Critical-Point Behavior of Classical Heisenberg Ferrimagnets

SMITH FREEMAN AND PETER J. WOJTOWICZ
RCA Laboratories, Princeton, New Jersey 08540
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This paper discusses in detail the critical properties of a class of particularly simple ferrimagnets typified by simple cubic and body-centered cubic lattices having alternate sites occupied by classical spins of different magnitudes, S_A and S_B. Heisenberg exchange interactions are assumed to act between nearest-neighbor sites. For such ferrimagnets, general arguments are presented which determine certain features of the dependence of the susceptibility χ and the specific heat on the variables $R=S_B/S_A$ and $j=S_AS_BJ/\kappa T$. It is easily shown that the critical point occurs at a fixed $|j|$ for either sign of the exchange and arbitrary spin values. Moreover, the exponent γ of the singularity in χ has a value independent of the sign of the exchange and of the spin values. The only exception permitted occurs at the singularity of the simple antiferromagnet ($R=1, j<0$), which has a reduced critical exponent. The nature of the dependence of χ on R permits one to obtain its expansion in powers of j from the known expansion coefficients for the related ferromagnet. A Padé-approximant study of χ for a range of R values is described. For $R\neq 1$, the dominant singularity for positive and negative j shows the familiar power-law behavior and is consistent with the general features of the R dependence described above. The critical points and exponents are estimated for the cases studied by a new method making use of the freedom to vary R in the ferrimagnet. A method of characterizing the weaker singularities in χ is described and investigated numerically. The implications of these results for real ferrimagnets are examined.

I. INTRODUCTION

IN recent years there has been considerable effort devoted to the study of the critical properties of Heisenberg ferromagnets. Relatively little comparable work has been on Heisenberg ferrimagnets despite the great physical interest of such systems. We are currently engaged in a study of the critical properties of Heisenberg ferrimagnets based on the first six coefficients in the high-temperature susceptibility series. We have noticed, however, that certain striking simplifications occur when one passes to the classical limit within a class of especially simple lattices. Rigorous statements can then be made concerning the manner in which the susceptibility, χ, depends on the ratio of the spin magnitudes involved. In particular, the coefficients in the series expansion of χ are simply related to those for the ferromagnetic problem. This feature has enabled us to obtain the first eight coefficients for ferrimagnets based on the simple and body-centered cubic lattices from those for the analogous ferromagnets published by Wood and Rushbrooke.[1]

The classical Heisenberg ferrimagnet should provide a useful preview of the properties of the quantum system. A close relation is assured by the correspondence principle, which states that the quantum system goes over into the classical in the limit of large spin. Indeed, results obtained for the classical cubic Heisenberg ferromagnets by Wood and Rushbrooke,[1] Stanley and Kaplan[2,3] and Stanley[4] do demonstrate a close similarity in the critical behavior of classical and quantum Heisenberg systems.

In this paper, we discuss the critical properties of a particular type of classical Heisenberg ferrimagnet which is based on a lattice composed of two alternate, crystallographically equivalent sublattices. The two sublattices are occupied by classical spins having different magnitudes S_A and S_B. The spins interact via nearest-neighbor Heisenberg exchange. With these specializations, it turns out that the heat capacity in zero magnetic field, C_H, is a function of j^2 alone, where $j=S_AS_BJ/\kappa T$. Thus the magnitudes of the spins may be regarded as simply scaling the temperature dependence; in terms of the variable j they have no affect at all. The susceptibility χ is a function of the same scaled variable j; its only additional dependence on $R=S_B/S_A$, the spin ratio, is a factor $2R/(1+R^2)$ which weights the part of χ which is odd in j. These special forms of χ and C_H permit us to conclude that the critical points and critical exponents are independent of R and are symmetric between positive and negative values of the exchange constant J. There is only a single exception. The critical exponent for the susceptibility γ is decreased at the antiferromagnetic singularity ($R=1$ and $J<0$). For all other values of R and both signs of J, the value of γ is identical to that appropriate to the ferromagnet ($R=1$ and $J>0$). These results are established in Sec. II.

In Sec. III, we report the results of a Padé approximant study of the critical properties of χ for ferrimagnets based on the simple and body-centered cubic lattices. The coefficients of the high-temperature series were readily available through trivial modification of the values for the ferromagnetic series presented by Wood and Rushbrooke.[1] The features derived in Sec. II, in particular the constancy of the critical points and exponents, have been verified for these lattices in this approximation. Moreover, it has proved possible to use the invariance of γ with R to obtain an improved

[1] P. J. Wood and G. S. Rushbrooke, Phys. Rev. Letters **17**, 307 (1966).
[2] H. E. Stanley and T. A. Kaplan, Phys. Rev. Letters **16**, 981 (1966).
[3] H. E. Stanley and T. A. Kaplan, J. Appl. Phys. **38**, 977 (1967).
[4] H. E. Stanley, Phys. Rev. **158**, 546 (1967).

numerical estimate of its value. In this fashion, the following values of γ are obtained: 1.345 for the bcc and 1.385 for the sc lattices.

