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Anomalous magnetic and transport behavior in the magnetic insulator $Sr_3Ir_2O_7$

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We report magnetic and transport properties of single-crystal $Sr_3Ir_2O_7$, a bilayered orthorhombic system with Bbca symmetry. Our study reveals that $Sr_3Ir_2O_7$ is a ferromagnetic insulator with a Curie temperature of $T_C = 285$ K and a small ordered magnetic moment. The saturation moment μ_S is found to be less than $0.04\mu_B$/Ir or 4% of the expected moment for a localized spin $S=\frac{1}{2}$. For fields larger than 1 T the magnetic susceptibility is the superposition of a Curie law (with negligible Weiss temperature) and a constant term. Surprisingly, at low temperatures (<20 K) the magnetization is *opposite* to the applied magnetic field when $Sr_3Ir_2O_7$ is cooled in a modest magnetic field ($\leqslant 0.2$ T). The resistivity $\rho(T)$ measured from 1.7 to 1000 K along two principal crystallographic directions exhibits insulating behavior with anomalies corresponding to the magnetic properties. Both the magnetization and the resistivity are strongly anisotropic as a consequence of the layered structure. Possible crystalline field and band scenarios are briefly discussed.

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I. INTRODUCTION

Although $4d$-electron based ruthenates have drawn increasing attention in recent years, the $4d$- and $5d$-electron transition-metal oxides are mostly still an unexplored territory, rich in novel physical phenomena that often deviate from conventional expectations. In this paper, we present the anomalous magnetic and transport behavior displayed by the $5d$-electron based $Sr_3Ir_2O_7$, which provides another example illustrating the intriguing and complex physics in the $4d$ and $5d$ transition-metal oxides.

An important feature characteristic of $4d$ and $5d$ electrons is the large spatial extent of d-electron orbitals. The extension of the orbitals enhances crystalline field splittings and the $4d$ or $5d$ $2p$ hybridization between the transition metal and the oxygen octahedron surrounding it. This also leads to strong electron-lattice coupling, which can alter and distort the metal-oxygen bonding lengths and angles, in this way lifting degeneracies of narrow bands and gaps. It is this feature that is at the heart of the novel physical phenomena in the transition-metal oxides. This point is well illustrated in recent studies on the $4d$-based ruthenates, whose behavior departs from that for most $3d$-electron based materials.[1–5] Like the ruthenates, many of the iridates ($5d$ electrons) also defy the notion that $5d$ transition-metal oxides should be much more conducting than their $3d$ counterparts because of the more extended $5d$ orbitals that significantly reduce the Coulomb interactions. Many of the iridates known to date are nonmetallic and display exotic magnetism with high magnetic ordering temperatures and yet small magnetic moments.[6–15]

This study reveals that $Sr_3Ir_2O_7$ is a magnetic insulator with a transition to weak ferromagnetism at $T_C = 285$ K. The magnetic moment at 2 K and 7 T is only $0.037\mu_B$/Ir, i.e., less than 4% of the moment expected for localized spin $S = \frac{1}{2}$. The field-cooled magnetization at constant temperature has a rapid increase as a function of field at low fields ($B < 0.5$ T) and at higher fields it follows a Curie law (with very small Weiss temperature) in the background of a small temperature-independent susceptibility. A low ordered magnetic moment and a high transition temperature have previously been observed in the sister compound Sr_2IrO_4.[13] The resistivity displays insulating behavior up to 1000 K with a temperature dependence that reflects the magnetic anomalies (magnetic scattering). The resistivity is insensitive to the magnetic field except at very low temperatures where a weak dependence is observed in the field-cooled case. Consistent with the quasi-two-dimensional structure, $Sr_3Ir_2O_7$ displays a large anisotropy in the resistivity and in the magnetization.

The most striking phenomenon in $Sr_3Ir_2O_7$ is a magnetization reversal in the basal plane below 20 K under *field-cooled* (FC) conditions. This means the system has an anomalous ‘‘diamagnetic’’ response, with the magnetization pointing opposite to the magnetic field. In contrast, no anomaly is discerned in the entire measured temperature range under *zero-field-cooled* (ZFC) conditions. The magnetization reversal is gradually suppressed with increasing magnetic field and eventually, when the magnetic field is large enough ($0.2 < B < 4$ T), the weak ferromagnetic state is restored.[19] The observed magnetoresistivity at low temperatures seemingly mirrors the magnetization reversal.

