The $J_{\text{eff}} = 1/2$ Antiferromagnet Sr$_2$IrO$_4$: A Golden Avenue toward New Physics and Functions

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Iridates have been providing a fertile ground for studying emergent phases of matter that arise from the delicate interplay of various fundamental interactions with approximate energy scale. Among these highly focused quantum materials, the perovskite Sr$_2$IrO$_4$, which belongs to the Ruddlesden–Popper series, stands out and has been intensively addressed in the last decade, since it hosts a novel $J_{\text{eff}} = 1/2$ state that is a profound manifestation of strong spin–orbit coupling. Moreover, the $J_{\text{eff}} = 1/2$ state represents a rare example of iridates that is better understood both theoretically and experimentally. Here, Sr$_2$IrO$_4$ is taken as an example to review the recent advances of the $J_{\text{eff}} = 1/2$ state in two aspects: materials fundamentals and functionality potentials. In the fundamentals part, the basic issues for the layered canted antiferromagnetic order of the $J_{\text{eff}} = 1/2$ magnetic moments in Sr$_2$IrO$_4$ are illustrated, and then the progress of the antiferromagnetic order modulation through diverse routes is highlighted. Subsequently, for the functionality potentials, fascinating properties such as atomic-scale giant magnetoresistance, anisotropic magnetoresistance, and nonvolatile memory, are addressed. To conclude, prospective remarks and an outlook are given.

1. Introduction

The electron–electron interaction (also known as Hubbard repulsion) ($U$) and spin–orbit coupling (SOC, measured by strength factor $\lambda$) are two critical fundamental ingredients that determine the electronic properties and functionalities of a quantum material.$^{[11]}$ The correlated electron materials have been greatly emphasized in 3d transition metal oxides where diverse magnetic textures, metal-insulator transitions, unconventional superconductivity, etc. were found. However, the SOC of 3d elements is weak and usually treated as perturbations to the electron correlations in materials ($\lambda \approx 0.01$ eV and $U \approx 5.0$ eV for 3d ions). One exception could be multiferroics with noncollinear magnetic configuration where SOC does not have to be large but still plays a dominant role in generating ferroelectricity.$^{[2,3]}$ It is known that SOC is effectively increased with atomic number, and consequently, emergent nontrivial quantum states can be induced. Topological insulators and semimetals are profound manifestations of the enhanced SOC.$^{[4,5]}$ In these extremely hot topics, the Hubbard repulsion $U$ is usually small (if not all), giving rise to a barrier to develop intrinsic magnetism.$^{[1,6]}$

Iridates certainly are another group of materials that show remarkably enhanced SOC ($\lambda \approx 0.5$ eV). Importantly and also differently, iridates in the meanwhile possess a moderate $U (=2.0$ eV), which provides a novel platform for cooperative effects of $U$ and SOC.$^{[7,8]}$ and this cooperation is unique, characterized by the fact that various exchange energy scales, Hubbard repulsion $U$, SOC, and other competing interactions, are comparable with each other. Theoretical investigations revealed that the delicate interplay of these fundamental interactions can cause a large array of novel electronic states, including the $J_{\text{eff}} = 1/2$ Mott state.$^{[9,10]}$ Weyl semimetals with Fermi arcs,$^{[5]}$ correlated topological insulator,$^{[3]}$ Kitaev spin liquid,$^{[11]}$ excitonic magnetism of pentavalent Ir$^{5+}$ (5d$^4$),$^{[12]}$ Probably due to the rapid developing nature of this field, there are just a few of these proposals that have been demonstrated experimentally, resulting in a huge gap between theoretical predictions and experimental findings. Among these, the $J_{\text{eff}} = 1/2$ Mott state represents a rare but unique example that has been better understood both theoretically and experimentally as well as mostly addressed in the past decade.

Since the 5d orbitals of Ir are more extended in comparison with 3d elements, a common perception assumes that iridates would be more metallic and less magnetic than the 3d compounds. However, since 1990s and early 2000s, a couple of magnetic and insulating iridates have been successfully synthesized,$^{[13-15]}$ and the observed results are distinctly different from our current consensus. This striking conflict was partially solved by identification of the $J_{\text{eff}} = 1/2$ picture in Sr$_2$IrO$_4$ in 2008.$^{[9,10]}$ As shown in Figure 1, because of crystal
field effect, the 5d orbitals of Ir are split into the fourfold degenerate $e_g$ orbitals and sixfold $t_{2g}$ orbitals, and the $e_g$ state takes a much higher energy than the $t_{2g}$ one. In this sense, the five electrons (Ir$^{4+}$) reside in the $t_{2g}$ orbitals with an effective orbital angular momentum $L = 1$. In the presence of strong SOC which acts within the $t_{2g}$ manifold as $-\lambda L \cdot S$, where $L$ is the effective angular momentum and $S$ is the spin moment, the $t_{2g}$ band is split into an effective $J_{\text{eff}} = 2$ doublet and an effective $J_{\text{eff}} = 3/2$ quartet. As a result, the $J_{\text{eff}} = 3/2$ band is fully occupied with four electrons due to its lower energy, and the energetically higher $J_{\text{eff}} = 1/2$ band is half-filled with one remaining electron. The $J_{\text{eff}} = 1/2$ band is thin enough, and a moderate $U$ can thus open a charge-gap ranging from $\approx 0.1$ to $\approx 0.5$ eV.\[16\]

The $J_{\text{eff}} = 1/2$ model has been successfully utilized to explain the insulating nature of many iridates,\[9,15,17-19\] and it works quite well although the ideal $J_{\text{eff}} = 1/2$ state is only expected for a perfect cubic symmetry. For instance, layered perovskites Sr$_2$IrO$_4$ and Sr$_2$Ir$_2$O$_7$ (Ruddlesden–Popper compounds, Sr$_{n+1}$Ir$_n$O$_{3n+1}$, $n = 1$ and 2) have been extensively discussed within the $J_{\text{eff}} = 1/2$ scenario, although theoretical calculations have found that the both show certain deviation from the ideal $J_{\text{eff}} = 1/2$ situation related to the tetragonal distortion.\[20\] Nevertheless, a deviation from a cubic crystal environment still exhibits non-negligible influence to the electronic properties, especially to magnetism, which has been a central issue within the domain of iridates researches.

