Thermodynamics of the frustrated ferromagnetic spin-1/2 Heisenberg chain

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Abstract. We studied the thermodynamics of the one-dimensional $J_1$-$J_2$ spin-1/2 Heisenberg chain for ferromagnetic nearest-neighbor bonds $J_1 < 0$ and frustrating antiferromagnetic next-nearest-neighbor bonds $J_2 > 0$ using full diagonalization of finite rings and a second-order Green-function formalism. Thereby we focus on $J_2 < |J_1|/4$ where the ground state is still ferromagnetic, but the frustration influences the thermodynamic properties. We found that their critical indices are not changed by $J_2$. The analysis of the low-temperature behavior of the susceptibility $\chi$ leads to the conclusion that this behavior changes from $\chi \propto T^{-2}$ at $J_2 < |J_1|/4$ to $\chi \propto T^{-3/2}$ at the quantum-critical point $J_2 = |J_1|/4$. Another effect of the frustration is the appearance of an extra low-$T$ maximum in the specific heat $C_v(T)$ for $J_2 \gtrsim |J_1|/8$, indicating its strong influence on the low-energy spectrum.

Introduction: In low-dimensional frustrated quantum magnets thermal and quantum fluctuations strongly influence the low-temperature physics [1,2]. Special attention has been paid to one-dimensional (1D) $J_1$-$J_2$ quantum Heisenberg magnets, see Ref. [3] and references therein. Recent experimental studies have shown that edge-shared chain cuprates, such as LiVCuO$_4$, Li(Na)Cu$_2$O$_2$, Li$_2$ZrCuO$_4$, and Li$_2$CuO$_2$ [4–13], represent a family of quantum magnets for which the 1D $J_1$-$J_2$ Heisenberg model is a good starting point for a theoretical description. The above listed compounds are quasi-1D frustrated spin-1/2 magnets with a ferromagnetic (FM) nearest-neighbor (NN) in-chain coupling $J_1 < 0$ and an antiferromagnetic (AFM) next-nearest-neighbor (NNN) in-chain coupling $J_2 > 0$.

The model: The Hamiltonian of the 1D $J_1$-$J_2$ Heisenberg ferromagnet is given by

$$H = J_1 \sum_{\langle i,j \rangle} S_i S_j + J_2 \sum_{[i,j]} S_i S_j,$$

where the first sum runs over the NN bonds and the second sum over the NNN bonds. Henceforth we set $J_1 = -1$. For the model [11] a quantum critical point at $J_2 = 0.25$ exists where the FM ground state (GS) gives way for a singlet GS with spiral correlations for $J_2 > 0.25$ [14–16]. For most of the edge-shared chain cuprates $J_2$ is large enough to realize such a spiral GS. However, several materials considered as model systems for 1D spin-1/2 ferromagnets, such as TMCuC[(CH$_3$)$_4$NCuCl$_3$] [17] and p-NPNN (C$_{13}$H$_{16}$N$_3$O$_4$) [18], might have also a weak frustrating NNN interaction $J_2 < 0.25$. Moreover, recent studies [13] lead to the conclusion that Li$_2$CuO$_2$ is a quasi-1D spin-1/2 system with a dominant FM $J_1$ and weak frustrating AFM...
Here we focus on the parameter region $J_2 \leq 0.25$, i.e. the GS is ferromagnetic. Only at $J_2 = 0.25$ the FM GS multiplet is degenerate with a spiral singlet GS [14–16]. On the other hand, the frustrating $J_2$ influences the low-energy excitations, in particular, if $J_2$ is close to the quantum critical point. Hence, the frustration may have a strong effect on the low-$T$ thermodynamics. We mention that previous studies [19, 20] of the thermodynamics of the 1D $J_1$–$J_2$ model with FM $J_1$ did not consider values of $J_2$ near the quantum critical point $J_2 \lesssim 0.25$.

