A Simplified Model of Phase Transition in Amorphous Antiferromagnets. II

Effect of the Magnetic Field

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Investigation of a simplified statistical model of Ising amorphous antiferromagnets is continued. The effect of the magnetic field on various thermodynamical quantities is studied with use of the Monte Carlo method. It is found that application of a certain range of finite magnetic field changes nature of the spin freezing transition qualitatively. The low temperature phase becomes essentially like “ferrimagnetic” when viewed in a properly defined “curved” space. The field cooled magnetization at low temperatures has a plateau region versus the magnetic field whereas the zero field cooled magnetization shows almost linear behavior.

§ 1. Introduction

In a previous paper (referred to as (I) hereafter), we investigated thermodynamical properties and nature of the spin freezing phenomena of amorphous antiferromagnets with use of a simplified two-dimensional model and the Monte Carlo technique. The model consists of Ising spins on a spatially random lattice in two dimensions which is constructed by closely packing the plane with equilateral triangles and squares (triangular-square lattice). By changing the ratio between the numbers of triangles and squares, the lattice can be varied continuously from the regular triangular lattice to the square lattice through various kinds of amorphous lattices. When this ratio takes a certain intermediate value, the lattice becomes highly amorphous and can be regarded as a reasonable structural model of amorphous materials (see Fig. 2 in (I)). Note that the short-range order is always preserved in this lattice in the sense that all bonds have equal lengths. Each Ising spin is put on every vertex of this lattice and is assumed to be coupled antiferromagnetically with nearest neighboring spins. In terms of the “frustration” concept, the triangular unit causes frustration whereas the square unit does not, and the low temperature properties of this model are somewhat interesting. In (I), we generated the most random lattice configuration by the computer which corresponds to the highest entropy value, and carried out some Monte Carlo simulations for the antiferromagnetic Ising model on this sample to investigate various thermodynamical properties under zero magnetic field.

It was found that the system exhibits a cooperative spin freezing phenomenon at a certain finite temperature whereas the susceptibility rises monotonically without showing any clear anomaly. The results were analyzed in comparison with some experimental results for amorphous antiferromagnets. In the present paper, we shall investigate the effect of the magnetic field with use of the same model and the Monte Carlo technique in order to study how the magnetic field influences the thermodynamical properties of amorphous antiferromagnets.
The most impressive finding of the present calculation is the following. When the system is cooled under the constant magnetic field of a certain intermediate strength, the large ground state degeneracy which exists in zero or small fields is drastically decreased and the low temperature spin configuration becomes essentially like "ferrimagnetic". This does not mean that the low temperature spin configuration has any kind of spatial long-range order which is intrinsically impossible because the underlying lattice does not have any translational long-range order in our model. Nevertheless, the low temperature state in these intermediate fields can be regarded as the ferrimagnetic state in a properly defined "curved" space. This effect is most clearly observed in the field cooled magnetization (the magnetization measured by cooling the sample under the constant magnetic field). When the field cooled magnetization well below the freezing temperature is plotted versus the magnetic field, the resulting magnetization curve has a clear plateau for certain intermediate fields. The magnetization value corresponding to this plateau is one third of its saturation value. When the applied field is small, the system shows qualitatively similar behavior with the zero field case which was studied in detail in (1). When the applied field is large enough, the system behaves like paramagnetic spins arranged in the field.

Our model and the method of the calculation are explained in §2. The calculated thermodynamical properties are presented in §3. In §4, the ground state spin configurations obtained in the present Monte Carlo simulations are classified and statistically analyzed. Section 5 is devoted to summary and discussions.

§ 2. Model and the method of the calculation

Our model is the same as the one used in (1) except that the finite magnetic field is applied in the present case. We consider the antiferromagnetic Ising model on the triangular-square lattice which is assumed to be quenched. The Hamiltonian is given by

$$\mathcal{H} = J \sum_{ij} \sigma_i \sigma_j - h \sum_i \sigma_i, \quad (J > 0)$$

