

Strong anisotropy within a Heisenberg model in the $J_{\text{eff}}=1/2$ insulating state of $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$

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The dispersive magnetic excitations in Sr_2IrO_4 have previously been well described within an isospin- $1/2$ Heisenberg model on a square lattice that revealed parallels with La_2CuO_4 . Here we investigate the inelastic spectra of $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ with resonant inelastic x-ray scattering (RIXS) at the Ir L_3 -edge. The results are well described using linear spin-wave theory within a similar Heisenberg model applicable to Sr_2IrO_4 , however the disorder induced by the substitution of 20% Ir^{4+} ions for Ru^{4+} removes longer range exchange interactions. A large spin-gap (40 meV) is measured indicating strong anisotropy from spin-orbit coupling that is manifest due to the altered magnetic structure in $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ with c-axis aligned moments compared to the basal plane moments in the parent. Collectively the results indicate the robustness of a Heisenberg model description even when the magnetic structure is altered and the $J_{\text{eff}}=1/2$ moments diluted.

Understanding the underlying mechanisms that create unconventional superconductivity remains one of central unsolved problems in condensed matter physics and has driven research on cuprates for decades. The search for related materials with similar phenomena in which to either observe superconductivity, or explain the lack thereof, has recently led to the investigation of Sr_2IrO_4 [1–3]. Concurrently iridates, and $5d$ -based transition metal oxides in general, have undergone increased interest unrelated to superconductivity where the delicate balance of competing interactions of strong spinorbit coupling (SOC), increased hybridization and reduced Coulomb interactions lead to novel magnetic phenomena [4].

Sr_2IrO_4 hosts a spin-orbit entangled ground state with $J_{\text{eff}}=1/2$ magnetic moments [1]. Mapping of these pseudo-spins onto several distinct crystal structures has led to the uncovering of a variety of novel properties, such as Kitaev physics and Weyl semimetals [5–9]. Specific interest related to Sr_2IrO_4 has been the predictions of unconventional superconductivity driven by analogous structural and spin properties found in the cuprates. In particular, proximity to a superconducting phase was strongly suggested by the magnetic excitation spectra which revealed striking similarities to La_2CuO_4 and was able to be described within a pure Heisenberg model consisting of spin- $1/2$ on a square lattice [2, 10]. A further surprising aspect of the excitation spectra for Sr_2IrO_4 was the absence of a spin-gap in several measurements [2, 11–13], however a separate reanalysis of the data in Ref. 11 indicated a spin-gap [14]. Overall any spin-gap would be

expected to be large in a SOC dominated system with strong exchange interactions and the absence was explained as being due to an accidental balance of symmetric and antisymmetric anisotropic terms for the basal plane moment case [15]. In these studies of Sr_2IrO_4 XY -anisotropy would be the dominant term in the Hamiltonian for any spin-gap, however we show here that by perturbing the system we not only open a sizable spin-gap but that further terms beyond the proposed XY -anisotropy come to the fore as we alter the moment direction.

Parallels of Sr_2IrO_4 with the cuprates are continuing to mount as was shown with the observation of fermi arcs in surface doped Sr_2IrO_4 [16]. Given the various similarities would suggest bulk superconductivity in the iridates is achievable. Although similarly a consistent lack of superconductivity would also be revealing in finding the key ingredients required for high T_c superconductivity amongst the plethora of candidate phenomena. As with cuprates to achieve bulk superconductivity in iridates a promising route is to dope the parent compound [3]. Moreover in the case of Sr_2IrO_4 doping has proven instructive in understanding the $J_{\text{eff}}=1/2$ moments as the magnetic interactions are altered and diluted. For example studies of the series $\text{Sr}_2\text{Ir}_{1-x}\text{Ru}_x\text{O}_4$ and $\text{Sr}_2\text{Ir}_{1-x}\text{Rh}_x\text{O}_4$ have found a percolative description appropriate as the Ir ions are diluted in explaining the presence of magnetism up to relatively high doping of 20-30% [17, 18]. Moreover, with regards to the explanation of the spin-gap in Sr_2IrO_4 perturbing the system to break the purported accidental

degeneracy that suppresses the spin-gap will allow this reasoning to be directly tested.

