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Nonequilibrium low temperature phase in pyrochlore iridate $Y_2Ir_2O_7$: Possibility of glass-like dynamics

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ABSTRACT

Geometrical frustration and spin–orbit coupling effect together play vital role to influence properties in pyrochlore based iridium oxides. Here we have investigated detailed structural, magnetic, thermodynamic and transport properties of pyrochlore iridate $Y_2Ir_2O_7$. Magnetization data show onset of magnetic irreversibility around temperature $T_{irr} \sim 160$ K, however, no sign of long-range type ferromagnetic ordering is observed below T_{irr}. Specific heat data show no visible anomaly across T_{irr}, and the analysis of data indicate sizable density of states across Fermi level. Temperature dependent x-ray diffraction measurements show no change in structural symmetry down to low temperature. The material, on the other hand, shows significant relaxation and aging behavior similar to glassy dynamics. The electronic charge transport in this highly insulating system is found to follow power law dependence with temperature. The material shows negative magnetoresistance which is explained with quantum interference effect.

1. Introduction

Pyrochlores with chemical formula $A_2B_2O_7$ (A and B generally stands for rare earth and transition metal ions, respectively) have attracted surge of scientific interest over last one or two decades [1,2]. These materials have in-built frustration arising from structural geometry where the A and B cations form corner-shared tetrahedra. As a result, many interesting physical phenomena are observed. Most notable examples are spin glass [3,4], spin ice [5,6], spin liquid [7,8], anomalous Hall effect [9,10], superconductivity [11], Kondo-like behavior [10], etc.

In recent times, Ir-based pyrochlores ($A_2Ir_2O_7$) have been at center place for investigation. In addition to its geometric frustration, these materials exhibit sizable spin–orbit coupling (SOC) effect due to the presence of heavy Ir atoms. However, the electron–electron correlation effect (U), which is usually prominent in transition metal oxides, turns out relatively small in these systems due to extended character of 5*d* orbitals of Ir. Nonetheless, the SOC and electronic correlation effect exhibit comparable energy scale, therefore, these Ir-based oxides systems offer an ideal playground where the interesting physics coming out as an interplay between these two effects can be studied with control manner. The theoretical calculation, indeed, has predicted various exotic topological insulating phases in pyrochlore iridates based on this intriguing interplay between SOC and electronic correlation effect [12]. Moreover, recent study has shown that physical properties in pyrochlore iridates $A_2Ir_2O_7$ largely depend on ionic radius of A^{3+} where the material gradually shifts from its insulating/magnetic behavior to metallic/nonmagnetic one with increasing ionic radius [13].

Within the family of pyrochlore iridates, $Y_2Ir_2O_7$ has special interest as it has nonmagnetic Y^{3+} residing at A-site therefore, the magnetic properties are mostly determined by the contribution from Ir sublattice. In addition, one can exclude the possibility of *f*–*d* exchange interactions which has been discussed to introduce complicated magnetic interactions in Ir-based pyrochlores [14]. This is also important considering obvious interconnection between magnetic and electronic properties in these materials. This shows that $Y_2Ir_2O_7$ is an ideal system to study the intercoupling effect of SOC and U while neglecting other associated contributions. This material by nature is insulator where the resistivity increases by couple of orders at low temperature [16]. The magnetic state is, however, still debated. While the neutron diffraction/scattering measurements reveal no sign of magnetic long-range ordering [15], the muon spin rotation/relaxation measurement [17], on the other hand, has shown well-defined spontaneous oscillations in muon symmetry indicating long-range ordering. Considering the in-built frustration in pyrochlore lattice, the glass-like behavior could also be a low temperature magnetic state in $Y_2Ir_2O_7$. Note that Y-based other pyrochlores, i.e., $Y_2Mo_2O_7$ and $Y_2Ru_2O_7$, have shown spin-glass behavior [3,4]. This underlines the fact that there is need for conclusive understanding of magnetic

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state in $Y_2Ir_2O_7$ which is further amplified with the fact that recent calculation proposes topological Weyl semimetallic phase in $Y_2Ir_2O_7$ based on its ground state magnetic character [19].

In this work, we present detailed structural, magnetic, thermodynamic and transport properties of pyrochlore iridate $Y_2Ir_2O_7$. Magnetic measurements show onset of magnetic irreversibility at temperature T_{irr}~160 K, however, no sharp peak/cusp is observed at this temperature. Analysis of magnetization data shows no spontaneous moment at low temperature. Instead, reasonable magnetic relaxation and aging behavior similar to metamagnetic systems such as spin-glasses and superparamagnet has been observed. Temperature dependent x-ray diffraction (XRD) measurements show that there is no crystallographic phase/symmetry transition down to low temperature. The material is found to be insulating throughout the temperature range where the resistivity follows power law behavior with temperature. The negative magnetoresistance at low temperature can be explained with quantum interference effect.

