Temperature evolution of magnetic and transport behavior in 5d Mott insulator Sr$_2$IrO$_4$: significance of magneto-structural coupling

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Abstract

We have investigated the temperature evolution of magnetism and its interrelation with structural parameters in the perovskite-based layered compound Sr$_2$IrO$_4$, which is believed to be a $J_{\text{eff}} = 1/2$ Mott insulator. The structural distortion plays an important role in this material and induces a weak ferromagnetism in an otherwise antiferromagnetically ordered magnetic state with a transition temperature around 240 K. Interestingly, at low temperatures, below around 100 K, a change in the magnetic moment has been observed. Temperature dependent x-ray diffraction measurements show that sudden changes in structural parameters around 100 K are responsible for this. Resistivity measurements show insulating behavior throughout the temperature range across the magnetic phase transition. The electronic transport can be described with Mott’s two-dimensional variable range hopping (VRH) mechanism, however, three different temperature ranges are found for VRH, which is a result of varying the localization length with temperature. A negative magnetoresistance (MR) has been observed at all temperatures in contrast to positive behavior generally observed in strongly spin-orbit coupled materials. The quadratic field dependence of MR implies the relevance of a quantum interference effect.

Keywords: 5d transition metal oxides, x-ray diffraction, magnetism, canted antiferromagnet, hopping transport, quantum interference

(Some figures may appear in colour only in the online journal)

1. Introduction

The Ir-based 5d transition metal oxides (TMOs) have been the topic of intense research in recent times due to their unusual properties [1–12]. With increasing $d$ character in 5d TMOs, the orbitals are spatially extended which substantially reduces the electronic correlation effect ($U$). On the other hand, extended orbitals contribute to enhanced bonding interactions and orbital dependent physical phenomena such as, orbital ordering, unusual magnetic properties, etc. Apart from these, the spin orbit coupling (SOC) effect becomes very prominent in these materials due to the high value of the atomic number of 5d transition metals. This SOC effect is an additional key factor in governing their detailed physical properties. With this reduced $U$, the 5d TMOs are expected to be more metallic than their 3d and 4d counterparts. Surprisingly, however, they happen to be strong insulators. It is commonly believed that the strong SOC in iridium oxides causes splitting of the $t_{2g}$ band into $J_{\text{eff}} = 1/2$ and $J_{\text{eff}} = 3/2$ multiples [1, 2]. Since Ir$^{4+}$ with five $d$ electrons adopts a low spin state, four of which fill the lower $J_{\text{eff}} = 3/2$ while one partially fills the upper $J_{\text{eff}} = 1/2$. The presence of $U$, though small,
further splits the narrow $J_{\text{eff}} = 1/2$ band opening a Mott type gap which renders an insulating behavior. Nonetheless, the delicate competition among the various interactions such as the crystal field effect (CEF), SOC and $U$ plays a crucial role in realizing the $J_{\text{eff}} = 1/2$ intriguing physics in these materials.

The perovskite-based layered compound Sr$_2$IrO$_4$ with a celebrated K$_2$NiF$_4$ structure is an interesting member of the 5$d$ TMOs. The Sr$_2$IrO$_4$ crystallizes in a tetragonal structure with a $I4_1/acd$ space group where the IrO$_6$ octahedra exhibit a rotation of about $11^\circ$ around the c axis (see figure 1) [13]. The crystallographic structure creates an important interest in this material. For instance, the isostructural nature of Sr$_2$IrO$_4$ with the 3$d$- and 4$d$-based superconducting materials La$_2$CuO$_4$ and Sr$_2$RuO$_4$, respectively has induced a special interest in search of superconductivity in this material [10, 11]. Moreover, the octahedral distortion (rotation) is believed to bring in a Dzyaloshinsky–Moriya (DM) type antisymmetric exchange interaction which leads to a transition to a weak ferromagnetic (FM) phase with a transition temperature ($T_c$) $\sim 240$ K. This material otherwise would show a collinear type antiferromagnetic (AFM) ordering [13, 14]. However, at low temperature ($T$) below $\sim 100$ K, a change in magnetization ($M$) is observed [15–17], which it appears is not due to the magnetic phase transition. In the present scenario of canted AFM spin ordering the change in the moment could be related to the spin ordering of Ir ions. In fact, a recent theoretical investigation has put forward an interrelation between the spin canting angle and the structural parameters in Sr$_2$IrO$_4$ [9]. These observations emphasize a temperature dependent structural analysis to comprehend the temperature evolution magnetic behavior. It should be mentioned that temperature dependent structural studies employing x-ray and neutron diffraction have been performed for Sr$_2$IrO$_4$, however, detailed analysis of the data as well as finding a correlation between the structural and magnetic behavior has not been attempted [13, 18]. Moreover, given an exotic insulating phase in the present material, the electronic transport mechanism needs to be investigated thoroughly including when in the presence of a magnetic field ($H$).

