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Signature of Thermal Rigidity Percolation

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Abstract. To explore the role that temperature and percolation of rigidity play in determining the macroscopic properties, we propose a model that adds translational degrees of freedom to the spins of the well known Ising hamiltonian. In particular, the Ising model illustrate the long-standing idea that the growth of correlations on approach to a critical point could be describable in terms of the percolation of some sort of "physical cluster". For certain parameters of this model we observe two well defined peaks of C_V, that suggest the existence of two kinds of "physical percolation", namely connectivity and rigidity percolation. Thermal fluctuations give rise to two different kinds of elementary excitations, i.e. droplets and configuron, as suggested by Angell in the framework of a bond lattice model approach. The later is reflected in the fluctuations of redundant constraints that gives stability to the structure and correlate with the order parameter.

1. Introduction

In order to explore the role that the lost of degrees of freedom (dof) plays due to the decrease of temperature in a magnetic network bond forming lattice, we propose an extension of the Ising Model. In this extension, each particle, that has a spin associated, and can freely translate inside a cell, but not escape from it. The later in a similar way to the early cell-theory of liquids, where each Wigner-Seitz cell is occupied by at most one particle [1], to mimic the conditions of a particle caged inside its neighbors [2]. Although mainly of academic interest, this model could have applications in the study of real systems as the spin-crossover in solid materials [3] or in the study of thermodynamical or structural properties of magnetic gels [4], where sudden sharp properties are mainly attributed to the percolation of the bond network [5, 6]. For simplicity in this preliminary version of the model we have considered the case where all cells are occupied by one point-like particle (i.e. does not have excluded volume or area in 2D), then no interactions far from first neighbor cells can occurs, as we describe in the following.

1.1. The Model

In 2D each particle has two dof for free translation inside a cell (i.e. cell confinement restricts the motion of the particle by non-holonomic non-lagrangian constraints but does not lock or jam). The model particles has no volume exclusion to prevent other entropic effects, allowing the particles to interact only by energetic bonds. To simulate the bond we use an Ising-like interaction of the particles with its neighboring cells, the later to mimic a constraint formation. If the orientation and distance between spins are favorable, the interacting particles become correlated, in that way the later represents the decrease of the particles dof by one. Assume

that the size of the cell is larger than the vibrations of the bond length. The hamiltonian is the following:

$$H = \sum_{<ij>} J_{ij}\sigma_i\sigma_j + \sum_i U_i^{cell}, \tag{1}$$

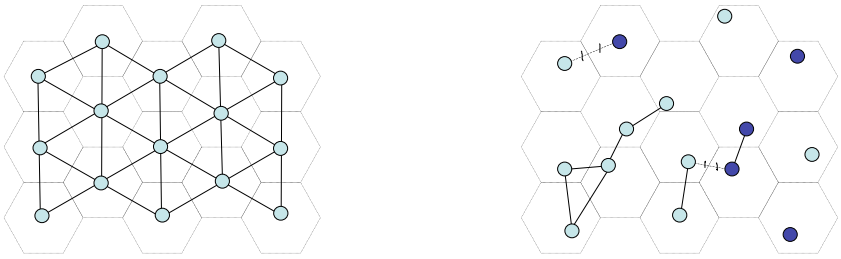
where $\sigma_i = -1, 1$, $< ij >$ means interactions between nearest neighboring cells and J_{ij} corresponds to the attractive energetic interaction given by

$$J_{ij} = \begin{cases} -\varepsilon & \text{for} \quad l - \Delta/2 < |r_i - r_j| < l + \Delta/2 \\ 0 & otherwise \end{cases}, \tag{2}$$

whereas U_i^{cell} represents the cell where the particle can freely move,

$$U_i^{cell} = \begin{cases} 0 & \text{inside the cell} \\ \infty & \text{outside the cell} \end{cases}, \tag{3}$$

a nonzero Δ allows bond vibrations. A representation of the system is given in Figure 1, only attractive interactions are considered as a constriction. Non-favorable energetic interactions have low probability to be observed because the particles prefer to move out to the zero interaction energy length, due to the availability of the free volume inside the corresponding cell.



a) b)

Figure 1: Extension of the Ising model on a triangular lattice, where each particle can translate inside the hexagonal cell and interacts as described in the text. a) represents a ground state configuration ($T = 0$), where all the spins points in the same direction (i.e. the magnetization is 1) and all the particles are at the favorable distance to form a bond. b) represents an idealized configuration where the temperature effect produces bond fluctuations, allowing the particles to move inside the corresponding cell. Continuous lines between particles represent favorable energetic interactions, whereas crossed lines represent non-favorable energetic interactions.

