Electronic properties in itinerant ferromagnet SrRu$_{1-x}$Ti$_x$O$_3$

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Abstract

Here, we study the electrical transport and specific heat in 4$d$ based ferromagnetic material SrRuO$_3$ and its Ti substituted SrRu$_{1-x}$Ti$_x$O$_3$ series ($x \leq 0.7$). The SrRuO$_3$ is a metal and shows itinerant ferromagnetism with transition temperature $T_c \sim 160$ K. The nonmagnetic Ti$^{4+}$ ($3d^0$) substitution would not only weaken the active Ru–O–Ru channel but is also expected to tune the electronic density and electron correlation effect. A metal to insulator transition has been observed around $x \sim 0.4$. The nature of charge transport in paramagnetic-metallic state ($x \leq 0.4$) and in insulating state ($x > 0.4$) follows modified Mott’s variable range hopping model. In ferromagnetic-metallic state, resistivity shows a $T^2$ dependence below $T_c$ which though modifies to $T^3/2$ dependence at low temperature. In Ti substituted samples, temperature range for $T^3/2$ dependence extends to higher temperature. Interestingly, this $T^3/2$ dependence dominates in whole ferromagnetic regime in presence of magnetic field. This evolution of electronic transport behavior can be explained within the framework of Fermi liquid theory and electron–magnon scattering mechanism. The negative magnetoresistance exhibits a hysteresis and a crossover between negative and positive value with magnetic field which is connected with magnetic behavior in series. The decreasing electronic coefficient of specific heat with $x$ supports the increasing insulating behavior in present series. We calculate a high Kadowaki–Woods ratio ($x \leq 0.3$) for SrRuO$_3$ which increases with substitution concentration. This signifies an increasing electronic correlation effect with substitution concentration.

Keywords: transition metal oxides, electrical resistivity, specific heat, magnetoresistance, electronic correlation

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

1. Introduction

Due to its complex magnetic and transport properties, the 4$d$ based oxide SrRuO$_3$ continue to attract attention where many of the observed properties are important for technological applications [1–3]. This material is commonly believed to an itinerant ferromagnet (FM) with a transition temperature $T_c \sim 160$ K [1, 4–8]. SrRuO$_3$ further exhibits a metallic behavior where the resistivity ($\rho$) shows a linear increase till temperature as high as 1000 K, even crossing the Ioffe–Regel limit which is considered as limit for good metallic conduction [7]. The electrical charge conduction, however, has direct correlation with the magnetic ordering in this material as $\rho(T)$ shows a distinct slope change across $T_c$ where its value decreases with faster rate with decreasing temperature. While photoemission spectroscopy indicates a weak or moderate electron correlation strength ($U$) in SrRuO$_3$ [10–12], this material shows a reasonably high electronic coefficient of specific heat $\gamma$ ($\sim 30$ mJ mol$^{-1}$K$^{-2}$) and a Fermi-liquid (FL) behavior at low temperature [4, 7].

To understand its exotic magnetic as well as transport properties, the chemical substitution with suitable dopant character...
at Ru-site has often been used. In present study, we have used nonmagnetic Ti$^{4+}$ (3$d^0$) substitution to replace magnetic Ru$^{4+}$ (4$d^5$). While a least structural modification is expected due their matching ionic radii (Ru$^{4+}$ = 0.62 Å and Ti$^{4+}$ = 0.605 Å), but this substitution is expected to introduce random potential through site disorder in active Ru–O–Ru channel, create hole substitution by reducing electron density and modify the electronic correlation effect which would have significant ramifications on the magnetic and electronic properties of material. Therefore, one can expect an increase in $U$ and decrease in effective density of states at Fermi level $N(\varepsilon_F)$ in SrRuO$_3$ with substitution concentration. The photo-emission spectroscopy studies, indeed, have indicated a moderate increase of electronic correlation effect with progressive substitution of Ti [15]. The band structure calculation has been done with a home-built setup following semi adiabatic method.

**2. Experimental details**

Previous studies have shown that Ti substitution induces a metal-insulator transition around $x = 0.5$ in SrRu$_{1-x}$Ti$_x$O$_3$, however, the detail study of electrical transport behavior in presence of magnetic field and the study related to evolution of electronic correlation is lacking [17, 19]. Our study shows a metal-insulator transition is induced for $x \geq 0.4$. While the charge transport in PM-metallic (up to $x \leq 0.4$) and in insulating state ($x > 0.5$) follows a modified Mott’s variable range hopping (VRH) model, the FM-metallic state follows a $T^2$ and $T^{3/2}$ dependence and its crossover with temperature. The electronic coefficient of specific heat decreases but the Kadowaki–Woods ratio increases over the series indicating an increasing of electronic correlation effect with progressive substitution of Ti.

### 2. Experimental details

The series of polycrystalline samples SrRu$_{1-x}$Ti$_x$O$_3$ with $x = 0.0, 0.1, 0.2, 0.3, 0.4, 0.5$ and $0.7$ have been prepared using solid-state reaction method. The details of samples preparation and characterization of ingredients and temperature used have been discussed elsewhere [13, 14]. Temperature and field dependent magnetic measurements have been done using SQUID. Further, the dc electrical resistivity $\rho(T)$ and magnetoresistance (MR) data are collected using a home-built setup attached with Oxford superconducting magnet by following the four probe technique, in the temperature. The low temperature specific heat $C_p(T)$ measurements have been done with a home-built setup following semi adiabatic method.

