Weak localization in the layered iridates $Sr_2Ir_{1-x}M_xO_4(M = Ru, Ti)$: Role of interplay between spin-orbit coupling and magnetism

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The combined role of the spin-orbit effect (SOC), electron correlation (*U*), and magnetism has had a significant impact on the electron transport behavior in disordered systems. Here, we report an experimental investigation of weak localization (WL) behavior in layered SOC-dominated iridate Sr_2IrO_4 where these impacting parameters are further tuned with substitution of Ru and Ti, which have different magnetic and electronic characters. Magnetic Ru^{4+} ($4d^4$) participates in magnetic interaction with existing Ir^{4+} as well as with itself, while nonmagnetic Ti^{4+} ($3d^0$) will lead to the dilution of a magnetic lattice. Sr_2IrO_4 shows a crossover from negative to positive magnetoconductivity (MC) with the onset of magnetic ordering where the WL behavior is observed in a magnetically ordered state. In $Sr_2Ir_{1-x}Ru_xO_4$ series, a complete suppression of the magnetic and insulating state is observed with $x \sim 0.6$. However, an opposite result is seen in $Sr_2Ir_{1-x}Ti_xO_4$ series, where the magnetic ordering temperature is minimally influenced, though the magnetic moment is weakened and the insulating state is significantly strengthened. This contrasting effect on magnetism has a different impact on the crossover in MC and allied WL behavior, which basically follows the evolution of the magnetic and conducting state in both series. The Ti⁴⁺ further reverses the negative MC in the nonmagnetic state to positive MC with a reduction in SOC. This work demonstrates the critical role of magnetism, SOC, and *U* in manipulating the quantum transport properties in one of the iridate materials which we believe has significance in other similar materials.

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I. INTRODUCTION

Electron transport in disordered materials continues to be a widely studied topic in condensed-matter physics, where the combined role of magnetism, electronic correlation (U), and spin-orbit coupling has a significant effect [1]. The quantum interference (QI) effect, which is regarded as a quantum correction to the classical Drude model, has been investigated quite intensively in these materials in order to understand the charge conduction mechanism. QI has mainly two dominant sources: (i) strong U and related modification in the density of states at the Fermi level, and (ii) the weak localization (WL) effect, which arises due to constructive interference between two time-reversed self-intersecting electron paths as they experience coherent backscattering by impurities or other defects while passing through the diffusive system. While both of these effects lead to a decrease in conductance, the WL effect is generally evidenced in systems with weak disorder. In systems with strong spin-orbit coupling (SOC), however, prominent spin-momentum locking induces an additional π phase in the spin part of the wave function. This leads to a destructive interference that nullifies the backscattering, an effect known as weak antilocalization (WAL). The applied magnetic field or the magnetic moment has a profound effect on the WL/WAL behavior as it acts as a dephasing agent. Generally, positive or negative magnetoconductivity (MC) with a sharp cusp around zero field is considered to be a defining feature for WL and WAL, respectively [2].

The 5*d*-based layered Sr_2IrO_4 (n = 1 in the Ruddlesden-Popper phase, $Sr_{n+1}Ir_nO_{3n+1}$) is currently attracting a great deal of interest [3-7]. Because Sr₂IrO₄ is isostructural to La2CuO4, the goal of many theoretical and experimental studies has been to understand the possible superconducting phase in its doped compounds [8,9]. This material is basically a magnetic insulator with a $J_{\text{eff}} = 1/2$ ground state [10]. Due to the heavy weight and extended orbitals of Ir, a strong SOC and reduced U are realized. The strong SOC leads to a splitting of t_{2g} orbitals into $J_{\text{eff}} = 1/2$ and 3/2 states. With the $5d^5$ electronic structure of Ir⁴⁺, a fully filled $J_{\text{eff}} = 3/2$ quartet and a half-filled $J_{\text{eff}} = 1/2$ doublet are obtained. Due to weak U, the narrow $J_{\text{eff}} = 1/2$ band splits and opens a Mott-like gap in the electronic band. The $J_{eff} = 1/2$ pseudospins of Ir⁴⁺ sitting on an in-plane square lattice in a Heisenberg-type antiferromagnetic (AFM) exchange interaction with ordering temperature $T_N \sim 225$ K [11–13]. The reduced lattice symmetry in Sr_2IrO_4 plays a vital role in which a rotational distortion of IrO₆ octahedra induces a Dzyaloshinskii-Moriya (DM) -type antisymmetric interaction, which is believed to give rise to weak ferromagnetic (FM) behavior in this material [14,15]. Therefore, the unique combination of a quasi-twodimensional layered structure, strong SOC, moderate U, and exotic electromagnetic behavior makes this material ideal for studying quantum transport behavior.

In this paper, we have studied WL behavior in Sr_2IrO_4 . Further, to examine the effect of the interplay between SOC

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and magnetism on WL properties, the magnetic and electronic states in the original material are tuned with a substitution of $\operatorname{Ru}^{4+}(4d^4, S = 1, 0.62 \text{ Å}) \text{ and } \operatorname{Ti}^{4+}(3d^0, S = 0, 0.605 \text{ Å}) \text{ for}$ Ir⁴⁺ (5 d^5 , $J_{\text{eff}} = 1/2$, 0.625 Å). Both of these elements have different d characters, but they will introduce hole doping and tune the parameters such as SOC and U accordingly. Due to their matching ionic radii, these substitutions will not cause major structural modifications, but the magnetic Ru⁴⁺ will supposedly participate in exchange interaction with Ir^{4+} and itself, while nonmagnetic Ti⁴⁺ will simply dilute the magnetic lattice. In fact, there have only been a few studies on $Sr_2Ir_{1-x}Ru_xO_4$ showing a total suppression of the magnetic and insulating state in Sr_2IrO_4 with x around 0.5–0.6 [16–19], which is also connected to the fact that the end compound (x = 1.0), i.e., Sr₂RuO₄, is a nonmagnet [20]. Our present study shows that Ru⁴⁺ suppresses both the magnetic and insulating state at $x \sim 0.6$. In the case of Ti⁴⁺ substitution, T_N does not change much (though the magnetic moment is weakened) while the insulating state is significantly enhanced. These contrasting behaviors have a different impact on the WL state, thus underlining the complex role of SOC and magnetism in charge-transport behavior.

II. EXPERIMENTAL DETAILS

Two different series of polycrystalline samples $Sr_2Ir_{1-x}M_xO_4$ with M = Ru and Ti (x = 0, 0.05, 0.10, 0.20,0.40, and 0.60) are prepared by a conventional solid-state method, as detailed elsewhere [13,21-24]. The high-purity powder ingredients SrCO₃, IrO₂, RuO₂, and TiO₂ (Sigma Aldrich, purity ~99.99%) are mixed in a 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 $3 \,^{\circ}$ C min¹. The calcinated powders are then pressed into pellets and sintered at 1000 and 1100 °C for 24 h at the same heating and cooling rates with intermediate grinding. The phase purity of prepared materials is checked with powder x-ray diffraction (XRD) using a Rigaku made diffractometer. The XRD data are collected at room temperature in a 2θ range of 10° – 90° with a step of 0.02° and a scan speed of 2° /min. Structural analysis has been performed by Rietveld refinement of XRD data using the FULLPROF program. The x-ray photoemission spectroscopy (XPS) study has been done to probe the oxidation states of cations in the present series. The XPS measurements are performed with a base pressure of $\sim 10^{-10}$ mbar using a commercial electron energy analyzer (Omnicron nanotechnology) and a nonmonochromatic Al $K\alpha$ x-ray source (hv = 1486.6 eV). The CASAXPS software has been used to analyze the XPS data. The samples used for the XPS study are in pellet form, where ion beam sputtering has been done to expose the clean surface before measurements. The dc magnetization data are collected in a Quantum Design made Physical Properties Measurement System (PPMS). Resistivity measurements have been done following a four-probe technique on a homemade system attached to an Oxford magnet. Magnetoconductivity data are collected at different temperatures up to a magnetic field of ± 80 kOe. For both magnetic and transport measurements, samples are taken in a cuboid shape, with each side a few mm in length.

