ESRF	**Experiment title:** Orbital state and metal-insulator transition of Ca3Ru2O7 investigated by x-ray absorption linear dichroism and magnetic circular dichroism	**Experiment number**: HC-2990
Beamline: ID32	**Date of experiment**: from: 13/04/2017 to: 18/04/2017	**Date of report**: 19/06/2017
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Report:

The double-layer perovskite $Ca_3Ru_2O_7$ system shows a particularly interesting and complex phase diagram as multiple phase transitions are easily induced by applied magnetic field, uniaxial or hydrostatic pressure, and doping. $Ca_3Ru_2O_7$ undergoes an AFM order transition at T_N = 56 K while remaining metallic. It is followed by a metal-to-insulator transition (MIT) at T_{MI} = 48 K [1]. Hence this system is a rare example of oxide exhibiting an antiferromagnetic metallic phase. At T = 2K, a metamagnetic transition accompanied by a drop of resistivity occurs when a field of B = 6 T is applied along the b-axis.

For the approved proposal at the ID32 beamline of ESRF we proposed a X-ray linear dichroism (XLD) study at the O *K*-edge and a X-ray magnetic circular dichroism (XMCD) study at the Ru $M_{2,3}$ edge. In the proposal we planned to carry out the XLD at O *K*-edge across the MIT in order to probe directly the evolution of the occupation of the Ru t_{2g} orbitals. The goal of the XLD investigation is to verify the scenario, proposed in literature, of a Jahn-Teller distortion-induced change in the Ru t_{2g} orbitals occupation at T_{MI} which would, via orbital ordering, cause the MIT in $Ca_3Ru_2O_7$ [1]. The same technique was previously used to reveal large changes in the orbital occupation in single layer $Ca_{2-x}Sr_xRuO_4$ [2]. The purpose of XMCD is to determine the orbital momentum in $Ca_3Ru_2O_7$. Recent XMCD studies in $Ca_{2-x}Sr_xRuO_4$ revealed a large unquenched orbital moment [3]. On the contrary, the orbital moment are nearly completely quenched in $SrRuO_3$. The determination of orbital moment would be essential for revealing the spin-orbital coupling and its role in the transitions.

For the present investigation several single crystals of $Ca_3Ru_2O_7$ were grown by floating zone method. The values of T_N and T_{MI} given by transport measurement are in agreement with literature [3]. The magnetic hysteresis loops confirms the metamagnetic transition at 6 T in 7 K. The Ru $M_{2,3}$-edge and O *K*-edge 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 or b-axis parallel to the X-ray beams as well as the magnetic field. Both the grazing- and normal-incidence conditions were used to measure spectra along various axes.

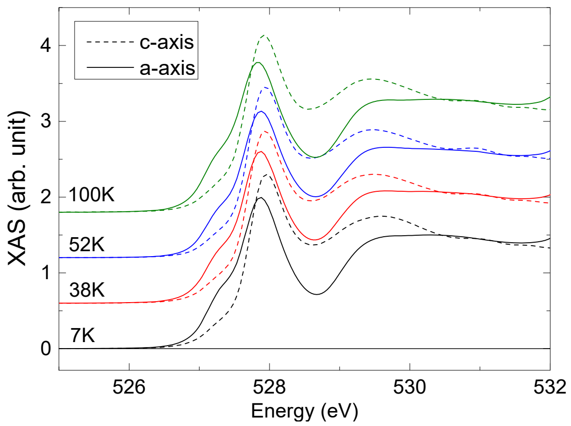

Fig. 1. O K-edge XLD along a- and c-axis in various temperatures.

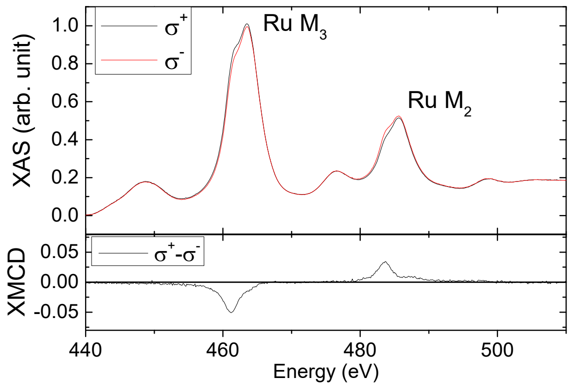

Fig. 2. Ru $M_{2,3}$-edge XMCD at 4 Tesla, 7 K along b-axis.

The XLD spectra at the O K-edge are shown in Fig. 1. The XLD between a- and c-axes at 100 K (metallic phase) indicates that the holes in the Ru t_{2g} are mainly located in the d_{xy} orbital, which corresponds to an elongated Jahn-Teller distortion [2]. Surprisingly, the XLD does not show significant changes at T_{MI} or at any temperature. These results rule out the proposed rearrangement of the Ru t_{2g} orbital occupation as the cause of the MIT and suggest the persistence of the RuO_6 octahedra elongation below T_{MI} till the lowest temperature. Our data agree with the results of neutron scattering in literature, which showed a negligible change on deformation of RuO_6 octahedron [4]. In additions, our XAS measurements show a negligible XLD between a- and b-axes both above and below T_{MI}.

The XMCD spectrum measured at 7 K under 4 T along b-axis is shown in Figure 2. The application of the sum rules to our data gives a ratio $\langle L_z \rangle / \langle S_z \rangle = 0.13(1)$ at 4 K. The same value was also obtained from the data measured at 33 K. The ratio increases to 0.17(2) after the metamagnetic transition at 6 T (ferromagnetic phase.) These values show a smaller orbital moment in $Ca_3Ru_2O_7$ compared to that in $Ca_{1.91}Sr_{0.9}RuO_4$ ($\langle L_z \rangle / \langle S_z \rangle \sim 0.4\ (1)$) [3]. These results provide us new clues for the understanding of the ground-state symmetry in $Ca_3Ru_2O_7$.

In summary, the results of the present experiment for $Ca_3Ru_2O_7$ reveal a negligible change of deformation in RuO_6 octahedron across the T_{MI} and a more quenched orbital moment in $Ca_3Ru_2O_7$ compared to $Ca_{1.91}Sr_{0.9}RuO_4$. They also suggest that other structural parameters such as Ru-O-Ru bond angle must be taken in consideration to explain the metal-insulator transition and AFM ordering. The orbital moment derived from XMCD would also provide a clue to unveil the mechanism of transition.

Reference:

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