

Observation of Bethe strings in the quantum spin chain antiferromagnet $\text{BaCo}_2\text{V}_2\text{O}_8$

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Despite being predicted almost a century ago as the excitations of the spin-1/2 antiferromagnetic chain, Bethe strings were overshadowed by the more dominant multi-spinon-pair excitations and were only observed recently. Here we confirm their existence in the quasi-one-dimensional quantum antiferromagnet $\text{BaCo}_2\text{V}_2\text{O}_8$ using a magnetic field to separate them from the spinons. We find excellent quantitative agreement over an extended field range between these experimental results and our theoretical predictions based on the algebraic Bethe ansatz formalism. This work proves the existence of Bethe strings (multimagnon bound states) up to length-3 in $\text{BaCo}_2\text{V}_2\text{O}_8$.

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I. INTRODUCTION

The quasi-one-dimensional (quasi-1D) spin-1/2 Heisenberg antiferromagnet with nearest neighbor exchange interactions has long been studied as a paradigmatic system for the exploration of exotic physics such as spinon excitations [1–5], quantum critical scaling [6,7], and quantum entanglement [8]. As fractional excitations carrying spin-1/2 [9,10], spinon excitations are well established by both theory [11,12] and inelastic neutron scattering (INS) experiments [4,5] to explain the dynamical spin structure factors of the 1D spin-1/2 Heisenberg antiferromagnet at zero field.

However, the spinon picture fails when a magnetic field is applied, while the Bethe ansatz formalism identifies the excitations regardless of the presence of the magnetic field, such as psinon-psinon pair, psinon-antipsinon pair, and Bethe strings [13,14]. Among these excitations, Bethe strings, as bound states of magnons, were predicted in the spin-1/2 antiferromagnetic (AFM) Heisenberg chain long ago by Bethe [15]. As characteristic excitations of the AFM Heisenberg chain, Bethe strings can still play a crucial role in a Heisenberg chain with integrability breaking terms [16]. Although their existence was confirmed theoretically, they were only observed very

recently [17–20], as explored, for example, in the Heisenberg ($\Delta = 1$) AFM spin-1/2 chain YbAlO_3 [21]. Strings up to length-2 were also observed in CsCoCl_3 , which realizes an AFM spin-1/2 chain with very large Ising anisotropy ($\Delta = 6.7$) [22]. ESR measurements showed that they evolve into two-magnon states above the saturation field.

The difficulty in detecting Bethe strings is due to their weak intensity compared to the spinon continuum and the fact that they overlap with it. These problems were overcome in the compound $\text{SrCo}_2\text{V}_2\text{O}_8$ (SCVO), which realizes the spin-1/2 AFM XXZ chain with Ising anisotropy [23–26]. By applying a longitudinal magnetic field (along the easy axis) to separate the Bethe strings from the spinons excitations in energy, they became observable via terahertz (THz) spectroscopy [18] and INS experiments [19]. In this paper we observe Bethe strings up to length-3 [27] using INS in a second compound, namely $\text{BaCo}_2\text{V}_2\text{O}_8$ (BCVO). The accurate agreement with theoretical prediction based on algebraic Bethe ansatz (ABA) formalism [14] provides further confirmation for the existence of these exotic states.

Similar to SCVO and $\text{PbCo}_2\text{V}_2\text{O}_8$ [28], BCVO is a good example of the spin-1/2 AFM XXZ chain and has attracted considerable attention for hosting exotic magnetism [29–44]. BCVO crystallizes in the tetragonal space group $I4_1/acd$ (no. 142), with lattice parameters $a = b = 12.444 \text{ \AA}$ and $c = 8.415 \text{ \AA}$ at $T = 2.0 \text{ K}$ [30]. The magnetic Co^{2+} ions (effective spin-1/2) are arranged in edge-sharing CoO_6 octahedra that form fourfold screw chains running along the tetragonal c axis. There are four screw chains in each unit cell, with two rotating clockwise and the other two counterclockwise. Neighboring Co^{2+} ions are coupled by strong

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nearest neighbor AFM interactions within each chain, which have partial Ising anisotropy along the $\mathbf{c}$ axis. The chains are separated by nonmagnetic V^{5+} and Ba^{2+} ions allowing for only weak interchain interactions, which helps to stabilize 3D AFM long-range order (LRO) below the Néel temperature $T_N = 5.5$ K [32]. The magnetic structure is a Néel AFM (NAF) order along the screw chains with the spins pointing parallel to the $\mathbf{c}$ axis (easy axis) [37]. The gapped spinon continuum was observed in the spectrum for temperatures above T_N , indicating the presence of free spinons. Below T_N this broad continuous spectrum is replaced at low energy by sharp modes, indicating that the spinons are confined into pairs with integer spin due to the mean field of the 3D NAF order [33].

