PHYSICAL REVIEW B VOLUME 13, NUMBER 7 1 APRIL 1976

Phase transitions in layered isotropic systems

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(Received 7 July 1975)

A theory of phase transitions for systems of weakly coupled layers with an isotropic order parameter is developed. The properties of the two-dimensional (2D) system are assumed to be known, and the interlayer coupling is treated by a mean-field or random-phase type approximation leading to an appropriate Landau-Ginzburg free-energy functional for the 3D system. The assumption that the pure 2D system has a phase transition and that it satisfies scaling is shown to lead to a number of measurable consequences. The 3D ordering temperature, the 2D-3D crossover region, the 3D critical region, and mean-field specific-heat jump as well as other mean-field properties are obtained as functions of the interlayer coupling. Comparison with existing experimental results for almost isotropic layered ferromagnets supports the existence of a finite transition temperature in 2D. However, the non-symmetry-broken low-temperature phase is found within our approximation to be unattainable owing to the finite interlayer fields.

I. INTRODUCTION

The effect of fluctuations on a phase transition in a two-dimensional (2D) system where the order parameter has a continuous symmetry group (and is therefore isotropic in an appropriately chosen space) is a challenging theoretical question. It has been proven[1] that spontaneous symmetry breaking (i.e., a nonzero average of the order parameter for zero external field) is impossible at nonzero temperatures. On the other hand, there exist theoretical indications[2-7] for the existence of an unusual kind of phase transition where, although the order parameter vanishes, its susceptibility diverges in the low-temperature phase. Experimentally, one may only consider approximations to 2D systems consisting of, for example, layered systems where the ordering interactions among the layers are much weaker than those within the layers. Examples of such systems are intercalated superconductors, anisotropic lattices, and layered magnets where a beautiful systematic study of the phase transitions as the interlayer coupling is made weaker was recently done[8-10] in the system $(C_nH_{2n+1}NH_3)_2CuX_4$, where $X = Cl$ (Br) and $n = 1, 2, 3, 4, 5, 6, 10$. Even an extremely weak interlayer coupling causes a 3D ordering below a well-defined transition temperature; thus it is important to understand the effect of the interlayer coupling in order to reach conclusions on the behavior of the pure 2D systems.

In this paper we shall use an approximation which assumes that the properties of a pure 2D system are known and treats the interlayer coupling in a mean-field way.[11] By comparing the ensuing measurable results with experiment, one can get valuable information on the 2D properties.

We shall present theoretical results on the thermodynamic properties, correlation functions, and susceptibilities, and obtain some indications concerning the properties of the 2D systems. We shall show that our approximation is excellent in the sense that the appropriate critical regions, which indicate the breakdown of our modified mean-field theory, become small for weak interlayer couplings. The results of our approximation are consistent with the exact crossover results of Liu and Stanley,[12] but we obtain further approximate predictions. We shall also derive an effective Landau-Ginzburg-Wilson free-energy functional that can be used in the 3D critical regime. An analogous treatment for weakly coupled chains was recently given in Ref. 11.

Comparison with existing experimental results appears to favor the existence of the 2D transition. However, a perhaps disappointing but in fact quite an obvious conclusion of our approximation is that the low-temperature non-symmetry-broken phase of the pure 2D system, which is of great theoretical interest, is physically unrealizable for a finite, however weak, interlayer coupling. This conclusion depends, however, on our mean-field approximation for the interlayer coupling, and it may or may not follow from a more accurate picture.

In Sec. II we describe the model and derive its thermodynamics and susceptibility. In Sec. III we derive the correlation function and obtain the Ginzburg criterion and the appropriate Ginzburg-Landau functional for the 3D regime. Measurable predictions are summarized in Sec. IV, along with a brief comparison with existing experimental results. We conclude by discussing the fluctuation-induced shift in the 3D ordering transition temperature T_c.

