Experimental report

30/05/2022

Proposal: **5-41-1132** **Council:** 10/2020

Title: Field-induced behavior of $(NH_4)_2IrCl_6$ with the near ideal j_eff = 1/2state of Ir^{4+}

Research area: Physics

This proposal is a new proposal

Main proposer: **Nazir KHAN**

Experimental team: Bachir OULADDIAF

Local contacts: Bachir OULADDIAF

Samples: $(ND_4)_2IrCl_6$

Instrument	Requested days	Allocated days	From	To
D10	9	7	09/06/2021	18/06/2021

Abstract:

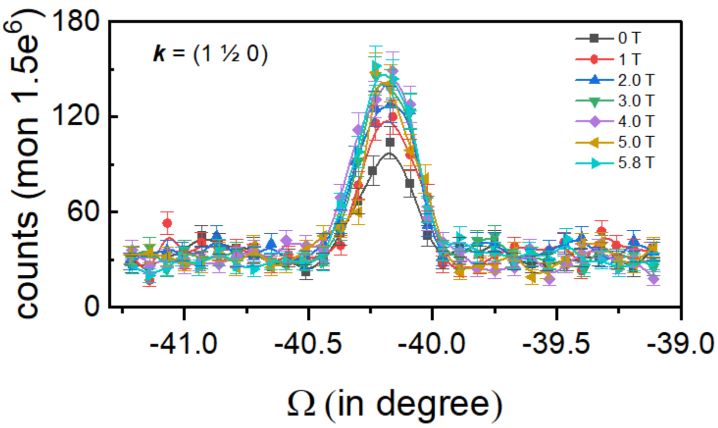
$(NH_4)_2IrCl_6$ offers the best possible cubic condition to realize the novel j_eff=1/2 state of Ir^{4+} down to a very low temperature. The interplay of the geometric frustration and the exchange frustration due to the bond dependent Kitaev interactions on the fcc lattice results in type-IIIA (k=1 0.5 0) collinear antiferromagnetic (AFM) ground state. Its ground state is one of the rich magnetic phases proposed by the fcc Heisenberg-Kitaev (J1-J2-K) model and it appears to lie close to the phase boundary. Therefore, its magnetic ground state could be tuned by external stimuli quite easily. In fact, a preliminary study on isostructural K_2IrCl_6 compound suggested that in the high-field phase the AFM state is a non-linear combination of both type-I, k = (1 0 0) and type-IIIA, k = (1 0.5 0) magnetic structures. Secondly, a strong volume effect has been observed just below the magnetic transition from thermal expansion measurement, suggesting spin-lattice coupling also playing a role in the magnetic ordering phenomena. Thus $(NH_4)_2IrCl_6$ may act as a unique case of an ideal j_eff= 1/2 system and a 5d frustrated AFM with the strong spin-lattice coupling and deserves a detailed investigation.

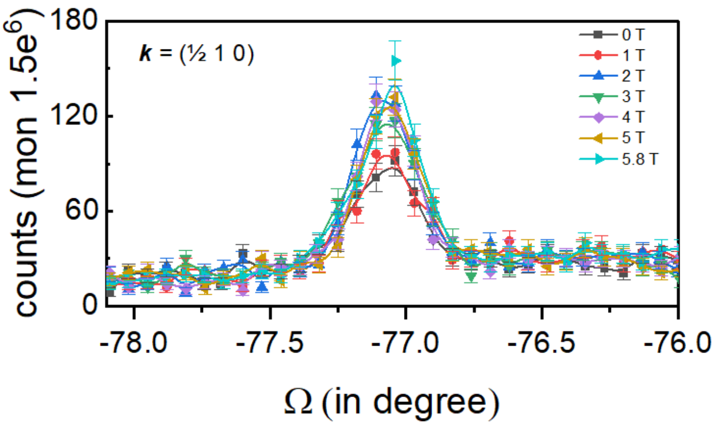
Experimental report (Proposal no. 5-41-1132, Instrument: D10)

Field-induced behavior of (NH4)2IrCl6 with the near ideal j_eff = ½ state of Ir4+

