aim is to achieve ferroelectricity in a crystalline and thin oxide film grown epitaxially on a semiconductor. This would be the key to realize non-volatile field-effect transistors in which the state of the system is retained by the oxide polarization. In this work, we study oxides that are not ferroelectric in the bulk but become ferroelectric as an ultrathin film on a semiconductor such as silicon. Thanks to the recent progress in epitaxial growth methods fabrication of such systems are also feasible.

In our study, we use density functional theory to first examine the interface of a single monolayer of ZrO_2 and Si. We show that a set of structures with a variety of positive and negative out-of-plane ferroelectric polarizations are stabilized. We present geometrical and electronic analyses of these structures. We also propose that a monolayer of ZrO_2 can be used as a buffer layer to induce ferroelectricity in thin perovskite oxides such as $SrTiO_3$ on Si which can couple the polarization of the oxide to the silicon carrier density. We analyze the layer-by-layer polarization profile of the Si-1ML ZrO_2-$SrTiO_3$ system and demonstrate that our proposal is viable. Finally we explore some aspects of switching between states of different polarization. We examine the transition barriers between metastable states using the nudged elastic bands method. We also analyze formation of differently polarized domains by mapping the system to an anisotropic Ising model with parameters determined by our *ab initio* theory.

Strain-dependent electronic and optical properties of metals for photocatalytic applications

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Photocatalytic production of hydrogen fuel from untreated sources of water is one of the highly sought methods for solar-to-fuel conversion, mainly due to its higher power density, ease of storage and transportation, and the possibility to ameliorate the intermittent nature of solar energy. However, photocatalytic systems performance ($< 10\%$) has been hindered due to the poor absorption of visible light on wide band gap oxides.

Recently, it has been shown that photo-excited hot carriers produced by surface plasmons can be used to carry out highly energetic chemical reactions, or used in photo -voltaic and -electrochemical devices through their transfer into a semiconductor. Initial charge-carriers distributions generated by plasmon decay are heavily dependent on the electronic structure of the metals, and particularly through interband transitions (affecting those involving d-bands) that dominate at higher plasmon energies.
Here, we use a band-structure engineering approach on a series of prototypical noble metals and other metal alloys under various values of strain. Electronic structure and optical properties are analysed to determine changing features and their origin, and their possible impact on the initial charge-carrier densities at different electron excitation energies —including those related to decay of surface plasmons. We will discuss our results in terms of carriers distributions for different materials and strain conditions, and will hint design principles for enhanced hot-carriers generation.

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A10

Scalable GW-BSE code development

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Electronic excitations can play a significant role in designing new functional materials. Although density functional theory has been widely applied to study ground state properties of materials, it has showed limitations in describing excited states of electrons, such as band gaps and optical spectra. The GW-Bethe Salpeter Equation (GW-BSE) method is the fully *ab initio* method of choice for general materials problems involving one- and two-electron excitations. For large unit cells and/or nanoscale systems, the GW calculation is typically *extremely* expensive computationally and is the "show stopper". This is in part due to the computational load typically requiring enormous numbers of FFTs for plane wave bases; separately, the memory requirements for most GW algorithms can be daunting.

We describe our efforts to develop new algorithms that permit GW calculations to be performed on large-scale parallel computers efficiently. We will focus on the computation of the dielectric screening matrix which is one of the most expensive parts of a GW calculation. Our approach uses a real-space representation of the polarizability and avoids extensive use of FFTs, and we compare its behavior to the more conventional G-space GW approaches. Our GW software is in the process of being interfaced with the highly scalable Car-Parinello *ab initio* molecular dynamics simulation package "OpenAtom" [1]. We briefly describe how OpenAtom leverages the charm++ parallel libraries [2, 3] to achieve admirable parallel scaling on large problems as well as greatly reduce the complexity of the parallel code.

[1] `http://charm.cs.uiuc.edu/OpenAtom`

[2] E. Bohm, A. Bhatele, L. V. Kale, M. E. Tuckerman, S. Kumar, J. A. Gunnels and G. J. Martyna: "Fine grained parallelization of the Car-Parrinello ab initio MD method on Blue Gene/L" IBM J. RES. & DEV. VOL. 52 NO. 1/2, 2008.

