Experimental report

15/02/2021

Proposal: **4-05-791** **Council:** 4/2020

Title: Anisotropy gap in the magnon dispersion of Na2IrO3

Research area: Physics

This proposal is a new proposal

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Experimental team: Alexandre BERTIN

Local contacts: Paul STEFFENS

Samples: Na2IrO3

Instrument	Requested days	Allocated days	From	To
THALES	5	5	23/09/2020	28/09/2020

Abstract:

We recently succeeded in performing INS experiments on the magnetic excitations on Na2IrO3, which in spite of its AFM ground state is considered to be a candidate for the Kitaev model on a honeycomb lattice. The experiments reveal the low energy dispersion in one direction but show now magnon gap above 2meV. In order to determine this gap, higher resolution data is required that will also allow us to determine the small dispersion perpendicular to the layers.

Experimental Report

Instrument	Thales
Proposal Number	4-05-791
Proposal	Anisotropy gap in the magnon dispersion of Na_2IrO_3
Experimentalist	Alexandre Bertin, Markus Braden
Local Contact	Paul Steffens

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, i.e. along each of the three bond directions only a single (and different) orthogonal spin component is coupled, the so-called Kitaev interaction *K.* 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. Spins fractionalize into Majorana fermions with interesting properties in the context of 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 below T_c=15K which points to additional magnetic interaction parameters beyond the pure Kitaev interaction. The magnetic order is found to be of the zigzag type [3], which can be stabilized by different additional terms. Already the nearest neighbor interaction needs to be extended to an isotropic Heisenberg interaction, *J*, and a symmetric anisotropy term Γ. In addition there are further couplings (supposedly isotropic) for pairs at larger distances. However, there is little information about the magnetic interaction parameters in these honeycomb iridates. Only a powder INS study is reported [3], which remains qualitative. With the purpose to get information about the magnetic exchange parameters, we previously conducted experiments on IN8 [4]: magnon modes have been resolved by constant energy scans in *K* direction across the AFM Bragg peak (*0KL*) with half-integer *L* in the monoclinic *C2/m* notation. An intriguing feature was the absence of a large gap in such an anisotropic system with strong spin-orbit coupling. Magnon modes could be resolved at 2meV at the AFM zone center, and its energy dependence suggests the gap to be not much smaller than that. However, with the thermal TAS it is not possible to study the energy range below 2meV.

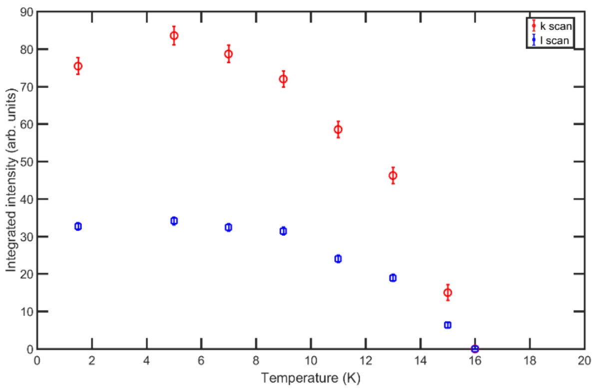

Fig. 1: Integrated intensity of the magnetic Bragg peak (0 1 0.5) scanned across the *K* (red circle) and *L* direction (blue square).

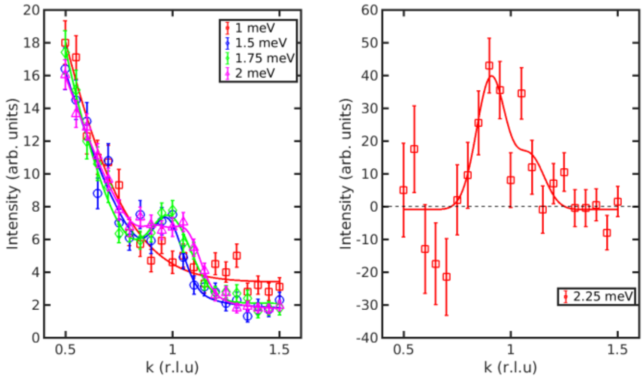

Fig. 2: Constant energy scans across the magnetic zone center (0 K 0.5) for several energy transfers at *T*=1.5K. In the right panel, background measured by rotating the sample by 45° has been subtracted to the raw data.

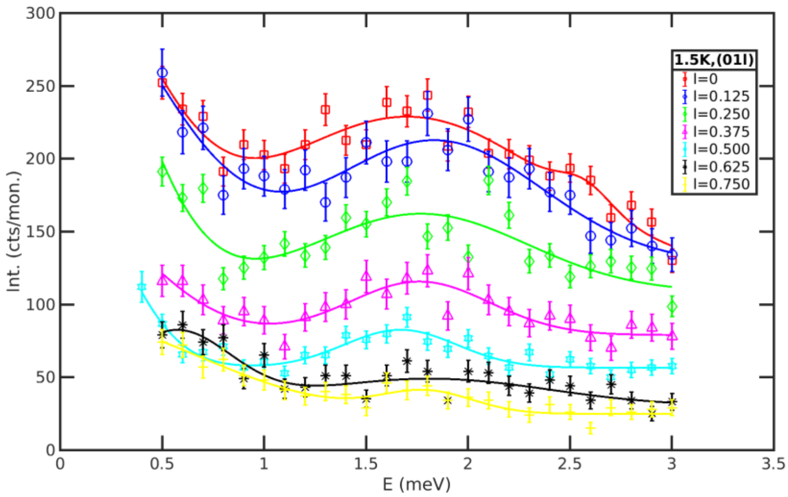

Fig. 3: Energy scans across the magnetic zone center (0 1 *L*) for several *L* values at *T*=1.5K. In the left panel, background has been subtracted to the raw data.

