

Field-Induced Magnon Decay, Magnon Shadows, and Rotonlike Excitations in the Honeycomb Antiferromagnet YbBr₃

J. A. Hernández¹, A. A. Eberharter², M. Schuler³, J. Lass¹, D. G. Mazzone¹, R. Sibille¹, S. Raymond⁴, K. W. Krämer⁵, B. Normand^{2,6}, B. Roessli¹, A. M. Läuchli^{2,6}, and M. Kenzelmann¹

¹PSI Center for Neutron and Muon Sciences, CH-5232 Villigen-PSI, Switzerland

²PSI Center for Scientific Computing, Theory and Data, CH-5232 Villigen-PSI, Switzerland

³Institut für Theoretische Physik, Universität Innsbruck, A-6020 Innsbruck, Austria

⁴Université Grenoble Alpes, CEA, IRIG, MEM, MDN, 38000 Grenoble, France

⁵Department of Chemistry, Biochemistry and Pharmaceutical Sciences, University of Bern, Freiestrasse 3, CH-3012 Bern, Switzerland

⁶Institute of Physics, Ecole Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland



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Although the search for quantum many-body phenomena in magnetic materials has a strong focus on highly frustrated systems, even unfrustrated quantum magnets show a multitude of unconventional phenomena in their spin excitation spectra. YbBr₃ is an excellent realization of the $S = 1/2$ antiferromagnetic Heisenberg model on the honeycomb lattice, and we have performed detailed spectroscopic experiments with both unpolarized and polarized neutrons at all applied magnetic fields up to saturation. We observe extensive excitation continua, which cause strong renormalization and the decay of single magnons at higher fields, while coherent features include field-induced “shadows” of the single magnons and the spectacular emergence of a rotonlike excitation. To guide and interpret our experiments, we performed systematic calculations by the method of cylinder matrix-product states that provide quantitative agreement with the neutron scattering data and a qualitative benchmark for the spectral signatures of strong quantum fluctuations even in the absence of magnetic frustration.

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Introduction—Quantum magnetic models and materials provide an excellent proving ground for highly entangled quantum many-body phenomena. Much of this research has focused on systems with low dimensionality and high frustration, such that quantum fluctuation effects cause the complete destabilization of magnetic order, and on fingerprints of the resulting quantum spin liquid (QSL) [1,2]. Much more common, however, is “renormalized classical” behavior, in which ordered magnetic properties including the sublattice magnetization, spin stiffness, and spin-wave velocity are finite, but strongly renormalized by quantum fluctuations. Nevertheless, some well-documented quantum phenomena in unfrustrated systems question whether a renormalization description can be sufficient, or if some hallmarks of the QSL may appear at finite energies. Key examples for the paradigm square-lattice Heisenberg antiferromagnet (SLHAF) are the theoretical discussion of magnon decay at high fields [3–7] and

the experimental observation of the $(\pi, 0)$ scattering anomaly [8–10], which sharpened the question of whether the spectral function reflects many multimagnon bound and scattering states [11,12] or a magnon fractionalization [10,13–18].

One of the “most quantum” unfrustrated systems on a regular two-dimensional (2D) geometry is the $S = 1/2$ honeycomb lattice, with coordination number $z = 3$. Although the honeycomb Heisenberg model was studied intensively for its ground-state phase diagram in the presence of frustrating next- (J_2) and third-neighbor (J_3) coupling [19–27], and also for the topological magnon excitations of the nearest-neighbor (J_1) model [28–30], studies of the excitation spectrum have only recently been extended from the semiclassical level [31] to discuss many-body quantum phenomena [32,33]. Layered van der Waals materials offer the possibility of measuring the magnetic properties of 2D systems, and the AX₃ family of lanthanide (A) trihalides ($X = \text{Cl}, \text{Br}, \text{I}$) offers two ideal properties, namely, the precise honeycomb geometry shown in Fig. 1(a) and, for the Yb-based systems, effective spin $1/2$ and saturation fields (B_{sat}) within range of experiment. Early measurements on YbBr₃ found a complex zero-field spectrum [34], while studies of YbCl₃ discussed one-magnon physics [35] and sharp features in the scattering

