The classical Generalized Constant Coupling method for Geometrically Frustrated Magnets revisited: Microscopic formulation and effect of perturbations beyond nearest neighbor interactions.

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A microscopic derivation of the classical Generalized Constant Coupling (GCC) model for geometrically frustrated magnets is presented. Starting from the classical Heisenberg Hamiltonian, the partition function for clusters with \( p = 2, 3, 4, \ldots \) spins in the presence of the inhomogeneous symmetry breaking fields (SBF) created by spins outside the unit is calculated. The effective fields characterizing the interaction between units naturally arise as averages over the SBF. These effective fields are fixed by a self-consistency condition. In the paramagnetic regime, we recover all the results previously obtained in a more phenomenological way, which were shown to be in excellent agreement with Monte Carlo calculations for these lattices. However, this microscopic formulation allows us to also study the behavior of the system in the absence of applied magnetic field. It is found that, for antiferromagnetic interactions, the equilibrium configuration is a non-collinear configuration in which the total magnetization of the unit is zero, and the condition under which such an ordered state occurs is also obtained from the calculation. However, frustration inhibits the formation of such an state, and the system remains paramagnetic down to 0 K, if only nearest neighbor interactions are taken into account, for all the systems considered, in agreement with the now generally accepted idea. For completeness, the ferromagnetic case is also studied. Furthermore, as the present method is formulated in the real space, it is very easy to study the effect of further perturbations in the Hamiltonian by modifying the form of the SBF. We do this to include the effect of next nearest neighbor interactions (NNN) and site dilution by non-magnetic impurities for the pyrochlore lattice. It is found that NNN interactions can stabilize a non-collinear ordered state, or ferromagnetic one, depending on the relation between NN and NNN interactions, in agreement with mean field calculations, and the phase diagram is calculated. However, site dilution is not enough by itself to form such an ordered state.

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I. INTRODUCTION

The study of the magnetic ordering in geometrically frustrated antiferromagnets (GFAF) is not a new problem in Condensed Matter Physics. It was initiated in the fifties by Wannier, Houtappel, and Anderson. A further milestone was set by Villain, who first argued that the pyrochlore Heisenberg antiferromagnet remains paramagnetic down to 0 K. However, there has been a renewed interest during recent years on these lattices, mainly because, experimentally, a rich phenomenology has been found at low temperatures and novel phases have been identified, even though these systems are relatively simple crystals. In these materials, the elementary unit of the magnetic structure is the triangle, which makes it impossible to satisfy all the antiferromagnetic bonds at the same time, with the result of a macroscopically degenerate ground state. Among the lattices with the highest degree of frustration are the pyrochlore and kagome ones. In the former, the magnetic ions occupy the corners of a 3D arrangement of corner sharing tetrahedra; in the latter, the magnetic ions occupy the corners of a 2D arrangement of corner sharing triangles (see Fig. 1). To summarize a very long story, it is found that the susceptibility in materials belonging to this class exhibits a high temperature phase in which it follows the Curie–Weiss (CW) law. Below the Curie–Weiss temperature, geometrical frustration inhibits the formation of a long range ordered (LRO) state, and the system remains paramagnetic, even though there are strong correlations between units. This phase is universally present in these systems, and it is called the cooperative paramagnetic phase. Finally, at a certain temperature, \( T_f \), which depends on the particular material and, usually, is well below the CW temperature, there
appear non universal phases: some of the systems remain paramagnetic down to the lowest temperature reached experimentally, some of them exhibit non collinear ordered states, or even some of them form a spin glass state, even though the amount of disorder in the structure is very small. It is thus easy to understand the amount of theoretical attention these systems have generated.

In the simplest theoretical description of GFAF, the spins on the lattice are regarded as Heisenberg spins with only nearest neighbor (NN) interactions. In this picture, it is predicted that the non trivial degeneracy of the ground state inhibits the formation of a LRO state, and the system remains paramagnetic down to zero temperature. However, due to the presence of frustration, the NN exchange does not fix an energy scale on the problem, and any small perturbation can break the non trivial degeneracy of the ground state and lead to some kind of ordered state. Therefore, it is especially important to incorporate these possible perturbations in any model that tries to explain the low temperature phases of these systems. Examples of perturbations present in real systems are next nearest neighbor (NNN) interactions, small anisotropies, long range dipole–dipole interaction, or dilution by non magnetic impurities, to cite some.

![Kagome lattice](image1.png) ![Pyrochlore lattice](image2.png) ![Linear chain](image3.png)

Figure 1: The lattices we will consider in this work. A, B, C, . . . denote the sublattices in which the whole lattice is subdivided. All these lattices share the following property: if we consider a spin inside a cluster formed by \( p \) spins, there are \( p - 1 \) NN spins outside the cluster. Notice also that one spin belonging to one sublattice does not have NN belonging to the same sublattice.

In a series of recent papers, the present authors have generalized the well known Constant Coupling method of magnetism, to deal with geometrically frustrated lattices, in both the classical and quantum cases, in the cooperative paramagnetic region. Also, the connection between this technique and the so called Phenomenological Renormalization Group (RG) methods for Ising spins has been furnished in Ref. where it was shown that the Generalized Constant Coupling (GCC) method can be regarded as the zero-th order approximation of one of these RG techniques, for vectorial or scalar spin systems, the so
called, Mean Field Renormalization Group method, when correlations outside a cluster are neglected, and the anomalous critical exponent of the system is taken to be zero. Therefore, it is not so surprising that magnetic properties calculated within the framework of the classical GCC method were found to be in excellent agreement with Monte Carlo calculations for those same quantities, for both the kagome and pyrochlore lattices. The main idea behind this technique is very simple, and it is based in the experimental and numerical observation that correlations in GFAF are always short ranged. Therefore, in order to calculate any thermodynamic quantity, we can start by calculating the partition function of an isolated cluster with \( p \) spins and, later, add the interactions with the surrounding \( p - 1 \) spins outside the cluster (see Fig. 1) by means of an effective field which is fixed by a self-consistency condition. This, almost phenomenological, formulation of the method, allows one to easily compute thermodynamic quantities, mainly the susceptibility, in the paramagnetic region, by using a single effective field to characterize all the interactions with spins outside the cluster. However, it is not very useful to study the critical properties of these systems, except for ferromagnetic interactions, which is not as interesting as the antiferromagnetic case, due to the absence of geometrical frustration (for Heisenberg spins). The reason is very simple. It is now generally accepted that the ordered state of these GFAF, if any, is not a simple antiferromagnetic configuration, as in bipartite lattices, but a non-collinear (NC) one, in which the total spin momentum of the unit is identically zero. Obviously, we cannot characterize such an ordered state by using a single order parameter fixed by a single effective field (EF). What is even worse, even we know that we have to introduce different order parameters for each of the spins in the cluster, and different (EF) for the neighboring spins outside the cluster, it is not easy to intuitively see how these EF enter into the expressions of the order parameters. Therefore, we need to go beyond the phenomenological formulation of the GCC method introduced in the aforementioned works. This is precisely the intention of this work. We construct the GCC method from first principles, by using rigorous identities in terms of averages over finite size clusters. In this way, the EF naturally emerge as averages over the microscopic spin variables. This microscopic formulation of the GCC method not only allows us to recover all the results obtained Refs. 25 and 27 in the paramagnetic regime but, also, gives us the possible ordered configurations of the system, and the conditions for a transition to one of these states to occur. Moreover, as the GCC method is constructed in the real space, it is especially easy to include further perturbations in the system. Particularly, we study the effect of next nearest neighbor (NNN) interactions and site dilution by non-magnetic impurities.

