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We present a ^{14}N nuclear magnetic resonance study of a single crystal of $CuBr_4(C_5H_{12}N)_2$ consisting of weakly coupled spin-$1/2$ Heisenberg antiferromagnetic ladders. When placed in a magnetic field, such a ladder is theoretically predicted to exhibit a quantum critical Luttinger liquid (LL) behaviour in the gapless phase, i.e. between the two critical fields. Treating ladders as LLs and interaction between them as a perturbation, we are indeed able to fully account for (i) the spin dynamics accessed by measuring the spin-lattice relaxation rate T_1^{-1}, and for (ii) the phase transition to a 3D ordered phase occuring below 110 mK due to the weak interladder coupling.

1 Introduction Applying an external magnetic field H to a spin-$1/2$ antiferromagnetic ladder decreases the spin gap between the singlet ground state and the lowest triplet ($S_z = -1$) excited states. At H_{c1} the ladder enters a magnetic gapless phase, while at H_{c2} it gets fully polarized and the gap reopens [1]. The gapless phase can be essentially mapped onto a partially filled spinless fermion band, described as a Luttinger liquid (LL) [2–5]. Low-energy physics of the LL is fully characterized by two interaction dependent LL parameters: the velocity of excitations u, and the dimensionless exponent K. Correlation functions decay as power laws, with exponents which are simple functions of K. The interaction term in the fermion picture is *uniquely* determined by the exchange coupling constants, which can be experimentally extracted [$J_\perp$ on the rungs and $J_\parallel$ on the legs of the ladder, see Figure 1(a)], and by H, which controls the filling of the fermion band. Once $J_\perp$ and $J_\parallel$ are known, the LL parameters can be obtained numerically for arbitrary H. The associated LL prediction can then be checked *quantitatively* over the *whole* fermion band, which extends between the critical fields H_{c1} and H_{c2}.

2 Experimental We performed a ^{14}N nuclear magnetic resonance (NMR) study of a single crystal of $CuBr_4(C_5H_{12}N)_2$ (BPCB). The crystal structure of BPCB contains weakly coupled spin-$1/2$ Heisenberg antiferromagnetic ladders formed by the pairs of spin-$1/2$ Cu^{2+} ions, which are stacked along the crystallographic a axis, as shown in Fig. 1(a) [6]. As ^{14}N has spin $I = 1$ and thus a quadrupole moment, ^{14}N NMR spectrum [upper spectrum in Fig. 1(b)] generally consists of four quadrupole doublets, for two crystallographically inequivalent nitrogen sites [N(1) and N(11) in Fig. 1(a)] in two crystallographically equivalent ladders (denoted by I and II), which are made physically inequivalent by the applied magnetic field $\mathbf{H}$. We chose the sample orientation with external magnetic field in the a^*b plane ($a^* \perp b, c$), at an angle of

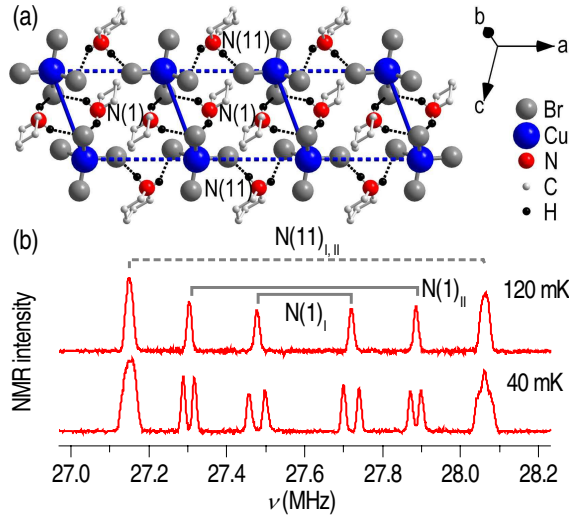

Figure 1 (a) A ladder formed by the supposed exchange interactions, $J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j$, between $S = 1/2$ spins of Cu^{2+} ions in the crystal structure of BPCB. Solid and dashed thick blue lines stand for $J_\perp$ and $J_\parallel$, respectively. The 10 protons attached to the C atoms are not shown. (b) ^{14}N NMR spectra at 120 mK and 40 mK recorded at $H = 9.0$ T.

9° to the b axis. In this orientation, the N(1) doublets are well resolved, while the N(11) doublets overlap as shown in Fig. 1(b). All the measurements presented here were performed on the $N(1)_I$ lines.

