Non-linear magnons in the 1/3 magnetization plateau of a proximate quantum spin liquid

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Quantum spin liquids (QSL) are theoretical states of matter with long-range entanglement and exotic quasiparticles. However, they elude quantitative theory, rendering their underlying phases mysterious, and hampering experimental efforts to determine candidate materials’ interactions. Here we study resonating valence bond QSL candidate material KYbSe2 in its 1/3 plateau phase, where quantitative theory is tractable. We measure the magnon modes within this phase using inelastic neutron scattering and fit them using nonlinear spin wave theory, and we also fit the heat capacity using high temperature series expansion. Both fits yield the same magnetic Hamiltonian to within uncertainty, confirming previous estimates and showing the Heisenberg $J_2/J_1$ to be an accurate model for KYbSe2, and confirming its status as a proximate QSL.

Triangular lattice quantum magnets have been of intense interest since Anderson’s 1973 prediction of a resonating valence bond (RVB) quantum spin liquid (QSL) [1], but no clear triangular QSL materials exist [2]. In this state, antiferromagnetic interacting spins on a two-dimensional triangular lattice produce a long-range entangled quantum spin liquid rather than conventional long range order [3]. This state has intriguing theoretical properties [4], and quantum spin liquids holds great potential for quantum electronic technologies [5, 6]. Although Anderson’s original proposal was for the nearest neighbor Heisenberg antiferromagnet, subsequent studies showed the pure nearest neighbor model actually orders into a 120° phase at the lowest temperatures [7, 8]. Instead, the QSL state requires a small second neighbor $J_2$ exchange between $\approx 6\%$ and $\approx 16\%$ of the nearest neighbor exchange to stabilize [9–15]. (A similar role is played by nearest neighbor anisotropic exchange [16].) Although it is unclear whether this $J_2$-stabilized phase is Anderson’s RVB or a different type of QSL [15, 17], it is clear that the QSL state should exist—if a material with the right Hamiltonian could be found.

Many materials have been proposed as 2D triangular lattice antiferromagnets, including Ba$_2$CoSB$_2$O$_8$ [18–20], YbMgGaO$_4$ [21–25], and organic salts [26–28]. However, none of these have been found to have an RVB ground state. A promising new class of materials is the Yb$^{3+}$ delafossites, which have magnetic Yb$^{3+}$ in a crystallographically perfect 2D triangular lattice [3, 29–32, 34]. However, the crucial test in evaluating candidate RVB materials is whether their magnetic exchange Hamiltonians are indeed within a theoretical QSL phase.

Part of the difficulty in experimentally studying an RVB liquid is that its lacks sharp spectral features: its elementary quasiparticles are $S = 1/2$ spinons, which are created in pairs and thus produce a diffuse continuum [35, 36]. And unfortunately, diffuse continua are also produced by disordered or glassy states and are thus ambiguous [25, 37]. However, an important feature of the quantum $S = 1/2$ triangular lattice antiferromagnet is that an applied magnetic field produces a 13-magnetization plateau phase corresponding to up-up-down long range magnetic order [38, 39]. This is an inherently quantum mechanical effect, whereby quantum fluctuations select a collinear spin ordering [40]. Crucially, this produces

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FIG. 1. KYbSe2 crystal structure and phase diagram. (a) The triangular lattice plane with the $J_1$ and $J_2$ exchanges between magnetic Yb sites. (b) The 0.42 K in-plane magnetization from Ref. [3], showing a 1/3 magnetization plateau phase at 4 T. (c) The $J_2/J_1$ phase diagram, showing the refined KYbSe$_2$ $J_2/J_1$ is extremely close to the resonating valence bond (RVB) spin liquid state.
well-defined magnon modes within the plateau phase, whose gapped dispersion can be calculated via nonlinear spin wave theory [6, 40]. This observation allows us to extract the Hamiltonian parameters without the need for reaching the fully polarized magnetic state (the saturation field is prohibitively large for most RVB candidate materials [6, 42]). The problem thus becomes tractable by studying the magnons in the plateau phase.

