Electron spin resonance in a strong-rung spin-1/2 Heisenberg ladder.

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$Cu(C_8H_6N_2)Cl_2$, a strong-rung spin-1/2 Heisenberg ladder compound, is probed by means of electron spin resonance (ESR) spectroscopy in the field-induced gapless phase above H_{c1}. The temperature dependence of the ESR linewidth is analyzed in the quantum field theory framework, suggesting that the anisotropy of magnetic interactions plays a crucial role, determining the peculiar low-temperature ESR linewidth behavior. In particular, it is argued that the uniform Dzyaloshinskii-Moriya interaction (which is allowed on the bonds along the ladder legs) can be the source of this behavior in $Cu(C_8H_6N_2)Cl_2$.

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Quantum spin ladders exhibit a variety of exotic strongly correlated states continuing to attract a great deal of attention. One of the most remarkable discoveries on this family of low-dimensional spin systems is superconductivity, observed in $Sr_{0.4}Ca_{13.6}Cu_{24}O_{41.84}$ under pressure [1]. Recently, spin ladders have been used to address a number of fundamental questions, related to field-induced phase transitions in quantum matter. In magnetic fields, spin ladders undergo a transition from a disordered gapped to gapless phase, where they can be mapped onto a system of interacting bosons with the density of states easily controlled by the applied magnetic field. At low-enough temperatures, when three-dimensional interactions become important, the system undergoes a transition into the magnetically ordered state (often recalled as to the magnon Bose-Einstein condensation [2]), while above these temperatures (when one-dimensional interactions become dominant) the quantum spin ladders provide a remarkable realization of the Tomonaga-Luttinger liquid (TLL) state [3].

Among other spin-Hamiltonian parameters, the anisotropy of spin-spin interactions (hereafter magnetic anisotropy) is known to play a particularly significant role [4, 5], being, for instance, of crucial importance for the realization of the magnon Bose-Einstein condensation in gapped quantum magnets (where the presence of the uniaxial U(1) symmetry, corresponding to the global rotational symmetry of the bosonic field phase, is one of the most essential conditions). Electron spin resonance (ESR) spectroscopy is traditionally recognized as one of the most sensitive tools to probe the magnetic anisotropy in magnets. In particular, a theory of the low-temperature ESR was developed for uniform spin-1/2 Heisenberg antiferromagnetic (AF) chains and spin-1/2 Heisenberg AF chains perturbed by a staggered magnetization [6–8]. The theory allowed to explain the effect of the anisotropy on the ESR linewidth and field shift in a number of spin-chain systems (see [8–10] and the references therein). A new approach to determine anisotropy parameters from the low-temperature ESR frequency shift in a spin-1/2 strong-rung ladder has been recently developed by Furuya et al. [11]. The theory was applied to $(C_5H_{12}N)_2CuBr_4$ (also known as BCPB or $(Hpip)_2CuBr_4$), indicating a good agreement with the experimental data [12]. It was shown also, that the anisotropy strongly affects the ESR linewidth in spin-1/2 strong-leg ladders [13], in particular in the TLL phase [14].

The strong-rung spin-1/2 ladder material, $Cu(C_8H_6N_2)Cl_2$ [hereafter $Cu(Qnx)Cl_2$, where Qnx indicates quinoxaline, $C_8H_6N_2$] with $J_r/k_B = 34.2$ K and $J_l/k_B = 18.7$ K [15], where J_r and J_l are the exchange coupling constants along the rungs and legs, respectively. This material is characterized by $H_{c1} \simeq 14.3$ T [15, 16], which can be easily reached using a superconducting magnet (the second critical field corresponding to the fully spin polarized phase is $H_{c2} \simeq 52$ T [17]). The relatively low H_{c1} makes this compound a perfect model system for studying spectral and magnetic properties of strong-rung spin-1/2 ladders in the field-induced gapless phase, where the TLL regime can be realized [18].

