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

P. Rubin, A. Sherman

Institute of Physics, University of Tartu, Estonia

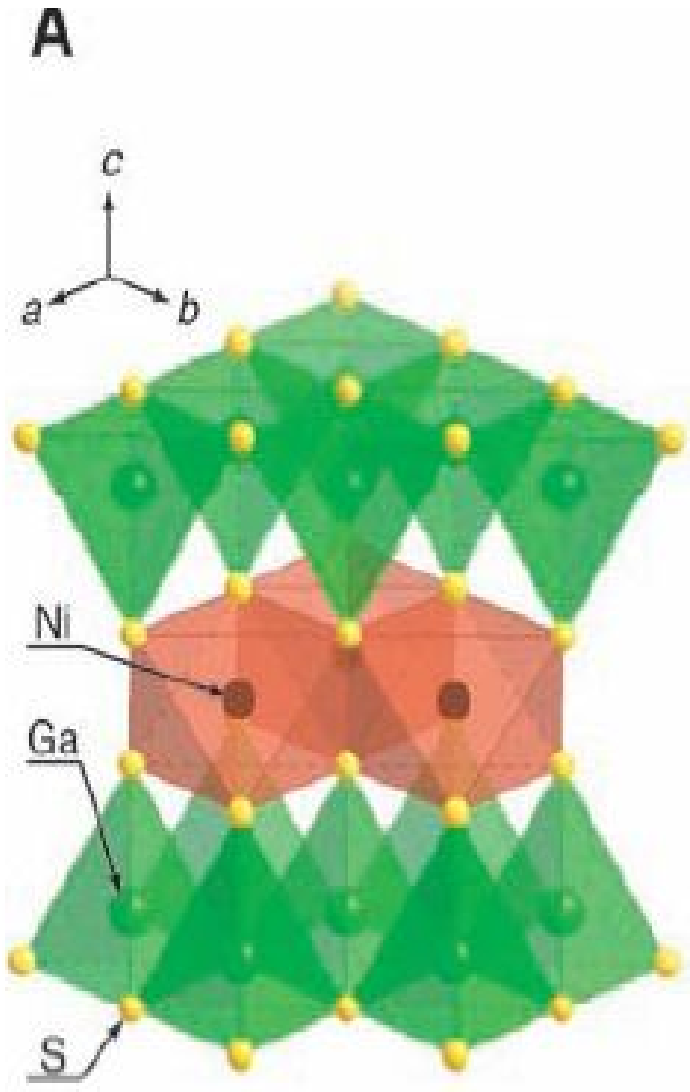
M. Schreiber

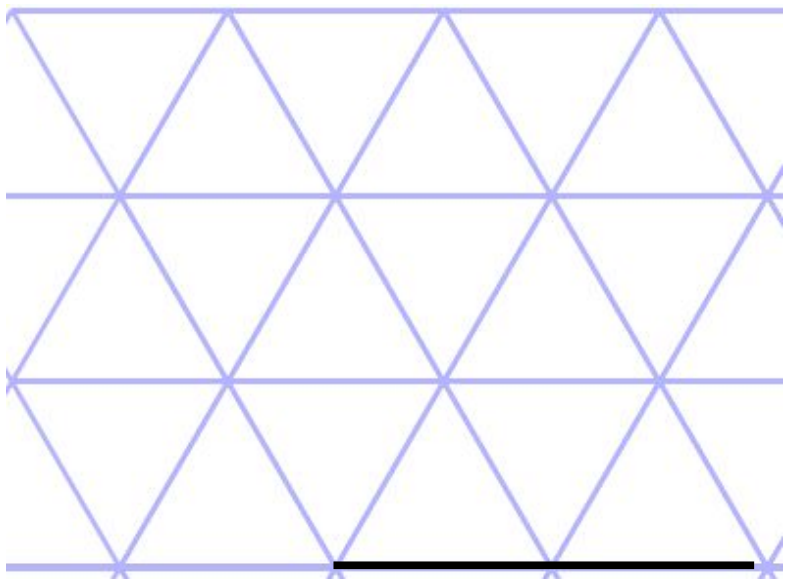
Institut für Physik,Technische Universität,Chemnitz,Germany