Apart from the fundamental aspects, the $J_{\text{eff}} = 1/2$ state also shows tantalizing functionalities, which has been reported typically in the leading iridate Sr$_2$IrO$_4$ hosting a canted antiferromagnetic (AFM) structure.\[21–28\] Since Sr$_2$IrO$_4$ and La$_4$CuO$_4$ show couple of similarities in terms of structure and magnetic configuration, possible unconventional superconducting behaviors (even $p$-wave superconductivity and multipole superconductivity related to an odd parity magnetic hidden order) was frequently discussed.\[29–32\] This however remains elusive in experiments, and the two signature features of superconductivity, that is, zero resistance and diamagnetism, are still missed. Another noteworthy phenomenon is the significantly large magnetoresistance (MR) in Sr$_2$IrO$_4$ with a robust AFM order, unveiling tantalizing functionalities of the $J_{\text{eff}} = 1/2$ state toward the antiferromagnetronics (AFMtronics, referred to AFM spintronics in some occasions).\[21–28,33\] These phenomena have underscored the uniqueness of iridates with the $J_{\text{eff}} = 1/2$ state as promising and unique functional materials.

Surely, the family of iridates as a new group of quantum materials possess many more fascinating properties beyond the $J_{\text{eff}} = 1/2$ state, and there are indeed several review articles which cover this topic partially or to some extent.\[1,7,8,34\] In this progress report, we take Sr$_2$IrO$_4$ as representative material to address recent advances of the $J_{\text{eff}} = 1/2$ state, from fundamentals to functionalities, while so far no such framework has been touched with sufficient depth and breadth. The earlier reviews mainly covered the basic properties of iridates including Sr$_2$IrO$_4$,\[8,35\] here we focus on the novel functionalities associated with the $J_{\text{eff}} = 1/2$ state, including the SOC twisted antiferromagnetism and AFMtronics effects. This report is thus organized as follows. In Section 2, we illustrate the fundamental aspects of the canted AFM order with Sr$_2$IrO$_4$. In Section 3, we overview the recent advances in terms of the SOC twisted antiferromagnetism. In Section 4, we describe the distinctly large MR in Sr$_2$IrO$_4$, which underscores the uniqueness

Figure 1. A sketch of the $J_{\text{eff}} = 1/2$ model where the 5d orbitals of Ir$^{4+}$ are split by crystal field and spin–orbit coupling (which assumes $L \cdot S$ coupling), and finally a charge-gap is opened due to the Coulomb interaction. LHB and UHB represent the lower Hubbard band and upper Hubbard band, respectively.

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of Sr$_2$IrO$_4$ as one of AFM electronic materials. Remarkable and controllable anisotropic magnetoresistance (AMR) and nonvolatile memory effect in Sr$_2$IrO$_4$ are further discussed in Section 5. At the end of this progress report, concluding remarks and outlook, likely with our personal bias, will be presented. Given the rapidly evolving nature of this field, we certainly do not have any attempt to cover everything in this short article, and may miss some important results in this field.

2. $J_{\text{eff}} = 1/2$ Antiferromagnetism in Sr$_2$IrO$_4$

Sr$_2$IrO$_4$ is a single-layer perovskite, belonging to the Ruddlesden–Popper series Sr$_n$Ir$_2$O$_{3n+1}$ with $n = 1$.[36] It was reported to crystallize in a tetragonal structure with space group $I4_1/acd$. This assignment was challenged, and a reduced structural symmetry with space group $I4_1/a$ has been proposed.[37,18] The IrO$_6$ octahedra rotate about the $c$-axis by an angle $\alpha \approx 11^\circ$, leading to an expanded unit cell with $a = b = 5.4846$ Å and $c = 25.804$ Å.[37,39] The tetragonal distortion makes Sr$_2$IrO$_4$ deviate from the ideal $J_{\text{eff}} = 1/2$ situation which is only expected in a cubic crystal environment. Theoretical calculations found that the deviation is only $\approx 3%$.[39] confirmed by further nonresonant magnetic X-ray scattering (NRMXS) on Sr$_2$IrO$_4$.[40] In the meanwhile, the NRMXS experiments revealed a significantly enhanced orbital component of the $J_{\text{eff}} = 1/2$ magnetic moment (hereafter called the pseudospin), which is much larger than the ideal $J_{\text{eff}} = 1/2$ state.[40] Possible hybridization of $e_g$ and $t_{2g}$ states, and the weak Mott character were suggested as additional physical sources beside the elongated octahedral along the $c$-axis. According to the $J_{\text{eff}} = 1/2$ model, the pseudospin with equal contributions from the three $t_{2g}$ orbitals is isotropic and SU(2) invariant, and carries a magnetic moment of 1.0 $\mu_B$/Ir.[30] However, for Sr$_2$IrO$_4$, a much smaller magnetic moment $\approx 0.3$ $\mu_B$/Ir due to the extended orbital has been identified, and the SU(2) invariance is broken because of the tetragonal distortion.[37,41]

