

Robustness of Basal-Plane Antiferromagnetic Order and the $J_{\text{eff}} = 1/2$ State in Single-Layer Iridate Spin-Orbit Mott Insulators

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The magnetic structure and electronic ground state of the layered perovskite Ba_2IrO_4 have been investigated using x-ray resonant magnetic scattering. Our results are compared with those for Sr_2IrO_4 , for which we provide supplementary data on its magnetic structure. We find that the dominant, long-range antiferromagnetic order is remarkably similar in the two compounds and that the electronic ground state in Ba_2IrO_4 , deduced from an investigation of the x-ray resonant magnetic scattering L_3/L_2 intensity ratio, is consistent with a $J_{\text{eff}} = 1/2$ description. The robustness of these two key electronic properties to the considerable structural differences between the Ba and Sr analogues is discussed in terms of the enhanced role of the spin-orbit interaction in $5d$ transition metal oxides.

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Transition metal oxides containing a $5d$ element are increasingly attracting attention as an arena in which to search for novel electronic states [1–4]. These are proposed to derive from the strong spin-orbit interaction (SOI) in the $5d$'s, which in essence entangles spin and orbital moments, strongly mixing spin and spatial coordinates. Iridium-based compounds have featured predominantly in this quest, with considerable focus on the layered perovskites of which Sr_2IrO_4 is the prototypical example [5]. In this case, the SOI leads to a $J_{\text{eff}} = 1/2$ ground state for the Ir^{4+} ($5d^5$) ions, from which a Mott-like insulator then emerges through the action of relatively weak electronic correlations which would otherwise lead to a metallic state. Direct evidence for the existence of a $J_{\text{eff}} = 1/2$ ground state in Sr_2IrO_4 was provided by x-ray resonant magnetic scattering (XRMS) experiments, which revealed a much stronger resonance at the L_3 edge than at the L_2 [6].

The structural similarity of the single-layer iridates to La_2CuO_4 adds further impetus to the study of these materials, opening as it does a possible route to the discovery of new families of superconductors [7]. In this context, a particularly interesting compound is Ba_2IrO_4 , since structurally it is a closer $5d$ analogue of La_2CuO_4 than the Sr compound. Ba_2IrO_4 crystallizes in the K_2NiF_4 -type structure (space group $I4/mmm$) with 180° Ir-O-Ir bonds in the

basal plane (Fig. 1) and with a 7% tetragonal distortion of the IrO_6 octahedra along the $[0\ 0\ 1]$ direction [8]. In contrast, in Sr_2IrO_4 , there is a tetragonal distortion of 4% [9] and a staggered, correlated rotation of the IrO_6 octahedra by 11° about the c axis that reduces the space group symmetry to $I4_1/acd$. This generates a larger unit cell, $\sqrt{2}a \times \sqrt{2}b \times 2c$, under the rotation of the original cell by 45° .

From a theoretical point of view, both the tetragonal distortion and the presence or otherwise of octahedral rotations have significant consequences for the electronic and magnetic properties. First, it should be noted that the $J_{\text{eff}} = 1/2$ state itself is only strictly realized in a system of cubic symmetry [10]. Second, the loss of inversion symmetry in Sr_2IrO_4 gives rise to a finite Dzyaloshinskii-Moriya (DM) interaction, allowing the formation of non-collinear magnetic structures [11]. Both of these effects on the magnetism in Ba_2IrO_4 and Sr_2IrO_4 have been investigated using *ab initio* methods [12].

