times in a superconductor such as electron-electron, electron-phonon, and branch mixing¹⁰ must be considerably shorter. In addition, the behavior of the energy gap of an illuminated superconductor can be used to measure the quasiparticle recombination time in superconductors.

We are pleased to acknowledge valuable discussions with Dr. C. Owen and Professor D. J. Scalapino.

*Research supported by the National Science Foundation.

¹L. R. Testardi, Phys. Rev. B 4, 2189 (1971).

²C. S. Owen and D. J. Scalapino, Phys. Rev. Lett. <u>28</u>, 1559 (1972).

³The effective recombination time τ is not the quasi-

particle recombination time τ_R since the phonons produced by recombination of the quasiparticles will themselves create quasiparticles. See A. Rothwarf and B. N. Taylor, Phys. Rev. Lett. <u>19</u>, 27 (1967).

⁴S. Y. Hsieh and J. L. Levine, Phys. Rev. Lett. <u>20</u>, 1502 (1968).

⁵A. Rothwarf and M. Cohen, Phys. Rev. <u>130</u>, 1401 (1963).

⁶Rothwarf and Taylor, Ref. 3.

⁷C. Kittel, Introduction to Solid State Physics (Wiley, New York, 1971), 4th ed., p. 249.

⁸B. N. Taylor, Ph.D. thesis, University of Pennsylvania, 1963 (unpublished).

⁹We would like to thank C. Owen for sending us the results of computer calculations of μ * as a function of n and reduced temperature.

¹⁰J. Clarke, Phys. Rev. Lett. <u>28</u>, 1363 (1972); M. Tinkham and J. Clarke, Phys. Rev. Lett. <u>28</u>, 1366 (1972).

Some Results Concerning the Crossover Behavior of Quasi–Two-Dimensional and Quasi–One-Dimensional Systems*

Luke L. Liu and H. Eugene Stanley

Physics Department, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 27 April 1972)

A magnetic system with intraplanar and interplanar interaction strengths J and RJ is is treated. Rigorous relations are established concerning the first few derivatives with respect to R of the susceptibility $\chi(R)$. Considering $\chi(R) = b_0 + b_1 R + b_2 R^2 + \cdots$, we find b_1 and the order of magnitude of b_2 . Hence we can predict when the system "crosses over" from d-dimensional to \overline{d} -dimensional behavior (e.g., for quasi-two-dimensional systems, d=2, $\overline{d}=3$, while for quasi-one-dimensional systems, d=1, $\overline{d}=3$). These results also support scaling with respect to the anisotropy parameter R.

There has recently been considerable interest¹⁻³ in systems with "lattice anisotropy" (different coupling strengths in different lattice directions). Consider, e.g., the *d*-dimensional nearest-neighbor (nn) Ising system with Hamiltonian

$$\mathcal{K} = -J \sum_{\vec{v}_i = \vec{v}_j}^{nn} s_i s_j - RJ \sum_{\vec{u}_i = \vec{u}_j}^{nn} s_i s_j \equiv \mathcal{H}_0 + R\mathcal{H}_1, \qquad (1)$$

where $\vec{r}_i \equiv (x_1, x_2, \cdots, x_{\overline{d}}) \equiv (\vec{u}_i, \vec{v}_i)$ with $u_i = (x_1, \cdots, x_d)$, and $\vec{v}_i \equiv (x_{d+1}, \cdots, x_{\overline{d}})$. For example, very recently there have been extensive calculations¹ concerning the case d = 2, $\vec{d} = 3$, corresponding to a "square to simple-cubic crossover." Hence-forth we shall consider this system for the purpose of specificity and clarity; thus $\vec{r}_i \equiv (x_i, y_i, z_i) \equiv (\vec{u}_i, z_i)$, $J = J_{xy}$, $RJ = J_z$. In the last paragraph we treat briefly the case d = 1, $\vec{d} = 3$.

