Quasi-One-Dimensional and Quasi-Two-Dimensional Magnetic Systems: Determination of Crossover Temperature and Scaling with Anisotropy Parameters*

Luke L. Liu and H. Eugene Stanley
Physics Department, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
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Magnetic systems of lattice dimensionality $\bar{d}$ are considered, with interaction strength J in d lattice directions and RJ in the remaining $(\bar{d}\text{-}d)$ directions; recent experimental work on quasi-one-dimensional and quasi-two-dimensional systems are germane to the cases $d = 1, 2$, respectively (and $\bar{d} = 3$). Rigorous relations are established for the first few derivatives with respect to R of the susceptibility $\chi(R)$, the second moment of the correlation function $\mu_2(R)$, and the specific heat $C_H(R)$. These results permit detailed statements about the coefficients in the expansions of $\chi(R)$, $\mu_2(R)$, and $C_H(R)$ in Taylor expansions in R about the d-dimensional limit ($R = 0$), and thus permit estimates concerning the temperatures at which each of these functions exhibits significant departures as $T \to T_c^+$ from its high-temperature d-dimensional behavior. These are the first estimates of the "crossover temperature" at which the system crosses over to $\bar{d}$-dimensional behavior from its limiting high-temperature behavior (at which the weak interactions RJ are ineffective compared to the strong interactions J in the d lattice directions). In particular, they emphasize the fact that the crossover temperature depends upon the *function* being considered as well as upon the system and the value of R. These results also give strong support to the generalized scaling hypothesis in which R is scaled.

I. INTRODUCTION

An extremely large number of real magnetic materials[1-4] are not "directionally isotropic" in the sense that the coupling strengths in all lattice directions are not identical. Many systems displaying such lattice anisotropy are described by special cases [e.g., Eqs. (1.2), (1.3) and Fig. 1] of the general interaction Hamiltonian

$$\mathcal{H}(d,\bar{d}) = -\sum_{\vec{u}_i=\vec{u}_j} J_{ij}\vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)} - R\sum_{\vec{u}_i\neq\vec{u}_j} J_{ij}\vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)} . \tag{1.1}$$

Here the $\bar{d}$-dimensional lattice vector $\vec{r} = (x_1, x_2, \ldots, x_{\bar{d}})$ is partitioned into a $(\bar{d}\text{-}d)$-dimensional vector $\vec{u} = (x_1, x_2, \ldots, x_{\bar{d}-d})$ and a d-dimensional vector $\vec{v} = (x_{\bar{d}-d+1}, \ldots, x_{\bar{d}})$, and $\vec{r} = (\vec{u}, \vec{v})$. The first summation is over pairs of magnetic sites whose relative displacement vector $\vec{r}_{ij} = (\vec{u}_i - \vec{u}_j, 0)$ has no component in the directions $\vec{v} = (x_{\bar{d}-d+1}, \ldots, x_{\bar{d}})$, while the second summation is over pairs of sites for which $\vec{v}_i \neq \vec{v}_j$. The "spins" $\vec{S}_i^{(D)}$ of (1.1) are D-dimensional unit vectors, so that $D = 1$, 2, 3, and ∞ correspond to the $S = \frac{1}{2}$ Ising, classical planar, classical Heisenberg, and spherical models, respectively.[5,6]

Although many of the theoretical results of this paper are valid for the general Hamiltonian (1.1), a principal motivation of the authors has been interpretation of the plethora of recent experiments[1-4] (pioneered by Birgeneau, Shirane, and others), on quasi-two-dimensional and, most recently, on quasi-one-dimensional systems. These cases, sketched in Figs. 1(a) and 1(b) respectively, correspond to $d = 2$ and $d = 1$, respectively, in (1.1), with, of course, $\bar{d} = 3$. Four particular examples are the following:

(i) Quasi-two-dimensional magnets,[1] many of the K_2NiF_4 structure, have $\bar{d} = 3$, $d = 2$, and $R \sim 10^{-3}$ or smaller.

(ii) The very recently discovered quasi-one-dimensional[2] magnet $(CD_3)_4NMnCl_3$ has $\bar{d} = 3$, $d = 1$.

(iii) The familiar "three-dimensional" Heisenberg ferromagnet $CrBr_3$ actually possesses considerable lattice anisotropy, with $\bar{d} = 3$, $d = 2$, and $R \sim 0.06$.[3]

(iv) It has recently been proposed[4] that the metamagnet $FeCl_2$ is describable by a spin $S = 1$ Ising Hamiltonian with $\bar{d} = 3$, $d = 2$, and $R = -0.05$.

For a simple cubic (sc) lattice, if we further assume that one can neglect all coupling strengths except those between nearest-neighbor pairs of sites, then the Hamiltonian (1.1) reduces for quasi-two-dimensional [Fig. 1(a)] and quasi-one-dimensional [Fig. 1(b)] systems, respectively, to

$$\mathcal{H}(2,3) = -J\sum_{\langle ij\rangle}^{xy}\vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)} - RJ\sum_{\langle ij\rangle}^{z}\vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)} \tag{1.2}$$

and

$$\mathcal{H}(1,3) = -J\sum_{\langle ij\rangle}^{z}\vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)} - RJ\sum_{\langle ij\rangle}^{xy}\vec{S}_i^{(D)}\cdot\vec{S}_j^{(D)} . \tag{1.3}$$

For the sake of simplicity, some of the proofs presented below are carried out in detail for the Hamiltonian (1.2) rather than for (1.1). Note that (1.2) reduces to (1.3) when $J \to J/R$ and $R \to \infty$.

A most dramatic finding of the earliest detailed neutron scattering experiments[1(b)] (which were on

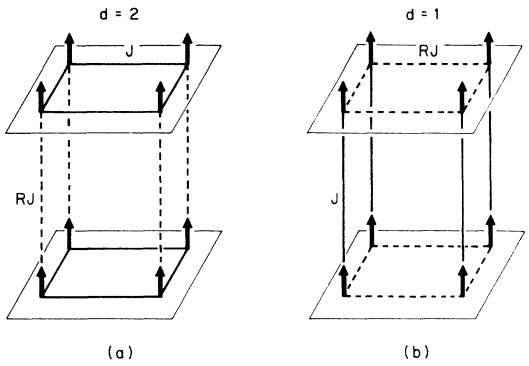

FIG. 1. Schematic representations of (a) quasi-two-dimensional lattice ($d=2$, $\bar{d}=3$), and (b) quasi-one-dimensional lattice ($d=1$, $\bar{d}=3$). Here the solid lines indicate interaction bonds of strength J, while the dashed lines indicate bonds of strength RJ with $R \ll 1$. For the graphs in Figs. 2–5, the RJ bonds are indicated by a heavy solid line rather than a dashed line.

systems with $d=2$ and small R) was that well above the critical temperature $T_c = T_c(R)$, the system displays behavior that is essentially d dimensional. This is intuitively plausible since well above $T_c(R)$, the correlations between the moments are governed by the stronger interactions J so that the order will begin to appear in these directions first; only as one gets close to $T_c(R)$ will the correlations arising from the weaker interactions RJ (in the remaining $3-d$ lattice directions) begin to manifest themselves, and accordingly the system will cross over from d-dimensional to three-dimensional behavior as $T \to T_c^+(R)$.

This crossing over is of course not infinitely sharp, and hence the frequently used term "crossover temperature," $T_x(R)$, is somewhat of a misnomer. In fact, the crossover from d-dimensional to three-dimensional behavior begins, literally, at infinite temperature and is not complete until $T = T_c^+(R)$. However, we shall see that it makes sense to speak of a crossover *region* $T_A(R) \geq T \geq T_B(R)$ defined such that within it "most of the crossing over" takes place. Clearly $T_A(R)$ and $T_B(R)$ will depend upon the accuracy of one's measuring instruments and, as we shall see, upon the function being measured. (See the discussion in Sec. VI C.)

The crossover behavior has been a well-established experimental fact,[1,2] but apart from scaling-type arguments,[7–9] little has been done to attempt to understand this fact. Actually, the crossover region has never been defined precisely. A first purpose of this work is to provide a practical and useful definition of this region. This purpose is achieved by considering the expansion with respect to the anisotropy parameter R of the function in question. For example, for the reduced susceptibility $\bar{\chi}(R) = \chi(R)/\chi_{\mathrm{Curie}}(R)$,

$$\bar{\chi}(R) = \bar{\chi}_0(0) + \bar{\chi}_1(0)R + \bar{\chi}_2(0)\frac{R^2}{2!} + \cdots + \bar{\chi}_n(0)\frac{R^n}{n!} + \cdots , \quad (1.4)$$

where

$$\bar{\chi}_n(0) \equiv \left(\frac{\partial^n \bar{\chi}(R)}{\partial R^n}\right)_{R=0} . \quad (1.5)$$

In Sec. II we shall show that $\bar{\chi}_0(0) = \bar{\chi}_d$, the d-dimensional susceptibility. At high temperature, as well as at small R, the d-dimensional approximation is satisfactory and $\bar{\chi}(R) \cong \bar{\chi}_d$. Thus if we could get the magnitude of the first few "coefficients" $\bar{\chi}_n(0)$, we should be able to estimate the temperature at which the d-dimensional approximation is no longer satisfactory. This temperature is the "crossover temperature"[10] $T_A(R)$ at which the effect of the RJ interactions (and hence the three-dimensional behavior) becomes experimentally noticeable.

A second purpose of this work concerns testing the scaling hypothesis[7–9] in situations such as this for which the parameter R that changes the universal class of the system is scaled like an external field. In this case when $R=0$, we have d-dimensional exponents and when $R \neq 0$ we have a $\bar{d}$-dimensional system and, *sufficiently close* to $T_c(R)$, have $\bar{d}$-dimensional exponents. Scaling with a parameter has been strongly questioned by Oitmaa and Enting,[11] whose numerical results indicate a failure in predictions of scaling for the sequence of exponents γ_n characterizing the behavior as $T \to T_c^+$ of the quantities $\bar{\chi}_n(R=0)$. We shall see that in fact many of the numerical results of Oitmaa and Enting[11] are contradicted by the rigorous relations that appear to exist among certain of the γ_n.

In Secs. II, III, and IV we consider, respectively, the first, second, and third derivatives with respect to R of certain thermodynamic functions and of quantities related to the pair correlation function. For simplicity and clarity of derivation, we shall consider the Hamiltonian of Eq. (1.2) with $D=1$; specifically, we shall consider a $S=\frac{1}{2}$ Ising model situated on a sc lattice with coupling strengths J in the x and y directions and RJ in the z direction. Many of the results are of more general validity, and we shall indicate when this is the case. In Sec. V we tabulate our results and examine their possible range of validity, and the reader not interested in the details of our derivations is urged to skip directly to Sec. V. Finally, in Sec. VI we demonstrate various applications of the results of Sec. V, showing, in particular (i) how to predict when the system crosses over from d dimensional to $\bar{d}$ dimensional [i.e., what is $T_A(R)$] and (ii) how our results lend strong support to the scaling hypothesis for scaling with respect to an active parameter.[7–9]

II. FIRST-ORDER DERIVATIVES WITH RESPECT TO R OF THERMODYNAMIC FUNCTIONS AND OF QUANTITIES RELATED TO THE PAIR CORRELATION FUNCTION

In this section we derive relations for the first-order derivatives with respect to the anisotropy parameter R of various thermodynamic functions (Sec. II A) and of various quantities related to the two-spin correlation function (Sec. II B). In Secs. III and IV analogous relations are considered for second and third derivatives, respectively.

The system which shall be considered in this and the following two sections is the spin-$\frac{1}{2}$ Ising model for the case of a simple cubic ($\bar{d}=3$) to square ($d=2$) crossover. In the presence of an external field H, the system is described by the Hamiltonian

$$\mathcal{H}=\mathcal{H}_0+R\mathcal{H}_1-mH\sum_{\vec{u},z}s_{\vec{u},z} \quad , \tag{2.1}$$

where $\mathcal{H}_0$ and $R\mathcal{H}_1$ are, respectively, the first and second terms of Eq. (1.2) for $D=1$, $s_i\equiv\vec{S}_i^{(1)}$, and $m\equiv g\mu_B$, where g is the g factor and μ_B is the Bohr magneton. The "quasi-two-dimensional" lattice is sketched in Fig. 1(a); analogs of all of the results of Secs. II–IV can be obtained for the "quasi-one-dimensional" lattice of Fig. 1(b) [cf. Eq. (1.3)].

In Sec. V we discuss the generalization, where possible, to the arbitrary spin Ising model, to more complex lattices (e.g., the square to fcc crossover), and to arbitrary spin dimensionality D.

Note that $\mathcal{H}_0$ is in fact an assembly of two-dimensional (noninteracting) square lattices; i.e.,

$$\mathcal{H}_0\equiv-J\sum_z\sum_{\vec{u},\vec{u}'}^{nn}s_{\vec{u}z}s_{\vec{u}'z}\equiv\sum_z\mathcal{H}_d(z) \quad , \tag{2.2}$$

where $\vec{u}$, $\vec{u}'$ are two-dimensional lattice vectors with Cartesian coordinates $\vec{u}=(x,y)$. Each layer, described by $\mathcal{H}_d(z)$, is a conventional two-dimensional Ising lattice, although our notation looks more complex because we must perforce carry along the component z of the lattice vector.

One similarly observes that $\mathcal{H}_1$ is an assembly of one-dimensional linear chain lattices.

Before commencing with the derivations, we explain in detail two sorts of thermal averages to be utilized. The first sort of thermal average, denoted by the symbol $\langle\cdots\rangle_R$, is the average appropriate for the "three-dimensional" system described by the Hamiltonian $\mathcal{H}$ of Eq. (2.1); thus

$$\langle\cdots\rangle_R=\mathrm{Tr}(\cdots e^{-\beta\mathcal{H}})/\mathrm{Tr}(e^{-\beta\mathcal{H}}) \quad , \tag{2.3}$$

where $\beta\equiv1/kT$, k is the Boltzmann constant, and T is the temperature.

The second sort of thermal average, denoted by the symbol $\langle\cdots\rangle_d$, is the average appropriate for *one* of the two-dimensional layers described by the Hamiltonian $\mathcal{H}_d(z)$ of Eq. (2.2),

$$\langle\cdots\rangle_d=\mathrm{Tr}(\cdots e^{-\beta\mathcal{H}_d})/\mathrm{Tr}(e^{-\beta\mathcal{H}_d}) \quad . \tag{2.4}$$

Thus $\langle\cdots\rangle_d$ is a conventional average for a two-dimensional system.

Frequently we shall consider the system described by the Hamiltonian of (2.1) for the case $R=0$. Thus the symbol $\langle\cdots\rangle_0$ is defined through the relation

$$\langle\cdots\rangle_0=\langle\cdots\rangle_{R=0}\equiv\lim_{R\to0}\langle\cdots\rangle_R \quad . \tag{2.5}$$

Note that the thermal average $\langle\cdots\rangle_0$ is quite different from the thermal average $\langle\cdots\rangle_d$, although they are related by the following theorem.

