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Monte Carlo simulation of fcc Heisenberg antiferromagnet with nearest- and next-nearest-neighbor interactions

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We report the results of Monte Carlo simulation of the ordering phenomena and phase transition in a classical Heisenberg fcc spin system ($S = \infty$) with antiferromagnetic nearest- and next-nearest-neighbor interactions ($J_{NNN} = 0.1\,J_{NN}$). At low temperatures the system exhibits Type III collinear antiferromagnetic order. At $T = 0.47 J_{NN} k_B$ there is a transition to disordered phase. Unlike the second-order transition in a "frustrated" fcc antiferromagnet (only $J_{NN} \neq 0$), the transition in the system studied is of the first order, as indicated by discontinuities in system energy and relaxation time versus temperature. The system we investigated may be considered as a classical analog of the Type III antiferromagnet β-MnS. We compare the calculated results with the experimental data obtained for this compound.

I. INTRODUCTION

The best known Type III fcc antiferromagnet[1] is the zinc-blende structured modification β-MnS. Type III order was detected in this system almost three decades ago[2] by neutron diffraction measurements on a powder sample. Later, it was pointed out that the structure might also be a complex noncollinear spin alignment[3] but the existing neutron data did not allow to resolve this dilemma. A renewed interest in β-MnS has been evoked by the discussion on the nature of phase transition in Type III systems. Considerations based on group-theoretical methods and renormalization group techniques led to the conclusion that the transition is of the first order.[4] This result was confirmed by another neutron experiment.[5] Still, the discussion has not resolved the problem of the structure, which likely will remain unanswered until single crystals of β-MnS become available.

Current interest in the magnetic properties of systems known as the diluted magnetic semiconductors[6] (DMS) points to additional reasons for concern about β-MnS data. Several compounds of the DMS group can be considered as diluted zinc-blende structured Mn chalcogenides (e.g., $Cd_{1-x}Mn_xTe$, $Zn_{1-x}Mn_xTe$ for $x \leqslant 0.70$, $Cd_{1-x}Mn_xSe$ for $x \leqslant 0.50$, or $Zn_{1-x}Mn_xS$ for $x \leqslant 0.10$; see Refs. 7 and 8). From the point of view of DMS, β-MnS is therefore of particular interest, being the only existing system of this kind with $x = 1$.

Model studies on fcc antiferromagnets performed in relation with DMS utilized mostly computer simulation techniques.[9-11] The cases investigated were the simplest analogs of DMS—Ising or Heisenberg "frustrated" fcc lattices (i.e., with nearest-neighbor interactions only). These results can not apply to β-MnS, as Type III order requires nonzero next-nearest-neighbor interactions ($0 < J_{NNN} < 0.5 J_{NN}$, according to Ref. 1). However, there is a growing amount of evidence that J_{NNN} should also be taken into consideration for DMS in order to achieve a quantitative agreement between the calculated and measured data.[12-15] For $x = 1$ such a model becomes at the same time an analog of β-MnS.

We initiated a series of Monte Carlo studies of antiferromagnetic Heisenberg fcc systems with nearest- and next-nearest-neighbor interactions. Some of the results obtained for a diluted system, with $x = 0.7$, have been previously reported[16] and compared with the experimental data on $Cd_{0.3}Mn_{0.7}Te$. In this paper we show that the results obtained for $x = 1$ give a reasonable description of some of the magnetic properties of β-MnS.

II. METHOD OF CALCULATION

The sample investigated was a fcc supercell of $N = 4 \times 10^3$ sites with periodic boundary conditions. The sites were occupied by unit vectors $\mathbf{S}_n$, representing classical spins. The energy of interaction of the nth spin with its neighbors was calculated using the formula

$$E_n = J_{NN} \sum_{i=NN} \mathbf{S}_n \cdot \mathbf{S}_i + J_{NNN} \sum_{j=NNN} \mathbf{S}_n \cdot \mathbf{S}_j, \qquad (1)$$

where the sums are over the nearest and next-nearest neighbors, and J_{NN}, J_{NNN} are the corresponding exchange constants (in all calculations, $J_{NNN} = 0.1 J_{NN}$ was taken). The computer program utilized the Monte Carlo algorithm developed by Metropolis *et al.*[17,18]

One of the objectives of our work was to compare the properties of the system with $J_{NN}, J_{NNN} \neq 0$ with the results obtained previously by Fernandez *et al.*[10] for a Heisenberg system with only $J_{NN} \neq 0$. Similarly as in this work, we studied for various temperatures the time-dependent Edwards-Anderson[19] order parameter $q(t)$ and the relaxation time τ using the formulae

$$q(t) = N^{-1} \sum_n \langle \mathbf{S}_n(0) \cdot \mathbf{S}_n(t) \rangle \qquad (2)$$

and

$$\tau = \int_0^\infty q(t)\,dt \,/ q(t=0), \qquad (3)$$

where time is expressed in Monte Carlo steps/spin.

