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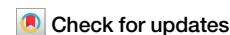
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RIXS observation of bond-directional excitations in the Kitaev material Na_2IrO_3



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Spin-orbit coupling locks spin direction and spatial orientation and generates, in semi-classical magnets, a local spin easy-axis and associated ordering. Quantum spin-1/2's defy this fate: rather than spins becoming locally anisotropic, the spin-spin interactions do. Consequently interactions become dependent on the spatial orientation of bonds between spins, prime theoretical examples of which are Kitaev magnets. Bond-directional interactions imply the existence of bond-directional magnetic modes, spin excitations that render crystallographically equivalent bonds magnetically inequivalent, which yet have remained elusive experimentally. Here we show that resonant inelastic X-ray scattering allows us to explicitly probe the bond-directional character of magnetic excitations. To do so, we use a scattering plane spanned by one bond and the corresponding spin component and scan a range of momentum transfer that encompasses multiple Brillouin zones. Applying this approach to Na_2IrO_3 we establish the different bond-directional characters of magnetic excitations at ~10 meV and ~45 meV. The bond-directional nearest-neighbor excitations are a fingerprint of the dominant Kitaev interactions and even prevail in the magnetically ordered state.

The physics of quantum magnets with bond-directional interactions can be captured by so-called compass models¹, quantum theories of matter in which the couplings between different spin components are inherently spatially (typically, direction) dependent. Theoretically, this class of models harbors a range of interesting emergent physical phenomena, including the frustration of (semi-)classical ordered states on unfrustrated lattices, and in general a boost of quantum effects, prompting, in certain cases, the appearance of quantum spin liquids^{2–4}—Kitaev spin liquids are well-known examples^{5,6}. In spin-1/2 materials, spin-orbit coupling naturally induces bond-directional spin-spin interactions. These can dominate when spin-orbit coupling becomes large, e.g., in *4d* and *5d* transition-metal compounds^{7–11}. However, it has remained a principal challenge to experimentally identify the fingerprints of bond-directional magnetic interactions^{12–16} and to establish methods to systematically explore their consequences for elementary magnetic properties.

Experimentally, spin-spin interactions are typically studied via the energies of magnetic excitations. To address the bond-directional character of the interactions, we propose a complementary approach, focusing on the character of the excitations. Here we show that bond-directional *excitations* (BDE)—spin excitations that render crystallographically equivalent directions magnetically inequivalent—are a hallmark of bond-directional magnetic *interactions* and demonstrate how to use resonant inelastic X-ray scattering (RIXS) to directly probe these BDE. The challenge of resolving the bond-directional character of magnetic modes is that it requires simultaneous knowledge of both the spin operator creating the excitation and the direction of the bond involved. We introduce a RIXS geometry that yields the former via the polarization dependence and the latter by measuring across *multiple* Brillouin zones. Using this method on Na_2IrO_3 , a Kitaev material exhibiting a proximate spin liquid regime^{17–19}, we establish the different bond-directional characters of magnetic spin-conserving and spin-flip excitations at ~10 meV and ~45 meV, respectively.

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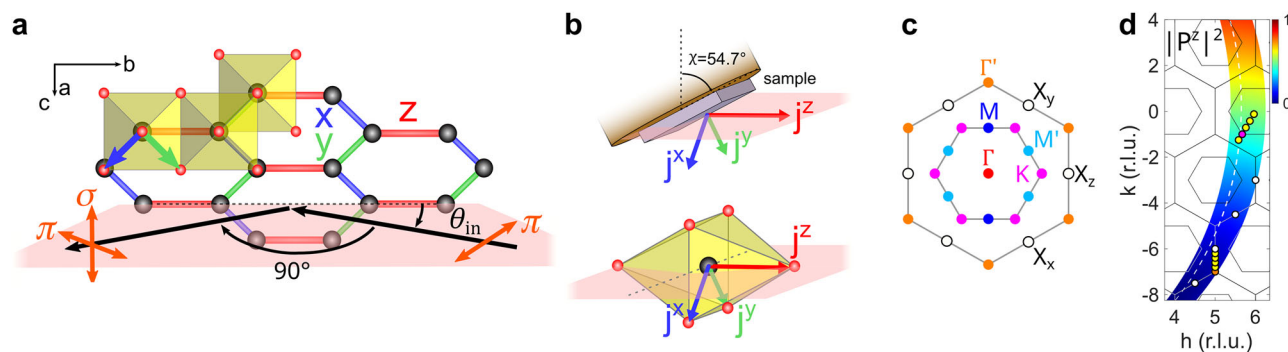


