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 started, aiming at extending the previous projects (Q13367 and G14013) of the electronic structure calculations on pyrochlore iridates $Pr_2Ir_2O_7$ and $Y_2Ir_2O_7$, which clarified a phase diagram, including a paramagnetic semimetal, "all-in, all-out" antiferromagnetic Weyl semimetal and insulator, and Z_2 topological insulator, of the materials with varying lattice constant, oxygen position parameter, and on-site Coulomb interaction. Recent developments of experiments on Mott-insulating pyrochlore iridate $R_2Ir_2O_7$ have shown a qualitative change in the conducting properties as the rare-earth element R is varied from Nd to Sm or late lanthanide series. It has also been found experimentally that doped holes yield a temperature-linear dc conductivity below the Neel temperature, as expected for Weyl semimetals. There also exists yet another class of epitaxially grown iridates Ir_2O_4 on certain substrates. It forms a spinel structure but the Ir network forms the pyrochlore lattice as in $R_2Ir_2O_7$, Hence it might be another candidate for hosting time-reversal broken Weyl semimetal. Motivated by these observations, we clarify electronic and magnetic structures of these materials.

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, the OPENMX package. We have launched the project in November and used roughly 200kh of CPU time on the MPC system by Feb. 23, using 3 to 4 nodes for each computational job.

3. We have succeeded in obtaining the energy gain of the all-in, all-out antiferromagnetic state over the paramagnetic and the ordered moment per Ir as a function of U for $A_2Ir_2O_7$ with A = Pr, Nd, Sm, Eu, and Y, as shown in Fig.1. In particular, all the cases host Weyl points slightly above the Fermi level when the ordered magnetic moment is up to about 0.2 μ_B per Ir, as shown in Fig.2.

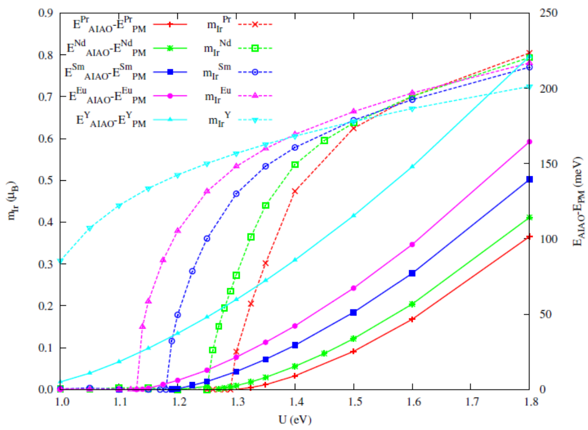

Fig.1: The energy gain and the ordered Ir moment of the all-in, all-out antiferromagnetically ordered state over the paramagnetic semimetal in $A_2Ir_2O_7$ for A=Pr, Nd, Sm, Eu, and Y.

Stable crystal structures of Ir_2O_4 have been identified in the hypothetical cubic case and in the cases grown on tetragonal MgO(001) and rhombohedral $LiNbO_3$(0001) substrates. We have found that the all-in, all-out magnetic structure is unstable and that this system involves two-in, two-out like ferromagnetic interactions, as in spin ice.

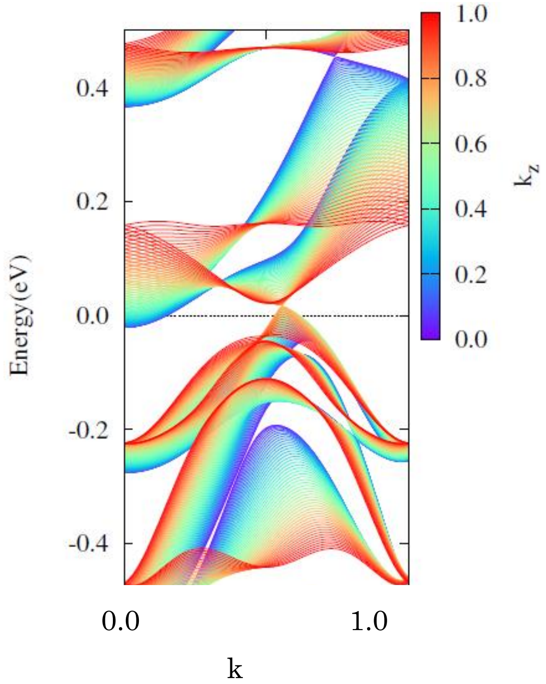

Fig.2: Electronic band structure of Nd2Ir2O7 with U=1.35 eV in the momentum plane of (k,k,kz), in which a Weyl point appears near the Fermi level set to zero energy.

4 . Our first-principles calculations with U~1.3 eV explain the metal-insulaor transition and the all-in, all-out magnetic order observed in $A_2Ir_2O_7$ except for A=Pr. Weyl semimetal phase should be present when the ordered moment is small, though it has not been detected experimentally. It has also been found that the spinel iridate Ir_2O_4 involves ferromagnetic interaction as in spin ice. The results on each material series are to be submitted as a separate paper.

5 . We plan to continue the study on $A_2Ir_2O_7$ in the doped case in the next fiscal year, and on $R_2Ir_2O_7$ to finish some calculations yet to be completed, in particular, calculations of exchange interactions and the optical conductivity.

Usage Report for Fiscal Year 2015

Fiscal Year 2015 List of Publications Resulting from the Use of the supercomputer

[Oral presentation at an international symposium]

Shigeki Onoda, "Quantum spin ice: current issues and beyond", 2016 Quantum Materials Symposium (Muui Island Homeplus Academy, Korea, Feb. 22-26, 2016).