Generalized Landau-Peierls formula for orbital magnetism of many-body systems: Effects of spin fluctuations

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A simple and general formula for the orbital magnetism has been obtained for a many-body system in which the one-particle self-energy function depends on both energy and momentum variables. The result is an extension of the Landau-Peierls formula, which is valid only in the case of an energy-independent self-energy. Using this formula the effects of spin fluctuations on the orbital susceptibility of itinerant electrons in a nondegenerate band are examined near the Curie temperature T_C. If the spin-orbit interaction is neglected, the orbital susceptibility is modified only to order $(T_C/\epsilon_F)^2$, where ϵ_F is the Fermi energy. In the presence of the spin-orbit interaction, there is a contribution proportional to the static spin susceptibility which is divergent at $T = T_C$. However this contribution is generally small since the proportionality constant is of order α^2, where $\alpha = (137)^{-1}$ is the fine-structure constant.

I. INTRODUCTION

It is a well-established fact that the Coulomb interactions between electrons give rise to exchange forces that enhance the spin susceptibility, resulting in the existence of strong magnetism such as ferromagnetism, antiferromagnetism, etc.[1] In the previous discussions, it was assumed that the orbital susceptibility is not modified appreciably even when the paramagnetic spin susceptibility becomes singular. Although this assumption seems reasonable, there has been no detailed examination of the problem thus far. In this paper we examine the validity of this assumption by investigating the effects of spin fluctuations[2] in the paramagnetic phase near the Curie temperature. The model we take is the itinerant-electron system represented by the Hubbard model[3]

$$\mathcal{H} = \sum_{ij,\sigma} t_{ij} a^{\dagger}_{i,\sigma} a_{j,\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} , \tag{1.1}$$

where t_{ij} and U are the transfer integral and the Coulomb repulsive energy, respectively.

Before examining the problem, we first derive a formula for the orbital susceptibility which has a wide range of applicability. We should mention that deriving a simple formula which can be applied to the present problem was not a trivial step. This is because the main reason for the unsatisfactory understanding of orbital magnetism is simply the complex way magnetic fields affect the free energy, although there is nothing very profound in the fundamental principles.[4-6] Such complexity arises from the fact that the magnetic field affects the energy and the wave function of each electron below the Fermi energy, so that arguments similar to the Fermi-liquid theory[7] seem not to hold in general.

Historically speaking, however, Sampson and Seitz,[8] who first examined the effects of Coulomb interactions between otherwise free electrons, applied the Landau-Peierls formula[9] to the one-particle energy spectrum obtained in the Hartree-Fock approximation. This formula is given by

$$\chi = \frac{e^2}{6c^2} \sum_{k} \left[\frac{\partial^2 E}{\partial k_x^2} \frac{\partial^2 E}{\partial k_y^2} - \left(\frac{\partial^2 E}{\partial k_x \partial k_y} \right)^2 \right] \frac{\partial}{\partial E} f , \tag{1.2}$$

where $E(k)$ is the one-particle energy, $f(E) = [e^{\beta(E-\mu)} + 1]^{-1}$. $\beta^{-1} = kT$ and μ are the temperature and the chemical potential. Since the orbital magnetism in this formula is determined by electrons at the Fermi energy only, we may doubt its validity in the presence of electron-electron interactions. This procedure, the application of the Landau-Peierls formula to the quasiparticle energy spectrum, has been termed the Sampson-Seitz (SS) prescription.[10] After Sampson and Seitz, several workers[11] applied this prescription to the more elaborate quasiparticle energy calculated in the Bohm-Pines theory[12] of correlations.

Due to the obscurity of its foundations, the SS prescription has not been used very much. Instead, a simple perturbative method has usually been used to examine the effects of Coulomb interactions. In particular, Kanazawa and Matsudaira[13] evaluated the correction terms due to a statically screened Coulomb interaction by use of the Green's-function technique. They expanded the one-particle Green's function and vertex corrections to the current operator in terms of screened Coulomb forces and retained the first-order term in such a way as to maintain the gauge invariance. In this respect, their calculations are based on the free-electron energy spectrum and they did not take into account the full form of self-energy and

vertex corrections. Although some works[14] examined higher-order corrections in this scheme, they are simple perturbative treatments.

On the other hand, the SS prescription, if it is correct, is more satisfactory since it includes the quasiparticle energy spectrum itself, or the pole of the full Green's function. Philippas and McClure[10] noticed this fact and *proved* the validity of this prescription for the electron gas with statically screened interactions. In their proof it was essential that the self-energy function does not depend on the energy variable. Thus it is known that the orbital susceptibility χ is solely determined by the properties at the Fermi energy if the self-energy function depends only on the momentum variable. At about the same time, Rajagopal and Jain[15] reached similar conclusions by a different approach. Using the general expression of the electromagnetic response based on the Maxwell equations, they reduced the problem of determining the orbital susceptibility to that of the vertex function of the current operator at finite wave vectors, and solved it variationally in the case of static screening. Their final results agree with those of Ref. 10.

In general, however, the self-energy function in the interacting electron system depends not only upon the momentum but also upon the energy variable. Contrary to the case where the SS prescription holds, we encounter situations where the self-energy function is solely dependent on the energy variable. An example is a strongly correlated electron system in a narrow band represented by the Hubbard model, Eq. (1.1). The approximations[3] thus far developed to treat the effects of strong correlations yield a self-energy function dependent only on the energy variable. The orbital magnetism in such cases has been discussed by one of the present authors,[16] who first derived a simple formula valid in this case and found that, even if the band is split off by correlations, there exist interband contributions caused by a mechanism similar to Van Vleck paramagnetism.[17] In between these two limiting cases there exist various situations where both momentum and energy dependences are important. For example the dynamical screening in an otherwise free-electron gas[18] introduces these dependences, which yield various important consequences. Moreover in magnetic metals, which are of interest here, spin fluctuations[2] with low excitation energies affect the one-particle energy appreciably, especially near the second-order phase transition, resulting in a strong energy and momentum dependence of the self-energy function. Since we lacked a simple formula for the orbital susceptibility applicable to such a case, we first derive it in Sec. II by taking an electron gas with Coulomb interactions as an example. The Green's-function technique[19] similar to Ref. 16 is employed so that the gauge invariance is clearly seen. The form of the derivation is conceptionally similar to that of Rajagopal and Jain[15] in the sense that we are concerned with the vertex function of the current operator at a finite wave vector $\vec{Q}$. In contrast to Ref. 15, however, we do not treat this function for general values of Q but derive the rigorous expression for the expansion coefficient to second order in Q, which is sufficient to give the orbital susceptibility in a uniform field. Thus applications, such as the cross section for neutron scattering,[6] are beyond our scope. The formula thus obtained is shown to have a wide range of applicability, which is discussed in Sec. III. The effects of spin fluctuations are examined in Sec. IV, and discussion is given in Sec. V, where the effects of spin-orbit interactions are also examined.