### 3. Results and discussions

#### 3.1. Electrical resistivity study

The temperature dependent magnetic and electrical transport data of SrRuO$_3$ are shown in figure 1. The left axis of figure 1 shows the dc magnetization data of SrRuO$_3$ in temperature range between 5 K to 300 K where the data have been collected following field cooled (FC) and zero field cooled (ZFC) protocol in 100 Oe magnetic fields. The $M(T)$ data show a ferromagnetic (FM) to paramagnetic (PM) transition at temperature $T_c \sim 163$ K which is second-order phase transition in nature. The $T_c$ is indicated by a vertical dotted line and arrow. The wide bifurcation between $M_{ZFC}$ and $M_{FC}$ below $T_c$ indicates a large anisotropy associated with SrRuO$_3$ [20, 21]. The critical analysis of this PM–FM transition has shown a mean-field like magnetic interaction in SrRuO$_3$ [5, 6, 13, 14, 22, 23]. The right axis of figure 1 shows temperature dependent electrical resistivity $\rho(T)$ of SrRuO$_3$, measured in 0 and 8 T magnetic field. The $\rho(T)$ of SrRuO$_3$ shows metallic behavior throughout the temperature range. Interestingly, the $\rho(T)$ shows a slope change around $T_c$ which implies a close correlation between magnetic and transport behavior in SrRuO$_3$. Above $T_c$ in PM region, the $\rho(T)$ increases linearly without any saturation till temperature as high as 1000 K [7]. Even, $\rho(T)$ crosses the Ioffe–Regel limit for conductivity, which is an indication for bad metallic nature in SrRuO$_3$ [24–26]. Below $T_c$ in FM region, the $\rho(T)$ decreases sharply which is attributed to spin disorder effect. Further, a minimum in $\rho(T)$ (indicated by a dotted circle in figure 1) has been observed around 15 K ($T_m$), where $\rho(T)$ increases as the temperature is lowered (discussed later). The $\rho(T)$, however, does not show any major change in magnitude from 5 K (1.21 mΩ cm) to 300 K (1.98 mΩ cm) at zero magnetic field. The residual resistivity ratio (RRR), $\rho(300 K)/\rho(5 K)$, for SrRuO$_3$ is obtained to be ~1.62 which matches with other study. By applying of magnetic field (8 T), the $\rho(T)$ of SrRuO$_3$ decreases that gives a negative magnetoresistance (MR). The kink in $\rho(T)$ around $T_c$ is suppressed in presence of magnetic field which is due to broadening of magnetic transition. The MR has been calculated using following relation,

$$\text{MR}\% = \frac{\Delta \rho}{\rho(0)} \times 100 = \left(\frac{\rho(H) - \rho(0)}{\rho(0)}\right) \times 100 \quad (1)$$

where $\rho(H)$ is the resistivity in magnetic field and $\rho(0)$ is the resistivity recorded in zero magnetic field. The inset of figure 1 shows the calculated negative MR with pronounced dips around $T_c$ and at low temperature ~50 K which is also matches with reported studies [27, 28].

The $\rho(T)$ data for SrRu$_{1-x}$Ti$_x$O$_3$ series are shown in figures 2(a) and (b), respectively. It is evident in figure 2(a) that with an increase of Ti substitution, the $\rho(T)$ increases but shows an identical metallic behavior till $x \sim 0.3$. Further, figure 2(a) shows a temperature driven metal to insulator transition (MIT) around $74$ K ($T_{MIT}$) for $x = 0.4$ sample. The insulating behavior in SrRu$_{1-x}$Ti$_x$O$_3$ series increases as the Ti substitution concentration increases for $x = 0.5$ and 0.7, shown in figure 2(b) [15–17, 29]. It may be noted that for samples
with higher $x$ values, the increased magnitude of resistivity probably arises due to less number of Ru–O–Ru conducting paths as Ti$^{4+}$ substitution acts for site dilution for Ru$^{4+}$. The magnitude of $\rho(T)$ at low temperature (5 K) increases from 1.21 m$\Omega$ cm and 62.89 m$\Omega$ cm for $x = 0.7$ sample indicating a substantial change toward the insulating behavior. The $\rho(T)$ does not saturate at high temperature for metallic samples ($x = 0.0, 0.1, 0.2, 0.3$ and $0.4$) and the derivative $d\rho/dT$ (not shown here) remains finite down to the lowest temperatures. The ground state of $\rho(T)$ switches from FM-metallic to FM-insulating with Ti substitution around $x = 0.4$.

