Histogram Monte Carlo study of multicritical behavior in the hexagonal easy-axis Heisenberg antiferromagnet

A. Mailhot,* M. L. Plumer, and A. Caillé
Centre de Recherche en Physique du Solide et Département de Physique, Université de Sherbrooke, Sherbrooke, Québec, Canada J1K 2R1
(Received 10 March 1993)

The results of a detailed histogram Monte Carlo study of critical-fluctuation effects on the magnetic-field temperature phase diagram associated with the hexagonal Heisenberg antiferromagnet with weak axial anisotropy are reported. The multiphase point where three lines of continuous transitions merge at the spin-flop boundary exhibits a structure consistent with scaling theory but without the usual umbilicus as found in the case of a bicritical point.

The frustration of antiferromagnetically coupled sites in a triangular array gives rise to noncolinear spin order and magnetic-field temperature phase diagrams which exhibit a rich variety of structure.[1,2] In the case of a hexagonal lattice with weak (c-axis) axial anisotropy, a novel type of multicritical point occurs at which three lines of continuous transitions merge at the spin-flop and paramagnetic phase boundaries. This type of phase diagram has been observed in the quasi-one-dimensional hexagonal antiferromagnets $CsNiCl_3$,[3,4] $CsMnI_3$,[5,6] and $CsNiBr_3$.[6–8] Similar behavior is expected in[1] $RbNiCl_3$. Gross features of the experimental results for $CsNiCl_3$ have been reproduced by the analysis of a phenomenological Landau-type free energy constructed from symmetry arguments.[9] In zero field, two continuous transitions occur, identified by the components of the spin vector $\mathbf{M}$.[10] As the temperature is lowered from the paramagnetic phase 1, a linear phase 2 with M_z (where $\mathbf{z}\|\mathbf{c}$) is stabilized at T_{N1}, and at $T_{N2} < T_{N1}$ an elliptically polarized phase 3 with M_z and M_x occurs. In a magnetic field applied along the c axis, a first-order spin-flop transition to a helically polarized phase 4 with M_x and M_y is found. Each of the ordered states is characterized by a period-3 modulation in the basal plane and a period-2 structure along the c axis. (The helical phase 4 is similar to the well-known 120° spin struture of triangular antiferromagnets.) Scaling analysis of the multicritical point where phase 1, 2, 3, and 4 meet suggests that the behavior of all the three lines of continuous transitions are governed by the same crossover exponent ϕ.[11] It is the purpose of the present work to examine in detail this behavior by means of accurate histogram Monte Carlo simulations of the anisotropic Heisenberg antiferromagnet.

The easy-axis Hamiltonian studied here is given by[12]

$$\mathcal{H} = J_{\|} \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_{\perp} \sum_{\langle kl \rangle} \mathbf{S}_k \cdot \mathbf{S}_l + D \sum_i (S_{zi})^2 - H \sum_i S_{zi} \, , \tag{1}$$

where $J_{\|} > 0$ and $J_{\perp} > 0$ are the nearest-neighbor antiferromagnetic exchange interactions along the c axis and in the basal plane, respectively, $D < 0$ is the single-ion anisotropy, and H is magnetic field applied along the c axis. Although $J_{\|} >> J_{\perp}$ for $CsNiCl_3$, we consider here the isotropic case $J_{\|} = J_{\perp} = 1$ for simplicity and since Monte Carlo simulations of models with strongly anisotropic parameters require considerably more computing effort. For this reason, we also chose a value for $D = -0.2$, which is small enough for our purposes; the ground states which occur as a function of H are consistent with those observed experimentally[12] (in contrast with the case $D = -1$, for example, which yields a phase diagram with a completely different structure[1,12]). The present work serves to compliment and extend the Monte Carlo studies performed at $H = 0$ on this and similar Hamiltonians,[13,14] as well as one cursory examination of the phase diagram.[15]

