PHYSICAL REVIEW B VOLUME 19, NUMBER 9 1 MAY 1979

Local-band theory of itinerant ferromagnetism. V. Statistical mechanics of spin waves

R. E. Prange and V. Korenman
Department of Physics and Astronomy, University of Maryland, College Park, Maryland 20742
(Received 14 August 1978)

The functional-integral method of the previous paper is generalized to allow a discussion of the dynamical spin fluctuations in itinerant-electron magnets. The magnitude fluctuations and nonstatic spin fluctuations are treated as a perturbation on the nonlinearly interacting static spin fluctuations. The result is a free energy which is a sum of a Stoner contribution together with correlation corrections, a classical Heisenberg free energy as found in the previous paper, and a quantum spin-wave free energy minus its zero-frequency part.

I. INTRODUCTION

In this paper we are concerned with the effects on the thermodynamics of the dynamics of the motion of the magnetization of itinerant ferromagnets. The best known such effect is the $T^{3/2}$ law for the magnetization. In contrast, the phase transition itself is primarily a consequence of classical statistical mechanics and the dynamics of the spin motion is a secondary effect.

The method we use is that of expressing the thermodynamic quantities as a functional integral via an appropriate Stratonovich-Hubbard transformation. As we discussed in the previous paper,[1] earlier attempts using this method have certain serious shortcomings. This has led us to use a generalization of the so-called "two-field" approach, which makes possible spin-rotationally invariant approximations, while retaining the virtues of the two-field method.

Illustrating the method with the one-band Hubbard model, we have an exact expression for the partition function

$$Z=\int \mathfrak{D}x\mathfrak{D}\vec{\mu}\,\exp\left(-\tfrac{1}{4}U\int_0^\beta d\tau\sum_i x^2 - U\int_0^\beta d\tau\sum_i \vec{\mu}^{\,2}\right)e^{-\beta\mathfrak{F}_0[x,\vec{\mu}]} \quad , \tag{1}$$

where

$$e^{-\beta\mathfrak{F}_0[x,\vec{\mu}]}=\mathrm{Tr}\; e^{-\beta H_0}\left[\exp\left(\tfrac{1}{2}iU\int_0^\beta d\tau\sum_i nx\right)\exp\left(2U\int_0^\beta d\tau\sum_i \vec{\mu}\cdot\vec{\mathrm{M}}\right)\right]_+ \quad . \tag{2}$$

Here $x=x_i(\tau)$, $\vec{\mu}=\vec{\mu}_i(\tau)$ are functions of site i and (imaginary) time τ;

$$n=n_i(\tau)=e^{\tau H_0}\sum_s c_{is}^\dagger c_{is}e^{-\tau H_0},\quad \vec{\mathrm{M}}_i(\tau)=\tfrac{1}{2}e^{\tau H_0}\sum_{ss'}c_{is}^\dagger\vec{\sigma}_{ss'}c_{is'}e^{-\tau H_0}$$

are the position- and "time"-dependent density and magnetization density operators. The Hubbard Hamiltonian may be written

$$H=H_0+\tfrac{1}{4}U\sum_i n_i^2-\sum_i U(\vec{\mathrm{M}}_i\cdot\hat{\mu}_i)^2 \quad , \tag{3}$$

where $\hat{\mu}_i$ is an arbitrary unit vector. We use this form to motivate the expression (1). Finally, the functional integral is $\mathfrak{D}x=C\prod_{i\tau}dx_i(\tau)$ and

$$\mathfrak{D}\vec{\mu}=C\prod_{i\tau}d\mu_i(\tau)\,d^2\hat{\mu}_i(\tau) \quad , \tag{4}$$

where we have indicated certain constants, irrelevant for our purposes, by C. The form (4) for $\mathfrak{D}\vec{\mu}$, which is less convenient and certainly less familiar than one involving $d^3\mu_i(\tau)$, is the price paid for this choice of transformation. It differs from the choice in Paper IV by the τ dependence of the unit vectors $\hat{\mu}$. Clearly, approximate treatments will distinguish between magnitude fluctuations and angle fluctuations. We have argued before that this is physically correct in the cases of interest, particularly, for iron and nickel.

