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Hysteretic behavior of the site-diluted Ising antiferromagnet in uniform applied fields

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We report a Monte Carlo study of the hysteretic behavior of the site-diluted Ising antiferromagnet on a simple-cubic lattice. States found by reducing the magnetic field from an initial value in the paramagnetic phase show no long-range antiferromagnetic order, whereas states found by increasing the magnetic field from an initially ordered state do show long-range order until the field is increased to a critical value. These "increasing-field" states are presumably nearly representative of true thermodynamic equilibrium. This hysteretic behavior is due to metastable interfaces which are formed when the field is decreased from the paramagnetic phase. To see the physical origin of the formation of the metastable interfaces, we do computer experiments for the dynamics of initially flat interfaces with and without uniform applied fields. From this result, we show that metastable interfaces are formed, because even a small uniform applied field roughens the interface.

I. INTRODUCTION

Ever since the analysis of Imry and Ma\cite{1} based on a domain argument which predicted that the lower critical dimension \( d_c \) of the ferromagnetic Ising model in the random fields to be 2, there has been much interest\cite{2-9} in this subject. In particular, the arguments based on field-theoretical perturbation expansions\cite{2-4} and supersymmetry arguments\cite{5,6} seemed to show that \( d_c = 3 \). Also, Pytte et al.\cite{7} have argued that a roughening of the domain walls adds to the disorder of the system, so that \( d_c \) ought to be 3. Thus, for several years there have been contradicting theoretical conclusions as to the value of \( d_c \) for the random-field Ising model (RFIM). More recently, further analysis\cite{8,9} of the roughening transition based on renormalization-group calculations has predicted that, even though interface roughening may occur for \( d < 5 \), the lower critical dimension of the RFIM is still 2.

Although the experimental test of this dispute seems to be unrealizable, Fishman and Aharony\cite{10} have argued that a bond-diluted antiferromagnetic Ising model in a uniform applied field behaves like a RFIM. Also, Wong, von Molnar, and Dimon\cite{11} have pointed out that there is another mechanism to generate random fields besides that of Fishman and Aharony when a uniform field is applied to the site-diluted Ising antiferromagnet (SIAF). Thus, several groups\cite{12-15} have done experiments to determine \( d_c \) of the RFIM, using the correspondence of a SIAF in uniform applied fields to a RFIM, but their conclusions are also contradictory to one another, as were the theoretical studies.

These contradictory experimental conclusions have recently been at least partly explained by the work of Wong and Cable\cite{15} who have shown the existence of hysteretic behavior for a SIAF in uniform applied fields. It is this hysteresis which causes different experimental approaches to a point in the ordered phase characterized by uniform applied field \( h \) and temperature \( T \) to differ, and, as a result, greatly complicates the experimental determination as to whether or not an ordered phase exists. To be specific, the neutron-scattering studies of the dilute antiferromagnet Fe\(_x\)Mg\(_{1-x}\)Cl\(_2\) have shown that the scattering intensities for momentum transfer along the antiferromagnetic axis in decreasing uniform fields are quite different from those in the increasing fields. The data have also shown that the hysteretic behavior becomes stronger as the temperature is decreased. Furthermore, above some temperature, there is no hysteretic behavior. Similar hysteretic behavior has been found by King et al.\cite{16} in the very-low-temperature measurement of magnetic susceptibility of the dilute antiferromagnet and zero-temperature Monte Carlo (MC) simulation studies. However, all the systems studied by the experiments\cite{11-15} have complex crystal symmetry\cite{17} which might affect the results of experiments. Recently, there have been several theoretical attempts\cite{18-20} to explain these experimental hysteretic behaviors in the context of interface roughening of the RFIM. These attempts have all been based on similar physical arguments and have obtained results which are similar qualitatively but different in detail from one another. In any event, it is clear that to interpret the experimental studies of \( d_c \), we must understand the hysteretic behavior of the SIAF in applied fields. Accordingly, in this paper we have performed MC studies of the SIAF on a simple-cubic lattice under various conditions of uniform applied fields and initial preparation of the samples in order to understand this hysteretic behavior.

II. MONTE CARLO STUDY OF Hysteretic BEHAVIOR

In this section we explain the methods of MC simulation we used to study the hysteretic behavior of the SIAF and present the results so obtained.