Apart from the microscopic formulation of the GCC method itself, we present various interesting results in this work. We find the possible ordered configurations for systems formed by corner sharing units. These systems have the interesting property that the corresponding lattices are formed by units with \( p = d + 1 \) spins \( (d \) the dimensionality), and each of the spins in the unit has \( p - 1 \) NN outside the unit. Thus, for \( d = 3 \), we have the pyrochlore lattice; for \( d = 2 \), the kagome lattice, and for \( d = 1 \), the linear chain. We have studied this last case for the sake of completeness, even though this system is not frustrated. It is found that, apart from the trivial ferromagnetic configuration for ferromagnetic interactions, the ordered state would correspond to a non-collinear configuration given by the condition that the total magnetization of the unit is zero (for the special case of the linear chain this corresponds to standard antiferromagnetic order). For NN interactions only, there is no transition to such an ordered state at any finite temperature, for none of the systems considered, as it is now generally accepted. However, when NNN interactions are taken into account, there is a transition to such a NC ordered state for some ranges of the coupling parameters. We have only studied the pyrochlore lattice in this case, as we do not think the results of the present method for the critical behavior of these systems are reliable in dimension lower than 3 (actually, the results of any model based on the idea of finite size clusters), as the long wavelength fluctuations around the NC ordered state are not properly taken into account, and they are very likely to be specially important in 2 dimensions (kagome lattice). Also, the effect of site dilution is considered in the present work. It is found that, in this framework, dilution by itself is not enough to stabilize a NC ordered state. Finally, we also consider both dilution and NNN together, and the phase diagrams for different amounts of dilution are calculated.

The remaining of the paper is organized as follows: In the following Section, we introduce the systems
studied in this work, the Hamiltonian of these systems, the notation, and the self-consistency condition that fixes the EF. In Section III, we present the calculation of the order parameters for finite cluster of size 1 and \( p \) (\( p = 4, 3, \) and 2 for the pyrochlore, kagome, and linear chain, respectively). The connection with the original formulation of the GCC method is furnished in Section IV. In Section V, we study the conditions for a transition to a long range ordered state. In Section VI, we include NNN interactions and study how this additional interaction modifies the conclusions of Section V. In Section VII, we develop the formalism to deal with site dilution by non-magnetic impurities, and apply it to the present problem. In Section VIII, we consider simultaneously the effect of both NNN interactions and site dilution. Section IX is devoted to the discussion of the results and the conclusions. Finally, we have included two appendixes to clarify some of the most involved details of the evaluation of the partition function of the different clusters subject to inhomogeneous EF.

II. PRELIMINARIES

As stated above, the original formulation of the GCC method is not well suited to study the critical behavior of GFAF. Indeed, it is now generally accepted that the ground state of the pyrochlore and kagome lattices is fixed by the condition that the total spin momentum of the tetrahedral or triangular unit, respectively, is zero, though frustration inhibits the formation of such a state, and the system remains paramagnetic in the whole temperature range, if only NN interactions are present. Obviously, we cannot characterize such an ordered state with a single order parameter, as we did in Refs. 25 and 26, which restricts the applicability of the GCC method to the study of the paramagnetic region or, at most, the ferromagnetic case, which is of less interest, due to the absence of geometrical frustration. A relatively simple way of circumventing this shortcoming of the method has been pointed out in Refs. 14 and 33, and essentially consists on introducing different sublattices characterized by different order parameters. The minimum number of sublattices we need to introduce is given by the number of spins in the cluster, that is, 3 and 4 for the kagome and pyrochlore lattices, respectively (see Fig. 1). With this subdivision, spins belonging to one sublattice only interact with spins belonging to different sublattices. Therefore, the dimensionless Heisenberg Hamiltonian of a lattice formed by corner sharing clusters with \( p \) spins, and only NN interactions, in the presence of a uniform magnetic field can be put in the form

\[
\mathcal{H} = K \sum_{\alpha \neq \beta} \sum_{\langle i, j \rangle} \vec{s}_{i\alpha} \cdot \vec{s}_{j\beta} + \vec{H}_0 \cdot \sum_{\alpha, i} \vec{s}_{i\alpha},
\]

where \( \vec{s}_{i\alpha} \) are the classical Heisenberg spins of unit length. The \( \alpha \) index labels the sublattice to which the considered spin belongs (and takes the values \( \alpha = A, B, C, \ldots \)), whereas the \( i \) index labels the spins belonging to a given sublattice. \( K = \frac{J}{T} \) (\( K > 0 \) for ferromagnetic interactions and \( K < 0 \) for antiferromagnetic ones) and \( \vec{H}_0 = \frac{\vec{h}_0}{T} \), with \( \vec{h}_0 \) the applied magnetic field.

The central idea of the GCC method is to calculate the order parameter (for each sublattice) for two clusters of different sizes, \( p \) and \( p' \), replacing the effect of the spins not included in the cluster by fixed effective fields, which act as a symmetry breaking field (SBF) and, by using the self-consistency condition

\[
\vec{m}_{p'}(K) = \vec{m}_p(K),
\]

obtain an equation to determine these effective fields and, thus, the magnetization. The order parameter is calculated by making use of the Callen-Suzuki identity

\[
\vec{m}_{p\alpha} = \langle \vec{s}_{i\alpha} \rangle = \left\langle \frac{\text{Tr}_p \vec{s}_{i\alpha} e^{\mathcal{H}_p}}{\text{Tr}_p e^{\mathcal{H}_p}} \right\rangle_{\mathcal{H}},
\]

where the partial trace is taken over the set of \( p \) variables specified by the finite size cluster Hamiltonian \( \mathcal{H}_p \); \( \vec{m}_{i\alpha} \) is the corresponding order parameter, and \( \langle \ldots \rangle_{\mathcal{H}} \) indicates the usual canonical thermal average over the ensemble defined by the complete Hamiltonian \( \mathcal{H} \).
III. EVALUATION OF THE ORDER PARAMETERS IN THE PRESENCE OF THE SBF

A. 1-spin cluster order parameter

Let us consider the cluster defined by the spin 1 in sublattice \( \alpha \) (see Fig. 2). The Hamiltonian of this cluster is given by

\[
\mathcal{H}_{1\alpha} = s_{1\alpha} \left( K \sum_{\beta \neq \alpha} z \sum_i s_{i\beta} + H_0 \right) = s_{1\alpha} \cdot \xi_{1\alpha},
\]

(4)

where the subindex 1\( \alpha \) makes reference to the fact that the Hamiltonian corresponds to a cluster of 1 spin belonging to sublattice \( \alpha \), and \( \xi_{1\alpha} \) stands for the symmetry breaking field acting on the spin of the 1-spin cluster, which belongs to sublattice \( \alpha \). The sum over the \( i \) index is performed over the NN of the 1\( \alpha \) spin, with \( z \) the number of NN in each of the sublattices (\( z = 2 \) for both the kagome and pyrochlore lattices).

By using the Callen–Suzuki identity we can calculate the order parameter in sublattice \( \alpha \) for a cluster formed by 1 spin

\[
\bar{m}_{1\alpha} = \langle s_{1\alpha} \rangle = \frac{Tr_{1\alpha} s_{1\alpha} e^{\mathcal{H}_{1\alpha}}}{Tr_{1\alpha} e^{\mathcal{H}_{1\alpha}}}.
\]

(5)

As commented above, \( Tr_{1\alpha} \) represents the partial trace with respect to the variables of the spin 1\( \alpha \), and the symbol \( \langle \ldots \rangle_{\mathcal{H}} \) stands for the usual canonical thermal average taken over the ensemble defined by the complete Hamiltonian \( \mathcal{H} \). This equation can be put in the more convenient form

\[
\bar{m}_{1\alpha} = \left< \nabla \xi_{1\alpha} \ln Tr_{1\alpha} e^{\mathcal{H}_{1\alpha}} \right>_{\mathcal{H}}.
\]

(6)
For classical vectorial spins of fixed length, the trace operation is given by

\[ Z_{1\alpha}(K) = Tr_{1\alpha} e^{\hat{s}_{1\alpha} \cdot \hat{\xi}_{1\alpha}} = \int d\Omega e^{\hat{s}_{1\alpha} \cdot \hat{\xi}_{1\alpha}}, \]  

(7)

where the integration is performed over the angular degrees of freedom of the spin. This integral is trivially evaluated to give

\[ Z_{1\alpha}(K) = 2 \sinh \xi_{1\alpha} \frac{\xi_{1\alpha}}{\xi_{1\alpha}}, \]

(8)

and, taking into account the well known relation \( \nabla \hat{r} = \hat{n} \), where \( \hat{n} \) is a unitary vector in the direction of \( \hat{r} \), we can easily calculate the order parameter for the 1-spin cluster as

\[ m_{1\alpha} = \langle L(\xi_{1\alpha}) \hat{n}_{1\alpha} \rangle, \]

(9)

where \( L(x) \) is the Langevin function and \( \hat{n}_{1\alpha} \) is an unitary vector in the direction of the corresponding SBF.

