# Mean-field study of the disordered ground state in the $\beta$-Mn lattice

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The type-II atoms in the $\beta$ phase of manganese form a three-dimensional network of corner-sharing triangles in a cubic symmetry. This peculiar geometry induces a very strong topological frustration that can explain the absence of magnetic ordering in the $\beta$ phase of manganese. Within a mean-field theory, we show that the classical antiferromagnetic Heisenberg model with nearest-neighbor interactions exhibits a line of zero energy modes. Thus this model does not order at any temperature on this lattice in the mean-field approximation. Longer-range interactions or slight deformations do not always remove the ground-state degeneracy, and the model will order only for peculiar sets of exchange parameters.

*Introduction.* During the last 30 years, there has been great interest in the study of frustrated models. The problem of ordering in frustrated regular lattices was initiated by Anderson, who first proposed a resonating valence bond ground state for the $S=1/2$ Heisenberg antiferromagnet on a triangular lattice.[1] Since then, the possibility of stabilizing a spin liquid (SL), quantum or classical, in two- and three-dimensional lattices has stimulated a large amount of experimental and theoretical work. These SL's are charaterized by very weak magnetic correlations, decreasing exponentially with the distance like structural correlations in real liquids. Up to now, two particular frustrated lattices, have been studied: the kagomé and the pyrochlore lattices where frustation is expected to overcome any magnetic ordering. Both lattices belong to the category of ''fully frustrated lattices'' (FFL) and have been theoretically identified to be quantum[2,3] SL and classical[4–6] SL.

A new candidate for a similar ground state is $\beta$-manganese. Nakamura *et al.*[7] have shown recently that this compound remains disordered down to the lowest temperatures (1.4 K) and exhibits strong spin fluctuations. Moreover, a weak doping with Al drives a transition to an unconventional spin-glass-like ground state. These characteristics are very similar to the behaviors of the kagomé and the pyrochlore compounds.[7–16]

The crystallographic structure of $\beta$-Mn is the cubic $A13$-type structure that contains 20 atoms in the unit cell.[17,18] In this structure, there are two inequivalent sites, usually called I ($8c$ sites) and II ($12d$ sites). Magnetic characterization of the $\beta$-Mn (Refs. 7 and 17) has shown that the magnetic properties of this system can be interpreted considering only the type II sites, the type I sites being almost nonmagnetic. Since we are interested in magnetic properties of $\beta$-Mn, only the 12 type II sites will be considered in the following. The positions of these atoms are described in the A13-type structure with one parameter: $y$. Structural refinements give different values for this parameter [$y=0.206$ (Ref. 17), $y=0.202$ (Ref. 19)], both of them defining nearest-neighbor (NN) and next-nearest-neighbor (NNN) distances very close to each other. It was proposed in Ref. 7 that these two distances become identical if the parameter $y$ has a ''perfect'' value $y_0=(9-\sqrt{33})/16\approx 0.203\,46$. In this case, the 12 magnetic Mn sites are forming a network of corner-sharing regular triangles and each Mn site belongs to three of these triangles.

In the following we shall consider first the case where the parameter $y$ is equal to $y_0$, and then we consider small deviations from the perfect value $y_0$. For $y=y_0$, the NN distance is $d_1=0.420$ (in units of the cubic cell parameter), while for $y=0.202$ ($<y_0$) proposed in Ref. 19 one gets two values for the NN distance: $d_1=0.419$ and $d_1'=0.424$ and for $y=0.206$ ($>y_0$),[17] one gets $d_1=0.414$ and $d_1'=0.422$.

We use the Heisenberg Hamiltonian:

$$H=-\sum_{\langle i,j\rangle} J_{ij}S_i\cdot S_j\,, \tag{1}$$

where $J_{ij}$ is the exchange interaction between two sites and is negative for an antiferromagnet. $S_i$ is a classical three-dimensional spin representing a moment located on a site $i$ of the lattice. Within a mean-field approximation, the ground-state energy is related to the maximum of the eigenvalues of the Fourier transform of the interactions.[20] This means that we have to calculate the matrix $J_q$ ($12\times 12$ in the present case) defined as

$$J_q^{\alpha\beta}=\frac{1}{N}\sum_{ij} J_{ij}^{\alpha\beta}e^{-iq\cdot(r_\alpha-r_\beta)}e^{-iq\cdot(R_i-R_j)}, \tag{2}$$

where $i$ and $j$ refer to crystallographic cells of the cubic lattice and $\alpha$ and $\beta$ label the different sites of the crystallographic cell ($1\leqslant\alpha,\beta\leqslant 12$). Defining $U_q$ as a unitary matrix which diagonalizes $J_q$ with eigenvalues $\lambda_q$, we have

$$\sum_\beta J_q^{\alpha\beta}U_q^{\beta\mu}=\lambda_q^\mu U_q^{\alpha\mu}\,, \tag{3}$$