The system described by (1) is interesting because critical-point exponents, according to the universality hypothesis,² should depend only upon lattice dimensionality; and hence when $R \rightarrow 0$ (and the lattice "crosses over" from \overline{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 $T_A(R) \ge T$ $\ge T_B(R)$ where the system transits from d=2 to $\overline{d}=3$ (cf. Fig. 1).

The crossover region is only a loosely defined concept. To be quantitatively precise, we shall define $T_p(R)$ as the solution of $\chi(T_p, R)/\chi(T_p, 0)$ -1=p and we arbitrarily choose $T_A(R) = T_{0.01}(R)$, since this is roughly the temperature for which the deviation (1%) of the reduced susceptibility $\overline{\chi}(R) \equiv \chi/\chi_{\text{Curie}}$ from the two-dimensional value $\overline{\chi}(0)$ becomes experimentally appreciable.⁴

This crossover behavior is most easily explained in the context of the scaling hypothesis,² where we assume that there exist three numbers

(3)

FIG. 1. 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.] $T_{0,1}(R)$ is the temperature at which Eq. (4) is no longer satisfactory. $T_c(R)$ 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 $\overline{\chi}$ 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 (1) with $R = R_1$.

$$a_{\tau}$$
, a_{H} , and a_{R} , such that for all positive λ

$$G(\lambda^{a_{\tau}}\tau,\lambda^{a_{H}}H,\lambda^{a_{R}}R) = \lambda G(\tau,H,R), \qquad (2)$$

where G is the singular part of the Gibbs potential, $\tau = T - T_c$, and H is the magnetic field. From the scaling hypothesis, the situation sketched in Fig. 1 follows immediately.

In particular, we note that (2) implies that $T_c(R) - T_c(0) \sim CR^{1/\varphi}$ and $T_p(R) - T_c(0) \sim A_p R^{1/\varphi}$, where $\varphi \equiv a_R/a_\tau$ is the "crossover exponent." On differentiating (2) twice with respect to H and n times with respect to R, we have $\overline{\chi}_n(\tau, H=0, R=0) \sim |\tau|^{-\gamma_n}$, with $\gamma_n = \gamma_0 + n\varphi$, where $\overline{\chi}_n \equiv (\partial^n \overline{\chi}/dR^n)_{H,\tau}$.



The exponents γ_n cannot be calculated exactly, but they *can* be estimated by extrapolations based upon high-temperature series expansions. There presently exists a dispute^{1 b, 1 c} in the literature concerning numerical values of γ_m and the most recent estimates^{1 b} seem to challenge the scaling prediction $\gamma_n = \gamma_0 + n\varphi$.

We shall first derive rigorously the relation

$$\overline{\chi}_1(0) = 2g\overline{\chi}_0^2$$

where $\mathfrak{g} \equiv \beta J$. This relation is useful for the following purposes:

(i) Equation (3) is the first-order correction term of the two-dimensional approximation of the threedimensional quantity $\overline{\chi}(R)$. In other words, we have

$$\overline{\chi}(R) = \overline{\chi}_0 + R\overline{\chi}_1(0) + \frac{1}{2}R^2\overline{\chi}_2(0) + \dots = \overline{\chi}_0 + R(2g\overline{\chi}_0^2) + O(R^2).$$
(4)

To O(R), $T_{b}(R)$ may therefore be found from the solution of $2gR\overline{\chi_{0}}=p$.

(ii) Equation (4) may be used as a checking method for the leading coefficient of the general-R high-temperature series expansion

$$\overline{\chi}(R) = \sum_{n=0}^{\infty} \sum_{j=0}^{n} a_{nj} R^{j} \mathcal{G}^{n}.$$
(5)

(iii) If (2) is valid, Eq. (3), which yields $\gamma_1 = 2\gamma_0$, furnishes a simple but rigorous proof of $\varphi = \gamma_0$, and hence $\gamma_n = (n+1)\gamma_0$.

