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Signature of a Griffiths phase in layered canted antiferromagnet Sr_2IrO_4

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ABSTRACT

The complex magnetic nature of 5d layered iridate Sr_2IrO_4, owing to IrO_6 octahedral rotation has triggered great interest in recent years. In this article, we investigated the magnetic excitations in layered canted antiferromagnet Sr_2IrO_4 via dc magnetization measurements and report the Griffiths phase (GP) signatures above the magnetic ordering temperature T_N. The non-analytic nature of GP leads to unique critical exponent, $\beta = 0.75(1)$ extracted from modified Arrott plot, in corroboration with magneto-caloric study. However, the analysis by Bray model in GP regime yields a reliable critical exponent value of $\beta = 0.18(1)$, belonging to two-dimensional XYh_4 universality class. The study also suggests a new picture of largely debated insulating nature of Sr_2IrO_4 in context of GP above T_N.

1. Introduction

The physics of 5d transition metal oxides (TMOs) has been widely investigated recently owing to their inherently large spin-orbit coupling (SOC) competing with other energy scales i.e. electronic correlations, Hund's coupling, etc. The 5d TMOs studies were mainly initialized by pioneering experiments on layered iridate Sr_2IrO_4, claiming unconventional spin-orbital Mott state [1] and "isotropic" two-dimensional (2D) quantum pseudospin-1/2 (j = 1/2) Heisenberg antiferromagnetic (2DQHAFM) excitations [2], despite weak electronic correlations and large SOC, respectively in 5d TMOs. Nevertheless, these unconventional observations make Sr_2IrO_4 analogous to novel layered cuprates [3]. Recent theoretical calculations also show high-temperature superconductivity (HTSc) in doped-Sr_2IrO_4 with optimal Hund's coupling [4]. The Sr_2IrO_4 has a layered structure, similar to cuprate La_2CuO_4, however with IrO_6 octahedral rotation of $\sim 11^0$ around c-axis [5]. This rotation breaks in-plane inversion symmetry and the system adopts an in-plane canted AFM structure below $T_N \sim 225$ K [6], whereas along c-axis, the weakly AFM coupled IrO_2 planes exhibit a ferromagnetic (FM) ordering in a weak external magnetic field of $H_c \sim 2$ kOe [1]. Moreover, the neutron diffraction [6,7], X-ray resonant magnetic scattering (XRMS) [8] and muon spin spectroscopy (μSR) [9] techniques suggest anisotropic 2D XY interactions through critical exponent study. Furthermore, the electronic nature of Sr_2IrO_4 is also not clear. Recent studies even question the believed Mott state in Sr_2IrO_4 and suggest Slater behavior [10,11]. Despite these complications, recently observed HTSc signatures in electron-doped Sr_2IrO_4 [12] trigger reinvestigation of magnetism and insulating nature in Sr_2IrO_4.

More surprisingly, the XRMS studies on Sr_2IrO_4 show the persistence of in-plane magnetic correlations between j = 1/2 spins in paramagnetic (PM) state [8,13]. Furthermore, the Raman study [14] reveals the presence of two-magnon scattering well above T_N indicating strong pseudospin excitations in PM state. These observations suggest the existence of short-range magnetic clusters in a PM matrix, which can happen due to presence of *Griffiths singularity* [15–17]. In such scenario, the magnetic system breaks into different sized short-range magnetic clusters with a distribution of ordering temperature T_N (p) depending upon the intrinsic disorder (p) and the long-range magnetic ordering is stabilized at much lower temperature T_N rather than at T_G, the Griffiths temperature, for a perfectly ordered system. The intermediate regime between T_N and T_G is referred as Griffiths phase (GP).

Here, we report the Griffiths phase signatures in Sr_2IrO_4 via detailed *dc* magnetization measurements. The non-analytic form of free energy in a Griffiths system [15] causes unrealistic critical exponents through conventional modified *Arrott* plot (MAP) method. We utilize Bray model to investigate the critical behavior of Sr_2IrO_4 and further correlate its unconventional insulating nature with GP above T_N.

