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# Critical properties and Bose Einstein Condensation in dimer spin systems

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We analyze the spin relaxation time  $1/T_1$  for a system made of weakly coupled one dimensional ladders. This system allows to probe the dimensional crossover between a Luttinger liquid and a Bose-Einstein condensate of magnons. We obtain the temperature dependence of  $1/T_1$  in the various dimensional regimes, and discuss the experimental consequences.

Bose-Einstein condensation (BEC) is a particularly spectacular manifestation of quantum physics and the indiscernibility of the particles. Traditional systems where to observe such a phenomenon are bosonic systems such as<sup>1</sup> Helium 4 or more recently cold atomic gases<sup>2</sup>. However BEC is also present in many other condensed matter systems. Superconductivity can be viewed as a BEC of Cooper pairs and BEC has also long been sought in excitonic systems<sup>3</sup>. Because spin-1/2 operators can be faithfully represented by hard core bosons<sup>4</sup>, spin systems are also very natural systems where to look for such a phenomenon<sup>5,6,7</sup> and various applications have thus been investigated. However, in practice, both the experimentally large exchange constants and, on a more theoretical level the hard core constraint, which corresponds to very strong repulsion of the bosons, restrict considerably the usefulness of these concepts for simple regular spin systems. In the extreme case of one dimension it is in fact much more fruitful to think of spins-1/2 in terms of spinless fermions<sup>8</sup> than in terms of bosons.

Nevertheless, it has been realized<sup>7</sup> that the BEC physics is extremely useful to describe the spin systems made of coupled spin-1/2 dimers. In such systems the application of a magnetic field induces a zero temperature quantum phase transition (QPT) between a spin-gap phase with zero magnetization and a phase where some of the dimers are polarized in a triplet state giving a nonzero magnetization. This transition has been shown<sup>7</sup> to be in the universality class of BEC, where the bosons represent the dimers in the triplet state. The BEC order parameter is then the staggered magnetization in the plane orthogonal to the applied field. These predictions have been spectacularly confirmed by magnetization<sup>9</sup> and neutron<sup>10</sup> measurements on  $\text{TlCuCl}_3$  and related dimer compounds. The active discovery and analysis of new compounds in which this phenomenon could be observed thus triggered further theoretical<sup>11,12,13,14,15,16,17,18,19</sup> and experimental work<sup>20,21,22,23,24,25,26,27,28,29,30</sup> on this magnon BEC.

In connection with the above mentioned magnon BEC,

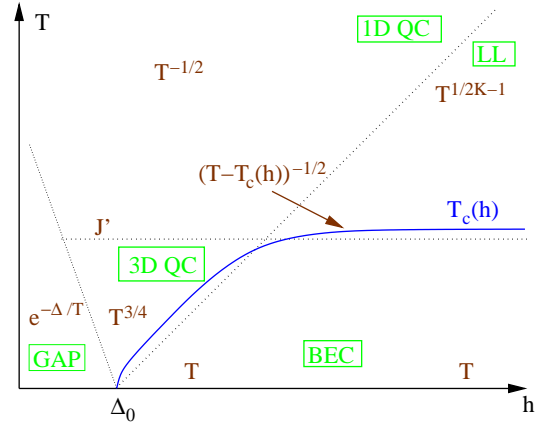


FIG. 1: Summary of the temperature dependence of  $1/T_1$ .  $T_c(h)$  represents the BEC condensation temperature.  $\Delta_0$  is the gap in the absence of magnetic field. The temperature dependence is indicated for the various regimes (see text).

an important question is to understand how the dimensional crossover, between the one dimensional and the three dimensional behavior can occur in systems made of weakly coupled ladders. From a theoretical point of view, in such systems, at temperatures much larger than the inter-ladder coupling, the system can be viewed as a collection of one dimensional ladders, with the very peculiar associated one dimensional behavior<sup>7,31,32,33</sup>, more fermionic in nature, and in the universality class of Luttinger liquids at high field<sup>8</sup>. At lower temperature, interladder coupling cannot be ignored anymore, and the system falls into the universality class of magnon BEC condensates. From the experimental point of view, such a change of regime between the one dimensional limit and the three dimensional one is relevant for compounds such as BPCP<sup>34</sup> that are a very good realization of weakly coupled two legs ladders, with strong dimers along the ladder rungs. To probe these different regimes and the dimensional crossover, it is crucial to understand the

spin dynamics as a function of temperature and magnetic field. An important probe in that respect is provided by Nuclear Magnetic Resonance (NMR) measurements. Quantities such as the relaxation rate  $1/T_1$  have been computed in the condensed phase and in the one dimensional Luttinger liquid regime<sup>7,32</sup>. However, predictions for their behavior close to the BEC quantum critical point or in the other one-dimensional regimes are still lacking, although some results for two dimensional systems have recently appeared<sup>35</sup>.

