

# Quantum generalized constant-coupling model for geometrically frustrated antiferromagnets

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A generalized constant-coupling approximation for quantum geometrically frustrated antiferromagnets is presented. Starting from a frustrated unit, we introduce the interactions with the surrounding units in terms of an internal effective field that is fixed by a self-consistency condition. Results for the static magnetic susceptibility and specific heat are compared with previous results in the framework of this same model for the classical limit. The range of applicability of the model is discussed.

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## I. INTRODUCTION

In the last several years, geometrically frustrated antiferromagnets (GFAF) have emerged as a new class of magnetic materials with uncommon physical properties and have received a great deal of attention (see Refs. 1–3, and references therein). 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. Examples of GFAF are the pyrochlore and the *kagomé* lattices. In the former, the magnetic ions occupy the corners of a three-dimensional arrangement of corner-sharing tetrahedra; in the latter, the magnetic ions occupy the corners of a two-dimensional (2D) arrangement of corner-sharing triangles (see Fig. 1). In the case of materials that crystallize in the pyrochlore structure, the static magnetic susceptibility follows the Curie-Weiss law down to temperatures well below the Curie-Weiss temperature. At this point, usually one to two orders of magnitude smaller than the Néel point predicted by the standard mean-field (MF) theory, some systems exhibit some kind of long-range order (LRO), whereas others show a transition to a spin-glass state (SG). This is a striking feature for a system with only a marginal amount of disorder. Finally, there are some pyrochlores that do not exhibit any form of order whatsoever, and these are usually regarded as spin liquids. In the case of the *kagomé* lattice, even though there are very few real systems where this structure is realized, the magnetic properties fall in two major categories: the vast majority of the compounds studied show a transition to a LRO state with a noncollinear configuration of spins, and a few systems exhibit no LRO, but a SG-like transition.

For these reasons, it is easy to understand the large amount of attention these systems have attracted, both from the experimental and the theoretical points of view. From the theoretical point of view, a number of techniques have been used to try to understand the origin of the puzzling properties mentioned above. All the theoretical results seem to indicate that the classical Heisenberg model with only nearest-neighbor interactions does not display any long-range order for these geometries, in agreement with Monte Carlo (MC) results.<sup>4–13</sup>

There are also relatively few works that have dealt with the quantum effects in these systems,<sup>14–22</sup> even though the main interest over the last years has been the classical GFAF.

However, in a recent work,<sup>19</sup> the present authors showed that there are some features of the experimental data for the susceptibility in pyrochlore compounds that can only be understood in a quantum framework, such as, for example, the maxima appearing in this quantity at temperatures well below the Curie-Weiss temperature.

In another recent work,<sup>23</sup> the present authors developed a generalization of the well-known constant-coupling (CC) method<sup>24</sup> that can be applied to frustrated geometries, the so-called generalized constant-coupling (GCC) method. This technique was applied successfully to the study of the classical Heisenberg Hamiltonian with nearest-neighbor (NN) interactions in both the pyrochlore and *kagomé* lattices. In spite of the mathematical simplicity of this technique, the results obtained for the susceptibility are essentially exact when compared with Monte Carlo data in both types of lattices. Moreover, the calculated specific heat is also in very good agreement with Monte Carlo data, even though there are some deviations at very small temperatures that can be understood in light of the fact that a MF theory cannot properly describe this property (or, equivalently, the internal energy) at  $T=0$  K, due to the distinct nature of the excitations

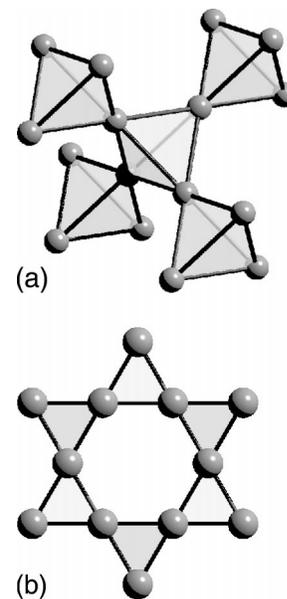


FIG. 1. The magnetic lattices considered in this work. (a) Pyrochlore lattice. (b) *kagomé* lattice.

that are important in this limit, namely, the spin waves. In any case, the GCC method provides an excellent starting point for studying the magnetic properties of the frustrated systems in the paramagnetic region.