The metal-insulator transition has been a subject of intense research in condensed matter physics for long time. There are many possible reasons for metal-insulator transition such as, disorder, electron correlation, hole substitution, electron–phonon coupling, etc. The sources of disorder in real materials are many that include lattice impurities, crystal defects, chemical substitution and inhomogeneity, etc. Disorder usually provides random potential in the system that leads to localization of electronic wave-function, hence Anderson like insulating phenomena is realized. The strong electron correlation effect, on the other hand, induces Mott like insulating state in a material even with partially filled band(s). Similarly, hole substitution amounts to depletion of density of state across Fermi level which results in a metal-insulator transition behavior. The effect of both disorder and electron correlation on electronic conduction mechanism have been studied quite intensively for last few decades. In present SrRu$_{1-x}$Ti$_x$O$_3$ series, Ti$^{4+}$ replaces Ru$^{4+}$ which has $3d^3$ and $4d^3$ electronic structure, respectively. The immediate effects of present substitution are it introduces hole substitution, increases electron correlation effect and induces site disorder creating local potential where all are in favor of transition toward an insulating phase. The SrRuO$_3$ is commonly believed to a correlated metal sitting on edge of Mott transition while the other end member i.e., SrTiO$_3$ is a band insulator with an energy gap around 3.7 eV [7, 9]. Previous study on series of SrRu$_{1-x}$Ti$_x$O$_3$ films has shown metal-insulator transition occurs around $x = 0.5$, and various electronic states such as, correlated metal ($x = 0$), disordered metal ($x = 0.3$), Anderson insulator ($x = 0.5$), Coulomb gap insulator ($x = 0.6$), disordered correlation insulator ($x = 0.8$) and band insulator ($x = 1.0$) are observed across the series [17]. Our study shows a metal-insulator transition in present SrRu$_{1-x}$Ti$_x$O$_3$ series at $x ~ 0.4$ which is consistent with previous study, even considering the fact that films are accompanied by substrate strain and electronic confinement. The photo-emission spectroscopy study has, however, shown an opening of soft gap at Fermi level around $x ~ 0.5$ and a hard gap at higher values of $x$ where the metal-insulator transition is ascribed to an increasing amount of local disorder and electron correlation in the system. Nonetheless, the present series offer an ideal background to study the combined effect of disorder and electronic correlation on charge conduction on oxide system. Interestingly, it can be further mentioned that the $x$ value for metal-insulator transition in present series is close to percolation threshold ($\sim 30\%$) for 3-dimensional simple cubic lattice [18].

3.2. Electrical transport in PM state

In this section, we discuss about the charge transport mechanism in high temperature PM state for SrRu$_{1-x}$Ti$_x$O$_3$ series. Figure 2(a) shows $\rho(T)$ in PM state ($T > T_c$) continuously increases almost linearly till $x = 0.4$. On the other hand, figure 2(b) shows $\rho(T)$ continuously decreases showing an insulating behavior. For insulating disordered materials, electronic transport usually occurs due to thermally activated hopping of charge carriers among localized states around Fermi level, therefore the detail nature of density of states across Fermi level plays an important role. In most of the cases charge conduction follows Mott’s variable range hopping (VRH) model which considers hopping of trapped charge carriers in disorder materials and assumes a nearly constant density of states [31]. The disorder is, however, another parameter which has large influence on the charge conduction mechanism. To take an account of the role of disorders in...
materials, the originally proposed Mott’s VRH model has further been modified by Greaves including a \( \sqrt{T} \) term \([32]\). According to Greaves prescription, the modified Mott’s VRH model is given by

\[
\rho(T) = \rho_{\text{OM}} \sqrt{T} \exp \left( \frac{\rho_{\text{OM}} \sqrt{T}}{T} \right)^{1/4}
\]

(2)

where \( \rho_{\text{OM}} \) is the pre-exponential constant due to the electron–phonon interaction and \( T_M \) is the Mott’s characteristic temperature that depends on density of states. The \( T_M \) basically measures the degree of disorder in system and its relation with \( \rho_{\text{OM}} \) at Fermi level is given by through \( N(\epsilon_F) \).

\[
\rho_{\text{OM}} = \frac{1}{3} \nu_{\text{ph}} e^2 \left[ \frac{8 \pi \alpha k_B}{N(\epsilon_F)} \right]^{1/2}
\]

(3)

\[
T_M = 19.45 \left[ \frac{\alpha^3}{N(\epsilon_F) k_B} \right]
\]

(4)

where \( \nu_{\text{ph}} \) is the optical phonon frequency, \( k_B \) is the Boltzmann constant, \( e \) is the electronic charge and \( \alpha (=1/\xi) \) is the inverse localization length of the localized states. The \( \rho(T) \) data both in high temperature paramagnetic–metallic and in insulating state have been fitted well with equation (2) in figures 3(a) and (b), respectively. Here, we mention that tried to fit our \( \rho(T) \) data with all the available thermally activated hopping models but we find equation (2) is more suitable in terms of fitting range as well as fitting indicator. The same model (equation (2)) has been used by Kim et al. \([17]\) to fit the \( \rho(T) \) in insulating state of SrRu\(_{1-x}\)-Ti\(_x\)O\(_3\) (\( x = 0.5 \)), terming it as an Anderson insulator highlighting the role of disorder on charge conduction mechanism. Here, it can be noted that single equation (2) can be used to explain the charge transport in PM state both for metallic (\( x < 0.4 \)) as well as insulating (\( x > 0.4 \)) samples. The validity of equation (2) is extended in FM state for insulating samples. It is evident in figure 3(b) that equation (2) can be fitted in whole temperature range for samples with high Ti concentration. The obtained fitting parameters such as, \( \rho_{\text{OM}} \) and \( T_M \) are given in table 1 for the present series. In PM-metallic state the \( T_M \) changes nonmonotonically showing an oscillatory change till \( x = 0.4 \), but its value increases significantly by order of two in case of insulating materials (\( x = 0.5 \) and 0.7). While the electronic density of state \( N(\epsilon_F) \) \((\propto \text{electronic coefficient to specific heat } \gamma, \text{ equation (11)) decreases continuously with } x, \text{ the nonmonotonic changes of } T_M \text{ is surprising, which probably arises due to a nonmonotonic change of } \alpha \text{ in equation (4). The substantial increase of } T_M, \text{ however, in insulating materials is due to significant depletion of density of states in higher substituted samples.}

