Critical Behavior of Magnets with Lattice Coupling

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(Received 6 June 1973)

The exact spin Hamiltonian, induced by linear exchange coupling to a harmonic lattice with fixed periodic boundary conditions, is considered in the framework of renormalization-group recursion relations. Neglecting irrelevant variables, the Hamiltonian amounts to a replacement of the four-spin amplitude u_0 by $a(T - T_1)$, with T_1 proportional to the lattice compressibility. Hence the system exhibits a critical point with unrenormalized exponent values when $T_c > T_t$, but presumably a first-order transition for $T_c < T_t$, where $T_t \simeq T_1$. The point $T_c = T_t$ is expected to be a classical tricritical point. Experiments on NH_4Cl are considered briefly.

I. INTRODUCTION

The critical behavior of a system which has additional "hidden" variables, such as a compressible ferromagnet, has been the subject of considerable controversy in recent years. The general effects of constraints upon the critical behavior of thermodynamic systems were first investigated systematically by Fisher.[1] For the compressible ferromagnet, Fisher's arguments suggested that only for zero pressure (or zero external forces) would the system behave precisely as in the rigid model; in most other cases the transition remains continuous, but with renormalized exponents.[1]

Indeed, Baker and Essam[2] have recently constructed a soluble model of a compressible Ising lattice, but without shear forces, which exhibits Fisher's renormalized behavior for the constraint of constant volume. This model was later extended by Gunther, Bergman, and Imry[3] to include other types of constraints, and then exhibited renormalized critical behavior for positive pressures or fixed volume, but a first-order transition for negative pressures. A first-order transition is also found by Baker and Essam[4] in an approximate treatment of a model with nonzero shearing forces. Similar conclusions are obtained by Larkin and Pikin,[5] who use a continuum model for the elastic lattice terms.

Since the Baker-Essam model is oversimplified, it is interesting to consider a Hamiltonian with a somewhat more realistic representation of the lattice degrees of freedom, as studied by Wagner.[6] By integrating exactly over the harmonic-lattice degrees of freedom, for the constraint of zero external forces, Wagner generated an exact effective-spin Hamiltonian, with long range four-spin interactions. To analyze this spin Hamiltonian, Wagner applied the approach of Fisher's droplet model.[7] He concluded, in contradistinction to Fisher's renormalization theory, that the critical behavior differed significantly from that of the rigid lattice.

With the aid of Wilson' s renormalization-group approach,[8] one is now able to treat Wagner' s Hamiltonian by more precise methods. In this note we report the result of such a treatment, and show that under the constraint of zero pressure (or zero external forces) one may obtain *either* nonrenormalized second-order transition, as predicted by Fisher (without Wagner' s modifications) or an instability which may lead to a first-order transition. The latter may be related to the behavior found (at negative pressures) by Gunther, Bergman, and Imry[3] on the basis of the Baker-Essam model. These results are clearly in disagreement with Larkin and Pikin,[5] and we shall discuss this later.

II. WAGNER HAMILTONIAN

The actual calculation starts with the Wagner-Swift Hamiltonian,

$$\mathcal{H} = \Phi + \Psi - K_m^\mu \gamma_m^\mu + \Psi_m^\mu u_m^\mu + (1/2M) p_m^\mu p_m^\mu + \tfrac{1}{2} u_m^\mu (\Phi_{mn}^{\mu\nu} + \Psi_{mn}^{\mu\nu}) u_n^\nu , \quad (1)$$

where $\vec{u}_m$ is the displacement of the ion at the lattice point $\vec{r}_m^{\,0}$ [defined by the lattice equilibrium condition $\Phi_m^\mu(\vec{r}_n^{\,0}) = K_m^\mu$], $\vec{P}_m$ is the conjugate momentum, M is the atomic mass, Φ is the harmonic potential, and

$$\Psi = -\tfrac{1}{2} \sum_{mn} J(\vec{r}_m^{\,0} - \vec{r}_n^{\,0}) s_m s_n \quad (2)$$

