Adiabatic demagnetization of antiferromagnetic systems: A computer simulation of a planar-rotor model

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We report a computer simulation of the adiabatic demagnetization of a two-dimensional antiferromagnetic (AFM) model system, based on the molecular-dynamics method, and motivated by recent studies in the nuclear magnetism of copper. As opposed to Monte Carlo computations, we are able to calculate directly the isentropes $B(T)$ with perfect control of adiabaticity, having macroscopic reversibility available as a signature. We then find a distinct minimum temperature at a nonzero external field in the vicinity of the phase-transition line separating the antiferromagnetically ordered phase from the high-temperature region. This is compatible with both earlier Monte Carlo calculations of Lindgard, Viertiö, and Mouritsen [Phys. Rev. B **38**, 6798 (1988)] and a mean-field-approximation analysis also presented in this paper. We propose that the shape of the isentropes is a feature of simple AFM systems with next-neighbor interactions.

I. INTRODUCTION

For about the past decade, there has been a continuous period of interest in nuclear magnetism. In 1982 Huiku and Loponen[1] reported a phase transition in the spin system of copper at nanokelvin temperatures. The experimental methods have been improved a lot since, and different methods of investigation have been employed (e.g., recent neutron-diffraction studies of nuclear ordering). Yet a sound theoretical understanding of the ordering process in antiferromagnetic systems subject to an external magnetic field must still be developed. As is often the case, one resorts to MFA analysis and computer simulations to get first ideas of how nature actually works.

Our interest in the demagnetization of antiferromagnets is based on this background. However, we find it is appealing by itself to show the counterintuitive features of the isentropes in such systems.

II. OUR MODEL

Interestingly, the Hamiltonian of a nuclear spin system like copper is quite well known.[2] Disregarding all the complications of the spin Hamiltonian of copper our model is classical, contains no contribution of long-range interactions, and particularly no dipolar forces: We study a two-dimensional array of 32×32 classical rotors at fixed positions, each possessing one rotational degree of freedom with next-neighbor interaction. Our Hamiltonian is

$$H=-J\sum_{\langle i,j\rangle}\cos\alpha_i\cos\alpha_j-B\sum_i\cos\alpha_i \qquad (1)$$
$$J<0,\quad B>0\ ,$$

with an Ising-like anisotropic interaction, well suited for a continuous multistep algorithm in a molecular-dynamics calculation. A similar model has been studied by one of us[3] earlier and in a different context. As discussed there, we add a small ferromagnetic coupling $-K\sum_{\langle i,j\rangle}\sin\alpha_i\sin\alpha_j$, with $K>0$, $|K|\ll|J|$, which at low T prevents the separation of the system into independent oscillators and thus facilitates transport through the lattice. We leave aside a further discussion of this perturbation, because its only measurable effect is the improvement of the data in regions where the decoupling of rotors would effectively inhibit equilibration.

To avoid misunderstanding, our model is not from the very start meant to model true nuclear magnetism, but is a vehicle to show the qualitative behavior of antiferromagnetic isentropes, which we assume to be independent of the particular Hamiltonian. (In the same sense as the plateau and cusplike behavior of the perpendicular and parallel antiferromagnetic susceptibility, respectively, is discussed as a universal feature of such systems.)

In the following paragraphs we first report the results of a MFA analysis of our model system, thereby heuristically discussing the expected behavior of antiferromagnetic isentropes. Then we give a short overview on the simulation techniques which we employed and finally we present the results of the computer simulation.

