Experimental report

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Proposal: **4-05-759**

Council: 4/2019

Title: Magnetic excitations in Na_2IrO_3

Research area: Physics

This proposal is a new proposal

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Local contacts: Alexandre IVANOV

Samples: Na_2IrO_3

Instrument	Requested days	Allocated days	From	To
IN8	7	7	10/02/2020	17/02/2020
IN3	1	1	31/01/2020	01/02/2020

Abstract:

The pure Kitaev model on a honeycomb structure attracts enormous interest because the excitations in this exactly solvable model with a spin-liquid ground state behave like Majorana Fermions. Iridates, in particular Na_2IrO_3, can be considered as realizations of this model, although all these materials exhibit AFM order. Clearly other interaction beyond the Kitaev one are relevant in this material. With the progress in crystal growth for Na_2IrO_3 we propose to perform inelastic neutron scattering studies on a thermal TAS, which can yield much more insight than existing RIXS, REXS and powder INS studies.

Experimental Report

Instrument	IN8
Proposal Number	4-05-759
Proposal	Magnetic interactions in Na_2IrO_3
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, i.e. along each of the three bond directions only a single (and different) orthogonal spin component is coupled. This coupling is labelled 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.

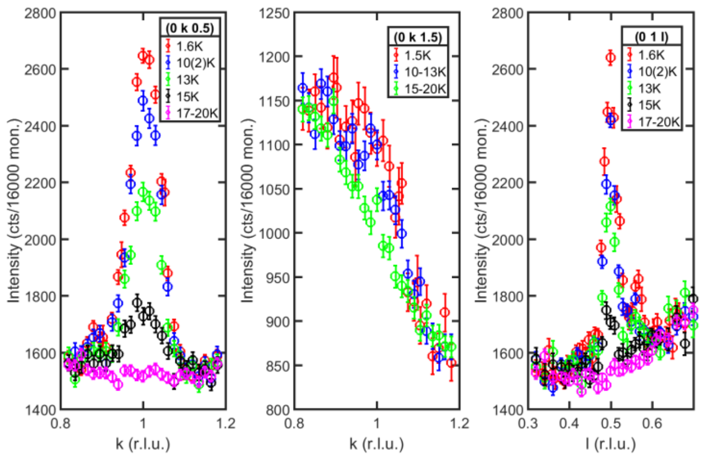

Fig. 1: Elastic scans across the magnetic zone center (0 1 0.5) k-direction (left), (0 1 1.5) k-direction (middle), and (0 1 0.5) l-direction (right).

Na_2IrO_3 as well as the other discussed candidates, however, exhibits long-range antiferromagnetic order below ~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. Unfortunately it seems impossible to grow large crystals, Ir is highly absorbing and the form factor falls off very rapidly. For the iridates there is only a powder INS study reported, which remains qualitative and X-ray studies analyzing either the diffuse scattering [5] or the resonant inelastic signals [4]. The latter two experiments 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 find the clear signature of the Kitaev interaction superposed on the antiferromagnetic magnons.

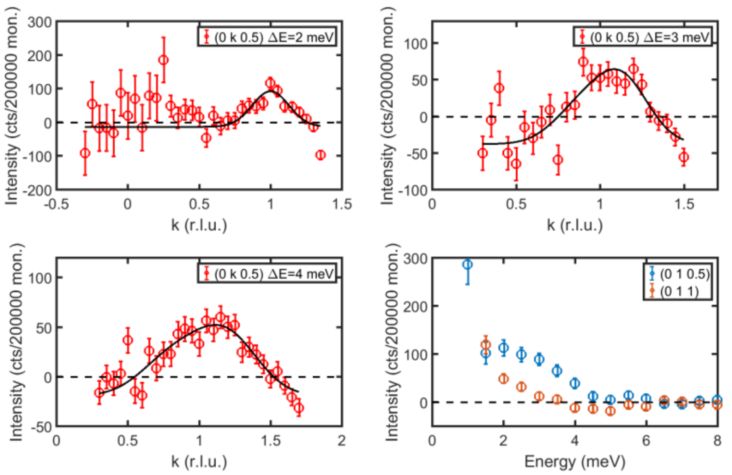

Fig. 2: Scans across the magnetic zone center (0,1,0.5) for 2, 3 and 4 meV and energy scan. The data were obtained by subtracting the background measured in scans with rotated crystal.

