

Evolution of structural and magnetic properties of Ti doped Sr_2IrO_4 : a novel magnetic insulator

Imtiaz Noor Bhatti¹ and A. K. Pramanik

School of Physical Sciences, Jawaharlal Nehru University, New Delhi-110067, India

ABSTRACT

5d transition metal oxides (TMOs) recently caught attention of researchers because of potential to show exotic phenomenon. Crystal field effect (CFE), spin-orbital interaction (SOI) and onsite Coulomb interaction (U) in 5d oxides are on comparable scale and their interplay among themselves setting new balance of energies and drive exotic physics in these materials. Among several 5d oxides iridates are extensively studied, where Sr_2IrO_4 is prototype of novel physics and stabilize in $J_{\text{eff}} = 1/2$ ground state. The Sr_2IrO_4 is first member of Rudellson-Popper series $\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$ with $n = 1$ and crystallize in reduced tetragonal structure and adopt $I4_1/acd$ space group. The physical properties of Sr_2IrO_4 are influenced by this reduced symmetry due to rotation of IrO_6 octahedral. The magnetic ground state in Sr_2IrO_4 is canted type antiferromagnetic (AFM), it's this canted spin structure which gives ferromagnetic character to this material with ordering temperature around 225 K to this material. The spin canting is rendered by Dzyaloshinskii-Moriyam anti-symmetric interaction driven by SOI and rotated IrO_6 octahedra. We endeavour to perturb the interactions viz. SOI and U in Sr_2IrO_4 by substituting Ti^{4+} ($3d^0$ $S = 0$) for Ir^{4+} ($5d^5$ $J_{\text{eff}} = 1/2$). We have studied the effect of Ti doping on structural and magnetic properties. We observe evolution in unit cell lattice parameters, in the meantime Ir-O-Ir bond angle also increases which reduces octahedral rotation (θ_{Oct}). Further, our magnetization study shows decrease in transition temperature (T_C), increase in effective magnetic moment and doping influence the coercively and remnant magnetization. Thus we observe Ti doping prominently effect the structural and magnetic properties in Sr_2IrO_4 . The results presented here provide insight into the effect of Ti doping on structure and magnetic phase in strong spin-orbit coupled Sr_2IrO_4 .

Keywords : Doping, Insulator, magnetization, spin-orbit coupling

Corroresponding Author: inbhatti07@gmail.com



1. INTRODUCTION

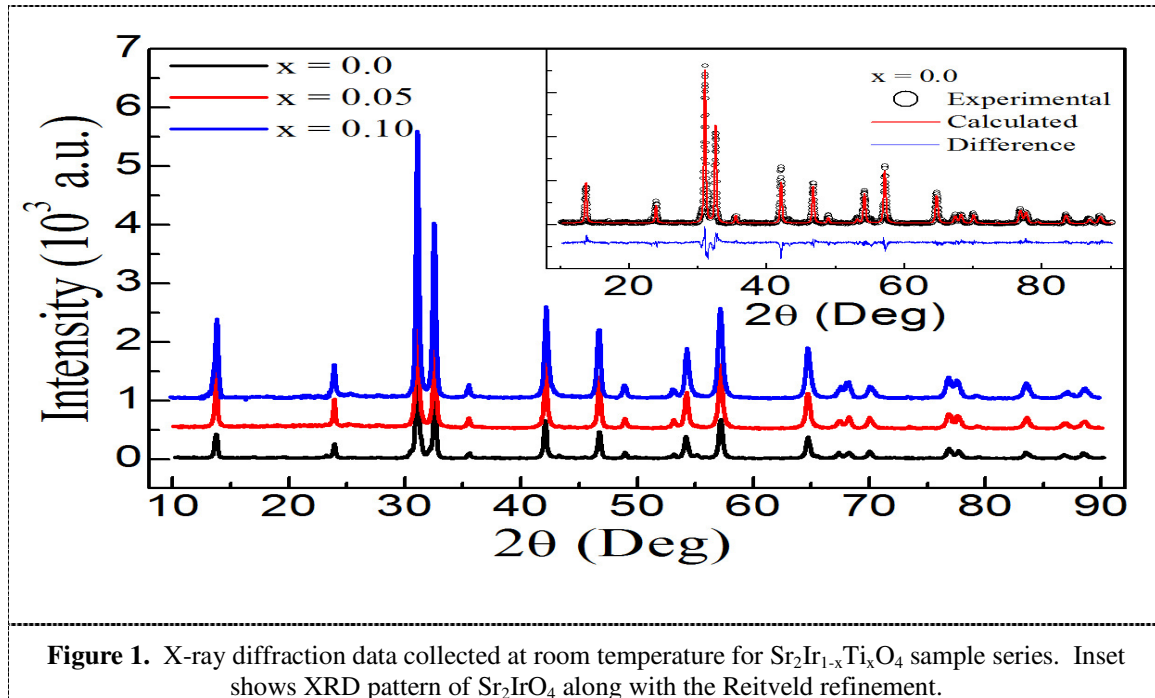
Recently 5d-based iridates and osmates have received much attention of researchers due to exotic physics arising from competing interactions viz. crystal field effect (CFE), spin-orbital interaction (SOI) and onsite Coulomb interaction (U).[1-5] Among them the most extensively studied iridate is the layered perovskite Sr_2IrO_4 , believed to stabilize in $J_{\text{eff}} = 1/2$ ground state and it is a spin orbital coupling driven Mott insulator. Sr_2IrO_4 having similarities with high T_C cupric superconductor La_2CuO_4 draw interest, as it has also been theoretically predicted to be superconductor electrons/hole doping. In 5d iridates active interactions such as: (CEF), (U) and (SOI), compete among themselves setting new level of energies and endorse novel magnetic phases in these materials. It is recently found that the ground state is driven by strong SOI of Ir ion in an octahedral environment of oxygen atoms which splits the t_{2g} band into lower $J_{\text{eff}} = 3/2$ quartet and upper $J_{\text{eff}} = 1/2$ doublet. The $J_{\text{eff}} = 1/2$ band is so narrow that even reduced U opens a Mott gap by splitting it into upper and lower Hubbard bands, thus driving it to unconventional Mott insulator.[1-2] The Sr_2IrO_4 crystallize in reduced tetragonal structure with space group $I4_1/acd$. The reduced symmetry is due to rotation of IrO_6 octahedra around c-axis and play important role in deciding the physical properties of Sr_2IrO_4 . [1-7]

