Magnetic-Field-Induced Bound States in Spin-$\frac{1}{2}$ Ladders

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Motivated by the recently observed intriguing mode splittings in a magnetic field with inelastic neutron scattering in the spin ladder compound $(C_5H_{12}N)_2CuBr_4$ (BPCB), we investigate the nature of the spin ladder excitations using a density matrix renormalization group and analytical arguments. Starting from the fully frustrated ladder, for which we derive the low-energy spectrum, we show that bound states are generically present close to $k = 0$ in the dynamical structure factor of spin ladders above H_{c1}, and that they are characterized by a field-independent binding energy and an intensity that grows with $H - H_{c1}$. These predictions are shown to explain quantitatively the split modes observed in BPCB.

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Intermediate between chains and 2D lattices, spin ladders have played a central role in the investigation of quantum effects in magnetism [1]. Being effectively 1D, they can be studied by state-of-the-art methods of quantum 1D physics such as bosonization [2–4] and density matrix renormalization group (DMRG) [5,6]. On the experimental side, numerous spin ladder compounds have been discovered and extensively studied [7–12]. Accordingly, their physics is very well understood. In zero field, the spectrum of a ladder is gapped regardless of the ratio of leg to rung coupling [4], and when they are of the same order, the ground state can be interpreted as some kind of resonating valence bond state, implementing an idea put forward by Anderson in his seminal paper on the theory of high T_c superconductors [13]. The spin gap can be closed by a magnetic field, leading to a Tomonaga-Luttinger liquid phase with gapless excitations and incommensurate correlations [14–21]. All these properties have been observed in many systems. For instance, inelastic neutron scattering (INS) has been used to study the fate of the low-lying excitations across the quantum phase transition at which the gap closes [22–25], and a continuum of excitations has been observed in the gapless phase in perfect agreement with theoretical expectations. It has even been possible to check the universal finite-temperature scaling of the transverse local dynamical structure factor at the gapped-gapless quantum critical point [26].

Some puzzles remain however. One of them has to do with the higher energy excitations in the gapless phase. In the presence of a magnetic field, triplet excitations are split. The lowest branch crosses the singlet ground state and gives rise to the low-energy continuum, while the other two branches lie at a finite energy. The dynamical spin structure factor of these branches turns out to have a rather complex structure however, as revealed by neutron scattering on $(C_5H_{12}N)_2CuBr_4$ (BPCB) and time-dependent DMRG [27–29] on a ladder with strong rungs. The main features, which will be discussed in great detail below, are summarized in Figs. 1 and 2. Let us concentrate on the intermediate branch. It consists of three main features, and a weak continuum barely visible on that scale. The two upper features (black dashed lines in Fig. 1), and the associated continuum, have been nicely explained by Bouillot *et al.* [30] in terms of an effective $t - J$ model where the ground state is described by an effective spin chain with a finite

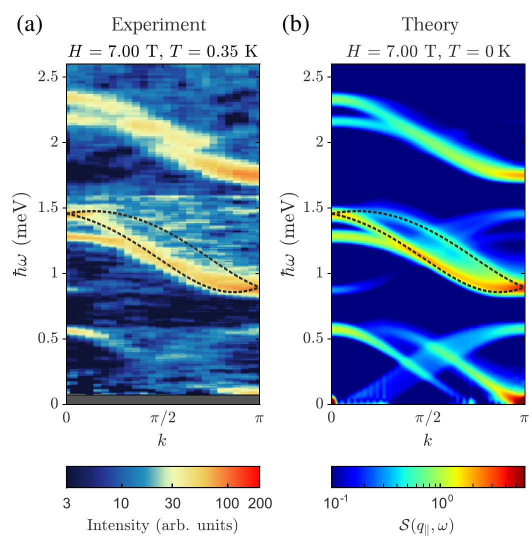

FIG. 1. (a) INS spectrum measured in BPCB in the slightly magnetized regime. Three strongly Zeeman-split triplet branches are clearly visible. Near $k = 0$ a distinct splitting of the middle and upper triplet band is observed. These data were previously published in Ref. [26]. (b) DMRG simulation of the dynamic structure factor calculated at the same magnetic field. The black dashed lines in the DSF plots of both panels mark the boundaries of the continuum associated to the dispersion of t^0 rung triplets as predicted by the mapping on the $t - J$ model [30].

magnetization, and the $S_z = 0$ triplet excitation is represented by a mobile hole.

