On the estimate of the spin-gap in quasi-1D Heisenberg antiferromagnets from nuclear spin-lattice relaxation

R. Melzi and P. Carretta

Department of Physics "A. Volta", Unitá INFN di Pavia, 27100 Pavia, ITALY

Abstract

We present a careful analysis of the temperature dependence of the nuclear spin-lattice relaxation rate $1/T_1$ in gapped quasi-1D Heisenberg antiferromagnets. It is found that in order to estimate the value of the gap correctly from $1/T_1$ the peculiar features of the dispersion curve for the triplet excitations must be taken into account. The temperature dependence of $1/T_1$ due to two-magnon processes, is reported for different values of the ratio $r = J_1/J_2$ between the superexchange constants in a 2-leg-ladder. As an illustrative example we compare our results to the experimental findings for $^{63}$Cu $1/T_1$ in the dimerized chains and 2-leg-ladders contained in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$.

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The many peculiar aspects of quasi one-dimensional quantum Heisenberg antiferromagnets (1DQHAF) have stimulated an intense research activity during the last decade [1]. Moreover, the recent observation of superconductivity in the 2-leg-ladder compound $(\text{Sr},\text{Ca})_{14}\text{Cu}_{24}\text{O}_{41}$ [2] and the occurrence of a phase separation in high temperature superconductors (HTSC) in hole-rich and hole-depleted regions analogous to spin-ladders [3], have brought to a renewed interest on 1DQHAF. One of the relevant issues is whether the spin-gap observed in some of these 1DQHAF is related to the one observed in the normal state of HTSC [4]. For these reasons many NMR groups working on HTSC have focused their attention on these systems and on the determination of the spin-gap values in pure and hole-doped compounds [5–16]. However, since the early measurements, a clear discrepancy between the values for the gap ($\Delta$) estimated by means of nuclear spin-lattice relaxation ($1/T_1$) and susceptibility (or Knight shift) measurements has emerged [5]. In many compounds the gap estimated by means of $1/T_1$ using the activated form $1/T_1 \propto \exp(-\Delta/T)$ derived by Troyer et al. [17], turned out to be $\approx 1.5$ times larger than the one estimated by using susceptibility or inelastic neutron scattering measurements (see Tab. 1). Many attempt models, theoretical [18] or phenomenological [19], have tried to explain these differences, however, while they were able to describe the findings for some compounds they were not able to explain the results obtained in other gapped 1DQHAF. In fact, as can be observed in Tab. 1, while for certain 2-leg-ladders [13] an agreement between the gap estimated from $T_1$ and through other techniques is found, in several other systems it is not [3–12]. It is interesting to observe that the 1DQHAF where the agreement is observed are the ones in the strong coupling limit, namely either dimerized chains or 2-leg-ladders with a superexchange coupling along the rungs much larger than the one along the chains. Therefore, one can conclude that the disagreement is not always present and has to be associated with the peculiar properties of the spin excitations in each system, i.e. with the form of the dispersion curve for the triplet excitations. In this manuscript we will show that the discrepancy relies essentially on the use for $1/T_1$ of an expression which is valid in general only at very low temperatures ($T \ll 0.2\Delta$) and its application to higher temperatures depends on the form of the dispersion curve for the triplet spin excitations. In particular, for dimerized chains the validity of a simple activated expression extends to higher temperatures than for a 2-leg-ladder. As an illustrative example we will analyse the temperature dependence of $1/T_1$ for the $^{63}$Cu nuclei in the dimerized chains (Cu(1)) and in the 2-leg-ladders (Cu(2)) contained in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ [19].

In the following we will consider the contribution to nuclear relaxation arising from 2-magnon Raman processes only. Namely, we will assume that although the system is not in the very low temperature limit ($T \ll \Delta$), the temperature is low enough ($T \ll \Delta$) so that 3-magnon processes as well as the spin damping can be neglected. If the large value of the gap derived by means of $1/T_1$ was due to these contributions, which are proportional to $\exp(-2\Delta/T)$, one should observe some discrepancy also for the 1DQHAF in the strong coupling limit, at variance with the experimental findings (see Tab. 1). The approach we use follows exactly the same steps outlined in the paper by Troyer et al. [17] where, by assuming a quadratic dispersion for the triplet excitations (valid for $T \ll \Delta$), namely

$$E(k_x) = 1 + \alpha(k - \pi)^2$$

(1)

in units of $\Delta$, they found that

$$1/T_1 = \frac{3\gamma^2A^2}{4\alpha\pi^2} \frac{h}{k_B\Delta} \exp(\omega_o/2T) \exp(-\Delta/T) \left(0.80908 - ln(\omega_o/T)\right)$$

