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Spectrum of a Magnetized Strong-Leg Quantum Spin Ladder


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Inelastic neutron scattering is used to measure the spin excitation spectrum of the Heisenberg $S = 1/2$ ladder material $(C_3H_{10}N)_2CuBr_4$ in its entirety, both in the gapped spin liquid and the magnetic field-induced Tomonaga-Luttinger spin liquid regimes. A fundamental change of the spin dynamics is observed between these two regimes. Density matrix renormalization group calculations quantitatively reproduce and help understand the observed commensurate and incommensurate excitations. The results validate long-standing quantum field-theoretical predictions but also test the limits of that approach.

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One of the main attractions of low-dimensional quantum magnets is that these simple systems can be employed as quantum simulators [1] for fundamental field theories of particle and many body physics [2,3]. Conversely, much of what we know about spin chains and ladders is based on continuum quantum field-theoretical (QFT) nonlinear $\sigma$ models [4,5]. Such theories predicted and explained the gapped “spin liquid” ground states in integer-spin chains [6] and even-leg spin ladders [7], and provided a unified “Tomonaga-Luttinger spin liquid” (TLSL) picture of all gapless one-dimensional magnets [8]. Moreover, they painted a road map to how the phases in real materials are transformed one into another through quantum phase transitions [9] and provided new insights on their quantum critical behavior [10].

The recent discovery of new materials that experimentally realize spin chain and ladder models, as well as progress in neutron spectroscopy techniques and numerical methods, allows a quantitative verification of the relevant QFT predictions. Moreover, one can now push beyond the low-energy limit to explore features of spin chains and ladders that are not covered by continuous mappings of the actual spin Hamiltonians. In this context, in the present work, we focus on the quintessential model of the strong-leg antiferromagnetic $S = 1/2$ Heisenberg spin ladder [11]. We study the spin excitation spectrum on either side of a field-induced quantum phase transition between gapped and gapless phases, in its entire dynamic range. An excellent agreement with QFT predictions is obtained for some low-energy excitations. At the same time, we uncover numerous prominent higher-energy spectral features that were not covered by existing field-theoretical approaches.

The basic mechanism of the field-induced transition in a quantum spin ladder is well understood through a continuum mapping of the Heisenberg Hamiltonian based on Abelian bosonization [12–14]. It is a soft-mode transition, driven by a Zeeman splitting of the lowest-energy magnon triplet in the gapped phase. These magnons initially have a “relativistic” dispersion relation with a mass $\Delta$, and can be viewed as $S = 1$ confined states [15] of elementary $S = 1/2$ excitations called spinons [16]. Beyond the critical field $H_{c1} = \Delta / g \mu_B$, the spinons are deconfined and have a linear dispersion relation at small momenta. The spectrum is a gapless incommensurate multispinon continuum, described by TLSL theory at low energies. The incommensurability is directly related to the field-dependent magnetization [12].

Experimentally, a field-induced deconfinement of spinons in the TLSL regime has been beautifully demonstrated in neutron scattering experiments and numerical simulations on the strong-rung ladder material $(C_5H_{12}N)_2CuBr_4$ (BPCB) [17,18]. In such strong-rung ladders, due to the large gap and small bandwidth, spectral components arising from the three members of the initial triplet of magnons remain energetically separated in a wide range of fields above $H_{c1}$. In particular, the lower-energy excitations studied in BPCB [17] are all descendants of the $M_S = +1$ magnon mode. The main idea of the present work is to study the field evolution of excitations in the strong-leg case. Here, the spin gap is much smaller than the excitation bandwidth, and components of different spin polarization are no longer energetically separated. Since transitions between different spin states now occur at relatively low energies, the resulting spectrum is expected to be considerably more complex with numerous interesting high-energy features.

We study the compound $(C_7H_{10}N)_2CuBr_4$ (DIMPY) [19,20], which is structurally somewhat similar to BPCB. Unlike BPCB, though, it realizes the strong-leg antiferromagnetic Heisenberg $S = 1/2$ ladder model.
As described in Refs. [20,21], the ladders are built of Cu$^{2+}$ and run along the $a$ axis of the monoclinic crystal structure. Previous experiments and their comparison to density matrix renormalization group (DMRG) calculations established the spin Hamiltonian [21–23]. The bulk of all experimental data are adequately reproduced, assuming just two Heisenberg exchange constants $J_{\text{leg}} = 1.42$ meV and $J_{\text{rung}} = 0.82$ meV for the ladder legs and rungs. Such small energy scales, much smaller than those in the known strong-leg ladder compound La$_4$Sr$_{10}$Cu$_{24}$O$_{41}$ [24], make the quantum phase transition in DIMPY at $H_{c1} \approx 2.6$ T easily accessible in neutron experiments.