is the magnetic exchange Hamiltonian, in which s_m is the single-component spin variable of the ion at the site $\vec{r}_m$. All our calculations may be repeated with $\vec{s}_m$ replacing s_m; thus the results also apply to compressible Heisenberg magnets. The subscript on Ψ_m^μ means $\partial\Psi/\partial\gamma_m^\mu$, and Φ_m^μ, $\Psi_{mn}^{\mu\nu}$, and $\Phi_{mn}^{\mu\nu}$ are defined similarly. The external force on the ion at $\vec{r}_m$ is denoted by $\vec{K}_m$. A summation over repeated indices is implied everywhere. Following Wagner and Swift,[6] we shift

$\vec{u}_m$ so that the term $\Psi^\mu_m u^\mu_m$ in (1) is eliminated.[9] This shift amounts to finding the local equilibrium positions of the ions. After this transformation $\mathcal{H}$ becomes exactly

$$\mathcal{H} = \Phi + \Psi - K^\mu_m \gamma^\mu_m + \tfrac{1}{2}\Psi^\mu_m \mathfrak{D}^{\mu\nu}_{mn}\Psi^\nu_n + (1/2M)p^\mu_m p^\mu_m + \tfrac{1}{2}u^\mu_m(\Phi^{\mu\nu}_{mn} + \Psi^{\mu\nu}_{mn})u^\nu_n , \tag{3}$$

where the Green's function $\mathfrak{D}^{\mu\nu}_{mn}$ is defined by

$$(\Phi^{\mu\lambda}_{ml} + \Psi^{\mu\lambda}_{ml})\mathfrak{D}^{\lambda\nu}_{ln} = -\delta_{mn}\delta_{\mu\nu} . \tag{4}$$

If one sets $\Psi^{\mu\nu}_{mn} = 0$, which is equivalent, for cubic lattices, to assuming $J(\vec{r})$ to be a *linear* function of $|\vec{r}|$, then (4) can be solved using the known lattice potentials. If, in addition, one assumes fixed *periodic boundary conditions*, the solution becomes

$$\mathfrak{D}^{\mu\nu}_{mn} = -\sum_j \int_{\vec{q}} \frac{e^\mu_j(\vec{q})\, e^\nu_j(\vec{q})}{\rho\omega^2_j(\vec{q})} e^{i\vec{q}\cdot(\vec{r}^0_m - \vec{r}^0_n)} , \tag{5}$$

where ρ is the mass density of the lattice, $\omega_j(\vec{q})$ is the dispersion relation for the lattice mode of wave vector $\vec{q}$ and polarization vector $\vec{e}_j(\vec{q})$ ($j = 1, 2, \ldots d$), and $\int_{\vec{q}}$ denotes $(2\pi)^{-d}$ times an integral over the first Brillouin zone. With these assumptions Wagner obtained a complete separation between the lattice and the spin Hamiltonians, with the exact effective-spin Hamiltonian

$$\mathcal{H}_s = \Psi + \tfrac{1}{2}\Psi^\mu_m \mathfrak{D}^{\mu\nu}_{mn}\Psi^\nu_n . \tag{6}$$

The appearance of $\Psi^{\mu\nu}_{mn}$ in (3) is shown by Wagner and Swift[6] to be important for retaining the symmetry properties of the elastic constants. They actually estimate the contribution of $\Psi^{\mu\nu}_{mn}$ to the elastic constants by using an average $\langle\Psi^{\mu\nu}_{mn}\rangle$, calculated with (6), and so obtain renormalized values for the elastic constants. Note that if $\Psi^{\mu\nu}_{mn} = 0$, the elastic constants, and the complete phonon spectrum $\omega_j(\vec{q})$, remain as in the nonmagnetically coupled lattice.

