

Supplementary Information for: Spectrally sharp magnetic excitations above the critical temperature in a frustrated Weyl semimetal

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S1 X-ray characterization of the $\text{Y}_2\text{Ir}_2\text{O}_7$ film

To assess the epitaxial relationship and strain condition between the film and the substrate, reciprocal space mapping (RSM) measurements were taken around the YSZ (4 0 2) peak. Measurements were taken at Rutgers University with an Empyrean X-ray diffractometer manufactured by Malvern Panalytical. The voltage and current were set to 45 kV and 40 mA, respectively. The sample was probed with monochromated Cu $K\alpha$ light ($\lambda = 1.5406\text{\AA}$) and the diffracted beam was detected with a PIXcel^{3D} area detector. Fig. 1 clearly shows the YIO (8 0 4) peak, indicating epitaxial growth as expected. Furthermore, the YIO (8 0 4) peak lies along the radial line (e.g. goes through the origin of the reciprocal space), indicating that the strain of the film is fully relaxed.

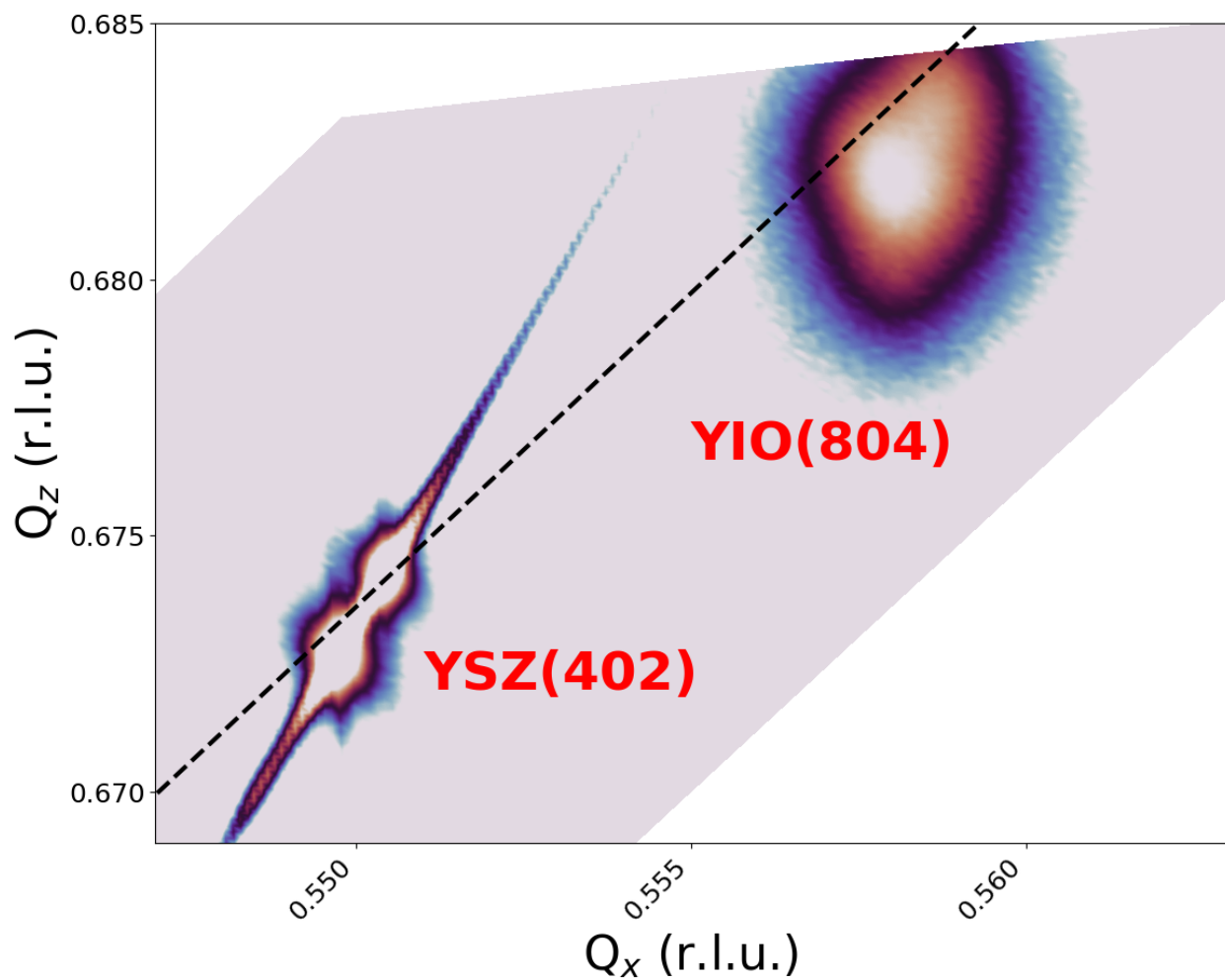


Figure S1: **Reciprocal space mapping around the YSZ (4 0 2) Bragg peak.** The YIO (8 0 4) diffraction is observed along the radial direction in the reciprocal space. Axes plotted in reciprocal lattice units of YSZ.

S2 Transport

To compare the resistivity behavior between $\text{Y}_2\text{Ir}_2\text{O}_7$ and $\text{Eu}_2\text{Ir}_2\text{O}_7$, electrical transport measurements were taken between 2K and 300K. Measurements were taken at Rutgers University with a Physical Properties Measurement System manufactured by Quantum Design. Fig. S2 shows that $\text{Eu}_2\text{Ir}_2\text{O}_7$ has a minimum in resistivity around 105 K, indicating a metal-insulator transition at T_N . $\text{Y}_2\text{Ir}_2\text{O}_7$ lacks such a transition, instead resembling more semimetallic behavior.

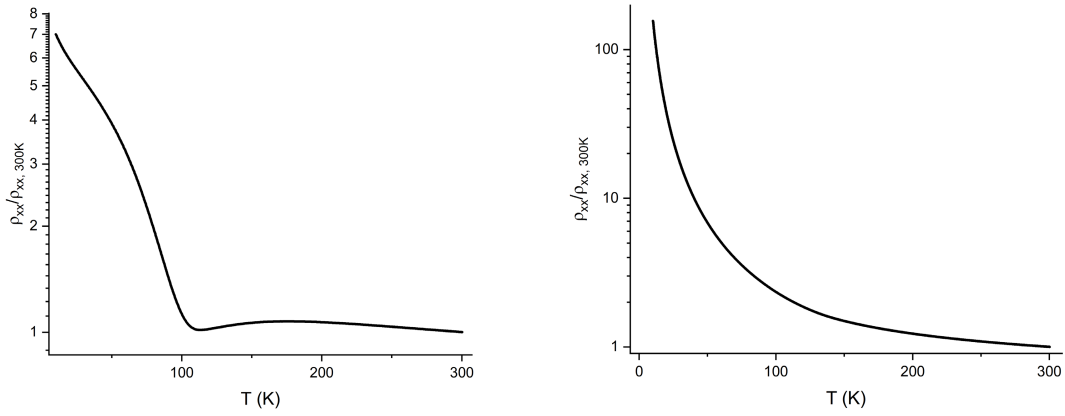


Figure S2: Temperature-dependent resistivity curves for (left) $\text{Eu}_2\text{Ir}_2\text{O}_7$ and (right) $\text{Y}_2\text{Ir}_2\text{O}_7$.

S3 REXS ATS subtraction

The anisotropy tensor susceptibility (ATS) scattering due to local IrO_6 tetrahedra distortions contributes to the $(0\ 0\ 4n+2)$ peaks^{1,2}. With linearly-polarized incident light perpendicular to the scattering plane, a polarization analyzer was used to only collect scattered X-rays in the scattering plane (σ - π channel detected). The analyzers used were pyro-graphite (008) at APS and Al (333) at BNL. This setup was used to suppress the contribution from non-magnetic scattering as much as possible.

To assess and isolate the ATS contribution to the signal, the rocking curves at $T > T_N$ were averaged, as the ATS contribution is expected to be temperature-independent. The averaged rocking curve was then used as the background and was subtracted from all $T < T_N$ rocking curves. Fig. 2 shows the averaged background rocking curves compared to one curve at 103K.