III. STRUCTURAL STUDY

Sr₂IrO₄ is a layered material that belongs to the K₂NiF₄ class of compounds. The layered structure is evident in the unit cell where each SrIrO₃ layer is separated by a SrO layer, thus making it a quasi-two-dimensional structure [Fig. 1(a)]. Further, the IrO₆ octahedra show a rotational distortion (θ_{oct}) along the *c*-axis [Fig. 1(b)]. This θ_{oct} is believed to induce a Dzyaloshinskii-Moriya (DM) -type antisymmetric interaction, hence it has a significant influence on its magnetic and electronic properties [13,15]. Figures 1(c)-1(e) show the XRD pattern along with Rietveld refinement for the representative compositions of studied series $Sr_2Ir_{1-x}M_xO_4$ (M = Ru and Ti), i.e., x = 0.0 [Fig. 1(c)], x (Ru) = 0.60 [Fig. 1(d)], and x (Ti) = 0.60 [Fig. 1(e)]. The structural parameters, atomic positions, Ir-O bond angles, octahedral distortion θ_{oct} , and refinement judging parameters, such as χ^2 and "goodness of fit (GOF)" (R_{wp}/R_{exp}) as obtained from Rietveld refinement, are summarized in Table I for representative x = 0, 0.2, and0.6 samples. We have obtained similar low values for both χ^2 and GOF across both series, which suggests the refinement is reasonably good and in line with earlier works [25,26]. Rietveld refinement further shows that all the samples are in single-phase and crystallize in tetragonal- $I4_1/acd$ structure with reduced symmetry. This reduced symmetry is due to IrO₆ octahedral distortion, as discussed before. The Ru/Ti substitution does not induce any structural phase transition; however, a slight variation in diffraction peak intensities and position is observed, which may be due to the evolution of lattice parameters. Considering the slight mismatch between ionic radii of Ru^{4+} (0.62 Å), Ti^{4+} (0.605 Å), and Ir^{4+} (0.625 Å), no major structural modification is observed until the highest doping concentration, x = 0.6.

Figures 1(f) and 1(g) show the evolution of lattice constants *a* and *c* for M = Ru and Ti, respectively. It is evident from the figures that lattice constant *a* (left axis) increases while *c* (right axis) decreases monotonically with *x*. Substitution of smaller size $\text{Ru}^{4+}/\text{Ti}^{4+}$ causes a contraction in parameter *c*, while the slight increase in *a* is due to the straightening of the basal plan $\langle \text{Ir-O1-Ir} \rangle$ bond angle. The estimated θ_{oct} has been shown in Fig. 1(h) for both series. While θ_{oct} decreases in both series, the effect is very prominent in the case of Ru showing a variation from 11.3° for x = 0.0 to 2.1° for x = 0.60. This large decrease of θ_{oct} will have a large influence on the physical properties.

IV. X-RAY PHOTOEMISSION SPECTROSCOPY STUDY

Considering that the electronic charge state of transition metals plays a crucial role in the physical properties of oxide materials, we have carried out x-ray photoemission spectroscopy (XPS) measurements at room temperature on parent as well as on Ru- and Ti-substituted samples. Figure 2 presents XPS data along with fittings for representative samples. The Ir-4*f* core-level XPS spectra have been shown in Figs. 2(a), 2(b) and 2(c) for parent x = 0.0, 0.40(Ru), and 0.40 (Ti) samples, respectively. The solid red lines in these figures are due to an overall fitting of Ir-4*f* data taking individual contributions of both Ir⁴⁺ (solid blue line) and Ir⁵⁺ (solid orange line). The analysis indicates that Ir mostly are



FIG. 1. (a) Unit cell of Sr₂IrO₄ showing the structural organization of IrO₂ and SrO layers along with IrO₆ octahedra. (b) Rotation of IrO₆ octahedra around the *c*-axis and Ir-O1-Ir bond angle in the *ab* plane. An x-ray diffraction pattern is shown along with Rietveld refinement for representative samples (c) x = 0.0, (d) x (Ru) = 0.60, and (e) x (Ti) = 0.60 for Sr₂Ir_{1-x} M_x O₄ (M = Ru and Ti) series. A variation of lattice parameter *a* (left axis) and *c* (right axis) for Ru series (f) and Ti series (g) is shown. (h) The octahedral rotational distortion θ_{oct} related to IrO₆ octahedra are shown as a function of doping concentration *x* for these two series.

in an Ir⁴⁺ ionic state, however some contribution of Ir⁵⁺ has also been observed. Spin-orbit-split Ir⁴⁺ spectra, i.e., $4f_{7/2}$ and $4f_{5/2}$ electronic states, appear around 62 and 65 eV of binding energies, respectively [27,28]. Similar spin-orbit-split doublets for Ir⁵⁺ are found around 63 and 68 eV, respectively. For Sr₂IrO₄, the analysis further implies that the amount of Ir⁴⁺ and Ir⁵⁺ is about ~96% and ~4%, respectively. In the case of Ru/Ti substitution, the content of Ir⁵⁺ remains almost the same at ~4%. The presence of Ir⁵⁺ at this minimal level, which may be due to nonstoichiometry during sample growth, has also been reported in other iridates [21,27].

Figure 2(d) shows Sr-3*d* spectra for the x = 0.0 sample, where the open circle is the experimental data and the solid red line is the fitted envelope curve. The solid blue lines are due to spin-orbit split Sr-3*d*_{3/2} and Sr-3*d*_{1/2} peaks arising at binding energies around 132.9 and 134.2 eV [29,30]. The analysis indicates an Sr²⁺ electronic state in all samples.

The Ru-3*d* core-level spectra for x = 0.4 are shown in Fig. 2(e), which shows two peaks corresponding to spin-orbit-split $3d_{5/2}$ and $3d_{3/2}$ at binding energies of 279.21 and 284.89 eV, respectively, with a splitting of ~5.68 eV [29].

The analysis of XPS data reveals that Ru is in the Ru⁴⁺ charge state. Similarly, the core-level spectra of Ti-2*p* are presented in Fig. 2(f) for x = 0.40, showing two spin-orbit-split peaks $2p_{3/2}$ and $2p_{1/2}$ at binding energies 463.5 and 458 eV, respectively, with a splitting of ~6.5 eV [31,32]. This confirms the Ti⁴⁺ charge state. Due to matching charge states, it is expected that both Ru⁴⁺ and Ti⁴⁺ will replace Ir⁴⁺ in the original Sr₂IrO₄, hence hole doping will be introduced accordingly.

V. MAGNETIZATION STUDY

The magnetism in Sr₂IrO₄ is mainly decided by Ir⁴⁺ (5*d*⁵), which forms a square lattice in a layer. Due to strong spinorbit coupling, the filled t_{2g} state in Ir⁴⁺ is further split into a fully filled $J_{\text{eff}} = 3/2$ quartet and a half-filled $J_{\text{eff}} = 1/2$ doublet. This gives the realization of a $J_{\text{eff}} = 1/2$ magnetic state in this material. The $J_{\text{eff}} = 1/2$ pseudospins engage in Heisenberg-type antiferromagnetic interaction where the octahedral distortion induces DM interaction, which gives rise to a weak ferromagnetic interaction with $T_N \sim 225$ K [11,13]. TABLE I. Structural parameters, atomic positions, Ir-O bond angles, octahedral distortion, and refinement judging parameters as determined from the Rietveld refinement of the powder XRD patterns are shown for representative samples of $Sr_2Ir_{1-x}M_xO_4$ (M = Ru,Ti) series at room temperature with $I4_1/acd$ space group. Here O2 refers to apical oxygen, and O1 refers to basal oxygen, which lies in the plane of the perovskite layer.

