Irreversibility and metastability in spin-glasses. I. Ising model

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By studying the evolution (with field H and temperature T) of the free-energy surface, we numerically calculate various history-dependent magnetizations. The interactions are assumed to be of short range with a Gaussian probability distribution. In this paper, only Ising spin-glasses are discussed. In a companion paper, we treat vector spins. Only the simplest mean-field (MF) approximation to the free-energy functional $F[\{m_i\}]$ (where m_i is the thermally averaged spin at the ith site) is studied. For finite-size systems, it is difficult to include the reaction corrections to F within a self-consistent formalism. Good qualitative agreement with experiment is found for the T and H dependence of the field-cooled and zero-field-cooled, and thermal and isothermal remanent magnetizations. Our hysteresis loops are qualitatively similar to those observed in AuFe spin-glasses. The characteristic changes in the shape of the loop as the Fe concentration increases can also be reproduced theoretically. At $T=0$, the corrections to MF theory are negligible and it is shown that the ground states generated iteratively by cooling solutions of the MF equations are obtained much more rapidly and are of lower energy than those found in Monte Carlo simulations. It is demonstrated that the field-cooled state, which is obtained by a series of reversible procedures, is one for which linear-response theory (properly interpreted) and Maxwell's relations all hold. The close correspondence between our theory and experiment supports the hypothesis that on intermediate-time scales the behavior of spin-glasses is primarily determined by the properties of the free-energy surface: Irreversibility occurs when minima of F are destroyed with changing H or T.

I. INTRODUCTION

Recently, experimental[1-6] and theoretical[7-13] studies of spin-glasses have become increasingly concerned with understanding irreversible and time-dependent effects. The primary focus of new theoretical work[7-13] in the field has been to characterize spin-glass states within a nonequilibrium framework. This represents a shift from an earlier emphasis[14-16] in which it was hoped that a thermodynamic equilibrium state existed and could be treated at the level of generalized mean-field theories. Such equilibrium approaches have not been particularly successful and it is not clear under which circumstances, if any, true thermodynamic equilibrium holds in a laboratory spin-glass. The most likely candidate for the equilibrium state is that obtained upon cooling at constant field.[8,17] The failure of equilibrium thermodynamics, which has been claimed[18] for finite-range interactions, may be connected to the breakdown of ergodicity. The spin-glass does not sample equally all accessible states in the time scale of a laboratory experiment.

Attempts to characterize the nonequilibrium aspects of spin-glasses have proceeded along three main lines. (i) Sompolinsky[9,10] and Hertz[11] have shown that Parisi's[7] novel replica-symmetry-breaking ideas can be more readily interpreted within a dynamical framework. The time evolution of the spin-glass (macro)states plays a central role in their work. (ii) Monte Carlo simulations[12] reproduce the experimentally observed irreversibility effects[1-6] and give evidence for nonergodicity.[19] More importantly, however, recent work in which Monte Carlo and transfer-matrix techniques[18] are combined allows a direct comparison of properties of the finite-N thermodynamic equilibrium state with those of a "laboratory" spin-glass. (iii) Finally, in recent work the present authors have focused on metastable states of the spin-glass free energy.[20,21] These are believed to be relevant to intermediate time-scale experiments during which a spin-glass can "follow" a given minimum on the free-energy surface as it evolves with changing field H or temperature T. The results obtained numerically by following the evolution of a free-energy minimum have been found to yield various history-dependent magnetizations which are in good qualitative agreement with experiment. It is the purpose of the present paper to expand upon our earlier[20] Letter on this subject, which addressed Ising spin-glasses only. A companion paper[22] is devoted to a study of Heisenberg spins, which follows a second Letter[23] on the vector spin-glasses.

Our approach is implicitly nonergodic. A given minimum on the free-energy surface, in which the spin-glass is originally prepared, is singled out and followed with temperature or field. Clearly, on a long time scale the system will generally find its way out of this minimum either by tunneling or thermal activation. On too short a time scale the spin-glass may not be able to relax to the nearest minimum since a change in temperature or field distorts the free-energy surface so that the initial minimum may be displaced or disappear altogether. It is

only on this intermediate time scale that a characterization of the free-energy surface can be directly relevant to the behavior of the various physical properties of the system. The time intervals of interest correspond experimentally to those observed after the initial rapid decrease in magnetic remanence[7] but before the $\ln t$ (where t is the time variable) decay has become appreciable.

A number of investigators[5,24–27] have argued at a qualitative level that the characteristics of the free-energy surface are directly related to irreversibility effects in spin-glasses. Our work represents the first attempt to make this interrelationship quantitative. We propose that the appropriate configuration space for studying the free energy F is as a function of the thermally averaged values $\{\langle S_i \rangle\} \equiv \{m_i\}$ at each site i. An exact expansion for the functional $F[\{m_i\}]$ was determined for the infinite-range Ising model by Thouless, Anderson, and Palmer (TAP).[28] Numerical attempts to follow (with temperature) minima of the TAP free energy have consistently yielded[29,30] unphysical results. This will be discussed in more detail in Sec. II. For this reason we will consider here (and in the companion paper) the free-energy surface corresponding to simple mean-field theory.[20] It should be noted that in the low- and high-temperature limits, the TAP expression for F reduces to that derived in mean-field theory. While this provides some justification, the major support for adopting this approximation is that it seems to work extremely well. It also leads to useful insights into the behavior of spin-glasses which would not otherwise be obtainable.

A major advantage of our mean-field approach is that the metastability or variational condition $\partial F/\partial m_i = 0$ can be solved by simple iterative techniques for fixed H and T. All states so generated are minima on the free-energy surface.[30] They can be readily followed with H and T. Because no matrix algebra is required we can treat fairly large systems ($N \lesssim 5\times 10^4$ for Ising spins). Furthermore, the results are easily generalized to the vector-spin case, as will be discussed in the following paper.[22]

The main content of this work is the presentation of results for the field-cooled (FC) and zero-field-cooled (ZFC) magnetizations, for magnetic hysteresis and for thermal remanent (TRM) and isothermal remanent magnetizations (IRM) as functions of temperature T and field H. We also discuss the field-dependent specific-heat and ground-state properties. It is pointed out that our $T=0$ algorithm is equivalent to that used in Monte Carlo calculations for obtaining ground states. However, our iterative mean-field approach is a factor of 10—100 times faster and yields slightly lower energy ground states.

Our results can be directly compared to experiment as well as to previous theoretical calculations. Kinzel[12] has performed Monte Carlo simulations on a 50×50 spin-glass system. The FC, ZFC, and remanent magnetizations he obtains all compare favorably with our results (as well as with experiment). However, Monte Carlo techniques allow a direct comparison of the time dependence of various properties which we will not address here. Dasgupta, Ma, and Hu[13] have also calculated dynamical properties of spin-glasses using a semiphenomenological model. They compute the distribution of barrier heights between ground states, generated by Monte Carlo techniques (with the use of a 20×20 Ising lattice). These are then combined with the simple dynamics of the kinetic Ising model to obtain various (time-dependent) magnetizations. When comparison is possible, our results are in qualitative agreement with those of Ref. 13. It should be stressed, however, that our approach is basically nonergodic, whereas Dasgupta *et al.*[13] treat all degenerate ground states on an equivalent basis. The states that we focus on for a fixed T or H are those which are accessible, given the history of the system. While these authors can treat the time evolution of various spin-glass properties, they are not able to associate the onset of reversibility with any particular temperature. Thus the transition temperature T_c, which is found experimentally[2,5,6] to be intimately connected with reversibility properties, does not have a role in the work of Ref. 13.

An outline of the paper is as follows. In Sec. II we review the equations and describe the numerical techniques used to follow the various free-energy minima. In Sec. III we list our general results which pertain to (i) the transition temperature, (ii) fluctuation and time-dependent effects, and (iii) the validity of linear-response theory. Section IV presents a comparison of Monte Carlo ground states with those we generate by cooling (in zero field). Our two-dimensional (2D), $S=\frac{1}{2}$ free energies are essentially the same at $T=0$ as those found by "exact" techniques.[18] We discuss the temperature dependence of the field-cooled and zero-field-cooled magnetizations in Sec. V, magnetic hysteresis in Sec. VI, and the various remanences in Sec. VII. The properties of the field-dependent specific heat are discussed in Sec. VIII. Section IX presents results for the distribution of local molecular fields $P(H_i)$. Finally, in Sec. X we list our conclusions.

Throughout this paper considerable effort is made to present our theoretical results alongside with experimental data. However, it should be stressed that we do not make any attempt to quantitatively fit experiments. Furthermore, it appears inappropriate to do so. We are dealing with an oversimplified [non-Ruderman-Kittel-Kasuya-Yosida (RKKY)] Hamiltonian, treated within the mean-field approximation and solved for finite-size systems. While this approach is clearly oversimplified, it provides useful insight into the highly complex free-energy surface, which we believe is intimately connected with irreversibility in spin-glasses.