Bethe string excitations are not detectable via INS measurements on $\text{BaCo}_2\text{V}_2\text{O}_8$ at 0 T. Indications for the string states were however found in THz spectroscopy in a small longitudinal magnetic field of $4.5 \leq \mu_0 H \leq 7$ T applied parallel to the easy axis ($\mathbf{c}$ axis) [20]. The goal of this work is to use a combined experimental and theoretical approach to establish the existence of Bethe strings up to length-3 in BCVO using longitudinal magnetic fields as strong as $\mu_0 H = 14.9$ T to separate them from the spinons. BCVO is described by the Hamiltonian,

$$\mathcal{H} = J \sum_{i,\mu} [\Delta S_{i,\mu}^z S_{i+1,\mu}^z + (S_{i,\mu}^x S_{i+1,\mu}^x + S_{i,\mu}^y S_{i+1,\mu}^y)] + J' \sum_i \sum_{\langle \mu, \nu \rangle} \mathbf{S}_{i,\mu} \cdot \mathbf{S}_{i,\nu} - g \mu_B \mu_0 H \sum_{i,\mu} S_{i,\mu}^z, \quad (1)$$

where $\mathbf{S}_{i,\mu} = (S_{i,\mu}^x, S_{i,\mu}^y, S_{i,\mu}^z)$ are the spin operators for the i th site of the μ th chain. For BCVO the intrachain coupling, interchain coupling, Ising anisotropy, and Landé factor are $J = 3.07$ meV, $J' = 0.17$ meV, $\Delta = 1.9$, and $g = 6.07$ [43,44], respectively, while μ_B is the Bohr magneton and $\mu_0 H$ is the longitudinal magnetic field along the $\mathbf{c}$ axis. The NAF order is suppressed by the longitudinal field and disappears above the critical field of $\mu_0 H^c = 3.8$ T at zero temperature [45]. At higher fields in the absence of interchain coupling, a gapless quantum critical state known as the Tomonaga-Luttinger liquid (TLL) is predicted for the XXZ chain over a large field range. Although the weak interchain couplings in BCVO induce a series of long-range ordered phases below $T \approx 1$ K for $\mu_0 H > \mu_0 H^c$ [35,43], at $T = 1.5$ K where our experiments were performed BCVO is expected to behave as a critical TLL [43,44,46].

II. EXPERIMENTAL DETAILS

For this investigation, a single crystal of BCVO with mass 4.13 g was grown using the floating-zone method at the Core Lab Quantum Materials, Helmholtz Zentrum Berlin. INS measurements were performed using the triple-axis spectrometer ThALES at the Institut Laue-Langevin. The sample was aligned with the $(H, K, 0)$ plane in the horizontal scattering plane and a vertical cryomagnet capable of producing a field of up to 14.9 T along the out-of-plane $\mathbf{c}$ axis was used. All data were collected at a fixed final wave vector of $k_f = 1.50 \text{ \AA}^{-1}$ giving an energy resolution of $\Delta E = 0.17$ meV. Energy scans were performed at the two different scattering wave vectors $Q_1 = (3.25, 0, 0)$ and $Q_2 = (2.3, 2.3, 0)$. Both wave vectors

have the same excitations since they have the same wave vector along the chain of $q_l = 0$. However, the relative intensities of the modes are different (due to the screw chain structure of the cobalt chains). Thus by combining measurements at both wave vectors the modes could be measured more accurately. The sample was cooled to $T = 1.5$ K just above the LRO phases to be in the TLL regime. Energy scans were performed over the energy range $0 \leq E \leq 15.3$ meV in steps of 0.10 meV for different applied magnetic fields up to 14.9 T [47].

