Evolution of structural and magnetic properties of Ti doped Sr$_2$IrO$_4$: a novel magnetic insulator

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ABSTRACT

5$d$ 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 5$d$ oxides are on comparable scale and their interplay among themselves setting new balance of energies and drive exotic physics in these materials. Among several 5$d$ oxides iridates are extensively studied, where Sr$_2$IrO$_4$ is prototype of novel physics and stabilize in $J_{\text{eff}} = \frac{1}{2}$ ground state. The Sr$_2$IrO$_4$ is first member of Rudellson-Popper series Sr$_{n+1}$Ir$_n$O$_{3n+1}$ with $n=1$ and crystallize in reduced tetragonal structure and adopt $I4_1/acd$ space group. The physical properties of Sr$_2$IrO$_4$ are influenced by this reduced symmetry due to rotation of IrO$_6$ octahedral. The magnetic ground state in Sr$_2$IrO$_4$ 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$_2$IrO$_4$ 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 ($\theta_{\text{Od}}$). 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$_2$IrO$_4$. The results presented here provide insight into the effect of Ti doping on structure and magnetic phase in strong spin-orbit coupled Sr$_2$IrO$_4$.

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

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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$_2$IrO$_4$, believed to stabilize in $J_{\text{eff}} = \frac{1}{2}$ ground state and it is a spin orbital coupling driven Mott insulator. Sr$_2$IrO$_4$ having similarities with high T$_c$ cupric superconductor La$_2$CuO$_4$ draw interest, as it has also been theoretically predicted to be superconductor electrons/hole doping. In 5$d$ iridates active interactions such as: (CEF), (U) and (SOI), compete among themself setting new level of energies and indorse 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 $J_{\text{eff}} = \frac{3}{2}$ quartet and upper $J_{\text{eff}} = \frac{1}{2}$ doublet. The $J_{\text{eff}} = \frac{1}{2}$ 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$_2$IrO$_4$ crystallize in reduced tetragonal structure with space group I4$_1$/amd. The reduced symmetry is due to rotation of IrO$_6$ octahedral around c-axis and play important role in deciding the physical properties of Sr$_2$IrO$_4$. [1-7]

The magnetic ground state in Sr$_2$IrO$_4$ 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 perturb and destabilized by tuning the 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 introducing holes and reduces the SOC while increase U.

In this paper we investigate the evolution of structural and magnetic properties of Sr$_2$Ir$_{1-x}$Ti$_x$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 as effective magnetic moment increases. The isothermal magnetization shows decrease in coercively and remnant magnetization.

2. EXPERIMENTAL DETAIL

The polycrystalline samples of Sr$_2$Ir$_{1-x}$Ti$_x$O$_4$ with composition x = 0.0, 0.05 and 0.1 are prepared using slandered solid state reaction technique. The starting material Ir$_2$O$_3$ (Sigma Aldrich 99.99 %), TiO$_2$ (Sigma Aldrich 99.99 %), and SrCO$_3$ (Sigma Aldrich 99.95 %), are taken in stoichiometric composition and grinded well. The fine powder is then calcinated at 900 °C for one day. The calcinated 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 FullProof 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 Sr$_2$Ir$_{1-x}$Ti$_x$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 crystalizes 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 $8^\circ$ for $x=0.1$ around c axis.
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 $T_C \approx 225$ 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 $\chi$ is magnetic susceptibility, $C$ is the Curie constant and $\theta_p$ is the Curie temperature. We have calculated effective paramagnetic moment using fitting parameters for all samples $\mu_{\text{eff}} = 0.56 \mu_B/$f.u. for parent compound. With Ti doping $\mu_{\text{eff}}$ 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 $\mu_S = 0.05 \mu_B$/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 reminance 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 2. (a) ZFC and FC magnetization data measured in 10 kOe are plotted as a function of temperature for Sr$_2$Ir$_{1-x}$Ti$_x$O$_4$. (b) Inverse susceptibility ($\chi^{-1}$) shows Curie-Weiss behaviour.

Figure 3. (a) Isothermal magnetization is shown for Sr$_2$Ir$_{1-x}$Ti$_x$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 Sr$_2$Ir$_{1-x}$Ti$_x$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.

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