

Proximate spin liquid and fractionalization in the triangular antiferromagnet KYbSe₂

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I. LOW TEMPERATURE MAGNETIC ORDER

Diffuse scattering Hamiltonian fits predicted that KYbSe₂ is within the 120° ordered phase, so that it should order magnetically at the lowest temperatures. To test this, we measured the specific heat of KYbSe₂ using quasi-adiabatic method in a dilution refrigerator. Heater was mounted on one side of the sapphire stage, with one large single sample (1.19 mg) mounted on the other side with GE varnish. Ruthenium oxide resistance thermometer was glued on the top of the samples. A heat pulse is delivered to heat capacity stage, and the temperature of the thermometer is measured as a function of time. The results are shown in Fig. 1(a). A clear kink is visible at 290 mK consistent with an ordering transition.

To evaluate any possible sample-dependent properties, we remeasured heat capacity of a collection of 11 small pieces (2.33 mg) [see the grey data in Fig. 1(a)], which shows a somewhat broadened transition. Neither sample had a measurable amount of crystalline disorder, suggesting that an appreciable amount of crystalline disorder may suppress the transition entirely.

More clear evidence of static magnetism comes from the upturn at the lowest temperatures. We fit this with a nuclear Schottky anomaly using the hyperfine parameters for Yb³⁺ in Ref. [1]. As shown by the colored lines in Fig. 1(a), the Yb moment size is well-constrained by the data, giving a fitted $\mu = (0.579 \pm 0.010) \mu_B$. This method of determining the magnetic moment has the advantage of (a) being a local probe and insensitive to the particular type of magnetic order, and (b) is a fit extrapolated to zero temperature, and thus is an estimate of the $T = 0$ ordered moment.

We also measured the neutron diffraction of KYbSe₂ using the CTAX spectrometer at the Oak Ridge National Laboratory HFIR reactor, using the same sample and sample mount as used in the CNCS experiment, but mounted in a dilution refrigerator. We measured with $E_i = E_f = 4.8$ meV neutrons focusing on the $Q = (1/3, 1/3, 0)$ point, the wavevector associated with 120° order. This data is shown in Fig. 1(b), and shows a clear onset of elastic scattering at around 300 mK. Panel (c) shows cuts along the $(hh0)$ direction at several different temperatures, showing the emergence of the Bragg intensity. The steepest part of the order parameter curve is at 290 mK, confirming that the bump observed in heat capacity is indeed the transition to 120° magnetic order. Additionally, the agreement between the heat capacity and neutron order parameter curves shows that the sample mount used for the neutron experiments provides adequate thermal equilibration down to at least 290 mK. In summary, both heat capacity and neutron diffraction confirm that the KYbSe₂ J_2/J_1 ratio is indeed within the 120° ordered phase.

II. CRYSTAL CHARACTERIZATION

To investigate the quality of the KYbSe₂ crystals used in this experiment, we measured the single crystal X-ray diffraction using a Bruker Quest D8 single-crystal X-ray diffractometer. The data were collected at room temperature utilizing a Mo

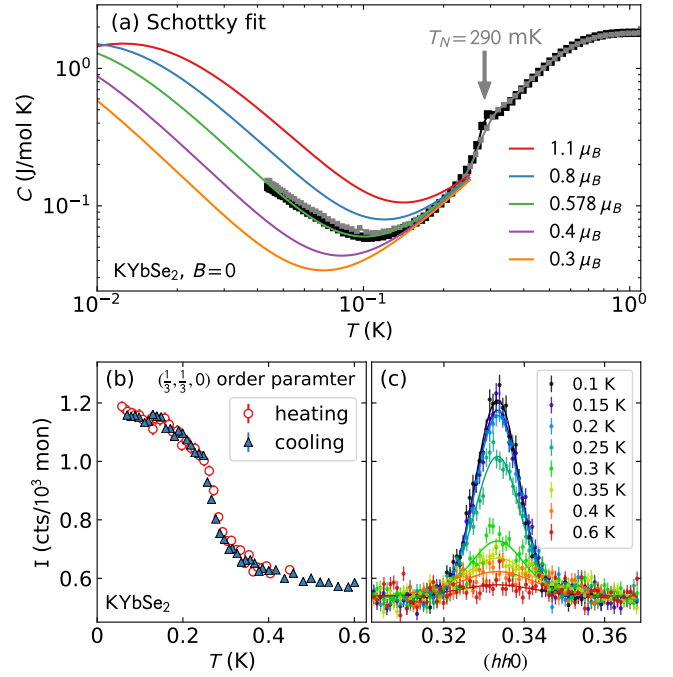


FIG. 1. Evidence of magnetic order in KYbSe₂. (a) Zero field heat capacity of a large (black) and small (grey) sample, showing a kink at 290 mK. An upturn at low temperatures is fitted with a nuclear Schottky anomaly, which uniquely constrains the local ordered Yb³⁺ moment as shown by the colored curves. (b) Neutron scattering at the $(1/3, 1/3, 0)$ Bragg peak as a function of temperature, showing a discontinuity at 290 mK. (c) Rocking curves Error bars indicate one standard deviation uncertainty.

K α radiation, $\lambda = 0.71073$. The crystal diffraction images were collected using ϕ and ω -scans. The diffractometer was equipped with an Incoatec I μ S source using the APEXIII software suite for data setup, collection, and processing [2]. The structure was resolved using intrinsic phasing and full-matrix least square methods with refinement on F². Structure refinements were performed using the SHELXTL software suite [3]. All atoms were first refined with isotropic displacement parameters and then they were refined anisotropically. The final refinement was confirmed with *CheckCif* [4]. A refinement with no site mixing fits the data extremely well.

