Partial Test of the Universality Hypothesis: The Case of Next-Nearest-Neighbor Interactions*

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High-temperature series expansions are used to examine the dependence of critical-point exponents upon the presence of second-neighbor interactions. We consider the Hamiltonian

$$\mathcal{H}_{\rm nnn} = -J_1 \sum_{\langle ij\rangle}^{\rm nn} \vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)} - J_2 \sum_{\langle ij\rangle}^{\rm nnn} \vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)},$$

where the first and second sums are over pairs of nearest-neighbor (nn) and next-nearest-neighbor (nnn) sites, and where the spins $\vec{S}^{(D)}$ are D-dimensional unit vectors. The two-spin correlation function, $C_2(\vec{r})$, is calculated to tenth, ninth, and eighth order in $1/k_BT$ for the Ising ($D=1$), classical-planar ($D=2$), and classical-Heisenberg ($D=3$) models, respectively, for various values of the parameter $R'\equiv J_2/J_1$ and for various cubic lattices (fcc, bcc, and sample cubic). These represent the first series expansions of the spin correlation function for nnn interactions. From $C_2(\vec{r})$ we obtain series for the specific heat, susceptibility, and second moment. Analysis of these series and detailed comparisons with the exactly soluble spherical model ($D=\infty$) lead us to conclude that the exponents γ (susceptibility) and ν (correlation length) may be independent of R'; this suggestion is consistent with the universality hypothesis.

I. INTRODUCTION

In this work we present evidence from series expansions germane to the question "Do critical-point exponents depend upon the range of the exchange interaction?"

One motivation for considering this question is that almost all materials in nature involve interactions that are greater than "nearest neighbors only" in range, while the great majority of theoretical calculations are restricted to the simplest, nearest-neighbors-only case. A second motivation is provided by our desire to test the universality hypothesis,[1] which predicts that for systems with interaction strengths that are finite in range[2] all critical-point exponents should assume the same values as for the case of nearest-neighbor interactions only.

To this end we consider a system with both nearest-neighbor (nn) and next-nearest-neighbor (nnn) interactions:

$$\mathcal{H}_{\rm nnn} = -J_1 \sum_{\langle ij\rangle}^{\rm nn} \vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)} - J_2 \sum_{\langle ij\rangle}^{\rm nnn} \vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)}$$

$$\equiv -J_1\Big(\sum_{\langle ij\rangle}^{\rm nn} \vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)} + R' \sum_{\langle ij\rangle}^{\rm nnn} \vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)}\Big), \quad (1.1)$$

where $R'\equiv J_2/J_1$ and J_1, J_2 denote, respectively, the nn and nnn exchange interactions. Here $\vec{S}_i^{(D)}$ and $\vec{S}_j^{(D)}$ denote isotropically interacting D-dimensional classical spins situated on sites i and j of a regular three-dimensional ($d=3$) lattice, where $D=1$, 2, 3, and ∞ correspond, respectively, to the Ising, plane-rotator (or classical-planar), classical-Heisenberg, and spherical models.

A. Previous Work

One can show rigorously that for $D=\infty$ (the spherical model) critical-point exponents are independent of the parameter R' for all values of R' (cf. Appendix A of Paper I[3]). However, aside from certain one-dimensional ($d=1$) models, there exist no exact results for finite D.

Moreover, previous *approximation* procedures leave this an open question. In fact, the most *recent* calculations[4] using the method of high-temperature series expansions suggest that the susceptibility critical-point exponent γ for the $S=\frac{7}{2}$ Heisenberg model actually varies continuously with R', at least for R' in the range $-0.2\le R'\le 2$. As the authors emphasized, however, these results were based upon the calculation of rather short series and therefore the rather marked dependence of γ upon R' might be spurious.

Indeed, a large literature does exist concerning the application of series-expansion techniques to the problem of further neighbor interactions,[4–14] and previous workers who had noticed a possible dependence of exponents upon R' were generally inclined to dismiss their results as spurious, although their reasons given were not always convincing.

Using both high- and low-temperature series expansions, Dalton and Wood[12] have extensively analyzed the Ising model ($D=1$) on two- and three-dimensional lattices ($d=2$, 3). Analysis of the low-temperature series yielded estimates for the exponents γ' and β consistent with the universality hypothesis.

From high-temperature series, Dalton and Wood concluded that, for $d=2, 3, \gamma$ remains unchanged when second and third neighbors are introduced. However, these conclusions were based only on analysis of the special case of equivalent bonds (e.g., $J_1 = J_2$ or $J_1 = J_2 = J_3$). Although it is quite plausible that invariance of exponents for this special case implies invariance for *all* values of the interaction strengths, this is by no means obvious. Furthermore, the conclusions reached were based on the following observation: Although a series of estimates, $\{\gamma_n\}$, for γ are consistently *lower* than the nearest-neighbor ($R' = 0$) values, the $\{\gamma_n\}$ are very slightly increasing—apparently toward the nearest-neighbor values. It would be interesting to see if this trend (toward the $R' = 0$ values of γ) continued with the introduction of more coefficients of the series. More importantly, it would be desirable to calculate the series for *arbitrary* J_1 and J_2 and hence study $\gamma = \gamma(R')$.

