Ref. 8, we estimate that for the $Al\text{-}Al_2O_3\text{-}GeTe$ junctions σ^{-1} is approximately 0.45 eV and for $Al\text{-}Al_2O_3\text{-}SnTe$ junctions σ^{-1} is approximately 0.35 eV. For the $In\text{-}SrTiO_3$ junction used in Ref. 10, σ^{-1} is smaller.

If we consider a positive voltage applied to the normal metal so that tunneling occurs from the superconducting semiconductor into the metal, we have

$$j_{r\to l}=\int_{A(V)}^{B(V)}\frac{d\epsilon_{k'}}{Z(\epsilon_{k'})}\mathfrak{F}'(\epsilon_{k'})V(\xi)\,, \tag{35}$$

where $\mathfrak{F}'(\epsilon_{k'})$ is obtained from $\mathfrak{F}(\epsilon_{k'})$ by replacing Φ_e by $\Phi_{e'}=\frac{1}{2}(\Phi_l+\Phi_r-qV)$.

Expressions analogous to Eqs. (28) and (29) may be obtained for tunneling from the superconductor. We have, for example,

$$\begin{aligned} dj_{r\to l}/dV=&-q\mathfrak{F}'(B)\tfrac{1}{2}[1-V/(V^2-\Delta^2)^{1/2}]S_1\\ &+q\mathfrak{F}'(A)\tfrac{1}{2}[1+V/(V^2-\Delta^2)^{1/2}]S_2\\ &-\tfrac{1}{8}C\alpha q e^{-\alpha(\Phi_e'+\epsilon_F)^{1/2}}\left\{\frac{B-A}{Z}\right.\\ &\left.-\left[\left(\frac{B+\chi}{Z}\right)^2+\Delta^2\right]^{1/2}+\left[\left(\frac{A+\chi}{Z}\right)^2+\Delta^2\right]^{1/2}\right\}\\ &+I(V)' \end{aligned} \tag{36}$$

and

$$I(V)'=+\frac{C\alpha q}{4Z}\int_A^B d\epsilon\left[v\left(\frac{\epsilon+\chi}{Z}\right)\right]^2 e^{-\alpha(\Phi_e'-\epsilon)^{1/2}}, \tag{37}$$

which correspond to Eqs. (29) and (30). When $\epsilon_F\ll\Phi_a$ and $qV\ll\Phi_a$, $\mathfrak{F}'(\epsilon)$ can be obtained from Eq. (32) by replacing V by $-V$. Also, $I(V)'$ can be obtained from Eq. (33) by replacing V by $-V$ and $u(\xi)^2$ by $v(\xi)^2$.

The derivative of current with respect to voltage for tunneling into a low-carrier-density superconductor obtained from Eq. (29) with $Z=1$, $\chi=0$, and $\Delta=$ const is plotted as a function of voltage in Fig. 2. Tunneling from the superconductor into the normal metal using the same parameters in Eq. (36) is given in Fig. 3.

The decrease in conductance shown in Fig. 3 for all voltages shown may be contrasted with tunneling from a nonsuperconducting semiconductor into a metal, where the conductance increases when $qV>\epsilon_F$. The decreasing conductance shown in Fig. 3 arises from the superconducting interaction, specifically from the first term of Eq. (36), which is zero when $\Delta=0$.

V. CONCLUSIONS

The equations derived for dj/dV in Secs. III and IV may be used to determine the energy gap function from experimental tunneling curves in low-carrier-density superconductors. These equations may also be used in conjunction with equations for the energy gap[12] and knowledge of barrier dimensions to predict tunneling characteristics of metal-insulator-superconducting semiconductor junctions.

These equations, with slight modifications, may also be used to obtain the conductance for tunneling from a degenerate semiconductor into a metallic superconductor.

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Localized Impurity Spin Excitations in a Ferromagnetic Metal

T. Wolfram

Science Center, North American Rockwell Corporation, Thousand Oaks, California 91360

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The structure of the theory of impurity spin excitations in a one-band model of a ferromagnetic metal is examined. The existence of a local magnetic moment and an off-diagonal spin correlation in the impurity ground state is shown to be closely related to the occurrence of localized spin excitations (localized magnons). Expressions for both the transverse and longitudinal reduced susceptibility functions are obtained in terms of single-particle Green's functions. Poles of the reduced transverse susceptibility function are shown to correspond to local magnon states, and poles of the reduced longitudinal susceptibility are identified with fluctuations in the magnitude of the impurity moment. The role of localized electronic states is briefly discussed. Some of the well-known results for the unperturbed ferromagnet are derived in the Appendix.

I. INTRODUCTION

The nature of impurity spin excitations in magnetic insulators which may be described by the Heisenberg spin Hamiltonian were first studied in detail by Wolfram and Callaway[1] and more recently by others.[2,3] Spin-wave impurity states (localized magnons) outside of the host ferromagnetic spin-wave band, as well as virtual or resonance impurity states within the spin-wave band, may occur when the im-

[1] T. Wolfram and J. Callaway, Phys. Rev. **130**, 2207 (1963).

[2] S. Takeno, Progr. Theoret. Phys. (Kyoto) **30**, 731 (1963).

[3] D. Hone, H. Callen, and L. R. Walker, Phys. Rev. **144**, 283 (1966).

purity spin and exchange interaction differs from that of the host system. Comments by Jaccarino, Walker, and Wertheim[4] on ferromagnetic Fe samples with dilute concentrations of Mn stimulated calculations of the temperature dependence of the impurity magnetization by Hone and Callen[3] and also by Wolfram and Hall[5] who in addition showed that a spin-wave specific-heat anomaly should result when low-lying spin-wave impurity states occur.

We are concerned here with the question of how the above qualitative features of the impure insulating ferromagnet carry over to the case of the metallic ferromagnet. To date, no detailed analysis of localized spin excitations in ferromagnetic metals has been given, although, the problem has been briefly considered by Lederer[6] who pointed out that localized magnons should show up as additional poles in the transverse susceptibility function. In this paper we formulate a theory for the impure ferromagnetic metal and examine some of its properties which are relevant to the localized spin-excitation problem.

The theory of impurities in a ferromagnetic metal is very complex, and therefore very interesting, because of the rich structure of the problem. Many impurity effects, which have been studied individually, are simultaneously present in the impure ferromagnetic system and must be treated with equal respect. Questions concerning the existence of localized electronic states, localized magnetic moments, and localized spin canting in the impure ferromagnetic ground state are inseparable from the questions we wish to ask about the impurity spin excitations.

In principle, the problem is straightforward: calculate the susceptibility of the impure ferromagnetic system, identify the poles associated with the formation of localized spin excitations and determine the dependence of the localized excitation energy on the important parameters. The problem is greatly complicated by the fact that the proper impurity ground state, which depends upon the impurity perturbation, is not known *a priori* but must be calculated self-consistently. This leads to the situation described above in which the problem of the stability of the assumed ground state must be considered concomitantly with the excited spin-state problem. The connection between these two problems is a central feature of this paper.

We consider the Wolf model[7] for an impure ferromagnetic metal having a single band with an intra-atomic Coulomb repulsion between electrons occupying Wannier states of different spin on the same atomic site. The theory is developed in the Wannier representation using the generalized random phase approximation (RPA). We assume that the Hamiltonian describes a system whose ground corresponds to a spatially uniform ferromagnet in the absence of the impurity. The impurity is characterized by the perturbation in the Coulomb interaction ΔU and the core scattering potential V. In Sec. II, we first calculate by self-consistent perturbation theory the conditions for the existence of off-diagonal spin correlation (ODSC) in the impurity ground state. We obtain an equation which determines sets of values for ΔU and V corresponding to a transition curve in the $\Delta U-V$ plane. A similar equation is derived for the existence of a local magnetic moment (different from the moment of the ferromagnetic host). In Sec. III, expressions for the local reduced transverse and longitudinal susceptibility functions are derived in terms of the single-particle Green's functions. Equations are derived for the localized spin excitations and it is shown that the conditions for the existence of a low-lying spin excitation is directly related to the condition for the existence of either ODSC or local moment formation in the ground state. In particular, it is shown that the condition for a zero-energy local magnon is identical to the condition for the formation of ODSC or spin canting in the ground state. Similarly, the condition for the formation of a local magnetic moment is shown to be identical to the condition for a zero-energy pole in the longitudinal susceptibility. The role of the localized electronic state is briefly discussed. In Sec. IV, the results are briefly summarized and some of the important features of the unperturbed ferromagnetic metal are discussed in the Appendix. A brief discussion of some of the results in this paper has been given elsewhere.[8]

II. GROUND-STATE PROPERTIES

A. Hamiltonian

We consider the simple one-band strong-correlation model[7] for a ferromagnetic metal with an impurity atom at the lattice position $\mathbf{R}_0$

$$H=\sum_{ij\sigma} \mathcal{E}_{ij}C_{i\sigma}^\dagger C_{j\sigma}+\sum_i U_i n_{i\uparrow}n_{i\downarrow}+V\sum_\sigma n_{0\sigma}, \qquad (1)$$

where $C_{i\sigma}^\dagger$ ($C_{i\sigma}$) creates (destroys) an electron in a Wannier state at the lattice position $\mathbf{R}_i$ with spin σ ($\sigma=\uparrow,\downarrow$), and $n_{i\sigma}$ is the number operator for the Wannier state. The $\mathcal{E}_{ij}$ are the matrix elements of the single-particle Hamiltonian between Wannier states located at $\mathbf{R}_i$ and $\mathbf{R}_j$. These matrix elements are related to the band energy, $\epsilon_\mathbf{k}$, by the relation,

$$\mathcal{E}_{ij}=\frac{1}{N}\sum_\mathbf{k} \epsilon_\mathbf{k} e^{i\mathbf{k}\cdot(\mathbf{R}_j-\mathbf{R}_i)}. \qquad (2)$$

Single-particle scattering from the impurity core potential V is represented by the last term of Eq. (1). Elec-

[4] V. Jaccarino, L. R. Walker, and G. K. Wertheim, Phys. Rev. Letters **13**, 752 (1964).

