Experimental report

22/09/2022

Proposal: **4-05-841** **Council:** 4/2021

Title: Magnetic excitations in Na2IrO3

Research area: Physics

This proposal is a continuation of 4-05-790

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Samples: Na2IrO3

Instrument	Requested days	Allocated days	From	To
IN8	5	5	27/09/2021	02/10/2021

Abstract:

INS experiments on IN8 and Thales have been conducted on the Kitaev candidate material Na2IrO3, allowing to characterize the low energy AFM magnon dispersion and to determine the magnon gap. Looking at a Kitaev signature, no magnetic signal has been found at low energy at the 2D zone center and only a weakly dispersive feature has been found at 23meV. The understanding of the magnetic excitations requires a complex Hamiltonian, whose numerous exchange parameters cannot be unambiguously determined. So far, due to the large activation of the sample, only measurements in the (0kl) scattering plane have been conducted. We propose to continue investigating the magnetic excitations in Na2IrO3 with a different scattering plane, which will help not only to discriminate the large parameters sets, but also facilitate the interpretation of magnetic signals in both INS and RIXS measurements.

Experimental Report

Instrument	IN8
Proposal Number	4-05-841
Proposal	Magnetic excitations in Na2IrO3
Experimentalist	Alexandre Bertin, Markus Braden
Local Contact	Andrea Piovano

The honeycomb iridates attract considerable interest as a potential model of what is called now the Kitaev model [1,2]. In this model, magnetic ions occupy a honeycomb lattice and the interaction between two nearest neighbors is bond directional. The beauty of this model and concept lies in the fact that it can be exactly solved [2] giving a topological quantum spin liquid (QSL) ground state and exotic excitations with a possible impact for quantum computing. Therefore, there is an ongoing race to realize this model in a material, and Na_2IrO_3 is one of the first candidates for such a system. Na_2IrO_3 as well as the other discussed realizations, however, exhibits long-range antiferromagnetic order, found to be of the zigzag type [3-7], below ~15K which points to additional magnetic interaction parameters beyond the pure Kitaev interaction. X-ray studies analyzing either the diffuse scattering [5] or the resonant inelastic signals [4,7] claim evidence for a dominating Kitaev interaction. On the other hand there is a series of papers on INS of the honeycomb α-$RuCl_3$ [6], which finds the clear signature of the Kitaev interaction superposed to the antiferromagnetic magnons. More recently the progress in RIXS at the Ir edge permitted measurements of the magnetic excitations in Na_2IrO_3 [7] but the energy resolution is still limiting. A singular experiment with the best RIXS resolution of 12meV [7] yields dispersive features that cannot easily be attributed to a fully dominant Kitaev interaction. In our previous experiments on Na_2IrO_3 on IN8 and on Thales we could observe antiferromagnetic magnons by INS in spite of the difficult experimental conditions. We could determine the low-energy dispersion for the direction parallel to the in-plane component of the magnetic Bragg reflections and perpendicular to the layers. There is no dispersion visible perpendicular to the layers and the magnon gap amounts to 1.75 meV which is unexpectedly low for a supposedly very anisotropic system. The low-energy magnon response is located near the antiferromagnetic Bragg peaks, which contrasts to the findings in α-$RuCl_3$ which show the strongest magnetic response at the ferromagnetic scattering vectors [6]. Our results are thus in agreement with the recent theoretical analysis proposing that the Heisenberg interaction even differs in sign between α-$RuCl_3$ and Na_2IrO_3 [8] and that the ferromagnetic-like signals in α-$RuCl_3$ are not fully due to the Kitaev interaction.

The aim of this new experiment was to explore the magnon in the second in-plane direction, i.e. at 30 degrees to that previously studied. Here one reaches the **K** point, for which the RIXS experiments claim a strong response [7]. The same sample – 63 H-free glued crystals on an Aluminum plate with thicknesses mostly below 1mm limiting neutron absorption – but a different scattering plane defined as (100)/(001) have been used (in the monoclinic *C2/m* notation). However, it turned out that, due to an imperfect atmosphere, the sample considerably deteriorated during the storage at ILL between this and the previous experiment, while similarly long periods did not have any impact before. In addition the experimental conditions concerning the background were not optimum. In the previous experiments we got much lower background by using an orange-type cryostat with a larger isolation vacuum that was not available during the experiment. A considerable amount of beam time needed to be invested to improve the background.

In the (100)/(001) scattering plane one does not find a strong magnetic Bragg peak but can nevertheless reach low-energy antiferromagnetic magnons near Q_h=1. We do find some signal that can be attributed to the antiferromagnetic magnons steeply dispersing near this magnetic zone center. But this signal is rapidly lost in constant energy scans at higher energy transfer, see Fig. 1.

