Magnetization profiles and NMR spectra of doped Haldane chains at finite temperatures

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I. INTRODUCTION

The physics of low-dimensional spin systems is extremely rich and shows many surprising effects. Using a valence bond picture it has been suggested that open segments of $S = 1$ antiferromagnetic spin chains should have $S = 1/2$ excitations localized at the ends of the chain [1,2], that are experimentally observable. In a recent experiment by Tedoldi et al [3] the presence of these $S = 1/2$ chain end spins was beautifully demonstrated by measuring the NMR profile of Mg doped Y$_2$BaNiO$_5$, a model $S = 1$ spin chain system, showing well defined satellite peaks indicative of the chain end excitations. These satellite peaks were, perhaps surprisingly, very well defined at temperatures close to and well above the gap, $\Delta \approx 100K$. At temperatures below the gap, where one would expect the signal from the chain end spins to be most clearly observable, the satellite peaks were smeared into a single broad line. The inter chain coupling $J^\perp$ in Y$_2$BaNiO$_5$ is very small ($|J^\perp/J| \leq 5 \times 10^{-4}$, $J \approx 285K$) [3] and it therefore remains a question why no well defined satellite peaks were observed below the gap. Bulk susceptibility and magnetization measurements [4] on Zn doped Y$_2$BaNiO$_5$ have also been interpreted in terms of $S = 1/2$ chain end excitations, but in this case at temperatures all the way down to 4-10K, well below the gap. Very interestingly, a sub-Curie power law divergence was observed at temperatures below 4K hinting at the presence of a gapless phase. In light of these two experiments it is then natural to ask, from a theoretical point of view, if chain end excitations are observable at finite fields and temperatures well above the gap and when a simple one-dimensional model will fail to describe the experiments. In the present paper we systematically address these issues by calculating the on-site magnetization, $S_i^z$, at finite temperatures and fields using continuous time quantum Monte Carlo techniques and reconstructing the associated NMR spectra for a given concentration of impurities.

It is by now well established that integer spin chains display a gap above a spin liquid ground-state [5] as opposed to half-integer spin chains which are gapless. The gap in integer spin chain systems has been observed experimentally in numerous compounds [6] as well as in many numerical studies [7]. For $S = 1$ chains the gap is known to be $\Delta \sim 0.4$105/J [8]. For integer spin chains, described by a simple antiferromagnetic Heisenberg model, it is possible to understand the low temperature physics using the intuitive valence bond picture. From this point of view the ground-state is close to the valence bond solid (VBS) state. Each integer spin $S$ is decomposed into $2S$ spin $S = 1/2$ in the symmetric state and each of the $S = 1/2$ on neighboring sites are paired into singlets. It is possible to determine Hamiltonians where this VBS state is the exact ground-state. For $S = 1$ spin chains the corresponding Hamiltonian is rather simple and apart from the usual Heisenberg term includes a biquadratic term $\beta(S_i \cdot S_{i+1})^2$. For $\beta = -1/3$, the AKLT point, the VBS state is the exact ground-state as was shown by Affleck-Kennedy-Lieb and Tasaki [9] (AKLT). Under periodic boundary conditions the VBS state is always a singlet. However, as was noted by Kennedy [1] for open $S = 1$ chains, the ground-state is four-fold degenerate in the thermodynamic limit. In the VBS picture this is easy to understand since the $S = 1/2$ spins at the end of the chain are not paired into a singlet state, leading to a four-fold degeneracy for a $S = 1$ system. In general a spin-$S$ chain should then have spin-$S/2$ chain-end excitations. Many real compounds are well described by the antiferromagnetic Heisenberg chain:

$$H = J \sum_i S_i \cdot S_{i+1}. \quad (1)$$

Following the above discussion the ground-state should
then be close to the VBS state. For a $S = 1$ system, the low-energy states of an open segment is then a singlet and a triplet separated by a gap decaying exponentially with $L$, the length of the segment. For $L$ odd the interaction between the two chain end excitations is ferromagnetic and the triplet is the lowest lying state, for $L$ even the interaction is antiferromagnetic and the singlet is lowest.

