Usage Report for Fiscal Year 2024

Project Title:

Point defect engineering in functional metal oxides.

Name:

○ Supparat Charoenphon(7,2), Dita Puspita Sari(1,2), Wan Nurfadhilah Binti Zaharim(3,2), Muhammad Redo Ramadhan(4,2), Irwan Ramli(5,2), Phantip Jaikaew(6,2).

Laboratory at RIKEN:

(1) Shibaura Institute of Technology, Japan

(2) Nuclear Structure Research Group, RIKEN Nishina Center, Japan

(3) Universiti Kebangsaan Malaysia, Malaysia

(4) UPN Veteran, Yogyakarta, Indonesia

(5) Universitas Cokroaminoto Palopo, Indonesia

(6) Chiang Mai University, Thailand

(7) Kasetsart University, Thailand

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

Neodymium-based iridate, $Nd_2Ir_2O_7$, has drawn significant interest due to the interplay between strong spin-orbit coupling (SOC) and electron-electron interactions, leading to intriguing phenomena such as the metal-insulator transition (MIT), unconventional superconductivity, the magnetoelectric effect, and Weyl semimetal phases. Its pyrochlore lattice induces geometrical frustration, resulting in complex magnetic interactions between the 4f electrons of rare-earth ions and the 5d electrons of transition metals. Recent studies on the $R_2Ir_2O_7$ family show a systematic decrease in MIT temperature with increasing ionic radius of R, vanishing between Nd and Pr. Initially thought to be metallic at all temperatures, improved sample quality revealed an MIT at 37 K with a 45 meV energy gap. Reports on its magnetic order vary, with some suggesting an all-in-all-out (AIAO) structure for Ir^{4+} and Nd^{3+} moments (0.34 μB and 1.27 μB at 1.8 K), while others propose an antiferromagnetic long-range order with a propagation vector of (0,0,0) and higher Nd moments.

Density Functional Theory (DFT) is an effective tool for studying such systems, where SOC and the on-site Coulomb potential (U) play a crucial role. Previous work shows that GGA better describes electronic states in strongly correlated materials than LDA or PBE. However, LDA+U tends to overestimate insulating magnetic states. Prior studies using GGA+SOC predicted a metallic state in $Y_2Ir_2O_7$ and $Pr_2Ir_2O_7$ but an insulating state in $Eu_2Ir_2O_7$, while GGA+SOC+U opened a band gap in $Y_2Ir_2O_7$ and $Eu_2Ir_2O_7$, aligning with experimental results. For $Nd_2Ir_2O_7$, previous DFT and dipole-field calculations underestimated the magnetic moments of Ir and Nd, highlighting the need for improved methods. In this study, we used GGA+SOC+U to systematically vary U (0–8 eV) and examine its impact on structural and electronic properties, aiming to determine the ground state of $Nd_2Ir_2O_7$ and reproduce experimental results as accurately as possible.

2. Specific usage status of the system and calculation method

DFT calculations were performed using the GGA scheme with HOKUSAI within VASP software. A 400 eV energy cutoff was used for plane waves. DFT+U and SOC effects were included on Nd. A simple unit cell (space group Fd-3m, 88 atoms) was used, with structural relaxations converged to residual forces < 0.02 eV and an energy threshold of 10^{-5} eV. Lattice parameters and band gaps were calculated using a 22-atom primitive cell with a 7×7×7 MP k-point mesh. Magnetic properties of Ir and Nd were studied under NM, FM, and AFM

configurations with SOC on Nd, using an 88-atom supercell and a non-centered MP 2×2×2 k-point mesh.

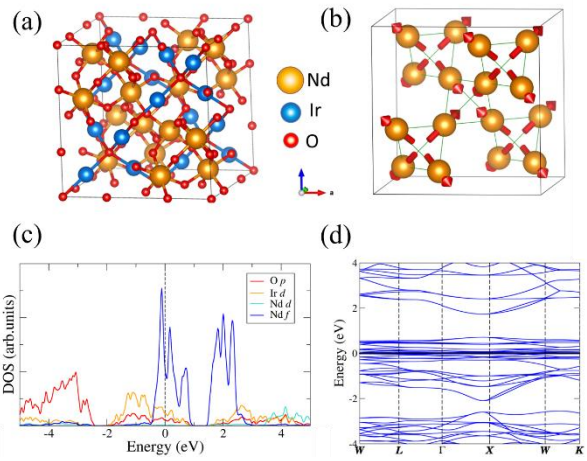

Figure 1. (a) An illustration of $Nd_2Ir_2O_7$ cubic cell containing 88 atoms and the its local atomic structure (b) Model of All-in all-out magnetic structure of $Nd_2Ir_2O_7$. The orange balls represent Nd3+ ions, and the red arrows represent the direction of Nd magnetic moments. (c)The GGA+SOC calculated projected density of states (PDOS), (d) Electronic band structure of $Nd_2Ir_2O_7$ show the first Brillouin zone with the path W-L--X-W-K

