Experimental report

12/01/2021

Proposal:	**4-03-1738**	**Council:**	4/2019
Title:	Complex magnetic fluctuations in Ca2RuO4 above the Néel temperature		
Research area:	Physics		

This proposal is a new proposal

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Samples: Ca3Ru2O7

Instrument	Requested days	Allocated days	From	To
THALES	3	3	17/02/2020	20/02/2020

Abstract:

The 4d-electron system Ca2RuO4 (CRO) plays host to a complex interplay between magnetic and electronic correlations. Recent neutron experiments revealed a novel soft magnetism, with strong single-ion anisotropy, and 'Higgs' amplitude fluctuations in the spin wave spectrum. On the other hand, the nature of the electronic order in CRO above the magnetic ordering temperature is still under debate, and the emergence of an exotic spin-nematic or an orbitally ordered state have been suggested. Here, propose to study the magnetic fluctuations prevalent in proximity to TN that are fundamentally related to the nature of the magnetism, with the magnetic order parameter possibly coupling to the electronic states. Thus, a thorough understanding of the magnetic dynamics above the magnetic transition will help elucidate the nature of the electronic order in CRO.

Experimental Report for proposal: 4-03-1738

Title: Complex magnetic fluctuations in Ca_2RuO_4 above the Néel temperature

Date: From 17/02/2020 to 20/02/2020

Remark: Sample was changed to $Ca_3Ru_2O_7$

The layered Ca-ruthenates Ca_2RuO_4 (CRO214) and $Ca_3Ru_2O_7$ (CRO327) host various intriguing physical phases due to an interplay of crystal field splitting, spin-orbit coupling, and Hund's coupling. An avenue to obtain deep insights into complex magnetic phase transitions is the exploration of critical dynamics. Specifically, the fluctuations prevalent in proximity to T_N are fundamentally related to the nature of the magnetic order. Therefore, we aim to determine the critical exponents of spin fluctuations (ν, z) in CRO214 and CRO327 using high-resolution neutron spectroscopy to check whether the 2D to 3D-like crossover in the crystal structure [Fig. 1a] and electronic correlations are similarly reflected in critical dynamics or if other effects govern the magnetism.

The following measurements were carried out on the high-resolution triple-axis spectrometer THALES to determine the temperature dependence of the energy and momentum linewidth Γ and κ, respectively, and eventually derive the critical exponents via the scaling relation $\Gamma \sim \kappa^z \sim (T/T_N - 1)^{\nu z}$.

Note that, in the scope of this experiment, we studied the critical dynamics of the bilayer compound CRO327. To realize a reasonable scattering intensity, we arranged about 30 co-aligned single crystals with a total mass of ~800 mg on a Si-plate [Fig. 1b].

The sample alignment was done in the ordered antiferromagnetic (AFM) phase at T = 10K with a neutron wave vector of k = 1.5 1/Angst. . The instrumental configuration was (+-+) and the scattering plane was defined as (100)/(001). For the actual alignment, we used the nuclear peaks (200) and (002), respectively. By performing Q_H-scans around (200) we were able to obtain the ab-twinning (1:1) of the crystals [Fig. 1c]. Furthermore, the sample mosaicity of ~2° was determined through a rocking scan at (002).

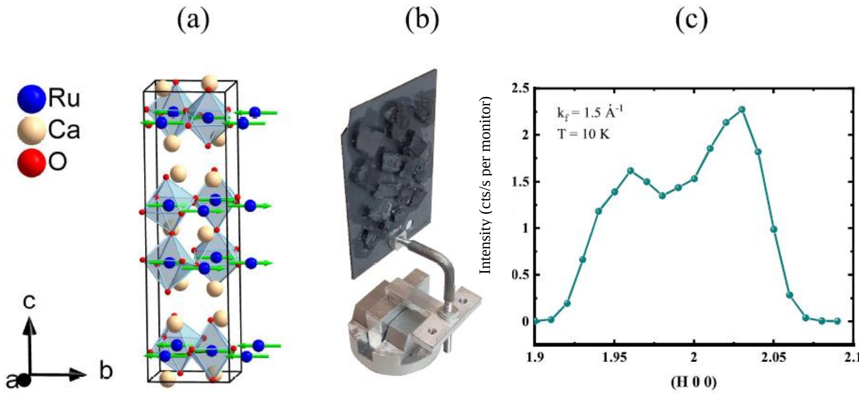

Fig. 1: (a) Crystal and magnetic structure of CRO327. The magnetic moments (green arrows) are FM aligned along the crystallographic b-axis within the bilayers and AFM coupled along the c-direction. (b) Photograph of the CRO327 sample. (c) QH-scan around (200) to determine the ab-twinning of the crystals.

After the alignment, we changed the neutron wave vector to 1.3 1/Angst. and tracked the (001) magnetic peak intensity to determine the magnetic transition temperature $T_N \sim 54K$ and the metal-to-insulator transition $T_{MIT} \sim 48K$ for the present thermometry on THALES [Fig. 2]. A preliminary power-law fit on the resulting intensities leads to a 2D-like critical exponent $\beta \sim 0.15$ of the sublattice magnetization.