For Sr_2IrO_4 , the consequences of these structural features for the electronic and magnetic properties have been comprehensively explored in a number of experimental and theoretical studies [5,6,11,13]. By contrast, for Ba_2IrO_4 there are a number of important open questions, including whether or not its ground state can reasonably be assigned as $J_{\text{eff}} = 1/2$ and the exact nature of its magnetic

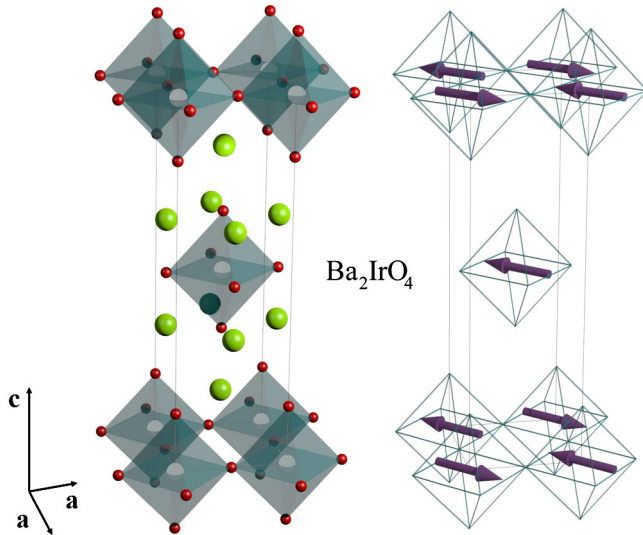


FIG. 1 (color online). The left-hand panel shows the crystal structure of Ba_2IrO_4 . Perovskite IrO_6 layers, where the Ir atoms (grey) lie at the center of corner sharing oxygen (red) octahedra, are separated by Ba atoms (light green). The right-hand panel shows the basal-plane antiferromagnetic structure of the Ba_2IrO_4 , where the magnetic moments are pointing along the $[1\ 1\ 0]$ direction.

structure. The latter question is of particular relevance to the prospect of Ba_2IrO_4 becoming the parent compound of a new family of unconventional, magnetically mediated superconductors. Both cuprate and pnictide superconductors, for example, emerge when doping destabilizes long-range antiferromagnetic order, and in each case obtaining a microscopic understanding of the magnetic groundstate of the parent compound has played a pivotal role in our knowledge [14,15]. From a range of bulk probes and muon spin rotation (μSR) it is known that Ba_2IrO_4 exhibits a magnetic transition below $\sim 240\text{ K}$ [8], close to the magnetic transition in Sr_2IrO_4 of $T_N \sim 230\text{ K}$, below which the magnetic moments in Sr_2IrO_4 form a canted antiferromagnetic structure [6]. Whether or not the ferromagnetic moment resulting from the canting is inimical for superconductivity when Sr_2IrO_4 is doped to form a metal is another important open question.

In this Letter we report the results of our XRMS investigation of Ba_2IrO_4 , which addresses both the question of the magnetic structure in Ba_2IrO_4 and the relevance of the $J_{\text{eff}} = 1/2$ description to its electronic groundstate. Our results are compared with corresponding measurements on Sr_2IrO_4 , for which we also supply supplementary data, and discussed in terms of current theoretical models. The major achievement of our study is to establish that both antiferromagnetic order and the $J_{\text{eff}} = 1/2$ state are, to a remarkable degree, robust to structural distortions in the single layered iridate perovskites.

The XRMS measurements were performed at the Ir L_2 (12.831 keV) and L_3 (11.217 keV) edges, probing dipolar