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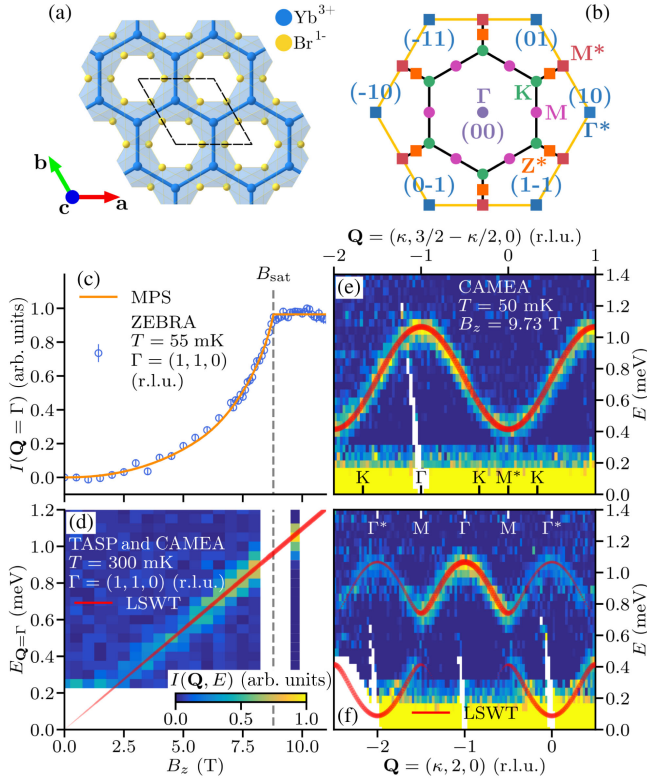


FIG. 1. Crystal structure and interaction parameters. (a) Crystal structure of a single YbBr_3 layer. Edge-sharing Br^- octahedra surround the magnetic Yb^{3+} ions. (b) Reciprocal space for the honeycomb lattice, showing the BZ (black hexagon) and irreducible BZ (orange hexagon). (c) Complete magnetization response deduced from neutron diffraction measurements of the ordered moment at $T = 55$ mK. The orange line is the result calculated by MPS. (d) Measurements at the Γ point showing the Larmor-mode frequency, $\hbar\omega_L = g_{zz}\mu_B B_z$. The red line is the result calculated by linear spin-wave theory (LSWT). The vertical dashed line marks the deduced saturation field, $B_{\text{sat}} = 8.78(2)$ T. (e, f) INS spectra measured in the fully polarized phase, at $B_z = 9.73$ T, along the two high-symmetry directions in the BZ. Red lines indicate the optimal fit by LSWT used to extract the interaction parameters. The widths of the LSWT lines are proportional to the calculated intensities.

continua ascribed to two noninteracting magnons [36], but achieved only a partial account of the observed spectra.

In this Letter, we report our comprehensive investigation of the honeycomb Heisenberg antiferromagnet (HHAF) YbBr_3 by inelastic neutron scattering (INS) experiments and cylinder matrix-product-state (MPS) calculations of the spectral function. We find excellent quantitative agreement between the measured INS data and an MPS treatment of the nearest-neighbor HHAF for all applied magnetic fields up to saturation. At finite fields, we find four remarkable phenomena: the field-induced restoration of a magnonlike excitation from the continuum scattering at the K point of the Brillouin zone (BZ); the almost complete field-induced destruction of magnonlike excitations close to the critical

field, caused by spectral-weight transfer into broad continua; an additional excitation at low fields that shadows the one-magnon modes; a nonmonotonic one-magnon dispersion reminiscent of the roton in superfluid helium. Having benchmarked the complete spectral function, we discuss the candidate theoretical descriptions accounting for the coexistence of the magnonlike excitations and complex continua we observe.