Parameters	Sr_2IrO_4	$Sr_2Ir_{0.8}Ru_{0.2}O_4$	$Sr_2Ir_{0.4}Ru_{0.6}O_4$	$Sr_2Ir_{0.8}Ti_{0.2}O_4$	$Sr_2Ir_{0.4}Ti_{0.6}O_4$
a (Å)	5.4980(2)	5.5031(1)	5.5079(5)	5.5025(2)	5.5060(3)
c (Å)	25.779(1)	25.616(1)	25.560(2)	25.606(2)	25.490(3)
$V(Å^3)$	779.24(7)	775.75(7)	775.41(2)	775.28(5)	772.75(5)
Sr site (16d)					
x	0.0	0.0	0.0	0.0	0.0
v	0.0	0.0	0.0	0.0	0.0
z	0.17506(2)	0.17680(1)	0.17683(1)	0.17679(2)	0.17672(6)
Ir/Ti/Ru site (8a)	. ,				. ,
x	0.0	0.0	0.0	0.0	0.0
y	0.0	0.0	0.0	0.0	0.0
z	0.0	0.0	0.0	0.0	0.0
O1 site (16f)					
x	0.20021(5)	0.21584(6)	0.27296(3)	0.21469(4)	0.21973(5)
y	0.20021(5)	0.21584(6)	0.27296(3)	0.21469(4)	0.21973(5)
z	0.25000	0.25000	0.25000	0.25000	0.25000
O2 site (16d)					
x	0.0	0.0	0.0	0.0	0.0
v	0.0	0.0	0.0	0.0	0.0
z	0.07995(3)	0.07680(2)	0.07323(4)	0.07678(3)	0.07212(3)
R factors %					
R _{wp}	7.25	8.35	8.73	6.80	9.05
Rexp	5.62	6.00	6.51	5.15	6.24
$R_{\rm wp}/R_{\rm exp}$	1.29	1.39	1.34	1.32	1.45
χ^2	1.5	1.8	2.1	1.45	2.4
(Ir-O1-Ir)	157.4°	164.5°	175.9°	163.4	164.20
$\theta_{\rm oct}$	11.30°	7.75°	2.05°	8.3	7.9

The substitution of Ru and Ti shows a distinct effect on the magnetic state in Sr_2IrO_4 .

The temperature-dependent magnetization M(T) measured under a zero-field-cooled (ZFC) and a field-cooled (FC) condition in a field of 10 kOe are shown in Figs. 3(a) and 3(c) for Ru and Ti substitutions, respectively. As is evident in the figures, both Ru and Ti suppress the magnetic state by lowering the moment. While for Ru the T_N decreases drastically and the highest x = 0.6 material turns out to be paramagnetic (PM), the T_N appears to change minimally with Ti. Here, it can be noted that the present results of $Sr_2Ir_{1-x}Ru_xO_4$ closely match the reported studies, which show a total suppression of the magnetic and insulating state with x around 0.5-0.6 [16-19]. This suggests that the Ru⁴⁺, despite being magnetic, does not favor the existing magnetic in host lattice, which is also connected to the fact that the end compound (x = 1.0), i.e., Sr₂RuO₄, is a nonmagnet [20]. The high-temperature paramagnetic (PM) state is studied with temperature-dependent inverse magnetic susceptibility χ^{-1} (= H/M), as shown in Fig. 4(a) for the representative samples. For x = 0.0, the $\chi^{-1}(T)$ shows linear behavior above T_N ; however, above 280 K the $\chi^{-1}(T)$ deviates from linearity. This anomaly in $\chi^{-1}(T)$ at high temperature is likely due to in-plane exchange interaction, which survives in the PM state well above T_N for Sr_2IrO_4 [11]. With substitution, we also observe similar behavior in the PM state, though the linearity region of $\chi^{-1}(T)$

extends with x. The magnetic susceptibility, $\chi (= M/H)$, in the high-temperature PM state is fitted with the Curie-Weiss law,

$$\chi = \frac{C}{T - \theta_P},\tag{1}$$

where C is the Curie constant and θ_P is the Curie-Weiss constant. The extracted θ_P and the calculated effective PM moment $\mu_{\rm eff}$ from C are shown in Figs. 3(b) and 3(d) for Ru and Ti, respectively. A nearly vanishing and a constant θ_P over the series for Ru and Ti, respectively, agree with M(T) data in Figs. 3(a) and 3(c). We have further plotted the temperature derivative of magnetization, dM/dT, for selected x = 0 and 0.1 (Ru and Ti) in Fig. 4(b). Considering the inflection point in dM/dT as T_N , we have plotted the composition variation of T_N in the inset of Fig. 4(b) for both series. $T_N(x)$ shows a similar variation as for $\theta_P(x)$ (Fig. 3) for both series. The M(T), T_N , and θ_P in combination suggest an almost vanishing and constant transition temperature in Ru- and Ti-based series, respectively. The μ_{eff} , on the other hand, shows an initial increase and then saturation $(x \ge 0.2)$ for Ru substitution, while Ti shows an opposing trend. However, $x \sim 0.2$ marks a boundary above which the effects of substitution are found to be drastic in both series.

The magnetic and electronic state in Sr_2IrO_4 has been investigated using different substitutions both at the Sr- and



FIG. 2. X-ray photoemission spectra (XPS) are shown for the $Sr_2Ir_{1-x}(Ru/Ti)_xO_4$ series. Parts (a), (b), and (c) show the Ir-4*f* core-level spectra for x = 0.0, x (Ru) = 0.4, and x (Ti) = 0.4, respectively. Open black circles represent the experimental data, and red solid lines are a fitted envelope curve taking individual contributions of Ir^{4+} and Ir^{5+} components, which are plotted with blue and orange solid lines, respectively. Part (d) shows the Sr-3*d* core-level spectrum for x = 0.0. Parts (e) and (f) show the Ru-3*d* and Ti-2*p* core-level spectrum of the x = 0.40 sample for Ru and Ti substitution, respectively.

the Ir-site. At the Ir-site, mostly Rh, Ru, Cu, and Tb have been used, while La and Y are used at the Sr-site, which introduce a hole and electron doping accordingly. The Ir-site substitution, in addition, modifies the SOC and U in original material. While there remains a controversy between the Rh³⁺ $(4d^6)$ and Rh⁴⁺ $(4d^5)$ state, which has a role in band filling, the Rh generally causes a total suppression of a magnetic and insulating state around 17% of doping concentration [33–37]. Similarly, the $\operatorname{Ru}^{4+}(4d^4)$ substitution causes a total suppression of magnetic and insulating state in $Sr_2Ir_{1-x}Ru_xO_4$ with x around 0.5-0.6 [16-19]. A decoupling of magnetic and insulating states is shown with Tb^{4+} (4 f^7) in Sr₂Ir_{1-x}Tb_xO₄, where a complete suppression of magnetic state is observed with only 3% of Tb while retaining the insulating state [38]. In a recent study, we have shown that the Cu^{2+} (3 d^9) substitution causes a complete destabilization of a long-range magnetic state in $\text{Sr}_2 \text{Ir}_{1-x} \text{Cu}_x \text{O}_4$ with $x \sim 0.2$ [21]. The substitution of trivalent La^{3+}/Y^{3+} at the Sr-site, however, does not alter the basic interactions such as SOC and U, but it acts as electron doping. The La in $(Sr_{1-r}La_r)_2 IrO_4$ has shown a drastic effect where a complete suppression of the magnetic and insulating state has been observed with $x \leq 0.05$ [39,40]. The Y, on the

other hand, has shown a minimum influence on the groundstate properties in original material [22].

The suppression of a magnetic state in Sr₂IrO₄ with Ru⁴⁺ (S = 1) appears quite surprising, though the results are in line with other magnetic substitutions (Rh³⁺, Cu²⁺, Tb⁴⁺) where the total suppression of a magnetic state occurs at different concentration (discussed above) [21,34,38]. Given that the possible exchange interactions are $Ir^{4+}-O^{2-}-Ir^{4+}$, $Ir^{4+}-O^{2-}-Ru^{4+}$, and $Ru^{4+}-O^{2-}-Ru^{4+}$, the present results imply that is is unlikely that Ir-Ru interacts toward the original AFM interaction. Rather, with increasing concentration of Ru, the Ru-Ru interaction dominates, thus leading to a PM behavior in x = 0.6 that agrees with total Ru-based Sr_2RuO_4 [20]. The obtained $\mu_{eff} = 0.55 \,\mu_B/f.u.$ for x = 0.0appears much lower than the calculated spin-only value [= $g\sqrt{S(S+1)}\mu_B$]1.72 $\mu_B/f.u.$ for spin-1/2 material. The steep rise of μ_{eff} around x = 0.2 and then its saturation indicates the dominance of the Ru moment. As $Ti^{4+}(S = 0)$ acts for site dilution without participating in magnetic interaction, with increasing concentration it will create pockets of locally ordered Ir moments with different sizes. This will lead to a broadening of transition without much change in the onset of magnetic



FIG. 3. (a) The ZFC and FC magnetization data measured in a 10 kOe field are shown against temperature for $Sr_2Ir_{1-x}Ru_xO_4$ series. Part (b) shows the obtained effective PM moment μ_{eff} (left axis) and Curie-Weiss temperature θ_P (right axis) with Ru concentration; (c) and (d) show the same for $Sr_2Ir_{1-x}Ti_xO_4$ series.