As a reference point, in Fig. 1(a), we show the magnetic excitation spectrum measured in DIMPY in zero applied field at $T = 70$ mK. These data were taken on the same sample as in previous studies [21–23], using the neutron time-of-flight spectrometer LET at the ISIS facility [25]. The sample was mounted on a $^3$He-$^4$He dilution refrigerator inside a vertical cryomagnet. Taking advantage of repetition rate multiplication, low- and high-resolution spectra with an initial energy of $E_i = 4.2$ meV and 2.2 meV were recorded at the same time. The inelastic background originating from the sample and instrument components was determined from the zero-field measurement and subtracted pixel by pixel [26]. All data shown are integrated along the non-dispersive $b^*$ and $c^*$ directions and projected onto the plane defined by energy transfer $\hbar \omega$ and momentum transfer along the leg direction $q_{||} = \mathbf{Q} \cdot \mathbf{a}$ [23].

As discussed in detail in Refs. [22,23], this spectrum shows several distinct features. The lowest-energy excitations are a triplet of single-magnon states that are the key prediction of the nonlinear $\sigma$ models [15]. In DIMPY, they have a gap of $\Delta = 0.33$ meV at $q_{||} = \pi$ and a dispersion with a relativistic velocity $c = 2.36$ meV [27]. At higher-energy transfers, there is an extended two-magnon bound state separated from a diffuse multimagnon continuum at even higher energies. While both a continuum and bound state [23] are predicted by the field-theoretical approach of Ref. [15], it is not able to describe their dispersion and internal structure. The experiment is in almost perfect quantitative agreement with DMRG calculations [Fig. 1(b)] based on just the two exchange constants quoted above. Here, the numerical result was convoluted with the instrumental resolution, allowing a direct comparison to the experimental data.

An external magnetic field $H$ lifts the threefold degeneracy of the excitation spectrum. This is clearly visible in Figs. 2(a) and 2(b), which summarizes the data collected at $H = 2.55$ T $< H_{c1}$ applied along the crystallographic $b$ axis in (a) low- and (b) high-resolution setups. As long as the critical field is not exceeded, the singlet ground state of the spin ladder remains unchanged. Since the Zeeman term commutes with the Heisenberg Hamiltonian, the spectrum can be viewed as a superposition of three distinct polarization channels with excitations carrying $M_S = 0$, $\pm 1$, respectively, $M_S$ being the eigenvalue of the $S^z_{\text{tot}}$ operator and $z$ the direction of applied field. Since the temperature is low enough to ensure a negligible population of excited states, the three components are identical to the spectrum at $H = 0$, except for an overall Zeeman energy shift. This

FIG. 1 (color online). (a) Time-of-flight inelastic neutron scattering spectrum measured on DIMPY in zero applied field ($E_i = 4.2$ meV). (b) A DMRG calculation of the same spectrum, folded with the known resolution function of the neutron instrument for a direct comparison with experiment.

FIG. 2 (color online). Spin excitations in DIMPY at $H = 2.55$ T. Inelastic neutron data were measured at $T = 70$ mK in the low-resolution [(a) $E_i = 4.2$ meV] and high-resolution [(b) $E_i = 2.2$ meV] modes. (c),(d) Numerical DMRG calculations, convoluted with experimental resolution. Dashed lines indicate the onset of the elastic line in the experiment. Solid lines indicate the low-energy magnon excitations with relativistic dispersion.
applies not only to the magnon branch, but to all features, including the bound states and continua. In a neutron scattering experiment, the relative intensities of the three polarization contributions scale as 1:2:1, for $M_S = -1, 0,$ and 1, respectively, due to the intrinsic polarization dependence of the neutron scattering cross section. A DMRG calculation [Figs. 2(c) and 2(d)] for the same conditions as the neutron experiment illustrates this simple yet important result.

The central result of this study is the measurement of excitations in the gapless TLSL phase. As can be seen from the data measured in DIMPY at $H = 7$ T [Figs. 3(a) and 3(b)], beyond the critical field, the spectrum undergoes qualitative changes compared to that at low fields. The previously sharp transitions in the gapless TLSL phase. As can be seen from the experiment illustrates this simple yet important result.

$q_\parallel$ modes decompose into structured and overlapping continua. A gapless linearly dispersive excitation emanating from $q_\parallel = \pi$ is seen at low energies. Many more distinct gapped features, with minima at either commensurate or incommensurate wave vectors, appear at higher energies.

