Local-band theory of itinerant ferromagnetism. IV. Equivalent Heisenberg model

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The main results of the picture of itinerant ferromagnetism previously presented are obtained by the functional-integral method of Stratonovitch and Hubbard. A new form of this technique is introduced which is spin rotationally invariant in all approximations and which respects the Pauli principle. By a generalization of the saddle-point method, the itinerant-electron thermodynamics is transformed to that of a classical Heisenberg model with Ruderman-Kittel exchange.

I. INTRODUCTION

A new picture of itinerant-electron ferromagnetism, which is particularly apt for the nonweak magnets, iron, nickel, and cobalt, has been recently introduced.[1–7] This "local band" picture is based on the concept of short-range magnetic order (SRMO). The idea is that the crucial magnetic fluctuations, those responsible for the phase transition, are fluctuations in the direction of the local magnetization. It is further argued that this direction changes in space (and time) slowly enough that locally and momentarily, the state of the system is rather like that of the ground state.

In this paper, we present a formal development of these ideas using the Hubbard-Stratonovich functional-integral procedure[8,9] and the one-band short-range exchange model (Hubbard model).

One purpose of this paper is to give comfort to those readers who consider the functional-integral methods to be first principled and microscopic in contrast to the semiphenomenological methods used earlier. Indeed, the method *is* free of some of the ambiguities of the previous approach, and does confirm that the microscopic parameters enter the problem in the way earlier found. This is, of course, not the first attempt to solve the problem of itinerant ferromagnetism by the functional-integral method.[10–14]

The method is a transformation from one set of variables, the single-particle variables, to another, the fluctuating-magnetization variables. The result for the partition function is a functional integral over the magnetization field variables, which can be interpreted as the partition function of a classical spin system, i.e., of a classical Heisenberg model.

The only functional integrals which can be exactly evaluated are of Gaussian form. In consequence, most previous studies of this problem[12–14] have relied on approximation schemes which replace the correct integrand by an approximate one of Gaussian form. We are quite confident, however, that the key to the problem of itinerant-electron ferromagnetism does not lie under the Gaussian street lamp and are resigned to searching in mathematically murkier areas. In compensation it turns out that the physics of the situation becomes remarkably clear.

We adopt the point of view taken by Feynman,[15] Hubbard[9] and others,[16] which is to seek the class of "paths", i.e., field configurations, which maximize the integrand. This "saddle-point" approximation is known to yield a mean-field theory. Small fluctuations about it give the corresponding random-phase approximation (RPA).

There are also many other field configurations, typically varying violently and discontinuously in space and time, which make a contribution. The number of such paths is so large that their net contribution is finite, even though individually such paths are of no importance.

The aim of our approach is to provide a formulation in which the neighborhood of the saddle-point paths dominates the physical description of the problem. The remaining paths then contribute a renormalization of the saddle-point result.

It is not obvious that this program can work. Indeed, in the formulations heretofore used, it is quite apparent that the saddle-point paths are an insufficient starting point. However, we have devised a formulation which is at least free of the known diseases[17,18] which have afflicted the others. The success of Fermi-liquid phenomonology gives hope that the program is a feasible one.

Our main result is an expression for the partition function which in spite of its apparent intractability has several appealing features. It is the partition function of a classical spin system which has interactions between nearest-neighbor *pairs* of spins, i.e., among a total of four spins. The self-interaction of a pair is ferromagnetic, while the interaction between different pairs tends to be antiferromagnetic. The net result is on average ferromagnetic. This provides a mechanism by which SRMO can exist above the

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transition temperature T_C. The temperature at which SRMO disappears is given by the nearest-neighbor interaction. T_C, on the other hand, is given in mean-field theory (which is an overestimate especially in this case) by the average interaction. In fact, further approximation reduces the result to a classical Heisenberg model with Ruderman-Kittel-Kasaya-Yosida (RKKY) interaction in *ferromagnetic* bands. In lowest approximation this model gives quite reasonable results for the transition temperature and susceptibility of iron and nickel.

In Sec. II we formulate the theory. This is followed by a description of the approximation scheme and the results. In this paper, we consider only the first nontrivial approximation (which is a static approximation). We shall deal with dynamic and quantum effects later. We shall remark on the technical problems of this method elsewhere.

