Random-field critical scaling in a model of dipolar glass and relaxor ferroelectric behavior

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Heat-bath Monte Carlo simulations have been used to study a 12-state discretized Heisenberg model with a type of random field on simple cubic lattices of size $128 \times 128 \times 128$. The 12 states correspond to the [110] directions of a cube. The model has the standard nonrandom two-spin exchange term with coupling energy J and a random field which consists of adding an energy h_R to two of the 12 spin states, chosen randomly and independently at each site. We report on the case $h_R/J = 3$, which has a sharp phase transition at about $T_c/J = 1.406\,25$. Below T_c, the model has long-range ferroelectric order oriented along one of the eight [111] directions. At T_c, the behavior of the peak in the structure factor, $S(\mathbf{k})$, at small $|\mathbf{k}|$ is a straight line on a log-log plot, which gives the result $\bar{\eta} = 1.214 \pm 0.014$. The onset of orientational order below T_c is very rapid for this value of h_R. There are peaks in the specific heat and longitudinal susceptibility at T_c. Below T_c there is a strong correction to ordinary scaling, which is probably caused by the cubic anisotropy, which is a dangerous irrelevant variable.

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I. INTRODUCTION

The O_{12} model [1,2] is a model of discretized Heisenberg spins, i.e., O_{12} is a discrete subgroup of O(3). We will put this model on a simple cubic lattice of size $L \times L \times L$, with periodic boundary conditions. It is now almost 25 years since the original Monte Carlo study [1] of the three-dimensional (3D) random-field O_{12} model. The numerical results presented there, which used $L \leqslant 64$, are crude by current standards, but the reasons given for why this model is worthy of study are just as valid now as they were then. This model is designed to represent a ferroelectric dipolar glass [3,4]. It is also believed to be relevant [5] for ferroelectric phases in strongly disordered cubic perovskite alloys, which are often referred to as relaxor ferroelectrics [6–11]. From this point of view, the essential element for relaxor ferroelectric behavior is the alloy disorder, rather than a particular chemistry. A cubic perovskite which has a supercell ordering of a few perovskite unit cells in size should not be considered a relaxor ferroelectric, regardless of its chemical composition.

The work reported here uses modern computing power and a somewhat improved Monte Carlo algorithm to obtain results for lattices of size $L = 128$, which was not possible in the earlier work. We also make a more sophisticated choice of the random-field distribution to obtain new results which should be of great interest to many people. We feel that the new results presented here demonstrate that the hope of Halperin and Varma [5], that this type of model can provide new insights into the nature of perovskite ferroelectric materials, is finally being realized.

Our Hamiltonian starts with the classical Heisenberg exchange form for a ferromagnet:

$$H_{\rm ex} = -J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j. \tag{1}$$

Each spin $\mathbf{S}_i$ is a three-component dynamical variable which has 12 allowed states, which are unit vectors pointing in the [110] directions. The $\langle ij \rangle$ indicates a sum over nearest neighbors of the simple cubic lattice. The symbol H denotes the uniform external field. It does not appear explicitly in our Hamiltonian, because we have set H to zero. There is no loss of generality in setting $J = 1$. We will also set Boltzmann's constant to 1. This merely establishes the units of energy and temperature. The restriction of the spin variables to O_{12} builds a temperature-dependent cubic anisotropy into the model. A useful review of the effects of such a cubic anisotropy has been given by Pfeuty and Toulouse [12].

We add to $H_{\rm ex}$ a random field which, at each site, is the sum of two Potts field terms:

$$H_{\rm RP2} = h_R \sum_i \left(\delta_{\mathbf{S}_i, \hat{\mathbf{u}}_i} + \delta_{\mathbf{S}_i, \hat{\mathbf{v}}_i} \right). \tag{2}$$

Each $\hat{\mathbf{u}}_i$ and $\hat{\mathbf{v}}_i$ is an independent quenched random variable. which assumes one of the 12 [110] allowed states with equal probability. Since $\hat{\mathbf{u}}_i$ is allowed to be equal to $\hat{\mathbf{v}}_i$, the random field at each site has 78 possible types. The reader should note that, for an XY model, adding a second Potts field would generate nothing new, due to the Abelian nature of the O(2) group. However, in the current case, adding the second Potts field can substantially reduce the corrections to finite size scaling.