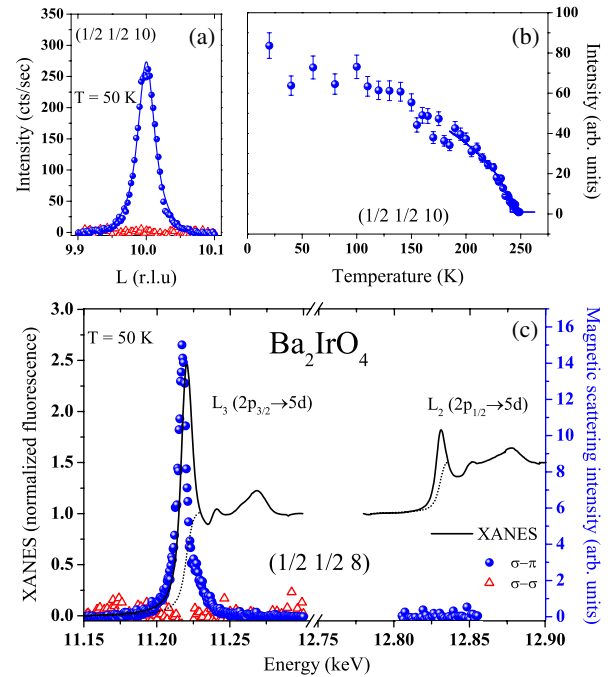


FIG. 2 (color online). (a) L scans across the $(\frac{1}{2}\ \frac{1}{2}\ 10)$ magnetic reflection, $T = 50\text{ K}$ in Ba_2IrO_4 . (b) The temperature dependence of the $(\frac{1}{2}\ \frac{1}{2}\ 10)$ magnetic reflection in Ba_2IrO_4 . The solid blue line is a fit to a power law. (c) Resonant enhancement of the $(\frac{1}{2}\ \frac{1}{2}\ 8)$ magnetic reflection across the $L_{2,3}$ edges at $T = 50\text{ K}$ in Ba_2IrO_4 . The solid black line shows the XANES spectra normalized to the number of initial states. The blue spheres and red triangles show the intensity of the $(\frac{1}{2}\ \frac{1}{2}\ 8)$ reflection. The black dashed line demarcates the integrated white line used to calculate the branching ratio.

transitions from $2p_{1/2}$ to $5d$ and from $2p_{3/2}$ to $5d$, respectively. The experiment on the Ba_2IrO_4 crystal was conducted at the I16 beam line of the Diamond Light Source, Didcot, UK. The XRMS study on Sr_2IrO_4 was performed at the P09 beam line [16] of Petra III, at DESY, Germany. An Au (3 3 3) crystal and a pyrolytic graphite (0 0 8) crystal were exploited to analyze the polarization of the scattered beam on I16 and on P09, respectively.

In Ba_2IrO_4 , with the photon energy tuned to be close to the L_3 edge (11.222 keV) and the sample cooled to 50 K, sharp peaks were found at the reciprocal lattice points $(\frac{1}{2}\ \frac{1}{2}\ L)$ with L even. These peaks existed in the rotated photon polarization channel $\sigma - \pi$ only (see Fig. 2(a)) as expected from the selection rules for XRMS arising from dipolar transitions [17]. We thus deduce that the Ir^{4+} magnetic moments order in an antiferromagnetic structure, with a doubling of the unit cell along the in-plane directions ($\sqrt{2}a \times \sqrt{2}b$), described by a magnetic propagation vector of $\mathbf{k} = [\frac{1}{2}\ \frac{1}{2}\ 0]$.

In Fig. 2(c) we present the energy dependence of the magnetic scattering at $(\frac{1}{2}\ \frac{1}{2}\ 8)$ together with x-ray absorption near edge structure (XANES) measurements for energies in the vicinity of the L_3 and L_2 edges. The most

notable features of this data are the existence of a well-defined resonance at the L_3 edge, and the complete absence of a response at the L_2 edge within experimental uncertainty. Concerted attempts to find a magnetic response at the L_2 edge by investigating various magnetic reflections all ended in failure. In their study of Sr_2IrO_4 , Kim *et al.* [6] argued that the observed large XRMS intensity ratio, I_{L_3}/I_{L_2} , served as a unique fingerprint of the $J_{\text{eff}} = 1/2$ state, since for the pure $J_{\text{eff}} = 1/2$ state I_{L_2} is identically zero. Our results, interpreted in this spirit, establish that even in the presence of a large tetragonal distortion, Ba_2IrO_4 belongs to the same class of $J_{\text{eff}} = 1/2$ spin-orbit Mott insulators as Sr_2IrO_4 .