This study was made in an attempt to accurately estimate the phase-boundary lines close to the multicritical point at (T_m, H_m). It was anticipated to be a numerically challenging problem since fluctuations involving all three components of $\mathbf{M}$ are important in this region of the phase diagram. The Ferrenberg-Swendsen histogram method of Monte Carlo simulations offers the possibility of the precise determination of transition points by the temperature at which extrema occur in thermodynamic functions for finite-size systems.[16] Relevant components of the staggered susceptibility, defined according to the components of the order parameter $\mathbf{M}$ involved in the transition of interest,[12] were used for this purpose. Simulations were performed on a lattice of size $12 \times 12 \times 12$. Runs of 1.2×10^6 Monte Carlo steps per spin were made, with the initial 2×10^5 steps discarded for thermalization. (Guided by previously reported Monte Carlo simulations of frustrated systems,[16,17] this latter number was chosen to be about 15% of the total number of steps. After this number of steps, relatively small statistical fluctuations are observed.) For a given value of magnetic field, single histograms were made at one or more T to ensure that the maxima in the susceptibility occured close to at least one simulation temperature.

The results shown in Fig. 1 confirm the general stucture determined by the phenomenological Landau model[9] as well as by a molecular-field treatment of the Hamiltonian (1).[1] [In the latter case, two different types of linear and elliptical states ($2A$, $2B$, $3A$, and $3B$) were distinguished by a relative phase angle, as in the Monte Carlo studies at $H = 0$.[13,14] Such distinctions are beyond, and not relevant to, the goals of the present work.] The detailed behavior near the multicritical point at $T_m = 0.915(5)$, $H_m = 2.62(4)$, however, is quite different from both mean-field results. There is clear indication that both the 1-2 and 1-4 transition lines approach this point with slopes that are the same in magnitude as that of the 3-4 spin-flop line. This is an effect of critical fluctuations and is predicted by scaling theory.[11] Less clear is that the 2-3 transition line exhibits the same predicted tendency (i.e., having the same slope as the 3-4 line at the multicritical point). A remarkable feature of the 1-4 line is its initial curvature to the left as the field increases. In the usual case of the bicritical point associated with unfrustrated antiferromagnets with weak axial anisotropy, only two critical lines are involved.[18] These also approach this point asymptotically with slopes that are the same in magnitude as the spin-flop line and are governed by the same crossover exponent ϕ. In contrast with the present case, both of these lines have always been found to approach the bicritical point from the right, forming an umbilicus structure.[19] This behavior has also been observed in Monte Carlo simulations.[20] There appears to be no argument from scaling theory, however, that relates the *signs* of the initial slopes of critical lines emanating from a multicritical point. We note that this opposite-slope behavior has not been observed experimentally. This is not surprising since the model parameters used in here are not relevant for quasi-one-dimensional materials, a feature which may obscure this unusual effect.

In conclusion, these Monte Carlo results demonstrate that significant critical-fluctuation effects are associated with this novel multicritcial point. While there are strong symmetry arguments to support the conclusion made in Ref. 11 that the 1-2 and 2-3 transitions belong to the *xy* universality class, the nature of the 1-4 transition, and that of the multicritical point itself, remains somewhat unsetteled. The interpretation made in Ref. 11 is that these transitions are related to the new chiral universality classes proposed by Kawamura.[17] (The resulting crossover-exponent value $\phi \simeq 1.06$ is not inconsistent with the results of Fig. 1.) Azaria *et al.*[21–24] have argued that such transitions exhibit nonuniversal critical behavior, where a first-order, mean-field tricritical or $O(4)$ universality can occur. It is not clear what type of scaling behavior for the multicritcal point can be expected in these cases.

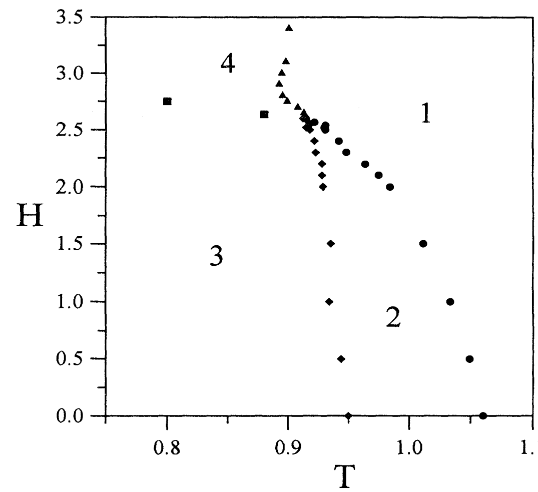

FIG. 1. Phase diagram with $\mathbf{H}\|\mathbf{z}\|\mathbf{c}$ near the multicritical point at $T_m = 0.915(5)$, $H_m = 2.62(4)$ as determined by Monte Carlo simulations (points). Indicated are the paramagnetic phase 1, linear phase 2 with M_z, elliptical phase 3 with M_z, M_x, and spin-flopped helical phase 4 with M_x, M_y.