High-temperature series expansions for the next-nearest-neighbor classical Heisenberg model ($D = 3$) have been analyzed by Bowers and Woolf,[13] who also treated only the case of "equivalent bonds," $R' = 1$. We feel that their analysis, which concluded that $\gamma(R' = 1) = \gamma(R' = 0)$, was not a valid test of the universality hypothesis. Bowers and Woolf proceeded as follows. They first obtained an estimate of the critical temperature $T_c(R' = 1)$ by assuming that $\gamma(R' = 1) = \gamma(R' = 0)$. They then argued that since this critical temperature yielded consistent estimates for $\gamma(R' = 1)$ equal to $\gamma(R' = 0)$ the exponent must be independent of R'. There are two possible pitfalls in this type of argument: (i) It is not clear that there is a *unique* pair (T_c, γ) which yield consistent results and (ii) consistency in itself is not sufficient to justify the choice of a pair (T_c, γ). With regard to this second point we note that because of correction terms to pure power-low behavior a given series may yield estimates for an exponent which, while not constant, may extrapolate to the correct value for the exponent. An attempt to choose T_c so as to make the series more consistent may result in incorrect conclusions.[15]

The $S = \frac{1}{2}$ Heisenberg model with next-nearest-neighbor interactions of arbitrary strength has been considered by Dalton and Wood,[6] who obtained five terms in the expansion of the zero-field susceptibility. They analyzed the series for $0 \le R' \le 1$ and concluded that for this range of R', $\gamma \cong 1.33$. This value of γ was consistent with the work of earlier authors who had estimated $\gamma(R' = 0) \cong 1.33$, though more recent analysis of longer series has indicated larger values for $\gamma(R' = 0)$.[16]

B. Relevant Experimental Results

EuO is an insulating ferromagnet which can be represented by a $S = \frac{7}{2}$ Heisenberg model with first- and second-neighbor interactions. Early experimental investigation of this material led certain authors to conclude that $J_2/J_1 \cong -0.1$ with J_1 positive.[17] On the other hand, the recent work of Menyuk, Dwight, and Reed[4] indicated $J_2/J_1 \cong 0.5$. Furthermore, Menyuk *et al.* concluded from their measurements (using a vibrating-coil magnetometer) that $\gamma \cong 1.29$. This value disagrees both with the estimates of $\gamma(R' = 0)$ from high-temperature series expansions and with the very recent work of Als-Nielsen, Dietrich, Kunnmann, and Passell,[18] who studied EuO and also EuS ($J_2/J_1 \cong 0.4$, $S = \frac{7}{2}$) using neutron scattering. These authors concluded that for both EuO and EuS, $\gamma \cong 1.39$ in agreement with series-expansion results for $\gamma(R' = 0)$.

We feel that the present work may shed some light on the disagreements noted above. In particular, a conclusion that universality *holds* would support the results for γ of Als-Nielsen *et al.* while a conclusion that universality *breaks down* would support the result for γ of Menyuk *et al.*[19]

The longer series we obtain will also be useful because the value $J_2/J_1 \cong 0.5$ estimated by Menyuk *et al.* was obtained by comparison of their experimental data with predictions of high-temperature series which were rather short.

In Sec. III we will give a possible explanation for experimentally observed low values of γ, consistent with universality but based upon some peculiar features of the next-nearest-neighbor series we obtain.

C. Present Work

Using the methods described in Sec. ID of I we have calculated the coefficients in the high-temperature series expansion for the two-spin correlation function

$$C_2(\vec{r}) = \sum_{n=0}^{\infty} g_n(\vec{r})\, x^n \tag{1.2}$$

through order g_{10}, g_9, and g_8, respectively, for $D = 1$, 2, and 3 (Ising, planar, and Heisenberg models) for $\mathcal{H}_{\rm nnn}$ for various values of the parameter R'. Here $x \equiv 1/k_B T$. From the coefficients $g_n(\vec{r})$, series of corresponding lengths were calculated for the reduced isothermal susceptibility $\bar{\chi}_T$, for the "second moment" μ_2, and for the reduced specific heat $\bar{C}_H$. Series for $\bar{\chi}$, μ_2, and $\bar{C}_H$ are available upon request from the authors.

We also calculated 20 terms in the high-temperature series expansion of $\bar{\chi}$ and μ_2 for the exactly soluble spherical model ($D = \infty$) (cf. Appendix). This calculation will be found to play an important role in the analysis which follows.

As far as we know this is the first calculation for $\mathcal{H}_{\rm nnn}$ of $C_2(\vec{r})$ and hence μ_2. Our work also significantly extends the number of known coefficients in the series for $\bar{\chi}$ and $\bar{C}_H$ (cf. Table I).

In the limits $R' \to 0$ and $R' \to \infty$, series for the corresponding nearest-neighbor problems were generated, thereby providing a strong check on the calculation. Additional checks were carried out, and of course agreement with previous cal-

TABLE I. Comparison of number of expansion coefficients in the series obtained in the present work for $\mathcal{H}_{nnn}$ and the longest previously published series. An asterisk indicates that series were obtained only for special case, $R'=1$. The quantities $\bar{C}_H$, $\bar{\chi}$, $C_2(\vec{r})$, and $\mu_2(\vec{r})$ are defined in I in Eqs. (1.4)–(1.7).

	$\bar{C}_H$ Previous	$\bar{C}_H$ Present	$\bar{\chi}$ Previous	$\bar{\chi}$ Present	$C_2(\vec{r})$ (and thus μ_2) Previous	$C_2(\vec{r})$ (and thus μ_2) Present
Ising	8 (Ref. 14, fcc only) 6* (Ref. 8) 5 (Ref. 9)	10	7* (Ref. 8) 5 (Ref. 9)	10		10
Classical planar		9		9		9
Classical Heisenberg	5 (Ref. 9)	8	7* (Ref. 13) 6 (Ref. 11)	8		8
Spherical		20		20		20

culations was obtained in the regions of overlap.

II. ANALYSIS OF SERIES FOR ISING, PLANAR, HEISENBERG, AND SPHERICAL MODELS

We will see below that support for the universality predictions for $\mathcal{H}_{nnn}$ is less direct than the support for the predictions for $\mathcal{H}_{l\,\text{anis.}}$.[3] In particular, our arguments will depend heavily upon a comparison between the series analysis for the Ising ($D=1$), planar ($D=2$), and Heisenberg ($D=3$) models, and the analysis for the spherical model ($D=\infty$). In fact, *without this comparison* there is little to counter strong (but we think misleading) evidence for the failure of universality (i.e., for γ and ν

TABLE II. Estimates (in units of 10^{-2}) for the critical-point exponent γ from PA's to $(d/dx)\ln\bar{\chi}(x)$ for the Ising model on the sc lattice. Here and in all PA tables which follow, the notation "0" indicates that either the singularity closest to the origin was not on the positive real axis or that there were two singularities on the positive real axis very close to each other, thereby making determination of an estimate of the exponent difficult. For all three cubic lattices the estimates are decreasing with R', at least for $R' \lesssim 10$, and for certain R' there is a remarkable consistency in the estimates.

