

Spin dynamics and possible topological magnons in the nonstoichiometric pyrochlore iridate $\text{Tb}_2\text{Ir}_2\text{O}_7$ studied by RIXS

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We report on a resonant inelastic x-ray scattering study on a single crystal of a nonstoichiometric pyrochlore iridate $\text{Tb}_{2+x}\text{Ir}_{2-x}\text{O}_{7-y}$ ($x \simeq 0.25$) that magnetically orders at $T_N \simeq 50$ K. We observe a propagating gapped magnon mode at low energy and model it using a Hamiltonian consisting of a Heisenberg exchange [$J = 16.2(9)$ meV] and Dzyaloshinskii-Moriya interactions [$D = 5.2(3)$ meV], which shows the robustness of interactions despite Tb stuffing at the Ir site. Strikingly, the ratio $D/J = 0.32(3)$ supports possible nontrivial topological magnon band crossing. This material may thus host coexisting fermionic and bosonic topology, with potential for manipulating electronic and magnonic topological bands thanks to the d - f interaction.

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Iridium-based compounds hosting a $j_{\text{eff}} = \frac{1}{2}$ ground state have attracted a lot of interest due to the delicate interplay between large spin-orbit coupling (SOC), moderate electronic correlations, and the crystal field that can give rise to exotic electronic and magnetic states [1–3]. Topological phases of matter are predicted to arise such as topological insulators, axion insulators, and Weyl semimetal (WSM) phases [4].

In this context, pyrochlore iridates with the formula $A_2\text{Ir}_2\text{O}_7$ ($A = \text{Y}$, or rare-earth Lu-Pr) have been extensively studied, as these systems were potential candidates to realize WSM phases driven by magnetic ordering [5–10]. These compounds crystallize in the $Fd\bar{3}m$ cubic structure (space group No. 227) consisting of two interpenetrating rare-earth and iridium pyrochlore sublattices (Fig. 1) that are connected via a d - f interaction, making them a fertile playground to discover phases and to study frustrated magnetism [1,2,5,11]. The whole family of pyrochlore iridates exhibits a metal-to-insulator transition (MIT) when cooling down in temperature, concomitant with magnetic ordering (except for $A = \text{Pr}$) [12]. The iridium sublattice was shown to order in an all-in-all-out (AIAO) magnetic structure (Fig. 1) compatible with the realization of a WSM state [8], using various techniques

such as μSR , neutron powder diffraction, and resonant elastic x-ray scattering [13–19]. While the existence of the WSM phase in pyrochlore iridates is still theoretically debated [7–11,20–22], experimental signatures of fermionic band topology in these compounds were observed, e.g., through optical terahertz spectroscopy [23] or magnetoresistivity measurements, where field-induced WSM phases were observed in $\text{Nd}_2\text{Ir}_2\text{O}_7$ [24–26]. Authors of studies of thin films also showed topological properties of the WSM phase by breaking cubic symmetry through epitaxial strain [27–29].

Interestingly, topological properties in pyrochlore iridates extend beyond fermions. Indeed, like its ferromagnetic counterpart found in the pyrochlore compound $\text{Lu}_2\text{V}_2\text{O}_7$, where a magnon Hall effect [30,31] was observed, authors of recent studies have proposed that pyrochlore iridates could host topological bosons, i.e., topologically nontrivial magnon bands depending on the ratio of the Dzyaloshinskii-Moriya interaction (DMI) D and Heisenberg interaction J [32,33]. More precisely, if $D/J > 0.28$, magnon band crossing is predicted to occur close to the Γ point. This makes pyrochlore iridates unique systems where both fermionic and bosonic band topology coexist and hence potential platforms to realize topological electronic and magnonic devices [34]. A better understanding of the spin dynamics in these compounds is thus crucial yet challenging. The Ir^{4+} cation is indeed a strong neutron absorber and possesses a small magnetic moment ($m_{\text{Ir}} \simeq 0.3 \mu_B$), making inelastic neutron scattering unsuccessful up to now. Resonant inelastic x-ray scattering (RIXS), on the other hand, is a powerful tool since it allows us to probe spin and orbital dynamics in iridium compounds due

