Some Critical Properties of Quantum-Mechanical Heisenberg Ferro- and Antiferromagnets

H. E. STANLEY*

Lincoln Laboratory,† Massachusetts Institute of Technology, Lexington, Massachusetts and

Physics Department, University of California, Berkeley, California

High-temperature expansions of the *staggered* susceptibility for the Heisenberg antiferromagnet are analyzed for two-dimensional lattices. The evidence favoring a divergent staggered susceptibility $\tilde{\chi}$ is found to be essentially as strong as that presented earlier in support of a divergent susceptibility χ for the two-dimensional Heisenberg ferromagnet. Thus the very recent experimental and theoretical findings which favor a phase transition in various "planar antiferromagnets" (or materials with the K_2NiF_4 structure in which the intraplanar interactions may be as much as 10^5 times stronger than any interplanar interactions) would appear to be supported by the series expansions. We also consider the question of the dependence of the susceptibility exponent $\gamma[\chi\sim(T-T_c)^{-\gamma}]$ upon spin quantum number S for three-dimensional lattices. The previously proposed behavior must definitely be revised in the light of additional terms in the expansions and the availability of more refined extrapolation procedures. Here we argue (a) that $\gamma(\infty)=1.38\pm0.01$ and (b) that $\gamma(S)$ does not appear to jump, for S finite, to 1.43 (its presumed value for $S=\frac{1}{2}$).

I. STAGGERED SUSCEPTIBILITY OF TWO-DIMENSIONAL HEISENBERG ANTIFERROMAGNETS

Initially it was assumed that a system of spins situated on a two-dimensional lattice and interacting with a Heisenberg Hamiltonian would possess a finite zero-field susceptibility χ for all positive temperatures T. In 1966 this assumption was questioned on two grounds[1]: (i) the coefficients in the high-temperature expansions for χ suggested that χ diverged at a nonzero temperature T_c, and (ii) there existed absolutely no evidence *against* a divergent χ; the only evidence at all was a nonrigorous argument for zero spontaneous magnetization M. Shortly thereafter, it was rigorously proved[2] that $M=0$ for $T>0$, leaving only the possibility, as suggested in Ref. 1, of a phase transition to a low-temperature phase in which there is no "infinite-range order" $[M\propto\lim_{R\to\infty}\Gamma_R^{1/2}]$, yet there is sufficient "long-range order" that $\chi\propto\sum_R\Gamma_R\to\infty$; $\Gamma_R\equiv\langle\mathbf{S}_0\cdot\mathbf{S}_R\rangle$ is the two-spin correlation function. This possibility[1] derived some support from a low-temperature approximation for the quantum-mechanical Heisenberg model,[3] subsequently applied also to the two-dimensional harmonic solid[4]; these calculations predict that at large spin separation R, $\Gamma_R\propto R^{-\lambda T}$, so that $M=0$ yet $\chi=\infty$ for low T. A different picture of the proposed $M=0$ "low-temperature phase" has recently arisen from a Green's function calculation.[5]

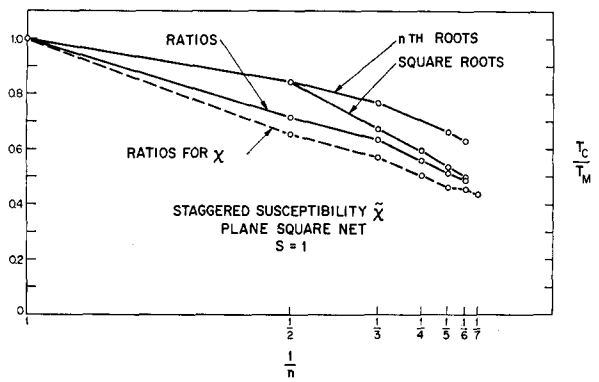

FIG. 1. Ratios $\tilde{a}_n/\tilde{a}_{n-1}$, square roots $(\tilde{a}_n/\tilde{a}_{n-1})^{1/2}$, and nth roots $(\tilde{a}_n)^{1/n}$ formed from coefficients $\tilde{a}_n$ in the staggered susceptibility series; for comparison are shown the ratios for the ferromagnetic susceptibility series. All the plots are normalized by dividing by $\tilde{a}_1$ in order that the intercept at $1/n=0$ be the ratio of the critical temperature to the molecular-field critical temperature.