In the following we will focus on $S = 1$ spin chains relevant to the experiments on $Y_2$BaNiO$_5$. Numerically the $S = 1/2$ chain end excitations have been observed by various methods: exact diagonalisation [1], Quantum Monte Carlo (QMC) [10], Density Matrix Renormalization Group (DMRG) [11]. They subsist all through the Haldane phase $-1 < \beta < 1$ [11] but as $\beta$ is increased from 0 to 1 the peak in the on-site magnetization, $S_z^x$, moves away from the first site of the chain. At $\beta = 1$ a dimerized phase is reached where the $S = 1/2$ chain end excitations cease to exist. At the AKLT point, $\beta = -1/3$, it can be analytically shown that the decay of the on-site magnetization away from the end of the chain has a pure exponential form with a length scale exactly equal to the bulk correlation length. Experimentally the $S = 1/2$ chain end excitations have been observed in EPR measurements in $S = 1$ chains doped with non magnetic ions [2,12–15] and even in microscopically fractured undoped systems [16]. Initially, experiments measuring the Schottky anomaly in Zn doped $Y_2$BaNiO$_5$ [17] were interpreted in terms of $S = 1$ excitations but later work [18] showed that an explanation in terms of $S = 1/2$ chain end excitations is also possible when anisotropy is taken into account. These studies have mainly dealt with low temperature (i.e. $T \ll \Delta$) behavior of these systems where the only relevant states are the low-lying singlet and triplet. In the recent experiments by Tedoldi et al [3], a $^{89}$Y NMR study of doped system $Y_2$BaNi$_{1-x}$Mg$_x$O$_5$ was performed, clearly indicating the $S = 1/2$ chain end excitations for temperatures well above the gap where many excited states apart from the low-lying triplet and singlet should be populated. In order to understand the NMR spectra in this high temperature range we simulated the isotropic Heisenberg spin 1 chain using a continuous time Quantum Monte Carlo algorithm at finite temperature and finite field. The QMC method samples the partition function so that all (excited) states are correctly treated. For the purpose of understanding the physics at temperatures close to $T \approx 0$ we have performed Density Matrix Renormalisation Group calculations of the NMR spectra at $T = 0$. Our results for temperatures above the gap are in very good agreement with the experimental data and clearly show that the $S = 1/2$ chain end excitations indeed are observable at such high temperatures. At temperatures below the gap down to $T = 0$ we find very well defined satellite peaks that at $T = 0$ only are broadened by the distribution of chain lengths. In the experiments of Tedoldi [3] et al, no clear satellite peaks were observed at $T = 77K$, well below the gap. This could be due to a limited resolution at this temperature [19]. In the experiments of Payen et al [4] on doped $Y_2$BaNiO$_5$ the results were interpreted in terms of the chain end excitations at temperatures down to 4K and it therefore remains unclear when and if higher dimensional physics become important in doped $Y_2$BaNiO$_5$.

The paper is organized as follows. In section II we discuss the numerical methods used. Section III presents our results for the on site magnetization for different temperatures and fields. In section IV we turn to a discussion of the NMR spectra obtained from the magnetization profiles and finally in section V we discuss possible explanations why signals from the $S = 1/2$ excitations are not clearly seen below the gap.

II. NUMERICAL METHOD

We use a single-cluster continuous time [20] version of the loop algorithm [21]. Spins 1 are simulated by two spins 1/2 with symmetrized boundary conditions [22,23] in the imaginary time direction. In order to take into account the magnetic field, we use a hybrid method [24] where field-dependent loop flip probability and global flip of worldlines are used. Each Monte Carlo run consists of $10^7$ sweeps (a measurement is taken every 10 sweeps to avoid autocorrelations) preceded by $10^5$ sweeps of thermalization. The DMRG [25] calculations were performed keeping $m = 243$ states for chain lengths between $L=1,100$ and $m = 81$ states for chain lengths $L=101,501$. Only negligible differences were observed between the results for $m = 81$ and $m = 243$. The calculations were performed using the total $z$-component of the spin, $S_{tot}^z$, and parity, $P$, with respect to a reflection about the middle of the chain as quantum numbers. All the magnetization profiles were calculated in the $S_{tot}^z = 1, P = -1$ state.