The rest of the paper is organized as follows. In Sec. II we

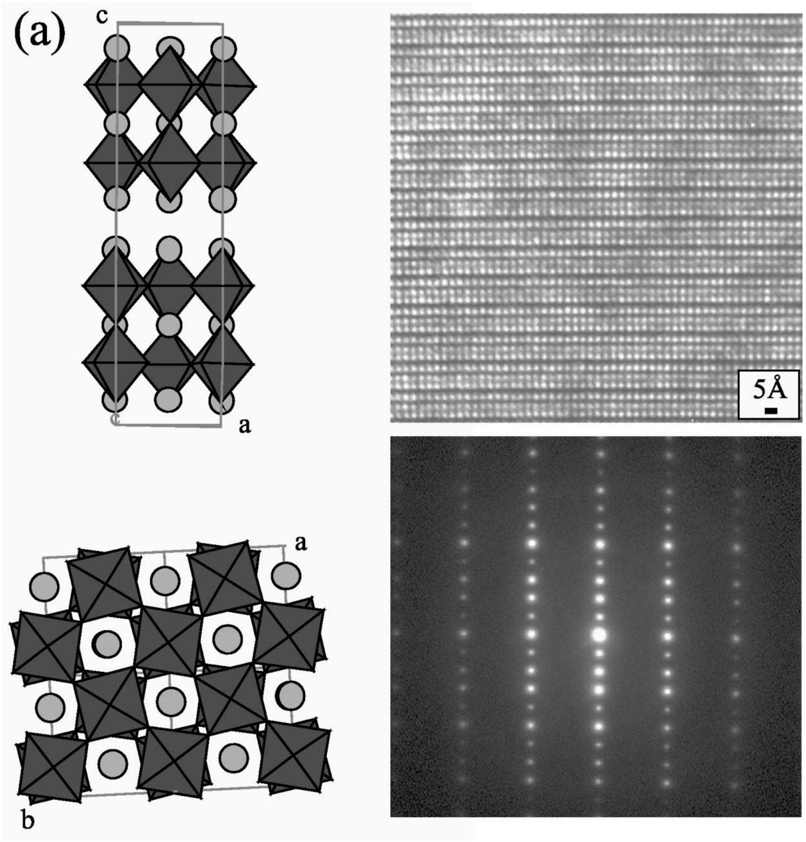

FIG. 1. (a) Diagram of the $Sr_3Ir_2O_7$ unit cell projected on the *ac* plane (upper) and the *ab* plane (lower). Note the rotation of IrO_6 about the *c* axis and that subsequent bilayers are displaced by half a lattice spacing. (b) A high-resolution TEM image of the crystal along the *a* axis showing a perfect stacking of the atomic layers: (c) A [100] x-ray-diffraction pattern.

briefly summarize the experimental methods used. The results are presented in Sec. III. The discussion of possible scenarios for the properties of the ferromagnetic insulator are given in Sec. IV and concluding remarks are presented in Sec. V.

II. EXPERIMENT

Single crystals were grown in Pt crucibles using self-flux techniques from off-stoichiometric quantities of IrO_2, $SrCO_3$, and $SrCl_2$. These mixtures were heated to 1480 °C in Pt crucibles, fired for 20 h, cooled at 5 °C/h to 1440 °C, and then rapidly quenched to room temperature. It is found that the quenching process is critical to ensure an exclusion of precursor phases such as Sr_2IrO_4, which is a stronger magnet with $T_C = 240$ K. It needs to be pointed out that a slight inclusion of the Sr_2IrO_4 phase could fully overshadow the diamagnetic response, or eliminate the negative magnetization by lifting the magnetic background although temperature dependence remains unchanged below 50 K. A good indication of the inclusion of Sr_2IrO_4 in $Sr_3Ir_2O_7$ is an anomaly near $T_C = 240$ K in ZFC magnetization. ZFC magnetization for pure $Sr_3Ir_2O_7$ shows no features in that temperature range. The resulting shapes of $Sr_3Ir_2O_7$ are platelike with an average size of $0.6\times0.5\times0.1\ \mathrm{mm}^3$ with the shortest dimension along the *c* axis. The crystal structure of $Sr_3Ir_2O_7$ has been refined by the single-crystal x-ray diffraction performed at the Experiment Station, du Pont. The single crystals studied were also characterized by transmission electron microscopy (TEM) using a JEOL 2010 microscope operated at 200 kV. The composition of the crystals was examined by energy-dispersive x-ray (EDX) spectroscopy, confirming the ratio of Sr:Ir to be 3:2. The electrical resistivity was measured with a standard four-probe technique and the magnetization with a commercial superconducting quantum interference device (SQUID) magnetometer. All crystals used in this study were as-grown.

