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Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** **Possible J=1/2 state in thin films of Ir-based double perovskites Sr_2MnIrO_6**	**Experiment number**: HC 3251
Beamline: ID-12	**Date of experiment**: from: 30.08.17 to: 04.09.17	**Date of report**: 02.11.17
Shifts: 15	**Local contact(s)**: ROGALEV Andrei	*Received at ESRF:*

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Report:

3d-5d based compounds have drawn an increased scientific interest due to interplay between strong correlation effects in 3d-ions and large spin-orbit-coupling (SOC) in 5d-ions[1]. Recent theory and experiments reveal Ir^{4+} ($5d^5$) to exhibit effective total angular momentum $J = ½$ due to large SOC[2,3]. Even a small Coulomb repulsion U may open a Mott gapped state in Iridates, which results in $J = ½$ due to the spin-orbital Mott state. Here, we report on investigation of the magnetic properties of epitaxial thin films of new Ir^{4+} based compounds Sr_2MnIrO_6 and $LaSrMnIrO_6$ with different epitaxial strain stabilized using pulsed laser deposition on $SrTiO_3$, $DyScO_3$, and $(LaAlO_3)_{0.3}(Sr_2TaAlO_6)_{0.7}$ substrates.

The total magnetic moments of Sr_2MnIrO_6 and $LaSrMnIrO_6$ thin films on the $SrTiO_3$ substrates were characterized using a superconducting quantum interference device (SQUID). The total magnetization curves for both compounds are shown in Fig. 1 as functions of the applied field (a, c) and temperature (b, d). The magnetization *vs.* field curves for both compounds display well defined hysteresis loops with different anisotropies and the saturation magnetizations of 0.4 and 0.6 μ_B/f.u. for the Sr_2MnIrO_6 and $LaSrMnIrO_6$ thin films, respectively. In Sr_2MnIrO_6, interaction between anti-ferromagnetic insulator $SrMnO_3$ and paramagnetic semi-metal $SrIrO_3$ layers gives rise to a ferri-magnetic low-band gap insulating behaviour. As compared to Sr_2MnIrO_6, the La-doped $LaSrMnIrO_6$ compound shows higher saturation magnetization and magnetic Curie temperature (T_c). In order to characterize the contribution of spin-orbit coupling and to compare the role of magnetism in Ir for both compounds, x-ray magnetic circular dichroism (XMCD) measurements of the Sr_2MnIrO_6 and $LaSrMnIrO_6$ thin films were performed at the Ir $L_{2,3}$ x-ray absorption edges.

Absorption spectra were recorded using the total fluorescence yield detection mode. The XMCD spectra for both Sr_2MnIrO_6 and $LaSrMnIrO_6$ films were obtained as direct difference between consecutive X-ray Absorption Near Edge Spectrum (XANES) scans recorded with opposite helicities of the incoming x-ray beam. All measurements were performed at 5 K which is well below the observed T_c for both materials.