II. FUNCTIONAL-INTEGRAL FORMULATION

The Hubbard-Stratonovitch transformation relies on the identity

$$e^{a^2} = \pi^{-1/2} \int dx\, e^{-x^2+2ax} .$$

To utilize this formula, one must rewrite the interaction term in the Hamiltonian as a sum of squares. That this can be done in an infinite number of ways is the source of many difficulties.

Our point of view is that a useful expression must be one in which the important paths in function space are those in some sense near the saddle-point path. Thus, the effects of spin waves and spin fluctuations have to be included in this class of paths. For this to be the case, it is essential that this class of paths maintains spin rotational invariance.

Another requirement, considered to be essential,[17] but as a rule ignored in work on ferromagnetism, is that the approximations used must respect the Pauli principle. In fact, heretofore no approximation scheme has been employed which satisfies both of these criteria.[19]

We illustrate our approach for the case of the one band Hubbard model, with Hamiltonian

$$H = \sum T_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} + U \sum n_{i+} n_{i-}$$

$$\equiv H_0 + U \sum n_{i+} n_{i-} .$$

The simplest method previously employed which does not do violence to the Pauli principle is the so-called two-field method, in which the interaction term is written,

$$n_{i+} n_{i-} = \tfrac{1}{4} (n_i)^2 - (M_i^z)^2 ,$$

where

$$\vec{\mathrm{M}}_i = \tfrac{1}{2} \sum_{ss'} c_{is}^{\dagger} \vec{\sigma}_{ss'} c_{is'} ,$$

$\vec{\sigma}$ being the Pauli matrices, and $n_i = n_{i+} + n_{i-}$. Two fields are introduced, one for n_i, one for M_i^z, corresponding to density and magnetization density fluctuations, respectively. If corresponding approximations are made on the two fields, the Pauli principle is preserved.[18]

However, magnetization fluctuations which correspond physically to long-wavelength deviations of spin direction are not readily described by the two fields. To achieve a more convenient description, $(M_i^z)^2$ must be replaced by a squared operator which is rotationally invariant. Usually[12,13,20] this is done by the identity $(M_i^z)^2 = \frac{1}{3} \vec{\mathrm{M}}_i \cdot \vec{\mathrm{M}}_i$, followed by the introduction of a vector magnetization fluctuation field. Unfortunately, approximations again violate the Pauli principle. This can be checked by noting that the saddle-point method does not give the lowest order in U correctly.

To avoid these problems, we use the identity (valid for spin $\frac{1}{2}$)

$$(M_i^z)^2 = (\vec{\mathrm{M}}_i \cdot \hat{\mu}_i)^2 ,$$

where $\hat{\mu}_i$ is an arbitrary unit vector. The two-field method is then employed and the directions of $\hat{\mu}_i$ are averaged over to restore rotational invariance. A more detailed analysis of this method will be presented elsewhere. We note here that the saddle-point approximation yields the standard Hartree-Fock, and Gaussian fluctuations about that solution gives the RPA corrections to the Hartree-Fock, including the standard spin-wave results. These approximations are known to be consistent with the Pauli principle and, after Fermi-liquid renormalizations, are believed to be numerically quite good at low temperaure.

The expression for the partition function, using this procedure, is

$$Z = \mathrm{Tr} e^{-\beta H} = \int \mathfrak{D}x \mathfrak{D}\mu \exp\left[-U \int_0^{\beta} d\tau \sum_i (\tfrac{1}{4} x^2 + \vec{\mu}^2) \right] \mathrm{Tr} e^{-\beta H_0} \left[\exp\left[\int_0^{\beta} d\tau \sum_i (2U \vec{\mathrm{M}} \cdot \vec{\mu} + i \tfrac{1}{2} U x n) \right] \right]_+ ; \qquad (1)$$

x, μ, M, n are functions of i, τ. The "time" dependence is given by $M_i(\tau) = e^{\tau H_0} M_i e^{-\tau H_0}$. The functional integrals are defined by