If we allow site mixing between K and Yb, similar to what was observed in NaYbSe₂ [5], the K-Yb site mixing in KYbSe₂ refines to $(0.2 \pm 0.3)\%$, where the error bar indicates one standard deviation uncertainty as calculated by reduced χ^2 contour. This is a full order of magnitude less site mixing than the 3% site disorder found in NaYbSe₂ [5]. If we force the refined model to have a site-mixing greater than 1%, as shown in Fig. 2, we find a worse *R*-value and a visibly worse fit. Thus, to within uncertainty, KYbSe₂ has no K-Yb site mixing and can be considered as an ideal 2D triangular lattice.

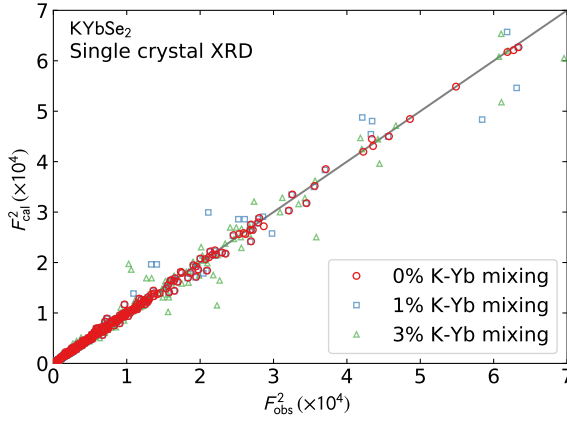


FIG. 2. Single crystal KYbSe₂ X-ray diffraction, showing the observed peak intensities versus the peak intensities of a calculated model with no site mixing or disorder (red circles), 1% Yb-K site mixing (blue squares), and 3% Yb-K site mixing (green triangles). The 0% site mixing visibly fits the data the best, indicating very high crystal quality.

III. ENTANGLEMENT WITNESSES

A. One tangle

One-tangle is calculated from the nuclear Schottky anomaly as explained in the main text. Dividing the total static moment by the in-plane g -tensor (as appropriate for 120° order) gives a measure of the static spin: $\langle S \rangle = \langle \mu \rangle / g_{xx} = 0.193 \pm 0.013$, giving a one-tangle $\tau_1 = 1 - 4\langle S \rangle^2 = 0.85 \pm 0.02$.

As noted in the main text, the static moment from nuclear Schottky fits indicates a moment 39(2) % of the maximal static moment. This is close to the fully static moment for the 2D pure J_1 model predicted by various theoretical techniques: 41% from quantum Monte Carlo [6], 41% from DMRG [7], 38% from series expansion [8], and 44% from Schwinger Boson theory [9]. Experimentally, inter-plane and anisotropic exchange (such as are unavoidable in real materials) would tend to increase the static moment relative to the pure 2D isotropic case. The fact that KYbSe₂ orders at finite temperature is evidence of these effects. However, the addition of J_2 would tend to decrease the static moment relative to the 2D isotropic case. The fact that KYbSe₂, despite its 3D and weak anisotropic interactions, has a static moment slightly below the theoretical value for the pure 2D isotropic case is evidence that the J_2 is destabilizing the magnetic order. This concurs with our conclusion that KYbSe₂ is a proximate QSL.

B. Two tangle

We calculate Two-tangle from the real-space spin correlations, which we obtain from the Fourier transform of the energy-integrated $(hk0)$ plane scattering. For an isotropic $S = 1/2$ system (which KYbSe₂ is to a good approximation—

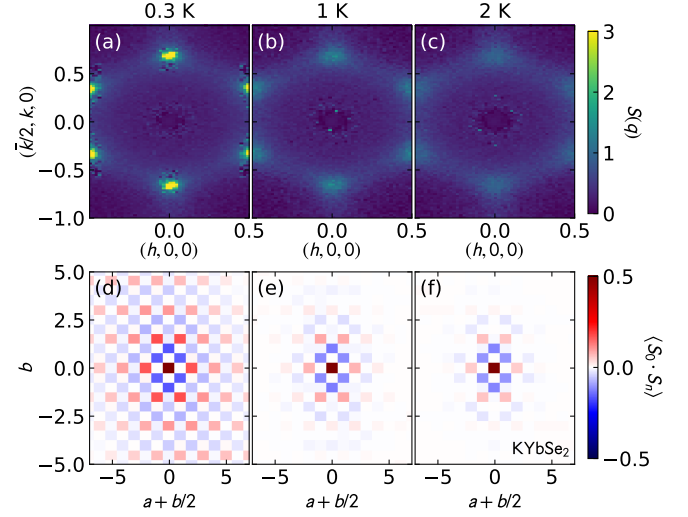


FIG. 3. Energy-integrated KYbSe₂ scattering (a)-(c) Fourier transformed to obtain the static spin-spin correlation in real space (d)-(f). Red indicates ferromagnetic correlation, blue indicates antiferromagnetic correlation. Quasi-long-range order is visible at 0.3 K, but higher temperatures show a much shorter correlation length.

see Onsager reaction field fits), the two-tangle is defined as

$$\tau_2 = 8 \sum_{r \neq 0} \left(\max \left\{ 0, 2|g_r^{zz}| - \left| \frac{1}{4} + g_r^{zz} \right| \right\} \right)^2, \quad (\text{S.1})$$

where $g_r^{zz} = \langle S_i^z S_{i+r}^z \rangle$ [10]. As eq. S.1 shows, g_r^{zz} must exceed the classical threshold of 1/4 for two-tangle to be nonzero. As is shown in main text Fig. 4(b), none of the first four neighbor distances exceed this threshold, and thus two-tangle is zero for all temperatures in KYbSe₂.