In Sec. IV, the relevance of our results to physical ferrimagnets is considered. A preliminary account of this research was presented at the International Congress on Magnetism.[5]

II. FORMAL STATISTICAL MECHANICS

We shall be concerned with a ferrimagnet based on a lattice in which each site belongs to one of two crystallographically equivalent sublattices, "A" and "B." Each nearest neighbor of an A site is a B site and vice versa. Each A site is occupied by a spin S_A and each B site by a different spin S_B. We assume the spins to interact via Heisenberg exchange terms between nearest neighbor spins on alternate sublattices. The Hamiltonian is then

$$\begin{aligned} \mathcal{H}&=-2JS_AS_B\mathbf{P}-g\mu H\mathbf{Q}\,,\\ \mathbf{P}&=(S_AS_B)^{-1}\sum_{\langle i,j\rangle}\mathbf{S}_i^{(A)}\cdot\mathbf{S}_j^{(B)}\,,\\ \mathbf{Q}&=S_AQ_A+S_BQ_B\,,\\ Q_A&=S_A^{-1}\sum_i S_{iz}^{(A)}\,,\\ Q_B&=S_B^{-1}\sum_j S_{jz}^{(B)}\,. \end{aligned}\tag{1}$$

If now we assume the spins to be classical, then

$$\begin{aligned} \mathbf{P}&=\sum_{\langle i,j\rangle}\mathbf{n}_i^{(A)}\cdot\mathbf{n}_j^{(B)}\,,\\ Q_A&=\sum_i n_{iz}^{(A)}\,,\\ Q_B&=\sum_j n_{jz}^{(B)}\,. \end{aligned}\tag{2}$$

The vectors $\mathbf{n}$ are ordinary unit vectors.

The susceptibility for zero magnetic field is

$$\chi=\kappa T(\partial^2/\partial H^2)\ln[\mathrm{Tr}\{\exp(-\beta\mathcal{H})\}]|_{H=0}\,,\tag{3}$$

where κ is Boltzmann's constant. The heat capacity for zero magnetic field is

$$C_H=(\partial/\partial T)\kappa T^2(\partial/\partial T)\ln[\mathrm{Tr}\{\exp(-\beta\mathcal{H})\}]|_{H=0}\,.\tag{4}$$

The trace appearing in these expressions is simply an integration over all possible directions of all unit vectors $\mathbf{n}_i^{(A)}$, $\mathbf{n}_j^{(B)}$. We shall write

$$\begin{aligned} \chi&=(S_A^2+S_B^2)(Ng^2\mu^2/3\kappa T)\hat{\chi}(j)\,,\\ C_H&=(4zN\kappa/3)j^2\hat{C}_H(j)\,, \end{aligned}\tag{5}$$

where $j=S_AS_BJ/\kappa T$. The region $j<0$ has, as usual, the physical interpretation of describing systems with antiferromagnetic exchange, $J<0$. The factors multiplying $\hat{\chi}$ and $\hat{C}_H$ are chosen so that these functions are unity for $j=0$. The total number of A or B sites present is N while z is the number of nearest neighbors. In general, we shall mean $\hat{\chi}$ or $\hat{C}_H$ when we speak of the susceptibility or heat capacity, respectively. These quantities may now be expressed in the form

$$\begin{aligned} \hat{\chi}&=\frac{\mathrm{Tr}\{\mathbf{Q}^2\exp(-2j\mathbf{P})\}\ \mathrm{Tr}\{1\}}{\mathrm{Tr}\{\exp(-2j\mathbf{P})\}\ \mathrm{Tr}\{\mathbf{Q}^2\}}\,,\\ \hat{C}_H&=\frac{\mathrm{Tr}\{\mathbf{P}^2\exp(-2j\mathbf{P})\}\ \mathrm{Tr}\{1\}}{\mathrm{Tr}\{\exp(-2j\mathbf{P})\}\ \mathrm{Tr}\{\mathbf{P}^2\}}\,. \end{aligned}\tag{6}$$

Since the traces involve an integration over the directions of the spins on the lattice sites, their value is unchanged by the substitution $\mathbf{n}_i\to-\mathbf{n}_i$. Thus, only that part of the integrand even in unit vectors referring to a particular sublattice gives a nonzero contribution. Therefore,

$$\begin{aligned} \hat{\chi}&=3N^{-1}(S_A^2+S_B^2)^{-1}\,\mathrm{Tr}\{(S_A^2Q_A^2+S_B^2Q_B^2)\cosh(2j\mathbf{P})\\ &\qquad+2S_AS_BQ_AQ_B\sinh(2j\mathbf{P})\}/\mathrm{Tr}\{\cosh(2j\mathbf{P})\}\,,\\ \hat{C}_H&=3(Nz)^{-1}\,\mathrm{Tr}\{\mathbf{P}^2\cosh(2j\mathbf{P})\}/\mathrm{Tr}\{\cosh(2j\mathbf{P})\}\,. \end{aligned}\tag{7}$$