where $\sigma_i = \pm 1$ and the sum is taken over all nearest neighbors. The configuration average is formally defined to be taken over all possible lattice configurations for a given value of the ratio between the numbers of triangles and squares. In actual calculations, we take the most random lattice configuration corresponding to the highest entropy state. In such a state, the number of squares to that of triangles is evaluated to be about 0.45 which corresponds to the average coordination number about equal to 5.1. The way of sample preparation was explained in detail in (1). In the present calculation, the system size $N$ is set equal to $24 \times 24$. For this purpose, the sample of the
size $26 \times 26$ is prepared and the outermost layer is used to impose the periodic boundary condition on the spins. The configuration average is actually taken over only a small number of samples, often only one sample. As was shown in (I), the triangular-square lattice can be represented by the bond-diluted triangular lattice with a certain restriction. The dilution rule is given in Fig. 5 in (I). As an example, the sample most frequently used in the present simulation is shown explicitly in Fig. 1 in such a form. The method of the Monte Carlo simulation is the same as the one used in (I). In each run, initial 1000 Monte Carlo steps per spin [MCS/ spin] are discarded and subsequent 10000 MCS/ spin are used to calculate the average.

§ 3. Results

The temperature dependence of the field cooled magnetization is shown in Fig. 2 for several values of the field. The data are obtained for the sample shown in Fig. 1. (The dimensionless temperature $\theta$ is defined by $k_B T / J$. ) It can be clearly seen that for the fields $2J \leq \theta \leq 4J$, the magnetization curves are eventually attracted to the one third of its saturation value at low enough temperatures. In fact, we have found that the low temperature spin configuration with these fields are essentially ferrimagnetic. In order to see this, we make use of the fact that the triangular-square lattice is topologically equivalent to the bond-diluted triangular lattice as is shown in Fig. 1. Via this mapping procedure, one can map the spatially random triangular-square lattice onto the regular triangular lattice in a deformed or a “curved” space. With this mapping, the ferrimagnetic magnetization per spin can be defined without ambiguity by the relation:

Fig. 2. Temperature dependence of the field cooled magnetization for several values of the magnetic field.

Fig. 3. Temperature dependence of the ferrimagnetic magnetization $M_F$ for several values of the magnetic field. For the definition of $M_F$, see the text.
where $M_I$, $M_B$, and $M_W$ denote the uniform magnetization per spin of the three sublattices in the corresponding triangular lattice, respectively. The value of $M_F$ is normalized so that it gives unity when the system is completely ferrimagnetic, i.e., when the spins in two sublattices are up and in one sublattice down. The temperature dependence of $M_F$ is shown in Fig. 3 for several representative values of the field. This quantity becomes completely (or almost) unity at lower temperatures when the field lies in the range $2J \leq h \leq 4J$. (Only in the case $h=2J$, the obtained ground state is imperfectly ferrimagnetic ($M_F \approx 0.92$) and a small size of misfitting domain appears in the system.) This result suggests that for these fields the large ground state degeneracy is drastically decreased and the unique long-range order prevails in the system at low temperatures. Moreover, this long-range order is the ferrimagnetic one when viewed in the mapped curved space (2-sublattice ferri in the triangular lattice), though it does not give any spatially long-range order in the original flat space. In the original space, it at most gives the random ordered phase (ROP). In order to check the possible sample dependence of this phenomenon, we have repeated the same Monte Carlo simulations for the case $h=3J$ by using a different sequence of random numbers and by using different samples (one more $24 \times 24$ lattice and five $18 \times 18$ lattices). Almost all results give completely ferrimagnetic ground state ($M_F=1$). Only one run gives nearly ferrimagnetic ground state ($M_F \approx 0.94$) where a small size of misfitting domain appears.

The field dependence of the ground state energy obtained in the present Monte Carlo runs is shown in Fig. 4. The straight line represents the energy of the perfectly ferrimagnetic state.

![Field dependence of the ground state energy obtained in the field cooled Monte Carlo runs.](image)

Fig. 4. Field dependence of the ground state energy obtained in the field cooled Monte Carlo runs. The straight line represents the energy of the perfectly ferrimagnetic state.

![Field dependence of the field cooled magnetization (FCM, •) and the zero field cooled magnetization (ZFCM, □) at the temperatures $T=0.2J$ (a), $0.7J$ (b) and $1.4J$ (c), respectively.](image)

Fig. 5. Field dependence of the field cooled magnetization (FCM, •) and the zero field cooled magnetization (ZFCM, □) at the temperatures $T=0.2J$ (a), $0.7J$ (b) and $1.4J$ (c), respectively.
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Carlo simulation is shown in Fig. 4. The straight line corresponding to the energy of the perfect "ferrimagnetic" state is also shown. In the cases \(2.5J \leq h \leq 4J\), the calculated energies lie just on this straight line reflecting the fact that the obtained ground state spin configurations are perfectly ferrimagnetic. Note that in the cases \(h=2.0J\) and \(4.5J\), the energies obtained in the Monte Carlo simulation lie slightly above the ferrimagnetic energy. This means that within the time scale of the present simulation the system cannot reach the true ground state which is most probably ferrimagnetic. In the cases \(h \leq 1.5J\) and \(h \geq 5J\), on the other hand, the obtained ground state energies lie below the ferrimagnetic energy.