In addition to that, we calculated the magnetic susceptibility versus temperature, using the method based on fluctuations in magnetization.[18] We applied also various methods of analyzing the spin order in the sample (which will be described in detail elsewhere).

The calculations were started from initial states which were either random spin configurations, or ground-state

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configurations (corresponding to $T=\infty$ and $T=0$, respectively). For a given temperature the program was run until a well-established thermal equilibrium state was reached, and then the calculations proceeded for a time necessary for obtaining sufficiently accurate averages of the calculated quantities (no less than 5000 MCS/spin were used for each temperature, and up to 20 000 in the vicinity of the transition point). In order to study the reversibility of the processes in the sample we carried out a series of calculations corresponding to "heating" and "cooling," in which the equilibrium state generated for one temperature was subsequently used as the initial state for the next temperature.

III. RESULTS

At low temperatures ($\Upsilon = k_B T/J_{NN} \leqslant 0.47$) the spins in the sample formed long-range collinear Type III antiferromagnetic order. It was of some surprise that the collinearity occured in all cases, and did not depend on the initial configuration. A spin system with antiferromagnetic J_{NN} and J_{NNN}, satisfying $0 < J_{NNN} < 0.5 J_{NN}$, possesses a degenerate ground state. In particular, the ground state may be a collinear Type III order, associated with a single wave vector of the type $\mathbf{k} = (2\pi/a)(1, \frac{1}{2}, 0)$. In general, the energy minimization requirement may be also satisfied by a noncollinear spin alignment associated with three inequivalent such vectors.[5] However, this latter configuration turned out to be unstable for $T > 0$. In runs starting from noncollinear ground states the system reached in the beginning a quasi-equilibrium state; usually, after several thousand MCS/spin a "domination" of one of the wavevectors became visible, and after $\simeq 10^4$ more MCS/spin the order components corresponding to the other two vectors practically disappeared.

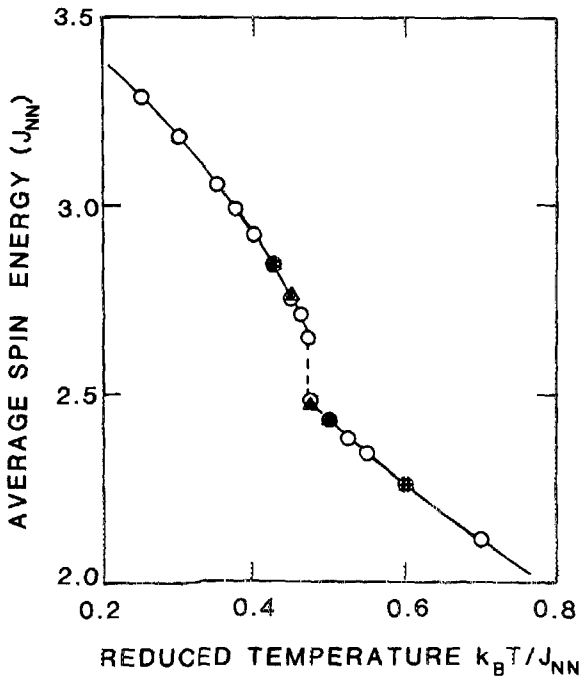

FIG. 1. Average energy per spin (absolute value—in fact, the energy is negative) vs temperature. The circles correspond to a sequence of runs in which the temperature was raised starting form $T=0$, and the triangles to "cooling" started from $T=\infty$. The curves are guides for the eye.

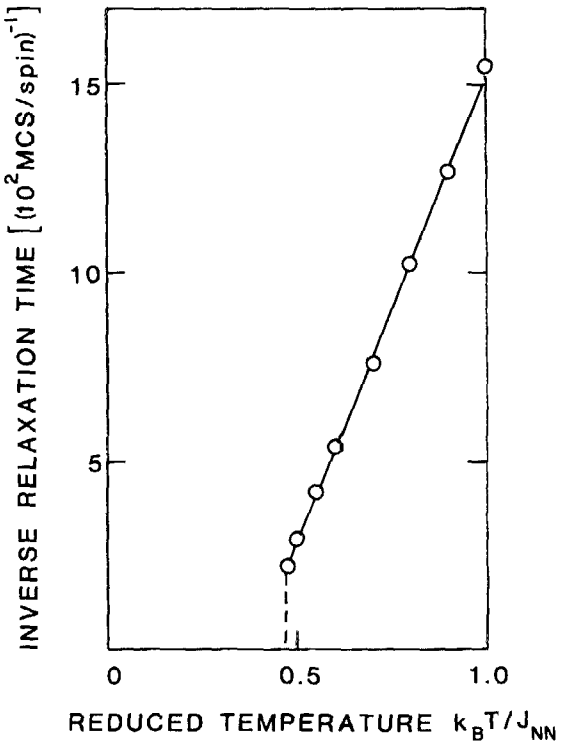

FIG. 2. Inverse relaxation time τ^{-1} vs temperature. For reduced temperatures lower than 0.475, τ^{-1} is zero.