Near a critical point, as we expect the SBF to be very small, we can expand the previous expression and keep only linear terms in the SBF

\[ m_{1\alpha} \simeq \frac{\langle \xi_{1\alpha} \rangle}{3}. \]

(10)

By defining the effective fields \( \vec{h}_{\alpha} = J \langle \hat{s}_{i\alpha} \rangle_{\mathcal{H}} = J \langle \hat{s}_{j\alpha} \rangle_{\mathcal{H}} (i, j \neq 1) \), we arrive to the final expression for the order parameter

\[ m_{1\alpha} = \frac{2}{3T} \sum_{\beta \neq \alpha} \vec{h}_{\beta} + \vec{h}_{0} + \frac{\vec{h}_{0}}{3T}, \]

(11)

where we have taken into account that each spin has 2 NN on each sublattice different from the considered one. For example, for the pyrochlore lattice with no applied magnetic field we have

\[ m_{1\alpha} = \frac{2}{3T} (\vec{h}_{\beta} + \vec{h}_{\gamma} + \vec{h}_{\delta}), \]

(12)

where \( \alpha, \beta, \gamma, \) and \( \delta \) can take the values \( A, B, C, \) or \( D \). For the kagome lattice

\[ m_{1\alpha} = \frac{2}{3T} (\vec{h}_{\beta} + \vec{h}_{\gamma}), \]

where \( \alpha, \beta, \) and \( \gamma \) can take the values \( A, B, \) or \( C \). For a cluster with 2 spins, we have a linear chain, for which

\[ m_{1\alpha} = \frac{2}{3T} \vec{h}_{\beta}, \]

(13)

where \( \alpha \) and \( \beta \) can take the values \( A \) or \( B \).

Another interesting point is that if we keep all the terms in the expansion of the order parameter, and make use of the decoupling approximation

\[ \langle s_{i\alpha} s_{j\beta} \cdots s_{k\gamma} \rangle_{\mathcal{H}} \simeq \langle s_{i\alpha} \rangle_{\mathcal{H}} \langle s_{j\beta} \rangle_{\mathcal{H}} \cdots \langle s_{k\gamma} \rangle_{\mathcal{H}}, \]

(14)
and further assume $\vec{h}_\alpha' = \vec{m}_{1\alpha}$, we recover the usual self consistent Curie–Weiss equation for the order parameter

$$m_{1\alpha} = L \left( 2K \sum_{\beta \neq \alpha} m_{1\beta} + \vec{H}_0 \right).$$  \hspace{1cm} (15)

However, this expression would predict a transition to an ordered state (for antiferromagnetic interactions) at a finite temperature given by $T_N = -\Theta_{CW} = \frac{2(p-1)|J|}{3}$, which it is well known to be incorrect for GFAF. That is one of the reasons why constructing the effective fields by the self-consistency condition (2) provides a much better theory for GFAF, as we will see below.

**B. $p$-spin cluster order parameter**

The calculation of the order parameter for a cluster with $p$ spins in the presence of the SBF created by the neighboring spins is a highly non-trivial problem, as we will see in the following. The size of the clusters is chosen so that we consider the smallest number of spins that include all the different sublattices. That is, $p = 3$ for a kagome lattice (a triangular cluster), $p = 4$ for a pyrochlore (a tetrahedral cluster), etc (see Fig. 3). The Hamiltonian for such a cluster can be put as

$$H_p = K \sum_{\alpha \neq \beta} \vec{s}_{1\alpha} \cdot \vec{s}_{1\beta} + \sum_\alpha \vec{s}_{1\alpha} \cdot \vec{\xi}_{p\alpha}.$$  \hspace{1cm} (16)

In this Hamiltonian, the first term represents the interaction between the spins inside the cluster of size $p$, whereas the second term represents the interaction of those same spins with the symmetry breaking fields

$$\vec{\xi}_{p\alpha} = K \sum_{\beta \neq \alpha} \sum_i z^{-1} \vec{s}_{i\beta} + \vec{H}_0,$$  \hspace{1cm} (17)

created by the $p - 1$ spins outside the cluster. The sum over $i$ goes over all the NN spins except the ones already included in the cluster.

The order parameter for each of the sublattices included in the cluster, obtained from the $p$-spin cluster can be now put as

$$\vec{m}_{p\alpha} = \left\langle \frac{Tr_{1A,1B,1C...}\vec{s}_{1\alpha} e^{H_p}}{Tr_{1A,1B,1C...} e^{H_p}} \right\rangle_{H},$$  \hspace{1cm} (18)

or, more conveniently

$$\vec{m}_{p\alpha} = \left\langle \nabla_{\vec{\xi}_{p\alpha}} \ln Tr_{1A,1B,1C...} e^{H_p} \right\rangle_{H}. $$  \hspace{1cm} (19)

In these expressions, $Tr_{1A,1B,1C...}$ represents the trace over the spin degrees of freedom in the $p$-spin cluster. The reduced trace operation for Heisenberg spins is given by

$$Z_p(K) = \int \prod_\alpha d\Omega_\alpha e^{H_p}$$  \hspace{1cm} (20)

It is impossible to find a closed expression for this partition function of the cluster in the presence of inhomogeneous SBF (the case with no SBF has been studied by Moessner and Berlinsky in Ref. [15]). However, as we will be mainly interested in the region near the critical point (if any) in the absence of applied field, where the SBF are expected to be small, and in the paramagnetic region, where the SBF will be proportional...
Figure 3: $p$-spin clusters for the lattices considered in this work. Again, we have represented the interactions with the spins creating the EF by dashed bonds. Notice that there is 1 spin outside the cluster on each sublattice different from the considered spin of the cluster.

to the applied field and, again, small, it is enough for our purposes to calculate reduced partition function up to second order in the SBF. The main steps of the calculation are presented in Appendix A, and the result is given by

$$Z_p(K) \simeq z^0_p(K) + \frac{z^0_p(K)}{6} \sum_{\alpha} \xi_{p\alpha}^2 - \frac{z^1_p(K)}{6} \sum_{\alpha \neq \beta} \xi_{p\alpha} \cdot \xi_{p\beta},$$  \hspace{1cm} (21)

for $p > 1$, where

$$z^0_p(K) = \int_0^\infty dq \, q^2 \, e^{\frac{q^2}{2K}} \left( \frac{\sin q}{q} \right)^p$$  \hspace{1cm} (22)

and

$$z^1_p(K) = \int_0^\infty dq \, q^2 \, e^{\frac{q^2}{2K}} \left( \frac{\sin q}{q} \right)^{p-2} \left( \frac{\cos q}{q} - \frac{\sin q}{q^2} \right)^2.$$  \hspace{1cm} (23)

Expressions for these functions are given in Appendix B. Incidentally, the first term in (21) corresponds to the partition function for non-interacting clusters with no applied field, which were first calculated by Moessner and Berlinsky. The additional terms in (21) allow us to study not only the paramagnetic regime, but also the behavior of the system near a critical point, as we will see below.
Once we have this reduced partition function we can calculate the order parameter up to first order in the SBF by taking into account (19)

\[ \bar{m}_{p\alpha} = \frac{\bar{\xi}_{p\alpha}}{3} - \frac{z_p^{1}(K)}{3z_p^{0}(K)} \sum_{\beta \neq \alpha} \langle \bar{\xi}_{p\beta} \rangle. \] (24)

Taking into account the definition of the SBF, and the fact that \( J \langle \vec{s}_i\rangle_H = J \langle \vec{s}_j\rangle_H = \vec{h}'_{\alpha} \), after a little algebra we arrive at

\[ \bar{m}_{p\alpha} = \frac{1}{3T} \left\{ [1 - (p - 1)a_p(K)] \bar{h}_0 + (1 - (p - 2)a_p(K)) \sum_{\beta \neq \alpha} \bar{h}'_{\beta} - (p - 1)a_p(K)\bar{h}'_{\alpha} \right\}, \] (25)

where we have introduced the function

\[ a_p(K) = \frac{z_p^{1}(K)}{z_p^{0}(K)}. \] (26)

**IV. CALCULATION OF THE SUSCEPTIBILITY: CONNECTION WITH THE ORIGINAL FORMULATION OF THE GCC METHOD**

In the following, we will find more useful to express all the thermodynamic quantities in terms of the dimensionless temperature

\[ \bar{T} = \frac{1}{|K|} = \frac{T}{|J|}. \] (27)