[3] G. Martyna, E. Bohm, R. Venkataraman, L. Kale, and A. Bhatele: Chapter 5: OpenAtom: Ab-initio Molecular Dynamics for Petascale Platforms, BOOK: *Parallel Science and Engineering Applications: The Charm++ Approach*

Abstract Submitted
for the MAR13 Meeting of
The American Physical Society

On The Geometric Nature of "Singlet Fission" in Certain Crystalline Conjugated Polymers NOAH RAHMAN, Department of Chemistry, University of California - Santa Barbara — In recent years, the coherent fission of low-lying singlet electronic excitations in conjugated polymers has attracted interest as a possible way to exceed the Shockley-Queisser limit in organic photovoltaics. Femtosecond spectroscopic and fluorescence measurements of such singlets and the resulting triplets in crystalline anthracene, tetracene and naphthalene reveal curious phenomena associated with certain vibrational modes, such as ultrafast propagation on a timescale inconsistent with conventional intersystem crossing, long-lived electronic coherence, and triplet magnetic anisotropy whose structure is consistent across all three materials. This conflicts with NRG and quantum chemical simulations, which posit isotropic triplets. I explain this by a dynamical Rashba spin-orbit interaction that decays as R^{-6}. This arises from a geometric SU(2) gauge potential generated by a nuclear-motion-induced parametric near-degeneracy of the molecular electronic states. The anisotropy is shown to follow from the work of Affleck and Oshikawa on spin one-half Heisenberg chains. Possible directions for future work are discussed, especially with regard to adiabatic pumping and topological insulators.

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Date submitted: 21 Dec 2012 Electronic form version 1.4

Gauge-discontinuity contribution to Chern-Simons orbital magnetoelectric coupling

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A quantity that is closely related to the physics of topological insulators is an isotropic contribution to the orbital magnetoelectric coupling that is known as the "Chern-Simons" or "axion" or "θ coupling" term. This last name comes from the fact that this contribution can be written in the form $(e^2/h)(\theta/2\pi)$, where θ is a phase angle, i.e., is only gauge-invariant modulo 2π. We propose here a new method for calculating θ, which is defined as a 3D Brillouin-zone integral of the Chern-Simons 3-form defined in terms of occupied Bloch functions. However, a straightforward finite-difference evaluation of this formula is only practical if a smooth and periodic gauge has been chosen in the entire Brillouin zone. Moreover, previous calculations have shown that for interesting systems expected to exhibit a large θ, such as topological insulators and systems derived from them, it is very difficult to converge the results with respect to **k**-point sampling.

In order to solve this problem, we divide the Brillouin zone into subvolumes, and the gauge is chosen to be smooth within each subvolume. These subvolumes meet at 2D planes in **k**-space where there is gauge discontinuity. The total θ response is then divided into contributions of two kinds: 3D integrals of the Chern-Simons 3-form over the subvolumes, and 2D integrals of a planar contribution associated with the gauge discontinuities on the boundary planes. Furthermore, in some cases it is necessary to subdivide the boundary planes into subregions separated by "vortex loops," which make yet a third contribution in terms of Berry phases defined around the vortex loops. The total θ thus consists of three kinds of terms, expressed as integrals over 3D, 2D and 1D manifolds. Interestingly, in the presence of time-reversal (TR) symmetry, both the 3D and 2D integrals vanish (assuming the gauge has been chosen to respect TR symmetry), and the 1D vortex-loop integral is either 0 or π, corresponding to the $\mathbb{Z}_2$ classification of 3D time-reversal invariant insulators. We illustrate our method by applying it to the Fu-Kane-Mele model with applied staggered Zeeman field.

Quantum transport studies of CNT-Based DNA Polymerase Nano-Circuits

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DNA polymerases are important enzymes that replicate double-helix DNA from single stranded template with both very high replication rate, hundreds of bases per second, and very low error rate, one per ~10^5 bases. Recently, it was found that the replication process can be electrically monitored by attaching a "Klenow fragment" (KF) of *polymerase I* to the surface of a carbon nanotube (CNT) and monitoring the current through the nanotube during replication. In this presentation, we report results from computational studies of DNA polymerase nano-circuits. We have first performed classical molecular dynamics simulations to obtain snapshots of different enzymatic stages, particularly the *open* state (when the enzyme is not replicating) and the *closed* state (when the enzyme is synthesizing a new base pair). We then used density functional theory and Keldysh non-equilibrium Green's function formalism to calculate transmission coefficients and currents for different enzymatic states. Our calculations show that the transmission spectrum and the current change significantly when the enzyme moves from the open to the closed state. While the original CNT-KF setup did not find significant differences between dissimilar bases, our theoretical work shows that both nucleotide analogs and gate potential scanning help distinguish between different DNA bases. Current work in progress is investigating conditions in which different bases might show larger variations in currents, which would allow for electrical sequencing of DNA.