III. MOLECULAR FIELD ANALYSIS

From the MFA Hamiltonian $H^{\pm}=-(zJm^{\mp}+B)\cos\alpha_{\pm}$ (where the index denotes two sublattices with respective magnetizations $m^{\pm}\equiv\langle\cos\alpha_{\pm}\rangle$ and z is the coordination number) one has the coupled self-consistency equations

$$m^{\pm}=(1/Z^{\pm})\mathrm{Tr}\{\cos\alpha_{\pm}\exp[\beta(zJm^{\mp}+B)\cos\alpha_{\pm}]\}$$

with (2)

$$Z^{\pm}=\mathrm{Tr}\{\exp[\beta(zJm^{\mp}+B)\cos\alpha_{\pm}]\}$$

which are easily stated in terms of modified Bessel functions of zeroth and first order.[4]

$$m^{\pm}=(1/Z^{\pm})2\pi I_1(zJ\beta m^{\mp}+\beta B)\ ,$$
$$Z^{\pm}=2\pi I_0(zJ\beta m^{\mp}+\beta B)\ . \tag{3}$$

There is no difficulty in obtaining numerically exact solutions of these MFA equations and to extract the isentropes $B(T)$.

A consideration of the ground-state energy at zero temperature reveals that there is indeed such a simple two-lattice antiferromagnetic structure in the ordered phase, as one expects. The staggered magnetization is therefore a suitable order parameter.

As our system makes no distinction between antiferromagnetic (AFM) and ferromagnetic (FM) coupling as long as there is no external field applied, both FM and AFM isentropes will have the same value $T(B=0)$. At temperatures high enough for the interactions to become negligible, they again will converge to a common limit.

In low but finite fields AFM systems will tend to stay in the ordered zero-field phase. A higher entropy and more important disorder is expected under the external force which is in competition with the interaction. So one recognizes that AFM isentropes will lead to lower temperatures in moderate fields.

Interestingly, there is even an heuristic argument why the minimum temperature should be assumed at finite fields, well in the paramagnetic region: If the field is high enough to polarize both sublattices in the same direction the interaction energy will take a maximum value. Lowering the field will set free some potential energy and so, in a thermally isolated system, will raise the temperature.

Both considerations together show, at least in the case of next-neighbor coupling, that AFM isentropes will allow lower temperatures to be reached at nonzero fields. This will prove to come true for our MFA evaluation as well as the simulations.

Some results of MFA computations for the model (1) as well as for the antiferromagnetic planar Ising system are shown in Figs. 1 and 2. The most striking feature is the distinct minimum temperature along isentropes of low enough entropy, in the vicinity of the phase separation line.

Remarkably, this phenomenon is even amplified in the simple antiferromagnetic Ising system where, for entropies lower than $S=0.5S_{\max}$, the temperature is lowered down to $T=0$(!) at the critical field $B_c/|zJ|=1$. For all those isentropes passing through the point $(B,T)=(|zJ|,0)$ a first-order transition occurs from an AFM phase to a pseudo-spin-flop phase in which both sublattice magnetizations have the same direction but different magnitude. At this transition the parallel sublattice magnetization takes its maximum value (complete alignment) while for the other lattice the sign of magnetization is reversed. For the other isentropes both phases can still be distinguished, but there is no more a phase transition. These pathological features disappear in our model (1). There is only one ordered phase (AFM) and zero temperature is approached only in the limit of low entropy.

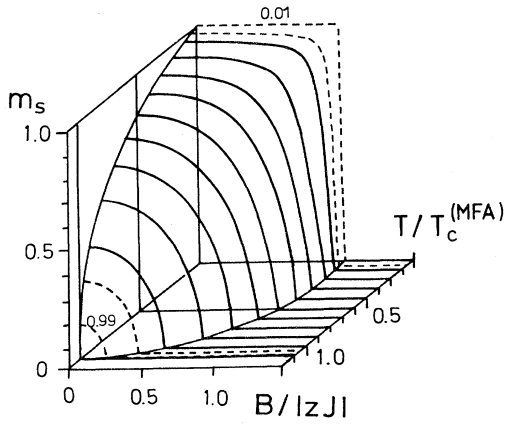

FIG. 1. Order parameter (staggered magnetization m_S) of the model under study in mean field approximation (MFA).