MOTIVATION

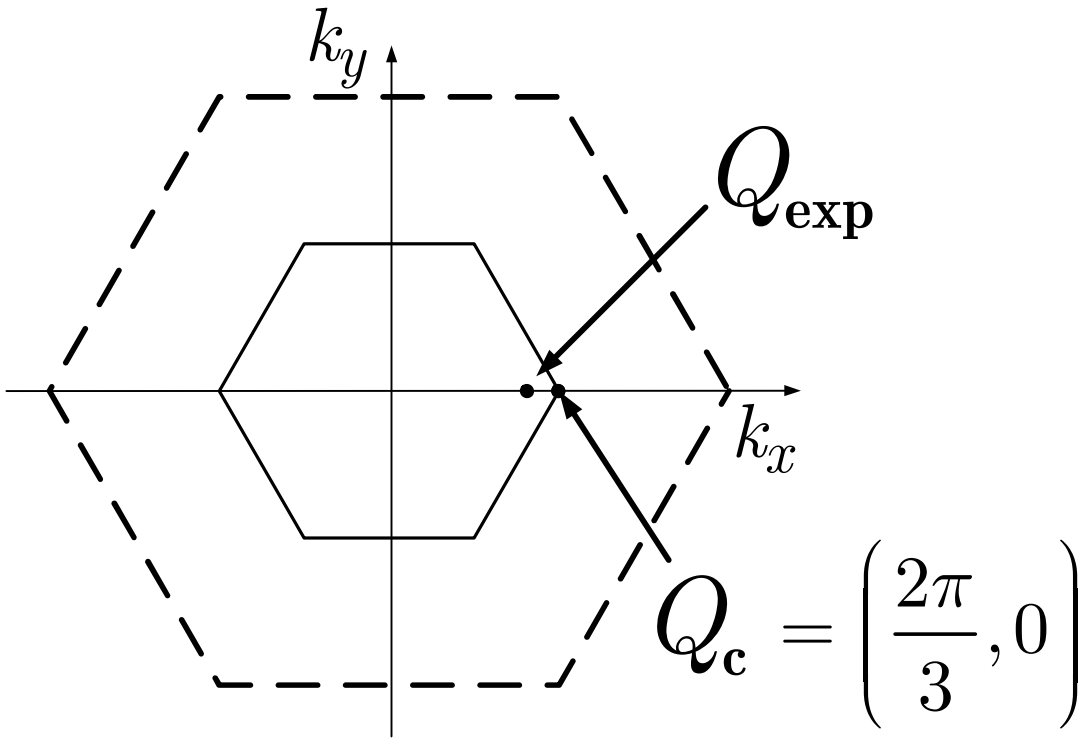
A number of systems contain *two-dimensional triangular lattices* formed by ions with **S=1**: **$NiGa_2S_4$**, **$Ba_3NiSb_2O_9$**.

Despite strong **AF** interactions **$NiGa_2S_4$** demonstrates an incommensurate **AF** short-range order at temperatures near T=0 (S. Nakatsuji et al., Science **309** (2005) 1697).





$J_1<0$ $J_3>0$



The J_1 - J_3 classical model with dominating 3rd-nearest-neighbor AF interaction and weak nearest-neighbor ferromagnetic interaction was proposed by S. Nakatsuji (Science **309** (2005) 1697) to describe the incommensurate shot-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

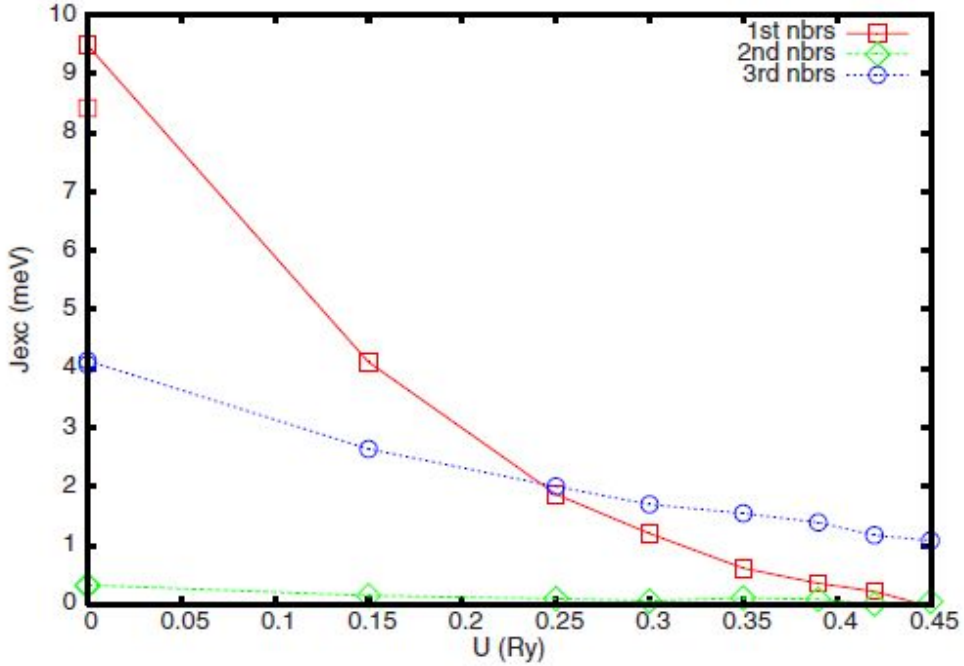


FIG. 3. (Color online) Calculated exchange constants for the first three neighbor shells in $NiGa_2S_4$, in meV, as a function of the Hubbard U, assuming the intra-atomic J=0.07 Ry. The additional entry at U=0 corresponds to J=0.

S=1 two-dimensional Heisenberg model

$$H = \frac{1}{2}\sum_{\mathbf{mn}} J_{\mathbf{mn}} \left(s_{\mathbf{m}}^{z} s_{\mathbf{n}}^{z} + s_{\mathbf{m}}^{+1} s_{\mathbf{n}}^{-1} \right),$$

$$s_{\mathbf{n}}^{z} = \sum_{\sigma=\pm 1} \sigma \left| n\sigma \right\rangle \left\langle n\sigma \right|,$$

$$s_{\mathbf{n}}^{\sigma} = \sqrt{2}\left(\left| n0 \right\rangle \left\langle n, -\sigma \right| + \left| n\sigma \right\rangle \left\langle n, 0 \right| \right)$$

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 $D(\mathbf{k},t) = -i\theta(t)\left\langle\left[s^z_{\mathbf{k}}(t), s^z_{-\mathbf{k}} \right]\right\rangle,$