FIG. 4. (a) Temperature-dependent inverse magnetic susceptibility, i.e., χ^{-1} vs *T*, is shown for representative samples of $\text{Sr}_2\text{Ir}_{1-x}(\text{Ru},\text{Ti})_x\text{O}_4$ series. Solid lines are due to Curie-Weiss law [Eq. (1)] fitting. Part (b) shows dM/dT vs *T* for representative samples, where the point of inflection is taken as T_N . The inset shows the composition variation of T_N for both series.



FIG. 5. Isothermal magnetization as a function of applied field up to ± 70 kOe collected at 5 K is shown for (a),(b) Ru and (c),(d) Ti series. Parts (e) and (f) show a moment at the highest applied field, i.e., μ_H , and a coercive field H_c for respective sample series.

transition temperature. This explains nearly constant T_N and θ_P and decreasing μ_{eff} in Ti series [Figs. 3(c) and 3(d)].

The magnetic-field-dependent magnetization M(H) at 5 K is shown in Figs. 5(a) and 5(b) for $Sr_2Ir_{1-x}Ru_xO_4$ series measured in a field range of \pm 70 kOe. The M(H) for Sr₂IrO₄ shows large hysteresis with a coercive field $H_c \sim 9370$ Oe. With Ru, the H_c decreases substantially, while for $x \ge 0.2$ the H_c is very minimal [Fig. 5(e)]. The moment μ_H at the highest field (70 kOe), however, shows anomalous behavior showing a dip around x = 0.2. Although this behavior of μ_H is consistent with M(T) above $x \sim 0.2$, the system turns out to be PM-like suppressing the AFM exchange, which promotes μ_H . In the case of Ti [Figs. 5(c) and 5(d)], an hysteresis loop with reasonable H_c exists until the highest value x = 0.6. Unlike Ru, the μ_H continue to decrease with x. As Ti only dilutes the magnetic lattice without promoting PM behavior, the reasonable AFM exchange survives until the highest x, which explains the variation of both H_c and μ_H [Fig. 5(f)].

VI. CHARGE-TRANSPORT STUDY

Both Ru and Ti have a contrasting effect on electron transport behavior in Sr_2IrO_4 , where the former suppresses



FIG. 6. Temperature-dependent resistivity is shown for (a) $\text{Sr}_2\text{Ir}_{1-x}\text{Ru}_x\text{O}_4$ and (b) $\text{Sr}_2\text{Ir}_{1-x}\text{Ti}_x\text{O}_4$ series. The inset of (a) shows the $\rho(T)$ with a metal-insulator-like transition for x = 0.6(Ru).

the insulating state and induces a metal-insulator transition (MIT) for the highest x = 0.6, while the latter substantially promotes the insulating state. The temperature-dependent resistivity $\rho(T)$ is shown in Figs. 6(a) and 6(b) for Ru and Ti series, respectively. Sr₂IrO₄ shows a semiconducting behavior with an increase in $\rho(T)$ of around four orders at low temperature. The charge-transport behavior follows a twodimensional Mott's variable-range-hopping (VRH) model $(\rho \propto \exp T^{-1/3})$, which is consistent with its layered structure consisting of Ir [13]. In contrast with 3d materials, 5d materials have extended orbitals (less U), which does not explain the insulating behavior in Sr₂IrO₄. Given that this material has a magnetic transition ($T_N \sim 225$ K) as well as an intermediate U, the insulating state has been described as either Mott-type (due to an interplay between SOC and U) or Slater-type (driven by magnetic ordering). Their coexistence has also been discussed both theoretically and experimentally [41–43]. Note that Ru (4 d^4) and Ti (3 d^0) have different d characters compared to Ir $(5d^5)$, hence their substitution will have a different impact on SOC, U, and band-filling. The Ru leads to a decreasing resistivity across the temperature, where the

TABLE II. The temperature range and fitting parameter T_0 obtained from fitting of the 2D VRH model in Fig. 7(a) are given for $Sr_2Ir_{1-x}Ru_xO_4, 0.0 \le x \le 0.10$, samples.

Sample $Sr_2Ir_{1-x}Ru_xO_4$	Temperature range (K)	T_0 (10 ⁵ K)
x = 0.0	300-240	1.44(1)
	240-70	0.46(8)
	40–5	0.04(8)
x = 0.05	300-130	2.82(8)
	130-30	1.26(2)
x = 0.10	300–20	1.22(2)

effect is very prominent for x > 0.2 giving a metal-insulator transition (MIT) for x = 0.6 around 90 K, which is probably due to a proximity of metallic behavior in Sr₂RuO₄ [44]. The Ti, on the other hand, depletes the *d*-band significantly, hence a highly insulating state is established [Fig. 6(b)]. Moreover, the nature of charge conduction is modified with two different substitutions.

In parent Sr₂IrO₄, the charge conduction mechanism follows the 2D Mott's VRH model [45], $\rho = \rho_0 \exp[(T_0/T)^{1/3}]$, where $T_0 (=21.2/[k_B N(E_F)\xi^3])$ is the characteristic temperature, k_B is the Boltzmann constant, $N(E_F)$ is the density of states (DOS) at the Fermi surface, and ξ is the localization length. Figure 7(a) shows the plotting of $\ln \rho$ versus $T^{-1/3}$, where the straight line fitting reveals the validity of the VRH model in a different temperature regime for x = 0.0, 0.05, and 0.1 in $Sr_2Ir_{1-x}Ru_xO_4$ series. The fitting parameters are given in Table II, showing that different temperature regimes are closely associated with the magnetic behavior in this material [13]. However, the charge conduction converts to power-law behavior ($\rho = \rho_0 T^{-n}$) for x = 0.2 and 0.4, which is confirmed by plotting $\ln \rho$ versus $\ln T$ in Fig. 7(b). Following MIT in x =0.6, the charge conduction in a high-temperature metallic state follows Fermi-liquid behavior, $\rho \propto T^2$, in an extended temperature range [Fig. 7(c)]. The charge-transport mechanism in $Sr_2Ir_{1-x}Ti_xO_4$, on the other hand, can be well explained with Mott's 2D VRH model for all samples. Figure 7(d) shows



FIG. 7. Part (a) shows $\ln \rho$ vs $T^{-1/3}$ plotted along with a straight line fitting following Mott's VRH model for x = 0.0, 0.05, and 0.1 of $\operatorname{Sr}_2\operatorname{Ir}_{1-x}\operatorname{Ru}_x\operatorname{O}_4$ series. Part (b) shows $\ln \rho$ vs $\ln T$ plotting following power-law behavior for x = 0.2 and 0.4. Part (c) shows ρ vs T^2 plotted following Fermi-liquid behavior for x = 0.6 of $\operatorname{Sr}_2\operatorname{Ir}_{1-x}\operatorname{Ru}_x\operatorname{O}_4$ series. Part (d) shows $\ln \rho$ vs $T^{-1/3}$ plotted for $\operatorname{Sr}_2\operatorname{Ir}_{1-x}\operatorname{Ti}_x\operatorname{O}_4$ series showing the validity of Mott's VRH model.

TABLE III. The temperature range and fitting parameter T_0 obtained from fitting of the 2D VRH model in Fig. 7(d) are given for $Sr_2Ir_{1-x}Ti_xO_4$ series.

Sample $Sr_2Ir_{1-x}Ti_xO_4$	Temperature range T (K)	T_0 (K) (10 ⁵)
x = 0.0	300–240	1.44(1)
	240-70	0.46(8)
	40–5	0.04(8)
x = 0.05	300-150	2.78(1)
	150-50	4.72(2)
	50-5	2.07(3)
x = 0.10	300-60	1.96(1)
	60-20	0.64(3)
x = 0.20	300-60	6.18(9)
x = 0.40	300-80	11.24(2)
x = 0.60	300-115	24.74(4)
x = 0.80	300-160	28.63(7)
x = 1.0	300–200	29.36(2)

reasonably good fitting until x = 0.6; the extracted fitting parameters are shown in Table III.

