Experimental report

12/09/2018

Proposal:	**4-01-1497**	**Council:**	4/2016
Title:	Higgs amplitude mode in Ca_2RuO_4		
Research area:	Physics		

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Samples: Ca_2RuO_4

Instrument	Requested days	Allocated days	From	To
IN20	6	8	08/07/2016	16/07/2016

Abstract:

The phenomenon of Bose-Einstein condensation (BEC) takes many different guises in nature ranging from superfluidity in liquid helium and ultracold atomic gases to superconductivity in metals. An analogy is also found in magnetic insulators, in which a quantum phase transition takes place as magnons condensate. A well-known example is the BEC of triplons in a dimer system $TlCuCl_3$ induced by tuning the boson density using magnetic field, providing a connection between complex quantum properties of matter and BEC.
Recently, a novel type of BEC of spin-orbit excitons has been proposed in Ca_2RuO_4. Using TOF and polarized neutron scattering measurements, we have found strong indications for a magnetic mode compatible with the theoretical predictions. In order to complete our study, it is crucial to follow the dispersion of the additional mode, and by doing polarization analysis on a detwinned sample confirm its longitudinal nature. Additionally, following the broadening of the mode as the wavevector approaches \$(\pi,\pi)\$ will shed light on the decay process of the Higgs mode.

Higgs amplitude mode in Ca_2RuO_4

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We report on our polarized inelastic neutron scattering measurement of the magnetic excitations in Ca_2RuO_4 following our previous results on the same system, where two in-plane modulated magnetic modes were detected: transverse and longitudinal magnon modes. In this experiment, we attempted to follow the dispersion of the longitudinal mode to study its coupling and decay as the momentum is increased towards the antiferromagnetic zone center (π, π). Unfortunately, an increased background compared to our previous experiment on the same sample using the same instrument made the measurement challenging.

OUR RECENT RESULTS

We have recently investigated the magnetic excitations in Ca_2RuO_4 by means of time-of-flight (TOF) and polarized inelastic neutron scattering (INS)[1]. The mapping of the transverse magnetic excitation using TOF revealed a spin-wave dispersion deviating from both Heisenberg or X-Y antiferromagnets. The dispersion is well accounted within a model calculation for an S=1 system[1], which predicts three modes: the main transverse mode T seen in the TOF data, a folded mode T' and the Higgs amplitude mode L, which are not clearly visible. The polarized neutron scattering measurements close to (0,0) reveal three distinct well defined peaks: out-of-plane polarized folded mode T' (Fig.1a), in-plane polarized transverse T and longitudinal L (Fig.1b). Measurements close to (π, π) (Fig.2c), reveals broad magnetic intensity in addition to the sharp transverse mode. This can be understood as the decay of the longitudinal mode due to the interaction with the two-magnon continuum[1].

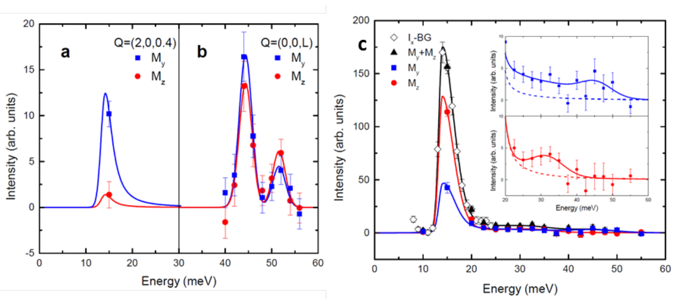

FIG. 1: **Energy scans at (a) Q = (2,0,0.4), (b) Q = (0,0,L) and (c) Q = (1,0,L).** The value of L was varied along the scan to minimize the magnitude of **Q**. The data denoted I_x-BG (empty black diamonds) is obtained from the raw data in the X channel after subtraction of a small background. The intensities M_y+M_z(filled black triangles), M_y (blue squares), and M_z(red circles) are obtained from the standard XYZ differences and the lines are guides to the eye. The inset of **(c)** shows in detail the region above 20 meV.

EXPERIMENTAL SETUP AND PROCEDURE

We measured the magnetic excitations in Ca_2RuO_4 using the thermal neutron three-axis spectrometer IN20 with polarized neutrons at the ILL. Both Heusler monochromator and analyzer were used and the polarization direction was chosen via Helmholtz coils and the final wavevector $k_f = 2.662$ Å^{-1} was chosen to match the effective window for PG filters. The same sample mosaic was used as in the previous study, consisting of ∼100 "twinned" crystals, i.e., approximately half of them are rotated 90° about the c axis with respect to the other half, therefore we can only distinguish between the in-plane (ab) and out-of-plane (c) polarized modes. The sample was mounted with the ($H0L$) plane horizontally, to perform energy scans at constant H={0,0.25,0.5,1} from 30 to 56 meV. We knew from our previous experiment that performing XYZ polarization analysis was imperative for substracting non-magnetic background and distinguishing different modes, which implies long counting times per point to disentangle the signal from the background. These measurements were taken during the first three days.