$$\mathfrak{D}x \equiv C \prod_{i\tau} dx_i(\tau) , \tag{2}$$

$$\mathfrak{D}\mu = C \prod_i d^2\hat{\mu}_i \prod_\tau d\mu_i(\tau) , \tag{3}$$

with C a constant, irrelevant for our purposes. The time ordering, symbolized by $(\,)_+$ has been introduced for the usual reasons. The price paid for the proper treatment of the Pauli principle is Eq. (3) which defines a functional integral of a somewhat inconvenient and unfamiliar form. It is equally exact, and more conducive to approximations which take into account dynamic and quantum effects to average $\hat{\mu}_i$ over directions independently for each τ, but in this paper we shall be concerned with the static approximation, which already gives the major results.

The saddle-point paths are the ones which minimize

$$\mathfrak{F}(\vec{\mu},x) \equiv T \int_0^\beta d\tau \sum_i (\tfrac{1}{4} U x^2 + U \vec{\mu}^2) + \mathfrak{F}_0 , \tag{4}$$

where

$$\mathfrak{F}_0 = -T \ln \mathrm{Tr} e^{-\beta H_0} \left[\exp\left(\int_0^\beta d\tau \sum_i (2U\vec{\mathrm{M}} \cdot \vec{\mu} + i\tfrac{1}{2} Unx) \right) \right]_+ . \tag{5}$$

The paths satisfy

$$x_j(\tau) = i \langle n_j(\tau) \rangle_{x\mu} , \tag{6}$$

$$\vec{\mu}_j(\tau) = \langle \vec{\mathrm{M}}_j(\tau) \rangle_{x\mu} , \tag{7}$$

where for arbitrary operators $O_j(\tau)$ we define

$$\langle O_j(\tau) \rangle_{x\mu} = e^{\beta F_0} \mathrm{Tr} e^{-\beta H_0} \left[O_j \exp\left(\int_0^\beta d\tau \sum_i (2U\vec{\mathrm{M}} \cdot \vec{\mu} + i\tfrac{1}{2} Unx) \right) \right]_+ . \tag{8}$$

Since we are interested in ferromagnetism, we assume that the parameters are such that ferromagnetic solutions of Eqs. (6) and (7) are the most stable. Then

$$x_j(\tau) = i\bar{n} , \qquad \vec{\mu}_i(\tau) = M_s \hat{e}_z \tag{9}$$

are the saddle-point paths, constant in space and time, and $\mathfrak{F}(M_s\hat{e}_z, i\bar{n})$ is precisely the Stoner free energy, M_s is the Stoner magnetization at temperature $T = \beta^{-1}$, and $\bar{n}$ is the density.

III. FLUCTUATIONS

We have thus found that the best single path already gives the Stoner approximation; a satisfying result since it has long been apparent that the band theory is on the right track (at any rate for low temperatures). At higher temperatures, the Stoner theory fails. From the vantage point of the functional-integral formulation that simply means that paths exhibiting significant global deviation from the path of Eqs. (6) and (7) become important. In principle there is a competition: Is the system stiffer against temperature effects on the dominant path or against temperature effects inducing fluctuations away from the dominant path? In practice, a ferromagnet with spin rotational symmetry is intrinsically weak against fluctuations in the direction of the magnetization. In fact, it is clear that any path with $|\vec{\mu}_i(\tau)| \approx M_s$, and which is slowly varying in i and τ, will give a free energy $\mathfrak{F}$ close to the minimum, and therefore will contribute heavily to the functional integral.

Our approximation consists in keeping just such paths. To the extent that such slowly varying field configurations dominate the integral, there will be a form of short-range magnetic order (SRMO). We have argued previously that there is considerable experimental evidence in favor of the existence of SRMO, even to temperatures well above T_C.[1-4] We shall also see that the present formulation provides a way to study the reasons for its existence.

The next step is to evaluate the expression (5) for the paths slowly varying in direction; $|\vec{\mu}_i(\tau)| = M_s$, $x_j(\tau) = i\bar{n}$, $|\hat{\mu}_i - \hat{\mu}_j|$ small (i,j neighbors). Fluctuations in the magnitude $\mu_i(\tau)$ and $x_j(\tau)$ can be reasonably handled by a Gaussian expansion, and give RPA correlation corrections to the free energy. These contributions will be discussed elsewhere, but they do not significantly affect the phase transition, except possibly in the case of truly weak itinerant electron ferromagnetism.