In our simulation we take processes in which applied field is varied at a certain temperature. We did not attempt "field cooling" or "field heating" in which the temperature is varied at constant applied field. Such conditions would correspond to paths labeled 3 and 4 in the...
FIG. 1. Equilibrium phase diagram for the site-diluted antiferromagnet in an applied field \( h \) at temperature \( T \). Here, AF and PA denote the antiferromagnetic and paramagnetic phases, respectively. We show the following processes: (1) constant \( T \), decreasing \( h \); (2) constant \( T \), increasing \( h \); (3) decreasing \( T \), constant \( h \); (4) increasing \( T \), constant \( h \).

qualitative equilibrium phase diagram of the SIAF in an applied field shown in Fig. 1. For equilibrium critical phenomena it is well established that the angle at which the phase boundary is crossed has only a rather trivial affect on the characteristics of the observed phase transition. In the present case we expect paths 1 and 3 starting in the paramagnetic phase to be equivalent to each other, and, similarly, paths 2 and 4 starting in the ordered phase to be equivalent to each other.

We treated the model on a simple-cubic lattice of size \( 20 \times 20 \times 20 \). The Hamiltonian for the site-diluted Ising antiferromagnet is

\[
H = J \sum_{\langle i,j \rangle} \sigma_i \sigma_j - h \sum_i \sigma_i \epsilon_i ,
\]

where \( i \) and \( j \) are site indices, and, for each site \( i \), \( \epsilon_i \) is an independent random variable assuming the value 0 for vacant sites with probability \( 1 - p \) and 1 for occupied sites with probability \( p \). The exchange interaction between spins is assumed to be only between nearest neighbors, and \( \sigma_i \) is the Ising spin on site \( i \) and is either 1 or \(-1\). In Eq. (1) the notation \( \langle i,j \rangle \) indicates that each pair \( i,j \) is counted once in the summation. The interaction constant \( J \) is positive and \( h \) is the uniform applied field. Quenched averages over the distribution of \( \epsilon_i \) will be denoted by \( \langle \cdot \rangle \), so that, for instance, \( \langle \epsilon_i \rangle = p \). Throughout this paper we use periodic boundary conditions.21

In the MC simulation, first we make randomly chosen sites vacant one by one from the completely occupied lattice until the given occupation probability \( p = \langle \epsilon_i \rangle \) is fulfilled. In this process, \( p \) is prepared to be sufficiently far above the critical concentration, \( p_c = 0.31 \), to avoid the complex behavior at the percolation threshold. Next, to discard the effect of isolated clusters, the cluster structure of the prepared sample is analyzed, and only the sites on the largest cluster (which we call the “infinite” cluster) are retained. Sites in the smaller isolated clusters are made vacant. Because the concentration \( p \) in our simulation is far above \( p_c \), the identification of the largest cluster is always unambiguous and the number of sites, \( n_\infty \), in the “infinite” cluster differs by only a fraction of a percent from the total number of occupied sites including all clusters. Also, the temperatures are taken to be sufficiently far below the antiferromagnetic phase-transition temperature so as not to “see” the large thermal fluctuations which are difficult to handle in the finite-sized lattice we treated. We have used Glauber dynamics,\(^ {21-23} \) in which a randomly chosen spin is flipped only if the transition probability \( w \) satisfies

\[
w = \tau^{-1} \exp (-\Delta E / k_B T) \geq r ,
\]

where \( \tau \) sets the timescale and is independent of temperature if the temperature is not too low,\(^ {21} \) \( \Delta E \) is the energy difference between the case when the spin is flipped and that when the spin is not flipped, and \( r \) is a random number \((0 \leq r \leq 1)\). One Monte Carlo “step” refers to examining each occupied site on the average once.

For the prepared sample at a given temperature, we did two kinds of Monte Carlo study under the above dynamics. One is the low-field approach (LFA, process 2 in Fig. 1), in which the data are taken for the increasing uniform applied fields from the sample which is initially prepared in the completely antiferromagnetic ground state, i.e., \( \sigma_i \)'s of the occupied sites in one sublattice (\( A \) sublattice) are 1 and those in the other sublattice (\( B \) sublattice) are \(-1\). The other is the high-field approach (HFA, process 1 in Fig. 1), in which data are taken for the decreasing fields from the sample which is initially prepared in the completely ferromagnetic ground state, i.e., \( \sigma_i \)'s of all occupied sites are 1. Experiments\(^ {1-15} \) in uniform applied fields have been performed under various conditions of initial preparation and at temperatures above and below the phase-transition temperature. Here we limit our attention to a systematic comparison of the HFA and LFA processes outside the critical region.

