Finite-size calculations of spin-lattice relaxation rates in Heisenberg spin-ladders

Martin P. Gelfand* and Mohan Mahadevan†

Department of Physics, Colorado State University, Fort Collins, Colorado 80523

(Dated: October 24, 2018)

Abstract

Calculations of nuclear spin-lattice relaxation rates are carried out by means of exact diagonalization on small (2 × 6) antiferromagnetic Heisenberg ladders, using the simplest forms permitted by symmetry for the hyperfine couplings for the three nuclear sites in Cu$_2$O$_3$ ladders. Several values of the rung/chain exchange ratio $J_\perp/J_\parallel$ have been considered. Comparisons with experimental results, field theoretic calculations, and the Gaussian approximation highlight some open problems.

PACS numbers: 75.10.Jm, 75.40.Gb, 75.40.Mg, 76.60.-k
I. INTRODUCTION

Spin-ladder systems, in particular the two-leg, $S = 1/2$, antiferromagnetic variety, have been the subject of considerable theoretical and experimental investigation. Spin ladders are appealing because they are one-dimensional systems and thus can be effectively investigated using many powerful theoretical tools, while offering a wider parameter space of “simple,” and potentially experimentally realizable, Heisenberg Hamiltonians than spin chains. The simplest Heisenberg ladder Hamiltonian has the form

$$H = \sum_n J_\parallel (S_{n,1} \cdot S_{n+1,1} + S_{n,2} \cdot S_{n+1,2}) + J_\perp S_{n,1} \cdot S_{n,2}$$

(1)

which offers a dimensionless parameter $J_\perp / J_\parallel$ which is in principle tunable by chemistry or pressure. In addition, compounds containing weakly coupled Cu$_2$O$_3$ ladders are appealing because of possible connections with cuprate superconductivity.

The present work was motivated by the nuclear spin-lattice relaxation measurements in La$_6$Ca$_8$Cu$_{24}$O$_{41}$, an undoped ladder compound, by Imai et al. These measurements were carried out for all of the nuclear sites on the ladder, namely the copper, the “rung” oxygen, and the “ladder” (or “chain”) oxygen, over a wide temperature range, from low temperatures up to nearly 900 K. Because the principal exchange interactions in cuprates are so large, on the order of 1000 K, it is quite challenging to do experimental work at temperatures significantly greater than the spin gap ($\Delta \approx 500$ K).

The experimental results (see Figure 1(c) of Imai et al.) have the following noteworthy features. At temperatures below about 425 K, the relaxation rates for all three sites follow a common (activated) temperature dependence up to a scale factor. However, on increasing $T$ the copper $1/T_1$ (which we will refer to as $1/CuT_1$) exhibits a rather sharp departure from that of the two oxygen sites ($1/O(1)T_1$ and $1/O(2)T_1$ for ladder and rung, respectively). There seems to be a nearly discontinuous decrease in the derivative of $1/CuT_1$; moreover, above 425 K the $1/CuT_1$ data appear nearly linear with an almost vanishing intercept. The relaxation rates for the two oxygen sites, in contrast, exhibit no particular features in the vicinity of 425 K.

Several aspects of the wavevector dependence of the low-frequency spin susceptibility can be gleaned directly from the data.

One can express the spin-lattice relaxation rate in terms of the dynamic structure factor
for the Cu$^{2+}$ spins

$$\frac{1}{T_1} \propto \int dq H_n(q) S(q, \omega_n)$$

(2)

where $H_n$ is the hyperfine form factor associated with nucleus $n$, $\omega_n$ is the NMR frequency (which we will take to be zero in everything that follows), and $S$ is the structure factor. The proportionality constants can be neglected for our present purposes. The spin correlations are isotropic, so there is no need to consider the various components, $S^{xx}$ and so forth, individually. The hyperfine interactions are not isotropic, so the orientation of the magnetic field in the NMR experiment does affect the results; however, all of the results of present interest can be obtained with a single field orientation, which then specifies $H_n(q)$ uniquely.

The largest hyperfine couplings are between a given nuclear site and the closest spins; at that level of approximation, and taking the intra- and inter-chain lattice constants to be of unit length, one has

$$H_{Cu} = A^2, \quad H_{O(1)} = 4C^2 \cos^2(q_x/2),$$

$$H_{O(2)} = 4F^2 \cos^2(q_y/2) + D^2$$

(3)

where $C$, $F$, and $D$ are the hyperfine couplings identified in Fig. 1(a) of Imai et al.\textsuperscript{4} $A$ is the on-site hyperfine interaction for copper, and we have elided the orientation dependence of the hyperfine interactions (so, for example, $A^2$ should really be $A_x^2 + A_y^2$ if the static field is along the $z$ axis).