We have investigated the interrelation between temperature dependent magnetic behavior and the structural parameters by means of temperature dependent x-ray diffraction (XRD) measurements. The measured magnetization data show a transition to a weak ferromagnetic phase around 238 K, however, at low temperatures below around 95 K we find $M(T)$ shows a sharp fall with lowering the temperature. The analysis of temperature dependent XRD data shows that a sudden change in structural parameters around this temperature is responsible for this behavior. Resistivity ($\rho$) data show an insulating behavior where it rises drastically below 50 K. The mode of electronic transport has been understood to follow Mott’s variable range hopping mechanism at three different temperature ranges. The quadratic nature of the field dependence of magnetoresistance (MR) at low temperature implies the significance of the quantum interference effect in this material.

2. Experimental details

A polycrystalline sample of Sr$_2$IrO$_4$ has been prepared using the standard solid state ceramic method. The ingredient powder materials SrCO$_3$ and IrO$_2$ with phase purity greater than 99.99% (Sigma-Aldrich) are taken in stoichiometric ratio and ground well. The fine powders are calcined in air at 900°C for 24 h with a heating and cooling rate of 3°C min$^{-1}$. The calcinated powders are then palletized and sintered at 1000°C and 1100°C for 24 h at the same heating and cooling rates with intermediate grinding. The phase purity of the sample is checked using powder x-ray diffraction (XRD) with a Rigaku MiniFlex diffractometer with CuK$_{\alpha}$ radiation.
at room temperature. The reitveld analysis of the XRD data shows the sample is in the single phase without any chemical impurity and crystallizes in the tetragonal phase with 14I/acd symmetry. The temperature dependent XRD measurements were performed using a PANalytical X’Pert powder diffractometer in the temperature range of 298–20 K. A helium closed cycle refrigerator (CCR) based cold head is used to achieve the low temperature. Care has been taken for proper temperature stabilization by allowing sufficient wait time before collecting the data. The data was collected in the 2θ range of 10–90° at a step of Δ2θ = 0.033° and a scan rate of 2° min⁻¹. The XRD data were analyzed using the Reitveld refinement program (FULLPROF) by Young et al [19]. DC magnetization data was collected using a vibrating sample magnetometer (PPMS, Quantum Design) and the electrical transport properties were measured using a home-built insert fitted with an Oxford superconducting magnet.

3. Results and discussions

3.1. Magnetization study

Figure 2 shows the temperature dependent magnetization data for Sr₂IrO₄ measured in an applied magnetic field of 10 kOe following the zero field cooling (ZFC) and field cooled (FC) protocols. When the temperature decreases, $M(T)$ shows a sharp increase around 240 K which is marked by a paramagnetic (PM) to FM transition in this material as has previously been observed [13]. We find the FM transition temperature $T_c \approx 233$ K which is estimated from the inflection point in the d$M$/dT plot. The obtained $T_c$ is in agreement with the reported studies of this material [13, 20, 21]. On further lowering the temperature there is a bifurcation between the ZFC and FC branches of the magnetization data. A close observation reveals that bifurcation starts around 150 K. It is, however, interesting to notice that while $M_{ZFC}$ monotonically increases with a small slope, the $M_{ZFC}$ shows a steep decrease below 95 K ($T_d$) which continues down to the lowest measuring temperature. This fall in moment below around 100 K has also been observed in the case of both single and polycrystalline samples [15–17] and may be intrinsic to this material. There may be many underlying mechanisms behind this behavior, yet the most obvious one could be that the structural phase/parameters change around this temperature. In fact, a theoretical calculation has predicted an interrelation between the rotation of IrO₆ octahedra and the spin canting angle [9]. This implies that a detailed structural analysis with temperature is required to understand the low-$T$ magnetic behavior.

