Thermodynamics comparison of a monatomic systems modeled as an Antiferromagnetic Ising model on Husimi and cubic recursive lattices of the same coordination number

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Abstract

Two kinds of recursive lattices having the same coordination number but different unit cells (2-D square and 3-D cubic cells) are constructed and an antiferromagnetic Ising model is solved exactly on them to study stable and metastable states. The Ising model with multi-particle interactions is designed to represent a monatomic system or an alloy and exhibits the phenomenon of supercooled liquid and the ideal glass transition. Based on the solutions, the thermodynamics on both lattices was examined. In particular, the free energy, energy and entropy of the ideal glass, supercooled liquid, crystal and liquid state of the model on each lattice were calculated and compared. Interactions between particles farther away than the nearest neighbor distance are taken into consideration. The two lattices allow us to compare the effects of the unit cells on thermodynamics and to indicate the advantages and disadvantages of each lattice.
I. INTRODUCTION

The glass transition in which the amorphous state becomes brittle on cooling or soft on heating has been studied and investigated for many years. The situation is, however, is still unclear and remains controversial [1-6]. Numerous models and methods have been developed to study the glass transition [7] with regards to its thermodynamic or dynamic aspect. In this work, we are only interested in the thermodynamic aspect of the so-called ”equilibrium glass transition”, commonly known as the ideal glass transition, which is a hypothetical transition that is believed to occur in the limit of infinitely slow cooling rate. The hope is that a clear and complete understanding of the ideal transition will give us insight into the real glass transition that is observed in the laboratory. Among these efforts to study the ideal glass transition, one method developed in our group involves exact thermodynamic calculation on recursive lattices, such as a Bethe lattice or a Husimi lattice, and has been demonstrated to be a practical way to study the ideal glass transition [8-11]. As the name implies, recursive lattices are constructed recursively by using basic units of regular lattices to approximate them, and have the advantage to allow exact calculation [11-13]. Based on the structures of different recursive lattices, their capability to deal with systems with various complexity are different. In this paper, we are going to consider a Husimi lattice and compare its prediction with a cubic recursive lattice (CRL) [14] to be introduced below, both of which are designed to approximate the regular 3-D cubic lattice for which the coordination number $q$ is 6. We will specifically study an antiferromagnetic Ising model on them and solve it exactly to study various equilibrium and time-independent metastable phases. The calculation will allow us to compare the consequences on the two lattices.

II. RECURSIVE LATTICE GEOMETRY

A. Recursive Lattice Approximation

Lattices have been demonstrated to be a practical method to approximate and study a real system. For example, the Flory-Huggins theory [15, 16], which employs a lattice model, is a successful approach to investigate polymers. The regular lattices of 1-D, 2-D and 3-D are shown in Fig. 1.

Except in some rare cases [17, 18, 19], a many-body system with interactions on a regular
lattice is difficult to be solved exactly because of the complexity involved with treating the combinatorics generated by the interaction terms in the Hamiltonian when summing over all states. Usually the mean-field approximation is adopted to solve this problem. Such is the case with the Flory model of semiflexible polymers [15, 20]. On the other hand, recursive lattices enable us the explicit treatment of combinatorics on these lattices and no approximation is necessary [11, 29-32]. The recursive lattice is chosen to have the same coordination number as the regular lattice it is designed to describe. As usual, the coordination number \( q \) is the number of nearest-neighbor sites of a site. This number is 2 in the linear lattice (1-D case), 4 in the square lattice (2-D case) and 6 in cubic lattice (3-D case).

Typical recursive lattices, the Bethe lattice [21] and the Husimi lattice, along with the new cubic recursive lattice (CRL), as the analogs of the 1-D, 2-D and 3-D lattices shown in Fig. 1 are shown in Fig. 2. In Husimi lattice shown in Fig. 2 the coordination number is 4, which is the same as in a regular square lattice, while in CRL the coordination number is 6 to match the coordination number of a regular cubic lattice. On the other hand, the 3-coordinated Bethe lattice may be used to approximate an hexagonal lattice. The recursive lattice calculations have been demonstrated to be highly reliable approximations to regular lattices [11]. The most impressive property of these recursive lattices is that a certain number

\[
\text{FIG. 1: Regular 1-D, 2-D and 3-D lattices.}
\]
of independent lattice units are joint at a site recursively (see below for a clear prescription) to give rise to a tree-like structure. To see this, imagine replacing each unit (square or a cube) by a point and connect each connected pair of units by a bond. Here, a connected pair shares a common face of a dimension one less than the units. Thus, for two cubes in a cubic lattice are connected when they share a common square. This will result in Bethe lattice, which is a tree and is the most simple recursive lattice. We can use any kind of an artificial geometrical structure, called a unit cell, containing any number of sites to construct a recursive lattice with the provision that a unit is only connected to the neighbor unit by a shared site, and the neighbor unit connects to the next neighbor unit by the next shared site, and so on in a recursive fashion. In Fig. 2, the unit in the Bethe lattice is a line segment with two sites on the ends. The unit in Husimi lattice is a square formed by four sites. And in CRL, the basic unit is a cube. We label the unit at the center of the tree by \( m = 0 \) and all other units by \( m = 1, 2, 3, ... \) as we move outwards from the center. The labeling is shown explicitly for the Bethe lattice, but the same scheme applies to all the recursive lattices that have a tree structure. It follows from the tree structure that the units on level 1 (e.g. the three sites on the circle 1 in the Bethe lattice figure) are not connected directly with each other; their indirect connection is only through the unit at level 0. Of course, each unit at level \( m \) is connected to one unit at level \( m - 1 \) and \( q - 1 \) units at level \( m + 1 \).
FIG. 3: A 3-branched Husimi lattice. Only few branches are drawn. Note that all the diamonds are equal in size although they cannot be drawn in that way here. All the sites have the same number of neighbors.