The celebrated Imry-Ma construction [13] argues that random fields built from groups with continuous symmetries cannot have conventional long-range order when the number of spatial dimensions is less than or equal to four. The Imry-Ma argument implicitly assumes that the spin group is Abelian. What is assumed is that the coupling of the random field to the spins is purely of vector form. However, at a renormalization group fixed point, a non-Abelian

random vector field has the ability to generate higher-order tensor couplings to the effective spins. Whether or not such higher-order couplings are relevant at the critical fixed point must be studied explicitly, on a case-by-case basis.

The results we find here demonstrate that the Imry-Ma instability does not necessarily exist for random fields applied to a Heisenberg model. For a non-Abelian spin group the Halperin-Saslow effect [14] changes the long-wavelength dispersion from quadratic to linear. This can cure the Imry-Ma instability. This mechanism is also closely related to the ordering in the 3D Heisenberg spin glass [15]. From the hydrodynamic viewpoint, a spin glass and a random-field model for the same type of spins should have the same lower critical dimension. However, the Imry-Ma argument can only be applied to systems which do not have a Kramers degeneracy. The problem with the Imry-Ma argument for the Heisenberg model is that for non-Abelian spins the sum of two noncollinear vectors does not transform like a vector.

The reader may be tempted to object that the analysis of Halperin and Saslow should not be applicable in the presence of the cubic anisotropy. However, what we are really interested in here is the issue of the stability of the random-field Heisenberg critical point. It is believed that in 3D the pure Heisenberg critical point is stable against the presence of cubic anisotropy favoring the [111] directions [12]. For the O_{12} model, this was verified numerically by the author [1]. Here we will demonstrate numerically that the necessary condition for the 3D random-field Heisenberg critical point to be stable against [111] cubic anisotropy is also satisfied. This type of cubic anisotropy is often referred to as a "dangerous irrelevant variable."

In the work presented here, we will set $h_R = 3$. Study of the model for other values of h_R is under way. Note that having h_R positive means that $\mathbf{S}_i$ has an increased energy if it points in the direction of $\hat{\mathbf{u}}_i$ or $\hat{\mathbf{v}}_i$. As shown by density functional calculations [16], and studied previously in the author's work on a four-state model [17], a positive h_R is an appropriate choice for a model of a relaxor ferroelectric. The author believes that a negative value of h_R should be used for modeling a dipolar glass.

It is likely, however, that critical exponents will not depend on whether h_R is positive or negative, or even whether the values of h_R are equal for $\hat{\mathbf{u}}_i$ and $\hat{\mathbf{v}}_i$. Of course, we do not know yet whether this is true. In any case, the size of corrections to scaling are not universal. The author expects that corrections to scaling will be smallest when h_R is the same for both $\hat{\mathbf{u}}_i$ and $\hat{\mathbf{v}}_i$, because that is when the Halperin-Saslow effect is the strongest. The reader should note that, with this type of a random field, the model does not become trivial in the limit $h_R \to \infty$.

The range of distributions of the random fields for which this effect will be found remains unclear. Note that the pure Heisenberg critical point is not stable against the introduction of cubic anisotropy which favors the [100] directions. In that case, the phase transition becomes first order [12]. However, a first-order phase transition with a latent heat is not allowed in a model with quenched disorder [18,19].

II. NUMERICAL PROCEDURES

If h_R is chosen to be an integer, then the energy of any state of the model is an integer. Then it becomes straightforward to design a heat-bath Monte Carlo computer algorithm to study it which uses integer arithmetic for the energies, and a look-up table to calculate Boltzmann factors. This procedure substantially improves the performance of the computer program, and was used for all the calculations reported here. (The program currently has the ability to use half-integer values of h_R.) Lattices with periodic boundary conditions were used throughout.

Three different linear congruential random number generators are used. The first chooses the $\hat{\mathbf{u}}_i$, the second chooses the $\hat{\mathbf{v}}_i$, and the third is used in the Monte Carlo routine which flips the spins. The generator used for the spin flips, which needs very long strings of random numbers, was the Intel library program *random_number*. In principle, Intel *random_number* can be used for multicore parallel processing. However, our program is so efficient in single-core mode that no speedup was seen when the program was run in parallel mode.