Material, experiments, and methods—For our study, we used the high-quality single-crystal samples of YbBr_3 [Fig. 1(a)] measured at zero field in Ref. [34], whose growth and structurally 2D character we summarize in Sec. S1 of the Supplemental Material (SM) [37]. We performed unpolarized INS experiments to measure the magnetic excitations at all fields on the cold-neutron triple-axis spectrometer (TAS) TASP and the multiplexing spectrometer CAMEA [56] at the Swiss Spallation Neutron Source, SINQ, at the Paul Scherrer Institut (PSI), as well as polarized INS on the TAS IN12 at the Institut Laue Langevin (ILL) [57]; these measurements and the accompanying data analysis are detailed in Sec. S2 of the SM [37]. The low energy scale of the magnetic interactions in YbBr_3 demands precision measurements at sub-Kelvin temperatures in superconducting cryomagnets. However, it also allows experimental access to the saturation field, which we determined to high accuracy by measuring the intensity of the $\mathbf{Q} = \Gamma$ reflection as a function of magnetic field applied along the crystalline c axis on the thermal neutron diffractometer ZEBRA (PSI), as shown in Fig. 1(c), obtaining the result $B_{\text{sat}} = 8.78(2)$ T.

To interpret our INS spectra, we performed extensive calculations by the MPS method, which provides near-unbiased spectral functions for 2D systems wrapped on a cylinder [58–60]. Section S4 of the SM [37] explains the application and benchmarking of cylinder MPS for the HHAF, extraction of the dynamical structure factor from MPS correlation functions, and resolution convolution using TAKIN [61], by which we achieved a fully quantitative comparison of INS and MPS.

Fully polarized phase—All quantum fluctuations are suppressed in the fully polarized phase above B_{sat} . Any spin-flip excitation is then described exactly by linear spin-wave theory (LSWT), whose implementation we summarize in Sec. S3 of the SM [37]. This makes it possible to determine very precisely the microscopic interaction parameters that govern the low-energy physics of YbBr_3 at all fields (taking magnetostriction effects in octahedrally coordinated Yb^{3+} materials to be negligible below 10 T [62]). We remark that any Heisenberg magnet has an excitation at $\mathbf{Q} = \Gamma$ whose energy ($\hbar\omega_L$) rises linearly with the field [63]. As Fig. 1(d) shows, this Larmor mode, or precession mode, is clearly visible and very accurately linear in YbBr_3 , providing no evidence for magnetostriction or non-Heisenberg terms in the spin Hamiltonian.