(a) γ: Ising, sc, $R'=1.00$

D \ N	1	2	3	4	5	6	7
2	125	122	122	122	123	123	123
3	122	122	123	123	123	131	
4	122	123	123	123	123		
5	122	123	123	123			
6	123	123	123				
7	123	125					
8	123						

(b) γ: Ising, sc, $R'=2.00$

D \ N	1	2	3	4	5	6	7
2	117	121	126	131	124	122	122
3	121	128	134	127	119	122	
4	126	135	128	123	122		
5	131	127	123	122			
6	124	119	122				
7	122	122					
8	122						

(c) γ: Ising, sc, $R'=5.00$

D \ N	1	2	3	4	5	6	7
2	117	113	114	115	115	115	0
3	114	114	113	116	116	130	
4	114	114	116	116	116		
5	115	116	116	116			
6	115	116	116				
7	115	150					
8	174						

(d) γ: Ising, sc, $R'=10.00$

D \ N	1	2	3	4	5	6	7
2	120	0	0	115	115	115	115
3	0	0	0	116	115	115	
4	0	0	116	115	115		
5	115	116	115	115			
6	115	115	115				
7	115	115					
8	115						

(e) γ: Ising, sc, $R'=20.00$

D \ N	1	2	3	4	5	6	7
2	122	123	121	116	117	115	113
3	122	121	123	118	117	110	
4	121	122	118	115	116		
5	116	118	115	116			
6	117	117	116				
7	115	112					
8	113						

dependent on R').

A. Padé Approximants

As with $\mathcal{H}_{l\ \mathrm{anis.}}$, the Padé approximants (PA's) for $\mathcal{H}_{\mathrm{nnn}}$ consistently indicated ferromagnetic and antiferromagnetic singularities at x_c and x_{af}, respectively. With the introduction of second-neighbor interactions, Eq. (3.1) of I holds only in the limit of loose-packed lattices, i.e., for $R' = 0$ (sc and bcc) and for $R' = \infty$ (bcc and fcc). Thus in general $|x_{af}|$ should not equal x_c. Furthermore, when J_1, J_2 are both negative the interactions are competing in determining the ordered state.[20] Thus, it follows that $T_{af} \lesssim T_c$, or

$$|x_{af}| \gtrsim x_c \tag{2.1}$$

for all R'. Equation (2.1) was verified by the PA analysis.

A sample or "cross section" of the PA estimates for γ and 2ν for the $D = 1$, 2, and 3 models is presented in Tables II–VII. We note that the estimates for $\gamma(R')$ and $2\nu(R')$ are decreasing with R' at least until $R' \cong 10$. We point out especially the consistency at $R' \cong 5$–10 [cf. Tables II(d), III(d), IV(d), V(c), VI(c), and VII(c)]. For example, from the PA's *alone* it would appear that γ(Ising, fcc, $R' = 10$) $\cong 1.10$, so that $\gamma - 1$ has decreased to less than half of the $R' = 0$ value, 0.25.

On the other hand, consider the PA's for the spherical model [cf. Table VIII and Table V(a) of I] for which $\gamma(R') = 2$ for all R'. If only 11 coefficients were known in the susceptibility series for the spherical model (so that $N + D \leq 10$ in Table VIII), we would be led to conclude from the PA analysis that for $R' = 10$, $\gamma \cong 1.31$. On examination of higher-order PA's ($10 < N + D \leq 19$), however, we see that the residues become much less consistent and are generally increasing, although on the basis of 20 coefficients it is hard to tell for sure whether the residues are in fact converging to 2. The behavior of the spherical-model PA's clearly illustrates the possibility that *we do not have enough coefficients to see asymptotic behavior* for the $D = 1$, 2, and 3 models. In Sec. II B we present stronger evidence for this possibility.

B. Park's Method and "T_c Renormalization"

We have applied Park's method to the series for $\bar{\chi}$, $\mu_2/\bar{\chi}$, and μ_2. For $R' \gtrsim 1$ on the sc lattice application of a transformation [of the type Eq. (3.26)

TABLE III. Estimates (in units of 10^{-2}) for the critical-point exponent γ from PA's to $(d/dx)\ln\bar{\chi}(x)$ for the Ising model on the bcc lattice.

(a) γ: Ising, bcc, $R' = 1.00$

D \ N	1	2	3	4	5	6	7
2	128	125	124	124	124	124	124
3	125	125	124	124	124	124	
4	124	124	124	124	124		
5	125	124	124	124			
6	124	124	124				
7	124	124					
8	124						

(b) γ: Ising, bcc, $R' = 2.00$

D \ N	1	2	3	4	5	6	7
2	118	123	123	124	124	124	124
3	123	124	124	124	124	125	
4	123	124	124	124	125		
5	124	124	124	124			
6	124	124	124				
7	124	144					
8	124						

(c) γ: Ising, bcc, $R' = 5.00$

D \ N	1	2	3	4	5	6	7
2	116	117	118	120	121	121	122
3	117	118	124	121	123	124	
4	118	124	120	122	124		
5	120	121	122	122			
6	121	123	125				
7	121	124					
8	122						

(d) γ: Ising, bcc, $R' = 10.00$

D \ N	1	2	3	4	5	6	7
2	117	115	116	117	117	118	118
3	115	116	116	117	117	117	
4	116	115	117	117	117		
5	117	117	118	0			
6	117	117	117				
7	118	117					
8	118						

(e) γ: Ising, bcc, $R' = 20.00$

D \ N	1	2	3	4	5	6	7
2	119	117	117	117	116	115	116
3	117	117	117	117	115	116	
4	117	117	117	117	116		
5	117	117	117	115			
6	116	115	116				
7	116	116					
8	116						

of I] is not necessary because $|x_{af}| \gg x_c$ (the sc lattice reduces to an fcc lattice for $R' \to \infty$). Transformations were performed on the series for the bcc and fcc lattices for which $|x_{af}| \cong x_c$ for large R'.