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

$$\left(\left(s^z_{\mathbf{k}}, s^z_{-\mathbf{k}}\right)\right) = \cfrac{\left(s^z_{\mathbf{k}}, s^z_{-\mathbf{k}}\right)}{\omega - E_0 - \cfrac{F_0}{\omega - E_1 - \cfrac{F_1}{\ddots}}},$$

$$D(k,\omega) = ((s^z_{\kappa}, s^z_{-\kappa}))\omega - (s^z_{\kappa}, s^z_{-\kappa}),$$

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

$$\left[A_n,H\right]=E_nA_n+A_{n+1}+F_{n-1}A_{n-1},\quad n=0,1,2\ldots$$

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

The continued fraction is cut off by setting $F_1=0$. F_0 and $\left(s_{\mathbf{k}}^z,s_{-\mathbf{k}}^z\right)$ 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

$$D(\mathbf{k}\omega) = \frac{6J\left(-c_1(\gamma_{\mathrm{k}}-1)(1-p)+c_{2a}(\gamma_{2\mathrm{k}}-1)p\right)}{\omega^2-\omega_{\mathbf{k}}^2},$$

where

$$\omega_{\mathbf{k}}^2 = 36J^2\alpha\left\{(1-p)^2(\gamma_{\mathbf{k}}-1)\left[\frac{c_1}{6}+c_1\gamma_{\mathbf{k}}-c_2-\frac{2(1-\alpha)}{9\alpha}\right]\right.$$

$$+p^2(\gamma_{2\mathbf{k}}-1)\left[\frac{c_{2a}}{6}+c_{2a}\gamma_{2\mathbf{k}}-c_2^{'}-\frac{2(1-\alpha)}{9\alpha}\right]$$

$$\left.-p(1-p)\left[(1-\gamma_{\mathbf{k}})(c^{''}-\gamma_{2\mathbf{k}}c_1)+(1-\gamma_{2\mathbf{k}})(c^{''}-\gamma_{\mathbf{k}}c_{2a})\right]\right\}.$$

$$c_2 = \frac{1}{6}\left(\frac{4}{3} + 2\langle s_{\mathbf{n}}^{+1} s_{\mathbf{n+d}}^{-1}\rangle + 2c_1 + c_{2a}\right),$$

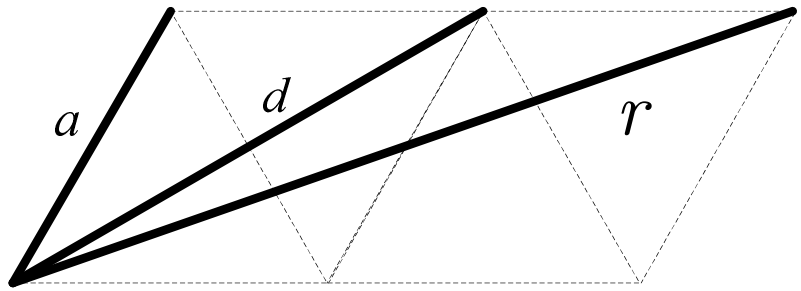
$$c'' = \frac{1}{6}\left(2\langle s_{\mathbf{n}}^{+1} s_{\mathbf{n+r}}^{-1}\rangle + 2\langle s_{\mathbf{n}}^{+1} s_{\mathbf{n+d}}^{-1}\rangle + c_1 + \langle s_{\mathbf{n}}^{+1} s_{\mathbf{n+3a}}^{-1}\rangle\right),$$

$$c_2' = \frac{1}{6}\left(\frac{4}{3} + \langle s_{\mathbf{n}}^{+1} s_{\mathbf{n+4a}}^{-1}\rangle + 2c_{2a} + 2\langle s_{\mathbf{n}}^{+1} s_{\mathbf{n+2d}}^{-1}\rangle\right),\ c_1 = \langle s_{\mathbf{n}}^{+1} s_{\mathbf{n+a}}^{-1}\rangle,\ c_{2a} = \langle s_{\mathbf{n}}^{+1} s_{\mathbf{n+2a}}^{-1}\rangle$$

are the spin correlations on neighboring sites,

$$\gamma_{\mathbf{k}} = \frac{1}{6}\sum_{\mathbf{k}} \exp[i\mathbf{ka}],$$

α 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

$$\left\langle s_{\mathbf{n}}^{+1} s_{\mathbf{m}}^{-1} \right\rangle = \frac{6J}{N} \sum_{\mathbf{k}} \exp\left[i\mathbf{k}\left(\mathbf{n}-\mathbf{m}\right) \right] \frac{\left(c_1(\gamma_{\mathbf{k}} - 1)(p-1) + c_{2a}(\gamma_{2\mathbf{k}} - 1)p \right)}{\omega_{\mathbf{k}}} \coth\left(\frac{\omega_{\mathbf{k}}}{2T} \right)$$

and the equation $\left\langle s_{\mathbf{n}}^{+} s_{\mathbf{n}}^{-} \right\rangle = \frac{4}{3}$, which follows from the constraint $\left\langle s_{\mathbf{n}}^{2} \right\rangle = 2$

LRO or SRO ? One has to analyze $\left\langle s_{\mathbf{n}}^{+1} s_{\mathbf{m}}^{-1} \right\rangle$ at **T=0**

$$\left\langle s_{\mathbf{n}}^{+1} s_{\mathbf{m}}^{-1} \right\rangle = \frac{1}{N} \sum_{\mathbf{k} \neq \mathbf{Q}} \exp\left[i\mathbf{k}\left(\mathbf{n}-\mathbf{m}\right) \right] c_{o\mathbf{k}} + \frac{1}{N} \sum_{\mathbf{Q}} \exp\left[i\mathbf{Q}\left(\mathbf{n}-\mathbf{m}\right) \right] c_{\mathbf{Q}}.$$

The 1st term describes local correlations, this term vanishes for large $\left|\mathbf{n}-\mathbf{m}\right|$.
The 2nd term - the condensation part – is connected to the LRO.