This work was supported by NSERC of Canada and FCAR du Québec.

* Present address: INRS-EAU, 2800 rue Einstein, C.P. 7500, Sainte-Foy, Québec, Canada G1V 4C7.

[1] M.L. Plumer and A. Caillé, J. Appl. Phys. **70**, 5961 (1991).
[2] M.L. Plumer, A. Caillé, and K. Hood, Phys. Rev. B **39**, 4489 (1989).
[3] P.B. Johnson, J.A. Rayne, and S.A. Friedberg, J. Appl. Phys. **50**, 1853 (1979).
[4] M. Poirier, A. Caillé, and M.L. Plumer, Phys. Rev. B **41**, 4869 (1990).
[5] H. Kadowaki, T. Inami, Y. Ajiro, K. Nakajima, and Y. Endoh, J. Phys. Soc. Jpn. **60**, 1708 (1991).
[6] H.A. Katori, T. Goto, and Y. Ajiro, J. Phys. Soc. Jpn. **62**, 743 (1993).
[7] S. Maegawa, T. Kohmoto, T. Goto, and N. Fujiwara, Phys. Rev. B **44**, 12 617 (1991).
[8] M. Chiba, Y. Ajiro, H. Kikuchi, S. Maegawa, and T. Morimoto, J. Phys. Soc. Jpn. **61**, 1758 (1992).
[9] M.L. Plumer, K. Hood, and A. Caillé, Phys. Rev. Lett. **60**, 45 (1988); **60**, 1885(E) (1988).
[10] X. Zhu and M.B. Walker, Phys. Rev. B **36**, 3830 (1987).
[11] H. Kawamura, A. Caillé, and M.L. Plumer, Phys. Rev. B **41**, 4416 (1990).
[12] A. Mailhot, M.L. Plumer, and A. Caillé, J. Appl. Phys. **67**, 5418 (1990).
[13] S. Miyashita, in *Computer Simulation Studies in Condensed Matter Physics IV*, edited by D.P. Landau, K.K. Mon, and H.B. Schüttler (Springer-Verlag, Berlin, in press).
[14] T. Ohyama and H. Shiba, J. Phys. Soc. Jpn. **61**, 4174 (1992).
[15] G.A. Petrakovskiĭ, N.V. Fedoseeva, S.S. Aplesnin, and V.K. Korolev, Fiz. Tverd. Tela (Leningrad) **31**, 169 (1989) [Sov. Phys. Solid State **31**, 1372 (1989)].
[16] A.M. Ferrenberg and R.H. Swendsen, Phys. Rev. Lett. **61**, 2635 (1988); A.M. Ferrenberg and D.P. Landau, Phys. Rev. B **44**, 5081 (1991); J.N. Reimers, J.E. Greedan, and M. Björgvinsson, *ibid.* **45**, 7295 (1992).
[17] H. Kawamura, J. Phys. Soc. Jpn. **55**, 2095 (1986); Phys. Rev. B **38**, 4916 (1988); **42**, 2610(E) (1990); J. Phys. Soc. Jpn. **61**, 1299 (1992); **59**, 2305 (1990).

[18] M.E. Fisher, Phys. Rev. Lett. **34**, 1634 (1975); J.M. Kosterlitz, D.R. Nelson, and M.E. Fisher, Phys. Rev. B **13**, 412 (1976).
[19] Y. Shapira, J. Appl. Phys. **57**, 3268 (1985).
[20] D.P. Landau and K. Binder, Phys. Rev. B **17**, 2328 (1978).
[21] P. Azaria, B. Delamotte, and T. Jolicoeur, Phys. Rev. Lett. **64**, 3175 (1990); J. Appl. Phys. **69**, 6170 (1991).
[22] T. Bhattacharya, A. Billoire, R. Lacaze, and Th. Jolicoeur (unpublished).
[23] M.L. Plumer, A. Mailhot, and A. Caillé, Phys. Rev. B **48**, 3840 (1993).
[24] A. Mailhot, M.L. Plumer, and A. Caillé (unpublished).