In this paper we thus calculate the NMR relaxation rate  $1/T_1$  as a function of temperature and magnetic field in a system made of weakly three-dimensionally coupled ladders, in the one-dimensional and in the three-dimensional regime. Our findings are summarized in Fig. 1

We consider a system of coupled one dimensional two leg ladders described by the Hamiltonian

$$H = J \sum_{\alpha,i,l=1,2} \mathbf{S}_{\alpha,i,l} \cdot \mathbf{S}_{\alpha,i+e_x,l} + J_{\perp} \sum_{\alpha,i} \mathbf{S}_{\alpha,i,1} \cdot \mathbf{S}_{\alpha,i,2} - h \sum_{\alpha,i,l=1,2} S_{\alpha,i,l}^z + H' \quad (1)$$

where  $\alpha$  is the ladder index,  $l = 1, 2$  denote the two legs of the ladder,  $e_x$  is the direction parallel to the ladder leg and  $h$  the applied magnetic field.  $H'$  is the interladder exchange term. We take here the simple form for  $H'$  :

$$H' = J' \sum_{\alpha,i} \mathbf{S}_{\alpha,i,2} \cdot \mathbf{S}_{\alpha,i+e_y,1} + J'' \sum_{\alpha,i,l} \mathbf{S}_{\alpha,i,l} \cdot \mathbf{S}_{\alpha,i+e_z,l} \quad (2)$$

where  $e_y$  is the direction of the rungs, and  $e_z$  is the direction orthogonal to the ladder planes. We take for simplicity  $J' = J''$ , generalization is immediate. The NMR relaxation rate  $T_1$  can be expressed<sup>36</sup> as a function of the retarded spin-spin correlation function. We concentrate in this paper on the contribution of the transverse local spin-spin correlation,

$$\frac{1}{T_1} = T \lim_{\omega \rightarrow 0} \sum_q \frac{\text{Im} \langle S^+(q, \omega) S^-(q, \omega) \rangle}{\omega}, \quad (3)$$

as one can show that the longitudinal contribution  $\langle S^z S^z \rangle$  gives only subdominant corrections as far as the temperature dependence is concerned<sup>37</sup>.

When the temperature is larger than the interladder coupling  $J'$  the system can be essentially described by a set of independent ladders. This one-dimensional regime is well understood<sup>7,31,32</sup>. For fields  $h$  lower than the spin gap of the ladder  $\Delta_0$ , and at sufficiently low temperature, the system remains in the spin gap state, and  $1/T_1$  has an activated form<sup>38</sup>. For fields  $h > \Delta_0$ , and such that  $h - \Delta_0 \gg T$ , the system is in a Luttinger liquid regime<sup>7,32,33</sup> and one finds  $1/T_1 \sim T^{1/(2K)-1}$  for  $h > \Delta_0$ , with  $K$  the Luttinger exponent. Close to the critical field the ladder system can be mapped to a two state system for which only the singlet and lowest triplet are

important<sup>7,39,40,41,42</sup>. This allows to use a pseudo spin 1/2 representation. For high temperatures,  $T \gg |h - \Delta_0|$ , the system is in a universal quantum critical regime<sup>31</sup> with a dynamical exponent  $z = 2$ . There,  $1/T_1$  varies as a power law with a universal exponent which can be determined by a scaling argument. Indeed, scaling requires a local spin correlation function of the form  $T^{-1/2} f(\omega/T)$  in one dimension. The requirement that  $f$  be analytical in  $\omega$  for  $T > 0$  with a vanishing imaginary part for  $\omega = 0$  implies  $T/\omega \times \text{Im} f(\omega/T) \rightarrow \text{Ct.}$ , leading to  $1/T_1 \sim T^{-1/2}$ . We note that since at the commensurate-incommensurate transition induced by the magnetic field, one predicts<sup>7,32</sup> that  $K = 1$ , the behavior of  $1/T_1$  is continuous at the crossover from the Luttinger liquid to the  $z = 2$  quantum critical regime. The exponent simply becomes constant once the  $z = 2$  quantum critical regime has been reached. Such a behavior is shown in Fig. 1.