Results: To study the thermodynamic properties we use the full exact diagonalization (ED) of finite rings of up to $N = 22$ lattice sites, complemented by data obtained by a spin-rotation-invariant second-order Green-function method (RGM) [21–24]. Note that by contrast to ED the RGM is limited to values $J_2 \leq 0.2$ [24] but yields results for $N \to \infty$, that allows the calculation of the correlation length by the RGM. Here we will present data for the spin-spin correlation functions $\langle S_0 S_n \rangle$, the uniform static spin susceptibility $\chi$, and the specific heat $C_v$. For the discussion of the correlation length of the model (1), see Ref. [24]. For the unfrustrated model we will compare our results with available Bethe-ansatz data [25] and transfer-matrix renormalization group (TMRG) results [19].

The temperature dependence of the spin correlation functions $\langle S_0 S_n \rangle$ is shown for $n = 1$ (NN) and $n = 10$ for various $J_2$ in Fig. 1. With increasing frustration the correlation functions decrease, where the further-distant correlators decay much stronger than the NN correlator. Near and at the quantum critical point the large-distance correlator $\langle S_0 S_{10} \rangle$ vanishes already at $T \gtrsim 0.05$. Interestingly, for $J_2 = 0.2$, 0.24, and 0.25 the correlator $\langle S_0 S_{10} \rangle$ changes the sign and goes through a minimum. This behavior is not affected by finite-size effects, e.g., the correlators $\langle S_0 S_8 \rangle$ for $N = 16, 20$ and $\langle S_0 S_6 \rangle$ for $N = 12, 16, 20$ also change the sign and go through a minimum for $J_2 = 0.2$, 0.24, and 0.25.

Next we discuss the low-temperature properties of the susceptibility $\chi = \lim_{T \to 0} d\langle S_z \rangle/dh$. Due to the FM GS $\chi$ diverges at $T \to 0$. Using Bethe-ansatz for $J_2 = 0$ the critical behavior has been determined as $\chi \propto T^{-2}$ [25]. Using the RGM, recently it has been confirmed that the critical indices for the susceptibility and the correlation length, $\gamma = 2$ and $\nu = 1$, respectively, are not changed by frustration for $J_2 < 0.25$. However, at the quantum critical point $J_2 = 0.25$ a change of the low-temperature physics is expected [1]. To study that question we consider the

\begin{figure}
\centering
\includegraphics[width=0.45\textwidth]{fig1}
\caption{Spin correlation function $\langle S_0 S_1 \rangle$ (NN) and $\langle S_0 S_{10} \rangle$ calculated by ED for $N = 20$ sites.}
\end{figure}

\begin{figure}
\centering
\includegraphics[width=0.45\textwidth]{fig2}
\caption{$\chi T^2$ versus $\sqrt{T}$ calculated by ED for $N = 22$ (thick lines – calculated data, thin lines – extrapolation to $T \to 0$, see Eq. (2) and text) and RGM (circles) as well as Bethe-ansatz data (squares) for $J_2 = 0$ from Ref. [25]. The inset shows the coefficient $y_0 = \lim_{T \to 0} \chi T^2$ versus $J_2$.}
\end{figure}
In Fig. 2 we plot $\chi T^2 = y_0 + y_1 \sqrt{T} + y_2 T + O(T^{3/2})$ \label{eq:chiT2}
related to the existence of the FM critical point at $T = 0$. It has been derived for $J_2 = 0$ in Ref. [25]. For the frustrated system 1 the coefficients $y_0$, $y_1$, and $y_2$ depend on $J_2$. In Fig. 2 we plot $\chi T^2$ versus $\sqrt{T}$. We find a good agreement of the ED data for $\chi T^2$ with Bethe-ansatz results down to quite low temperature $T$. The RGM results for $\chi T^2$ deviate slightly from the Bethe-ansatz results for finite $T$, but approach the Bethe-ansatz data for $T \to 0$, see also Ref. [22]. The behavior of the leading coefficient $y_0$ and the next-order coefficient $y_1$ can be extracted from the results for $\chi T^2$ by fitting them to Eq. \ref{eq:chiT2}. For the RGM we use data points up to a cut-off temperature $T = T_{cut} = 0.005$. To deal with finite-size effects in the ED data at very low $T$, we use the specific heat per site $C_v(T)$, see below, to determine that temperature $T_{ED}$ down to which the first four digits of $C_v(T)$ for $N = 20$ and $N = 22$ coincide. Then we fit the ED data in the interval $T_{ED} \leq T \leq T_{ED} + T_{cut}$ to Eq. \ref{eq:chiT2}. Note that $T_{ED}$ becomes smaller for increasing $J_2$, we find e.g., $T_{ED} = 0.22, 0.13, 0.09, 0.04, 0.03, 0.02$ at $J_2 = 0, 0.1, 0.15, 0.2, 0.24, 0.25$, respectively. For $J_2 = 0$ we found $y_0 = 1/24$ ($y_0 = 0.0418$) for the RGM (ED), which agrees with the Bethe-ansatz results of Ref. [25]. [Note the different definitions of $\chi$ in our paper and in Ref. [25].] Including frustration $J_2 > 0$ we observe a linear decrease of $y_0$ with $J_2$ down to zero at $J_2 = 0.25$ given by
\begin{equation}
y_0 = (1 - 4J_2)/24, \label{eq:y0}
\end{equation}