Theorem T1. Let $f_l(z_l)$ denote products of spins in one of L layers with $z=z_l$. Then

$$\left\langle\prod_{l=1}^{L}f_l(z_l)\right\rangle_0=\prod_{l=1}^{L}\langle f_l\rangle_d \quad . \tag{2.6}$$

The proof of Theorem T1 follows directly from the observation that at $R=0$, there is no interaction among different layers; hence the three-dimensional system for $R=0$ is in fact an ensemble of (infinitely many) statistically independent two-dimensional systems. Hence from (2.1) and (2.2) it follows that

$$\mathrm{Tr}\left\{\prod_{l=1}^{L}f_l(z_l)\exp\left(-\beta\mathcal{H}_0-m\beta H\sum_{\vec{u},z}s_{\vec{u},z}\right)\right\}=\mathrm{Tr}\left\{\prod_{l=1}^{L}\left[f_l(z_l)\exp\left(-\beta\mathcal{H}_d(z_l)-m\beta H\sum_{\vec{u}}s_{u,z_l}\right)\right]\right\}$$

$$=\prod_{l=1}^{L}\left\{\mathrm{Tr}_l\left[f_l(z_l)\exp\left(-\beta\mathcal{H}_d(z_l)-m\beta H\sum_{\vec{u}}s_{\vec{u},z_l}\right)\right]\right\} \quad , \tag{2.7}$$

where the notation Tr denotes a trace over all spins on the three-dimensional lattice, and the notation Tr_l denotes a trace taken over spins on the layer with $z=z_l$. A similar relation may be derived for the denominator of Eq. (2.3) by choosing the operator $f_l(z_l)=1$ for all l in Eq. (2.7). Thus it follows from the definitions (2.3) and (2.4) that

$$\left\langle\prod_{l=1}^{L}f_l(z_l)\right\rangle_0=\prod_{l=1}^{L}\frac{\mathrm{Tr}_l\{f_l(z_l)\exp[-\beta\mathcal{H}_d(z_l)-m\beta H\sum_{\vec{u}}s_{\vec{u},z_l}]\}}{\mathrm{Tr}_l\{\exp[-\beta\mathcal{H}_d(z_l)-m\beta H\sum_{\vec{u}}s_{\vec{u},z_l}]\}}=\prod_{l=1}^{L}\langle f_l\rangle_d \quad , \tag{2.8}$$

and the proof of Theorem T1 is complete. [In the last expression of (2.8) we dropped the coordinate z_l because $\langle \cdots \rangle_d$ is the conventional average for a two-dimensional lattice and hence is independent of z_l.]

As a useful illustrative example of the application of Theorem T1, consider the thermal average $\langle s_{\vec{r}_i} s_{\vec{r}_j} \rangle_R$ of the product of two spins on sites $\vec{r}_i$ and $\vec{r}_j$ of a three-dimensional lattice $[\vec{r}_i = (\vec{u}_i, z_i)]$. Then at $R = 0$ we have

$$\langle s_{\vec{r}_i} s_{\vec{r}_j} \rangle_0 = \begin{cases} \langle s_{\vec{u}_i} s_{\vec{u}_j} \rangle_d & \text{if } z_i = z_j \quad (2.9a) \\ \langle s_{\vec{u}_i} \rangle_d \langle s_{\vec{u}_j} \rangle_d = \langle s_{\vec{u}} \rangle_d^2 & \text{if } z_i \neq z_j \quad . \quad (2.9b) \end{cases}$$

We should emphasize that in Eq. (2.6) we assume the existence of the thermodynamic limit, and we assume that the thermodynamic limit can be exchanged with the limit $R \to 0$. We cannot justify these assumptions, and hence we must be satisfied with this degree of rigor.

We are now prepared to derive, in Secs. II A and II B, relations concerning the first derivatives with respect to R of thermodynamic functions and quantities related to the two-spin correlation function.

A. Gibbs Function, Magnetization, and Susceptibility

The Gibbs potential G at temperature T and magnetic field H for a system with anisotropy parameter R is given by

$$G(T, H, R) = -kT \ln Z \quad , \quad (2.10)$$

where

$$Z \equiv \mathrm{Tr}(e^{-\beta \mathcal{H}}) \quad (2.11)$$

is the partition function. Hence from (2.1)

$$G_1(R) = \frac{\partial G(T, H, R)}{\partial R} = \frac{\mathrm{Tr}(\mathcal{H}_1 e^{-\beta \mathcal{H}})}{\mathrm{Tr}(e^{-\beta \mathcal{H}})} = \langle \mathcal{H}_1 \rangle_R \quad . \quad (2.12)$$

Since $\mathcal{H}_1$ consists only of products of pairs of spins on *different* layers l, we may use Theorem T1 [cf. Eq. (2.9b)] to obtain

$$G_1(R=0) = \langle \mathcal{H}_1 \rangle_0 = -J \sum_{\vec{u}, z} \langle s_{\vec{u}, z} s_{\vec{u}, z+1} \rangle_0 = -J \sum_{\vec{u}, z} \langle s_{\vec{u}} \rangle_d^2 \quad . \quad (2.13)$$

Thus in the thermodynamic limit, we have

$$G_1(R=0) = -NJ \langle s_{\vec{u}} \rangle_d^2 \quad , \quad (2.14)$$

where $N \equiv K^2 L$ is the total number of lattice sites in the full $\bar{d} = 3$ lattice.

To simplify the subsequent manipulations, it is useful to introduce the dimensionless exchange energy,

$$\mathcal{J} \equiv J/kT \quad , \quad (2.15)$$

magnetic field,

$$h \equiv mH/kT \quad , \quad (2.16)$$

reduced magnetization,

$$\overline{M}(R) \equiv \frac{M(T, H, R)}{M(T=0, R)} = \langle s_{\vec{r}} \rangle_R = -\frac{\beta}{N} \frac{\partial G(T, H, R)}{\partial h} \quad , \quad (2.17)$$

and reduced susceptibility,

$$\overline{\chi}(R) \equiv \frac{\chi}{\chi_{\mathrm{Curie}}} = \sum_{\vec{r}} \{ \langle s_0 s_{\vec{r}} \rangle_R - \langle s_0 \rangle_R \langle s_{\vec{r}} \rangle_R \} = \frac{-\beta}{N} \frac{\partial^2 G(T, H, R)}{\partial h^2} = \frac{\partial \overline{M}(R)}{\partial h} \quad . \quad (2.18)$$

Differentiating (2.13) with respect to h and using (2.17) and (2.18), we have for the first derivative of the reduced magnetization

$$\overline{M}_1(R) \equiv \frac{\partial \overline{M}(R)}{\partial R} \quad , \quad (2.19)$$

the relation

$$\overline{M}_1(R=0) = \frac{-\beta}{N} \frac{\partial G_1(R=0)}{\partial h} = 2\mathcal{J} \overline{M}_d \chi_d \quad , \quad (2.20)$$

where $\overline{M}_d$ and $\overline{\chi}_d$ are the d-dimensional reduced magnetization and reduced susceptibility, respectively.

For the first derivative of the reduced susceptibility,

$$\overline{\chi}_1(R) \equiv \frac{\partial \overline{\chi}(R)}{\partial R} \quad , \quad (2.21)$$

we have, on differentiating (2.13) twice, the result

$$\overline{\chi}_1(R=0) = \frac{-1}{N} \frac{\partial^2 G_1(R=0)}{\partial h^2} = 2\mathcal{J} \left\{ \overline{\chi}_d^2 + \overline{M}_d \frac{\partial \overline{\chi}_d}{\partial h} \right\} \quad . \quad (2.22)$$

Equations (2.13), (2.20), and (2.22) relate $G_1(R=0)$, $\overline{M}_1(R=0)$, and $\overline{\chi}_1(R=0)$ (the first derivatives with respect to R of the Gibbs potential, the magnetization, and the susceptibility) to the d-dimensional quantities $\overline{M}_d$ and $\overline{\chi}_d$. The utility of these three equations, the principal results of this subsection, will become clear in Secs. V and VI.

B. Two-Spin Correlation Function, Correlation Length, and Structure Factor

The two-spin correlation function

$$C_2(T, H, \vec{r}, R) \equiv \langle s_0 s_{\vec{r}} \rangle_R - \langle s_0 \rangle_R \langle s_{\vec{r}} \rangle_R \quad (2.23)$$

is of both theoretical and experimental interest. In particular, the correlation length $\xi(T, H, R)$ is directly related to $C_2(T, H, \vec{r}, R)$, where $\xi(T, H, R)$ is conventionally defined as

$$\xi(T, H, R) = [\mu_2(T, H, R)/\mu_0(T, H, R)]^{1/2} \quad , \quad (2.24)$$

where the moments μ_m of the correlation function are defined through

$$\mu_m(T,H,R) \equiv \sum_{\vec{r}} |\vec{r}|^m C_2(T,H,\vec{r},R) \tag{2.25}$$

and, in particular, from (2.18) it follows that the reduced susceptibility is simply the zeroth moment of the pair correlation function

$$\bar{\chi}(T,H,R) = \mu_0(T,H,R) \ . \tag{2.26a}$$

The static structure factor, measured in scattering experiments, is also related to the correlation function

$$S(T,H,\vec{q},R) \equiv \sum_{\vec{r}} C_2(T,H,\vec{r},R) e^{i\vec{q}\cdot\vec{r}} \ . \tag{2.27}$$

In particular, Eqs. (2.25)–(2.27) yield

$$\bar{\chi}(T,H,R) \equiv S(T,H,0,R) = \sum_{\vec{r}} C_2(T,H,\vec{r},R) \ . \tag{2.26b}$$

The first-order derivative with respect to R of $C_2(T,H,\vec{r}_i,R)$,

$$C_{2,1}(\vec{r}_i,R) \equiv \frac{\partial C_2(T,H,\vec{r}_i,R)}{\partial R} \ , \tag{2.28}$$

is given directly from (2.1):

$$\begin{aligned} C_{2,1}(\vec{r}_i,R) = -\beta\{&\langle s_0 s_i \mathcal{H}_1\rangle_R - \langle s_0 s_i\rangle_R \langle \mathcal{H}_1\rangle_R \\ &- \langle s_0\rangle_R[\langle s_i \mathcal{H}_1\rangle_R - \langle s_i\rangle_R \langle \mathcal{H}_1\rangle_R] \\ &- \langle s_i\rangle_R[\langle s_0 \mathcal{H}_1\rangle_R - \langle s_0\rangle_R \langle \mathcal{H}_1\rangle_R]\} \ . \end{aligned} \tag{2.29}$$

We are interested in the behavior of $C_{2,1}(\vec{r}_i,R)$ for $R=0$; to this end, it is helpful to introduce the diagrammatic considerations outlined in Fig. 2. Each horizontal thin line of Fig. 2 represents a *layer*, so that all lattice points on the same line have the same value of the Cartesian coordinate z. The two special spins s_0 and s_i of (2.29) are situated at the origin and at $\vec{r}_i$, respectively; these two sites are indicated by crosses in the diagrams of Fig. 2. Since $\mathcal{H}_1$ consists only of products of pairs of spins situated on *adjacent* layers, it is convenient to denote each such product by a vertical bond (the thick vertical lines of Fig. 2).

Thus, for example, in Figs. 2(a)–2(d) we show situations in which the vector $\vec{r}_i$ extends to the layer above the layer containing the origin, (i.e., $z_i=1$). In Fig. 2(a) there is one vertical arrow, corresponding to the term $s_{\vec{u}_j,0} s_{\vec{u}_j,1}$ of $\mathcal{H}_1$. In Fig. 2(b),

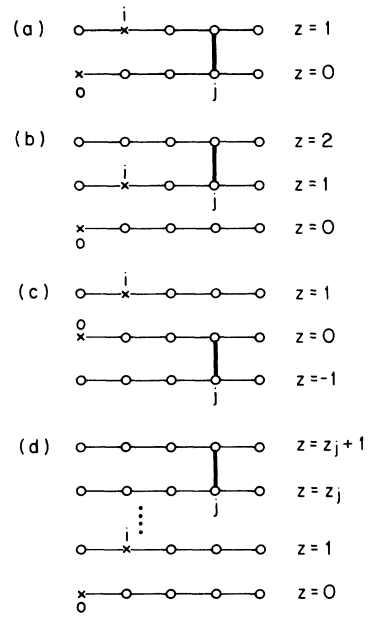

FIG. 2. Possible configurations obtained from placing one R bond on the lattice, with spins s_0 and s_i being on the $z_0=0$ and $z_i=1$ layers, respectively. The spins s_0 and s_i are denoted by X, while the small circles denote lattice sites. Each horizontal line symbolizes a layer whose z coordinate is given in the right-hand margin. The vertical dark line is the R bond coming from $\mathcal{H}_1$ in Eq. (2.24). These conformations are those which must be considered in evaluating $C_{2,1}(T,H,\vec{r}_i,R=0)$ for $z_i=1$. See also the discussion in Sec. II.

the spins s_0 and s_i are at the same positions as in Fig. 2(a), but the vertical bond connects another pair of neighboring layers ($s_{\vec{u}_j,1} s_{\vec{u}_j,2}$ of $\mathcal{H}_1$). Similarly, the conformation shown in Fig. 2(c) corresponds to the term $s_{\vec{u}_j,-1} s_{\vec{u}_j,0}$ of $\mathcal{H}_1$. Figure 2(d) represents the final topological possibility for the vertical bond, given the location of the crosses for s_0 and s_i—namely, the vertical bond connects neighboring layers, neither of which has $z=0$ or 1.

Utilizing the diagrammatic representations of Fig. 2 for terms in $\mathcal{H}_1$, we can evaluate the first derivative of the correlation function, (2.29), at $R=0$, for the case in which $z_i=1$ (i.e., the two correlated spins are s_0 and $s_{\vec{u}_k,1}$). The contribution to (2.29) due to configurations of the same topological type as Fig. 2(a) is given by

$$\begin{aligned} A \equiv \mathcal{J}\{&\langle s_0 s_{\vec{u}_i,1} s_{\vec{u}_j,0} s_{\vec{u}_j,1}\rangle_0 - \langle s_0 s_{\vec{u}_i,1}\rangle_0 \langle s_{\vec{u}_j,0} s_{\vec{u}_j,1}\rangle_0 - \langle s_0\rangle_0[\langle s_{\vec{u}_i,1} s_{\vec{u}_j,0} s_{\vec{u}_j,1}\rangle_0 \\ &- \langle s_{\vec{u}_i,1}\rangle_0 \langle s_{\vec{u}_j,0} s_{\vec{u}_j,1}\rangle_0] - \langle s_{\vec{u}_i,1}\rangle_0[\langle s_0 s_{\vec{u}_j,0} s_{\vec{u}_j,1}\rangle_0 - \langle s_0\rangle_0 \langle s_{\vec{u}_j,0} s_{\vec{u}_j,1}\rangle_0]\} \ . \end{aligned} \tag{2.30}$$

The right-hand side of (2.30) may be "decoupled" using Theorem T1, with the result

$$\begin{aligned} A = \mathcal{J}\{&\langle 0j\rangle\langle ij\rangle - \langle 0\rangle\langle i\rangle\langle j\rangle\langle j\rangle \\ &- \langle 0\rangle\langle j\rangle[\langle ij\rangle - \langle i\rangle\langle j\rangle] \\ &- \langle i\rangle\langle j\rangle[\langle 0j\rangle - \langle 0\rangle\langle j\rangle]\} \ , \end{aligned} \tag{2.31}$$

where we have introduced the notation

$$\langle s_0 s_{\vec{u}_i} s_{\vec{u}_j}\rangle_d \equiv \langle 0ij\rangle \tag{2.32}$$

for the two-dimensional average. Making the obvious cancellations in (2.31), we have

$$\begin{aligned} A &= \mathcal{J}\{\langle 0j\rangle - \langle 0\rangle\langle j\rangle\}\{\langle ij\rangle - \langle i\rangle\langle j\rangle\} \\ &= \mathcal{J} C_d(\vec{u}_j) C_d(\vec{u}_i - \vec{u}_j) \ , \end{aligned} \tag{2.33}$$

where

$$C_d(\vec{u}) \equiv \langle s_0 s_{\vec{u}}\rangle_d - \langle s_0\rangle_d \langle s_{\vec{u}}\rangle_d \tag{2.34}$$

is the correlation function of the *two-dimensional* lattice.

