Random site dilution properties of frustrated magnets on a hierarchical lattice

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Abstract

We present a method to analyze the magnetic properties of frustrated Ising spin models on specific hierarchical lattices with random dilution. Disorder is induced by dilution and geometrical frustration rather than randomness in the internal couplings of the original Hamiltonian. The two-dimensional model presented here possesses a macroscopic entropy at zero temperature in the large size limit, very close to the Pauling estimate for spin-ice on the pyrochlore lattice, and a crossover towards a paramagnetic phase. The disorder due to dilution is taken into account by considering a replicated version of the recursion equations between partition functions at different lattice sizes. An analysis to first order in replica number allows a systematic reorganization of the disorder configurations, leading to a recurrence scheme. This method is numerically implemented to evaluate thermodynamical quantities such as specific heat and susceptibility in an external field.

(Some figures may appear in colour only in the online journal)

1. Introduction

Hierarchical lattices possess some interesting features of geometrical scale invariance, and several methods have been developed extensively in the past to study thermodynamical properties of spin models considered on such structures [1, 2]. The similarity to the Migdal–Kadanoff renormalization procedure for magnetic spin systems [3], with exact recursion relations between coupling constants at the successive stages of lattice construction, makes such models suitable for the study of critical properties. Indeed, phase transitions in these systems have non-mean-field critical exponents, effective dimensions and frustration effects within local loops. However, the absence of translation and the site dependent connectivity inherent to such a theoretical construction make the physics apparently different from real systems. The importance of exact solutions in disordered lattices are highlighted by the possibility to obtain controllable recursion relations which gives interesting properties for Ising spin glass and quenched disorder models [4, 5], or disordered Potts models [6]. Multicritical point locations can moreover be checked carefully using Nishimori symmetric lines [7–9] and duality exact properties [10]. Frustration on hierarchical lattices has been studied for example on a diamond shape ferromagnetic structure with an additional transverse antiferromagnetic coupling between two spins [11]. Important properties of the phase diagram when the coupling is increased concern the low-temperature entropy, which presents steps at specific values of this coupling, reflecting the frustration character of the lattice geometry. These transitions are generated by slight variations of the transverse coupling inducing a change in the nature of the ground states. Recursion relations can be found exactly in this non-disordered but frustrated model, hence equations for ground state structure, residual entropy and magnetization can be obtained using scaling properties of the hierarchical geometry.

Experimental realizations of frustrated magnetic structures with spin dilution can be found in spin-ice materials doped with rare earth elements, such as Dy$_{2-x}$Lu$_x$Ti$_2$O$_7$, Dy$_{2-x}$Y$_x$Ti$_2$O$_7$, and Ho$_{2-x}$Y$_x$Ti$_2$O$_7$ [12], where $x$ parametrizes the randomness [13]. These compounds are obtained from primary materials Dy$_2$Ti$_2$O$_7$ or Ho$_2$Ti$_2$O$_7$. 

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where magnetic ions Dy$^{3+}$ and Ho$^{3+}$ occupy the sites of a pyrochlore lattice made of tetrahedra connected to each other by their vertices. The first magnetic rule for each individual tetrahedron is given by two spins out of the tetrahedron and two spins in, known as the first ice rule [14]. These magnetic ions are then replaced by non-magnetic atoms Y or Lu, giving rise to an experimental system of site dilution where a macroscopic fraction of sites is non-magnetic. One of the interesting physical properties of these systems is the non-monotonic behavior of the zero-temperature entropy with respect with the dilution level [13] from the measured specific heat, which can be explained quite accurately within the Pauling approximation, where tetrahedra are treated as independent [13]. Refined calculations with Monte Carlo simulations on a Husimi cactus [15] is a step beyond the model of Pauling which takes into account the correlations between tetrahedra on the pyrochlore lattice [16]. In the Bethe–Peierls approximation, a tree-like structure is constructed from a central site, where the number of branches grows with the distance from the central site. However, in this geometrical approach, the branches never reconnect together, a construction which forbids geometrical loops, but recursion equations can in general be written in a systematic manner. In this paper, we address the question of site disorder importance on thermal properties in the same spirit as [11], where frustration plays an important role.

We indeed consider explicitly an antiferromagnetic tetrahedron-like hierarchical structure with Ising spins $\sigma_i = \pm 1$ located on the vertices $i$. Disorder configurations are given by a set of additional quenched random variables $\epsilon_i = 0, 1$, with probability $x$ and $(1-x)$ respectively, located on the same sites (vertices) $i$, as shown in figure 1. The replicative procedure to construct the lattice iteratively is implemented starting from one single antiferromagnetic bond at level $r = 0$, until level $r > 0$ is reached, as exemplified on figure 1 for the first two steps. Such a structure is known to have a well defined thermodynamical limit, when $r$ is large, for the free energy per site [2, 17]. The zero-temperature entropy of the tetrahedron-like structure at level $r = 1$ can be evaluated exactly, as we will see in the following sections and it appears to be non-monotonic as a function of the dilution.

We can then generalize the calculation for larger structures $r > 1$ using recursion equations in the replica space, and obtain quantitative information about frustration and disorder effects in the thermodynamical limit. This simple model can be considered as a good example on how to characterize spin-ice or spin-liquid states and how these states may emerge, for example, from the specific heat or susceptibility measurements.

2. Notations and method

We consider the hierarchical lattice seen as a graph construction (set of vertices connected here by links or edges), built recursively an arbitrary number of times. Vertices can then be occupied by a spin with probability $(1 - x)$, with $0 \leq x \leq 1$, or stay vacant with probability $x$, which is the dilution factor. Initially, at step $r = 0$, the graph consists of a single antiferromagnetic link, as displayed in figure 1(a), with two vertices occupied eventually by spins. This link has a Boltzmann strength $K = J/T$ with coupling unity, $J = 1$. The structure at the next step is formed by replicating the previous structure four times, and by connecting the lower and upper vertices together, as in figures 1(b), (c) and in general (d). The diamond shape structures that are obtained are connected two by two to form a larger graph. Additionally, two transverse antiferromagnetic bonds of value $K' > 0$ (see figures 1(b)–(d)) are added at each step, in order to enhance frustration and to mimic tetrahedral structures. For example, step $r = 1$ is obtained by connecting four single edges to form a square, on which two antiferromagnetic edges of value $K'$ are added transversely and longitudinally.

The spins considered in this paper are Ising spins $\sigma_i = \pm 1$ on each site (vertex) $i$, and we use an additional set of variables $\epsilon_i = 0$ when the spin is vacant, with probability $x$, or $\epsilon_i = 1$ when it is present, with probability $1 - x$. It is straightforward to compute recursively the total number of vertices $N_r$ at level $r$.

![Figure 1](image-url)

**Figure 1.** Construction of the hierarchical lattice, starting from a single link $K > 0$ between two spins (a). This link is replicated to form a diamond shape, and couplings $K' > 0$ are added successively between boundary sites, see (b) and (c), to form a frustrated lattice. Shaded areas in (d) define the previous lattice structures at level $r - 1$, with bonds $K'$ added furthermore between the boundary sites.
\[ N_0 = 2, \quad N_1 = 4, \quad N_{r+1} = 4N_r - 4, \quad N_r = \frac{4^{r+1} + 8}{6}. \tag{1} \]

Also the total number of bonds \( B_r \) is equal to \( B_r = 4^r + 2^{r+1} - 2 \), and the maximum number of bonds connecting the two upper and lower sites is equal to \( L_r = 2^r \). We may define an effective dimension \( d \) such that \( B_r = L_d^r \), which gives \( d = 2 \) in the large size limit \( r \to \infty \). In the following we will mainly concentrate on the frustrated case \( K' = K \). The partition function for this dilute hierarchical lattice made of antiferromagnetic bonds can be constructed starting at step \( r = 0 \) using the following standard method. Also an external magnetic field \( h_x \) is taken into account in order to evaluate the spin susceptibility and magnetization.