3 Luttinger liquid description We extract the values of H_{c1}, H_{c2} and the coupling ratio $J_\perp/J_\parallel = 3.6$ from the magnetic field dependence of the longitudinal (parallel to $\mathbf{H}$) uniform magnetization of Cu^{2+} ions, as measured at 40 mK via the ^{14}N hyerfine shift of $N(1)_I$ NMR lines [7]. Once the coupling ratio $J_\perp/J_\parallel$ is known, the variation of the LL parameters with H is determined and the LL theory for the gapless phase in BPCB is left without any adjustable parameter. For an isolated ladder with $J_\perp/J_\parallel = 3.6$ we numerically calculate $K(H)$ and $u(H)$ (in kelvin units), combining DMRG method with bosonization as in Ref. [5]. The result is displayed in Fig. 2(a) together with the corresponding result in the XXZ chain model (from Ref. [4]) obtained in the strong-coupling limit ($J_\perp \gg J_\parallel$) [4]. Close to H_{c1} (H_{c2}) the LL exponent approaches the value $K = 1$ of noninteracting fermion system indicating a nearly empty (full) fermion band.

4 Test of the Luttinger liquid model We test the LL behavior of BPCB via the dynamical spin-spin correlation functions, which are experimentally accessible through ^{14}N NMR observables due to the hyperfine coupling of spin-1/2 Cu^{2+} ions to the ^{14}N nuclei. For the above determined range of $K(H)$, the transverse (perpendicular to $\mathbf{H}$) staggered spin-spin correlation turns out to be the

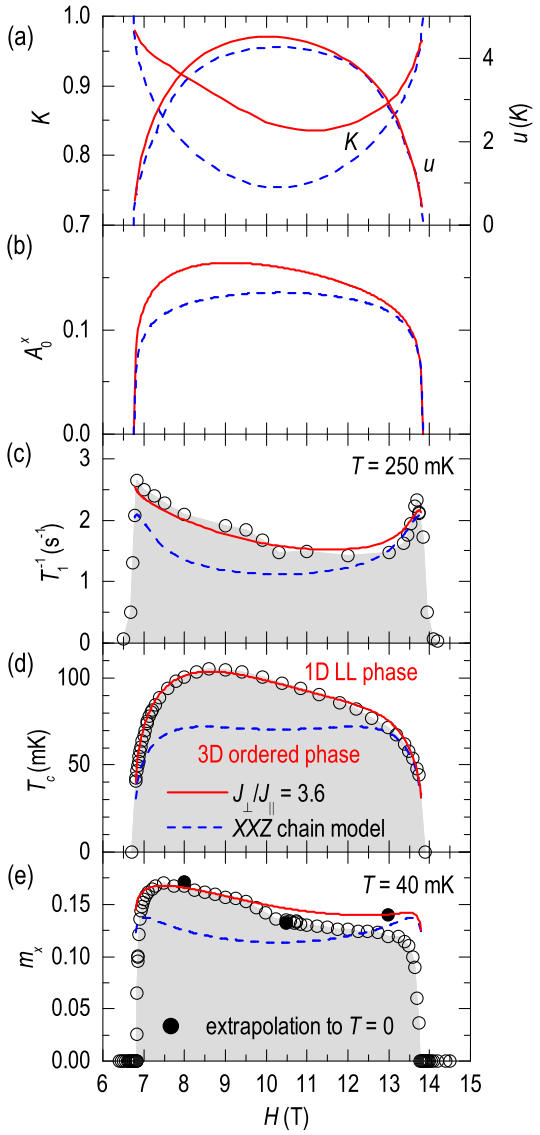

Figure 2 Variation (a) of the LL parameters $K(H)$ and $u(H)$ (in kelvin units) and (b) of the correlation amplitude $A_0^x(H)$ over the fermion band as calculated for $J_\perp/J_\parallel = 3.6$ (solid line) and for the XXZ chain model (dashed line). Magnetic field dependence of (c) ^{14}N T_1^{-1} at 250 mK (○), in the 1D LL phase, (d) the temperature of the transition between the 3D ordered phase and the 1D LL phase (○), (e) the transverse staggered magnetization m_x per Cu^{2+} ion at 40 mK (○) and its extrapolation to zero temperature (●), all measured on $N(1)_1$ site. The data sets in (c), (d) and (e) are compared to the corresponding predictions of the LL model based on $K(H)$, $u(H)$ [from (a)] and $A_0^x(H)$ [from (b)] for $J_\perp/J_\parallel = 3.6$ (solid line) and to the prediction of the simplified XXZ chain model (dashed line). The *only* adjustable parameter in each case is an overall scaling factor.

