Magnetic phase diagram of the spin-1 two-dimensional J1-J3 Heisenberg model on a triangular lattice

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MOTIVATION

A number of systems contain two-dimensional triangular lattices formed by ions with $S=1$: $\text{NiGa}_2\text{S}_4$, $\text{Ba}_3\text{NiSb}_2\text{O}_9$.

Despite strong AF interactions $\text{NiGa}_2\text{S}_4$ demonstrates an incommensurate AF short-range order at temperatures near $T=0$ (S. Nakatsuji et al., Science 309 (2005) 1697).
The $J_1$ - $J_3$ classical model with dominating 3\textsuperscript{rd}-nearest-neighbor AF interaction and weak nearest-neighbor ferromagnetic interaction was proposed by S. Nakatsuji (Science 309 (2005) 1697) to describe the incommensurate short-range order.
Ab-initio estimation of the exchange constants

(I.I.Mazin, PRB 76, 140406 (2007))

$J_3 > 0$ is anomalously large

$J_2$- negligibly small

$J_1$ is smaller then $J_3$ and possibly ferromagnetic

**FIG. 3.** (Color online) Calculated exchange constants for the first three neighbor shells in NiGa$_2$S$_4$, in meV, as a function of the Hubbard $U$, assuming the intra-atomic $J=0.07$ Ry. The additional entry at $U=0$ corresponds to $J=0$. 

S=1 two-dimensional Heisenberg model

\[ H = \frac{1}{2} \sum_{mn} J_{mn} (S^z_m S^z_n + S^{+1}_m S^{-1}_n), \]

\[ S^z_n = \sum_{\sigma = \pm 1} \sigma |n\sigma\rangle\langle n\sigma|, \]

\[ S^\sigma_n = \sqrt{2} (|n0\rangle\langle n, -\sigma| + |n\sigma\rangle\langle n, 0|) \]

We will consider this model on a triangular lattice with account of the nearest \( J_1 = (-1+p)J \) and the 3rd-nearest \( (J_3 = pJ) \) couplings, \( J > 0 \).
Magnetic Properties

The spin Green’s function \( D(k, t) = -i\theta(t)\left\langle [s^z_{k}(t), s^z_{-k}] \right\rangle \),

is obtained with the use of Mori’s projection technique (Mori, Progr. Theor. Phys. 34 (1965) 399):

\[
\left( \left(s^z_{k}, s^z_{-k} \right) \right) = \frac{\left(s^z_{k}, s^z_{-k} \right)}{\omega - E_0 - \sum F_0},
\]

\[
D(k, \omega) = (\left(s^z_{k}, s^z_{-k} \right))\omega - \left(s^z_{k}, s^z_{-k} \right),
\]

\[
\omega - E_1 - \sum F_1.
\]
where \((A, B) = i \int_0^\infty dt \exp(-\eta t) \langle [A(t), B] \rangle, \eta \to 0\). The elements of the fraction \(E_n\) and \(F_n\) are determined from the recursive procedure:

\[
[A_n, H] = E_n A_n + A_{n+1} + F_{n-1} A_{n-1}, \quad n = 0, 1, 2 \ldots
\]

\[
E_n = ([A_n, H], A_n^\dagger)(A_n, A_n^\dagger)^{-1}, \quad F_n = (A_{n+1}, A_{n+1}^\dagger)(A_n, A_n^\dagger)^{-1}, F_{-1} = 0.
\]

The continued fraction is cut off by setting \(F_1 = 0\). \(F_0\) and \((s_k^z, s_{-k}^z)\) contain many-particle Green’s functions. These Green’s functions are calculated approximately with the use of a decoupling.
The obtained spin Green’s function reads

\[
D(k\omega) = \frac{6J \left(-c_1(\gamma_k - 1)(1-p) + c_2a(\gamma_{2k} - 1)p\right)}{\omega^2 - \omega_k^2},
\]

where

\[
\omega_k^2 = 36J^2 \alpha \left\{ (1-p)^2(\gamma_k - 1) \left[ \frac{c_1}{6} + c_1\gamma_k - c_2 - \frac{2(1-\alpha)}{9\alpha} \right] + p^2(\gamma_{2k} - 1) \left[ \frac{c_2a}{6} + c_2a\gamma_{2k} - c_2 - \frac{2(1-\alpha)}{9\alpha} \right] - p(1-p) \left[ (1-\gamma_k)(c'' - \gamma_{2k}c_1) + (1-\gamma_{2k})(c'' - \gamma_k c_{2a}) \right] \right\}.
\]
\[ c_2 = \frac{1}{6} \left( \frac{4}{3} + 2\langle s_{n}^{+1} s_{n+d}^{-1} \rangle + 2c_1 + c_{2a} \right), \]

\[ c'' = \frac{1}{6} \left( 2\langle s_{n}^{+1} s_{n+r}^{-1} \rangle + 2\langle s_{n}^{+1} s_{n+d}^{-1} \rangle + c_1 + \langle s_{n}^{+1} s_{n+3a}^{-1} \rangle \right), \]

\[ c_{2}' = \frac{1}{6} \left( \frac{4}{3} + \langle s_{n}^{+1} s_{n+4a}^{-1} \rangle + 2c_{2a} + 2\langle s_{n}^{+1} s_{n+2d}^{-1} \rangle \right), \]

\[ c_1 = \langle s_{n}^{+1} s_{n+a}^{-1} \rangle, \quad c_{2a} = \langle s_{n}^{+1} s_{n+2a}^{-1} \rangle \]

are the spin correlations on neighboring sites,

\[ \gamma_k = \frac{1}{6} \sum_k \exp[ika], \]