II. DESCRIPTION OF THE METHOD

As in most theoretical descriptions of Ising spin-glasses we consider the Hamiltonian with near-neighbor interactions

$$\mathcal{H} = -\sum_{i,j} J_{ij} S_i S_j - \sum_i S_i H \ , \tag{2.1}$$

where i,j are taken over all sites in the lattice and J_{ij} is given by a Gaussian probability distribution $P(J_{ij})$ of width $\bar{J}$ and center J_0. In what follows, T, H, J_0, etc., will all be measured in units of $\bar{J}$; also we take the magnetic moment $g\mu_B \equiv 1$. The general free-energy functional $F[\{m_i\}]$ can be decomposed into the mean-field and so-called reaction terms

$$F[\{m_i\}] = F^{\mathrm{MF}}[\{m_i\}] + F^{\mathrm{reac}}[\{m_i\}] \ , \tag{2.2}$$

where for general S, such that $-S \le m_i \le S$,

$$F^{\mathrm{MF}}[\{m_i\}]=\tfrac{1}{2}\sum_{i,j}J_{ij}m_im_j - k_BT\sum_i \ln\frac{\sinh[(S+1/2)\bar{H}_i]}{\sinh(\bar{H}_i/2)} \quad (2.3a)$$

We will use the symbols M_0 and S interchangeably throughout this paper. Here

$$\bar{H}_i\equiv\beta\sum_j J_{ij}m_j+\beta H,\quad \beta^{-1}=k_BT\ .$$

In the case $S=\frac{1}{2}$,

$$F^{\mathrm{MF}}[\{m_i\}]=-k_BT\sum_i \bar{H}_im_i\sum_i H_im_i + k_BT\sum_i[(\tfrac{1}{2}+m_i)\ln(\tfrac{1}{2}+m_i) + (\tfrac{1}{2}-m_i)\ln(\tfrac{1}{2}-m_i)]\ . \quad (2.3b)$$

For the infinite-range Ising spin-$\frac{1}{2}$ model, Thouless, Anderson, and Palmer have shown that[28]

$$F^{\mathrm{reac}}[\{m_i\}]=-\tfrac{1}{2}\beta\sum_{i,j}J_{ij}^2(1-4m_i^2)(1-4m_j^2)\ . \quad (2.4)$$

In general, the reaction term derives from the fact that a "self-orientation" effect, present in F^{MF}, should be cancelled out. TAP have pointed out that their expression for F is valid subject to a "convergence criterion." Thus not all solutions of the extremal condition $\partial F/\partial m_i=0$ are physical.

A number of different groups have searched for minima of the TAP free-energy surface. Bray and Moore,[31] using iterative techniques, found a solution (at a given T) in about 10% of all bond configurations. Later attempts[29,30] to follow these solutions with varying temperature indicated that they quickly disappeared and that no nearby state was found to which the system flowed. More recently Ling *et al.*[30] reexamined these earlier calculations. In an attempt to determine whether the unphysical results obtained were a consequence of inadequate numerical approaches, an improved scheme was devised. Even with this more precise approach, unphysical results are found; it was shown that the FC state evolved into one of negative entropy at low T and that the solutions to the TAP equations were, indeed, piecewise continuous, at best. Dasgupta and Sompolinsky[32] have recently generated some solutions using Monte Carlo slow-cooling procedures. These also appear to have unphysical properties.[30] Presumably these unphysical results arise because the reaction term in Eq. (2.4) must be corrected in small-size systems.[29] At present there is no fully consistent theory of these corrections.

Because a characterization of the spin-glass free-energy surface appears to be so important to our understanding of these materials, we have set out to study the more well-behaved mean-field limit. It should be recalled that at very low and high temperatures the TAP self-consistent (or variational) equations are equivalent to those derived from F^{MF}. In contrast to the TAP case, however, there are no unphysical extrema: The entropy is always well behaved. This seems to be in part responsible for the fact that mean-field theory "works" as well as it does.

We solve iteratively the self-consistent equations deriving from $\partial F/\partial m_i=0$:

$$m_i=SB_s(\bar{H}_i)\ , \quad (2.5a)$$

where B_s is the Brillouin function for general S values. In the limit $S=\frac{1}{2}$, this equation reduces to

$$m_i=\tfrac{1}{2}\tanh\left[\frac{\beta}{2}\left(\sum_j J_{ij}m_j+H\right)\right]\ . \quad (2.5b)$$

Convergence is assumed when

$$\frac{\sum_i[(m_i)_n-(m_i)_{n-1}]^2}{\sum_i[(m_i)_n]^2}\le 10^{-6}\ , \quad (2.6)$$

where the subscript n denotes the nth iteration. The results were not different (to within five decimals) from those obtained when a much more stringent convergence criterion of 10^{-12} was applied.[20] However, we found that introducing larger values for this cutoff led to inaccuracies. It has been shown[30] that all solutions to Eqs. (2.5) which are obtained iteratively are minima. However, not all minima are generated in this way. It is possible, in principle, to "lose" a solution as T or H varies due to the failure of iterative convergence, rather than to the disappearance of a minimum. However, it has been pointed out[30] that in mean-field theory the disappearance of a given minimum closely coincides with the failure of iterative convergence. This result was observed empirically and has not been proven analytically. On this basis it appears that a simple iteration procedure is sufficient for mean-field theory and that solutions to the self-consistent equations will not be "lost" due to numerical artifacts. It should be noted that all of these results were established in the limit where Eqs. (2.5) are solved iteratively, without updating. That is, only after all the new m_i are computed are these values substituted into the argument of the Brillouin function. As in most numerical approaches, however, we will take advantage of the fact that updating the m_i as we iterate leads to much more rapid convergence. To monitor the validity of this updating scheme, we have checked our numerical results by varying the order in which we rotate through the site indices i in Eqs. (2.5). The effect of changing this sequencing is discussed below. In general, because of the insensitivity of our results to the sequencing order, we argue that those results obtained after updating our iterations are not significantly different from those obtained with no updating.

The numerical calculations were done on an IBM 4341 computer. In most instances we began at high $T=2T_c$, where T_c is the spin-glass transition temperature, and cooled in zero or finite field. The iterations at high T were started by choosing the values of the $\{m_i\}$ randomly. Convergence (to an accuracy of 10^{-6}) typically required 50 iterations for a 100×100 Ising system. The temperature was then decreased in small steps of about $0.2T_c$. At each subsequent T, the converged values at the previous temperature were used to start the iterations. For $T\approx0$ only a few (less than 10) iterations were needed. For more stringent convergence criteria the number of iterations clearly increased. In general, the order in which the site indices i were chosen, was generated randomly. Field changes were typically made after cooling in zero field. To study magnetic hysteresis, for example, the iterations were started at the converged values for $H=0$ and the

magnitude of the field was then increased in units of $\Delta H=0.2\bar{J}$ (for large values of H) and $\Delta H=0.05\bar{J}$ (for smaller H). In this way it is assumed that the system "follows" a given minimum of the free-energy surface as it evolves in H or T, and when a minimum disappears the system "hops" to a new, but nearby state.

Anderson[33] has pointed out that localization effects may be important in the mean-field-theoretic treatment of spin-glasses. He based his argument on the observation that the *linearized* mean-field self-consistent equations reduce to an eigenvalue problem. Localized eigenstates cannot correspond to long-range order; as a consequence T_c must be renormalized to lower temperatures. In our calculations we have found that the eigenfunction solution for the m_i is not generated by iterative techniques. It is a higher-order minimum or saddle point. While localization effects presumably play a role in renormalizing T_c, it may not be inappropriate to neglect them at this stage, since finite-size and magnetic field effects make any determination of T_c inaccurate.

III. GENERAL RESULTS

A. The transition temperature

The character of the free-energy surface is believed to be strongly temperature dependent. Above the transition temperature T_c there is only one minimum in F (for any H). Below T_c, at $H=0$, the system continuously develops additional new minima, as T is decreased. At finite $H\neq 0$, the temperature at which the number of minima first exceeds unity is less than T_c. The phase diagram for this field-dependent transition $T_c(H)$ has been determined experimentally[5,17,34] as well as theoretically.[35] In numerical calculations one can determine $T_c(H)$ only approximately because of finite-size effects. We define T_c as the lowest temperature at which (in an extrapolated thermodynamic limit)

$$Q\equiv\frac{1}{N}\sum_i m_i^2 \tag{3.1}$$

is nonzero. Finite-size effects always lead to a "tail" in Q at high T, so that it is necessary to consider the systematic variation of Q as the size of the system N increases. We estimate (for a nearest-neighbor interaction model) $T_c=0.86\bar{J}$ for a 2D Ising system and $T_c=1.125\bar{J}$ for the three-dimensional (3D) case, with $S=\frac{1}{2}$. For 3D spin-glasses with $S=1$ and $\frac{5}{2}$, $T_c=3.1\bar{J}$ and $13.0\bar{J}$, respectively. It is important to note that these transition temperatures coincide closely with the temperature at which the ZFC magnetization (M^{ZFC}) has a maximum at the lowest H values we considered.

In order to obtain the field dependence of T_c we studied the variation with H of the maximum in M^{ZFC}.[36] According to predictions[8,35] for the infinite-range model, at low H, $T_c(H)/T_c(0)=1-(\frac{3}{4})^{1/3}H^{2/3}$. We find for a 2D, $S=\frac{1}{2}$ spin-glass that our calculated $T_c(H)$ fits this dependence very accurately; the coefficient of the $H^{2/3}$ term is $0.91\approx(\frac{3}{4})^{1/3}$. Experimentally,[5,17] however, it is found that this coefficient is roughly 10 times larger. This discrepancy appears to be a general problem which has plagued spin-glass theories. A possible resolution[37] of this difference is to assume that the effective moment of the local spins is enhanced by a factor of 10, as would be the case if there were short-range ferromagnetic order present. In this way, one obtains a stronger sensitivity to magnetic fields in χ and other properties, as is observed experimentally.

B. Fluctuation and time-dependent effects

Within the context of our mean-field-theoretical calculation, fluctuation effects are ignored. The spin-glass is assumed to "sit" in the bottom of a given well as long as it remains a minimum and, otherwise, to hop into a nearby state. We cannot treat the dynamics of this hopping process; it is assumed to occur "instantaneously." Furthermore, we do not consider tunneling or thermal activation processes which take the system from one metastable state to another. Nevertheless, our picture may shed light on these two phenomena; the experimentally observed "fast" and "slow" decay of, say, magnetic remanence, may be associated with a rapid relaxation to the nearest minimum and very much slower ($\sim\ln t$) thermal activation processes, respectively. The fast and slow time scales presumably vary from system to system. Krey[25] has argued that in *Cu*Mn the former corresponds to $t<10^{-11}$ sec and the latter to $t>10^{-10}$ sec. These times are somewhat temperature dependent. Our mean-field-theoretical approach is designed to treat only those experimental measurements which have very slow time dependences ($\sim\ln t$, or slower), so that the system is always "quasiequilibrated." It should be viewed as the first of a series of studies. Once the behavior of the minima on the free-energy surface is understood, the next step is to determine barrier heights between minima and to, thereby, build in fluctuation and dynamical effects.