III. RESULTS

The data is summarized in Fig. 1, where the individual energy scans for $\mu_0 H \leq 14.9$ T are shown in panels (c) and (e) for wave vectors Q_1 and Q_2 , respectively. The data are combined into color maps of energy transfer versus magnetic field for these two wave vectors in panels (a) and (b), respectively [the data in the panels (a) and (b) are raw data and the data sets in the panels (c) and (e) have the offsets of 100 a.u.]. These maps reveal that a sudden change takes place at $\mu_0 H^c$ (indicated by the vertical white dotted line) where the NAF order is destroyed and the TLL state is reached. In this phase various modes are observed with distinct linear field dependences. The peaks in the energy scans for $\mu_0 H > \mu_0 H^c$ [Figs. 1(c) and 1(e)] were fitted by the sum of Gaussians to determine the energy of the modes. Their linear dependences are emphasized by the black dashed lines. For Q_1 a weak mode was observed at high energies ($E > 8$ meV), which is highlighted in the panel on the right-hand side of Fig. 1(c). The extracted peak positions are added to the energy transfer versus magnetic field maps [Figs. 1(a) and 1(b)] as filled colored circles to emphasize the field dependences.

Our theoretical calculations were performed for the spin-1/2, AFM, XXZ chain in a longitudinal magnetic field using the ABA formalism [14]. The dynamical spin structure factors relevant to INS were calculated with anisotropy parameter $\Delta = 1.9$ for the transverse (D^{+-}, D^{+}) and longitudinal (D^z) polarizations as a function of energy transfer versus wave vector q_l along the chain. An energy resolution of 0.15 meV was used to match the experimental resolution of 0.17 meV. Various types of excitations were identified, such as the psinon-psinon pair (R^{PP}) and psinon-antipsinon pair (R^{PAP}), which are gapless at commensurate or incommensurate wave vectors and related to the spinon-pair continuum observed in zero field. The length-2 ($\chi^{(2)}$) and length-3 ($\chi^{(3)}$) Bethe string states were also identified, which are gapped throughout the Brillouin zone and lie mostly at higher energies than the psinon excitations.

The energies of the excitations were extracted as a function of longitudinal field at wave vectors along the chain of $q_l = 0, \pi/2, \pi$ for $J = 3.07$ meV and $g = 6.07$. For moderate field strengths, the 2- and 3-string excitations are well separated in energy from the psinon-type excitations at certain wave vectors, with the 3-string lying at a higher energy than the 2-string, allowing them to be distinguished. Moreover, these calculations show that for longitudinal fields greater than the critical field $\mu_0 H^c$, the individual excitations have distinct, linear field dependences of the form $E = \beta \mu_0 H + \alpha$, which can be used to identify them.