The essential difference between copper and oxygen sites is that in the latter the hyperfine interaction in the vicinity of $q = (\pi, \pi)$ is much smaller than in the vicinity of $q = (0, 0)$. If, at all temperatures of experimental relevance, $S(q, 0)$ had most of its weight in the vicinity of $q = (0, 0)$, then then all three relaxation rates would have tracked one another. The marked decrease of $1/T_1$ relative to the other two relaxation rates at 425 K indicates that this cannot be the case, and in fact suggests that at temperatures below 425 K the ratio of the spectral weight near $(\pi, \pi)$ to that near $(0, 0)$ is roughly constant and of order unity, while above 425 K the ratio falls markedly. (The decrease is crucial. If there were an increase in $1/T_1$ relative to the oxygen rates with increasing $T$, one could ascribe that to a turn-on of $S((\pi, \pi), 0)$ for $T \gtrsim \Delta$ but $S((\pi, \pi), 0)$ might have been negligible compared to $S((0, 0), 0)$ at lower temperatures.)

Why the emphasis on $q = (0, 0)$ and $(\pi, \pi)$? In gapped systems such as spin ladders, the low-energy spin fluctuations are Raman processes, and at low temperatures one needs to
consider only the lowest energy magnons, namely those near \( q = (\pi, \pi) \). Spin fluctuations near \( (0,0) \) are associated with two-magnon processes, and those near \((\pi, \pi)\) with three-magnon processes, and on the face of it one would be justified in neglecting the three-magnon processes entirely at low temperatures: see Ref. 3 and references cited therein. However, as we have just seen, this appears to be inconsistent with the experimental data for \( \text{La}_6\text{Ca}_8\text{Cu}_{24}\text{O}_{41} \), and it is also inconsistent with the quantum Monte Carlo calculations of spin-lattice relaxation in a particular Heisenberg ladder \((J_\perp/J_\parallel = 1)\) by Sandvik, Dagotto, and Scalapino at least at temperatures greater than half the magnon gap.

An extensive theoretical treatment of spin dynamics in gapped one-dimensional Heisenberg models, including spin ladders, has been presented by Damle and Sachdev. Their analysis of \( S(q, \omega \approx 0) \) was restricted to \( q \) near \((0,0)\), but they did find the quite interesting result that the activation energy for \( 1/T_1 \) is larger, by a factor of \( 3/2 \), than the activation energy for the uniform static susceptibility (which is simply the spin gap). An analysis of \( S(q, \omega \approx 0) \) for \( q \) near \((\pi, \pi)\), for systems with \( J_\parallel \gg J_\perp \) has been presented by Ivanov and Lee. Their results are suggestive of a fairly sharp crossover from low- to high-temperature regimes at \( T \approx \Delta \), and also indicate that the \((\pi, \pi)\) contribution to \( 1/T_1 \) “overshoots” its \( T = \infty \) value and thus decreases as \( T \to \infty \).

In the present work, we have applied exact diagonalization to evaluate spin-lattice relaxation rates, following the method of Sokol, Gagliano and Bacci. We have considered three different ladder Hamiltonians, namely \( J_\perp/J_\parallel = 0.5, 1.0, \text{ and } 2.0 \), and have obtained \( 1/T_1 \) for Cu, O(1), and O(2) sites taking the simplest conceivable hyperfine couplings, namely \( A = C = F = 1 \), with all other interactions neglected. All of the calculations were for rather small systems, \( 2 \times 6 \), such that exact diagonalization could be carried out in an extremely straightforward manner.