The first question that arises is which features of these magnetic properties are different in a quantum formulation of the GCC method, and that is precisely what we try to answer in this work. However, in contrast with the classical limit, there are no reliable quantum Monte Carlo calculations for these systems, due to well-known difficulties that arise in the quantum version of this technique, which makes it very difficult to check the accuracy of the present quantum version of the GCC method. It is tempting to compare the results of this method with experimental data available in the literature. However, here we are considering the most restrictive approach to the problem by focusing on Heisenberg models with NN interactions only, whereas in real systems, as pointed out above, there are always additional effects, so we think such a comparison makes no sense at this point. Therefore, the most we can do is to give the results for the quantum version of the GCC method, and study how they evolve towards the classical results for large values of the individual spin quantum number. If this transition from the quantum to the classical limit is smooth, we can expect the predictions of the model to be, at least, reasonable in the paramagnetic region.

## II. THE MODEL

The Heisenberg model with only NN interactions in the presence of a magnetic applied field  $H_0$  is described by the Hamiltonian<sup>25</sup>

$$H = J \sum_{\langle i,j \rangle} \mathbf{s}_i \cdot \mathbf{s}_j - H_0 \sum_i s_{z_i}, \quad (1)$$

where  $J$  is the positive antiferromagnetic coupling,  $\mathbf{s}_i$  and  $\mathbf{s}_j$  represent quantum spins of modulus  $s_0$  located in a pyrochlore or *kagomé* lattice,  $s_{z_i}$  the corresponding component along the applied field, and the sum is done over pairs of NN's.

The idea of our approximate method is based on the experimental fact that the spin-spin correlations in the GFAF lattices are short ranged.<sup>4</sup> Therefore, it is a reasonable approximation to start by considering isolated units (tetrahedra or triangles for the pyrochlore and *kagomé* lattices, respectively) and later add the interactions with the surrounding units in an approximate way. Thus, it is important to first study the properties of the individual units. This task has been carried out by García-Adeva and Huber for the quantum case,<sup>19</sup> and we will not repeat the derivation here. It will suffice to say that the susceptibility per spin of an isolated unit with  $p$  spins ( $p=4$  for the pyrochlore and  $p=3$  for the *kagomé*) is given by

TABLE I. Values of  $g(S)$  for some representative values of  $s_0$ . Integer values of  $S$  correspond to the pyrochlore case ( $p=4$ ) for any value of  $s_0$  and the *kagomé* lattice for integer values of  $s_0$ . Half-integer values of  $S$  correspond to the *kagomé* lattice ( $p=3$ ) for half-integer values of  $s_0$ .

		$S$										
		$\frac{1}{2}$	$\frac{3}{2}$	$\frac{5}{2}$	$\frac{7}{2}$	$\frac{9}{2}$	$\frac{11}{2}$	$\frac{13}{2}$	$\frac{15}{2}$	8	9	10
		0	1	2	3	4	5	6	7	8	9	10
$p=3$	$s_0=\frac{1}{2}$	2	1									
	$s_0=1$	1	3	2	1							
	$s_0=\frac{3}{2}$	2	4	3	2	1						
	$s_0=\frac{5}{2}$	2	4	6	5	4	3	2	1			
$p=4$	$s_0=\frac{1}{2}$		2	3	1							
	$s_0=1$	3	6	6	3	1						
	$s_0=\frac{3}{2}$	4	9	11	10	6	3	1				
	$s_0=\frac{5}{2}$	6	15	21	24	24	21	15	10	6	3	1