The contribution due to configurations of the type shown in Fig. 2(b) is now

$$\begin{aligned} B \equiv \mathcal{J}\{&\langle 0\rangle\langle ij\rangle\langle j\rangle - \langle 0\rangle\langle i\rangle\langle j\rangle\langle j\rangle \\ &- \langle 0\rangle[\langle ij\rangle\langle j\rangle - \langle i\rangle\langle j\rangle\langle j\rangle] \\ &- \langle i\rangle[\langle 0\rangle\langle j\rangle\langle j\rangle - \langle 0\rangle\langle j\rangle\langle j\rangle]\} \ , \end{aligned} \tag{2.35}$$

which sums to zero. Proceeding in exactly the same fashion, one can easily see that the contributions corresponding to Figs. 2(c) and 2(d) are also zero. In other words, for $z_i = 1$, only those vertical bonds (from $\mathcal{H}_1$) which have the topological configuration of Fig. 2(a) will have a nonzero contribution to (2.29), and the contribution is given by (2.33). Thus for the case that $\vec{r}_i = (\vec{u}_i, 1)$ we have the final result

$$C_{2,1}(\vec{r}_i, R=0) = \mathcal{J} \sum_{\vec{u}_j} C_d(\vec{u}_j) C_d(\vec{u}_i - \vec{u}_j) \ . \tag{2.36}$$

From symmetry arguments, it follows that (2.36) also holds for $\vec{r}_i = (\vec{u}_i, -1)$.

For $z_i = 0$, the only configurations which have a nonzero contribution to (2.29) are those shown in Figs. 3(a) and 3(b). The contribution of Fig. 3(a) is

$$\begin{aligned} \langle j\rangle[&\langle 0ij\rangle - \langle 0i\rangle\langle j\rangle - \langle ij\rangle\langle 0\rangle \\ &- \langle 0j\rangle\langle i\rangle + 2\langle i\rangle\langle j\rangle\langle 0\rangle] \ . \end{aligned} \tag{2.37}$$

The contribution of Fig. 3(b) is the same, since Figs. 3(a) and 3(b) are symmetries about the $z = 0$ layer. Summing over $\vec{u}_j$ and observing that in (2.37) $\langle j\rangle \equiv \langle S_{\vec{u}_j}\rangle_d \equiv \overline{M}_d$ is independent of $\vec{u}_j$, we finally obtain, for $\vec{r}_i = (\vec{u}_i, 0)$, the result

$$\begin{aligned} C_{2,1}(\vec{r}_i, R=0) &= 2\mathcal{J}\overline{M}_d \sum_{\vec{u}_j}[\langle 0ij\rangle - \langle 0i\rangle\langle j\rangle \\ &\quad - \langle ij\rangle\langle 0\rangle - \langle 0j\rangle\langle i\rangle + 2\langle i\rangle\langle j\rangle\langle 0\rangle] \\ &= 2\mathcal{J}\overline{M}_d \frac{\partial C_d(\vec{u}_i)}{\partial h} \ . \end{aligned} \tag{2.38}$$

For $z_i \neq 0, 1$ one can easily see that $C_{2,1}(\vec{r}_i, R=0) = 0$. In terms of the graphs of Figs. 2 and 3, this result corresponds to the fact that with only one R bond from $\mathcal{H}_1$, it is impossible to draw a connected graph from one spin s_0 to another $s_{\vec{r}_i}$ if $s_{\vec{r}_i}$ is further than one layer away.

Thus Eqs. (2.36) and (2.38) are sufficient to obtain the first derivative of the pair correlation function for *all* site separations $\vec{r}_i$ when $R = 0$. As a check on these expressions, we use the fluctuation-dissipation result (2.18) to find

$$\begin{aligned} \chi_1(0) &= \sum_{r_i} C_{2,1}(\vec{r}_i, 0) \\ &= 2\mathcal{J} \sum_{\vec{u}_i, \vec{u}_j} C_2(\vec{u}_j) C_d(\vec{u}_i - \vec{u}_j) + 2\mathcal{J}\overline{M}_d \frac{\partial}{\partial h} \sum_{\vec{u}_i} C_d(\vec{u}_i) \\ &= 2\mathcal{J}\overline{\chi}_d^2 + 2\mathcal{J}\overline{M}_d \frac{\partial \overline{\chi}_d}{\partial h} \ , \end{aligned} \tag{2.39}$$

which is exactly (2.22).

To consider the second moment, we utilize the identity

$$\vec{r}_i \equiv \vec{u}_i \pm \hat{z} = \vec{u} \pm \hat{z} + (\vec{u}_i - \vec{u}) \ . \tag{2.40}$$

Hence

$$|\vec{r}_i|^2 = 1 + |\vec{u}|^2 + |\vec{u}_i - \vec{u}|^2 + 2\vec{u}\cdot(\vec{u}_i - \vec{u}) \tag{2.41}$$

for the case that $z_i = \pm 1$. Substituting (2.36), (2.38), and (2.41) into (2.25), we have for

$$\mu_{2,1}(R) \equiv \frac{\partial \mu_2(T, H, R)}{\partial R} \tag{2.42}$$

the expression

$$\mu_{2,1}(R=0) = 2\mathcal{J}\overline{M}_d \frac{\partial}{\partial h} \mu_2(0) + 2\mathcal{J}[\overline{\chi}_d^2 + 2\overline{\chi}_d \mu_2(0)] \ . \tag{2.43}$$

Similarly, it follows from (2.27) that for the first derivative of the structure factor,

$$S_1(\vec{q}, R) \equiv \frac{\partial S(T, H, \vec{q}, R)}{\partial R} \tag{2.44}$$

that

$$\begin{aligned} S_1(\vec{q}, R=0) &= 2\mathcal{J}\overline{M}_d \frac{\partial}{\partial h} S_d(\vec{q}) \\ &\quad + 2\mathcal{J}\cos(\vec{q}\cdot\hat{z}) S_d(\vec{q}) S_d(-\vec{q}) \ . \end{aligned} \tag{2.45}$$

Note that (2.45) reduces to (2.22), in accordance with the fact that $S(0, R) = \overline{\chi}(R)$.

In summary, we have seen that Theorem T1, Eq. (2.6), permits straightforward expressions to

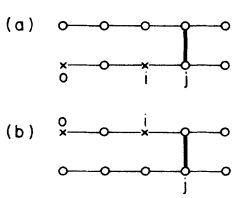

FIG. 3. One-R-bond configurations which have nonzero contributions to $C_{2,1}(\vec{r}_i, 0)$ for $z_i = 0$ (i.e., s_0 and s_i lying in the same layer $z_i = 0$). See definitions in caption to Fig. 2 and discussion in Sec. II.

be obtained for the first derivatives with respect to R of various thermodynamic functions and quantities related to the pair correlation function. The following expressions are thereby obtained: Eq. (2.14) for the free energy, Eq. (2.20) for the magnetization, Eq. (2.22) for the susceptibility, Eqs. (2.36) and (2.38) for the pair correlation function, Eq. (2.43) for the second moment, and Eq. (2.45) for the structure factor.

III. SECOND-ORDER DERIVATIVES WITH RESPECT TO R OF THERMODYNAMIC FUNCTIONS AND OF QUANTITIES RELATED TO THE PAIR CORRELATION FUNCTION

A. Gibbs Potential, Magnetization, and Susceptibility

1. Gibbs Potential

The second-order derivative of the Gibbs potential with respect to R is

$$G_2(R) \equiv \frac{\partial^2 G(T,H,R)}{\partial R^2} = -\beta[\langle \mathcal{H}_1^2\rangle_R - \langle \mathcal{H}_1\rangle_R^2] \ . \tag{3.1}$$

Hence for $R=0$, we have—in analogy to Eq. (2.13)—the result

$$-G_2(R=0) = 2N\beta J^2 \sum_{\vec{u}} \langle s_0\rangle_d^2 [\langle s_0 s_u\rangle_d - \langle s_0\rangle_d^2] + N\beta J^2 \sum_{\vec{u}} [\langle s_0 s_{\vec{u}}\rangle_d^2 - \langle s_0\rangle_d^2 \langle s_{\vec{u}}\rangle_d^2] \ . \tag{3.2}$$

After some rearrangement, (3.2) becomes

$$-G_2(R=0) = 4N\beta J^2 \overline{M}_d^2 \overline{\chi}_d + N\beta J^2 \sum_{\vec{u}} [C_d(\vec{u})]^2 \ . \tag{3.3}$$

2. Magnetization

Differentiating (3.3) with respect to h yields, for the reduced magnetization, the expression

$$\begin{aligned} \mathcal{J}^{-2}\overline{M}_2(0) &\equiv \mathcal{J}^{-2}\left(\frac{\partial^2 M(T,H,R)}{\partial R^2}\right)_{R=0} \\ &= -\mathcal{J}^{-2}\frac{\beta}{N}\left(\frac{\partial G_2(R=0)}{\partial h}\right) \\ &= \Sigma_M + 8\overline{M}_d\overline{\chi}_d^2 + 4\overline{M}_d^2\frac{\partial \overline{\chi}_d}{\partial h} \ , \end{aligned} \tag{3.4}$$

where Σ_M denotes the summation:

$$\begin{aligned} \Sigma_M \equiv \sum_{\vec{u}}\sum_{\vec{u}'} & [\langle s_0 s_{\vec{u}}\rangle_d - \langle s_0\rangle_d\langle s_{\vec{u}}\rangle_d] \\ & \times[\langle s_0 s_{\vec{u}} s_{\vec{u}'}\rangle_d - \langle s_0 s_{\vec{u}}\rangle_d\langle s_{\vec{u}'}\rangle_d - \langle s_0 s_{\vec{u}'}\rangle_d\langle s_{\vec{u}}\rangle_d \\ & - \langle s_{\vec{u}} s_{\vec{u}'}\rangle_d\langle s_0\rangle_d + 2\langle s_0\rangle_d\langle s_{\vec{u}}\rangle_d\langle s_{\vec{u}'}\rangle_d] \ . \end{aligned} \tag{3.5}$$

Although (3.4) is an exact expression, the summation Σ_M is too complicated for us to estimate even its order of magnitude. Therefore, we shall proceed to find the upper and lower bounds of Σ_M in terms of quantities which are more familiar. Specifically, we shall show that

$$0 \geq \Sigma_M \tag{3.6a}$$

and

$$\Sigma_M \geq -4\overline{M}_d\overline{\chi}_d^2 \ . \tag{3.6b}$$

If we can prove (3.6), then we can combine these results with Eq. (3.4) to obtain bounds on $\overline{M}_2(0)$:

$$8\overline{M}_d\overline{\chi}_d + 4\overline{M}_d^2\frac{\partial \overline{\chi}_d}{\partial h} \geq \mathcal{J}^{-2}\overline{M}_2(0) \tag{3.7a}$$

$$\mathcal{J}^{-2}\overline{M}_2(0) \geq 4\overline{M}_d\left(\overline{\chi}_d^2 + \overline{M}_d\frac{\partial \overline{\chi}_d}{\partial h}\right) \ . \tag{3.7b}$$

The proof of (3.6a) and (3.6b) is based upon three rigorous inequalities, which we denote by G1, G2, and G3a, b; they were proved, respectively, by Griffiths,[12] Ginibre,[13] and Griffiths, Hurst, and Sherman.[14]

Theorem G1. (See Ref. 12.) If s_A, s_B are spins on sites A, B or if they denote arbitrary products of spins, then

$$\langle s_A s_B\rangle \geq \langle s_A\rangle\langle s_B\rangle \geq 0 \ . \tag{3.8}$$

In particular, we shall utilize the following special cases of Theorem G1:

$$C_2(T,H,\vec{u},R=0) = \langle s_0 s_{\vec{u}}\rangle_d - \langle s_0\rangle_d\langle s_{\vec{u}}\rangle_d \geq 0 \tag{3.9}$$

and

$$\langle s_0 s_{\vec{u}} s_{\vec{u}'}, s_{\vec{u}''}\rangle_d - \langle s_0 s_{\vec{u}}\rangle_d\langle s_{\vec{u}'}, s_{\vec{u}''}\rangle_d \geq 0 \ . \tag{3.10}$$

Theorem G2. (See Ref. 13.) If s_A, s_B, s_ρ denote single spins or arbitrary products of spins, then

$$\sum_{\{s_\rho\}} \langle s_\rho\rangle\langle s_A s_B s_\rho\rangle - \langle s_A s_\rho\rangle\langle s_B s_\rho\rangle \geq 0 \ , \tag{3.11}$$

where the summation ranges over all elements $\{s_\rho\}$ of a set, whose elements form a group under the operation of multiplication. In particular, if $s_A = s_0 s_{\vec{u}}$, and $s_B = s_{\vec{u}'}$, and $\{s_\rho\}$ is a set consisting of two elements, $\{s_\rho\} = \{1, s_{\vec{u}}\}$, then Theorem G2 yields

$$\langle s_0 s_{\vec{u}} s_{\vec{u}'}\rangle_d - \langle s_0 s_{\vec{u}}\rangle_d\langle s_{\vec{u}'}\rangle_d + \langle s_{\vec{u}}\rangle_d\langle s_0 s_{\vec{u}'}\rangle_d - \langle s_0\rangle_d\langle s_{\vec{u}} s_{\vec{u}'}\rangle_d \geq 0 \tag{3.12}$$

or, equivalently,

$$\begin{aligned} C_3(\vec{u},\vec{u}') &\equiv \langle s_0 s_{\vec{u}} s_{\vec{u}'}\rangle_d - \langle s_0 s_{\vec{u}}\rangle_d\langle s_{\vec{u}'}\rangle_d - \langle s_{\vec{u}}\rangle_d\langle s_0 s_{\vec{u}'}\rangle_d \\ &\quad - \langle s_0\rangle_d\langle s_{\vec{u}} s_{\vec{u}'}\rangle_d + 2\langle s_0\rangle_d\langle s_{\vec{u}}\rangle_d\langle s_{\vec{u}'}\rangle_d \\ &\geq -2\langle s_{\vec{u}}\rangle_d[\langle s_0 s_{\vec{u}'}\rangle_d - \langle s_0\rangle_d\langle s_{\vec{u}'}\rangle_d] \ , \end{aligned} \tag{3.13}$$

where $C_3(\vec{u},\vec{u}')$ is of interest because it is the three-spin correlation function that will appear in our subsequent analysis.