B. Construction of a Recursive Lattice

We described how to construct a recursive lattice to approximate a regular lattice with the same coordination number. The Husimi lattice is formed by joining two basic square units of the regular square lattice at each site to form an approximation of the square lattice. And it is the same for CRL. This may cause a misunderstanding that the recursive lattice of a given coordination number $q$ can only be constructed out of a single unit of a particular structure, which is not true. We first recognize that the coordination number alone does not determine the structure of basic unit. For example, we can modify the above Husimi lattice by joining three square units at each site, see Fig. B to result in $q = 6$. But such a recursive lattice is very different from the CRL. In this case, we may ask the following questions: Is this modified Husimi lattice also capable to approximate a 3-D regular lattice like a CRL based on the same coordination number? If yes, what advantages or disadvantages does it have compared to the CRL? These questions will be investigated in this work.

III. ANTIFERROMAGNETIC ISING MODEL

An Ising spin $S$ has two possible values, which usually are assigned as $+1$ (up or +) and $-1$ (down or −) [17, 18, 22] and can also represent a particle or void or particle A or B in an alloy, respectively. The Ising model with nearest-neighbor interactions between neighboring spins $S_i$ and $S_j$ and in the presence of a uniform magnetic field $H$ has its energy given by
\[ E = -J \sum_{<i,j>} S_i S_j - H \sum S_i, \]

where \(<i,j>\) is the nearest-neighbor pair of lattice sites \(i\) and \(j\) and \(S_i = \pm 1\) is the spin at site \(i\). The interaction has the value \(-J\) when the two neighboring spins are parallel, and the value \(+J\) when they are antiparallel.

The Ising spin \(S = 1\) to be also denoted by \(+\), \(S = -1\) to be also denoted by \(-\) can also represent a particle or void or particle A or B in an alloy, respectively.

For the ferromagnetic case, \(J > 0\), and favors parallel arrangement at absolute zero, so that the spins have a ferromagnetic ordering. This means that in the case of an alloy, we either have all A or all B particles at absolute zero. This state does not represent the alloy ordering in an alloy. The antiparallel arrangement at absolute zero, see Fig. 4 is favored by the antiferromagnetic interaction \(J < 0\). This ordering represents the perfect ordering in an alloy. Thus, we will focus on the antiferromagnetic case here for which the low temperature equilibrium phase will represent a crystalline ordering in an alloy or a pure system. The ordered spin arrangement becomes more disordered which corresponds to a crystal at positive temperature until it melts into a liquid with the increase of temperature. This liquid can be supercooled to a metastable state, which we will call the supercooled liquid. As we will see, this state is disordered and will eventually yield a glass at a low enough temperature.

The standard Ising model only includes the interaction between the nearest neighbor spins. However it is not realistic that particles farther away than the nearest distance do not interact with each other in real systems. For a better approximation of a real system, more interaction energy terms such as the interactions between second neighbor sites, interaction between three and four spins are employed in our model.

IV. MODEL, FIX-POINT SOLUTIONS AND THERMODYNAMICS

The calculation of Husimi lattice and CRL are similar in spirit. As a basic unit or cell at a higher level shares a single site connecting it to the lower level, this site in each unit plays a role different from the other sites. We will call this the base site and the corresponding spin at that site will be generically be denoted by \(S^b\). In particular, we can define the energy \(e_\alpha\) associated with a cell \(\alpha\) by paying special attention to this site in such a way that the sum
over all cells will give the energy $E$ of the model over the entire recursive lattice. Here we will take the multi-branched Husimi lattice for example to demonstrate the calculation method, and briefly give the generalized formula for CRL, for which the details of calculations were described in our previous work [14]. We will, however, provide all relevant details here.

