Dependence of Critical Properties of Heisenberg Magnets upon Spin and Lattice

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High-temperature expansion methods have recently been used to predict the *form* of the divergence of the zero-field static susceptibility χ at the critical temperature T_c. These studies have suggested that $\chi \sim A(T-T_c)^{-\gamma}$, with $\gamma=\frac{4}{3}$ independent of both lattice structure and spin quantum number S. Here we argue that the proposal $\gamma=\frac{4}{3}$ for all S is unjustified; we find a slow but nevertheless clear variation of γ with S. We further point out that there exists at least one physically interesting lattice —a normal cubic spinel with nearest-neighbor ferromagnetic B–B interactions—for which the theoretical evidence indicates that if the power-law form of divergence is correct, γ may differ from $\frac{4}{3}$ by as much as 50%.

I. INTRODUCTION

HIGH-TEMPERATURE extrapolation methods[1] have recently been used to predict not only the radius of convergence, $z_c=J/kT_c$, of the power series for the zero-field static susceptibility

$$\chi(z)/\chi_{\mathrm{Curie}}=1+\sum_{l=1}^{\infty} a_l z^l, \tag{1}$$

but also the *form* of the divergence at the critical temperature T_c.[2,3] These studies have assumed the divergence to be of the power law form $\chi(z)\sim A(z_c-z)^{-\gamma}$, and have proposed on the basis of the six terms a_l known for general spin quantum number S, that for three-dimensional lattices $\gamma(S)\cong\frac{4}{3}$. These studies[2,3] have suggested that the value $\gamma(S)=\frac{4}{3}$ is "universal" in the sense that it is independent of both lattice structure and spin quantum number S.

We point out that, using only the first six terms a_l, the conclusion of Refs. 2 and 3 as to the independence of γ on S is unwarranted. We find, instead, a slow, but nevertheless clear, variation of γ with S. We further point out that there exists at least one three-dimensional lattice—a normal cubic spinel with nearest-neighbor ferromagnetic interactions between the B-site cations—for which the theoretical evidence indicates that γ differs *appreciably* from $\frac{4}{3}$. This is of more than academic interest, as very recently several insulating ferromagnetic spinels with nonmagnetic A sites (e.g., $CdCr_2S_4$) have been discovered.[4,5]

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II. DEPENDENCE OF γ UPON SPIN

The basic idea behind the extrapolation method of determining T_c is that one guesses the radius of convergence of the power series (1) by extrapolating to $l=\infty$ from the first several a_l, when these a_l behave regularly. The curves of Fig. 1 are plots of a_l/a_1a_{l-1} for the face-centered cubic lattice with $S=\frac{1}{2}$, 1, $\frac{3}{2}$, and ∞. (Figure 1 includes the additional terms available[6,7] for the special cases $S=\frac{1}{2}$, ∞.) The observation that each of these plots appears to approach a straight line for large l motivates the extrapolation to $l=\infty$ shown by the dashed lines. It follows that the intercept is the ratio of T_c to the ordering temperature T_M predicted by the Weiss molecular field approximation. Moreover, if χ is to diverge as $T\rightarrow T_c^+$ with a power law, then for large l, $a_l/a_1a_{l-1}\cong(T_c/T_M)[1+(\gamma-1)/l]$. The slopes of the four curves of Fig. 1 correspond to the estimates $\gamma(\frac{1}{2})\cong1.41$, $\gamma(1)\cong1.38$, $\gamma(\frac{3}{2})\cong1.36$, and $\gamma(\infty)\cong1.33$. It is seen that the additional terms available for $S=\frac{1}{2}$ and $S=\infty$ improve the reliability of the estimates of γ.[8] Our results for the fcc are conveniently summarized, to within a few percent, by the formula

$$\gamma(S)\cong1.33+0.05/S. \tag{2}$$

Identical values for γ are obtained if one uses the Domb–Sykes criterion[2] for estimating γ.

[6] G. A. Baker, H. E. Gilbert, J. Eve, and G. S. Rushbrooke, Phys. Letters **20**, 146 (1966).

