Usage Report for Fiscal Year 2019

Project Title:

Numerical studies on superlattices and interfaces of quantum spin ice

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1. Background and purpose of the project, relationship of the project with other projects

Quantum spin liquids are novel states of magnetic materials where localized magnetic moments of electrons do not show any symmetry-breaking long-range magnetic order, but their excitations fractionalize into new quasiparticles, called spinons, coupled to deconfined gauge fields [1]. They are prototypical examples of correlated topological states and exhibit a nontrivial intrinsic topological order and the associated quantum entanglement [2]. Despite progress in basic theoretical understandings and in accumulating candidate materials, phenomena that provide compelling evidence of quantum spin liquids with a complete identification of their classes remain open. In this project, we focus on quantum spin ice, which describes low-energy magnetic properties of magnetic rare-earth pyrochlore oxides $A_2B_2O_7$ (A=Pr, Tb, or Yb, B=Ti, Zr, or Hf) [3,4,5] and A-site deintercalated spinel iridate Ir_2O_4 [6].

So far, using quantum Monte-Carlo simulations, we have numerically verified the stability of a U(1) quantum spin liquid phase and/or regime[7] in the phase diagram of quantum spin ice [8]. The U(1) quantum spin liquid phase of quantum spin ice can be viewed as a magnetic analogue of an electrical insulator in quantum electrodynamics. It shows gapped bosonic spinons carrying monopole charges of magnetization and the spinons are coupled to emergent U(1) gauge fields, i.e., fictitious electromagnetic fields. It also exhibits gapless photon modes as fluctuations of gauge fields, as in quantum electrodynamics.

Among the above candidate materials, Ir_2O_4 is advantageous for performing experiments as well as for potential applications, because of its much higher energy and thus temperature scales [6]. However, Ir_2O_4 needs to be grown as epitaxial thin films. Thus, the presence of the interface with and strain from the substrate may drastically change the electronic structure. Now, it is called for to demonstrate from first-principles electronic structure calculations the case where the low-energy effective spin model derived from first principles indeed gives the U(1) quantum spin liquid.

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[3] K. Kimura, S. Nakatsuji, J. Wen, C. Broholm, M. Stone, E. Nishibori, H. Sawa, Nature Comms. 4, 1934 (2013).
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Tsuei, Y. Yasui, K. Kakurai, M. R. Lees, Nature Comms. 3 (2012) 992.
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2. Specific usage status of the system and calculation method

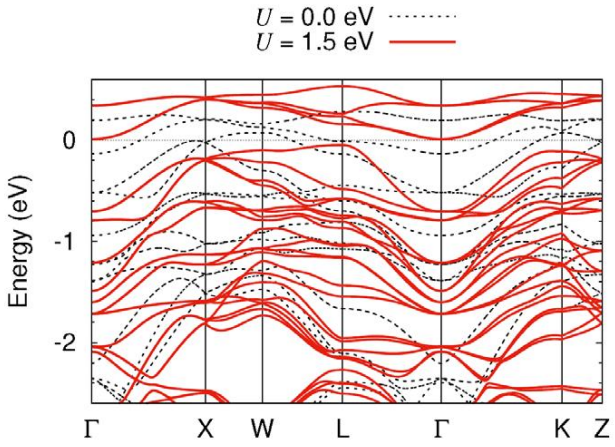
In the very end of the last fiscal year, we performed first-principles calculations on Mott-insulating Ir_2O_4 and band-insulating ZnIr2O4 in the project G18032, using the OPENMX package [9]. In the current project, G19026, we have tried to optimize the crystal structure of the $2Ir_2O_4$-$2ZnIr_2O_4$ superlattice stacked in the (001) direction on several potential substrates by the molecular dynamics simulation implemented in the OPENMX package, using the GWMPC system with a massive parallelization maximally using both the MPI and OpenMP. We also performed preliminary LDA+U calculations on the $2Ir_2O_4$-$2ZnIr_2O_4$ superlattices, using the GWMPC system

[9] T. Ozaki, H. Kino, J. Yu, M. J. Han, N. Kobayashi, M. Ohfuti, F. Ishii, T. Ohwaki, H.Weng, K. Terakura, Open source package for Material Explorer, http://www.openmxsquare.org/.