A. Spins Conformation

We first consider a Husimi lattice made up by squares as the unit cells. The four spins in such a square with its base site at the $m$-th level are shown in Fig. 5 with $S_m$ at the base site. The peak site is at level $m + 2$ and the remaining intermediate sites are at level $m + 1$. The spins at the intermediate sites are denoted by $S_{m+1}$ and $S'_{m+1}$, respectively. The energetics of the model is defined by considering these squares. We now introduce following quantities for a square cell:

$$A_{\text{mag}} = S_{m+1} + S'_{m+1} + S_{m+2};$$
$$A_{\text{nb}} = S_m S_{m+1} + S_0 S'_{m+1} + S_{m+1} S_{m+2} + S'_{m+1} S_{m+2};$$
$$A_{\text{sd}} = S_m S_{m+2} + S_{m+1} S'_{m+1};$$
$$A_{\text{tri}} = S_m S_{m+1} S'_{m+1} + S_m S'_{m+1} S_{m+2} + S_m S_{m+1} S_{m+2} + S_{m+1} S'_{m+1} S_{m+2};$$
$$A_{\text{qua}} = S_m S_{m+1} S'_{m+1} S_{m+2}.$$

Note that the base site is not included in the magnetic term $A_{\text{mag}}$; instead we just count the contribution from the sites of the unit cell above the site $S_m$. The missing contribution from this site will be accounted for when we construct the recursion relations below.
Let \( \gamma \) denote a particular spin state in the cell. The energy of the Ising model in the cell for a particular cell state \( \gamma \) is:

\[
e(\gamma) = -J_{A_{ab}}(\gamma) - J_P A_{sd}(\gamma) - J' A_{tri}(\gamma) - J'' A_{qua}(\gamma) - H A_{mag}(\gamma),
\]

where \( J_P \) is the interaction energy between the second nearest sites, \( J' \) the interaction energy of three sites (triplet), and \( J'' \) the interaction energy of the four sites (quadruplet).

The total energy of the Ising model mentioned above is the sum of the energy of all cells \( \alpha \) on our recursive lattice:

\[
E(\Gamma) = \sum_{\alpha} e(\gamma_{\alpha}) - HS_0,
\]

where \( \Gamma = \bigotimes_{\alpha} \gamma_{\alpha} \) denotes the spin state of the lattice, and where \( S_0 \) now represents the spin at the origin of the recursive lattice. There are four sites in a square cell; hence, the number of possible states of a cell is \( 2^4 = 16 \). The 1st to the 8th states correspond to the base spin \( S_m = +1 \), and the 9th to the 16th states to \( S_m = -1 \) on the base site. The number of lattice conformations \( \Gamma \) for a lattice with \( N_C \) cells is \( 16^{N_C/r} \).

The Boltzmann weights are given by

\[
w(\gamma) = \exp(-\beta e(\gamma)), \quad w(\Gamma) = \exp(\beta HS_0) \prod_{\alpha} w(\gamma_{\alpha}).
\]

The partition function (PF) is a sum over lattice configurations:

\[
Z(T) = \sum_{\Gamma} w(\Gamma).
\]

By choosing proper values of interaction energies, we can simulate various systems to study their thermodynamic properties and phase transition under different conditions. As a convention, we set \( J = -1 \) to set the temperature scale for our antiferromagnetic model.

**B. Fix-point solution**

Since units just join together at one site, the connected units on higher levels from some site may be treated as a branch at this site. The branch \( T_m \) is obtained by cutting the lattice at the \( m \)-th site; see Fig. 6. This branch is said to have its base site at the \( m \)-th level. A partial partition function (PPF) \( Z_{m+1}(S_{m+1}) \) for the branch \( T_{m+1} \) with its base
at the level $m + 1$ can be introduced to represent the contribution of the branch with spin $S_{m+1}$ as its base site. Then the PPF $Z_m(S_m)$ at the lower level $m$ can be expressed in terms of PPF’s $Z_{m+1}(S_{m+1}), Z_{m+1}(S'_{m+1})$ and $Z_{m+2}(S_{m+2})$ at higher levels as discussed below; see Fig. 6. Thus, recursively we can start from an initial site at the highest level from the origin to calculate the contribution of the entire lattice. In this way, we can obtain the thermodynamics of the entire system.
For a multi-branched lattice, we introduce a coefficient $r$ as the number of branches joined at a site. The partition function $Z_0$ of the lattice can be represented by the contribution to the origin site from the $r$ branches $T_0$ meeting at the origin with either $S_0$:

$$Z_0 = Z_0^r(+)e^{\beta H} + Z_0^r(-)e^{-\beta H}$$ (5)

The contribution from the magnetic field above is due to the contribution from the base site that is not included in $A_{mag}$.

We now turn to the PPF’s. As each PPF at a base site in the square is a sum of the 8 configurations of the remaining three sites in the square, we have ($r' \equiv r - 1$)

$$Z_m(+) = \sum_{\gamma=1}^{8} Z_{m+1}(S_{m+1})Z_{m+1}'(S_{m+1}')Z_{m+2}(S_{m+2})w(\gamma),$$ (6)

$$Z_m(-) = \sum_{\gamma=0}^{16} Z_{m+1}(S_{m+1})Z_{m+1}'(S_{m+1}')Z_{m+2}(S_{m+2})w(\gamma),$$ (7)

with a product from the $r'$ branches at the two sites on level $m + 1$ and the $r'$ branches at the peak site at $m + 2$.