The isothermal resistivity as a function of magnetic field $(\pm 80 \text{ kOe})$ is measured at different temperatures for both series. Figures 8(a) and 8(b) show the $\rho(H)$ plot for Ru series at 300 K and at an intermediate temperature 150 K, respectively. The change in resistivity with field is not very significant, and a broken axis is used for better comparison of the data with a different composition. For parent Sr₂IrO₄, an increasing



FIG. 8. (a),(b) Magnetic field dependence of resistivity (ρ vs *H*) Sr₂Ir_{1-x}Ru_xO₄ series at 300 and 150 K, respectively, where $\rho(H)$ shows a switching from increasing to decreasing behavior with a magnetic transition. (c),(d) Similar plot for Sr₂Ir_{1-x}Ti_xO₄ series at 300 and 150 K, respectively, showing a decreasing $\rho(H)$ that extends until 300 K in doped samples due to weakening of both the magnetic state and SOC.

 $\rho(H)$ with a parabolic feature is observed in the PM state at 300 K, while in a magnetic state at 150 K, the $\rho(H)$ switches to a decreasing feature with a sharp cusp near H = 0. For Ru series, an increasing $\rho(H)$ is seen for all composition at 300 K, but similar switching to decreasing $\rho(H)$ has been observed with x, which suggests that this changeover in $\rho(H)$ behavior is related to the magnetic phase transition [Figs. 8(a)and 8(b)]. In contrast, there is significant weakening of both the magnetic state and SOC in Ti-doped series, therefore the $\rho(H)$ exhibits a decreasing behavior even up to 300 K in doped materials [Figs. 8(c) and 8(d)]. The cusp in $\rho(H)$ and its opposite evolution in both series are quite noteworthy, and they are often observed in materials with strong spin-orbit coupling showing weak localization or weak antilocalization. In the following, we present a detailed investigation about the effect of magnetic field on electronic charge transport in the present set of materials.

VII. MAGNETOCONDUCTANCE

The degree of disorder in an electron system contributing to charge localization is usually described using dimensionless parameter $g \equiv \sigma/(e^2/h)$, where $\sigma (= 1/\rho)$ is the conductivity and $h/e^2 (\approx 25.8 \text{ k}\Omega)$ is the resistance constant [46]. The materials with g > 1 and g < 1 are normally considered as weakly disordered with diffusive charge conduction (i.e., WL or WAL) and a strongly disordered (i.e., Anderson localization) system, respectively. In that sense, the comparatively less resistive regime in Sr₂IrO₄ [i.e., $\rho(T) < \sim h/e^2$ above $\sim 20 \text{ K}$] is likely to represent the diffusive charge conduction.

The calculated MC $[\Delta\sigma(B) = 1/\rho(B) - 1/\rho(0)]$ at different temperatures is shown in Fig. 9 in terms of $e^2/\pi h$ for $\text{Sr}_2\text{Ir}_{1-x}M_x\text{O}_4$ (M = Ru,Ti). For $\text{Sr}_2\text{Ir}\text{O}_4$, MC in a PM state (250 and 300 K) is negative, showing a quadratic field dependance ($\propto B^2$) that is likely due to the usual Lorentz contribution [Fig. 9(e)]. The fitting of Kohler's rule to magneto resistance data, $\Delta \rho / \rho(0) = [\rho(B) - \rho(0)] / \rho(0) = (\mu B)^2$, gives charge carrier mobility (μ) ~ 250 cm² V⁻¹ s⁻¹, which agrees with semiconducting materials [47]. On going to a magnetically ordered state below T_N , the MC interestingly makes a crossover to a positive value, indicating the promotion of charge conduction in the presence of magnetic field [Fig. 9(a)]. This positive MC data show a sharp cusp close to B = 0, which is considered to be typical of the WL effect. Upon lowering the temperature (Fig. 9), as σ decreases, the MC decreases and the cusp in the MC data is softened significantly. However, the cusp in MC below 20 K is not very clear, which is probably due to reduced conductivity at low temperature. Here, it can be mentioned that the observed positive MC dos not follow a B or B^2 dependence, which otherwise has been attributed to another type of QI effect in the hopping-dominated conduction regime [48,49].

The role of Ru and Ti on MC or WL behavior in the present SOC-dominated Sr₂IrO₄ is quite interesting. With drastic suppression of T_N using Ru [Fig. 3(a)], the negative MC is now observed in the extended PM regime. The cusp in positive MC below T_N is, however, no longer evidenced for $x \ge 0.2$, where the MC turns out to be parabolic-like [Figs. 9(b), 9(c) and 9(d)]. The B^2 dependence of MC data, for instance, has been shown for representative x = 0.2 at 100 K, which is well



FIG. 9. The magnetoconductivity $\Delta \sigma$ vs magnetic field at different temperatures is shown for selective samples in Sr₂Ir_{1-x}Ru_xO₄ (a)–(d) and Sr₂Ir_{1-x}Tu_xO₄ (f)–(h) series. Part (e) presents MC for x = 0.0 at 300 K and for x = 0.2 K at 100 K in Sr₂Ir_{1-x}Ru_xO₄ series, showing a parabolic dependence. The lines are due to B^2 fitting.

within the magnetic state [Fig. 9(e)]. This implies that with a weakening of the magnetic state in $\text{Sr}_2\text{Ir}_{1-x}\text{Ru}_x\text{O}_4$, the MC crossover between negative and positive T_N still resembles the parent $\text{Sr}_2\text{Ir}\text{O}_4$, but the WL properties cease to exist for $x \ge$ 0.2. The Ti substitution, on the other hand, shows contrasting behavior where, with increasing percentage, only positive MC is observed throughout the temperature range, even up to 300 K [Figs. 9(f), 9(g) and 9(h)]. This conversion from negative to positive MC in the PM state is believed to be due to the weakening of SOC and increased disorder with Ti^{4+} ($3d^0$) substitution. However, the cusps in MC at 250 and 300 K are largely softened. Nonetheless, the persistence of WL up to ~250 K signifies a nearly constant T_N in $\text{Sr}_2\text{Ir}_{1-x}\text{Ti}_x\text{O}_4$ as seen in M(T).

The effect of charge localization due to 2D MC has been theoretically described by the Hikami-Larkin-Nagaoka (HLN) equation, which considers scatterings due to the inelastic-dephasing (ϕ), spin-orbit (SO), and elastic (*e*) channel [50],

$$\Delta \sigma = -\frac{e^2}{2\pi h} \left[\Psi \left(\frac{1}{2} + \frac{B_{\phi}}{B} \right) - \ln \left(\frac{B_{\phi}}{B} \right) \right]$$
$$- \frac{e^2}{\pi h} \left[\Psi \left(\frac{1}{2} + \frac{B_{SO} + B_e}{B} \right) - \ln \left(\frac{B_{SO} + B_e}{B} \right) \right]$$
$$+ \frac{3e^2}{2\pi h} \left[\Psi \left(\frac{1}{2} + \frac{(4/3)B_{SO} + B_{\phi}}{B} \right) \right]$$
$$- \ln \left(\frac{(4/3)B_{SO} + B_{\phi}}{B} \right) \right], \qquad (2)$$

where *e* is the electronic charge, *h* is Planck's constant, $\Psi(x)$ is the digamma function, and $B_i (= \hbar/4eL_i^2)$ is the characteristic field related to each scattering channel, $i = \phi$, SO, and *e*. The L_{ϕ} , L_{SO} , and L_e are the phase coherence length, spinorbit scattering length, and elastic scattering length (mean free path), respectively. The dephasing magnetic field B_{ϕ} gives a rough estimate of the WL/WAL cusp [51], where L_{ϕ} signifies the lengthscale over which electrons maintain their phase coherence, and it usually dominates among the three characteristic lengths.