### 3.3 Electrical transport in FM state

For SrRuO\(_3\), the \( \rho(T) \) below \( T_c \) in FM state also shows metallic behavior, however, a distinct slope change is evident across \( T_c \) (figure 1). This implies that magnetic ordering has profound influence on the charge conduction mechanism which reduces spin disorder scattering. The charge conduction below \( T_c \) has been observed to follow the functional form,

\[
\rho(T) = \rho_{\text{OM}} + AT^2
\]

(5)

where \( \rho_{\text{OM}} \) is the residual resistivity due to impurity scattering and \( A \) is the coefficient which signifies the scattering rate. The \( T^2 \) dependence of \( \rho \) is an indication of dominant electron–electron interactions forming a Fermi liquid (FL) state which gives a different temperature dependence of \( \rho(T) \), compared to simple metals \([33]\).

We have plotted the resistivity of metallic samples (\( x \leq 0.3 \)) as a function of \( T^2 \), as shown in figure 4(a). It is evident in figure 4(a) that \( \rho(T) \) of SrRuO\(_3\) follows a quadratic temperature dependence below \( T_c \). However, this \( T^2 \) dependence deviates at low temperature which is marked by vertical arrows in figure 4(a). For Ti substituted materials (\( x = 0.1, 0.2 \) and 0.3), we also observe similar \( T^2 \) dependence below \( T_c \), however, the range of fitting modifies with Ti substitution. The temperature range for \( T^2 \) dependence as well as the values of \( \rho_{\text{OM}} \) and \( A \) (equation (5)) have been shown in table 2. For SrRuO\(_3\), \( \rho(T) \) follows a \( T^2 \) dependence down to \( \sim 67 \) K. In substituted materials the fitting range decreases while keeping upper temperature \( T_c \) fixed. This suggests that breakdown of \( T^2 \) dependence is triggered by other phenomenon, active at

![Figure 3](image-url)
Following temperature dependence, \[ T^2 \] has been calculated to be around 4.15 cm K^{-2} for SrRuO3 series with \( x = 0.0, 0.1, 0.2, \) and 0.3. (b) The same have been shown and the straight lines are due to fitting with equation (7) at low temperature region. The dotted lines represent \( T_c \).

low temperatures. Note, that parameter \( \rho_{0A} \) shows a comparatively higher value than that for single crystals or thin films but the value of \( A \) closely matches with those values \[7, 34\].

In parallel to Fermi liquid behavior, the \( T^2 \) dependence of \( \rho(T) \) has also been discussed to originate at low temperatures due to prominent electron–magnon (e–m) scattering with following temperature dependence, \[34\]

\[
\rho_{e-m} = \frac{3\pi^2 S}{16e^2\left(\frac{\mu_k B J_0}{m_e E_F}\right)^2} \frac{h^2}{2\pi k_B T^2} (6)
\]

where \( \mu \) is an effective mass of magnon, \( J_0 \) is the spin–orbit coupling constant, \( E_F \) is the Fermi energy and the \( k_p \) is the Fermi wave vector. The \( T^2 \) coefficient \( (A_{e-m}) \) in equation (6) has been calculated to be around \( 4.15 \times 10^{-7} \) mΩ cm K^{-2} for SrRuO3 taking an appropriate value of \( E_F \), Fermi velocity \( V_F \) and \( N J_0 \) \[34\]. This calculated \( A_{e-m} \) shows a low value than our obtained \( A \) in table 2. Here, it can be noted that this \( T^2 \) dependence of \( \rho(T) \) is evident at low temperatures (favorably below 40 K) in other studies of SrRuO3 \[7, 34\] but we observe this dependence at higher temperature below \( T_c \). Nonetheless, a sudden slope change in \( \rho(T) \) across \( T_c \) a significant fractional value of \( A_{e-m} \) imply that this electron–magnon scattering has contribution to \( T^2 \) dependence of \( \rho(T) \) along with electron–electron scattering related to FL behavior. Given that the electron correlation is the key factor in FL behavior, modification of \( U \) as well as electron density with Ti^{4+} (3d^4) substitution will have a significant influence on electron transport behavior, hence on the observed \( T^2 \) dependence. For instance, coefficient \( A \), which signifies the quasiparticle scattering rate, is expected to increase with the decrease of electronic density. The table 2 shows an increasing \( A \) with \( x \) which can be explained with the depletion of charge carriers as Ti^{4+} (3d^4) is introduced in the system.

