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# Anisotropic antiferromagnetic order in spin-orbit coupled trigonal lattice $\text{Ca}_2\text{Sr}_2\text{IrO}_6$

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We used single-crystal X-ray and neutron diffraction to investigate the crystal and magnetic structures of trigonal lattice iridate  $\text{Ca}_2\text{Sr}_2\text{IrO}_6$ . The crystal structure is determined to be  $R\bar{3}$  with two distinct Ir sites. The system exhibits long-range antiferromagnetic order below  $T_N = 13.1$  K. The magnetic wavevector is identified as  $(0, 0.5, 1)$  with ferromagnetic coupling along the  $a$  axis and antiferromagnetic correlation along the  $b$  axis. Spins align dominantly within the basal plane along the  $[1, 2, 0]$  direction and tilt  $34^\circ$  towards the  $c$  axis. The ordered moment is  $0.66(3) \mu_B/\text{Ir}$ , larger than other iridates where iridium ions form corner- or edge sharing  $\text{IrO}_6$  octahedral networks. The tilting angle is reduced to  $\approx 19^\circ$  when a magnetic field of 4.9 Tesla is applied along the  $c$  axis. Density functional theory calculations confirm that the experimentally determined magnetic configuration is the most probable ground state with an insulating gap  $\sim 0.5$  eV.

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## INTRODUCTION

Controlling the balance between spin-orbit interactions (SOI), on-site Coulomb interactions and crystalline electric field splitting in  $5d$  iridates is the central theme behind searching for novel quantum phenomena such as  $j_{\text{eff}} = 1/2$  Mott insulating states [1–3], correlated topological insulators [4, 5], spin liquid phases [6], superconductivity [7, 8], and Kitaev models [9–11]. Due to the entangled spin and orbital degrees of freedom, the form of magnetic interactions is no longer dictated by a global spin  $SU(2)$  symmetry. This leads to physics that is dramatically different from the  $3d$  systems where SOI is of a perturbative nature. The wave functions are composed by the superposition of different orbital and spin states and the resulting magnetic interactions depend critically on the lattice symmetry. In the case of a  $180^\circ$  Ir-O-Ir bond, the Hamiltonian is governed by an isotropic Heisenberg term plus a weak dipolar-like anisotropy term due to the Hund's coupling. While for a  $90^\circ$  bond, the anisotropic term due to the off-diagonal hopping matrix results in a quantum analog of the compass model [9]. The strong SOI limit also assumes local cubic symmetry of the  $\text{IrO}_6$  octahedra, which is rare in real materials. It was discovered that nearly all iridate families have a certain degree of noncubic distortions. For example, the O-Ir-O bond angle in pyrochlores  $R_2\text{Ir}_2\text{O}_7$  ( $R$ : rare earth) is an average  $6^\circ$ – $10^\circ$  away from  $90^\circ$  [12]; a substantial elongation (tetragonal distortion) of the  $\text{IrO}_6$  octahedra occurs in the  $j_{\text{eff}} = 1/2$  Mott insulator  $\text{Sr}_2\text{IrO}_4$  [13]; an appreciable trigonal distortion was revealed in honeycomb lattice  $\text{Na}_2\text{IrO}_3$  with O-Ir-O bond angles  $\sim 4 - 9^\circ$  deviating from the cubic case [14, 15]. Although it was claimed that the  $j_{\text{eff}} = 1/2$  state is robust against distortion, re-

cent resonant inelastic X-ray scattering (RIXS) studies have shown that the distortion of  $\text{IrO}_6$  octahedra leads to modification of the isotropic wave functions [16, 17]. This underscores the need for extending the  $j_{\text{eff}} = 1/2$  picture to correctly describe the Mott insulating ground states.