The free energy may be evaluated by the technique introduced in Ref 3, of utilizing a locally rotated spin coordinate system (LRSCS). The transformation to this system is of course canonical. The expression for $\mathfrak{F}_0$, Eq. (5), becomes, in the LRSCS,

$$\mathfrak{F}_0 = -T \ln \mathrm{Tr} e^{-\beta \tilde{H}} , \tag{10}$$

with

$$\tilde{H} = H_0 + U \sum_{i,\sigma} \bar{n}_\sigma c_i^\dagger{}_{-\sigma} c_{i\,-\sigma} + H_1 + H_2 . \tag{11}$$

The terms perturbing the Hartree-Fock single-particle Hamiltonian are

$$H_1 = -i \sum_{k,l} T_{kl} (c_{ks}^\dagger \sigma_{ss'}^+ c_{ls'} a_{kl}{}^* + c_{ks}^\dagger \sigma_{ss'}^- c_{ls'} a_{kl} - c_{ks}^\dagger \sigma_{ss'}^z c_{ls'} g_{kl}) , \tag{12}$$

$$H_2 = \sum_{kl,s} T_{kl} d_{kl} c_{ks}^\dagger c_{ls} . \tag{13}$$

The central quantity a_{kl} is

$$a_{kl} = (\sin \tfrac{1}{2}\phi_{kl} \sin \bar{\theta}_{kl} - i \cos \tfrac{1}{2}\phi_{kl} \sin \tfrac{1}{2}\theta_{kl}) e^{-i\bar{b}_{kl}} \approx \tfrac{1}{2}(\phi_{kl} \sin \bar{\theta}_{kl} - i\theta_{kl}) e^{-i\bar{b}_{kl}} . \tag{14}$$

Here the direction $\hat{\mu}_i$ is labelled by polar angles θ_i, ϕ_i, and b_i is the (arbitrary) third Euler angle necessary to specify the local coordinate system. Also, $\theta_{ij} = \theta_i - \theta_j$, etc., $\bar{\theta}_{ij} = \frac{1}{2}(\theta_i + \theta_j)$. The other quantities are

$$g_{kl} = \sin \tfrac{1}{2} b_{kl} \cos \tfrac{1}{2}\phi_{kl} \cos \tfrac{1}{2}\theta_{kl} + \cos \tfrac{1}{2} b_{kl} \sin \tfrac{1}{2} \phi_{kl} \cos \bar{\theta}_{kl} \approx \tfrac{1}{2}(b_{kl} + \phi_{kl} \cos \bar{\theta}_{kl}) \tag{15}$$

and

$$d_{kl} = \cos \tfrac{1}{2} b_{kl} \cos \tfrac{1}{2}\phi_{kl} \cos \tfrac{1}{2}\theta_{kl} - \sin \tfrac{1}{2} b_{kl} \sin \tfrac{1}{2}\phi_{kl} \cos \bar{\theta}_{kl} - 1 \approx -\tfrac{1}{8}(b_{kl}^2 + \phi_{kl}^2 + \theta_{kl}^2 + 2\phi_{kl} b_{kl} \cos \bar{\theta}_{kl}) = -\tfrac{1}{2}(|a_{kl}|^2 + |g_{kl}|^2) . \tag{16}$$

It is straightforward to evaluate the contributions of H_1 and H_2 in perturbation theory. The result, to second order, is

$$\mathfrak{F} = \mathfrak{F}_{\mathrm{Stoner}} + \mathfrak{F}_1 , \tag{17}$$

where

$$\mathfrak{F}_1 = \mathfrak{F}_\perp + \mathfrak{F}_z , \tag{18}$$

with

$$\mathfrak{F}_\perp = -\tfrac{1}{2} \sum_{kl} |a_{kl}|^2 T_{kl} f_{kl} - \tfrac{1}{2} \sum_{klrs\,\sigma} a_{kl}^*\, a_{rs}\, T_{kl} T_{rs} f_{ks,\sigma;rl\,-\sigma} \equiv \mathfrak{F}_\perp' + \mathfrak{F}_\perp'' \tag{19}$$

and

$$\mathfrak{F}_z = -\tfrac{1}{2} \sum_{kl} g_{kl}^2 T_{kl} f_{kl} - \tfrac{1}{2} \sum_{klrs,\,\sigma} g_{kl} g_{rs} T_{kl} T_{rs} f_{ks\,\sigma,rl\,\sigma} . \tag{20}$$