Here we investigate spin excitations as the magnetic interactions in Sr_2IrO_4 are tuned by substitution of Ru^{4+} onto the Ir^{4+} site in the form $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$. This region of the phase diagram has well defined $J_{\text{eff}}=1/2$ magnetic moments along the c -axis, as opposed to the basal plane in undoped Sr_2IrO_4 [18], and is an insulator within the relativistic $J_{\text{eff}}=1/2$ Mott-like description of the par-

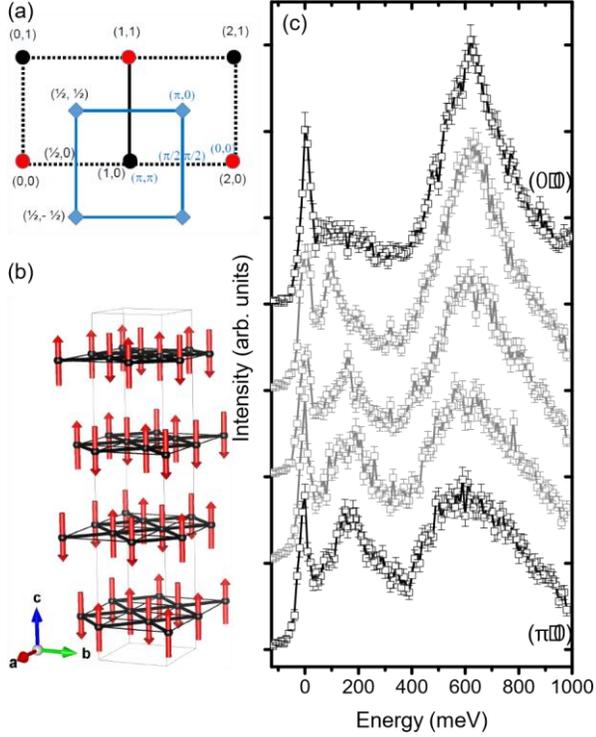


FIG. 1. (a) Structural (red circles) and magnetic (black circles) reciprocal space. High symmetry magnetic zone boundary points are indicated by the blue diamonds. H and K points are labeled along with the labeling in the square lattice notation used to describe the dispersions. The square lattice is rotated from the conventional lattice of $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$. (b) Magnetic ground state of $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ has spins (red arrows) aligned along the c -axis. The nearest and next nearest bonds are shown as thick and thin black lines, respectively. (c) RIXS measurements at the Ir L-edge along several high symmetry directions in the low resolution set-up ($\Delta E=90$ meV).

ent [1, 18]. The results presented here reveal the presence of robust but damped spin-waves in this doped system that are well described within a Heisenberg model with a relative renormalization of the measured dispersion with some qualitatively similar features towards that observed for La_2CuO_4 . Distinct from doping on the Sr site in Sr_2IrO_4 is the observation of a large spin-gap indicating strong anisotropy.

To access the excitation spectra resonant inelastic x-ray scattering (RIXS) measurements at the Ir L_3 -edge (11.215 keV) were performed at the Advanced Photon Source (APS), Argonne National Laboratory. To aid comparison with separate studies on doped and undoped Sr_2IrO_4 and La_2CuO_4 we use the tetragonal $I4/mmm$ space group to describe reciprocal space. A map of the structural and magnetic reciprocal space is shown in Fig. 1(a). The magnetic structure of $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ has been well characterized using both polarized and unpolarized neutron diffraction and resonant x-ray scattering to consist of c -axis aligned spins [18]. The magnetic