To make sense of this multitude of spectral features, we are going to classify them by their spin projections and by their parity with respect to a permutation of the equivalent ladder legs. Both quantities are conserved by the Heisenberg and Zeeman Hamiltonians. One has to keep in mind, though, that at $H > H_{c1}$, the spin ladder is magnetized, and therefore the contributions of different polarizations are no longer identical. The total neutron scattering cross section can be broken up into six independent parts [18]:

$$
\frac{d^2\sigma}{d\Omega dE} \propto 4 \left(1 - \frac{Q^2}{Q^2}\right)[s^+(Q)S^0_z + s^-(Q)S^0_z] + \left(1 + \frac{Q^2}{Q^2}\right)[s^+(Q)(S^0_z + S^0_z)]
$$

Here, $Q$, denotes the component of the momentum transfer $Q$ along magnetic field. $S^{\alpha\beta}_{q\perp} = S^{\alpha\beta}(q\perp , \omega)$ are dynamic correlation functions associated with the different symmetry channels. The superscripts $\alpha$ and $\beta$ label the correlated spin components: $(\alpha, \beta) = (zz)$ for correlations longitudinal to the field and $(\pm \pi)$ for transverse ones. The subscript $q_\perp = 0, \pi$ is the momentum transfer along the ladder rungs. The corresponding structure factors represent one-dimensional correlations of sums $(q_\perp = 0)$ and differences $(q_\perp = \pi)$ between the two spins on each rung, as discussed in detail in Refs. [21,23].

It is, in principle, possible to separate the six channels in inelastic neutron scattering experiments by performing measurements at different wave vectors in different Brillouin zones or by applying a horizontal magnetic field. In practice, this procedure is extremely challenging. Instead, to identify the various spectral components in the experimental data, we took guidance from DMRG calculations. In such simulations, the individual contributions $S^{\alpha\beta}_{q\perp}$ can be accurately obtained. For DIMPY, $H = 7$ T and $g = 2.17$ [22] for Cu$^{2+}$; the result is shown in Fig. 4 [28]. It corresponds to a net magnetization per site of $\langle S_z \rangle = 0.065$. The complete calculated cross section, for a direct comparison with experiment, is shown in Figs. 3(c) and 3(d). The spectacular agreement with the neutron data gives us confidence that the measured spectra can be deciphered using the numerical “key” of Fig. 4.

Our goal is, wherever possible, to relate the features observed to those predicted by the field-theoretical mapping of Refs. [12,13,15]. Starting with low energies, at $H > H_{c1}$, one expects several gapless excitations, generic to the TLSL state [12,13]. These continua are descendants of the soft $M_S = +1$ magnon. They have linear lower thresholds of $(h\omega)^2 = \nu^2(q_\parallel - q^*)^2$, as indicated by solid lines in Figs. 3 and 4. Commensurate gapless excitations with $q^* = \pi$ occur in the $S^{\pi+}_0$ channel and are readily observed experimentally. Incommensurate excitations around $q^* = \pi \pm 4\pi(S_z)$ and $q^* = \pm 4\pi(S_z)$ are predicted in the $S^{\pi+}_0$ and $S^{\pi-}_0$ channels, respectively. They are detected by our DMRG calculations but appear at least 2 orders of magnitude weaker than the commensurate ones, and therefore remain undetected in our experiments. Contrarily, gapless incommensurate excitations were observed in strong-rung ladders [17]. This remarkable difference can be explained by the different nature of interactions between spinons (spinons are repulsive in
FIG. 4 (color online). Calculated components of the $H = 7 \text{T}$ excitation spectrum of DIMPY, classified by their spin projection quantum number (top to bottom) and parity with respect to ladder-leg interchange (left, asymmetric; right, symmetric). Lines and symbols are as in Fig. 3.

strong-rung and attractive in strong-leg ladders), leading to distinct TLSL exponents $K$ but also to different amplitudes of commensurate and incommensurate excitations [18]. The ratios $(\alpha_1, \alpha_2) = (A_x/A_S, A_y/B_y)$ between commensurate (incommensurate) excitations in $S_0^{x\pm}$ with amplitude $A_x$ ($B_y$) and incommensurate excitations in $S_0^{z\pm}$ (amplitude $A_z$) at $\langle S_x \rangle = 0.065$ were numerically calculated to be $(\alpha_1, \alpha_2) = (13.1, 8.9)$ for DIMPY [22,29] and (2.5, 1.3) for BPCB [18], proving that incommensurate contributions are suppressed in strong-leg ladders. For the commensurate modes in DIMPY, linear fits of the experimental and numerical low-energy spectra allow us to extract the TLSL velocity $v$ [18]. Experimentally, we obtain $v = 2.5(2) \text{meV}$, which is comparable to $v = 2.0(1) \text{meV}$, as estimated [30] from the calculated spectrum $S_{z\pm}^2$ or $v = 1.87(1) \text{meV}$ from static correlations in Ref. [22].

