Experimental report

13/03/2019

Proposal: **5-41-946**

Council: 4/2017

Title: Metal-insulator transition in Ti-substituted Ca1.78Sr0.22RuO4

Research area: Physics

This proposal is a new proposal

Main proposer: **Kevin JENNI**

Experimental team: Kevin JENNI

Local contacts: Bachir OULADDIAF

Samples: $Ca_{1.78} Sr_{0.22} Ru_{0.9} Ti_{0.1} O_4$

Instrument	Requested days	Allocated days	From	To
D10	6	6	27/03/2018	02/04/2018

Abstract:

The phase diagram of $Ca_{2-x}Sr_xRuO_4$ shows a metal insulator transition at low Sr concentration ($x<0.2$) and a metamagnetic transition due to the competition of various magnetic instabilities at slightly higher Sr content ($0.2<x<0.6$). We recently found that Ti substitution in this latter concentration range induces a metal-insulator transition to occur as function of temperature, whose magnetic and structural properties are proposed here to be studied by four-circle neutron diffraction.

Experimental Report

Instrument	D10
Proposal Number	5-41-496
Proposal	Metal-insulator transition in Ti-substituted $Ca_{1.78}Sr_{0.22}RuO_4$
Experimentalist	Kevin Jenni, Sebastian Hoffmann, Stefan Kunkemöller, Markus Braden
Local Contact	Bachir Ouladdiaf

The material system $Ca_{2-x}Sr_xRuO_4$ exhibits a rich phase diagram showing superconductivity, metamagnetic transitions, Mott insulating states and metal-insulator transition (MIT) in the region x<0.2 [1-3]. This MIT occurs also for x>0.2 at low temperatures if Ru is substituted by a small amount of Ti. We could successfully grow single crystals of $Ca_{1.78}Sr_{0.22}RuO_4$ with 10 % Ti and wanted to study the magnetic and nuclear structure in both, the metallic and insulating phase.

a) b) c) d)

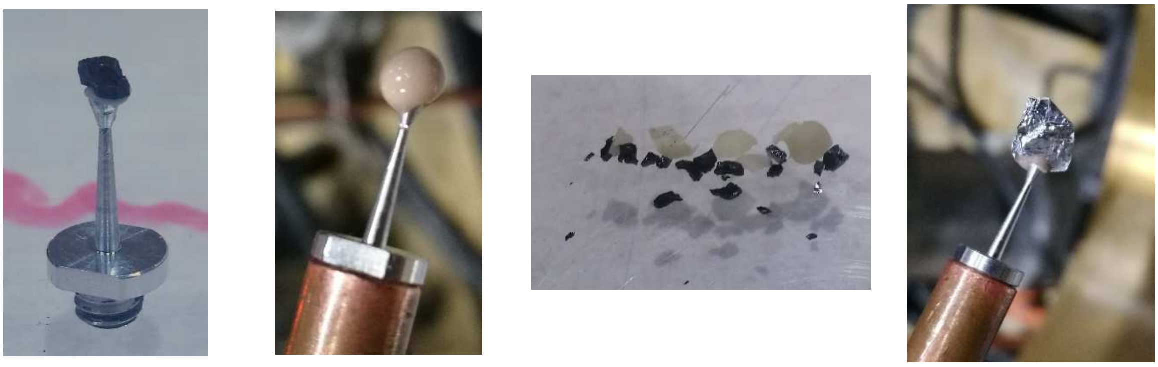

Figure 1: different samples attached to sample holder using different approaches. a) first sample attached with two-component glue, b) second sample surrounded by glue layer, c) pieces of sample 2 and hardened glue taken out of cryostat after cooling down the sample, d) sample wrapped in aluminum foil and glued to sample holder

The material $Ca_{1.78}Sr_{0.22}Ru0.9Ti_{0.1}O_4$ shows two transitions between room temperature (RT) and 2 K: a structural transition around 200 K where the crystal system changes from tetragonal to orthorhombic and the MIT around 50 K which is accompanied by a significant increase of magnetization comparable to a meta- or ferromagnetic systems. Therefore, we wanted to measure a set of nuclear and magnetic reflections at three different temperatures 295 K, 75 K and 2 K.

