The spin-1 $J_1$-$J_3$ Heisenberg model on a triangular lattice

P Rubin and A Sherman
Institute of Physics, University of Tartu, W. Ostwaldi 1, 50411 Tartu, Estonia
E-mail: rubin@ut.ee

Abstract. Motivated by the experimental data for NiGa$_2$S$_4$, the spin-1 Heisenberg model on a triangular lattice with the ferromagnetic nearest- and antiferromagnetic third-nearest-neighbor exchange interactions, $J_1 = -(1 - p)J$ and $J_3 = pJ, J > 0$, is studied in the range $0 \leq p \leq 1$. Mori's projection operator technique and the Lanczos exact diagonalization are used. Mori's method retains the rotation symmetry of spin components and does not anticipate any magnetic ordering. For zero temperature several phase transitions are observed. At $p_{cr} \approx 0.2$ the ground state is transformed from the ferromagnetic spin structure into a disordered state, which in its turn is changed to an antiferromagnetic long-range ordered state with the incommensurate ordering vector $Q' \approx (1.16, 0)$ at $p \approx 0.31$. With growing $p$ the ordering vector moves along the $X$ axis to the commensurate point $Q_c = (2\pi/3, 0)$ which is reached at $p = 1$. The final state with an antiferromagnetic long-range order can be conceived as four interpenetrating sublattices with the 120$^\circ$ spin structure on each of them. The model is able to describe the state with the incommensurate short-range order observed in NiGa$_2$S$_4$. To verify the used approach the ground state energy and corresponding spin-spin correlations are compared with exact-diagonalization results obtained with the SPINPACK code (the Lanczos exact diagonalization). Results of the two methods are in qualitative agreement.

1. Introduction
Investigation of the spin-1 Heisenberg model on a two-dimensional (2D) triangular lattice is of interest in connection with recently synthesized compound NiGa$_2$S$_4$ [1]. Magnetism in this system is mainly connected with Ni$^{2+}$ ($S = 1$) ions, which form a 2D triangular lattice. Experiments on neutron scattering reveal a short-range antiferromagnetic order, being characterized by an incommensurate ordering vector. For the description of the observed order the classical $J_1 - J_3$ 2D Heisenberg model on a triangular lattice was proposed [1]. The dominating antiferromagnetic interaction $J_3$ between the third nearest neighbors (TNN) and the weak ferromagnetic interaction $J_1$ between the nearest neighbors (NN) were taken into account. This model agrees with first-principle calculations of the exchange parameters [2]. In this work the corresponding quantum model with $S = 1$ is considered with the aim to describe all phases of the model.

2. Ground state properties in Mori’s approach
The Hamiltonian of the model reads

$$ H = \frac{1}{2} \sum_{nn} J_{nn} \left( s_n^z s_m^z + s_n^{+1} s_m^{-1} \right), \quad (1) $$
where $s_n^±$ and $s_m^{±1}$ are components of the spin-1 operators, $n$ and $m$ label sites of the triangular lattice. Only the NN and TNN interactions are taken into account,

$$J_{nm} = \sum_a (J_1 \delta_{n,m+a} + J_3 \delta_{n,m+2a}),$$

with the vectors $a$ and $2a$ connecting the NN and TNN sites. We use the lattice spacing $a = |a|$ as the unit of length. The exchange interactions, $J_1 = -(1 - p)J$ and $J_3 = pJ$, $J > 0$, are expressed through the frustration parameter $p$, which varies in the full range from 0 to 1 ($J$ is set as the unit of energy). Mori’s projection operator technique [3] gives the following expression for the spin Green’s function

$$D(k\omega) = \frac{6J[(1 - p)[1 - \gamma(k)]C_1 - p[1 - \gamma(2k)]C_{2a}]}{\omega^2 - \omega_k^2},$$

(2)