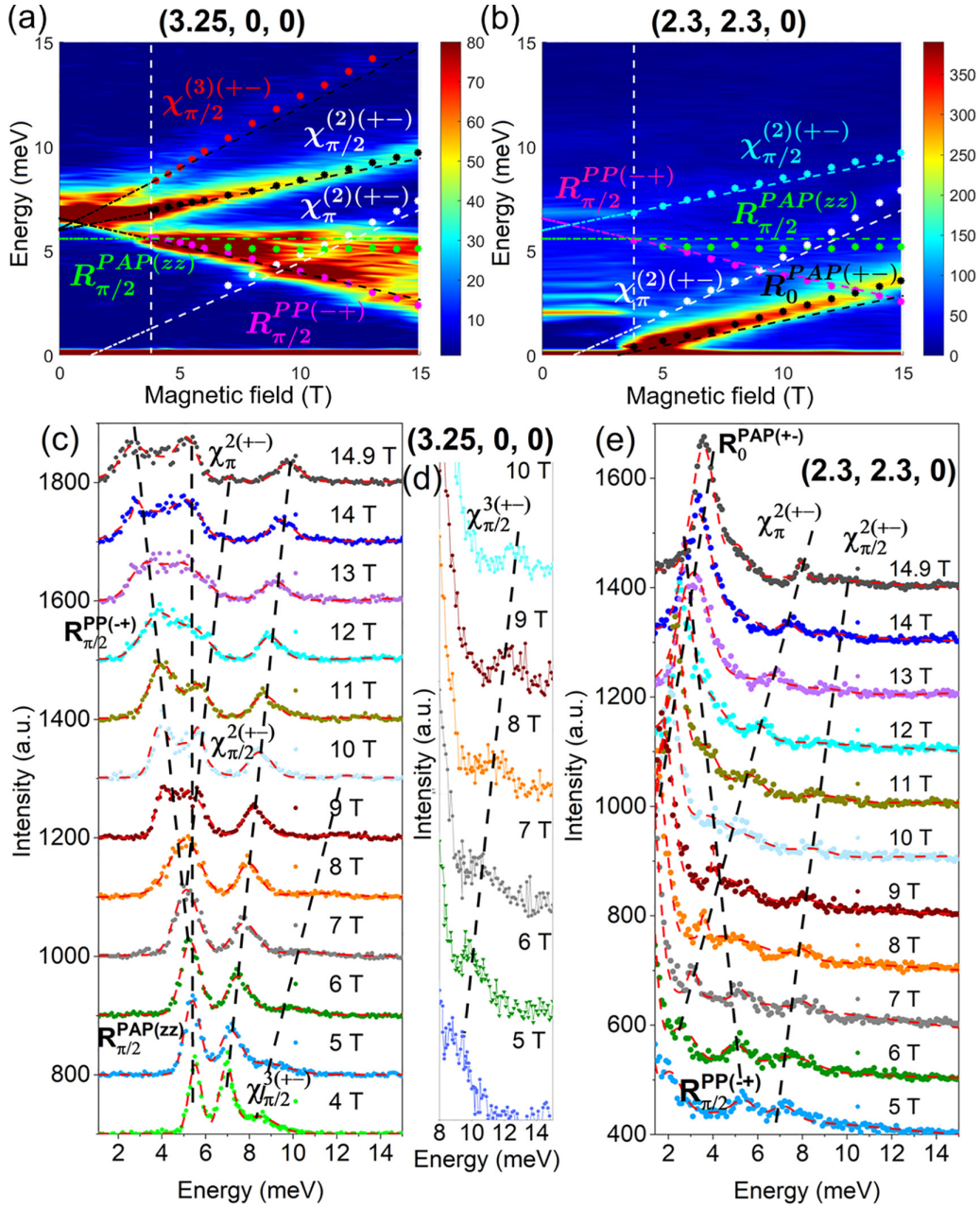


FIG. 1. Field dependence of magnetic excitations in BCVO under longitudinal applied field at $T = 1.50$ K. Energy transfer versus field color maps are shown for INS measurements at (a) $Q_1 = (3.25, 0, 0)$ and (b) $Q_2 = (2.3, 2.3, 0)$. The critical field $\mu_0 H^c = 3.8$ T is indicated by the vertical dashed white line. At lower fields the bound-spinon modes are observed at Q_2 . For $\mu_0 H > \mu_0 H^c$, the position of the peaks extracted by fitting the individual energy scans to Gaussians are indicated by the colored dots, while the tilted dashed lines show the predictions of the ABA formalism for the various excitations labeled $\chi_{\pi/2}^{(3)(+-)}$, $\chi_{\pi/2}^{(2)(+-)}$, $\chi_{\pi/2}^{(2)(+-)}$, $R_{\pi/2}^{PAP(zz)}$, $R_0^{PAP(+)}$, and $R_{\pi/2}^{PP(-+)}$. Selected energy scans for $\mu_0 H^c < \mu_0 H < 14.9$ T are shown for (c) Q_1 and (e) Q_2 together with Gaussian fits (red dashed lines). The sloping black dashed lines emphasize the observed linear field dependences. Panel (d) is a plot of the scans from 5 to 10 T rescaled to highlight the weak $\chi_{\pi/2}^{(3)}$ mode.

The slopes β and offsets α of the theoretical field dependences of the various excitations can be determined by linear fitting. These fits are shown over the experimental data in Figs. 1(a) and 1(b) by the colored dashed lines. In BCVO where the chains are fourfold screw chains, zone-folding effects mean that there are four copies of the spectra each shifted by a reciprocal lattice unit; thus an INS measurement at $q_l = 0$ also records the spectrum at $q_l = \pi/2, \pi$, and $3\pi/2$.

Therefore, the field dependences for all these wave vectors are included and are labeled according to type of mode, wave vector, and polarization, e.g., $\chi_{\pi/2}^{(2)(+-)}$ is the 2-string excitation at $q_l = \pi/2$ observed in the D^{+-} structure factor. The theoretical slopes and offsets for BCVO are listed in the fourth row of Table I.