Fig. 1 | Exploiting polarization via the scattering geometry. In our RIXS experiment, both the z bond (red solid line) and the spin component j^z (red arrow, perpendicular to the z bond) are lying in the scattering plane. **a, b** The sample surface is parallel to the honeycomb ab plane with x (blue), y (green), and z bonds. The honeycomb plane is tilted by $\chi = 54.7^\circ$ around the b axis with respect to the scattering plane, yielding the desired orientation of j^z . We use incident π polarization, while the RIXS intensity is summed over outgoing π and σ polarizations. The sketch in (a) further shows the angle of incidence θ_{in} and the scattering angle $2\theta = 90^\circ$. In the coordinate system, the projections of a and c axes fall on top of each other. **b** The two

sketches use the same perspective, showing the tilted sample and the corresponding orientation of the IrO_6 octahedra. Arrows denote spin components j^y . **c** 2D high-symmetry points in the first and second Brillouin zones around Γ . A Γ point occurs for $h + k$ even and $k = 3m$ with integer m and h , see **d**. BDE is most evident around X_y . **d** Polarization factor $|P^z(\mathbf{q})|^2$ across the covered $\mathbf{q}$ range. The dashed line corresponds to scanning θ_{in} from 5° to 65° for $\chi = 54.7^\circ$ and scattering angle $2\theta = 90^\circ$. Experimentally, we vary χ between 52° and 64° . Symbols mark the $\mathbf{q}$ points addressed in Fig. 2. For the high-symmetry points, symbol colors refer to the sketch in **c**.

Resolving BDE with RIXS on Na_2IrO_3 is based on polarization selection rules, but a polarization analysis of the scattered X-rays is not available at the commonly used Ir L edge. We have solved this problem by exploiting a *tilted* sample geometry, which highlights polarization effects over the large range of $\mathbf{q}$ space that can be covered with hard X-rays, see Fig. 1. Tilting the sample puts the spin-orbit entangled j^z component of the local pseudo-spin $j = 1/2$ moments into the scattering plane. This allows us to disentangle excitations of j^z from those of j^x or j^y . The anisotropy in j space translates into a characteristic $\mathbf{q}$ dependence of the intensity that provides a direct signature of bond-directional behavior.

To illustrate the conceptual relation between bond-directional interactions and BDE we consider a central spin on site i surrounded by sites j , connected by bonds $\gamma_{(ij)}$ along crystallographically equivalent directions. When exchange interactions are bond-directional, the magnetic Hamiltonian H_γ for two spins on bond γ has the property that in general $H_\gamma \neq H_{\gamma'}$, even if bonds γ and γ' are equivalent from a structural point of view. When one now creates a magnetic excitation by perturbing the central spin by operator $\hat{O}_i$, the commutator $[H_{\gamma(ij)}, \hat{O}_i]$ in general depends on γ . As a consequence this magnetic excitation distributes *unevenly* over the crystallographically equivalent bonds, breaking lattice symmetry.

An elementary example in which BDE emerge is the honeycomb Kitaev model with $H_\gamma = K S_i^\gamma S_j^\gamma$ where $\gamma = x, y, z$ simultaneously denotes the three nearest-neighbor bonds and the three spin components, signifying the bond-directional character of the interaction. A local operator S_i^x commutes with H_x but not with H_y and H_z on the other two bonds, so that perturbing the system by S_i^x renders the three crystallographic bonds inequivalent and produces BDE. In particular, S_i^x creates localized flux excitations on the two hexagons that share the x bond, while the third hexagon connected to site i remains unaffected, manifestly breaking the three-fold rotational symmetry of the flux distribution and consequently the Majorana modes scattered by the fluxes.

Results

We address the magnetic excitations of the $j = 1/2$ moments in Na_2IrO_3 via RIXS at the Ir L_3 edge. In the following, the term *spin* refers to spin-orbit entangled $j = 1/2$ pseudo-spins. To reveal the bond-directional character of the excitations, we put both the z bond (parallel to b) and the spin component j^z into the horizontal scattering plane, see Fig. 1. Note that j^y is perpendicular to the γ bond. The x, y, z bonds are lying in the honeycomb ab plane, which is parallel to the sample surface. The components j^y are tied to the Ir-O directions within an IrO_6 octahedron. To have j^z in the scattering plane, we tilt the sample by $\chi = 54.7^\circ$ around b , where χ is the angle between the vertical

and the honeycomb ab plane. For a vanishing angle of incidence θ_{in} , the incident beam is parallel to b and in particular the incident polarization is parallel to j^z , which is favorable for our goal. This scattering geometry suppresses the polarization factor $|P^z(\mathbf{q})|^2$ for a large range of $\mathbf{q}$, see Fig. 1d.