II. DERIVATION OF GENERAL FORMULA FOR ORBITAL MAGNETISM

For the sake of convenience, we consider a system in a unit volume represented by the Hamiltonian

$$\mathcal{H}=\sum_i \frac{1}{2m}\left(p_i-\frac{e}{c}A_i\right)^2+\frac{1}{2}\sum_{i\neq j} v(r_i-r_j) \quad , \qquad (2.1)$$

where $v(r)=e^2/r$ is the Coulomb interaction and $A(r)$ is the vector potential related to the magnetic field by $\vec{H}=\nabla\times\vec{A}$. Although in a rigorous sense, A includes not only the external magnetic field but also the local-field correction, we can usually neglect the latter since χ itself is small except in the case of superconductors, which are not treated in this paper. The static susceptibility in the weak-field limit is given by[20]

$$\chi=-\left(\frac{1}{H}\frac{\partial\Omega}{\partial H}\right)_{H=0} \quad , \qquad (2.2)$$

where Ω is the thermodynamic potential, defined by

$$e^{-\Omega/kT}=\mathrm{Tr}\, e^{-\beta(\mathcal{H}-\mu N)} \quad . \qquad (2.3)$$

In Eq. (2.2), μ is held fixed in the course of differentiation. As is seen, the expansion of Ω in terms of H, or A, up to the second order is sufficient to determine χ. We take for the vector potential[6]

$$\vec{A}(\vec{r})=\vec{A}_Q(e^{i\vec{Q}\cdot\vec{r}}-e^{-i\vec{Q}\cdot\vec{r}})/i \quad , \qquad (2.4)$$

or $\vec{H}=2\vec{Q}\times\vec{A}_Q$ in the limit of uniform fields. By use of Eq. (2.4), we rewrite Eq. (2.1) as follows:

$$\mathcal{H}=\sum_k \epsilon(k)a_k^\dagger a_k+\sum_k v(q)a_{k+q}^\dagger a_{k'-q}^\dagger a_{k'}a_k$$

$$-\frac{e}{ci}A_Q(j_Q-j_{-Q})-\frac{e^2}{2mc^2}A_Q^2(\rho_{2Q}+\rho_{2Q}-2\rho_0)\ , \tag{2.5}$$

where $\epsilon(k)=k^2/2m$, $v(q)=4\pi e^2/q^2$, and

$$j_Q=\sum_k \frac{k}{m}a_{k^+}^\dagger a_{k^-}\ , \tag{2.6}$$

$$\rho_Q=\sum_k a_{k^+}^\dagger a_{k^-}\ , \tag{2.7}$$

$$\vec{k}^\pm=\vec{k}\pm\tfrac{1}{2}\vec{Q}\ . \tag{2.8}$$

Ω, Eq. (2.3), can be calculated as a perturbation series in A_Q by use of the thermal Green's function $\mathcal{G}$ of the system without the magnetic field, defined by[21]

$$\mathcal{G}(k,i\epsilon_n)=-\frac{1}{\beta}\int_0^\beta d\tau\int_0^\beta d\tau' e^{i\epsilon_n(\tau-\tau')}\langle Ta_k(\tau)a_k^\dagger(\tau')\rangle$$

$$=[i\epsilon_n+\mu-\epsilon(k)-\Sigma(k,i\epsilon_n)]^{-1}\ , \tag{2.9}$$

where $\epsilon_n=(2n+1)\pi T$ and $\Sigma(k,i\epsilon_n)$ is the self-energy function. The correction to Ω in the second order of A_Q, $\Omega^{(2)}$, can be written as[19]

$$\Omega^{(2)}=-2\left(\frac{e}{c}\right)^2\sum_{\lambda,\nu}A_{Q,\lambda}A_{Q,\nu}\,T$$

$$\times\sum_n\sum_k\left[\frac{\partial\epsilon}{\partial k_\lambda}\mathcal{G}(k^+,i\epsilon_n)\Gamma_\nu(k^+,k^-,i\epsilon_n)\right.$$

$$\left.\times\mathcal{G}(k^-,i\epsilon_n)+(\lambda\leftrightarrow\nu)\right]$$

$$-(4c^2/mc^2)A_Q^2T\sum_n\sum_k\mathcal{G}(k,i\epsilon_n)\ , \tag{2.10}$$

where $\Gamma_\nu(k^+,k^-;i\epsilon_n)$ is the vertex function of $\partial\epsilon/\partial k_\nu$, which is explicitly defined below by Eq. (2.13), and the spin-degeneracy factor 2 is included. Equation (2.10) is quite general. To obtain an explicit answer for χ we need to specify an approximation for the self-energy function Σ and consequently for the vertex function Γ_ν. In this paper we employ the random-phase-approximation (RPA),[18] in which Σ is given by

$$\Sigma(k,i\epsilon_n)=T\sum_l\sum_q\mathfrak{D}(q,i\omega_l)\mathcal{G}(k-q,i\epsilon_n-i\omega_l)\ , \tag{2.11}$$

where $\omega_l=2\pi lT$ and $\mathfrak{D}(q,i\omega_l)$ is the dynamically screened Coulomb interaction