The magnetic ground state in Sr_2IrO_4 is believed to be canted type antiferromagnetic (AFM) which gives ferromagnetic component with magnetic ordering around 225 K to this material. [5-11] The magnetic ground state is quite delicate and can be perturbed and destabilized by tuning any of the interaction band filling, SOC and U. To achieve this motivation chemical doping is often employed. Here we have doped Ti^{4+} as substitution for Ir^{4+} which introduces holes and reduces the SOC while increasing U.

In this paper we investigate the evolution of structural and magnetic properties of $\text{Sr}_2\text{Ir}_{1-x}\text{Ti}_x\text{O}_4$ ($x = 0.0, 0.05$ and 0.1). We observe evolution of structure parameters with Ti doping but no structure phase transition is observed. The magnetization is studied in detail; all samples are ferromagnetic in nature. However, the transition temperature decreases with doping, whereas effective magnetic moment increases. The isothermal magnetization shows decrease in coercivity and remnant magnetization.

2. EXPERIMENTAL DETAIL

The polycrystalline samples of $\text{Sr}_2\text{Ir}_{1-x}\text{Ti}_x\text{O}_4$ with composition $x = 0.0, 0.05$ and 0.1 are prepared using a solid state reaction technique. The starting materials IrO_2 (Sigma Aldrich 99.99 %), TiO_2 (Sigma Aldrich 99.99 %), and SrCO_3 (Sigma Aldrich 99.95 %), are taken in stoichiometric composition and ground well. The fine powder is then calcined at 900°C for one day. The calcined powder is sintered twice at 1000°C and 1100°C with intermediate grindings, the detail procedure is described elsewhere.[9,10,11] Samples in powder form are characterized by x-ray diffraction (XRD). The structural analysis is done on XRD data using FullProf software. DC magnetization (M) data have been collected using a vibrating sample magnetometer (PPMS, Quantum Design).



3. RESULTS AND DISCUSSION

3.1. Structural study

Fig. 1 shows the XRD pattern of $\text{Sr}_2\text{Ir}_{1-x}\text{Ti}_x\text{O}_4$ sample series, we observe that there is no extra peak present in doped samples which shows samples are in single phase. Inset Fig. 1 shows Rietveld refinement of parent sample with $x = 0.0$. The structural analysis on all samples shows the samples crystallizes in single phase and adopts tetragonal structure with $I4_1/acd$ space group. The refined lattice parameters are $a = b = 5.4980(2)$ Å and $c = 25.779(1)$ Å for parent sample, with Ti doping lattice parameter evolve slightly. We also find a rotation of IrO_6 octahedra decreases from $\sim 11.3^\circ$ for $x = 0.0$ to 8° for $x = 0.1$ around c axis.