At low temperature the system enters a three dimensional regime, where the interladder coupling must be considered from the start. Two distinct regimes occur depending on the magnetic field. If  $h - \Delta_0 \gg J'$ , then the system can be viewed as a collection of one dimensional ladders, with a fully developed Luttinger regime for each ladder. The coupling between the ladders then induces a three dimensional ordering for the magnetization in the XY plane, in the same universality class than a Bose-Einstein condensation<sup>7</sup>. The one dimensional fluctuations strongly affect the three dimensional ordering temperature to lead to  $T_{BEC} = J(J'/J)^{1/(2-2K)}$ . This regime can essentially be described by an RPA treatment of the one dimensional ladders. If  $h - \Delta_0 \ll J'$  then the coupling between the ladder is too strong for the ladders to have a one dimensional regime. One must then view the system as an anisotropic, but three dimensional, spin system. Note that this regime is universal and depends only on the presence of dimers in a three dimensional environment and not on whether a ladder structure exists. The results we obtain in this paper for the 3D quantum critical regime thus also apply to compounds for which  $J_{\perp} \gg (J, J')$  without any specific relation between  $J$  and  $J'$ . A useful representation<sup>7,9</sup> is to introduce a boson excitation for each rung in the ladder. The presence (resp. absence) of the boson indicates that the rung is the triplet (resp. singlet) state. In the regime with three-dimensional coupling, and above the BEC condensation temperature, we can describe the coupled dimer system (1) using the following effective Hamiltonian<sup>7,9,16</sup>:

$$H = \sum_k (\epsilon(k) - h) b_k^{\dagger} b_k + \frac{g}{L^3} \sum_{k_1, k_2, q} b_{k_1+q}^{\dagger} b_{k_2-q}^{\dagger} b_{k_2} b_{k_1}, \quad (4)$$

where  $\epsilon(k) = \Delta_0 + \frac{k^2}{2m}$  is the dispersion of the  $S^z = 1$  gapped magnons,  $b_k^{\dagger}$  creates a magnon excitation with momentum  $k$  and  $g$  is an effective repulsion between the magnons. Since the spin operator  $S^+$  is proportional to the magnon creation operator  $b^{\dagger}$ , the correlation function in (3) can be obtained from the Matsubara Green's function of the magnon operator. Above the BEC tem-

perature, anomalous propagators are vanishing, and this Matsubara Green's function is obtained as a function of

the magnon self-energy  $\Sigma(k, \omega + i0)$  from the usual Dyson equation. We therefore have:

$$\frac{1}{T_1} \sim \lim_{\omega \rightarrow 0} \frac{T}{\omega} \int \frac{d^3 k}{(2\pi)^3} \frac{\text{Im}\Sigma(k, \omega)}{(\omega + h - \epsilon(k) - \text{Re}\Sigma(k, \omega))^2 + \text{Im}^2\Sigma(k, \omega)}. \quad (5)$$

Just above the BEC temperature, the magnon density is small, making a low density expansion applicable. At first order<sup>9</sup> only Hartree-Fock diagrams are present, however they do not contribute to the imaginary part of the self-energy. The first non-trivial contribution to the imaginary part of the magnon self-energy comes from the second order graphs shown on Fig. 2. In order to evalu-

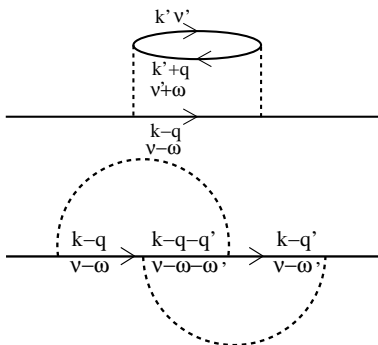


FIG. 2: Skeleton self-energy graphs at second order. The dotted line represents the interaction and the straight line the magnon propagator. The magnon propagator is computed with the first order self energy. Because of the low density limit close to the quantum critical point this expansion is well behaved.

ate the expression (5), we use the fact that when  $\omega \rightarrow 0$ ,  $T\text{Im}\Sigma(k, \omega)/\omega$  has a finite limit. This implies that the denominator in Eq. (5) is dominated by the contribution coming from the real part of the Green's function. Since the contribution coming from the denominator is strongly peaked near  $k = 0$  when  $h$  is near  $\Delta_0$ , the dominant contribution in Eq. (5) comes from the small momenta. The width in momentum of the expression in the numerator<sup>37</sup> being of order  $T^{2/3}$ , it is less strongly peaked near  $k = 0$  than the contribution from the denominator for  $T \rightarrow 0$ . This justifies the replacement in the numerator, of  $T\text{Im}\Sigma(k, \omega)/\omega$  by its value for  $k = 0, \omega = 0$ , yielding  $1/T_1 \sim T[m^3/2(\Delta - h)]^{1/2} \lim_{\omega \rightarrow 0} \text{Im}\Sigma(k = 0, \omega)/2\pi\omega$ , with  $\Delta$  the effective gap containing the Hartree-Fock renormalization of  $\Delta_0$ .