Consider first the exponent γ and the sc lattice for which no transformation need be performed [cf. Figs. 1(a)–1(c)]. For $R' = 1$, 2, the estimates γ_n have an *upward* trend, possibly extrapolating to the $R' = 0$ values at $n = \infty$. For $R' = 10$, 20, however, there is a *downward* trend with no indication that the series will bend up again. The only positive statement we can make is that whatever is happening for $D = 1$ is clearly happening for $D = 2$ and 3. Similar behavior is observed for other lattices even after transforming the original series (cf. Fig. 2), for the exponent ν [cf. Figs. 3(a) and 3(b)] and using other methods of analysis [cf. Figs. 3(b) and 3(c)].

Consider now the spherical model [Fig. 1(d)]. The general behavior of the first 8–10 estimates is *exactly* the same as for the Ising, planar, and Heisenberg models.[21] The only quantitive difference in the behavior of the series for the $D = 1$, 2, 3, and ∞ models seems to be the actual value of the exponents. We now discuss what can be inferred from this similarity.

C. Conclusions about $\gamma(R')$ and $\nu(R')$

We have seen above a striking similarity between the series analyses for the $D = 1$, 2, 3, and ∞ models.[21] On the basis of this similarity and the fact that $\gamma_{\text{spherical}}(R') = \text{const}$, we speculate that the predictions of universality hold for $\mathcal{H}_{\text{nnn}}$ for $D = 1$, 2, 3 (and probably for all D). That is, we suggest that the series which indicated a downward trend in the estimates for γ and ν will eventually show a bending up to the $R' = 0$ values upon the introduction of a sufficient number of higher-order coefficients.

What does puzzle us is why the series should show such great curvature for $R' \gg 1$ in light of the fact that as $R' \to \infty$ each cubic-type lattice reduces to another cubic-type lattice, all of which are believed to have equal exponents. If any curvature should be present at all, we might have expected it to be greatest near the "symmetrical point" $R' = 1$.[22]

III. SUMMARY

A. Conclusions for Exponents

We have generated what we believe are the first

TABLE IV. Estimates (in units of 10^{-2}) for the critical-point exponent γ from PA's to $(d/dx)\ln\overline{\chi}(x)$ for the Ising model on the fcc lattice.

(a) γ: Ising, fcc, $R' = 1.00$

D \ N	1	2	3	4	5	6	7
2	125	121	123	122	122	122	123
3	121	122	122	122	123	122	
4	123	122	122	124	123		
5	122	122	124	123			
6	122	123	123				
7	123	122					
8	123						

γ: Ising, fcc, $R' = 2.00$

D \ N	1	2	3	4	5	6	7
2	113	116	116	119	118	120	126
3	116	123	121	120	121	121	
4	116	121	120	120	120		
5	119	120	120	121			
6	142	120	120				
7	120	121					
8	123						

(c) γ: Ising, fcc, $R' = 5.00$

D \ N	1	2	3	4	5	6	7
2	111	110	111	113	113	114	115
3	110	111	0	117	119	119	
4	111	121	114	118	119		
5	113	117	119	119			
6	113	119	119				
7	115	119					
8	115						

(d) γ: Ising, fcc, $R' = 10.00$

D \ N	1	2	3	4	5	6	7
2	113	111	110	110	110	111	111
3	111	110	110	110	110	110	
4	110	110	110	110	110		
5	110	110	110	110			
6	110	110	110				
7	111	110					
8	111						

(e) γ: Ising, fcc, $R' = 20.00$

D \ N	1	2	3	4	5	6	7
2	116	113	112	112	111	110	110
3	109	112	112	113	109	110	
4	112	112	112	0	110		
5	112	113	0	109			
6	111	109	110				
7	110	110					
8	110						

high-temperature series expansions for the two-spin correlation function for the Ising, classical-planar, classical-Heisenberg, and spherical models of magnetism ($D=1$, 2, 3, and ∞) with next-nearest-neighbor interactions. We have also significantly extended the series for the zero-field

