Gyrate quantum states in frustrated magnetism: continuous transitions on the

\( J_1 - J_2 - J_3 \) globe

V. E. Valiulin,1,2,3 A. V. Mikheyenkov,1,2,3 N. M. Cluchekkatchev,1,2,4,5 and A. F. Barabanov1

1 Institute for High Pressure Physics, Russian Academy of Sciences, Moscow (Troitsk) 108840, Russia
2 Department of Theoretical Physics, Moscow Institute of Physics and Technology (State University), Moscow 141700, Russia
3 National Research Centre “Kurchatov Institute”, Moscow 123182, Russia
4 L.D. Landau Institute for Theoretical Physics, Russian Academy of Sciences, Moscow 119334, Russia
5 Institute of Metallurgy, Ural Branch, Russian Academy of Sciences, Ekaterinburg 620016, Russia

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Frustrated magnetic compounds, in particular, low-dimensional, are topical research due to persistent uncover of novel nontrivial quantum states and potential applications. The problem of this field is that many important results are scattered over the localized islands of parameters, while nebular areas in between still contain hidden new physics. We have found new local order in spin liquids: antiferromagnetic isotropical helices. On the structure factor we see gyrate concentric dispersionless structures, while on any radial direction the excitation spectrum has “roton” minima. That implies nontrivial magnetic excitations and consequences in magnetic susceptibility and thermodynamics. On the \( J_1 - J_2 - J_3 \) exchange globe we discover a continuous pass from antiferromagnetic-like local order to ferromagnetic-like; we find stripe-like order in the middle of this pass. In fact, our “quasielastic” approach allows investigation of the whole \( J_1 - J_2 - J_3 \) globe.

I. INTRODUCTION

One of the key topical questions is how strong frustration in magnetic systems coexist with ordering [1–4]. Intense research today addresses systems with multiple frustrating agents. The problems is how the number of frustrating agents and the relations between them affect the order and the structure of disordered state. The theoretical activity in the field is continuously fed by regular experimental achievements. New possibilities to construct and control quantum states of matter emerge this way including transport of skyrmions and antiskyrmions [5–7], chiral spin liquids with robust edge modes [8–9], nontrivial quasiparticles like semions [10].

Frustration agents in magnetic systems have different nature, including magnetoelastic coupling [11], spin-orbital interaction [12–15], geometrical constrains [16–18], doping, competing interactions (both exchange [3, 4, 19, 20] and long-range order — Dzyaloshinskii-Moriya [21] and dipole-dipole [22] ones).

There is a wide class of magnetically frustrated systems that can be well enough described as a set of weakly interacting magnetic planes with strong multi-exchange Heisenberg interaction within the plane. This concept is during decades widely used for spin system of HTSC cuprates [23, 24] and for long known other layered compounds [25, 29]. Later several other layered (quasi-two-dimensional) \( J_1 - J_2 \) compounds were discovered covering great variety of relationships between first and second exchange parameters. In particular, these are \( \text{Pb}_2\text{VO}_4\text{PO}_4\) [30–33], \( \text{(CuCl)}\text{LaNb}_2\text{O}_5\) [29], \( \text{SrZnVO}_4\text{PO}_4\) [33–36], \( \text{BaCdVO}_4\text{PO}_4\) [32, 31, 37], \( \text{K}_2\text{CuF}_4\), \( \text{Cs}_2\text{CuF}_4\), \( \text{Cs}_2\text{AgF}_4\), \( \text{La}_2\text{BaCuO}_5\), \( \text{Rb}_2\text{CrCl}_4\), [32, 45, 88, 42] and others.

Today multi-exchange, in particular \( J_1-J_2-J_3 \) strongly frustrated low-dimensional Heisenberg systems are in the centre of attraction due to the progress of material science, development of new theoretical tools and new physics emerging from competition of \( J \)-frustrating agents [3, 4, 16, 19, 22, 43, 47]. The problem of this field is that many important results are scattered over the localized islands of parameters, while nebular areas in between still contain hidden new effects. We suggest the approach [49, 52] that gives an opportunity to uncover “white spots” on \( J_1 - J_2 - J_3 \)-“globe”.

We have found new local order in spin liquids: antiferromagnetic isotropical helices. On the structure factor we see gyrate concentric dispersionless structures, while on any radial direction the excitation spectrum has “roton” minima. That implies nontrivial magnetic excitations and consequences in magnetic susceptibility and thermodynamics. On the \( J_1 - J_2 - J_3 \) exchange globe we discover a continuous pass from antiferromagnetic-like (AFM) local order to ferromagnetic-like (FM); we find stripe-like order in the middle of this pass.