The inset of figure 2 shows an inverse susceptibility ($\chi^{-1}$), deduced from the dc magnetization data $(M_{ZFC}/H)^{-1}$, as a function of temperature. Above $T_c$, the $\chi^{-1}(T)$ shows a linear behavior over a limited temperature range up to around 270 K following a Curie–Weiss behavior $\chi = M/H = C/(T - \theta_p)$, where $C$ and $\theta_p$ are the Curie constant and Curie temperature, respectively. From the straight line fitting (the inset of figure 2) we estimate $\theta_p = 233.02$ K. The value of $\theta_p$ is not only very close to the estimated $T_c$ of this material, it also suggests that the spin ordering is FM in nature. Moreover, using the fitted parameters, we have calculated the effective PM moment ($\mu_{eff}$) in terms of the Bohr magneton per formula unit as 0.56 $\mu_B$ f.u⁻¹. The estimated $\mu_{eff}$ agrees with the other studies [20, 21] and also the value matches with the expected one ($\mu_{eff} = g_J \sqrt{J_{zz}(J_{zz} + 1)}$) for this material with a single unpaired 5d electron residing in the $J_{zz} = 1/2$ state. With $g_J = 2/3$, the expected value of $\mu_{eff}$ comes as 0.57 $\mu_B$ f.u⁻¹.

To understand the magnetic field induced magnetic behavior in Sr₂IrO₄ with canted AFM-type spin ordering, we have recorded a magnetic hysteresis loop $M(H)$ at 5 K (figure 3(a)) with a field range of ±70 kOe. At the highest applied field of 70 kOe, the magnetization does not saturate, but instead increases monotonically yielding a moment $\mu_H = 0.05 \mu_B$ f.u⁻¹. The measured $\mu_H$ is comparable with other studies of this material [13, 15, 20–22]. This measured $\mu_H$ is significantly low compared to the expected value ($\mu_{eff} = g_J J_{zz}(\mu_{eff} \mu_B)$) for this SOC dominated 5d oxide material Sr₂IrO₄ which can be calculated as 1.3 $\mu_B$ f.u⁻¹. This disagreement between the measured and expected values of $\mu_H$ is not very clearly understood. However, an uncompensated antiferromagnetic type of spin ordering in the present material would likely explain such a low moment and the unsaturated $M(H)$ hysteresis loop. Given that there is a strong coupling between structural and magnetic ordering in this material a structural investigation in the presence of the magnetic field and/or an investigation introducing chemical doping would shed some light on understanding this unusual behavior. On the other hand, this reduced moment has been explained from an itinerant based band FM model using the Rhodes–Wohlfarth plot [24]: with the ratio, $\mu_{eff}/\mu_H > 1$ the low moment in Sr₂IrO₄ has been ascribed due to the itinerant FM behavior [20]. However, a very high field measurement of the moment is required to check the saturation moment before assigning it to band ferromagnetism. Note that a reasonably...
Figure 3. (a) Magnetic field dependent magnetization is shown for Sr$_2$IrO$_4$ at 5 K. (b) An Arrott plot ($M^2$ versus $H/M$) of magnetization data is shown at 5 K.

high value of remnant magnetization $M_{rem} = 0.024 \mu_B$ f.u$^{-1}$.

