Thermodynamics of a two-dimensional frustrated spin-$\frac{1}{2}$ Heisenberg ferromagnet

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Using the spin-rotation-invariant Green’s function method we calculate the thermodynamic quantities (correlation functions $\langle S_i S_j \rangle$, uniform static spin susceptibility $\chi$, correlation length $\xi$, and specific heat $C_V$) of the two-dimensional spin-1/2 $J_1$-$J_2$ Heisenberg ferromagnet for $J_2 < J_2^c \approx 0.44|J_1|$, where $J_2^c$ is the critical frustrating antiferromagnetic next-nearest neighbor coupling at which the ferromagnetic ground state gives way for a ground-state phase with zero magnetization. Examining the low-temperature behavior of $\chi$ and $\xi$, in the limit $T \to 0$ both quantities diverge exponentially, i.e., $\chi \propto \exp(b/T)$ and $\xi \propto \exp(b/2T)$, respectively. We find a linear decrease in the coefficient $b$ with increasing frustration according to $b = -\frac{1}{2} (J_1 + 2J_2)$, i.e., the exponential divergence of $\chi$ and $\xi$ is present up to $J_2^c$. Furthermore, we find an additional low-temperature maximum in the specific heat when approaching the critical point, $J_2 \to J_2^c$.

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I. INTRODUCTION

Low-dimensional quantum magnets have attracted much attention during the last decades. They are predestined to study the influence of strong thermal and quantum fluctuations. Much attention has been paid to the theoretical investigation of the two-dimensional (2D) spin-1/2 $J_1$-$J_2$ quantum Heisenberg antiferromagnet which may serve as a canonical model to study the interplay of frustration effects and quantum fluctuations (see, e.g., Ref. [3]). An additional motivation to study this model comes from the experimental side. Very recently, several quasi-2D magnetic materials with a ferromagnetic nearest-neighbor (NN) coupling $J_1 < 0$ and a frustrating antiferromagnetic next-nearest neighbor (NNN) coupling $J_2 > 0$ have been investigated experimentally, e.g., Pb$_2$VO(PO$_4$)$_2$ [28], (CuCl)LaNb$_2$O$_7$ [29], SrZnVO(PO$_4$)$_2$ [28-30], and BaCdVO(PO$_4$)$_2$ [28,31]. The quite large frustrating $J_2$ drives these materials out of the ferromagnetic phase. The experimental findings have stimulated several theoretical studies of the ground-state and thermodynamic properties of the $J_1$-$J_2$ model with $J_1 < 0$ and frustrating $J_2 > 0$. It was found that the ferromagnetic ground state for the spin-1/2 model breaks down at $J_2 = J_2^c \approx 0.4|J_1|$ [28,31,32]. Note that for the classical model (spin $s \to \infty$) the corresponding transition point is at $J_2 = 0.5|J_1|$. On the other hand, some materials considered as 2D spin-1/2 ferromagnets, such as K$_2$CuF$_4$, Cs$_2$CuF$_4$, Cs$_2$AgF$_4$, La$_2$BaCuO$_5$, and Rb$_2$CrCl$_4$ might have also a weak frustrating NNN interaction $J_2 < J_2^c$. In contrast to the previous investigations of the 2D $J_1$-$J_2$ model with ferromagnetic $J_1 > 0$, which have considered predominantly the case of strong frustration, in the present paper we will focus on the region of weak frustration $J_2 < J_2^c$.

Although for $J_2 < J_2^c$ the ground state remains ferromagnetic, the frustrating $J_2$ may influence the thermodynamics substantially, in particular near a zero-temperature transition. This has been demonstrated for the one-dimensional (1D) frustrated ferromagnet in Ref. [28], where a change in the low-temperature behavior of the susceptibility and the correlation length as well as an additional low-temperature maximum in the specific heat have been found when approaching the zero-temperature critical point from the ferromagnetic side.

The Hamiltonian of the system considered in this paper is given by

$$H = J_1 \sum_{\langle i,j \rangle} S_i S_j + J_2 \sum_{\langle i,j \rangle} S_i S_j : J_1 < 0 ; J_2 \geq 0 ,$$

where $\langle S_i \rangle^2 = 3/4$, and $\langle i,j \rangle$ denotes NN and $\langle i,j \rangle$ NNN bonds. The unfrustrated 2D ferromagnet ($J_2 = 0$) has been widely investigated, e.g., by the modified spin-wave theory renormalization group approaches [28] and by a spin-rotation-invariant second-order Green’s function method (RGM) [28]. However, in presence of frustration ($J_2 > 0$) the choice of appropriate methods for studying temperature dependent quantities of this system is restricted due to frustration and dimensionality. For instance, the powerful quantum Monte Carlo method is not applicable due to the minus sign problem, whereas finite-temperature density matrix renormalization group studies are restricted to 1D systems.