where $\gamma(k) = \frac{1}{2} \cos(k_x) + \frac{2}{3} \cos \left(\frac{k_y}{2}\right) \cos \left(\frac{k_z \sqrt{3}}{2}\right)$, the frequency of spin excitations $\omega_k$ contains products of $[1 - \gamma(k)]$, $[1 - \gamma(2k)]$ and spin correlations such as $C_1 = \langle s_n^{+1} s_{n+a}^{-1}\rangle$ and $C_{2a} = \langle s_n^{+1} s_{n+2a}^{-1}\rangle$. These correlations can be expressed through $D(k\omega)$ and calculated self-consistently. Details of our approach can be found in [4, 5]. If the system has a long-range order at $T = 0$ the summation over the wave vector in an expression for the spin correlation function can be divided into the contribution yielded at the ordering vector $k = \mathbf{Q}$, which is proportional to the condensation part $C$, and the fluctuation contribution given by other wave vectors [6]. $C$ plays the role of the order parameter. In the case $|J_1| > J_3$ the ground state is ferromagnetic and the ordering vector $\mathbf{Q} = (0,0)$. We have found that the long-range ferromagnetic order exists for $0 \leq p \leq p_c$, $p_c \approx 0.2$. In this range of $p$ the ferromagnetic condensation part is practically constant, and it vanishes abruptly at $p \approx p_c$. At $T = 0$ the spectra of magnetic excitations for different values of $p$ are shown in figure 1. As it can be seen, in the ferromagnetic range the dispersion $\omega_k$ is parabolic near the $\Gamma$-point, which is typical for a ferromagnet. With the growth of the antiferromagnetic exchange $J_3$ the system passes into a disordered state, where the condensation part is equal to zero. When the value $p \approx 0.31$ is reached the system goes into the state with the long-range antiferromagnetic order with the incommensurate ordering vector $\mathbf{Q}' \approx (1.16, 0)$ . The frequency of magnetic excitations vanishes at this wave vector (see figure 1) and a corresponding condensation part is finite. With further growth of the frustration parameter $p$ the ordering vector moves along the $X$ axis from the point $\mathbf{Q}'$ to $\mathbf{Q}_c = (2\pi/3, 0)$, which is reached at $p = 1$. For this $p$ the incommensurate antiferromagnetic order transforms to a commensurate phase, which can be conceived as four interpenetrating $120^\circ$ spin structures on sublattices with twice as large lattice spacing. Spin orientations in these sublattices are independent of one another.

At $p \approx 0.82$ the calculated ordering vector is close to that observed in NiGa$_2$S$_4$ [1]. For the finite temperature of the experiment, in accord with the Mermin-Wagner theorem, our long-range order passes into the short-range one. However, its calculated correlation length is an order of magnitude larger than the experimental value. This discrepancy can be related to the influence of impurities.

3. Lanczos exact diagonalization

Applying Mori’s method one has to use a number of approximations. Therefore, it is of interest to study the same model with another method and compare obtained results [7]. To do this we employ Schulenburg’s SPINPACK code [8]. This package is intended for exact diagonalization (ED) of finite spin systems using the Lanczos algorithm. For $0 \leq p \leq 1$ we found energies of the ground, low-lying excited states and spin-spin correlation functions for lattices containing
Figure 1. The dispersion of spin excitations $\omega_k$ along the $X$ axis for different values of the frustration parameter $p$ at $T=0$.

Figure 2. The triangular lattices with $N=16$ and $N=20$ sites studied in this work.

$N=16$ and $N=20$ sites. These lattices are shown in figure 2. The periodic boundary conditions were used.

Dependencies of the ground and low-lying excited state energies of the $N=20$ lattice on the frustration parameter $p$ are shown in figure 3 (corresponding dependencies for the $N=16$ lattice are, in general, analogous). As seen, the ground state (GS) of the $N$-site lattice is transformed from the classic ferromagnet ($S_{\text{tot}} = N$) to the singlet state at some critical value of $p$. For the lattices with $N=16$ and $N=20$ these critical values are $p_{16} \approx 0.45$ and $p_{20} \approx 0.28$, respectively. As mentioned above, in Mori’s approach we found that the transition from the ferromagnetically ordered state to a spin disorder occurs at the value of the frustration parameter $p_{cr} \approx 0.2$. The condensation part vanishes at this value of the frustration parameter. Thus $p_N$ and $p_{cr}$ indicate a border of the ferromagnetic region. It can be supposed that with the rise of the lattice size $p_N$ will tend to the value $p_{cr}$ obtained in Mori’s approach because $p_{cr}$ was obtained in a $216 \times 216$ lattice [5]. In figure 3a the dependencies of the GS energy ($E_{\text{GS}}, S_{\text{rot}} = N = 20$) and the first excited state ($E_1, S_{\text{tot}} = 19$) on $p$ are shown. These dependencies are linear. Differences between the GS and excited-state energies disappear at the critical value $p_{20}$. For $p > p_{20}$ the energies
of the low-lying states are shown in figure 3b. The GS is a singlet, $S_{\text{tot}} = 0$. The lowest excited states are characterized by $S_{\text{tot}} = 0$ ($E_2$) and $S_{\text{tot}} = 1$ ($E_3$).