III. CONTINUOUS-SPIN MODEL

We now consider a continuous spin model, with a weighting factor $\exp(-\frac{1}{2}s^2_m - us^2_m - vs^6_m - \ldots)$. On Fourier transforming the variables in (6), we finally arrive at a reduced Hamiltonian of the form

$$\overline{\mathcal{H}} = -\tfrac{1}{2}\int_{\vec{q}}(r+q^2)\sigma_{\vec{q}}\sigma_{-\vec{q}} - \int_{\vec{q}_1}\int_{\vec{q}_2}\int_{\vec{q}_3} U(\vec{q}_1, \vec{q}_2, \vec{q}_3, -\vec{q}_1, -\vec{q}_2, -\vec{q}_3)\sigma_{\vec{q}_1}\sigma_{\vec{q}_2}\sigma_{\vec{q}_3}\sigma_{-\vec{q}_1-\vec{q}_2-\vec{q}_3}$$
$$+ \int_{\vec{q}_1}\int_{\vec{q}_2}\int_{\vec{q}_3}\int_{\vec{q}_4}\int_{\vec{q}_5} V(\vec{q}_1, \cdots, \vec{q}_5, -\vec{q}_1 - \cdots - \vec{q}_5)\sigma_{\vec{q}_1}\sigma_{\vec{q}_2}\sigma_{\vec{q}_3}\sigma_{\vec{q}_4}\sigma_{\vec{q}_5}\sigma_{-\vec{q}_1-\vec{q}_2-\vec{q}_3-\vec{q}_4-\vec{q}_5}\cdots , \tag{7}$$

where the integrals now run over $0 < |\vec{q}| < \pi$ (the equilibrium lattice spacing a has been chosen as 1), and the spins are rescaled by a factor $s_0 = (2dk_B T/cJa^{d+2})^{1/2}$, in which J is the equilibrium nearest-neighbor exchange parameter, and c is the coordination number. The four-spin coupling potential, using (5), is

$$U(\vec{q}_1, \vec{q}_2, \vec{q}_3, \vec{q}_4) = s^4_0 a^{3d}\left(u + \sum_j \frac{\hat{\vec{J}}(\vec{q}_1)\cdot\vec{e}_j(\vec{q}_1+\vec{q}_2)\hat{\vec{J}}_1(\vec{q}_3)\cdot\vec{e}_j(\vec{q}_1+\vec{q}_2)}{2k_B TM\omega^2_j(\vec{q}_1+\vec{q}_2)}\right) \tag{8}$$

where M, the atomic mass, replaces ρa^d, and

$$i\hat{J}^\mu_1(\vec{q}) = iq^\mu[cJ\gamma_M + O(q^2)] \tag{9}$$

is the Fourier transform of $\partial J(\vec{r})/\partial r^\mu$ [we assume that $J(\vec{r})$ is a function of $|\vec{r}|$, and we sum only over nearest neighbors]. The parameter γ_M is $d^{-1}d\ln J/d\ln r \simeq d\ln J/d\ln V$, with the derivative calculated at the nearest-neighbor site. The six-spin coupling potential is simply proportional to the parameter v of the weighting function. More generally, V could include six-spin terms from the spin Hamiltonian.[10] We shall assume $V \simeq v_0$.

For small $\vec{q}$, we follow Wagner[6] in approximating $\omega_j(\vec{q})$ by $\bar{v}|\vec{q}|$, where $\bar{v}$ is an average phonon velocity. [The simplification that $\bar{v}$ is independent of j may easily be avoided. Similarly, the assumption that $\omega_j(\vec{q})$ depends only on the magnitude of $\vec{q}$ may be lifted, and angular variation characteristic of cubic symmetry may be introduced.] Thus, we obtain the crucial expression

$$U(\vec{q}_1, \vec{q}_2, \vec{q}_3, \vec{q}_4) = \gamma^2_M(2d)^2 a^{4-d}\left[u\left(\frac{k_B T}{\gamma_M cJ}\right)^2 + \frac{k_B T}{2M\bar{v}^2}\frac{\vec{q}_1\cdot\vec{q}_3}{(\vec{q}_1+\vec{q}_2)^2} + O(q^2_i)\right] \tag{10}$$

in which we do not need the explicit forms of the $O(q^2_i)$ terms, since they are irrelevant in the renormalization group sense near four dimensions. [Their exponent is $\epsilon - 2 + O(\epsilon^2) < 0$ for $\epsilon = 4 - d \ll 1$.]