LRO: Ferromagnetic order ($|J_1| >> J_3$), Q=Γ=[0,0],

Near the Γ point the serial expansion of $\omega_{\mathbf{k}}^2$ starts from the terms proportional to k^4.

$$\left\langle s_{\mathbf{n}}^{+1} s_{\mathbf{m}}^{-1} \right\rangle = \frac{1}{N} \sum_{\mathbf{k} \neq \Gamma} \exp\left[i\mathbf{k}\left(\mathbf{n}-\mathbf{m}\right)\right] c_{o\mathbf{k}} + C_F .$$

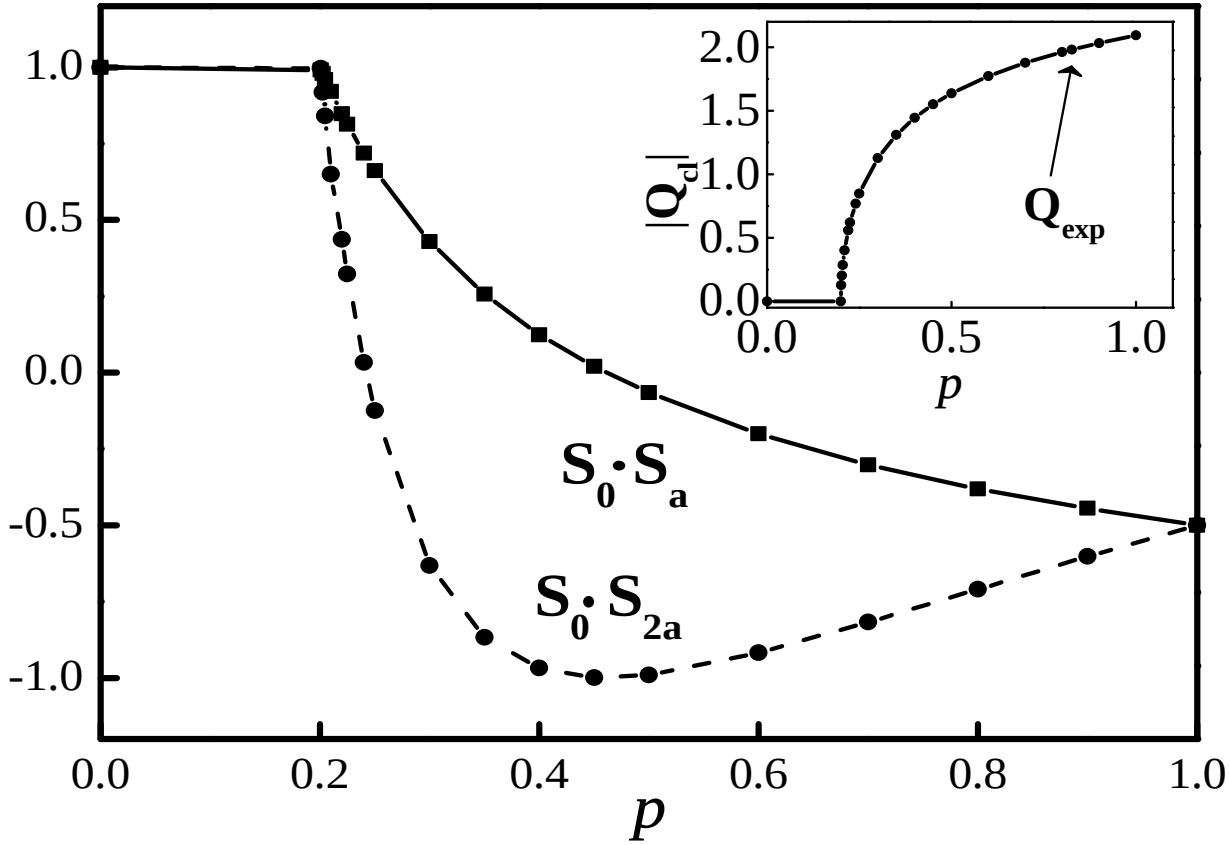
Antiferromagnetic order:

$\omega_{\mathbf{k}}$ is vanishing at the nonzero ordering vector $\mathbf{Q_i}$.

$$\left\langle s_{\mathbf{n}}^{+1} s_{\mathbf{m}}^{-1} \right\rangle = \frac{1}{N} \sum_{\mathbf{k} \neq \mathbf{Q}} \exp\left[i\mathbf{k}\left(\mathbf{n}-\mathbf{m}\right)\right] c_{o\mathbf{k}} + \sum_{i} \cos\left[\mathbf{Q}_i \left(\mathbf{n}-\mathbf{m}\right)\right] C_{AF} .$$

