

## Electron spin resonance modes in a strong-leg ladder in the Tomonaga-Luttinger liquid phase

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Magnetic excitations in the strong-leg quantum spin ladder compound  $(\text{C}_7\text{H}_{10}\text{N})_2\text{CuBr}_4$  (known as DIMPY) in the field-induced Tomonaga-Luttinger spin-liquid phase are studied by means of high-field electron spin resonance (ESR) spectroscopy. The presence of a gapped ESR mode with unusual nonlinear frequency-field dependence is revealed experimentally. Using a combination of analytic and exact-diagonalization methods, we compute the dynamical structure factor and identify this mode with longitudinal excitations in the antisymmetric channel. We argue that these excitations constitute a fingerprint of the spin dynamics in a strong-leg spin-1/2 Heisenberg antiferromagnetic ladder and owe their ESR observability to the uniform Dzyaloshinskii-Moriya interaction.

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The investigation of spin systems where quantum effects play a dominant role has become a very active branch of quantum many-body physics. Although the spin Hamiltonian describing quantum magnets is quite simple and often very well controlled [1], the interplay of all spin degrees of freedom can be very complex, leading to a large diversity of phases ranging from long-range magnetic order to spin liquids of various types [2]. In addition, the ground state can possess not only local types of order but also more complex and subtle nonlocal topological orders [3,4]. Understanding such behavior is thus a frontier of fundamental knowledge, providing, on the other hand, a potential means for quantum computation [5] or quantum simulators of some itinerant problems [6,7].

Due to enhanced quantum effects, one- and quasi-one-dimensional (1D) spin systems, such as spin chains and ladders, are of particular interest [8]. In these systems, interactions between excitations can play a very important role, giving rise to exotic states [9], including quasi-long-range order, known as Tomonaga-Luttinger liquids (TLLs), or phases where correlations between magnetic excitations are of short range (e.g., in the case of Haldane spin-1 chains [3]).

Recent progress in material science makes it possible to synthesize new materials with exchange parameters permitting the manipulation of the ground states by accessible magnetic fields, with drastic effects on the physical properties. This and the progress in both analytical and numerical techniques provide access to a host of novel physics, allowing, e.g., the obser-

vation of the Bose-Einstein condensation of magnons [6,10], the quantitative test for TLL predictions [11,12], the observation of fractionalization of spin excitations [13,14], spinon attraction [15], and remarkable effects of disorder [16–18].

Even very tiny anisotropies can play an important role, reducing local symmetries and drastically affecting the low-energy spin dynamics. Electron spin resonance (ESR) spectroscopy has proven to be one of the most sensitive tools to probe such interactions and effects in exchange-coupled spin systems [19]. One remarkable advantage of this technique is that ESR allows experiments in very high magnetic fields, far beyond the superconducting magnet limit [20–23]. Theoretical studies of predicted ESR parameters are available for spin chains and ladders [24–30], and have been applied with good success to, e.g., spin chains [31–33] and strong-rung ladders [34,35]. However, relatively little is known about the spin dynamics in strong-leg ladder systems, which can be very different from that in spin chains and strong-rung ladders in terms of the spinon interactions.

In this Rapid Communication, we report on high-field ESR studies of the spin ladder  $(\text{C}_7\text{H}_{10}\text{N})_2\text{CuBr}_4$  [bis(2,3-dimethylpyridinium) tetrabromocuprate(II) or (2,3-dmpyH)<sub>2</sub>-CuBr<sub>4</sub>, abbreviated as DIMPY], currently known as the best realization of a strong-leg spin-1/2 Heisenberg antiferromagnetic ladder [36] with moderate exchange-coupling constants. We reveal experimentally the presence of an ESR excitation mode in the TLL phase that is absent in a strong-rung ladder and was not observed in previous ESR work on DIMPY [37]. We describe the unusual excitation spectrum of DIMPY, using a combination of analytic techniques and exact-diagonalization (ED) methods. We demonstrate that the appearance and magnetic-field dependence of parameters

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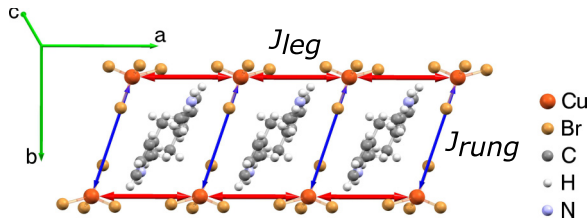


FIG. 1. (Color online) Schematic view of the crystal structure of DIMPY [36]. The copper (Cu) ions form a ladderlike structure with the dominant exchange couplings indicated in the figure.

of our mode can be understood by taking into account the dynamic spin-spin correlation function for the strong-leg spin-1/2 Heisenberg antiferromagnetic ladder model, thus providing important information on the spin excitations as well as the anisotropy of magnetic interactions in this system.

