Experimental Report

Proposal: **5-51-496** **Council:** 4/2014

Title: Spin density distribution on Ru inCa2RuO4: A polarised neutron diffraction study

This proposal is a new proposal

Researh Area: Physics

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Samples: Ca2RuO4

Instrument	Req. Days	All. Days	From	To
D3	7	7	29/09/2014	06/10/2014
D9	5	5	24/09/2014	29/09/2014

Abstract:

The recent finding [12] that spin-orbit coupling can have a major effect on the electronic structures calls for a detailed investigation of the role of SOC in 4d oxides. In the proposed work, we plan to investigate the magnetization distribution in Ca2RuO4 single crystals by polarized neutron diffraction measurements. We would like to compare the experimentally observed spin density maps with simulated ones for the S = 1 case (crystal field dominates) and J = 0 case (SOC dominates) to understand the effect of SOC on the ground state electronic configuration. The outcome of the proposed experiment will help in determining the radial part of the Ru magnetic form factor and possibly to establish the symmetry of Ru 4d orbitals in Ca2RuO4.

Spin density distribution on Ru in Ca_2RuO_4: A polarised neutron diffraction study

(Experiment No: 5-51-496, Instrument: D3 and D9)

Ruthenates have been at the forefront of the condensed matter physics following the discovery of unconventional electronic ordering phenomena [1–6] including spin-triplet superconductivity in Sr_2RuO_4 and electronic nematicity in $Sr_3Ru_2O_7$. Other ruthenates including $(Tl,Hg)_2Ru_2O_7$, $La_4Ru_2O_{10}$, and $PbRuO_3$ exhibit prominent metal-insulator transitions as well as unusual orbitally and magnetically ordered states whose microscopic origin has not yet been explored in detail. All of these phenomena result from a delicate interplay among spin, orbital, and lattice degrees of freedom, which are coupled through the crystal field and cooperative Jahn-Teller effect, Hund's coupling, magnetic exchange coupling, and to the some extent, spin-orbit coupling (SOC).

Despite clear experimental indications of strong SOC in a number of spectroscopic studies [7, 8], many theoretical models have neglected it [9–11]. However, the recent finding that SOC can have a major effect on the electronic structures of related 5*d* transition-metal oxides [12] calls for a detailed investigation of the role of SOC in 4*d* oxides. To this end, we particularly focus on a Mott insulator Ca_2RuO_4 with Van-Vleck type transition metal ion, having t_{2g}^4 electronic configuration. It crystallizes in a layered perovskite structure (space group $Pbca$) containing RuO_6 octahedra that are tilted through the *ab*-planes and rotated about the *c*-axis. It undergoes metal-insulator transition temperature at 357 K[13] and orders in the A-type AFM structure (with moment pointing along the *b*-axis) below 110 K[1]. A strong spin-orbit coupling, confirmed by x-ray absorption study, can drastically change the electronic ground state of this compound. In the limit of zero SOC, low-spin t_{2g}^4 electrons with nominally $S = 1$ moments (triplet) will interact with each other through the usual Goodenough-Kanamori type superexchange interactions. However, in the limit where SOC is much larger than the tetragonal crystal field splitting, coupling of spins to $L = 1$ orbital moments will result in the $J = 0$ nonmagnetic ground state (singlet). Since SOC (which give rise to the singlet-triplet splitting) in this compound is comparable to superexchange energy, a Bose condensation of the exciton into a magnetic state has been theoretically predicted by G. Khaliullin [15]. Formally, this singlet-triplet structure is very similar to that in the dimer system, which hints at a possible BEC analogous to the one discovered in $TlCuCl_3$ [14]. Note, however, that the singlet-triplet structure is realized within a single ion by the orbital angular momentum L ($L = 1$) that is equal in magnitude and opposite in direction with respect to the spin angular momentum *S* ($S = 1$). In a magnetically ordered state, this leads to vanishing total angular momentum *J* [15], so that the magnetic moment wildly fluctuates in size provided that one is in the dilute boson density limit. Thus an intense and sharp longitudinal magnon mode is expected in such a situation. The construction of spin-density map in Ca_2RuO_4 is required to understand the singlet-triplet excitation in Ca_2RuO_4.