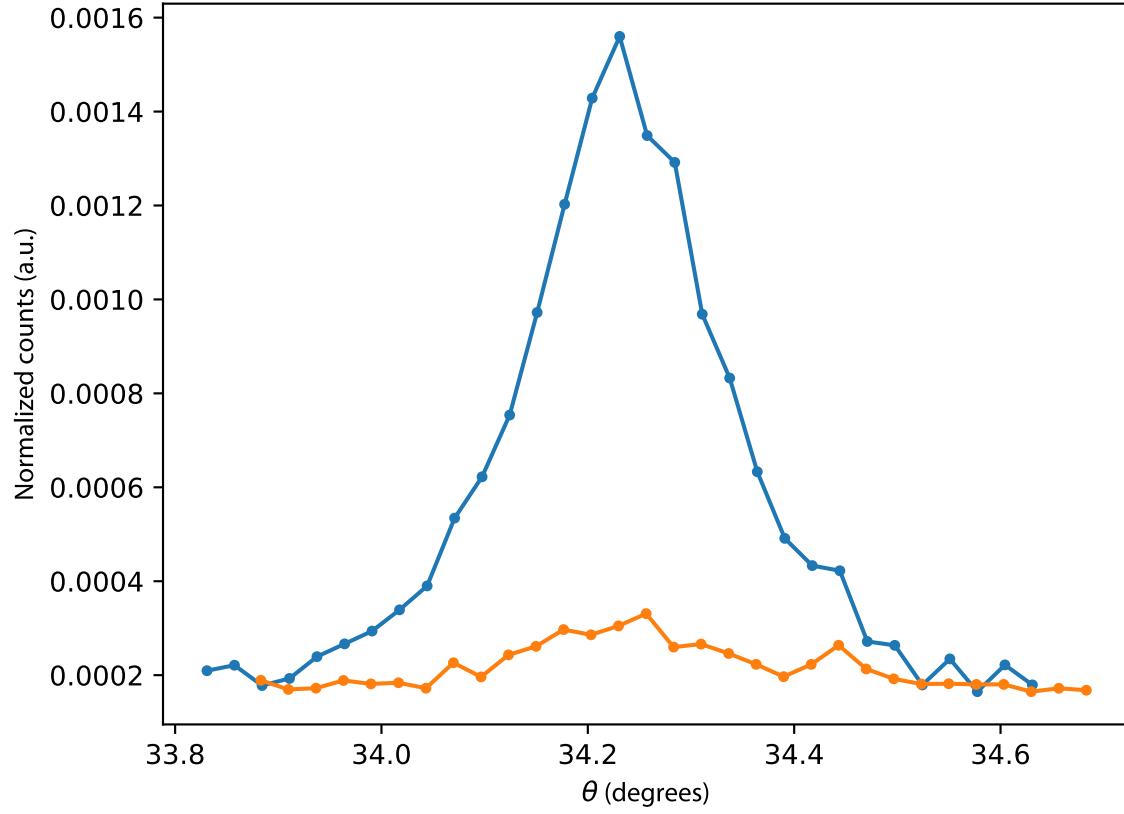


Figure S3: **Rocking scan through the (0 0 10) magnetic Bragg peak.** Orange curve is $T = 103K$ (below T_N), with blue curve (averaged for all $T > T_N$ scans) for comparison.

S4 Fitting of critical exponent β

The REXS scattering intensity was fitted against the reduced temperature using

$$I(T) = I_0 + A(1 - T/T_N)^{2\beta}, \quad (1)$$

with $T_N = 140$ K. As shown in Figure S4, the fitted value of $\beta = 0.30 \pm 0.04$ remains robust up to $(1 - T/T_N)^{0.6} < 0.66$.

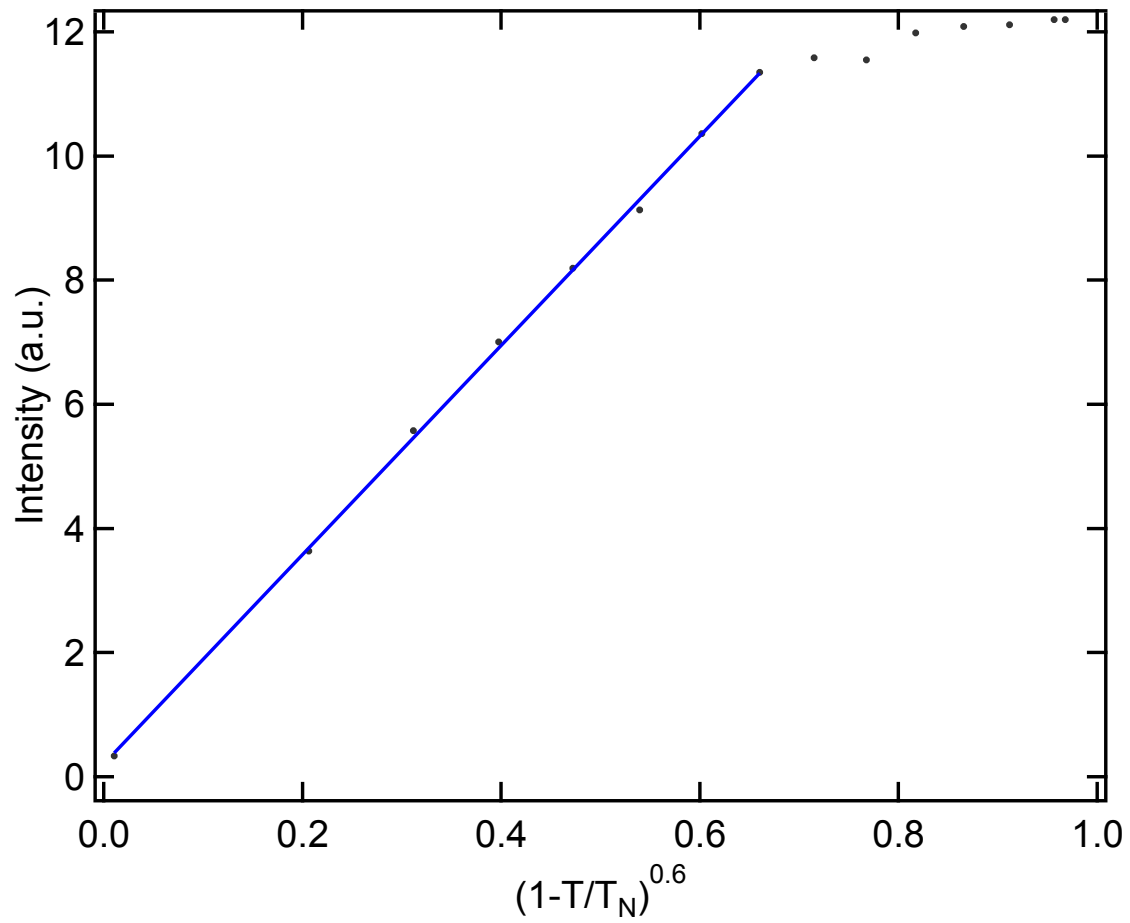


Figure S4: REXS scattering intensity as a function of $(1 - T/T_N)^{0.6}$, corresponding to the value of $\beta = 0.3$.

S5 Geometry of the RIXS experiment

RIXS measurements on thin films are experimentally challenging due to the low volume of material in the film compared to the substrate. For this reason, we employed the following restrictions in finding a magnetic Bragg peak:

- $2\theta \sim 90^\circ$ ³⁻⁷
- Incident angle $\alpha < 1^\circ$
- Bragg peak which is structurally forbidden but magnetically allowed

Both YIO and the underlying YSZ substrate have space group $Fd\bar{3}m$ with the following selection rules for allowed structural Bragg peaks (hkl):

$$h + k = 2n$$

$$h + l, k + l = 2n$$

We collected the RIXS spectra exclusively in the vicinity of the (5 12 -1) structural peak, which was structurally forbidden, so we could therefore isolate the magnetic signal, to reduce the elastic line that usually hinders RIXS measurements of thin films in the hard X-ray regime. This allowed us to map magnon dispersion in pyrochlore iridate thin-films for the first time.

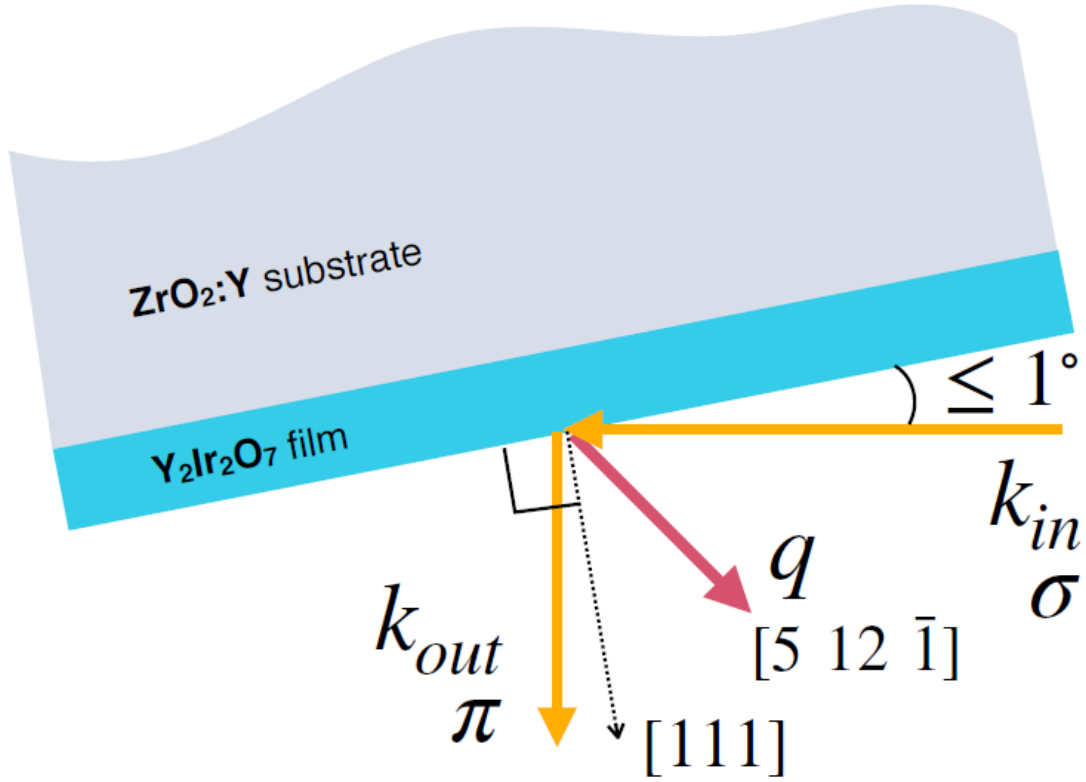


Figure S5: **RIXS experimental geometry.** The setup for the RIXS experiment followed the restrictions of $2\theta \sim 90^\circ$, low incidence angle, and structurally forbidden Bragg peak.