Here we have

$$f_{kl} = \frac{1}{N} \sum_{q,\,\sigma} e^{i\vec{q}\cdot(\vec{r}_k - \vec{r}_l)} f(E_{q\sigma}) , \tag{21}$$

$$f_{ks\,\sigma,rl\,\sigma'} = \frac{1}{N^2} \sum_q \sum_p e^{i\vec{q}\cdot(\vec{r}_s - \vec{r}_k)} e^{i\vec{p}\cdot(\vec{r}_l - \vec{r}_r)} \times \frac{f(E_{q\sigma}) - f(E_{p\sigma'})}{E_{q\sigma} - E_{p\sigma'}} . \tag{22}$$

The quantities $f(E)$ are the Fermi factors, $E_{q\sigma}$ the single-electron eigenenergies of H_0, and N is the total number of sites.

It remains to perform the functional integral over directions of the field $\hat{\mu}_i$.

IV. FUNCTIONAL INTEGRATION

Functional integrals of the type here encountered,

$$Z = e^{-\beta \mathfrak{F}_{\text{Stoner}}} \int \prod_i d^2\mu_i \, e^{-\beta \mathfrak{F}_1} , \tag{23}$$

have been studied in relation to the classical Heisenberg model. In our previous work,[3] we arrived at an expression of this form in which the long-wavelength limit of the expression for $\mathfrak{F}_1$ appeared. The limit is obtained here by the replacement $a_{kl} = \vec{a} \cdot (\vec{r}_k - \vec{r}_l)$, where $\vec{a}$ is the continuous limit form, used in Ref. 3, i.e.,

$$\vec{a} = \tfrac{1}{2} e^{-ib} (\sin\theta \, \nabla\phi - i \nabla\theta) .$$

In this limit the terms in g cancel, and $\mathfrak{F}_\perp$ is

$$\mathfrak{F}_\perp \approx \int A (\nabla \hat{M})^2 \, d^3x ,$$

with

$$A = \frac{1}{24} \frac{1}{V} \sum_{k,\sigma} \left[\nabla_k^2 E_{k\sigma} f(E_{k\sigma}) - 2(\nabla_k E_{k\sigma})^2 \frac{\sigma f(E_{k,\sigma})}{\Delta} \right] . \tag{24}$$

We have introduced the exchange splitting $\Delta = 2M_s U$. This is just the standard expression for the Bloch wall stiffness in the RPA.[21] The first term in Eq. (24) is $\mathfrak{F}_\perp'$, the second is $\mathfrak{F}_\perp''$.

The renormalization-group study of integrals of the form (23) shows that the short-wavelength cutoff is of numerical importance. In Ref. 3, it was assumed that this cutoff was the zone-boundary wave vector, and at first sight that appears to be plausible here. The explicit expression for $\mathfrak{F}_1$ provides a basis for numerical study of this question.

We next study the instructive case of one electron per atom, for very large Δ. This is expected to describe a localized magnet. Then $f_{kl} = \delta_{kl}$ and the first, positive, term $\mathfrak{F}_\perp'$ vanishes. It also follows that

$$f_{ks\sigma;rl-\sigma} \cong -(1/\Delta) \delta_{sk} \delta_{rl} ,$$

so that, in this limit

$$\mathfrak{F}_\perp = -\sum_{kl} |a_{kl}|^2 T_{kl}^2 / \Delta \tag{25}$$

The terms in g again vanish. As discussed in Ref. 3, they can only be important at short wavelength and we continue to neglect them here.

As expected, the one electron per site, large-U limit, is unstable to the ferromagnetic solution, and favors a staggered, antiferromagnetic field configuration.