DIMPY crystallizes in a monoclinic lattice with space group  $P2(1)/n$  and lattice constants  $a = 7.504 \text{ \AA}$ ,  $b = 31.61 \text{ \AA}$ ,  $c = 8.202 \text{ \AA}$ ,  $\beta = 98.98^\circ$  (number of formula units per unit cell  $Z = 4$ ) [36] with  $S = 1/2$   $\text{Cu}^{2+}$  ions arranged in a ladderlike structure (Fig. 1). Each unit cell contains two rungs, each from a different symmetry-equivalent ladder, running parallel to the  $a$  axis. The spin Hamiltonian of DIMPY can be written as

$$\mathcal{H} = J_{\text{leg}} \sum_{(l,j)} \mathbf{S}_{l,j} \cdot \mathbf{S}_{l+1,j} + J_{\text{rung}} \sum_{(l)} \mathbf{S}_{l,1} \cdot \mathbf{S}_{l,2} - g\mu_B H \sum_{l,j} S_{l,j}^z + \mathcal{H}_\delta, \quad (1)$$

where  $J_{\text{leg}}$  and  $J_{\text{rung}}$  are exchange-coupling constants along the legs and rungs, respectively,  $\mathbf{S}_{l,j}$  are the spin operators on site  $l$  of the leg  $j = 1, 2$  of the ladder, and  $g\mu_B H$  is the Zeeman term ( $g$  is the  $g$  factor,  $\mu_B$  is the Bohr magneton, and  $H$  is the applied magnetic field). The fourth term represents various possible, usually small, anisotropic contributions. Exchange constants along the rungs and legs of the ladder have been determined by use of inelastic neutron scattering (INS) as  $J_{\text{rung}}/k_B \approx 9.5 \text{ K}$  and  $J_{\text{leg}}/k_B \approx 16.5 \text{ K}$ , respectively ( $J_{\text{leg}}/J_{\text{rung}} \sim 1.73$ ) [38]. The ladders are coupled via very weak exchange interactions,  $J'/k_B \lesssim 5\text{--}7 \text{ mK}$  [36–38], resulting in a transition into a field-induced magnetically ordered phase at temperatures below  $\sim 0.35 \text{ K}$  [39].

In a strong-leg ladder, the transverse interchain interaction couples two spin chains. As a result, two spinons are confined to magnons, opening a spin gap in the excitation spectrum. In the presence of a magnetic field, the gap in DIMPY closes at a critical field  $H_{c1} = 2.8 \text{ T}$ , where the system undergoes a transition into the gapless TLL phase [40]. Above  $H_{c2} = 29 \text{ T}$ , the system is in the magnetically saturated spin-polarized phase [41]. Inelastic neutron scattering experiments revealed the presence of several gapless continua as well as a number of gapped excitations in DIMPY [38,42,43]; some of the excitations have been interpreted theoretically. Investigating the field-induced evolution of the magnetic excitation spectrum of a strong-leg ladder in the TLL state is of particular interest. Such a study would allow one to obtain a better understanding of the peculiarities of the spin dynamics in a strong-leg ladder in the TLL phase, which is, as shown below, rather different