We obtained the real space correlations in main text Fig. 4 for the two-tangle by taking Fourier transform of the energy-integrated data in the $(hk0)$ plane. To do this, we cut out a section of reciprocal space from $-0.5 < h < 0.5$ and $-1 < k < 1$. Data were corrected for the Yb³⁺ form factor and background subtracted as described above. Empty data near $Q = 0$ was filled in with data from the next Brillouin zones, and then the data were integrated over all energies, yielding the 2D slices shown in Fig. 3.

We then took the 2D Fourier transform of the data to obtain the real-space spin-spin correlation function, which is shown in the bottom row of Fig. 3. Note that the correlations noticeably decrease as temperature increases, although they retain an overall antiferromagnetic correlation to the nearest neighbors. The average neighbor correlations for the two-tangle calculation in main text Fig. 4 were extracted from these plots.

The two-tangle is dependent upon the background subtraction scheme, and the uncertainty from this is difficult to estimate. We re-ran the analysis with no inelastic background subtraction whatsoever, and found a 0.3 K nearest neighbor two-tangle of -0.082 instead of the background subtracted value of -0.159. As a “worst-case” estimate of uncertainty, it indicates a 48% error bar—which still leaves the nearest neighbor value below the threshold for witnessed bipartite

entanglement. (This is most certainly an overestimate, as it leaves a fair amount of background and artifacts in the data near $Q = 0$.) Thus, the two-tangle is most certainly zero for KYbSe_2 .

C. Quantum Fisher Information

Quantum Fisher Information can be calculated from the neutron spectrum by an integral over energy. When normalized by spin length, QFI is defined as

$$\text{nQFI} = \frac{1}{3\pi S^2} \int_0^\infty d(\hbar\omega) \tanh\left(\frac{\hbar\omega}{2k_B T}\right) \chi''(\hbar\omega, T), \quad [11, 12] \quad (\text{S.2})$$

where $\chi''(\mathbf{q}, \omega)$ is obtained via the fluctuation dissipation theorem $\chi''(\omega) = \pi(1 - e^{-\hbar\omega\beta})S(\omega)$ [13]. In our case, because the correlations are 120° , we calculate the nQFI at $\mathbf{q} = (1/3, 1/3)$ as shown in main text Fig. 4(b). The magnetic excitations at $\mathbf{q} = (1/3, 1/3)$ are gapless to within $40 \mu\text{eV}$ and the QFI integral begins to diverge as temperature decreases.

If we calculate nQFI from Schwinger boson theory and DMRG, we get $\text{nQFI} = 5.049$ for Schwinger Bosons and $\text{nQFI} = 0.107$ for DMRG (both assuming $T = 0.3 \text{ K}$). The tiny DMRG value is because of the large broadening present from finite size effects. We expect the Schwinger Boson result to more accurately reflect the theoretical QFI, and it is in good agreement with our experimental value of $3.4(2)$. (We expect the experimental value is smaller because of (i) experimental broadening, (ii) small 3D and anisotropic exchanges in KYbSe_2 , and (iii) the fact that the experimental data is taken at finite temperature while the theoretical calculation is for $T = 0$.) These experimental considerations notwithstanding, the 0.3 K nQFI far exceeds the unentangled threshold, even if the 0.3 K nQFI uncertainty were much larger. This allows us to conclusively rule out a trivial unentangled phase.

We also note that the nature of the nQFI integral guarantees that nQFI will increase as temperature decreases. This is for two reasons: (1) the tanh factor allows more low-energy intensity into the integral as $T \rightarrow 0$, leading to an increase in QFI as T decreases because the most intense features are at the lowest energies. (2) scattering features generally sharpen as temperature decreases. This is certainly the trend in KYbSe_2 as we compare the 1 K , 2 K , and 0.3 K scattering. As the scattering features sharpen, the energy integrated intensity at $Q = K$ will increase as well. Therefore the $T \rightarrow 0$ nQFI will certainly increase even more.

IV. FITTING THE ROTON MODE

To quantify the extent and the gap of the roton-like mode, we fitted the intensity vs energy of many constant- Q cuts as shown in Fig. 4. We used an asymmetric Gaussian to model the mode, and with the exception of two data points near M , it picks out the peak maximum very well. We then fitted these data points to a sinusoidal function $A \sin(Q) + B \sin(3Q) + C$ to estimate the mode maximum and minimum. These fits

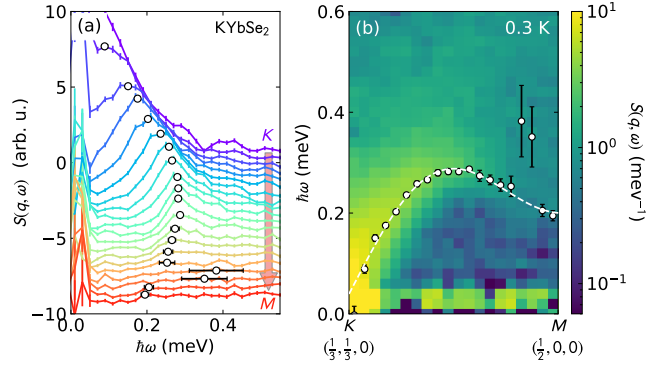


FIG. 4. Fits to the KYbSe_2 roton mode. (a) shows constant Q cuts between K and M , and the fitted mode maximum. Each constant Q slice is offset on the y axis, and the colors show the variation from K and M . (b) shows the mode maximum overplotted on the colormap data, along with a fitted sinusoidal dispersion function. Error bars indicate one standard deviation uncertainty.