In the present study, we have used polarized and unpolarized single crystal neutron diffraction to determine the spatial distribution of the magnetization density in the orthorhombic phase of Ca_2RuO_4. The flipping ratio measurement (Fig.1) was carried out on untwined single crystal at 160 K (well above T_N 110K) under an applied magnetic field of 9 T along the crystallographic *c*-axis. The flipping ratios were measured at 160 K considering that (1) in-plane AFM correlation will die out up (2) the magnetization was high at this temperature, (iii) AFM peak (101) seen in the previous neutron powder diffraction measurements[ ] was not observed in our sample down to 140 K [23] on the measurements carried out at D9 instrument (which confirms high quality of the sample). In addition, we have determined the magnetic structure at 10 K, which shows that the Ru magnetic moment is canted away from the crystallographic b axis by ∼15 deg. The maximum entropy reconstruction (Fig.2) shows clearly that most of the magnetization is confined to the region around the Ruthenium atoms whereas there is no significant magnetization associated with oxygen atoms. However, in recent inelastic neutron scattering study, we have observed that the strong AFM correlation in Ca_2RuO_4 survives up to 280 K. Moreover, AFM elastic diffuse peak has been observed till 250 K. The presence of AFM correlation up to high temperatures put a question mark behind the reliability of the measured magnetization density in Ca_2RuO_4. we would therefore like to repeat the measurements at high temperature. Note that the magnetization is small at high temperatures, therefore the measuring time for the Flipping ratio will be high. At least 2 weeks of beam time on D3 instrument will be required to finish the measurements at high temperature. In addition, 2 days of beam time on D9 instrument will required to get the extinction parameters at high temperatures.

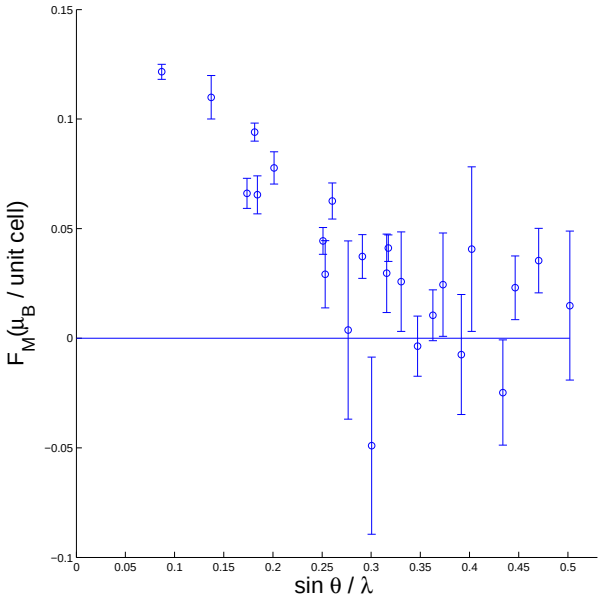

Fig. 1: Magnetic amplitudes of Ca_2RuO_4 at 160 K and 9 T.(with $B \parallel a$).

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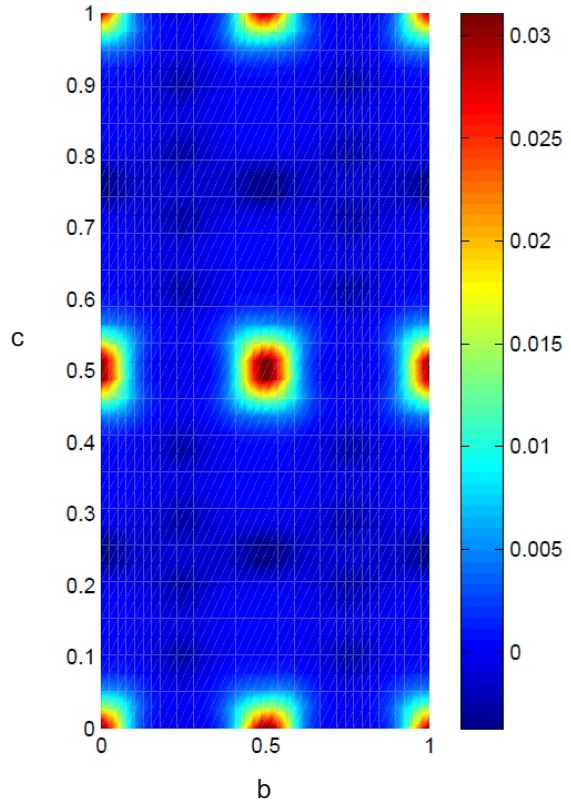

Fig. 2: Maximum entropy reconstruction of the magnetization distribution in Ca_2RuO_4 at 160 K and 9 T.