RIXS spectra measured at 5 K at selected $\mathbf{q}$ points show two inelastic features peaking around 10 meV and 45 meV, see Fig. 2. Their magnetic character is demonstrated by the resonance behavior measured at 30 K, i.e., above the 3D ordering temperature $T_N = 15$ K, see Supplementary Information. Our focus is on the bond-directional character of excitations expected for a Kitaev material, not on low-energy magnons of the ordered state. In previous RIXS studies, the broad continuum peaking around 45 meV has been established as a generic, quasi-2D magnetic excitation of the $j = 1/2$ honeycomb iridates which persists up to 300 K^{19–24}. A similar continuum has been observed in the $j = 1/2$ honeycomb compound α - RuCl_3 ^{25,26}. The 10 meV peak is particularly pronounced at the K point, which agrees with first RIXS results collected with improved energy resolution $\delta E = 12$ meV²⁰. Observation of the 10 meV feature with $\delta E = 25$ meV requires excellent suppression of elastic Thomson scattering which we achieve by using $2\theta = 90^\circ$. For comparison, reference spectra (open symbols) measured with $2\theta \neq 90^\circ$ are dominated by a strong elastic line peaking at zero energy loss.

In general, RIXS spectra as a function of energy loss are appropriate to study dispersive modes. However, for both inelastic features the peak energies hardly depend on $\mathbf{q}$, see Fig. 2. The key to a microscopic understanding of the two predominantly local excitations is the $\mathbf{q}$ -dependent intensity $I(\mathbf{q}, \omega)$. This is most evident from the astounding behavior at X_x and X_z , see Fig. 2c. These $\mathbf{q}$ points are fully equivalent for the honeycomb lattice but probe different bond directions in the case of nearest-neighbor excitations, as argued below. We will show that a small value of the polarization factor $|P^z(\mathbf{q})|^2$ depicted in Fig. 1d suppresses j^z -conserving excitations and enhances j^z -flip modes. This suppresses the spin-conserving 10 meV feature and enhances the 45 meV spin-flip mode at X_z but not at X_x , i.e., on z bonds but not on x bonds. Hence $I(\mathbf{q}, \omega)$ demonstrates that the magnetic honeycomb lattice of Na_2IrO_3 hosts BDE that have spin-flip or spin-conserving character.

RIXS intensity maps

Figure 3 depicts 2D $\mathbf{q}$ -space maps of $I(\mathbf{q})$ integrated below 15 meV (top row) and above 45 meV (bottom), respectively. The integration ranges have been chosen to disentangle the different behaviors of the two peaks observed in the spectra. For the continuum peaking around 45 meV, the quasi-2D character is supported by the gradual evolution with temperature T , see

Fig. 2 | RIXS spectra at 5 K at selected q points. a Γ' to X_z , b Γ to K , c X_x and X_z , cf. Fig. 1. The spectra show two peaks at about 10 meV and 45 meV, which we attribute to predominantly local nearest-neighbor excitations. Dash-dotted lines in (a) distinguish the two peaks at (5 -6.95). Reference spectra recorded with $2\theta \neq 90^\circ$ and a corresponding large elastic line mark zero energy loss (open symbols, scaled down as indicated). In (c), the different intensities of the two peaks at X_x and X_z provide direct evidence of BDE.