The crystallographic identity of the sublattices implies

$$\mathrm{Tr}\{Q_A^2\cosh(2j\mathbf{P})\}=\mathrm{Tr}\{Q_B^2\cosh(2j\mathbf{P})\}\,,$$

and hence, writing R for S_B/S_A,

$$\begin{aligned} \hat{\chi}&=\hat{\chi}_1(j^2)+[2R/(1+R^2)]j\hat{\chi}_2(j^2)\,,\\ \hat{C}_H&=\hat{C}(j^2)\,;\\ \hat{\chi}_1(j^2)&=3N^{-1}\,\mathrm{Tr}\{Q_A^2\cosh(2j\mathbf{P})\}/\mathrm{Tr}\{\cosh(2j\mathbf{P})\}\,,\\ \hat{\chi}_2(j^2)&=3(Nj)^{-1}\,\mathrm{Tr}\{Q_AQ_B\sinh(2j\mathbf{P})\}/\mathrm{Tr}\{\cosh(2j\mathbf{P})\}\,,\\ \hat{C}(j^2)&=3(Nz)^{-1}\,\mathrm{Tr}\{\mathbf{P}^2\cosh(2j\mathbf{P})\}/\mathrm{Tr}\{\cosh(2j\mathbf{P})\}\,. \end{aligned}\tag{8}$$

The functions $\hat{\chi}_1$, $\hat{\chi}_2$ and $\hat{C}$ depend on the lattice, but have no explicit R dependence. Of course, the variable $j=S_AS_BJ/\kappa T$ is uniformly scaled, for a fixed T, by the factor S_AS_B. In terms of j itself, however, the only R dependence in either function, $\hat{\chi}$ or $\hat{C}_H$, is the factor weighting the odd part of $\hat{\chi}$.

For the purposes of the numerical investigation to be described in the next section we shall assume that the functions, $\hat{\chi}$ and $\hat{C}_H$ may be expanded in powers of j:

$$\begin{aligned} \hat{\chi}&=1+\sum_{n=1}^{\infty}A_nj^n\,,\\ \hat{C}_H&=1+\sum_{n=1}^{\infty}C_nj^n\,. \end{aligned}\tag{9}$$

The coefficients C_n are precisely those for the ferromagnetic case, $R=1$. The relation of the susceptibility coefficients, A_n, to those for the ferromagnet is apparent at once from Eq. (8). Writing a_n to distinguish the

[5] S. Freeman and P. J. Wojtowicz, J. Appl. Phys. 39, 622 (1968).

ferromagnetic coefficients, we have $A_n = a_n$ for even n and $A_n = [2R/(1+R^2)]a_n$ for odd n.

The simple form of the dependence of $\hat{C}_H$ and $\hat{\chi}$ on R has profound implications for the manner in which the positive and negative J critical points and critical exponents vary with R. In terms of j, the location and nature of the singularities of $\hat{C}_H$ are independent of the spin magnitude, and are symmetrical between positive and negative J. The properties of the susceptibility are less straightforward.

It is usually assumed that near the ferromagnetic critical point, the susceptibility in the paramagnetic state has the form

$$\chi \approx A(1-j/j_c)^{-\gamma}. \tag{10}$$

The exponent γ describes the strength of the singularity in $\hat{\chi}$ at the critical point. The factor A gives an overall weight which scales the susceptibility. We shall define quantities analogous to j_c, γ, and A. The definitions will be chosen partly for convenience in view of the particular properties of the system under investigation, and partly so as to presuppose as little as possible about the nature of the singularity in $\hat{\chi}$. Nonetheless, the definitions will be such as to reduce to the same j_c, γ and A as in Eq. (10) when $\hat{\chi}$ has this simple form near the critical point. This, of course, is the most interesting case.

It will prove convenient to define a family of functions $\mathfrak{X}$ containing the various $\hat{\chi}_\lambda(j) = \hat{\chi}_1(j^2) + \lambda j \hat{\chi}_2(j^2)$, where we have written λ for $2R/(1+R^2)$ so that we may label the members of the family with λ as a subscript. The range of values of λ for which $\hat{\chi}_\lambda$ is to be contained in $\mathfrak{X}$ will be chosen as $-1 \leq \lambda \leq 1$. The functions having $\lambda < 0$ are given physical meaning through the relation

$$\hat{\chi}_{-\lambda}(j) = \hat{\chi}_\lambda(-j). \tag{11}$$

The function $\hat{\chi}_{-\lambda}$ may be regarded as giving, for positive values of j, information on the system with $J<0$ and the spin ratio corresponding to $|\lambda|$. Alternatively, $\hat{\chi}_{-\lambda}$ may be regarded as the staggered susceptibility of the system whose susceptibility is $\hat{\chi}_\lambda$. This follows from the invariance of the partition function of this classical system under simultaneous replacement of J by $-J$ and Q_A by $-Q_A$.

The fundamental property of the family $\mathfrak{X}$ which we exploit is that any member $\hat{\chi}_\lambda$ may be expressed as a linear combination of any two arbitrarily chosen distinct members of the family, $\hat{\chi}_\alpha$ and $\hat{\chi}_\beta$:

$$\hat{\chi}_\lambda(j) = (\lambda-\alpha/\beta-\alpha)\hat{\chi}_\beta(j) + (\lambda-\beta/\alpha-\beta)\hat{\chi}_\alpha(j). \tag{12}$$

Thus, if some property of $\hat{\chi}_\lambda$ is defined by a linear operation independent of λ and holds for two distinct members of $\mathfrak{X}$, Eq. (12) implies that it must hold for every member of $\mathfrak{X}$.