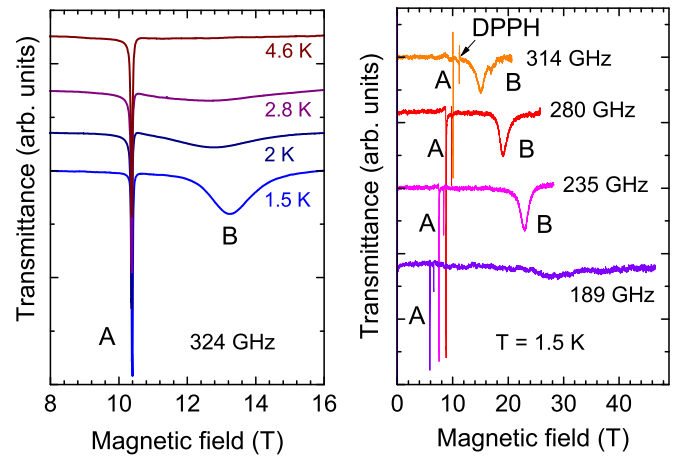


FIG. 2. (Color online) Left panel: Examples of ESR spectra obtained at a frequency of 324 GHz at 1.5, 2, 2.8, and 4.6 K. Right panel: Examples of pulsed-field ESR spectra obtained at the frequencies 189, 235, 280, and 314 GHz ( $T = 1.5 \text{ K}$ ).

from that known for quantum spin-1/2 chains and strong-leg ladders.

ESR experiments were performed at the Dresden High Magnetic Field Laboratory (Hochfeld Magnetlabor-Dresden), using transmission-type ESR spectrometers (similar to that described in Ref. [44]) equipped with 16 T superconducting and 50 T pulsed-field [45] magnets. VDI modular transmitters (product of Virginia Diodes, Inc., USA) and backward-wave oscillators (PO Istok, Russia) were employed as sub-mm radiation sources. High-quality single-crystal samples of DIMPY with typical sizes of  $2 \times 1 \times 1 \text{ mm}^3$  were used in our experiments. The magnetic field was applied along the  $b$  axis. In our experiments, 2,2-diphenyl-1-picrylhydrazyl (DPPH) with  $g = 2.0036$  was used as a standard ESR marker.

A single resonance line (mode A, Fig. 2) was observed at temperatures above  $\sim 4 \text{ K}$ . At lower temperatures, the ESR spectrum undergoes remarkable changes. In addition to mode A, we detected a relatively broad resonance absorption line (mode B, Fig. 2). With decreasing temperature, mode B becomes more intensive and narrower, shifting towards higher fields. Corresponding examples of the ESR spectra as well as the dependences of ESR linewidth (mode B) on temperature and magnetic field are shown in Figs. 2 and 3, respectively.

The frequency-field diagram of the magnetic excitations in DIMPY is shown in Fig. 4. Mode A (white boxes in Fig. 4) can be described using the equation  $h\nu = g_b\mu_B H$ , where  $h$  is the Planck's constant,  $\nu$  is the excitation frequency, and  $g_b = 2.23$ . Mode B (white circles in Fig. 4) has a more complex behavior: This mode is gapped for all fields and has a nonlinear frequency-field dependence. From the extrapolation of the frequency-field dependence to zero field, the energy gap,  $\Delta \sim 350 \text{ GHz}$ , can be estimated. This value agrees well with the size of the gap between the spin-singlet ground and first-excited triplet states observed by means of INS in zero magnetic field at  $k = 0$  [42,43], where the system is in the gapped spin-liquid state.

It is worth mentioning that the ESR excitation spectrum in DIMPY is very different from that in the strong-rung spin ladder  $(\text{C}_5\text{H}_{12}\text{N})_2\text{CuBr}_4$  (BPCB) [35], where only one gapless

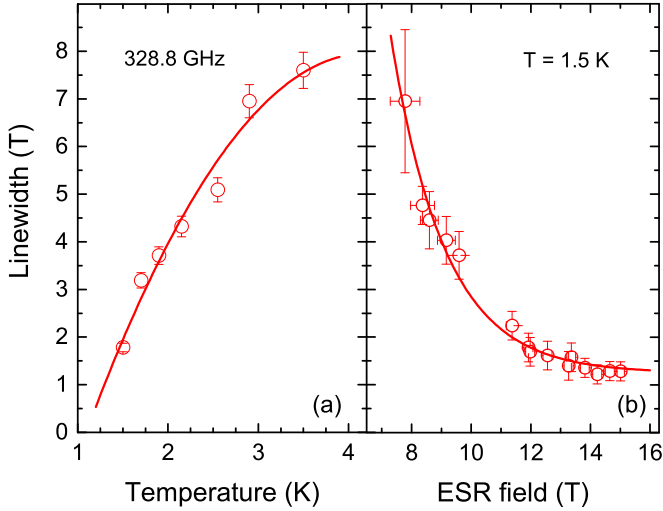


FIG. 3. (Color online) (a) Temperature dependence of the linewidth of mode B at a frequency of 328.8 GHz. (b) Linewidth of mode B for different resonance fields ( $T = 1.5$  K). Lines are guides to the eye.