In terms of this parameter, we have

\[ \bar{m}_{1\alpha} = \frac{1}{3|J|T} \left( \bar{h}_0 + 2 \sum_{\beta \neq \alpha} \bar{h}'_{\beta} \right), \] (28)

and

\[ \bar{m}_{p\alpha} = \frac{1}{3|J|T} \left\{ [1 - (p - 1)A_p(\bar{T})] \bar{h}_0 + \left(1 - (p - 2)A_p(\bar{T})\right) \sum_{\beta \neq \alpha} \bar{h}'_{\beta} - (p - 1)A_p(\bar{T})\bar{h}'_{\alpha} \right\}, \] (29)

where

\[ A_p(\bar{T}) = a_p(\bar{T}) \] (30)

for ferromagnetic interactions and

\[ A_p(\bar{T}) = a_p(-\bar{T}) \] (31)

for antiferromagnetic ones. The self consistency condition (see eq. (3))

\[ \bar{m}_{1\alpha} = \bar{m}_{p\alpha} \] (32)

leads to the following system of linear equations

\[ (p - 1) A_p(\bar{T}) \bar{h}'_{\alpha} + \left[ 1 + (p - 2) A_p(\bar{T}) \right] \sum_{\beta \neq \alpha} \bar{h}'_{\beta} = -(p - 1) A_p(\bar{T}) \bar{h}_0, \] (33)
that has the solution

\[ \vec{h}'_\alpha = \vec{h}' = -\frac{A_p(\vec{T})}{1 + (p - 1)A_p(\vec{T})} \vec{h}_0. \]  

Substituting back in (28) we find

\[ \vec{m}_{p\alpha} = \frac{1}{3 |J| T} \frac{1 - (p - 1)A_p(\vec{T})}{1 + (p - 1)A_p(\vec{T})} \vec{h}_0, \]  

that is, the susceptibility is given by

\[ \chi^{gce}_p = \frac{1}{3 |J| T} \frac{1 - (p - 1)A_p(\vec{T})}{1 + (p - 1)A_p(\vec{T})}, \]  

By direct substitution, it is simple to verify the relation

\[ \varepsilon_p(\vec{T}) = -(p - 1) A_p(\vec{T}), \]  

where \( \varepsilon_p(\vec{T}) \) is defined as

\[ \varepsilon_p(\vec{T}) = \frac{2 \vec{T}^2}{p} \frac{\partial}{\partial T} \ln z_p(T) - 1 \]  

so we can finally put

\[ \chi^{gce}_p = \frac{1}{3 |J| T} \frac{1 + \varepsilon_p(\vec{T})}{1 - \varepsilon_p(\vec{T})}, \]  

in complete agreement with the result in Ref. 25.

V. CALCULATION OF THE CRITICAL POINTS

In the absence of applied field, the system of equations (33) takes the simpler form

\[ (p - 1) A_p(\vec{T}) \vec{h}'_\alpha + \left[ 1 + (p - 2)A_p(\vec{T}) \right] \sum_{\beta \neq \alpha} \vec{h}'_{\beta} = 0, \]  

and it has two different non-trivial solutions. The first one occurs if

\[ A_p(\vec{T}) = 1 \]  

or, equivalently,

\[ \varepsilon_p(\vec{T}) = -(p - 1), \]  

and corresponds to an ordered state in which

\[ \sum_\alpha \vec{m}_\alpha = 0. \]  

We will call this kind of state a non-collinear (NC) ordered state. Actually, this condition includes, as particular cases, states in which spins in different sublattices are antiferromagnetically aligned with each
other, that is, collinear states. However, the most general state described by this condition is a non-collinear one. The second solution of (40) occurs if

\[ A_p(K) = -\frac{1}{p - 1} \]  

(44)

or, equivalently,

\[ \varepsilon_p(\tilde{T}) = 1, \]  

(45)

and corresponds to an ordered state in which

\[ \vec{m}_\alpha = \vec{m}_\beta = \vec{m}_\gamma = \ldots \quad \forall \alpha, \beta, \gamma, \ldots, \]  

(46)

that is, ferromagnetic order.

Figure 4: Graphical representation of the conditions for (a) Non-collinear order and (b) Ferromagnetic order.

The previous condition for the existence at any finite temperature of a ferromagnetic ordered state is equivalent to studying if the function \( G_p(\tilde{T}) = \varepsilon_p(\tilde{T}) - 1 \) changes its sign for any finite value of \( \tilde{T} \). This function has been depicted in Fig. 4(b) for the pyrochlore lattice, kagome lattice, and linear chain. We can see that the present method correctly predicts the existence of a transition for the pyrochlore lattice for a value finite value of \( \tilde{T} \) \((K_c = 0.71)\), and no finite \( \tilde{T} \) transition for the linear chain, but incorrectly predicts a transition for the kagome lattice \((K_c = 1.33)\). This is due to the fact that the long wavelength fluctuations around the ferromagnetic state are not taken into account in the present method, and these fluctuations destroy the ordered state in 2 dimensions. On the other hand, in the linear chain case, even short range correlations, properly described in the model, are enough to destroy any form of long range order.

The condition for the existence of a non-collinear ordered state is equivalent to study if the function \( F_p(\tilde{T}) = \varepsilon_p(\tilde{T}) + (p - 1) \) changes its sign for any finite value of \( \tilde{T} \). This function has been depicted in Fig. 4(a). We can see that the model correctly predicts that there is no transition to an ordered state for any of the systems considered.
For comparison, we have also applied the present GCC method to the Heisenberg model with NN interactions on a simple cubic (s.c.) lattice. In the framework of the CW theory, it is predicted that both the s.c. and the pyrochlore lattices should have the same transition temperature to a LRO state, as they have the same number of NN. For the s.c., it is enough, for our purposes, to consider 2 different sublattices. Each spin in one sublattice has 6 NN in the other sublattice. Once we take into account this difference with respect to the pyrochlore lattice (which has \( p - 1 \) NN, as mentioned above), we obtain that, for ferromagnetic NN interactions, there is a transition to a ferromagnetic state for \( K_c = 0.615 \), to be compared with more refined estimates of this quantity from high-precision MC calculations, \( K_c = 0.693 \). For antiferromagnetic interactions, a transition to an antiferromagnetic (AF) state occurs at \( K_c = -0.615 \). Therefore, we can see that the present method is able to distinguish between the different topologies of the lattices, even when they have the same number of NN. Furthermore, the estimation of the critical point for the s.c. is very good (up to \( \sim 10\% \)), even though we have considered the smallest possible cluster that can describe the AF ordered state of the s.c. lattice.

As mentioned in the Introduction, one consequence of the presence of geometrical frustration in the system is that the NN exchange does not define an energy scale in the problem. Therefore, any perturbation is a strong perturbation, and can break the non-trivial degeneracy of the ground state, selecting one ordered state. In the following sections, we will study if NNN interactions and site dilution by non-magnetic impurities, which are always present in real systems, really break this non-trivial degeneracy, that is, if these effects are able to select an ordered state at finite temperature. Furthermore, we will focus the discussion on the pyrochlore lattice, for the reasons explained in the Introduction.

VI. EFFECT OF NNN INTERACTIONS

In order to include the effect of NNN interactions, we only need to redefine the SBF in the following way

\[
\langle \xi_{1\alpha} \rangle = \tilde{H}_0 + K_1 \sum_{\beta \neq \alpha} \sum_i z_i \langle \tilde{s}_{i\beta} \rangle + K_2 \sum_{\beta \neq \alpha} \sum_i z'_{i\beta} \langle \tilde{s}_{i\beta} \rangle = \frac{1}{3 |J_1| T} \left[ \tilde{h}_0 + 2(1 + z' \lambda) \sum_{\beta \neq \alpha} \tilde{h}_{\beta} \right],
\]

where \( z' \) stands for the number of NNN on each sublattice (\( z' = 2 \) for the kagome lattice and \( z' = 4 \) for the pyrochlore lattice) and \( \lambda = \frac{J_2}{J_1} \), with \( J_1 \) the NN exchange coupling and \( J_2 \) the NNN exchange coupling. For the cluster with \( p \) spins

\[
\langle \xi_{p\alpha} \rangle = \tilde{H}_0 + K_1 \sum_{\beta \neq \alpha} \sum_i z_{i\beta}^{-1} \langle \tilde{s}_{i\beta} \rangle + K_2 \sum_{\beta \neq \alpha} \sum_i z'_{i\beta} \langle \tilde{s}_{i\beta} \rangle = \frac{1}{3 |J_1| T} \left[ \tilde{h}_0 + (1 + z' \lambda) \sum_{\beta \neq \alpha} \tilde{h}_{\beta} \right],
\]

and

\[
\sum_{\beta \neq \alpha} \langle \xi_{p\beta} \rangle = \frac{1}{3 |J_1| T} \left\{ (p - 1)\tilde{h}_0 + (1 + z' \lambda) \left[ (p - 1)\tilde{h}'_{\alpha} + (p - 2) \sum_{\beta \neq \alpha} \tilde{h}'_{\beta} \right] \right\}.
\]

