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Emergence of non-Fermi liquid behaviors in 5d perovskite SrIrO₃ thin films: Interplay between correlation, disorder, and spin-orbit coupling

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ABSTRACT

We investigate the effects of compressive strain on the electrical resistivity of 5d iridium based perovskite SrIrO₃ by depositing epitaxial films of thickness 35 nm on various substrates such as GdScO₃ (110), DyScO₃ (110), and SrTiO₃ (001). Surprisingly, we find anomalous transport behaviors as expressed by $\rho \propto T^\epsilon$ in the temperature dependent resistivity, where the temperature exponent ϵ evolves continuously from 4/5 to 1 and to 3/2 with an increase of compressive strain. Furthermore, magnetoresistance always remains positive irrespective of resistivity upturns at low temperatures. These observations imply that the delicate interplay between correlation and disorder in the presence of strong spin-orbit coupling is responsible for the emergence of the non-Fermi liquid behaviors in 5d perovskite SrIrO₃ thin films. We offer a theoretical framework for the interpretation of the experimental results.

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1. Introduction

Understanding the microscopic origin of non-Fermi liquid physics has been one of the long-standing problems in strongly correlated systems [1]. Although various mechanisms were provided to explain anomalous non-Fermi liquid transport phenomena, it is believed that for many strongly correlated systems the non-Fermi liquid behaviors are most likely triggered by the interplay of quenched disorder and strong electronic correlation [2]. This non-Fermi liquid phenomenology is rather ubiquitous near metal-insulator transitions [3]. Here, we wish to report non-Fermi liquid (NFL) behaviors over a wide temperature range near the metal-insulator transition (MIT) found in 5d element perovskite (Pv) SrIrO₃ thin films. We interpret that these NFL behaviors are the emerging result from a delicate interplay between correlation and disorder in the presence of strong spin-orbit coupling (SOC). Pv-SrIrO₃ would be an example of strong spin-orbit coupled correlated systems.

5d transition metal oxides (TMO) have become a topic of intense activities in condensed matter physics following the discovery of $J_{\text{eff}}=1/2$ Mott insulating ground state in Sr₂IrO₄ [4,5]. In fact, many intriguing properties such as correlated insulator, charge-density wave, Weyl semimetal and possible topological insulator have been studied in 5d TMOs by taking into account the

energy scale of large SOC [6]. Among 5d oxides, probably Rudlesden-Popper series Sr_{n+1}Ir_nO_{3n+1} ($n=1,2$, and ∞) have been the most investigated materials as they show a rich phase diagram involving MITs with increasing n [7]. With increasing n , i.e. increasing the number of IrO₂ planes, the bandwidth of the Ir 5d band becomes broader, and as a result Pv-SrIrO₃ ($n=\infty$) becomes a correlated semimetal [8–11]. This novel phenomenon is due to the interplay between local Coulomb interaction (U) and strong SOC as the strength of SOC can be as large as 0.3~0.5 eV in 5d oxides ($\text{SOC} \propto z^4$ where z is the atomic number), comparable to the corresponding bandwidth or Coulomb interaction, and thus play a decisive role in the physics of 5d oxides [6].

From the viewpoint of crystal structures, the ground state of SrIrO₃ at room temperature and atmospheric pressure is the structure of hexagonal BaTiO₃ [12]. Studies on hexagonal SrIrO₃ single crystals revealed that it is a metallic system exhibiting NFL behaviors; the specific heat displays a typical logarithmic divergence and the electrical resistivity follows $T^{3/2}$ [13]. This NFL physics has been attributed to quantum criticality. Pv-SrIrO₃ can be obtained only at elevated pressure (40 kbar) and temperature (1000 °C) and is a paramagnetic metal [12,14,15]. In fact, Pv-SrIrO₃ is supposed to be a correlated bad metal, i.e., mean free path comparable to the inter-atomic distance, and is presumably close to a metal-insulator phase boundary [7]. Thus, if Pv-SrIrO₃ can be stabilized at room temperature, it would open up a possibility of investigating new emergent phenomena because Pv-SrIrO₃ would be susceptible to external perturbations due to its vicinity to MIT.