In figure 4 we compare the dependencies of the GS energy per site on the frustration parameter $p$, obtained by ED in the $N = 20$, $N = 16$ lattices and obtained by Mori’s projection operator technique in a $216 \times 216$ lattice. As a whole these dependencies are similar. As one can see, the dependencies are linear in the ferromagnetic region ($p < p_{16}$, $p_{20}$ or $p_{cr}$). Besides, $p_{cr} < p_{20} < p_{16}$. This sequence of the critical values of $p$ seems reasonable taking into account the sizes of the lattices under consideration. For $p$ larger than these critical values all curves have maxima. However, positions of these maxima are different – the dependence is peaked at $p \approx 0.4$ in the results obtained with Mori’s technique, at $p \approx 0.7$ in the $N = 16$ and at $p \approx 0.9$ in the $N = 20$ ED results.

Spin correlation functions for nearest and third-nearest neighbors are shown in figure 5. The data were obtained by the exact diagonalization in the $N = 20$ lattice and by Mori’s technique in a $216 \times 216$ lattice. In panels a) and b) ED correlations are constant and equal to unity in the ferromagnetic phase for $p < p_{20}$. In this region correlation functions obtained by Mori’s method are also constant. However, they are somewhat smaller than one due to approximations made for the considered $S = 1$ Heisenberg model [5]. The interaction between nearest neighbor spins vanishes at $p = 1$, which manifests itself in the vanishing correlation $\langle S_0 S_a \rangle$. The correlation $\langle S_0 S_{2a} \rangle$ depends only weakly on $p$ in the range $p > p_{20}$ and $p > p_{cr}$. Notice that the phase transition from the spin disordered state to the state with the incommensurate antiferromagnetic long-range order at $p \approx 0.31$ does not reveal itself in figure 5. As seen from the figure, curves obtained in the larger lattice are more smooth than those in the $N = 20$ lattice, which at least

![Figure 3](image-url)
Figure 4. The dependencies of the ground state energy per site ($E_{GS}/N$) on the frustration parameter $p$ obtained by ED in the $N = 20$ lattice (red circles and solid line), $N = 16$ lattice (blue triangles and dash-dotted line), and obtained by Mori’s approach in a $216 \times 216$ lattice (black squares and dashed line). The parameters $p_{20}$ and $p_{16}$ are values of $p$, at which the transition from the ferromagnetic to the singlet GS occurs. $p_{cr}$ corresponds to the transition to a disordered state.

Figure 5. The nearest neighbor (a) and third-nearest neighbor spin correlations obtained by ED in the $N = 20$ lattice (circles and dashed lines) and by Mori’s technique in a $216 \times 216$ lattice at $T = 0$ (squares and solid lines).

partly may be connected with finite-size effects. However, spin correlations calculated in the two lattices by two different methods are in general close and behave similarly with changing the frustration parameter. We can conclude that results obtained with the approximate approach based on Mori’s projection operator technique are in reasonable agreement with the exact-diagonalization data.

4. Concluding remarks
In this work, Mori’s projection operator technique and the Lanczos exact diagonalization have been applied for investigating magnetic properties of the quantum $J_1$-$J_3$ Heisenberg model
on a triangular lattice in the entire range of the frustration parameter $0 \leq p \leq 1$, where $p = J_3/(J_3 - J_1)$. $J_1$ and $J_3$ are the nearest and third-nearest exchange constants. With the use of Mori’s method we have shown that at zero temperature for different values of $p$ the system is characterized by the ferromagnetic ordering, spin disorder, incommensurate and commensurate antiferromagnetic ordering. At $p \approx 0.82$ the model describes the key feature observed in NiGa$_2$S$_4$ [1] – the incommensurate short-range antiferromagnetic order at finite temperature. The results were verified using the exact diagonalization of small lattices with periodic boundary conditions. The SPINPACK package implementing the Lanczos algorithm was applied to find energies of the ground and low-lying exited states. Besides, spin-spin correlation functions between nearest- and third-nearest spins were calculated. We found qualitative and in some cases quantitative agreement between results obtained by exact diagonalization of small clusters and by Mori’s projection technique in larger lattices.

Acknowledgments

This work was supported by the research project IUT2-27 and the ESF Grant No. 9371.

References

[1] Nakatsuji S, Nambu Y, Tomonura H, Sakai O, Jonas S, Broholm C, Tsunetsugu H, Qiu Y and Maeno Y 2005 Science 309 1697-1700
[2] Mazin I I 2007 Phys. Rev. B 76 40406
[3] Mori H 1965 Prog. Theor. Phys. 34 399-416
[4] Sherman A and Schreiber M 2002 Phys. Rev. B 65 134520
[5] Rubin P, Sherman A and Schreiber M 2012 Phys. Lett. A 376 1062-66
[6] Shimahara H, Takada S 1991 J. Phys. Soc. Jpn. 60 2394-2405
[7] Rubin P, Sherman A and Schreiber M 2014 Phys. Lett. A 378 3572-74
[8] Schulenburg J Program package SPINPACK https://www-e.uni-magdeburg.de/jschulen/spin/.