(2)

with $\omega_o$ the resonance frequency and $A_o$ the hyperfine coupling constant. We remark that there is a factor 4
difference with respect to the equation reported by Troyer et al. [7], which is related to a different definition of the hyperfine Hamiltonian and of the dispersion curve. The values of the hyperfine constants are $A_\parallel = 120$ Koe for $^{63}$Cu(2) and $A_\perp = 29$ Koe for $^{63}$Cu(1) [3,4]. In the case of a general form for the dispersion relation, by considering that the low-energy processes are the ones corresponding to an exchanged momentum $q \simeq 0$ and $q \simeq -2k_x$, one can write the contribution related to 2-magnon Raman processes in the form [17]

$$\frac{1}{T_1} = \frac{3\gamma^2 A_0^2}{\pi^2 k_B \Delta} \int_0^\pi dk_x \frac{e^{-E(k_x)/T}}{\sqrt{v^2(k_x) + 2\omega_o^2 \partial E(k_x)/\partial k_x}}$$

(3)

where $E(k_x)$ is the dispersion relation for the triplet spin excitations, normalized to the gap value, whereas $v(k_x) = \partial E(k_x)/\partial k_x$. For a 2-leg-ladder a general form describing $E(k_x)$ is

$$E(k_x)^2 = E(k_x = 0)^2 \cos^2\left(\frac{k_x}{2}\right) + \sin^2\left(\frac{k_x}{2}\right) + c_o \sin^2(k_x)$$

(4)

which is strongly dependent on the ratio $r = J_\perp/J_\parallel$ between the superexchange coupling along the rungs and along the legs. We have taken the dispersion curves derived by Oitmaa et al. [20] from extensive series studies and estimated the parameters $E(k_x = 0)$ and $c_o$ accordingly. Then, starting from Eqs. 3 and 4, by means of a numerical integration one can derive directly $1/T_1$ for a 2-leg-ladder for different values of $r$. It should be remarked that for $r$ of the order of unity the dispersion curve for the triplet excitations has a maximum around a wave-vector $k_m$ [20] (see Fig. 1) and also low-energy processes from $k_m - k_x$ to $k_m + k_x$ could contribute to the relaxation. However, this processes should become relevant only at $T \gtrsim \Delta$, where also 3-magnon processes and the damping of the spin excitations become relevant.

In Fig. 2 we report the results obtained on the basis of Eqs. 3 and 4 for $^{63}$Cu(2) for different values of the superexchange anisotropy $r$. One observes that while for the dimerized chains, corresponding to the limit $r \gg 1$, $1/T_1$ follows an activated behavior as the one given in Eq. 2, for the 2-leg-ladders with $r \sim 1$ one observes some differences with respect to the simple activated behavior already at temperatures $T \lesssim \Delta/4$. This analysis points out that for a 2-leg-ladder with $r$ of the order of unity it is not correct to estimate the gap from 1/$T_1$ by using Eq. 2, at least for $T \lesssim \Delta/4$. In fact, it is noticed that the quadratic approximation for the dispersion curve becomes valid for a more restricted range of $k_x$ around $\pi/a$ as $r$ decreases (see Fig. 1). This seems to contradict the results reported in Fig. 2a, where the departure from the quadratic approximation is found more pronounced for $r = 1$ than for $r = 0.5$. However, this artifact is related to the choice of the horizontal scale, namely to have reported 1/$T_1$ vs. $\Delta/T$, since $\Delta$ increases with $r$ [1]. In fact, if we report 1/$T_1$ vs $J_\parallel/T$ (Fig. 2b), with $J_\parallel$ independent of $r$, one immediately notices that the deviation from the quadratic approximation starts at lower temperatures for the lowest value of $r$.

One can then analyse the experimental data on the basis of Eq. 3 by taking the value for the gap estimated by other techniques and check if there is an agreement. We have fit the experimental data for $^{63}$Cu(2) (Fig. 3b) and $^{63}$Cu(1) (Fig.3a) in Sr$_{14}$Cu$_{24}$O$_{41}$ by taking $\Delta = 450$ K and $\Delta = 120$ K, respectively, as estimated from susceptibility or NMR shift data [1-11]. In both cases we find a good agreement between theory and experiment by taking $1 \geq r \geq 0.5$ for the ladder site and $r \gg 1$ for the chain site. If the data for $^{63}$Cu(2) were fitted according to Eq. 2 one would derive a value for the gap around 650 K, a factor 1.5 larger than the actual value (see Tab. 1).

For $r = 1$ also a quantitative agreement with the experimental data for $^{63}$Cu(2) is found. However, this fact seems to be at variance with the estimates by Johnston [21] based on the analysis of DC susceptibility data and with the recent findings by Inmi et al. [10] based on the study of $^{17}$O NMR shift anisotropy, where a value for $r \simeq 0.5$ was derived. If we take this value for $r$ we find that the experimental data are a factor $\simeq 8$ larger than expected. This disagreement could originate, at least partially, from having considered for the $q_x = 2k_x$ processes the values for the $|< -k_x|[S_z][k_x]|^2$ matrix elements estimated by Troyer for the case $r = 1$ [7]. One has also to mention that the estimate of the hyperfine coupling constants could suffer from some uncertainties, particularly the contribution from the transferred hyperfine interaction with the neighbouring Cu$^{2+}$ spins. This contribution should be particularly relevant for the $^{63}$Cu(1) nuclei while it should be small for $^{63}$Cu(2). However, it must be recalled that since 1/$T_1$ depends quadratically on the hyperfine coupling constant even for $^{63}$Cu(2) sizeable corrections can be expected. Finally it has to be observed that in these systems the low-frequency divergence of 1/$T_1$ is cut because of the finite coupling among the ladders (or chains), introducing another correction to the absolute value of 1/$T_1$.