 

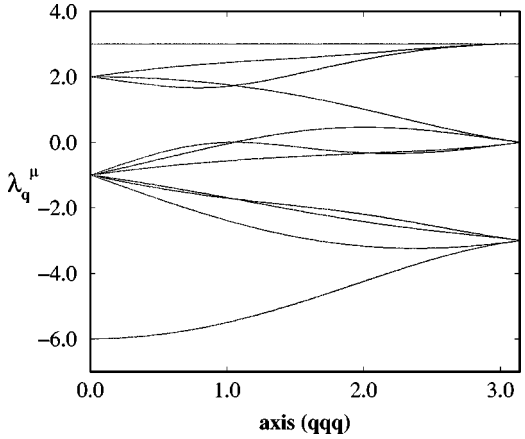


FIG. 1. Dispersion curves of the 12 eigenvalues in the ''perfect case'' along the [111] axis. The highest eigenvalue is independent of $q$ and twofold degenerate [case I(a)].

where $q$ is a vector of the first Brillouin zone and $\mu$ labels the 12 eigenvalues. Then the mean-field ground-state energy is given by the maximum of $\lambda_q^\mu$ ($\lambda_{q_0}^{\mu_0}$), through $E_{g.s.}=-\lambda_{q_0}^{\mu_0}$, and the Néel temperature $T_c$ is proportional to $\lambda_{q_0}^{\mu_0}$. Therefore in mean-field approximation the wave vector of the ordered magnetic structure is $q_0$, while the corresponding eigenvector determines the arrangement inside the crystallographic cell.[20]

However, this scheme is useful only when there is one maximum eigenvalue (or several but equivalent in reciprocal space). In fully frustrated systems, there are an infinite number of maxima indicating that the mean-field ground state is degenerate and $T_c$ does not define a Néel transition temperature.[21]

*The ''perfect'' case*. We first consider the case where $y$ is ''perfect'' and only the nearest-neighbor exchange $J(<0)$ is included. The eigenvalues of the $12\times12$ matrix defined above are shown on Fig. 1 for $q$ along the [111] axis of the Brillouin zone: the highest eigenvalue, is completely independent of $q$ for all $q$ values on this axis, and moreover, it is twofold degenerate. However, in other directions of the reciprocal space, this mode becomes dispersive. This indicates that, within a mean-field treatment, there exists an infinite number of zero energy modes and the system is able to explore all these modes without any cost in energy as in FFL. Therefore the magnetic ground state is disordered. However, the present result is different from what is observed in the kagomé and pyrochlore lattices,[21] where it was found that the largest eigenvalue is independent of $q$ over the whole Brillouin zone. In the present case the ground state is also degenerate but the degeneracy is much smaller: in FFL the number of degenerate ground states in the mean-field approximation is extensive, i.e., it is proportional to the number of sites $N$, while in the present case the number of degenerate states is only proportional to $N^{1/3}$.

This lattice is somewhat similar to the antiferromagnetic rhombohedral lattice used by Rastelli and Tassi for the description of $\beta$-oxygen:[22] in this lattice the classical spectrum contains a ''soft line'' (the [001] axis), leading to a degenerate helix and large effect of quantum and thermal excitations. Here also, the classical ground state is a degenerate helix with wave vector $(qqq)$. As a consequence, the spin wave energy spectrum in any of the degenerate helicoidal states contains a line of zero energy modes that destroy long range order at any finite temperature, but it is likely that quantum or thermal fluctuations will restore long-range order in $\beta$-Mn in the same way as it occurs in the rhombohedral antiferromagnet.

As in the kagomé and pyrochlore lattices, it is possible to make a link between the existence of soft branches and a local constraint in real space. First, we write the Hamiltonian as a sum over triangles in the lattice, and, as each spin belongs to three triangles, we obtain:

$$H=-\frac{J}{2}\sum_{triangles}(S_1+S_2+S_3)^2+\frac{3J}{2}\sum_i S_i^2. \qquad (4)$$

Thus the energies per site of Hamiltonian (4) is minimum when the sum $S_1+S_2+S_3=0$ for all triangles $(3J/2)$. The peculiarity of this lattice (as for the kagomé and the pyrochlore lattices) is that it is possible to satisfy this constraint in many ways (an infinite number) due to the low coordination of each site; in fact what is important in these systems is not the real coordination (six in the present case as in the triangular lattice), but the number of triangles at each site (three in the present case); in the following we will use this definition of the coordination since it determines the number of constraints attached to each site.