By contrast, the lowest feature of the intermediate branch, the split mode around $k = 0$ below the continuum bounded by the black dashed lines in Fig. 1, has not been discussed. This splitting, which is also present in the upper branch, clearly calls for an explanation.

In this Letter, we provide strong evidence that this split mode is a bound state of the excited triplet with one of the triplets induced in the ground state by the magnetic field in the gapless phase. This identification relies on three results: (i) the exact solution of the low-lying spectrum of the fully frustrated ladder with cross couplings equal to leg couplings in terms of triplets and of bound states of pairs of triplets; (ii) the smooth evolution of the lowest bound state upon reducing frustration, as revealed by extensive DMRG simulations; and (iii) the prediction that the intensity of this mode grows with the distance to the critical field while its binding energy is essentially independent of the field, a prediction in perfect agreement with the neutron scattering results on BPCB in the gapless phase. Note that bound states have been discussed before in zero magnetic field [31–34] as multitriplet excitations from the singlet ground state. To the best of our knowledge, the possibility of observing them above the first critical field with inelastic neutron scattering as split features of the main triplet bands with significant spectral weight had not been anticipated however.

Our experimental results were obtained on the compound $(C_5H_{12}N)_2CuBr_4$, a particularly well-studied $S = 1/2$ strong rung spin ladder material [19,20,22,23,26,35]. The ladders are formed by magnetic Cu^{2+} cations and linking Br^- anions [35]. The rung and leg couplings respectively are given by $J_\perp = 12.67(6)$ K and $J_\parallel = 3.54(3)$ K and the critical field is $\mu_0 H_{c1} = 6.66(6)$ T [26]. All the inelastic neutron scattering data [36] were previously published in Ref. [26], and experimental details can be found therein. The mentioned study exclusively focused on the universal low energy spin dynamics. The subject of the present Letter on the other hand is the high energy triplet excitations.

All theoretical results reported in this Letter have been obtained on the model

$$\mathcal{H} = J_\parallel \sum_{i,j=1,2} \vec{S}_{i,j}\vec{S}_{i+1,j} + J_\perp \sum_i \vec{S}_{i,1}\vec{S}_{i,2} + J_\times \sum_i (\vec{S}_{i,1}\vec{S}_{i+1,2} + \vec{S}_{i,2}\vec{S}_{i+1,1}) - h \sum_{i,j=1,2} S^z_{i,j},$$

where $J_\perp$ is the rung coupling, $J_\parallel$ is the leg coupling, and $J_\times$ is a cross coupling (not present in BPCB) that frustrates the leg coupling. The component of the vector operators $\vec{S}_{i,j}$ are spin-1/2 operators. The first index keeps track of the rung, the second one of the leg. The g factor and the Bohr magneton μ_B have been included in the magnetic field h. The numerical results have been obtained using the time-dependent DMRG method pioneered in this context by Bouillot *et al.* [30], and we have benchmarked our code by reproducing the results of Bouillot *et al.* in the unfrustrated case ($J_\times = 0$) [30]. The dynamical structure factor (DSF) can be defined in the Lehmann representation by

$$S^{\alpha\alpha}_{k_\perp}(k, \omega) = \frac{2\pi}{N_r} \sum_\eta |\langle \eta | S^\alpha_{k_\perp}(k) | \mathrm{GS} \rangle|^2 \delta(\omega + E_{\mathrm{GS}} - E_\eta)$$

where N_r is the number of rungs, $|\mathrm{GS}\rangle$ is the ground state and the sum over η runs over the excited states. The rung operators (in position basis) are defined as $S^\alpha_{i,k_\perp=0,\pi} = S^\alpha_{i,1} \pm S^\alpha_{i,2}$. The action of the various components on the eigenstate of a rung dimer are summarized in Table I.