The code was checked by correctly reproducing the known results [1] of the $h_R = 0$ case, and extending them to $L = 128$. For $h_R = 3$, 32 samples of size $L = 128$ were studied. The same set of 32 samples was used for all values of T, so it is meaningful to talk about heating and cooling of a sample. Each sample was initially in a random state, and then cooled slowly, starting from $T = 1.5625$, and ending at $T = 1.3125$. Both cooling and heating were done in temperature steps of 0.015 625, with times of 20 480 Monte Carlo steps per spin (MCS) at each stage. (The reader should note that the temperatures we use in the computer code are simple binary fractions, even though they may appear to be unnatural when written in decimal notation.)

For the pure O_{12} system, the Heisenberg critical temperature [1] is $T_c = 1.453$. For $h_R = 3$, the spin correlations are only short-ranged above this temperature, so no extended data runs were made in this region of T. For 30 of the 32 samples studied, the system was strongly oriented along one of the [111] directions by the time it had been cooled to $T = 1.3125$. The other two samples had become trapped in metastable states. These two samples were then initialized in [110] states which had a positive overlap of approximately 0.5 with their metastable states. They were run at $T = 1.25$, where they were easily able to relax to low-energy [111] states. These states were then warmed slowly to $T = 1.4375$. At $T = 1.343\,75$, these [111] states were seen to have significantly lower energies than the metastable states found by cooling for those two samples.

The fact that it was not difficult to find a stable state oriented along [111] at $T = 1.25$ means that, for $h_R = 3$, an $L = 128$ sample does not have many metastable states. However, it is expected that for large enough values of L it should be true that in the [111] ferroelectric phase there would be a metastable state corresponding to each of the [111] directions. Whether or not this will continue to be true for larger values of h_R is not yet known. In the proposed power-law correlated phase [1], it would no longer be possible to assign low-energy metastable states to particular [111] directions.

TABLE I. Thermodynamic data for $128 \times 128 \times 128$ lattices at $h_R = 3$, for various T. (h) and (c) signify data obtained relaxing from hotter and colder initial conditions, respectively. The one σ statistical errors shown are due to the sample-to-sample variations.

T	$\lvert\mathbf{M}\rvert$	$\chi_\parallel$	E	c_H	COO
1.34375 (c)	0.4270 ± 0.0024	25.9 ± 3.8	-1.1955 ± 0.0001	2.782 ± 0.008	0.985 ± 0.006
1.375 (c)	0.3108 ± 0.0041	86.0 ± 7.8	-1.1047 ± 0.0002	2.997 ± 0.012	0.869 ± 0.029
1.390625 (c)	0.2309 ± 0.0051	146 ± 26	-1.0569 ± 0.0002	3.103 ± 0.021	0.726 ± 0.054
1.40625 (c)	0.1253 ± 0.0055	291 ± 29	-1.0072 ± 0.0002	3.185 ± 0.018	0.616 ± 0.041
1.4375 (h)	0.0238 ± 0.0012	72.4 ± 1.7	-0.9173 ± 0.0001	2.472 ± 0.009	0.599 ± 0.012

Two trial runs made on $L = 64$ lattices indicated the presence of a [111] to [110] phase transition at roughly $T = 0.875$ for $h_R = 3$. However, a detailed study of this second transition was not undertaken.

Extended runs for data collection using the hot initial condition were made at $T = 1.4375$, 1.406 25, 1.375, and 1.343 75. For the cold initial condition, data collection runs were made at $T = 1.343\ 75$, 1.375, 1.390 625 and 1.406 25. As we shall see, relaxation of the hot and cold initial conditions gave essentially indistinguishable results at $T = 1.406\ 25$.

A data collection run for each sample consisted of a relaxation stage and a data stage. For the three lower values of T, a relaxation stage was a run of length 122 880 MCS. In most cases this was sufficient to bring the sample to an apparent local minimum in the phase space. This was followed by a data collection stage of the same length. If further relaxation was observed during the data collection stage, it was reclassified as a relaxation stage. Then a new data collection stage was run. The energy and magnetization of each sample were recorded every 20 MCS. A spin state of the sample was saved after each 20 480 MCS. Thus there were six spin states saved from the data collection stage for each run. These six spin states were Fourier transformed and averaged to obtain the structure factor for each sample. At $T = 1.4375$, a relaxation stage of only 20 480 MCS was judged to be sufficient.