We define the positive-j critical point of $\hat{\chi}_\lambda$, $j_+(\lambda)$, as the largest value of j having the property that $\hat{\chi}_\lambda(j)$ is analytic for all j': $0 \leq j' < j$. Similarly, $j_-(\lambda)$ is the smallest value of j such that $\hat{\chi}_\lambda(j)$ is analytic for all j': $j < j' \leq 0$. This definition presupposes that $\hat{\chi}_\lambda(j)$ is analytic at the origin, of course.

If $\hat{\chi}_\alpha$ and $\hat{\chi}_\beta$ are analytic for some j, then it follows from Eq. (12) that all the other members of $\mathfrak{X}$ are analytic at j as well. We conclude from this that the value of $j_+(\lambda)$ is the same for all λ,

$$j_+(\lambda) = j_c, \tag{13}$$

except possibly for a single exceptional value, $\lambda = \lambda_0$, for which $j_+(\lambda_0) > j_c$. Moreover, since $j_-(\lambda) = -j_+(-\lambda)$; it follows that

$$j_-(\lambda) = -j_c \tag{14}$$

for all λ except $-\lambda_0$, for which $j_-(-\lambda_0) = -j_+(\lambda_0) < -j_c$. Thus, the critical points of the susceptibility are symmetric between positive and negative J and do not change with spin ratio except that there may be a single exceptional spin ratio for which the critical j's are exceptionally large in magnitude. If we make the customary physical assumption that the critical points given by $\hat{\chi}$ and $\hat{C}_H$ are necessarily the same, then we are assured that there is no such exceptional spin ratio. We assume this to be the case throughout the following. What we have deduced concerning the location of the critical points of $\hat{\chi}$ constitutes evidence that this assumption is consistent in the present case. The critical temperature, T_c, is given in terms of the critical j value, j_c, by $T_c = |J| S_A S_B / \kappa j_c$. This holds for either sign of J.

For each λ, we now define the positive j critical exponent, $\gamma_+(\lambda)$, as the greatest lower bound of the values of γ' for which

$$\lim_{j \to j_c -} (j-j_c)^{\gamma'} \hat{\chi}_\lambda(j) = 0. \tag{15}$$

The negative j critical exponent, $\gamma_-(\lambda)$, is defined analogously. The symmetry of $\hat{\chi}$ implies that

$$\gamma_+(\lambda) = \gamma_-(-\lambda). \tag{16}$$

Suppose that for some value of γ', the limit in Eq. (15) is zero for both $\lambda = \alpha$ and $\lambda = \beta$. Then it follows from Eq. (12) that γ' makes this limit vanish for every $\hat{\chi}_\lambda$ in $\mathfrak{X}$; thus, for all λ, $\gamma_+(\lambda) \leq \gamma'$. We may conclude that for all values of λ, $\gamma_+(\lambda)$ has the same constant value,

$$\gamma_+(\lambda) \equiv \gamma, \tag{17}$$

except that possibly for a single value, $\lambda = \lambda_1$, the value of $\gamma_+(\lambda)$ may be exceptionally small:

$$\gamma_+(\lambda_1) < \gamma. \tag{18}$$

From Eq. (16), it follows that

$$\gamma_-(\lambda) = \gamma \tag{19}$$

for all λ except $-\lambda_1$, for which

$$\gamma_-(-\lambda_1) = \gamma_+(\lambda_1) < \gamma. \tag{20}$$

Now one certainly expects that the γ for the simple antiferromagnet ($R=1, J<0$) is less than that for the ferromagnet ($R=1, J>0$). We shall assume that this is the case: $\gamma_-(1)<\gamma_+(1)$. Then the discussion above implies that for all the properly *ferri*magnetic systems, regardless of the spin ratio or the sign of the exchange, the critical exponent is the same as that of the simple *ferro*magnet ($R=1, J>0$). For $0\leqslant\lambda<1$,

$$\gamma_-(1)<\gamma_-(\lambda)=\gamma_+(\lambda)=\gamma_+(1)\equiv\gamma. \tag{21}$$

The staggered susceptibility also has this same exponent for all R and either sign of J, except for the ferromagnet itself. This has a smaller value, as required by Eq. (20), when $\lambda_1=-1$ is substituted.

Following the form of $\hat{\chi}$ assumed in Eq. (10), we would naturally wish to define $A(\lambda)$ as

$$A(\lambda)=\lim_{j\to j_{c-}}(1-j/j_c)^\gamma\hat{\chi}_\lambda(j). \tag{22}$$

However, it is not necessarily obvious that $A(\lambda)$ is then finite and nonzero. This certainly is not a property required by the definition of γ. We may illustrate these pathological cases by imagining A in Eq. (10) replaced by $|\ln(1-j/j_c)|$ or by $|\ln(1-j/j_c)|^{-1}$. Now for $\lambda=1$, Eq. (22) holds with $A(-1)=0$. This follows from the assumption $\gamma_-(1)<\gamma_+(1)$. Thus if Eq. (22) holds with finite $A(\lambda)$ for some other $\lambda\neq-1$, it follows from Eq. (12) that it holds for all λ. We shall assume this to be the case. This assumption is consistent with the results of the numerical study described in Sec. III. Then Eq. (12) implies that $A(\lambda)$ is a linear function of λ. Since $A(-1)=0$, we can write

$$A(\lambda)=(1+\lambda)\mathcal{A}, \tag{23}$$

where $\mathcal{A}$ is a constant.