To separate the confinement of the geometric cell from the energetic confinement due to the bond formation we take $l >> \Delta$ (at least one order of magnitude). An important aspect is that the configurations produced with this model are not static, due to thermal motion of particles inside the cells and changes of the spin orientation, allows the bond formation if distance and spins are pointing in a favorable direction.

Ising model phase transition have been subject of many studies that relate it with the connectivity percolation of a proper defined cluster, namely the Fortuin-Kasteleyn "physical cluster", [7, 8]. Initially it was believed that a simple geometrical cluster made of favorable spin interactions percolates at critical temperature. However, using this definition now is clear that

it cannot be identified with the thermal phase transition [9], since on average it is larger than a "physical cluster". Accepting this fact Coniglio and Klein suggest to distinguish between active and inactive bonds of the geometrical cluster with certain probability, [10]. On the other hand, important aspects of rigidity percolation in a field have been addressed by C. Moukarzel [11]. Of course, other very interesting aspects that we do not treat for the moment are nucleation, metastability, frustration and dilution [7, 12, 13, 14, 15].

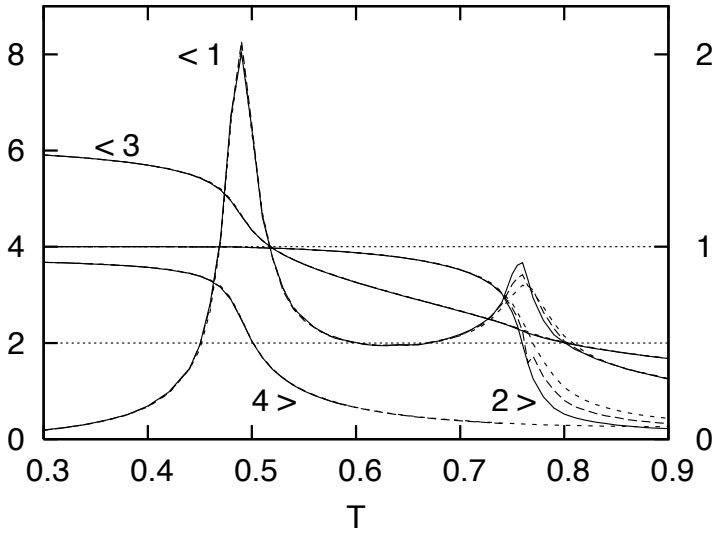


Figure 2: Energy Fluctuations (C_V, 1), magnetization (2) average coordination (3) and ψ_6 bond-orientational order parameter (4). Symbols < and >, represent left and right scales respectively.

2. Results

The traditional Ising Model shows a second order phase transition that is reflected in the magnetization order parameter and the energy fluctuations, C_V. For the extension proposed in this paper, besides magnetization, other parameters can take the form of translational and/or bond-orientational order. Translational order can be studied through the radial distribution function and is not an issue of the present study. A quantitative measure of bond-orientational (hexagonal) order is provided by the so-called "global" bond-orientational order parameter, ψ_6, that was evaluated using the following definition:

$$\psi_6 = \left| \frac{1}{N_{nn}} \sum_{jk} \exp(i6\theta_{jk}) \right|, \tag{4}$$

where j runs over all particles in the system, k runs over all bonded nearest neighbors (nn) of particle j and N_{nn} is the total number of such nearest neighbors in the system. The angle θ_{jk} is defined between some fixed reference axis in the system and the vectors ("bonds") connecting nearest neighbors j and k.