In this paper, we report a single crystal X-ray and neutron diffraction investigation of a trigonal lattice iridate  $\text{Ca}_2\text{Sr}_2\text{IrO}_6$  (CSIO). The crystal orders antiferromagnetically (AFM) below 13.1 K with no structural anomaly across the transition. The wavevector of the spin structure is  $(0, 0.5, 1)$  indicating strong anisotropic magnetic interactions. The iridium moments align nearly along the diagonal O-Ir-O direction within the  $\text{IrO}_6$  octahedra. The ordered moment reaches  $0.66(3) \mu_B/\text{Ir}$ , larger than other iridates which form corner- or edge sharing  $\text{IrO}_6$  octahedral networks. Most importantly, the local environment of  $\text{IrO}_6$  is close to the cubic limit and there is no direct connectivity between individual  $\text{IrO}_6$  octahedra, making this system a canonical candidate to study the novel magnetism arising from the SOI.

## EXPERIMENTAL RESULTS

Single crystals of CSIO were grown using a self-flux method similar to the one reported in Ref. [18], from off-stoichiometric quantities of  $\text{IrO}_2$ ,  $\text{CaCO}_3$  and  $\text{SrCO}_3$  that were mixed with  $\text{CaCl}_2$  and/or  $\text{SrCl}_2$ . The starting ratio of Ir to (Ca, Sr) is approximately 1:5. The mixed powders were fired to  $1460^\circ\text{C}$  for 4 hours and then slowly cooled at a rate of  $4^\circ\text{C}/\text{hour}$ . The compositions were independently checked to be consistent using both energy dispersive X-ray analysis (EDX) (Hitachi/Oxford 3030 Plus) and single-crystal X-ray diffraction. The mag-

netic susceptibility and specific heat were measured using a Quantum Design Magnetic Property Measurement System. X-ray diffraction data were collected using a Rigaku XtaLAB PRO diffractometer at the Oak Ridge National Laboratory (ORNL). A molybdenum anode was used to generate X-ray with wavelength  $\lambda = 0.7107$  Å. Neutron diffraction measurement was carried out using the TOPAZ diffractometer with crystal size of  $1 \times 1 \times 1.5$  mm<sup>3</sup> at Spallation Neutron Source (SNS), ORNL. A larger piece with dimension of  $1.5 \times 1.5 \times 4$  mm<sup>3</sup> was chosen for magnetic structure determination using the single crystal diffuse scattering diffractometer CORELLI at SNS [19]. The  $\pm 28.5^\circ$  vertical angular coverage of the detector allows an extensive survey in reciprocal space. A 5 Tesla vertical field superconducting magnet was used to study the field evolution of the spin structure.

Pure and Sr-doped Ca<sub>4</sub>IrO<sub>6</sub> were reported to crystallize in a rhombohedral, K<sub>4</sub>CdCl<sub>6</sub>-type structure with  $R\bar{3}c$  space group (SG No. 167) from X-ray powder diffraction studies [20]. The lattice parameters increase monotonically with Sr doping. The values become  $a = b = 9.588$  Å and  $c = 11.414$  Å for CSIO at room temperature. The crystal structure in Fig. 1(a) shows the one-dimensional (1D) chains of alternating IrO<sub>6</sub> octahedra and CaO<sub>6</sub> trigonal prisms parallel to the  $c$  axis. The single crystal X-ray diffraction measurement on CSIO reveals that the majority of reflections are consistent with the reported  $R\bar{3}c$  space group, with a significant portion of peaks violating the reflection conditions [345 out of 3152 reflections with  $I > 3\sigma(I)$ ]. To further confirm the finding, we employed single crystal neutron diffraction to characterize the structure. Fig. 1(b) presents a typical contour plot in the  $(h, k, l = 1)$  scattering plane. Indeed, several marked reflections cannot be indexed using SG 167, which requires both  $h + l = 3n$  and  $l = 2n$  in the  $(hkl)$  scattering plane. The presence of (4,0,1) in Fig. 1(b) clearly indicates the breakdown of the reflection condition and suggests a reduced crystal structure symmetry. Based on the X-ray and neutron observation, the maximal non-isomorphic subgroup  $R\bar{3}$  (No. 148) that lacks the  $c$ -axis glide is the most likely space group, where the unique Ir site ( $6b$  site in SG 167) splits into  $3a$  and  $3b$  Wyckoff positions. Such different surrounding oxygen environments allow the two Ir sites to have independent spin orientations. Each IrO<sub>6</sub> octahedron contains six identical Ir-O bond length of 2.036 Å. There is a small trigonal distortion with the octahedron stretched along the  $c$  axis, the corresponding O-Ir1-O and O-Ir2-O bond angles at 100 K are  $88.92^\circ$  and  $88.68(8)^\circ$ , respectively. The overall local environment surrounding the Ir atoms is close to the ideal cubic limit. Furthermore, the alkaline earth atoms connecting neighboring IrO<sub>6</sub> along the  $c$  axis are dominated by Ca ions while the sites between the IrO<sub>6</sub>/CaO<sub>6</sub> chains have mixed Sr:Ca ions with a 2:1 ratio [Figure 1(a)]. This atomic arrangement is probably due to the longer distance from the mixed site to the oxy-