The same technique outlined above can be used to characterize the weaker singularities in $\hat{\chi}_\lambda$. Consider the functions $\psi_\lambda(j)$ defined for each λ, $-1\leq\lambda\leq1$, by

$$\psi_\lambda(j)=\hat{\chi}_\lambda(j)-\mathcal{A}\left[\frac{1+\lambda}{(1-j/j_c)^\gamma}+\frac{1-\lambda}{(1+j/j_c)^\gamma}\right]. \tag{24}$$

The totality of such functions has the same linear property, Eq. (12), as the original collection, $\mathfrak{X}$. Clearly $\psi_\lambda(j)$ has no singularities for $|j|<j_c$. Moreover, for $\lambda=+1$, it must have a singularity at $j=-j_c$. This corresponds to the critical point of the simple antiferromagnet at which point $\hat{\chi}$ was supposed to be singular. The leading terms which we have subtracted out have no singularity for this value of j and λ so this particular singularity must still be present in ψ_λ. Therefore $\psi_\lambda(j)$ has singularities at $\pm j_c$ for all λ, by the linear property of Eq. (12), except possibly for exceptional values as mentioned after Eqs. (13) and (14). We cannot in this case argue on physical grounds against the existence of these exceptional values. Critical exponents may be defined for ψ_λ just as they were for $\hat{\chi}_\lambda$. The argument given before for the constancy of such exponents holds in this case as well. It is still, however, not necessary that these next smaller exponents characterize the singularity of the simple antiferromagnet. Their weighting factor, analogous to $A(\lambda)$, might be such that they too make no contribution to this particular singularity. This is an interesting question in itself and hopefully could be resolved by a Padé approximant analysis.

III. PADÉ APPROXIMANT STUDY OF CUBIC LATTICES

Coefficients a_1 through a_8 for the classical Heisenberg ferromagnet have been published by Wood and Rushbrooke[1] for the simple and body-centered cubic lattices. The associated ferrimagnets, which have spins of different magnitudes on the two sublattices, have the properties described in the previous section. Thus, the first eight coefficients A_1 through A_8 of the susceptibility series for these ferrimagnets are available for analysis by Padé approximants [see discussion following Eq. (9)]. We have carried out this numerical study for two reasons: first, to verify the properties predicted above and to observe to what extent they are embodied in the truncated series; second, to exploit the additional degree of freedom present in R for the study of the critical parameters. As a preliminary step, Padé approximants to $\hat{\chi}_\lambda$, $\hat{\chi}_\lambda^{3/4}$ and $\hat{\chi}_\lambda'/\hat{\chi}_\lambda$ (where $\hat{\chi}_\lambda'=d\hat{\chi}_\lambda/dj$) were constructed for $\lambda=0.1, 0.2, \cdots, 0.9, 1.0$. The locations and residues of the poles of these approximants confirmed, in a general way, the behavior predicted in Sec. II. The approximants to $\hat{\chi}_\lambda$ had poles at both positive and negative j near the published ferromagnetic values. The negative pole disappeared as λ approached 1 and the positive pole joined smoothly to the ferromagnetic limit, $\lambda=1.0$. The residues at $+j_c$ increase nearly linearly as λ increases from 0.1 to 1.0 while the residues at $-j_c$ decrease and appear to pass through zero of λ near unity. These residues measure $A(\lambda)$ as it was defined in Eq. (22). The approximants to $\hat{\chi}_\lambda^{3/4}$ show these same features. However, the locations of the poles of $\hat{\chi}_\lambda^{3/4}$ are more nearly constant with λ and reproduce more closely the value of j_c expected from the previous studies of the ferromagnet.[1]

The logarithmic derivative, $\hat{\chi}'/\hat{\chi}$ should have poles at $j=\pm j_c$. The residue at each should be $-\gamma$ for all $\lambda\neq1$. This is observed to a degree for a range of intermediate values of λ. The negative-j pole occurs at a value nearly symmetrical to the positive pole for values of λ less than about 0.6, it then moves away and becomes complex. For the smaller values for λ the residue at the negative-j pole is consistent with a value of γ near 1.4.

As expected, the positive j pole is much better behaved. For values of λ greater than about 0.3, the estimates of j_c and γ for the positive-j singularity obtained from the more consistent entries in the Padé table are constant to within a few parts per thousand up

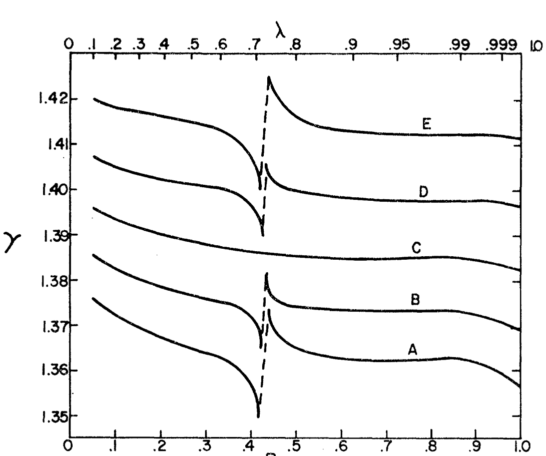

FIG. 1. Simple cubic lattice. Value of [4,3] approximant to $(j_c-j)\hat{\chi}_\lambda'/\hat{\chi}_\lambda$ at $j=j_c$. The labeled curves correspond to choices of j_c as follows: (A) 0.345, (B) 0.3455, (C) 0.346, (D) 0.3465, (E) 0.347.