III. THERMODYNAMIC FUNCTIONS

The thermodynamic data calculated from our Monte Carlo data on the 32 $L = 128$ samples are shown in Table I.

For a random-field model, unlike a random-bond model, the average value of the local spin, $\langle \mathbf{S}_i \rangle$, is not zero even in the high-temperature phase. The angle brackets here denote a thermal average. Thus the longitudinal part of the susceptibility, $\chi_\parallel$, is given by

$$T\chi_\parallel(\mathbf{k}) = 1 - |\mathbf{M}|^2 + L^{-3} \sum_{i \neq j} \cos(\mathbf{k} \cdot \mathbf{r}_{ij})(\mathbf{S}_i \cdot \mathbf{S}_j - Q_{ij}). \tag{3}$$

For Heisenberg spins,

$$Q_{ij} = \langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_j \rangle, \tag{4}$$

and

$$|\mathbf{M}|^2 = L^{-3} \sum_i Q_{ii} = L^{-3} \sum_i [\langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_i \rangle]_t, \tag{5}$$

where the square brackets $[\cdots]_t$ indicate a time average. Q_{ij} must be included for all T. Note that the $\chi_\parallel$ we define here is not exactly what would be called the longitudinal susceptibility in a nonrandom system. The distinction between longitudinal modes and transverse modes is not completely well defined in a system which has a local order parameter that is not fully aligned with the sample-averaged order parameter. In any case, our system has no soft modes below T_c in the ferroelectric phase, due to the cubic anisotropy.

The specific heat, c_H, may be calculated by taking the finite differences $\Delta E/\Delta T$, where E is the energy per spin. Alternatively, it may be calculated as the variance of E divided by T^2. The second method was used for the c_H numbers shown in Table I.

The data in Table I show that there are peaks in $\chi_\parallel$ and c_H at T_c. However, accurate estimates of the values of the critical exponents α and γ would require collecting a lot more data at temperatures close to T_c. As we shall see shortly, our data below T_c do not take the form of the usual critical scaling behavior. This may be partly a reflection of the fact that this type of model is expected to show replica-symmetry breaking [20] (RSB) below T_c. It has been understood for a long time that, in a mean-field approximation, RSB is a type of ergodicity breaking [21]. We are very far from mean-field theory here, however.

Define $M_x(t)$, $M_y(t)$, and $M_z(t)$ to be the averages over the lattice at time t of the components of $\mathbf{S}_i$, in the usual way. Then the cubic orientational order (COO) parameter is measured by calculating the quantity

$$\mathrm{COO} = 3\left[\left(M_x^2 M_y^2 + M_x^2 M_z^2 + M_y^2 M_z^2\right)/|\mathbf{M}|^4\right]_t, \tag{6}$$

The square brackets, $[\ldots]_t$, indicate a time average. The possible values of COO range from 0 when $\mathbf{M}$ points in a [100] direction to 1 when $\mathbf{M}$ points along a [111] direction. Note that if all of the spins were fully aligned along one of the [110] directions, the value of COO would be 3/4. Table I shows that the sample average of COO is approximately 0.60 in the paraelectric phase, and that it rapidly increases to 1 as T is reduced below T_c. This behavior indicates that the [111] orientational ordering is irrelevant at the random-field Heisenberg critical point, just as it is irrelevant at the pure Heisenberg critical point.