Following a deviation of \( T^2 \) dependence at low temperature, we have analyzed the \( \rho(T) \) with following temperature dependence form,

\[
\rho(T) = \rho_{0B} + BT^{3/2} (7)
\]

where \( \rho_{0B} \) is the temperature independent residual resistivity and \( B \) is the coefficient. The \( T^{3/2} \) dependence in \( \rho(T) \) is mostly attributed to non-Fermi liquid (NFL) behavior which naturally arises with the breakdown of FL behavior. However, other theoretical models such as, antiferromagnetic spin fluctuations \[39\] or incoherent electron–magnon scattering \[40\] have also explained this unconventional \( T^{3/2} \) dependence of resistivity. While the former model can be excluded for this present FM SrRuO3, the later one arises from a nonconserving-wave-vector (incoherent part) scattering of electron–magnon showing a prominent effect at low temperature below the characteristic temperature \( T_m \). The coherent part of electron–magnon scattering, on the other hand, is not significant at low temperature because there are no available long-wave-vector thermal magnons to participate in scattering process with electrons. This model \[40\] further suggests a strengthening of \( T^{3/2} \) dependence with disorder. Figure 4b presents the plotting of \( \rho(T) \) vs \( T^{3/2} \) showing a linear dependence at low temperature, below the temperature range where a \( T^{2} \) dependence has been observed (figure 4a). Table 2 shows that the \( T^{3/2} \) dependence is valid in temperature range of 20 to 70 K for SrRuO3, but in Ti substituted samples this range extends to higher temperatures.

The appearance of NFL behavior usually occurs in the strange metals such as, heavy fermion materials close to quantum critical point, copper oxide based superconductors, etc \[41–44\]. The prominent example of FL to NFL crossover with substitution is system like Sr1-xCaRxO3 which shows a quantum-phase-transition across \( x = 0.7 \) \[4, 5\]. The NFL behavior has previously been reported in SrRuO3 through optical conductivity/reflectivity measurements by Kostic et al \[45\], and that has been further theoretically explained by Laad et al \[46\] from its structural organization. SrRuO3 has complex transport behavior where both FL and NFL behavior have been observed by several groups but in most cases FL behavior (\( T^2 \) dependence) has been observed at low temperatures, in contrast to present study \[25, 30, 34, 47–51\]. The \( \rho(T) \) in SrRuO3 shows a continuous linear increase (till at least 1000 K) crossing the Ioffe–Regel limit which is considered as metallic conductivity limit based on system’s lattice parameters. The FL to NFL crossover in these studies has been attributed to this bad metallic character of SrRuO3.

The crossover from \( T^2 \) to \( T^{3/2} \) dependence at low temperature in present series is quite noteworthy. We believe that this breakdown of \( T^2 \) dependence or appearance of \( T^{3/2} \) is likely to be caused by an incoherent part of electron–magnetic scattering \[40\]. An estimation of \( T_m \sim 70 \) K for SrRuO3 by Mazin and Singh \[8\] matches with the upper temperature limit (70 K).
Nonetheless, the observation of both $T$ higher than the case where magnetic field. However, our $B$ parameter is roughly one order higher than the case where $T^{3/2}$ dependence has been observed at high temperature (50 K) ascribing to NFL behavior. This mostly insensitive behavior of $T_K$ and the nature of shifting of $T_K$ in magnetic field implies this minimum in $TK$ is unlikely due to intergrain transport or weak localization effect. The Kondo effect arises due to scattering of conduction electrons with the magnetic impurities resulting in a continuous increase in resistivity at low temperature [35, 36]. In case of Kondo behavior, resistivity follows logarithmic of temperature dependence with following functional form,

$$\rho(T) = \rho_{OK} - C \ln T$$  \hspace{1cm} (8)

where $\rho_{OK}$ is the Kondo residual resistivity (i.e., sample impurity) and C is the Kondo coefficient. Following equation (8), we have plotted $\rho(T)$ vs $\ln T$ in figure 6 where the linear behavior close to $T_K$ indicates Kondo-like behavior till Ti substitution concentration $x$ = 0.3. As we increase the Ti substitution level ($x > 0.3$), $\rho(T)$ shows an insulating behavior and the Kondo effect disappears which also suggests this minimum in $\rho(T)$ is unlikely due to intergrain transport mechanism. Recently, the possibility of Kondo behavior has been discussed in SrRuO$_3$ films [32]. The parameters ($\rho_{OK}$ and C) obtained from fitting boundaries where in zero magnetic field the spins in neighboring grains have non-parallel alignment which restricts the charge carrier movement, giving an increasing resistivity at low temperature. With increasing temperature, the thermal energy helps for spin reorientation that results in a minimum in $\rho(T)$. This minimum in $\rho(T)$ due to intergrain transport mechanism is highly sensitive to the applied magnetic field which lowers the minimum toward lower temperature or even vanishes the minimum [53]. Weak localization effect, on the other hand, mostly arises in disordered materials due to interference of electronic wavefunctions when they are coherently backscattered by randomly distributed scattering centers. In weak localization effect, the conductivity ($\sigma$) follows a $T^{1/2}$ dependence. In present SrRuO$_3$, we find $\sigma \propto T^{1/2}$ dependence is followed up to around 8 K (not shown) which is much lower than the $T_K$. The left inset of figure 1 shows the minimum in $\rho(T)$ or $T_K$ does not shift significantly with magnetic field. For instance, in magnetic field as high as 8 T the $T_K$ shifts to higher temperature only by ~1.5 K. This mostly insensitive behavior of $T_K$ and the nature of shifting of $T_K$ in magnetic field implies this minimum in $\rho(K)$ is unlikely due to intergrain transport or weak localization effect. The Kondo effect arises due to scattering of conduction electrons with the magnetic impurities resulting in a continuous increase in resistivity at low temperature [35, 36]. In case of Kondo behavior, resistivity follows logarithmic of temperature dependence with following functional form,