to $\lambda=1.0$. The γ's are rather less constant but show a broad region of λ's less than one where their values are steady. The values of j_c and γ for this plateau are

$$\begin{aligned} &\text{sc} \quad j_c=0.3456\,, \quad \gamma=1.378\,,\\ &\text{bcc} \quad j_c=0.2425\,, \quad \gamma=1.347\,. \end{aligned} \tag{25}$$

These estimates agree quite closely with those of Wood and Rushbrooke[1] for $\lambda=1$. The behavior of the approximants to $\hat{\chi}_\lambda'/\hat{\chi}_\lambda$, as well as to $\hat{\chi}_\lambda$ itself and $\hat{\chi}_\lambda^{3/4}$, is consistent with the outline of the critical properties of $\hat{\chi}_\lambda$ given in Sec. II. However, there are several reasons why further work is necessary before the quantitative estimates of j_c and γ given in Eq. (25) can be accepted. Although the best estimates of j_c and γ obtained from the Padé table in the region, $\lambda>0.3$, are quite constant, nonetheless, a given order of Padé approximant often shows irregularities somewhere within this region. Moreover, the consistency of the Padé table becomes markedly worse as $\lambda\to 1.0$. This is most noticeable for the sc lattice but can be discerned for the bcc as well. In view of the fact that this is precisely where the ferrimagnetic system goes over into the simple ferromagnet, this lack of perfect regularity is unfortunate.

In order to study a function whose behavior could be more easily interpreted, we formed Padé approximants to $(j_c-j)\hat{\chi}'/\hat{\chi}$ and evaluated at $j=j_c$. If j_c is known, such a procedure estimates γ; in our case we wished to study the behavior of these estimates with varying λ for a succession of choices of j_c. Constructing Padé tables and estimating from them the best value of γ, the resulting estimate is quite constant, as a function of λ, for appropriately chosen j_c. However, more can be learned by study of the estimates obtained from a single-order Padé approximant, the [4,3] for example. In general, such an estimate is reasonably constant except near a particular intermediate value of λ ($\lambda\approx 0.7$ for the sc and $\lambda\approx 0.85$ for the bcc lattice). For most values of j_c, the estimate of γ regarded as a function of λ, $\gamma(\lambda)$, shows a rapid variation near this value of λ; in fact, it appears to have a pole there. However, if j_c is chosen within a rather narrow special range, this singularity is absent and the resulting curve $\gamma(\lambda)$ is strikingly smooth and level down to quite small values of λ. This behavior is illustrated for simple and body-centered cubic lattices in Figs. 1 and 2, respectively. We cannot claim to understand what causes $\gamma(\lambda)$ to behave in this fashion. Nevertheless, the criterion of smooth, level behavior is sufficiently straightforward and firmly based theoretically, and the departure from this required mode of behavior sufficiently striking for us to feel justified in picking the exceptional value of j_c which makes $\gamma(\lambda)$ level as the best estimate of j_c. The level plateau in $\gamma(\lambda)$ as $\lambda\to 1$ provides a corresponding estimate of γ. This type of behavior was found in both [4,3] and [3,4] Padé approximants (also in the [5,2] approximant in the case of the bcc lattice). The special choices of j_c which must be made to suit the different orders of approximant differ only by a few parts per thousand. The best estimates of j_c and γ for the two lattices are

$$\begin{aligned} &\text{bcc} \quad j_c=0.2425\pm 0.0002\,,\\ &\qquad\quad \gamma=1.345\pm 0.005\,,\\ &\text{sc} \quad j_c=0.3460\pm 0.0003\,,\\ &\qquad\quad \gamma=1.385\pm 0.01\,. \end{aligned} \tag{26}$$

The errors given are intended to indicate the range within which we could locate j_c by this method, as well as the degree of consistency of the estimates based on different orders of approximant. The Padé table for the estimation of γ from the various order approximants showed a high degree of consistency for the chosen values of j_c over almost all of the range of λ.

The values of j_c given in Eq. (26) are identical with those which Wood and Rushbrooke[1] found in their study of the ferromagnetic systems ($\lambda=1$) based on these lattices. These authors found $\gamma\sim 1.36$ for both lattices. Our estimates of γ are in qualitative agreement with this value as well as with the value $\gamma=1.38$ for both lattices obtained recently by Stanley.[4,6] However, we do see a clear and consistent difference between the two lattices. The discrepancies between our results for γ and those of the previous investigators is somewhat larger than the errors cited in Eq. (26). The safest interpretation of these discrepancies is to regard γ as being determined only to within a few percent as indicated by the several estimates. We feel, however, that our method (Padé approximants plus use of the theoretically justified nonvariation of γ with λ) is more precise and reliable than techniques previously employed, especially those involving linear extrapolations as used in Refs. 4

[6] H. E. Stanley, Phys. Rev. 164, 709 (1967).

and 6 in particular. We feel this to be the case despite the use of an additional coefficient in the susceptibility series (unfortunately not available to us when the present work was completed) in Refs. 4 and 6.