After \( r \) recurrences, we define the partial partition function \( Z_r^{[\eta]}(\epsilon\sigma, \epsilon'\sigma') \), which depends on some disorder configuration \( \eta \) given by a vacancy distribution set \( \{ \epsilon_i = 0, 1 \} \), with \( i \) corresponding to the sites located inside the structure delimited by the two extreme sites \( (\epsilon, \epsilon') \). These latter sites have a given disorder configuration \( (\epsilon, \epsilon') \) which does not belong to \( \eta \). A recursion relation can be written between steps \( r \) and \( r + 1 \) of the lattice construction

\[
Z_{r+1}^{[\eta]}(\epsilon\sigma, \epsilon'\sigma') = \text{Tr}_{\sigma_1, \sigma_2} Z_r^{[\eta]}(\epsilon\sigma, \epsilon_1 \sigma_1) Z_r^{[\eta]}(\epsilon_1 \sigma_1, \epsilon'\sigma') \times \exp \left( -K'\epsilon_1 \sigma_1 \epsilon_2 \sigma_2 + \frac{h_x}{T} (\epsilon_1 + \epsilon_2) \right) \times \exp(-K\epsilon\epsilon') 2^{\epsilon_1 - 2} 2^{\epsilon_2 - 1} \tag{2} \]

where \( \eta \) represents the disorder configuration \( \eta = \{ \eta_1, \eta_2, \eta_3, \eta_4, \epsilon_1, \epsilon_2 \} \) and the Tr signifies the sum over all the possible spin values. The additional factors \( 2^{\epsilon_1 - 2} 2^{\epsilon_2 - 1} \) compensate the fact that we sum over eventual ghost spins located on vacant sites. The initial condition figure 1(a) is obtained from the two-spin partition function

\[
Z_0^{[\eta]}(\epsilon\sigma, \epsilon'\sigma') = \exp(-K\epsilon\epsilon' \sigma\sigma') \tag{3} \]

which has actually no internal disorder configuration, \( \eta = \emptyset \). The magnetic field is implemented in (2) only for spins not belonging to the top and bottom vertices. Using the replica method, we consider \( n \) copies of (2) for a given configuration \( \eta \) and perform the average

\[
Z_{r+1}^{[\eta]}(\epsilon\sigma, \epsilon'\sigma') \equiv \left[ \prod_{\alpha=1}^{n} Z_{r+1}^{[\eta]}(\epsilon\sigma, \epsilon'\sigma') \right] \eta. \tag{4} \]

Here \[ ]_n stands for averaging over disorder \( \eta \). In this case, the recursion relation (2) becomes

\[
Z_{r+1}(\epsilon\sigma, \epsilon'\sigma') = \exp \left( -K\epsilon\epsilon' \sum_{\sigma} \sigma\sigma' \right) \times \int P(\epsilon_1)P(\epsilon_2) d\epsilon_1 d\epsilon_2 \text{Tr}_{\sigma_1'\sigma_2'} \times Z_r(\epsilon\sigma_1, \epsilon_2\sigma_2) Z_r(\epsilon_2\sigma_2, \epsilon'\sigma') \times \exp \left[ -K\epsilon_1 \epsilon_2 \sum_{\alpha} \sigma_1^{\alpha} \sigma_2^{\alpha} \right] + \frac{h_x}{T} \sum_{\alpha} (\sigma_1^{\alpha} + \sigma_2^{\alpha})^2 2^{\epsilon_1 - 1} 2^{\epsilon_2 - 1} \tag{5} \]

where \( P \) is the bimodal distribution for the dilute sites \( P(\epsilon) = x\delta(\epsilon) + (1-x)\delta(\epsilon - 1) \). In the previous expression, we perform only the sum over the two spins \( \sigma_1^{\alpha} \) and \( \sigma_2^{\alpha} \) connecting pairs of diamond shape structures. Starting with the initial condition

\[
Z_0(\epsilon\sigma, \epsilon'\sigma') = \exp \left( -K\epsilon\epsilon' \sum_{\sigma} \sigma\sigma' \right) \tag{6} \]

all quantities \( Z_r(\epsilon\sigma, \epsilon'\sigma') \) can in principle be computed by step by step. Free energy \( F(r) \) is evaluated directly after averaging over the remaining disorder and summation over the two spin variables. We define the complete partition function as

\[
z_r(n) = \int P(\epsilon)P(\epsilon') d\epsilon' d\epsilon 2^{\epsilon_1 - 1} 2^{\epsilon_2 - 1} \text{Tr}_{\sigma_1\sigma_2} \times Z_r(\epsilon\sigma, \epsilon'\sigma') \exp \left[ \frac{h_x}{T} \sum_{\alpha} (\sigma_1^{\alpha} + \sigma_2^{\alpha}) \right] \tag{7} \]

and take the limit \( n \to 0 \)

\[
-KF(r) \equiv \lim_{n \to 0} \frac{z_r(n) - 1}{z_r(1)} = z_r'(0) \tag{8} \]

after noticing that \( z_0(0) = 1 \). The disorder averaged entropy can be expressed in terms of \( z_r'(0) \) via the usual thermodynamical relations

\[
S(r)(T) = -\frac{\partial F(r)}{\partial T} = z_r'(0) - K \frac{\partial}{\partial K} z_r'(0) \tag{9} \]

as well as the averaged specific heat \( C_v^{(r)} = -T^2 \frac{\partial F(r)}{\partial T^2} = K^2 z_0^{(r)}/(\partial K)^2 \). The relation between entropy and specific heat is given by the integral

\[
S(r)(T) = N_x(1 - x) \ln(2) - \int_T^\infty \frac{C_v^{(r)}}{T^2} dT \tag{10} \]

which is a direct experimental way to measure the zero-temperature entropy by extrapolation, knowing that all spins are in the paramagnetic phase at high temperature, each contributing with a factor \( \ln(2) \). The linear susceptibility corresponds to the excitation of the magnetic order parameter \( M = \sum_{\sigma} \epsilon \) under a field \( h_x \) and is defined as \( \chi^{(r)} = \partial z[M]/\partial h_x \), after averaging over disorder. We can also relate \( \chi^{(r)} \) to the free energy, using \( T \chi^{(r)} = [\langle M^2 \rangle]_n - [\langle M \rangle]^2_n \) and \( [\langle M \rangle]_n = -\partial F^{(r)}/\partial h_x \), in particular

\[
\chi^{(r)} = \frac{\partial z[M]_n}{\partial h_x} = -\frac{\partial^2 F^{(r)}}{\partial h_x^2}. \tag{11} \]

3. Residual entropy for \( r = 1 \) structure

As a simple application, we consider the case \( r = 1 \) consisting of only four spins figure 1(b), where all thermodynamical
quantities can be computed exactly. The zero-temperature entropy is identical to the entropy of a three-dimensional tetrahedron made of antiferromagnetic bonds. After iterating (5), we obtain

\[ Z_1(\epsilon \sigma^a, \epsilon' \sigma^a') = \exp \left( -K' \epsilon' \sum_a \sigma^a \sigma^a' \right) \left[ (1 - x)^2 e^{2\rho} - 2 \frac{h_e}{T} \right] \]

\[ + 2x(1 - x)^2 \sum_a \cosh \left( K(\epsilon \sigma^a + \epsilon' \sigma^a') - \frac{h_e}{T} \right) \]  

(12)

Then the function \( z_1(n) \) is equal, after some computation, to

\[ z_1(n) = (1 - x)^4 2^n \left[ 1 + \cosh(4K) \right. \]

\[ + e^{-4K} \sinh^2 \left( \frac{h_e}{T} \right) \left( e^{2K} \right. \]

\[ + e^{2K} + \cosh \left( \frac{h_e}{T} \right) \left( e^{2K} \right. \]

\[ + 4x(1 - x)^3 4^n \left[ \cosh \left( \frac{h_e}{T} \right) e^{2K} \right. \]

\[ + \left. \cosh(2K) \cosh \left( \frac{h_e}{T} \right) \cosh \left( \frac{2h_e}{T} \right) \right] \]

\[ - \sinh(2K) \sinh \left( \frac{h_e}{T} \right) \sinh \left( \frac{2h_e}{T} \right) \right] \left( e^{-K} \right)^n \]

\[ + 2x^2(1 - x) 2^n \left[ \cosh \left( \frac{h_e}{T} \right) e^{K} \right. \]

\[ \left. + 2 \left( \cosh \left( \frac{h_e}{T} \right) e^{-K} + e^{K} \right)^2 \right] \]

\[ + 4x^3(1 - x) 2^n \cosh \left( \frac{h_e}{T} \right) + x^4. \]  