In addition to the gapless spectrum, Refs. [12,13] predict gapped excitations that are descendants of the $M_S = 0$ magnon. They appear in the $S_{zz}^{\pm}$ as well as in the $S_0^{x\pm}$ channels and show incommensurate minima at $q_{iz} = \pi \pm 2\pi \langle S_x \rangle$ (gray circles in Figs. 3 and 4) and $q_{iz} = \pm 2\pi \langle S_x \rangle$ (white circles in Figs. 3 and 4), respectively. The minima around $q_{iz} = \pi$ are readily observed in our measured spectrum (grey circles in Fig. 3). They are located at $q_{iz}/2\pi = 0.345(3)$ and $0.560(3)$, in excellent agreement with expectation, based on the known value of $\langle S_x \rangle$. Moreover, we observe a hint of an incommensurate minimum at $q_{iz}/2\pi = 0.06(2)$ [white circle in Fig. 3(a)] which agrees with the predicted excitation at $q_{iz} = \pm 2\pi \langle S_x \rangle$ in the $S_0^{x\pm}$ channel. However, while we reproduce the predicted $q_{iz}$ position of the incommensurate gapped minima, we observe an additional energy shift due to a renormalization of the gap outside the vicinity of the quantum critical point.

Quantum field theory [13] predicts that excitations at the zone center $q_{iz} = 0$ (in $S_{zz}^{x\pm}$) as well as at the magnetic zone center $q_{iz} = \pi$ (in $S_{zz}^{x\pm}$) appear directly at $\hbar \omega = g \mu_B H$. In our data on DIMPY, we observe corresponding excitation energies of $\hbar \omega = 0.90(1) \text{meV}$ at $q_{iz} = \pi$ (grey stars in Figs. 3 and 4) and $\hbar \omega = 0.896(4) \text{meV}$ at $q_{iz} = 0$ (white stars in Figs. 3 and 4), in excellent agreement with $g \mu_B H = 0.89 \text{meV}$ for $g = 2.17$ and $H = 7 \text{T}$.

We hence experimentally verify most of the QFT predictions in Refs. [12,13]. However, many of the observed features remain unaccounted for. These include the internal structure of the continua stemming from the $M_S = 0$ magnon branch, as well as the descendant of the $M_S = -1$ magnon in the $S_{zz}^{x\pm}$ and $S_{zz}^{x\pm}$ channels, respectively [Figs. 4(a) and 4(c)]. Moreover, the existing QFT results do not bring any insight on how two-magnon excitations evolve in the TLSL regime. In Figs. 4(b), 4(d), and 4(f), remains of the two-magnon bound state and continuum excitations are still visible in the $S_{zz}^{x\pm}$ and $S_{zz}^{x\pm}$ channels. Apart from a shift in energy, a slightly reduced bandwidth, and a loss of intensity, two-magnon excitations in the $S_{zz}^{x\pm}$ and $S_{zz}^{x\pm}$ channels resemble the corresponding contribution in zero field. At the same time, two-magnon excitations in the $S_{zz}^{x\pm}$ channel [at highest energies in Fig. 4(d)] gain a novel internal structure.

We conclude that the spin correlations in even the simplest magnetized quantum spin ladders are rather complex and cannot be fully captured by continuum theories. Fortunately, the recent progress in neutron instrumentation and numerical methods provides the necessary tools to quantitatively study the novel lattice effects and high-energy spectral features.

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In this work, we measure the velocity of spin excitations in energy units, as defined by a linear one-dimensional dispersion relation written as $\hbar \omega = \epsilon \mathbf{Q} \cdot \mathbf{a}$. The values of $\Delta$ and $c$ were extracted from the previously measured dispersion relation in Ref. [21].

The DMRG computation parameters were the same as in Ref. [22].

The amplitudes $A_x$ and $B_x$ were calculated as in Ref. [22] but not shown there. The values of the TLSL exponent $K$ at $\langle S_\perp \rangle = 0.065$ are $K = 0.966$ and 1.170 for BPCB and DIMPY, respectively.

The fit errors are estimated using different fitting procedures.