The structure factor, $S(\mathbf{k}) = \langle |\mathbf{M}(\mathbf{k})|^2 \rangle$, for Heisenberg spins is

$$S(\mathbf{k}) = L^{-3} \sum_{i,j} \cos(\mathbf{k} \cdot \mathbf{r}_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j, \tag{7}$$

where $\mathbf{r}_{ij}$ is the vector on the lattice which starts at site i and ends at site j. When there is a phase transition into a state with

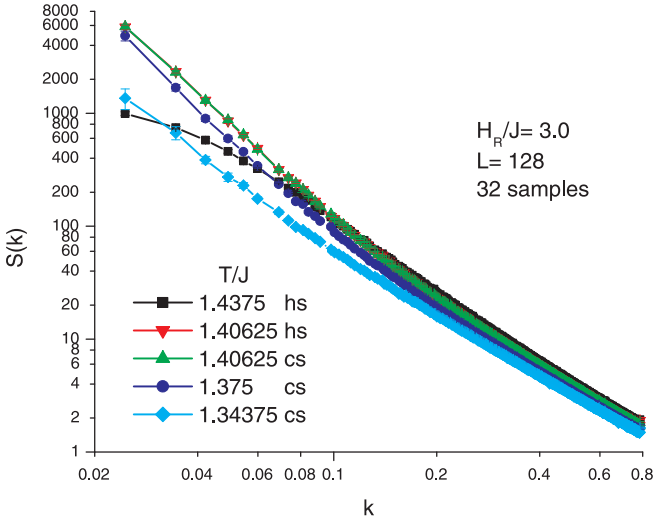

FIG. 1. Angle-averaged magnetic structure factor, $S(|\mathbf{k}|)$, at a sequence of temperatures for the RP2 $\mathbf{O}_{12}$ model with $h_R = 3$ on $128 \times 128 \times 128$ simple cubic lattices, log-log plot. The points show averaged data from 32 samples, at a series of temperatures. (h) and (c) signify data obtained by relaxing from hotter and colder initial conditions, respectively.

long-range spin order, $S(\mathbf{k} = 0)$ has a stronger divergence than $\chi(\mathbf{k} = 0)$ does.

Values of $S(|\mathbf{k}|)$ calculated by taking an angular average for each sample and then averaging the results for the 32 $L = 128$ samples are shown in Fig. 1. In Fig. 2, the results for the 15 smallest nonzero values of $|\mathbf{k}|$ at $T = 1.375$, 1.390 625, and 1.406 25 are shown. Note that the data shown for the cold initial condition and the hot initial condition at $T = 1.406\ 25$ are virtually indistinguishable. At lower temperatures, two of

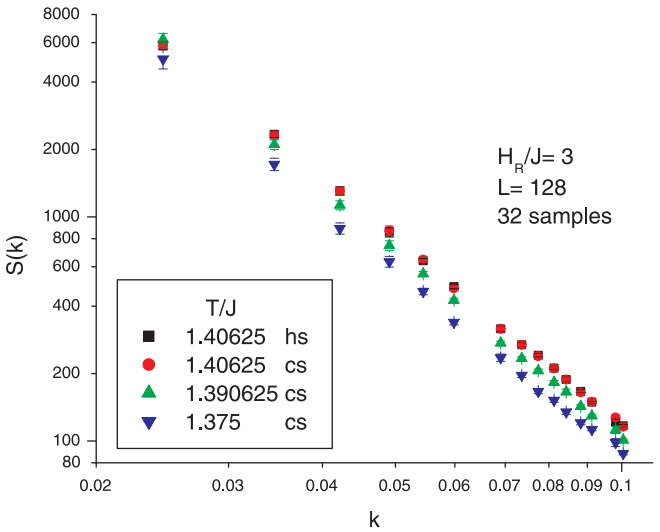

FIG. 2. Angle-averaged magnetic structure factor, $S(|\mathbf{k}|)$, at $T/J = 1.375$, 1.390625 and 1.40625 for the RP2 $\mathbf{O}_{12}$ model with $h_R = 3$ on $128 \times 128 \times 128$ simple cubic lattices, log-log plot. The points show averaged data from 32 samples. (h) and (c) signify data obtained by relaxing from hotter and colder initial conditions, respectively.

the hot initial condition samples became trapped in metastable states.

IV. DISCUSSION

The RP2 random-field distribution defined by Eq. (2) is analogous to a quantum random field built with spin-2 operators. Since a single-site operator term in a Hamiltonian of quantum O(3) spins which favors the [111] directions is also a spin-2 operator, it seems natural that this type of random field can give a direct transition from the paraelectric phase into a ferroelectric phase having random-field characteristics. This will only happen if h_R is chosen appropriately. The author expects that when h_R becomes very large, a phase with quasi-long-range order will be seen [1]. It remains to demonstrate this. If that does not happen, then the next step would be to add a third random Potts field, i.e., to study the RP3 model.