The pseudospins in Sr$_2$IrO$_4$, entangling both spin and orbital momenta due to the strong SOC at Ir-site, are aligned into an AFM lattice below the Néel temperature $T_N = 240$ K.[13,24,25,28] as illustrated in Figure 2. The absence of anomaly at $T_N$ in the electrical transport data is in agreement with the scenario of $J_{\text{eff}} = 1/2$ Mott state. All the pseudospins lie in the $ab$-plane, and show uniform deviation of $\approx 13^\circ$ from the $a$-axis. The canting is within the $ab$-plane and there is no canting along the $c$-axis. Various techniques, including neutron scattering and resonant X-ray scattering (RXS), demonstrated that the pseudospin canting rigidly tracks the rotation of IrO$_2$ octahedra, resulting in the well-known locking relation: $\alpha = \phi$.[37,39,41] This locking effect between the pseudospin canting and lattice distortion was reproduced theoretically by Jackeli et al.[42] Because of the canting, net magnetic moments appear alternatively in each IrO$_2$ planes, and are coupled antiferromagnetically along the $c$-axis. In this sense, Sr$_2$IrO$_4$ is fully compensated at the ground state without showing macroscopic magnetization. A magnetic field $H$ larger than the critical value $H_{\text{flop}}$ can cause a flop transition, and the net moments of IrO$_2$ layers are then ferromagnetically aligned along the $c$-axis, resulting in a weak ferromagnetic (FM) phase.[13,44] as sketched in Figure 2. This critical field $H_{\text{flop}}$ is fortunately not large ($\approx 0.1$ T), and it together with the flop transition makes Sr$_2$IrO$_4$ very unique. Consequently a number of emergent phenomena have been observed.
Benefiting from recent technological advances in resonant X-ray scattering, direct probing of the Ir pseudospins is allowed, noting that Ir has large neutron absorption ratio, which challenges related characterizations using neutron scattering technique.\cite{37} Extensive efforts in the last several years revealed that the AFM order is mainly stabilized by the strong nearest-neighbor (NN) intralayer AFM interaction of $\approx 0.1$ eV.\cite{45–47} Regarding the interlayer coupling, it was found to be as small as $\approx 1.0 \mu$eV, $10^5$ times weaker than the intra-plane AFM coupling.\cite{47} Such a large magnetic anisotropy allows the so-called flop transition, also known as the AFM to weak FM (AFM-wFM) transition, as demonstrated with RXS.\cite{43} The AFM order of Sr$_2$IrO$_4$ looks fairly robust and the AFM fluctuation with long-range in-plane correlation was found to persist at even $\approx 20$ K above $T_N$, which can be described by the 2D $S = 1/2$ quantum Heisenberg model.\cite{47} Another work addressing the critical behavior of Sr$_2$IrO$_4$ revealed that the critical scattering can be followed out to a much higher temperature ($T_N + 73$ K), and the XY anisotropy is important for accounting for the pseudospin interactions.\cite{48} High magnetic field experiments revealed that the AFM order does not show any trace of breakdown up to $H = 60$ T.\cite{25} The weak FM phase was found to be quenched at hydrostatic pressure $P \approx 20$ GPa, which is probably due to the reorientation of pseudospins from the $ab$-plane to the $c$-axis rather than the AFM order collapse.\cite{49}

While the NN interaction in developing the long range AFM order has been emphasized, some further next-order interactions with smaller energy scales seem to be non-negligible to the low energy magnetic properties of Sr$_2$IrO$_4$. By performing theoretical calculations and various microscopic measurements, Porras et al. revealed that pseudospin–lattice coupling is crucial to understand the static magnetism and low-energy pseudospin dynamics in Sr$_2$IrO$_4$.\cite{44} This was discussed and confirmed in the meantime by pure theoretical considerations.\cite{50} After incorporating the term of pseudospin–lattice coupling into the effective Hamiltonian, one may unravel many puzzles of Sr$_2$IrO$_4$ such as in-plane magnetic anisotropy and reduced structural symmetry. It is worthy to note that the in-plane magnetic anisotropy has been a long-term controversial issue, which is now clarified by improving crystal quality.\cite{25,43,51,52} This issue will be further discussed in the following, as it hosts emergent functionalities, that is, as an AFMtronics ingredient.

### 3. Tuning the Antiferromagnetism

#### 3.1. Tuning via Carrier Doping

In parallel to continuous interest in the $J_{\text{eff}} = 1/2$ antiferromagnetism, enthusiastic endeavors have been given also to carrier doping in Sr$_2$IrO$_4$, including electron- and hole-dopings, owing to not only the proposed unconventional superconductivity. Indeed, a bunch of features such as Fermi arcs, pseudogaps, and inhomogeneous electronic order have been observed,\cite{53–57} which are highly parallel to those seen in cuprates. However, the two signatures of superconductivity, that is, zero resistivity and diamagnetism, have not yet been observed. Recently, a hidden broken-symmetry phase was identified in Sr$_2$IrO$_4$, and Rh-doping (hole-doping) can enlarge the region of hidden phase in the phase diagram.\cite{38,58} Probably driving this hidden phase to a quantum critical point by heavier doping would be an approach to the missed superconductivity. Since electron-doping behaves more efficient than hole-doping in generating metallicity in Sr$_2$IrO$_4$, an interested question is how the hidden order would evolve upon electron-doping, which is still unexplored.