In our previous zero-field neutron diffraction experiment (proposal no. 5-41-1068) on the $(ND_4)_2IrCl_6$ single crystal, we found that the crystal structure remains cubic (Fm-3m) down to a temperature well below the antiferromagnetic transition temperature T_N=2.28 K. At 1.8 K, we were able to measure several magnetic reflections such as (1 ½ 0), (0 ½ -1), (½ 1 0), (-½ 0 1), (0 -½ -1), (3 ½ 0), and (1 -½ 2) (see the experimental report of 5-41-1068). The absence of these magnetic reflections was also checked by measuring them above the transition temperature at T=5.0 K. The evolution of the (1 ½ 0) magnetic reflection around T_N (1.84 K <T< 5 K) was investigated which suggests a very sharp magnetic transition. Two strong nuclear reflections (1 1 -1) and (0 -2 0) were also investigated around the transition temperature to check the presence of any lattice effects due to the magnetic ordering. The intensities of these nuclear reflections increase just below the magnetic transition.

In the present experiment, we have successfully measured the field dependence of the intensities of (1 ½ 0) and (½ 1 0) magnetic Bragg reflections. The magnetic field was applied along the (001) cubic axis using a vertical 6 T CNRS magnet and the horizontal scattering plane was (100) – (010). Below T_N, the crystal forms three magnetic domains due to the cell doubling along the three cubic axes, if the magnetic propagation vector ***k*** = (1 ½ 0) is single-k in nature. Since the moments in the single-***k*** structure are along the half integer component of the propagation vector, which implies the *c*-direction for the (1 0 ½) domain, this means that the magnetic field will suppress the (1 0 ½) domain and will stabilize the (½ 1 0) and (1 ½ 0) domains. So, our magnetic reflections measured in the scattering plane should increase in intensity up to 50% when the magnetic field fully suppresses the (1 0 ½) domain. This is what we see in our field dependence measurement of the (1 ½ 0) and (½ 1 0) magnetic Bragg reflections at T= 1.7 K as shown in the figure below.



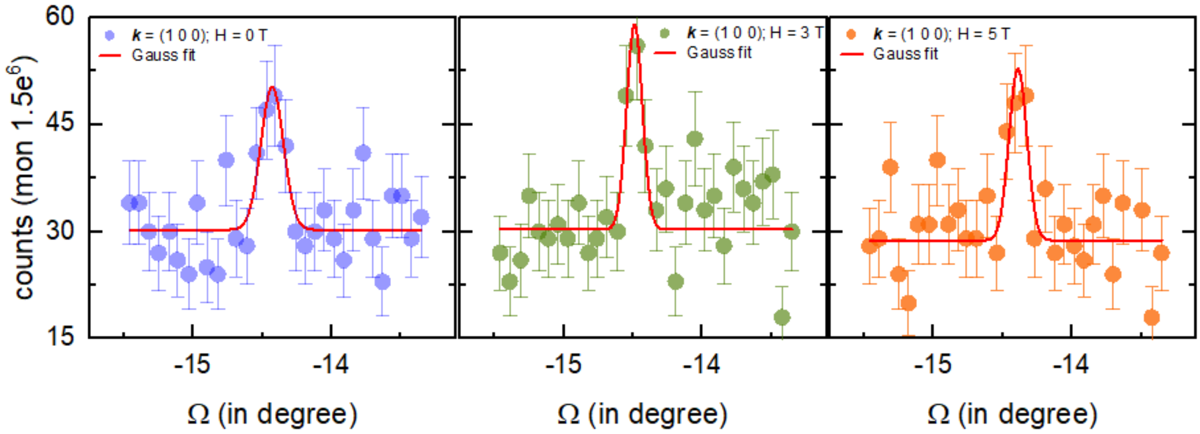


Experimental report (Proposal no. 5-41-1132, Instrument: D10)

