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Gang Cao  
*University of Colorado Boulder*

Jasminka Terzic  
*University of Colorado Boulder*

H. D. Zhao  
*University of Colorado Boulder*

H. Zheng  
*University of Colorado Boulder*

Lance E. De Long  
*University of Kentucky*, lance.delong@uky.edu

See next page for additional authors

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Authors

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Electrical Control of Structural and Physical Properties via Strong Spin-Orbit Interactions in Sr$_2$IrO$_4$

G. Cao,$^{1,3}$ J. Terzic,$^1$ H. D. Zhao,$^1$ H. Zheng,$^1$ L. E. De Long,$^2$ and Peter S. Riseborough$^3$

$^1$Department of Physics, University of Colorado at Boulder, Boulder, Colorado 80309, USA
$^2$Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506, USA
$^3$Department of Physics, Temple University, Philadelphia, Pennsylvania 19122, USA

Abstract: We have conducted a new study of Sr$_2$IrO$_4$ that centers on unconventional, single-crystal x-ray diffraction measurements with simultaneous application of electrical current to diffraction samples, as well as the $I$-$V$ characteristics, electrical resistivity and magnetization as functions of temperature, electrical current, and magnetic field. Our central finding is that application of electrical current causes the $a$-axis lattice parameter to expand by an astonishing 1% that, in turn, precipitates profound changes in physical properties. The current-controlled lattice expansion closely tracks the long-range magnetic order, causing a considerable decrease in both the Néel temperature $T_N$ and magnetization, due to the strong SOI that rigidly locks the Ir moments to the lattice. The current dependence of the $a$-axis expansion is highly nonlinear, which induces the novel $I$-$V$ characteristics of Sr$_2$IrO$_4$.

Simultaneous control of structural and physical properties via electrical current is a rare but extremely desirable contemporary goal because of its great technological potential. Our discovery of such behavior in Sr$_2$IrO$_4$ opens new avenues for understanding the fundamental consequences of strong SOI in crystalline solids, and provides a new paradigm for development of functional materials and devices.

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It is now widely recognized that a unique competition between spin-orbit interactions (SOI) and Coulomb correlations $U$ in 4$d$ and 5$d$ elements and their compounds drives unusual physical behaviors that markedly differ from those of their 3$d$ counterparts [1,2]. The 5$d$ iridates display particularly strong and surprising influences of SOI on their physical properties. Early studies [3–8] indicated that iridates exhibit a preference for magnetic, insulating ground states, a trend now recognized as a consequence of a combined effect of $U$ and strong SOI. An important example of this effect is the $J_{\text{eff}} = 1/2$ Mott state identified in Sr$_2$IrO$_4$ [9], whose defining characteristic is the strong locking of the lattice and Ir magnetic moments [1,2,10].

We have conducted a new study of Sr$_2$IrO$_4$ that centers on unconventional, single-crystal x-ray diffraction measurements with simultaneous application of electrical current to diffraction samples, as well as the $I$-$V$ characteristics, electrical resistivity and magnetization as functions of temperature, electrical current, and magnetic field. Our central finding is that application of electrical current causes the $a$-axis lattice parameter to expand by an astonishing 1% that, in turn, precipitates profound changes in physical properties. The current-controlled lattice expansion closely tracks the long-range magnetic order, causing a considerable decrease in both the Néel temperature $T_N$ and magnetization, due to the strong SOI that rigidly locks the Ir moments to the lattice. The current dependence of the $a$-axis expansion is highly nonlinear, which induces the novel $I$-$V$ characteristics of Sr$_2$IrO$_4$.

Simultaneous control of structural and physical properties via electrical current is a rare but extremely desirable contemporary goal because of its great technological potential. Our discovery of such behavior in Sr$_2$IrO$_4$ opens new avenues for understanding the fundamental consequences of strong SOI in crystalline solids, and provides a new paradigm for development of functional materials and devices.

An explanation of experimental techniques and additional discussion are presented in the Supplemental Material [11].