$$\chi_p(T) = \frac{\langle S_p^2 \rangle}{3pT} = \frac{1}{3pT} \frac{\sum_S g(S) S(S+1)(2S+1) e^{-jS(S+1)/2}}{\sum_S g(S)(2S+1) e^{-jS(S+1)/2}}, \quad (2)$$

where  $j=J/T$ ,  $\langle S_p^2 \rangle$  represents the average value of the square of the total spin of the unit, and  $g(S)$  is the number of configurations with total spin  $S$  (see Table I).

For some derivations, it is better to express all the thermodynamic quantities in terms of the function

$$\varepsilon_p(T) = \frac{\langle S_p^2 \rangle}{ps_0(s_0+1)} - 1. \quad (3)$$

Therefore, the susceptibility of the isolated unit can be expressed as

$$\chi_p(T) = \frac{s_0(s_0+1)[1 + \varepsilon_p(T)]}{3T}. \quad (4)$$

It is important to notice that the only difference from the derivation carried out in Ref. 23 is in the definition of  $\langle S_p^2 \rangle$ . The rest of the method is essentially the same.

Next, the interaction with neighboring units is introduced as an unknown internal effective field  $H_1$  created by the  $(p-1)$  NN ions outside the unit. The CC approximation consists of taking this internal field as  $H_1 = (p-1)H'$ , where  $H'$  is the average internal field acting on a spin due to each of its NN's. By comparison, in the case of the standard Curie-Weiss (CW) model, the internal effective field is given by  $H_1 = 2(p-1)H'$  as the ions are considered separately, and each has  $2(p-1)$  NN's in the corner-sharing structures considered in this work (see Fig. 1). The internal field is evaluated by imposing the self-consistent condition of equating the magnetization per spin in the field with that of a unit in the field, which can be mathematically stated as

$$s_0 B_{s_0}(s_0[H_0 + 2(p-1)H']/T) = \frac{m_p(H_0 + (p-1)H')}{p}, \quad (5)$$

where the left side of the equation corresponds to the value of the magnetization per spin in the Curie-Weiss model, with

$$B_{s_0}(x) = \frac{2s_0 + 1}{s_0} \coth\left(\frac{2s_0 + 1}{s_0}x\right) - \frac{1}{2s_0} \coth\left(\frac{x}{2s_0}\right) \quad (6)$$

the so-called Brillouin function.<sup>25</sup> The right side corresponds to the magnetization per spin of the isolated unit under the influence of the internal field. In the general case, Eq. (5) can only be solved numerically. However, in the paramagnetic regime, we can put, for small fields,  $(H_0 + H_1)/T \ll 1$ ,

$$\frac{m_p(H_0 + (p-1)H')}{p} \approx \chi_p(T)[H_0 + (p-1)H'] \quad (7)$$

for the magnetization per spin in the cluster, where the susceptibility per spin is given by Eq. (4), and

$$s_0 B_{s_0}(s_0[H_0 + 2(p-1)H']/T) \approx \frac{s_0 + 1}{3T} [H_0 + 2(p-1)H']. \quad (8)$$

Taking into account Eqs. (7) and (8) we can easily solve Eq. (5) to give

$$H' = \frac{\varepsilon_p(j)}{(p-1)[1 - \varepsilon_p(j)]} H_0, \quad (9)$$

which, upon substitution in Eqs. (7) or (8), leads to an expression for the susceptibility per spin in the paramagnetic region<sup>23</sup>

$$\chi_p^{\text{GCC}}(T) = \frac{s_0(s_0 + 1)}{3T} \frac{1 + \varepsilon_p(T)}{1 - \varepsilon_p(T)}. \quad (10)$$

The corresponding expressions for the internal energy<sup>26</sup> and the specific heat per spin are given by

$$u_p(T) = J s_0 (s_0 + 1) \varepsilon_p(T) \quad (11)$$

and

$$c_p(T) = J s_0 (s_0 + 1) \frac{\partial}{\partial T} \varepsilon_p(T), \quad (12)$$

respectively.