[5] T. Wolfram and W. Hall, Phys. Rev. **143**, 284 (1966).

[6] P. Lederer, thesis, A. La Faculte Des Sciences De L'universite De Paris, 1967 (unpublished).

[7] P. A. Wolff, Phys. Rev. **124**, 1030 (1961).

[8] T. Wolfram J. Appl. Phys. (to be published).

trons with different spins experience a Coulomb repulsion U_i when they occupy Wannier states on the same lattice site $\mathbf{R}_i$. For the impurity system we have

$$\begin{aligned} U_i &= U, \qquad (i\neq 0) \\ &= U+\Delta U \quad (i=0). \end{aligned} \tag{3}$$

B. Single-Particle Green's Functions

The single-particle retarded Green's functions[9] are defined by

$$[\hat{G}_{\sigma,\sigma'}(t)]_{jk} = -i\theta(t)\langle[C_{j\sigma}(t),C_{k\sigma'}^\dagger(0)]_+\rangle, \tag{4}$$

where the brackets indicate the zero-temperature ($T=0$) expectation value of the enclosed operator for the impurity ground state of the system and $[A,B]_{+(-)}$ indicates the anticommutator (commutator) of the operators A and B. The function $\theta(t)$ is unity for $t>0$ and vanishes otherwise. We use a caret over the symbols for functions to indicate a matrix. We introduce the Fourier-transform Green's functions

$$[\hat{G}_{\sigma,\sigma'}(\omega)]_{jk} = (\tfrac{1}{2}\pi)\int_{-\infty}^{\infty} dt\, e^{i\omega t}[\hat{G}_{\sigma,\sigma'}(t)]_{jk}. \tag{5}$$

The equations of motion for the Green's functions are obtained in the usual manner.[9] Use of the generalized RPA yields the matrix equation,

$$\omega\hat{G}_{\sigma\sigma'}(\omega) = (\delta_{\sigma\sigma'}/2\pi)\hat{I} + (\hat{E}+\hat{P}_\sigma)\hat{G}_{\sigma\sigma'}(\omega) + \hat{M}_\sigma\hat{G}_{\bar{\sigma}\sigma'}(\omega), \tag{6}$$

where $\hat{G}_{\sigma\sigma'}(\omega)$ is an N by N matrix whose elements are $[\hat{G}_{\sigma\sigma'}(\omega)]_{jk}$, $\delta_{\sigma\sigma'}$ is a Kroneker δ function which is unity for $\sigma=\sigma'$ and vanishes otherwise, and we use $\bar{\sigma}$ to denote the spin state opposite to that of σ. The remaining matrices in Eq. (6) are defined as follows:

$$\begin{aligned} [\hat{E}]_{ij} &= \mathcal{E}_{ij}, \\ [\hat{P}_\sigma]_{ij} &= U_i\langle n_{i\bar{\sigma}}\rangle\delta_{ij} + V\delta_{0i}\delta_{0j}, \\ [\hat{M}_\sigma]_{ij} &= U_i\langle C_{i\sigma}C_{i\bar{\sigma}}^\dagger\rangle\delta_{ij}, \\ [\hat{I}]_{ij} &= \delta_{ij}. \end{aligned} \tag{7}$$

We shall refer to the quantity $\langle C_{i\sigma}C_{j\bar{\sigma}}^\dagger\rangle$ as the ODSC. The real and imaginary parts of the Green's functions matrix are defined by

$$\hat{G}_{\sigma\sigma'}(\omega) = (2\pi)^{-1}[\hat{R}_{\sigma\sigma'}(\omega) + i\pi\hat{N}_{\sigma\sigma'}(\omega)]. \tag{8}$$

The ground-state averages, $\langle n_{i\sigma}\rangle$ and $\langle C_{i\sigma}C_{i\bar{\sigma}}^\dagger\rangle$ are related at $T=0$ to the Green's function by the relation[9]

$$\langle C_{i\sigma'}^\dagger C_{i\sigma}\rangle = \int_{-\infty}^{\epsilon_f} d\omega[N_{\sigma\sigma'}(\omega)]_{ii}, \tag{9}$$

where ϵ_f is the Fermi energy. Thus in order to obtain $\hat{G}_{\sigma\sigma}(\omega)$, Eq. (6) must be solved self-consistently.

C. Off-Diagonal Spin Correlation

In this paper we assume that the Hamiltonian of Eq. (1) describes a system whose ground state, in the absence of the impurity, is a spatially uniform ferromagnetic state with $\langle n_{i\sigma}\rangle = n_\sigma$, and vanishing ODSC. The latter condition means that the local and bulk magnetization vectors are parallel to the spin-quantization axis and that there is no component of local or bulk magnetization perpendicular to the spin-quantization axis.

The conditions for which Eq. (6) has self-consistent solutions with a small but finite ODSC can be determined by means of a self-consistent perturbation calculation. There may exist critical values of V and ΔU which define a transition curve in the V-ΔU plane. On one side of this curve (region Ia) the ODSC exists, while on the other side (region IIa) the ODSC vanishes.[10] Let $\hat{G}_{\sigma\sigma'}{}^a(\omega)$ be the solution of Eq. (6) for V and ΔU corresponding to a point infinitesimally near the transition curve in region IIa. If V and ΔU are changed infinitesimally to give a point in region Ia we find the new Green's function to first order in the ODSC to be

$$\begin{aligned} \hat{G}_{\sigma\sigma}(\omega) &= \hat{G}_{\sigma\sigma}{}^a(\omega), \\ \hat{G}_{\sigma\bar{\sigma}}(\omega) &= 2\pi\hat{G}_{\sigma\sigma}{}^a(\omega)\hat{M}_\sigma\hat{G}_{\bar{\sigma}\bar{\sigma}}{}^a(\omega). \end{aligned} \tag{10}$$

If we impose self-consistency by means of Eq. (9), then we must have,

$$\langle C_{i\sigma}C_{i\bar{\sigma}}^\dagger\rangle = 4\,\mathrm{Im}\left\{\int_{-\infty}^{\epsilon_f} d\omega \sum_{jk} [\hat{G}_{\sigma\sigma}{}^a(\omega)]_{ij}[\hat{M}_\sigma]_{jk} \times[\hat{G}_{\bar{\sigma}\bar{\sigma}}{}^a(\omega)]_{ki}\right\}. \tag{11}$$

Substitution of the matrix elements of $\hat{M}_\sigma$ from Eq. (7) and some matrix algebra yields the matrix equation

$$\boldsymbol{\phi}_{\sigma\bar{\sigma}} = \Delta U(\hat{I} - U\hat{\boldsymbol{\lambda}}_{\sigma\bar{\sigma}})^{-1}\hat{\boldsymbol{\lambda}}_{\sigma\bar{\sigma}}\Delta\boldsymbol{\phi}_{\sigma\bar{\sigma}}, \tag{12}$$

where the N-element columns vectors $\boldsymbol{\phi}_{\sigma\bar{\sigma}}$ and $\Delta\boldsymbol{\phi}_{\sigma\bar{\sigma}}$ are given by

$$\boldsymbol{\phi}_{\sigma\bar{\sigma}} = \begin{bmatrix} \langle C_{0\sigma}C_{0\bar{\sigma}}^\dagger\rangle \\ \langle C_{1\sigma}C_{1\bar{\sigma}}^\dagger\rangle \\ \cdots \\ \langle C_{N\sigma}C_{N\bar{\sigma}}^\dagger\rangle \end{bmatrix} \tag{13}$$

and

$$\Delta\boldsymbol{\phi}_{\sigma\bar{\sigma}} = \begin{bmatrix} \langle C_{0\sigma}C_{0\bar{\sigma}}^\dagger\rangle \\ 0 \\ 0 \\ \cdots \\ 0 \end{bmatrix}. \tag{14}$$

[9] See, for example, D. N. Zubarev, Usp. Fiz. Nauk **71**, 71 (1960) [English transl.: Soviet Phys.—Usp. **3**, 320 (1960)].

[10] Transition curves for the various magnetically ordered states of the pure host are discussed by D. R. Penn, Phys. Rev. **142**, 350 (1966).

The matrix $\hat{\lambda}_{\sigma\bar{\sigma}}$ has elements

$$[\hat{\lambda}_{\sigma\bar{\sigma}}]_{ij}=-\int_{-\infty}^{\epsilon_f} d\omega\{[\hat{R}_{\sigma\sigma}{}^a(\omega)]_{ij}[\hat{N}_{\bar{\sigma}\bar{\sigma}}{}^a(\omega)]_{ji}+[\hat{R}_{\bar{\sigma}\bar{\sigma}}{}^a(\omega)]_{ji}[\hat{N}_{\sigma\sigma}{}^a(\omega)]_{ij}\}. \quad (15)$$

In a later section, we shall relate $\hat{\lambda}_{\sigma\bar{\sigma}}$ to the local reduced transverse susceptibility matrix. The components of Eq. (12) give the ODSC functions

$$\langle C_{i\sigma}C_{i\bar{\sigma}}{}^{\dagger}\rangle=\Delta U[(I-U\hat{\lambda}_{\sigma\bar{\sigma}})^{-1}\hat{\lambda}_{\sigma\bar{\sigma}}]_{i0}\langle C_{0\sigma}C_{0\bar{\sigma}}{}^{\dagger}\rangle. \quad (16)$$

For $i=0$, Eq. (16) requires that

$$1=\Delta U[(I-U\hat{\lambda}_{\sigma\bar{\sigma}})^{-1}\hat{\lambda}_{\sigma\bar{\sigma}}]_{00}, \quad (17)$$

in order that ODSC exist. The existence of ODSC for the ground state would imply that the magnetization is canted with respect to the unperturbed ferromagnetic ground state. The magnitude or $\langle C_{i\sigma}C_{i\bar{\sigma}}{}^{\dagger}\rangle$ is expected to be maximum at the impurity site and to decrease rapidly with increasing distance from the impurity.[11] We shall refer to Eq. (17) as the local spin-canting condition (LSCC). We are assuming here that a state in region Ia lies lower in energy than a state without the ODSC having the same values for V and ΔU. Although this assumption has not yet been established rigorously, we show in a later section that the converse leads to a situation in which the ground state is unstable to the formation of localized magnons.[12]

It should be noted that it is necessary to use the correct impurity ground state for $\hat{G}_{\sigma\sigma'}{}^a$ when investigating the LSCC. In the derivation of the LSCC no assumptions were made concerning the quantities $\langle n_{i\sigma}\rangle$. The ground state corresponding to $\hat{G}_{\sigma\sigma'}{}^a$ may or may not have uniform magnetization depending upon the values of V and ΔU.