gen atoms, which is more suitable to host the larger Sr ions. This feature also agrees with a 2.7% increase in  $a$ , but only 1.6% increase in  $c$  from Ca<sub>4</sub>IrO<sub>6</sub> to CSIO. It is noteworthy that the formation of two distinct octahedral sites is not common in the K<sub>4</sub>CdCl<sub>6</sub>-type compounds. The observation of  $R\bar{3}$  space group in CSIO might be related to the preferred site occupancy of the mixed Ca/Sr ions. It is certainly interesting to verify whether this is the case in similar material with  $R\bar{3}$  space group discovered in the future.

Figure 1(c) shows the  $T$  dependence of specific heat of CSIO single crystal. A sharp anomaly appears near 13 K indicating a magnetic transition similar to pure Ca<sub>4</sub>IrO<sub>6</sub> [18, 21]. Fig. 1(d) shows the temperature evolution of the magnetic susceptibility  $\chi$  with applied magnetic field of 0.5 T. The peaks observed around 13.5 K confirm the phase transition. Fits of  $1/\chi_{ab}$  ( $1/\chi_c$ ) for  $30 \text{ K} < T < 300 \text{ K}$  to a Curie-Weiss law yield effective magnetic moments  $\mu_{\text{eff}}$  of 1.25 (2.14)  $\mu_B$  and Curie temperatures  $\theta_{CW}$  of -0.13 (-12.69) K for the field applied in the basal plane (parallel to the  $c$  axis). The negative value in  $\theta_{CW}$  implies AFM interactions between the neighboring Ir ions. The large difference in both  $\mu_{\text{eff}}$  and  $\theta_{CW}$  within the  $ab$  plane and along the  $c$  axis indicates strong anisotropy in magnetic property and is consistent with the chain-like topology of the crystal structure. The average value  $(2/3\theta_{CW}^{ab} + 1/3\theta_{CW}^c)$  agrees well with the powder sample, where  $\theta_{CW}$  decreases steadily with Sr doping [20]. For systems with trigonal or triangular lattice, it is generally expected there is a certain amount of magnetic frustration [22]. However, the small value of frustration parameter ( $\theta_{CW}/T_N \approx 0.32$ ) implies it is absent in CSIO.

The spin structure of CSIO in zero applied magnetic field ( $H=0$ ) was characterized by surveying a large portion of the reciprocal volume at 5 K. The sample was oriented with the  $c$  axis perpendicular to the horizontal scattering plane. The diffraction data were collected with the crystal rotating along the  $c$  axis for 210 degrees. Earlier studies of undoped Ca<sub>4</sub>IrO<sub>6</sub> reported a spin configuration with magnetic wavevector  $q_m = (0.5, 0.5, 0)$  [21]. As shown in Fig. 2(a), the low- $T$  contour plot in the  $(h, k, l = 0)$  scattering plane does not show extra intensities at this reflection and equivalent positions. In contrast, new reflections appear in the plane with  $l = 2n + 1$ . All observed magnetic reflections at  $(h, k, l = 1)$  can be indexed using a magnetic wavevector  $q_m = (0, 0.5, 1)$  plus two additional magnetic domains -120 and 120 degrees apart [Fig. 2(b)]. The volume fraction ratio of the three magnetic domains is 35:33:32, and is consistent with the trigonal symmetry. The observed magnetic propagation wavevector  $(0, 0.5, 1)$  in CSIO is the same as the isostructural Sr<sub>3</sub>ZnIrO<sub>6</sub> [24]. The  $T$  dependence of the strongest magnetic reflection shows a clear second order phase transition. Fitting the data using  $I \sim (1 - T/T_N)^{2\beta}$  yields  $T_N = 13.1(3)$  K and  $\beta = 0.25(1)$ . The value of  $\beta$

