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Evolution of magnetic and transport properties in hole doped $Y_2Ir_2O_7$

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Abstract. We report the effect of Ru^{4+} substitution at Ir-site on structural, magnetic and electrical properties in pyrochlore iridate $Y_2Ir_2O_7$. Structural investigation has been done using x-ray powder diffraction and Rietveld analysis for both samples. There is no structural phase transition with Ru doping. Magnetization study shows that on slightly substitution of Ru at Ir-site effectively shifts the magnetic irreversible temperature T_{irr} towards low temperature and reduce the bifurcation between M_{ZFC} and M_{FC} branches. The electrical resistivity shows an insulating behavior in whole temperature range, however, resistivity decreases with doping of Ru.

1. Introduction

Pyrochlore iridates $A_2B_2O_7$ (A = rare earth elements) are very interesting showing exotic electronic and magnetic properties [1]. Within the extended $5d$ character, Ir exhibit reduced electronic correlation (U), however, the heavy nature of Ir gives prominent spin orbit coupling (SOC) effect. Moreover, the interpenetrating layers of IrO_6 octahedra and OA_4 tetrahedra introduce geometrical frustration. This frustration is seen to give rise many interesting phenomena such as, spin glass, spin ice, spin liquid, etc. The physical properties interestingly evolve with A-site element: from magnetic insulating to complex metallic phases [2]. When the A^{3+} ion is magnetic, there is a possibility of $f-d$ exchange interactions between Ir^{4+} and A^{3+} ions which naturally introduces complicated magnetic interactions in Ir-based pyrochlores [3]. The $Y_2Ir_2O_7$ is of special interest as the nonmagnetic Y^{3+} resides at A-site. In this sense, Ir^{4+} ion ($5d^5$) with single unpaired electron generate frustration in this material. The $Y_2Ir_2O_7$ shows an insulating behavior where the resistivity increases with decreasing temperature. $Y_2Ir_2O_7$ shows irreversibility in magnetization data around 160 K [4, 5, 6]. Given that frustration plays vital role in pyrochlore lattice the spin freezing to glassy state could also be a probable magnetic state in $Y_2Ir_2O_7$ at low temperature [4, 7, 8].

In this work, we have studied the effect of Ru^{4+} substitution in pyrochlore iridate $Y_2Ir_2O_7$. The $5d$ based Ir has strong SOC and less U, on other hand, $4d$ Ru has more U and less SOC. Thus, this substitution is expected to reduce the SOC and intensify the electron correlation which will result in change in ground state properties. Substitution of Ru^{4+} ($4d^4$) at Ir^{4+} ($5d^5$) site not only reduce the SOC but also introduce a hole in t_{2g} band, which reduce the level of Fermi surfaces E_F, thus, moving the system away from the insulating to metallic state. We have studied structural, magnetic and electrical transport properties. we find that magnetic moment, Curie temperature, frustration parameters (f) and magnetic irreversible temperature

T_{irr} decreases. Moreover, Resistivity data of $Y_2Ir_2O_7$ show insulating behavior throughout the temperature range, however, on Ru substitution, resistivity is decreased.

2. Experimental Procedure

Polycrystalline samples of $Y_2Ir_{2x}Ru_xO_7$ (x = 0.0 and 0.1) have been prepared using standard solid state method. The mixture of ingredient powder materials Y_2O_3 , RuO_2 and IrO_2 with phase purity 99.99% (Sigma-Aldrich) taken in stoichiometric ratio are ground well. Pellet is heated in air in temperature range 1000 ^{o}C to 1160 ^{o}C for 18 days with several intermediated grindings. The material is characterized by powder x-ray diffraction (XRD) measured with Rigaku-made Miniflex600 and PANalytical XPert powder diffractometer. The XRD pattern is refined with Rietveld program. DC Magnetization (χ) data have been collected using a vibrating sample magnetometer (PPMS, Quantum Design).

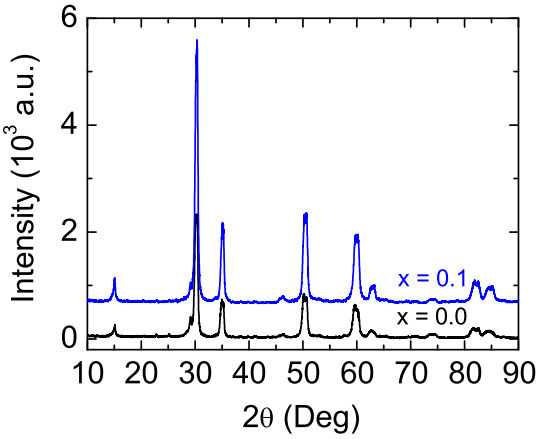

Figure 1. X-ray diffraction patterns of polycrystalline samples with compositions $Y_2Ir_{2x}Ru_xO_7$ series.

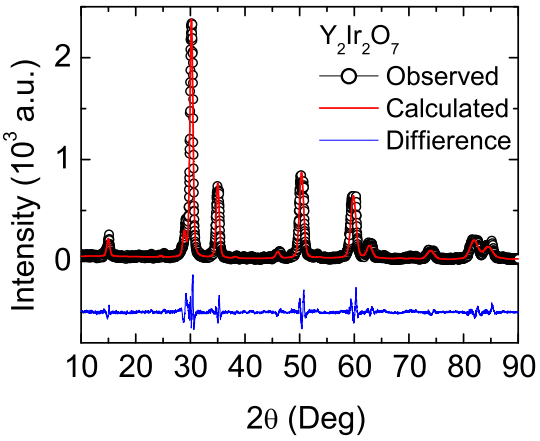

Figure 2. XRD pattern for $Y_2Ir_2O_7$ (x = 0) along with reitveld refinement.