KYbSe₂ [3] was recently studied using neutron scattering, and fits to paramagnetic diffuse scattering show it to be a proximate QSL: it orders magnetically at 290 mK, but its fitted $J_2/J_1$ ratio is close enough to the QSL phase to exhibit some of its exotic behaviors (see Fig. 1), including diffuse zero-field neutron spectra from bound spinons [2]. However, the previous fit relied upon classical models compared to paramagnetic scattering. A robust Hamiltonian fit requires quantum calculations. Here we provide such analysis using inelastic magnetic scattering and heat capacity, showing KYbSe₂ to be a remarkably good realization of the $J_2/J_1$ triangular lattice Heisenberg antiferromagnet.

Like the ideal 2D triangular Heisenberg model [38, 39], KYbSe₂ has a 1/3 saturation magnetization plateau at an in-plane magnetic field of 4 T [Fig. 1(b)], which also accompanies a reentrant magnetic ordered phase [3]. Presumably, this ordered phase is the up-up-down phase of the quantum $S = 1/2$ triangular lattice antiferromagnet. This finite field ordered phase is itself evidence of a highly quantum ground state, and also provides an opportunity to observe coherent spin wave modes at modest magnetic field, and fit them to a non-linear spin wave theory (NLSWT) [6, 40]. This provides an independent test of whether the $J_1$-$J_2$ Hamiltonian is appropriate to KYbSe₂, and what the $J_3/J_1$ ratio is.

We assume a $J_1$-$J_2$ Hamiltonian in a magnetic field

$$\hat{H} = J_1 \sum_{(i,j)} \hat{S}_i \cdot \hat{S}_j + J_2 \sum_{(i,j)} \hat{S}_i \cdot \hat{S}_j - h \sum_i \hat{S}_i^z (1)$$

where $h = g_{xx} \mu_B B$, $B$ is the magnitude of the applied magnetic field along the $x$-direction and $g_{xx}$ is the $g$-tensor.

By following the procedure described in Ref. [6], we fit the NLSWT magnon dispersion to the mode energies as a function of wavevector extracted from Gaussian profiles at constant $Q$ slices [see Fig. 3(a)]. Treating the three Hamiltonian parameters $J_1$, $J_2$, and $h$ as fitted constants (magnetic field $h$ is a fitted parameter because $g_{xx}$ has large uncertainty for KYbSe₂ [48]), we fit the NLSWT mode energies to the experimentally measured dispersions as shown in Fig. 3(c). This gives a very good reproduction of the lower energy modes, as shown in Fig. 3(b). To define uncertainty, we calculated a contour $\chi^2_{red} = 1$ above the reduced $\chi^2$ minimum [49]. One standard deviation and two standard deviation contours are plotted in Fig. 3(d). Using a one standard deviation uncertainty, the best fit values are $J_1 = (0.456 \pm 0.013)$ meV, $J_2/J_1 = 0.043 \pm 0.010$, and $h/J_1 = 1.73 \pm 0.05$. This $J_2/J_1$ ratio agrees to within uncertainty with the $J_2/J_1 = 0.047 \pm 0.007$ from the Onsager reaction field fits in Ref. [2]. We note that the exact saturation field for the proposed model is $h_{sat}/J_1 = 9/2$, implying that $h/h_{sat} = 0.38 \pm 0.01$.

Single-magnon NLSWT calculations did not capture the dispersive feature above 1 meV in Fig. 3(a). However, two-magnon continuum calculations (shown in the Supplemental Information [46]) do capture this feature. The sharpness of this mode in an otherwise diffuse continuum arises from the two-dimensional nature of the system, such that the density of two-magnon states has a Van Hove singularities at an energies that depend on their total momentum. These singularities are broadened by the finite experimental resolution, leading to a dispersive peak in the continuum scattering arising from the longitudinal spin structure factor, as was seen in YbCl₃ [50].