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### Table 2. The coefficients $A$, $B$ and $C$ in equations (5), (7) and (8), respectively are shown along with their effective temperature range of fitting for SrRu$_{1-x}$Ti$_x$O$_3$ series.

| Field | Property | Parameter | $x = 0.0$ | 0.1 | 0.2 | 0.3 |
|-------|----------|-----------|-----------|-----|-----|-----|
| 0 T   | $T^2$ $\rho_{OA}$ (m$\Omega$ cm) | 1.25 | 1.37 | 1.93 | 2.18 |
|       | $A$ (m$\Omega$ cm K$^{-2}$) $\times$ 10$^{-5}$ | 1.89 | 3.48 | 2.81 | 4.25 |
|       | $T$ range (K) | 67–163 | 119–163 | 117–163 | 91–163 |
| 0 T   | $T^{3/2}$ $\rho_{OB}$ (m$\Omega$ cm) | 1.18 | 1.31 | 1.90 | 2.11 |
|       | $B$ (m$\Omega$ cm K$^{-3/2}$) $\times$ 10$^{-4}$ | 2.98 | 4.33 | 3.30 | 4.96 |
|       | $T$ range (K) | 20–70 | 38–131 | 85–123 | 41–106 |
| 8 T   | $T^{3/2}$ $\rho_{OB}$ (m$\Omega$ cm) | 1.17 | 1.25 | 1.86 | 1.98 |
|       | $B$ (m$\Omega$ cm K$^{-3/2}$) $\times$ 10$^{-4}$ | 2.58 | 4.44 | 3.42 | 5.60 |
|       | $T$ range (K) | 27–163 | 47–163 | 87–163 | 83–163 |

**Figure 5.** The resistivity vs temperature data in 0 and 8 T field are shown for representative (a) $x = 0.0$ and (b) $x = 0.3$ material of SrRu$_{1-x}$Ti$_x$O$_3$ series. The $\rho(T)$ of 8 T has been shifted vertically by 0.2 for clarity. The straight lines are due to fitting with equation (7).
Figure 6. Temperature dependent resistivity are plotted in semi-log scale at low temperature for SrRu$_{1-x}$Ti$_x$O$_3$ series with $x = 0.0$, 0.1, 0.2, and 0.3. The straight lines are due to fitting following Kondo behavior (equation (8)).

of equation (8) are given in table 2 in 0 and 8 T field for $x \leq 0.3$. As evident, both the parameters do not change appreciably and remain nearly constant within the limit of error bar. This field-independent nature of coefficient $C$ is further regarded as typical feature of Kondo behavior [52–54]. Therefore, this low temperature minimum in $\rho(T)$ in present series ($x \leq 0.3$) is likely due to Kondo-like behavior.

Figure 7. The magnetoresistance (equation (1)) with variation of field are shown (a) at 5 K and (b) at 200 K for SrRu$_{1-x}$Ti$_x$O$_3$ series. (c) Shows MR at 200 K with quadratic dependance of field. (d) and (e) Show the value of MR at 8 T field of SrRu$_{1-x}$Ti$_x$O$_3$ series at 5 K and 200 K, respectively.

3.5. Magnetoresistance

The electrical resistivity have further been measured in presence of magnetic field. The MR calculated for SrRuO$_3$ shows a negative value throughout the temperature range where two prominent dips around $T_c$ and 50 K are observed (figure 1) [27, 28]. The dip in MR across $T_c$ is associated with broadening of phase transition where $\rho(T)$ shows a smooth change. Magnetic field dependent MR have been measured at different temperatures to understand the effect of magnetic state on charge transport. For instance, we have plotted the isothermal MR for present series at two selected temperatures i.e., 5 K and 200 K, which represents FM and PM regions, respectively in figures 7(a) and (b). A negative MR has been observed at both these temperatures. The MR varies linearly with the applied magnetic field in FM regime at 5 K (figure 7(a)) while in PM state, this variation is nonlinear for all the samples (figure 7(b)). However, a linear variation of MR in PM state at 200 K is observed with square of magnetic field in figure 7(c). The quadratic field dependance of MR has been observed for both metallic as well as nonmetallic samples without spin ordering (PM state) at high temperatures. In PM state above $T_c$, MR $\propto H^2$ basically implies an MR $\propto M^2$ due to linear relationship $M = \chi H$ where $M$ is the magnetic moment and $\chi$ is the magnetic susceptibility. In PM state, as the charge carriers are scattered by thermally fluctuating spins, this increasing moment with magnetic field reduces the spin fluctuations which in turn increases the charge conductivity, hence a negative MR is realized. In figure 7(d), we have shown the MR value observed at highest magnetic field of 8 T for different Ti concentration in FM region at 5 K. For the metallic samples till $x = 0.3$, the MR is not impressive and its value does not change much with $x$. With the introduction insulating phase ($x \geq 0.4$) however, MR increases even in same FM state, implying spin ordering with magnetic field promotes charge conduction. Interestingly, an opposite evolution of MR has been observed at high temperature PM state where MR value decreases with $x$, though its values are not significantly different. This can be explained due to site dilution effect. This signifies the role of magnetic ordering and magnetic field on the charge transport behavior in present SrRu$_{1-x}$Ti$_x$O$_3$ series.