Therefore, the self-consistency condition that fixes the effective fields is given by

\[
(1 + z' \lambda)(p - 1)A_p(\tilde{T})\tilde{h}_\alpha' + \left[ 1 + (1 + z' \lambda)(p - 2)A_p(\tilde{T}) \right] \sum_{\beta \neq \alpha} \tilde{h}_{\beta}' = -(p - 1)A_p(\tilde{T})\tilde{h}_0,
\]

and its solution is given by

\[
\tilde{h}_\alpha' = -\frac{A_p(\tilde{T})}{1 + (1 + z' \lambda)(p - 1)A_p(\tilde{T})} \tilde{h}_0.
\]
Substituting back into the expression of the 1-spin cluster order parameter we find the expression for the susceptibility

\[ \chi_{gcc}^p = \frac{1}{3 |J_1| T} \frac{1 - (p - 1) A_p(T)}{1 + (1 + z' \lambda)(p - 1) A_p(T)} \]

or, in terms of the \( \varepsilon_p \) function defined above

\[ \chi_{gcc}^p = \frac{1}{3 |J_1| T} \frac{1 + \varepsilon_p(T)}{1 - (1 + z' \lambda) \varepsilon_p(T)}. \]

This susceptibility has been depicted in Fig. 5 for different values of \( \lambda \) and antiferromagnetic NN interactions.

![Figure 5: Effect of NNN interactions on the susceptibility of the pyrochlore lattice.](image)

In zero applied field, the system has two non-trivial solutions, similar to the ones found for NN interactions only. The ferromagnetic solution occurs if

\[ \varepsilon_p(T) = \frac{1}{1 + z' \lambda}, \]

and the non-collinear solution occurs if

\[ \varepsilon_p(T) = -\frac{(p - 1)}{1 + z' \lambda}. \]

These conditions have been studied and the main results are summarized in Fig. 6. As we can see from those figures, NNN interactions can stabilize different ordered states depending on the value of \( \lambda \) and the sign of the NN interaction. For antiferromagnetic NN interactions and \( |J_2| < 0.5 |J_1| \), the system remains paramagnetic down to 0 K. For antiferromagnetic NN and NNN interactions, a NC ordered estate is selected at finite temperature for \( \lambda > \frac{1}{2} \). However, for antiferromagnetic NN interactions, and ferromagnetic NNN interactions, a ferromagnetic ordered state is selected for \( \lambda < -\frac{1}{2} \). On the other hand, the case of ferromagnetic NN interactions is slightly more complex. For ferromagnetic NNN interactions, a ferromagnetic
ordered state is selected for $-\frac{1}{6} < \lambda$. In the range $-\frac{1}{2} < \lambda < -\frac{1}{6}$, the system remains paramagnetic down to 0 K and, in the range $\lambda < -\frac{1}{2}$, a NC ordered state is selected.

In any case, from the analysis of this section, it seems very unlikely that the non collinear ordered states experimentally found in some pyrochlore systems are due to NNN interactions, as in real pyrochlore systems $|J_2| \sim 0.1 |J_1|$, a value well inside the region of the phase diagram in which the system is found to remain paramagnetic down to 0 K.

It is also important to stress that the present treatment of NNN interactions is only approximate. In fact, in order to treat this problem consistently, we should include new sublattices such as spins in one sublattice do not interact with spins on the same sublattice. In doing so, it can happen that new kinds of ordered states arise. However, our main intention here is to check if the NC state given by (43) is selected by NNN interactions. The general problem with more sublattices will be studied elsewhere.

VII. EFFECTS OF SITE DILUTION BY NON-MAGNETIC IMPURITIES

In order to study the effect of site dilution on the susceptibility and on the conditions for the appearance of an ordered state, we need to average both the SBF and the order parameters over the distribution of non-magnetic impurities. For simplicity, we will assume that the distribution of non-magnetic impurities is purely random. Under this condition, the number of units with $q$ spins, for a lattice which, in the absence of dilution, is formed by units with $p$ spins, for a concentration of non-magnetic impurities $x$, is given by

$$P^p_q(x) = \binom{p}{q} (1 - x)^q x^{p-q}. \quad (56)$$
However, this distribution is normalized with respect to units, whereas we need a distribution function normalized with respect to a single spin. The adequate distribution function, for the SBF, is then

\[ P_p^p(x) = \frac{q}{p(1 - x)} P_q^p(x), \] (57)

and verifies the important relations we will use below

\[ \sum_{q=1}^{p} P_q^p(x) = 1, \] (58)

\[ \sum_{q=1}^{p} (q - 1) P_q^p(x) = (p - 1)(1 - x). \] (59)

The corresponding distribution function for averaging over the order parameter, however, has a different normalization. Indeed, if we average the SBF for the 1-spin cluster with ferromagnetic interactions with respect to (57) we have

\[ \sum_{q=1}^{p} P_q^p(x) \left\langle \vec{\xi}_{1\alpha} \right\rangle = \frac{\vec{h}_0}{|J| T} + \frac{2(p - 1)(1 - x)}{3|J| T} \vec{H}', \] (60)

in agreement with the intuitive idea that, upon dilution, the applied field is unaffected, and the EF changes according with the reduction of the average coordination number by a factor \((1 - x)\), indicating that the distribution function for the SBF is properly normalized. If we now average the order parameter with respect to the same distribution function, we find

\[ \sum_{q=1}^{p} P_q^p(x) \vec{m}_{q\alpha} = \frac{\vec{h}_0}{3|J| T} + \frac{2(p - 1)(1 - x)}{3|J| T} \vec{H}', \] (61)

and this result is incorrect, as it does not take into account the reduction in the total number of magnetic ions in the lattice upon dilution. Therefore, the correctly normalized distribution function we have to use in order to average over the order parameter is given by

\[ Q_p^p(x) = (1 - x) P_q^p(x). \] (62)

In the following, we will use the following notation to denote averages with respect to dilution

\[ [f_q]_p = \sum_{q=1}^{p} P_q^p(x) f_q, \] (63)

where \( f_q \) is any quantity, and it will be understood that we are using the distribution (57) when averaging over SBF and the distribution (62) when averaging over the order parameter.

Furthermore, for the sake of clarity, we will consider the antiferromagnetic case with no applied field separately from the paramagnetic and ferromagnetic with no applied field cases. In the paramagnetic case and ferromagnetic case in the absence of an applied field, we have seen that the system can be described by a single effective field, \( \vec{H}' \). The average with respect to dilution of the SBF for the 1-spin cluster, as we have seen above, is given by