TABLE I. Ratios $\rho_n\equiv\tilde{a}_n/\tilde{a}_1\tilde{a}_{n-1}$ of successive coefficients $\tilde{a}_n$ in the staggered susceptibility series for the plane square lattice. The $\tilde{a}_n$ themselves are obtained using $\tilde{a}_1=2zS(S+1)/3$. Classically (as $S\to\infty$) $\tilde{\chi}=\chi$ and the $\tilde{\rho}_n(=\rho_n)$ are given, through order $n=9$, in Table III of Ref. 15.

n	$S=\frac{1}{2}$	$S=1$	$S=\frac{3}{2}$	$S=\frac{5}{2}$	$S=\frac{7}{2}$
1	1.000000000	1.000000000	1.000000000	1.000000000	1.000000000
2	0.666666667	0.718750000	0.733333333	0.742857143	0.746031746
3	0.500000000	0.635869565	0.667272727	0.686373626	0.692502533
4	0.365625000	0.560202991	0.598109673	0.620767148	0.628021258
5	0.329059829	0.511681129	0.555934626	0.583790668	0.592892590
6	0.518939394	0.488380669	0.532227615	0.560477020	0.569711310

* Fellow, Miller Institute for Basic Research in Science.

† Operated with support from the U.S. Air Force.

[1] H. E. Stanley and T. A. Kaplan, Phys. Rev. Letters **17**, 913 (1966).
[2] N. D. Mermin and H. Wagner, Phys. Rev. Letters **17**, 1133 (1966).
[3] F. J. Dyson (unpublished).
[4] B. Jancovici, Phys. Rev. Letters **19**, 20 (1967).
[5] V. Mubayi and R. V. Lange, Phys. Rev. (in press).

Experimentally, it appears impossible to actually *build* a truly two-dimensional lattice (although a recent computer simulation of the classical Heisenberg model[6] suggests the persistence of considerable "order" for $T>0$). However two groups of workers[7,8] have found evidence supporting a "phase transition" in a class of layered structures (generally with the K_2NiF_4 structure) for which the intraplanar interactions are apparently several orders of magnitude stronger than any interplanar interactions. Since the magnetic structure within the planes is that of an *antiferromagnetic* square net, we have calculated the coefficients in the high-T expansions for the *staggered* susceptibility $\tilde{\chi}$ of a square net antiferromagnet[9]; the results are summarized in Table I. Various extrapolation procedures (see, e.g., Fig. 1) suggest that the "numerical evidence" favoring a divergent staggered susceptibility for the antiferromagnet is just as convincing as that presented earlier in favor of a divergent susceptibility for the ferromagnet. *Note added in proof*: Extremely convincing evidence from neutron scattering experiments for "two-dimensional ordering" in K_2NiF_4 has recently been obtained by R. J. Birqeneau, H. J. Guggenheim and G. Shirane, Phys. Rev. Letters (in press).

II. DEPENDENCE OF THE HEISENBERG-MODEL SUSCEPTIBILITY EXPONENT ON SPIN QUANTUM NUMBER

It was once thought that the exponent γ describing the divergence of the zero-field susceptibility, $\chi\sim(T-T_c)^{-\gamma}$, had the value $\frac{4}{3}$ for all values of the spin quantum number S. In 1964 it was suggested[10] that γ might have a weak spin dependence, $\gamma(S)\cong 1.33+0.05/S$. As additional terms in the various series became available, it was argued that[11] $\gamma(\frac{1}{2})=1.43\pm0.03$ and that[12] $\gamma(\infty)\cong 1.36$–1.38. Hence it would now appear that any spin dependence, if it exists at all, would be barely outside the margin of "experimental error" inherent in the extrapolation procedures.[13]

