Project Title:

First-principles calculations of topological electronic states in Ir/Rh pyrochlore oxides

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1. Low-temperature properties of materials strongly depend on the electronic structure, which is strongly affected by possible nontrivial topologies as well as symmetries. For instance, electronic structures endowed with U(1) or Z_2 topology may host unconventional phenomena, such as quantum Hall or spin Hall effects. If it is combined with electron correlation effects, materials may show even more fascinating properties. Because of a moderately large spin-orbit coupling for Ir 5d electrons, pyrochlore iridate oxides $A_2Ir_2O_7$ with A being rare-earth elements and Y have recently been studied intensively as candidate materials to correlated topological materials, such as Weyl or Dirac semimetals and axion insulators. I have aimed at designing and predicting electronic structures of the related materials, including $A_2Ir_2O_7$ and $A_2Rh_2O_7$, under ambient and hydrostatic pressures. This project is continued from our Quick Use (Q13367) for test LDA calculations on $La_2Ir_2O_7$ at the end of the last fiscal year.
2. Electronic structures of pyrochlore iridates and rhodates with and without strong electron correlation effects have been investigated by performing fully relativistic first-principles local-density-approximation (LDA) and LDA+U calculations based on the OPENMX method for hypothetical $A_2M_2O_7$ (A=La, Y; M = Ir, Rh) with RICC supercomputers.

We have finished performing LDA calculations on both $La_2Ir_2O_7$ and $La_2Rh_2O_7$, which are summarized for $La_2Ir_2O_7$ in Fig.1. On the contrary to the previous calculations reported in X. Wang, A. Turner, A. Vishwanath, and S. Y. Savrasov, Phys. Rev. B **83**, 205101 (2011), the ground state of both $La_2Ir_2O_7$ and $La_2Rh_2O_7$, have been found to be paramagnetic without any onsite Coulomb repulsion. Depending on the lattice constant a and the oxygen position parameter x, we have obtained the following changes in the four Kramers pairs of electron band dispersions around the chemical potential. (See Fig.1.) For x being compared to that for $Pr_2Ir_2O_7$ ($x \sim 0.33$), the system is a metal (M) forming small electron and hole Fermi surfaces around the Γ point and near the L points. For $x \sim 0.31-0.32$, these Fermi surfaces disappear and the two electron band dispersions touch at the Γ point exactly at the chemical potential, yielding Fermi-point semimetal (FPSM). For slightly smaller x, the fourfold degenerate Γ_8 energy level crosses the Γ_6, leading to a Z2 topological insulator (TI). Similar tendency has been obtained for hypothetical $La_2Rh_2O_7$.

Effects of strong correlations have also been examined for $Pr_2Ir_2O_7$ by using the LDA+U method based on the same OPENMX package, to check if they induce magnetic orders or metal-insulator transitions. In sharp contrast to the previous results by Wang et al., the ground state of Pr2Ir2O7 is found to remain paramagnetic and metallic for small onsite Coulomb repulsion U. A moderately large value of U=1.3 eV is required for stabilizing an all-in, all-out antiferromagnetic metal, which is nearly degenerate with an all-in, all-out

antiferromagnetic insulator. This value of U is reasonable, since for $Y_2Ir_2O_7$, it yields an antiferromagnetic insulator with a small ordered magnetic moment of $0.51\mu_B$ / Ir in reasonable agreement with findings by neutron scattering experiments.

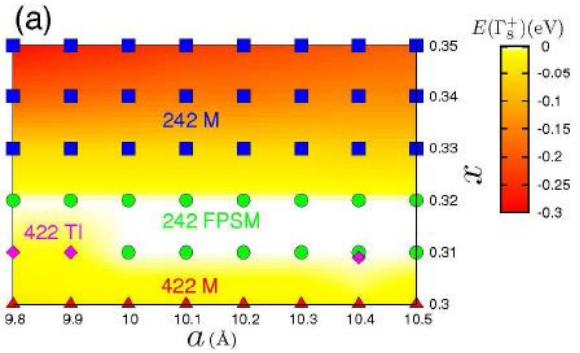

Fig.1. Phase diagram of $La_2Ir_2O_7$ as a function of the lattice constant a and the oxygen position parameter x.

3. We have performed intensive extensive first-principles LDA and LDA+U calculations on $A_2M_2O_7$ (A=La, Y; M = Ir, Rh) on a basis of the OPENMX package. We obtained a paramagnetic metal, Fermi-point semimetal, and Z_2 topological insulator phases, which are stable against at least weak Coulomb interaction. We have also obtained a reasonable value of the onsite Coulomb repulsion U, which reproduces an antiferromagnetic insulator for $Y_2Ir_2O_7$, which most likely shows the all-in, all-out magnetic structure and a paramagnetic metal for $Pr_2Ir_2O_7$.

4. We need to perform more LDA+U calculations to complete the phase diagram in the space of (x,U). This will reveal a more practically correct phase diagram which is direclyt relevance to pyrochlore iridates, and thereby uncover possible nontrivial phases, including Fermi-point semimetal, Z2-topological insulator, axion insulator, Weyl semimetal phase, all-in, all-out antiferromagnetic metal/insulator, ferromagnetic metal/insulator.

Fiscal Year 2014 List of Publications Resulting from the Use of RICC

[Others]

F. Ishii, Y. P. Mizuta, T. Kato, T. Ozaki, H. Weng, S. Onoda, “First-principles study on cubic pyrochlore iridates: Fermi-point semimetal and Z2 topological insulator”, submitted to Phys. Rev. Lett.

F. Ishii, Y. P. Mizuta, T. Kato, T. Ozaki, H. Weng, S. Onoda, “First-principles study on pyrochlore iridate and rhodate oxides: metal-insulator transition, magnetism, and topology” 2014 Japan Physical Society Fall Meeting (Chubu University, Kasugai, Sep. 7-10, 2014)." 2014 Japan 日本物理学会 2014 年秋季大会(中部大学, 春日井, 2014 年 9 月 7-10 日)

S. Onoda, “Topology, magnetism, and Mott insulator in pyrochlore iridates and rhodates”, to be presented at and invited to KITP workshop: “New Phases and Emergent Phenomena in Correlated Materials with Strong Spin-Orbit Coupling” (Kavli Institute of Theoretical Physics, University of California, Santa Barbara, 2015 July-August).