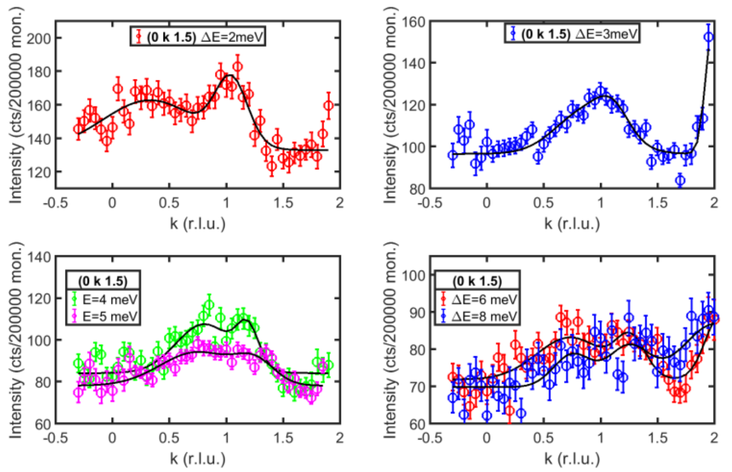

Fig. 3: Scans across the magnetic zone center (0,1,1.5) for higher energies. The raw scans show the AFM magnons but no signatures of the Kitaev interaction expected at (0,0,1.5).

It was thus the aim of this experiment to study magnetic excitations in Na_2IrO_3. We glued 63 thin crystals (from the group of G. Cao) on thin Al plates using a H-free glue (total sample mass of 207.8mg). Crystals grow as plates parallel to the layers with thicknesses mostly below 1mm, so that the absorption remains tolerable. The crystals exhibit clear edges which simplifies the orientation.

Fig. 1 shows the emergence of antiferromagnetic order in our crystal in perfect agreement with previous studies [3]. We use the monoclinic C2/m lattice and notation, in which the magnetic Bragg peaks appear at (0 1 l) with l half integer. Note that the order is fully three-dimensional in Na_2IrO_3 (right panel).

For the INS experiments problems arise from the necessity to keep the scattering vector small (due to the form factor) and a sharply increasing background. In particular for l=0.5 and scanning along the k direction the background cannot be flat at low energies. We have therefore measured the background by rotating the crystal sufficiently to run the scan outside the expected magnetic signals. The results of the differences are shown in Fig. 2. One clearly recognizes the magnon signals emerging at k=1. Further scans were performed at the (0 1 1.5) magnetic zone center, see Fig. 3, and at other l values, in particular l=1, which would be the magnetic zone boundary. However, from the constant energy scans we cannot deduce any finite l-dispersion. Just the energy scans shown in Fig. 1 seem to indicate some difference. There is also no clear evidence for a magnon gap which appears astonishing for such an anisotropic system. The energy scans in Fig. 1 may indicate that the gap is not much below 2meV.

The results obtained for Na_2IrO_3 significantly differ from those reported for α-$RuCl_3$ [6], which exhibits a dominating feature at (0,0,l) in addition to the spin-wave signals centered around the AFM q-values (01l) in our notation. The scans shown in Fig. 3 obtained at higher energies did not yield an indication for a dominant signal around (0,0,l) up to energies of 10meV. The preliminary magnon dispersion deduced from the data is shown in Fig. 4. It clearly cannot be described by a single nearest-neighbor interaction (which also contradicts the zigzag structure) but requires additional terms. Most likely there is a low-lying optical mode and a reflection of the dispersion at the antiferromagnetic zone boundary, which need to be taken into account in full calculation of folding the dispersion with the instrumental resolution.

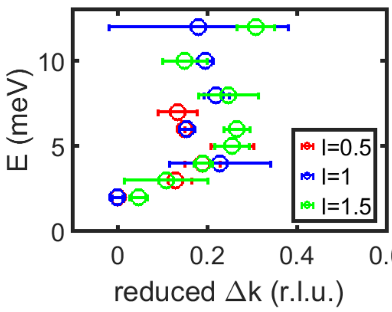

Fig. 4: Dispersion of the AFM spin-waves in Na_2IrO_3 deduced from constant-E scans.

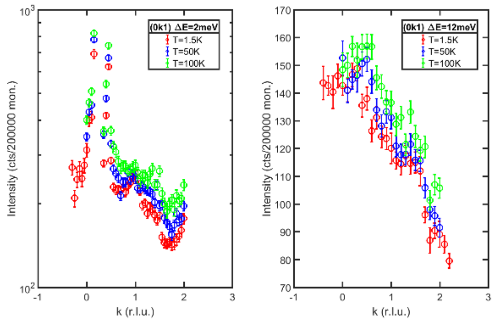

Fig. 5: (0k1) scans at l=1 and E=2 and 12 meV for different temperatures

We also performed preliminary studies as function of temperature, see Fig. 5. The sharp magnon signal at 2meV fades out with heating but survives well above T_N due to the two-dimensional character of the AFM ordering. Also the magnon signals at 12 meV remain visible up to 100K but there seems also to be enhancement of scattering around the zone centre, where Kitaev correlations can be expected.

In conclusion the first INS experiments on single-crystalline Na_2IrO_3 yield the low-energy dispersion of spin-waves in the ordered state. Up to 12 meV there is a clear difference to the INS studies on α-$RuCl_3$, which reveal a clear scattering related to the Kitaev interaction, which rapidly dominates the pattern for increasing temperature or energy.

References

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