The first sample was attached with a two-component glue to the sample holder (Fig. 1a) and cooled down after some alignments scans at room temperature. While cooling down we recorded three nuclear reflections which disappeared under 20 K. The sample fell off during cooling and could only be recovered in pieces from the cryostat. The second sample (Fig. 4b) was covered completely in glue und cooled down to 75 K. It was possible to record a reflection set at 295 K in the tetragonal phase and another one at 75 K in the orthorhombic phase. This orthorhombic phase corresponds to a rotation of the RuO6 octahedron and does not alter the metallic properties. In order to study the insulating and magnetically ordered phase, we cooled the sample further down to base temperature and recorded again nuclear reflections while cooling. These reflections disappeared again around 18 K because the sample exploded into pieces (Fig. 4c). Apparently, the MIT in $Ca_{1.78}Sr_{0.22}Ru0.9Ti_{0.1}O_4$ is accompanied with a large structural change which leads to strains in the sample and let it break. During our macroscopic measurements this decomposition seemed to be less harmful, because smaller crystals were used, and because these crystals were grown shortly before the laboratory measurements.

Eventually crystals deteriorate with time, which is not visible as long the samples stay at room temperature, but their capacity to survive the structural phase transition accompanying the metal-insulator transition is drastically reduced. Therefore, we wrapped a third sample in aluminum foil (Fig. 4d) to keep the crystal together even when it goes through the transition and breaks. With this setup it was possible to cool the crystal down to 2 K and record a set of magnetic reflections.

The reflection set at room temperature could be refined by using the expected tetragonal space group $I4_1/acd$ with a satisfactory R-value of 5%. The reflection set at 75 K was refined with the orthorhombic space group Pbca which allows for a tilting of the RuO_6 octahedron. For the refinement the orthorhombic twins (a $\leftrightarrow$ b) occurring at the structural phase transition have to be taken into account. Because of the small amount of structural reflections at 2 K collected during the last part of beam time, it was not possible to refine the complex crystal structure with a decent R-value. However, the crystal structure of this phase is accessible by X-ray diffraction on single crystals.

The temperature dependence of two magnetic reflections (0 1 1) and (0 1 5) recorded upon cooling unambiguously established the antiferromagnetic order emerging below 50 K (Fig. 2). This result is in agreement with the macroscopic magnetization measurements. The propagation vector corresponds to an in-plane antiferromagnetic arrangement similar to the fully insulating phase of pure Ca_2RuO_4 and magnetic moments seems to point again along the b direction.

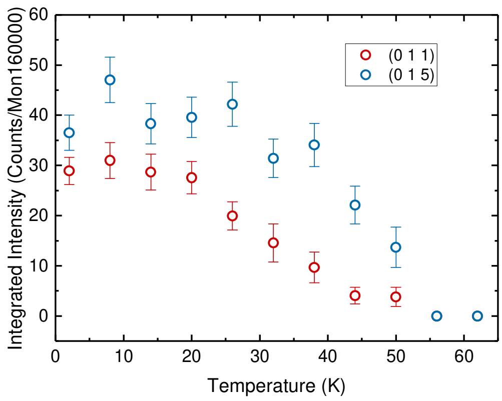

Fig. 2: Integrated Intensity of two magnetic Bragg peaks while cooling down.

References

[1] S. Nakatsuji et al.: PRB 62, 6458 (2000) and PRL 84, 2666 (2000).
[2] M. Braden et al.: PRB 58, 847 (1998); O. Friedt et al.: PRB 63, 174432 (2001).
[3] P. Steffens, et al. PRL 99, 217402 (2007); ibid. PRB 83, 054429, (2011).