While the proposed superconductivity warrants further verification in experiments, the studies that have been made so far focusing on this topic have revealed interesting phenomena on the magnetism. The Neél temperature $T_N$ as a function of doping level for various dopants is shown in Figure 3. First, electron-doping and hole-doping in Sr$_2$IrO$_4$ show highly asymmetric effect on the AFM phase. For the electron-doping, that is, La-substitution at Sr-site, the long-range AFM order is rapidly lost at a doping level of only $\approx 3\%$, but the short-range correlated orders persist well into the metallic region.\cite{28,59–62} Further increasing the La-content slightly can cause a unidirectional spin density wave state.\cite{63} On the contrary, the long-range AFM order exhibits much more robustness against the hole-doping. For instance, in Sr$_3$Ir$_{1-x}$Ga$_x$O$_4$, the long range AFM phase can be well preserved, and $T_N$ just shows modest decrease from $\approx 240$ to $\approx 180$ K with increasing $x$ up to $\approx 9\%$.\cite{24} While Rh was demonstrated as hole-dopant, the AFM phase transition can still be detected even at a doping level as high as $x = 11\%$.\cite{64,65} The slight K-substitution of Sr can break the Mott state easily akin to electron doping effect, but it does not show any apparent impact onto the AMF order.\cite{28} Oxygen deficiency in materials is known to be equivalent to electron-doping. In Sr$_2$IrO$_4$, generating oxygen vacancies can break the Mott state quickly, but the canted AFM order remains nicely.\cite{66} This is similar to the K-doping effect (hole doping), but distinctly different from the case of La-doping (electron doping) which destroys both the charge-gap and the long-range AFM order simultaneously.

Second, the Ir-site substitution in Sr$_2$IrO$_4$ looks quite complicated. The 10% Mn-doping at Ir-site can flop the pseudospins from the basal plane to the $c$-axis, but does not show...
much effect on the magnetization and electric transport.\cite{67} Similar pseudospin reorientation has also been observed in \textit{Sr}_2\textit{Ir}_{1-x}\textit{Ru}_x\textit{O}_4.\cite{68} According to the model often used for magnetism in layered iridates,\cite{42,69} a modified interlayer coupling may be responsible for such a flop transition from the \textit{ab}-plane to the \textit{c}-axis. A slight isovalent substitution of \textit{Ir} with \textit{Tb} (≈3\%) causes the long-range AFM order to collapse, which was interpreted by a compass impurity model.\cite{70,71} These works have shown the important role of magnetic cation doping at \textit{Ir}-site. It has been found that the Ga- and Rh-doping can also trigger the AFM-wFM transition,\cite{24,72} resembling the case driven by magnetic field \textit{H}. Theoretical calculations suggest that a hole-doping may be important for the in-plane flopping transition.\cite{73} However, similar effect was recently reported in \textit{Sr}_2\textit{Ir}_{1-x}\textit{Sn}_x\textit{O}_4 where \textit{Sn} is expected to take a tetravalent state as \textit{Ir} \textit{IV}+\textit{74}.

Third, substitution of \textit{Sr} with isovalent cation such as \textit{Ca} or \textit{Ba} is expected to cause lattice distortion only, without implanting charge carriers and additional magnetic perturbations. The Ca-doping tends to shrink the lattice, while the Ba doping prefers to tune the \textit{Ir}-O-\textit{Ir} bond angle,\cite{75} which can be increased by ≈1° due to 4\% Ba-doping, and 15\% Ca-doping can shrink the cell volume by ≈1\%. It is noted that the electronic band structure of \textit{Sr}_2\textit{IrO}_4 is correlated with the \textit{Ir}-O-\textit{Ir} bond angle, and indeed a sudden drop of the in-plane resistance at low temperature has been observed for the sample with 2\% Ba. Regarding the magnetic properties, no apparent change of the Néel temperature \textit{T}_\textit{N} with the Ca- or Ba-doping can be seen as shown in Figure 3. Nevertheless, the Ca-doping can evidently reduce the ratio of the out-of-plane magnetization to the in-plane one, \textit{M}_\textit{a}/\textit{M}_\textit{c}, indicating the suppressed magnetic anisotropy.

It should be mentioned that one of the major difficulties for chemical substitution in \textit{Sr}_2\textit{IrO}_4 is the limited tolerance for doping, which hinders the role of carrier density modulation. Alternative approaches include the field effect transistor (FET) scheme which is one of the most elegant ways to inject carriers into materials without generating parasitic effects like quenching disorder. This scheme with a modified structure, that is, electric-double-layer transistor (EDLT) with electrolyte as the gating layer, has been recognized as a very powerful method to tune the carrier density.\cite{77} Recently, the EDLT with \textit{Sr}_2\textit{IrO}_4 as the channel layer was successfully fabricated to tune the \textit{J}_{\textit{eff}} = 1/2 state,\cite{33,78} as shown in Figure 4. Indeed, the carrier density in the \textit{Sr}_2\textit{IrO}_4 layer can be drastically enhanced up to ≈4 \times 10^{15} \text{cm}^{-2}, about two orders of magnitude higher than that with the non-gated sample.\cite{33} Unfortunately, no superconductivity was yet detected and the transport data show insulating behavior. It should be mentioned that this carrier density is already higher than that in a lot of superconductors.\cite{79} On the other hand, the enhanced carrier density did evidence significant impact on magnetotransport such as standard MR and AMR, indicating the gating-modified magnetic properties in \textit{Sr}_2\textit{IrO}_4.\cite{33,78} Furthermore, a linear relationship between conductivity and channel thickness implies a bulk gating effect of the EDLT structure.