\[ \left\langle \vec{\xi}_{1\alpha} \right\rangle_p = \frac{2(p - 1)(1 - x)}{|J| T} \vec{H}' + \frac{\vec{h}_0}{|J| T}, \] (64)
where we have only considered the contribution of NN to the SBF. Now, the averaged 1-spin order parameter is given by
\[
[\vec{m}_{1\alpha}]_p = \frac{1-x}{3|J|T} \left[ \tilde{h}_0 + 2(p-1)(1-x)\tilde{h}' \right]. \tag{65}
\]
Analogously, for the \(p\)-spin cluster we have
\[
\left[ \langle \vec{\xi}_{q\alpha} \rangle \right]_p = \frac{(p-1)(1-x)}{|J|T} \tilde{h}' + \frac{\tilde{h}_0}{|J|T}, \tag{66}
\]
\[
\sum_{\beta \neq \alpha} \left[ \langle \vec{\xi}_{q\beta} \rangle \right]_p = \frac{q-1}{|J|T} \left( (p-1)(1-x)\tilde{h}' + \tilde{h}_0 \right), \tag{67}
\]
and the corresponding averaged order parameter is given by
\[
[\vec{m}_{q\alpha}]_p = \frac{1-x}{3|J|T} \left( 1 - \tilde{A}_p(\tilde{T}) \right) \left( \tilde{h}_0 + (p-1)(1-x)\tilde{h}' \right), \tag{68}
\]
where
\[
\tilde{A}_p(\tilde{T}) = \sum_{q=1}^p P_q^p(x) (q-1) A_q(\tilde{T}). \tag{69}
\]
Therefore the averaged self-consistency condition
\[
[\vec{m}_{1\alpha}]_p = [\vec{m}_{q\alpha}]_p, \tag{70}
\]
leads us to the following equation for the effective field \(\tilde{h}'\)
\[
\tilde{h}_0 + 2(p-1)(1-x)\tilde{h}' = (1 - \tilde{A}_p) \left( \tilde{h}_0 + (p-1)(1-x)\tilde{h}' \right), \tag{71}
\]
that has the solution
\[
\tilde{h}' = -\frac{\tilde{A}_p}{(p-1)(1-x)(1+\tilde{A}_p)} \tilde{h}_0, \tag{72}
\]
and the corresponding susceptibility is found to be
\[
[\chi]_p = \frac{1-x}{3|J|T} \frac{1 - \tilde{A}_p}{1 + \tilde{A}_p}, \tag{73}
\]
which is of the same form as \((36)\), except for that \((p-1)A_p\) is replaced by the corresponding average over dilution. Interestingly, it is easily shown that this average can be expressed in terms of an average over the \(\varepsilon\) function introduced before. Indeed, if we define
\[
\tilde{\varepsilon}_p = \frac{2\tilde{T}^2}{p(1-x)} \frac{\partial}{\partial \tilde{T}} \ln \tilde{z}_p^0(\tilde{T}, x) - 1, \tag{74}
\]
where \(\tilde{z}_p^0\) is the average of \(\varepsilon_p^0\) with respect to dilution, given by
\[
\tilde{z}_p^0 = \prod_{q=1}^p \tilde{z}_p^0(\tilde{T}) P_q^p(x), \tag{75}
\]
we have the identity

\[ \bar{\varepsilon}_p = -\bar{A}_p, \]  

and we can put the averaged susceptibility in the form

\[ [\chi]_p = \frac{1 - x}{3 |J| T} \left( 1 + \frac{1}{1 - \bar{\varepsilon}_p} \right). \]  

Incidentally, this is precisely the susceptibility calculated by the present authors in Ref. [27], so we recover all the results presented there for the paramagnetic case.

![Phase diagram](image1)

**Figure 7:** (a) Phase diagram for the ferromagnetic pyrochlore with site dilution. (b) Study of the condition for the existence of non-collinear order. $F_4$ stands for the function $F_p = \bar{\varepsilon}_p - (p - 1)(1 - x)$ for $p = 4$ (see text).

In the absence of applied field, it can be easily shown from eq. (71) that the condition for the formation of a ferromagnetic ordered state is given by

\[ \bar{\varepsilon}_p = 1. \]  

(78)

To check the consistency of the results, we have studied this condition for different amounts of dilution for the ferromagnetic pyrochlore, and the dependence of the transition temperature with $x$ is depicted in Fig. 7. We can see that there is a critical value $x_c$ above which there is no transition. This value is in reasonable agreement with the value of the percolation threshold for the pyrochlore lattice, $x_c \approx 0.6$, whereas our model predicts a value $x_c \approx 0.66$. This result is reasonable, as the transition temperatures predicted by our model are slightly larger than the real ones, due to neglecting long wavelength fluctuations.

Let us now consider the antiferromagnetic case with no applied field, and let us calculate the condition for the existence of non-collinear order upon dilution. In this case, we know from the analysis in the previous sections that the effective fields $\tilde{h}'_{\alpha}$ verify the relation

\[ \sum_{\beta \neq \alpha} \tilde{h}'_{\beta} = -\tilde{h}'_{\alpha} \]  

(79)
at the transition point, if any. Therefore, we have the following expressions for the dilution averaged SBF
in this case
\[
\langle \xi_{1\alpha} \rangle_p = 2 \langle \xi_{q\alpha} \rangle_p = -\frac{2\tilde{h}_\alpha'}{|J| T},
\]
and
\[
\sum_{\beta \neq \alpha} \langle \xi_{q\beta} \rangle_p = \frac{\tilde{h}_\alpha}{|J| T}.
\]
The order parameter for a unit with \( q < p \) spins is then given by
\[
\bar{m}_{q\alpha} = \frac{1}{3|J| T} [1 + A_q] \tilde{h}_\alpha'.
\]
If we now naively average the self consistency condition as above, we would not be able to obtain the
condition for the transition to a non-collinear state in terms of the \( \bar{A}_p \) functions introduced before, as
a \((q - 1)\) prefactor is lacking in the expression for \( \bar{m}_{q\alpha} \). Thus, we construct the following averages instead
\[
[(q - 1) \bar{m}_{1\alpha}]_p = -\frac{2(p - 1)(1 - x)}{3|J| T} \tilde{h}_\alpha',
\]
and
\[
[(q - 1) \bar{m}_{q\alpha}]_p = -\frac{x}{3|J| T} ((p - 1)(1 - x) + \bar{A}_p') \tilde{h}_\alpha'.
\]
Equating both quantities, we easily obtain the condition for the existence of non-collinear order upon site
dilution
\[
\bar{e}_p = -(p - 1)(1 - x),
\]
where we have made use of relation (76).

As can be seen from Fig. 7(b), the function \( F_p = \bar{e}_p - (p - 1)(1 - x) \) does not change its sign for any
value of \( x \), indicating that a non-collinear ordered state is not selected by site dilution, in contrast with
the qualitative picture suggested in other works.

VIII. COMBINED EFFECT OF NNN INTERACTIONS AND SITE DILUTION

Of course, in real systems, the effects analyzed above are always present simultaneously. Therefore, it
is important to study how the conclusions reached in the previous sections are modified when we simulta-
neously consider both NNN interactions and site dilution. We will only present the main results of the
analysis, as the derivation of the final expressions follows very closely the previous derivations.

The averaged susceptibility is obtained to be
\[
[\chi]_p = \frac{1 - x}{3 |J_1| T} \frac{1 + \bar{e}_p}{1 - (1 + z'\lambda) \bar{e}_p}.
\]
The condition for the existence of a ferromagnetic ordered state is given by \( \bar{e}_p = \frac{1}{1 + z'\lambda} \), whereas the
corresponding one for a non-collinear ordered state is given by \( \bar{e}_p = -\frac{(p - 1)(1 - x)}{1 + z'\lambda} \). The phase diagrams,
for different concentrations of non-magnetic impurities are depicted in Fig. 8. Also, in Fig. 8 we have
depicted the values of \( \lambda_c(x) \), that is, the value of \( \frac{J_2}{J_1} \) above which a non-collinear ordered state is formed at
a finite temperature, for a given value of \( x \). We have only depicted this quantity for antiferromagnetic NN
interactions, as the corresponding \( \lambda_c \) for ferromagnetic NN interactions does not change upon dilution.
IX. SUMMARY AND CONCLUSIONS

In this work we have presented a microscopic formulation of the Generalized Constant Coupling method for geometrically frustrated magnets. By introducing different Bravais sublattices characterized by different order parameters we are able to describe more general kinds of long range ordered states, including non-collinear ones. These order parameters are evaluated by making use of the Suzuki-Callen identity for finite size clusters, in terms of symmetry breaking fields arising from interactions with units surrounding the
considered one. The symmetry breaking fields are easily related with the phenomenological effective fields introduced in the original formulation of the generalized constant coupling method, in terms of averages over the neighboring spin variables outside the unit, and they are fixed by a self-consistency condition. Though we have not stressed this point, this self-consistency condition is, in fact, a particular case of the more general scaling hypothesis between the order parameters for finite clusters of different sizes.