Following the same steps as in Ref. 43, the graphs of Fig. 2 are evaluated in the limit  $k \rightarrow 0$  as:

$$\text{Im}\Sigma(k = 0, \omega) = \frac{m^3 g^2 T}{4\pi^3} (1 - e^{-\omega/T}) \int_0^\infty d\epsilon_1 \frac{e^{-(\Delta + \epsilon_1 - \omega)/T}}{(1 - e^{-(\Delta + \epsilon_1 - \omega)/T})(1 - e^{-(\Delta + \epsilon_1)/T})} \ln \left( 1 + \frac{1 - e^{-(\epsilon_1 + \Delta - \omega)/T}}{e^{\frac{1}{T} \left( \frac{(\omega - \Delta)^2}{4\epsilon_1} + \Delta \right)} - 1} \right), \quad (6)$$

In the vicinity of the quantum critical point,  $\Delta \ll T$  and the integral (6) yields:

$$\frac{1}{T_1} \sim m^{9/2} g^2 \frac{T^3}{(\Delta - h)^{3/2}} \quad (7)$$

Let us note that by a power-counting argument<sup>44</sup>, we expect the real part of the  $n$ -th order correction to the self-energy to behave as  $g^n T^{1+n/2}$ . This implies that at low temperature the Hartree-Fock contribution dominates so that  $\Delta(T) - h \sim gT^{3/2}$  in the 3D quantum critical regime. Therefore, in the 3D quantum critical regime:

$$1/T_1 \sim T^{3/4}. \quad (8)$$

In the BEC regime, we find instead that  $\Delta(T) - h \propto (T - T_{BEC}(h))$  and  $1/T_1 \propto (T - T_{BEC}(h))^{-3/2}$ . This behavior is valid when  $T - T_{BEC}(h) \gg |\Delta_0 - h|$  where  $\Delta_0$  is the bare gap. This result breaks down when  $T$  is sufficiently close to  $T_{BEC}(h)$ . Then, one has the mean-field behavior  $1/T_1 \sim (T - T_{BEC}(h))^{-1/2}$ . Below the BEC condensation temperature, due to the presence of a massless Goldstone mode associated with phase fluctuations, one can show<sup>7</sup> that  $1/T_1 \sim T$ . At the other extreme, in the gapped regime,  $|\Delta - h| \gg T$  and evaluating (6) yields  $1/T_1 \propto T^{3/2} e^{-3\Delta/T}$ . Our findings are summarized on Fig. 1 for all the different regimes.

The predictions of Fig. 1 are directly testable on coupled ladder systems. For systems made of weakly coupled ladders such as BPCB<sup>34</sup>, given the accessible magnetic fields and with the condition  $J' \ll J$ , the one dimensional regime is probably the easiest to probe. Thus the clearest test is the  $T^{-1/2}$  divergence in the quantum critical regime. In principle, one could also observe the gradual change of the exponent as one moves inside the Luttinger regime towards quantum criticality. However, since the exponent varies between 1/2 and 1/3, this change is rather small and this effect is likely to be hard to observe. Unless one is very close to the quantum critical point the pure one dimensional divergence will merge with the divergence as  $1/(T - T_c)^{1/2}$  at the finite temperature BEC transition due to the three dimensional coupling. For systems for which one is able to probe both the one dimensional and the three dimensional regimes, a clear signature of the dimensional crossover will be the divergence of  $1/T_1$  in the one dimensional regime followed by a drop of  $1/T_1$  in the quantum critical 3D regime. Note that going towards the BEC regime will reveal a more com-

plex behavior where the initial drop when entering the 3D quantum critical regime will be followed by a divergence at  $T = T_{BEC}$  and then by another drop inside the BEC phase. Finally more isotropic systems would show a broad 3D regime, allowing to test the quantum critical prediction  $T^{3/4}$  in the quantum critical regime or  $T$  in the BEC one. Unfortunately in  $\text{TiCuCl}_3$  the 3D quantum critical regime is affected by a simultaneous lattice distortion<sup>45</sup> and one must find other candidates to probe for this regime. An interesting one might be provided by hydrated copper nitrate<sup>46,47</sup>.

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