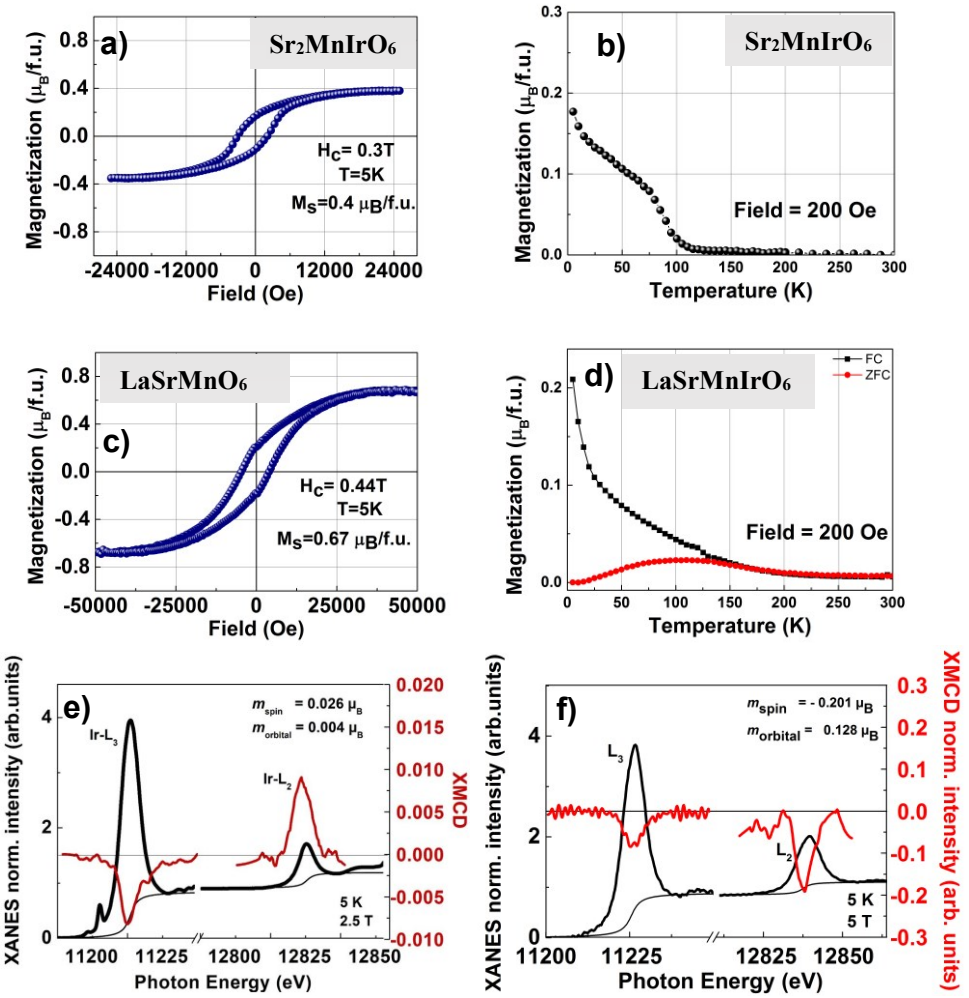

Figure 1: (a,c) Magnetization *vs.* field and (b,d) magnetization *vs.* temperature curves for Sr_2MnIrO_6 and $LaSrMnIrO_6$ thin films measured with a SQUID magnetometer. XANES and XMCD spectra of the Ir$L_{2,3}$ edges for (e) Sr_2MnIrO_6 and (f) $LaSrMnIrO_6$ thin films measured at 5 K.

The measured XANES spectra with the correspondent XMCD signals for Sr_2MnIrO_6 and $LaSrMnIrO_6$ are shown in Fig. 1 (e) and (f), respectively. For Sr_2MnIrO_6, we derived a positive XMCD signal with the corresponding spin magnetic moment of $m_{spin} \approx 0.026\ \mu_B$ and orbital moment of $m_{orbital} \approx 0.004\ \mu_B$ ($|m_{orbital}/m_{spin}| \approx 0.15$) resulting in a total paramagnetic moment of $m_{tot} \approx 0.030\ \mu_B$ per Ir atom. For $LaSrMnIrO_6$, the negative XMCD signal has been observed and the total magnetic moment of Ir is aligned opposite to the net magnetization. Quantitavely, a spin magnetic moment of $m_{spin} \approx -0.201\ \mu_B$ and an orbital magnetic moment of $m_{orbital} \approx 0.129\ \mu_B$ were calculated using the sum rules, i.e. $|m_{orbital}/m_{spin}| \approx 0.64$ and $m_{tot} = -0.072\ \mu_B$ at the Ir-site. Thus, doping of Sr_2MnIrO_6 with La results in the Mn^{3+} ($3d^4$) state in $LaSrMnIrO_6$ with an extra e_g electron, which is effectively hybridized with the Ir 5d shell leading to the observed induced negative total magnetic moment at the Ir-site.

In summary, based on our XMCD measurements, we confirm presence of an induced magnetic moment at the Ir-site in $LaSrMnIrO_6$ due to hybridization of the e_g electron between the Mn^{3+} ($3d^4$) and Ir^{4+} ($5d^5$) states. Quantitative estimations of spin and orbital moment in the case of $LaSrMnIrO_6$ confirm an unquenched orbital moment at the Ir-site, which contributes to the observed total magnetic moment. In Sr_2MnIrO_6, the observed small paramagnetic moment at the Ir-site indicates that hybridization between the t_{2g} electrons of the Mn^{4+} ($3d^3$) configuration and the Ir 5d shell is supressed. Unfortunately, we could not perform XMCD measurements as a function of the Sr_2MnIrO_6 strain state as the thin-film samples on $DyScO_3$ substrates were detached from the sample holders on application of strong magnetic fields due to high paramagnetic moment of Dy. Moreover, in the case of $(LaAlO_3)_{0.3}(Sr_2TaAlO_6)_{0.7}$ substrates, XANES spectra at the Ir $L_{2,3}$-edges were masked by signal from nearby Ta-edge.

[1] A. Kolchinskaya et al Phys. Rev. B **85**, 224422 (2012).
[2] H. Zhang et al Phys. Rev. Lett. **111**, 246402
[3] Woo Jin Kim, et al Phys. Rev. B **93**, 045104