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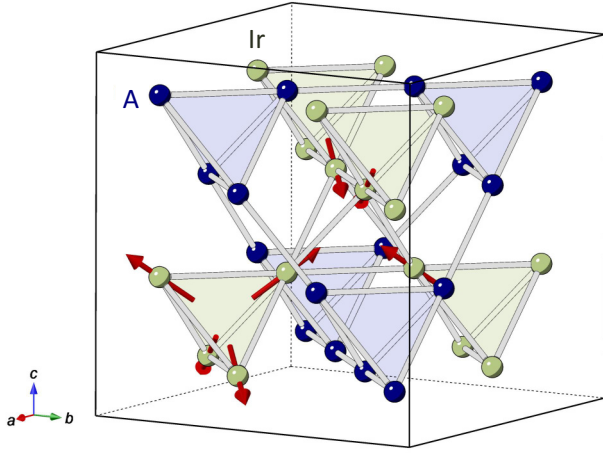


FIG. 1. Pyrochlore structure of $A_2\text{Ir}_2\text{O}_7$ with two interpenetrating rare-earth (blue spheres) and iridium sublattices (yellow spheres) forming corner-shared tetrahedra. The all-in-all-out (AIAO) magnetic order corresponds to spins (red arrows) pointing either within or out of a given tetrahedra. Such a magnetic configuration is depicted only on two tetrahedra for better clarity.

to resonant processes enhancing magnetic scattering cross-sections [35]. It is worth noting that, while the spin dynamics in pyrochlore iridates has been explored in systems with nonmagnetic rare-earth ions [36–38], little attention has been given to those with magnetic rare-earth ions, where the d - f interaction may modify electronic and magnonic band structure and thus drive topological phases [1, 11, 39]. Specifically, $\text{Tb}_2\text{Ir}_2\text{O}_7$ is of interest owing to the relatively large moments of the Tb^{3+} cations ($m_{\text{Tb}} \simeq 5.2 \mu_B$) [19] that could easily couple to a magnetic field and thus potentially drive exotic phase transitions, as observed in $\text{Nd}_2\text{Ir}_2\text{O}_7$ [24–26].

In this letter, we report on a study using RIXS at the Ir L_3 edge of a nonstoichiometric single crystal of $\text{Tb}_{2+x}\text{Ir}_{2-x}\text{O}_{7-y}$ ($x \simeq 0.25$) in which the transition temperature is strongly suppressed down to 50 K (instead of 130 K for stoichiometric powder $\text{Tb}_2\text{Ir}_2\text{O}_7$). Analysis of the orbital dd intra- t_{2g} excitations yields SOC $\xi = 0.40(1)$ eV and trigonal distortion of the IrO_6 octahedra $\Delta = 0.52(2)$ eV. We observe a propagating gapped magnetic excitation and analyze the dispersion spectra with a linear spin-wave model. This allows us to extract Heisenberg exchange interaction $J = 16.2(9)$ meV and DMI $D = 5.2(3)$ meV, leading to the ratio $D/J = 0.32(3)$. Our findings demonstrate that interactions remain robust despite Tb stuffing on the Ir site. More strikingly, the found $D/J = 0.32(3)$ ratio supports the possible presence of topological magnons in our stuffed sample of $\text{Tb}_2\text{Ir}_2\text{O}_7$. This material may thus host coexisting fermionic and bosonic topology, with potential for manipulating electronic and magnonic topological bands via the d - f interaction.

Here, $\text{Tb}_{2+x}\text{Ir}_{2-x}\text{O}_{7-y}$ single crystals were flux-grown as described in Ref. [40] (see inset of Fig. 2). Electron dispersive x-ray spectroscopy (EDX) was used to probe the cation ratio, which revealed an excess of Tb ions of $x \simeq 0.25$ with no reliable estimation of the oxygen content (details in the Supplemental Material [41]). The crystal structure probed by single-crystal x-ray diffraction gives a

