Impact of La Doping on the Local Electronic Properties of the Ru^{4+} Ion: A Ru L-Edge XMCD Study on $Ca_{2-x}La_xRuO_4$

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Ca_2RuO_4 has attracted considerable attention as the Mott insulating analogue of the unconventional superconductor Sr_2RuO_4 [1]. It displays a metal to insulator transition (MIT) at T_{MIT}~357 K, concomitant with a first-order structural phase transition and an antiferromagnetic (AFM) ordering below T_N~110 K [2]. Contrary to many Mott insulators, the nature of the insulating ground state of Ca_2RuO_4 arises from the combination of Hund's and spin-orbit couplings (SOC) as well as crystal fields, which makes the low-energy electronic structure very sensitive to structural distortions acting on the local Ru^{4+} environment [3].

In this context, dramatic changes in the insulating state of Ca_2RuO_4 have been achieved by internal chemical pressure, where the substitution of Ca by La [4] suppress the MIT and drive the system to a metallic state. The different radii of the La^{3+} and Ca^{2+} ions cause the compressed RuO_6 octahedra of pure Ca_2RuO_4 to be progressively stretched along the c axis for increasing doping levels. This, in turn, is expected to significantly change the local physics of the Ru^{4+}.

Using a variety of X-ray scattering (REXS) [4] and absorption techniques (O K-edge XANES [5] and Ru L-edge XMCD [6]) we address in detail the impact of the structural changes on the electronic and magnetic properties of $Ca_{2-x}La_xRuO_4$ with x= 0, 0.05(1), 0.07(1) and 0.12(1). The results show that La doping induces significant changes in the local crystalline environment at the Ru sites. The AFM ordering observed in Ca_2RuO_4 parent compound persists upon La substitution [4], thus excluding the presence of a predicted ferromagnetic phase [7]. The crystal field tuning caused by the structural distortion has a visible effect on the hole population of the Ru t_{2g} orbitals [5]. Finally, both the non-statistical absorption branching ratio BR and the sizeable Ru orbital moment revealed by XANES and XMCD measurements confirm a significant impact of SOC in the low-energy physics of the insulating phase [6].

References

[1] - G. Cao, PRB 56, R2916(R) (1997).
[2] - T. Mizokawa et. al., PRL 87, 077202 (2001).
[3] - D. Sutter et. al., Nat. Comm. 8, 15176 (2017).
[4] - D. Pincini et. al., PRB 98, 014429 (2018).
[5] - D. Pincini et. al., PRB 99, 075125 (2019).
[6] - D. Pincini et. al., in preparation.
[7] - J. Chaloupka and G. Khaliullin, PRL 116, 017203 (2016).