Experimental report

12/03/2021

Proposal: **5-31-2793** **Council:** 4/2020

Title: Neutron diffraction studies on themagnetic structure of Re2ZnIrO6

Research area: Physics

This proposal is a new proposal

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Experimental team:

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Samples: Nd2ZnIrO6
La2ZnIrO6

Instrument	Requested days	Allocated days	From	To
D20	2	2	26/08/2020	27/08/2020
			05/09/2020	06/09/2020

Abstract:

Double perovskite with 5d transition metal at the B site, namely, Re2ZnIrO6 (Re = rare earth) show complex magnetism due to the interplay between the 4f electrons and the 5d electrons with large spin-orbit coupling. The small size of Ir4+ moments and the relatively large neutron absorption of Ir, however, hinder the detailed determination of the magnetic structure. Thanks to the extremely high flux of the D20 diffractometer, we propose to study two characteristic compounds with Re = La and Re = Nd in order to reveal the nature of the magnetic ground state, especially the electronic state of the interesting Ir4+ ions. In order to fulfill all our purposes, we ask for 2 days beamtime on the D20 diffractometer.

Neutron diffraction studies on the magnetic structure of RE_2ZnIrO_6

Double perovskite iridate R_2ZnIrO_6 show complex magnetic behaviors ranging from weak ferromagnetism to successive magnetic transitions at low temperatures. Due to relatively large neutron absorption by the naturally abundant Ir atoms and the small moment size of Ir^{4+}, previous studies were not able to resolve the magnetic structure including the Ir sublattice unambiguously.

In this experiment [1], we have prepared two high quality powder samples of Nd_2ZnIrO_6 and La_2ZnIrO_6. Thanks to the high flux of the D20 diffractometer, we managed to observed the magnetic signal which can be explained only if the Ir sublattice is taken into account. Experimentally, about 7 g samples were loaded into a double-wall vanadium can. The R = La sample was measured for 10.25 h at 1.8 and 20 K, respectively. Due to the stronger magnetic signal from the Nd_2ZnIrO_6 sample, 2-hour measurements at 1.8, 13.5, 15.1 and 19.3 K, were performed. In addition, a series of 15-min measurements were carried out while ramping the temperature at 0.1 K/200sec and 0.1 K/450sec between 1.8 - 12 K and 12 – 20 K respectively. These data allowed the temperature dependence of the magnetic peak intensities to be determined, which is proportional to the magnetization squared.

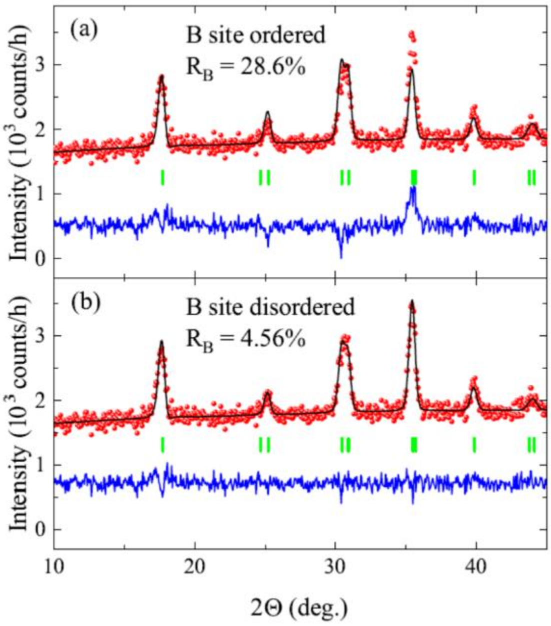

Fig. 1 Rietveld refinement of the magnetic diffraction from La_2ZnIrO_6 at 1.8 K. A paramagnetic data set, taken at 20 K has been subtracted to remove the scattering from the crystal lattice. The calculated patterns are according to the IR Γ_3 with (a) a perfectly ordered B-site model and (b) a B-site disordered model where the best model found 13(4)% site substitution between Zn and Ir ions.

The low temperature magnetic intensities were extracted by subtracting the paramagnetic signal measured at ~20 K. The magnetic peaks can be indexed with a k = 0 and k = (0 0 1/2) propagation vector for the R = La and Nd sample, respectively. Magnetic symmetry analysis for the R = La sample shows that the magnetic representation is decomposed into two irreducible representations (IRs) as $\Gamma = \Gamma_1 + \Gamma_3$. Rietveld magnetic refinements show that the magnetic structure can be described well by the IR Γ_3, which corresponds to a magnetic structure with the Ir moments confined within the bc plane, and with a nonzero ferromagnetic component along the c axis. Moreover, there is a site mixing between the B' and B'' site, namely, between the Zn and Ir site for the R = La sample, which amounts to about 13(4)% from the magnetic structure refinement, and about 6(4)% from the nuclear structure refinement on the D2B data set. The best refinement is shown in Fig. 1 and the magnetic structure is shown in Fig. 2(a).