II. THE MODEL AND ITS THERMODYNAMICS

Let us start from a general model of the following form:

$$\mathcal{H}=\sum_i \mathcal{H}_{2D}(i)+\sum_{\langle i,j\rangle} V(i,j), \quad (1)$$

where the variable i characterizes the ith layer, $\mathcal{H}_{2D}$ is the Hamiltonian of a single layer, and the second term, in which the summation is on nearest-neighbor planes, is an interlayer interaction. Possible forms for $\mathcal{H}_{2D}$ and V are

$$\mathcal{H}_{2D}=\int d^2x\,[\,c_\parallel(\nabla\psi_i)^2+a\psi_i^2+b(\psi_i^2)^2-h\psi_i] \quad (2a)$$

or

$$\mathcal{H}_{2D}=\sum_{\vec{R}_i,\vec{\delta}_\parallel} -J_\parallel \vec{S}(\vec{R}_i)\cdot\vec{S}(\vec{R}_i+\vec{\delta}_\parallel)-\vec{h}\cdot\vec{S}(\vec{R}_i), \quad (2b)$$

$$V(i,j)=2c_\perp\int d^2x\,\psi_i(x)\psi_j(x) \quad (3a)$$

or

$$\sum_j V(i,j)=-J_\perp\sum_{\vec{R}_i,\vec{\delta}_\perp} S(\vec{R}_i)\cdot S(\vec{R}_i+\vec{\delta}_\perp), \quad (3b)$$

for the cases of Landau-Ginzburg fields and spin Hamiltonians, respectively. Where $a=\bar{a}(T-T_c^0)/T_c^0$; $\bar{a}$, b, $c_\parallel$, $c_\perp$, $J_\parallel$, and $J_\perp$ are positive constants (the case of antiferromagnetic interactions, on a cubic lattice and in the classical approximation, can be described by modifying the definitions of alternate spins). $\vec{R}_i$ are the lattice vectors in the ith plane, $\vec{\delta}_\parallel$ are nearest-neighbor lattice vectors in the plane, and $\vec{\delta}_\perp$ are nearest-neighbor lattice vectors connecting nearest-neighbor planes [the ith to the jth plane, in Eq. (3)]. $\psi_i(x)$ is the Landau-Ginzburg field [having in general n components; ψ_i^2 is understood to be the length of this n-dimensional vector, $(\nabla\psi_i)^2$ has a sum over the components of ψ_i, and the products of ψ and h are scalar products in the n-dimensional ψ space]. $\vec{S}(\vec{R}_i)$ is the spin operator on the site R_i, and $\vec{h}$ is an external ("magnetic") field.

We shall start with the case of zero external field and will treat the interlayer coupling by a mean-field approximation, assuming that we know the solution for the 2D problem:

$$F_{2D}(T,\vec{h})=-k_BT\ln\mathrm{Tr}\,e^{-\mathcal{H}_{2D}/k_BT}, \quad (4)$$

leading to the 2D order parameter $[\beta=(k_BT)^{-1}]$

$$m_{2D}(T,h)=-\frac{\partial[\beta F_{2D}(T,h)]}{\partial(\beta h)}, \quad (5)$$

where m is $\langle\psi\rangle$ or $\langle s\rangle$ (in the direction of $\vec{h}$). The interlayer mean-field approximation consists of taking in (4) and (5)

$$h=Jm, \quad (6)$$

where

$$J=4c_\perp \quad\text{or}\quad J=4J_\perp \quad (7)$$

for the Landau-Ginzburg and the spin cases, respectively. We find that m is zero above T_c and is nonzero and starts to grow below the 3D phase transition temperature T_c given by

$$1=J\chi_{2D}(T_c,0), \quad (8)$$

where χ_{2D} is the 2D susceptibility defined by

$$\chi_{2D}(T,h)=\frac{\partial m(T,h)}{\partial(h)}. \quad (9)$$

The thermodynamics of the model for $h_{\text{ext}}=0$ is given above T_c by the pure 2D result (4); below T_c we have to take the mean field into account. In order to do that we have to know the linear and nonlinear susceptibilities of the pure 2D systems, which we shall obtain from the expansion of the free energy in powers of βh,

$$\beta F_{2D}(T,h)=f(T)+\tfrac{1}{2}G_1(T)(\beta h)^2+\tfrac{1}{4}G_2(T)(\beta h)^4, \quad (10)$$

where G_1 is proportional to the linear susceptibility $[\beta G_1(T)=-\chi_{2D}(T,0)]$ and G_2 to the next order nonlinear one.