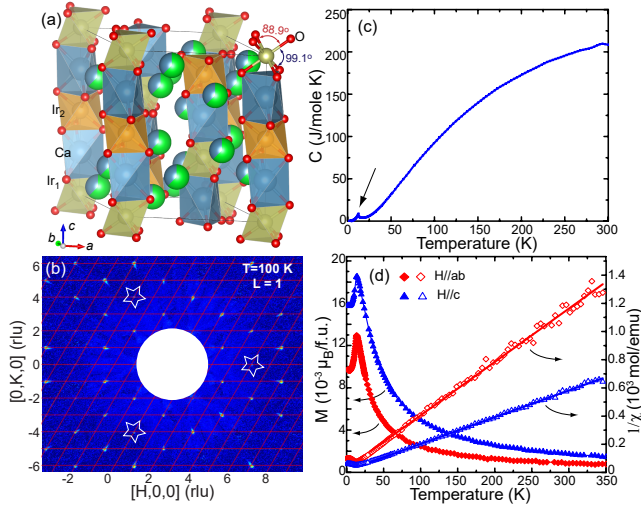


FIG. 1. (a) Crystal structure of  $\text{Ca}_2\text{Sr}_2\text{IrO}_6$  in SG  $R\bar{3}c$ . There are two distinct Ir1 and Ir2 sites located at  $(0,0,0)$  and  $(0,0,0.5)$  with different bonding oxygen environment. The trigonal distortion of  $\text{IrO}_6$  leads to out-of-plane  $91.1^\circ$  and in-plane  $88.9^\circ$  bond angles. The structure are drawn using VESTA software [23]. (b) The reciprocal-space image in the  $(h,k,l=1)$  scattering plane at 100 K with data collected from TOPAZ. The nuclear peaks in white stars are forbidden reflections of SG  $R\bar{3}c$ . (c) Temperature dependence of specific heat  $C_p(T)$ . (d)  $T$ -dependence of magnetization  $M(T)$  and inverse magnetic susceptibility  $1/\chi(T)$  in an applied magnetic field of 0.5 T parallel to the  $ab$  plane and the  $c$  axis in the field cooling protocol. Solid lines are the fits using Curie-Weiss law above transition temperature.

deviates from the critical exponent of a 3D spin system but is consistent with pure  $\text{Ca}_4\text{IrO}_6$  [21].

The magnetic structure of CSIO is determined by analyzing over eighty reflections in conjunction with representational analysis [25]. For the SG  $R\bar{3}c$  with two inequivalent Ir sites and propagation wavevector  $(0, 0.5, 1)$ , the spin configuration is described by an irreducible representation (IR) that allows moments of Ir1(Ir2) along all three crystallographic axes. The relative phase between the two iridium sites could either be ferromagnetic (FM) or AFM. In the former case, one expects strong magnetic reflections in the scattering plane with  $l$  equal to even numbers. However, all major magnetic peaks are observed in the scattering plane with  $l$  equal to odd numbers, indicating a dominant AFM coupling between the two iridium sites. This feature is verified using simulated annealing method [25]. There are weak peaks in the  $l = 0$  plane, e.g. the  $(2, 0.5, 0)$  peak. This indicates the spins at those two sites are not exactly out of phase and further confirms the reduced crystal symmetry. A reliable refinement of the spin configuration can only be reached by collecting a complete set of magnetic reflections in reciprocal space with domain populations correctly refined, since certain reflections result from the summation of different domains. Full details are given in the supplemental