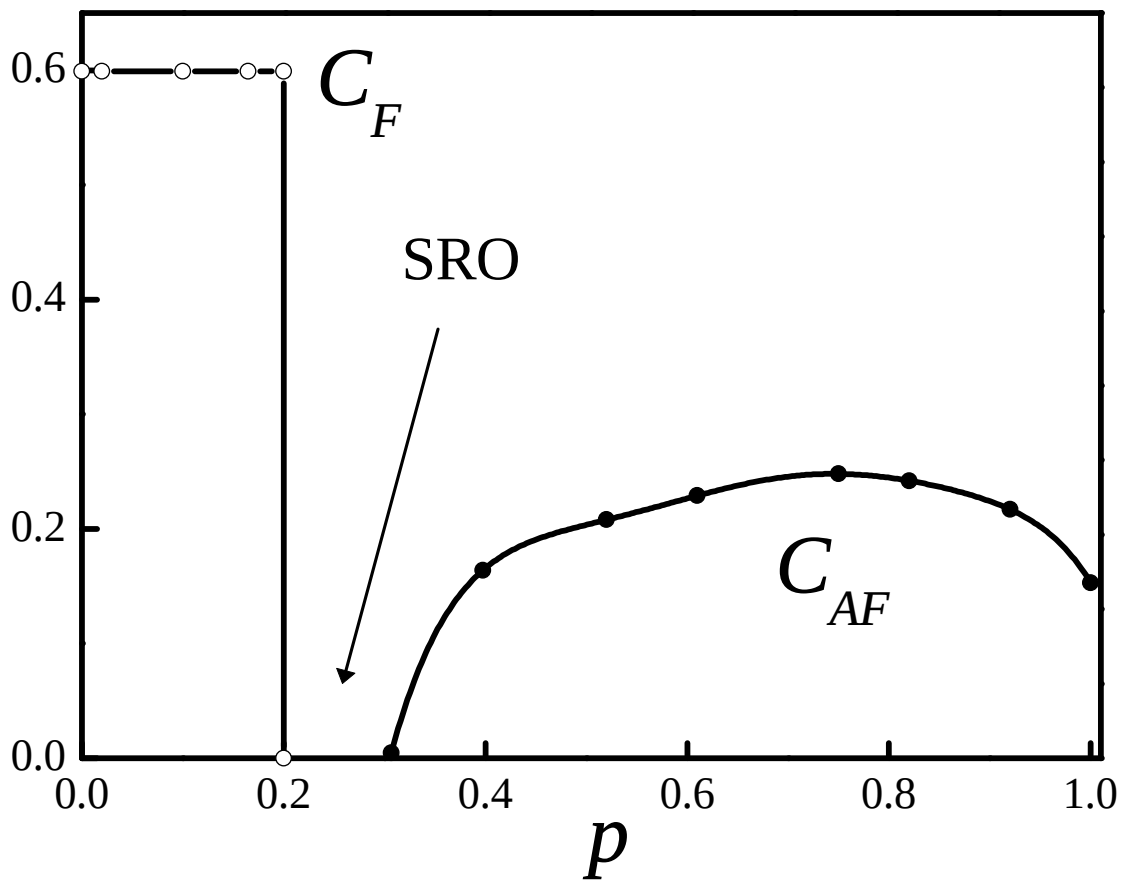
SRO: CAF =0, CF=0

Classical model : the AF ordering vector Q corresponds to a minimum of $J(\mathbf{k}) = \sum_{i,j} J_{ij} \cos(\mathbf{k}(\mathbf{R}_i - \mathbf{R}_j)), \mathbf{S}_i = \mathbf{u}\cos(\mathbf{Q}\mathbf{R}_i) + \mathbf{v}\sin(\mathbf{Q}\mathbf{R}_i)$.



The dependencies of the scalar products between the nearest ($\mathbf{S_0}\cdot\mathbf{S_a}$) and third nearest neighbor ($\mathbf{S_0}\cdot\mathbf{S_{2a}}$) classical spins on the frustration parameter *p*. The inset shows the dependence of the length of the ordering vector Q_{cl} on *p*.