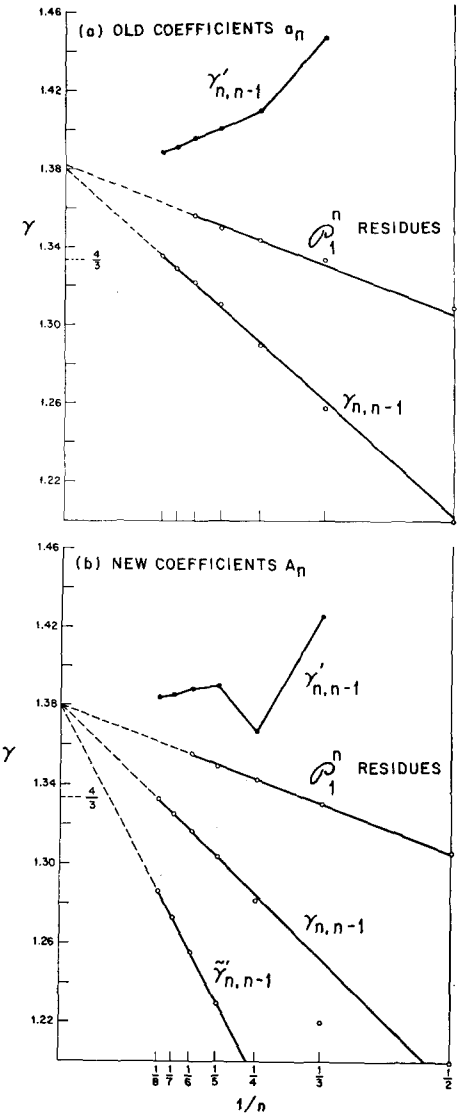

FIG. 2. Fcc lattice for the classical ($S=\infty$) Heisenberg model. (a) The function $\gamma_{n,n-1}$ gives the value of γ which would be obtained by placing a straight line through successive ratios plotted against $1/n$ (see Ref. 15 for discussion). More precisely, $\gamma_{n,n-1}\equiv 1-n+n\rho_n/t_{n,n-1}$, where $t_{n,n-1}\equiv n\rho_n-(n-1)\rho_{n-1}$, and $\rho_n\equiv a_n/a_1a_{n-1}$, and a_n are the coefficients in the susceptibility series with expansion parameter $x\equiv J/kT$. The function γ_{nn-1}' is defined by the same equations with primes on all quantities, where $\rho_n'\equiv(a_n/a_{n-2})^{1/2}/a_1$. Finally, P_1^n are the $[1, n]$ sequence of Padé approximants. (b) The same functions are plotted as in part (a), with a_n replaced by A_n, where A_n are the coefficients in the susceptibility series with expansion parameter $y=L(x)$. The additional function plotted is $\tilde{\gamma}_{n,n-1}'\equiv 1-n+n\rho_{n+1}'/t_{n+1,n}'$.

Here we apply various extrapolation procedures not used before (and use a new term[14] in the general-S susceptibility series) (a) to argue more convincingly that $\gamma(\infty)=1.38\pm0.01$, and (b) to suggest that $\gamma(S)$ does not appear to jump suddenly to 1.43 for finite S [as apparently some workers had expected].

[6] R. E. Watson, M. Blume, and G. Vineyard (unpublished).

[7] J. Koppen, R. Hamersma, J. V. Lebesque, and A. R. Miedema, Phys. Letters **25A**, 376 (1967); D. J. Breed, Physica **37**, 35 (1967); G. De Vries, D. J. Breed, E. P. Maarschall, and A. R. Miedema, J. Appl. Phys. **39**, 1207 (1968).

[8] G. K. Wertheim, H. J. Guggenheim, H. J. Levinstein, D. N. E. Buchanan, and R C. Sherwood, Phys. Rev. (in press). See also M. E. Lines, Phys. Letters **24A**, 591 (1967); Phys. Rev. **164**, 736 (1967).

[9] We wish to thank Prof. G. S. Rushbrooke for sending us the "general-lattice expressions" for the coefficients in the staggered susceptibility series (obtained by him and P. J. Wood); see, e.g., Mol. Phys. **6**, 409 (1963).

[10] H. E. Stanley and T. A. Kaplan (Comment at 1964 Magnetism Conf.); J. Appl. Phys. **38**, 977 (1967).

[11] G. A. Baker, H. E. Gilbert, J. Eve, and G. S. Rushbrooke, Phys. Letters **20**, 146 (1966); Phys. Rev. **164**, 800 (1967).

[12] It was P. J. Wood and G. S. Rushbrooke, Phys. Rev. Letters **17**, 307 (1966) who first suggested that $\gamma(\infty)$ was appreciably larger than 4/3. *Note added in proof*: R. G. Bowers (unpublished) has recently put forth arguments (supporting ours) that $\gamma(\infty)\leqq 11/8$.

[13] In fact, most current theoretical ideas concerning the critical exponents predict there should be no spin dependence whatsoever.

[14] R. L. Stephenson, K. Pirnie, P. J. Wood, and J. Eve, Phys. Letters **27A**, 2 (1968). Incidentally, the additional general-S term for *two-dimensional* lattices supports the arguments in Ref. 1 for a divergent susceptibility.