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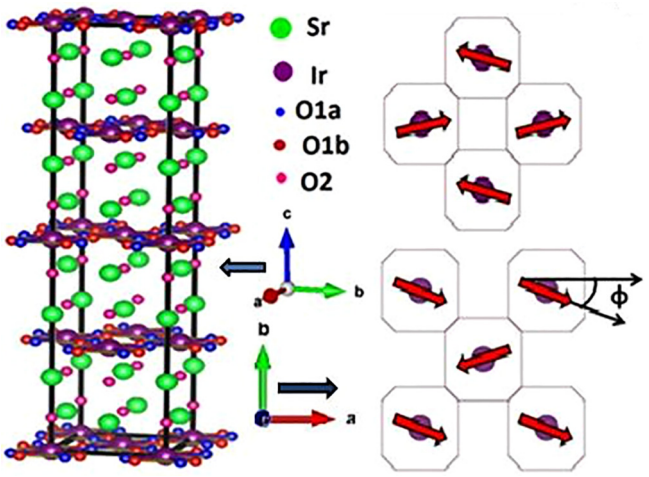

Fig. 1. The layered perovskite crystal structure (left panel) and the schematic magnetic structure (right panel) of Sr_2IrO_4, showing clockwise and anticlockwise rotation of alternate basal (ab-) planes around c-axis.

2. Experimental

The Sr_2IrO_4 polycrystalline sample was synthesized through solid state reaction method. The starting oxides $SrCO_3$ (99.99% purity) and IrO_2 (99.99% purity) were mixed in stoichiometric ratio. The mixture was heated at 800, 900 and 1000 °C for 12 h each with intermediate grindings. Finally, the mixture was pressed into pellet and sintered at 1000 °C for 24 h. The room-temperature X-ray diffraction measurement confirms the single tetragonal phase (space group $I4_1/acd$) with Rietveld refined lattice parameters, a = b = 5.495 Å and c = 25.773 Å, which are in close agreement with earlier report on Sr_2IrO_4 [5]. Furthermore, high resolution transmission electron microscopy (HRTEM) micrographs clearly reveal the randomly-oriented crystallites of Sr_2IrO_4 asserting high crystallinity of the sample [see Fig. S1 in the Supplementary material]. The layered crystal structure and (schematic) magnetic structure, generated from the Rietveld fit, are shown in Fig. 1, where the alternate basal (ab) planes rotate by $\phi \sim 11^\circ$ around c-axis, in clockwise and anticlockwise direction, respectively. The temperature and field-dependent dc magnetization were measured using SQUID magnetometer, whereas the magnetic isotherms were recorded in the vicinity of critical temperature using a physical property measurement system (PPMS-9T, Quantum Design).

3. Result and discussion

3.1. dc magnetization

Fig. 2 shows the temperature-dependent dc magnetization, $M(T)$ of Sr_2IrO_4 measured in the field-cooled (m^{FC}) and zero-field-cooled (m^{ZFC}) modes at 100 Oe (a) and 10 kOe (b). The $M(T)$ at 100 Oe shows the magnetic ordering at $T_N \sim 221$ K, as determined from dip in thermal derivative plot of magnetization, which is in close agreement with earlier report on Sr_2IrO_4 [6,8]. On cooling, the large bifurcation between m^{FC} and m^{ZFC} curves along with a broad maxima in m^{ZFC} curve suggest a large magnetic anisotropy and freezing of weakly magnetic domains at low fields. However, this bifurcation is largely suppressed to low temperatures in a high-field of 10 kOe [Fig. 2b], which indicates a large reduction in magnetic anisotropy in the system. Taking account of weak temperature-independent van-vleck susceptibility contribution in Iridates [18], the high temperature m^{FC} is fitted with modified Curie-Weiss (CW) equation, $\chi_T = \chi - \chi_0 = C/(T - \theta_{CW})$, which gives $\theta_{CW} = 165.7$ K and $\mu_{eff} = 0.68\,\mu_B$ with $\chi_0 = 2 \times 10^{-4}$ emu/mole at 100 Oe [see inset of Fig. 2a]. The positive θ_{CW} value, contrary to what expected for a conventional AFM, proves a significant canting of in-plane spins, whereas the obtained μ_{eff} value is much smaller than the theoretical $\mu_{eff} = 1.73\,\mu_B$ with $g_J = 2$ for $J_{eff} = 1/2$ state due to strong hybridization between Ir 5d and O 2p orbitals, but still larger than $0.5\,\mu_B$ obtained for single crystal studies [19,20]. More importantly, the $1/\chi_T$ (T) is not fully described by CW behavior down to T_N, but shows negative downturn at much higher temperature, $T_G \sim 285$ K. Furthermore, this downturn in $1/\chi_T$ disappears at high field of 10 kOe, as shown in inset of Fig. 2b. These observations suggest the presence of short-range AFM ordering (or equivalently, finite spin-canted clusters sustained by a strong basal-plane anisotropy) in microscopically phase-separated regions much above the expected bulk T_N. These are the signatures of Griffiths singularity [15], as previously observed in 3d counterpart single-layered perovskite $LaSrCoO_4$ [21] and other magnetic systems [22,23]. Here, we want to point out that the χ_T (T) above T_N for 1 kOe has been alternatively explained taking into account the interlayer couplings [24], however, the study does not explain the violation of CW behavior at lower fields. The possible GP, its origin and impact on Sr_2IrO_4 insulating nature have been discussed later on.