We now introduce the ratios

$$x_m = \frac{Z_m(+)}{Z_m(+) + Z_m(-)}, \quad y_m = \frac{Z_m(-)}{Z_m(+) + Z_m(-)}.$$ (8)

and

$$z_m(S_m) = \begin{cases} x_m & \text{if } S_m = +1 \\ y_m & \text{if } S_m = -1 \end{cases}$$ (9)

In terms of

$$B_m = Z_m(+) + Z_m(-),$$

we have $Z_m(+) = B_mx_m$ and $Z_m(-) = B_my_m$. By use of (6), we obtain

$$Z_m(\pm) = \sum z_{m+1}^{r'}(S_{m+1})z_{m+1}'(S_{m+1}')z_{m+2}^{r'}(S_{m+2})w(\gamma)/Q(x_{m+1}, x_{m+2}),$$

where the sum is over $\gamma = 1, 2, 3, \ldots, 8$ for $S_m = +1$, and over $\gamma = 9, 10, 11, \ldots, 16$ for $S_m = -1$, and where

$$Q(x_{m+1}, x_{m+2}) = \left[B_m/B_{m+1}^2B_{m+2}\right]^{r'};$$

10
it is related to the polynomials

\[
Q_+ (x_{m+1}, x_{m+2}) = \sum_{\gamma=1}^{8} z'^\gamma_{m+1} (S^\gamma_{m+1}) z'^\gamma_{m+1} (S'^\gamma_{m+1}) z'^\gamma_{m+2} (S_{m+2}) w(\gamma),
\]

\[
Q_- (x_{m+1}, x_{m+2}) = \sum_{\Gamma=9}^{16} z'^\Gamma_{m+1} (S^\Gamma_{m+1}) z'^\Gamma_{m+1} (S'^\Gamma_{m+1}) z'^\Gamma_{m+2} (S_{m+2}) w(\gamma),
\]

according to

\[
Q(x_{m+1}, x_{m+2}) = Q_+(x_{m+1}, x_{m+2}) + Q_-(y_{m+1}, y_{m+2}).
\]

In terms of the above polynomials, we can express the recursive relation for the ratio \(x_m\) in terms of \(x_{m+1}\) and \(x_{m+2}\):

\[
x_m = \frac{Q_+(x_{m+1}, x_{m+2})}{Q(x_{m+1}, x_{m+2})}.
\]

(10)

A similar relation holds for \(y_m \equiv 1 - x_m\):

\[
y_m = \frac{Q_-(x_{m+1}, x_{m+2})}{Q(x_{m+1}, x_{m+2})}.
\]

(11)

The entire bulk thermodynamic calculation just focuses on solving Eqs. (10, 11) and determine the fix-point (FP) solution. Because of the recursive property of these equations, we would expect cycle-form solutions, which is called fix-point solution. It is clear from the definition of \(x\) that it determines the probability that a site is occupied by the spin \(S = +1\).

At high temperatures, we find a uniform FP solution \(x\) on various levels, which we call a 1-cycle solution:

\[
x = \frac{Q_+(x, x)}{Q(x, x)}.
\]

At low temperatures, we find alternate solutions \(x_1, x_2\) on two successive levels, which we call a 2-cycle solution:

\[
x_1 = \frac{Q_+(x_2, x_1)}{Q(x_2, x_1)}, x_2 = \frac{Q_+(x_1, x_2)}{Q(x_1, x_2)}.
\]

This describes the antiferromagnetic ordering. Because of this ordering at low temperatures, a three or higher cycle solutions is hard to imagine. Although we cannot prove
that there are no other solutions than the 1- and 2-cycle solutions, we have not found any. From the definition of the recursive equation it is clear that the solutions depends on the interaction energy parameters of the system and the temperature $T$. A typical $x$ solution for the 2-branched Husimi lattice ($r = 2$), with $J = -1$, $J_p = 0$, $J' = 0$, $J'' = 0$ and $H = 0$ on a wide temperature range is shown in Fig. [7]. From Fig. [7] we can see that the 1-cycle solution is $x = 0.5$ everywhere, which implies the probability of a site occupied by $S = +1$ is fifty percent; note that there is no magnetic field $H$ in the Hamiltonian. This situation corresponds to a disordered phase and represents a high free energy state at low temperature (supercooled liquid) [1, 2, 23]. For the 2-cycle solution, one branch approaches to 1 and the other branch approaches to 0 as $T \to 0$; thus we have $S = +1$ and $S = -1$ alternatively occupying neighboring sites in the lattice at absolute zero. This structure represents the ordered states (crystal) and is characterized by the complete antiferromagnetic order in the Ising model. At high temperatures, the 2-cycle solution turns to the 1-cycle solution and there occurs a transition at melting temperature $T_K = 2.75$, where the crystal state turns into the disordered liquid state. This temperature represents the melting transition. [24, 25] Obviously, the simple Ising model, which we will call the reference model in this work, is not realistic enough to give a discontinuous melting.

C. FP Solution and the Bulk Thermodynamics

1. Free Energy

It is clear that the 1-cycle solution is a special case of the 2-cycle solution with $x_1 = x_2$. Therefore, we will assume a 2-cycle solution and determine its the free energy. If it happens that $x_1 = x_2$, we obtain the 1-cycle free energy. The solution with a higher free energy will then describe a metastable state, which will be useful below to identify the supercooled liquid and the glass.