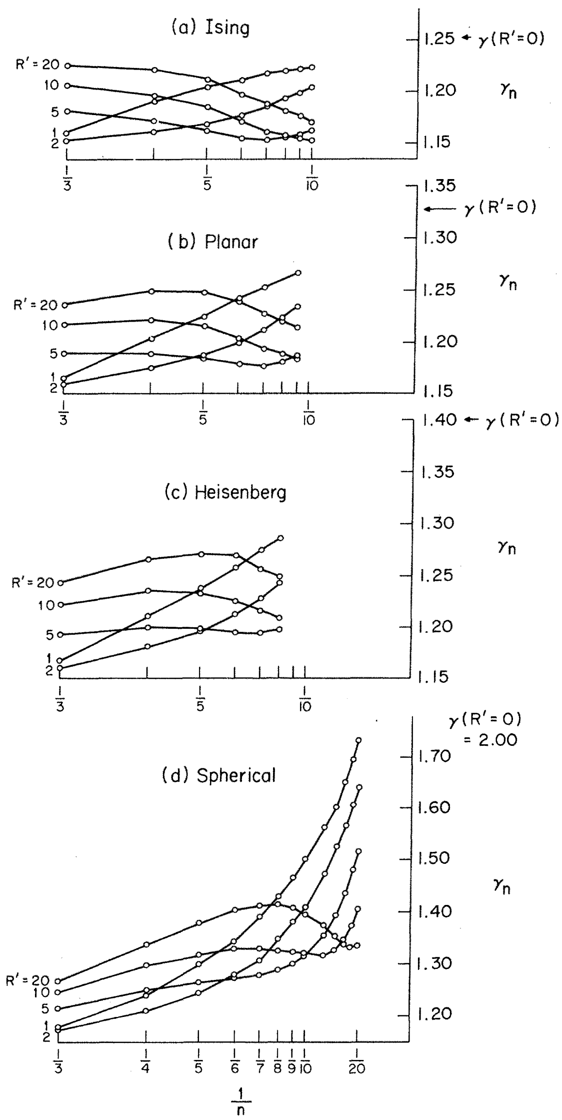

FIG. 1. Estimates for γ from Park's method for the (a) Ising [$\gamma(R'=0)\cong 1.25$], (b) classical-planar [$\gamma(R'=0)\cong 1.33$], (c) classical-Heisenberg [$\gamma(R'=0)\cong 1.40$], and (d) spherical [$\gamma(R'=0)=2.00$] models on the sc lattice. We note the similar behavior for all four models. The reader should note that later terms of the series for $R'=1$, 2, and 5 indicate a "turning up" to larger values of γ. Moreover, this bending occurs at *larger* order n for *larger* values of R', suggesting that perhaps a similar turning up might occur for very large R' ($R'=20$, for example) if a sufficiently large number of terms in the series were available. This *must* occur in the spherical model for which γ is rigorously independent of R'.

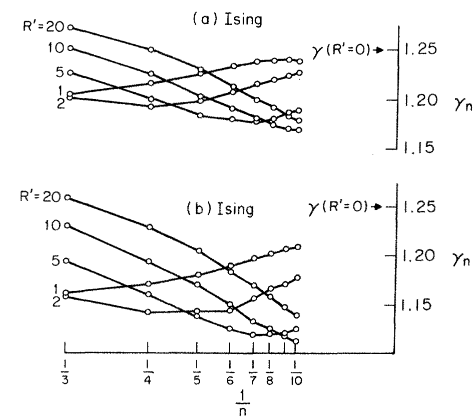

FIG. 2. Estimates for γ from Park's method applied to Ising model series for the (a) bcc and (b) fcc lattices. The series were first transformed to reduce the effects of antiferromagnetic singularities. By comparison with Fig. 1(a) we see that the behavior of the estimates appears to be lattice independent.

isothermal susceptibility and the specific heat for these models.

Straightforward analyses using a number of different techniques indicate that the exponents γ and ν are decreasing with the parameter R' at least for $R'\lesssim 10$. However, comparison with similar anal-

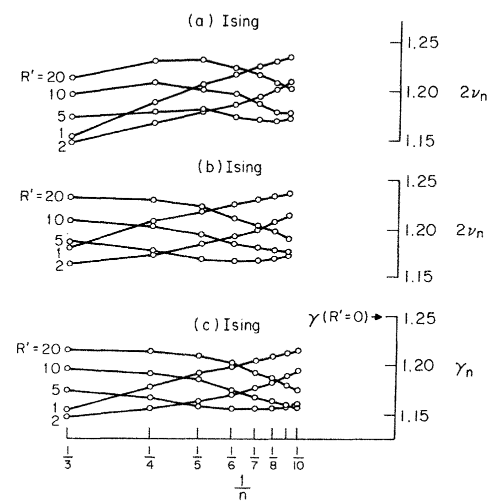

FIG. 3. Ising model, sc lattice. (a) and (b) Estimates of 2ν from application of Park's method and "T_c renormalization," respectively; (c) estimates for γ from a variation [H. E. Stanley, Phys. Rev. 158, 546 (1967)] of the ratio method in which $\gamma_n = 1 - n[1-\rho_n(x_c)_n]$ [cf. Eq. (2.8) of I], where $(x_c)_n$ is found from Eq. (2.7) of I. The similar behavior for the estimates in (a)–(c) and in Figs. 1 and 2 indicates that the general behavior noted in Fig. 1 is not confined to the exponent γ, to a specific lattice, or to a specific method of analysis.

TABLE V. Estimates (in units of 10^{-2}) for the critical-point exponent γ from PA's to $(d/dx)\ln\bar{\chi}(x)$ for the planar model on the sc lattice. We see the same decrease with R' in the estimates for γ as seen for the estimates for the Ising model.

(a) γ: Planar, sc, $R'=1.00$

D \ N	1	2	3	4	5	6
2	132	125	127	127	128	128
3	125	126	127	128	128	
4	127	127	128	128		
5	127	128	128			
6	128	128				
7	128					

(b) γ: Planar, sc, $R'=2.00$

D \ N	1	2	3	4	5	6
2	120	122	130	134	129	127
3	122	0	135	132	121	
4	131	135	126	126		
5	134	132	126			
6	129	122				
7	127					

(c) γ: Planar, sc, $R'=5.00$

D \ N	1	2	3	4	5	6
2	119	119	107	118	118	118
3	119	115	118	118	119	
4	110	118	118	118		
5	118	118	118			
6	118	119				
7	118					

(d) γ: Planar, sc, $R'=10.00$

D \ N	1	2	3	4	5	6
2	122	122	122	116	118	118
3	122	122	120	118	118	
4	122	120	119	117		
5	117	118	117			
6	118	118				
7	118					

(e) γ: Planar, sc, $R'=20.00$

D \ N	1	2	3	4	5	6
2	125	125	125	111	120	118
3	125	125	123	121	119	
4	125	123	121	109		
5	115	121	112			
6	120	119				
7	118					

TABLE VI. Estimates (in units of 10^{-2}) for the critical-point exponent γ from PA's to $(d/dx)\ln\bar{\chi}(x)$ for the classical-Heisenberg model.