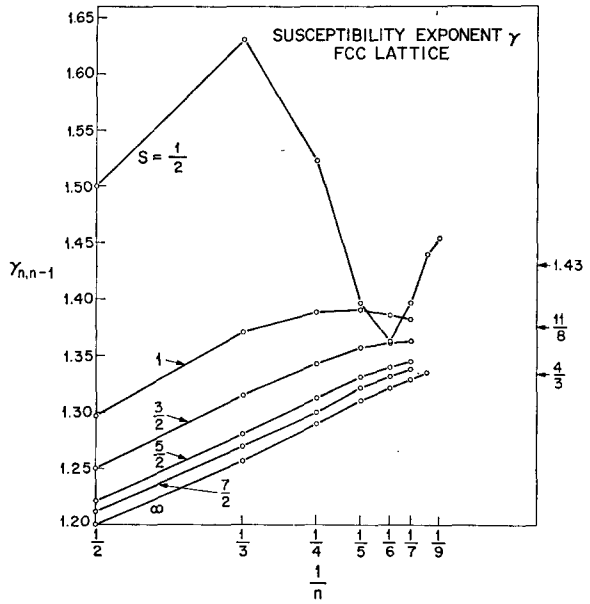

FIG. 3. The function $\gamma_{n,n-1}$ (see Caption to Fig. 2 for definition) for 6 different values of spin quantum number. The evidence for $\gamma(\frac{1}{2})=1.43\pm0.03$ was based upon Padé analysis.[11]

(a) It has been observed that smoother series for the classical Heisenberg model ($S\to\infty$ limit) are frequently obtainable by expanding not in the variable $x\equiv J/kT$ but rather in the new variable $y\equiv L(x)\equiv\coth x-x^{-1}$. The improvement in using the new expansion parameter is exemplified for the *fcc* lattice in Fig. 2, where the extrapolation techniques described elsewhere[15] are used first for the old expansion [Fig. 2(a)] and then for the new expansion [Fig. 2(b)]. Certainly the values $\gamma=\frac{4}{3}$ and $\gamma=1.43$ are not indicated; however it would be difficult to exclude the possibility, say, that $\gamma=1.375$ $(=11/8)$.

(b) When we apply various extrapolation procedures to the series for finite S, we find no indication that γ jumps to 1.43. In fact, there exists a little evidence (see, e.g., Fig. 3) suggesting that *perhaps* $\gamma=1.38$ for all $S>\frac{1}{2}$. However, we feel that the series are still too short to permit a firm answer to the question, "Does γ vary smoothly or discontinuously with S as S goes from $\frac{1}{2}$ to ∞?"

[15] H. E. Stanley, Phys. Rev. **158**, 546 (1967).

JOURNAL OF APPLIED PHYSICS VOLUME 40, NUMBER 3 1 MARCH 1969

Entropy and Susceptibility of a Pure Dipole–Dipole System (Cerous Magnesium Nitrate)*

PAUL H. E. MEIJER
National Bureau of Standards and *The Catholic University of America, Washington, D. C. 20017*
AND
DAVID J. O'KEEFFE
The Catholic University of America, Washington, D. C. and *The U.S. Naval Ordnance Laboratory, Silver Spring, Maryland*

The Van Vleck moment expansion is applied to a pure dipole system. On the basis of the long-range nature of the forces, sequences of diagrams are selected that give the dominant contributions in the lattice sums. A selection of diagrams contributing to the entropy and susceptibility is given. There are three different summations to be performed for each type of diagram: the trace over the spin variables, the lattice summation, and the summation over the Cartesian coordinates. The second is performed on the computer and the last by means of the Kramers–Wannier diagonalization. In order to obtain the contributions of diagrams of higher order a Fourier transform is performed. The calculations were performed for Cerous Magnesium Nitrate using a g factor that is zero along the c axis.

In order to determine the coefficients in the Van Vleck[1] moment expansion for Cerous Magnesium Nitrate, which has an almost pure dipole–dipole interaction, we evaluated a number of terms indicated by the diagrams in Fig. 1. The contribution of each diagram is the product of three factors, a weight factor, factors from the traces of the various spin contractions in the vertices, and a lattice summation. The last is done on a computer.

ENTROPY DIAGRAMS

The leading diagram for each power n of $1/kT$ ($n=$ the number of bonds) is the ring diagram with n vertices. The various components xx, xy, etc. of the dipole–dipole interaction is taken care of by a Kramers–Wannier[2] diagonalization. In the case of CMN this needs only a two-by-two diagonalization since $g_{\|}\approx0$; the second- and third-order terms were computed directly. The leading contribution to the fourth-order

* Supported by AF-AFOSR-66-0445a.
[1] J. H. Van Vleck, J. Chem. Phys. **5**, 320 (1937).
[2] H. A. Kramers and G. H. Wannier, Phys. Rev. **60**, 252 (1941).