Pseudospin-lattice coupling and electric control of the square-lattice iridate $\text{Sr}_2\text{IrO}_4$

Feng Ye,1 Christina Hoffmann,1 Wei Tian,1 Hengdi Zhao,2 and G. Cao2

1Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
2Department of Physics, University of Colorado at Boulder, Boulder, Colorado 80309, USA

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$\text{Sr}_2\text{IrO}_4$ is an archetypal spin-orbit-coupled Mott insulator and has been extensively studied in part because of a wide range of predicted novel states. Limited experimental characterization of these states thus far brings to light the extraordinary susceptibility of the physical properties to the lattice, particularly, the Ir-O-Ir bond angle. Here, we report a newly observed microscopic rotation of the IrO$_6$ octahedra below 50 K measured by single crystal neutron diffraction. This sharp lattice anomaly provides keys to understanding the anomalous low-temperature physics and a direct confirmation of a crucial role that the Ir-O-Ir bond angle plays in determining the ground state. Indeed, as also demonstrated in this study, applied electric current readily weakens the antiferromagnetic order via the straightening of the Ir-O-Ir bond angle, highlighting that even slight change in the local structure can disproportionately affect the physical properties in the spin-orbit-coupled system.

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Strong spin-orbit interactions (SOI), along with appreciable Coulomb interactions, crystalline electric field, and large orbital hybridization in 5$d$-electron based oxides has produced a wide range of quantum phenomena such as spin liquid phases [1], superconductivity [2, 3], and Kitaev magnetism [4–6]. One prominent example is the observation of the so-called effective $j_{\text{eff}}=1/2$ Mott insulating state in iridates [7–9], where the magnetism is attributed to an isotropic pseudospin [10–12]. Unlike the situation in the 3$d$ transition metal oxides which have a distinct energy scale in both orbital and spin parts and sequential orbital and magnetic orders, the notion of spin-orbital separation in the 5$d$ systems is no longer valid due to the strong SOI. Furthermore, the Jahn-Teller effect, which measures the couples between the lattice and orbital degrees of freedom and is a common occurrence in 3$d$-electron based oxides, remains largely unexplored in the iridates. However, recent experimental and theoretical studies indicate that the magnetic properties and low-energy spin dynamics [13, 14], can be better explained using a pseudospin-lattice coupling mechanism. The introduction of such a term in the spin Hamiltonian not only correctly describes the metamagnetic transitions [13], but also explains a finite in-plane magnon gap [15–17]. It also predicts an orthorhombic structural distortion in the order of $10^{-4}$, which can be probed using a high resolution Larmor precession technique [18].

The extraordinarily strong coupling between the electronic and structural properties in the iridates provides unprecedented opportunities to uncover novel quantum phases by merely modifying the local structure. It is particularly interesting that application of hydrostatic pressure steadily reduces the charge gap of approximately 500 meV and diminishes the antiferromagnetic (AFM) state existent in the square lattice Sr$_2$IrO$_4$ (Sr-214), yet retains the insulating state [19–21]. A possible quantum paramagnetic state at pressures greater than 20 GPa is proposed, attributing to the suppression of the interlayer exchange coupling and enhanced magnetic frustration within the IrO$_2$ layer [22]. Alternative approaches to modify the structure using strain engineering are also reported in epitaxial grown thin films of Sr-214 [23–27], where a compressive or tensile strain can drastically enhance or reduce the transition temperature $T_N$. In some cases, the strengthened in-plane exchange interaction can promote a short range magnetic correlation well above the nominal transition [27]. In this paper, we report a newly observed lattice anomaly at 50 K. Given the exceptionally strong coupling between the lattice and physical properties in Sr-214, this structural anomaly, which has escaped all previous studies until now. Our study provides much-needed keys to better understand the low-temperature magnetic and transport behavior, which remains a focus of current studies on the iridates. Indeed, the susceptibility of the physical properties to the lattice is further revealed by the application of in-plane $dc$ electric current as a new stimulus that straightens the Ir-O-Ir bond angle, thus suppresses the canted AFM order. These results reveal crucial insights into the crystal structure and demonstrates that the subtle changes in the crystal structure, whether ambient or induced, dictate the physical properties in this spin-orbit-coupled Mott insulator.