S6 Analysis of the RIXS spectra

To analyze the RIXS spectra, we developed a multi-peak fitting procedure.

The elastic lines were fitted to a Voigt lineshape; we report a FWHM of 28-29 meV which was consistent with the FWHM of the (6 6 6) Bragg peak, giving us a measure of the experimental resolution. The inelastic peaks (low-energy magnon modes and a higher-energy incoherent peak) were fitted to anti-symmetrized Lorentzian line shapes^{8,9} of the form

$$S(\mathbf{Q}, \omega) = \frac{A(\mathbf{Q})\gamma}{2\pi} \left[\frac{1}{(\omega - \omega_0)^2 + (\gamma/2)^2} - \frac{1}{(\omega + \omega_0)^2 + (\gamma/2)^2} \right]$$

where A is the peak amplitude, γ is the full width at half maximum of the peak, and ω_0 is the peak position. The amplitude was set to zero for all negative loss energy values.

The momentum-dependent scans were taken at 10 K, where we were able to track two low-energy magnon modes. The fits are shown in Fig. S6 for RIXS curves taken at (5 12 -1) and (5 11.5 -0.5). We note that the intensity of the elastic line increases as we move away from Γ . This is because the elastic line intensity is very sensitive to the geometric constraints of the experiment (see Section S5), and these conditions became less favorable as we moved to the zone edge.

The temperature-dependent data was all taken at Γ . Due to the increased contribution from elastic line intensity at higher temperatures, it became more difficult to resolve two modes throughout the temperature range. For consistency, only one low-energy magnon mode is fitted for the

temperature-dependent RIXS curves. This choice is justifiable since we are only making qualitative claims about the gap from the temperature-dependent data.

We then developed a subtraction method to observe the presence of the gap as shown in Fig. 3 of the main text. We subtracted both the contributions from the elastic line and the magnetic continuum. Since we observed a temperature-independent behavior of the continuum, we averaged all continuum curves before subtraction.

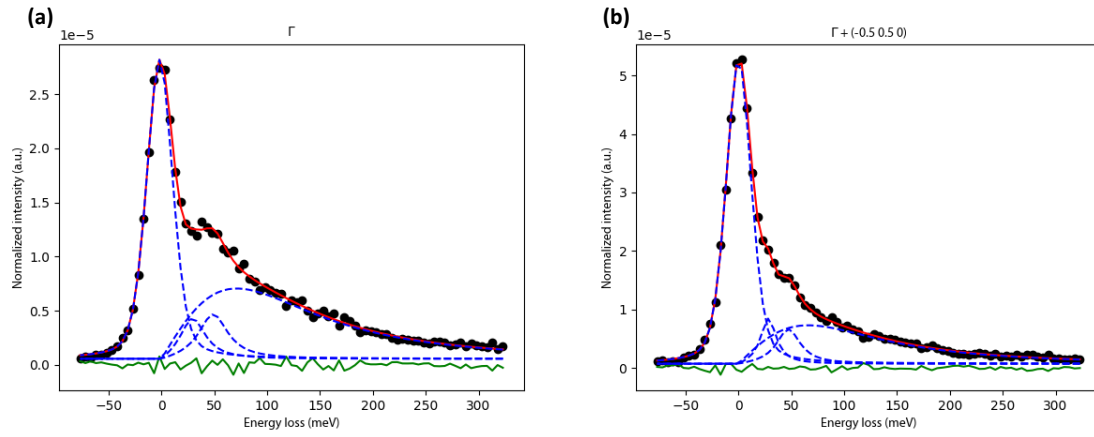


Figure S6: **Fits for select RIXS scans.** Fits displayed for scans taken at (a) Γ and (b) $\Gamma + (-0.5, 0.5, 0)$. All momentum-dependent scans were measured at 10K. Black dots represent raw data. Blue dashed lines show the individual components of the multi-peak fit. Red curve is the total fit, and green curve is the residual.

S7 Details of the spin-wave calculations

We treat our film as a bulk system, following the approach described in Refs. ^{10,11}. In light of the REXS results, the magnetic ground state is assumed to be the 4i-4o order, as illustrated in Fig. 1 of the main text. PyIr systems are commonly modeled with the nearest-neighbor spin Hamiltonian

$$H = \sum_{\langle i,j \rangle} [J \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) + S_i^a \Gamma_{ij}^{ab} S_j^b], \quad (2)$$

where the first term represents antiferromagnetic Heisenberg exchange ($J > 0$), the second represents the antisymmetric Dzyaloshinskii-Moriya (DM) interaction, and the last term represents symmetric anisotropic exchange^{12,13}. The repeated indices $a, b \in \{x, y, z\}$ are summed over. The traceless symmetric anisotropic term was neglected in the main text, but is included here for completeness. As a rule of thumb, it is subleading to the DM interaction as it represents a process of higher order in the spin-orbit coupling. We use a convention where $\mathbf{D}_{ij} = D \hat{d}_{ij}$, where the $\hat{d}_{ij}$ are normalized DM vectors in the direct configuration¹⁴. We note that an alternative convention where the DM strength is instead defined as $D' = |\mathbf{D}_{ij}| = D/\sqrt{2}$ also exists in the literature.

It is convenient to first rewrite the spin Hamiltonian as

$$H = \sum_{\langle i,j \rangle} S_i^a \Lambda_{ij}^{ab} S_j^b. \quad (3)$$

The noncollinear 4i-4o order is accounted for by a sublattice-dependent rotation onto a new coordinate system where the spin quantization axis on each site points along the direction of the local moment, i.e.

$$S_i^a = \left[R_i \left(\tilde{S}_i \right) \right]^a = R_i^{ab} \tilde{S}_i^b, \quad (4)$$

where S_i^a is the spin operator in the global coordinate system and $\tilde{S}_i^a$ is the spin operator in the sublattice-specific coordinate system. Deviations about the ground state are described using the Holstein-Primakoff representation $\tilde{S}_i^z = s - a_i^\dagger a_i$,

$$\tilde{S}_i^+ = \sqrt{2s - a_i^\dagger a_i} a_i, \quad \tilde{S}_i^- = a_i^\dagger \sqrt{2s - a_i^\dagger a_i}, \quad (5)$$

where the spin length $s = 1/2$ for the $J_{\text{eff}} = 1/2$ states and a_i is the bosonic annihilation operator on site i . The four sublattices of the magnetic order results in four distinct bosonic species. The Hamiltonian is then truncated to quadratic order in the boson operators, Fourier transformed into momentum space, and written in matrix form,

$$H_{\mathbf{k}} = \mathbf{X}^\dagger h \mathbf{X}, \quad h(\mathbf{k}) = \begin{bmatrix} A(\mathbf{k}) & B(\mathbf{k}) \\ B^*(-\mathbf{k}) & A^*(-\mathbf{k}) \end{bmatrix} \quad (6)$$

where $\mathbf{X} = \left(a_1(\mathbf{k}), \dots, a_4(\mathbf{k}), a_1^\dagger(-\mathbf{k}), \dots, a_4^\dagger(-\mathbf{k}) \right)^T$ with numerical indices labeling the sublattices. The system is diagonalized “paraunitarily” to preserve the canonical bosonic commutation relations. This is achieved by selecting the positive eigenvalues of $gh(\mathbf{k})$, where

$$g = \begin{pmatrix} I_{4 \times 4} & 0 \\ 0 & -I_{4 \times 4} \end{pmatrix}, \quad (7)$$

and $I_{4 \times 4}$ is the 4×4 identity matrix. Thus theoretical dispersions for four magnon bands are obtained.