For the R = Nd sample, DC magnetic susceptibility measurements exhibit two successive transitions at ~17 and ~13 K. However, our neutron diffraction

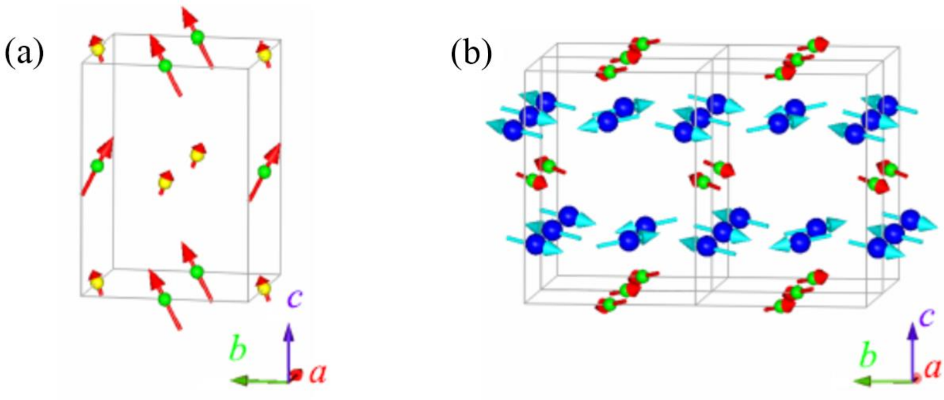

Fig. 3 Representive magnetic structure of (a) La_2ZnIrO_6 and (b) Nd_2ZnIrO_6.

measurements show no anomaly at the second transition temperature. Refinement results at three different temperatures are shown in Fig. 3, where the inclusion of both the Nd and Ir sublattices is necessary in order to account for the experimental data, as can be seen from the expanded region in the right column. The corresponding magnetic structure is shown in Fig. 2(b). As can be seen, the magnetic easy plane is rotated from the bc plane for the R = La sample to the ab plane for the R = Nd sample, possibly due to the anisotropy of Nd moments. Moreover, the moment size of the Ir ions at 15 K is already about 70% of the low temperature value, while that is only about 39% for the Nd ions, indicating that the Ir sublattice is the driving force of the magnetic ordering. The increase in the transition temperature by about 70% for the R = Nd sample compared to that of the R = La sample, on the other hand, showing the importance of the 5d-4f interactions. Together with Dzyaloshinskii-Moriya interaction and crystal field schemes, these comparable energy scales within the double perovskite iridate results in complex magnetic behaviors, which will deserve further investigations.

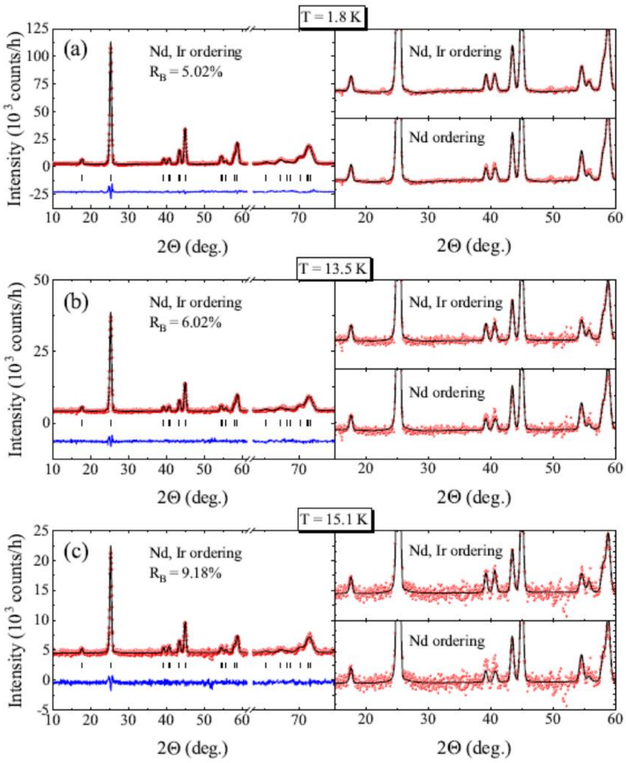

Fig. 2 Rietveld magnetic refinement of the difference pattern at various temperatures for Nd_2ZnIrO_6 according to IR $\Gamma_1 + \Gamma_3$. The left panels show the refinement results with perfectly ordered Nd and Ir sublattices, and the right panels highlights an expanded region of low peak intensities comparing our best model and that with only the Nd sublattice ordering.

References:

[1] H. Guo et al., Phys. Rev. B 103, L060402 (2021).