III. MAGNETIZATION PROFILES

A. Influence of temperature

The first question we address is the temperature dependence of the on-site magnetization $S_z$. Due to symmetry the on-site magnetization is always zero in the absence of a magnetic field. In order to compare to experiments we therefore perform calculations in the sector with $S_{tot}^z = 1$. In Fig. 1 we show the magnetization profile for an open chain of 29 spins for various temperatures in the sector $S_{tot}^z = 1$. Filled circles are $T=0$ results obtained by DMRG, the other symbols are QMC data. We observe the well-known alternating structure due to the antiferromagnetic interactions which tends to be diminished as the temperature is raised. For the lowest temperature

...
$T = 0.1J$ (well below the gap), the profile is almost identical to the ground-state result. The decay of the on-site magnetization away from the end of the chain is at $T = 0$ known to be exponential [26,11] with a length scale equal to the bulk correlation length $\xi \simeq 6$. At high temperature the staggered magnetization decays rapidly from its value at the end of the chain corresponding to a shorter correlation length. At the highest temperatures, the middle spins (around $i = 15$) have become polarized and show an average magnetization of $\langle S_i^z \rangle = \frac{1}{L} \sim 0.034$. At temperatures relevant to the experiments of Tedoldi et al [3], $T = 0.3J - 0.8J$, the structure from the $S = 1/2$ chain end excitations is still clearly visible. All these results confirm the existence of $S = 1/2$ excitations at temperature above the gap, even if the magnetization at the end of the chain is significantly reduced above the gap.

**B. Influence of magnetic field**

Lifting the constraint of working in the $S_{tot}^z = 1$ sector we now consider the on-site magnetization as a function of the applied magnetic field at temperatures and fields relevant to the experiments. We calculated the dependence on the magnetic field of the profiles for two temperatures: $T = 0.1J \ll \Delta$ and $T = 0.526J > \Delta$.

For $T = 0.1J$ (figure 2), the zero-field magnetization is zero for all spins (by symmetry). At this temperature, the only magnetic states are in the sectors $S_{tot}^z = \pm 1$ of the triplet (which is the lower state for an odd chain) and are the only ones affected by the field. As we increase the field, the $S_{tot}^z = 1$ state will be energetically favored and hence the magnetization starts to take on the alternating structure showing the presence of the chain end excitations. At sufficiently large fields we should obtain results comparable to the results discussed in the previous section obtained in the absence of a magnetic field but in the $S_{tot}^z = 1$ sector. This is clearly the case and for $H = 0.3J$, we nearly recover the zero-field structure of the $S_{tot}^z = 1$ sector at $T = 0.1J$ shown in Fig. 1. If the field is too strong (say $H > 0.3J$), magnetic states from different sectors above the Haldane gap contribute to the thermal expectation of the on-site magnetization and, even if the alternating structure is still present, we don’t recover the results from the zero-field case. This effect is visible on the $H = 0.4J$ data (left triangles).

For this field, $\langle S_i^z \rangle$ for odd spins (which all have a positive expectation value) is nearly equal to the value in the $S_{tot}^z = 1$ sector (filled circles) but for even spins the value for $H = 0.4J$ is greater than in the $S_{tot}^z = 1$ sector (this is particularly clear for the middle even spins).

At higher temperature ($T = 0.526J$), slightly above the gap $\Delta \simeq 0.35 - 0.4J$ (see figure 3), end effects are smaller, especially for low fields. At $H = 0$, the on-site magnetization is zero by symmetry and is not shown in the figure. Increasing the field, we notice two effects: the first is simply progressive magnetization of the middle spins, unaffected by end effects. When we later discuss the NMR spectra this will result in a positive shift of the NMR spectra for Y atoms close to the middle of the chain segments, i.e. of the central peak. The second effect is the progressive magnetization of the end spins with increasing field. The largest value of the field is equal to the field used in the experiments by Tedoldi et al [3]. Clearly at this field and temperature there is
Here, \( L \) from the chain end. When the temperature is large, the correlation length is small and the correlation length. In order to compare to the experimental results \[23\] \[27\] and the experimental results taken from NMR experiments \[3\].

Due to the rather large variation in chain lengths and the resulting variation in the on-site magnetization the satellite peaks should be broadened. Averaging over a large number of chain lengths it is then possible to re-construct the observed NMR spectra up to an overall rescaling factor.

In order to perform this calculation we have simulated open spin 1 segments in a magnetic field of \( H = 0.08J \), corresponding to the experimental situation, for four temperatures between \( \sim 0.2J \) and \( \sim 1J \) equal to the experimental values, for chain lengths between \( L=1,200 \). At zero temperature we have computed the profiles in a slightly different fashion by selecting the \( S^2_{\text{tot}} = 1 \) subspace for chain lengths between \( L=1,501 \) using DMRG techniques. For very short even length chains, which are quite probable, this approach might lead to a slight bias since the magnetic field applied in the experimental situation might not be sufficiently strong to select the \( S^2_{\text{tot}} = 1 \) state as the ground-state. However, we estimate this effect to be rather small.