D. Local Magnetic Moments

Next we turn our attention to the problem of determining the conditions under which the quantity $\langle n_{i\sigma}\rangle$ of the impurity system is nonuniform. We write the matrix elements of $\hat{P}_\sigma$ appearing in Eq. (7) in the form

$$[\hat{P}_\sigma]_{ij}=[\hat{P}_\sigma{}^b]_{ij}+[\Delta\hat{P}_\sigma]_{ij}, \quad (18)$$

where

$$\begin{aligned}[\hat{P}_\sigma{}^b]_{ij}&=(Un_{\bar{\sigma}}+V\delta_{0i})\delta_{ij}+\Delta Un_{\bar{\sigma}}\delta_{0i}\delta_{ij},\\ [\Delta\hat{P}_\sigma]_{ij}&=U\Delta n_{i\bar{\sigma}}\delta_{ij}+\Delta U\Delta n_{i\bar{\sigma}}\delta_{0i}\delta_{0j}.\end{aligned} \quad (19)$$

In Eq. (19), n_σ is the uniform value of $\langle n_{i\sigma}\rangle$ for the unperturbed ferromagnet and $\Delta n_{i\sigma}=\langle n_{i\sigma}\rangle-n_\sigma$ is impurity-induced change. There exist critical values for V and ΔU which define a transition curve. On one side of this curve (region Ib) $\Delta n_{i\sigma}$ is nonvanishing while on the other side (region IIb), $\Delta n_{i\sigma}=0$. Choose V and ΔU corresponding to a point infinitesimally near the transition curve in region IIb and let $\hat{G}_{\sigma\sigma'}{}^b(\omega)$ be the solution of Eq. (6) for these values of V and ΔU. If V and ΔU change infinitesimally, so that they now correspond to a point near the transition curve in region Ib, then the matrix elements of $\Delta\hat{P}_\sigma$ are small but finite, and first-order perturbation in $\Delta\hat{P}_\sigma$ yields the new Green's function

$$\begin{aligned}\hat{G}_{\sigma\bar{\sigma}}(\omega)&=\hat{G}_{\sigma\bar{\sigma}}{}^b(\omega),\\ \hat{G}_{\sigma\sigma}(\omega)&=\hat{G}_{\sigma\sigma}{}^b(\omega)\Delta\hat{P}_\sigma\hat{G}_{\sigma\sigma}{}^b(\omega)+\hat{G}_{\sigma\sigma}{}^b(\omega).\end{aligned} \quad (20)$$

Using Eq. (9) the following set of coupled equations for the change in the population factors is obtained:

$$-\Delta n_{i\sigma}=U\sum_j(\hat{\lambda}_\sigma)_{ij}\Delta n_{j\bar{\sigma}}+\Delta U(\hat{\lambda}_\sigma)_{i0}\Delta n_{0\bar{\sigma}}. \quad (21)$$

The matrix elements of $\hat{\lambda}_\sigma$ are given by

$$(\hat{\lambda}_\sigma)_{ij}=-\int_{-\infty}^{\epsilon_f} d\omega\{[\hat{R}_{\sigma\sigma}{}^b(\omega)]_{ij}[\hat{N}_{\sigma\sigma}{}^b(\omega)]_{ji}+[\hat{R}_{\sigma\sigma}{}^b(\omega)]_{ji}[\hat{N}_{\sigma\sigma}{}^b(\omega)]_{ij}\}. \quad (22)$$

We show in a later section that $\hat{\lambda}_\sigma$ is related to the local reduced longitudinal susceptibility of the system. Equation (21) may be written in the supermatrix form

$$\begin{pmatrix}\Delta\mathbf{n}_\uparrow\\ \Delta\mathbf{n}_\downarrow\end{pmatrix}=\begin{pmatrix}0 & -U\hat{\lambda}_\uparrow-\Delta U\Delta\hat{\lambda}_\uparrow\\ -U\hat{\lambda}_\downarrow-\Delta U\Delta\hat{\lambda}_\downarrow & 0\end{pmatrix}\times\begin{pmatrix}\Delta\mathbf{n}_\uparrow\\ \Delta\mathbf{n}_\downarrow\end{pmatrix}, \quad (23)$$

where the N-element column vectors $\Delta\mathbf{n}_\sigma$ are defined by

$$\Delta\mathbf{n}_\sigma=\begin{bmatrix}\Delta n_{0\sigma}\\ \Delta n_{1\sigma}\\ \cdots\\ \Delta n_{N\sigma}\end{bmatrix} \quad (24)$$

and

$$(\Delta\hat{\lambda}_\sigma)_{ij}=\delta_{0j}[\hat{\lambda}_\sigma]_{i0}. \quad (25)$$

In order to obtain nonzero values for the $\Delta n_{i\sigma}$ it is necessary that the determinant of the coefficients vanish;

$$\begin{vmatrix}I & U\hat{\lambda}_\uparrow+\Delta U\Delta\hat{\lambda}_\uparrow\\ U\lambda_\downarrow+\Delta U\Delta\hat{\lambda}_\downarrow & I\end{vmatrix}=0. \quad (26)$$

We may express this requirement in the form

$$\left|\begin{pmatrix}\hat{I} & 0\\ 0 & \hat{I}\end{pmatrix}+\Delta U\hat{A}^{-1}\begin{pmatrix}0 & \Delta\hat{\lambda}_\uparrow\\ \Delta\hat{\lambda}_\downarrow & 0\end{pmatrix}\right|=0, \quad (27)$$

where

$$\hat{A}=\begin{pmatrix}\hat{I} & \Delta\hat{\lambda}_\uparrow\\ U\hat{\lambda}_\downarrow & \hat{I}\end{pmatrix} \quad (28)$$

[11] This conjecture is based upon an approximate calculation of the function $\{[I-U\lambda_{\sigma\bar{\sigma}}]^{-1}\lambda_{\sigma\bar{\sigma}}\}_{i0}$ using an unperturbed ferromagnetic ground state.

[12] The author is indebted to L. M. Falicov for valuable suggestions concerning this point.

and

$$\hat{A}^{-1}=\begin{pmatrix} (\hat{I}-U^2\hat{\lambda}_\uparrow\hat{\lambda}_\downarrow)^{-1} & -U\hat{\lambda}_\uparrow(I-U^2\hat{\lambda}_\downarrow\hat{\lambda}_\uparrow)^{-1} \\ -U\hat{\lambda}_\downarrow(\hat{I}-U^2\hat{\lambda}_\uparrow\hat{\lambda}_\downarrow)^{-1} & (\hat{I}-U^2\hat{\lambda}_\downarrow\hat{\lambda}_\uparrow)^{-1} \end{pmatrix}. \tag{29}$$

Because of the simple form of $\Delta\hat{\lambda}_\sigma$ the determinant of Eq. (27) is easily evaluated, with the result that

$$\begin{aligned}\{1-U\Delta U[\hat{\lambda}_\uparrow(\hat{I}-U^2\hat{\lambda}_\downarrow\hat{\lambda}_\uparrow)^{-1}\hat{\lambda}_\downarrow]_{00}\}\\ \times\{\hat{I}-U\Delta U[\hat{\lambda}_\downarrow(\hat{I}-U^2\hat{\lambda}_\uparrow\hat{\lambda}_\downarrow)^{-1}\lambda_\uparrow]_{00}\}\\ -(\Delta U)^2[(\hat{I}-U^2\hat{\lambda}_\downarrow\hat{\lambda}_\uparrow)^{-1}\lambda_\downarrow]_{00}\\ \times[(\hat{I}-U^2\hat{\lambda}_\uparrow\hat{\lambda}_\downarrow)^{-1}\hat{\lambda}_\uparrow]_{00}=0.\end{aligned} \tag{30}$$

Equation (30) defines the transition curve for the existence of nonzero $\Delta n_{i\sigma}$ and also for the existence of local magnetic moments different from that of the host ferromagnet,

$$\langle n_{i\sigma}-n_{i\bar{\sigma}}\rangle\neq n_\sigma-n_{\bar{\sigma}}. \tag{31}$$

The magnitude of the local moment is expected to be maximum on the impurity atom and to decrease rapidly with increasing distance from the impurity site. We refer to Eq. (30) as the local-moment stability condition (LMSC). In the derivation of the LMSC no assumptions have been made concerning the ODSC. The V-ΔU plane may contain two transition curves; one corresponding to the LSCC and a second corresponding to the LMSC. These curves divide the V-ΔU plane into the following possible four regions; (i) no local moment exists and no ODSC exists, (ii) no local moments exist but ODSC does exist, (iii) a local moment exists but ODSC does not, and (iv) a local moment and ODSC both exist.

In a paramagnetic host the two transition curves coincide and the LSCC is simply a statement of the rotational invariance of the local moment. A ferromagnetic host is not isotropic since there is a preferred direction associated with the magnetization and, in general, the local-moment transition curve and the spin-canting transition curve are different.