As another independent test of our $J_2/J_1$ model, we also compared the KYbSe₂ heat capacity to theoretical calculations. Using the zero field heat capacity reported in Refs. [2, 3], subtracted by KLuSe₂ heat capacity to isolate the magnetic heat capacity (see the Supplemental Information for details [46]), we compared the data to the high temperature series expansion interpolation calculations [51–54], which give accurate quantum calculations of the high temperature heat capacity of the 2D triangular lattice as a function of $J_2/J_1$. The series were calculated out to the 14th order (one order more than Ref. [54]). These data are shown in Fig. 4.

The zero field heat capacity was measured in a dilution refrigerator below 2.5 K and a $^4$He cryostat above 1.8 K separately and combined. For temperatures greater than 2 K, the theory and experiment match very closely, with subtle differences depending on the precise $J_2/J_1$ value. To quantitatively compare theory and experiment, we define a $\chi^2_{red}$ for $2 K < T < 8 K$. (8 K is where the lattice heat capacity becomes much larger than the magnetic contribution, and the
FIG. 2. KYbSe$_2$ field dependent scattering. The top row shows the zero field neutron spectrum, showing a diffuse continuum with a lower bound coming to zero energy at $\hbar \omega = (1/3, 1/3)$ (in reciprocal lattice units). The middle row shows the same slices for $B = 4$ T and the bottom row for $B = 8$ T. The left two columns show cuts along $\hbar \omega$ (the far left integrated over $1/5 \leq \ell \leq 1/5$ r.l.u., the center-left column integrated over $1.5 < \ell < 4.5$), and the right two columns show cuts along $\ell$. For $B = 0$ T and $B = 8$ T, there is no appreciable $\ell$ dependence to the scattering, but at $B = 4$ T there is a modulated intensity to some of the features as shown most clearly in panel (g).

FIG. 3. Nonlinear spin wave fit to KYbSe$_2$ scattering at 4 T. The left panel shows the experimental scattering integrated along ($\hbar \omega$), with the grey data points indicating the mode centers determined using constant $Q$ cuts fitted to a Gaussian profile in energy. Panel (b) shows the best calculated neutron spectrum from nonlinear spin wave theory (NLSWT). Note that the highest energy mode is not captured by the single-magnon scattering calculation. Panel (c) shows the fitted points from panel (a) with the NLSWT dispersion curves used to fit the data. Panel (d) shows the one and two standard deviation $\chi^2$ contours of $J_2/J_1$ and magnetic field $h$.

Subtracted values are more questionable. For each theoretical $J_2/J_1$ value, we fit $J_1$ to minimize $\chi^2$ and thus obtain $\chi^2_{\text{red}}$ as a function of $J_2/J_1$, shown as an inset to Fig. 4. This fit yields best fit values of $J_1 = (0.439 \pm 0.010)$ meV and $J_2/J_1 = 0.037 \pm 0.013$. This agrees to within uncertainty with the results from NLSWT fits.