[7] H. E. Stanley and T. A. Kaplan, Phys. Rev. Letters **16**, 981 (1966); P. J. Wood and G. S. Rushbrooke, Phys. Rev. Letters **17**, 307 (1966); H. E. Stanley (submitted to Phys. Rev.)

[8] G. A. Baker [Phys. Rev. **136**, A1376 (1964)] has claimed to prove, on the basis of six terms, that for the fcc $\gamma(\frac{1}{2})\leq1.34$. But recently Baker *et al.* (Ref. 6) proposed, on the basis of nine terms in the series (extended for $S=\frac{1}{2}$), that $\gamma(\frac{1}{2})=1.43\pm0.03$. Our result, $\gamma(\frac{1}{2})=1.41$, with the very small uncertainty indicated by the regularity of the last four ratios in Fig. 1 is clearly inconsistent with Baker's upper bound and is consistent with the more recent value of Baker *et al.*

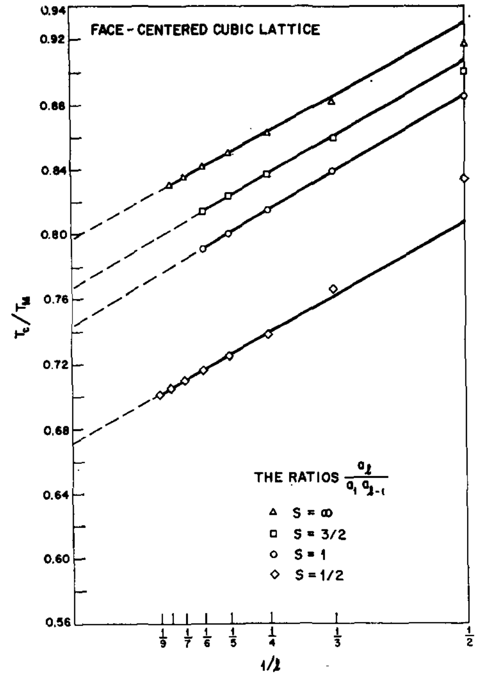

FIG. 1. The ratios a_l/a_1a_{l-1} of the susceptibility series (1) are plotted against $1/l$ for the face-centered cubic lattice for $S=\frac{1}{2}$, 1, $\frac{3}{2}$, and ∞. Note that the value of γ (for $\gamma \gtreqqless 1$) determined by the slope method is not very sensitive to one's choice of an asymptotic straight line, since its slope is proportional to $\gamma-1$.

We find, applying both of these criteria to the bcc and sc lattices, that the values of γ, though less reliable, are consistent with (2). We also used both criteria to study the variation of γ with S for the plane square and plane triangular lattices[9]; for both of these two-dimensional lattices, we found a more pronounced variation of $\gamma^{(2)}$ with S than for the three-dimensional cubic lattices considered. This variation is conveniently summarized by a mnemonic formula analogous to Eq. (2): $\gamma^{(2)}(S)\cong 2.5+0.67/S^2$.[10]

A second method of determining γ, given the assumption that χ diverges with a power law, is the method of Padé approximants. For three cubic lattices (sc, bcc, and fcc), Gammel, Marshall, and Morgan[3] found that for $S\geq 1$ the Padé approximants seem to converge to some value of γ within 10% of $\frac{4}{3}$. A careful study of their numerical results reveals a slow, but nevertheless clear, variation of γ with S, consistent with Eq. (2).