When a magnetic field is applied to the system, this method allows us to easily compute the susceptibility, and we recover previous results obtained in the phenomenological formulation of the GCC method, which were shown to be in excellent agreement with MC results for both the kagome and pyrochlore lattice. However, the present formulation, allows us to also study the critical behavior of the system when no applied magnetic field is present. The possible ordered configurations for these systems naturally arise from the equations that fix the order parameters. In completely agreement with the accepted idea, for ferromagnetic interactions a ferromagnetic configuration in which all the spins point along the same direction is found. For antiferromagnetic interactions, a non-collinear state in which the total magnetization of each cluster is identically zero is found. Also, the conditions for the formation of such ordered states arise from the calculation. It is found that, for nearest neighbor antiferromagnetic interactions, frustration inhibits the formation of a long range ordered state, and the system remains paramagnetic down to 0 K. We have briefly compared the results for these frustrated geometries with the results for a simple cubic lattice obtained with this same method, and we found that the generalized constant coupling method correctly distinguishes between different topologies of the lattice, even in case where the number of nearest neighbors is the same, as it happens with the pyrochlore and cubic lattices. Another advantage of the present formulation is that it is constructed in the real space, which makes it very easy to include additional perturbations to the system, by modifying the form of the symmetry breaking fields. Particularly, we have studied the effect of next nearest neighbor interactions and site dilution by non-magnetic impurities for the pyrochlore lattice. In the former case, the phase diagram has been calculated. It is found that, for certain values of the ratio \( \lambda = \frac{J_2}{J_1} \), a non-collinear or ferromagnetic ordered state can be stabilized. However, the minimum value of \( \lambda \) above which a non-collinear configuration is stable seems to be too large to explain the formation of long range order found in some systems, notably in Gd_{2}Ti_{2}O_{7}. Moreover, it seems clear from neutron scattering that the ordered configuration in this case is not compatible with the condition that the total magnetization of the tetrahedral cluster is zero. However, the main perturbation in this system seems to come from long range dipolar interactions, which are highly anisotropic. Further theoretical work in this direction, in the present framework, is still necessary.

Regarding the effects of site dilution, it has been argued that this kind of effect, always present in real materials, could break the non-trivial degeneracy of the ground state and cause a transition to some kind of long range ordered state at finite temperature. However, in our calculation, we find that this is not case: Site dilution by itself does not induce a transition, for any amount of dilution. It is necessary to include simultaneously both dilution and next nearest neighbor interactions.

A word of caution must be said here, however. In the last part of this work, we have focused our attention on studying if a non-collinear ordered state given by the rule that the total magnetization of the tetrahedral unit is zero is stabilized by any of these perturbations. However, these are not the only possible ordered configurations when we include these effects. For example, Reimers and coworkers showed that, in the framework of mean field theory, next nearest neighbor interactions can induce both non-collinear ordered states as the ones studied in this work, or ordered states characterized by an incommensurate wave vector. It is not easy to describe these incommensurate states with a real space method, and it could be the case that site dilution would lead to such an incommensurate state. Therefore, more theoretical work in that direction should be necessary before ruling out the possibility of a long range ordered state induced by small amounts of site dilution or, what would be even more interesting, the possibility of a transition to some kind of spin glass state induced by dilution.

In conclusion, the formulation of the generalized constant coupling method presented in this work provides us a conceptual framework in which both thermodynamic quantities and the critical behavior of geo-
metrically frustrated magnets can be properly described. The method can be easily generalized to deal with further perturbations always present in real systems. We have presented the classical limit of the method. However, in order to compare the calculated quantities with experimental data for these systems, the corresponding quantum generalized constant coupling method should be used. This issue, the occurrence of incommensurate states due to dilution, and the effect of the inclusion of long range dipolar interactions in the Hamiltonian are open issues which deserve further theoretical work.

Appendix A: EVALUATION OF THE $P$-SPIN CLUSTER PARTITION FUNCTION.

In this appendix we present the main step for the evaluation of the integral (20)

$$Z_p(K) = \int \prod_{\alpha} d\Omega_{\alpha} e^{\mathcal{H}_p},$$  \hspace{1cm} (A1)

which can be put in the more convenient form

$$Z_p(K) = \int d^3\vec{S} \prod_{\alpha} d^3\vec{s}_{1\alpha} \delta \left( \vec{S} - \sum_{\alpha} \vec{s}_{1\alpha} \right) \exp \left( K \sum_{\alpha \neq \beta} \vec{s}_{1\alpha} \cdot \vec{s}_{1\beta} + \sum_{\alpha} \vec{s}_{1\alpha} \cdot \vec{\xi}_{p\alpha} \right).$$  \hspace{1cm} (A2)

By using the Fourier transform representation of the Dirac-$\delta$

$$\delta(\vec{x}) = \frac{1}{(2\pi)^3} \int d^3\vec{q} e^{i\vec{q} \cdot \vec{x}},$$  \hspace{1cm} (A3)

we can further put

$$Z_p(K) = \frac{1}{(2\pi)^3} \int d^3\vec{q} \int d^3\vec{S} e^{K \vec{S} \cdot \vec{s}} \prod_{\alpha} \int d^3\vec{s}_{\alpha} e^{i\vec{a}_{\alpha} \cdot \vec{s}_{\alpha}},$$  \hspace{1cm} (A4)

where we have introduced the vector $\vec{a} = \vec{\xi}_{p\alpha} - i\vec{q}$, and made use of the fact

$$\frac{1}{2} \sum_{\alpha \neq \beta} \vec{s}_{1\alpha} \cdot \vec{s}_{1\beta} = \frac{S^2}{2},$$  \hspace{1cm} (A5)

with $\vec{S}$ the total spin of the unit, up to an unimportant global constant in the Hamiltonian. The integral over $\vec{s}_{1\alpha}$ can be done as follows. Taking into account

$$\vec{a} \cdot \vec{s} = a_x \sin \theta \cos \varphi + a_y \sin \theta \sin \varphi + a_z \cos \theta,$$  \hspace{1cm} (A6)

where we have assumed that the length of $\vec{s}_{1\alpha}$ is 1 (this can be always done by redefining $K \rightarrow K s_0^2$ with $s_0$ the length of the spin), the integral over $\vec{s}_{1\alpha}$ can be put as

$$\int_0^{2\pi} d\varphi \int_0^{\pi} d\theta \sin \theta e^{a_z \cos \theta + a_y \sin \theta \sin \varphi + a_z \cos \theta} = 2\pi \int_0^{\pi} d\theta \sin \theta e^{a_z \cos \theta} I_0 \left( \sqrt{a_x^2 + a_y^2} \sin \theta \right)$$

$$= 2\pi \int_{-1}^{1} dx e^{-ax} I_0 \left( b\sqrt{1 - x^2} \right) = 4\pi \sinh \frac{\sqrt{a_x^2 + b^2}}{\sqrt{a_x^2 + b^2}} = 4\pi \sinh \frac{\sqrt{a_x^2 + a_y^2 + a_z^2}}{\sqrt{a_x^2 + a_y^2 + a_z^2}} = 4\pi \frac{\sin a_x}{a_x},$$  \hspace{1cm} (A7)

where $a_{\alpha} = q + i\xi_{p\alpha}$ (we have factored out a (-1) in the definition of $a_{\alpha}$).
Next, we can do the integral over $\vec{S}$

$$\int d^3 \vec{S} e^{\frac{K}{2} \vec{S}^2} e^{i \vec{q} \cdot \vec{S}} = 4\pi \sqrt{\frac{\pi}{2}} (-K)^{3/2} e^{-q^2/2(-K)},$$  \hspace{1cm} (A8)

which only converges for $K < 0$, and we are left with an integral over $\vec{q}$

$$Z_p(K) \sim \int d^3 \vec{q} e^{\frac{q^2}{4K} \prod_\alpha \sin \sqrt{q^2 + 2i \vec{q} \cdot \vec{\xi}_p \xi - \xi_{p\alpha}^2 \xi_{p\alpha}^2}},$$  \hspace{1cm} (A9)

which cannot be calculated in a closed form. An interesting point here is that, if we take $\vec{\xi}_{p\alpha} = \vec{0}$, we recover the partition function calculated in the original GCC model. The only thing we can do at this point is to expand the partition function in powers of the SBF, up to second order, as the rest of terms will be assumed to be small near the critical point once we take the thermal average. With a little algebra it is easy to show that

$$\sin \sqrt{q^2 + 2i \vec{q} \cdot \vec{\xi}_p \xi - \xi_{p\alpha}^2 \xi_{p\alpha}^2} \simeq f_0(q) + i f_1(q) \xi_{p\alpha} \cos \theta_\alpha + f_2(q) \xi_{p\alpha}^2 + f_3(q) \xi_{p\alpha}^2 \cos^2 \theta_\alpha,$$  \hspace{1cm} (A10)

where $\theta_\alpha$ is the angle defined by $\vec{\xi}_{p\alpha}$ and $\vec{q}$, and

$$f_0(q) = \frac{\sin q}{q}$$  \hspace{1cm} (A11)

$$f_1(q) = \frac{\cos q}{q} - \frac{\sin q}{q^2}$$  \hspace{1cm} (A12)

$$f_2(q) = \frac{\sin q}{2q^3} - \frac{\cos q}{2q^2}$$  \hspace{1cm} (A13)

$$f_3(q) = \frac{3\cos q}{2q^2} - \frac{3\sin q}{2q^3} + \frac{\sin q}{2q}.$$  \hspace{1cm} (A14)