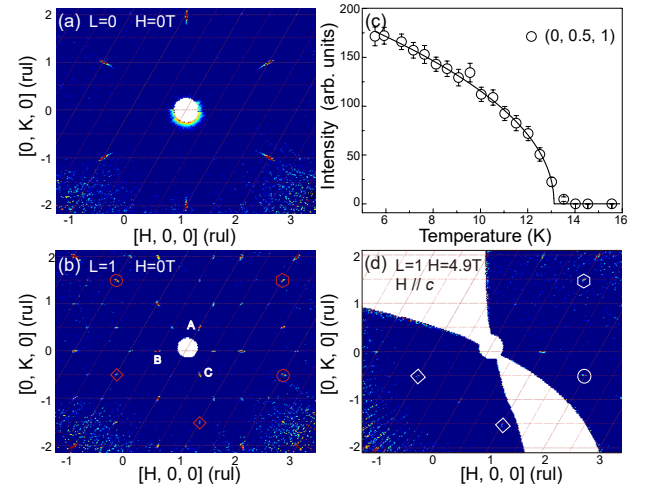


FIG. 2. The diffraction image in the (a)  $(h,k,l=0)$  and (b)  $(h,k,l=1)$  scattering plane at 5 K with zero field. Capital letters A, B, C label the three magnetic domains. Reflections encircled in square, circle and hexagons are from the same magnetic domain with the wavevector  $q_m = (0, 0.5, 1)$ . (c)  $T$  dependence of the  $(0,0.5,1)$  magnetic peak. The solid line is the fit of integrated intensity. (d) The image in the  $(h,k,l=1)$  scattering plane with a magnetic field of  $H=4.9$  Tesla applied along the  $c$  axis. The marked peaks are the magnetic reflections.

Material [26]. Using the symmetry-adapted model and the magnetic form factor for  $\text{Ir}^{4+}$  [27], we obtained the spin structure with detailed information listed in Table I. As illustrated in Figs. 3(a)-3(b), The spins have staggered  $+-$  patterns between neighboring sites along the  $c$  axis. The in-plane spin configuration is highly anisotropic despite the trigonal symmetry of the lattice. The moments are coupled ferromagnetically along the  $a$  axis but are antiferromagnetic along the  $b$  axis. The spin moments are dominantly aligned within the  $ab$  plane with out-of-plane tilt angle of  $34^\circ$  towards the  $c$  axis. The projection in the basal plane is parallel to the  $[1,2,0]$  direction. The moment direction is close to the diagonal O-Ir-O bond of within the  $\text{IrO}_6$ . The nearly collinear spin configuration is in contrast with the noncollinear spin order reported in the isostructural  $\text{Sr}_3\text{ZnIrO}_6$  [24]. The ordered moment in CSIO is  $0.66(3) \mu_B/\text{Ir}$  site. It is larger than other iridates compounds with corner- or edge sharing  $\text{IrO}_6$  octahedra [15, 28–32], but comparable with the value of  $0.87 \mu_B/\text{Ir}$  in  $\text{Sr}_3\text{ZnIrO}_6$  and  $0.6 \mu_B/\text{Ir}$  in the transition metal element substituted  $\text{Sr}_3\text{CoIrO}_6$  that has similar crystal structure with quasi-1D chains along the  $c$  axis [24, 33]. Thus, the relative large ordered moment is most likely due to the suppressed electron hopping between the isolated  $\text{IrO}_6$  octahedra.

The magnetic field effect on the spin structure is investigated with a field applied along the  $c$  axis. The diffraction pattern in the  $(h,k,l=1)$  scattering plane

TABLE I. The basis vectors (BV) and refined spin components for the SG  $R\bar{3}$  with  $q_m = (0, 0.5, 1)$ . Two independent Ir sites are located at  $(0, 0, 0)$  and  $(0, 0, 1/2)$ , respectively.