These four bands are then fit to the two experimentally extracted magnon peaks (measured relative to the fitted elastic peak) along the Γ –K path (Fig. 3B in the main text). The fit is implemented as a minimization problem in (J, D) for a two-parameter model or (J, D, Γ) for

the three-parameter model, where the quantity being minimized is the reduced χ_{red}^2 between the experimental data and the linear spin-wave theory (LSWT) energies,

$$\chi_{\text{red}}^2 = \frac{\chi^2}{\nu} = \frac{\sum_i \frac{(O_i - C_i)^2}{\sigma_i^2}}{\nu}. \quad (8)$$

where O_i is the i th observed (i.e. experimentally extracted) magnon peak energy, C_i is the corresponding calculated value, and σ_i^2 is the variance, which we approximate with the uncertainty in the position of the experimental magnon peak. This uncertainty is on the order of a few meV. Since there are four calculated but only two experimentally extracted modes, we use the LSWT mode closest in energy as C_i for evaluating χ_{red}^2 . The denominator $\nu = n - m$ is the number of observations n minus the number of fitted parameters m , and thus represents the degrees of freedom.

The best fits obtained for the two- and three-parameter models are

- $J = 33.9468 \text{ meV}$, $D = 5.64612 \text{ meV}$, $\chi_{\text{red}}^2 = 12.9231$,
- $J = 32.6832 \text{ meV}$, $D = 6.31624 \text{ meV}$, $\Gamma = 1.8954$, $\chi_{\text{red}}^2 = 17.2308$.

Both the χ_{red}^2 values and the limited number of experimental data points used in the fits suggest that the two-parameter model should be preferred. The corresponding dispersions are plotted in Fig. S7. We obtain reasonable agreement except at the Γ point, where an outlier data point is found. However, there the theoretically predicted gap is small; both fitted models find magnon modes at $\approx 24 \text{ meV}$ and $\approx 29.5 \text{ meV}$, for a gap of 5.5 meV . We note that the energy resolution in the RIXS

experiment is 29 meV, meaning that gaps between closely situated magnon bands (with gaps below ≈ 10 meV) may fail to be resolved. We also note that a Raman scattering study on a polycrystalline $\text{Y}_2\text{Ir}_2\text{O}_7$ sample likewise found a small gap between two modes argued to be of magnetic origin, with peaks at 28.6 meV and 34.4 meV¹⁵. Higher energy resolution is required to fully determine the magnon band structure at the Γ point. Future experimental work may also collect a larger data volume in reciprocal space, which would permit more comprehensive fits that could take into account additional interactions that are allowed by symmetry, such as further-range exchange. In principle, symmetry also allows for the nearest-neighbor symmetric anisotropic exchange to be parameterized by two independent parameters Γ_1 and Γ_2 ¹⁶. These are found to be proportional to each other in perturbation-theory derivations of the spin Hamiltonian^{12,13}, motivating the use of two- and three-parameter models here, but we note that this relationship can break down in more general models¹⁶. We have attempted to fit our data also to the four-parameter model $(J, D, \Gamma_1, \Gamma_2)$ but did not obtain robust fitting results, presumably due in part to the limited amount of data.

We have performed classical Monte Carlo simulations using the ESpinS software¹⁷ to estimate the ordering temperature of the two-parameter model. We find a T_N of at least 127 K in reasonable agreement with the experimental value; see Figure S8. The T_N value extracted from specific heat grows with system size and could further approach the experimental value of 145 K in the thermodynamic limit. Due to uncertainties with fitting and simulation, as well as the expectation that quantum effects could renormalize the value, we have not carried out a detailed finite-size scaling here. The simulations used clusters with $N = 4 \times L \times L \times L$ sites. Following Ref.¹⁴ 10^6 Monte Carlo steps were used for thermalization, and 2×10^6 steps were used for the sampling.

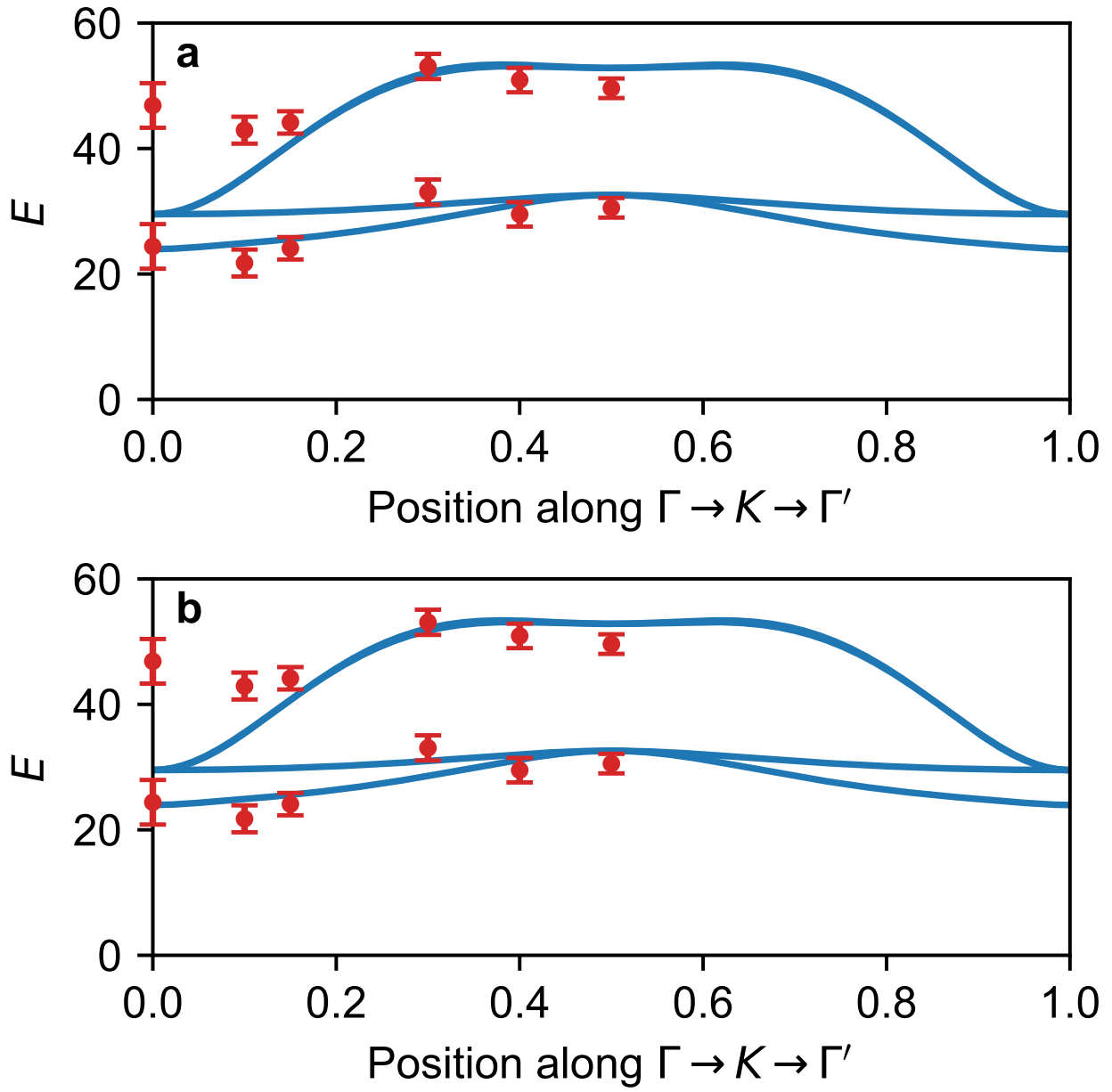


Figure S7: **Spin-wave fits.** **a** Best fitted two-parameter model compared with magnon peaks extracted from the RIXS data. **b** Same for the three-parameter model.