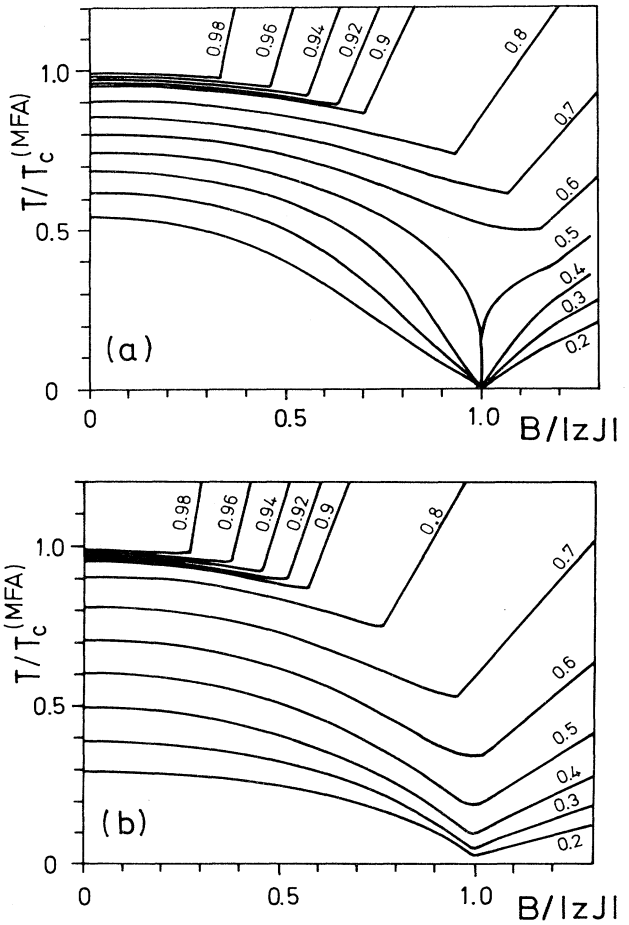

FIG. 2. Mean field (MFA) isentropes $B(T)$ of the antiferromagnetic Ising system (a) and the model under study (b). The parameters at the different curves denote the entropy per particle, measured in units of the respective maximum entropy ln2 (a) and ln2π (b). Despite some differences there is an obvious correspondence in the shape of the isentropes in both, the Ising system and our model (1).

IV. COMPUTER SIMULATION

First of all a survey of the methods will be given and it will be shown how one can use a simple calculation of the rotor dynamics for extraction of thermodynamical quantities.

Until now we have only specified an interaction to define the AFM model (1). For the purpose of molecular dynamics we have to add a prescription for the dynamics of the system. As usual this is done by adding a kinetic-energy term to the interaction Hamiltonian. In our case it is the rotational kinetic energy of classical rotors with a fixed center of mass and one angular degree of freedom. We will refer to the model system governed by this modified Hamiltonian as the "rotor model." It should be emphasized at this point that we introduce the rotor model for dynamical reasons only while we are finally interested in the magnetic model (1). Consequently we have to extract observables for the rotor model from simulation data and then to transform these into the equivalent observables of model (1). Obviously this is a problem for such observables which are based on the momentum distribution, as, e.g., temperature and entropy particularly. We will address this topic in some detail below.

To find the observables of the rotor model we study the classical motion of the system. In fact, we numerically solve Newton's equation of motion of such a classical rotor system, using a simple multistep algorithm of Beeman[5] which is correct to third order in the time step and—most importantly—stable for our model system, even over a range of several hundreds of thousand steps.

The interesting thermodynamical quantities are gathered by time-averaging certain mechanical quantities of the system. It is well established that some purely thermodynamical variables can thus be related to mechanics by an asymptotic equivalence or—more problematically—by application of fluctuation-dissipation theorems. Examples are the magnetic susceptibility and temperature. Specifically, from the classical equipartition theorem it follows that the mean kinetic energy is a measure for the (canonical) temperature which in turn is equivalent to the microcanonical temperature in the macroscopic limit.