The Helmholtz free energy is given by

$$ F = -T \log Z $$

and requires obtaining the partition function $Z$. As the lattice is infinitely large, there is no sense in calculating $Z$ or $F$, each of which will be infinitely large. At the FP solution, we
FIG. 7: Reference $x$ solutions corresponding to the 2-cycle at low temperatures and the 1-cycle at higher temperatures ($J = -1$ and other parameters zero) for $r = 2$ Husimi lattice. Note the singular behavior around $T = 2.75$.

can use its homogeneity over two levels to obtain the free energy per site by following the Gujrati trick [11], which we briefly describe below. We consider the Husimi lattice made up by connecting $r$ squares at each site. The partition function of the whole system is given by considering the $r$ branches $T_0$ meeting at the origin. It is given by Eq. (5).

Let us image that we cut off the $rr'$ branches $T_2$ on level 2 and hook up them together to form $r'$ smaller lattices. The partition function of each of these smaller lattice is:

$$Z_2 = Z_2^r (+) e^{\beta H} + Z_2^r (-) e^{-\beta H}.$$  

Similarly we can image to cut off the $2rr'$ branches $T_1$ and hook them up to form $2r'$ smaller
lattices. The partition function for each smaller lattice is

\[ Z_1 = Z_1^r (+) e^{\beta H} + Z_1^r (-) e^{-\beta H}. \]

Since free energy is an extensive quantity, the free energy of the whole system is the sum of the free energies of the smaller lattices and the \( r \) local squares that are left out after dissecting the original lattice. These squares contain \( 2r \) sites. Their free energy is given by

\[ F_{\text{local}} = -T \log \left[ \frac{Z_0}{(Z_1^r Z_2^r)^r} \right]. \]

Thus, the free energy per site is:

\[ F = -\frac{F_{\text{local}}}{2r}. \]  

(12)

By substituting \( Z_m(+) = B_m x_m \) and \( Z_m(-) = B_m y_m \), we finally obtain

\[ F = -\frac{1}{2r} kT \log \left( Q^{2r} \frac{1}{\left\{ [x_1^r e^{\beta H} + (1 - x_1)^r e^{-\beta H}]^r + \left[ x_2^r e^{\beta H} + (1 - x_2)^r e^{-\beta H} \right]^r \right\}^r} \right) \]  

(13)

Using the FP solution \( x_1 \) and \( x_2 \) from fix-point calculation, the numerical value of free energy can be easily achieved.

2. Conformation Probability

A square unit with 4 spins on each site has sixteen possible conformations \( \gamma \). The probability of a given conformation \( \gamma \) is

\[ P(\gamma) = \frac{[Z_0(S_0)Z_1(S_1)Z_1(S_1')Z_2(S_2)]^r \exp(\beta HS_0)w(\gamma)}{Z_0}; \]

It obviously satisfies the sum rule

\[ \sum_{\Gamma}^1 P(\gamma) = 1. \]

It is easy to see that

\[ P(\gamma) = \frac{[z_0(S_0)z_1(S_1)z_1(S_1')z_2(S_2)]^r \exp(\beta HS_0)w(\gamma)}{Q[x_0^r e^{\beta H} + (1 - x_0)^r e^{-\beta H}]} \]
3. Energy Density

The energy density is defined as the summation over the product of the energy and the probability of a conformation state. Once we have the conformation probabilities, energy density (per site) can be simply calculated by evaluating the sum of the product of energy of each conformation and the conformation probability:

\[ E = \frac{1}{2r} \sum e(\gamma)P(\gamma) \]
where
\[ e_\alpha = -J A_{nb} - J_P A_{sd} - J'_A A_{tri} - J'' A_{qua} - H(S^b_\alpha + A_{mag})/r \]
is the energy of a unit cell \( \alpha \) in a configuration \( \gamma \) as mentioned before. The base site \( S^b_\alpha \) is included in the magnetic term as it contributes to the energy of each cell.

4. Entropy

With the free energy and energy density, by the thermodynamic definition we can simply obtain the entropy by
\[ S = \beta (E - F), \]
or by the first derivative of the free energy with respect to the temperature:
\[ S = -\partial F/\partial T. \]

A negative entropy is possible to be obtained from 1-cycle solution at the some temperature \( T_K \) below the melting transition temperature \( T_M \), since it represents the disordered supercooled liquid. However negative entropy is known as unphysical in nature, thus the metastable state must undergo a transition and change to the glass state at \( T_K \), which in this way is defined as the ideal glass transition temperature.

D. The CRL calculation

The CRL calculation is similar to the method introduced in previous section and can be found in ref. [14]. Here we briefly give the scheme of the CRL calculation. For cubic recursive lattice, the sites on a cube unit are divided into four layers and labeled as follows:
\[ S_m, (S_{m+1,1}, S_{m+1,2}, S_{m+1,3}), (S_{m+2,1}, S_{m+2,2}, S_{m+2,3}), S_{m+3}; \]
see Fig. 9 where we have taken \( m = 0 \).

The sub-tree structures with three sub-trees at each site are easily defined by considering Fig [10].