Sr$_2$IrO$_4$ is the archetype, SOI-driven insulator [9] with $T_N = 240$ K [3–6], and an electronic energy gap $\Delta < 0.62$ eV [9,12–15]. It crystallizes in a tetragonal structure with space group $I4_1/acd$ (no. 142) with $a = b = 5.4846$ Å and $c = 25.804$ Å at temperature $T = 13$ K [3–5]. Recent studies suggest a further reduced space group $I4_1/a$ (no. 88) for Sr$_2$IrO$_4$ [16,17]. Two signature characteristics of Sr$_2$IrO$_4$ are essential for understanding the results of this study. (1) Rotation of the IrO$_6$ octahedra about the $c$ axis by approximately 12°, which corresponds to a distorted in-plane Ir1-O2-Ir1 bond angle $\theta$, has a critical effect on the ground state [2,18–25]. (2) The magnetic structure is composed of ordered moments [0.208(3)$\mu_B$/Ir] canted within the basal plane [16]. The 13(1)° canting of the moments away from the $a$ axis closely tracks the staggered rotation of the IrO$_6$ octahedra [26], which sharply contrasts the behavior of 3$d$ oxides [27].

A representative diffraction pattern taken with the basal plane of Sr$_2$IrO$_4$ aligned nearly perpendicular to the incident x-ray beam is shown in Fig. 1(a). The Bragg peaks for Miller indices (220) or (0016) are representative for the discussion that follows. The position and intensity of the (0016) peak for temperature $T = 200$ K are shown in
Figs. 1(b) and 1(c), respectively, and undergo remarkable changes upon the application of basal-plane electrical currents $I$ up to 105 mA. The (0016) peak shifts up and to the right with a threefold reduction in intensity that is sensitive to the atomic positions within a unit cell [Fig. 1(c)]. Other Bragg peaks exhibit similar shifts with $I$, and either enhanced or reduced intensities, which reflects changing interference generated by shifts in atom positions. The current-induced lattice changes are also accompanied by a subtle but visible color and size change of the sample, as seen with the aid of a polarizing microscope [11].

The current-controlled changes in the $a$- and $c$-axis lattice parameters were quantitatively characterized by x-ray diffraction for $I$ applied along either the basal plane or the $c$ axis. The lattice responds more strongly to current in the basal plane than along the $c$ axis, which suggests that the orientation of the Ir moments is important, and that Joule heating is not affecting the data [11].

We now focus on normalized changes in the $a$- and $c$-axis lattice parameters $\Delta a/a$ and $\Delta c/c$ with basal-plane $I$, where $\Delta a/a = |a(I) - a(0)|/a(0)$ and $0 \leq I \leq 105$ mA; $\Delta c/c$ is similarly defined. Figure 2(a) shows $\Delta a/a$ peaks at nearly 1% near $T_N$, then decreases to 0.2% at 300 K, whereas $\Delta c/c < 0.1\%$. The clear difference between $\Delta a/a$ and $\Delta c/c$ once again does not support a Joule heating effect [11], and further confirms an important role for the in-plane Ir moments. A more striking observation is that the temperature dependence of $\Delta a/a$ closely tracks that of the $a$-axis magnetization $M_a$ [Fig. 2(a)]; this is direct evidence that the current-controlled expansion of the $a$ axis involves interlocking of cooperative magnetic order and the lattice. The reduced magnetic canting must be accompanied by a simultaneous increase of $\theta$, which is a critical parameter for determining the ground state [24,25].

We expect the current-controlled lattice expansion to be strongly associated with long-range antiferromagnetic (AFM) order. We therefore undertook a parallel study of Sr$_2$Ir$_{0.97}$Tb$_{0.03}$O$_4$, since a 3% replacement of Ir$^4+$ by Tb$^{4+}$ suppresses $T_N$ to zero, but conveniently retains the insulating state and the original crystal structure [28]. We found that the absolute values of $\Delta a/a$ and $\Delta c/c$ for Sr$_2$Ir$_{0.97}$Tb$_{0.03}$O$_4$ for $I = 105$ mA are small ($<0.2\%$) and weakly temperature dependent in the absence of AFM order [Fig. 2(b)]. A comparison of Figs. 2(a) and 2(b) clearly points to a critical role played by long-range AFM in the current-controlled lattice expansion, and essentially eliminates the possibility of a Joule heating effect [11].