III. DEPENDENCE OF γ UPON LATTICE

For the spinel with ferromagnetic interactions between nearest-neighbor B sites, general expressions[1] for the zero-field susceptibility may be used to get the a_l by taking Rushbrooke and Wood's lattice constants[1] to be $z=6$, $p_1=p_2=p_4=r=2$, $p_3=q=0$. We plot the ratios a_l/a_1a_{l-1} for $S=\frac{3}{2}$ (corresponding to the spin-only moment of Cr^{3+}) in Fig. 2. The plot does not seem to be approaching a straight line but rather "turns around," so that how best to extrapolate to $l=\infty$ is not clear. The plot in Fig. 2 of $(a_l/a_{l-2})^{1/2}/a_1$ should also approach T_c/T_M with slope proportional to $\gamma-1$ if χ diverges with a power law. Again, there is no clear limiting behavior. Following Ref. 3, we also formed all Padé approximants to the logarithmic derivative of χ which can be obtained, given the six known terms a_l; we found neither a consistent pole location z_c nor a consistent residue γ. Finally, we plot (Fig. 2) the lth roots of a_l against $1/l$. It is seen that this plot is nearly a straight line (with a slight "upward" curvature) and one might be tempted to use this plot as a basis for extrapolation to $l=\infty$, as has been done in the past.[1] However, one should be particularly careful in extrapolating the lth roots if one assumes a power-law divergence of χ—indeed, the slope of $(a_l)^{1/l}$ vs $1/l$ would, for large l, become proportional to

$$(a_l)^{1/l}\,[\text{constant}+(\gamma-1)\ln l]$$

which approaches $\pm\infty$ for $\gamma\neq 1$. Such a rapid variation if $\gamma\neq 1$ would seem to mean that it is impossible to

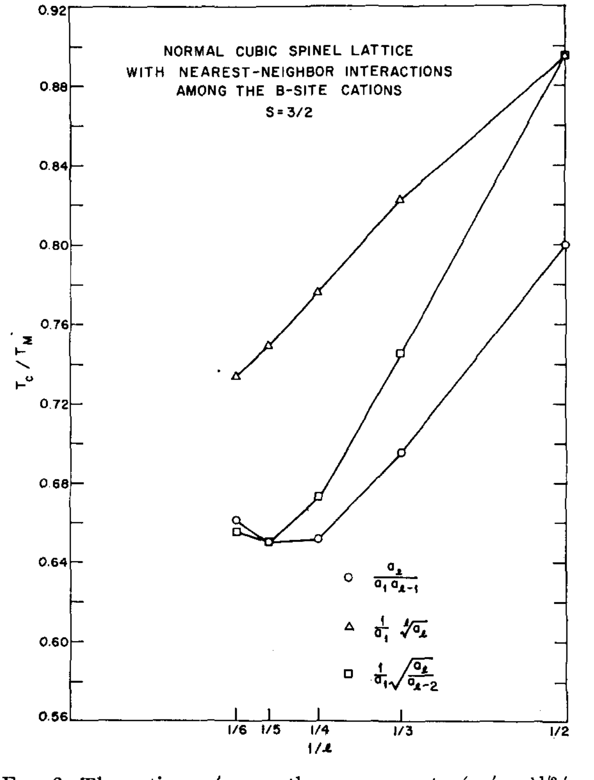

FIG. 2. The ratios a_l/a_1a_{l-1}, the square roots $(a_l/a_{l-2})^{1/2}/a_1$, and the lth roots $(a_l)^{1/l}/a_1$ plotted against $1/l$ for the spinel lattice with nearest-neighbor ferromagnetic interactions among the B-site cations and $S=\frac{3}{2}$ (corresponding to the spin-only moment of Cr^{3+} in the ferromagnetic spinels $CdCr_2X_4$ studied experimentally[4,5]).

[9] H. E. Stanley and T. A. Kaplan, Phys. Rev. Letters **17**, 913 (1966) and these Proceedings. Also, H. E. Stanley (unpublished work).

[10] That $\gamma^{(3)}$ is smaller than $\gamma^{(2)}$ is consistent with the speculation [M. E. Fisher and D. S. Gaunt, Phys. Rev. **133**, A224 (1964)] that γ should decrease with increasing dimensionality (and approach the molecular field value, $\gamma=1$, in the limit of an infinite-dimensional lattice).

reasonably extrapolate from a plot of lth roots unless $\gamma=1$. On the other hand, plots of the lth roots, together with plots of a_l/a_{l-1} and $(a_l/a_{l-2})^{1/2}$, *have* been used to reliably determine T_c for three-dimensional cubic lattices for which γ differs from unity by as much as 40%. We have found that the degree of upward concavity of the plot in Fig. 2 is considerably less than the degree of downward concavity for corresponding plots of $(a_l)^{1/l}/a_1$ for the three-dimensional cubic lattices. This suggests that for the spinel lattice with nearest-neighbor ferromagnetic interactions between the B-site cations, $\gamma\cong1$. Clearly additional terms in the high-temperature expansions are needed, and we have begun the extensions of the series for $S=\frac{1}{2}$, ∞.