Let’s turn for a moment to the classical limit of the problem. Fig. 1 shows the phase portrait of the system in hand. Even this simple case demonstrates the variety of spin structures. In the present work we focus on the quantum case. The most interesting fragment of spin correlation portrait in the quantum case is shown in Fig. 3.

By now only the domain \( J_2 > 0, J_3 = 0 \) (that is, half of the globe equator) can be considered as deeply investigated, see, e.g., [1, 12, 51, 52] and Refs. therein. Briefly, the generally accepted picture is the following. At \( T = 0 \) for \( J_1 > 0 \) there are two phase transitions in the system: from AFM long-range order to spin liquid and then to stripe-like long-range order. For \( J_1 < 0 \) there is a sequence of transitions: stripe – spin liquid – FM order [54, 56, 61]. At nonzero temperature the same applies to the short-range order structure.

Still there is no full clarity on the nature of successive quantum phase transitions, fine details of the disordered
state, influence of finite temperature (at least in quasi-
two-dimensional case) and nonzero $J_3$.

The “quasielastic” approach adopted here allows to re-
solve or dampen the mentioned problems. In particular,
it is possible to investigate the whole $J_1 - J_2 - J_3$ globe.
We can find out spin-spin Green’s and correlation func-
tions, structure factor, correlation length, spin suscep-
tibility and heat capacity in the wide temperature and
exchange parameters range.

II. MULTI-EXCHANGE HEISENBERG
SYSTEM: FROM SIMPLE FRUSTRATION TO
QUANTUM HELICES

A. Model Hamiltonian

We address two-dimensional $J_1 - J_2 - J_3$ Heisenberg
model with spin $S = 1/2$ on the square lattice, see Fig. 2.

The Hamiltonian of the model reads

$$H = J_1 \sum_{\langle i,j \rangle} \hat{S}_i \hat{S}_j + J_2 \sum_{[i,j]} \hat{S}_i \hat{S}_j + J_3 \sum_{\{i,j\}} \hat{S}_i \hat{S}_j$$  \hspace{1cm} (1)

where $(\hat{S}_i)^2 = 3/4$, $(i,j)$ denotes NN (nearest neighbor) bonds, $[i,j]$ denotes NNN (next-nearest neighbor) bonds and $\{i,j\}$ denotes NNNN (next-to-next-nearest neighbor) bonds of the square lattice sites $i, j$.

Expression (1) provides the minimal possible model, since quantum (and classical in the limit $S \to \infty$) hel-
ciles appear starting from “$J_3$”-level of multi-exchange
Heisenberg Hamiltonian. In other words, $J_1 - J_2$ yet
does not lead to helical state.

We first briefly remind the classical limit of the prob-
lem. For classical spins in 2D any order, commensurate or
incommensurate, can be set by the simple ansatz (plane
spiral) \[S_r = e_1 \cos(q_0 r) + e_2 \sin(q_0 r), \hspace{1cm} (2)\]

where $e_1$ and $e_2$ are in-plane orthogonal orths. For fixed
values of exchanges $J_1, J_2, J_3$ the spin structure is deter-
ned by the energy minimization with respect to control
point $q_0$ position.

First of all this means that only long-range order
(LRO) is realised in the classical limit, no short-range or-
der (SRO), that is no spin liquid. Apparently (2) means
$\delta$-like spin-spin correlation functions.

In the quantum case under consideration ($S = 1/2$),
we underline, average site spin is zero

$$\langle S_r \rangle = 0,$$  \hspace{1cm} (3)

and the spin order is defined by the structure factor which
usually is a complicated continuous function of momen-
tum $q$ in the Brillouin zone with more or less pronounced
maximum.

B. The method

We use the so called spherically symmetric self-
consistent approach for spin-spin Green’s functions
(SSSA) \[12, 48–52\].

FIG. 1. (Color online) A sketch of the $J_1 - J_2 - J_3$-model
phase diagram in the classical limit. The labels mark the
positions of structure factor $\delta$-peak, see text and Eqs. (2),
(3). Top: “Globe” representation of the phase diagram when
$J_1 = \cos(\psi) \cos(\phi), J_2 = \cos(\psi) \sin(\phi), J_3 = \sin(\psi)$, see
Sec. II C. Bottom: “Flat” representation of the phase dia-
gram. The phases are: $(0,0)$ — ferromagnetic (FM), $(\pi, \pi)$ —
antiferromagnetic (AFM), $(\pi, 0)$ — stripe, while $(\pi, q), (q, 0)$
and $(q, q)$ are three different incommensurate helical phases.
Note, that FM and AFM phases are not seen on the visible
side of the globe.