Taking their contributions at each site easily gives the recursion relations for the PPFs:
FIG. 9: The sites label in a cube unit of CRL

FIG. 10: The sub-tree contributions in CRL
\[Z_m(+) = \sum_{\gamma=1}^{128} \left[ \prod_{\{m+1\}} z_{m+1}(S_{m+1}) \right] \left[ \prod_{\{m+2\}} z_{m+2}(S_{m+2}) \right] z_{m+3}(S_{m+3}) w(\gamma),\]

\[Z_m(-) = \sum_{\gamma=129}^{256} \left[ \prod_{\{m+1\}} z_{m+1}(S_{m+1}) \right] \left[ \prod_{\{m+2\}} z_{m+2}(S_{m+2}) \right] z_{m+3}(S_{m+3}) w(\gamma),\]

By introducing \(B_m\) and the ratios \(x_m\) and \(y_m\), and \(z_m(S_m)\) as before, we have

\[Q_+(x_{m+1}, x_{m+2}, x_{m+3}) = \sum_{\gamma=1}^{128} \left[ \prod_{\{m+1\}} z_{m+1}(S_{m+1}) \right] \left[ \prod_{\{m+2\}} z_{m+2}(S_{m+2}) \right] z_{m+3}(S_{m+3}) w(\gamma),\]

\[Q_-(x_{m+1}, x_{m+2}, x_{m+3}) = \sum_{\gamma=129}^{256} \left[ \prod_{\{m+1\}} z_{m+1}(S_{m+1}) \right] \left[ \prod_{\{m+2\}} z_{m+2}(S_{m+2}) \right] z_{m+3}(S_{m+3}) w(\gamma),\]

and

\[Q(x_{m+1}, x_{m+2}, x_{m+3}) = Q_+(x_{m+1}, x_{m+2}, x_{m+3}) + Q_-(y_{m+1}, y_{m+2}, y_{m+3}) = \frac{B_m}{B_{m+1}B_{m+2}B_{m+3}}.\]

We can now obtain the recursive relation for the ratio \(x_m\) in terms of \(x_{m+1}, x_{m+2}\) and \(x_{m+3}\):

\[x_m = \frac{Q_+(x_{m+1}, x_{m+2}, x_{m+3})}{Q(x_{m+1}, x_{m+2}, x_{m+3})}.\]

A similar relation holds for \(y_m = 1 - x_m\):

\[y_m = \frac{Q_-(y_{m+1}, y_{m+2}, y_{m+3})}{Q(y_{m+1}, y_{m+2}, y_{m+3})}.\]

For the CRL case, we also have the 1-cycle solution, \(x_m = x_{m+1} = x_{m+2} = x_{m+3}\), which represents the disordered state, and the 2-cycle solution, \(x_m = x_{m+2}\) and \(x_{m+1} = x_{m+3}\), which represents the ordered stable state. Thermodynamic properties such as free energy and entropy can be calculated from the solutions using similar techniques as before. The local area around the origin is chosen to be the two cubes joint on the origin point, and it has eight sites. The final expression of the free energy per site is:

\[F = \frac{1}{8} F_{local} = -\frac{1}{8} kT \log(Q^2 \left[ x_1^2 e^{\beta H} + (1 - x_1)^2 e^{-\beta H} \right] \left[ x_2^2 e^{\beta H} + (1 - x_2)^2 e^{-\beta H} \right]^4) - \frac{1}{2} \left[ x_1^2 e^{\beta H} + (1 - x_1)^2 e^{-\beta H} \right] \left[ x_2^2 e^{\beta H} + (1 - x_2)^2 e^{-\beta H} \right]^2 \tag{14}\]
V. RESULTS AND DISCUSSION

The thermodynamic calculations have been done on 3-branched Husimi lattice and CRL. Since both models are to approximate 3-D regular lattice, the results will be compared in this section. The role of energy parameters will be studied in both cases..

A. General solutions with \( J = -1 \) and all other parameter are zero

For a reference comparison, we keep \( J = -1 \) and set all other parameters to zero in both \( r = 3 \) Husimi lattice and the CRL. The 1-cycle and 2-cycle solutions are shown in Fig. 11 and 12. The solutions in Fig. 7 and 11 and 12 clearly show that, with branch number 3 in Husimi lattice the melting temperature increases significantly and becomes very close to the one in CRL. The melting temperature is found to be 4.84 in CRL and 4.89 in 3-branched Husimi lattice. These values are comparable to the results obtained by other methods on the Ising antiferromagnetic model on a simple cubic lattice: 4.51 by Monte Carlo [26], 4.93 by mean field renormalization group [27] and 4.52 by series expansion [28]. We clearly see that the singular temperature moves towards the ”correct” temperature 4.51 or 4.52 obtained by simulation or series analysis as we increase the coordination number in the case of the two Husimi lattices or use a more realistic the unit cell. However, they drift away from the mean field renormalization group result.