The field dependence of the field cooled magnetization is shown in Fig. 5(a) at a temperature \(t=0.2\) well below the freezing temperature. (The freezing temperature in zero field is about equal to \(t=0.7\).) The magnetization curve has a plateau just at the one third of the saturation value when the field lies in the range \(2J \leq h \leq 4J\). (In the case \(h=2J\), this value is equal to 0.313 a little smaller than \(1/3\) and the obtained ground state spin configuration is not perfectly ferrimagnetic with some domain structures.) Our previous analysis of the ground state energy suggests, however, that the non-equilibrium effect exists in the boundary region in the present simulation and the plateau region extends from the field \(h=2.0J\) up to the field \(h=4.5J\) in complete thermal equilibrium. In Fig. 5, the corresponding results for the zero field cooled magnetization are also shown. In our present simulation, the zero field cooled magnetization is evaluated by the following procedure. First, the sample is gradually cooled without applying the magnetic field under the same condition with the field cooled calculation. Then the field of the prescribed strength is applied and the resulting magnetization is measured after waiting for a hundred MCS/ spin. In contrast to the field cooled magnetization, it shows almost linear behavior without showing the plateau phenomena.

Such remarkable difference between the behaviors of the field cooled and the zero field cooled magnetizations is probably originated from the fact that in the case of the field cooling the system can reach thermal equilibrium even in our Monte Carlo runs except for the boundary cases (e.g., \(h=2.0J\) and \(h=4.5J\)). In the case of the zero field cooling, on the other hand, the system is trapped in the metastable valley due to the kinetic effect. In order to check this viewpoint, we have carried out extremely long Monte Carlo runs for the zero field cooled samples. That is after switching the fields \((h=2J\) and \(h=4J\), respectively) on the zero field cooled sample, the system is evolved at a temperature \(t=0.2\) during 150000 MCS/ spin. Such calculations show as expected that both the uniform magnetization and the ferrimagnetic magnetization gradually approach the corresponding field cooled values (i.e., \(M=1/3\) and \(M_F=1.0\)). During the 150000 MCS/ spin runs, the magnetization changes its value from 0.196 to 0.240 and from 0.424 to 0.379 in the cases \(h=2J\) and \(h=4J\), respectively. The ferrimagnetic magnetization changes its value from 0.226 to 0.272 and from 0.189 to 0.510 in both the cases, respectively. In Figs. 5(b) and (c), we have also shown the field dependence of the field cooled and the zero field cooled magnetizations at higher temperatures. Two kinds of curves agree in a good accuracy, which indicates that the non-equilibrium effect associated with the spin freezing is negligible at these higher temperatures.

We have also investigated whether the system exhibits appreciable remanence effect. It is found that even if the system is initially in the completely polarized state, the magnetization drops very quickly to zero (or the fluctuation value around zero) when the
The specific heat per spin calculated from the energy fluctuation is shown in Fig. 6. The peak height becomes maximum at an intermediate field \((h=3J)\) indicating that the ordering tendency is enhanced around this field. The susceptibility per spin calculated from the magnetization fluctuation is shown in Fig. 7. When the field is in the range \(2J \leq h \leq 4J\), it has a cusp-like structure which is similar to the one found in the usual antiferromagnet. At higher fields, on the other hand, the susceptibility shows a paramagnetic behavior. These behaviors of the specific heat and the susceptibility seem consistent with our observation that in the field range \(2J \leq h \leq 4J\) the "ferrimagnetic" long-range order appears in the system.