TABLE I. Action of the rung operators on a single rung.

	S^z_0	S^z_π	S^x_0	S^x_π
$\lvert s\rangle$	0	$\lvert t^0\rangle$	0	$-\sqrt{2}(\lvert t^+\rangle - \lvert t^-\rangle)$
$\lvert t^+\rangle$	$\lvert t^+\rangle$	0	$\sqrt{2}\lvert t^0\rangle$	$-\sqrt{2}\lvert s\rangle$
$\lvert t^0\rangle$	0	$\lvert s\rangle$	$\sqrt{2}(\lvert t^+\rangle + \lvert t^-\rangle)$	0
$\lvert t^-\rangle$	$-\lvert t^-\rangle$	0	$\sqrt{2}\lvert t^0\rangle$	$\sqrt{2}\lvert s\rangle$

In Fig. 1(b), a false color plot of the sum of the longitudinal and transverse symmetric and antisymmetric DSF components is shown. The field has been adjusted to the experimental value of Fig. 1(a). These results agree qualitatively with those of Bouillot *et al.*, obtained at a slightly larger field. The inelastic neutron scattering cross section contains the same DSF components with k-dependent weights [24,26]. In the scattering geometry of the present experiment, these weights are near unity. Further, the experimental and numerical resolution are quite comparable. Indeed, we find good qualitative agreement between the experiment and the numerical simulation.

Let us start by discussing the spectrum of the fully frustrated ladder ($J_\times = J_\parallel \equiv J$) in the absence of a field

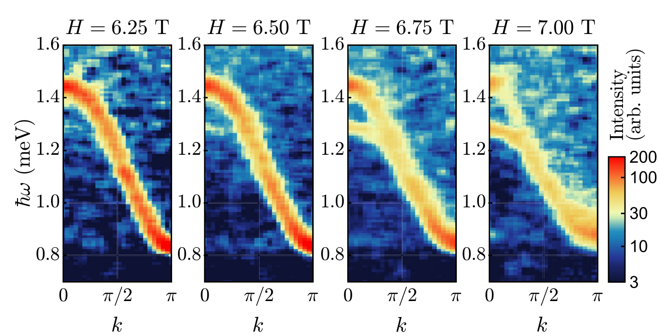

FIG. 2. Evolution of the INS spectrum measured in BPCB at 0.35 K at different magnetic fields near the critical field of $H = 6.66(6)$ T. In the plotted range of energy transfer only the middle triplet branch of excitations is visible. Upon increasing the magnetic field beyond H_{c1}, a distinct splitting is observed near the band maximum. These data were previously published in Ref. [26].