$$\mathfrak{D}(q,i\omega_l)=v(q)\left(1-v(q)T\sum_n\sum_k\mathcal{G}(k,i\epsilon_n)\right.$$

$$\left.\times\mathcal{G}(k+q,i\epsilon_n+i\omega_l)\right)^{-1}\ . \tag{2.12}$$

Equations (2.11) and (2.12) are diagrammatically shown in Fig. 1, where the dotted and the wavy lines represent the bare Coulomb interaction $v(q)$ and the dynamically screened interaction $\mathfrak{D}(q,i\omega_l)$, respectively, and $\mathcal{G}$ is represented by a solid line. The vertex function Γ_ν, consistent with the approximation to the self-energy function, Eqs. (2.11) and (2.12), is determined by the following integral equation:

$$\Gamma_\nu(k^+,k^-;i\epsilon_n)=\frac{\partial\epsilon}{\partial k_\nu}+T\sum_l\sum_q\Gamma_\nu(k^+-q,k^--q;i\epsilon_n-i\omega_l)\mathcal{G}(k^+-q,i\epsilon_n-i\omega_l)\mathcal{G}(k^--q,i\epsilon_n-i\omega_l)\mathfrak{D}(q,i\omega_l)$$

$$+T\sum_l\sum_q T\sum_{n'}\sum_{k'}\Gamma_\nu(k'^+,k'^-;i\epsilon_{n'})\mathcal{G}(k'^+,i\epsilon_{n'})\mathcal{G}(k'^-,i\epsilon_{n'})[\mathcal{G}(k'-q,i\epsilon_{n'}-i\omega_l)+\mathcal{G}(k'+q,i\epsilon_{n'}+i\omega_l)]$$

$$\times\mathfrak{D}(q^+,i\omega_l)\mathfrak{D}(q^-,i\omega_l)\mathcal{G}(k-q,i\epsilon_n-i\omega_l)\ , \tag{2.13}$$

where $q^\pm=q\pm Q/2$. Equation (2.13) is represented in Fig. 2, where Γ_ν is represented by a doubly shaded vertex. Before discussing Γ_ν for finite values of Q, we note that Γ_ν at $Q=0$, $\Gamma_\nu(k,k,i\epsilon_n)\equiv\Gamma_\nu^0(k,i\epsilon_n)$, is given by

$$\Gamma_\nu^0(k,i\epsilon_n)=\partial_\nu[\epsilon(k)+\Sigma(k,i\epsilon_n)] \tag{2.14}$$

or

$$-\partial_\nu\mathcal{G}(k,i\epsilon_n)=\mathcal{G}^2(k,i\epsilon_n)\Gamma_\nu^0(k,i\epsilon_n)\ . \tag{2.15}$$

In Eqs. (2.14) and (2.15), and thereafter, we use the notation $\partial_\nu F\equiv\partial F/\partial k_\nu$. That Γ_ν^0 [Eq. (2.14)] is the solution of Eq. (2.13) in the case of $Q=0$, is seen as follows. By use of Eq. (2.15), the second term of Eq. (2.13) can be transformed as

$$-T\sum_l\sum_q\partial_\nu\mathcal{G}(k-q,i\epsilon_n-i\omega_l)\mathfrak{D}(q,i\omega_l)=\partial_\nu\Sigma(k,i\epsilon_n), \tag{2.16}$$

whereas in this case of $Q=0$ the summations over k' and n' in the last term of Eq. (2.13) can be performed as

$$T\sum_{n'}\sum_{k'}\Gamma_\nu^0(k',i\epsilon_{n'})\mathcal{G}^2(k',i\epsilon_{n'})[\mathcal{G}(k'-q,i\epsilon_{n'}-i\omega_l)+\mathcal{G}(k'+q,i\epsilon_{n'}+\omega_l)]$$

$$=-T\sum_{n'}\sum_{k'}\partial_\nu'\mathcal{G}(k',i\epsilon_{n'})[\mathcal{G}(k'-q,i\epsilon_{n'}-i\omega_l)+\mathcal{G}(k'+q,i\epsilon_{n'}+i\omega_l)]$$

$$= -T\sum_{n'}\sum_{k'}[\mathcal{G}(k'-q, i\epsilon_{n'}-i\omega_l)\partial'_\nu \mathcal{G}(k', i\epsilon_{n'}) + \mathcal{G}(k', i\epsilon_{n'})\partial'_\nu \mathcal{G}(k'-q, i\epsilon_{n'}-i\omega_l)]$$

$$= -T\sum_{n'}\sum_{k'}\partial'_\nu[\mathcal{G}(k'-q, i\epsilon_{n'}-i\omega_l)\mathcal{G}(k', i\epsilon_{n'})]$$

$$= 0 \quad . \tag{2.17}$$

Here $\partial'_\nu = \partial/\partial k'_\nu$. The second equality is due to the change of variables $k' \to k'-q$, $\epsilon_{n'} \to \epsilon_{n'} - \omega_l$, in the second term in the bracket and the last equality is due to the integration over k'. The schematic representation of Eq. (2.17) is shown in Fig. 3 (compared with Fig. 2), where $\Gamma^0_\nu(k, i\epsilon_n)$ is represented by a singly shaded vertex in order to stress the difference between Γ_ν, Eq. (2.13) and Fig. 2, and Γ^0_ν. Thus the right-hand side of Eq. (2.13) is seen to be equal to Γ^0_ν, which is self-consistent. Because of Eq. (2.15), $\Omega^{(2)}$, Eq. (2.10), vanishes in the limit of $Q=0$; the first term of Eq. (2.10) canceling the second after a partial integration. As we are concerned with a uniform magnetic field, the expansion of $\Omega^{(2)}$ in terms of Q up to the second order is sufficient, which we will outline in the following.