In the Figure 2 we show the results of the standard Metropolis Monte Carlo simulation tecnique for C_V, average magnetization, average coordination and orientational-order parameter ψ_6, all as a function of the temperature for several systems sizes (900, 1600, 3600 particles). From high to low temperatures the first peak of C_V corresponds to the spontaneous magnetic order of the system as happens in the usual Ising model. However, the main difference that we clearly

see is a second maximum of C_V at lower temperature. In Figure 3 we show some examples of the configurations at low, intermediate and high temperatures (i.e. below, between and above the C_V peaks). At intermediate temperatures the magnetic order is high but ψ_6 still shows high degree of orientational disorder. In Figure 3b we can see spins pointing in the opposite direction as happens in the usual Ising Model due to the elementary thermal excitations (that produce the usual droplet formation) but in this case in an orientational disordered state. The fact that at low temperatures ψ_6 shows high degree of bond-orientational order, rises the idea of a different kind of elementary excitation that produces orientational disorder. We can see the effect of these excitations in the disordered spots surrounded by large regions of ordered particles in Figure 3a. In other words, when the magnetic order is large, thermal fluctuations produce islands of spin pointing in the opposite direction (droplets). In a similar way in this model when the orientational order is large, thermal fluctuations produce orientational disorder, due to the addition of translational degrees of freedom allowing the particles to displace by 'breaking and forming bonds' with their neighboring cells. Austen Angell refers to that kind of configurational excitations as configuron [16].

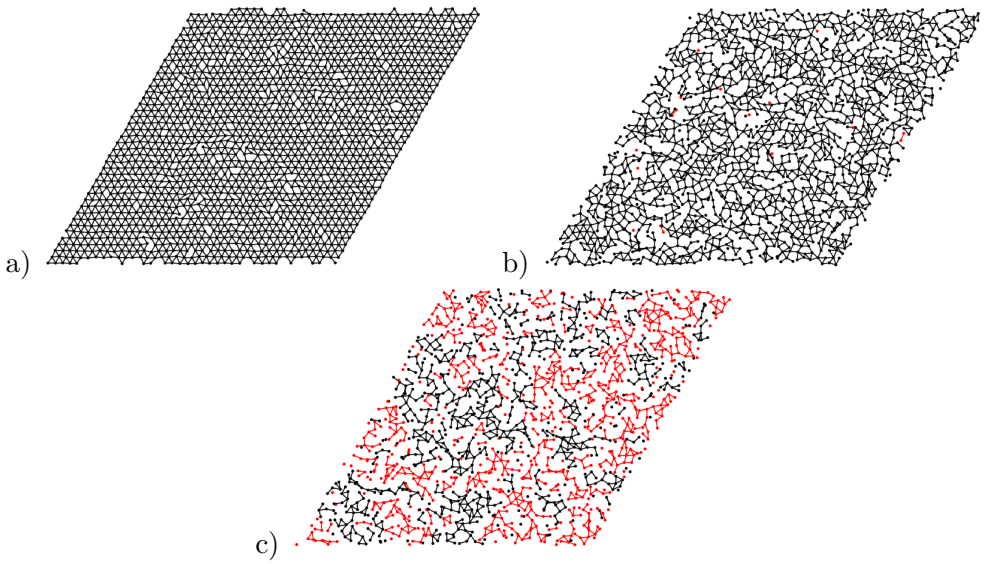


Figure 3: Configurations at a) $T = 0.4$, b) $T = 0.6$ and c) $T = 0.9$. The spin of particles in black point up, whereas particles in red point down.

To give a description of the configurational excitations we have mesured the orientational order of the system, ψ_6. We have observed that the orientational parameter correlates well with the second peak of C_V, which does not show the usual finite size effect with the increase of the size of the system, as we can see in Figure 2. Up to droplet formation, the second peak appears when the system is fully magnetized (i.e. when most of the spins are pointing in the same direction). Then, for temperatures below the maximum of the second peak, a different kind of order appears and is reflected in the consequent increase of the value of ψ_6, as temperature decreases.

3. Discussion

From the average coordination plot, shown in Figure 2, we can recognize that magnetization occurs even when the system is not full coordinated and corresponds to coordinations slightly larger than 2 and below 4.

The coordination 4 is crucial from the rigidity percolation point of view, since mean field Maxwell constraint counting in 2D predicts the loss of dof at this coordination for central forces. The interaction between particles only occurs between those particles that belong to neighboring cells. This interaction contributes with a favorable term in the hamiltonian when the orientation of the spin and the position between them are appropriate. The later allows us to consider a favorable interaction as a constriction, so the number of remaining dof of the structure is given by the total number of dof for N sites (equal to $2N$) minus the number of independent constraints. A redundant or linearly dependent constraint only imposes additional reinforcement to an already rigid cluster connected by favorable interactions, and that should be reflected in the energy fluctuations.