### III. RESULTS FOR THE SUSCEPTIBILITY AND THE SPECIFIC HEAT

In this section, we will present the main results of the model introduced in the previous section. One is tempted to compare them with the available experimental data. However, such a comparison makes no sense at this point because, as stated in the Introduction, there are several effects not included in this simplified model such as dipole-dipole interactions, anisotropies, or further neighbor interactions, to cite some examples. Therefore, we will present the results of the present model and study how the quantum case evolves

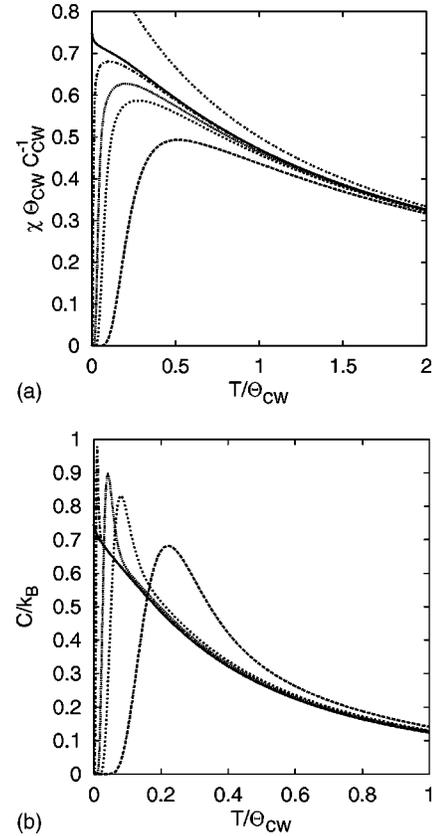


FIG. 2. Susceptibility and specific heat for the pyrochlore lattice for various values of  $s_0$ : (long-dashed line)  $s_0 = \frac{1}{2}$  (short-dashed line)  $s_0 = 1$ ; (dotted line)  $s_0 = \frac{3}{2}$ ; and (dot dashed line)  $s_0 = \frac{7}{2}$ . The solid line is the classical limit of the model. The double dotted line in the case of the susceptibility represents the Curie-Weiss law.  $\Theta_{\text{CW}} = 2(p-1)s_0(s_0+1)J/3$  is the Curie-Weiss temperature and  $C_{\text{CW}} = s_0(s_0+1)/3$  is the Curie constant. (a) Magnetic susceptibility. (b) Specific heat.

towards the classical limit for relatively large values of the individual spins. If the quantum behavior of the physical quantities of interest (namely, susceptibility and specific heat) is not so different from the classical one for relatively large values of  $s_0$ , we can feel confident that the predictions are essentially correct, at least in the temperature region where deviations from the classical behavior are small, as we know that the GCC model gives an accurate description of these quantities in the whole temperature range. It would be desirable also to compare the predictions of this model with the ones obtained from more sophisticated techniques (quantum Monte Carlo, high-temperature series expansions, density-matrix renormalization group, exact diagonalization, and so on). However, that comparison is not possible in general, due to the geometrical complexity of GFAP that prevents one from applying any of these techniques to arbitrary values of  $s_0$ , and these kind of calculations have been only carried out for the quantum  $s_0 = \frac{1}{2}$ , which, as we will see below, is the case where we expect to have the worst results.