C. The behavior of the free-energy surface

Our most general observations about the way in which the free-energy surface evolves with H and T are as follows:

(i) A free-energy minimum will never disappear upon cooling (at constant H).

(ii) Below $T_c(H)$, a minimum will generally disappear upon heating (unless the minimum was obtained by a cooling procedure).

(iii) For $T<T_c(H)$ minima appear to be continuously created as well as destroyed, upon changing H, by small but finite amounts.

There is thus a "directionality" associated with the way in which the surface evolves with temperature changes. This is plausible since at high T there is only one minimum in $F[\{m_i\}]$ corresponding to the paramagnetic state $m_i=0$, for all i, whereas at low T the number of minima is very large.[31] It is not surprising that, as the spin-glass is warmed, minima are destroyed. As T increases, a finite-size system ($N<100$) may make a series of observable first-order transitions (if it was not initially in a state obtained by cooling at constant H). Each time a state becomes unstable, the spin-glass finds its way to a nearby minimum. As the system gets arbitrarily large, the m_i appear to change nearly continuously with increasing T. However, cooling from $T_0+\Delta T$ to T_0 after heating from

T_0 to $T_0+\Delta T$ leads to a different state at T_0. This suggests that there have been many first-order jumps in the heating process. The converse is not true. If the system is cooled (at constant field) from T_0 to $T_0-\Delta T$ and then warmed back up to T_0, the same state is found.

As in a warming procedure (at constant H), magnetic hysteresis is found when H is changed at constant T. This hysteresis, or irreversibility, arises from the destruction of minima on the free-energy surface, as H varies. There is no directionality to this irreversibility. Changing H by positive or negative amounts always leads to magnetic hysteresis below $T_c(H)$.

D. Zero-field-cooled states

Zero-field-cooled (ZFC) states were obtained by cooling in zero H from high $T=(1.5-2.0)T_c$ to $T\approx 0$. As expected these states were found to be reversible with respect to subsequent heating, as long as the magnetic field H was kept at a constant (zero) value. We found that for fixed $\{J_{ij}\}$ and $H<0.01\bar{J}$, several different states were obtained depending on the choice of random numbers m_i which were used at the highest T for the first iteration. Once this was fixed, the order in which we rotated through the site indices in our subsequent iterations did not affect any results. While we did not generate unique ZFC states, we found that when a small constant ($H>0.01\bar{J}$) field was applied during cooling, the result was a unique state at each T. It is probable that the critical field (above which a unique low-temperature state is obtained) varies with the size of the system as $\sim 1/\sqrt{N}$. We could not ascertain unambiguously whether there is a unique ZFC state. It is possible that when the cooling procedure is started at sufficiently high T only one ZFC state would be found. However, as T is increased the values of the m_i are so close to zero that we ran into numerical accuracy problems deriving from the convergence criterion [Eq. (2.6)]. We consistently found that the lowest-energy (ZFC) ground states were obtained by starting at the highest possible T that our numerical program could handle.

To measure the magnetization in a ZFC state, a field was rapidly applied after cooling to the lowest T. The temperature-dependent magnetization curve was then obtained by warming from low T. This sequence of steps corresponds to the experimental procedure.[3,5] It is important to realize that after the field is applied, and the temperature raised, a subsequent cooling leads to irreversibility.[5] The resulting magnetizations are discussed in detail in Sec. V. Our results for $M^{\mathrm{ZFC}}(T)$ are slightly sensitive (within 5%) to the order in which we sequence the site indices as the field is turned on or the temperature is raised. This follows because as H or T changes, minima are destroyed and the system goes to a nearby state. It is not surprising that the state to which the system flows depends on the order of sequencing.

E. Field-cooled states

The states obtained upon cooling at constant field $H\neq 0$ were found to be unique: They were insensitive to our choice of random variables for the m_i at the highest T and to our sequencing procedure. It follows from the discussion in Sec. III C that the field-cooled magnetization is reversible. In a field-cooled measurement, the system does not "minima hop."

We observed that the free energies of the FC states were the lowest of any states we generated [at the same (H,T)]. Since we did not make a systematic search, it is possible that there are other states degenerate with or of even lower energy than the FC state. Experimental evidence[2,3] suggests that there is no relaxation out of the FC state so that if there are deeper minima, they appear to be inaccessible on a laboratory time scale. Theoretical studies[18] for $d\leq 3$ and, in the case of finite-range interactions, suggest that there may be no spin-glass order in thermodynamic equilibrium. A way of reconciling these two points of view is that the FC state is a state of very long-lived metastable equilibrium. If so, then even the field-cooled state represents a nonergodic situation.

F. Validity of Maxwell's relations

In order to test the Maxwell relation[38–43]

$$\left[\frac{\partial^2 M}{\partial T^2}\right]_H=\frac{1}{T}\left[\frac{\partial C_H}{\partial H}\right]_T, \tag{3.2}$$

we numerically studied the lower-order derivatives

$$M=-\left[\frac{\partial F}{\partial H}\right]_T \tag{3.3a}$$

and

$$S=-\left[\frac{\partial F}{\partial T}\right]_H \tag{3.3b}$$

in both ZFC and FC states. We found that for finite-size systems these equations were *only* satisfied in the FC state. For rather small ($N<100$) ZFC systems, M vs H consists of a series of jumps between smooth regions; it is, thus, not surprising that the derivative $\partial F/\partial H$ is ill defined. These jumps are present whenever H is changed below T_c. For $T>T_c(H)$, M vs H is a continuous function (since there is only one minimum at each H). Because the FC state involves changing H above T_c and then cooling (both of which are reversible processes), the derivatives in Eqs. (3.3) are well behaved and satisfy the equalities. For the ZFC state, when the size of the system increases, M vs H smooths out considerably below T_c, and Eqs. (3.3) are closer to being satisfied. However, for finite-size systems these equations are never strictly satisfied in the ZFC state. It is not clear what happens in the thermodynamic limit. We believe this issue, which depends on the smoothness or differentiability of the M vs H curves, is directly relevant to the validity of linear-response theory. This will be discussed in the following section (III G).

The Maxwell relations appear to hold experimentally[39,41,42] for the FC state, in agreement with our theoretical conclusions. It should be stressed that this is a unique state: Only under field-cooled conditions are changes in H and T made in a reversible manner. These relations will hold even in the presence of nonergodicity, provided there is no relaxation out of the field-cooled state.

G. Validity of linear-response theory

Whether linear-response theory (LRT) is valid in spin-glasses is a question which has received much atten-

tion[9,11,24] in recent years. The issue is somewhat clouded by the fact that LRT, in which the spin susceptibility χ is given by

$$\chi=\beta\sum_{i,j}[\langle S_iS_j\rangle-\langle S_i\rangle\langle S_j\rangle]\ , \tag{3.4}$$

and the Fischer relation[16] $\chi=(1-\tilde{Q})/T$ are sometimes viewed as equivalent. Here $\tilde{Q}$ is the thermodynamic spin-glass order parameter

$$\tilde{Q}=\langle(m_i)^2\rangle_c\ ,$$

where the angular brackets with subscript c denote a configuration average. As Young and Kirkpatrick[44] and Fischer and Hertz[45] have pointed out, lack of ergodicity and/or the presence of a small magnetic field may lead to violations of the Fischer relation, even in instances where the fluctuation-dissipation theorem (from which this relation is derived) is valid. To avoid this confusion, in what follows we define LRT to be equivalent to Eq. (3.4), where the thermal averages over both sides of the equation are taken over an appropriately restricted subset of states.

Another assumption which is implicit in the derivation of LRT is that the limit $\delta H\rightarrow 0$ of $\delta M/\delta H$, or any other thermodynamic quantity, must be well behaved. Experimentally, it appears[5] that even for changes in H as small as 40 mOe, there is irreversibility, as indicated by long time relaxation processes. In the present picture, this suggests that the minimum in F at 40 mOe has not evolved continuously from a zero-field minimum, i.e., there is "minima hopping" even for this extremely small field change. As Chamberlin *et al.*[5] point out "the field truly 'scrambles' the surface."

Bray and Moore[24] have proposed, on the basis of replica symmetry breaking, that LRT may be violated due to the fact that $\delta M/\delta H$ is not well defined as $\delta H\rightarrow 0$. Our numerical studies can provide insight into whether or not this "scenario" is reasonable. From our calculations (extrapolated to $N\rightarrow\infty$) we have been unable to find a δH sufficiently small such that M is clearly reversible with respect to variations of order δH, below T_c. That is, we always see magnetic hysteresis so that the value of $\delta M/\delta H$ is path dependent. While this is consistent with experiment,[5] any extrapolation procedure is suspect; it is extremely difficult to determine whether in the true thermodynamic limit these results are valid. On this basis it cannot be proved or disproved that LRT holds in the ZFC state. By contrast, in the FC state, in which H is always changed above T_c, there is no ambiguity and the limit of $\delta M/\delta H$ as $\delta H\rightarrow 0$ is well defined. In this case, we do not believe LRT, as we have defined it, can be violated. In summary, the fundamental issue which determines the validity of LRT under more general circumstances, is whether or not $\delta M/\delta H$ reaches a well-defined limit for small enough δH. Even if this does occur, it is important to note that experimentally controllable field changes appear to be beyond the limits of the validity of LRT.

IV. PROPERTIES OF THE GROUND STATE

The algorithm for defining metastable $T=0$ states in Monte Carlo simulations is equivalent to that obtained from mean-field theory at $T=0$ [Eqs. (2.5)]. It is, therefore, useful to compare the lowest-energy ground states found using a slowly cooled Monte Carlo procedure and a "slow-cooling" iterative solution of the mean-field equations.