Assuming that the distribution of the Mg impurities is uncorrelated we use a geometric distribution for chain lengths where the probability of having a chain of length \( L \) is given by:

\[
p(L) = x(1 - x)^L.
\]

Here \( x \) is the impurity concentration. The mean chain length is then

\[
\bar{L} = \sum_{l=0}^{\infty}lx(1 - x)^{l} = \frac{1}{x} - 1,
\]

which, if \( x \) is sufficiently small, reduces to \( \bar{L} = \frac{1}{x} \). The non magnetic impurity concentration we used is \( x = 5\% \), which corresponds to a mean chain length of 19. One should note that the geometric distribution is quite wide and even though large chain lengths are less probable than short ones they contribute significantly to the observed NMR spectrum. The most probable chain length is in fact \( L = 0 \) corresponding to an impurity site and the possibility of having one or more impurities next to each other is non-negligible. Hence, in order to obtain reliable results we have computed profiles for chains of long lengths. For \( T \neq 0 \), we calculated profiles for chains of lengths 1 up to 200, and for \( T = 0 \) we simulated chains of up to 500 sites \[28\].

C. Estimation of correlation length

As previously mentioned, the on-site magnetization should decay exponentially away from the end of the chain with a characteristic length related to the bulk correlation length. In order to compare to the experiments, we estimated the correlation length for different temperatures for the longest chains studied in a field of \( H = 0.08J \). We fit the exponential decay of the on-site magnetization away from the end of the chain using the following form:

\[
\langle S^z_i \rangle = \langle S^z_1 \rangle \left( (-1)^{-l}e^{-\frac{L}{\xi(T)}} + (-1)^{L-l}e^{-\frac{L-l}{\xi(T)}} \right) + \langle S^z_h \rangle. \tag{2}
\]

Here, \( L \) is the length of the chain and the last term \( \langle S^z_h \rangle \) in this equation corresponds to the magnetization induced by the field at high temperature. Our results for the correlation length obtained using this method are shown in table I and are in agreement with previous numerical results \[23\] \[27\] and the experimental results taken from NMR experiments \[3\].

| \( T(J) \) | 0.993 | 0.702 | 0.526 | 0.27 |
|-----------|-------|-------|-------|------|
| \( \xi(T) \) | \( 1.57 \pm 0.23 \) | \( 2.37 \pm 0.37 \) | \( 2.97 \pm 0.13 \) | \( 4.62 \pm 0.11 \) |

**TABLE I.** The temperature dependence of the correlation length as determined from the on-site magnetization profiles in a field of \( H = 0.08J \).
According to the experimental situation, we randomly choose two lengths of chains and compute the sum of the two chains magnetizations site by site as we go along the chains. The two sites whose magnetizations are added at each step stand for the two next nearest neighbors spins of the NMR probe in the experiments [3]. When the end of one chain is reached we insert a site with zero on-site magnetization corresponding to the impurity. We then select a new chain following the geometrical distribution and continue to go along the system until the end of another chain is reached and we repeat the process till the thermodynamic limit is reached. If the length of the chain is 0 we immediately insert an impurity site resulting in two or more impurities sitting next to each other. The NMR spectrum is then computed by plotting the histogram of the effective field seen on the Y site, i.e. the sum of the on-site magnetization coming from the two sites sitting closest to the Y on the two chains. This procedure should be quite close the experimental situation. In order to compare to the experimental data we rescale the y and x axis of the experimental NMR spectrum by a scale factor. This is done by adjusting to the first satellite peak (for the x axis we use a scale factor of roughly 0.8 – 0.9 which is in good agreement with the experimental estimate of hyperfine interaction).

In figures 4 and 5, we show the results of our calculations for the five temperatures and compare them to the experimental spectra. The central peak corresponds to the situation where the effective field at the Y site cancels either due to the fact that the two contributions are of roughly equal and opposite sign or because the Y is sitting close to the middle of both of the two chain segments where the on-site magnetization is very close to zero. If the Y is closest to the first site of a chain segment it should experience a positive shift away from the central peak and assuming that the magnetization on the other chain is close to zero we should observe the most pronounced satellite peak corresponding to the average value of the on-site magnetization on the first site. If we have exponentially localized excitations at the end of the chains the most probably value of the magnetization on the “other” chain is close to zero. However, this is of course not strictly the case, resulting in a broadening of the peaks. If the Y is close to the second site of one of the chain we should observe a satellite peak shifted in the negative direction away from the central peak but with a smaller absolute shift. It is then possible to identify the various satellite peaks with the on-site magnetization as one goes away from the end of the chain. As the distance from the end of the chain is increased the position of the satellite peak should move closer and closer to the central peak. Due to the significant broadening of the peaks only the first few satellite peaks are resolvable. With exponentially localized excitations at the end of the chains there is also a small probability that the Y is close to firsts sites on both chains (for example first-first, first-third ..., or second-second, second-fourth...). Theoretically, this should result in a number of secondary peaks beginning at twice the absolute value of the main peaks, also contributing to a general broadening of the spectrum. However, the intensity of such peaks should be rather small. In our calculations at $T = 0$, a secondary peak corresponding to the Y close to the firsts sites on both chains is clearly visible at twice the value of the first satellite peak (See Fig. 5).