It was noted above that calculations of spin lattice relaxation rates for spin ladders have already been carried out by means of large scale quantum Monte Carlo, but those calculations were limited to the Cu sites. Our goal is somewhat different than that of Sandvik, Dagotto, and Scalapino’s work. We are not trying to fit the data in detail, rather we want to see what can be learned from modest numerical calculations. One reason not to fit the data is that to get the gap correct to 10% by exact diagonalization for \( J_\perp/J_\parallel = 0.5 \) would require a system at least \( 2 \times 12 \). Another is that we do not treat the spin diffusion contribution to the relaxation rates correctly: our calculations effectively introduce an artificial cut-off
so that we obtain a finite spin-lattice relaxation rate. Finally, the precise form of the spin Hamiltonian for the cuprate ladder compounds is still subject to argument. Although the Knight-shift results of Imai et al. appear to be consistent with the simple spin-ladder Hamiltonian of Eq. (1) for $J_{\perp}/J_{\parallel} \approx 0.5$, it has been suggested by Brehmer et al. that instead $J_{\perp}/J_{\parallel} \approx 1$ and in addition there is a modest amount of plaquette “ring exchange” in the Hamiltonian. A quantum-chemical analysis of the exchange interactions in various cuprates provides some support for the latter proposal, since it concludes that $J_{\perp}/J_{\parallel} \approx 0.9$ for Sr$_{14}$Cu$_{24}$O$_{41}$ (which is a lightly-self-doped version of the undoped La$_6$Ca$_8$Cu$_{24}$O$_{41}$ compound).

To be precise, the goals of our calculation are as follows. First, we want to verify that $S(q,0)$ has significant weight near $q = (\pi, \pi)$ as well as near $(0,0)$ and see if there are any noticeable trends with varying $J_{\perp}/J_{\parallel}$. Second, we want to explore the crossover from low to high temperature behavior in $1/T_1$: can we see anything like the experimental results, or like the theoretical results of Ivanov and Lee? Third, we want to keep our eyes open for any unanticipated patterns that might emerge in the numerical results.

II. METHOD OF CALCULATION AND RESULTS

The finite-size calculations of spin-lattice relaxation rates are carried out following Sokol, Gagliano, and Bacci. Rather than repeating their discussion of the method let us make a few remarks. We take $J_{\parallel}$ as the unit of energy.

The first step in the calculation is a complete diagonalization of the Hamiltonian and evaluation of matrix elements for certain local spin operators (depending on which nuclear site one is interested in). For the $2 \times 6$ lattices all of the calculations could be done using the simplest possible representations of the states in terms of local $S^z$ values; it was not even necessary to use translational invariance to classify states by wave vector.

The second step is the construction of an auxiliary function which Sokol et al. refer to as $I(\omega)$. This is implicitly dependent on $T$ and the hyperfine couplings. We considered temperatures ranging from 0.3 to 50. Typically we constructed $I(\omega)$ at intervals of 0.02 in $\omega$ up to at least $\omega = 0.6$.

Finally, one needs to estimate the zero-frequency derivative of $I(\omega)$, because $1/T_1$ is proportional to $T(\partial I/\partial \omega)|_{\omega=0}$. At high temperatures $I(\omega)$ is quite smooth, but at temper-
atures comparable to the gap significant structure develops (see Fig. 1). In order to avoid introducing spurious temperature dependences into \(1/T_1\) it is important to use a consistent procedure for extracting the derivative from the data. What we did was to fit a zero-intercept line through all the data points up to a cutoff \(\omega_{\text{max}}\), weighting all points equally in the fit. We did all of the calculations using both \(\omega_{\text{max}} = 0.5\) and 0.3. While there are noticeable differences in the results using these two cutoffs, as shown in Fig. 2, our conclusions turn out the same no matter which is chosen. The use of a much smaller cutoff, which might seem to be preferred on the grounds that one is really looking for a zero-frequency derivative, is not beneficial. The structure that develops in \(I(\omega)\) as \(T\) is lowered, making it look like a Devil’s staircase, is a finite-size artifact and must be averaged over, using a suitably large \(\omega_{\text{max}}\), to obtain results that are representative of the thermodynamic limit.

We now turn to the results of the calculations for the three nuclear sites and three values of \(J_{\perp}\) considered (0.5, 1.0, and 2.0). In every case we take \(\omega_{\text{max}} = 0.5\). In Fig. 3 we present results on a linear temperature scale, for \(T \leq 2\). The behavior of the spin-lattice relaxation rate at high temperatures is a bit surprising: comparing the plots in Fig. 3(a) through (c) it is apparent that while the Cu and O(1) rates decrease strongly as \(J_{\perp}\) increases, the trend for the O(2) rate is different. This is made more explicit in Fig. 4, where we show \(1/T_1\) for all three sites as a function of \(J_{\perp}\) at \(T = 50\) (effectively infinite temperature). In contrast, at low temperatures \(1/T_1\) decreases with increasing \(J_{\perp}\) at all sites, as one would expect since the spin gap is an increasing function of \(J_{\perp}\).