III. RESULTS

$Sr_3Ir_2O_7$ has strongly coupled double Ir-O layers with adjacent double layers offset along the *c* axis and separated by Sr-O interlayers [see Fig. 1(a)]. The system belongs to the Ruddlesden-Popper series $Sr_{n+1}Ir_nO_{3n+1}$ with $n=2$. The compound was reported about 30 years ago.[16] The only previous crystal structure study of $Sr_3Ir_2O_7$ (Ref. 17) reported a tetragonal cell ($a=3.896$ and $c=20.879$ Å) with space group $I4/mmm$.

Our newly refined crystal structure determined by single-crystal x-ray diffraction reveals an orthorhombic cell with $a=5.5221$ Å, $b=5.5214$ Å, and $c=20.9174$ Å with a Bbca symmetry.[18] The refinement shows that the IrO_6 octahedra are elongated along the crystallographic *c* axis, i.e., the Ir-O bond distances are in average 2.035 Å (apical, along the *c* axis) and 1.989 Å (in the IrO planes). The octahedra are rotated about the *c* axis by 11° at room temperature. It is found that within a layer the rotations of the octahedra alternate in sign, leading to a staggered structure, with between the two layers of a double layer being out of phase to each other [see Fig. 1(a)]. The x-ray-diffraction data display su-

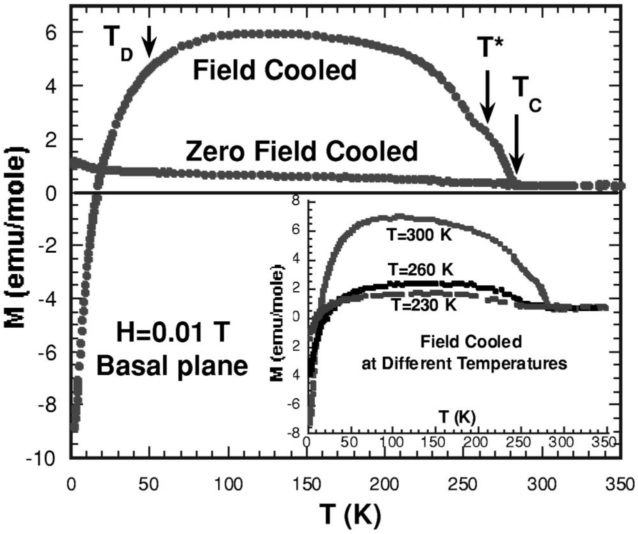

FIG. 2. Magnetization M for the basal plane as a function of temperature at 100 Oe. Inset: M vs T measured using the FC sequence that starts at $T=300$, 260, and 230 K, respectively.

perlattice reflections due to the presence of correlated rotations of the IrO_6 octahedra. The previous crystal structural study[17] reported similar rotations, which, however, were believed to be noncorrelated because no superlattice reflections could be discerned.

Also shown in Fig. 1 is a high-resolution TEM image along the *a* axis [Fig. 1(b)] that illustrates a perfect stacking sequence of atomic layers of $Sr_3Ir_2O_7$. The TEM diffraction pattern along the *a* axis shown in Fig. 1(c) displays a characteristic single-crystal pattern that is consistent with the newly refined crystal structure described above. No defects, such as planar defects or second phases such as Sr_2IrO_4 were discerned in the crystals studied.