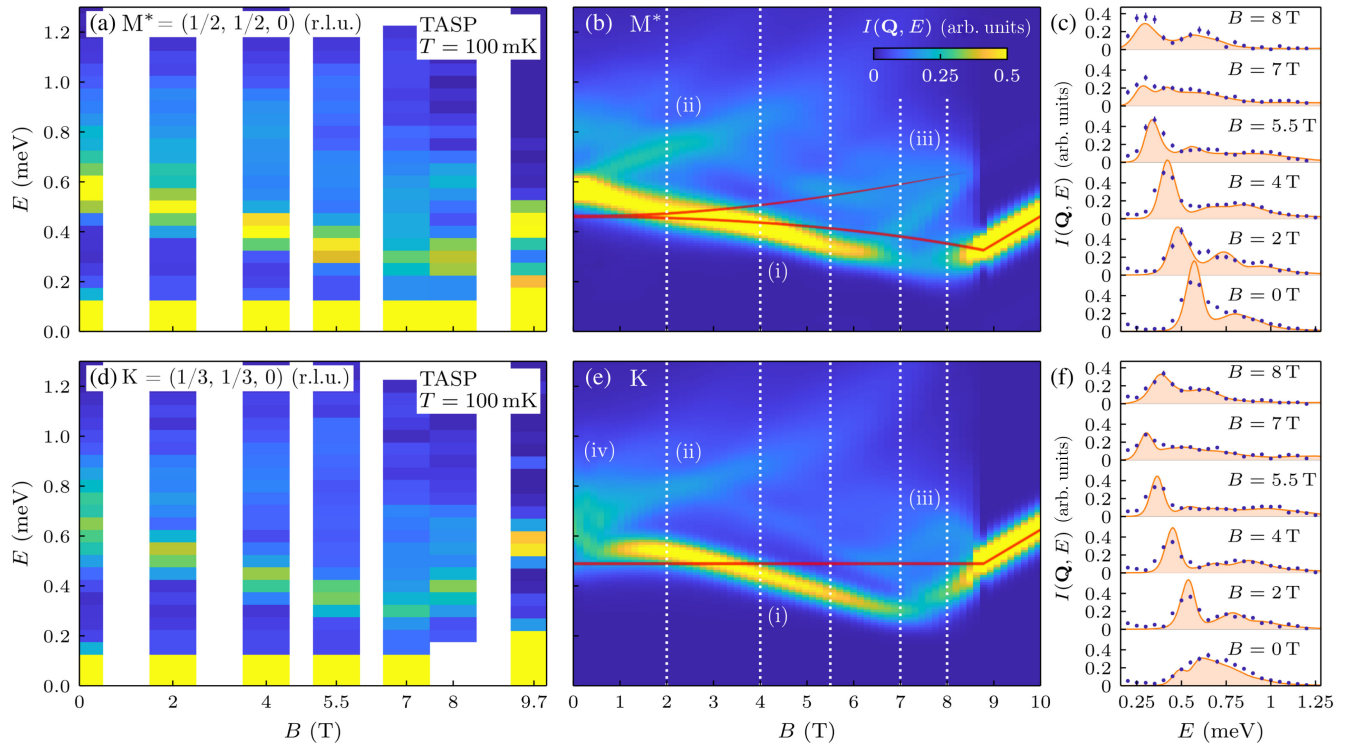


FIG. 2. Field dependence of magnetic excitations. (a) INS measurements and (b) MPS calculations of the field-induced spectral function at the M^* point. (c) Quantitative comparison of INS and MPS at M^* for all fields applied in experiment. (d) INS measurements and (e) MPS calculations at the K point. (f) Comparison of INS and MPS at K. Red lines in (b) and (e) show the magnon branches of LSWT. The labels (i)–(iv) in (b) and (e) refer to the specific excitation features discussed in the text.

Working above B_{sat} , in a field $B_z = 9.73$ T applied perpendicular to the honeycomb plane, we measured the magnon dispersion at $T = 50$ mK along the two high-symmetry directions of the BZ as shown in Figs. 1(e) and 1(f). Neglecting single-ion anisotropies for an $S_{\text{eff}} = 1/2$ system, we fit these data to a J_1 - J_2 - J_3 - Δ model with Heisenberg interactions between the first three near-neighbor sites and an additive XXZ anisotropy on the J_1 bond [making the full z -component term $\sum_{\langle i,j \rangle} (J_1 + \Delta) \hat{S}_i^z \hat{S}_j^z$]. The optimal values of J_2 , J_3 , and Δ are all zero within the precision afforded by the INS data, as we show in detail in Sec. S3 of the SM [37]. An appropriate minimal model to describe the measured dispersion is therefore the pure nearest-neighbor (NN) HHAF Hamiltonian,

$$\hat{\mathcal{H}} = J_1 \sum_{\langle i,j \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j - g_{zz} \mu_B B_z \sum_i \hat{S}_i^z, \quad (1)$$

with $J_1 = 0.326(22)$ meV, where the g -tensor component $g_{zz} = 1.89(3)$ is obtained from the gradient of the Larmor mode [Fig. 1(d)]; the LSWT dispersion obtained from this model, marked by the red lines in Figs. 1(e) and 1(f), indeed fits extremely well. The model we deduce is consistent with the prediction of $S_{\text{eff}} = 1/2$ character, minimal anisotropy (other than the g tensor), and predominant antiferromagnetic

Heisenberg superexchange in Yb^{3+} magnets with edge-sharing octahedral coordination made in Ref. [64].