In the itinerant case, f_{kl} has a range determined by the Fermi-surface structure, i.e., typically short, but extending certainly to nearest neighbors. If ferromagnetism is stable, i.e., if $A > 0$, then $\mathfrak{F}_\perp'$ will be positive, as can be seen from Eq. (24), and sufficiently so to more than compensate for the essentially negative contribution of $\mathfrak{F}_\perp''$.

V. HEISENBERG MODEL

Aside from the static approximation, the main simplification we have so far made is that $|\hat{\mu}_i - \hat{\mu}_j| \ll 1$ when i,j are neighbors. If we are willing to use the approximation that all four $\hat{\mu}_i$ appearing in a typical term in Eq. (19) are nearly in the same direction, further significant simplifications can be made. In this case we have

$$a_{kl}^* \, a_{rs} = \tfrac{1}{4} (\hat{\mu}_k - \hat{\mu}_l) \cdot (\hat{\mu}_r - \hat{\mu}_s) . \tag{26}$$

It is thus apparent that, with the continued neglect of $\mathfrak{F}_z, \mathfrak{F}_\perp$ is of the form

$$\mathfrak{F}_\perp = -\sum_{kl} J_{kl} \hat{\mu}_k \cdot \hat{\mu}_l . \tag{27}$$

The above expression is of interest only for $k \neq l$. Many simplifications are possible in the second term of Eq. (19) because Eq. (26) can be broken up into terms depending on only two subscripts at a time. There results

$$J_{kl} = \frac{1}{4} \frac{\Delta^2}{N} \sum_{p,q,\sigma} \frac{f(E_{p\sigma})}{E_{p+q,-\sigma} - E_{p\sigma}} e^{i\vec{q}\cdot\vec{r}_{kl}} . \tag{28}$$

This expression is just of the familiar Ruderman-Kittel type. It is to be emphasized, however, that the bands involved are ferromagnetic and that the usual oscillations of wave number $q = 2k_F$ are replaced by ones which span Fermi surfaces of opposite spin. It thus differs from the interaction advocated by Stearns.[22]

The Fourier transform of J_{kl} is of interest. In particular

$$L(q) = \sum_l J_{kl} (1 - e^{i\vec{q}\cdot\vec{r}_{kl}})$$

is

$$L(q) = \tfrac{1}{4} \Delta \left[M_s - \frac{\Delta}{N} \sum_{p\sigma} \frac{f(E_{p\sigma})}{E_{p+q,-\sigma} - E_{p\sigma}} \right] . \tag{29}$$

At small q,

$$L(q) \sim (V/N) A q^2 , \tag{30}$$

where A is given by Eq. (24). Thus the long-wavelength limit of this Heisenberg model reduces appropriately to the continuum limit. The mean-field value of the Curie temperature T_C is given by

$$kT_C^{\mathrm{mf}} = \frac{2}{3}\frac{1}{N}\sum_q L(q) \ . \tag{31}$$

The Green's function scheme[23] gives

$$kT_C^{\mathrm{Gf}} = \frac{2}{3}\left[\frac{1}{N}\sum_q \frac{1}{L(q)}\right]^{-1} \ . \tag{32}$$

If $L(q)$ is replaced by the estimate (30) throughout the entire Brillouin zone, and this is estimated by a sphere, Eq. (32) yields

$$kT_C^{\mathrm{Gf}} \approx \frac{4\pi^2}{3}\frac{A}{G} \ , \tag{33}$$

where G is the Brillouin-zone radius. This is a factor $\frac{1}{3}\pi$ greater than the estimate obtained in Ref. 3, and yields estimates of T_C, 35% too high for iron and cobalt, 10% too high for nickel.

The susceptibility may be calculated in the mean-field theory and gives (for $T > T_C$)

$$\chi = \tfrac{1}{3}M_s^2/(T - T_C) \ . \tag{34}$$

This estimate of the Curie constant $\frac{1}{3}M_s^2$ is considerably smaller than that found by experiment for iron and nickel. However, two effects have been left out. The first is the quantum effect, which is neglected in the static approximation, and which will go in the direction of replacing M_s^2 by $M_s(M_s+1)$. The second is the effect of short-range order, which should also increase the Curie constant, but decrease the quantum effect.