A14

Abstract Submitted
for the Meeting of
The American Physical Society

Sorting Category:

Raman spectra calculations for nanostructures using *ab initio* real-space methods[1] N. SCOTT BOBBITT, JAMES R. CHELIKOWSKY, The University of Texas at Austin — We use a real-space pseudopotential method within density functional theory to calculate Raman spectra for Si nanocrystals. We examine the effects of quantum confinement and the presence of impurities, including both substitutional and interstitial impurities, such as lithium. The ability to predict the effects of dopant location on a Raman spectrum from first principles suggests that this calculation technique could be coupled with spectroscopic experiments to identify the size and nature of doped nanocrystals.

[1]This work is supported by the DOE under grant number DE-FG02-06ER46286. Computations were performed on machines at TACC and NERSC.

☐ Prefer Oral Session
☒ Prefer Poster Session

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A15

TITLE: Elasto-optic effect in semiconductors: a first principle approach using Maximally Localized Wannier Functions

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ABSTRACT:

Strain-induced changes of optical properties are of use in the design and functioning of devices that couple photons and phonons. The elasto-optic (or photo-elastic) effect describes a general materials property where strain induces a change in the dielectric tensor. Despite a number of experimental and computational works on this area, it is fair to say that a basic physical understanding of the effect and its materials dependence is lacking: for example, we know of no materials design rules for enhancing (or suppressing) elasto-optic response. Here, we describe an electronic structure method that helps us isolate the physics that determines this property. By analyzing dielectric response to strain, we explain why Maximally Localized Wannier Functions (MLWFs) is very useful in understanding the elasto-optic effect.

A16

Effects of surface step on Cu_2O thin film growth and Cu_2O surface reactivity

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Fundamental understanding of metal oxidation has received extensive interest due to its significant importance in many fields including high temperature corrosion, catalytic reactions and thin film processing. However, many questions still remain unresolved concerning the early stages of oxidation, which is inaccessible by the traditional surface science and ‘‘bulk’’ materials science techniques. The nucleation and growth of surface oxide is often complicated by surface inhomogeneities caused by the presence of surface defects such as steps. In this work, through the use of in-situ transmission electron microscopy (TEM) we observe that the presence of surface steps leads to the decomposition of the Cu_2O overlayer at the growth front of the Cu substrate, thereby resulting in oscillatory Cu_2O film growth. Using density-functional theory (DFT) total energy calculations and *ab initio* molecular dynamics (AIMD) simulations, we show that oxygen adsorption on the lower terrace destabilizes the Cu_2O thin film formed on the upper terrace that leads to oxide decomposition. Our results reveal the unique role of surface defects in oxide film growth and may have broader implications for understanding the fundamental process governing gas-surface reaction kinetics as modulated by atomic defects on a solid surface [1]. We will also discuss the effects of step edge on the surface reactivity of Cu_2O, revealed via DFT calculations of gas species adsorption on the terrace and step edges of Cu_2O surfaces.

[1] L. Li, L. Luo, J. Ciston, W. A. Saidi, E. A. Stach, J. C. Yang, and G. Zhou, Phys. Rev. Lett. **113**, 136104 (2014).

Ferroelectric switching path of polar corundum derivatives

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Polar magnets form one of the most promising classes of materials for multiferroics and spintronics. While much attention has focused on ABO_3 perovskites, recent studies have demonstrated that ABO_3 corundum-derived materials also have remarkable polarization and magnetization properties. In 2008, Fennie studied corundum-structure $FeMO_3$ (M = Fe, Mn, Ni) in the $LiNbO_3$-type phase using symmetry analysis and first-principles calculations.[1] He found that these novel materials exhibit large spontaneous polarizations and weak ferromagnetism. Most interestingly, he found that the two order parameters are coupled so that the magnetization can be switched by 180° when the direction of polarization is reversed by an applied electric field. Motivated by this pioneering work, novel ABO_3 and $A_2BB'O_6$ corundum derivatives have been successfully synthesized in high-temperature and high-pressure conditions.[2–5] Even though these noncentrosymmetric structures exhibit a spontaneous electric polarization, it has remained unclear whether the polarization can be switched by an external electric field. In this work, we use first-principles density-functional methods to study the intrinsic switching path and energy barrier for some of the ABO_3 and $A_2BB'O_6$ corundum derivatives that have been synthesized to date. Generalizing from our results, we clarify the general conditions under which FE switching is possible in this class of materials.

1. C. J. Fennie, Phys. Rev. Lett. **100**, 167203 (2008).
2. T. Varga *et al*, Phys. Rev. Lett. **103**, 047601 (2009).
3. M. Li *et al*, Angew. Chem. Int. Ed. **52**, 8406 (2013).
4. M. Li *et al*, Angew. Chem. Int. Ed. **53**, 10774 (2014).
5. M. Li *et al*, J. Adv. Mater. **27**, 2177 (2015).