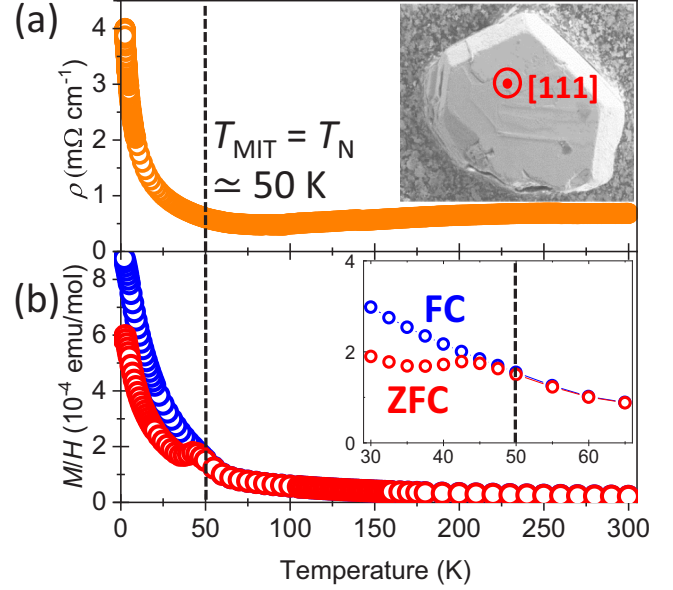


FIG. 2. (a) Resistivity ρ vs temperature. Inset shows a photograph of the sample of $\text{Tb}_{2+x}\text{Ir}_{2-x}\text{O}_{7-y}$ of size $L \times l \times h \simeq 0.3 \times 0.3 \times 0.2$ mm³. The sample has a hexagonal shape with the [111] direction perpendicular to its surface. (b) Zero-field-cooled (ZFC)-field-cooled (FC) magnetization data. The inset shows a zoom around the transition temperature $T_N = T_{\text{MIT}} \simeq 50$ K, where a bifurcation between the ZFC-FC curves is observed.

$Fd\bar{3}m$ pyrochlore structure with lattice parameter $a = 10.26$ Å, slightly larger than previously reported in $\text{Tb}_2\text{Ir}_2\text{O}_7$ powder samples [17, 19].

Electrical and magnetic macroscopic properties were characterized by four-probe resistivity and magnetization measurements using PPMS and MPMS Quantum Design apparatus, respectively. Resistivity vs temperature measurements show a MIT concomitant with a magnetic ordering of the iridium sublattice at $T_N = T_{\text{MIT}} \simeq 50$ K, as observed by the bifurcation between the zero-field-cooled (ZFC) and field-cooled (FC) magnetization data (Fig. 2). This magnetic ordering is further confirmed by resonant x-ray magnetic diffraction measurements (details in the Supplemental Material [41]). This value is much lower than for pure $\text{Tb}_2\text{Ir}_2\text{O}_7$ [12], indicating that stuffing tends to suppress the onset of magnetic order. This is associated with a resistivity which below T_{MIT} is 3–4 orders of magnitude lower than for $\text{Tb}_2\text{Ir}_2\text{O}_7$ [12]. These observations are consistent with previous studies on stuffed pyrochlore iridates and especially $\text{Tb}_{2+x}\text{Ir}_{2-x}\text{O}_{7-y}$ ($x = 0.18$) [49], where off-stoichiometry was shown to favor metallic or semimetallic behaviors [18, 50, 51].

A RIXS experiment at the iridium L_3 edge was performed on the beamline ID20 of the European Synchrotron Radiation Facility with an overall energy resolution of 25 meV [42] (details in the Supplemental Material [41]). The scattering plane and incident photon polarization were both horizontal in the laboratory frame, i.e., π incident polarization was used (inset in Fig. 3). To enhance the intra- t_{2g} magnetic and orbital signal, we tuned E_{in} to a resonant condition, specifically the Ir L_3 edge in this case. We thus set $E_{\text{in}} = 11.215$ keV in this letter.

