Green’s function theory of quasi-two-dimensional spin-half Heisenberg ferromagnets: stacked square versus stacked kagomé lattice

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We consider the thermodynamic properties of the quasi-two-dimensional spin-half Heisenberg ferromagnet on the stacked square and the stacked kagomé lattices by using the spin-rotation-invariant Green’s function method. We calculate the critical temperature $T_C$, the uniform static susceptibility $\chi$, the correlation lengths $\xi$, and the magnetization $M$ and investigate the short-range order above $T_C$. We find that $T_C$ and $M$ at $T > 0$ are smaller for the stacked kagomé lattice which we attribute to frustration effects becoming relevant at finite temperatures.

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Introduction: Quasi-two-dimensional magnets have attracted much attention in recent years. According to the Mermin-Wagner theorem magnetically strictly two-dimensional (2D) Heisenberg magnets do not possess magnetic long-range order (LRO) at any finite temperature. Therefore, the exchange coupling between planes is crucial for the existence of a finite critical temperature $T_C$. Though most of the quasi-2D magnetic insulators are antiferromagnets there are also some quasi-2D ferromagnetic insulators like $K_2CuF_4$, $La_2BaCuO_{5}$, $Cs_2AgF_4$. The calculation of thermodynamic properties for quasi-2D magnets is an important issue in particular with respect to the interpretation of experimental results. Especially in the limit of weak interlayer coupling the evaluation of $T_C$ is challenging. Recently accurate values for the critical temperature of a Heisenberg magnet on a stacked square lattice by Monte Carlo calculations have been presented. Another promising approach to calculate the thermodynamics of quasi-2D magnetic systems is a spin-rotation invariant Green’s function method (RGM). The RGM allows a consistent description of LRO as well as of short-range order at arbitrary temperatures in magnetic systems of arbitrary dimension.

In this paper we use the RGM for the calculation of thermodynamic quantities of the quasi-2D spin-half Heisenberg ferromagnet on the stacked square and the stacked kagomé lattice. Both lattices have the same coordination number, but differ in their lattice geometry within the layers. Note that corresponding RGM treatments for the respective stacked antiferromagnets were given in Refs. 16 and 18. The Heisenberg antiferromagnet on the kagomé lattice is a canonical model to study frustration effects in 2D spin-half systems, see, e.g. Refs. 17, 21, 22, 23. Due to strong frustration there is most likely no magnetic LRO in the kagomé antiferromagnet. It has been argued recently that even the increase of the dimension in a stacked kagomé antiferromagnet does not lead to magnetic LRO.

In this paper we consider layered ferromagnets. Then geometric frustration is irrelevant in the ground state. However, at finite temperatures excited states with antiferromagnetic correlations contribute to the partition function. The energy of such states is influenced by triangular spin configurations and thus the frustrating geometry of the kagomé lattice may become more and more relevant with increasing temperature. By comparison with the unfrustrated stacked square lattice we can discuss these frustration effects.

We consider the Heisenberg model of $N$ spins 1/2

$$H = \frac{1}{2} \sum_{m,n} J_{m,n} \mathbf{s}_m \cdot \mathbf{s}_n, \quad J_{m,n} \leq 0, \quad (1)$$

where the sum runs over all $N$ unit cells (labeled by $m$ and $n$) and all spins within a unit cell (labeled by running indices $\alpha$ and $\beta$, see Fig. 1 for the kagomé case). The exchange coupling $J_{m,n}$ is nonzero for nearest neighbors, only. The ferromagnetic exchange parameter within the layers $J_\parallel$ is fixed to $J_\parallel = -1$.

Spin-Rotation-Invariant Green’s Function Method:
The rotation-invariant decoupling scheme introduced by Kondo and Yamaji\textsuperscript{12} goes one step beyond the random-phase approximation (RPA). An advantage of this method is the possibility to treat magnetic order-disorder transitions driven by quantum fluctuations as well as by thermal fluctuations and to provide a good description of magnetic SRO\textsuperscript{12,13,14,15,16,17,18,19,20}. In this paper we only illustrate some basic features of the RGM. For more details the reader is referred to Refs. 12,13,14,15,16,17,18,19,20 and in particular to Refs. 16,18,20 where the corresponding stacked antiferromagnets are considered.

To evaluate all relevant correlation functions (S\textsubscript{mα}S\textsubscript{nβ}) and several thermodynamic quantities we have to calculate a set of Fourier-transformed commutator Green’s functions \((\langle S^+_{qα}S^-_{qβ} \rangle_ω)\) which are connected with the dynamic spin susceptibilities by \(\chi_{qαβ}(ω) = -\langle S^+_{qα}S^-_{qβ} \rangle_ω\). Using the equations of motion and supposing spin-rotational invariance, i.e. \((S^+_{ma}) = 0\), we obtain \(ω^2(\langle S^2_{qα}S^2_{qβ} \rangle_ω = \langle \langle S^2_{qα}S^2_{qβ} \rangle \rangle_ω \)-the operator \(-\hat{S}^+_q = [\hat{S}^+_q, H], [H]\) written in site representation contains products of three spin operators along nearest-neighbor sequences which are treated in the spirit of the decoupling scheme of Shimahara and Takada.\textsuperscript{13} For example, the product \(S^-_{qα}S^+_{qβ}S^-_{qγ}C\) is replaced by \(η_{αβ}\hat{S}^+_{q}\hat{S}^+_{q}\hat{S}^-_{q}\), where \(A, B, C\) represent spin sites. The vertex parameters \(η_{µ}\) are introduced to improve the approximation. In the minimal version of the RGM we introduce just as many corresponding stacked antiferromagnets are considered.