Single crystals of Sr-214 were grown using the self-flux method [28, 29]. The magnetic susceptibility and specific heat were measured using a Quantum Design Magnetic Property Measurement System. The neutron diffraction measurements were carried out at the HB1A triple axis spectrometer at the High Flux Isotope Reactor and the TOPAZ diffractometer at the Spallation Neutron Source, ORNL. For measurements at HB1A, a sample assembly of over 40 single crystals (mass ~ 25 mg, mosaicity ~ 1.5°) was aligned at various scattering planes to probe the magnetic and nuclear re-
flections. A closed-cycle refrigerator was used to regulate the sample temperature (T). For measurements at TOPAZ, a plate-like single crystal with dimensions of 1.8 mm × 1.8 mm × 0.55 mm was used. The sample temperature is controlled using the Oxford cryostream cooler Cobra 800. Electric current was applied using the power supply from BioLogic science potentiostat with a voltage range of ±12 V and current range ±200 mA.

Figures 1(a)-1(b) show the main structural features of the Sr-214 with Ir ions forming a square-lattice network. The system exhibits a characteristic staggered rotation of IrO$_6$ octahedra along the c axis at about $\sim 11.8^\circ$ [30, 31]. Because of the layered structure, the corresponding IrO$_6$ octahedra elongate slightly along the c-axis causing a crystal electric field (CEF) level $\Delta_{CEF} \sim 200$ meV [4, 32]. Neutron scattering studies reveal a long-range magnetic order below 240 K [Fig. 1(e)]. Magnetic structure refinement indicates that the spins of Ir ions form a canted AFM configuration. The in-plane magnetic components closely track the rotation of the IrO$_6$ [31, 33], which results directly from the SOI; the canting angle is governed by a characteristic ratio, $\phi \sim D/2J$, of the antisymmetric Dzyaloshinskii-Moriya (DM) interaction $D$ to the isotropic Heisenberg interaction $J$ [4].

![Diagram of Sr$_2$IrO$_4$ crystal structure](image)

**FIG. 1:** (a) The crystal structure of Sr$_2$IrO$_4$ in the tetragonal setting. For simplicity, only the iridium and oxygen atoms are shown. (b) The connecting IrO$_6$ network is viewed along the c axis. The T-dependence of (c) the in-plane and c-axis resistivity $\rho_{ab}(T)$ and $\rho_c(T)$, (d) the lattice parameters $a$ and $c$ measured from neutron diffraction, and (e) the strongest AFM peak (1,0,2). The solid line in panel (e) is a guide to the eye.

It is known that the onset of long range magnetic order at $T_N$ causes no anomaly in $\rho_{ab}$ or $\rho_c$ [28, 34], specific heat (data not shown), and the lattice parameters [Figs. 1(c)-1(d)]. The conventionally anticipated correlation between the structural and physical properties is conspicuously missing at $T_N$. However, such a correlation is instead established below a newly observed lattice anomaly, $T_M = 50$ K. As shown in Fig. 1(c), both the in-plane and c-axis resistivity exhibit noticeable kinks below $T_M$. Recently, independent experiments have confirmed the anomalous character at low T. Among them, a magnetic study shows that the residual magnetization increases below $T_N$, peaks at $T_M$, and diminishes on further cooling [35]. Such behavior was initially attributed to the reorientation of the canted spin moment [34]. A muon spin rotation study by Franke et al. shows that the oscillation of single precession signal below the magnetic transition splits into high and low frequency components below 20 K [36]. The authors of Ref. [36] suggest that the modification of the magnetic structure leads to structurally equivalent muon sites that experience increasingly distinct local field at low temperatures.