Shown in Fig. 2 is the magnetization M as a function of temperature T for the basal plane. The zero-field-cooled (ZFC) M was measured on heating at an applied field $B=0.01$ T after cooling in zero magnetic field to 1.7 K from room temperature whereas the field-cooled (FC) M was obtained on cooling at $B=0.01$ T from room temperature (or different temperatures if specified). It is clear that the FC M shows an abrupt ferromagnetic transition at $T_C=285$ K that is closely followed by a noticeable change of slope near $T^*=260$ K. Most strikingly, the FC M undergoes a rapid drop below $T_D=50$ K, and anomalously becomes negative at $T<20$ K, unambiguously signaling a magnetization reversal or a rotation of the magnetic moment that is now in opposition to the magnetic field. This behavior is robust and not observed in the ZFC magnetization, which instead remains positive and displays no anomalies that are seen in the FC M (see Fig. 2). In the entire temperature range measured, the ZFC M shows a severely reduced moment and a nearly straight line with an extremely weak temperature dependence that only becomes slightly stronger below 20 K. While the irreversibility is characteristic of a ferromagnet, it is conspicuously unusual that all magnetic anomalies are absent in the case of ZFC measurements, implying a strong spin disordering or a random orientation of magnetic domains that is persistent through T_C. It is found that this drastic irreversibility is strongly correlated with both T_C and T^*. If $Sr_3Ir_2O_7$ is field cooled from the vicinity below T^* rather than room temperature, all anomalies becomes weakened. Shown in the inset of Fig. 2 is M vs T obtained using a FC sequence that starts at $T=300$, 260, and 230 K, respectively (the data were taken from a different crystal that shows a slightly different value of M). It is apparent that both ferromagnetic and diamagnetic responses diminish considerably with decreasing temperature from which the FC sequence starts. Interestingly, if $Sr_3Ir_2O_7$ is field cooled from the temperature range of $T^*<T<T_C$, M shows an essentially unchanged diamagnetic response but a significantly reduced ferromagnetic response. While the nature of T^* is unclear, it cannot be ruled out that a magnetostriction may occur near T^* and "lock" up a certain spin configuration that may facilitate the magnetization reversal. Nevertheless, these observations suggest that any spin disordering below T^* has to be eliminated in order to preserve the diamagnetic state that occurs at lower temperatures.

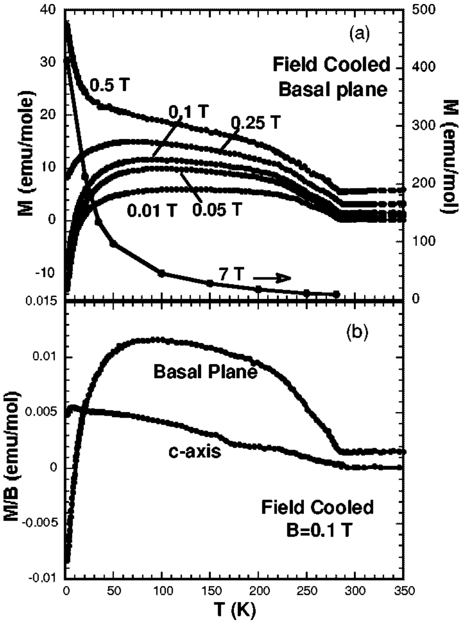

FIG. 3. (a) Magnetization M for the basal plane as a function of temperature for $B=0.01$, 0.05, 0.1, 0.25, 0.5, and 7 T (right scale). (b) Magnetic susceptibility (defined as M/B) as a function of temperature for the basal plane and the *c* axis.

The diamagnetic state is metastable, and exists only at modest magnetic fields. Shown in Fig. 3(a) is the FC M for the basal plane vs T for various magnetic fields. As the magnetic field increases, the ferromagnetic response becomes stronger whereas the diamagnetic state diminishes and eventually vanishes at $B=0.25$ T, indicating that the magnetic moment parallel with B or the ferromagnetic moment is dominant although the downturn of M below T_D still remains. At $B=0.5$ T, the downturn of M develops into a sharp upturn below T_D. This enhancement apparently suggests that all magnetic components that were in opposition to the modest magnetic fields are now aligned with the stronger magnetic field and ferromagnetically added to the overall magnetization. Remarkably, M can flip back to the direction opposite to the magnetic field without hysteresis when $H < 0.25$ T so long as the FC $Sr_3Ir_2O_7$ stays below T^*. This full reversibility of the spin reorientation once again implies that the diamagnetic state or the irreversibility seen in the FC M and the ZFC M shown in Fig. 2 is more likely to be associated with an effect from the crystal structure or a magnetostrictive distortion that stabilizes the metastable state. In addition, T_C remains sharp without shifting and T^* stays well defined, unlike ferromagnetic transitions which will be broadened or antiferromagnetic transitions which will be suppressed to lower temperatures when a higher magnetic field is applied.