We do not wish to dwell here on the effects of density fluctuations,[2] so we replace $x_j(\tau)$ by its most important value, $x_j(\tau) = i\langle n\rangle$, and suppress further mention of it. A similar replacement of $\vec{\mu}_j(\tau)$ by $\vec{M}_s = \langle\vec{M}_j(\tau)\rangle$ gives the Hartree-Fock approximation.

In Paper IV, we neglected the time dependence of $\vec{\mu}_j(\tau)$, to obtain a formulation of the thermodynamics. Here, we wish to account for the time dependence in an approximate way. This is necessary if quantum effects are to be studied. For example, this must be done to obtain the $T^{3/2}$ law alluded to earlier.

We find in Sec. II that the neighborhood of the minimizing path gives the free energy conventionally found from RPA spin waves. This is satisfying as it has not been previously obtained by functional integral methods because up to now transverse spin fluctuations have not been treated properly.

At higher temperatures paths with deviations far from the uniform field solution are important. In Sec. III, we make an approximation keeping paths whose high frequency excursions are small, although the time-average deviation may be large.

II. SPIN WAVES

We now turn to a study of the expression

$$e^{-\beta\mathfrak{F}[\mu]} = \exp\left[-U\int_0^\beta d\tau \sum_i \vec{\mu}^2\right] \operatorname{Tr} e^{-\beta H_0} \times \left[\exp\left[2U\int_0^\beta d\tau \sum_i \vec{\mu}\cdot\vec{M}\right]\right]_+ , \tag{5}$$

where the effects of x are incorporated into H_0. Our previous point of view was that the paths which were of most interest were those minimizing $\mathfrak{F}[\mu]$, in other words, those satisfying

$$\vec{\mu}_i(\tau) = \langle\vec{M}_i(\tau)\rangle_\mu , \tag{6}$$

where

$$\langle\vec{M}_i(\tau)\rangle_\mu = e^{\beta\mathfrak{F}_0} \operatorname{Tr} e^{-\beta H_0} \times \left[\vec{M}_i(\tau)\exp\left[2U\int_0^\beta d\tau \sum_i \vec{\mu}\cdot\vec{M}\right]\right]_+ . \tag{7}$$

The basic solution of Eq. (6) is

$$\vec{\mu}_i(\tau) = \vec{M}_s \tag{8}$$

and M_s is the Stoner magnetization at temperature $T = 1/\beta$.

We here keep small fluctuations about this solution, i.e., we expand $\mathfrak{F}[\mu]$ in powers of $\vec{\mu} - \vec{M}_s \equiv \delta\vec{\mu}_i(\tau)$. This is appropriate well below the transition temperature. Then we have

$$\mathfrak{F}[\mu] = \mathfrak{F}_s(T) + \sum_{\alpha\beta}\int_0^\beta\int_0^\beta d\tau\, d\tau' \sum_{ij} K_{ij}^{\alpha\beta}(\tau-\tau')\,\delta\mu_i^\alpha(\tau)\,\delta\mu_j^\beta(\tau') . \tag{9}$$

$\mathfrak{F}_s(T)$ is the Stoner free energy. The kernel K is closely related to the reciprocal of the random-phase approximation (RPA) susceptibility,

$$K_{ij}^{\alpha\beta}(\tau-\tau') = U[\delta_{\alpha\beta}\delta_{ij}\delta(\tau-\tau') - 2U\chi_{ij}^{\alpha\beta}(\tau-\tau')] \tag{10}$$

and

$$\chi_{ij}^{\alpha\beta}(\tau-\tau') = \langle(M_i^\alpha(\tau)M_j^\beta(\tau'))_+\rangle - \langle M_i^\alpha\rangle\langle M_j^\beta\rangle \tag{11}$$

is the susceptibility of free electrons (Hamiltonian H_0) in a constant exchange field $2UM_s$.

The kernel is diagonalized as usual, by the introduction of the Fourier representation, and the transverse parts of $\vec{\mu}$ are represented by $\delta\mu^\pm = \delta\mu^x \pm i\delta\mu^y$. We also have

$$d\mu^x d\mu^y \simeq M_s^2\theta\, d\theta\, d\phi = M_s^2\, d^2\hat{\mu} \tag{12}$$

valid for small fluctuations.