3. Results and Discussion

3.1. structral properties

Fig. 1 shows XRD pattern of $Y_2Ir_{2x}Ru_xO_7$ with x = 0.0 and 0.1 at room temperature. The figure shows there is no major modification in terms of peak splitting and/or peak positions in XRD data with Ru doping. This imply that Ru doping does not induce structural phase transition. Fig. 2 shows XRD pattern of $Y_2Ir_2O_7$ along with Rietveld refinement. The Rietveld analysis of XRD data shows the sample crystallizes in cubic structure with Fd-3m space group. The material has been found in single phase; however, a small fraction of unreacted ingredient materials is observed which is very common in this material [4, 5, 6, 9]. Considering the diamagnetic nature of ingredients, the magnetic properties of $Y_2Ir_2O_7$ are minimally influenced. The lattice constant is found to be 10.2445 and 10.2397 $\AA$ for x = 0.0 and 0.1 respectively.

3.2. Magnetic Properties

Fig. 3 shows temperature dependent magnetic susceptibility of $Y_2Ir_{2x}Ru_xO_7$ measured in 10 kOe magnetic field (H) following zero field cooling (ZFC) and field cooling (FC) protocol. A bifurcation between ZFC and FC branches of magnetic susceptibility has been observed around T_{irr} = 160 K and 153 K for x = 0.0 and 0.1 respectively, which implies spins are ordered below this temperature. The bifurcation between M_{ZFC} and M_{FC} braches could arise due to

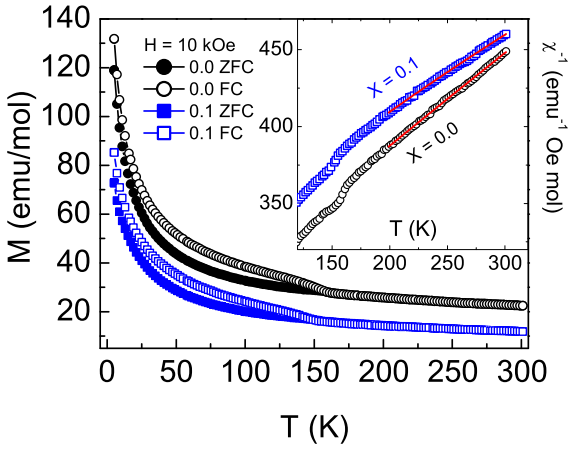

Figure 3. Temperature dependent dc magnetization from 5 K to 300 K at 10 kOe field (Magnetization data for x = 0.1 shifted downward by 10 (emu/mol) for clear understanding the data). Inset shows the modified CWL fitting of parent and doped sample. (discussed in text).

glassy behavior which is quite expected in pyrochlore system with geometrical frustration. The inset of Fig. 3 shows temperature dependence of inverse susceptibility (χ =M/H) which can be explained with modified Curie-Weiss law (CWL).

$$\chi = M/H = \chi_0 + C/(T - \theta_P) \tag{1}$$

Where χ_0 is the temperature independent susceptibility, C is the Curie constant and θ_P is the Curie temperature. The straight line fitting of χ^{-1} data gives Curie temperature θ_P = -386 K and -335 K for x = 0.0 and 0.1 respectively, which is indicative of non-ferromagnetic (FM) behavior. The relevant frustration parameters (f) which is defined as $|\theta_p|/T_{irr}$, has been calculated to be 2.4 and 2.1 for x = 0.0 and 0.1 respectively, which suggest that frustration level is decreased with Ru-substitution.

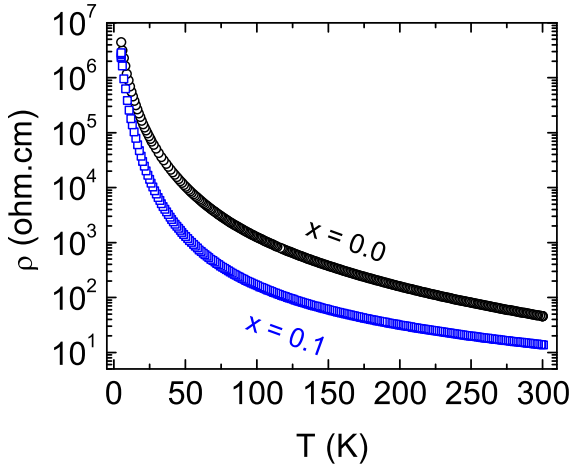

Figure 4. The temperature dependence electrical resistivity from 5 K to 300 K.

3.3. Transport properties

Fig. 4 shows the temperature dependent electrical resistivity for $Y_2Ir_{2x}Ru_xO_7$ (x = 0.0 and 0.1). The ρ (T) shows an insulating behavior for all composition which is intrinsic to most of the iridium based oxides due opening of an insulating gap [10] i.e., the resistivity increases with

decreasing temperature. At low temperature below 40 K, a sudden rise in resistivity of $Y_2Ir_2O_7$ is observed where it increases by about five orders. We observed that resistivity decreases with Ru doping. However, decrease in resistivity in $Y_2Ir_{2x}Ru_xO_7$ can be attributed to tuning of SOC, U with Ru^{4+} ($4d^4$) substitution. Moreover, Ru electrons are itinerant in character can result in increase conduction. However, detail investigation will be done in future with more doping concentration to understand this.

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