We also examined the conventional thermal expansion of Sr$_2$IrO$_4$ measured without application of $I$. The temperature dependences of the $a$- and $c$-axis lattice parameters and their corresponding changes $\delta a/a$ and $\delta c/c$ due to pure thermal expansion $\delta a/a = [a(T) - a(90\,K)]/a(90\,K)$ and $\delta c/c$ is similarly defined) demonstrate that the $a$ axis expands linearly and only slightly ($\sim0.1\%$) from 90 to 300 K [Figs. 2(c) and 2(d)]. The corresponding coefficient of linear thermal expansion $\alpha = 1/a (da/dT)$ is approximately $5.0 \times 10^{-6} \, ^\circ\text{C}^{-1}$, which is small and comparable to those of many materials [29]. The small thermal expansion of Sr$_2$IrO$_4$ is also consistent with its high melting point ($>1900^\circ\text{C}$), which reflects bond energies on the order of electron volts. The sharp contrast between the conventional thermal expansion $\delta a/a (0.1\%$) and the novel current-controlled $\Delta a/a (~1\%$) highlights the extraordinary coupling between current and the AFM state.

We also observe significant changes in the $a$-axis magnetic susceptibility $\chi_a(T)$ and the $a$-axis magnetization $M_a$ when current is applied (Fig. 3). $T_N$ is drastically decreased by 40 K for $I = 80$ mA [Figs. 3(a) and 3(b)], and the value of $M_a$ is reduced by 16% [Figs. 3(c) and 3(d)]. Magnetic canting is ascribed to a Dzyaloshinsky-Moriya interaction [24,30] that is closely associated with $\theta$; the canting decreases with increasing $\theta$ and vanishes when $\theta = 180^\circ$ [24]. This is consistent with the reduced $M_a$ that signals enhanced itinerancy due to increased $\theta$.

Another prominent consequence of the current-controlled lattice expansion is non-Ohmic behavior that
features a negative differential resistance (NDR). NDR \[31–34\] is a nonlinear phenomenon with a negative ratio of voltage shift in response to a current change, \(\Delta V = \Delta I < 0\), contrary to Ohm’s law, which describes the traditional positive, linear relationship, \(\Delta V = \Delta I > 0\). The NDR phenomenon is in general attributed to either an "electrothermal" effect or a "transferred carrier" effect \[31,32\]. The more common form of NDR is manifest in N-shaped I-V characteristics \[31–34\]. Alternatively, an S-shaped NDR has been observed in various memory devices \[35,36\] and a few bulk materials such as VO\(_2\), CuIr\(_2\)S\(_4\), Ca\(_3\)Ru\(_2\)O\(_7\), and 1T-TaS\(_2\) [36–40]. These bulk materials are characterized by a first-order metal-insulator transition (MIT) and, except for Ca\(_3\)Ru\(_2\)O\(_7\), are without an AFM state. The S-shaped NDR in these materials is closely associated with the first-order MIT, and is attributed to drastic differences in crystal and electronic structures below and above the MIT \[41\]. In contrast, Sr\(_2\)IrO\(_4\) features strong AFM order and a Mott insulating state that persists up to at least 600 K without a MIT \[1,2,18,20,21\], indicating a different mechanism that drives the NDR in Sr\(_2\)IrO\(_4\).

An S-shaped NDR was observed in an earlier study of Sr\(_2\)IrO\(_4\) [6], but the underlying mechanism remained unclear up to now. The I-V curves for \(I\) applied along either the \(a\) or \(c\) axis at a few temperatures are presented in Figs. 4(a) and 4(b), along with the strong anisotropy of the response in Fig. 4(c). A linear I-V response during an initial current ramp is followed by a sharp threshold voltage \(V_{th}\) that marks a switching point where \(V\) abruptly drops with increasing \(I\), thus signaling a NDR. Another broad turning point emerges with further current increase, and is more distinct in the \(c\)-axis I-V curves for \(T < 100\) K.

