High-temperature expansion methods have recently been used to predict the form of the divergence of the zero-field static susceptibility $\chi$ at the critical temperature $T_c$. These studies have suggested that $\chi \sim A (T - T_c)^{-\gamma}$, with $\gamma = \frac{1}{2}$ independent of both lattice structure and spin quantum number $S$. Here we argue that the proposal $\gamma = \frac{1}{2}$ for all $S$ is unjustified; we find a slow but nevertheless clear variation of $\gamma$ 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 the power-law form of divergence is correct, $\gamma$ may differ from $\frac{1}{2}$ by as much as 50%.

I. INTRODUCTION

High-temperature extrapolation methods 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

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

(1)

but also the form of the divergence at the critical temperature $T_c$. These studies have assumed the divergence to be of the power law form $\chi(z) \sim A (z - z_c)^{-\gamma}$, and have proposed on the basis of the six terms $a_l$ known for general spin quantum number $S$, that for three-dimensional lattices $\gamma(S) \approx \frac{1}{2}$. These studies have suggested that the value $\gamma(S) = \frac{1}{2}$ 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 $\gamma$ on $S$ is unwarranted. We find, instead, a slow, but nevertheless clear, variation of $\gamma$ 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 $\gamma$ differs appreciably from $\frac{1}{2}$. This is of more than academic interest, as very recently several insulating ferromagnetic spinels with nonmagnetic A sites (e.g., CdCr$_2$S$_4$) have been discovered.\footnote{G. A. Baker, H. E. Gilbert, J. Eve, and G. S. Rushbrooke, Phys. Letters 20, 146 (1966).} \footnote{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.)}

II. DEPENDENCE OF $\gamma$ 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 $a_l/a_{l-1}$ for the face-centered cubic lattice with $S = \frac{1}{2}$, 1, $\frac{3}{2}$, and $\infty$. (Figure 1 includes the additional terms available for the special cases $S = \frac{1}{2}$, $\infty$.) 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 $\chi$ is to diverge as $T \to T_c^+$ with a power law, then for large $l$, $a_l/a_{l-1} \to (T_c/T_M)[1 + (\gamma - 1)/l]$. The slopes of the four curves of Fig. 1 correspond to the estimates $\gamma(S) \approx 1.41$, $\gamma(1) \approx 1.38$, $\gamma(\frac{3}{2}) \approx 1.36$, and $\gamma(\infty) \approx 1.33$. It is seen that the additional terms available for $S = \frac{1}{2}$ and $S = \infty$ improve the reliability of the estimates of $\gamma$. Our results for the fcc are conveniently summarized, to within a few percent, by the formula

$$\gamma(S) \approx 1.33 + 0.05/S.$$

(2)

Identical values for $\gamma$ are obtained if one uses the Domb–Sykes criterion for estimating $\gamma$.\footnote{G. A. Baker [Phys. Rev. 136, A1376 (1964)] has claimed to prove, on the basis of six terms, that for the fcc $\gamma(\frac{1}{2}) \leq 1.34$. But recently Baker et al. (Ref. 6) proposed, on the basis of nine terms in the series (extended for $S = 4$), that $\gamma(\frac{1}{2}) = 1.43 \pm 0.03$. 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.}
a_l by taking Rushbrooke and Wood’s lattice constants to be \( z = 6 \), \( p_1 = p_2 = p_3 = r = 2 \), \( p_2 = q = 0 \). We plot the ratios \( a_l/a_{l-1} \) for \( S = \frac{3}{2} \) (corresponding to the spin-only moment of Cr\(^{4+}\)) 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 \( (a_l/a_{l-2})^{1/2}/a_l \) should also approach \( T_C/T_M \) with slope proportional to \( \gamma - 1 \) if \( \chi \) 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 \( \chi \) which can be obtained, given the six known terms \( a_l \); we found neither a consistent pole location \( z_{\epsilon} \) nor a consistent residue \( \gamma \). Finally, we plot (Fig. 2) the \( l \)th 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. However, one should be particularly careful in extrapolating the \( l \)th roots if one assumes a power-law divergence of \( \chi \)—indeed, the slope of \( (a_l)^{1/2} \) vs \( 1/l \) would, for large \( l \), become proportional to

\[
(a_l)^{1/2} \left[ \text{constant} + (\gamma - 1) \ln l \right]
\]

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

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Fig. 1. The ratios \( a_l/a_{l-1} \) 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 \( \infty \). Note that the value of \( \gamma \) (for \( \gamma \leq 1 \)) 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 \( \gamma \), though less reliable, are consistent with (2). We also used both criteria to study the variation of \( \gamma \) with \( S \) for the plane square and plane triangular lattices; 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):

\[
\gamma^{(S)} = 2.5 + 0.67/S.
\]

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

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III. DEPENDENCE OF \( \gamma \) UPON LATTICE

For the spinel with ferromagnetic interactions between nearest-neighbor B sites, general expressions for the zero-field susceptibility may be used to get the

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10 That \( \gamma^{(0)} \) is smaller than \( \gamma^{(W)} \) is consistent with the speculation [M. E. Fisher and D. S. Gaunt, Phys. Rev. 133, A224 (1964)] that \( \gamma \) 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 $l$th roots unless $\gamma = 1$. On the other hand, plots of the $l$th roots, together with plots of $a_1/a_{l-1}$ and $(a_1/a_{l-2})^{1/3}$, have been used to reliably determine $T_c$ for three-dimensional cubic lattices for which $\gamma$ 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 $(a_1)^{1/3}/a_1$ for the three-dimensional cubic lattices. This suggests that for the spinel lattice with nearest-neighbor ferromagnetic interactions between the B-site cations, $\gamma \approx 1$. Clearly additional terms in the high-temperature expansions are needed, and we have begun the extensions of the series for $S = \frac{3}{2}, \infty$.

**Critical Properties of Heisenberg Magnets**

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The magnetization $\sigma$ 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 $\sigma^2$ vs $H/\sigma$, the initial susceptibility $\chi_0$ above $T_c$ was obtained, which when the resistance against the relationship $\chi_0 = 1.63 \pm 0.02$ from just above $T_c$ (386.5 K) up to about 1.15 $T_c$. This $\gamma$ value contrasts with the values near $\frac{3}{4}$ recently computed for the Heisenberg model and later found experimentally in various ferromagnetic metals and compounds. At higher temperatures the effective $\gamma$ decreases rapidly towards unity. Up to the highest field used (25 kOe), the critical isotherm obeys the relationship $\gamma \propto H^{1/4}$ with $\delta = 3.75 \pm 0.05$, which differs markedly from the theoretical $\delta$ values of 3 from the 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, $\sigma/\sigma_0 = f(H/H_0)$, in which $\sigma = (T - T_c)^{1/(4+\delta)}$, 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.

**Magnetic Critical-Point Behavior of CrO$_2$**