Series of $Cu(Qnx)Cl_2$ single crystals of typically 1 mm^3

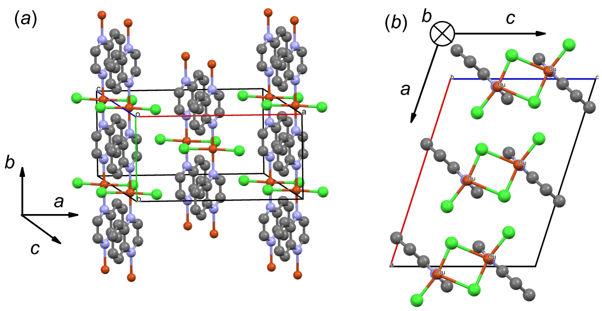

FIG. 1: (color online) Two views of the crystal structure of the spin-1/2 spin-ladder compound Cu(Qnx)Cl_2. The Cu, N, C, and Cl ions are shown in red, blue, gray, and green, respectively. The ladders run along the b-axis direction.

size were synthesized at the ETH Zürich using slow diffusion in methanol solution, as described in Ref. [16]. The crystal structure of Cu(Qnx)Cl_2 is shown schematically in Fig. 1. The compound belongs to the monoclinic space group $C2/m$ with unit-cell parameters a = 13.237 Å, b = 6.935 Å, c = 9.775 Å, and β = 107.88° ($Z = 4$), measured at room temperature [19, 20]. The spin-1/2 Cu^{2+} ions (shown in red in Fig. 1) are bridged by quinoxaline molecules, forming chains along the two-fold rotation axis b. Two chains are coupled into a two-leg ladder over two Cl^- ions in each rung.

The samples first were characterized using the X-band Bruker Elexsys E500 ESR spectrometer operated at a frequency of 9.4 GHz. High-quality, twin-free samples were chosen for high-field experiments. At room temperature X-band ESR experiments revealed $g_a = 2.16(1)$, $g_b = 2.03(1)$, and $g_c = 2.09(1)$. High-field ESR experiments were performed using a spectrometer, similar to that described in Ref. [21]. The spectrometer was equipped with a 16 T superconducting magnet, transmission-type probe in the Faraday configuration, and VDI radiation sources (product of Virginia Diodes Inc.). The magnetic field was applied along the b axis.

Examples of ESR spectra for the frequency 446.7 GHz are shown in Fig. 2. The ESR spectra were fit using the Lorentzian lineshape function. With decreasing temperature, starting at $T \sim J_l/k_B$ the ESR line shifts towards smaller magnetic fields (Fig. 3), indicating the enhancement of short-range spin correlations. These correlations appear also to be be responsible for the pronounced ESR narrowing revealed by us in Cu(Qnx)Cl_2 (Fig. 4).

As shown in Ref. [22], the Hamiltonian of a spin ladder for $H > H_{c1}$ can be reduced to an effective spin-1/2 Heisenberg AF chain Hamiltonian. To set the stage we can describe the low-temperature behavior of the system with the Hamiltonian $\mathcal{H}$ using the bosonization (or conformal field theory) description, namely

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{pert}, \tag{1}$$

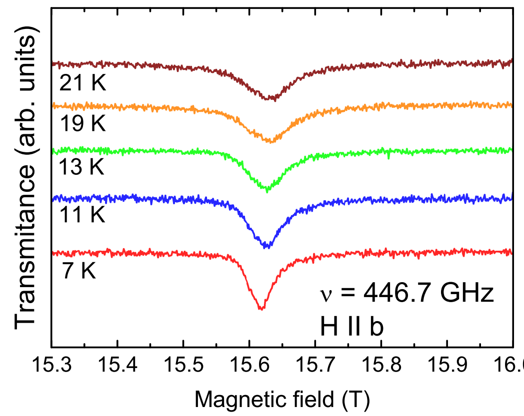

FIG. 2: (color online) Examples of ESR spectra obtained at 446.7 GHz.