So far, only a few theoretical papers deal with the thermodynamics of the model for $J_2 > 0$, see, e.g., Refs. [13,17] and [17] where finite lattices of $N = 16$ and $N = 20$
sites are considered. As we will see below, for these lattice sizes the finite-size effects at low temperatures are large. Therefore, methods studying infinite systems are highly desirable. Among others, the Green’s function method is a powerful tool to study magnetic systems at finite temperatures, see e.g. Refs. 10–12 and references therein. A specific variant appropriate for frustrated systems is the RGM. In the present paper, we use the RGM to investigate thermodynamic properties of the frustrated model (1), focusing on $J_2 \leq J_1 \approx 0.4|J_1|$. The RGM has been applied successfully to low-dimensional (frustrated) infinite quantum spin systems. 28–37,43–45. In particular, for the 1D spin-1/2 Heisenberg ferromagnet it was shown that the RGM reproduces Bethe-ansatz results 28,37. Since the RGM can be formulated also for finite systems, we use full exact diagonalization (ED) of finite square lattices of $N = 16$ and $N = 20$ sites to compare the RGM with ED data for finite lattices.

The paper is organized as follows: In Sec. II the RGM applied to model (1) is presented. The results for the thermodynamic quantities are discussed in Sec. III. Following the arguments of Ref. 28 we put $\gamma_{n,m} = 0$, which we adopt the decoupling

\[
S_i^+ S_j^- = \alpha_{i,k} (S_i^+ S_k^-) S_j^+ + \alpha_{j,k} (S_j^+ S_k^-) S_i^+. 
\]

Following the arguments of Ref. 28 we put $\alpha_{i,k} = \alpha$ in the whole temperature region. We obtain $-S^+_q = \omega^2 S^+_q$ and

\[
\chi^+ (\omega) = -\langle S^+_q S^+_q \rangle =\frac{M_q}{\omega^2 - \omega^2}. 
\]
finite $N$. In this case the quantity $C$ is not related to the magnetization and stays nonzero in the whole temperature region. In Ref. [43] it was shown that $C = 2T_X/N$.

To evaluate the thermodynamic quantities, a coupled system of six (for finite systems: seven) nonlinear algebraic self-consistency equations, including the sum rule $C_0 = \frac{1}{2}$, has to be solved numerically to determine the correlators $C_{1,0}$, $C_{1,1}$, $C_{2,0}$, $C_{2,1}$, $C_{2,2}$, and $C$ and the vertex parameter $\alpha$. To solve this system we use Broyden’s method[49] which yields the solutions with a relative error of about $10^{-8}$ on the average. The momentum integrals are done by Gaussian integration. To find the numerical solution of the RGM equations for $T > 0$, we start at high temperatures and decrease $T$ in small steps. Below a certain (low) temperature $T_0(J_2)$ no solutions of the RGM equations (except at $T = 0$) could be found, since the quantity $\Delta(T, J_2)$ in Eqs. (9) and (10) becomes exponentially small which leads to numerical instabilities.

Presenting our results in the next section we put $J_1 = -1$.

III. RESULTS

A. Phase transition at $T = 0$

In a first step we use the RGM as described above to determine the transition point $J_2^c$, where the ferromagnetic ground state gives way for a ground-state phase with zero magnetization. For that we solve the RGM equations at $T = 0$ (i) starting from $J_2 = 0$ with increasing $J_2$ and (ii) starting from large $J_2 \gg 1$ with decreasing $J_2$. In case (i) the RGM equations at $T = 0$ can be solved until the classical transition at $J_2 = 0.5$. In case (ii) we find solutions of the RGM ground-state equations down to $J_2 = 0.46$. This value lies certainly above the transition point $J_2^c$. To fix $J_2^c$ we may extrapolate the ground-state energy obtained for case (ii). The crossing point of both energies at $J_2 = 0.44$ can be considered as the RGM estimate of $J_2^c$, see the inset of Fig. 1. This value is in reasonable agreement with values for $J_2^c$ reported in other papers.[14,18,21,22]

