Fluctuations in itinerant-electron paramagnets*

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We use a functional-integral approach to study spin fluctuations in strongly paramagnetic systems. Our basic approximation is to replace the exact free energy functional by a variationally chosen quadratic form in the fluctuating (paramagnon) fields. This leads to a susceptibility χ of the form $\bar{\varphi}(1-U\bar{\varphi})^{-1}$, where $\bar{\varphi}$ is an averaged electron-hole bubble in the presence of a space- and time-varying random external potential. The random potential has Gaussian statistics, and its covariance matrix is determined self-consistently. In another language, $\bar{\varphi}$ is a polarization bubble dressed with paramagnons in all orders of perturbation theory. When the fluctuations are small and effectively only one paramagnon dresses the bubble at a time, we recover the results of Murata and Doniach and of Moriya and Kawabata. For intermediate coupling and at temperatures well above the spin-fluctuation temperature, we find that $\bar{\varphi}$ is given approximately by an average of the corresponding random-phase-approximation (RPA) bubble over a distribution of Fermi levels of width $\approx(UkT)^{1/2}$, producing approximate Curie-Weiss behavior in χ. These conclusions are supported by calculations of χ for two model systems—one, for simplicity, with a Gaussian density of states, and the other with the density of states of paramagnetic Ni.

I. INTRODUCTION

The phenomenological Stoner theory of itinerant-electron magnetism has provided a useful and generally very successful basis for understanding the electronic and magnetic properties of many metals and alloys since it was first proposed. It seeks to explain these properties in terms of a self-consistent theoretical account of the interactions between up- and down-spin electronic quasiparticles. In the simplest interpretation, these quasiparticles are the bare Bloch electrons, and hence one may use Stoner theory to predict magnetic properties on the basis of a knowledge of the one-electron band structure.

Less naively, however, the Stoner quasiparticles are complicated many-body excitations like those of Landau-Fermi-liquid theory, having no direct relation to single-particle Bloch states. A central problem, then, is this connection between quasiparticle and bare-particle states. This relationship has been thoroughly investigated for normal Fermi liquids at very low temperatures, but its general structure, particularly in magnetic systems, remains only partly understood. The importance of this problem is underlined by several recent photoemission experiments, in both ferromagnetic and paramagnetic states of transition-metal ferromagnets.[1] It has proved difficult to reconcile the observed structure of the photoelectron spectra with a Stoner-theoretical picture of band magnetism if the Stoner quasiparticles are identified directly with the Bloch states of the band structure.

The most obvious shortcoming of Stoner theory is its neglect of correlation, which renders it incapable of describing the incipient local-moment formation which is so characteristic of actual transition-metal ferromagnets. Nickel is commonly held to have the least-localized spins of the pure transition-metal ferromagnetic series, yet we know of no Stoner-theoretical account, based on an even remotely realistic band structural model, of the temperature dependence of its susceptibility. Even the very weak itinerant ferromagnets Sc_3In and $ZrZn_2$ have Curie-Weiss susceptibilities which cannot be explained in Stoner theory.[2]

These, then, are the sorts of problems we want to study here—the collective spin fluctuations, the structure of the single-particle excitations, and the interactions between them. We will develop a quite general approach to these problems, focusing particularly on the spin fluctuations. As a starting point we use the Hubbard Hamiltonian.[3] For a single nondegenerate band the interaction takes the simple local form

$$H' = U\sum_i n_{i\uparrow} n_{i\downarrow} \tag{1}$$

in Wannier representation. Although interatomic exchange is no doubt present in real transition metals, and d-band degeneracy, together with interband interactions, may be essential to a quantitative understanding of their magnetic properties, we ignore these complications. The simple one-band Hubbard model contains sufficient physics to

give a qualitative account of the properties we want to understand.

A random-phase-approximation (RPA) theory of the Hubbard Hamiltonian predicts a ferromagnetic instability when $UN(E_F) > 1$, but correlation effects of the sort mentioned above make the ferromagnetic state more difficult to realize. In this paper we want to focus our attention on the intermediate-coupling region where $UN(E_F) \gtrsim 1$, but the system is still paramagnetic.

At finite temperature, the RPA criterion for ferromagnetism is the same as that above, except that $N(E_F)$ is replaced by an average of $N(E)$ over an energy region of approximate width kT around E_F. The principal qualitative result of the theory we describe below is that incipient local-moment formation leads to a fluctuating local Zeeman energy whose rms value is, in the intermediate coupling region, of the order of $(UkT)^{1/2}$. Consequently, the effective density of states in the criterion for ferromagnetism, or for determining the susceptibility in general, is an average of $N(E)$ over an energy range of this much larger width. The temperature dependence of physical quantities will be qualitatively affected by this fact.

Some progress in the many-body theory of metallic magnetism has been gained through functional integral techniques, and we adopt this approach here.[4,5] We use the so-called two-variable linearization scheme, and since we believe that for systems of interest here, density fluctuations are unimportant, we approximate the functional integral over the spin-independent field by its stationary value.[5,6] The remaining functional integral, over the spin-dependent external field, may be written in the form

$$Z = \int \prod_i \mathfrak{D}\xi_i(\tau) \exp\left(-\sum_i \frac{1}{\beta}\int_0^\beta \xi_i^2(\tau)\,d\tau + \sum_\sigma \mathrm{Tr}\ln(1 - V^\sigma G^0)\right), \quad (2)$$

where the matrix V^σ, with elements $V^\sigma_{ij}(\tau,\tau') = \sigma c \xi_i(\tau)\,\delta_{ij}\,\delta(\tau-\tau')$, represents the fictitious potential in which electrons of spin-σ propagate, and G^0 is the Bloch propagator in the absence of this potential. In momentum space

$$G^0(k,k';\, i\omega_n, i\omega_{n'}) = G^0(k, i\omega_n)\,\delta_{kk'}\,\delta_{nn'} = (i\omega_n - \epsilon_k)^{-1}\,\delta_{kk'}\,\delta_{nn'}\,, \quad (3)$$

the $i\omega_n$ are the fermion Matsubara frequencies $2\pi i(n+1)/\beta$.

It is also possible to express the single-particle Green's function for the interacting system in terms of a functional integral.[5] If we dispose of the spin-independent field in the same way that we did above, we have

$$G_{ij}(t,t') = -\langle T(c_i(t)\,c_j^\dagger(t'))\rangle = -\frac{1}{Z}\int \prod_i \mathfrak{D}\xi_i(\tau) \exp\left(-\sum_i \frac{1}{\beta}\int_0^\beta \xi_i^2(\tau)\,d\tau\right) \mathrm{Tr}\,T\left[c_i(t)\,c_j^\dagger(t')\exp\left(-\sum_{i\sigma}\int_0^\beta c\sigma\,\xi_i(\tau) \times n_{i\sigma}(\tau)\,d\tau\right)\right] \equiv \int \prod_i \mathfrak{D}\xi_i(\tau)\, W\{\xi\} G_{ij}(t,t',\{\xi\}) \Big/ \int \prod_i \mathfrak{D}\xi_i(\tau)\, W\{\xi\} \quad (4)$$

where

$$G_{ij}(t,t',\{\xi\}) = \mathrm{Tr}\,T\left[c_i(t)\,c_j(t')\exp\left(-\sum_{i\sigma}\int_0^\beta c\sigma\,\xi_i(\tau)\,n_{i\sigma}(\tau)\,d\tau\right)\right] \Big/ \mathrm{Tr}\,T\exp\left(-\sum_{i\sigma}\int_0^\beta c\sigma\,\xi_i(\tau)\,n_{i\sigma}(\tau)\,d\tau\right) \quad (5)$$

is the Green's function in the presence of the fluctuating field $\sigma c\xi_i(\tau)$, the distribution of whose values is described by a probability density function $W\{\xi\}$, which is the integrand of Eq. (2). [Here and henceforth the notation $y\{x\}$ indicates that y is a functional of a set of functions $x_i(\tau)$.] That is, the full Green's function is the averaged propagator for an electron propagating in the random external potential whose distribution is $W\{\xi\}$.