It should be noted that the high temperature expansion does not capture either the maximum in heat capacity at 1 K or the ordering transition at 290 mK. The lack of ordering transition is not surprising as the theory is based off a perfectly isotropic 2D lattice, which only orders at $T = 0$ [55]. The 1 K “bump”, also seen in NaYbO$_2$ [31], is more challenging to explain. The failure of the high-temperature expansion in capturing this peak suggests that it is either induced by an increase of the magnetic in-plane correlation length beyond several lattice spaces, $\xi \gtrsim 10a$, or by the onset of three-dimensional correlations in KYbSe$_2$, which ultimately leads to long range magnetic order. Be that as it may, the series expansion theory
that the Onsager reaction field theory from Ref. [2] cannot estimate
different experiments and three different theoretical techniques. Note
that the Onsager reaction field theory from Ref. [2] cannot estimate
theoretical techniques to fit the KYbSe$_2$ from three
different fits of the KYbSe$_2$ heat capacity, and
allows us to extract both $J_1$ and $J_2$ from the data.
At this point, we have three independent fits of the KYbSe$_2$
magnetic exchange Hamiltonian, which are summarized in
Table I. Each fit used a different experiment and a different
experimental technique, so it is remarkable that all three agree
to within uncertainty. The two theoretical fits performed in
this study (NLSWT and high temperature heat capacity series
expansion) include quantum effects, and are appropriate for the
highly quantum KYbSe$_2$. That being said, the Onsager reaction
field theory fit agrees to within uncertainty with these two
other methods even though it neglects quantum effects. This
is evidence that fitting paramagnetic scattering with classical
methods [56] works even for highly quantum systems. Also,
this is a remarkable confirmation of the results and methodology
in Ref. [2].
The study here shows that KYbSe$_2$ is an excellent example of a
spin-$1/2$ $J_2/J_1$ Heisenberg magnet on a triangular lattice. The use of nonlinear spin-wave theory at an intermediate—rather than fully saturated—field provides an alternative method to that in Ref. [42] which requires a fully
polarized system. This is then applicable not only to other delafossite materials but also other candidate spin liquids where the saturation field cannot be reached. The confirmation that
KYbSe$_2$ is very close to a quantum spin liquid state provides
strong impetus to further study this class of compounds as a
rational route to quantum spin liquids.

In conclusion, we have used two quantum simulation tech-
niques to fit the KYbSe$_2$ magnetic exchange Hamiltonian to
developmental: nonlinear spin waves to finite-field scattering
in a plateau phase, and high temperature expansions to zero
field heat capacity. Both give extremely good agreement with
experiment (down to an energy scale of 2 K), showing that
the Heisenberg $J_2/J_1$ model is an excellent minimal model for
KYbSe$_2$. Furthermore, we find a $J_2/J_1$ ratio extremely close
to the 2D triangular lattice spin liquid state at $J_2/J_1 \approx 0.06$.
With three independent experimental measurements all giving
the same result, this definitively verifies the model proposed in
Ref. [2]. Thus we expect to find approximate spin liquid behavior in KYbSe$_2$ and in related materials.

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TABLE I. Best fit exchange Hamiltonian value for KYbSe$_2$ from three
different experiments and three different theoretical techniques. Note
that the Onsager reaction field theory from Ref. [2] cannot estimate
$J_1$ as it is classical. The last row gives the weighted mean.

| Theoretical technique       | $J_1$ (meV) | $J_2/J_1$ |
|----------------------------|-------------|-----------|
| Onsager reaction field      | NA          | 0.047 ± 0.007 |
| Nonlinear spin waves        | 0.456 ± 0.013 | 0.043 ± 0.010 |
| Heat capacity               | 0.429 ± 0.010 | 0.037 ± 0.013 |
| Weighted mean               | 0.438 ± 0.008 | 0.044 ± 0.005 |

FIG. 4. KYbSe$_2$ zero-field heat capacity (black) compared to theoretical calculations of the heat capacity from Ref. [54]. For the theory, the only adjustable parameter is the $T$ axis scaling (corresponding to $J_1$). The inset shows reduced $x^2$ as a function of $J_2/J_1$, showing a minimum at $J_2/J_1 = 0.040(12)$.
SUPPLEMENTAL INFORMATION FOR NON-LINEAR MAGNONS IN THE 1/3 MAGNETIZATION PLATEAU OF A PROXIMATE QUANTUM SPIN LIQUID