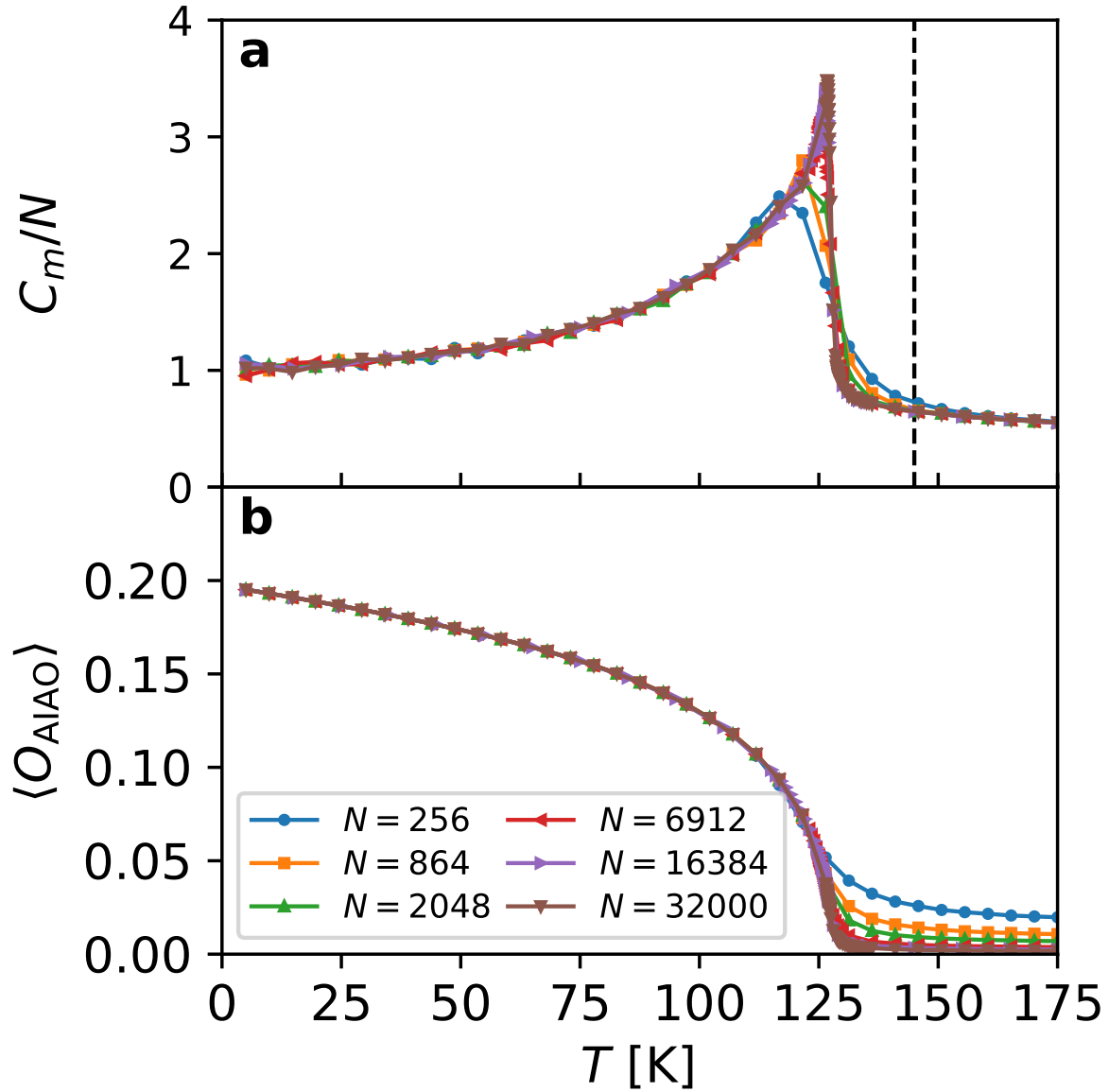


Figure S8: **Classical Monte Carlo simulations.** **a** Magnetic specific heat predicts a transition temperature of at least $T_N \approx 127$ K. The dashed vertical line indicates the experimentally observed transition temperature 145 K. **b** the four-in-four-out order parameter. All calculations were done for the best fitted two-parameter model with $J = 33.9$ meV and $D = 5.6$ meV on clusters with $N = 4L^3$ sites.

S8 The derivation of the spin- and charge-resolved channels of the Lindhard function

Here, we derive the expressions for the spin- and charge-resolved channels of the Lindhard function.

The non-interacting multiorbital itinerant Hamiltonian for YIO, in the presence of spin-orbit coupling (SOC), is given as

$$\mathcal{H}_0 = \sum_{ij} \sum_{ab} \sum_{\sigma\sigma'} (t_{ia\sigma,jb\sigma'} - \mu\delta_{ij}\delta_{ab}) c_{ia\sigma}^\dagger c_{jb\sigma'}, \quad (9)$$

where $t_{ia\sigma,jb\sigma'}$ are hopping matrix elements from the Wannier model derived from DFT+ U calculations including SOC. In our calculation we consider full 24-orbital spin-resolved Wannier model with electrons originating from t_{2g} orbitals of four Ir atoms in the unit cell. The chemical potential μ is fixed to a filling of 20 electrons per unit cell. Here the operator $c_{ia\sigma}^\dagger$ creates an electron at site i in orbital $a \in \{1, \dots, 12\}$ with spin polarization $\sigma \in \{\uparrow, \downarrow\}$.

We consider the orbitally-resolved spin operator

$$S_{ab}^i(\mathbf{q}, \tau) = \frac{1}{2} \sum_{\mathbf{k}} \sum_{\alpha\beta} c_{\mathbf{k}+\mathbf{q}a\alpha}^\dagger(\tau) \sigma_{\alpha\beta}^i c_{\mathbf{k}b\beta}(\tau), \quad (10)$$

and the density operator

$$n_{ab}(\mathbf{q}, \tau) = \frac{1}{2} \sum_{\mathbf{k}} \sum_{\alpha} c_{\mathbf{k}+\mathbf{q}a\alpha}^\dagger(\tau) c_{\mathbf{k}b\alpha}(\tau) \equiv S_{ab}^0(\mathbf{q}, \tau), \quad (11)$$

where σ^i corresponds to the Pauli matrices $i \in \{x, y, z\}$ in eq. (10) and over $i = 0$ in eq. (11), a, b denote orbitals, and α, β denote spin degrees of freedom. The corresponding components of the generalized imaginary-time spin-spin ($i, j \in \{x, y, z\}$) and the density-density ($i = j = 0$)

correlation functions are

$$(\chi_0)_{abcd}^{ij}(\mathbf{q}, i\omega_n) = \int_0^\beta d\tau e^{i\omega_n \tau} \langle T_\tau S_{ad}^i(\mathbf{q}, \tau) S_{bc}^j(-\mathbf{q}, 0) \rangle_0, \quad (12)$$

where T_τ is the time-ordering operator in imaginary time τ and a, b, c, d denote orbitals. The brackets $\langle \cdot \rangle_0$ denote the thermal expectation value with respect to the non-interacting Hamiltonian $\mathcal{H}_0$, and $\beta \equiv 1/T$ is the inverse temperature.

By applying Wick's theorem and summing over Matsubara frequencies, followed by a summation over orbital degrees of freedom, we derive the spin ($i, j \in \{x, y, z\}$) and the charge ($i = j = 0$) channels of the Lindhard response function^{18,19}

$$\chi_0^{ij}(\mathbf{q}, i\omega_n) = \frac{1}{N} \sum_{\mathbf{k}} \sum_{mn} \sum_{ab} \mathcal{M}_{abba}^{ij,mn}(\mathbf{k}, \mathbf{q}) \frac{n_F(\varepsilon_{\mathbf{k}}^m) - n_F(\varepsilon_{\mathbf{k}+\mathbf{q}}^n)}{\varepsilon_{\mathbf{k}+\mathbf{q}}^n - \varepsilon_{\mathbf{k}}^m - i\omega_n}. \quad (13)$$

Here, N is the total number of $\mathbf{k}$ -points considered for the discretization of the reciprocal primitive unit cell, m, n index energy bands $\varepsilon_{\mathbf{k}}^n$ of the tight-binding Hamiltonian with respect to the Fermi level, and $n_F(\varepsilon_{\mathbf{k}}^n) = 1/[e^{\varepsilon_{\mathbf{k}}^n/T} + 1]$ is the Fermi-Dirac distribution function at temperature T . In general, tensor $\mathcal{M}_{abcd}^{ij,mn}(\mathbf{k}, \mathbf{q})$ contains information about the Wannier orbitals via the eigenfunctions of the tight-binding Hamiltonian when transformed from the original orbital to band basis

$$\mathcal{M}_{abcd}^{ij,mn}(\mathbf{k}, \mathbf{q}) = \frac{1}{4} \sum_{\alpha\beta\gamma\delta} \sigma_{\alpha\delta}^i \sigma_{\beta\gamma}^j \left(u_{a\alpha}^n(\mathbf{k} + \mathbf{q})^* u_{b\beta}^m(\mathbf{k})^* u_{c\gamma}^n(\mathbf{k} + \mathbf{q}) u_{d\delta}^m(\mathbf{k}) \right), \quad (14)$$

where $u_{a\alpha}^n(\mathbf{k})$ is the n th eigenstate of the tight-binding Hamiltonian at momentum $\mathbf{k}$ whose eigenenergy is $\varepsilon_{\mathbf{k}}^n$.

The physical susceptibility channels given in eq. (13) are obtained by performing a trace over

the orbital indices, considering that the spin operators given in eq. (10) are localized on a single orbital $S_a^i(\mathbf{q}, \tau) = \sum_b \delta_{ab} S_{ab}^i(\mathbf{q}, \tau)$.

Static and dynamical spin- and charge- susceptibility channels from eq. (13) are obtained by performing analytic continuation $i\omega_n \rightarrow \omega + i\eta$, considering a small smearing parameter $\eta > 0$.

We distinguish the longitudinal $\chi_0^{\parallel}(\mathbf{q}, \omega) \equiv \chi_0^{zz}(\mathbf{q}, \omega)$ and the transverse bare spin susceptibility $\chi_0^{\perp}(\mathbf{q}, \omega) \equiv \chi_0^{xx}(\mathbf{q}, \omega) + \chi_0^{yy}(\mathbf{q}, \omega)$.