A similar expression applies for other response functions, including the spin susceptibility, but we can also write χ in terms of the mean square fluctuations of the auxiliary field ξ (Ref. 4):

$$\chi(q, i\nu_m) = (2/U)(\langle\,|\,\xi_{qm}\,|^2\rangle - \tfrac{1}{2})\,, \quad (6)$$

where

$$\langle\,|\,\xi_{qm}\,|^2\rangle = \frac{1}{Z}\int \prod_{q',m'}\left(\frac{d\xi_{q'm'}}{\sqrt{\pi}}\right)\,|\,\xi_{qm}\,|^2 \times \exp\left(-\sum_{q'm'}\,|\,\xi_{q'm'}\,|^2 + \sum_\sigma \mathrm{Tr}\ln(1 - V^\sigma G^0)\right) \quad (7)$$

and ξ_{qm} are the Fourier components of $\xi_i(\tau)$. Details of all the above algebra can be found in Refs. 4 and 5.

Equation (2) is our starting point in this paper. For the Anderson model, where the interaction acts at only one site (and hence there are no site or momentum indices on any quantities), Wang *et al.*[4] were able to show how the local moment (Curie

susceptibility) emerged as a consequence of the form of Eq. (2). By restricting themselves to temperatures above any characteristic spin fluctuation energy, they could ignore all but the time-averaged ($m = 0$) fluctuations of the ξ field. They could then obtain an explicit expression for the Tr ln term as a function of ξ_0. Writing the exponent in Eq. (2) as $-\psi(\xi_0)$, the function ψ develops two separated minima when U exceeds the value that produces the RPA instability. It is this two-well structure which gives rise to the Curie susceptibility. Numerical evaluations are easy because the remaining integral is one dimensional.

In the full N-center Hubbard problem, ψ is a function of N variables ξ_i, $i = 1, 2, \ldots, N$, even in the static approximation, and its structure is difficult to ascertain precisely. Its projection onto any ξ_i axis is likely to have a form similar to that of the $\psi(\xi_0)$ in the Anderson model, although the details of the structure may be washed out somewhat by interaction between sites. In general, there are of the order of $2N$ local minima of ψ in the N-dimensional space. But unless we decouple completely the spin-fluctuation fields on different sites (which reduces the problem to an ensemble of identical one-site problems such as the one considered by Wang *et al.*), we cannot hope even to evaluate $\psi(\xi_1, \ldots, \xi_N)$ explicitly, much less carry out the N-dimensional integration. We have to resort to finding approximations to ψ [or of the entire exponent of Eq. (2), if nonzero frequency fluctuations are retained], for which the N-fold integration is not prohibitively difficult.

In order to proceed in this direction, it is useful to look at the problem from a somewhat different physical viewpoint. We review the interacting paramagnon field theoretical picture of this system, which has been discussed in the context of the Anderson model and localized spin fluctuations by Schrieffer *et al.*[7]

We study the structure of the Tr ln term by expanding it in a power series in V. In the paramagnetic state, all odd order terms vanish because of the spin sum, since V^σ includes a factor σ. The second-order term is

$$-\frac{1}{2}\sum_\sigma \mathrm{Tr}(V^\sigma G^0)^2$$

$$= -c^2 \sum_{q,m} |\xi_{qm}|^2 \sum_{k,n} G^0(k, i\omega_n)\, G^0(k+q, i\omega_{n+m})$$

$$\equiv U \sum_{qm} |\xi_{qm}|^2 \varphi_2(q, i\nu_m)\ , \tag{8}$$

where φ_2 is just the zero-order polarization bubble [Fig. 1(a)], the generalized Lindhard function appropriate to the band structure of the system. To this order, the spin susceptibility is just

$$\chi(q, i\nu_m) = \frac{\varphi_2(q, i\nu_m)}{1 - U\varphi_2(q, i\nu_m)} \tag{9}$$

[using Eqs. (6) and (7)], which is just the RPA result. Now consider the next nonvanishing term in the expansion:

$$-\frac{1}{4}\sum_\sigma \mathrm{Tr}(V^\sigma G^0)^4 = -\tfrac{1}{2}c^4 \sum_{\substack{q_1,q_2,q_3\\ m_1,m_2,m_3}} \xi_{q_1 m_1}\xi_{q_2 m_2}\xi_{q_3 m_3}\xi_{-q_1-q_2-q_3,\,-m_1-m_2-m_3}$$

$$\times \sum_{kn} G^0(k, i\omega_n)\, G^0(k+q_1, i\omega_{n+m_1}) G^0(k+q_1+q_2, i\omega_{n+m_1+m_2})\, G^0(k+q_1+q_2+q_3, i\omega_{n+m_1+m_2+m_3})$$

$$= \tfrac{1}{2}\beta c^4 \sum_{\substack{q_1 q_2 q_3\\ m_1 m_2 m_3}} \xi_{q_1 m_1}\xi_{q_2 m_2}\xi_{q_3 m_3}\xi_{-q_1-q_2-q_3,\,-m_1-m_2-m_3}\, \varphi_4(q_1, q_2, q_3, i\nu_{m_1}, i\nu_{m_2}, i\nu_{m_3})\ . \tag{10}$$

The vertex φ_4 is pictured in Fig. 1(b). If we think of the functional integration in Eq. (2) as a trace operation for the scalar field $\xi_i(\tau)$, then the quadratic terms in the exponent can be identified as a kind of zero-order Hamiltonian describing the free propagation of this field, and the quartic terms can be interpreted as describing an interaction between modes of different q. Since φ_4 depends in general on all the q_i and ν_{m_i}, this interaction is nonlocal in space and time. Because we can identify the noninteracting limit of this theory with RPA spin-fluctuation (paramagnon) theory, we have here the basis of a field theory of interacting paramagnons.

In this language, the nth-order term in the expansion of the Tr ln in Eq. (2),

$$-\frac{1}{n}\sum_\sigma \mathrm{Tr}(V^\sigma G^0)^n = \frac{2}{n}\,\beta c^n \sum_{q_i, m_i} \prod_{i=1}^{n} \xi_{q_i, m_i} \times \delta\left(\sum_i q_i\right) \delta\left(\sum_i m_i\right) \varphi_n(q_i, i\nu_{m_i})\ , \tag{11}$$

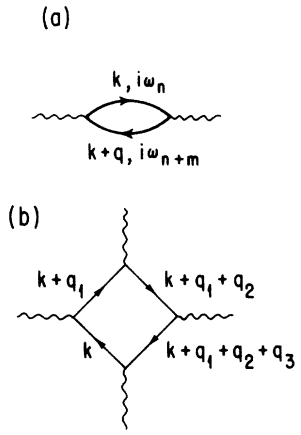

FIG. 1. (a) second- and (b) fourth-order terms in the expansion of Tr ln $(1-VG^0)$. Solid lines represent electron propagators; wavy ones, the random field V.

where φ_n, an electron loop with n vertices, describes an nth-order anharmonic interaction between paramagnons. The functional-integration representation has converted an interacting fermion problem into an interacting boson one. The interaction of the bosons, involving anharmonic, nonlocal vertices φ_n of all orders, is intrinsically more complex than that of the fermions (1), which is local and only quartic in the field operators, but as compensation the zero-order bosons already include an infinite series of perturbation-theoretical corrections to the zero-order fermion problem. This interacting-paramagnon picture may often be valid and useful even when the simple Hubbard model from which we derived it here is not valid. Of course we do need to know the correct underlying electronic model in order properly to evaluate the paramagnon-paramagnon vertices, but the general structure of the theory should apply to any system where spin fluctuations of some sort are important excitations.