A key quantity is the remaining dof normalized per dof, $f = dof/2N$. We define the total number of constraints per dof as $n_c = n_i + n_r$, where the number of independent (redundant) is n_i (n_r), and then we can generally write,

$$f = \frac{2N - (2Nn_c - 2Nn_r)}{2N} = 1 - n_c + n_r = 1 - n_i \tag{5}$$

Neglecting the redundant constraints n_r in the last equation, as first done by Maxwell, we come to Maxwell counting:

$$f \approx f_M = 1 - n_c \tag{6}$$

Now the idea is to associate the rigidity percolation transition with the point where f_M goes to zero. The Maxwell approximation gives a good account of the number of remaining degrees of freedom, but it ultimately fails, because some constraints are redundant. An exact way to count the redundant constrictions can be carried out using the "the pebble game" algorithm [17, 18], that is a combinatorial procedure and is based in the Lamman theorem of graph theory which is exact in 2D. It is convenient to introduce the mean coordination $< r >$ as an average number of bonds stemming from a site. It is given by $< r >= pz$, where p is the probability of the bond being present, z is the maximum coordination of the underlying regular lattice (eg. 6 for the triangular, 4 for the square and 3 for the honeycomb). If the total number of sites is N, the number of bonds is $N_B = N < r > /2$. Each of these bonds represents one constraint, as always in central force networks, and therefore the number of constraints per dof is given by

$$n_c = \frac{N_B}{2N} = \frac{< r >}{4} \tag{7}$$

Therefore, according with the Maxwell counting we obtain

$$f \approx f_M = 1 - \frac{< r >}{4}. \tag{8}$$

This quantity goes to zero at $< r >_c = 4$, which we associate with the rigidity percolation transition. A similar expression when we consider angular constraints, that also corresponds to the case of connectivity percolation in a rigid lattice, gives $< r >_c = 2$ [19].

Now, since we are considering as constraints all the favorable contributions of the hamiltonian, the total number of lattice bonds (N_{LB}) has either a favorable or non-favorable interaction then the energy is given by

$$E = -J(N_c) + J(N_{LB} - N_c) = -2J(N_i + N_r) + JN_{LB}, \tag{9}$$

where we have used the fact that the total number of constraints is given by the linearly independent plus redundant (i.e. $N_c = N_i + N_r$).

Since the specific heat is given by $C_V = \frac{1}{kT^2}\{< E_i^2 > - < E_i >^2\}$, we can separate the independent and redundant contributions to each energy contribution, obtaining the following expression

$$\begin{aligned} kT^2 C_V = -2J\{&< N_i^2 > - < N_i >^2 + < N_r^2 > - < N_r >^2 \\ &+ 2(< N_i N_r > - < N_i >< N_r >)\} \end{aligned} \tag{10}$$

Since we are considering each favorable interaction as a constriction, using the 'the pebble game' we are able to determinate how many independent and redundant constrictions each configuration has, and then calculate their fluctuations. We have plotted the different contributions in Figure 4. Where we see that the main contribution to the second peak is due to the fluctuations of redundant constraints. Redundant constrictions are not localized as we show in the Figure 5a, where we observe that once a system is rigid and over constrained in this case by only one redundant constriction, all bonds are candidates to be the redundant constraint too. So if elementary excitations produce a bond breaking, as in the case of the Figure 4, that excitation always breaks a redundant constraint leaving the system still rigid. In that case a redundant constraint gives stability, and it produces a smeared out heat C_V bumps known as Schottky anomalies as Angell describes for a "bond lattice model" [20]. Moreover, if we plot the number of redundant constraints we see how it goes to zero near the second peak. To explore this observation we have evaluated the C_V behavior of larger systems as we mention before, Figure 2. Whereas, the first C_V jump, the one that promotes the magnetization of the system, presents the usual increase of the C_V as we increase the system size, the second C_V jump do not, as expected in a Schottky anomaly.