At this point, it is natural to consider Padé approximants to $\hat{\chi}^{1/\gamma}$ for a succession of values of γ. The location of the appropriate pole gives an estimate of j_c and the variation of this estimate with λ may be investigated just as above. Unfortunately the curves $j_c(\lambda)$ obtained in this manner are considerably more complex and less easily interpreted than those obtained from $(j_c-j)\hat{\chi}'/\hat{\chi}$ above. For both lattices, the behavior of $j_c(\lambda)$ is irregular both for R close to unity and for small values of R (λ less than about 0.6). Attempts to minimize each of these structures in turn give contradictory estimates of γ. Thus, for the simple cubic lattice, the rapid variation near $R=1$ may be progressively diminished by increasing γ from 1.37 to 1.42; however, this increase in γ makes the singularity at small R increasingly prominent. On the other hand, for the body-centered cubic lattice, decreasing γ from 1.37 to 1.35 makes the singularity for R near one vanish, but the reverse change in γ is required to make the small R singularity disappear. In the absence of any understanding of the origin or meaning of this behavior, we shall simply disregard these properties of $\hat{\chi}^{1/\gamma}$ except for the followlng remarks. First, a choice of γ in agreement with Eq. (26) above does, at least, make the location of the positive pole of $\hat{\chi}^{1/\gamma}$ constant to within a few parts per thousand over most of the range of λ (or of R if we think of it as varying between zero and one). This is true for any of the higher-order Padé approximants. These, moreover, show a high degree of mutual consistency. Second, the procedure used to extract j_c and γ from the Padé approximants to $(j_c-j)\hat{\chi}'/\hat{\chi}$ is partly empirical. The mysterious behavior of the approximants of $\hat{\chi}^{1/\gamma}$ may decrease one's confidence in this method; on the other hand, its correct evaluation of the critical points, reliably known from the ferromagnets, is a strong point in its favor. In balance, we regard the estimates of the critical exponent, γ given above as being the best currently available for these systems.

We may now use the value of j_c obtained above to investigate the behavior of $\hat{\chi}_\lambda$ for negative j. The critical exponent for the singularity at $-j_c$ can be evaluated from $-(j+j_c)\hat{\chi}'/\hat{\chi}$; the Padé approximants to this function are evaluated at $j=-j_c$. The exponent shows a less regular behavior over a range of λ than for the $j>0$ singularity. This is to be expected since the term singular at $-j_c$ appears in $\hat{\chi}$ multiplied by the small weight factor $(1-\lambda)\mathfrak{a}$. Nonetheless, in the bcc case, the value obtained for the negative j exponent lies within one percent of 1.36 for $0.1\leq\lambda\leq 0.6$; and in the sc case, it lies within one percent of 1.40 for $1\leq\lambda\leq 0.5$. The higher-order Padé approximants in these regions yield fairly consistent estimates of γ. For large λ less than unity, the values of the exponent at the negative j singularity obtained in this fashion vary wildly and

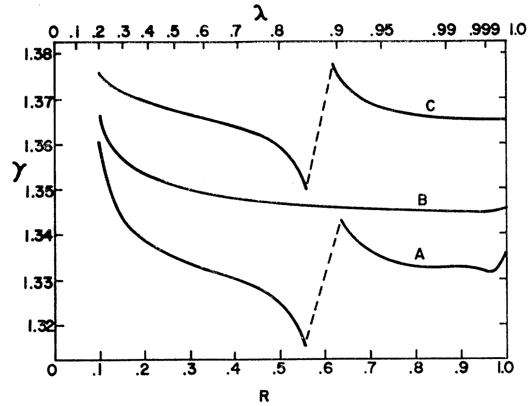

FIG. 2. Body-centered cubic lattice. Value of [4,3] approximant to $(j_c-j)\hat{\chi}_\lambda'/\hat{\chi}_\lambda$ at $j=j_c$. The labeled curves correspond to choices of j_c as follows: (A) 0.2420, (B) 0.2425, (C) 0.2430.

show little consistency. However, for $\lambda=1$ itself, the Padé table is again very regular. The best estimates are

$$\begin{aligned} &\text{bcc} \quad \gamma(-1)=-0.01\pm 0.01\,, \\ &\text{sc} \quad \gamma(-1)=-0.05\pm 0.05\,. \end{aligned} \tag{27}$$

It is virtually certain that, in fact, $\gamma(-1)=0$ for both lattices since one expects a constant term to be present in the antiferromagnetic susceptibility at this point.

We have evaluated the over-all multiplicative factor, $A(\lambda)$, which weights the singular term $(1-j/j_c)^{-\gamma}$ in $\hat{\chi}$. Padé approximants were formed to $(1-j/j_c)^\gamma\hat{\chi}_\lambda(j)$ and evaluated at $j=j_c$. The values of j_c and γ used were, respectively, 0.2425 and 1.345 for the bcc, and 0.346 and 1.385 for the sc. The Padé tables were quite consistent and showed very clearly the expected linear dependence of A on λ. The general linear form of $A(\lambda)$ may be written

$$A(\lambda)=\mathfrak{a}_1+\mathfrak{a}_2\lambda\,. \tag{28}$$

Using the best estimates from the Padé tables for $A(\lambda)$, we obtained the estimates of $\mathfrak{a}_1$ and $\mathfrak{a}_2$:

$$\begin{aligned} &\text{sc} \quad \mathfrak{a}_1=0.471\,, \\ &\qquad \mathfrak{a}_2=0.477\,; \\ &\text{bcc} \quad \mathfrak{a}_1=0.455\,, \\ &\qquad \mathfrak{a}_2=0.460\,. \end{aligned} \tag{29}$$