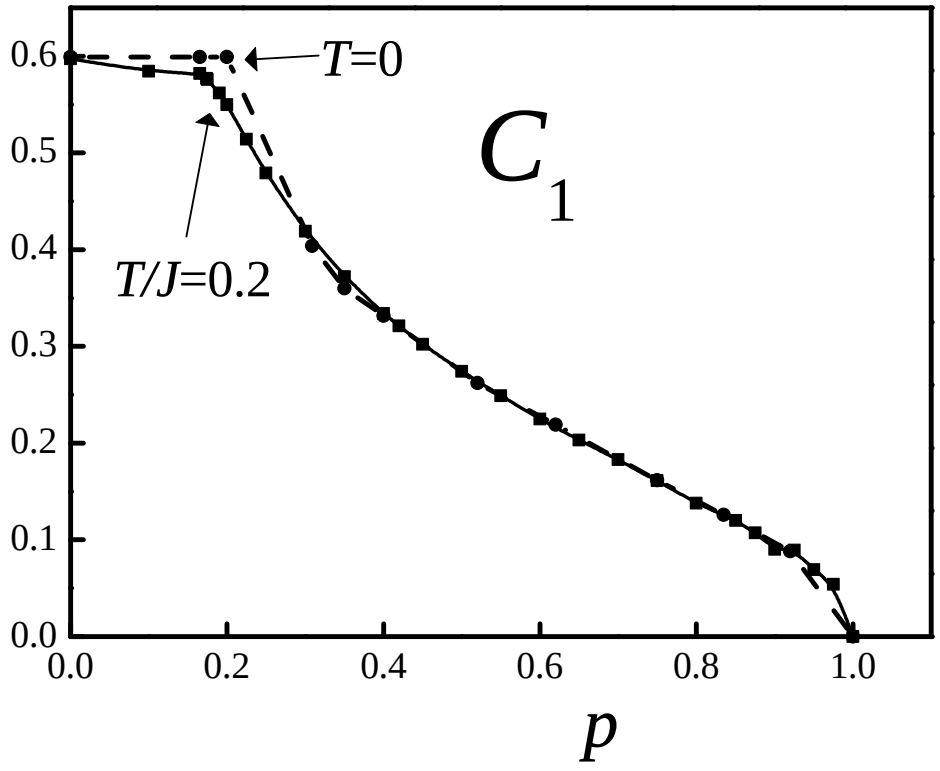
Quantum model. Mori's approach. T=0

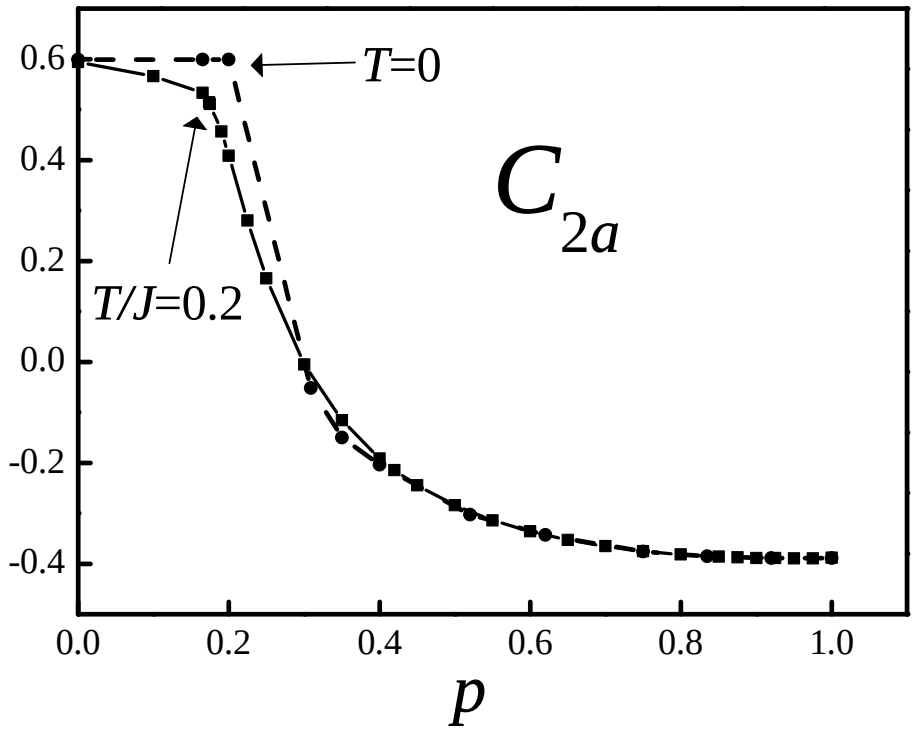


The dependencies of the ferromagnetic condensation part C_F and the antiferromagnetic condensation part C_{AF} on the frustration parameter *p*.