The result for the longitudinal susceptibility is

$$K^{zz}(q,i\omega_n) = U\left[1 - \tfrac{1}{2}U\sum_\sigma\int\frac{d^3k}{(2\pi)^3}\frac{f(E_{k+q,\sigma}) - f(E_{k\sigma})}{i\omega_n - (E_{k+q,\sigma} - E_{k\sigma})}\right] . \tag{13}$$

Since K^{zz} has period β, $\omega_n = 2\pi n T$. Near $q=0$, this formula gives (for $\vec{v}_k \cdot \vec{q} << 2\pi T$),

$$K^{zz}(q,i\omega_n) \sim U, \quad \omega_n \neq 0 \ , \qquad (14)$$

$$K^{zz}(q,0) = U[1 - \tfrac{1}{2}U(N_+ + N_-)] \ ,$$

where $N_\pm$ is the density of states at the ($\pm$) Fermi surface. Actually, in this case the coupling to the fluctuations in total density is of some importance. It is usually taken to be a better approximation to require the total electron density to be fixed, (although this is not a consequence of the simple Hubbard model we are using). In that case, the expressions for $K^{zz}(q,0)$ becomes

$$K^{zz}(q,0) = U\left[1 - 2U\left(\frac{1}{N_+} + \frac{1}{N_-}\right)^{-1}\right] \ . \qquad (15)$$

Since K^{zz} does not vanish or become small, its contribution to the total free energy represents a correlation correction of quantitative interest only. It would predict large fluctuation effects only near the Stoner transition temperature.

The configurations of $\vec{\mu}$ such that K vanishes or is unusually small are of special importance. These correspond to low-lying elementary excitations and the system is soft against distortion in that direction. In this case, it is the long-wavelength low-frequency transverse fluctuations which are soft. In fact, the transverse static long wave susceptibility χ is just M/H, i.e., $M_s/2UM_s$, which shows that Eq. (10) vanishes in this limit.

The transverse responses, K^{+-} and $K^{-+} = (K^{+-})^*$ are given by

$$K^{+-}(q,i\omega_n) = 2U\left[1 - U\int \frac{d^3k}{(2\pi)^3} \times \frac{f(E_{k,+}) - f(E_{k+q,-})}{i\omega_n + E_{k+q,-} - E_{k,+}}\right] \ . \qquad (16)$$

For $v_k q << \Delta$, $\omega_n << \Delta$, Eq. (16) is approximated by

$$K^{+-}(q,i\omega_n) \simeq \frac{1}{M_s}(i\omega_n + \omega_q) \ , \qquad (17)$$

with $\omega_q = Dq^2$ at small q. The contribution of these terms to the free energy is

$$F_{\text{sw}} = T\sum_q \sum_n \ln\frac{(i\omega_n + \omega_q)}{T} + \text{const} \ . \qquad (18)$$

This free energy is, up to an irrelevant constant term, the standard expression for the free-energy contribution of spin waves,

$$F_{\text{sw}} = (T\sum_q \ln(1 - e^{-\beta\omega_q}) \ . \qquad (19)$$

It follows that the magnetization obeys the usual $T^{3/2}$ law. Thus, this formulation is capable of giving the correct low-temperature properties, and is the first formulation of the Stratonovich-Hubbard variety to do so.

III. NONLINEAR CASE

As the temperature rises, the magnetization can fluctuate in direction over substantial angles. The question arises how to deal with the time dependence of such fluctuations.

We here assume that those configurations which do not deviate too much from their time average, are of special importance.

We put $\vec{\mu}_i(\tau) = M_s\hat{\nu}_i + \delta\vec{\mu}_i(\tau)$ and expanding in $\delta\vec{\mu}_i$, we evaluate Eq. (5) as

$$\beta\mathfrak{F}[\vec{\mu}] = U\int_0^\beta d\tau \sum_i \delta\vec{\mu}^{\,2} + \beta UM_s^2 + 2UM_s\int_0^\beta d\tau\sum_i \delta\vec{\mu}\cdot\hat{\nu} + \mathfrak{F}_0[M_s\hat{\nu}_i] - 2U\int_0^\beta d\tau \sum_i \delta\vec{\mu}_i\cdot\langle\vec{M}_i\rangle$$
$$- 2U^2\int_0^\beta\int_0^\beta d\tau\, d\tau' \sum_{ij}\sum_{\alpha,\beta} \delta\mu_i^\alpha(\tau)\chi_{ij}^{\alpha\beta}(\tau,\tau')\delta\mu_j^\beta(\tau') \ . \qquad (20)$$