I. EXPERIMENTS

A. Cold Neutron Chopper Spectrometer (CNCS)

For the CNCS experiment, we used a sample made of 19 coaligned crystals (total mass ~ 200 mg) oriented in the (h0l) scattering plane glued to aluminum plates (see Fig. S1). The sample was mounted in a helium dilution refrigerator with an 8 T magnet and measured with double-disc chopper frequency 300.0 Hz (high-flux mode, 9 degree opening on the double disk) for an incident energy $E_i = 3.32$ meV. At each field the sample was rotated $180^\circ$ to map the neutron spectrum. The processed CNCS data was then corrected for the isotropic Yb$^{3+}$ form factor [S1]. This data (prior to background subtraction) is shown in Fig. S2. The left two columns are integrated over $\ell \pm 1.5$ reciprocal lattice units (RLU), and the right two columns are integrated over $h\ell \pm 0.02$ RLU, and the integration ranges for the main text data are the same.

Fig. S1. KYbSe$_2$ sample used to measure the field-dependent spin excitations on CNCS. 19 plate-like crystals were coaligned and glued to two aluminum plates with the [110] direction vertical and the [001] direction orthogonal to the aluminum surface.

To clearly resolve the low-energy magnetic scattering, it was necessary to subtract the background (sample environment scattering and phonon scattering) from the data. Because a perfect background is not available for this data, we created a phenomenological background using 12 K scattering data where magnetic correlations are negligible [S2, S3], shown in Fig. S3(a)-(d). Because the higher temperature intensities phonon scattering, we removed certain intense inelastic features in the slices and filled in via Astropy’s interpolation routine [S4]. Then, following the method in Ref. [S2], we subtracted the median intensity for each energy from the scattering data for inelastic ($> 0.38$ meV) data. Finally, we convolved the inelastic ($> 0.38$ meV) data with a two-dimensional Gaussian profile to reduce the noise. This final phenomenological background is shown in Fig. S3(e)-(f). By comparing the supplemental information Fig. S2 to the main text Fig. 1, it is clear that this approach approximately isolates the magnetic scattering quite nicely.

Although the data have a large elastic background, substantial elastic scattering appears in the finite-field plateau phase, plotted in Fig. S4. At 4 T, a streak of elastic scattering appears at (1/3, 1/3, $\ell$), in accord with in-plane magnetic order of the plateau phase. If we isolate the magnetic scattering by subtracting the 0 T data (where the magnetic order is extremely weak and barely any elastic magnetic signal is visible) from the 4 T data as in Fig. S4(e), we see that the 4 T magnetic signal has a sinusoidal intensity modulation with $\ell$, as shown in Fig. S4(f). The intensity peaks at $\ell = 0$ and $\ell = \pm 3$, the same periodicity as the triangular planes, signaling short-ranged ferromagnetic correlations between the triangular lattice planes with a propagation vector $Q = (1/3, 1/3, 0)$. This is markedly different from the elastic magnetic scattering at 4 T measured in CsYbSe$_2$, which had maximal intensity at $\ell = \pm 1$ and a propagation vector $Q = (1/3, 1/3, 1)$ [S5]. Furthermore, in CsYbSe$_2$ the correlations extend to at least three triangular lattice planes, but in KYbSe$_2$ the magnetic correlations appear to extend only to the neighboring planes. Thus KYbSe$_2$ appears to be more two-dimensional than CsYbSe$_2$.

B. Heat capacity uncertainty

The heat capacity data presented in the main text is made of two different measurements: one measured in a dilution refrigerator [S2], and another on a Quantum Design PPMS with a $^3$He insert [S3]. The KYbSe$_2$ sample mass was measured more precisely for the dilution refrigerator experiment, and thus we scaled the higher temperature PPMS data to match the dilution refrigerator measurement for the temperature window $0.4$ K $< T < 2.0$ K, giving a renormalizing scale factor of 0.99 to the high temperature data. Heat capacity of nonmagnetic KLuSe$_2$ was subtracted from all data to isolate the magnetic heat capacity.