To examine the nature of ground state magnetism we have plotted the virgin plot of $M(H)$ in the form of an Arrott plot [23]: $M^2$ versus $H/M$. The Arrott plot has been successfully used to understand the magnetic state as the intercept on the positive $M^2$ axis due to an extrapolation of high field data in the Arrott plot suggests the presence of spontaneous magnetization ($M_s$) in the system. Figure 3(b) shows such an Arrott plot related to 5 K $M(H)$ plot in figure 3(a), where one can see a positive intercept on the $M^2$ axis implying a FM ordering. From the intercept the $M_s$ has been calculated to be $6.85 \times 10^{-3} \mu_B$ f.u$^{-1}$ which indicates the ground state of Sr$_2$IrO$_4$ is of a weak FM nature.

3.2. Temperature dependent XRD studies

Figure 4. The XRD pattern collected at 298 K for Sr$_2$IrO$_4$ has been shown along with the Rietveld refinement. The refinement shows the material crystallizes in the tetragonal structure with $I4_1/acd$ symmetry.

detailed temperature dependent structural studies by means of XRD measurements. The data have been collected at 298, 250, 230, 200, 150, 100, 80, 60, 40 and 20 K encompassing the PM, FM and low temperature magnetic state below $T_M$. Figure 4 shows the XRD pattern at 298 K along with Rietveld refinement fitting which suggests a good quality of sample without any chemically impure phase. The refinement result suggests that the sample crystallizes in the tetragonal structure with $I4_1/acd$ symmetry [13]. The details of the structural parameters and crystallographic positions obtained from Rietveld analysis of the XRD data at 298 K have been summarized in table 1. The ratio $R_{wp}/R_{exp}$, which is a known goodness of fit (GOF), is found to be around 1.38. These $R$-factors are mathematically constructed in a Rietveld refinement program which employs the least square approach to refine the calculated diffraction pattern based on the structural model and tries to match with the observed diffraction pattern [19]. The $R_{wp}$ is called the ‘weighted pattern $R$-factor’ and $R_{wp}^2$ represents the ratio between the summation of the weighted squared difference between the calculated and the observed value and the summation of the weighted squared observed value. On the other hand, $R_{exp}$ is called the ‘expected $R$-factor’ and represents the ‘best possible $R_{wp}$’ where the calculated and observed patterns have a perfect match. A value of the ratio $R_{wp}/R_{exp}$ around 1.3–1.4 or less is considered to be a reasonably good fitting while the large and small values imply inaccuracy of the applied model and/or erroneous statistical counting. In this sense, our obtained $R_{wp}/R_{exp}$ ratio is suggestive of a reasonably good fitting. The Ir–O–Ir bond lengths and bond angles, calculated using the unit cell parameters and crystallographic positions have been shown in table 2. The IrO$_6$ octahedra have two distinct oxygen positions [two apical (O1) and four basal (O2)] where they are found to be slightly apically elongated (table 2). The $<\text{Ir–O1–Ir}>$ is found to be 180$^\circ$ showing they are not distorted along the c axis. The refinement, however, shows IrO$_6$ octahedra are rotated around the c axis by an angle...
also shows two vertical dotted lines representing unit cell volume (Figures 6).

Table 1. Structural parameters and crystallographic positions determined from the Rietveld profile refinement of the powder XRD patterns for Sr$_2$IrO$_4$ at 298 K. Here O1 refers to the apical oxygen, O2 refers to the basal oxygen which lies in the plane of the perovskite layer.

| Parameters | 298 K |
|------------|-------|
| $a$ (Å) | 5.4980(2) |
| $c$ (Å) | 25.779(1) |
| $V$ (Å$^3$) | 779.27(6) |
| Sr site | 16d |
| $x$ | 0.0 |
| $y$ | 0.0 |
| $z$ | 0.17506(2) |
| Ir site | 8a |
| $x$ | 0.0 |
| $y$ | 0.0 |
| $z$ | 0.0 |
| O1 site | 16d |
| $x$ | 0.0 |
| $y$ | 0.0 |
| $z$ | 0.0 |
| O2 site | 16f |
| $x$ | 0.20021(5) |
| $y$ | 0.20021(5) |
| $z$ | 0.25000 |
| $R_w$ | 23.6 |
| $R_{wp}$ | 17.0 |