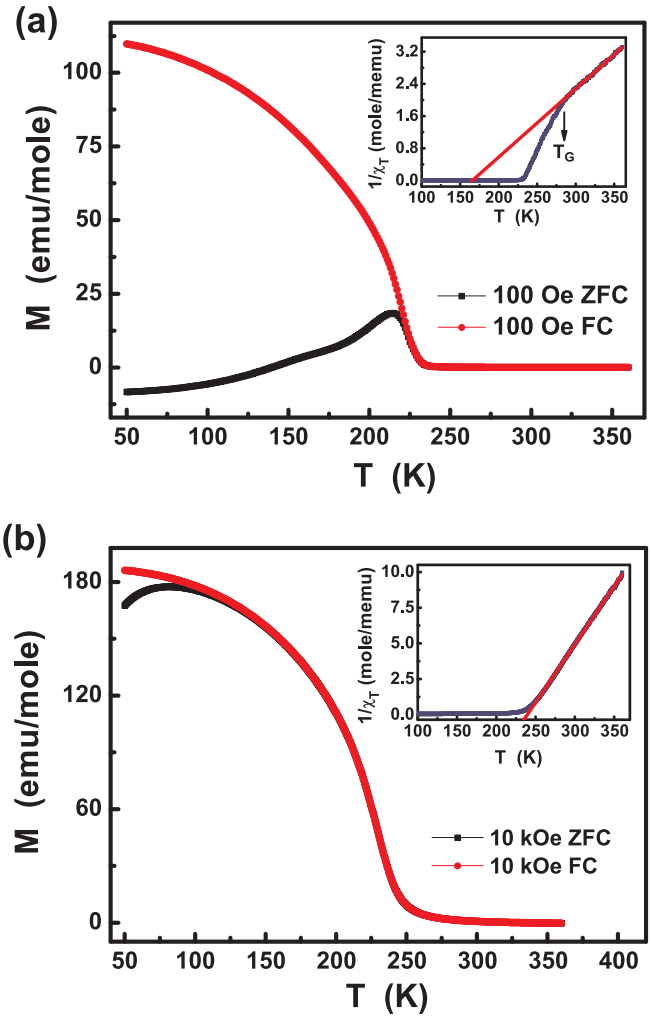

Fig. 2. The temperature dependence of dc magnetization, $M(T)$ measured in field-cooled (m^{FC}) and zero-field-cooled (m^{ZFC}) modes at 100 Oe (a) and 10 kOe (b). The insets shows $1/\chi_T$ (T) plot along with Curie-Weiss law fit (red line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

3.2. Critical phenomena

Next, we investigate the magnetic interactions in Sr_2IrO_4 through critical exponent study using $M(H)$ isotherms in the vicinity of