(13)

When \( K \) is large and in absence of external field, this expression can be put into the following form, by keeping the dominant contributions in the exponential terms

\[ z_1(n) = \sum_{k \geq 0} \rho_k e^{n(s_k - K \epsilon_k)}, \quad \sum_{k \geq 0} \rho_k = 1. \]  

(14)

This expansion is useful in order to identify the zero-temperature entropy which can be written as a sum of contributions \( S^{(1)}(T = 0) = \sum_k \rho_k s_k \). Each partial entropy \( s_k \) and energy \( e_k \) physically corresponds to a configuration of disorder with weight \( \rho_k \). In this limit, the asymptotic form for \( z_1(n) \) when \( 0 \leq K' < K \) is indeed equal to

\[ z_1(n) \simeq (1 - x)^4 2^n e^{4K - 2K'} + 4x(1 - x)^3 2^n e^{2K'} \]

\[ + 4x^2(1 - x)^2 2^n e^{K'} + 2x^2(1 - x) 2^n e^{K'} \]

\[ + 4x^3(1 - x) 2^n + x^4. \]  

(15)

After taking the limit \( n \rightarrow 0 \), the entropy and energy per spin are respectively equal to \( S^{(1)}(T = 0)/4(1 - x) = (1 - x^4)/[4(1 - x)] \ln(2) \) and \( E^{(1)}/[4(1 - x)] = -(1 - x)^2(1 - \gamma/2) - x(1 - x)^2(2 - \gamma) - x^2(1 - x)(1 + \gamma/2) \), where \( \gamma := K'/K \) is the coupling ratio. The frustrated case \( K' = K \) is particular since we obtain instead

\[ z_1(n) \simeq (1 - x)^4 6^n e^{2K} + 4x(1 - x)^3 6^n e^{K} \]

\[ + 6x^2(1 - x)^2 2^n e^{K'} + 4x^3(1 - x)^2 + x^4. \]  

(16)

and, in this specific case, the entropy and energy per spin have a different expression

\[ S^{(1)}(T = 0) = \frac{1 - x}{4(1 - x)} \ln(6) + \frac{1}{4} x(1 - x)^2 \ln(6) \]

\[ + 3x^2(1 - x) \ln(2) + x^3 \ln(2), \]

\[ E^{(1)}(T = 0) = -\frac{1}{2} (1 - x)^3 - x(1 - x)^2 - \frac{3}{2} x^2(1 - x). \]  

(17)

As expected, the entropy for a single tetrahedron in the frustrated case is larger. The plot of the entropy is given in figure 5 as function of \( x \) and appears to be non-monotonic, with a local maximum around \( x = 0.295 \). This maximum, intrinsic to a four-spin system or single uncoupled tetrahedra, is probably due to the non-monotonic variation of the fraction of the spin configurations with the lowest energy as function of dilution in a system of four spins where there are at most 16 possible states, in a similar way to that described in [13] for coupled tetrahedra. This maximum disappears for \( r > 1 \), as shown on the same figure, because the varying connectivity and coupling arrangement on the hierarchical structure tends to modify the fractions of these acceptable states in the different tetrahedral units. The exact entropy value in the absence of dilution is given by \( \ln(6)/4 \simeq 0.4479 \), which is larger than for a system of coupled tetrahedra in the large size limit (see sections below).

Susceptibility \( \chi^{(1)} \) is derived exactly using the definition (11) and is plotted in figure 2 as function of the external field for different values of \( x \). As expected, when \( x > 0 \), a peak appears at \( h_e = 0 \) when the probability to find a single spin in the tetrahedron is nonzero. A simple calculation for the non-disordered case leads to three possible ground states per spin \( E_G \), depending on the value of the field: two spins up and two spins down with \( E_G = -2h_e \), and one spin up and one spin down with \( E_G = -2h_e \), and all spins up with \( E_G = 6 - 4h_e \). Two transitions occur respectively at \( h_e = 1 \) and \( h_e = 3 \).

For larger sizes however, it is interesting to analyze the susceptibility using recursive equations (5) for the partition function \( Z_r(\sigma, \sigma') \) in the absence of disorder \( x = 0 \). The recursive equations are detailed in appendix A. The method is to assume an effective form for the partition function, with Ising effective coupling, magnetic field, and weight coefficient at all recursion levels. The exact recurrence equations for these three quantities as function of the external magnetic field \( h_e \) allow us directly to compute the susceptibility. Plots of \( \chi^{(r=6)}(h_e) \) and magnetization per spin \( m_r^{(r=6)}(h_e) \) are displayed in figure 3, showing multiple field transitions and magnetization plateaus up to \( h_e = 127 \) for \( N_6 = 2732 \) spins. In section 4, the analysis is extended to the dilute case. We use the set of recursive equations (5) to compute the
partition function at a given step \( r \). The main technique is to reorganize the partition function in the same manner as (16), where we easily identify weights \( \rho_k = x^k (1-x)^{n-2} C_{n_r}^k \).

One difficulty with dilution is that an iterative procedure at low temperature on effective Ising constant couplings to obtain the limiting coupling distribution can in principle be performed, but not for the free energy, unlike the case of spin glass models. For example, for the disordered Potts model [6], where the disorder is given by a discrete distribution of couplings, zero-temperature free energy satisfies some recursive equations. In general, when the disorder is located only in the couplings, the four structures represented as shaded areas in figure 1(c) have independent internal disorder, even if they are connected together by the four vertices. Therefore disorder can be factorized and direct iteration of Ising couplings by renormalization can be performed, as well as the partition function weights such as \( I_r \) in (A.1), which are important for the determination of free energy and entropy. In the case of dilution, however, the disorder configuration in the boundary vertex sites given by set \( \{ \epsilon, \epsilon', \epsilon_1, \epsilon_2 \} \) is actually shared between the four diamond structures. For example, variable \( \epsilon_1 \) is common to the two shaded diamonds on the left hand side of figure 1(c). This makes the evaluation of weight coefficients \( I_r \) (A.1) problematic in the disordered case, since after few iterations correlations will develop rapidly. This is main reason why it is more convenient to consider a replica version of (A.1) and (2) with partial integration over the disorder located strictly inside the diamond structures, as explicitly defined previously by (5). As we will see, an approximation scheme can be derived from the replica method and thermodynamical functions can be obtained.

4. Recursion relations in the general case

At step \( r \), we assume from the previous analysis an expansion of the partition function in terms of configurational weights \( x^k (1-x)^{n_r-2} C_{n_r}^k \), where \( n_r \) is here the number of sites localized between the two extreme top and bottom sites and which satisfies the equation \( n_{r+1} = 4n_r + 2 \), with initial condition \( n_0 = 0 \) and \( N_r = n_r + 2 \). The main idea, as discussed before, is to keep a generic and minimal expression for the partial partition function, which depends on unknown functions satisfying recurrence equations such that

\[
Z_r(\epsilon \sigma^\alpha, \epsilon' \sigma'^\alpha) = \exp \left( -K' \epsilon' \sum_{\alpha} \sigma^\alpha \sigma'^\alpha \right) \sum_{n=0}^{n_r} \chi^k (1-x)^{n_r-k} C_{n_r}^k \\
\times \exp \left( nH^{(r)}_0 (\epsilon, \epsilon') + K'_r \epsilon' \sum_{\alpha} \sigma^\alpha \sigma'^\alpha \right) + H^{(r)}_k (\epsilon, \epsilon') \sum_{\alpha} \left[ \epsilon \sigma^\alpha + \epsilon' \sigma'^\alpha \right] \right)
\]

(18)

where \( H^{(r)}_r (\epsilon, \epsilon') \) and \( I^{(r)}_0 (\epsilon, \epsilon') \) are symmetric functions of \( \epsilon \) and \( \epsilon' \). Coupling \( K'_r \) is independent of the boundary disorder. All these functions depend implicitly on \( K' \) and \( h_r \). As initial condition we impose \( K^{(0)}_0 = -K + K' \) and \( H^{(0)}_0 (\epsilon, \epsilon') = I^{(0)}_r (\epsilon, \epsilon') = 0 \). We also take \( K' = K \) in the following for the frustrated version. At the next level, \( Z_{r+1} \) is evaluated by considering the product of four partition functions \( Z_r \), as written in (5).