E. Localized Perturbation

It is interesting to consider the form of the LSCC and the LMSC in limit that only the impurity site is perturbed. If we require that the ODSC vanish except at the impurity site then Eq. (12) yields the LSCC,

$$1=(U+\Delta U)[\hat{\lambda}_{\sigma\bar{\sigma}}]_{00}. \tag{32}$$

This condition is structurally the same as the usual local moment criterion for an impurity in a paramagnetic host.[13] Here $[\lambda_{\sigma\bar{\sigma}}]_{00}$ is related to the local transverse susceptibility for a band-split ferromagnet while in the paramagnetic case. $[\lambda_{\sigma\bar{\sigma}}]_{00}$ is the paramagnetic local susceptibility at the impurity site. Similarly the LMSC obtained from Eq. (21) is

$$1=(U+\Delta U)^2[\hat{\lambda}_\uparrow]_{00}[\hat{\lambda}_\downarrow]_{00}. \tag{33}$$

In a paramagnetic system $\hat{\lambda}_\uparrow=\hat{\lambda}_\downarrow=\lambda_{\sigma\bar{\sigma}}$, and proper solution of Eq. (33) coincides with that of Eq. (32). In Eq. (33) for the ferromagnetic system $\hat{\lambda}_\sigma$ and $\hat{\lambda}_{\bar{\sigma}}$ are the components of the local longitudinal susceptibility matrix as will be shown in Sec. III.

III. LOCAL REDUCED SUSCEPTIBILITY FUNCTIONS

In this section, we calculate expressions for the local reduced transverse susceptibility matrix $\hat{\chi}(\omega)$ and the longitudinal susceptibility matrix $\hat{\chi}_z(\omega)$. We show that the functions $\hat{\lambda}_{\sigma\bar{\sigma}}$ and λ_σ of Sec. II are related to the transverse and longitudinal susceptibility function $\hat{\chi}(\omega)$ and $\hat{\chi}_z(\omega)$ at zero energy. The impurity spin excitations may be associated with the occurrence of new poles or structure in the susceptibility functions. We show that the existence of low-energy impurity spin excitations is closely related to the instability of the impurity ground state to the formation of local spin canting and/or local magnetic moment formation. In fact, it is shown that condition for the formation of a zero-frequency localized magnon is identical with the LSCC. New poles may also appear in the longitudinal susceptibility corresponding to fluctuations in the magnitude of the impurity moment. We refer to these modes as polar spin excitations. The condition for the occurrence of a zero-frequency polar spin excitation is shown to be identical with the LMSC.

A. Local Reduced Transverse Susceptibility

We begin by examining the properties of the two-particle Green's-function matrix $\hat{S}(t)$ defined by[14]

$$[\hat{S}(t)]_{ijkl}=-i\theta(t)\langle[C_{i\uparrow}^\dagger(t)C_{j\downarrow}(t),C_{k\downarrow}^\dagger(0)C_{l\uparrow}(0)]_-\rangle. \tag{34}$$

The Fourier transform $\hat{S}(\omega)$ defined by

$$\hat{S}(\omega)=(\tfrac{1}{2}\pi)\int_{-\infty}^{\infty}dt\,\hat{S}(t)e^{i\omega t} \tag{35}$$

satisfies in the generalized RPA, the matrix equation,

$$\omega\hat{S}(\omega)=(\hat{K}+\hat{W}+\hat{J}+\hat{j})\hat{S}(\omega)+\hat{D}, \tag{36}$$

if the ODSC vanishes for the ground state. The matrices

[13] See, for example, D. L. Mills and P. Lederer, Phys. Rev. **160**, 590 (1967).

[14] These functions are the Wannier components of the momentum space (Bloch representation) operators $[C_{k+q\uparrow}^\dagger(t)C_{k\downarrow}(t), C_{k'}^\dagger(0)C_{k'+q'}(0)]$ usually studied; see, for example, Refs. 13 and 15.

have the following elements:

$$
\begin{aligned}
[\hat{K}]_{ijkl} &= \mathcal{E}_{lj}\delta_{ik} - \mathcal{E}_{ik}\delta_{lj}\,, \\
[\hat{W}]_{ijkl} &= [\hat{V}_\downarrow]_{ijkl} - [\hat{V}_\uparrow]_{ijkl}\,, \\
[\hat{V}_\uparrow]_{ijkl} &= \delta_{ik}\delta_{lj}\{U\langle n_{j\downarrow}\rangle + [\Delta U\langle n_{0\downarrow}\rangle + V]\delta_{0j}\}\,, \\
[\hat{V}_\downarrow]_{ijkl} &= \delta_{ik}\delta_{lj}\{U\langle n_{i\uparrow}\rangle + [\Delta U\langle n_{0\uparrow}\rangle + V]\delta_{0i}\}\,, \\
[\hat{J}]_{ijkl} &= U\delta_{kl}\{\delta_{ik}\langle C_{i\downarrow}{}^\dagger C_{j\downarrow}\rangle - \delta_{jl}\langle C_{i\uparrow}{}^\dagger C_{j\uparrow}\rangle\}\,, \\
[\hat{j}]_{ijkl} &= \Delta U\delta_{kl}\{\delta_{ik}\delta_{i0}\langle C_{0\downarrow}{}^\dagger C_{j\downarrow}\rangle - \delta_{jl}\delta_{0j}\langle C_{i\uparrow}{}^\dagger C_{0\uparrow}\rangle\}\,, \\
[\hat{D}]_{ijkl} &= (\tfrac{1}{2}\pi)\{\delta_{jk}\langle C_{i\uparrow}{}^\dagger C_{l\uparrow}\rangle - \delta_{il}\langle C_{k\downarrow}{}^\dagger C_{j\downarrow}\rangle\}\;.
\end{aligned} \tag{37}
$$

In Eq. (36) matrix multiplication is defined such that if $\hat{A}=\hat{B}\hat{C}$, then

$$[\hat{A}]_{ijkl}=\sum_{mn}[\hat{B}]_{ijmn}[\hat{C}]_{mnkl}\,, \tag{38}$$

and the unit matrix has components

$$[\hat{I}]_{ijkl}=\delta_{ik}\delta_{jl}. \tag{39}$$

We are concerned here with the calculation of the local reduced suceptibility functions.[15] The local reduced transverse susceptibility matrix, $\hat{\chi}(\omega)$, is an $N\times N$ matrix whose elements are derived from $\hat{S}(\omega)$ by means of the relation

$$[\hat{\chi}(\omega)]_{ij}=-2\pi[S(\omega)]_{iijj}. \tag{40}$$

Our purpose in this section is to express these functions in terms of the single-particle Green's functions introduced in Sec. II. We accomplish this task by a generalization of the procedure used by Mills and Lederer[13] in their treatment of the impure paramagnetic system.

Let $\hat{S}^{(1)}(\omega)$ be the solution of Eq. (36) in the absence of the matrices $\hat{J}$ and $\hat{j}$, then we find that

$$\hat{S}(\omega)=\hat{S}^{(1)}(\omega)+\hat{S}^{(1)}(\omega)(\hat{F}+\Delta\hat{F})\hat{S}(\omega)\,, \tag{41}$$

where the matrixes $\hat{F}$ and $\Delta\hat{F}$ have elements

$$
\begin{aligned}
[\hat{F}]_{ijkl} &= -2\pi U\delta_{il}\delta_{jk}\delta_{ik}\,, \\
[\Delta\hat{F}]_{ijkl} &= -2\pi\Delta U\delta_{il}\delta_{jk}\delta_{0i}\delta_{0j}.
\end{aligned} \tag{42}
$$

Using Eqs. (41) and (42) yields the result

$$
\begin{aligned}
[\hat{S}(\omega)]_{ijkl} = {} & [\hat{S}^{(1)}(\omega)]_{ijkl} - 2\pi U\sum_m [\hat{S}^{(1)}(\omega)]_{ijmm} \\
& \times[\hat{S}(\omega)]_{mmkl} - 2\pi\Delta U[\hat{S}^{(1)}(\omega)]_{ij00}[\hat{S}(\omega)]_{00kl}\,,
\end{aligned} \tag{43}
$$

so that with Eq. (40) one obtains

$$\hat{\chi}(\omega)=\hat{\chi}^{(1)}(\omega)+U\hat{\chi}^{(1)}(\omega)\hat{\chi}(\omega)+\Delta U\Delta\hat{\chi}^{(1)}(\omega)\hat{\chi}(\omega). \tag{44}$$

In Eq. (44),

$$[\hat{\chi}^{(1)}(\omega)]_{ij}=-2\pi[\hat{S}^{(1)}(\omega)]_{iijj} \tag{45}$$

and

$$[\Delta\hat{\chi}^{(1)}(\omega)]_{ij}=[\hat{\chi}^{(1)}(\omega)]_{i0}\delta_{0j}. \tag{46}$$

[15] For a discussion of the unperturbed ferromagnetic and the reduced susceptibility functions see T. Izuyama, D. J. Kim, and R. Kubo, J. Phys. Soc. Japan 18, 1025 (1963).