By doing the appropriate thermodynamic manipulations we find that the Landau free energy which corresponds to our interlayer mean-field approximation is, for $h_{\text{ext}}=0$,

$$F_L(T,m)=\tfrac{1}{2}Jm^2+F_{2D}(T,h=Jm)$$
$$=f(T)+a_Lm^2+b_Lm^4, \quad (11)$$

where the Landau coefficients a_L and b_L are given by

$$a_L=\tfrac{1}{2}J(1+\beta JG_1)=\tfrac{1}{2}J(1-J\chi_{2D}),$$
$$b_L=\tfrac{1}{4}G_2\beta^3J^4. \quad (12)$$

Close to T_c, b_L can be approximated by a constant, and a_L can be written as

$$a_L=\bar{a}t \quad\text{with}\quad t=(T-T_c)/T_c. \quad (13)$$

Thus the order parameter builds up below T_c in a mean-field fashion

$$m^2=-(\bar{a}/2b)t. \quad (14)$$

The specific heat has a mean-field-type discontinuity at T_c given by

$$\Delta c=\bar{a}^2/2b_LT_c. \quad (15)$$

We shall find that for weak interlayer couplings the coefficient of t in (14) and Δc of (15) will typically be much larger and much smaller than the respective quantities in the spatially isotropic case (in which the interlayer and intralayer couplings are comparable).

It is now of great interest to find out how specific properties of the 2D system show up in our approximate picture of the layered system. Let us start by making the mild assumption that the 2D system has a phase transition at T_c^{2D} ($T_c^{2D}=0$ is included!) and that the susceptibility diverges as T is decreased towards T_c^{2D}. This assumption probably holds always. Even in the least favorable case[6,7] of the spherical model ($n=\infty$) or the ideal Bose gas at a constant density, it can easily be shown that $T_c^{2D}=0$ and that $\chi \propto e^{T_0/T}$. It is then clear that even for an arbitrarily weak interlayer coupling, Eq. (8) will be satisfied at a finite $T_c > T_c^{2D}$. We start with the case of a nonzero T_c^{2D} and a power-law divergence of χ_{2D},

$$\chi_{2D}=A\beta_c[(T-T_c^{2D})/T_c^{2D}]^{-\gamma_2}. \tag{16}$$

A is a dimensionless constant which may be expected in many cases to be of order unity. We find from (8) that

$$(T_c-T_c^{2D})/T_c^{2D}=(A\beta_c J)^{1/\gamma_2}. \tag{17}$$

For an exponential divergence of χ_{2D}, a logarithmic dependence will be obtained in the right-hand side of Eq. (17).

Two important remarks can be made on Eq. (17):

(a) The physical meaning of the condition (17) is the following: Close to T_c^{2D}, strong correlations are developed within each layer and each spin is strongly correlated with $\sim(\xi_{2D}/\delta_\parallel)^2$ neighboring spins. Here ξ_{2D} is the 2D correlation length and $k_B T\chi_{2D}\sim(\xi_{2D}/\delta_\parallel)^2$ (we ignore the critical exponent η in this rough qualitative argument). The energy required to flip a spin (which in the mean-field theory is of the same order of magnitude as T_c) is *not* just of the order of $2J_\perp$. Owing to the strong 2D correlations, we have to flip on the order of $(\xi_{2D}/\delta_\parallel)^2$ spins together; thus the relevant coupling constant is of the order of $2J_\perp k_B T\chi_{2D}$, which is consistent with (17).

(b) The result (17) for T_c also follows from the exact result of Liu and Stanley[12] for the crossover behavior as a function of the anisotropy parameter R. This is also consistent with "strong scaling" of the thermodynamic functions with R. However, our approximate treatment can be used to obtain further (approximate) specific relations.

Let us first estimate $\bar{a}$ [of Eq. (13)]. From (12) and (8) we find

$$\bar{a}=\frac{\gamma_2 J}{2(J\beta_c A)^{1/\gamma_2}}=\frac{\gamma_2 J}{2\tau_0}, \tag{18}$$

where

$$\tau_0=\frac{T_c-T_c^{2D}}{T_c^{2D}}\quad(\tau_0\ll 1). \tag{19}$$

We can also easily obtain the magnetic susceptibility χ. In our modified mean-field approximation it is given by the random-phase-approximation expression [cf. Eq. (30)]

$$\chi=\frac{\chi_{2D}}{1-J\chi_{2D}}\cong\begin{cases}\chi_{2D} & \text{for } t\gg\tau_0, & (20)\\ 1/2\bar{a}t=(1/\gamma_2 J)\tau_0/t & \text{for } t\ll\tau_0, & (21)\end{cases}$$

i.e., χ_2 is 2D-like for $t\gg\tau_0$ and 3D mean-field-like for $t\ll\tau_0$. Thus τ_0 determines the crossover between 2D and 3D behaviors. Interestingly [Eq. (19)], τ_0 is also the relative shift between the 2D and 3D T_c. It will turn out that τ_0 is also of the same order of magnitude as the 3D Ginzburg critical region.[13]