Figure 7(e) shows MR ratio at 8 T magnetic field for different Ti substitution concentration at 200 K in the paramagnetic region, as evaluated from figure 7(b). It is evident from the data that in PM region MR does not have a significant change in magnitude as compared to FM regime. In PM region, a maximum MR around 1% is observed for $x = 0.1$ and the MR value decreases with increase in Ti substitution concentration to 0.2% for $x = 0.7$.

Further, the evolution of MR has been studied with sweeping of magnetic field from 0 to $+8$ T and then to $-8$ T to 0 to $+8$ T at 5 K, indicated by arrows in figure 8. For SrRuO$_3$, MR is observed to be negative and it shows a reasonable hysteresis below $\sim 4$ T. It also shows a remnant MR at zero field after returning from higher field. Interestingly, when field is reversed toward negative direction, the negative MR continue to decrease and the MR shows a small positive value and then with further increase of field, MR becomes again negative. The field where this positive MR arises closely matches with the coercive field $H_c$ of magnetic hysteresis loop. This shows positive MR arises due to weakening of moment at low
temperature. Similar hysteresis and remnant value in MR has also been observed in negative field cycle (figure 8(a)). This behavior of MR appears to be associated with the nature of magnetism in the material. For instance, the inset of figure 8(a) shows the magnetic hysteresis loop $M(H)$ of SrRuO$_3$ at 5 K which similarly shows a wide hysteresis below $\sim 4$ T as well as high remnant magnetization at zero magnetic field. This underlines that the spin ordering and charge transport behavior in SrRuO$_3$ is closely related. While returning from high magnetic field (8 T), system retains its induced moment which results in a higher negative MR and a remnant MR, this causes hysteresis in both $M(H)$ and MR. With increasing temperature, both the hysteresis as well as remnant MR decreases due to softening of spin ordering (figure 8(b)). We, however, have not found hysteresis in high temperature PM state (not shown). Further, with dilution of spin ordering through Ti substitution, we find both the hysteresis and remnant MR reduces, as seen in figures 8(c)–(f) for $x = 0.1$, 0.3, 0.4 and 0.7, respectively. Here, it can be noted that with increasing temperature the similar decrease in hysteresis and remnant in MR has been observed for Ti substituted samples (not shown). This signifies the dominant role of magnetism on transport behavior in SrRuO$_3$.

3.6. Specific heat

The specific heat ($C_p$) has mainly been measured to understand an evolution of charge transport behavior and electron correlation effect in present series. In figure 9(a), we have presented the temperature dependent specific heat for two representative samples i.e., $x = 0$ and 0.2. It is clear in figure 9 that at low temperature ($< 20$ K) $C_p(T)$ does not vary much, but above this temperature the $C_p(T)$ increases almost linearly. For $x = 0$ parent compound, $C_p(T)$ exhibits a pronounced jump around $T_c$ which is shown in a magnified view in upper inset of figure 9(a). The $C_p(T)$ for $x = 0.2$ sample shows similar behavior at low temperature but the jump across $T_c$ is much softened. This implies that with Ti substitution, magnetic transition has been broadened, as also evidenced with a dip in MR across $T_c$ (see figure 1). Similar signature of broadening of transition has also been observed in temperature dependent magnetization data which are shown in lower inset of figure 9(a). Here, it can be mentioned that similar behavior of $C_p(T)$ data has been observed in other materials in present SrRu$_{1-x}$Ti$_x$O$_3$ series. The specific heat has mainly two contributions namely, electronic and lattice contribution which has given below,

$$C_p = \gamma T + \beta T^3$$  \hspace{1cm} (9)\]

where $\gamma$ and $\beta$ are the electronic ($C_e$) and lattice ($C_l$) coefficients of specific heat, respectively. To identify the individual contribution of electronic and lattice part, the $C_p(T)$ data of equation (9) have been plotted as following,

$$C_p/T = \gamma + \beta T^2$$  \hspace{1cm} (10)\]

The straight line fitting of $C_p(T)$ data using equation (10) gives $\gamma = 30$ mJ mol$^{-1}$K$^{-1}$ and $\beta = 0.138$ mJ mol$^{-1}$K$^{-4}$, respectively.
The electronic specific heat coefficient $\gamma$ (left axis) and the Debye temperature $\Theta_D$ (right axis) are shown for SrRu1-xTiO3 series. (b) The Kadowaki–Woods ratio (see text) is shown for the metallic samples of present series.