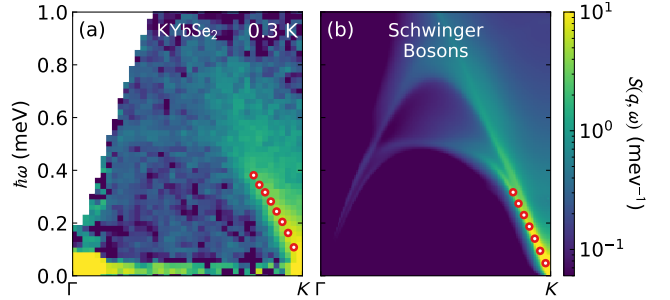


FIG. 5. Fits to the mode maximum between Γ and K for both (a) KYbSe_2 and (b) Schwinger boson simulations. The red dots show the center of a fitted Gaussian, the slope of which was used to determine the energy scale for Schwinger boson simulations. The experimental slope appears to have a nonzero $\hbar\omega$ intercept, but is too small to be directly resolved with this experiment.

show a mode maximum of $0.288(12) \text{ meV}$, a roton minimum $0.200(13) \text{ meV}$, and a fitted gap of $0.059(7) \text{ meV}$. The fitted gap may be an artifact of the mode's deviation from the idealized sin function rather than an actual gap—the higher resolution scan in Fig. 8 of the main text do not reveal a clear gap above $40 \mu\text{eV}$.

We also fitted the low-energy intense mode emanating from K toward Γ in order to match energy scales between theory and experiment, shown in Fig. 5. We fit constant Q cuts to Gaussian curves to define the center of the mode in both KYbSe_2 scattering data and Schwinger boson simulations. We then fit these fitted points to a sinusoidal curve between Γ and K , and scaled the slope of the sin curves at K so that theory matched experiment. This led to a fitted energy scale $J_1 = 0.56(3) \text{ meV}$. The Schwinger boson simulations show the fitted maxima extrapolating toward $\hbar\omega = 0$ at K , but the KYbSe_2 mode maxima appear to have a nonzero intercept. Allowing for a gap in the fitted sin function, we estimate a KYbSe_2 fitted gap of $0.030(5) \text{ meV}$ —too small to be directly resolved using these data.

We can cross-check this fitted J_1 value by comparing to the saturation magnetization. Although saturation magnetization has not been measured for KYbSe₂, it has been measured for the sister compound NaYbSe₂, which has an ab -plane saturation magnetization of ~ 12 T [14]. Assuming the KYbSe₂ value to be close to 12 T, this gives an exchange energy scale of $J_1 = 5.36$ K = 0.462 meV.

V. CRYSTAL ELECTRIC FIELD FITS

Here we describe the procedure used, and the results from the crystal electric field (CEF) fits to the KYbSe₂ ARCS data.

Because of the three-fold Yb³⁺ rotational symmetry in KYbSe₂, there are six nonzero crystal field parameters in the Stevens operator formalism for the Yb³⁺ ion: B_2^0 , B_4^0 , B_4^3 , B_6^0 , B_6^3 , B_6^6 [15]. The effective $J = 7/2$ of Yb³⁺ allows for four energetically-distinct Kramers doublet states, which means three crystal field excitation peaks should be visible in the neutron spectrum.

The crystal field excitations in Fig. 6 can be distinguished from the phonon background by the dependence upon Q : phonon intensity grows with Q while magnetic intensity decreases with Q according to the magnetic form factor. Three of the latter excitations are visible: one near 32 meV, one near 23 meV, and one near 17 meV. Unfortunately, as shown in Fig. 6, the 17 meV mode sits atop an intense flat phonon band which extends to low Q (it is clearly a phonon because its intensity grows with both temperature and $|Q|$), which potentially indicates coupling between phonons and the CEF excitation. (Alternatively, flat-band phonons can have $Q = 0$ intensity from multiple scattering [16].) To verify that the low-energy mode is indeed the third CEF excited level, we measured the CEF spectrum up to 130 meV [Fig. 7], and found no additional visible CEF levels. Furthermore, the observed energies are close to (i) point charge calculations which predict excited modes at 6.4 meV, 18.4 meV, and 33.0 meV, and (ii) measured crystal field excitations of sister compound NaYbSe₂ of 15.8 meV, 24.3 meV, and 30.5 meV [17]. Therefore, we are confident that the 17 meV, 23 meV, and 32 meV peaks are the three excited Yb³⁺ CEF levels.

1. Fitting procedure

To fit the six crystal field parameters to the data, we started with a point-charge model calculation of the crystal field levels, which predicts energies at 6.4 meV, 18.4 meV, and 33.0 meV. We then used *PyCrystalField* [18] to fit the model to the neutron scattering data between 1 Å⁻¹ and 2 Å⁻¹ using the point charge model as starting values.