In the calculation of the longitudinal spin susceptibility, the orbital factor given in eq. (14) becomes

$$\mathcal{M}_{abcd}^{\parallel, mn}(\mathbf{k}, \mathbf{q}) = \frac{1}{4} \sum_{\sigma\sigma'} \text{sgn}(\sigma\sigma') u_{a\sigma}^n(\mathbf{k} + \mathbf{q})^* u_{b\sigma'}^m(\mathbf{k})^* u_{c\sigma'}^n(\mathbf{k} + \mathbf{q}) u_{d\sigma}^m(\mathbf{k}). \quad (15)$$

The transverse spin susceptibility can be rewritten in terms of the raising and lowering spin operators via $\sigma^{\pm} = \frac{1}{2}(\sigma_x \pm i\sigma_y)$, so the summation in eq. (14) simplifies to

$$\mathcal{M}_{abcd}^{\perp, mn}(\mathbf{k}, \mathbf{q}) = \frac{1}{2} \sum_{\sigma} u_{a\sigma}^n(\mathbf{k} + \mathbf{q})^* u_{b\bar{\sigma}}^m(\mathbf{k})^* u_{c\sigma}^n(\mathbf{k} + \mathbf{q}) u_{d\bar{\sigma}}^m(\mathbf{k}), \quad (16)$$

using notation $\bar{\sigma} = -\sigma$.

For the charge susceptibility $\chi_0^C(\mathbf{q}, \omega) \equiv \chi_0^{00}(\mathbf{q}, \omega)$, the orbital-dependent factor given in eq. (14) becomes

$$\mathcal{M}_{abcd}^{C, mn}(\mathbf{k}, \mathbf{q}) = \frac{1}{4} \sum_{\sigma\sigma'} u_{a\sigma}^n(\mathbf{k} + \mathbf{q})^* u_{b\sigma'}^m(\mathbf{k})^* u_{c\sigma'}^n(\mathbf{k} + \mathbf{q}) u_{d\sigma}^m(\mathbf{k}). \quad (17)$$

S9 Investigating the connection to Weyl semimetal

Below, we investigate the potential role of Weyl fermions in stabilizing the magnetic fluctuations in YIO. A mechanism of Weyl fermions coupling to other magnetic degrees of freedom was proposed theoretically^{20,21}, and reported experimentally^{22,23}. A recent experimental study of magnetic WSM CeAlGe suggests that Weyl fermions coupled to magnetic domains stabilize magnetic fluctuations above the long-range ordering temperature²⁴. This condition is met when the magnetic ordering wavevector is close to the Weyl point nesting vector. From our DFT+ U calculations with $U = 0.85$ eV, we predict a Weyl point nesting vector of 0.08 r.l.u. in the Weyl semimetal phase (see Fig. S11a inset for the locations of the Weyl nodes), in close proximity to the $\mathbf{q} = (0, 0, 0)$ magnetic order as established through our REXS measurements. Our experimental finding – the persistence of strong spin fluctuations above the Néel temperature in the WSM YIO – suggests the survival of short-range magnetic order with the ordering vector close to the Weyl nesting vector, so the proposed scenario in²⁴ could play out in the new pyrochlore iridate class of magnetic topological materials. To test this hypothesis, we study the paramagnetic states of both Weyl semimetal and topological insulator phases. In Fig. 4c, shown is the imaginary part of the integrated intensity of the dynamical bare transverse spin susceptibility around Γ . The lower-energy peak that corresponds to the contribution from paramagnons shows the same intensity in all models considered, suggesting that the strong low-energy magnetic fluctuations survive also in the topologically trivial regime. Furthermore, a comparison of the electronic density-of-states (DOS) in all considered models in the PM phase reveals the robust peak around the Fermi energy. The existence of this peak suggests the itinerant origin of the observed paramagnons in the system. The presence of this

peak, however, also suggests the presence of many itinerant states that are available to potentially couple with Weyl nodes. The potential interplay between the itinerant and Weyl fermions in YIO is a natural direction for future research.

S10 The study of the origin of the low-energy and high-energy peaks from the spin- and charge- susceptibility channels

Here we show the momentum- and frequency-resolved dynamical bare transverse spin and charge susceptibility referred to in the main text. The comparison of the spin- and charge- channels from the Lindhard function calculations uncovers the origin of different low-energy excitations observed in the paramagnetic state in YIO.

In the main text in Fig. 4(b) we show the integrated intensity of the imaginary part of the dynamical bare transverse spin susceptibility that features the low-energy (~ 25 meV) and the high-energy ($\sim 70 - 90$ meV) peaks in all models considered. As a complementary calculation, in Fig. S9 we focus on the tight-binding model obtained for $U = 0.85$ eV and show the momentum-resolved imaginary part of the dynamical bare transverse spin susceptibility Fig. S9(a), as well as the charge susceptibility channel Fig. S9(b). The results are obtained considering full 24-orbital models, where the different channels are calculated as given in eqs. (16) and (17). The calculations in Fig. S9(a-b) are obtained for $k_B T = 0.015$ eV, with the k-point grid discretization $25 \times 25 \times 25$ and broadening $\eta = 3$ meV.

As for the origin of the stronger low-energy peak observed in the integrated intensity in Fig. 4(b), we associate it with the sharp paramagnon-like excitations that propagate from the Γ point. These excitations are seen in both the transverse spin- and the charge-susceptibility channels in Fig. S9(a-b). In order to better resolve the gap at zone center and the number of branches along Γ -K direction, we focus on the low-energy window and map out the excitations considering

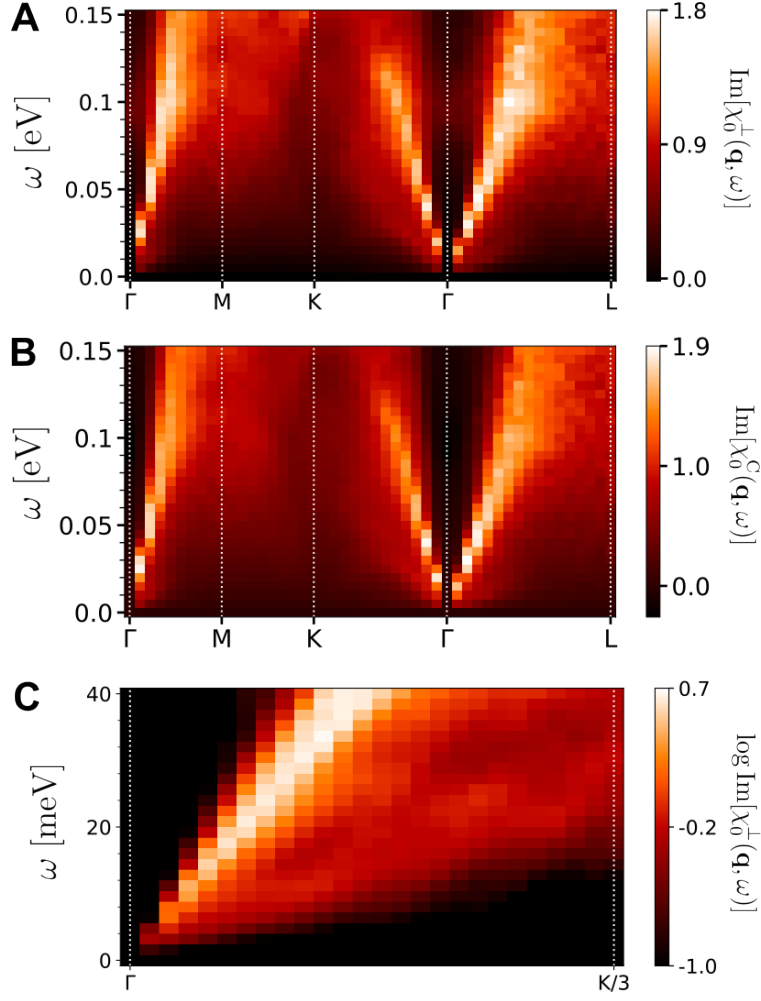


Figure S9: **Dynamical bare transverse susceptibilities.** (a-b) The imaginary part of the dynamical bare transverse spin (a) and charge (b) susceptibility channels from the Lindhard function calculation for the model at $U = 0.85$ eV. (c) Low-energy window of the dynamical bare transverse spin susceptibility along Γ -K direction around the zone center for the model at $U = 0.85$ eV. The values of parameters used for calculations in all panels are given in the text.

finer momentum- and ω -grids, and a smaller broadening. The obtained results with the color bar shown on the log scale, are given in Fig. S9(c) (the same data is also shown in the main text in Fig. 4(c)). These calculations are performed at the temperature $k_B T = 0.015$ eV, with the $\mathbf{k}$ -grid discretization $30 \times 30 \times 30$ and the smaller broadening $\eta = 2$ meV. Within our itinerant model, we do not reproduce the experimentally observed value of the gap of magnetic excitations. We ascribe this difference to the pronounced Fermi surface nesting near the BZ center and the magnetic anisotropy that arises from spin-orbit coupling (SOC) in our itinerant model being smaller than its experimental value. As the magnetic anisotropy in our itinerant model arises from SOC, we argue its smaller than expected value due to the high-symmetry cubic space group and large sensitivity of the value of magnetic anisotropy to the small structural distortions of the lattice. As for the number of the branches along $\Gamma - K$ direction, besides the stronger upper branch, we also identify an additional weaker lower branch propagating with a smaller velocity, which is in the agreement with the experimental finding.