dominant one at low temperature [2,4]. In the following we focus on three ^{14}N NMR observables driven by this

spin-spin correlation alone. For the nuclear spin-lattice relaxation rate T_1^{-1} in the LL phase we find the following expression [7]:

$$T_1^{-1} = \frac{\hbar\gamma^2 A_\perp^2 A_0^x}{k_B u}\cos\Big(\frac{\pi}{4K}\Big)\,B\Big(\frac{1}{4K},1-\frac{1}{2K}\Big)\Big(\frac{2\pi T}{u}\Big)^{\frac{1}{2K}-1}, \tag{1}$$

where A_0^x is the amplitude of the correlation function, $\gamma/(2\pi) = 3.076$ MHz/T is ^{14}N nuclear gyromagnetic ratio, $A_\perp$ the transverse hyperfine coupling constant, and $B(x,y) = \Gamma(x)\Gamma(y)/\Gamma(x+y)$. The amplitude A_0^x is defined in the following way:

$$\langle S^x_{l,1} S^x_{l',1}\rangle = A_0^x \frac{(-1)^{l-l'}}{\mid l-l'\mid^{\frac{1}{2K}}},$$

where $S^x_{l,i}$ is the spin-$1/2$ operator at site l for the ladder leg $i = 1, 2$. The variation of A_0^x with H is numerically calculated along with $K(H)$ (as in Ref. [5]). Our result for the ladder with $J_\perp/J_\parallel = 3.6$ is shown in Fig. 2(b) together with the corresponding result in the XXZ chain model.

As the transverse staggered spin-spin correlation function diverges with decreasing temperature, any interladder exchange coupling (i.e., the coupling between the LLs) leads to the transverse staggered 3D magnetic ordering at low enough temperature [4], no matter how small the coupling is. Treating it in the mean-field approximation, we get the following expression for the temperature T_c of transition from the 1D LL phase to the 3D ordered phase [7]:

$$T_c = \frac{u}{2\pi}\left[\sin\Big(\frac{\pi}{4K}\Big)\,B^2\Big(\frac{1}{8K},1-\frac{1}{4K}\Big)\,\frac{zJ'A_0^x}{2u}\right]^{\frac{2K}{4K-1}}. \tag{2}$$

Here J' is the exchange coupling between the Cu^{2+} ions in neighboring ladders and z the coordination number. Although exchange paths between the ladders have not been identified yet, the structure of BPCB suggests that $z = 4$.

The order parameter in the 3D ordered phase is a transverse staggered magnetization m_x. Its $T = 0$ prediction $m_x = -(-1)^l(-1)^i\langle S^x_{l,i}\rangle$ can be expressed within the same mean-field approximation [7] as:

$$m_x = F(K)\sqrt{A_0^x}\left(\frac{\pi z J' A_0^x}{2u}\right)^{\frac{1}{8K-2}}, \tag{3}$$

where

$$F(K) = \left\{\frac{\frac{\pi^2}{\sin(\pi/(8K-1))}\frac{8K}{8K-1}\Big[\frac{\Gamma(1-(1/8K))}{\Gamma(1/8K)}\Big]^{\frac{8K}{8K-1}}}{\Gamma\big(\frac{4K}{8K-1}\big)^2\Gamma\big(\frac{16K-3}{16K-2}\big)^2}\right\}^{\frac{8K-1}{8K-2}}.$$

The emergence of the transverse staggered magnetic order breaks the inversion symmetry between the pairs of Cu^{2+} ions forming the rungs [6]. This leads to the splitting of ^{14}N NMR lines as demonstrated in Fig. 1(b) for the case when BPCB sample is cooled from 120 to 40 mK at $H = 9.0$ T. The splitting of ^{14}N NMR lines is proportional to the order parameter m_x [7].

Our measurements of T_1^{-1} at 250 mK, temperature T_c of the transition to a 3D ordered phase, and the order parameter m_x at 40 mK, all as a function of the magnetic field H in the gapless phase of BPCB, are compared to the LL predictions from Eqs. (1), (2) and (3) in Figs. 2(c), (d) and (e), respectively. The *only* adjustable parameter in each case is an overall scaling factor [7], while the shape of the three curves is fixed through the known pair of LL parameters $K(H)$ and $u(H)$ [from Fig. 2(a)] and the correlation amplitude $A_0^x(H)$ [from Fig. 2(b)] for the ladders with $J_\perp/J_\parallel = 3.6$. Excellent agreement over the whole field range for all three observables provides a remarkable confirmation of the LL model. The obtained $J' = 20$ mK compares well to the value obtained in neutron diffraction experiments [8]. In contrast, the LL prediction based on $K(H)$, $u(H)$ and $A_0^x(H)$ in the XXZ chain model [from Figs. 2(a) and (b)] fails to reproduce the biased shape of the three observables. This demonstrates the sensitivity of the observables to the applied set of LL parameters $K(H)$, $u(H)$ and, consequently, to the coupling ratio $J_\perp/J_\parallel$.

5 Conclusion We showed that ^{14}N NMR data for the spin-ladder material BPCB can be fully accounted for by treating the ladders in the gapless phase as LLs through bosonization and DMRG techniques. We thus demonstrated that BPCB is an excellent model system offering the possibility to control LL parameters in a continuous manner over the whole fermion band. Our results show that the whole physics of weakly coupled spin ladders can be captured in a *single* theory based on LLs, and provide the *first* quantitative check of the LL model.

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