Spin Dynamics of a S=1/2 Antiferromagnetic Heisenberg Ladder: ^{1}H NMR in $Cu_2(C_5H_{12}N_2)_2Cl_4$

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We have studied the low-energy dynamical properties of the spin-1/2 two-leg ladder material $Cu_2(C_5H_{12}N_2)_2Cl_4$ by ^{1}H Nuclear Magnetic Resonance (NMR). Both exchange constants and spin-triplet gap $\Delta \approx 10.5$K have been determined previously from susceptibility and high-field magnetization measurements. The nuclear spin-lattice relaxation rate T_1^1 has been measured below and above the critical field $H_{c1} = 7.5\text{T} \equiv \Delta$. In the "gapped" regime ($T \ll H_{c1} - H$), the evolution of $1/T_1$ is governed by the longitudinal correlator $S_{zz}(q, \omega_n)$. Close and above H_{c1}, $1/T_1$ is progressively dominated by staggered processes which contribute to $S_\perp(q, \omega_n)$. It is shown a relaxation rate which diverges at low temperature with a power law dependence ($1/T_1 \propto T^{-\alpha}$). Experimental values of α are agreement with quantum phase transition theory predictions.

The crossover from one to two dimensions in $S = 1/2$ Heisenberg antiferromagnets (HAF) was recently predicted not to be smooth in the sense that an even number of $S = 1/2$ HAF chains coupled by any exchange $J_\perp$ possess a non-magnetic ($S = 0$) ground state, with a gap to $S = 1$ excitations, while an odd number of such chains behave essentially as a single chain [1,2]. A number of experiments [2,3] have now convincingly born out this remarkable conjecture, also reminiscent of the distinction between integer (disordered) and half-integer (quasi-ordered) quantum spin chains [4]. There remain however several unresolved issues raised by experiments. For instance, susceptibility and spin-lattice relaxation measurements seem to indicate different values of the spin gap in ladder materials [3,5], and in some Haldane chains as well [6,7]. Another point of interest are the dynamical properties of such antiferromagnets in the presence of a large magnetic field. In fact, if static properties like susceptibility or magnetization may not give a clear idea of what is the microscopic Hamiltonian [8], low-frequency dynamical properties as revealed by nuclear magnetic relaxation can, in principle, discriminate between these systems *if* the magnetic field H is larger than the gap energy Δ [9]. A finite magnetic field lifts the S=1 states degeneracy and a field-induced second order phase transition occurs at $H_{c1} = \Delta/g\mu_B$ where the lowest branch of the triplet crosses the ground state. At the critical field H_{c1}, the system experiences a $T = 0$ quantum phase transition defined by universal critical exponents [10,11].

In contrast with other ladder materials, the organometallic $Cu_2(C_5H_{12}N_2)_2Cl_4$ [12] is the unique representative of S=1/2 antiferromagnetic Heisenberg ladders with relatively small exchange constants ($J_\perp \simeq 13.2$K, $J_\parallel \simeq 2.5$K, $\Delta \simeq 10.5 \pm 0.3$K) and enables experimental studies in the "magnetic" phase ($g\mu_B H > \Delta$) [13].

NMR experiments have been performed with the magnetic field direction along the $\vec{b}$ axis (perpendicular to the ladder direction) of a set of five small single crystals (typically 100-200μg each). We used a conventionnal pulse spin-echo sequence in the range 180−410MHz. The spectra exhibits relatively well defined lines corresponding to the various proton sites.

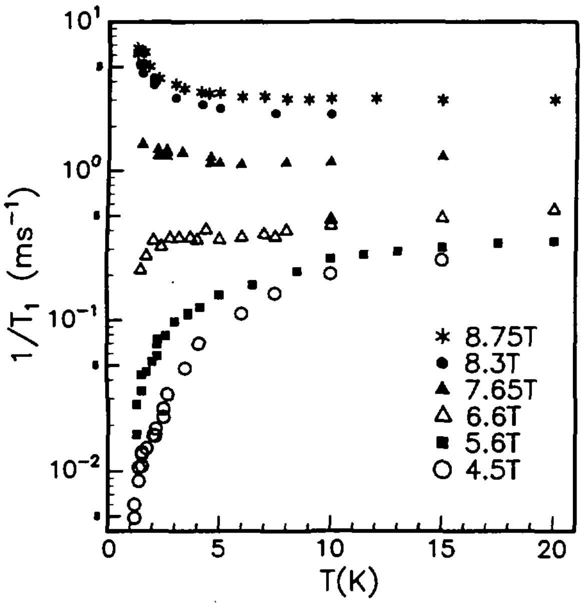

FIG. 1. ^{1}H spin-lattice nuclear relaxation rate $1/T_1$ as a function of T for different magnetic field. Below H_{c1}, low temperature $1/T_1$ goes as $\exp(-\Delta_{eff}/T)$ where $\Delta_{eff} \approx \Delta - g\mu_B H$. Above H_{c1}, $1/T_1$ diverges at low temperatures with a field dependent exponent α.