Figures 1(a) and 1(b) show that the theoretical field dependences of the modes are in good agreement with the

TABLE I. Slopes $\beta = \frac{dE}{d(\mu_0 H)}$ (meV/T) and offsets α (meV) from the linear fits ($E = \beta\mu_0 H + \alpha$) of the field dependence of the various excitations listed in the first row. Experimental values for BCVO from the INS data presented here fitted over 4 to 14.9 T (second row) and THz spectroscopy over 5 to 7 T [20] (third row) are compared to the values predicted by the ABA formalism calculated for $\Delta = 1.9$ with the parameters $J = 3.07$ meV and $g = 6.07$. The field dependences for SCVO from the INS data [19] and ABA calculations [14] are given in the fourth and fifth rows, respectively. The parameters for SCVO are $\Delta = 2$, $J = 3.55$ meV, and $g = 5.50$. The error bars of the fitted slopes are given in the third row; they are all <0.01 meV/T.

Field dependence (β, α)	$\chi_{\pi/2}^{(3)(+-)}$	$\chi_{\pi/2}^{(2)(+-)}$	$R_{\pi/2}^{(PAP)(zz)}$	$R_{\pi/2}^{(PP)(-+)}$	$\chi_{\pi}^{(2)(+-)}$	$R_0^{(PAP)(+-)}$
BCVO Expt. INS	(0.65, 5.80)	(0.25, 5.95)	(−0.01, 5.20)	(−0.26, 6.48)	(0.59, −1.09)	(0.29, −0.84)
Error bars of BCVO Expt. INS	(0.007, 0.06)	(0.002, 0.02)	(0.003, 0.03)	(0.004, 0.04)	(0.009, 0.10)	(0.003, 0.03)
BCVO Expt. THz and ESR [20,48]	(0.44, 6.59)	(0.22, 6.08)		(−0.22, 6.19)	(0.53, −0.64)	(0.27, −0.76)
BCVO theory	(0.58, 6.06)	(0.23, 5.99)	(0.00, 5.58)	(−0.26, 6.65)	(0.51, −0.66)	(0.24, −0.75)
SCVO Expt. INS [19]	(0.60, 7.16)	(0.26, 6.97)	(0.00, 7.06)	(−0.30, 8.19)	(0.44, −0.18)	(0.27, −0.90)
Error bars of SCVO Expt. fitted (slopes)	0.02	0.01		0.02	0.05	0.01
SCVO theory [14]	(0.63, 7.27)	(0.27, 7.27)	(0.00, 6.40)	(−0.27, 7.27)	(0.50, 0.06)	(0.27, −1.24)
Error bars of SCVO theory (slopes)	0.03	0.01		0.01	0.02	0.01

features observed in the experimental data with the extracted peak positions from the neutron energy scans (colored circles) matching closely the colored lines from theory, allowing us to identify the excitations found in the data. We identify the psinon-psinon-pair excitation at $q_l = \pi/2$ in the D^{-+} structure factor ($R_{\pi/2}^{PP(-+)}$) whose energy decreases with increasing field. We also identify the psinon-antipsinon-pair excitation at $q_l = \pi/2$ in the D^{zz} structure factor ($R_{\pi/2}^{PAP(zz)}$), which is field independent, as well as at $q_l = 0$ in the D^{+-} structure factor ($R_0^{PAP(+-)}$), whose energy increases with field. Most interestingly, the Bethe string excitations could be identified. The 2-string excitation was observed at $q_l = \pi/2$ and $q_l = \pi$ in D^{+-} with increasing energy as a function of field ($\chi_{\pi/2}^{(2)(+-)}$ and $\chi_{\pi}^{(2)(+-)}$). The elusive 3-string was also observed at highest energies at $q_l = \pi/2$ in D^{+-} ($\chi_{\pi/2}^{(3)(+-)}$). Although weak, this excitation is clearly present as shown by the energy scans in Fig. 1(d).

Having identified the observed excitations, we linearly fit their field dependences to make a quantitative comparison with the theoretical predictions. The fits are shown in Fig. 2 by the solid lines along with the corresponding peak positions (symbols) and the theoretical field dependences (dashed lines). The fitted slopes and offsets are also listed in the second row of Table I alongside the theoretical predictions (fourth row). Excellent quantitative agreement between the experiment and theory is found for all excitations. Small systematic shifts between the experimental and calculated energies could be related to the presence of weak interchain interactions in BCVO which influence the extracted values of Δ and J used in calculation, whereas the theory is based on a purely 1D model. Furthermore, the theory assumes a simple Ising anisotropy, whereas off-diagonal terms in the J tensor are possible for the Co^{2+} ion. The INS and theory are also in agreement with the THz spectroscopy data [20] and the ESR data [48] as shown in Fig. 2. It is important to mention that for both scattering wave vectors $Q_1 = (3.25, 0, 0)$ and $Q_2 = (2.3, 2.3, 0)$ no phonon modes are expected as the modes we observe are strongly field dependent, whereas phonons are not field dependent.