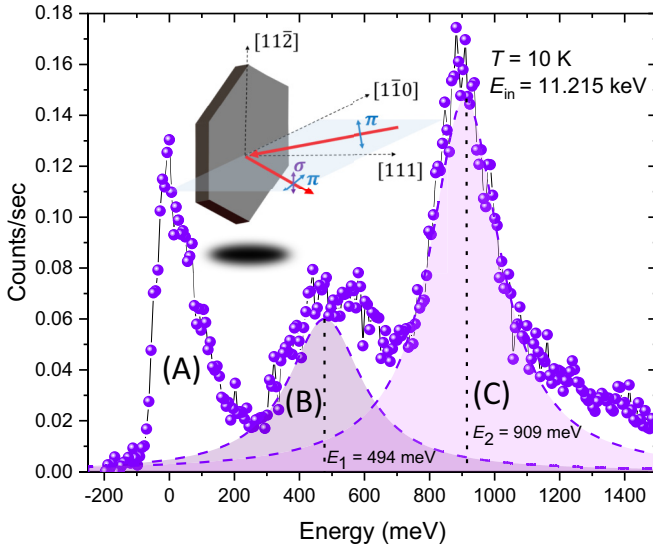


FIG. 3. High-resolution resonant inelastic x-ray scattering (RIXS) spectrum collected at $T = 10$ K and $\mathbf{Q} = (7.5, 7.5, 7.5)$. The incident energy is set to $E_{\text{in}} = 11.215$ keV. Full purple circles denote the experimental data. An incoherent elastic contribution with low-energy excitations (A) is observed. Two higher-energy dd intra- t_{2g} excitations (B) and (C) are visible and fitted with Lorentzian functions (dashed purple and pink curves), leading to $E_1 = 494(7)$ meV and $E_2 = 909(3)$ meV. The inset shows the horizontal geometry used in the RIXS experiment with the scattering plane defined by the $[111]$ and $[1\bar{1}0]$ directions of the sample.

Figure 3 depicts a high-energy resolution RIXS spectrum measured at $T = 10$ K. Within the spectrum, three features are observed. First, there is an incoherent elastic contribution followed by a low-energy inelastic signal (A) down to 300 meV. Then two higher-energy excitations (B) and (C) are observed ~ 0.5 and 1 eV. By fitting the latter two excitations with Lorentzian functions, their energies are found to be $E_1 = 494(7)$ meV and $E_2 = 909(3)$ meV, respectively. Note that these excitations do not disperse, as previously observed in other pyrochlore iridates [41]. *Ab initio* calculations [52] along with RIXS measurements [18] have shown that these two excitations correspond to dd excitations within the t_{2g} manifold. The presence of these two features is explained by the combination of SOC ξ and trigonal distortion Δ of the IrO_6 octahedra, which can be described by a single-ion model for Ir^{4+} in a $5d$ t_{2g} basis subjected to trigonal distortion [44,52]:

$$H = \xi \mathbf{LS} - \Delta L_z^2,$$

where ξ denotes the SOC and Δ the trigonal distortion with the quantization axis z corresponding to the threefold rotation axis of the octahedron. From an effective model, one can extract ξ and Δ from the dd excitations with energies E_1 and E_2 with the following relations [44,52]: $\lambda = 2(2E_1 - E_2)/(3 - \delta)$, $\Delta = \lambda\delta/2$, with $\delta = -b - \sqrt{b^2 - 9}$, $b = (1 + 3a^2)/(1 - a^2)$ and $a = E_2/(E_2 - 2E_1)$. We thus extracted the following values: $\xi = 0.40(1)$ eV and $\Delta = 0.52(2)$ eV, which are consistent with the values found for other pyrochlore iridates [18,52]. We now turn to the analysis of the low-energy excitations < 300 meV.

Figures 4(a)–4(c) show the RIXS spectra measured at $T = 10$ K at the zone-center $\mathbf{Q} = \mathbf{G} = (7, 7, 7)$, $\mathbf{Q} = (7.1, 7.1, 7.1)$, and $\mathbf{Q} = (7.5, 7.5, 7.5)$. Three main features appear: (1) an incoherent elastic contribution centered at zero energy loss, (2) a sharp dispersing mode ~ 20 –50 meV, and (3) a broad continuum of excitations from 50 to 300 meV. The dispersing sharp mode (2) corresponds to a magnetic excitation, as it is not present at room temperature [41]. The continuum of excitations (3) does not seem to disperse, and its nature is still unclear at present. Indeed, this broad continuum, previously seen in other RIXS studies of pyrochlore iridates [18,36,37], has a broad linewidth and is temperature independent. Its origin has been attributed to a particle-hole continuum, e.g., interband excitations, or incoherent multimagnetic excitations [37].