There exists, however, no unique direct method of finding the entropy for a system under simulation. Yet in our case the system degenerates into a system of independent particles in the high-temperature limit and into a harmonic system in both the high-field and the low-temperature limit. One knows the classical entropy of these systems; so if we are able to adiabatically take the system into one of these regions, we know the entropy. One might argue that in the limiting case of known entropy the system is not suited for computer simulation due to the lack of equilibration processes. This is a serious objection, but our computations show that we can maintain macroscopic reversibility up to fields high enough to show the expected high temperature behavior.

Now we will show in the following argument that for large systems ($N=1024$ in our case) $T=2K/N+O(1/N^2)$ is an approximation to the microcanonical temperature of the magnetic model system (setting $k_B=1$). Let $\Omega(E,K)=\Omega_K(K)\Omega_\Phi(E-K)$ be the number of states available to the system for a fixed kinetic energy K and total energy $E=K+\Phi$. $\Omega_K(K)$ is the number of available states in momentum space at fixed kinetic energy [$\Omega_\Phi(E-K)$ in configuration space at fixed $\Phi=E-K$], respectively. If $\Omega_\Phi(E-K)$ is a monotonically increasing function of its argument (which needs not be true if the system undergoes a first-order phase transition) then $\Omega(E,K)$ has a single sharp maximum at $K=K^*(E)$. There the partial derivative of $\ln\Omega(E,K)$ with respect to K vanishes and consequently $\beta_K(K^*)=\beta_\Phi(E-K^*)$ where $\beta_\Phi=(d/d\Phi)\ln\Omega_\Phi(\Phi)$ (and similarly β_K) is the/p microcanonical definition of inverse temperature. $\Omega_K(K)=\text{const}\times K^{(N-1)/2}$ is known so that $\beta_\Phi=\beta_K=(N-1)/2K$.

Next we consider the determination of the entropy for model (1). In a large enough system the entropy is determined from the number of states at the mean value of the kinetic (potential) energy: one simply writes

$$S_\Phi(E-K^*)=S(E)-S_K(K^*)\,, \tag{4}$$

where S is the observed entropy of the rotor system and S_Φ is the entropy in configuration space (S_K in momentum space). Since $S_K(K)$ is known, as well as $S(E)$ for a system of harmonic oscillators, we have

$$\frac{S_\Phi(\Phi)}{N}=\frac{(2N-1)}{2N}\ln\frac{(E-E_0)}{N} -\ln\omega-\frac{(N-1)}{2}\ln\frac{2K}{N}+C(N)\,, \tag{5}$$

where ω is the frequency of the system of identical classical oscillators with ground-state energy E_0 toward which the model system degenerates in the high field limit; particularly $\omega=(zJ+B)^{1/2}$. $C(N)$ is a term which depends only on N and vanishes as $(1/N)\ln N$. For constant and not too small N this term may be neglected and Eq. (5) simplifies to

$$\frac{S_\Phi(\Phi)}{N}=\ln\frac{(E-E_0)}{N}-\ln\omega-\tfrac{1}{2}\ln\frac{2K}{N}\,. \tag{6}$$

Equation (4), however, is known to be an exact equation only in an infinite system. For a finite system one generally has to add a "defect" term $D(E,N)$ to the right-hand side of (4) which represents the contribution of states with kinetic energy different from the mean value. The defect D is generally assumed to converge to zero as the number of particles approaches infinity as constant E/N. Yet in our case (harmonic system) we can even calculate D exactly. It is of order $O((1/N)\ln(E/N))$ and negligible against the terms in (6). This justifies the use of (6) for entropy determination in the finite system with $N=1024$.

All simulations have been performed for the same Hamiltonian. The moment of inertia of all rotors is chosen to be 1. The interaction constant J is set to be -0.5 leading to a MFA critical temperature $T_c=z|J|/2=1$ and a critical field $B_c(T=0)=|zJ|=2$. Our goal was to generate isentropes in and near to the or-

dered phase region. In zero external field we know $T_c \approx 0.68$ and we have an estimate of the range of total energies which yield isentropes in the interesting region when an external field is adiabatically imposed. The time step was $dt = 0.05$ and a typical measurement took 100 000 steps after 10 000 to 30 000 steps of equilibration.