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In order to obtain Pv-SrIrO₃ and also to induce new physical phenomena different from bulk properties, we resorted to thin film technology, where underlying substrates help stabilize the perovskite phase. We demonstrate that Pv-SrIrO₃ thin films display NFL behaviors and, more surprisingly, with an increase of compressive strain, the temperature dependence of the resistivity as expressed $\rho \propto T^\epsilon$ evolves from that with $\epsilon=4/5$ to 1 and to $3/2$, due to a subtle interplay between correlation, disorder, and SOC.

2. Materials and methods

While bulk Pv-SrIrO₃ is metastable at room temperature, the metastable phase can be stabilized with thin film growth, and, in addition, key parameters of Pv-SrIrO₃ such as band width and correlation can be tuned with strained thin films by changing underlying substrates. We have grown Pv-SrIrO₃ thin films (typical thickness ~ 35 nm) on various lattice mismatched substrates such as GdScO₃ (110), DyScO₃ (110), and SrTiO₃ (001) [9]. We used pulsed laser deposition (KrF laser with $\lambda=248$ nm) to grow SrIrO₃ thin films from a polycrystalline target. The target was prepared by a solid-state reaction method; stoichiometric mixing of SrCO₃ and IrO₂ raw powders is followed by sintering at 1000 °C for 48 h. The laser was operated at frequency 4 Hz, and the substrate temperature and oxygen partial pressure were 550 °C and 20 mTorr, respectively. The target to substrate distance was kept ~ 50 mm. After growth, all the films were annealed at the same oxygen partial pressure and temperature to compensate for any oxygen deficiency. Crystalline quality of the films was checked by X-ray diffraction (XRD) measurements; XRD was carried out with the Empyrean XRD System from PANalytical. Four-probe van der Pauw geometry was used for electrical transport measurements. For magnetoresistance measurements, the applied magnetic field strength was $-9 \text{ T} \leq B \leq 9 \text{ T}$ in the out-of-plane direction.

3. Results and discussion

Structurally, bulk Pv-SrIrO₃ is of orthorhombic structure ($a=5.60 \text{ \AA}$, $b=5.58 \text{ \AA}$, $c=7.89 \text{ \AA}$) [12]. Note that GdScO₃ (110) and DyScO₃ (110) substrates are also orthorhombic and orthorhombic indices are typically used to indicate their orientation. SrTiO₃ (001) substrates, on the other hand, are cubic. For thin films on substrates, pseudo-cubic indices are preferred for easy comparison. If the *pseudo-cubic* (a_{pc}) lattice parameter is converted from the orthorhombic lattice parameters of bulk Pv-SrIrO₃, we obtain $a_{pc} \sim 3.96 \text{ \AA}$. This would match very well GdScO₃ substrates with $a_{pc} \sim 3.96 \text{ \AA}$, correspond to $+0.50\%$ compressive strain for the films on DyScO₃ substrates with $a_{pc} \sim 3.94 \text{ \AA}$, and $+1.54\%$ compression for those on SrTiO₃ substrates with $a_{pc} \sim 3.90 \text{ \AA}$ as illustrated in Fig. 1a. The epitaxial nature of the perovskite thin films were confirmed by XRD measurements. The XRD analysis (θ - 2θ scan) of the Pv-SrIrO₃ films grown on aforementioned substrates show crystalline peaks without any impurity or additional peaks; for clarity, only the low angle data including the (001)_{pc} peak are shown in Fig. 1b. The films on all the substrates exhibit clear layer thickness fringes (results of coherent scattering from a finite number of lattice planes with thickness of the film), showing the surface smoothness as well as high crystalline quality. AFM characterization of the film surfaces also confirmed that they were nearly flat with maximum roughness of 1.5 nm (not shown here). From Fig. 1b, it is noted that with an increasing lattice mismatch, the 2θ value of the film peak maximum decreases and thus the film's out-of-plane lattice constant increases correspondingly. This fact indicates the in-plane film lattice constant is locked with that of the underlying substrate and indeed the films are under compression