Table 2. Ir–O–Ir bond length ($d$) in Å, bond angle ($<$) in Deg are given for Sr$_2$IrO$_4$ at 298 K. Here O1 refers to the apical oxygen, O2 refers to the basal oxygen which lies in the plane of the perovskite layer.

| Parameters | 298 K |
|------------|-------|
| $d_{Ir-O1}$ (Å) | 2.0610(4) |
| $d_{Ir-O2}$ (Å) | 1.9820(3) |
| $<\text{Ir-O1-Ir}>$ (Deg) | 180 |
| $<\text{Ir-O2-Ir}>$ (Deg) | 157.47 |
| $\theta_{Oct}$ (Deg) | 11.26 |

c increases, $c/a$ increases and $V$ decreases with decreasing temperature. The increase of parameter $c/a$ implies the tetragonal structure of this compound is further elongated along the $c$ axis. However, the temperature evolution of the parameters is not monotonous. With decreasing temperature, all the parameters exhibit a change in slope at the onset of the FM phase transition i.e. around $T_c$. Within the FM phase, although the parameters change with temperature, their rate of change is almost maintained. Interestingly, around $T_M$ all the parameters show a sudden drastic change and with a further decrease in temperature the parameters change with a slower rate toward saturation. We do not, however, find any structural phase change cooling across $T_c$ and $T_M$ (figure 5). These changes of the structural parameters around $T_c$ and $T_M$ are quite intriguing.

To further elucidate this issue we have looked into the temperature evolution of the bond angle ($<$) and the bond length ($d$) of IrO$_6$ octahedra. Figures 7(a)–(d) show that with decreasing temperature the basal bond angle $<\text{Ir-O2-Ir}>$ increases, the rotation angle ($\theta_{Oct}$) of IrO$_6$ octahedra around the $c$ axis decreases, the basal bond length $d_{Ir-O2}$ decreases and the apical bond length $d_{Ir-O1}$ increases. It is observed that similar to the unit cell parameters in figure 6, the bond angle and length also exhibit a change in slope around $T_c$ and $T_M$. While there is no structural phase transition across the whole temperature range, the changes in slope in the structural parameters in figures 6 and 7 are intriguing. This structural behavior in figures 6 and 7 could be related to temperature evolution of the magnetization ($M_{ZFC}$) behavior in figure 2. It is a commonly accepted fact that magnetism in this material is governed by structural distortion i.e. cooperative rotation of corner shared IrO$_6$ octahedra [2, 13]. The Sr$_2$IrO$_4$, which is isostructural with La$_2$CuO$_4$, is a $J_{eff} = 1/2$ Mott insulator with a single unpaired electron in a spin orbit coupled $J_{eff} = 1/2$ state. The ground state of Sr$_2$IrO$_4$ is expected to be a Heisenberg like a magnetic state with AFM spin ordering [9]. However, a weak ferromagnetic state is established with an onset temperature $T_c \sim 238$ K (figure 2), which is believed
to be a result of canted antiferromagnetic ordering induced by a DM type antisymmetric exchange interaction [13]. The DM interaction mainly arises in magnetic materials without inversion symmetry and favors a spin canting in an otherwise AFM system resulting in weak ferromagnetism. The distortion of the IrO$_6$ octahedra in Sr$_2$IrO$_4$ breaks the required inversion symmetry of the Ir–O$_2$–Ir network. A dominant SOC effect ($\sim 0.4$ eV) in Sr$_2$IrO$_4$ also contributes to strengthening the DM interaction in this compound.