3. Result

Our first-principles calculations have shown that the spinel $ZnIr_2O_4$, which has been successfully synthesized only as thin films, possesses the lattice constant of 8.582 A, which is 2% smaller than that of Ir_2O_4 [6], and the oxygen position parameter x = 0.258301, which is also smaller than that (x=0.26911) of Ir2O4 [6]. This means that Ir_2O_4 should be compressed when it has an interface with ZnIr2O4. We then established the out-of-plane lattice constant c = 8.798 A of $ZnIr_2O_4$ on the MgO(100) substrate with the in-plane lattice constant of 8.422 A. In both cases, $ZnIr_2O_4$ serves as a band insulator with fully filled Ir 5d t2g electrons, in contrast to the Mott-insulating Ir_2O_4 with a single Ir 5d t2g hole per Ir. (See Fig.1 below. The tetragonal strain weakly split the degeneracy of the band dispersions.) Using these parameters as initial inputs, we obtained a preliminary solution for the paramagnetic state of the $2(Ir_2O_4)$-$2(Ir_2O_4)$ superlattice on the MgO(100) substrate. This solution has similar fully filled t2g electrons in the $2ZnIr_2O_4$ layer and a single t2g hole per Ir in the Ir_2O_4 layer, as we expected, though the convergence has yet to be established,

(a)



(b)

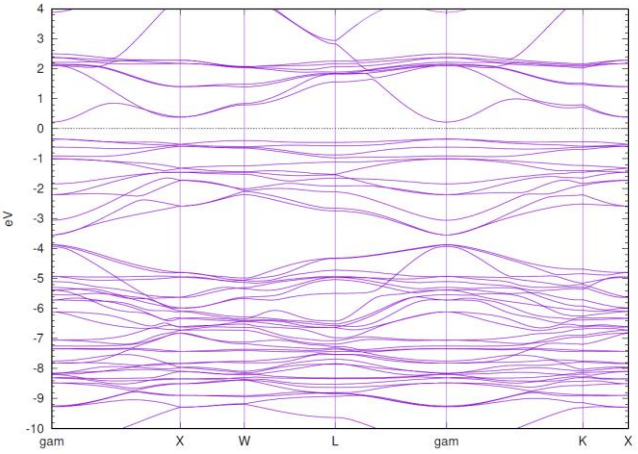


Fig.1: (a) Paramagnetic (U=0.0 eV) and Mott-insulating (U=1.5 eV) electronic band structures of the cubic bulk Ir_2O_4. (b) Band-insulating electronic band structure of Zn

Ir_2O_4 on the MgO(100) substrate.

4. Conclusion

We established the stable crystal structures of the spinel ZnIr2O4 both in the cubic bulk and on the MgO(100) substrate and investigated the superlattice system $2(Ir_2O_4)$-$2(ZnIr_2O_4)$ on the MgO(100) substrate. In the preliminary solution of the superlattice system, the paramagnetic electronic structure near the chemical potential is almost confined into each two-dimensional Ir_2O_4 layer, which accommodates an unpaired single hole per Ir. This suggests that these systems are promising candidates of hosting interfaces of Mott-insulating quantum spin ice with a band insulator. Also, our preliminary LDA+U calculations show that these unpaired holes can show both ferromagnetic and antiferromagnetic states, though the convergence issue and finding the true global energy minimum, as well as the application to much larger system sizes, has yet to be solved.

5. Schedule and prospect for the future

In this fiscal year, we had challenges for achieving a fast and reliable convergence of the crystal structures of both paramagnetic and magnetically ordered Ir_2O_4-$ZnIr_2O_4$ superlattices. This is because of the following fact. Preliminary LDA+U calculations we performed on these systems in this fiscal year show that the magnetic structures are significantly sensitive to and vary with small changes in the crystal structure. In other words, we have found unusually large correlations between the lattice and the magnetism. It forced us to perform molecular dynamics calculations by which both the lattice and the magnetism are simultaneously optimized. However, the molecular dynamics calculations on such large systems require a large computational cost, as we estimated when we applied for the CPU time. Magnetically ordered states require additional computational costs, typically, by 3-4 times. Therefore, we needed to improve the method for a fast convergence before embarking on extensive calculations, while we investigated the substrate dependence of the crystal structure of the paramagnetic solutions. Now that we could speed up the convergence of molecular dynamics calculations of magnetically ordered states by a factor of two, in the remaining two months and in the next fiscal year, we will investigate the seven different magnetically ordered solutions within the LDA+U calculations on the superlattice systems. Then, we will achieve our main of deriving effective spin model for the superlattice systems from the energy differences and ordered magnetic moments obtained from the LDA+U calculations, as we did in the previous projects Q17307 and G18032 [6].

Fiscal Year 2019 List of Publications Resulting from the Use of the supercomputer

[Oral presentation]

Shigeki Onoda, “Quantum spin ice under an electric field”, IoP Magnetism- TCM Group Symposium (Abingdon, UK, July 17th, 2019). Invited.