Experimental report

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Proposal: **5-51-504**

Council: 10/2014

Title: Magnetic correlations in Electric-field-induced ferromagnetic metal in Ca2RuO4

Research area: Physics

This proposal is a new proposal

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Samples: Ca2RuO4

Instrument	Requested days	Allocated days	From	To
D9	5	5	28/04/2015	04/05/2015
D3	7	7	18/06/2015	25/06/2015
D23	7	0		

Abstract:

Recently, a novel type of BEC of spin-orbit excitons has been proposed in
Mott insulators with Van Vleck-type d4 ions, such as Re3+, Ru4+, Os4+ and Ir5+
and Ca2RuO4 was suggested as a possible candidate material based on the
experimental observation of an unquenched SOC. Ca2RuO4 is a Mott insulator and It undergoes metal-insulator transition at T = 357 K [13] and orders in the A-type AFM structure below 110 K [1]. Recently, an electric field induced insulatormetal switching was reported in Ca2RuO4 with low electric field
Eth ~ 40V/cm. This switching was accompanied by a bulk
first-order structural transition. The present proposal aims to investigate the nature of magnetic ordering under an applied electric field by single crystal neutron diffraction measurements.

Experimental Report: Magnetic correlations in Electric-field-induced ferromagnetic metal in Ca_2RuO_4

Proposal ID: 5-51-504, Instrument: D9, Attending: N. Gurung, J. Bertinshaw

Ca_2RuO_4 is a *4d*-electron layered perovskite compound that exhibits a competition between electron correlation and spin-orbit coupling energies. 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 a metal-insulator transition (MIT) at 357 K and orders in the A-type antiferromagnetic (AFM) structure (with moment pointing along the *b*-axis) below 110 K [1]. A strong spin-orbit coupling (SOC) drastically changes the electronic ground state of this compound. In the limit of zero SOC, low-spin four t_{2g} electrons with nominally $S = 1$ moments (triplet) interacts 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 ($S = 1$) to orbital moments ($L = 1$) will result in the $J = 0$ non-magnetic ground state (singlet). Since SOC (which give rise to the singlet-triplet splitting) in this compound is comparable to superexchange energy, a Bose–Einstein condensation (BEC) of the exciton into a magnetic state has been theoretically predicted [2,3]. Our recent neutron scattering data for Ca_2RuO_4 support this theoretical picture [4]. The comparable energy scales of strong electron correlations and SOC in this ground state mean that Ca_2RuO_4 is highly susceptible to outside perturbation, such as application of pressure [5], chemical substitution [6], or even applied electric fields [7,8] result in a transition from an AFM insulator to FM metal ($T_C = 12$ K). Intriguingly, the electric field induced transition is unique in that the transition is stabilized with a constant applied current, even down to lowest temperatures [7].

The aim of the present experiment was to investigate the evolution of the nuclear structure, magnetic order, spin direction, and magnetic moment upon crossing the metal-insulator transition under applied electric fields. A high quality single crystal, characterized as having an AFM transition at 112 K in the normal state, was used for experiment. The sample was mounted such that horizontal scattering plane was (H0L), enabling access to strong reflections of the A-type AFM phase. Neutron diffraction experiments were carried out using the D9 diffractometer (wavelength = 0.8416 Å) at ILL, Grenoble. Contacts were made directly to the sample such that in-situ electric fields could be applied along the *c*-axis.

We started the experiment by collecting a subset of reflections at 300K in the normal state to provide a control for changes that arise to the nuclear structure after applied electric currents. At this point an electric field dependency study was performed. The applied electric field was increased systematically while studying the (0 0 6) and (1 1 6) reflections. It was identified that the metallic state was fully stabilized under an applied current of 300 mA, with a significant shift of the (006) position corresponding to an expanded *c*-axis lattice parameter, shown in Fig 1a). Upon cooling the system retained its metallic state. The MIT was completely reversible, with the original structure returning when the applied electric current was removed. The temperature evolution of the system was studied by probing the (0 0 6) and (1 1 6). In Fig 1b) clear peak splitting is evident in the (006) reflection at $T = 130$ K, indicative of a mixed phase under an applied current of $I = 300$ mA.