Figure 2 shows the data taken with spin-flip X polarization, which is sensitive to magnetic scattering, compared to the calculated background from polarization analysis. In comparison to our previous experiment the background level has increased tremendously making it difficult to disentangle small signals like the longitudinal mode or the transverse mode away from (1,0). Moreover the background seems to have an energy dependence in the interesting region where the signal is expected. The experiments were done as close to the same conditions as possible. One possibility would be some sample degradation, but from the intensity of the transverse magnon at (1,0) this does not seem to be the case.

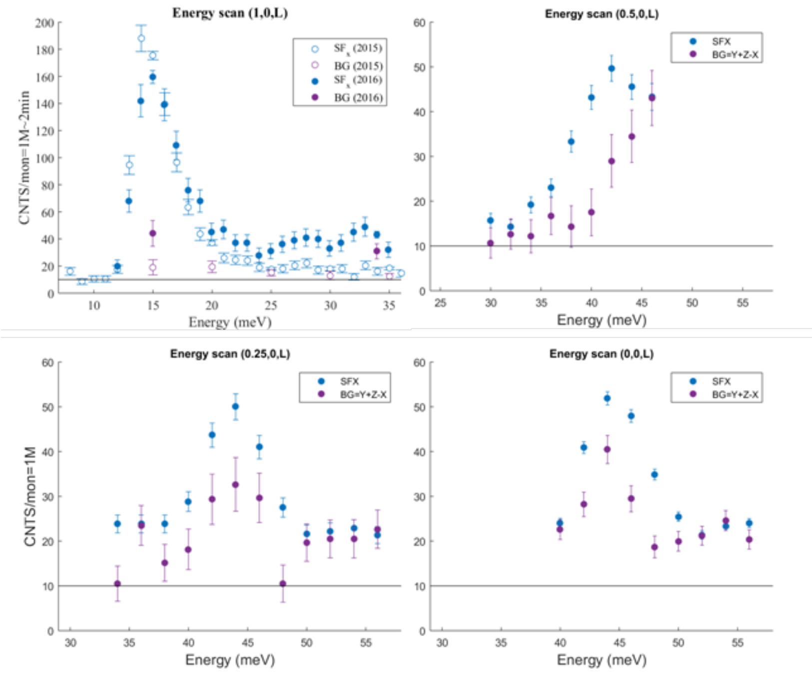

FIG. 2: Energy scans at constant $H = 1$(top-left), $H = 0.5$(top-right),$H = 0.25$(bottom-left) and $H = 0$(bottom-right). The solid blue and purple markers reperesent data taken with spin-flip X polarization and calculated background (spin flip Y+Z-X) respectively. Empty markers correspond to the data taken in the previous experiment for comparison.The solid line is an estimate of the background level in previous experiments.

In order to improve the background, we explored two other possible changes: the size of the virtual source slits and increasing the thickness of the PG filter after the sample. In the first case, the looser beam profile before the monochromator can cause more of the sample holder and sample environment to be in the beam which increases the extrinsic background. For this check we reduce the size of the slits from $30 \times 30\text{mm}^2$ to $10 \times 10\text{mm}^2$. Although the cross section is reduced by 90% the flux at the sample is estimated to be reduced by 40% as the density of neutrons increases towards the center of the beam. This approach was not very effective, as the required time to measure with the same statistics is increased substantially and the signal to background ratio was only minimally affected. The second approach was to replace the PG filter by a thicker one, the estimated transmission of the 2nd filter compared to the first was of 75% for neutrons with $k_f = 2.662$ as calculated from the reduction in the transverse magnon signal. Although the background for the mode around 15 meV was not improved, at higher energies, which was the main focus of the experiment, it was greatly reduced as seen in Fig. 3. Specifically, the background level reached similar values to what has been achieved in previous experiments. More importantly the energy dependence of the background was greatly reduced, making it more reliable for the analysis of the data.

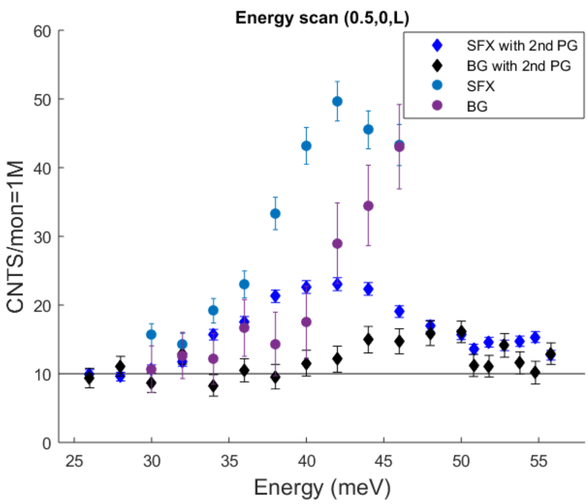

FIG. 3: Comparison of the energy scan at constant $H = 0.5$ before (circles) and after (diamonds) replacing the PG filter.