Theorem G3*a*. (See Ref. 14.) If s_i, s_j, s_k denote three arbitrary spins, then

$$0 \geq \langle s_i s_j s_k\rangle - \langle s_i s_j\rangle\langle s_k\rangle - \langle s_i s_k\rangle\langle s_j\rangle - \langle s_j s_k\rangle\langle s_i\rangle + 2\langle s_i\rangle\langle s_j\rangle\langle s_k\rangle \ . \tag{3.14}$$

In particular,

$$0 \geq C_3(\vec{u}, \vec{u}') \ . \tag{3.15}$$

Theorem G3b. (See Ref. 14.) If s_0, s_i, s_j, s_k denote four arbitrary spins, then

$$0 \geq \langle s_0 s_i s_j s_k \rangle - \langle s_0 s_i \rangle \langle s_j s_k \rangle - \langle s_0 s_j \rangle \langle s_i s_k \rangle - \langle s_0 s_k \rangle \langle s_i s_j \rangle \ . \tag{3.16}$$

To apply Theorems G1–G3 in proving (3.6), we begin by observing that the summand of the quantity Σ_M [defined in (3.5)] consists of products of two factors in square brackets. The term in the first square bracket is known by Eq. (3.9) to be positive, while the term in the second square bracket is precisely the function $C_3(\vec{u}, \vec{u}')$ [defined in (3.13)] which was shown to be negative in (3.15). Thus each term in the summand of Σ_M is negative, and (3.6a) is proved.

To prove (3.6b), we first substitute the inequality (3.13) into (3.5) with the result

$$\Sigma_M \geq -2 \sum_{\vec{u}, \vec{u}'} C_d(\vec{u}) \langle s_{\vec{u}} \rangle_d C_d(\vec{u}') \ . \tag{3.17}$$

We may now use the fluctuation-dissipation relation (2.26b) to see that the right-hand side of (3.17) is simply $-2\overline{M}_d \overline{\chi}_d^2$. Thus (3.6b) is proved. With both (3.6a) and (3.6b) established, (3.7a) and (3.7b) follow at once.

3. Susceptibility

Although one may differentiate (3.4) with respect to h to obtain an exact and general expression for $\overline{\chi}_2(R) = (\partial^2 \overline{\chi}/\partial R^2)$, the result is too complicated for one to extract any useful information. However, for $H = 0$ and $T \geq T_c(R = 0)$, thermal averages of products of odd numbers of spins vanish:

$$\langle s_i \rangle_d = \langle s_i s_j s_k \rangle_d = \langle s_i s_j s_k s_l s_m \rangle = \cdots = 0 \ . \tag{3.18}$$

Thus it is straightforward to see that

$$\mathcal{J}^{-2} \overline{\chi}_2(R = 0) = 2\Sigma_2 + 4\overline{\chi}_d^3 \ , \tag{3.19}$$

where

$$\Sigma_2 \equiv \sum_{\vec{u}} \sum_{\vec{u}'} \sum_{\vec{u}''} \langle s_0 s_{\vec{u}} \rangle_d [\langle s_0 s_{\vec{u}} s_{\vec{u}'} s_{\vec{u}''} \rangle_d - \langle s_0 s_{\vec{u}} \rangle_d \langle s_{\vec{u}'} s_{\vec{u}''} \rangle_d] \ . \tag{3.20}$$

From Theorem G1 [Eqs. (3.8) and (3.10)] it follows that the summand of (3.20) is the product of two positive factors and hence we have a lower bound on Σ_2,

$$\Sigma_2 \geq 0 \ . \tag{3.21a}$$

An upper bound may be obtained by observing that Theorem G3b [Eq. (3.16)] implies

$$\langle s_0 s_{\vec{u}'} \rangle_d \langle s_{\vec{u}} s_{\vec{u}''} \rangle_d + \langle s_0 s_{\vec{u}''} \rangle_d \langle s_{\vec{u}} s_{\vec{u}'} \rangle_d \geq \langle s_0 s_{\vec{u}} s_{\vec{u}'} s_{\vec{u}''} \rangle_d - \langle s_0 s_{\vec{u}} \rangle_d \langle s_{\vec{u}'} s_{\vec{u}''} \rangle_d \ , \tag{3.22}$$

and substituting (3.22) into (3.20),

$$\sum_{u} \sum_{u'} \sum_{u''} \langle s_0 s_{\vec{u}} \rangle_d [\langle s_0 s_{\vec{u}'} \rangle_d \langle s_{\vec{u}} s_{\vec{u}''} \rangle_d + \langle s_0 s_{\vec{u}''} \rangle_d \langle s_{\vec{u}} s_{\vec{u}''} \rangle_d \geq \Sigma_2 \ . \tag{3.23}$$

Finally, on using (2.26b), we have the desired upper bound,

$$2[\overline{\chi}_d]^3 \geq \Sigma_2 \ . \tag{3.21b}$$

Combining (3.19) with (3.21a), we obtain a lower bound on $\overline{\chi}_2$,

$$\overline{\chi}_2(0) \geq 4\mathcal{J}^2 [\overline{\chi}_d]^3 \ , \tag{3.24a}$$

while on combining (3.19) with (3.21b), we obtain the upper bound

$$8\mathcal{J}^2 [\overline{\chi}_d]^3 \geq \overline{\chi}_2(0) \ . \tag{3.24b}$$

B. Two-Spin Correlation Function and Correlation Length

The second-order derivative with respect to R of the correlation function $C_2(T, H = 0, \vec{r}_i, R)$,

$$C_{2,2}(\vec{r}_i, R) = \frac{\partial^2 C_2(T, H = 0, \vec{r}_i, R)}{\partial R^2} \ , \tag{3.25}$$

is obtained in a relatively simple form for $H = 0$ and $T \geq T_c(R = 0)$ by using Eq. (3.18), with the result [cf. (2.29)]

$$C_{2,2}(\vec{r}_i, R = 0) \equiv \beta^2 [\langle s_0 s_i \mathcal{H}_1^2 \rangle_{R=0} - \langle s_0 s_i \rangle_{R=0} \langle \mathcal{H}_1^2 \rangle_{R=0}] \ . \tag{3.26}$$

It is now straightforward to show, with the help of Theorem T1 and (3.18), that the only graphical configurations making nonzero contributions to (3.26) are those corresponding to the topological forms shown in Fig. 4. Thus for Fig. 4a, we have

$$\mathcal{J}^{-2} C_{2,2}(\vec{u}_i, z_i = 2, R = 0) = \sum_{z_j, z_k} [\delta(z_j = 0)\delta(z_k = 1) + \delta(z_j = 1)\delta(z_k = 0)] \sum_{\vec{u}_j, \vec{u}_k} \langle s_0 s_{\vec{u}_j} \rangle_d \langle s_{\vec{u}_j} s_{\vec{u}_k} \rangle_d \langle s_{\vec{u}_k} s_{\vec{u}_i} \rangle_d$$

$$= 2 \sum_{\vec{u}_j, \vec{u}_k} \langle s_0 s_{\vec{u}_j} \rangle_d \langle s_{\vec{u}_j} s_{\vec{u}_k} \rangle_d \langle s_{\vec{u}_k} s_{\vec{u}_i} \rangle_d \ . \tag{3.27}$$

For Fig. 4(b), with $z_i = -2$, the contribution to (3.26) is the same as is given in (3.27) for the case $z_i = +2$.

For Figs. 4(c) and 4(d) we have $z_i = 0$. For both configurations, we have

$$\mathcal{J}^{-2} C_{2,2}(\vec{r}_i, R = 0) = 2 \sum_{\vec{u}_j, \vec{u}_k} \langle s_{\vec{u}_j} s_{\vec{u}_k} \rangle_d$$

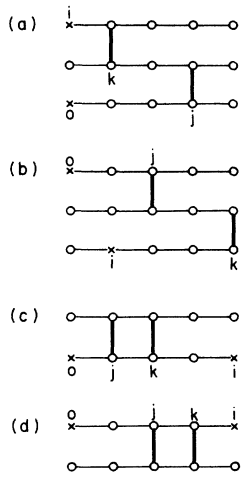

FIG. 4. Two-R-bond configurations which have nonzero contributions to $C_{2,2}(\vec{r}_i,0)$ at $T>T_c$, $H=0$ for (a) $z_i=2$, (b) $z_i=-2$, and (c) and (d) $z_i=0$. See definitions in caption to Fig. 2, and discussion in Sec. III.

$$\times[\langle s_0 s_{\vec{u}_i} s_{\vec{u}_j} s_{\vec{u}_k}\rangle_d - \langle s_0 s_{\vec{u}_i}\rangle_d \langle s_{\vec{u}_j} s_{\vec{u}_k}\rangle_d] , \tag{3.28}$$

where the factor of 2 arises from the fact that $z_j(=z_k)$ may be either zero or -1.

As a check on (3.27) and (3.28), we note that they may be combined with the fluctuation-dissipation relation to recover (3.19).

The second derivative of the second moment of the correlation function is given by

$$\mu_{2,2}(R=0)\equiv\left(\frac{\partial^2\mu_2(R)}{\partial R^2}\right)_{R=0}=\sum_{\vec{r}_i} C_{2,2}(\vec{r}_i,0)|\vec{r}_i|^2 . \tag{3.29}$$

There are only three possible values of z_i; for $z_i=0$, we have simply

$$\vec{r}_i=\vec{u}_i , \tag{3.30}$$

while for $z_i=\pm 2$, we have

$$\vec{r}_i=\vec{u}_j+\vec{u}_k-\vec{u}_j\pm 2\hat{z}+\vec{u}_i-\vec{u}_k , \tag{3.31}$$

so that

$$|\vec{r}_i|^2=|\vec{u}_j|^2+|\vec{u}_k-\vec{u}_j|^2+|\vec{u}_i-\vec{u}_k|^2+4 + 2[\vec{u}_j\cdot(\vec{u}_k-\vec{u}_j)+\vec{u}_j\cdot(\vec{u}_i-\vec{u}_k) + (\vec{u}_k-\vec{u}_j)\cdot(\vec{u}_i-\vec{u}_k)] , \tag{3.32}$$

where $\hat{z}$ denotes a unit vector in the z direction (so that z is orthogonal to all vectors lying in a layer). It is now straightforward to find from (3.32) and (3.27) the contribution to $\mu_{2,2}(R=0)$ of the configuration shown in Figs. 4(a) and 4(b) with $z_i=\pm 2$, which we denote by the symbol $\Sigma'_{\pm 2}$. We get

$$\Sigma'_{\pm 2}\equiv\sum_{\vec{r}_i(z_i=\pm 2)} C_{2,2}(\vec{r}_i,0)|\vec{r}_i|^2 = 4\mathcal{J}^2[3\mu_2(0)\overline{\chi}_d^2+4\overline{\chi}_d^3] . \tag{3.33}$$

In obtaining (3.33) we have used the fact that since $\vec{u}_j$, $(\vec{u}_i-\vec{u}_k)$, and $(\vec{u}_k-\vec{u}_j)$ are independent of each other and since $\sum_{\vec{u}}\vec{u}\langle s_0 s_{\vec{u}}\rangle_d=0$, we can virtually neglect all the inner products on the right-hand side of (3.32).

We next obtain the contribution to $\mu_{2,2}(R=0)$ of the configurations of Figs. 4(c) and 4(d) with $z_i=0$, which we denote by Σ'_0. We note that (3.28)–(3.30) yield

$$\Sigma'_0\equiv\sum_{\vec{r}_i(z_i=0)} C_{2,2}(\vec{r}_i,0)|\vec{r}_i|^2 = 2\mathcal{J}^2\sum_{\vec{u}_i,\vec{u}_j,\vec{u}_k}\langle jk\rangle[\langle 0ijk\rangle-\langle 0i\rangle\langle jk\rangle]|\vec{u}_i|^2 , \tag{3.34}$$

where the notation $\langle jk\rangle$ is defined in Eq. (2.32) above. We can obtain a lower bound on Σ'_0 from Theorem G1, Eq. (3.10),

$$\Sigma'_0\geq 0 . \tag{3.35}$$

To obtain an upper bound, we use Theorem G3b [specifically Eq. (3.22)] to show that

$$2\mathcal{J}^2\sum_{\vec{u}_i\vec{u}_j\vec{u}_k}\langle jk\rangle[\langle 0j\rangle\langle ik\rangle+\langle 0k\rangle\langle ij\rangle]|\vec{u}_i|^2\geq\Sigma'_0 . \tag{3.36}$$

Observe that

$$\sum_{\vec{u}_i\vec{u}_j\vec{u}_k}\langle jk\rangle\langle 0j\rangle\langle ik\rangle|\vec{u}_i|^2=\sum_{(\vec{u}_i-\vec{u}_k)}\sum_{(\vec{u}_k-\vec{u}_j)}\sum_{\vec{u}_j}\langle 0j\rangle\langle jk\rangle\langle ki\rangle|(\vec{u}_i-\vec{u}_k)+(\vec{u}_k-\vec{u}_j)+\vec{u}_j|^2$$
$$=3\mu_2(0)\overline{\chi}_d^2+2\sum\sum\sum\langle 0j\rangle\langle jk\rangle\langle ki\rangle[\vec{u}_j\cdot(\vec{u}_i-\vec{u}_k)+\vec{u}_j\cdot(\vec{u}_k-\vec{u}_i)+(\vec{u}_i-\vec{u}_k)\cdot(\vec{u}_k-\vec{u}_i)] . \tag{3.37}$$

Since $\vec{u}_i-\vec{u}_k$, $\vec{u}_k-\vec{u}_j$, and $\vec{u}_j$ are all independent variables, the three cross terms inside the square brackets of the last expression of (3.37) make no contribution to the final summation. Hence from (3.36) and (3.37) we find the upper bound on Σ'_0,

$$\mathcal{J}^2 4\cdot 3\mu_2(0)\overline{\chi}_d^2\geq\Sigma'_0 . \tag{3.38}$$

Corresponding to the lower and upper bounds on Σ'_0 of Eqs. (3.35) and (3.38) there are lower and upper bounds on the second moment. Thus on substituting (3.33) and (3.35) into (3.29), we find the lower bound

$$\mathcal{J}^{-2}\mu_{2,2}(R=0)\geq 12\mu_2(0)\overline{\chi}_d^2+16\overline{\chi}_d^3 , \tag{3.39a}$$

while on using (3.38) instead of (3.35), we find the upper bound

$$24\mu_2(0)\bar{\chi}_d^2 + 16\bar{\chi}_d^3 \geq \mu_{2,2}(R=0)\mathcal{J}^{-2} \ . \qquad (3.39b)$$

IV. THIRD-ORDER DERIVATIVES WITH RESPECT TO R OF THERMODYNAMIC FUNCTIONS AND OF QUANTITIES RELATED TO THE PAIR CORRELATION FUNCTION

In this section we shall consider only the supercritical region, $T > T_c$, and the case $H = 0$. The magnetization is zero for this region and hence we shall begin our discussion with the pair correlation function. The third derivative with respect to R,

$$C_{2,3}(\vec{r}_i, R) \equiv \frac{\partial^3 C_2(T, H=0, \vec{r}_i, R)}{\partial R^3} \ , \qquad (4.1)$$

is given by an expression analogous to Eq. (3.26) and to the $H=0$, $T > T_c$ value of (2.29),

$$C_{2,3}(r_i, R=0) = -\beta^3[\langle s_0 s_i \mathcal{H}_1^3\rangle_{R=0} - 3\langle s_0 s_i \mathcal{H}_1\rangle_{R=0}\langle \mathcal{H}_1^2\rangle_{R=0}] \ . \qquad (4.2)$$

The only configurations with nonzero contributions to (4.1) are of the topological types shown in Fig. 5. As before, it is convenient to consider different values of z_i *seriatum*.