Having generated starting configurations of different entropies in zero field— by shifting the total energy step by step in a series of runs at constant energy—the system is guided along an isentrope by gently sweeping the magnetic field upward between two measurements. The sweep speed must be balanced between total simulation time cost and the goal of reversibility conservation. We found a value of 1 000 000 steps per unit of magnetic field to fit these needs.

The basic technique to assert adiabaticity is to prove long-term reversibility. This is established by a reverse sweep, back over the full range of up-sweeping. The observables are found to be reversible within (approximately) 1% precision, except for very low entropies. There we observe irreversibility, but only in the ordered phase for low temperatures.

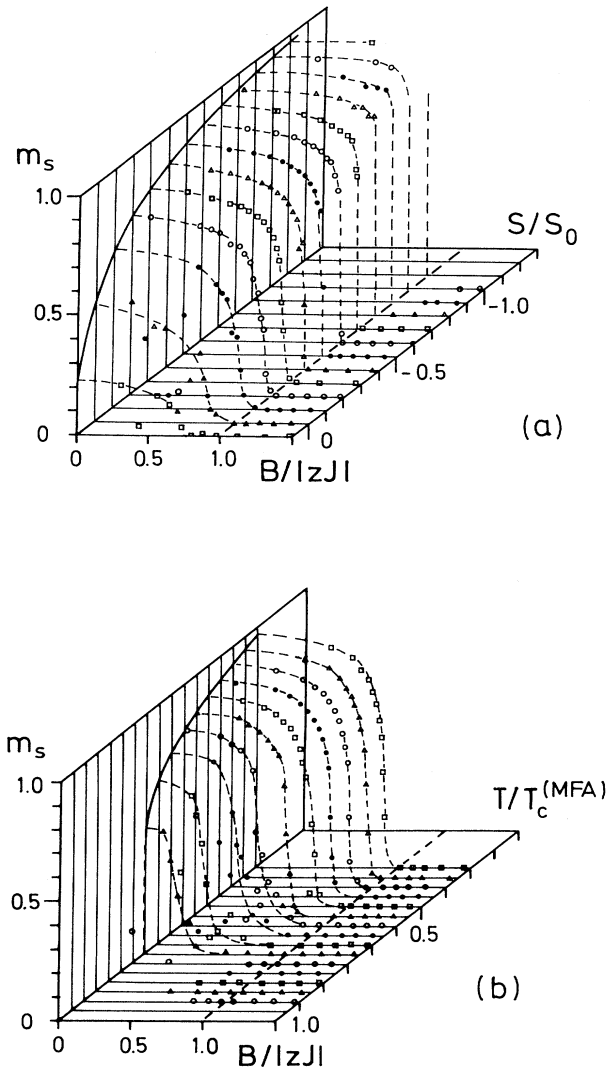

FIG. 3. The order parameter of the model under study on both, the (B,S) plane (a) and the (B,T) plane (b). The values of entropy S/S_0 shown in (a) are computed from formula (6), where S_0 is the corresponding dimensionless unit entropy (note $k_B = 1$). This (B,S) plot is more reliable due to the fact that entropy is a well-controlled parameter of our simulation. On the contrary, temperature is determined from a fluctuating quantity and obviously suffers from finite-size effects (b).