Finally, we briefly focus on the origin of the high-energy peak. We find that the transverse spin channel shows the incoherent continuum bands around Γ at energies ~ 100 meV. On the other hand, the absence of the high-energy peak in the charge susceptibility channel indicates the magnetic origin of these excitations.

S11 Fermi surface nesting near the zone center

We also identify strong Fermi surface nesting near the center of the BZ²⁵ that characterizes the paramagnetic phase, as shown in Fig. S10, visualized using FermiSurfer²⁶. From Fig. S10(b), we can see that the PM phases given by the three values of U , regardless of whether they give a topological or trivial 4i-4o phase or not, always show a cube-like pocket at the BZ center, of size about $2/9$ of the linear scale of the BZ along Γ -X direction. The observed pronounced nesting from the cube-like pocket would also contribute to huge peaks in the real part of the Lindhard response function at $\mathbf{q} \approx \mathbf{0}$ we obtain from calculations and consequently could explain the appearance excitations that appear to propagate from Γ . We expect the role of this pronounced nesting will be renormalized in the full calculation including interactions.

Here, we note that the so-called paramagnetic (PM) state that we accessed using DFT+ U is not a real paramagnetic state but a mean-field approximation, and the same applies to the 4i-4o low-energy state. To obtain PM state, we initialized the magnetic configuration at each site to be exactly 0, with noncollinear magnetic self-consistent field calculations converging to a zero-magnetic-moment final state without on-site spin fluctuations. A more appropriate simulation of a true paramagnetic state involves random sampling of the spin configurations at large supercells, which is beyond the scope of our calculations.

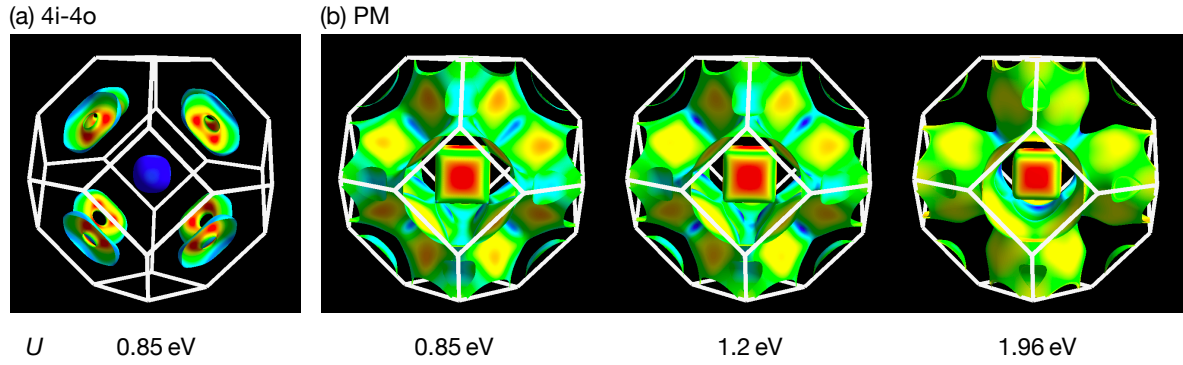


Figure S10: **Fermi surfaces.** The Fermi surfaces of the bulk YIO for (a) the 4i-4o state and (b) the paramagnetic (PM) state as given by the DFT+ U calculations, using U values of 0.85 eV, 1.2 eV, and 1.96 eV. The 4i-4o state of 1.2 and 1.96 eV are insulating, so no Fermi surface is available. Fermi surface nesting is visible near the center of the BZ for the paramagnetic states for all values of U , regardless of whether the corresponding 4i-4o state is topologically non-trivial (0.85 eV) or trivial (1.2 and 1.96 eV).

S12 Details of the DFT+ U calculations

To obtain the electronic structure of YIO, we performed the first principles density functional calculations implemented in the Vienna ab initio simulation package^{27,28} with an exchange-correlation PBE functional²⁹ and an on-site U applied to the Ir d orbitals, with non-collinear spin configurations and spin-orbit coupling included. A $9 \times 9 \times 9$ Monkhorst-Pack k grid was used. We have identified the range of $0.75 \text{ eV} \leq U \leq 0.9 \text{ eV}$ to be the range where the ground state appears as a 4i-4o Weyl semimetal (still with small electron and hole pockets near the Fermi level), below which the system is a Weyl semimetal (with small electron and hole pockets near the Fermi level), and above which the system is gapped. Notably, we estimated the value of U to be 1.96 eV for the Ir d orbitals using the linear response theory within DFT+ U ³⁰, where YIO's ground state appears as a 4i-4o insulator with a 0.38 eV gap.

We then computed the spectral function (see Fig. S11) using WANNIERTOOLS³¹ for a 41-layer [111]-slab, with the top (bottom) terminated surfaces as kagome (triangle) Ir atomic planes, as described by a tight-binding Hamiltonian downfolded from the DFT+ U calculations to the Ir t_{2g} orbitals using WANNIER90³². The Weyl nodes are identified near the M point (see the inset of Fig. S11a for the locations of the Weyl nodes). For a non-magnetic approximation of the paramagnetic phases (see spectral function shown in Fig. S11c, d), the magnetic configurations were initialized at each site to be exactly 0, with noncollinear magnetic self-consistent field calculations converging to a zero-magnetic-moment final state without on-site spin fluctuations. We stress that to nail down definitively the exact ground state and the finite-temperature properties of YIO,

further studies using methods beyond DFT+ U , which was already available in literatures^{33–35}, are needed.

Here, we provide an estimate of the Hubbard U value of the Ir t_{2g} manifold using linear response theory based on the method described in Ref. ^{36,37}. We compute the changes of the occupation numbers n_d in the t_{2g} manifold as we vary the U value slightly in a spin-polarized PBE+ U calculation. Figure S12 shows the occupation n_d versus the U in a self-consistent field (SCF) and a non-self-consistent field (NSCF) calculations where the ground state charge density of $U = 0.0$ was used, respectively. We then fit lines to these data, since

$$\begin{aligned}\chi &= \frac{\partial n_d^{\text{SCF}}}{\partial U}, \\ \chi_0 &= \frac{\partial n_d^{\text{NSCF}}}{\partial U}.\end{aligned}\tag{18}$$

Note that the χ and χ_0 are the slopes of fitted lines, which are $\chi = 0.356 \text{ eV}^{-1}$ and $\chi_0 = 1.180 \text{ eV}^{-1}$. Then the Hubbard U can be estimated as

$$U = \chi^{-1} - \chi_0^{-1} = 1.96 \text{ eV}.\tag{19}$$

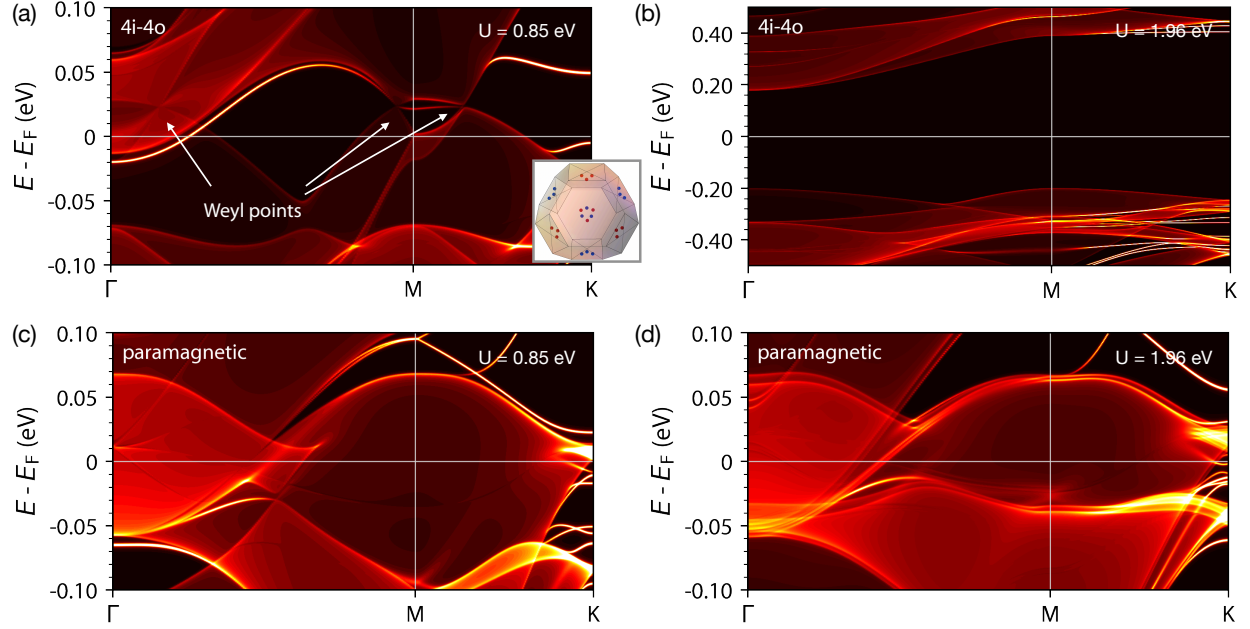


Figure S11: **Spectral function of YIO along the high-symmetry k-path of a (111)-slab.** Spectral functions computed from the tight-binding model downfolded from DFT+ U calculations, with (a) $U = 0.85$ eV, 4i-4o phase, (b) $U = 1.96$ eV, 4i-4o phase, (c) $U = 0.85$ eV, paramagnetic phase, and (d) $U = 1.96$ eV, paramagnetic phase. The $U = 0.85$ eV self-consistent field calculations converge to a 4i-4o Weyl semimetal with small electron and hole pockets near the Fermi level, while $U = 1.96$ eV gives a 4i-4o insulating phase with a gap size of 0.38 eV. Both paramagnetic phases yield higher SCF total energy compared with the corresponding 4i-4o phases and appear to be metallic.