We now make a further, rather conservative assumption on the 2D system, namely, that it satisfies the usual scaling or homogeneity in the variables

$$\tau=(T-T_c^{2D})/T_c^{2D} \tag{22}$$

and h. This is a property that most systems seem to have at least approximately. Let us thus assume that there exist constants a_τ and a_H such that for any value of λ

$$F_{2D}(\lambda^{a_\tau}\tau,\lambda^{a_h}h)=\lambda F_{2D}(\tau,h). \tag{23}$$

From this we find that if G_1 diverges as $\tau^{-\gamma_2}$, then G_2 has to diverge like $\tau^{-(2\gamma_2+2-\alpha_2)}$, where α_2 is the specific-heat critical exponent of the 2D system, from which it follows using (8) and (12) that

$$b_L(T_c)=JB/\tau_0^{2-\alpha_2-\gamma_2}=JB/\tau_0^{\eta_2\nu_2}, \tag{24}$$

where B is a dimensionless constant (which will be of order unity if indeed A is a constant of the same order of magnitude with an analogous assumption on the amplitude of χ''); we have used the usual scaling law $\gamma_2=(2-\eta_2)\nu_2$, where η and ν are the correlation function critical indices.

Using a_L and b_L [Eqs. (18) and (24)] we find the behavior of the order parameter below T_c, in our approximation, to be

$$m^2=(\text{const})\tau_0^{-1+\eta_2\nu_2}\tau. \tag{25}$$

For reasonable values of η and ν, $\tau_0^{-1+\eta_2\nu_2}\gg 1$, and the rise of the order parameter below T_c is extremely sharp. We also obtain the specific-heat jump [Eq. (15)]

$$\Delta c = k_B(\text{const})\tau_0^{\gamma_2 - 2 + \eta_2\nu_2} = k_B(\text{const})\tau_0^{2(\nu-1)}. \tag{26}$$

Since ν is expected to be larger than unity,[2] Δc will be significantly less here than for a spatially isotropic model. The numerical constants in (25) and (26) will be of order unity provided the amplitudes of χ and χ'' are of the orders of magnitude mentioned above.

It is straightforward to repeat the same procedure for the case $T_c^{2D}=0$. We replace $k_B T_c$ in the above analysis by $J_\parallel$ and the variable τ by $k_B T/J_\parallel$, where $J_\parallel$ is the relevant 2D coupling constant, and make the same considerations as those made for $T_c^{2D} \neq 0$. The results are of course radically different in that $T_c \to 0$ with J. We shall see that the experimental results do not appear to support this possibility.

It is also straightforward to consider the case where the external field h_{ext} is nonzero. All that one has to do is to replace Eq. (6) by

$$h = Jm + h_{\text{ext}} \tag{6'}$$

and repeat the above analysis.

III. CORRELATION FUNCTIONS, 3D GINZBURG-LANDAU FUNCTIONAL, AND 3D CRITICAL REGION

Let us consider the response of our system to a spatially varying external field. For a given Fourier component $\vec{h}_{\vec{q}}$, $\vec{q} = (\vec{q}_\parallel, q_\perp)$, where $\vec{q}_\parallel$ and $q_\perp$ are the components of $\vec{q}$ in the plane of the layer and perpendicular to that plane, respectively, we are interested in the susceptibility $\chi(q)$ defined by

$$\vec{m}_{\vec{q}} = \chi(\vec{q})\vec{h}_{\vec{q}}, \tag{27}$$

where $\vec{m}_{\vec{q}}$ is the $\vec{q}$th component of the order parameter. We shall take both $\vec{h}_{\vec{q}}$ and $\vec{m}_{\vec{q}}$ to point in an arbitrary direction in the order-parameter space. Let us first write the $\vec{q}_\parallel$-dependent susceptibility of a single layer in the simple form for small $q_\parallel$:

$$\chi_{2D}(\vec{q}_\parallel) = \chi_{2D}(0)/(1 + \xi_{2D}^2 q_\parallel^2), \tag{28}$$