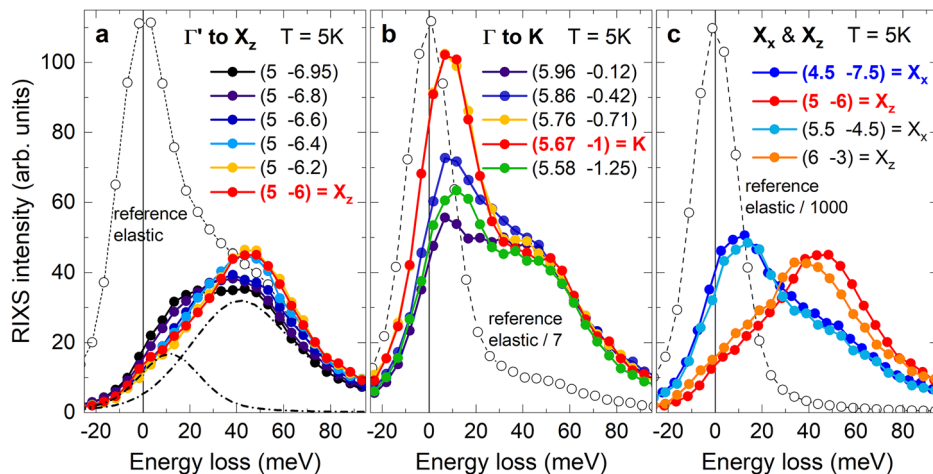
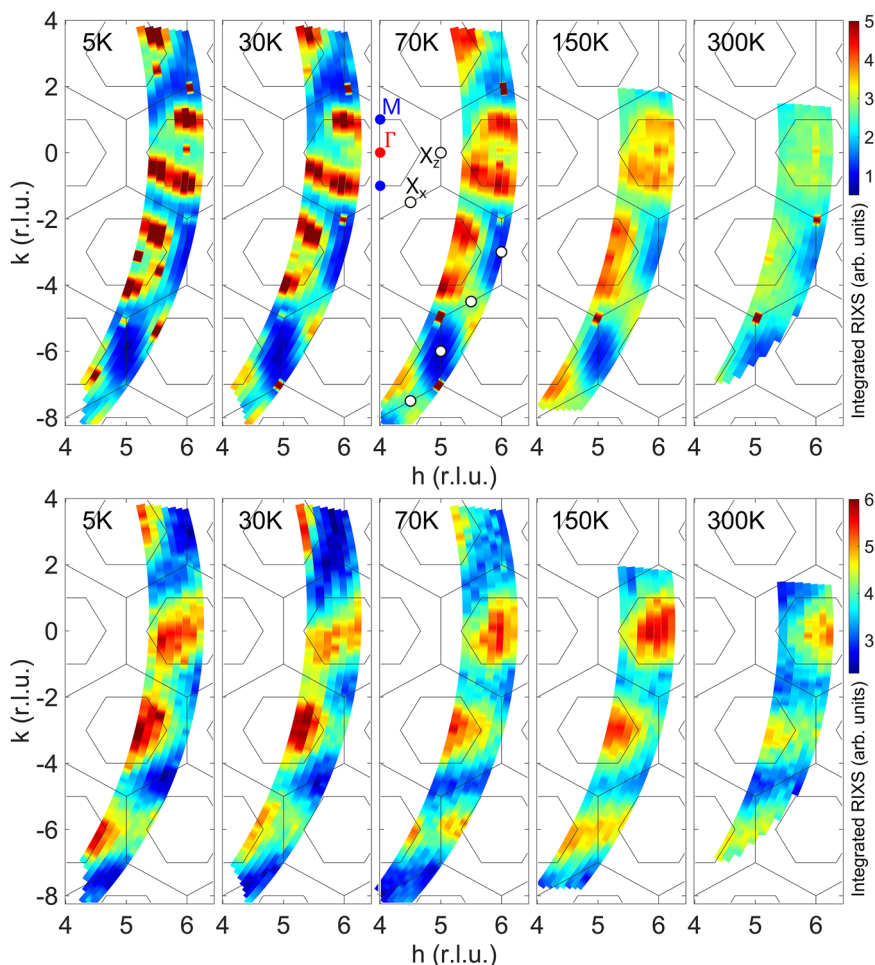


Fig. 3 | RIXS intensity maps at different temperatures, focusing on the behavior of the two peaks shown in the spectra, cf. Fig. 2. The data have been integrated from -15 to 15 meV (top) and from 45 to 125 meV (bottom). At low energy, the narrow spots for integer h and k such as at (6 2) correspond to tails of structural Bragg peaks. White symbols denote X_x and X_z points, where the bond-directional behavior is evident.



bottom panels in Fig. 3. This insensitivity to T and in particular to T_N agrees with previous RIXS results¹⁹ for selected values of q . The overall behavior with the RIXS intensity peaking at Γ can be described by nearest-neighbor correlations (see below).

To study the 10 meV peak, we have to cope with the fact that the dominant contribution to the low-energy intensity below T_N stems from elastic magnetic Bragg scattering with the 3D ordering wave vector $Q_0 = (0 \ 1 \ 1/2)^{27,28}$ and from low-energy magnons that emerge from there²⁹. In the 2D ($h \ k$) maps, the magnetic Bragg spots are not hit

perfectly since the value of l is adapted to achieve $2\theta = 90^\circ$. Nevertheless we find pronounced maxima at $M = \Gamma \pm (0 \ 1)$ and enhanced intensity at M' , cf. Fig. 1c. The narrow features at M and M' are evident in Fig. 4a, which shows the same 5 K data as panel b but on another color scale. Chun et al.¹² analyzed the elastic scattering at M and M' to derive the existence of dominant Kitaev exchange interactions in Na_2IrO_3 . In contrast, we focus on the inelastic response not too close to M and M' .

With decreasing temperature, the low-energy maps in the top panels of Fig. 3 show the building up of intensity close to M , reflecting the evolution of

Fig. 4 | Maps of the RIXS intensity at 5 K. The data have been integrated from -15 to 15 meV (a, b) and from 45 to 125 meV in (d). Compared to Fig. 3, the data were measured on a finer mesh in $\mathbf{q}$ space. a, b show the same data on different color scales. c, e Predictions of a simple bond model for BDE with spin-conserving and spin-flip character, respectively, see Eqs. (1) and (2). Excitations are restricted to either an x , y , or z bond with corresponding correlations of j^x, j^y , or j^z , respectively. The calculated modulation pattern reflects nearest-neighbor correlations, cf. Fig. 5. White circles indicate X_y points. At these, the bond-directional character is most apparent based on polarization-related differences of the RIXS intensity.