We regard these results as consistent with the form anticipated for $A(\lambda)$:

$$A(\lambda)=(1+\lambda)\mathfrak{a}\,. \tag{30}$$

The difference in the estimates of $\mathfrak{a}_1$ and $\mathfrak{a}_2$ probably indicates the extent to which numerical errors have accumulated by this stage in the calculation. Thus, basing the error estimates on the extent of disagreement in Eq. (29),

$$\begin{aligned} &\text{sc} \quad \mathfrak{a}=0.474\pm 0.005\,, \\ &\text{bcc} \quad \mathfrak{a}=0.457\pm 0.005\,. \end{aligned} \tag{31}$$

TABLE I. Padé approximant estimates of η for a range of λ values.

λ	[4,3]	[3,4]	λ	[4,3]	[3,4]
1.0	0.916	0.912	−0.1	1.139	1.008
0.95	1.051	1.027	−0.2	1.164	1.100
0.9	1.006	0.977	−0.3	1.189	1.146
0.85	1.009	0.972	−0.4	1.213	1.184
0.8	1.021	0.973	−0.5	1.239	1.219
0.7	1.063	0.979	−0.6	1.265	1.251
0.6	1.140	0.983	−0.7	1.290	1.281
0.5	1.453	0.987	−0.8	1.316	1.310
0.4	0.682	0.995	−0.85	1.328	1.323
0.3	0.954	1.011	−0.9	1.340	1.336
0.2	1.031	1.038	−0.95	1.351	1.348
0.1	1.077	1.080	−1.0	1.362	1.360

It should be mentioned that the estimates of $\mathcal{A}$ depend quite strongly on the values used for j_c and γ. If the uncertainty in the estimates of these latter quantities were included in the errors indicated in Eq. (31), these errors would be larger by a factor of at least three.

The parameters j_c, γ, $\mathcal{A}$ characterize completely the leading singularities in $\hat{\chi}_\lambda$—that is to say, the most rapidly divergent part at the real singularities closest to $j=0$. These leading singular terms have the form

$$\mathcal{A}\left[\frac{1+\lambda}{(1-j/j_c)^\gamma}+\frac{1-\lambda}{(1+j/j_c)^\gamma}\right]. \tag{32}$$

The possibility of subtracting this term out of $\hat{\chi}$, and investigating successively weaker singularities was discussed in Sec. II. Unfortunately, the function ψ_λ defined in Eq. (24) is not well suited to analysis by Padé approximants. The error terms due to the approximate values we must use for the parameters j_c, γ and $\mathcal{A}$ are so highly singular that they dominate the behavior of the Padé approximants even though their numerical contribution to the value of the function is small over most of its domain. The function

$$\Omega_\lambda=(1-j/j_c)^\gamma\hat{\chi}_\lambda-(1+\lambda)\mathcal{A} \tag{33}$$

is more tractable. We have investigated the critical exponent of this function at $+j_c$ for $-1\leq\lambda<1$ by the same method used with $\hat{\chi}_\lambda$. The Padé approximants to $(j-j_c)\Omega_\lambda'/\Omega_\lambda$ were formed and evaluated at j_c. The results for the bcc lattice are given in Table I. The values of the parameters j_c, γ and $\mathcal{A}$ used were respectively 0.2425, 1.385, 0.457. The behavior for the sc lattice was essentially the same though less regular. Note that the exponent η in this table represents a singular term in $\hat{\chi}_\lambda$ proportional to

$$(1-j/j_c)^{\eta-\gamma}. \tag{34}$$

The evidence suggests that a singular term of the power-law type with exponent $\gamma'=\gamma-1$ is present in $\hat{\chi}$ except at the critical point of the simple antiferromagnet ($\lambda=-1$ and $j=j_c$ or equivalently $\lambda=+1$ and $j=-j_c$). At this latter exceptional point the leading exponent seems to be of order zero. This is presumably just the constant term expected to be present.

IV. EXPERIMENTAL IMPLICATIONS

For the special type of classical system studied in the preceding sections, there is a close similarity between the properly ferrimagnetic and the simple ferromagnetic systems in their critical properties. In fact, the critical index, γ, is precisely the same regardless of the magnitudes of the spins on the two sublattices or the sign of the exchange (excepting the simple antiferromagnet). We believe that these findings have strong implications for the critical point behavior of real ferrimagnets. The exponents of the classical and quantum Heisenberg ferromagnets have proved to be quite similar.[1,3] It is reasonable to expect that this will hold true for the ferrimagnets as well. Thus we expect that the exponents of the real ferrimagnets will be in the neighborhood of 1.35 (barring unforeseen sensitivity to lattice structure).

Preliminary experimental results[7] for yttrium iron garnet have indicated a value of γ less than one. The experiments, however, were not performed with the intention of observing the critical behavior. We believe that further experiments are highly desirable.

[7] E. E. Anderson, Phys. Rev. Letters 17, 375 (1966).