The behavior of the spin-spin correlation functions at $T = 0$ near the transition point $J_2^c$ is illustrated in Fig. 1. For $J_2 \gtrsim J_2^c$ there is a noticeable variation in the correlation functions. With increasing $J_2$, at $J_2 \gtrsim 1$, the spin-spin correlation functions approach the corresponding values of the limiting case $J_2 \gg 1$. This behavior is in qualitative agreement with data from Lanczos ED and coupled-cluster method.[24] At the critical point the correlation functions jump to the exact results of the ferromagnetic phase, i.e., $\langle S_0 S_R \rangle = 0.25$. Together with the kink in the energy, this indicates a first-order transition. These results confirm the findings in previous papers.[14,19,22]

B. Thermodynamic properties

Now we investigate the thermodynamic properties of the model for $J_2 < J_2^c \approx 0.44$, where the ground state is ferromagnetic. First we consider the temperature dependence of the spin-spin correlation functions. The NN and NNN correlators are shown in Fig. 2 for various values of $J_2$. For comparison we show also ED data for $N = 20$ sites. The RGM results agree qualitatively with the ED data. With increasing frustration, the correlation functions decrease more rapidly with temperature. Interestingly, for large frustration the NNN correlation function changes its sign at a certain temperature, e.g.,
for $J_2 = 0.3$, at $T \approx 1.25$ (RGM data). The faster decay of the spin-spin correlators due to frustration is related to the decrease in the correlation length $\xi$ with increasing $J_2$, see the discussion below.

The uniform static spin susceptibility $\chi$ and the correlation length $\xi$ depicted in Figs. 3 and 4 show a similar behavior. Due to the ferromagnetic ground state, both quantities diverge at $T = 0$ exponentially, see below. With increasing frustration $J_2$ the rapid increase in both quantities is shifted to lower temperatures. As shown in the insets of Figs. 3 and 4 at a certain fixed temperature both $\chi$ and $\xi$ decrease rapidly with $J_2$.

Since the susceptibility (together with the correlation length) is an important quantity to analyze the critical properties for $T \to 0$, we first test the quality of our RGM approach in more detail by comparing the RGM data for $N = 16$ and $N = 20$ with ED data for the same system sizes, see Fig. 5. It is obvious, that the RGM and the ED data for $\chi$ are in excellent agreement. This is consistent with our previous finding\textsuperscript{28} that for the 1D Heisenberg ferromagnet the RGM data for $\chi(T \to 0)$ and $\xi(T \to 0)$ are in perfect agreement with exact Bethe ansatz results. Moreover, the RGM calculations of $\chi$ for finite $N$ allow to estimate the magnitude of the finite-size effects. As can be clearly seen in Fig. 5, the finite-size effects at $T \lesssim 0.6$ ($T \lesssim 0.4$) for $J_2 = 0.1$ ($J_2 = 0.3$) become very large. Since the correlation length $\xi$ becomes smaller with increasing $J_2$ (see the inset in Fig. 4), the finite-size effects become less pronounced with growing frustration. Nevertheless, based on our data we argue that the finite systems of $N = 16$ and $N = 20$, accessible...
by exact full diagonalization, are not representative for the thermodynamic limit, see Refs. 10 and 17. Note that for the 1D case, the ED data for \( N = 20 \) are in good agreement with RGM data for \( N \to \infty \).\textsuperscript{28} Consistent to that observation, in two dimensions the system of \( N = 400 \) sites with the same linear extension is already close to the thermodynamic limit, see Fig. 5.