CORRELATION FUNCTIONS

$$c_1 = \langle s_{\mathbf{n}}^{+1} s_{\mathbf{n+a}}^{-1} \rangle$$

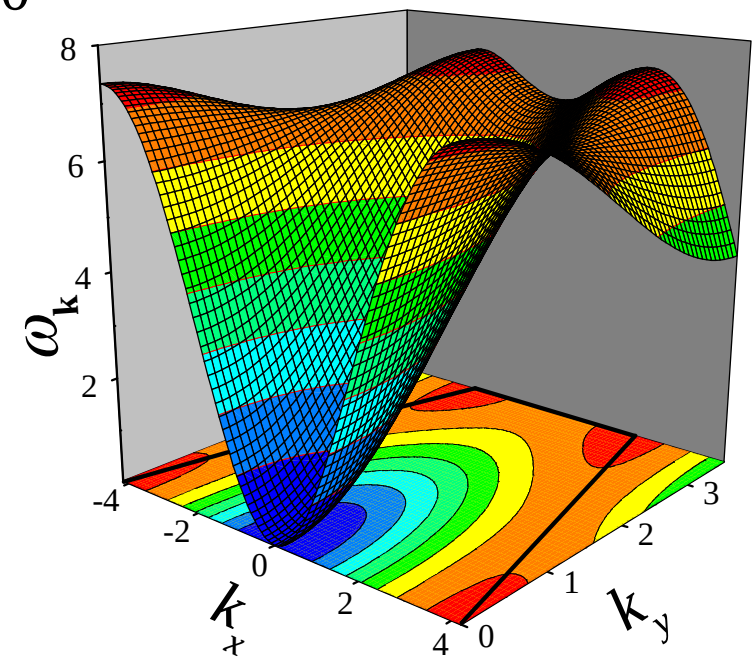




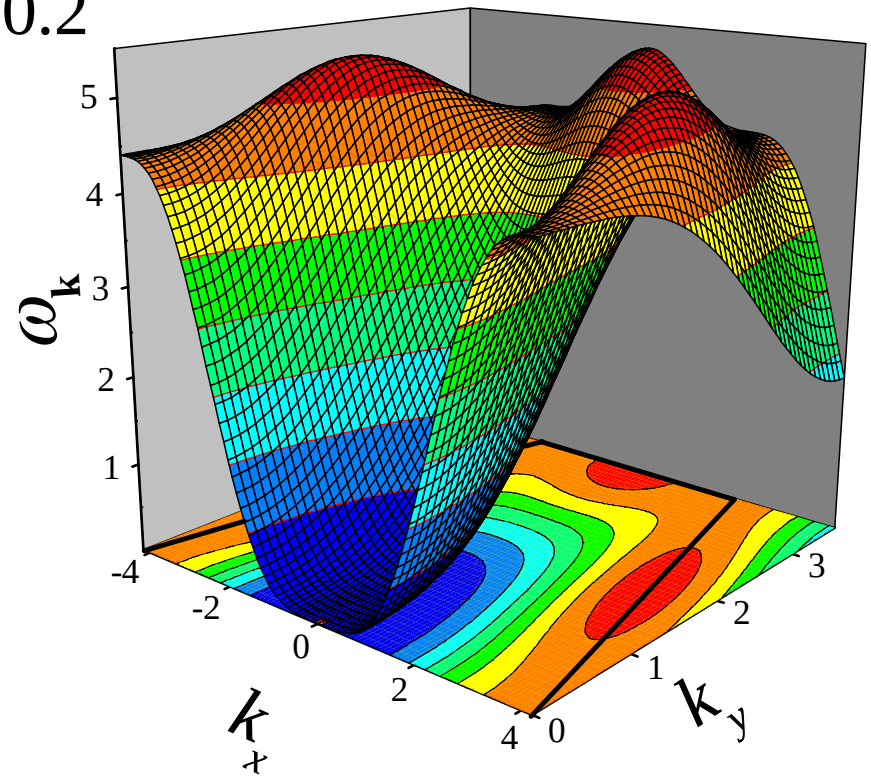
$$c_{2a} = \langle s_{\mathbf{n}}^{+1} s_{\mathbf{n+2a}}^{-1} \rangle$$