Using the ansatz (18), this product is expanded as

\[
Z_{r+1} = \exp \left( -K' \epsilon' \sum_{\alpha} \sigma^\alpha \sigma'^\alpha \right) \times \sum_{k_1,k_2,k_3,k_4=0}^{n_r} C_{n_r}^{k_1} C_{n_r}^{k_2} C_{n_r}^{k_3} C_{n_r}^{k_4} x^{k_1+k_2+k_3+k_4} \\
\times (1-x)^{n_r-k_1-k_2-k_3-k_4} \\
\times \int \int P(\epsilon_1) P(\epsilon_2) \, d\epsilon_1 \, d\epsilon_2 \, Tr \sigma^\alpha \sigma'^\alpha
\]
order in $n$, and perform an expansion at first addition, we impose the constraint

$$K' = \sum_\alpha (\epsilon_1 \sigma_1^\alpha + \epsilon_2 \sigma_2^\alpha \epsilon_\alpha + \epsilon' \sigma_\alpha')$$

$$\times \exp \left( -\frac{2\pi(n-1) \epsilon(n-2)}{2} \right) \exp \left( K'^{(r)} (H_{1(r)} \sum_\alpha \epsilon_\alpha \sigma_\alpha + \epsilon_1 \sigma_1^\alpha) + H_{2(r)} \sum_\alpha \epsilon_\alpha \sigma_\alpha + \epsilon_1 \sigma_1^\alpha \right)$$

$$\times \exp \left( H_{3(r)} \sum_\alpha \epsilon_\alpha \sigma_\alpha + \epsilon_2 \sigma_2^\alpha \right)$$

$$\times \exp \left( H_{4(r)} \sum_\alpha \epsilon_\alpha \sigma_\alpha + \epsilon_2 \sigma_2^\alpha \right)$$

$$\times \exp \left( \frac{h_0}{T} \sum_\alpha (\epsilon_1 \sigma_1^\alpha + \epsilon_2 \sigma_2^\alpha) \right).$$

The last term takes into account the missing field on former boundary spins $\sigma_1^\alpha$ and $\sigma_2^\alpha$, which are now summed up. It is useful to introduce the operator $1 = \sum_{k=0}^{4n_r} \delta_{k,k_1+k_2+k_3+k_4}$ or the integral form

$$1 = \int_0^{4n_r} \frac{2\pi}{2\pi} \exp(-v(k_1+k_2+k_3+k_4))$$

which is then inserted in the previous expression in order to reorganize the sum over the $k$ into a single sum over weights $\chi^k(1-x)^4n_r-k \chi^k$. The integral over 0 can be performed using a first-order expansion in $n$, sufficient to obtain $\alpha'(0)$. For example, given integer $p > 1$ and $0 \leq k \leq pn_r$, let us consider $n_r+1$ field variables $\varphi_i$ and define the quantity $W_{n,k} \equiv \exp(p \varphi_i)$ made of the product of $p$ sums $\sum_k \chi^k \exp(p\varphi_i)$, $i = 1, \ldots, p$ by analogy with the product of sums that appears in (19). In addition, we impose the constraint $\sum_{i=1}^p \varphi_i = k$ by using the Kronecker integral (20), and perform an expansion at first order in $n$:

$$W_{n,k} \equiv \left( \int_0^{2\pi} \frac{2\pi}{2\pi} \exp(-v(k_1+k_2+k_3+k_4)) \prod_{i=1}^p C_{n_i} \exp(p\varphi_i) \right)^k$$

$$\simeq \int_0^{2\pi} \frac{2\pi}{2\pi} \exp(-v(k_1+k_2+k_3+k_4)) \prod_{i=1}^p C_{n_i} \exp(p\varphi_i)$$

$$\simeq \int_0^{2\pi} \frac{2\pi}{2\pi} \exp(-v(k_1+k_2+k_3+k_4)) \prod_{i=1}^p C_{n_i} \exp(p\varphi_i)$$

$$\simeq \int_0^{2\pi} \frac{2\pi}{2\pi} \exp(-v(k_1+k_2+k_3+k_4)) \prod_{i=1}^p C_{n_i} \exp(p\varphi_i)$$

$$\simeq \int_0^{2\pi} \frac{2\pi}{2\pi} \exp(-v(k_1+k_2+k_3+k_4)) \prod_{i=1}^p C_{n_i} \exp(p\varphi_i)$$

(21)

The exponentiation in the last line allows us, at first order in $n$, to reorganize the product of the $p = 4$ sums in (5) as a single sum over configurations of $k$ vacant sites, with combinatorial factor $C^k_{pn_n}$. In particular, taking a constant value $\varphi_i \equiv \varphi$, we easily find $W_{n,k} \equiv C^k_{pn_n} e^{p\varphi}$, and therefore the identity

$$W_{n,k} \equiv C^k_{pn_n}$$

(22)

from which we deduce that the factors

$$D^k_{n_r, p} \equiv \frac{C^k_{pn_n}}{C^k_{pn_n}}$$

(23)

can be considered as natural weights since the sum over integers $k_1$ is normalized. We may apply this technique to the spin operators $\sum_\alpha \sigma_\alpha \sigma_\alpha$ appearing in (19) as well, which are the sum of $n$ terms. Considering, for example, linear spin operator $\sum_\alpha \sigma_\alpha$, and instead of (21) the function

$$W_{n,k} \equiv \left( \int_0^{2\pi} \frac{2\pi}{2\pi} \exp(-v(k_1+k_2+k_3+k_4)) \prod_{i=1}^p C_{n_i} \exp(p\varphi_i) \right)^k$$

(24)

We then take the limit $n \to 0$ and define $w_k \equiv \partial W_{n,k} \equiv \partial W_{n,k} \partial n$, so that

$$w_k \equiv \left( \int_0^{2\pi} \frac{2\pi}{2\pi} \exp(-v(k_1+k_2+k_3+k_4)) \prod_{i=1}^p C_{n_i} \exp(p\varphi_i) \right)^k$$

(25)

We can compare this expression with the approximation

$$w_k \equiv \tilde{w}_k \equiv C_{pn_n} \ln \left[ 2 \cosh \left( \sum_{i=1}^p \varphi_i \right) \right]$$

(26)
coming from the same analysis made in (21), we can discuss two different cases. First, let choose $\varphi_i := \varphi$, both functions $w_k$ and $\tilde{w}_k$ are identical, using the normalization (22). Then, we may try non-constant fields such as $\varphi_i \equiv \varphi_i$, which gives after summation the exact result

$$w_k[\varphi_i = \varphi_i] = C_{\rho m}^k \ln[2 \cosh(k K \varphi)],$$

which is also identical to $\tilde{w}_k$ in (26) using equality

$$\sum_{k} D_{n_k}^{(a)} D_{n_k}^{(b)} k_1 = k/p.$$  

In the more general case, when the arguments $\varphi_i \leq \varphi_i$, are random variables, we can try to evaluate the accuracy of (26). Let us consider, for example, a Poisson distribution for the $\varphi_i > 0$, with mean and variance unity, prob($\varphi_i = \exp(-\varphi_i)$, and $K$ fixed. A measure of the accuracy can be given by the relative error function

$$g(\varphi_i) := \langle (\tilde{w}_k[\varphi_i]/w_k[\varphi_i] - 1)^2 \rangle^{1/2},$$

where the brackets are the average over random realizations. In figure 4 we present $g(\varphi_i)$ as function of $k$ for $n = 10$ and for different temperatures $1/K$.