The same model on the triangular lattice can also be written in terms of triangle sums. But in this lattice, triangles are not corner sharing but edge sharing, and this does not provide enough degrees of freedom to build a large number of configurations. The consequence of the constraint in the triangular system is to order the classical model, which is exactly the opposite of what is obtained in the kagomé and the pyrochlore lattices. So the origin of these zero energy modes is the local constraint on each triangle and the possibility to build a large number of states within this constraint, due to the peculiar connectivity of the structure. However in $\beta$-Mn, each site belongs to three triangles and the lattice is more connected than the kagomé one, making this lattice intermediate between FFL and ''ordered frustrated lattices'' such as the triangular lattice.

Recently, Moessner and Chalker[5] intoduced a very general argument to determine if order by disorder will occur in a given model: they compared the number of degrees of freedom ($F$) of the system with the number of constraints ($K$) in the minimum energy state. In the present case, $F=N(n-1)$, where $N$ is the number of triangles and each spin has n components ($n=3$ for Heisenberg spins), while, if all constraints of the type $S_1+S_2+S_3=0$ are independent, one should have $K=nN$. In fact, it is not possible to impose more constraints than the number of available degrees of freedom $F$; this means that the number of independent constraints $K_i$ is at most equal to $F$; this is the main difference with the kagomé and pyrochlore lattices where all constraints are independent and, following the definition of Ref. 5, one finds that $F=K$ (kagomé) or $F>K$ (pyrochlore). In Ref. 5 it was also argued that, only when $F>K$, the ground state remains degenerate and order by disorder does not occur. To conclude, since we do not know the number of independent constraints due to the complexity of the lattice, order by

TABLE I. The different situations that have been studied: cases I(a), I(b), and I(c) correspond to the perfect case with first $[J(d_1)]$ and second $[J(d_2)]$ exchange; cases II(a), II(b), II(c), II(d), and II(e) correspond to $y \neq y_0$; there are 2 types of first-neighbor interactions at distances $d_1$ and $d_1'$ $(d_1 < d_1')$: we consider $J(d_1) = -1$, $J(d_1') \neq J(d_1)$ and all second neighbors are at the same distance $d_2$.

| Value of $y$ | Value of $J$ parameter | | | Model | Ordering wave vector |
|---|---|---|---|---|---|
| $y = y_0$ | $J(d_1)$ | | $J(d_2)$ | | |
| | -1 | | 0 | I(a) | Degenerate |
| | -1 | | $>0$ | I(b) | $q=0$ |
| | -1 | | $<0$ | I(c) | Incommensurate |
| $y \neq y_0$ | $J(d_1)$ | $J(d_1')$ | $J(d_2)$ | | |
| | -1 | $>0$ | 0 | II(a) | Incommensurate |
| | -1 | $-1-\epsilon$ | 0 | II(b) | Degenerate |
| | -1 | $-1+\epsilon$ | 0 | II(c) | Degenerate |
| | -1 | -1 | $>0$ | II(d) | $q=0$ |
| | -1 | -1 | $<0$ | II(e) | Incommensurate |

disorder is not excluded, but further studies, similar to Ref. 22, are necessary to answer this question.

*Deviations from the perfect case*. Longer-range interactions can arise for two reasons. First, $y$ is not ''perfect'' so nearest neighbors may be at slightly different distances $d_1$ and $d_1'$ defined above and exchange couplings are very sensitive to atomic displacements. Second, there can be longer range interactions, due to a type of Ruderman-Kittel-Kasuya-Yosida mechanism. In order to study the different possible cases, we investigate several models described in Table I. The perfect case with only NN exchange is called I(a) in the table, other cases describe the perfect case with second-neighbor exchange [I(b) and I(c)] and the nonperfect case either with only NN exchange [II(a), II(b), and II(c)] or with second-neighbor exchange [II(d) and II(e)]. We have investigated both cases $y > y_0$ and $y < y_0$ since there are some discrepancies in experimental results: the results for $y > y_0$ are similar to the case $y < y_0$, i.e., they depend only on the sign of the interactions. Thus in Table I, we made no distinction between the cases $y > y_0$ and $y < y_0$. Model II(a) describes the case where the structural displacement implies ferromagnetic coupling; this would be the case if exchange is extremely sensitive to distance. Models II(b,c) correspond to a slight change in the coupling (which remains antiferromagnetic), between nearest neighbors; this occurs if the interaction is not very sensitive to small variations of the distance. Finally, models II(d,e) and I(b,c) are general cases that describe the effect of next nearest neighbor exchange. By studying the eigenvalues of the matrix in each of these cases, we find that the highest eigenvalue is always along the [111] axis, and we obtain three types of ground states:

(1) The first one concerns the models where the degeneracy along the [111] axis is not lifted: I(a) and II(b,c) (see Fig. 1). Within this set, mean-field theory predicts critical modes with zero stiffness in one direction of the reciprocal space. Thus, no long range order is expected but order by disorder might occur, as discussed above.