A physically sensible simplification of the theory consists in making a local approximation to all the anharmonic vertices, that is, approximating the functions $\varphi_n(q_i, i\nu_{m_i})$ by constants independent of any of the q_i and ν_{m_i}. For illustrative purposes, consider the first φ, $\varphi_2(q, i\nu_m)$. For simplicity we restrict ourselves to $\nu_m = 0$ and examine the analytic form of $\varphi_2(q, 0)$ at zero temperature. In a free-electron model, φ_2 falls off slowly with q until the neighborhood of $q = 2k_F$, where it has an infinite negative derivative. It then falls off more slowly toward zero for $q > 2k_F$. Any system whose RPA instability is ferromagnetic rather than antiferromagnetic must have this general sort of q dependence. Its maximum must occur at $q = 0$, and it must have the $(q - 2k_F)\ln|q - 2k_F|$ singularity at $2k_F$. It is then not a bad approximation to take $\varphi_2(q) \approx \varphi_2(0)$ for $q < 2k_F$, and $\varphi_2(q) = 0$ otherwise. The analytic form of the higher order φ_n is much more difficult to calculate, but we do know that they also vary with momenta on a scale $\approx 2k_F$, so we can take $\varphi_n(q_1, \ldots, q_n) \approx \varphi_n(0, \ldots, 0)$ for all $q_i < 2k_F$, and $\varphi_n = 0$ when any $q_i > 2k_F$. This form of q dependence can be put into the functional integral expression for Z by writing it in the Fourier-analyzed form and cutting all momentum space sums off at $q = 2k_F$. Back in real space, this means that the paramagnon-paramagnon interactions have a finite effective range $1/2k_F$. Similar arguments can be made in frequency as well as wave vector; the reasonable cutoff is of the order of E_F. Of course we do not want to make this approximation for the quadratic term φ_2; if we did so for every φ_n the effective free-energy functional [the exponent of Eq. (2)] would be completely local and there would be no correlation of spins at different sites. Furthermore, in the intermediate coupling region, $U\varphi_2(0) \approx 1$, so even though φ_2 varies slowly with q, the total quadratic coefficient $1 - U\varphi_2$ has significant q dependence because the $q = 0$ part is very small. We therefore retain the true q and ν_m dependence of φ_2, which allows the free paramagnons to propagate from site to site. The resulting approximate theory describes local (but nonpolynomial) self-interactions of the paramagnon field.

The simplest sort of model effective free-energy functional which contains the physics of this mode-mode coupling picture is obtained by ignoring all interaction vertices of higher order than quartic, and making the local approximation as above for the quartic vertex. If we also make the static approximation of neglecting all but the $m = 0$ components of the ξ field, we have the most basic kind of interacting classical field theory. Murata and Doniach took this picture as the starting point of their analysis of spin fluctuations in itinerant paramagnets.[8] This sort of free-energy functional is also the starting point for the Wilson theory of critical fluctuations.[9]

In what follows we develop a generalized Hartree picture of paramagnon-paramagnon coupling, including vertices of all orders and nonlocal effects, which reduces to the Murata-Doniach theory in the quartic, local limit. Another way of describing it is as a very general kind of paramagnetic-polaron theory.

II. PARAMAGNETIC-POLARON THEORY

We follow Murata and Doniach and the earlier work of Muhlschlegel and Zittartz[10] on the Ising model in taking a variational approach to the problem of approximating the exact free-energy functional by one for which the functional integration

can easily be performed. Let ψ be the exact functional, that is

$$\psi\{\xi\}=\frac{1}{\beta}\int_0^\beta d\tau \sum_i \xi_i^2(\tau) - \sum_\sigma \mathrm{Tr}\ln(1-V^\sigma G^0) \quad (12)$$

in the present problem, and $\psi_0\{\xi\}$ be the approximate functional, whose parameters we want to determine. To lowest order in $\psi-\psi_0$, the true free energy is

$$F = F_0 + kT\langle \psi-\psi_0\rangle_0 , \quad (13)$$

where

$$-\beta F = \ln Z = \ln\int \prod_i \mathfrak{D}\,\xi_i(\tau)\, e^{-\psi\{\xi\}} , \quad (14)$$

$$-\beta F_0 = \ln\int \prod_i \mathfrak{D}\,\xi_i(\tau)\, e^{-\psi_0\{\xi\}} , \quad (15)$$

and $\langle A\rangle_0$ for some quantity A means

$$\langle A\rangle_0 = \int \prod_i \mathfrak{D}\xi_i(\tau) A e^{-\psi_0\{\xi\}} \Big/ \int \prod_i \mathfrak{D}\,\xi_i(\tau)\, e^{-\psi_0\{\xi\}} . \quad (16)$$

The higher-order corrections are positive, so Eq. (13) is an upper bound on the true free energy, and the optimal choice of ψ_0 is obtained by varying its parameters to minimize $\beta F_0 + \langle \psi-\psi_0\rangle_0$.

Like the above authors we use a quadratic ψ_0; because of translational invariance it is diagonal in q:

$$\psi_0 = \sum_{qm} A_{qm} \mid \xi_{qm}\mid^2 . \quad (17)$$

We can now evaluate

$$\beta F_0 + \langle \psi-\psi_0\rangle_0 = -\frac{1}{2}\sum_{qm}(1-\ln A_{qm}) + \int \prod_{qm}\left(\frac{dx_{qm}}{\sqrt{\pi}}\right) \exp\left(-\sum_{qm}\mid x_{qm}\mid^2\right) \times \psi\{xA^{-1/2}\}, \quad (18)$$

where we have made the change of variable $x_{qm} = A_{qm}^{1/2}\xi_{qm}$. The notation $xA^{-1/2}$ means that for each Fourier component, $(xA^{-1/2})_{qm} = x_{qm}A_{qm}^{-1/2}$. Stationarity with respect to variation of A_{qm} requires

$$0 = \frac{\partial}{\partial A_{qm}}(\beta F_0 + \langle \psi-\psi_0\rangle_0) = \frac{1}{2A_{qm}} - \frac{1}{2A_{qm}^{3/2}}\int \prod_{qm}\left(\frac{dx_{qm}}{\sqrt{\pi}}\right) \times \exp\left(-\sum_{qm}\mid x_{qm}\mid^2\right) x_{qm}\,\frac{\partial\psi\{xA^{-1/2}\}}{\partial(x_{qm}/A_{qm}^{1/2})} . \quad (19)$$

Integrating by parts and reverting to the original variables ξ_{qm}, we find

$$A_{qm} = \frac{1}{2}\left\langle \frac{\partial^2\psi\{\xi\}}{\partial\xi_{qm}\,\partial\xi_{-q,-m}}\right\rangle , \quad (20)$$

which might almost have been guessed in advance—the optimal quadratic functional coefficients are just (self-consistently) averaged second derivatives of the true functional. This result should be contrasted with the RPA (steepest descents), in which the second derivatives are evaluated at the origin:

$$A_{qm}^{\mathrm{RPA}} = \frac{1}{2}\left.\frac{\partial^2\psi\{\xi\}}{\partial\xi_{qm}\,\partial\xi_{-q,-m}}\right|_{\xi=0} . \quad (21)$$

Applying Eq. (20) to the free energy functional of our model Eq. (12) is most easily accomplished by working in the representation where the matrix $K^\sigma = V^\sigma G^0$ is diagonal:

$$\sum_\sigma \mathrm{Tr}\ln(1-K^\sigma) = \sum_{\sigma,\alpha}\ln(1-K_\alpha^\sigma) . \quad (22)$$

The index α labels the eigenvalues of K. Then

$$\frac{\partial^2}{\partial\xi_q\,\partial\xi_q}\left(-\sum_\sigma \mathrm{Tr}\ln(1-K^\sigma)\right) = \sum_{\sigma,\alpha}(1-K_\alpha^\sigma)^{-1}\,\frac{\partial K_\alpha^\sigma}{\partial\xi_q}\,\frac{\partial K_\alpha^\sigma}{\partial\xi_{-q}} . \quad (23)$$

In this section we use q to stand for the four-vector $(q, i\nu_m)$. Now define $\langle\alpha\mid k\rangle$ as the matrix elements of the transformation that diagonalizes K,

$$K_\alpha^\sigma = \sum_{kk'}\langle\alpha\mid k\rangle K_{kk'}^\sigma\langle k'\mid\alpha\rangle = \sigma c\sum_{kk'}\langle\alpha\mid k\rangle\,\xi_{k-k'}\,G_{k'}^0\langle k'\mid\alpha\rangle . \quad (24)$$