mode was observed in the TLL phase. The comparison of our ESR data with results of INS studies [42] and ED calculations for DIMPY in the TLL regime strongly suggests that the observed ESR mode A corresponds to magnetic excitations in the  $S_0^\pm$  channel, while mode B corresponds to ESR excitations in the channel  $S_\pi^{zz}$ , which are nominally forbidden in the purely isotropic case. To demonstrate that  $S_\pi^{zz}$  indeed gives rise to mode B, we calculated the field dependence of the dynamical structure factor employing ED of the model (1), where the anisotropic contribution  $\mathcal{H}_\delta$  has been omitted. We used the parameters  $J_{\text{leg}}/J_{\text{rung}} = 1.73$ ,  $J_{\text{rung}} = 9.51$  K, and  $g = 2.23$  as determined above from the frequency-field dependence of mode A [47]. The transverse dynamical structure factor  $S_0^\pm$  in the symmetric channel of the legs and the longitudinal dynamical structure factor  $S_\pi^{zz}$  in the antisymmetric channel

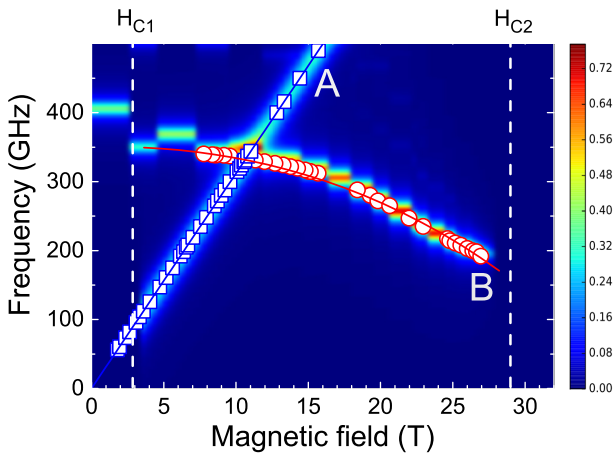


FIG. 4. (Color online) The frequency-field diagram of magnetic excitations in DIMPY. Data of the structure factors  $S_\pi^{zz}$  and  $S_0^\pm$  obtained by use of exact-diagonalization calculations for chains from  $N = 32$  to  $N = 64$  sites are given in bright colors [46]. Blue and red solid lines are guides to the eye. The first and second critical fields are denoted by vertical dashed lines.

are calculated for finite systems of up to 64 sites using the expression

$$S_{k_\perp}^{\alpha\beta}(\omega) = \frac{1}{\pi} \sum_n \text{Im} \frac{\langle 0 | S_{k_\perp}^\alpha | n \rangle \langle n | S_{k_\perp}^\beta | 0 \rangle}{\omega - (\epsilon_n - \epsilon_0 + i\eta)}, \quad (2)$$

for  $T = 0$ , where  $|n\rangle$  are the eigenstates with energy  $\epsilon_n$  ( $|0\rangle$  is the ground state).  $\eta$  is a Lorentzian broadening that we set to  $\eta = 0.05J_{\text{rung}}$ . The Fourier-transformed spin operators are given by

$$S_{k_\perp}^\alpha = \frac{1}{\sqrt{N}} \sum_{l,j} \exp(ik_\perp j) S_{l,j}^\alpha. \quad (3)$$

Thus,  $k_\perp$  is the momentum perpendicular to the ladder, while we have assumed zero momentum along the ladder direction, as is common for ESR.

We exploit the conservation of total  $S^z$  of the model (1). When the dimension of the subspace is sufficiently small, we use full diagonalization to evaluate (2), while for bigger dimensions we use first the Lanczos algorithm [48,49] to find the ground state  $|0\rangle$  and then a continued-fraction expansion [49–51] to obtain the spectral function (2).