As can be noted in Fig. 3(a), at $B > 0.5$ T, there is a strong upturn at low temperatures, which becomes further stronger with increasing B. At $B = 7$ T, M behaves more like a paramagnet. It is plausible although not without speculations that the system is weak ferromagnetic above 50 K at weak magnetic fields and then undergoes a change possibly due to a magnetoelastic effect, becoming nonmagnetic below 50 K, accordingly, the diamagnetic response at low fields and the paramagnetic behavior at higher fields. This paramagnetic phase grows and eventually dominates the entire temperature range as the field increases. Applying Curie-Weiss law to magnetic susceptibility at $B = 7$ T yields an effective moment μ_{eff} of $0.69\mu_B$ (vs $1.73\mu_B$ expected for $S = \frac{1}{2}$) and a Weiss temperature Θ, of -17 K, suggesting an antiferromagnetic coupling.

$Sr_3Ir_2O_7$ has a strong magnetocrystalline anisotropy energy. Figure 3(b) demonstrates a FC magnetic susceptibility defined as M/B vs T for the basal plane and the c axis at $B = 0.1$ T. The magnetic susceptibility is much weaker for the c axis than for the basal plane. Furthermore, the magnetic transition at T_C for the c axis, though still observable, is largely broadened. In addition, there exists no magnetization reversal but a well-defined drop in M/B near 10 K following a slight slope change of M/B between 150 and 250 K. The easy axis for magnetization apparently lies within the basal plane. The anisotropy between the a and b axis is quite small and yields no significant difference in the magnetic behavior.

Figure 4(a) demonstrates the isothermal magnetization M as a function of magnetic field B parallel to the basal plane at $T = 2$, 20, 35, and 50 K (left scale) and various higher temperatures measured in a FC sequence. This set of data shows a few remarkable features: (i) the FC $M < 0$ at modest fields for the basal plane but $dM/dB > 0$, confirming that the diamagnetic state is originated from nothing else but spin canting. The ZFC M shows no magnetization reversal [see Fig. 4(b)]; (ii) M shows a significant slope change as T rises. It appears to be more "saturated" and yet with an extremely low moment when $T > 150$ K (right scale), i.e., less than $0.003\mu_B$/Ir. A linear field dependence at higher fields for $2 < T < 50$ K may indicate an antiferromagnetic state intermediate between the high-temperature weak ferromagnetic state and the low-temperature diamagnetic state. Relevantly, M becomes slightly curved at low fields when $T > 35$ K, suggesting a development of weak ferromagnetic component with increasing T; (iii) $Sr_3Ir_2O_7$ shows no hysteresis in either the FC M or the ZFC M cycled to 7 T, i.e., the spin rotation is fully reversible, indicating a possible association with the magnetostriction that, if taking place at $T^* < T < T_C$, already stabilizes the spin structure that favors the spin reversal at lower temperatures; (iv) the magnetic moment for the basal plane at 7 T is about $0.037\mu_B$/Ir, which means a canting angle of ~2.5° for $1\mu_B$/Ir expected for $S = \frac{1}{2}$.

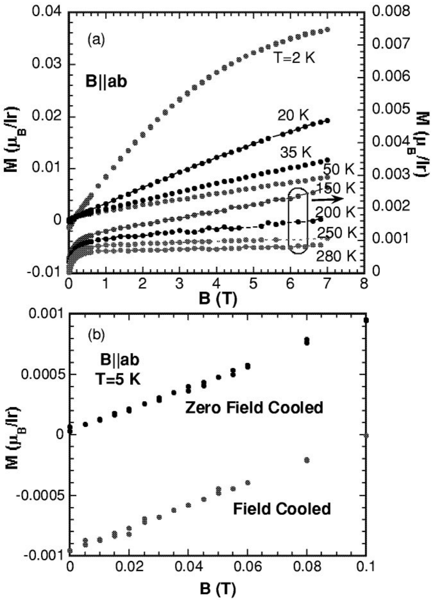

FIG. 4. (a) Field-cooled isothermal magnetization M for the basal plane at 2, 20, 35, and 50 K (left scale) and higher temperatures (right scale). (b) Enlarged low field M at 5 K for the FC and ZFC sequences.