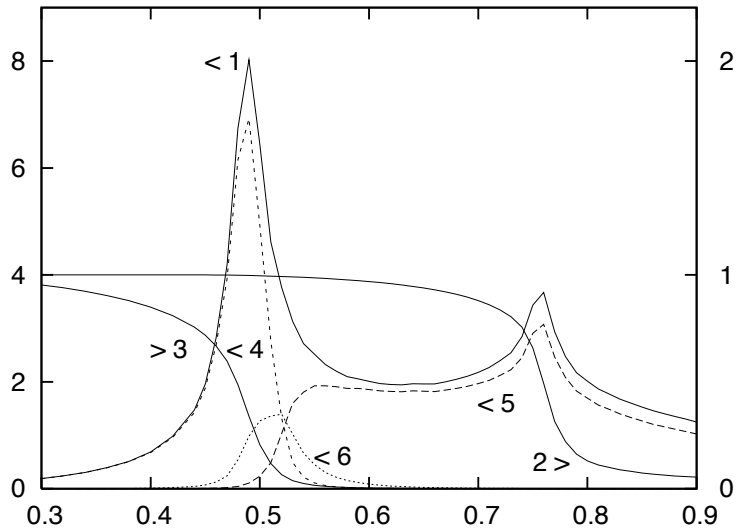


Figure 4: Energy Fluctuations (C_V, 1), magnetization (2) and average of normalized redundant constraints (3). Different contributions to C_V $< N_r^2 > - < N_r >^2$ (4), $< N_i^2 > - < N_i >^2$ (5) and $2(< N_i N_r > - < N_i >< N_r >)$ (6), as described in the text. Symbols $<$ and $>$, represent left and right scales respectively.

Using "the pebble game" Thorpe and Chubynsky [19], report a study of the generic triangular lattice where they have observed that the athermal rigidity percolation occurs very near coordination 4. They also draw parallels between the rigidity and connectivity percolation, relating over-constraints (excess of constrictions than necessary to lost dof) with a loop formation

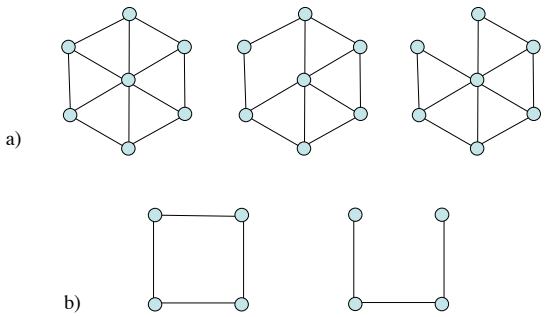


Figure 5: Example of the effect of an elementary excitation that produces fluctuations on redundant constraints. a) Once the system becomes magnetized and rigid at low temperatures near and below the second peak, the broken constriction is always redundant in the rigidity sense, since it leaves the body rigid. b) Since loops play the role of redundancies, if temperature breaks a bond of the loop, this bond is redundant but in the connectivity sense.

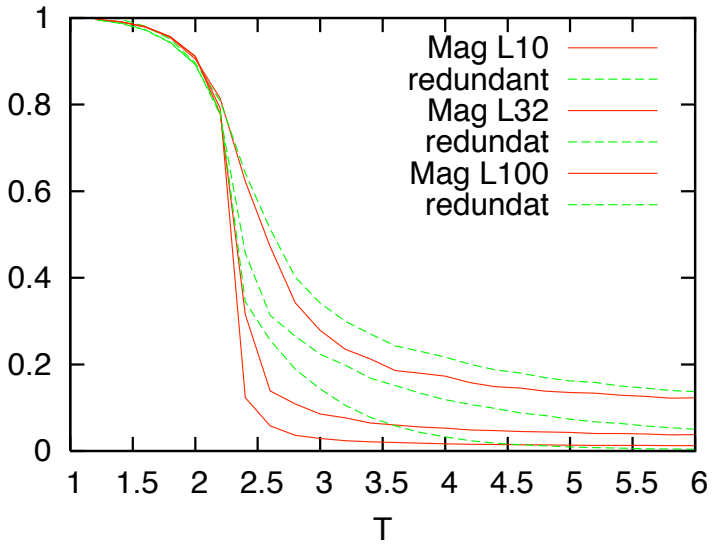


Figure 6: Comparison of the magnetization (continuos line) and percolation of the fraction of redundant constraints (dashed line).

between two sites, i.e. redundant ways to arrive between two sites, as shown in Figure 5b. Now the fact that magnetization occurs for coordinations larger than 2 is understandable and is consistent with active bonds defining a "physical cluster" rather than a geometrical one, as explained before.