Equation (35) is easily solved and gives the result that

$$
\begin{aligned}
[\hat{\chi}(\omega)]_{ij} = [\hat{K}(\omega)]_{ij} + (\Delta U)^2\{[\hat{K}(\omega)]_{i0}[\hat{K}(\omega)]_{0j}\}/ \\
\{1-\Delta U[\hat{K}(\omega)]_{00}\}\,,
\end{aligned} \tag{47}
$$

where

$$[\hat{K}(\omega)]_{ij}=\{[I-U\hat{\chi}^{(1)}(\omega)]^{-1}\hat{\chi}^{(1)}(\omega)\}_{ij}. \tag{48}$$

The matrix $\hat{\chi}^{(1)}(\omega)$ is the transverse susceptibility matrix for a system of noninteracting electrons moving in the effective one-particle spin-dependent potential $\hat{W}$. The susceptibility function $\hat{K}(\omega)$, includes the host exchange terms contained in $\hat{J}$. The poles of $[I-U\hat{\chi}^{(1)}(\omega)]^{-1}$ (see Appendix) correspond to spin waves perturbed by the impurity part of $\hat{W}$ and also by the effect of the entire impurity perturbation on the ground state of the system. The structure of $\hat{K}(\omega)$ is similar to that obtained for the susceptibility of an exchange-enhanced paramagnetic system.[13]

From Eq. (47) we find the local reduced transverse susceptibility of the impurity site to be

$$[\hat{\chi}(\omega)]_{00}=[\hat{K}(\omega)]_{00}/\{1-\Delta U[\hat{K}(\omega)]_{00}\}\,, \tag{49}$$

which has the same form as impurity exchange enhancement in a paramagnetic system.[13]

Next we express $\hat{\chi}^{(1)}(\omega)$ in terms of the single-particle Green's functions of Sec. II. We find that

$$
\begin{aligned}
[\hat{S}^{(1)}(t)]_{ijkl} \Rightarrow {} & \langle C_{i\uparrow}{}^\dagger(t)C_{l\downarrow}(0)\rangle[\hat{G}_{\downarrow\downarrow}(t)]_{jk} \\
& + \langle C_k{}^\dagger(0)C_j(t)\rangle[G_{\uparrow\uparrow}{}^*(t)]_{il}\,,
\end{aligned} \tag{50}
$$

where the symbol $\Rightarrow$ indicates that the function on the right-hand side of Eq. (50) satisfies the same matrix equation as the function on the left-hand side in the generalized RPA .The Fourier transform functions are given by

$$
\begin{aligned}
\mathrm{Re}[\hat{S}^{(1)}(\omega)]_{ijkl} = \frac{1}{2\pi}\int_{-\infty}^{\epsilon_f} d\Omega\{ & [\hat{N}_{\uparrow\uparrow}(\Omega)]_{li}[\hat{R}_{\downarrow\downarrow}(\Omega+\omega)]_{jk} \\
& + [\hat{N}_{\downarrow\downarrow}(\Omega)]_{jk}[\hat{R}_{\uparrow\uparrow}(\Omega-\omega)]_{li}\}
\end{aligned} \tag{51}
$$

and

$$
\begin{aligned}
\mathrm{Im}[\hat{S}^{(1)}(\omega)]_{ijkl} = \frac{1}{2}\int_{-\infty}^{\epsilon_f} d\Omega\{ & [\hat{N}_{\uparrow\uparrow}(\Omega)]_{li}[\hat{N}_{\downarrow\downarrow}(\Omega+\omega)]_{jk} \\
& - [\hat{N}_{\downarrow\downarrow}(\Omega)]_{jk}[\hat{N}_{\uparrow\uparrow}(\Omega-\omega)]_{li}\}\,.
\end{aligned} \tag{52}
$$

It is easily verified that the real and imaginary parts of $\hat{S}^{(1)}(\omega)$ satisfy the Kromers-Kronig relation

$$\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\omega'}{\omega-\omega'}\mathrm{Im}\hat{S}^{(1)}(\omega')=\mathrm{Re}\hat{S}^{(1)}(\omega)\,, \tag{53}$$

since

$$[\hat{R}_{\sigma\sigma'}(\omega)]_{ij}=\int_{\infty}^{\infty}d\omega'\frac{[N_{\sigma\sigma'}(\omega')]_{ij}}{\omega-\omega'}\,, \tag{54}$$

with the convention that ω possesses an infinitesimal negative imaginary part.

B. Localized Magnons

The transverse spin excitations of the impurity system correspond to the poles of the transverse susceptibility matrix $\hat{\chi}(\omega)$. In general, there are three types of such excitations. First, are the Stoner excitations[15] involving the transfer of an electron from a Bloch state with momentum $\mathbf{k}$ in one spin band to a Bloch state with momentum $\mathbf{k}+\mathbf{q}$ in the other spin band. These states are separated from the ground state by an energy approximately equal to the band-splitting energy Δ for $\mathbf{q}=0$. Secondly, there exist bulk-type spin-wave states with energy $\omega_q \propto q^2$ for small q. At large $\mathbf{q}$ the spin waves intersect the Stoner excitation band and become highly damped due to decay into Stoner excitations.[15,16] Last, but of central importance here, are the localized impurity excitations or localized magnon excitations. These excitations correspond to nonpropagating spin modes in which the amplitude of excitation is maximum at (or near) the impurity site and decreases rapidly with increasing distance from the impurity site.[1,5]

For a large system in which the Bloch momentum $\mathbf{k}$ is considered to be a continuous variable only the poles associated with the impurity localized magnons remain in the transverse susceptibility functions. The bulk spin waves and the Stoner excitations are characterized by densities of states and give imaginary contributions to the transverse susceptibility functions. The lifetime of the localized magnons are limited by decay into both bulk magnon states as well as Stoner excitations. The poles of $\hat{\chi}(\omega)$ associated with the localized magnons are according to Eq. (49), determined by the condition

$$1=\Delta U[\hat{K}(\omega_L)]_{00}, \tag{55}$$

where ω_L is the energy for which Eq. (55) is satisfied. If such a pole exists it indicates that the impurity system possesses a new spin excitation eigenstate. The new state is a localized magnon state in which the amplitude of spin excitation is maximum at the impurity site and decreases rapidly with increasing distance from the impurity site. If $\omega_L=0$, then Eq. (55) gives

$$1=\Delta U[\{I-U\hat{\chi}^{(1)}(0)\}^{-1}\hat{\chi}^{(1)}(0)]_{00}. \tag{56}$$

It follows from Eq. (52) that $\mathrm{Im}\{\hat{\chi}^{(1)}(0)\}$ vanishes and from Eq. (51) we see that

$$\mathrm{Re}\chi^{(1)}(0)=\hat{\lambda}_{\sigma\bar{\sigma}}, \tag{57}$$

so that the condition [Eq. (17)] for the existence of ODSC in the ground state is identical to the condition for the existence of a zero-frequency localized magnon. This result also shows what was stated in Sec. II, namely, that if the LSCC is satisfied, then a ground state without ODSC is unstable to the formation of localized magnons. The localized magnon states necessarily possess ODSC and the zero-frequency state is degenerate with the ground state. Consequently, it it appears that in region Ia (see Sec. II C) the proper ground state will possess ODSC. Low-energy local magnons are expected when the ground state is nearly unstable to the formation of the ODSC.

[16] D. C. Mattis, Phys. Rev. **132**, 2521 (1963).

C. Local Reduced Longitudinal Susceptibility

We now turn our attention to the problem of calculating the longitudinal susceptibility. We proceed in a manner similar to that used for the transverse susceptibility. Expressions for the elements of the local reduced longitudinal susceptibility matrix are derived in terms of the single-particle Green's functions. It is established that the condition for a zero-frequency pole in the longitudinal susceptibility is identical with the LMSC derived in Sec. II.

We consider the two-particle Green's-function matrix $\hat{S}_z(t)$,

$$[\hat{S}_z(t)]_{ijkl}=[\hat{S}_\uparrow(t)]_{ijkl}-[\hat{S}_\downarrow(t)]_{ijkl}, \tag{58}$$

where

$$[\hat{S}_\sigma(t)]_{ijkl}=-i\theta(t)\langle[C_{i\sigma}{}^\dagger C_{j\sigma},\, C_{k\uparrow}{}^\dagger C_{l\uparrow}-C_{k\downarrow}{}^\dagger C_{l\downarrow}]_-\rangle. \quad (59$$

The local longitudinal susceptibility functions $[\hat{\chi}_z(\omega)]_{ij}$ are defined by[15]

$$[\hat{\chi}_z(\omega)]_{ij}=\tfrac{1}{4}\sum_\sigma [\hat{\chi}_\sigma(\omega)]_{ij}, \tag{60}$$

where

$$[\chi_\sigma(\omega)]_{ij}=-2\pi\eta_\sigma[\hat{S}_\sigma(\omega)]_{iijj}. \tag{61}$$

In Eq. (61), $\hat{S}_\sigma(\omega)$ is the Fourier transform of $\hat{S}_\sigma(t)$ and η_σ is $+1$ (-1) for spin $\uparrow$ $(\downarrow)$. In the RPA, we obtain the supermatrix equation

$$\omega\begin{pmatrix}\hat{S}_\uparrow(\omega)\\ \hat{S}_\downarrow(\omega)\end{pmatrix}=\begin{pmatrix}(\hat{K}+\hat{W}_\uparrow) & (\hat{J}_\uparrow+\hat{j}_\uparrow)\\ (\hat{J}_\downarrow+\hat{j}_\downarrow) & (\hat{K}+\hat{W}_\downarrow)\end{pmatrix}\begin{pmatrix}\hat{S}_\uparrow(\omega)\\ \hat{S}_\downarrow(\omega)\end{pmatrix}+\frac{1}{2\pi}\begin{pmatrix}\hat{D}_\uparrow\\ \hat{D}_\downarrow\end{pmatrix}, \tag{62}$$

where the matrices are defined by

$$\begin{aligned}
[\hat{W}_\sigma]_{ijkl}&=\delta_{ik}\delta_{jl}\{U(\langle n_{j\bar{\sigma}}\rangle-\langle n_{i\bar{\sigma}}\rangle)\\
&\qquad+(V+\Delta U\langle n_{0\bar{\sigma}}\rangle)(\delta_{0j}-\delta_{0i})\},\\
[\hat{J}_\sigma]_{ijkl}&=U\langle C_{i\sigma}{}^\dagger C_{j\sigma}\rangle\delta_{kl}(\delta_{jl}-\delta_{ik}),\\
[\hat{j}_\sigma]_{ijkl}&=\Delta U\langle C_{i\sigma}{}^\dagger C_{j\bar{\sigma}}\rangle\delta_{kl}(\delta_{jl}\delta_{0j}-\delta_{ik}\delta_{i0}),\\
[\hat{D}_\sigma]_{ijkl}&=(\eta_\sigma/2\pi)\{\langle C_{i\sigma}{}^\dagger C_{l\sigma}\rangle\delta_{jk}-\langle C_{k\sigma}{}^\dagger C_{j\sigma}\rangle\delta_{il}\}.
\end{aligned} \tag{63}$$