The calculation of the susceptibility is along the usual lines. It should be shown, however, that the magnetic field enters in the ordinary way.

To see this, one may note that an external field $\vec{\mathrm{H}}$, changes F_0, to first order in $\vec{\mathrm{H}}$, by an additional term, $\int d\tau \langle \vec{\mathrm{M}}_i(\tau)\rangle_{x\mu} \cdot \vec{\mathrm{H}}$ which is approximately $\beta M_s \hat{\mu}_i \cdot \vec{\mathrm{H}}$. Corrections to this are of order a_{kl}, and give a correction to the Curie constant of second order.

We mention for the sake of comparison some results of Moriya and Takahashi,[14] using the Gaussian method designated earlier. They obtain for T_C an expression

$$T_C = \frac{2}{3}\eta\left[\frac{1}{N}\sum \tilde{L}(q)^{-1}\right]^{-1} \ ,$$

where η is a parameter between $\frac{3}{5}$ and one, and $\tilde{L}(q) = 2(\frac{2}{3}U)^2 M_s^2(\chi_0 - \chi_q)$. The quantity χ_q is to be chosen phenomenologically and is a paramagnetic, noninteracting, susceptibility. (Their U' is our U.) In order that $\tilde{L}(q) = L(q)$, one must choose

$$\chi_q = \tfrac{1}{2}(\tfrac{3}{2})^2 \sum_{p,\sigma} f(E_{p\sigma})/(E_{p+q,-\sigma} - E_{p\sigma})$$

$$= \tfrac{1}{2}(\tfrac{3}{2})^2 \chi_q^{+-} \ ,$$

where χ_q^{+-} is the susceptibility of free electrons in an exchange field strong enough to give the splitting Δ. The factor $(\frac{3}{2})^2$ is a result of their choice of a Stratonovich transformation which does not respect the Pauli principle in low approximation. The factor 2 may be related to the fact that their χ_q represents a *longitudinal, paramagnetic* susceptibility, not the *transverse, ferromagnetic* susceptibility of our formula.

For the Curie constant, they obtain $\frac{1}{3}\eta M_s^2$, close to our result.

VI. DISCUSSION

It has been the goal of theorists for many years to relate the thermodynamics of itinerant magnets to that of a Heisenberg model. A Heisenberg model can indeed come close to fitting the "magnetic part" of the thermodynamic data. To do this, it is known that the effective exchange must be of long range and oscillatory in sign, as is the RKKY.

Earlier studies using the functional-integral method include those of Cyrot[11] and of Evanson, Wang, and Schrieffer.[10] The spirit of these approaches was rather different than ours, (and in addition spin rotational invariance was lost at an early stage). They concentrated on special configurations in which the field μ_i was nonvanishing only at one or two sites. Their approach thus sought to build up the magnet as a collection of local moments of the Friedel-Anderson variety.

Great difficulties have been encountered in previous efforts which sought an appropriate transformation at the operator level. Herring[21] has reviewed and criticized some of these efforts.

The result which comes closest to ours is that of Capellman.[24] By a Hartree-Fock method he obtained an energy precisely equal to our Eq. (27), with the exception that the Fermi functions in Eq. (28) were taken at zero temperature and $\hat{\mu}_i$ was replaced by $\langle \vec{\mathrm{M}}_i\rangle/M_s$ [as is true if Eq. (26) holds]. The meaning of the result was unclear, however, and Capellman replaced $\langle \vec{\mathrm{M}}_i\rangle$ by a spin operator $\vec{\mathrm{S}}_i$, a step both unjustified and unjustifiable, since $2M_s$ is not integral. This illustrates the central difficulty in the program of finding an effective Heisenberg Hamiltonian at the operator level.

As a matter of principle, the density fluctuations δx and $\delta|\mu|$ may always be integrated out of Eq. (1) to yield an expression for Z of the form (23). Thus a classical Heisenberg-like free energy can always be found. The question is whether the approximation

we have made of ignoring the coupling between the density and the orientation fluctuations is adequate as a first approximation. In a similar way, dynamical effects corresponding to having spin *operators* will be sought here as corrections to the static approximation. These corrections will be discussed in a subsequent paper. Thus, this approach renounces the attempt to express the itinerant-electron Hamiltonian as a Heisenberg Hamiltonian in favor of the less ambitious plan of expressing the free energy as arising from a classical Heisenberg Hamiltonian with corrections for density and dynamic fluctuations.