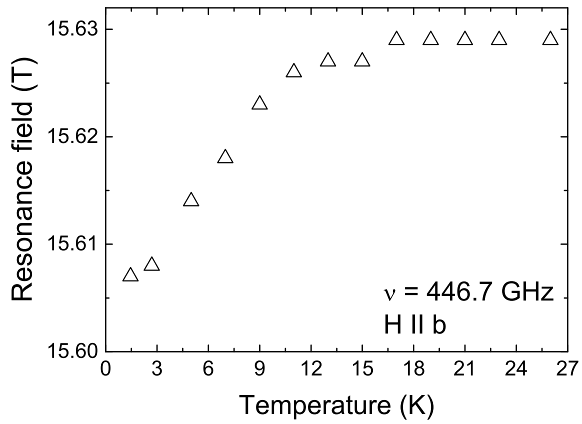

FIG. 3: ESR field as function of temperature obtained at 446.7 GHz.

where

$$\mathcal{H}_0 = \frac{v}{2} \int dx [\Pi^2 + (\partial_x \phi)^2] \tag{2}$$

is the Hamiltonian of the free boson field ϕ and its conjugated momentum Π. The velocity of low-lying spin excitations, v, can be written as

$$v = \frac{\pi J_{eff} \sqrt{1-\Delta^2}}{2 \arccos \Delta}. \tag{3}$$

Here, J_{eff} is the effective exchange interaction in the chain (proportional to the exchange coupling J_l, [22]) and Δ is a parameter describing the uniaxial anisotropy.

The perturbation can be written as

$$\mathcal{H}_{pert} = \lambda \int dx \cos(\sqrt{8\pi K}\phi) \ , \tag{4}$$

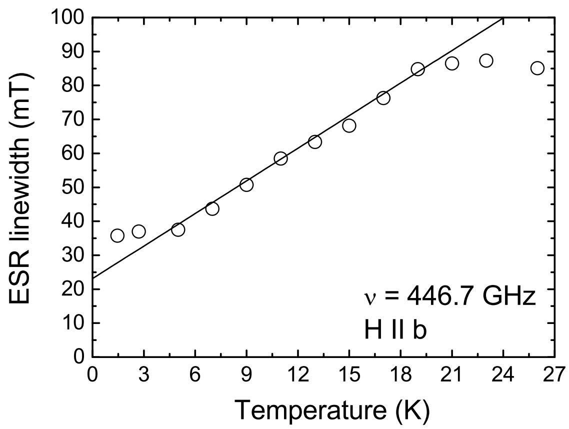

FIG. 4: ESR linewidth as function of temperature obtained at 446.7 GHz (symbols). The line is the fit results using Eq. (11) (see the text for details).

with the conformal field theory exponent, K, given by

$$K = \frac{\pi}{\pi - \arccos\Delta}. \quad (5)$$

The ESR intensity, $I(\omega, H, T)$, in the standard geometry can be expressed in the framework of the bosonization approach [6] for $H < J_l$ (we use the units in which $\hbar = g\mu_B = k_B = 1$) as

$$I(\omega, H, T) \propto -\chi(T, H)\omega \mathrm{Im} \frac{H^2 + \omega^2}{\omega^2 - H^2 - \Pi(\omega, H, T)} , \quad (6)$$

where $\chi(T, H)$ is the static susceptibility of the chain, ω is the frequency of the ac field and $vq = H$ (where q is the wave vector). In the above expression the self energy $\Pi(\omega, H, T)$ can be calculated in the framework of the perturbation theory with respect to λ. One can show that

$$\Pi(\omega, q) = 4\pi K v^2 \lambda^2 [F^r(\omega, q) - F^r(0, 0)] , \quad (7)$$