The difference between the 2-cycle solutions in both lattices is also remarkable. It is obvious that Husimi lattice with \( r = 3 \) is more suited to describe the 3-D case rather than the 2-D case according to our expectation. However, in Fig. 11 the curves are more like stretched curves in Fig. 7 while the solutions of CRL in Fig. 12 are more rounded curves. The difference between \( x_0 \) and \( x_1 \) is larger in CRL. Since the 1 and 0 cyclic solution at the zero temperature corresponds to the ideal crystal stable state, a 2-cycle can be identified to be more stable if its two branches are closer to 1 and 0 respectively, or vice versa, in this way the 2-cycle solutions in CRL are more stable below the melting temperature \( T_M \) than the 3-branched Husimi lattice. This difference can also be observed in thermal properties given in Fig. 13 and 14.

The entropy curves of 1-cycle solution (supercooled liquid) in Fig. 13 and 14 go to negative values at some low temperature, below the melting transition, which demonstrates
FIG. 11: Reference solutions of the 2-cycle and the 1-cycle for $r = 3$ Husimi lattice with $J = -1$ and other parameters zero.

the Kauzmann paradox [8, 9], or the ideal glass transition. The Kauzmann paradox was originally referred to where the entropy of supercooled liquid comes to be lower than the crystal entropy. This is because at the same temperature the kinetic entropy of crystal and supercooled liquid are the same, and the entropy of crystal was approximated to be the kinetic entropy, then the difference between the entropy of crystal and supercooled liquid, i.e. the excess entropy, equals to the configurational entropy which cannot be negative. However the entropy of crystal at non-zero temperature is not only contributed by kinetic entropy, it also has entropy from defects. In this way it is possible to have a supercooled liquid state with less defects and lower entropy than the crystal state at the same temperature. Thus we take the negative entropy of supercooled liquid as the paradox. At the melting transition, the entropy of 2-cycle solution falls dramatically due to crystallization, while the entropy of 1-cycle solution continue to decrease gradually following the behavior of supercooled liquid. The rapid drop in the entropy in the 2-cycle reflects that the current model is too unrealistic to capture the discontinuity in entropy due to crystallization and a better model might
produce a discontinuity. This adds faith that antiferromagnetic model as not too far from a realistic model. With the lowering of the temperature, the entropy of supercooled liquid goes negative at some point, which is unacceptable. Thus there must exist a transition to avoid the negative entropy, the so called ideal glass transition, and the transition temperature is called the Kauzmann temperature $T_K$.

**B. The effect of $J_p$**

Beside the interaction between the nearest spins $J$, the interactions between spins farther apart also play important roles in our lattice model. With the set of various values of these parameters, the model is capable to simulate many different cases, i.e. to manipulate the transition temperatures. As we vary the interaction along the surface diagonal $J_P$, it significantly changes the transition temperatures as shown in Figs. 15 and tables 1, 2.
FIG. 13: Reference thermal properties of the 2-cycle and the 1-cycle case for \( r = 3 \) Husimi lattice \((J = -1 \text{ and other parameters are zero})\).

Table 1. The transition temperature variation with different \( J_P \) in CRL

| \( J_P \) | \( T_M \) | \( T_K \) |
|---------|--------|--------|
| -0.1    | 4.2    | 1.6    |
| -0.2    | 3.5    | 1.3    |
| -0.3    | 2.8    | 1.1    |
| -0.4    | 2.0    | 0.8    |
| -0.5    | 1.1    | 0.6    |
| 0.3     | 6.6    | 2.4    |
| 0.5     | 7.7    | 2.9    |
FIG. 14: Reference thermal properties of the 2-cycle and the 1-cycle case for CRL ($J = -1$ and other parameters are zero).

FIG. 15: The transition temperature variation with different $J_P$ in CRL and the Husimi lattice.
Table 2. The transition temperatures variation with different $J_P$ in the Husimi lattice

| $J_P$ | $T_M$ | $T_K$ |
|-------|-------|-------|
| -0.1  | 4.6   | 1.4   |
| -0.2  | 4.3   | 1.3   |
| -0.3  | 4.0   | 1.2   |
| -0.4  | 3.6   | 1.1   |
| -0.5  | 3.2   | 1.0   |
| 0.3   | 5.8   | 1.8   |
| 0.5   | 6.4   | 2.0   |

In both case, the increase of $J_P$ increases the transition in a linear fashion. From Eq. [1] it is easy to see the negative value of $J_P$ competes with negative $J$. The negative $J$ prefers different states of the nearest spins and consequently the same states of the spins on surface diagonal, while negative $J_P$ forces the diagonal spins to be in different states. Thus, in both the CRL and the Husimi lattice the decrease of $J_P$ decreases the transition temperature, that is, makes it easier for the system to be melted. On the other hand, positive $J_P$ favors antiferromagnetic spin configuration and increase the transition temperature for the same reason. In CRL, the role of $J_P$ is more significant than it is in the Husimi lattice, because a cubic unit has 12 surface diagonal interactions and 12 nearest interactions, while a square unit has 4 $J$’s and 2 $J_P$’s therefore the diagonal interactions have less weight.

C. The effect of $J'$

An important feature of $J'$ is that its sign (positive or negative) does not affect the solutions. With the increase of $J'$, the transition temperature decrease although by a very small amount in CRL, and much smaller in the Husimi lattice. That is because the three-spins interactions would disturb the ordered state formation; nevertheless its contribution is very small in the total energy in both cases.