In the iterative approach very-low-energy ground states are found. As shown in Table I, these are lower in energy than those generated by Monte Carlo techniques; in 2D, the energies E_0/M_0^2 are extremely close to the "exact" values discussed by Morgenstern and Binder.[18] It is possible that, because the iterative approach closely follows the (unique) high-temperature state as it evolves with temperature (rather than hopping over barriers as in Monte Carlo calculations), it always finds the best ground state. An added advantage of the mean-field method is that it is considerably faster than simulation techniques. The numbers shown in Table I represent an average over the bond configurations of 100×100 Ising spin-glasses (in 2D) in $30\times 30\times 30$ (in 3D). The numbers shown in parentheses were obtained from a specially selected bond distribution for which the first four moments were within 10^{-4} of the exact Gaussian values. For the 2D system the mean-field ground-state energy, in this case, coincided with that predicted by Morgenstern and Binder.[18] We also performed Monte Carlo simulations for a nearly exact Gaussian distribution and found $E_0/M_0^2=-1.28$. To estimate the relative times involved in slow cooling in Monte Carlo and iterative mean-field theory, we now outline the steps in the two procedures, in some detail, for a 100×100 spin-glass with a nearly perfect Gaussian distribution. It should be noted that it is difficult to make strict comparisons since the ground states found were slightly different (the mean-field result was of lower energy). We performed a slow-cooling Monte Carlo run by starting from a random state at $T=3.0$. We cooled in temperature decrements of $\Delta T=0.5$ for $T>1.5$ and $\Delta T=0.2$ for $T<1.5$ using 100 Monte Carlo steps (MCS) per spin at each T. This procedure took 70 min on an IBM 4341 computer and yielded a ground-state energy $E_0/M_0^2=-1.27$. Cooling at a slower rate (400 MCS per T) improved E_0/M_0^2 to -1.28

TABLE I. Comparison of ground-state energies (obtained by iterating mean-field equations) with "exact" results (from Ref. 18) and Monte Carlo simulations.

	Exact recursive method[a]	Mean-field theory[b]	Monte Carlo[a,b]
2D	-1.31 ± 0.01	-1.29 ± 0.01 (-1.30)	-1.26 ± 0.02 (-1.28)
3D	-1.71 ± 0.03	-1.65 ± 0.01 (-1.665)	-1.63 ± 0.02

[a]Reference 18.
[b]This work.

but increased the time to 280 min, while cooling more rapidly (50 MCS per T) gave $E_0/M_0^2=-1.26$ in only 35 min. These final energies are to be compared to a value of -1.13 obtained after a rapid quench from $T\gg T_c$ to $T=0$. By contrast, using iterative mean-field theory, we found a final ground-state energy -1.30 in less than 3 min. This was obtained by starting from a random state at $T=3.6$ and cooling in decrements of $\Delta T=0.2$. The convergence criterion [Eq. (2.6)] was fixed at 10^{-5}. Choosing a more stringent criterion increased the computer time necessary, but gave no improvement in E_0. Relaxing the criterion to 10^{-2} improves the computing time to 1.5 min but yields $E_0/M_0^2=-1.29$. An obvious advantage to the use of mean-field theory is the relatively rapid (by factors of 10—100) computational speed. This difference is all the more significant when it is realized that the Monte Carlo results were not run sufficiently long to obtain the best possible ground states.

It should be noted that our 3D results were not in as good agreement with those of Morgenstern and Binder[18] as our results in 2D. It is possible that, since Ref. 18 considered only very small lattices ($4\times4\times10$), the actual error bars may be larger than those quoted. Our experience for the 2D system suggests that our method should be closer to the exact answer in 3D than Table I indicates.

Monte Carlo methods have recently been applied to a number of optimization problems by Kirkpatrick, Gelatt, and Vecci.[46] They have shown that simulated annealing can be a very important way of optimizing very large and complex systems; since they have found the analogy with spin-glass ground states to be useful, it appears that our approach may also be relevant to numerical methods of optimization. Our results indicate that, at least for a certain class of problems, which can be written in terms of a spin Hamiltonian, further improvement can be achieved using iterative mean theory to find ground-state properties. One example is the electron glass which can be mapped[47] directly on to an infinite-range spin Hamiltonian.

V. RESULTS FOR THE TEMPERATURE-DEPENDENT MAGNETIZATION

In this section we discuss the temperature dependence of the field-cooled and zero-field-cooled magnetizations, called, respectively, $M^{\rm FC}$ and $M^{\rm ZFC}$ for various magnetic fields H. Our studies involved both two- and three-dimensional Ising lattices with nearest-neighbor Gaussian probability distributions, centered at J_0 and of width $\bar{J}$. We considered the values $S=\frac{1}{2}$, 1, and $\frac{5}{2}$ for the spin.

In Fig. 1, we plot the temperature dependence of the Edwards-Anderson[14] order parameter Q, for a $10\times10\times10$, $S=1$ Ising system for various J_0. The highest value, $J_0=1.0$, exceeds the critical value (~0.55) for ferromagnetism. The effect of increasing J_0 is to enhance the values of Q and to, therefore, raise T_c. Our estimated transition temperature (for $N\to\infty$) is $3.1\bar{J}$ (at $J_0=0$).

Figure 2 plots the temperature dependence of $M^{\rm ZFC}$ and $M^{\rm FC}$ (corresponding always to the lower and upper curves, respectively, in each pair at fixed H). The values of the magnetic field (in units of $\bar{J}$) are as indicated. In Fig. 2(a) is shown a 30×30, $S=\frac{1}{2}$ Ising system with $J_0=0$ and in

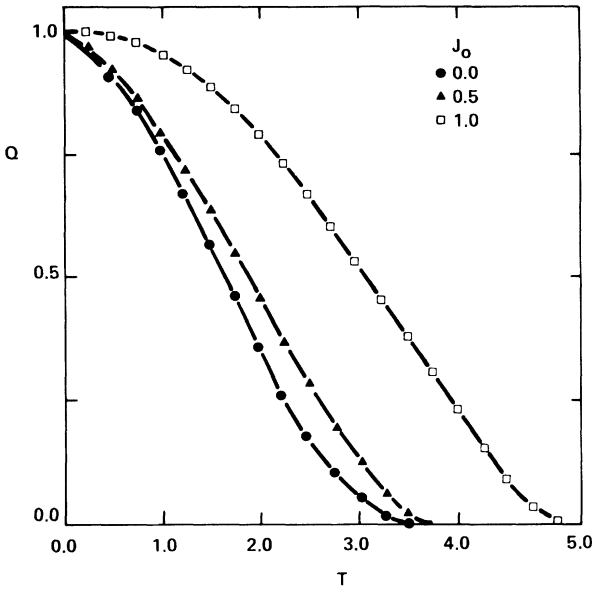

FIG. 1. Temperature dependence of spin-glass order parameter Q for $N=10^3$, $S=1$, and various J_0. The highest value, $J_0=1.0$, is ferromagnetic. All energies are measured in units of $\bar{J}$.

Fig. 2(b) a $10\times10\times10$, $S=1$ spin-glass, with $J_0=0.5$. [In the latter case, the corresponding $Q(T)$ curve is shown in Fig. 1.] This one configuration will be discussed frequently throughout the remainder of the paper. We have also studied other configurations to verify that our results are not qualitatively affected by varying the J_{ij}. All the results shown in this and subsequent sections were found to be fairly typical. The arrows in the bottom sets of curves in Fig. 2(a) indicate that the ZFC curve is obtained upon warming only. The FC curve, by contrast, is completely reversible with respect to temperature variations. A subsequent cooling of a ZFC curve, at low temperatures, leads to a new magnetization, which is indicated in Fig. 2(a) by the nearly constant, reversible curve attached to $M^{\rm ZFC}$. Note that in both figures $M^{\rm ZFC}$ has a maximum with respect to T, for sufficiently low fields. When $J_0=0$, $M^{\rm FC}$ shows the familiar temperature-independent plateau, for low T,H. This occurs, roughly, for all temperatures below that at which $M^{\rm ZFC}$ has a maximum. Presumably, even for $J_0\neq0$, this saturation of $M^{\rm FC}$ will occur for sufficiently low T and H. However, because J_0 acts to enhance the applied field, this requires much lower field values than we were able to study in Fig. 2(b). Finite-size effects necessitate the use of rather large fields in order for the finite-size-effect magnetization to be negligible compared to that induced by the field. These finite-size effects are the reason the lowest-H curve in Fig. 2(b) is not as smooth as those at higher fields. When our smallest-H value equal to $0.1\bar{J}$ is converted into gauss (assuming $T_c=5$ K), $H\approx1$ kG. However, as pointed out in Sec. III, it appears[5,17] that these estimates must be rescaled by about a factor of 10. For the purposes of comparing theory and experiment, the lowest fields we can consider ($H=0.1\bar{J}$) probably correspond to ~100 G. From both figures it is evident that as the field increases, the splitting of the FC and ZFC curves decreases. This is a direct reflection of