The width of the L_3 resonance is $\text{FWHM } L_3 = 7.6(1)$ eV, comparable to the values found in Sr_2IrO_4 and in $\text{Sr}_3\text{Ir}_2\text{O}_7$ [6,18]. The position of the resonance, similarly to those of Sr_2IrO_4 and $\text{Sr}_3\text{Ir}_2\text{O}_7$, is 3 eV below the L_3 white line. From the analysis of the XANES spectra, we find a very large branching ratio $\text{BR} = 5.45$ [19]. This is a further confirmation of the strong SOI regime in Ba_2IrO_4 .

The thermal evolution of the antiferromagnetic order was determined by performing $\theta - 2\theta$ scans of the $(\frac{1}{2} \frac{1}{2} 10)$ reflection in the $\sigma - \pi$ channel at the energy (11.219 keV) that maximizes the XRMS response. Figure 2(b) shows the integrated intensity obtained by fitting a Lorentzian peak shape to the individual scans as a function of temperature. The transition appears to be second order, and from the fit to a $A(1 - \frac{T}{T_N})^{2\beta}$ function we obtain the Neel temperature $T_N = 241(2)$ K and $\beta = 0.25(4)$, in good agreement with the values found by μSR measurements [8].

In order to determine the magnetic structure of Ba_2IrO_4 , we performed azimuthal scans of the $(\frac{1}{2} \frac{1}{2} 10)$ magnetic reflection. This method consists in measuring $\theta - 2\theta$ scans for different Ψ angles, rotating the sample around the scattering vector. From the azimuthal modulation of the intensity of the XRMS signal it is possible to determine the orientation of the magnetic moments in an antiferromagnetic material [20]. Figure 3 shows the azimuthal dependence of the $(\frac{1}{2} \frac{1}{2} 10)$ reflection (blue solid points). The dashed black line, solid blue line and dotted red line are the azimuthal dependencies calculated by means of the FDMNES package [21] for three different orientations of the moments, $m \parallel [001]$ (Γ_3), $m \parallel [110]$ (Γ_5), and $m \parallel [1-10]$ (Γ_7), that are the allowed magnetic structures and irreducible representations (Γ_i) according to group theory calculations [22]. The experimental curve most closely resembles the calculation for the Γ_5 representation. We therefore conclude unambiguously that Ba_2IrO_4 exhibits a basal-plane antiferromagnetic order with the magnetic moments pointing along the $[1 1 0]$ direction. The magnetic structure of Ba_2IrO_4 is shown in Fig. 1.

To understand the dependence of the $J_{\text{eff}} = 1/2$ state and the associated Hamiltonian on symmetry and lattice distortions, we have investigated the magnetic structure of

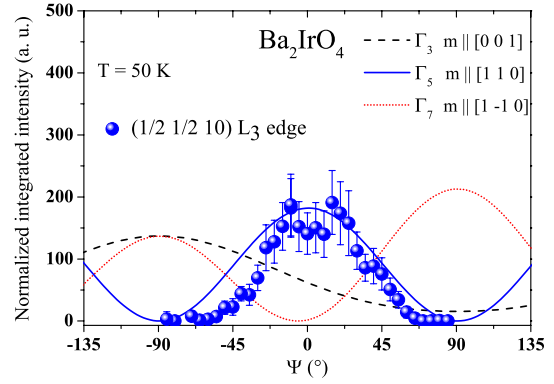


FIG. 3 (color online). The azimuthal dependence of the $(\frac{1}{2} \frac{1}{2} 10)$ magnetic reflection (solid blue spheres) in Ba_2IrO_4 . The lines are the azimuthal dependencies calculated for the allowed moment orientations in Ba_2IrO_4 . The azimuthal angle Ψ is defined with respect to the reference vector $[1 1 0]$ in the $I4/mmm$ space group.