FIG. 2. (Color online) The sketch of the square lattice and
three exchange bonds.
The long-range order is zero at any temperature, see Eq. (3)). In our case it means in particular that average site spin and allows:

$$G(q, \omega, T) = \frac{F_q}{\omega^2 - \omega_q^2}, \quad (5)$$

acquires the form

where $$q_0 = 0, r_i (i = 1 \div 8)$$ belongs to $$i$$-th coordination spheres, the structure factor

$$c_q = \langle S^z_q S^z_{-q} \rangle = -\frac{1}{\pi} \int_0^{\infty} d\omega \coth \left( \frac{\omega}{2T} \right) \text{Im} G(q, \omega, T). \quad (7)$$

The system of self-consistent equations (5)–(7) is analyzed numerically. Hereafter all the energy-related parameters are set in the units of $$J = \sqrt{J_1^2 + J_2^2 + J_3^2}$$. All the foregoing results have been obtained at low temperature $$T = 0.02$$.

C. Results and discussion

In the classical limit the structure factor is always $$\delta$$-function like (see Eq. (2)). This means that there is only one unique wave vector $$q$$ defining spin order (apart from symmetry equivalent points in the Brillouin zone).

In the quantum case $$S = 1/2$$ the structure factor is usually a smooth complicated continuous function of momentum $$q$$. Nevertheless at not very high temperatures local spin order can be distinguished by the positions of the structure factor maxima (see Fig. 3).

The most interesting situation corresponds to continuous degeneracy of the structure factor maxima: in this case they merge into the curve in the $$q$$-space (this is hardly possible in the classical limit). Sometimes this curve is topologically equivalent to circle, then we can discuss the “gyrate” quantum states.

We underline, that the last picture is natural only for strongly frustrated model. For example such continuous degeneracy does not appear in $$J_1 - J_2$$ square lattice model: the third frustrating agent $$J_3$$ is necessary.

1. Phase diagram: general properties

In $$J_1 - J_2 - J_3$$ model the norm $$\sqrt{J_1^2 + J_2^2 + J_3^2}$$ is irrelevant for short-range order and the phase diagram.
FIG. 4. (Color online) Contour lines for excitation spectra $\omega(q)$ (upper raw) and the structure factor $c_q$ (lower raw). Exchanges $J_1$, $J_2$ and $J_3$ are parameterized by spherical angles $\phi$ and $\psi$ (in degrees): $J_1 = \cos(\psi) \cos(\phi)$, $J_2 = \cos(\psi) \sin(\phi)$, $J_3 = \sin(\psi)$. Here $\psi = 10^\circ$ and $\phi = 0^\circ \div 20^\circ$. On the first column $\omega(q)$ minimum and $c_q$ maximum at AFM point $(\pi, \pi)$ indicate AFM short-range order. With growing $\phi$ AFM gap is opening and gyrate $c_q$ structure is developed, acquiring then square features.

So the kind of “globe” parametrisation is convenient

$$J_1 = \cos(\psi) \cos(\phi),$$
$$J_2 = \cos(\psi) \sin(\phi),$$
$$J_3 = \sin(\psi).$$

Here $\psi = \pi/2 - \theta$, and $\theta$ is the standard spherical angle. This choice improves the observables readability.

Like on the earth globe, there is a “no man’s land” at the “poles” ($\psi = \pm \pi/2$, that is $J_1 = J_2 = 0, J_3 = \pm 1$), where there is almost nothing interesting and experimentally relevant on the phase diagram. The most intriguing are the “equatorial” latitudes of the “north” hemisphere, $0 \leq \phi \leq 2\pi, -\pi/2 \leq \psi \leq \pi/2$. One can see, that this region, depicted in Fig. 3, is the most frustrated.

We choose the trajectory on the phase diagram, see thick blue arrow line ($J_3 = 0.17$) in Fig. 3 that passes the following states:

- AFM with structure factor maxima at $q_0 = (\pm \pi, \pm \pi)$;
- stripe $- q_0 = (\pm \pi, 0), (0, \pm \pi)$, in the classical limit it would be alternating stripes along a lattice with spins up and down;
- FM $q_0 = (0, 0)$;
- helicoid $q_0 = (\pm q, 0), (0, \pm q)$;
- helicoid $q_0 = (\pm \pi, q), (q, \pm \pi)$;
- helicoid $q_0 = (\pm q, \pm q)$.

