Usage Report for Fiscal Year 2018

**Project Title:**

# Numerical studies on topologically nontrivial frustrated magnets

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

Uncovering novel quantum phases with nontrivial topological structures and the associated phenomena has been a central issue of great interest in condensed matter physics. In particular, frustrated magnets offer a fertile laboratory for correlated topological materials, such as quantum spin liquids (QSLs). The QSLs are characterized by the absence of any symmetry-breaking long-range order but by a quasi-particle excitations fractionalized from spins, a long-range quantum entanglement, and the associated intrinsic nontrivial topology. In this project, we plan to search for these topologically nontrivial magnetic states in experimentally accessible frustrated magnets. Specifically, we planned to work on three subtopics in this project.

i) First-principles study on the Mott-insulating A-site-deintercalated spinel iridate. This topic has been taken over from Q17307, where we have shown that the material is a promising candidate for high-temperature quantum spin ice if it is fabricated on a proper substrate. After a submission of the manuscript, it has been required by referees to strengthen the case by checking the dependence of the results on different LDA+U schemes.

ii) Furthermore, it remains to be an open question, how one can tune the electronic structure through the substrate in order to host the U(1) QSL, which hosts a U(1) QSL with deconfined bosonic spinons carrying emergent magnetic monopole charges. However, the presence of an interface and/or strain from the substrate may alter the physics from that realized in the bulk systems. Therefore, it is now called for to clarify whether the bulk properties of high-temperature quantum spin ice may survive in the presence of the interface. Therefore, it is important to understand the electronic structure in the presence of the interface. One of the simplest example is a superlattice of the band-insulating $ZnIr_2O_4$ and the Mott-insulating $Ir_2O_4$.

iii) We also planned to investigate a so-called quantum spin ice model, which describes low-energy magnetic properties of magnetic rare-earth pyrochlore oxides as well as $Ir_2O_4$, under an applied electric field. Using the gauge theory description of the system in terms of spinons coupled to U(1) gauge fields, we have already shown that the electric field gives rise to an effective gauge flux, penetrating each hexagonal plaquette, for the spinons. Then, the model has to be solved numerically for a set of rational numbers of the flux, as studied in the case of the Hofstadter problem for electrons forming Landau levels on lattices. This may offer a unique intriguing phenomenon of spinons carrying monopole charges of the magnetization under the electric field.

2. Specific usage status of the system and calculation method

We used the GWMPC system for 3.4M CPU hours by February 12th.

i, ii) We used the OpenMX package for the LDA and LDA+U calculations for $Ir_2O_4$ as well as superlattices of $Ir_2O_4$ and $ZnIr_2O_4$.

iii) We performed numerical exact diagonalizations of the bilinear spinon Hamiltonian on the diamond lattice dual to the original pyrochore lattice, in the presence of the uniform gauge flux $\phi$ that penetrates each hexagonal plaquette in the (001) direction and takes various rational numbers. Then, we computed the spinon spectra forming Landau levels.

3. Result

i) We have performed LDA+U calculations on the ideal cubic $Ir_2O_4$ in the Lichtenstein form for the exchange-correlation functional, which depends only on the local charge density, with a finite onsite Hund's-rule coupling constant $J_H$=0.65 eV. The total energy differences of several (meta)stable magnetically ordered solutions from the paramagnetic one are plotted in Fig.1(a). When these results are viewed as a function of an effective Coulomb repulsion U'=U-$J_H$, they are rather similar to those obtained in the Dudarev form for the exchange correlation functional, which depends on the local spin and charge densities. Note that in most of the materials studied so far, the Lichtenstein form shows a better agreement with experiments. Hence the above results indicates that our results, obtained by using the Dudarev form in Q17307, on high-temperature quantum spin ice in $Ir_2O_4$ are indeed trustworthy. Actually, in the current case, only one electron per Ir site is directly relevant to Mott physics. In this case, it is rather reasonable that the onsite Hund's-rule coupling can be simply renormalized into U. This can be clearly in Fig.1(b), which shows a comparison between the electron band dispersions obtained in the two different schemes. The results well agree with each other for the eight bands near the chemical potential 0, while they differ for the other bands deeply below the chemical potential.

ii) We have performed large-scale LDA and GGA calculations to obtain the stable crystal structures of superlattices of $ZnIr_2O_4$ and $Ir_2O_4$ on the MgO(100) substrate. At the moment, however, the calculations are restricted only to the case with the smallest superlattice unit cell, comprising stacked $2ZnIr_2O_4$ and $2Ir_2O_4$ in the (100) direction. And the stable crystal structure has not been determined yet, because of a slow convergence and longer computational time for the larger system size. We originally planned to study the cases with larger superlattice unit cells and to reveal the electronic structures with a finite onsite Coulomb repulsion U and Hund's-rule coupling $J_H$. Thus, this work needs to be continued in the next fiscal year.