Applying (21) for spin operators will be useful to simplify expression (19) and obtain recursive equations in the presence of dilution. Computing recursive equations for $H_k^{(t)}(\epsilon, \epsilon')$, $I_k^{(t)}(\epsilon, \epsilon')$, and $K_0^{(t)}$ follows two steps: the integral over variable $\theta$ coming from the constraint $k_1 + k_2 + k_3 + k_4 = k$, with $k_i = 0, \ldots, n_r$, introduced in (19), is performed using transformation (21) over the different spin operators with $p = 4$. This will allow the partial summation over spins $\sigma_1^x$ and $\sigma_2^x$, and averaging over random variables $\epsilon_1$ and $\epsilon_2$, see figure 1(c). We obtain an expression for $Z_{r+1}$ as a summation over configurations $\chi^{r+1}(1 - x)^{n_r + 2} C_4^{n_r} C_2^I$, with $4n_r$ sites coming from inside the shaded diamond structures in figure 1(c), plus the two sites coming from the integration over $\epsilon_1$ and $\epsilon_2$. This sum can be furthermore reorganized by again using identity (21) in order to finally obtain (18) at level $r + 1$ as a sum over weights $\chi^{r+1}(1 - x)^{n_r + 2} C_4^{n_r} C_2^I$ (with $4n_r + 2 = n_r+1$) and new coupling values. All details of this development are presented in appendix B, where recurrence equations are written explicitly in (B.11).

Using ansatz (18), and integrating over the boundary site degrees of freedom, the complete partition function (7) is then equal to

$$Z_{r}(n) = \sum_{k=0}^{n_r} C_{n_r}^k \chi^{r+1}(1 - x)^{n_r - k} \left[ \left(1 - x\right)^2 \left(\ln(2) + \left(\frac{2}{T} \ln(2) + \frac{2}{T} \left(2H_0^{(t)}(1, 1) + 2\frac{h_c}{T} + e^{K_0^{(r)} + K} \right) \right) \right]^n \right.$$  

and the free energy is derived directly from the previous equation

$$-KF^{(t)} = \zeta^{(0)}$$  

$$= \sum_{k=0}^{n_r} C_{n_r}^k \chi^{r+1}(1 - x)^{n_r - k} \left[ \left(1 - x\right)^2 \left(\ln(2) + \left(\frac{2}{T} \ln(2) + \frac{2}{T} \left(2H_0^{(t)}(1, 1) + 2\frac{h_c}{T} + e^{K_0^{(r)} + K} \right) \right) \right]^n \right.$$  

After a few steps, the number of configurations grows rapidly, as the number of weights $C_{n_r}^k \chi^{r+1}(1 - x)^{n_r - k}$ becomes exponentially large as well as the number of iterative functions to evaluate. However, we can obtain a very good approximation if we notice that these weights are distributed
closely around a Gaussian when $n_r$ is sufficiently large

$$C_n^x x^k(1-x)^{n_r-k} \approx \frac{\exp\left(-\frac{(k-n_r-x+\frac{1}{2})^2}{2a(1-a)n_r}ight)}{\sqrt{2\pi x(1-x)n_r}}. \quad (29)$$

For $r = 4$, for example, the number of internal sites is equal to $n_r = 170$, and the previous approximation is very accurate. Numerically, we solved the iterative functions up to level $r = 4$ included, using (B.11), and then apply for higher levels $r > 4$ the Gaussian approximation for $k$ distributed within four standard deviations around the mean value $x n_r + x - 1/2$, which gives precise results.

5. Calorimetry and thermodynamical functions in the dilute case

In this section, we evaluate different thermodynamical quantities as function of dilution using (28). The residual entropy per spin is plotted as a function of $x$ in figure 5 for $r$ between 1 and 6. For the single tetrahedron structure $r = 1$, the entropy is numerically identical to the exact expression (17) and presents a non-monotonic dependence with increasing dilution. For $r$ larger, the entropy is reduced, but saturates rapidly after $r = 5$, which corresponds to 684 sites. In the limit of extreme dilution, the entropy per spin is simply equal to $\ln(2)$, as expected. It is interesting to compare the resulting entropy with the Pauling estimation $S_P$ for an infinite number of tetrahedra, treated as independent, as function of dilution [13]

$$S_P(x) = \ln(2) - 3x^2(1-x) \ln(2) - 2x(1-x)^2 \ln(4/3) - \frac{1}{2}(1-x)^3 \ln(8/3). \quad (30)$$

Comparison between $S_P$, the experimental data for spin-ice Dy$_{2-x}$Y$_x$Ti$_2$O$_7$ in figure 4 of [13], and $S^{(6)}$ shows very similar values at low and moderate dilution, especially the entropy difference in the undiluted case is quite small, $S_P(0) = \frac{1}{2} \ln(3/2) \approx 0.2027$ and $S^{(6)}/N_6 \approx 0.20804$, which is an upper bound (see also inset of figure 5). Exact values for the undiluted hierarchical structure can be computed up to a certain order, but the entropy shows a behavior similar to spin-ice models. Approximations on a pyrochlore lattice made of Ising antiferromagnet tetrahedra give a closer value around 0.204 10 [18].

The specific heat $C_v^{(r)}$ is displayed in figure 6 as a function of temperature for five different values of $x$. The curve presents, in general, a broad maximum or Schottky anomaly at a temperature around $T = 1$, corresponding to the typical coupling $J = 1$ and associated with a crossover between a low-temperature spin-ice state and a paramagnetic state. However, the system stays antiferromagnetic in the low-temperature regime but is highly degenerated. The ground state energy per spin can be computed exactly for the first terms in the absence of dilution $E^{(1)}/4 = -1/2$, $E^{(2)}/12 = -5/6$, and the limiting value is estimated to be $E^{(r\rightarrow\infty)}/N_r \approx -0.9$ using the recurrence equations in appendix A. The main peak location behaves non-monotonically with dilution, as for the dilute compound Dy$_{2-x}$Y$_x$Ti$_2$O$_7$ in [13], which results from the non-monotonic fraction of ground states in elementary tetrahedral structures, as seen for the entropy. At large dilution, the specific heat can accurately be fitted with a two-level model with gap $\Delta$ and constant $C_0$, as can be seen in figure 6

$$C_v^{\text{approx}} = C_0 \frac{\Delta^2}{T^2} \frac{e^{\Delta/T}}{(1 + e^{\Delta/T})^2}. \quad (31)$$

For example, the curve for $x = 0.95$ was fitted with the previous formula using $C_0 \approx 0.114$ and $\Delta = 1.988$, which corresponds to the specific heat for a gas of dilute pairs of spins with $C_0 \approx 2(1-x) = 0.1$ and energy coupling very close to $J = \Delta/2 = 1$. However, at lower temperature, $T \approx 0.1$, the specific heat presents a second broad peak at an intermediate dilution factor $x \approx 0.3$ (see inset of figures 6 and 7) which cannot be reproduced by a two-level model.

These characteristics are exemplified in figure 7 for $T \leq 0.1$. The exponential-like Arrhenius behavior of the pure system seems to evolve to more complex features associated with a very small and broad peak contribution at intermediate dilutions and non-exponential deviations. Arrhenius behavior is then recovered when we approach large dilution, modeled by (31). We have actually rescaled the specific heat in figure 7 by a factor $1/T^2$, in order to check if excitations such as phonons or elastic modes are present in the intermediate dilute regime. In this case $C_v^{(r)}$ should scale like $T^2$, with $d = 2$, in our model for a two-dimensional Debye contribution. Such elastic modes (in the low-temperature dynamics of domain walls for example) could result from the non-trivial effect of a long-range and random distribution of the couplings, due to the additional bonds added at each step of the lattice construction, which tend to couple remote spins and induce non-local interactions. This could generate a random distribution of local fields, or small gaps at different scales.
Figure 7. Low-temperature behavior of the specific heat per spin \( C(r) / [N_r (1-x) T^2] \) at zero field and level \( r = 6 \) (2732 sites) for different disorder probability values. A local maximum develops for low dilution \( 0.1 \leq x \leq 0.3 \), then non-exponential behavior is observed for intermediate values \( x \approx 0.5 \).