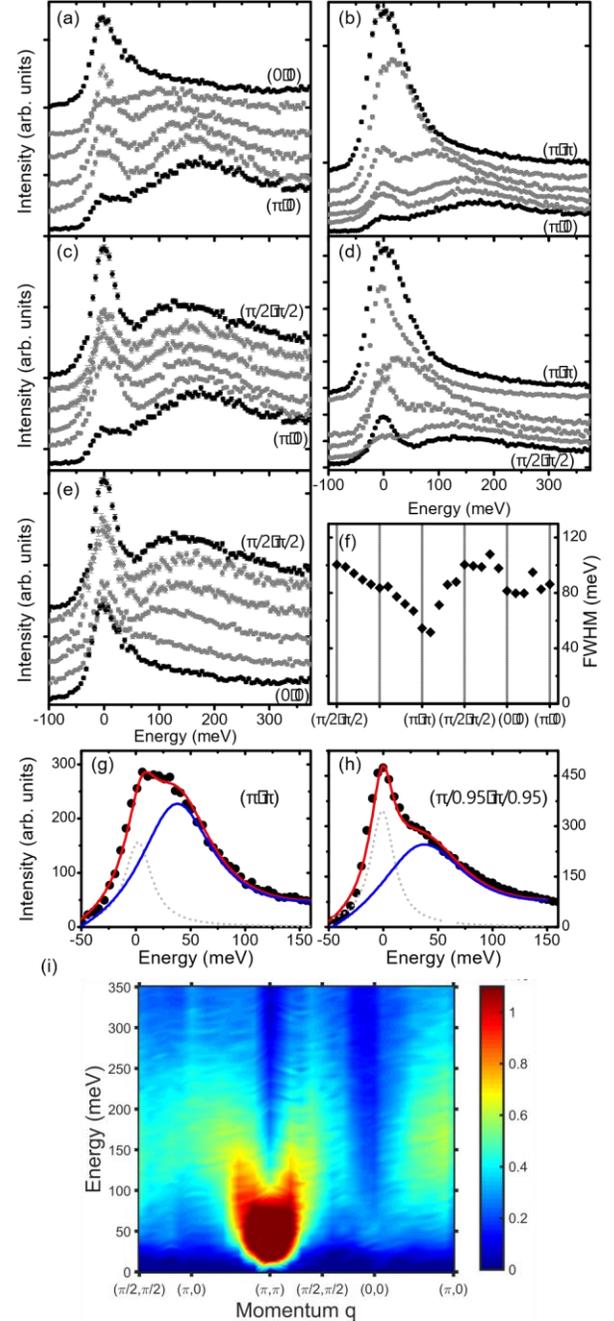


FIG. 2. (a)-(e) High resolution RIXS ($\Delta E=35$ meV) at the Ir L_3 -edge following the low energy magnetic excitation along high symmetry directions. (f) FWHM of the magnetic excitation. (g)-(h) RIXS spectra around (π,π) zone center. The elastic line (gray dashed line) and excitation (blue solid line) are shown. (i) RIXS map with the elastic line subtracted.

structure is shown in Fig. 1(b).

Preliminary RIXS measurements were performed on sector 9-ID, followed by detailed higher energy resolution measurements with $\Delta E=35$ meV FWHM, based on fitting the quasi-elastic line to a charge peak, on MERIX at the APS [19]. The scattering plane and incident photon polarization were both horizontal, i.e. π incident polarization, with the incident beam focused to a size of $41 \times 25 \mu\text{m}^2$ (H \times V) at the sample position on MERIX. To minimize elastic scattering measurements were performed with 2θ close to 90 degrees. This was achieved throughout the Brillouin zone by assuming L independence, in-line with previous RIXS measurements performed [2, 12, 13, 20]. The single crystal of $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ was grown and characterized as described in Ref. 18. The initial 9-ID measurements, with a resolution of $\Delta E=90$ meV FWHM, were used to follow higher energy excitations involving d - d transitions. RIXS measurements to 1 eV energy loss are shown in Fig. 1(c). An inelastic excitation is observed around 650 meV. This excitation has been well characterized in the undoped compound as a spin-orbit exciton arising from transitions between the J_{eff} ground states and the presence here confirms that the $J_{\text{eff}}=1/2$ model remains unbroken in $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ [1]. Additionally the intensity variation, with a maximum at $(0,0)$, corresponds to the behavior of Sr_2IrO_4 . As with Sr_2IrO_4 a lower energy excitation is also observed below 200 meV. This appears to show some dispersion but given the resolution the results are hard to accurately interpret, therefore this scattering was probed with higher resolution RIXS.

To follow the dispersions below 400 meV RIXS measurements with the high resolution set-up ($\Delta E=35$ meV) were performed along high symmetry directions in the magnetic Brillouin zone as shown in Fig. 2(a)-(e). The elastic line and excitation were each fit to a Lorentzian on top of a sloping background to account for the higher energy scattering from the d - d excitations. The broader than resolution FWHM of the excitation is shown in Fig. 2(f) and reveals the excitation is strongly damped, particularly towards the zone boundary, as would be expected for a doped system. The excitation showed dispersive behavior typical of a spin-wave excitation, with maxima at zone boundary and minima at zone center. Fitting the elastic line to a Lorentzian with width restricted to the instrumental resolution and the

excitation at zone center (π,π) revealed a large spin gap of 40 meV, see Fig. 2(g)-(h). This can be seen qualitatively in the RIXS map that shows the scattering with the elastic line removed, Fig. 2(i).