The unit vector $\hat{\nu}_i$ is determined by the requirement that

$$\int_0^\beta d\tau[\delta\vec{\mu}_i(\tau)\times\hat{\nu}_i] = 0 \ .$$

The quantity $\delta\vec{\mu}_i\cdot\hat{\nu}_i$ is regarded as small, but does not have to vanish, on the average.

We again make the assumption of short-range order,[3–9] and expand in powers of $\hat{\nu}_i - \hat{\nu}_j$, where i,j are neighbors. The expression (20) is most easily studied in the spin coordinate system (LRSCS) locally rotated so that $\hat{\nu}_i$ is the z direction. The form of the expression is preserved by this rotation, but the expectation values $\langle M^\alpha \rangle$ and $\chi^{\alpha\beta}$ are to be evaluated with the Hamiltonian

$$H = H_0 - 2UM_s M^z + H_1 + H_2 \quad , \tag{21}$$

where H_1 and H_2 are given in Paper IV. The single-particle eigenstates of Eq. (21) with neglect of H_1 and H_2 are just those of the Stoner model, $E_{p\sigma}$.

In Paper IV, we expanded $\mathfrak{F}_0$ to second order in the small quantity a_{ij},

$$|a_{ij}|^2 = \tfrac{1}{4} |\hat{\nu}_i - \hat{\nu}_j|^2 \quad .$$

The result was an effective Heisenberg free energy in addition to the Stoner free energy. We shall expand $\langle M_i^\alpha \rangle$ to first order, and χ is evaluated to zeroth order. Thus we have

$$\langle M_i^\alpha \rangle = \delta_{\alpha z} M_s + m_i^\alpha \quad . \tag{22}$$

In the LRSCS, m_i^α is purely transverse, to the order calculated, and independent of time.

Thus, to the order calculated, the fluctuations in the magnitude of the order parameter remain uncoupled from the transverse and their contribution is exactly that given earlier. The temperature, in other words, enters only through the Fermi factors appearing in Eq. (13), and this dependence is weak for T less than the Stoner temperature.

The transverse fluctuations may be diagonalized as before, and since their time average vanishes, the zero frequency term is eliminated. The term in m_i does not contribute because of this. The contribution to the free energy is just the spin-wave contribution, Eq. (18), less the zero frequency term; i.e.,

$$\mathfrak{F}_{\rm sw}' = T \sum_q [\ln(1 - e^{-\beta\omega_q}) - \ln\beta\omega_q] \quad . \tag{23}$$

IV. DISCUSSION

The total free energy is thus a sum of four contributions. The first is the Stoner free energy, i.e., the free energy of a system of noninteracting metallic electrons. A contribution of this type is certainly needed on experimental grounds. The second is from the longitudinal (and density) fluctuations. This is a correlation correction of a familiar type. The third is from the effective Heisenberg Hamiltonian, i.e., from the static magnetization deviations. This contribution was the subject of Paper IV. At low temperatures this contribution just cancels the second term in Eq. (23), but at high temperatures the nonlinear effects are important. The phase transition is dominated by this term. Finally, there is a "finite frequency" spin-wave contribution.

To this order, there is no coupling between the various contributions. It is straightforward, though tedious, to extend the calculation somewhat to include some such coupling. One may calculate, for example, $\chi_{ij}^{\alpha\beta}$ of Eq. (20) treating the terms H_1, and H_2, in the Hamiltonian of Eq. (21) as a perturbation. This will introduce terms coupling $\delta\mu^z$ and $\delta\mu^{x,y}$. The effect on $K_q(\omega_n)$ will be most prominent at small q, which, according to Eq. (23), is not too important anyway.

There is also a coupling we have neglected between the finite frequency and static transverse fluctuations. This comes from the linearization of the $d^2\hat{\mu}$. We have in essence assumed that the time average of $\hat{\mu}_i(\tau)$ is a unit vector, $\hat{\nu}_i$. This time average will be approximately a unit vector if $|\hat{\nu}_i \times \hat{\mu}_i|^2$ is small on the average. The smallness of this quantity is the basis of our approximation, however.