3. Electronic and Magnetic Structure

The $Nd_2Ir_2O_7$ structure belongs to the cubic Fd-3m space group, with Nd3+ ions forming distorted body-centered cubic bonds to O^{2-} atoms and Ir^{4+} ions forming corner-sharing IrO_6 octahedra. The magnetic moments of both Ir and Nd ions exhibit AIAO structures, with both ferromagnetic and antiferromagnetic configurations possible. Density of states (DOS) and band structure calculations in the GGA+SOC approximation show that the O-*p*, Ir-*d*, and Nd-*f* states contribute near the Fermi level. However, the material shows metallic behavior, contradicting experimental reports suggesting a Mott insulator. To address this, we apply the GGA+SOC+*U* method.

3.1 U_d= 0, U_f= 0 (Ir only)

Applying Hubbard *U* (U_d) to the Ir atoms affects the d orbital energy levels. From U_d = 0 to 3 eV, the O-*p* and Ir-*d* states become more localized near the Fermi energy. At U_d= 5-7 eV, the 5*d* states hybridize with the O-2*p* orbitals, but the Nd-*f* states remain unaffected, preventing the formation of a band gap. The calculated lattice parameters and magnetic moments are compared to experimental values, showing a maximum percentage error of 1.57% for U_d= 8 eV. The best agreement with neutron diffraction data occurs at U_d= 6 eV, with Ir magnetic moment increasing with U_d.

3.2 U_d= 0, U_f= 0 (Nd only)

Applying Hubbard U (U_f) to the Nd atoms affects the f orbitals. The Nd-*f* states near the Fermi level decrease as U_f increases, but the Ir-*d* and O-*p* states remain unaffected. Despite these adjustments, the band gap remains metallic, similar to the case of U_d. The lattice parameters show a small deviation from experimental values, and the Nd magnetic moment is higher than the experimental value of 2.3 μB.

3.3 Combined U_d and U_f

Adjusting both U_d (5 eV) and U_f(5-8 eV) opens a band gap, with the best agreement with experimental data occurring at U_d = 5 eV and U_f = 5 eV, producing an indirect band gap of 1.58 eV. The lattice parameter values are in good agreement with experiment, with a maximum error of 0.29%. The magnetic moment of Ir is close to the experimental value, but the Nd magnetic moment remains higher than the experimental value. The lowest energy configuration for $Nd_2Ir_2O_7$ is ferromagnetic when both Ud and Uf are adjusted around 5 eV.

4. Conclusion

The electronic structures and magnetic properties of $Nd_2Ir_2O_7$ were studied using GGA+*U* and spin-orbit coupling (SOC). It was found that using Hubbard *U* values of 5 eV for Nd-4*f* and Ir-5*d*

orbitals helps open a gap from metal to insulator. The calculations show a stable ferromagnetic phase with Ir^{4+} magnetic moment of 0.32 μB and Nd^{3+} magnetic moment of 3.00 μB, which is higher than the experimental value.

5. Schedule and prospect for the future

Next, we will compare computational results with experimental muon spin rotation (μSR) data to validate the predictions. The model will then be refined to optimize muon implantation and study the impact on the material's electronic and magnetic properties. Ultimately, this research will open avenues for applying muon spectroscopy to probe unique behaviors in $Nd_2Ir_2O_7$ and other related materials, contributing to the advancement of condensed matter physics.

6. If no job was executed, specify the reason.

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Usage Report for Fiscal Year 2024

Fiscal Year 2024 List of Publications Resulting from the Use of the supercomputer

[Conference Proceedings]

1. **Charoenphon, S**., Widyaiswari, U., Syakuur, M. A., Asih, R., Reunchan, P., & Watanabe, I. (2024). Structural, electronic, and magnetic properties of pyrochlore $Nd_2Ir_2O_7$ studied by first-principles calculations. Interactions, 245(1), 73.

[Oral presentation]

1. **Supparat Charoenphon**, Utami Widyaiswari, Retno Asih, Pakpoom Reunchan and Isao Watanabe "Structural, electronic, and magnetic properties of pyrochlore Nd2Ir2O7 studied by first-principle calculations" at 1st Conference of Accelerator-Based Sciences and Technology (CAST 2024). National Research and Innovation Agency of Indonesia (BRIN), Indonesia: Oral presentation

2. **Supparat Charoenphon**, Pakpoom Reunchan and Isao Watanabe "First principal calculation of the magnetic properties and muon stopping site in metal oxide with different Electronic correlational Functionals" at The 2024 Muon User Meeting. ISIS Neutron and Muon Source, Oxford, UK: Oral presentation

[Poster presentation]