The measured dispersion for $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$, Fig. 3(a), shows deviations from Sr_2IrO_4 in several key regards. (i) the $(\pi,0)$ zone boundary maximum is reduced by 20% to 156 meV indicating a reduction of exchange interactions. (ii) The $(\pi,0) \rightarrow (\pi/2,\pi/2)$ dispersion softens since the $(\pi/2,\pi/2)$ value is close to the parent but the $(\pi,0)$ is appreciably reduced. This has implications on exchange interactions beyond nearest and next nearest. (iii) The $(0,0) \rightarrow (\pi,\pi)$ nodal direction is hardened and the $(0,0) \rightarrow (\pi,0)$ antinodal direction is softened compared to Sr_2IrO_4 , directly opposite to the behavior of electron doping on the Sr

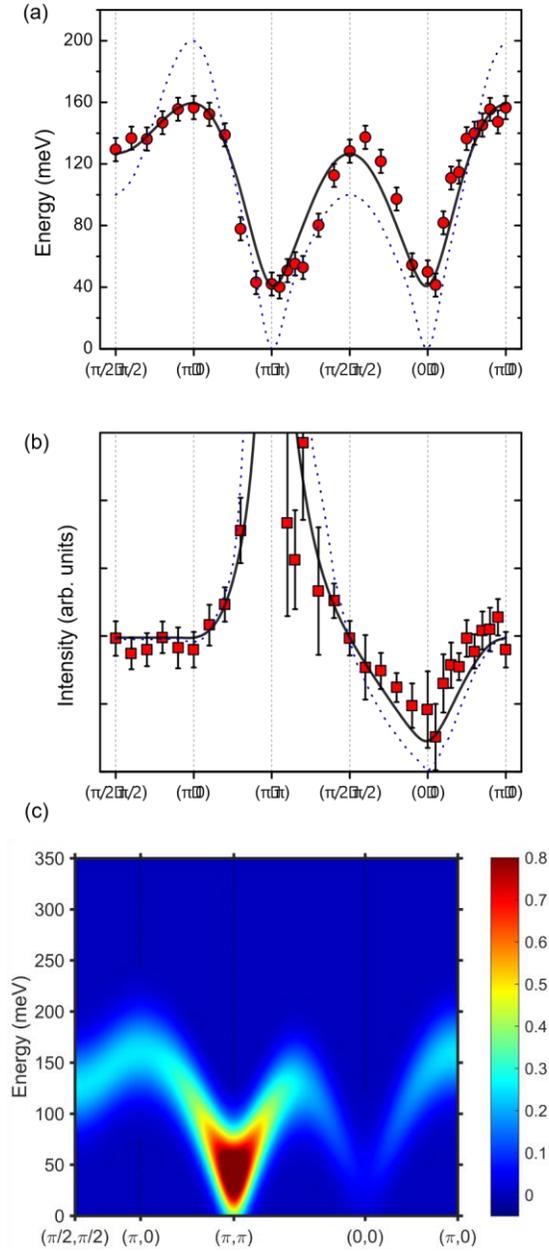


FIG. 3. (a) Measured (circles) and calculated (solid line) dispersion along high symmetry directions and (b) measured (squares) and calculated (solid line) intensity. The dashed blue line is the dispersion for undoped Sr_2IrO_4 based on the Hamiltonian in Ref. 2. (c) Calculated map of the dispersion and intensity.

site [12, 13]. (iv) The excitation is strongly gapped.

The final point is the most striking and a departure from RIXS measurements of doping on the Sr site that shows no spin gap in the RIXS spectra [12, 13] and is consistent with recent measurements on higher Ru doping [20]. As noted there is some debate in the literature for the undoped case with several studies showing no spin-gap, Refs. 2, 11–13,

however the analysis in Ref. 14 indicted a spin-gap. The current debate surrounding a spin-gap is somewhat surprising given the expected presence of anisotropic magnetic interactions in a strongly SOC system with large orbital component. The best RIXS resolution of $\sim 30\text{meV}$, however, precludes the measurement of any small gaps but evidence of such a spin-gap was reported from electron spin resonance of 0.8 meV [15]. This small gap, contrasting with the 90 meV giant magnon-gap of the next member of the RuddlesonPopper series $\text{Sr}_3\text{Ir}_2\text{O}_7$ [21], was explained in Ref. 15 for Sr_2IrO_4 as being due to the quasi-degeneracy of the large antisymmetric DM anisotropy that leads to the observed net FM moment and the large symmetric anisotropies, such as single-ion and exchange anisotropy, resulting in a canceling out of in-plane and out-of-plane anisotropies and a suppressed spin-gap. Therefore following this reasoning in $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ that has c -axis aligned pseudospins this quasi-degeneracy is broken and a large gap can emerge. This observation has further implications since in the undoped case if a large spin-gap does indeed emerge then it is a result of dominant XY -anisotropy [14]. Our results here indicate that there is a further dominant term present in $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ beyond XY anisotropy responsible for the gap. This suggests that in general perturbing Sr_2IrO_4 allows for a control and alteration of the large anisotropic magnetic interactions as the magnetic spin direction is altered from basal plane to c -axis aligned spins. Considering points (i)-(iii) reveals an anisotropic alteration of the dispersion compared to the undoped case and, with the smaller dispersion from $(\pi, 0) \rightarrow (\pi/2, \pi/2)$ behavior that qualitatively appears closer to the cuprate case.