Let us start by considering the results for the susceptibility and specific heat in the pyrochlore lattice, which can be seen in Fig. 2 for some values of  $s_0$ . As we can see from the

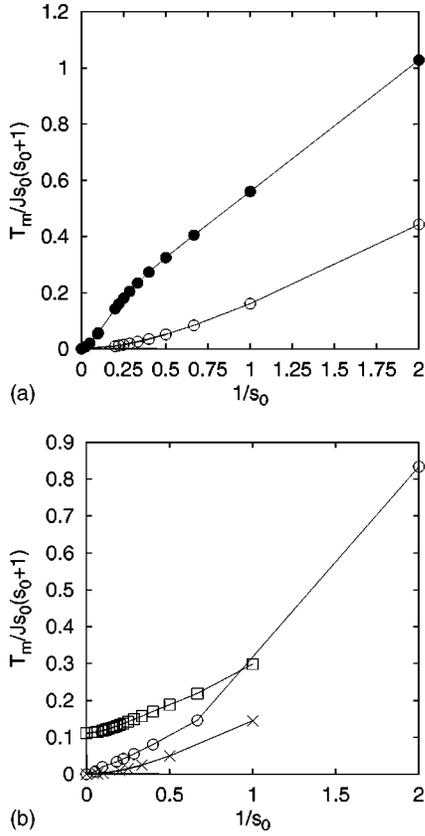


FIG. 3. Position of the maxima appearing in the susceptibility and specific heat. Solid lines are a guide to the eye. (a) Pyrochlore lattice. (●) Maxima in the susceptibility; (○) maxima in the specific heat. (b) *kagomé* lattice. (□) Maxima in the susceptibility, (○) maxima in the specific heat for half-integer  $s_0$  and (×) maxima in the specific heat for integer values of  $s_0$ .

observation of that figure, the classical susceptibility deviates from the Curie-Weiss behavior below  $\Theta_{CW}$ . The quantum case is qualitatively similar to the classical behavior except at very low temperatures, where it passes through a maximum and then falls to zero. However, we can also see that even for relatively small values of  $s_0$  the quantum behavior is not so different from the classical one down to the maximum. Regarding the predictions for the specific heat, again, the main difference is that it goes through a maximum, and later falls to zero, in contrast with the classical situation, where it goes to a constant value at  $T=0$  K. Additional information can be gained by studying the dependence of the position of the maxima with the value of  $s_0$  in Fig. 3. In that figure can be seen how nicely both the maxima in the susceptibility and specific heat go to the zero classical value.

Let us now turn to the *kagomé* lattice. In this case, it is reasonable to expect the model to work less well than in the pyrochlore lattice, as it is well-known that the role of long-wavelength fluctuations is more important in 2D lattices, which are explicitly neglected in any MF approach. However, from the MC results for the classical limit, it turns out that long-wavelength thermal fluctuations are unimportant for GFAF at finite temperatures, which partially explains why the classical limit of our model is essentially exact when

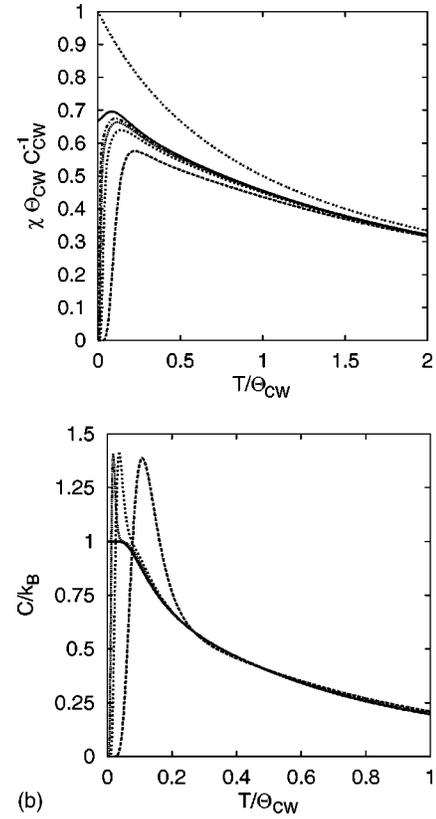


FIG. 4. Susceptibility and specific heat for the *kagomé* lattice with integer values of  $s_0$ : (long-dashed line)  $s_0=1$ ; (short-dashed line)  $s_0=2$ ; (dotted line)  $s_0=3$ ; and (dot dashed line)  $s_0=4$ . The solid line is the classical limit of the model. The double dotted line in the case of the susceptibility represents the Curie-Weiss law. (a) Magnetic susceptibility. (b) Specific heat.