IR	BV	atom	$m_a$	$m_b$	$m_c$
$\Gamma_1$	$\psi_1$	Ir1	1	0	0
	$\psi_2$	Ir1	0	1	0
	$\psi_3$	Ir1	0	0	1
$\Gamma_1$	$\psi_1$	Ir2	1	0	0
	$\psi_2$	Ir2	0	1	0
	$\psi_3$	Ir2	0	0	1
Refinement ( $T = 5\text{K}$ )			$m_a(\mu_{\text{B}})$	$m_b(\mu_{\text{B}})$	$m_c(\mu_{\text{B}})$
Ir1			-0.33(2)	-0.64(2)	0.41(5)
Ir2			0.32(2)	0.63(2)	-0.34(5)

is presented in Fig. 2(d). Compared to the zero field data, only one of the three magnetic domains survives at  $H=4.9$  T. This supports the conclusion that the observed magnetic reflections at  $H=0$  results from multiple domains instead of one single magnetic domain with multi- $k$  structure [34]. Limited number of magnetic reflections are collected due to the partial block of neutron beam by the magnet. With the spin configuration constrained to be similar to that at  $H=0$ , the refined spins tilt further towards to the basal plane with a canting angle of  $19^\circ$  and the ordered magnetic moment decreases to  $0.60(7) \mu_B/\text{Ir}$  [Fig. 3(d)]. The tilting of the moment direction towards to the basal plane is expected since the state with field  $H$  perpendicular to the easy magnetization is energetically more favorable. On the other hand, the absence of spin-flop transition with field up to 4.9 T (the transition temperature  $T_N$  reduces from 12.5 K at  $H=0$  to 10.6 K at  $H=14$  T from specific heat measurement) indicates the magnetic structure is rather robust and consistent with observation in other iridates [15].

To further understand the nature of the observed magnetic ordering, we performed density functional theory (DFT) calculations using the Vienna Ab initio Simulation Package VASP [35, 36] with the modified Perdew-Burke-Ernzerhof exchange-correlation designed for solids (PBEsol) [37]. We employed PAW potentials [38] with the following electronic configurations Ca: $3p^6 3s^2$ , Sr: $4s^2 4p^6 5s^2$ , Ir: $6s^1 5d^8$ , and O: $2s^2 2p^4$ . The cation arrangement was chosen using a random number generator to assign Ca or Sr with the correct distribution on each site. The calculations were found to be converged with a 500 eV cutoff. To allow for the two antiferromagnetic configurations to be studied in a commensurate unit cell,  $2 \times 2 \times 1$  unit cells were employed with a  $1 \times 1 \times 2$  Monkhorst-Pack  $k$ -point mesh. All ionic coordinates were relaxed until all Hellman-Feynman forces were less than  $0.015 \text{ eV/\AA}$ . A Hubbard  $U$  of 2.0 eV and intrasite Hund's coupling  $J_H = 0.2 \text{ eV}$  for Ir  $d$ -states were employed [39]. The magnetic structure shown in Fig. 3

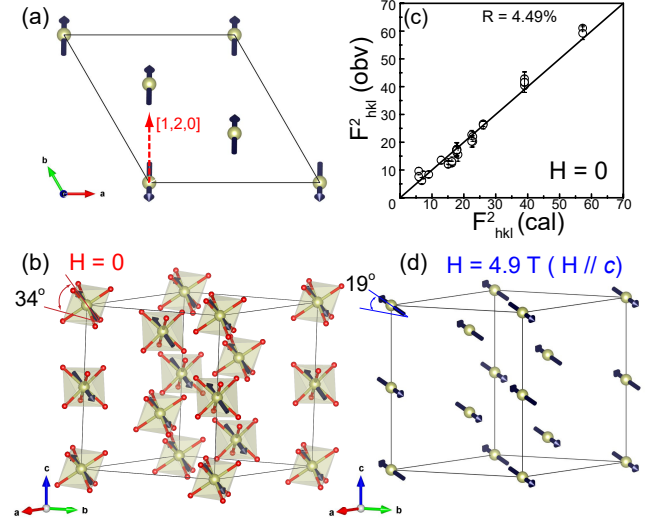


FIG. 3. (a) The spin configuration projected on the  $ab$  plane with moment along the  $[1,2,0]$  direction. (b) The refined magnetic structure at  $H = 0$  from single crystal neutron diffraction measurement. the spin moments tilt  $34^\circ$  away from the  $ab$  plane. (c) The calculated magnetic intensities versus observation using the model described in the text. (d) The magnetic structure with  $H = 4.9$  T applied along the  $c$  axis, the canting angle away from the basal plane reduces to  $19^\circ$ .