The long-range order persisted in the sample up to $\Upsilon = 0.47$ and broke down abruptly at $\Upsilon = 0.475$. At the same time, the system energy showed a distinct discontinuity (Fig. 1). A corresponding behaviour was seen for the relaxation time: the inverse τ shows approximately linear dependence on temperature for $\Upsilon \geqslant 0.475$ (Fig. 2), falling abruptly to 0 between 0.47 and 0.475 [for $\Upsilon \leqslant 0.47$ $q(t)$ remained finite for the entire run lengths, indicating $\tau = \infty$, within the framework of our time scale]. In some Monte Carlo works on similar systems (e.g., in Ref. 9) if was found useful to analyze $q(t)$ vs temperature below the transition point, after performing the same number of MCS/spin at each temperature. However, we do not present such data, as we observed occasional a slow "drifts" of the sublattice magnetization direction, which might artificially change the data; this kind of process could not occur in the Ising model studied in Ref. 9.

The reversibility of the observed transition process was studied by "cooling" the sample starting from disordered phase. The sample returned to the ordered state for $\Upsilon = 0.45$, but not for $\Upsilon = 0.46$ after applying 20 000 MCS/spin. This fact may indicate a hysteresis, which is likely to occur in the first-order phase transition. On the other hand, the establishing of order for $\Upsilon = 0.45$ was extremely slow a process, lasting over 10 000 MCS/spin; 20 000 used at $\Upsilon = 0.46$ might therefore be too short a run, so the hysteresis may be actually narrower.

The calculated magnetic susceptibility displayed in Fig. 3 is a "polycrystalline" average over [100], [010], and [001] directions. The results show typical features of $\chi(T)$ curves of antiferromagnets, namely a sharp kink at T_c, and Curie–Weiss behavior $\chi \propto (T+\theta)^{-1}$ at high temperatures. The fit

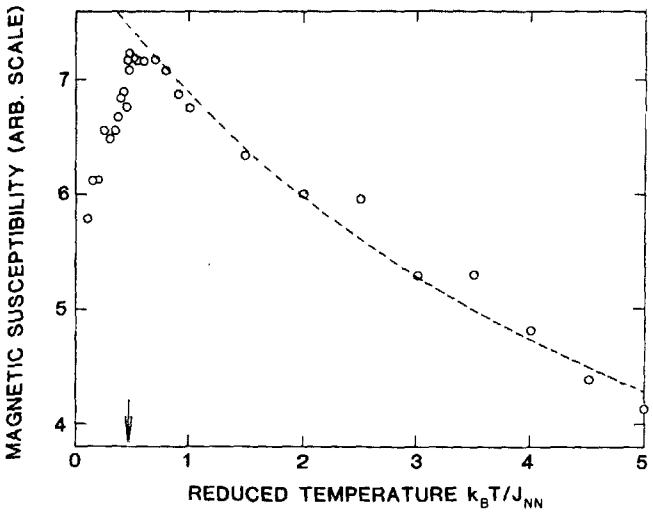

FIG. 3. Temperature dependence of the magnetic susceptibility averaged for the three cubic axes. The arrow shows the transition temperature indicated by the temperature variations of the system energy and the relaxation time. The dashed curve is the fitted Curie-Weiss function, corresponding to $\theta = 5.6J_{NN}/k_B$.

value of the Curie Weiss constant was $\theta = 5.6J_{NN}/k_B$. Large fluctuations seen in Fig. 3 indicate that the lengths of our runs were still too small for accurate calculations of the magnetic susceptibility. Indeed, as was pointed out by Bray and Moore,[20] such studied require especially long runs, $> 10^5$ MCS/spin.