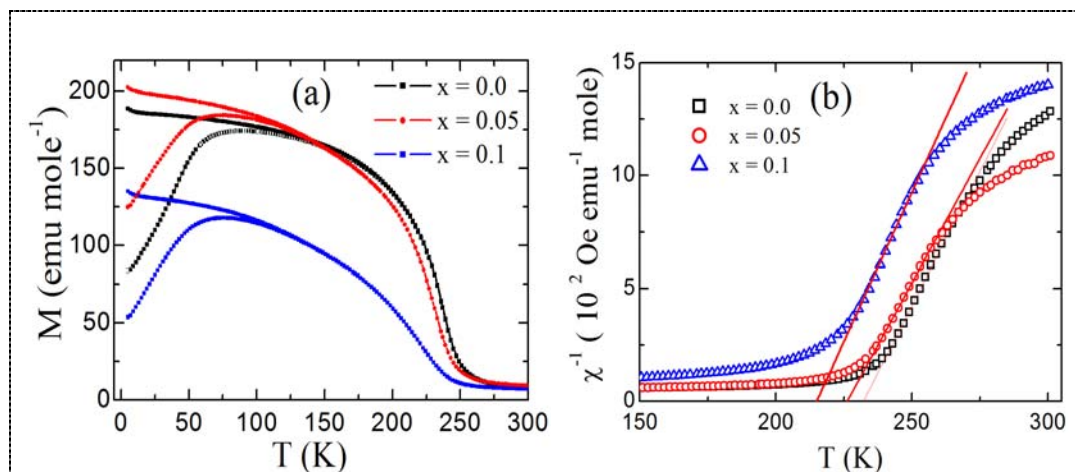


Figure 2. (a) ZFC and FC magnetization data measured in 10 kOe are plotted as a function of temperature for $\text{Sr}_2\text{Ir}_{1-x}\text{Ti}_x\text{O}_4$. (b) Inverse susceptibility (χ^{-1}) shows Curie-Weiss behaviour.

3.2. Magnetization Study

Fig. 2(a) shows the temperature dependant magnetization $M(T)$ data collected in ZFC and FCW protocol in the temperature range of 5 K – 300 K in an applied field of 10 kOe. The $M(T)$ data show a phase transition from paramagnetic (PM) to weak ferromagnetic (FM) state with $T_C \sim 225$ K for the parent compound. However, with Ti doping moment and T_C decreases. Fig. 2(b) shows the inverse susceptibility as a function of temperature. In PM state the susceptibility is fitted with Curie-Weiss law expressed as:

$$\chi = \frac{C}{T - \theta_p}$$

Where χ is magnetic susceptibility, C is the Curie constant and θ_p is the Curie temperature.

We have calculated effective paramagnetic moment using fitting parameters for all samples $\mu_{\text{eff}} = 0.56 \mu_B/\text{f.u.}$ for parent compound. With Ti doping μ_{eff} increases which reflect the weakening of exchange interaction, making spins free to align in field direction easily.

To further understand the magnetic properties we measured isothermal magnetization $M(H)$ at $T = 5$ K shown in Fig. 3(a). The hysteresis in $M(H)$ is suggestive of FM ordering in this material. At 5 K, the magnetic moment $\mu_S = 0.05 \mu_B/\text{f.u.}$ at $H = 70$ kOe is observed, which decreases with Ti doping. It is quite evident from the Fig. 3(a) that coercivity and remanance decreases with doping. Fig. 3(b) shows the Arrott plot (M^2 vs H/M), the extrapolation of Arrott plot at high field intercept the positive M^2 axis which realize the presence of spontaneous moment in these samples.

The suppression of transition temperature T_C and moment can be explain by considering the effect of Ti substitution since Ti^{4+} is $S = 0$ is a non-magnetic and hence break the Ir-O-Ir magnetic channel thus weaken the exchange interactions and suppress the T_C and decreases magnetic moment.