Figure 10. The electronic specific heat coefficient $\gamma$ (left axis) and the Debye temperature $\Theta_D$ (right axis) are shown for SrRu1-xTiO3 series. (b) The Kadowaki–Woods ratio (see text) is shown for the metallic samples of present series.

respectively for SrRuO3, where these values closely match with the reported values for this material [4, 7]. This high value of $\gamma$ suggests a considerable electronic correlation in SrRuO3. The electronic density of states $N(\epsilon_F)$ has been calculated directly from $\gamma$ as,

$$\gamma = \frac{\pi^2 k_B^2 N_a N(\epsilon_F)}{3} \tag{11}$$

that gives $N(\epsilon_F) = 174$ states Ry f.u. for SrRuO3 (for both spin directions) with $N_a$ is the Avogadro number. Similarly, the Debye temperature ($\Theta_D$) has been calculated from $\beta$ using the following relation

$$\Theta_D = \left[ \frac{12\pi^4 n R}{5\beta} \right]^{1/3} \tag{12}$$

where $R = 8.314$ JK⁻¹mol⁻¹ is the universal molar gas constant and $n = 5$ is the number of atoms per formula unit of SrRuO3. Using equation (12), we have calculated $\Theta_D \sim 413$ K for SrRuO3. The estimated values of $\gamma$ and $\Theta_D$ have been shown in figure 10(a) for present series where $\gamma$ or equivalent $N(\epsilon_F)$ (equation (11)) and $\Theta_D$ decrease with progressive substitution of Ti. This behavior of $\Theta_D$ is likely due to substitution of lighter element for Ru. The decrease in density of states is reflected in an increase of resistivity with $x$ in SrRu1-xTiO3 series, as shown in figure 2(a). Here it can be noted that our result is in agreement with photo-emission data which similarly shows a decrease of density of states in SrRu1-xTiO3 with $x$ [15].

3.7. Kadowaki–Woods ratio

The Kadowaki–Woods ratio (KWR), $A/\gamma^2$, is about comparing the coefficient of the quadratic term in $\rho(T)$ ($A$ in equation (5)) with the coefficient of linear term in $C_p(T)$ ($\gamma$ in equation (9)) at low temperature. According to FL model, the $A$ is squarely proportional to quasiparticle mass enhancement due to electron correlation effect. This KWR is believed to be suggestive of electron correlation strength in a material, and shows a constant or near constant value for a definite class of material [55]. For instance, its value has been found to be 0.4 and 10 $\mu\Omega$ cm mol⁻¹ K² J⁻¹ in case of transition metals and heavy fermions, respectively [55–58]. For SrRuO3, using $A = 1.9 \times 10^{-5} \mu\Omega$ cm K⁻² and $\gamma = 30$ mJ mol⁻¹ K⁻², we calculate KWR to be around 10 $\mu\Omega$ cm mol⁻¹ K² J⁻¹ that matches with other report [34]. This value of KWR for SrRuO3 is almost double of that for heavy fermion systems but in the range of transition metal oxides [55]. For example, La1.5Sr0.5CuO4 shows KWR about 50 $\mu\Omega$ cm mol⁻¹ K² J⁻¹, while a very high value around 500 $\mu\Omega$ cm mol⁻¹ K² J⁻¹ has been observed in Na0.7CoO2 [59, 60]. With Ti concentration, KWR increases reaching around 80 $\mu\Omega$ cm mol⁻¹ K² J⁻¹ for $x = 0.3$ sample. This increase of KWR by around four times with 30% of Ti is quite noteworthy which underlines that SrRuO3 has reasonable $U$ which further enhances with introduction of 3d element in present SrRu1-xTiO3 series. This behavior is consistent with our previous report where the constant $T_c$ in present series has been explained due to simultaneous increase of $U$ and decrease of density of states at Fermi level with substitution of Ti4⁺ [13].

4. Conclusion

To understand the evolution of electronic correlation and charge transport behavior, a series of polycrystalline SrRu1-xTiO3 samples are prepared. The parent SrRuO3 shows a metallic behavior throughout the temperature range, however, a distinct correlation between charge conduction and spin ordering is evident below $T_c$. The electrical resistivity of SrRu1-xTiO3 series increases and a metal to insulator transition has been observed around 40% of Ti substitution. The electrical resistivity both in PM-metallic state ($x \leq 0.4$) as well as in insulating state ($x > 0.4$) follows a modified Mott’s VRH model. In FM metallic region, a crossover from $T^2$ to $T^{3/2}$ dependence in resistivity has been observed at low temperature where with applied magnetic field the $T^{3/2}$ dependence prevails over entire FM state. The Fermi liquid and electron–magnon scattering are believed to be reason for this behavior. At low temperature, Kondo like behavior found for samples up to $x \leq 0.4$. The negative MR shows a linear and quadratic increase with magnetic field in FM and PM regime, respectively. Further, a reasonable hysterisis and a crossover from negative to positive value in MR have been observed at low temperature which decrease both with increasing temperature and substitution concentration. This evolution of MR has been associated with the magnetic ordering in the materials. The decreasing value of electronic coefficient of specific heat are in agreement with increasing...
insulating behavior in present series. We observe a relatively high value of Kadawaki–Woods ratio for SrRuO$_3$ which further increases with substitution indicating an increase of electronic correlation effect with Ti substitution.

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