To fit our data and follow precisely the dispersion of the magnetic excitation, we used a damped harmonic-oscillator model, also used in Ref. [37] (note that fits using a Lorentzian curve for the single-magnon gave the same results on its energy position):

$$S(\mathbf{q}, \omega) = \frac{A_{\mathbf{q}}}{(1 - e^{-\omega/T})} \left[\frac{\gamma_{\mathbf{q}}}{(\omega - \omega_{\mathbf{q}}) + \gamma_{\mathbf{q}}^2} - \frac{\gamma_{\mathbf{q}}}{(\omega + \omega_{\mathbf{q}}) + \gamma_{\mathbf{q}}^2} \right],$$

where $\omega_{\mathbf{q}}$ is the energy peak position, $\gamma_{\mathbf{q}}$ the peak width, and $A_{\mathbf{q}}$ the overall amplitude. The broad inelastic continuum (3) was fitted using the same function, while the elastic background (1) was fitted using a Pseudo-Voigt function. Figures 4(d)–4(h) show scans along high-symmetry directions within the Brillouin-zone [see inset of Fig. 4(a)]: Γ -X, X-W, W-L, Γ -L, and Γ -K, respectively. By fitting the whole set of data with the above fitting function, we were able to precisely obtain the full dispersion of the magnetic mode, as depicted in Fig. 4(i). Our data indicate a spin gap of 27 meV and a dispersion width of 12 meV. This gapped spectrum is consistent with an AIAO magnetic order [36,37]. The magnon mode is strongly dispersing along Γ -L and Γ -K while almost not dispersing along X-W and W-L. Interestingly, it weakly disperses along Γ -X, in contrast with what was reported in $\text{Sm}_2\text{Ir}_2\text{O}_7$ and $\text{Eu}_2\text{Ir}_2\text{O}_7$ [36,37]. The intensity of the magnon mode is strongly suppressed when moving away from the Γ point, consistent with what was observed previously in other pyrochlore iridates [36,37].

To model our data, we use a linear spin-wave approach including the minimal spin Hamiltonian for AIAO pyrochlore iridates (details in the Supplemental Material [41]):

$$H = \sum_{\langle i,j \rangle} J \mathbf{S}_i \mathbf{S}_j + \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j),$$

where J corresponds to the Heisenberg exchange interaction between nearest neighbors $\langle i, j \rangle$ and $\mathbf{D}_{ij}$ the antisymmetric DMI. Using SpinW [48], the experimental magnon dispersion was fitted through linear spin-wave calculations [see Fig. 4(i)], yielding $J = 16.2(9)$ meV and $D = |\mathbf{D}_{ij}| = 5.2(3)$ meV and thus a ratio $D/J = 0.32(3)$. The calculated curves capture very well the dispersion along Γ -K, Γ -L, and L-W. However, discrepancies appear along X-W, and only the lowest-energy magnon band captures the dispersion of the magnon mode along Γ -X. These discrepancies could be explained by the