**Figure 4.** a) A sketch of the field-effect transistor with \textit{Sr}_2\textit{IrO}_4 where the gate layer is an ionic liquid. b) The temperature dependence of resistance \textit{R}_\textit{S} under various gating voltage \textit{V}_\textit{G}. c) The charge carrier density and mobility as a function of gating voltage \textit{V}_\textit{G}. d) Magnetoresistance under \textit{V}_\textit{G} = 0 and −2.0 V at \textit{T} = 35 K. e,f) present the anisotropic magnetoresistance (AMR) measured under \textit{V}_\textit{G} = 0 V and −2.0 V at \textit{T} = 35 K, respectively. The dashed and solid lines in (f) represent the AMR trace and retrace. b–d) Reproduced with permission.\cite{33} Copyright 2014, American Physical Society.
Besides the chemical doping, Sr$_2$IrO$_4$ offers another roadmap of tunability, considering the locking effect $\alpha = \phi$, which suggests another idea to engineer the AFM order of epitaxial thin films via lattice strain. This idea was initially examined theoretically, and indeed remarkable strain effects on magnetism were revealed. Upon a strain, the ground state can be shifted toward or from the SU(2) point, and the canting angle of pseudospins and the $c/a$ ratio of IrO$_6$ octahedra are all linearly proportional to strain. As shown in Figure 5, when parameters $c/a$ and $\alpha$ are small, an $ab$-plane collinear AFM state appears. Increasing the $c/a$ ratio would rotate the pseudospins from the $ab$-plane to the $c$-axis, yet keeping the collinear AFM configuration. However, upon the increased rotation of IrO$_6$ octahedra, the pseudospins are expected to be clearly canted because of the locking effect $\alpha = \phi$. The electronic structure is predicted to be sensitive to strain, evidenced by the enhanced electron correlation and spin–orbit coupling with increasing strain, while the response of electronic structure to strain is highly correlated with the direction of magnetic moment. The density-functional calculations do suggest the strong strain-dependence of transport properties for hole-doped Sr$_2$IrO$_4$.

Experimentally, it was found that the Néel point $T_N$ depends remarkably on strain, and $T_N$ can be drastically enhanced (reduced) by tensile (compressive) strain in Sr$_2$IrO$_4$ epitaxial films. In comparison with the bulk counterpart, $T_N$ of the (001) Sr$_2$IrO$_4$/SrTiO$_3$ thin films with a $\sim 0.5\%$ tensile strain is enhanced by $\sim 30\%$. The underlying physics can be ascribed to the renormalization of interlayer and intralayer interactions due to the structural engineering by strain. However, this observation was questioned by other reports on the same system where the observed $T_N$ is even lower than that of the bulk counterpart. Similar inconsistency can be found for the electrical transport data. In fact, for bulk Sr$_2$IrO$_4$, a tiny La-doping is sufficient to drive the insulating to metallic transition, and this transition is understandable if one considers the small band gap of the $J_{\text{eff}} = 1/2$ state. However, the insulating ground state in epitaxial thin films seems to be highly robust even at a much higher doping level. Another example to show such inconsistency is (Sr$_{1-x}$Ba$_x$)$_2$IrO$_4$ which in the bulk form shows strong metallic behavior at $x = 0.02$. The intriguing insulating transport remains robust even at very high Ba-content ($x = 37.5\%$).

It should be mentioned that pure Sr$_2$IrO$_4$ phase can only be stabilized within a relatively narrow growth window and the electronic properties show high sensitivity to chemical environment (i.e., defects and interfaces). In fact, even for high quality Sr$_2$IrO$_4$ epitaxial films, crossover behavior in terms of conduction mechanism upon varying thickness and temperature was observed, as shown in Figure 6. Nevertheless, one has to admit that high doping concentration is allowed in thin film samples, a favored advantage that is not accessible in the bulk. In agreement with theoretical calculations, the strain modified electron correlation and electronic band structure were experimentally revealed in Sr$_2$IrO$_4$ thin films grown on various substrates, while such agreement does not happen for the bulk crystals.

### 3.3. Tuning via Electric Current

Besides the strain engineering, another efficient route to tune the lattice of Sr$_2$IrO$_4$ is the application of large electric current which was recently addressed by Cao’s group, an unusual tunability. It was found that the current induced variations along the $a$-axis and $c$-axis are anisotropic, and the $a$-axis expansion can be as large as $\sim 1\%$ if the current reaches a high level, as shown in Figure 7. More importantly, in accompanying with the huge in-plane lattice expansion, remarkable reductions in both $T_N (\Delta T_N = 40\, \text{K})$ if the current $I$ is as high as $\sim 80\, \text{mA}$ and in-plane magnetization ($\Delta M_a = 0.012\, \mu_B$/Ir at $H = 7\, \text{T}$ and $I = 100\, \text{mA}$) have been observed. The simultaneous modulation of lattice and magnetization can be understood in the framework of pseudospin–lattice locking ($\alpha = \phi$). In the meanwhile, other unusual transport behaviors were also observed when the current was huge, including the negative differential resistance and reversible resistance switching. The large current effect demonstrates the potentials for an electro-control of the $J_{\text{eff}} = 1/2$ state in Sr$_2$IrO$_4$, although the underlying physics remains elusive.