We shall now derive the following results:

$$8g^{2}\chi_{0}^{3} \ge \overline{\chi}_{2}(0) \ge 4g^{2}\overline{\chi}_{0}^{3},$$

$$(6)$$

$$48g^{3}\chi_{0}^{4} \ge \overline{\chi}_{3}(0) \ge 8g^{3}\overline{\chi}_{0}^{4}.$$

$$(7)$$

Equations (6) and (7) yield $\gamma_2 = 3\gamma_0$, and $\gamma_3 = 4\gamma_0$, which agree exactly with the scaling predictions $\gamma_n = (n+1)\gamma_0$; thus the reported estimates^{1b} are unreliable. From (6) we have a fairly good estimate of

(8)

the second-order correction term; therefore (4) may be improved, with the result

$$\overline{\chi}(R) \cong \overline{\chi}_0 \left[1 + 2Rg \,\overline{\chi}_0 + R^2 4g^2 \overline{\chi}_0^2 f_2(g) + O(R^3) \right],$$

where $2 \ge f_2(g) \ge 1$.

The proof of (3) proceeds from observing that at R=0, spins on different layers (i.e., if $z_k \neq z_i$) are not coupled. Hence for $T > T_c(0)$ and H=0 (i.e., $\langle s_i \rangle_d = 0$), the thermal average of spin product is

$$\langle s_k s_l \rangle_{R=0} = \delta(z_k, z_l) \langle s_k s_l \rangle_d, \tag{9}$$

where $\langle \cdots \rangle_R$ denotes a thermal average of a \overline{d} -dimensional Hamiltonian (1) and $\langle \cdots \rangle_d$ denotes a thermal average of a d-dimensional Hamiltonian. Note that on the right-hand side of Eq. (9) we set R = 0in the Hamiltonian *before* taking a thermal average, while on the left-hand side we set R = 0 after calculating the thermal average. Furthermore we have, for $z_b \neq z_1$,

$$\langle s_i s_j s_k s_l \rangle_{R=0} = \delta(z_i, z_k) \,\delta(z_j, z_l) \langle s_i s_k \rangle_d \,\langle s_j s_l \rangle_d + \delta(z_i, z_l) \,\delta(z_j, z_k) \langle s_i s_l \rangle_d \,\langle s_j s_k \rangle_d. \tag{10}$$

The reduced susceptibility of a lattice which is composed of N+1 layers with M^2 spins in each layer⁵ is given by

$$\overline{\chi}(T, H=0; R) = [(N+1)M^2]^{-1} \sum_{\overline{r}_i, \overline{r}_j} [\langle s_i s_j \rangle - \langle s_i \rangle \langle s_j \rangle].$$
(11)

Differentiating (11) and using (9) and (10), we have

$$(N+1)M^{2}\overline{\chi}_{1}(0) = - \beta \sum_{\vec{t}_{i},\vec{t}_{j}} \langle s_{i}s_{j}\mathcal{H}_{1} \rangle_{R=0} = \mathcal{G} \sum_{\vec{t}_{k}} \sum_{z_{k}=1}^{N} \{ \sum_{\vec{u}_{i}} \langle s_{\vec{u}_{i}z_{k}} s_{\vec{u}_{k}z_{k}} \rangle_{d} \sum_{\vec{t}_{j}} \langle s_{\vec{u}_{j}z_{k}+1} s_{\vec{u}_{k}z_{k}+1} \rangle_{d} + \sum_{\vec{t}_{i}} \langle s_{\vec{u}_{i}z_{k}+1} s_{\vec{t}_{k}z_{k}+1} \rangle_{d} \sum_{\vec{t}_{j}} \langle s_{\vec{t}_{j}z_{k}} s_{\vec{t}_{k}z_{k}} \rangle_{d} \}.$$
(12)

Now each of the four summations inside the curly brackets of (12) is over all spins which lie in a single plane, and thus each summation is exactly the reduced susceptibility of a *two*-dimensional lattice. Thus in the thermodynamic limit, we have Eq. (3).

Four remarks are worth making at this point:

(i) Equation (3) may be derived directly using graphical methods that are valid for arbitrary spin dimensionality D, and hence its validity is *not* restricted to the Ising model. In particular, for the spherical model $(D = \infty)$, (3) can be derived directly from the general-R partition function as well.

