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Report:

Iridate materials attracted a lot of attention because of the new physics that evolves due to the strong spin orbit coupling (SOC). The strong SOC splits the $5d$ t_{2g} bands into a lower lying J_{eff}=3/2 quadruplet and a higher lying J_{eff}=1/2 doublet. The main attention has been focussed on Ir^{4+} ($5d^5$) compounds where electronic ground states with non-trivial band topologies and the signatures for compass exchange interactions have been predicted.

Far less work has been done on the Ir^{5+} ($5d^4$) primarily because the strong SO coupling limit is expected to lead to a non-magnetic singlet ground state J_{eff} = 0 state arising from four electrons filling the J_{eff}=3/2 quadruplet. According to this picture the double perovskite Sr_2YIrO_6 should be a band insulator with no magnetism. Contrary to this a magnetic moment [1,2] and magnetic transition at 1.3 K [1] has been observed, both associated with a small magnetic moment of Ir. According to Cao *et al.* the small magnetic moment is due to a *novel magnetism* of Ir^{5+} ($5d^4$) where the J_{eff} = 0 is affected by the entanglement of non-cubic CEF interactions, local exchange interactions and SOC [1] thus invalidating the strong SOC limit and opening up an array of interesting new physical questions. Contrary to this explanation, Wakeshima *et al.* suspect the magnetism to be due to some minor impurity of magnetic Ir^{4+} ($5d^5$) [2]. Alternative scenarios, like Van Vleck-type excitation or superexchange stabilization of J_{eff} = 1, 2 states have been proposed as possible origin of the magnetic ground state because the singlet and triplet states are close in energy [3,4].

For the allocated beamtime we proposed to carry out x-ray magnetic circular dichroism (XMCD) experiments at the ID12 beamline on several Ir^{5+} oxides at the Ir-$L_{2,3}$ edges. We measured also Sr_3CuIrO_6 as an Ir^{4+} ($5d^5$) reference sample. High quality single crystals of Sr_2YIrO_6, Sr_2GdIrO_6, Sr_2CoIrO_6 and Sr_3CuIrO_6 were grown and high quality polycrystalline samples of Sr_2CoIrO_6 and $Sr_2Co_{0.5}Ir_{0.5}O_4$ were synthesized for the proposed experiment. The XMCD measurements on ID12 were carried out at T = 2 K under an applied field of B = 17 T and the results are plotted in Figure 1. Sr_2GdIrO_6 was not measured because there was not sufficient time. XMCD of Sr_2CoIrO_6 was measured on the polycrystalline sample. It was annealed under high oxygen pressure and preliminary soft-XAS measurements at the Co $L_{2,3}$ revealed that is has a better oxygen stoichiometry than the single crystal.

The XMCD signal of Sr_2YIrO_6, shown in Fig 1 (a), is extremely small, as expected for a Van Vleck system, and long data collecting time was necessary to have sufficient statistics. The XMCD signals of Sr_2CoIrO_6 and $Sr_2Co_{0.5}Ir_{0.5}O_4$ (Fig 1 (b) and (c)) are also small, but approximately 5 times larger than that of

Sr$_2$YIrO$_6$. The difference in the XMCD signal size might be explained as an effect of the exchange field felt by the Ir^{5+} ions in Sr$_2$CoIrO$_6$ and Sr$_2$Co$_{0.5}$Ir$_{0.5}$O$_4$, and caused by the neighbouring magnetic Co^{3+} (S=2) ions.

Sr$_3$CuIrO$_6$, our Ir^{4+} ($5d^5$) reference system, has also a quite small XMCD signal, which could be attributed to an antiferromagnetic arrangement of the iridium magnetic moments still persisting at an applied field as high as 17 Tesla.

The XMCD signal at the Ir L_3 edge is much larger than the signal at Ir L_3 edge for nearly all samples, which indicates the presence of a large orbital moment. The only exception is the double perovskite Sr$_2$CoIrO$_6$, where the XMCD at the Ir L_3 and L_2 edges are approximately equal in size and opposite in sign. Hence, our measurements suggest that the Ir orbital moment in Sr$_2$CoIrO$_6$ is nearly quenched. In order to be more quantitative, we have applied the sum rules developed by Thole et al. and Carra et al. to extract directly from the spectrum the ratio:

$$\frac{L_z}{2S_z + 7T_z} = \frac{2\int_{L3}\Delta\mu(E)dE + \int_{L2}\Delta\mu(E)dE}{3\int_{L3}\Delta\mu(E)dE - 2\int_{L2}\Delta\mu(E)dE}$$