where $\chi_{2D}(0) = \chi_{2D}$ and ξ_{2D} is the 2D correlation length assumed to diverge with an exponent ν_2 as $T \to T_c^{2D}$. More sophisticated forms for $\chi_{2D}(\vec{q})$ are possible, involving the critical index η_2, but we shall not consider them here. By our interlayer mean-field (or RPA-type) approximation, generalized to deal with χ_q, the effective field on each spin is the external field plus the mean field owing to the neighboring layers,

$$h_{q,\text{eff}} = h_{q\,\text{ext}} + (J_\perp)_{q_\perp}\sigma_q, \tag{29}$$

where $(J_\perp)_{q_\perp}$ is the $q_\perp$ transform of the interlayer interactions

$$(J_\perp)_{q_\perp} = 2J_\perp(e^{iq_\perp d_\perp} + e^{-iq_\perp d_\perp}) = 4J_\perp \cos q_\perp d_\perp = J\cos q_\perp d_\perp. \tag{30}$$

Since $\sigma_q = h_{q,\text{eff}}\chi_{2D}(q_\parallel)$, we find that the response to the *external* magnetic field is

$$\chi(\vec{q}) = \chi_{2D}(\vec{q}_\parallel)/[1 - J_{\perp,q_\perp}\chi_{2D}(\vec{q}_\parallel)]. \tag{31}$$

[Eqs. (20) and (21) are the $\vec{q}=0$ cases of this Eq. (31).] Let us analyze $\chi(\vec{q})$ for small $\vec{q}$ and t ($q_\parallel \ll 1/\xi_{2D}$, $q_\perp \ll 1/d_\perp$, $t \ll \tau_0$):

$$\chi(\vec{q}) \cong \frac{\chi_{2D}}{2a_L/J + \frac{1}{2}d_\perp^2 q_\perp^2 + q_\parallel^2\xi_{2D}^2} = \frac{\chi_{2D}}{\gamma_2 t/\tau_0 + \frac{1}{2}d_\perp^2 q_\perp^2 + q_\parallel^2 \xi_{2D}^2}. \tag{32}$$

This defines two new characteristic length scales $\xi_\parallel$ and $\xi_\perp$,

$$\xi_\parallel^2 = \xi_{2D}^2\tau_0/t, \quad \xi_\perp^2 = \tfrac{1}{2}d^2\tau_0/t. \tag{33}$$

On the other hand, for $t \gg \tau_0$

$$\chi(\vec{q}) \cong \chi_{2D}(q_\parallel), \tag{34}$$

i.e., the interlayer correlations vanish. This is why τ_0 defines the crossover from 2D to 3D behavior.

Using our results for the correlation lengths and the Landau free energy, it is easy to estimate the Ginzburg[13] critical region t_g signaling the breakdown of our mean-field approximation. This is given by requiring at t_g (we display the equation for the spin model)

$$\frac{a^2}{2b}\left(\frac{\xi_\parallel}{d_\parallel}\right)^2 \frac{\xi_\perp}{d_\perp} \cong kT_c. \tag{35}$$

It follows, using the fact that

$$\chi_{2D}(T_0) = A\beta_c\,\tau_0^{-(2-\eta_2)\nu_2},$$

that (again assuming that A is of order unity)

$$t_g \sim \tau_0. \tag{36}$$

This result provides some justification for our mean-field approximation, since the size of the critical region will vanish with J/T_c^{2D}. However, our results in the 3D regime are quantitatively incorrect, because it follows from (36) that once the system goes into the 3D regime, it also enters the critical region. One thus would expect three-dimensional critical behavior for $t \ll \tau_0$. What this means is that the various singularities for $T \to T_c$ will have the usual 3D critical indices, not the mean-field indices, as in Eqs. (14), (15), (20), (21), and (33). To convince ourselves that the problem for $t \ll \tau_0$ is indeed equivalent to the usual problem of the critical behavior in an isotropic