The low-frequency divergence of 1/$T_1$ was found to follow the logarithmic behavior reported by Troyer et al. [7] (see also Eq. 2) and does not change upon varying the anisotropy factor $r$, for $r > 0$. In fact, the form of this divergence is related to the shape of the dispersion curve close to $k_x = \pi/a$, where it is always correctly approximated by a quadratic form for $r > 0$.

In conclusion we have presented a careful analysis of the problem of estimating the spin-gap from nuclear spin-lattice relaxation measurements in 1DQHAF. It is found that in order to estimate correctly the gap one should either perform the experiments at temperatures $T \lesssim 0.2\Delta$ where in many cases other contributions to the relaxation process emerge [12,14], or use an appropriate ex-
pression for $1/T_1$ which takes into account the form of the dispersion curve for the triplet excitations. Then a good agreement for the gap value estimated by means of $1/T_1$ and other techniques is found, allowing also to derive information on the anisotropy of the superexchange constants.

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TABLE I. Values for the gap $\Delta$ between singlet and triplet excitations for different 1DQHAF, estimated from $1/T_1$ using Eq. 2 and from DC susceptibility (or NMR shift) measurements. In the last column the ratio for the values of the gap estimated by the two techniques is reported.

FIG. 1. The dispersion curves for the triplet excitations in a 2-leg-ladder are reported for different values of the superexchange anisotropy $r = J_\perp / J_\parallel$. The dotted line shows the quadratic approximation of the dispersion curve for $r >> 1$ (see Eq. 1).

FIG. 2. a) Nuclear spin-lattice relaxation rate as a function of $\Delta/T$ for three different values of the ratio $r = J_\perp / J_\parallel$. The solid lines give the results obtained from Eqs. 3 and 4 after a numerical integration for $\omega_0/2\pi = 15$ MHz, while the dotted lines show the corresponding behavior by using the quadratic approximation (see Eq. 2). b) The same data as in a) are now reported as a function of $J_\parallel/T$.

FIG. 3. a) Temperature dependence of $^{63}\text{Cu}(1) 1/T_1$ in a Sr$_{14}$Cu$_{24}$O$_{41}$ single crystal for $\vec{H} \parallel b$. The solid line shows the behavior expected from Eq. 3 by taking a gap $\Delta = 120$ K. b) Temperature dependence of $^{63}\text{Cu}(2) 1/T_1$ in Sr$_{14}$Cu$_{24}$O$_{41}$ for $\vec{H} \parallel b$. The data were obtained either in oriented powders (circles) or single crystals (squares). The solid line shows the behavior according to Eq. 3 for $r = 0.5$ by using the same value for the gap derived from $^{63}\text{Cu}(2)$ NMR shift ($\Delta = 450$ K), while the dotted line gives the corresponding behavior obtained using the quadratic approximation (Eq. 2) with $\Delta = 650$ K.
Fig. 1 (Melzi and Carretta)
Fig. 3 (Melzi and Carretta)

(a) Graph showing the relation between $1/T_1$ (in s$^{-1}$) and $T$ (in K).

(b) Graph showing the relation between $1/T_1$ (in s$^{-1}$) and $T$ (in K), with two lines indicating different values of $\Delta$: $0.5$ and $450$ K, and $650$ K.
| Compound                              | $\Delta T_1$ | $\Delta \chi$ | $\Delta T_1/\Delta \chi$ |
|--------------------------------------|---------------|----------------|--------------------------|
| Sr$_{14}$Cu$_{24}$O$_{41}$ (2-leg-ladder) | 650 K         | 450 K          | 1.45 Refs. 7-10          |
| Sr$_{14}$Cu$_{24}$O$_{41}$ (dimerized chain) | 120 K         | 120 K          | 1 Refs. 7-10             |
| VO(HPO$_4$)0.5H$_2$O (dimerized chain) | 75 K          | 75 K           | 1 Ref. 16                |
| Cu(CHN)Cl (2-leg-ladder)             | 11 K          | 11 K           | 1 Ref. 15                |
| CaV$_2$O$_5$ (dimers)                | 650 K         | 660 K          | 1                        |
| SrCu$_2$O$_3$ (2-leg-ladder)         | 700 K         | 450 K          | 1.55 Ref. 5              |
| AgVP$_2$S$_6$ (S=1 chain)            | 400 K         | 320 K          | 1.25 Ref. 6              |
| YBa$_2$NiO$_5$ (S=1 chain)           | 200 K         | 100 K          | 2.0 Ref. 13              |

Tab. 1