### 3.4. Superlattice Tuning

Finally, one may be noticed that a striking feature of Sr$_2$IrO$_4$ is the layered magnetic configuration with alternatively stacked IrO$_2$ and SrO layers, in which the cooperation between the interlayer coupling and intralayer interaction determines the magnetic properties. Such layered magnetic structure can be artificially synthesized and engineered, owing to the advance of thin film fabrications. One typical example is the SrTiO$_3$/SrIrO$_2$ multilayers superlattice, where the TiO$_2$ layer is used to replace the IrO$_2$ layer, so that the IrO$_2$ interlayer distance can...
be enlarged for weakening the interlayer coupling and suppressing $T_C$. As expected, an increasing thickness of TiO$_2$ layer sandwiched by two IrO$_2$ layers moves $T_C$ down to low-$T$ side significantly, evidencing the pivotal role of interlayer coupling for the long-range AFM order. In the case of $m = 2$, the superlattice behaves more like a 2D antiferromagnet which can be effectively controlled by magnetic field $H$. As shown in Figure 8, the AFM state can be switched on and off by a magnetic field as small as $H < 0.5$ T. Another observation for this case is the weak FM characteristic in the $m = 1$ superlattice, resembling the cases triggered by magnetic field $H$ or Ir-site chemical substitution. It can be concluded that a modulation of the IrO$_2$ interlayer coupling may be crucial for the appearance of weak FM phase. An atomic scale synthesis of the IrO$_2$ layered magnetic structure has been often utilized as an effective tool for cruising fantastic phenomena, which nonetheless are non-accessible to the AFM nature of Sr$_2$IrO$_4$ in terms of the pseudospin alignment and that the magnetic IrO$_2$ layer is atomically thin, different from the artificial GMR heterostructures.

The quest for large MR effect in antiferromagnets has been intensively discussed within the domain of AFM spintronics in the past years. This was initially exploited in AFM heterostructures, that is, AFM/nonmagnetic-spacer/AFM, in analogy to traditional magnetic multilayers. The AFM counterpart to the GMR, called the AFM-GMR, was theoretically proposed in such AFM structures, and it was found to be one-to-one correspondence with the observed $M(H)$ loop, suggesting the magnetic order origin for the magnetotransport in Sr$_2$IrO$_4$. Notable MR effect can also be seen if current flows within the $ab$-plane (simply called the in-plane MR), but it is clearly smaller than out-of-plane MR as shown in Figure 9c,d. Such difference between the in-plane and out-of-plane MR data was also observed in traditional GMR structures, where the difference between the out-of-plane and in-plane MR data was also observed in traditional GMR structures. The atomic scale GMR-like effect observed in Sr$_2$IrO$_4$ may share similar physics. Nonetheless, we should bear in mind the AFM nature of Sr$_2$IrO$_4$ in terms of the pseudospin alignment and that the magnetic IrO$_2$ layer is atomically thin, different from the traditional GMR heterostructures.

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where the function of temperature under high electric current $I$ in SrTiO$_3$ (001) substrate as the best choice to preserve the films in comparison with bulk single crystals even though observe sharp and complete GMR-like effect in Sr$_2$IrO$_4$ thin films easily destroyed by external perturbations. It is difficult to destruction of the intrinsic AFM order, and thus can be just originates from the pseudospin flop rather than aelastic properties and emergent functions.

The MR effect driven by the out-of-plane magnetic field ($H_{/c}$-) also deserves for some discussion, while it looks a bit more complicated. First, whether the flop transition can be triggered by $H_{/c}$ is still under debates. Second, multiple step-like anomalies in the MR curves were observed in the high field region ($H \approx 2 T > H_{\text{flop}}$), but no slope change can be observed in $M(H)$ correspondingly. Therefore, the magnetic scattering scenario may be excluded or it is not the dominant one at least. While the intriguing MR effects for $H_{/c}$ remain elusive, $H$-driven lattice distortion may be relevant noting the fact that the pseudospin–lattice coupling is essential to describe the magnetism in Sr$_2$IrO$_4$. Striking magneto-dielectric response was reported in the high field region, supporting the argument of strong pseudospin–lattice (phonon) coupling in Sr$_2$IrO$_4$. Third, the MR magnitudes for the two geometries ($H_{/c}$- and $H_{/a}$-axis) are similar, but the critical fields for pseudospin flop are very different. In addition, so far reported data on the MR data in the $H_{/c}$-axis geometry seem to be authors-dependent, an issue to be clarified.

5. Anisotropic Magnetoresistance toward Antiferromagnetronics

The atomic-scale GMR-like effect has illustrated the uniqueness of Sr$_2$IrO$_4$ as an AFMtronics candidate. More than this, a system with a novel $J_{\text{ir}} = 1/2$ state does host more exciting functionalities, including remarkable and controllable anisotropic magnetoresistance (AMR) and nonvolatile memory. The AMR effect is the magnetotransport counterpart of the relativistic energy anisotropy, a powerful and widely utilized function for detecting reorientation of magnetic moment typically in FM materials. Recently, this effect was demonstrated in some AFM materials since it is an even function of magnetization. A seminal work on this topic is the observation of large tunneling AMR in IrMn-based AFM tunnel junctions, and the essential role of large SOC in inducing such AFM-based AMR (AFM-AMR) effect has been emphasized.