In order to isolate the crystal field excitations from the phonon background, we employed a self-consistent background subtraction scheme. We used the highest temperature data (300 K for $E_i = 50$ meV and 200 K for $E_i = 35$ meV) as background, but subtracted off the simulated CEF intensities and then rescaled the subtracted data to match the lower-temperature phonons. This way, the background improves as

the CEF model improves, such that the best fit CEF model subtracts off the visible CEF excitations at high temperatures. Because the frequencies of the phonon spectrum are not precisely known, we created a phenomenological energy-dependent scale function to apply the background to lower temperatures. It was a step function of the form

$$\frac{a}{\exp[(\hbar\omega - \mu)/k_B T] + 1} + b$$

where a , b , and μ were fitted to the ratio of high- T to low- T scattering data at energy transfers where no crystal electric excitations are present. As shown in Fig. 6, it produces a reasonable background for the fits.

As in NaYbSe₂ [17], the crystal field levels broaden in energy and shift to higher energies as temperature increases, as shown in Fig. 8. The broadening indicates a shorter excitation lifetime, and is typical for crystal field levels at high temperatures. The shift in energy indicates CEF-phonon coupling, which is not surprising given that the lowest energy CEF mode is at nearly the same energy as an intense phonon band. To account for this in our fits, we applied an ad-hoc shift to the higher temperature energy eigenvalues so that they match the data. In theory, these shifts occur because of slight shifts in the CEF Hamiltonian and require a separate CEF fit—but in order to constrain the low-temperature Hamiltonian it was necessary to include the higher temperature data. Thus we assume that the slight shift in energy indicates a negligible change in the mode intensities, and the resulting fit matches the data very well.

We simulated the crystal field excitations with a Voigt profile, with a temperature-dependent Lorentzian width to account for finite lifetime and a Gaussian width to account for instrumental resolution. The Lorentzian widths were fitted to the two highest peaks for each temperature prior to the Hamiltonian fit and were kept fixed throughout the fit. The resolution function was treated as a fitted parameter, and was allowed to vary linearly as a function of energy transfer but not temperature. The fitted resolution was allowed to vary between $E_i = 50$ meV and $E_i = 35$ meV. Also, an overall scale factor was fitted to the data, one for each incident energy. We simultaneously fit the 7 K, 100 K, and 200 K $E_i = 50$ meV data and the 7 K and 100 K $E_i = 35$ meV data. χ^2 minimization was performed with Powell's method [19] as implemented by Scipy [20]. The best fit crystal field parameter values are given in Table I, and the resulting crystal field Hamiltonian eigenstates are listed in Table II. The best fit calculated g -tensor is given in the main text.

We calculated the uncertainty for these parameters by using a Monte Carlo stochastic search method to map out the χ^2 contour around the best fit model [21]. Using a series of Markov chains, we generated several thousand solutions within $\Delta\chi^2 = 1$ of the best fit minimum $\chi_{red}^2 = 65.86$. This search was aided by principal component analysis of the valid solutions using Scikit [22], such that the random guesses were more along principal component axes. The CEF parameter, g -tensor, and eigenvector uncertainties were calculated from the range of valid values in this set.