If the function $\hat{S}_\sigma{}^{(1)}(\omega)$ satisfies Eq. (62) in the absence of the matrices $\hat{J}_\sigma$ and $\hat{j}_\sigma$, then

$$\hat{S}_\sigma(\omega)=\hat{S}_\sigma{}^{(1)}(\omega)+\hat{S}_\sigma{}^{(1)}(\omega)\hat{A}_\sigma\hat{S}_{\bar{\sigma}}{}^{(1)}(\omega), \tag{64}$$

(note $\bar{\sigma}$ in the equation) where the matrix $\hat{A}_\sigma$ is defined by

$$[\hat{A}_\sigma]_{ijkl}=2\pi\delta_{ij}\delta_{jk}\delta_{kl}\eta_\sigma(U+\Delta U\delta_{i0}). \tag{65}$$

Equation (64) leads to the result that

$$[\hat{S}_\sigma(\omega)]_{ijkl}=[\hat{S}_\sigma{}^{(1)}(\omega)]_{ijkl}+2\pi U\eta_\sigma\sum_m[\hat{S}_\sigma{}^{(1)}(\omega)]_{ijmm}\times[\hat{S}_{\bar{\sigma}}(\omega)]_{mmkl}+2\pi\Delta U\eta_\sigma[\hat{S}_\sigma{}^{(1)}(\omega)]_{ij00}\times[\hat{S}_\sigma(\omega)]_{00kl}. \quad (66)$$

Using Eq. (61) we find the matrix equation for the reduced longitudinal susceptibility matrix,

$$\hat{\chi}_\sigma(\omega)=\hat{\chi}_\sigma{}^{(1)}(\omega)+U\hat{\chi}_\sigma{}^{(1)}(\omega)\hat{\chi}_{\bar{\sigma}}(\omega)+\Delta U\Delta\hat{\chi}^{(1)}(\omega)\hat{\chi}_{\bar{\sigma}}(\omega)\,, \quad (67)$$

where

$$\begin{aligned}[\hat{\chi}^{(1)}(\omega)]_{ij}&=-2\pi\eta_\sigma[\hat{S}^{(1)}(\omega)]_{iijj}\,,\\ [\Delta\hat{\chi}_\sigma{}^{(1)}(\omega)]_{ij}&=[\hat{\chi}_\sigma{}^{(1)}(\omega)]_{i0}\delta_{0j}.\end{aligned} \quad (68)$$

We note that Eq. (67) is similar in form to Eq. (23) relating to the local moment criterion. We solve Eq. (67) for $\hat{\chi}_\sigma(\omega)$ and obtain

$$\begin{pmatrix}\hat{\chi}_\uparrow(\omega)\\ \hat{\chi}_\downarrow(\omega)\end{pmatrix}=\left\{I-\Delta U\hat{B}^{-1}\begin{pmatrix}0 & \Delta\hat{\chi}_\uparrow{}^{(1)}(\omega)\\ \Delta\hat{\chi}_\downarrow{}^{(1)}(\omega) & 0\end{pmatrix}\right\}^{-1}\hat{B}^{-1}\times\begin{pmatrix}\hat{\chi}_\uparrow{}^{(1)}(\omega)\\ \hat{\chi}_\downarrow{}^{(1)}(\omega)\end{pmatrix}, \quad (69)$$

where $\hat{B}^{-1}(\omega)$ is obtained from $\hat{A}^{-1}$ [see Eq. (29)] by replacing λ_σ by $\hat{\chi}_\sigma{}^{(1)}(\omega)$ and also replacing U by $-U$. Therefore, it follows that the perturbed poles of $\hat{\chi}_\sigma(\omega)$ are determined by Eq. (30) if the same replacement of symbols is made. In order to show that the condition for a zero-frequency pole in the longitudinal susceptibility is identical to the LMSC it is now only necessary to show that

$$\hat{\chi}_\sigma{}^{(1)}(\omega)\,|_{\omega=0}=\lambda_\sigma. \quad (70)$$

We proceed as in the treatment of the transverse susceptibility. It is easily verified (in the generalized RPA) that the function

$$\langle C_{i\sigma}{}^\dagger(t)C_{l\sigma}(0)\rangle[G_{\sigma\sigma}(t)]_{jk}+\langle C_{k\sigma}{}^\dagger(0)C_{j\sigma}(t)\rangle[G_{\sigma\sigma}{}^*(t)]_{il} \quad (71)$$

satisfies the same matrix equation as does the function

$$\eta_\sigma[\hat{S}_\sigma{}^{(1)}(t)]_{ijkl}. \quad (72)$$

Thus, we find in this approximation

$$\mathrm{Re}\{\eta_\sigma[\hat{S}_\sigma{}^{(1)}(\omega)]_{ijkl}\}=\frac{1}{2\pi}\int_{-\infty}^{\epsilon_f}d\Omega\{[\hat{N}_{\sigma\sigma}(\Omega)]_{il}\times[\hat{R}_{\sigma\sigma}(\Omega+\omega)]_{jk}+[N_{\sigma\sigma}(\Omega)]_{jk}[\hat{R}_{\sigma\sigma}(\Omega-\omega)]_{li}\}\,, \quad (73)$$

and

$$\mathrm{Im}\{\eta_\sigma[S_\sigma{}^{(1)}(\omega)]_{ijkl}\}=\frac{1}{2}\int_{-\infty}^{\epsilon_f}d\Omega\{[N_{\sigma\sigma}(\Omega)]_{il}\times[N_{\sigma\sigma}(\Omega+\omega)]_{ij}-[N_{\sigma\sigma}(\Omega)]_{jk}[N_{\sigma\sigma}(\Omega-\omega)]_{li}\}\,. \quad (74)$$

The real and imaginary parts of $\hat{S}^{(1)}(\omega)$ are also related by the Kramers-Kronig relation. For $\omega=0$, the imaginary part of $\hat{S}_\sigma{}^{(1)}(\omega)$ vanishes and it is easily verified that

$$\hat{\chi}_\sigma(\omega)\,|_{\omega=0}=\lambda_\sigma. \quad (75)$$

This establishes the result that an $\omega=0$ pole in the longitudinal susceptibility is associated with the formation of a local magnetic moment in the impure ferromagnetic system.

We now develop further the expression for the local reduced longitudinal susceptibility given in Eq. (69). The elements of the supermatrix $\hat{B}^{-1}(\omega)$ are

$$\hat{B}^{-1}(\omega)=\begin{pmatrix}\hat{B}_{\uparrow\uparrow}{}^{-1} & \hat{B}_{\uparrow\downarrow}{}^{-1}\\ \hat{B}_{\downarrow\uparrow}{}^{-1} & \hat{B}_{\downarrow\downarrow}{}^{-1}\end{pmatrix}=\begin{pmatrix}(\hat{I}-U^2\hat{\chi}_\uparrow{}^{(1)}\hat{\chi}_\downarrow{}^{(1)})^{-1} & U\hat{\chi}_\uparrow{}^{(1)}(\hat{I}-U^2\hat{\chi}_\downarrow{}^{(1)}\hat{\chi}_\uparrow{}^{(1)})^{-1}\\ \hat{\chi}_\downarrow{}^{(1)}(\hat{I}-U^2\hat{\chi}_\uparrow{}^{(1)}\hat{\chi}_\downarrow{}^{(1)})^{-1} & (\hat{I}-U^2\hat{\chi}_\downarrow{}^{(1)}\hat{\chi}_\uparrow{}^{(1)})^{-1}\end{pmatrix}. \quad (76)$$

(For simplicity we shall omit the argument ω in some of the complicated expressions.) If we define

$$\hat{L}(\omega)=\begin{pmatrix}\hat{L}_{\uparrow\uparrow}(\omega) & \hat{L}_{\downarrow\uparrow}(\omega)\\ \hat{L}_{\downarrow\uparrow}(\omega) & \hat{L}_{\downarrow\downarrow}(\omega)\end{pmatrix}=\hat{I}-\Delta U\hat{B}(\omega)^{-1}\times\begin{pmatrix}0 & \Delta\hat{\chi}_\uparrow{}^{(1)}(\omega)\\ \Delta\hat{\chi}_\downarrow{}^{(1)}(\omega) & 0\end{pmatrix}, \quad (77)$$

then we find

$$[\mathfrak{L}_{\sigma\sigma}{}^{-1}(\omega)]_{ij}=\delta_{ij}-\Delta U\delta_{0j}\{[\hat{B}_{\sigma\sigma}{}^{-1}\hat{\chi}_\sigma{}^{(1)}]_{i0}[\hat{B}_{\bar{\sigma}\bar{\sigma}}{}^{-1}\hat{\chi}_{\bar{\sigma}}{}^{(1)}]_{00}-[\hat{B}_{\sigma\bar{\sigma}}{}^{-1}\hat{\chi}_{\bar{\sigma}}{}^{(1)}]_{i0}(1+[\hat{B}_{\bar{\sigma}\sigma}{}^{-1}\hat{\chi}_\sigma{}^{(1)}]_{00})\}\{\det\mathfrak{L}\}^{-1} \quad (78)$$

and

$$[\mathfrak{L}_{\sigma\bar{\sigma}}{}^{-1}(\omega)]_{ij}=-\Delta U\delta_{0j}\{[\hat{B}_{\sigma\bar{\sigma}}{}^{-1}\hat{\chi}_{\bar{\sigma}}{}^{(1)}]_{i0}[\hat{B}_{\sigma\sigma}{}^{-1}\hat{\chi}_\sigma{}^{(1)}]_{00}-[\hat{B}_{\sigma\sigma}\hat{\chi}_\sigma{}^{(1)}]_{i0}(1+[\hat{B}_{\sigma\bar{\sigma}}{}^{-1}\hat{\chi}_{\bar{\sigma}}{}^{(1)}]_{00})\}\{\det\mathfrak{L}\}^{-1}, \quad (79)$$

where

$$\det\mathfrak{L}=(1-\Delta U[\hat{B}_{\uparrow\downarrow}{}^{-1}\hat{\chi}_\downarrow{}^{(1)}]_{00})(1-\Delta U[\hat{B}_{\downarrow\uparrow}{}^{-1}\hat{\chi}_\uparrow{}^{(1)}]_{00})-(\Delta U)^2[\hat{B}_{\uparrow\uparrow}{}^{-1}\hat{\chi}_\uparrow{}^{(1)}]_{00}[\hat{B}_{\downarrow\downarrow}{}^{-1}\hat{\chi}_\downarrow{}^{(1)}]_{00}. \quad (80)$$