(AFM3 state) and the one reported for pure  $\text{Ca}_4\text{IrO}_6$  (AFM1 state with  $q_m = (0.5, 0.5, 0)$  as shown in Ref. [21]) were chosen as the initial magnetic configurations. With SOI taken into account, the density of states (DOS) for both magnetic structures exhibit similar features. However, the energy of the AFM3 state is  $2.5 \text{ meV/Ir}$  site lower than that of AFM1 state, thereby confirms the observed magnetic structure as the most probable ground state. The initial magnetic structure tested in the calculation has moments direction arbitrary chosen while keeping the configuration consistent with the magnetic wavevector, but the converged state has moments relaxed nearly along the Ir-O bond direction. There is an insulating gap  $\sim 0.5 \text{ eV}$  near  $E_F$ , mainly from the  $t_{2g}$  orbital of the  $\text{Ir}^{5+}$  ions. This is consistent with resistivity measurement shown in Fig. 4(b) where an band gap ( $2\Delta_{ag}$ )  $\sim 1.26 \text{ eV}$  is obtained by fitting the data to the form of  $\rho(T) = \rho_0 \exp(\Delta_{ag}/k_B T)$  for  $300 \text{ K} < T < 500 \text{ K}$ . The calculated spin moment of  $0.5 \mu_B/\text{Ir}$  agrees well with the experimental observation.

## DISCUSSION

The anisotropic magnetic configuration that breaks the trigonal symmetry and the moment direction following the diagonal O-Ir-O bond strongly suggests the emergence of relativistic SOI, where the exact form of the magnetic Hamiltonian depends on the lattice geometry



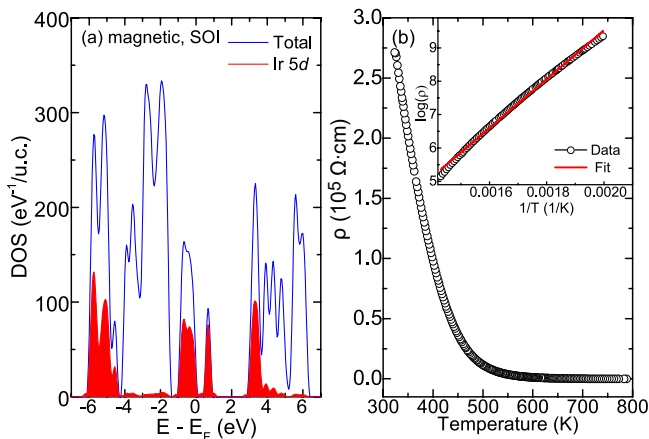


FIG. 4. The density of states of CSIO from DFT calculation with spin-orbit interaction included. The initial magnetic structure is similar to the one illustrated in Fig. 3. (b) The temperature dependence of resistivity of CSIO. Inset shows the fit to  $\rho(T) = \rho_0 \exp(\Delta_{ag}/k_B T)$  with  $\Delta_{ag} = 0.63$  eV.

