ESRF	**Experiment title:** X-ray magnetic dichroism study of magnetic ground state across the metal insulating transition of the $Ca_{2-x}Sr_xRuO_4$ system	**Experiment number**: **HC-2381**
Beamline: ID32	**Date of experiment**: from: 03/06/2016 to: 06/06/2016	**Date of report**: 29/08/2016
Shifts: 9	**Local contact(s)**: KUMMER Kurt	*Received at ESRF:*

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Report:

In the past years the layered perovskite $Ca_{2-x}Sr_xRuO_4$ system has aroused a tremendous interest in the scientific community due to the large variety of physical properties displayed. The end member Ca_2RuO_4 is an antiferromagnetic insulator below T_N = 110 K and shows a metal-insulator transition (MIT) at 357 K [1]. In the range of x<0.2, the MIT takes place at continuously decreasing temperature as the Sr content approaches the quantum critical point at x=0.2. At the quantum critical point x = 0.2 an anomalous magnetic metallic phase appears with the occurrence of a metamagnetic transition from a state with low susceptibility to a state with high magnetic polarization [2]. A further increase of the Sr content leads the system, first, to a critical enhancement of magnetic susceptibility at x=0.5 and, finally, for x=2 (i.e. Sr_2RuO_4) to spin-triplet superconductivity below 1.5 K [3].

In the proposal we planned a X-ray magnetic circular dichroism (XMCD) study at the Ru-$M_{2,3}$ edge of $Ca_{2-x}Sr_xRuO_4$, with the goal of identifying the electronic and local magnetic structure of the Ru ions. In fact, XMCD is a powerful technique, which allows unraveling separately the contributions of spin and orbital magnetic moments. Very important for the present case, XMCD can distinguish parallel from antiparallel arrangement between spin and orbital moment, enabling us to determine the ground state symmetry. In addition the comparison of the spectral lineshape to full-multiplet calculations will allow us to unravel the details of the electronic structure of the Ru^{4+} and, in particular, will provide us with the necessary information concerning the importance of the SOC for the occurrence of metamagnetism in the x=0.2 composition and for the mechanism for the spin-triplet superconductivity pairing in Sr_2RuO_4.

For the investigation several single crystals with Sr concentrations x = 0.00 and 0.09 were grown by floating zone method. The Ru-$M_{2,3}$ XAS spectra were collected using the total electron yield method, i.e., by measuring the sample and beam drain currents. Oriented crystals were cleaved in situ in order to obtain a clean surface. The crystals were mounted with the c-axis (easy axis for magnetization) parallel to the magnetic field. As the MCD signal of the field-induced moment in the paramagnetic phase and of the canted moment in the antiferromagnetic phase is quite small many XAS spectra were collected for each temperature in order to have a good signal to noise ratio. The XAS spectra were collected in both H = 8.5 T and –8.5 T applied fields and in groups of four or quartet (*paap* or *appa*, where *a* and *p* indicate photon spin parallel or antiparallel to the applied field, respectively) in order to minimize the effect of any time dependence in the X-ray beam on the measured spectra. Ti $L_{2,3}$ XAS spectra of Sr_2TiO_4 were recorded simultaneously as energy calibration for the Ru-$M_{2,3}$ edge.

The beamtime HC-2381 is a continuation of a previous beamtime HC-2101 at the ID32 beamline of ESRF. The continuation was needed because during beamtime HC-2101 repeated problems with the new

2.5m Apple II undulator of ID32 caused the loss of two days out of the awarded four days. Hence, in the beamtime HC-2101 we could measure only one sample, a $Ca_{2-x}Sr_xRuO_4$ crystal with x=0.09 [4]. During the continuation beamtime (HC-2381) we successfully completed the planned study by measuring also the pure Ca_2RuO_4. The measurements on Ca_2RuO_4 were carried out below and above the antiferromagnetic transition (T_N = 110 K) in order to probe the antiferromagnetic and paramagnetic insulating phases.

