Oxygen 1s excitation in a strongly spin-orbit coupled Sr_2IrO_4 system

Cheng, S.[1], Rocquefelte, X.[2], Zhang, H.[3], Wei, J.[4], Kim, Y.[5], Seo, A.[6] and Botton, G.[7]

[1] Department of Materials Science and Engineering, Mcmaster University, Canada, [2] Institut des Sciences Chemiques de Rennes, Univ. de Rennes, France, [3] Department of Physics, Univerity of Toronto, Canada, [4] Department of Physics, Univerisity of Toronto, Canada, [5] Department of Physics, University of Toronto, Canada, [6] Department of Physics and Astronomy, University of Kentucky, United States

The 5d transition-metal oxides have drawn much research interest in the correlated electrons physics community due to their novel physics and electronic structure behaviour. Sr_2IrO_4, a typical 5d compound, where strong spin-orbit coupling exists, has been regarded as a potential candidate for new superconductors.[1] The octahedral crystal field, combined with large spin-orbit coupling, produce one hole in t_{2g} band, forming a $j_{eff}=1/2$ state that mimics the situation in cuprates: a single hole in a square lattice, with antiferromagnetic correlations dominating, but with the t_{2g} states being involved in this case instead of the e_g that are affecting the cuprates.[2] The strong spin-orbit coupling systems are also interesting because the role of spin-orbit coupling in the electronic structure is not completely understood at this point.
We have investigated Sr_2IrO_4 thin films prepared by pulsed laser deposition [3] as a good model system for the diverse spin-orbit coupling oxides, to probe the unoccupied electronic structure as reflected by oxygen 1s excitation spectrum. For this work, we have systematically investigated, by transmission electron microscopy (TEM) methods and electron energy loss spectroscopy (EELS), both bulk regions and interfaces between films with their substrates. Using atomic resolved EELS mapping, we have explored the structure of the thin films (figure 1) and changes at interfaces, and antisite defects. For this work, we have used high energy resolution EELS spectra acquired with high energy resolution (using a FEI Titan STEM equipped with monochromator and Gatan K2 Summit direct electron detector camera attached to a Gatan Quantum spectrometer).[4] The changes in the fine structure between the bulk Sr_2IrO_4 and $SrTiO_3$ substrate show the significant differences in the unoccupied states induced by the presence of hybridization between 5d states with the oxygen 2p states (figure 2). This indicates very strong sensitivity of the O 1s spectrum to changes in the electronic structure, particularly at the interface. Through density functional calculations, we simulate the EELS near edge structures by considering LDA+U and spin-orbit coupling effects. The combination of spatially resolved high energy resolution EELS with first principle calculations is demonstrated as a powerful approach necessary for a comprehensive understanding of the electronic structure of strong spin-orbit coupling systems, offering new possibilities for elucidating their physical properties.

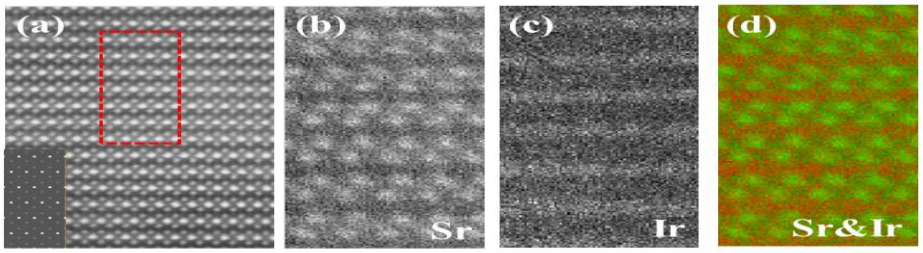

FIG. 1. High resolution STEM image and the corresponding EELS mapping results. (a) High resolution STEM image showing the high quality of the Sr_2IrO_4 thin film. The corresponding simulated Sr_2IrO_4 image is embedded at the bottom left corner. (b) EELS mapping for the Sr L_{23} edge from the red rectangle area in (a); (c) Ir M_{45} edge mapping (c). (d) Colour coded maps of Sr (Green) and Ir (red) signals.

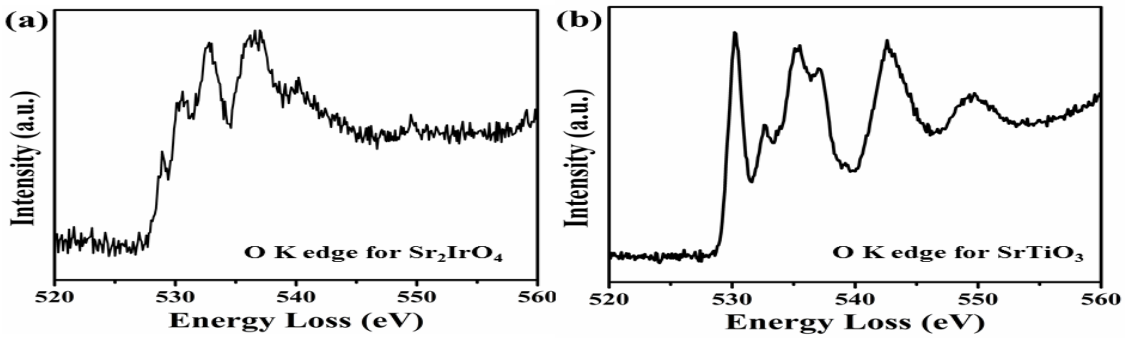

FIG. 2. The typical oxygen K edge EELS spectra for the Sr_2IrO_4 thin film (a) and $SrTiO_3$ substrate (b).

References:

[1] Y. K. Kim, N. H. Sung, J. D. Denlinger, and B. J. Kim, Nat. Phys. **12**, 37 (2016)
[2] B. J. Kim, Hosub Jin, S. J. Moon, et al. Phys. Rev. Lett. **101**, 076402 (2008)
[3] A. Seo, J. Nichols, J. Hwang *et al.*, Appl. Phys. Lett. **109**, 201901 (2016)
[4] J. L. Hart, A. C. Lang, A. C. Leff, P. Longo, C. Trevor, R. D. Twesten, M. L. Taheri, Sci. Rep. **7**, 8243 (2017)

Acknowledgement:

G. A. B and S. C. are grateful to NSERC for a Discovery Grant supporting this work. Electron microscopy was carried out at the Canadian Centre for Electron Microscopy (CCEM), a national facility supported by NSERC, the Canada Foundation for Innovation, and McMaster University.