Figure 8. Surface plot of the specific heat per spin \( C(r) / [N_r (1-x) T^2] \) at zero field and level \( r = 6 \) (2732 sites) as a function of temperature and dilution factor \( x \). The Schottky peak amplitude is reduced as \( x \) increases.

Such scaling was analyzed, for example, in the pyrochlore compound \( \text{Bi}_2\text{Ti}_2\text{O}_7 \) (with \( d = 3 \)) in order to measure the excess of specific heat due to additional Einstein oscillator contributions that could give rise to a broad peak at low temperature [19]. The scaling in \( T^2 \) in figure 7 is more appropriate, since a \( T^3 \) scaling would clearly present a divergence.

Non-usual low-temperature specific heat behavior in dilute systems was analyzed, in a different context, for Heisenberg magnets, within the low-temperature spin-wave approximation [20, 21], where dilution induced non-trivial temperature exponents depending on the nature of the couplings.

To summarize, the specific heat per spin as a function of both temperature \( T \) and dilution factor \( x \) is displayed in figure 8, where the variation of the main Schottky peak amplitude with \( x \) shows a decreasing behavior towards a system made of individual pairs of spins with a broader extension.

Fluctuations of the ferromagnetic order parameter defined by \( \Delta_F = [(\langle M^2 \rangle)_T]_r / [(1-x) N_r] \) can be evaluated directly from the free energy using a small field [11]

\[
\Delta_F = \frac{T}{(1-x) N_r} \left. \frac{\partial^2 F(r)}{\partial h_e^2} \right|_{h_e=0} - \left( \frac{1}{(1-x) N_r} \left. \frac{\partial F(r)}{\partial h_e} \right|_{h_e=0} \right)^2
\]  

and are plotted in figure 9. It takes noticeable values at low temperature for intermediate dilution, where short-range ferromagnetic order appears to be well developed. Such fluctuations could be associated with a classical spin-liquid phase, as opposed to a gas state at higher temperatures [11].

Susceptibility curves as a function of dilution and field are plotted in figure 10. We chose to represent only the low-field excitations \( h_e < 1 \) in order to follow the displacement and amplitude of the first peaks with dilution, in particular those corresponding to figure 3 in the same low-field region. As dilution is increased, a new peak appears at \( h_e = 0 \), corresponding to excitations of uncoupled and isolated spins. The location of the peak at \( h_e = 1/2 \) does not change, only its amplitude. It is associated with excitations which appear numerically only at recursion level \( r = 4 \) (172 sites), and might probably consist in flipping two distant spins.
the replica method and reorganization of configurational hierarchical lattices in the presence of dilution based on In this paper we propose a method to study frustrated energy configurations, such as three spins up and one spin of two spins up and two spins down (\(E_1\)) where, from a ground state undiluted structures with the same configurational weight or configurations.

The peak located at \(h_c = 1/3\) which appears at recursion level \(r = 3\) is instead moving towards higher field values, with several intermediate peaks in the range \(1/5 < h_c < 1/2\). Smaller peaks at \(h_c < 1/3\) are moving towards the origin instead. A surface plot figure 11 gives a general view of how peaks are moving with field with respect to dilution, and how their amplitude vanishes as we approach the high dilution regime. For higher field, transitions occur in small structures of four spins (\(r = 1\)) where, from a ground state of two spins up and two spins down (\(E = -2J\)), increasing the field leads successively to spin–flip processes with lower energy configurations, such as three spins up and one spin down at \(h_c = 1\) and all spins up at \(h_c = 3\).

6. Conclusion

In this paper we propose a method to study frustrated hierarchical lattices in the presence of dilution based on the replica method and reorganization of configurational weights at first order in the replica parameter \(n\). Interesting properties of the dilute spin–ice state at low temperature can be examined within this approximation by implementing recursive equations for the partition function, leading to the specific heat and susceptibility as functions of temperature and external field. Clear crossover evidence is seen between spin–ice and paramagnetic states in the specific heat, with the presence of a Schottky peak, and the zero-temperature entropy follows closely the Pauling approximation, at least at moderate dilution. Specific heat presents also a secondary contribution at low dilution (\(x \sim 0.3\)) and very low temperature, with non-Arrhenius behavior, at least at the temperatures considered numerically. This feature is probably due to the effect of dilution on the long-range couplings across the lattice, which involves a bimodal distribution of random antiferromagnetic couplings between sites at different scales, and a broad distribution of random effective fields or small gaps. This makes this the hierarchical model a good candidate for exploring in detail the physics of spin-ices or spin-liquids. Additional analysis can probably be made, for example, using correlation functions or short-range ferromagnetic order parameter \([11]\) to probe the spin correlations in the low-temperature state. This approximation scheme based on the replica method may probably be implemented more easily to hierarchical spin glass models with modal distribution of couplings, since the quenched disorder is treated as independent between the recursive diamond structures, making the need of a partial partition function unnecessary and therefore simplifying the analytical recurrence.

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Appendix A. Recursion relations for the non-disordered model

In this appendix we write the recursion relations for the non-disordered case (\(x = 0\)). Starting from the partition function \(Z_0(\sigma, \sigma') = \exp(-K \sigma \sigma')\) of a single antiferromagnetic link between two spins \(\sigma\) and \(\sigma'\), and \(K' = K\), we can generally assume the following recursive and stable form at any step \(r\)

\[
Z_r(\sigma, \sigma') = \exp (I_r - K_r \sigma \sigma' + H_r (\sigma + \sigma'))
\]

with \(I_0 = 0\), \(K_0 = K\), and \(H_0 = 0\) as initial conditions. At the next level \(r + 1\), we form the product

\[
Z_{r+1}(\sigma, \sigma') = \text{Tr}_{\sigma_1, \sigma_2} Z_r(\sigma, \sigma_1) Z_r(\sigma_1, \sigma') Z_r(\sigma, \sigma_2) Z_r(\sigma_2, \sigma') \times \exp \left( -K \sigma \sigma' - K \sigma_1 \sigma_2 + \frac{h_c}{T} (\sigma_1 + \sigma_2) \right).
\]

After replacing \(Z_r\) by its ansatz (A.1), and performing the sum over the internal spin degrees of freedom, we obtain

\[
Z_{r+1}(\sigma, \sigma') = \exp (4I_r - K \sigma \sigma' + 2H_r (\sigma + \sigma')) \times 2 \cosh \left[ 2K_r (\sigma + \sigma') - 4H_r - \frac{2h_c}{T} \right] e^{-K} + 2e^K.
\]
The term into bracket \( \{ \cdots \} \) can be rewritten as
\[
2 \cosh \left[ 2K_r (\sigma + \sigma') - 4H_r - \frac{\hbar}{T} \right] e^{-K} + 2e^K
= \exp \left( \bar{I}_r - \bar{K}_r \alpha' + \bar{H}_r (\sigma + \sigma') \right) \tag{A.3}
\]
with the following equations for \( \bar{I}_r, \bar{K}_r, \) and \( \bar{H}_r \)
\[
\exp (\bar{I}_r - \bar{K}_r + 2H_r) = 2 \cosh [4K_r - 4H_r - 2H_r] e^{K} + 2e^K,
\exp (\bar{I}_r + \bar{K}_r - 2H_r) = 2 \cosh [4K_r + 4H_r + 2H_r] e^{-K} + 2e^K.
\]
This set of equations can be solved by eliminating successively the arguments in the exponential terms. Then the recursive solutions for the new couplings of \( Z_{r+1} \) are given by
\[
I_{r+1} = 4H_r + \bar{I}_r, \quad K_{r+1} = K + \bar{K}_r, \quad H_{r+1} = 2H_r + \bar{H}_r. \tag{A.4}
\]
These relations can be easily implemented in order to compute numerically the different thermodynamical quantities from free energy \( F_r = -T \ln Z_r \).