Nevertheless, the abnormal low-T behavior demands a detailed examination of the crystal structure. The neutron diffraction measurements indicate that the system remains tetragonal at all temperature and does not show a lower symmetry below the magnetic transition [30, 31, 37, 38]. Since the neutron coherent scattering length of the light oxygen atom ($b_O = 5.8$ fm) is comparable to the much heavier strontium ($b_{Sr} = 7.02$ fm) and iridium ($b_{Ir} = 10.6$ fm), the subtle change of the IrO$_6$ rotation can be readily characterized using neutron diffraction and used to quantify its coupling to the magnetic order. The structure factor of the nuclear scattering at a wavevector transfer $\vec{q}$ is

$$|F(\vec{q})|^2 = \left| \sum_i b_i \exp(\vec{r}_i \cdot \vec{q}) \right|^2,$$

where $b_i$ is the coherent scattering length for individual atom inside the unit cell and $\vec{r}_i$ is the corresponding atomic coordinate. The rotation of the IrO$_6$ octahedra leads to the nonzero peak intensities forbidden for the space group $I4/mmm$, e.g., $(2,1,2n+1)$, and is solely determined by the in-plane oxygen atom $O_1$ located at $(1/4 - \delta,1/2 - \delta,1/8)$ and equivalent positions, where $\delta \approx 0.05$ characterizes the deviation from the undistorted IrO$_6$ scenario. The intensity of the superlattice peak $(2,1,1)$, $I_{211}$, can be derived as $b_O(\sin \delta + \sin 3\delta)^2$ and is proportional to $\delta^2$ in the small rotation limit.

The red squares in Fig. 2(c) show the $(2,1,1)$ peak intensity upon cooling. The gradual increase in intensity implies an enhancement of the IrO$_6$ rotation. Surprisingly, the monotonic evolution experiences an abrupt change below 50 K. To further verify the feature, $\theta - 2\theta$ scans across the peak are performed (blue circles). The marked decrease at low-T reveals that the suppression of IrO$_6$ octahedral rotation is intrinsic. Comparing to 50 K, the value at 5 K is reduced by 5%, which trans-
FIG. 2: (a) The magnetization $M$ in the zero-field-cooled (ZFC) and field-cooled (FC) protocols, with a magnetic field of 0.1 Tesla applied along the [1, 0, 0] and [1, 1, 0] directions. (b) The $T$-dependence of the peak intensity of the magnetic (0,0,3) reflection which probes the canted spin component. The solid line is a guide to the eye. (c) The $T$-evolution of the structural peak (2,1,1) associated with the in-plane rotation of the IrO$_6$ octahedra. The (blue) circles are the integrated intensities, the (red) squares are the peak intensities. Inset shows the comparison of the $\theta - 2\theta$ scans at 5 K and 50 K. Both peaks are fit using Gaussian profile.

lates to a suppression of rotation angle by $0.3^\circ$. As the moment strictly follows the octahedral rotation, one expects a sudden reduction of the rotation or straightening of the Ir-O-Ir bond angle. This can be probed by examining the magnetic reflection (0,0,3) that is exclusively sensitive to the canted component [31]. Fig. 2(b) displays the $T$-dependence of the magnetic peak with long counting time. A similar kink is observed below 50 K, reinforcing the structural anomaly in Fig. 2(c). This is also consistent with the abrupt reduction of the magnetization shown in Figure 2(a) with an external magnetic field of $\mu_0 H = 0.1$ T applied along either the [1, 0, 0] or the [1,1,0] directions, which also agrees well with our earlier report [39]. These data point out a close association between the crystal structure and magnetic order, confirming an essential role of the pseudospin-lattice coupling that dictates the low-energy magnetic properties.