Since the integral in (7) is symmetric under permutations of $\vec{q}_1$, $\vec{q}_2$, $\vec{q}_3$, $\vec{q}_4$, we may replace $U(q_1, \cdots, q_4)$ by an average over all these twelve permutations. Using the constraint $\vec{q}_1 + \vec{q}_2 + \vec{q}_3 + \vec{q}_4$

$= 0$, simple algebra yields

$$U(\vec{q}_1, \vec{q}_2, \vec{q}_3, \vec{q}_4) = \gamma_M^2 (2d)^2 a^{d-4} \times \left[u \left(\frac{k_B T}{\gamma_M c J} \right)^2 - \frac{k_B T}{8 M \bar{v}^2} + O(q_i^2) \right] , \quad (11)$$

which leads precisely to the Wilson[8] Hamiltonian, with

$$u_0 = (2d)^2 a^{d-4} (cJ)^{-2} u T k_B^2 (T - T_1) , \quad (12)$$

where

$$k_B T_1 = \gamma_M^2 (cJ)^2 / 8 M \bar{v}^2 u = \gamma_M^2 (cJ)^2 n \kappa_T / 8u , \quad (13)$$

in which n is the number density, while κ_T is the lattice compressibility.

IV. RENORMALIZATION GROUP NEAR FOUR DIMENSIONS

We now consider the renormalization-group recursion relations near four dimensions. If $T_c > T_1$, then u_0 is positive near the critical point, and the iterations lead to the usual fixed points. For $\epsilon > 0$ we obtain exactly the same nontrivial fixed point, and hence the same exponents, as found for the rigid lattice. Since the Fisher renormalization of critical exponents[1] involves a factor $(1 - \alpha)$, and α is of order ϵ,[8] we thus predict a nonrenormalized behavior already in order ϵ. We expect more generally that *the second-order transition for the Wagner-Swift Hamiltonian has nonrenormalized exponents to all orders in* ϵ.

If $T_c < T_1$, then for temperatures in the range $T_c < T < T_1$, u_0 is negative. If $v_0 \gtrsim |u_0|$, then the v_l term in the recursion relation for u_l may build up a positive u_l, which will still go to the usual nontrivial fixed point. However, if $|u_0| \gg v_0$, this will not happen, since v_l is an irrelevant parameter for $d \geq 3$, and hence decays to zero, while u_l will grow exponentially to $-\infty$. Numerical studies on the approximate recursion relations suggest[11] that such a situation usually leads to an effective Hamiltonian with two minima, and hence to a first-order transition. We are thus led to the conclusion that there exists a tricritical point, with T_t differing from T_1 by terms of order $T_1 v_0$, such that *for* $T_c < T_t$ *one finds a first-order transition.* Note that in all the above discussion we must have $v_0 > 0$, since otherwise the Hamiltonian $\bar{\mathcal{H}}$ of (7) is unstable (the partition function is undefined). Clearly one can always choose v in the weighting factor so that this will be the case. (If the Hamiltonian includes six-spin terms with large negative coefficients, then one will have to go to higher-order terms.)

The situation with $u_0 < 0$, $v_0 > 0$ was studied by Riedel and Wegner,[12] who found that the point $r^* = u^* = v^* = 0$ is a fixed point, which, for $d \geq 3$, corresponds to the Blume-Emery-Griffiths[13] tricritical point with classical exponents (with logarithmic corrections at $d = 3$). Thus, for appropriate values of r_0, u_0, and v_0 (which give the actual value of T_t) the recursion relations will lead to the above fixed point. *We thus expect classical tricritical behavior at* T_t.