III. DISCUSSION AND CONCLUSIONS

It is evident that for \(J_{\perp} = 0.5\) and 1.0, \(1/T_1\) for all three sites is nearly equal for temperatures below the spin gap. (Of course we do not claim that this holds to arbitrarily low temperatures, just that it seems correct for temperatures as low as we dare to estimate \(1/T_1\).) Because of our choice of hyperfine interactions, this suggests that in such cases the weight in \(S(q, 0)\) for \(q \approx (\pi, \pi)\) is approximately three times that for \(q \approx (0, 0)\). This is in quantitative agreement with the results of Sandvik et al. at \(J_{\perp} = 1.0\). However, the story is rather different at \(J_{\perp} = 2.0\), where the spin-lattice relaxation rates for all three sites, including the two oxygen sites, are significantly different even at \(T = \Delta/2\). In the strong-coupling limit, then, the simple picture for \(S(q, 0)\) in which its weight is concentrated
at \((0, 0)\) and \((\pi, \pi)\) does not work even for temperatures that are a modest fraction of \(\Delta\).

What can we say about the low-to-high temperature crossover in the spin-lattice relaxation rates? First of all, the sort of behavior seen experimentally, in which \(1/T_1\) for the oxygen sites track each other closely while \(1/CuT_1\) splits off, appears to be a special feature of \(J_\perp \approx 1\) in the present calculations; it is not at all generic and does not hold for the putative experimental value \(J_\perp \approx 0.5\). Second, in no case does \(1/CuT_1\) exhibit any sort of sharp “break” as seen experimentally; nor does \(1/CuT_1\) exhibit linear-in-\(T\) behavior (with zero intercept, or otherwise) in the high temperature regime, even over a restricted temperature range (say \(\Delta\) to \(2\Delta\)). Finally, in no case does \(1/CuT_1\) exhibit an “overshoot” during the crossover: the spin-lattice relaxation rate associated with all sites monotonically increases with \(T\).

Our calculations thus suggest that there are quite a few open problems in this field. Almost none of the prominent experimental facts concerning \(1/CuT_1\) in \(La_6Ca_8Cu_{24}O_{41}\) are reproduced in our finite-size calculations. Furthermore, the work of Ivanov and Lee⁴ does not seem to have much to say about our results, either. Their calculation is controlled only in the \(J_\perp \ll 1\) regime, so we should only look at the \(J_\perp = 0.5\) data. Here we have no evidence of overshoot in \(1/CuT_1\), and no reason to believe that one can just examine the spectral weight near \((\pi, \pi)\) since \(1/O(2)T_1\) “peels off” from \(1/O(1)T_1\) in a manner not very different from \(1/CuT_1\).

At this point we face several alternatives. It is possible that our results are simply unreliable, because we are considering systems that are too small (especially for \(J_\perp = 0.5\)) and our procedure for estimating \(dI(\omega)/d\omega\) is flawed. We cannot rule this out, but we strongly suspect that the trends in the results as a function of \(J_\perp\) are robust. It is possible that the spin Hamiltonian for the ladders in \(La_6Ca_8Cu_{24}O_{41}\) is more complicated than the model we have considered. Whether the Hamiltonian of Brehmer et al.⁷ can reproduce the spin-lattice relaxation data requires another calculation. Another possibility that must be considered, given the remarkably sharp feature in \(1/CuT_1\) found in the experimental data, is that \(La_6Ca_8Cu_{24}O_{41}\) undergoes, by coincidence, a subtle structural transition at 425 K. This could introduce an anomalously strong \(T\)-dependence to the hyperfine interactions, though why the effect should be so much stronger in \(H_{Cu}(q)\) than \(H_{O(1)}(q)\) and \(H_{O(2)}(q)\) is difficult to envision.

Let us now turn to the results of our calculations for spin-lattice relaxation at very
high temperatures, shown in Fig. 4. The most natural way to think about these results is in
terms of the Gaussian approximation. The basic idea of this approach is to assume that
\[ \int dq H_n(q) S(q, \omega) \] is a Gaussian function of \( \omega \), and then evaluate the frequency cumulants of
this function by means of short-time expansions of time-dependent correlation functions. At
\( T = \infty \) the calculations are especially simple, because the expectation values of correlators
\( \langle S_i \cdot S_j \rangle \) vanish for sites \( i \neq j \). For three sites of interest in Heisenberg ladders, the Gaussian
approximation yields the following exchange dependences of the spin-lattice relaxation rates
at \( T = \infty \):