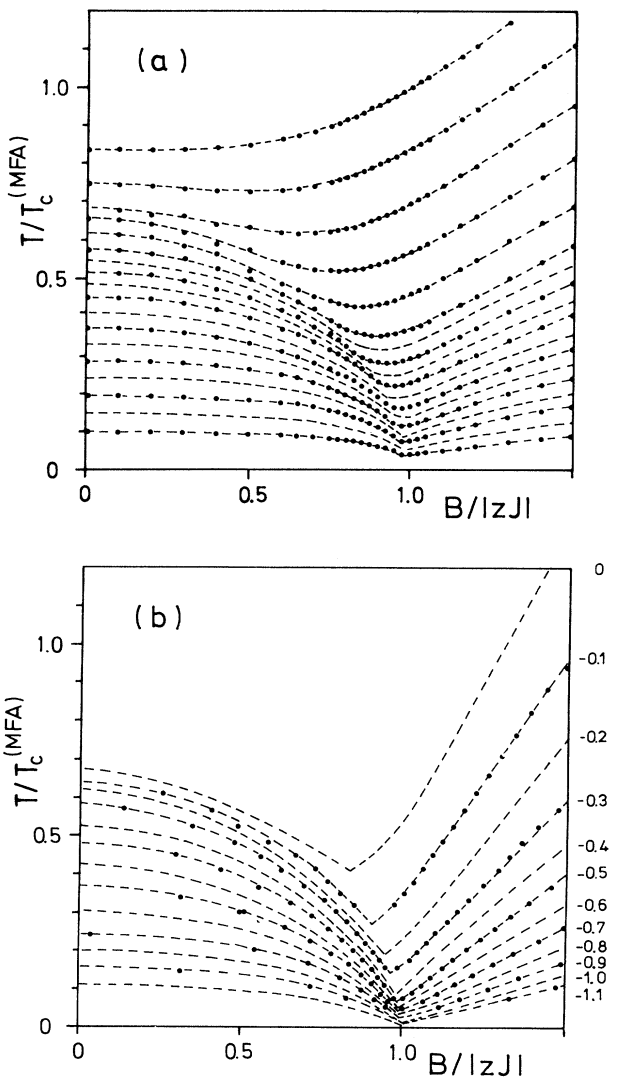

FIG. 4. Isentropes $B(T)$ of the model actually simulated (a) demonstrate the quality of original data. The lower plot (b) shows isentropes of the model (1). Only such data points have been drawn which directly result from the interpolation being used to extract the entropy for model (1) from simulation data. The shape of the isentropes is in fact much better determined than the relatively few data points may indicate. The curve parameters are entropy values in the same units as in Fig. 3.

All simulations have been executed on the AMT-DAP parallel computer in Erlangen, which is perfectly suited for our purpose. It is using a 32×32 array of processors with hardware support for data transport between neighbors and for toroidal boundary conditions, both of which have been heavily used.

In Figs. 3–5 we present some results of simulation. Figure 3 shows the order parameter of the model system. The very steep descent in (B,S) plot of the order parameter could naively be interpreted as a signature for a first-order phase transition. However, based solely on our data for a single lattice size this proposition cannot be proven.