Case I: $z_i = \pm 3$. From Fig. 5(a) for $z_i = 3$ and Fig. 5(b) for $z_i = -3$, we have

$$\mathcal{J}^{-3}C_{2,3}(\vec{r}_i, R=0) = 3! \sum_{\vec{u}_j, \vec{u}_k, \vec{u}_l} \langle s_0 s_{\vec{u}_j}\rangle_d \times \langle s_{\vec{u}_j} s_{\vec{u}_k}\rangle_d \langle s_{\vec{u}_k} s_{\vec{u}_l}\rangle_d \langle s_{\vec{u}_l} s_{\vec{u}_i}\rangle_d \ , \qquad (4.3)$$

where the factor of 3! is analogous to the factor of 2! in (3.27) and arises from the permutation of the three vertical bonds (representing the interactions of strength RJ).

Case II: $z_i = \pm 1$. From Figs. 5(c), 5(e), and 5(g) for $z_i = 1$ and Figs. 5(d), 5(f), and 5(h) for $z_i = -1$ we have

$$C_{2,3}(\vec{r}_i, R=0) = 3!\ \Sigma_{ce}(\vec{u}_i) + \Sigma_g(\vec{u}_i) \ , \qquad (4.4)$$

where

$$\vec{\Sigma}_{ce}(\vec{u}_i) \equiv \sum_{\vec{u}_j, \vec{u}_k, \vec{u}_l} \langle s_0 s_{\vec{u}_j}\rangle_0 \langle s_{\vec{u}_k} s_{\vec{u}_l}\rangle_0 \times [\langle s_{\vec{u}_i} s_{\vec{u}_j} s_{\vec{u}_k} s_{\vec{u}_l}\rangle_0 - \langle s_{\vec{u}_i} s_{\vec{u}_j}\rangle_0 \langle s_{\vec{u}_k} s_{\vec{u}_l}\rangle_0] \qquad (4.5)$$

represents the contribution to the correlation function for $z_i = 1$ from diagrams 5(c), 5(e), and

$$\vec{\Sigma}_g(\vec{u}_i) \equiv \sum_{\vec{u}_j, \vec{u}_k, \vec{u}_l} \{\langle s_0 s_{\vec{u}_j} s_{\vec{u}_k} s_{\vec{u}_l}\rangle_0 \langle s_{\vec{u}_i} s_{\vec{u}_j} s_{\vec{u}_k} s_{\vec{u}_l}\rangle_0 - 3\langle s_0 s_{\vec{u}_j}\rangle_0 \langle s_{\vec{u}_i} s_{\vec{u}_j}\rangle_0 \langle s_{\vec{u}_k} s_{\vec{u}_l}\rangle_0 \langle s_{\vec{u}_k} s_{\vec{u}_l}\rangle_0\} \qquad (4.6)$$

represents the contribution from diagram 5(g).

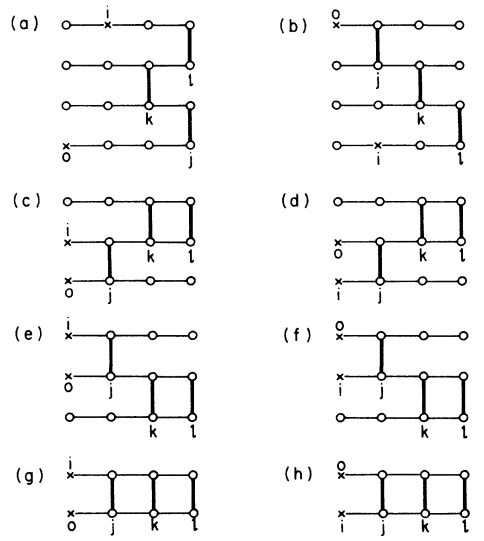

FIG. 5. Three-R-bond configurations that have nonzero contribution to $C_{2,3}(\vec{r}_i, R=0)$ at $T > T_c$, $H=0$ for (a) $z_i = 3$, (b) $z_i = -3$, (c), (e), and (g) $z_i = 1$, and (d), (f), (h) $z_i = -1$. See the definitions in caption to Fig. 2 and the discussion in Sec. IV.

A lower bound on $\Sigma_{ce}(u_i)$,

$$\Sigma_{ce} \geq 0 \ , \qquad (4.7a)$$

follows immediately from Theorem G1 [Eq. (3.10)], while an upper bound,

$$\sum_{\vec{u}_j, \vec{u}_k, \vec{u}_l} \langle 0j\rangle\langle kl\rangle[\langle lj\rangle\langle ik\rangle + \langle li\rangle\langle jk\rangle] \geq \Sigma_{ce}(\vec{u}_i) \ , \qquad (4.7b)$$

arises from Theorem G3b [cf. Eq. (3.22)].

The bounds for the summation $\Sigma_g(\vec{u}_i)$ are somewhat more complicated to derive. We begin by defining the function $A(i,j,k,l)$,

$$A(i,j,k,l) \equiv \langle ij\rangle\langle kl\rangle - \langle ik\rangle\langle jl\rangle + \langle il\rangle\langle kj\rangle \ , \qquad (4.8)$$

and the function $B(\vec{u}_i)$,

$$B(\vec{u}_i) = \frac{1}{2}\sum_{jkl}\{[\langle 0jkl\rangle - A(0,j,k,l)] \times [\langle ijkl\rangle + A(i,j,k,l)] + 0 \longleftrightarrow i\} \qquad (4.9)$$

where the symbol $0 \longleftrightarrow i$ in (4.9) denotes the terms obtained by exchanging the symbols s_0 and $s_{\vec{u}_i}$ everywhere. To relate $\Sigma_g(\vec{u}_i)$ to $B(\vec{u}_i)$, we note that

$$B(\vec{u}_i) = \sum_{j,k,l}\{\langle 0jkl\rangle\langle ijkl\rangle - A(0,j,k,l)A(i,j,k,l)\}$$

$$= \sum_{j,k,l}\{\langle 0jkl\rangle\langle ijkl\rangle - \langle 0j\rangle\langle ij\rangle\langle kl\rangle^2 - \langle 0k\rangle\langle ik\rangle\langle jl\rangle^2 - \langle 0l\rangle\langle il\rangle\langle jk\rangle^2 + 2\langle 0j\rangle\langle jk\rangle\langle kl\rangle\langle li\rangle\} \ . \qquad (4.10)$$

From (4.6) it follows that

$$\Sigma_g(\vec{u}_i) \equiv \sum_{jkl} \{\langle 0jkl\rangle\langle ijkl\rangle - 3\langle 0j\rangle\langle ij\rangle\langle kl\rangle^2\} = B(\vec{u}_i) - 2\sum_{j,k,l}\langle 0j\rangle\langle jk\rangle\langle kl\rangle\langle li\rangle \ . \quad (4.11)$$

To obtain lower and upper bounds for $B(\vec{u}_i)$, we make the following four observations:

Observation (i). From (4.8) we have for the *second* quantity in square brackets in (4.9),

$$\langle ijkl\rangle + A(i,j,k,l) = \langle ijkl\rangle + \langle ij\rangle\langle kl\rangle - \langle ik\rangle\langle jl\rangle + \langle il\rangle\langle kj\rangle \ . \quad (4.12)$$

On using the result of Theorem G1 that quantities of the form of $\langle ij\rangle$ are always positive,

$$\langle ijkl\rangle + A(i,j,k,l) \geq \langle ijkl\rangle - \langle ik\rangle\langle jl\rangle \geq 0 \ , \quad (4.13a)$$

where the last inequality follows from (3.10).

Observation (ii). Using a relation analogous to (3.22) to "eliminate" the expression $\langle ijkl\rangle - \langle ik\rangle\langle jl\rangle$ in (4.12), we get the following upper bound for (4.12):

$$2\langle ij\rangle\langle kl\rangle + 2\langle il\rangle\langle kj\rangle \geq \langle ijkl\rangle + A(i,j,k,l) \ . \quad (4.13b)$$

Observation (iii). The quantity appearing in the *first* curly brackets of (4.10),

$$\langle ijkl\rangle - A(i,j,k,l) = \langle ijkl\rangle - \langle ij\rangle\langle kl\rangle + \langle ik\rangle\langle jl\rangle - \langle il\rangle\langle kj\rangle \ , \quad (4.14)$$

obeys the inequality

$$\langle ijkl\rangle - A(i,j,k,l) \geq 0 \ . \quad (4.15a)$$

This inequality follows from Theorem G2 by choosing $s_A \equiv s_i s_j$, $s_B \equiv s_k s_l$, and $\{s_p\} = \{1, s_j s_k\}$.

Observation (iv). From (4.14) we have, on using the same reasoning as in (ii), that

$$2\langle ik\rangle\langle jl\rangle \geq \langle ijkl\rangle - A(i,j,k,l) \ . \quad (4.15b)$$

Note that (4.13a), and (4.13b) and (4.15a), and (4.15b) are also valid when i is replaced by 0. Thus from (4.13a), (4.15a), and (4.9), we have the desired lower bound on $B(\vec{u}_i)$,

$$B(\vec{u}_i) \geq 0 \ . \quad (4.16a)$$

Substituting (4.13b) and (4.15b) into (4.9), we obtain the desired upper bound

$$2\sum_{jkl}\{[\langle ij\rangle\langle kl\rangle + \langle il\rangle\langle kj\rangle] \times \langle 0k\rangle\langle jl\rangle + i \longrightarrow 0\} \geq B(\vec{u}_i) \ ,$$

or simply

$$8\sum_{jkl}\langle 0j\rangle\langle jk\rangle\langle kl\rangle\langle li\rangle \geq B(\vec{u}_i) \ . \quad (4.16b)$$

Finally, we utilize the bounds (4.16a) and (4.16b) on $B(\vec{u}_i)$ together with (4.11) and the bounds (4.7a), and (4.7b) on Σ_{ce} to obtain the desired lower and upper bounds on $C_{2,3}$ of (4.4); for the case of $z_i = \pm 1$,

$$\mathcal{J}^{-3}C_{2,3}(r_i, R=0) \geq -2\sum_{u_j,u_k,u_l}\langle s_0 s_{u_j}\rangle\langle s_{u_j}s_{u_k}\rangle_d \times \langle s_{u_k}s_{u_l}\rangle_d\langle s_{u_l}s_{u_i}\rangle_d \quad (4.17a)$$

and

$$18\sum_{u_j,u_k,u_l}\langle s_0 s_{u_j}\rangle_d\langle s_{u_j}s_{u_k}\rangle_d\langle s_{u_k}s_{u_l}\rangle_d\langle s_{u_l}s_{u_i}\rangle \geq \mathcal{J}^{-3}C_{2,3}(\vec{r}_i R=0) \ . \quad (4.17b)$$

Using (4.3) for case I and the bounds of (4.17a), and (4.17b) for case II, we can in principle present bounds on all functions obtainable from the zero-field pair correlation function. While in Secs. II and III we derived all statements about the susceptibility derivatives first from differentiation of the field-dependent Gibbs potential, here we obtain bounds on the zero-field susceptibility derivative for $T > T_c$ by direct summation of the correlation function,

$$\chi_3(R=0) = \frac{\partial^3\chi(T,H=0,R)}{\partial R^3}\bigg|_{R=0} = \sum_{\vec{r}_i} C_{2,3}(\vec{r}_i, R=0) \ . \quad (4.18)$$

Combining (4.18) with (4.3) and (4.17a) and (4.17b), and using (2.26b), we have the lower and upper bounds

$$\overline{\chi}_3(0) \geq 8\mathcal{J}^3\overline{\chi}_d^4 \quad (4.19a)$$

and

$$48\mathcal{J}^3\overline{\chi}_d^4 \geq \overline{\chi}_3(0) \quad (4.19b)$$

To discuss the third-order derivatives with respect to R of the second moment,

$$\mu_{2,3}(R=0) \equiv \frac{\partial^3\mu_2(T,H=0,R)}{\partial R^3}\bigg|_{R=0} = \sum_{\vec{r}_i}|\vec{r}_i|^2 C_{2,3}(\vec{r}_i, R=0) \ , \quad (4.20)$$

one must carefully distinguish cases I and II. For case I ($z_i = \pm 3$),

$$\vec{r}_i = 3\hat{z} + \vec{u}_i = 3\hat{z} + (\vec{u}_i - \vec{u}_l) + (\vec{u}_l - \vec{u}_k) + (\vec{u}_k - \vec{u}_j) + \vec{u}_j \ . \quad (4.21)$$

Combining (4.3) and (4.21), we obtain

$$\sum_i^{\mathrm{I}}|\vec{r}_i|^2 C_{2,3}(\vec{r}_i, R=0) = 2\cdot 3!\,\mathcal{J}^3[9\overline{\chi}_d^4 + 4\overline{\chi}_d^3\mu_2(0)] \ . \quad (4.22)$$

For case II ($z_i = \pm 1$), Eq. (4.21) is replaced by

$$\vec{r}_i = \hat{z} + \vec{u}_i = \hat{z} + (\vec{u}_i - \vec{u}_l) + (\vec{u}_l - \vec{u}_k) + (\vec{u}_k - \vec{u}_j) + \vec{u}_j \ . \quad (4.23)$$

Combining (4.23) with the lower and upper bounds of (4.17a) and (4.17b), we get

$$\sum_i^{\mathrm{II}}|\vec{r}_i|^2 C_{2,3}(\vec{r}_i, R=0) \geq -4\mathcal{J}^3[\overline{\chi}_d^4 + 4\overline{\chi}_d^3\mu_2(0)] \quad (4.24a)$$

and

$$36\mathcal{J}^3[\bar{\chi}_d^4+4\bar{\chi}_d^3\mu_2(0)]\geq\sum_i|\vec{r}_i|^2C_{2,3}(\vec{r}_i,R=0)\ , \tag{4.24b}$$

where, as noted in Sec. III, the new summation variables are $(\vec{u}_i-\vec{u}_l)$, $(\vec{u}_l-\vec{u}_k)$, $(\vec{u}_k-\vec{u}_j)$, and $\vec{u}_j$. The inner products between any two of these summation variables will be zero after taking the summation.

Finally, on combining the results (4.22) and (4.24a) and (4.24b) for cases I and II respectively, we obtain the desired lower and upper bounds for the second-moment derivative defined in (4.20),

$$\mu_{2,3}(0)\geq\mathcal{J}^3[104\bar{\chi}_d^4+32\bar{\chi}_d^3\mu_2(0) \tag{4.25a}$$

and

$$48\mathcal{J}^3[3\bar{\chi}_d^4+4\bar{\chi}_d^3\mu_2(0)]\geq\mu_{2,3}(0)\ . \tag{4.25b}$$

V. SUMMARY OF RESULTS AND POSSIBLE GENERALIZATION TO OTHER SYSTEMS

In Secs. II, III, and IV we have examined, respectively, the first, second, and third derivatives with respect to R of various thermodynamic functions and quantities related to the two-spin correlation function. Our principal results are summarized in Table I, using notation defined in Table II. As emphasized above, these results were derived only for the special case of a system of spins situated on a simple cubic lattice ($\bar{d}=3$, $d=2$) and interacting with their nearest neighbors through a $S=\frac{1}{2}$ Ising interaction. This system [cf. Fig. 1(a)] is appropriate for the recent numerical calculations concerning the square → sc crossover.

In this section we shall discuss how certain of these results may be easily generalized to other systems. Specifically, we shall treat the generalization to (a) lattice pairs other than the square and sc, (b) systems for which the perturbation term $R\mathcal{H}_1$[cf. Eq. (2.1)] contains other than nearest-neighbor interactions, and (c) systems in which the spin-spin interaction is other than that described by the $S=\frac{1}{2}$ Ising model. Finally, we shall comment on the plausibility of extending our results to higher-order derivatives (and in the Appendix we shall present a rigorous analysis of the self-avoiding walk problem for general nth order derivatives).