First we note the following expansion:

$$\mathcal{G}(k^+, i\epsilon_n)\mathcal{G}(k^-, i\epsilon_n) = \mathcal{G}^2 - 4^{-1}Q_\alpha Q_\beta(2-\delta_{\alpha\beta}) \times[\partial_\alpha \mathcal{G}\partial_\beta \mathcal{G} - \mathcal{G}\partial_{\alpha\beta}\mathcal{G}]$$

$$\equiv \mathcal{G}^2 + Q_\alpha Q_\beta \pi_{\alpha\beta} \quad . \tag{2.18}$$

In this equation and hereafter, the summation symbol over components of vectors, α and β, is suppressed in the case where two same indices appear in one expression. In order to determine the coefficient of $Q_\alpha Q_\beta$ in Γ_ν, we formally write Eq. (2.13) as follows:

$$\Gamma_\nu = \partial_\nu \epsilon + \Gamma_\nu \mathcal{G}_+\mathcal{G}_- K \quad , \tag{2.19}$$

where K is the kernel of the integral equation represented by the second and third term of Eq. (2.13) and is diagrammatically given in Fig. 4. In Eq. (2.19), $\mathcal{G}_\pm = \mathcal{G}(k'^{\pm}, i\epsilon_{n'})$ and the integration symbol is suppressed. We expand Eq. (2.19) in terms of Q. Since $\mathcal{G}_+\mathcal{G}_- K$ does not include a term linear in Q, Γ_ν can be written as

$$\Gamma_\nu = \Gamma^0_\nu + Q_\alpha Q_\beta \Gamma^{\alpha\beta}_\nu \quad , \tag{2.20}$$

where the equation for $\Gamma^{\alpha\beta}_\nu$ can be obtained by inserting Eq. (2.10) into Eq. (2.19),

$$\Gamma^{\alpha\beta}_\nu = \Gamma^0_\nu \pi^{\alpha\beta} K + \Gamma^{\alpha\beta}_\nu \mathcal{G}^2 K \quad . \tag{2.21}$$

In Eq. (2.21), use is made of the fact that the contribution coming from the expansion of $\mathfrak{D}(q^+, i\omega_l)\mathfrak{D}(q^-, i\omega_l)$ vanishes. The product $\mathfrak{D}(q^+, i\omega_l)\mathfrak{D}(q^-, i\omega_l)$ has no linear term in Q. In evaluating the effect of the quadratic term for this product, all other Q's in Eq. (2.13) may be put to zero and the result vanishes by use of Eq. (2.17). By use of $\pi^{\alpha\beta}$ and $\Gamma^{\alpha\beta}_\nu$ in Eq. (2.10) for $\Omega^{(2)}$, we obtain

$$\Omega^{(2)} = -2\left(\frac{e}{c}\right)^2 \sum_{\lambda,\nu,\alpha,\beta} A_{Q,\lambda}A_{Q,\nu}Q_\alpha Q_\beta T \times\sum_n\sum_k[(\partial_\lambda \epsilon \mathcal{G}^2\Gamma^{\alpha\beta}_\nu) + (\nu \leftrightarrow \lambda)] \quad . \tag{2.22}$$

Next, we will find the relationship between $\Gamma^{\alpha\beta}_\lambda$ and Γ^0_ν. For this purpose, we iterate Eq. (2.21) to get

$$\Gamma^{\alpha\beta}_\nu = \Gamma^0_\nu \pi^{\alpha\beta}(K + K\mathcal{G}^2 K + K\mathcal{G}^2 K\mathcal{G}^2 K + \cdots) \quad . \tag{2.23}$$

On the other hand, Eq. (2.19) leads to

$$\Gamma^0_\lambda = \partial_\lambda \epsilon(1 + \mathcal{G}^2 K + \mathcal{G}^2 K\mathcal{G}^2 K + \cdots) \tag{2.24}$$

or

$$\Gamma^0_\lambda - \partial_\lambda \epsilon = \partial_\lambda \epsilon \mathcal{G}^2(K + K\mathcal{G}^2 K + \cdots) \quad . \tag{2.25}$$

Thus we find

$$T\sum_n\sum_k \partial_\lambda \epsilon \mathcal{G}^2 \Gamma^{\alpha\beta}_\nu = T\sum_n\sum_k (\Gamma^0_\lambda - \partial_\lambda \epsilon)\pi^{\alpha\beta}\Gamma^0_\nu \quad . \tag{2.26}$$

Consequently, we obtain

$$\Omega^{(2)} = -4\left(\frac{e}{c}\right)^2 \sum_{\lambda,\nu,\alpha,\beta} A_{Q,\lambda}A_{Q,\nu}Q_\alpha Q_\beta T\sum_{n,k}\Gamma^0_\lambda\Gamma^0_\nu\pi^{\alpha\beta} \quad . \tag{2.27}$$

Hereafter we suppress the superscript 0 for Γ^0_λ. That $\Omega^{(2)}$ given by Eq. (2.27) is proportional to the observable magnetic field $H^2 = (2\vec{Q}\times\vec{A}_Q)^2$ irrespective of the gauge of the vector potential A_Q is shown by the following equations:

$$T\sum_n\sum_k \Gamma^2_x \pi^{xx} = -4^{-1}T\sum_n\sum_k \Gamma^2_x[(\partial_x \mathcal{G})^2 - \mathcal{G}\partial_{xx}\mathcal{G}]$$

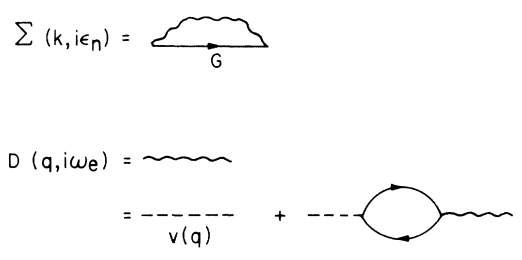

FIG. 1. Self-energy function Σ due to the dynamically screened exchange interaction. $v(q)$ (broken line) and $\mathfrak{T}(q, i\omega_l)$ (wavy line) are bare and screened Coulomb interaction, respectively. $\mathcal{G}$ (solid line) is the one-particle Green's function.