Therefore,

$$\frac{\partial K_\alpha^\sigma}{\partial\xi_q} = \sigma c\sum_k\langle\alpha\mid k+q\rangle G_k^0\langle k\mid\alpha\rangle \quad (25)$$

and

$$\frac{\partial^2}{\partial\xi_q\,\partial\xi_{-q}}\left(-\sum_\sigma \mathrm{Tr}\ln(1-K^\sigma)\right) = c^2\sum_{\alpha\sigma kk'} G_k^0\langle k\mid\alpha\rangle(1-K_\alpha^\sigma)^{-1}\langle\alpha\mid k'\rangle \times G_{k'+q}^0\langle k'+q\mid\alpha\rangle(1-K_\alpha^\sigma)^{-1}\langle\alpha\mid k+q\rangle . \quad (26)$$

In Eq. (26) we have rearranged the factors to make it clear that

$$\frac{\partial^2}{\partial\xi_q\,\partial\xi_q}\left(-\sum_\sigma \mathrm{Tr}\ln(1-K^\sigma)\right) = c^2\sum_{\sigma kk'} G_{kk'}^\sigma\{\xi\}G_{k'+q,k+q}^\sigma\{\xi\} , \quad (27)$$

where $G^\sigma\{\xi\} = G^0(1-V^\sigma G^0)^{-1}$ is the propagator of an electron subject to the potential $\sigma c\,\xi_i(\tau)$. This expression is then just an electron-hole bubble in the presence of this random Gaussian potential.

Alternatively, we can derive Eq. (27) diagrammatically, as follows. The expansion of the Tr ln in Eq. (12) generates a set of closed-loop diagrams such as Fig. 2(a). The nth-order diagram contains n random fields ξ_{q_i}, n factors of c, an electron

loop with n Green's functions, a factor $1/n$ because we are expanding a log, and a sum on σ. (Only even-order terms survive the spin sum, but we leave all terms in for the moment.) Differentiating with respect to ξ_q gives a factor of c and removes any one of the ξ (wiggly) lines of momentum q. We indicate this by replacing the wiggly line by a dotted one, as shown in Fig. 2(b). Because of cyclic invariance, there are n ways to do this in the nth-order diagram, cancelling the factor of $1/n$. Then differentiating again removes any of the remaining wiggly lines of momentum $-q$ and gives another factor of c [Fig. 2(c)]. This time, however, there is no cyclic symmetry, and the $n-1$ different ways to do this have to be counted separately. So we have the sum of all electron loops having two dotted line vertices with four-momentum transfers $\pm q$, and arbitrary numbers of wiggly lines dressing the electron lines between these vertices. But an electron line dressed in this manner is just the propagator in the external field $\sigma c \xi_i(\tau)$, so what we have is a diagrammatic representation of Eq. (27).

Either way, when we add in the derivative of the first term in ψ, we are led to an expression for A_{qm} of the form

$$A_{qm} = 1 - U\overline{\varphi}(q, i\nu_m) , \tag{28}$$

where $U\overline{\varphi}$ is half the average (over the distribution $e^{-\psi_0}$) of Eq. (27),

$$\overline{\varphi}(q, i\nu_m) = -\frac{1}{\beta}\sum_{k',k} \langle G^\sigma(k, k', i\omega_n, i\omega_{n'}; \{\xi\}) \times G^\sigma(k+q, k'+q, i\omega_{n+m}, i\omega_{n'+m}; \{\xi\})\rangle , \tag{29}$$

that is, $\overline{\varphi}$ is the averaged electron-hole bubble (of either spin) in the presence of the random fields. (To lowest order in U, we ignore the random fields, $\overline{\varphi}$ reduces to the bare bubble φ_2, and we recover the RPA result.)

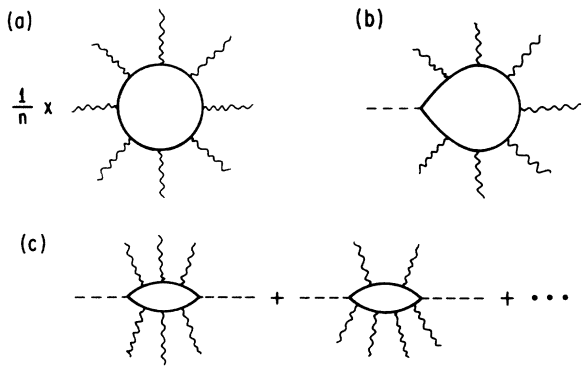

FIG. 2. (a) Diagrammatic form of the nth order term in Tr ln $(1-VG^0)$. (b) Result of one differentiation with respect to V. (c) Series of diagrams resulting from a second differentiation with respect to V.

Equation (29) allows us to exploit our knowledge of the properties of response functions of disordered systems. We can also use the Gaussian nature of the distribution of the random fields to formally carry out the averaging procedure in any of the diagrams of Fig. 2(c). These contain products of Fourier components of V^σ, whose average

$$P_n(q_1, q_2, \ldots) = \left\langle V^\sigma(q_1)V^\sigma(q_2)\cdots V^\sigma(q_{n-1})\, V^\sigma\left(-\sum_{i=1}^{n-1} q_{n_i}\right)\right\rangle \tag{30}$$

is simple to compute for a Gaussian distribution. First, all P_n for odd n vanish. Then, for even n, because there are no cumulants of order higher than two, we have simply

$$P_n(q_1, q_2, \ldots) = \sum_P \langle V^\sigma(q_{j_1})V^\sigma(q_{j_2})\rangle \langle V^\sigma(q_{j_3})V^\sigma(q_{j_4})\rangle \times\cdots\langle V^\sigma(q_{j_{n-1}})V^\sigma(q_{j_n})\rangle , \tag{31}$$

where the sum is over all permutations $(1, \ldots, n) \to (j_1, \ldots, j_n)$. Because of translational invariance, the averaged pair products vanish unless in each pair one four-momentum argument is the negative of the other:

$$P_n(q_1, q_2, \ldots) = \sum_P \langle |V^\sigma(q_{j_1})|^2\rangle \delta(q_{j_1}+q_{j_2}) \langle |V^\sigma(q_{j_3})|^2\rangle \times \delta(q_{j_3}+q_{j_4})\cdots\langle |V^\sigma(q_{j_{n-1}})|^2\rangle \delta(q_{j_{n-1}}+q_{j_n}) . \tag{32}$$

In diagrammatic terms, this means that we connect all possible pairs of wiggly lines and conserve the four momenta carried by these lines, as shown in Fig. 3. The natural physical interpretation is of an electron-hole pair dressed by the emission and reabsorption of paramagnons in all pairs of perturbation theory, hence the name paramagnetic-polaron theory. We may take each of the wiggly lines here to represent the dimensionless paramagnon propagator $\langle |\xi_{qm}|^2\rangle$, and give each electron-paramagnon vertex a value c. All intermediate momentum and energy indices are summed over, *without* the factor $1/\beta$ which accompanies a Matsubara sum in the usual many-body formalism.[11] Here the $1/\beta$ comes from the vertex factors of $c = (U/\beta)^{1/2}$.

A similar analysis applies to the Green's function of the interacting system, with the functional integral representation (4). If we replace the exact distribution of random fields $W\{\xi\} = e^{-\psi\{\xi\}}$ by the optimal variational one $e^{-\psi_0\{\xi\}}$ and cancel the de-

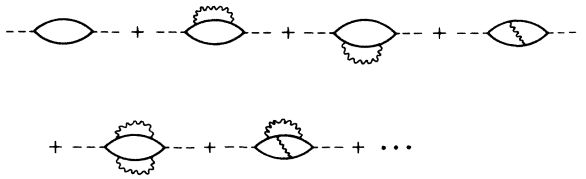

FIG. 3. Diagrammatic expansion of $\overline{\varphi}$. Wavy lines represent the paramagnon propagators.

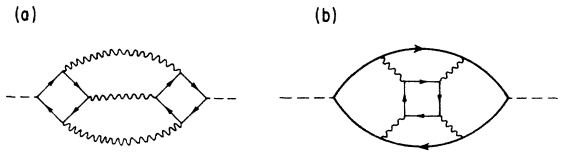

FIG. 5. Some diagrams present in the exact $\overline{\varphi}$, but not included in the approximation of this paper.

nominator with all the unlinked graphs in the expansion of the numerator, we have the series for G given in Fig. 4.