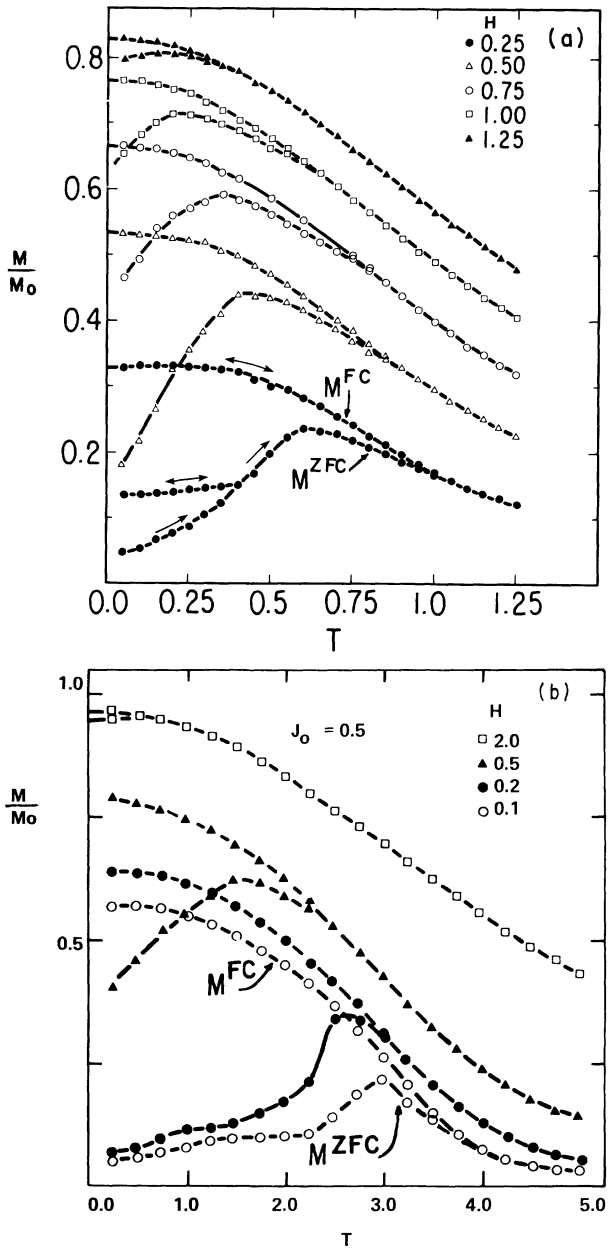

FIG. 2. (a) Temperature dependence of field-cooled (FC) and zero-field-cooled (ZFC) magnetization for $J_0=0$, $N=30^2$, $S=\frac{1}{2}$, and various magnetic fields H (in units of $\bar{J}$). Here $M_0=\frac{1}{2}$. (b) Temperature dependence of FC and ZFC magnetization for $J_0=0.5$, $N=10^3$, and $S=1$ for various magnetic fields H.

the fact that the number of minima on the free-energy surface becomes fewer and fewer as H gets very large. We have explored the effects of varying S and find them to be qualitatively insignificant. Results for the case $S=\frac{5}{2}$ are presented in Ref. 21.

Our theoretical curves may be compared with those measured experimentally, as shown in Fig. 3. The data are taken from Ref. 5. It should be noted that the experimental field values were not sufficient to appreciably shift $T_c(H)$; the tendency to merge the FC and ZFC curves at high H is, thus, not apparent in the data. The theoretical results are in qualitative agreement with the data; experi-

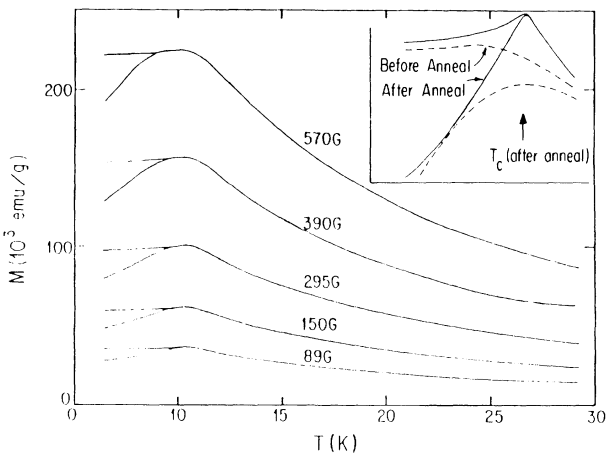

FIG. 3. Measured temperature-dependent FC and ZFC magnetizations for 2.6 at. % *Ag*Mn for various H (after Ref. 5). The inset shows the effects of annealing for 4.0 at. % *Cu*Mn (after Ref. 48).

mentally, it is found that the FC curve is reversible with respect to changes in T and that the ZFC is not. In fact, Chamberlin *et al.*[5] find a curve similar to that shown at the bottom in Fig. 2(a), for the effect of cooling after warming in a ZFC measurement. This small cooling curve is found to be reversible and nearly constant in temperature, as in our calculations. It should be noted that experimentally the temperature at which M^{ZFC} reaches its maximum value coincides with the temperature at which $M^{FC}=M^{ZFC}$. That this is not the case in our mean-field theory probably is related to finite-size effects, which tend to "blur out" the transition region. In fact, our numerical results are in strikingly good agreement with data taken[48] on nonannealed spin-glasses, which show irreversibility above T_c. It is presumed that this is due to inhomogeneity effects which give rise to a distribution of T_c's. This behavior, which is illustrated in the inset of Fig. 3, seems to mimic that which we find to be associated with finite-size effects.

VI. RESULTS FOR MAGNETIC HYSTERESIS CURVES

There is an abundance of data on magnetic hysteresis effects in spin-glasses.[6,49–51] Qualitatively, the results are of two types. The hysteresis loops[51] seen in (dilute) *Au*Fe are smooth and continuous as functions of H. By contrast, more concentrated *Au*Fe,[51] as well as *Cu*Mn and *Ag*Mn alloys show sharp magnetization reversals in magnetic hysteresis.[6,50] In Mn-containing spin-glasses, one also sees the so-called "displaced loops" obtained under certain circumstances in field-cooled measurements.[6,50] Such effects are not seen in *Au*Fe. Monte Carlo simulation results are rather scarce. We are only aware of one result by Binder and Kinzel[52] which yielded a characteristic (dilute) *Au*Fe-type loop for an Ising 80×80 system. In this section we discuss hysteresis loops obtained in a variety of different situations. We find the parameter to which the magnetic hysteresis is most sensitive is J_0, the center of the Gaussian. The larger the tendency toward ferromagnetism is, the sharper are the magnetization reversals. However, for Ising spins, displaced loops are nev-

er found to be present. This situation is to be contrasted with that obtained for Heisenberg spin-glasses,[22] with Dzyaloshinsky-Moriya[53] anisotropy; in this case (as will be discussed in the following paper) displaced loops will appear under the appropriate circumstances.

It should be emphasized that in our calculations magnetic hysteresis arises directly from the behavior of the free-energy surface. As H changes, a given minimum of the free energy becomes unstable and the system hops to a nearby state. The fact that the new minimum at $H+\Delta H$ has nearly the same magnetization as that at H leads to a very gradual, smooth hysteresis curve. This behavior is characteristic of Ising spin-glasses, with $J_0=0$. While we believe the shape of the hysteresis curve reflects primarily the character of the various metastable states in spin-glasses, it is clear that there are also time-dependent or relaxational effects which may play a role. Presumably, these are most clearly evident experimentally when the hysteresis loop is not well closed.

In Fig. 4 are plotted two hysteresis loops corresponding to $S=\frac{1}{2}$ and $\frac{5}{2}$, for an Ising system of $10\times10\times10$ spins. In these two cases, for convenience, the loops were obtained by slowly decreasing the field from the high-field limit ($H=2T_c$) to an equally large (in magnitude) negative field and then closing the loop. (Here $T_c=1.125\bar{J}$ and $13\bar{J}$ for $S=\frac{1}{2}$ and $\frac{5}{2}$, respectively.) At the highest fields the magnetization assumes a unique value, so that the resulting loop is independent of the way it was generated. This is not the case for smaller field sweeps. An example of the latter was shown in Fig. 2(a) of Ref. 20. As may be seen from Fig. 4, the effect of increasing S is to narrow the loop slightly and to position it more vertically around the origin. In Fig. 5 is plotted an experimentally measured hysteresis loop for *Au*Fe. The data, which are taken from Ref. 50, are in qualitative agreement with the Ising model results and with Monte Carlo simulations.[52] The similarity of results obtained in the two different theoretical approaches is not surprising since the calculations were performed at reasonably low T.

In more concentrated *Au*Fe as well as in *Cu*Mn, magnetic hysteresis loops are found[6,50] to exhibit sharp magnetization reversals. To obtain this behavior we studied the effect of increasing J_0 from 0.0 to 0.25 and finally to 0.50. The three corresponding loops are shown for the case $S=1$ in Fig. 6. As expected, when J_0 increases, the magnetization exhibits rather sharp jumps, with small

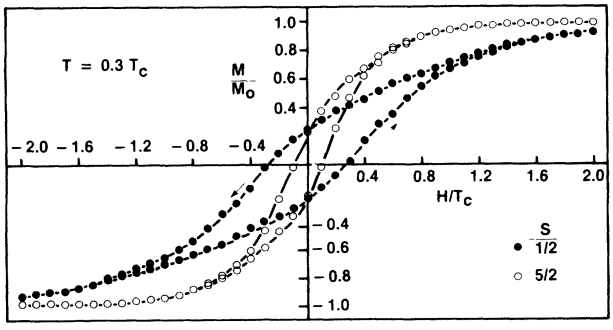

FIG. 4. Magnetic hysteresis curves, M vs H, for a symmetric field sweep for high maximum H for $T=0.3T_c$, $N=10^3$, and $J_0=0$ for two values of S.

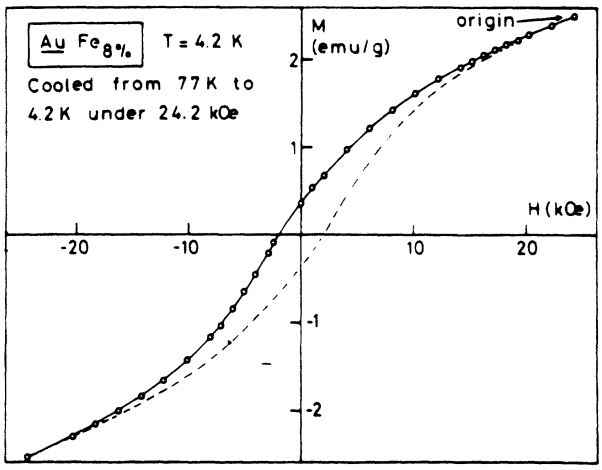

FIG. 5. Experimentally determined hysteresis for 8 at.% *Au*Fe which was field cooled in a field of 24.2 kOe (after Ref. 50).

changes in the field. This is due to the fact that many spins tend to flip in parallel when J_0 is large. This cooperative behavior is clearly evident from the data[51] shown in Fig. 7 for an $Au_{88}Fe_{12}$ spin-glass. This sharp, vertical loop should be contrasted with that observed for the more dilute *Au*Fe system shown in Fig. 5. Once J_0 exceeds the critical value for ferromagnetism the loop becomes almost rectangular (at low T). In the extreme limit of large J_0, the system is like an ordered Ising ferromagnet: There are exactly two minima on the free-energy surface, and the sharp reversals represent passing from one minimum to another.

