Experimental report

14/02/2021

Proposal:	**4-05-790**	**Council:**	4/2020
Title:	Kitaev excitations and magnons temperature behaviour in Na_2IrO_3		
Research area:	Physics		

This proposal is a new proposal

Main proposer: **Alexandre BERTIN**

Experimental team: Alexandre BERTIN

Local contacts: Alexandre IVANOV

Samples: Na_2IrO_3

Instrument	Requested days	Allocated days	From	To
IN8	7	8	04/09/2020	12/09/2020

Abstract:

We recently succeeded in performing INS experiments on the magnetic excitations on Na_2IrO_3, which in spite of its AFM ground state is considered to be a candidate for the Kitaev model on a honeycomb lattice. We propose to extend these experiments by studying higher energies and a second scattering plane and by measurements at higher temperatures where Kitaev type correlations should be relatively enhanced.

Experimental Report

Instrument	IN8
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Proposal	Kitaev excitations and magnons temperature behavior in Na2IrO3
Experimentalist	Alexandre Bertin, Markus Braden
Local Contact	Alexander Ivanov

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 was one of first proposals for such a system. Na_2IrO_3 as well as the other discussed candidates, 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. This IN8 experiment is a continuation of experiment 4-05-759. The same sample – 63 H-free glued crystals on an Aluminum plate with thicknesses mostly below 1mm limiting neutron absorption – and same scattering plane defined as (010)/(001) have been used. Magnetic Bragg peaks appear at (0 1*L*) with *L* half-integer in the monoclinic *C2/m* notation. The aim of this experiment was to look for the Kitaev signature: a broad high-energy mode expected to appear at the 2D zone center, whose intensity does not drop or even strengthens with temperature.

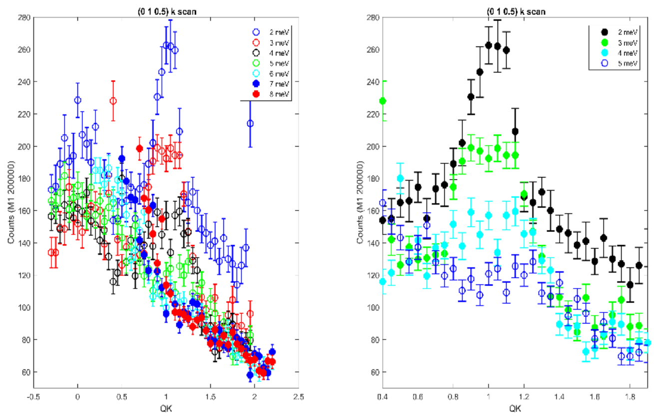

Fig. 1: *Constant energy scans up to ΔE=8meV along the K direction across the AFM Bragg peak at (0 1 0.5) at T=1.5K. The dispersing magnons can be detected up to 5meV at this* ***Q*** *vector.*

First of all, the background has been significantly improved compared to the previous IN8 experiment where a large background arose at small Q values and made the magnon analysis difficult. A new cryostat with a larger isolation vacuum and an additional diaphragm placed between the sample and the secondary spectrometer has significantly reduced the background related to air scattering by a factor of up to 10, therefore significantly enhancing the data quality of the low energy measurements at 1.5K. Constant energy scans along *K* across the 2D magnetic zone center at (0 1 *L*) with *L* =0.5 and 1.5 are shown in Fig.1 and Fig. 2, respectively, additional scans were taken at *L*=1. Magnon excitations could be resolved up to ~14meV for *L*=1.5. With increasing energy, it is necessary to study larger *L* values, because otherwise the small scattering angle cause a drastic increase in background. The constant energy *K*-scans have been extended up to the zone center, where the Kitaev signal is expected. For *L*=0.5, the increased signal with increasing energy transfer seen at low ***Q*** still comes from the proximity of the direct beam. For *L*=1 and 1.5, the higher signal at the zone center is likely due to air scattering, increasing with higher incident energy. These long scans show an intrinsic difference to the INS results reported for α-$RuCl_3$ [6], which find clearly dominating scattering at the 2D zone center that is attributed to a dominating Kitaev interaction. In Na_2IrO_3 these low-energy scans do not find clear signatures of the Kitaev bond directed interaction; the strongest magnetic excitations are doubtlessly stemming from the zigzag antiferromagnetic structure that is stabilized through Heisenberg exchange at larger distances.