Appendix B. Recursion relations at finite temperature

In this section the recursive equations for the different couplings in (18) are derived. Using the Kronecker integral (20), we can rewrite \( Z_{r+1} \) as
\[
Z_{r+1} = \sum_{k=0}^{4n_r} \chi^k (1 - x)^{4n_r - k} \int P(\epsilon_1)P(\epsilon_2) \, d\epsilon_1 \, d\epsilon_2 \times \text{Tr} \sigma_1^\sigma_2^\sigma_{1'}^\sigma_{2'}^\sigma_{1}^{\sigma_{2}} \times \exp \left( -K' \epsilon \sum_{\alpha} \sigma_{\alpha} \sigma_{\alpha'} - K' \epsilon \sum_{\alpha} \sigma_{1}^{\alpha} \sigma_{2}^{\alpha} \right.
\]
\[
- K' \sum_{\alpha} (\sigma_{1}^{\alpha} \sigma_{2}^{\alpha}) (\sigma_{\alpha} \sigma_{\alpha'} + \epsilon \sigma_{\alpha} \sigma_{\alpha'}) \right.
\]
\[
\times \exp \left[ \frac{\hbar}{T} \sum_{\alpha} (\sigma_{1}^{\alpha} \sigma_{2}^{\alpha} \sigma_{1'}^{\alpha} \sigma_{2'}^{\alpha}) \int_0^{2\pi} \frac{d\theta}{2\pi} e^{-\omega k} \right.
\]
\[
\times \sum_{k_1, k_2, k_3, k_4=0} C_n^{k_1} C_n^{k_2} C_n^{k_3} C_n^{k_4} e^{i\theta(k_1+k_2+k_3+k_4)}
\]
\[
\times \exp \left[ nH_{k_1}^{(r)} (\epsilon_1, \epsilon_1) + nl_{k_2}^{(r)} (\epsilon_1, \epsilon') \right.
\]
\[
+ nH_{k_3}^{(r)} (\epsilon, \epsilon_2) + nl_{k_4}^{(r)} (\epsilon_2, \epsilon') \right.
\]
\[
+ K_{k_1}^{(r)} \epsilon \sum_{\alpha} \sigma_{\alpha} \sigma_{1}^{\alpha} \right.
\]
\[
+ K_{k_2}^{(r)} \epsilon \sum_{\alpha} \sigma_{1}^{\alpha} \sigma_{\alpha'} \right.
\]
\[
+ K_{k_3}^{(r)} \epsilon \sum_{\alpha} \sigma_{2}^{\alpha} \sigma_{\alpha} \right.
\]
\[
+ K_{k_4}^{(r)} \epsilon \sum_{\alpha} \sigma_{\alpha} \sigma_{\alpha'} \right.
\]
\[
+ H_{k_1}^{(r)} (\epsilon_1, \epsilon_1) \sum_{\alpha} (\epsilon \sigma_{\alpha} + \epsilon_1 \sigma_{1}^{\alpha})
\]
\[
+ H_{k_2}^{(r)} (\epsilon, \epsilon_1) \sum_{\alpha} (\epsilon' \sigma_{\alpha} + \epsilon_1 \sigma_{1}^{\alpha})
\]
\[
+ H_{k_3}^{(r)} (\epsilon, \epsilon_2) \sum_{\alpha} (\epsilon \sigma_{\alpha} + \epsilon_2 \sigma_{2}^{\alpha})
\]
\[
+ H_{k_4}^{(r)} (\epsilon', \epsilon_2) \sum_{\alpha} (\epsilon' \sigma_{\alpha} + \epsilon_2 \sigma_{2}^{\alpha}) \right]. \tag{B.1}
\]

Using (21), we can integrate over \( \theta \) and rewrite (B.1) as
\[
Z_{r+1} = \sum_{k=0}^{4n_r} \chi^k (1 - x)^{4n_r - k}
\]
\[
\times \int P(\epsilon_1)P(\epsilon_2) \, d\epsilon_1 \, d\epsilon_2 \times \text{Tr} \sigma_1^{\sigma_2} \times 2^{n(\epsilon_1 - 1) - 2n(\epsilon_2 - 1)} \times \exp \left( -K' \epsilon \sum_{\alpha} \sigma_{\alpha} \sigma_{\alpha'} - K' \epsilon \sum_{\alpha} \sigma_{1}^{\alpha} \sigma_{2}^{\alpha} \right.
\]
\[
- K' \sum_{\alpha} (\sigma_{1}^{\alpha} \sigma_{2}^{\alpha}) (\sigma_{\alpha} \sigma_{\alpha'} + \epsilon \sigma_{\alpha} \sigma_{\alpha'}) \right.
\]
\[
\times \sum_{k_1=\max(0,k-3n_r)} nD_{k_1}^{k_2} \left[ l_{k_1}^{(r)} (\epsilon_1, \epsilon_1) + l_{k_1}^{(r)} (\epsilon_1, \epsilon') \right] \right.
\]
\[
+ \sum_{k_1=\max(0,k-3n_r)} D_{k_1}^{k_2} K_{k_1}^{(r)} \right.
\]
\[
\times \sum_{\alpha} (\epsilon \sigma_{\alpha} + \epsilon' \sigma_{\alpha'})(\sigma_{1}^{\alpha} \sigma_{2}^{\alpha} + \epsilon_2 \sigma_{2}^{\alpha}) \right.
\]
\[
+ \sum_{\alpha} (\epsilon \sigma_{\alpha} + \epsilon' \sigma_{\alpha'})(\sigma_{1}^{\alpha} \sigma_{2}^{\alpha} + \epsilon_2 \sigma_{2}^{\alpha}) \right]
\]
\[
+ \sum_{\alpha} (\epsilon \sigma_{\alpha} + \epsilon' \sigma_{\alpha'})(\sigma_{1}^{\alpha} \sigma_{2}^{\alpha} + \epsilon_2 \sigma_{2}^{\alpha}) \right]. \tag{B.2}
\]

Now the sum over intermediate spins \( \sigma_{1}^{\alpha} \) and \( \sigma_{2}^{\alpha} \) can be performed directly. Let first define intermediate couplings
\[
\bar{K}_k^{(r)} = \sum_{k_1=\max(0,k-3n_r)} D_{k_1}^{k_2} K_{k_1}^{(r)} = K', \tag{B.3}
\]
and new functions
\[
l_{k_1}^{(r)} (\epsilon, \epsilon', \epsilon_1, \epsilon_2) \]
\[
:= \sum_{k_1=\max(0,k-3n_r)} D_{k_1}^{k_2} \left[ l_{k_1}^{(r)} (\epsilon_1, \epsilon_1) + l_{k_1}^{(r)} (\epsilon_1, \epsilon') \right] \right.
\]
\[
+ \sum_{\alpha} (\epsilon \sigma_{\alpha} + \epsilon' \sigma_{\alpha'})(\sigma_{1}^{\alpha} \sigma_{2}^{\alpha} + \epsilon_2 \sigma_{2}^{\alpha}) \right]
\]
\[
+ \sum_{\alpha} (\epsilon \sigma_{\alpha} + \epsilon' \sigma_{\alpha'})(\sigma_{1}^{\alpha} \sigma_{2}^{\alpha} + \epsilon_2 \sigma_{2}^{\alpha}) \right]. \tag{B.2}
\]
\[ H_k^{(r)}(\epsilon, \epsilon', \epsilon_1) := \sum_{k_1=\max(0,k-3n)}^{\min(n_\alpha,k)} D_{n_\alpha,4}^{k_1} \left[ H_k^{(r)}(\epsilon, \epsilon_1) + H_k^{(r)}(\epsilon', \epsilon_1) \right]. \] (B.4)