There are a few more important points to note about this polaronic-model theory and its relation to the exact theory. First, as far as the fermion propagators are concerned, evaluating G is a one-electron problem, and evaluating $\overline{\varphi}$ is a one-electron and one-hole problem. No diagrams involving additional electron-hole pairs are generated in this approximation. Therefore, some terms in the full diagrammatic perturbation theory (derivable from Eq. (2), for example, by a direct expansion of the exponential) are omitted. Some examples are shown in Fig. 5. Note that among the omitted diagrams are any with interactions between the emitted paramagnons [such as Fig. 5(b)]. This is consistent with the independence of different paramagnon modes implicit in the quadratic-model free-energy functional (17). In these respects this theory is analogous to conventional polaron theory, where one studies the motion of a single electron (or electron-hole pair, if collective or transport properties are of interest) under the influence of a boson field whose motion is entirely harmonic. It differs from ordinary polaron theory in that the parameters describing the paramagnon field, the A_{qm}, depend on the electronic properties through $\overline{\varphi}$. Note also that $\overline{\varphi}$ is not merely a bubble consisting of dressed electron and dressed hole. This would omit all vertex corrections, where the paramagnon is emitted by the electron and absorbed by the hole, or vice versa, as in the fourth diagram of Fig. 3.

Finally, we can easily connect this picture with a generalized Hartree approximation in the diagrammatic perturbation theory of the interacting paramagnon-field theory. We define the paramagnon propagator

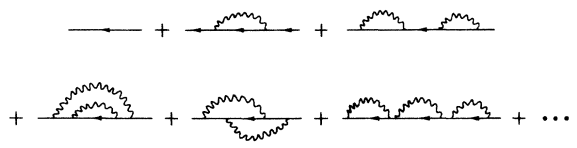

FIG. 4. Diagrammatic expansion for the electron propagator in the presence of the paramagnons.

$$D(q, i\nu_m) = 2\langle\,|\,\xi_{qm}\,|^2\rangle = [1 - U\overline{\varphi}(q, i\nu_m)]^{-1} \tag{33}$$

and a self-energy

$$\Pi(q, i\nu_m) = U[\overline{\varphi}(q, i\nu_m) - \varphi_2(q, i\nu_m)]\ , \tag{34}$$

so that

$$D^{-1}(q, i\nu_m) = D_0^{-1}(q, i\nu_m) - \Pi(q, i\nu_m)\ , \tag{35}$$

where D_0 is the free- (RPA) paramagnon propagator. We can then look at the diagrams of Fig. 3 as paramagnon self-energy diagrams. The constraints placed on the diagrams by the exclusion of extra electron-hole pairs and interactions between paramagnons naturally restricts the series to those which can be described as self-consistent Hartree terms.

III. FURTHER APPROXIMATIONS WITHIN PARAMAGNETIC-POLARON THEORY

From now on we will work within the static approximation, where all Fourier components ξ_{qm} of the fluctuation field with nonzero ν_m are ignored. Physically, this approximation makes sense whenever the temperature is high enough for thermal fluctuations to wash out the details of the dynamics of the spin fluctuations, that is, whenever kT is much larger than the characteristic spin-fluctuation relaxation rate kT_{sf}. In the elemental transition metal ferromagnets, where most of the magnetic spectral weight is concentrated in the collective modes, a simple estimate for T_{sf} is just T_C, the local energy required to destroy the magnetic order, so the static approximation should be valid for $T \gg T_C$. In fact, as T is lowered toward T_C and the spin fluctuations exhibit critical slowing down, the validity of the static approximation should be preserved for all $T > T_C$. (Current theories of critical fluctuations make this hypothesis near T_C.) In systems which are only weakly ferromagnetic or almost ferromagnetic, there is still a characteristic $T_{sf} \approx T_F(\chi/\chi_{Pauli})$. Most of the spectral weight in χ lies at energies $\leq T_{sf}$, so what follows should be meaningful for $T > T_{sf}$ in these systems.

We are now faced with a problem in the theory of disordered systems—the evaluation of the average susceptibility $(\overline{\varphi})$ in the presence of static ran-

dom potentials. The statistics of these potentials are described by the covariance matrix

$$\langle V_i V_j\rangle = UkT\langle \xi_{i,\nu=0}\,\xi_{j,\nu=0}\rangle = \tfrac{1}{2}UkT\sum_q e^{i\vec{q}\cdot(\vec{r}_i-\vec{r}_j)}(1-U\bar{\varphi}(q,0))^{-1}\ . \quad (36)$$

If the dimensionless field ξ_i has an rms size of order unity (and we shall argue that this is reasonable in the intermediate coupling region), we have $\langle V_i^2\rangle \approx UkT$, implying a strong incoherent mixing of electron states whose energies lie within roughly this range of each other.

Such a picture emerges naturally if we make the local interaction approximation in the paramagnon-field theory picture described in Sec. I, and sum all the Hartree diagrams for the self-energy $\Pi(q)$. The lowest-order diagrams (fourth order in c) are the second, third, and fourth in Fig. 3, and each of them has the value

$$c^4\beta\varphi_4(0,0,0,0)\xi_{\rm rms}^2\ , \quad (37)$$

where

$$\xi_{\rm rms}^2 = \langle \xi_i^2\rangle = \frac{1}{N}\sum_q \langle\,|\,\xi_q\,|^2\rangle\ . \quad (38)$$

Similarly, in nth order, each of the $(n-1)!!$ $=(n-1)(n-3)\cdots 3\times 1$ distinct diagrams is equal to

$$c^n\beta\varphi_n(0,\ldots)\xi_{\rm rms}^{n-2}\ . \quad (39)$$

The vertices $\varphi_n \equiv \varphi_n(0,\ldots)$ are loops of n electron Green's functions, all of the same four momentum:

$$\varphi_n = -\frac{1}{\beta}\sum_{k,n'} G^n(k, i\omega_{n'})\ . \quad (40)$$

We use the Poisson summation formula to convert Eq. (40) into a contour integral

$$\varphi_n = \sum_k \int_C \frac{dE}{2\pi i}\,\frac{f(E)}{(E-\epsilon_k)^n}\ , \quad (41)$$

where C encloses the fermion Matsubara frequencies in a counterclockwise direction, and $f(E)$ is the Fermi function. C may be continuously deformed into a path which encircles the nth-order pole at ϵ_k, clockwise. We then expand $f(E)$ in a power series around ϵ_k. Only the $(n-1)$st-order term survives the contour integral, so we obtain

$$\varphi_n = -\frac{1}{(n-1)!}\sum_k f^{(n-1)}(\epsilon_k) = -\frac{1}{(n-1)!}\int d\epsilon\, N_0(\epsilon) f^{(n-1)}(\epsilon)\ , \quad (42)$$

where $N_0(E)$ is the band density of states. φ_n can also be expressed in a slightly different way by integrating Eq. (42) by parts, $n-2$ times:

$$\varphi_n = \frac{1}{(n-1)!}\int d\epsilon\, N_0^{(n-2)}(\epsilon)\left(\frac{-\partial f}{\partial\epsilon}\right) \equiv \frac{1}{(n-1)!}\,N_T^{(n-2)}(E_F)\ . \quad (43)$$

That is, φ_n is proportional to the $(n-2)$nd derivative of the density of states near the Fermi energy, averaged over an energy range whose width is of order kT through the factor $(-\partial f/\partial E)$. As $T\to 0$, we have simply

$$\varphi_n \to \frac{1}{(n-1)!}\,N_0^{(n-2)}\ . \quad (44)$$

We can now sum the entire series of Hartree self-energy terms as follows. From Eqs. (39) and (43) we have

$$\Pi(q) = \sum_{n=2}^{\infty}\beta c^{2n}\frac{1}{(2n-1)!} \times\int d\epsilon\, N_0^{(2n-2)}(\epsilon)\left(\frac{-\partial f}{\partial\epsilon}\right)(2n-1)!!\,\xi_{\rm rms}^{2n-2}\ . \quad (45)$$