It is often the case that crystal field fits to neutron data are

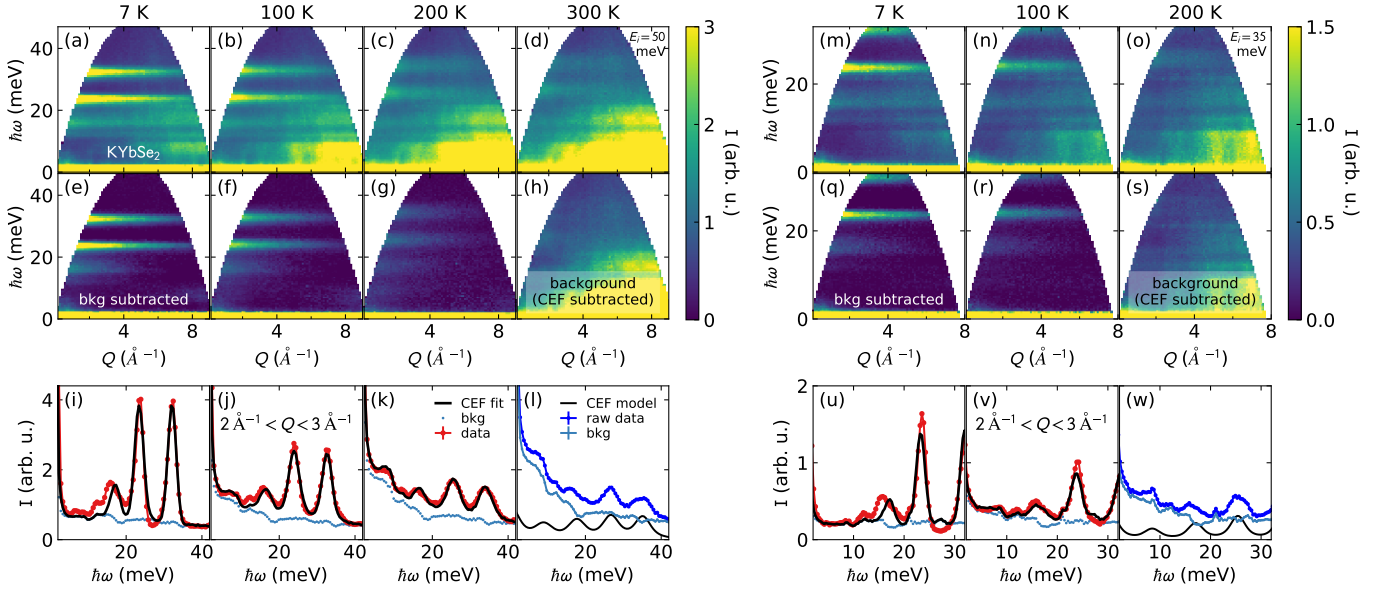


FIG. 6. KYbSe₂ crystal field fit. The top row shows the raw data measured with $E_i = 50$ meV (left section) and $E_i = 35$ meV neutrons (right section). The middle row shows the background subtracted data, with the model-subtracted backgrounds shown in panels (h) and (s). The bottom row shows the fitted data between $2 \text{ \AA}^{-1}$ and $3 \text{ \AA}^{-1}$. Red data shows the raw data, light blue data shows the rescaled high-temperature background. The black line shows the CEF model plus the fitted background. The backgrounds are shown in panels (l) and (w). All error bars indicate one standard deviation uncertainty.

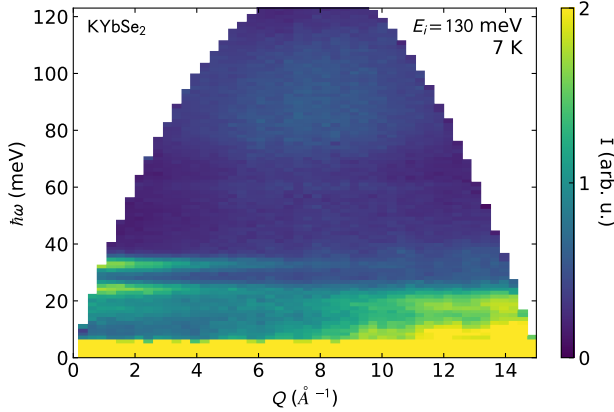


FIG. 7. Crystal field spectrum of KYbSe₂ measured with $E_i = 130$ meV neutrons. No crystal field excitations are visible above 40 meV, confirming the model derived in this study.

TABLE I. Best fit crystal field parameters for KYbSe₂. The middle column gives the nearest neighbor point charge model for KYbSe₂ and the right column shows the best fit values.

Crystal field parameter	Point charge model	Best fit
B_0^0	-0.556	-0.16(2)
B_0^0	0.0088	0.004(2)
B_3^3	-0.281	-0.15(2)
B_0^0	0.00005	0.00038(5)
B_3^3	0.0002	0.0143(5)
B_6^6	0.00043	0.0103(5)

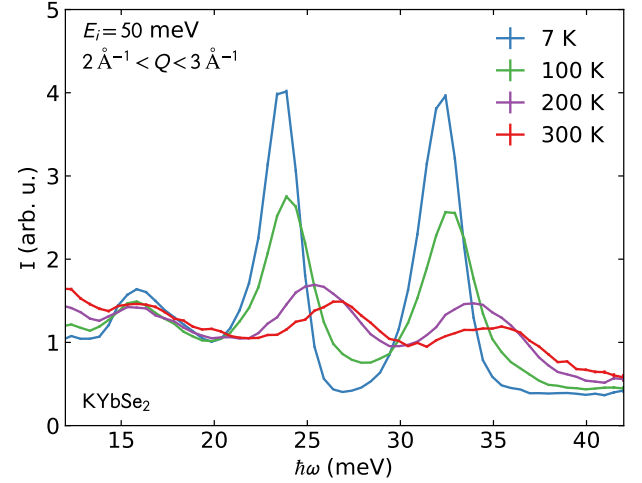


FIG. 8. KYbSe₂ crystal field excitations as a function of temperature. The top two peaks noticeably shift to higher energies as temperature increases, while the bottom level stays constant. This effect was accounted for in the model fit. Error bars indicate one standard deviation statistical uncertainty.

underconstrained, and wildly different Hamiltonians can fit the data equally well [21, 23]. The same is true here: two different models emerged from the fits, one with easy-axis magnetism and one with easy-plane. To select the correct Hamiltonian, we compared the calculated single-ion susceptibility to the measured susceptibility in Fig. 9. Measured susceptibility clearly shows an easy-plane magnetism at low temperature shifting to easy-axis magnetism at high temperature. The

TABLE II. Eigenvectors and eigenvalues for the best fit KYbSe₂ CEF Hamiltonian. Numbers in parenthesis are one standard deviation uncertainty.

E (meV)	$ - \frac{7}{2} \rangle$	$ - \frac{5}{2} \rangle$	$ - \frac{3}{2} \rangle$	$ - \frac{1}{2} \rangle$	$ \frac{1}{2} \rangle$	$ \frac{3}{2} \rangle$	$ \frac{5}{2} \rangle$	$ \frac{7}{2} \rangle$
0.0	0.0	0.78(3)	0.0	0.0	-0.44(4)	0.0	0.0	-0.44(3)
0.0	-0.44(3)	0.0	0.0	0.44(4)	0.0	0.0	0.78(3)	0.0
17.1(3)	0.0	-0.09(3)	0.0	0.0	0.61(3)	0.0	0.0	-0.79(2)
17.1(3)	-0.79(2)	0.0	0.0	-0.61(3)	0.0	0.0	-0.09(3)	0.0
23.24(5)	0.0	0.0	1.0	0.0	0.0	0.0	0.0	0.0
23.24(5)	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0
31.93(5)	0.43(3)	0.0	0.0	-0.66(4)	0.0	0.0	0.62(4)	0.0
31.93(5)	0.0	0.62(4)	0.0	0.0	0.66(4)	0.0	0.0	0.43(3)