By making use of this result, we can put

$$\prod_\alpha \sin \sqrt{q^2 + 2i \vec{q} \cdot \vec{\xi}_p \xi - \xi_{p\alpha}^2 \xi_{p\alpha}^2} \simeq f_0^p + f_0^{p-1} f_2 \sum_\alpha \xi_{p\alpha}^2 + f_0^{p-1} f_3 \sum_\alpha \cos^2 \theta_\alpha \xi_{p\alpha}^2$$

$$- \frac{f_0^{p-2} f_1^2}{2} \sum_{\alpha \neq \beta} \cos \theta_\alpha \cos \theta_\beta \xi_{p\alpha} \xi_{p\beta} + i f_0^{p-1} f_1 \sum_\alpha \cos \theta_\alpha \xi_{p\alpha}.$$  \hspace{1cm} (A15)

The expressions of the form $\xi_{p\alpha} \cos \theta_\alpha$ are the projection of $\vec{\xi}_{p\alpha}$ over $\vec{q}$, and can be put in the more convenient form

$$\xi_{p\alpha} \cos \theta_\alpha = \xi_{p\alpha}^x \sin \theta \cos \varphi + \xi_{p\alpha}^y \sin \theta \sin \varphi + \xi_{p\alpha}^z \cos \theta.$$  \hspace{1cm} (A16)
If we consider the imaginary part of the previous series, and integrate over the angular part of $\vec{q}$ in (A9), we have integrals of the form

$$\int_0^\pi d\theta \int_0^{2\pi} d\varphi \sin \theta \, \xi_{\rho\alpha} \cos \theta_{\alpha},$$

which are easily shown to be zero.

We also have integrals of the form

$$\int_0^\pi d\theta \int_0^{2\pi} d\varphi \sin \theta \, (\xi_{\rho\alpha} \cos \theta_{\alpha})^2 = \frac{4\pi}{3} \left[ (\xi_{\rho\alpha}^x)^2 + (\xi_{\rho\alpha}^y)^2 + (\xi_{\rho\alpha}^z)^2 \right] = \frac{4\pi}{3} \xi_{\rho\alpha}^2$$

(A18)

and

$$\int_0^\pi d\theta \int_0^{2\pi} d\varphi \sin \theta \, \xi_{\rho\alpha} \cos \theta_{\alpha} \xi_{\rho\beta} \cos \theta_{\beta} = \frac{4\pi}{3} \left[ \xi_{\rho\alpha\beta}^x \xi_{\rho\alpha}^x + \xi_{\rho\alpha\beta}^y \xi_{\rho\alpha}^y + \xi_{\rho\alpha\beta}^z \xi_{\rho\alpha}^z \right] = \frac{4\pi}{3} \xi_{\rho\alpha} \cdot \xi_{\rho\beta}. $$

(A19)

Substituting these results back in (A15) we are left with the integral over $q$

$$Z_p(K) \sim \int_0^\infty dq \, q^2 e^{\frac{q^2}{2K}} \left( f_0^p + f_0^{p-1} f_2 \sum_\alpha \xi_{\rho\alpha}^2 + \frac{f_0^{p-1} f_3}{3} \sum_\alpha \xi_{\rho\alpha}^2 - \frac{f_0^{p-2} f_1^2}{6} \sum_{\alpha \neq \beta} \xi_{\rho\alpha} \cdot \xi_{\rho\beta} \right)$$

(A20)

which, taking into account

$$f_2 + \frac{f_3}{3} = \frac{f_0}{6}$$

(A21)

can be simplified to the form

$$Z_p \sim \int_0^\infty dq \, q^2 e^{\frac{q^2}{2K}} \left( f_0^p + \frac{f_0^p}{6} \sum_\alpha \xi_{\rho\alpha}^2 - \frac{f_0^{p-2} f_1^2}{6} \sum_{\alpha \neq \beta} \xi_{\rho\alpha} \cdot \xi_{\rho\beta} \right) .$$

(A22)

Each of the integrals can be put as a closed expressions, and are given in the next section. For the moment, we will use the notation

$$z_0^K = \int_0^\infty dq \, q^2 e^{\frac{q^2}{2K}} f_0^p$$

(A23)

and

$$z_1^K = \int_0^\infty dq \, q^2 e^{\frac{q^2}{2K}} f_0^{p-2} f_1^2 .$$

(A24)

Finally, up to second order in the SBF, we can put

$$Z_p(K) \sim z_0^K + \frac{z_0^K}{6} \sum_\alpha \xi_{\rho\alpha}^2 - \frac{z_1^K}{6} \sum_{\alpha \neq \beta} \xi_{\rho\alpha} \cdot \xi_{\rho\beta} .$$

(A25)
Appendix B: EXPRESSIONS FOR $Z^0_p(K)$ AND $Z^0_p(K)$

The $z^0_p(K)$ function for different values of $p$ and antiferromagnetic interactions ($K < 0$) is given as follows

\begin{align*}
z^0_1(K) &= e^\frac{K}{2}, \\
z^0_2(K) &= e^{2K} - 1, \\
z^0_3(K) &= \frac{3 \text{erf} \left(\sqrt{-\frac{K}{2}}\right) + \text{erf} \left(3\sqrt{-\frac{K}{2}}\right)}{(-K)^{5/2}}, \\
z^0_4(K) &= \frac{2 \text{erf} \left(\sqrt{-2K}\right) - \text{erf} \left(\sqrt{-8K}\right)}{(-K)^{3/2}} - \frac{3 + e^{8K} - 4e^{-2K}}{\sqrt{8\pi} K^2}. \tag{B4}
\end{align*}

In these expressions, $\text{erf}$ represents the error function. It is important to notice that, if we make the replacement $K \rightarrow -\frac{1}{t}$, we immediately recover the expressions of Ref. [3]. For ferromagnetic interactions ($K > 0$), the expressions are completely similar to the ones above, with the substitutions $\sqrt{-K} \rightarrow \sqrt{K}$ and $\text{erf} \rightarrow \text{erfi}$, with $\text{erfi}(x) = \frac{\text{erf}(ix)}{i}$ the imaginary error function. To quote an example

\begin{align*}
z^0_4(K) &= \frac{2 \text{erfi} \left(\sqrt{2K}\right) - \text{erfi} \left(\sqrt{8K}\right)}{K^{3/2}} - \frac{3 + e^{8K} - 4e^{-2K}}{\sqrt{8\pi} K^2}. \tag{B5}
\end{align*}

The $z^1_p(K)$ functions are obtained to be, for antiferromagnetic interactions

\begin{align*}
z^1_2(K) &= \frac{e^K}{K^2} (\sinh K - K \cosh K), \\
z^1_3(K) &= \frac{e^{3K}(e^{-4K} - 1)}{K^2} - \sqrt{\frac{\pi}{2}} \frac{1 + K}{(-K)^{3/2}} \left[3 \text{erf} \left(\sqrt{-\frac{K}{2}}\right) - \text{erf} \left(3\sqrt{-\frac{K}{2}}\right)\right], \tag{B7}
\end{align*}

\begin{align*}
z^1_4(K) &= \sqrt{\frac{\pi}{2}} \frac{4K + 3}{(-K)^{5/2}} \left[2 \text{erf} \left(\sqrt{-2K}\right) - \text{erf} \left(\sqrt{-8K}\right)\right] - \frac{(1 + K)(3 + e^{8K} - 4e^{2K})}{K^3}. \tag{B8}
\end{align*}

The corresponding expressions for the ferromagnetic case can be obtained by using the prescription indicated above. It is important to notice that $z^1_p(K)$ is not defined for $p = 1$ but it easy to see that this function does not appear in any calculation, as it corresponds to non-interacting spins.

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