Another interesting effect of $J'$ is that, although the transition temperature change weakly with $J'$, it changes the thermodynamic curves dramatically, and with $J' = 1$ it could even destroy the ideal glass transition by having the supercooled liquid continue to zero temperature, as shown in Fig. [17]
FIG. 16: The transition temperature variation with different $J'$ in CRL and the Husimi lattice.

Table 3. The transition temperature variation with different $J'$ in CRL

| $|J'|$ | $T_M$ | $T_K$ |
|------|------|------|
| 0.3  | 4.8  | 1.7  |
| 0.6  | 4.7  | 1.4  |
| 0.8  | 4.6  | 1.1  |
| 1.0  | 4.3  | N/A  |

Table 4. The transition temperature variation with different $J'$ in Husimi lattice

| $|J'|$ | $T_M$ | $T_K$ |
|------|------|------|
| 0.3  | 4.9  | 1.5  |
| 0.6  | 4.8  | 1.4  |
| 0.8  | 4.7  | 1.3  |
| 1.0  | 4.7  | 1.2  |
FIG. 17: The transition temperature variation with different $J'$ in CRL

**D. The effect of $J''$**

The roles of four-spins interaction $J''$ in CRL and the Husimi lattice are very different. In the CRL, there are six squares in one cubic unit. the value of $J''$ affects the spins states the same way as $J_P$, i.e. it increases the transition temperature. However it does not change $T_M$ and $T_K$ in the Husimi lattice because the 4-spins interaction is mainly the interaction of a whole unit, thus its effect is like a magnetic field $H$ for each single spin.
FIG. 18: The transition temperature variation with different $J''$ in CRL

| $J''$ | $T_M$ | $T_K$ |
|-------|-------|-------|
| -0.5  | 3.9   | 1.2   |
| -0.9  | 3.1   | 0.3   |
| -1.0  | 2.8   | N/A   |
| 0.5   | 5.5   | 2.3   |
| 0.9   | 6.1   | 2.7   |

Table 5. The transition temperature variation with different $J''$ in CRL

| $J''$ | $T_M$ | $T_K$ |
|-------|-------|-------|
| -0.5  | 4.9   | 1.5   |
| -0.9  | 4.9   | 1.5   |
| -1.0  | 4.9   | 1.5   |
| 0.5   | 4.9   | 1.5   |
| 0.9   | 4.9   | 1.5   |

Table 6. The transition temperature variation with different $J''$ in Husimi lattice
E. Conclusion

The Ising model on a recursive lattice enables us to do exact calculations without any approximation to study the supercooled liquid, the crystal formation and phase transitions. We have constructed a multi-branched 2D Husimi lattice to represent a model with higher dimension and a cubic recursive lattice of the same coordination number to simulate a 3D system. The branch number is set to be 3 in the multi-branched 2D Husimi lattice to make the coordination number to be 6. Because of the same coordination number, both lattice are designed to approximate regular 3D lattice. We applied the antiferromagnetic Ising model on the two lattices and the alternative spin arrangement to represent the ordered phase i.e. crystal state, whereas the homogeneous spins arrangement represents the metastable state, i.e. the liquid and the supercooled liquid states. A set of solutions relating to the probability that one site being occupied by a particular spin state can be exactly calculated on the lattice from the ratio of partial partition functions. Then the thermal properties of the two models were calculated from the solutions and compared with each other. Two kinds of solutions, 2-cycle and 1-cycle solutions, representing the ordered and disordered states respectively, give the thermodynamics of crystal and metastable states. The behavior of the free energy and the entropy of two states allows us to locate the melting transition at $T_M$ and the Kauzmann temperature $T_K$. The $T_M$ is determined by the differing of two states with temperature decreasing, above that temperature two solutions have the same disordered behavior as liquid. When the system goes below the $T_M$ an ordered state (from 2-cycle solution) appears with lower free energy and entropy, while the disordered solution (1-cycle solution) still exists and the system is possible to continue in supercooled state without phase transition. The entropy of this disordered state will gradually decrease to zero and then negative at a non-zero temperature, which is determined to be the ideal glass transition temperature, or Kauzmann temperature $T_K$.

The transition temperatures determined on both lattices are very close to each other, and fairly agrees with the results from other methods such as MC simulation and series analysis. The results confirms our expectation that 1) CRL is a good approximation of regular 3D cubic lattice, and 2) with the same coordination number the multi-branched Husimi lattice is also capable to describe systems in higher dimension.

In addition to the nearest neighbor interactions, the effects of second nearest neighbor
interaction, interaction between triplets and quadruplet are studied in this work. All their effects are similar for the two lattices except $J''$. This enables us to obtain expected transition temperature or thermodynamic behaviors by adjusting the interaction parameters for an extensive use of our models. We conclude that the 3-branched Husimi lattice provides a less versatile and accurate method compared to CRL. For example, the latter provides a transition temperature closer to that obtained by series analysis than the one obtained by the Husimi lattice. However, the multi-branched Husimi lattice calculation is simpler and less time-consuming, which still prove very helpful in applications.

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