The XMCD spectrum (green curve) measured on Ca_2RuO_4 at T=150 K (paramagnetic insulating) is reported in Fig. 1, together with the XMCD spectrum measured on the $Ca_{1.91}Sr_{0.09}RuO_4$ crystal with x=0.09 at T=115 K (antiferromagnetic insulating phase) during the previous beamtime HC-2101. From the XMCD measurements we did obtain indications for a scenario quite different from the pure spin description of magnetism in this system as commonly considered in the literature: the XMCD spectra of both $Ca_{2-x}Sr_xRuO_4$ compositions show the M_3 signal approximately twice larger in spectral weight than the M_2 one. By applying the magneto-optical sum rules to our data we could estimate the ratio L_z/S_z = 0.4(1). Similar results were obtained from the XMCD measurements of the antiferromagnetic insulating phase of Ca_2RuO_4 at T=100 K, and that of metallic phase of $Ca_{1.91}Sr_{0.09}RuO_4$ at T=160 K (>T_{MIT}). This result suggests the presence of a large orbital moment, a presence which has far reaching consequences for the interpretation of the magnetic properties and, in particular for the origin of the metamagnetic transition. As comparison we report in Fig 1 (bottom) the measurements of Okamoto et al. [4] on the metallic ferromagnet $SrRuO_3$ at the Ru-$M_{2,3}$ edge, where the M_3 and M_2 edges show an equal XMCD spectra weight, which corresponds to a quenched orbital moment for Ru^{4+}.

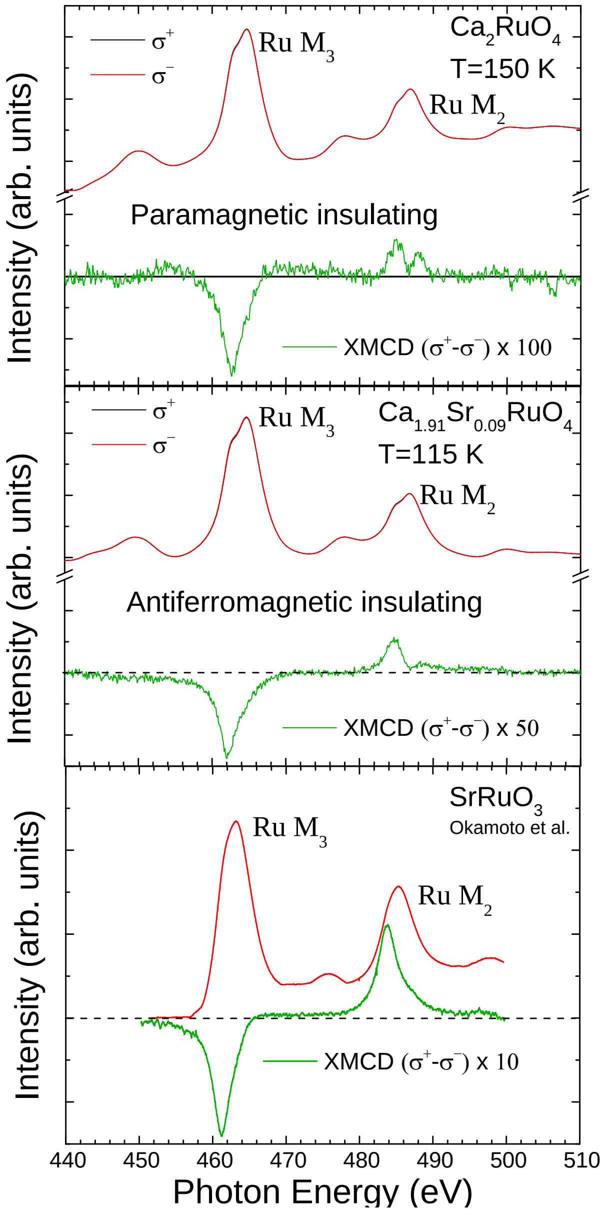

Fig.1. Top: Ru-$M_{2,3}$ XAS and XMCD spectra of Ca_2RuO_4 (top panel), $Ca_{1.81}Sr_{0.09}RuO_4$ (middle panel) single crystals measured under applied field H = 8.5 T at ID32 beamline with circular polarized soft X-rays. Bottom panel: Ru-$M_{2,3}$ XAS and XMCD spectra of $SrRuO_3$ taken from [4].

References

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