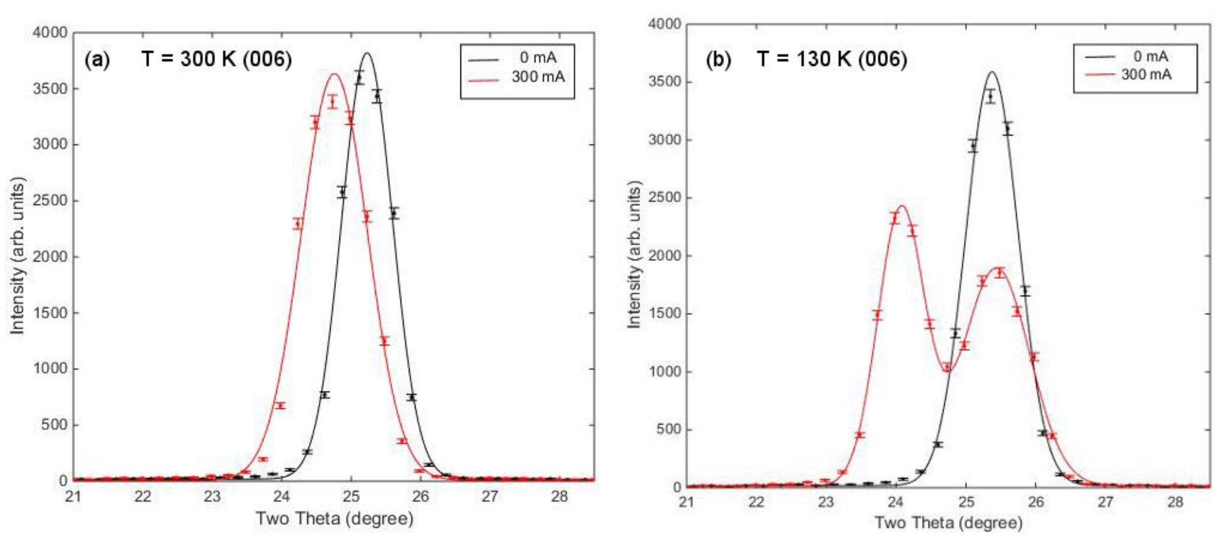

Figure 1. (a) Upon transitioning into the metallic state, the (006) reflection shows a distinct shift. This corresponded to an expansion of the c-axis lattice parameter from ~11.552 Å to ~11.755 Å at I=300 mA. (b) At 130 K, distinct peak splitting is present, indicative of a mixed phase state.

A set of ~800 reflections were measured at 130K (20K above TN) with and without an applied electric field of 300mA. All accessible reflections were measured with a *Q*-range up to 0.8 $Å^{-1}$, with further reflections corresponding to the *Pbca* space group. Preliminary refinement of the two states reveals significant structural modifications, with an expansion of the *c*-axis lattice parameter and reduced octahedral canting angles arising in the metallic state.

It was only possible to reach a minimum temperature of 40K, due to the effect of joule heating under applied electric currents. However, it was identified in this state that the AFM phase had completely vanished. Indeed no magnetic signal could be identified at *T*=40 K after a thorough exploration of reflections corresponding to possible commensurate and incommensurate spin structures. It is possible that ground state stabilized with applied electric currents is similar to the high pressure state of Ca_2RuO_4, which has been shown to exhibit ferromagnetism with T_C = 12 K. We aim to continue this experiment using the instrument D9 in order to perform a more thorough E-field dependency at lower temperatures in order to develop a full picture of the evolution of the magnetic and nuclear structures. To achieve this, modifications to the sample preparation and sample environment setup are planned to enable cooling to below *T*=12 K in order to study the potential ferromagnetic state.

References:

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