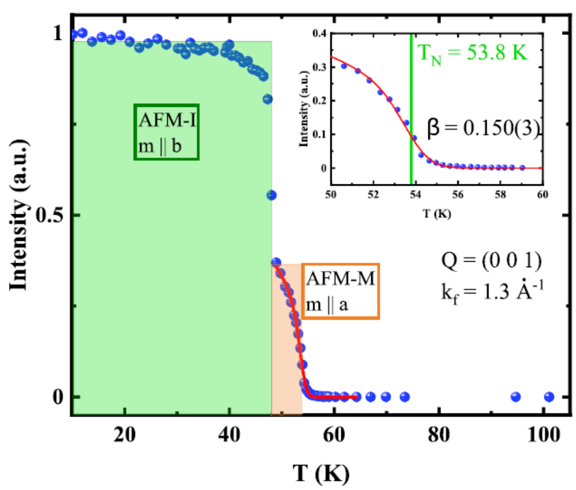

Fig. 2: T-dependence of the AFM (001) peak intensity. The step of intensity at $T_{MIT} \sim 48K$ indicates the first order metal-to-insulator transition.

Finally, we performed various scans in energy and momentum space at different temperatures [Fig. 3], determined the corresponding widths Γ(T) and κ(T), and thus, were able to derive preliminary values for the critical exponents [Fig. 4]. While the static critical exponent *ν ~ 0.5* unexpectedly indicates the mean-field value, the dynamic critical exponent of *z ~ 3* does not match any universal value.

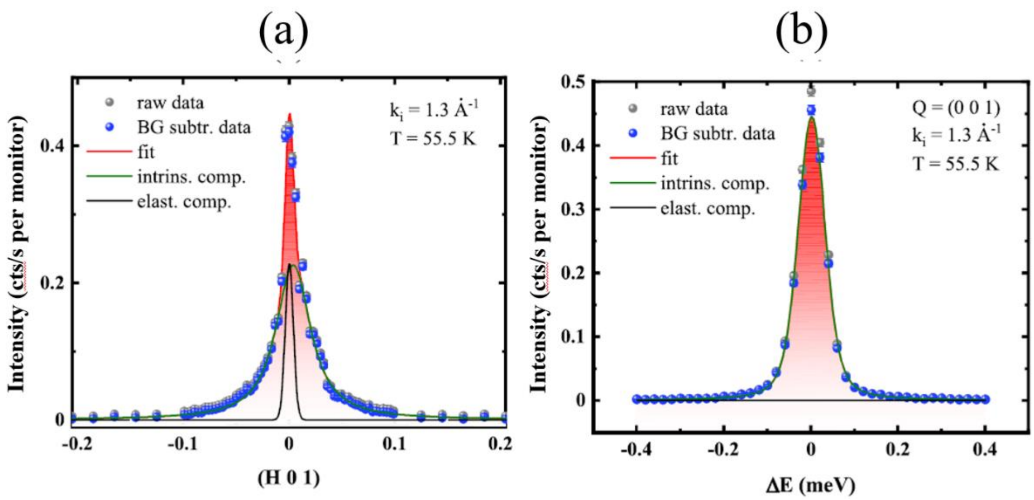

Fig. 4: Examples of (a) an energy integrated Q_H-scan and (b) an E-scan at T = T_N +2K. Note that, we observed a resolution limited signal with constant linewidth (black solid line) in the Q_H-scans besides a signal with temperature dependent Γ(T), which we assigned to the critical fluctuations (green solid line).

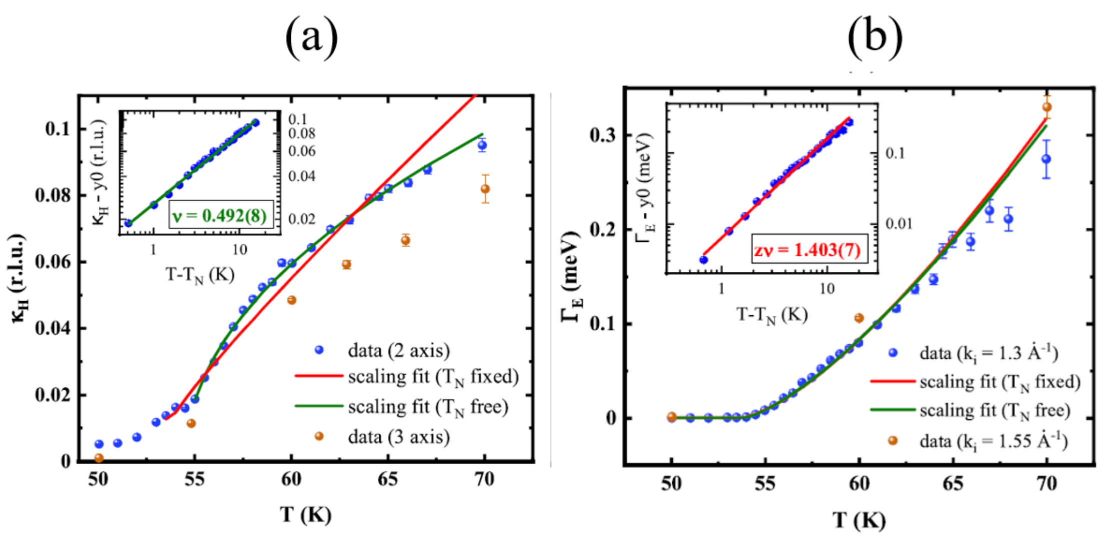

Fig. 3: (a) T-dependence of the Q-linewidth κ_H. For comparison, data taken in energy-integrated (Two axis) and triple-axis mode are shown. (b) T-dependence of the energy linewidth Γ. The inset each show a double logarithmic plot.