Before the above results can be seriously compared with experiment a number of additional problems must be resolved.

First, one must generalize the procedure to a more realistic Hamiltonian, say that of the combined interpolation scheme. Such a choice assumes that Fermi liquid and other correlation corrections can be accounted for by the effective parameters of the Hamiltonian.

Corrections to the short-range-order approximation must also be investigated. Actually, the smallness of $|\hat{\mu}_i - \hat{\mu}_j|$ is a sufficient, not a necessary criterion. What is necessary is that the perturbation treatment of H_1 and H_2 be adequate. We have estimated this to be the case, at least for nickel. In making the replacement of Eq. (26), however, the small angle approximation has been used, even for further than nearest neighbors. Since we found in Ref. 3, that a typical angle between nearest-neighbor $\hat{\mu}_i$ vectors is 45°, at and above T_C in Fe and Ni, this will be a source of error.

There is finally the question of the prediction of short-range magnetic order. This question hinges on the spatial structure of the function $f_{ks\sigma,rl-\sigma}$. Roughly speaking, the situation is promising in that $F_\perp'$ is of the shortest possible range and is ferromagnetic, whereas $F_\perp''$ is of longer range and is antiferromagnetic. Thus $|a_{kl}|^2$ is inhibited from becoming too large, while there is a correlation between nearby pairs which favors an accumulation of spin deviations on a larger scale. The details of this depend on the band structure, and may well be more sensitive to the approximations previously mentioned than the estimate of the Curie temperature.

In spite of these questions, it is satisfying that this crude first treatment is in order-of-magnitude agreement with experiment and that the expected corrections go in the right direction.

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[1]R. E. Prange and V. Korenman, AIP Conf. Proc. 24, 325 (1975).
[2]V. Korenman, J. L. Murray, and R. E. Prange, AIP Conf. Proc. 29, 321 (1978).
[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]V. Korenman and R. E. Prange, Inst. Phys. Conf. Ser. 39, 562 (1978).
[7]R. E. Prange and V. Korenman, Inst. Phys. Conf. Ser. 39, 567 (1978).
[8]R. L. Stratonovich, Sov. Phys. Dok. 2, 416 (1958).
[9]J. Hubbard, Phys. Rev. Lett. 3, 77 (1959).
[10]W. E. Evanson, S. Q. Wang, and J. R. Schrieffer, J. Appl. Phys. 41, 1199 (1970).
[11]M. Cyrot, Phys. Rev. Lett. 25, 871 (1970).
[12]K. K. Murata and S. Doniach, Phys. Rev. Lett. 29, 285 (1972).
[13]J. A. Hertz and M. A. Klenin, Physica (Utrecht) B 91, 49 (1977).
[14]T. Moriya and Y. Takahashi J. Phys. Soc. Jpn. 45, 397 (1978).
[15]R. P. Feynman and A. R. Hibbs, *Quantum Mechanics and Path Integrals* (McGraw-Hill, New York, 1965).
[16]D. R. Hamann, Phys. Rev. B 2, 1373 (1970).
[17]H. Keiter, Phys. Rev. B 2, 3777 (1970).
[18]G. Morandi, E. Galliani D'Agliano, F. Napoli, and G. F. Ratto, Adv. Phys. 23, 867 (1974).
[19]Recently we received a report by Hubbard [J. Hubbard, Phys. Rev. 19, 2626 (1979).] which independently introduces precisely the formulation we advocate.
[20]A. A. Gomes and P. Lederer, J. Phys. (Paris) 38, 231 (1977).
[21]C. Herring, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic, New York, 1966), Vol. IV.
[22]M. B. Stearns, Phys. Rev. 8, 4383 (1973).
[23]R. Tahir-Kheli and G. ter Haar, Phys. Rev. 127, 88 (1962).
[24]H. Capellman, J. Phys. F 4, 1466 (1974).