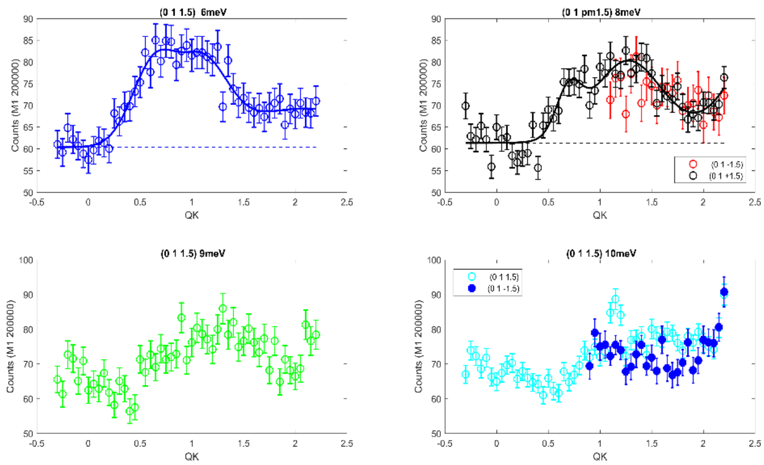

Fig. 2: *Constant energy scans up to ΔE=10meV along the K direction across the AFM Bragg peak at (0 1 1.5) at T=1.5K. The dispersing magnons can be detected up to ~14meV at this **Q** vector. These scans were performed with a* PG002 *monochromator and k_f=2.662Å^{-1}.*

This significant difference between the INS scattering for α-$RuCl_3$ [6] and for Na_2IrO_3 were corroborated by scanning with larger final energy which strengthens the relative weight of an excitations that is less defined in energy or Q space, these scans are shown in Fig. 3, but again the moderate intensity uptake around the 2D zone center can mostly be attributed to the increasing background. (In our previous experiment this issue was further studied by scans with a rotated sample.)

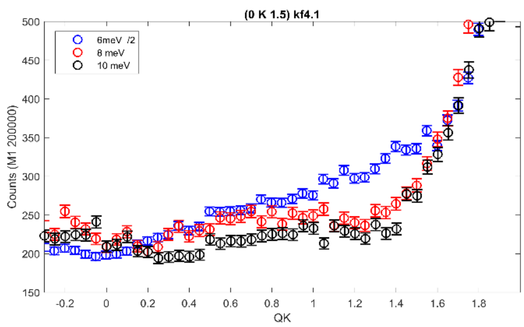

Fig. 3: *Constant energy scans up to ΔE=10meV along the K direction across the AFM Bragg peak at (0 1 1.5) at T=1.5K. These scans were performed with a* PG002 *monochromator and k_f=4.1Å^{-1} and thus emphasize any broader contribution.*

The temperature dependence of the antiferromagnetic magnon scattering is shown in Fig. 4. The magnons become rapidly broad but the clear signatures remain visible up to at least 100K. There is no qualitative change occurring that can be attributed to Kitaev excitations with a more robust temperature dependence. Even at 100K the antiferromagnetic magnon scattering dominates the low-energy response.

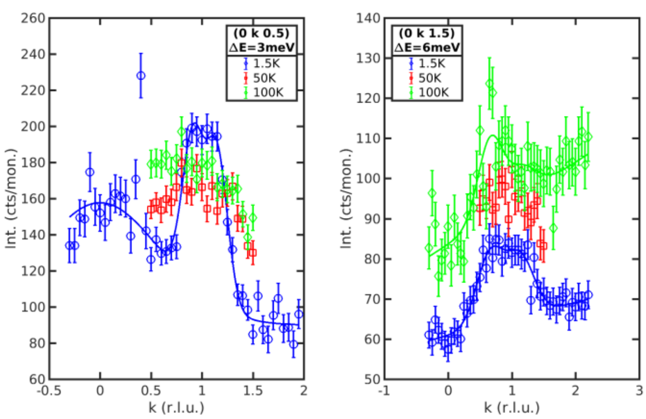

Fig. 4: *Temperature dependence of constant energy scans at (0 1 0.5) and 3meV, and at (0 1 1.5) and 6meV. The magnon response does not change qualitatively up to 100K.*

