Experimental report

12/09/2016

Proposal: 5-51-519 **Council:** 4/2016

Title: Spin density distribution in untwinned ferromagnetic SrRuO3

Research area: Physics

This proposal is a continuation of 5-51-499

Main proposer: Stefan KUNKEMOELLER

Experimental team: Markus BRADEN
Stefan KUNKEMOELLER

Local contacts: Anne STUNAULT

Samples: SrRuO3

Instrument	Requested days	Allocated days	From	To
D3 High field >1T	7	7	08/07/2016	16/07/2016

Abstract:

SrRuO3 is the perovskite member of the well known Ruddlesden-Popper series of ruthenates Srn+1RunO3n+1. It is the only simple material (otherwise one has to study three or higher-number layer members of the Ruddlesden-Popper series) that exhibits ferromagnetic order at ambient conditions. Magnetic hysteresis loops reveal strong anisotropic properties which are only accessible with twin-free crystals. We recently succseed in obtaining them by applying uniaxial pressure during cooling through the structural phase transitions. Such strong magnetic anisotropy can only result from some orbital preference rendering a more deep analysis of the spin density desirable. Also in view of the nearly half-metallic character it would be very interesting to resolve the orbitals carrying the magnetic moment in SrRuO3.

Experimental report 5-51-519
Spin density distribution in untwinned ferromagnetic $SrRuO_3$

1 Experimental

We used a a nearly cubic peace of single crystalline $SrRuO_3$with a mass of 60 mg [1].The crystal edges correspond to the orthorhombic directions [a]. The magnetic field-direction is the c-direction. The crystal was detwinned by cooling through the structural transition under applied stress.
First, the sample was cooled to 200 K in zero-field. There some unique nuclear Bragg-reflections of each of the six twins were measured. Then a magnetic field of 9 T was applied and the sample was cooled to 2 K. Some Bragg reflections were measured again at this temperature with an applied field followed by a large set of Flipping ratios. Further the sample was heated to 200 K and the Bragg-reflections and Flipping ratios were measured. Finally, the field was set to zero and the sample heated to 300 K and the nuclear Bragg reflections were measured a last time. The sets of flipping ratios were treated with the maximum entropy algorithm implemented in the CCSL directly. This helped to valuate the quality of the data and made it possible to optimize the data collecting strategy.

2 Results of the Flipping ratio measurement

In figure 1 the spin density distribution as obtained with the maximum entropy algorithm from the CCSL is shown for 2 K and 200 K in subfigure a) and b), respectively. Cuts parallel to the ab-plane are shown with c=0. Around the ruthenium positions at (0.5,0), (0.5,0.5) and (0.5,1) the spin density is the highest, but also at the surrounding oxygen sites a sizable magnetic density is seen. The shape of the magnetic density does not

[a]We use the Pnma setting with a=5.53 Å, b=7.84 Å and c=5.57 Å.