2. Experimental details

Polycrystalline samples of $Y_2Ir_2O_7$ have been prepared using standard solid state method. The ingredient powder materials Y_2O_3 and IrO_2 with phase purity >99.99% (Sigma-Aldrich) are taken in stoichiometric ratio. The mixture are ground well, subsequently pelletized and heated in air at 1000 °C for 96 h, at 1100 °C for 96 h and at 1160 °C for 252 h with intermediate grinding. The heating and cooling rate has been used 3 °C/min. The material has been characterized by powder x-ray diffraction (XRD) using a Rigaku made diffractometer (model: Miniflex600) with CuK_α radiation at room temperature. The XRD data have also been collected at different temperatures in the range of 300–20 K using a PANalytical X'Pert powder diffractomer. The low temperature is achieved using a helium close cycle refrigerator (CCR) based cold head where proper temperature stabilization is ensured by waiting sufficiently before collecting data. Data have been collected in the range of $2\theta = 10$–$90°$ with a step of $\Delta 2\theta = 0.033°$ and scan rate of 2°/min. The collected XRD data have been analyzed using Reitveld refinement program (FULLPROF) by Young et al. [20]. The Rietveld analysis shows that the material is in single phase except the presence of small fraction of unreacted ingredient materials. It can be noted that after certain heat treatment, no further improvement in terms of unreacted ingredients is observed, rather they are found to grow. Magnetically, Y_2O_3 is diamagnetic and IrO_2 is paramagnetic therefore these residual impurities are expected to influence the magnetic properties of desired material minimally. Note that similar impurity phases have also been observed in previous studies [15,17,21].

In general, pyrochlore materials adopt cubic crystallographic phase with $Fd\bar{3}m$ symmetry [1]. There are two sets of oxygen atoms, therefore pyrochlore structures consist of four nonequivalent crystallographic positions (Table 1). The resulting composition can be written as $Y_2Ir_2O_6O'$. The Ir^{4+} cations are six coordinated and they form corner shared IrO_6 octahedra with equal length (d_{Ir-O}) of Ir–O bonds. The eight coordinated (six O and two O′) Y^{3+} cations form distorted cubes where Y–O bonds are equal in length and larger than the Y–O′ ones. The only variable position in pyrochlore structures is x position with O (Table 1) which determines its structural stability. For instance, $x=0.3125$ gives perfect IrO_6 octahedra while $x=0.375$ results in perfect cubes around Y. The pyrochlore structure that can be viewed as interpenetrating two sub-lattices consists of each type of tetrahedra, i.e., Y_4O' and $Ir_4\diamond$, where $\diamond$ implies empty center-site (8a site). As these tetrahedra's are corner shared the magnetic atoms sitting at vertices of tetrahedra induce frustration. The Rietveld analysis of our XRD data confirms that $Y_2Ir_2O_7$ crystallizes in cubic phase with $Fd\bar{3}m$ symmetry (Fig. 1). The unit cell lattice parameter is found to be $a=10.2445$ Å. The bond lengths (d) and angles ($\langle\rangle$) are determined from structural parameters as given in Table 2. In IrO_6 octahedra, bond angles ⟨O–Ir–O⟩ in basal plane are not equal while opposite ones are same in value. The bond lengths and bond angles for present materials (Table 2) show good agreement with the reported values [15]. DC magnetization data have been collected

Table 1
Unit cell parameters and crystallographic positions determined from the Rietveld profile refinement of the powder XRD patterns for $Y_2Ir_2O_7$ at 300 and 20 K. Here O and O′ refers to two different oxygen position (discussed in text).

Parameters	300 K $Fd\bar{3}m$	20 K $Fd\bar{3}m$
a (Å)	10.237(2)	10.219(2)
V (Å^3)	1072.7(3)	1067.2(3)
Y site	16d	16d
x	0.5	0.5
y	0.5	0.5
z	0.5	0.5
Ir site	16c	16c
x	0.0	0.0
y	0.0	0.0
z	0.0	0.0
O site	48f	48f
x	0.336(3)	0.332(2)
y	0.125	0.125
z	0.125	0.125
O′ site	8b	8b
x	0.375	0.375
y	0.375	0.375
z	0.375	0.375

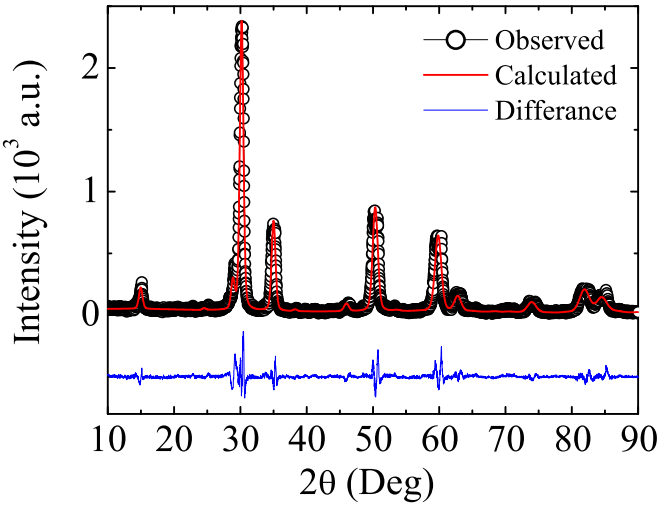

Fig. 1. XRD plot for $Y_2Ir_2O_7$ is shown at room temperature along with Rietveld analysis.

Table 2
Bond lengths and bond angles determined using structural parameters for $Y_2Ir_2O_7$ at 300 and 20 K. Here O and O′ refers to two different oxygen position (discussed in text).

Parameters	300 K $Fd\bar{3}m$	20 K $Fd\bar{3}m$
d_{Ir-O} (Å)	2.0169(2)	1.9914(3)
d_{Y-O} (Å)	2.4615(2)	2.4921(1)
$d_{Y-O'}$ (Å)	2.2164(2)	2.2125(2)
⟨O–Ir–O⟩ (deg)	80.9(1)	82.6(1)
⟨O–Ir–O⟩ (deg)	99.1(1)	97.4(1)