Along this line, it was reported that the Sr$_2$IrO$_4$/La$_{2/3}$Sr$_{1/3}$MnO$_3$ heterostructure indeed exhibits AMR effect up to $15\%$, where the FM La$_{2/3}$Sr$_{1/3}$MnO$_3$ layer was used to control the AFM lattice of Sr$_2$IrO$_4$ through the interface coupling. Subsequent experiments with Sr$_2$IrO$_4$ bulk crystals found that FM buffer layer is not necessary to generate AMR, because Sr$_2$IrO$_4$ has the nonzero net magnetization at $H > H_{\text{flop}}$. It is believed that realizing the AMR effect in a pure AFM material without any auxiliary reference layer is an
Figure 8. a) The paramagnetic to antiferromagnetic transition under various magnetic fields, where $T_0$ indicates the crossover temperature. b) By switching on/off magnetic field $H = 0.2$ T at $T = 50$ K, the magnetic peak intensity of the AFM phase shows the corresponding response. a,b) Reproduced with permission.[99] Copyright 2018, Springer Nature.

Figure 9. a–c) Magnetoresistances (MR$_c$ with current along the $c$-axis and MR$_{ab}$ with current along the $ab$-plane) and in-plane magnetization $M$ as a function of $H$ measured at: a) $T = 35$ K, b) $T = 50$ K, c) $T = 90$ K. d) For a comparison, the MR$_{ab}$ data with the in-plane current are also plotted. e,f) The MR data of (001) Sr$_2$IrO$_4$/SrTiO$_3$ thin films (e) and (001) Sr$_2$IrO$_4$/NdGaO$_3$ thin films (f) at different temperatures. e,f) Reproduced with permission.[26] Copyright 2014, American Physical Society.
The AFM-AMR was also identified in Sr$_2$IrO$_4$ thin films grown on (001) SrTiO$_3$ substrates without reference layer.[23,26,33] Because of the good lattice fit between Sr$_2$IrO$_4$ and SrTiO$_3$, it is allowed to preserve the spin–orbit coupled AFM order in the epitaxial thin films. The MR curves with $H$ applied along the [100] (easy axis) and the [110] (hard axis) directions present an intriguing intercross in the high field region, indicating unconventional AMR in the films.[23] This was further confirmed by the detailed $R(\Phi)$ measurements under various $H$, as shown in Figure 10. At low field, the fourfold crystalline AMR with minima at the easy axes is presented. Upon increasing $H$, the AMR contour is rotated by $\approx 45^\circ$, indicating that the pristine fourfold AMR minima are now changed to the maximal positions in the induced fourfold AMR. This AMR contour rotation can be even observed in the high-$T$ range close to $T_N \approx 240$ K. The first-principles calculations revealed the different charge-gaps as the pseudospins point along the easy- and hard-axes. For instance, a larger charge-gap is expected when the pseudospins are aligned along the easy axis, in comparison with the case of pseudospins along the hard axis. This is consistent with the $J_{\text{eff}} = 1/2$ picture in which the strong SOC is essentially involved in developing electronic structure of Sr$_2$IrO$_4$.

Similar AMR contour rotation was also observed in Sr$_2$IrO$_4$ bulk crystals while the critical field is smaller. Surprisingly, a giant AMR ratio reaching $\approx 160\%$ was identified in bulk Sr$_2$IrO$_4$, which is a record in the field of AFMtronics.[25] The MR curves show sudden change at low field associated with the pseudospin flop transition, as discussed in Section 4. As shown in Figure 11, the critical fields for the MR-jumps show clear difference when $H$ is applied along the easy axis (the [100] direction) and in-plane hard axis (the [110] direction), leading to a window in this region. The difference in measured $R$ for the two cases can be as large as $\Delta R \approx 38$ k$\Omega$ at $T = 35$ K, a value larger than the most reported values in the literature, including those in semiconductors and tunnel junctions which usually show relatively large $\Delta R$ and AMR. By plotting the AMR ratio as a function of $H$, one could easily see that the remarkable AMR enhancement is tightly related to the flop transition.

There are two factors pivotal for the observation of large AMR in Sr$_2$IrO$_4$. One is that the flop transition has to be sharp and complete, which promises a giant MR. This looks sensitive to external perturbations such as chemical doping and strain in thin films. The other one is the in-plane magnetic anisotropy, allowing us to have a window between the MR curves
with $H_{//[100]}$ and $H_{//[110]}$. The in-plane magnetic anisotropy of Sr$_2$IrO$_4$ has been a missed issue until very recently. In previous studies focusing on the exploration of AFM-AMR, magnetocrystalline anisotropy has been considered solely to realize AMR in antiferromagnets, and the observed AMR ratio is unfortunately small overall.[107,108,116] Therefore, the observation of giant AMR in Sr$_2$IrO$_4$ has illustrated an efficient mean, that is, combination of various mechanisms, to enhance the AFM-AMR remarkably.

By tiny Ga-doping (1.0%) at Ir-site in Sr$_2$IrO$_4$, nonvolatile memory was demonstrated, and the resistive ratio of two memory states can be as large as 4.5%,[25] as shown in Figure 12. The AFM-based memory phenomena have been identified in several antiferromagnets such as MnTe and FeRh, and a heat assist magnetic recording method was generally used to manipulate the memory states.[113,114] For instance, in order to set the memory states in FeRh which represents a rare room temperature AFM memory resistor, the sample has to be warmed up above $T_N$, and then cooled down with $H$. In Sr$_{1.99}$Ga$_{0.01}$IrO$_4$, the memory states can be switched in situ by changing the direction of $H$, and the memory effect is nonvolatile and fully reproducible in the successive write–read cycles, which looks more compatible with realistic devices.