3D system, we shall exhibit the Landau-Ginzburg free-energy functional, built from our approximation, in an isotropic 3D form. Using (2) and (33) we can write for $t \ll \tau_0$

$$F_{\rm GL}[\psi] = \frac{1}{d_\perp d_\parallel^2} \int d^3x \{ a_L \psi^2 + b_L \psi^4 + \tfrac{1}{2} J [\xi_{\rm 2D}^2 (\nabla_\parallel \psi)^2 + \tfrac{1}{2} d^2 (\nabla_\perp \psi)^2] \}, \tag{37}$$

where $\nabla_\parallel$ and $\nabla_\perp$ are the gradient operators in the plane and perpendicular to the plane, respectively. Here $\psi(x)$ is a field which can be thought of as being obtained by a partial trace over the Fourier components of the field $[\psi(R_i)]$ with $q_\parallel > 1/\xi_{\rm 2D}$, $q_\perp > 1/d_\perp$, or by a partial trace over the Fourier components of $\vec{S}_q$ for $q_\parallel \gtrsim 1/\xi_{\rm 2D}$, $q_\perp \gtrsim 1/d_\perp$. Thus it is very reasonable that all of the static properties of the system will be obtained by the appropriate functional integrals over ψ; e.g., the partition function is $z = \int \delta\psi e^{-\beta F_{\rm GL}[\psi]}$. By scaling distances in the plane and perpendicular to the plane by $\xi_{\rm 2D}$ and $d_\perp/\sqrt{2}$, respectively, we may obtain the above functional in the following isotropic form:

$$F_{\rm GL}^{\rm iso}(\psi) = J(\xi_{\rm 2D}/d_\parallel)^2 \times \int d^3\vec{x} [a_L \psi^2 + b_L \psi^4 + (\nabla\psi)^2]. \tag{38}$$

which is just the usual 3D form as used extensively in the theory of critical phenomena.[14]

IV. CONCLUSIONS AND COMPARISON WITH EXISTING EXPERIMENTS

Our results for the effects of a weak interlayer coupling[15] on the isotropic 2D systems are summarized as follows:

(a) 3D ordering is achieved at a temperature slightly above $T_c^{\rm 2D}$ given by Eq. (8). The quantity $(T_c^{\rm 3D} - T_c^{\rm 2D})/T_c^{\rm 2D}$ defines a small parameter τ_0. There may be a small shift owing to 3D fluctuations in T_c; this shift is further discussed towards the end of this section.

(b) In a temperature region determined by τ_0 above T_c, 3D correlations start to be felt. For $t \gg \tau_0$ the behavior is 2D; for $t \ll t_0$ the behavior is 3D critical.

(c) Below T_c the order parameter should rise with a 3D exponent and with an enhanced amplitude of the kind appearing in Eq. (25).

(d) If measurements of $C(T)$ are not accurate enough to display the true 3D anomaly, the specific heat will appear to have a mean-field-like jump.[11] The magnitude of the jump is given by (26), and it goes to zero in a well-defined way with the interlayer couplings.

We hope that these results will constitute guide lines for the interpretation of experiments on these systems which will completely reveal the pure 2D properties. A set of systematic experiments was already reported in Refs. 8–10 as a function of the interlayer coupling. As we shall see below, the results are in at least a qualitative agreement with the theory, which further supports the tentative conclusion that $T_c^{\rm 2D} \neq 0$. Let us discuss the agreement of existing experimental results with our above predictions (a)–(d).

(a) It was found in Refs. 8–10 that $k_B T_c$ tends to a finite fraction of $J_\parallel$ when $J_\perp \to 0$. In Fig. 1 we have attempted to fit their results with Eq. (17). Assuming $kT_c^{\rm 2D}/J_\parallel \cong 0.44$ as in Ref. 10, the log-log plot of $T_c/J_\parallel - 0.44$ as a function of $J_\perp/J_\parallel$ appears to be a straight line with a slope of $1/\gamma_2$, where $\gamma_2 \sim 2$–3. This is in agreement with the direct measurement of γ_2 in Refs. 10 and 16 as well as with theoretical estimates.[2] This rather high value of γ_2, allowing for the experimental errors, may be also consistent with an exponential divergence of the susceptibility in 2D.[4,17] On the other hand, taking $T_c^{\rm 2D} = 0$ and plotting $T_c/J_\parallel$ as a function of $J_\perp/J_\parallel$ on a log-log scale gives a definite large curvature. Thus the assumption $T_c^{\rm 2D} = 0$ with a power-law divergence of $\chi_{\rm 2D}$ seems to be unjustified. A similar plot (not shown) of $T_c/J_\parallel$ as a function of $\ln(J_\perp/J_\parallel)$ likewise reveals a substantial curvature, so that the assumption $T_c^{\rm 2D} = 0$ with an exponential divergence of $\chi_{\rm 2D}$ also appears to be inconsistent with experiment.