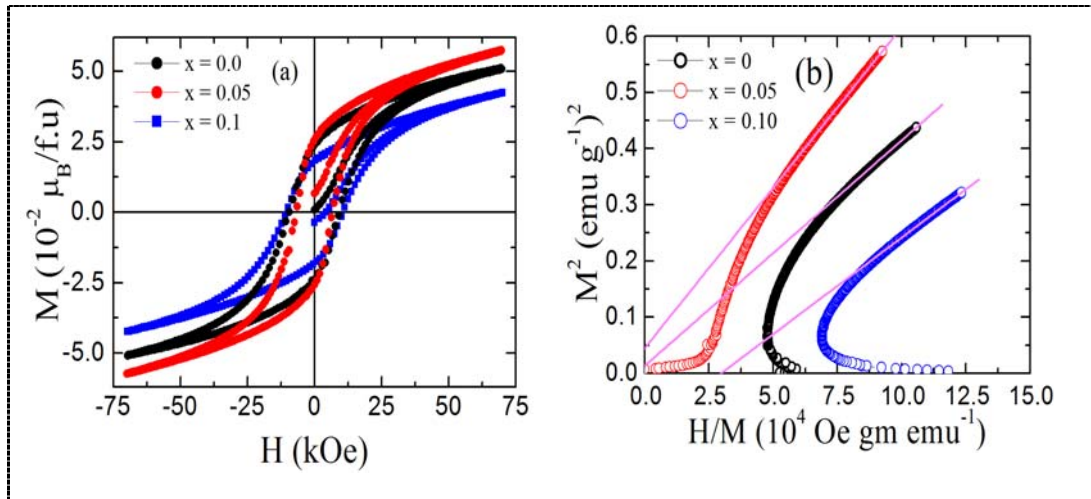


Figure 3. (a) Isothermal magnetization is shown for $\text{Sr}_2\text{Ir}_{1-x}\text{Ti}_x\text{O}_4$ at 5 K. (b) Arrott Plot (M^2 vs H/M) solid lines due to extrapolation at high field.

4. CONCLUSIONS

We have prepared single phase polycrystalline samples of $\text{Sr}_2\text{Ir}_{1-x}\text{Ti}_x\text{O}_4$. Rietveld analysis of XRD data shows samples adopt tetragonal structure with crystal structure $I4_1/acd$ space group. The detail temperature dependent magnetization $M(T)$ study shows the moment decreases with Ti doping and suppression in T_C . Ti doping breaks the Ir-O-Ir chain thus weakens the exchange interaction. $M(H)$ data shows that all samples are ferromagnetic in nature. However, coercivity and remnant magnetization decreases with doping.

ACKNOWLEDGMENTS

We acknowledge UGC-DAE CSR, Indore for measurement facilities. One of the authors (INB) acknowledges the CSIR-India for fellowship.

REFERENCES

- [1] B.J. Kim, Hosub Jin, S.J. Moon, J.-Y. Kim, B.-G. Park, C.S. Leem, Jaejun Yu, T.W. Noh, C. Kim, S.-J. Oh, J.-H. Park, V. Durairaj, G. Cao, and E. Rotenberg, 2008, Physical Review Letter, Vol. 101, pp. 076402.
- [2] S.J. Moon, H. Jin, K.W. Kim, W.S. Choi, Y.S. Lee, J. Yu, G. Cao, A. Sumi, H. Funakubo, C. Bernhard, and T.W. Noh, 2008, Physical Review Letter, Vol. 101, pp. 226401.
- [3] M.K. Crawford, M.A. Subramanian, R.L. Harlow, J.A. Fernandez-Baca, Z.R. Wang, and D.C. Johnston, 1994, Physical Review B, Vol. 49, 9198.
- [4] M. Ge, T.F. Qi, O.B. Korneta, D.E. De Long, P. Schlottmann, W.P. Crummett, and G. Cao, 2011, Physical Review B, Vol. 84, pp. 100402.
- [5] J.S. Lee, Y. Krockenberger, K.S. Takahashi, M. Kawasaki, and Y. Tokura, 2012, Physical Review B, Vol. 85, pp. 035101.
- [6] T.F. Qi, O.B. Korneta, L. Li, K. Butrouna, V.S. Cao, X. Wan, P. Schlottmann, R.K. Kaul, and G. Cao, 2012, Physical Review B, Vol. 86, pp. 125105.
- [7] T. Shimura, Y. Inaguma, T. Nakamura, M. Itoh, and Y. Morii, 1995, Physical Review B, Vol. 52, pp. 9143.
- [8] S. Calder, G.-X. Cao, M.D. Lumsden, J.W. Kim, Z. Gai, B.C. Sales, D. Mandrus, and A.D. Christianson, 2012, Physical Review B, Vol. 86, pp. 220403.
- [9] I.N. Bhatti, R. Rawat, A. Banerjee, and A.K. Pramanik, 2014, Journal of Physics: Condense Matter, Vol.27. No. 1, pp. 016005.
- [10] I.N. Bhatti and A K Pramanik, 2017, Journal of Magnetism and Magnetic Material, Vol. 422, pp. 141.
- [11] I.N. Bhatti and A.K. Pramanik, 2017, American Institute of Physics Conference Proceedings, Vol. 1731, pp. 110046.
- [12] I.N. Bhatti, R.S. Dhaka, and A.K. Pramanik, 2017, Physical Review B, Vol. 96. No. 14, pp. 144433.