Intermediate-field spectra—We proceed to the regime at all finite fields up to saturation, focusing in Fig. 2 on the M^* and K points. First, at the descriptive level, we draw attention to four field-induced phenomena that are visible in experiment [Figs. 2(a) and 2(d)] and can be studied both qualitatively [Figs. 2(b) and 2(e)] and quantitatively [Figs. 2(c) and 2(f)] by MPS. (i) There is a significant renormalization of the one-magnon branch compared to LSWT, which is strongly downward with increasing field. (ii) The clearest scattering continuum is a broadened shadow of the one-magnon branch that rises linearly above it with the applied field at $B < \frac{2}{3} B_{\text{sat}}$ and, in fact, appears at all $\mathbf{Q}$ [36]. (iii) The second-strongest continuum is a low-energy feature that develops at higher fields ($B > \frac{1}{2} B_{\text{sat}}$) and seems to drain weight away from the one-magnon branch. Continua (ii) and (iii) are more complex than the two-magnon continuum obtained from SWT [36], which corresponds to two energetically renormalized (at order $1/S$) but noninteracting magnons, and our results at fields $B > \frac{1}{2} B_{\text{sat}}$ reveal significantly more insight into interaction effects. (iv) At the K point, we observe a complete decay of the one-magnon branch as the field is reduced below 1.5 T [Figs. 2(d)–2(f)], and below we discuss the apparent zero-field continuum that results;