The theoretical calculation [9] has, in fact, predicted an intrinsic connection between the structural parameters and the magnetic ordering. The study shows the spin canting angle ($\phi$) and the octahedral distortion angle ($\theta_{\text{Oct}}$) are somehow connected and their relation depends on the lattice parameters. For $c < a$, the angle $\phi$ is more than the angle $\theta_{\text{Oct}}$, however, $\phi$ decreases with an increasing $c/a$ ratio, being equal for a cubic structure. The decreasing trend of the spin canting angle $\phi$, implies more moment cancellation and hence a reduced FM component of the moment. The temperature evolution of $c/a$ for Sr$_2$IrO$_4$ in figure 6(c) shows a drastic increase in $c/a$ across $T_M$. Similar behavior is also evident in figure 7(b) where the octahedra rotation angle $\theta_{\text{Oct}}(T)$ exhibits a decrease with decreasing temperature. These observations are suggestive of a weakening of the DM interaction and explain fairly the evolution of magnetization at low temperature where $M_{\text{ZFC}}$ shows a downfall below $T_M$ (figure 2).

3.3. Electronic transport study

To understand the interesting insulating phase in Sr$_2$IrO$_4$, we measured resistivity ($\rho$) as a function of both temperature
as well as magnetic field. Figure 8(a) shows that at room temperature resistivity is very low with value around 1 Ω cm. With decreasing temperature the ρ(T) increases exhibiting an insulating behavior [15, 21], however, the increase is very drastic at low temperature where resistivity increases by about five orders. We have also measured ρ(T) in presence of applied field of 80 kOe. In presence of magnetic field the qualitative feature of ρ(T) does not change but there is small decrease in ρ where the effect is prominent at lower temperatures (discussed later). This thermally activated conduction behavior has been described using Mott’s 2-dimensional (D) variable range hopping (VRH) model [25]:

\[
\rho = \rho_0 \exp \left( \frac{T_0}{T} \right)^{1/3}
\]

where \( T_0 \) is the characteristic temperature and can be expressed as:

\[
T_0 = \frac{21.2}{k_B N(E_F) \xi^3}
\]

where \( k_B \) is the Boltzmann constant, \( N(E_F) \) is the density of states (DOS) at Fermi surface and \( \xi \) is the localization length. Figure 8(b) shows straight line fitting of \( \rho(T) \) data following equation (1). Interestingly, our data can be fitted in three different temperature ranges and the parameter \( T_0 \) has been estimated from the respective fittings. Now taking the electronic coefficient of specific heat \( \gamma = 2 \text{mJ K}^{-2} \text{mole}^{-1} \) [21, 26], we have calculated \( N(E_F) \), \( \gamma = \pi^2 k_B^2 V_m N(E_F)/3 \), where \( V_m \) is the molar volume of Ir. The \( N(E_F) \) comes out very low (6 \( \times \) 10^28 eV\(^{-1}\) m\(^{-3}\)) which can be assumed constant across whole temperature as it shows throughout insulating behavior. The so obtained \( N(E_F) \) has been used to calculate \( \xi \) using equation (2). The values are given in table 3. The obtained \( T_0 \) compares well with other insulating oxide materials [21, 27]. The \( \xi \), which is of the order of in-plane lattice constant \( a \) (table 1), increases in lower temperature range where \( \rho \) shows higher values which looks counterintuitive, yet, considering lower thermal energy this increase in resistivity is justified. Moreover, figure 8 and table 3 show that temperature range for VRH changes around 240 K (\( \sim T_c \)) and 70 K (\( < T_M \)), though at low temperature the range is extended up to 40 K below which \( \rho(T) \) exhibits steep increase. These behavior is somehow related with both magnetization and structural parameters as they exhibit sudden changes around \( T_c \) and \( T_M \) (see figures 2, 6 and 7). Given that 2D charge transport in this layered tetragonal material the sudden changes of slope in lattice parameter \( a \) and basal bond angle \( \angle \text{Ir–O2–Ir} \) around \( T_c \) and \( T_M \) would imply a certain modification in charge hopping which is realized through change in parameter \( \xi \) (table 3). It can be noted that the apical bond angle \( \angle \text{Ir–O1–Ir} \) does not go through a change retaining value at 180°. Nonetheless, it is important that the three independent measurements i.e. magnetization, structural and resistivity show reasonable agreement which rather authenticates our study.