The different checks and optimizations took about a day and a half leaving us with about 3 days left of beamtime and given the reduced intensity, we decided to focus most of the remaining time, on the polarization analysis of the energy scan at $H = 0.5$.

Figure 4 shows the results of the polarization analysis for the total magnetic intensity and the two different channels My and Mz. The results of gaussian fits are shown as solid lines, where the position and width of the high energy mode were fixed to the values at (0,0) from Fig. 1(b). From the orientation factor we know the

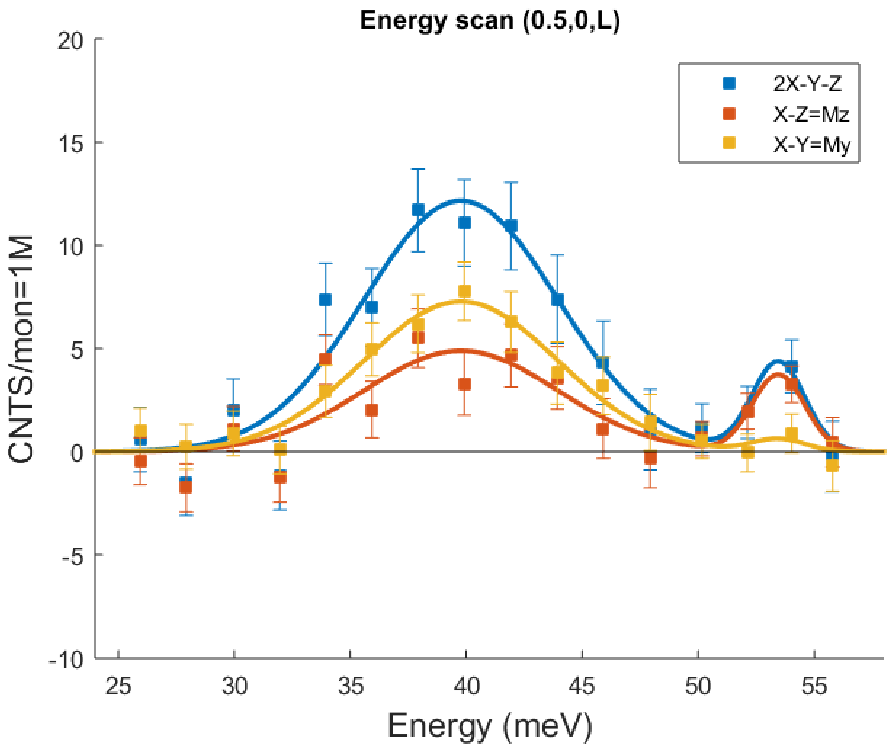

FIG. 4: Polarization analysis of the energy scans taken at $H = 0.5$. The total magnetic intensity is represented in blue and is calculated as spin-flip 2X-Y-Z, the magnetic intensity along the vertical direction Mz=X-Z is shown in orange, whereas along the direction perpendicular to both $\vec{Q}$ and $\vec{z}$, My=X-Y is shown in yellow. Solid lines are the result of gaussian fits.

measurement is not very sensitive to magnetic excitations along the c-axis and it is close to equally sensistive to excitations along a or b. In the case of equally populated twin domains, we would get equal contributions to My and Mz for an excitation that is only along a or along b, as $\vec{Q}$ is almost parallel to $\vec{c}$. The difference between the two channels is similar to what we have observed previously and is probably due to a reduced flipping ratio for the spin-flip Y channel. If we were to assign each peak to a different excitation mode, then the transverse mode would be centered at 40 meV with the longitudinal at 53 meV. Nevertheless the increased width in the first peak needs to be explained. A precise resolution calculation would be able to distinguish whether the increase is solely due to the convolution of the instrumental resolution with the dispersion (likely) or some broadening might be coming from the predicted decay of the longitudinal mode.

CONCLUSIONS AND OUTLOOK

We have measured magnetic excitations in Ca_2RuO_4 using polarized inelastic neutron scattering at intermediate momentum transfer between (0,0) and the antiferromagnetic zone center (1,0) to follow the longitudinal fluctuations. Due to time constraints and an increased background that was neccessary to remove, we were only able to measure the modes at (0.5,0). At this position, no evident decay is seen yet, but careful resolution calculations might reveal some minor effects. In retrospective we could have chosen to study the excitations closer to (1,0) where the decay is expected, for example at (0.75,0).

REFERENCES

Instruments: IN20

[1] A. Jain *et al.*, Nature Physics **13**, 633-637 (2017)

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