# Experimental report

19/01/2024

**Proposal:** **4-01-1789**

**Council:** 4/2023

**Title:** Study of bond-directional excitations in the proximate Kitaev QSL ¿-RuCl3

**Research area:** Physics

**This proposal is a new proposal**


**Main proposer:** **Alexandre BERTIN**

**Experimental team:** Yixi SU
Markus BRADEN
Alexandre BERTIN

**Local contacts:** Paul STEFFENS


**Samples:** alpha-RuCl3

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| THALES | 6 | 6 | 10/11/2023 | 16/11/2023 |


## Abstract:

¿-RuCl3 is the most promising candidate to study the Kitaev model, in which Ising nearest-neighbor exchange coupling are bond-directional. This sample develops a long range magnetic order below TN=7K, characterized by a zigzag type AFM order with propagation vector [0 1 1/2] (in our orthorombic notation). The ordered moment is estimated to be ¿~0.45¿B. The main feature revealed with INS experiments is a broad excitation continuum, of magnetic origin, at the Brillouin zone center which extends up to ~12meV and survives well above TN up to ~120K. The aim of this experiment is to use longitudinal polarization analysis on the cold neutron TAS Thales in order to both investigate the anisotropy of the continuum at the Brillouin zone center and to look for the anisotropy gaps of the in-plane and out-of-plane spin waves excitations at the 2D AFM zone center.

## Experimental Report

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|---|---|
| **Instrument** | THALES |
| **Proposal Number** | 4-01-1789 |
| **Proposal** | Study of bond-directional excitations in the proximate Kitaev QSL α-$RuCl_3$ |
| **Experimentalist** | Alexandre Bertin, Yixi Su, Markus Braden |
| **Local Contact** | Paul Steffens |

The aim of the experiment was to study the anisotropy of magnetic excitations in the candidate material for a Kitaev quantum spin-liquid α-$RuCl_3$. α-$RuCl_3$ consists of honeycomb $RuCl_3$ layers and shows antiferromagnetic (AFM) order with a zickzack arrangement at low temperature [1-4]. A large α-$RuCl_3$ single-crystal of weight 0.7g was grown at MLZ in the group of Y. Su and mounted in the [010]/[001] orientation (in the monoclinic notation with space group *C2/m* [2]). In this setting magnetic Bragg peaks appear at (0 1 $Q_L$) but the crystal consists of domains that are rotated by 120 degrees against each other. Note that other space groups were proposed for α-$RuCl_3$ at low temperature [5], but this does not affect the analysis of the multidomain magnetism described here. We used focusing Heusler crystals as polarizing monochromator and analyzer, and a Helmholtz-coil setup permitted to adjust the neutron polarization direction at the sample. Flipping ratios determined on nuclear Bragg peaks amounted to above ~20. To suppress higher order contaminations, we inserted a velocity selector and a Beryllium filter with most scans performed with fixed $k_f$=1.50Å$^{-1}$.

Fig. 1 shows results of elastic scan across the magnetic Bragg peaks. There is a single magnetic transition visible at $T_N$~8.2K in perfect agreement with the recent literature on high-quality crystals with a small amount of stacking faults [5]. The polarization analysis of *e.g.,* the (0 1 $Q_L$) Bragg peak shows that magnetic moments are aligned mostly parallel to the layers, only in qualitative agreement with non-polarized studies indicating a larger *c* component [6]. The sharp SF scattering for polarization direction *x* vanishes above the Néel temperature. For the definition of the *x,y,z* coordinate system used in neutron polarization analysis see the caption of Fig. 1. Throughout the report, we use the red-green-blue color sequence for data along coordinates *x,y,z.*

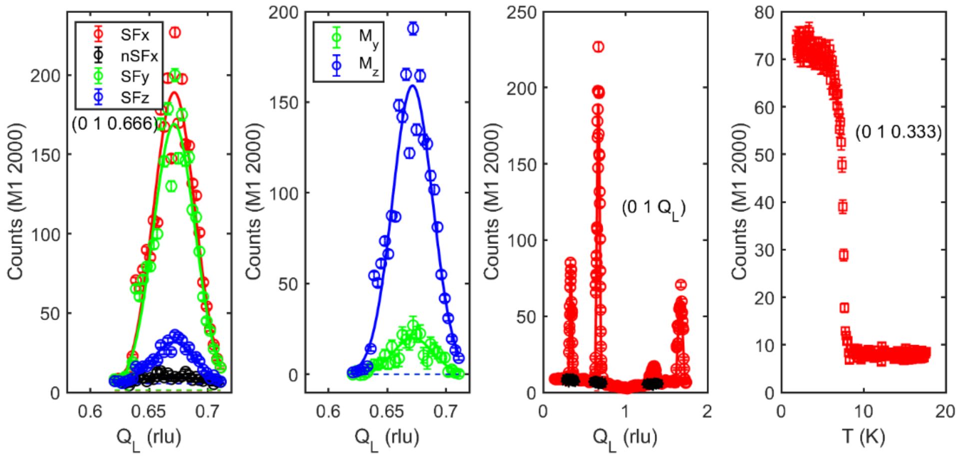


**Fig. 1:** Elastic scans in the direction perpendicular to the $RuCl_3$ layers across the positions of AFM Bragg scattering. From the spin-flip (SF) data taken along three directions we calculate the contributions of the vertical component parallel to the layers, corresponding to coordinate *z,* and of the direction *y* parallel to the scattering plane and perpendicular to the scattering vector. In this commonly used notation the coordinate *x* is parallel to the scattering vector.