(2) In the second set of models the degeneracy is lifted (Fig. 2), and one (or several equivalent) wave vector is selected: II(a,e) and I(c). In these cases a usual long-range order occurs. When varying the exchange coupling, the selected wave vector moves continuously on the [111] axis and is in general incommensurate. We have verified that the related eigenmodes correspond to an antiferromagnetic phase where the sum of the 12 spins of the unit cell vanishes.

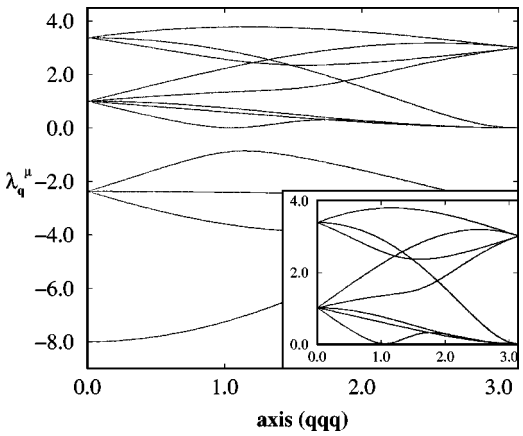


FIG. 2. The highest eigenvalues along the [111] axis for the case $J(d_1) = -1$, $J(d_2) = -1$ [case I(c)]. The inset shows the upper part of the spectrum; the maximum occurs at an incommensurate $q$ value.

(3) For the last set of models the selected propagation vector is $q=0$: I(b) and II(d) (Fig. 3). Note that selection of this vector does not mean a ferromagnetic phase (as it is in Bravais lattices) because the inner structure of the unit cell is governed by the eigenmode. It simply means that the magnetic unit cell is the crystallographic unit cell. Moreover, for these cases, the $q=0$ mode is still doubly degenerate as can be seen on Fig. 3 and this may result in a complex mixing of different eigenmodes.

*Conclusion*. We have derived some properties of the antiferromagnetic Heisenberg Hamiltonian on the $\beta$-manganese lattice. Within a mean field theory, we have shown that there is no long-range order at any temperature if we only consider nearest neighbor interactions. This property is due to the peculiar geometry and especially, to the coincidence of a local constraint and a weak connectivity. Even when deviations from the perfect case are included, the sys-

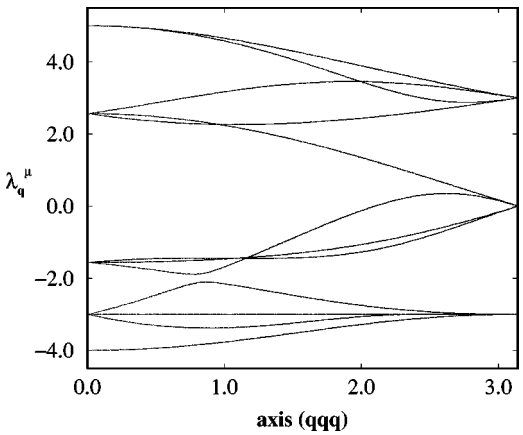


FIG. 3. The highest eigenvalues along the [111] axis for the case $J(d_1) = -1$, $J(d_2) = +1$ [case I(b)]. The maximum occurs at $q = 0$.

tem remains disordered and only longer-range interactions remove the degeneracy as in the kagomé and pyrochlore lattices. As we mentioned, the possibility of order by disorder due to thermal or quantum fluctuations cannot be excluded.

Recent experiments (neutrons and muon spin resonance) performed on $\beta$-Mn and $\beta$-Mn-Al alloys show that this system is very similar to $YMn_2$.[23–25] From a microscopic point of view, both lattices are strongly frustrated, which can explain the existence of a SL ground state at $T=0$ K. However, in both cases, Mn exhibits itinerant magnetism and this should be taken into account; the large specific heat $\gamma$ value observed in $YMn_2$ ($\gamma=180$ mJ K$^{-2}$ mol$^{-1}$) as in $\beta$-Mn ($\gamma=70$ mJ K$^{-2}$ mol$^{-1}$) should be explained in a model where both frustration and itinerancy are taken into account as proposed in Ref. 26. In fact it should be noticed that two ''3*d* heavy fermion'' compounds have been identified, namely $Y(Sc)Mn_2$ and $LiV_2O_4$;[27] in both cases, the 3*d* atoms are placed on the same frustrated lattice (pyrochlorelike). We propose that $\beta$-Mn belongs to the same class of materials, i.e., metallic systems where the absence of ordering is due to strong frustration; in this picture the large $\gamma$ value is then related to the large number of low-energy states.

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