For each data point, we have discarded the initial 1500 MC steps and obtained \( \langle \sigma_i \rangle \), which is defined to be the average of the magnetization of the \( i \)th site over the final 500 MC steps. Then the staggered magnetization \( S \) and the magnetization \( M \) are defined as

\[
S = \sum_{i \in A} \epsilon_i \langle \sigma_i \rangle - \sum_{j \in B} \epsilon_j \langle \sigma_j \rangle \equiv n_m s ,
\]

\[
M = \sum_i \epsilon_i \langle \sigma_i \rangle \equiv n_\infty m ,
\]

so that \( s \) is the staggered magnetization and \( m \) the magnetization, both per occupied site of the “infinite” cluster. Also, \( i \in A, (j \in B) \) indicates a sum in which \( i (j) \) ranges over sites in the \( A (B) \) sublattice. In Fig. 2 we present the data of the HFA and LFA for three differently prepared samples with the same concentration probability, \( p = 0.52 \), and at the temperature \( T = 0.25 T_c (1) \), where \( T_c (1) = 4.5 J \) is the phase-transition temperature of the nonrandom sample. For the LFA there is nearly no dispersion of \( s \) in three differently prepared samples, except for the critical thermal fluctuations near the critical field, which represents a typical phenomenon in a finite-sized lattice. The field dependence of \( s \) for the LFA is very much like a typical order-parameter diagram for a second-order phase transition. Since the uniform applied field is not the con-
jugate field of the order parameter $s$, if there is an antiferromagnetic phase transition at some field, the order-parameter-versus-field curve should resemble that of the LFA in Fig. 2. As we discuss in Sec. IV, we believe that the LFA states are essentially (but not exactly) the equilibrium states for the SIAF in uniform applied fields in three dimensions. In contrast, for the HFA the $s$'s of the three samples are quite different from one another and the magnitudes are much smaller than those of the LFA in small fields. This is a rather striking fact. There is a hysteretic behavior which depends on the preparation of the initial states. To test that this hysteresis is not an artifact of our numerical procedures, we show, in Fig. 3,
similar results for the staggered magnetization of a pure system. The results for the magnetizations \( m \) for the LFA and HFA is presented in Fig. 4. The \( m \)'s for the LFA in small fields are slightly different from those of the HFA. This fact explains that the states of the HFA in small fields are very stable and have nearly the same free energy as those of the LFA. For each data point we have recorded the final snapshots of configurations of Ising spins in the planes \( z = 9,10,11 \) and of the averaged configurations for the final 100 MC steps of the same planes. (See Fig. 5.) Here the averaged configurations are meant to be the configurations in which the Ising spin of each occupied site is summed over the final 100 MC steps and is replaced by \(+1\) or \(-1\) according to the sign of the sum. The final snapshots are nearly the same as the averaged configurations for all data points for both the HFA and LFA. The LFA configurations consist of a single antiferromagnetic domain whose phase is the same as the initial state. In contrast, the configurations of the HFA are found to consist of two domains of nearly equal size. Also, the location of the domain wall hardly changes as a function of field for \( h/k_B T \leq 0.6 \). This means that the domain wall (or interface) is locally very stable in small fields. An important fact concerning the domain-wall formation is that the domain walls (interfaces) are formed preferentially at vacant bonds and cut only small numbers of occupied bonds.\(^{23}\)

III. MONTE CARLO STUDY OF INTERFACIAL DYNAMICS

In order to understand the dynamical origin for the metastable interfaces (domain walls), we have done another computer experiment which we discuss in this section. To see the results more clearly, instead of studying the site-diluted Ising antiferromagnet in uniform fields, we study the site-diluted Ising ferromagnet in the uniform staggered fields, which is mathematically equivalent to the SIAF in uniform fields. This Hamiltonian is

\[
H = -J \sum_{(i,j)} \epsilon_j \sigma_i \sigma_j - h \left[ \sum_{i \in A} \epsilon_i \sigma_i - \sum_{j \in B} \epsilon_j \sigma_j \right],
\]

where all the indices are the same as those in Eq. (1), and the sublattice notation is as in Eq. (3). Thus the magnetization of this section corresponds to the staggered magnetization of the preceding section and vice versa. Initially, we gave two domains to the sample C in Fig. 2 with the flat interfaces as follows. The Ising spins of the occupied sites in eight planes, \( p = 7 - 14 \), are set to be \(-1\), and those in the other planes are set to be \(1\). Thus initially the cross section of the sample at the \( z = 10 \) plane is that shown in Fig. 6(a). From this initial preparation, and at temperature \( T = 0.25T_c (1) \), we have simulated the time evolution of magnetization \( M \) [see Eq. (3b)] under two conditions. One is that there is no applied staggered field, i.e., \( h = 0 \), and the other is that there is small applied staggered field \( h/k_B T = 0.2 \). We have done each experiment for 6000 MC steps (for each occupied site). The magnetization so obtained is shown in Fig. 7, and the final drawings of the \( z = 10 \) plane with no field and with small staggered applied field are presented in Figs. 6(b) and 6(c). When there is no field, the sandwiched domain (i.e., "down" domain) begin to collapse after some initial fluctuations, and thus the magnetization \( M \) increases. (We remind the reader that the magnetization here corresponds to the staggered magnetization in, say, Fig. 2.) Eventually, the divided "up" domains meet each other and thus the sample will be all "up" phase after some
shown in Fig. 8. When a small magnetic field is applied parallel to the down spins, the spins of this dense region must reverse their directions to remain in thermodynamic equilibrium if the condition