The positive MCs in Fig. 9 qualitatively suggest a WL behavior in these samples. The basic idea for charge localization (WL) due to weak disorder is coherent backscattering by impurities where, due to constructive interference between self-intersecting time-reversed electron paths, a phase is accumulated that amounts to $2\psi_0 \cos(\varphi/2)$, where ψ_0 is due to the phase accumulated in the spatial part, and φ is the angle accumulated in the spin part of the wave function. In a system without spin-momentum locking (as applies to WL), the φ is significantly small. This leads to the probability of backscattering $4|\psi_0|^2$, which eventually reduces the conductivity and causes the WL effect. In the case of WAL with strong SOC and spin-momentum locking, the φ accrues a π phase that then cancels the backscattering. The applied magnetic field breaks the time-reversal symmetry, hence two time-reversed paths acquire equal but opposite phases, $\beta =$ $e\Phi/\hbar$, with Φ the attached magnetic flux. This additional phase adds to the spin part, modifying the previous phase to $2\psi_0 \cos(\varphi/2 + \beta)$. As the phase β increases with magnetic field, the constructive interference (WL) will be changed,

which will increase the conductivity, hence a positive MC is realized.

As the time-reversed electron paths are self-intersecting, this implies that the amplitude of backscattering strongly depends on the dimensionality of material where the reduced dimensionality (1D or 2D) enhances the possibility of selfintersection. This WL/WAL is usually believed to be a small correction to the conductivity in the limit of weak disorder, however the effects become prominent in low-dimensional materials. Further, the magnetic field requirement is perpendicular to the charge conduction paths to maximize the flux. Therefore, the HLN equation [Eq. (2)] is best suited for 2D charge conduction with a perpendicular magnetic field. This equation has been used extensively in the case of topological insulators (TIs) with a distinct conducting surface and insulating bulk states [52]. In addition to thin films of TIs, the HLN equation has been successfully used to analyze the MC data in thick films as well as in bulk materials, where studies have indicated a strong coupling between surface and bulk states or a bulk mediated intersurface coupling, suggesting the charge conduction is not purely 2D in nature. In many cases, the used systems have a high resistance in the range of $k\Omega$ [53–58].

In the present study, the materials used are polycrystalline bulk, where the response to electrical transport measurements is orientation average. The parent Sr₂IrO₄ has an antiferromagnetic-type magnetic transition around 225 K [11–13], and it shows a Mott-type insulating state [10]. The Sr₂IrO₄ is structurally layered, where magnetically and electrically active Ir-O forms quasi-2D layers that are separated by inactive Sr-O layers [see Fig. 1(a)]. A significant anisotropy and nearly 2D magnetic interaction have been evidenced in different studies [7,11,59,60]. In fact, the signature of 2D charge conduction has been observed from analysis of $\rho(T)$ data (Fig. 7). Though the used materials are bulk, the 2Dlike character is very much inherent in its structure. Here, we mentioned that a relatively low MC and its sign change with temperature rules out the possibility of "spin-polarizedtunneling" driven MC, which is usually seen in polycrystalline materials due to grain boundaries [22,61,62]. All these suggest that the HLN equation [Eq. (2)], which is considered to describe the 2D MC due to charge localization, can be suitably used for the present set of materials.

The fitting of the HLN equation [Eq. (2)] to MC data is shown for Ru-based series with x = 0.0, 0.05, and 0.1 and for Ti-based series with x = 0.0, 0.05, 0.1, 0.2, 0.4, and 0.6 at representative 150 K in Figs. 10(a) and 11(a), respectively. As is evident in the respective figures, the fittings are reasonable good. Here, it can be mentioned that Eq. (2) is the original full HLN equation, while in many cases (in the limit of very strong and weak spin-orbit scattering) this equation is approximated by neglecting the spin-orbit and elastic terms and by inserting a prefactor α in place of 1/2 in the inelastic-dephasing term [first part in Eq. (2)]. The value of α is related to the number of conducting channels, where each channel contributes a value of 1/2. While the sign of prefactor α is intrinsically connected to WL/WAL behavior, its value has been used to understand the bulk-surface coupling or degree of disorder in different systems [46,53–56,63,64]. It has been known that a decreasing conductance in different disordered systems has given a low α . A theoretical study has shown that in low conducting



FIG. 10. (a) Magnetoconductivity $\Delta\sigma$ vs magnetic field for $\mathrm{Sr}_{2}\mathrm{Ir}_{1-x}\mathrm{Ru}_{x}\mathrm{O}_{4}$ with x = 0.0, 0.05, and 0.1 at representative 150 K. The solid lines are due to fitting with the HLN equation [Eq. (2)]. Parts (b), (c), and (d) show the obtained L_{ϕ} , L_{SO} , and L_{e} with temperature, respectively.

systems $(g \sim 1)$, the HLN equation is still valid to describe the MC data but the prefactor reduces as $\alpha = 1 - \frac{2}{\pi g}$ [63]. We note here that we checked our fittings by replacing 1/2 with α as a fitting parameter in the full HLN equation [Eq. (2)], but its value is around 1/2 without much change in fitting.

The extracted values of characteristic lengths (L_{ϕ} , L_{SO} , and L_e) are shown with temperature in Figs. 10 and 11 for Ru- and Ti-based materials, respectively. The L_{ϕ} [Figs. 10(b) and 11(b)], which is the lengthscale responsible for coherent scattering, decreases with lowering temperature, though for higher Ti-doped materials no clear pattern is observed. While the values of L_{ϕ} are quite reasonable at higher temperature, it decreases rapidly upon lowering the temperature, and at around 20 K L_{ϕ} almost vanishes. This rapid decrease of L_{ϕ} implies that the mechanism for inelastic scattering in the present materials increases drastically at low temperature. However, the fact that L_{ϕ} is much larger than the interlayer separation of Ir atoms in the unit cell [\sim 1 nm, Fig. 1(a)] suggests that the charge transport in the present systems is within the 2D quantum diffusion regime. In materials without magnetic ordering, the L_{ϕ} usually shows a $\propto T^{-n}$ dependence where the electron-electron interaction mainly contributes to dephasing at low temperature [65,66]. However, an opposite temperature dependence of L_{ϕ} for low-doped materials [Figs. 10(b) and 11(b)] suggests another type of inelastic scattering in the present materials. The onset of positive MC or WL behavior with magnetic ordering suggests magnetic scattering is the



FIG. 11. (a) Magnetoconductivity $\Delta\sigma$ vs magnetic field for $\text{Sr}_2\text{Ir}_{1-x}\text{Ti}_x\text{O}_4$ with x = 0.0, 0.05, 0.1, 0.2, 0.4, and 0.6 at representative 150 K. The solid lines are due to fitting with the HLN equation [Eq. (2)]. Parts (b), (c), and (d) show the obtained L_{ϕ} , L_{SO} , and L_e with temperature, respectively.

likely dephasing agent here. Magnetic scattering [67], which is considered as an interaction between the magnetic ions with conduction electrons, becomes stronger with decreasing temperature as the thermal demagnetization effect is minimized at lower temperature. Recent studies have shown a rapid suppression of both WL/WAL behavior and dephasing length with magnetic elements, which act as a dominating source of dephasing, in Fe-doped film of TI Bi₂Te₃ [51], and in TI-PM based heterostructure Bi₂Se₃-Co₇Se₈-Bi₂Se₃, where the magnetic Co²⁺ acts as a strong dephasing element at the interface [68]. However, for higher Ti doping [Fig. 11(b)], L_{ϕ} is significantly reduced and almost temperature-independent, which is primarily due to a weakened magnetic and conducting state. This is also evident in Fig. 11 as the cusp in the MC data becomes very broad for $x \ge 0.1$ of Ti. The evolution of L_{ϕ} with both temperature and doping in the present study is clearly governed by the magnetic and conducting state of the studied materials. L_{SO} , on the other hand, turns out to be very small and does not show any clear temperature dependence. L_{SO} , which roughly measures the strength of SOC, is usually not very prominent in the case of WL, although a high value has been seen in other materials [65,66]. This low value of L_{SO} is rather interesting as 5*d*-based Ir usually has high SOC. Nonetheless, one can speculate that the localized nature of the $J_{\rm eff}$ state may lead to such low $L_{\rm SO}$ in the present materials or iridates in general.