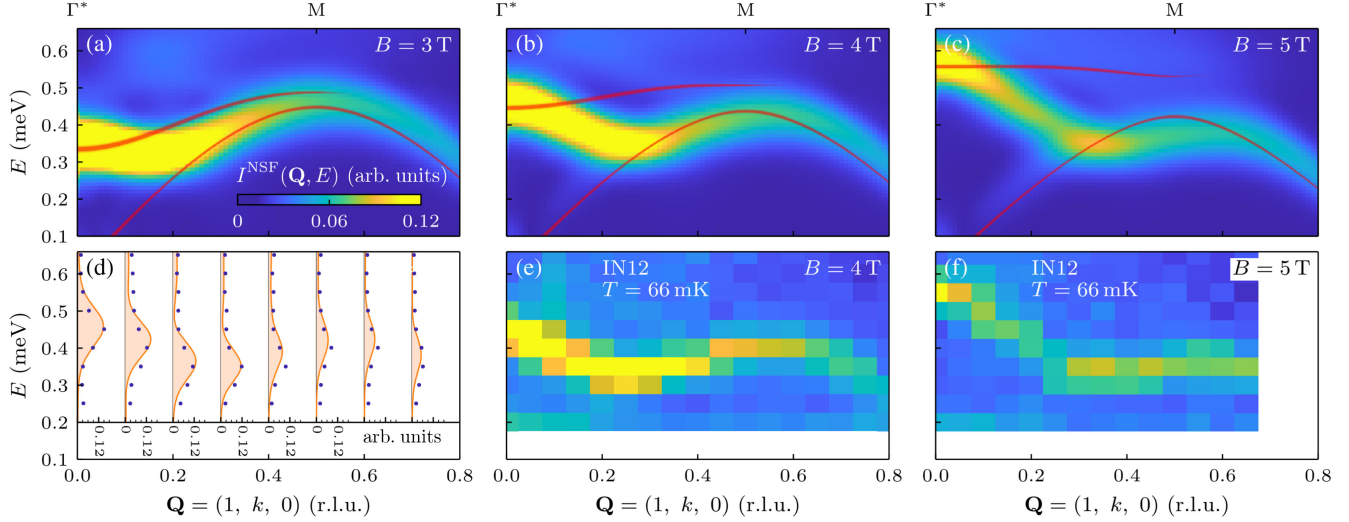


FIG. 3. Rotonlike excitation. (a)–(c) MPS calculations of the longitudinal (non-spin-flip) spectral response, $I^{\text{NSF}}(\mathbf{Q}, E)$, along the Γ^* -M direction for fields of 3, 4, and 5 T, showing the field-induced evolution of a rotonlike feature. Red lines show the magnon branches within LSWT. (d) Constant- $|\mathbf{Q}|$ cuts through the INS and MPS spectral functions at 4 T for eight wave vectors. Error bars on the INS data are smaller than the symbol sizes. (e), (f) Intensity in the NSF channel measured by polarized INS at $T = 66$ mK, showing the rotonlike mode at 4 and 5 T.

we remark that the energy of the one-magnon branch at M^* also shows a strong upward trend from 1.5 to 0 T [Figs. 2(a) and 2(b)]. We stress again that the quantitative agreement between INS and MPS in Fig. 2 is achieved with a single scale factor for the intensity (Sec. S4 of the SM [37]), confirming qualitatively (1) that the NN HHAF is the correct model for YbBr_3 and (2) that all of the observed features are intrinsic to the HHAF.

Although the field-induced downward renormalization of the one-magnon branch at M^* and K (i) is captured qualitatively at the $1/S$ level [36], our demonstration of continuum scattering at all higher energies is consistent with the picture [65] of a strong mutual repulsion between this branch and the developing continua, in turn, suggesting strong magnon interactions and a multimagnon origin for the latter. However, the dominant effect of these continua, particularly continuum (iii), is the near-complete disappearance of the single magnon branch at fields approaching B_{sat} . While this magnon decay could be regarded as a diverging width or as a field-induced fractionalization, its progressive nature and the energetic separation of the magnon from continuum (iii) reinforce the picture of strong two-magnon interactions [65], rather than the abrupt decay at a well-defined two-magnon continuum boundary [7,31] characteristic of noninteracting magnons.

Turning to the low-field continuum (ii), it is easy to see that its energy offset from the one-magnon branch is the Larmor energy $\hbar\omega_L$, which explains its linear rise with the field (Fig. 2). This suggests an interpretation as a collective two-magnon state formed from a single magnon at $\mathbf{Q}$ and a Larmor mode at Γ . We remark that this collective state is remarkably sharp at all wave vectors in the SLHAF, where it merits the name “shadow mode” [66]. In the HHAF we use

the term “magnon shadow” because both our MPS and INS measurements on YbCl_3 [36] show that it is considerably broader at most $\mathbf{Q}$ points, illustrating significant differences between these two paradigm unfrustrated quantum spin models. The continuum of two noninteracting magnons [36] captures the position of the shadow qualitatively, but finds intensities that are more diffuse in energy and are dominated by van Hove points that both INS and our MPS results show to be artificial over a wider field range. We note here that the field-controlled spectrum changes very rapidly as the field is reduced below B_{sat} , reflecting a very low energy scale associated with the linear part of an otherwise quadratic magnon dispersion, and hence a very low threshold to form bound and scattering states [continuum (iii)] contributing to magnon decay in this regime.

Zero field—The zero-field spectrum was measured by Wessler *et al.* [34], and we replot these data in Fig. 4 of the End Matter for comparison with our calculated spectral function. The MPS results quantify the extent of continuum formation on the zone boundary seen in INS, which progresses from a robust magnon branch with only weak high-energy continuum scattering at M to a dominant continuum with minimal magnon intensity at K [Figs. 4(b)–4(e)].

Rotonlike mode—We close by considering the $\mathbf{Q}$ dependence of the spectrum in more detail. The intensity measured by unpolarized INS is a sum of transverse and longitudinal spin excitation channels, which we define relative to the field direction ($\mathbf{z}$) and can separate readily in our MPS calculations. In this way, we noticed highly unconventional behavior of the longitudinal spectral weight at intermediate fields that is reminiscent of the roton in liquid ^4He [67], with a distinctive dip of the mode energy at

small but finite $|\mathbf{Q}|$. As Figs. 3(a)–3(c) show, this mode involves an abrupt transfer of spectral weight between the two one-magnon branches of LSWT to produce a sharp magnonlike mode in a part of reciprocal space absolutely not predicted by SWT.

