Usage Report for Fiscal Year 2016

Project Title: First-principles studies on electronic structures of magnetic pyrochlore oxides $A_2Ir_2O_7$ and Ir_2O_4

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1. This project was taken over from the previous one (Q15307), where strongly correlated iridium pyrochlore oxides $R_2Ir_2O_7$ were systematically studied using first-principles calculations. The previous calculations successfully explained experimental findings on the phase transition from a paramagnetic semimetal for R=Pr through a possible all-in, all-out antiferromagnetic Weyl semimetal to an all-in, all-out antiferromagnetic Mott insulator for R=Nd, Sm, Eu, and Y. This year, in addition to some more calculations to understand recent experimental reports by collaborators on carrier-doped Mott-insulating pyrochlore iridates $R_2Ir_2O_7$, we have performed extensive first-principles calculations on the other material, A-site-deintercalated spinel iridate Ir_2O_4 thin films. This material had been considered to be another candidate for a topological Weyl semimetal, since Ir ions form the same lattice structure as in the pyrochlore case.

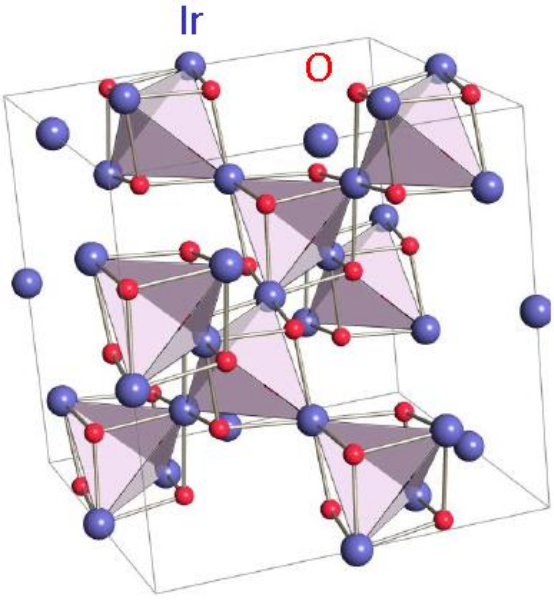

Fig.1: Crystal structure of the ideal cubic A-site-deintercalated spinel iridate Ir_2O_4.

2. We employ first-principles electronic structure calculations based on the fully relativistic local spin density functional with the onsite Coulomb interaction U, in particular, using the OPENMX package. We have used roughly 400k hours of CPU time on the MPC system and 60k hours on the ACSG system by Feb. 23, occupying 3 to 32 nodes for each computational job.

3. We have investigated the ideal cubic case of Ir_2O_4 as well as the tetragonal and rhombohedral cases that have already been successfully grown on the MgO and $LiNbO_3$ substrates, respectively. Using the stable crystal structures that were determined from molecular dynamics calculations implemented into the OpenMX package last year, we have successfully obtained a set of (meta)stable magnetically ordered states, which are categorized according to the magnetic space group. Figure 2 presents a comparison among several (meta)stable states labeled by the magnetic space group in the ideal cubic Ir_2O_4. In sharp contrast to pyrochlore iridates $R_2Ir_2O_7$, all-in, all-out antiferromagnetic state is unstable in any range of U. At the ground state, the system undergoes a couple of phase transitions from a metallic paramagnet ($Fd\bar{3}m1'$) through a metallic splayed ferromagnet ($I4_1/am'd'$) to another antiferromagnet ($I4_1'/amd'$), which is realized in the insulating magnetic rare-earth pyrochlore material $Er_2Ti_2O_7$. Furthermore, it is found that a large-U region can be modeled as nearest-neighbor quantum spin ice with a

large spin-ice-rule coupling of the order of 30 meV, and introducing a tetragonal distortion drives a phase transition from the insulating $I4_1'/amd'$ antiferromagnet to an insulating splayed ferromagnet $Fdd'd'$, which differs from the $I4_1/am'd'$ ferromagnet in that the ferromagnetic moment lies within the tetragonal plane. The possibility of having Weyl points near the Fermi level have also been critically examined and excluded for all the cases.

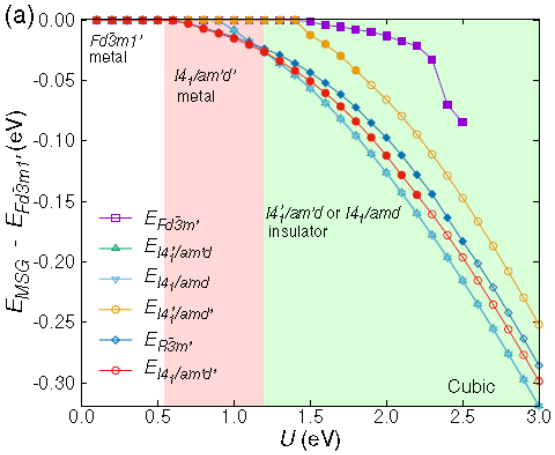

Fig.2: Energies of (meta)stable magnetically ordered states compared to the paramagnetic state as functions of U, in the ideal cubic case

4. Our first-principles calculations have revealed that A-site-deintercalated spinel iridates Ir_2O_4 are candidates for quantum spin ice with a spin-ice-rule coupling that is two orders of magnitude larger than in magnetic rare-earth pyrochlores, for instance, $Pr_2Zr_2O_7$, $Tb_2Ti_2O_7$, and $Yb_2Ti_2O_7$.

5. Calculations for understanding the latest experimental reports in $(R_{1-x}Ca_x)_2Ir_2O_7$ and the magnetic neutron-scattering diffraction pattern of $R_2Ir_2O_7$ are still on the way. We plan to continue these calculations in the next fiscal year.

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Fiscal Year 2016 List of Publications Resulting from the Use of the supercomputer

Shigeki Onoda, Fumiyuki Ishii, "First-principles design of the iridate spinel Ir_2O_4 for high-temperature quantum spin ice", arXiv:1612.00553.