Next we use the RGM for \( N \to \infty \) to investigate the critical behavior of \( \chi \) and \( \xi \) for \( T \to 0 \) in more detail. We start with a brief discussion of the low-temperature behavior of the unfrustrated ferromagnet. Contrary to the 1D case, where for \( J_2 = 0 \) exact Bethe results are available,\textsuperscript{28} we have only approximate results for the 2D model. From low-temperature expansions of the susceptibility and the correlation length for the model with \( J_2 = 0 \) using renormalization group approaches,\textsuperscript{29,31} the modified spin-wave theory,\textsuperscript{29} and the RGM,\textsuperscript{32} it is known that for \( T \to 0 \) the susceptibility behaves as \( \chi \propto T^\alpha \exp \left( b/T \right) \) and the correlation length as \( \xi \propto T^\beta \exp \left( \beta/T \right) \). While different leading exponents \( \alpha \) and \( \beta \) (for the less important) preexponential factor were obtained by different methods, the exponential divergence is obtained by all authors.\textsuperscript{29–31,37} However, different values for the coefficients \( b \) and \( \beta \) were reported, namely, \( b(J_2 = 0) = \pi/2 \) using RGM\textsuperscript{32} \( b(J_2 = 0) = \pi \) using modified spin-wave theory\textsuperscript{29} or renormalization group approach\textsuperscript{33} and \( b(J_2 = 0) = 0.1327\pi \) using a different version of the renormalization-group approach.\textsuperscript{33} In all these papers\textsuperscript{29–31,37} it was found that the coefficients in the exponents fulfill the relation \( \beta = b/2 \). We mention further, that early numerical studies based on quantum Monte-Carlo calculations (using, however, \( \chi \) and \( \xi \) data only for quite large temperatures) give \( b(J_2 = 0) \approx 4.5 \approx 1.43\pi \) (Ref. 33) and \( \beta(J_2 = 0) = 0.254\pi \) (Ref. 32). Let us finally argue, that the results for the coefficient \( b \) obtained by modified spin-wave theory\textsuperscript{29} as well as renormalization group approach\textsuperscript{31} and by the RGM\textsuperscript{32} seem to be most reliable, since these methods are well-tested. Moreover, for the 1D spin-1/2 Heisenberg ferromagnet it was shown that the RGM\textsuperscript{28,37} as well as the modified spin-wave theory\textsuperscript{29} reproduce the exact Bethe-ansatz results for the low-temperature behavior of \( \chi \) and \( \xi \). Hence, it is to some extent surprising, that there is a difference by a factor of 2 in the coefficient \( b \) between the value obtained by modified spin-wave theory\textsuperscript{29} (as well as renormalization group approach\textsuperscript{31}) and the RGM for the 2D unfrustrated ferromagnet. This discrepancy is a known but unresolved problem.\textsuperscript{34,37} However, we believe that our general conclusions concerning the exponential divergence of the frustrated model, see the discussion below, are not affected by this problem.

Next we use the RGM results to determine the coefficients \( b \) and \( \beta \) as functions of \( J_2 \). We assume that the general low-\( T \) behavior of these quantities is preserved for \( 0 < J_2 < J_2^c \), cf. Ref. 28, and fit our numerical RGM data for \( \chi \) and \( \xi \) at low temperatures using the ansatz of \( \chi = \left( a_0 T^{\pi} + a_1 + a_2 T \right) \exp \left( \frac{b}{T} \right) \) and \( \xi = \sqrt{(a_0 T^{\pi-1} + a_1 + a_2 T)} \exp \left( \frac{b}{T} \right) \). Note that the leading power \( T^{-1} \) in the preexponential functions was derived for the unfrustrated case with the RGM in Ref. 37. We consider \( b, a_0, a_1, \) and \( a_2 \) as well as \( \beta, a_0, a_1, \) and \( a_2 \) as independent fit parameters. Recall that we can calculate RGM data only for temperatures down to a certain \( T_0 \), where \( T_0 \) (e.g., \( T_0 = 0.161 \) for \( J_2 = 0 \) and \( T_0 = 0.052 \) for \( J_2 = 0.4 \)) is the lowest temperature where the system of RGM equations converges (see Sec. 11). Thus, in addition to the leading power \( T^{-1} \) in the preexponential function it is reasonable to consider higher order terms to achieve an optimal fit of the RGM data. For the fit we use 500 equidistant data points in the interval \( T_0 \ldots T_{cut} \), where \( T_{cut} \) is set to 0.05. The fit of the numerical RGM data reproduces the analytical results of Ref. 37 for \( J_2 = 0 \), i.e., \( b(J_2 = 0) = 2\beta(J_2 = 0) = \pi/2 \), with a precision of four digits.

After having tested our fitting procedure by comparison with the analytical predictions for \( J_2 = 0 \), we now consider the frustrated model, where (to the best of our knowledge) no other results are available. From our numerical data for \( \chi \) and \( \xi \) we determine the \( J_2 \) dependence of the coefficients \( b \) and \( \beta \). We find that the numerical data for \( b \) and \( \beta \) obtained by the fitting procedure described above are very well described by a linear decrease in both parameters with increasing frustration,

\[
\begin{align*}
b &= 2\beta = -\frac{\pi}{2} (J_1 + 2J_2).
\end{align*}
\]