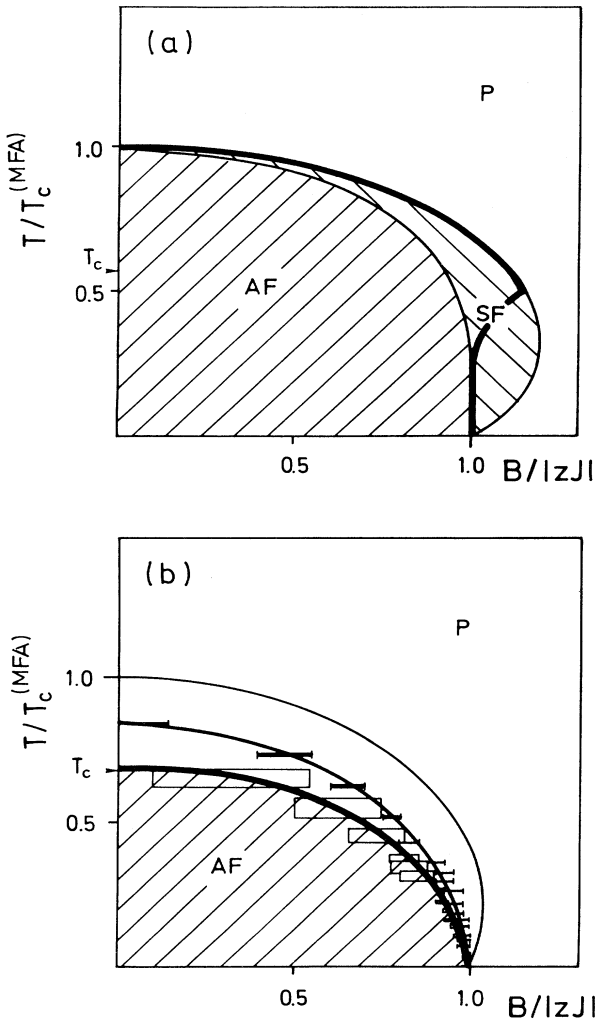

FIG. 5. Phase diagram of the antiferromagnetic Ising system (a) (as calculated by mean field theory) and of model under study (b) (MFA as well as simulation results). For the Ising system (a) the bold line shows the line of minimum values of temperature along the isentropes which is partially identical to the phase boundary between the paramagnetic phase (P) and the ordered phase SF/AF. (There is no phase transition between AF and SF.) For our model system (b) the bold line marks the phase boundary between AF and P. The line of minimum values of temperature is shown between the true one and the MFA phase boundary.

It should be noted that the points shown are very "diluted" when compared to the original simulation data (not shown) for the rotor model. Reducing the data to the entropy of the system of interest a two-dimensional set of data points may be obtained. Only the points along the original isentropes have been recuperated and are shown in the figure.

In Fig. 4 the isentropes are depicted. Again, the most striking result is the shape of the isentropes: the characteristic feature is a distinct minimum value of the temperature at a finite field and approximate linearity for high fields.

Qualitatively the shape of isentropes is found to be quite well reproduced by the MFA. Even numerically, the correspondence between MFA and simulation data is good (entropy differences being regarded), particularly in the low temperature region. But this point should not be overestimated, as we assume the precision of our entropy determination not to be better than a few percent.

In Fig. 5 the phase diagrams are represented. To construct a phase diagram the transition point has been located within intervals outside of which the order parameter is definitely zero (nonzero, respectively); furthermore the well-defined maxima of magnetic susceptibility (not shown) are found within these intervals. Despite the relatively large error bars, a knowledge of the zero-field transition temperature allows us to determine the separation line with good precision. As one is expecting, MFA yields too extended an area of the ordered phase in the B-T diagram.

The line of minimum temperatures along each isentrope is located between the MFA phase separation line and the actual phase boundary, that is well in the paramagnetic region. In MFA of both, Ising system and the model under study (but not shown for the latter), it is partially inside the ordered region and partially at the transition line.

V. CONCLUSIONS

The method of molecular dynamics computations has certainly proven to be successfully applicable. But we feel that the computer time needed to perform our simulations is close to the limit which is acceptable. This big amount of computer time required is due to the intrinsic necessity to simulate a continuous change of state, when one wants to directly find the isentropes in a model system. The time needed to sweep to the next point of measurement actually triples the simulation time.

Our investigation obviously lacks a finite-size analysis as well as a true quantitative evaluation of simulation data. It has not been our goal to obtain accurate information about the nonanalytical behavior at the phase transition (critical exponents). Therefore we have not attempted a finite-size analysis, in particular on the background of simulation times needed. This is also the main reason why we avoid statements on the nature of the observed phase transition, which one would naively classify as first order like, due to the very steep descent of order

parameter. Based on other work by us,[6] we have learned to be very cautious in establishing the nature of a phase transition in a finite system. A serious discussion of this topic is definitely impossible on the background of presented results.

We find and thereby confirm the observations of Lindgard *et al.*[7] that isentropes in antiferromagnetic systems tend to have a distinct minimum temperature if entropy is low enough in order that the phase separation line is passed. This statement is supported by a molecular field consideration. The minimum temperature is reached in the paramagnetic region and the temperature attained may be much lower than the zero-field limit.

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