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Magnetic Critical-Point Behavior of CrO_2*

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The magnetization σ of the ferromagnetic compound CrO_2 was measured as a function of field H and temperature T near the Curie point T_c. From isotherms of σ^2 vs H/σ, the initial susceptibility χ_0 above T_c was obtained, which when tested against the relationship $\chi_0^{-1}\propto(T-T_c)^\gamma$ gives a constant γ of 1.63 ± 0.02 from just above T_c (386.5°K) up to about 1.15 T_c. This γ value contrasts with the values near $\frac{4}{3}$ recently computed for the Heisenberg model and later found experimentally in various ferromagnetic metals and compounds. At higher temperatures the effective γ decreases rapidly towards unity. Up to the highest field used (25 kOe), the critical isotherm obeys the relationship $\sigma\propto H^{1/\delta}$ with $\delta=5.75\pm0.05$, which differs markedly from the theoretical δ values of 3 (molecular field) and 5.2 (3-dimensional Ising) and from various experimental values. Gradual departure from this relationship below 1.5 kOe is attributed to the magnetocrystalline anisotropy that persists at T_c. Furthermore, we find that all the $\sigma(H, T)$ data for CrO_2 just above T_c can be represented by a universal function of the form, $\sigma/\sigma'=f(H/H')$, in which $\sigma'\propto(T-T_c)^\lambda$ and $H'\propto(T-T_c)^{\lambda+\gamma}$, where $\lambda=0.34$. This "corresponding states" representation is the exact magnetic analog of an equation of state recently proposed by Widom for a fluid near its critical point.

WITHIN the context of current interest in magnetic critical-point phenomena, the rutile CrO_2 seemed to us an excellent choice for detailed study near its Curie point ($T_c\approx390$°K) since it is one of very few stoichiometric ionic compounds that orders ferromagnetically. Its saturation moment at 0°K of about 130 emu/g ($2\mu_B$ per chromium ion) is consistent with a Cr^{4+} state; its electrical conductivity, however, is metallic.[1] The present work reports the initial susceptibility χ_0 vs temperature just above T_c and the magnetization σ vs field at T_c (the critical isotherm).

All theories for the susceptibility predict

$$\chi_0^{-1}\propto(T-T_c)^\gamma. \tag{1}$$

Whereas the molecular field model gives $\gamma=1$, recent exact calculations for the Ising[2] and Heisenberg[3] ferromagnets yield γ values close to $\frac{5}{4}$ and $\frac{4}{3}$, respectively, for any spin on any basic cubic lattice. Experiments have shown that various ferromagnetic materials (Fe,[4] Ni,[5,6] Gd,[7] $YFeO_3$,[8] some complex copper salts[9]) obey Eq. (1) with $\gamma\approx\frac{4}{3}$, in essential agreement with the Heisenberg model calculations; however, for cobalt, $\gamma\approx\frac{5}{4}$ has been reported.[10] For the critical isotherm, the molecular field and three-dimensional Ising[11] models predict

$$\sigma\propto H^{1/\delta}, \tag{2}$$

with $\delta=3$ and 5.2, respectively. Recent experiments have yielded δ values of 4.22 (Ni),[5] 4 (Gd),[7] and 2.8 ($YFeO_3$).[8]

Our CrO_2 specimen was a cylinder, 0.37-cm diam by 1-cm long, of compressed powder of stoichiometric material prepared here by DeVries. The magnetization was measured at regular intervals between 370° and 530°K in fields of 500 Oe up to 25 kOe; the results

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