$$nh > 2J$$

is fulfilled. If $h$ is small, i.e., if $h \ll J$, then $n$ must be very large to satisfy condition (5). For large $n$ the probability of finding such a configuration is of the same order as the probability that an isolated cluster of $n$ sites occurs. Above the percolation threshold ($p > p_c$), the probability $P(n)$ that an isolated cluster of $n$ sites occur is \(26, 27\)

$$P(n) \sim \exp(-Cn^{1-d^{-1}}),$$

where $d$ is the spatial dimensionality. We can estimate the difference between the thermodynamic value of the staggered magnetization, $\langle s \rangle$, and its value in the LFA state, as

$$\langle s \rangle - \langle s \rangle_{\text{LFA}} \sim \exp(-Ch^{-1+d^{-1}}).$$

Although this is a small effect, it is important to help us keep in mind that even the LFA state is not the true thermodynamic equilibrium state.

Next we consider the physical origin of the states of the HFA and we now neglect the fact that the LFA states are not, strictly speaking, the true equilibrium states. The HFA states are metastable states and thus, dynamically, must have long relaxation time. Now we want to suggest

**IV. DYNAMICAL MODEL FOR HYSTERETIC BEHAVIOR**

In this section the physical interpretations of the data and the dynamical model for the metastable interface formation in SIAF in the small applied fields are given.

The first question we address is whether any of the states obtained in simulations such as these are to be interpreted as the absolute equilibrium states. First, it is clear that the states obtained at small fields via the HFA are not the equilibrium states, because the domains which were nucleated at high field cannot be stable in the zero-field limit. It remains to ascertain whether the LFA field states are the equilibrium states for small fields. That this is not strictly the case can be argued as follows. Consider a sample prepared at zero field to which an arbitrarily small uniform field is applied. As prepared, this sample clearly consists of a single domain. On the other hand, even in an arbitrarily small applied field, one can infer the existence of domains in true thermodynamic equilibrium by an argument similar to that given by Lischitz\(^24\) to discuss states near the exact band edge for an electron in a random potential. In dilute samples, there will always occur, albeit infrequently, any imaginable configuration of occupied and unoccupied bonds. In particular, there will occur a densely occupied region consisting of $N_1$ up spins and $N_2$ down spins, with $n = N_1 - N_2 > 0$ connected to the infinite cluster by a single bond, or by a few bonds, as
a dynamical model for these metastable states from our MC-simulation study of the interfacial dynamics. In the LFA the initial state is one of the two degenerate antiferromagnetic ground states. When the applied field is increased, some spins which initially were antiparallel to the field are flipped into the field direction, and as a result, the staggered magnetization \( S \) decreases. In fields less than the critical value, the phase throughout sample must still be the same phase with the initially given state, even though there are some thermal fluctuations. In the HFA, however, at very large applied field, all spins are in the paramagnetic phases and aligned along the field. When the fields decrease, antiferromagnetic nucleation starts. Owing to local fluctuation of the concentration, there must be locally dense regions, i.e., occupied sites with more than the average number of occupied neighbors. Such regions of high density are the initial nucleation centers for formation of the antiferromagnetic phase. However, since there are two antiferromagnetic phases, the phases at different nucleation centers need not be in registry. When the field is decreased further from this stage, the two phases will grow until all the occupied spins are in one or the other of the antiferromagnetic phases, if the temperature is sufficiently low and the field sufficiently small. Then there should be at least two domains with associated interfaces. Thus the important question is whether this kind of domain formation with interfaces is stable or not. If it is stable, is it metastable or equilibrium? To answer these questions quantitatively may be an arduous task. Here we suggest only the qualitative argument using methods similar to those of Grinstein and Ma, Villain, and Aeppli.