Motivated by this observation, we performed a polarized INS experiment on the TAS spectrometer IN12 at the ILL, as detailed in Sec. S2 of the SM [37]. Equating the longitudinal response directly with the non-spin-flip (NSF) channel, the measured spectra [Figs. 3(e) and 3(f)] find the “honeycomb roton” to be a resolution-limited mode that agrees quantitatively in both position and intensity with the MPS predictions over a range of fields [Fig. 3(d)]. Here we have already identified the mutual repulsion of the magnon branch and higher-lying continua, and in this context the field-dependent rotonic minimum in Figs. 3(a)–3(c) corresponds well to the changing $\mathbf{Q}$ position of the minimum in the nearby continuum. Thus, the honeycomb and liquid-He rotons share the property that both constitute collective behavior emerging from strong multiparticle interactions that cause strong mutual renormalization of both the lower branch and the higher continuum. We note that this rotonlike behavior is also visible in the 4 T INS data on YbCl_3 [36], but not in the spectrum obtained by nonlinear SWT, pointing again to the effects of strong magnon-magnon interactions that are captured by our MPS calculations.

Discussion—We reiterate that our INS measurements and MPS calculations show full quantitative equivalence over the entire range of $\mathbf{Q}$, E , and B , confirming both the model and the spectral function. Thus, our results reveal multiple phenomena in the HHAF spectrum which are hallmarks of intrinsic many-body interactions that are manifestly strong, although they are not of the type required to produce a QSL. Over most of the BZ and field range, we find well-defined magnonic modes, albeit with significant energetic renormalization that appears to be a consequence of mutual repulsion from extensive higher-lying continua (Fig. 2) [65]. The exception is around the K point at low fields, where it is hard to argue for a true quasiparticle [Figs. 2(e) and 4(f)]. As in the SLHAF at $(\pi, 0)$, one anticipates an interpretation of this behavior in terms of multimagnon states [11,12] or of magnon fractionalization [16–18]. Two-magnon binding has been found in the square lattice in a field [66] and a further example of multimagnon resonances would be the correlated plaquette fluctuations proposed in Ref. [34]. Fractional quasiparticles are expected around deconfined quantum critical points (DQCPs), and extensive studies of the frustrated HHAF [19–27] show that J_2 causes the spectrum to soften at K, where Néel order gives way to a plaquette (hexagon) valence-bond crystal at a DQCP. Possible fingerprints of this deconfinement have been reported [32,33] at finite energies even for $J_2 = 0$, and our demonstration that a

magnonic excitation forms above 1.5 T [Fig. 2(e)] would be consistent with a field-induced spinon reconfinement.

One of our signature findings is field-induced magnon decay, which we show to be a progressive phenomenon following the strong-interaction scenario [65], where spectral weight shifts systematically to higher-lying continua with no sharp onset in field or wave vector. In fact, the magnons never disappear completely, even if the vast majority of their weight has moved to higher energies near saturation (Fig. 2). In this context, the coherent collective modes we observe come with a caveat: the magnon shadow forming at low fields (Fig. 2, [36]) still has continuum rather than single-mode character, not least because the magnons themselves become broadened entities near the BZ boundary. Where the magnon branches are largely conventional, in the inner BZ and at lower fields, then we find a highly unconventional shift of spectral weight between them, and this is the rotonlike excitation (Fig. 3).