\( \alpha \) is the vortex correction introduced to improve the decoupling procedure.
To find the parameters $\alpha, c_1, c_{2a}, c_2, c_2'$ and $c''$ one has to use relations connecting the spin correlations with Green's functions

$$
\langle s_{n}^{+} s_{m}^{-} \rangle = \frac{6J}{N} \sum_{k} \exp \left[ ik(n-m) \right] \frac{\left( c_{1}(\gamma_{k}-1)(p-1)+c_{2a}(\gamma_{2k}-1)p \right)}{\alpha_{k}} \coth \left( \frac{\alpha_{k}}{2T} \right)
$$

and the equation $\langle s_{n}^{+} s_{n}^{-} \rangle = \frac{4}{3}$, which follows from the constraint $\langle s_{n}^{2} \rangle = 2$
LRO or SRO? One has to analyze $\langle s_n^+ s_m^- \rangle$ at $T=0$

$$
\langle s_n^+ s_m^- \rangle = \frac{1}{N} \sum_{k \neq Q} \exp\left[ ik(n-m) \right] c_{ok} + \frac{1}{N} \sum_Q \exp\left[ iQ(n-m) \right] c_{Q}.
$$

The $1^{st}$ term describes local correlations, this term vanishes for large $|n-m|$. The $2^{nd}$ term - the condensation part – is connected to the LRO.
**LRO:** Ferromagnetic order (\(|J_1| \gg J_3\)), \(Q=\Gamma=[0,0]\),
Near the \(\Gamma\) point the serial expansion of \(\omega_k^2\) starts from the
terms proportional to \(k^4\).

\[
\langle s_n^+ s_m^- \rangle = \frac{1}{N} \sum_{k \neq \Gamma} \exp \left[ ik(n-m) \right] c_{ok} + C_F.
\]

**Antiferromagnetic order:**
\(\omega_k\) is vanishing at the nonzero ordering vector \(Q_i\).

\[
\langle s_n^+ s_m^- \rangle = \frac{1}{N} \sum_{k \neq Q} \exp \left[ ik(n-m) \right] c_{ok} + \sum_i \cos \left[ Q_i (n-m) \right] C_{AF}.
\]

**SRO:** \(\text{CAF} = 0, \text{CF} = 0\)
**Classical model**: the AF ordering vector $Q$ corresponds to a minimum of $J(k) = \sum_{i,j} J_{ij} \cos(k(R_i - R_j))$, $S_i = u \cos(QR_i) + v \sin(QR_i)$.

The dependencies of the scalar products between the nearest ($S_0 \cdot S_a$) and third nearest neighbor ($S_0 \cdot S_{2a}$) classical spins on the frustration parameter $p$. The inset shows the dependence of the length of the ordering vector $Q_{\text{cl}}$ on $p$. 
Quantum model. Mori’s approach. $T=0$

The dependencies of the ferromagnetic condensation part $C_F$ and the antiferromagnetic condensation part $C_{AF}$ on the frustration parameter $p$. 
CORRELATION FUNCTIONS

\[ C_1 = \langle S_{n}^{+1} S_{n+a}^{-1} \rangle \]

\[ C_{2a} = \langle S_{n}^{+1} S_{n+2a}^{-1} \rangle \]
Frequency of spin excitations

$p=0$

$p=0.2$
$p = 0.52$

$\omega_k$

$\kappa$

$p = 1$

$\omega_k$

$\kappa$

$\gamma$
The dispersion of spin excitations $\omega_k$ for different values of the frustration parameter $p$ at $T = 0$ along the $x$ axis. The inset shows $\omega_k$ at zero temperature (solid line) and at $T / J = 0.2$ (dashed line) for $p = 0.52$. 
RESUME

Using Mori's projection operator technique the phase diagram of the S=1 $J_1$-$J_3$ Heisenberg model on a 2D triangular lattice was found.

At the frustration parameter $p \approx 0.2$ the ground state is transformed from the long-range ordered ferromagnet into a disordered state, which is changed to an antiferromagnetic long-range ordered state with the incommensurate ordering vector $Q = Q_c \approx (1.16, 0)$ at $p \approx 0.31$. With increasing $p$ the ordering vector moves along the line $Q - Q_c$ to the commensurate point $Q_c = (2\pi/3, 0)$, which is reached at $p = 1$. The final state can be conceived as four interpenetrating sublattices with the $120^\circ$ spin structure on each of them. The results are applicable to NiGa$_2$S$_4$.