$$= -12^{-1}T\sum_n\sum_k \partial_x(\mathcal{G}\Gamma_x)^3 = 0 \qquad (2.28)$$

by use of Eq. (2.15). Similarly,

$$T\sum_n\sum_k \Gamma_x^2\pi^{yy} = -4^{-1}T\sum_n\sum_k \Gamma_x^2[(\partial_y\mathcal{G})^2 - \mathcal{G}\partial_{yy}\mathcal{G}]$$
$$= -2^{-1}T\sum_n\sum_k (\Gamma_x^2\Gamma_y^2\mathcal{G}^4 + \Gamma_x\Gamma_y\Gamma_{xy}\mathcal{G}^3) \quad , \qquad (2.29)$$

where $\Gamma_{xy} = \partial_x\Gamma_y = \partial_y\Gamma_x$, and

$$T\sum_n\sum_k \Gamma_x\Gamma_y\pi^{xy} = -2^{-1}T\sum_n\sum_k \Gamma_x\Gamma_y(\partial_x\mathcal{G}\partial_y\mathcal{G} - \mathcal{G}\partial_{xy}\mathcal{G})$$
$$= 2^{-1}T\sum_n\sum_k (\Gamma_x^2\Gamma_y^2\mathcal{G}^4 + \Gamma_x\Gamma_y\Gamma_{xy}\mathcal{G}^3) \quad . \qquad (2.30)$$

Thus

$$\Omega^{(2)} = 2\left(\frac{e}{c}\right)^2 (\vec{Q}\times\vec{A}_Q)^2 T\sum_n\sum_k (\Gamma_x^2\Gamma_y^2\mathcal{G}^4 + \Gamma_x\Gamma_y\Gamma_{xy}\mathcal{G}^3)$$
$$\equiv -\tfrac{1}{2}\chi H^2 \quad , \qquad (2.31)$$

or

$$\chi = -\left(\frac{e}{c}\right)^2 T\sum_n\sum_k (\Gamma_x^2\Gamma_y^2\mathcal{G}^4 + \Gamma_x\Gamma_y\Gamma_{xy}\mathcal{G}^3) \quad . \qquad (2.32)$$

By partial integration, Eq. (2.32) can be rewritten as (see Appendix)

$$\chi = \frac{1}{6}\left(\frac{e}{c}\right)^2 T\sum_n\sum_k [\Gamma_{xx}\Gamma_{yy} - (\Gamma_{xy})^2]\mathcal{G}^2 \quad , \qquad (2.33)$$

where

$$\Gamma_{\lambda\nu} = -\frac{\partial^2}{\partial k_\lambda \partial k_\nu}\mathcal{G}^{-1}(k, i\epsilon_n) = \frac{\partial^2}{\partial k_\lambda \partial k_\nu}[\epsilon(k) + \Sigma(k, i\epsilon_n)] \quad .$$

We emphasize that $\mathcal{G}$ is the one-particle Green's function including the self-energy correction. Equation (2.33) is the fundamental result obtained in this paper.

III. DISCUSSION OF THE GENERAL FORMULA

A. Validity of Eq. (2.33) in some known cases

1. *Free-electron gas*

In this case $\Sigma = 0$ and $\epsilon(k) = k^2/2m$, so that $\Gamma_{xx} = \Gamma_{yy} = m^{-1}$ and $\Gamma_{xy} = 0$. Thus by use of $\mathcal{G}^{-1} = i\epsilon_n + \mu - \epsilon(k)$, we have

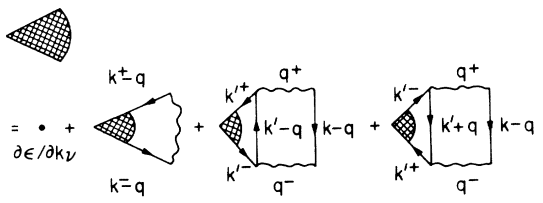

FIG. 2. Integral equation for the vertex function Γ_ν of the velocity operator $\partial\epsilon/\partial k_\nu$. Γ_ν is represented by a doubly shaded segment, and $k^\pm = k \pm Q/2$, etc.

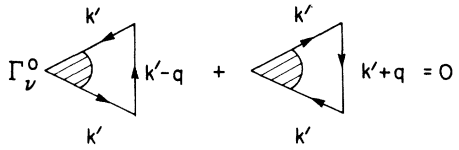

FIG. 3. Diagrammatic representation of Eq. (2.17), the fact that the third term of Eq. (2.13) vanishes if $Q = 0$. The vertex function at $Q = 0$ is written as Γ^0 and shown as singly shaded segment.

$$\chi_L = \frac{2\mu_B^2}{3}T\sum_n\sum_k \mathcal{G}_0^2 = \frac{2\mu_B^2}{3}\sum_k \frac{\partial}{\partial\epsilon}f(\epsilon) = -\frac{mk_F}{3\pi^2}\mu_B^2 \quad , \qquad (3.1)$$

where $\mu_B = e/2mc$ is the Bohr magneton. Equation (3.1) is the Landau diamagnetism.[9]

2. *Bloch electrons*

Although Eq. (2.33) is explicitly derived for an electron gas in the presence of the Coulomb interaction without the periodic potential, it gives us the Landau-Peierls formula if we take $\epsilon(k)$ as the energy function of a Bloch band and take $\Sigma = 0$. In that case it is seen that Eq. (2.33) reduces to Eq. (1.2). That we get the Landau-Peierls formula even if we ignore the effects of the periodic potential is due to the fact that this formula only takes account of the modifications of the motion of Bloch electrons by the magnetic field in the form of $\epsilon(k - (e/c)A)$, disregarding the interband coupling completely. This is parallel to the derivation of Eq. (2.33), where the expansion in terms of Q affects only the one-particle Green's function. Thus Eq. (2.33) is applicable to perturbed Bloch electrons with the same range of applicability as the Landau-Peierls formula.