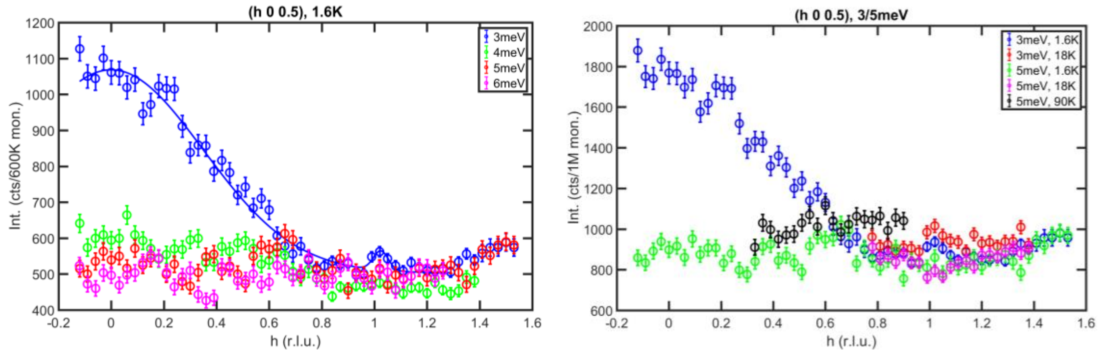

Fig. 1: *Constant-energy scans (h, 0, 0.5) with closed slits at 3, 4, 5, and 6 meV; there is a small signal visible at the AFM peak that is rapidly lost.*

The main part of the experiment was dedicated to determine the magnetic signal at the **K** point corresponding to (0.666, 0, q_l) for which the RIXS experiments yielded a strong signal, see reference [7]. This is supported by our linear spin-wave calculations with the *SpinW* code for the twinned system. These calculations show a degeneracy of magnon branches leading to a strong response at **K**. In the raw data of the constant-Q scans it was not possible to clearly identify such a magnon signal because the background was too high. We therefore recorded not only the scans at various (0.666, 0, q_l) and (1.333, 0, q_l) but also by rotating the sample away from this position in both directions where possible. These results are shown in Fig. 2.

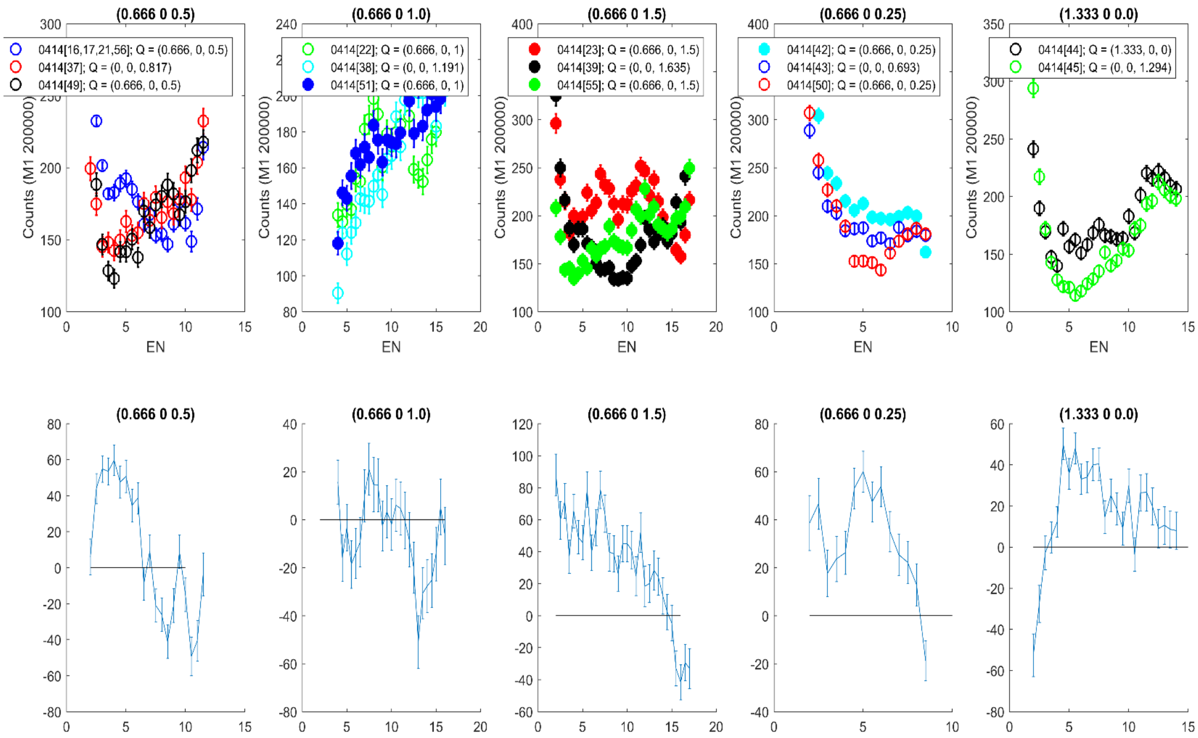

Fig. 2: *Constant Q-scans at two K points in reciprocal space,* (0.666, 0, q_l) *and* (1.333, 0, q_l) *in orthorhombic (or monoclinic) notation. Besides the scans at K we also performed scans by considerably rotating the sample in both directions where possible. One background scan was performed at (0,0,l) with the same absolute value of the scattering vector. The other background scan was performed by changing the zero of A3 (lowest line in legend). The lower panels give the differences between the intensity at K and at the background positions.*

Due to the rather large rotation angles between the scans at the **K** points and at the background positions one may not expect to obtain a precise determination of the background arising from the sample as well as from the environment. Nevertheless the combined data indicate a magnetic signal at the **K** point to appear at 5-7meV which roughly agrees with our preliminary magnetic model.

A similar analysis was also performed at the antiferromagnetic zone center (1, 0, 0) and yields a signal consistent with the steep magnon dispersion and the qualitatively much better results obtained in the scattering geometry of the previous experiments.

Our studies indicate that inelastic neutron scattering on iridates using a multi-crystal sample are possible on the most powerful triple-axis spectrometers at the ILL (both thermal and cold neutrons), but great care is needed to suppress the background for measuring the tiny signals.

References

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