While our numerical results provide insight into what is going on in *Au*Fe spin-glasses, the origin of the sharp magnetization reversals seen in *Cu*Mn is less clear. It has been argued[49,54,55] that dilute *Cu*Mn has a tendency towards ferromagnetism; the next-nearest-neighbor Mn-Mn RKKY interactions are believed to be ferromagnetic. Within our theoretical framework, we have found that only the presence of a ferromagnetic tendency can lead to cooperative magnetization reversals such as are found in *Cu*Mn. This will be discussed in more detail for anisotropic Heisenberg spin-glasses in the following paper. However, it is possible that in *Cu*Mn there are relaxation effects present (so that the system is not always "quasiequilibrated" at each value of H) and thus our theoretical picture may break down. In summary, the presence of cooperative behavior in a highly disorganized system, such as a spin-glass, appears difficult to understand unless there is a tendency towards ferromagnetism.

It has been observed that in *Cu*Mn, when the field sweep is nonsymmetric, the hysteresis loop is displaced[6,50] with respect to the $H=0$ axis. Similar results may also be obtained upon field cooling, under the appropriate conditions. Such "displaced" loops have evidently not been seen in *Au*Fe.[51] In order to ascertain whether they are present in our theoretical model for Ising spin-glasses, we have generated loops which start at large positive values and reverse direction at much smaller (in magnitude) negative values of the field. For the case $J_0=0$, the results were published in Fig. 2(b) of Ref. 20. There it was seen

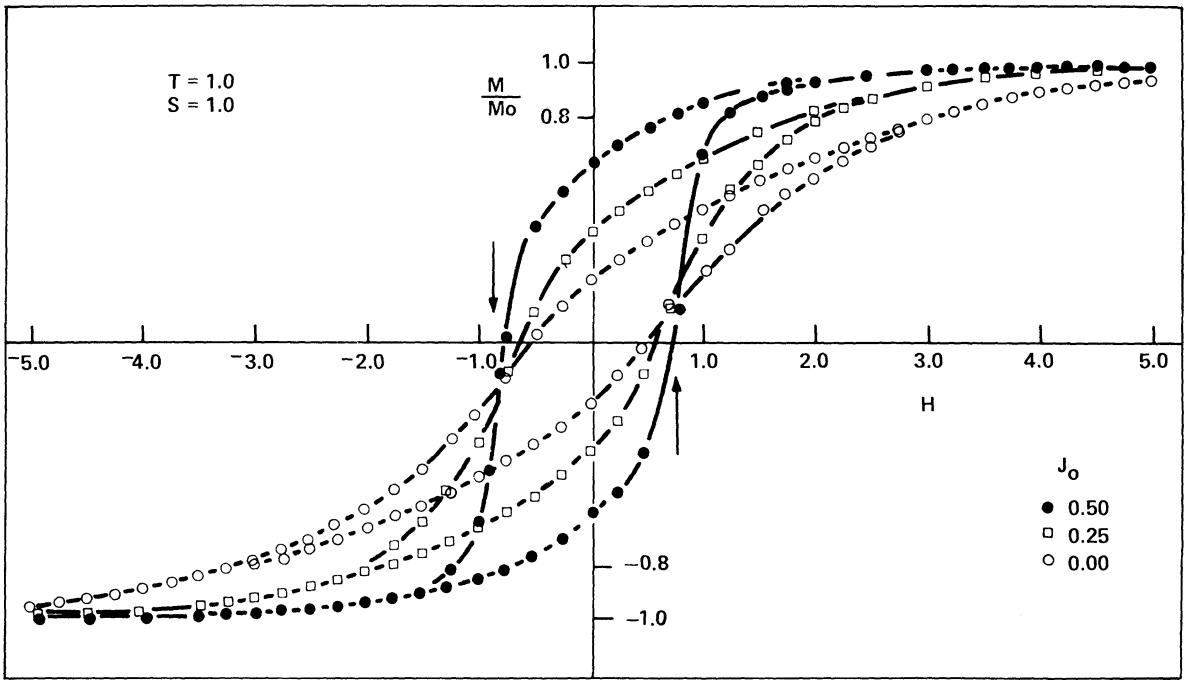

FIG. 6. Magnetic hysteresis curves, M vs H, for symmetric field sweeps for high maximum H at $T=1.0$ and $S=1.0$ for several values of J_0 and $N=10^3$.

that the hysteresis loop shifted primarily upward, i.e., with respect to the $M=0$ axis. The character of the hysteresis is analogous to what is sometimes called a "minor loop." In this paper we have studied the effects of increasing J_0, since this yields a more "boxlike" hysteresis loop. The result for the same $10\times10\times10$ system with $J_0=0.5$ and $S=1$, which was studied above, is shown in Fig. 8. While the loop is reasonably sharp, the effect of a nonsymmetric field sweep is similar to that found for $J_0=0$: The loop is raised relative to the $M=0$ axis. Hence we see no evidence for displaced loops in Ising spin-glasses, with or without a large J_0.

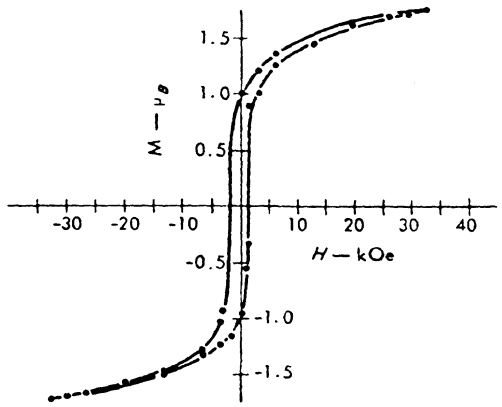

FIG. 7. Experimental hysteresis curve at 4.2 K in 12 at. % *Au*Fe (after Ref. 51).

VII. PROPERTIES OF THE MAGNETIC REMANENCE

When the field is turned off after creating a FC or ZFC state, one observes the so-called thermal remanent magnetization (TRM) or isothermal remanent magnetization (IRM), respectively.[1,2,6] Both the field and temperature dependences of these remanent magnetizations have been well studied. In this section we compare our theoretical values for TRM and IRM with those measured experimentally. Our physical picture is that the remanence arises because the spin-glass is "trapped" in a metastable state on the ($H=0$) free-energy surface which has finite magnetization. Only through very slow activation procedures can the spin-glass find its way over barriers to the "best" ($M=0$) free-energy minimum. This evidently takes place with a characteristic $\ln t$ dependence.[1,2] Presumably

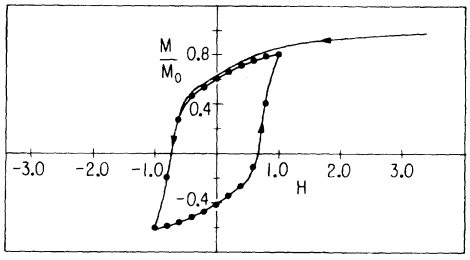

FIG. 8. Effects of nonsymmetric field sweep on hysteresis curve for $T=1$, $S=1$, $N=10^3$, and $J_0=0.5$. Note loop is displaced slightly upward.

the remanent ($M\neq 0$) metastable state is one which is close to the original FC or ZFC state created before the field is turned off. We calculate the values of the TRM (or IRM) by first creating FC (or ZFC) states and then slowly turning off the field, using small decrements ΔH. At a given value of H, we chose $\Delta H=0.05H$ or $0.10H$ for small and large values of the field, respectively. In this way the remanent state is chosen to be close to the original $H\neq 0$ state. It is believed that our results are appropriate for a computation of the remanence for times greater than those needed to obtain the "fast" relaxation which was discussed in Sec. III B.

The results for the calculated TRM and IRM as a function of field are shown in Fig. 9 for a 30×30, $S=\frac{1}{2}$ Ising spin-glass at low temperature $T=0.1T_c$. For comparison purposes we plot experimental data[1] obtained in *Au*Fe (Fig. 10). As can be seen, our calculated IRM has the characteristic "S shape" found in the data. However, the TRM has a somewhat weaker maximum than seen experimentally. (We have verified that there is, indeed, a maximum in our TRM.) Similar results were obtained in Monte Carlo simulations[12] on a 50×50 system. While the "noise" appears to be somewhat greater in the simulations, the maximum in the TRM is more pronounced, although there is also an unexpected weak maximum in IRM. It is not surprising that our results are of roughly the same quality as found in Monte Carlo simulations; at low T, our algorithm for defining metastable states coincides with that used in simulations. The manner in which the system is allowed to flow from one metastable state to another seems, however, to be clearly different in the two procedures.