Increasing Ga-doping content in Sr$_2$Ir$_{1-x}$Ga$_x$O$_4$ induced metallic transport behavior, while the canted AFM order was maintained just showing gentle decrease in $T_N$ from 240 to 180 K.[24] Note that it seems a bit difficult to successively tune electric transport from insulating to metallic without breaking the pristine magnetic order in AFM materials. Upon Ga-doping, the fourfold crystal AMR symmetry can be well preserved, although its magnitude is reduced from $\approx 16\%$ to 1.0%, as shown in Figure 13. The suppression of AMR magnitude is much more obvious when samples host insulating transport. In the samples with $x > 0.05$ showing metallic behavior, the AMR magnitude evolves with $x$ steadily at a level of $\approx 1.0\%$ at low temperature. This is in fact a relatively large AMR ratio in comparison with other metallic AFM materials.

6. Final Remarks and Outlook

To this end, we have presented a brief overview on the recent progress of Sr$_2$IrO$_4$ with novel $J_{\text{eff}} = 1/2$ state, addressing the magnetic ground state, fantastic electronic properties, canted antiferromagnetism, and related magnetotransport behaviors. Sr$_2$IrO$_4$ represents a leading iridate that has been intensively addressed in recent years. Although the predicted unconventional superconductivity has not yet been observed, a bunch of fascinating phenomena have been identified, including low energy excitations, Fermi arcs, pseudogap, electronic inhomogeneity, and broken symmetry phases. In the meanwhile, our understanding of the quasi 2D
antiferromagnetism associated with the $J_{\text{eff}} = 1/2$ moments has been advanced largely. Apart from the metal-insulator transition, the highly tunable layered AFM phase and associated phenomena such as spin-density wave, time-reversal symmetry breaking hidden order, and pseudospin flop transitions have been identified. Especially, the observations of atomic scale GMR-like effect, remarkable and controllable AMR, and nonvolatile memory have highlighted Sr$_2$IrO$_4$ as a promising AFMtronic candidate.

While these observations on one hand underscore the uniqueness of Sr$_2$IrO$_4$ in both physical and functional aspects, a large array of focusing issues remain elusive on the other hand. One of the most attractive topics is experimentally yet missed unconventional superconductivity in carrier-doped Sr$_2$IrO$_4$. Although many strategies have been proposed, such as pushing the hidden phase into its quantum critical point, increasing chemical doping concentration effectively, efficient means to approach the elusive superconducting state remain yet unexplored. This is obviously a very meaningful issue, and extensive works are required to fill the huge gap between the related theories and experiments. Regarding the canted AFM phase, it can now be more precisely described using a modified Heisenberg Hamiltonian. However, after involving stimuli such as dopants, strain, and electric field, things are going complex and no certain tendency and principle can be concluded. For instance, in some cases, the long-range order collapses simultaneously when the Mott gap is closed, indicating the close correlation between charge and magnetic degrees of freedom. This is consistent with the close energy scale between the exchange ($\approx 0.1$ eV) and charge gap ($\approx 0.5$ eV). However, there are still a few of experimental results showing the decoupled long-range order and insulating state.

The demonstration of AFMtronic functionalities made Sr$_2$IrO$_4$ even more tantalizing, distinguishing this material from other iridates. Obviously, the magnitude of MR and AMR are already sufficiently large. However, these superior properties are more frequently observed in low temperature range. For all iridates that have been discovered, one may fall frustrated indeed, since the highest reported Néel point is just $\approx 280$ K in Sr$_3$Ir$_2$O$_7$. Nevertheless, it has been reported that $T_N$ can be enhanced significantly by simply enhancing the interlayer coupling. Another interesting phenomenon is the nonvolatile memory in Sr$_2$IrO$_4$ with 1% Ga-doping. Again, the operating temperature is low. Although these works have shown the fascinating aspects of Sr$_2$IrO$_4$, there is much room for utilizing these functionalities in practical devices.

Thin film fabrication is a key step to integrate this leading iridate, Sr$_2$IrO$_4$, into practical devices, and it has been revealed that the electronic properties of Sr$_2$IrO$_4$ can be efficiently tuned through strain indeed. However, related experiments are largely unexplored. One possible reason is the lack of high quality samples. High quality pure phase Sr$_2$IrO$_4$ can only be stabilized within a narrow growth window deposition. In fact, even for Sr$_2$IrO$_4$ bulk crystals, the results from different groups can be somehow scattering. For instance, the step-anomaly appearing in the $M(H)$ curves with $H//c$-axis is not yet understood. In some cases, strong metallicity can arise in La-doped Sr$_2$IrO$_4$. However, qualitatively different electric transport behavior with clear upturn in the $R(T)$ curve at low temperature has also been reported. These inconsistent results suggest the important role of sample quality, which has been much less focused but surely deserves special attention.
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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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Figure 13. a) A sketch of the AMR measurements for Sr$_2$Ir$_{1-x}$Ga$_x$O$_4$ crystals ($0 \leq x \leq 0.09$), where the current is applied along the c-axis and $H$ is rotated within the ab-plane. b) The AMR magnitude as a function of Ga-doping content. c) The fourfold AMR symmetry is observed for all samples with different Ga-content. d) The AMR ratio as a function of $T$ for different Sr$_2$Ir$_{1-x}$Ga$_x$O$_4$ crystals. b,d) Adapted with permission. Copyright 2018, American Physical Society.