3. *Sampson-Seitz prescription*

If the mutual Coulomb interaction is treated using a statically screened interaction, the self-energy function is independent of the energy variable. Thus the sum on n in Eq. (2.33) affects the $\mathcal{G}^2$ factor only, and Eq. (1.2) is obtained with $E(k) = \epsilon(k) + \Sigma(k)$. By use of the fact that ϵ and Σ depend on $|k|$ only, the result can be written

$$\chi = -\frac{1}{12}\left(\frac{e}{\pi c}\right)^2\left(\frac{1}{3}\frac{dE}{dk} + \frac{2}{3}k\frac{d^2E}{dk^2}\right)_{k=k_F} \quad . \qquad (3.2)$$

Equation (3.2) is the SS prescription.[8,10] As is evident, the energy independence of $\Sigma(k, i\epsilon_n)$ is crucial to this prescription.

B. Range of applicability of Eq. (2.33)

Equation (2.33) was derived for the case of the electron gas. In this case

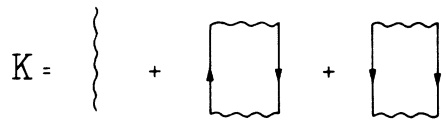

FIG. 4. The four-vertex function, K, defined by Eqs. (2.13) and (2.19).

$$\mathfrak{D}(q, i\omega_l) = \frac{4\pi e^2}{q^2 \epsilon(q, i\omega_l)} \quad , \tag{3.3}$$

where $\epsilon(q, i\omega_l)$ is the dielectric function. In the past the dynamical effects, the contribution coming from the ω_l dependence of $\epsilon(q, i\omega_l)$, have been supposed to be minor and χ was estimated within the static approximation, which neglects the energy of an electron-hole pair compared to the plasmon energy. Because of the fact that the plasmon energy can be much smaller than the Fermi energy in the high-density limit, we cast doubt on the validity of this approximation, although we do not treat this problem here.

We list in the following other cases where Eq. (2.33) can be applied.

(1) *The electron-phonon interaction.*

This case has been examined by Tani[22] by a quite different method. In this case, $\mathfrak{D}$ is the phonon propagator.

(2) *The elastic scattering potential with a finite force range treated within the Born approximation.*[23]

In this case, $\mathfrak{D}$ is given by

$$\mathfrak{D}(q, i\omega_l) = \delta_{\omega_l} n_i \,|\, u(q) \,|^2 g(q) \quad , \tag{3.4}$$

where n_i, $u(q)$, and $g(q)$ are the number density of impurities, the Fourier transform of the scattering potential and the Fourier transform of the pair-distribution function of the impurities. The existence of the Kronecker δ, δ_{ω_l} represents the elasticity of the scattering.

If the perturbing potential has a short range, Eq. (2.33) is applicable to binary alloys treated in the coherent-potential approximation (CPA)[24] as was explicitly derived in Ref. 16 and applied later.[25] In this case the fact that the self-energy function is independent of the momentum variable even if we sum all orders of $u(q)$ makes the formula Eq. (2.33) valid.

Note that the periodic potential can also be treated within this equation by use of the pseudopotential method.[26] The liquid metal can similarly be treated.[27]

IV. EFFECT OF SPIN FLUCTUATIONS IN A SIMPLE CASE

We employ the model Hamiltonian given by Eq. (1.1). We confine ourselves to the paramagnetic state, where the spin fluctuations[2] are represented by the transverse component of the spin susceptibility, $\chi_{sp}(q, i\omega_l)$. Near $T = T_C$, $\mathfrak{D}(q, i\omega_l)$ is approximately related to $\chi_{sp}(q, i\omega_l)$ as follows:

$$\mathfrak{D}(q, i\omega_l) = -\tfrac{3}{2} U \left(1 + \frac{U}{\mu_B^2} \chi_{sp}(q, i\omega_l)\right) , \tag{4.1}$$

$$\chi_{sp}(q, i\omega_l) = -\mu_B^2 T \sum_n \sum_k \mathcal{G}(k, i\epsilon_n) \mathcal{G}(k+q, i\epsilon_n + i\omega_l) \times \left(1 + U T \sum_n \sum_k \mathcal{G}(k, i\epsilon_n) \mathcal{G}(k+q, i\epsilon_n + i\omega_l)\right)^{-1} . \tag{4.2}$$

The factor $\frac{3}{2}$ takes account of both longitudinal and transverse components of the spin fluctuations. The self-energy function given by Eq. (2.11) includes two processes like those shown in Fig. 5. In the calculations to follow, we assume that the band energy is given by that of nearly free electrons, i.e., $\epsilon(k) = k^2/2m^*$ with the effective mass m^*. Considering the spherical symmetry of $\Sigma(k, i\epsilon_n)$ in momentum space, we first rewrite Eq. (2.33) as follows:

$$\chi = \frac{1}{18}\left(\frac{e}{c}\right)^2 T \sum_n \sum_k \frac{1}{k^2} [E^2_{(1)} + 2kE_{(1)}E_{(2)}] \mathcal{G}^2 , \tag{4.3}$$

where

$$E_{(n)} = \frac{d^n}{dk^n} [\epsilon(k) + \Sigma(k, i\epsilon_n)] \quad .$$

Expecting that the effect is small, which is justified later, we expand Eq. (4.3) in terms of Σ and retain the lowest-order term,

$$\chi = \chi_0 + \Delta\chi \quad ,$$

$$\chi_0 = \frac{1}{6}\left(\frac{e}{m^* c}\right)^2 T \sum_n \sum_k \mathcal{G}_0^2 = -\frac{1}{6}\left(\frac{e}{m^* c}\right)^2 \rho(\epsilon_F) = -\frac{1}{12\pi^2}\left(\frac{e}{c}\right)^2 \frac{k_F}{m^*} \quad , \tag{4.4}$$

$$\Delta\chi = \frac{1}{18 m^*}\left(\frac{e}{\pi c}\right)^2 T \sum_n \int_0^\infty dk\, \mathcal{G}_0^2 [-3\Sigma_{(0)} + k\Sigma_{(1)} + 2k^2 \Sigma_{(2)}] \quad , \tag{4.5}$$

where $\mathcal{G}_0^{-1} = i\epsilon_n + \mu - \epsilon(k)$, $\Sigma_{(n)} = d^n\Sigma/dk^n$, $\epsilon_F = k_F^2/2m^*$ and $\rho(\epsilon_F)$ is the density of states at the Fermi energy in the presence of spin fluctuations. In deriving Eq. (4.5), use has been made of Eq. (2.15) ap-