where $F^r(\omega, q)$ is the retarded propagator of the boson field given by

$$F^r(\omega, q) \propto -\frac{2^{4K-2} v}{\pi^2 T^2} \left(\frac{\pi T}{v}\right)^{4K} \Gamma^2(1 - 2K) \times$$
$$\times \frac{\Gamma(K - \frac{i(\omega+vq)}{2\pi})\Gamma(K - \frac{i(\omega-vq)}{2\pi})}{\Gamma(1 - K - \frac{i(\omega+vq)}{2\pi})\Gamma(1 - K - \frac{i(\omega-vq)}{2\pi})} ,$$

where $\Gamma(x)$ is the Gamma function. In the case of resonance we have to use $\omega = H$. Then, it is clear that the shift of the resonance position due to spin-spin interactions and anisotropy is given by the real part of the self energy, while the ESR linewidth is related to its imaginary part. In particular, the linewidth, ΔB, can be expressed as

$$\Delta B \approx \lambda^2 v^2 \frac{\sqrt{\pi} K}{2^{2K}} T^{4K-3} \left(\frac{2\pi}{v}\right)^{4K-2} \times$$
$$\frac{\Gamma(\frac{1}{2} - K)\Gamma^2(K)\Gamma(1 - 2K)}{\Gamma(1 - K)}. \quad (8)$$

We can consider the weak anisotropy $A = \Delta J_{eff}$ of the system as a perturbation with respect to the isotropic antiferromagnetic case $K = 1$. Notice that only the real (not effective) magnetic anisotropy reveals itself in the ESR experiments. Then, according to the above and neglecting corrections due to marginal perturbations, such as logarithmic ones, one finds

$$\Delta B \approx \left(\frac{A}{J_{eff}}\right)^2 T. \quad (9)$$

Such a linear dependence between ΔB and T should exist up to $T \approx J_{eff}$. Taking into account logarithmic corrections, one obtains

$$\Delta B(T) \approx A^2 \ln^2 \left(\frac{v}{T}\right) \frac{T}{v^2}. \quad (10)$$

The linear fit of the ESR linewidth behavior for 5 K $< T <$ 21 K [23], using the equation

$$\Delta B_{tot} = \Delta B_0 + \frac{\delta(\Delta B)}{\delta T} T, \quad (11)$$

gives $\Delta B_{TI} = 21.7(5)$ mT and $\frac{\delta(\Delta B)}{\delta T} = 3.2(1)$ mT/K, for the temperature-independent and temperature-dependent contributions, respectively (Fig. 4). The first, temperature-independent, contribution can be of the van Vleck origin. Accordingly to Eq. (9), the second term of Eq. (11) gives

$$A \approx 0.066 J_{eff}. \quad (12)$$

Assuming $J_{eff} = J_l = 18.7$ K, one obtains $A \approx 1.2$ K.

One source of ESR line broadening is the exchange anisotropy, which can be roughly estimated using the formula $D_E \approx J_{eff}(\Delta g/g)^2$ (where Δg is the deviation of the g factor from the free electron value and assuming $J_{eff} = J_l$) [24], giving $D_E/k_B \sim 0.1$ K. On the other hand, it is worthwhile to mention that the uniform Dzyaloshinskii-Moriya (DM) interaction is allowed on the bonds along the ladder legs in $Cu(Qnx)Cl_2$ by the symmetry. The rough estimate using the formula $D_{DM} \approx J_{eff}(\Delta g/g)$ [24] gives for the DM interaction $D_{DM}/k_B \sim 1.5$ K. This value agrees well with the one obtained using Eq. (12), $A \approx 1.2$ K, suggesting that the uniform DM interaction can be the source of the ESR linewidth dependence observed by us in $Cu(Qnx)Cl_2$.

In conclusion, we have presented systematic ESR studies of the spin-1/2 strong-rung Heisenberg ladder compound $Cu(C_8H_6N_2)Cl_2$. Employing the quantum field theory approach, we argue that the anisotropy of magnetic interactions plays a crucial role, determining the peculiar ESR linewidth behavior at low temperatures.

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