The temperature evolution of $1/T_1$ at fixed magnetic fields is shown in Fig. 1. The nuclear spin-lattice relaxation is expressed in terms of the magnetic structure factor components [14]

$$\frac{1}{T_1} = \gamma_N^2 \sum_{q,\alpha} |F_{\alpha\alpha}(q)|^2 \cdot S_{\alpha\alpha}(q, \omega_n)\,, \tag{1}$$

where $|F_{\alpha\alpha}(q)|^2$ are hyperfine form factors of dipolar origin [15] and $\alpha = z, \pm$ represents the longitudinal and transverse components, respectively.

The structure factor $S_{\alpha\alpha}(q,\omega_n)$ is related to the Fourier transform at ω_n of the two-spin correlation functions $< S_{z,+}(q,t)S_{z,-}(-q,0) >$ which is given in terms of matrix algebra by

$$S_{\alpha\alpha}(q,\omega_n) \approx \sum_{k,\sigma,\sigma'} |< k+q,\sigma \mid S_q^\alpha \mid k,\sigma' >|^2 \times \delta(E_{k+q,\sigma} - E_{k,\sigma} - \hbar\omega_n)e^{-E_{k,\sigma}/k_BT} , \quad (2)$$

where $\mid k,\sigma\rangle$ represents a one-magnon state with $\sigma = 0,\pm1$ and energy $E_{k,\sigma}$. The one-magnon dispersion spectrum in the strong coupling regime $J_\perp \gg J_\parallel$ is given by perturbation theory, $E_{k,\sigma} = J_\perp + J_\parallel \cos(k) + \sigma g\mu_B H$, and is minimum at $k = \pi$ [13].

In the gapped phase $T \ll H_{c1} - H$, only excitations near $k = \pi$ with small momentum transfer q contribute significantly to the relaxation [16]. In such limit, the low-T limit is dominated by "uniform" ($q \approx 0$) transitions within the lowest Zeeman branch $\sigma = -1$ (intra-branch transitions, $\Delta M_S = 0$). These matrix elements contribute only to the longitudinal correlator $S_{zz}(q,\omega_n)$ with a low-T Boltzmann factor $\exp(-\Delta_{eff}/T)$. This regime is characterized by a fall-off of all correlators and all thermodynamical quantities. Fig. 1 shows that $1/T_1$ is linearly reduced by magnetic field. In this regime, relaxation is driven by $S_{zz}(q \approx 0,\omega_n)$ with a linearly reduced effective gap $\Delta_{eff} = \Delta - g\mu_B H$.

In the magnetic phase $H > H_{c1}$ the ground state becomes magnetic, gapless and populated by excitations. At T=0, the number of excitations n in the ground state is directly related to the chemical potential $\mu = g\mu_B H - \Delta$ in a free Fermions picture [17,11] and "staggered" processes dominates the region $k_BT \approx |H - \Delta|$ relaxation. In this situation, one would expect a relaxation enhancement related to the non-zero magnetization ($M \propto n$) of the electronic system. At low temperatures $1/T_1$ turns upward in contrast with the gapped phase (see Fig. 1). The low temperature divergence is already visible below 2-3K very close to H_{c1} and becomes more prononced at higher fields, extending to higher temperatures. This persistence of the divergent behavior might be specific of ladders [9] and could not originates from the onset of 3D ordering [19]. At $H = 7.6\ T \approx H_{c1}$, $\alpha \approx 0.5$: this is in good agreement with theoretical predictions based on impenetrable Bosons (*i.e* free Fermions) $T_1 \sim \sqrt{T}$ ($T \to 0$) [18]. This region refers to the "Luttinger liquid" regime.

In conclusion, we have demonstrated in the spin-1/2 two-leg Heisenberg ladder $Cu_2(C_5H_{12}N_2)_2Cl_4$ the possibility to study dynamical properties through a quantum phase transition induced by magnetic field. The transition from a "gapped" regime to a "Luttinger" regime has been identified by proton NMR. More, these low-temperature regimes are dominated by different contributions of the correlation functions.

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