A negative and parabolic MC in the SOC-dominated PM regime, and then its crossover to a positive value (WL) with the onset of magnetic ordering, is quite noteworthy in the present Sr₂IrO₄. The tuning of WL/WAL and even their crossover were evidenced in magnetically doped as well as ultrathin TIs, which are caused by opening a gap at the Dirac point [46,51,64]. Such a WL/WAL crossover has further been shown in another Dirac material graphene, showing that it is driven by an intervalley scattering that reduces the chiral nature of Dirac fermions [69,70]. Further, a disorder-driven sign change and a crossover from anisotropic to isotropic MC data in films of topological materials have been discussed for future device application in terms of spin memory [71,72]. Iridates with sizable SOC and diverse magnetic/lattice structure are ideal to study the disorder-driven quantum transport behavior, akin to WL/WAL properties. Except for lowtemperature WAL in Na_2IrO_3 film [73], the iridates have not been explored yet in that direction. Note that a crossover in MC with temperature and field was shown for Sr₂IrO₄ film, however the MC data do not show any sharp cusp, and further the crossover is not connected with a magnetic transition [74,75]. It is rather interesting that 5d-based Sr_2IrO_4 has high SOC, and it still exhibits WL behavior, where the role of dimensionality needs to be investigated. Although the physics in iridates is quite different from that of TIs and graphene, the breaking of time-reversal symmetry appears to be the root cause for such exotic behavior. Further, it can be noted that WL in materials without strong SOC is distinctly different, where WL diminishes with the inclusion of a magnetic moment [76]. In that sense, the present report of SOC and magnetism-driven WL behavior in bulk iridates is quite noteworthy, and it should inspire similar experimental and theoretical studies.

VIII. CONCLUSION

In summary, we have demonstrated a crossover from negative to positive magnetoconductivity accompanied by weak localization behavior with an onset of magnetic ordering in layered Sr_2IrO_4 , which is mainly driven by a complex interplay between SOC and ordered moments. To understand this interplay in greater detail, the SOC as well as magnetic and electronic states in pure material are tuned with the substitution of Ru and Ti having different magnetic and electronic characteristics. In both cases, WL follows the evolved magnetic and conducting state, but Ti additionally converts the negative MC to a positive one above T_N with suppression of SOC. This tuning of quantum transport behavior with selective substitution through modification of inherent SOC and magnetism makes iridates promising materials for spintronic applications.

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- P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. 57, 287 (1985).
- [2] M. Brahlek, N. Koirala, N. Bansal, and S. Oh, Solid State Commun. 215-216, 54 (2015).
- [3] G. Cao and L. DeLong, *Physics of Spin-Orbit-Coupled Oxides* (Oxford University Press, Oxford, 2021).
- [4] T. Takayama, J. Chaloupka, A. Smerald, G. Khaliullin, and H. Takagi, J. Phys. Soc. Jpn. 90, 062001 (2021).
- [5] J. G. Rau, E. Kin-Ho Lee, and H.-Y. Kee, Annu. Rev. Condens. Matter Phys. 7, 195 (2016).
- [6] W. Witczak-Krempa, G. Chen, Y. B. Kim, and L. Balents, Annu. Rev. Condens. Matter Phys. 5, 57 (2014).
- [7] G. Cao, J. Bolivar, S. McCall, J. E. Crow, and R. P. Guertin, Phys. Rev. B 57, R11039(R) (1998).
- [8] Z. Y. Meng, Y. B. Kim, and H.-Y. Kee, Phys. Rev. Lett. 113, 177003 (2014).
- [9] Y. Yang, W.-S. Wang, J.-G. Liu, H. Chen, J.-H. Dai and Q.-H. Wang, Phys. Rev. B 89, 094518 (2014).
- [10] B. J. Kim, Hosub Jin, S. J. Moon, J.-Y. Kim, B.-G. Park, C. S. Leem, Jaejun Yu, T. W. Noh, C. Kim, S.-J. Oh, J.-H. Park, V. Durairaj, G. Cao, and E. Rotenberg, Phys. Rev. Lett. 101, 076402 (2008).
- [11] S. Fujiyama, H. Ohsumi, T. Komesu, J. Matsuno, B. J. Kim, M. Takata, T. Arima, and H. Takagi, Phys. Rev. Lett. 108, 247212 (2012).
- [12] F. Wang and T. Senthil, Phys. Rev. Lett. 106, 136402 (2011).
- [13] I. N. Bhatti, R. Rawat, A. Banerjee and A. K. Pramanik, J. Phys.: Condens. Matter 27, 016005 (2015).
- [14] M. K. Crawford, M. A. Subramanian, and R. L. Harlow, J. A. Fernandez-Baca, Z. R. Wang, and D. C. Johnston, Phys. Rev. B 49, 9198 (1994).
- [15] G. Jackeli and G. Khaliullin, Phys. Rev. Lett. 102, 017205 (2009).
- [16] R. J. Cava, B. Batlogg, K. Kiyono, H. Takagi, J. J. Krajewski, W. F. Peck, Jr., L. W. Rupp, Jr., and C. H. Chen, Phys. Rev. B 49, 11890 (1994).
- [17] S. A. Carter, B. Batlogg, R. J. Cava, J. J. Krajewski, W. F. Peck, Jr., and L. W. Rupp, Jr., Phys. Rev. B 51, 17184 (1995).
- [18] S. Calder, J. W. Kim, G.-X. Cao, C. Cantoni, A. F. May, H. B. Cao, A. A. Aczel, M. Matsuda, Y. Choi, D. Haskel, B. C. Sales, D. Mandrus, M. D. Lumsden, and A. D. Christianson, Phys. Rev. B 92, 165128 (2015).
- [19] S. J. Yuan, S. Aswartham, J. Terzic, H. Zheng, H. D. Zhao, P. Schlottmann, and G. Cao, Phys. Rev. B 92, 245103 (2015).
- [20] J. E. Ortmann, J. Y. Liu, J. Hu, M. Zhu, J. Peng, M. Matsuda, X. Ke, and Z. Q. Mao, Sci. Rep. 3, 2950 (2013).
- [21] I. N. Bhatti, R. S. Dhaka and A. K. Pramanik, Phys. Rev. B 96, 144433 (2017).
- [22] I. N. Bhatti, and A. K. Pramanik, J. Magn. Magn. Mater. 528, 167749 (2021).
- [23] I. N. Bhatti, and A. K. Pramanik, J. Magn. Magn. Mater. 422, 141 (2017).
- [24] I. N. Bhatti, and A. K. Pramanik, Phys. Lett. A 383, 1806 (2019).
- [25] Q. Huang, Jr., J. L. Soubeyroux, O. Chmaissem, I. Natali Sora, A. Santoro, R. J. Cava, J. J. Krajewski, W. F. Peck, Jr., J. Solid State Chem. 112, 355 (1994).
- [26] T. Shimura, Y. Inaguma, T. Nakamura, M. Itoh, and Y. Morii, Phys. Rev. B 52, 9143 (1995).