Because the interface at sufficiently low temperature in small fields hardly moves and is instead roughened, we can pretend that there is only one interface. Then the free energy \( F \) of this interface in \( d \) dimensions can be written as

\[
F = \int dz J(r) [1 + (\nabla f)^2]^{1/2} + h \int dz \sigma_0(r),
\]

where \( z = f(x) \) is the interface profile, such as in Fig. 9(a), and \( r = (x, z) \) is a \((d - 1, 1)\)-dimensional vector. We assume that the interface thickness is negligible because we are at a sufficiently low temperature, and use the continuum hypothesis. For simplicity we assume that the model is the site-diluted ferromagnetic Ising model in a small uniform staggered field, as in Eq. (4), and \( \sigma_0 \) is the local average of staggered magnetization. Also, \( J(r) \) is the local surface tension and, at low temperature, is proportional to the local-exchange coefficient. The term proportional to \( h \) in Eq. (8) is absent in the nonrandom model, except for a contribution confined to the interfacial region. This effect will be neglected because the fields \( h \) are much smaller than \( J_0 = J(r) \), where \([ ]\) means the average over occupation \( \epsilon_i \) as before. Now consider a part of an interface like Fig. 9(b), the free energy of which is estimated as follows. The contribution of the first term in Eq. (8) can be approximated by

\[
\int dz \frac{d-3}{2} J(r) \left[ 1 + \frac{(\nabla f)^2}{2} \right]
\]

within the harmonic approximation, which will be justified later. Then the exchange-energy cost by the roughening is roughly of order \( J_0 d^{-1} + J_0 w^2 L^{d-3} \) because \( |\nabla f| \approx w/L \). The fluctuation of \( \epsilon_A - \epsilon_B \) per site is estimated by assuming \( \epsilon_i \) and \( \epsilon_j \) to be uncorrelated. Then we have

\[
[\epsilon_A - \epsilon_B]^2 = 2\langle [\epsilon_A]^2 \rangle - \langle [\epsilon_A]^2 \rangle
\]

\[
= \langle [\epsilon_i]^2 \rangle = \langle [\epsilon_j]^2 \rangle \approx p (1 - p) = \delta
\]

where \( \epsilon_A \) is 0 when a lattice site in the \( A \) sublattice is vacant and 1 when occupied, and \( \epsilon_B \) is 0 when a site in the \( B \) sublattice is vacant and 1 when occupied. Thus, the field energy gain is of order \( h (w L^{d-1} \delta)^{1/2} \). Thus the excess free energy associated with the existence of the wall, \( E_w \), is the order of

\[
E_w \approx J_0 L^{d-1} + J_0 w^2 L^{d-3} - h (w L^{d-1} \delta)^{1/2}
\]

Minimization of this with respect to \( w \) yields

\[
w \approx h^{3/2} \delta^{1/2} J_0^{1/2} L^{(5-d)/3}
\]

\[
|\nabla f| \approx w/L \approx h^{3/2} \delta^{1/2} J_0^{1/2} L^{(5-d)/3}
\]

We thereby find that the energy gain via the interface roughening, compared to the energy when the interface is flat, \( \delta E_w \), is

\[
\delta E_w \approx h^{4/3} \delta^{2/3} J_0^{-1/3} L^{(d+1)/3}
\]

If \( d > 2 \), \( |\nabla f| \) is negligible when \( L \to \infty \), and thus the harmonic approximation is justified. Thus the domain roughening is negligible compared to the total interface size. Consequently, an argument similar to that of Imry and Ma should be applicable to this case, and the lower critical dimension is two. In three dimensions there must

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**FIG. 9.** (a) Schematic diagram for the analysis of the smooth interface in \( d \) dimensions. (b) Part of a roughened interface.
be long-range order, and thereby we conclude that the LFA states are essentially those of equilibrium. As one can see from Eq. (13), however, the interface roughening is always metastable, and thus, if \( d < 5 \), the interface can be pinned by roughening. This suggests why there is a hysteretic behavior of the SIAF in applied uniform fields.

V. FINAL REMARKS

To see interfacial roughening in the finite-sized samples, there must be a sufficient number of vacancies. The reason for this is the following. If \( L \rightarrow \infty \), the field energy gain \( h (\omega L^{d-8})^{1/2} \) in Eq. (11) always exceeds the exchange-energy cost \( J_0 \omega L^{d-3} \) if \( d < 5 \). When \( L \) is limited by the sample size to be finite, however, then, unless \( \delta \approx \rho (1 - p) \) is sufficiently large, we cannot see the interface roughening because field energy gain cannot overcome the exchange-energy cost.

When the temperature is not sufficiently low, the analysis of the preceding section may not be applicable in two senses. Near the critical temperature, not only is the domain size large but also the interface is not very sharp. The situation as depicted by Eq. (8) or Fig. 9(a) may not exist. Also, local order parameter \( m \) or \( s \) should not be approximated by unity. In this case we need to invoke an analysis like Villain's.

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