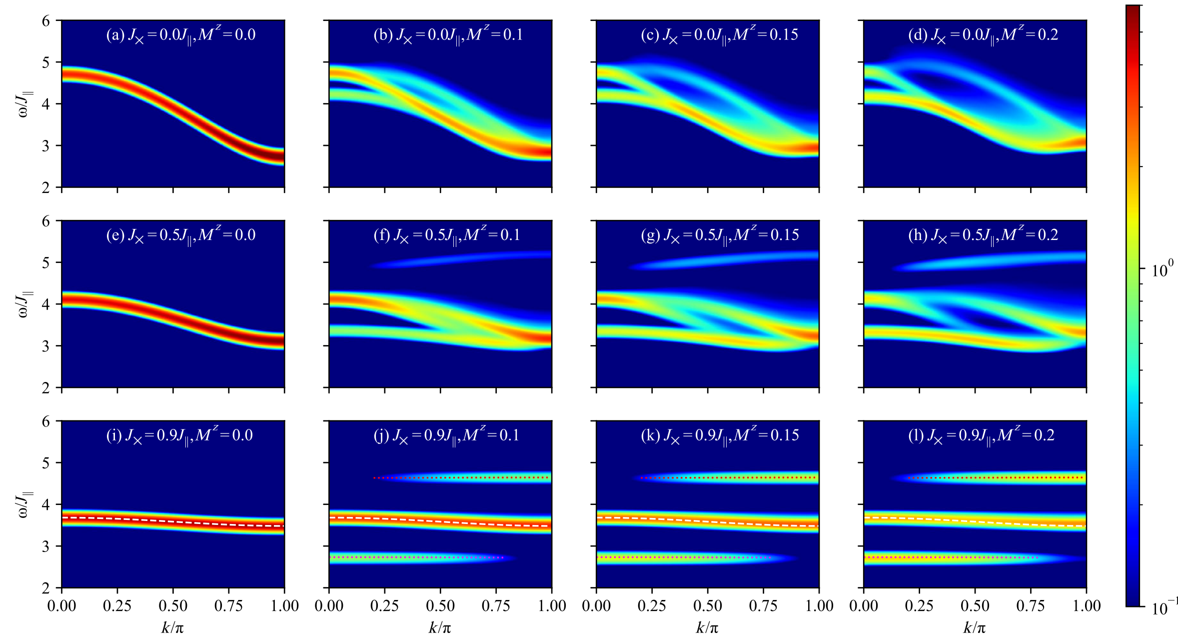

FIG. 3. Evolution of the longitudinal antisymmetric DSF $S_\pi^{zz}(k,\omega)$ with magnetization and frustration as obtained with DMRG (color plots) and perturbation theory (dashed lines, lower panels). At strong frustration, the three branches correspond to a single-triplet branch (middle) and two bound states with total spin 2 (upper mode) and total spin 1 (lower mode). Upon reducing the frustration, the upper bound state progressively loses its intensity, while the lower bound state evolves smoothly into the split mode at $k = 0$ of the unfrustrated case, leading to the interpretation of this mode as a spin-1 bound state.

[37–42]. The Hamiltonian can be rewritten entirely in terms of the sum of the spins on each rung so that the total spin of each rung is a conserved quantity. Accordingly, there are 2^{N_r} decoupled sectors of the Hilbert space, corresponding to the possible values of the spin (0 or 1) of each rung. For $J_\perp/J > 1.401$, the ground state is a product of singlets. The lowest energy excitation consists of creating a triplet on a rung. It is immobile because both neighbors are singlets, its energy is equal to $J_\perp$, and it is $(3 \times N_r)$-fold degenerate, the factor 3 coming from the three possible values of S_z, -1, 0, or 1. The next excitations correspond to the creation of two triplets. If the triplets are separated by at least one singlet, they are immobile. The energy of such an excitation is equal to $2J_\perp$, and it is $[9 \times N_r(N_r - 3)/2]$-fold degenerate. By contrast, if the triplets are nearest neighbors, the spectrum is that of two $S = 1$ spins coupled by J. The pair is immobile because it is surrounded by singlets. This will result in a singlet, a triplet, and a quintuplet bound state, with energies $2J_\perp - 2J$, $2J_\perp - J$, and $2J_\perp + J$, respectively. Each of these bound states is N_r-fold degenerate. The wave functions of these states are given in the Supplemental Material [43]. Higher energy states will not enter the discussion of the DSF, so we do not quote them.

In a magnetic field, and for $J_\perp$ large enough, the fully frustrated ladder has a direct transition from the product of singlets to a $1/2$ plateau state with triplets on every other bond at $h = J_\perp$ [45–47], followed by another direct transition to the fully saturated state at $h = J_\perp + 2J$. So to discuss the DSF at small magnetization, we need to leave the fully frustrated limit and restore a continuous magnetization curve by introducing a small perturbation defined by $\delta J = J_\parallel - J_\times$ such that, for $\delta J = 0$, the ladder is fully frustrated, while, for $\delta J = J_\parallel$, we get the regular ladder relevant for BPCB. In zero field, the degeneracy of the first excited states is lifted to first order, and the excitations acquire a dispersion given by (see the Supplemental Material [43])