[9]. An Heisenberg interaction  $\vec{S}_i \cdot \vec{S}_j$  dominates in a corner-shared iridate, e.g., the square lattice  $\text{Sr}_2\text{IrO}_4$ . The canted spin moments that rigidly follow the staggered rotation of octahedra [30, 40] is naturally explained by the strong SOI. In contrast, the highly anisotropic interactions appear due to the off-diagonal hopping matrix in the edge-shared case. This maps the system into a quantum compass model and has been extensively studied in the honeycomb lattice  $\text{A}_2\text{IrO}_3$  [41, 42]. Since the building block of perovskite-derived iridates are made of individual  $\text{IrO}_6$  octahedron, the isolated  $\text{IrO}_6$  without corner- or edge sharing connectivity, makes the CSIO a ideal system to study the SOI in the single ion limit. On the other hand, the influence of nonoctahedral crystal field splitting ( $\Delta$ ) can not be ignored. Although the  $j_{\text{eff}} = 1/2$  state was initially thought to be a robust feature in iridates as evidenced by the vanishing intensity at the  $L_2$  absorption edge [3], it is now recognized that the local distortion could dramatically modify the ground states. For example, RIXS measurements on quasi-one-dimensional spin-chain  $\text{Sr}_3\text{CuIrO}_6$  has revealed  $\Delta = 0.31$  eV caused by the reduction of O-Ir-O bond angle to  $82^\circ$ , and contributed a significant mixing between  $j_{\text{eff}} = 1/2$  and  $j_{\text{eff}} = 3/2$  states [16]. A similar result is reported in post perovskite  $\text{CaIrO}_3$  [17], where the energy scale of the octahedral compression along the local  $z$  axis ( $\Delta = -0.71$  meV) is comparable with the SOI strength  $\lambda = 0.52$  meV and signifies a departure from the  $j_{\text{eff}} = 1/2$  state. While the local symmetry of these compounds is not the same, tetragonal for perovskites and trigonal for non-perovskites, distortions of  $\text{IrO}_6$  octahedron seems to be ubiquitously present in the so-called  $j_{\text{eff}} = 1/2$  iridates (e.g.  $\text{Na}_2\text{IrO}_3$  with  $|\Delta| = 0.11$  eV [43],  $\text{Y}_2\text{Ir}_2\text{O}_7$  with  $|\Delta| = 0.59$  eV [44]). A nonoctahedral crystal field must be considered in realistic models

since the electronic structure highly depend on the relative orbital contributions. In this respect, the  $\text{A}_4\text{BO}_6$  ( $A$  is alkaline earth ions and  $B$  is  $4d$  or  $5d$  element) system featuring a chain-like structure with minimal local distortion of  $\text{BO}_6$  octahedra represents a new family of platform to realize the spin-orbit-entangled state. Yet, distinct magnetic configurations have been reported in isostructural iridates such as spin order with wavevector  $(0.5, 0.5, 0)$  in  $\text{Ca}_4\text{IrO}_6$  [21], or a noncollinear spin structure in  $\text{Sr}_3\text{ZnIrO}_6$  [24]. The difference indicates the magnetic coupling between seemingly isolated Ir octahedra might depend on the overall averaged lattice and warrant more experimental investigation. Unlike the transition metal substituted  $\text{Sr}_3\text{NiIrO}_6$  or  $\text{Sr}_3\text{CoIrO}_6$  where the magnetism is influenced by the interplay between transition metal and the  $5d$  ions, or the  $4d$  counterpart  $\text{Sr}_4\text{RhO}_6$  [45] where the strength of SOI is smaller than the  $5d$  systems, the CSIO can be regarded as a suitable example to further explore the electronic and magnetic properties arising from the SOI.

In summary, neutron and X-ray diffraction have been employed to investigate the crystal and magnetic structures of the trigonal lattice iridate  $\text{Ca}_2\text{Sr}_2\text{IrO}_6$ . The well-separated  $\text{IrO}_6$  octahedra are close to the cubic limit with six equal Ir-O bond distance and O-Ir-O bond angles  $\approx 90^\circ$ . The  $\text{Ir}^{4+}$  spins form an anisotropic three-dimensional antiferromagnetic configuration with wavevector  $(0, 0.5, 1)$ . The ordered moment is  $0.66(3) \mu_B/\text{Ir}$ , larger than iridates with corner- and edge sharing  $\text{IrO}_6$  octahedral networks. The DFT calculation confirms that the observed magnetic ordering is the most probable ground state and indicates that the insulating behavior is enhanced by the spin-orbit interaction.

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