(a) γ: Heisenberg, sc, $R'=1.00$

D \ N	1	2	3	4	5
2	138	128	131	131	132
3	128	130	131	0	
4	131	131	132		
5	131	0			
6	132				

(b) γ: Heisenberg, sc, $R'=2.00$

D \ N	1	2	3	4	5
2	122	124	135	138	134
3	124	124	139	136	
4	135	139	133		
5	139	136			
6	134				

(c) γ: Heisenberg, sc, $R'=5.00$

D \ N	1	2	3	4	5
2	120	120	120	119	120
3	120	116	120	120	
4	120	120	120		
5	119	120			
6	120				

(d) γ: Heisenberg, sc, $R'=10.00$

D \ N	1	2	3	4	5
2	124	123	124	114	120
3	123	124	122	120	
4	124	122	121		
5	117	120			
6	120				

(e) γ: Heisenberg, sc, $R'=20.00$

D \ N	1	2	3	4	5
2	128	127	127	127	121
3	127	128	125	123	
4	127	125	124		
5	0	123			
6	122				

TABLE VII. Estimates (in units of 10^{-2}) for the critical-point exponent 2ν from the PA's to $(d/dx)\,[\ln x^{-1}\mu_2(x)/\bar{\chi}(x)]$ for the Ising model on the sc lattice. These estimates show the same decrease with R' as the estimates for γ.

(a) 2ν: Ising, sc, $R'=1.00$

$D \backslash N$	1	2	3	4	5	6
2	130	122	124	124	124	124
3	122	124	124	124	124	
4	124	124	124	127		
5	124	124	127			
6	124	125				
7	124					

(b) 2ν: Ising, sc, $R'=2.00$

$D \backslash N$	1	2	3	4	5	6
2	120	119	125	124	124	123
3	119	120	124	124	124	
4	125	124	124	124		
5	124	124	125			
6	124	125				
7	123					

(c) 2ν: Ising, sc, $R'=5.00$

$D \backslash N$	1	2	3	4	5	6
2	119	118	20	117	117	117
3	118	117	118	118	118	
4	85	118	118	118		
5	117	118	118			
6	117	117				
7	117					

(d) 2ν: Ising, sc, $R'=10.00$

$D \backslash N$	1	2	3	4	5	6
2	122	121	122	116	116	117
3	121	118	119	116	116	
4	122	119	118	117		
5	117	116	117			
6	116	116				
7	117					

(e) 2ν: Ising, sc, $R'=20.00$

$D \backslash N$	1	2	3	4	5	6
2	125	124	124	115	115	116
3	124	120	121	115	115	
4	124	121	121	116		
5	118	115	116			
6	115	116				
7	116					

yses for the exactly soluble spherical model [for which $\gamma(R')=2$ for all R'] leads us to put forth the hypothesis that this decrease is probably spurious and would disappear if more terms in the series were known. We thus conclude from this *indirect* evidence that the predictions of universality are

TABLE VIII. Estimates (in units of 10^{-2}) for the critical-point exponent γ from PA's to $(d/dx)\,[\ln\bar{\chi}(x)]$ for the spherical model on the sc lattice with $R'=10$. For $N+D \lesssim 10$ the estimates for γ are consistently ~ 1.3. For larger values of $N+D$ the estimates are generally increasing although it is not clear that the estimates are converging to the known exact value of γ, 2.0.

γ: Spherical model, sc lattice, $R'=10.00$

$D \backslash N$	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	125	130	132	133	133	133	133	132	132	132	132	133	134	135	136	137	139	134
2	137	135	134	133	133	0	131	132	132	132	131	131	130	138	0	0	0	
3	135	126	132	130	131	132	132	133	134	136	140	146	153	159	165	170		
4	134	132	132	131	130	133	136	158	165	166	171	175	177	179	181			
5	133	131	131	131	133	135	0	165	166	165	184	181	182	185				
6	134	131	130	133	138	143	160	166	165	0	180	181	0					
7	127	132	133	135	144	0	169	178	179	181	181	248						
8	131	132	136	0	161	169	173	179	178	212	0							
9	132	133	162	171	167	180	179	182	162	0								
10	132	135	170	168	169	179	178	193	89									
11	132	138	168	169	166	182	175	144										
12	131	144	171	198	180	182	133											
13	131	151	175	181	179	206												
14	131	158	178	182	0													
15	143	164	180	185														
16	0	169	181															
17	0	173																
18	0																	

correct for the Ising, classical-planar, and classical-Heisenberg models. This conclusion is in agreement with the conclusions of most other authors (cf. Sec. I B) who analyzed $\mathcal{H}_{nnn}$ using shorter series, primarily for the special case $R'=1$. Furthermore, assuming the spin independence of exponents our work would indicate that the decrease in γ observed by Menyuk *et al.*[4] for the $S=\frac{7}{2}$ Heisenberg model is also spurious and is related to the shortness of the series that they analyzed.

The series for $\overline{C}_H$ were not regular enough to permit reliable predictions for the exponent α.

B. Relation with Experiment

In Sec. I B and in I we discussed certain experiments, the results of which would indicate a *possible* breakdown in universality. While our high-temperature series analysis leads us to believe that universality *is* obeyed, it also gives us one possible reason for the disagreements between theory and experiment noted above. We note that for $\mathcal{H}_{l\,\text{anis.}}$ and $\mathcal{H}_{mn}$ there were ranges of values for the parameters R and R', respectively, for which the series exhibited considerable curvature; there was so much curvature, in fact, that a superficial analysis might lead to incorrect predictions for exponents. We feel that a similar phenomenon may be affecting experiments to determine exponents.