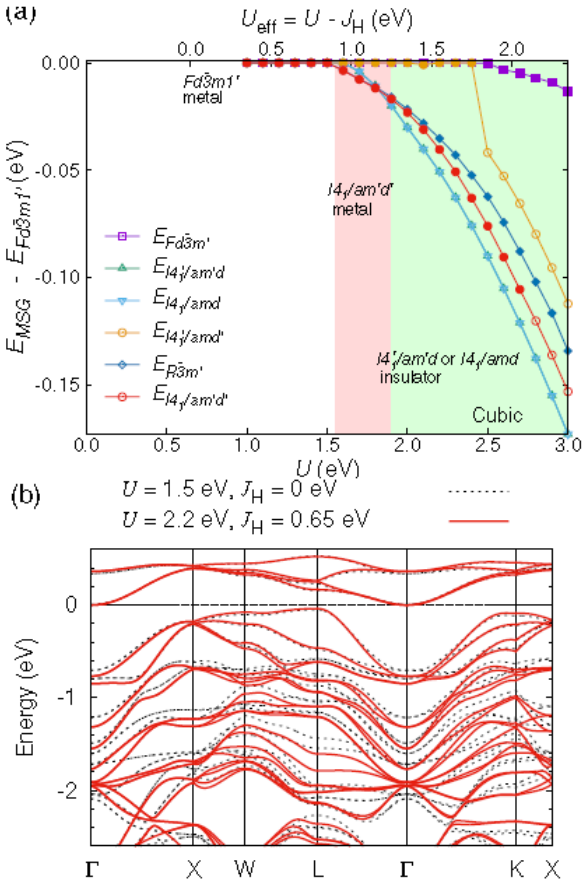


Fig.1. Energy differences of six magnetically ordered solutions from the paramagnetic one in the LDA+U calculations in the Lichtenstein form with $J_H$ = 0.65 eV. Filled and open symbols stand for (semi)metallic and insulating states, respectively. (b) Comparison in the electronic band structures of the ideal cubic spinel iridate $Ir_2O_4$. Black and red curves represent the LSDA+U results with U = 1.5 eV and $J_H$ = 0 and the LDA+U results with U = 2.2 eV and $J_H$ = 0.65 eV (red curves) in the antiferromagnetically ordered ($I4_1'/am'd$) ground states. The results are from [S. Onoda, F. Ishii, Phys. Rev. Lett. 122, 067201 (2019)].

iii) Spinon spectra for the quantum spin ice model have been obtained in the case where the flux is given by rational numbers, i.e., $\phi=2\pi p/q$, with integers $p$ and $q$. In each case, there appear $q$ spinon bands with Weyl points in the Brillouin zone that is folded in $q$. In particular, the Weyl points mediate a transfer of Chern numbers for spinon wavefunctions. Figure 2 shows a specific example for $p$=2 and $q$=3 in the simplest quantum spin ice model, namely, the spin-1/2 XXZ model with ferromagnetic $J_{xx}$ and antiferromagnetic $J_z$.

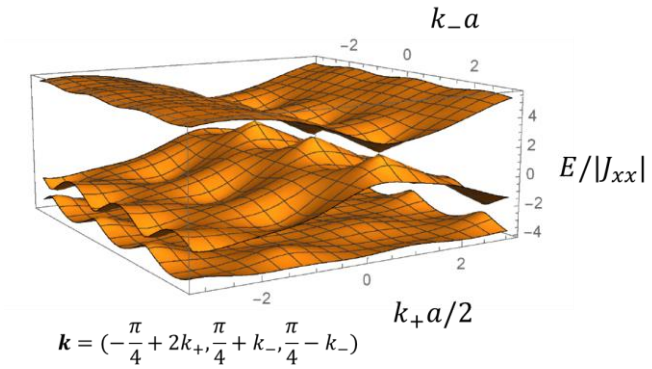


Fig.2. Energy band dispersions of spinons in the case of $\phi=4\pi/3$ in a certain gauge. The chemical potential is determined to satisfy the local constraint on the spinon density. (a: the cubic lattice constant.)

## 4. Conclusion

i,ii) The A-site de-intercalated spinel iridate $Ir_2O_4$ should behave as high-temperature quantum spin ice, where the spin ice rule sets in at room temperature. In particular, when it is grown as epitaxial thin films on a proper tetragonal substrate, it provides a promising candidate to host a U(1) QSL. Superlattices and an interface of $Ir_2O_4$ and $ZnIr_2O_4$ are chemically stable and free from charging effects, and may provide a nice laboratory for fabricating high-temperature quantum spin ice material.

iii) Quantum spin ice under an applied electric field can host a U(1) QSL with topologically nontrivial spinon bands that show a transfer of their Chen numbers through Weyl points.

## 5. Schedule and prospect for the future

In this fiscal year, referee comments forced us to perform more intensive and extensive first-principles calculations using various LDA+U schemes, to show that the results are really trustworthy. As the result, we could not finish all the first-principles calculations on superlattices of $ZnIr_2O_4$ and $Ir_2O_4$. We plan to perform the remaining calculations in the next fiscal year.

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## Fiscal Year 2018 List of Publications Resulting from the Use of the supercomputer

### [Paper accepted by a journal]

Shigeki Onoda, Fumiyuki Ishii, First-principles design of the spinel iridate $Ir_2O_4$ for high-temperature quantum spin ice. Physical Review Letters **122**, 067201 (2019).

### [Oral presentation]

Shigeki Onoda, Magnetic monopoles in quantum spin ice under an electric field. International Conference on Highly Frustrated Magnetism 2018 (July 10-14, 2018, Univ. California, Davis, USA).

Shigeki Onoda, Transport properties of magnetic monopoles in quantum spin ice. 1st-NCKU-RIKEN Joint Workshop on Quantum Topological Materials (January 10-12, 2019, Tainan, Taiwan). Invited.

Sho Nakosai, Shigeki Onoda, Magnetic monopole supercurrent through a quantum spin ice junction. 1st-NCKU-RIKEN Joint Workshop on Quantum Topological Materials (January 10-12, 2019, Tainan, Taiwan). Invited.

### [Poster presentation]

Shigeki Onoda, Quantum spin ice under an electric field. International Conference on Magnetism 2018 (July 16-20, 2018, San Francisco, USA).

### [Others (Book, Press release, etc.)]

Press release (in Japanese) 「単極子を制御できる新たな物質」 (February 12, 2019).

English version, to be released.