(b) The temperature regions where χ starts to deviate from the 2D result (see Fig. 4 of Ref. 10) for varying $J_\perp/J_\parallel$ are in a qualitative agreement

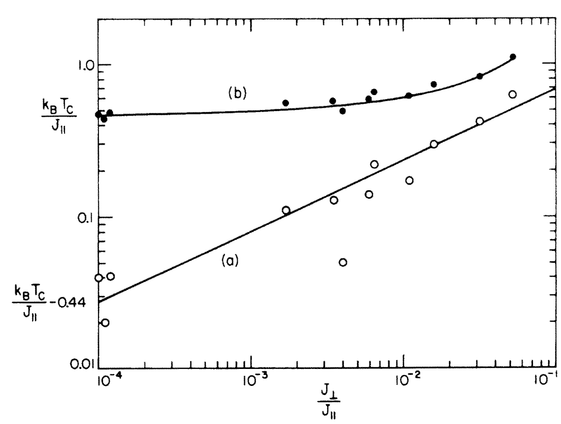

FIG. 1. Log-log plots of $kT_c/J_\parallel$ [curve (a)] and $kT_c/J_\parallel - 0.44$ [curve (b)] versus $J_\perp/J_\parallel$. Line (a) appears to have a definite curvature, while line (b) is approximately a straight line with a slope of 0.35 to 0.55.

with this prediction.

(c) Experimental results on the buildup of the order parameter below T_c as a function of $J_\perp/J_\parallel$ will be of a great relevance.

(d) The area under the specific-heat peak does go to zero with $J_\perp/J_\parallel$. However, more quantitative results are needed to check the theoretical prediction. The anomaly does visually appear to be closer to the mean-field one for weaker $J_\perp$, in agreement with our findings about t_g.

It must be noted that these systems are not exactly isotropic Heisenberg ferromagnets; they do have small xy anisotropies that may in principle make the critical behavior of some of them xy like. This circumstance is of no qualitative importance since the *existence* of a phase transition in the 2D xy model seems to pose problems similar to those appearing in the Heisenberg case.[18] Clearly, more experiments are needed for a full quantitative check of these points, especially regarding point (c) above. The weight of the existing experimental information supports the existence of a finite T_c^{2D} in isotropic ferromagnets.

Unfortunately, as discussed in Sec. I, the very interesting low-temperature zero-field phase of the pure 2D system is not physically realizable within our approximation, owing to the interlayer fields which exist below T_c and have finite values below $T_c^{2D} < T_c$. However, the finite field properties of the pure 2D system are observable in principle in layered samples. Scaling theories of these effects below T_c^{2D} were recently given in Refs. 5 and 15. One should be aware of the downward renormalization of T_c due to fluctuations. This T_c shift, $t_s = [T_c - T_c(\text{mean field})]/T_c(\text{mean field})$, is presumably small because t_g is small. However, it is possible that $t_s > \tau_0$. This is in fact true for spatially isotropic systems for which $t_s \sim t_g^{1/2} \gg \tau_0$, for $t_g \ll 1$. Since our theory does not apply below T_c^{2D}, we cannot use it to estimate t_s. Were t_s larger than t_g, the pure low-temperature 2D phase would have existed at a small temperature region of a relative size $t_s - \tau_0$ below T_c^{2D}. It is interesting that effects of this kind were claimed to have been observed[19] experimentally. However, this question is extremely delicate and further experimental and theoretical work in this direction is needed. It would also be of great interest to obtain the amplitudes of the 3D critical singularities from our free-energy functional (37) or (38).

A basic question which we have not answered fully is: What is the validity of the interlayer mean-field approximation? Since the critical region is small, this type of approximation should be valid over substantial temperature regions. Within the critical region, one feels that the Ginzburg-Landau functional derived here [Eqs. (37) and (38)] should be the appropriate one to use in a more complete (e.g., a renormalization group) treatment, as discussed at the end of Sec. III. This is based on plausible physical ideas, but we do not claim to have established it rigorously.

ACKNOWLEDGMENTS

The author would like to thank T. A. Kaplan, P. Pincus, and D. J. Scalapino for fruitful discussions.

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*Research at the University of California, Santa Barbara, supported by the U. S. Army Research Office, Durham, N. C.

†Research at Tel-Aviv University supported by The Commission for Basic Research, Israel Academy of Sciences, Jerusalem, Israel.

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