The results of the application of the sum rules to our data are reported in Tabel 1. Please be aware that the values reported in the table do not correspond to the ratio of orbital moment to the spin moment, $L_z/2S_z$, because for $5d$ transition metals the intra-atomic magnetic dipole moment T_z is not negligible. In any case, the sum rules confirm that the orbital moment in Sr$_2$CoIrO$_6$ is surprisingly small. Cluster calculations to simulate the measured spectra and to estimate T_z are in progress.

sample	configuration	$L_z/(2S_z+7T_z)$
Sr$_2$YIrO$_6$	d^4	0.29
Sr$_2$CoIrO$_6$	d^4	0.006
Sr$_2$Co$_{0.5}$Ir$_{0.5}$O$_4$	d^4	0.45
Sr$_3$CuIrO$_6$	d^5	0.89

Table 1. Results of the application of the sum rules to the measured Ir-$L_{2,3}$ XMCD spectra

References

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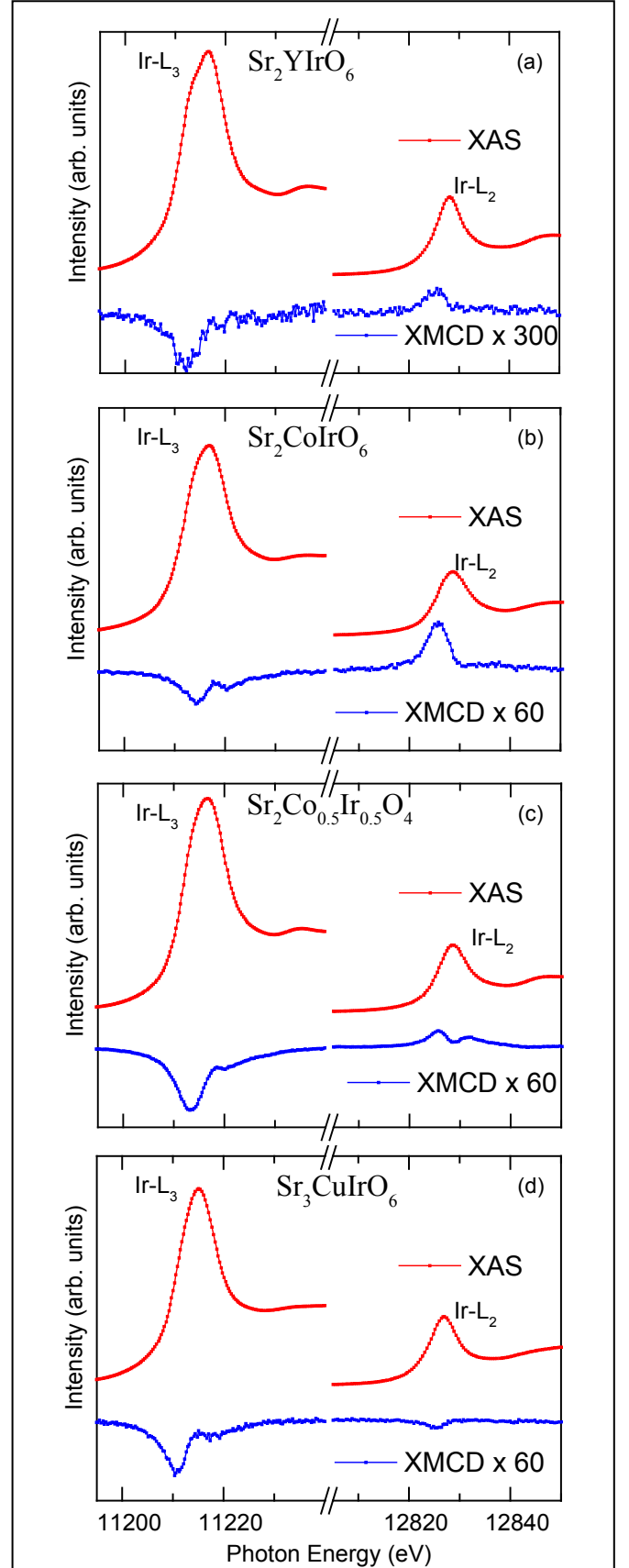
Fig.1. Ir-$L_{2,3}$ XAS and XMCD spectra of Sr$_2$YIrO$_6$ (a), Sr$_2$CoIrO$_6$ (b), Sr$_2$Co$_{0.5}$Ir$_{0.5}$O$_4$ (c) and Sr$_3$CuIrO$_6$ (d) measured at T = 2 K under applied field H = 17 T at ID12 beamline with circular polarized soft X-rays.