The most important inelastic result is shown in Fig. 2 presenting energy scans at the magnetic Bragg position (0 1 0.333). In agreement with unpolarized experiments, there is a sharp mode at E=2meV. However, the polarization analysis shows that this signal is equally strong in the SFx and SFy channels documenting that this mode is almost completely polarized vertical to the scattering plane, coordinate *z*, that is parallel to the layers and perpendicular to the in-plane component of the scattering vector, (010). The inelastic response is thus parallel to the static one, which is just opposite to the expectation for a simple AFM system. In a usual AFM system, low-energy magnons near the AFM zone center correspond to a transversal precession around the direction of the ordered moment and are thus polarized perpendicular to the static moment, they acquire finite energy through moderate anisotropy [7]. This discrepancy illustrates that AFM ordering in α-$RuCl_3$ fundamentally differs from a Heisenberg model. Our finding can, however, be perfectly described by a dominant bond-directional anisotropy [8,9], which pushes the transversal precession mode to very high energies so that there is no longer a dispersion minimum near the AFM zone center. The strong signals in Fig. 2 and 3 correspond to contributions of the rotated domains that do not contribute to the static signal at the scattering vector chosen in our experiment. Also, these non-condensed instabilities are thus fully anisotropic. In α-$RuCl_3$ each propagation vector of magnetic fluctuations is tight to a single spin component.

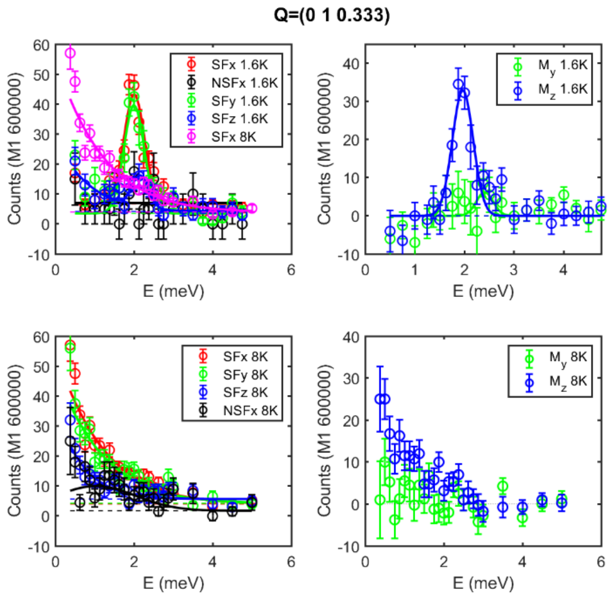


**Fig. 2:** Energy scans at the magnetic zone center (0 1 0.333) at 1.6 and 8 K in the upper and lower panels, respectively. From the SF data in the left panels, we calculate contributions of the vertical and of the *y* components in the right panels. The inelastic mode at 2 meV appears in the same polarization direction as the static scattering.

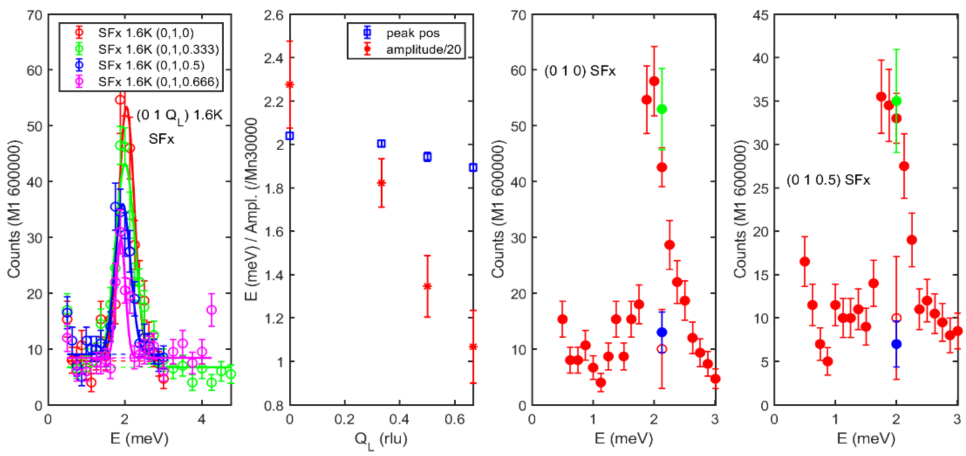


**Fig. 3:** Energy scans at the magnetic zone center (0 1 $Q_L$). From the SFx data we deduce weak energy and pronounced amplitude dispersion. Also for $Q_L$ =0 and 0.5, the signal fully stems from the SFy polarization channel.