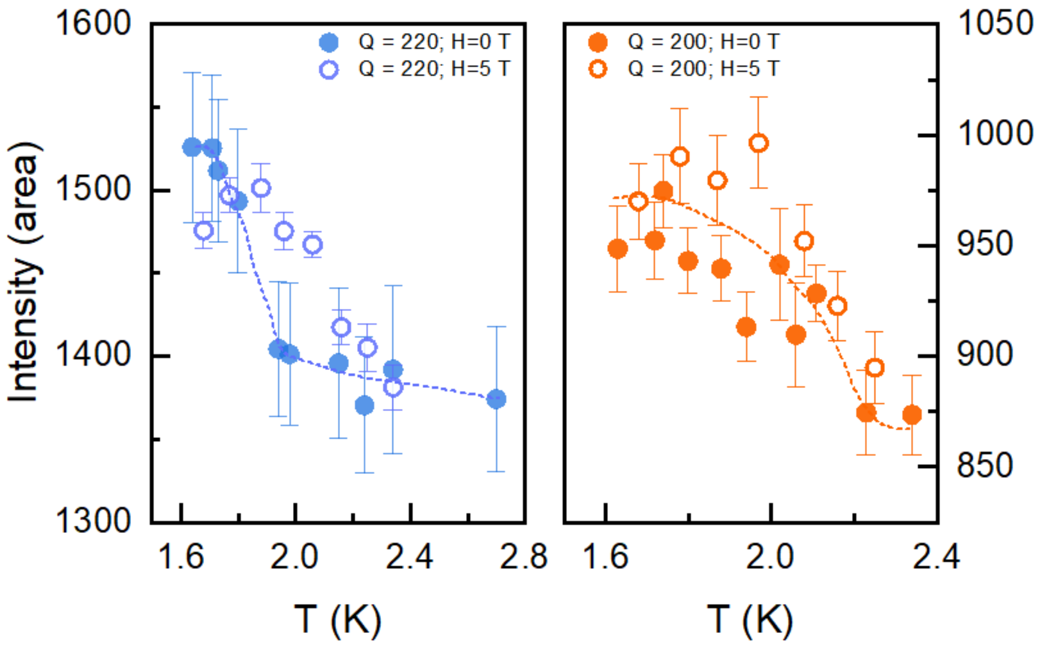
Field-induced behavior of (NH4)2IrCl6 with the near ideal j_eff = ½ state of Ir4+

The present experiment confirms that the magnetic propagation vector is a single-***k*** type with a type-IIIA collinear antiferromagnetic ground state.

It could be mentioned that in part 2 of the experiment we additionally tried to find whether the compound shows any field-induced phase transition as has been observed previously in the isomorphic K_2IrCl_6 compound where (1 0 0) magnetic reflection was detected above H = 5 T [Ref. **1**]. We found that there is a very weak (1 0 0) peak at the base temperature T= 1.7 K due to λ/2 contamination whose intensity shows no change up to 5 T as shown in the figure below. Thus, the presence of a field-induced (1 0 0) magnetic reflection in $(ND_4)_2IrCl_6$ cannot be concluded from this experiment.



Our proposal also aimed at searching for any magneto-structural effect just below the transition temperature. We measured temperature dependence of the intensity of two strong nuclear reflections (2 0 0) and (2 2 0) across the Neel temperature T_N in zero field and in an applied field of 5 T. The zero-field and 5 T data show that the integrated intensity of the two nuclear peaks increases with lowering the temperature below T_N=2.28 K as shown below.



However, whether such change in intensities indicates a symmetry lowering from Fm-3m cannot be inferred from the single crystal data. The extinction due to the formation of magnetic domains below T_N may also cause such changes in the intensity of nuclear Bragg reflections. Indeed, our powder diffraction experiment at the WISH diffractometer at ISIS, UK reveals no changes in the intensities of (2 0 0) and (2 2 0) nuclear Bragg reflections in the powder neutron diffraction patterns of $(NH_4)_2IrCl_6$ taken at T = 0.25 K and T = 3 K. Therefore, we conclude that the crystal structure remains Fm-3m in the ground state with type-III A [***k*** = (1 ½ 0)] collinear AFM ordering. No field induced type-I [***k*** = (1 0 0)] magnetic ordering is observed up to a magnetic field of 5.8 T. The field-induced phase is the conventional spin-flop phase of a collinear AFM.

Ref. 1: M. Meschke *et al.* J. Magn. Magn. Mater. **226-230**, 621 (2001).