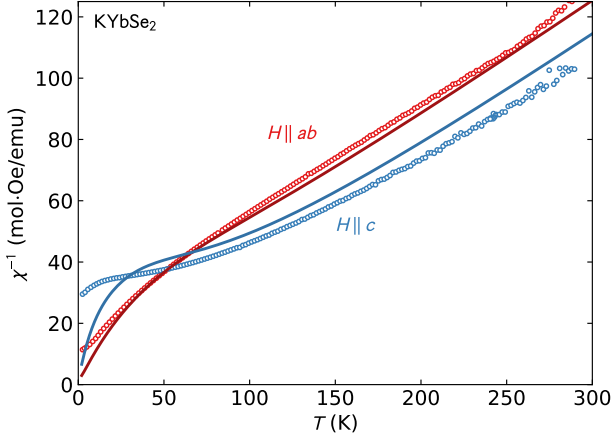


FIG. 9. KYbSe₂ susceptibility compared to best fit CEF calculated single crystal susceptibility. Qualitatively, the simulation matches experiment, though the experiments show a higher susceptibility in the *c* direction.

easy-plane model matches this behavior very well, and thus we select it as the correct model. However, this highlights the need to cross-check any fitted Hamiltonian with a different measure of magnetic anisotropy.

Although the calculated susceptibility qualitatively matches the measured susceptibility, the correspondence is not perfect (especially in the *c* direction). Although this discrepancy is within error bars of the fitted Hamiltonian, two additional complications may also prevent perfect agreement: (i) magnetic exchange which shifts measured susceptibility values, and (ii) a shifting CEF Hamiltonian as a function of temperature. Because of these effects, we did not use susceptibility data to constrain the fit itself.

One final cross-check of the crystal field model can be made by comparing the calculated saturation magnetization to the measured KYbSe₂ 1/3 magnetization plateau. According to ref. [24], the 0.42 K 1/3 magnetization plateau occurs at $\mu_0 H = 4.2$ T, $M = 0.569 \mu_B$. However, this value is offset by Van Vleck susceptibility which at low fields adds a linear offset to the ground state CEF magnetization. According to the fitted KYbSe₂ CEF model, the Van Vleck susceptibility is $0.0176 \mu_B/T$ at 0.42 K—which means we must subtract

$(0.0176 \mu_B/T)(4.2 \text{ T}) = 0.074 \mu_B$ from the measured plateau magnetization for a true 1/3 magnetization of $0.495 \mu_B$. This is one third of the CEF predicted *ab*-plane saturation magnetization $1.48(9) \mu_B$. Thus we have high confidence that our fitted CEF model and associated *g*-tensor is correct.

VI. ONSAGER REACTION FIELD FITS

Here we outline the Onsager reaction field (ORF) approach for completeness. We consider the spin Hamiltonian

$$\mathcal{H} = \sum_{\langle i,j \rangle} \left\{ J_X \left(S_i^x S_j^x + S_i^y S_j^y \right) + J_Z S_i^z S_j^z \right. \\ \left. + J_A \left[(S_i^x S_j^x - S_i^y S_j^y) \cos \phi_{ij} - (S_i^x S_j^y + S_i^y S_j^x) \sin \phi_{ij} \right] \right\}, \quad (\text{S.3})$$

in which $\alpha \in \{x, y, z\}$ denote spin components with respect to Cartesian axes $\mathbf{x}, \mathbf{y}, \mathbf{z}$, and $\phi_{ij} \in \{\frac{2\pi}{3}, -\frac{2\pi}{3}, 0\}$ as specified in Ref. [25]. We use the Onsager reaction-field (ORF) approach [26–28] to calculate magnetic diffuse scattering patterns. The Fourier transform of the interactions is given by

$$J_{\alpha\beta}(\mathbf{Q}) \equiv - \sum_{\mathbf{R}} J_{\alpha\beta}(\mathbf{R}) e^{-i\mathbf{Q} \cdot \mathbf{R}}, \quad (\text{S.4})$$

where $J_{\alpha\beta}(\mathbf{R})$ is the coefficient of $S_i^\alpha S_j^\beta$ in Eq. (S.3) for sites *i* and *j* separated by a lattice vector $\mathbf{R}$.

The magnetic diffuse scattering intensity is given, in the reaction-field approximation, by

$$I_{\text{ORF}}(\mathbf{Q}) \propto [f(Q)]^2 \sum_{\mu=1}^3 \frac{|s_\mu(\mathbf{Q})|^2}{1 - \chi_0(\lambda_\mu(\mathbf{Q}) - \lambda)}, \quad (\text{S.5})$$

where $\chi_0 = 1/3T$ is the Curie susceptibility and λ_μ denotes the eigenvalues of the interaction matrix, where μ labels its 3 eigenmodes. The structure factor

$$s_\mu(\mathbf{Q}) = \sum_{\alpha} (\hat{\mathbf{n}}_\alpha - \mathbf{Q} \hat{\mathbf{n}}_\alpha \cdot \mathbf{Q} / Q^2) g_\alpha U_\mu^\alpha, \quad (\text{S.6})$$

where $\hat{\mathbf{n}}_\alpha \in \{\mathbf{x}, \mathbf{y}, \mathbf{z}\}$, g_α denotes components of the diagonal *g*-tensor, and U_μ^α denotes the eigenvector components

of the interaction matrix. At each temperature, we obtain the reaction field λ self-consistently by enforcing that $\sum_{\mu, \mathbf{q}} [1 - \chi_0(\lambda_{\mu}(\mathbf{q}) - \lambda)]^{-1} = 3N_{\mathbf{q}}$ for a grid of $N_{\mathbf{q}} = 40^3$ wavevectors in the Brillouin zone. The best fit values are given in the methods section of the main text.