- [27] W. K. Zhu, M. Wang, B. Seradjeh, F. Yang, and S. X. Zhang, Phys. Rev. B 90, 054419 (2014).
- [28] J. M. Kahk, C. G. Poll, F. E. Oropeza, J. M. Ablett, D. Céolin, J.-P. Rueff, S. Agrestini, Y. Utsumi, K. D. Tsuei, Y. F. Liao, F. Borgatti, G. Panaccione, A. Regoutz, R. G. Egdell, B. J. Morgan, D. O. Scanlon and D. J. Payne, Phys. Rev. Lett. 112, 117601 (2014).
- [29] K. Maiti, and R. S. Singh, Sci. Lett. J. 3, 55 (2014).
- [30] B. N. Lin, C. Y. Lin, Y. S. Wu, and H. C. Ku, J. Magn. Magn. Mater. 272-276, 479 (2004).
- [31] C. Lin, A. Posadas, T. Hadamek, and Alexander A. Demkov, Phys. Rev. B 92, 035110 (2015).
- [32] T. Uozumi, K. Okada, A. Kotani, Y. Tezuka, and S. Shin, J. Phys. Soc. Jpn. 65, 1150 (1996).
- [33] T. F. Qi, O. B. Korneta, L. Li, K. Butrouna, V. S. Cao, X. Wan, P. Schlottmann, R. K. Kaul, and G. Cao, Phys. Rev. B 86, 125105 (2012).
- [34] J. P. Clancy, A. Lupascu, H. Gretarsson, Z. Islam, Y. F. Hu, D. Casa, C. S. Nelson, S. C. LaMarra, G. Cao, and Y.-J. Kim, Phys. Rev. B 89, 054409 (2014).
- [35] Y. Cao, Q. Wang, J. A. Waugh, T. J. Reber, H. Li, X. Zhou, S. Parham, S.-R. Park, N. C. Plumb, E. Rotenberg, A. Bostwick, and J. D. Denlinger, Nat. Commun. 7, 11367 (2016).
- [36] Y. Klein and I. Terasaki, J. Phys.: Condens. Matter 20, 295201 (2008).
- [37] V. Brouet, P. Foulquier, A. Louat, F. Bertran, P. Le Fèvre, J. E. Rault, and D. Colson, Phys. Rev. B 104, L121104 (2021).
- [38] J. C. Wang, S. Aswartham, Feng Ye, J. Terzic, H. Zheng, Daniel Haskel, Shalinee Chikara, Yong Choi, P. Schlottmann, Radu Custelcean, S. J. Yuan, and G. Cao, Phys. Rev. B 92, 214411 (2015).
- [39] X. Chen, T. Hogan, D. Walkup, W. Zhou, M. Pokharel, M. Yao, W. Tian, T. Z. Ward, Y. Zhao, D. Parshall, C. Opeil, J. W. Lynn, V. Madhavan, and S. D. Wilson, Phys. Rev. B 92, 075125 (2015).
- [40] H. Gretarsson, N. H. Sung, J. Porras, J. Bertinshaw, C. Dietl, Jan A. N. Bruin, A. F. Bangura, Y. K. Kim, R. Dinnebier, J. Kim, A. Al-Zein, M. Moretti Sala, M. Krisch, M. Le Tacon, B. Keimer, and B. J. Kim, Phys. Rev. Lett. **117**, 107001 (2016).
- [41] A. Yamasaki, S. Tachibana, H. Fujiwara, A. Higashiya, A. Irizawa, O. Kirilmaz, F. Pfaff, P. Scheiderer, J. Gabel, M. Sing *et al.*, Phys. Rev. B 89, 121111(R) (2014).
- [42] H. Watanabe, T. Shirakawa, and S. Yunoki, Phys. Rev. B 89, 165115 (2014).
- [43] Q. Li, G. Cao, S. Okamoto, J. Yi, W. Lin, B. C. Sales, J. Yan, R. Arita, J. Kuneš, A. V. Kozhevnikov *et al.*, Sci. Rep. **3**, 3073 (2013).
- [44] D. Pavuna, H. Berger, and L. Forro, J. Eur. Ceram. Soc. 19, 1515 (1999).
- [45] N. Mott, Conduction in Non-Crystalline Materials (Clarendon Press, Oxford, 1993).
- [46] J. Liao, Y. Ou, X. Feng, S. Yang, C. Lin, W. Yang, K. Wu, K. He, X. Ma, Q.-K. Xue, and Y. Li, Phys. Rev. Lett. 114, 216601 (2015).
- [47] D. Neumaier, K. Wagner, S. Geiβler, U. Wurstbauer, J. Sadowski, W. Wegscheider, and D. Weiss, Phys. Rev. Lett. 99, 116803 (2007).
- [48] L. Nguyen, B. Z. Spivak, and B. I. Shklovskii, Zh. Eksp. Teor. Fiz. 89, 1770 (1985).

- [49] U. Sivan, O. Entin-Wohlman, and Y. Imry, Phys. Rev. Lett. 60, 1566 (1988).
- [50] S. Hikami, A. I. Larkin, and Y. Nagaoka, Prog. Theor. Phys. 63, 707 (1980).
- [51] H.-T. He, G. Wang, T. Zhang, I.-K. Sou, G. K. L. Wong, J.-N. Wang, H.-Z. Lu, S.-Q. Shen, and F.-C. Zhang, Phys. Rev. Lett. 106, 166805 (2011).
- [52] M. Z. Hasan and C. L. Kane, Rev. Mod. Phys. 82, 3045 (2010).
- [53] J. Chen, H. J. Qin, F. Yang, J. Liu, T. Guan, F. M. Qu, G. H. Zhang, J. R. Shi, X. C. Xie, C. L. Yang, K. H. Wu, Y. Q. Li, and L. Lu, Phys. Rev. Lett. **105**, 176602 (2010).
- [54] N. Bansal, Y. S. Kim, M. Brahlek, E. Edrey, and S. Oh, Phys. Rev. Lett. **109**, 116804 (2012).
- [55] Y. S. Kim, M. Brahlek, N. Bansal, E. Edrey, G. A. Kapilevich, K. Iida, M. Tanimura, Y. Horibe, S.-W. Cheong, and S. Oh, Phys. Rev. B 84, 073109 (2011).
- [56] Z. Li, I. Garate, J. Pan, X. Wan, T. Chen, W. Ning, X. Zhang, F. Song, Y. Meng, X. Hong, X. Wang, Li Pi, X. Wang, B. Wang, S. Li, M. A. Reed, L. Glazman, and G. Wang, Phys. Rev. B 91, 041401(R) (2015).
- [57] J. G. Checkelsky, Y. S. Hor, M.-H. Liu, D.-X. Qu, R. J. Cava, and N. P. Ong, Phys. Rev. Lett. **103**, 246601 (2009)
- [58] G. Xu, W. Wang, X. Zhang, Y. Du, E. Liu, S. Wang, G. Wu, Z. Liu, and X. X. Zhang, Sci. Rep. 4, 5709 (2014).
- [59] Y. Hong, Y. Jo, H. Y. Choi, and N. Lee, Y. J. Choi and W. Kang, Phys. Rev. B 93, 094406 (2016).
- [60] S. Calder, D. M. Pajerowski, M. B. Stone, and A. F. May, Phys. Rev. B 98, 220402(R) (2018).
- [61] J. M. D. Coey, J. Appl. Phys. 85, 5576 (1999).
- [62] H. Y. Hwang, S.-W. Cheong, N. P. Ong, and B. Batlogg, Phys. Rev. Lett. 77, 2041 (1996).

- [63] G. M. Minkov, A. V. Germanenko, and I. V. Gornyi, Phys. Rev. B 70, 245423 (2004).
- [64] M. Liu, J. Zhang, C.-Z. Chang, Z. Zhang, X. Feng, K. Li, K. He, L.-L. Wang, X. Chen, X. Dai, Z. Fang, Q.-K. Xue, X. Ma, and Y. Wang, Phys. Rev. Lett. 108, 036805 (2012).
- [65] M. Meng, S. Huang, C. Tan, J. Wu, Y. Jing, H. Peng, and H. Q. Xu, Nanoscale 10, 2704 (2018).
- [66] X. Zhang, J. M. Woods, Judy J. Cha, and X. Shi, Phys. Rev. B 102, 115161 (2020).
- [67] G. Bergmann, Phys. Rep. 107, 1 (1984).
- [68] J. Lapano, A. R. Mazza, H. Li, D. Mukherjee, E. M. Skoropata, J. Mok Ok, H. Miao, R. G. Moore, T. Z. Ward, G. Eres, H. Nyung Lee, and M. Brahlek, APL Mater. 8, 091113 (2020).
- [69] E. McCann, K. Kechedzhi, Vladimir I. Falko, H. Suzuura, T. Ando, and B. L. Altshuler, Phys. Rev. Lett. 97, 146805 (2006).
- [70] F. V. Tikhonenko, A. A. Kozikov, A. K. Savchenko, and R. V. Gorbachev, Phys. Rev. Lett. **103**, 226801 (2009).
- [71] J. Reindl, H. Volker, N. P. Breznay, and M. Wuttig, npj Quantum Mater. 4, 57 (2019).
- [72] I. Korzhovska, H. Deng, L. Zhao, Y. Deshko, Z. Chen, M. Konczykowski, S. Zhao, S. Raoux, and L. Krusin-Elbaum, npj Quantum Mater. 5, 39 (2020).
- [73] M. Jenderka, J. Barzola-Quiquia, Z. Zhang, H. Frenzel, M. Grundmann, and M. Lorenz, Phys. Rev. B 88, 045111 (2013).
- [74] J. Ravichandran, C. R. Serrao, D. K. Efetov, D. Yi, Y. S. Oh, S.-W. Cheong, R. Ramesh and P. Kim, J. Phys.: Condens. Matter 28, 505304 (2016).
- [75] C. Lu, Shuai Dong, A. Quindeau, D. Preziosi, Ni Hu, and M. Alexe, Phys. Rev. B 91, 104401 (2015).
- [76] T. Dietl, J. Phys. Soc. Jpn. 77, 031005 (2008).