Conclusions—We have measured and computed the spectral function of the honeycomb antiferromagnet YbBr_3 at all fields from zero to saturation. We use the spectra in the fully polarized phase to establish that the spin Hamiltonian is of pure Heisenberg and nearest-neighbor form. We find a rich and complex field-induced spectral response that deviates from spin-wave predictions in many regards. At low energies, magnonic modes appear at unexpected positions, influenced by repulsive interactions with nearby excitation continua. At low fields, quantum fluctuations destroy the magnon around the K point. At higher fields, the magnons are broadened by their interactions to the threshold of complete decay. However, interaction effects are not only destructive, but also create coherent features over significant regions of the spectrum, including collective magnon shadows and rotonlike modes. We conclude that a wealth of interacting many-body phenomena remain to be understood in the complete field-induced spectral functions of quantum magnetic systems, even in the absence of frustration.

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Data availability—The data that support the findings of this Letter are openly available [68], embargo periods may apply.

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End Matter

Zero-field spectrum—In Fig. 4(a), we replot the INS data of Ref. [34] to analyze the previously reported tendency that magnonlike excitations in the inner BZ evolve into excitation continua near the BZ boundary. Our MPS calculations reflect the maximal quantum entanglement at zero field through some persisting weak dependence on the cylinder size and bond dimension, although these differences vanish when the spectrum is convolved with the instrumental resolution (Sec. S4 of the SM [37]). As noted in the main text, at M we find a rather well-defined magnon accompanied by only weak continuum scattering at higher energies [Fig. 4(b)]. At M^* , the magnonic peak remains and the continuum is slightly stronger [Fig. 4(c)]. On proceeding toward K, there is a progressive weakening of the magnon and strengthening of the continuum [Fig. 4(d)], but only at the K point do we find a near-complete loss of sharp magnon intensity [Fig. 4(e)]. The MPS results make clear that this continuumlike K-point spectral function, proposed [36] as the honeycomb analog of the $(\pi, 0)$ anomaly in the SLHAF [8–10], is an intrinsic quantum many-body property of the HHAF, as are the continua lying above the putative one-magnon branch for all of the zone-boundary points [Figs. 4(b)–4(d)]. We remark that the value of J_1 extracted from our high-field data continues to provide an optimal fit at zero field.

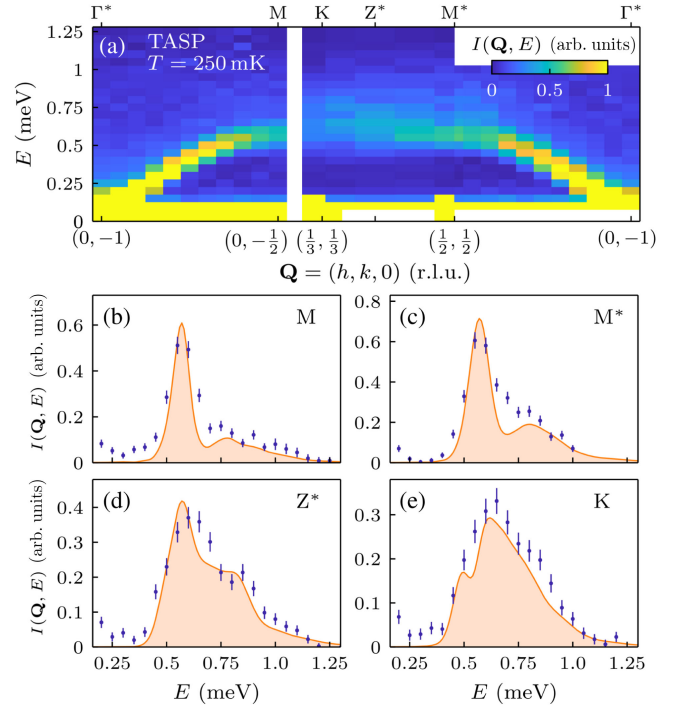


FIG. 4. Zero-field spectrum. (a) INS intensity measured along the paths Γ^* -M and K - M^* - Γ^* by INS. (b)–(e) Quantitative comparison of INS (symbols) and MPS results (beige shading) for constant- $\mathbf{Q}$ spectra measured and computed at (b) M, (c) M^* , (d) Z^* , and (e) K.