Finally in Fig. 8, we have shown the field dependence of the Edwards-Anderson order parameter \(q\) for several temperatures. It is also clear from this figure that the ordering field is turned off. This is in sharp contrast with the spin glass case in which remarkable remanence effect is commonly observed.\(^5\)\(^,\)\(^6\) Such difference suggests that in amorphous antiferromagnets the ordering field is "orthogonal" to the uniform field in distinction with the spin glass case. On the other hand, if we observe the ferrimagnetic magnetization starting from the completely ferrimagnetic initial state, clear remanence effect is found. We note that a similar behavior is also reported by Grest and Gabl for the site-diluted antiferromagnet on the triangular and the fcc lattices.\(^7\)
is much enhanced in the field range \(2J \leq h \leq 4J\). Obviously, this phenomenon cannot be interpreted simply as the field induced ordering of paramagnetic spins because at the fields a little above \(h=4J\) the value of \(q\) is decreased by increasing the field. On the other hand, the saturation of \(q\) observed in the fields higher than \(h\approx 7J\) is caused due to this effect. This non-monotonic behavior is reflecting the meta-magnetic feature of this model, that is disorder (spin glass)→ferri→polarized para as the field increases as we see in Fig. 5(a).

§ 4. Analysis of the ground state spin configuration

In the present section, we shall statistically analyze the ground state spin configurations obtained in the present Monte Carlo runs. The triangular-square lattice consists of only two elementary units, i.e., the triangular unit and the square unit. For the triangular unit, there are four different spin configurations which are described as \((+++\)\) \((++-\)\) \((-+-\) and \((---\)\), respectively, as are shown in Fig. 9. Here we do not distinguish the spin configuration which can be interchanged with each other by mere rotation. The magnetic field is assumed to be applied in the upward direction. For the square unit, there are six different spin configurations, i.e., \((++++)\) \((++++-)\) \((++-+)\) \((++--+)\) \((+--++)\) and \((-----)\), as shown in Fig. 9. The ratio of the number of these particular spin configurations is examined for the ground states obtained in our field cooled runs. The results for several representative fields are summarized in

Table 1. The ratio of the number of each spin configuration which appeared in the field cooled Monte Carlo runs. For the meaning of the symbol, see the text and Fig. 9. The data are given for several representative values of the field in the case of triangles (a) and in the case of squares (b).

<table>
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<tr>
<td>(h=0)</td>
<td>48.3 %</td>
<td>52.5 %</td>
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<tr>
<td>(h=1.5J)</td>
<td>85.6 %</td>
<td>14.1 %</td>
<td>0.3 %</td>
<td>0 %</td>
</tr>
<tr>
<td>(h=3.0J)</td>
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<td>0 %</td>
<td>0 %</td>
<td>0 %</td>
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<tr>
<td>(h=4.5J)</td>
<td>95.5 %</td>
<td>0 %</td>
<td>4.5 %</td>
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<tr>
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<td>70.6 %</td>
<td>14.9 %</td>
<td>5.6 %</td>
<td>8.9 %</td>
<td>0 %</td>
<td>0 %</td>
</tr>
<tr>
<td>(h=1.5J)</td>
<td>53.2 %</td>
<td>4.0 %</td>
<td>42.3 %</td>
<td>0.4 %</td>
<td>0 %</td>
<td>0 %</td>
</tr>
<tr>
<td>(h=3.0J)</td>
<td>34.3 %</td>
<td>0 %</td>
<td>65.7 %</td>
<td>0 %</td>
<td>0 %</td>
<td>0 %</td>
</tr>
<tr>
<td>(h=4.5J)</td>
<td>21.8 %</td>
<td>0 %</td>
<td>6.9 %</td>
<td>0 %</td>
<td>9.3 %</td>
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Table I. Up to the field as large as $h = 4.0J$, completely polarized spin configurations, i.e., $(+++)$ triangles and $(++++)$ squares are relatively rare. (Especially, $(++++)$ squares are not found at all in this field range.) In the case of zero magnetic field, the energy associated with the triangular part of the sample approximately takes its possible minimum value which is equal to $-J$ per triangle. On the other hand, not all squares can take its minimum energy value $-4J$ per square because the existence of triangles necessarily causes "frustration" in the squares. The ground state spin configuration in zero field should be determined so as to maximize the number of $(+-+-)$ squares within such geometrical restriction. In the present simulation, about 70% of squares are in the $(+-+-)$ state. When the magnetic field is applied, the number of $(+-+-)$ squares is decreased and that of $(++-)$ squares is increased. The appearance of the perfect ferrimagnetic order means the disappearance of $(++-)$ squares and $(+-)$ triangles. (These states are incompatible with the ferrimagnetic long-range order.) In the perfect ferrimagnetic state, the ratio between the number of $(+-+-)$ squares and that of $(++-)$ squares is about 1:2 and all triangles are in the $(+++)$ state. Breakdown of the ferrimagnetic order at higher fields is characterized by the appearance of the fully polarized configurations such as $(++++)$ squares and $(+++)$ triangles.