To understand the electron transport behavior in further detail, we have measured isothermal resistivity as a function of magnetic field up to field of 80 kOe. Figure 9 shows normalized magnetoresistance (MR) expressed as, \( \Delta \rho / \rho(0) = [\rho(H) - \rho(0)] / \rho(0) \) for Sr\(_2\)IrO\(_4\) at few representative temperatures. As evident in figure, the calculated MR is not very high and at all temperatures its value is negative i.e. resistance decreases in presence of magnetic field in agreement with previous study [15]. However, the characteristic nature as well as magnitude of MR changes with temperature. For instance, with lowering in temperature the magnitude of MR increases, reaching its value about 4.6% at 5 K and in magnetic field of 80 kOe. At higher temperatures the MR shows sharp fall in low field regime and then decreases very slowly. In

Table 3. Temperature range, fitting parameter \( T_0 \) (equation (1)) and localization length \( \xi \) calculated using equation (2) are given for Sr\(_2\)IrO\(_4\).

| Temperature range (K) | \( T_0 \) (K) | \( \xi \) (Å) |
|-----------------------|--------------|--------------|
| 300–240               | 1.44 \( \times \) 10^4 | 3.04 |
| 240–70                | 4.68 \( \times \) 10^4 | 4.42 |
| 40–5                  | 4.82 \( \times \) 10^3 | 9.44 |

\( k_B \) is the Boltzmann constant, \( N(E_F) \) is the density of states (DOS) at Fermi surface and \( \xi \) is the localization length. Figure 8(b) shows straight line fitting of \( \rho(T) \) data following equation (1). Interestingly, our data can be fitted in three different temperature ranges and the parameter \( T_0 \) has been estimated from the respective fittings. Now taking the electronic coefficient of specific heat \( \gamma = 2 \text{mJ K}^{-2} \text{mole}^{-1} \) [21, 26], we have calculated \( N(E_F) \), \( \gamma = \pi^2 k_B^2 V_m N(E_F)/3 \), where \( V_m \) is the molar volume of Ir. The \( N(E_F) \) comes out very low (6 \( \times \) 10^28 eV\(^{-1}\) m\(^{-3}\)) which can be assumed constant across whole temperature as it shows throughout insulating behavior. The so obtained \( N(E_F) \) has been used to calculate \( \xi \) using equation (2). The values are given in table 3. The obtained \( T_0 \) compares well with other insulating oxide materials [21, 27]. The \( \xi \), which is of the order of in-plane lattice constant \( a \) (table 1), increases in lower temperature range where \( \rho \) shows higher values which looks counterintuitive, yet, considering lower thermal energy this increase in resistivity is justified. Moreover, figure 8 and table 3 show that temperature range for VRH changes around 240 K (\( \sim T_c \)) and 70 K (\( < T_M \)), though at low temperature the range is extended up to 40 K below which \( \rho(T) \) exhibits steep increase. These behavior is somehow related with both magnetization and structural parameters as they exhibit sudden changes around \( T_c \) and \( T_M \) (see figures 2, 6 and 7). Given that 2D charge transport in this layered tetragonal material the sudden changes of slope in lattice parameter \( a \) and basal bond angle \( \angle \text{Ir–O2–Ir} \) around \( T_c \) and \( T_M \) would imply a certain modification in charge hopping which is realized through change in parameter \( \xi \) (table 3). It can be noted that the apical bond angle \( \angle \text{Ir–O1–Ir} \) does not go through a change retaining value at 180°. Nonetheless, it is important that the three independent measurements i.e. magnetization, structural and resistivity show reasonable agreement which rather authenticates our study.