(ii) The argument used in deriving (3) may be extended to any pair of lattices that can be related by a "lattice anisotropy" term in the Hamiltonian, with the result

$$\overline{\chi}_1^{\ d}(0) = g^{\ dd} \, g_{\overline{\chi}_0^{\ 2}}^2, \tag{13}$$

where $g^{d\bar{d}}$ is the number of "additional nearest neighbors" that a site acquires when the lattice anisotropy term transforms the lattice from being *d*-dimensional to being \bar{d} -dimensional [e.g., g=2 for sq \rightarrow sc, linear chain \rightarrow sq, but g=4 for linear chain (lc) \rightarrow sc and g=8 for sq \rightarrow fcc]. If \mathcal{X}_1 contains nonnearest-neighbor terms, we may simply replace $g^{d\bar{d}}g$ by $\sum_i g_i d\bar{d} g_i$, where g_i^{dd} is the number of neighbors interacting with a site with strength RJ_i .

Equation (13) may be used as a checking procedure for the coefficients of (5) (e.g., at R=0, the a_{n1} are the coefficients in $2\chi_{sq}^2$, while in the limit of $J \rightarrow 0$, RJ = const, the $a_{n,n-1}$ are the coefficients in $4\overline{\chi}_{1c}^2$).

(iii) Note that since our derivation starts from the correlation function, we may obtain the following relations for the second moment μ_2 of the correlation function:

$$(\partial \mu_2 / \partial R)_{R=0} = 2g[\bar{\chi}_0^2 + 2\bar{\chi}_0 \mu_2(0)], \qquad (14)$$

and the general \bar{q} susceptibility (where \bar{q} is a \bar{d} = 3-dimensional vector):

$$\left[\frac{\partial \overline{\chi}(\mathbf{q})}{\partial R} \right]_{R=0} = 2\vartheta \left[\overline{\chi}_0(\mathbf{q}) \right]^2 \cos(\mathbf{q} \cdot \hat{z}),$$
with $\overline{\chi}(\mathbf{q}) \equiv \int e^{i \mathbf{q} \cdot \mathbf{r}^T} C_2(\mathbf{r}) d^3 r.$
(15)

929

(17)

To derive (7) we may proceed in exactly the same fashion and arrive at

$$\mathcal{G}^{-2}\overline{\chi}_{2}(0) = 4[\overline{\chi}_{0}(0)]^{3} + 2\sum_{\vec{u},\vec{v}_{i},\vec{v}_{j}} \langle s_{0}s_{\vec{u}}\rangle_{d} \left(\langle s_{0}s_{\vec{u}}s_{\vec{u}_{i}}s_{\vec{u}_{j}}\rangle_{d} - \langle s_{0}s_{\vec{u}}\rangle_{d} \langle s_{\vec{u}_{i}}s_{\vec{u}_{j}}\rangle_{d} + 2\sum_{\vec{u}} \langle s_{0}s_{\vec{u}}(\sum s_{\vec{u}_{i}})\rangle_{d}^{2}.$$
(16)

The Griffiths inequality⁶

$$\langle s_0 s_{\vec{u}} s_{\vec{u}_i} s_{\vec{u}_j} \rangle - \langle s_0 s_{\vec{u}} \rangle \langle s_{\vec{u}_i} s_{\vec{u}_j} \rangle \ge 0$$

permits us to cancel the second term on the right-hand side of (16). Noting that the third term is positive, we have the second inequality of (6). Using⁷

$$\langle s_0 s_{\vec{u}_i} \rangle \langle s_{\vec{u}} s_{\vec{u}_j} \rangle + \langle s_0 s_{\vec{u}_j} \rangle \langle s_{\vec{u}_i} s_{\vec{u}} \rangle \geq \langle s_0 s_{\vec{u}} s_{\vec{u}_i} s_{\vec{u}_j} \rangle - \langle s_0 s_{\vec{u}} \rangle \langle s_{\vec{u}_i} s_{\vec{u}_j} \rangle$$

$$\tag{18}$$

and the fact that $\langle s_0 s_{\vec{u}} s_{\vec{u}} \rangle \equiv 0$ for the Ising system, we immediately have from (16) the first inequality of (6).