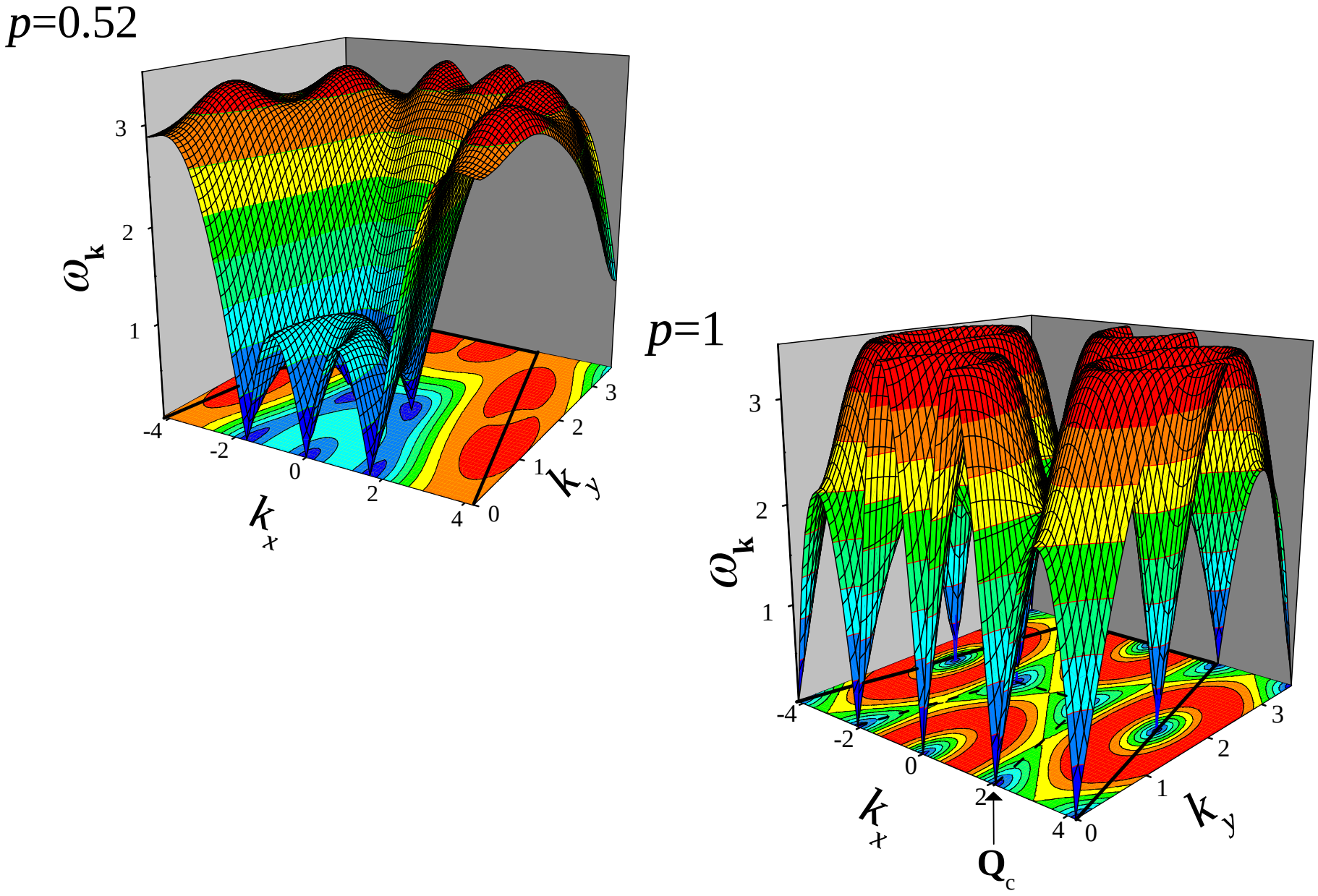
Frequency of spin excitations

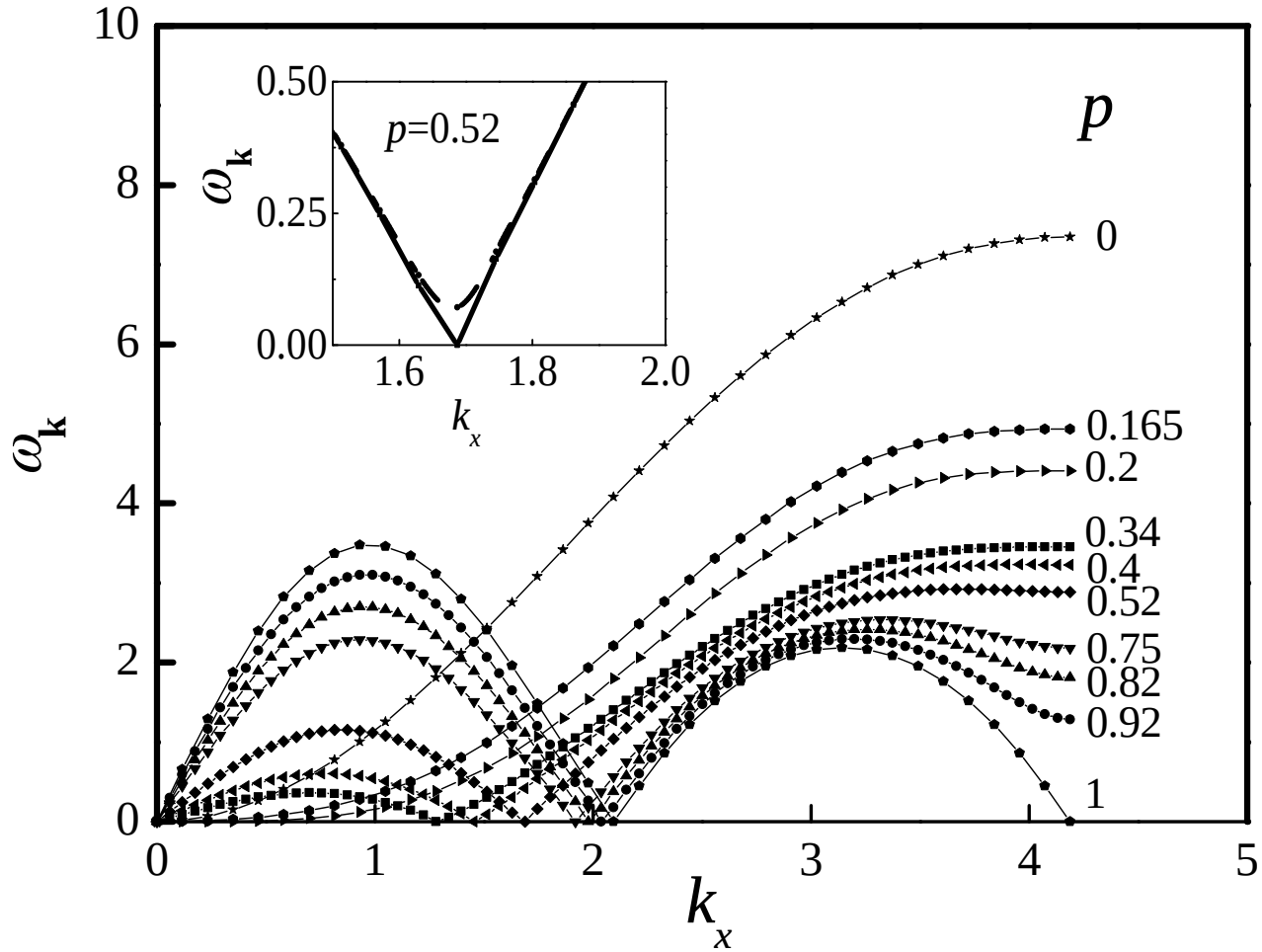
p=0



p=0.2




p=0.52
p=1
ω_k
k_x
k_y
Q_c
-4
-2
0
2
4
1
2
3



The dispersion of spin excitations ω_k for different values of the frustration parameter p at T = 0 along the x axis. The inset shows ω_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 *J1-J3* 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° spin structure on each of them. The results are applicable to $NiGa_2S_4$.

P.Rubin, A. Sherman, M. Schreiber, Physics Letters A **376** (2012), 1062