The fact that for the range of $|\mathbf{k}|$ shown in Fig. 2 the straight-line fit to the data for $T = 1.406\ 25$ is essentially perfect is remarkable. This only a numerical accident, however, since we did not tune the temperature to find this condition. The fit of the hot initial condition data to a straight line has a slope of -2.788 ± 0.014, and the fit to the cold initial condition data has a slope of -2.784 ± 0.015. Averaging these gives

$$-(4 - \bar{\eta}) = -2.786 \pm 0.014 \tag{8}$$

as the slope of $S(|\mathbf{k}|)$ on the log-log plot at the critical point in the scaling region. We do not reduce our error estimate, because the data for the different initial conditions on the same set of samples cannot be considered to be statistically independent. Thus

$$\bar{\eta} = 1.214 \pm 0.014, \tag{9}$$

which is a quite reasonable value for this quantity. The value of $\bar{\eta}$ should not be sensitive to varying T by a small amount away from $T = 1.406\ 25$. This is shown by the fact that the data in Fig. 2 for $T = 1.390\ 625$ run parallel to the data for $T = 1.406\ 25$ over a range of $|\mathbf{k}|$, and for smaller $|\mathbf{k}|$ the slope of $S(|\mathbf{k}|)$ becomes more negative. What this means is that $\bar{\eta}$ is not a continuous function of T. We are not seeing a critical phase of the Kosterlitz-Thouless type.

This effect will eventually be cut off by the cubic anisotropy, which stabilizes the ferroelectric order. What this means is that there is a strong correction to finite-size scaling of the ferroelectric order parameter below T_c. This effect was already seen in the author's earlier work [1]. This strong correction to scaling, which is dramatically different from what is usually seen at a second-order phase transition, is likely what gives rise to the experimentally observed relaxor ferroelectric effect. The author believes that this strong correction to scaling is a direct result of the dangerous irrelevant variable, the cubic anisotropy. This causes the COO to vary rapidly as a function of T just below T_c, as shown in Table I.

For larger $|\mathbf{k}|$ the slope of S is less negative, because the system is then in the crossover region between the pure system fixed point and the random-field fixed point. A calculation for $h_R = 4$, similar to the one described here, is currently in progress. The scaling region should then extend to larger values of $|\mathbf{k}|$. Incomplete results appear to show that the value

of $\bar{\eta}$ is smaller for $h_R = 4$. We are also obtaining data at negative values of h_R. The results for negative h_R are qualitatively similar to the results for positive h_R.

We are not claiming to understand completely why this behavior occurs in our fairly simple model. We only claim that we have shown that this behavior, which is similar to the observed central peak in $S(|\mathbf{k}|)$ seen in experiments, does not require the inclusion of any additional feature, such as chemical correlations, in the model.

V. CONCLUSION

In this work we have used Monte Carlo computer simulations to study a model of discretized Heisenberg spins on simple cubic lattices in 3D, with a carefully chosen type of random field. We have found that, for the random-field distribution used here, our system shows a sharp critical point and a transition into a phase with long-range order. The phase transition shows characteristics expected of random-field behavior. This example shows that the extreme difficulty in reaching equilibrium which is usually associated with glassy behavior is not an inevitable consequence of the presence of random fields. Standard renormalization group arguments imply that a similar transition ought to exist in a 3D model of this type with continuous spins and random fields which have an isotropic probability distribution. The primary reason why the existence of such behavior has been considered impossible until now is that the non-Abelian nature of the O(3) group, which can give rise to relevant higher-order tensor couplings to the random field, was not properly taken into account. We have also identified the relaxor ferroelectric effect as arising from a strong correction to scaling of the ferroelectric order parameter near T_c, which is probably caused by the cubic anisotropy, which is present in this model and also in the cubic perovskite relaxor ferroelectrics. This strong correction to scaling may not occur in a model with isotropic spins and an isotropic distribution of random fields, or in an experimental system which is not a crystalline alloy.

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