To gain a quantitative understanding of the low energy excitation and underlying Hamiltonian we extracted the energy and intensity of the excitation by fitting to a Lorentzian with a sloping background and modeled the resulting dispersion within linear spin wave theory [22]. As a natural starting point we follow the approach for Sr_2IrO_4 by using an isospin-1/2 Heisenberg model on a square lattice with JJ^0, J^{00} for the first, second and third nearest neighbor exchange interactions in the basal plane. For the undoped case these values correspond to $J = 60\text{ meV}$, $J^0 = -20\text{ meV}$ and $J^{00} = 15\text{ meV}$. The undoped dispersion is shown in Fig. 3(a). The undoped case did not have an observable gap in the RIXS measurements however we account for this spin-gap here by introducing a single-ion term $\Delta = D^P(S_z^2)_i$. We note alternative terms such as Dzyaloshinskii-Moriya (DM) interaction and anisotropic exchange interaction likely contribute to this Δ parameter, however we do not pursue the relative contribution of different terms further.

Allowing the exchange interactions to refine to the measured dispersion consistently resulted in J^{00}

converging towards zero, within error. The dominant effect of J^{00} on the dispersion is to decrease (positive J^{00}) or increase (negative J^{00}) the energy at $(\pi/2, \pi/2)$ relative to $(\pi, 0)$ that, when non-zero, is inconsistent with the extracted dispersion data. Considering the crystal structure of $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ one in five Ir atoms are disrupted by Ru substitution. Since a good fit for the dispersion is found without J^{00} then this indicates that longer range exchange interactions beyond nearest and next nearest neighbors are disrupted. This is distinct from the case of doping on the Sr site of Sr_2IrO_4 where J, J^0, J^{00} are still needed to describe the dispersion [12, 13]. Considering J, J^0 and Δ as a minimal model gives good agreement to the measured dispersion, as shown in Fig. 3(a) with values of $J = 42$ meV, $J^0 = -16$ meV, $J^{00} = 0$ meV and

$D=9$ meV. We based our fitting on the extracted energies rather than the observed intensity due to the incomplete understanding of RIXS scattered cross sections and interferences from elastic line. However, when using the values for exchange interactions to calculate the intensity we find reasonable agreement, as shown in Fig. 3(b). Taking this further we show the purely magnetic scattering cross section from our spin-wave calculations in Fig. 3(c) that show qualitative agreement with the RIXS map we obtained once subtracting the elastic line in Fig. 2(f).

Collectively our results reveal the introduction of strong anisotropy that can be explained by the breaking of the quasi-degeneracy found in Sr_2IrO_4 that artificially suppresses the observation of a spin-gap. Given the strongly SOC dominant behavior previously observed in $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ then the measurement of such a large spin-gap provides further evidence of the importance of SOC in the magnetism, despite the dilution with Ru^{4+} ions. Within this anisotropic picture, coupled with the disordered effect of doping, may suggest a departure from a purely Heisenberg model. However, we were able to apply such a model to describe the excitations in a similarly robust methodology to the parent compound. The dispersion is altered, however, from Sr_2IrO_4 . The principle distinction can be explained as the Ru ions breaking the $J''_{\text{Ir-Ir}}$ third nearest interactions. This behavior is in-line with the percolative picture where the dopant ions disrupt the magnetic ordering rather than interact directly with the Ir ions.

In conclusion, the RIXS spectra for $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ was measured out to 1 eV energy loss. The signature of the $J_{\text{eff}}=1/2$ state was observed as a 650 meV momentum dependent excitation. A lower energy excitation was measured with high resolution ($\Delta E=35$ meV) along high symmetry directions in the magnetic Brillouin zone. The results were well described within a 2D Heisenberg

minimal model of nearest and next nearest interactions. A large spin-gap of 40 meV was observed that is consistent with the magnetic structure in $\text{Sr}_2\text{Ir}_{0.8}\text{Ru}_{0.2}\text{O}_4$ and a large orbital moment on the Ir ion. Modeling of the dispersion for the hole doped case here provides a contrast to the electron doped RIXS studies with alternative behavior that will guide future studies aimed at uncovering bulk superconductivity in doped Sr_2IrO_4 .

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