IV. SUMMARY AND CONCLUSIONS

The sample that we studied exhibited Type III long-range antiferromagnetic order at low temperatures. The transition to the disordered phase was associated with discontinuities in system energy and the relaxation time, indicating that the process is of the first order. Thus, the model reproduces correctly the basic experimental characteristics of β-MnS. A comparison in quantitative terms is more difficult, as the exchange constants for β-MnS are not known. An estimate of J_{NN} can be made based on the reported[1] Curie–Weiss temperature $\theta = 982$ K. Using the molecular field theory expressions, one obtains $S(S+1)J_{NN} = 246\ \text{K} \cdot k_B$. Using this value for J_{NN} in our classical model, we obtain 115 and 1378 K for T_c and θ, as compared to the corresponding experimental values 98 K (Ref. 5) and 982 K, which seems to be a reasonable result, taking into consideration the classical approximation used in our model, and the rather arbitrary choice of $J_{NNN} = 0.1J_{NN}$.

The phase transition in fcc antiferromagnets with Heisenberg nearest-neighbor interactions only, studied by Fernandez *et al.*[10] using the same technique was found to be of the second order, and the transition temperature obtained was $0.42\,J_{NN}/k_B$. Introduction of even relatively weak second-nearest-neighbor interactions thus changes considerably the qualitative and quantitative transition characteristics of the system.

Our results confirm the conclusions made in Refs. 4 and 5 that the phase transition in case of collinear Type III allignment is of the first order, and so do not bring any new arguments for the discussion on the structure of the low-temperature phase in β-MnS. Our only finding that may be in some relation to this problem is the observed instability of the noncollinear structures in the presence of thermal excitations. Based on the discussion presented in the paper of Fernandez *et al.*,[10] we can see the following general explanation for this phenomenon: Type III structure is built up of (100)-type sheets of antiferromagnetically coupled spins. The sequence of such planes can be written as $ABABAB\ldots$, where only planes A (or B) interact between themselves via J_{NNN}, but interactions AB reduce to zero. This allows an arbitrary mutual orientation of spins in planes A and B. However, at $T > 0$ planes A and B may become additionally coupled by thermal excitations in a way preferring the collinearity. Nevertheless, it is difficult to predict whether such a mechanism would be able to compete with the Moriya–Dzialoshinski interactions, which are likely to favor the noncollinear structure in β-MnS.[3,5]

ACKNOWLEDGMENT

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[1] P. Anderson, Phys. Rev. **79**, 705 (1950).
[2] L. M. Corliss, N. Elliot, and J. Hastings, Phys. Rev. **104**, 924 (1956).
[3] F. Keffer and W. O'Sullivan, Phys. Rev. **108**, 637 (1957); also F. Keffer, Phys. Rev. **126**, 896 (1962).
[4] P. Bak and D. Mukamel, Phys. Rev. B **13**, 5086 (1976); David Mukamel, Phys. Rev. **B 17**, 4477 (1978).
[5] J. M. Hastings, L. M. Corliss, W. Kunnmann, and David Mukamel, Phys. Rev. B **24**, 1388 (1981).
[6] For a review, see e. g., J. K. Furdyna, J. Appl. Phys. **53**, 7637 (1982).
[7] A. Pajaczkowska, Prog. Cryst. Growth Charact. **1**, 289 (1978).
[8] R. R. Galazka, *Proceedings of the 14th International Congress on Physics of Semiconductors*, Edinburgh, 1978, edited by B. L. H. Wilson, IOP Conf. Ser. **43**, 133 (1978).
[9] G. S. Grest, and E. F. Gabl, Phys. Rev. Lett. **43**, 1182 (1979).
[10] J. A. Fernandez, H. A. Farah, C. P. Poole, and M. Puma, Phys. Rev. B **27**, 4274 (1983).
[11] W. Y. Ching and D. L. Huber, Phys. Rev. B **25**, 5761 (1982); **26**, 6164 (1982).
[12] R. R. Galazka, S. Nagata, and P. H. Keesom, Phys. Rev. B **22**, 3331 (1980).
[13] M. Escorne and A. Mauger, Phys. Rev. B **25**, 4674 (1982).
[14] W. Y. Ching and D. L. Huber, Phys. Rev. B **30**, 179 (1984).
[15] T. M. Giebultowicz, J. J. Rhyne, W. Y. Ching, and D. L. Huber (these proceedings).
[16] T. M. Giebultowicz, W. Minor, H. Kepa, B. Lebech, B. Buras, and R. R. Galazka, J. Appl. Phys. **55**, 2305 (1984).
[17] H. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, and E. Teller, J. Chem. Phys. **21**, 1087 (1953).
[18] *Monte Carlo Methods in Statistical Physics*, edited by K. Binder (Springer, New York, 1978).
[19] S. F. Edwards and P. W. Anderson, J. Phys. F **5**, 965 (1975).
[20] A. J. Bray and M. A. Moore, J. Phys. F **7**, L333 (1977).