Finally, using Eq. (69), we obtain

$$[\chi_\sigma(\omega)]_{ij}=[\hat{L}_{\sigma\sigma}{}^{-1}(\omega)]_{i0}[\hat{\chi}_\sigma{}^{(2)}(\omega)]_{0j}+[\hat{\chi}_\sigma{}^{(2)}(\omega)]_{ij}\times(1-\delta_{0i})+[\hat{L}_{\sigma\bar{\sigma}}{}^{-1}(\omega)]_{i0}[\hat{\chi}_{\bar{\sigma}}{}^{(2)}(\omega)]_{0j}, \quad (81)$$

where

$$\begin{pmatrix}\hat{\chi}_\uparrow{}^{(2)}(\omega)\\ \hat{\chi}_\downarrow{}^{(2)}(\omega)\end{pmatrix}=\hat{B}^{-1}(\omega)\begin{pmatrix}\hat{\chi}_\uparrow{}^{(1)}(\omega)\\ \hat{\chi}_\downarrow{}^{(1)}(\omega)\end{pmatrix}. \quad (82)$$

The functions $\hat{\chi}_\sigma{}^{(2)}(\omega)$ are the components of the local reduced longitudinal susceptibility matrix which satisfies Eq. (62) in the absence of the impurity-exchange perturbation matrix $\hat{J}_\sigma$. The components at the impurity site may be obtained from the two-by-two matrix equation

$$\begin{pmatrix}[\hat{\chi}_\uparrow(\omega)]_{00}\\ [\hat{\chi}_\downarrow(\omega)]_{00}\end{pmatrix}=\begin{pmatrix}[\hat{L}_{\uparrow\uparrow}{}^{-1}]_{00}[\hat{L}_{\uparrow\downarrow}{}^{-1}]_{00}\\ [\hat{L}_{\downarrow\uparrow}{}^{-1}]_{00}[\hat{L}_{\downarrow\downarrow}{}^{-1}]_{00}\end{pmatrix}\times\begin{pmatrix}[\hat{\chi}_\uparrow{}^{(2)}]_{00}\\ [\hat{\chi}_\downarrow{}^{(2)}]_{00}\end{pmatrix}. \quad (83)$$

D. Localized Longitudinal Excitations

According to Eq. (80), the reduced longitudinal susceptibility will have a pole at ω_L if

$$\det\hat{L}(\omega_L)=0. \quad (84)$$

It has been shown that an $\omega_L=0$ pole corresponds to the formation of a local magnetic moment in the ground state. If poles exist for $\omega_L\neq 0$ they may be associated with longitudinal or polar spin excitations involving fluctuations in the spin population at and near the impurity site. Low-energy excitation of this type would indicate that the ground was nearly unstable to the formation of a local moment.

In general, it is expected that the localized spin excitations (transverse or longitudinal) will be damped by decay into bulk spin wave and/or Stoner excitations in which case the excitation energy ω_L will be complex. Since the lifetime of the excitation is inversely proportional to the imaginary part of ω_L the concept of the localized mode is meaningful only if the imaginary part is small compared to the real part. In such a case, we may define the excitation energy of the virtual state by the condition

$$\mathrm{Re}\{\det\hat{L}(\omega)\}=0, \quad (85)$$

for the longitudinal excitations and for the local magnons by

$$\mathrm{Re}\{1-\Delta U[\hat{K}(\omega)]_{00}\}=0. \quad (86)$$

E. Localized Electronic States

Localized electronic states may be formed with or without a local moment or ODSC. In previous sections, we have expressed the local reduced susceptibility functions in terms of the single-particle Green's functions. [See Eqs. (50) and (71).] The single-particle Green's functions contain information about the electronic structure of the impurity system. The formation of localized electronic impurity states is indicated by the appearance of new poles in the one-particle Green's functions. Usually these poles do not occur for real energy so that the localized electronic states are virtual states which decay into the band states.

If the impurity ground state has vanishing ODSC then from Eq. (6) we have that

$$\hat{G}_{\sigma\sigma}(\omega)+(2\pi)^{-1}[\omega-\hat{\epsilon}-\hat{P}_\sigma]^{-1}. \quad (87)$$

In order to illustrate the features of the localized electronic state we approximate the single-particle potential $\hat{P}_\sigma$ by retaining in $\hat{P}_\sigma$ only the change in the impurity population, $\langle n_{0\sigma}\rangle$, and replace all other population factors in $\hat{P}_\sigma$ by their unperturbed values n_σ. This approximation is not generally self-consistent and is not necessarily a good approximation. We employ it here in order to reveal the gross structure of the electronic impurity state. With this approximation for $\hat{P}_\sigma$ we find that

$$[\hat{G}_{\sigma\sigma}(\omega)]_{ij}=[\hat{g}_\sigma(\omega)]_{ij}+\nu_\sigma\{[\hat{g}_\sigma(\omega)]_{i0}[\hat{g}_\sigma(\omega)]_{0j}/(1-\nu_\sigma[\hat{g}_\sigma(\omega)]_{00})\}, \quad (88)$$

where

$$[\hat{g}_\sigma(\omega)]_{ij}=(2\pi N)^{-1}\sum_{\mathbf{k}}[e^{i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)}/(\omega-\epsilon_{\mathbf{k}\sigma}-i0^\dagger)] \quad (89)$$

and

$$\epsilon_{\mathbf{k}\sigma}=\epsilon_{\mathbf{k}}+Un_{\bar{\sigma}},\qquad \epsilon_{\mathbf{k}}=\frac{1}{N}\sum_{\mathbf{R}_i\mathbf{R}_j}\mathcal{E}_{ij}e^{i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)}. \quad (90)$$

In Eq. (90), $\epsilon_{\mathbf{k}\sigma}$ is the band energy plus a contribution due to the Coulomb repulsion and $\mathbf{k}$ is the propagation vector which lies in the first Brillouin zone. The effective single-particle potential ν_σ is given by

$$\nu_\sigma=2\pi[(U+\Delta U)\langle n_{0\bar{\sigma}}\rangle-Un_{\bar{\sigma}}+V]. \quad (91)$$

Localized electronic states will exist when

$$[\hat{g}_\sigma(\omega)]_{00}=1/\nu_\sigma, \quad (92)$$

and as before we may have virtual or resonance states when the real part of Eq. (92) is satisfied. The width of a virtual state is proportional to the imaginary part of $[\hat{g}_\sigma(\omega)]_{00}$, that is to the single-particle density of states $[\hat{N}_{\sigma\sigma}(\omega)]_{00}$ evaluated at the energy of the resonance. Since the potential ν_σ is spin-dependent, localized states occur at different energies for different spins in the ferromagnet. Thus, we expect a local moment will also exist. It can also occur that one spin state is localized while the other is not. We also note that localization does not necessarily require the presence of V, the impurity core scattering potential.

The occurrence of a localized or virtual electronic state will lead to an impurity Green's function with an energy dependence quite different from that of the unperturbed system. From Eqs. (51), (52), (73), and (74) we see that this also leads to significant modifications of the local reduced susceptibility functions. The effect of the localized electronic states will be largest on the local susceptibility at the impurity site, $[\hat{\chi}(\omega)]_{00}$ or $[\hat{\chi}_z(\omega)]_{00}$, and it is just these quantities which enter into the localized spin-excitation resonance equations [Eqs. (85) and (86)]. One is led, therefore, to the conjecture that the localized spin excitations are likely to be associated with the presence of localized electronic states.

IV. SUMMARY AND CONCLUSION

In the preceding sections we have exposed some of the structure of the theory of impurity effects in the one-band model of the ferromagnetic metal. The connection between the formation of ODSC or localized magnetic moments in the ground state and localized spin excitations is established. Expression for the local reduced transverse and longitudinal susceptibility functions are given in terms of the single-particle Green's functions. The theory is developed in the Wannier representation because the impurity perturbation is expected to be short range due to screening effects by the metallic host electrons. This assumption is, of course, implicit in the short-range Coulomb-interaction model Hamiltonian we use. Experimental[17–19] and theoretical[20–22] results on magnetic transition-metal impurities in ferromagnetic Ni indicate that localized magnetic moments are usually formed. In the case of Co, Fe, and Mn in Ni the excess moment is believed to be concentrated almost entirely at the impurity site and while the excess moment associated with Cr or V is more extended it is still localized within a few nearest neighbors.

It remains now to apply the theory developed in this paper to particular situations. It is important to establish whether or not region Ia (local spin canting) exists for realistic values of the perturbation parameters.[23] Preliminary studies indicate that the results are strongly dependent upon the type of energy band employed as well as the degree of self-consistency achieved.

When a highly localized magnetic moment is formed the localized spin tends to be decoupled from the bulk spin waves. The spin excitation energy then depends principally upon the spin splitting of the localized electronic levels and a situation similar to that envisioned by Jaccarino *et al.*[4] results in which the behavior of the impurity spin may be approximately described by a molecular field model. In this case, an Anderson-type extra-orbital Hamiltonian[24] may also serve as an appropriate model.

APPENDIX: UNPERTURBED FERROMAGNET

In this section we apply the formalism developed in the previous sections to the unperturbed ferromagnetic system in order to display some of the features which were mentioned in the text.