It is important to emphasize that the range of validity of our results is not necessarily restricted to the special systems considered here; for example, if Theorems G1, G2, and G3a, b were to be valid for a wider class of Hamiltonians than the class for which they have thus far been proved, then certain of our results would be correspondingly generalized.

A. Lattice Pairs Other Than Square and Simple Cubic

All the theorems used in our analysis are lattice independent and therefore analogs of all the fundamental relations proved above may be obtained for other lattice pairs that can be related by some anisotropic term $R\mathcal{H}_1$ in the Hamiltonian [cf. Eq. (2.1)] which connects an assembly of d-dimensional systems to form a $\bar{d}$-dimensional system. The final formulas are altered only by the multiplicative factors shown in Table II.

To appreciate the origin of these multiplicative factors we consider first the case $n=1$ (first derivatives with respect to R). Let $g_1(\lambda\rightarrow\bar{\lambda})$ be the number of "RJ bonds" at a lattice site—i.e., the number of additional nearest neighbors that a site acquires when the lattice anisotropy term in (2.1) transforms the lattice λ from being d-dimensional to a lattice $\bar{\lambda}$ which is $\bar{d}$-dimensional. For the sake of convenience, we shall denote $g_1(\lambda\rightarrow\bar{\lambda})$ by simply g_1. The total number of nearest-neighbor bonds with strength RJ is $\frac{1}{2}(g_1)N$; this number is $\frac{1}{2}(2N)=N$ for the square → sc crossover system. Thus Eq. (2.14) for G_1—and hence each equation for quantities obtained by differentiating G_1—should be multiplied by a factor $\frac{1}{2}g_1$. To explain the occurrence of the factor g_1 in the quantities derived from the correlation function, we note that for each of the topological configurations shown in Figs. 2 and 3, there are g_1 ways to place the single "RJ bond", and for each way of drawing this bond the contribution to the correlation function is the same.

Consider next the case $n=2$ (second derivatives). The generalization here is somewhat more subtle than an over-all multiplicative factor, and we must consider in some detail the topological configurations of Fig. 4. In Figs. 4(a) and 4(b) the two RJ bonds are connecting three consecutive layers, while in Figs. 4(c) and 4(d) they are connecting the same pair of layers. There will be different multiplicative factors for each configuration. The factor $g_{21}(\lambda\rightarrow\bar{\lambda})=g_{21}$ associated with the graphs of Figs. 4(a) and 4(b) is equal to the number of ways of placing two consecutive RJ bonds without returning to the original layer. In particular, note that for the linear chain → sc crossover, $g_{21}(\text{lc}\rightarrow\text{sc})=(4)(3)=12$ is just the two-step self-avoiding walk problem on a square lattice.

Similarly, the factor g_{22} associated with Figs. 4(c) and 4(d) is the number of possible ways of placing two consecutive RJ bonds in such a fashion that one returns to the original layer.

Using these considerations, the formulas for $n=2$ become somewhat more complex. In particular, the bounds on $\bar{\chi}_2(R=0)$ become

$$\bar{\chi}_2(0)\geq 4\mathcal{J}^2g_{21}\bar{\chi}_d^3\ , \tag{5.1a}$$

$$4\mathcal{J}^2[g_{21}+g_{22}]\bar{\chi}_d^3\geq\bar{\chi}_2(0)\ . \tag{5.1b}$$

Finally, we consider the case $n=3$ (third-order derivatives). Here the argument is entirely analogous to the case $n=2$, except that there are three

TABLE I. Summary of results derived in this work. Shown in column 2 is the equation number where the formula is given for the special case of a square → sc crossover, and given in columns 3, 4 are the *general-lattice* expressions. In the last columns we give the range of validity and the theorems used in deriving the bounds. Theorems T1, G1, G2, G3a, b are given in Eqs. (2.6), (3.8), (3.11), (3.14), and (3.16), respectively. Note that lines 1–5 for the first derivatives are equalities rather than bounds.

	Physical quantity	Equation	Lower bound	Upper bound	Range of (T,H)	Theorem used: lower bound	Theorem used: upper bound
1.	$\left(\frac{\partial}{\partial R}G(T,H,R)\right)_{R=0}$	(2.14)	$\frac{1}{2}Ng_1 J\bar{M}_d^2$		Arbitrary	T1	
2.	$\left(\frac{\partial}{\partial R}C_H(T,H,R)\right)_{R=0}$		$-\frac{1}{2}Ng_1 JT\frac{\partial^2}{\partial T^2}[\bar{M}_d^2]$		Arbitrary	T1	
3.	$\left(\frac{\partial}{\partial R}\bar{M}(T,H,R)\right)_{R=0}$	(2.20)	$g_1\mathcal{J}\bar{M}_d\bar{\chi}_d$		Arbitrary	T1	
4.	$\left(\frac{\partial}{\partial R}\mu_2(T,H,R)\right)_{R=0}$	(2.43)	$g_1\mathcal{J}\bar{M}_d\frac{\partial}{\partial h}\mu_2(0)+g_1\mathcal{J}\bar{\chi}_d[\bar{\chi}_d+2\mu_2(0)]$		Arbitrary	T1	
5.	$\left(\frac{\partial}{\partial R}S(T,H,\vec{q},R)\right)_{R=0}$	(2.45)	$g_1\mathcal{J}\bar{M}_d\frac{\partial S_d(q)}{\partial h}+g_1\mathcal{J}\cos(\vec{q}\cdot z)S_d(\vec{q})S_d(-\vec{q})$		Arbitrary	T1	
5a.	$\left(\frac{\partial}{\partial R}\bar{\chi}(T,H,R)\right)_{R=0}$	(2.22) (2.39)	$g_1\mathcal{J}\bar{M}_d\frac{\partial}{\partial h}\bar{\chi}_d+g_1\mathcal{J}\bar{\chi}_d^2$		Arbitrary	T1	
6.	$\left(\frac{\partial^2}{\partial R^2}\bar{M}(T,H,R)\right)_{R=0}$	(3.7a) (3.7b)	$2g_{21}\mathcal{J}^2\bar{M}_d\bar{\chi}_d^2+(g_{21}+g_{22})\mathcal{J}^2\bar{M}_d^2\frac{\partial\bar{\chi}_d}{\partial h}$	$(g_{21}+g_{22})\mathcal{J}^2\left(2\bar{M}_d\bar{\chi}_d^2+\bar{M}_d^2\frac{\partial\bar{\chi}_d}{\partial h}\right)$	Arbitrary	T1, G2	T1, G3a
7.	$\left(\frac{\partial^2}{\partial R^2}\bar{\chi}(T,H=0,R)\right)_{R=0}$	(3.24a) (3.24b)	$2g_{21}\mathcal{J}^2\bar{\chi}_d^3$	$2(g_{21}+g_{22})\mathcal{J}^2\bar{\chi}_d^3$	$T>T_c$, $H=0$	T1, G1	T1, G3b
8.	$\left(\frac{\partial^2}{\partial R^2}\mu_2(T,H=0,R)\right)_{R=0}$	(3.39a) (3.39b)	$2g_{21}\mathcal{J}^2[3\mu_2(0)+4\bar{\chi}_d]\bar{\chi}_d^2$	$2(g_{21}+g_{22})\mathcal{J}^2[3\mu_2(0)\bar{\chi}_d^2]+8g_{21}\mathcal{J}^2\bar{\chi}_d^3$	$T>T_c$, $H=0$	T1, G1	T1, G3b
9.	$\left(\frac{\partial^3}{\partial R^3}\bar{\chi}(T,H=0,R)\right)_{R=0}$	(4.19a) (4.19b)	$(6g_{31}-2g_{33})\mathcal{J}^3\bar{\chi}_d^4$	$(6g_{31}+12g_{32}+6g_{33})\mathcal{J}^3\bar{\chi}_d^4$	$T>T_c$, $H=0$	T1, G2	T1, G3b
10.	$\left(\frac{\partial^3}{\partial R^3}\mu_2(T,H=0,R)\right)_{R=0}$	(4.25a) (4.25b)	$(6g_{31}-2g_{33})4\mathcal{J}^3\bar{\chi}_d^3\mu_2(0)+(54g_{31}-2g_{33})\mathcal{J}^3\bar{\chi}_d^4$	$(6g_{31}+12g_{32}+6g_{33})4\mathcal{J}^3\bar{\chi}_d^3\mu_2(0)+(54g_{31}+12g_{32}+6g_{33})\mathcal{J}^3\bar{\chi}_d^4$	$T>T_c$, $H=0$	T1, G2	T1, G3b

TABLE II. Expressions for the lattice factors $g(\lambda \rightarrow \bar{\lambda})$ for several possible pairs of lattices λ and $\bar{\lambda}$ in order that the reader can use Table I to easily calculate the upper and lower bounds for a wide variety of lattice crossovers $\lambda \rightarrow \bar{\lambda}$). (Square, sq; simple cubic, sc; linear chain, lc).

	Conformations	sq → sc	sq → fcc	lc → sc	lc → sq
g_1	Figs. 2 and 3	2	8	4	2
g_{21}	Figs. 4(a) and 4(b)	2	32	12	2
g_{22}	Figs. 4(c) and 4(d)	2	32	4	2
g_{31}	Figs. 5(a) and 5(b)	2	128	36	2
g_{32}	Figs. 5(c), 5(d), 5(e), and 5(f)	2	128	12	2
g_{33}	Figs. 5(g) and 5(h)	2	128	4	2

factors instead of two. Thus the factor g_{31} corresponds to Figs. 5(a) and 5(b) and is the number of ways of placing three consecutive RJ bonds without returning to any layer, and again for the particular case of a linear chain → sc crossover, g_{31} is the self-avoiding walk on a square lattice [g_{31}(lc → sc = (4)(3)(3) = 36 for a three-step self-avoiding walk]. The factor g_{32} corresponds to Figs. 5(c)–5(f) and is the number of ways of placing three consecutive RJ bonds such that the second and third bonds join the same pair of layers. Lastly, the factor g_{33} corresponds to Figs. 5(g) and 5(h) and is the number of ways of placing three RJ bonds such that they all join the same two layers.

B. Interaction Hamiltonians Involving Other Than Nearest Neighbors

The second generalization of the above results concerns interaction Hamiltonians that include other than only nearest-neighbor interactions Since changing the unperturbed Hamiltonian $\mathcal{H}_0$ does not change any formal results, the only terms which affect the results are those in the perturbation Hamiltonian $\mathcal{H}_1$. For example, if $\mathcal{H}_1$ contains g nearest-neighbor pairs of spin products with coupling strength RJ and g' second nearest-neighbor pairs with coupling strength RJ', then multiplicative factors of $2J$ for the first-derivative results of Sec. II are changed to multiplicative factors of $(gJ+g'J')$. The appropriate changes in the results of Secs. III and IV are also easily obtainable using similar reasoning.

C. Interaction Hamiltonian Other Than $S = \frac{1}{2}$ Ising Model

The third direction of generalization is to consider models other than the $S = \frac{1}{2}$ Ising model. Clearly the results of Sec. II–IV apply for any model Hamiltonian for which the theorems utilized in deriving that result are valid. Therefore, we have included in the last column of Table I a listing of which theorems were utilized in obtaining each result.

All of the results of Sec. II for first derivatives with respect to R require for their proof only Theorem T1. This result holds for an extremely wide class of interaction Hamiltonians. In particular, if our perturbation Hamiltonian is

$$\mathcal{H}_1 = -\sum_{\langle ij \rangle} \sum_{\mu=1}^{D} J_\mu S_i^{(\mu)} S_j^{(\mu)} , \tag{5.2}$$

then we should have

$$G_1(R=0) = -N \sum_{\mu=1}^{D} J_\mu \langle S^{(\mu)} \rangle^2 ; \tag{5.3}$$

similar changes affect the other results of Sec. II. Here D is the dimensionality of a classical spin vector $\vec{S} \equiv [S^{(1)}, S^{(2)}, \ldots, S^{(D)}]$; for the *isotropic* limit of (5.2) in which $J_\mu = J$ for all $\mu = 1, 2, \ldots, D$, then (5.2) reduces to the $S = \frac{1}{2}$ Ising, $S = \infty$ planar, $S = \infty$ Heisenberg, and spherical[6] models, respectively, for $D = 1, 2, 3$, and ∞.

Many of the results of Secs. III and IV for the higher-order derivatives necessitated for their proofs Theorems G1, G2, and G3a,b. Theorem G1 is knowns to be valid for the general spin Ising model,[15] and so are G3a and G3b.[16] However, to the best of our knowledge Theorem G2 has been proved only for the $S = \frac{1}{2}$ Ising model. Thus the specific ranges of validity of our results may easily be known by consulting the last column of Table I.

It is worth emphasizing that to the extent that the certain of theorems are plausible assumptions for a wider range of model Hamiltonians, our results can be regarded as being as plausible as the theorems underlying them.

D. Possible Validity of Relations for Derivatives of Order Higher Than Three

The last, and probably the most difficult, generalization is to the case of derivatives of order n higher than three. Although we have not succeeded in finding bounds for $n > 3$, it is not difficult to find *both upper and lower bounds for general n* of a closely related system, the self-avoiding walk

(SAW) problem. This analysis is carried out in the Appendix.

We shall see in Sec. VI that the scaling hypothesis, when stated in its weakest form, implies the validity of the results for general n, and this prediction has been confirmed numerically through $n=5$ for the $S=\frac{1}{2}$ Ising model for both the susceptibility and second moment, and through order $n=8$ for the specific heat.[17]

VI. DISCUSSION AND APPLICATION OF RESULTS

In this final section, we shall briefly discuss some of the possible applications of our results. In Sec. VI A we illustrate the application of our relations for first derivatives as a checking method for high-temperature series. In Sec. VI B the implications of our results in providing a check of scaling theory are considered. Lastly, in Sec. VI C we consider possible applications of the present results to experimental systems. In particular, we shall discuss (i) the concept of a "crossover temperature," (ii) how our results may possibly be used to determine the upper bound for the anisotropy parameter R, and (iii) the plausibility of observing an exponent change as $T \to T_c(R_1)$ in a system with a fixed (small) value of R, $R=R_1$ [cf. Fig. 6(a)].

A. Checking Procedure for High-Temperature Series Expansions

Most of our information concerning critical phenomena in model systems (except those few that can be solved exactly) has come from extrapolations based upon exactly calculated coefficients in series expansions. For example, the high-temperature expansion, in powers of $\mathcal{J} \equiv \beta J \equiv J/kT$, of the zero-field reduced susceptibility for the simple cubic lattice ($R=1$) is of the form

$$\bar{\chi}(R=1)=\sum_{l=1}^{\infty} A_l \mathcal{J}^l \ , \tag{6.1}$$

where the coefficients in the expansion A_l may be related to the number of graphs on the lattice containing l "J bonds." For an anisotropic lattice ($R \neq 1$), each graph may have $0, 1, 2, \ldots, n$ bonds in the R direction ("RJ bonds"). Thus each coefficient a_l in (6.1) is, in general, a polynomial of lth degree in R, $A_l = a_{l0} + a_{l1}R + \cdots + a_{ll}R^l$ and (6.1) becomes

$$\bar{\chi}(R)=\sum_{l=0}^{\infty}\sum_{m=0}^{l} a_{lm} R^m \mathcal{J}^l \ . \tag{6.2}$$

The coefficients a_{lm} in (6.2) have recently been obtained through order $l=10$ and 11 various Ising model lattices ($D=1$),[11,18,19] through order $l=10$ for the planar model ($D=2$),[20] and through order $l=10$ for the Heisenberg model ($D=3$).[20] Since the calculation of the coefficients has historically resulted in numerous errors,[21] it is important to have checking methods available. To our knowledge, the methods we present below are the only checks usable on the series (6.2).