The uncertainties for the heat capacity values came from three sources: (i) uncertainty in mass normalization, which we estimate from the the difference between the absolute heat capacities of the two different dilution refrigerator measurements on two different KYbSe$_2$ samples reported in Ref. [S2]. (ii) Variance in heat capacity measured at the same temperature: in the $^3$He heat capacity measurement, data points were repeated three times at each temperature. (iii) Difference in heat capacity between two separate $^3$He refrigerator measurements. The uncertainties from these three sources were added in quadrature to yield the uncertainties plotted in the main text Fig. 4.

II. NONLINEAR SPIN WAVES

A. Two magnon continuum

In the experimental KYbSe$_2$ scattering, an additional broadened mode is visible above 1 meV which is not captured by the single-magnon NLSWT calculation (main text Fig. 3). However, when we calculate the two-magnon continuum scattering
FIG. S2. KYbSe₂ field dependent scattering without background subtraction. The top row shows the zero field neutron spectrum, the middle row shows the same slices for $B = 4$ T, and the bottom row for $B = 8$ T. These data should be compared with main text Fig. 1. At high energy transfers the features are clear, but background subtraction (Fig. S3) is necessary to resolve low energy features.

FIG. S3. KYbSe₂ scattering background. The top row shows the raw $T = 12$ K data. To process this and use it as a background for the data in Fig. S2, we removed the intense features [e.g., at 0.7 meV in panel (b)], subtracted the energy-dependent median inelastic scattering intensity from every energy, and convolved with a Gaussian function to smooth the data. This resulted in the background in the bottom row, which was subtracted from the scattering data in Fig. S2 to generate the data in main text Fig 1.

(following the method in Ref. [S6]), we see a similar mode appear in the simulation, as shown Fig. S5(b).

Although the overall character and bandwidth of the calculated two-magnon continuum matches experiment, Fig. S5(c) shows the calculated intensity is slightly weaker than what is experimentally observed. There could be several explanations for this: (i) the experimental background subtraction was imperfect and left some nonlinear anomalous intensity above 1 meV. (ii) The $g$-tensor is different than the best fit crystal field values ($g_{zz}$ has a very large error bar [S2]), which leads to different weight being given to $S_{xx}$ vs $S_{yy}$ and $S_{zz}$ and more intensity in the continuum. (iii) The NLSWT approximation neglects higher order effects, and thus it may be slightly underestimating the weight of the continuum scattering. Despite this discrepancy, because of the close resemblance to the experimental dispersion, we are confident that the observed mode above 1 meV is in fact a two-magnon scattering effect.
B. Nonlinear effects

For a highly quantum system like a $S = 1/2$ triangular lattice in the $1/3$ magnetization plateau phase, nonlinear corrections to magnon dispersions become significant. To illustrate this, Figure S6 shows the linear spin wave dispersions compared to the nonlinear dispersions for the best fit KYbSe$_2$ parameters from the main text. Particularly at low energies, the nonlinear corrections are very significant. Note that the existence of a gap (signaling a finite-field plateau phase) requires nonlinear effects to capture; this is because the plateau phase is an inherently quantum-mechanical phenomenon [S6]. Thus accurately extracting the Hamiltonian parameters from the neutron scattering measurements on KYbSe$_2$ requires a nonlinear spin wave model.

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FIG. S5. Two magnon continuum of KYbSe$_2$. Panel (a) shows the experimental data at $B = 4$ T, and panel (b) shows the nonlinear spin wave calculated intensity with the two-magnon contribution included. Panel (c) compares the experiment with the theory at $(1/3, 1/3, 3)$. If we assume some small remnant background the overall shape and intensity of the two-magnon contribution is close to what we observe in experiment, though the calculated two-magnon intensity is slightly lower than experiment.

FIG. S6. Nonlinear spin wave calculations (blue) compared to linear spin wave calculations (red) for the triangular lattice using best fit parameters in the main text. Note that at this field, nonlinear corrections to the spin wave modes are significant, especially at low energies.