The temperature dependence of the TRM is shown in Fig. 11 for the same parameters as in Fig. 9. Each point is obtained after cooling at constant field to the temperature in question and then turning the field off. At low H we find that the TRM decreases roughly linearly with increasing T. Because of finite-size effects, there is a small "tail" above T_c. The results of a series of measurements[2] on different *Au*Fe alloys is shown in Fig. 12. At those low fields, the TRM is found to be linear in T. However, at higher H, the remanence decreases more rapidly. This is not seen in our numerical results, which show a rounding at low T; note also that all theoretical curves appear to intersect at $T\approx 0.6T_c$. These results seem to be generally true for a range of different configurations of the $\{J_{ij}\}$. A low-temperature rounding of the TRM has been reported

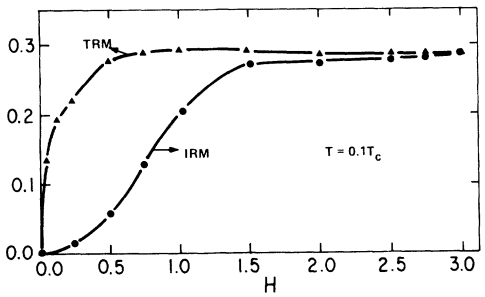

FIG. 9. Field dependence of TRM and IRM at $T=0.1T_c$ for $S=\frac{1}{2}$, $N=30^2$. Here $T_c=3.4\bar{J}$.

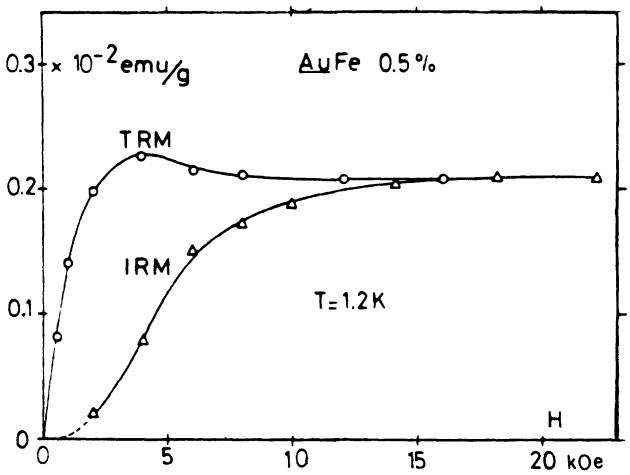

FIG. 10. Measured field dependence of TRM and IRM for 0.5 at. % *Au*Fe (after Ref. 1).

in Ref. 56 for alloys which are close to ferromagnetic. However, the fact that these measurements[56] have been made by a warming procedure may affect the shape of the TRM curve. The high-field results in Fig. 11 seem to represent the most serious discrepancy between mean-field theory and experiment that we have encountered. It is interesting to note that in Monte Carlo calculations Kinzel[12] also fails to see a more rapid decrease (than linear) in TRM with temperature, at high fields.

The temperature dependence of the "incremental" IRM ($=\Delta$IRM) is shown in Fig. 13 for the same parameters as in Figs. 9 and 11. The results are presented for three different values of H. To calculate the "incremental" IRM, we subtract the IRM from the values of the magnetization obtained just before the field was removed (i.e., the ZFC magnetization). Since both of these curves have maxima in their temperature dependences, it is not surprising that ΔIRM does also. While the errors are large (due to finite-size "noise" effects), the quantity ΔIRM peaks at $T\cong 0.6T_c$ for all three field values. This feature has also been observed by Guy,[2] who attributes some physical sig-

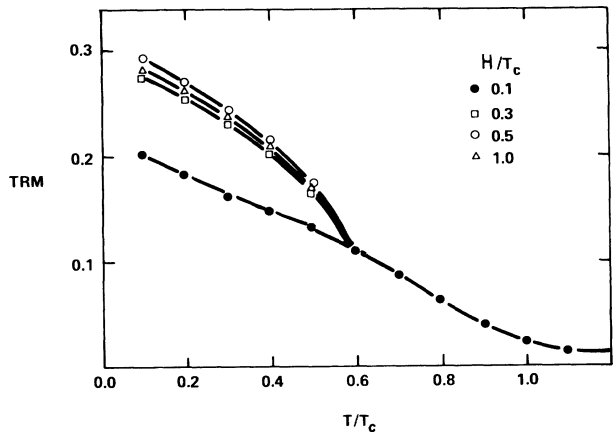

FIG. 11. Temperature dependence of TRM for various H for $S=\frac{1}{2}$, $N=30^2$.

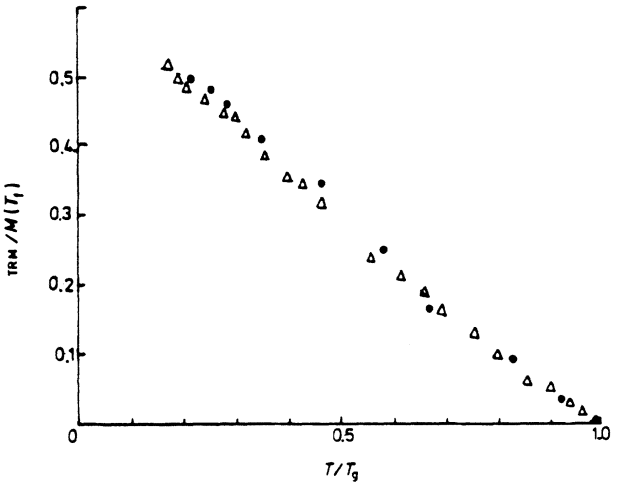

FIG. 12. Temperature dependence of TRM in Au–2 at. % Fe (●), Au–7 at. % Fe (△). Here $T_g \equiv T_c$ (after Ref. 2).

nificance to the temperature $T=0.6T_c$. It is difficult for us to assign any particular meaning to this temperature, although it is also associated with a feature in our TRM results.

VIII. PROPERTIES OF THE FIELD-DEPENDENT SPECIFIC HEAT

A series of very precise field-dependent specific-heat C_H measurements by Fogel and co-workers[38] was originally interpreted as suggesting that the Maxwell relation [Eq. (3.2)] was not satisfied in the FC configuration. A subsequent reanalysis of these measurements based on experimental[39,41,42] and theoretical[43] considerations led to the opposite conclusion: that Eq. (3.2) was valid for field-cooled measurements. These data also gave the first clear evidence that T_c played a role in thermal measurements. This is not surprising in view of Maxwell's relations, which indicate that structure in the T dependence of M^{FC} must be reflected in the field dependence of C_H. When C_H vs H was fit to a parabola, the coefficient of the quadratic term had a minimum at T_c.

Our numerical results for the T dependence of C_H for various H are plotted in Fig. 14 for a 100×100, spin-$\frac{1}{2}$ Is-

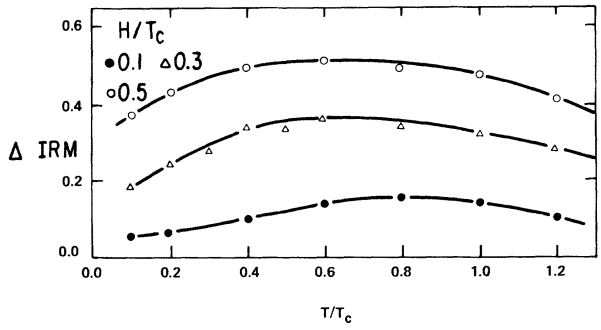

FIG. 13. Temperature dependence of incremental IRM (computed relative to magnetization before field removed). $S=\frac{1}{2}$, $N=30^2$ for three values of magnetic field H/T_c.

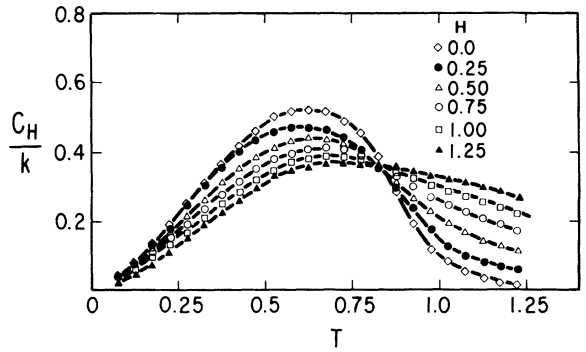

FIG. 14. Temperature-dependent specific heat for various H, in field-cooled configuration.

ing spin-glass. The numbers were obtained by taking small differences in the energy divided by temperature increments. The corresponding measurements of Brodale *et al.*[40] are plotted in Fig. 15. Both theory and experiment show that for $H>0$, $\partial C_H/\partial H$ changes sign at a temperature somewhat above T_c, corresponding to the temperature at which $M^{FC}(T)$ has a maximum negative slope. The quality of the agreement between theory and experiment, to some extent, reflects that found in Sec. V for the temperature and field dependence of M^{FC}. However, it should be pointed out that the theoretical curve for C_H at $H=0$ has a maximum at approximately T_c; by contrast, the experimental curve reaches a maximum significantly above T_c. This discrepancy, which is not related to Maxwell's relations, was also found[14,15] in early analytical theories. It is possible that when short-range-order effects are included (as for example with $J_0\neq0$) the theoretical maximum will increase to higher T, as was observed in previous "cluster" theories.[37] Monte Carlo results,[52] however, do seem to yield a maximum in C_H at $H=0$ above T_c, although the field dependence of C_H has not been explored.

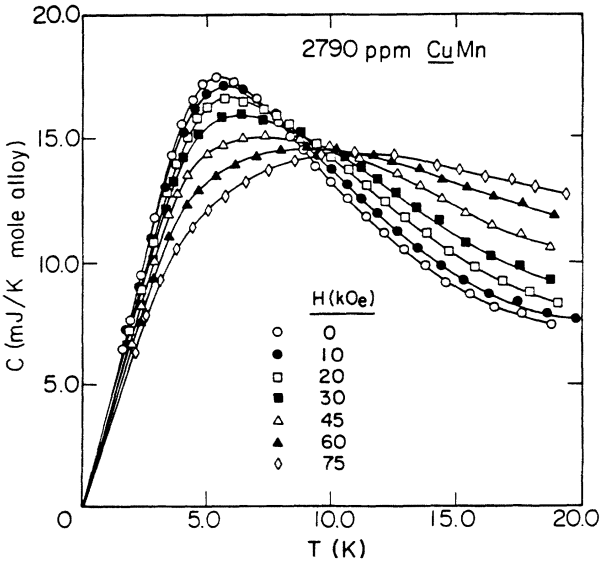

FIG. 15. Temperature-dependent specific heat for different field-cooled measurements (after Ref. 40) on *Cu*Mn.