For higher derivatives, the algebra becomes much more involved. However, we have been able to derive (7) for systems for which Ginibre's generalization⁸ of Griffiths's inequality⁶ is valid. Furthermore, for the self-avoiding walk on an anisotropic lattice, by means of simple graphic theoretical arguments we have proved $\gamma_n = (n+1)\gamma_0$ for all n.

We conclude with several remarks. (i) The exponent φ obtained from the γ_n 's does not necessarily characterize the asymptotic behavior of $T_{c}(R) - T_{c}(0)$ except within the framework of scaling-type arguments. (ii) The previous derivation³ of $\varphi = \gamma_0$ exploits the relation $d\nu = 2 - \alpha$, for which numerical estimates indicate there is a 2% discrepancy for d=3. Our derivation of (3) is rigorously true, *independent* of the relation $d\nu = 2 - \alpha$. (iii) It is clear that for different thermodynamic quantities, the crossover region may be different. In particular, since the firstorder correction term $(\partial C_{H}/\partial R)$ in the analog of Eq. (4) for the specific heat is zero, it would be more difficult for one to use specific-heat measurements to cross over into the d=3 region. (iv) One may also use (4) to determine the anisotropy parameter R. For example, experiments⁹ show that $(CD_3)_4 NMnCl_3$ is essentially an $S = \frac{5}{2}$ antiferromagnetic Heisenberg linear chain for $T \ge 1.1$ °K with intrachain coupling J = -6.3°K. Assuming the experimental error bar is 5%, we may substitute $T = 1.1^{\circ}$ K, S, and J into the equation $4R\beta J\chi_{1c} \leq 0.05$; we get $R \leq 10^{-6}$, in contrast to the value $R \sim 10^{-5}$ obtained by a Green's function analysis.⁹

We wish to thank Professor R. B. Griffiths for several extremely useful remarks.

*Work supported by the National Science Foundation, the Office of Naval Research, and the Air Force Office of Scientific Research.

^{1a}G. Paul and H. E. Stanley, Phys. Lett. 37A, 347 (1971), and Phys. Rev. B <u>5</u>, 2578 (1972). ^{1b}J. Oitmaa and I. G. Enting, Phys. Lett. <u>36A</u>, 91

(1971); I. G. Enting and J. Oitmaa, Phys. Lett. 38A,

107 (1971); J. Oitmaa and I. G. Enting, J. Phys. C:

Proc. Phys. Soc., London 5, 231 (1972).

^{1c}D. C. Rapaport, Phys. Lett. 37A, 407 (1971). ²See, e.g., A. Hankey and H. E. Stanley, Phys. Rev.

B (to be published), and references contained therein. ³R. Abe, Progr. Theor. Phys. 44, 339 (1970); M. Su-

zuki, Progr. Theor. Phys. 46, 1054 (1971).

⁴Clearly, p depends on the precision of the experimental situation. A more accurate experiment corresponds to a lower value of p.

⁵We shall assume the validity of our results in the thermodynamic limit.

⁶R. B. Griffiths, J. Math. Phys. 10, 1559 (1969).

⁷R. B. Griffiths, C. A. Hurst, and S. Sherman, J. Math. Phys. 11, 790 (1970). Although their theorem 2 has only been proven for the $S = \frac{1}{2}$ Ising system, Equation (18) may easily be generalized to arbitrary spin (R. B. Griffiths, private communication).

⁸J. Ginibre, Phys. Rev. Lett. 23, 828 (1969).

⁹R. J. Birgeneau, R. Dingle, M. T. Hutchings, G. Shirane, and S. L. Holt, Phys. Rev. Lett. 26, 718 (1971); R. Dingle, M. E. Lines, and S. L. Holt, Phys. Rev. 187, 643 (1969).