V. COMPARISON WITH EXPERIMENTS AND DISCUSSION

To estimate T_1, we need a value for u; but this is a feature of the particular physical model considered. For $d = 3$, we expect reasonable values of u to be of order unity. Thus, using $cJ = 2k_B T_c$,[14] we find $T_1/T_c = \frac{1}{2} k_B T_c n \kappa_T \cdot \gamma_M^2$. For Wagner's illustrative values[6] $T_c = 100\,^\circ\mathrm{K}$, $n = 10^{22}\ \mathrm{cm}^{-3}$, $\kappa_T = 10^{-12}\ \mathrm{cm}^2/\mathrm{dyn}$, $\gamma_M = 1$, one finds $T_1/T_c \simeq 10^{-4}$. Our model thus predicts that typical Ising-like systems will behave exactly as rigid lattices. Indeed, the data for β-brass[15] is not inconsistent with this conclusion.

The above numbers change appreciably for a highly compressible lattice, like NH_4Cl. For this material, which seems to be well described by a compressible Ising model, Garland and Weiner[16] indeed find a change from a first-order to a second-order transition at $T_t = 250\,^\circ\mathrm{K}$ and $p_t = 1500$ bar. Although the assumptions of zero external forces and of harmonic lattice coupling may not be justified at this pressure, it is instructive to examine the value of T_1 for this case. As we noted after (6), the elastic constants of Wagner's model are the same as for the rigid lattice, unless one takes into account some average magnetic contribution. From the Weiner-Garland[16] results, the rigid-lattice compressibility (far from T_c) is $\kappa_T \simeq 5.91 \times 10^{-6}\ \mathrm{bar}^{-1}$. The value of γ_M may be estimated from the variation of the critical temperature with the volume. From the Renard-Garland measurements[17] one finds $\gamma_M \simeq -10.7$. Together with $n = 1.7 \times 10^{22}\ \mathrm{cm}^{-3}$ we find $T_1/T_c \simeq 0.2$. Remembering (i) that u might well be equal to 0.1, (ii) that we do not know the order-v contribution to $(T_1 - T_t)$, and (iii) that the actual compressibility is larger than the above value, because of the average magnetic contribution, this is an encouraging order-of-magnitude agreement.

Another positive indication concerns the specific-heat exponent α, for which Weiner and Garland[16] find a value which is quite consistent with the classical tricritical $\alpha = \frac{1}{2}$. On the other hand, measurements near the tricritical point in ND_4Cl[18] give $2\beta = 0.36 \pm 0.01$, which seems significantly lower than the classical tricritical $2\beta = \frac{1}{2}$. However, this discrepancy may be related to the Wegner-Riedel[12] logarithmic corrections at $d = 3$.

The main difference between the present model and that of Larkin and Pikin[5] lies in our assumption of fixed periodic boundary conditions, which

led to (5). Under this assumption, it is impossible to obtain the nonzero uniform deformations indicated by Larkin and Pikin, which lead in their work to a first-order transition whenever the magnetic specific heat becomes large enough. Although it is not clear if this assumption is justified, the observation on NH_4Cl suggests quite strongly that a real tricritical point can in fact occur. Since the specific-heat exponent near the tricritical point is relatively large, one might expect a strong divergence of the specific heat, and hence the Larkin-Pikin theory would predict a first-order transition instead of the observed behavior.

Note added in proof. The important role of boundary conditions for the present problem has been demonstrated in one dimension by S. R. Salinas [J. Phys. A (to be published) and private communication], who also, independently, derived the result (12).

ACKNOWLEDGMENTS

The author is indebted to Professor Michael E. Fisher, for many useful comments on the manuscript and on the subject, and to Professor Robert B. Griffiths, Professor Kenneth G. Wilson, Professor Yoseph Imry, Professor A. I. Larkin, and Professor Franz J. Wegner for informative discussions. The support of the National Science Foundation, partly through the Materials Science Center at Cornell University, and a scholarship received from the Fulbright-Hays Committee are also acknowledged.

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