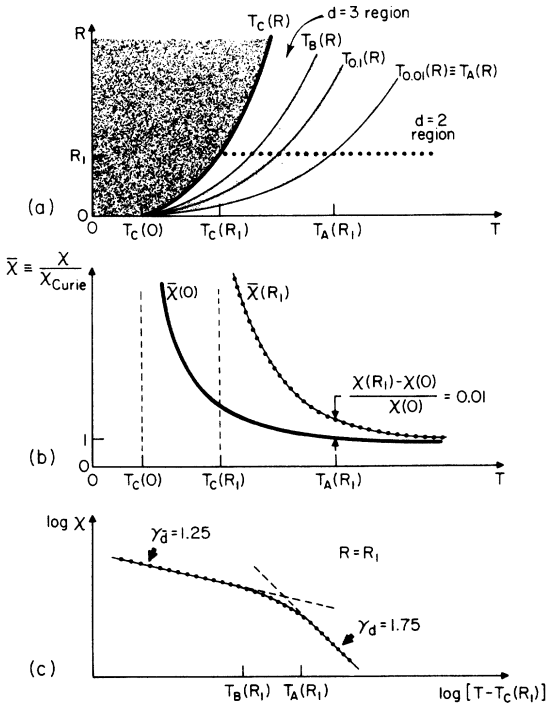

FIG. 6. Schematic diagram of the crossover behavior. (a) The crossover region (shaded area) is bounded by $T_A(R)$ and $T_B(R)$. [$T_A(R) = T_{0.01}(R)$ is the temperature at which the system differs appreciably (1%) from being two dimensional.] $T_c(R)$ is the critical temperature. The generalized scaling hypothesis predicts that all curves should approach $T_c(0)$ via the power law $R^{1/\varphi}$. (b) Dependence of reduced susceptibility $\bar{\chi}$ upon T for $R=0$ and for $R=R_1$, indicating the definition of $T_A(R)$. Note that this drawing is not to scale. (c) Sketch of hypothetical experimental data, plotted in the conventional log-log plot, for a system which is described by the Hamiltonian with $R=R_1$.

1. Limiting Results at $R=0$, ∞, and 1 (First Column, Main Diagonal, and Sum of Each Row)

For $R=0$, the system is d dimensional and the series are generally known. Hence we have

$$\bar{\chi}(0)=\sum_{l=0}^{\infty} a_{l0}\mathcal{J}^l = \bar{\chi}_d \ . \tag{6.3a}$$

Similarly, for $R=\infty$ the lattice reduces to a known case. For example, the sc lattice reduces to uncoupled linear chains, and we have

$$\bar{\chi}(\infty)=\sum_{l=0}^{\infty} a_{ll}(R\mathcal{J})^l = \bar{\chi}_{1c} \ . \tag{6.3b}$$

Finally, for $R=1$ we have the isotropic case of (6.1) and hence we have the "sum rule"

$$A_l = \sum_{m=0}^{l} a_{lm} \quad . \tag{6.3c}$$

Thus if the coefficients a_{lm} are arranged in a triangular array with the rows labeled by the index l and the columns by m ($m = 0, 1, 2, \ldots, l$) (cf. Tables I–IV of Ref. 19), then (6.3a) permits one to check the first column, (6.3b) the main diagonal, and (6.3c) the sum of the terms in each row.

2. First-Derivative Results (Second Column)

From Sec. II and the general-lattice discussion of Sec. V (cf. Table I), it follows that the first-order derivative of $\bar{\chi}(R)$ at $R = 0$ is exactly $g_1(\bar{\chi}_d)^2$. Hence we have a very powerful check on the entries of the second column of Tables I–IV of Ref. 19:

$$\sum_{l=1}^{\infty} a_{l,1}\mathcal{J}^l = g_1\left(\sum_{l=0}^{\infty} a_{l,0}\mathcal{J}^l\right)^2 \quad . \tag{6.3d}$$

As $R \to \infty$, the sc lattice reduces to uncoupled linear chains and we can also check the diagonal just below the main diagonal,

$$\sum_{l=1}^{\infty} a_{l,l-1}(R\mathcal{J})^{l-1} = 4[\bar{\chi}_{lc}]^2 \quad . \tag{6.3e}$$

To check the second column of the second moment series, one needs the somewhat more complicated result of line 4, Table I.

We have used the recently calculated series expansion coefficients to verify this relation for the second moment—together with relations (6.3a)–(6.3e) for the susceptibility—for the Ising, planar, and Heisenberg models ($D = 1$–3).

3. System with Both First- and Second-Neighbor Interactions

Another example is a system with both nearest-neighbor (nn) and next-nearest-neighbor (nnn) interactions. Consider a magnetic system on a square (sq) lattice with nn distance c and with nn coupling strength RJ and nnn coupling strength J. In the limit $R \to 0$, this system reduces to two interpenetrating independent sq lattices, for each of which the nn distance is $\sqrt{2}c$ and the nn coupling strength is J. Thus for this system the first-order derivative relations of Table I apply with $g_1 = 4$. General R series for systems with arbitrary nn and nnn interactions have been obtained for various systems, and these relations corroborated numerically.[22]

4. Application to Baxter and Ashkin-Teller Models

Lastly, it is worth mentioning that similar relations are always obtainable providing there exists a certain RJ bond that couples two (or more) independent lattice systems. One such example was given above in Sec. VI A 3. Another example is provided by the eight-vertex model[23] (Baxter model) and the four-state Ising model (Ashkin-Teller model). These have been shown to be equivalent to two interpenetrating regular Ising systems coupled together by a four-spin interaction term $Rs_{1i}s_{1j}s_{2i}s_{2j}$, where s_{1i}, s_{1j} are spins on one lattice and s_{2i}, s_{2j} are spins on the other lattice. At $R = 0$, these models decouple into two independent regular Ising systems. Hence it is not difficult to show for the Baxter-model susceptibility $\bar{\chi}_B(R)$ that

$$\left(\frac{\partial \bar{\chi}_B(R)}{\partial R}\right)_{R=0} \propto \left(\frac{\partial \bar{\chi}}{\partial T}\right) U \tag{6.4}$$

where in this equation $\bar{\chi}$ and U are, respectively, the susceptibility and internal energy (i.e., the nn correlation function) for the $R = 0$ system.[24]

B. Scaling with Respect to Anisotropy Parameter R

1. Thermodynamic Functions

Historically the scaling hypothesis has proved to be a valuable qualitative guide in our attempt to understand critical phenomena. Recently this hypothesis has been extended to treat systems that depend upon a parameter (the anisotropy parameter R in this case).[7] In the formulation of Ref. 9, we assume that there exist three numbers a_τ, a_H, and a_R, such that for all positive λ,

$$G(\lambda^{a_\tau}\tau,\ \lambda^{a_H}H,\ \lambda^{a_R}R) = \lambda\ G(\tau, H, R) \quad , \tag{6.5}$$

where G is the singular part of the Gibbs potential, $\tau \equiv T - T_c$, and H is the magnetic field. In particular, we note that (6.5) implies that

$$T_c(R) - T_c(0) \sim CR^{1/\varphi} \quad , \tag{6.6}$$

where

$$\varphi \equiv a_R/a_\tau \tag{6.7}$$

is the "crossover exponent." On differentiating (6.5) twice with respect to H and n times with respect to R, we have

$$\bar{\chi}_n(T, H=0, R=0) \equiv \left(\frac{\partial^n \bar{\chi}}{\partial R^n}\right)_{H,\tau} = |\ \tau\ |^{-\gamma_n} \tag{6.8}$$

with

$$\gamma_n = \gamma_0 + n\varphi \quad . \tag{6.9}$$

Two questions arise at this point: (i) Is the hypothesis (6.5) of scaling with respect to R valid? (ii) If it is valid, what is the value of $\varphi = a_R/a_\tau$? In view of our rigorous relation

$$\bar{\chi}_1(0) \propto |\bar{\chi}_d|^2 \quad , \tag{6.10}$$

we have

$$\gamma_1 = 2\gamma_0 \quad . \tag{6.11a}$$

Combining the rigorous relation (6.11) with the scaling prediction (6.9), it follows that if scaling with R is valid, then

$$\varphi = \gamma_0 \quad . \tag{6.12}$$

Thus (6.12) provides a "rigorous" answer to question (ii).[25]

We have not succeeded in answering question (i) in any rigorous or even semirigorous fashion. However, we can verify rigorously certain of the predictions of the scaling hypothesis (6.5). Specifically, for the Ising model lines 7 and 10 of Table I imply that

$$\gamma_2 = 3\gamma_0 \tag{6.11b}$$

and

$$\gamma_3 = 4\gamma_0 \ , \tag{6.11c}$$

respectively, in accordance with the scaling prediction (6.9). Equations (6.11b) and (6.11c) are in conflict with Ising model ($\gamma_0 = 1.75$) estimates of the γ_n from Ref. 11, where it is reported that for the sq → sc crossover, $\gamma_2 = 5.0 \pm 0.1$ and $\gamma_3 = 6.5 \pm 0.2$. However, they are consistent with the much more extensive work of Ref. 17 and with the remarks of Ref. 26 and the extensive calculations on both the sc → sq and the fcc → sq crossovers presented in Ref. 17. Thus one application of our rigorous relations (6.11) is to show that the extrapolations of Refs. 11 are not valid.

2. *Two-Spin Correlation Function*

One can make a scaling hypothesis, analogous to (6.5), for the two-spin correlation function, namely, that there exist four numbers b_τ, b_H, b_r, and b_R such that for all positive λ, $C_2(\tau, H, r, R)$ is a generalized homogeneous function[9]

$$C_2(\lambda^{b_\tau}\tau,\ \lambda^{b_H}H,\ \lambda^{b_r}\vec{r},\ \lambda^{b_R}R) = \lambda\, C_2(T, H, \vec{r}, R) \ . \tag{6.13}$$

From (6.13) it follows that

$$\mu_{2n}(\tau, H=0, R=0) \sim |\ \tau\ |^{-(\gamma_0+2\nu_n)} \tag{6.14}$$

with

$$2\nu_n = 2\nu_0 + n\gamma_0 \ . \tag{6.15}$$

From lines 4, 8, and 10 of Table I the validity of the scaling prediction (6.15) follows at once for the cases $n = 1$, 2, and 3, respectively. Thus these results suggest that not only thermodynamic functions, but also the two-spin correlation fuctions, scale with the anisotropy parameter R. Indeed, the predictions of both (6.5) and (6.13) are borne out for the sq → sc and the sq → fcc crossovers for μ_2 and C_H (the specific heat) by the work of Ref. 17.

It is interesting to point out that although our results for the zero-field high-temperature susceptibility $\bar{\chi}(T, H=0, R)$ and the second moment $\mu_2(T, H=0, R)$ seem to support the hypothesis that both T and R scale, this result does *not* imply that all thermodynamic functions scale in R. The following counterexample illustrates this point. As proved above, if $\bar{\chi}(T, H=0,\ R=0) \sim \tau^{-\gamma_0}$, then the second-derivative function varies as $\bar{\chi}_2(T, H=0,\ R=0) \sim \tau^{-3\gamma_0}$; this result is consistent with the scaling prediction coupled with the result $\varphi = \gamma_0$. Thus if the same scaling hypothesis holds for the Gibbs function and the specific heat, one would expect that $G_2(T, H=0,\ R=0)$ diverges with exponent $\alpha_0 - 2 + 2\gamma_0$ and that $C_{H,2}(T, H=0,\ R=0)$ diverges with exponent $\alpha_0 + 2\gamma_0$. However, as one can easily see from Eq. (3.3) at $H=0$ and $T > T_c(0)$, the first term on the right-hand side vanishes. If we assume that the correlation function $C_2(T, H, \vec{r})$ for the d-dimensional system scales, then one can show using arguments of Sec. IV of Ref. 9 that the second term on the right-hand side of Eq. (3.3) diverges at $T(0)$ with exponent $-d\nu + 2\gamma_0 = \alpha_0 - 2 + 2\gamma_0 - [d\nu - (2 - \alpha_0)]$. Hence if $d\nu > 2 - \alpha_0$ (as many workers believe to be the case for $d = 3$), then our results would indicate that the scaling hypothesis in R [which works for $\chi(T, H\,0, R)$ and $\mu_2(T, H=0, R)$] may not work for $G(T, H=0, R)$ and $C_H(T, H=0, R)$.

C. Precise Meaning and Approximation Determination of Crossover Temperature

The anisotropic system described by (1.1) is interesting because critical-point exponents, according to the universality hypothesis, should depend only upon lattice dimensionality; and hence when $R \to 0$ (and the lattice "crosses over" from d dimensions), we expect anomalous behavior. This crossover behavior would be observable if we could vary R continuously to zero.

Another interesting property of the weakly coupled layers is that even for $R \neq 0$ the system is essentially two dimensional at high temperature. Yet when it is sufficiently close to the critical temperature $T_c(R)$, it is three dimensional. Hence there is a crossover region $T_A(R) \geq T \geq T_B(R)$ where the system transits from $d = 2$ to $\bar{d} = 3$ (cf. Fig. 6).

The crossover region is only a loosely defined concept. To be quantitatively precise, we shall consider the reduced susceptibility $\bar{\chi}(R)$ for the Hamiltonian (1.1). If we consider the coupling in the weak direction as a perturbation, then we may expand $\chi(R)$ in a Taylor series about $R = 0$:

$$\bar{\chi}(R) \equiv \chi_0(0) + \chi_1(0)R + \bar{\chi}_2(0)\frac{R^2}{2!} + \cdots + \bar{\chi}_n(0)\frac{R^n}{n!} + \cdots \ , \tag{6.16}$$

where

$$\bar{\chi}_n(0) \equiv \frac{\partial^n \bar{\chi}(R)}{\partial R^n}\bigg|_{R=0} \ , \tag{6.17}$$

and $\bar{\chi}_0(0) = \bar{\chi}_d$ is the susceptibility of the two-dimensional system. From Table I, it is clear that for systems described by (1.1), we have

$$\bar{\chi}(R) = \bar{\chi}_d[1 + 2R\mathcal{J}\bar{\chi}_d + (2R\mathcal{J}\bar{\chi}_d)^2 f_2(\mathcal{J}) + (2R\mathcal{J}\bar{\chi}_d)^3 f_3(\mathcal{J}) + O(R^4)\cdots] \ , \tag{6.18}$$

where $2 \geq f_2(\mathcal{J}) \geq 1$ and $6 \geq f_3(\mathcal{J}) \geq 1$.

The effective crossover temperature is now determined by the following argument. If $\bar{\chi}(R)$ versus T were known exactly, for a given R, there would be no sudden transition from two-to three-dimensional behavior; rather, as the temperature approached $T_c(R)$, higher-order terms in Eq. (6.18) would gradually become more significant. Thus we can define a p-percent crossover temperature, $T_{0.01p}(R)$, as that temperature at which the second term in Eq. (6.18) causes a p-percent deviation of $\chi(R)$ from the two-dimensional value $\bar{\chi}_d$:

$$2R\,\beta J\,\bar{\chi}_d = 0.01p \tag{6.19}$$

where the left-hand side is expressed as a function of T, and the equation is then solved for $T_{0.01p}$.