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1. Local Reduced Transverse Susceptibility

The single-particle Green's function for the unperturbed ferromagnetic metal, $\hat{g}_\sigma(\omega)$, is given by Eq. (88). The matrix elements of $\hat{\chi}^{(1)}(\omega)$ may be calculated from Eqs. (51) and (52). We have

$$[\hat{R}_{0\sigma\sigma}(\omega)]_{ij}=\frac{1}{N}P\sum_k \frac{e^{i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)}}{\omega-\epsilon_{\mathbf{k}\sigma}}, \tag{A1}$$

where P indicates the principal value and

$$[\hat{N}_{0\sigma\sigma}(\omega)]_{ij}=\frac{1}{N}\sum_k \delta(\omega-\epsilon_{\mathbf{k}\sigma})e^{i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)}. \tag{A2}$$

We use a subscript 0 to indicate that these quantities refer to the unperturbed system. The real and imaginary parts of $\hat{\chi}_0^{(1)}(\omega)$ combine into a single expression

$$[\hat{\chi}_0^{(1)}(\omega)]_{ij}=\frac{1}{N}\sum_{\mathbf{q}} e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\Gamma_{\mathbf{q}}(\omega), \tag{A3}$$

where

$$\Gamma_{\mathbf{q}}(\omega)=\frac{1}{N}\sum_k \frac{f_{\mathbf{k}\uparrow}-f_{\mathbf{k}+\mathbf{q}\downarrow}}{\epsilon_{\mathbf{k}+\mathbf{q}\downarrow}-\epsilon_{\mathbf{k}\uparrow}-\omega+i0^+}. \tag{A4}$$

The sum over $\mathbf{k}$ and $\mathbf{q}$ are over the first Brillouin zone with the usual convention that $\mathbf{k}+\mathbf{q}$ is modulus a reciprocal lattice vector. In Eq. (A4) the Fermi factors $f_{k\sigma}$ are unity for $\epsilon_{k\sigma}$ less than the Fermi energy ϵ_f and vanish otherwise and 0^+ is a positive infinitesimal. The matrix $\hat{\chi}_0^{(1)}(\omega)$ is diagonal in the Bloch representation;

$$\begin{aligned}[\hat{\chi}_0^{(1)}(\omega)]_{\mathbf{q}\mathbf{q}'}&=\frac{1}{N}\sum_{\mathbf{R}_i\mathbf{R}_j} e^{-i\mathbf{q}\cdot\mathbf{R}_i}[\hat{\chi}_0^{(1)}(\omega)]_{ij}e^{i\mathbf{q}'\cdot\mathbf{R}_j}\\&=\delta_{\mathbf{q}\mathbf{q}'}\Gamma_{\mathbf{q}}(\omega).\end{aligned} \tag{A5}$$

In the Bloch representation

$$[(I-U\hat{\chi}_0^{(1)}(\omega))^{-1}]_{\mathbf{q}\mathbf{q}'}=\delta_{\mathbf{q}\mathbf{q}'}/[1-U\Gamma_{\mathbf{q}}(\omega)], \tag{A6}$$

and the transverse susceptibility [for the unperturbed ferromagnet this is equal to $\hat{K}(\omega)$] $\hat{\chi}_0(\omega)$ is

$$[\hat{\chi}_0(\omega)]_{\mathbf{q}\mathbf{q}'}=\delta_{\mathbf{q}\mathbf{q}'}(\Gamma_{\mathbf{q}}/(1-U\Gamma_{\mathbf{q}})). \tag{A7}$$

The local transverse susceptibility is obtained by transforming to the Wannier representation with the result that

$$[\hat{\chi}_0(\omega)]_{ij}=\frac{1}{N}\sum_{\mathbf{q}} e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\frac{\Gamma_{\mathbf{q}}}{1-U\Gamma_{\mathbf{q}}}. \tag{A8}$$

2. Local Reduced Longitudinal Susceptibility

We use Eqs. (73) and (74) to calculate $\hat{\chi}_{0\sigma}^{(1)}(\omega)$ for the unperturbed ferromagnet with the result that

$$[\hat{\chi}_{0\sigma}^{(1)}(\omega)]_{ij}=\frac{1}{N}\sum_{\mathbf{q}} e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\Gamma_{\mathbf{q}\sigma}(\omega), \tag{A9}$$

with

$$\Gamma_{k\sigma}(\omega)=\frac{1}{N}\sum_{k}\frac{f_{k\sigma}-f_{k+q\sigma}}{\epsilon_{k+q\sigma}-\epsilon_{k\sigma}-\omega+i0^{\dagger}}. \quad (A10)$$

Next we construct the matrix $\hat{B}^{-1}(\omega)$ defined by Eq. (76) and find

$$[\hat{B}_{0\sigma\sigma}{}^{-1}(\omega)]_{qq'}=\delta_{qq'}/[1-U^2\Gamma_{q\sigma}(\omega)\Gamma_{q\bar{\sigma}}(\omega)] \quad (A11)$$

and

$$[\hat{B}_{0\sigma\bar{\sigma}}{}^{-1}(\omega)]_{qq'}=\delta_{qq'}U\Gamma_{q\sigma}(\omega)/[1-U^2\Gamma_{q\sigma}(\omega)\Gamma_{q\bar{\sigma}}(\omega)]. \quad (A12)$$

For the case of the unperturbed ferromagnet the longitudinal susceptibility is given by

$$\hat{\chi}_{0z}(\omega)=\tfrac{1}{4}[\hat{\chi}_{0\uparrow}{}^{(2)}(\omega)+\hat{\chi}_{0\downarrow}{}^{(2)}(\omega)], \quad (A13)$$

since $\hat{\chi}_{0\sigma}{}^{(2)}(\omega)$ is equal to $\hat{\chi}_{0\sigma}(\omega)$. Equation (82) may be employed to obtain

$$[\hat{\chi}_{0\sigma}(\omega)]_{qq'}=[\hat{\chi}_{0\sigma}{}^{(2)}(\omega)]_{qq'}=\delta_{qq'}\left\{\frac{\Gamma_{q\sigma}(\omega)(1+U\Gamma_{q\bar{\sigma}}(\omega))}{1-U^2\Gamma_{q\sigma}(\omega)\Gamma_{q\bar{\sigma}}(\omega)}\right\}. \quad (A14)$$

The reduced longitudinal susceptibility in this representation is therefore given by

$$[\hat{\chi}_{0z}(\omega)]_{qq'}=\tfrac{1}{4}\delta_{qq'}\left\{\frac{\Gamma_{q\uparrow}(\omega)+\Gamma_{q\downarrow}(\omega)+2U\Gamma_{q\uparrow}(\omega)\Gamma_{q\downarrow}(\omega)}{1-U^2\Gamma_{q\uparrow}(\omega)\Gamma_{q\downarrow}(\omega)}\right\}. \quad (A15)$$

In the Wannier representation, the elements of the local reduced longitudinal susceptibility are determined by

$$[\hat{\chi}_{0z}(\omega)]_{ij}=\frac{1}{N}\sum_{q}e^{iq\cdot(R_i-R_j)}[\hat{\chi}_{0z}(\omega)]_{qq}. \quad (A16)$$

3. Unperturbed Spin Waves

The spin-wave spectrum of the unperturbed ferromagnet is determined by the poles of the transverse susceptibility. The spectrum is found from the equation

$$U\Gamma_q(\omega_q)=1. \quad (A17)$$

Equation (A17) also determines the spectrum for the Stoner excitations. In the limit of a large system the sum over the $\mathbf{k}$ in Eq. (A4) is replaced by the integral

$$\Gamma_q(\omega)=\frac{1}{V_B}\int d\mathbf{k}\frac{f_{k\uparrow}-f_{k+q\downarrow}}{\epsilon_{k+q\downarrow}-\epsilon_{k\uparrow}-\omega+i0^+}, \quad (A18)$$

where V_B is the volume of the Brillouin zone. In this limit, the discrete poles of $\hat{\chi}(\omega)$ associated with the Stoner excitations are replaced by a density of states. The function $\Gamma_q(\omega)$ is then a complex function. The spin-wave energy is determined by the equation

$$1-U\,\mathrm{Re}[\Gamma_q(\omega_q)]. \quad (A19)$$

The imaginary part of $\Gamma_q(\omega)$ determines the spin-wave relaxation time for decay into Stoner excitations. It is easily verified that the imaginary part of $\Gamma_q(\omega)$ vanishes for small $\mathbf{q}$ so that in the RPA long-wavelength spin waves are undamped. The character of the long-wavelength spin wave may be established by expanding $\Gamma_q(\omega)$. If the spin-wave energy is small compared to the Coulomb integral U then

$$\mathrm{Re}[\Gamma_q(\omega)]=\frac{1}{V_B\Delta}\sum_{n=0}^{\infty}\left(\frac{-1}{\Delta}\right)^n\int d\mathbf{k}(f_{k\uparrow}-f_{k+q\downarrow})\times(\epsilon_{k+q}-\epsilon_k-\omega)^n, \quad (A20)$$

where Δ is the band splitting energy

$$\Delta=U(n_\uparrow-n_\downarrow). \quad (A21)$$

Consider the case of a parabolic energy band with

$$\epsilon_k=(\hbar^2/2m^*)k^2, \quad (A22)$$

where m^* is the effective electron mass for the band. From Eqs. (A19) and (A20) we find to order q^2 that

$$1-U\,\mathrm{Re}[\Gamma_q(\omega)]=1/\Delta\{(\hbar^2/2m_s)q^2-\omega\}+O(q^4), \quad (A23)$$

where the effective spin wave mass is given by[16]

$$m^*/m_s=\{1+\beta^3-\tfrac{4}{5}(\epsilon_f/\Delta)(1-\beta^5)\}/(1-\beta^3). \quad (A24)$$

In Eq. (A24) we have introduced β defined by

$$\beta=1-\Delta/\epsilon_f, \quad (A25)$$

and have assumed that $\Delta<\epsilon_f$. In the case that $\Delta\geqslant\epsilon_f$, $m^*/m_s=\frac{1}{5}$. Thus, in the long-wavelength limit for the parabolic band the spin-wave energy is given by

$$\omega_q=(\hbar^2/2m_s)q^2. \quad (A26)$$

The imaginary part of $\Gamma_q(\omega)$ may also be calculated but will not be given here. We mention here only that there is a critical value for q, above which decay into Stoner excitations can occur.

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