The second part of the experiment was dedicated to the investigation of the Kitaev magnetic signal or more generally to any signal with an essentially local character. This part of the experiment is by far more challenging, because the higher energies require a larger scattering vector which reduces magnetic scattering and enhances phonon contributions. Therefore, it is very difficult to isolate a weak magnetic signal. A wide energy scan at Q= (0 1 1.5) up to 37meV has been carried out combining k_f=2.662Å^{-1} and 4.1Å^{-1} at base temperature and at 100K. Background data sets have been measured by rotating the sample by 30° and subtracted from the raw data. These difference spectra did not yield any indication for an additional magnetic contribution. Moreover, energy scans at the 2D zone center have been performed for several (00*L)* values at base temperature, some scans are displayed in Fig.5 where background data sets have been subtracted. A broad signal is observed in the differences around 23meV. At *L*=1.8, data measured with the PG002 (k_f=4.1Å^{-1}) and the Si monochromator (k_f=2.662Å^{-1}) perfectly agree, ruling out a spurious origin. Furthermore, energy scans at constant |***Q***|, but at different reciprocal lattice points, show that the signal survives while getting away from the 2D zone center, see Fig. 5. Finally, the temperature comparison of the energy scans measured at the 2D

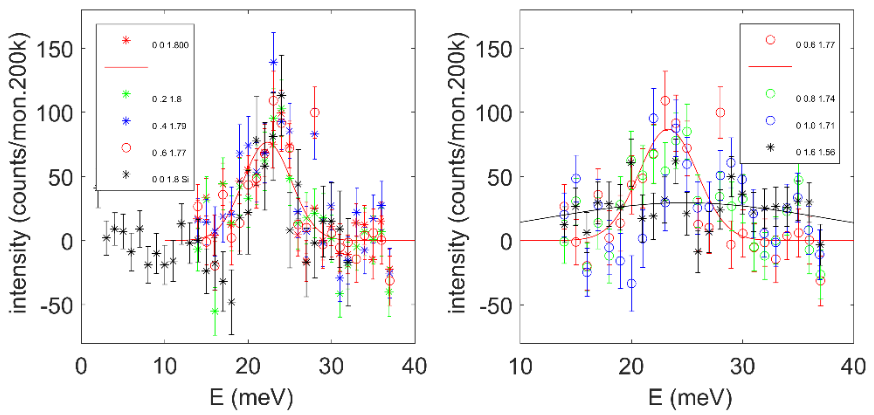

Fig. 5: *Energy scans at **Q**=(0 0 1.8) and at positions with the same absolute |Q| value. Background was determined by rotating the sample by 90° and subtracted from the data. There is evidence for a scattering contribution at ~23meV that is little Q dependent.*

zone center at (0 0 1.8) and at the same absolute |***Q***|-value, but away from the zone center at (0 1 1.71), show that the signal intensity grows with temperature at both reciprocal lattice points and even survives up to room temperature. While the signal is a candidate for flat magnetic excitations a reliable interpretation requires further information.

Finally, due to the large activation of the sample after only a few days, it was impossible to conduct the experiment in the rotated scattering plane, and this aspect will be hopefully the purpose of a continuation on IN8. In conclusion the continuation of INS experiments on single-crystalline Na_2IrO_3 improved the characterization of the low-energy dispersion of antiferromagnetic spin-waves in the ordered state. Unlike α-$RuCl_3$, no clear evidence of magnetic excitations related to the Kitaev interaction has been found neither at low nor at higher energies, and the low-energy scans reveal a clearly different hierarchy of magnetic interaction in Na_2IrO_3.

References

[1] G. Cao et al., Rep. Prog. Phys. **81**, 042502 (2018) **[2]** H. Takagi et al., Nature Reviews Physics **1**, 264 (2019) **[3]** Ye F *et al.*, Phys. Rev. B **85** 180403 (2012) ; Choi S K *et al.*, Phys. Rev. Lett. **108** 127204 (2012) **[4]** A. Revelli *et al.*, Phys. Rev. Research **2**, 043094 (2020) **[5]** S.H. Chun *et al.*, Nature Physics **11**, 462 (2015) **[6]** A. Banerjee *et al.*, Nat. Mater. **15**, 733 (2016); ibid. Science **356**, 1055 (2017); ibid. npj Quantum Mater. **3**, 8 (2018) **[7]** Jungho Kim et al., Phys. Rev. X 10, 021034 (2020).