$$\omega_1(k) = J_\perp + \delta J \cos k.$$

For the two triplet excitations, the excitations corresponding to triplets far apart give rise to a two-magnon continuum built out of the dispersion $\omega_1(k)$. By contrast, the bound states acquire a dispersion, but since the basic process induced by δJ is to let a triplet hop, moving a pair is a second-order process, and the effective hopping is of the order $(\delta J)^2/J$ [43]. In a magnetic field, the first level to cross the singlet ground state is the one triplet level sitting at the bottom of the band. The only competitors are the two triplet states with $S_z = 2$. Since the spin-2 bound state is higher than the state with two triplets far apart, the next state to cross will be a two-magnon state. This state will cross slightly later than the one triplet state because its energy is more than twice the ground state of the single triplet states—these triplets behave roughly speaking as spinless fermions with large nearest neighbor repulsion. Upon increasing the magnetization, the same argument will carry over with many-magnon states. The ground-state magnetization increases smoothly [43], and the ground state wave function is essentially a linear combination of states with the appropriate number of isolated triplets.

On the basis of these simple arguments, we are now in the position to predict the form of the DSF close to the fully frustrated limit. In the fully frustrated case, and in the limit $J = 0$, the only excitation that has a nonzero $k_\perp = \pi$ matrix elements with a state with a few isolated triplets and that has energy $J_\perp$ corresponds to a transition from the singlet to the t^0 triplet ($S_z = 0$) induced by S^z_π. This band will be split into three bands upon introducing J: a band of isolated triplets at energy $J_\perp$, and two bands of bound states corresponding to the states that contain the configuration $|t^+t^0\rangle$ or $|t^0t^+\rangle$, namely $|S=1,S^z=1\rangle = (|t^+t^0\rangle - |t^0t^+\rangle)/\sqrt{2}$ and $|S=2,S^z=1\rangle = (|t^+t^0\rangle + |t^0t^+\rangle)/\sqrt{2}$, of energy $J_\perp - J$ and $J_\perp + J$ respectively. Upon introducing the perturbation δJ, the isolated triplet acquires a dispersion of amplitude δJ, while the bound states acquire a dispersion of order $(\delta J)^2/J$, with an intensity proportional to $1 + \cos k$ ($1 - \cos k$) for the lower (upper) mode and a momentum shift by π [43]. Finally, the intensity of the isolated triplet is expected to be much larger at low magnetization because the probability to excite a singlet not adjacent to a triplet is almost equal to 1 (it is 1 in the limit of vanishing magnetization), while the bound states are expected to have an intensity proportional to the number of triplets in the ground state, i.e., to the magnetization.

These predictions are fully supported by DMRG simulations (see Fig. 3). The spectrum of a ladder close to full frustration ($\delta J = 0.1J$) is shown in the bottom panels as a function of magnetization. As expected, a single branch with small but visible dispersion is present at zero magnetization. Upon increasing the magnetization, two additional branches appear with essentially no dispersion, and with an intensity that increases with the magnetization.

To make connection with the standard ladder relevant to BPCB, we have performed extensive DMRG simulations for several values of $J_\times$. The spectrum acquires a complicated structure, but the evolution from the fully frustrated limit is transparent: (i) two copies of the single triplet band appear, shifted by a vector that increases with the magnetization, as predicted by the $t - J$ model analogy [30]; (ii) the upper bound state loses its intensity and is no longer visible in the standard ladder; and (iii) the lower bound is unaffected close to $k = 0$ while for larger k it gets mixed with the dispersive single triplet excitation.

These results strongly suggest that the split mode is the $S = 1, S_z = 1$ bound state of the fully frustrated case that survives as a well-defined excitation close to $k = 0$. To lend further support to this interpretation, let us look at the position and intensity of this branch. In the limit of full frustration, the position of this bound state is at J below the single triplet branch independent of the magnetic field, while its intensity grows linearly with magnetization.