Since the local field ξ_i is a sum of independent Gaussian random variables $(N^{-1/2}e^{iq\cdot r_i}\xi_q)$, its distribution is also Gaussian, and hence (for even n)

$$\langle \xi_i^n\rangle = (n-1)!!\,\langle \xi_i^2\rangle = (n-1)!!\,\xi_{\rm rms}^2\ . \quad (46)$$

(From now on we leave out the subscript i in $\langle \xi_i^n\rangle$, since it does not depend on i.) Then

$$\Pi(q) = \sum_{n=2}^{\infty}\beta c^{2n}\frac{1}{(2n-2)!} \times\int d\epsilon\, N_0^{(2n-2)}(\epsilon)\left(\frac{-\partial f}{\partial\epsilon}\right)\langle \xi^{2n-2}\rangle = \beta c^2\int d\epsilon\left(\frac{-\partial f}{\partial\epsilon}\right)\sum_{n=1}^{\infty}\frac{1}{(2n)!}\,N_0^{(2n)}(\epsilon)\,c^{2n}\langle \xi^{2n}\rangle\ . \quad (47)$$

Since the odd moments vanish, we may equally well write this as

$$\Pi(q) = \beta c^2\int d\epsilon\left(\frac{-\partial f}{\partial\epsilon}\right)\sum_{n=1}^{\infty}\frac{1}{n!}\,N_0^{(n)}(\epsilon)\,c^n\langle \xi^n\rangle\ , \quad (48)$$

which sums up to

$$\Pi(q) = U\int d\epsilon\left(\frac{-\partial f}{\partial\epsilon}\right)[\langle N_0(\epsilon + c\xi)\rangle - N_0(\epsilon)] = U\langle N_T(E_F + c\xi)\rangle - U\varphi_2(0)\ . \quad (49)$$

The average is over the distribution of the local ξ field. The zeroth-order term of the series in Eq. (48), which is added in and subtracted in Eq. (49), just cancels the RPA part of the inverse paramagnon propagator at $q=0$.

$$D(q) = [1 - U\langle N_T(\epsilon_F + c\xi)\rangle + U(\varphi_2(0) - \varphi_2(q))]^{-1}\ . \quad (50)$$

That is, in this local interaction approximation, the q-dependent part of D^{-1} is given by RPA, and the constant part is like the corresponding RPA expression $1 - UN_T(E_F)$, except that $N_T(E)$ is averaged over an energy region of width $c\xi_{\rm rms}$, in agreement with the qualitative picture we suggested earlier. The averaging in Eq. (49) is, explicitly,

$$\langle N_T\rangle = \langle N_T(E_F + c\xi)\rangle = \int \frac{d\xi}{(2\pi\xi_{\rm rms}^2)^{1/2}}\, N_T(E_F + c\xi) \times \exp(-\xi^2/2\xi_{\rm rms}^2) \,. \quad (51)$$

and it must be solved simultaneously with the self-consistency condition (38), which we write here as

$$\xi_{\rm rms}^2 = \frac{1}{2N}\sum_q \frac{1}{1 - U\langle N_T\rangle + U(\varphi_2(0) - \varphi_2(q))} \,. \quad (52)$$

The reason the q dependence of the paramagnon propagator is the same as in RPA is that the local approximation imposes a momentum-independent self-energy (within the Hartree approximation). We can improve on this approximation somewhat by allowing the external paramagnon lines (dotted lines in Fig. 3) to have general values of q and evaluating the bubble as if only the *internal* (wiggly) paramagnons had zero momentum. To see where this leads, it is simplest to go back to the expression for the electron-hole bubble before averaging over the distribution of the random fields [Fig. 2(c)]. Evaluating the bubble as if all the internal momentum transfers were zero means evaluating the propagators *in a uniform field,* or, in other words, with a shifted Fermi surface. This expression is then averaged over the distribution of local fields, as in Eq. (51):

$$\overline{\varphi}(q) = \langle \varphi_2(q; E_F + c\xi)\rangle = \int \frac{d\xi}{(2\pi\xi_{\rm rms}^2)^{1/2}}\, \varphi_2(q; E_F + c\xi) \times \exp(-\xi^2/2\xi_{\rm rms}^2) \quad (53)$$

or just $\langle\varphi_2(q)\rangle$ for short, and the propagator has the simple form

$$D(q) = [1 - U\langle\varphi_2(q)\rangle]^{-1} \,. \quad (54)$$

The self-consistency condition is now

$$\xi_{\rm rms}^2 = \frac{1}{2N}\sum_q \frac{1}{1 - U\langle\varphi_2(q)\rangle} \,, \quad (55)$$

so the entire Lindhard function is averaged over a distribution of Fermi levels in the same way that its constant part was in the strictly local approximation.

We can get a reasonable idea of the size of the $\xi_{\rm rms}^2$ as follows. $\overline{\varphi}(q) = \langle\varphi_2(q)\rangle$ should vary with q on the scale of inverse interatomic distances, or of the Fermi momentum. So let us make a Debye cutoff $q_c = (6\pi^2 N)^{1/3}$ in the sum in Eq. (55) and approximate $\langle\varphi_2(q)\rangle$ by

$$\overline{\varphi}(q) \approx \overline{\varphi}(0)(1 - q^2/q_c^2) \,. \quad (56)$$

Now if the enhancement of χ is weak, i.e., $U\overline{\varphi}(0) \ll 1$, we have

$$(\xi_{\rm rms}^2)_{\rm min} = \tfrac{1}{2} \,, \quad (57)$$

while in the opposite limit $1 - U\overline{\varphi}(0) \ll 1$, we find

$$(\xi_{\rm rms}^2)_{\rm max} = \tfrac{3}{2} \,. \quad (58)$$

Hence $\xi_{\rm rms}$ is always of order unity, so $\langle V_i^2\rangle^{1/2} = (UkT)^{1/2}\xi_{\rm rms}$ is of order $(UkT)^{1/2}$ and is only weakly dependent on the degree of enhancement. The latter feature occurs because of the assumed q dependence of Eq. (56) and the fact that large q fluctuations make the most important contribution to $\xi_{\rm rms}$. Our calculation thus supports the qualitative picture we suggested earlier of an effective density of states (or better, effective Lindhard function) obtained by averaging the corresponding RPA quantity over an energy range of width $\approx (UkT)^{1/2}$.

Any finite order approximation to $\Pi(q)$ effectively approximates $N_T(E)$ by a *finite* power series expansion around E_F {as an examination of the structure of the vertices φ_n [Eq. (43)] makes clear}. For example, the Murata-Doniach approximation, which stops at fourth order in c, approximates $N_T(E)$ by its quadratic expansion around E_F. It therefore must break down whenever $\langle V_i^2\rangle^{1/2} \approx (UkT)^{1/2}$ gets much larger than the distance from E_F where this expansion becomes inaccurate. This may explain the breakdown of the Murata-Doniach theory in Sc_3In above about 15 °K, and in $ZrZn_2$ in the entire paramagnetic region ($T_c = 25$ °K). In general, if the Fermi level lies in a peak of width δ in the density of states, higher-order interactions will be important whenever $kT \gtrsim \delta^2/U$. The Fermi level is generally believed to lie near peaks in $N_0(E)$ in most itinerant ferromagnets and nearly ferromagnetic metals, as well as in the $A15$-structure transition-metal compounds, important because of the relatively large superconducting transition temperatures which many of them have. This theory should be relevant to all these systems.

The susceptibility follows directly from Eq. (6) with the definition of $\overline{\varphi}$ in Eq. (29). In the approximation (55), we have

$$\chi(q) = \langle\varphi_2(q)\rangle/[1 - U\langle\varphi_2(q)\rangle] \,. \quad (59)$$

Again, the result is analogous to RPA, with φ_2 replaced by its average over the local-field distribution.

The Fermi energy E_F referred to in all the foregoing is the temperature-dependent chemical potential, not the zero-temperature Fermi level. It is determined by fixing the total number of electrons:

$$n = \int f(\epsilon)\,\overline{N}(\epsilon)\,d\epsilon \,, \quad (60)$$

where the many-body density of states

$$\overline{N}(\epsilon) = \frac{1}{\pi}\sum_k |\,\mathrm{Im}\,G(k,\epsilon)\,| = \frac{1}{\pi}\,|\,\mathrm{Im}\,G_{ii}(E)\,| \quad (61)$$

depends on a knowledge of the Green's function, Eq. (4), or the diagrammatic series, Fig. 4.