Obviously, both parameters would be zero at the classical transition point \( J_2 = 0.5 \), but they are still finite at the transition point \( J_2^c \approx 0.44 \) of the quantum model. Hence, the exponential divergence is present in the full parameter range \( J_2 \leq J_2^c \) where the ground state is ferromagnetic. We emphasize that this result is contrary to the behavior observed for the 1D frustrated spin-1/2 ferromagnet, where the critical properties change at the zero-temperature transition point.\textsuperscript{28} We mention further that the leading coefficients \( a_0 \) and \( a_0 \) of the preexponential functions, see the expressions for \( \chi \) and \( \xi \) given above, vanish at the transition point \( J_2^c \), whereas the next coefficients \( a_1 \) and \( a_1 \) remain finite at \( J_2^c \). Hence, the preexponential temperature dependence is changed approaching the zero-temperature transition. Let us mention, that the linear decrease in the coefficients \( b \) and \( \beta \), see Eq. (11), found by fitting the low-temperature behavior of \( \chi \) and \( \xi \) is the same as that obtained analytically for the zero-temperature spin-stiffness \( \rho_s \), see Sec. 11. This relation between \( \rho_s \) and the divergence of the correlation length and the susceptibility is in accordance with general arguments\textsuperscript{35,39} concerning the low-temperature physics of low-dimensional Heisenberg ferromagnets.

Another interesting quantity is the specific heat \( C_V \) shown in Fig. 6. For \( J_2 = 0 \) the specific heat exhibits a typical broad maximum at about \( T = 0.562 \). Increasing the frustration the height of this maximum becomes smaller and it is shifted to lower temperatures.
Interestingly, within our RGM approach the shape of the $C_V(T)$ curve changes for large frustration. For $J_2 \gtrsim 0.34$ the specific heat shows two maxima, one at low temperatures ($T < 0.193$) and another one at high temperatures ($T > 0.6$). This extra low-temperature maximum signals the emergence of an additional low-energy scale when approaching the transition point $J_2^\ast$. Then many low-lying multiplets appear above the fully polarized ferromagnetic ground-state multiplet. Hence, the appearance of the additional low-temperature maximum in $C_V(T)$ can be attributed to a subtle interplay between all of these low-lying states. Note that a similar behavior was found for the 1D case.

**IV. SUMMARY**

In this paper we investigated the influence of the frustrating NNN coupling $J_2 < J_2^\ast$ on the thermodynamic properties of the 2D spin-1/2 Heisenberg ferromagnet using the spin-rotation-invariant Green’s function method (RGM). The RGM estimate for the critical frustration $J_2^\ast$, where the ferromagnetic ground state breaks down, is $J_2^\ast \approx 0.44|J_1|$.

We tested the method by comparing RGM data for the spin-spin correlation functions and the uniform susceptibility $\chi$ calculated for finite lattices of $N = 16$ and $N = 20$ sites with corresponding exact-diagonalization data for the same lattice sizes and found a good agreement between both methods. However, the comparison of RGM data for finite lattices and for $N \to \infty$ indicates strong finite-size effects at low temperatures. This leads to the conclusion that the finite systems of $N = 16$ and $N = 20$ are not representative for the low-temperature thermodynamics of large systems.

As it is known from the Mermin-Wagner theorem the thermal fluctuations are strong enough to suppress magnetic LRO for the Heisenberg ferromagnet in dimension $D < 3$ at any finite temperature. Due to frustration the fluctuations are further enhanced. Thus, frustration leads to a significant suppression of magnetic correlations at finite temperatures even if the ground state remains ferromagnetic.

The low-temperature behavior of the susceptibility $\chi$ and the correlation length $\xi$ in the ferromagnetic ground-state region $J_2 < J_2^\ast$ exhibits the exponential divergences $\chi \propto \exp \left( \frac{\pi |(J_1 - 2J_2)|}{2D} \right)$ and $\xi \propto \exp \left( \frac{\pi |(J_1 - 2J_2)|}{4D} \right)$. Although the numerator in the exponent decreases with growing $J_2$ the exponential divergence perpetuates in the whole parameter region $J_2 \leq J_2^\ast$.

The specific heat calculated within the RGM exhibits a double-maximum structure for $J_2 \gtrsim 0.34$ analogous to the 1D model.

To test our theoretical predictions it would be interesting to reconsider experimentally quasi-2D spin-1/2 ferromagnets, such as $K_2CuF_4$, $Cs_2CuF_4$, $Cs_2AgF_4$, $La_2BaCuO_5$, and $Rb_2CrCl_3$ with respect to a possible frustration and its influence on low-temperature thermodynamics.

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