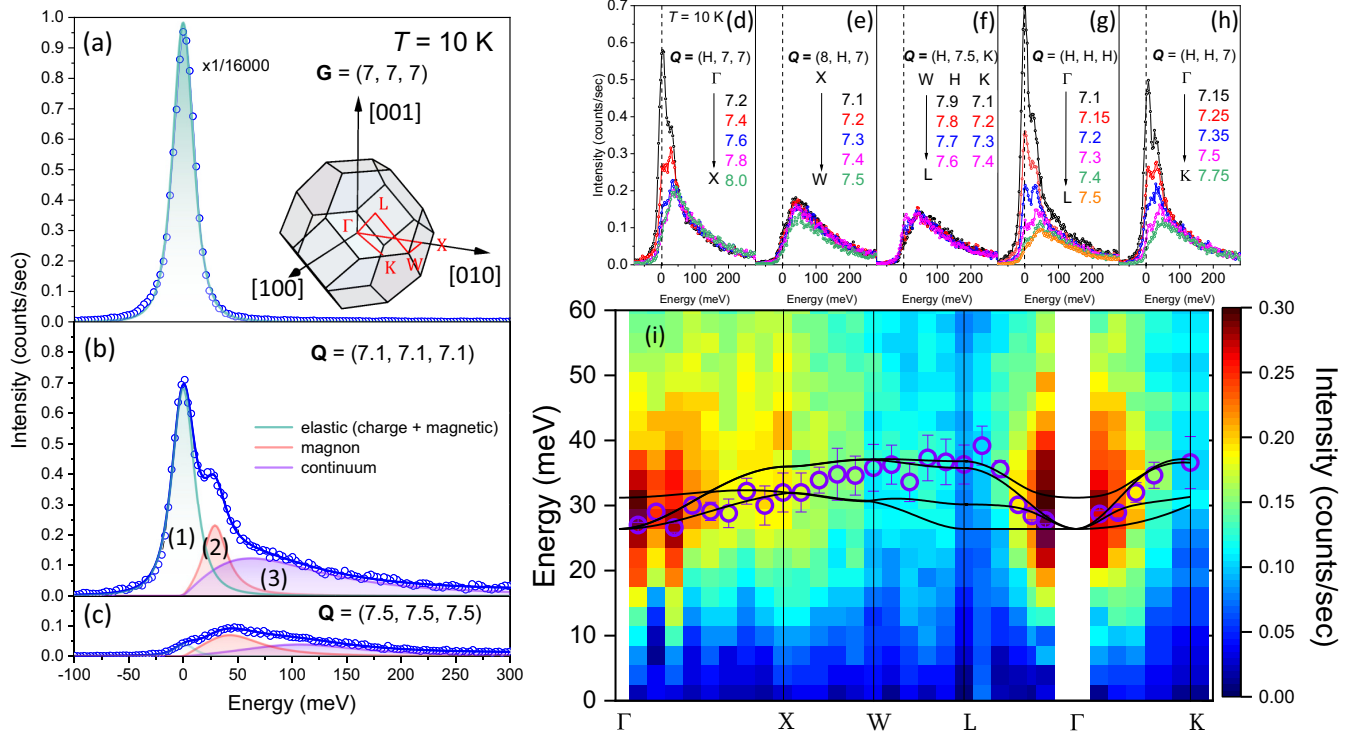


FIG. 4. High-resolution resonant inelastic x-ray scattering (RIXS) spectra collected at $T = 10$ K and (a) $\mathbf{Q} = \mathbf{G} = (7, 7, 7)$, (b) $\mathbf{Q} = (7.1, 7.1, 7.1)$, and (c) $\mathbf{Q} = (7.5, 7.5, 7.5)$. The intensity in (a) is reduced by a factor of 16 000 for comparison with other spectra. The elastic line (1) (green curve) is fitted using a Pseudo-Voigt function. The spectra in (b) and (c) show a sharp inelastic feature (2) that corresponds to the magnon (red solid line) and an inelastic continuum (3) (purple solid line). Open blue circles correspond to raw data. The global fitting curve (blue solid line) corresponds to the sum of those three features. Inset: First Brillouin zone (BZ) of the face-centered cubic lattice with high-symmetry points and directions. RIXS spectra recorded at $T = 10$ K and along the high-symmetry directions within the BZ: (d) Γ -X, (e) X-W, (f) W-L, (g) Γ -L, and (h) Γ -K. (i) Dispersion spectra obtained from (d)–(h). The elastic line has been subtracted for more clarity. The open purple circles denote the energy of the magnetic excitations extracted from the fits of the spectra in (d)–(h) and the associated error bars. The four black lines denote the calculated four nondegenerate magnon dispersions using SpinW [48] with $J = 16.2$ meV and $D = 5.2$ meV, leading to the best agreement with the experimental dispersion.

fact that a spin-wave approach assumes strong electron correlations U , while pyrochlore iridates were shown to be in a more itinerant regime [37]. To go further, a more sophisticated approach such as random phase approximation of the dynamical spin susceptibility would be necessary [53]. The presence of Ir^{4+} vacancies should also be considered, but it is expected to mainly introduce a broadening of the magnon bands. Given the low-energy resolution of RIXS, this does not appear crucial in the present analysis, and one can consider the J and D values as average values over the random distribution of Ir^{4+} vacancies in $\text{Tb}_{2+x}\text{Ir}_{2-x}\text{O}_{7-y}$.