There is a wealth of approximate techniques for both evaluating fermion propagators in the presence

of a boson field, and evaluating the average propagator in the presence of a disordered potential. Many of these may be of use here, but for qualitative purposes we will only discuss a particularly simple one. It is the analog in the evaluation of G of the local interaction approximation in the evaluation of the paramagnon propagator, and shares with it the feature of *counting* all diagrams properly, while making rather crude estimates of the value of individual diagrams.

As in the arguments leading to Eqs. (53) and (54), we evaluate all the electron propagators in the diagrams as if the momentum transfer to the paramagnon lines were zero, or, equivalently, as if the ξ field were uniform. The result is an average over the distribution of the local field ξ_i of

$$G(k, i\omega_n; E_F + c\xi) = (i\omega_n - \epsilon_k + c\xi)^{-1} \tag{62}$$

and, after summing on k and taking the imaginary part, an averaging of the band structural $N_0(E)$ over this Gaussian distribution, exactly as in Eq. (51), to give the many-body density of states. [This result can also be obtained by explicitly summing the series of diagrams of Fig. 4, letting every internal G line have the same four-momentum as the external ones. Then each term with n paramagnon lines is just n factors of $c^2\xi_{\rm rms}^2$ and $2n+1$ factors of $G(k, i\omega_n)$, and each of the $(2n-1)!!$ distinct nth-order diagrams has an identical value. The summation is then expressible as a power series expansion of the average of Eq. (62), in the same way that the series for $\Pi(q)$ led to an average of $N_T(E)$. Notice incidentally that this convenient summation was possible because we made *no* separation of reducible and irreducible diagrams for G, nor attempted to proceed via calculating a self-energy.]

Certainly more powerful and less crude approximations should be brought to bear on the calculations of both D and G, but we feel that the foregoing discussion at least contains the elements of the problem, and has the virtue of a transparent physical interpretation in the local fluctuating Zeeman field of size $c\xi_{\rm rms}$.

IV. CALCULATIONS

A. Gaussian density of states

To illustrate the above ideas, we have calculated the temperature dependence of the susceptibility for a model density of states whose form allows most of the calculations to be performed analytically. We take

$$N_0(\epsilon) = (2\pi W^2)^{-1/2} e^{-\epsilon^2/2W^2} \tag{63}$$

and assume that $\overline{\varphi}(q)$ varies with q as in Eq. (56). We should obtain $\overline{\varphi}(0) = \langle N_T(c\xi)\rangle$ by first convoluting N_0 with the derivative of the Fermi function to obtain N_T, and then convoluting the result with the Gaussian distribution of local fluctuating ξ fields. We will approximate the first stage of this process by replacing the derivative of the Fermi function by a Gaussian with the same second moment $\frac{1}{3}\pi^2 \times (kT)^2$. This will give a qualitatively correct description of the thermal smearing effects (which are not very important until $kT \gtrsim U$) and now both convolutions are analytically trivial. We obtain

$$N_T(\epsilon) = \{2\pi[W^2 + \tfrac{1}{3}(\pi kT)^2]\}^{-1/2} \times \exp\{-\epsilon^2/2[W^2 + \tfrac{1}{3}(\pi kT)^2]\}\,, \tag{64}$$

and, using Eq. (51),

$$\overline{\varphi}(0) = \{2\pi[W^2 + \tfrac{1}{3}(\pi kT)^2 + c^2\xi_{\rm rms}^2]\}^{-1/2}\,. \tag{65}$$

Now taking the form (56) for $\overline{\varphi}(q)$ and evaluating $\xi_{\rm rms}^2$,

$$\xi_{\rm rms}^2 = \tfrac{3}{2}(1+\lambda)(1 - \lambda^{1/2}\tan^{-1}\lambda^{-1/2})\,, \tag{66}$$

where

$$\lambda = (U\chi)^{-1} = [U\overline{\varphi}(0)]^{-1} - 1\,. \tag{67}$$

Eliminating two of the three unknowns in Eqs. (65)–(67), $\overline{\varphi}(0)$ and $\xi_{\rm rms}^2$, leads to a single equation for λ:

$$(U^2/2\pi)(1+\lambda)^2 - W^2 - \tfrac{1}{3}(\pi kT)^2 = \tfrac{3}{2}UkT(1+\lambda)(1 - \lambda^{1/2}\tan^{-1}\lambda^{-1/2})\,. \tag{68}$$

We have solved Eq. (68) by numerical iteration, and the resulting inverse susceptibilities are plotted in Fig. 6, for three different values of U. For the smallest of these, the system remains paramagnetic down to $T=0$. The second value is just at the threshold for zero-temperature ferromagnetism, and the third has a finite Curie temperature of about $0.32\,W$. When a solution of Eq. (68) exists at $T=0$, χ is the same there as in RPA, since the RPA result is obtained by ignoring the right-hand side, which vanishes when $T=0$. As T increases, however, the inverse susceptibility obtained here rises with something much more like a linear temperature dependence than does the RPA χ^{-1}. For the threshold value of U, our χ obeys nearly a perfect Curie law at all temperatures. Of course, these results have only a qualitative significance at very low temperatures $\lesssim T_{sf}$, the spin-fluctuation temperature, since the time dependence of the fluctuations must be taken into account there. For U larger than the threshold value, χ^{-1} starts out like $(T - T_c)^2$, and then quickly straightens out to approximate Curie-Weiss behavior. The quadratic dependence near T_c derives analytically from the fact that the right-hand side of Eq. (68) is proportional to $1 - (1/2)\pi\lambda^{1/2}$ for small λ. Such a temperature dependence is characteristic of averaged-fluctuation approximation schemes like the present one—it occurs in the Murata-Doniach theory and in the Berlin-Kac spherical model[12] as well. The T_c

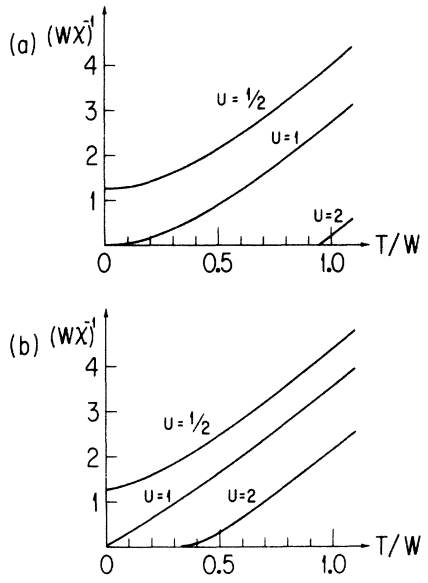

FIG. 6. Inverse susceptibility as a function of temperature for the model of Sec. IVA. (a) The RPA result; (b) obtained from Eq. (68). U is in units of $U_{\text{crit}} = (2\pi W^2)^{1/2}$, the largest U for which the system remains paramagnetic at $T = 0$.

obtained here is about a third of the RPA T_c.

B. Application to nickel

We turn now to numerical calculations carried out for a system with a complex band structure, and for whose magnetic properties an intermediate-coupling description is probably appropriate. As is well known, the bare zero-temperature d-band density of states $N_0(E)$ in nickel has a rather-high rather-narrow peak just below the band edge, and the Fermi level lies very near its maximum.[13] The thermally averaged $N_T(E)$ may be significantly smeared out only when kT is greater than about the width of the peak (about 0.5 eV). As remarked above, the ξ fluctuations produce an averaging over an energy range $(UkT)^{1/2}$. Thus, for nickel ($T_c = 631\,^\circ\text{K}$, $U = 4$ eV) we expect the averaging over the ξ distribution to be important and to dominate the thermal smearing in determining the susceptibility as a function of temperature above T_c.