Going back to the case of the usual Ising model, where the magnetization effect is related with connectivity percolation of the so called "physical clusters". From the previous discussion a relation of magnetization with percolation of redundant bonds is also suggested, in this case redundancy interpreted as those sites that are already connected as we mention above [19]. To explore this point in Figure 6, we calculate the fraction of redundant bonds in the over-constraint percolating cluster, defined by those bonds that belong to a loop. We can see how that fraction follows qualitatively the magnetization of the system in a square lattice, and it shows similar finite size effects, in a similar way of the "physical cluster" description [9]. The fraction of redundant bonds was obtained during the MC simulations using "pebble game" algorithm, since connectivity percolation can be considered as rigidity percolation with the sites having one degree of freedom [19]. The later is supported by the fact that in the so called high temperature series expansions of the pure Ising model, that is valid for all temperatures [21, 22], only the terms that belong to a loop [14, 23] survive.

It is also expected that reducing redundancy, the fluctuations of the second peak gradually disappear. A simple way to decrease redundant constraints but not rigidity is to eliminate diagonal interactions of the proposed cell model. The later corresponds topologically to a transformation of the triangular lattice to a square like one, when the maximum coordination is 4, then the over-constraints reduce notoriously. On the Figure 8, we show how the reduction of over-constraints (redundant bonds) the second peak associated with rigidity percolation C_V disappears, stressing the importance of redundant constrictions.

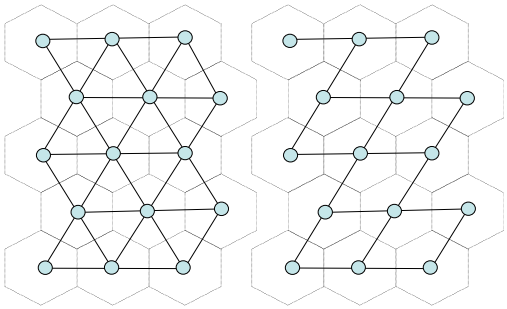


Figure 7: Decreasing redundant constraints of the triangular lattice, producing a topological square lattice by cutting the diagonal bonds.

4. Conclusion

We have proposed an extension of the Ising model to add translational degrees of freedom to each spin. As temperature decreases we have observed that the fluctuations of redundant constraints produce a non-divergent second peak on C_V, which correlates well with the increase of the order parameter ψ_6. The later corresponds to a configurational elementary excitation that is different

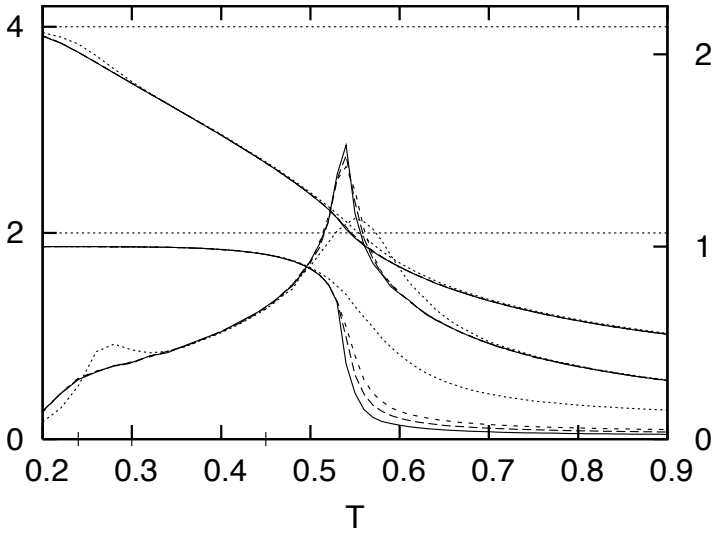


Figure 8: The same as in Figure 2 but using the model where we have decreased over constraint, see previous figure). C_V plot shows only one well defined peak that scales with the system size. It corresponds to the spontaneous magnetization - similar perhaps as occurs with the decrease of loops in the Ising model on a Cayley tree. However, thermodynamic limit should be carefully studied.

from the usual droplets produced by spin orientation. The configurational excitations in this model are similar to a bond lattice model proposed by Austen Angell. Our interpretation is that below the temperature where the second peak of C_V appears, redundant constraints give support to all structure but temperature produces local disorder by the bond breaking of some redundant constraints, similar to droplets in the usual Ising model below T_C. More detailed studies should be done to describe this observation. An interpretation in terms of the rigidity percolation of "physical clusters" suggests a close relation of the active bonds proposed by Coniglio and Klein with the redundant constrictions in both kinds of percolation, rigidity and connectivity [10, 14]. Also, the introduction of an external load that plays the role of a field in the usual Ising model, should be interesting to address in order to give place to disordered like phase counterpart.

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