Experimentally, this means that if one's confidence limits are p percent, the two-dimensional susceptibility $\bar{\chi}_d$ should be a good enough approximation to $\chi(R)$ down to $T_{0.01p}$. Note that $T_{0.01p}$ is the temperature at which the first-order correction term on the right-hand side of Eq. (6.18) equals to $0.01p$. This temperature may be slightly different from the true p-percent temperature for which $(\chi(R)/\bar{\chi}_d) - 1 = 0.01p$. However, from (6.18), one may reasonably expect that these two temperatures are essentially identical.

Equation (6.19) is the major result of this subsection. It is useful in the following respects:

(i) It may be used to estimate the crossover temperature, since it is generally easier to obtain theoretical information about $\bar{\chi}_d$ than about $\bar{\chi}(R)$.

(ii) By inverting the above procedure, Eq. (6.18) may be used to estimate R when R is not known, the crossover temperature $T_{0.01p}(R)$ being obtained from a plot of $\chi(R)$ vs T. As an illustration, consider the antiferromagnet $(CD_3)_4NMnCl_3$ (TMMC),[6] known to be a spin-$\frac{5}{2}$ quasi-one-dimensional system for $T \geq 1.1$ K, with $J = -6.3$ K. Assuming the experimental error to be 5%, Eq. (6) requires that

$$4R\,\beta J\,\bar{\chi}_{1.c.}(J, T, S) \leq 0.05 \tag{6.20}$$

for $T \geq 1.1$ K, where $\bar{\chi}_{1.c.}$ is the linear chain susceptibility, known exactly, and evaluated at $J = -6.3$ K and $S = \frac{5}{2}$. From this relation we obtain an upper bound on R, $R \leq 10^{-6}$.[27]

(iii) Equation (6.18) may be used in making quantitative estimates for the experimentalist. For example, as we have discussed earlier, current theory predicts that for any given value of R, the critical behavior should be d dimensional at high temperature and three-dimensional near $T_c(R)$. A schematic illustration of the expected behavior is shown in Fig. 6(c) for hypothetical susceptibility data on a "quasi-two-dimensional Ising system." Data similar to these hypothetical experiments have been reported,[28] and subsequently challenged.[29] However, from the theoretical point of view, it would be interesting if we could estimate under what conditions it would be feasible to observe this kind of dramatic crossover behavior. To achieve this aim, we proceed in two steps

(a) To be able to observe the two-dimensional exponent, the system must be well above the critical temperature $T_c(R)$. Let us assume for the sake of argument that the two-dimensional behavior does not set in until $T - T_c(0)/T_c(0)$ is less than about 10^{-2}—i.e., above this temperature, there is no meaningful exponent behavior. Then in order to reliably estimate the two-dimensional exponent, let us assume that we need at least a decade in temperature (i.e., from 10^{-2} to 10^{-3}) before the system deviates from the d-dimensional behavior by, say, 10%. Thus we require that

$$\frac{T_{0.10} - T_c(0)}{T_c(0)} \leq 10^{-3} \tag{6.21}$$

where $T_{0.01p}$ represents the temperature at which the deviation from two-dimensional behavior is p percent; e.g., $T_{0.01p} = T_{0.10}$ if the deviation is 10%. Since $T_{0.01p}$ is given by Eq. (6.19) and since for the $d = 2$ Ising system,

$$\bar{\chi}_{sq}(T) \cong \mathcal{C}_+[(T - T_c)/T_c]^{-7/4} \ , \tag{6.22}$$

we have

$$0.1 \cong 2R\,\beta J\,\bar{\chi}_{sq}(T_{0.1})$$

$$\geq 2R\left(\frac{J}{kT_c(0)}\right)\mathcal{C}_+(10^{-3})^{-7/4} \sim 10^5 R \tag{6.23}$$

or

$$10^{-6} \geq R \ . \tag{6.24}$$

(b) To be able to observe three-dimensional exponents with any confidence, one *probably* will have to reduce ϵ by two decades or more (one decade for the transition to three-dimensional behavior, and one decade in order to accurately measure the slope of the linear three-dimensional behavior). Hence the experimental apparatus should at least be able to achieve the minimum reduced temperature

$$\epsilon_{\min} = \frac{T_{\min} - T_c(R)}{T_c(R)} \cong \frac{T_{\min} - T_c(0)}{T_c(0)} \cong 10^{-5} \ .$$

Combining (a) and (b), we conclude that the most optimistic situation to observe the dramatic behavior of Fig. 6(c) would be to use a system with R on the order of 10^{-6} and to use apparatus capable of making measurements down as low as $\epsilon_{\min} = 10^{-5}$. If R were much *bigger*, the two-dimensional exponent might not be measurable, while if R were much *smaller*, the three-dimensional region may be too small to observe.

We conclude by remarking that the crossover temperature (more precisely, T_p) should be depen-

dent upon the quantity being measured; i.e., for a different physical quantity, $T_{0.01p}$ may well differ. One example of this is the specific heat. At high temperature [$T > T_c(R)$, $H = 0$] it is clear from line 3 of Table I that the first-order correction term $(\partial/\partial R)C_H(R)$ is zero, and the second-order derivative may be diverging less strongly than $\bar{\chi}_2(0)$ (cf. the discussion in Sec. VIB). *Hence, the crossover into the three-dimensional region for the specific heat may occur for a much smaller value of* ϵ.

Note added in proof. Very recently L. J. de Jongh (private communication) has confirmed the utility of the relation (6.19) for the quasi-two-dimensional ferromagnetic compound $(C_nH_{2n+1}NH_3)_2CuCl_4$ with $n = 2$. This work is described in L. J. de Jongh and A. R. Miedema, Adv. Phys. (to be published). This work also reports "crossover behavior" described by considerations such as those that governed the construction of Fig. 6(c).

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APPENDIX: UPPER AND LOWER BOUNDS FOR GENERAL nTH DERIVATIVES FOR SELF-AVOIDING WALK PROBLEMS

As mentioned in Sec. V D, although we have not succeeded in finding bounds for $n > 3$, we show in this appendix that there exist both upper and lower bounds for arbitrary n for a closely related system, the SAW problem.[30]

For simplicity, we shall consider a SAW problem on an anisotropic square lattice; the susceptibility is defined by

$$\bar{\chi}^{\mathrm{SAW}}(v) = \sum_{l=0}^{\infty}\sum_{m=0}^{\infty} C_{lm}^{(d)} u^l v^m \quad , \tag{A1}$$

where the expansion variables are $u \equiv \tanh J/kT$ and $v \equiv \tanh RJ/kT$, and $C_{lm}^{(\bar{d})}$ is the number of self-avoiding walks with l horizontal bonds ("u bonds") and m vertical bonds ("v bonds").

The nth derivative with respect to $v \equiv \tanh RJ$ is

$$\bar{\chi}^{\mathrm{SAW}}(0) = \frac{1}{n!}\left(\frac{\partial^n \bar{\chi}^{\mathrm{SAW}}(v)}{\partial v^n}\right)_{R=0}$$

$$= \sum_{l=0}^{\infty} C_{ln}^{(\bar{d})} u^l \quad . \tag{A2}$$

Now some of the SAW paths contributing to $C_{mn}^{(\bar{d})}$ (with n vertical bonds) go from one "layer" to another without ever returning to any layer that has been passed; an example is shown in Fig. 7(a). Others of the SAW paths will return to a given layer or layers, as shown in Fig. 7(b). If we ignore all paths of the second type, we can obtain a *lower bound* on $C_{mn}^{(\bar{d})}$ and hence a lower bound on $\bar{\chi}_m^{\mathrm{SAW}}(R = 0)$. However, the paths of type one are simply the set of paths which, starting from the origin, take a self-avoiding walk of m_0 steps in the layer $z = 0$, then takes a step to the second layer $z = \pm 1$, and takes another self-avoiding walk of m_1 steps, then another step to the next layer, and so on. Note that the number of steps in each layer m_i, might be zero, and that the first step out of the layer $z = 0$ can be to either of *two* layers. Thus we have the simple inequality, for $n \geq 1$,

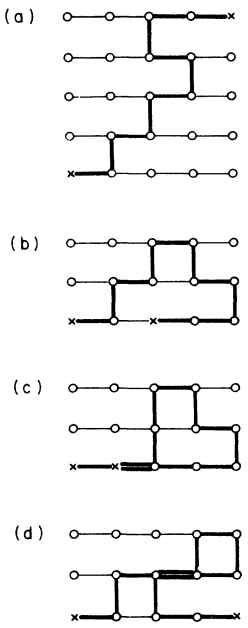

FIG. 7. (a) and (b) are contributions to the SAW problem where (b) does *not* contribute to the lower bound. (c) and (d) are contributions to the upper bound of Eq. (A6) that represents overcounting of configurations (neither walk is self-avoiding). See the discussion in the Appendix.

$$\bar{\chi}_n^{\mathrm{SAW}}(R = 0) \geq 2 \sum_{m_0=0}^{\infty} \cdots \sum_{m_n=0}^{\infty} \left(\prod_{i=0}^{n} C_{m_i}^{(d)} u^{m_i}\right) . \tag{A3}$$

where $C_{m_i}^{(d)}$ is the number of self-avoiding walks of m_i in the d-dimensional lattice forming the "layers" (here $d = 1$). Since the number of SAW steps taken on the ith layer, m_i, is independent of the number of SAW steps on the jth layer ($i \neq j$), it is straightforward to evaluate the expression of (A3), with the result

$$\bar{\chi}_n^{\mathrm{SAW}}(0) \geq 2[\bar{\chi}_d^{\mathrm{SAW}}]^{n+1} \quad . \tag{A4}$$

To obtain an upper bound for $\chi_n^{\mathrm{SAW}}(R=0)$, we shall consider the following type of path. Starting from the origin, we first take a SAW of m_0 steps in the $z_0=0$ layer, then take a single step (through a v bond) to either the layer above ($z_1=z_0+1=+1$) or the layer below ($z_1=z_0-1=-1$). In the layer z_i we take another SAW of m_1 steps, and then we proceed to another layer—*either the layer above or the layer below*. We take another self-avoiding walk in the new layer, but we assume we lost all memory of previous self-avoiding walks in that layer. We thereby overcount the number of self-avoiding walks, in the sense that although we include all SAW's contributing to (A1), we also include numerous paths that are not allowed. Thus we obtain a rigorous upper bound to χ_n^{SAW},

$$2^n \sum_{m_0=0}^{\infty} \cdots \sum_{m_n=0}^{\infty} \left(\prod_{i=0}^{n} C_{m_i}^{(d)} u^{m_i} \right) \geq \overline{\chi}_n^{\mathrm{SAW}}(R=0) \quad , \tag{A5}$$

where the factor 2^n arises from the fact that we may have two choices for *each* step from layer z_i to the next layer. Evaluating the expression on the left-hand side of (A5), we finally obtain the desired result:

$$2^n [\overline{\chi}^{\mathrm{SAW}}]^{n+1} \geq \overline{\chi}_n^{\mathrm{SAW}}(R=0) \quad . \tag{A6}$$

One can compare the results for $\overline{\chi}_n^{\mathrm{SAW}}(0)$ in (A4) and (A6) with the results tabulated in Table I for $\overline{\chi}_n(0)$, and the forms of the upper and lower bounds are similar to each other. In particular (A4) and (A6) yield the exponent equality

$$\gamma_n^{\mathrm{SAW}} = \gamma_0^{\mathrm{SAW}} + n\gamma_0^{\mathrm{SAW}} \quad , \tag{A7}$$

consistent with the scaling predictions.

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[1](a) For a recent review, see L. J. de Jongh, P. Bloembergen, and J. H. P. Colpa, Physica (Utr.) **58**, 305 (1972); (b) see, also, the original neutron-scattering experiments of R. J. Birgeneau, G. Shirane, and H. J. Guggenheim, Phys. Rev. Lett. **22**, 720 (1969).

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[10]If we fix the temperature and allow R to decrease, the correlation between spins of different planes diminishes, and the system becomes more and more two dimensional. If, on the other hand, we fix R (as is the case with most experiments) at high temperatures, the correlation between spins of different planes is destroyed by the fluctuations and the system appears two dimensional. However, as the temperature is lowered toward $T_c = T_c(R)$, interplanar correlations will become significant and the system will exhibit three-dimensional behavior. Thus, as the critical temperature is approached from above, the critical exponents will cross over from two-dimensional values to those characteristic of three-dimensional systems. See Fig. 6 and Sec. VI C.

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[15]*Theorem G1* has in fact been proved for several other model systems. For an extensive review, see, e.g., R. B. Griffiths, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1972), Vol. 1.

[16]Although Theorem 2 of Reg. 14 was proved only for the $S=\frac{1}{2}$ Ising model, our Theorem *G* 3*b* may easily be shown to be valid for general spin quantum number S. We wish to thank Professor R. B. Griffiths for pointing out this result.

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[27]In this example, the validity of our formula was assumed and it was used to obtain an upper bound on R for a system for which R is *not* known; to test our formula more thoroughly, we need a system for which all the parameters, including R, are known. Very recently J. de Jongh (private communication and unpublished) has supplied data on just such a system!

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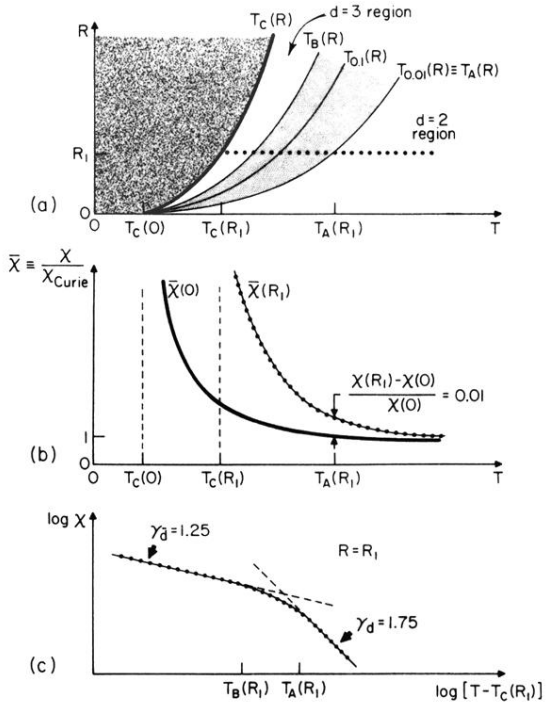

FIG. 6. Schematic diagram of the crossover behavior. (a) The crossover region (shaded area) is bounded by $T_A(R)$ and $T_B(R)$. [$T_A(R) = T_{0.01}(R)$ is the temperature at which the system differs appreciably (1%) from being two dimensional.] $T_c(R)$ is the critical temperature. The generalized scaling hypothesis predicts that all curves should approach $T_c(0)$ via the power law $R^{1/\varphi}$. (b) Dependence of reduced susceptibility $\bar{\chi}$ upon T for $R = 0$ and for $R = R_1$, indicating the definition of $T_A(R)$. Note that this drawing is not to scale. (c) Sketch of hypothetical experimental data, plotted in the conventional log-log plot, for a system which is described by the Hamiltonian with $R = R_1$.