A18

Warm Dense Crystallography

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The femtosecond scale of intense x-ray pulses from x-ray free electron lasers (XFELs) is now well established as being able to create interesting states of matter characterized by long-lived non-equilibrium semicore or core electron occupancies or by the heating of dense phases via the relaxation cascade initiated by the photoelectric effect. We address here the latter case of so-called 'warm dense matter' (WDM) and seek to better understand how experiments can be best designed to investigate the consequences of electronic heating in tepid WDM states of crystalline phases where the lattice degrees of freedom have insufficient time to respond. We report temperature-dependent density functional theory calculations for the x-ray diffraction from perfectly crystalline LiF, graphite, diamond, and Be. We find strong signatures of condensed-phase effects that are testable by experiment and that emphasize the importance of wide-angle scattering, i.e., that requires the use of high energy x-rays to obtain the Bragg intensities for a wide range of momentum transfers.

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A19

A Local Representation of the Dielectric Response Function

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The screened dielectric response function (χ) is a fundamental physical quantity that captures the many-electron correlation effect, key to a range of excited state properties formulated in the correlated electron theories. Although χ is non-local by definition, a real space partition of χ onto local structural motifs can help us gain further physical insight into, e.g., effective local screening properties. Because the bare response function, χ_0, is normally expressed in electron - hole pairs, standard localization procedures for electron wave functions can not be directly applied. In this work, we propose a new method to decompose χ_0 and χ into contributions from local response of Wannier orbitals. We demonstrate that the localization properties of the local dielectric response can be exploited to build a local basis set for χ_0, which can be used to construct a tight-binding Hamiltonian to perform dielectric band structure interpolation.

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A Linear Response DFT + U(Fe) Study of the α-Fe_2O_3(0001) Surface

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Abstract

The surfaces of iron oxides are essential components to a wide range of environmental and technological processes such as contaminant adsorption and heterogeneous catalysis. However, even for the most stable and abundant iron oxides surface, hematite α-Fe_2O_3(0001), our understanding of the phase diagram is incomplete. DFT modeling using an $ab\ initio$ thermodynamics framework [1] has the potential to provide insight on the preferred surface structure as a function of T and p conditions, but these strongly correlated materials pose challenges to the methodology. Various predictions for the (0001) surface phase diagram have been summarized in the literature [2] and note that supposedly improved calculations using GGA + U actually lead to worsened stability predictions. Herein, we aim to identify methods for reliable theoretical predictions for the surface phase diagrams of strongly correlated materials by studying the α-Fe_2O_3(0001) surface. We focus on four terminations: -O3Fe, -O3Fe2, -Fe=O and -Fe2O3. Only the -O3Fe and -Fe2O3 terminations are reported experimentally, while GGA + U predicts the -Fe=O surface to be stable over a wide range of T and p_{O_2} conditions [3]. We use a linear response method to derive U values for chemically distinct Fe sites in each surface structure. We go on to show that this U(Fe) approach alone does not recover a realistic phase diagram. Further investigation shows that the hybridization between transition metal d- and oxygen p-orbital is strong enough to warrant an additional Coulomb correction, U^p, to balance their repulsion effects. Our results show that a U(Fe) + U^p approach does yield a reasonable α-Fe_2O_3(0001) phase diagram, as well as good predictions of the physical properties of hematite such as lattice constant, bulk modulus, and band gap. Finally, we demonstrate how the U(Fe) + U^p method impacts predictions for heterogeneous reactivity on the hematite surface.

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Automatically tuned PAW pseudopotentials for accuracy and efficiency

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Pseudopotentials for electronic structure calculations using density functional theory (DFT) can lower computational expense compared to all-electron DFT. However, pseudopotentials that are accurate, efficient, and transferable are difficult to generate because these objectives can compete with each other. We have used a multi-objective genetic algorithm to optimize PAW pseudopotentials based on accuracy and efficiency. The parameters varied for optimization were cutoff radii and projector energies. In all optimizations, the computational work requirements of pseudopotentials are based on an estimate of floating point operations required to perform electronic structure calculations using those pseudopotentials. Pseudopotential accuracy was determined by calculating lattice constant and bulk modulus and comparing to the lattice constant and bulk modulus given by all-electron DFT. To encourage transferability, pseudopotentials were simultaneously optimized for different crystal systems. Additional pseudopotentials were optimized using an alternative force-based method where pseudopotential accuracy is determined by the interatomic forces in a perturbed crystal system. Motivations for our force-based approach are applications where atoms are not in a perfect-crystal environment, such as defects in lattices, material interfaces, and molecular dynamics.