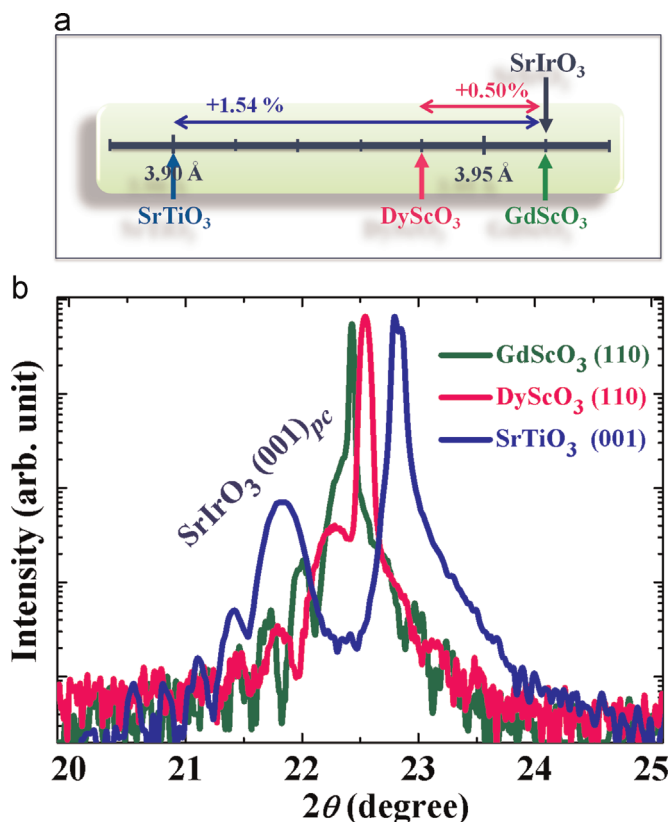


Fig. 1. (a) *Pseudo-cubic* (or cubic) lattice constants of perovskite SrIrO₃ and substrates GdScO₃ (110), DyScO₃ (110), and SrTiO₃ (001). Corresponding amount of compressive strain is also shown. (b) X-ray θ - 2θ scan of epitaxial SrIrO₃ thin films of 35 nm thickness grown on three different substrates. Bragg peaks and thickness fringes are seen. Only low angle *pseudo-cubic* (001)_{pc} peaks are shown for clarity.

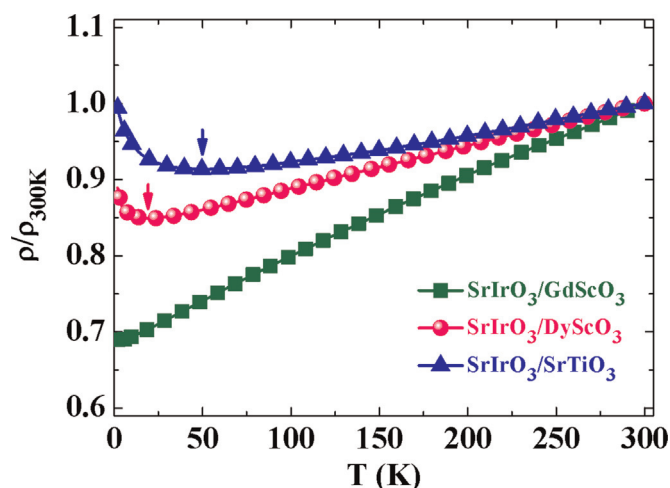


Fig. 2. Temperature dependent electrical resistivity of epitaxial SrIrO₃ thin films grown on three different substrates. With increasing compressive strain, resistivity at $T=300 \text{ K}$ increases. Best lattice-matched film on GdScO₃ shows a fully metallic behavior down to lowest temperature. For the strained films, low temperature upturns in resistivity ($T=20 \text{ K}$ for films on DyScO₃ and $T=50 \text{ K}$ for films on SrTiO₃) are indicated by arrow.

[9].

Having confirmed the epitaxial quality, we then measured resistivity (ρ) of the Pv-SrIrO₃ films grown on various lattice-mismatched substrates as shown in Fig. 2. The film on the best lattice-matched substrate GdScO₃ shows $\rho \sim 1.45 \text{ m}\Omega \text{ cm}$ at $T=300 \text{ K}$, which is much smaller than that of the bulk polycrystalline Pv-SrIrO₃ $\rho \sim 4 \text{ m}\Omega \text{ cm}$ at $T=300 \text{ K}$ [12]. Obviously, the resistivity of

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