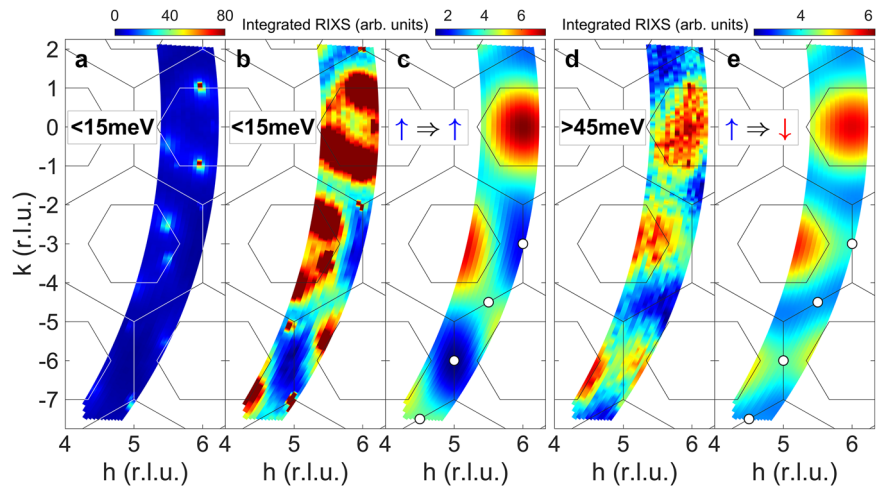
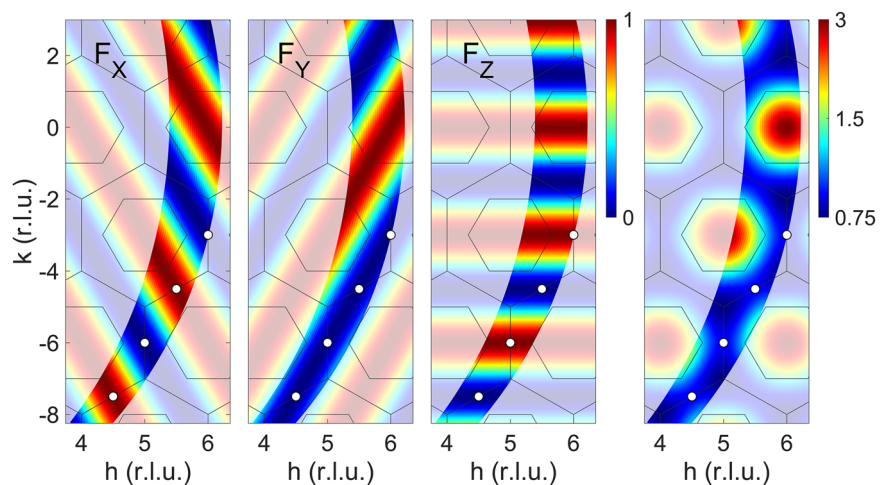


Fig. 5 | Sinusoidal two-site modulation patterns. The three panels on the left depict the structure factor $F_\gamma = \cos^2(\mathbf{q} \cdot \Delta\mathbf{R}_\gamma/2)$ for individual γ bonds with $\gamma = x, y, z$, see Eq. (1). The measured $\mathbf{q}$ range is highlighted, cf. Fig. 3. Right: Sum over x, y , and z bonds. White circles mark X_x and X_z points, these are relevant for the discussion of the bond-directional character, see Fig. 2c.



longer-range 3D correlations. However, central to our study is the behavior in $\mathbf{q}$ ranges not too close to M such as around the four X_y points marked by white symbols in Fig. 3. There, also the low-energy maps are insensitive to T , and the data at 5 K and $30 \text{ K} \approx 2T_N$ are very similar. In agreement with the spectra, cf. Fig. 2c, the low-energy maps show extended ranges of low intensity around $X_z = (5 \text{ } -6)$ and $X_z = (6 \text{ } -3)$ but larger intensity around $X_x = (4.5 \text{ } -7.5)$ and $X_x = (5.5 \text{ } -4.5)$. The opposite behavior is observed for the 45 meV continuum in the high-energy maps in the lower part of Fig. 3, again in agreement with the spectra. The tilted sample geometry of our experiment has been designed to address this particular behavior via the polarization factors, revealing a clear signature of bond-directional behavior, as we will show below.