\begin{figure}[h]
\centering
\includegraphics[width=0.45\textwidth]{fig3}
\caption{ED data for the specific heat for $N = 22$. For comparison we show TMRG data (squares) from Ref. [19] for $J_2 = 0$.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.45\textwidth]{fig4}
\caption{Finite-size dependence of the specific heat for $J_2 = 0.22$. The inset shows $C_v^p$ and $T^p$ versus $J_2$, see text.}
\end{figure}

cf. the inset of Fig. 2. The vanishing of $y_0$ at $J_2 = 0.25$ indicates the change of the low-$T$ behavior of the physical quantities at the quantum critical point [1]. Indeed, a polynomial fit according to $y_1 = a_y + b_y J_2 + c_y J_2^2$ yields the finite value $y_1 \approx 0.05 (0.04)$ for RGM (ED). Hence, our data provide evidence for a change of the low-$T$ behavior of $\chi$ from $\chi \propto T^{-2}$ at $J_2 < 0.25$ to $\chi \propto T^{-3/2}$ at the quantum critical point $J_2 = 0.25$. For a a similar discussion of the correlation length $\xi$, see Ref. [24], where it was found that the low-$T$ behavior of $\xi$ changes from $\xi \propto T^{-1}$ at $J_2 < 0.25$ to $\xi \propto T^{-1/2}$ at $J_2 = 0.25$.

In Fig. 3 we present ED results for the specific heat $C_v$. For $J_2 = 0$ we found a broad maximum at $T \approx 0.332$ and a steep decay to zero starting at about $T = 0.05$ in accord with the TMRG [19]. For $J_2 \gtrsim 0.125$ the specific heat exhibits a minimum located at around $T = 0.2$, and two maxima, namely a high-$T$ maximum at around $T = 0.6$ and an additional low-$T$ maximum at $T < 0.1$. If $J_2$ approaches $J_2 = 0.25$, a further quite sharp peak at very low $T$ appears,
that is, however, strongly size dependent, see Fig. 4. From Fig. 4 it is obvious that the extra low-\(T\) finite-size peak behaves monotonously with \(N\). Hence, we have performed a finite-size extrapolation to \(N \to \infty\) of the height \(C^p\) and the position \(T^p\) of the peak in \(C_v(T)\) using the formula \(a(N) = \alpha_\infty + a_1/N^2 + a_2/N^4\). The extrapolated values \(C^p_\infty\) and \(T^p_\infty\) are shown in the insets of Fig. 4. Obviously, \(C^p_\infty > 0\) even near the quantum critical point \(J_1 = 0.25\), where \(C^p_\infty \approx 0.05\). On the other hand, \(T^p_\infty\) decreases with \(J_2\) and becomes very small near \(J_2 = 0.25\). This behavior suggests that a characteristic steep decay of \(C_v(T)\) down to zero starts at very low \(T\) when approaching \(J_2 = 0.25\).

**Summary:** We discussed the thermodynamics of frustrated FM spin-1/2 \(J_1-J_2\) Heisenberg chains and found as prominent features (i) a change of the low-\(T\) critical behavior at the quantum critical point \(J_2 = \left|J_1\right|/4\), (ii) and an additional low-\(T\) maximum in the specific heat for \(\left|J_1\right|/4 > J_2 \gtrsim \left|J_1\right|/8\).

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