Shown in Fig. 5 is the electrical resistivity $\rho(T)$ as a function of T for the basal plane and the c axis $(1.5 < T < 1000\ \mathrm{K})$. ρ for both the basal plane and the c axis increases slowly as T decreases from 1000 to 300 K and then rises rapidly in the vicinity of $T_C (= 285\ \mathrm{K})$ and $T_D (= 50\ \mathrm{K})$, characteristic of a coupling of magnetic and transport properties. ρ (T) can be fit to $1/T$ dependence for the high-temperature range $300 < T < 1000$ K (not activation behavior). In ρ_{ab} fits well to $1/T^{1/4}$ for $15 < T < 100$ K, suggesting variable range hopping (VRH) behavior with the negligible Coulomb interaction. $\rho_c(T)$, however, does not obey

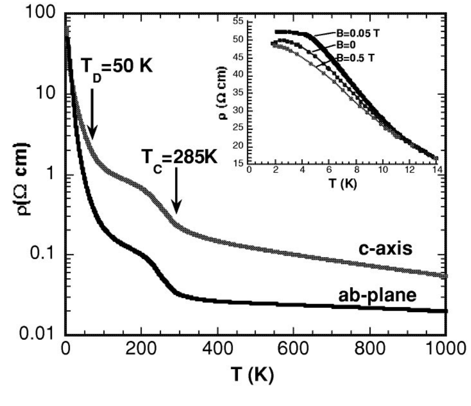

FIG. 5. Resistivity ρ as a function of temperature for the basal plane and the c axis. Inset: ρ vs T at $B = 0$, 0.05, and 0.5 T.

any known power laws or exponentials over the same temperature interval. The anisotropy between the c axis and the basal plane, or ρ_c/ρ_{ab} reaches as high as 7 but diminishes at low temperatures. It is worth noting that the system remains to be semiconducting or insulating without a metal-insulator transition even up to 1000 K. A similar localized behavior has been also observed in Sr_2IrO_4.

Although the magnetization and the resistivity seem to suggest a simultaneous transition at $T_C = 285$ K, $\rho(T)$ for $T > 15$ K is surprisingly insensitive to the magnetic field up to 10 T, i.e., neither magnetoresistivity nor a shift in the transition temperature is observed. The similar behavior has been observed in other iridates such as Sr_2IrO_4 (Ref. 13) and $BaIrO_3$ (Ref. 14). It is interesting, however, that $\rho(T)$ below 15 K appears to show some sensitivity to the magnetic field when $Sr_3Ir_2O_7$ is field cooled through T_C (see the inset of Fig. 5). More notably, ρ shows changes that seemingly reflect the magnetic states: Positive magnetoresitivity $[\Delta\rho/\rho(0T) = 8\%]$ for $B = 0.05$ T at which the net magnetic moment is opposite to B (the diamagnetic state), and negative magnetoresistivity $[\Delta\rho/\rho(0T) = 6\%]$ for $B = 0.5$ T at which the weak ferromagnetic state is restored [see Fig. 3(a)].

IV. DISCUSSION

The occurrence of the magnetization reversal is not without precedent. It has been observed in the ferrimagnetic spinels Co_2VO_4 (Ref. 20) and Co_2TiO_4 (Ref. 21) characterized by two inequivalent magnetic sublattices that are antiferromagnetically coupled. The reversal of magnetization in these compounds is believed to be due to different temperature dependence of the individual magnetic sublattices.[20] A similar phenomenon though with distinct features has been recently observed in $LaVO_3$ (Refs. 22–24) and YVO_3 (Ref. 25), which are canted antiferromagnetic insulators with only one type of magnetic ions (V^{3+}). Although $LaVO_3$ and YVO_3 are isostructural, the diamagnetic state of these vanadates differs significantly. For $LaVO_3$, it is suggested that the phenomenon is essentially a result of the first-order Jahn-Teller phase transition occurring at $T = 138$ K, a few degrees below the Néel temperature ($T_N = 142$ K). This structural transition maximizes the orbital angular momentum and therefore enhances the spin-orbit coupling that consequently reverses the Dzyaloshinski vector, thus a resultant magnetic moment opposite to the small magnetic field.[24] For YVO_3, which displays multiple magnetization reversals at $T = 77$ and 95 K with $T_N = 116$ K, the anomalous behavior is attributed to a change of orbital ordering and a delicate combined effect driven by the single-ion anisotropy and antisymmetric Dzyaloshinski-Moriya (DM) interactions defined as $\mathbf{D}\cdot(\mathbf{S}_1 \times \mathbf{S}_2)$ (where $\mathbf{D}$ is the Dzyaloshinski vector, and $\mathbf{S}_1$ and $\mathbf{S}_2$ are neighboring spins).[25]