Below T_N , magnetic order breaks the three-fold rotational symmetry of the honeycomb lattice. However, immediately above T_N short-range zigzag correlations of all three domains were observed with equal strength¹². Therefore, the inequivalence of X_x and X_z above T_N and the corresponding breaking of rotational symmetry cannot be attributed to long-range magnetic order. Furthermore, the RIXS intensity at X_y is insensitive to l , and the intensity maps measured at 5 K on two different magnetic domains are very similar, see Supplementary Information. In sum, the quasi-2D character of the studied excitations is firmly established.

Prevailing nearest-neighbor correlations

From the RIXS intensity maps, we now establish the predominance of nearest-neighbor correlations, which will allow us to, e.g., identify the

response at X_y with the γ bond. A magnetically ordered state is characterized by long-range spin-spin correlations. Above the ordering temperature T_N , thermal fluctuations yield a strong decrease of the correlation length. In quasi-2D compounds, short-range correlations typically survive at temperatures much higher than T_N ³⁰. The Kitaev model describes a very different case in which spin-spin correlations are strictly restricted to two nearest neighbors on a single bond.

This unusual scenario of nearest-neighbor correlations leaves clear fingerprints in the $\mathbf{q}$ dependence of the RIXS intensity¹⁹. The dynamical structure factor of two sites has a sinusoidal shape, i.e., the two-site scattering problem is equivalent to an inelastic incarnation of Young's double-slit experiment³¹. Summing over the three different bonds one finds

$$I_{\text{nn}}(\mathbf{q}) = I_0 + \sum_{\gamma} I_{\gamma}(\mathbf{q}) \cos^2(\mathbf{q} \cdot \Delta\mathbf{R}_{\gamma}/2), \quad (1)$$

where $\Delta\mathbf{R}_{\gamma} = \mathbf{R}_2 - \mathbf{R}_1$ denotes a γ bond with two correlated nearest-neighbor Ir sites, I_0 is a background intensity, and I_{γ} depends on the polarization. Such sinusoidal behavior has been observed for the continuum in the Kitaev material $\alpha\text{-RuCl}_3$ in inelastic neutron scattering²⁶. In RIXS on the honeycomb iridates, Eq. (1) has been found to describe the integrated intensity of the continuum along $\Gamma\text{-}M\text{-}\Gamma'$ and $\Gamma\text{-}K\text{-}X'$ ¹⁹, treating I_0 and I_{γ} as empirical fit parameters. In Fig. 5, we visualize the sinusoidal intensity modulation of each bond for $I_{\gamma} = 1$ and $I_0 = 0$. The sum $I_{\text{nn}}(\mathbf{q})$ in the right panel yields broad maxima at Γ and reduced intensity at Γ' . Comparing

$I_{\text{nn}}(\mathbf{q})$ with the high-energy maps in the bottom panels of Fig. 3 demonstrates that already this simple picture of individual bonds describes the behavior of the continuum surprisingly well. For the low-energy maps, integrated below 15 meV, the RIXS intensity in the first Brillouin zone is dominated, at low temperature, by longer-range correlations of zigzag type, as discussed above. For $\mathbf{q}$ points not too close to M and M' , in particular in the vicinity of X_γ points, we achieve a convincing description in terms of the nearest-neighbor model for both energy ranges. This requires to consider the polarization-dependent matrix elements, as done in Figs. 4c and e and discussed below.

In such a nearest-neighbor scenario with two-site structure factors $F_\gamma = \cos^2(\mathbf{q} \cdot \Delta \mathbf{R}_\gamma/2)$, the data at X_γ selectively probe the response of the γ bond. For instance, $X_z = (5-6)$ hosts a maximum of F_z with vanishing contributions from x and y bonds, see Fig. 5, while $X_x = (4.5-7.5)$ shows a maximum in F_x with $F_y = F_z = 0$.

Bond-directional excitations

With this information on the bond direction, we utilize polarization selection rules to address bond-directional behavior, i.e., whether the application of, e.g., the local operator S_i^x creates an excitation that is connected to a specific bond. In the pure Kitaev model, a RIXS excitation on the γ bond requires to apply $P^\gamma S_i^{\gamma 32}$ with the polarization factor $\mathbf{P} = i\epsilon' \times \epsilon$ and the incident and outgoing polarizations ϵ and ϵ' , respectively. As shown below, this describes the behavior of the 10 meV feature with $I(X_x) > I(X_z)$ but disagrees with the different polarization properties of the 45 meV continuum, with $I(X_x) < I(X_z)$. Therefore, we employ a complementary microscopic approach for both peaks and start from the RIXS matrix element for magnetic excitations of a $j = 1/2$ moment at site $\mathbf{R}_i^{33}$. The general expression for the single-site matrix element is independent of the chosen low-energy model. With the outgoing π and σ polarizations being perpendicular to the incident polarization, see Fig. 1, this matrix element is proportional to $\mathbf{P} \cdot \mathbf{S}_i$, where $\mathbf{S}_i$ operates within the $j = 1/2$ subspace. We apply this matrix element in a bond-directional scenario of nearest-neighbor excitations. More precisely, for the two sites of, e.g., a z bond, we consider that S_i^z creates a spin-conserving (sc) excitation while S_i^x or S_i^y yield a spin flip (sf) of the z component. For $I_\gamma(\mathbf{q})$ in Eq. (1) this yields