Sr_2IrO_4 . In particular we focus on the polarization and azimuthal dependencies of the XRMS, neither of which have been reported [6]. With the photon energy tuned to the Ir L_3 edge, well defined magnetic peaks were found at the $(1 0 4n)$ and $(0 1 4n + 2)$ Bragg positions, which are forbidden within the $I4_1/acd$ space group and correspond to the $(\frac{1}{2} \frac{1}{2} L)$ peaks observed in the Ba_2IrO_4 [as illustrated in the inset of Fig. 4(c)]. Figures 4(a) and 4(b) shows the L scan and the energy scan of the $(1 0 24)$ magnetic reflection at the Ir L_3 edge at $T = 90$ K. The well-defined L scan supports the existence of a long-range antiferromagnetic order. The Lorentzian shape of the energy scan ($\text{FWHM}_{L_3} = 6.26(9)$ eV) and the absence of any $\sigma - \sigma$ scattering mechanism confirms the magnetic nature of the peaks, similarly to Ba_2IrO_4 . These results are in agreement with the first XRMS study of Sr_2IrO_4 [6].

In order to determine the direction of the magnetic moments in Sr_2IrO_4 we performed azimuthal scans at the Ir L_3 edge, $T = 90$ K. The results, together with the FDMNES calculation using the same moment direction as in the irreducible representation Γ_5 of Ba_2IrO_4 , are presented in Fig. 4(c). Note the equivalence of the Ψ angles in Fig. 4 with those in Fig. 3, for the correspondence of the $[1 1 0]$ direction in $I4/mmm$ to the $[1 0 0]$ direction in the $I4_1/acd$. By comparing the azimuthal dependence of the $(1 0 24)$ reflection in Sr_2IrO_4 with the azimuthal dependence of $(\frac{1}{2} \frac{1}{2} 10)$ reflection in Ba_2IrO_4 , we deduce that in Sr_2IrO_4 the antiferromagnetic component is oriented along the $[1 1 0]$ direction of the $I4/mmm$ reference system. We therefore conclude that the two compounds have essentially the same basal-plane antiferromagnetic structure [23].

We now turn to the discussion of our results. An effective, low-energy Hamiltonian for the layered iridates, valid in the strong SOI limit, incorporating both the effects of a tetragonal crystal field and rotation of the IrO_6 octahedra

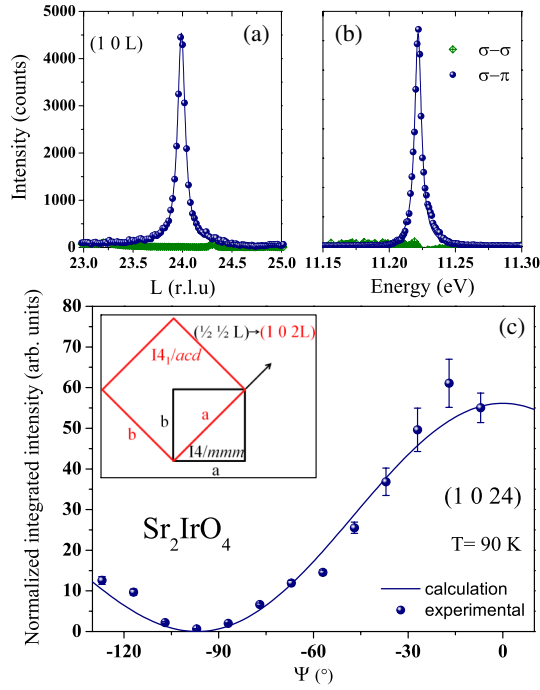


FIG. 4 (color online). Reciprocal space L scan (a) and energy dependence (b) of the XRMS intensity of the $(1\ 0\ 24)$ reflection in Sr_2IrO_4 . The solid blue line is a fit to a Lorentzian peak shape. The azimuthal dependence of the same reflection (c) is compared with a calculation for collinear moments along $[1\ 0\ 0]$. The azimuthal angle Ψ is defined with respect to the reference vector $[1\ 0\ 0]$ in $I4_1/acd$, which corresponds to the $[1\ 1\ 0]$ in the $I4/mmm$ space group, as demonstrated in the inset by means of a two-dimensional projection onto the basal plane of the unit cell.