compared to MC data for the susceptibility. The quantum case is a more delicate one, as quantum fluctuations can play an important role. However, these fluctuations will be only important at low temperatures, a region in which our model does not apply for other reasons we will explain below.

In order to proceed further, we will split the discussion for the *kagomé* lattice in two cases, corresponding to integer and half-integer values of the individual spins.

Let us first analyze the results for integer values of  $s_0$ , which can be seen in Fig. 4. There are no remarkable differences with respect to the discussion for the pyrochlore, except for the fact that the maximum in the susceptibility is present even in the classical limit. The only qualitative difference is the later fall to zero of both the susceptibility and specific heat. Also, we have computed the evolution of the maxima for both the susceptibility and specific heat. Again, both quantities go smoothly to their classical values. It is important to stress that the fact that the susceptibility goes to zero is related to the ground-state total spin of the cluster, which, for integer values of the individual spins, is always a singlet. This is not the case for half-integer values of  $s_0$ .

For half-integer values of  $s_0$ , there are very important differences with respect to the previous cases, as is evident in Fig. 5. The susceptibility goes through a maximum, then reaches a minimum, and diverges as  $T \rightarrow 0$ . This is a conse-

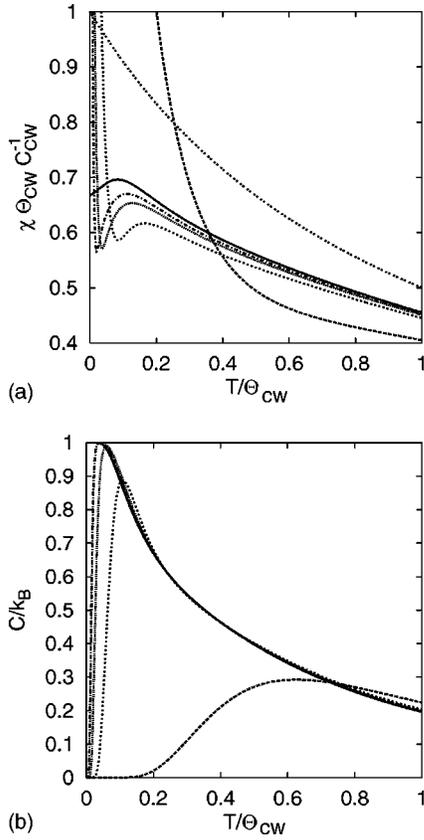


FIG. 5. Susceptibility and specific heat for the *kagomé* lattice with half-integer values of  $s_0$ : (long-dashed line)  $s_0 = \frac{1}{2}$  (short-dashed line)  $s_0 = \frac{3}{2}$ , (dotted line)  $s_0 = \frac{5}{2}$ , and (dot-dashed line)  $s_0 = \frac{7}{2}$ . The solid line is the classical limit of the model. The double dotted line in the case of the susceptibility represents the Curie-Weiss law. (a) Susceptibility per ion. (b) Specific heat per ion.