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between two suitable sites separated by a distance of hopping mediated conduction, the probability of hopping (QI) effect which is largely viewed as quantum correction effect. This can be interpreted in terms of quantum interference is commonly understood in picture of ‘weak localization’ MR at low temperature in VRH dominated conduction process to negative with temperature around 90 K [31]. The negative while in thin film the MR shows a sign change from positive to negative with temperature around 90 K [31]. In case of Sr2IrO4, a negative MR has been evidenced in bulk sample [15] while in thin film the MR shows a sign change from positive to negative with temperature around 90 K [31]. Negative MR at low temperature in VRH dominated conduction process is commonly understood in picture of ‘weak localization’ effect. This can be interpreted in terms of quantum interference (QI) effect which is largely viewed as quantum correction in the classical Drude equation for conductivity. In case of hopping mediated conduction, the probability of hopping between two suitable sites separated by a distance \( R_{hop} \) depends on interference of connecting paths. The effect of magnetic field leads to a destruction of QI effect producing a negative MR [25]. However, the theoretical calculations employing two different approaches have shown two different dependence of MR on applied field. Nguyen et al [32] considered the approach of averaging the logarithm of conductivity over many random paths and they found the effect of magnetic field is linear. On contrary, a quadratic field dependence of MR was obtained by Sivan, Entin-Wohlman and Imry [33] who applied a method of critical path analysis to the same problem. Nonetheless, independent of field dependence the negative MR in VRH dominated conduction has been shown as a result of QI effect.

The inset of figure 9 shows MR at 5 K follows a quadratic field dependence in lower field regime up to field about 46 kOe. Note, that the MR at higher temperatures (150 and 180 K) does not exhibit such field dependence. This agreement shows QI effect is present in this compound. It can be mentioned that negative MR with quadratic dependence has been observed for many insulating disordered materials such as, \( n \)-type CdSe [34], indium oxide films [35], \( n \)-type GaAs [36], etc. However, the observation of negative MR in Sr2IrO4 with arguably high SOC is quite surprising while a positive MR is generally evidenced in strong SOC based materials. It is worth noting that a positive MR is observed at low temperature in thin film of Sr2IrO4 [31]. This implies, even though 2D hopping based charge transport is observed in present bulk Sr2IrO4 (figure 8), a real 2D localization of charges, as in thin films, is required for ‘weak antilocalization’ induced positive MR effect. Nonetheless, more investigations employing films with varying thickness are required to understand the MR behavior in Ir-based oxides.

At this point, we will add that our experimental results are very inspiring for further theoretical investigations. Our observed interrelation between structural parameters and magnetic behavior is mostly in conformity with the theoretical explanation discussed in [9], however, the result showing electrical transport behavior too linked with structural evolution and magnetic ordering need to be examined through theoretical works. Origin of weak moment in present material is an another issue which also requires theoretical investigation, though this has been primarily discussed within framework of SOC effect and distortion of IrO6 octahedra [9, 37]. Moreover, the origin of negative MR or rather QI effect needs to be theoretically understood in this bulk material where SOC effect is very prominent.

4. Conclusion

In conclusion, we have prepared polycrystalline sample of layered 5\textit{d} oxide material Sr2IrO4. Analysis of room temperature XRD data shows crystallographically the material adopts a tetragonal structure with \( I4_1/acd \) symmetry where the IrO6 octahedra exhibits a rotation around \( c \) axis by angle 11.26°. This is in conformity with previous studies. DC magnetization measurement shows a transition from paramagnetic to weak ferromagnetic state around 238 K, which is also confirmed by Arrott plot. At low temperature below 95 K, a sharp and continuous decrease in magnetization is observed. The analysis of temperature dependent XRD data shows a continuous change in structural parameters, however, a sudden change in parameters is observed around 95 K which is ascribed to observed magnetic behavior. Resistivity measurements show a throughout insulating behavior where the resistivity shows a drastic increase at low temperature increasing by about five orders. The nature of charge transport in all temperatures has been found to follow Mott’s 2D VRH model, yet three different temperature ranges have been found where the localization length changes accordingly. A negative MR is observed in all temperatures and the quadratic field dependence of MR at low temperature suggests a presence of quantum interference effect.
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