Because experiments cannot actually get to temperatures arbitrarily close to T_c, what is actually measured is a temperature-dependent exponent γ^* defined through[23]

$$\gamma^* \equiv (T-T_c)\frac{d}{dT}\ln\chi^{-1}\ , \tag{3.1}$$

which has the property that

$$\lim_{T\to T_c}\gamma^*(T)=\gamma\ . \tag{3.2}$$

If the series expansion for χ exhibits much curvature, then the experimentally measured $\gamma^*(T)$ will do so also. This can be seen, for example, by calculating $\gamma^*(T)$ for the model function in Eq. (2.40) of I. Here we find

$$\gamma^*(T)=a-\frac{b}{AR}\,\epsilon^b+(\text{higher-order terms in }\epsilon)\ . \tag{3.3}$$

In order to measure the correct value for γ we must have

$$\frac{b}{aAR}\ll 1\ , \tag{3.4}$$

which implies for $b\sim 1$ that ϵ must be 10 times as small for $R=0.1$ as for $R=1.0$ (cf. Sec. II F of I).

We thus see that when there is considerable curvature in a series, not only are the series analyses likely to yield incorrect estimates but experimental investigations are likely to do so also. We are by no means claiming that this is *the* reason for the disagreement between theory and experiment; we present it only as one possibility.

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APPENDIX: SELECTED SERIES FOR THE SPHERICAL MODEL

Coefficients in the spherical-model susceptibility series for selected values of $R'\equiv J_2/J_1$ are listed below; shown are the first 20 terms for $R'=0$, 1, 2, 5, 10, and 20. An arbitrary number of terms can be straightforwardly calculated using methods explained in Appendix A of Paper I.[3]

	Spherical Model on sc Lattice: Susceptibility		
	$J_1=1.00,\ J_2=0.00$	$J_1=1.00,\ J_2=1.00$	$J_1=0.50,\ J_2=1.00$
0	0.100 000 000 0D 01	0.100 000 000 0D 01	0.100 000 000 0D 01
1	0.600 000 000 0D 01	0.180 000 000 0D 02	0.150 000 000 0D 02
2	0.300 000 000 0D 02	0.306 000 000 0D 03	0.211 500 000 0D 03
3	0.144 000 000 0D 03	0.506 400 000 0D 04	0.290 400 000 0D 04
4	0.666 000 000 0D 03	0.823 500 000 0D 05	0.392 456 250 0D 05
5	0.302 400 000 0D 04	0.132 249 600 0D 07	0.524 659 500 0D 06
6	0.134 760 000 0D 05	0.210 366 360 0D 08	0.695 747 756 2D 07
7	0.593 280 000 0D 05	0.332 097 840 0D 09	0.916 757 932 5D 08
8	0.258 354 000 0D 06	0.521 035 594 2D 10	0.120 167 546 5D 10
9	0.111 585 600 0D 07	0.813 250 818 2D 11	0.156 823 726 4D 11
10	0.478 450 800 0D 07	0.126 378 992 0D 13	0.203 893 421 2D 12

TABLE. (*Continued*)

Spherical Model on sc Lattice: Susceptibility			
	$J_1=1.00,\ J_2=0.00$	$J_1=1.00,\ J_2=1.00$	$J_1=0.50,\ J_2=1.00$
11	0.203 938 560 0*D* 08	0.195 650 811 4*D* 14	0.264 226 907 4*D* 13
12	0.864 735 480 0*D* 08	0.301 893 941 5*D* 15	0.341 433 757 9*D* 14
13	0.365 034 816 0*D* 09	0.464 475 829 6*D* 16	0.440 081 655 7*D* 15
14	0.153 482 796 0*D* 10	0.712 767 454 4*D* 17	0.565 946 375 0*D* 16
15	0.643 100 083 2*D* 10	0.109 125 324 0*D* 19	0.726 326 009 7*D* 17
16	0.268 622 284 5*D* 11	0.166 722 778 3*D* 20	0.930 435 813 5*D* 18
17	0.111 891 970 5*D* 12	0.254 237 919 0*D* 21	0.118 990 413 7*D* 20
18	0.464 902 263 4*D* 12	0.387 020 784 3*D* 22	0.151 939 509 4*D* 21
19	0.192 724 355 2*D* 13	0.588 220 074 2*D* 23	0.193 739 003 7*D* 22
20	0.797 276 776 9*D* 13	0.892 713 457 0*D* 24	0.246 716 796 4*D* 23
	$J_1=0.20,\ J_2=1.00$	$J_1=0.10,\ J_2=1.00$	$J_1=0.05,\ J_2=1.00$
0	0.100 000 000 0*D* 01	0.100 000 000 0*D* 01	0.100 000 000 0*D* 01
1	0.132 000 000 0*D* 02	0.126 000 000 0*D* 02	0.123 000 000 0*D* 02
2	0.162 000 000 0*D* 03	0.146 700 000 0*D* 03	0.139 275 000 0*D* 03
3	0.192 595 200 0*D* 04	0.164 774 400 0*D* 04	0.151 711 800 0*D* 04
4	0.224 814 816 0*D* 05	0.181 178 586 0*D* 05	0.161 403 881 6*D* 05
5	0.259 340 820 5*D* 06	0.196 448 243 0*D* 06	0.168 998 758 2*D* 06
6	0.296 720 481 3*D* 07	0.210 937 092 8*D* 07	0.174 947 323 2*D* 07
7	0.337 435 195 5*D* 08	0.224 888 917 8*D* 08	0.179 578 309 6*D* 08
8	0.381 937 794 7*D* 09	0.238 476 815 4*D* 09	0.183 139 318 9*D* 09
9	0.430 675 527 3*D* 10	0.251 826 667 0*D* 10	0.185 821 601 4*D* 10
10	0.484 104 896 2*D* 11	0.265 031 549 5*D* 11	0.187 775 535 6*D* 11
11	0.542 702 097 0*D* 12	0.278 161 085 6*D* 12	0.189 120 906 2*D* 12
12	0.606 971 006 3*D* 13	0.291 267 806 5*D* 13	0.189 954 098 2*D* 13
13	0.677 449 763 1*D* 14	0.304 391 627 0*D* 14	0.190 353 323 4*D* 14
14	0.754 716 549 9*D* 15	0.317 563 067 8*D* 15	0.190 382 520 4*D* 15
15	0.839 394 954 9*D* 16	0.330 805 613 2*D* 16	0.190 094 321 7*D* 16
16	0.932 159 163 4*D* 17	0.344 137 458 7*D* 17	0.189 532 344 2*D* 17
17	0.103 373 915 4*D* 19	0.357 572 815 9*D* 18	0.188 732 976 4*D* 18
18	0.114 492 602 7*D* 20	0.371 122 893 5*D* 19	0.187 726 783 3*D* 19
19	0.126 657 756 2*D* 21	0.384 796 637 1*D* 20	0.186 539 618 8*D* 20
20	0.139 962 408 6*D* 22	0.398 601 288 1*D* 21	0.185 193 509 4*D* 21