\[ \frac{1}{C_{\text{Cu}}} T_1 \propto 1/\sqrt{1 + \frac{1}{2} J_\perp^2}, \]  
\[ \frac{1}{O(1)} T_1 \propto 1/\sqrt{1 + J_\perp^2}, \]  
\[ \frac{1}{O(2)} T_1 \] does not have any \( J_\perp \) dependence at all. (Recall that \( J_\parallel \equiv 1 \); in all of these
results there is an overall factor of \( 1/J_\parallel \).) If this last result seems peculiar, let us note that it
can be derived in another way, by considering the strong-\( J_\perp \) limit. Then one most naturally
thinks about the states in terms of singlets and triplets on the rungs. The relevant energy
scale for the dynamics of the total spin on a rung, which is relevant to \( 1/O(2) T_1 \), would seem
to be proportional to \( J_\parallel \) (that is, the bandwidth in lowest-order perturbation theory for a
triplet excitation in a single background), and with the hypothesis of a single energy scale
in \( \int dq H_{O(2)} S(q, \omega) \) one reproduces the Gaussian approximation result.

We see in Fig. 4 that \( 1/T_1 \) for the copper and ladder oxygen sites decreases with increasing
\( J_\perp \), qualitatively in agreement with the Gaussian approximation, although the dependence
on \( J_\perp \) is not as strong as that approximation suggests. Furthermore, \( 1/O(2) T_1 \) exhibits an
increase with \( J_\perp \). The rather poor performance of the Gaussian approximation is somewhat
disappointing, considering how well it works for estimating spin-lattice relaxation rates in
square-lattice Heisenberg antiferromagnets. It is not too surprising, perhaps, given that
the dynamic correlations in the \( S = 1/2 \) Heisenberg chain are far from Gaussian at \( T = \infty \). So, there is yet another open problem in the area of low-energy spin dynamics of Heisenberg
ladders.

Acknowledgments

This work was supported by the US National Science Foundation through grant DMR
94–57928. We thank T. Imai for several stimulating discussions and also for communicating
the results of his group’s experiments prior to publication.

\[ gelfand@lamar.colostate.edu \]

† Permanent address: KLA-Tencor, 3 Technology Drive, Malpitas, California 95035

1 E. Dagotto and T. M. Rice, Science 271, 618 (1996).

2 T. Imai, K. R. Thurber, K. M. Shen, A. W. Hunt, and F. C. Chou, Phys. Rev. Lett. 81, 220 (1998).

3 D. A. Ivanov and P. A. Lee, Phys. Rev. B 59, 4803 (1999).

4 A. W. Sandvik, E. Dagotto, and D. J. Scalapino, Phys. Rev. B 53, R2934 (1996).

5 K. Damle and S. Sachdev, Phys. Rev. B 57, 8307 (1998).

6 A. Sokol, E. Gagliano, and S. Bacci, Phys. Rev. B 47, 14646 (1993).

7 S. Brehmer, H.-J. Mikeska, M. Müller, N. Nagaosa, and S. Uchida, Phys. Rev. B 60, 329 (1999).

8 Y. Mizonu, T. Tohyama, and S. Maekawa, Phys. Rev. B 58, R14713 (1998).

9 P. W. Anderson and P. R. Weiss, Rev. Mod. Phys. 25, 269 (1953).

10 T. Moriya, Prog. Theor. Phys. 16, 23 (1956).

11 T. Moriya, Prog. Theor. Phys. 16, 641 (1956).

12 T. Barnes, E. Dagotto, J. Riera, and E. S. Swanson, Phys. Rev. B 47, 3196 (1993).

13 M. P. Gelfand and R. R. P. Singh, Phys. Rev. B 47, 14413 (1993).

14 J. M. R. Roldan, B. M. McCoy, and J. H. H. Perk, Physica 136A, 255 (1986).
FIG. 1: The auxiliary function $I(\omega)$ for the Cu site with $J_\perp = 1.0$ and $T = 0.5$ (squares), 1.0 (circles), and 5.0 (diamonds).

FIG. 2: Estimated values of $1/CuT_1$ for $J_\perp = 1.0$ as a function of temperature, taking $\omega_{\text{max}} = 0.3$ (+) and 0.5 (×).
FIG. 3: Spin lattice relaxation rates as a function of temperature (in units of $J_\parallel$) for copper (circles), ladder oxygen (squares) and rung oxygen (diamonds) sites, for $J_\perp/J_\parallel = 0.5, 1.0,$ and $2.0$ in (a), (b), and (c) respectively. The upside-down triangle on each graph indicates the value of the spin gap $\Delta$ for the corresponding system.
FIG. 4: Spin lattice relaxation rates at $T = 50$ as a function of $J_\perp$. 