§ 5. Discussion

We first summarize the transition behavior of the present model on the basis of the results obtained in the present paper and in (I). Qualitatively speaking, three distinct cases are possible depending on the strength of the applied field. In zero or small fields ($h \leq 2J$), the ground state spin configuration is highly degenerate and the system exhibits a spin freezing transition at a certain finite temperature which is most probably driven by the kinetic effect. In this case, the low temperature phase is essentially the "metastable spin glass" state, though the susceptibility does not show a clear cusp in contrast to the canonical spin glasses. Such different behavior of the susceptibility might be attributed to the fact that all interactions are antiferromagnetic in the present model and the uniform field is "orthogonal" to the ordering field. In the intermediate fields ($2J \leq h \leq 4.5J$), the ground state degeneracy is drastically suppressed and the system seems to have a certain long-range order at low temperatures. This long-range order is ferrimagnetic when viewed in the properly defined curved space. In the original flat space, however, it does not give any spatial long-range order and looks at best like the random ordered phase (ROP) reflecting the fact that the triangular-square lattice does not have any translational long-range order. This ordering property most clearly manifests itself in the plateau behavior of the field.
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dependence of the field cooled magnetization. In higher fields \( (h \geq 4.5J) \), the system behaves like paramagnetic spins arranged in the applied field and does not show any phase transition. All that we have discussed are summarized in the temperature-magnetic field phase diagram Fig. 10. (Note that the figure is only schematic.)

Now we add some discussions about related problems. The most intriguing question is that whether the phenomena observed in the present model calculation are the universal ones in general amorphous antiferromagnets or the accidental ones peculiar to the present model. In other words, how sensitive the results are on several details of the model and what are the essential elements of the observed phenomena? Especially it seems important to clarify to what extent the details of the atomic structure influence the transition behavior. One can check this point, for example, by studying various kinds of random structures such as the dense random packed array of hard particles or the quenched random structure obtained by molecular dynamics simulation. Moreover, it might be interesting to ask whether a similar behavior could be found in some three-dimensional systems. If one might dare to regard the three-dimensional dense random packed structure as the bond-diluted fcc lattice in some curved space in analogy with the present two-dimensional model, the possibility that the field cooled magnetization curve has a plateau at one half of the saturation value is suggested. (The transition behavior of the antiferromagnetic Ising model on the fcc lattice under the magnetic field was studied by Binder by using the Monte Carlo method.) We are planning to check these points by carrying out further Monte Carlo studies.

It also seems quite probable that the anisotropy (or isotropy) of the spin-spin interaction might give a large effect on the results when we take into account the fact that the antiferromagnets on the regular triangular lattice show quite different kinds of behavior in the case of the Ising, the plane rotator and the isotropic Heisenberg models. These points need further clarification.

Finally we discuss the experimental possibility to observe the predicted phenomena in real amorphous antiferromagnets. As was discussed in (1), the oxygen adsorbed on amorphous substrate can be regarded as nearly two-dimensional amorphous antiferromagnets. It is interesting to study whether the plateau phenomena are observed in the field cooled magnetization of this system. We must note, however, that the value of the field necessary to observe this plateau (if it exists) is relatively large. If we estimate the strength of the exchange interaction \( J \) about 100 K as in (1), our Monte Carlo results suggest that the necessary field is more than about 200 K, i.e., more than 200 KG. Note that the zero field cooled magnetization shows the conventional linear behavior in contrast to the field cooled magnetization.

To conclude, we have carried out some Monte Carlo studies for a simplified two-dimensional Ising model for amorphous antiferromagnets in order to investigate how the magnetic field influences the thermodynamical properties. It is found that application of a certain range of finite magnetic field changes the nature of the spin freezing transition qualitatively and leads to the uniquely determined random ordered phase at low temperatures. This state is the ferrimagnetic state in the properly defined curved space. Experimentally, such effect could be most directly observed in the plateau phenomena of the field cooled magnetization.
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References

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