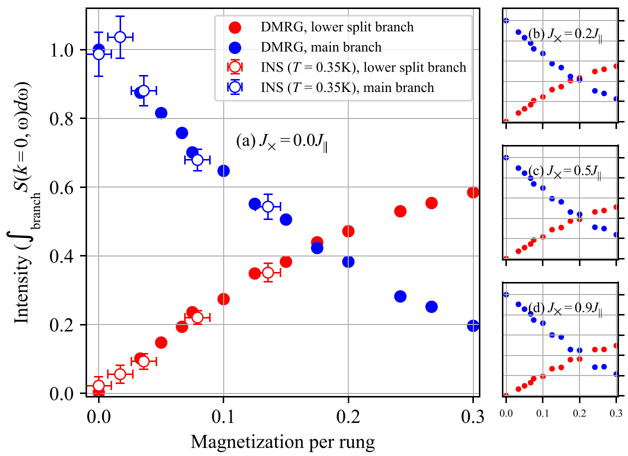

FIG. 4. Main panel: intensity of the two main branches at $k = 0$ as a function of magnetization from DMRG (solid symbols) and experiments (open symbols). The agreement between theory and experiment is quantitative, and the main effects are very clear: the intensity of the split mode increases with magnetization, as it should for a bound state, while that of the single triplet branch decreases. Right panels: DMRG results for frustrated ladders. The results are qualitatively the same.

These properties are shared by the split mode at $k = 0$ of both the DMRG results of the standard ladder and the INS results on BPCB (see Fig. 2) with a remarkable degree of accuracy. Indeed, the energy of the split mode does not change in any significant way from its value when it first appears. By contrast, its intensity increases quite fast with magnetization, as shown in Fig. 4, while at the same time the intensity of the single triplet mode decreases. The DMRG simulations show that this is essentially independent of frustration [see Figs. 4(b)–4(d)], supporting the continuity between the fully frustrated and the standard limits, hence the interpretation as a bound state. The comparison between the standard ladder and BPCB is shown in the main panel. To make this plot, the experimental branch intensities have been extracted from narrow slices $k/2\pi \in [-0.05, 0.05]$ of the neutron scattering data obtained at $\mu_0 H = 6.00$, 6.25, 6.50, 6.75, and 7.00 T. The corresponding magnetization was directly measured at the same fields using a Faraday balance magnetometer [43]. After multiplying the arbitrarily normalized neutron scattering data with a single overall prefactor, these points have been included in Fig. 4 as open symbols. The DMRG intensities have been obtained along similar lines [43].

The same kind of analysis can be made for the upper branch of excitation. This branch corresponds to the excitation of a rung singlet to the $S^z = -1$ triplet under the action of S^x_π. In the fully frustrated case, it gives rise to a main branch at $2J_\perp$, and to three bound state branches corresponding to the singlet, triplet, and quintuplet bound states of energy $2J_\perp - 2J$, $2J_\perp - J$, and $2J_\perp + J$ since all of them have a component with $|t^+t^-\rangle$. Upon reducing frustration, the only bound state that remains visible is the triplet with energy $2J_\perp - J$, giving rise to a split mode very similar to that of the intermediate branch [43].

In conclusion, we have solved one of the main remaining open questions in the physics of spin-1/2 ladders, the origin of the split modes observed in inelastic neutron scattering. Starting from the fully frustrated ladder, in which single particle and bound state excitations are very transparent, we have shown by continuity using DMRG simulations that the split modes originate from spin-1 bound states. It would be interesting to see if this conclusion can also be reached as a consequence of the nearest-neighbor attractive interaction between a hole and the up triplet in the context of the $t-J$ description of the excitations [43]. This goes beyond the scope of the present Letter however.

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[43] See the Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.124.087203, which includes Ref. [44], for details about the magnetization, the extraction of the intensities, the perturbative calculation of the dispersion of all the modes close to the fully frustrated case, the calculation of the dynamical structure factor in that limit, a discussion of the higher energy excitations, and a comparison with the $t-J$ model.

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