$$I_z^{\text{sc}}(\mathbf{q}) \propto |P^z(\mathbf{q})|^2 \text{ and } I_z^{\text{sf}}(\mathbf{q}) \propto |P^x(\mathbf{q})|^2 + |P^y(\mathbf{q})|^2 \quad (2)$$

and equivalent expressions for x and y bonds. We have chosen the experimental geometry with the aim to strongly suppress $|P^z(\mathbf{q})|^2$ in the lower parts of the maps, see Fig. 1d. More precisely, $|P^z|^2 \propto \sin^2(\theta_{\text{in}})$, and the angle of incidence θ_{in} is varied from about 5° to 65° . This corresponds to an (h,k) range where k roughly runs from -8 to 4 and depends roughly linearly on θ_{in} . In contrast, $|P^x(\mathbf{q})|^2$ and $|P^y(\mathbf{q})|^2$ are nearly constant and large, see Supplementary Information. In the bond-directional scenario described by Eq. (2), the suppression of $|P^z|^2$ switches off spin-conserving excitations on z bonds while it reduces the intensity of spin-flip excitations on x or y bonds by about a factor 2. This explains the stunning difference of the spectra measured at X_z and X_∞ , see Fig. 2c, if we attribute the peaks at 10 and 45 meV to spin-conserving and spin-flip excitations, respectively, and identify X_γ with the γ bond, as appropriate for nearest-neighbor correlations.

Finally, we calculate maps of the RIXS intensity $I_{\text{nn}}(\mathbf{q})$ expected for BDE, see Eq. (1). This combines the sinusoidal structure factors F_γ of the nearest-neighbor model, containing information on the bond direction, with the polarization-dependent $I_\gamma(\mathbf{q})$ given in Eq. (2), reflecting the involved spin component. Figure 4c, e plot $I_{\text{nn}}(\mathbf{q})$ for spin-conserving and spin-flip excitations, respectively. The excellent agreement with the RIXS data in panels b and d, in particular in the vicinity of the X_γ points, corroborates the above assignment of spin-conserving and spin-flip excitations at low and high energies, respectively. The continuum, integrated above 45 meV, is described very well over the entire range of $\mathbf{q}$ and for all studied temperatures. For integration below 15 meV, our nearest-neighbor model represents the RIXS data very well at high T , such as 70 K or 150 K. At 5 K, it still captures the behavior not too close to M , while the response in the

vicinity of M reflects longer-range correlations. Altogether, bond-directional behavior is most evident around X_γ points. For instance, X_x and X_z are equivalent on the honeycomb lattice and within the nearest-neighbor model for isotropic $I_\gamma = 1$, see Fig. 5. The anisotropic, bond-directional character of the magnetic excitations in Na_2IrO_3 yields very different RIXS intensities around X_x and X_z as well as different behavior at low and high energy.

Discussion

The honeycomb compound Na_2IrO_3 , like related materials, has been dubbed a *proximate spin liquid*^{17–19}. It exhibits aspects of a spin liquid despite showing long-range magnetic order at low temperature. This can be traced back to additional exchange terms beyond dominant Kitaev exchange, such as isotropic Heisenberg coupling³⁴. Our model calculations do not explicitly rely on the Kitaev model but rather assume that spin-spin correlations for the considered excitations are bond-directional and restricted to two sites. On this basis, we demonstrate the existence of bond-directional magnetic excitations, establishing a hallmark of systems with bond-directional interactions. In particular the high-energy mode around 45 meV is very well described by a bond-directional nearest-neighbor scenario. Note that such a spin-flip mode does not arise in the pure Kitaev model. Remarkably, the RIXS response around 10 meV, not too close to the M points, is also very well described by bond-directional nearest-neighbor excitations, even in the magnetically ordered state. Previously, RIXS on Kitaev iridates has been used to address the coupling constants of a minimal magnetic model²⁰. Such experiments typically focus on the excitation energies within a restricted $\mathbf{q}$ range such as the first Brillouin zone. Our results demonstrate that one has to investigate the RIXS intensity over an extended range of $\mathbf{q}$ to achieve a comprehensive picture of the magnetic excitations.