One can of course, *a posteriori*, estimate $\langle |\delta\hat{\mu}_i(\tau)|^2 \rangle$, to be approximately

$$\frac{1}{M_s^2} \frac{T}{N} \sum_q \sum_n \frac{1}{K_q^{+-}(\omega_n)} \quad .$$

Using Eq. (16) we see that there is an ultraviolet divergence, since K becomes constant for large n. This divergence means that the paths are varying with infinite amplitude at infinite frequency. Such paths are incorrectly treated by Gaussian expansion, and it must be imagined that a renormalization has been carried out in which such paths have been integrated out in favor of a renormalized Hamiltonian.

As usual with approximations involving the time dependence in the functional-integral formulation, the degree of validity of the scheme is difficult to assess. We feel that the most important error is the neglect of the coupling between the static and dynamic spin fluctuations at short-wave lengths, which will appear as a stiffening of the effective Heisenberg exchange against short-wave length fluctuations. In other words, we have not yet succeeded in making an approximation which would be the equivalent of the quantum replacement of S^2 by $S(S+1)$. This error has little or no qualitative importance but may have significance in determining the Curie temperature and constant numerically.

We finally remark on the role of dynamic equations of motion. In the functional integral formulations of field theory, the integrand is $\exp(-i \int L\, d^4x)$ and the stationary condition is just the condition that the field equations are obeyed. Equation (6) here does not look like an equation of motion. However, if one removes its static part, say by taking a time derivative, it may be shown that a dynamic equation results. This equation in the long-wave length limit is just the Landau-Lifshitz equation, as we shall show elsewhere.[10]

Minimization of Eq. (9) (for transverse fluctuations) gives $K\delta\mu=0$; Eq. (17) shows this is just the linearized Landau-Lifshitz equation, the static part $\vec{\mu}=M_s\hat{e}_z$ having been removed. Minimization of Eq. (20) gives a Landau-Lifshitz equation, linearized about a typical static fluctuation. This technique has been exploited in the third paper of this series,[5] and elsewhere.[11]

It would be of interest to cast the functional integral in a form which displayed more clearly the role of the nonlinear Landau-Lifshitz equation, and then to discuss the statistical mechanics in terms of solutions to it. In one dimension at least, the Landau-Lifshitz equations can be completely integrated,[12] and there is a possibility of discussing its statistical mechanics as the statistical mechanics of solitons.

ACKNOWLEDGMENT

This work was supported in part by NSF Grant No. DMR-77-08602.

[1]R. E. Prange and V. Korenman, Phys. Rev. B 19, 4691 (1979) (preceding paper, hereafter referred to as IV). The notation of this paper follows Paper IV.

[2]A. A. Gomes and P. Lederer, J. Phys. (Paris) 38, 231 (1977); D. J. Kim, B. B. Schwartz, and H. C. Praddaude, Phys. Rev. B 7, 205 (1973).

[3]V. Korenman, J. L. Murray, and R. E. Prange, Phys. Rev. B 16, 4032 (1977).

[4]V. Korenman, J. L. Murray, and R. E. Prange, Phys. Rev. B 16, 4048 (1977).

[5]V. Korenman, J. L. Murray, and R. E. Prange, Phys. Rev. B 16, 4058 (1977).

[6]R. E. Prange and V. Korenman, AIP Conf. Proc. 24, 325 (1975).

[7]V. Korenman and R. E. Prange, AIP Conf. Proc. 29, 321 (1978).

[8]V. Korenman and R. E. Prange, Inst. Phys. Conf. Ser. 39, 562 (1978).

[9]R. E. Prange and V. Korenman, Inst. Phys. Conf. Ser. 39, 567 (1978)

[10]R. E. Prange and V. Korenman (unpublished).

[11]F. B. McLean and M. Blume, Phys. Rev. 7, 1149 (1973); V. Korenman and R. E. Prange (unpublished).

[12]H. H. Chen (private communications); L. A. Takhtajan, Phys. Lett. A 64, 235 (1977); V. Korenman and R. E. Prange (unpublished).