We also performed the fit including the off-diagonal J_B component. [25]. This quantity is difficult to determine because it depends upon distinguishing K from K' , and there is some degree of twinning in KYbSe₂ which means we can only fit the magnitude of J_B . Nevertheless, for completeness we performed the ORF fit assuming a twinning model and found

$$\begin{aligned} J_X &= 2.33(10) \text{ K} & J_Z &= 2.28(10) \text{ K} \\ J_A &= -0.018(8) \text{ K} & J_2 &= 0.11(2) \text{ K} \end{aligned} \quad (\text{S.7})$$

$$|J_B| = 0.00(5) \text{ K}. \quad (\text{S.8})$$

The error bar indicates $|J_B|$ could be larger than $|J_A|$, but this is still much smaller than J_X and J_Y , indicating that the Heisenberg model is still appropriate for KYbSe₂.

VII. SCHWINGER BOSON CALCULATIONS

Figure 10 shows additional Schwinger boson calculated spectra for various values of nearest neighbor exchange anisotropy J_Z/J_X , where $J_X = J_Y$. As anisotropy increases, a low-energy mode at K becomes gapped. However, high-resolution experimental scattering shows no such mode, even at the highest resolution setting (0.02 meV FWHM). This is consistent with the Onsager Reaction Field fits, which show $J_{ZZ} = J_{XX}$ to within uncertainty.

Figure 11 shows the calculated Schwinger Boson spectra for different values of J_2/J_1 . As J_2/J_1 increases, the spectra bandwidth slightly decreases, while the gap at M grows smaller.

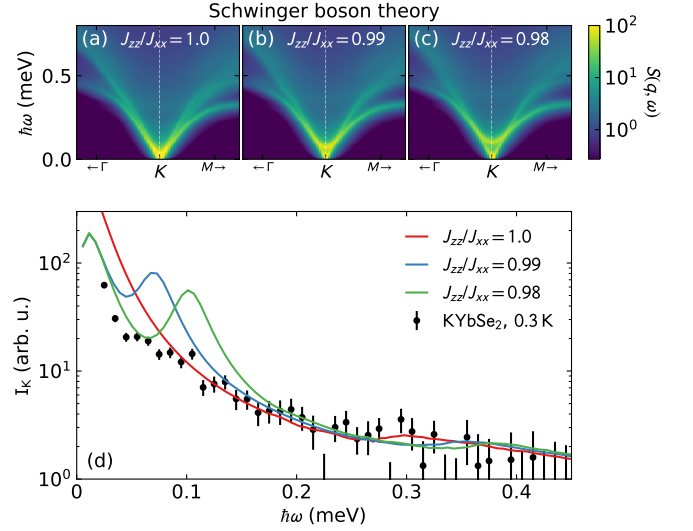


FIG. 10. Effects of exchange anisotropy on the magnetic structure factor. Panels (a)-(c) show the low-energy structure factor predicted by Schwinger boson theory for different values of exchange anisotropy, where $J_X = J_Y$. As anisotropy increases, a well-defined mode becomes gapped. Panel (d) shows the energy-dependent scattering at K compared to KYbSe₂ scattering at 0.3 K and $E_i = 1$ meV (0.02 meV FWHM). No finite-energy modes are seen on the scale that is predicted by Schwinger boson theory, suggesting a highly isotropic nearest neighbor exchange. Error bars indicate one standard deviation uncertainty.

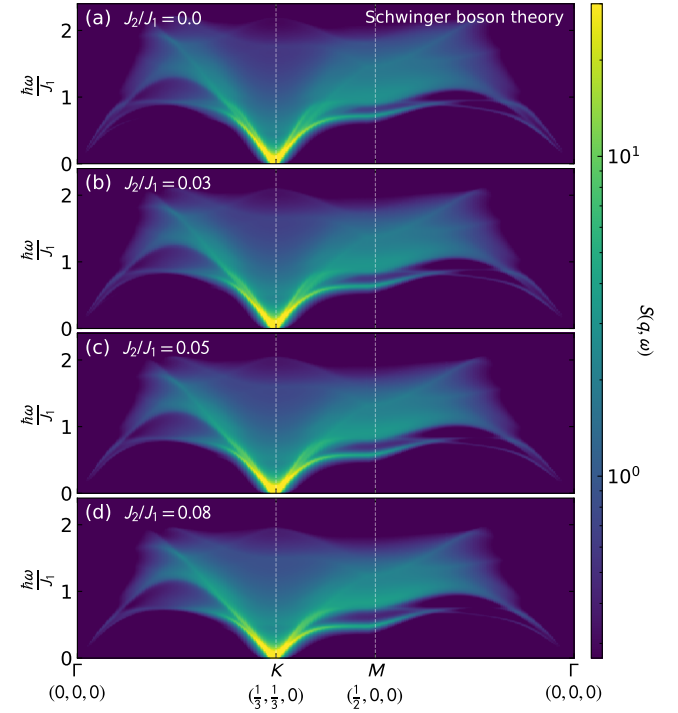


FIG. 11. Schwinger boson calculations for the Heisenberg triangular lattice antiferromagnet for varying values of second nearest neighbor exchange.

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