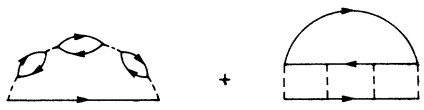

FIG. 5. Electron self-energy function due to spin fluctuations. Broken lines are Coulomb repulsive force U.

plied to $\mathcal{G}_0$, and a partial integration has been performed. In order to evaluate Eq. (4.5), we need Σ, which is determined in the following. Since we are interested in temperatures near a finite Curie temperature T_C, it is sufficient to take out the $\omega_l = 0$ component in Eq. (2.11), and then to neglect the contribution with $\omega_l \neq 0$. This comes from the fact that $\mathfrak{D}(q, i\omega_l)$ $(\omega_l \neq 0)$ does not have a singularity in small-q limit even if $T = T_C$.[2,28] In this case, $\mathfrak{D}(q, 0)$ can be written as[28]

$$\mathfrak{D}(q, 0) = -\tfrac{3}{2}\pi\,[\eta + Dq^2]^{-1} \quad , \tag{4.6}$$

where $\eta = (T - T_C)/T_C$ and $D = U/6\pi^2 v_F$. Here $v_F = k_F/m^*$ is the Fermi velocity. Inserting Eq. (4.6) into Eq. (2.11) and integrating over q, we have

$$\Sigma\,(k, i\epsilon_n) = \frac{3i}{8\pi}\, UT_C m^* (Dk)^{-1} \ln\left(\frac{iq_0 + q_n + k}{iq_0 + q_n - k}\right) \quad , \tag{4.7}$$

where $q_0^2 = \eta D^{-1}$ and $q_n = [2m^*(i\epsilon_n + \epsilon_F)]^{1/2}$ $(\mathrm{Im} q_n > 0)$. Using Eq. (4.7) we evaluate Eq. (4.5) as follows:

$$\Delta\chi = i\frac{m^{*2}}{6\pi D}\left(\frac{e}{\pi c}\right)^2 UT_C^2 \times \sum_n \int_0^\infty dk\, \frac{(q_n + iq_0)[7k^2 - 3(iq_0 + q_n)^2]}{(q_n^2 - k^2)^2[(iq_0 + q_n)^2 - k^2]^2} \quad . \tag{4.8}$$

Equation (4.8) is seen to be convergent even if $T = T_C$ or $q_0 = 0$, and $\Delta\chi$ at $T = T_C$ can be estimated as

$$\Delta\chi = -\frac{11\pi^2}{96}\left(\frac{e}{\pi c}\right)^2 T_C^2 UD^{-1} \sum_n q_n^{-4} \quad . \tag{4.9}$$

The summation in Eq. (4.9) can be performed as follows:

$$\sum_{n=-\infty}^{\infty} q_n^{-4} = \sum_{n=-\infty}^{\infty} \frac{1}{[2m^*(i\epsilon_n + \epsilon_F)]^2} = -\frac{1}{16m^{*2}(\pi T_C)^2} \sum_{n=0}^{\infty}\left[\left(n + \frac{1}{2} - \frac{i\epsilon_F}{2\pi T_C}\right)^{-2} + \left(n + \frac{1}{2} + \frac{i\epsilon_F}{2\pi T_C}\right)^{-2}\right]$$

$$= -\frac{1}{16m^{*2}(\pi T_C)^2}\left[\psi'\left(\frac{1}{2} - \frac{i\epsilon_F}{2\pi T_C}\right) + \psi'\left(\frac{1}{2} + \frac{i\epsilon_F}{2\pi T_C}\right)\right] \simeq -\frac{1}{8m^{*2}\epsilon_F^2} \quad , \tag{4.10}$$

where $\psi'(z) = \sum_{n=0}^{\infty}(z + n)^{-2}$, and we have made use of the fact that $(T_C/\epsilon_F) \ll 1$ in deriving the last line.[29] Thus

$$\Delta\chi = \frac{11}{768}\left(\frac{e}{\pi c}\right)^2 (T_C/\epsilon_F)^2\, UD^{-1} = \frac{11}{124}\left(\frac{e}{c}\right)^2 \frac{k_F}{m^*}(T_C/\epsilon_F)^2 \quad , \tag{4.11}$$

or

$$\Delta\chi/\chi_0 = -\tfrac{33}{31}\pi^2 (T_C/\epsilon_F)^2 \quad . \tag{4.12}$$

From Eq. (4.12) we conclude that the correction due to spin fluctuations is very small in the present model of a simple nondegenerate band without spin-orbit interactions.

V. GENERAL DISCUSSION OF EFFECT OF SPIN FLUCTUATIONS

In Sec. IV we showed that the orbital magnetism is not appreciably influenced by the spin fluctuations near the Curie temperature if the band is nondegenerate and the spin-orbit interaction is neglected. Although we confined ourselves to the region near T_C, the fact that the correction is small will not be changed in the system at $T = 0$ near the ferromagnetic instability (paramagnons). We examined the case based on Eq. (1.1) where the spin fluctuations and the orbital magnetism originate from the same kind of electrons. The conclusions, however, are valid even if the spin system has a different origin from that of mobile electrons.[30]

The physical reason for the small correction is that the orbital motions are not directly coupled to the large fluctuating magnetic field due to spin ordering. In this sense the situation is different if the band is degenerate or if the spin-orbit interaction is present. As regards the former, degeneracy of the band, some workers[31] pointed out the possibility of an orbitally aligned state besides the usual spin-ordered state in the model of the doubly degenerate band at zero temperature, which might suggest the divergence of the orbital susceptibility at the finite temperature T_C in some cases. In realistic fivefold d-orbital metals the situation is far from clear, although there have been discussions[32] of the orbital magnetism of a dilute nonmagnetic $3d$ impurity in metals based on the Anderson model[33] with orbital degeneracy.