Then we isolate the part in (B.2) containing only \( \sigma_1^a \) and \( \sigma_2^a \), and perform the sum:

\[
\begin{align*}
& \text{Tr}_{\sigma_1^a,\sigma_2^a} \exp \left( -K' \epsilon_1 \epsilon_2 \sum_a \sigma_1^a \sigma_2^a \right) + \hat{H}_k^{(r)}(\epsilon, \epsilon', \epsilon_1) + \frac{\hbar c}{T} \sum_a \epsilon_1 \sigma_1^a \\
& + \left( \hat{H}_k^{(r)}(\epsilon, \epsilon', \epsilon_2) + \frac{\hbar c}{T} \right) \sum_a \epsilon_2 \sigma_2^a \\
& = \prod_a \left\{ \exp(-K' \epsilon_1 \epsilon_2) 2 \cosh \left( \hat{K}_k^{(r)}(\epsilon_1 + \epsilon_2)(\sigma_1^a + \epsilon' \sigma_1^a) \right) \right. \\
& + \left( \hat{H}_k^{(r)}(\epsilon, \epsilon', \epsilon_1) + \frac{\hbar c}{T} \right) \epsilon_1 + \left( \hat{H}_k^{(r)}(\epsilon, \epsilon', \epsilon_2) + \frac{\hbar c}{T} \right) \epsilon_2 \\
& + \exp(K' \epsilon_1 \epsilon_2) 2 \cosh \left( \hat{K}_k^{(r)}(\epsilon_1 - \epsilon_2)(\sigma_1^a + \epsilon' \sigma_1^a) \right) \\
& - \left( \hat{H}_k^{(r)}(\epsilon, \epsilon', \epsilon_2) + \frac{\hbar c}{T} \right) \epsilon_2 \right\}. \quad (B.5)
\]

Next, we perform the integration over \( \epsilon_1 \) and \( \epsilon_2 \)

\[ Z_{r+1} = \sum_{k=0}^{4n_\alpha} C_{4n_\alpha}^k (1-x)^{4n_\alpha-k} \exp \left( -K' \epsilon \epsilon' \sum_a \sigma_1^a \sigma_2^a \right) \]
\[ \times \left[ (1-x)^2 \exp \left( nI_k(\epsilon, \epsilon', 1, 1) \right) + \hat{H}_k^{(r)}(1, 1, 1) \sum_a \sigma_1^a \right] \]
\[ \times 2^n \prod_a \left\{ \exp(-K') \cosh \left( \frac{2\hat{K}_k^{(r)}(\epsilon) \sigma_1^a + \epsilon' \sigma_1^a}{2} \right) + 2\hat{H}_k^{(r)}(\epsilon, \epsilon', 1) + \frac{\hbar c}{T} \right\} \]
\[ + 2x(1-x) \exp \left( nI_k(\epsilon, \epsilon', 0, 1) \right) + \hat{H}_k^{(r)}(1, 0, 1) \sum_a \sigma_1^a \right] \]
\[ \times 2^n \prod_a \cos \left( \frac{2\hat{K}_k^{(r)}(\epsilon) \sigma_1^a + \epsilon' \sigma_1^a}{2} \right) + \hat{H}_k^{(r)}(\epsilon, \epsilon', 1) + \frac{\hbar c}{T} \right\} \]
\[ + x^2 \exp \left( nI_k(\epsilon, \epsilon', 0, 0) \right) + \hat{H}_k^{(r)}(0, 0, 1) \sum_a \sigma_1^a \right\}. \quad (B.6)
\]

We introduce now a set of functions \([M_{k,j}(\epsilon, \epsilon'), Q_{k,j} \hat{H}_{k,j}(\epsilon, \epsilon')\)] for each of the three terms appearing in the previous expression and proportional to \((1-x)^2 \) (\( l = 0 \)), \(2x(1-x) \) (\( l = 1 \)), and \( x^2 \) (\( l = 2 \)) respectively. The first factor associated with \((1-x)^2\) can be exponentiated such that

\[ \exp \left( I_k(\epsilon, \epsilon', 1, 1) + \hat{H}_k^{(r)}(1, 1, 1)(\epsilon^a + \epsilon' \sigma_1^a) \right) \]
\[ \times 2 \left\{ \exp(-K') \cosh \left( 2\hat{K}_k^{(r)}(\epsilon^a + \epsilon' \sigma_1^a) \right) + 2\hat{H}_k^{(r)}(\epsilon, \epsilon', 1) + \frac{\hbar c}{T} \right\} \]
\[ = \exp \left( M_{k,0}(\epsilon, \epsilon') + Q_{k,0} \epsilon \sigma_1^a \sigma_1^a \right) \]
\[ + \hat{H}_{k,0}(\epsilon, \epsilon')(\epsilon^a + \epsilon' \sigma_1^a). \quad (B.7) \]

Similarly we have for the second term proportional to \(2x(1-x)\)

\[ \exp \left( I_k(\epsilon, \epsilon', 0, 1) + \hat{H}_k^{(r)}(1, 0, 1)(\epsilon^a + \epsilon' \sigma_1^a) \right) \]
\[ \times 2 \cosh \left[ \hat{K}_k^{(r)}(\epsilon^a + \epsilon' \sigma_1^a) + \hat{H}_k^{(r)}(\epsilon, \epsilon', 1) + \frac{\hbar c}{T} \right] \]
\[ = \exp \left( M_{k,1}(\epsilon, \epsilon') + Q_{k,1} \epsilon \sigma_1^a \sigma_1^a \right) \]
\[ + \hat{H}_{k,1}(\epsilon, \epsilon')(\epsilon^a + \epsilon' \sigma_1^a). \quad (B.8) \]

The last term associated with \(x^2\) can be exponentiated using the values \(M_{k,2}(\epsilon, \epsilon') := I_k(\epsilon, \epsilon', 0, 0) \), \(Q_{k,2} := 0 \) and \(H_{k,2}(\epsilon, \epsilon') := \hat{H}_k^{(r)}(0, 0, 1) \). All these functions can be identified in a unique way by using the four possible configurations for \(\sigma_1^a\) and \(\sigma_1^a\). Then the partition function can be rewritten as

\[ Z_{r+1} = \sum_{k=0}^{4n_\alpha} C_{4n_\alpha}^k (1-x)^{4n_\alpha-k} \exp \left( -K' \epsilon \epsilon' \sum_a \sigma_1^a \sigma_1^a \right) \]
\[ \times \left[ \sum_{l=0}^{4n_\alpha} C_{4n_\alpha}^l (1-x)^{4n_\alpha-l} \exp \left( nI_{k,l}(\epsilon, \epsilon') \right) + Q_{k,l} \epsilon \sigma_1^a \sigma_1^a \right] \]
\[ + \hat{H}_{k,l}(\epsilon, \epsilon')(\epsilon^a + \epsilon' \sigma_1^a). \quad (B.9) \]

As before we can expand the exponential terms at first order in \(n\) and rearrange the powers in \(x\) such that

\[ Z_{r+1}(\epsilon^a, \epsilon' \sigma_1^a) = \exp \left( -K' \epsilon \epsilon' \sum_a \sigma_1^a \sigma_1^a \right) \]
\begin{equation}
\times \sum_{k=0}^{4n_{l}+2m_{n_{l}+1}} C^{k}_{4n_{l}+2} k (1-x)^{4n_{l}+2-k} \\
\times \exp \left( \frac{\min(2,k)}{\max(0,k-4n_{l})} C^{k-1}_{4n_{l}+2} \left[ nM_{k-l}(\epsilon, \epsilon') \right. \right.
\left. + Q_{k-l}(\epsilon, \epsilon') \sum_{\alpha} \sigma^{\alpha} \sigma'^{\alpha} \right) \right) \end{equation}

From this result, we can deduce finally the recursive equation for the couplings

\begin{equation}
K^{(r+1)}_{k} = \min(2,k) \sum_{l=\max(0,k-4n_{l})}^{\min(2,k)} C^{k-1}_{4n_{l}+2} M_{k-l}(\epsilon, \epsilon'), \\
\ell^{(r+1)}_{k} (\epsilon, \epsilon') = \sum_{l=\max(0,k-4n_{l})}^{\min(2,k)} C^{k-1}_{4n_{l}+2} M_{k-l}(\epsilon, \epsilon'), \\
\text{and} \quad H^{(r+1)}_{k} (\epsilon, \epsilon') = \min(2,k) \sum_{l=\max(0,k-4n_{l})}^{\min(2,k)} C^{k-1}_{4n_{l}+2} H_{k-l}(\epsilon, \epsilon'). \end{equation}

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