As pointed out in Sec. III, our analysis indicates that in the FC configuration Maxwell's relations are clearly satisfied. This feature is (probably) unique to the field-cooled state; it is the only spin-glass state in which changes in H and T are produced reversibly.

IX. THE EFFECTIVE-FIELD DISTRIBUTION

The distribution of effective fields

$$H_i \equiv \sum_j J_{ij} m_j + H$$

may be readily calculated in all the states we considered. In Fig. 16 this distribution is plotted for a 100×100, $S = \frac{1}{2}$ spin-glass in zero external field. The three panels correspond to three different temperatures. As may be seen, at the lowest T (and presumably also at $T=0$) the distribution does not vanish at $H=0$. This is to be contrasted with the results obtained for infinite-range spin-glasses, which have $P(H) \propto H$ as $H \to 0$. Using Monte Carlo simulations, Binder and Kinzel[52] have obtained histograms similar to those in Fig. 16 for nearest-neighbor interactions.

In Fig. 17 are plotted the probability distributions for the same system as in Fig. 16 but with $H = 0.5 T_c$. Figures 17(a)—17(c) correspond to ZFC states at the indicated

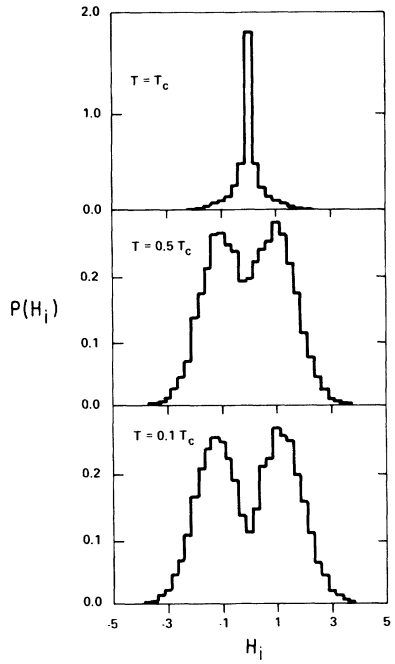

FIG. 16. Distribution of molecular fields for three temperatures for $S=1$, $N=10^4$, and no external field.

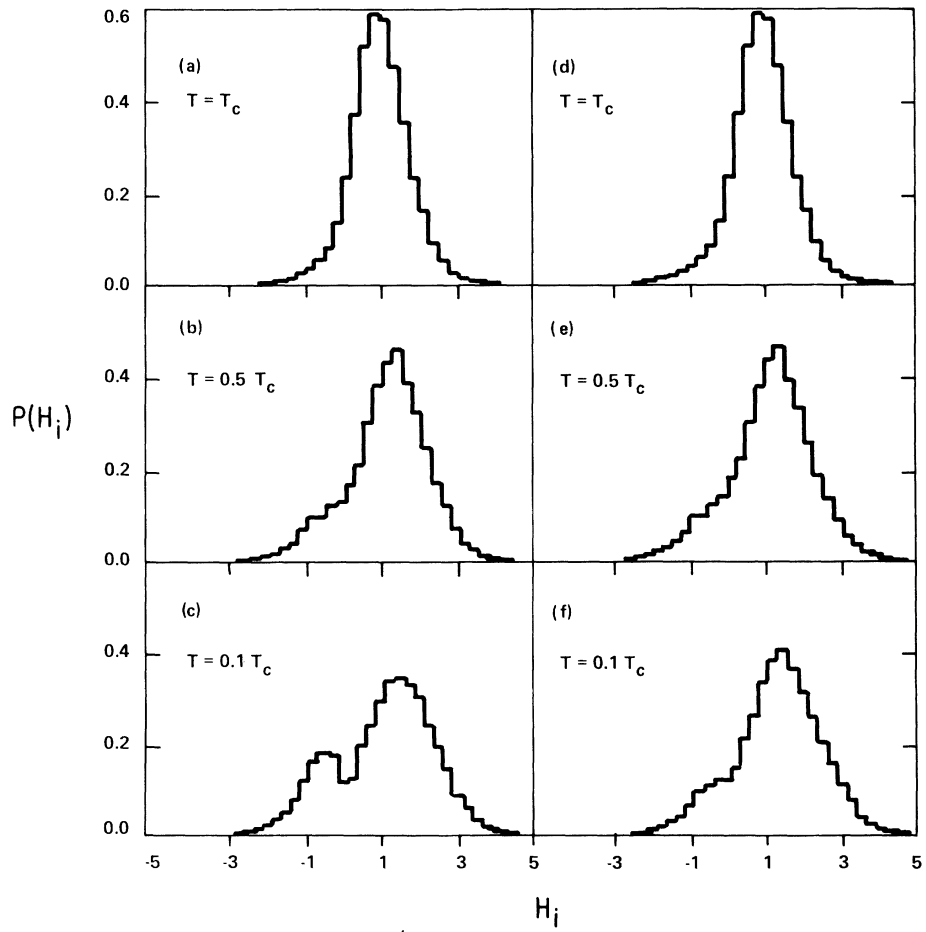

FIG. 17. Distribution of molecular fields for $S=1$, $N=10^4$, and $H=0.5T_c$. (a)—(c) for the ZFC and (d)—(f) for FC configurations.

temperatures and Figs. 17(d)–17(f) to FC states. The contrast between the two states is most apparent at $T=0.1T_c$. There is a "hole" at $H\sim 0$ present for the ZFC case but not for the FC state. Presumably this hole is a remnant of that obtained when no field was applied as in Fig. 16; the ZFC state is expected to be more similar to the $H=0$ state than is the FC one. In general, the effect of the external field is to shift $P(H_i)$ along the positive field direction. The effect of increasing temperature is to make $P(H_i)$ into a Lorentzian-shaped function. Our results for the ZFC case at $T=0.1T_c$ are similar to those obtained by Bantilan and Palmer,[57] for infinite-range spin-glasses in a magnetic field.

X. CONCLUSIONS

The most striking conclusion of this study is that the experimentally observed hysteresis, field-cooled and zero-field-cooled and isothermal and thermal remanent, magnetizations can all be reasonably well reproduced by a theoretical model which focuses only on the properties of the free-energy surface. Presumably, the reason for the success of this approach is that there are two important time scales in spin-glass experiments. "Fast times" (which may correspond[25] to $t<10^{-11}$ sec) during which the system makes small adjustments to find the nearest minimum of the free energy, and "long times" during which the system finds its way over energy barriers to more stable states. These latter processes appear[1,2] to vary as $\ln t$, suggestive of thermal activation. It is primarily because of these long-time processes that our quasistatic or quasiequilibrium viewpoint has some validity. The fast-time "dynamics" are automatically included in our calculations.

Our approach has features in common with that proposed by Tholence and Tournier,[1] following Néel's ideas on superparamagnetic particles. Nearby free-energy minima are found to differ by rotations of a few isolated clusters of spins. (This observation has also been made by Dasgupta *et al.*[13]) These clusters form the basis of the Néel picture. Their rotations are hindered by "anisotropy fields" or free-energy barriers, which in turn give rise to irreversible phenomena (e.g., hysteresis or remanence). However, a significant weakness of this Néel approach is that there is no sharp temperature onset for irreversibility. This is in contrast to both experiment and the present picture, in which the properties of the free-energy surface are found to vary dramatically with T.

Our viewpoint can be compared with recent dynamical[9–11] theories of spin-glasses. Such dynamical theories are essential in order to describe how the system evolves in time from a metastable (say, zero-field-cooled) state to the, presumably, equilibrium field-cooled state. However, the zero-field-cooled magnetization is only weakly time dependent, on a laboratory time scale. Thus we believe there are additional nondynamical effects which play an important role in determining, for example, the temperature dependence of M^{ZFC}. Indeed, it is very likely that the behavior of the free-energy surface is important. To see this, it should be recalled that the T dependence of M^{ZFC} is measured[5] in a warming procedure. If below $T_c(H)$ this process is interrupted and the spin-glass cooled back down, a different magnetization is obtained.[5] This irreversibility of M^{ZFC} (and the absence of irreversibility in M^{FC}) are readily found in our study of the free-energy surface. They, therefore, do not appear to be primarily dynamical effects. While dynamical theories often associate[58] the so-called nonequilibrium susceptibility $\chi^{\mathrm{ne}}(T)$ with the ZFC susceptibility, it is, perhaps, more appropriate to view $\chi^{\mathrm{ne}}(T)$ as the low-frequency ac response. Unlike M^{ZFC}, this susceptibility is reversible in T.[5]

Presumably the truth lies somewhere between the quasistatic or quasiequilibrium approach that we have proposed and the dynamical pictures of Hertz,[11] Sompolinsky,[9] and Shastry.[59] The extreme sensitivity of the free-energy surface to changes in temperature and field is well supported by our calculations. This appears to be one (of several) important ingredients needed in order to understand the spin-glasses, in all their complexity.

In addition to comparing our approach with previous dynamic theories, it can also be contrasted with standard numerical simulation techniques. Our primary thesis is that the evolution of the free-energy surface dominates that component of irreversible behavior which is weakly time dependent. Because we have reduced the complexity of the problem (on the basis of a physical model), our approach is not strictly equivalent to a Monte Carlo simulation. Furthermore, because of the reduction in complexity we are able to handle larger systems more rapidly; this also makes it possible, for the first time, to address Heisenberg spin-glasses in considerable detail, as will be seen in the following paper.

The approach taken here should be assessed at two levels; it should be evaluated according to the validity of the physical picture which we have proposed (and others[5,24–27] have sketched in less detail) as well as that of the numerical scheme used to test the model. We regard the model as an appealing way of understanding irreversible behavior in spin-glasses and the numerical scheme, although approximate, appears to effectively illustrate and implement this physical picture.

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