quence of the fact that the ground state of the triangular cluster for half-integer values of  $s_0$  is a doublet in our model. However, it is now generally accepted that the real ground state of the *kagomé* lattice is a singlet,<sup>20,21,27</sup> and may have a gap in the spectrum so, actually, the susceptibility should either be finite or go to zero as  $T \rightarrow 0$ . Obviously, the present breakdown at very low temperatures is not a particular feature of the mean-field approach, but it is shared by every model that starts from finite cluster calculations, with an odd number of spins in the elementary cluster. Moreover, the maxima in the specific heat follow a different trend than in the integer  $s_0$  case, even though the classical limit is correct (see Fig. 3). Even more striking is the fact that both the susceptibility and specific heat for  $s_0 = \frac{1}{2}$  are qualitatively different from all the previously presented results. The susceptibility does not show a maximum, but diverges as  $1/T$  as  $T$  approaches 0. The maximum in the specific heat does not follow the extrapolated behavior for other half-integer values of  $s_0$ . These kinds of problems have been already pointed out in the literature for more rigorous methods than ours.<sup>20,22</sup> A qualitative argument that can help to understand these deviations is the fact that  $s_0 = \frac{1}{2}$  is the “most quantum” case, in which the physical quantities are more sensitive to the discrete structure of the energy levels of the system. Taking into

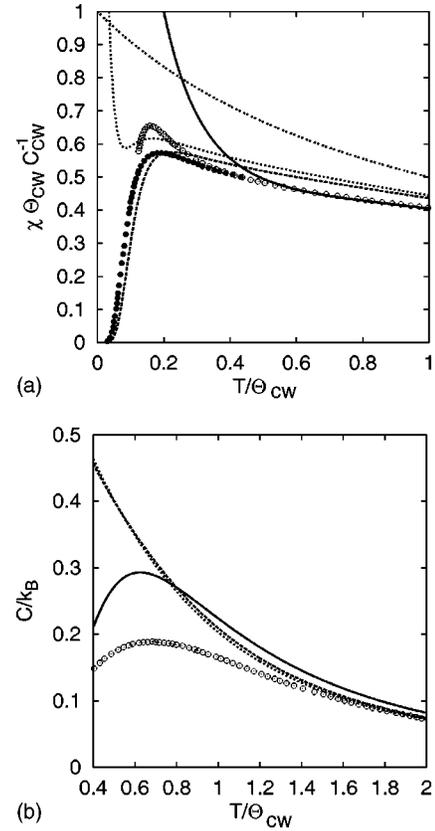


FIG. 6. Comparison of the GCC predictions with results from finite-size clusters calculations and high-temperatures series for the *kagomé*  $\frac{1}{2}$  lattice (Ref. 20) (solid line) GCC model for  $s_0 = \frac{1}{2}$ , (long-dashed line) GCC model for  $s_0 = 1$ , (dashed line) GCC model for  $s_0 = \frac{3}{2}$ , (○) results from high-temperature series expansions, and (●) results from exact diagonalization of a cluster with 18 spins. The double dotted line in the case of the susceptibility represents the Curie-Weiss law. (a) Susceptibility. (b) Specific heat.

account that for half-integer values of  $s_0$  the energy-level structure of our model is qualitatively incorrect, we should not be surprised by this special behavior.

As commented above, if we want to further check the range of applicability of the model, it would be desirable to compare its predictions with the ones obtained from more sophisticated methods, such as exact diagonalization of small clusters or high-temperature series expansions. Unfortunately, these calculations have been carried out only for the *kagomé* with  $s_0 = \frac{1}{2}$  (Refs. 20,21) where, as we have argued above, we can expect the model to be least successful. However, even though we do not expect complete agreement, we have carried out such a comparison with results obtained from the aforementioned methods and the results are presented in Fig. 6. In that figure we have also plotted the curves corresponding to  $s_0 = 1$  and  $s_0 = \frac{3}{2}$  for comparison at a qualitative level. It is important to stress that the curves depicted there do not contain any fitting parameter nor have they been rescaled in any sense. Surprisingly, the susceptibility calculated from our model for  $s_0 = \frac{1}{2}$  is in very good agreement down to  $\approx 0.5\Theta_{CW}$  with both high-temperature series expansions and the diagonalization of a cluster with 18

spins. The corresponding curve for the specific heat is in poor quantitative agreement with the results from high-temperature series expansions. However, it describes accurately the position of the maximum. In any case, we can expect the agreement with the specific heat to be worse, as it considers independent units, neglecting the emerging correlations at low temperatures. Moreover, we can see that the calculated susceptibilities for  $s_0=1$  and  $s_0=\frac{3}{2}$  reproduce the main qualitative features of the high-temperature expansion and finite cluster results. At this point, it is difficult to say if this a fortunate coincidence or an indication of the accuracy of our model for higher values of  $s_0$ . Numerical data for these values of the spins would be necessary to decide between the two cases.