The last three in the classical limit would be spin helices rotating along one of the axis or along the diagonal of the square lattice. In Fig. 3 and hereafter we label the local orders with one of the equivalent points $q_0$.

Below evolution of the structure factor and the spin excitations spectra along the trajectory is investigated.

The situation in the “depth” of each phase is more or less clear, at least qualitatively. But the transitions between definite spin-liquid local orders is much more intriguing. Note, that the physical picture here is some sense similar to liquid-liquid transitions [68–73].

We are to remind some general properties of the spectrum [49–52]. The spin gap is always closed at trivial point $q_0 = (0, 0)$ at any temperature. At $T = 0$ it might be closed at nontrivial points in the Brilloin zone with $\delta$-peak of structure factor at the same point. These means the corresponding long-range order (AFM, FM, stripe or helical). At $T = 0$ spin-liquid states are also possible.

We are interested in the case of $T > 0$, when the long-range order is always absent, but the short-range order remains pronounced and complicated. The local order is defined by the structure factor maximum and the spectrum minimum at nontrivial points.

FIG. 5. (Color online) The a) “volcanic” and b) “spider” figures show structure factor $c_q$ and the spin excitations spectrum $\omega_q$. Here $\psi = 10^\circ$, $\phi = 20^\circ$ that correspond to the region of AFM gyrate states.
FIG. 6. (Color online) The same as in Fig. 4 (contour lines for $\omega(q)$ and $c_q$), for $\psi = 10^\circ$ but $\phi = 25^\circ \div 50^\circ$. Here local order is evolving from complex $(\pi, q)$ helix with $c_q$ maxima forming the modulated square line to stipe order with $c_q$ maximum at $(\pi, 0)$, see also Fig. 7.

2. From AFM via two helices to stripe

a. From $(\pi, \pi)$ via $(q, q)$ to $(\pi, q)$. The spectrum and structure factor evolution in this domain is shown in Fig. 4. We have chosen the frame of reference for the Brillouin zone ($0 \leq q \leq 2\pi$). In this case the AFM maxima are located in the centre of the Brillouin zone.

The first figure-column in Fig. 4 just corresponds to AFM with sharp maximum of the structure factor $c_q$ and local minimum of the spin excitations spectrum $\omega_q$ at the AFM point $(\pi, \pi)$. For large enough $\phi$ ($\phi \gg 40^\circ$) the short-range order becomes clearly stripe-like (see Fig. 6) with $c_q$ maximum and $\omega_q$ minimum at the stripe point $(\pi, 0)$ and the equivalent ones. The half-width of the mentioned maxima in these limits defines the correlation length correspondingly for AFM and stripe order.

In between these limits $c_q$ evolves smoothly and its peak becomes much wider implying the correlation length’s diminishing, see second figure-column in Fig. 4.

At higher $\phi$ (that is $J_2$) the top of the $c_q$ peak starts collapsing down and the peak acquires “volcanic” shape, see the evolution between second and fourth figure-columns in Fig. 4.

The form of the structure factor defines the symmetry and the structure of the underlying quantum state. Thus we get the desired “gyrate” quantum states, with the $c_q$ maxima forming the circle structure centered at $(\pi, \pi)$ see Fig. 5. This indicates local order of the antiferomagnetic isotropical helix. The continuous gyrate degeneracy can be treated as the quantum superposition of incommensurate spiral states propagating in all directions.

The $c_q$ in Fig. 5 with the volcanic shape being imaginatively squeezed to the point $(\pi, \pi)$ acquires purely AFM local order. The nonzero diameter of the $c_q$ crater is the incommensurability parameter for the degenerate set of helices and the width of the walls of the crater defines the correlation length.

b. From $(\pi, q)$ to $(\pi, 0)$. The spectrum and structure factor evolution in this domain is shown in Fig. 6. With the growth of $\phi$ (that is $J_2$) gyrate “volcanic” structure of $c_q$ acquires four-fold modulation that finally transforms into four distinct peaks. The last is the quan-
FIG. 8. (Color online) The same as in Fig. 4 (contour lines for $\omega(q)$ and $c_q$), for $\psi = 10^\circ$ but $\phi = 55^\circ \div 145^\circ$. The first three figure-columns represent the stipe-state with $\omega(q)$ minimum and $c_q$ maximum at point $(\pi,0)$ (and at equivalent points). We remind that the correlation length is related to the width of $c_q$ maximum. The correlation length diminishes from left to right. The last two figure-columns correspond to continuous splitting of stripe $c_q$ maximum, that can be interpreted as the crossover to the $(q,0)$ incommensurate helical state, more exactly, to the quantum superposition of several such states. See also Fig. 9.

tum stripe state: the superposition of local stripes along perpendicular directions, see Fig. 7.