The spin-orbit interactions have different aspects, which we briefly discuss in the following. To discuss this problem we add to Eq. (1.1) the following energy[34]:

$$\mathcal{H} = \frac{1}{4m^2c^2}\sum_i \vec{\sigma}(r_i) \times \nabla V(r_i)\left(p_i - \frac{e}{c}A_i\right) + \tfrac{1}{2} g_0 \mu_B H \sum_i \sigma_z(r_i) \quad , \tag{5.1}$$

where $\vec{\sigma}(r)$ is the Pauli spin matrix. V and g_0 are the crystal potential and g factor of the free elec-

tron, respectively.

The susceptibility coming exclusively from the second term in Eq. (5.1) is the Pauli spin paramagnetism. Since the spin-orbit interaction is usually weak, we examine its effect in first order. In this order the thermodynamic potential has an excess contribution, $\Delta\Omega$, due to the mixing of the two terms in Eq. (5.1). Another possible contribution due the mixing of the first term in Eq. (5.1) and $\mathcal{H}$, Eq. (2.1), vanishes after the summation over spin direction. By employing procedures similar to those in Sec. II, we obtain the following contribution to $\Delta\Omega$. We find

$$\Delta\Omega = \frac{e}{4m^2c_i^3} g_0\mu_B H A_{Q,l} \times \int_0^\beta d\lambda \int_0^\beta d\lambda' \langle T_0 \Lambda_l(Q)(\lambda)\sigma_z(Q)(\lambda')\rangle_0 \ , \tag{5.2}$$

where

$$\sigma_z(Q) = \sum_k (a^\dagger_{k^+,} a_{k^-,} - a^\dagger_{k^+,} a_{k^-,}) \ , \tag{5.3}$$

$$\Lambda_l(Q) = \langle k^+ \mid (\vec{\sigma}\times\nabla V)_l e^{iQr} \mid k^-\rangle \ , \tag{5.4}$$

T_0 is the time-ordering operator and $|k^\pm\rangle$ is the Bloch wave function. The time evolution and the thermal average $\langle\ \rangle_0$ in Eq. (5.2) refer to the total Hamiltonian without magnetic fields. In the tight-binding approximation Eq. (5.4) can be evaluated to the linear order of Q as follows:

$$\Lambda_l(Q) = i(\vec{\sigma}(0)\times\vec{Q})_l \xi \ , \tag{5.5}$$

where

$$\xi = \int d\vec{r}\, x \frac{\partial u}{\partial x}\varphi^2(r) . \tag{5.6}$$

In Eq. (5.6) the spherical symmetry of the atomic potential $u(r)$ is assumed and $\varphi(r)$ is the atomic wave function; $\Delta\Omega$ [Eq. (5.2)] yields an excess susceptibility $\Delta\chi$,

$$\Delta\chi = -\frac{g_0\xi}{2mc^2}\chi_{sp}(0,0) \ , \tag{5.7}$$

where $\chi_{sp}(0,0)$ is given by $\chi_{sp}(q,i\omega_l)$, Eq. (4.2), as $\chi_{sp}(0,0) = \lim_{q\to 0}\chi_{sp}(q,0)$. Since ξ is of the order of the average of the atomic potential $U(r) = -ze^2/r$ (z is the atomic number) with respect to the state $\varphi(r)$,[34] we have

$$\Delta\chi/\chi_{sp}(0,0) \approx -ze^2/2mc^2a = -z\alpha^2/2 \ , \tag{5.8}$$

where a is the Bohr radius and $\alpha = (137)^{-1}$ is the fine-structure constant. Thus in the pure system without band degeneracy the spin-orbit interaction has only a minor effect on the orbital magnetism.

It might be appropriate to point out the close formal similarity between present case and that of fluctuating superconductors.[35] In the latter we have large contributions near the critical temperature due to the presence of the Meissner effect below T_C. This physical difference is represented by the fact that in superconductors, where the electron-electron correlation becomes singular, the equation corresponding to Eq. (2.17) does not vanish and the process with two fluctuation propagators remains finite resulting in the divergence at $T = T_C$. To the contrary, the spin fluctuations, which are neutral in charge, do not yield large diamagnetic currents.

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APPENDIX: TRANSFORMATIONS OF EQ. (2.32) TO EQ. (2.33)

We first note the following:

$$T\sum_n\sum_k [\Gamma_x^2\Gamma_x^2\mathcal{G}^4 + \Gamma_x\Gamma_y\Gamma_{xx}\mathcal{G}^3] = +T\sum_n\sum_k [\tfrac{1}{3}\Gamma_x^2\Gamma_x\partial_y\mathcal{G}^3 + \tfrac{1}{2}\Gamma_y\Gamma_{xy}\partial_x\mathcal{G}^2] . \tag{A1}$$

By partially integrating Eq. (A1) we get

$$-T\sum_n\sum_k [\tfrac{1}{3}\mathcal{G}^3\partial_y(\Gamma_x^2\Gamma_y) + \tfrac{1}{2}\mathcal{G}^2\partial_x(\Gamma_y\Gamma_{xy})] \ . \tag{A2}$$

The first term on the right-hand side of Eq. (A2) can be transformed as follows, again by partial integrations:

$$\begin{aligned} -\tfrac{1}{3}T\sum_n\sum_k \mathcal{G}^3\partial_y(\Gamma_x^2\Gamma_y) &= -\tfrac{1}{3}T\sum_n\sum_k \mathcal{G}^3\Gamma_x \times(2\Gamma_y\Gamma_{xy} + \Gamma_x\Gamma_{yy}) \\ &= -\tfrac{1}{6}T\sum_n\sum_k \partial_x\mathcal{G}^2 \times(2\Gamma_y\Gamma_{xy} + \Gamma_x\Gamma_{yy}) \\ &= \tfrac{1}{6}T\sum_n \mathcal{G}^2\partial_x(2\Gamma_y\Gamma_{xy} + \Gamma_x\Gamma_{yy}) . \end{aligned} \tag{A3}$$

Equation (A3) and the second term of Eq. (A2) add to give Eq. (2.33), after an additional partial integration.

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