In any case, it is important to notice that some of the qualitative features obtained in this simple model have been observed in experimental studies.<sup>1,28-33</sup> Again, it is difficult to say at this point if those features are due to additional interactions not accounted for in this model. For example, the existence of a maximum in the susceptibility for the pyrochlore lattice has been systematically observed in all the experiments at low enough temperatures. Actually, these maxima were successfully interpreted by the present authors with an even simpler model in which the interactions with nearest neighbors and next-nearest neighbors were taken into account.<sup>19</sup> Moreover, in the work by Wills *et al.*<sup>28</sup> on iron jarosites, which is one of the few systems where the *kagomé* lattice has an experimental realization, the magnetic susceptibility exhibits the maximum and a later upturn at a temperature comparable with that predicted by our model. However, in that temperature region, it seems likely that the dilution of the magnetic lattice by nonmagnetic impurities plays an important role, giving rise to some kind of spin-glass behavior, for which it is very difficult to extract any conclusion.

In the light of these results, we think that a prudent estimate of the breakdown of our model is set by the position of the maximum in the specific heat. As we approach this temperature, emerging correlations that cannot be described in the GCC formalism enter into play. For the pyrochlore lattice and the *kagomé* with integer values of  $s_0$ , it is very likely that the present description is qualitatively correct for temperatures well below the peak temperature of the specific heat. However, we think that the description below that tem-

perature for half integer  $s_0$  in the *kagomé* lattice is incorrect. Probably, there is a maximum and a minimum in the susceptibility, but we expect the susceptibility either vanishes or goes to a finite value at  $T\rightarrow 0$ , not to diverge. In any case, it is still a significant improvement with respect to the Curie-Weiss theory and other MF theories. However, more rigorous techniques are needed in order to verify these assertions.

#### IV. CONCLUSIONS

In this work we have presented the quantum version of the generalized constant-coupling model that was shown by the authors to be in excellent agreement with Monte Carlo data for the susceptibility and specific heat in the classical limit for both the pyrochlore and *kagomé* lattices. There are some important qualitative differences between the classical and quantum behaviors that are important at low temperatures.

The main features of the susceptibility in this model are the presence of maxima in both the susceptibility and specific heat, similar to those found experimentally. The susceptibility for the *kagomé* lattice with half-integer values of the individual spins is found to pass through a maximum and then, after a minimum, diverges as  $T\rightarrow 0$ . This divergence is due to a nonzero value of the total spin of the ground state of the elementary units for these values of the individual spins. There is some experimental evidence for the existence of such an upturn in the susceptibility in the iron jarosite systems.<sup>28</sup> However, the divergence of this quantity in our model that is due to the finite-size cluster with an odd number of spins is incorrect.

The results for the *kagomé*  $\frac{1}{2}$  are compared with high-temperature series expansions and exact diagonalization of small clusters. The susceptibility is found to be in good agreement with those results down to  $T\approx 0.5\Theta_{CW}$ , which is a remarkable fact for so simple a model. Even though the specific heat is not in quantitative agreement at this temperature, the position of the maximum is adequately predicted. Results from the aforementioned techniques for these lattices for higher values of  $s_0$  would be desirable in order to check the accuracy of the GCC model.

In any case, we feel that the present model provides an adequate description of the nearest-neighbor *kagomé* and pyrochlore systems down to the temperature of the peak in the specific heat.

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