In terms of spin excitations spectrum this transformation is the shift of $\omega_q$ local minimum from incommensurate point $(\pi,q)$ to stripe point $(\pi,0)$ with the simultaneous reduction of the corresponding spin gap, see Figs. 4-5.

Note that the spectrum $\omega_q$ in Figs. 4-5 has in some directions roton form. The same is true for Fig. 5.

3. From stripe via two helices to FM

a. From $(\pi,0)$ via $(q,0)$ towards $(q,q)$. The spectrum and structure factor evolution in this domain is shown in Fig. 6-9. We remind that the frame of reference for the Brillouin zone here is $0 \leq q_{x,y} \leq 2\pi$.

In the range $\phi \sim 90^\circ \pm 30^\circ$ the local order is stripe-like. The correlation length is maximal for $\phi = 90^\circ$ and decays on both sides. After leaving the stripe region ($\phi \gtrsim 120^\circ$) the peaks of $c_q$ split and the local order acquires $(q,0)$ helical structure.

Correspondingly, the excitation spectrum $\omega_q$ undergoes the splitting of local minima and transforms from “spider” to “squid” shape.

Structure factor $c_q$ maximum underdoes similar splitting, that can be interpreted as the crossover to the $(q,0)$ incommensurate helical state, more exactly, to the quantum superposition of several such states.

b. Reentrance from $(q,q)$ to $(q,0)$. The spectrum and structure factor evolution in this domain is shown in Figs. 10-12.

In contrast to the classical limit there exists the island of $(q,q)$ helical local order with the subsequent reentrance to $(q,0)$ helical local order.

The complex helix state, that in contrast to AFM gyrate, see Fig. 5, is to be labeled as FM gyrate, appears in the borderland (see the fourth figure-column in Fig. 10). Similar observation have been made recently in Ref. 2 using purely numerical tools (quantum Monte-Carlo simulation).

The correlation length shows nontrivial nonmonotonic evolution while passing from purely $(q,q)$ to purely $(q,0)$ helix. It dramatically drops in the borderland being suffi-
c) d) e) f)
Note in addition, that the correlation length of the FM gyrate state is much larger than the correlation length of the AFM gyrate state (compare Fig. 5 and Fig. 11).

III. CONCLUSIONS

To conclude, we have considered the topical case of the systems with multiple frustrating agents — $S = 1/2$ two-dimensional $J_1 - J_2 - J_3$ Heisenberg model. Many important results for this problem are scattered over the localized islands of parameters, while nebular areas in between still contain hidden new physics.

We use the spherically symmetric self-consistent approach for spin-spin Green’s functions. It conserves all the symmetries of the problem, the $SU(2)$-spin symmetry and the translational invariance and strictly holds the characteristic limitation of low-dimensionality.

Let us underline that the problem in hand is difficult for first principle numerical simulation: the frustration especially multi-agent increases the well-known “sign problem”. The method we use here allows to bypass this problem analytically with the cost of some uncertainty related to the accuracy of multi-spin Greens-function approximation. The method reproduces most of the well investigated cases.

In the considered case of low, but nonzero temperature, the spin state for any set of parameters is a singlet spin-liquid without long-range order. Nontrivial challenge of this long standing problem is to determine the local structure of the disordered state. Our consideration shows that in some parameter domains the structure acquires a gyrate form — quantum helical isotropical states. Gyrate state is a continuous quantum superposition of helical states; the manifold of helices directions fills the circle-like curve.

The token of a gyrate state is a tube-like form of $c_q$ and the circle-like manifold of spectrum $w_q$ local minima. These key features enriched with traditional $c_q$ and $w_q$ parts lead to the zoo of peculiar spectra and structure factors.

Finally, we uncovered a number of nebular areas in the phase diagram when one local order state of spin-liquid transforms into another one. The nontrivial gyrate states that we have found are located just in the borderlands.

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