The computed spectra are in extremely good agreement with the experimental results for the three higher temperatures. Note that the central peak is always greater in the NMR experiments than in our simulations, this might be due to a different distribution of impurities in the sample than the simple geometrical distribution (we don’t consider correlations between impurities in this model). There is a central peak for all the temperatures; the position of this peak is increased as temperature is raised due to the fact that (middle) spins are more easily
magnetized by the magnetic field for high temperatures, see section III B. The spectrum is wider at lower temperatures since the magnetization profiles are much more developed, as discussed in the previous section, resulting in larger shifts. For each figure, apart from the central one, several peaks can be observed. As explained, the shift from the central peak to the others correspond to the intensity of the magnetization on neighboring sites. On the figures, we have labelled the different satellite peaks according to the proximity to a site on the chain. As can be clearly seen, the neighboring magnetic structure of a site is more and more well defined as the temperature is lowered. For example, at the highest temperature $T = 0.993J$, only the two first satellite peaks are resolved, the more distant satellites cannot be distinguished and contribute to the central peak. For $T = 0.526J$, up to six satellite peaks are resolved, and even more at the lowest temperature, $T = 0.27J$. This temperature is well below the gap and corresponds to the $T = 77K$ experimental results in Ref. [3]. In this case the experiment show a single broad line and the signal from satellite peaks is lost. Tedoldi et al [3] argues that one possible explanation of this is the large distribution of the chain lengths which could become more important at temperatures below the gap. This effect is taken correctly into account in our calculations and since we observe well-defined satellite peaks well below the gap and at $T = 0$ the explanation is likely elsewhere. The resolution of the $T = 77K$ spectrum is less than for the other temperatures [19] and it is possible that higher resolution experiments will be capable of resolving the satellite peaks.

Finally, in figure 5 we show results calculated at $T = 0$ using DMRG techniques. The calculation is performed in zero field in the $S_{tot}^z = 1$ subspace. Many well defined peaks are now visible. In particular we see secondary peaks corresponding to the case where the $Y$ is between sites which both have large on-site magnetizations ($1'$).

V. DISCUSSION

Our results clearly indicate that the $S = 1/2$ chain excitations should be visible at temperatures well above the gap in a finite field. It is well known that the gap in $S = 1$ chains corresponds to a single magnon branch at $k = \pi$. Around $k = 0$ a two-magnon continuum should begin at energies of $2\Delta$ and a three-magnon continuum at $k = \pi$ should begin at an energy of $3\Delta$ [29]. It is natural to expect that chain end effects are visible until the two-magnon and three-magnon continuum become significantly populated, i.e. for temperatures above 300K in agreement with the numerical and experimental results.

The discrepancy between our theoretical results and the experimental results for temperatures below the gap could have several explanations. As suggested by Tedoldi et al [3] anisotropy effects could be important. However, the single ion anisotropy term $D = 8K$ is rather small and it is not obvious that it would give rise to a large effect. Such a term would split the low-lying triplet into a doublet and a singlet and could likely cause a systematic shift in the spectra rather than a complete smearing of the profile. Still another explanation could come from the small coupling between the chains. However, it would seem unlikely that this effect could be important at temperatures as high as $T = 77K$. Payen et al [4] argue that such effects become important at temperatures around 4K, much lower than the gap, $\Delta \approx 100K$. However, these experiments are concerned with bulk susceptibility and magnetization which might be a less sensitive probe. Another simple explanation is that below the gap so many secondary peaks appear that the experimental spectrum appears as a single broad line. This is not unreasonable considering the results shown for $T = 77K$ in figure 4. Lastly, the actual impurity concentration is not known to a very high precision and a slight modification in this concentration greatly modifies the observed spectra. High resolution NMR experiments at low temperatures will be needed to resolve these issues.

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