This key characteristic of the lattice degree of freedom provides an effective “knob” to control the physical properties of the iridates using small external stimuli that readily couple to the lattice. Electrical current, as a novel stimulus, is particularly effective in controlling the lattice, and thus the magnetic properties [41]. The previous x-ray study has shown a prominent lattice expansion for the undoped Sr-214 under electric current, accompanied by remarkable reduction in both transition temperature and in-plane magnetization. No structural anomaly is observed in Tb-doped Sr-214 where long range magnetic order is absent [42]. To better characterize the structural response, we perform single crystal neutron diffraction measurements with in situ electric current application. The orientation of the Sr-214 crystal is determined using the x-ray Laue method to ensure the currents are applied in the basal plane [inset of Fig. 3]. Since the diffractometer TOPAZ has a white incident neutron beam with large reciprocal coverage, the sample is mounted such that the characteristic (4,0,0) and (0,0,12) peaks can be simultaneously accessed [Fig. 3(a)]. In such fixed-orientation configuration, we collected a total number of around 2000 nuclear reflections at each temperature that allow a reliable structural determination. A electric current of 80
mA (current density $\sim 8\ A/cm^2$) is employed at 300 K and the sample is cooled to 100 K while the electric current is maintained. The crystal is subsequently warmed to 300 K at a rate of 0.5 K/min. Figs. 3(b)-3(c) display the $T$-dependence for the out-of-plane and in-plane lattice parameters with and without electric current, respectively. It is evident that the $c$-axis lattice parameter is insensitive to the current. In contrast, the in-plane lattice parameter displays a dramatic response to the current, consistent with the fact that the magnetic moments lie within the basal plane. The overall $T$-dependence of lattice parameter $a$ at 80 mA stays above the one at $I = 0$. While both data can be well described by a quadratic form, a slope change near the AFM transition is clearly visible [see the dashed curve in Fig. 3(c)]. The detailed structure refinement reveals that the Ir-O bond distances remain unchanged at $I = 80$ mA. The corresponding IrO$_6$ octahedron also remains slightly elongated, with a ratio of the Ir-O$_2$ to the Ir-O$_1$ bond distances being 1.04 throughout the temperature range measured.

Further reduction of temperature. A slope change occurs at the magnetic transition $T_N$ at $I = 0$ mA. In sharp contrast, the bond angle becomes notably straighter or relaxed by $0.3^\circ$ when a current of $I = 80$ mA is applied in the basal plane. In addition, these bond angle values show a smooth decrease upon cooling; the anomaly observed near $T_N$ at the ambient condition is significantly suppressed. Indeed, the corresponding in-plane magnetic susceptibility displays the behavior consistent with the structural data [Fig. 4(a)]. The $\chi_a$ at 80 mA shows reduction in amplitude along with the suppression and broadening of the transition temperature. The reduced magnetization is expected because the DM interaction $D$ becomes smaller as the Ir-O-Ir bond angle relaxes due to the applied current (recall $\phi \sim D / J$).

Tuning the physical properties, in particular magnetism using electric current, is highly desirable. It opens a new frontier for studies of the correlated and spin-orbit-coupled systems [41, 43, 44]. For instance, a reorientation of the AFM order is observed in the metallic Fe$_{1/3}$ NbS$_2$ at a remarkable low current density ($\sim 10^4\ A/cm^2$) showing the potential to build AFM spintronics devices [43]. Spatially inhomogeneous rotation of the skyrmion lattice in the metallic helimagnet MnSi occurs when a current flows across the sample. This emphasizes the role of friction near the sample edges for skyrmion-based applications [44]. Much efforts have also been devoted to insulating materials such as ruthenate oxide Ca$_2$RuO$_4$ [40, 45-50]. Zhao et al. have shown that a extremely small electric current density ($\sim 0.15\ A/cm^2$) can reduce the orthorhombic distortion and AFM order, and further induce a new orbital state featuring a simultaneous jump in both magnetization and electrical resistivity [40]. The authors propose that orbital occupancies stabilized by the nonequilibrium current drive the lattice changes and the novel phenomena. Because of the distinct energy hierarchy in the SOI system, the narrow band Mott insulator Sr-214 is more susceptible to such a perturbative approach. It is anticipated the crystallographic details through this structural study would feed critical information for the electronic band structure calculations, and assist to explain the emerging phenomena including the negative differential resistivity, reversible resistance switching in Sr-214 [41, 51, 52], and the nonlinear conductivity in other spin-orbit-coupled iridates [53, 54].

In summary, neutron diffraction work of the square lattice Sr-214 reveals a pronounced reduction of the staggered IrO$_6$ distortion below 50 K. This important observation will help better understand the low-temperature physics of the archetypal spin-orbit-couple Mott insulator as the magnetic and transport properties closely tracks the underlying lattice. This point is further strengthened in that applying a steady electric current at 8 A/cm$^2$ readily reduces the in-plane IrO$_6$ octahedral rotation, which aligns well with the diminishing bulk magnetization. Our study demonstrates that the application of
electric current is an effective and appealing route to tune the lattice and probe the rich physics in the SOI system.

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