Interestingly, the $D/J = 0.32(3)$ value found here supports nontrivial topological magnon band crossing as pointed out by authors of recent theoretical studies [33,38,54]. Indeed, for $D/J > 0.28$ magnon band crossing occurs along Γ -X, which is reproduced by our spin-wave calculations. The D/J ratio depends on the Ir-O-Ir bond angle, which itself depends linearly on the ionic radius of the A^{3+} cation in $A_2\text{Ir}_2\text{O}_7$. Hence, D/J is expected to decrease smoothly within the Lu-Nd series of magnetically AIAO-ordered pyrochlore iridates. More specifically, J and D decrease and increase, respectively, with the size of the ionic radius. Authors of recent Raman studies showed that, in the case of Y^{3+} , owing to a small ionic radius, $\text{Y}_2\text{Ir}_2\text{O}_7$ hosts $D/J = 0.68$, with $D = 9.0$ meV

and $J = 15.1$ meV. Consistently, authors of RIXS studies on $\text{Eu}_2\text{Ir}_2\text{O}_7$ and $\text{Sm}_2\text{Ir}_2\text{O}_7$, thus with larger ionic radius, found a ratio of $D/J = 0.26, 0.18$ [33,36,37]. Our findings are consistent with this dependence of J and D with the ionic radius, i.e., Tb^{3+} has an intermediate size and is placed between Y^{3+} and Eu^{3+} , Sm^{3+} [38]. These results nevertheless contrast with the ones calculated for $\text{Tb}_2\text{Ir}_2\text{O}_7$ through quantum chemistry calculations, which proposed a scenario where $D = 5$ meV $\gg J = -1.5$ meV [47]. This may be due to a different value of the Ir-O-Ir angle used in the calculations compared with the material [38]. Finally, we demonstrate that the exchange couplings are robust despite stuffing. This could be explained by the large extended $5d$ orbitals of the Ir^{4+} cations and the three-dimensional character of this system. The precise effect of stuffing and how the exchange couplings are affected in comparison with pure $\text{Tb}_2\text{Ir}_2\text{O}_7$ are nevertheless unclear, and we leave this open question for future studies. Note that recent results in Nd-based pyrochlores have shown that magnetic interactions are little affected even in the presence of strong disorder and high off-stoichiometry [55].

In summary, we investigated a stuffed sample of $\text{Tb}_{2+x}\text{Ir}_{2-x}\text{O}_{7-y}$ ($x \simeq 0.25$) by means of RIXS. The low-energy spectra exhibit a dispersive magnon gapped excitation consistent with AIAO order. A linear spin-wave modeling

of the experimental magnon dispersion allowed us to extract the Heisenberg interaction $J = 16.2(9)$ meV and the DMI $D = 5.2(3)$ meV, leading to $D/J = 0.32(3)$. These values are consistent with the trend of J and D that depends on Ir-O-Ir bond angle and hence ionic radius. Strikingly, the ratio D/J found here supports topological magnon band crossing in stuffed $\text{Tb}_2\text{Ir}_2\text{O}_7$. This establishes this material as an interesting platform where both fermionic and magnonic topology coexist and where electronic and magnonic bands could be modulated thanks to the d - f exchange. We also demonstrate that magnetic interactions are robust despite the presence of stuffing. We hope our findings will motivate future complementary measurements, e.g., Raman scattering, thermal Hall measurements, or RIXS with higher-energy resolution

to demonstrate the presence of nontrivial topological magnon bands in stuffed and pure $\text{Tb}_2\text{Ir}_2\text{O}_7$.

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