A plot of \(V_{th}\) as a function of temperature displays a pronounced slope change near 100 K, where an anomaly in \(M_g\) occurs [Fig. 4(d)]. Previous studies \[18,21\] have shown that \(M_g\) undergoes two additional anomalies at \(T_M \approx 100\) and...
indicating an incipient metallic state. The representative temperature after peaking around 11 K [Fig. 4(e), inset], and the corresponding activation energy gap drops by nearly 3 orders of magnitude at low temperatures (estimated from data covering the range 100–270 K), which decreases from 81 to 32 meV, as $I$ increases from 0.1 to 20 mA. There is a clear drop of $\rho_a$ with decreasing temperature after peaking around 11 K [Fig. 4(e), inset], indicating an incipient metallic state. The representative

differential resistance $dV/dI$ [11] at 100 K reveals two anomalies near 10 and 45 mA, marked as $I_{C1}$ and $I_{C2}$, respectively [Fig. 4(f)]. Corresponding I-V curves at 100 K feature a sharp switching point ($V_{th}$) at $I_{C1}$ and a broader turning point near $I_{C2}$ [Fig. 5(a)].

It is crucial to note that the current-controlled $a$-axis expansion $\Delta a/a$ closely tracks the I-V curves with nonlinear changes at $I_{C1}$ and $I_{C2}$, respectively [upper horizontal axis in Fig. 5(a)]. The slope changes in $\Delta a/a$ signal successively more rapid expansions of the $a$ axis at $I_{C1}$ and $I_{C2}$, each accompanied by a more significant increase in $\theta$, which, in turn, enhances electron hopping [Fig. 5(b)] (more discussion below). As the current further increases above $I_{C2} = 45$ mA, $\Delta a/a$ appears to saturate. This explains why a magnetic field $H$ reduces $V$ considerably only between $I_{C1}$ and $I_{C2}$ but shows no visible effect above $I_{C2}$ [see green curve in Fig. 5(a)], because $H$ can only increase $\theta$ via realigning the Ir moments below $I_{C2}$: Above $I_{C2}$, the saturation of $\Delta a/a$ corresponds to $\theta$ approaching 180°, which precludes further increases, and magnetic field can therefore no longer affect the I-V curves. The close association between $\Delta a/a$, moment canting, and the I-V curves reveals how current-controlled basal-plane expansion drives the nonlinear I-V characteristics.

Fundamentally, the formation of the Mott insulating state with canted IrO$_6$ octahedra and canted (locked) moments is caused by a cooperative transition in which the electronic structure gaps, thereby lowering its energy relative to the paramagnetic metallic state. The gapping mechanism involves electronic correlations that involve both spin-orbit coupling and scattering through the magnetic reciprocal lattice vector. The electronic correlation is expected to manifest itself in the unoccupied states (electron-carrier)
and in the occupied (hole-carrier) states. The momentum shift associated with a finite current is usually negligible in uncorrelated systems, but in correlated systems close to quantum critical points, theory shows that relatively small changes in the low-energy electronic structure can cause large (nonlinear) changes in the ordered structure. In short, a slight modification of the electronic structure induced by current may result in strong modifications of the electronic correlations. The NDR data are interpreted in terms of a reduction in the gapping, as suggested in Fig. 4(e), and the concomitant decrease in the carrier effective mass induced by current.

We have shown that a combination of strong SOI and canted AFM order can lead to a highly desirable paradigm for simultaneous electrical control of the crystal structure and physical properties of Sr$_2$IrO$_4$. (1) Strong SOI lock canted Ir moments to the IrO$_6$ octahedra, which rigidly rotate together (Fig. 2). (2) Strong SOI dictate the low-energy Hamiltonian and create small gaps in the electronic structure, which ultimately affect electron mobility (i.e., an increase in $\theta$ favors electron hopping; see Figs. 4 and 5). (3) Applied current effectively drives a lattice expansion by increasing $\theta$ and reducing small gaps in the electronic structure [Fig. 4(e)], which also reduces $T_N$ and the Ir moments (Fig. 3).

Our ongoing research on the physical consequences of strong SOI in materials suggests that similar behavior may be widespread in other iridates. This work offers a new paradigm for studies of the physics driven by the SOI and may help unlock a world of possibilities for functional materials and devices.

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*Corresponding author. 
gang.cao@colorado.edu