Our procedure is to seek ξ^2_{rms} from the conditions on $\overline{\varphi}$ given in Sec. III [Eqs. (53) and (55)], and at the same time to adjust the Fermi energy so that

$$\int_{-\infty}^{E_F} d\epsilon \int \frac{d\xi}{(2\pi\xi^2_{\text{rms}})^{1/2}} \times \exp\left(\frac{-\xi^2}{2\xi^2_{\text{rms}}}\right) N_T(\epsilon + c\xi) = \int_{-\infty}^{E_F^0} N_0(\epsilon)\, d\epsilon \,, \qquad (69)$$

where E_F^0 is the zero-temperature Fermi energy of the noninteracting system. We assume the form for $\overline{\varphi}(q)$ given in Eq. (56) and write

$$\frac{1}{N}\sum_q \langle |\, \xi_q \,|^2 \rangle = \frac{1}{2\pi^2}\int_0^{q_c} q^2\, dq\, [1 - U\overline{\varphi}(q)]^{-1} \,. \qquad (70)$$

In Sec. I we argued that the natural choice for q_c was $2k_F$, and in nickel a typical k_F for the d band holes is perhaps a quarter or a third of the Debye wave vector. The solutions of the set of self-consistent equations are then used to calculate the susceptibility (6).

Figure 7 shows the results of numerical calculations of χ^{-1} carried out using the density of states given in Ref. 13 for values of U between 4.0 and 6.4 eV, for two values of q_c. For comparison, we also display the inverse of the RPA susceptibility (9). The larger q_c is, the more we expect the calculated curves to differ from the RPA ones.

First we remark that, as in the case of the Gaussian $N_0(E)$, we still have the RPA criterion for the existence of zero-temperature ferromagnetism [$N_0(E_F)\, U \geq 1$], but the T_c obtained here for a given U is lower than the RPA value—that is, the averaging effect introduced by the fluctuating ξ field suppresses the tendency toward magnetic order at finite T. The values of T_c obtained here for given values of U are obviously not to be taken quantitatively seriously, given the crudeness of the approximations made in evaluating $\overline{\varphi}(q)$ and in neglecting effects which arise from detailed consideration of the multiple band structure. However, it seems clear that this shift in T_c will occur in general for a peaked density of states.

Secondly, for large enough q_c, χ^{-1} curves downward at high T from the straight line which one would obtain by extrapolating the Curie-Weiss law at lower temperatures. The strength of this effect depends strongly on the size of the region of q space in which fluctuations contribute to ξ^2_{rms}, so it is difficult to make a quantitative estimate of χ here. However, the tendency to curve over at high T is observed in experimental data,[14] and this effect is not very pronounced in the RPA susceptibility [Fig. 7(c)].

The effective magnetic moments obtained from these curves vary somewhat with U and q_c, but they are all around one Bohr magneton. For the curves shown for $q_c = 0.77 q_0$ the effective moment varies from 0.9 μ_B ($U = 4$ eV) to 1.3 μ_B ($U = 6.4$ eV). The experimentally observed moment (between $\approx T_c$ and 1000 °K) is somewhat larger, 1.61 μ_B.[15]

We have not attempted to calculate χ very close to T_c, where our iteration procedure for solving the self-consistent equations (53)–(55) does not converge very quickly. If we were to do so, we would find the quadratic dependence of χ^{-1} on $(T - T_c)$ discussed above for the case of the Gaussian $N_0(E)$. As in that case, however, this behavior

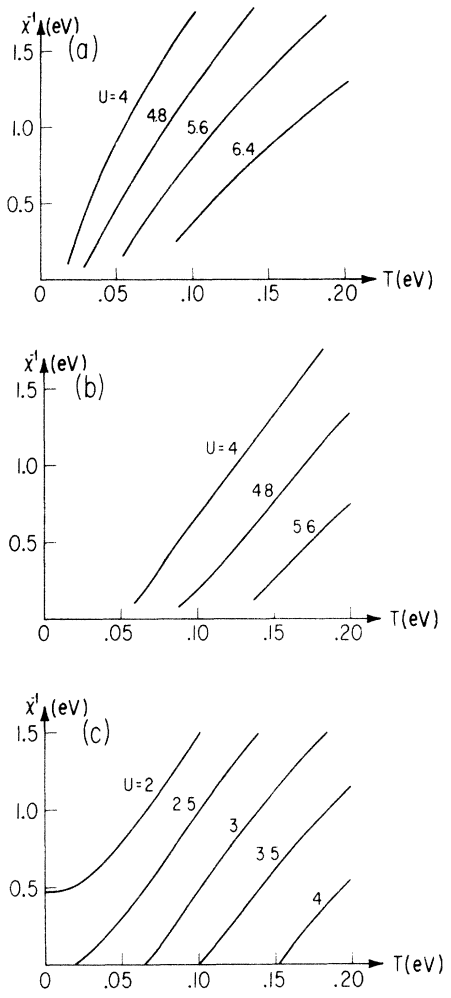

FIG. 7. Inverse susceptibility using the density of states of Ni, for two different values of the cutoff q_c. (a) $q_c = 0.77\ q_D$, (b) $q_c = 1/2\ q_D$, (c) the RPA results (with smaller values of U).

is unphysical and results because our approximation is insufficient to describe critical fluctuations.

V. CONCLUSIONS

We have presented a framework for an intermediate coupling theory of fluctuations in metallic paramagnets or ferromagnets above T_c. It gives a physical picture of how the RPA paramagnon propagator is renormalized by the fluctuations, and, for temperatures above the spin fluctuation temperature, a very simple interpretation in terms of a dynamically smeared effective density of states emerges if we make a local approximation to the paramagnon-paramagnon interaction vertices. This effective density of states allows us to begin to answer the question of the relationship between the Bloch states and the quasiparticle states of the phenomenological Stoner theory by giving us an explicit approximation to the latter as a functional of the former.

In the past, it has been suggested[16] that a reasonable Stoner theory could be obtained from Hartree-Fock band theory by simply replacing the interaction U by a smaller U_{eff}, to account for electron correlation effects of the sort discussed by Kanamori.[17] The validity of such an approach, however, depends on bare electrons being dressed by interactions with relatively few electron-hole pair excitations out of the Fermi sea, a condition grossly violated when there are many spin fluctuations excited either thermally ($T > T_{sf}$) or virtually ($T \ll T_{sf}$). Under these conditions, the present approach, leading to a temperature-dependent effective $N(E)$ instead of an effective U, makes more sense.

Our calculations of this effective density of states or of the effective Lindhard function $\overline{\varphi}(q)$ are only qualitatively correct because we made the local approximation for the mode-mode coupling vertices. It would be of interest to apply techniques used in other sorts of polaron problems to improve on these estimates.

Another aspect of this problem on which we have not concentrated here is the effect of the time dependence of the fluctuating fields on $\overline{\varphi}(q)$ and the paramagnon propagator. It is formally included in the discussion of Sec. II, but we have not tried to calculate $\overline{\varphi}$ except at sufficiently high temperatures that only the time-averaged components matter. An extension of the theory of Sec. III to temperatures below the spin fluctuation temperature is potentially important. It might permit us to understand why current paramagnon theory is only partially successful in explaining the effective mass enhancement in low-temperature nearly ferromagnetic Fermi systems. [Murata and Doniach[8,18] attempt to account for the dynamical part of the fluctuations by integrating out the ξ_q fields with characteristic frequencies greater than T, leaving renormalized coefficients of the remaining modes, which can then be treated in a static approximation. Their theory therefore contains a temperature-dependent cutoff wave vector which will in general be less than our q_c. Moriya and Kawabata have taken a different approach to the dynamical effects, based on a sum rule relating the susceptibility and the free energy.[19] Their theory contains no explicit cutoff, although an effective cutoff like that in the Murata-Doniach theory can occur via the Bose factor in frequency integrals. However, in both of these theories, the density of states is effectively approximated by its quadratic expansion around E_F. We stress that whenever E_F lies in a narrow peak in $N_0(E)$, or, more generally when $N_0(E)$ changes very rapidly near E_F, it is probably essential to take higher-order terms into account,

as we do here in Sec. III for the high-temperature case.]

Another important generalization of this work is to the ferromagnetic temperature region. We leave this, as well as the extensions suggested above, to future papers.

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