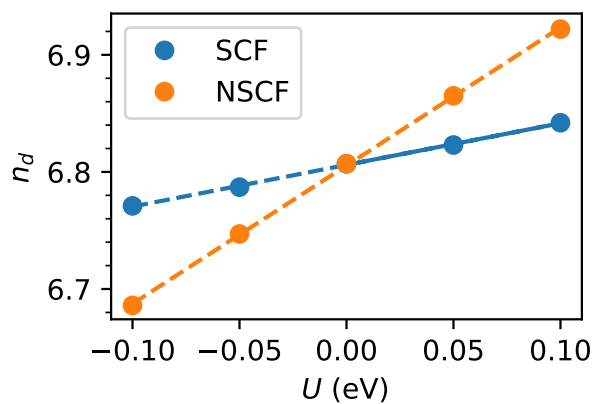


Figure S12: **Occupancy n_d as a function of U .** The occupation on Ir t_{2g} manifold as we change the U value away from 0.0 in a spin-polarized PBE+ U calculation in a self-consistent field (SCF) and non-self-consistent field (NSCF) calculation, where the ground state charge density of $U = 0.0$ was used.

S13 DFT+eDMFT electronic structure of the paramagnetic phase

In this section, we present the results of the electronic structure calculations obtained using DFT+embedded dynamical mean-field theory (eDMFT) for the paramagnetic phase of YIO. The DFT part of the calculation is performed using the WIEN2K code³⁸, using an all-electron LAPW basis set and a PBE exchange-correlation functional on a Monkhorst-Pack mesh of 11x11x11 k points. We have thoroughly checked that the electronic structures obtained by the VASP and WIEN2K agree when $U=0$. The eDMFT calculations^{39–41} start with projecting to a set of real-space orbitals that span the correlated subspace, and the bath orbitals are treated by DFT. The hybridization between the impurity and the bath is then self-consistently determined by solving the DMFT equations. The projector is chosen to be the atomic-like t_{2g} orbitals of one isolated Ir atom. A window of 20 eV around the Fermi level is used for computing the hybridization between the impurity and the bath, and during the self-consistency loops, continuous time quantum Monte Carlo^{42,43} was performed to solve the quantum impurity until the charge density, the impurity self-energy, and the lattice Green's function are converged. The double-counting correction is computed using the nominal double-counting scheme^{39,43}. Then the maximum entropy method⁴⁴ is used to perform the analytical continuation of the self-energy to real frequencies.

Figure S13 shows a comparison between the electronic structures of bulk YIO computed using DFT+eDMFT (a-b, paramagnetic state) and DFT+ U (c-d, zero local spin magnetic moments are assumed). We notice that eDMFT captures the metal-insulator transition of the system around $U = 5.0$ eV. In contrast, when using DFT+ U , the system remains metallic even with large U . This

is because in our DFT+ U calculations, we have constrained the local spin magnetic moments to be zero. The on-site spin fluctuations would only be accessible if the disordered local moments approach⁴⁵ were used. We highlight the similar two-peak feature in the DOS that appears at the Fermi level in the metallic regime from both DFT+ U and DMFT results, suggesting that one would expect to see a similar behavior in their transverse spin susceptibilities.

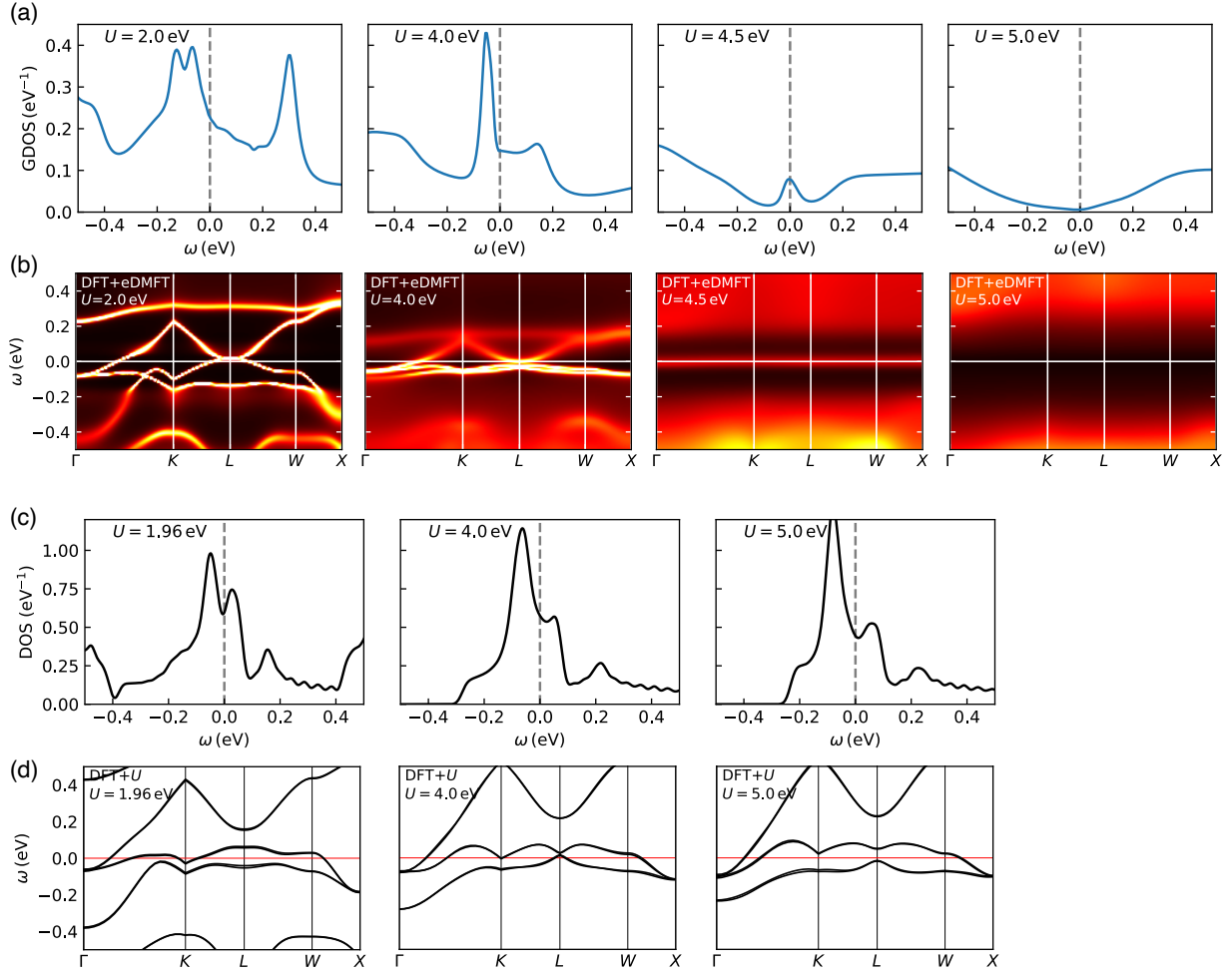


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A comparison between the electronic structures of bulk YIO obtained using DFT+ U (zero local spin magnetic moments) and DFT+eDMFT (paramagnetic state) (a) The global density of states (GDOS) and (b) the spectral functions of the paramagnetic state computed using DFT+eDMFT, with $U = 2.0, 4.0, 4.5$ eV, $J = 0.8$ eV, and $\beta = 50$ meV. (c) The density of states (DOS) and (d) the band structures as given by DFT+ U , with $U = 1.96, 4.0, 5.0$ eV. See the details of the DMFT calculations in the text. Note that for DFT+eDMFT, larger U significantly decreases the bandwidth of the $J_{\text{eff}} = 1/2$ bands, and eventually brings the system from metallic to insulating state around 5 eV. This is not captured with our calculations using DFT+ U where the zero on-site spin magnetic moments are assumed.

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