To make a comparison with the excitations previously observed in SCVO, including the 2- and 3-strings [18,19], the

values of experimental and theoretical slopes and offsets that fit the data in Ref. [19] are listed in the sixth and eighth rows of Table I. In this compound, the excitations were also observed with linear field dependences but at different energies due to differences in the values of anisotropy ($\Delta = 2.0$) as well as intrachain interaction ($J = 3.55$ meV) and g factor ($g = 5.5$). It is interesting to note that the agreement with theory for BCVO is better than for SCVO. This difference in agreement with theory for these two compounds may be due to the fact that the interchain exchange interaction (which

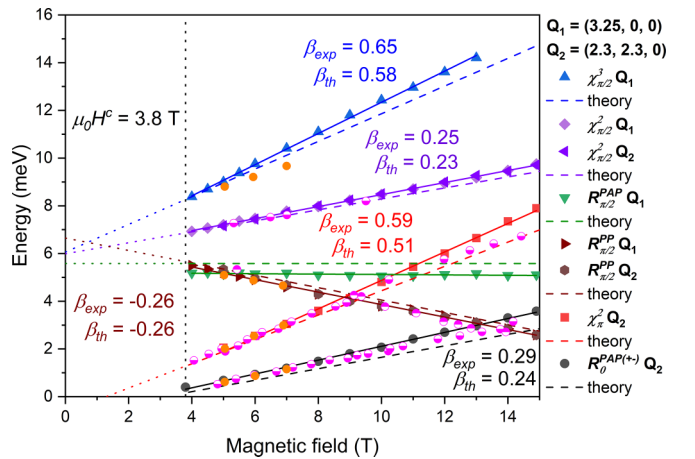


FIG. 2. Field dependence of the various magnetic excitations. The excitation energies extracted at $Q_1 = (3.25, 0, 0)$ and $Q_2 = (2.3, 2.3, 0)$ shown in Figs. 1(a) and 1(b), respectively, are combined and represented by the colored symbols. Their field dependences are linearly fitted over the field range 4–14.9 T (solid lines) and the fitted slope and offset values are listed in the second row of Table I. The theoretical field dependences are also shown (dashed lines) and are extrapolated to 0 T (dotted lines). The different observed excitations $R_{\pi/2}^{PP(zz)}$, $R_{\pi/2}^{PAP(zz)}$, $R_0^{PAP(+-)}$, $\chi_{\pi/2}^{(2)(+-)}$, $\chi_{\pi}^{(2)(+-)}$, and $\chi_{\pi/2}^{(3)(+-)}$ are represented by the colors brown, green, black, purple, red, and blue, respectively. The orange dots show the THz data for $R_0^{PAP(+-)}$, $\chi_{\pi}^{(2)(+-)}$, $R_{\pi/2}^{PP(-+)}$, and $\chi_{\pi/2}^{(3)(+-)}$ reported in [20] and the magenta dots show the ESR data for $R_{\pi/2}^{PP(-+)}$, $\chi_{\pi/2}^{(2)(+-)}$, $\chi_{\pi}^{(2)(+-)}$, and $R_0^{PAP(+-)}$ reported in [48].

will slightly shift the energy positions of some excitations) for BCVO is weaker than for SCVO. It should also be noted that, for both compounds, deviations from 1D physics due to selective interchain couplings are a cause of the enhancement of the spectral-weight intensity of the longitudinal (D^z) polarization [49].

IV. SUMMARY

To conclude, the spin dynamics of the spin-1/2 AFM XXZ chain compound BCVO was studied in a longitudinal field using a combination of experiment and theory and we identified a variety of spin excitations. Below the $\mu_0 H^c$ at $T = 1.5$ K, the spectrum is characterized by confined spinon pairs. Above $\mu_0 H^c$, the spectrum is governed by psinon-psinon and psinon-antipsinon pairs. At higher energies we identified the elusive Bethe string excitations of length-2 and -3 by their characteristic field dependences and found excellent agreement between our measurements and theoretical calculations. This investigation provides a key step in the experimental verification of the long-sought Bethe string excitations and gives proof of their existence up to length-3 in a second compound after their first observation in SCVO. This result is of strong importance

in revealing the novel high energy many-body excitations in quantum spin chains.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [47].

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