(by an angle α), has been derived by Jackeli and Khaliullin [11], which we write as

$$\mathcal{H}_{ij} = J\vec{S}_i \cdot \vec{S}_j + J_z S_i^z S_j^z + D \cdot [\vec{S}_i \times \vec{S}_j] + \mathcal{H}^l. \quad (1)$$

The terms on the right-hand side are the isotropic Heisenberg exchange, the symmetric and asymmetric DM anisotropies, and finally an anisotropic contribution from the Hund's coupling [11]. This Hamiltonian has been used to successfully account for the canted magnetic structure observed in Sr_2IrO_4 [6] and additionally for a dimensionally driven spin reorientation in its bilayer counterpart $\text{Sr}_3\text{Ir}_2\text{O}_7$ [18,20,24]. For Ba_2IrO_4 , it would also seem to offer a natural explanation of our results: with $\alpha = 0$, the second and third terms are identically zero, leaving a leading isotropic exchange along with a weaker anisotropy, a Hamiltonian that readily supports the commensurate antiferromagnetic order observed in our experiments. One important proviso, however, is that the magnetic ground state supported by this Hamiltonian becomes unstable above a critical value of tetragonal distortion leading to a spin reorientation where the moments point along the $[0\ 0\ 1]$ direction. Nevertheless, it seems that nearly doubling the tetragonal distortion in moving from Sr to Ba is insufficient to exceed the critical threshold.

Although the above analysis provides a general framework for us to understand the formation of magnetic structures in the layered perovskites, and most especially the canting of the moments in Sr_2IrO_4 , it does not address the key fact revealed in our experiments that the antiferromagnetic components in the two compounds are essentially identical. For this we refer to explicit calculations of J by Katukuri *et al.* [12], who used an *ab initio* many-body approach. Their calculations show that when the SOI is switched off, the ground state and the magnetic interactions are extremely sensitive to the local symmetry and so very different in the two systems: Ba_2IrO_4 has a hole in the d_{xz}/d_{yz} states and a strong antiferromagnetic J interaction (~ 15.4 meV), Sr_2IrO_4 has a hole in the d_{xy} state and a ferromagnetic J interaction (~ -19.2 meV). However, upon including the SOI, the hole acquires an equal d_{xy} , d_{zx} , and d_{yz} character in both compounds, and J in Sr_2IrO_4 becomes antiferromagnetic (~ 51.3 meV) and almost identical to that in Ba_2IrO_4 (~ 58 meV). Therefore, the robustness of antiferromagnetic order in the layered perovskites to structural distortions is ultimately linked to the strong SOI, which produces a ground state wave function that is three dimensional and inherently less perturbed by structural distortions.

In this Letter we have presented a detailed XRMS study of the magnetic and electronic structures of the single layered iridates Ba_2IrO_4 and Sr_2IrO_4 . Ba_2IrO_4 is found to be a basal-plane commensurate antiferromagnet below $T_N = 241$ K. Azimuthal scans combined with group theory calculations have been employed to prove that the moments order along the $[1\ 1\ 0]$ direction. From a comparison with XRMS data on the related compound Sr_2IrO_4 , we establish that both compounds have essentially the same basal-plane antiferromagnetic structure, in spite of their structural differences. We also conclude from our results for the intensity ratio I_{L_3}/I_{L_2} of the XRMS signal that Ba_2IrO_4 is also in the same class of $J_{\text{eff}} = 1/2$ spin-orbit Mott insulators as Sr_2IrO_4 . Thus both the magnetic and electronic structures in the layered perovskites are remarkably robust to structural distortions, a fact that can be linked directly to the unique three-dimensional character of the $J_{\text{eff}} = 1/2$ state produced by the strong SOI, which renders it insensitive to the perturbations in local symmetry.

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