The measurements at other AFM zone centers (0 1 $Q_L$) fully agree with this interpretation. Fig. 3 shows that there is little energy dispersion for varying $Q_L$ but that the signal is rapidly suppressed. Also, at $Q_L$

values not corresponding to the 3D order the signal is only found in the vertical component underlining the bond-directional anisotropy.

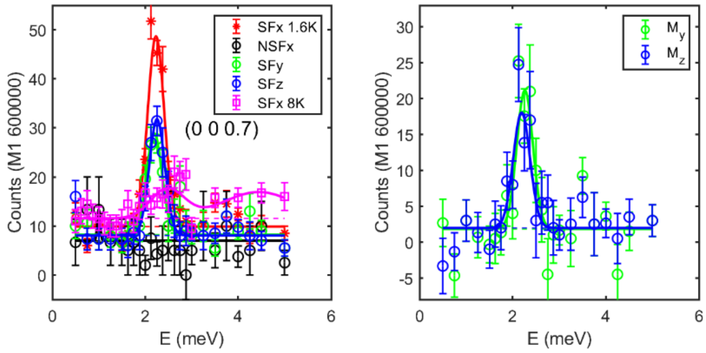


**Fig. 4** Polarization analysis at ***Q***=(0 0 0.7) corresponding to the ferromagnetic zone center. There is a sharp mode that appears equally strong in SFy and SFz channels yielding comparable contributions from both components.

Fig. 4 shows the results of polarized scans at the ferromagnetic ***Q*** position (0 0 0.7). One finds again a rather sharp mode at 2.2meV that, however, appears equally strong in SFy and SFz channels revealing that this signal is fully isotropic within the layers. Such behavior must result from the rotational twinning. At the ferromagnetic in-plane component one expects the impact of the Kitaev interaction, here the scan at 8 K shows the broad response emerging from the suppression of the AFM ordering, which was reported in unpolarized experiments.

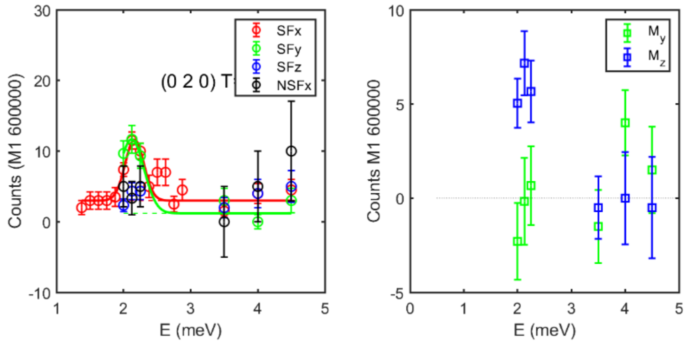


**Fig. 5** Polarization analysis at ***Q***=(0 2 0) corresponding to the ferromagnetic zone center. One observes a sharp signal in agreement with the data in Fig. 4, but here the finite in-plane component allows one to study the anisotropy.

Studying the ferromagnetic response at (0 0 $Q_L$) suppresses any out-of-plane magnetic contribution and does thus not allow one to investigate the anisotropy of these signals that are attributed to the Kitaev interaction. Fur this purpose we examined at ***Q***=(0 2 0), see Fig. 5. The signal is strongly reduced due to the magnetic form factor but a sharp feature is clearly visible at the same energy. This signal is entirely contained in the SFy channel indicating a full polarization parallel to the layers. This is rather intriguing as the response of dominant Kitaev interactions along the three bond types should cover all directions equally.

In conclusion, due to the high-quality crystal of large size polarized inelastic experiments on Thales were quite efficient yielding good statistics. The polarization of magnetic excitations in the zickzack AFM ordered state is governed by anisotropy and is fully opposite to the expectations for a Heisenberg model. The strong coupling between the propagation vector and the polarization of the fluctuations persists in the paramagnetic state. Importantly we also find a strong anisotropy of the quasiferromagnetic response.


**References** **[1]** K. W. Plumb et al., Phys. Rev. B **90**, 041112(R) (2014). **[2]** R.D. Johnson et al., Phys. Rev. B **92**, 235119 (2015). **[3]** H.B. Cao et al., Phys. Rev. B **93**, 134423 (2016). **[4]** G. Cao et al., Rep. Prog. Phys. **81**, 042502 (2018). **[5]** Subin Kim et al., arXiv2311.04000. **[6]** J.A. Sears *et al.*, Phys. Rev. B **91**, 144420 (2015). **[7]** N. Qureshi et al., Phys. Rev. B **86**, 060410(R) (2012). **[8]** S. M. Winter et al., *Nat Commun* **8**, 1152 (2017). **[9]** H. Liu et al., Phys. Rev. B **105**, 214411 (2022).