[*]For the values of the exact series coefficients, the reader may order document NAPS 01762 from Asis-National Auxiliary Publications Service, c/o CCM Information Corporation, 866 Third Ave., N. Y., N. Y. 10022, remitting $2.00 for each microfiche or $5.00 for each photocopy.

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[15]Remarks similar to these have been made concerning the Bowers-Woolf procedure by M. Ferer, M. A. Moore,

and M. Wortis [Phys. Rev. B 4, 3954 (1971)] in a rather different context, namely, in connection with their attempt to answer the question of what is $\gamma(R'=0)$ for the case $D=3$. Ferer *et al.* conclude that γ is 1.405 ± 0.02, a value somewhat larger than the Bowers-Woolf estimate $\gamma=1.375\pm 0.002$, but closer to the $S=\frac{1}{2}$ estimate of $\gamma=1.43\pm 0.01$ of Baker *et al.* (Ref. 16).

[16]G. A. Baker, Jr., H. E. Gilbert, J. Eve, and G. S. Rushbrooke [Phys. Rev. 164, 800 (1967)] estimate $\gamma(R'=0)=1.43\pm 0.01$ for $S=\frac{1}{2}$; see also M. H. Lee and H. E. Stanley [Phys. Rev. B 4, 1613 (1971)] who estimate $\gamma(R'=0)=1.36\pm 0.04$.

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[19]If one accepts the spin independence of critical-point exponent γ, if one believes that the values for all S are equal to the $S=\frac{1}{2}$ value, and if one accepts the estimate of Baker *et al.* (Ref. 16) for $S=\frac{1}{2}$, then both the Menyuk *et al.* (Ref. 4) and the Als-Nielsen *et al.* (Ref. 18) values are somewhat low. Hence one need not conclude that a violation of universality favors the Menyuk value or the Als-Nielsen value.

[20]When J_1 and J_2 are allowed to vary arbitrarily between $-\infty$ and $+\infty$, we find domains where the competing interactions affect the nature of the state to which the system orders. This state is determined by the precise *type* of lattice structure (fcc, bcc, sc, . . .) as well as by the values of J_1 and J_2. The ordered state has been studied heretofore by Green's-function methods (and, of course, by mean-field approaches); the application of high-temperature series-expansion methods to this problem is the subject of another work just completed. In any case, the universality hypothesis predicts that the exponent for the appropriate diverging staggered susceptibility would be the same as for the case of $R'=0$ and J_1 positive. See G. Paul and H. F. Stanley (unpublished).

[21]The similarity between the series for $D=1$, 2, 3, and ∞ has been observed by the authors from detailed comparison of numerous plots; the reader can obtain a simple impression by masking out points in Figs. 1(a)–1(d) with $n>8$ (and, if he wishes, with $n>9$ and then with $n>10$).

[22]That is, if we reformulate the Hamiltonian as $\mathcal{H}=-J_2(\mathcal{H}_{nnn}+R''\mathcal{H}_{nn})$, with $R''\equiv J_1/J_2$, then we find that the apparent change of exponents with R'' is much larger than the apparent change (in the original problem) with R'.

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Effects of Weak Covalency in Iron Fluoride Salts*

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The configuration-interaction method is utilized to investigate the effects of weak covalency on the crystal-field splittings, the g factors, the spin Hamiltonian, the spin-orbit factors, and the nuclear-quadrupole splitting in the salts FeF_2 and $KFeF_3$. Recent x-ray data for FeF_2 allow predictions to be made concerning the pressure dependence of the above-mentioned parameters in that salt. In addition, predictions are made for the pressure dependence of the Néel temperature and the saturation ($T=0$) value of the magnetic-hyperfine field based upon the calculated pressure dependence of the spin-Hamiltonian parameters for FeF_2.

INTRODUCTION

The effects of weak covalency have been observed in transition-metal salts for many years. Even in the highly electronegative fluoride salts one observes significant charge transfers. As has been shown previously,[1-5] these covalency effects must be taken into account if one expects to deal with the problem of calculating atomic parameters such as the crystal-field splittings, g factors, the spin Hamiltonian, etc. In addition, certain nuclear parameters (i.e., electric-quadrupole and magnetic-hyperfine splittings and the isomer shift) are coupled to the charge environment of the nucleus and are thereby affected by the covalent bond.

In the ensuing sections we investigate, respectively, the crystal-field splittings for $KFeF_3$ and FeF_2 (ionic and covalent), the spin Hamiltonian (including covalent reduction), and the Fe^{57} nuclear-quadrupole splitting and magnetic-hyperfine field.

CRYSTAL-FIELD SPLITTINGS

The formalism utilized here (configuration interaction) was developed by Hubbard, Rimmer, and Hopgood[4] (HRH) in a first-principles treatment of the crystal-field splittings and the transferred hyperfine field in the perovskite salts $KNiF_3$ and $KMnF_3$. In order to effect this variational calculation, HRH assume a trial wave function of the form

$$\psi=\sum_i \xi_i\,|\,i\rangle+\sum_\epsilon\sum_j\sum_k \alpha^\epsilon_{jk}\,|\,jk\epsilon\rangle\,, \tag{1}$$

where the $|\,\rangle$'s are representative of determinantal wave functions with ξ_i and α^ϵ_{jk} being the appropriate mixing coefficients. Here the basis set will con-