Unlike the ferrimagnetic spinels or the vanadates, $Sr_3Ir_2O_7$ has neither two inequivalent magnetic sublattices whose different temperature dependences lead to the compensation effect in the spinels, nor the orbital angular momentum and spin-orbit coupling (due to quenching by crystalline fields), which is thought to be crucial to the magnetization reversal in the vandates. $Sr_3Ir_2O_7$ has also a much higher Curie temperature and a far broader temperature range between T_C and T_D, the onset of the reversed magnetization.

Ferromagnetism with a small ordered magnetic moment has also been found for Sr_2IrO_4 below 250 K.[8] This system is the $n = 1$ compound of the Ruddlesden-Popper series. Structurally it corresponds to the tetragonal space group $I4_1/acd$, i.e., four layers of IrO_6 octahedra, which are slightly stretched along the c axis and rotated by 11° about the c axis in a staggered fashion. The deformation and rotation of the octahedra is then quite similar to $Sr_3Ir_2O_7$. This is also the case for the weak ferromagnetism, except at low T. For Sr_2IrO_4 the magnetic order has been attributed to the Dzyaloshinski-Moriya interaction with $\mathbf{D}$ originating from the rotation of the octahedra. Sr_2IrO_4 does not reverse the magnetization at low T, although there appears to be a small decrease at lowest temperature measured (see top panel of Fig. 3 in Ref. 8).

The Ir^{4+} ($5d^5$) ions in $Sr_3Ir_2O_7$ are presumed to be in a low spin state. The large extension of the $5d$ orbitals leads to large crystal-field splittings and a reduced Coulomb repulsion. In perfectly symmetric octahedra, the states are split into a lower t_{2g} triplet and an excited e_g doublet. Only the t_{2g} orbitals play a role here, because in contrast to the manganites the exchange between the $5d$ electrons is too small to align the spins into a $S = \frac{5}{2}$ (first Hund's rule). Since the octahedra are elongated along the c axis, the degeneracy of the t_{2g} states is lifted, such that the d_{xy} orbital has lower energy than the d_{xz} and d_{yz} orbitals, which are nearly degenerate (they would be degenerate in tetragonal symmetry). With five electrons to be accommodated, the d_{xy} orbital is filled with two electrons (up spin and down spin) and the d_{xz} and d_{yz} orbitals are occupied by three electrons, i.e., the d_{xz} and d_{yz} orbitals accommodate one hole [see Fig. 6(a)].

The twofold orbital degeneracy of the d_{xz} and d_{yz} is now lifted by the rotation of the octahedra about the c axis by 11°. This can be accomplished via a lattice distortion of the Jahn-Teller type by coupling to a phonon at the Brillouin-zone boundary. The consequence of the rotations is a halving of the Brillouin zone in the ab plane. The lifting of the degeneracy reduces the energy by opening gaps at the boundaries of the reduced Brillouin zone. This does not affect the d_{xy} bands (which remain filled), but it opens the possibility of various scenarios for the occupation of the now four bands (in the reduced Brillouin zone) originating from the d_{xz} and d_{yz} states. Note that each band can accommodate $\frac{1}{2}$ electron per spin direction. The bands are expected to be narrow, because shielding of the Ir ions by the octahedra reduces the overlap of $5d$ orbitals on neighboring sites. The gaps are expected to be small since they arise from a lattice distortion (Jahn-Teller type). There are two possible scenarios leading to an insulating system.

Scenario (i): Two of the four bands in the reduced Brillouin zone are filled (for up and down spins) and the other two are filled for up-spin electrons but empty for down-spin electrons. This situation corresponds to a magnetic insulator and its realization would require that the exchange interaction is sufficiently strong (compared to the band width and the gaps) to align the spins, so that an effective spin $\frac{1}{2}$ can be