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The ground-state energy (lower bound of the spectrum) is $E_{\text{min}} = -0.5N$ for both lattices. However, the upper bound for the kagomé lattice $E_{\text{max}} = 0.434N$ is much lower than the corresponding value $E_{\text{max}} = 0.670N$ for the square lattice (see, e.g., Ref. 1). This upper bound is equal to the ground-state energy of the kagomé antiferromagnet (i.e., $J_s = +1$) multiplied by minus one, and its small value signals strong frustration effects being relevant for eigenstates with antiferromagnetic spin-spin correlations. Hence, one can expect that excited states with antiferromagnetic spin correlations have lower energy for the kagomé ferromagnet resulting in a larger contribution to the partition function at $T > 0$ in comparison with the square-lattice ferromagnet. The observation $T_C^{\text{kagomé}} < T_C^{\text{square}}$ corresponds to the data for the susceptibility $\chi$ and the correlation lengths $\xi_s$, i.e., $\chi$ and $\xi_s$ for the stacked kagomé lattice are smaller than the corresponding values for the stacked square lattice (see below, Fig. 3).

In the limit $J_\parallel/J_s \ll 1$ we derive more explicit expressions for $T_C$ following Ref. 4. We find $T_C = -J_s \pi/(2 \ln(-20T_C/|J_s|))$ for the stacked square lattice and $T_C = -3J_s \pi/(8 \ln(-20T_C/|J_s|))$ for the stacked kagomé lattice. Note that these expressions are of similar form as corresponding formulas for the Neél temperature $T_N$ obtained by selfconsistent spin-wave theories. We compare our data for $T_C$ in Tab. 1 with results for the Neél temperature $T_N$ of the stacked square-lattice antiferromagnet. For the simple cubic lattice (i.e., $J_s = J_\parallel$) our result for $T_C$ exceeds the best available value for $T_C$ obtained by high-order high-temperature series expansion by 9% which can be considered as a test of the accuracy of the RGM results. This accuracy is much better than that of the Schwinger boson approach (compare 4th and 5th columns in Tab. 1). Note that the accurate results of Ref. 11 show that $T_N > T_C$ for unfrustrated spin-half Heisenberg magnets, whereas an RPA treatment yields $T_N = T_C$. Therefore, the relation between $T_C$ and $T_N$ can be used as a further test of the RGM. Indeed, using the same set of vertex parameters for $J_s = J_\parallel$ we obtain $T_N = 1.1225T_C$ which is in excellent agreement with Ref. 11.

The overall dependence of $T_C$ versus $J_\parallel$ resembles the results of Yasuda et al. for $T_N$, see Tab. 1. Therefore, we adopt their empirical formula

$$T_C = \frac{A}{B - \ln(J_s/J_\parallel)}.$$  (2)

The best agreement with our data is obtained for $A = 2.414$, $B = 2.049$ and $B = 2.506$, $2.315$ for the stacked square (kagomé) ferromagnet, cf. Fig. 2.

The correlation lengths and the uniform static susceptibility are shown as functions of $T$ in Fig. 3 for
interlayer coupling $J_\perp = -0.05$ in Fig. 3. For this value of $J_\perp$ we have $T_C = 0.4388$, i.e. for $T > 0.4388$ the long-range part of the spin-spin correlation function vanishes. It is obvious that the short-range intralayer and interlayer correlators behave differently. The interlayer correlators become very small at $T = T_C$ whereas the intralayer spin-spin-correlations are still well-pronounced. This behavior corresponds to the data for the intra- and interlayer correlation lengths, see Fig. 2 and indicates the crossover from three-dimensional magnetic LRO at $T < T_C$ to 2D SRO at $T > T_C$, if $|J_\parallel| \ll |J_\perp|$.

**Summary:** In this paper the thermodynamics of the spin-half Heisenberg ferromagnet on the stacked square and kagomé lattices has been investigated applying a spin-rotation-invariant Green’s function method. We have calculated the Curie temperatures $T_C$ in dependence on the interlayer coupling $J_\perp$, and simple empirical formulas for $T_C(J_\perp)$ are obtained from the numerical data. Studying short-range spin-spin correlation functions we see clearly a dimensional crossover at $T \sim T_C$ for strongly anisotropic systems. Comparing the values of $T_C$, of the magnetization, the correlation lengths and of the uniform static susceptibility we come to the conclusion that at $T > 0$ frustration becomes relevant for the stacked kagomé lattice leading to a weakening of magnetic order at finite temperatures and to $T_{C}^{\text{kagome}} < T_{C}^{\text{square}}$.

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The stacked square-lattice Heisenberg antiferromagnet was treated within the RGM in Ref. 16. However, in that paper an additional vertex parameter was introduced which results in slightly different values for $T_N$ compared to the scheme used in the present paper.