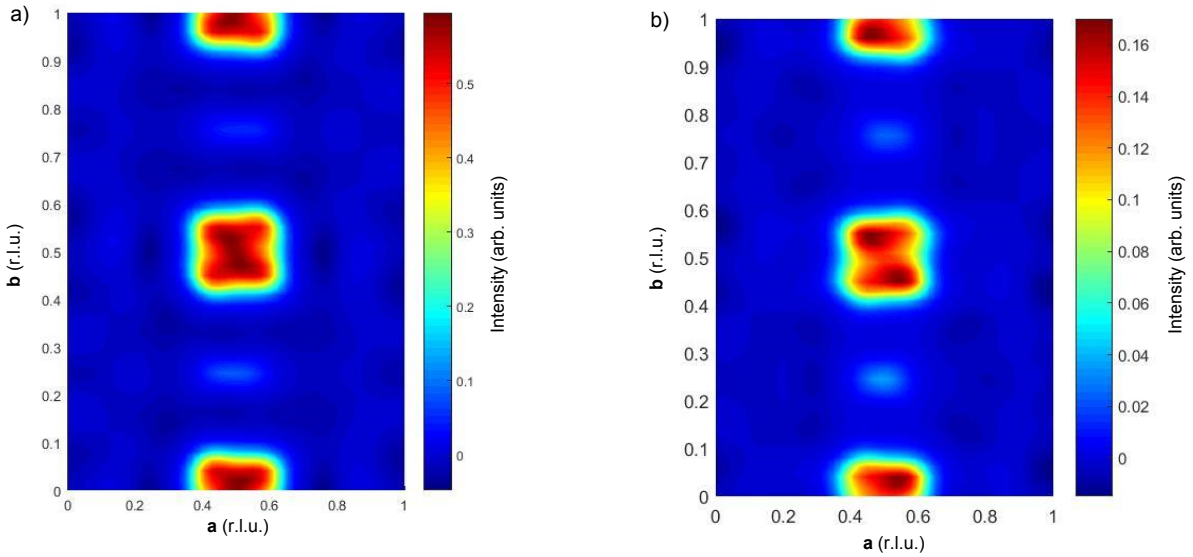

Figure 1: *Cuts parallel to the ab plane at z=0 of the spin density distribution of $SrRuO_3$ at 2 K (a)) and 200 K (b)) obtained with the maximum entropy algorithm of the CCSL. The highest spin density is found around the ruthenium positions which are at (0.5,0), (0.5,0.5) and (0.5,1).*

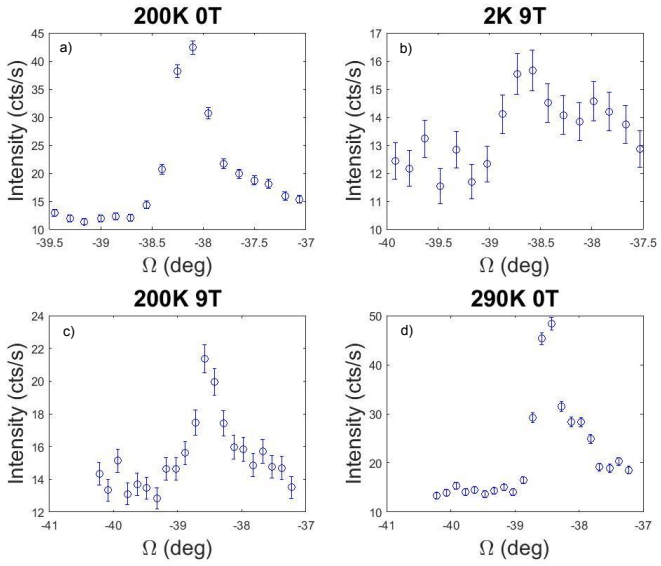

Figure 2: *(1 3 0)-reflection at different ambient conditions. The time series of the measurements is from a) to d). There were no other ambient conditions applied to the crystal in the meanwhile.*

change a lot in the paramagnetic phase at 200 K in comparison to the ferromagnetic phase at 2 K. The absolute value of the magnetic density is roughly lowered by a factor of four at 200 K. Due to the untwinned sample the precision of the spin-density maps is remarkable. We can clearly distinguish the magnetization on the two distinct O positions. In addition the spin density at the Ru position is anisotropic pointing to an imbalance in the orbital occupation of the t_{2g} orbitals.

3 Analysis of the twinning fractions

$SrRuO_3$ crystallizes in the orthorhombic perovskite structure. The symmetry of the crystal structure becomes tetragonal and then cubic on heating at about 700 K. Thus the crystal has 6 possible structural twins which considerably complicates microscopic and macroscopic studies. The ruthenium ions sit on positions with higher symmetry, so that the twins can only be distinguished by the scattering intensities resulting from the lighter ions, especially from the oxygen ions. Reflections of the kind (even odd 0) are unique in every twin. But the intensities of these reflections are very weak. By measuring these reflections at various ambient conditions we could see that the twinning fractions can be changed with a magnetic field in the ferromagnetic phase. This is remarkable because the anisotropy in this cubic system is expected to be small and the structural transition appears at much higher temperatures. The change of twinning can explain strange results previously reported for magnetoresistance. Our neutron observation that magnetic fields at low temperature may change the domain is further corroborated by our in-house studies of magneto-striction and magnetization. There seems to be a memory effect in our crystal. This was further hardened by measuring the forbidden (1 3 0) reflection. In all of the other 5 possible twins, the corresponding reflections ((0 3 1), (2 -1 -1), (-1 -1 2) , (2 -1 -1), (2 1 1)) have a sizable structure factor. So the appearance and disappearance is a proof for having only one twin or more twins in a crystal (see figure 1). It can be clearly seen that the crystal was not perfectly single-crystalline at the beginning. About 15 % of the crystal was not the desired twin of the 6 possible ones which is concluded from a comparison of the intensities of the (0 3 1) and (1 3 0) reflections. In a magnetic field of 9 T which was already applied during cooling the crystal becomes untwined. This field induced detwinning is reversed on heating and zero magnetic field.

References

[1] S. Kunkemöller, F. Sauer, A. A. Nugroho and M. Braden, Crys. Res. Technol. **51** 299–305 (2016). 1