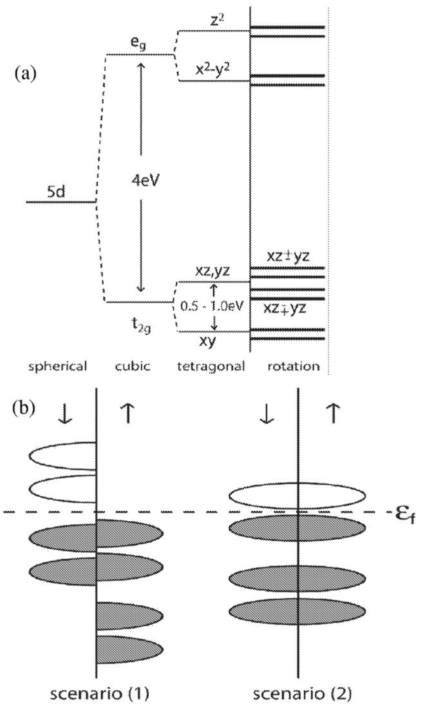

FIG. 6. (a) Effects of crystal structural distortions on t_{2g} and e_g levels for Ir^{4+} $(5d^5)$; (b) two possible scenarios leading to an insulating system (see text).

formed at each Ir site. This case is physically analogous to the picture of local magnetism. The spins would have to order antiferromagnetically (there are several possible arrangements) aligned in plane without a net magnetic moment. The small ordered ferromagnetic moment could originate from a small canting of the spins, induced, e.g., by the Dzyaloshinski-Moriya interaction [see Fig. 6(b)].

Scenario (ii): Three bands are filled and one is empty for both spin components, which corresponds to a nonmagnetic insulator. However, the Zeeman splitting could induce changes in the overlap matrix elements of $5d$ orbitals on neighboring sites, which are different for the up-spin and down-spin subbands. This could give rise to a magnetic response of the system, similar to that of a small magnetic moment. The effect could become collective if the induced magnetic moment slightly changes the rotation angle of the octahedra [see Fig. 6(b)].

It is likely that the rotation of the octahedra is not completely uniform throughout the crystal, but that there is a distribution of angles. This distribution gives rise to scattering and to localization of some electron states. The localization of states close to the band edges may be the origin of the variable range hopping observed in the temperature interval $15 < T < 100$ K.

Scenario (i) is very similar to current interpretations for the high-T_c compound $\mathrm{La_2CuO_4}$ and the one for the sister compound $\mathrm{Sr_2IrO_4}$ presented in Ref. 8. Unfortunately, there is no theory available for the magnetization reversal. The mechanisms proposed for the ferrimagnetic spinels and the vanadates do not seem to apply to the present case. However, the arguments invoked in scenario (ii), i.e., a change in the hopping matrix elements different for the two spin components via a Zeeman splitting could lead to an increase in the energy and hence to a diamagnetic response.

V. CONCLUDING REMARKS

While $\mathrm{Sr_3Ir_2O_7}$ is indeed a rare magnetic system, it shares a number of unique features with its sister compound $\mathrm{Sr_2IrO_4}$ and the nine-layer hexagonal $\mathrm{BaIrO_3}$. They are all characteristically insulating with unusual temperature dependence of resistivity and negative differential resistivity or non-ohmic behavior[13,14] in spite of the largely extended $5d$-electron orbitals, which are conventionally thought to favor a metallic ground state. It is striking that all their counterparts in the ruthenates ($\mathrm{Sr_2RuO_4}$, $\mathrm{Sr_3Ru_2O_7}$, and $\mathrm{BaRuO_3}$) are metallic or even superconducting! These iridates are all weakly ferromagnetic with relatively high magnetic ordering temperatures ($T_C = 240$ and 175 K for $\mathrm{Sr_2IrO_4}$ and $\mathrm{BaIrO_3}$, respectively) and yet only a fraction of the expected ordered moment ($\mu_o = 0.14\ \mu_B$/Ir and $0.03 \mu_B$/Ir for $\mathrm{Sr_2IrO_4}$ and $\mathrm{BaIrO_3}$, respectively).[13,14] Should the low moment be due to spin canting alone, then it would be extraordinarily unusual that the magnetic field as high as 55 T cannot even achieve 30% of the anticipated moment and induce any spin flop or metamagnetic transitions.[14] In addition, all these materials often show a simultaneous transition in magnetization and resistivity, but none displays sensitivity of T_C to high magnetic fields and detectable magnetoresistance near and below T_C except at very low temperatures.[13,14] Without doubt, this class of the materials, which are often characterized by overwhelming interatomic interactions, presents new intellectual challenges and merits systematic investigations.

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