The identification of the bond-directional character via the $\mathbf{q}$ -dependent RIXS intensity is similar in spirit to identifying the dipolar character of spin correlations in spin ice via the existence of pinch points in spin-flip neutron scattering³⁵. Concerning the $\mathbf{q}$ dependence, it is interesting to consider the differences between RIXS and inelastic neutron scattering. Magnetic neutron scattering depends on the spatial extent of the magnetization density, which yields a magnetic form factor that decays quickly with $\mathbf{q}$ and may be anisotropic, as discussed for $\alpha\text{-RuCl}_3$ ³⁶, or reflect hybridization with the ligands³⁷. In contrast, the RIXS process involves an almost point-like core hole, which is equivalent to a nearly constant form factor³¹. This allows us to study the intensity modulation over a large range of $\mathbf{q}$.

In the ongoing quest to identify Kitaev materials, the observation of bond-directional excitations via our advanced RIXS scheme can play a decisive role in validating, for a given candidate compound, the presence of bond-directional interactions. These are appreciated as an alternative source of frustration beyond the geometric frustration of non-bipartite lattices and key to the emergence of non-conventional forms of magnetism (such as discussed for the fcc $j = 1/2$ material $\text{Ba}_2\text{CeIrO}_6$ ³⁸) and potentially spin-liquid ground states (as hypothesized³⁹ for the $j = 1/2$ pyrochlore material $\text{Yb}_2\text{Ti}_2\text{O}_7$). A first step in this direction might be to apply our approach to $\alpha\text{-Li}_2\text{IrO}_3$ and $\alpha\text{-RuCl}_3$, two widely studied honeycomb Kitaev materials, but also to $j = 1/2$ systems which have not attracted primary interest for bond-directional exchanges, such as Sr_2IrO_4 , where their occurrence would add to the puzzle to identify routes to superconductivity in the presence of spin-orbit coupling in materials isostructural to cuprate superconductors. In further developing our experimental approach a natural next step is to validate other forms of bond-directional interactions, such as dominating off-diagonal Γ -interactions^{8,9}, and their manifestation in bond-directional excitations.

Methods

Crystal growth and characterization

Single crystals of Na_2IrO_3 were grown following the procedure described in ref. 40 with 10% extra Ir at 1323 K for 14 days. The crystal quality has been checked by X-ray diffraction and magnetic measurements. Na_2IrO_3 exhibits the monoclinic space group $C2/m$ with the lattice parameters $a = 5.427(1)$ Å,

$b = 9.395(1) \text{ \AA}$, $c = 5.614(1) \text{ \AA}$, and $\beta = 109.037(18)^\circ$ at 300 K^{28} . It shows antiferromagnetic zigzag order below $T_N = 15 \text{ K}^{34}$. Laue diffraction of our samples indicates structural 120° twinning in the honeycomb plane, which below T_N dictates the formation of corresponding magnetic domains. These have no impact on our results, as discussed in Supplementary Information.

RIXS

We measured RIXS at the Ir L_3 edge at beamline ID20 at the ESRF^{41,42}. The incident energy $E_{\text{in}} = 11.2145 \text{ keV}$ resonantly enhances magnetic excitations of the $j = 1/2$ moments. The energy resolution is $\delta E = 25 \text{ meV}$. The elastic contribution due to Thomson scattering is almost fully suppressed by strictly using a scattering angle $2\theta = 90^\circ$, where all outgoing polarization contributions are perpendicular to the incident π polarization, see Fig. 1. The resolution $\delta \mathbf{q}$ of the transferred momentum approximately amounts to $(\pm 0.05 \pm 0.1 \pm 0.05)$ reciprocal lattice units (r.l.u.) using an iris with 60 mm diameter on the $R = 2 \text{ m Si}(844)$ spherical diced analyzer. We measured on a sample surface of $2 \text{ mm} \times 2 \text{ mm}$ within the honeycomb plane. All RIXS data were corrected for self-absorption⁴³.

Data availability

The data shown in the figures are available on Zenodo⁴⁴ <https://doi.org/10.5281/zenodo.21810173>.

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Author contributions

M.M. and M.G. conceived the experiment. M.M., M.M.S., M.H.U., J.K., P.H.M.v.L., and M.G. carried out a proof-of-principles RIXS study at beamline 27-ID at APS. M.M., K.H., C.J.S., M.M.S., G.M., and M.G. performed the RIXS experiments at beamline ID20 of the ESRF. M.M. analyzed the experimental data. I.M.P., F.B., and P.G. synthesized and characterized the single crystals used for the measurements. M.M. and M.G. performed the theoretical calculations with support from J.A., C.H., S.T., and J.v.d.B. Finally, M.M., M.G., S.T., and J.v.d.B. wrote the manuscript with input from all authors.

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Competing interests

The authors declare no competing interests.

Additional information

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