Our ED results for the zero-temperature dynamical structure factors  $S_0^\pm$  and  $S_\pi^{zz}$  are shown as intensity plots in Fig. 4. Finite-size effects are strongest for low magnetic fields where they may amount to errors of up to 50 GHz for mode B [46]. For magnetic fields  $H > 12.5$  T, the main finite-size effects are the steps observed in “line” B, and thus one may estimate them to not exceed 10 GHz here. The agreement of the position of the intensity maxima for  $H \gtrsim 7$  T with the experimental ESR and INS [42] data is excellent, including not only the downward slope, but also the curvature of mode B. We note further that application of ED to finite temperature [46] also reproduces the qualitative trends observed in Figs. 2 and 3(a), in particular, a substantial thermal broadening of mode B.

As mentioned, ESR transitions corresponding to mode B are nominally forbidden in the purely isotropic case. On the other hand, Fig. 1 shows that there is no inversion center on bonds along the ladder legs in the crystal structure of DIMPY and successive tetrabromocuprate units are related by unit cell translations [36]. This allows for the presence of a uniform Dzyaloshinskii-Moriya (DM) interaction along the legs of the form  $\sum_l \sum_{j=1,2} (-1)^j \mathbf{D} \cdot (\mathbf{S}_{l,j} \times \mathbf{S}_{l+1,j})$ . It is important to mention that the uniform DM interaction has been found to be responsible for a number of unusual effects, including, e.g., broadening of resonance line A, observed in DIMPY by means of low-frequency ESR spectroscopy [37] and the zero-field gap opening in the triangular-lattice antiferromagnet  $\text{Cs}_2\text{CuCl}_4$  [52]. On the other hand, such a term accounts for the intensity of mode B, which is directly proportional to the spin-spin correlations as discussed above [46]. In the low-field limit, mode B can be described using the non-Abelian bosonization approach [53,54], where it is understood as a complex of two Majorana fermions [46]. The magnetic field couples symmetrically to the two legs of the ladder whereas the Majorana fermions are antisymmetric under the exchange  $j = 1 \leftrightarrow j = 2$  of the two legs. Thus, to first approximation, mode B is not affected by the applied field. This accounts for the almost flat behavior of mode B observed

in Fig. 4 up to about 15 T [46]. At higher magnetic fields, renormalization effects of these Majorana fermions are more important, resulting in the observed nonlinear frequency-field dependence of mode B.

Our observations of the  $S_{\pi}^{zz}$  mode can have a broader impact in the context of the SO(5) ladder model [55,56]. In this model, the quantum phase transition driven by the chemical potential can be mapped to the field-induced phase transitions in the Heisenberg ladder. In that case, the gapless excitations in the TLL state of spin ladders (mode A) are interpreted as massless  $t_{i+}$  bosons, while the gapped excitations (mode B) correspond to massive  $t_{i,0}$  bosons [56]. The former contribution is characteristic of the TLL state and is commonly found in spin-1/2 Heisenberg chains (and can be interpreted as originating from the Bose condensate of  $\Delta S^z = 1$  magnons), while the latter have  $\Delta S^z = 0$  magnons as their origin. The boson mass is determined by the Luttinger constant  $K$  (describing the nature of interactions between particles) and the velocity  $u$ ; both parameters are field dependent [38]. The complex contributions of these two variables to the gapped excitation give a hint for understanding the nonlinear dependence of mode B in a magnetic field as observed in our experiments.

To summarize, the excitation spectrum in DIMPY, a spin-1/2 Heisenberg antiferromagnetic strong-leg ladder compound, was probed by means of high-field ESR in magnetic fields up to 50 T. Two ESR modes were observed. One of them has a linear frequency-field dependence, and corresponds to Zeeman-split massless  $S_0^{\pm}$  excitations, commonly found in spin-1/2 Heisenberg chains and strong-rung ladders in the Tomonaga-Luttinger liquid regime. On the other hand, we show that a key property of the ESR spectrum in a spin-1/2 Heisenberg strong-leg ladder in the TLL phase is the presence of gapped  $S_{\pi}^{zz}$  excitations that derive from the gapped  $\Delta S^z = 0$  boson. Good agreement between the results of exact-diagonalization calculations and the experimental data was demonstrated.

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