The aim of this experiment was to resolve the energy gap of the magnon excitations by using cold neutrons, and to track its *L* dependency which will yield valuable information on the inter-plane couplings. We used the same Al sample plate as used on the previous experiments on IN8, and consisting of 63 H-free glued thin crystals (grown in the group of Prof. G. Cao at Boulder University, USA) for a total sample mass of 207.8mg. Crystal thicknesses are mainly below 1mm, limiting neutron absorption. Fig. 1 shows the temperature dependence of the integrated intensity of the magnetic Bragg peak at (0 1 0.5), in the monoclinic *C2/m* notation, scanned across the *K* and *L* directions, and showing the emergence of 3D antiferromagnetic order at T_c=15K, in agreement with previous studies [3]. Using cold neutrons allowed us to avoid the large background arising at small *Q* on IN8, and to resolve the magnon mode at the AFM Bragg peak with *L*=0.5. Constant energy scans along *K* across the magnetic zone center and measured at *T*=1.5K are shown in Fig. 2. The magnon signal clearly emerges at *ΔE*=1.5meV. At *ΔE*=2.25meV, the signal becomes weaker. The data at 1meV clearly does not exhibit the magnon peak and thus unambiguously proves the finite spin gap in Na_2IrO_3. Background is a delicate issue in the quantitative analysis and we measured it by rotating the sample by ±45°. This background can be subtracted from the raw data in order to resolve the magnon peak, see right panel of Fig.2. At higher energy transfer, the signal could not be observed within reasonable time. The low energy part of the magnon dispersion has been reconstructed and is shown in the left panel of Fig.4. The *L* dependence of the magnon gap has been investigated by energy scans measured at *T*=1.5K that are reported in Fig.3 for several *L* values. It turned out that determining the background by rotating the sample was not reliable enough for the quantitative analysis, instead we obtain the magnon energies by fitting the raw data with Gaussians. We can follow the magnon at (0 1 *L*) for 0 < *L* < 0.75, the Ir form factor and the less favorable geometry suppresses the signal rapidly with increasing *L*. There is no finite *L* dispersion detectable documenting the two-dimensional character of magnetism in Na_2IrO_3. Finally, the temperature dependence of the low-energy magnon mode has been investigated. Energy scans at the magnetic zone center (0 1 0.5) were measured at base temperature, at 13 K and above the transition temperature at T=18K and 40K. The results with the background subtracted from the raw data are shown in Fig.5. The finite-energy magnon mode at the zone-center vanishes at T_N, as it is expected for such an anisotropy mode. In contrast the magnon signals measured at larger energy on IN8 remain visible up to well above T_N. Close to the transition there is clear enhancement of low-energy fluctuations. To conclude, we succeeded to identify the magnon gap at 1.7meV, a value surprisingly low for such an anisotropic system containing Ir. The low-energy part of the magnon dispersion around the AFM Bragg peak at *L*=0.5 could be determined and the absence of significant *L* dispersion is documented. This THALES experiment characterizes Na_2IrO_3 as a two dimensional magnetic system sensing only small anisotropy.

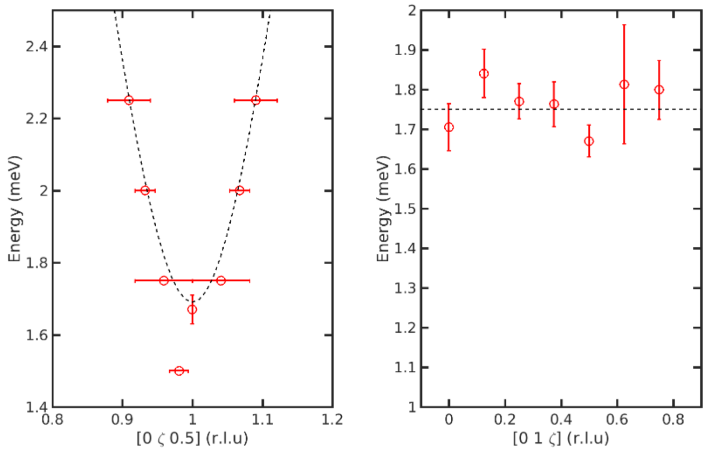

Fig. 4: Left: Low energy dispersion of the AFM spin-waves in Na_2IrO_3 across the magnetic zone center. The dashed line is a guide to the eye, modelled with $E=(\Delta^2+(c*(K-K_0))^2)^{1/2}$, with Δ =1.69meV and c=16.5meV.Å^{-1}. Right: L-dependence of the magnon energy gap. The constant-E dashed line at ΔE=1.75meV is a guide to the eye.

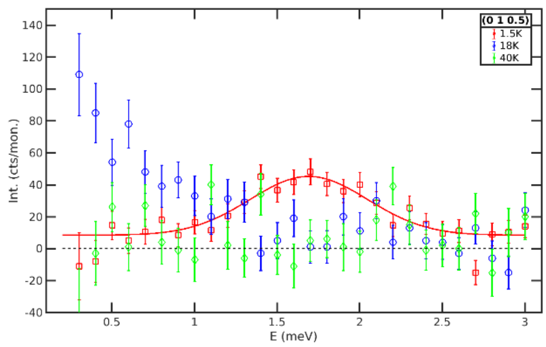

Fig. 5: Energy scans at the magnetic zone center (0 1 0.5) for temperatures below and above the AFM transition temperature at T_c=15K. Background has been subtracted.

References

[1] Gang Cao and P. Schlottmann, 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] Experimental reports 81606 and 84735